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NUCLEAR DATA FOR REACTORS

VOL. II

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PROCEEDINGS SERIES

NUCLEAR DATA FOR REACTORS

PROCEEDINGS OF A CONFERENCE ON NUCLEAR DATA - MICROSCOPIC CROSS-SECTIONS AND OTHER DATA BASIC FOR REACTORS HELD BY THE INTERNATIONAL ATOMIC ENERGY AGENCY IN PARIS, 17-21 OCTOBER 1966

In two volumes

VOL. II

INTERNATIONAL ATOMIC ENERGY AGENCY VIENNA, 1967

NUCLEAR DATA FOR REACTORS (Proceedings Series)

ABSTRACT. Proceedings of a Conference organized by the IAEA and held in Paris, 17-21 October 1966, at the invitation of the French Government and the French Atomic Energy Commission. The meeting was attended by 213 participants from 24 countries and 4 international organizations.

The main theme of the Conference was the discussion of neutron cross-sections and other neutron data with regard to their use for nuclear reactor calculations, emphasizing the roles of the compiler and the evaluator in this connection.

Contents: (Vol.1) Keynote address; Nuclear data requirements (4 papers); Cross-section and resonance parameters of non-fissile nuclides in the resonance energy region (13 papers); Statistical properties of resonance parameters (8 papers); (n, p), (n, p), (n, 2n)-reactions, etc. and inverse reactions (5 papers); Cross-sections and constants used as standards (7 papers); Neutron cross-sections above the resonance energy region (13 papers); Neutron radiative capture (16 papers); (Vol. II) Cross-sections and parameters of fissile nuclides (23 papers); Comparison of fission cross-sections in the resonance energy region (3 papers); Neutron data evaluation (12 papers); International co-operation in the field of nuclear data (1 paper); Panel discussion.

Of the 106 papers contained in these Proceedings, 83 are published in full with abstracts and 23 are presented by title and abstract only. Each paper is in its original language (69 English, 19 French and 18 Russian), the abstracts being in English and the original language if this is not English. Discussions are in English.

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NUCLEAR DATA FOR REACTORS, IAEA, VIENNA, 1967 STI/PUB/140

FOREWORD

A Conference on Nuclear Data for Reactors, held in Paris at the invitation of the French Government, was convened by the International Atomic Energy Agency on 17-21 October 1966. The meeting was held as a result of recommendations made by the International Nuclear Data Scientific Working Group. Over 200 delegates attended, representing 24 countries and four international organizations, and 106 papers were submitted. Of the 106 papers, 23 are given here by title and abstract only, since they were not presented at the Conference; copies of these papers, however, can be obtained on request from the Nuclear Data Unit of IAEA, Vienna.

The main purpose of the Conference was to provide an opportunity for reviewing results from recent basic neutron physics investigations against a background of need for basic information, especially as it applies to reactors.

After a keynote address, the meeting proceeded with three papers dealing with reactor and shielding needs. These were followed by sessions on basic neutron physics, which included the following topics: thermal and resonance fission cross-sections; cross-sections of non-fissile nuclides in and above the resonance region; neutron-induced particle reactions (and their inverse); radiative capture; the interpretation of fission cross-sections and the application of the optical and statistical models to the calculation of neutron cross-sections beyond the resonance region.

In his keynote address Dr. Usachev of the USSR gave special emphasis to the widening gulf of communication between data measurers and data users. Several sessions had been designed to try to bridge this gap. These consisted of: a session on standards, which was concerned with crosssections and constants suitable for use as standards and which included a paper summarizing the precision of present standards in relation to need; a session on the critical comparison of the cross-sections of fissile nuclides; a session on neutron data evaluation; and a session on the international exchange of neutron data.

A summary discussion by a panel of experts concluded the Conference. Their opinion was that there now exists essentially adequate information for thermal reactor calculations, that gaps and discrepancies in information for present needs will almost certainly be resolved as techniques improve, but that unforeseen needs and benefits justify a continued long-term search for fundamental understanding. The benefits from international co-operation were stressed throughout.

An assessment of the Conference in retrospect should take note of the interest shown in the sessions on standards, cross-sections of fissile nuclides, neutron data evaluations and international data exchange. Of these,

the session on data evaluation was especially noteworthy, not only because the need to include it as a session was due to the number and quality of the papers submitted by the evaluators, but also because of the recognition of the importance of the professional evaluator as the indispensable middleman between the physicist concerned with basic measurements and the physicist concerned with reactor applications. The growth in size and importance of this professional group of evaluators should give great impetus to the organizing of future conferences on the lines of the one held in Paris.

EDITORIAL NOTE

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For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies.

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^{*} This paper is presented by title and abstract only, since it was not read at the Conference.

Session VIII

CROSS-SECTIONS AND PARAMETERS OF FISSILE NUCLIDES

HARWELL MASS SPECTROMETRIC MEASUREMENTS OF THE RATIO OF NEUTRON CAPTURE TO FISSION FOR ²³³U, ²³⁵U, ²³⁹Pu AND ²⁴¹Pu IN REACTOR AND MAXWELLIAN NEUTRON SPECTRA

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Abstract

HARWELL MASS SPECTROMETRIC MEASUREMENTS OF THE RATIO OF NEUTRON CAPTURE TO FISSION FOR ²³³U, ²³⁵U, ²³⁵Pu AND ²⁴¹Pu, IN REACTOR AND MAXWELLIAN NEUTRON SPECTRA. During the past five years the mass spectrometric method has been used at Harwell for the measurement of alpha, the ratio of neutron capture to fission, for the four nuclides ²³³U, ²³⁵U, ²³⁹Pu and ²⁴¹Pu. Initially irradiations were carried out in the core of a nuclear reactor but more recently a position in the graphite reflector has been used (as yet for ²³⁹Pu and ²⁴¹Pu only).

In this paper the basic technique and its limitations are described. The expression used to derive 2200 m/s values of alpha (α_0) from the experimental results is given, its implications are discussed, and the neutron spectral and other data which are necessary for the calculations are considered.

The results which were obtained are set out in such a way that errors in the derived values of α_0 which arise from these particular measurements are clearly distinguished from those which are common to all measurements of this type. It is then shown that they are in good agreement with presently accepted "best" values.

1. INTRODUCTION

The mass spectrometric method is the most direct way of determining alpha, the relative probability of an absorbed neutron being captured by a fissile nuclide rather than causing fission. During the past five years it has been employed at Harwell for the measurement of alpha for the four nuclides 233U, 235U, 239Pu and 241Pu. Initially, in order to obtain useful results as rapidly as practicable, the high neutron fluxes present in the core of a nuclear reactor were employed for the irradiations. Values of a_0 (the value for 2200 m/sec neutrons) could then be obtained from the measured values in a reactor spectrum ($\hat{\alpha}$) and a knowledge of the spectrum parameters. More recently similar measurements have been made in the thermal column of a reactor (as yet for 239Pu and 241Pu only), in an effort to realise the greater potential accuracy with which a_0 can be derived from $\hat{\alpha}$ under these conditions.

Results from the earlier work were published as soon as they became available. However, since then, as a result of new measurements and critical surveys, small but significant changes have become necessary in some of the basic nuclear data originally assumed (changes in the 2200 m/sec cross-sections, in the g values, and in the resonance integrals of the four nuclides etc.). The opportunity has been taken here to take these and other changes into account, and to calculate the results from the complete series of measurements afresh, using a set of correction and conversion factors common to all. Some of the experiments included have been completed recently and have not previously been described in the literature.

In the present paper the basic method is described, its limitations are listed and methods of minimising their effects are given. The derivation of α_0 from $\hat{\alpha}$ is then described, together with an account of the spectral measurements which are necessary for the calculation, and a discussion of the way in which the choice of an irradiation position affects the

CABELL

TABLE I

Measurement made	Monitor or monitoring method used	Accuracy of measurement	Comments
Thermal neutron dose i.e. (nv _o)t	0.005 in. diameter cobalt wire	Originally ± 5-10%; improved to ± 3%	Co ⁶⁰ activity counted in a calibrated ionisation chamber
Epithermal index (r)	a) U ²³⁸ diluted with graphite	± 5%	Mass spectrometer and $4\pi \beta$ -proportional counter needed
	b) Pu ²⁴⁰ diluted with silica	± 12%	Mass spectrometer needed
	c) Activity ratio of Ag/Al and Co wires	Originally $\pm 12\%$; improved to $\pm 5\%$	Ratio of 110mAg and 60Co activi- ties measured in a calibrated ionisation chamber
	d) Cadmium ratio for gold	Depends on r	Applicable only to short irradia- tions or when r can be assumed constant. Simple γ -counter needed.
Neutron temperature (T)	a) Sm ¹⁴⁹ diluted with graphite	± 16 ⁰	Mass spectrometer needed
	b) Mn/Lu ceramics or alloy wires	± 5 ⁰ -10 ⁰ depending on r	γ -counter or spectrometer needed

DETAILS OF NEUTRON SPECTRUM AND DOSE MONITORS

accuracy of the result. The importance of avoiding neutron self-shielding is stressed, then some experimental details are given, including a description of a complete measurement. Finally the results are presented and compared with present "best" values.

2. BASIS OF THE METHOD

The basic principle of the method is very simple. Consider first the ideal case in which an isotopically-pure mixture of two isotopes of an element A i.e. ^XA and ^{X±n}A (assume $n \ge 2$), is irradiated with neutrons. Suppose ^XA to be the fissile nuclide under investigation and ^{X±n}A to be a reference nuclide which has a negligible neutron absorption cross-section. Suppose further that the mixture is isotopically analysed both before and after irradiation and that <u>before</u> irradiation

> <u>Number of atoms ^xA initially present</u> = R_0 Number of atoms ^{x±n}A present

whereas after irradiation

<u>Number of atoms ^xA remaining</u> = R_1 Number of atoms ^{x±n}A present

<u>Number of atoms x+1A formed by neutron capture</u> = R₂ Number of atoms $x\pm nA$ present

4

and

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TABLE II

Nuclide	Absorption cross- section	Fission cross- section	Capture cross- section	References
233 _U	576.3 ± 2.3	527.7 ± 2.1	48.6 ± 1.5	[11]
234 _U	-	-	95 ± 7	[12]
235 _U	679.9 ± 2.3	579.5 ± 2.0	100.5 ± 1.4	[11]
236 _U	-	-	6 ±•1	[12]
238 _U	-	-	2.73 ± 0.04	[12]
239 _{Pu}	1008.1 ± 4.9	742.4 ± 3.5	265.7 ± 3.7	[11]
240 _{Pu}	-	. ^	· 281 ± 7	[9]
241 _{Pu}	1376.1 ± 24.7	1012.7 ± 6.7	359 ± 16	[13,10]
242 _{Pu}	· · · ·	-	19.8 ± 1.1	[9]
59 _{Co}	-	-	37.4 ± 0.6	[14]
149 _{Sm}		-	43,200 ± 540	[15]

2200 m/sec NEUTRON CROSS-SECTIONS (in barns) FOR FISSILE, REFERENCE AND MONITOR NUCLIDES

Then, the neutron absorption cross-section of ^xA is directly proportional to $(R_0 - R_1)$, whereas its neutron capture cross-section is directly proportional to R_2 . It follows that α is given by

$$(R_{0} - R_{1}) / R_{2} = 1 + 1/\alpha$$
(1)

so that α can be determined directly from the measured isotopic ratios.

3. CORRECTION FACTORS

In practice this simple scheme may be complicated by a number of reactions for which allowance must be made. These include:-

1) The initial mixture may not be isotopically pure ^xA and ^{x±n}A, but contain small amounts of other isotopes of A which also undergo neutron capture during irradiation to produce some ^xA, ^{x+1}A and/or ^{x±n}A.

2) The reference nuclide is almost certainly not entirely impervious to neutron irradiation and some will be destroyed.

3) Some of the neutron capture product x+1A may undergo subsequent neutron absorption and be destroyed also, and

4) Some of the neutron capture product x+1A may be formed by nuclear transformations other than the one under investigation. (For example, when 241 Pu is irradiated with

TABLE III

Nuclide	g _a	^g f	g _γ
233 _U	0.9983 ± 0.20%	1.0003 ± 0.28%	, <u>-</u>
235 _U	0.9971 ± 0.13%	0.9781 ± 0.17%	- ·
238 _U	-	-	1.0017 ± 0.05%
239 _{Pu}	1.0723 ± 0.13%	1.0487 ± 0.17%	-
240 _{Pu}	-	-	1.0270 ± 0.15%
241 _{Pu}	1.030 ± 0.3%	1.0395 ± 0.6%	-
149 _{Sm}	-	-	1.6170 ± 0.26%

WESTCOTT's g-FACTORS FOR 20°C [2,3,13]

neutrons 242 Pu is formed, not only by direct neutron capture, but also by the sequence 241 Pu $\beta \rightarrow ^{241}$ Am $n.\gamma \rightarrow ^{242}$ gAm <u>k-capture</u> 242 Pu).

The corrections made necessary by reactions such as these can be made in one of two ways. Either they can be calculated, or they can be obtained directly from subsidiary irradiations made simultaneously and under the same conditions as the main irradiation.

If the former procedure i.e. calculation, is adopted, it is not only necessary to assume values for the neutron absorption and capture cross-sections of the nuclides concerned, for both thermal and epithermal neutrons, but also to rely on measurements of, and assumptions concerning, the shape of the neutron flux spectrum employed for the irradiation (see later). By making direct measurements of correction factors, however, most of these difficulties are circumvented and the results are usually more certain. For this reason, in the work described here; correction factors have been measured directly whenever practicable and calculations have been relied on only when the corrections are small, or when their direct measurement was not possible.

4. DERIVATION OF a₀

The quantity α given in equation 1 applies only to the particular neutron spectrum employed during the irradiation, and will be written henceforth as $\hat{\alpha}$ to denote this fact. By itself $\hat{\alpha}$ is only of limited value, particularly if the neutron spectrum was not well defined. For reactor design it is necessary to know α for specified conditions; in particular it is desirable to know α_0 , the value for 2200 m/sec neutrons.

The 2200 m/sec value can be obtained from a by means of the expression

$$(1 + a_0) = (1 + \hat{a})(g_f + rs_f)/(g_a + rs_a)$$
⁽²⁾

in which the subscripts f and a imply neutron fission and absorption respectively, g and s are factors which describe the departure of the nuclide from 1/v behaviour in the thermal and epithermal regions respectively, and r is Westcott's epithermal index [1].

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TABLE IV

Nuclide	Nuclide For absorption		For capture	Reference
233 _U	892 ± 20	761 ± 17	131 ± 27	[16]
234 _U	-	-	700 ± 70	[17]
235 _U	421 ± 7	277 ± 5	144 ± 5	[3,18]
236 _U	-	-	400 ± 40	[12]
238 _U	-	-	280 ± 12	[12]
239 _{Pu}	519 ± 15	324 ± 9	195 ± 12	[9]
240 _{Pu}	-	-	8453 ± 600	[9]
241 _{Pu}	707	541 ± 14	166	[9]
242 _{Pu}	-	-	1220 ± 200	[9]
59 _{Co}	-	-	72.6 ± 4.6	[14]
149 _{Sm}	-	-	2500	[2]

RESONANCE INTEGRALS (above 0.45eV) IN BARNS, FOR FISSILE, REFERENCE AND MONITOR NUCLIDES

Values of g and s for the four nuclides 2^{33} U, 2^{35} U, 2^{39} Pu and 2^{41} Pu (and others) have been computed for different neutron temperatures [2-4] so that, provided the neutron temperature (T) and the epithermal index are known, expression (2) may be applied. T and r can be measured by employing monitors during the irradiation.

The simplest case is when a purely Maxwellian spectrum is used for the irradiation. Equation (2) then simplifies to

$$(1 + a_0) = (1 + \hat{a})g_f/g_a$$

and, since g factors are generally known more precisely than s factors (which are derived from resonance integrals), a_0 is, in principle, obtained most accurately under these conditions.

When irradiations are carried out in the core of a reactor, however, the rs terms of equation (2) make substantial contributions and accuracy is lost. In addition, unless other evidence is available to the contrary, not only must the assumptions implicit in equation (2) be made (i.e. that the neutron spectrum consists of an unperturbed Maxwellian component joined to an unperturbed 1/E distribution of epithermal flux) but some form of the epithermal cut-off function must be assumed also. These assumptions are other sources of uncertainty. In this work Westcott's Λ_4 form [2] has been used for the epithermal cut-off function

In contrast, from a purely practical point of view, the use of reactor core spectra is to be preferred, since they give considerably higher neutron flux densities and, in consequence, measurable changes are produced more rapidly.

(3)

TABLE V

Irradiation Position	Neutron Temperature, T	Epithermal Index, r	Neutron Dose $i \times 10^{-20}$	â	Conversion* Factor	a ₀
C5 - 3	87 ⁰	0.070	4.23	0.0953	0.9950	0.0898
- 4	99	"	3.71	0.0965	n	0.0910
- 6	7	79	4.03	0.1018	"	0.0963
B6 - 3	94 ⁰	0.050	2.90	0.0920	0.9974	0.0891
- 4	77	7	3.21	0.0960	æ	0.0931
- 5	77	"	3.22	0.0947	7	0.0918
- 6	"	×	2.76	0.1003	7	0.0974
D1 - 3	990	0.058	3.26	0.0964	0.9962	0.0922
- 4	n	* 19	3.02	0.0989	39	0.0947
- 5	79 ¢	"	3.03	0.0989	"	0.0947
- 6	31	7	2.75	0.0940	"	0.0898

RESULTS FOR ²³³U

*Defined here, and in the following, as $(g_f + rs_f)/(g_a + rs_a)$

At the time the work summarised here was started very few measurements of \hat{a} for the four nuclides had been made, and these almost invariably under ill-defined conditions. It was decided therefore to make initial measurements in core spectra, defining these as precisely as possible with the monitors available, then to follow these by repeated measurements in a Maxwellian spectrum, to see if improvements could be made.

At the present time \hat{a} has been measured for all four nuclides in reactor spectra, and additional measurements have also been made for 239 Pu and 241 Pu in a spectrum almost devoid of epithermal neutrons. For the reasons stated above, other things being equal, the latter results are to be preferred.

5. OTHER CONSIDERATIONS

It is evident that during irradiations of the type described it is essential to ensure that erroneous results do not arise in either samples or monitors due to neutron self-shielding. To avoid this possibility the samples and monitors were diluted by materials which are virtually transparent to neutrons. Thus uranium samples were diluted with over one hundred times their own weight of magnesium oxide (which was also utilised in the subsequent purification of the uranium from fission products), whereas plutonium samples were diluted with up to a thousand times their own weight of precipitated silica. Monitors were treated in a similar manner.

TABLE VI

Position*	r	i × 10 ⁻²⁰	â	Conversion Factor	a ₀
1	0.057	3.01	0.1901	0.9898	0.1780
2	0.060	3.44	0.1872	0.9892	0.1744
3	0.062	3.89	0.1839	0.9888	0.1706
4	0.064	4.09	0.1844	0.9883	0.1705
5	0.067	4.23	0.1876	0.9879	0.1732
7	0.059	4.35	0.1794	0.9894	0.1669
8	0.054	4.06	0.1847	0.9903	0.1732
9	0.050	4.04	0.1786	0.9911	0.1681
10	0.046	4.22	0.1795	0.9919	0.1699

RESULTS FOR 235₁₁

 $T = 103^{\circ}$ for all positions

An exception to this general rule was the use of 0.005 in. diameter cobalt wire for thermal neutron dose measurements. The corrections necessary for neutron self-shielding effects in this material are well established [5].

6. EXPERIMENTAL DETAILS

Most of the experimental details of individual measurements have already been described in full in a number of publications [6-10], so only a brief summary will be given here. For further details the original accounts should be consulted.

A complete measurement can be divided into a number of stages, as follows:-

1) Isotopically enriched samples of the fission and reference nuclides are obtained, mixed in suitable proportions and mounted on a diluent. A number of samples of the mixture (only micrograms of fissile material are required) are sealed in individual evacuated silica ampoules.

2) Samples for the subsidiary measurements are prepared in a similar way. Neutron dose, epithermal index and neutron temperature monitors are prepared also.

3) The sample ampoules and monitors are packed into irradiation cans, alongside each other if space permits, otherwise in adjacent cans of a string. Samples for the subsidiary measurements are incorporated in the same way.

4) The samples and monitors are irradiated for a suitable time in the reactor position chosen.

5) After allowing a suitable time for most of the gross fission product activity to decay, the ampoules are broken open and the irradiated samples are separated from

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remaining fission products and other impurities. Remote operation inside a shielded cell is necessary.

6) The remaining un-irradiated samples are treated in a similar way.

7) Several microgram aliquots of the unirradiated and of the irradiated samples are mounted for individual isotopic analysis with a mass spectrometer. By repeated measurements their isotopic compositions are determined as precisely as possible.

8) The irradiated monitors are used to determine the neutron dose, epithermal index and neutron temperature appropriate to each irradiated sample.

A.E.I. Type M.S 5 mass spectrometers, with tungsten triple-filament surface ionisation sources and electron multiplication of the output current, have been used for all isotopic analyses.

A variety of monitors have been used at different times. They are listed in Table I (column 2), together with estimations of the accuracy of the measurements made with them (column 3) - based on the precision of the measurements made with the apparatus available and the accuracy with which the nuclear parameters involved are known - and some comments on their use (column 4).

Purification of irradiated samples from fission products and other impurities have been achieved by normal radiochemical techniques; liquid-liquid solvent extraction for uranium samples and anion exchange chromatography for plutonium samples.

Irradiations have been either in the 0.25 in. diameter aluminium flux scanning tubes which are attached to, or run through the centre of the hollow multi-plate fuel elements of the PLUTO reactor or, more recently, in a position in the thermal column of the DIDO reactor.

7. RESULTS

Information from the monitors and (where necessary) the reaction rates which were required in order to apply corrections to the measured isotopic ratios, were calculated from the general expression [1]

$$R = (nv_0)\sigma_0(g + rs)$$

(4)

in which R is the reaction rate per target atom per second, (nv_0) is the conventional neutron flux, σ_0 is the cross-section of the target atom for 2200 m/sec neutrons, and g, r and s have the meanings previously assigned to them.

The values for σ_0 which were used for application of expression 4 are listed in Table II and, in the author's view, are the most reliable at the time of writing. The data for 241 Pu in this table requires special comment. In view of the fact that the results of the most recent review of 241 Pu data by Westcott et al. [11] are greatly influenced by the present author's previously published values for \hat{a} for this nuclide [9], values which should now be amended, the later results have been discarded in favour of those from an earlier review [13], which were not influenced in this way. For neutron capture by 241 Pu the present author's own values have been used [10], since these are believed to be the most reliable at the present time.

g values at the appropriate neutron temperature, for use in expression 4, were normally obtained from tables [2-4]. These are based on the 20^oC values given in Table III; the same percentage error in g was assumed irrespective of the temperature. Where tabulated values were not available g was assumed to be unity.

s values were usually obtained from tables as well [2,3], although, where necessary, the tabulated values were adjusted to be consistent with the resonance integral data given in

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TABLE VII

Position*	ſ	i × 10 ⁻²⁰	â	Conversion Factor	a ₀
1	0.053	2.36	0.450	0.9359	0.357
4 🗸	0.066	3.01	0.460	0.9311	0.359
5	0.071	3.28	0.453	0.9294	0.351
7	0.075	3.43	0.452	0.9281	0.348
8	0.074	3.49	0.458	0.9283	0.354
10	0.073	3.21	0.452	0.9287	0.349
11	0.072	3.17	0.452	0.9289	0.349

RESULTS FOR ²³⁹Pu (reactor core spectra)

$T = 101^{\circ}$ for all positions

Table IV. For this purpose the expression [3]

$$\Delta s = \sqrt{\frac{4T}{\pi T_o}} \Delta \left(\frac{\Sigma}{\varphi_o}\right) - b\Delta g +$$

was employed. Where not directly available, s values were calculated from Westcott's formulae [2].

In the case of 0.005 in. diameter cobalt wires, self-shielding corrections were made in conjunction with the data of Tables II and IV, G_{th} and G_r were taken to be 0.97 \pm 0.01 and 0.52 \pm 0.02 respectively [5].

Results from the experiments and the derived values of α_0 are given below under the appropriate headings and comments. Three separate errors are quoted for the final value obtained for α_0 . The first is the standard error i.e. it takes into account random errors in the experiment only, and is a measure of the precision of the result. The second error takes into account all possible systematic errors (errors in measurements of the neutron temperature, epithermal index, neutron dose, isotopic ratios of the target materials etc. and errors in the assumed neutron cross-sections) also, except that introduced by the factor $(g_f + rs_f)/(g_a + rs_a)$ see equation (2) - used in the conversion of $\hat{\alpha}$ to α_0 . The third error introduced by the conversion factor as well. A distinction has been made between these last two errors because the former should be used when the results given here are compared with those obtained by other workers making similar measurements (as, for example, in reviews such as those by Westcott et al. [11,13]), whereas the latter is applicable when the result is considered in isolation, and an absolute value for α_0 is required.

7.1 Uranium - 233

Thirteen samples of 233 U mixed with 238 U were irradiated in three different flux-scanning tubes. Separate samples of 238 U were used for epithermal index measurements.

(5)

TABLE VIII

Position*	r	i × 10 ⁻²⁰	â	Conversion factor	a ₀
2	0.051	3.74	0.315	1.0114	0.330
4	0.078	4.12	0.323	2	0.338
6	0.080	4.79	0.343	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	0.358
. 8	0.080	5.02	0.308	77	0.323
12	0.046	4.96	0.330	9	0.345

RESULTS FOR ²⁴¹Pu (reactor core spectra)

 $T = 90^{\circ}$ for all positions

 $149_{\rm Sm}$ was used for temperature measurements and the "burn-up" of $233_{\rm U}$ was used for neutron dose determinations (neutron dose, i neutrons per cm², is given by i = (nv₀)t, where t is the duration of the irradiation in seconds).

The results, which replace those quoted previously [6], are listed in Table V. From them a_0 was computed to be:

 $\alpha_0 = 0.0923 \pm 0.0005$ (standard error)

or $a_0 = 0.0923 \pm 0.0012$ (excluding error in conversion factor)

or $a_0 = 0.0923 \pm 0.0049$ (absolute error).

7.2 Uranium - 235

Nine samples of 235 U mixed with 238 U were irradiated in a flux scanning tube. Neutron dose and spectrum parameter measurements were similar to those for 233 U. The results, which replace those quoted previously [7], are listed in Table VI and gave:

 $a_0 = 0.1716 \pm 0.0012$ (standard error)

or $a_0 = 0.1716 \pm 0.0019$ (excluding error in conversion factor)

or $a_0 = 0.1716 \pm 0.0033$ (absolute error).

7.3 Plutonium - 239

(a) In reactor core spectra

Seven samples of ²³⁹Pu mixed with ²⁴²Pu were irradiated in a flux scanning tube. Separate measurements were made of the "burn-up" of ²⁴⁰Pu, both in order to allow precise corrections to be made to the main measurements, and also to provide values of r. Cobalt monitors were used for neutron dose measurements, ¹⁴⁹Sm monitors for the determination of neutron temperature.

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TABLE IX

RESULTS FOR ²⁴¹Pu (Maxwellian spectrum) $r = (7.5 \pm 1.0) \times 10^{-4}$ $T = 116 \pm 4^{\circ}$

Sample	i × 10 ⁻²⁰	â
1	1.308	0.337
2	1.301	0.330
3	1.316	0.345
· 4	1.362	0.338
5	1.344	0.359

The results are summarised in Table VII and replace those previously quoted [8]. They gave the following values for a_0 :

 $a_0 = 0.352 \pm 0.002$ (standard error)

or $a_0 = 0.352 \pm 0.006$ (excluding error in conversion factor) or $a_0 = 0.352 \pm 0.007$ (absolute error).

(b) In a Maxwellian spectrum

Five samples of ²³⁹Pu mixed with ²⁴²Pu were irradiated in the graphite reflector of DIDO [10]. Samples of ²⁴¹Pu mixed with ²³⁹Pu, and of ²⁴⁰Pu mixed with ²⁴²Pu, were also irradiated to provide additional information. Cobalt monitors were used for neutron dose measurements, the neutron temperature (116 ± 4°C) was determined by the manganese-lutetium activity ratio method (by Mr. E. D. Jones of Research Reactors Division, A.E.R.E.), and the epithermal index was determined to be (7.5 ± 1.0) × 10⁻⁴ from measurements of the cadmium ratio for gold.

The results which, in view of their imprecision, are not set out in detail, gave:-

 $a_0 = 0.377 \pm 0.011$ (standard error)

(or $a_0 = 0.377 \pm 0.020$)(excluding error in conversion factor)

or $a_0 = 0.377 \pm 0.021$ (absolute error).

7.4 Plutonium - 241

Completely different exp.t Convelated errors probably sum ll

Five samples of ²⁴¹Pu mixed with ²³⁹Pu were irradiated in a flux scanning tube. Samples of ²⁴⁰Pu mixed with ²⁴²Pu were included, as for measurements with ²³⁹Pu, to allow corrections to be applied to the main results and for the measurement of r. Epithermal indices were also measured by the silver-cobalt activity ratio method [14,19]; neutron temperature was again determined by the manganese-lutetium activity ratio method and neutron dose

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TABLE X

SUMMARY OF α_0 RESULTS

Nuclide	This v	From reviews by Westcott et al. [11,13]	
	uclide With experimental error only Absolute value		
233 _U	0.0923 ± 0.0012	0.0923 ± 0.0049	0.0921 ± 0.0029
235 _U	0.1716 ± 0.0019	0.1716 ± 0.0033	0.1734 ± 0.0025
239 _{Pu}	0.356 ± 0.006	0.356 ± 0.007	0.3580 ± 0.0054
241 _{Pu}	0.355 ± 0.013	0.355 ± 0.015	0.3589 ± 0.0252

by cobalt monitors. Corrections for the production of ^{242}Pu by the sequence $^{241}Pu \beta^{-}_{241Am n, \gamma} 242g_{Am K-capture} 242Pu$, were also applied; these corrections were omitted from the original calculations [9] which are therefore inaccurate.

The results are given in detail in Table VIII and resulted in:

 $a_0 = 0.339 \pm 0.006$ (standard error)

or $a_0 = 0.339 \pm 0.030$ (excluding error in the conversion factor)

or $a_0 = 0.339 \pm 0.031$ (absolute error)

(b) In a Maxwellian spectrum

These measurements were made using an irradiation position in the graphite reflector of DIDO [10], at the same time as those for 239 Pu (see above); the spectrum conditions were therefore similar. Corrections were applied for the production of 242 Pu via 241 Am. The results are listed in Table IX and, with $(g_f + rs_f)/(g_a + rs_a)$ equal to 1.0114 ± 0.0069 gave:-

 $a_0 = 0.357 \pm 0.006$ (standard error) or $a_0 = 0.357 \pm 0.014$ (excluding error in conversion factor) or $a_0 = 0.357 \pm 0.016$ (absolute error).

8. CONCLUSIONS

The final values for α_0 obtained as a result of this work are listed in Table X (columns 2 and 3). For 233U and 235 U the data were obtained from irradiations in reactor core spectra only. For 239 Pu and 241 Pu the values are weighted means obtained from irradiations in both reactor core spectra and in a thermal column spectrum. (In this weighting process we have, somewhat arbitrarily, doubled the weight given to thermal column results relative to reactor core results, in order to take some account of our preference for the former - see above).

Errors in Table X have been expressed in two ways. Those in column 2 exclude uncertainty in the factor used to obtain α_0 from $\hat{\alpha}$ and should be used when the values given

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here are compared with those from other measurements of a similar type. In column 3 the uncertainty in the correction factor is included as well. As might be concluded from an examination of equation (2), other things being equal the error in the conversion factor has a proportionally greater effect when a_0 is smaller than when it is larger. This effect, which is the main source of error when a_0 is small, can be seen if the errors in column 2 and 3 for 233U are compared with those for 239Pu or 241Pu.

It is not our intention to make an assessment of the value of the results given here compared with those obtained by other workers (not necessarily by the mass spectrometric method), since the latter values have been the subjects of recent reviews [11,13]. It is sufficient to say that, for 2^{33} U and 2^{35} U, the agreement, in general, is good, particularly with the more recent results. In the case of 2^{39} Pu only one other measurement, with a result in good agreement with the one given here, has been reported. No other measurement has yet been reported for 2^{41} Pu.

For the sake of completeness, however, we list in column 4 of Table X the current "best" values of α_0 for the four nuclides. These values were obtained, not only as a result of direct measurements of alpha, but also from a consideration of all measurements which have been made of the individual nuclear parameters of the nuclides, and of the relationships between them. It will be seen that agreement between these values and the results of this work is well within the quoted limits of error. The precision we are able to quote for α_0 for 2^{41} Pu is particularly gratifying, since information concerning this nuclide is particularly sparse.

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RATIO OF CAPTURE TO FISSION IN $^{235}\mathrm{U}$ AND $^{239}\mathrm{Pu}$

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Abstract

RATIO OF CAPTURE TO FISSION IN ²³⁵U AND ²³⁹Pu. Thin samples of uranium and plutonium have been irradiated in the pneumatic carrier of the NRU reactor, and their changes in isotopic composition measured by a mass spectrometer, to obtain information in radiative capture in ²³⁵U at thermal and epithermal neutron energies and on capture and absorption in ²³⁹Pu.

The irradiation position, a vacant lattice site of the NRU reactor (natural uranium fuelling) was characterized by a well-moderated high-intensity thermal flux ($\sim 2, 5 \times 10^{14} n/cm^2$ s) with an epithermal component having a relatively small departure from a 1/E shape and an intensity r of 0.018 (Westcott convention). Two experimental irradiations were carried out, in the first of which a sample of natural uranium was irradiated bare and a sample enriched in ²³⁵U was irradiated under cadmium; the integrated flux was 0.32 n/kb and the neutron temperature 60°C. In the second experiment (0.71 n/kb, 47°C) similar uranium samples were irradiated, and, in addition, samples of ²³⁹Pu "spiked" with ²⁴²Pu were irradiated in the same capsule.

The change in ²³⁵U content of the bare sample relative to the almost constant ²³⁸U gave the integrated flux and, when combined with the measured ²³⁶U production, the value of α for ²³⁵U. In the cadmiumcovered sample, measurement of the ²³⁶U, when combined with subsidiary silver-activation measurements of the epithermal flux, gave a value of the resonance integral for radiative capture in ²³⁵U.

The change in ²³³Pu and (²⁴⁰Pu ^{‡241}Pu) contents relative to the comparatively constant ²⁴²Pu content gave values of α and absorption cross-section for ²³⁹Pu; corrections for epithermal effects were made by calculation, but they introduce only minor uncertainties. Corrections from the measured values for a Maxwellian neutron distribution to a neutron velocity of 2200 m/s involve uncertainties that are comparable with the experimental errors.

Introduction

The Chalk River Nuclear Laboratories have a long-standing interest in the burn-up that can be attained in natural-uranium fuelled heavy-water moderated reactors. These reactors are characterized by a well thermalized neutron spectrum so that the factors affecting burn-up are largely dependent on thermal neutron cross-sections, which accounts for the strong Canadian interest in accurate values of these cross-sections. Correspondingly, measurements directly related to burn-up studies are capable of interpretation in terms of 2200 m/s cross-sections.

A summary of early work in this field was given at the 1958 Geneva Conference [1] where thermal neutron cross-section data derived from measurements on irradiated fuel were presented. Extensions of this work have given more reliable values for a (the ratio of capture-tofission cross-sections) of 235 U [2] and for the capture cross-section of 239 Pu [3]. These results will be mentioned later in the present paper.

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TABLE I

Measurements of Epithermal Index

Date	Monitor	$r\sqrt{T/T_0}$
August 1960	Au-Al	0.0189
August-September 1961	first irradiation	
April-May 1962	second irradiation	
October 1962	Ag-Al	0.0192
August 1963	Au-Al	0.0196*

* T.A. Eastwood, private communication

The precise interpretation of such measurements depends on an accurate knowledge of the neutron spectrum. As with other reactor types there is a continuing improvement in methods of calculating spectra, which justifies attempts to improve the accuracy of the basic cross-section data. This consideration has led to a study at Chalk River of small samples irradiated in an empty lattice position where the neutron spectrum is better known than in the fuel itself. These measurements are the subject of the present paper. Further studies are presently in progress on samples irradiated in a position where the flux is lower but more completely thermalized, with an almost negligible contribution from the slowing-down spectrum.

In our present state of knowledge of the thermal neutron cross-sections of the principal fissile nuclei, the results of any particular measurement can have only a relatively minor impact on any set of self-consistent values obtained by least-squares fitting to all available data (e.g. [4]). However, direct measurements of fission cross-sections are more widely dispersed than would be expected from the accuracies claimed, so that least-squares fitting demands a more or less arbitrary treatment of the experimental data. This is especially true of ²³⁹Pu and, in spite of a recent direct measurement of the 2200 m/s fission cross-section [5], reliable measurements of the absorption and capture cross-sections are still of great interest.

The results of the present work have been made available in preliminary form in various reports of limited circulation [6]. All previously quoted results are hereby superseded.

Experimental Procedures

Two irradiations were carried out. On each occasion the samples were irradiated in the pneumatic carrier of the Chalk River NRU reactor during the period when this reactor was fuelled with natural uranium [7]. The neutron flux in the pneumatic carrier, which is located at an empty lattice position, was about 2.5×10^{14} n cm⁻²sec⁻¹. The spectrum closely approximated a Maxwellian distribution plus an epithermal component of relative intensity $r \approx 0.02$ on the Westcott convention [8].

The temperature of the Maxwellian component was calculated from the moderator temperature (supplied by the reactor operations staff) by adding the correction factor (450 r) suggested by Bigham following his extensive studies of spectra in reactor lattices [9].

The intensity of the epithermal component was measured by irradiating gold-aluminum and silver-aluminum wires in a subsidiary series of experiments which bracketed the uranium and plutonium irradiations (the first of these did not include an internal epithermal monitor; in the second it was lost). The reactor was operated in a manner not expected to produce changes in the epithermal/thermal flux ratio, and the gold and silver measurements, and also the agreement between the results obtained in the two uranium irradiations, support the assumption of a constant value of r. The results are given in Table I; they are based on a value of s $\sqrt{T_0/T} = 17.0$ for gold i.e. on a value of 1535 barns for the resonance integral above 0.5 eV [see 10]. The silver measurements depend on the gold-silver comparison made by Hart et al. [11].

The departure from 1/E shape of the epithermal spectrum in an empty lattice site has been discussed by Bigham and Pearce [12]. The necessary corrections to the present measurements are quite small.

In the first irradiation a sample of natural uranium was irradiated bare and a sample enriched in 235 U was irradiated under cadmium; the integrated flux was 0.32 neutrons per kilobarn. The samples, located at opposite ends of an aluminum capsule, each consisted of about 25 mg U₃O₈ spread out over an area of about 1.1 cm² in a small aluminum can. The enriched sample was enclosed in a cadmium box 0.8 mm thick, 1.3 cm diameter and 1.3 cm high, located about 6 cm below the bare sample. The capsule was irradiated close enough to the central plane of the reactor for the effects of the vertical flux gradient to be negligible.

The arrangements for the second irradiation were similar except that both the bare natural uranium and cadmium-covered enriched uranium samples were accompanied by samples of $Pu0_2$ ($\sim 1 \text{ mg/cm}^2$) containing approximately 10% of ²⁴²Pu. Each plutonium sample was about 6 mm away from the corresponding uranium sample, and the bare and cadmium-covered samples were, again, 6 cm apart. The integrated flux in this irradiation was 0.71 neutrons per kilobarn.

After cooling, the capsules were cut open and the irradiated samples dissolved in nitric acid. The solutions of the uranium samples were analyzed for plutonium by alpha counting, and for uranium by colorimetry using KCNS; this Pu/U analysis measured the destruction of ²³⁸U during the course of the irradiation. Purified uranium samples were prepared from these solutions for isotopic analysis by mass spectrometry. The irradiated plutonium samples were purified and analyzed mass-spectrometrically; ²⁴¹Am was removed immediately before analysis.

Samples of approximately 30 μ g were used for the isotopic analyses, which were carried out on two rather similar mass spectrometers, Consolidated Electrodynamics Models 21-702B (uranium) and 21-703 (plutonium). These instruments are of the single-focusing 60⁰ sector type, with a radius of 30 cm, and were operated with a resolving power of 400. They employ triple-filament surface-ionization sources and the faraday-cup collectors were used. In these experiments mass

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TABLE II

Sample		Isotopic Analysis ²³⁹ Pu+ ²⁴⁰ P Atom Ratio dpm/mg T			
Natural U unirradiated	MS-702-192 MS-702-230	²³⁵ U/ ²³⁸ U ²³⁵ U/ ²³⁸ U	$(0.7251 \pm 0.0040)10^{-2}$ $(0.7254 \pm 0.0019)10^{-2}$		
Natural U irradiated (bare)	MS-702-206 MS-702-207	²³⁵ U/ ²³⁸ U ²³⁶ U/ ²³⁸ U ²³⁵ U/ ²³⁸ U ²³⁶ U/ ²³⁸ U	$\begin{array}{c} (0.5870 \pm 0.0021)10^{-2} \\ (0.0213 \pm 0.0004)10^{-2} \\ (0.5856 \pm 0.0025)10^{-2} \\ (0.0211 \pm 0.0004)10^{-2} \\ (0.0211 \pm 0.0004)10^{-2} \end{array}$	2.78 × 10 ⁵ 2.84 × 10 ⁵ 2.80 × 10 ⁵	
	MS-702-231	²³⁵ U/ ²³⁸ U ²³⁶ U/ ²³⁸ U	$(0.5878 \pm 0.0015)10^{-2}$ $(0.0213 \pm 0.0004)10^{-2}$	2.83×10 ⁵	
Enriched U unirradiated	MS-702-232 MS-702-234	²³⁶ U/ ²³⁵ U ²³⁸ U/ ²³⁵ U ²³⁶ U/ ²³⁵ U ²³⁸ U/ ²³⁵ U	$(0.0680 \pm 0.0006)10^{-2}$ 1.060 ± 0.004 $(0.0679 \pm 0.0006)10^{-2}$ 1.062 ± 0.003		
Enriched U irradiated (under Cd)	MS-702-233 MS-702-235	²³⁶ U/ ²³⁵ U ²³⁸ U/ ²³⁵ U ²³⁶ U/ ²³⁵ U ²³⁸ U/ ²³⁵ U	$(0.1604 \pm 0.0007)10^{-2}$ 1.070 ± 0.004 (0.1604 ± 0.0007)10^{-2} 1.067 ± 0.004	9.11 × 10 ⁴ 9.15 × 10 ⁴ 9.20 × 10 ⁴ 9.06 × 10 ⁴	

First Irradiation, 20 August-6 September 1961 Results of Chemical and Mass Spectrometric Analyses

TABLE III

Second Irradiation, 12 April-28 May 1962 Results of Mass Spectrometric Analyses

Sample	Isotopic Analysis			
	Atom Ratio			
Natural U irradiated (bare)	MS-702-402 MS-702-403	²³⁵ U / ²³⁸ U ²³⁶ U / ²³⁸ U ²³⁵ U / ²³⁸ U ²³⁶ U / ²³⁸ U	$(0.4552 \pm 0.0008)10^{-2} (0.0416 \pm 0.0004)10^{-2} (0.4550 \pm 0.0008)10^{-2} (0.0412 \pm 0.0004)10^{-2}$	
Enriched U irradiated (under Cd)	MS-702-404	²³⁶ U/ ²³⁵ U ²³⁸ U/ ²³⁵ U	$(0.2711 \pm 0.0005)10^{-2}$ 1.0708 ± 0.0018	
Plutonium unirradiated	MS-703-09 (13 May 63)	²³⁹ Pu ²⁴⁰ Pu ²⁴¹ Pu ²⁴² Pu	$\frac{\text{Abundance (atom \%)}}{88.497 \pm 0.010}$ 1.586 ± 0.002 0.219 ± 0.001 9.698 ± 0.010	
Plutonium irradiated (bare)	MS-703-10 (15 May 63)	²³⁹ Pu ²⁴⁰ Pu ²⁴¹ Pu ²⁴² Pu	60.37 ± 0.06 21.23 ± 0.04 3.13 ± 0.01 15.27 ± 0.04	

discrimination effects could be ignored [13]. Tables II and III summarize the results of the analyses.

Each mass spectrometric analysis represents the averaging of about thirty individual spectrograms, and the assigned errors are calculated from the dispersion of these individual results (or set equal to 0.1%, whichever is larger). More repeat analyses were made on the first irradiation material because of the smaller changes in isotopic composition.

The same starting materials were used for both uranium irradiations, and their composition is given in Table II. The natural uranium was from a stock that has been analyzed repeatedly at Chalk River on the model 21-702B mass spectrometer, and a standard value of $(0.7256 \pm 0.0007)10^{-2}$ for the 235/238 atom ratio has been obtained from all these measurements. (A value of $(0.7257 \pm 0.0007)10^{-2}$ was obtained earlier on an older instrument [13].) The present results agree with the standard value within their errors, but the standard value has been used in all subsequent calculations. Use of the present analyses instead would not appreciably affect the results except to increase some of the quoted errors slightly.

No measurements of the Pu/U ratio were made for the second irradiation; it was judged that the conditions were sufficiently similar that the effective absorption cross-section of 238 U derived from the first irradiation could be used to make the relatively small correction for 238 U destruction in the second irradiation. Because of experimental mis-fortunes no satisfactory samples were obtained of the plutonium irradiated under cadmium.

Interpretation

(a) Uranium-irradiations - bare samples

If no 234 U were present, the ratio of radiative capture and fission cross-sections for 235 U, \hat{a}_5 , would be given by

$$\hat{a}_5 / (1 + \hat{a}_5) = (1 - \delta_6)z / \{y^{(1 - \delta_6) / (1 - \delta_8)} - 1\}$$

where $y = \frac{238/235 \text{ atom ratio in the irradiated uranium}}{238/235 \text{ atom ratio in the initial (natural) uranium}}$

z = 236/235 atom ratio in the irradiated uranium

 $\delta_6 = \hat{\sigma}_6 / \hat{\sigma}_5$ and $\delta_8 = \hat{\sigma}_8 / \hat{\sigma}_5$

where $\hat{\sigma}_5$, $\hat{\sigma}_6$ and $\hat{\sigma}_8$ are the neutron absorption cross-sections of ²³⁵U, ²³⁶U and ²³⁸U respectively. The symbol $\hat{\sigma}$ denotes the effective crosssection of the Westcott convention [8].

The correction required for the $^{234}U(n,\gamma)^{235}U(n,\gamma)^{236}U$ reaction chain is quite small. The main effect is the production of ^{235}U from ^{234}U ; neglecting this leads to an under-estimate of the burnup of ^{235}U and hence an over-estimate of \hat{a}_5 . The correction to \hat{a}_5 was -0.17% for the first irradiation and -0.19% for the second irradiation.

A value of 0.0097 for δ_8 was obtained from the measured Pu/U ratio. This corresponds to a value of 6.4 b for $\hat{\sigma}_8$; if there were no self-shielding of resonance neutrons it would be expected to be 8.4 b, and the difference is considered to be reasonable. For capture in ²³⁶U

the 2200 m/s cross-section has been measured by Eastwood et al.[14] as 5.5 b; the resonance integral was taken to be 290 b as a compromise between values of 240 ± 20 b [14] 330 ± ? b [15] and 310 ± 70 b [16]. These data give $\hat{\sigma}_6 = 11.8$ b, and hence $\delta_6 = 0.0178$. The effects on \hat{a}_5 of errors in any of these cross-sections are quite small, for example an error of about 0.1% in \hat{a}_5 would result from errors of 10% in δ_8 or 20% in δ_6 (second irradiation) or 40% in δ_6 (first irradiation).

The values of \hat{a}_5 obtained from the first and second irradiations are 0.1790 ± 0.0033 and $0.1787_5 \pm 0.0017$ respectively. The correction to values for 2200 m/s neutrons was made through the Westcott [8] formalism $\hat{\sigma} = \sigma_0(g+rs)$ with values of g and s₄ from Critoph's tables [17]. (The differences between s₄ and s₂, which correspond to different epithermal spectrum shapes near cut off, are negligible.) The s₄ values were slightly modified so as to be consistent with the data obtained in the present experiments on ²³⁶U production under cadmium. The value of a_5 from the first irradiation (neutron temperature $60^{\circ}C$) is 0.1768, and that from the second ($47^{\circ}C$) is 0.1764. The weighted mean is

$a_5 = 0.1765 \pm 0.0015$ at 2200 m/s

not including the uncertainty in the conversion to 2200 m/s. The uncertainties in the epithermal correction and in the neutron temperature are not important, but those in the g factors are. Westcott et al.[4] assigned an uncertainty of 0.2% to g_{η} corresponding to $\pm 1.3\%$ in g_{α} i.e. to ± 0.0024 in a. This estimate was based on the spread of g factors listed in the available compilations; the recent analytical fits of Fluharty et al.[18] give g factors lying within these limits.

The present value of a_5 is significantly higher than the value of 0.1715 ± 0.0015 due to Cabell and Slee (see Table VIIIa of [4]) and that of 0.1718 ± 0.0006 reported by Okazaki et al. [16]. More recent unpublished work by Lisman et al.[19] also gives a lower value, 0.1717 ± 0.0015. These uncertainties exclude g-factor errors. The explanation for the discrepancy cannot lie in the conversion to 2200 m/s - similar g and s values were used in all cases - but must be due to either to unsuspected experimental errors, or to differences between actual and assumed neutron spectra. The present experiment is believed to have the advantage in this respect since the neutron spectrum was more completely thermalized. Furthermore, the simultaneous sub-cadmium irradiations essentially eliminated uncertainties in epithermal capture (epithermal fission is much less important). A value of 0.173 ± 0.004 was obtained earlier from fuel-rod analyses [2]; the large uncertainty is due to the relatively poor knowledge of the neutron spectrum.

With a ²³⁵U absorption cross-section of 680.57 ± 2.70 b at 2200 m/s (weighted mean of transmission measurements corrected for scattering from Table IVb of ref [4]) the integrated fluxes are $0.3240 \pm$ 0.0035 n/kb and 0.7123 ± 0.0038 n/kb for the first and second irradiations respectively, where the flux is the Westcott-convention product (total neutron density X 2200 m/s). The epithermal corrections do not introduce any appreciable uncertainty; the effect of errors in neutron temperature will be considered later in connection with the plutonium irradiation.

b. Uranium irradiations - cadmium covered samples

It follows from the definition of r (see [8] especially equation 3b) that the cadmium-covered rate of production of ^{236}U per ^{235}U atom in a l/E spectrum is

$$nv_0(2/\sqrt{\pi})(r\sqrt{T/T_0}) \int_0^{\infty} \theta\sigma dE/E = nv_0\sigma(Cd)$$
, say,

where (nv_0) is the Westcott-convention flux and θ is the (energy-dependent) transmission of the cadmium cover. The relation between this integral and the resonance integral will be discussed below, along with the effect of departures from a 1/E shape.

From the measured changes $in^{236}U/^{235}U$ ratios under cadmium, and the values of $\int nv_0 dt$ from section (a) above, values of $\sigma(Cd)$ equal to 2,853 b and 2,852 b were obtained from the first and second irradiations respectively. With $r\sqrt{T/T_0} \approx 0.019$ these give $\int_0^{\infty} \sigma \sigma dE/E \approx 133.0$ b, assuming a 1/E shape.

The energy dependence of the 235 U radiative capture crosssection was taken from the measurements of Shore and Sailor [20] and the value of the correction term that allows for the non-ideal behaviour of cadmium

 $\left[\int_{0.5eV}^{\infty} \sigma \, dE/E - \int_{0}^{\infty} \theta \sigma \, dE/E\right]$

was calculated on the extreme assumptions of (i) a small uranium sample at the centre of a spherical cadmium shell 0.8 mm thick, and (ii) an infinite thin layer of uranium between two infinite cadmium sheets 0.8 mm thick, the actual geometry being intermediate between these two extremes. The results were, respectively, 1 b and 5 b, and a compromise correction cf 3 b was therefore applied to give a value of 136 b for the resonance integral above 0.5 eV.

The experimental uncertainty is almost entirely associated with the epithermal flux. The value of $r\sqrt{T/T_0}$ is probably accurate to $\pm 4\%$, and the uncertainty in the interpretation of the cadmium cut off is ± 2 b; the resulting (quadrature-sum) error is ± 6 b.

The effect of the departure from 1/E shape (a deficiency of higher energy neutrons) was estimated to be an underestimate of the resonance integral by $7 \pm 3b$. The corrected result is accordingly

 $\int_{0.5eV}^{\infty} \sigma dE/E = 143 \pm 7 b$

This value agrees well with those of Feiner and Esch [21, 22] 148 \pm 7 b, and Conway and Gunst [21, 23], 133 \pm 7 b. Measurements with monokinetic neutrons now appear to be giving similar results [24, 25].

c. Plutonium irradiation

The value of a for ²³⁹Pu is obtained by comparing the number of atoms of capture products produced with the number of ²³⁹Pu atoms destroyed. Furthermore, since the integrated flux is available from the

TABLE IV

	σ _a (²⁴¹ Pu)	a(²⁴¹ Pu)	$\sigma_a^{(242}Pu)$
2200 m/s value	$1371.8 \pm 16.9 b(a)$	$0.3574 \pm 0.0149(a)$	19.8 ± 1.1 b(b)
Effective value	1454.7 b (c)	0.3367(c)	45.6 b(b)
Uncertainty in effective value	± 2.5%	± 7%	± 10%
Effect on $\hat{a}(^{239}Pu)$	± 0.28%	\pm 0.35%	± 0.82%
Effect on $\hat{\sigma}(^{239}Pu)$	± 0.05%	\pm 0.17%	± 0.38%
Effect on $\hat{\sigma}_{f}(^{239}Pu)$	± 0.13%	\pm 0.27%	± 0.62%

Effect of Input Data on Results of Plutonium Irradiation

(a) K. Ekberg, private communication (1966); least squares fit as [4] but including recent results on $a(^{241}Pu)$ by Cabell and Wilkins (this conference). (b) Cross section and resonance integral values as in [26].

(c) g factors from [27], s factors from [28] adjusted for changed g factors and for more recent resonance integral data [29, 30].

accompanying uranium sample, the destruction of 239 Pu gives a measure of its absorption cross-section relative to that of 235 U. Ideally the reference isotope, 242 Pu, would be unaffected by the irradiation, so that the decrease in the $(^{239}$ Pu/ 242 Pu) ratio would measure the destruction of 239 Pu, and the increase in $(^{240}$ Pu + 241 Pu)/ 242 Pu the amount of radiative capture. In practice corrections are necessary for the destruction of the 241 Pu, representing a loss of 239 Pu capture products, and for changes in the 242 Pu content resulting from capture in 241 Pu, and 242 Pu. In this irradiation 95.1% of capture in 239 Pu remains as 240 Pu + 241 Pu, and 3.2% of the 242 Pu is destroyed, but an amount corresponding to 2.7% of its initial concentration is produced. Of the 240 Pu + 241 Pu initially present 18% is destroyed; expressed as a correction to the amount of 240 Pu + 241 Pu produced this amounts to 2.3%.

These corrections are calculated from the known integrated flux with assumed values for $\hat{\sigma}_1$, $\hat{\sigma}_{C1}$ and $\hat{\sigma}_2$, the values of $\hat{\sigma}_9$, $\hat{\sigma}_{C9}$ and $\hat{\sigma}_0$ being themselves derivable from the observed change in isotopic composition. ($\hat{\sigma}_1$ represents the absorption cross-section of ²⁴¹Pu, $\hat{\sigma}_{C1}$ its capture cross-section, etc.) Table IV lists the input data, and the effects of their estimated uncertainties on the accuracy of the results. The uncertainties in the mass spectrometer analyses produce uncertainties of $\pm 0.66\%$ in $\hat{\alpha}_9$, $\pm 0.31\%$ in $\hat{\sigma}_9$, and $\pm 0.50\%$ in $\hat{\sigma}_{f9}$. The results are $\hat{\alpha}_9 = 0.4138 \pm 0.0047$, $\hat{\sigma}_9 = 1188.5 \pm 9.0$ b, and $\hat{\sigma}_{f9} = 840.6 \pm 8.4$ b, where the cross-section values include a correction of 0.6% to allow for the flux depression in the plutonium sample.

The conversion to 2200 m/s values was made with g factors derived from fits to more recent $\sigma(E)$ data than were available to West-cott [28]. Table V lists these g factors, at 20^oC for convenience of comparison with other tabulations; their temperature dependence is known well enough. The s₄ factors were taken from Critoph [17] and slightly modified for the changed g factors. At 47^oC the (g+rs) factors
TABLE V

Source of data	g _a	g _f	g_a/g_f
Westcott [28, 17]	1.0723	1,0487	1,0225
Fluharty et al (a)	1.0818	1.0571	1.0234
BNW Data 🚺 (b)	1.0793	1.0535	1.0245
Library [32] 🕻 (c)	1.0817	1,0571	1,0233
(a)	1.0805	1.0553	1.0239
Adopted (mean of	4 - 1		
(a) and (d))	1.0812	1.0562	1.0237

A Comparison of ²³⁹Pu g factors at 20^oC

(a) Integration by G.C. Hanna of analytical fits of [18]

(b) Integration by G.C. Hanna of smooth curves through data points.

(c) Private communication from R.C. Liikala.

(d) Mean of (b) and (c). Differences attributed to differences in smoothing procedures.

NOTE: The data of [32] are based on the careful fitting by Leonard [33].

are (1.1073 + 0.0562) for absorption and (1.0755 + 0.0435) for fission, and give

 $a_{9} = 0.3597 \pm 0.0046$ $\sigma_{9} = 1021 \pm 12 \text{ b} \quad \text{at } 2200 \text{ m/s}$ $\sigma_{f9} = 751 \pm 10 \text{ b}$

where the errors now allow for uncertainties in neutron temperature and epithermal index, and in the shape of the slowing down spectrum near cut off. These spectrum-shape uncertainties were taken to be \pm 50% of the (roughly 1%) differences that would result from the use of the s₂ rather than the s₄ factors of [17]. The temperature was known to \pm 6°C; this produces an uncertainty of \pm 0.5% in a₉, and, allowing for the temperature variation of the ²³⁵U absorption cross-section, uncertainties of \pm 0.7% in σ_9 and \pm 0.6% in σ_{f9} .

The uncertainties in the resonance integrals are unimportant, and those in g_a and g_f , tentatively set at $\pm 0.3\%$, do not appreciably increase the errors of the cross-sections. The uncertainty in $g_{1+a}(=g_a/g_f)$ appears to be considerably less than this, to judge from the spread of values in Table V. It may be noted that the measurements by Smith et al. [31] of the variation of η with neutron energy support the cross-section data from which the present g values were obtained. An uncertainty of $\pm 0.2\%$ in g_{1+a} would produce an uncertainty of $\pm 0.76\%$ in a.

The present value of a_9 is in good agreement with the value obtained by Cabell and Slee, 0.356 ± 0.009 as recalculated by Westcott et al.([4] Table VIIIa). The measurements of the isotopic composition of plutonium produced in reactor fuel rods [3] gave a value of 265 ± 7 b for σ_{c9} , also in agreement with the present results.

Regarding σ_9 , the result of 1021 ± 12 b is in satisfactory agreement with the value of 1006.6 ± 6.4 b obtained by Westcott et al. ([4] Table IVc) as a weighted mean of the available transmission

measurements corrected for scattering. If this value of σ_9 is combined with the present value of a_9 a fission cross-section of 740 ± 6 b is obtained.

This result for σ_f , and the one of 751 ± 10 b given above, may be compared with the recent direct monokinetic measurements of 723 ± 15 b by White et al.[5] and 738 ± 7 b by Fraysse and Prosdocimi [34]. It may be noted that the value of 751 ± 5 b attributed to Bigham et al.[35] in [5] is obtained from the Bigham ²³⁹Pu/²³⁵U ratio and ignores the comparison with gold which gave 742.7 ± 5.6 b (see Table V of ref [4]). These Bigham values would be reduced by 0.7% if the g_f factors of the present paper were used. Altogether, because of new measurements [5] and changes to g factors, the value of 742.2 ± 3.5 b from the least-squares fit of [4] is probably somewhat too high.

Summary

The value of a for 235 U has been measured as 0.1765 ± 0.0015 . for 2200 m/s neutrons. This is higher than other recent measurements, but the neutron spectrum is believed to be better known in the present experiment. The resonance capture integral has been measured to be 143 ± 7 b above 0.5 eV. This is in good agreement with other results.

For ²³⁹Pu 2200 m/s values of $a = 0.3597 \pm 0.0046$, $\sigma_a = 1021 \pm 12$ b, and $\sigma_f = 751 \pm 10$ b were obtained. If the value of σ_a from transmission measurements, 1006.6 ± 6.4 b, is combined with the present a measurement, a value of 740 ± 6 b is obtained for the fission cross-section.

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MEASUREMENTS OF THE FISSION CROSS-SECTIONS OF ²³⁹Pu AND ²⁴¹Pu RELATIVE TO THAT OF ²³⁵U IN THE NEUTRON ENERGY RANGE 0.016 TO 0.55 eV

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Abstract

MEASUREMENTS OF THE FISSION CROSS-SECTIONS OF 239 Pu AND 241 Pu RELATIVE TO THAT OF 235 U IN THE NEUTRON ENERGY RANGE 0.016 TO 0.55 eV. Measurements have been made to an accuracy of 229 wo of the ratios of the fission cross-sections of 239 Pu and 241 Pu to that of 235 U in the energy range 0.016 to 0.55 eV and also in two thermalized fields of neutrons.

The monoenergetic neutrons were obtained from a crystal spectrometer on the AWRE reactor HERALD. At the three lowest neutron energies a velocity selector was used to remove neutrons diffracted into the second and higher orders. At the other energies resonance filters were used. One thermal neutron field was an extracted beam from the graphite thermal column and its spectrum was measured by a time-of-flight method. The other thermal neutron field was in a cavity inside the thermal column.

Some discrepancies between the present results and previous measurements are discussed. Corrections and errors due to the neutron spectra of both the monoenergetic neutrons and the thermal beam and errors introduced by beam non-uniformity are described.

1. Introduction

The fission cross-sections of ²⁹⁹Pu and ²⁴¹Pu in the thermal and epithermal regions are of importance in reactor design and have been measured many times using a variety of techniques. Summaries of these measurements have been made by Stehn et al. [1], by Westcott et al. [2] and by Hughes et al. [3,4]. All these measurements have been relative to the fission cross-section of ²⁹⁵U or to the capture cross-section of gold. The results for a particular cross-section sometimes differ by up to 10% and may differ depending upon whether monoenergetic neutrons or a broad spectrum of neutrons has been used for measurement. In view of these uncertainties it was considered worthwhile to remeasure the fission cross-sections of ²³⁹Pu and ²⁴¹Pu relative to ²³⁵U in monoenergetic neutron fluxes of energies 0.016, 0.0253, 0.051, 0.117, 0.161, 0.27 and 0.545 eV, in a broad spectrum of neutrons in a cavity in a graphite thermal column and in collimated beam of neutrons extracted from the thermal column.

2. Monoenergetic neutron sources

A monoenergetic beam of neutrons of energy in the range 0.016 to 0.55 eV was obtained from a crystal spectrometer used in conjunction with the light-water-moderated reactor HERALD at AWRE [5,6].

Fig. 1 shows the experimental arrangement. The mean angle of diffraction of the aluminium crystal was adjusted to give the required neu-



POSITION A. IN A MONOENERGETIC NEUTRON BEAM POSITION B. IN AN EXTRACTED BEAM OF THERMAL NEUTRONS POSITION C. IN A THERMAL NEUTRON FLUX INSIDE THE THERMAL COLUMN NEUTRON SPECTRA A AND A' ARE SHOWN IN FIGURE 2 NEUTRON SPECTRUM B IS SHOWN IN FIGURE 5



FIG.1. Apparatus used for fission cross-section ratio experiments

FIG. 2. Neutron spectra from crystal spectrometer (A) and at position of fission counters (A'); E = energy of first-order beam of neutrons

TABLE I

Details of mono chromating system

1. Crystal Spectrometer	
Number of slits in in-pile collimator	3
Neutron beam divergence of in-pile collimator	30' FWHM
Mosaic spread of aluminium diffracting crystal	20' FWHM
Number of slits in external collimator	11
Neutron beam divergence of external collimator	10' FWHM
2. Velocity Selector	
Number of rotating discs	3
Separation of discs	40 cm
Thickness of cadmium on each disc	0.05 cm
Number of slits on each disc	50





tron energy using the known crystal lattice spacing (2.3333 Å for the 111 plane [7]) in the Bragg equation for first order diffraction $\lambda = 2d \sin \theta_{\rm B}$. However the aluminium diffracting crystal produced a neutron spec-

trum containing many orders of diffraction, each order having an unknown resolution function. The spectrum is shown diagrammatically in Fig. 2. It was known from previous work that the neutron flux in the second and higher order diffracted beams was comparable with the flux in the first order beam and to produce a monoenergetic neutron beam suitable for the fission cross-section ratio measurements it was necessary to attenuate the second and higher order neutrons.

2.1 Attenuation of the second and third order neutrons

At the neutron energies 0.016, 0.0253 and 0.51 eV a three disc mechanical velocity selector shown in Fig. 1 was used to attenuate the second order diffracted beam by a factor of more than 100 and except at 0.051 eV to attenuate the third order beam by a factor of more than 300. Details of the velocity selector are given in Table I. The angular velocity was electrically stabilised to within \pm 0.5% of its nominal value. Fig. 3 shows the count rate in a BF₃ counter which was exposed to neutrons transmitted by the velocity selector. The primary neutron energy of 0.0253 eV was set by the diffraction angle of the crystal spectrometer. After correction for the variation of efficiency with neutron energy of the BF₃ counter the ratio of the neutron flux in the second order to that in the first order can be derived. Table II shows this ratio for the three energies mentioned and also the neutron flux that was available for the measurement of the fission cross-section ratios.

Between 0.117 and 0.55 eV the second and third order diffracted neutron beams lie at energies greater than the cadmium cut-off as also does the third order beam at 0.051 eV primary neutron energy. For these energies the velocity filter provided negligible attenuation of the higher orders and resonance filters were used. The precise neutron energies were determined by the availability of resonance absorbers which would provide reasonable attenuation of the second and third order neutron

TABLE II

Measured intensity of the second and third order diffracted neutrons' relative to the first order and the filters used

First Order			Second Order			Third Order		
Neutron Energy (eV)	Neutron Energy Spread (%)	Neutron flux (n cm ⁻² s ⁻¹)	Neutron Energy (eV)	Filter	Intensity Relative to 1st Order (%)	Neutron Energy (eV)	Filter	Intensity Relative to 1st Order (%)
0.016	1.0	0.3 × 10 ⁵	0.064	Velocity selector	290	0.144	Velocity selector	NM
0.0253	1.4	1.0 × 10 ⁵	0.101	Velocity selector	91	0. 228	Velecity selector	NM
0.051	4.0	1.8 × 10 ⁵	0.204	Velocity selector	<30	0.459	Erbium	NM
0.117	6.6	3.4×10^{6}	0.468	Erbium	3.5	1.05	Hafnium	0.75
0.161	7.8	1.4×10^{6}	0.644	Indium	4.6	1.449	Indium	1.6
0.270	10.2	1.0 × 10 ⁶	1.08	Hafnium	10	2.43	Hafnium	NM
0.545	14.4	0.8 × 10 ⁶	2.180	Rhenium	~15	4.90	Gold	NM

NM Not measured.

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beams while not appreciably attenuating the first order beam. Absorber thicknesses were chosen to attenuate the second and third order beams by factors of 100 and 30 respectively more than the attenuation of the first order beam. With each pair of absorbers at the end of the external collimator scans of the crystal spectrometer across the chosen energies produced transmission dips in reasonable agreement with that calculated from the known cross-sections and thicknesses of the absorbers. Table II includes the resonance filters used and the measured amounts of second and third order beams relative to that of the first order. These ratios



were found in a subsidiary experiment by using cadmium and the resonance filters to eliminate successively the first, second and third order neutrons. A measurement by time-of-flight of the order contamination from an aluminium crystal similar to the one used in the crystal spectrometer gave 120%, 12% and 7% for the second, third and fourth orders relative to the first order respectively at 0.020 eV primary neutron energy. The amount of the second order is in reasonable agreement with that measured by the velocity selector. The effect of the fourth and higher order diffracted beams which are not attenuated by the velocity selector or the resonance filters will be discussed in Section 4.

2.2 Neutron energy spread

The energy resolution width on the first order diffracted beam of neutrons can be calculated from the diffraction angle θ_{p} , the acceptance

angles of the collimators and the mosaic spread of the crystal and is included in Table II. Multiple scattering in the aluminium crystal produces an asymmetric resolution function and hence the mean energy of the neutron beam differs from the neutron energy at the mean collimator angle. This difference is of no significance at the lower neutron energies where



FIG.5. Energy spectrum of extracted thermal neutron beam. Each spectrum normalized to unit area — Spectrum measured by a time-of-flight method

--- Theoretical Maxwellian neutron spectrum of temperature 50°C

--- Theoretical Maxwellian neutron spectrum of temperature 100°C

.... Theoretical Maxwellian neutron spectrum of temperature 150°C

the energy spread is small but at the three highest energies the resolution function was measured using a second analysing crystal. The mean neutron energy was calculated from the known variation of fission crosssections with energy and the measured resolution function.

2.3 Neutron beam profile

A profile of the 3.8×7.5 cm neutron beam was measured at each neutron energy by scanning a 0.2-cm diameter hole in a 5-cm thick block of borated wax across the neutron beam at the position at which the fission cross-section ratios were measured. The neutrons were detected in a BF₃ counter mounted behind the 0.2-cm hole. Fig. 4 shows profiles in the horizontal plane at two neutron energies. The structure observed is due partly to the in-pile and external collimators and partly to attenuation effects in the crystal. Beam profiles in a vertical plane were measured to be of uniform intensity to within ± 5% over the region of the fissile foils.

3. Thermal neutron sources

3.1 Extracted thermal beam

A thermal beam of neutrons 7 cm in diameter was extracted from the graphite thermal column on HERALD. The neutron spectrum was measured by a time-of-flight method similar to the one described by Reichelt [8] and is shown in Fig. 5. Also shown in the figure are several theoretical spectra having Maxwell-Boltzman distributions of energies. It was found that a theoretical spectrum of temperature $90^{\circ}C$ approximated best to the observed neutron spectrum. The neutron temperature is significantly higher than the graphite temperature of $20 - 40^{\circ}C$ mainly because the neutron beam is the leakage flux rather than the internal flux of the thermal column. A check on the neutron temperature was obtained by measuring the transmission of a 2.65 g cm⁻² gold sample [9,10]. The value found was $90 \pm 10^{\circ}$ C in good agreement with the temperature measured by time-of-flight.

The flux of neutrons of energy greater than ~ 0.6 eV was determined by measuring the cadmium ratio for a BF₃ counter and showed that the epithermal flux per unit lethargy as a fraction of the thermal flux (the epithermal index, r in the Westcott convention) was 0.001 ± 0.0003 . Measurements with counters containing ²³⁵U, ²³⁹Pu and ²⁴¹Pu gave similar ratios but with larger errors due to the low count rates involved.

The profile of the thermal beam is included in Fig. 4.

3.2 Thermal neutron source inside the thermal column

For fission ratio measurements inside the graphite thermal column of HERALD the fission counters were introduced into the 11.5-cm diameter access hole (J2 facility) in the thermal column. This hole is ~ 3 m long and curved over the outer 1.5 m to prevent neutron streaming along the tube and into the reactor hall. The counters were inserted to a position such that at least 60 cm of graphite surrounded the counters in all directions. To reduce neutron flux gradients along the hole, 15-cm long graphite plugs were placed in the hole in front of, and behind the fission counters. The reactor was operated at the low power of 1 kW in order to maintain a uniform temperature in the thermal column and for the duration of the fission counting the temperature at any position in the thermal column did not differ from the mean value of 30°C by more than 7°C.

The cadmium ratio was measured by enclosing the counters in a 1-mm thick cadmium can and gave for r, the epithermal index, a value 0.001. Fully shielding the counters proved to be a difficult task and small holes may have been present in the cadmium can. Because of this the measured epithermal index must be considered an upper limit to the true value.

4. Measurement of the fission rates

The fission rates were measured by counting fission fragments from thin foils of ²³⁹Pu or ²⁴¹Pu and ²³⁵U placed back to back in the neutron beam. The foil backings were thick so that fragments could only be counted in 2π geometry from each foil. Details of the fission counters and foils have been described by White [11], White et al. [12] and Perkin et al. [13]. To reduce the error in the fission cross-section ratio due to the non-uniformity of the fissile deposit two foils of each isotope were used and details of the foils are given in Table III. The fission counters were positioned centrally in the neutron beam to within ± 0.5 mm.

A single measurement with one pair of counters consisted of eight runs each with a statistical counting accuracy of about 1%. The counters were placed in different orientations in each of the eight runs to eliminate effects due to attenuation of neutrons in the foil backings, to displacement of the counters on rotation and to reduce as far as possible errors due to the interaction of the non-uniform neutron flux with the non-uniform beam

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TABLE III

Counton	Nuclide		Mass Number				Assay	Foil Thickness
Counter	Nucilue	234	235	236	238	(mg cm ⁻²)	Error (%)	Correction (%)
1.2	235U	1.19	92.95	0.18	5.68	0.5	1.0	5.9
2.7	295U	0.11	99.33	0.25	0.31	0.5	1.0	5.7
		239	240	. 241	242			
6.7	239Pu	99.985	0.012	0.003	-	0.2	0.5	3.2
6.12	239Pu	99.985	0.012	0.003	-	0.06	0.5	0.7
241.1	²⁴¹ Pu	5.12	5.53	88 .9 5	0.40	0.25	2.0	3.0
241.2	241 _{Pu}	5.12	5.53	88.95	0.40	0.08	2.0	1.1

Fissile foil isotopic compositions (weight %), nominal thickness, assay errors and foil thickness corrections

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fissile deposit. In no case did a single run differ from the mean by more than 3% except at the highest neutron energy where because 1% statistical accuracy was not achieved on each run the maximum difference was 5%. For monoenergetic measurements three sources of neutron background were found to be present at the position of the fission counters. Firstly the stray neutron flux in the reactor hall produced a background fission rate in the fission counters of between 0.3% and 3.0% of that induced by the monoenergetic neutron beams and was measured by attenuating the neutron beam from the crystal spectrometer with a cadmium filter. Secondly there are neutrons in the fourth and higher orders from the crystal spectrometer. The energies of these neutrons are sufficiently high for them to be transmitted through the cadmium of the mechanical velocity selector and through the resonance filters without appreciable attenuation. The cadmium filter placed in the neutron beam to measure the reactor hall background also transmits these high order neutrons and the 0.3 - 3.0% background attributed to the general neutron background in the reactor hall includes fission induced by the fourth and higher order neutrons. No further correction has been made for neutrons in the fourth and higher orders. Thirdly the neutron background from the crystal spectrometer due to incoherent scattering in the aluminium crystal was determined by measurement of the rocking curves for the spectrometer. The fission rate induced by this background was less than 0.5% of the fission rate induced by the monoenergetic neutrons.

Backgrounds were measured to be negligible (<0.1%) for measurements made using thermal neutron spectra. Cross-section ratios measured without the velocity selector or resonance filters in the neutron beam agreed to within 1 - 2% with the ratios calculated from the monoenergetic data and the measured amount of second order diffracted neutron flux and the measured or estimated amount of third order diffracted flux. A check comparison of one ²³⁵U foil with the other gave a result which agreed to 0.3 \pm 1.3% with the ratio of the masses of fissile material on the foils.

5. Experimental errors

A summary of the estimated errors on the fission cross-section ratios is given in Table IV. For ratios measured with monoenergetic neutrons the largest contributions to the total error are from foil assay which has been described by White [11,14] and from beam non-uniformity. The latter source of error occurs when fission events are detected from a non-uniform fissile foil in a non-uniform neutron flux and is small if either the foil thickness or the beam intensity were uniform over the foil area. Analysis of the fission counting data taken with monoenergetic neutrons gave no systematic variation of fission count rate with the orientation of the fission counter and indicated that the error introduced by beam non-uniformity was less than the statistical error of 1%. Other estimates of the error were calculated from the known beam intensity profiles and the measured thickness profiles of other foils prepared by the same painting technique. The maximum error was again 1%. Detailed profiles of the foils used in the present work were not known

TABLE IV

Table of estimated errors (%) on final cross-section ratios

1. Monoenergetic measurements

Source of error	Comment	299 _{Pu} ∕285 _U Ratio	241 _{Pu/} 235 _U Ratio
Weighing and) U assay of foil Pu	Corrections and errors described by White [11,14]	1.07	1.0 2.0
Fission fragments} U absorbed in foil }Pu	Corrections and errors described by White [11,14]	0.5 0.4	0.5 0.4
Extrapolation of fission U fragment spectra to Pu zero pulse height	Corrections and errors described by White [11,14]	0.5 0.4	0.5 0.4
Statistical(except 0.545 eV) Statistical at 0.545 eV		0.5 1.0	0.5 1.0
Isotopic composition	Fission in other isotopes of U and Pu assumed zero except for ³³⁹ Puin ²⁴¹ Pufoil	کا 1مہ 0	0.1
Fourth and higher order neutrons		0.3	0.3
Incoherent background from crystal spectrometer		0.5	0.5
Beam non-uniformity (except for thermal beams)	See text	1.0	1.0
Decay of ²⁴¹ Puin sample since its separation from ³⁴¹ Am	Calculated from $\tau_{K} = 13.24 \pm 0.24$ yrs [15]		0.1
Total error for monoenergetic me	easurements	1.8	2.7

2. Additional errors for thermal measurements

Hardening of neutron spectrum by captive in counter mat- erials	[16]	0.1	0.1
Beam non-uniformity Uncertainty in neutron spectrum	See text	0.1 1.0	0.1 1.0
Total error for thermal measurem	ents	1.8	2.7

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TABLE V

Measured ratios of the fission cross-sections of 23 Pu and 241Pu to that of ²⁸⁵U and the fission cross-sections of ²⁸⁹Pu and ²⁴¹Pu at 0.0253 eV and in a thermal neutron flux

		At 0.0	0253 eV	Neutron Energ	IY .	
Monoenergetic	Fission Cross-	Section	Ratios	Fission Cr	055-5	Sections ^a (b)
Measurements	239 _{Pu} /235U	241 Pu/	2.95U	289Pu		241 Pu
Present work Fraysse and Prosdocimi [17] Leonard [18] Raffle [19] Leonard et al.[20] Tunnicliffe [21] Watenabe and Simpson [22] Raffle and Price [23] Jaffey et al.[24]	$\begin{array}{c} 1.253 \pm 0.022 \\ 1.264 \pm 0.03 \\ 1.23 \pm 0.04 \\ 1.21 \pm 0.04^{\text{b}} \end{array}$	1,772 = 1.618 =	± 0.048 ± 0.07	723 ± 1 729 ± 1 710 ± 2 704 ± 2 680 ± 2 680 ± 6	5 7 3 20 3 0 0	1022 ± 29 935 ± 42 962 ± 38 987 ± 42 1100 ± 30
Deduced from thermal measurements						
Present work (extracted beam Present work (inside thermal	$\begin{array}{c}1.235 \pm 0.022\\1.277 \pm 0.025\end{array}$	1.722 1.782	0.048	713 ± 1 737 ± 1	5 7	994 ± 29 1028 ± 29
Pratt et al.[25] Bigham et al.[16] Raffle [19]	$\begin{array}{r} 1.233 \pm 0.027 \\ 1.307 \pm 0.010 \\ 1.20 \pm 0.04^{b} \end{array}$	$\begin{array}{c} 233 \pm 0.027 \\ 307 \pm 0.010 \\ 20 \pm 0.04^{\text{b}} \end{array} $ 1.780 $\pm 0.010^{\text{c}}$		713 ± 15 751 ± 5 701 ± 20		1014 ± 8 ^d
Compilation values						
Westcott et al.[2] Stehn et al.[1]	1.281 ± 0.008 1.283 ± 0.006	1.741 ±	0.017	742.4 ± 740.6 ±	5.3 3.5	1012 ± 9 950 ± 30
	Neutron Temperature (°C)	-	Ratio	s in Thermal	Neuti	rons Spectrum
Present work (extracted beam) Present work (inside thermal column) Pratt et al.[25] Bigham et al.[16] Raffle [19]	~ 90 30 ± 7 222 20 30		1.40 1.31 1.72 1.40 1.29	$\begin{array}{r} 0.05 \pm 0.020 \\ 0.020 \\ 0.025 \pm 0.027 \\ 0.027 \\ 0.007 \\ 0.007 \\ 0.041 \end{array}$	1.9 1.9 1.8	966 ± 0.048 920 ± 0.047 937 ± 0.010 53 ± 0.07

a ²⁹⁵Ufission cross-section assumed to be 577 barn.

b Deduced from cross-section measurement. c Deduced from ³⁴¹Pu/²³⁹Pu and ²⁸⁴Pu/³³⁵U ratios-d ²⁸⁴Pu fission cross-section assumed to be 742 barns.

but were expected to be similar to other foils painted by the same operator.

The uncertainty in the energy of the monoenergetic neutrons is due mainly to $a \pm 5'$ uncertainty in the direction of the neutrons entering the diffracting aluminium crystal and to a lesser extent upon the uncertainty in the resolution function of the beam of neutrons.

For ratios measured with thermal neutron fluxes the largest single errors are from the foil assay and from uncertainty in the neutron spectrum incident on the counters. The latter error is discussed in more detail in the next section.

6. Results and comments

The measured ratios of the fission cross-sections of ²³⁹Pu and ²⁴¹Pu to that of ²³⁵U for monoenergetic neutrons of 0.0253 eV energy are

TABLE VIMeasured ratios of the cross-sections of 239Pu and 241Pu relative to thatof 235U in the energy range 0.016 to 0.545 eV

Neutron Total	Uncertainty	Cross-Secti ²³⁹ Pu/ ²	on Ratio 350	Cross-Section Ratio 241pu/235U		
(eV)	Spread (%)	ad (%) (eV) Present Other Measurements Measurements		Other Measurements	Present Measurements	Other Measurements
0.016	1.0	± 0,00004	1.208 ± 0.022	1.230	1.681 ± 0.044	1.594
0.0253	1.4	± 0.0001	1.254 ± 0.022	1.279	1.771 ± 0.048	1.663
0.051	4.0	± 0.0005	1.374 ± 0.024	1.438	1.876 ± 0.05	1.858
0.117	6.6	± 0.002	2.010 ± 0.036	2.091	2.719 ± 0.07	2.753
0.161	7.8	± 0.003	2.93 ± 0.05	3.09	3.96 ± 0.10	3.84
0.270	10.2	± 0.007	12.51 ± 0.3	14.25	8.23 ± 0.3	7.58
0.545	14.4	± 0.02	1.94 ± 0.05	1 .6 8	0.806 ± 0.02	0.688

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given in Table V together with the results quoted by other authors. The present results are an improvement on accuracy on previous measurements and agree well with the cross-section ratios quoted by Fraysse and Prosdocimi [17], Leonard [18] and Raffle [19]. If one assumes the cross-section of ²³⁵U to be 577 b [1] then the fission cross-sections of ²³⁹Pu and ²⁴¹Pu deduced from the present work are also shown in Table V together with other measurements of these cross-sections made with monoenergetic neutrons. The present results again agree well with the other measurements.

Also shown in Table V are the fission cross-section ratios measured with thermal neutron fluxes and the fission cross-sections and fission crosssection ratios at 0.0253 eV calculated from the measured ratios. For the measurements made using the extracted beam of thermal neutrons the g-factors* required to transform the measured fission cross-section ratios to ratios at 0.0253 eV were calculated from the known variation of the fission cross-sections with energy and the neutron spectrum measured by a time-of-flight method. These factors were 1.138 ± 0.01 and 1.142 ± 0.01 for 239 Pu and 241 Pu relative to 235 U respectively. The Maxwellian neutron spectrum of temperature 90°C which gives the best visual fit to the measured neutron spectrum has corresponding g-factors of 1.151 and 1.132 [26,27]. The residual discrepancy of ~ 1% is probably due to the measured neutron spectrum not having a Maxwell-Boltzman distribution of energies as is normally assumed.

The g-factors used to convert the fission cross-section ratio measurements made inside the thermal column to ratios at 0.0253 eV have been calculated assuming the neutron spectrum to be Maxwellian with a neutron temperature equal to the thermodynamic temperature of the graphite. The fission cross-section ratios measured in thermal neutron fluxes, when corrected to 0,0253 eV agree within error with the ratios measured with monoenergetic neutrons and agree within slightly wider limits with the results of Bigham et al. [16]. Pratt et al. [25] and Raffle [19] which were also determined in thermal neutron fluxes. The average of the three fission cross-section ratios measured with thermal neutron fluxes and with monoenergetic neutrons gives 1.255 \pm 0.022 and 1.760 \pm 0.048 for the ratios ²³⁹ Pu to ²³⁵U and ²⁴¹ Pu to ²³⁵U respectively. These ratios agree satisfactorily with the compilation values of Westcott [2] but the measured fission cross-section ratio of 241 Pu relative to 235 U is significantly higher than the assessed value of Stehn [1]. The error on the average of the three results has not been reduced by averaging since most of the errors in the present work are common to the three ratio measurements.

The fission cross-sections and fission cross-section ratios at other monoenergetic neutron energies are given in Table VI. Also included in the table are ratios deduced from the assessments of Stehn et al. [1] and of Hughes et al. [4]. These assessments represent an average of the

^{*}For a description of the g-factors of the Westcott conventions see Westcott [26] or Beckurtz and Wirtz [10].

results of many authors and are for practical purposes normalised to the 'recommended' cross-sections at 0.0253 eV.

At all seven neutron energies within error the present results agree with the ratios deduced from these assessments as would be expected if present and previous measurements have the same variation of fission cross-sections with neutron energy. The larger discrepancies at the two highest neutron energies are probably due to the uncertainty in the mean neutron energy.

7. Acknowledgments

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DISCUSSION

(on papers CN-23/21, CN-23/2 and CN-23/58)

C.H. WESTCOTT: With reference to our two surveys of 2200 -m/s. constants^{1,2}, I would like to report that the IAEA has made a revision for ²⁴¹Pu, and an even fuller revision for the other three fissile nuclides too will probably be made in 1967. Difficulty arises in estimating the accuracy of g-factors (as mentioned in paper CN-23/2), and this may affect the date of the revision. A new fitting is needed for ²⁴¹Pu as the values available at present are not very accurate. The results of the recent revision for ²⁴¹Pu (2200 m/s) are as follows:

σ_{a}	=	1372	Ъ	$\overline{\nu}$ = 2.969
$\sigma_{\rm f}$	=	1011	b	$\eta = 2.187$
σγ	=	361	b ·	$\alpha = 0.357$

The changes since the 1965 survey² are due to the inclusion of the new results obtained by Cabell et al. and the withdrawal of their earlier values. Our values in fact now lie very close to those given in the 1964 survey¹. It should be noted that the σ_f value also agrees with the mean of the values reported by White et al. in paper CN-23/58.

M.J. CABELL: The fact that the recommended values for the 2200-m/s constants for ^{241}Pu given in the 1965 paper by Westcott et al. were greatly influenced by our preliminary results for alpha is mentioned in the paper. This measurement is particularly difficult: ²⁴²Pu cannot be used as a reference nuclide for the measurement since it is the product of neutron capture by ²⁴¹Pu. The ²³⁹Pu content of the sample must be used instead and this is itself, of course, greatly affected by neutron irradiation. The preliminary value for alpha, obtained using reactor spectra, has been withdrawn, as Dr. Westcott just said. It was wrong for two reasons. Firstly, as stated in my paper, a correction for the amount of ²⁴²Pu formed via ²⁴¹Am was omitted and secondly the results were heavily weighted in the light of simultaneous measurements made on the destruction of ²³⁹Pu under similar experimental conditions. We now believe this weighting was unjustified. That these changes in the value of alpha have a direct effect on the recommended values resulting from the least-squares fitting procedure for all the 2200 - m/s data emphasizes the value of these measurements, particularly when other data are sparse, as in this case.

It is a pleasure to hear that Dr. Westcott hopes to derive a new set of recommended values in the near future taking into account the latest results. His comments that the accuracy of g-values is being looked at again are particularly welcome since these values can be important sources of error.

¹ WESTCOTT, C.H. et al., "Survey of nuclear data for reactor calculations", Proc. 3rd UN Int. Conf. PUAE II (1965) 412.

² WESTCOTT, C.H. et al., "A survey of values of the 2200 m/s constants for four fissile nuclides", Atomic Energy Rev. 3 2 IAEA, Vienna (1965) 3.

РЕАКЦИЯ n, nf И ВЫХОДЫ ЗАПАЗДЫВАЮЩИХ НЕЙТРОНОВ

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Abstract — Аннотация

THE n, nf REACTION AND DELAYED NEUTRON YIELDS. The author has studied the effect of the n, nf reaction on the yield ratio of delayed-neutron groups. The experiments were carried out on a Van de Graaff generator. A solid zirconium-deuterium target of thickness 20 mg/cm² was used. The maximum energies of the neutron spectrum were in the 5-7.75 MeV range. The decay curves were analysed on a digital computer for given values of half-life - 55, 24, 15, 5, 5.2 and 2.2 s. The neutron background, after taking off the voltage on the generator, made it impossible to isolate shorter groups. Errors were calculated according to the inverse matrix by the normal method. The two elements which have so far been studied in most detail are uranium-238 (7 points in the above-mentioned energy range) and thorium-232 (9 points). The results are presented in the form of the ratio of the i-th group of delayed neutrons to the first group (half-life 55 s). The way in which this ratio varies as the energy increases is the same in both elements (for corresponding groups). In both cases the ratio of the yield from each of the groups with half-lives of 24 and 5.2 s, in the energy region where the n, nf reaction occurs, has the shape of a resonance curve. The ratio of the 15.5-s group to the first group shows a clearly defined minimum, while the ratio of the 2.2-s group shows a more complex structure with maxima and minima; this is possibly due to the fact that it is a mixture of the two groups.

РЕАКЦИЯ n, nf И ВЫХОДЫ ЗАПАЗДЫВАЮЩИХ НЕЙТРОНОВ. Изучалось влияние реакции n, nf на отношение выходов групп запаздывающих нейтронов. Опыты проводились на генераторе Ван-де-Граафа. Использовалась твердая цирконий-дейтериевая мишень толщиной 20 мг/см². Максимальные энергии спектра нейтронов в данных опытах были в интервале 5 - 7,75 Мэв. Разложение кривых распада производилось на ЦВМ при заданных значениях периодов полураспада: 55; 24; 15,5; 5,2 и 2,2 сек. Фон нейтронов после снятия напряжения на генераторе не позволял выделить более короткие группы. Ошибки вычислялись по обратной матрице общепринятым способом. В настоящее время наиболее подробно изучены два элемента: уран-238 (7 точек в указанном интервале энергий) и торий-232 (9 точек). Результаты представлены в виде отношения і -й группы запаздывающих нейтронов к первой (период полураспада 55 сек). Характер изменения этого отношения с ростом энергии у обоих элементов для соответствующих групп одинаков. У обонх элементов отношение выхода для каждой из групп с периодами полураспада 24 и 5,2 сек. В области энергий, где идет реакция n, nf имеет вид резонансной кривой. Отношение 15,5 сек. группы к первой дает резкий минимум, а отношение 2,2 сек. группы проявляет более сложную структуру с минимумами и максимумами; возможно, это связано с тем, что она представляет смесь двух групп.

Изучалось вдияние реакции n, nf на отношение выходов групп запаздывающих нейтронов из тория-232 и урана-238 (диапазон знергий нейтронов, вызывающих деление 5 – 7,75 Мэв). Опыты проводились на генераторе Ван-де-Граафа. Использовалась твердая цирконий-дейтериевая мишень толщиной 20 мг/см². Разложение кривых распада производилось на цифровой вычислительной машине при заданных значениях постоянных распада. Округленные значения периодов полураспада, принятые в данных расчета, составляют 55; 24; 15,5; 5,2; 2,2 сек.

Период полураспада, сек		55	24	15,5	5,2	2,2
	5	I	2,385±0,072	2,42 ±0,10	4,87 ±0,16	10,13±0,34
B	6	I	2,54 ±0,08	2,32 ±0,09	4,79 ±0,16	10,71±0,26
бодо	6,2	I	3,18 ±0,18	1,43 ±0,25	8,07 ±0,45	7,74±0,80
BBI	6,4	I	2,329±0,043	$2,319 \pm 0,063$	4,44 ±0,10	10,84±0,20
ие	6,6	I	$2,423 \pm 0,056$	2,288±0,083	4,370±0,13	10,10±0,27
пен	6,8	I	2,23 ±0,07	2,29 ±0,08	3,54 ±0,11	$11,66 \pm 0,27$
ILHOI	7,25	I	$1,969 \pm 0,045$	2,113±0,065	3,70 ±0,11	6,94 ±0,21
0	7,5	I	1,85 ±0,05	$2,12 \pm 0,07$	3,04 ±0,09	8,87 ±0,20
	7,75	I	1,474±0,087	2,38 ±0,13	2,47 ±0,21	7,91 ±0,52

ТАБЛИЦА 1. ТОРИЙ-232 (Энергия нейтронов, Мэв).



1 2 3 4 5 6 7 8 En (Mas)

Рис. 1. Значения выходов, отнесенные к выходу группы с периодом полураспада 55 сек для тория-232.



Рис. 2. Значения выходов, отнесенные к выходу группы с периодом полураспада 55 сек для урана-238.

В связи с тем, что мишень толстая, полученные значения отношения выходов есть

$$R_{i9} = \int_{E\min}^{E\max} \sigma_f(E) \Phi(E) a_i(E) dE / \int_{E\min}^{E\max} \sigma_f(E) \Phi(E) a_i(E) dE,$$

где R_{іэ} — полученное при разложении кривой распада нейтронов отношение выхода і-й группы к первой (с периодом полураспада 55 сек),

σ_f - сечение деления,

Ф(Е) dE - поток нейтронов,

 а_i(E) – абсолютный выход i-й группы запаздывающих нейтронов при делении нейтронами с энергией Е.

В табл.1 приведены значения R_{i_3} для обоих элементов. Эти же данные представлены на рис.1, 2 и 3 (точки отнесены к максимальной энергии спектра нейтронов, σ_f - кривая сечения деления).



Рис. 3. Значения выходов, отнесенные к выходу группы с периодом полураспада 55 сек для урана-238.

ТАБЛИЦА 2.	УРАН-238	(Энергия	нейтронов,	Мэв).
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Период полураспада, сек		55	24	15,5	5,2	2,2
_	5	I	7,56±0,17	3,55±0,16	10,7 ±0,3	27,3 ±0,7
BOB	6	I	6,93±0,31	3,81±0,15	9,18 ±0,66	30,69±0,78
TOX	6,4	I	$7,95 \pm 0,18$	$2,60 \pm 0,16$	10,2 ±0,3	19,2 ±0,6
Bh	6,6	I	7,19±0,32	$3,76 \pm 0,32$	11,48±0,54	24,9 ±1,3
ние	6,8	I	12,31±0,37	0,59±0,23	13,3±0,6	45,7 ± 3,4
olie	6,9	I	$8,42 \pm 0,24$	3,16±0,20	11,6 ±0,4	24,4 ±0,8
HT O	7,1	I	8,30 ±0,20	$3,54 \pm 0,19$	10,39±0,32	25,16±0,70
Ŭ	7,3	I	$7,95 \pm 0,15$	24,0 ±1,4	$8,68 \pm 0,25$	14,0 ±0,5
	7,5	I	8,08±0,14	20,6 ±1,2	8,64±0,22	13,3 ± 0,43
	7,8	I	7,32±0,42	$3,74 \pm 0,42$	7,04±0,73	28,0 ±2,0

Видно, что там, где начинает идти реакция n, nf, наблюдается резко выраженная структура в отношениях выходов групп. Ясно, что эффект следует приписать второму делящему ядру (торию-232 и урану-238).

Интерпретация явления осложняется отсутствием данных по распределению масс и заряда в этой области энергий и недостаточно надежной химической идентификацией.

Предварительные данные по распределениям масс (для урана-233 и урана-235) показывают резкое изменение в отношениях выходов в области масс, приписываемых родоначальникам запаздывающих нейтронов. Непосредственно на ступеньке отношение выходов масс (85 – 95 и 135 – 145) к массе 87 уменьшается и нигде не наблюдается противоположный эффект, имеющий место для некоторых групп запаздывающих нейтронов. По-видимому, следует ожидать для этих осколков резкое изменение в распределении заряда.

Для трех точек торий-232 (6; 6,2; 6,4 Мэв) было проведено разложение кривых распада при заданных значениях периодов полураспада, варьируя последние по таблицам случайных величин при заданных значениях ошибок (всего проделано 10 вариантов). Результаты показывают, что эффект сохраняется.

ЛИТЕРАТУРА

[1] ВОРОБЪЕВА В.Г. и др. Многопараметровые исследования осколков при делении урана-235 тепловыми нейтронами и нейтронами с энергией 5, 6 и 7 Мэв. "Ядерная физика" (в печати).

ANALYTICAL DESCRIPTION OF NEUTRON CROSS-SECTIONS AND THE EFFECT OF THEIR ENERGY DEPENDENCE UPON THEIR 2200-m/s VALUES

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Abstract

ANALYTICAL DESCRIPTION OF NEUTRON CROSS-SECTIONS AND THE EFFECT OF THEIR ENERGY DEPENDENCE UPON THEIR 2200-m/s VALUES. In the calculation of reaction rates in a reactor, the energy dependence of the cross-sections involved must be known with a precision comparable to that of their absolute values. Many of the measurements of absolute values of nuclear parameters are made in a broad spectrum of neutron energies, and the accuracy with which the 2200-m/s values can be extracted from such measurements depends directly upon the accuracy with which the energy dependences involved are known. With these considerations in mind an effort has been made to obtain precise descriptions of the shape of the low-energy cross-sections of ²³³U, ²³⁵U, ²³⁹ Pu and ²⁴¹ Pu. Emphasis has been placed on accounting for all available data, including the information given by the measurements of the energy variation of n. The experimental data are fitted by the least-squares technique to obtain the best values of the analytical parameters and their error estimates. The analytical expressions of Fluharty et al, have been used. This formalism has the advantage of simplicity of calculation. It employs a simple polynomial of second order to describe the cross-sections in the energy region below appreciable influence of the lowest observed energy resonance. Above approximately 0.1 eV, resonance terms were added. For each indicated level separate symmetric and asymmetric resonance terms are added, corresponding to the symmetric capture term of non-fission resonances and to the asymmetry introduced by interference in the fission channels, respectively. From the analytical expressions are derived values and error estimates for the Westcott g-values.

PROMPT p IN NEUTRON-INDUCED FISSION OF ²⁴¹Pu

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Abstract

PROMPT $\overline{\nu}$ IN NEUTRON-INDUCED FISSION OF ²⁴¹Pu. The energy dependence of prompt $\overline{\nu}$ for the neutron-induced fission of ²⁴¹Pu has been investigated. The fission neutron detector was a large liquid scintillator. The data from 0.5 to 14.8 MeV of incident neutron energy can be fitted by a straight line $\overline{\nu}$ (E_n) = 2.905 + 0.146 E_n.

INTRODUCTION

Values for $\bar{\nu}$ for neutron-induced fission of ²⁴¹Pu are requested for reactor calculations with an accuracy of about 5% from thermal up to 15 MeV of incident neutron energy.

In the neutron-induced fission of ²⁴¹Pu the compound nucleus, which undergoes fission, is ²⁴²Pu. Thus the dependence of $\bar{\nu}$ on excitation energy of the compound nucleus may be studied from zero energy, corresponding to the spontaneous fission of ²⁴²Pu, up to higher energies given as the sum of the incident neutron energy and the binding energy of the last neutron in ²⁴²Pu.

This gives an opportunity to study the energy dependence of $\bar{\nu}$ over a wide range of excitation energy and to compare with the results given for the compound nuclei ²⁴⁰Pu and ²³⁸U [1, 2]. For these isotopes the $\bar{\nu}$ -values for spontaneous fission are higher than the expected ones, calculated from an extrapolation of the energy dependence of $\bar{\nu}$ obtained at higher excitation energies.

Earlier measurements of $\bar{\nu}$ for the neutron-induced fission of ²⁴¹Pu, which are made only at thermal incident neutron energy, are given in a data compilation by Asplund-Nilsson [3], together with the $\bar{\nu}$ -values of the spontaneous fission of ²⁴²Pu.

The aim of the present investigation has been to measure D for ²⁴¹Pu relative to D for the spontaneous fission of ²⁵²Cf from 0 to 15 MeV with an accuracy of about 3% in each value. This paper gives the preliminary results of the *D*-values at five different energies.

EXPERIMENTAL METHOD

The experimental arrangement, with a large liquid scintillator as the fission neutron detector is described in detail in Ref. [4].

A total amount of 20 mg of 241 Pu, deposited on three platinum plates with a coating thickness of 0.5 mg/cm², was used. The mass analysis of the deposition gave 1.33% 242 Pu, 96.95% 241 Pu, 1.18% 240 Pu and

0.53% ²³⁹Pu [5]. The amounts of ²⁴⁰Pu and ²⁴²Pu were enough to give about 4.5 spontaneous fissions per minute.

The counting rate of neutron-induced fissions in ²⁴¹Pu was limited by the maximum beam current from the accelerator and by the background counting rate in the scintillator. Thus the counting rate of ²⁴¹Pu was not larger than about 1 fission per minute.

To improve the counting ratio between the induced and spontaneous fissions the measurement was made with a time-of-flight technique according to Ref. [4]. With this technique a selection was also made of the fissions induced by the proper neutron energy in case neutrons of lower energy were produced in the target or in the collimating system. In Fig. 1 the block diagram of the electronic circuits to the time-of-flight equipment is shown together with a typical time-of-flight spectrum for the fission events.

The $\bar{\nu}$ -value of ²⁵²Cf and the ratio between the gate-length of the fission neutron counter and the background counter were checked according to Ref. [4]. The $\bar{\nu}$ -value and the number per minute of the spontaneous fission events from the contamination of ²⁴⁰Pu and ²⁴²Pu within the ²⁴¹Pu sample were measured with an accuracy of about 1%. This was made by counting all fission events occurring without a neutron beam and with the ²⁴¹Pu fission chamber in the centre of the large liquid scintillator tank.

To check the position and width of the one-channel analyser in the time-of-flight measurement the time-of-flight spectrum of the target neutrons was recorded on a 256-channel analyser with and without gating the analyser by the pulse from the one-channel analyser (see Fig. 1). The number of ²⁵²Cf fissions was counted at a certain time with and without gating the fission neutron counting system by the pulse from the one-channel analyser. In this way the percentage of spontaneous fission events occurring within the time interval corresponding to the window of the one-channel analyser could be calculated. This figure was measured with an error of less than 2%.

RESULTS AND CORRECTIONS

After the background had been subtracted from the observed $\bar{\nu}$ -values these were corrected for the contributions from the spontaneous fission of ²⁴⁰Pu and ²⁴²Pu. This correction was of the order of 10 - 15% with an estimated experimental error of about 0.5%, which was mainly due to the uncertainty in the determination of the width of the one-channel analyser. However, the statistical errors in the observed $\bar{\nu}$ -values were increased by about 10% by the correction. This depends on the statistical uncertainty in the number of spontaneous fissions and the number of neutrons from these fissions that occur within the window of the one-channel analyser.

A correction of (2 ± 0.5) % was applied to the $\bar{\nu}$ -value at 14.8 MeV for fissions induced by thermal neutrons. Other corrections were applied according to Ref. [4] but the magnitude of the sum of these corrections was less than 1%.

The corrected results, referred to $\bar{\nu} = 3.767 \pm 0.000$ for the spontaneous fission of ²⁵²Cf [6], are given in Table I and are also shown in Fig. 2. The $\bar{\nu}$ -values of the spontaneous fission of ²⁴²Pu [7, 8] and of the thermal neutron-induced fission of ²⁴¹Pu [9] are also shown in Fig. 2.



FIG.1. Block diagram of the electronic circuits of the time-of-flight equipment for the measurements of $\overline{\nu}$, and a typical time-of-flight spectrum of the fission events. The dotted lines, marked D, indicate the window of the one-channel analyser.

The straight line $p = 2.905 \pm 0.146 E_n$ is a least-squares fit to the present results. The slope of the line, 0.15 neutrons per MeV, is in agreement with that observed for other plutonium and uranium isotopes [4, 6, 10, 11].

As can be seen from Fig. 2 no certain conclusion can be drawn about a difference between the experimental $\bar{\nu}$ -value of ²⁴²Pu spontaneous and the ν -value obtained by an extrapolation of the straight line for ²⁴¹Pu + n.

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TABLE I. CORRECTED EXPERIMENTAL $\bar{\nu}$ -VALUES OF ²⁴¹Pu. The $\bar{\nu}$ -VALUES ARE BASED ON $\bar{\nu}$ = 3.767 ± 0.000 FOR THE SPONTANEOUS FISSION OF ²⁵²Cf. THE STATED UNCERTAINTIES ARE DUE TO COUNTING STATISTICS ONLY.

Neutron energy (MeV)	Ũ
0.52 ± 0.02	2. 89 ± 0. 11
2.71 ± 0.01	3.37 ± 0.11
4.19 ± 0.02	3.50 ± 0.10
5.88 \pm 0.12	3.84 ± 0.12
14.8 ± 0.2	5. 02 ± 0.14



FIG. 2. $\overline{\nu}$ -values in the fission of the compound nucleus ²⁴²Pu. The open circles are the present investigation, the filled circle is from Jaffey et al. [9], the open triangle is from Hicks et al. [7] and the filled triangle is from Crane et al. [8].

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DISCUSSION

N. STARFELT: Dr. Holmberg pointed out the equal slopes of the $\overline{\nu}$ curves of different isotopes. I should like to add that for all the isotopes of a given element there is also equality in the absolute values to within a few percent. This means that if you want to know for instance $\overline{\nu}$ for 240 Pu at a certain energy where a measurement is difficult, you can just take the measured value of 239 Pu and feel confident that you have got the right value within a few percent. The more exotic uranium isotopes could be dealt with in a similar way.

NEUTRONS PROMPTS DE FISSION-MESURE DE $\overline{\nu}$ ET DES PROBABILITES P(ν) D'EMISSION DE ν NEUTRONS

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Abstract - Résumé

PROMPT FISSION NEUTRONS - MEASUREMENT OF \vec{v} AND OF THE PROBABILITIES P(v) OF EMISSION OF v-NEUTRONS. With a large liquid scintillator in conjunction with a fission-fragment detector, we hope to measure the values of \vec{v} and P(v) for different fissile materials to better than 1% in fissions produced by neutrons in the energy range 4-14 MeV.

To estimate the various sources of error in these measurements, we have carried out preliminary work on the spontaneous fissions of 252 Cf and 240 Pu; measurement of the efficiency of the liquid scintillator; variation of $\overline{\nu}$ with the kinetic energy of the fragment detected; variation of the efficiency of the liquid scintillator with the geometry of the fission-fragment detector; correction of the experimental results and experimental check of these corrections.

We measured the P(ν) of ²⁵²Cf and the $\overline{\nu}$ and P(ν) of ²⁴⁰Pu in spontaneous fission. In absolute values, it is the inexact knowledge of the reference $\overline{\nu}$ (²⁵²Cf) which limits the accuracy of these measurements.

NEUTRONS PROMPTS DE FISSION – MESURE DE $\overline{\nu}$ ET DES PROBABILITES P (ν) D'EMISSION DE ν NEUTRONS. En utilisant la technique du gros scintillateur liquide associé à un détecteur de fragments de fission, les auteurs envisagent de mesurer à mieux que 1% $\overline{\nu}$ et P(ν) pour différents matériaux fissiles, dans les fissions provoquées par des neutrons dans le domaine d'énergie compris entre 4 et 14 MeV.

A cette fin, pour apprécier l'importance des différentes sources d'erreurs intervenant dans ces mesures, ils ont effectué différents travaux préliminaires pour les fissions spontanées de 252 Cf et de 240 Pu; mesure de l'efficacité du scintillateur liquide; variation de $\overline{\nu}$ avec l'énergie cinétique du fragment détecté; variation de l'efficacité du scintillateur liquide avec la géométrie du détecteur de fragments de fission; corrections des résultats expérimentaux, contrôle expérimental de ces corrections.

Les résultats présentés concernent la distribution des P(v) pour la fission spontanée de ²⁵²Cf et les valeurs de \tilde{v} et de P(v) pour la fission spontanée de ²⁴⁰Pu. En mesure absolue, c'est l'imprécision avec laquelle on connaît la valeur du \bar{v} de référence (²⁵²Cf) qui limite la précision des mesures.

I - PRINCIPE ET DISPOSITIF EXPERIMENTAL

Rappelons rapidement que le principe de l'expérience consiste à distribuer dans le temps les instants de capture des neutrons émis simultanément, et à détecter à l'aide de l2 photomultiplicateurs les gammas correspondants. Les neutrons thermalisés essentiellement par diffusion élastique dans le liquide scintillant sont capturés par le gadolinium dissous dans celui-ci. La Fig. 1 représente la courbe expérimentale de répartition des probabilités du délai de capture d'un neutron après son émission au centre de la sphère. L'instant de capture le plus probable est à 7 microsecondes et on constate qu'un neutron, détecté, a 99,3 chances sur 100 de l'être dans les 50 microsecondes qui suivent l'instant d'émission.

En enregistrant le nombre d'événements détectés au-dessus d'un certain seuil par les l2 photomultiplicateurs dans les 50 microsecondes qui suivent une fission, nous mesurons le nombre de neutrons détectés, au bruit de fond et aux pertes par coïncidences fortuites près.

Le bruit de fond est mesuré en enregistrant, toujours pendant 50 microsecondes les événements détectés 200 microsecondes après une fission.



FIG.1. Probabilité de capture d'un neutron en fonction du temps suivant son émission



FIG.2. Schéma général

La Fig. 2 représente le schéma de principe du dispositif expérimental utilisé :

Le scintillateur liquide sphérique, fabriqué par "Nuclear Enterprise ", type NE 5562, contient 230 litres de liquide scintillant NE 323. Au centre, dans un tube, est placé le détecteur qui enregistre l'un des 2 fragments de fission émis.

Simultanément les photomultiplicateurs détectent les gammas de fission auxquels s'ajoute la scintillation due aux premiers protons de recul. Les l2 photomultiplicateurs sont groupés en 2 bancs de 6, ces bancs après amplification sont en coïncidence de façon à réduire le taux de bruit de fond.

Les informations des 2 détecteurs sont mises en coïncidence de façon à déterminer l'instant de fission.

Si une fission est détectée, nous ouvrons pendant 50 microsecondes une porte qui laisse entrer sur une échelle rapide les informations provenant du scintillateur liquide. Le contenu de l'échelle est alors enregistré dans un bloc mémoire : si l'échelle a enregistré n événements dans la porte fission, on fait monter d'un coup le canal n . L'échelle est alors remise à zéro, et 200 microsecondes après la fission, le même cycle enregistre le bruit de fond : si l'échelle a enregistré p bruits de fond, on fait monter d'un coup le canal A + p (avec A > n_{max}).

Le stockage de l'information est interdit par 2 types de veto.

Le "veto cosmique " qui intervient si une impulsion de niveau supérieur aux plus grands gammas de capture est détectée dans les 50 microsecondes qui suivent ou précédent l'ouverture de la porte. En effet, le phénomène de post-impulsions dans les photomultiplicateurs entraîne sur la voie associée une distribution anormale d'événements, impossible à corriger.

Le "veto empilement fission " élimine le stockage de l'information d'une part si des neutrons appartenant à plusieurs fissions peuvent être enregistrés dans une seule " porte fission ", d'autre part si des neutrons de fission risquent d'être enregistrés dans la " porte bruit de fond ". L'efficacité de ce veto n'est complète que si l'efficacité du détecteur de fragments de fission et celle du scintillateur liquide pour les gammas de fission sont voisines de 100%.

II - EXPLOITATION DES RESULTATS EXPERIMENTAUX

L'expérience permet de mesurer les probabilités Q_n de détecter n événements dans les 50 microsecondes qui suivent une fission, et les probabilités En d'enregistrer n bruits de fond en 50 microsecondes. Les Q_n doivent être corrigés pour tenir compte du bruit de fond, de l'efficacité du scintillateur liquide, et éventuellement des coïncidences fortuites neutron-neutron, et neutron-bruit de fond.

1 - Correction bruit de fond

n

Si le temps de résolution de l'électronique est infiniment bref, la formule de correction est (DIVEN et al.[1]) :

$$Q_n = \sum_{j=0}^{n} Q_j^{i} B_{n-j}$$

où Q_n^i est la probabilité de détecter effectivement n neutrons par fission. Si le temps mort est petit mais non nul, et le bruit de fond important, la formule précédente entraîne une erreur par défaut, car on soustrait les cas du le bruit de fond est en coïncidence avec un neutron. Une première approximation consiste à supposer qu'il ne peut exister que k coïncidences du premier ordre (l bruit de fond et l neutron) quand n neutrons et p bruits de fond sont détectés (probabilité notée : $P_{n,p}^k$). Alors :

$$Q_n = \sum_{j=0}^{n} Q_j \sum_{p=n-j}^{p \max} B_p P_{j,p}^{p+j-n}$$

avec :

$$P_{n,p}^{k} = k! C_{n}^{k} C_{p}^{k} r^{k} P_{n-k, p-k}^{0}$$

où r = t b t : temps mort de l'électronique associée b : taux moyen de bruit de fond par unité de temps

2 - Correction temps mort

S

Si le temps mort est très faible, on peut admettre qu'il n'y a au plus qu'une coïncidence du premier ordre (l neutron - l neutron) par fission, quel que soit le nombre de neutrons émis. Les probabilités non corrigées Q'_n s'expriment en fonction des probabilités F_n corrigées par la formule donnée par DIVEN et al. [1]:

$$Q'_{n} = F_{n} (1 - s C_{n}^{2}) + F_{n+1} s C_{n+1}^{2}$$

avec :

$$= 2\tau \int_{0}^{\infty} P^{2}(t) dt$$

P(t) : probabilité qu'un neutron émis à l'instant t=0 soit détecté à l'instant t (Fig. l).

Une approximation d'ordre supérieur consiste à admettre qu'il y a k coincidences de 2 neutrons par fission à n neutrons (probabilité notée P_n^K), la formule de correction est :

$$Q_{n}' = \sum_{p=0}^{n} F_{n+p} P_{n+p}^{p}$$

$$n! s^{k} = 0$$

avec :

$$P_{n}^{k} = \frac{n! s^{k}}{k! 2^{k} (n - 2k)!} P_{n}^{0} - 2k$$

3 - Correction d'efficacité

Les probabilités physiques P(v) se déduisent des F_n par la relation :

$$P(\mathbf{v}) \approx \sum_{j=1}^{\sqrt{\max}} F_j C_j^{\mathbf{v}} \left(\frac{1}{e}\right)^{\mathbf{v}} \left(1 - \frac{1}{e}\right)^{j-\mathbf{v}}$$

avec e : efficacité de détection.

III - CONTROLE EXPERIMENTAL DES CORRECTIONS

La fission spontanée du Californium-252 est prise comme étalon. Nous avons contrôlé la validité des formules de correction utilisées en faisant varier les différents paramètres caractéristiques.

La Fig. 3 représente l'efficacité apparente du scintillateur après les différentes étapes des corrections, pour différents temps morts de l'électronique (les points expérimentaux sont numérotés de l à 8 dans l'ordre du taux de bruit de fond croissant).

Après la correction temps mort, on obtient une efficacité constante quel que soit le temps mort dans la gamme 120 à 300 nanosecondes.

La Fig. 4 représente les différents $P(\mathbf{v})$ calculés en fonction de 2 paramètres : le temps mort (tirets correspondant à la même mesure que ci-dessus) et l'efficacité (points).

60


FIG. 3. Etapes successives de l'exploitation des résultats expérimentaux obtenus pour différents temps morts (²⁵²Cf)

Nous voyons que quelles que soient les conditions expérimentales nous obtenons, à la statistique près, les mêmes P(v).

La correction coïncidences fortuites neutron-bruit de fond est superflue en fission spontanée où le taux de bruit de fond est de 0,05 par 50 microsecondes, par contre cette correction peut atteindre 1% sur les P(v) pour un taux de bruit de fond de 0,6.

IV - AUTRES SOURCES D'ERREUR SYSTEMATIQUE

1 - Electronique associée au scintillateur liquide

Une source d'erreur systématique pouvant entraîner des mesures de $\overline{\nu}$ par défaut de l à 2% est due au temps de récupération des circuits de mise en forme précédant sur chaque voie de photomultiplicateurs le circuit de coincidences. Ce temps de récupération introduit une perte d'efficacité de coincidences d'autant plus grande que le taux de bruit de fond et le temps de récupération sont élevés. Cette perte n'est pas génante lorsque toutes les mesures sont faites avec le même taux de bruit de fond. Lors des mesures en fissions provoquées du le bruit de fond est plus élevé que lors de l'étalonnage avec le Californium-252, le phénomène précédent entraîne une mesure de $\overline{\nu}$ par défaut.

Pour minimiser ce phénomène, nous avons utilisé un circuit de coincidences rapides pouvant être attaqué par les 2 voies venant du scintillateur liquide, directement après amplification. Le temps mort est réalisé après le circuit de coincidences.



FIG.4. Probabilités d'émission de ν neutrons par fission – Résultats obtenus pour différents temps morts (tirets) et pour différentes efficacités (points) (Cas du ²⁵²Cf)

2 - Fissions perdues

Une cause éventuelle d'erreur systématique est la perte de fissions due par exemple à l'épaisseur de la source. Une mesure de $\overline{\nu}$ faite en perdant des fissions de basse énergie entraîne une erreur systématique par défaut dont il convient de connaître l'ordre de grandeur.

La Fig. 5 représente l'erreur systématique commise en fonction du pourcentage de fissions perdues lors de la mesure de $\overline{\gamma}$ du 252 Cf faite en détectant le fragment de fission avec un détecteur solide.

3 - Géométrie du détecteur de fragments de fission par rapport au scintillateur liquide

Les neutrons émis dans la direction du tube central (diamètre 7,5 cm) sont détectés avec une efficacité plus faible que ceux correspondant aux fragments émis perpendiculairement à l'axe. Quand on utilise un détecteur solide présentant une géométrie légèrement inférieure à 2π par rapport à la source (cas du 252 Cf), nous avons constaté une diminution de 1% de l'efficacité quand la source est perpendiculaire à l'axe du tube, par rapport au cas où elle est parallèle à cet axe.

Pour minimiser cette erreur, il convient de travailler avec la même géométrie pour le corps étalon et pour le corps à mesurer.

4 - Mesure de l'efficacité du scintillateur liquide

L'efficacité est mesurée en utilisant les neutrons de fission du ²⁵²Cf. L'efficacité ne peut donc être connue avec une précision supérieure à celle



FIG.5. Erreur introduite sur les mesures de $\overline{\nu}$ en fonction du pourcentage des fissions perdues (Détecteur solide - Cas du ²⁵²Cf)

	Nos résultats	DIVEN	HICKS
Réf.	²⁵² cf 3,782±0,024	²⁵² cf 3,869±0,078	²⁴⁰ Pu 2,257±0,046
PO	0,0022±0,0003	0,005±0,002	0,001±0,001
Pl	0,0245±0,0012	0,004±0,009	0,021±0,007
P2	0,1225±0,0030	0,138±0,019	0,111 ±0,019
P3	0,2713±0,0024	0,223±0,032	0,271±0,019
P4	0,3053±0,0030	0,356±0,035	0,326±0,018
P5	0,1873±0,0030	0,175±0,034	0,178±0,016
P6	0,0688±0,0018	0,071±0,028	0,077±0,013
P7	0,0157±0,0007	0,022±0,017	0,013±0,004
P8	0,0024±0,0003	0,006±0,007	0,003±0,001
$D_{=}\langle \gamma^2 \rangle - \overline{\gamma}^2$	1,618		
√ D - 1/12	1,239		
$\frac{\langle v^2 \rangle_{-} \overline{v}}{\overline{v}^2}$	0,849	0,850	-
Nbre de fissions	1 700 000	4545	24 579

TABLEAU I. $P(\nu)$: CALIFORNIUM-252

BARON et al.

avec laquelle on connaît le $\overline{\nu}$ de référence. Pour exploiter les résultats présentés ici, nous avons utilisé une compilation des mesures effectuées par HOPKINS et DIVEN [2], ASPIUND, NILSON, H.CONDE et N.STARFELT [3], MOAT, MATHER et M.C.TAGGART [4], COLVIN et SOWERBY [5].

La valeur moyenne choisie, identique à celle utilisée par MOAT et al. [4],est de 3,782±0,024. Cette imprécision engendre donc une erreur systématique de 0,6% sur la mesure de l'efficacité.

Si les résultats des différentes mesures étaient compatibles avec cette valeur moyenne jusqu'en 1963, en 1965, COLVIN et SOWERBY [6] ont présenté le résultat d'une nouvelle mesure : 3,713±0,015. Cette valeur est donc inférieure de 2% à la compilation de 1963. Il semble prématuré actuellement de faire un choix entre ces différentes mesures.

V - RESULTATS

Le tableau I concerne les $P(\mathbf{v})$ du ²⁵² Cf calculés à partir des résultats expérimentaux. Pour déterminer la précision des mesures de $P(\mathbf{v})$ et de $\overline{\mathbf{v}}$, nous tenons compte de 3 types d'erreur :

- L'erreur due à l'imprécision sur la valeur de référence du ²⁵² Cf.
 L'erreur due à l'incertitude sur le temps mort et l'intégrale de la
 - probabilité de capture d'un neutron en fonction du temps.
- Les fluctuations statistiques sur le nombre d'événements enregistrés pour chaque probabilité.



FIG. 6. Probabilités d'émission de v neutrons par fission - Résultats comparés



FIG. 7. Probabilités d'émission de v neutrons par fission - Résultats comparés

La Fig. 6 permet de comparer ces résultats avec ceux de B.C.DIVEN, H. C.MARTIN, R.F.TASCHEK, J.TERREL [1], D.A.HICKS, J.ISE, Jr. et R.V.PYLE [7]. Bien que les valeurs de référence ne soient pas exactement les mêmes, il y a assez bon accord entre ces 3 types de mesures.

a assez bon accord entre ces 3 types de mesures. Le tableau II et la Fig. 7 permettent de comparer nos résultats en Pu avec ceux de DIVEN et al [1], HICKS et al [7], J.E.HAMMEL et J.F. KEPHART [8].

De nos résultats nous déduisons :

 $\overline{v}_{spon}^{240}$ Pu = 2,153±0,020

Ces mesures ont été effectuées avec une chambre à fission, c'est-à-dire avec une géométrie légèrement différente de celle utilisée lors de la mesure de l'efficacité. D'autre part, nous n'avons pas tenu compte de la différence des spectres en énergie pour les neutrons du ²⁵² Cf et ceux du ²⁴⁰ Pu.

Ce résultat est en bon accord avec la mesure de ASPLUND, NILSON, CONDE, STARFELT [3] de 1963 :

 $\overline{v}_{spon}^{240}$ Pu = 2,154 ± 0,028

	Nos résultats	DIVEN	HICKS	HAMMÉL
Réf.	²⁵² cf 3,782±0,024	²⁵² Cf 3,869±0,078	240 _{Pu} 2,257±0,046	²⁴⁰ . Pu 2,20±0,03
PO	0,0673±0,0025	0,049±0,006	0,041±0,009	0,062±0,006
Pl	0,2298±0,0046	0,214±0,012	0,219±0,021	0,198±0,017
P2	0,3288±0,0050	0,321±0,014	0,351±0,021	0,374±0,022
P3	0,2534±0,0060	0,282±0,017	0,241±0,020	0,228±0,024
Pų	0,0995±0,0030	0,112±0,013	0,127±0,018	0,114±0,022
P5	0,0192±0,0015	0,021±0,008	0,020±0,006	0,027±0,013
P6	0,0019±0,0004	0,001±0,003	0,001±0,002	0,000±0,005
$D = \langle \gamma^2 \rangle - \overline{\gamma}^2$	1,335			
√D - 1/12	1,118			
<u><v< u="">²>- v v²>- v</v<></u>	0,823	0,807		
Nbre de fissions	335 883	8 355	3 269	4 197

TABLEAU II. $P(\nu)$: PLUTONIUM-240

faite en prenant comme référence $\overline{\nu}$ (252 Cf)₌ 3,80±0,03. La plus grande partie de l'erreur affectant ces résultats est due à l'imprécision avec laquelle on connaît le $\overline{\nu}$ du 252 Cf.

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DISCUSSION

P. FIELDHOUSE: In precise $\overline{\nu}$ determinations of the kind described in Dr. Soleilhac's paper, using a large liquid scintillator as the neutron detector, one important source of error is the uncertainty in the correction one must make for the spectral difference between the reference standard, ²⁵²Cf, and the fissile nuclide under investigation. This uncertainty can be as large as $\frac{1}{2}$ % and I think this emphasizes the need for further work to be carried out on energy spectra.

КАНАЛОВЫЕ ЭФФЕКТЫ В ЭНЕРГЕТИЧЕСКОЙ ЗАВИСИМОСТИ $\overline{\nu}$ УРАНА-235 И ТОРИЯ-232

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Abstract — Аннотация

CHANNELLING EFFECTS IN THE ENERGY DEPENDENCE OF $\tilde{\nu}$ FOR ²³²U AND ²³²Th. The paper gives the results of measurements of the relationship between the mean number of secondary neutrons $\tilde{\nu}$ and the energy of the neutrons E_n inducing the fission of ²³⁵U and ²³²Th.

Measurement of $\bar{\nu}$ was performed by the method consisting of selecting pulse coincidences from $^{10}BF_{\rm g}$ counters in a paraffin block, inside which was located a multi-layer fission chamber. The method has been described in detail in numerous studies.

The neutron source was the T(p, n) reaction, obtained with a Van de Graaff accelerator.

The measurements of \vec{v} for ²³²Th were performed in the neutron energy range from 1.6 to 3.2 MeV, and those of \vec{v} for ²³⁵U in the range from 0.4 to 3.2 MeV; the energy step was ~0.2 MeV and the energy resolution ~ 0.06 MeV. The accuracy of the \vec{v} -values was about 1% for ²³⁵U and about 2% for ²³²Th.

The measured values for $\overline{\nu}$ were compared with those calculated from the mean kinetic energy of the fission fragments in the same energy range.

In general, the measured dependence of $\vec{\nu}$ on the energy of the neutrons E_n inducing ²⁸⁵U fission takes the form of a somewhat stepped curve. In particular, in the energy region from 1 to 2 MeV the slope $d\nu/dE_n$ represents 0.06 MeV⁻¹, whereas at higher neutron energies $d\vec{\nu}/dE_n \sim 0.15$ MeV⁻¹.

One of the possible explanations of the stepped dependence of $\overline{\nu}$ on the energy of neutrons inducing fission of the even-even compound nucleus ²³⁶U could be the existence of an energy gap in the ²³⁶U fission channel spectrum. The stepped form of the energy dependence of $\overline{\nu}$ is in good qualitative agreement with the conclusions of V.M. Strutinsky and V.A. Pavlinchuk regarding the effect of nucleon pairing on the internal excitation spectrum of fissionable nuclei.

КАНАЛОВЫЕ ЭФФЕКТЫ В ЭНЕРГЕТИЧЕСКОЙ ЗАВИСИМОСТИ $\overline{\nu}$ УРАНА-235 И ТОРИЯ-232. В работе приводятся результаты измерений зависимости среднего числа вторичных нейтронов $\overline{\nu}$ от энергии нейтронов E_n , вызывающих деление урана-235 и тория-232.

Измерения $\overline{\nu}$ проводились методом отбора совпадений импульсом от B¹⁰ F₃ счетчиков в парафиновом блоке и помещенной внутрь него многослойной камеры деления. Методика подробно описана во многих работах.

Источником нейтронов была реакция T(p, n) на ускорителе Ван-де-Граафа.

Измерения $\overline{\nu}$ тория-232 были выполнены в интервале энергий нейтронов от 1,6 Мэв до 3,2 Мэв, измерения $\overline{\nu} U^{235}$ в интервале энергий от 0,4 Мэв до 3,2 Мэв с шагом по энергии ~0,2 Мзв и энергетическим разрешением ~0,06 Мэв. Точность эначений $\overline{\nu}$ около 1% для U^{235} и около 2% для Th 232 .

Проведено сравнение измеренного v со значениями, вычисленными из величины средней кинетической энергии осколков в этой же энергетической области.

В целом измеренная зависимость $\overline{\nu}$ от энергии нейтронов E_n , вызывающих деление урана-235, представляет собой некую ступенчатую кривую. В частности, на участке энергий от 1 Мэв до 2 Мэв наклон $d\overline{\nu}/dE_n$ составляет 0,06 Мэв⁻¹, в то время, как при более высоких энергиях нейтронов $d\overline{\nu}/dE_n \sim 0,15$ Мэв⁻¹.

Одним из возможных объяснений ступенчатой зависимости $\overline{\nu}$ от энергий нейтронов, вызывающих деление четно-четного компаунда ядра урана-236 могло бы быть наличие энергетической щели в спектре каналов деления урана-236. Ступенчатая структура энергетической зависимости $\overline{\nu}$ качественно хорошо согласуется с заключениями работы Струтинского В.М. и Павлинчука В.А. о влиянии спаривания нуклонов на спектр внутреннего возбуждения делящихся ядер. Измерения среднего числа мгновенных нейтронов $\overline{\nu}$, выполненные в последние годы [1 ÷ 5], показали приближенность принимавшихся прежде представлений о линейной зависимости $\overline{\nu}$ от энергии нейтронов E_n , вызывающих деление [6, 7]:

$$\overline{\nu}(\mathbf{E}_{n}) = \nu_{\tau} + \mathbf{a}\mathbf{E}_{n}, \tag{1}$$

где а≈0,13-0,15 Мэв⁻¹ - константа,приблиэительно равная обратной средней энергии, требуемой для отделения нейтрона от осколка.

Исследования энергетической зависимости средней кинетической энергии осколков Е_к от Е_п [3, 8] также обнаруживают вполне заметные отступления от гипотезы Фаулера о независимости Е_к от Е_п, положенной в основу соотношения (1).

Зависимость (1) предполагает, таким образом, что вся избыточная над барьером энергия возбуждения полностью переходит в нуклонные степени свободы образующихся осколков. Андреев [9] впервые обратил внимание на то, что существование в процессе деления стадии охлажденного переходного ядра может вызвать ряд эффектов, которые приведут к отступлениям от "универсального" линейного роста. Происхождение их связывается с дискретностью спектра уровней ядра в переходном состоянии (каналов деления). Возбуждение каналов деления, имеющих коллективную природу, в случае слабой связи между коллективными и нуклонными степенями свободы при спуске с вершины барьера, может привести не только к уменьшению $d\nu/dE_n$ [1, 2, 10], но и к нерегулярному изменению $\overline{\nu}$ [3]. Нерегулярности в ходе зависимости $\overline{\nu}$ (E_n) могут возникнуть также в результате изменения от канала к каналу конфигурации делящегося ядра ("точки" разрыва) [3]. С точки зрения наблюдения подобных эффектов, область энергий вблизи порога деления представляет наибольший интерес. Наиболее благоприятным объектом исследования, по-видимому, являются четно-четные делящиеся ядра, обладающие энергетической щелью в спектре внутренних возбуждений.

Из ядер этого класса наиболее подробно изучен уран-236 в реакции уран-235 (n, f). Мы предприняли изучение ядра мишени урана-235 не только из-за его большой роли в качестве ядерного горючего в реакторах, но и в связи с тем, что из всех изученных четно-четных делящихся ядер урана-236 обладает наименьшим по абсолютной величине значением пороговой энергии нейтронов $E_{n,f} = -0,6$ Мэв [11]. Последнее обстоятельство позволило рассчитывать на то, что у этого ядра область E_n , в которой можно было ожидать проявления каналовых эффектов, будет наиболее протяженной. В данной работе сообщаются результаты измерений $\overline{\nu}$ урана-235 в области E_n от 0,4 до 3,2 Мэв.

Кроме того, мы приводим некоторые данные для нечетного ядра тория-233, делящегося в реакции торий-232 (n, f). Эти измерения будут продолжены.

YPAH-235

Измерения $\overline{\nu}$ производились широкораспространенным методом: путем отбора совпадений между BF₃ - счетчиками в парафиновом блоке и помещенной внутрь него многослойной камерой делений. Схема опыта приведена на рис.1. Эффективность детектора к нейтронам деления составляла ~5%. Низкая эффективность детектора нейтронов была



Рис. 1. Схема опыта

скомпенсирована использованием больших количеств делящегося вещества: в камеру деления было загружено около 2 г урана-235 90%-ного обогащения. Непосредственно в опыте измерялось отношение

$$\mathbf{R} = \frac{(\mathbf{N}/\mathbf{N}_{f})\mathbf{E}_{n}}{(\mathbf{N}/\mathbf{N}_{f})\mathbf{E}_{n}} = \mathbf{A}\frac{\overline{\nu}(\mathbf{E}_{n})}{\overline{\nu}(\mathbf{E}_{n})},$$

где $\tilde{\mathbf{E}}_n = 0,39$ Мэв — опорное значение энергий нейтронов, для которого $\overline{\nu}$ было определено в работе [3] и составляет (1,025±0,007) $\overline{\nu}_{\tau}$ (уран-235); N и N_f — число делений в камере и истинных совпадений соответственно; A — поправочный множитель, близкий к единице, учитывает зависимость эффективности от энергии падающих нейтронов.

В табл.1 приведены значения R/A, и среднее число мгновенных нейтронов $\overline{\nu}$. При вычислении A мы учитывали угловую корреляцию нейтрон деления — падающий нейтрон, возникающую вследствие угловой

ТАБЛИЦА 1. 7 УРАНА-235

D + 276 superedid & D + 522

Энергия нейтронов Е _п	R	$\overline{ u}$
0,37±0,10	1,000 ±0,007	2,474 ±0,017
$0,59 \pm 0,10$	0,998±0,014	$2,469 \pm 0,035$
0,81 ±0,09	$0,993\pm0,014$	$2,457 \pm 0,035$
$1\text{,}02\pm0\text{,}08$	$1,024 \pm 0,011$	$2,534 \pm 0,027$
$1,23 \pm 0,08$	1,031±0,015	$2,551 \pm 0,037$
$1,44 \pm 0,07$	$1,\!033\pm0,\!015$	$2,555 \pm 0,037$
$1,64 \pm 0,07$	$1\text{,}044\pm0\text{,}014$	$2,583 \pm 0,034$
$1,85 \pm 0,07$	$1,055 \pm 0,013$	$2,610 \pm 0,032$
$2,05 \pm 0,06$	$1,050 \pm 0,012$	$2,598 \pm 0,029$
$2,25 \pm 0,06$	$1,077 \pm 0,014$	$2,665 \pm 0,035$
$2,46\pm0,06$	1,108±0,015	$2,741 \pm 0,038$
$2,76 \pm 0,06$	1,130±0,014	2,795±0,034
3,06±0,05	1,133±0,016	2,803±0,046
$3,25 \pm 0,05$	1,144 ±0,017	$2,830 \pm 0,042$



Рис. 2. а) Схематическое изображение энергетической зависимости $\overline{\kappa}^2$ от энергии возбуждения Е* переходного ядра в реакции плутония-239 (d, pf) [16]. б) Сравнение результатов данной работы (ϕ) с измерениями $\overline{\nu}$ урана-235 других авторов: $\phi = [12]$, $\phi = [13], \phi = [10, 21], \phi = [4], \phi = [2], \psi = [22].$

анизотропии деления, изменение спектра нейтронов деления с ростом $\overline{\nu}$, вклад делений урана-235 медленными закадмиевыми нейтронами.

На рис.2б результаты настоящих измерений сравниваются с данными других работ [2, 4, 5, 12 - 14], точность которых не хуже 3%. Полученные нами значения 7 согласуются с другими данными, за исключением, быть может, точки при E_n = 2,0 Мэв, на которую в связи с этим было обращено особое внимание.

Совокупность приведенных данных показывает, что реальное поведение $\overline{\nu}(E_n)$ существенно отличается от линейной зависимости (1) с постоянным значением $d\overline{\nu}/dE_n$. Характер отступлений от соотношения (1) в работах [2, 4] представлен различным образом. Гопкинс и Дайвен [2] считают, что зависимость $\overline{\nu}(E_n)$ можно приблизительно описать ломаной линией с изломом при $E_n = 2$ Мэв, для которой $d\overline{\nu}/dE_n =$ 0,085 Мэв⁻¹ при $E_n < 2$ Мэв $d\overline{\nu}/dE_n = 0,16$ Мэв⁻¹ при $E_n > 2$ Мэв. В работе Мазера, Филдхауза и Моата [4] рассматриваются два способа описания экспериментальных данных: с помощью двух отрезков ломаной линии с $d\overline{\nu}/dE_n = 0,109$ Мэв⁻¹ при $E_n < 3$ Мэв и 0,181 Мэв⁻¹ при $E_n > 3$ Мэв и квадратичной параболы с $d^2\nu/dE_n^2 > 0$. Оба эти представления совершенно не передают тонких деталей зависимости $\overline{\nu}(E_n)$ при низких E_n и отражают лишь самый грубый, но весьма существенный эффект: возрастание фактора $d\nu/dE_n$ с увеличением E_n .

В настоящее время есть основания говорить о наличии более сложной структуры в ходе $\overline{\nu}$ (E_n). Совокупность данных вполне отчетливо обнаруживает два подъема в районе 0,2 и 1 Мэв. На участке 0,2 – 1 Мэв

 $\overline{
u}$, по крайней мере, не возрастает. После "скачка" при 1 Мэв $\overline{
u}$ медленно растет с наклоном, который характеризуется dv/dE,≈0.1 Мэв⁻¹ начиная с Е_n = 2,5 - 3 Мэв устанавливается более крутой рост с d_ν/dE_n ≈ 0,16 Мэв⁻¹. Производная $d\overline{\nu}/dE_n$ вновь увеличивается со вступлением процесса (n, nf) при E_n = 5,5 - 6 Мэв. Данная структура зависимости $\overline{\nu}$ от E_n у урана-235 обсуждалась нами в предварительной публикации [15], где ее происхождение было связано с предположением об аномально большей величине энергетической щели 22 = 2,5 Мэв для ядра в переходном состоянии, которое было выдвинуто авторами работ [16, 17] для объяснения поведения угловой анизотропии осколков в реакции плутония-239 (d, pf). Угловая анизотропия деления согласно статистической теории [18] однозначно связана со средним квадратом проекции углового момента на ось ядра \overline{K}^2 . Энергетическая зависимость \overline{K}^2 от энергии возбуждения Е* переходного ядра урана-236 схематически показана на рис.2а, на котором шкалы E* и E_n совмещены так, чтобы выполнялась очевидная связь $E_n - E^* = -0,6$ Мэв. Обращает на себя внимание поразительное сходство между зависимостями на рис.2а и 2б. Сходство проявляется даже у энергий, при которых происходят скачки $\overline{\nu}$ и \overline{K}^2 .

Ступенчатый характер зависимостей $\overline{\nu}$ (E_n) и \overline{K}^2 (E*) при E*<2,5 Мэв и монотонный рост при E*> 2,5 Мэв, казалось бы, получает естественное объяснение в предположении, что, начиная с E* = 2,5 Мэв (E_n \approx 2 Мэв), вступают каналы внутреннего возбуждения.

Впоследствие Струтинский и Павлинчук [19] дали единообразное истолкование поведения $\overline{\nu}$ и K² и корреляции между ними без привлечения предположения об аномально большей энергетической щели. Эти эффекты объяснены ими скачкообразным изменением числа возбужденных квазичастиц n с ростом E* в интервале нескольких Δ : n=0, если E*<2 Δ и n=2, когда 2 Δ <E*<(4 \div 5) Δ . При больших E*n(E*) растет практически монотонно. Задолго до обнаружения обсуждаемых эффектов Струтинский [20] показал, что $\overline{K}^2 \sim n$. Если поставить в соответствие с положением второго подъема \overline{K}^2 величину 2 Δ = 1,4 Мэв для равновесных ядер, то положение третьего подъема при (4 \div 5) Δ ~3 Мэв окажется в удовлетворительном согласии с опытом. Скачок \overline{K}^2 внутри щели связывается со вступлением каналов γ -вибрационной природы с K = 2⁺.

Расчет $\overline{\nu}$ в [19] производился в предположении, что:

а) избыток над энергией состояний внутреннего возбуждения E*- U переходит в кинетическую энергию осколков;

б) коллективные состояния внутри щели вклада в $\overline{\nu}$ не дают.

Тогда, как показано в работе [19], в интервале $E^* < 2\Delta (d\overline{U})/(dE^*) = 0$, $2\Delta < E^* < (4 \div 5) \Delta (d\overline{U})/(dE) = 2/3$, при больших $E^* (d\overline{U})/(dE) \simeq 1$. Авторы [19] считают, что переходная область E^* , в которой $(d\overline{U})/(dE)$ изменяется от 2/3 до 1, невелика. В этом случае отношение $(d\overline{\nu})/(dE_n) =$ $(d\overline{\nu})/(d\overline{U}) \cdot (d\overline{U})/(dE)$ для последних двух участков составляет 2/3 в превосходном соответствии с результатами опыта. "Скачок" $\overline{\nu}$ при $E_n \leqslant 0,2$ Мэв связывается с состоянием с K = 2⁺, через которые преимущественно и происходит деление урана-235 (7/2) р-нейтронами. Таким образом, ступенчатый рост $\overline{\nu}$ находит не только качественное, но и количественное объяснение в рамках последовательного рассмотрения эффектов спарирования нуклонов.

En	ν
1,48±0,03	2,175±0,096
1,56±0,05	2,088±0,073
1,64 ±0,07	2,103 ± 0,069
2,05 ±0,06	$2,158 \pm 0,065$
2,46±0,06	2,231 ±0,048
2,86±0,05	2,231 ±0,050
$3,27 \pm 0,04$	$2,425 \pm 0,070$

ТАБЛИЦА 2. 🗄	⊽ ТОРИЯ-232
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торий-232

В опыте использовалось около 4 г тория-232. Измерения производились по отношению к $\overline{\nu}$ урана-235 при $E_n = 1$ Мэв. Экспериментальные данные приводятся в табл.2 и на рис.3, где они сравниваются с результатами других работ. Результаты настоящих измерений в согласии со всей имевшейся в нашем распоряжении экспериментальной информации и обнаруживают значительный подъем $\overline{\nu}$ на пороге деления тория-232. Этот типично каналовый эффект, по-видимому, связан с делением через разные состояния переходного ядра одночастичного происхождения [3]. Аналогичное явление было обнаружено и в ходе средней кинетической энергии осколков урана-235 в реакции урана-234 (d, pf) [8].

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Рис. 3. Сравнение результатов измерения $\overline{\nu}$ тория-232 настоящих измерений ϕ с данными других авторов: $\phi = [24], \ \phi = [23], \ \phi = [25]$. Все данные нормированы к ν_p (калифорний-252) = 3,763; $\nu_p^{\rm T}$ (уран-235) = 2,414.

CN-23/95

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ТОНКАЯ СТРУКТУРА ЭНЕРГЕТИЧЕСКОЙ ЗАВИСИМОСТИ 7 УРАНА-233 И УРАНА-235 ПРИ ДЕЛЕНИИ НЕЙТРОНАМИ НИЖЕ 1 Мэв

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Abstract

FINE STRUCTURE OF THE ENERGY DEPENDENCE OF $\vec{\nu}$ FOR ²³³U AND ²⁸⁵U IN FISSION INDUCED BY NEUTRONS OF LESS THAN 1 MeV. The authors give the results of comparative measurements of the average number of secondary neutrons for ²⁹³U and ²³⁵U in the energy range of fission-inducing neutrons up to 1 MeV.

The results of the comparative measurements were converted to absolute values on the basis of the relationship:

$$\overline{\nu}(E_n) = \overline{\nu}_{th} + a(E_n - \Delta \overline{E}_n)$$

where $\overline{\nu}(E_n)$ and $\overline{\nu}_{th}$ are respectively the mean number of secondary neutrons for any primary-neutron energy E_n and for thermal energy, a is a parameter inverse to the mean energy of neutron emission and $\Delta \overline{E}_n$ is the difference between the mean kinetic energy of fragments in the case of fission induced by neutrons of energy E_n and by thermal neutrons.

Use of this relationship is based on the fact that, within the limits of the neutron energy range studied, a change in the mass yield curve and the energy removed by neutrons and fission gamma rays have no pronounced effect on the redistribution of energy between the collective and internal degrees of freedom of the fissioning nucleus. The feasibility of this indirect method of arriving at absolute figures was corroborated by deriving the absolute values for ²³⁵ U directly. The authors present possible interpretations of the results obtained,

ТОНКАЯ СТРУКТУРА ЭНЕРГЕТИЧЕСКОЙ ЗАВИСИМОСТИ $\overline{\nu}$ УРАНА-23⁵ И УРАНА-235 ПРИ ДЕЛЕНИИ НЕЙТРОНАМИ НИЖЕ 1 Мэв. Приводятся результаты относительных измерений среднего числа вторичных нейтронов для урана-233 и урана-235 в диапазоне энергий нейтронов, вызывающих деление до 1 Мэв.

Абсолютизация результатов относительных измерений осуществлялась на основе соотношения

$$\overline{\nu}(\mathbf{E}_{n}) = \overline{\nu} \mathbf{t} \mathbf{h} + \mathbf{a} (\mathbf{E}_{n} - \Delta \overline{\mathbf{E}}_{n}),$$

где $\overline{\nu}$ (E_n) и $\overline{\nu}$ th соответственно среднее число вторичных нейтронов при некоторой энергии первичных нейтронов E_n и тепловой, а – параметр, обратный средней энергии отделения нейтрона, Δ E_n – разность средней книетической энергии осколков при делении нейтронами с энергией E_n и тепловыми.

Использование приведенного соотношения основано на том, что в пределах исследованного диапазона энергий нейтронов изменение кривой выхода масс, энергия, уносимая нейтронами и у-лучами деления, не вляяют существенно на перераспределение энергии между коллективными и внутренними степенями свободы делящегося ядра. Прямая абсолютизация данных для урана-235 подтвердила возможность такого непрямого метода абсолютизация. Приводятся возможные интерпретации полученных результатов.

введение

В работе [1] были сообщены результаты измерений среднего числа вторичных нейтронов $\overline{
u}$ для урана-235 и средней кинетической энергии осколков Ек урана-233 и урана-235 при делении указанных ядер-мишеней нейтронами. Совокупные данные по $\overline{\nu}$ и Е_к для урана-235 указывали на наличие отступлений от линейной зависимости $\overline{\nu}$ от энергии E_n нейтронов. вызывающих деление, и E_к(E_n) от постоянства, принимавшихся в соответствии с [2]. В работе [1] была приведена также интерпретация обнаруженных эффектов, которая связывала нерегулярности в $\overline{\nu}$ и \overline{E}_{κ} с проявлением дискретной структуры каналов в переходном состоянии делящегося ядра. В основу анализа экспериментальных данных по $\overline{\nu}$ и \overline{E}_{κ} была положена модель неоксиально-симметричного грушевидного ядра, совершающего туннельные переходы между зеркальносимметричными формами. В соответствии с этой моделью в переходном состоянии ядро имело две полосы уровней положительной 0 *, 2 *, 3 *, 4 * . . . и отрицательной 1, 2, 3, 4, ... четности, разделенные энергетическим интервалом $E_{inv} = t \omega_{inv} \approx 0.8$ Мэв, где ω_{inv} — частота туннельного перехода. При делении урана-235 (7/2) s-нейтронами в седловой точке реализовывались состояния 37, 47. При вступлении в игру р-нейтронов деление осуществлялось через состояния $2^{+}-5^{+}$, лежащие на $\sim 0,8$ Мэв ниже. Далее предполагалось, что:

1) энергия возбуждения каналов отрицательной четности E_{inv} переходит в кинетическую энергию осколков вследствие слабой связи инверсии ядра с внутренними степенями свободы;

 кинетическая энергия деформации, поглощающая избыток энергии переходного ядра над энергией возбуждения участвующих в делении каналов диссипируется в нуклонные степени свободы;

3) энергия возбуждения каналов нуклонной природы идет на нагрев осколков;

4) деление через каналы нуклонной природы благоприятствует изменению среднего расстояния, на котором происходит разрыв стенки ядра \overline{r}_{sc} , что сопровождается соответствующими изменениями \overline{E}_{sc} .

Падение $\overline{\mathbf{E}}_{\kappa}$ и соответственно рост $\overline{\nu}$ при переходе от s- и ρ -нейтронам в рамках этих представлений объясняется как переход от каналов З⁻, 4⁻, при давлении через которые $\overline{\mathbf{E}}_{\overline{\kappa}}$ на величину \mathbf{E}_{inv} больше, к каналам 2⁺ - 5⁺.

Для урана-233 (5/2⁺) с противоположной четностью основного состояния общая тенденция в $\overline{E}_{\vec{k}}$ (E_n) и $\overline{\nu}$ (E_n) должна быть обратной только что рассмотренной. Деление s-нейтронами урана-233 идет через состояние 2⁺, 3⁺ нижней полосы, а ρ -нейтронами – через состояние 1⁻ - 4⁻ полосы отрицательной четности приподнятой на $E_{inv} \sim 0,8$ Мэв. Поэтому переход от s- к ρ -нейтронам должен сопровождаться увеличением \overline{E}_{κ} и соответствующим падением $\overline{\nu}$. Экспериментальные данные по $\overline{E}_{\kappa}(E_n)$ подтвердили этот вывод. Желательно было проведение измерений и $\overline{\nu}(E_n)$ для урана-233 в этой области энергий E_n . Наряду с измерениями $\overline{\nu}(E_n)$ для урана-233 было признано целесообразным проведение повторных, более детальных измерений $\overline{\nu}(E_n)$ и для урана-235. В основу экспериментальной методики была положена описанная в [1] методика относительных измерений, которая, как оказалось, позволяет при сравнительно несложной аппаратуре производительные измерения $\overline{\nu}$ с точностью лучшей 1%. (С точки зрения выявления структуры в $\overline{\nu}(E_n)$ эта методика пол-



Рис. 1. Расположение аппаратуры при относительных измерениях: 1 - мониторная камера; 2 - диск из делящегося изотопа; 3 - мониторные слои; 4 - камера деления; 5 - слои тория-232; 6 - мишень ускорителя.

ностью решала поставленную задачу. Для возможности практического использования данные относительных измерений необходимо абсолютизировать либо прямым способом [1], либо непрямым — с помощью привлечения данных по кинетической энергии осколков деления, как это сделано в настоящей работе.

ОТНОСИТЕЛЬНЫЕ ИЗМЕРЕНИЯ $\overline{\nu}$

В пучок нейтронов, получаемых в реакции Н³(р, n) Не³ с помощью электростатического ускорителя, помещается мониторная ионизационная камера (1) (рис.1), содержащая диск (2) из исследуемого изотопа диаметром 30 мм и толщиной 3 мм, окруженного тонкими $\sim 1 \text{ мг/см}^2$ слоями (3) одинакового с диском изотопного состава, которые нанесены на подложки диаметром также 30 мм. Количество делящегося вещества в слоях подбиралось одинаковым с точностью ±10%. Вторичные нейтроны, вылетающие, в основном, из диска, регистрируются пороговым детектором вторичных нейтронов с величиной порога В, превышающей максимальную энергию первичных нейтронов из исследуемого диапазона. В данном случае в качестве порогового детектора вторичных нейтронов использовалась ионизационная камера деления (4) со слоями тория-232 (5). Число отсчетов, зарегистрированное за некоторое время детектором вторичных нейтронов Nn, отнесенное к числу импульсов в мониторной камере nf с точностью до некоторых весьма слабо зависящих от E_n факторов, равно $\overline{\nu}(\mathbf{E}_n)$:

$$\rho = \frac{N_n(E_n)}{n_f(E_n)} = c^{\dagger} \overline{\nu}(E_n).$$

Отношение двух значений ρ, измеренных при энергии первичных нейтронов E_n, меняющейся от опыта к опыту, и некоторой \widetilde{E}_n , постоянной для всех измерений, с точностью до коэффициента с равно отношению соот-

КУЗНЕЦОВ и СМИРЕНКИН

ветствующих \mathbf{E}_n и $\widetilde{\mathbf{E}}_n$ значениям $\overline{\boldsymbol{\nu}}$:

$$\mathbf{R}_{\mathfrak{skcn}} = \frac{\rho(\mathbf{E}_n)}{\rho(\mathbf{\widetilde{E}}_n)} = \mathbf{c}(\mathbf{E}_n, \mathbf{\widetilde{E}}_n) \frac{\overline{\nu}(\mathbf{E}_n)}{\overline{\nu}(\mathbf{\widetilde{E}}_n)} = \mathbf{c}(\mathbf{E}_n, \mathbf{\widetilde{E}}_n) \mathbf{R}(\mathbf{E}_n, \mathbf{\widetilde{E}}_n) \dots$$
(1)

Расчет коэффициента с позволяет найти искомую величину R. Выбор толщины диска определялся оптимумом, складывающимся из соображений достаточно высокой статистики отсчетов и малости поправочного коэффициента с в (1). Измерения производились с использованием твердой $Zr - {}^{3}H$ или Ti - {}^{3}H мишени толщиной ~ 0,3 мг/см². Нейтроны с энергией 0.2 Мэв получались установкой устройства (рис.1) под углом 60° к пучку протонов. В этом случае на диск попадали нейтроны с E_n от 0,12 Мэв до 0,28 Мэв и $\overline{E}_n = 0,2$ Мэв; нейтроны с $E_n = 0,08$ Мэв получались под 0° к пучку протонов превышением энергии протонов над порогом реакции ³ H(p, n)³ He на 25 кэв. При этом на диск попадали нейтроны с E_n от 0,02 до 0,14 Мэв. Для всех остальных точек разница между максимальной и минимальной энергией нейтронов не превышала 0,06 Мэв. Апертура диска в пучке нейтронов составляла 40°. Измерения при энергии нейтронов Е, чередовались с измерениями при опорной энергии Е,. Величина River, получаемая в одной такой серии, имела точность по статистическому фактору ~3-4%. Число серий измерений подбиралось таким, чтобы получить итоговую ошибку 0,5-0,7% по разбросам отдельных серий. Аппаратура обеспечивала набор импульсов детектора вторичных нейтронов на уровне ~ 5 н/сек. Значения $R_{3\kappa cn}$, полученные в измерениях, приведены в таблицах 1 и 2. В табл.2 внесены результаты, полученные в [1] со сцинтилляционным пороговым детектором вторичных нейтронов. Поскольку точность, достигнутая в [1] с ториевой камерой невысока и составляет 1,5 - 2%, в качестве окончательных результатов с этим детектором вторичных нейтронов рассмариваются данные публикуемой работы. При расчете коэффициента с в (1) учитывались следующие факторы, приводящие к отклонению с от единицы:

1. Зависимость от E_n коэффициента пропорциональности λ между полным числом делений в диске F и числом делений в поверхностных слоях n_f .

Зависимость эффективности регистрации вторичных нейтронов
 От пространственной корреляции первичный нейтрон-осколок деления – вторичный нейтрон, воэникающий в результате наличия угловой анизотропии разлета осколков деления относительно направления первичных нейтронов и сильной угловой зависимости вероятности испускания вторичных нейтронов относительно направления разлета осколков в лабораторной системе координат.

 Зависимость эффективности регистрации вторичных нейтронов пороговым детектором <Σ> от изменения средней энергии нейтронов деления, связанного с изменением энергии возбуждения делящихся ядер.

4. Зависимость эффективности регистрации вторичных нейтронов <µ> от пространственных характеристик распределения делений по диску. Для сцинтилляционного детектора эта зависимость пренебрежимо мала из-за большого расстояния от детектора до диска.

5. Вклад процессов, связанных с упругим, неупругим взаимодействием первичных нейтронов с веществом диска и мультипликацией нейтронов в среде диска. Этот вклад учитывается коэффициентом X. Расчет указанных поправочных коэффициентов производился только с учетом вклада нейтронов первого поколения, т.е. нейтронов, возникших в результате однократного соударения первичных нейтронов с ядрами диска. Такое приближение оказалось достаточным, поскольку геометрические характеристики диска выбирались такими, чтобы параметр, характеризующий вклад многократных соударений $\Sigma_t \cdot t$ (Σ_t – макроскопическое сечение взаимодействия нейтронов с ядрами, а t – наиболее характерный геометрический размер, в данном случае толщина диска), оказался малым:

$$\Sigma_t \cdot t \sim 0,1$$

Структура уравнения (1) такова, что все поправочные коэффициенты входят в него в виде отношения их значений при энергии первичных нейтронов E_n к значениям при энергии \tilde{E}_n . Это весьма важное и благоприятное для данной методики обстоятельство, поскольку указанные коэффициенты от E_n зависят весьма слабо, а их отношение незначительно отличается от единицы. В таблицах 1 и 2 приведены значения отношения поправочных коэффициентов для энергий нейтронов E_n , \tilde{E}_n ; $\lambda/\overline{\lambda}$, $<\Omega>/<\widetilde{\Omega}>$, μ/μ , $\chi/\tilde{\chi}$. Поправка $<\Sigma>/<\tilde{\Sigma}>$ в явном виде не представлена. Она учитывалась непосредственно при переходе от $R_{\rm эксn}$ и

$$\Xi = \frac{\lambda}{\lambda} \cdot \frac{\langle \Omega \rangle}{\langle \overline{\Omega} \rangle} \cdot \frac{\chi}{\overline{\chi}} \cdot \frac{\mu}{\overline{\mu}} \, \mathrm{k} \, \mathrm{R} \, .$$

В последней колонке таблиц 1 и 2 приведены окончательные значения R. В ошибку R включена ошибка, связанная с неточностью введения поправок. Поправки не превышают 0,4% от измеряемой величины, т.е. меньше среднеквадратичной ошибки результатов измерений. Поскольку поправки изменяют R_{эксп} в противоположных направлениях, окончательное значение слабо отличается от R_{эксп}, т.е. измеряемая в эксперименте величина устойчива к действию различных искажающих эффектов.

АБСОЛЮТИЗАЦИЯ РЕЗУЛЬТАТОВ ОТНОСИТЕЛЬНЫХ ИЗМЕРЕНИЙ

Абсолютизация результатов относительных измерений состоит в сравнении какого-либо значения $\overline{\nu}$ из исследованного диапазона E_n (например, при \widetilde{E}_n) с хорошо известным значением $\overline{\nu}$. Чаще всего в качестве последнего используют $\overline{\nu}$ при делении тепловыми нейтронами ($\overline{\nu}_{T}$). В работе [1] в отдельном эксперименте было проведено сравнение $\overline{\nu}$ (\widetilde{E}_n) с $\overline{\nu}_{T}$, оказавшееся равным

$$\frac{\overline{\nu}_{\mathrm{T}}}{\overline{\nu}(\widehat{\mathbf{E}}_{\mathrm{n}})} = 1,025 \pm 0,007.$$

Принятая в [1] методика не позволяла привести такое сравнение с $\overline{\nu}_{\rm T}$ для урана-233 ввиду того, что из-за повышенной у-активности этих изотопов в ионизационной камере невозможно было получить необходимую статистическую точность из-за недостаточного количества вещества. Поэтому в настоящей работе было решено обратиться к непрямому методу абсолютизации путем привлечения весьма подробных и точных данных



Рис. 2. Сравнение результатов измерения $\overline{\nu}$ (E_n) для урана-233: ● - результаты данной работы; ▼ - значения $\overline{\nu}$ (E_n), полученные по $\Delta \overline{E}_k(E_n)$ [1] с помощью уравнения баланса [2]; \diamond - данные Колвина и Соуэрби [3]; О - данные Дивена и Гопкинса [4]; Δ - данные Мазера и др. [5].

по разности средних кинетических энергий осколков при делении нейтронами с энергией Е_п и тепловыми нейтронами

$$\Delta \overline{E}_{\kappa} = \overline{E}_{\kappa} (E_{n}) - \overline{E}_{\kappa}^{T}$$
⁽¹⁾

В предположении неизменности кривой выхода масс и зарядов Y (M, z) и постоянства средней энергии \overline{E}_{γ} мгновенных ү-лучей в диапазоне $E_n \le 1$ Мэв из разности уравнений баланса энергии, реализующейся при делении быстрыми и тепловыми нейтронами, можно получить следующее соотношение:

$$\overline{\nu}(\mathbf{E}_{n}) = \overline{\nu}_{T} + \mathbf{a}[\mathbf{E}_{n} - \Delta \overline{\mathbf{E}}_{K}(\mathbf{E}_{n})],$$
 (2)

где а — величина, обратная энергии отделения нейтрона, равной сумме средней энергии связи нейтрона в осколках и средней кинетической энергии вторичных нейтронов. Согласно имеющимся оценкам значение а находится в пределах 0,13 - 0,15 Мэв⁻¹. Если энергетический эквивалент, деформация поверхности Y (M, z) и Е_γ при переходе от деления тепловыми нейтронами к быстрым с энергией Е_n обозначить через ΔMc^2 , то разность уравнений энергетического баланса при делении нейтронами с энергией Е_n и тепловыми запишется в виде:

$$\Delta M c^{2} = - \left[E_{n} - \Delta \overline{E}_{\kappa}(E_{n}) \right] + \frac{\overline{\nu} \left(E_{n} - \overline{\nu}_{T} \right)}{a} \dots$$
(3)

Видно, что при $\Delta Mc^2 = 0$ (т.е., если Y (M, z) = const и \overline{E}_{γ} =const) соотношение (3) переходит в (2). Примем в качестве первого приближения весьма реальное предположение Y (M, z) = const и \overline{E}_{γ} = const и разделим правую и левую части (2) на $\overline{\nu} \widetilde{E}_n$):

$$\frac{\overline{\nu} (\mathbf{E}_n)}{\overline{\nu} (\mathbf{E}_n)} = \frac{\overline{\nu} \mathbf{r}}{\overline{\nu} (\mathbf{E}_n)} + \frac{\mathbf{a}}{\overline{\nu} (\mathbf{E}_n)} \left[\mathbf{E}_n - \Delta \overline{\mathbf{E}}_{\mathbf{k}} (\mathbf{E}_n) \right] \dots$$
(4)

В (4) слева имеем величину $R = \overline{\nu}(E_n)/\overline{\nu}(\widetilde{E}_n)$, найденную в относительных измерениях. Справа стоит искомое соотношение $\overline{\nu}_{\tau}/\overline{\nu}(\widetilde{E}_n)$, коэффициент $a/\overline{\nu}(\widetilde{E}_n)$ и член в скобках, величина которого известна из измерений

Е _л , Мэв	R эксп	٨/٦	$<\Omega_0>/<\widetilde\Omega_0>$	x/x	$\mu_0/\widetilde{\mu_0}$	R
0,08	1,014±0,005	1,0022	0,9968	0,9989	1,0035	1,011±0,007
0 ,20	1,000 ±0,006	1,0004	0,9980	1,0000	1,0003	1,002±0,008
0,30	0,990±0,005	1,0003	0,9997	1,0007	1,0002	$0,992 \pm 0,005$
0,40	1,000 ± 0,000	1,0000	1,0000	1,0000	1,0000	1,000±0,000
0,50	1,005±0,004	0,9997	1,0005	0,9996	0,9998	1,004±0,005
0,60	1,014±0,005	0,9994	1,0005	0,9996	0,9997	1,012±0,005
0,70	1,025±0,005	0,9995	1,0004	0,9986	0,9996	1,022±0,006

ТАБЛИЦА 1. РЕЗУЛЬТАТЫ ОТНОСИТЕЛЬНЫХ ИЗМЕРЕНИЙ ДЛЯ УРАНА-233



Рис. 3. Сравнение результатов измерения $\overline{\nu}(E_n)$ для урана-235: • – результаты данной работы (камера деления с торием-232 в качестве детектора вторичных нейтронов); • – результаты работы [1] с учетом поправок по методике, изложенной в данной работе (детектор вторичных нейтронов – сцинтилляционный счетчик); ∇ – значения $\overline{\nu}(E_n)$, полученные по $\Delta \overline{E}_k(E_n)$ [1] с помощью уравнения баланса (2); о – данные Мазера и др. [6]; — – данные Медоуза и Уолена [7]; • – данные Медоуза и Уолена [10]; r_{Δ} – данные Гопкинса и Дивена [4]; • – данные Колдо [9].

 $\Delta \mathbf{E}_{\mathbf{x}}$ при энергии нейтронов \mathbf{E}_n . Для всех экспериментальных значений R (E_n) и $\Delta \mathbf{E}_{\mathbf{x}}$ (E_n) из исследованного диапазона E_n получаем систему уравнений с неизвестными коэффициентами $\overline{\nu}_{\tau}/\overline{\nu}$ ($\widetilde{\mathbf{E}}_n$) и а/ $\overline{\nu}$ ($\widetilde{\mathbf{E}}_n$), которые находятся по методу наименьших квадратов. В результате расчетов получаем величину $\overline{\nu}$ ($\widetilde{\mathbf{E}}_n$) и а. Найденные таким образом значения а оказались равными 0,070 ±0,005 Мэв и 0,080 ±0,005 Мэв⁻¹ для урана-233 и урана-235 соответственно, что существенно ниже ожидаемых значений 0,13 - 0,15 Мэв⁻¹.

Такая разница могла возникнуть в результате приближенности предположения $\Delta Mc^2 = 0$. Воспользуемся соотношением (3) и подставим в правую часть величину ν (E_n), определенные в результате расчетов по методу наименьших квадратов, принимая значение а равным 0,14 Мэв⁻¹. Получаем значение ΔMc^2 для E_n из исследованного диапазона. Для урана-235 величина ΔMc^2 оказалась равной $\approx -0,3\pm0,3$ Мэв, начиная с 0,08 до 1 Мэв. Иными словами, изменение энергии, реализуемой в виде E_k и ν , за счет деформации Y (M, z) и вариаций \overline{E}_{γ} оказалось невелико. Для урана-235 до E_n = 0,5 Мэв $\Delta Mc^2 \approx 0\pm0,3$ Мэв. При энергиях

КУЗНЕЦОВ и СМИРЕНКИН

Е _п , Мэв	R _{экслі}	λ/λ	< ດ> /< ດີ>	x/x	μ /μ	R
0,08	0,982±0,005	1,0014	0,9940	1,0007	1,0031	0,98620,006
0,08*	0,970±0,006	1,0014	0,9941	1,0007	1,0000	0,979±0,008
0,20	1,015±0,006	1,0008	0,9973	0,9998	1,0005	-1,013±0,007
0,30	1,009±0,006	1,0002	0,9994	0,9996	1,0002	E1,008 ±0,006
0,31* [.]	$0,995 \pm 0,006$	1,0002	0,9994	0,9996	1,0000	0,997±0,006
0,40	1,000±0,000	1,0000	1,0000	1,0000	1,0000	1,0000 ±0,000
0,50	0,998±0,005	0,9998	1,0006	1,0002	1,9998	-0,998±0,005
0,55*	0,976±0,006	0,9998	1,0010	1,0003	1,0000	0,980±0,006-
0,60	$0,994 \pm 0,004$	0,9997	1,0016	1,0004	0,9997	0,995 +0,005
0,67*	0,994±0,006	0,9996	1,0026	1,0004	1,0000	0,992±0,006~
0,70	0,994±0,005	0,9996	1,0030	1,0004	0,0097 (10,994 ±0,005
0,78*	0,993±0,006	0,9996	1,0034	1,0004	1,0000	0,992±0,007-
0,99	1,010±0,008	0,9995	1,0034	1,0005	1,0000	1,005±0,009-
	ł			1	1	· ·

ТАБЛИЦА 2. РЕЗУЛЬТАТЫ ОТНОСИТЕЛЬНЫХ ИЗМЕРЕНИЙ $\vec{R} = \overline{\nu} (E_n) / \overline{\nu} (\widetilde{E}_n) ДЛЯ УРАНА-235 (звездочкой помечены результаты измерений с сцинтилляционным пороговым детектором [1])$

 $E_n = 0,6$ и 0,7 Мэв величина ΔMc^2 уменьшается, достигая при $E_n = 0,7$ Мэв значения – 0,6±0,3 Мэв.

В качестве второго приближения учитывались значения ΔMc^2 для каждого E_n и процедура обсчета методом наименьших квадратов повторялась вновь. Для урана-233, кроме того, точки для $E_n = 0,6$ и 0,7 Мэв не учитывались. Для урана-235 полученное во втором приближении значение $\overline{\nu_r}/\overline{\nu}$ (\widetilde{E}_n) оказалось равным 0,974 ±0,001, что превосходно согласуется с $\overline{\nu_r}/\overline{\nu}$ (\widetilde{E}_n) = 0,975 ±0,07, определенным из эксперимента путем прямого сравнения. Для урана-233 $\overline{\nu_r}/\overline{\nu}$ (\widetilde{E}_n) = 0,013 ±0,0001, a = 0,120 ±0,007.

Ошибки определения $\overline{\nu}_{T}/\overline{\nu}/\widetilde{E}_{n}$) расчетом по методу наименьших квадратов не учитывают того факта, что использовавшиеся значения а нам известны только по оценкам. Поэтому $\overline{\nu}(\widetilde{E}_{n})$ анализировались по коэффициенту а. Для этого с помощью (3) находились значения $\overline{\nu}(\widetilde{E}_{n})$ для а, изменявшихся от 0,04 до 0,20. Полагая, что истинное значение а находится в интервале 0,10 до 0,18, по кривой $\overline{\nu}(\widetilde{E}_{n}) = f(a)$ определялся разброс значений $\overline{\nu}(E_{n})$, который оказался равным 0,2% урана-233 и 0,9% для урана-235. Для большей гарантии от возможно неучтенных факторов ошибка $\overline{\nu}(\widetilde{E}_{n})$ для урана-233 была принята равной 1%, а для урана-235 взято значение $\overline{\nu}(\widetilde{E}_{n}) = 2,491 \pm 0,007$ из эксперимента. Величина $\overline{\nu}_{T-}$ с-учетом-запаздывающих нейтронов для урана-233 принималась равной 2,494 ± 0,000,/для урана-235 2,430 ± 0,000 в соответствии с [3].

На рис.2 и 3-представлено сравнение полученных в настоящей работе данных с имеющейся совокупностью. В данных других авторов учтен вклад запаздывающих нейтронов. Они нормированы на значения $v_{\rm T}$, принятые в данной работе, причем опорные значения $\overline{v}_{\rm T}$ принимались всюду без ошибки.

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Как видно из рис.2 и табл.1 тенденция $\overline{\nu}(E_n)$ для урана-233 обнаруживает спад на≈1,5% от теплового значения до области E_n = 0,2-0,3 Мэв в полном соответствии с предсказаниями работы [1]. Новые данные для урана-235 хорошо согласуются с данными, полученными в [1], (табл.2, рис.3), причем подъем $\overline{\nu}$ при $E_n = 0,2$ и 0,3 Мэв подчеркнут еще сильнее.

Недавно появилась еще одна работа [11], объясняющая структуру в $\overline{\nu}$ (Е_n) для урана-235. Нерегулярности в ходе $\overline{\nu}$ при Е_n <1 Мэв в этой работе связываются с вкладом состояний с К = 2 и положительной четностью, аналогичных у-вибрационным состояниям стабильных ядер. По мнению авторов [11] энергия возбуждения этих каналов при делении р-нейтронами переходит в нагрев осколков, что приводит к увеличению $\overline{\nu}$ в области $E_n \approx 0.2$ Мэв. Энергия возбуждения всех прочих коллективных состояний внутри энергетической щели согласно [11] идет в кинетическую энергию осколков. Относительно кинетической энергии деформации принимается предположение, противоположное работе [1]: эта энергия не переходит во внутреннее возбуждение, т.е. добавляется к Ек.

Нетрудно показать, что участием каналов с К = 2⁺ можно объяснить и результаты для урана-233.

Исходя из имеющихся экспериментальных данных и представлений о переходном состоянии делящегося ядра в настоящее время нельзя отдать предпочтения какой-либо из этих интерпретаций. Необходима экспериментальная проверка альтернативных предположений, принимавшихся авторами [1, 11].

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О ВЛИЯНИИ ВАРИАЦИЙ ЭНЕРГЕТИЧЕСКИХ И МАССОВЫХ РАСПРЕДЕЛЕНИЙ ОСКОЛКОВ ДЕЛЕНИЯ НА ЭНЕРГЕТИЧЕСКУЮ ЗАВИСИМОСТЬ 7

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ФИЗИКО-ЭНЕРГЕТИЧЕСКИЙ ИНСТИТУТ, ОБНИНСК СССР

Abstract — Аннотация

THE EFFECT OF VARIATIONS IN THE ENERGY AND MASS DISTRIBUTIONS OF FISSION FRAGMENTS ON ENERGY DEPENDENCE. The average number of neutrons emitted in a single fission event is determined mainly by the excitation energy of the fragments. It has been shown in a number of studies that an increase in the excitation energy of a fissionable nucleus is usually accompanied by an increase in the number of fission neutrons. The quantity $\overline{\nu}$ being measured is an average for all the fragment masses. When there is a change in the excitation energy of a nucleus undergoing fission, the yield and the kinetic energy of the fragments also change, so that these factors have an effect on the value of $\overline{\nu}$.

We can then write

$$\overline{E}_{excit.} = \int E_{fiss} Y(M) dM - \int E_{kin.} Y(M) dM + E_n + B_n$$

or, approximately,

$$\Delta \overline{\nu} = a \left[\overline{\Delta E}_{\text{fiss.}} - \overline{\Delta E}_{\text{kin.}} + \Delta E_{\text{n}} \right]$$

The paper discussed the effect of changes in the kinetic energy and yield of fragments in fast-neutron fission of 233 U, 255 U and 238 U on the energy dependence of $\overline{\nu}$ for neutron energies in the range 0-7 MeV. Information is obtained regarding the kinetic energy and yield of fragments by simultaneous measurement (with semiconductor detectors) of the kinetic energies of fragment pairs.

It is shown that the changes in the average kinetic energy and yield of fragments of different mass, which have been observed in many cases, can make an appreciable contribution to the change in $\overline{\nu}$.

О ВЛИЯНИИ ВАРИАЦИЙ ЭНЕРГЕТИЧЕСКИХ И МАССОВЫХ РАСПРЕДЕЛЕНИЙ ОСКОЛКОВ ДЕЛЕНИЯ НА ЭНЕРГЕТИЧЕСКУЮ ЗАВИСИМОСТЬ **7**. Среднее число нейт-

ронов, испускаемых на один акт деления, определяется, главным образом, энергией возбуждения осколков. Э целом ряде работ было показано, что с увеличением энергии возбуждения делящегося ядра, как правило, увеличивается и число нейтронов деления. Измеряемая величина $\overline{\nu}$ является усредненной по всем массам осколков. При изменении энергии возбуждения делящегося ядра меняется как выход осколков, так и их кииетическая энергия, поэтому все эти факторы отразятся на величине $\overline{\nu}$.

Можно записать, что

$$\overline{\mathbf{E}}_{\mathbf{B}036} = \int \mathbf{E}_{\mathbf{gen}} \mathbf{Y}(\mathbf{M}) \, d\mathbf{M} - \int \mathbf{E}_{\mathbf{K}\mathbf{H}\mathbf{H}} \mathbf{Y}(\mathbf{M}) \, d\mathbf{M} + \mathbf{E}_{\mathbf{n}} + \mathbf{B}_{\mathbf{n}}$$

Или, приблизительно

$$\Delta \overline{\nu} = \mathbf{a} \left[\Delta \overline{\mathbf{E}}_{gen} - \Delta \mathbf{E}_{KHH} + \Delta \mathbf{E}_{n} \right]$$

* В настоящее время имеются данные [2, 3], свидетельствующие о том, что часть мгновенных нейтронов испускается делящимся ядром. Рассматривается вопрос о влиянии изменений кинетических энергий и выходов осколков при делении урана-233, урана-235 и урана-238 быстрыми нейтронами на энергетическую зависимость $\overline{\nu}$ при энергии нейтронов от 0 до 7 Мэв. Информация о кинетической энергии и выходах осколков получалась путем одновременного измерения кинетических энергий парных осколков с помощью полупроводниковых счетчиков.

Показано, что в ряде случаев наблюдавшиеся изменения средней кинетической энергии и изменения выходов осколков с различными массами могут внести заметный вклад в изменеиме $\overline{\nu}$.

Тщательное экспериментальное исследование зависимости среднего числа мгновенных нейтронов деления $\overline{\nu}$ от энергии нейтронов, вызывающих деления, показало, что эта зависимость не может быть описана линейной функцией [1]. В той же работе отмечалось наличие корреляции между изменениями $\overline{\nu}$ и кинетической энергии осколков деления. С увеличением энергии возбуждения делящегося ядра меняется целый ряд параметров, характеризующих осколки деления, и это приводит к отклонению от линейного роста $\overline{\nu}$. Если принять, что все мгновенные нейтроны испускаются осколками деления*, то величину $\overline{\nu}$ можно выразить следующим образом:

$$\bar{\nu} = \sum_{i} \left(\frac{\mathbf{E}_{f}^{i} - \mathbf{E}_{\kappa}^{i} - \mathbf{E}_{\gamma}^{i} + \mathbf{E}_{x}}{\eta_{i} + \mathbf{E}_{B}^{i}} \right) \mathbf{Y}_{i}$$
(1)

Здесь введены следующие обозначения: E_i^i – энергия деления; E_k^i – средняя кинетическая энергия осколков; η_i – средняя кинетическая энергия нейтронов, испускаемых из данного осколка; E_B^i – энергия связи нейтронов в ядре-осколке; E_j^i – средняя энергия, уносимая мгновенными γ квантами из осколков деления; Y_i – выход осколков с данными массами; E_x – энергия возбуждения делящегося ядра.

Изменение $\tilde{\nu}$, обусловленное вариацией параметров выражения [1], можно приблизительно записать в следующем виде:

$$\begin{split} \delta \overline{\nu} &= \mathbf{K}_0 \Sigma \delta \mathbf{E}_f^{i} \mathbf{Y}_i - \mathbf{K}_0 \Sigma \delta \mathbf{E}_{\mathbf{K}}^{i} \mathbf{Y}_i - \mathbf{K}_0 \Sigma \delta \mathbf{E}_{\mathbf{\gamma}}^{i} \mathbf{Y}_i + \mathbf{K}_0 \delta \mathbf{E}_{\mathbf{x}} - \\ &- \overline{\nu}_0 \mathbf{K}_0 \Sigma \left(\delta \eta_i + \delta \mathbf{E}_B^{i} \right) \mathbf{Y}_i + \Sigma \nu_i \delta \mathbf{Y}_i \qquad \qquad \mathbf{K}_0 = \langle \frac{1}{\eta + \mathbf{E}_B} \rangle \end{split}$$

Таким образом, величина измерения $\overline{\nu}$ зависит от изменения энергии возбуждения делящегося ядра, от изменения кинетической энергии и выходов осколков с данными массами и от перераспределения заряда между осколками.

В данной работе были измерены распределения осколков по массам и кинетическим энергиям при делении урана-233, урана-235, урана-238 быстрыми нейтронами. Это дало возможность определить вклад в $\delta \overline{\nu}$, обусловленный изменениями кинетической энергии и выходов осколков.

Экспериментальный метод [4] состоял в изменении кинетической энергии парных осколков с помощью поверхностно-барьерных полупроводниковых счетчиков. Экспериментальные выходы осколков с разными массами были приведены к выходам осколков до испускания нейтронов.

Результаты измерений приведены в таблице. Изменения $\delta \overline{\nu}$, обусловленные изменением кинетической энергии и выходов осколков, для

			Уран-233	-		
Еп, Мэв	0,4±0,05	1,0±0,1	2,6±0,09	$5,6\pm0,2$	$6,0 \pm 0,2$	7,0±0,19
$\Sigma \nu_i \Delta Y_i$	-0,0083	-0,0046	-0,0064	- 0,0017	0,012	0,0013
ΚοΣΔΕκΥί	0,031	0,083	0,035	0,031	0,043	-0,0056
δν	- 0,039	- 0,088	- 0,041	- 0,033	- 0,031	0,007
······································	P Bi sa bi ri an ri da	1	Уран-235	5		
Еп, Мэв	0,75±0,1	2,0±0,1	5,0±0,3	6,0±0,2	7,0±0,18	15,0±0,5
$\Sigma \nu_i \Delta Y_i$	-0,007	0,014	0,005	0,022	0,022	0,125
$K_0 \Sigma \Delta E_{\kappa} Y_i$	0,004	-0,021	-0,021	0,004	0,029	-0,174
δν	-0,011	0,035	0,035	0,018	-0,007	0,299
			Уран-238			
Еп, Мэв	5,0±0,2	7,0±0,18				
$\Sigma \nu_i \Delta \mathbf{Y}_i$	0,014	0,013				
$K_0 \Sigma \Delta E_\kappa Y_i$	0,006	- 0,022				
δν	0,008	0,035				

ТАБЛИЦА. ВКЛАД, ОБУСЛОВЛЕННЫЙ ВАРИАЦИЯМИ ЭНЕРГЕТИЧЕСКИХ И МАССОВЫХ РАСПРЕДЕЛЕНИЙ ОСКОЛКОВ ПРИ ДЕЛЕНИИ УРАНА-233, УРАНА-235 И УРАНА-238 БЫСТРЫМИ НЕЙТРОНАМИ

урана-233 и урана-235 вычислены относительно $\overline{\nu}$ при делении тепловыми нейтронами, а для урана-238 — относительно $\overline{\nu}$ при делении нейтронами с энергией 2 Мэв. Точность измерений, выраженная в единицах $\overline{\nu}$, всюду составляет около ±0,025. Изменения выходов осколков при делении рассматриваемых ядер нейтронами с энергией от 0 до 7 Мэв слабо отражаются на величине $\delta\overline{\nu}$. В этой области энергий основной вклад в $\delta\overline{\nu}$ вносят изменения кинетической энергии осколков.

При делении нейтронами с энергией 15 Мэв величина $\delta \overline{\nu}$ составляет около 8% от $\overline{\nu}$ и состоит приблизительно из одинаковых вкладов от изменения кинетической энергии осколков и от изменения выходов осколков.

Вопрос об изменении распределения заряда между осколками с фиксированными массами при увеличении энергии возбуждения делящегося ядра изучен недостаточно глубоко, поэтому трудно заключить что-либо определенное относительно величин $\delta \mathbf{E}_{j}^{i}$; $\delta \mathbf{E}_{j}^{i}$; $\delta \eta_{i}$ и $\delta \mathbf{E}_{g}^{i}$.

Отклонения экспериментальных значений $\overline{\nu}$ от линейной зависимости имеют тот же знак и тот же порядок величины, что и величины $\delta\overline{\nu}$, подсчитанные в данной работе. Этот факт свидетельствует о том, что перераспределение заряда между осколками, если оно имеет место, оказывает слабое влияние на $\delta\overline{\nu}$ в рассматриваемой области энергий ($E_{\pi} < 6$ Мэв).

КУЗЬМИНОВ и др.

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ЗАВИСИМОСТЬ СЕЧЕНИЯ СИММЕТРИЧНОГО ДЕЛЕНИЯ УРАНА-238 ОТ ЭНЕРГИИ НЕЙТРОНОВ

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Abstract — Аннотация

THE DEPENDENCE OF THE SYMMETRICAL ²³⁸U FISSION CROSS-SECTION ON NEUTRON ENERGY. The yields of various products of symmetrical and asymmetrical ²³⁸U fission were compared by radiochemical means. Fission was produced by monoenergetic neutrons. Measurements were made for neutron energies of 1.5, 2, 3, 4, 5, 13, 15, 16, 5 and 18 MeV. The ratios of the product yields for symmetrical

fission to those for asymmetrical fission at neutron energies of 2, 15, 13 and 18 MeV were $\frac{1}{500}$, $\frac{1}{90}$, $\frac{1}{8}$ and $\frac{1}{8}$, respectively.

The experimental results are compared with those obtained from existing fission models.

ЗАВИСИМОСТЬ СЕЧЕНИЯ СИММЕТРИЧНОГО ДЕЛЕНИЯ УРАНА-238 ОТ ЭНЕРГИИ НЕЙТРОНОВ. Радиохимическим методом сравнивались выходы некоторых продуктов симметричного и асимметричного деления урана-238. Деление вызывалось моноэнергетическими нейтронами. Измерения проведены при энергиях нейтронов I,5; 2; 3; 4; 5; 13; 15; 16,5; 18 Мэв. Выход продуктов симметричного деления при энергиях нейтронов 2; 5; 13 и 18 Мэв соответственно равен 1/500; 1/90; 1/8; 1/8.

Результаты эксперимента сравниваются с предсказаниями существующих моделей деления.

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INTERPRETATION OF NEUTRON-INDUCED FISSION CROSS-SECTIONS AND RELATED DATA

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Abstract

INTERPRETATION OF NEUTRON-INDUCED FISSION CROSS-SECTIONS AND RELATED DATA. The paper discusses the interpretation of the slow neutron cross-sections, ratios of capture-to-fission crosssections in the keV region, and degree of asymmetry in the mass distribution of fission products in the framework of the channel theory of fission. Problems associated with the fission cross-sections of even nuclei are mentioned, and a new hypothesis concerning the behaviour of mean fission widths are advanced as a possible solution.

1. INTRODUCTION

Our understanding of the neutron cross-sections of the non-fissile nuclei now seems fairly complete. The resonance spacings, s-wave neutron strength functions and radiative capture widths have been measured for many nuclei and their phenomenological interpretation by means of sophisticated complex potential models is quite satisfactory. The various elastic and inelastic scattering data at higher energies can mostly be interpreted with the same kind of model. As a result it is possible to make reasonable, though not completely accurate, attempts at calculating unmeasured cross-sections required for reactor design purposes. Such cross-sections may be averages over rather wide energy intervals containing very many resonances, or may be statistical models of the microscopic resonance structure such as required for calculations of the Doppler temperature coefficient.

The question I pose in this paper is: how far is it possible to carry out the same programme for fissionable nuclei ? To a considerable extent this reduces to asking how well we understand the fission process at least as far as the scission point. The basic theory for understanding fission cross-sections at low and medium neutron energies is the channel theory of A. Bohr [1], and in this paper I shall rest my discussion on this. The main imponderables of this theory are generally assumed to be the energies and nature of the internal nuclear states associated with the transition of the nucleus through the fission barrier as well as the penetrability of the latter. These states are sometimes conveniently termed the transition states (although this is not quite the meaning of the term as used by Wigner [2] in his original paper on unimolecular reactions) or are, more often, known as the fission channels. So far, their properties have been deduced mainly from our views on nuclear collective behaviour and partly from a variety of experimental evidence. In the first place I shall review the interpretation of the slow neutron cross-sections of the common fissionable nuclei.

2. CHANNEL SCHEMES FOR THE COMMON FISSIONABLE NUCLEI

2.1 Analysis of Resonance Cross-Section Data

In principle, the direct observation of the resonances in low energy cross-sections should provide estimates of the number and penetrability of

<u>TABLE I</u> Fission channel scheme for s-wave resonances of $235_{\rm U}$ + n

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis (K)	Spin and parity relevant to compound nucleus (J ^T)	Energy of transition state with respect to neutron threshold of ²³⁶ U (in MeV)
Mass asymmetry vibration (+ rotation)	0	3-	~ -0.7
Bending vibration (+ rotation)	1	3-, 4-	+0.1
Combination state of gamma vibration and mass asymmetry vibration (+ rotation)	2	• 3 ⁻ , 4 ⁻	+0.25

Chosen to fit resonance data

the accessible fission channels. In practice, there are various effects, due to the rather large ratio of fission width to spacing of the compound mucleus levels, that can mislead one in interpreting these data.

The simplest of these is the low peak cross-section and large width, due to a low neutron width to fission width ratio, of many simple resonances, causing them to be hidden against the general background cross-section [3]. The second is the occurrence of "mixed" resonances, a single peak in the cross-section due to the superposition of two resonances of different total angular momentum [4]. The third effect, which occurs rather less frequently than the last, is the quasi-resonance effect [4]. In this, two (or more) close eigenlevels with the same quantum numbers can interfere in a dramatic way to cause a single peak in the cross-section that is narrower than the width associated with either level. The remainder of the sum of the widths occurs as a low, broad resonance term underlying, and hidden by, the sharp peak. All these effects have the same result: if the cross-section is analysed in the usual way as essentially a sequence of single level terms, the resulting ratio of mean fission width to level spacing can be considerably underestimated.

In view of these difficulties I have favoured a statistical treatment of the data that compares the results of single level analysis of the resonance peaks, as well as other material such as minimum cross-section behaviour and ratios of capture to fission, with similar quantities derived from statistical simulations of cross-sections using R-matrix theory. The method and results of this kind of analysis have been described previously [4].

2.2 Fission Channels for the Compound Nucleus 236

The spin and parity of the ²³⁵U target nucleus are $I^{\pi} = 7/2^{-}$. From this, compound nucleus levels formed in ²³⁶U by s-wave neutron bombardment have total angular momentum and parity $J^{\pi} = 3^{-}$, 4⁻. The transition states which best fitted the resonance cross-section data [4] are given in Table I. With these thresholds I have used the Hill-Wheeler [5] harmonic barrier penetration formula to calculate the total fission strength function,

$$\frac{2\pi T_{(F)}}{\overline{D}} = \frac{\sum_{f} \frac{1}{1 + \exp\left[-2\pi (E - E_f)/\hbar\omega_f\right]},$$
(1)

with $\hbar\omega_r$ always 0.5 MeV; 0.4 to 0.5 MeV is the value suggested by spontaneous fission data [6] and the slope of the yield curve of (d,pf) reactions [7]. Assuming these properties for the fission channels, one can calculate the average fission and capture cross-sections in the s-wave region and compare with other data. For example, at 1 keV the ratio a of these two cross-sections is calculated to be 0.91.

In this calculation the usual simple expression for the average crosssection of a compound nucleus reaction has been used; it is

$$\langle \sigma_{\alpha\beta} \rangle = 2\pi \lambda_{\alpha}^2 q(J) \langle \frac{\Gamma_{(\alpha)}}{\Gamma} \rangle$$
 (2)

TABLE II

Alternative fission channel scheme for s-wave resonances of $\frac{235_{\text{U}}}{1000} + n$

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis (K)	Spin and parity relevant to compound nucleus (J^{π})	Energy of transition state with respect to neutron separation energy (in MeV)
Mass asymmetry vibration (+ rotation)	0	. 3	~ -0.6
Bending vibration (+ rotation)	1	3-, 4-	~ -0.3
Combination state of gamma vibration and mass asymmetry vibration (+ rotation)	2 .	37,4	~ +0.4

Chosen to fit resonance data and a-value to 1 keV

which is usually written

$$\langle \sigma_{\kappa\beta} \rangle = 2 \pi^2 \chi_{\kappa}^2 g(J) \frac{\overline{\Gamma}_{(\kappa)}}{\overline{\Gamma}} \frac{\overline{\Gamma}_{(\kappa)}}{\overline{\Gamma}} \delta$$
(3)

where 8 can differ from unity to some degree) to express the fact that the statistical behaviour of resonance partial widths must be considered [8]. This expression is derived from averaging over a sequence of single-level Breit-Wigner terms in the cross-section. The fissile nucleus crosssections, however, exhibit considerable deviations from single-level behaviour, and we must consider the effect of these on the average crosssection. The simplest way to derive a many-level average cross-section expression having Eq. (3) as its leading term is to start from the S-matrix formalism of Humblet and Rosenfeld [9]. With this procedure it is found that additional terms become increasingly important with increasing value of $\overline{\mathbf{T}}$ /D, but also that these terms vanish if the channel-pole phase factors have certain properties of randomness. Unfortunately, this attractive randomness assumption cannot be made. The requirement that unitarity (conservation of flux in the stationary wave representation of the nuclear reaction) be imposed on the S-matrix leads to correlations in these phase factors and prevents the higher order terms from disappearing (see Appendix 1). In addition, the statistical behaviour of the partial widths will differ from those of the narrow resonance case, with the consequence that the & -factors in Eq. (3) will be different from those normally calculated. These correlations and statistical properties of broad resonances have by no means been fully investigated, with the result that our average cross-section calculations cannot be made completely accurate.

In the case of 235 U, I have made a few calculations by direct computations over many randomly chosen resonances of the average crosssection and compared them with the "no interference" form (Eq. 3). It is found that the many-level effects increase a by roughly 10°/o. On the other hand, it is necessary to take into account the (n,Yf) reaction [10] whereby a radiative transition from the resonance state to a state below the neutron threshold but above the fission threshold can be followed by fission. This is, of course, measured as a fission event and decreases the effective value of a by about the same amount. These compensatory effects are in any case of the same order of magnitude as the uncertainties in the radiation width value assumed for the calculations so we neglect them in all our subsequent calculations.

Now, although this calculated value of a agrees reasonably well with the experimentally observed value in the range 0 - 100 eV, the observed value of a for 100-eV intervalsup to about 2 keV fluctuates about the value 0.5. In fact, the mean value over 100 eV intervals from 100 eV to 1 keV is $0.57 \pm .19$ [10]. This strongly suggests that the cross-sections in the 0 to 100-eV range are dominated by a statistically extreme sample of levels (such as we are quite familiar with in the non-fissile nuclei). If we look at the statistical comparisons with simulated cross-sections again, we find that the scheme of channels shown in Table II would quite well explain the 235 U resonance data apart from the pseudo-fission strength function value, which is rather low in the data.

Average cross-sections calculated with these channels result in an a-value at 1 keV of about 0.52, in rather good agreement with the data. It is interesting to calculate how a is expected to fluctuate over finite intervals. Twelve calculations of a were made by selecting level parameters

TABLE III

Fission channel scheme for p-wave resonances of 235 U + n

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Deduced from s-wave scheme of Table II

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Nature of transition state	Projection of angular momentum on cylindrical symmetry axis K	Spin and parity relevant to compound nucleus (J ^{TA})	Energy of transition state with respect to neutron separation energy (in MeV)	Comment
"Ground" (+ rotation)	0	2*, 4*	-0.8	This energy is established by the (d,pf) reaction [12].
Combination state of bending vibration and mass asymmetry vibration (+ rotation)	1	2 ⁺ , 3 ⁺ , 4 ⁺ , 5 ⁺	0.1	This energy is fixed by the s-wave scheme assumed in Table II.
Gamma vibration (+ rotation)	2	2 ⁺ , 3 ⁺ , 4 ⁺ , 5 ⁺	+0.2	This energy is fixed by the s-wave scheme assumed in Table II.

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TABLE IV

Fission channel scheme for s-wave resonances of 233U + n

Suggested by analysis of resonance data and other phenomena

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis K	Spin and parity relevant to compound nucleus (J^{π})	Energy of transition state relevant to neutron separation energy (in MeV)	Comments
"Ground" (+ rotation)	0	2+	-1.4	Energy established by (d,pf) reaction [12]
Gamma vibration (+ rotation)	2	2 ⁺ , 3 ⁺	-0.6	Perhaps indicated by angular distribution of fission products in (d,pf) reaction [14]
Combination state of mass asymmetry vibration and bending vibration (+ rotation)	1	2 ⁺ , 3 ⁺	~ -0.3	•
2 phonons in gamma vibration (+ rotation)	ο.	2 ⁺	~ +0.1	

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TABLE V

Fission channel scheme for p-wave resonances of $\frac{233}{U + n}$

Deduced from s-wave scheme of Table IV

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis K	Spin and parity relevant to compound nucleus (J^{π})	Energy of transition state relevant to neutron separation energy (in MeV)
Mass asymmetry vibration (+ rotation)	0	1", 3	~ -0.9
Bending vibration (+ rotation)	1	1, 2, 3, 4	~ -0.8
Combination state of gamma vibration and mass asymmetry vibration (+ rotation)	2	1, 2, 3, 4	~ -0.2

.
TABLE VI

Fission channel scheme for s-wave resonances of 239Pu + n

Deduced from resonance data

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis K	Spin and parity relevant to compound nucleus (J ^{TT})	Energy of transition state relevant to neutron separation energy (in MeV)	Comments
"Ground"	0	o ⁺	-1.5	Given by (d,pf) reaction [12].
2 phonons of gamma vibration	0	0 ⁺	0.0	Also indicated by (d,of) angular distribution [14]
Combination state of mass asymmetry vibration and bending vibration	1	1+	+0.15	

TABLE VII

Fission channel scheme for p-wave resonances of 239Pu + n

Inferred from s-wave scheme

Nature of transition state	Projection of angular momentum on cylindrical symmetry axis (K)	Spin and parity relevant to compound nucleus (J^{π})	Energy of transition state relevant to neutron separation energy (in MeV)
Mass asymmetry vibration	0	1-	~ -0.9
Bending vibration (+ rotation)	1	1, 2	~ -0.45
Combination state of gamma vibration and mass asymmetry vibration	2	2	-0.15
Combination state of gamma vibration and bending vibration (+ rotation)	1	1 ⁻ , 2 ⁻	~ +0.3

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randomly in 80 eV intervals. The results varied from 0.31 to 0.68. The mean and root mean square values of these calculations were 0.56 and 0.11. After the quoted experimental errors are allowed for in the a-values measured in 100-eV intervals from 100 eV to 1 keV their root mean square value is 0.12. This encourages the belief that the second set of assumed fission channels is better than the first, and that the experimental data leading to the first are due to a statistically extreme sample of level parameters.

It is now interesting to see if we can extrapolate into the p-wave region. The channel scheme could well be that shown in Table III. Notice now that the channels in both schemes together are so uniformly spaced that no structure would be observed in the (d,pf) reaction on 2350 below neutron threshold if $\hbar\omega_{\rm f}$, the tunnelling parameter, is assumed to be ~ 0.5 MeV. If the average cross-sections are now calculated at, say, 60 keV, allowing for inelastic scattering to the known states of 2350 and using a p-wave neutron strength function of $\overline{T_n}^{(1)}/\overline{D} = 2 \times 10^{-4}$ as established by Uttley [13], we obtain the result that $\alpha = 0.375$. This is to be compared with the experimental datum of ref. [13], $\alpha = 0.33 \pm 0.02$. The agreement is very encouraging.

2.3 Fission Channels for the Compound Nucleus 234U

The statistical analysis of the resonance data of 233 U would suggest a channel scheme for the s-wave region like that in Table IV. The calculation of a at 1 keV from this channel scheme gives a ~ 0.23. This is to be compared with the experimental value which is fluctuating about 0.3 in the 0.6 to 1-keV region [16]. From the even parity channel scheme we might deduce the odd-parity channel scheme for the p-wave region shown in Table V. The channel schemes of Tables IV and V could give the structure observed in the (d,pf) reaction [12]. With these channels the calculated values of a at 30 keV is 0.23. The experimental value is 0.11 ± .02 [15]. This disagreement suggests that we may still be underestimating the number of fission channels available at low neutron energies.

2.4 Fission Channels for the Compound Nucleus 240 Pu

The analysis of the available resonance data is rather more tentative than in the previous two cases. Since the analysis in ref. [4], which suggested more than one fully open channel for the 0⁺ levels, more extensive spin assignments [17, 18] indicate that the 1⁺ levels have a penetration factor of about 0.1. The suggested channel scheme is shown in Table VI. From this the value of a at 1 keV is calculated to be 0.7.

This is in rather unsatisfactory disagreement with very recent experimental evidence [19] indicating that a is rather higher than this in the keV region. The latter value suggests that the K = 1, $J^{\pi} = 1^+$ channel should be a little higher than given above. Clearly very many more data on the resonances are desirable in order to establish this point.

The odd parity channel scheme inferred from the s-wave scheme is shown in Table VII.

At 30 keV a is calculated to be 0.26. Very recent measurements [20] indicate a value a little above 0.3. The raising of the K = 1, J^{π} = 1⁺ state by as much as 150 keV only increases this estimate of a to 0.32, so this set of channels for the p-wave region seems quite satisfactory.



FIG.1. Vibration levels in the octupole deformation parameter to explain the channel theory of asymmetric mass division.

3. ASYMMETRY IN THE FISSION-PRODUCT MASS DISTRIBUTION

The idea suggested by A. Bohr [1] and J. Wheeler [21] that not only the angular distribution of fission products but also their mass distribution could be governed by the properties of the transition states at the saddle point has recently received strong experimental support. The group of Cowan and others at Los Alamos [22] have carried out a radiochemical analysis of the products of neutron-induced fission of ²³⁹Pu in the resonance region. Thus they found the ratio of ¹¹⁵Cd (a nearly symmetric fission product) to ⁹⁹Mo formed in a large sequence of slow neutron resonances. In many resonances known to be associated with total angular momentum 1 this ratio was found to be low (~ 1 x 10⁻²) while in a few others known to have zero spin there was more symmetric fission (r:Cd/Mo ~ 5 x 10⁻²). In addition, where resonance spins were not known, it was found that a high ratio of ¹¹⁵Cd to ⁹⁹Mo was formed in the resonances of large fission width, which are most likely to have spin zero.

The channel theory explanation of asymmetric mass division employs the principal collective parameter describing the asymmetry of the shape with respect to a plane perpendicular to the cylindrical symmetry axis; this is the octupole expansion coefficient β_3 . If the potential energy as a function of β_3 has the form shown in Figure 1, and it may well have this. CN-23/122



FIG. 2. Asymmetric to symmetric fission mass yield for 235 U+n and 239 Pu+n (from Ref. [25]).

form in the region of the saddle point according to nuclear structure calculations of Johansson [23], the lowest wave function for the octupole parameter has the symmetric form shown. This is interpreted as allowing a small amount of purely symmetric fission ($\beta_3 = 0$). The next eigenstate in the well is raised by only a small energy and has the anti-symmetric form shown. This indicates no symmetric mass yield for fission proceeding through a channel with this collective character. This is the mass asymmetry vibration mentioned several times above. The next anti-symmetric eigenstate of this potential well is expected to lie at a considerably higher energy and need not be considered for the low energy phenomena under discussion here.

From this simple consideration we can immediately pick out the channels in the schemes we have considered which are likely to give rise to particularly low symmetric fission. In the case of 239Pu + n there is only one candidate, the one originally suggested by Griffin [24], with K = 1, $J^{\pi} = 1^+$ and thought to be a combination of mass asymmetry and bending vibration (see Table VI). Thus, the 1⁺ resonances, expected on the average to have very much smaller fission widths than the 0⁺ resonances, are also expected to have much smaller symmetric/asymmetric fission ratios in agreement with the observations (Cowan et al. [22]). In the p-wave region we have a greater choice of channels with a component of the mass-asymmetry vibration (see Table VII) but also some without. Using this scheme and the results of Cowan et al. a calculation of the $99Mo/11^3$ Ag ratio as a



FIG. 3. The neutron-induced fission cross-section of ²³²Th.



FIG. 4. Level density calculated from the pairing correlation model.

function of energy is shown in Figure 2. Here we have also tried to extrapolate the channel scheme into the d-wave region. The curves shown are in fair agreement with radiochemical measurements by Cuninghame et al. [25]. In the same figure the measurements of Cuninghame, Kitt and Rae [26] on the 99Mo/113Ag ratio produced in neutron-induced fission of 2350 are shown. Now it is known that in the slow neutron resonance region of this cross-section the asymmetric/symmetric fission ratio varies only slightly from the thermal neutron value (indicated here by the arrow) [27]. It was always a puzzle, therefore, why the 60-keV value should indicate a smaller degree of symmetric fission than thermal, because this energy is well within the p-wave region which decays through even parity channels towards fission while the pure mass asymmetry vibration has odd parity. The answer to this seems to lie in the channel schemes I have already indicated (Tables II and III). At 1 keV the ratio of the cross-section for fission proceeding through a purely asymmetric fission channel (σ_{aB}) to that for fission proceeding through a channel allowing some symmetric fission (σ_s) is calculated to be 0.37. At 60 keV however it is calculated to be 0.84 in qualitative accord with the observations. This is because the suggested channel schemes allow the combination state of bending and mass asymmetry vibration, with even parity, to come strongly within the p-wave region. Notice that this interpretation would fall if I had attempted to build on the channel scheme suggested by ref. [4] (see Table I), and notice also that it would still fall if the statistical sample of fission widths that actually seems to occur in the 0-to 100-eV region were due to unusually narrow $J^{\pi} = 4^{-}$ resonances rather than to narrow 3⁻ resonances.

4. NEUTRON-INDUCED FISSION OF EVEN TARGET NUCLEI

4.1 Anomalies in Fission Cross-Sections

So far it seems that the channel theory can be made to explain quite a lot of things rather well. But now let us look at the other side of the coin. In contrast to the very heavy odd mass nuclei, even targets are not normally fissile to slow neutrons but show a threshold effect at several hundred keV. Above the threshold, dips are often found in the fission cross-sections; these are usually explained as the effect of inelastic neutron competition after a single channel (or group of channels in a rotational band) has become fully open for fission [28].

One wonders, however, if the theory of inelastic scattering competition is fully capable of explaining these dips. In the cross-section of 232 Th (shown in Fig. 3), for example, the first peak falls by nearly a factor of 2 with an energy increase of only 80 keV. Assuming only about 15 levels below about 1.6 MeV in 232 Th available for inelastic scattering, we calculate that the level density in this nucleus would suddenly have to increase to at least a few thousand per MeV at 1.6 MeV to provide sufficient inelastic scattering to explain this fall. Now a sudden increase in the level density at about this energy is certainly possible; it is associated with the top of the energy gap in the pairing correlation theory of nuclear structure. A calculated level density curve (by Kluge [29]) from this theory is shown in the Fig. 4. However this calculation indicates that the density above the energy gap is only a few hundred per MeV. Furthermore, the required degree of inelastic scattering should also have its effect on the radiative capture cross-section, but the rather sparse data on this [30], indicate that it falls by less than a factor of two between 1.5 MeV and 2.0 MeV. Yet again, the available fission channels are not expected to appear with a density comparable to that of the inelastic scattering channels until a neutron energy of nearly 4 MeV is reached. One wonders, then, why the fission cross-section does not show an overall decline from 1.5 to 4 MeV.

Our doubts about the inelastic scattering competition theory are further intensified when we turn to the fission cross-section of 2380 (Fig. 5). This is a rather similar nucleus to 232 Th and the fission threshold is very close. Yet apart from some breaks in the sub-threshold region, its cross-section is very smooth.

But the greatest difficulty that the competition theory has to face is provided by the fission cross-section of 230 Th (Fig. 6). This exhibits



FIG. 6. The neutron-induced fission cross-section of ²³⁰Th.



FIG. 7. Fission cross-section calculated for a $K^{\pi} = 7/2^{-1}$ channel with threshold energy $E_f = 700$ keV, Hill-Wheeler tunnelling parameter $\hbar \omega_f = 100$ keV, and inelastic scattering competition.

a narrow peak [31] in what is commonly called the sub-threshold region rising to about 50 mb and then falling to about one quarter of this value. The energy at which this occurs is only 700 keV so there is no chance at all of invoking the large onset of inelastic scattering channels at the top of the energy gap to explain it. In fact the most favourable assumptions about the states known to exist in 230 Th will only allow a small drop in the fission cross-section, as shown in Fig. 7. In this calculation the fission channel is assumed to have spin projection (on the symmetry axis) and parity of $K^{\pi} = \frac{1}{2^{\pi}}$; this, and the higher spin members of its rotational band, would allow the fission of compound nucleus levels excited only by neutron f-waves and waves of higher odd orbital angular momentum. A smaller choice of K would allow states excited by lower orbital angular momenta to fission and a much higher fission cross-section at the plateau would be expected. This in itself is a difficulty: measurements of the angular distribution of fission products (due to Vorotnikov et al, [32] indicate that a K = 1/2 channel is open, so the observed peak in the cross-section really is a subthreshold effect and is not just the contribution from a partial wave below the neutron centrifugal potential barrier.

4.2 Possibility of Structure in Fission Channel Strength Functions

A possible weak point at the root of a quantitative channel theory of fission may be the estimate of channel fission width, both in magnitude and in barrier penetration. The estimate that $\overline{\mathbf{v}}_{f}/D = 1/2\pi$ for a single open fission channel really comes from the classical limit of quantal statistical mechanics. Furthermore, perhaps the penetrability formula of Hill and Wheeler leaves room for improvement. This latter visualises the motion of the fissioning nucleus over the barrier as a wave progressing from an infinitely low value of the deformation parameter, β , of the nucleus through an inverted harmonic oscillator potential towards an indefinitely high value (see Fig. 8).

Now we know that the potential energy of an actual transuranic nucleus as a function of the deformation parameter becomes very high for



NUCLEAR POTENTIAL ENERGY AS FUNCTION OF DEFORMATION



FIG. 8. Above: Hill-Wheeler fission barrier model.

Below: more realistic picture of behaviour of potential energy as a function of deformation.



FIG. 9. Complex potential model used for fission strength function calculations.

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small negative values of β , reaches a minimum at a moderate positive value, corresponding to the permanent stable deformation of the nucleus in its ground state, rises through a maximum (the saddle point) then falls towards scission. In describing this potential curve we think of all the other coordinates of the nucleus as being lumped together to provide some intrinsic excitation of the nucleus, or collective excitation in other modes. This picture of the deformation potential suggests that we obtain the fission strength function by a calculation in which a wave in the deformation parameter is scattered off this potential with partial absorption inside the well. The absorption, which represents the coupling of the transition state to the compound nucleus wave function, is provided, as usual, by an imaginary component in the potential energy. Thus we have a complex potential model for the fission strength, but it is vital to remember that this model describes a particular collective motion of the nucleus corresponding to a particular Bohr fission channel, and is not intended to describe an inverse particle reaction such as heavy ion bombardment.

In order to think of such a collective motion beyond the solssion point, we would have to consider a superposition of the wave function of all the fission product pairs in all their different states of excitation and relative angular momenta, and all such wave functions would have coefficients and relative phases governed by the nature of the transition state.

Some preliminary calculations have been made with such a model. For computational simplicity the potential well has the form shown in Figure 9. For values of the deformation parameter greater than zero the real part of the potential energy is given by the expression

$$V = \frac{V_c}{1 + \exp\left[\left(\beta - \beta_c\right)/d_c\right]} + \frac{V_e}{1 + \exp\left[-\left(\beta - \beta_e\right)/d_e\right]}$$
(4)

The values of β_c , d_c , β_e and d_e are suggested by the liquid drop model. The value of V_e is suggested by the observed release of kinetic energy in low energy fission (i.e. about 180 MeV) and V_e is obtained from the known energy of the transition state E_F with respect to the nuclear ground state; typical values for this are of the order of 6 MeV. The mass parameter associated with the deformation is not a well-known quantity and probably varies with the degree of deformation. For small deformation the path towards fission is governed primarily by the quadrupole parameter β_2 , and the mass parameter associated with this is given by hydrodynamical theory [34] as

$$B_2 = \frac{3 \text{ m A R}_o^2}{8 \pi}$$
(5)

where m is the nucleon mass, A the nuclear mass number and $R_{\rm C}$ is the nuclear radius.

Substitution of typical nuclear quantities in this expression gives a mass parameter B_2 of the order of 4.5 x 10^{-47} gm.cm².

The wave function of such a potential is computed by numerical integration of the Schrödinger equation starting from a regular function within the well. At a point where the external potential has become



FIG.10. Strong coupling calculation of fission strength function for $V_c = -5.4$ MeV, $\beta_c = 0.42$, $d_c = 0.06$, $\beta_e = 3.0$, $d_e = 0.2$, $V_e = -180$ MeV, W = -1.0 MeV (full curve). The dashed curve is the Hill-Wheeler fission strength function for $\hbar\omega_f = 0.38$ MeV.

asymptotically constant the wave function is matched to the external wave, which is the sum of an ingoing wave and the collision function multiplied by an outgoing wave. From this matching the collision function \overline{U} is determined. This is the quantity required from the model as representing the average collision function of the nuclear problem. The microscopic collision function has the form [9]:

$$U_{cc} = \delta_{cc} + k_c \mathcal{P}_c Q_{cc} - i \sum_{m} \frac{k_c \mathcal{P}_c G_{m(c)}^2 \exp(2i\xi_{m(c)})}{E - E_m + \frac{1}{2}i\Gamma_m}$$

The average over many resonances of this expression is

Ū

$$\overline{J}_{cc} = e^{i\phi} \left[1 - \pi \frac{k_c \mathcal{P}}{\overline{D}} \overline{G}_{(c)}^2 \right]$$

in the case $\overline{\Gamma} << \overline{D}$ (see Appendix 2). Using this expression the fission strength function is obtained from the model.

The results of a typical "strong-coupling" calculation are shown in Fig. 10. Here the mass parameter B is 1.9×10^{-47} gm om² and the imaginary

(7)



FIG.11 Weak coupling calculation of fission strength function for W = -0.03 MeV (other parameters as in Fig.10).

part of the potential (cutting off at $\beta = 0.9$) is W = -1.0 MeV. The curve of fission strength function against energy about the channel threshold recalls the Hill-Wheeler strength function (shown as a broken curve) but is shifted upwards by about 100 keV and rises above the classical value of $1/2\pi$ at high energies. The sub-threshold slope of the curve corresponds to a Hill-Wheeler tunnelling parameter $K\omega_{\rm f} = 0.38$ MeV.

The intriguing features of the model are found in the "weak coupling" calculations. In the result of Fig. 11, W is only -30 keV. A rather sharp peak is found in the fission strength function very close to the threshold energy. One is tempted to hypothesize that this may be the explanation of the peaks in the fission cross-section of 232Th. The peak corresponds to a virtual state in the (real) potential well of the model. With the kinds of parameters used the spacing of such states is of the order of 2 or 3 MeV. Fairly small changes in the parameters will move the virtual states to different energies with respect to threshold. If V_e is changed from -5.4 MeV to -6.0 MeV the virtual state moves from just above threshold to a binding energy of 130 keV. The fission strength function then has the form shown in Fig. 12 and could provide the explanation of the sub-threshold fission strength function of ²³⁰Th. The occurrence of the virtual state at an energy considerably above threshold would give a much damped peak owing to the large partial width associated with the motion in the deformation parameter (see Fig. 13). Thus the model seems to have the property of being able to explain the variety of structure observed in fission cross-sections ranging from the fully-developed peaks in the $^{232}Th(n,f)$ cross-section, through the sub-threshold



FIG.12. Weak coupling calculation of fission strength function with deeper well, V_c = -6.0 MeV (other parameters as in Fig.11).



FIG.13. Weak coupling calculation with virtual state at positive energy (V_c = -3.6 MeV).

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peaks in 230 Th(n,f) to the rather featureless cross-section of 238 U(n,f). This hypothesis is, of course, only a phenomenological model for what appears to be happening in these reactions. The explanation at a deeper level would lie in answering why the imaginary component is so low. This question has not yet been completely satisfactorily answered in the related problem of nuclear scattering by nuclei, but at least existing theory suggests that there is no reason for expecting W in this case to be the same as in the nuclear scattering case. There are other examples of elementary types of excitation being only weakly coupled to the compound nucleus. The outstanding example is the isobaric analogue state which has been found to be spread over energy intervals as small as 10 keV even at excitation energies greater than 10 MeV.

5. CONCLUSION

The considerations of this paper indicate that extrapolation of existing data to unmeasured regions using nuclear theory may be somewhat uncertain. In spite of their success in explaining many data, it is to be stressed that the fission channel schemes of even compound nuclei presented in the earlier part of the paper as possible bases for such extrapolation may require reconsideration in the light of the model discussed in Section 4b. It is hoped that these discussions will stimulate even more careful measurements in this subject.

APPENDIX I

Calculation of average cross-sections

Perhaps the formally simplest many-level cross-section expression is the S-matrix version of Humblet and Rosenfeld [9]. The cross-section for the process (α,β) is

$$\sigma_{\alpha\beta} = \frac{\Pi}{k_{\alpha}^{2}} \left| \left(Q_{\alpha\beta} - i \sum_{m} \frac{G_{m(\alpha)} G_{m(\beta)} e^{i \left(\xi_{m(\alpha)} + \xi_{m(\beta)} \right)}}{E - E_{m} + \frac{1}{2} i \Gamma_{m}} \right) \sqrt{k_{\alpha} k_{\beta} \mathcal{P}_{\alpha} \mathcal{P}_{\beta}} \right|^{2}$$
(A.1)

where $Q_{\alpha\beta}$ is a slowly varying background term, the k_{α} are wave numbers in the channels α , and the \mathcal{P}_{α} are threshold factors depending on centrifugal and Coulomb potentials. Poles of the S-matrix occur at complex energies $\mathbb{E}_m - \frac{1}{2}tT_m$ corresponding to the energies of decaying states of the system. The residues of the poles factorise into components $G_{m(\alpha)} \exp(i\xi_{m(\alpha)})$ which give rise to the partial widths of resonance terms in the cross section. The averaging of this expression over energy is readily achieved using contour integration. On ignoring the background term $Q_{\alpha\beta}$ which is expected to be small in the typical compound nucleus region the average cross-section over an energy interval ε containing many resonances is

$$\frac{1}{\varepsilon} \int_{\varepsilon} dE \sigma_{\alpha\beta} \approx \frac{\pi^{2}}{\varepsilon} \frac{k_{\beta}}{k_{\alpha}} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \left\{ \frac{(\Gamma_{m} + \Gamma_{m'}) G_{m(\alpha)} G_{m(\beta)} G_{m'(\beta)} G_{m'(\beta)} G_{m'(\beta)} G_{m'(\beta)}}{(E_{m'} - E_{m})^{2} + \frac{1}{4} (\Gamma_{m} + \Gamma_{m'})^{2}} - \frac{(E_{m'} - E_{m}) G_{m(\alpha)} G_{m(\beta)} G_{m'(\beta)} G_{m'(\beta)} G_{m'(\beta)}}{(E_{m'} - E_{m})^{2} + \frac{1}{4} (\Gamma_{m} + \Gamma_{m'})^{2}} \right\}$$

$$(A.2)$$

where $\overline{\Phi}_{mm'} = \xi_{m(m)} - \xi_{m'(m)} + \xi_{m(S)} - \xi_{m'(n)}$. Now, if it is assumed that the phases $\overline{\Phi}_{mm'}$ (which is zero for m = m')are randomly distributed around the unit circle for all $m \neq m'$, or alternatively have similar distributions about zero and π , then the average cross-section reduces statistically to the single sum

$$\left\langle \sigma_{\alpha\beta} \right\rangle = \frac{\pi^{2}}{\varepsilon} \frac{k_{\beta}}{k_{\alpha}} \frac{\Gamma_{\alpha}}{\Gamma_{\beta}} \sum_{m} \frac{2 \ G_{m}(\alpha)}{\Gamma_{m}} \frac{G_{m}(\beta)}{\Gamma_{m}}$$

$$\approx 2\pi^{2} \frac{k_{\beta}}{k_{\alpha}} \frac{1}{\overline{D}} \left\langle \stackrel{P}{\sigma_{\alpha}} \frac{G_{m}(\alpha) R_{\beta}}{\Gamma_{m}} \frac{G_{m}(\beta)}{\Gamma_{m}} \right\rangle$$
(A.3)

However, these phases cannot be assumed to be random. This is because Humblet and Rosenfeld's version of the S-matrix theory is not automatically unitary in form. If unitarity is imposed upon the S-matrix, correlations are introduced amongst the resonance parameters. A particular example of this is the imposition of unitarity on the two-level S-matrix without background. It is found that the following condition is imposed on the pole residues if the energy variation of the threshold term is ignored

$$G_{I(\alpha)}G_{I(\beta)}\sin(\xi_{I(\alpha)} + \xi_{I(\beta)}) = -G_{2(\alpha)}G_{2(\beta)}\sin(\xi_{2(\alpha)} + \xi_{2(\beta)})$$
(A.4)

This clearly imposes a strong correlation on the channel-pole phase factors, and since the latter depart increasingly from the narrow resonance limits 0 and π as the width to spacing ratio increases, the contributions from the $m \neq m'$ terms in Eq. (A.2) becomes increasingly important as $\overline{\Gamma}/\overline{D}$ becomes large. In addition, the distribution of the $G^2_{m(d)}$ etc. and the resonance spacings depart from the known statistics of the corresponding quantities (reduced widths and spacings) of R-matrix theory in the same limit; these statistics are invariant to things like barrier penetrability because the basic quantities of R-matrix theory are the eigensolutions of the Schrödinger equation within an internal region constricted only by real boundary conditions. The quasi-resonances [4] provide an example of how the statistics are distorted; the S-matrix expansion of the two-level interference results in a pole spacing smaller than the level spacing and widths respectively less than and greater than the two R-matrix level widths. Unfortunately these correlations and statistics of the S-matrix parameters are not nearly well enough established at the present time to allow calculations of the average cross-sections on the basis of Eq. (A.2).

Appendix 2

The average collision function

A similar problem appears in the application of S-matrix theory to the interpretation of a model average collision function. The diagonal S-matrix element has the form

$$S_{\alpha\alpha} = U_{\alpha\alpha} - \delta_{\alpha\alpha}$$

= $k_{\alpha} \mathcal{P}_{\alpha} \left\{ Q_{\alpha\alpha} - i \sum_{m} \frac{G_{m}(\alpha) \exp(2i\xi_{m}(\alpha))}{E - E_{m} + \frac{1}{2}iT_{m}} \right\}$
(B.1)

From this the average collision function is found to be

$$\overline{U}_{xx} = S_{xx} + k_{x} \partial_{x} \left[Q_{xx} - \underline{T} \quad \underline{G}_{m(x)}^{2} \cos 2 \underline{S}_{m(x)} - i \underline{T} \quad \underline{G}_{m(x)}^{2} \sin 2 \underline{S}_{m(x)} \right]$$

$$\overline{D} \qquad \overline{D} \qquad \overline{D} \qquad (B.2)$$

The application of unitarity to the background function together with the assumption that it is diagonal reveals that the term $\delta_{wx} + k_{x} \mathcal{R}_{x} \mathcal{Q}_{wx}$ has modulus unity. If its phase angle is ϕ' , say, then \overline{U}_{GG} can be rewritten

$$\begin{split} \overline{U}_{\text{AXX}} &= e^{i\left(\phi + \phi'\right)} / \left[1 - \frac{2\pi k_{x} \partial_{x} \overline{G_{m(x)}} \cos 2\xi_{m(x)}}{\overline{D}} \cos \phi - \frac{2\pi k_{x} \partial_{x} \overline{G_{m(x)}} \sin 2\xi_{m(x)}}{\overline{D}} + \frac{\pi^{2} k_{x}^{2} \partial_{x}^{2} \left(\overline{G_{m(x)}} \cos 2\xi_{m(x)}\right)^{2}}{\overline{D}^{2}} + \frac{\pi^{2} k_{x}^{2} \partial_{x}^{2} \left(\overline{G_{m(x)}} \sin 2\xi_{m(x)}}{\overline{D}^{2}}\right)^{2} \right] \end{split}$$
(B.3)

where ϕ' is the phase angle due to the remaining part of $\overline{U}_{\alpha\alpha}$. Now if interference amongst the resonance terms of the S-matrix can be ignored it can be shown by imposition of the unitarity condition that $2\xi_{m(\alpha)} = \phi$ and the collision function reduces to

$$\overline{U}_{xx} \approx e^{i(\phi + \phi')} / \left[1 - 2\pi k_{x} \frac{g_{x}}{\overline{D}} \frac{\overline{G}_{m(x)}^{2}}{\overline{D}} + \pi^{2} k_{x}^{2} \frac{g_{x}^{2}}{\overline{D}^{2}} \frac{\overline{G}_{m(x)}^{2}}{\overline{D}^{2}} \right]$$

$$= e^{i(\phi + \phi')} \left[1 - \pi k_{xx} \frac{g_{x}}{\overline{D}} \right] \qquad (B.4)$$

which is the result quoted in the text. Neglect of resonance interference can only be justified, however, when $\Gamma << D$, and this condition is therefore essential for the validity of Eq. (B.4). If resonance interference becomes dominant then the <u>unitarity correlation</u> expressed in Eq. (A.4) suggests that the term $G_{m_{ext}}^{\infty} \sin 2\xi_{m(x)}$ in Eq. (B.3) is negligible. It then becomes clear that Eq. (B.4) expresses a lower limit for $|U_{\alpha\alpha}|$ i.e.

$$\frac{\pi k_{x} P_{x}}{\overline{D}} \frac{\overline{G_{m(x)}^{2}}}{\overline{D}} > 1 - |\overline{U}_{xx}|$$

(B.5)

The right hand side of inequality (B.5) gives a lower limit for the fission strength function calculated from our complex potential model.

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DISCUSSION

H. NIFENECKER: Comparison between the ²³³U cross-section curve and the curves simulated by a Monte Carlo method shows that about one resonance in two is not observable, and this is particularly the case for resonances of small neutron width; a value of 3 is thus obtained for $2\pi \Gamma_f/D$ per spin state. The analyses that we have performed have always employed single-level formalism and have given satisfactory results; could Dr. Lynn say how it would be possible to reveal the quasi-resonances?

J.E. LYNN: The cross-sections of the odd-mass fissionable nuclei are probably much too complex, owing to the presence of resonances of two spin sequences, for it to be possible to say with certainty that any given peak in the cross-section is, or is not, a quasi-resonance. It is more profitable to look at the even target nuclei for a suitable candidate. The most fissile of these appears to be 232 U, and with some luck it may be possible to identify as a quasi-resonance a single maximum by looking for the broad component (due to the other pole) underlying the sharper peak. The chances of doing this really depend on just how fissile 232 U is,

A+

i.e. whether the value of $2\pi \overline{\Gamma}_{\rm f}/\overline{D}$ is large enough to give appreciable probability of level overlap.

A. T.G. FERGUSON: Dr. Lynn has postulated certain collective kinds of levels at the nuclear saddle point. A characteristic of such collective levels, as found in the stable deformed nuclei, is that they occur at excitations that change only slowly from nucleus to nucleus. Has he examined the saddle-point collective levels to see if they show a similar systematic behaviour?

J.E. LYNN: It is also characteristic of such collective levels that they are expected to vary their energy with respect to the "ground" state potential energy curve as the deformation changes. Since the saddle-point deformation varies from nucleus to nucleus, any systematic behaviour of the kind Dr. Ferguson is pointing out may not be very apparent. In any case, in choosing the channel schemes I did, I tended to be guided by what one may expect for the energy behaviour of these collective states. This behaviour is discussed by Wheeler in his article in Fast Neutron Physics.

A. MICHAUDON: I should like to ask Dr. Lynn a question regarding the first part of the paper, i.e. the analysis of cross-sections in the resonance region. You have done "simulated" cross-section calculations using a Monte Carlo method and taking account of interference between these resonances due to fission. What proportion of quasi-resonances have you observed in the cross-sections calculated for the main fissile nuclei, ²³³U. ²³⁵U. ²³⁹Pu. ²⁴¹Pu?

J.E. LYNN: For ²³⁵U, with the channel scheme of Table I, there is a 5% probability, roughly, of a neighbouring pair of 3⁻ levels giving rise to a quasi-resonance, and a considerably smaller probability in the case of the 4⁻ resonances. The probability that two resonances of different spin will overlap is about 10%. With the channel scheme of Table II the quasiresonance probability is nearer 10% for the 3⁻ levels and 3% for the 4⁻ levels. In the case of ²³³U, the probability of occurrence of quasiresonances is about 19% for the 2⁺ levels and 10% for the 3⁺ levels, using the channel scheme of Table IV, and the corresponding figures for ²⁴¹Pu are around 13% and 7% respectively. For ²³⁹Pu the probability of 1⁺ quasi-resonance formation is probably negligible, but is perhaps about 7% for the 0⁺ levels.

C.D. BOWMAN: I should like to mention that we have measured the 232 U fission cross-section using the spark chamber and find a resonance structure which is similar to that for 239 Pu. We were able to detect up to 4000 events per channel in the peaks of the resonances and we measured peak-to-valley ratios of almost 10^3 . I hope we can get a meaningful fit using the multi-level formalism.

MEASUREMENTS OF ETA, ALPHA AND NEUTRON CROSS-SECTIONS FOR ²³⁹Pu ON THE HARWELL NEUTRON TIME-OF-FLIGHT SPECTROMETER

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Abstract

MEASUREMENTS OF ETA, ALPHA AND NEUTRON CROSS-SECTIONS FOR ²³⁹ Pu ON THE HARWELL NEUTRON TIME-OF-FLIGHT SPECTROMETER. Eta values and total and fission cross-sections have been measured using the Harwell 45-MeV electron linear accelerator neutron time-of-flight spectrometer. The 34.9-m flight path, with a resolution of 7.2 ns/m, was used for the energy range 10 eV to 1 keV and the 97.5-m flight path, with a resolution of 2.5 ns/m, was used for the energy range from 50 eV to 30 keV. The experimental methods employed were similar to those of Brooks et al., in which the fission neutron yield and the transmission of a number of samples of different thicknesses are measured together with the shape of the incident neutron spectrum, all the measurements being made with the same resolution.

In the yield measurements, fast neutrons from fission events were detected in liquid scintillators, and pulse-shape discrimination was used to reject events caused by gamma rays. The transmission measurements were performed with a lithium glass scintillator, and a 1/v detector was used to measure the spectrum. Backgrounds were measured using the "black resonances" technique.

The measurements at 34.9 m have been normalized in the region of 10 eV to the eta values obtained by Brooks et al. and the eta values from the 97.5-m measurements have been normalized to the 34.9-m measurements in the region of 50 eV. The results have been corrected for multiple scattering in the samples.

Capture cross-sections have been derived from the total and fission cross-sections by assuming values for the scattering cross-section. A direct measurement of alpha (the ratio of the capture-to-fission cross-sections) is in progress over the energy range 10 eV to 30 keV. This will lead to a better estimate of the capture cross-section since alpha measurements are not so sensitive to the magnitude of the scattering cross-section.

1. INTRODUCTION

The increasing interest in fast breeder reactors has led to a requirement for accurate, high resolution measurements of the total and partial cross-sections and related quantities (e.g. alpha, the ratio of capture and fission cross-sections) of ²³⁹Pu over an energy range extending far into the kilovolt region. The fission cross-section has been measured by a number of workers but the recent experiments of James [1] and de Saussure et al. [2] using gas scintillation counters and Shunk et al. [3] using solid-state fission fragment detectors and the Petrel nuclear explosion as a source of neutrons are the first to attempt high resolution measurements in the eV and keV regions. There are no high resolution measurements of eta, the number of neutrons produced per neutron absorbed, and alpha above 11 eV and data of any description on the capture cross-section are very scarce.

To obtain high resolution using time-of-flight techniques, it is necessary to use long flight paths. Using an accelerator to produce neutrons, the number incident on any reasonable size of sample at such distances becomes small and in practice background conditions make it impossible to use thin samples. Hence thick samples must be used and then the only method of measuring the fission cross-section is to detect the neutrons emitted in the fission process. This immediately raises a difficulty in the calculation of the fission cross-section because ingeneral the multiple scattering of the incident neutrons cannot be neglected except where the sample is "thin" (n $\sigma \ll 1$). However, if measurements of fission neutron yield and neutron transmission are made with identical energy resolution values of eta can be obtained which, for a "thick" sample $(n\sigma \gg 1)$, require negligible correction for multiple scattering. Therefore, if both measurements are made for samples of different thickness, the resulting data must be weight-averaged so that the results obtained from the thinnest samples have the highest weight for the fission crosssection and the lowest weight for eta. To normalize the results of the experiment it is necessary to do so in a region of high cross-section where the statistical errors are low and the data from previous experiments are best known. Since all samples are relatively thick in these energy regions the errors on the results are lowest if they are normalized through eta which is related to the fission cross-section by the formula:

$$\eta = \frac{\overline{\nu}_{\rm p} \sigma_{\rm f}}{\sigma_{\rm T} - \sigma_{\rm S}} \tag{1}$$

where \overline{v}_{p} = average number of prompt neutrons emitted per fission

 $\sigma_{\rm f}$ = fission cross-section

 $\sigma_{\rm T}$ = total cross-section

 σ_{s} = scattering cross-section

A measurement of eta and σ_f requires the measurement of the incident neutron spectrum in addition to the fission neutron yield and the transmission. Once we have calculated σ_T from the transmission data we can deduce the capture cross-section from the equation:

$$\sigma_{\rm T} = \sigma_{\rm f} + \sigma_{\rm c} + \sigma_{\rm S} \tag{2}$$

We can also calculate alpha from the relation

$$\alpha = \frac{\overline{\nu}_{\rm p}}{\eta} - 1 \tag{3}$$

In this paper preliminary results of σ_T and σ_f only will be presented.

2. EXPERIMENTAL METHOD

Two measurements of eta and the fission cross-section of ²³⁹Pu, covering the energy regions 10 eV to 2 keV and 50 eV to 30 keV, have been made on the 45-MeV Harwell Linear Accelerator time-of-flight spectrometer. The experiments will be referred to as the lower energy experiment and the higher energy experiment, respectively. In the experiments, fast fission neutrons from thick samples were detected by liquid scintillation counters and pulse shape discrimination was used to reject γ -rays resulting

	Lower energy experiment	Higher energy experiment	
Energy range	10 eV-2 keV	50 eV-30 keV	
Neutron pulse width (unmoderated)	220 ns	220 ns	
Timing channel width	125 ns	125 ns	
Flight path length	34. 9 m	97. 55 m	
Resolution	7.2 ns/m	2.5 ns/m	
Permanent "black" filter in beam	Sodium	Aluminium	
Detectors used to measure spectrum	BF ₃ 10 eV-100 eV ⁶ Liglass 100 eV-2 keV	⁶ Li glass 50 eV-30 keV ¹⁰ B plug 50 eV-30 keV	

TABLE I. EXPERIMENTAL DETAILS

from neutron capture and spontaneous decay of the samples. The experimental conditions for the two measurements are shown in Table I. Backgrounds were measured by the use of resonance filters. In each experiment the fission neutron yield, the transmission and the incident neutron spectrum were measured at the same resolution. Three 239 Pu samples 2.9 in. in diam. were used in the yield and transmission measurements with n-values of 0.02433, 0.00440 and 0.00115 atoms/barn. The two thickest samples contained 1.8% of 240 Pu and the thinnest 3.2%. For safety reasons, the 239 Pu samples were contained in aluminium cylinders 3 in. in diam. and 12 in. long, the samples being fixed at the centre to minimize the effect of neutrons scattering in the ends. Lithium-loaded glass scintillators were used in the transmission measurements and Table I shows the detectors used to measure the spectrum in each experiment.

The time-of-flight of each event was recorded on a l-in. magnetic tape recorder fitted with a five-word input buffer so that several events could be recorded per timing cycle. The number of time channels used was 8192. A PDP-4 computer was used to analyse the events into time spectra. The calculation of the cross-sections was done on an IBM 7030 computer using the programme originally written by Brooks [4] which had been extensively modified.

3. THE CALCULATION OF THE TOTAL CROSS-SECTION

The total cross-section was calculated from the transmission data of the three samples making allowances for the aluminium container and the

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impurities in the samples. Since the samples were not thin there was appreciable self-screening in the stronger resonances. In the absence of perfect resolution, this causes an error in the calculated cross-section. In an attempt to reduce errors caused by this effect, the minimum transmission acceptable for the calculation of the cross-section was reduced in steps as the energy and the resolution broadening increased. In this way, the thinner samples only were used in calculating the peaks of the crosssection. The total cross-section and error were calculated for each sample at each channel and the average found by weighting the value for each sample according to its error.

4. THE CALCULATION OF ETA AND FISSION CROSS-SECTION

The yield of fission neutrons in timing channel i is given by

$$Y_{i} = N_{i}\beta \left[A_{i}\overline{\nu}_{p}(D_{i} + H_{i} + L_{i}) + \overline{\nu}_{p}C_{i}\right]$$
(4)

where N_i = number of incident neutrons

 β = efficiency of the fission neutron detector

A_i = fraction of incident neutrons which interact in the sample

 D_i = probability of interaction being a direct fission

- H_i = probability of scattering from high Z materials in the sample, e.g. ²³⁹Pu and other fissile or fertile materials, and then causing fission
- L_i = probability of scattering from low Z impurities in the sample and then causing fission
- C_i = probability of scattering from the ends of the can and then causing fission

Now eta is given by

$$\eta_i = \frac{D_i \overline{v_p}}{I_i}$$
(5)

where I_i = fraction of incident neutrons absorbed in the ²³⁹Pu. Therefore

$$\eta_{i} = \frac{kY_{i}}{N_{i}A_{i}I_{i}} - \frac{\overline{\nu}_{p}}{I_{i}} \left[\frac{C_{i}}{A_{i}} + H_{i} + L_{i} \right]$$
(6)

with β replaced by a normalizing factor k.

The fission cross-section σ_f is then calculated from Eq.(1).

The most serious problem encountered when calculating the values of eta and σ_f at each energy is that of multiple scattering of the incident neutrons. We have assumed that in these measurements the scattering is elastic. In calculating the correction due to scattering the effect of only one scatter is taken into account. The correction is done starting at the low energy end of the range of energies used in the experiment and working channel by channel towards the high energy end. All scatters are assumed to take place on the axis of the sample at a depth equal to that penetrated by half of the neutrons which interact at the incident energy. At each timing channel the correction is calculated as follows. The energy of the next lower energy channel is found and the angle through which the neutron would have to scatter to have that energy is calculated. The probability of the scattered neutron subsequently producing a fission is calculated taking the finite size of the sample into account and using the data which have already been corrected. This probability is calculated for those channels corresponding to the range of possible energy loss and then averaged. The average probability is then multiplied by the probability of the neutron scattering once to give the correction factor.

5. THE SCATTERING CROSS-SECTION USED IN THE CALCULATIONS

The following considerations were made in deciding what to use for the scattering cross-section at each energy. The scattering cross-section is made up of two parts, the potential scattering and the resonant scattering. In ²³⁹Pu the ratio of the neutron width Γ_n to the total width Γ of the resonances is often as much as 20% and it is clear that the resonant scattering term cannot be neglected. However, since the scattering cross-section has not been measured over the energy range under consideration it must be found by other means. Up to 300 eV the neutron and total widths of many of the resonances have been measured [5, 6] and we have used these data to construct a scattering cross-section σ_{S_i} for timing channel i from the relation

$$\sigma_{S_i} = \left(\sigma_{T_i} - \sigma_{pot}\right) \frac{\Gamma_{n_i}}{\Gamma_i} + \sigma_{pot}$$

where σ_{T_i} = total cross-section for channel i σ_{pot} = potential scattering cross-section Γ_{n_i} = neutron width of the resonance appropriate to channel i Γ_i = total width of the resonance appropriate to channel i

This, of course is a very rough estimate of the scattering cross-section since it is only a single-level description and neglects interference.

Above 300 eV we have divided the energy range into 19 energy intervals and used average values. The total width Γ is given by

$$\Gamma = \Gamma_{\rm n} + \Gamma_{\rm f} + \Gamma_{\rm y} \tag{8}$$

where Γ_n , Γ_f and Γ_γ are the neutron, fission and radiation widths respectively. Now, averaged over many resonances Γ_f is reasonably constant as a function of energy. Γ_γ is also constant but average values of Γ_n show an energy dependence proportional to \sqrt{E} . Thus for Γ_f and Γ_γ in each interval above 300 eV we have used the average of the values below 300 eV. For Γ_n we have taken the average below 300 eV suitably weighted for the different spin states and modified this in each interval to account for the energy dependence. The total width in each interval was then found from the sum of the partial widths. The potential scattering cross-section has been measured by Uttley [7] and is essentially constant at 10.3 \pm 0.5 barns up to 30 keV.

(7)



FIG.1. Total cross-section of ²³⁹Pu

6. SPECTRUM MEASUREMENTS

The measurements of the spectrum on each flight path were initially done with a thin lithium glass scintillator. In estimating the efficiency of the detector it was assumed that the ${}^{6}Li(n, \alpha)$ cross-section varies as $1\sqrt{E}$ up to 30 keV and corrections for multiple scattering of the neutrons were made using a Monte Carlo programme [8]. When the spectra were examined they were found to disagree above 1 keV with previous measurements using detectors based on the ${}^{10}B(n, \alpha)$ reaction on other flight paths. It was thought that the discrepancy might be due to the $^{6}Li(n, \alpha)$ reaction deviating from a $1\sqrt{E}$ dependence. To test this, the spectra were remeasured using a BF_3 counter on the 34.9-m flight path and a thin ¹⁰B sample, with NaI counters to detect the γ -ray emitted in the reaction $^{10}B(n, \alpha \gamma)$, on the 97.5-m flight path. The spectra determined by these techniques agreed up to ≈ 25 keV with the first measurement to within an accuracy of \pm 5%. Thus, if the ¹⁰B(n, α) cross-section is proportional to $1\sqrt{E}$ up to 25 keV, we can say that the ⁶Li(n, α) cross-section has the same energy variation within \pm 5%. Although we are now satisfied that we have measured the correct spectrum shape the original discrepancy remains and it is now thought that it may be due to a difference in each flight path geometry with respect to the "booster" target.



FIG.2. Fission cross-section of ²³⁹Pu. The dotted lines indicate regions where the cross-section is too low to be measured.

7. NORMALIZATION OF ETA

The lower energy experiment has been normalized at 10.95 eV to $\eta = 2.041$ as measured by Brooks et al.[9]. The higher energy experiment was normalized to $\eta = 1.492$ at 65.7 eV as found by the lower energy experiment. In the calculations, the average number $\overline{\nu}_{\rm p}$ of prompt neutrons produced per fission was assumed to have the constant value of 2.864.

8. ERRORS

The computer programme used to calculate the final results also produced an estimate of the error on each quantity at each channel. Statistical errors together with an error for the assumed scattering cross-section were taken into account in the calculation of the final channel error. In the resonance region, the statistical error on the number of counts in each timing channel, after the subtraction of background, varied from $\approx 3\%$ on the peaks to $\approx 100\%$ in the valleys. At higher energies, although the crosssection is falling, there are more incident neutrons per channel and again statistical errors of $\approx 3\%$ were achieved.

The source of the largest error in the calculations lies in the correction for multiple scattering of the neutrons. Firstly, only single

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FIG. 3. Fission cross-section of ²³⁹Pu

scatters have been considered and where the correction for scattering is largest this will lead to an error of a few per cent. Secondly, the scattering cross-section used in the calculation is only a very rough estimate of the actual cross-section. The errors on Γ and Γ_n are typically 10-30% and it has been assumed that the error on the resonant part of σ_s is 50%. Eta and σ_f have been calculated using the scattering cross-section as previously described and also assuming that the only contribution to the scattering comes from the potential scattering term. In the worst case, the differences between the values of eta and σ_f obtained by the two calculations were 3% and 7%, respectively. The two scattering cross-sections at that point differ by a factor of eight and the results indicate that a 50% error on the scattering cross-section will introduce errors of less than 2% and 4% in eta and σ_f .

Systematic errors arise from two sources. The subtraction of background is the first and this will have its greatest effect at the low energy end of each experiment. In the yield measurements, most of the background originates from the spontaneous fission of the 240 Pu contaminant and since this is time independent, it can be measured accurately. Only in those valleys where the fission cross-section lies in the range zero to less than 10 b will the error in the background have any appreciable effect. In other regions, background subtraction should result in an error of < 3%.

The processes of normalization may also produce a systematic error. The low energy experiment is normalized to the results of an experiment which itself was normalized at thermal energy. The higher energy experiment has been normalized in a region where the error on eta is 3%.





FIG. 4. Fission cross-section of ²³⁹Pu

The use of three samples and the weighting of the individual results according to their errors to produce the final result has the effect of reducing the errors due to random sources.

9. RESULTS

Figures 1-4 show provisional σ_f and σ_T data as a function of neutron energy from the two experiments. Of particular interest are the regions of very low cross-section between 20 eV and 40 eV. In Fig.2 these are indicated by the dotted lines. In these regions the fission cross-section is too low to be measured. An upper limit of about 2 b can be placed on the cross-section since, as can be seen in Fig.1, the total cross-section in these regions becomes almost equal to the potential scattering crosssection of 10.3 b. The regions of almost zero fission cross-section agree well with those found by Shunk et al.[3]. In Figs.1-4, every tenth point shows the error, as calculated by the programme, resulting from statistics and the scattering cross-section error.

Values of alpha have been derived from averages of the capture and fission cross-sections. Present results indicate that alpha is high up to 10 keV and then falls off rapidly to about 0.5 at 25 keV. However, deductions of alpha from these data may be greatly in error if the scattering cross-section does not behave as we have assumed in section 5. In particular, at about 600 eV there is a break in the curve of average fission cross-section as a function of energy. The changes in the fission crosssection may be reflected in changes in the other partial cross-sections

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and therefore any deduced values of alpha may well be in error in this energy region.

The analysis of these data is continuing and the corrections are being studied in greater detail. One of the advantages of measuring σ_T and σ_f with identical energy resolution is that the capture cross-section σ_c can be deduced from Eq.(2) assuming a knowledge of the scattering cross-section. This technique has been used satisfactorily by Brooks et al. [10] for ²³⁵U. However in ²³⁵U the ratios of the neutron to total width of the resonances are on average very small and hence the resonant scattering contribution to σ_s is also small and can be neglected. In the case of ²³⁹Pu the neutron widths are much larger and the resonant scattering contribution becomes important. Thus the capture cross-section deduced from Eq.(2) will be subject to the errors introduced by the assumptions made in section 5.

10. DIRECT MEASUREMENT OF ALPHA

To measure the value of α , the ratio of the capture-to-fission crosssections, for ²³⁹Pu an experiment is in progress on the 35-m flight path. The detection system consists of liquid scintillation counters, which are used to detect the fast neutrons produced by fission events by using the pulse-shape discrimination technique, and Moxon-Rae counters which detect the γ -rays produced by the fission and capture events. If N_n and N_{γ} are the number of counts in the two counters respectively, then

$$N_n = \psi n_f \tag{9}$$

$$N_{\gamma} = \mu n_{\gamma} + \theta n_{f} \tag{10}$$

where n_{γ} and n_{f} are the number of capture and fission events, and ψ , μ and θ are constants. If these equations are combined α can be determined from the following formula

$$\alpha = \phi \frac{N_{\gamma}}{N_{n}} - \omega \tag{11}$$

where ϕ and ω are constants which can be determined by normalization.

This technique has the advantage over the measurements described earlier that for samples of thickness less than 0.001 atoms/barn the corrections required to allow for the multiple scattering of the incident neutron beam in the sample are small.

It is planned to make measurements using this technique over the energy range 10 eV to 30 keV.

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DISCUSSION

J. RYABOV: I should like to mention that similar measurements of radiative capture and fission cross-sections and of alpha have been carried out for 239 Pu using time-of-flight methods with a liquid detector containing Cd. This work was performed on the Joint Institute for Nuclear Research (J.I.N.R.) pulsed fast reactor at Dubna in the neutron energy range 5 eV to 23 keV with a resolution of about 60 ns/m (see Preprint P-2713, J.I.N.R. Dubna).

J.J. SCHMIDT: Your α -measurements in the range 1 to 10 keV on ²³⁹Pu yield values between 1.5 and 2.5 and these are higher than the corresponding KAPL (Knolls Atomic Power Laboratory) values of about 0.5 to 0.6 by a factor of 3 to 4. Comparison of multigroup calculations with measurements of the space dependence of $\overline{\alpha}$ in the EBR-I (Crouthamel et al.) seems to indicate that the α -values used in the calculations based on the above KAPL measurements are somewhat too low, but not by a factor of 3 or 4. I should therefore like to ask whether there is any explanation for the discrepancy between your α -measurements and the KAPL ones.

B.H. PATRICK: The values of alpha deduced from these measurements are subject to the assumptions made in the construction of the scattering cross-section. The average total cross-section falls off fairly smoothly as a function of energy, whereas the fission cross-section has a sharp change in level at $\approx 600 \text{ eV}$. Since we do not measure the capture or scattering directly, we do not know how this break is reflected in them and hence the scattering cross-section we have used may be incorrect. In the values of alpha deduced from this experiment we have assumed that the resonant part of the scattering cross-section is ≈ 2 b in the kilovolt region. An error in this quantity may have a considerable effect on the α -values deduced here.

SECTIONS EFFICACES TOTALE ET DE FISSION DE 237 Np

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Abstract - Résumé

TOTAL AND FISSION CROSS-SECTIONS OF ²³⁷ Np. The total and fission cross-sections of ²³⁷ Np were measured at Saclay by the time-of-flight method. The samples used in transmission were in oxide form, with thicknesses of 0.0066 and 0.002 atoms per barn. With resolutions of 7 ns/m (between 0.5 eV and 25 eV) and 2 ns/m (between 20 eV and 300 eV), the authors were able to detect a number of new resonances below 100 eV and to extend the previously known data beyond 100 eV. Analysis was extended up to 200 eV and yielded parameters for a large number of resonances on which only very little information had been available earlier.

The fission cross-section was measured by means of a large gas scintillator containing 2 g of fissionable material. The paper gives a brief description of the equipment used. The resolution was 10 ns/m, but the smallness of the cross-section below the fission threshold is such that only about 25 resonances (as against 250 in transmission) can be observed between 0.5 eV and 300 eV. The largest of these resonances corresponds to a $\sigma_0 \Gamma_f$ of about 1.7 b eV. The fission widths are given for all the resonances observed and a maximum value for the others. Extensive fluctuations are observed in the values of Γ_f accompanied, however, by a slow oscillation of the mean value with energy, which tends to group the resonances by packets.

SECTIONS EFFICACES TOTALE ET DE FISSION DE ²⁸⁷Np. La section efficace totale et la section efficace de fission de ²³⁷Np ont été mesurées à Saclay par la méthode du temps de vol. Les échantillons utilisés en transmission étaient sous forme d'oxyde avec des épaisseurs de 0,0066 et 0,002 at/b. Des résolutions de 7 ns/m entre 0,5 eV et 25 eV et de 2 ns/m entre 20 eV et 300 eV ont permis de mettre en évidence au-dessous de 100 eV un certain nombre de résonances nouvelles et d'étendre au-delà de 100 eV les données connues jusqu'ici. L'analyse a été poussée jusqu'à 200 eV et fournit les paramètres d'un grand nombre de résonances pour lesquelles on n'avait encore que très peu de renseignements.

La section de físsion a été mesurée au moyen d'un scintillateur gazeux de grandes dimensions contenant 2 g de matière fissile. On donne une brève description de l'appareillage. La résolution était de 10 ns/m, mais la faiblesse de la section efficace, au-dessous du seuil de fission, est telle qu'on n'a pu observer, entre 0,5 eV et 300 eV, qu'environ 25 résonances contre plus de 250 en transmission. La plus grande de ces résonances correspond à une valeur de $\sigma_0 \Gamma_f$ d'environ 1,7 b·eV. On donne les valeurs des largeurs de fission de toutes les résonances observées ainsi qu'une valeur maximale pour les autres. On observe de grandes fluctuations dans les valeurs de Γ_f avec cependant une oscillation lente de la valeur moyenne avec l'énergie qui tend à grouper les résonances par paquets.

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СЕЧЕНИЕ ДЕЛЕНИЯ ПЛУТОНИЯ-238 И АМЕРИЦИЯ-241 МОНОХРОМАТИЧЕСКИМИ РЕЗОНАНСНЫМИ НЕЙТРОНАМИ

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Abstract — Аннотация

FISSION CROSS-SECTIONS OF PLUTONIUM-238 AND AMERICIUM-241 FOR MONOCHROMATIC RESONANCE NEUTRONS. A study has been made of the dependence of the fission cross-sections of plutonium-238 on neutron energies in the range 0.02-100 eV and of americium-241 in the range 0.02-50 eV. The paper gives results of measurements of $\sigma_0 \Gamma$ for the first five resonances of plutonium-238. Fission widths have been estimated for three resonances of plutonium-238 and thirteen resonances of americium-241. Measurements of the dependence of the fission cross-sections of isotopes of transuranium elements in the resonance range of neutron energies is becoming of increasing importance, both as regards the design and installation of nuclear reactors with high neutron flux and in connection with fission physics. Special reference was made to the latter aspect of this work at the Salzburg Symposium on the Physics and Chemistry of Fission. The problems involved in making such measurements are mainly due to the marked instability of most of the isotopes of transuranium elements in respect of α -decay (and in some cases in respect of spontaneous fission) and also to the small cross-section values resulting from their sub-barrier fission. Thanks to the work carried out in the Soviet Union and other countries, it has been possible in recent years to make considerable progress in overcoming the difficulties due to the high specific α -activity of the materials in question. This has made it possible to measure the fission cross-sections of a number of isotopes of transuranium elements in the resonance range of neutron energies.

The paper reports measurements of the fission cross-sections of plutonium-238 and americium-241 using the time-of-flight method on the linear electron accelerator of the 1. V. Kurchatov Institute of A tomic Energy.

СЕЧЕНИЕ ДЕЛЕНИЯ ПЛУТОНИЯ-238 И АМЕРИЦИЯ-241 МОНОХРОМАТИЧЕСКИМИ РЕЗОНАНСНЫМИ НЕЙТРОНАМИ. Исследована зависимость сечения деления плутония-238 от энергии нейтронов в интервале 0,02 – 100 эв и америция-241 в интервале 0,2 – 50 эв. Приведены результаты определения величины σ₀Г для первых пяти резонансов плутония-238. Для трех резонансов плутония-238 и 13 резонансов америция-241 оценены делительные ширины.

Измерение зависийости сечений деления изотопов трансурановых элементов в резонансной области энергий нейтронов приобретает все большее значение как в связи с проектированием и строительством ядерных реакторов с высоким потоком нейтронов, так и с точки зрения интересов физики деления. На последний аспект этой задачи было указано, в частности, на симпозиуме по физике и химии деления в Залыцбурге. Трудности проведения подобных измерений связаны, в основном, с сильной неустойчивостью большинства изотопов трансурановых элементов по отношению к *о*-распаду (а в некоторых случаях и по отношению к спонтанному делению), а также с небольшими величинами сечений, вследствие подбарьерности их деления. В последние годы, благодаря исследованиям, проведенным в Советском Союзе и в ряде других стран, удалось существенно продвинуться по пути преодоления трудностей, вызываемых большой удельной *о*-активностью исследуемых материалов. Это позволило произвести измерения сечений деления некоторых изотопов трансурановых элементов в резонансной области знергии нейтронов.

В докладе сообщается об измерениях сечений деления плутония-238 и америция-241, выполненных по методу времени пролета на линейном ускорителе электронов Института атомной энергии им. И.В.Курчатова [1].

ГЕРАСИМОВ

Для улучшения соотношения между постоянным фоном, создаваемым α -излучением и осколками спонтанного деления (при работе с плутонием-238), измерения проводились на небольшом пролетном расстоянии ~210 см. Поэтому детектирующая аппаратура располагалась внутри биологической защиты ускорителя. Полное временное разрешение спектрометра нейтронов равнялось 2; 0,7 и 0,3 мксек/м при энергиях нейтронов 1; 10 и 100 эв соответственно (ширина канала анализатора – 0,25 мксек, длительность нейтронного импульса – 0,2 мксек). Существенный вклад в разрешение внесла неопределенность, связанная с размерами мишени ускорителя. Эта неопределенность была оценена из экспериментальных данных и оказалась равной ~4 см.

Измерения сечений деления производились одновременно с помощью двух искровых счетчиков, расположенных в общем объеме друг за другом. В одном из счетчиков находилась мишень с ~1,1 мг плутония-238, содержащая небольшие примеси других элементов:

Pu $^{239} - 7 \cdot 10^{-4}$ Am $^{241} - 5 \cdot 10^{-5}$ Cm $^{242} - 10^{-9}$ Pu $^{240} - 2 \cdot 10^{-4}$ Am $^{242} - 1,5 \cdot 10^{-5}$ Cm $^{244} - 10^{-3} *$ Am $^{243} - 7 \cdot 10^{-4}$

В другом счетчике была мишень с ~25-30 мг америция-241. Импульсы от счетчиков подавались на два 1024-канальных временных анализатора.

Нейтронный поток, необходимый для расчета относительного хода энергетической зависимости сечения деления, измерялся коронным счетчиком типа СНМ-13 с твердым борным покрытием. В результате измерений введена поправка на самоэкранирование при энергиях нейтронов <2 эв.

Фон для деления и нейтронного потока определялся в отдельных сериях измерений с резонансными фильтрами Cd, In, Ag, Мо и Со в пучке нейтронов. Постоянный фон от а-излучения и спонтанного деления, кроме того, измерялся в конце каждого цикла работы ускорителя в течение (2 – 4) 10³ мксек. До энергии ≤ 5 эв фон в основном складывался из счета α-частиц и осколков спонтанного деления (~0,4 имп/час • мксек для плутония-238 и ~0,8 имп/час • мксек для америция-241); при E_n>5 эв сильно возрастал нейтронный фон. Выше 15 эв в измерениях с америцием-241 нейтронный фон во много раз превыщал эффект от деления в провалах между резонансами, и статистическая точность результатов мала. Средняя скорость счета осколков в резонансах при энергии 1,27 и 10,05 эв равнялась ~4 и 1,7 имп/час соответственно. Всего в резонансе при энергии 1,27 эв в сериях измерений с шириной канала анализатора 0,25 мксек зарегистрировано 460 имп. Статистическая точность измерений сечения деления в провалах между резонансами не лучше 30 - 100%.

При измерениях с плутонием-238 до энергии <100 эв счет в провалах между резонансами практически совпадал с постоянным фоном. Выше 100 эв происходило возрастание нейтронного фона. Статистическая точность измерений даже в резонансах не превышала 5%.

Ниже приведены результаты измерений.

^{*} Анализ состава выполнен на магнитном α-спектрометре в лаборатории С.А.Баранова.



Рис. 1. Сечение деления плутония-238.

плутоний-238

. Сечение деления плутония-238 до энергии ~40 эв измерено с шириной канала анализатора 0,5 мксек, а выше 40 эв - 0,25 мксек.

Наибольший интерес при исследовании сечения деления плутония-238 могло бы представлять определение делительных ширин резонансов и анализ их распределений с точки эрения канальной модели деления. К сожалению, отсутствие надежных данных по полному сечению плутония-238 и небольшой интервал энергий, в котором произведены измерения сечения деления с достаточно хорошим разрешением, затрудняют такой анализ. Для первых трех резонансов, для которых известны значения полной и нейтронной ширины, можно приближенно оценить величину делительной ширины. Для остальных резонансов при энергии до 100 эв определены лишь значения $\sigma_0 \Gamma$. Эти данные приведены в табл.1.

Среднее значение делительной ширины для трех резонансов равно (7±4) Мэв. Это значение близко к величине, которую можно получить путем расчета из соотношения Хила-Уилера, взяв параметры для барьера деления из работы Воротникова и др., в которой проанализированы пороги деления в области энергий нейтронов до 1,5 Мэв [4].

АМЕРИЦИЙ-241

Сечение деления америция-241 в области энергий нейтронов до 0,9 эв измерено с шириной канала анализатора 1 мксек, а выше 0,9 эв

Е, эв	Г _f , мэв	σ ₀ Г, барн•эв
2,89	0,5±0,2	0,76±0,15
9,98	11 ±6	7,8 ±0,6
18,56	11 ±6	14,0 ±1,0
70,3	2	6,3 ±0,7
83,2		20,0 ±1,8
· ·		

ГЕРАСИМОВ



Рис. 3. Относительный ход о_f(E).

с 0,25 мксек. Результаты измерений до ~1,5 эв суммированы по четыре канала для уменьшения числа точек. Привязка отдельных серий произведена по площади резонанса при энергии 1,27 эв.

Относительный ход σ_f(E) (рис.2 и 3) нормирован при энергии 0,0253 эв к величине 3,13 барна [5].

Из экспериментальных данных для 13 резонансов при энергий <15 эв определено отношение параметров Г_f/Г методом анализа площадей. Значения gГn[•] для расчета взяты в [3]. Результаты приведены в табл. 2.

Среднее значение делительной ширины, рассчитанное из параметров первых резонансов при условии, что $\Gamma = 50$ Мэв, равно 0,22±0,08 Мэв.

Автор благодарит В.С.Загорнова за помощь при проведении эксперимента и обработке данных, В.С.Зенкевича и М.И.Певзнера за обсуждение результатов.
Е ₀ , эв	$(\Gamma_{\rm f}/\Gamma) \cdot 10^2$	Е ₀ , эв	$(\Gamma_{\rm f}/\Gamma) \cdot 10^2$
0,306	0,76±0,12	4,40	-
0,575	0,34±0,05	5,05	0,55±0,25
1,27	0,74 ±0,07	5,48	1,01±0,10
1,68	-	6,20	0,51±0,25
1 ,93	0,16±0,03	9,30	$0,59\pm0,25$
2,36	0,45±0,15	10,05	3,2 ±0,6
2,60	0,32±0,10	15,04	0,85±0,20
4,00	0,36±0,09		l.

ТАБЛИЦА 2. Г./Г ДЛЯ АМЕРИЦИЯ-241

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DISCUSSION

C.D. BOWMAN: We have measured the fission cross-sections of 238 Pu using the spark chamber as fission detector and the Livermore linac as neutron source, and have found areas of resonances up to 300 eV. When these results are combined with total cross-section measurements performed at Idaho Falls, a complete set of resonance parameters is obtained for many resonances. This nucleus is almost ideal for a search for the (n, γ f) reaction discussed by Dr. Lynn, where the nucleus captures a neutron and forms a compound nucleus, which decays by gamma emission to a new compound nucleus with different spin and parity. The nucleus might still have sufficient energy for fission, provided a channel for fission is open for the new spin and parity. Since fission from s-wave resonances is strongly inhibited in this nucleus, nearly all the observed fissions might be via the (n, γ f) reaction.

Since the fission event has a gamma ray as a precursor, the fission width distribution is characterized by many channels rather than by a few, as in ordinary fission. The fission width distribution for this nucleus is characterized by 7 ± 1.5 channels, providing strong evidence for this reaction in 238 Pu.

M.J.B. NEVE de MEVERGNIES (Chairman): Are the resonances which show a fission component in ²³⁸Pu associated with particularly small values of Γ_n (if the latter are known)?

V.F. GERASIMOV: At present the neutron widths for 238 Pu are known only for the first three resonances, so such a correlation can hardly be assumed.

MEASUREMENT OF THE NEUTRON TOTAL CROSS-SECTION OF ²⁴⁰Pu

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Abstract

MEASUREMENT OF THE NEUTRON TOTAL CROSS-SECTION OF ²⁴⁰Pu. The neutron total crosssection of ²⁴⁰Pu has been determined by time-of-flight techniques in the energy range from 20 eV to about 800 eV. For 36 of 42 resonances in this region values for Γ_0^0 have been determined by shape analysis. Between 800 eV and 5 keV the resonance structure has been investigated. In this energy range 116 resonances could be detected. The experiment was performed with the CBNM 60-MeV electron linear accelerator as a pulsed neutron source. Nominal resolutions are given.

1. INTRODUCTION

Because of its high neutron resonance capture cross-section ²⁴⁰Pu plays an important role in reactor neutron economy. On the other hand the knowledge of the resonance parameters of this isotope is rather limited. Up to now only a few very high resonances at low neutron energies could be investigated with a fair degree of accuracy. Above 120 eV no resonance widths are evaluated and resonance energies are only known up to 1 keV [1-4].

The published low-energy cross-section data may satisfy the design requirements for thermal reactors. Resonance parameters needed for fast reactor Doppler coefficient calculations are, however, almost completely missing.

This lack of data was chiefly caused by the limited quantities and enrichments of the available ²⁴⁰Pu samples. To improve this situation a batch of 7 samples of ²⁴⁰Pu with a total weight of about 87 g and excellent isotopic purity has been recently supplied by the United States Atomic Energy Commission. Three of these samples could be used in the experiment described below.

For ²⁴⁰Pu in principle the resonance energies and widths Γ , Γ_{n} and Γ_{γ} can be determined from transmission experiments alone applying shape and area analysis methods. The attainable accuracy depends, however, strongly on the range of available sample thicknesses.

The demands on sample quantity are less stringent and the attainable accuracy of the resonance width determination can be considerably improved if transmission measurements are combined with capture or scattering measurements or, even better, with both together [5-7].

The high resolution transmission measurements to be described here represent the first part of such combined experiments. Capture and scattering measurements are planned.

The results given in this paper have been obtained with the 60-MeV electron linear accelerator of EURATOM, which is operated by the



- FIG.1. Linac neutron time-of-flight spectrometer
- Total cross-section experiment -

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Central Bureau for Nuclear Measurements (C. B. N. M.) since April this year except for an intermediate period of two months.

2. SAMPLES

Three metallic discs of ²⁴⁰Pu enriched to 98% and alloyed with 1.15% (weight) aluminium have been used. The characteristics of these samples which were prepared by Los Alamos Scientific Laboratory are:

	7.62	± 0.013 cm
	0.675	\pm 0.002 g/cm ²
²³⁸ Pu:	0.004	±0.002%
²³⁹ Pu:	1.43	±0.02%
²⁴⁰ Pu:	97.92	±0.04%
²⁴¹ Pu:	0.52	±0.03%
²⁴² Pu:	0.12	±0.01%
	²³⁸ Pu: ²³⁹ Pu: ²⁴⁰ Pu: ²⁴¹ Pu: ²⁴² Pu:	$\begin{array}{rrrr} 7.\ 62 \\ 0.\ 675 \\ 2^{38} \mathrm{Pu:} & 0.\ 004 \\ 2^{39} \mathrm{Pu:} & 1.\ 43 \\ 2^{40} \mathrm{Pu:} & 97.\ 92 \\ 2^{41} \mathrm{Pu:} & 0.\ 52 \\ 2^{42} \mathrm{Pu:} & 0.\ 12 \end{array}$

3. MEASUREMENT PROCEDURE (see Fig. 1)

Short neutron bursts of high intensity are produced by the linear accelerator electron beam impinging on a mercury-cooled natural uranium target. Table I shows the main characteristics of this pulsed neutron source. The neutrons are moderated by a polyethylene slab. (This is separated from an opposite one by a ^{10}B layer to avoid resolution deterioration.) They then enter an evacuated flight path of 100-m length along which they are collimated by sand and B_4C-H_2O collimators as shown in Fig. 2. The samples are placed half-way between target and detector in an automatic sample changer.

The detector consists of a slab of 10 B powder (11 cm \times 11 cm \times 2 cm) viewed laterally by 4 NaI crystals (4 in.diam. \times 3 in.) and XP 1040 photomultipliers. The total detection system is shielded with lead, barytes concrete, Li₂CO₃ and paraffin.

Neutron flight times are measured with a 4096 channel time analyser. (Channel widths are selectable between 10 ns and 2.56 μ s.) The treatment of the detector signals follows conventional methods. The data acquisition is fully automatic. Extraneous backgrounds were determined by means of the "black resonance" technique.

¹⁰B overlap filters and Linac burst frequencies were chosen in such a way that the reaction rate due to overlap neutrons was less than 1% of that of the "useful" neutrons.

4. DATA HANDLING

The transmission data accumulated in the multichannel analyser are recorded on magnetic tape. These magnetic tape data are then checked for validity and reduced on a small computer (IBM 1401). A teleprocessing system (IBM 7702) transfers the reduced data at a speed of 150 characters/sec via telephone lines to the EURATOM Computer Centre

Burst width	Maximum frequency (Hz)	Peak current (A)
10 ns	1000	3.6
100 ns	880	1.2
1 µs	380	0.4
2 μs	250	0.4

TABLE I. MAIN CHARACTERISTICS OF THE C. B. N. M. LINAC

CETIS at Ispra (distance 1000 km). Here the data are analysed on an IBM 7090 computer to get the resonance parameters. Results of this analysis are retransferred to Geel by the tele-processing system and listed by the small computer or plotted by a digital plotter operated by a magnetic tape unit.

5. RESULTS

Total cross-sections, not corrected for Doppler and resolution effects, were determined in the energy range from 20 eV to 800 eV. They are partly shown in Fig. 3 and Fig. 4. In the energy range between 800 eV and 5 keV only sample-in measurements were performed. Figures 5a and b demonstrate the resonance structure of 240 Pu in that range. Nominal resolutions ($\Delta t_{burst} + \Delta t_{channe}$)/L varied as follows:

Energy range	Nominal resolution
20 eV - 210 eV	6 ns/m
210 eV - 310 eV	$1.8 \rm ns/m$
310 eV - 800 eV	0.9 ns/m
800 eV - 5000 eV	$0.9 \mathrm{ns/m}$

158 resonances have been found between 20 eV and 5 keV (Tables II and III). Figure 6 presents the cumulative number of levels versus neutron energy. It indicates that above about 1 keV an increasing number of levels remained undetected. Since the resolution of the spectrometer can still be increased by choosing channel and burst widths of 10 ns instead of 40 ns considerable improvements in level detection are possible.

The shape analysis programme of Atta and Harvey [8] has been used to determine the resonance parameters Γ and Γ_n^0 which are listed in Table II. (In this analysis Doppler broadening and instrument resolution are represented by Gaussian functions.) Only the predominant resonances could be analysed which are supposed to be due to s-wave neutrons. The errors for the resonance widths given in Table II are based on the assumption that the calculated widths of the resolution function have an uncertainty of \pm 15%. This value is mainly due to the uncertainties of the moderation time and the burst width.



FIG. 2. Experimental set-up for total cross-section experiments.



FIG.3. Total cross-section in the energy range from 80 to 320 eV.



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FIG. 4. Total cross-section in the energy range from 300 to 800 eV.







FIG.5b: Transmission in the energy range from 1600 to 5000 eV. (Sample-in measurements only).

For all resonances analysed by the shape method the Doppler and the resolution widths are greater than the natural widths. Therefore the obtained values of Γ_n are much more reliable than those of Γ . Nevertheless the latter have been included in Table II, indicating the limitation of this type of analysis. The Doppler constant has been calculated for an effective temperature equal to the ambient temperature.

E(eV)	Γ _n (meV)	r _n 0	Γ(meV) ^a
20.46 ± 0.009	(3.44 ± 0.09)	(0.76 ± 0.02)	(25 ± 3)
38.34 ± 0.02	18.3 ± 0.7	2.96 ± 0.12	43 ± 2
41.64 ± 0.02	16.1 \pm 0.5	2.50 ± 0.08	49 ± 2
66.66 ± 0.04	48.0 ± 1.6	5.88 ± 0.20	95 ± 5
72.83 ± 0.04	21.8 \pm 0.7	2.56 \pm 0.09	44 ± 5
90.78 ± 0.06	13.3 ± 0.3	1.39 ± 0.03	42 ± 6
92.50 ± 0.06	2.9 ± 0.1	0.30 ± 0.01	-
105.05 ± 0.07	44.2 ± 1.6	4.31 ± 0.16	76 ± 6
121.67 ± 0.09	13.9 ± 0.3	1.26 ± 0.03	49 ± 11
135.2 ± 0.1	17.8 ± 0.2	1.53 ± 0.02	56 ± 10
151.7 ± 0.1	13.6 ± 0.1	1.10 ± 0.01	46 ± 16
162.9 ± 0.1	8.6 ± 0.1	0.67 ± 0.01	52 ± 24
170.3 ± 0.1	13.4 ± 0.3	1.03 ± 0.02	50 ± 22
186.1 ± 0.2	16.0 ± 0.3	1.17 ± 0.02	57 ± 26
199.6 ± 0.2	-	-	-
239.3 ± 0.1	11.3 ± 0.3	0.73 ± 0.02	-
260.7 ± 0.1	-	-	-
287.3 ± 0.1	125.1 ± 3.7	7.38 ± 0.22	190 ± 16
305.1 ± 0.1	7.0 ± 0.4	0.40 ± 0.02	
318.5 ± 0.1	-	-	-
320.9 ± 0.1	18.6 ± 0.4	1.04 ± 0.02	96 ± 20
338.7 ± 0.1	5.7 ± 0.4	0.31 ± 0.02	-
346.2 ± 0.1	16.2 ± 0.4	0.87 ± 0.02	67 ± 27
364.0 ± 0.1	30.9 ± 0.4	1.62 ± 0.02	74 ± 23
372.3 ± 0.1	13.3 ± 0.4	0.69 ± 0.02	-
405.0 ± 0.1	102.5 ± 1.6	5.09 ± 0.08	173 ± 19
419.0 ± 0.1	-	-	
450.2 ± 0.2	16.8 ± 0.9	0.79 ± 0.04	119 ± 56
466.4 ± 0.2	-	-	-
473.2 ± 0.2	-	-	-
494.2 ± 0.2	5.1 \pm 0.4	0.23 ± 0.02	-
499.6 ± 0.2	18.6 ± 0.7	0.83 ± 0.03	-

TABLE II. RESONANCE ENERGIES AND WIDTHS BETWEEN 20 eV AND 750 eV

TABLE II (contd.)

E(eV)	$\Gamma_{n}(meV)$	Γ_n^0	Γ(meV) ^a
514.6±0.2	20.4 ± 0.9	0.90 ± 0.04	-
546.8 ± 0.2	29.9 ± 0.7	1.28 ± 0.03	-
553.5 ± 0.2	16. 7 ± 0.7	0.71 ± 0.03	-
566.6 ± 0.2	30.0 ± 0.7	1.26 ± 0.03	-
597.2 ± 0.2	53. 0 ± 1.0	2.17 ± 0.04	-
608.4 ± 0.2	20.5 ± 1.2	0.83 ± 0.05	-
632.6 ± 0.2	12.3 ± 1.0	0.49 ± 0.04	-
637.8 ± 0.2	· -	-	
665.5 ± 0.3	183 ± 3	7.11 ± 0.13	279 ± 40
678.9 ± 0.3	24.0 ± 1.0	0.92 ± 0.04	-

^a See section 5 of the text.



FIG. 6. Integral number N of levels versus neutron energy E.

IADLE III. RESONANCE ENERGIES BEIWEEN (SU eV and SIVU e	ABLE III. RESONA	NCE ENERGIES	BETWEEN	750 eV	and 5100 e
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	750.5 ± 0.3	1450.3 ± 0.7	2199 ± 1	3267 ± 2
	759.6 ± 0.3	1541.3 ± 0.7	2256 ± 1	34 65 ± 2
	791. 4 ± 0. 3	1550.0 ± 0.7	2279 ± 1	3 501 ± 2
	811. 0 ± 0. 3	1564.0 ± 0.7	2292 ± 1	3568 ± 2
	820.4 ± 0.3	1576.2 ± 0.7	2367 ± 1	3610 ± 2
	855.4 ± 0.3	1611.0 ± 0.7	2386 ± 1	3658 ± 2
	876.9±0.4	1621.6 ± 0.8	2405 ± 1	3727 ± 2
	891.8 ± 0.4	1643.3 ± 0.8	2417 ± 1	3854 ± 2
	904.1 ± 0.4	1663.1 ± 0.8	2435 ± 1	3902 ± 2
	909.5 ± 0.4	1688.6 ± 0.8	2471 ± 1	3918 ± 2
	915.5 ± 0.4	1724.8 ± 0.8	2521 ± 1	3953 ± 2
	944.0±0.4	1742.1 ± 0.8	2549 ± 1	3977 ± 2
	958.8 ± 0.4	1763.8 ± 0.8	2640 ± 1	4021 ± 2
	971.6 ± 0.4	1779.3 ± 0.8	2668 ± 1	4098 ± 2
	1002.4 ± 0.4	1841.5 ± 0.9	2693 ± 1	4121 ± 2
	1042.0 ± 0.4	1851.9 ± 0.9	2739 ± 1	4150 ± 3
	1072.8 ± 0.5	1873.9 ± 0.9	2750 ± 1	4203 ± 3
	1100.2±0.5	1882.8 ± 0.9	2819 ± 2	4269 ± 3
	1129.1 ± 0.5	1901.7 ± 0.9	2844 ± 2	4287 ± 3
	1148.4 ± 0.5	1918.1 ± 0.9	2904 ± 2	4331 ± 3
	1165.6 ± 0.5	1949.7 ± 0.9	2939 ± 2	4375 ± 3
	1191.2 ± 0.5	1957 ± 1	2969 ± 2	4458 ± 3
	1209.4 ± 0.5	1973 ± 1	2981 ± 2	4497 ± 3
	1255.2 ± 0.6	1992 ± 1	3015 ± 2	4571 ± 3
	1300.7 ± 0.6	2023 ± 1	3078 ± 2	4588 ± 3
	1328.8 ± 0.6	20 3 4 ± 1	3110 ± 2	4615 ± 3
	1350.7 ± 0.6	2055 ± 1	3173 ± 2	4719 ± 3
	1377.2 ± 0.6	2084 ± 1	3193 ± 2	4958 ± 3
	1426.5 ± 0.6	2182 ± 1	3239 ± 2	5071 ± 3

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NEUTRON RESONANCE PARAMETERS OF ²⁴⁰PU

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Abstract

NEUTRON RESONANCE PARAMETERS OF ²⁴⁰Pu. The Harwell 45-MeV electron linear accelerator pulsed neutron source has been used to measure the total, capture and scattering cross-sections of ²⁴⁰Pu over the energy range 10 to about 1000 eV by the time-of-flight method. Flight paths of 94, 55 and 32 m were used for the total, scattering and capture measurements, respectively, giving best resolutions of 2.6, 4,5 and 10 ns/m respectively. Two discs of Pu metal, containing about 98% ²⁴⁰Pu, of 7.6-cm diameter were used in the measurements, giving sample thicknesses of 0,0036, 0,00083 and 0,00119 atom/b.

Resonance parameters have been determined by shape analysis and by area analysis combining the three sets of data. A preliminary study of the resonances below 300 eV indicates a mean radiation width of ~ 25 MeV.

ИНТЕГРАЛЬНЫЕ И ДИФФЕРЕНЦИАЛЬНЫЕ СЕЧЕНИЯ ДЕЛЕНИЯ ТОРИЯ-232 НЕЙТРОНАМИ

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Abstract — Аннотация

INTEGRAL AND DIFFERENTIAL FISSION CROSS-SECTION OF 232 Th BY NEUTRONS. Measurements of fission cross-sections of 232 Th in the energy range from 0.6 to 3 MeV and angular distribution of fission fragments have been carried out. The measurements were carried out using electrostatic generators and T (p, n) reaction. Channel analysis was done and the results are given.

ИНТЕГРАЛЬНЫЕ И ДИФФЕРЕНЦИАЛЬНЫЕ СЕЧЕНИЯ ДЕЛЕНИЯ ТОРИЯ-232 НЕЙТ-РОНАМИ. На электростатических генераторах с использованием реакции T(p, n) измерены сечения деления тория-232 в диапазоне энергий нейтронов от 0,6 Мэв до 3 Мэв и измерены угловые распределения осколков деления.

Проведен каналовый анализ, и сообщаются результаты.

DISCUSSION

(on papers CN-23/89, CN-23/31 and CN-23/127)

N.J. PATTENDEN: I should like to comment on our paper CN-23/31. We have made measurements on ²⁴⁰Pu neutron cross-sections with the Harwell electron linac. Transmission, capture and scattering measurements have been carried out, and examples of the capture and scattering data are shown in Figs. 2 and 3. Each type of data gives a different Γ_n , Γ_y relationship, and we have combined the results to give best values of Γ_n and Γ_γ for 17 resonances. An example of this is shown in Fig. 4. The mean value of radiation width from these resonances is 18.1 ± 1.8 MeV. The transmission data alone gave Γ_n values for 40 resonances. Dr. Böckhoff has prepared slides comparing the Harwell and Geel Γ_n values, and with the exception of the 20 and 90 eV resonances the agreement between the two sets of data is in general very good. Further work is proceeding at Harwell on more samples covering a wider range of thickness.

J.S. STORY: I would draw attention to the fact that Pattenden et al. report very much lower values for the radiation widths of the higher ²⁴⁰Pu resonances than those commonly accepted for the very important 1-eV resonances. Is this a real effect or ought we to be asking for rather more direct measurements of the capture cross-section through the 1-eV resonance? The commonly used Breit-Wigner analysis of gaseous ²⁴⁰Pu may perhaps be inadequate for the 1-eV resonance.

N.J. PATTENDEN: We appreciate that our radiation widths are much lower than those accepted for the 1-eV resonance. As mentioned in the paper, our values are preliminary, but it does not seem likely that any revision will be sufficient to bring them into reasonable agreement with DISCUSSION

the 1-eV resonance. Hence, we feel that the 1-eV resonance could be reexamined, both experimentally and from the point of view of its analytical description.

M.A. HOLMBERG: I should like to make a comment about paper CN-23/127. I think that this measurement on the fission cross-section of ²³²Th has shown that the positions of the first plateaus in the cross-section correspond very well to the inelastic neutron scattering to the known levels at 800 keV and 1200 keV.

However, there is also a very marked plateau in the fission crosssection at 1.4 MeV and a strong decrease at 1.6 MeV. Dr. Lynn suggested that these effects were <u>not</u> due to the competition with the neutron reemission. Strömberg and I have made measurements on the inelastic neutron cross-section in this energy region, and at 1.4 MeV we have not observed any levels at all. At 1.6 MeV there is a level or levels, but the inelastic cross-section is not large enough to explain the strong decrease in the fission cross-section. In other words, our measurement seems to be in agreement with the suggestions made by Dr. Lynn.

SPARK CHAMBER MEASUREMENT OF NEUTRON FISSION CROSS-SECTIONS AND RELATED PARAMETERS*

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Abstract

SPARK CHAMBER MEASUREMENT OF NEUTRON FISSION CROSS-SECTIONS AND RELATED PARAMETERS. As reactor burn-up calculations become more detailed, a knowledge of the nuclear properties of the less common heavy nuclides becomes increasingly important. The fission cross-section of many of these nuclides can be measured with modern electron linear accelerators as pulsed neutron sources and a sample of several milligrams. Samples of this size are now becoming available for many nuclides. However, nearly all of these nuclides are much shorter-lived than the more common fissionable nuclides such as ²³⁹Pu. Therefore, a major problem in these measurements is the development of a detector which is capable of distinguishing fission fragments from the intense background radiation arising from the natural radioactivity of such samples. The insensitivity of the corona-spark counter to gamma rays, electrons and alpha particles makes it especially useful for measurements on these isotopes. With the exception of spontaneous fission, natural radioactivity appears to be a serious problem in the use of this detector for only a few of the most alpha-active and least fissionable nuclei. Since the detector is insensitive to the intense gamma flash associated with the electron pulse from a high-power linear electron accelerator, measurements can be made at very short times after the gamma burst; that is, into the MeV region. Measurements are, therefore, possible from thermal energy to several MeV with this detector. The cross-section can then be normalized at thermal energy to the usually well-known thermal fission cross-section so that a knowledge of detector efficiency, absolute neutron flux, and sample size are not required for determining absolute cross-sections. The usefulness and limitations of the technique are illustrated by measurements on ^{242 m}Am from 0. 02 eV to 6 MeV obtained with a 50-mg Am sample containing about 20% ²⁴² mAm and 80% ²⁴¹ Am. Other uses of this detector in measuring $\overline{\nu}$, alpha, and spontaneous fission half-lives are presented.

Introduction

As fission burn-up calculations become more detailed, a knowledge of the fission phenomena of the less-common heavy nuclides, such as 242Am, 241Am, and 232U, becomes increasingly important. In addition, the use of very α -active isotopes, such as 238Pu and 244Cm, in isotopic power sources for space applications requires accurate information on their fission properties. This kind of information will undoubtedly also contribute to a better understanding of the fission process in general. Useful measurements can be made using a modern linear electron accelerator as a pulsed neutron source with samples of only a few milligrams, and samples of this size are now becoming available for many of the rarer nuclides. However, nearly all of these are much shorter lived than the more common fissionable nuclides, such as ^{239}Pu . Therefore, a major problem in these measurements is the development of a detector which is capable of distinguishing fission fragments from the intense background radiation arising from natural radioactivity of such samples.

*Work performed under the auspices of the United States Atomic Energy Commission.

The technique usually employed for discrimination against alpha particles is that of reducing the resolving time of the detector until the probability is very small that a sufficient number of alpha particles may add their pulse heights so as to exceed a bias level set for fission fragments. The gas scintillator has proven to be quite useful for this purpose. [1] In principle, an ordinary parallel plate ionization chamber should be nearly as effective when the chamber is used with a current-sensitive preamplifier. [2] Ultimately, however, this general approach will prove inadequate as larger masses of nuclides with ever-shorter half-lives are studied.

The development of detectors which are sensitive to the ionization density of the particle and not the total energy appears to offer a considerable improvement in the discrimination against alpha particles which may be obtained in a practical detector. Such detectors may have a resolving time which need not be particularly short. For pile-up to occur, the particles must not only be emitted within this time, but they must traverse the same differential volume element of space in the detector. Only then will the ionization density of several alpha particles add together so as to be confused with a fission fragment. The critical parameters of such a detector then are the effective resolving time and the size of the differential volume element. Without carrying out a thorough study of the significance of these two parameters, two detectors operating on these principles have been shown to be successful: track detectors [3] such as mica which, of course, are not suitable for time-of-flight experiments; and the spark chamber, [4] which we discuss here. This detector appears to be of general usefulness; and, with the exception of spontaneous fission, natural radioactivity appears to be a serious problem for only a few of the most alpha-active and least-fissionable nuclei. Since the detector is insensitive to gamma radiation, the detector ignores the intense gamma flash associated with the electron pulse from a highpower linear accelerator. Therefore, fission measurements can be made immediately after or even during the gamma flash. Measurements are possible, therefore, with contemporary linacs from thermal energy to several MeV with this detector. The relative cross-section can then be normalized to the usually well-known thermal fission cross-section so that an accurate knowledge of detector efficiency, absolute neutron flux, and sample size are not required for determining absolute crosssections. These techniques will be illustrated by measurements [5] on 242mAm from .014 eV to 4 MeV and on 241Am over a more limited energy range. Briefer descriptions of other experiments are also included.

Description of the Detector

The detector is shown in Fig. 1. Each of two .0075-cm thick foils of copper were corrugated into thirty-two adjacent 0.32-cm diameter channels. The corrugated foils were then glued back-to-back into a supporting, non-conducting frame. Wires of 0.0123-cm diameter were stretched along the axis of each cylindrical corrugation. As many as four of these detecting units can be stacked one upon the other with fission foils between so that both sides of three fission foils can be observed and one side of two others at the top and bottom of the stack. The stack is enclosed in a chamber which provides means for gas circulation. The parameters of the detector, such as the voltage between the wire and the groove, the pressure, and the composition of the gas, can be varied to obtain the best discrimination against alpha particles



FIG.1. A single detector unit of the spark chamber.

while maintaining a satisfactory efficiency for fission fragments. Several gases have been studied, some more thoroughly than others. No mixture was found to be superior to natural air, which was used in these experiments.

Fission Cross-Section Measurements

We illustrate the application of the detector to fission crosssection measurements by describing recent measurements on $^{242}M_{Am}$. Measurements on this nucleus are complicated by the growth of the ^{242}Cm contaminant by beta decay from the ^{242}Am ground state. The pertinent transitions are:

$$^{242m}Am = \frac{I.T.}{152 \text{ yr}} > ^{242}Am = \frac{\beta^{-}}{16 \text{ h}} > ^{242}Cm = \frac{\alpha}{163 \text{ d}} > ^{238}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{242}Cm = \frac{\alpha}{163 \text{ d}} > ^{238}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{242}Cm = \frac{\alpha}{163 \text{ d}} > ^{238}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{242}Cm = \frac{\alpha}{163 \text{ d}} > ^{238}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{242}Cm = \frac{\alpha}{163 \text{ d}} > ^{248}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{248}Cm = \frac{\alpha}{163 \text{ d}} > ^{248}Pu = \frac{\alpha}{86.4 \text{ yr}} > ^{248}Cm = \frac{\alpha}{163 \text{ d}} > ^{248}Cm = \frac{\alpha}{163 \text{$$

Since the 242 Cm has a relatively short spontaneous fission half-life, the spontaneous fission background becomes prohibitively large, long before equilibrium is established. It is important, therefore, that the 242 Cm be well-separated from the 242m Am before the start of the experiment and that the measurements be completed in a short time. The 242 Cm was chemically removed until the Cm/Am atom ratio was less than 3 x 10⁻⁷. About 50 mg of a mixture containing 19.8% 242 Am, 79.5% 241 Am and 0.7% 243 Am was electroplated on both sides of a .075-mm thick 10-cm diameter nickel foil to a thickness of about 300 µg/cm². Three other foils were electroplated with better than 97% isotopically pure 239 Pu, 241 Am and 238 U. The 239 Pu was used as a flux monitor at the highest and lowest energies measured. The 241 Am allowed corrections for the large 241 Am contaminant in the Am mixture. The 238 U helped measure the background and resolution in the higher-energy region.



FIG. 2. Shielding and beam collimation in the neighbourhood of the detector.

The measurements were divided into three parts: the low-energy measurements below-5 eV, the intermediate measurements between 2.3 eV and 2.5 KeV, and the high-energy measurements from 1 KeV to 6 MeV. The arrangement of the detectors in the shielding and the beam collimation in the neighborhood of the detectors is shown in Fig. 2. The detector was placed in a large cylindrical shield which was filled with boric acid solution. This shield served essentially to eliminate the large extraneous neutron background which leaked through the relatively thin shielding around the neutron source. The diameter of the neutron beam was collimated to 10 cm by the steel collimator at the end of the evacuated portion of the flight tube. The neutrons passed down the axis of the cylindrical insert containing Ca₂B₆O₁.5H₂O which was placed inside the shield. A plastic bag filled with He to reduce neutron scattering was placed in this portion of the flight path. Both the spark chamber and the BF3 tube were surrounded by Cd to prevent neutrons which were scattered into the boric acid solution from returning to the detector and causing background fission events. The whole assembly could be moved easily from one flight path position to the other.

Signals from the spark chamber were attenuated, passed through a discriminator, and then to the multichannel time analyzer or to a gated scaler. Four scalers, one for each isotope and the BF3 tube, were operated in parallel with the time analyzer. These scalers were gated on during the recording portion of the analyzer cycle and will be referred to as the "prompt" scalers. Another four scalers in parallel with the first four, were gated on for an equal-time interval which just preceded the machine pulse, where no neutrons from the machine were present. In this way we determined, in effect, the machine-off, time-independent background from alpha pile-up, etc. The latter scalers are referred to as the delayed scalers. The conditions for the three measurements are summarized in Table I.

For the low-energy measurements, data were recorded only for the Am mixture and the 239 Pu foils. At the machine pulse rate of 50 pps,

Measurement	Interval (eV)	Filters	Pulse Rate (pps)	Pulse Width (µsec)	Channel Width (µsec)	Flight Path (meters)
Low Energy	0.019-3.7	None	50	2.0	1.0	6.50
Intermediate	2.3-2.5 x 10 ³	Cd,Mn	360	0.10	0.125	6.50
Intermediate Background	2.3-2.5 x 10 ³	Cđ,Mn,Ta	360	0.10	0.125	6.50
High Energy	1 x 10 ³ -6 x 10 ⁶	Cđ	360	0.10	0.3125	15.47

TABLE I. CONDITIONS FOR MEASUREMENTS

the delayed scaler indicated that no background correction due to overlap of neutrons from one pulse to the next was required. The relative cross-section of the mixture was obtained by using the ²³⁹Pu as a flux monitor. The ²³⁹Pu fission cross-section [6] was used for this purpose. The relative cross-section of the mixture was then normalized at thermal energy using the value of 7200 ± 300 barns of Wolfsberg et al. [7] The normalized cross-section for the mixture was corrected below 3.7 eV for the ²⁴¹Am contaminant with fission crosssections measured in an independent experiment at this laboratory. [8] The resulting, ²⁴²MAm data are shown in Fig. 3.

For the intermediate-energy measurements, data were recorded for the Am mixture, 239 Pu, and the BF₃ proportional counter used as a flux monitor. An Mn absorber in the beam during the measurement allowed us to determine the background at 335 eV. The time-independent portion of this background was determined with the delayed scaler as described



FIG. 3. The fission cross-section of ²⁴² mAm. The data have been corrected for the ²⁴¹ Am contaminant.

earlier. Measurements of the time- or energy-dependence of the background were carried out in a separate run with a Ta foil also included. The data were normalized to the 3.3 eV resonance of the low-energy measurements. The results are shown in Figs. 4, 5, and 6. No 241Am correction was applied to these data above 3.7 eV since the average 241Am cross-section is only about one per cent of the 242mAm crosssection and is not well known.



FIG. 4. The fission cross-section of ²⁴² Am. The data have not been corrected for the ²⁴¹ Am contaminant.



FIG.5. The fission cross-section of ^{242III}Am. The data have not been corrected for the ²⁴¹Am contaminant. The deep minimum at 91 eV is attributed to the influence of the Cd filter.

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FIG. 6. The fission cross-section of ²⁴²mAm between 0.1 and 100 keV. The solid line is the ²³⁹Pu fission cross-section used as the flux monitor for the ²⁴²mAm in the high-energy measurements.

For the high-energy measurements, time spectra were recorded for all four foils. A portion of the raw-time spectra are shown in Fig. 7. The peaks in all four spectra at low channel numbers result from photofission induced by the gamma flash from the accelerator. These peaks serve to measure the zero time and the resolution. The broad maximum observed at later times arise from fission induced by the essentially unmoderated portion of the neutron boil-off spectrum. The low energy side of the maxima for 2^{41} Am and 23^{80} foils are truncated due to the presence of threshold fission in these nuclei. A long tail in the 239Pu and Am mixture are indicative of considerable cross-section in the KeV region. The relative cross-section of the Am mixture was determined in the region from 1 KeV to 6 MeV using the known 2^{39} Pu fission cross-section [6] as the flux monitor. The cross-section for



FIG. 7. Time spectra for the high-energy measurements. The more narrow peak of the curves arises for photofission induced by the gamma flash from the accelerator. The broad maximum of the curves arises from the boil-off spectrum of neutrons from the (γ, n) and (γ, f) reactions in the neutron source.

the mixture was normalized in the 1.0 to 2.5 KeV region where it overlapped the intermediate data. The relative ^{241}Am and ^{238}U crosssections were determined in the same way. The ^{241}Am cross-section was normalized by simultaneous recording of data during the highenergy measurements near the 5.48 eV resonance in the ^{241}Am and the 7.85 eV resonance in ^{239}Pu . Since the area of both resonances is known, [8,6] the relative cross-section above the fast-fission threshold can be normalized. The value of the cross-section at 2.5 MeV of 1.85 ± .2 barns is consistent with the value of 1.95 ± 0.2 determined by Kazarinova et al. [9]

The results of the high-energy measurement are shown in Fig. 8. The cross-section for 242m Am obtained by taking the difference between the mixture and 241 Am is shown by the solid line. The 239 Pu crosssection, which was used to determine the 242m Am cross-section is also included in the figure. The data of Fig. 8 provide a means of checking the normalization of the Am mixture and the 241 Am fission cross-section. If we assume that the 242m Am cross-section changes slowly in the MeV region, the smooth behavior of the cross-section across the 241 Am fission threshold indicates that the large contribution of the 241 Am fission cross-section to the mixture, as shown in Fig. 8, has been correctly subtracted. The slope of the 241 Am fission cross-section across the threshold is not as high as that reported in other measurements. [10] We attribute this, primarily, to our much-poorer resolution.



FIG. 8. Cross-section results in the MeV region. The dashed line is the cross-section of the Am mixture $(^{242} \text{ mAm} + 4 \, ^{241}\text{Am})$. The data denoted by the rectangular points are the results for ^{241}Am . The circles give the fission cross-section for $^{242\text{mAm}}$ after the correction for the ^{241}Am contribution has been applied to the mixture. The solid curve is the ^{259}Pu cross-section which was assumed for the data reduction.

The $\bar{\mathbf{y}}$ Measurements

The value of $\overline{\mathbf{y}}$ for 242m Am at thermal energy has been measured by Fultz et al. [11] To check the method, measurements were also made on 2350 and 2330 . The apparatus is shown in Fig. 9. A paraffinmoderated neutron detector containing a small spark chamber was used.



FIG. 9. Experimental arrangement for $\overline{\nu}$ measurements: S denotes the 10^7 -n/sec source; A, the polyethylene moderator; B is a 7.5-cm plug of polyethylene; C, paraffin; D, pellets of borated polyethylene; E, the paraffin-moderated neutron detector; F, high-pressure BF₃ tubes; G, compressed boracic acid bricks; H, the spark chamber; I, electrical leads and air tubes to the spark chamber; J, a 7.5-cm diam. collimator of borated paraffin; K, 1.5-mm cadmium sheet.

The neutron detector consisted of a 60-cm paraffin cube containing 48 high-pressure 10BF3 counters arranged in four concentric rings around a 7.5-cm diameter axial hole. The neutron detection efficiency was 40 per cent. A corona-type spark chamber 7.5 cm in diameter was placed at the center of the axial hole. It contained about 10 mg of Am with the same isotopic mixture as the material used in the fission cross-section measurements. Fission was induced by neutrons from a 107 n/sec Pu-Be source. The neutron source was placed in the axial hole of a 35.5 cm diameter polyethylene moderator and aligned with the axis of the neutron detector. A moderating plug was located immediately in front of the source to produce low-energy neutrons. The detection of fission fragments from ^{242m}Am started a gate for several scaling circuits which recorded the detection of neutrons in the BF3 counters. The decay time for the paraffin cube was approximately 100 µsec. The primary source of backgrounds arose from source neutrons which penetrated the neutron shielding around the detector. This background was obtained by measuring the neutron counts per gate when the gate was triggered by the pulse generator instead of the fission counter. With the knowledge of the number of neutrons detected per fission corrected for the efficiency of the detector and background, it was possible to obtain a value for \bar{v} . The values for ^{242m}Am , ^{235}U and ²³³U are 3.24 + 0.12, 2.43 + 0.08 and 2.54 + 0.04, respectively. If a reactor had been available, the experiment obviously could have been done much more easily. The primary significance of the use of the spark chamber for this experiment lies in the fact that it allowed measurements to be carried out on a highly-radioactive material with a neutron flux of less than 100 n/cm^2 sec. Experiments of this kind using accelerators producing variable energy but monoenergetic neutrons therefore should be possible.

Other Measurements

The apparatus described in the previous section was ideally suited for measurement of the spontaneous fission half-life of $^{24}2m_{Am}$. Measurements [12] were performed in the manner described for $\bar{\mathbf{y}}$ but without the neutron source. The build-up of the ^{242}Cm decay was measured from a newly-purified Am sample and the growth curve was extrapolated back to zero time. The residual spontaneous fission of the sample was attributed to the 242m Am spontaneous fission. The weighted average of the two measurements on two separately purified samples is $8 \pm 3 \times 10^{11}$ years, which is at least two orders of magnitude shorter than spontaneous fission systematics have predicted.

Measurements of the energy dependence of α , the ratio of captureto-fission cross-sections, could be made by placing this detector at the center of a large liquid scintillator with high efficiency for capture gamma rays. The principal problem is the rather low efficiency of the spark chamber. For $\alpha = 0.5$ and an efficiency for fission events of 20 per cent, about two-thirds of the apparent capture events arise from gamma rays released in fission events and must be subtracted to determine the true capture rate. Except for the uncertainties introduced in the subtraction, useful measurements appear to be feasible.

Summary

Fission cross-section measurements have been made with the spark chamber over as many as eight decades of energy on the nuclei, ^{241}Am , ^{242m}Am , and the even shorter lived nuclei, ^{232}U and ^{238}Pu which are not reported here. The application of the detector to $\bar{\mathbf{y}}$ and spontaneous fission studies also have been demonstrated and measurements of the energy dependence of $\boldsymbol{\alpha}$ appear to be feasible. We believe that this detector is of general usefulness for experiments requiring the detection of fission fragments. As larger samples of the shorterlived nuclides become available, many experiments should become possible for which the spark chamber will offer distinct advantages over other types of detectors.

Acknowledgments

We wish to express our appreciation to Dr. S. C. Fultz for permission to describe the \bar{y} measurements before publication and to Dr. R. W. Hoff who participated in the 242m Am fission cross-section measurements. The success of these experiments was dependent, to a large degree, on the careful work of D. E. Petrich and W. L. Ridgwell in the construction of the detectors.

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DISCUSSION

P. FIELDHOUSE: Might I suggest measuring the spontaneous fission half-life of ²⁴⁰Pu with the spark chamber described by Dr. Bowman? Present published values range from about 1.2 to 1.4×10^{11} yr, and the spread here is far in excess of the individual errors. We at Aldermaston have recently deduced a further value of about 1.16×10^{11} yr by comparing the calculated and measured neutron outputs of spontaneous fission neutron sources. An independent determination by the spark chamber would be very valuable.

C.D. BOWMAN: Measurements of spontaneous fission half-life are at present underway on other isotopes. I will communicate this request to those doing the work at Livermore.

J.C. HOPKINS: I would like to ask Dr. Bowman for a brief comparison of his spark chamber technique with the underground nuclear explosion technique, which is ideally suited to the fission cross-section measurements of highly alpha-active samples.

C.D. BOWMAN: With the exception of measurements of radioactivity from spontaneous fission, I believe the spark chamber can be used on any sample that is available in quantities exceeding 100 μ g and provided a modern accelerator is used as the neutron source. Such a machine would give an improvement of at least a factor of 100 in the data acquisition rate as compared with the accelerator used in these experiments. The data obtained in this way should be adequate for a detailed multi-level fit provided the nucleus is "thermally fissionable". Of course, the resolution obtained with the nuclear explosion will greatly exceed that obtained by this technique in regions where both experiments overlap. In addition, the nuclear explosion should be capable of measurements on samples as small as 1 μ g.

Perhaps the primary advantage of our technique is that one facility suffices for the measurement of fission cross-sections from 0.02 eV to several MeV. In addition, this full energy range may be normalized to the thermal value, which makes it unnecessary to have accurate knowledge of the foil mass of the fission-fragment bias level. These sources of error are common to the bomb experiments and the experiments carried out with the variable-energy mono-energetic neutron source.

J. THEOBALD: How is the neutron detection efficiency determined for the $\overline{\nu}$ -measurement mentioned in the paper?

C.D. BOWMAN: The efficiency was measured by several techniques, but most reliance was placed on the value determined by means of a ^{252}Cf source using the Hopkins and Diven $\overline{\nu}$ -value of 3.78 neutrons/fission.

M.J.B. NEVE de MEVERGNIES (Chairman): Don't you experience trouble due to the rather large amount of material close to the sample itself? The windows, for instance, are certainly not thin.

C.D. BOWMAN: We were worried about this problem and attempted to make the components as thin as possible. The copper grids are as thin as we can go, 0.003 in. Our results show that the problem is not serious.

DETERMINATION DU SPIN DES RESONANCES DES NOYAUX FISSILES

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Abstract — Résumé

DETERMINATION OF THE RESONANCE SPIN OF FISSILE NUCLEI. The first part of this paper, which explains the direct and indirect methods of determining the resonance spin of fissile nuclei, describes how resonance spin can be determined either by the transmission of polarized neutrons through samples of polarized nuclei, or by resonance analysis using conventional measurements, including elastic scattering.

The second part deals with all methods which consist in deducing the resonance spin from the properties of the radiative capture and fission processes which may depend on the spin state, such as partial transitions of radiative capture, total widths of radiative capture, interference between resonances of the same spin due to the exit paths of the fission phenomena, fission widths, mass distribution of the fission products, number of neutrons released per fission, probability of ternary fission and the energy spectrum of long-range α -rays, anisotropy of fission fragments induced by non-polarized neutrons in the aligned nuclei, etc.

The author presents and discusses a number of experimental results obtained by these various methods, especially for ²³⁵ U and ²³⁹ Pu. In the case of ²³⁹ Pu, a fairly coherent set of values can be obtained independently by several different methods.

DETERMINATION DU SPIN DES RESONANCES DES NOYAUX FISSILES. L'auteur présente les différentes méthodes de détermination du spin des résonances des noyaux fissiles. Ces méthodes sont classées en deux catégories: méthodes dites directes et méthodes indirectes. Dans la première partie, l'auteur décrit comment le spin des résonances peut être connu soit par transmission de neutrons polarisés à travers des échantillons de noyaux polarisés, soit par l'analyse des résonances à partir de mesures classiques dont celle de diffusion élastique.

La deuxième catégorie comprend l'ensemble des méthodes qui consistent à déduire le spin des résonances des propriétés des processus de capture radiative et de fission qui peuvent dépendre de l'état du spin: transitions partielles de capture radiative, largeurs totales de capture radiative, interférence entre résonances de même spin due aux voies de sortie du phénomène de fission, largeurs de fission, distribution des produits de fission selon la masse, nombre de neutrons émis par fission, probabilité de fission ternaire et spectre en énergie des rayons α de long parcours, anisotropie des fragments de fission induite par des neutrons non polarisés dans des noyaux alignés, etc.

L'auteur présente et discute plusieurs résultats expérimentaux relatifs à ces diverses méthodes, particulièrement pour ²³⁵U et ²³⁹Pu. Pour ce dernier noyau, un ensemble assez cohérent des valeurs du spin peut être obtenu par plusieurs méthodes indépendantes.

1. INTRODUCTION

La plupart des noyaux fissiles (²³³U, ²³⁵U, ²³⁹Pu, ²⁴¹Pu,...) ont un nombre pair de protons et un nombre impair de neutrons. Par capture d'un neutron lent on obtient un noyau composé pair-pair dont l'énergie d'excitation est suffisamment élevée (à cause de l'énergie d'appariement de deux neutrons) pour permettre la désexcitation par fission. Comme le noyau cible a obligatoirement un spin $\frac{1}{2}$ entier, I, les résonances induites par des neutrons s appartiennent aux deux états de spin différents $J = (I) + \frac{1}{2}$ et $J = (I) - \frac{1}{2}$.

L'analyse et l'interprétation des résultats expérimentaux sont considérablement entravées par le mélange des deux états de spin. En particulier, la théorie des voies de sortie du phénomène de fission ne pourra MICHAUDON

être pleinement comprise que lorsque l'état de spin d'un grand nombre de résonances pourra être déterminé expérimentalement. C'est la raison pour laquelle plusieurs mesures ont été tentées (et le sont encore) dans plusieurs laboratoires pour apporter une information dans ce domaine.

Un moyen de tourner la difficulté est d'étudier un noyau fissile pairpair, tel que 232 U, dont toutes les résonances sont de spin $\frac{1}{2}$.

Mais la durée de vie très courte de cet isotope rend la mesure particulièrement délicate. Elle a cependant été réussie, mais pour un petit nombre de résonances, en fission [1] et en transmission [2].

Nous ignorerons cet aspect du sujet, qui n'entre pas tout à fait dans le cadre de ce mémoire, pour nous consacrer à la détermination du spin des résonances des principaux noyaux fissiles pairs-impairs dont nous rappelons le spin I, ainsi que les valeurs possibles du spin J et du facteur statistique g des résonances induites par des neutrons s:

233U	$I = \frac{5}{2} +$	$J_1 = 2 +$	g ₁ = 5/12
		J ₂ = 3+	g ₂ = 7/12
²³⁵ U	$I = \frac{7}{2} -$	$J_1 = 3 -$	g ₁ =7/16
		$J_2 \approx 4 -$	$g_2 = 9/16$
²³⁹ Pu	$I = \frac{1}{2} +$	$J_1 \approx 0^+$	$g_1 = \frac{1}{4}$
		$J_2 = 1 +$	$g_2 = \frac{3}{4}$
²⁴¹ Pu	$I = \frac{5}{2} +$	J ₁ = 2+	g ₁ = 5/12
		J ₂ = 3+	g ₂ = 7/12

Un peu arbitrairement, mais pour faciliter la présentation, nous séparerons les diverses méthodes de détermination du spin en deux grandes familles: les méthodes dites directes et les méthodes indirectes.

2. METHODES DIRECTES DE DETERMINATION DU SPIN

2.1. Transmission de neutrons polarisés à travers des noyaux polarisés

C'est probablement la méthode la plus directe. Elle est schématisée sur la figure 1. Pour des neutrons d'énergie E, elle consiste à effectuer deux taux de comptage, l'un lorsque les vecteurs de polarisation des neutrons (\vec{P}_n) et des noyaux (\vec{P}_N) sont parallèles, l'autre lorsqu'ils sont antiparallèles. Si l'on appelle I_P et I_A respectivement les deux taux de comptage ainsi obtenus, on peut en déduire le rapport d'asymétrie

E

$$=\frac{\mathbf{I}_{\mathbf{P}}-\mathbf{I}_{\mathbf{A}}}{\mathbf{I}_{\mathbf{P}}+\mathbf{I}_{\mathbf{A}}}$$
(1)

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FIG.1. Détermination du spin des résonances par polarisation

qui devient positif au voisinage d'une résonance de spin $J: I - \frac{1}{2}$ et négatif pour $J: I + \frac{1}{2}$.

Si cette méthode est particulièrement simple dans son principe, elle est par contre extrêmement difficile à appliquer dans la réalité. La production des faisceaux polarisés de neutrons de résonances est actuellement réalisée, soit par diffraction cristalline sur un cristal de Fe-Co qui donne également un faisceau monochromatique [3], soit par transmission d'un faisceau de neutrons non polarisés à travers une cible de protons polarisés [4]. Une autre méthode a également été proposée par Dabbs et al. pour produire un faisceau de neutrons ralentis et polarisés avec un accélérateur linéaire d'électrons [5]. La polarisation des noyaux est plus difficile: elle est donc dans un état beaucoup moins avancé. Le travail de pionnier en la matière a été accompli par Sailor pour la polarisation tant des neutrons que des noyaux. Le seul noyau fissile qui ait été polarisé jusqu'à présent est ²³⁵U, pour lequel des résultats encourageants ont été obtenus [6]. La radioactivité α des corps fissiles constitue une des difficultés de la polarisation, dans la mesure où la chaleur dégagée gêne la descente aux très basses températures (de l'ordre de 0,01°K) indispensable à l'obtention d'une bonne polarisation. Dans le cas de 235 U. l'échantillon a dû être appauvri en ²³⁴U, responsable de la plus grande partie de la radioactivité α .

Nous avons reproduit sur la figure 2 les premiers résultats obtenus par Sailor [6] avec de l'uranium sous la forme UCl₃ dilué dans un composé non magnétique et isomorphe (LaCl₃). La polarisation des noyaux d'uranium est de 10% environ; celle des neutrons est voisine de 100%. La mesure a été conduite de 0,075 eV à 2,04 eV. Elle comprend donc trois résonances à 0,28 eV, 1,14 eV et 2,05 eV. Il apparaît clairement sur la figure 2 que les deux résonances à 0,28 eV et 2,05 eV ont un spin différent de celle à 1,14 eV. (Sur la figure 2, Ip et I_A se réfèrent au vecteur champ magnétique produisant la polarisation des noyaux et non au vecteur polarisation des noyaux \vec{P}_N .) De plus, l'analyse des mesures à basse énergie (E < 0,15 eV) montre que la section efficace dans cette gamme d'énergie est un mélange des deux états de spin. Cependant, l'attribution exacte de l'état de spin à chaque résonance dépend de la



FIG. 2. Effet d'asymétrie observé par transmission d'un faisceau de neutrons polarisés à travers un échantillon de ²³⁵ U polarisé (extrait de [6]).

connaissance du signe de l'interaction hyperfine dans UCl₃ et de celui du moment de 235 U.

S'ils sont tous deux négatifs, alors les résonances à 0, 28 eV et à 2,05 eV sont de spin 3-, alors que celle à 1,12 eV est de spin 4-. Sailor a l'intention de reprendre ces mesures sur HFBR, non seulement sur UCl₃ mais aussi sur d'autres composés de l'uranium, afin de lever le doute sur l'attribution absolue du spin [7].

Ce genre de mesures va donc se poursuivre à plus haute énergie avec le faisceau de neutrons polarisés de HFBR. Il n'y a pas d'objection de principe à ce qu'elles soient étendues aux faisceaux de neutrons ralentis et polarisés de sources pulsées.

Par contre, il ne semble pas que l'on puisse l'appliquer dans le proche avenir aux autres noyaux fissiles à cause de la difficulté de les polariser.

2.2. Analyse des résonances sans mesure de diffusion élastique

Les paramètres d'une résonance d'un noyau fissile sont

l'énergie E_0 les largeurs partielles Γ_n , Γ_γ , Γ_f $(\Gamma_r + \Gamma_r + \Gamma_f)$

$$(1_{n} + 1_{\gamma} + 1_{f} = 1)$$

le facteur statistique $g = \frac{2J+1}{2(21+1)}$

Nous ignorons ici les largeurs partielles de transition radiative de même que les largeurs partielles de fission correspondant à un mode par-

(2)

ticulier de fission et nous supposons que l'analyse à un niveau est significative.

Deux mesures au moins sont nécessaires pour connaître toutes les largeurs partielles; par exemple, une mesure de fission et une de transmission. L'analyse de cette dernière donne $2 g \Gamma_n$ et Γ . De la mesure de fission on peut déduire $\sigma_0 \Gamma_f$ et, dans des conditions favorables, Γ (largeur déjà donnée par l'analyse de la transmission). On connaît alors $2 g \Gamma_n$, Γ , Γ_f et la quantité $\Gamma_{\gamma+n} = \Gamma_{\gamma} + \Gamma_n$ par la différence $\Gamma - \Gamma_f$.

Pour la plupart des noyaux fissiles, le spin est élevé (donc les deux valeurs possibles de g sont très voisines) et la largeur neutronique est relativement faible.

$$\frac{\langle 2 g \Gamma_n^0 \rangle}{\langle \Gamma \rangle} = 1, 3 \cdot 10^{-3} \text{ pour } {}^{235}\text{U et } 1, 7 \cdot 10^{-3} \text{ pour } {}^{239}\text{Pu}$$

Dans la plupart des cas, la valeur de g ne peut donc pas être extraite de ces mesures, et l'on fait couramment l'approximation $2 g \Gamma_n \# \Gamma_n$. La seule exception est ²³⁹Pu, dont les paramètres de certaines résonances ne sont compatibles qu'avec une seule valeur de g (sans faire l'hypothèse particulière sur les propriétés statistiques de Γ_y).

Exemple:	Résonance à 74,95 eV
	$2 g \Gamma_n = 33, 20 meV$
	Γ = 147 ± 14 meV
	$\Gamma_f = 84 \text{ meV}$

g = 3/4 donne Γ_{γ} = 40 meV et g = 1/4 donne Γ_{γ} = -4 meV, donc, obligatoirement g = 3/4 [8].

Cette méthode n'est valable que dans des conditions favorables qui ne sont que rarement satisfaites. Si on veut l'appliquer à un plus grand nombre de cas, il faut la compléter par une mesure de diffusion élastique.

2.3. Analyse des résonances avec une mesure de diffusion élastique

La mesure de diffusion élastique est indispensable lorsque les deux valeurs fissiles de g sont voisines et (ou) lorsque les largeurs neutroniques des résonances sont faibles, c'est-à-dire pratiquement dans tous les cas, exception faite de quelques résonances de ²³⁹Pu (paragraphe 2.2). Cependant, cette méthode se heurte à certaines difficultés comme nous le verrons plus loin.

La mesure consiste, soit à déterminer la surface A = $(\pi/2)\sigma_{0n}\Gamma$ de la résonance de diffusion, soit à mesurer directement la section efficace de diffusion σ_{0n} au-dessus de la section efficace potentielle, à l'énergie de la résonance.

Connaissant A, on en déduit le facteur statistique g par la relation simple

$$g \simeq \frac{(2 g \Gamma_{\rm n})^2}{A \times \Gamma} \tag{3}$$

Les quantités $2 g \Gamma_n$ et Γ sont obtenues par analyse de la résonance en transmission. On remarque que, dans ce cas, il n'est pas indispensable

			Sp	in J			P .		To an effe	Sp	in J	
(eV)	Vogt [39]	Fraser [9]	Sauter [11]	Asghar [10]	Cowan [38]	Derrien [8]	(meV)	(eV)	(eV)	Asghar [10]	Derrien [8]	(meV)
-1, 2	J ₁ a		·						90, 75		le	17
0,3	J2a						60		96, 49		0d	1670
7,82	J ₁ a	1	1				47		100, 25		0 d	6000
10, 93	J ₁ a	1	1				143		103	1		13
11, 5						0d	~ 500		105, 3	0		6
11, 89		1	1				24		106,67	(1)	1 ^f	26
14, 31	1		1 ^a				67		116, 03	0		215
14, 69		0	1	1			30		118,83	0	1e	43
15, 42			0a		(1)	0d	. 650		126, 2	0		ł
17,66	j	1	1	1	1		34		131, 75	0	0d	3300
22, 28		0	1	1	1		62		133, 78	1		7
26,29				0	1		55		136, 75	0		88
32, 38			0p		(0)		110		146, 25	1	(0) ^f	13
35, 43			1 ^b				5		147, 44		0q	1000
41, 42	1			_	-		3		157,08	0	0d	630
41, 66	1	1 ^c	1¢	1 ^c	1 ^C		54		164, 54	1	1e	8
44, 48	ļ	0	1	1	1		5		167, 1	0	(1) ^f	74
47, 60]		0	0	1	Į	240		171, 08		0d	1000

TABLEAU I. 1	DETERMINATION DU	SPIN DES	RESONANCES	DE 239	Ρu
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49,71					0d	690	177,22	1		5
50, 08			1	1		12	184, 87		0d	~1500
52, 60		1	1	1		9				
57,44	1			0	0d	~ 500	196, 7	0		59
58, 84				0	0d	~1100	199, 4	0	1 ^e	90
59, 22			0	1		133	207, 37	1	(1) ^f	7
60,94				0	0d	~6000	211, 1		0d	. 800
65, 71			1	1	(1) ^f	74	216,5	0	(0) ^f	10
66,57				0			220, 2		1f	4
74,05			1			32	223, 2	0		~ 2
74, 95		1	1	1	le	84	227,8		0d	6000
81,76				0	0d	~2000	231, 40	1	1 ^e	4
						<u> </u>	234, 3	0		14
							239, 1	0	(0) ^f	17
						242, 9	0	1 ^f	58	
							248, 86	1	1e	6
^a Spin détermi	'Spin déterminé entièrement ou partiellement par une analyse multiniveaux.						251, 2	1	1e	14
 Voir le commentaire dans le texte. Résonance double, non résolue dans la mesure du spin. Spin 0 attribué d'après la grande valeur de Γf. L'autre valeur du spin conduit à une valeur impossible ou très improbable de Γγ. Spin déterminé en supposant que la largeur Γγ est constante et indépendante du spin. L'énergie des résonances est celle donnée par Asghar. 							262, 5 ^g	0		
							273, 6 ^g	1		
						276, 9 ^g	0			
						281 ^g	0			
							284, 1 ^g	1		
							299, 78	1		

de disposer d'une autre section efficace partielle (fission, capture,...). De même, on peut déduire g des quantités σ_{0n} , $2g\Gamma_n$ et Γ .

La mesure de la diffusion élastique est particulièrement difficile dans le cas des noyaux fissiles. Non seulement il faut que le détecteur de neutrons diffusés soit peu sensible aux rayons γ (comme dans le cas des noyaux non fissiles), mais il faut également qu'il le soit peu aux neutrons de fission. Ces deux conditions sont d'autant plus sévères que les largeurs neutroniques des noyaux fissiles sont faibles devant les largeurs totales. Le cas le plus favorable est celui de ²³⁹ Pu pour les raisons déjà citées (plus grande valeur relative de Γ_n , et surtout deux valeurs possibles de g qui sont dans le rapport 3). De plus, la mesure de diffusion donnant un très faible taux de comptage et, de par sa nature, étant à mauvaise résolution, la séparation des résonances est malaisée, ce qui favorise encore ²³⁹Pu dont les résonances sont assez espacées.

Une première mesure a été tentée en 1961 par Fraser et Schwartz [9] auprès de l'accélérateur linéaire de Harwell avec une distance de vol de 14,6 m et en utilisant un banc de compteurs à BF3 comme détecteur de neutrons diffusés. La contribution des neutrons de fission a été évaluée en intercalant un fourreau de carbure de bore entre l'échantillon et le détecteur et en éliminant ainsi la contribution des neutrons diffusés. Malgré un bruit de fond très important et une contribution substantielle des neutrons de fission, Fraser et Schwartz ont pu déterminer le spin de huit résonances (7, 8 eV, 10, 9 eV, 11, 9 eV, 14, 7 eV, 17, 6 eV, 22, 2 eV, 41, 4 eV, 44, 5 eV) (voir tableau I).



FIG.3. Mesure de la section efficace de diffusion élastique de 239 Pu (extrait de [11])

Cette mesure a été reprise récemment à Harwell [10] avec une meilleure résolution (distance de vol 50 m) et avec un autre détecteur composé de plusieurs verres au lithium enrichi en ⁶Li. Une double correction à dû être appliquée, l'une pour la contribution des rayons γ , évaluée grâce à un verre au lithium naturel, l'autre pour les neutrons de fission, grâce à un cristal de stilbène. Par cette méthode, le spin d'un grand nombre de résonances a été déterminé jusqu'à 300 eV (tableau I).

Un moyen d'éliminer la contribution gênante des rayons γ et des neutrons de fission est d'utiliser la «bright line method» dans laquelle la séparation des événements provoqués par les neutrons diffusés des événements indésirables se fait par temps de vol (fig. 3). Cette méthode a été employée avec succès auprès de l'accélérateur linéaire de Livermore [11]. Si cette méthode donne des résultats moins entachés de phénomènes parasites, la résolution est néanmoins élargie par deux causes importantes:


FIG. 4. Comparaison des différents effets de résolution dans la méthode de Sauter et Bowman [11]



FIG. 5. Forme de la section efficace de diffusion au voisinage d'une résonance isolée

a) Le temps de ralentissement dans la boule de graphite tapissée d'une couche de polyéthylène à sa surface intérieure. A lui seul il correspond à une résolution en temps dont la largeur totale à mi-hauteur est de $\Delta t(\mu s) = 3$ [E (eV)]⁻¹, soit trois fois supérieure à celle d'une expérience classique.

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b) Le recul du noyau dans l'échantillon, qui se répercute directement sur le temps de vol du neutron par une fonction de résolution rectangulaire de largeur égale à E/(A+1). Elle varie en fonction de l'énergie comme le temps de ralentissement.

Dans cette mesure [11] ces effets sont tous les deux supérieurs à l'effet Doppler (fig. 4), ce qui impose une limite supérieure en énergie de 100 eV environ pour ²³⁹Pu. Les résultats de l'attribution du spin sont consignés dans le tableau I. Les trois mesures citées se réfèrent à ²³⁹Pu.

Très peu de résultats se rapportent à 235 U. Un mémoire est présenté à cette Conférence [12], qui donne les spins des résonances à 8,8 eV et 12,4 eV d'après la mesure de σ_{0n} . Nous ne disposons d'aucun résultat pour les autres noyaux fissiles.

Examinons rapidement quelles sont les conditions à remplir pour que ce type de mesure puisse conduire à des résultats significatifs.

La forme d'une résonance de diffusion élastique, compte tenu du terme d'interférence résonnant-potentiel, est représentée sur la figure 5. Deux conditions doivent être remplies:

a) La valeur de σ_{0n} , à l'énergie de la résonance, doit être suffisante comparée à la section efficace potentielle. On peut admettre un peu arbitrairement le critère $\sigma_{0n}/\sigma_p \ge 0, 1$.

b) Le terme d'interférence ne doit pas être trop important, sinon la résonance est très dissymétrique, ce qui peut fausser grossièrement l'analyse. Il s'agit soit de déterminer la section efficace de diffusion à l'énergie de la résonance, et alors elle est très sensible à la moindre erreur sur l'énergie, soit d'une analyse de surface qui doit être faite par différence de deux surfaces, l'une au-dessous, l'autre au-dessus de la section efficace potentielle.

On peut, là aussi assez arbitrairement, choisir le critère que le rapport A/B soit supérieur à une certaine quantité, 2 par exemple. Or, le minimum et le maximum de la section efficace élastique, pour une résonance isolée, sont égaux à [13]

$$\sigma_{\min} = \pi \lambda^2 \frac{(2J+1)}{2(2I+1)} \left\{ \left[1 + \left(1 - \frac{\Gamma_n}{\Gamma} \right)^2 - 2 \left(1 - \frac{\Gamma_n}{\Gamma} \right) \cos 2kR \right]^{\frac{1}{2}} - \frac{\Gamma_n}{\Gamma} \right\}^2$$
(4)

$$\sigma_{\text{Max}} = \pi \lambda^2 \frac{(2J+1)}{2(2I+1)} \left\{ \left[1 + \left(1 - \frac{\Gamma_n}{\Gamma} \right)^2 - 2 \left(1 - \frac{\Gamma_n}{\Gamma} \right) \cos 2kR \right]^{\frac{1}{2}} + \frac{\Gamma_n}{\Gamma} \right\}^2$$
(5)

$$A = \sigma_{Max} - \sigma_p \text{ et } B = \sigma_p - \sigma_{min}$$
 (6)

Le rapport A/B dépend uniquement de Γ_n^0/Γ et la variation de A/B en fonction de Γ_n^0/Γ est portée sur la figure 6. On constate que A/B>2 pour $\Gamma_n^0/\Gamma > 1, 4 \cdot 10^{-3}$.

On constate de même que $\sigma_{0n}\,/\sigma_p$ ne dépend que du même rapport Γ_n^0/Γ par la relation

$$\frac{\sigma_{0n}}{\sigma_{p}} = \sigma_{0} \frac{\Gamma_{n}}{\Gamma} \cdot \frac{1}{4\pi\lambda^{2}\sin^{2}k^{2}R} \not\# 2, 5 \cdot 10^{5} g \left(\frac{\Gamma_{n}^{0}}{\Gamma}\right)^{2}$$
(7)

d'où



FIG. 6. Importance de l'effet d'interférence potentiel-résonnant en diffusion élastique

En faisant, dans ce cas, l'approximation $g = \frac{1}{2}$ (ce qui est valable puisque nous ne faisons ici qu'un calcul approché), on constate que

$$\frac{\sigma_{0n}}{\sigma_p} > 0, 1 \text{ pour } \frac{\Gamma_n^0}{\Gamma} > 10^{-3}$$
(8)

On retrouve donc une condition très voisine de la précédente. Limitons-nous au cas ²³⁹Pu et calculons la probabilité $P(\alpha)$ que $\Gamma_n^0/\Gamma < \alpha$ (α est un nombre arbitraire) pour les états de spin 0 et 1. Un mémoire présenté à cette Conférence [8] suggère que

$$<\Gamma_{f}>=42$$
 meV pour J=1 et $<\Gamma_{f}>=1300$ meV pour J=0

La probabilité P(α), en fonction de α , est portée sur la figure 7 pour les deux états de spin, avec $< 2g \Gamma_n^{0} > = 0, 61 \text{ meV}$ et $\Gamma_{\gamma} = 40 \text{ meV}$. (Pour simplifier le calcul, il a été supposé que la distribution des largeurs de fission suivait une distribution en X² avec $\nu = 2$ degrés de liberté.)

Si l'on applique le critère $\Gamma_n^0/\Gamma < 10^{-3}$ à ces résonances (cela revient à évaluer la proportion des résonances qui ne peuvent pas être analysées en diffusion), le diagramme de la figure 7 donne

$$P_{(10^{-3})} = 0, 38 \text{ pour } J = 0$$
 (9)
 $P_{(10^{-3})} = 0, 08 \text{ pour } J = 1$ (10)



FIG.7. Probabilité $P(\alpha)$ pour que $\Gamma_n^0/\Gamma < \alpha$ (α quantité donnée) $A \rightarrow \Gamma_\gamma = 40 \text{ meV}, < \Gamma_n^0 > = 1,23 \text{ meV}, < \Gamma_f^> = 1300 \text{ meV}, résonances de spin 0 (?)$ $B \rightarrow \Gamma_\gamma = 40 \text{ meV}, < \Gamma_n^0 > = 0,41 \text{ meV}, < \Gamma_f^> = 46 \text{ meV}, résonances de spin 1 (?)$

Le rapport du nombre des résonances pour les deux états de spin qui peuvent être analysées en diffusion est

$$\frac{N(j=1)}{N(j=0)} = \frac{3}{1} \times \frac{1-0,08}{1-0,38} = 4,5$$
(11)

Cet effet accentue encore le rapport des populations entre les deux états de spin. Or, cela ne se retrouve pas dans les résultats d'Ashgar [10] entre 100 eV et 300 eV. Dans cette gamme d'énergie, Ashgar attribue le spin 0 à 19 résonances contre 12 résonances au spin 1. Le rapport (11) tel qu'il est observé est donc de 0, 63 au lieu de 4, 5, soit une différence d'un facteur 7. Nous reviendrons sur ce point plus tard. Cependant, nous pouvons remarquer dès maintenant que la sous-estimation du terme d'interférence conduit à une surestimation de la surface A de la résonance et donc à une diminution apparente du facteur statistique g. Cela reviendrait, dans certains cas, à donner le spin 0 à des résonances de spin 1.

Dans la gamme d'énergie en dessous d'une centaine d'eV, nous pouvons comparer trois mesures [9-11] (voir tableau I). On constate généralement un bon accord entre les mesures d'Ashgar et de Bowman, mais un moins bon accord entre ces deux dernières et celles de Fraser (notamment à 14,69 eV, 22,28 eV et 44,48 eV). Cependant, même lorsque l'accord est bon, il appelle quelques remarques:

- Le spin est quelquefois donné lorsque la résonance est double (p.ex. à 41,42 eV).

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- Bowman donne le spin de la résonance à 15, 42 eV d'après l'étude de l'interférence en fission et non d'après la mesure de diffusion. D'autre part, les spins des résonances à 32, 38 eV et 35, 49 eV sont attribués d'après la combinaison des paramètres qui donnent à ces deux résonances une surface à peu près égale en diffusion. Or, elles n'apparaissent que faiblement et elles sont fortement perturbées par le terme d'interférence. Quant à la résonance à 14, 41 eV elle n'apparaît pas dans cette mesure, mais cette propriété est quand même utilisée pour lui donner le spin 1 (confirmé par une étude de l'interférence dans la section efficace de fission).

- En tenant compte des attributions de spin pour 11 résonances jusqu'à 75, 3 eV, on arrive ainsi à 10 résonances de spin 1, contre une de spin 0. Ce rapport de 10 que l'on trouve ainsi, qui est assez incertain, est encore plus élevé que celui que nous avons calculé plus haut (formule 11).

Le critère fondé sur la valeur de Γ_n^0/Γ est encore plus sévère pour les noyaux fissiles autres que ²³⁹Pu.

2.4. Transitions de capture radiative

Cette méthode est utilisée couramment pour la détermination du spin des noyaux non fissiles par l'application des règles de sélection pour les transitions de l'état résonnant aux premiers états excités du noyau composé. Dans le cas des noyaux fissiles, elle n'a pas encore été employée à cause de la contribution des rayons γ et des neutrons de la fission, ainsi que de la radioactivité naturelle des échantillons. L'augmentation de l'intensité des sources de neutrons pulsées, d'une part, permettait à la fois l'emploi d'échantillons minces et de dispositifs d'anticoïncidence avec le phénomène de fission, et les nouveaux détecteurs au Ge, d'autre part, devraient étendre cette méthode aux noyaux fissiles. Pour ²³⁵U, noyau favorable à cause de sa parité négative, la transition électrique dipolaire au premier niveau excité 2+ est autorisée à partir d'une résonance 3- mais ne l'est pas lorsque le spin est 4-.

3. METHODES INDIRECTES DE DETERMINATION DU SPIN

Par méthodes indirectes, nous entendons celles qui font intervenir des propriétés qui sont supposées dépendre de l'état de spin. Nous allons en examiner quelques-unes.

3.1. Largeurs totales de capture radiative

Dans certains noyaux non fissiles, la valeur moyenne de Γ_{γ} dépend de l'état de spin (p. ex.: ⁷⁷Se, ¹⁹⁹Hg, ²⁰¹Hg, ...) [14]. Il est possible que ce phénomène se retrouve dans les noyaux fissiles, ainsi que l'avait déjà suggéré Wheeler [15]. Des fluctuations importantes de Γ_{γ} ont été observées dans ²³⁵U [16], qui peuvent être expliquées de cette façon, mais qui ne sont pas une preuve. En se référant aux travaux de Sailor [6] que nous avons cités plus haut, on constate que

 $<\Gamma_{\gamma}>$ = 32 meV pour les résonances à 0, 28 eV et 2, 05 eV (supposées 3-)

 Γ_{γ} = 43 meV pour la résonance à 1,14 eV (supposée 4-)

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Par contre, les résultats présentés par Poortmans et al. [12] ne semblent pas montrer une différence de Γ_{γ} pour les résonances à 8,8 eV et 12,4 eV de ²³⁵U qui sont mentionnées comme étant de spins différents. Des travaux supplémentaires sont nécessaires pour conclure sur ce point.

Fraser et Schwartz [9] avaient suggéré une valeur de Γ_{γ} différente suivant l'état de spin dans ²³⁹Pu, en se fondant sur la résonance à 41, 4 eV qu'ils trouvaient de spin égal à 1, ce qui conduisait à Γ_{γ} = 62, 9 meV [17] (au lieu de Γ_{γ} = 40 meV). En fait, cette résonance est double [8, 18], ce qui jette le doute sur la détermination du spin, et la nouvelle valeur de Γ_{γ} est plus basse (Γ_{γ} = 43, 5±8 meV) [8].

Pour conclure dans le cas de 239 Pu, il faudrait connaître plusieurs résonances de spin 0 qui soient étroites pour que la détermination de Γ_{γ} soit précise. Or, un calcul rapide montre que si l'on impose les deux conditions

$$\frac{\Gamma_{\rm n}^0}{\Gamma} > 10^{-3} \text{ et } \Gamma_{\rm f} \leqslant \Gamma_{\rm y} = 40 \, {\rm meV} \qquad (12)$$

pour les résonances larges supposées être de spin 0, on ne retient que 2 a 3% environ d'entre elles. Avec cette hypothèse, il est donc très peu probable de pouvoir mesurer une résonance étroite de spin 0, et donc de savoir si Γ_{Y} dépend de l'état de spin.

Si l'on admet alors que Γ_{γ} fluctue peu autour de la valeur moyenne de 40 meV, il est possible d'attribuer un plus grand nombre de spins par la méthode des paramètres de résonances (voir paragraphe 2.2 et tableau I). Il est intéressant de comparer les valeurs de spin ainsi obtenues à celle d'Asghar [10] entre 100 et 300 eV. On constate plusieurs désaccords, toujours dans le même sens: une résonance de spin 1 à Saclay devient une résonance de spin 0 à Harwell, ce qui confirmerait l'hypothèse que nous avons avancée au paragraphe 2.3 relative à un excès de résonances de spin 0 dans les mesures d'Asghar.

3.2. Interférences entre résonances

Seules peuvent interférer entre elles des résonances de même spin lorsque le processus de désexcitation comporte un faible nombre de voies de sortie (diffusion élastique, transitions individuelles de capture radiative, fission). Dans certains cas, des dissymétries de résonances peuvent être expliquées par ce phénomène d'interférence, ce qui permet de conclure que certaines résonances sont du même état de spin sans pouvoir toujours préciser lequel.

Cette propriété est utilisée dans le cas de la diffusion élastique [19] pour des noyaux non fissiles. Elle ne peut pas être appliquée aux noyaux fissiles (trop faible valeur relative de Γ_n / Γ) sauf peut-être à quelques résonances de ²³⁹Pu lorsque la résolution le permettra. L'interférence entre transitions partielles de capture radiative est aussi inapplicable aux noyaux fissiles pour le moment.

Par contre, l'interférence due au faible nombre de voies de sortie du phénomène de fission a été utilisée à plusieurs reprises avec un formalisme multiniveaux à 235 U [20-22], à 233 U [23, 24], à 241 Pu [25] et à 239 Pu. C'est ainsi que Sauter et Bowman déterminent le spin de trois résonances de 239 Pu entre 13, 3 eV et 16, 5 eV à partir d'une analyse



FIG. 8. Représentation schématique des états collectifs de transition d'un noyau pair-pair

multiniveaux de la section efficace de fission de ²³⁹Pu dans cette gamme d'énergie [11]. Cependant, cette méthode appelle quelques réserves:

- Il n'est pas possible de l'étendre à un grand nombre de résonances car le nombre de paramètres, donc de combinaisons de paramètres, augmente très rapidement. Même avec les calculatrices électroniques très puissantes, il est douteux qu'une méthode d'ajustement de la courbe théorique aux résultats expérimentaux par moindres carrés (avec choix des paramètres par la calculatrice) soit possible pour le moment.

- La solution obtenue n'est généralement pas unique (ce qui interdit l'attribution unique du spin).

- Les dissymétries des résonances peuvent non seulement être dues à des effets d'interférence mais aussi à des résonances masquées dans les résultats expérimentaux, et il est difficile de trancher entre les deux effets. Dans ²³⁵U, il est vraisemblable que l'on manque environ 20% des niveaux [16], dont la majorité resteront indétectables malgré l'amélioration de la résolution. En effet, ils sont masqués surtout par la largeur naturelle des résonances et un résidu d'effet Doppler qu'il est impossible d'éliminer même à très basse température.

Ces réserves sont justifiées par le fait que le classement des résonances effectué de cette façon par Vogt [21] et Shore est en désaccord avec les résultats de Sailor et al. [6] obtenus par polarisation (voir paragraphe 2.1). Par contre ces derniers sont en meilleur accord avec ceux de Kirpichnikov et al. [22].

En résumé, cette méthode ne doit être appliquée qu'avec précaution et ne peut donner des résultats sûrs que dans quelques cas isolés, pour un nombre restreint de résonances.

3.3. Propriétés du phénomène de fission

Le fil directeur reste toujours la théorie de Bohr [26] suivant laquelle, au point seuil, la plus grande partie dè l'énergie d'excitation se trouve

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FIG. 9. Distribution des largeurs de fission Γ_f de ²⁴¹Pu

sous la forme d'énergie de déformation. Le système ne peut donc exister que sous la forme de quelques états quantiques, dits états de transition, dont la spectroscopie est voisine de celle des premiers états excités du noyau. Ces états sont collectifs et chacun d'eux est caractérisé par les nombres quantiques I (spin), π (parité) et K (projection du spin I sur l'axe de symétrie). Nous reproduisons sur la figure 8 un diagramme schématique de ces états collectifs de transition tels qu'ils peuvent apparaître dans un noyau lourd pair-pair ayant une déformation quadrupolaire stable [27].

Les conséquences de cette théorie pour la fission induite par des neutrons de résonances ont été exposées à plusieurs reprises et récemment par Griffin [28] et Rae [29]. Nous n'allons pas reprendre cette question dans le détail, ce qui sortirait du cadre de ce mémoire, mais simplement nous attacher à en rappeler quelques aspects.

3.3.1. Différence possible du seuil de fission pour les deux états de spin

La différence doit surtout être marquée pour les isotopes fissiles à parité positive, où l'un des états de spin appartient à la bande de rotation de l'état fondamental qui est la plus basse. Les largeurs moyennes de fission peuvent donc être différentes pour les deux états de spin.

Cependant, du fait que les états de transition sont peu nombreux, les fluctuations des largeurs Γ_f sont importantes, d'où la difficulté de faire apparaître deux valeurs différentes de $<\Gamma_f >$ et (si elles le sont) d'en déduire le spin des résonances.





Dans ²⁴¹Pu [30], où il est fait état de deux groupes de résonances, les unes larges, les autres étroites, la distribution des largeurs Γ_f ne montre pas une telle séparation en deux groupes (fig. 9).

Dans le cas de ²³⁵U, où des fluctuations de Γ_{γ} ont été observées, on trouve à Saclay une corrélation positive entre Γ_{f} et Γ_{γ} [16], ce qui suggère deux familles pour les largeurs de fission. Mais, d'une part, ce résultat n'est pas retrouvé par le groupe de Dubna [31], d'autre part, le rapport entre les valeurs moyennes de Γ_{f} n'est pas suffisamment important pour permettre l'attribution du spin.

Le seul cas où deux familles apparaissent clairement dans la distribution des largeurs $\Gamma_{\rm f}$ est celui de ²³⁹Pu [8]. Nous reproduisons cette distribution sur la figure 10.

Les résonances de spin 1 mesurées en diffusion (en dessous de 76 eV) peuvent toutes appartenir à la famille des résonances étroites, ce qui correspond à la théorie de Bohr. Cette même théorie prédit que les résonances larges sont de spin 0. Dans cette hypothèse, les faibles valeurs de Γ_f correspondent à un mélange de résonances de spin 0 et de spin 1, avec une majorité de ces dernières. Par contre, les grandes largeurs de Γ_f ($\Gamma_f > 400$ meV) correspondent essentiellement à des résonances de spin 0. Cela constitue donc un critère simple d'attribution du spin 0. Nous reviendrons sur ce point.

3.3.2. Distribution en masse et énergie cinétique des fragments de fission

Il a été suggéré par Wheeler [32] que la distribution en masse des produits de fission pourrait dépendre du spin J de la résonance. La

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FIG. 11. Variation de la probabilité de fission symétrique dans les résonances de ²³⁹Pu (extrait de [38])

mesure du rapport de la fission symétrique à la fission asymétrique en fonction de l'énergie a été faite à plusieurs reprises, surtout pour 235 U et 239 Pu, l'effet le plus important ayant été trouvé pour 239 Pu,

Un grand nombre de travaux ont été poursuivis dans ce domaine, dont plusieurs ont été présentés à la Conférence de Salzbourg (mars 1965)¹. Nous allons examiner quelques-uns d'entre eux.

La mesure la plus complète sur la distribution en masse des produits de fission dans les résonances de ²³⁵ U a été faite par Cowan et al. [33] en utilisant un explosif nucléaire comme source de neutrons. Le rapport de l'activité de ¹¹¹Ag à celle de ⁹⁹Mo a pu être obtenu pour 17 résonances de 8, 8 eV à 40 eV. Ce rapport accuse des variations extrêmes de +22% à -50% comparées à la valeur mesurée pour la fission induite par des neutrons thermiques. Il est possible de séparer les résonances en deux groupes, quatre dans le premier groupe (rapport ¹¹¹Ag/⁹⁹Mo plus élevé) et 13 dans le second (rapport ¹¹¹Ag/⁹⁹Mo plus faible). La valeur moyenne $<\Gamma_f>$ pour ces deux groupes n'est pas très différente ($<\Gamma_f >= 63$ meV pour le premier groupe, $<\Gamma_f > = 52$ meV pour le second). Cela confirmerait que les deux seuils de fission pour les états de spin 3- et 4- sont voisins dans ²³⁵U.

Il a été montré expérimentalement [34, 35] et justifié théoriquement [36] que la distribution en masse des produits de fission était reliée à l'énergie cinétique totale de ces fragments. C'est en vue d'étendre ces résultats aux neutrons de résonances que Melkonian a mesuré les variations de l'énergie cinétique des fragments de fission pour une quinzaine de résonances de ²³⁵U jusque vers 40 eV [37]. Un accord est trouvé avec les résultats de Cowan pour sept résonances. Cet accord va dans le sens espéré puisque aux résonances ayant une faible proportion de fission symétrique (mesures de Cowan) correspondent des résonances dont l'énergie cinétique moyenne des fragments est plus élevée.

Cependant, on ne connaît le spin d'aucune de ces résonances (sauf peut-être celle à 8,8 eV), ce qui ne permet pas de vérifier la théorie et donc de faire confiance à cette méthode pour la détermination du spin.

Le plutonium-239 est un noyau plus favorable pour cette étude. Par la même méthode, Cowan et al. ont entrepris la détermination de la proportion de fission symétrique dans les résonances de ²³⁹Pu [38]. La mesure porte sur une vingtaine de résonances et le rapport des activités de ¹¹⁵ Cd et de ⁹⁹ Mo (voir fig. 11) accuse de grandes variations (les valeurs extrêmes

¹ AGENCE INTERNATIONALE DE L'ENERGIE ATOMIQUE, Physique et chimie de la fission, 2 vol., AIEA, Vienne (1965).

non corrigées sont dans un rapport 3). Après corrections des résonances voisines, les valeurs de ce rapport appartiennent nettement à deux groupes distincts pouvant correspondre aux deux états de spin. Dans cette hypothèse, le groupe ayant la plus grande proportion de fission symétrique correspondrait au spin 0+, l'autre groupe au spin 1+. Le dernier groupe comprend 12 résonances sûres, dont huit ont été mesurées en diffusion par Asghar et Sauter avec des résultats qui concordent. Sur ces huit résonances, une seule est en désaccord avec les résultats de Cowan, celle à 47, 74 eV, pour laquelle Sauter et Asghar trouvent J=0 alors que Cowan trouve J=1. Les résonances de spin J=0 (au nombre de six d'après Cowan) ne sont généralement pas analysées en diffusion, ce qui exclut une comparaison directe. Par contre, on remarque que sur ces six résonances cinq ont une largeur $\Gamma_{\rm f}$ supérieure à 500 meV et la largeur $\Gamma_{\rm f}$ de la sixième est de 180 meV, ce qui correspond très bien à la théorie et à l'hypothèse que nous avons avancée à l'alinéa 3.1.2 suivant laquelle les résonances à grande largeur Γ_f ($\Gamma_f \ge 400$ meV) sont pratiquement toutes de spin 0+.

Les résultats de Cowan sont en bon accord avec ceux de Melkonian [37] sur la variation d'énergie cinétique des fragments de fission.

Par contre, il faut citer un désaccord sur la résonance à 0,3 eV. Suivant l'analyse multiniveaux de Vogt [39] elle serait d'un spin différent des résonances à 7,82 eV et 10,93 eV, donc de spin 0. Or, la probabilité de fission symétrique est faible [40], ce qui au contraire devrait correspondre au spin 1.

Néanmoins, ²³⁹Pu donne donc un ensemble assez cohérent de résultats expérimentaux en accord qualitatif et parfois même quantitatif avec la théorie.

3.3.3. Probabilité de fission ternaire et neutrons de fission

Il a été montré expérimentalement [41, 42] et justifié théoriquement [36] que le nombre de neutrons de fission dépend de la division en masse au moment de la scission. Il en est de même pour l'émission des rayons α de long parcours dans le processus de fission ternaire. Ainsi, à une division en masse déterminée doit correspondre

- un nombre moyen et une distribution des neutrons de fission,
- une probabilité d'émission des rayons α de long parcours et un spectre d'énergie de ces rayons α .

Si la distribution en masse des produits de fission varie de résonance en résonance, alors ces différentes propriétés devraient aussi accuser des variations.

Les mesures faites jusqu'à présent ne montrent pas de variation de $\bar{\nu}$ (nombre moyen de neutrons par fission) d'une résonance à l'autre; elles devraient être reprises avec une meilleure précision.

Le processus de fission ternaire avec émission d'un rayon α de long parcours a été étudié, de résonance en résonance, dans plusieurs laboratoires, notamment à Saclay [43] où l'on a trouvé des variations significatives mais qu'il est difficile de relier au spin des résonances ou aux autres propriétés de capture ou de fission.

Des mesures sur les neutrons de fission et les rayons α de tripartition sont très souhaitées dans les résonances de ²³⁹Pu, là où les effets les plus grands sont attendus.

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	2 (Phc/k=	233 U (Phc/k=0,0277°K)		²³⁵ U 0, 0154°K)
ĸ	2+	3+	3-	4-
0	+ 0,074		+ 0,077	
±1	+ 0,037	+ 0,055	+ 0,058	+ 0,065
± 2	- 0, 037	0	0	+ 0,031
<u>+</u> 3		- 0,092	- 0, 96	- 0,027
± 4				- 0, 108

TABLEAU II. CALCUL DU COEFFICIENT A₂ D'ANISOTROPIE DES FRAGMENTS DE FISSION DANS LE CAS DE 233 U ET 235 U

Note: Ce tableau est extrait de [44].

3.3.4. Anisotropie des fragments de fission induite par des neutrons non polarisés sur des noyaux alignés

Le travail de pionnier dans ce domaine a été accompli par Dabbs et al. [44], qui ont montré que le coefficient A_2 de l'anisotropie des fragments de fission dans 235 U était de signe opposé à celui des rayons de la radioactivité naturelle. Ce rapport peut être calculé pour les différentes bandes des états collectifs (voir tableau II pour 233 U et 235 U, extrait de [44]).

Le rapport A_2 dépend des deux nombres quantiques K et J, mais plus du premier que du second. Il semble donc plus sensible à la voie de sortie du phénomène de fission qu'au spin de la résonance.

Cette méthode ne peut conduire à la détermination du spin que dans des cas favorables, par exemple si la plus grande contribution de fission dans chaque état de spin se trouve dans une bande ayant un K bien déterminé, la valeur de K étant alors différente pour chaque état de spin. En se limitant au cas de 235 U, on peut supposer que dans le noyau composé 236 U la contribution principale du spin 3- se trouve dans la bande de vibration octupolaire ayant K = 0 (la contribution de la bande K = 1 serait alors plus faible) alors que le spin 4- correspondrait uniquement à la bande K = 1.

Dabbs a publié des résultats sur ²³⁵U à basse énergie jusqu'à la résonance à 8, 8 eV. De 0, 14 eV à la résonance à 1, 14 eV, une analyse simple montre que les résultats d'anisotropie des fragments de fission sont en accord avec l'analyse multiniveaux de Kirpichnikov [22], ellemême en accord avec les récents travaux de Sailor et al. sur la polarisation [6] (voir paragraphe 2.1). Par contre, elle est en désaccord avec les analyses multiniveaux proposées antérieurement [20, 21] (ce qui paraît normal, voir paragraphe 3.2). Il existe également d'autres désaccords notés par Dabbs et al. [44], dont la présentation sortirait du cadre de ce mémoire. De ces mêmes résultats de Dabbs et al. il ressort que les résonances à 1, 14 eV et 8, 8 eV ont des valeurs de A_2 qui sont très voisines ($A_2 = 0, 052 \pm 0, 01$ environ) et différentes de celle à 1, 14 eV ($A_2 = 0, 031$ $\pm 0, 002$) ce qui suggère que les deux premières résonances sont du même état de spin, différent de celui de la troisième. Ce résultat n'est pas cohérent avec les valeurs suivantes:

Résonance à 0, 275 eV: spin 3- - Résonance à 1, 14 eV: spin 4- [16] (avec les réserves déjà formulées au paragraphe 2.1)

Résonance à 8,8 eV (spin 3-) [12].

Un désaccord existe donc entre trois déterminations du spin qui devra être examiné sérieusement si Sailor confirme l'attribution des spins qui a été proposée pour les résonances à 0, 27 eV et 1, 14 eV.

4. CONCLUSION

La mesure du spin des résonances et les propriétés du phénomène de fission sont deux aspects d'un même problème qui s'enchevêtrent étroitement. La façon logique de procéder serait d'abord de connaître le spin des résonances pour en déduire les propriétés de la fission. Cependant, la mesure des spins est très difficile et c'est la raison pour laquelle on se sert souvent des dernières, sans pour autant bien les connaître. Les travaux ont donc progressé parallèlement dans ces deux domaines et il nous a été impossible de les citer tous dans ce mémoire.

Les travaux commencent maintenant à converger sur une représentation cohérente du problème; c'est le cas de ²³⁹Pu où les mesures de largeurs de fission, de spin, de distribution en masse et d'énergie des fragments de fission correspondent à peu près à la théorie de Bohr. Plusieurs points restent encore à préciser. Des mesures de diffusion élastique devraient être améliorées et d'autres mesures seraient à entreprendre pour compléter notre compréhension du phénomène (fission ternaire, neutrons de fission, etc.).

L'uranium-235 est plus difficile à étudier, d'une part à cause des mesures de diffusion élastique, d'autre part parce que les propriétés de la fission ne semblent pas dépendre beaucoup de l'état de spin. Heureusement, par une sorte de bienfait de la nature, ce noyau est moins difficile à polariser que les autres. Par la polarisation, une source considérable d'informations commence à nous être fournie, qui va connaître d'assez grands développements dans un proche avenir.

Restent ²³³U et ²⁴¹Pu, tous deux de spin 5/2+, pour lesquels nous avons moins d'informations. L'énergie de liaison de ²⁴¹Pu est plus faible que celle de ²³³U, ce qui devrait faciliter l'analyse et l'interprétation des résultats (résonances plus espacées, plus étroites et énergie d'excitation au point seuil plus faible). Cependant, il est plus difficile à étudier expérimentalement.

Après avoir cheminé longtemps dans des voies difficiles et ingrates, il semble que l'étude de la fission connaisse un nouvel essor et s'ouvre sur des perspectives plus attrayantes. De nombreux résultats vont certainement être obtenus dans les années à venir et parmi ceux-ci la connaissance du spin des résonances représente une des clés de ce vaste problème.

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ANNEXE

COMPARAISON DES VALEURS DU SPIN DES RESONANCES DE ²³⁹ Pu

$\frac{\frac{\Gamma_n^0}{\Gamma}}{(\text{meV}^{-1})}$	E (eV)	gexp Harwell [10]	J Harwell [10]	J Saclay [8]
$2, 7 \cdot 10^{-2}$	164, 5	0,79±0,16	1	1]
$2, 2 \cdot 10^{-2}$	251, 2	1,03 ± 0,12	1	1
2, 15 · 10^{-2}	90, 7	0,82 ± 0,04ª	1	1 Accord
1,8 · 10 ⁻²	231, 4	0,77 ± 0,1	1	1
1,5 · 10 ⁻²	248, 9	0,6 ±0,06	1	1
1,5 · 10 ⁻²	118, 7	0,41 ± 0,10	0	1
5,1 · 10 ⁻³	199, 4	0,36 ± 0,12	0	1 Désaccord
4,2 · 10 ⁻³	242, 9	0,21 ± 0,06	0	1
2,3 · 10 ⁻³	157, 1	0,14 ± 0,03	0	0 Accord
10 ⁻³	131, 7	0,14 ± 0,03	0	0
8,5 10 ⁻⁴	66	0,01 ± 0,001 ^a	0(?)	0р

a Valeur de g_{exp} déduite de σ_{0n} Γ [10] (non corrigé des effets de diffusion multiple) et des paramètres 2 g Γ_n et Γ mesurés à Saclay [8].

^b Valeur de J mesurée par Cowan [38].

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DISCUSSION

C.D. BOWMAN: I would like to say a few words in favour of using resonance-resonance interference in the scattering cross-section for making spin assignments. We have measured scattering cross-sections at Livermore on the nuclides 233 U, 235 U and 241 Pu in addition to our earlier measurements on 239 Pu. We have developed a multi-level fitting code based on the Reich and Moore formalism; by means of a least-squares procedure this code searches for the best fit to the data, given a set of spins and the signs of interference. Of course, the fits used to assign the spins must also be consistent with the total fission cross-section, which has already been well measured. We are currently fitting this data and hope to be able to assign spins to as many as half the resonances below 15 eV in 241 Pu and 233 U. We expect very little success on 235 U.

J. THEOBALD: I should like to make a comment on the indirect methods of spin determination. I wonder why one has always considered only one kind of energy de-excitation during the fission process, e.g.

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ternary fission, fragment kinetic energy or neutron emission. I think that the summing of all measured de-excitation phenomena (including future measurements of neutron, beta and gamma de-excitation) from resonance to resonance should increase the energy differences for the two spin families.

A. MICHAUDON: It would certainly be of interest to sum fission processes which show the same trend – fission neutrons, ternary alpha rays, beta rays – to increase the difference from resonance to resonance.

J. JULIEN: The use of Li-Ge detectors to reveal weak transitions in the spectrum of gamma rays emitted after neutron capture will, I think, help in determining resonance spins for fissile nuclei. The radical difference that we now find for certain nuclei between these detectors and those employing NaI(Tl) crystals gives reason to hope that even without an anti-coincidence circuit it will be possible to reveal differences in gamma-ray spectra for fissile nuclei. We shall shortly be studying a method for doing this.

SPIN ASSIGNMENTS OF LOW-ENERGY RESONANCES IN ²³⁹Pu

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Abstract

SPIN ASSIGNMENTS OF LOW-ENERGY RESONANCES IN ²³⁹Pu. Neutron scattering measurements have been made up to about 300 eV on ²³⁹Pu with the existing neutron scattering detector, which was modified to take account of the fission neutrons. Combining these data with the published transmission data ($g\Gamma_n, \Gamma$) spin assignments of 45 resonances have been made. If the resonances assigned by other workers are included, the average values of fission widths for 0⁺ and 1⁺ resonances up to 200 eV are found to be 218, 2 and 42, 1 meV, respectively. It is shown that this average value of fission width (42, 1 meV) for 1⁺ resonances is inconsistent with the hypothesis of 1⁺ resonances fissioning through a 1⁺ two quasiparticle transition state and one needs a 1⁺ transition state at a much lower energy to account for this result. This state may be a 1⁺ combination transition state formed from mass asymmetry and bonding vibrations.

1. INTRODUCTION

The application of the collective model [1,2] to the fission process predicted that the height of the fission barrier should depend significantly on the spin and parity of the level of the compound nucleus involved. Since the probability of fission is closely related to the properties of the fission barrier, it means that among other things, such as the peak-to-valley ratio in fission fragment distributions, the fission widths of the compound nucleus resonances observed when s-wave neutrons interact with fissile nuclei should be correlated with the spins of these resonances which can be either $J = I + \frac{1}{2}$ or $I - \frac{1}{2}$, where I and $\frac{1}{2}$ are respectively the spins of the target nucleus and neutron.

Apart from this theoretical interest, the spin values of these compound nucleus s-wave resonances are involved in a direct way when multi-level resonance theories are used to fit the measured fission cross-section data [3]. A straightforward method of measuring the spins of these lowenergy s-wave resonances is to combine the total cross-section data $(g \Gamma_n \text{ and } \Gamma)$ with the data obtained from neutron resonant scattering measurements. But, unlike the non-fissile nuclei, the main difficulty in making scattering measurements on fissile nuclei is the presence of unwanted fission neutrons. Nevertheless, some neutron resonant scattering measurements have been made on fissile nuclei such as ²³⁹Pu up to about 90 eV using either the 'bright line' technique [4], where these fission neutrons and fission and capture γ -rays are eliminated by the time-offlight method, or a ¹⁰B sleeve [5] around the scattering sample to subtract out the fission neutrons. ¹⁰B has a high capture cross-section for low-energy neutrons, while for high-energy fission neutrons its capture cross-section is very low. Though the bright line technique is an ideal one to make scattering measurements on fissile nuclei, yet it is difficult to go beyond, say 50 eV, using this technique, because above this energy, the loss in energy of a scattered neutron due to the recoil of the scattering

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nucleus becomes so large that it is difficult to correlate the measured neutron energy with its real energy. We have modified our neutron scattering detector [6] at the Harwell 45-MeV electron linear accelerator, to take account of the fission neutrons (see section 3). In this paper we present our results on spin assignments of resonances in ²³⁹Pu up to about 300 eV.

2. EXPERIMENTAL ARRANGEMENT

The neutron resonant scattering measurements were made using the time-of-flight spectrometer based on the Harwell 45-MeV electron linear accelerator pulsed neutron source with its boosted neutron target [7], giving neutron pulses of duration about 200 ns, at a repetition rate of about 200 pulses/s.

The neutron scattering measurements were carried out on a flight path of about 50 m using the neutron scattering detector described in Ref. [6]. This detector was modified to take account of the fission neutrons from ²³⁹Pu. Ordinarily, in this detector, scattered neutrons are detected through the reaction ⁶Li (n, α)T + 4.8 MeV, using ⁶Li glass scintillators (called A). The γ -ray response of these scintillators is determined using natural lithium glass scintillators (called B). If the ratio of the γ -ray sensitivity of A relative to B (determined using ThC'' 2.6-MeV γ -rays) is K $_{\gamma}$, it can be shown that the number of neutrons scattered at energy E is

$$N' = \epsilon' \left(N_{A} - K_{\gamma} N_{B} \right)$$
(1)

where N_A and N_B are the number of events recorded, respectively, by A and B at energy E for the same recording time and ϵ' is an efficiency factor.

To take account of the fission neutrons, we used a stilbene crystal (called C) in conjunction with the neutron scattering detector. The response of this crystal to γ -rays was reduced to less than 0.3% using a pulse shape discrimination circuit [8]. The sensitivity of the neutron scattering detector (A) to fission neutrons relative to the fission neutron detector (C) was determined, using a ²⁴⁰Pu spontaneous fission source. It can be shown that for a fissile target nucleus, the number of neutrons scattered at an energy E is given by Eq.(1), when modified to

$$N = \epsilon (N_A - K_{\gamma} N_B - K_f N_C)$$
(2)

where N_C , like N_A and N_B , is the number of events recorded by C at energy E, and ϵ is again an efficiency factor.

Here it is assumed that the spontaneous fission spectrum of ²⁴⁰Pu is similar to the fission spectrum of fission occurring in the resonances of ²³⁹Pu and that the fission spectrum does not vary significantly from resonance to resonance.

It should also be pointed out that the neutron scattering detector has a low response to fission neutrons, for example, the fractional contributions (in terms of the total area of a resonance) of the fission neutrons to the 47.7, 52.6, 65.8 and 164.7 eV resonances are respectively 2.4, 2.6, 6.1 and 1.7%.

The data from A, B and C were recorded separately but simultaneously on a magnetic tape using 8192 timing channels of width 125 ns, giving an overall timing resolution of about 4.5 ns/m.

The efficiency factor, ϵ , was taken into account by making measurements relative to lead which has a smooth and accurately (about 1%) known cross-section.

One metallic sample (5-cm diameter disc) which contained about 98% of ²³⁹Pu alloyed with about 1% aluminium was used. The thickness of the sample was n = 5.79×10^{-4} atoms/b. A lead sample of n= 5.87×10^{-3} atoms/b was used to calibrate the scattering detector for this experiment.

3. ANALYSIS OF THE DATA

In Fig.1 some of the scattering data are presented as counts $(N_A - K_y N_B - K_f N_C)$ per timing channel. The slowly varying background which includes the effect of potential scattering, was fitted to a curve of the form $B(t) = B_0/(t+A)^C$ where A, B_0 and C are constants determined from the data. After the subtraction of the background the resonance data were analysed using the area method [9]. The area under each resonance was determined between finite limits. When the scattering sample is thin (n $\sigma_0 \ll 1$), the resonance scattering area with infinite limits is proportional to $\sigma_0 \Gamma_n$. In practice, however, this condition was not met and the resonance areas had to be corrected for the finite limits of integration, for the resonance self-shielding and Doppler effect and for the attenuation of the scattered neutrons due to multiple scattering in the sample. The corrections and the way they were applied are discussed in Ref. [10]. The published values of $g \Gamma_n$ from the transmission data [11,12] were used to apply the various corrections mentioned above. Table I lists the values of $\sigma_0 \Gamma_0$ for 53 resonances. The errors quoted on $\sigma_0 \Gamma_0$ are the statistical errors.' Systematic errors in the calibration of the detector were ignored, because the cross-section of lead, relative to which the measurements were made, is known to be about 1% (11.4 b) and multiple scattering corrections were small. Now the scattering area from a resonance is

$$\sigma_0 \Gamma_n = (2.603 \times 10^6 / E_R) g \Gamma_n^2 / \Gamma barn eV$$

where the resonance energy E_R , the partial level width Γ_n , and the total level width Γ are expressed in eV, and where the statistical factor g is (2J+1)/2(2I+1). Since $I=\frac{1}{2}$ for ^{239}Pu , the two possible g-values are $\frac{1}{4}$, corresponding to J=0, and $\frac{3}{4}$, corresponding to J = 1. If the values of $g\Gamma_n^2/\Gamma$ are obtained from scattering areas, previously determined values of g Γ_n and Γ from transmission data can be used to find g, hence J, for each resonance from the relation

$$g = (g \Gamma_n)^2 / \Gamma (g \Gamma_n^2 / \Gamma)$$

The g-values (hence values of J) of various resonances listed in Table I were obtained from Eq. (3) where our measured values of $g\Gamma_n^2/\Gamma$ were

(3)







FIG.1. Scattering data as counts (NA - $K_{\gamma}N_B$ - K_fN_C) per timing channel

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E _R	σ _o Γ _n	· ,	< ²	Spin T	Sauter and	Frazer and
(eV)	(b.eV)	Spin J = 0	Spin J = 1	chosen.	Bowman [4]	Schwartz [5
7.9			· · · · · · · · · · · · · · · · · · ·		1	1
10.9					1	1
11.9					1	1.
14.3					1	
14.68	5.72 ± 0.40	48.66	0.395	1	1	0
15.54					0	
17.65	4.95 ± 0.48	38,97	1.59	1	1	1
22.28	6.1 ± 0.35	54.75	1.0	1	1	0
26.26	2.26 ± 0.30	14.47	0.61	1.		
32.38					0	
35.78					1	
41.5	11.75 ± 0.76	23.78	1.8	1	1	1
44.5	27.4 ± 1.14	40.23	2.03	1	1	0
47.7	1.52 ± 0.33	0.106	8.24	0	· 0 ·	
50.2	4.55 ± 0.60	[.] 21.09	1.65	1	•	
52.6	40.2 ± 1.42	29.93	3.77	1	1 .	
55.7	1.62 ± 0.42	3.98	6.32	(0)		
59.3	12.0 ± 0.8	3.79	31.2	0		
65.8	29.5 ± 1.07	61.38	3.53	1		
74.3	2.65 ± 0.46	18.81	0.799	1		
75.01	75.71 ± 2.30	58.16	0.84	1	1	
85.7	32.0 ±1.56	2.94	9.57	0		
90.8	32.2 ± 1.20	26.57	4.90	1		
95.6	2.10 ± 0.54	0.459	15.82	0		
103.5	4.93 ± 1.0	2.95	75.08	0		
105.6	10.12 ± 1.02	14.93	0.93	1		
106.8	16.2 ± 1.26	50.67	2.3	1		
116.3	2.60 ± 0.9	0.002	40.93	0		
119.0	39.8 ± 1.65	25.62	1.370	1		
131,9	2.79 ± 1.14	1.90	45.91	0		
134.2	7.8 ± 0.98	11.06	0.008	1		
137.1	3.4 ± 0.52	0.226	17.74	0		

TABLE I. ²³⁹Pu RESONANCE AREAS AND SPINS^a

TABLE I cont'd

ER	o _o I _n	,	χ ²	Spin J	Sauter and	Frazer and
(eV)	(b.eV)	Spin $J = 0$	Spin J = 1	chosen	Bowman [4]	Schwartz [5]
146.7	9.4 ±1.1	29.51	0.0007	1		
157.5	9.0 ±1.1	1.111	95.40	0		
164.7	69.2 ± 2.1	48.31	1.59	1		
167.7	4.35 ± 0.9	7.30	0.78	1		
177.8	1.45 ± 0.7	2.34	0.74	1		
196.0	14.4 ± 1.4	0.865	20.51	0		
197.6	5.3 ±1.54	10.81	3.85	1		
200.0	6.2 ±1.6	3.42	0.002	1		
204.7	19.4 ±1.73	0.28	53.26	0		
208.0	6.67 ± 3.1	2.78	0.14	1		
217.0	7.85 ± 2.46	2.56	1.81	(1)		
232.0	8.5 ± 1.27	11.59	0.553	1		
235.4	35.5 ±1.83	0.30	78.7	0		
240.0	4.45 ± 1.18	3.85	1.04	1		
243.8	7.28 ± 1.94	1.31	9.11	0		
250.0	18.1 ±1.4	48.27	1.08	1		
251.5	28.4 ± 3.0	147.0	2.90	1		
257.2	4.05 ± 1.52	2.46	0.21	1		
262.5	2.62 ± 2.3	2.13	30.10	0		
273.6	38.2 ± 2.54	22.70	3.75	1		
276.9	25.6 ± 2.65	12.01	0.019	1		
281.0	10.1 ± 2.3	0.084	20.48	0		
284.1	47.3 ± 4.65	46.42	0,85	1		
299.7	4.85 ± 2.0	2,51	0.80	1		
303.2	17.5 ± 2.4	6.24	3.22	(1)		

^a This Table was made available as a replacement for the original Table I, just before going to press. Some of the spin values here presented are different from those originally provided because an error was found in the γ -sensitive part of the detector. Now the spin distribution of levels (unlike those in the conclusion of section 5 of the text) is not inconsistent with the (2J + 1) distribution.

combined with the published values of g Γ_n and Γ determined from total cross-section measurements data [11, 12]. Although scattering data were available for 53 resonances, only 45 spin assignments could be made,

because for the rest of the resonances either the values of both g_{Γ}^{n} and Γ were not available or they were of poor quality. The uncertain assignments are enclosed in parentheses. The errors quoted on the values of g for the resonances up to 100 eV include the errors only in g_{Γ}^{n2}/Γ , because the transmission data used do not quote errors in g_{Γ}^{n} and Γ . Beyond 100 eV the errors quoted on g were computed by adding in quadrature the errors in g_{Γ}^{n2}/Γ and g_{Γ}^{n} (and in Γ , when quoted). The errors are not realistic also, because the systematic errors that might be present in other data were not included.

4. **RESONANCE SPIN ASSIGNMENTS**

The results of spin assignments of 44 resonances (excluding the uncertain assignments) are presented in Table I. Table I lists also the spin assignments made by Sauter and Bowman [4], Frazer and Schwartz [5] and Bollinger et al.[13]. Bollinger's assignments of spins are in general merely guesses based on resonance shapes and, in his view, should not be taken very seriously. Frazer and Schwartz assigned their spins using their scattering data obtained by using a ^{10}B sleeve, while Sauter and Bowman's results are based on scattering measurements carried out using the bright line methods. While we (like Sauter and Bowman) disagree with Frazer and Schwartz in their spin assignments for 14.68, 22.28 and 44.5 eV resonances, all our assignments agree with the corresponding assignments made by Sauter and Bowman. Out of our 45 spin assignments, there are 22 resonances with J = 1 and 23 with J = 0, but if Sauter and Bowman's assignments are included, there are 52 spin assignments in all and out of these 27 resonances are with J = 1 and 25 resonances with J = 0.

5. DISCUSSION

From a total of 52 spin assignments, there are 27 resonances with J = 1 and 25 resonances with J = 0. This distribution is not consistent with a (2J + 1) dependence for level density. This may be due to a large number of small resonances not assigned and most of which may be of spin J = 1.

The collective model predicts [1, 2] that regular (even-even or oddodd spin-parity resonances should have greater average fission widths than the irregular (odd-even or even-odd) combinations. This prediction follows from the assumption that for even-even nuclei the transition state spectrum would exhibit in its low-lying states the correlation between spin and parity that dominates the spectra of deformed nuclei. Thus the lowest lying band in the transition spectrum of 239 Pu + n would be K = 0, J^{π} = 0⁺, 2⁺, 4⁺..., and the 0⁺ resonances would fission through the 0⁺ transition state. There should be a 1[±] transition state through which the 1[±] resonances can fission. This 1[±] transition state can be obtained either as a two quasi-particle state or from combination states formed from mass asymmetry (K = 0, J^{π} = 1⁻, 3⁻...) and bending (K = 0, J^{π} = 1⁻, 2⁻, 3⁻...) vibrations. Now the analysis of the experimental results on angular distributions of fission fragments from (d, pf) on ²³⁹Pu and from ²³⁹Pu + n using superfluid model show an energy gap of $2\Delta_0$ of 2.3 to 2.6 MeV

between the 0^+ transition state (the fission threshold for 0^+ resonances) and the two quasi-particle states (compared to about 1.4 MeV for the stable nucleus) [14-17]. If we take 6.4 MeV as the neutron binding energy for 239 Pu + n. 4.95 MeV as the 0⁺ fission threshold and assume that 1⁺ resonances fission through a 1⁺ transition state in a quasi-particle band, we find that there is a 0.85 to 1.15 MeV barrier against fission through this 1⁺ quasi-particle state. This means that the fission probability (hence the average fission width) for 1^+ resonances in 239 Pu + n should be, on the average, less by a factor of 10^7 to 10^9 than the corresponding value for 0⁺ resonances. (Here we assume that each factor of ten increase in half-life corresponds to an increase of about 0.125 MeV in the barrier height.) Though we find that very wide resonances in 239 Pu + n, such as 15.5, 132.1 and 262.5 eV, have, indeed, spin-parity 0⁺, yet if we use the resonance fission areas of de Saussure et al. [18], we find that up to 200 eV, the average fission widths $<\Gamma_f > J^{\pi} = 0^+$ (for 17 resonances, all wide resonances included) and $\langle \Gamma_i \rangle J^{\pi} = 1^+$ (for 19 resonances) are respectively 218.2 and 42.1 meV. This large value of $<\Gamma_{\!f}>~J^{\pi}$ = 1+ (42.1 meV), hence enhanced probability of fission through 1⁺ resonances, implies that the fission barrier for 1^+ resonances is much less than that suggested by a two quasi-particle state. In other words, it means there should be a 1⁺ transition state (other than the 1⁺ two quasi-particle state) at a lower energy to account for this large value of $< \Gamma_f > J^{\pi} = 1^+$. Recently Griffin [19] has claimed that 1⁺ resonances in ²³⁹Pu + n may fission through a 1⁺ combination state formed from mass asymmetry and bending vibrations. This combination would provide a transition state at an energy $E_{1+} \sim \hbar \omega_{MA} + \hbar \omega_{B}$, where the first and the second terms on the right hand side, are respectively the energies of mass asymmetry and bending vibration components. His liquid model calculations when combined with photo-fission data, show that a combination 1⁺ transition state occurs at an energy about 1.1 MeV above the 0⁺ fission threshold. We can determine the energy of a 1⁺ transition state, (hence the 1⁺ fission threshold) relative to the excitation energy, that would give the experimental value of $<\Gamma_f > J^{\pi} = 1^+$ of 42.1 meV, by using the Hill and Wheeler formula [20]

$$2\pi < \Gamma_{\rm f} > J^{\rm T}/<{\rm D} > J^{\rm T} = 1/[1 + \exp 2\pi ({\rm E}_{\rm f} - {\rm E})/\hbar\omega] \tag{4}$$

where $<\Gamma_f > J^{\pi}$ is the average fission width for a state of spin-parity J^{π} , $<D>J^{\pi}$ is the average level spacing for spin-parity J^{π} states (we take a value of 3 eV for 1⁺ resonances) [11], E_f is the fission threshold for spinparity J^{π} states, E is the excitation energy and is a measure of the barrier thickness. The value of $\hbar \omega$ can be taken as about 0.45 MeV for $J^{\pi} = 1^+$ states. Using this formula, we find that a barrier of about 0.062 MeV is required to obtain a value of 42.1 meV for $<\Gamma_f > J^{\pi} = 1^+$. Hence this 1⁺ combined state should lie at about 1.51 MeV above the 0⁺ fission threshold. This value of about 1.51 MeV is quite high compared to 1.1 MeV given by Griffin. It must be pointed out though, that Griffin's calculation [19] was a rough estimate made on the assumption that the values of average fission width for 0⁺ and 1⁺ resonances are the same.

In summary, we can say that if one accepts the validity of an energy gap of $2\Delta_0$ of 2.3 to 2.6 MeV between the 0⁺ fission threshold and 1⁺ two quasi-particle transition state, our result for the $<\Gamma_f > J^{\pi} = 1^+$ is inconsistent with the extremely low probability for the 1⁺ resonances to fission

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through the 1⁺ two quasi-particle transition state. It seems that one has to postulate a 1⁺ transition state such as a combination state suggested by Griffin, at an energy lower than the 1⁺ two quasi-particle state, in order to satisfy the experimental result. The theoretical estimate for the energy of this 1⁺ combination state is low compared to the value suggested by our experiment, but it is, as was pointed out before, a very rough estimate which, one hopes, can be improved upon.

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SECTIONS EFFICACES TOTALE ET DE FISSION DU ²³⁹Pu Etude statistique des paramètres de résonances

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Abstract — Résumé

TOTAL AND FISSION CROSS-SECTIONS OF ²³⁹Pu - STATISTICAL STUDY OF RESONANCE PARAMETERS. The authors measured the total and fission cross-sections of ²³⁹Pu with the linear accelerator at Saclay as a pulsed source of neutrons.

The total cross-section was measured in the range from 4 to 700 eV and the best resolution used was 1.5 ns/m; the fission cross-section was measured between 4 eV and 6 keV, the best resolution having been 6 ns/m. The transmission measurements on five samples were made at the temperature of liquid nitrogen, and comparisons made with supplementary experiments at ambient temperature made it possible to determine the Doppler broadening factor ($\Delta = \eta \sqrt{E}$). The resonances were identified from 4 to 500 eV in the total cross-section; the average level spacing was of the order of 2.4 eV. It would appear/that, in this energy range, nearly all the levels were identified.

The resonance parameters were determined by analysis of shape in conjunction with a least-squares programme on an IBM-7094 computer. The existence of a large number of broad resonances corresponding to very large fission widths has been shown to exist. Statistical study of the fission widths actually shows the existence of two families of resonances, one corresponding to a mean Γ_f of the order of 45 meV and the other to a mean Γ_f of about 750 meV. The authors were therefore able to postulate a classification of resonances in terms of two spin states, the level population ratio in each family being: $(2J_1+1)/(2J_2+1)=1/3$, $J_1 = 0$ corresponds to the broad resonances and $J_2 = 1$ to the narrow ones. The partial widths for radiative capture fluctuate slightly around a mean value of 40 meV. By using a multilevel programme, the authors were able to investigate the extent to which the existence of large fission widths might give rise to fictitious resonances (quasi-resonances) and perturbations and also to make a statistical study of the resonance parameters.

SECTIONS EFFICACES TOTALE ET DE FISSION DU ²³⁹Pu - ETUDE STATISTIQUE DES PARAMETRES DE RESONANCES. Les sections efficaces totale et de fission de ²³⁹Pu ont été mesurées auprès de l'accélérateur linéaire de Saclay utilisé comme source de neutrons pulsée.

La mesure a été faite de 4 à 700 eV pour la section efficace totale et la meilleure résolution utilisée était de 1,5 ns/m; ide 4 eV à 6 keV pour la section efficace de fission, la meilleure résolution étant de 6 ns/m. Les mesures de transmission sur cinq échantillons ont été faites à la température de l'azote liquide, et des comparaisons avec des expériences complémentaires faites à température ambiante ont permis aux auteurs de déterminer le coefficient d'élargissement Doppler ($\Delta = \eta \sqrt{E}$). Les résonances ont été identifiées de 4 à 500 eV en section efficace totale; l'espacement moyen des niveaux est de l'ordre de 2,4 eV; il semble que, dans cette gamme d'énergie, presque tous les niveaux soient identifiés.

Les paramètres des résonances ont été déterminés par analyse de forme à l'aide d'un programme de moindres carrés sur calculateur IBM 7094. Les auteurs ont mis en évidence l'existence d'un grand nombre de larges résonances correspondant à de très grandes largeurs de fission. L'étude statistique des largeurs de fission montre effectivement l'existence de deux familles de résonances, l'une correspondant à Γ_f moyen de l'ordre de 45 meV, et l'autre à Γ_f moyen de l'ordre de 750 meV. Il a donc été possible aux auteurs d'envisager une classification des résonances suivant les deux états de spin, le rapport de population des niveaux dans chaque famille étant de $(2J_1 + 1)/(2J_2 + 1) = 1/3$; $J_1 = 0$ correspond aux résonances larges, et $J_2 = 1$ aux résonances étroites. Les largeurs partielles de capture radiative fluctuent peu autour d'une valeur moyenne de 40 meV. Un programme multiniveaux a permis également aux auteurs d'étudier dans quelle mesure l'existence de grandes l'argeurs de fission pouvait créer des résonances fictives (quasi-résonances) et perturber ainsi l'étude statistique des paramètres de résonances.

Le spin du noyau du ²³⁹Pu est de 1/2+ et l'espacement des niveaux est plus grand que celui des autres noyaux fissiles connus, aussi son étude semble plus facile; en particulier, l'étude de la distribution des largeurs de fission et l'attribution des largeurs totales de capture radiative permet la détermination du spin d'un assez grand nombre de résonances. Dans l'interaction de ce noyau avec les neutrons lents, le noyau composé formé par l'absorption d'un neutron s peut se trouver dans deux états de spin différents: 0+ et 1+. On est en présence de deux familles de résonances, correspondant aux deux facteurs statistiques $g_0 = 1/4$ et $g_1 = 3/4$.

D'après la théorie développée par Bohr à la Conférence de Genève de 1955 [1], l'énergie potentielle de déformation au point seuil de fission devient telle qu'il reste peu d'énergie pour exciter les états quantiques du noyau composé; la configuration des niveaux est simple et s'apparente à celle qui existe au voisinage de l'état fondamental du noyau pair-pair correspondant. Dans le cas du ²³⁹Pu, à parité positive, l'état 0+ peut appartenir d'une part à la première bande de rotation (0+, 2+, 4+, ...)et d'autre part à la bande de vibration quadrupolaire; l'état 1+ correspondrait à un état de double vibration issu de la combinaison de deux états de vibrations octupolaires avec K = 0 et K = 1 [2], la bande de double vibration étant plus voisine du niveau fondamental 0+ que la région des états à deux quasi-particules.

Le seuil de fission le plus bas se trouve à peu près à 1,5 MeV audessous de l'énergie associée au noyau composé (239 Pu+neutron); les bandes de vibrations octupolaires et quadrupolaires sont à environ 0,7 MeV au-dessus de ce point seuil, et la bande de double vibrations au voisinage de l'énergie du noyau composé. Les valeurs de Γ_f pour l'état de spin 0+ peuvent être grandes; mais celles correspondant à l'état 1+ doivent être beaucoup plus faibles; l'intervalle d'énergie entre les seuils de fission correspondant aux deux états de spin est de 1,5 MeV environ. L'une des voies 0+ est totalement ouverte; l'autre ne peut que l'être partiellement; la voie 1+ n'est que partiellement ouverte.

En se fondant sur ces hypothèses, l'étude statistique des paramètres de résonances du ²³⁹Pu a permis de dégager les points suivants:

1° Il existe de très larges résonances dues à de grandes largeurs de fission.

2° La distribution des Γ_f montre bien l'existence de deux familles de résonances, l'une correspondant à une faible valeur moyenne ($\langle \Gamma_f \rangle = 42 \text{ meV}$) et une autre à une valeur moyenne supérieure à 1 eV.

3° Il n'a pas été possible de mettre en évidence deux valeurs moyennes différentes de Γ_y pour les deux familles, Γ_y pour les

résonances larges étant mal défini ou impossible à déterminer.

4° L'examen de la distribution de Γ_f et l'hypothèse que Γ_y est constant de résonance à résonance et ne dépend pas du spin permettent d'attribuer le spin à une trentaine de résonances.

Les sections efficaces totale et de fission ont été mesurées par la méthode du temps de vol auprès de l'accélérateur linéaire de Saclay. L'identification et l'étude des niveaux ont été faites jusqu'à 440 eV; un tableau détaillé des paramètres de résonances a pu être établi jusqu'à 250 eV; au-delà de 250 eV, nous donnons uniquement les valeurs de Γ et de $2g\Gamma_n$, les niveaux n'étant plus suffisamment séparés sur la courbe de section efficace de fission (tableaux I et II).

1. CONDITIONS EXPERIMENTALES DES MESURES

1.1. Section efficace totale

Les mesures de transmission ont été faites sur cinq épaisseurs d'échantillon d'un alliage de Pu-Al de 0,8% d'Al pour les fortes épaisseurs et de 80 à 88% d'Al pour les autres épaisseurs. Les échantillons contenaient respectivement 0, 1, 0, 3, 1, 4 et 14 g de ²³⁹Pu par cm². Dans le domaine des résonances, la transmission a été mesurée à la température de l'azote liquide (77°K), la largeur Doppler étant alors à peu près deux fois plus faible qu'à la température ambiante; le cryostat utilisé était mobile. L'intérêt d'un tel refroidissement a déjà été exposé [3,4]. Le tableau III résume les conditions expérimentales.

Des échantillons d'Au, de Co, de Mn ou de Bi mis en permanence dans le faisceau ont permis une bonne évaluation du bruit de fond. Pour une partie des mesures nous avons pu utiliser en ligne un calculateur CAE enregistrant les données sur bandes magnétiques, permettant ainsi des mesures quasi simultanées de la transmission, du spectre et des bruits de fond transmission et spectre (quatre séquences consécutives) [5].

1.2. Section efficace de fission

Les mesures ayant servi à cette analyse ont été effectuées à 16 m sur la base de vol inclinée à 18° par rapport à la perpendiculaire à la cible d'uranium, avec un scintillateur gazeux contenant un mélange argon-azote, de 300 g/cm² d'argon pour 25 g/cm² d'azote, la gamme d'énergie étudiée allant de 3,8 eV à 250 eV.

L'accélérateur fonctionnait à 500 c/s avec une largeur d'impulsion égale à 60 ns. La largeur des canaux du sélecteur de temps de vol était de 0,4 μ s de 3,8 eV à 37 eV et de 0,05 μ s de 37 eV à 250 eV. Un écran de Mn de 5 mm d'épaisseur a été utilisé comme écran de bruit de fond permanent. Nous avons utilisé des écrans de cobalt et de tantale pour les séquences bruit de fond. Nous avons obtenu pour une durée d'accumulation de 100 h

> $29\,300$ c à 10,9 eV dans un canal de 400 ns $12\,500$ c à 75 eV dans un canal de 50 ns.

2. ANALYSE DES COURBES EXPERIMENTALES

Dans un mémoire présenté également à cette Conférence [5] ont été décrites les méthodes utilisées pour le traitement des données. En particulier, les paramètres des résonances ont été obtenus par analyse de forme utilisant une méthode de moindres carrés dans laquelle la fonction théorique est une somme de formules de Breit et Wigner à un niveau élargies par effet Doppler; on tient compte d'un terme d'interférence de diffusion entre résonances. La résolution expérimentale est assimilée à une gaussienne. Dans la plupart des cas il y a un bon accord entre les points expérimentaux et la courbe théorique; l'existence de très larges résonances a été mise en évidence, telles les résonances à 82, 8, 96, 5, 131, 8, 147, 4, 212, 333, 9 eV, qui sont très bien séparées; d'autres

E (eV)	2g Г _п (meV)	σ₀Γ (b*eV)	a₀I] (b*eV)	Г (meV)	Γ _f (meV)	Γ _γ (meV)	η/ υ	g	
0.30	0, 121			99 + 4	60 ± 4	39 + 3	0.61	1/4	 (م
7 99	121 + 0.04	201 + 6.6	108 2 + 2	87 + 5	47 + 3	397+4	0.54	3/4	a /
10 93	284 ± 0.15	338 2 1 17 0	249 1 ± 5	200 + 20	143 + 16	55 1 + 9	0.72	3/4	t.
10,55	0.15	000,2 ± 11,5	272,1 1 0	200 - 20	140 - 10		0,12	0/ 4	ы
11 89	154 ± 0.10	168 6 + 11 0	60 9 + 2	67 + 7	24 + 3	42 0 + 4 6	0.37	3/4	5)
14 91	1,04 ± 0,10	100,0 ± 11,0	54.7 ± 2	102 - 8	24 ± 3 67 + 7	34 + 6	0,66	3/4	
14,51	0,31 ± 0,05	02,0 ± 4,0	107 5 + 9	70 + 7	30 + 3	37.6 + 4	0.00	1/2	
15,00	2,00 ± 0,00	203,0 ± 4,3	107,0 ± 2	700 + 50	650	0110	0,11	1/2	
10,40	$0,38 \pm 0,08$	$52,0 \pm 0,3$	00,0 ± 0	75 4 7	34 + 4	3974 47	0,37	2/4	
17,00	$2,74 \pm 0,03$	201,9 ± 3,0	$92, 1 \pm 2$	100 + 0	29 ± 2	JO, 1 ± 4, 1	0,47	3/4	
22,29	$4,00 \pm 0,10$	200,0 ± 0,8	132,3 ± 3	109 ± 9	62 ± 0 55	44,3 = 0,3	0,38	3/4	
23,94	$0,13 \pm 0,01$	7,1 ± 0,5	0,0 50 10	70 ± 12	55 44 ± 7		0,79	1/0	
26,24	$2,20 \pm 0,15$	109,1 ± 7,4	58 ± 3	83 ± 10	44 ± 7	37 ± 6	0,55	1/2	
27,24	$0,12 \pm 0,01$	$5,7 \pm 0,3$	1,1 10,0,1,0,5	42 ± 8	8 ± 4	34 ± 8	0,19	1/2	
32,31	$0,42 \pm 0,02$	$16,9 \pm 0,7$	12,2 ± 0,5	153 ± 20	110 ± 15	41 ± 8	0,73	1/4	
34,60	0,02								
35,50	0,43 ± 0,02	15,8 ± 0,6	1,6	47 ± 9	5 ± 2	42,4 ± 8,5	0,10	3/4	
41,42	6,20 ± 0,20	194,9 ± 6,2	9	52 ± 8	3	43,5 ± 8	0,05	1/2	
41,66	2,02 ± 0,25	63,1 ± 7,9	32,4	105 ± 16	54	49	0,52	1/2	
44,48	9,97 ±0,20	291,7 ± 5,9	23,0±0,8	58 ± ?	5 ± 1	46,8±7	û,09	3/4	
47,60	2,90 ± 0,15	79,3 ± 4,0	59,2 ± 2	322 ± 25	240 ± 24	74 ± 16	0,76	(1/4)	
49,71	2,2 ±0,2	58 ± 6	49,4 ± 5	810 ± 200	690 ± 200		0,86	1/4	
50,08	4,55 ± 0,20	118,2 ± 5,2	24,8 ± 2	57 ± 10	12 ± 3	42,0 ± 8	0,22	3/4	
52,60	15,70 ± 0,30	388,5 ± 7,4	51 ± 1	68 ± 10	9 ± 2	48,6 ± 9	0,16	3/4	
55,63	2,20 ± 0,12	51,5 ± 2,9	19 ± 2	59	22.	35	0,40	1/2	

TABLEAU I. PARAMETRES DE RESONANCES DU ²³⁹Pu

a) BNL 325.

 b) Explique un très fort résidu de section efficace entre 10, 93 et 11, 89 eV. DERRIEN et al.

TABLEAU I (suite)

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57,44	6,5	147	244	500	T				c)
58,84	5,5			1100	100 1 15	50 1 10	0.70	1/0	
59,22	8,37 ± 0,46	$184,0 \pm 10,1$	128 ± 8	191 ± 16	133 ± 15	50 ± 10	0,73	1/2	1
60,94	15	320	250	6000					1
63,08	1,20 ± 0,25	24,3 ± 5,2	17,8 ± 4	155 ± 17	111 ± 37	43	0,72	1/2	1
65,71	18,22 ± 0,50	372,8 ± 9,7	203 ± 6	137 ± 14	74 ± 8	49,5 ± 8	0,60	3/4	1
66,57	1,26 ± 0,25	24.6 ± 4.9	14	180			0,57		1
74,05	4,75 ± 0,20	86,1 ± 6,5	37,6 ± 2	71 ± 8	32 ± 4	37 ± 5	0,47	3/4	1
74,95	33,20 ± 1,50	576,5 ± 10,4	330,2±5	147 ± 14	84 ± 9	40,7 ± 6	0,67	3/4	1
78,95	0,16	2,6		180					
81,76	6	95,5	81	2050			0,86		[]
82,68	0,75 ± 0,15	11,8 ± 2,4	3	70			0.34		
83,52	1.2	18,7	17.9	1750			0,96		4
85,32	28 ± 4	427 ± 60	392	2300			0,94		
85.48	11.8 ± 0.36	179.7 ± 5.5	35	85 ± 10	17 ± 9		0.24		
90.75	18.45 ± 0.42	264.6 ± 6	38 ± 3	60 ± 10	9± 2	39 ± 6	0.18	3/4	1
92,97	1.05 ± 0.04	14.7 ± 0.6	2.3	57 ± 5	9	47 ± 6	0.16	1/2	
95.36	3.15 ± 0.15	43 ± 2	16.3 ± 1.5	98 ± 10	37 ± 5	58	0.39	1/2	
96.49	6.68 ± 0.26	90.1 ± 3.5	72.4 ± 2	1700 ± 350	1670 ± 400		0.81		1
100.25	5.60	72	72	6000			-,		
102,99	2.42 ± 0.08	30.6± 0.3	8 + 1	48± 5	13 ± 4	33 ± 5	0.28	1/2	
105.30	6.96 ± 0.40	86.0 ± 5	10 + 1	48 + 7	6+ 1	38 + 7	0.13	(3/4)	1
106 67	13 96 + 0 60	170.3 + 7.4	59 + 9	75+ 4	26 + 2	40 + 4	0.40	3/4)
110 38	0.66 + 0.07	7.8 + 0.8	24	43 + 16	12	30	0.31	1/9	1
114 44	0.20	0.3	1.5	10 - 10	10		0.66	1/2	
115 10	0.20 + 0.10	261 01	2,0	200			0,00		
116,10	$0,32 \pm 0,10$	0,0 ± 0,1		200	015 4 00	26	0,00	1/0]
110,03	0,41 20,13	00,1 = 1,0		100 1 0	210 = 20	40	0,00	1/2	
110,00	20,92 ± 0,60	203,91 0,9	113 20	102 2 0	43 x 3	144 I J	0,00	3/4	1
				-		• -			

c) Explique une très forte dissymétrie dans la résonance à 57,44 eV.
d) Ensemble très complexe sur lequel apparaît très nettement les deux résonances étroites. Deux résonances larges à 81,76 et 85,32 eV ne suffisent pas à expliquer la forme de la courbe. » **CN-23/**70

TABLEAU I (suite)

E (eV)	2g F 1 (me V)	σ ₀ Γ (b•eV)	σ₀Γ _f (b·eV)	Г (meV)	Γ _f (meV)	Γ _γ (meV)	ע/ח	g
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120,99	$3,70 \pm 0,15$	39,8 ± 1,6	20 ± 2	78 ± 12	39± 7	35 ± 7	0,53	1/2
123,44	0,70 ± 0,08	7,4±0,9	5	58 ± 13			0,69	
126,20	2,96±0,12	30,5 ± 1,2	8	96±10			0,27	
127,51	1,09 ± 0,12	11,1 ± 1,2	5	160 ± 30	0000 . 500		0,45	
131,75	18,26 ± 1,2	180,3 ± 12	155 ± 20	3830 ± 240	3300 ± 500		0,86	
133,78	8,38±0,30	81,5 ± 2,9	10	56± 6	7	43,5 ± 7	0,14	3/4
136,75	5,12±0,16	48,7 ± 1,5	34 ± 3	126 ± 10	88 ± 10	33 ± 8	0,73	1/2
139,28	0,18	1,7	1,2				0,71	
142,92	4,86 ± 0,21	44,3 ± 1,9	24,7 ± 3	137 ± 20	76 ± 15	56	0,58	1/2
143,47	$6,12 \pm 0,15$	$55,5 \pm 1,4$	$27,5 \pm 2$	83 ± 12	41 ± 7	36	0,53	1/2
146,25	10,58 ± 0,30	94,2 ± 2,7	17,1 ± 1	70 ± 7	13 ± 2	36	0,26	(1/4)
147,44	$1,20 \pm 0,60$	10,6 ± 5,3	7,5	1000			0,71	
148,21	0,70 ± 0,10	6,1 ± 0,9	5	150			0,82	
149,42	2,62 ± 0,12	22,8 ± 1,1	10,5 ± 2	120 ± 20	55 ± 14	62	0,47	1/2
157 , 08	17,2 ±0,4	142,5 ± 3,3	116 ± 5	670 ± 50			0,84	
160,8	0,2	1,7					·	
161,96	0,21 ± 0,07	1,7 ± 0,6		150				
164,54	42 ± 3	332 ± 22	35 ± 2	79 ± 10	8 * 1	42,7 ± 8	0,16	3/4
167,10	8,75 ± 0,40	68,2 ± 3,1	45 ± 3	112 ± 8	74 ± 7	32 ± 6	0,70	(3/4)
170,49	0,86±0,02	6,6±0,2	10.5	158 ± 60				
171,08	0,89±0,30	6,8 ± 2,3	J 10,0	1000				
174,56	0,05				1		ļ	
175,98	3,14 ± 0,10	23,2 ± 0,8	9,1	73 ± 5	29	41 ± 8	0,41	1/2
177,22	5,36±0,18	39,4 ± 1,3	3,8	51 ± 6	5	41 ± 6	0,11	1/2
178,90	1,83 ± 0,06	13,3 ± 0,5	3,2	58 ± 9	14	42 ± 7	0,25	1/2
183,64	2,29 ± 0,12	16,2 ± 0,08	5				0,33	
184,87	10 ± 2	70,4 ± 14	50 ± 10	2200 ± 200	1570 ± 500		0,71	

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188,27	0,92 ± 0,07	6,4 ± 0,5	1,3	53 ± 10	11	41	0,21	1/2		
190 ,64	2,51 ± 0,09	17,1 ± 0,6	2,2	67 ± 9	9	56	0,13	1/2		
195,36	30 ± 1,2	200 ± 8	157 ± 10	447 ± 40	350 ± 42	36 ± 12	0,91	1/4		
196,69	7,04 ± 0,42	46,6± 2,8	24,4 ± 3	112 ± 18	59 ± 12	46 ± 11	0,56	1/2		
199,39	14,49 ± 0,75	94,6±4,8	64 ± 3	133 ± 13	90 ± 10	33 ± 8	0,73	3/4		
203,46	6	38,4	17 170 5 1 0	200						
203,93	26,6 ±1	169,7 ± 6,4	1/0,5 x 0	430						
207,37	10,5 ±0,4	65,9 ± 2,5	7,4 ± 1,5	57 ± 5	7 ± 2	43,6 ± 5	0,14	3/4	(
211,09	1,4	8,6	8,5	800			-			
212,02	1,2	7.4	6,7	1500					e)	
213,28	0,7	4,3	3,5	200 ± 60			0,82			
216,53	9,40 ± 0,40	56,5 ± 7	8,1	67 ± 7	10	39 ± 6	0,20	(1/4)		
219,49	5,36 ± 0,27	31,8±1,6	17,7	70 ± 10		ļ	0,60			
220,22	11,14 ± 0,50	65,8 ± 1,5	6,2	45 ± 8	4	33	0,11	(3/4)		
223,16	5,12 ± 0,15	29,8 ± 1	1	59 ± 6			0,37		(2
224,89	2,56 ± 0,15	14,8 ± 0,9	2	85 ± 17			0,14			2
227,77	17	97	76	6 eV			0,78		f) 5	3
227,89	2,54 ± 0,15	14,5 ± 0,9	8	67 ± 10	37	28	0,57	1/2		ŝ
231,40	17,8 ±1,0	100,1 ± 5,6	9	43 ± 8	4	27	0,12	3/4		2
232,63	0,66 ± 0,15	3,7 ± 0,2	2,5	.130			0,68			
234,32	15,35 ± 0,60	85,2 ± 3,3	16	74 ± 9	14	45	0,24	1/2		
239,04	8,15 ± 0,40	44,4 ± 2,2	10,5	73± 8	17	47	0,27	1/2		
240,60	0,05									
242,88	9,92±0,45	53,2 ± 2,6	32 ± 3	97 ± 6	58 ± 6	32 ± 7	0,65	3/4		
247,50	1,39 ± 0,20	7,3 ± 1,2	6	312 ± 60]	0,82			
248,86	22,13 ± 0,72	115,7 ± 3,7	10	62 ± 6	6	41	0,12	3/4 ,		
251,23	41,2 ±1,2	213,4 ± 6,3	36,4 ± 3	83 ± 5	14 ± 2	41 ± 7	0,25	3/4		
					1	J		[
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e) Explique une très forte dissymétrie dans la résonance à 212,02 eV.

f) Explique un très fort résidu de section efficace entre 225 et 228 eV. Il y a peut-être deux résonances larges dont les largeurs totales sont de l'ordre de 4 à 5 eV.

E	2gr	r	E	²gr_	Г
(eV)	(meV)	(meV)	(eV)	(me V)	(meV)
(,	(((01)	(1107)	(mev)
254,58	4,2 ±0,3	55 ± 10	350,30	32,25 ± 0,90	97 ± 6
256,11	9,5 ±0,5	91 ± 16	352,82	5,84 ± 0,24	69 ± 13
259,00	0,4 ±0,10		354,89	0,60 ± 0,10	100
262,37	50	6100	357,87	4,5	6000
262,74	3,64 ± 0,45	60 ± 10	359,99	1,66 ± 0,18	114 ± 20
264,23	0,25		361,28	0,33 ± 0,10	
269,11	2,5 ± 1	160	366,00	4,9	5000 `
269,54	5,8 ±0,6	72 ± 20	368,33	0,60 ± 0,20	
272,62	41,8 ± 1,5	92 ± 10	370,31	3,90±0,24	105 ± 20
274,80	14,0 ± 2,0	800	371,72	11	3400
275,57	35,1 ±1,5	150 ± 30	375,02	4 ± 0,3	43 ± 12
277,23	8 '	5300	377,10	2,98 ± 0,30	101 ± 20
279,59	10,64 ± 0,33	111 ± 8	378,04	1,88 ± 0,30	223 ± 60
282,92	-37,8 ± 2,7	85 ± 6	382,43	0,63 ± 0,15	130
285,73	0,10		384,26	8,55 ± 0,50	109 ± 30
287	50	>6 eV	385,90	2	> 1 eV
288,30	0,08		389,51	2,09 ± 0,18	74 ± 14
292,33	5,82 ± 0,24	115 ± 13	391,52	1,89±0,18	142 ± 28
296,46	4,89±0,21	81 ± 12	394,43	9,78±0,33	106 ± 13
298,59	15,80 ± 0,60	74 ± 7	396, 91	3,17 ± 0,21	108 ± 20
301,81	27,3 ± 1,0	108 ± 6	401,56	29,1 ± 1,2	220 ± 20
308,20	4,40 ± 0,27	150 ± 30	404,24	34,8 ± 1,3	178 ± 16
309,01	21,10 ± 0,72	85 ± 12	406,03	2,73 ± 0,60	320
311,12	0,73 ± 0,18		406,95	1,46 ± 0,60	330
313,62	20,40 ± 0,72	62 ± 6	408,71	1,94 ± 0,30	150
316,66	7,75 ± 0,30	73 ± 10	412,31	13,41 ± 0,63	145 ± 15
321,75	0,20 ± 0,08		415,66	4,89 ± 0,36	152 ± 30
323,36	30,2 ±0,9	160 ± 16	417,60	2,57 ± 0,36	267 ± 64
325,30	$12,8 \pm 0.5$	105 ± 9	419,85	9,11 ± 0,45	139 ± 25
329,65	1,5	1500	425,67	0,40	
333,91	8,24 ± 0,30	67 ± 7	428,33	40	> 6 eV
335,93	26,60 ± 0,72	83 ± 6	429,64	5,66 ± 0,81	780
337,95	12,09 ± 0,42	74 ± 7	432,73	1,54 ± 0,30	00.1.15
339,24	$4,92 \pm 0,21$	81 ± 12	437,76	4,04 ± 0,30	62 ± 15
343,18	23,69 ± 0,78	75 ± 6	438,72	4,36 ± 0,30	61 ± 15
346,56	5,2 ±0,6	1200	440,07	$0,42 \pm 0,15$	
			442,41	10,5 ± 0,5	422 ± 50
	1	1	11	1	1

TABLEAU II. PARAMETRES DE RESONANCES DU ²³⁹Pu

sont moins apparentes. Dans certains cas leur présence est nécessaire pour expliquer de très fortes dissymétries, ou d'importants résidus de section efficace. Il semble peu probable que de telles dissymétries et de tels résidus de sections efficaces soient explicables par de seuls effets d'interférences dues à la fission. Aussi, dans l'interprétation de nos résultats d'analyse, nous admettons que ces résonances larges sont réelles. Une étude par un programme multiniveaux est en cours, qui nous permettra probablement de vérifier si une telle hypothèse est justifiée.

Energie (eV)	ie Largeur Largeur accélérateur sélecteur (µs) (µs)		Echantillons utilisés	Distance de vol (m)	
4 à 13 13 à 40 40 à 79	0,1 0,1 0,1	0,8 0,4 0,1	5 échantillons	53,7	
70 à 200	0,1 et 0,05	0,05	4 échantillons	53,7	
150 à 700	0,06	0,05	1 à 4 échantillons et 14 g/cm ²	103,7	

TABLEAU III. CONDITIONS EXPERIMENTALES

Pour l'évaluation de l'effet Doppler, le calcul de la température effective peut être fait en supposant que le réseau cristallin des échantillons suit la loi de Debye. La température de Debye du ²³⁹Pu métallique est voisine de 175°K [6,7]; la température effective obtenue est de 95,5°K pour une température réelle de 77°K. On obtient ainsi la largeur Doppler

$$\Delta = n\sqrt{E} = 0.0117 \sqrt{E} eV$$

Pour préciser cette valeur, des mesures complémentaires de transmission à température ambiante ont été faites sur quelques résonances isolées; les résultats ont montré que le coefficient η est sous-estimé. La valeur de η retenue pour l'analyse des courbes est égale à

$$\eta = 0,0122 \pm 0,0003$$
 (1bis)

2.1. Identification des niveaux

Les échantillons contenaient peu de 240 Pu; les résonances de cet isotope ont pu être repérées par comparaison de la transmission de l'échantillon de 14 g/cm² à celle d'un échantillon d'oxyde de Pu contenant 13 g/cm² de 239 Pu et 1, 3 g/cm² de 240 Pu. Les niveaux du 239 Pu ont été identifiés jusqu'à 440 eV. L'histogramme de la figure 1 est parfaitement rectiligne jusqu'à 300 eV et il semble que de 300 à 440 eV on perde une dizaine de niveaux (en se référant à la pente de la droite de 0 à 300 eV).

Jusqu'à 300 eV l'espacement moyen est de 2,39 eV, et de 0 à 440 eV il est de 2,47 eV. La figure 2 montre la distribution des espacements de 0 à 300 eV. Elle est compatible avec la superposition non corrélée de deux lois de Wigner dans le rapport 3 (rapport des populations correspondant aux deux états 0+ et 1+). On remarquera la présence de trois grands espacements dont la probabilité d'occurrence est pourtant faible.

(1)



FIG.1. Nombre de niveaux en fonction de l'énergie



FIG. 2. Distribution des espacements de niveaux

2.2. Largeurs neutroniques et fonction densité

La figure 3 représente $\sum_{0}^{E} 2g\Gamma_{n}^{0}$ en fonction de E. L'examen de cet histogramme montre que les fluctuations des fonctions densité locales sont assez importantes; la pente de la droite, déterminée par une méthode de moindres carrés, donne pour la fonction densité la valeur suivante:

$$S_0 = (1, 33 \pm 0, 14) \cdot 10^{-4}$$
 (2)


FIG.3. $\Sigma_0^{E} 2g \Gamma_n^0$ en fonction de E

Cette valeur est calculée sur 180 résonances; l'influence des erreurs sur les paramètres est très faible devant l'erreur d'échantillonnage: $(2/N)^{1/2}$ en valeur relative [8], N étant le nombre de résonances.

Dans les intervalles d'énergie de 100 à 200 eV et de 200 à 300 eV, les valeurs trouvées pour S_0 sont respectivement 1, $13 \cdot 10^{-4}$ et 1, $60 \cdot 10^{-4}$; elles sont en très bon accord avec celles obtenues par Uttley à partir des sections efficaces moyennes dans les mêmes intervalles d'énergie (1, $15 \cdot 10^{-4}$ et 1, $54 \cdot 10^{-4}$) [9]. Cela semble bien indiquer que les résonances larges qui ont été introduites dans l'analyse des résultats suffisent à expliquer les très forts résidus de section efficace dont il est fait mention plus haut.

La figure 4 montre un assez bon accord entre la distribution expérimentale des largeurs neutroniques réduites et la loi de Porter et Thomas. Toutefois, l'accord est meilleur en supposant que 5% de petits niveaux ont été omis. La valeur de $2g\Gamma_n^0$ pour ces niveaux serait inférieure à 0,003 eV $^{1/2}$.

2.3. Largeurs de fission

Les valeurs de Γ_f ont pu être déterminées pour une centaine de résonances. Pour les résonances très larges, les paramètres sont déterminés avec assez peu de précision. On peut admettre que pour ces très grandes valeurs de Γ , Γ_f est peu différent de $\Gamma(\Gamma_n + \Gamma_\gamma)$ est négligeable devant Γ pour des largeurs totales égales ou supérieures à 1 eV). L'histogramme de la figure 5 met nettement en évidence deux familles de résonances dont les valeurs de $\langle \Gamma_f \rangle$ sont très différentes. Pour la première famille $\langle \Gamma_f \rangle = 42$ meV, valeur déterminée avec assez de précision; pour la deuxième famille $\langle \Gamma_f \rangle$ est supérieur à 1 eV, probablement de l'ordre de 1,5 eV. L'histogramme expérimental est assez bien décrit par une somme de deux distributions en χ^2 dont les



FIG. 4. Distribution des largeurs neutroniques réduites



FIG. 5. Distribution des largeurs de fission

nombres de degrés de liberté sont $\nu = 1$ pour les resonances étroites et $\nu = 2$ pour les résonances larges. Ces valeurs ne sont données qu'à titre d'estimation; on peut admettre cependant que pour la famille des résonances larges, on a probablement $1 < \nu < 2$.

La population des niveaux étant proportionnelle à 2J+1 (en négligeant le facteur exponentiel exp $[-J(J+1)/2\sigma^2]$), il y a trois fois plus de résonances pour l'état de spin 1+ que pour 0+. Les espacements moyens sont donc respectivement 3,2 et 9,6 eV. La théorie des voies de sortie de la fission [10] permet de relier $\langle D \rangle$ et $\langle \Gamma_f \rangle$ au nombre effectif N_{eff} des voies de sortie:

$$\langle \Gamma_{\rm f} \rangle / \langle D \rangle = N_{\rm eff} / 2\pi$$
 (3)

Ainsi on obtient

$$N_{eff}(0+) = \frac{1,5}{9,6} \cdot 2\pi \simeq 1$$
(4)

$$N_{eff}(1+) = \frac{0,042}{3,2} \cdot 2\pi \simeq 0,08$$
(5)

Ces résultats sont assez compatibles avec les distributions expérimentales de $\Gamma_{\rm f}$ pour l'état de spin 0+ et pour l'état de spin 1+ (voie partiellement ouverte).

2.4. Largeurs totales de capture radiative

Les courbes expérimentales donnent directement $2g\Gamma_{n},\ \Gamma_{\gamma}$ a pour valeur

$$\Gamma_{v} = \Gamma - \Gamma_{f} - (2g\Gamma_{n})/2g \tag{6}$$

On en déduit que si Γ_n est assez grand, Γ_γ ne peut être déterminé d'une facon précise que si l'on connaît le facteur statistique g, c'est-àdire si le spin de la résonance a été attribué. D'autre part, pour les résonances très larges, Γ et Γ_f sont du même ordre de grandeur et la détermination de Γ_γ est pratiquement impossible. Différents auteurs ont fait des attributions de spin aux résonances du ²³⁹Pu [11-15] (voir tableau IV). On peut également essayer d'attribuer le spin en choisissant l'une ou l'autre des valeurs de g et en examinant les valeurs de Γ_γ obtenues; dans le paragraphe suivant nous examinerons ce cas. Dans la liste des Γ_γ du tableau I, pour les résonances dont le spin n'est pas attribué, le facteur statistique est pris égal à 1/2.

La valeur moyenne de Γ_{y} évaluée à partir de 59 niveaux est

$$<\Gamma_{v}>=41,6 \text{ meV}$$
 (7)

Si on l'évalue uniquement à partir des résonances dont le spin 1+ est connu avec certitude, on trouve

$$<\Gamma_{v}>=40,5 \text{ meV}$$
 (8)

Il n'est pas possible de calculer une valeur moyenne de Γ_{γ} pour l'autre état de spin.

L'ensemble des valeurs obtenues pour les 59 niveaux semble compatible, compte tenu de barres d'erreurs, avec une valeur unique. Mais un raisonnement rigoureux doit être fondé sur le calcul des variances. La distribution expérimentale est cohérente avec une distribution en χ^2 à 60 degrés de liberté, ce qui correspond à une variance expérimentale

$$\sigma_{\exp}^2 = \frac{2}{\nu_{\exp}} = \frac{2}{60} = 0,033$$

(9)

	(1958)	Schwartz (1962)	lgnatev (1964)	Sauter et Bowman (1965)	Asghar (1966)	Cowan (1966)	Saclay (1966)	Energie	Asghar (1966)	Saclay (1966)
0.3	1							05.4		
7.9	1	1		1		2		90,4	U	٥
10.9	1	1		1				100.2		0
11.5	1	-		-			0	100,2	1	0
11.9	1	1		1			v	105,0	1	
14.3	· ·			î .				106 6	(I)	1
14.7	0	0		1	1			116		-
15.4	n n	, v		n n	• •		0	118 7	ő	1
17.6	1	1 1	1	1 1	1	(-/	, ř	126.2	ő	•
22.3		0		1	1	1		131 7	0	n
26.3		-	ł	-	Ô	1	}	133 7	. 1	v
32.4				0		(0)		136.7	ō	
35.8				1				146 2	0	(0) >
41.51	0	1	(0)	1	1	l 1 .		147.4	-	0
41.7		-		-	-			157.1	0	0
44.5	1	0	a	1	1	1 1		164.5	1	1
47.7	-			0	0	1		167.1	0	(1)
49.8						0	0	171.1		0
50.2					1	1		177.2	1	
52,6	1		0	, 1	1	1		184.9	_	0
55.7			4	1	0	1		196,7	0	
57.6						0	0	199.4	0	1
58.8		· · ·				0	· 0	207.4	1	(1)
59.3					0	1		211.1		0
60,9						0	0	216.5	0	(0) ?
65.7		1	0		1	1	(1)	220,2	· _	1
66,6		1				0		223.2	0	-
74.1	· ·				1			227,8		0
74,9			1	1	1	1	1	231,4	1	1
81,7			1			0	0	234,4	0	
83,5		1		1	ł		o i	239,1	0	(0) ?
85,3]	· ·		l .			o ·	242,9	0	1
85,5]		· · · · ·		o ^a			248,9	1	. 1
90,7	1		1		1 ^a		1	251,2	1	. 1

TABLEAU IV. TABLEAU RECAPITULATIF D'ATTRIBUTION DE SPIN DES RESONANCES DU 239 Pu

^aValeurs non publiées par Asghar, mais déduites de ses valeurs de $\sigma_0\Gamma_n$ et des valeurs de $2g\Gamma_n$ et Γ de Saclay.

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Cette variance est la somme de la variance réelle et de la variance due aux erreurs expérimentales. Cette dernière est assez difficile à évaluer. Si nous admettons un écart σ de 10%, on obtient

$$2/\nu_{\rm réel} = 2/\nu_{\rm exp} - \sigma^2 = 0,023$$
 (10)

soit $v_{réel} = 90$.

TABLEAU V. RESONANCES DONT LE SPIN A PU ETRE ATTRIBUE

Energie	J	Γ(1+)-	Γ _γ (0+)	Energie	J	Γ _γ (1+)	Γ _γ (0+)
11,50 ^a	0			157,08	0		
15,46	0			164,54	1	42,3±8	-13
49,71	0			167,10	(1)	32 ± 6	20,5±4 ·
57,44	0			171,08	0		
58,84 ^a	0			184,87	0		
60,94	0						
65,71	(1)	49,5 ± 8	24,4 ± 4	199,39	1	33,3 ± 8	14 ± 4
74,95	1	40 ± 6	-4,4	207,37	(1)	43,6±5	29 .6 ± 3
81,76	0			211,09 ^a	0 -		ļ
83,52 ^a	0			216,53	?	51 ± 8	39 ± 6
85,32	0						
90,75	1	39 ± 6	14,5 ± 2,2	220,22	1	33,3	19,4
96,49	0			227,77	0		
100,25	0			231,40	1	27,5	3,5
106,47	1	40 ± 4	21 ± 2	239.1	\$	50	39
118,83	1	42 ± 5	7,4 ± 1	242,88	1	32 ± 7	19 ± 4
131,75	0						
146,25 ⁸	?	50,2 ± 7	36 ± 5	248,86	1	41,3 ± 7	12 ± 3
147,44	0			251,23	1	41,4 ± 7	-13

a Résonances expliquant de très fortes dissymétries ou d'importants résidus de sections efficaces.

2.5. Essai d'attribution de spin

Cette attribution peut être faite de deux façons différentes:

a) Par l'étude de la distribution des Γ_f . D'après l'histogramme de la figure 5, les résonances dont Γ_f est supérieur à 400 meV ont une probabilité pratiquement égale à 1 d'appartenir à la famille qui a une grande valeur de $\langle \Gamma_f \rangle$. A ces résonances on peut attribuer le spin 0+.

b) Cas des résonances à grandes valeurs de Γ_n . Les deux facteurs statistiques g sont 1/4 et 3/4. On suppose que Γ_{γ} est à peu près constant de résonance à résonance et ne dépend pas du spin. La valeur de Γ_{γ} doit être voisine de 40 meV. Dans certains cas il n'y a aucune ambiguîté; ainsi à 74,95, 164,54 et 251,23 eV l'attribution du spin 0+ conduirait à une valeur négative de Γ_{γ} ; ces résonances sont donc obligatoirement de spin 1+. Dans les autres cas, si l'une des valeurs de Γ_{γ} est voisine de 40 meV et l'autre très différente, il est également possible de se prononcer.

Dans le tableau V nous donnons les résonances dont le spin a pu être attribué.

CONCLUSION

En définitive, il semble que les propriétés du noyau du ²³⁹Pu sont bien celles auxquelles on doit s'attendre d'après la théorie des voies de fission; les deux familles de résonances correspondant aux deux états de spin possibles sont caractérisées par des valeurs de $\langle \Gamma_f \rangle$ très différentes, compatibles avec des seuils de fission très différents; le nombre de voies de sortie pour chaque état de spin est faible, et plus grand pour l'état 0+ que pour l'état 1+. D'autre part, la distribution en masse des produits de fission faite par Cowan [12] permet d'arriver aux mêmes conclusions: la fission est plus symétrique dans les résonances larges que dans les résonances étroites, et la théorie prévoit effectivement une plus grande symétrie dans l'état 0+ que dans l'état 1+.

En fait, l'étude des résonances du ²³⁹Pu ne peut être complète que par l'utilisation d'un formalisme multiniveaux. Si les effets d'interférence perturbent probablement peu les résultats pour les résonances de spin 1+, il risque de ne pas en être de même pour celles de spin 0+[16]; en particulier, l'étude des ensembles complexes tels que celui qui existe entre 70 et 90 eV peut être intéressante.

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SPIN MEASUREMENTS OF THE 8.8-eV AND 12.4-eV NEUTRON RESONANCES IN ²³⁵U

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Abstract

SPIN MEASUREMENTS OF THE 8.8-eV AND 12.4-eV NEUTRON RESONANCES IN ²³⁵U. Spin measurements of slow neutron resonances have been done by means of the resonance scattering method, using a ³He counter as scattering detector and a crystal spectrometer as neutron source. Results for the 8.8-eV and 12.4-eV resonances in ²³⁵U are given and discussed.

INTRODUCTION

The only information available at present about the spins of slow neutron resonances in 235 U are the recent results of Sailor et al.[1] for the three lowest resonances. A knowledge of the resonance spins combined with that of the fission widths, of the mass yield distribution in the resonances [2,3] and of the fission fragment directional anisotropies from oriented nuclei [4] could yield important information about the fission process, especially about the number and the nature of the fission channels available in 235 U+n.

The BR2 crystal spectrometer has been used as a monochromatic neutron source for the measurement of the ratio Γ_n / Γ in the 8.8-eV and the 12.4-eV resonances. From the known parameters $g\Gamma_n$ and Γ the spin J has been deduced for these resonances. One has of course to suppose that they are single resonances.

APPARATUS AND METHOD OF ANALYSIS

The scattering chamber with the ³He neutron detector has been described elsewhere [5]. The method of analysis has been explained in Refs. [5, 6]. Essentially the scattering counting rate at resonance energy is measured and compared with the counting rate from a standard sample such as lead with known scattering cross-section. The samples used are fairly thin, so only the experimental transmission of the samples, which is measured simultaneously, is needed for the absorption correction.

The targets used in all the experiments are laminated metallic sheets and have the following isotopic composition: 9.02% ²³⁸ U; 0.60% ²³⁶ U; 89.43% ²³⁵U; 0.95% ²³⁴U¹. A 2-mm thick ²³⁸U filter has been placed in the diffracted beam so as to avoid the scattering of neutrons in the ²³⁸U resonances at higher order energies.

¹ The base material was supplied by Oak Ridge National Laboratory, USA.

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The main experimental difficulties are the following:

(a) Background from fission neutrons

The fission neutrons can contribute to the scattering counting rate. This contribution has been determined in two ways. We measured first the scattering from a thin 235 U target (n = 12.7×10^{20} 235 U atoms per cm²) at 0.1 eV with a 1-mm-thick Cd shield between target and detector. The fission neutron contribution N_f can be written as follows:

$$N_f = N_0 \epsilon_f (1 - T) \frac{\sigma_f}{\sigma_r}$$

where N_0 = number of incident neutrons; it can be deduced from the scattering counting rate of a standard scatterer if one

calculates the efficiency of the scattering detector.

 ϵ_{f} = efficiency for fission neutrons.

T, = transmission.

 σ_f and σ_t = fission and total cross-sections. This measurement yielded:

$$\epsilon_{f} = (2.0 \pm 0.1) \times 10^{-3}$$

We also measured the scattering counting rate from the same target between 0.5 eV and 0.29 eV. In this energy range, $n\sigma_f$ varied from 0.10 to 0.24. After correction for absorption and subtraction of the contribution from the negative energy resonance, the data were fitted to:

$$n\sigma_s = n\sigma_n + \epsilon_f n \sigma_f$$

and we obtained:

 $n\sigma_p = (17.11 \pm 0.08) \times 10^{-3}$ $\epsilon_{\epsilon} = (2.0 \pm 0.24) \times 10^{-3}$

The measurements were performed using vanadium instead of lead as a standard scatterer so as to avoid any effect from coherent scattering. The contribution from the negative energy resonance has been calculated using the parameters given by Harvey and Sanders [7]. If the error on σ_s for V is taken into account [8], the following potential scattering cross-section for ²³⁵ U is deduced:

 $\sigma_{\rm p} = 11.9 \pm 0.4 \ {\rm b}$

in good agreement with the value obtained by Uttley [9] $(11.7 \pm 0.1 \text{ b})$.

(b) Non-resonant scattering contribution

Because of the low resolution of our spectrometer ($\Delta E/E = 0.07$ at 10 eV) and the small values of Γ_n/Γ , the ratio of resonant-to-potential scattering is rather low, so a precise determination of the non-resonant

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scattering cross-section is necessary. This has been done by measuring the scattering counting rate between 9.4 eV and 10.4 eV with a target of thickness n = 6.68×10^{20} atoms of 235 U per cm². In this energy range, the mean value for $n\sigma_p = (8.6 \pm 0.2) \times 10^{-3}$. After correction for absorption (the mean transmission in this energy range is 0.97) and subtraction of the small contributions from the most important neighbouring resonances



FIG.1. Net scattering counting rate and transmission between 7 eV and 13 eV with a target containing 6.68×10^{20} ²³⁵U atoms per cm². The crosses represent the scattering counting rates at resonance energies (8.8 eV and 12.4 eV) after correction for absorption. The correction for fission neutrons has not been subtracted; on the scale of Fig.1 it represents 16 counts at 8.8 eV and 8 counts at 12.4 eV.

(8.8 eV, 11.6 eV and 12.4 eV) the following value for σ_p of $^{235}\,U$ has been obtained:

$$\sigma_{\rm p} = 12 \pm 0.3 \, \rm b$$

in good agreement with the value quoted above.

RESULTS AND DISCUSSION

The net scattering counting rate and the transmission between 7 and 14 eV are shown in Fig.1. The total background which is only 25% of the potential scattering counting rate has been subtracted from the data. The counting rates on Fig.1 have been reduced to a fixed counting rate from the Pb target, so they are corrected for the changes of the detector efficiency and of the beam intensity. The measurements were performed using an automatic sample changer; the scattering signals from 235 U, from Pb and from the blank position of the sample

changer being measured alternatively during approximately 10 min. The data shown are the sum of different runs; each run consisted of 20 measurements at each energy. The results between 9.4 and 10.4 eV are the sum of 2 runs and the few points at or near resonance energies are the sum of 5 to 6 runs.

For the 8.8-eV resonance, a preliminary result has been presented at the Washington conference [5]. The efficiency for fission neutrons quoted at that time has been confirmed by the experiment in the 0.3-eV resonance described above and the statistical accuracy has been improved by a few additional runs at 8.8 eV. We now get the following result:

$$\Gamma_{\rm n} / \Gamma = (1 \pm 0.13) \times 10^{-2}$$

$$g = 0.46 \pm 0.08$$

so that J = 3. Only the scattering signal at 8.79 eV has been taken into account. The result quoted previously [5] ($g = 0.44 \pm 0.07$) has been obtained, taking into account the scattering signals at 8.73 eV, 8.79 eV and 8.85 eV.

For the 12.4-eV resonance we have the following result:

$$\Gamma_n / \Gamma = 1.46 \pm 0.25$$

$$g = 0.64 \pm 0.13$$
 so that $J = 4$.

In this case the scattering signals at 12.33 eV and 12.43 eV were taken into account in the analysis and the contribution from the 11.67-eV resonance was subtracted. This contribution, being only 7% of the total scattering counting rate, was calculated from the known Breit-Wigner parameters, not corrected for resolution and Doppler broadening. The parameters $g\Gamma_n$ and Γ are the recommended values of Rep. BNL 325[10].

The fission widths $\Gamma_{\rm f}$ for the 8.8-eV and the 12.4-eV resonances are 74 meV and 23 meV, respectively, but the number of resonances with known J-value being too small for the moment, it is not yet possible to draw any firm conclusion about the spin dependence of the fission width for ²³⁵U. As for the mass yield distribution, the results presently available [3] are not yet accurate enough to detect a correlation between this distribution and the resonance spins.

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DISCUSSION

(on papers CN-23/128, CN-23/70 and CN-23/79)

A. MICHAUDON: Could Dr. Asghar say whether there is some systematic reason why Frazer's measurements on 8 resonances in 1961¹ gave 3 resonances corresponding to spin 0, while the recent Harwell measurements associate spin 1 with them?

M. ASGHAR: Frazer et al. made their scattering measurements using a 10 B sleeve around the scattering sample. As assignments of spin 0 imply large resonance scattering areas, they may not have been able to take sufficient account of contributions from fission neutrons.

A. MICHAUDON: Dr. Poortmans showed that his measurements are very sensitive to any error in the potential energy measurement. Since the elastic scattering cross-section changes very quickly near the resonance energy E_0 , I should like to know whether there is any error due to the uncertain resonance energy E_0 or perhaps due to the resolution.

F. POORTMANS: I don't think so because with our poor resolution the scattering count rate at resonance energy does not change very much. Moreover, the energy can be well determined with the crystal spectrometer.

J. RYABOV: In connection with Dr. Asghar's paper, I should like to mention that in a previous work it has been shown that if the background I_p is above a certain level it is necessary to take account of the reduction in the number of neutrons undergoing potential scattering, as a result of resonance interaction: this may lead to a too high background in calculating the area from the expression $I_s = (I_{exp} - I_p)/(1 - \Omega)$, where I_{exp} is the experimental resonance count, I_p is the potential scattering background, I_s is the resonance scattering count, and Ω is a factor depending on various constants, the density of the sample, Γ_n and Γ . In Dr. Asghar's measurements Ω may attain a value of ~ 0.75 for certain resonances.

In connection with Dr. Poortman's paper, I want to mention that the results of the ²³⁵U resonance spin measurements for $E_0 = 8.7$ eV and 12.4 eV do not agree with the results from Melkonian's measurements of total kinetic energy for these resonances nor with the identical results of our own calibration measurements on the variation in $\overline{\nu}$.

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Session IX

COMPARISON OF FISSION CROSS-SECTIONS IN THE RESONANCE ENERGY REGION

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NEW TIME-OF-FLIGHT MEASUREMENTS MADE WITH AN INTENSE SOURCE*

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Abstract

NEW TIME-OF-FLIGHT MEASUREMENTS MADE WITH AN INTENSE SOURCE. In an experiment in Nevada in June 1965 a nuclear device with a yield equivalent of 1.2 kilotonnes of TNT provided the neutron source for time-of-flight measurements over a path of 185 m in vacuo. To exploit the combination of high flux and high energy resolution, new recording techniques have been required. Because more than a million data points are acquired in any one exposure of a set of targets, the general problems of data retrieval and processing have required special attention.

Measurements of fission cross-sections of the nuclides ²³³U, ²³⁵U, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴²Am are reported. In addition, capture-to-fission ratios of ²³³U and ²⁴⁰Pu are reported. The neutron energy range is 10 eV to 2 MeV. Individual resonances are resolved in the 100-eV range. Fission data in the resonance region are characterized by lower minima than are reported by most earlier investigators, indicating more favourable signal-to-background ratios. A unique feature of these experiments is the high rate of data acquisition, which allows cross-section measurements on short-lived nuclides. Even for the long-lived nuclides, these experiments provide an abundance of data required in current nuclear technology - data that could otherwise be acquired only by years of tedious measurement,

Introduction

Several preliminary experiments at the Nevada Test Site of the U. S. Atomic Energy Commission afforded the opportunity to develop instrumentation for time-of-flight measurements on neutrons from a nuclear detonation and to find typical levels of background signals. An experiment in December 1964 [1] pointed the way to measuring many fission cross-sections over a wide energy range. It is my purpose here to outline certain details of an experiment on 11 June 1965 and to present crosssection results of that test.

The source of neutrons was a nuclear explosion with an energy yield equivalent to 1.2 kilotonnes of chemical explosive and a neutron yield of 1.8 x 10^{23} with a duration of 0.1 µsec. It was located in desert alluvium at a depth of 185 m. A vertical pipe 35 cm in diameter evacuated to a pressure of 0.050 mmHg provided a flight path for neutrons. The pipe, shown in Fig. 1, contained many anti-scattering baffles and, at the lower end, a 10-cm thickness of polyethylene moderator. Near ground surface was a collimator 122 cm long, from which emerged a beam of neutrons 1.9 cm in diameter. The volume between the pipe and the casing of the drilled hole was filled with sand. The pipe was provided with several closure devices timed to allow transit of neutrons with energy greater

^{*} Work performed under the auspices of the United States Atomic Energy Commission.

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FIG.1. Line-of-sight pipe to provide vacuum flight path for neutrons originating in a nuclear explosion, showing eleven anti-scattering baffles and a two-holed collimator, and two charged particle detectors.



FIG. 2. View of moderator and anti-scattering baffles, looking through collimator.



FIG. 3. Moving film oscillogram of six signals plus 20-µsec time marks.



FIG.4. The number of neutrons emerging from the collimator, in neutrons/eV as a function of energy.

than 1 eV but to confine any flying debris or gaseous products of the explosion. Figure 2 shows a photograph of a set of concentric circles at the top of the moderator, taken through a telescope. In addition to the moderator, one can see the anti-scattering baffles. The view is through one of two holes in the collimator, each one several centimeters from center and designed to provide irradiation for two different sets of experiments.

A moderator is used because there would be relatively few neutrons below a few hundred eV without it. The moderator HEMMENDINGER



FIG. 5. Fission cross-section of ²³⁹Pu, 20 to 40 eV, [4] showing comparison with results of Bollinger [11] (+), Ignat'ev [12] (*), James [13] (x), and de Saussure [14] (0).



FIG.6. Fission cross-section of ²³⁹Pu, 40 to 60 eV, [4] showing comparison with results of Bollinger [11] (+), Ignat'ev [12] (*), James [13] (x), and de Saussure [14] (0).



FIG. 7. Fission cross-section of ²³⁹Pu, 500 to 1000 eV, [4] showing comparison with results of Bollinger [11] (+), and de Saussure [14] (0).







FIG. 9. Fission and capture cross-section of ²⁴⁰Pu, 600 to 1000 eV [5]

acts like a source of 20-eV thermal neutrons, but it has a sufficient velocity so that neutrons are thrown into a cone directed up the pipe with energy peaked at 45 eV.

In order to complete these time-of-flight experiments, careful investigation was required to develop logarithmic amplifiers [2] with suitable response to fast transients, to maximize the resolution of oscilloscope traces photographed on moving film, to maximize signal-to-noise ratios, to determine backgrounds, to protect equipment against ground shock, to understand transient response of solid state detectors, to



FIG.10. Fission cross-section of 240 Pu, 10³ to 2×10⁶ eV [5]



FIG.11. Fission cross-section of 241 Pu, 20 to 74 eV [6]. The solid curve represents a multi-level fit to the data.

establish the absolute sensitivity of the detectors to fission fragments, to establish suitable calibration procedures for time and for signal current, and to find schemes for digitizing the analog signals and processing the data to produce crosssections. These instrumental details are discussed in a report now in preparation.

It has proved convenient to record signals from several oscilloscopes on one film. It is possible to read the deflection produced by a single fission, but an experiment is arranged so that 10^3 or more fission fragments fall on the counter in one resolution interval. The plot of detector current against time can then be transformed to a product of



FIG.12. Fission cross-section of 241 Pu, 44 to 74 eV, [6] showing comparison with results from linac work at Rensselaer [15] (0) and Harwell [16] (Δ).



FIG. 13. Fission cross-section of ²³³U, 20 to 40 eV [7]

fission cross-section and neutron flux plotted as a function of neutron energy. In Fig. 3 is shown a record in which six signals from solid state detectors are displayed on three double beam oscilloscopes and photographed with 20:1 reduction on a 35-mm film; a time base is provided by moving the film at 30 m/sec. The peaks in the curves of Fig. 3, after background has been subtracted, are identified as fission resonances. Time marks from a small oscilloscope are also displayed. To get adequate resolution at energies above 1 keV, a repetitive 20-µsec long sweep photographed on film moving at 30 m/sec is required. Development is in progress of an alternative scheme for recording seven double beam oscilloscopes on film moving at 300 m/sec.

Results

To display all of the cross-section results would take too much space and time, and would serve little purpose. There are some 20 000 data points, and 85 figures would be required. The results have been published in a preliminary

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FIG.14. Fission and capture cross-sections for ²³³U, 45 to 65 eV [7]. The capture data are derived from measurement of the (relative) capture-to-fission ratio.



FIG.15. Fission cross-section of 25 U, 20 to 40 eV. [8] showing comparison with results of Adamchuk [20] (A), Yeater [21] (Y), Brooks [22] (+), Bowman [9] (*), and Michaudon [23] (o).



FIG.16. Fission cross-section of 235 U, 200 to 400 eV, [8] showing comparison with results of Yeater [21] (Y).



FIG. 17. Fission cross-section of ²⁴¹Am, 20 to 40 eV [10, 24]

form [3-8] and both listings and plots of the data appear in a recent report. [10] I will confine this presentation to a discussion of enough salient features of the data to indicate what was done and what are the possibilities for future work.

The neutron flux (in neutrons/eV) incident on the fission chamber is shown as a function of energy in Fig. 4. Cross-sections for 239 Pu [4] at low energies, along with results by other investigators, are plotted in Figs. 5-7.

The fission and capture cross-sections for 240 Pu [5] are shown in Figs. 8-10. Capture cross-sections were measured by recording the total gamma-ray production rate in a target. Since the neutron binding energy is known, a measurement of



FIG.18. Fission cross-section of ²⁴¹Am, 40 to 60 eV [10,24]



FIG. 19. Fission cross-section of ^{242}Am , 40 to 60 eV [10,24]. Crosses are original data, dots are ^{241}Am signals, and solid line is the cross-section of ^{242}Am .

the flux in a resonance where the target is black is adequate for calibrating the system.

The results of the fission measurements on 241 Pu, analyzed by the Idaho Falls group, [6] show that resonances are well resolved up to 45 eV. Data to which resonance parameters have been fitted are shown in Fig. 11, and Fig. 12 shows a comparison with other data.

New data on 233 U [7] are shown in Figs. 13 and 14. Relative capture-to-fission ratios (a) were measured by recording the total gamma-ray energy production rate in a target and combining this rate with the fission signal so as to fit a = 0.05 at 34 eV, a choice of α that seemed consistent with



FIG. 20. Fission cross-section of ²⁴²Am, 80 to 120 eV [10, 24], Crosses are original data, dots are ²⁴¹Am signals, and solid line is the cross-section of ²⁴²Am.



FIG.21. Fission cross-section of 242 Am, 10^4 to 10^6 eV [10,24]. Crosses are original data, dots are 241 Am signals, and solid line is the cross-section of 242 Am.

available data. Laboratory measurements to show what fraction of the gamma-ray signal was due to fission are required to extract more precise information about α . The fission cross-section for 233 U agrees with other measurements above 10 keV; near 100 eV these results are twice those measured elsewhere, but they are consistent with reported total crosssections. [17,18,19]

Results for 235 U [8] are available only for the lower energy region where fission of this nuclide was not used for measurement of neutron flux. In Figs. 15 and 16 results of this measurement are compared to other recent data.

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Measurements on two Am isotopes were made possible by the generous loan of target samples by the Radiochemistry Group at Lawrence Radiation Laboratory, Livermore, Calif. The cross-section for 2^{41} Am [10,24] is shown in Figs. 17 and 18. The isotope 242 is produced by neutron capture of 2^{41} Am followed by separation in a mass spectrometer to increase the 242/241 ratio. To get appreciably better data we would need a sample that had made a second pass through the mass separator. Since we do not anticipate getting a better sample for a long time, we are presenting these hitherto unpublished data [10,24] in Figs. 19-21. The errors are larger than we would like, but it can be seen that the cross-section for this odd-odd nuclide does not look markedly different from that of 239Pu above 10⁵ eV. These cross-sections agree well, in regions where they overlap, with recent measurements by Bowman. [25]

A feature shared by all of these data is the very low background as indicated by the deep valleys between resonances. Except in the valleys, these results are generally in good agreement with other measurements. Because multi-level fitting of resonance cross-sections is particularly sensitive to the shapes in the valleys where interference effects dominate, these data provide a rich source of resonance parameters; analysis by methods outlined by the Idaho Falls group [26] is in progress. The larger number of successive approximations which is required by these relatively precise data recommend the use of computer techniques that minimize the input-output time, and such work is also in progress. Another consequence of the low backgrounds found in these experiments is the possibility of producing extensive data on subthreshold fission, as is shown here for 240Pu (Figs. 8 and 9) and 241Am (Figs. 17 and 18).

A difficulty experienced in these measurements is the very low flux in the Pt resonances after the neutron beam has passed through many Pt foil backings. There are a few other holes in the flux due to resonances in residual oxygen in the pipe and various other absorbing materials in the beam. Efforts to make total and scattering cross-section measurements were not successful; these experiments came last in the neutron beam where there were additional holes in the flux due to 240Pu and 2330 samples. The 6Li-coated solid-state detectors used for scattering measurements had about equal response to scattered neutrons and to capture gamma rays. Further work on scattering must await the development of more suitable detectors.

Conclusion

We consider the fission cross-section results from the June 1965 experiment to be finished, but it has taken most of the 14 months since that experiment to process these data. The results for α need considerably more work. Our methods for reading film and handling the data have improved enough that a new set of data can be processed in a much shorter time. In any event, the effort required to produce such data using more conventional laboratory neutron sources would be many times greater than our own. Laboratory accelerators can produce, typically, 10^{21} neutrons per year, so it would take approximately 180 years of continuous running to match the

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time integrated flux from this one experiment using the same flight path. It is this combination of high flux and high resolution that makes this source uniquely suitable for measurements of fission cross-sections, where the target thickness must be small compared to the range of fission fragments. An experiment is now in progress to measure fission crosssections of 238Pu, 243Am, 244Cm, and the capture cross-section of 238Pu and 147Pm. The Pm is used in an exercise in handling very active nuclides--of the order of 10^4 curies--because measurements on such nuclides are prohibitively difficult with weaker sources. We are conducting another exercise in which we are attempting to measure the fission cross-section of 239Pu using a one microgram sample. If this can be done there are many exotic materials that can be prepared in only a small quantity that will be interesting targets.

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MEASUREMENT OF THE NEUTRON CAPTURE AND FISSION CROSS-SECTIONS AND OF THEIR RATIO ALPHA FOR ²³³U, ²³⁵U AND ²³⁹Pu*

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Abstract

MEASUREMENT OF THE NEUTRON CAPTURE AND FISSION CROSS-SECTIONS AND OF THEIR RATIO, ALPHA,FOR ²³⁵U, ²³⁵U AND ²³⁹Pu. A crucial requisite in the design of nuclear reactors, particularly breeder reactors, is the precise knowledge of the neutron capture and fission cross-sections of the fissionable materials involved. The need for accurate measurements was recently re-emphasized by the existence of a 20% discrepancy between the directly measured value of α_{RI} for ²³⁵U, the ratio of the capture resonance integral to the fission resonance integral, and the value of this parameter obtained by integrating differential measurements of the cross-sections as a function of energy.

A technique has been developed to measure simultaneously the neutron capture and fission crosssections of fissile nuclei. Since the two cross-sections are measured simultaneously, errors associated with uncertainties in the relative energy resolution and calibration of the two measurements are eliminated. Measurements of σ_c and σ_f for ²³⁵U in the neutron energy range of 3.25 to 25 eV have been published. These measurements have now been extended to cover the range of 1 to 100 eV, and the precision and energy resolution have been greatly improved. The fission cross-section is in good agreement with recent measurements using different techniques. At low energy, where the instrumental resolution is small compared to the Doppler broadening and where resonance scattering is unimportant, the directly measured capture cross-section is consistent over many of the resonances with that obtained indirectly by subtracting the fission and potential scattering from the total cross-section. The capture and fission resonance integrals and their ratio α_{RI} , obtained from our measurements on ²³⁵U and ²³³U over the neutron energy range of 1 eV to a few keV are now in progress. The limitations of the experimental method are discussed, and a detailed comparison of cross-section sobtained by different techniques are presented.

To compute the breeding ratio, the Doppler coefficient, and other parameters of large fast-power reactors, it is important to know α , the ratio of capture-to-fission, for the main fissile isotopes, and particularly for ²³⁹Pu, in the keV neutron energy region. Hopkins and Diven have performed direct measurements of α for ²³⁹U, ²³⁵U, and ²³⁹Pu with monoenergetic neutrons at 30, 60, and above 175 keV. But the value of α for ²³⁹Pu varies by more than a factor of two between 30 and 60 keV, and a detailed knowledge of the variation of this parameter with energy in the keV neutron energy range appears desirable. The application of the time-of-flight technique permits extending the direct measurements of α to energies where monoenergetic neutron sources are not readily available. Detailed measurements for α for ²³⁹Pu are now in progress, at few-keV intervals in the range of 100 to 600 keV. The technique has already been used with ²³⁵U and the results, now published, were found in agreement with those of Hopkins and Diven in the range of α obtained from direct measurements of α in a surface of Hopkins and Diven and with values of α obtained from direct measurements of η by Spivak et al. The factors limiting the precision of the measurements are discussed in some detail.

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1. INTRODUCTION

Simultaneous measurements of the capture cross-section, $\sigma_{\rm c}$, and the fission cross-section, $\sigma_{\rm c}$, have been performed for ²³³U over the neutron energy range from 0.4 to 100 eV and for ²³⁵U over the range from 0.4 eV to 3 keV. In addition, measurements of $\sigma_{\rm c}$ for ²³⁵U have been made up to 20 keV. In another set of measurements, values of $\alpha = \sigma/\sigma_{\rm c}$ have been measured directly over the range from 20 to 600 keV for ²³⁵U and ²³⁹Pu. This paper describes both types of experiments very briefly and compares the results with data from other laboratories.

TABLE I. EXPERIMENTAL PARAMETERS FOR THE ORNL-RPI SIMULTANEOUS MEASUREMENTS OF σ_c AND σ_f

	Low-Resolution Run	High-Resolution Run
Flight path, L	25.45 <u>+</u>	_ 0.05 m
Moderator	Polyeth	Nylene
Moderator thickness, d	2.54 <u>+</u>	0.5 cm
Sample composition	. U ₃ C) 8
Sample temperature, T	25 <u>+</u>	_ 5°C
Neutron burst width, $\tau_{\rm b}$	250 nsec	100 nsec
Repetition rate, R	240 pps	250 pps
Permanent filters	Cd, Co, Na	١٥ _B
Background filters	238 _U	Mn, ²³⁸ U;Co,Na;P
Channel width, τ (variable)	0.16-10.4 µsec	0.04-2.56 µвес

TABLE II. URANIUM SAMPLE PARAMETERS

	²³⁵ U Sample	²³³ U Sample		
Areal density, n	2.66 x 10 ⁻⁴ atoms/b	1.45 x 10 ⁵⁵ atoms/b		
Weight, wt	7.65 <u>+</u> 0.01 g	0.968 <u>+</u> 0.002 g		
Effective thickness, s	3.2 <u>+</u> 0.1 cm	3.2 ± 0.1 cm		
Isotopic composition				
233U		99•999 <u>+</u> 0•005%		
234 _U	0.040 <u>+</u> 0.01%			
235 _U	99.888 <u>+</u> 0.03%	•		
236 _U	0.006 <u>+</u> 0.006%			
238U	0.065 <u>+</u> 0.02%			

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FIG.1. Example of data from ORNL-RPI runs: Foreground (scintillator count rate not in coincidence with fission chamber) and background

2. SIMULTANEOUS MEASUREMENTS OF THE CAPTURE AND FISSION CROSS-SECTIONS

2.1. Experimental Technique

The LINAC at Rensselaer Polytechnic Institute [1] was used to produce a pulsed source of fast neutrons, some of which were slowed down by a moderator placed in the vicinity of the LINAC target. A detector consisting of a fission chamber surrounded by a large gamma-ray scintillator was positioned normal to the moderator surface at the end of a 25-m neutron flight path. The pulsed-neutron beam was collimated through the fission chamber; the scintillator detected the prompt gamma rays emitted following each neutron absorption in the uranium of the fission chamber, while the fission chamber detected only the fission. Thus, in principle, a fission was characterized by a pulse from the scintillator in coincidence with a pulse from the fission chamber, whereas a capture was characterized by a pulse from the scintillator alone. The energy of the neutrons absorbed in the uranium was measured by the time-of-flight technique, and the incident neutron beam spectrum was measured by a BF_3 ionization chamber placed between the moderator and the detector at a distance of 1 m from the detector.

The time-of-flight distributions of the signals from the scintillator in coincidence with the fission chamber, C(t), and not in coincidence with the fission chamber, A(t), were processed and stored in a computer. Simultaneously, the time-of-flight distribution of signals from the BF₃ detector was stored in another computer. A detailed description of the equipment has appeared elsewhere [2].

The measurements for 235 U were performed in two steps: the neutron energy range from 0.4 to 60 eV was covered by a "low-resolution" run,







FIG.3. Fission cross-sections for ²³⁵U from 20 to 50 eV

whereas the region above 15 eV was covered by a "high-resolution" run. The measurements for 233 U were performed with the low-resolution conditions only. Table I gives the experimental parameters for both types of runs, and Table II gives the uranium sample parameters.

During the low-resolution runs, filters of cadmium and cobalt were kept permanently in the beam so that the background level could be measured in the vicinity of 0.3 and 130 eV, where cadmium and cobalt, respectively,





FIG. 4. Fission cross-sections for ²³⁵U from 1 to 10 keV

have large resonances. The cadmium also eliminated "frame overlap" by absorbing all the neutrons below 0.3 eV.

Four types of measurements were performed: two with the fission chamber in place and two with a "dummy" chamber in place. In each case one of the two runs was made with the 238 U filter in the beam and one was made without the 238 U. The dummy chamber was identical to the fission chamber except that the uranium had been replaced by an equivalent amount of lead to mock up the scattering by the uranium.

Measurements with the 238 U filter were used to obtain a few additional background points at those energies where 238 U has large resonances. Measurements with the dummy were used to interpolate the background in between the 238 U resonance energies. Each individual measurement lasted about 3 hr, and the complete set of four measurements was repeated seven times to compensate for possible drifts in the electronics and in the beam spectrum, to obtain good statistics, and to permit some consistency tests on the data.

Figure 1 shows an example of the scintillator count rate not in coincidence with the fission chamber, A(t), and of the associated background, with the cadmium, cobalt, and ^{23B}U filters in the beam.

During the high-resolution runs, a ¹⁰B filter was kept permanently in the beam to eliminate overlap neutrons. The run consisted of six sets of measurements: measurements with the fission chamber and with the dummy, both with three different sets of filters in the beam [3].

2.3. Analysis of the Data

In the thin-sample approximation, the net count rates, C(t) and A(t), defined previously; are related to the cross-sections as a function of time of flight, t, by the following equations:

$$C(t) = \phi(t) \, \mathbb{N} \, \epsilon \, \epsilon_{f} \, \sigma_{f}(t) \quad , \qquad \qquad$$

(1)



FIG. 5. Capture cross-sections for ²³⁵U up to 15 eV

$$A(t) = \Phi(t) \ \mathbb{N}[\epsilon_{\sigma} \ \sigma_{\sigma}(t) + (1 - \epsilon) \ \epsilon_{\tau} \ \sigma_{\tau}(t)] , \qquad (2)$$

where $\phi(t)$ represents the neutron flux incident upon the fission chamber, N the number of uranium nuclei in the fission chamber, ϵ_c and ϵ_f the scintillator efficiencies for detecting capture and fission events, and ϵ the efficiency of the fission chamber for detecting fissions. ϵ can be obtained from the pulse-height spectra of the scintillator in coincidence and not in coincidence with the fission chamber, as has been shown elsewhere [2]. The two equations above can be solved for the cross-sections, giving

$$\sigma_{f}(t) = \frac{N_{1}}{\Phi(t)} C(t) , \qquad (3)$$

$$\sigma_{c}(t) = \frac{N_{2}}{\phi(t)} \left[A(t) - \frac{1 - \epsilon}{\epsilon} C(t)\right]$$
(4)

The constants N_1 and N_2 were obtained by normalizing the cross-sections over a low-energy interval where they are known from other experiments.

The major systematic errors on $\sigma_f \sqrt{E}$ for $\frac{235}{10}$ are estimated to be 1% associated with the normalization and 0.2 b $eV^{\overline{2}}$ associated with the background subtraction. On the values of $\sigma_c \sqrt{E}$, the major systematic errors are 3% for the normalization, 2 b $eV^{\overline{2}}$ for the background subtraction, and 0.04 $\sigma_f \sqrt{E}$ for the uncertainty in ϵ when solving Eq. 4. However, the magnitude of the systematic errors is very difficult to appreciate realistically

	· · · ·	Γ	Difference					
Energy				ORNL RPI	ORNL RPI: Bowman		ORNL RPI:Michaudon	
. (eV)	ORNL-RPI Bowman et al.		Michaudon et al.	' b	9k	b	%	
0.414 - 0.532	22.38 <u>+</u> 0.05	22.13	23.00	0.26	1.1	-0. 62	2.8	
0.532 - 0.683	16.55 <u>+</u> 0.05	16.27	17.14	0.28	1.7	-0.59	3.6	
0.683 - 0.876	13.90 <u>+</u> 0.05	13.75	14.28	0.145	1.1	-0.38	2.7	
0.876 - 1.13	18.10 <u>+</u> 0.08	17.98	18.05	0.12	0.7	0.05	0.3	
1.13 - 1.44	10.43 <u>+</u> 0.06	10.47	11.03	-0.04	0.4	-0.60	5.8	
1.44 - 1.86	3.74 <u>+</u> 0.06	3.70	3.90	0.04	1.1	- 0.16	4.3	
1.86 - 2.38	3.66 <u>+</u> 0.06	3.67	3.68	-0.01	0.3	-0.02	0.5	
2.38 - 3.06	3.38 <u>+</u> 0.06	3.26	3.29	0,12	3.7	0.09	2.7	
3.06 - 3.93	7 . 89 <u>+</u> 0.07	8.00	7.83	-0.11	1.4	0.06	0.8	
3.93 - 5.04	1.31 <u>+</u> 0.07	1,23	1.33	0,08	6.5	-0.02	1.5	
5.04 - 6.48	6.92 <u>+</u> 0.07	7.01	6.55	-0.09	1.3	0.37	5.6	
6.48 - 8.32	3.46 <u>+</u> 0.08	3.60	3.88	-0.14	4.0	-0.42	12.1	
8.32 - 10.7	25.54 <u>+</u> 0.10	26.55	24.86	-1.01	4.0	0.68	2.7	
10.7 - 13.7	11.62 <u>+</u> 0.09	11.86	11.11	-0,24	2.1	0.51	4.6	
13.7 - 17.6	8.17 <u>+</u> 0.09	8.38	8.15	-0.21	2.6	0.02	0.2	
17.6 - 22.6	16.56 <u>+</u> 0.09	17.32	15.19	-0.76	4.6	1.37	9.0	
22.6 - 29.0	10.92 <u>+</u> 0.10	11.35	10.17	-0.43	3.9	0.75	7.4	
29.0 - 37.3	13.50 <u>+</u> 0.10	14.09	12.01	-0.59	4.4	1.49	11.6	
37.3 - 47.9	8.45 <u>+</u> 0.11	8.95	8.02	-0.50	5.9	0.42	5.2	
47.9 - 61.4	13.67 <u>+</u> 0.15	14.17	12.56	-0.50	3.7	1.11	8.8	
0.414 - 61.4	220.0 <u>+</u> 1.5	224.0	216.0	-4.0	1.8	4.0	1.9	

TABLE III. COMPARISON OF FISSION RESONANCE INTEGRALS

and the only decisive test of the precision of the data must come from a careful comparison with results obtained independently, and when possible, by other techniques.

2.3. Results and Comparison with Other Data

2.3.1. Measurements on ²³⁵U

The ²³⁵U fission cross-section was normalized by making the fission resonance integral from 0.45 to 10 eV equal to 127.45 b, which is an average between the values of 127.08 b and 127.81 obtained from the data of Shore and Sailor [4] and Bowman et al. [5], respectively. Both measurements were normalized at 2200 m/sec. The capture cross-section was normalized by making the absorption resonance integral from 0.45 to 1.0 eV equal to 58.12 b, a value obtained by subtracting an estimated scattering contribution [6] from the total cross-section data of Shore and Sailor. The value is in good agreement with the value of 58.04 b reported by

	$\int \sigma_{\hat{\mathbf{f}}} d\mathbf{E} (\mathbf{b}-\mathbf{eV})$							
Energy Interval (eV)	ORNL-RPI	Brooks et al.	Bowman et al.	Brown et al.	Shore and Sailor	Michaudon et al.	Van Shi-Di et al.	
0.35 - 0.45		11.91	11.55		11.91			
0.45 - 0.50	4.36	4.34	4.32		4.37	4.47		
0.50 - 0.55	3.808	3.695	3.780		3.831	3•937		
0.55 - 0.70	9.63	9.39	9.47		9.42	9.98		
0.70 - 1.0	17.02	16.26	16.96		, 16.92	17.35		
1.0 - 1.3	20.85	20.85	20.78		21.09	21.14		
1.3 - 1.8	8.29	7.85	8.16		9.06	8.87		
1.8 - 4.5	46.53	42.66	43.28		48.15	46.51		
4.5 - 5.0	4.09	3•55	3.87		4.11	3.80		
5.0 - 10.0	281.7	257.0	292.1		272.4	275.0		
10.0 - 15.0	216.5	197.5	220.0			213.0		
15.0 - 20.5	315.8	278.85	332.6			287.3		
20.5 - 33.0	447 . 0	417.5	457•7	416		427.2		
33.0 - 41.0	496.3	415.2	517.5	465		445.3		
41.0 - 60.0	916.9	834.1	968.6	9 24		841.5		
60.0 - 73.0	308.1	245.7	302.6	318	,	.299•5		
73.0 - 100.0	665.8	580.5		682		632.0		
100.0 - 113.0	216.6	185.9		226		228.2		
113.0 - 200.0	1877.2	1600.8		1930		1809.3		
200.0 - 300.0	2081.8			2175		2052.5	1980	
300.0 - 1000.0	8124.8			8159		8122.4	7975	
1000.0 - 2000.0	7566			7656		7 5 45	7546	
2000.0 - 3000.0	5674			5461		5 7 61	5560	
3000.0 - 4000.0	5167			4678		4887	4880	
4000.0 - 5000.0	4656			4010		4502	¹ 4 ¹ 470	
5000.0 - 10000.0	17700			15920		18537	17460	

TABLE IV. COMPARISON OF FISSION INTEGRALS

Brooks et al. [7]. For the fission resonance integral over this interval, our data led to a value of 52.2 b, whereas Brooks et al. report a value of 52.1 b.

Figure 2 shows a comparison up to 15 eV of our fission cross-section data and those reported by Bowman et al. The solid line was obtained by
	$\int \sigma_{\mathbf{f}} \mathrm{d} \mathbf{E} /$	Έ(Ъ)	$\int \sigma_{\mathbf{c}} \mathrm{d}\mathbf{E}$	Έ (Ъ)	$\int \sigma_{\mathbf{a}} \mathrm{d}\mathbf{E}/$	Έ(Ъ)	Diff (in)	erence σ _a dE/E)
Energy Interval (eV)	ORNL-ÀRPI	Brooks et al.	ORNL-RPI	Brooks et al.	ORNL-RPI	Brooks et al.	%	barn
0.35 - 0.45		29•7		5.4		35.1		
0.45 - 0.5	9.21	10.4	1.13	1.29	10.34	11.69	13.1	-1.35
0.5 - 0.55	7•3	7.0	0.87	0.85	8.17	7.85	4.1	0.32
0.55 - 0.7	15.5	15.2	1.7	1.6	17.2	16.8	2.4	0.4
0.7 - 1.0	20,2	19.5	2.2	2.2	22.4	21.7	3.2	0.7
1.0 - 1.3	18.6	18.6	4.9	5.6	23.5	24.2	3.0	-0.7
1.3 - 1.8	5•5	5.2	1.4	1.8	6 .9	7.0	1.4	-0.1
1.8 - 4.5	15.8	14.7	8.5	9•3	24.3	24.0	1.2	0.3
4.5 - 5.0	0.85	0.73	4.6	4.6	5.45	5.33	2.3	0.12
5.0 - 10.0	34.5	31.7	24.4	27.9	58.9	59.6	1.2	-0.7
10.0 - 15.0	17.2	15.6	19.5	19.0	36.7	34.6	6.1	2.1
15.0 - 20.5	17.1	15.2	10.7	12.0	27.8	27.2	2.2	0.6
20.5 - 33.0	17.5	16.3	12.0	13.9	29.5	30.2	2.4	-0.7
33.0 - 41.0	13.8	11.6	7•9	8.4	21.7	20.0	8.5	1.7
41.0 - 60.0	17.9	16.3	8.9	9•9	26.8	26.2	´ 2 . 3	0.6
60.0 \$73.0	4.5	3•7	1.7	1.8	6.2	5.5	12.7	0.7
73.0 - 100.0	7.8	6.8	4.2	5.1	12.0	11.9	0.8	0.1
100.0 - 113.0	2.1	1.7	1.8	3.0	3.9	4.7	20.5	-0.8
113.0 - 200.0	12.5	10.8	6.4	7.8	18.9	18.6	1.6	0.3
0.45 - 200.0	237.86	221.03	122.8	136.04	360.66	357.07	1.0	3.59

TABLE V. COMPARISON OF RESONANCE INTEGRALS

joining our data at each time-of-flight channel by a straight segment. There are five data points near 13 eV in the data of Bowman et al. that are probably too high due to an instrumental pile-up effect.

In Fig. 3 our fission cross-section is compared in the range from 20 to 50 eV to that obtained by Brown et al. [8] with neutrons from an underground nuclear explosion. In the "valleys" between resonances, the crosssection of Brown et al. is considerably lower than ours and lower than that of Bowman et al. (not shown on the figure). In Fig. 4 the same two sets of data are compared in the range from 1 to 10 keV.

In Fig. 5 our capture cross-section up to 15 eV is compared to that of Brooks et al. The solid line was obtained in the same manner as that shown in Fig. 2.

TABLE VI. COMPARISON OF CAPTURE INTEGRALS

	<u></u>	$\int \sigma_{c} dE (b \cdot e)$	V)
Energy Interval (eV)	ORNL-RPI	Van Shi-Di et al.	Uttley
100 - 200	1171	1910	1 31 5
200 - 300	924	1210	1215
300 - 400	670	610	443
400 - 500	514	510	429
500 ~ 600	515	360	436
600 - 700	491	486	330
500 - 800	517	420	546
800 - 990	կկկ	410	288
900 - 1000	545	328	461
100 ~ 1000	5791	6244	5463
1000 - 1100	529	142	600
1100 - 1200	403	290	355
1200 - 1300	344	152	278
1300 - 1400	307	245	376
1400 - 1500	332	292	162
1500 - 1600	311	362	282
1600 - 1700	261	326	283
1700 - 1800	247	325	189
1800 - 1900	275	340	154
1900 - 2000	321	369	429
1000 - 2000	3330	2843	3108
2000 - 3000	1877	2200	2010
100 - 3000	10998	11287	10581

The fission resonance integrals over quarter lethargy intervals were computed using our data, the data of Bowman et al., and the data of Michaudon et al. [9]. The comparison of these integrals is given in Table III. The resonance integrals based on our data agree within 3% with those based on the data of Bowman et al. and within 6% with those based on the data of Michaudon et al. The discrepancies are clearly systematic, yet over the range from 0.4 to 61.4 eV the resonance integrals based on our data are within 2% of those based on the two other sets of data. The discrepancies between our resonance integrals and those of Bowman et al. are about four times as large as our estimated systematic errors.

TABLE VII. COMPARISON OF RESONANCE INTEGRALS. ABOVE 0.5 eV

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A) Values computed from cross sections

$$\int_{0}^{10} \frac{\text{keV}}{\text{g}} \sigma_{g} \frac{\text{dE}}{\text{E}} = 267 \pm 3 \text{ b} \qquad \text{ORNL-RFI (1$ uncertainty from normalization)} \\ \int_{10}^{1} \frac{\text{MeV}}{\text{g}} \sigma_{g} \frac{\text{dE}}{\text{E}} = 8 \pm 2.5 \text{ b} \qquad \text{Perkin et al., White, Diven} \\ (\text{estimated uncertainty}) \\ \int_{10}^{3} \frac{\text{keV}}{\text{g}} \sigma_{g} \frac{\text{dE}}{\text{E}} = 136 \pm 5 \text{ b} \qquad \text{ORNL-RFI (3$ uncertainty in normalization)} \\ 0.5 \text{ eV} \qquad \sigma_{g} \frac{\text{dE}}{\text{E}} = 136 \pm 5 \text{ b} \qquad \text{ORNL-RFI (3$ uncertainty in normalization)} \\ \int_{10}^{3} \frac{\text{keV}}{\text{g}} \sigma_{g} \frac{\text{dE}}{\text{E}} = 3.0 \pm 0.5 \text{ b} \qquad \text{Van Shi-Di et al., Uttley (estimated uncertainty)} \\ \int_{30}^{3} \frac{\text{keV}}{\text{keV}} \sigma_{g} \frac{\text{dE}}{\text{E}} = 1.65 \pm .05 \text{ b} \qquad \text{ORNL values of } \alpha, \text{ and Diven, Perkin et al.} \\ \text{Thus} \quad I_{\mathbf{F}} = 275 \pm 4 \text{ b} \qquad I_{\mathbf{c}} = 140.5 \pm 5 \text{ b} \qquad \alpha_{\mathbf{RI}} = 0.51 \pm 0.02 \\ \text{B}) \quad \text{Values obtained by direct measurements}; \\ \text{a}) \quad \text{Evaluation of Feiner and Esh:} \\ I_{\mathbf{F}} = 280 \pm 10\text{ b} \qquad I_{\mathbf{c}} = 140 \pm 8\text{ b} \qquad \alpha_{\mathbf{RI}} = 0.50 \pm 0.02 \\ \text{b}) \quad \text{Redman and Bretscher:} \qquad \alpha_{\mathbf{RI}} = 0.501 \pm 0.022 \\ \end{array}$$

In Table IV the fission integrals over various energy intervals, obtained from seven different measurements, are compared. The results are all within a few percent of each other except those obtained from the data of Brooks et al. which above 2 eV are systematically lower than all the others, with the discrepancy increasing with energy to about 12% in the vicinity of 100 eV.

Fission, capture, and absorption resonance integrals obtained from our data and from the data of Brooks et al. are compared in Table V. In spite of large systematic discrepancies in the fission and capture resonance integrals that increase with energy, the discrepancies in the absorption resonance integrals are small and not systematic.

In Table VI we compare our values of the capture cross-section integrals over various energy intervals with values obtained by a somewhat similar



FIG. 6. Fission cross-sections for ²³³U from 1 to 30 eV

technique by Van/Shi-Di et al. [10] and with values computed by Uttley [11] using his total cross-section measurement, the fission cross-section measured by Michaudon et al., and a value of 12.3 b for the potential scattering. The large discrepancies over intervals of 100 eV are probably due, in part, to the different energy resolutions and energy scales of the various measurements. For the integral from 100 eV to 3 keV our value is within 5% of the other two values.

Finally, in Table VII we compare the fission and capture resonance . integrals above 0.5 eV and their ratio $\alpha_{\rm RI}$, as obtained from our data below a few keV and other data [12-15] at higher energies, with direct measurements of these quantities [16-17].

2.3.2. Measurements on ²³³U

The ²³³U fission cross-section was normalized to a value of 420.47 b·eV for the integral of the fission cross-section from 1.0 to 2.1 eV, a value obtained from the data of Moore et al. [18]. Over this energy interval, the scattering cross-section was taken to be 12.4 b [18,19] and the capture cross-section was normalized by making the total cross-section integral $(\sigma_{\rm f} = \sigma_{\rm f} + \sigma_{\rm c} + \sigma_{\rm s})$ equal to 506.82 b·eV, a value obtained from the data of Pattenden and Harvey [20]. In the energy interval from 2.1 to 2.75 eV, the total cross-section integral obtained from our fission and capture cross-section measurements and from a scattering cross-section of 12.4 b is 210.68 b·eV, the same value as that obtained by integrating the data of Pattenden and Harvey.

In Fig. 6 our fission cross-section is compared to that measured by Nifenecker et al. [21] for energies below 30 eV. At low energies the two sets of data are in agreement but, as the energy increases, our cross-section is systematically larger. At 30 eV the discrepancy is about 10%.

In Fig. 7 the total cross-section obtained by Pattenden and Harvey is compared to that obtained by adding our fission and capture cross-sections and a scattering cross-section of 12.4 b.



FIG. 7. Total cross-sections for ²³³U from 1 to 30 eV

Our ²³³U measurements are still being checked for any possible systematic errors.

3. DIRECT MEASUREMENT OF α , THE RATIO OF CAPTURE TO FISSION

3.1. Experimental Technique

For the direct measurement of α , a pulsed-neutron beam was collimated onto a sample of the isotope under investigation (²³⁵U or ²³⁹Pu), which was placed in the center of a large hydrogenous gamma-ray scintillator "poisoned" with gadolinium. A capture event in the sample was characterized by a single pulse of the scintillator due to the cascade of capture-gamma rays, while a fission event was characterized by a pulse due to the promptfission gamma rays followed a few microseconds later by additional pulses due to the gamma rays produced when the thermalized fission neutrons were captured in the gadolinium of the scintillator.

The pulses of neutrons were produced by the 7 Li(p,n) reaction: 2-nsecwide bursts of protons were accelerated on a thick lithium target by the 3-MV ORNL Van de Graaff. Below 100 keV, neutron energies were measured by the time-of-flight technique using a 1-m flight path and a time resolution of 6 nsec. Above 100 keV the neutron energies were obtained from the energy of the protons impinging upon the lithium target, and the energy resolution was determined by the thickness of the target.

The two chief sources of error in the measurement were a 5 to 8% error on α due to the uncertainty in the relative efficiencies of the scintillator for detecting capture and fission events and a 0.007 absolute error on α associated with the uncertainty in the probability of detecting fission neutrons after a fission event.

	c	(α
E (keV)	^{эзө} Ри	^{азь} U	E (keV)	239 Pu	835 U
17.7	0.395 <u>+</u> 0.108	0.325 <u>+</u> 0.049	38.5	0.253 <u>+</u> 0.017	0.340 <u>+</u> 0.016
18.3	0.490 <u>+</u> 0.109	(0.274 <u>+</u> 0.044	40.5	0.226 <u>+</u> 0.016	0.360 <u>+</u> 0.016
18.8	0.443 <u>+</u> 0.097	0.337 <u>+</u> 0.043	42.3	0.246 <u>+</u> 0.016	0.365 <u>+</u> 0.016
19.4	0.442 <u>+</u> 0.089	0.342 <u>+</u> 0.041	44.5	0.244 ± 0.017	0.365 <u>+</u> 0.015
20.2 <u>+</u> 0.6	0.350 <u>+</u> 0.075	0.445 <u>+</u> 0.043	46.7	0.286 <u>+</u> 0.017	0.361 <u>+</u> 0.015
21.0	0.353 <u>+</u> 0.071	0.406 <u>+</u> 0.038	48.5	0.199 <u>+</u> 0.027	0.335 <u>+</u> 0.015
21.7	0.406 <u>+</u> 0.071	0.340 <u>+</u> 0.031	51.0 <u>+</u> 2 .5	0.198 + 0.028	0.339 <u>+</u> 0.016
22.4	0.409 <u>+</u> 0.048	0.360 <u>+</u> 0.031	54.5	0 . 195 <u>+</u> 0.030	0.322 <u>+</u> 0.013
23.1	0.371 <u>+</u> 0.040	0.391 <u>+</u> 0.031	57.5	0.178 <u>+</u> 0.032	0.329 <u>+</u> 0.012
23.9	0.353 <u>+</u> 0.036	0.373 <u>+</u> 0.027	60.7	0.176 <u>+</u> 0.025	0.300 <u>+</u> 0.011
24.8	0.350 <u>+</u> 0.034	0.382 <u>+</u> 0.025	64.0	0.174 + 0.022	0.355 <u>+</u> 0.012
25.7	0.355 <u>+</u> 0.030	0.368 <u>+</u> 0.024	68.0	0.169 <u>+</u> 0.021	0.315 <u>+</u> 0.012
26.8	0.327 <u>+</u> 0.027	0.372 <u>+</u> 0.022	72.0	0.165 <u>+</u> 0.020	0.329 <u>+</u> 0.013
27.9	0.289 <u>+</u> 0.025	0.349 ± 0.020	77.0	0.160 <u>+</u> 0.021	0.350 <u>+</u> 0.016
29.0	0.281 <u>+</u> 0.023	0.372 <u>+</u> 0.020	82.0 <u>+</u> 5.0	0.172 + 0.034	0.349 <u>+</u> 0.022
30.1 <u>+</u> 1.2	0.329 <u>+</u> 0.033*	0.384 <u>+</u> 0.033*			
31.0	0.297 <u>+</u> 0.020	0.370 <u>+</u> 0.018	200.0 ± 7.0	0.127 <u>+</u> 0.008	0.254 <u>+</u> 0.010
32.3	0 <i>1</i> 303 <u>+</u> 0.019	0.367 <u>+</u> 0.018	300.0 <u>+</u> 6.0	0.116 <u>+</u> 0.011	0.215 <u>+</u> 0.010
33.8	0.288 <u>+</u> 0.019	0.376 <u>+</u> 0.018	400.0 <u>+</u> 6.0	0.078 <u>+</u> 0.011	0.164 <u>+</u> 0.008
35•3	0.299 <u>+</u> 0.019	0.37 ⁴ ± 0.017	500.0 <u>+</u> 6.0	0.065 <u>+</u> 0.005	0.154 <u>+</u> 0.005
37.0	0.228 <u>+</u> 0.017	0.374 <u>+</u> 0.017	600.0 <u>+</u> 5.0	0.035 <u>+</u> 0.005	0.129 <u>+</u> 0.004

TABLE VIII. α of ²³⁹Pu and ²³⁵U

*The total error on α is given at 30.1 keV; values at other energies were normalized to this value, and only the statistical error is given for the other energies.

A detailed discussion of this technique and some results obtained on $^{\rm 235U}$ at ORNL have been published [22].

3.2. Results and Comparison with Other Data

The values of α obtained for ²³⁵U and ²³⁹Pu are given in Table VIII. In Fig. 8 the ²³⁵U data are compared to data obtained at ORNL by the same technique but with a poorer energy resolution [22], as well as with data obtained by very similar techniques [10,15], obtained by integral measurements [23], or derived from measurements of η [24]. Figure 9 shows similar comparisons for ²³⁹Pu. Figures 8 - 9 show that in the regions where they overlap the various sets of measurements are consistent within their rather large uncertainties.





FIG.8. Comparison of various values of α of ²³⁵U in the range from 1 to 1000 keV



FIG. 9. Comparison of various values of α of ²³⁹Pu in the range from 10 to 1000 keV

4. CONCLUSIONS

Reactor designers have requested the fission cross-section of 235 U in the range from thermal to 100 keV to 2% accuracy and the capture crosssection in the range up to 30 keV to an accuracy of 3% [25]. It is clear from an inspection of Tables III and IV that over small energy intervals the discrepancies between various recent measurements of of for 235 U are at least of the order of 5%. The discrepancies between values of $\sigma_{\rm C}$ are even larger. On the other hand, if the cross-sections are averaged over large energy intervals, the agreement between different measurements becomes comparable with the accuracy required: the fission resonance integral from 0.4 to 61 eV computed from our data is within 2% of the values obtained from the data of Bowman et al. [5] and Michaudon et al. [9]. The integral of the fission cross-section from 100 eV to 10 keV obtained with our data is 0.7% lower than that obtained from the data of Michaudon et al. and 2.2% lower than that obtained from Van Shi-Di's data [10]. In the range from 100 eV to 3 keV, our integrated capture cross-section is 2.6% lower than that derived by Uttley [11] and 3.9% higher that that measured by Van Shi-Di. Finally, the computed values of the fission and capture resonance integrals above 0.5 eV and of their ratio $\alpha_{\rm RI}$ are in very good agreement with the measured values of those quantities.

For 233 U an extended comparison of our results with other data has not yet been performed.

Finally, the various direct measurements of α in the keV range are reasonably consistent, but the uncertainties associated with the various sets of data are rather large (~10%). Furthermore, three direct measurements of α [10,15,22] were made with a technique very similar to that used in our measurements; thus, systematic errors associated with that technique may be present in all the data.

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A SINGLE-LEVEL ANALYSIS OF $^{235}\!U$ BASED ON RECENT σ_t , σ_f AND σ_c MEASUREMENTS*

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Abstract

A SINGLE-LEVEL ANALYSIS OF ²³⁵U BASED ON RECENT σ_f , σ_f , $AND \sigma_c$ MEASUREMENTS. Total, capture, and fission cross-section data have been simultaneously fit with a single-level Breit-Wigner least-squares analysis. The resonance capture and fission integrals calculated from the parameters agree within 3% with corresponding direct numerical integration of the data. The ratio of the capture resonance integral to the fission resonance integral agrees within 2% with reactor integral measurements. The effects of simultaneously fitting two cross-sections at a time: total and fission, and capture and fission have been studied.

I. INTRODUCTION

There has been a persistent discrepancy between the ratio, $\overline{\alpha}$, of the measured capture resonance integral to the fission resonance integral and the ratio, $\overline{\alpha}$, calculated from microscopic cross-section measurements. The integral measurements have given a value 15 to 20% lower(1) than that calculated from differential measurements. Feiner(2) recommends the follow-ing values for the resonance integrals: If = 280 + 11; I_c = 140 + 8; $\overline{\alpha}$ = .50 + .02. These values were obtained by averaging various integral measurements(3); they correspond to a 0.5 ev cutoff, and corrections to a 1/E spectrum. Brooks(4) points out that when the uncertainties of the measurements are considered perhaps there is no discrepancy. He gives for the Harwell data, which are based on the measurement of η (5, 6), $\overline{\alpha}$ = 0.57 + 0.06 (cutoff of 0.45), and for the integral measurements he cites a value of $\overline{\alpha}$ = 0.526 + 0.027.

Recently simultaneous measurements of the fission and the capture crosssections for ²³⁵U have been made by G. deSaussure⁽⁷⁾, et al. (ORNL-RPI) over the energy interval from 0.4 ev to 3 kev, in addition the fission crosssection was measured up to 20 kev, α has also been measured directly over the energy range from 20 kev to 600 kev. The value of $\overline{\alpha}$ calculated from these measurements, supplemented at the higher energies by other recent measurements, is in agreement with the experimental integral values. G. deSaussure, et al. have obtained the following values: If = 275 + 4 barns and I_c = 140.5 + 5 barns out to 1 Mev, and the ratio $\overline{\alpha}$ = 0.5I + 0.02. A numerical integration using available fission cross-section data⁽⁸⁾ gives a fission resonance integral value of 3 barns over the energy range 1 Mev - 10 Mev.

^{*} Work performed under the auspices of the Space Nuclear Propulsion Office. Prime Contractor Aerojet-General Corporation.

Using this fission data and extrapolating available measurements $^{(9)}$ of α one may conclude that the capture resonance integral is negligible over the energy range 1 Mev – 10 Mev.

At present the resonance parameters are being obtained from a simultaneous least-square fit (triple fit) to the ORNL-RPI capture and fission cross-sections and the Saclay⁽¹⁰⁾ total cross-section. In this paper the results are presented for the energy range out to 63 ev. In addition, some comparisons are made with the two sets of parameters obtained by simultaneous least-square fits to the Saclay fission and total cross-section measurements, as well as simultaneous fits to the ORNL-RPI fission and capture cross-sections. The resonance integrals which are calculated out to 37 ev using these different sets of resonance parameters are presented and compared with the resonance integrals obtained from a numerical integration using the ORNL-RPI data.

II. DISCUSSION OF CALCULATIONS

A. Theoretical Cross Sections

The single-level analysis embodied in the EXT⁽¹¹⁾ computer program has been used. EXT treats s-wave neutrons only; it includes interference between potential and resonance scattering, but not interference between levels, i.e. the fission and capture cross-sections for some incident neutron energy are obtained by summing the contributions from individual levels. The usual simplifying assumption that the neutron velocity is much greater than the velocity of the absorber nucleus has not been made. A Maxwellian velocity distribution is assumed for the target nucleii. The treatment of Doppler broadening and experimental resolution is the same as in a previous analysis (ref. 12).

B. Least-Square Calculation

The expressions for the fission, capture, and scattering cross-sections for some incident neutron energy, E, depend on the zero-temperature resonance parameters, the g-factor, the temperature of the sample during the measurement, and the experimental resolution.

As an example of the least-square calculation for obtaining the resonance parameters, consider fitting only the fission cross-section. Let $\sigma_{f,T}^e$ be the experimental data, and $\sigma_{f,Te}^{fn}$ the theoretical cross-section, where T^e is the "effective temperature" of the sample which is the true temperature, T, of the sample modified to account for binding effects and experimental resolution. Assume that a corrected fission cross-section is obtained from

$$\sigma_{f,Te} = \sigma_{f,Te} + \sum_{i} \left(\frac{\partial \sigma_{f,Te}}{\partial f_{i}} \right)_{o} S_{f_{i}}^{f_{i}}$$

where the subscript o indicates the term has been calculated using initial-guess parameters, and F; denotes the ith resonance parameter. By definition

$$S \equiv \sum_{\sigma_{f}, T} \left[\sigma_{f}, T_{e}^{e}(E_{j}) \right]^{2} w_{j}^{f} \qquad \text{where } w_{j}^{f} \text{ is a weight factor,}$$

and the Ei's are the incident neutron energies for which the cross-section has

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been measured.

$$S = \sum_{j} \left[\sigma_{\overline{F}, \overline{T}}^{e}(E_{j}) - \sigma_{\overline{F}, \overline{T}e}^{th}(E_{j}) \right]_{0} - \sum_{i=1}^{T} \left(\frac{\partial \sigma_{\overline{F}, \overline{T}e}(E_{j})}{\partial f_{i}} \right)_{0} S f_{i}^{t} \right]^{2} w_{j}^{t}$$

Minimizing this expression with respect to $S \xi_k$:

$$\frac{\partial S}{\partial (S\xi_{h})} = 0 = \sum_{j} \Delta w_{j}^{f} \left[\sigma_{f,Te}(E_{j}) - \sigma_{f,Te}(E_{j}) \right]_{o}^{-\sum_{i=1}^{I}} \left(\frac{\partial \sigma_{f,Te}(E_{j})}{\partial \xi_{i}} \right)_{o} SE_{i}^{-1}$$

There are 1 (k = 1 to 1) such equations which may be solved for the 1 unknown resonance parameters.

The simultaneous linear equations are solved using the Gauss elimination method. Once the ξ_k 's have been found the "corrected parameters" are taken as $\xi_i / + r \xi_i$

where r is a relaxation factor. Using these parameters another iteration is carried out, etc. On each iteration the percent change in each resonance parameter and also the root mean square value of the deviation between the experimental and theoretical cross-section is calculated, i.e.



where \boldsymbol{v} is the index for the iteration, and

where J is the number of energy points.

These quantities are a measure of the convergence of the theoretical crosssection to the experimental values.

If we wish to fit other cross-sections simultaneously with the fission, then the function S can be defined as

$$S \equiv \sum_{j} \left[\sigma_{f,T_{i}}^{e}(E_{j}) - \sigma_{f,T_{i}}^{e}(E_{j}) \right]^{2} w_{j}^{f} + \sum_{i} \left[\sigma_{\ell,T_{2}}^{e}(E_{i}) - \sigma_{\ell,T_{2}}^{e}(E_{i}) \right]^{2} w_{\ell}^{t} + \cdots$$

and the S_{k} 's are obtained as described above.

For the triple fit the Saclay data above 27 ev was shifted to higher energies by 0.026 ev to align the peaks more closely with the ORNL data. Either set of data could have been shifted. The Saclay data, of course, was not taken for the same neutron energies as the ORNL data; to obtain the Saclay cross-section for an arbitrary neutron energy a linear interpolation was carried out between the adjacent data points. The parameters of about seven resonances are iterated on simultaneously depending upon the number of data points. However, all resonances contribute to the cross-section at a given energy.

C. Test Cases

The least-square fitting program has been tested by using theoretical cross-sections (calculated using the EXT program) for which the resonance parameters are known in place of experimental cross-sections. Approximate parameters are used as initial theoretical guesses and the calculated parameters are checked on successive iterates to see how well they converge to the true parameters.

All of the parameters cannot be determined uniquely, of course, by fitting just one cross section, e.g. the fission cross-section. In testing such a case a set of parameters was obtained that gave a fission cross-section which fit the "experimental" cross-section very well yet the parameters were completely in error.

However, when two cross-sections are fit simultaneously, e.g. the fission and capture cross-sections or the fission and total cross-sections, the calculated parameters are unique and agree with the true parameters. As an example, the fission and total cross-sections were fit for the following two artificial resonances:

True	Е. 8.80	Γ <u>.</u> 1.18 × 10 ⁻³	Г <u>р</u> . 050	, 080	E. 9. 20	Γn 1.3 × 10-3	Г г . 100	ፍ . 035
lst Guess	8.72	1.50 x 10 ⁻³	. 030	. 110	9.28	1.0 x 10 ⁻³	. 080	. 046

The above parameters have units of ev. After three iterations the calculated parameters had converged till they differed from the true parameters by less than 0.1%. Similar results were obtained when fitting σ_c and σ_f , and also when doing triple fits to σ_c , σ_f , and σ_T .

To indicate how fitting data for two cross-sections defines the complete set of parameters uniquely, consider the special case of zero temperature. The E_o is determined, of course, by the energy at the peak of the resonance; the width of the resonance yields the value of $\Gamma = \Gamma_f + \Gamma_c + \Gamma_n \cong \Gamma_f + \Gamma_c$ since in general $\Gamma_n <<\Gamma_f$, Γ_c for U²³⁵. Also if the fission and the capture cross-sections are being fit, the peaks of these curves yield the values of

$$\frac{L_{s}}{r^{d}L^{u}L^{t}} = A \quad \text{and} \quad \frac{r^{d}}{r^{d}L^{u}L^{d}} = B.$$

Hence three equations and three unknowns.

The program was also checked to assure that it yields correct converged results when doing a triple fit with the sample at a different temperature for measuring the total cross-section than for measuring the fission and capture crosssections.

When fitting actual data it was found in several tests that the same converged parameters were obtained when using unit weighting or using a weighting factor proportional to the cross-section.

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III. COMPARISON OF THE THREE SETS OF PARAMETERS FOR NEUTRON ENERGIES LESS THAN 37 ev*

There are many resonances from 1.86 ev to 37 ev for which the single level analysis provides a good fit to the data. Figures 1, 2, and 3 are samples of the least-squares simultaneous fits to: (1) the ORNL-RPI fission and capture cross-section data and the Saclay total cross-section data, (2) the ORNL-RPI fission and capture cross-section data, and (3) the Saclay fission and the Saclay total cross-section data respectively. Unlike the idealized test cases described in Section 11 the three sets of parameters that have been obtained from fitting the actual experimental data vary considerably. In practice one might expect, and it is evident in Figure 2, that the largest deviation between the experimental data and the cross-sections calculated from the parameters which were obtained from fitting the ORNL-RPI capture and fission data occurs for the total cross-section. Similarly for the other double fit to the Saclay fission and the Saclay total cross-section (Figure 3) the largest deviation occurs for the capture cross-section. In general the triple fit shown in Figure 1 gives the best average fit to all of the data; for example frequently the theoretical total cross-section will be less than the experimental crosssection at a resonance peak whereas the theoretical capture and fission crosssections at the peak will be greater than the data, etc. The Γ_{n} 's obtained from the triple fit tend to agree with the Γ_n 's obtained from the double fit to the fission and total cross-sections, whereas the ratio Γ_p/Γ_f for each resonance tends to agree with the ratio obtained from the double fit to the capture and fission cross-sections.

Numerically integrating over the range 1.86 ev - 37.3 ev, and using the preliminary ORNL-RPI data one obtains for the resonance integrals $\Delta I_{f} = 114.4$ barns and $\Delta I_{c} = 93.0$ barns; using these partial integrals together with final normalized data over the remaining energy interval gives $\overline{\alpha} = 0.53$ over the range 0.5 ev - 10 Mev. (Using the final normalized ORNL-RPI data over the energy range 1.86 ev - 37.3 ev as well as the remaining energy interval results in $\Delta I_{f} = 112.6$ barns and $\Delta I_{c} = 86.1$ barns, also $\overline{\alpha} = 0.505$ over the energy range 0.5 ev - 10 Mev, - see Section IV.) Calculating the resonance integral from the resonance parameters obtained from the double fitting to the ORNL-RPI σ_{c} and σ_{f} gives $\Delta I_{f} = 118$ barns, $\Delta I_{c} = 94.7$ barns, and $\overline{\alpha} = 0.53$. From the parameters obtained from the double fit to the Saclay σ_{f} and the Saclay σ_{T} one obtains $\Delta I_{f} = 103$ barns, $\Delta I_{c} = 92.3$ barns, and $\overline{\alpha} = 0.53$ in good agreement with the numerical integration of the ORNL data.

Summarizing, the triple fit in general gives the best set of parameters. Fitting the new ORNL-RPI capture and fission cross-section data simultaneously with the Saclay total cross-section data does not greatly affect the values of Γ_n that are obtained from fitting the Saclay data alone, but it does apportion

^{*} Preliminary ORNL-RPI capture cross-section data was used for the calculations described in this section that were $\sim 8\%$ higher than the final normalized data of reference 7, the fission cross-section data was $\sim 2\%$ higher.



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FIG.1. The theoretical curves were calculated using the parameters obtained from a simultaneous least-square fit to the ORNL-RPI capture and fission cross-section data $(T = 300^{\circ}K)$ and the Saclay total cross-section data $(T = 77^{\circ}K)$.





FIG.2. The theoretical curves were calculated using the parameters obtained from a simultaneous least-square fit to the ORNL-RPI capture and fission cross-section data $(T = 300^{\circ}K)$.

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FIG.3. The theoretical curves were calculated using the parameters obtained from a simultaneous least-square fit to the Saclay total (77° K) and fission (300°K) cross-section data.

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TABLE I. TRIPLE-FIT RESONANCE PARAMETERS

	Eo	2g [n	Γ _f	Г _с	Type of Fit		E _o	2g Γ _n	۲ _۴	Ľ	Туре	
	(ev)	(mv)	(mv)	(mv)	0		(ev)	(mv)	(mv)	(mv)	of Fit	
1)	0.290	.0032	100	35	a	51)	28.371	.292	207	81.7	đ	
2)	1.135	.0154	42	115	a	52)	28,756	.030	72.7	23.1	d	
3)	2.04	.0077	10	37	a	53)	29.668	. 190	39.5	52.4	c	
4)	2.806	6010.	686	118	d	54)	30,627	. 259	131	67.2	ь	
5)	3.146	.0186	92.0	39.8	4	55)	30.891	.494	20.0	36.7	ь	
6)	3.608	.0458	54.0	38.2	ь	56)	32,088	1.871	65.9	46.1	ь	
7)	4.844	.0591	3.86	39.6	c	57)	33.540	1.824	32.8	44.0	ь	
8)	5.435	.00847	134	86,6	f	58)	34.379	2,177	47.9	47.8	ь	
9)	5.732	.0279	443	49.0	e	59)	34.866	1.246	116	40.8	ь	
10)	6.106	.00854	207	9.23	e	60)	35.205	4.547	105	51.9	Ь	
11)	6.203	.0323	79.8	26.7	di	61)	38.288	.562	471	83.6	F	
12)	6.381	.278	12.2	42.4	ь	62)	39.420	2.456	61.4	41.3	ь	
13)	7.076	.127	31.9	40.5	ь	63)	39.911	.311	153	22.3	f	
14)	8.777	1.128	85.0	39.3	ь	64)	40.56 2	.413	217	53.8	c	
15)	8.993	.0810	178	40.7	ь	65)	41.398	.773	399	77.0	c	
16)	9.287	.153	71.9	39.3	ь	66)	41.611	.089	0.9	0.8	d	
17)	9.665	.0936	500	12.2	ь	67)	41.887	1.229	22.3	49.7	c	
18)	10,180	.0632	60.1	43.1	ь	68)	42.240	.489	131	78.0	c	
19)	10.721	.0488	733	0.49	e	69)	42.723	.331	29.9	71.8	с	
20)	11,660	.622	7.4	40	ь	70)	43.407	.677	35.4	60.2	c	
21)	12,395	1.275	28.8	36.6	ь	71)	43.962	.682	129	63.7	с	
22)	12,860	.0513	60.4	52.2	ь	72}	44.619	.792	117	54.6	ь	
23)	13,270	.0300	76.7	43.3	ь	73)	44.948	.975	552	87.9	ç	
24)	13.695	.0097	4.4	0.7	c	74)	45.800	.196	92.4	59.0	ь	
25)	13.986	.650	495	84.1	ь	75)	46.808	.646	105	41.5	ь	
26)	14.541	.0994	17.7	35.1	9	76)	47.020	1.030	121	58.5	Ь	
27)	15,407	.235	44.5	38.2	ь	77)	47.980	.759	45.3	49.8	ь	
28)	16,089	.363	20.8	33.9	b	78)	48.319	1.201	266	81.7	9	
29)	16,664	.286	107.9	43.6	ь	79)	48.788	1.068	141	104	9	
30)	18.05	.402	134	49.5	ъ	80)	49.448	.928	25.6	63.6	ю	
31)	18,909	.0862	30,9	12.8		81)	50,123	.2.36	12,2	18.0	c	
32)	19,293	3.254	39.0	39.0	Ь	82)	50.482	1.215	/5,3	51.9	c	
33)	20,132	. 158	20/	90.0 20.4	5	83)	51.275	3.442	115	50.0	C	
34)	20.020	.10/	41.0	39.0	Ľ	64) 95)	51,003	0.320	34.3	14.5	Ē	
30)	21,004	429	46.8	40.0	ь Б	(66	52.245	2.720	82.6	40.2	ь Б	
271	22,733	.427	40.0	21.0			54 140	270	24.2	74 1	č	
37)	23,400	1.050	210	21.0	ь Б	07) 00)	55 112	2 224	5.04	14.1		
30)	24 221	202	18.9	30.1	ь Б	80/	55.870	1 512	126	74.04	8	
40)	24.251	179	77 2	44 1	ĥ	90)	56 745	5 710	035	41.5	Я	
41)	24.735	.322	780	6.9	F	91)	56.532	3.510	7.5	4.0	9	
42)	25 067	.121	252	34.3	ŕ	92)	57869	.811	93.7	39.0	å	
43)	25.239	.0974	192	7.1	ŕ	. 93)	58.092	1.102	12.3	12.6	d	
44)	25,473	.437	438	21.8	Ŧ	94)	58,689	1,213	89.9	43.9	c	
45)	25,590	.190	253	10.4	Ē	95)	59.762	.202	142	47.3	d	
46)	25,757	.0976	131	45.5	f	96)	60,205	1.114	205	58.1	d	
47)	26,482	.524	161	48.9	ь	97)	60.815	.155	101	41.0	g	
48)	26.822	.238	126	5.5	d	98)	61.081	.805	281	121	g	
49)	27.182	.053	0.53	0.63	f	99)	62.443	.601	722	124	9	
50)	27,823	.715	87.3	50.6	ь						-	

a) not fit, taken from BNL-325, 2nd edition, Supplement 2, Vol. 111 (1965).

a) not nr, toten non order sconverged to 3%.
 c) good fit, parameters converged to 3%.
 d) fair fit, but not converged.

e) fair fit, this resonance is inserted to provide a better fit to the data, however, it may not be needed in a multi-level analysis.
 (13)

fair fit, not converged, parameters should not be taken too seriously.
 g) rather poor fit, these parameters are simply our best estimates at this time.

the absorption width properly between the capture and fission widths so that the integral value of $\overline{\mathbf{a}}$ that is calculated from these parameters agrees with the measured value.

IV. RESULTS OF THE TRIPLE FIT FOR NEUTRON ENERGIES < 63 ev⁺

Table I is a list of the resonance parameters that have been obtained from a simultaneous least-square fit to the ORNL-RPI fission and capture crosssection data and the Saclay total cross-section data. With a few exceptions

1

the theoretical cross-sections that are calculated from these parameters give a good fit to the experimental data over the entire energy range (< 63 ev). This does not mean, of course, that the least-square fit has converged to yield the correct parameters in every case. Comments concerning the fit and accuracy of the parameters are included in the table.

In the range 24.6 to 26 ev it may be better for reactor calculations to use the experimental cross-sections directly rather than the resonance parameters. In this range the capture is small and the effect of doppler broadening on the fission cross-section through this broad "bump" should be small.

The resonance integrals over the range 1.86 ev - 37.3 ev which are calculated from the parameters given in Table I are $\Delta I_f = 113.12$ barns and $\Delta I_c = 89.41$ barns, also $\overline{\alpha} = 0.516$ which is in good agreement with the measured integral value of $\overline{\alpha}$. Included in each resonance integral there is a contribution from a negative energy resonance, viz. $I_f = 6.5$ barns, and $I_c = 1.79$ barns. In fitting the data, contributions to the fission and capture cross-sections of the form $\overline{\sigma \alpha} = K\alpha/\sqrt{F} (E-E_o)^2$ were used for the negative energy resonance. The following parameters were assumed: $E_o = -1.45$ ev, $K_f = 176.79$ barns ev^{5/2}, $K_c = 31.71$ barns ev^{5/2}.

From 37.3 ev - 63 ev, numerical integration of the data yields $\Delta l_f = 22.4$ barns and $\Delta l_c = 10.7$ barns; from the parameters one obtains $\Delta l_f = 23.3$ barns and $\Delta l_c = 11.1$ barns.

V. ACKNOWLEDGMENTS

We are indebted to A. Michaudon for information about the Saclay measurements. We are particularly grateful to G. deSaussure and co-workers for the use of their data before publication and also for providing information on the experimental resolution and results of integral calculations. The Sigma Center of the Brookhaven National Laboratory has also been most cooperative. The computer programs used were ably programmed by Miss Marian Melnick and Mr. Steven Page.

+The capture and fission cross-section data used in the calculations described in this section are the final normalized ORNL-RPI data.

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DISCUSSION

J.J. SCHMIDT: Your Γ_f values for 235 U resonances appear to be much higher than those obtained from other experimental analyses. How do you explain this?

D.W. DRAWBAUGH: I have not seen the results of earlier analyses of the ORNL-RPI data, so I do not have any explanation for such a difference. I would point out, however, that we obtained somewhat smaller Γ_f values from the double fit to the Saclay fission and total cross-section data. We used the resolution provided by the experimentalists, without tampering with it, and obtained resonance integrals in good agreement with direct numerical integrations of the data both for capture and for fission. Session X

NEUTRON DATA EVALUATION

EVALUATION OF NEUTRON CROSS-SECTIONS: CALCULATIONAL METHODS AND EVALUATED LIBRARIES

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Abstract

EVALUATION OF NEUTRON CROSS-SECTIONS: CALCULATIONAL METHODS AND EVALUATED LIBRARIES. A number of theoretical models are useful, in conjunction with experimental data, in providing evaluated sets of neutron cross-sections for reactor and shielding calculations. In addition to some of these several derived data sets which we currently recommend are described.

For light nuclei with well-spaced resonances the OPTIC code adds resonant terms, $\arctan [R_k^T P_{\ell}/(1 - R_k^T S_{\ell})]$ to conventional optical model phase shifts. This permits us to treat the rapid variations in angular distribution in fine detail, and to produce more accurate values for group-averaged Legendre moments than are obtainable from the optical model alone. Oxygen and carbon cross-sections based on this model plus recent good resolution experiments are presented. In the former, the important window at 2.37 MeV is well reproduced.

The coupling for nuclei with low-lying collective states to the ground state is not negligible and the single-channel optical model requires generalization to a coupled-channel theory. Examples of elastic and inelastic scattering for ⁴⁸ Ti, ⁵² Cr, ⁵⁵ Fe, ⁹² Zr, ⁹⁴ Zr, ²²² Th and ²²⁸U are presented. The inelastic scattering angular distributions show characteristic direct-interaction asymmetries which are not reproducible by compound nucleus theories, while the elastic scattering curves show structure which is not easily fit by optical model calculations. The coupling between channels can be obtained from coulomb-excitation measurements of reduced transition probabilities, or from a shell model calculation which incorporates residual pairing and quadrupole forces.

For use in activation analysis, detailed (n, p) cross-sections are often required, whose behaviour near threshold dominates their average value in a fission flux. We have found the Hauser-Feshbach compoundnucleus theory to be much more reliable in this respect than the conventional schematic penetrability methods which neglect nuclear structure. At energies too high to be treated by the Hauser-Feshbach theory, we must rely on experiment and evaporation-type models. Evaluated cross-sections for ³⁹K, ⁴⁶Ti, ⁵⁴Fe, ⁵⁶Fe and ⁵⁸Ni are presented.

Because current measurements of the differential capture cross-section in ²⁵⁵U are in conflict with quite reliable integral measurements, we discuss an evaluated data set which favours the latter, and which is in good agreement with a number of other experiments. In a similar vein, we discuss a set of boundhydrogen cross-sections for the near-thermal region which is designed to remove the inconsistency created by the use of up-scattering cross-sections in down-scattering reactor codes.

Two theoretical models which are useful, in conjunction with experimental data, in providing neutron cross-sections for reactor and shielding calculations are outlined in this paper. Several derived data sets which we currently recommend are also described.

1. OPTIC - A PROGRAMME FOR THE CALCULATION OF NUCLEAR CROSS-SECTIONS USING THE OPTICAL MODEL PLUS RESONANT PHASE SHIFTS

OPTIC [1] calculates total, shape-elastic, and reaction cross-sections, using an optical model plus resonant contributions described by R-matrix

FRANCIS et al.

I. Total cross-section $0 - 2.6$ $2.6 - 15.0$ BNL 325, KFK 120 Bockelman, FossanOPTIC ExperimentII. Elastic cross-section $0 - 15$ Total minus non- elastic (summed over reactions)ExperimentIII. Inelastic cross-section $0 - 15$ Total minus non- elastic (summed over reactions)ExperimentIII. Inelastic cross-section $0 - 10$ KFK 120, Hall and BonnerExperiment Interpolation $0 - 10$ 14 $10 - 15$ KFK 120, Hall and BonnerExperiment Interpolation 14 $10 - 15$ ConnerInterpolation $0 - 5$ $5 - 8.8$ 12 $12 - 15$ BNL 325Experiment Experiment Experiment et al. $8 - 12$ $12 - 15$ Bormann, Cierjacks, et al.Experiment Plus experiment $0 - 2 \cdot 15$ BNL 325Hauser-Feshbach plus experiment $0 - 2.6$ Lane, Langsdorf et al.OPTIC et al. Fowler and Cohn BNL 325, BNL 400 $0 - 2.6$ Lane, Langsdorf et al.OPTIC et al. Fowler and Sayres $4.7 - 5.0$ $5.0 - 15.0$ Chase et al.Optical model	Energy (MeV)	Experimental data source	Fitting procedure
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4.7 - 5.0Interpolation5.0 - 15.0Chase et al.Optical model	3.1 - 4.7	Lister and Sayres	Experiment
5.0 - 15.0 Chase et al. Optical model	4.7 - 5.0		Interpolation
	5.0 - 15.0	Chase et al.	Optical model

TABLE I.DATA SOURCES AND FITTING PROCEDURESFOR OXYGEN-16

theory. By including the resonance contributions, OPTIC is able to describe fluctuations in the scattering (total) cross-section which cannot be accounted for with a simple optical model.

For light nuclei, such as carbon and oxygen, resonance structure is clearly resolved below 10 MeV. Optical-model fits tend to become unsatisfactory below this energy. On the other hand, R-matrix theory

E (lab, MeV)	E (CM, MeV)	Q	3	γ² (MeV)
0.4444	0.4180	1	3/2	0.3340
1.000	0.94068 ^a	2	3/2	а
1.3161	1.2380	1	3/2	0.0756
1,6594	1.5610	3	7/2	0.8100
1.8295	1.7210	2	5/2	0.0676
1.9029	1.7900	- 1	1/2	0.0350
2.3700	2.2294	0	1/2	0.0484
3.2423	3.0500	2	3/2	1.4400
3.8270	3.6000	1	3/2	0.3600

TABLE II. PARAMETERS FOR THE RESONANCES FOR OXYGEN-16

^a This is a single-particle resonance and is obtained from a potential well instead of from the R-function.

has difficulties, stemming from the assumption that the nucleus scatters like a hard sphere. OPTIC therefore calculates the scattering from an optical potential instead of a hard-sphere model. This particular combination of optical and resonant phase shifts has no rigorous justification, but it is clear that a relationship exists between the optical model and the infinity of levels in R-matrix theory. Replacing the hard-sphere phase shift by an optical one is a reasonable way of accounting for these distant levels.

At energies sufficiently low so that only elastic scattering can occur, the quantum numbers j and ℓ (total and orbital angular momentum) characterize the phase shift, which for chargeless particles is given by

 $\tan^{-1} \left[R_{i\ell} (E) P_{\ell} (kR) / (1 - R_{i\ell} (E) \widetilde{S}_{\ell} (kR)) \right] + \phi_{i\ell} (kR)$

with the following notation:

 $R_{j\ell}$ (E) is the single-channel R-function: $\sum_{\lambda} \gamma_{\lambda}^{2} / (E_{\lambda} - E) + R_{j\ell 0}$

 $(R_{i\ell0}$ reflects the effect of distant levels); R is the nuclear radius; $P_{\ell}(kR)$ is the penetration factor, $P_{\ell}(kR) = kR [F_{\ell}(kR)^{2} + G_{\ell}(kR)^{2}]^{-1}$ (F_{ℓ} and G_{ℓ} are spherical Bessel functions); $\tilde{S}_{\ell}(kR)$ is the shift factor, $-b_{\ell} + kR(F_{\ell}F'_{\ell} + G_{\ell}G'_{\ell})(F^2_{\ell} + G^2_{\ell})^{-1}$ and $\phi_{j\ell}$ (kR) is either an optical model or a hard-sphere phase shift.



FIG.1. Resonance-plus-potential well fit to ¹⁶O total neutron cross-section below 3.0 MeV.

2. OXYGEN-16 CROSS-SECTIONS BASED ON AN OPTICAL MODEL PLUS RESONANT PHASE SHIFTS

A summary of data sources and fitting procedures is shown in Table I, and the parameters used for the resonances are shown in Table II.

In conjunction with our currently recommended hydrogen library, this set of oxygen cross-sections gives a water age of 26.5 cm^2 to indium, in good agreement with experiment. The fit to the total cross-section below 3 MeV is shown in Fig. 1.

Complete references are available in Ref. [2] which also gives the optical model parameters used in the calculation.

3. CARBON-12 CROSS-SECTIONS BASED ON AN OPTICAL MODEL PLUS RESONANT PHASE SHIFTS

A summary of data sources and fitting procedures is shown in Table III, and a summary of fitting parameters is shown in Table IV.

A segment of the fit to the total cross-section is shown in Fig. 2. Complete references are available in Ref.[3]. The programme 2 PLUS performs the coupled channel calculation described in the following section.

4. COUPLED-CHANNEL CALCULATION OF NEUTRON CROSS-SECTIONS

For nuclei with low-lying collective states, the latter's coupling to the ground state is not negligible and the single-channel optical model requires generalization to a coupled-channel theory. We have made coupled-channel calculations for even-even nuclei with first excited 2+ states using the vibrational nuclear model to obtain the

Energy (MeV)	Experimental data source	Fitting procedure
I. Total cross-sectio	n	
0 - 4	BNL 325, Bockelman, Wills KFK 120	OPTIC
4 - 15	BNL 325, Fossan	Experiment
II. Elastic cross-section	on	
0 - 9.2	Total - NE (summed over reactions)	OPTIC
9.2 15	Total - NE (MacGregor and Booth)	Interpolation, 2 PLUS
III. Legendre moment	S	
0 - 7		OPTIC
7 - 8.4		Interpolation
8.4 - 15		2 PLUS
IV. Inelastic cross-se	ction	
0 - 9.2	Hall and Bonner	Experiment
9.2 - 15	NE - other processes	-
V. (n, α) cross-section	n	
6.18 - 7.9	Coulomb penetrability	
7.9 - 8.65	Davis, Bonner et al.	
7.79.9	Risser, Price et al. (inverse)	Experiment
9.9 - 15		Interpolation
14.0	Al-Kital and Peck	
VI. (n, n [*] , 3α) cross-s	ection	
Threshold	Vasilev et al.	Experiment
-15	Frye, Rosen, et al.	

TABLE III.DATA SOURCES AND FITTING PROCEDURES FORCARBON-12

coupling between the two channels. In the vibrational model of Bohr and Mottelson the nuclear surface is assumed to be deformed dynamically and the radial co-ordinate of the surface may be defined by

$$\mathbf{R}(\mathbf{\hat{r}}) = \mathbf{R} \left[1 - \sum_{\mathbf{M}} \alpha_{2\mathbf{M}} \mathbf{Y}_{2}^{\mathbf{M}}(\mathbf{\hat{r}}) \right].$$

The optical potential experienced by a neutron at \vec{r}_0 is assumed to depend only on the distance from the neutron to the nuclear surface and may be

	l	ľ	γ2
I. E< 3.4 MeV	(+ hard-sphere phase	e shift: R=3.719, R _{1±0} =0.	.05, $R_{1}_{30}^{3} = 0.15$)
-6.0	0	1/2	-2- 4.0
1.915	2	5/2	0.025
2.733	2	3/2	0.175
3.383	2	3/2	1.638
3.55	2	3/2	optical
3.967	1	1/2	0.087
II. 2.9 < E < 7.0 (D 3/2 reson	MeV (+optical phas ance at 3.55 MeV d	e shift with energy-depend ue to potential well))	ent parameters
 II. 2.9 < E < 7.0 (D 3/2 reson 2.733 	MeV (+optical phas ance at 3.55 MeV d 2	e shift with energy-depend ue to potential well)) 3/2	ent parameters 0.3925
 II. 2.9 < E < 7.0 (D 3/2 reson) 2.733 3.940 	MeV (+optical phas ance at 3.55 MeV d 2 1	e shift with energy-depend ue to potential well)) 3/2 1/2	ent parameters 0.3925 0.1415
 II. 2.9 < E < 7.0 (D 3/2 reson) 2.733 3.940 4.548 	MeV (+optical phas ance at 3.55 MeV d 2 1 1	e shift with energy-depend ue to potential well)) 3/2 1/2 1/2	ent parameters 0.3925 0.1415 0.00622
II. 2.9 <e <7.0<br="">(D 3/2 reson 2.733 3.940 4.548 4.944</e>	MeV (+optical phas ance at 3.55 MeV d 2 1 1 1	e shift with energy-depend ue to potential well)) 3/2 1/2 1/2 3/2	ent parameters 0.3925 0.1415 0.00622 0.01736
II. 2.9 <e <7.0<br="">(D 3/2 reson 2.733 3.940 4.548 4.944 5.800</e>	MeV (+optical phas ance at 3.55 MeV d 2 1 1 1 3	e shift with energy-depend ue to potential well)) 3/2 1/2 1/2 3/2 7/2	ent parameters 0.3925 0.1415 0.00622 0.01736 0.9058
 II. 2.9 < E < 7.0 (D 3/2 reson) 2.733 3.940 4.548 4.944 5.800 III. 8.4 < E < 15 	MeV (+optical phas ance at 3.55 MeV d 2 1 1 1 3 MeV (2 PLUS param	e shift with energy-depend ue to potential well)) 3/2 1/2 1/2 3/2 7/2 neters)	ent parameters 0.3925 0.1415 0.00622 0.01736 0.9058
 II. 2.9 < E < 7.0 (D 3/2 reson) 2.733 3.940 4.548 4.944 5.800 III. 8.4 < E < 15 V_R = 50.36 	MeV (+optical phas ance at 3.55 MeV d 2 1 1 3 MeV (2 PLUS param $V_I = 0$	e shift with energy-depend ue to potential well)) 3/2 1/2 1/2 3/2 7/2 neters) VGAUS = 6.76	ent parameters 0.3925 0.1415 0.00622 0.01736 0.9058 $V_S = 5.0 R = 2.5$

 TABLE IV.
 FITTING PARAMETERS FOR CARBON-12



FIG. 2. Resonance-plus-potential well fit to ¹²C total neutron cross-section from 2.8 to 5.0 MeV.

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FIG.3. Coupled-channel fit to ⁴⁸ Ti and ⁵⁶Fe elastic and inelastic cross-sections at 2.45 MeV (Conventional potential wells plus deformation).

expanded to first order in the α_{2M} 's to yield

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$$V(\mathbf{r}_{0} - \mathbf{R}(\mathbf{\hat{r}}_{0})) \approx V_{0}(\mathbf{r}_{0}) + \mathbf{\hat{R}} \frac{dV_{0}}{d\mathbf{r}_{0}} \sum \alpha_{2M} Y_{2}^{M}(\mathbf{\hat{r}}_{0})$$

$$\langle \psi_{00} | \alpha_{2M} | \psi_{2\mu} \rangle = (-1)^{M} \langle \psi_{2-\mu} | \alpha_{2M} | \psi_{00} \rangle = -\frac{\beta}{\sqrt{5}} \delta_{M\mu}$$

$$\langle \psi_{2\mu} | \alpha_{2M} | \psi_{2\mu} \rangle = \langle \psi_{00} | \alpha_{2M} | \psi_{00} \rangle = 0$$

The central potential $V_0(r_0)$ is a Saxon-Woods well plus an imaginary Gaussian well and spin-orbit coupling. Also shown are the nuclear matrix elements which define the operators α_{2M} where ψ_{00} and the $\psi_{2\mu}$'s are the nuclear ground and first excited states respectively. The parameter β is a measure of the distortion of the nuclear surface. The coupling potential can then be written

$$\langle \psi_{2M} | V(r_0 - R(\hat{r}_0)) | \psi_{00} \rangle = -\frac{\beta}{\sqrt{5}} R \frac{dV_0}{dr_0} Y_2^M (\hat{r}_0)$$

The compound elastic and inelastic scattering are calculated using Hauser-Feshbach theory and the coupled equations are solved using the programme 2 PLUS [4].



FIG. 4. Coupled-channel fit to ⁴⁸ Ti and ⁵⁶Fe elastic and inelastic cross-sections at 2.45 MeV (Potential wells derived from a Yukawa two-body interaction).

Figure 3 shows fits obtained with this model for titanium and iron at 2.45 MeV. On the top is the fit to elastic differential cross-section data for natural Ti and inelastic cross-section data for exciting the 0.99 MeV 2+ level in ⁴⁸Ti. The difference in the elastic cross-section for natural Ti and ⁴⁸Ti is expected to be small. In the bottom curve the elastic data are for natural Fe and the inelastic data are for exciting the 0.845-MeV level in ⁵⁶Fe. The values for the distortion parameter required for the Ti and Fe fits were 0.21 and 0.28, respectively. The direct inelastic scattering cross-section calculated was significant, being about 20% of the total inelastic cross-section for both nuclei. The non-spherical part of the optical potential also had an appreciable effect on the shape of the differential elastic cross-section, decreasing it at the very forward and backward angles and at the diffraction minimum. The resulting shape is not easily fit with a spherical optical potential. Fits were also obtained for these nuclei at different energies and for Cr. Zr, and C. In all cases the calculated direct inelastic scattering was significant.

We have also calculated cross-sections for Ti and Fe using the shell model with residual pairing and quadrupole forces to describe these nuclei. The single-particle wave functions used in the nuclear wave function were calculated from a Saxon-Woods well with coulomb and spin-orbit forces. The single-particle energies used were those measured in deuteron stripping experiments and the pairing force CN-23/47

coupling constant was taken to be 23/A MeV where A is the atomic number. The nuclear wave functions were then calculated in the Boson approximation with the strength of the quadrupole force determined by matching the energy of the first excited 2+ state.

The interaction between the scattered neutron and the target nucleons is represented by a two-body, central, non-exchange potential with a Yukawa shape given by

$$V_{I} = \frac{C}{4\pi} \sum_{i} \frac{e - |\vec{r}_{0} - \vec{r}_{i}|/\mu}{|\vec{r}_{0} - \vec{r}_{j}|/\mu}$$

The sum is over all nucleons in the nucleus and the range μ is taken to be 1 fm. The coupled-channel equations arising in these calculations have exactly the same form as those based on the vibrational model just discussed. The spherical and non-spherical parts of the optical potential in this case are matrix elements of V₁ and the potentials shown above become

$$\begin{aligned} \mathbf{V}_{0}\left(\mathbf{r}_{0}\right) &= \langle \psi_{00} \mid \mathbf{V}_{1} \mid \psi_{00} \rangle \\ &- \frac{\beta}{\sqrt{5}} \operatorname{R} \frac{\mathrm{d}\mathbf{V}_{0}}{\mathrm{d}\mathbf{r}_{0}} \operatorname{Y}_{2}^{\mathsf{M}}\left(\mathbf{\hat{r}}_{0}\right) \rightarrow \langle \psi_{00} \mid \mathbf{V}_{1} \mid \psi_{2\mathsf{M}} \rangle \end{aligned}$$

The main difficulty with the formulation of the scattering problem at this point is that the spherical part of the optical potential is real and there is thus no compound nuclear reaction. We therefore assume that the imaginary potential arises from interactions with nucleons outside the closed shells, and consider it to arise from a two-body interaction with the same Yukawa shape between the neutron and extra-core nucleons (nucleons in the $f_{7/2}$ and higher shells). It thus has the form

 $iN \langle \psi'_{00} | V_I | \psi'_{00} \rangle$. Here ψ_{00}' is the ground state wave function with the core nucleons removed and N is a normalization constant.

We therefore have two adjustable parameters, C and N, to fit both the elastic and inelastic cross-sections. A value of 400 MeV was used for C and the strength of the imaginary potential was determined by fitting the total inelastic cross-section.

Figure 4 shows fits obtained in this way to the Ti and Fe data just shown. Here the calculated direct inelastic scattering was again about 20% of the total inelastic.

Therefore the calculations with both nuclear models used here indicate that the direct inelastic scattering is important for these nuclei even at 2.45 MeV and should be taken into account in order to make accurate calculations. Also, care should be used when calculating the elastic cross-sections of nuclei with low-lying collective states since removal of the non-spherical potential destroys the fits presented here.

5. URANIUM-235 CROSS-SECTIONS

No existing set of pointwise evaluated 235 U cross-sections that we are aware of has an epithermal alpha (= capture/fission ratio) as low as 0.50, the latest recommendation based on integral measurements [5]. (Typical values run from around 0.6 to around 0.7.)



FIG. 5. Evaluated activation cross-sections (Fission spectrum averages in mb given in square brackers).

It appears that the usual procedure of subtracting fission and scattering from the total cross-section to get capture values systematically overestimates the latter. This procedure also tends to obscure the characteristic differences in shape between capture and fission resonances which are ascribable to multi-level effects in the fission channels. Both of these difficulties are avoided in the present work, by relying where possible on the direct capture measurements made earlier this year by the Oak Ridge-Rensselaer Polytechnic Institute group [13].

Starting with Westcott's recent recommended set of 2200 m/s cross-sections [6], we used the fission measurements of Leonard [7] and the Livermore group's [8] 1966 data in the region up to 0.4 eV, fitting these, as needed, by least-squares polynomial fits. From 0.4 to 62 eV, we used the Oak Ridge-RPI fission data, normalizing it so that its integrated value from 0.4 to 62 eV matched the Livermore value. The latter has the advantage of being directly normalized to the 2200 m/s value. From 62 to 10000 eV we used Saclay data [9], and from 10000 to 2×10^7 eV a curve similar to the BNL 325 eyeguide, but adjusted to pass through the 24-keV value of Perkin et al. [10]. This composite curve gave a resonance integral of 281 b from 0.5 to 10^7 eV, compared to the Feiner and Esch value of 280 ± 11 .

For capture, we again used the ORNL-RPI data from 0.4 to 62 eV, but above and below that region we used capture-fission ratios times the previous fission curve. Below 0.4 eV the alpha values came from BNL 325 and Westcott [6]. From 62 to 10000 eV we used the values in KFK 120 [11]. From 10000 to 2×10^7 eV we used BNL 325 plus a smooth extrapolation.

This composite cross-section has a resonance integral of 139.8 b from 0.5 to 10^7 eV, compared to the Feiner-Esch value of 140 ± 8 . The alpha value of these two sets is 0.497, in good agreement with the Feiner-Esch value of 0.50 ± 0.02 . If the ORNL-RPI capture measurements
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can be independently verified, this will resolve the long-standing discrepancy between the differential and integral alpha values.

We are carrying out both Breit-Wigner single-level and Reich and Moore multi-level fitting to this set of cross-sections. The singlelevel procedure is relatively simple, but does not give a very good fit to the fission data. For example, the total widths of some resonances are 15% larger when seen in fission than when seen in capture. The twochannel Reich and Moore procedure can reproduce this type of behaviour but is difficult to parameterize. We are programming a version of the Reich and Moore procedure which will utilize an automatic parameter search but this programme is not yet operating. An interesting fact which emerges from even the preliminary analysis is that there is considerable fluctuation in the capture width distribution, suggesting collective effects in the gamma transitions.

6. ACTIVATION CROSS-SECTIONS

The procedure of using Hauser-Feshbach theory to generate the threshold dependence of (n, p) cross-sections has been described previously [12]. In Fig. 5 we show the results of combining this procedure with experimental data at higher energy, and then adjusting each curve to yield a recommended average value on a fission spectrum. The square bracket after the isotope designation in each curve gives the fission spectrum average. We show two curves for 46 Ti since the literature is evenly divided on whether the fission average is about 8 mb or about 13 mb.

Evaluated cross-sections have also been adopted for 27 Al (n, α), 32 S(n, p), 237 Np(n, f) and 238 U(n, f), but these were based entirely on experimental data.

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DISCUSSION

A. MICHAUDON: I should like to point out that, in trying to fit a theoretical curve to a set of experimental points, it is necessary to take into account two effects that are nearly always neglected because they are too difficult to estimate, namely, (1) interference between levels (due to fission) and (2) non-detected levels, which for 235 U amount to 20%. This effect may be more important than the first.

Dr. Adler (private communication) has obtained better agreement with experimental results by taking account of non-detected levels.

C.R. LUBITZ: With regard to the interference between levels, I realize that multi-level fitting is the best method. However, we found that one must first have a good idea of the single-level parameters. We therefore decided to carry out a single-level fit first, followed by a Reich and Moore one-channel fit.

H. NIFENECKER: I should like to point out, in connection with the question of non-detected levels, that, using only single-level formulae, it is possible to obtain an excellent fit to the cross-section curves of 233 U by adding a small number of "hidden" resonances. It has been proved by a Monte Carlo study of simulated cross-sections, however, that resonance parameters determined in this way involve considerable errors.

C.R. LUBITZ: There has been little discussion of the value of such fits in reactor design, where of course the requirements are quite different from those for obtaining fundamental information about 235 U or 238 U. Even though large fluctuations in the data do not represent individual levels in the compound nucleus, it is important – from the point of view of the use to which the data will be put – that we succeed in fitting such fluctuations. I have been interested to see how differently the resonance parameters come out when one fits the new Oak Ridge data, compared with the use of the older procedures of fitting total and fission data without any real knowledge of the capture widths.

PRINCIPLES AND PROBLEMS IN NEUTRON NUCLEAR DATA EVALUATION

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Abstract

PRINCIPLES AND PROBLEMS IN NEUTRON NUCLEAR DATA EVALUATION. The history of neutron nuclear data evaluation is briefly summarized. The physical problems involved in nuclear data evaluation, such as discrepancies and inconsistencies between different experimental data sets and gaps in experimental information, are discussed. The discrepancies in the capture cross-section data for molybdenum and iron are chosen to illustrate the great difficulties in systematizing and automatizing the evaluation process. The technical problems of data evaluation, such as computer storage and the establishment of nuclear data files, are not discussed.

In the last ten years the evaluation of nuclear data has evolved as a separate branch of applied nuclear science. Particularly the evaluation of low energy neutron nuclear data, with which we are dealing here, has attained eminent importance. In the course of development the term evaluation has become the common name to denote an activity which consists in the establishment of a complete chain of cross-sections or other nuclear data characterizing a certain reaction with a certain nuclide in a given energy range. This activity proceeds in several steps, beginning with the compilation of all available experimental references and data concerned, and continuing with a critical judgement and comparison of this information with the aim of elaborating it into a complete, unequivocal set of so-called best or recommended data. Complete means, in a larger sense, that no gaps are left, in a more specific sense, that the energy dependence of a cross-section is reproduced in an "almost monochromatic" way allowing a simple interpolation between adjacent data points.

The requirement of completeness involves the recourse to nuclear theory and systematics in the case of gaps and inconsistencies of the experimental data. Naturally evaluation also comprises, with a similar requirement of completeness, discontinuous and parametric data such as level schemes and resolved resonance parameters. In view of the fact that the gathering of experimental or theoretical data for comparison with a new theory or a new experiment is a very old and basic scientific job, the immediate question arises: What has made the evaluation, particularly of neutron nuclear data, almost a field of its own with such specific stringent terms of reference. A brief historical review will give an answer to this question.

Evaluation, as defined above, is primarily and closely related to the development of nuclear reactors. It never would have attained such importance, if, as at the beginning of reactor development, only thermal reactors had been designed. The physical behaviour of these reactors is mainly governed by the reactions the neutrons undergo in the small

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thermal energy range. Apart from the rather complex thermal inelastic scattering interactions, the energy dependence of the cross-sections is rather simple, the important capture cross-section for example following almost always the simple 1/v law. In principle, few group calculations with adaptation to integral parameters were thus sufficient to predict the neutronic behaviour of a thermal reactor; the gathering of the few necessary data could easily be done by the reactor physicists themselves. This was the somewhat simplified picture in the early stages of the reactor development.

With the design of fast and intermediate breeder reactors and with the rapid development of ever larger and faster electronic computers this situation changed completely. In small fast-critical assemblies and metal-fuelled fast reactors neutrons concentrate on keV and MeV energies. Large dilute oxide or carbide-fuelled intermediate power breeders contain neutrons down to the eV region. For shielding calculations the MeV range of neutron energies for calculating reactor neutron energy spectra and for safety considerations (calculations such as the Doppler, steam and sodium void coefficients) an extensive knowledge of the resonance properties of almost all reactor materials, in particular of the heaviest ones, became of urgent interest. Concurrently the computer development allowed and forced the development of advanced reactor theory programmes such as the one- and two-dimensional multigroup diffusion and transport codes and detailed Monte Carlo programmes. These comprehensive programmes enabled a more detailed and precise description of the nuclear properties of reactors, in particular of thermal reactors, than previously. The usefulness of these programmes and the reliability of their results depended to a large extent on the detailed and reliable knowledge of the microscopic nuclear data involved and their effective utilization in these programmes. The original, rather easy task of gathering thermal nuclear data grew to the more complicated and comprehensive task of gathering microscopic neutron data for all occurring nuclear interactions in the larger energy range, defined by nuclear physics as the low energy range (0 to 10 MeV and above); since the reactor neutrons do not leave out any energy sub-range or any reaction in this entire region, the gathering of the data had to be complete with regard to the energies and the reactions covered for a given nuclide. Thus the principal requirements for such a collection of neutron nuclear data outlined at the beginning of this paper evolved in quite a natural way.

At first one might think that the gathering of such data could not be such a large task and that nuclear theory calculations would give necessary and sufficient information. Unfortunately, however, as is well known, no unified nuclear theory exists, which allows the reliable prediction of all neutron nuclear interaction probabilities in the range from 0 to 10 MeV. If such a theory existed, one could conceive a coupling of nuclear and reactor theory programmes, the output of the former being the input of the latter, as a satisfactory solution of the entire data problem. Instead only nuclear models with a validity restricted to certain energy ranges, reactions and nuclides and parametric theories are available, which, for such accurate and reliable predictions of nuclear parameters and cross-sections as are needed in reactor calculations, can in most cases only be applied with reasonable success, if some or all parameters entering the theory are taken from experiment.

Thus, if this purely theoretical way does not exist or at best is only successful (and then no longer pure) with the help of experimental parameters, it is necessary to find a purely experimental way) e.g. to construct one or a few experimental apparatus and measure with these facilities the necessary data in the entire energy range of reactor neutrons with the high accuracy desired and a perfect energy resolution throughout. Such an ideal machine which, if available, would immediately solve the entire problem will probably never be invented: the machine could work on line with the reactor computer programmes and feed its data directly into these. In reality, however, one finds that just the opposite is true. A large variety of experimental facilities, such as choppers, Van de Graaff machines, linear accelerators and others, is available in almost all parts of the world, each type of machine being successfully applied only in certain energy ranges and for certain nuclear reactions. Actually, the development, refinement and rapid distribution of these machines was due to the data requirements of reactor physics, which were set forth, for example, in the well-known neutron nuclear data request lists of the EANDC. These machines now produce increasingly large amounts of data, which have first to undergo the rather complicated and lengthy procedures of mutual comparison, critical evaluation and selection, tabulation and computer storage, before being successfully fed and elaborated in the reactor theory programmes. Even so the purely experimental way does not suffice for yielding all necessary data. Unavoidable gaps exist due to the limited nature of the machines in energy resolution, energy ranges and reactions to be covered. Avoidable gaps exist because not all needed data have hitherto been measured. Finally more or less severe discrepancies and inconsistencies between different data sets are frequently encountered, reflecting the large difficulties connected with the experimental apparatus and measurement techniques, discrepancies which often cannot be solved on an experimental basis alone. However, reactor neutrons obviously do not know of these gaps and discrepancies, and the natural way to solve these difficulties is by recourse to estimates based on nuclear systematics and nuclear model calculations with the partial or full use of "best" guesses of the nuclear parameters involved.

This brief resumé reveals not only the principles, but also gives an idea of the difficulties and problems involved in every evaluation. An evaluation physicist has to be systematically aware of all experimental techniques and data concerned. At the same time he should be familiar with current and relevant nuclear models, theories and computer programmes. The emphasis he places on certain aspects of his work is governed by the needs and importance of reactor physics; he should therefore be aware of the current, main problems in reactor physics and work closely with reactor physicists. Finally he is always faced with a variety of computer organizational problems. Obviously neither the experimental or theoretical nuclear physicist nor the reactor physicist can perform the work of an evaluation physicist in addition to his own job; such a task is too large. The main problem therefore for an evaluation physicist is to be always up to date in, and aware of, all these aspects of his work. This explains our first assertion that evaluation has become a branch in itself in the domain of applied nuclear physics; it also makes

clear that evaluation can only be efficient when carried out in close cooperation with reactor and experimental and theoretical nuclear physics.

Now we turn more extensively to some of those problems encountered during the evaluation itself, which were already mentioned above. Technical problems connected with the computer storage of evaluated data and the establishment of nuclear data files, although forming an essential and difficult part of every evaluation, are omitted from our considerations, and the interest is focussed on physical problems.

Discrepancies and inconsistencies between different experimental data sets and their solution represent the largest problem in almost every evaluation, regardless of neutron energy, reaction and nucleus concerned. Naturally no two measurements are made under exactly the same conditions, but the results should be compatible and, if reduced to the same experimental conditions, be in agreement, at least within the range of the mutual uncertainties. However, frequently the results of two or more measurements, in spite of the corrections applied, differ systematically by a larger amount than the uncertainties of each individual measurement, showing that the sources of systematic errors have not been completely removed. One of the most easily recognizable reasons for such systematic discrepancies is different normalization; this is encountered often particularly in capture, (n, p) and (n, α) reactions. In principle this deficiency can be removed by measuring and/or evaluating accurate standard data and by renormalizing the original experimental results, an important task to which much work has already been devoted. In the majority of cases, however, the reasons for such systematic discrepancies are much more difficult to find, being most probably closely associated with the experimental apparatus and techniques, ambiguities in the interpretation of the measured raw data and so on. A famous example is the discrepancy between the Livermore data on the one hand and the Saclay and Harwell on the other for resonance fission crosssection measurements on ²³⁵U; lengthy discussions among the experts concerned and a subcommittee of the EANDC were necessary to solve this discrepancy. One of the immediate consequences of such unsolved discrepancies is that the uncertainties of the evaluated data, which are a more or less sophisticated average through such discrepant measurements, are larger than the accuracies asserted to be achieved in the underlying experiments.

Another kind of inconsistency is encountered rather often in the range of overlapping resonances in the keV range of neutron energies, which reflects particularly the difficulties of the accurate determination of the neutron flux in this energy region: measurements with broader energy resolution and worse statistics show larger cross-section fluctua-tions than those with finer energy resolution and better statistics. A prominent example for this kind of inconsistency are the discrepant fission cross-section measurements on ²³⁹Pu in the lower keV range. Unfortunately the parametric character of the nuclear theory already invoked before in general prohibits an unequivocal solution of those discrepancies.

Some of the above discrepancies for two typical cases, i.e. the evaluation of capture cross-section measurements on Mo and Fe in the keV energy range are discussed now. The first example, already discussed in Ref. [1], is mentioned here again because most of the discrepancies existing then have meanwhile been resolved. On the contrary, the Fe keV-capture cross-section represents at present one of the largest unsolved discrepancies.

Figure 1 shows the presently available σ_{γ} measurements on Mo in the energy range between 1 keV and 1 MeV; no measurements are available above 1 MeV (the reference numbers in Figs 1 and 2 are those from



FIG.1. σ_v for natural Mo from 1 keV to 10 MeV

Ref. [2]). In 1961, apart from the 30- and 65-keV values of Gibbons et al. [Mo - C57] and the 24, 220 and 830 keV results of Belanova [Mo -C51] the following more comprehensive measurements were available: Block et al. [Mo - C62] from below 1 keV to 6 keV; Staviskii, Shapar [Mo - C67] from 50 keV to 1 MeV and Diven et al. [Mo - C63] from 175 keV to 1 MeV. We noted then that Staviskii's results differed consistently from those of Gibbons and Diven by +50 to 100%. This discrepancy could not be resolved; an average curve through these discrepant results and through the measurements of Block et al. [Mo - C62] was therefore recommended. Afterwards, to solve the discrepancy semi-empirical statistical theory calculations were carried out by d'Auria and Schmidt [3] for all Mo isotopes in the range between 1 keV and 1 MeV and added to get σ_{y} for natural Mo.

In these calculations statistical s-wave resonance parameters determined from measured resolved s-wave resonance parameters were used and spin, energy and, if not defined from resonance measurements, atomic weight dependences of the average level spacings were determined from the Fermi gas model of the nucleus. The transmission coefficients for elastic and inelastic scattering of neutrons with higher orbital angular momenta than 0 were computed from the simple complex square well potential of Feshbach et al. [4]. Finally statistical fluctuation factors and inelastic scattering competition to all known levels below 1 MeV were taken into account. Without going into the numerical details, the results SCHMIDT

of these calculations agreed better with the data of Gibbons and Diven et al. than with those of Staviskii and Shapar. These results later on were confirmed when it became apparent that the discrepancy was solely due to a wrong standard value used by the Russian authors. Originally they normalized their σ_{ν} data to an ¹²⁷I capture cross-section value of 400 mb at 200 keV. A later comparison with the particularly extensive $\sigma_{\rm v}$ measurements on 127I in the keV range performed by Bame and Cubitt [5] showed that this value was a factor of 1.6 too high. By lowering their standard value by this factor to 245 mb and renormalizing their data, Staviskii and Shapar obtained very good agreement with the results of Diven, as can be noted from Fig. 1, in which the corrected Russian results have been inserted. However, the good agreement now attained between these two measurements does not necessarily involve the correctness of these data on an absolute scale. This, however, is rather confidentially ascertained by the fact that both measurements agree in spite of normalization to different standards, and that, in particular, the ²³⁵U capture and absorption cross-section values Diven used for normalization appear to be reliable, agreeing to within a few per cent with the best presently available measurements on ²³⁵U [2]. A rather large difference, however, still remains between Staviskii's and Gibbons' [Mo - C57] results between 30 and 65 keV and is so far unexplained; this disagreement is the worse, as Gibbons' σ_{γ} values are normalized to reliable In σ_v standards (763 mb at 30 keV and 448 mb at 65 keV). The high 200 keV value of Leipunsky et al. [Mo - C81] inserted unchanged in Fig. 1 is due to the same wrong ¹²⁷I standard as used by Staviskii and Shapar. The results of Belanova [Mo - C51] are believed to be rather unreliable; in her method σ_v is essentially obtained as the difference of total and scattering cross-sections, and, as these are about equally large, great errors are necessarily involved in the difference. This explains the large discrepancy of Belanova's 830-keV value to the other experiments and renders the good agreement of the 24-and 220-keV values with the other experiments purely incidental; for other elements like Cr for example Belanova's results at all three energies differ from other data by an order of magnitude.

Two measurement series below 50 keV are now available from Kapchigashev and Popov [Mo - C74] and from Mitzel and Plendl [Mo - C75], both being performed by the method of the neutron slowing down in a lead pile. In spite of the similar method used and the rather good agreement of both measurements below 1 keV, above 1 keV Mitzel and Plendl's results are systematically lower than those of Kapchigashev and Popov by as much as 60% at 30 keV. The decision, which of both measurements is correct, becomes almost irrelevant, since the errors involved in both measurements are of the order of the difference between both measurements, i.e. 30% and more; the method involves the estimate of σ_{γ} from the difference of the combined $\gamma\text{-spectra of Pb}$ and the investigated material and of the γ -spectrum of Pb alone and, as σ_{γ} is already rather small above 1 keV, the cross-section values in the keV range obtained by the lead pile method necessarily become rather unreliable. The only argument in favour of Kapchigashev's results is that they join better to the renormalized results of Staviskii and Shapar, which are considered reliable, than those of Mitzel and Plendl. Both lead pile measurements, however, concordantly show, outside their experimental errors and in

agreement with the calculations mentioned above, that the earlier measurements of Block et al. [Mo - C62] below 6 keV are too high by more than a factor of 2. This discrepancy can also be explained: it is due to the lack of corrections for multiple scattering before capture and resonance self shielding in Block's measurement, both of which render the measured capture cross-section too high; particularly the latter correction has been applied in the two lead pile experiments and is responsible for most of the discrepancy. On the basis of the foregoing arguments it is understandable why the curve called "presently recommended" in Fig. 1 has been chosen as a smooth average through the results of Kapchigashev and Popov, Staviskii and Shapar and Diven et al.



FIG. 2. σ_v for Fe in the range 10 eV to 1 MeV

For the Fe capture cross-section in the keV range even more discrepancies are encountered and fewer of them solved than for Mo. Figure 2 shows the available experimental data. The region 100 keV to 1 MeV does not interest us here; apparently the different measurements agree in this range. The region of discrepancies for most of the available experiments extends from 100 keV to \sim 100 eV. We begin with a comparison of the two lead pile measurements of Isakov et al. [Fe - R57] and again of Mitzel and Plendl [Fe - R80]. As in the case of Mo, both measurements were performed under essentially the same experimental conditions. In spite of the similarity in method, two characteristic differences are apparent, a shift in the energy scale of about 1 to 2 keV between both measurements at energies above 2 keV, and different heights of the peaks observed in both experiments between 100 and 1200 eV. With

regard to the first discrepancy, the energy scale in the Russian measurements is probably correct. This is suggested by a comparison of the broad peaks observed in the broadly resolved capture measurements with the more detailed resonance structure as observed in much finer resolved transmission experiments. The transmission experiments reveal a smaller resonance at 6.0 keV in ⁵⁷Fe and a larger resonance at 8.0 keV in 54Fe. In the capture measurements one would thus expect to see a large peak centred nearer to 8 keV than to 6 keV, and this is actually only the case for the Russian measurement. The second discrepancy can be unequivocally clarified. The peaks observed below 1 keV are clearly due to Mo, Co and Mn impurities in the samples of both authors; the difference in the amount of these admixtures explains the difference in the observed peak heights. The larger peak height Mitzel and Plendl observe for the 1.2-keV resonance in Fe is also due to a much larger impurity admixture than in the Russian sample of 55 Mn which has a resonance at 1.08 keV. Thus one is led to the conclusion that there is no resonance in Fe below the 1.2-keV resonance and that the capture cross-section there follows an undisturbed 1/v law. This conclusion is furthermore ascertained by the very careful and well-resolved capture and transmission studies of Moore et al. [6] in the vicinity of 1 keV, which do not reveal any other resonance than that at 1.2 keV.

Thus the differences between the two lead pile measurements are well understood. More disturbing are the much larger discrepancies between the lead pile measurements on the one hand and the Harwell linear accelerator [Fe - R79] and Oak Ridge Van de Graaff [Fe - R82] measurements on the other in the range from about 1 to 100 keV. The small differences between these two latter measurements are due to differences in isotopic sample composition and do not concern us here. The much smaller shape and much higher peak cross-section observed in the Harwell measurement compared with the two lead pile experiments is clearly the result of the much better energy resolution in the Harwell measurements. Capture areas and resonance parameters deduced from all three experiments for the 1.2-keV resonance agree well with each other after due correction for the impurity admixtures in the lead pile measurements. The much finer resonance structure observed by Harwell and Oak Ridge above 1 keV, however, is clearly explained by the much finer energy resolution. The systematic discrepancy by an average factor of about 2 to 3 between Van de Graaff and linear accelerator and the lead pile results still remains unexplained. It is true that the linear accelerator measurements are still not corrected for multiple neutron scattering before capture; this means that the corresponding σ_{γ} values plotted in Fig.2 are actually too high. However, at best this correction is expected to be important in the large 28-keV resonance in 56 Fe and thus does not explain the discrepancies below this resonance; furthermore the Oak Ridge measurements are corrected for this effect and are actually smaller in this resonance than the Harwell measurements, but still about twice as large as the lead pile results. From the known capture widths of nuclei heavier than Fe (see e.g. the survey of Ref. [7]) one would expect capture widths of Fe resonances to be from 0.4 to 0.5 eV, values which actually could be derived from the observed lead pile capture areas, whereas the Harwell and Oak Ridge capture areas which are about 3 times larger only agree with capture widths 3 times as large. Thus this

argument based on nuclear systematics of the capture widths would be in favour of the lead pile results. On the other hand some serious doubt is cast on these results by the large uncertainties already discussed for Mo, connected with the subtraction of the Pb γ -ray background in the keV range; however, it cannot be said whether the true capture cross-section values are larger or smaller than those observed and whether the resulting large uncertainties in the lead pile σ_{γ} do not account for the discrepancy to the other measurements.

A final possibility for deciding between the discrepant measurements is to compare direct determinations of the non-1/v part of the infinite dilute capture resonance integral with values calculated from the experimental capture cross-section data. Without going into numerical detail, the result is that five independent direct measurements of this quantity are all between 2 and 4 times as large as the value calculated from the highest differential, i.e. the Harwell data (for details see section III of Ref. [2]). Our obvious conclusion from the above arguments consists in the preliminary recommendation (valid so long as these discrepancies are not understood and removed) of a 1/v behaviour below 1 keV and of the Harwell σ_{γ} results in the range 1 to 100 keV as representing most closely the true energy dependence of σ_{γ} for Fe; the curve called "presently recommended" in Fig. 2 shows this compromise.

We elaborated rather extensively on these two examples, to which others could easily be added, because they show better than any general discussion the true bottle-neck of every evaluation. It is almost needless to mention the impossibility of the reliable prediction of the physical properties of a fast reactor working with steel as structural material, if such large discrepancies are not removed.

A second general problem in evaluation, but generally not as severe as the discrepancies, are the gaps in experimental information. The simplest case has to do with a smooth, energy-dependent cross-section which is well established in the entire energy range concerned except a certain interval in which no data are available; simple graphical interpolation closes such a gap reliably. Such a graphical or a statistical theory interpolation is more questionable in a region of overlapping resonance structure; the error of the interpolation is of the order of the variance of the cross-section concerned. A gap in the resolved resonance range can, in principle, not be closed adequately, because there is no theory which predicts the position and properties of the resonances; here only the experiment can help, and any interpolation is necessarily pure invention.

Certain limits in the experimental techniques render the experimental investigation of one or the other cross-section impossible. In many cases the finite-energy resolution in the detection of the scattered neutrons, for example, prohibits measurements of the inelastic excitation of wellknown separate nuclear levels. In these cases the simple statistical theory developed by Hauser and Feshbach [8] with transmission coefficients from realistic optical potentials is used; it has predicted neutron inelastic excitation cross-sections within the, however, still rather large uncertainties and inconsistencies of the experimental data. Naturally this does not mean that the theory is able to replace a good experiment.

Also in other cases nuclear theory can be successfully used for closing gaps, provided that important parameters entering the theory are sufficiently well known. Due apparently to some cancellation effect in the neutron transmission coefficients, the generally very narrow distribution of the capture widths as concluded from resonance experiments, and their weak spin and energy dependences, the Hauser-Feshbach theory applied to fast capture cross-sections of medium weight and heavy nuclei yields rather reliable results. Above 1 MeV for many nuclei large gaps in experimental capture cross-section data exist; at the same time the compound hypothesis underlying the Hauser-Feshbach theory becomes more and more invalid and the direct capture more and more important; the theory for this transition range is still insufficient. Fortunately, from the practical point of view, for most nuclei the absorption of neutrons by reactions other than capture, mainly by the (n, p) and (n, α) processes. is more important in the MeV range. Unfortunately the experimental information on these latter reaction cross-sections is often rather scarce. if at all available, and in addition often discrepant. Although extensive work has been devoted to the refinement and improvement of optical model transmission coefficients for neutrons and charged particles and the level density laws, the predictions of the evaporation theory are still not reliable enough, ranging from rather incidental good agreement to complete disagreement with experiment. One of the reasons is the strong dependence of the evaporation cross-section expressions upon parameters influenced by shell structure and still not accurately enough known, such as the single-particle level density. The usefulness of refined optical models for the prediction of elastic scattering angular distributions and non-elastic scattering cross-sections at energies where the compound elastic scattering has died out is so well known and has so often been described that we can neglect it here. The practically and fundamentally equally important problem of the correct interpretation of fission resonances is also only mentioned here; the author considered this problem extensively elsewhere [9].

Thirdly we wish to draw attention not so much to a problem, but rather an inconsistency encountered in every evaluated data set, which is the result of a similar inconsistency in the experimental information used. In a given energy sub-range measurements of different neutron cross-sections generally differ in the experimental conditions, in particular in the energy resolution. The better resolved cross-section might still exhibit something like a resonance structure, whereas the other poorer-resolved cross-section shows a smooth energy dependence. A well-known example of this is the very broadly resolved KAPL reactor spectra α -measurements and the 10 ns/m and better resolved Saclay and Harwell fission cross-section measurements on ²³⁵U in the 100 eV to low keV range. The only possibility to render two such measurements consistent with each other is to fold the better-resolved measurement with the energy resolution function of the poorer-resolved measurement. Generally this is not done, in order not to lose the detailed information contained in the better-resolved measurement. However, if these two cross-sections have to be used to determine a third one, because this is not, or only with large difficulties, attainable by experiment, this third cross-section can have large errors, on the average of the order of its variance, depending upon the size of the difference in resolution in the first two cross-sections. In our example this is true of the capture cross-section of 235 U determined from σ_{f} and α . This rather frequent

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observation has only a consequence for the experiment: to get a physically true picture of "derived" cross-sections and nuclear data, the experimental conditions, foremost the energy resolution, in the measurement of the "basic" cross-sections and nuclear data should be as similar as possible. The more this condition is fulfilled in the experiments, the more physically meaningful and consistent in all basic and derived quantities become the evaluations.

In the foregoing we discussed some of the main physical problems encountered in the evaluation of neutron cross-sections. Their solution in each individual case influences not only the reliability of the evaluated data for the accurate calculation of reactor physical properties, but also their usefulness for the checking of nuclear theories. The examples we discussed demonstrate the great difficulties one is faced with in systematizing and automatizing the evaluation process. However, to keep pace with the still increasing amount of experimental data flowing in and to utilize to the fullest extent the possibilities of the large computers, automatization, without loss of the individual physical aspects, will certainly be one of the main future problems in evaluation.

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DISCUSSION

H. WEIGMANN: Preliminary data obtained by us on the capture cross-section of molybdenum between 1 keV and 20 keV are in full agreement with those of Kapchigashev and Popov, and accordingly with the solid line shown in Fig. 1 of the paper. However, we observe a relative maximum near 1.5 keV; this is also visible in the curve of Block et al.

J.J. SCHMIDT: Thank you for this information. I should mention that the data of Mr. Kompe and of Dr. Ponitz for the capture cross-section of molybdenum in the keV range could not be included. There is fairly good agreement between the two sets of data, with smaller values (particularly at low keV energies) than those corresponding to our recommended curve.

R. BATCHELOR: Should an evaluator allow his evaluations to be influenced by the results of integral experiments, and, if so, to what extent?

J.J. SCHMIDT: As far as epithermal and fast neutron energies are concerned, the feedback from integral to differential data is generally not unequivocal. This has two consequences: first, different adjusted data sets are likely to fit a series of critical assemblies equally well; second, one cannot be sure that such a data set, adjusted to current critical assemblies with harder neutron spectra, will allow the correct prediction of the physical properties of large power reactors with softened neutron spectra. Therefore, in the case of discrepancies between predicted and experimental data, most likely due to nuclear constants, I would generally prefer a thorough review of the basic microscopic data most probably involved to a data adjustment that may be physically incorrect.

J. CHERNICK: In connection with Mr. Batchelor's question, I would say that sometimes one must change the differential cross-section data to obtain agreement with the integral data, when one can trust the integral data but not some of the differential data. For example, if the capture cross-section data are the least reliable, they may be changed to agree with the integral data.

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R.D. SMITH: Dr. Schmidt has shown how much work is involved in a major evaluation of the data for one nuclide.

How often does he think it will be necessary to undertake a major reevaluation of, firstly, a nuclide of major importance in reactor design (for example, ²³⁸U) and, secondly, a nuclide of lesser importance (such as chromium) during the next five to ten years?

J.J. SCHMIDT: Assuming that new experimental data continues to become available at the present rate, I would say that a re-evaluation will be necessary every year in the case of an important nuclide such as 238 U and every two years for a less important element such as chromium.

NEUTRON CROSS-SECTION EVALUATIONS – PAST, PRESENT AND FUTURE

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Abstract

NEUTRON CROSS-SECTION EVALUATIONS - PAST, PRESENT AND FUTURE. Before the many and increasing results of neutron cross-section measurements and calculations can be applied to practical nuclear energy problems it is generally necessary to produce evaluations of neutron cross-sections. Such evaluations should ideally be complete (all possible reaction parameters detailed), internally consistent and based on the best available experimental and theoretical information and should cover a wide energy range - typically 0,0001 eV-20 MeV.

Many such evaluations have been made and have been exchanged between laboratories. Indeed the limited effort available for this work makes it an ideal field for international co-operation. To encourage the exchange of such evaluations and the production of new evaluations one of the authors has previously published EANDC(UK)26 and AWRE 0-13/65 reviewing the position at September 1963 and November 1964, respectively. In the present paper these reviews are updated to 1 June 1966 and the possible display of the information in a form suitable for computer processing and retrieval is discussed.

In future the ENEA Neutron Data Compilation Centre at Saclay hopes to collect details of all evaluations and distribute descriptions to interested people, so that the main content of the present paper will be continually updated.

For each element and for certain important isotopes and compounds the paper sets out the evaluations available, the energy range covered, the reactions considered and the availability of the data in tabular form or on punched cards/magnetic tape, insofar as such information is known. Multigroup cross-sections are not considered.

1. INTRODUCTION

1.1. The need for cross-section evaluations in science and technology

Scientists and technologists who need to make calculations involving neutron cross-sections have often been surprised to find that, in spite of the vast effort spent in measuring and calculating these cross-sections during the last thirty years, information is not always readily available in an easily usable form. Depending on the cross-section and the energy range the data may be abundant but conflicting (different values for the same parameter often differing by more than the quoted error), internally contradictory (partial cross-sections inconsistent with total cross-section),

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sparse or non-existent. In any case the sheer volume of data - witness the $35\,000$ entries in CINDA 66 [1] - will daunt the casual enquirer. As with many other kinds of physical and chemical data there is a clear need for compilations and evaluations of the available data.

1.2. Scope of review

The terms compilation and evaluation require some discussion. In the present context compilation means the collection of all experimental measurements (and possibly theoretical calculations) of particular crosssections for particular materials. Compilation activities have been carried out in several laboratories but are now becoming increasingly concentrated in the Brookhaven, Obninsk, Saclay and Vienna data centres.

The term evaluation presents more difficulty. It can be justifiably defined as the logical derivation of preferred (best) values for individual physical parameters so that, for example, one might evaluate the fission cross-section of 235 U for 3-MeV neutrons. In this review the term evaluation is, in general, taken to refer to several, often interrelated parameters over a fairly large neutron energy range.

Such a definition is appropriate because the most important application of evaluated neutron cross-sections is in neutronics calculations where it is usually necessary to consider not only the overall crosssections for several reactions in any material but also differential crosssections in scattering angle and secondary neutron energy, all as continuous functions of incident neutron energy over a fairly large energy range. The energy range of most interest for nuclear energy applications is 0.0001 eV-20 MeV which suffices for all conventional reactor requirements but in analysing nuclear physics and experiments and in space applications much higher energies may be involved so that it seems impossible to impose any limits on the energy range which may be covered in the future.

2. THE GROWTH OF NEUTRON CROSS-SECTION EVALUATION WORK

The earliest method of obtaining evaluated neutron cross-sections was apparently to "ask Fermi" [2]. From its inception Brookhaven National Laboratory has been very active in the neutron cross-section compilation and evaluation fields and until around 1956 was virtually the sole contributor. The publication of the successive editions of BNL-325 [3] and BNL-400 [4] has been of immense help to nuclear and reactor physicists throughout the world. Although these reports have always been intended primarily as compilations of available experimental data they have included a good deal of evaluation - best values of resonance parameters and 2200-m/s cross-sections of both fissile and non-fissile nuclides and preferred curves through experimental cross-sections. However in the present sense the evaluation is generally incomplete in that no attempt is made to include theoretical curves for those cross-sections which have not been measured at any energy. There are many neutronics calculations for which nuclear data cannot be prepared using BNL-325 and BNL-400 alone.

From the mid-1950's onwards there has been a steady growth in neutron cross-section evaluation work for several reasons.

- (a) The development of high speed computers revolutionized neutronics calculations. No longer did calculations require only a few simple nuclear parameters. Multigroup transport and diffusion codes and Monte Carlo codes required large numbers of group cross-sections which in turn could only be properly prepared given all partial crosssections as functions of energy over a wide energy range.
- (b) The experimental nuclear physicists have become much much more efficient at turning out vast quantities of data by employing better nuclear physics machines, more people and, not least, computer techniques for handling numerical data. However discrepancies between different experiments are still numerous and there has been considerable pressure on evaluators to resolve these discrepancies as far as possible and so enable data acquired at considerable expense to be used in practical calculations.
- (c) The nuclear energy industry has developed rapidly and many more laboratories have found it necessary to undertake neutron crosssection evaluation work in the absence of any universally acceptable complete evaluation for all materials.

The late fifties saw several laboratories moving into the neutron cross-section evaluation field and the number increased again during the early sixties but gradually it was realized that the task of providing evaluations for all materials was beyond the means of any single laboratory. Inevitably this realization led to co-operation and exchange between laboratories and this was encouraged as EANDC and other organizations arranged meetings of evaluators starting with a small meeting of 7 people at the Atomic Energy Establishment, Winfrith in October 1962. The present conference is the latest in a line which have included meetings at Brussels in September 1963, at Brookhaven in May 1964 and May 1965 and at Washington in March 1966.

These have led to much greater contact between individual evaluators whilst simultaneously data centres such as those at Brookhaven and Saclay have become interested in promoting the exchange of evaluated data. The numbers and standards of evaluations have risen in response to this stimulation.

Mention should be made of efforts to standardize the presentation of evaluated cross-sections in a single international format. A detailed discussion has been given by Parker and Honeck [5]. Briefly it was the hope of many that a single standard format could be devised so as to facilitate the exchange of evaluated data on magnetic tape. The prior existence of some twenty different individual laboratory formats has presented great difficulty and to date progress is slow. Nevertheless in the United States strenuous efforts are being made to devise an evaluated nuclear data format (ENDF) based on proposals made by Honeck [6] at BNL. Facilities exist for translating into this format from that of the widely used UKAEA Nuclear Data Library Format [7] and there seems little doubt that in time an acceptable format will evolve both for data transmission and as input to processing programmes. It is unrealistic to expect laboratories to change codes which have taken years to develop except at a natural time such as the installation of a new computer. In spite of the absence of an international format much exchange of data on magnetic tape has occurred with the receiving laboratory writing suitable format translation programmes.

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3. DESIRABLE FEATURES OF NEUTRON CROSS-SECTION EVALUATIONS

Evaluations should be such that in the neutron energy range considered, whether small or large, the data are complete as far as the input to neutronics calculations is concerned. This implies, firstly, the reconciliation of conflicting data, secondly, the filling of gaps by theory and interpolation and, thidrly, the inclusion of sufficient data to enable the fate of a neutron in a single collision to be "determined" by sampling from various probability distributions. Evaluation is not an alternative to the making of good measurements. If the experimental data are in serious conflict the discrepancy should be underlined so that new and more careful measurements may be called for.

Evaluations should not be orientated to some particular method of calculation but should enable input parameters for all methods of calculation to be prepared readily. Thus diffusion theory calculations may only require the average cosine of the scattering angle in elastic scattering but other calculations may require the full angular distribution; the evaluation should therefore include the latter, the average cosine of the scattering angle being calculated as required or included as an optional extra.

It is useful to distinguish between evaluated data, as defined above, and processed data such as group cross-sections and collision probabilities which are often derived from evaluated data using suitable computer codes. Processed data are generally orientated towards particular methods of calculation such as multigroup diffusion theory or the Monte Carlo method. They are generally influenced by the particular needs of the originating laboratory; their origin is often obscure and their versatility limited in comparison with that of evaluated data. However it is not possible to make a hard and fast division between evaluated data and processed data. Many-group cross-sections can often be considered as good approximations to evaluated cross-sections and sometimes provide the most usable information available. (In this sense and only in this sense some reference is made to multigroup cross-sections in section 4.)

Convenience in use requires that the data of a particular evaluation are available altogether in one place, that the input to neutronics and other calculations can be readily obtained and that the justification for the choice of the data is available together with estimates of inaccuracies. With current techniques magnetic tape storage of data is well suited to many requirements. Data can be easily transmitted or processed and graphical and tabular displays are readily obtained.

Much exchange of evaluated data already takes place between laboratories and to a smaller but increasing extent through data centres such as Brookhaven and Saclay and should be encouraged particularly in view of the considerable labour involved in producing a first class evaluation over a wide energy range.

Adequate documentation of evaluation work is extremely desirable. There will always be an understandable reluctance to use data evaluated in other laboratories unless information on its general origin and accuracy is available. Quite recently two excellent journals have invited papers in this field – the Agency's Atomic Energy Review and the journal Nuclear Data and there now seems no reason why neutron cross-section evaluation

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work should not be routinely published in the same way as experimental cross-section measurements.

4. EVALUATIONS OF NEUTRON CROSS-SECTIONS AVAILABLE ON 1 JUNE 1966

In continuation and extension of similar tables given in previous reports by one of the authors [8, 9], a Table^{*} has been prepared from a computer printout, which attempts to summarize the neutron crosssection evaluations available on 1 June 1966. In constructing a table of this kind compromises are inevitable. The prospective user of evaluated data wants to ascertain quickly the main features of what is available. For this purpose the entries in CINDA are felt to be both too brief and too scattered. The Table has been constructed according to many of the principles of CINDA but allowing for the inclusion of additional information and of more comments than are normally made with CINDA entries. As a sample, the part of the Table on ²³⁸U is shown in Table I of this paper.

A brief discussion of certain features of the Table is in order.

The date of evaluation is given where known. This essentially means that the literature is fairly thoroughly surveyed to this date (a few later references may be included) which is often many months earlier than the date of any relevant report.

Data media indicate how the data are available. C means computer media – punched cards or magnetic tape – G graphs and T tables. Computer media, in particular, are the easiest way of transferring large blocks of data between laboratories. In these cases it often happens that the data are available on request from one or more of the international neutron data centres and appropriate symbols in the Data Bank columns show the position. BLS indicates availability of the data from ENDF/A bank at Brookhaven National Laboratory (B), the Lawrence Radiation Laboratory, Livermore (L) and the CCDN at Saclay (S). Otherwise requests for data should be sent to the originating laboratory.

The columns "reactions considered" and "data type" are intended to give the reader an idea of the extent of the data and should indicate whether a particular evaluation is sufficiently complete for a particular application. The scheme of identifying reactions is based on that used in the UKAEA Nuclear Data Library [7,12] and gives good detail in a small space.

The entries for the Table were prepared on punched cards and expanded by means of a simple programme similar to that used in preparing CINDA [1]. The formats and programme are deposited with the ENEA Neutron Data Compilation Centre at Saclay for further use and development.

An attempt has been made to limit entries in the Table to reports which are fairly freely available. Ideally the user would like to have an

^{*} This 77-page table, together with a list of references, is not included in these proceedings because of space limitations. It is available on request from the Nuclear Data Unit, IAEA, Kärntnerring 11-13, A-1010 Vienna, Austria. Revised versions of the table will be issued from time to time by the ENEA Neutron Data Compilation Centre, Saclay, Gif-sur-Yvette, Essonne, France, and/or other similar centres such as the Brookhaven Neutron Cross Section Evaluation Center, Brookhaven National Laboratory, Upton, Long Island, N.Y. 11973, USA.

MA S	T ER I AL	ENERGY MIN MAX	EVAL DATE	LAÐ	REFERENCE DATA DATA REACTIONS CONSIDERED Source date media Jama	DA TA Type
U	238	25-2 42 2	157	LAS	LA-2144 658 GT 1.2;102,RRP CROSS SECTIONS AT 14 TEMPERATURES IN RAMGE 0.0253-500EV BY DOPPLER BRUADENING OF Single Level Baett-Migner Analysis.	C
U	238	25-2 10 7		USA	NP-8216 058 TG 1,2,4,16,18,10Z CUMULATIVE ELASTIC ANGULAR DISTRIBUTION FOR MONTE CARLO CALCULATIONS.	8
U	230	50 5 15 7	58	LRL	UCRL-5351 N58 G 1.2.3.4.16.17.18.20.21	C .
U	238	50-3 25 0	561	GA	GA2113 661 GT 102	C.
U	Z38	41-1 10 7		GA	GA2451 VOL1 BO1 CT 2:4,16,18,27,NU,9RP,5RP,FS Ruarter Lethargy Group Cross Sections 0.416(V-10MeV, Poppi Scattering Matrices, Data for 5, resolved Resonametes. Interstic and 10,7%) transfer Matrices, Gami Data.	•
U	238	10-3 14 7		AL	NAA-SR-MEMB904 863 CGT 1,2,4,16,18,102,NU Gap in Range 564-2,4469, Maa-SR-Memd 645 Gives data to 14Mev and Elastic Scattering Anguar Distributions for Mont Carld Data. Supersedes Maa-SR-Memd 5861.	E
U	238	10-4 15 7	363	WIN	AEEW R351 264 CGT BLS 1.2.3.5.6.7.8.9.10.15.16.17.18.102 Data as ampe d-79/63 in Range 1.75kev-15nev. DFN5 of Ukaea Nuclear Data Library.	A
U	238	37-2 18 7		UNC	UNC-5099 D64 CT 1,2,5,6,7,8,9,15,16+17,18,102,NU.FS,	Ð
					ELASTIC LEGENDRE COEFFICIENTS TO PIG.	
U	238	10-9 15 7	N64	GA	GA-6087 165 CT 1,2,3,4,5,6,7,8,9,10,11,12,13,14,15, 10,17,18,31,102,NU,RRP ND PDINT VALUES OF CROSS SECTIONS BETWEEN 2,3850 AND IGKEV,AWRE 0-79/63 DATA USED FOR FISSION ABOVE 2REV,TOTAL ABOVE 10XEV AND 14,2NI AND 14,3NI CROSS SECTIONS. ELASTIC LEGENDRE COEFFICIENTS TO P12.	Ð
U	238	19 0 10 3		HAN	NANL-THE-1228 865 CT 2,4,16,18,27,NU,RRP,SRP Revision of Gam Library data in resonance region, only limited tables.	E
υ	238	0 0 10 7		BNW	BNWL-CC325VOL3 965 CT 2+15+16+10+102+NU+RRP Elastic Legendre Coefficients to P4- parameters for 55 resonances. Tho different Sets of Data Given.	Ð
U	238	THR 10 7	765	KFK	KFK 120/[D65 GT L.2.3.4.5.6.7.8.9.16.18.102.NU.RRP. SRP.MU	C
					J.J. SCHMIDT. TABLES IN KFK120/II 2ND ED. WHICH SUPERSEDES KFK120/II 1ST FD.BELOW 1 EV. CROSS SECTIONS IN RESOLVED REGION CALCULATED FROM RESONANCE PARAMETERS.	
U	238	25-2 10 3	265	ÓRL	ORNL-TH-1448 666 GT 1,2.102 CROSS-SECTIONS FOR 300 DEGREES K GENERATED FROM BNL325SUPPLEMENT2,VOLUME3 RESONANCE Praneters, 802 Energy Points.	C
U	238	36 5 15 7	666	KAP	C.R. LUBITZ+ 766 CT 18 Includes white data.	C
U	238	07	66	ORL	GIFT+MIMALCZO 66 1,2,4,18,102,NU Elastic legendre cdefficients to P14. Most reliable above lokev. Entry incomplete.	
U	Z3 B	'THR 15 7	466	FR	J.RAVIER + CGT 1,2,3,4,5,6,7,8,9,10,11,12,13,14,15, 16,17,18,102,MU,F5,RRP AVAILABLE AUTURN 1966 AS DFM-31 IN UKAEA MUCLERA DATA LIBRARY FORMAT.REVISES AMRE 6-79/63 AGUYE IKEV.MULTILEVEL BREIT-WIGNER FORMULA WITH DOPPLER BROADENING FOR DATA UP TO AGY.	•

indication of the best data available for a particular material but it is an almost impossible task for any reviewer to do this in an unbiased and comprehensive manner. Indeed, to assess several data sets for a particular nuclide it is really necessary to make a further evaluation. Most workers active in the evaluation field and most users will have their own definite views as to which data are the best for particular purposes. It is appropriate to point out that certain data referenced in previous reports [7, 8] are now definitely obsolete and are omitted from the Table.

It is appropriate to emphasize again the particular sense in which evaluation is used in this paper (see section 1.2). The lack of clear-cut divisions between compiled and evaluated data and between evaluated data and processed data (group cross-sections, etc.) causes difficulties. The work of Liskien and Paulsen [10] at BCMN (EURATOM), Geel is probably well known to most evaluators but currently no best curves are drawn through the many plots of data on (n, p), (n, α) and (n, 2n) cross-sections and hence there is no entry in the Table. Much Canadian and Russian work on the application of neutron cross-sections has led to some form of group cross-sections. Although best values of cross-sections may have

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been obtained during intermediate working those have not generally been published. This particularly applied to the Russian 26-group constant set where Abagyan et al. [11] discuss the derivation of evaluated crosssections in some detail without giving the unaveraged cross-sections.

In spite of these and other difficulties it is the hope of the authors that the Table of available evaluations will prove a valuable guide to the neutron cross-section evaluation work previously and presently undertaken and assist in the establishment of contacts. The main purpose of wide publicity in this respect is to encourage still further inter-laboratory and international co-operation.

Whilst every effort has been made to ensure the accuracy and completeness of the Table, both by studying papers and reports and by correspondence with laboratories and individuals concerned, there will quite likely be omissions and mistakes. Since it is intended to keep the Table up-to-date and revisions will be made, the authors will be very glad to be told of such omissions and mistakes so that future tables approach more closely to perfection.

5. CURRENT WORK AND THE FUTURE

Probably all laboratories active in the evaluation field now realize the task facing them and as a result there is a trend towards individual laboratories producing just a few good evaluations and obtaining other required information on an exchange basis. Thus the CEA establishment at Cadarache has taken a responsibility for nickel and chromium data but does not propose to spend a great effort on any other evaluation work; other requirements are met by data from the UKAEA Nuclear Data Library and elsewhere.

In the neutron cross-section evaluation field it seems desirable and possible to develop international co-operation along the lines which have been followed in the neutron cross-section measuring field, where it seems that the co-operation among the OECD countries, coordinated by the European-American Nuclear Data Committee, can be extended under the aegis of the newly established International Nuclear Data Committee to include effectively the whole world. Already the EANDC and the European-American Committee for Reactor Physics (EACRP) have set up a Joint Sub-Committee on Nuclear Data Evaluation. The co-operative venture being organized by the United States Cross Section Evaluation Working Group is described in these Proceeding [26].

The existence of request lists for new measurements has helped the measuring laboratories to plan their work sensibly and avoid undue duplication. There is every reason to draw up request lists also for evaluations and thereby offer similar guidance to the community of evaluators; this kind of activity has in fact been initiated.

In the compilation field, work is being increasingly concentrated at a few regional centres – Brookhaven, Obninsk, Saclay and Vienna. Compiled data is the starting material for evaluation work and it might be thought that the regional centres should eventually undertake evaluation. However it seems neither wise nor likely that this should happen on any large scale. Most scientists employed on evaluation work have other responsibilities in their own laboratories, particularly in answering a

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large miscellany of queries on various aspects of neutron cross-sections in relation to particular calculations and projects. Evaluation work is much more easily split down into small parts than is the case with compilation work where concentration in a few centres has many advantages. However the regional centres should play an increasing role in promoting the exchange of evaluations by acting as clearing houses. They are better equipped for this function than the often overworked individual evaluators.

The techniques of evaluation have not been referred to in this paper as they are the subject of a separate review by Schmidt [13]. It is in order here to mention the increasing interest in applying computer techniques to the analysis of the data now being collected at the regional centres. Briefly the argument may be put that if existing evaluation techniques (drawing "smooth" curves, etc.) are logical operations they can be analysed and programmed for computers. This type of development is likely to become increasingly important over the next few years.

An aspect of evaluation which requires increased attention is the analysis of a particular type of data for many nuclides rather than the analysis of all types of data for a particular nuclide. A particularly difficult area, at least until very recently, has been capture cross-sections in the keV energy region. An evaluator studying a particular nuclide has been faced with great difficulties regarding the best values of standard cross-sections such as those of gold and iodine. Studies have been made of large numbers of (n, 2n) cross-sections and of (n, p) and (n, α) crosssections at 14 MeV. This type of evaluation is a valuable complement to the evaluation of all reactions for one nuclide and should be encouraged. It is also important to encourage co-operation between evaluators and experts in particular methods of calculating cross-sections. For an evaluation covering the range 0.0001 eV-20 MeV many such methods may be profitably used and it is generally impossible for an evaluator to be really expert in all. The computer programme libraries such as those at Argonne and Ispra have a part to play in making suitable calculations a pratical possibility.

A survey of present evaluation activities, insofar as they are known to the authors, is given below. Where laboratories and individuals are mentioned it is with the object of promoting contacts and co-operation between those with similar interests.

5.1. Australia

An evaluated neutron cross-section library is maintained at the AAEC Establishment at Lucas Heights [14]. Much of the data is drawn from the UKAEA Nuclear Data Library. Evaluation work may well expand in the future; enquiries may be addressed to J.L. Symonds.

5.2. Canada

The Canadian atomic energy programme has so far been orientated towards well-moderated thermal reactors and this has stimulated the well known work of Westcott and co-workers on effective cross-section values for well-moderated thermal reactor spectra. However this work and that of Walker on effective cross-sections for fission products falls outside the scope of the present review.

5.3. EURATOM

The work of Liskien and Paulsen at CBNM, Geel [15] on (n, p) (n, α) and (n, 2n) reactions is essentially a type of compilation but there is a possibility that future issues will contain best curves.

A compilation and evaluation of data related to nuclear energy standards has been started by K. Gubernator, also at Geel. Work is well advanced on boron-10 cross-sections from thermal to 1 MeV, including particularly the (n, α) cross-section and its branching ratio. The fission cross-section of uranium-235 will be considered next.

5.4. European Nuclear Energy Agency

Although the ENEA's Neutron Data Compilation Centre at Saclay is not concerned with evaluation as such it is expected to play a part in disseminating evaluations and in particular to continue the informative work of the present paper.

5.5. France

The Table gives full details of evaluations of nickel and chromium cross-sections made by J.Ravier of Cadarache; later work was carried out in conjunction with M. Vastel of Electricité de France, Chatou. During recent months these two evaluators have been revising the current UKAEA Nuclear Data File for uranium-238, details of which are given in the Table.

5.6. Federal Republic of Germany

At the Kernforschungszentrum Karlsruhe J.J. Schmidt has made evaluations of cross-sections for several years; parts II (Tables) and III (Graphs) of Ref.[16] are well known. Recently several sections of part I (Theory and Basis for Evaluated Data) have been prepared and some revisions have been made to Part II. The description of the methods used and the detailed derivation is the best we know and provides an excellent example for other evaluators to follow. A magnetic tape nuclear data library is maintained at Karlsruhe.

5.7. International Atomic Energy Agency

As Head of the IAEA Nuclear Data Unit, C.H. Westcott¹ organized the most comprehensive evaluation of 2200 m/s parameters for fissile uranium and plutonium isotopes yet made [17].

5.8. India

The Atomic Energy Establishment Trombay has indicated its interest in the neutron cross-section evaluation field, but up-to-date information on plans and activities is not to hand.

¹ Dr. C.H. Westcott is now at the Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.

5.9. Israel

Israeli interest in cross-section evaluation is shown in the paper by Pazy et al. presented at this Conference [18].

5.10. Italy

At the Centro di Calcolo of the CNEN, Bologna, V. Banzi and coworkers have been active in the evaluation field since 1960 and have developed a number of computer programmes for associated optical and statistical model calculations. During recent months Benzi has prepared an evaluation of the cross-sections of copper which is available in UKAEA Nuclear Data File format.

5.11. Japan

At JAERI, Tokai-mura, Dr.T. Momota of the Japanese Nuclear Data Committee can be addressed for enquiries on cross-section evaluation activities.

5.12. Sweden

Evaluation work has been undertaken at both the Aktiebolaget (AB) Atomenergi and FOA, the Research Institute for National Defence; the Table contains appropriate information. Considerable use has been made of data from the Karlsruhe, United Nuclear Corporation and UKAEA Nuclear Data Libraries. H. Häggblom of the AB Atomenergi and M. Leimdörfer of FAO may be consulted.

5.13. Union of Soviet Socialist Republics

To an outside observer the approach to cross-section evaluation in the USSR appears rather different from that followed in the United States and Western Europe. In the well-known book "Group Constants for Nuclear Reactor Calculations" [11] the most comprehensive set of group crosssections yet prepared is given. It seems clear from the text that during the preparation of this most useful set many cross-section evaluations must have been made, although the preferred values of the basic differential cross-sections do not seem to have been published. The nearest approach to evaluation reports of the type listed in the Table seems to be the various editions of the "Handbook on Nuclear Physics Constants for Reactor Calculations" by I. V. Gordeev et al. [19]. More recently several bulletins of the Information Centre on Nuclear Data containing much work of an evaluation nature have been published. These bulletins are available in English translation from the IAEA Nuclear Data Unit.

5.14. United Kingdom

The UKAEA Nuclear Data Library was designed from its inception in 1961 for computer use (an earlier system developed at AWRE had given several years previous experience) and many of the processing programmes such as GALAXY and DICE have been operational for several

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years. At the 1964 Geneva Conference a general account of the data system was given [20] which also includes an extensive bibliography of reports giving detailed descriptions of the Library format, of the associated computer programmes and, of course, the data in use at that time. Later references may be found in a short paper presented at the Brookhaven evaluation seminar in May 1965 [21]. Much of the data and many of the programmes have been widely used in Europe, the United States and Australia and many of the ideas were adopted in the ENDF concept described by Chernick. Evaluation groups work at Winfrith (J.S. Story), Aldermaston (K. Parker) and Risley (W. Hart). The Table gives a clear picture of past activities. At present, evaluations aimed at improving data on deuterium, boron-10, boron-11, resonance reactions on chromium and nickel, manganese, iron, zirconium, tantalum, uranium-235 and plutonium-239 are in various stages of completion. The United Kingdom has been very aware of the need for international cooperation in the evaluated neutron cross-section field and has played a major role in stimulating such co-operation.

5.15. United States

A comprehensive picture of the United States effort in evaluation requires the consideration of contributions from many laboratories. As space is limited we must refer to the Table, and confine this paper to information not readily presented there. The Proceedings of the Conference on Neutron Cross Section Technology [22] give a recent and more detailed account of the American scene.

(a) Argonne National Laboratory, Argonne, Illinois

Published work from ANL is mainly in the form of group crosssection sets but in Ref. [23] will appear documentation of the data for 30 nuclides used in a 26-group cross-section set. Davey has made an evaluation of fast fission cross-sections. Moldauer has predicted many cross-sections particularly using the computer code NEAREX [24].

(b) Atomics International, Canoga Park, Calif.

As in the UK the Atomics International evaluation effort under H.Alter is highly computer-oriented and has made considerable use of programmes such as 2 PLUS (non-spherical optical model) and a modification of NEAREX. An analysis of resonance parameters and fission and capture cross-sections for uranium-233, uranium-235 and plutonium-239 in the resonance region will be published [25]. Mechanized evaluation of crosssections is also under study.

(c) Bettis Atomic Power Laboratory, Pa.

D.R. Harris and co-workers have devoted most effort to multi-group libraries, but unpublished evaluations exist below 1 eV for several elements and for hydrogen and oxgyen over a wide energy range.

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(d) Brookhaven National Laboratory, Brookhaven, N.Y.

Work at this Laboratory is described in J. Chernick's paper presented at this Conference [26]. Much of the laboratory's evaluation effort has been concerned with specific problems — optical model calculations, analysis of (n, 2n) cross-sections and statistical analysis of resonance parameters — rather than with comprehensive evaluations of the type included in the Table.

(e) General Atomic Division, General Dynamics Corporation, San Diego, Calif.

The General Atomic Library consists of sets of evaluated crosssections for almost all elements and includes detailed isotopic values for a large number of the elements (see the Table). M.K. Drake and D. Matthews are at present working on evaluations for sodium, chlorine, potassium, manganese and hafnium. Research on scattering law data is being carried out under James Young.

(f) General Dynamics, Fort Worth, Texas

Recent work concerns (n, 2n) cross-sections (by H.G. Carter and J.R. Williams) and elastic and inelastic scattering in neptunium, vanadium, carbon (by Weston et al.) [27].

(g) General Electric, Atomic Products Division, San Jose, Calif.

Greebler, Hutchins and Aline are preparing plutonium-239 data for ENDF/B.

(h) General Electric, Nuclear Materials and Propulsion Operation, Cincinatti, Ohio

After completing work on tantalum, tungsten isotopes, plutonium-238 and curium-244, A. Prince has now taken up a post at the Brookhaven Cross Section Evaluation Center.

(i) Knolls Atomic Power Laboratory, Schenectady, N.Y.

The position is similar to that at Bettis in that many data are available only in multigroup form. Specific information may be obtained from C.R. Lubitz. Data from Ref. [28] on oxygen-16 has been omitted from the references in the Table (but not from the oxygen entries).

(j) Lawrence Radiation Laboratory, Livermore, Calif.

The publication of R.J. Howerton's group is well known but full detail of the latest evaluated cross-section library covering 50 materials has not been published.

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(k) Los Alamos Scientific Laboratory, Los Alamos, New Mexico

Dr. Leona Stewart is working on evaluation of deuterium and helium cross-sections to parallel the report on tritium [29].

(1) Oak Ridge National Laboratory, Oak Ridge, Tenn.

The composition of multigroup libraries for reactor physics and shielding programmes at Oak Ridge are, in general, unpublished. Information on the two programmes can be obtained from C.W. Craven and S.K. Penny, respectively.

(m) Pacific Northwest Laboratory, Hanford, Conn.

The comprehensive BNWL-CC-325 (3 Volumes) covering 406 isotopes is surveyed in the Table.

(n) United Nuclear Corporation, Washington, D.C.

United Nuclear Corporation is a major contributor to existing evaluations. Available work from E.S. Troubetzkoy's group is described in the Table.

(o) Westinghouse Electric Corporation, Atomic Power Division, Pittsburgh, Pa.

N. Azziz and R. Dannels are completing evaluations for helium-4, iron and nickel, and are putting considerable effort into the ENDF/B project.

(p) Westinghouse Electric Corporation, Astronuclear Laboratory, Pittsburgh, Pa.

Recent work by Drawbaugh and Gibson on uranium-235 resonance cross-sections is described in a paper presented at this Conference [30]. Other evaluation work is covered in the Table.

Space does not permit the description of smaller efforts at other United States Laboratories and the contribution of several specialist information centres.

An increasing amount of money and effort is being devoted to making measurements of neutron cross-sections for use in the development of nuclear energy and it is important that more and better neutron crosssection evaluations become available so that the data obtained are put to maximum use. Indeed the carrying out of a full evaluation exercise together with the supporting neutronics calculations provides the only real criterion for deciding whether existing cross-section measurements meet requirements or whether further experimental (and theoretical) work is needed. Whilst the ideal state of having evaluated data on all natural elements and separate isotopes of many elements together with low energy data on the more important compounds is still some time away, there is clear evidence of an increasingly organized effort towards this desirable goal.

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We should like to thank the many laboratories and individuals who have responded to several appeals made through the ENEA and USAEC for information on their work in the evaluation field. This has proved invaluable in preparing the Table.

Mr.J.A. Price and Mrs.S.M. Offord of Atomic Weapons Research Establishment, Aldermaston are responsible for much of the detailed planning and programming relating to the Table.

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DISCUSSION

D.K. BUTLER: In most cross-section requests made by those engaged in fast-reactor calculations the precisions specified for capture and fission cross-sections are far better than can be obtained with current experimental techniques. Those making such requests do so in the hope that the precisions they require for calculating the properties of fast systems will one day be attainable. What do you consider to be the accuracy of present capture cross-section evaluations for materials such as ²³⁸U and iron?

K. PARKER: The best way of estimating the accuracy with which specific capture cross-sections are known is to get a corporate view from all the experimenters involved. An evaluator who has to provide many cross-sections (capture, fission, scattering, etc.) cannot generally do this. However, the perturbation analysis of integral measurements developed at Aldermaston, Bologna, Jerusalem and Obninsk (see papers CN-23/15 and CN-23/94) does offer a useful tool in indicating which of a number of conflicting experimental values is most likely to be correct.

J. CHERNICK: You appear to have omitted from your Table a considerable amount of valuable data that does not satisfy your criteria of completeness. For example, Davey's work at Argonne National Laboratory and the work carried out at General Atomic on thermal scattering kernels have been excluded because they are concerned with narrow energy ranges or particular cross-sections rather than with complete energy ranges and all cross-sections. The individual evaluator, however, should be aware of such work.

K. PARKER: The Table is intended for use by those who have to perform reactor calculations. However good a particular limited evaluation, such as that of Davey on fission cross-sections, it must be combined with other data (total, capture and scattering cross-sections, etc.) before it can be used in a reactor calculation. Naturally, evaluators preparing files of the type described in the Table are aware of, and make full use of, the more specialized, fragmentary evaluations. The Table is intended to help reactor designers who may not be knowledgeable in nuclear physics or evaluation.

INTEGRAL MEASUREMENTS AS SUPPLEMENTARY DATA IN NEUTRON CROSS-SECTION EVALUATION

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Abstract

INTEGRAL MEASUREMENTS AS SUPPLEMENTARY DATA IN NEUTRON CROSS-SECTION EVALUATION. The purpose of this paper is the exact formulation of a method to improve the microscopic cross-section evaluation by means of integral data. Considering a function $\sigma(E)$ and a set of functionals of this function $\gamma_1[\sigma]$, we propose that the function $\bar{\sigma}(E)$, which minimizes the quadratic form

$$\int \left(\frac{\sigma - \hat{\sigma}}{\Delta \hat{\sigma}}\right)^2 \omega \, dE + \sum_{i} \left(\frac{\gamma_i [\sigma] - \hat{\gamma}_i}{\Delta \hat{\gamma}_i}\right)^2$$

is the best estimate of $\sigma(E)$. $\hat{\sigma}$ is the estimate obtained from the analysis of direct measurements of σ , $\Delta\hat{\sigma}$ is the standard deviation of a single measurement, and $\Delta\hat{\gamma}_i$ is the significance attributed to the estimate in the interval dE. The $\hat{\gamma}_i$ are the experimental values of $\gamma_i[\sigma]$, and $\Delta\hat{\gamma}_i$ are their standard deviations.

Explicit expressions for $\overline{\sigma}$ and $\Delta\overline{\sigma}$ are derived, and various special cases and applications are discussed. To illustrate the method, data from a few simple systems are used to correct the appropriate cross-sections.

1. INTRODUCTION

Measurements of critical masses and flux distributions in critical and subcritical assemblies may be considered as indirect, "integral" measurements of the "elementary" nuclear parameters: cross-sections, number and spectra of fission neutrons, etc. So far, the information obtained by these integral measurements was hardly used in the evaluation of the elementary nuclear parameters. The purpose of this paper is the exact formulation of a method to improve the microscopic cross-section evaluation by means of such integral data.

To use data from discrete measurements, such as critical mass determinations, along with data from direct measurements of nuclear cross-sections, which are continuous functions of the energy, the method of least squares is generalized. This generalization is applied in sections 2, 3 and 4 to improve the microscopic cross-section evaluation by means of integral data. In section 5 the method is illustrated by means of a simple numerical example. In the concluding section some remarks on the practical application of the method are made.

2. STATEMENT OF THE PROBLEM

Let us consider a set of physical quantities $\sigma_i\left(E\right)$, the experimental estimates of which are

$$\hat{\sigma}_{i}(E) = \frac{\Delta \sigma_{i}(E)}{\sqrt{\omega_{i}(E)\Delta E}}$$
(2.1)

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where $\omega_i(E)$ is the number of measurements of $\sigma_i(E)$ per unit interval of E, and each single measurement has an experimental error $\Delta \hat{\sigma}_i(E)$. Circumflexes over symbols denote experimental results. Further, let us consider another set of quantities γ_j , which are functionals of the functions $\sigma_i(E)$,

$$\gamma_{i} = \gamma_{i}[\sigma_{1}(\mathbf{E}), \sigma_{2}(\mathbf{E}), \ldots] = \gamma_{i}[\sigma(\mathbf{E})] \qquad (2.2)$$

and assume experimental values of these also to be available, so that

$$\hat{\gamma}_{j} \hat{\gamma}_{j} \pm \hat{\Delta \gamma}_{j}$$
 (2.3)

We assume that the experimental estimates of the functions $\sigma_i(E)$ and the functionals $\gamma_j[\sigma]$ have normal distributions, the former with means $\sigma_i(E)$ (the true, unknown functions) and standard deviations $\Delta \hat{\sigma}_i (\omega_i(E)\Delta E)^{-1/2}$, and the latter with means $\gamma_i[\sigma]$ and standard deviations $\Delta \hat{\gamma}_i$.

Using this assumption, the probability of simultaneously obtaining the experimental estimates $\hat{\sigma}_i$ and $\hat{\gamma}_i$ is

$$P = C \exp \left\{-\sum_{i=1}^{1} \left[\frac{\sigma_{i}(E) - \hat{\sigma_{i}}(E)}{\Delta \hat{\sigma}_{i}(E)}\right]^{2} \omega_{i}(E) dE - \sum_{j=1}^{1} \left[\frac{\gamma_{j}[\sigma] - \hat{\gamma}_{j}}{\Delta \hat{\gamma}_{j}}\right]^{2}\right\}$$
(2.4)

The improved estimate of the functions considered consists of those functions $\sigma_i(E)$, for which the probability P is maximal. This is the fundamental proposition of the least-squares principle. Thus, the functions $\sigma_i(E)$ for which we are looking, should minimize the quadratic form

$$Q = \sum_{i} \int \left[\frac{\sigma_{i}(E) - \hat{\sigma}_{i}(E)}{\Delta \hat{\sigma}_{i}(E)} \right]^{2} \omega_{i}(E) dE + \sum_{j} \left[\frac{\gamma_{j}[\sigma] - \hat{\gamma}_{j}}{\Delta \hat{\gamma}_{j}} \right]^{2}$$
(2.5)

The functions $\omega_i(E)$ were introduced as the densities of the differential measurements of the $\sigma_i(E)$. It is now clear that they may be interpreted more generally as the over-all significance, the relative weights, attributed to the estimates $\hat{\sigma}_i(E)$. These weights are not necessarily just the densities of measurements; other considerations might be invoked in assessing the experimental estimates. If, for example, a given cross-section is constant within a certain energy interval, or of a known functional form, and is well measured around one energy value, then its estimated values at other energies should be just as reliable, even though there might be no experimental results available for these other energies.

Therefore, we shall henceforth consider the functions ω_i (E) to be the relative weights of the experimental estimates $\hat{\sigma}_i$ (E), as determined by all pertinent information.

3. FORMAL SOLUTION OF THE GENERAL PROBLEM

We shall treat the problem of improving the direct estimate of one function, using experimental data on N of its functionals, and then demonstrate that the general problem, with several functions, can always be reduced to this case.

We are thus seeking that function $\sigma(E),$ which minimizes the quadratic form

$$Q = \int \left[\frac{\sigma(E) - \hat{\sigma}(E)}{\Delta \hat{\sigma}(E)}\right]^2 \omega(E) dE + \sum_{i=1}^{N} \left[\frac{\gamma_i [\sigma] - \gamma_i}{\Delta \hat{\gamma}_i}\right]^2$$
(3.1)

It will be convenient to choose the square root of the integrand in Eq. (3, 1)

$$f(E) = \frac{\sigma(E) - \sigma(E)}{\Delta \sigma(E)} \sqrt{\omega(E)}$$
(3.2)

as our unknown function. We shall also introduce the functional derivatives $D_i(E)$ of the functionals $\gamma_i[\sigma]$. These derivatives are defined, as usual, by the relations

$$\delta \gamma_{i} = \int D_{i}(E) \, \delta \sigma(E) \, dE \tag{3.3}$$

Then, by Eqs. (3.2) and (3.3),

$$\gamma_{i}[\sigma] = \gamma_{i}[\hat{\sigma}] + \int D_{i}(E) \frac{\Delta \sigma(E)}{\sqrt{\omega(E)}} f(E) dE \qquad (3.4)$$

and the quadratic form (3.1) may now be expressed as

$$Q = \int f^{2}(E) dE + \sum_{i=1}^{N} [JG_{i}(E) f(E) dE - C_{i}]^{2}$$
(3.5)

where

$$G_{i}(E) = \frac{D_{i}(E)}{\Delta \hat{\gamma}_{i}} \frac{\Delta \hat{\sigma}(E)}{\sqrt{\omega(E)}}$$

$$C_{i} = \frac{\hat{\gamma}_{i} - \gamma_{i}[\hat{\sigma}]}{\Delta \hat{\gamma}_{i}}$$
(3.6)

The variation of Q with respect to f(E) is

$$\delta Q = 2 \int \{f(E) + \sum_{i=1}^{N} [\int G_{i}(E') f(E') dE' - C_{i}] G_{i}(E) \} \delta f(E) dE \qquad (3.7)$$

For that f(E) which minimizes Q, this variation vanishes for any arbitrary $\delta f(E)$. The function we are looking for is therefore the solution of the integral equation

$$f(E) + \int \sum_{i=1}^{N} G_{i}(E) G_{i}(E') f(E') dE' = \sum_{i=1}^{N} C_{i} G_{i}(E)$$
(3.8)

This is a symmetric Fredholm equation with a degenerate kernel. As long as +1 is not an eigenvalue of the homogeneous equation

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$$f(E) + \lambda \int_{i=1}^{N} G_{i}(E) G_{i}(E') f(E') dE' = 0 \qquad (3.9)$$

and this is indeed the case in all practical calculations, Eq.(3.8) can be solved by the usual procedure.

To solve Eq. (3.8) we first define

$$\begin{array}{c} \mathbf{x_{i}} = C_{i} - \int G_{i}(E) f(E) dE \\ \mathbf{A_{ij}} = \int G_{i}(E) G_{j}(E) dE \end{array}$$

$$(3.10)$$

With these notations Eq. (3.8) can be rewritten as

$$\mathbf{f}(\mathbf{E}) = \sum_{j=1}^{N} \mathbf{x}_{j} \mathbf{G}_{j}(\mathbf{E})$$
(3.11)

which, on multiplying by $G_i(E)$, and integrating over E, becomes

$$\mathbf{x}_{i} + \sum_{j=1}^{N} \mathbf{A}_{ij} \mathbf{x}_{j} = C_{i}$$
(3.12)

The solution of the integral Eq. (3.8) thus reduces to that of a system of N linear equation. By solving Eq. (3.12) for the x_i , and substituting the result in Eq. (3.11), we obtain

$$f(E) = \sum_{i,j=1}^{N} G_{i}(E) (B^{-1})_{ij} C_{j}$$
(3.13)

where the matrix B is

$$B_{ij} = \delta_{ij} + A_{ij} \tag{3.14}$$

and the improved estimate of $\sigma(E)$, by definition (3.2), is

$$\overline{\sigma}(\mathbf{E}) = \hat{\sigma}(\mathbf{E}) + \frac{[\Delta \hat{\sigma}(\mathbf{E})]^2}{\omega(\mathbf{E})} \sum_{i,j=1}^{N} \frac{D_i(\mathbf{E})}{\Delta \hat{\gamma}_i} (\mathbf{B}^{-1})_{ij} C_j \qquad (3.15)$$

In actual applications, however, functionals may depend on several functions, rather than one. The integral quantities that we propose to employ, to improve direct neutron cross-sections estimates, are practically functionals of the fission, capture, inelastic scattering and elastic scattering cross-sections, and the average number $\bar{\nu}(E)$ of neutrons emitted per fission induced by a neutron of energy E. There are actually as many sets of these functions as the number of isotopes of which the system under consideration is composed.

We shall refer to all these functions as cross-sections and denote them by $\sigma_{\rm m}^{\rm i}({\rm E})$, where j=1, 2, ..., 5 are the types of the functions, in their order above, and m=1, 2, ..., M indicate the various isotopes composing the system, M being their total number.

The effective energy range of all cross-sections (in the context of reactor physics) is finite, $0 \le E \le E_{max}$ (~10 MeV). We may therefore

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combine the 5M cross-sections, and construct a generalized single cross-section for each system, defined over the energy range $0 \le E \le 5ME_{max}$ by

$$\sigma(\mathbf{E}) = \sigma_{\mathbf{m}}^{j}(\mathbf{E} - (5\mathbf{m} + j - 6)\mathbf{E}_{\max})...(5\mathbf{m} + j - 6)\mathbf{E}_{\max} \leq \mathbf{E} < (5\mathbf{m} + j - 5)\mathbf{E}_{\max} (3.16)$$

Our problem in the general case of functionals depending on several functions, thus reduces to the case of functionals of just one function.

The definition (3.16) may now be used to express our results in terms of the separate components $\sigma_m^{j}(E)$ of the generalized cross-section.

Introducing the obvious modifications in the definitions, i.e. $D_{im}^{1}(E)$ is defined by the relation

$$\int (\delta \gamma_{i})_{m}^{j} = \int D_{im}^{j}(E) \delta \sigma_{m}^{j}(E) dE \qquad (3.17)$$

and

$$\mathbf{G}_{im}^{j}(\mathbf{E}) = \frac{D_{im}^{j}(\mathbf{E})}{\sqrt{\omega_{m}^{j}(\mathbf{E})}} \frac{\Delta \hat{\sigma}_{m}^{j}(\mathbf{E})}{\Delta \hat{\gamma}_{i}}$$
(3.18)

we obtain

$$\tilde{\sigma}_{\mathbf{m}}^{\mathbf{j}}(\mathbf{E}) = \hat{\sigma}_{\mathbf{m}}^{\mathbf{i}}(\mathbf{E}) + \frac{\left[\Delta \hat{\sigma}_{\mathbf{m}}^{\mathbf{j}}(\mathbf{E})\right]^{2}}{\omega_{\mathbf{m}}^{\mathbf{j}}(\mathbf{E})} \sum_{\mathbf{i},\mathbf{k}=\mathbf{1}}^{\mathbf{N}} \frac{D_{\mathbf{i}\mathbf{m}}^{\mathbf{j}}(\mathbf{E})}{\Delta \hat{\gamma}_{\mathbf{i}}} (\mathbf{B}^{-1})_{\mathbf{i}\mathbf{k}} C_{\mathbf{k}}$$
(3.19)

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where now the matrix A is given by

$$A = \int_{ik}^{5} \sum_{j=1}^{M} G_{im}^{j}(E) G_{km}^{j}(E) dE \qquad (3.20)$$

and the matrix B is defined by Eq. (3.14) with this A.

Finally, some remarks concerning the parameters used in the above theory are perhaps worthwhile. To apply our theory we need experimental data on the directly-measured cross-sections and on the integral quantities, as well as means and methods to calculate the integral quantities and their functional derivatives.

The integral quantities and their functional derivatives can be calculated, in principle, within any desired accuracy. This is usually done by solving the transport equation numerically and using perturbation methods to obtain the functional derivatives.

As to the experimental data, the only important data missing is the cross-section error function $\Delta \hat{\sigma}(E) / \sqrt{\omega(E)}$, which plays an essential role in our theory. The only information that we do possess is some crude estimate of the relative errors of individual cross-section measurements.

This information is conveniently given in terms of the relative error function $\beta(\mathbf{E})$ defined by

$$\frac{\Delta \hat{\sigma} (E)}{\sqrt{\omega (E) \Delta E}} = \beta (E) \hat{\sigma} (E)$$
(3.21)

Obviously, according to what has just been mentioned with respect to the sort of information we have about the relative error function, $\beta(E)$ is actually a "step function", i.e. piecewise constant.

4. THE MULTIGROUP APPROXIMATION

As we have just mentioned, we need experimental data on the directly-measured cross-sections and the integral quantities, and means and methods to calculate the integral quantities and their functional derivatives.

The integral quantities and their functional derivatives are usually calculated in a multigroup approximation. It seems, therefore, worthwhile to formulate a corresponding multigroup approximation of the theory which was developed in the previous section.

The multigroup approximation is based on the assumption that the true cross-sections can be replaced by appropriate fictitious cross-sections, which are piecewise constant functions, i.e. constant within each energy group.

With such an approximation, it seems meaningless to correct the cross-section in greater detail. In other words, if the corrected cross-sections are given by

$$\overline{\sigma}_{i}(E) = [1+p_{i}(E)]\hat{\sigma}_{i}(E) \qquad (4.1)$$

then the functions $p_i(E)$ can be estimated significantly only as piecewise constant functions, constant within each energy group.

For simplicity let us consider in this section only the case of one cross-section. The generalization elaborated at the end of section 3 applies to the present case as well.

Let us therefore assume that we use the multigroup approximation with group boundaries defined by the energies:

$$0 = E_0 < E_1 < \dots < E_C = E_{max}$$
(4.2)

The function p(E) is constant within each energy group,

$$p(E) = p_{g} \quad \dots \quad E_{g-1} \leq E \leq E_{g} \quad (4.3)$$

and the quadratic form (3.1) to be minimalized is transformed to

$$Q = \sum_{g=1}^{G} f_g^2 + \sum_{i=1}^{m} \left[\frac{\gamma_i [\sigma] - \gamma_i}{\Delta \hat{\gamma}_i} \right]^2$$
(4.4)

with

$$f_{g} = \frac{\beta_{g}}{\beta_{g}}$$

$$\frac{1}{\beta_{g}^{2}} = \int_{E_{g-1}}^{E_{g}} \frac{(E) dE}{(\Delta \hat{\sigma}(E) / \hat{\sigma}(E))^{2}}$$
(4.5)

Defining

$$G_{ig} = \frac{\beta_{g}}{\Delta \hat{\gamma}_{i}} \int D_{i}(E) \hat{\sigma}(E) dE \qquad (4.6)$$

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we obtain the following set of linear equations

$$\mathbf{f}_{g} + \sum_{i=1g}^{N} \sum_{g=1}^{G} \mathbf{G}_{ig} \mathbf{G}_{ig}, \mathbf{f}_{g} = \sum_{i=1}^{N} \mathbf{C}_{i} \mathbf{G}_{ig}$$
(4.7)

This set of equations is the discrete analogue of Eq. (3.8), and its solution is obtained in the same way. Thus,

$$\overline{\sigma}(E) = (1+p_g) \hat{\sigma}(E) \qquad E_{g-1} \leq E \leq E_g \qquad (4.8)$$

where p_g is given by

$$p_{g} = \beta_{g_{i,j=1}}^{N} G_{ig}(B^{-1})_{ij}C_{j}$$
(4.9)

The constants C_i are defined by Eq. (3, 6), and B = I+A where

$$A_{ij} = \sum_{g=1}^{G} G_{ig} G_{jg}$$
(4.10)

It should be noted that the number G of energy groups used in Eqs. (4. 2) to (4.10), can be any number less than or equal to the number G' of energy groups used in the calculation of the integral quantities and their derivatives $\partial \gamma_i / \partial \sigma_g$. The quantities G_{ig} defined by Eq. (4.6) are actually also calculated by a multigroup approximation and are thus approximated by

$$G_{ig} \approx \frac{\hat{\sigma}_{g}\beta_{g}}{\Delta \hat{\gamma}_{i}} = \frac{\partial \gamma_{i}(\hat{\sigma}_{g})}{\partial \hat{\sigma}_{g}}$$
 (4.11)

here $\gamma_i\,(\sigma_g$) is a function (and not a functional) of a finite number of group constants $\hat{\sigma}_g$.

5. A NUMERICAL EXAMPLE

The purpose of the following numerical example is merely to show how the theory developed in the previous sections works. In this sample calculation we used only a small number of integral measurements and a very crude approximation of the important relative error function, $\beta(\mathbf{E})$.

All the integral data used in our sample consist of six critical mass measurements. The essential information on these data is summarized in Table I.

The differential data used in the following example are all taken from the cross-section compilation of Schmidt [3].

The critical parameter γ , which we used in our calculations, is defined by the transport equation

$$\begin{bmatrix} \vec{\Lambda} \cdot \nabla + \sum_{m,j} n_{m}(\vec{x}) \sigma_{m}^{j}(E) \end{bmatrix} \psi(\vec{x}, E, \vec{\Lambda}) =$$

$$= \frac{1}{\gamma} \int_{m} \sum_{j} n_{m}(\vec{x}) \sigma_{m}^{j}(E) T_{m}^{j}(E', \vec{\Lambda}'; E, \vec{\Lambda}) \psi(\vec{x}, E', \vec{\Lambda}') dE' d\vec{\Lambda}'$$
(5.1)

Name	Critical Mass (Kg U-235)	Density of Core Alloy (g /cm ³)	Enrichment of Core (%)	Thickness of Reflector (cm)	Density of Reflector (g /cm ³)	Ref.
Ul	48.8	18.75	93.9			1
U2	34.0	18.75	93.9	1.77	19.0	2
U3	24.9	18.75	93.9	4.47	19.0	2
U4	19.2	18.75	93.9	8.94	19.0	2
U5	18.55	18.75	93.9	9.96	19.0	2
U6	16.65	18.62	93.2	18.11	19.0	2

TABLE I.	CRITICAL ASSEMBLIES,	ENRICHED	URANIUM	CORE,
NATURAL-	URANIUM REFLECTOR			

with the condition that no neutrons enter the system from without. The summation over j ranges from 1 to 4 (see the definition in section 3), and that over m from 1 to M, the number of isotopes of which the system is composed. The $n_m(\vec{x})$ are number densities of the various isotopes, and T_m^j (E', $\vec{\Omega}$ '; E, $\vec{\Omega}$) are transfer kernels of neutrons with energy E' and direction of flight $\vec{\Omega}$, to neutrons of energy E and direction of flight $\vec{\Omega}$.

The critical parameter γ , and the functional derivatives of γ with respect to the cross-sections, were calculated by the S₈ method of Carlson [4], using a 16-group approximation. The initial values of γ , as well as the values of γ calculated after the correction of the crosssections are given in Table II.

The cross-sections in the present example were corrected using three energy intervals. The correctors (correcting-factors) p_g for ²³⁵U and ²³⁸U are given in Tables III and IV.

Name	Experimental Error: Δγ×1000 (estimated)	(γ-1)×1000 with initial cross-sections	(y-1)×1000 with corrected cross-sections
Ul	1.00	-10.135	0.208
U2	0.80	-6.805	0.200
U3	0.95	-3.494	1.124
U4	1.30	-3.391	0.246
U5	0.65	-3.579	0.002
U6	1.40	-4.854	-1.3

TABLE II. VALUES OF CALCULATED γ BEFORE AND AFTER THE CORRECTION OF THE CROSS-SECTIONS

Reaction	Energy in Me∀	β×100	p×100
	.606	3.5	1.340
Fission	1.606	3.5	.955
	14.000	3.5	1.256
	.606	12.5	-2.595
Capture	1.606	21.0	-1.035
	14.000	21.0	434
Trolactic	.606	17.5	2.009
Costioning	1.606	17.5	5.186
Scattering	14.000	17.5	5.865
	.606	8.5	3.445
Conttoning	1.606	12.5	4.434
Scattering	14.000	17.0	4.675
	.606	3.0	1.341
Nu	1.606	3.0	.831
	14.000	3.0	1.095

TABLE III. CORRECTORS FOR ²³⁵U

6. CONCLUDING REMARKS

As we have already mentioned at the end of section 3, the new estimate of the cross-section, which is obtained by our method, depends strongly on the energy dependence of the estimates of the error of the measured cross-sections, and on the density of cross-section measurements. While some estimates for the mean error can be found in cross-section compilations, information on the exact energy dependence of the mean error, and especially information on the density of measurements, does not seem to be easily available, Because of this lack of information, we have to be satisfied with a crude correction of the crosssections, usually in the form of correcting factors over large energy intervals. The physical significance of such corrections is somewhat doubtful.

Nevertheless, the present method is still of great practical importance, since it provides us with an algorithm for calculating crosssection sets, which are consistent with the direct measurements and the integral measurements on many critical systems, as was illustrated,

Reaction	Energy in MeV	β×100	p×100
	.606	3.5	.000
Fission	1.606	3.5	.020
·	14.000	3.5	.090
· · · · · · · · · · · · · · · · · · ·	.606	12.5	-10.269
Capture	1.606	21.0	-2.636
	14.000	21.0	419
Thelestic	.606	17.5	-4.308
Inelastic	1.606	17.5	-5.228
Scattering	14.000	17.5	-12.635
	.606	8.5	-2.589
Elastic	1.606	12.5	-7.855
Scattering	14.000	17.0	-3.388
	.606	3.0	.000
Nu	1,606	3.0	.023
	14.000	3.0	.180

TABLE IV. CORRECTORS FOR ²³⁸U

for example, in Table II. We assume that such a set will be consistent even with integral measurements which were not considered in the evaluation, but do not differ significantly from those integral measurements considered in the evaluation. This assumption is confirmed by some preliminary calculations which we have performed.

The cross-sections obtained by the least-squares fitting can thus be considered as universal effective cross-sections for reactor calculations, provided that one uses one and the same method for critical calculations in the least-squares evaluation of the effective cross-sections and in the calculation of further reactor systems.

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ТОЧНОСТЬ РАСЧЕТА ХАРАКТЕРИСТИК РЕАКТОРОВ В ЗАВИСИМОСТИ ОТ ТОЧНОСТИ ЭЛЕМЕНТАРНЫХ КОНСТАНТ

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Abstract — Аннотация

THE ACCURACY OF CALCULATIONS OF REACTOR PARAMETERS AS A FUNCTION OF THE ACCURACY OF ELEMENTARY CONSTANTS. The paper describes an effective procedure for evaluating elementary constants from the viewpoint of their effect on various reactor parameters. This procedure has been developed on the basis of the perturbation theory evolved in a number of studies by two of the authors of this paper. This theory contributes linear relationships which link variations in elementary constants with the reactor parameter under consideration. The coefficients of these linear relationships are calculated by the method set forth in the studies. Storage in the memory of an electronic computer of the matrices of these coefficients for various reactor parameters and for an entire set of fast neutron reactors makes it possible, while operations are going on, to check the effect of different changes in the constants on variations in reactor parameters which are known experimentally.

In this way, new data on elementary constants can be evaluated during operation from the point of view of agreement between reactor calculations and experiment. Also, the required accuracy of the elementary constant measurements can be indicated in a simple manner, on the basis of the desired accuracy of the reactor parameter calculations.

The paper gives tables for the above-mentioned perturbation theory coefficients in respect of various parameters measured on a number of ZPR-3 critical assemblies and also for a typical fast neutron power reactor. Examples are given of using these coefficients to solve the problems involved.

ТОЧНОСТЬ РАСЧЕТА ХАРАКТЕРИСТИК РЕАКТОРОВ В ЗАВИСИМОСТИ ОТ ТОЧ-НОСТИ ЭЛЕМЕНТАРНЫХ КОНСТАНТ. Описывается эффективная процедура, употребляемая для оценки значений элементарных констант с точки зрения их влияния на различные характеристики реакторов. Указанная процедура разработана на основе теории возмущений, развитой в ряде работ авторов доклада. Эта теория возмущений дает линейные соотношения, связывающие вариации элементарных констант и рассматриваемого реакторного параметра. Коэффициенты этих линейных соотношений рассчитаны по разработанной в упомянутых работах методике. Хранение в памяти электронно-вычислительной машины матриц этих коэффициентов для различных параметров реактора и целого набора реакторов на быстрых нейтронах дает возможность оперативно проверять влияние тех или иных изменений в константах на вариации значений реакторных параметров, известных экспериментально.

Таким образом, появляется возможность оперативно оценивать новую информацию по элементарным константам с точки зрения согласия реакторных расчетов и эксперимента. Кроме того, становится возможным просто указать необходимую точность измерений элементарных констант, исходя из желаемой точности расчета реакторных характеристик.

В докладе приводятся таблицы упомянутых выше коэффициентов теории возмущений для некоторых параметров, измерявшихся на ряде критических сборок ZPR-3, а также для типичного энергетического реактора на быстрых нейтронах. Даются примеры решения указанных выше задач с использованием этих коэффициентов.

В данном докладе рассматривается вопрос о возможности систематического использования результатов макроскопических реакторных экспериментов при выработке рекомендуемых значений констант, а также вопрос об оценке погрешностей, которые могут быть в характеристиках реактора вследствие неопределенностей в ядерных данных.

ИСПОЛЬЗОВАНИЕ РЕЗУЛЬТАТОВ ИНТЕГРАЛЬНЫХ ЭКСПЕРИМЕН-ТОВ ПРИ ВЫРАБОТКЕ РЕКОМЕНДОВАННЫХ ЗНАЧЕНИЙ ЯДЕРНЫХ ДАННЫХ

Обычно проверка рекомендованных для расчета реакторов значений ядерных данных состоит в том, что с их помощью проводится расчет различных параметров, измеряемых на критических сборках и в других интегральных экспериментах. При этом чаще всего дело ограничивается констатацией полученных расхождений между экспериментом и расчетом, на основании чего делается вывод о той или иной степени пригодности этих констант для расчета реакторных систем, аналогичных рассмотренной. Результаты такого частного сравнения, как правило, не используются для внесения каких-либо изменений в систему констант. Надо отметить, что при создании известной 26-групповой системы констант [1]. разработанной под руководством покойного профессора И.И.Бондаренко, были использованы имевшиеся в то время результаты некоторых характерных макроскопических опытов. В результате эта система констант позволяет, например, достаточно точно вычислять эффективный коэффициент размножения kef и критическую загрузку быстрых реакторов на обогащенном уране [2], [3].

Что же касается других параметров, измеряемых на критических сборках, то они описываются существенно хуже, что свидетельствует о необходимости уточнения рекомендованных значений констант.

Мы хотим обратить внимание на метод, основанный на использовании теории возмущений [3-6], который позволяет проводить систематическую работу по улучшению констант с использованием совместно результатов как микроскопических, так и интегральных экспериментов. Речь идет о том, каким образом должны быть представлены результаты интегральных экспериментов для того, чтобы они могли быть эффективно использованы для выработки рекомендованных констант.

Проиллюстрируем это на примере обработки результатов критических опытов на сборках ZPR-III, моделирующих реакторы на быстрых нейтронах с урановым топливом. Эти опыты были проанализированы Дэви [7], который внес в экспериментальные результаты целый ряд поправок, к счастью, небольших, позволивших построить идеализированные модели, поддающиеся расчету. Поправки учитывали несферичность формы, изломанный характер поверхности сборки, гетерогенность и т.д. В константах, используемых для расчета этих моделей, учитывается резонансная самоэкранировка сечений при данном составе сборки [2]. В результате значения k_{ef}, полученные в расчете по константам [1], отличаются от единицы в среднем на 1,5%, но не более чем на 3% [2].

Наше предложение состоит в том, что после такой тщательной идеализации эксперимента следует использовать теорию возмущений, развитую для различных характеристик реакторов в цитированных выше работах [3-6]. В соответствии с этой теорией изменение любого параметра X, измеряемого на критической сборке и являющегося либо отношением интегралов по потокам нейтронов, т.е. отношениями чисел различных процессов, либо отношениями интегралов от произведения

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потоков и ценностей нейтронов, т.е. реактивностями образцов, временем жизни мгновенных нейтронов и т.д., может быть выражено через изменения констант следующим простым образом:

$$\Delta \chi / \chi = \sum_{\alpha \beta j} x_{\beta}^{\alpha j} \frac{\Delta \sigma_{\beta}^{\alpha j}}{\sigma_{a}^{\alpha j}}$$
(1)

Здесь α — индекс изотопа, β — индекс вида сечения, j — номер энергетической группы. В цитированных выше работах приведены формулы для коэффициентов $x_{\beta}^{\alpha j}$. Вычислив однажды эти коэффициенты для каждой сборки и для всех измеренных на ней характеристик, можно проводить эффективную и целенаправленную работу по улучшению рекомендованных значений констант $\sigma_{\beta}^{\alpha j}$, добиваясь наилучшего согласия экспериментальных и расчетных характеристик всех имеющихся сборок.

Покажем, как это можно сделать, на примере двух сборок ZPR-III: № 11 (загрузка урана-235 240,6 кг, обогащение 11,8%, объемная доля топлива 81,2%) и № 29 (420,7 кг, 33,3% и 15%, соответственно). Рассмотрим такой плохо описываемый параметр, как время жизни мгновенных нейтронов 1. В табл.1 и 2 приведены вычисленные нами коэффициенты формул (1) $k_{A}^{\alpha j}$ и $l_{A}^{\alpha j}$, описывающие зависимость $\Delta k_{ef} / k_{ef}$ и $\Delta l/l$ от изменений сечений поглощения ос и деления оf , а также от изменения числа вторичных нейтронов деления v для урана-235 и урана-238. Характерным для времени жизни является то, что расчетная величина его чувствительна к абсолютным значениям сечений, тогда как, например, для коэффициента размножения kef, в основном, существенен лишь их относительный ход. Экспериментальные значения1 равны 7,0·10⁻⁸ сек в сборке № 11 и 23,2.10-8 сек в сборке № 29, тогда как расчет по константам [1] дает 5,94·10⁻⁸ сек и 19,1·10⁻⁸ сек, соответственно [2]. Расчет показывает, что основной вклад в 1 в этих сборках вносят нейтроны активной зоны, энергия которых лежит в интервале от 2-3 кэв до 3-4 Мэв.

Проявившиеся недавно результаты измерений сечения деления урана-235 в области от 40 до 505 кэв [8], которые хорошо согласуются с рекомендованными значениями из атласа [9], лежат ниже, чем значения σ_f^{235} , принятые в системе констант [1]. Если исходить из этих результатов, групповые сечения деления урана-235 следует уменьшить примерно так, как указано ниже:

№ группы	6	7.	8	9
Интервал, Мэв	0,4-0,8	0,2-0,4	0,1 - 0,2	0,0465 - 0,1
$\Delta\sigma_{\mathrm{f}}^{235}/\sigma_{\mathrm{f}}^{235}$, %	- 6	- 7	-12	- 15
№ группы	10		11	12
Интервал, Мэв	0,0215 - 0,046	5 0,010	0-0,0215	0,00465 - 0,010
$\Delta\sigma_{ m f}^{235}/\sigma_{ m f}^{235}$, %	-17		- 20	- 22

Кроме того, последние измерения числа нейтронов спонтанного деления

лма	Су	18	17	16	15	14	13	12	11	10	9	8	7	6	5	4	3	2	1	<u>j</u>
)45	- (00	00	000	000	000	000	000	- 001	- 005	- 009	- 011	- 010	- 006	- 002	- 001	000	000	000	K _c ²³⁵
195	4	00	00	000	000	000	000	000	005	029	064	093	115	100	047	022	012	006	002	K_{f}^{235}
745	5	00	00	000	000	000	000	001	008	043	095	141	1 73	147	068	037	020	009	003	K_{ν}^{235}
244	- 2	00	00	000	000	000	000	000	- 003	- 021	~ 045	- 053	- 054	- 043	- 018	- 006	- 001	000	000	K ²³⁸
57	t I				ļ]			007	070	048	025	007	κ_{f}^{238}
256	2						}	ļ	ļ	1					009	115	078	039	015	K_{ν}^{238}
)69	- 0	00	00	000	000	000	000	- 001	- 008	- 0 2 2	- 021	- 015	005	001	001	001	000	000	000	1^{235}_{c}
756		00	00	000	000	000	000	002	014	028	- 017	- 109	- 253	- 211	- 112	- 050	- 030	- 014	- 004	1_{f}^{235}
64	- 5	00	00	000	000	000	000	005	031	086	058	- 046	- 223	- 215	- 124	- 071	- 040	- 019	- 006	1^{235}_{ν}
355	- 3	00	00	000	000	000	000	- 003	- 026	- 094	-124	- 087	- 041	005	008	006	001	000	000	1^{238}_{c}
\$79	- :					[ļ							- 015	- 172	-116	- 058	- 018	1_{f}^{238}
86	- 4														- 016	- 217	-150	- 075	- 028	1_{ν}^{238}
75 16 35 37	- 1 - 5 - 5 - 5 - 5 - 5	00 00 00	00 00 00	000	000	000	000	002 005 - 003	014 031 - 026	028 086 - 094	- 017 058 - 124	- 109 - 046 - 087	- 253 - 223 - 041	- 211 - 215 005	- 112 - 124 - 008 - 015 - 016	- 050 - 071 006 - 172 - 217	- 030 - 040 001 - 116 - 150	- 014 - 019 000 - 058 - 075	- 004 - 006 000 - 018 - 028	1_{p}^{235} 1_{ν}^{235} 1_{c}^{238} 1_{c}^{238} 1_{f}^{238} 1_{ν}^{238}

ТАБЛИЦА 1. КОЭФФИЦИЕНТЫ ФОРМУЛЫ (1) ДЛЯ СБОРКИ ZPR-III № 11 (·10³)

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усачев и зарицкий

j	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	Сумма
K ²³⁵	000	000	000	- 001	- 002	- 006	- 01 0	- 014	- 017	- 015	- 014	- 007	- 003	- 001	000	000	00	00	- 090
K_{f}^{235}	002	006	014	028	040	068	086	083	081	053	040	018	007	002	001	000	00	00	529
K_{n}^{235}	002	009	021	045	063	110	142	142	141	094	073	033	013	004	001	000	00	00	893
K ²³⁸ c	000	000	000	- 002	- 008	- 017	- 024	- 029	- 035	- 022	- 015	- 006	- 003	- 001	, 000	000	00	00	- 162
κ_{f}^{238}	003	009	020	034	002	· ·		· ·											068
K_{v}^{238}	005	014	031	053	004														107
1^{235}_{c}	000	000	000	001	002	003	004	- 002	- 013	- 024	- 039	- 031	- 020	- 009	- 003	- 001	00	00	- 132
1_{f}^{235}	- 004	- 014	- 033	- 070	- 101	-170	- 209	- 192	-160	- 082	- 030	004	012	008	003	000	00	00	-1038
1_{ν}^{235}	- 005	- 017	- 042	- 088	- 119	- 195	- 225	- 184	- 115	- 01 8	059	070	051	026	009	001	00	00	- 792
1^{238}_{c}	000	000	000	003	006	008	003	- 010	- 033	- 040	- 045	- 028	- 017	- 007	- 002	000	00	00	-162
1_{f}^{238}	- 007	- 023	- 048	- 083	- 006														- 167
1_{ν}^{238}	- 009	- 028	- 063	-104	- 007			}											- 211
	1					1		1			1			F	1				

ТАБЛИЦА 2.	коэффициенты	ФОРМУЛЫ (1)	для	СБОРКИ	ZPR-III № 29)(•10 ³)
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CN-23/94

калифорния-252 [10] указывают на 2%-ное уменьшение этой опорной величины и, следовательно, на такое же снижение значений v для других изотопов.

Посмотрим, к каким последствиям может привести указанное изменение величин σ^{235} , ν^{235} и ν^{238} . При этом будем считать $\alpha^{235} = \sigma_c^{235} / \sigma_f^{235}$ неизменной [1], [9], то есть соответственно уменьшению σ_f^{235} уменьшим и σ_c^{235} . Уменьшение размножения нейтронов скомпенсируем уменьшени~ ем сечения захвата в уране-238, поскольку это сечение сильно влияет на k_{ef} и в то же время в экспериментальных значениях этой величины наблюдается значительный разброс. Пользуясь данными табл.1 и 2, а также учитывая превышение расчетного значения kef на 2,6% для сборки № 29 [2], нетрудно получить, что для приведения k_{ef} к единице надо уменьщить σ²³⁸ на 22% и 23% для сборок № 29 и № 11, соответственно. Отметим, что такое уменьшение также находится в согласии с результатами последних весьма тщательных измерений, выполненных в сферической геометрии [11]. Из таблиц 1 и 2 видно, что все эти изменения констант действуют в сторону увеличения расчетного значения времени жизни. Расчеты по формуле (1) показывают, что увеличение 1 в случае сборки № 11 составит при этом 15%, а в случае сборки № 29 17%, в результате чего согласие с экспериментом существенно улучшится. Отметим кстати, что в пользу изменения констант в указанную сторону говорят и имеющиеся расхождения в таких характеристиках, как изменения реактивности, возникающие при помещении в сборку образцов урана-235 и урана-238. Отношение измеренной реактивности урана-235 к вычисленной составляет 0,891 и 0,947 для сборок №11 и №29, соответственно; те же отношения для урана-238 равны 0,811 и 0,889 [2] (реактивность, вносимая ураном-238, отрицательна).

На этом примере мы показали, как результаты инте́грального эксперимента выступают в поддержку наметившейся тенденции изменения микроскопических ядерных данных. Окончательное суждение по поводу сделанного выше изменения рекомендованных констант из 26-группового набора [1], можно будет сделать только после более детального анализа всех имеющихся экспериментальных работ по σ_f²³⁵ и σ_c²³⁸ и, конечно, после проверки этих изменений на всех параметрах всех известных быстрых сборок.

Хранение в памяти электронно-вычислительной машины матриц всех коэффициентов X_B^{CQ} для всех сборок даст возможность оперативно проверять совместимость тех или иных изменений в ядерных данных с известными результатами экспериментов на критических сборках. Варьируя при этом ядерно-физические константы в пределах разброса экспериментальных данных, можно будет подбирать рекомендованные значения этих констант.

Нам представляется, что результаты любого макроскопического эксперимента в физике реакторов должны аналогичным образом обрабатываться с помощью теории возмущений. Только в этом случае эти эксперименты могут сыграть свою роль при выработке рекомендованных значений констант.

2. ОЦЕНКА НЕТОЧНОСТИ В КРИТИЧЕСКОЙ ЗАГРУЗКЕ И КОЭФФИЦИЕНТЕ ВОСПРОИЗВОДСТВА ПЛУТОНИЕВОГО БРИДЕРА ИЗ-ЗА НЕОПРЕДЕЛЕННОСТИ В КОНСТАНТАХ

Выясним теперь, как повлияют рассмотренные выше изменения в константах урана-238, а также возможные изменения в константах плутония-239, на характеристики типичного энергетического реактора на быстрых нейтронах. Эти изменения соответствуют разбросу между рекомендованными 26-групповыми константами [1] и наиболее далеко отстоящими от них экспериментальными данными, появившимися в самое последнее время.

Рассмотрим реактор с топливом в виде смеси UO₂ и PuO₂ и натриевым охлаждением. Объем активной зоны 1800 л, загрузка плутония-239 740 кг, относительная концентрация плутония в топливе 11,6%, объемные доли топлива, натрия и стали составляют 50, 33 и 17%, соответственно. Начальное значение коэффициента воспроизводства (КВ) равно 1,57. В таблице 3 приведены коэффициенты формул (1) k^{αj}_β и m^{αj}_β, позволяющие вычислить изменения k_{ef} и КВ данного реактора при изменении σ_c, σ_f и ν плутония-239 и урана-238.

Поскольку выше мы, следуя работе [8], изменяли сечения деления урана-235, то логично рассмотреть возможные изменения σ_f плутония-239, также вытекающие из результатов этой работы. Эти изменения констант [1] должны быть примерно следующими:

№ группы	6	7	8	9
Интервал, Мэв	0,4-0,8	0,2-0,4	0,1-0,2	0,0465 - 0,1
$\Delta \sigma^{239} / \sigma^{239}$, %	- 5	- 8	-10	- 21
№ группы	10		11	12
Интервал, Мэв	0,0215 - 0,04	65 0,01	10 - 0,0215	0,00465 - 0,010
Δσ ²³⁹ /σ ²³⁹ , %	- 33		- 40	- 42

Расчет по формуле (1) с коэффициентами из таблицы 3 дает, что k_{ef} указанного реактора уменьшается при этом на 4,4%. Для компенсации такого уменьшения k_{ef} критическая загрузка реактора должна быть увеличена примерно на 8%. Расчетное значение КВ снижается на 5,6%. Однако темп расширенного воспроизводства плутония определяется величиной (КВ-I), которая в этом случае уменьшается примерно на 15%.

Таким образом, мы видим, что указанные изменения в сечениях приводят к весьма заметным изменениям основных характеристик энергетического быстрого реактора на плутониевом горючем. Эти изменения могут служить оценкой возможных погрешностей при расчете этих параметров, таких погрешностей, которые обусловлены неопределенностью в значениях ядерно-физических констант.

В заключение мы выражаем свою искреннюю благодарность H.O.Базазянц и М.Ф.Троянову, оказавшим нам большую помощь в работе над докладом.

jI	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	Сумма
K 239	000	000	000	000	000	- 002	- 003	- 005	- 007	- 008	- 009	- 004	- 002	- 004	- 007	- 001	00	00	- 052
K ²³⁹ f	002	007	015	031	035	069	079	089	084	068	057	022	010	017	020	003	00	00	608
К ²³⁹	002	010	023	046	050	097	112	114	115	093	077	030	013	024	030	005	00	00	841
K_{c}^{238}	000	000	- 001	- 004	- 009	- 018	- 024	- 034	-048	- 043	- 039	- 018	- 009	- 015	- 012	- 002	00	00	- 276
${ m K}_{ m f}^{238}$	005	014	029	048	003			ļ											099
K_{ν}^{238}	007	022	047	079	004														159
m_{c}^{239}	000	000	000	001	- 002	- 009	- 017	- 024	- 036	- 045	- 050	- 022	- 010	- 021	- 027	- 005	00	00	- 267
m_{f}^{239}	001	004	007	014	017	031	036	038	040	035	031	013	006	012	017	003	00	00	305
m_{ν}^{239}	004	018	041	082	089.	174	201	205	209	170	142	057	025	046	058	010	00	00	1531
m_c^{238}	000	000	000	000	002	005	009	016	029	032	036	019	011	018	015	003	00	00	195
m_{f}^{238}	009	027	057	098	005										1				196
m_{v}^{238}	015	043	095	161	008	1													322
			1																

ТАБЛИЦА З. КОЭФФИЦИЕНТЫ ФОРМУЛЫ (1) ДЛЯ ПЛУТОНИЕВОГО РЕАКТОРА-РАЗМНОЖИТЕЛЯ (·10³)

CN-23/94

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DISCUSSION

(on papers CN-23/15 and CN-23/94)

K. PARKER: I should like to underline the promise of the investigations reported by Reiss and Usachev. They offer the possibility of :

- (1) Systematic improvement of existing group cross-section sets
- (2) Systematic attainment of the desired accuracy as a function of energy for microscopic cross-section measurements
- (3) Pointers to the choice of preferred data when an evaluator is faced with a choice between conflicting microscopic data.

At Aldermaston, Hemment and Pendlebury have made extensive calculations using a 25-group set of cross-sections. Following transport and perturbation calculations on fast critical assemblies, adjustments to group cross-sections are calculated, leading to an improved set.

Full details of the work were given in a paper presented at the International Conference on Fast Critical Experiments and their Analysis, held at Argonne National Laboratory, 10-13 October 1966.

Figure 1 from that paper shows values of $\Delta B = B - 1$ (B being the inverse reactivity) for 25 spherical critical assemblies containing ²³⁵U and ²³⁸U; the values were calculated using unadjusted cross-sections based on an evaluation made in 1962. With perfect cross-section data the calculated values should lie between the horizontal broken lines.

Figure 2 shows that, after adjustments that are reasonable in relation to experimental cross-section errors, excellent results are obtained; it seems clear that the adjusted group cross-sections may be used with

DISCUSSION



FIG.1. ΔB as a function of core radius with unadjusted data.

considerable confidence to calculate similar properties of systems analogous to the 25 used.

Y. REISS: Did Hemment and Pendlebury, in their calculations, make corrections to the group constants or to the cross-sections themselves?

K. PARKER: They made corrections to the group constants. This, of course, means that there is no unique answer for the microscopic cross-section curves, although you can discern trends.

Y. REISS: I ask because, if one uses only corrected group constants, the fit is not valid for other critical systems. On the other hand, we hope that our method of correcting the cross-sections themselves will give a fit that can also be used for other critical systems.

K. PARKER: The crucial point, in my opinion, is whether the systems one calculates are sensitive to a particular cross-section; the question as to whether one makes corrections to the group cross-sections or to the basic microscopic cross-sections is less important.

I think you are implying that one can use the resulting cross-sections with reasonable confidence as an interpolation in the region bounded by the system studied, but that one has to be careful when one goes outside it.

H. GOLDSTEIN: Dr. Reiss, I confess to difficulty in understanding the parameter ω , "the number of measurements per unit energy interval". Does this imply that the measurements are uncorrelated with random statistical (but not systematic) errors? For example, what do you use for ω when the cross-sections are measured experimentally as a continuous function of energy, as in the recent experiments carried out in the United States with a nuclear explosion source? DISCUSSION



FIG. 2. $\triangle B$ as a function of core radius after data adjustment.

Y. REISS: Only the quantity ωdE has a physical meaning; on its own ω has none. The reason is that one cannot in any measurement obtain infinitely fine energy resolution. In measurements at energy E,

CROSS-SECTIONS AND RESONANCE PARAMETERS FOR ²³⁵U, ²³³U, ²³⁹Pu AND ²⁴¹Pu BETWEEN CADMIUM CUTOFF AND 10 keV

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Abstract

CROSS-SECTIONS AND RESONANCE PARAMETERS FOR ²³⁵ U, ²³³ U, ²³⁹ Pu AND ²⁴¹ Pu BETWEEN CADMIUM CUTOFF AND 10 keV. Fission and capture cross-sections between Cd cutoff and 10 keV have been evaluated for the fissile isotopes, based on the available experimental differential cross-sections, capture-to-fission ratios and total fission and capture resonance integrals. In particular, the recommended cross-sections contained in the 1965 edition of BNL-325 were compared with some other evaluations and with more recent results of cross-section measurements using nuclear explosives in space and underground.

As a result of this evaluation, it is felt that even for the most intensively studied nucleus, ²³⁵U, the uncertainties in the fission cross-section are about $\pm 5\%$ and in the capture cross-section $\pm 20\%$. For ²³⁹Pu, σ_f is known to $\pm 10\%$ and σ_c to $\pm 25\%$. For ²³³U and ²⁴¹Pu, the uncertainty in σ_f is $\pm 20\%$ and in σ_c , $\pm 30\%$.

Different sets of resonance parameters for the resolved resonances, which are recommended in BNL-325 or by other research laboratories, were compared by calculating infinitely dilute total and partial resonance integrals. Calculations were performed with the resonance integral code TRIX-1 which was developed at A tomics International and is based on the single-level Breit Wigner model. The well-known limitations of this model for nuclei, such as the fissile, with narrowly spaced interfering levels result in calculated fission resonance integrals, which are 10-20% too small when compared with integrated measured differential cross-sections. This result was found for energy regions where the cross-sections are sufficiently well known to make such a comparison possible.

Average parameters for the unresolved resonances have been extrapolated from resolved resonance parameters. Calculated partial resonance integrals, using these parameters, were compared with integrated measured cross-sections where these are available. As no published capture cross-section measurements exist in the resonance region for ²³³U, ²³⁹Pu, and ²⁴¹Pu these cross-sections were calculated with the determined average parameters and appropriate statistical distributions.

1. INTRODUCTION

Exact knowledge of the fission and capture cross-section as a function of neutron energy for the fissile nuclei in the resonance region is of great importance for the analysis of safety and economic characteristics (Doppler coefficient and breeding ratio) of modern, large, fastbreeder reactors.

Unfortunately, at the present time nuclear resonance data are not known with sufficient accuracy. Although there has been considerable progress in the measurement of cross-section data for resonance neutron interactions over the past few years, the results of the various experiments are in many cases not consistent. Thus, evaluation of the data is necessary.

Such evaluations are available from different sources, such as BNL-325, the 26 Russian group set by Bondarenko et al., or the compilation of Schmidt from Karlsruhe. They usually result in "best" or recommended cross-sections. These evaluations are very much appreciated by the reactor designer. But a new difficulty results from the fact that very often the recommended cross-sections of different evaluators

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still show a wide spread. This task was initiated to reduce these discrepancies by considering more recent experimental results.

To calculate self-shielding factors and their variation with temperature, an analytical representation of the evaluated fission and capture cross-section is necessary. The single-level Breit-Wigner model is most often used because of its simplicity. But, the small level spacing for the fissile isotopes produces strong interference effects between neighbouring levels. These interference effects are particularly evident for the fission resonances. They lead to asymmetric line shapes and relatively high fission cross-sections between resonances. Because of the many exit channels available in the capture process, the interference effects between capture resonances tend to cancel and the capture crosssection is small between neighbouring resonances. These strong interference effects raise doubts concerning the adequacy of the single-level model used to describe cross-sections for the fissile nuclei. Use of a multi-level formalism certainly would improve the accuracy, but is impractical for survey calculations because it is much more complicated.

For many cases the single-level model is adequate to describe cross-sections and calculate Doppler effects. The energy regions between resonances, for which the calculated cross-sections are too small, give only a small contribution to the Doppler effect. Therefore one may apply an empirical correction to the single-level results by adding a suitable smooth background to the capture and fission cross-sections which would produce agreement with measured data. To determine this background one has to compare the measured (or evaluated) cross-sections as a function of neutron energy with those derived from calculated infinitely dilute resonance integrals using single-level resonance parameters. Such a comparison is very tedious if it is carried out for each single resonance, or even in very narrow energy groups. Therefore, group resonance integrals have been calculated splitting the resonance region into about 10 groups with an energy structure commonly used in multigroup diffusion calculations.

Different sets of resonance parameters from several authors have been used for comparison. For the unresolved resonance region average parameters were extrapolated from resolved resonance parameters and modified if necessary to obtain agreement between calculated partial resonance integrals and integrated point cross-sections.

2. DESCRIPTION OF THE RESONANCE INTEGRAL CODE TRIX-1

Partial resonance integrals and multigroup cross-sections were calculated with TRIX-1 [1], an improved version of the ARES Resonance Integral Code [2]. The theory is based on the single-level Breit-Wigner line shape with no overlapping of neighbouring resonances. Resolved resonances can be treated in several approximations including a nonlinear combination of the NR and NRIM approximation, as developed by Goldstein and Cohen [3].

The unresolved resonances are treated in the NR approximation. Average resonance parameters for l=0 and l=1 neutrons, and statistical distributions of level widths, are used. The contribution to the resonance integral at each energy point is taken as the average of integrals calculated using several sets of parameters obtained by sampling from the level width distribution. For the scattering width, a Porter-Thomas distribution (the chi-squared distribution with one degree of freedom) is assumed, while a chi-squared distribution with three degrees of freedom is used for the fission widths. The energy dependence of the average neutron width $\langle \Gamma_n \rangle$ is given by $\langle \Gamma_n \rangle = \langle \Gamma_n^0 \rangle \cdot E^{\frac{1}{2}}$ (Γ_n^0 = reduced neutron width; E = neutron energy), while the radiation width Γ_y and the average level spacing $\langle D \rangle$ are taken to be energy independent. The average fission width $\langle \Gamma_f \rangle$ is chosen group-wise, so that experimental cross-sections and capture-to-fission ratios are best approximated. 1/v and negative-energy resonance contributions to the resonance integral can be calculated from resonance parameters; they are taken to be independent of temperature and concentration.

Group-wise cross-sections are defined as the quotient of the resonance integral and the integrated flux for the group.

For the energy dependence of the average fission width simple correlation functions are determined (see Fig.1). The average fission width $\langle \Gamma_f \rangle_g$ for each group is computed by the code from the correlation function f using the following relation:

$$\langle \Gamma_{\rm f} \rangle_{\rm g} = f \left(\sqrt{E_{\rm low} \cdot E_{\rm high}} \right)$$

3. EVALUATION OF CROSS-SECTIONS AND RESONANCE PARAMETERS

3.1.²³⁵U

3.1.1. Cross-sections

To compare resonance integrals calculated from resonance parameters with evaluated resonance cross-sections, the following partial resonance integrals were computed by numerical integration of 1/Espectrum-weighted differential cross-sections:

$$(\mathrm{RI}_{f})_{i} = \int \sigma_{f} \frac{\mathrm{dE}}{\mathrm{E}}; \quad (\mathrm{RI}_{c})_{i} = \int \sigma_{c} \frac{\mathrm{dE}}{\mathrm{E}}$$
$$\Delta E_{i} \qquad \Delta E_{i}$$

where i represents the i-th group.

Table I presents in the first columns several sets of group-wise fission integrals. Recommended data from the 1965 edition of BNL-325 [4], as well as other published data, have been weighted with a 1/Espectrum and are presented in a group-wise breakdown. Resonance integrals calculated in the same manner by Hanna and Walker [6], Schmidt [7], and Freemantle [8] are also given. The data presented by both BNL-325 [4] and Schmidt [7] are based on experimental results of Michaudon [9, 10].

The comparison illustrates that the agreement is not satisfactory and that, in general, the fission cross-section in the resonance region is known with an accuracy of about 5 to 10%. While the agreement is quite good

TABLE I. PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS FOR ²³⁵U. INTEGRATED CROSS-SECTIONS AND INTEGRALS CALCULATED WITH THE TRIX-1 CODE USING DIFFERENT SETS OF RESONANCE PARAMETERS.

				Fission resonan	nce integrals (1	Capture resonance integrals (b)								
		Integr	ated cross-se	ctions		Integra	red cross-sect	fons	c					
Energy						Parameters from:						Parameters from:		
(eV)	BNL 325	General Atomic	Hanna Walker	Karlsruhe	Winfrith	BNL	Schmidt	S chimidit	Karlsnihe	Hanna Walker	Winfrith	Schmidt	BNL	Schmidt
Ref:	[4]	[5]	[6]	[7]	[8]	[4]	[7]	(16]	[4]	[6]	[8]	(7)	[4]	[16]
0.45-1.8	77.06	77.06	78.0	77.07	76.40	65.74	67.31	67.10	15,12	15.0	13.6	16.11	16.16	16.25
1.8-5	16 .46	16.46	18.1	16.57	17.00	15.95	16.39	16,83	12.73	13.3	14.1	12.78	14.62	13.80
5 -1 0	35,24	35.24	30.9	35.86	32.50	28.04	33.79	31.11	24.03	31.4	27.3	26.20	31.27	28.60
10-20.5	31,00	30.84	30.3	32.53	25.30	23.29	26.23	27.25	39.88	34.6	28.0	40.63	37.59	35.79
20.5-41	29, 22	26.94	29.2	28.92	24.10	20.74	24.90	24.40	34.26	14.8	33.0	35.23	29,55	27.27
41-60	16.50	14.49	15.5	16.47	6.8	14.56	15.09	15.27	13.61	5.5	6.9	13.86	11.26	12.27
60-100	12,06	9.75	11.7	11.85	11.3		!	10.07	6,35	3.1	7.6		,	9.16
100-300		22.59	21.9	22.59	22.1			17,98	11.45	10,4	10.2			14.71
300-1000		14.86	12.8	14.86	14.1			13,79	7.13	6.4	6.3			7.36
1000-3000		7.67	6.8	7.67	7.6			7.50	3.45	3.1	3.5			3.28
3000-10 000		5.02	5.0	5,02	4.9			4,75	2 . 12	2.0	2.1			1,89

up to energies of about 10 eV, the discrepancies become larger with increasing energy.

The evaluations of General Atomic [5] and Winfrith [8] give very small values of the fission cross-section for several groups between 10 and 100 eV. These low values of the fission cross-section between 10 and 100 eV result from heavily weighting the measurements of Brooks and Jolly [11]. Data presented in BNL-325 [4] (page 92, 235-43) indicate that these experimental data appear to be much too low and need further corrections.

Group capture resonance integrals as determined by several evaluations including those of Schmidt [7], Hanna and Walker [6], and from Winfrith [8] are presented in Table I.

The available experimental data for the total fission resonance integral, the total capture resonance integral, and the capture-to-fission ratio, defined by

$$\alpha_{\rm RI} \approx \frac{\int \sigma_{\rm c}(E) \frac{dE}{E}}{\int \sigma_{\rm f}(E) \frac{dE}{E}} \\ > E_{\rm Cd}$$

have recently been evaluated by Feiner and Esch [12]. Their results are given in Table II. To allow for a comparison of these integral values with the data given in Table I, the following partial resonance integrals have been assumed:

Values of the capture-to-fission ratio α averaged over large energy intervals are given by de Saussure et al. [13], Freemantle [8] and Van Shi-di et al. [14]. The available data for α still show large discrepancies; in particular, there exists no good agreement between α -

TABLE II. TOTAL RESONANCE INTEGRALS AND α OF ²³⁵U FOR 1/E SPECTRUM; 0.50-eV Cd CUTOFF

Fission resonance integral	280±11 b
Capture resonance integral	140±8b
Capture-to-fission ratio α	0.50± 0.02
	-

Note: From evaluation by Feiner and Esch [12].

TABLE III. RECOMMENDED PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS AND DERIVED GROUP CONSTANTS FOR ²³⁵U, AND RESONANCE INTEGRALS CALCULATED BY TRIX-1 WITH THE RECOMMENDED RESONANCE PARAMETERS

Fnergy		Recommend integrals and (l	Resonance integrals from TRIX-1 with recommended parameters			
range (eV)	(RLf) _i	<af>g</af>	(RI _c) _i	<°c>g	(RI _f)i	(RI _c) _i
0.45-1.8	77.07	55.85	16.25	11.69	67.10	16.25
1.8-5	16.57	16.25	13.80	13.50	16.83	13.80
5-10	35,86	51.97	28.60	41.50	31.11	28.60
10-20.5	32.53	45.18	35.79	52.6	27.25	35.79
20.5-41	28.92	41.95	27,27	39.6	24.40	27.27
41-60	16.47	43.34	12.27	32.2	15.27	12.27
60-100	11.85	23.24	9.16	17.9	10.07	9.16
100-300	22.59	20.54	14.71	13.4	17.98	14.71
300-1000	14.86	12.38	7.36	6.12	13.79	7.36
1000-3000	7.67	6.97	3.28	2.97	7.50	3.28
3000-10 000	5.02	4.18	1.89	1.58	4.75	1.89

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values obtained from integral measurements, and integrated differential fission cross-section measurements and differential α -measurements. The integral-measured α of about 0.5 may be compared with $\alpha = 0.63 \pm 0.10$ which results from the new differential α -measurement of de Saussure et al. [13]. This result makes use of other measurements for the energy ranges which are not covered by their experiment. Figure 11 of Ref. [13] demonstrates that in the energy range 10 to 100 eV there are still large discrepancies among the results of various experimenters. Due to a remark made by Kinchin [15], a new analysis of the η , $\sigma_{\rm T}$, and $\sigma_{\rm f}$ measurements of Brooks and Jolly at Harwell led to a new lower value of $\alpha \approx 0.56-0.58$. Thus the differences between their α -values and those of other experimenters have been reduced.

Based on all this experimental information, recommended partial fission and capture resonance integrals have been chosen as presented in Table III. Also given are infinitely dilute group constants, which were calculated from the recommended resonance integrals as follows:

$$\langle \sigma \rangle_{g} = \frac{\int_{\Delta E_{g}} \sigma \frac{dE}{E}}{\int_{\Delta E_{g}} 1/E dE} = \frac{(RI)_{g}}{\Delta u}$$

where Δu is the lethargy width for the group.

The recommended fission data are taken from the measurements of Michaudon [10]. With those one obtains a total fission resonance integral (E > 0.5 eV) of

$$RI_{f} = 271 b$$

The value of 271 b is in reasonable agreement with recent experimental results for the 235 U fission resonance integral (see Table II).

For the recommended capture integrals and group constants, the results of a TRIX-1 calculation with resolved resonance parameters given by Schmidt and average parameters for the unresolved region as determined in this work (see section 3.1.2) have been chosen. The reason for this choice is that these capture resonance integrals combined with the recommended fission resonance integrals give group values of the capture-to-fission ratio α , which are in reasonable agreement with experimental results. Taking the fission and capture resonance integrals recommended in Table III one gets for $E_{Cd} = 0.5$ eV and appropriate consideration of the energy range > 10 keV and $\alpha = 0.58$. This value may be too high, when compared with the integral α -measurements, but it is within the range covered by the differential experimental results.

The recent measurements of de Saussure et al. [13] have shown that even for 235 U the accuracy of 1% for σ_{f} and 5% for α , which is desirable for reactor calculations, is far from being reached.

3.1.2. Resonance parameters

Three different sources for resonance parameters were available for the attempt to find the best representation of the recommended partial resonance integrals of Table III by TRIX-1 calculations. These are evaluated parameters for 80 resolved resonances up to 60 eV by Schmidt [7]' presented in 1965 at the Brookhaven Seminar, evaluated parameters up to 45 eV by BNL-325 [4], and the most recent parameter set of Schmidt [16], which was presented at the 1966 ANS-meeting in San Diego. This set extends the resolved region to 150 eV.

Table III contains the results of three TRIX-1 calculations for fission and capture resonance integrals with the different resonance parameter sets. In the column headed "Parameters from Schmidt [7]", the resonance integrals are based on the parameters for 80 resolved resonances. In the column headed "BNL" results are presented, which were obtained for the same 80 resonances, but when given, the recommended parameters of the recent BNL-325 [4] edition were used instead of the Karlsruhe parameters [7]. Finally (column Schmidt [16]), calculations based on parameters for 216 resolved resonances up to 150 eV were carried out. It is felt that the new Schmidt [16] data give the best cross-section representation.

In the unresolved region average parameters have been determined, which result in the best approximation of the recommended cross-sections over the whole resonance region. The TRIX-1 results obtained with these parameters are presented in Tables I and III. The average parameters are slightly different from those recommended by Schmidt [16]. The strength-function used for 1=0 neutrons of 1.01×10^{-4} is in agreement with the value given by Uttley [17].

The TRIX-1 results obtained with the recommended parameters are presented again in Table III for intercomparison with the recommended partial resonance integrals. The resulting recommended resonance parameters are presented in Table IV. Figure 1 shows the correlation for $\langle \Gamma_f \rangle$.

3.2.²³³U

3.2.1. Cross-sections

The new edition of BNL-325 [4] shows that there is still little information available about the fission cross-section and α in the resonance region and that the available measured data contain large discrepancies. For σ_f , the results of two measurements are given: Nifenecker [18] has measured the fission cross-section up to 60 eV, while measurements by Moore et al. [19] extend up to 900 eV. In addition there are older measurements of σ_f (Adamchuk et al. [20]) up to 750 eV. Until then, no σ_f measurements had been published for the energy region 1-10 keV.

This situation was considerably improved recently when the results of two additional high-energy resolution measurements became available. Albert [21] made fission cross-section measurements for 233 U between 30 eV and several MeV by means of a time-of-flight measurement using a nuclear explosive in space as the neutron source. The detectors were sent into space by rocket and the distance to the neutron source was about 800 miles. The results were obtained as fission ratio measurements be-

TABLE IV. RECOMMENDED RESONANCE PARAMETERS FOR 235 U

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Number of resolved resonances: (Parameters as given by Schmidt [16])

Parameters for negative energy resonance:

/E/ = 0.95 eVg = 0.5 $\Gamma_n^0 = 1.488 \times 10^{-3} \text{ eV}^{\frac{1}{2}}$ $\Gamma_\gamma = 27.6 \text{ meV}$ $\Gamma_f = 169 \text{ meV}$

Average parameters for unresolved resonances:





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Energy range (eV)	In: BNL-325	Integrated cross-sections Albert Bondarenko		TRIX-1 calculation BNL parameters
				L
Ref.:	[4]	[21]	[23]	
0.5-1	89.64		100.10 ^a	55.47
1-2.15	270.45		288.75	254.89
2.15-4.65	96.25		100.10	93.57
4.65-10	71.26		84.70	60.81
10-21,5	85.14		88.55	73.64
21.5-46.5	54.16		50,05	43.13
46.5-100	25.11		24.64	31.22
100-215	17.15	21.91	19.25	21.53
215-465	12.57	19.89	12.32	14.71
465-1000	9.04	12.06 ^b	9.24	10.05
1000-2150		7.32	6.78	6.90
2150-4650		5.23	5.93	4.73
46 50 -10 000		4.04	5.08	3.26

TABLE V. PARTIAL FISSION INTEGRALS (barns) FOR ²³³U. INTEGRATED CROSS-SECTIONS AND INTEGRALS CALCULATED WITH THE TRIX-1 CODE.

^a 0.465 - 1 eV group

b Interpolated data

tween the 233 U and a second detector containing 235 U. Between 30 and 100 eV the results were normalized to the 235 U values of Michaudon [10], which are almost identical to the numbers given as recommended in BNL-325 [4]. The results for σ_f for 233 U below 100 eV and especially between 100 and 500 eV are considerably higher than the available data. Unfortunately no results were measured between 500 and 900 eV, but for the first time differential data were measured between 1 and 10 keV. Above 20 keV the results are in good agreement with older data.

Time-of-flight measurements recently performed with an underground nuclear explosion by Hemmendinger et al. [22] show excellent energy resolution. Preliminary results give reasonable agreement with Albert's data for energies between 1 and 10 keV. But the experiment is still being analysed and final values were not available.

Partial fission resonance integrals found by 1/E-weighting and integrating the point cross-sections are presented in Table V. Numerically integrated were the data recommended in BNL-325 [4] and those of Albert [21]. Between 465 and 900 eV, Albert's data have been interpolated. For comparison the Bondarenko [23] data are also given. In this case the resonance integrals were calculated by multiplying the group constants by the lethargy width Δu .

TABLE VI.	TOTAL RESONANCE INTEGRALS
AND α OF 233	U FOR 1/E SPECTRUM AND
0.50-eV Cd C	CUTOFF

780±40 b	
137±7b	
0.175±0.008	
	780 ± 40 b 137 ± 7 b 0.175 ± 0.008

Note: From evaluation by Feiner and Esch [12]

There is still very little information available about capture resonance integrals or capture-to-fission ratios. Available information has large uncertainties, in particular the differential data. Between 10 and 1000 eV, the only measurements are those of Yeater et al. [24]. These values were derived from η -measurements and have large errors because of the small value of α for ²³³U.

The available results for total resonance integral measurements, as recently evaluated by Feiner and Esch [12], are given in Table VI. Based on this information, evaluated partial fission resonance integrals and group constants were obtained. They are presented in Table VII. Up to 21.5 eV, the BNL-325 recommended data have been taken without change. Between 21.5 eV and 1 keV, the different results have been averaged and heavily weighted by the measured data of Albert [21]. Above 1 keV, the recommended data are based on Albert's results. For these recommended fission resonance integrals, one obtains an integral of 770 b for the energy range 0.5 eV to 10 keV. By adding 13 b from Albert's data for energies above 10 keV, one gets a total resonance integral of 783 b. This value is in good agreement with experimental results, as can be seen from Table VI. But since the main part of the resonance integral comes from the low energy resonances, this comparison is not considered a severe test for the unresolved resonance region.

The partial capture resonance integrals and group constants calculated by the TRIX-1 code (with the parameters given in section 3.2.2) have been taken, without any change, as recommended values, although some of the resulting values are lower than the Yeater data. The values are given in Table VII. They result in a capture resonance integral of 147.3 b for the energy range 0.5 eV to 10 keV. By adding 1.5 b for

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TABLE VII. RECOMMENDED PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS AND DERIVED GROUP CONSTANTS FOR ²³³U, AND RESONANCE INTEGRALS CALCULATED BY TRIX-1 WITH THE RECOMMENDED RESONANCE PARAMETERS

Energy range (eV)	Recommended resonance integral and group constants (b)				Resonance integrals from TRIX-1 with recommended parameters	
	(RI _f)i	<°f ^{>} g	(RI _C)i	<σ _c >g	(RI _f) _i	(RI _c) _i
0.5-1	89.64	129.9	6.78	9.82	55.47	6.78
1-2.15	270.45	351.2	38.78	50.36	254.87	38,78
2.15-4.65	96.25	125.0	39.67	51.51	93.57	39.67
4.65-10	71.26	92,5	16.12	20.93	60.81	16.12
10-21.5	85.14	110.6	17.12	22.23	73.64	17.12
21.5-46.5	58.0	75.3	8.02	10.42	43.13	8.02
46.5-100	32.5	42.2	8.03	10.43	31.22	8.03
100-215	21.5	27,9	5.00	6.49	21.53	5.00
215-465	18.0	23.4	3.27	4.25	14.71	3.27
465-1000	11.0	14.3	2.00	2.60	10.05	2.00
1000-2150	7.32	9.51	1.23	1.59	6.90	1,23
2150-4650	5,23	6.79	0.78	1.01	4.73	0.78
4650-10000	4.04	5.25	0.45	0.58	3.26	0.45

energies above 10 keV, one gets 148.8 b for the total capture integral. This leads to a value $\alpha = 0.19$ when the recommended fission resonance integral as given in Table VII is used. All these numbers are in reasonable agreement with experimental results.

To gain more information about the capture cross-section, the results of the total cross-section measurement by Pattenden and Harvey [25] have been averaged over a 1/E-spectrum. The results are given in Table VIII together with group constants for potential scattering by Bondarenko [23] and the recommended group constants for fission as given in Table VII. It can be seen that by taking the difference

 $\sigma_{c} = \sigma_{total} - \sigma_{pot} - \sigma_{f}$

from these data (resonance scattering was neglected), no reasonable information about σ_c can be obtained. This is not surprising because of the small value of σ_c in comparison with σ_{total} and the large error for each experimental result. But, the result may be an indication that in this energy range α is probably smaller than the numbers given by Yeater et al. [24].

Energy range	<o<sub>total></o<sub>	<apre>capot></apre>	<σ _f >	
(eV)		(b)		
46.5-100	52.73	13.0	42.2	
100-215	40.84	13.0	27.9	
215-465	31.84	13.0	23.4	
465-1000	27.54	13.0	14.3	
1000-2150	22.30	12.0	9.51	
2150-4650	19.04	11.0	6.79	
4650-10 000	17.93	9.3	5.25	

TABLE VIII. INTEGRATED σ_{total} , σ_{pot} , AND σ_{f} FOR ²³³U (1/E-WEIGHTING)

3.2.2. Resonance parameters

Calculations with TRIX-1 led to the recommended resonance parameters presented in Table IX and Fig.1. The resulting partial resonance integrals are given in Tables V and VII for intercomparison with the evaluated cross-sections. Parameters for 31 resolved resonances up to an energy of 37 eV have been taken from BNL-325 [4] (recommended data). The average value of 54 meV for $\langle \Gamma_{\gamma} \rangle$ results from an unweighted average of the Γ_{γ} -values for the 31 resolved resonances. $\langle D \rangle = 0.87$ eV is based on a value given by Michaudon [26]. The value $\langle \Gamma_n^0 \rangle = 0.18 \times 10^{-3}$ is somewhat smaller than the average value of Γ_n^0 (0.225×10^{-3}) resulting from the 31 resolved resonances. This smaller value was chosen to match the fission cross-section in the unresolved region. The same criterion led to the recommended energy dependence of the average fission width. In the limit of low energies, the value of $\langle \Gamma_f \rangle$ approaches the average $\langle \Gamma_{\epsilon} \rangle$ from the 31 resolved resonances.

TABLE IX. RECOMMENDED RESONANCE PARAMETERS FOR $^{233}\mathrm{U}$

Number of resolved resonances: 31 (Parameters as given by BNL-325 [4])

Parameters for negative energy resonance:

/E/ = 0.3 eV g = 0.5 $\Gamma_{n}^{0} = 0.06 \times 10^{-3} \text{ eV}^{\frac{1}{2}}$ $\Gamma_{\gamma} = 45 \text{ meV}$ $\Gamma_{f} = 960 \text{ meV}$

Average parameters for unresolved resonances:

 $<\Gamma_{n}^{0} > = 0.18 \times 10^{-3} \text{ eV}^{\frac{1}{2}}$ $<\Gamma_{\gamma} > = 54 \text{ meV}$ <D > = 0.87 eV $S_{1} = 1.5 \times 10^{-4}$ $<\Gamma_{f} > = 0.09 \times \log_{10} \text{ E} + 0.72 \text{ (0.00036 < E < 0.00125)}$ $<\Gamma_{f} > = 0.2233 \times \log_{10} \text{ E} + 1.109 \text{ (0.00125 < E < 0.01)}$ $(<\Gamma_{f} > \text{ in eV}; \text{ E in MeV})$

3.3. ²³⁹Pu

3.3.1. Cross-sections

In Table X, partial fission and capture resonance integrals are given, which were calculated either by integration of differential cross-sections or by TRIX-1 calculations using different sets of parameters for the resolved resonances.

Unlike 235 U and 233 U, the calculation of resonance integrals was performed only for energies above 4.65 eV. The structure of the crosssection below this energy is dominated by the strong resonance at 0.296 eV.

TABLE X. PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS FOR ²³⁹Pu. INTEGRATED CROSS-SECTIONS AND INTEGRALS CALCULATED WITH THE TRIX-1 CODE USING DIFFERENT SETS OF RESONANCE PARAMETERS

	Fission resonance integrals (b)					Capture resonance integrals (b)		
Energy range (eV)	Integrated cross-sections TRIX-1 calculations parameters from :		lculations s from :	Integrated cross- sections	TRIX-1 calculations parameters from:			
	Karlsruhe	BNL 325	LASL	Karlsruhe	BNL	Karlsruhe	Karlsruhe	BNL
Ref.:	[27]	[4]	[22]	[16]	[4]	[27]	[16]	[4]
4.65-10	26.9	22.3		26.80	26.38	22.90	23.71	24.55
10 - 21,5	76.4 ^a	81.7		83.15	75.61	53,40 ^a	51.84	53.57
21,5-46.5	16.5	19.8	20.9	18.03	15.83	23.80	25.59	24.36
46,5-100	40.8	31.8	42.1	39,27	30,11	40.40	20.72	29.06
100-215	14.1	14.8	13.9	14,99		10.40	9,00	
215-465	10.0	10.0	9.66	9.30		6.92	6.20	
465-1000	5.80	6.00	5.94	6.03		3.78	3.40	
1000-2150	3.11	3.50	2.90	3.95		1.84	2.17	
2150 -4650	2.51	2.22	2.04	2.53		1.36	1.47	
4650-10 000	1,88	1.83	1.76	1.79		0.92	0.80	

a Aldermaston data **CN-**23/5

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Therefore small errors in the determination of the Cd cutoff energy may lead to large errors in the measured total resonance integral. Therefore, a comparison of experimental total resonance integrals with values calculated with TRIX-1 does not provide a critical test for the calculation method and the resonance parameters used.

For this study, cross-section information was available from the evaluated data of Schmidt [27], from the recommended data in BNL-325 [4], and from recent time-of-flight measurements performed with an underground nuclear explosion by Hemmendinger [22]. Partial resonance integrals found by integration of these data are presented in Table X. Resonance integrals were calculated from the constants of the Karlsruhe 26-group set. These numbers were recently published by Pasquer et al. [28] and compared with the group constants published by Bondarenko [23] and those used at Aldermaston [29]. The Aldermaston average crosssection has been used for the group 10-21 eV instead of the Karlsruhe value, which is about 10% lower than the Aldermaston result. Probably the data presented by Pasquer [28] did not contain Schmidt's [30] corrected value of Γ_n for the resonance at 14.68 eV. The results of Hemmendinger [22] are in good agreement with the other data up to 1-keV energy; especially good agreement is found with the evaluated Schmidt data, except in the group 21.5-46.5 eV. The Los Alamos data confirm the high value assumed by Schmidt in the group 46.5-100 eV. Between 1 and 10 keV the LASL-data are about 10% below the Schmidt values.

While the Karlsruhe group constants are based on information published before 1963, BNL-325 [4] contains some more recent data. But unfortunately three new measurements, which have very good energy resolution, were not yet taken into account. In Saclay, the fission crosssection has been measured by de Saussure et al. [31] between 0.16 eV and 5 keV. In Harwell, different σ_f measurements were done, which have been partially reported by James [32]. The most recent time-of-flight measurements, which were already mentioned, are those of Hemmendinger [22]. James and Endacott [33] mention that their results for σ_f between 1 and 8 keV agree within 5% with the Karlsruhe evaluated data [27]. The curve published by James [32] for σ_f between 50 and 100 eV shows good agreement with the older data of Bollinger [34], which are the basis of the Karlsruhe evaluation.

Direct measurements of the capture cross-section do not exist. This cross-section is calculated from σ_{total} , σ_{nn} , and α or η and ν measurements. Therefore, the uncertainties in the reported capture crosssections are large. Since the individual cross-sections are mostly measured by different laboratories with different experimental energy resolution, it is difficult to produce a consistent set of data. As the α -values are only poorly known [30], the capture cross-section has very large uncertainties in the resonance region. Evaluated data of Schmidt [27] are given in Table X. Total resonance integral measurements are available for fission only. They have recently been evaluated by Feiner and Esch [12], resulting in RI_f = 310 ± 20 b for E_{Cd} = 0.5 eV.

Based on the available information, the data given in Table XI are found as recommended data. Partial fission resonance integrals between 4.65 eV and 46.5 eV have been taken from BNL-325, while at higher energies the new LASL data are believed to be reasonable. In spite of the difficulties in comparing total resonance integrals, one may compare
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TABLE XI. RECOMMENDED PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS AND DERIVED GROUP CONSTANTS FOR ²³⁹Pu, AND RESONANCE INTEGRALS CALCULATED BY TRIX-1 WITH THE RECOMMENDED RESONANCE PARAMETERS

Energy range (eV)	Recommended resonance integrals and group constants (b)				Resonance integrals from TRIX-1 with recommended parameters	
	(RLf)i	<of>g</of>	(RI _c) _i	<σ _c >	(RL)i	(RI _c)i
4.65-10	22.3	28.9	23.71	30.8	26,80	23.71
10-21.5	81.7	106.1	51.84	67.4	83.15	51.84
21.5-46.5	19.8	25.7	25.59	33.2	18.03	25,59
46.5-100	42.1	54.6	20.72	26.9	39.27	20.72
100-215	13.9	18.1	9.00	11.7	14.99	9,00
215-465	9.66	12.5	6.20	8.06	9.30	6.20
465-1000	5.94	7.72	3.40	4.41	6.03	3.40
1000-2150	2.90	3.76	2.17	2.82	3.95	2.17
2150-4650	2.04	2.64	1.47	1.91	2.53	1.47
4650-10000	1.76	2.28	0.80	1.04	1.79	0.80
	202.10		744.90			

the fission integral which results from a summation of the recommended partial integrals in Table XI with the experimental results $(310 \pm 20 \text{ b})$. To do that one has to add the partial resonance integrals for energies 0.5-4.65 eV and >10 keV to the value of 202 b. With the values of Schmidt [30], one gets 89.7 b for 0.5-4.65 eV and 9 b for E >10 keV. With these numbers a total fission resonance integral of 301 b results for energies >0.5 eV.

For further evaluation of the fission cross-section below 1 keV one may use the measurements of Hellstrand [35], who measured the fission resonance integral in a 1/E flux with boron filters of different thickness. For the thin filter 50% of all fission occurs above 125 eV while for the thick filter the corresponding number of 750 eV. Hellstrand compares his experimental results with calculations based on measured transmission curves and the microscopic fission cross-section given by Schmidt in KFK 120 [27]. While reasonable agreement is found for the thin filter, the calculated values for the thick filter are 10% high. This discrepancy would no longer exist if the new LASL data would be used. The difference between the two measurements with the thin and the thick filter no longer contains the quite uncertain contribution of higher energies to the resonance integrals but is mostly formed by neutrons between 20 and 200 eV energy. While from the experiment a difference of 32.9 b is found a calculation with the Schmidt cross-sections yields 30.6 b. As the LASL data in this energy region are about 6% higher than the Schmidt

data, again better agreement with the Hellstrand experiment is obtained with the new LASL results.

Because of the assigned errors and the integral character of Hellstrand's experiment not too much weight should be given to the agreement found. But as no better possibility for solving the discrepancies in the ²³⁹Pu resonance fission cross-section is available, this agreement is taken as a basis for preferring the LASL data.

Since no better experimental data are available for the capture cross-section, the results of a TRIX-1 calculation with the recommended resonance parameters (see 3.3.2) have been taken without change as the recommended partial resonance integrals and group constants (Table XI). These data result in reasonable α -values and the total of $\sigma_f + \sigma_c + \sigma_n$ is in general in good agreement with the total cross-section as measured by Uttley [36]. But it must be pointed out that this comparison gives little information about the exact capture cross-section above 1 keV because the scattering cross-section is so much larger than the capture cross-section.

3.3.2. Resonance parameters

The most complete set of evaluated resonance parameters was recently presented by Schmidt [16] at the ANS-meeting in San Diego, Feb.1966. These extend the resolved region in 239 Pu to 300 eV, due to incorporation of the results of Saclay above 50 eV and those of Saclay and Harwell between 100 and 300 eV.

TRIX-1 calculations with these parameters recommended by Schmidt have been performed. Results are presented in Tables X and XI. The only change to the Schmidt parameters is a consideration of new spin assignments and resulting new Γ_n -values for 15 resonances at low energies, which follow from the recent scattering experiments of Bowman and Sauter at Livermore [37]. For intercomparison a TRIX-1 calculation was performed using the parameters recommended in BNL-325 [4] for 19 resonances below 53 eV in combination with the latest available parameters from Saclay [31] for five resonances between 50 and 100 eV (see Table X).

For the average resonance parameters for the unresolved resonances above 300 eV, the recommended parameters listed in Table XII have been used. These have been chosen to match the measured differential crosssections. The values for $\langle D \rangle$ and $\langle \Gamma_{\gamma} \rangle$ were taken as recommended by Schmidt [16]. The S₀-strength function used is 15% smaller than the Schmidt value. $\langle \Gamma_{f} \rangle$ = 1.5 eV for the J=0 state has been chosen as energy independent, while $\langle \Gamma_{f} \rangle$ for the J=1 state has been assumed energy dependent as given by the correlation function of Table XII and Fig.1.

Very broad fission resonances had been found in Harwell by Uttley. A direct separation of the fission widths found according to the two J-series is not possible from that experiment as no spin assignments for the resonances in that region exist. But making the assumption that broad resonances belong to J=0 states and small resonances to J=1 states, one can reach average values for $\langle \Gamma_f \rangle_J$ from the measured parameters. The "boundary" between both distributions has to be viewed

TABLE XII. RECOMMENDED RESONANCE PARAMETERS FOR $^{239}\mathrm{Pu}$

Number of resolved resonances: 124 (Parameters as given by Schmidt [16] and Bowman and Sauter [37]).

Negative energy resonance parameters:

$$/E/$$
 = 1.2 eV
g = 0.75
 Γ_n^0 = 0.771×10⁻³ eV²
 Γ_y = 39 meV
 Γ_f = 201 meV

Average parameters for unresolved resonances:

$$\langle \Gamma_{n}^{0} \rangle_{J=0} = 0.797 \times 10^{-3}$$

$$\langle \Gamma_{n}^{0} \rangle_{J=1} = 0.284 \times 10^{-3}$$

$$\langle \Gamma_{y} \rangle = 38.7 \text{ meV}$$

$$\langle D \rangle_{J=0} = 8.78 \text{ eV}$$

$$\langle D \rangle_{J=1} = 3.12 \text{ eV}$$

$$S_{1} = 2.5 \times 10^{-4}$$

$$\langle \Gamma_{f} \rangle_{J=0} = 1.5 \text{ eV}$$

$$\langle \Gamma_{f} \rangle_{J=1} = 0.0075 \times \log_{10} \text{ E+ } 0.0762$$

$$(\langle \Gamma_{f} \rangle \text{ in eV, E in MeV})$$

$$\text{ valid for 300 eV} \langle E \langle 10 \text{ keV}$$

as rather arbitrary. Doing this with a boundary at 300 meV, Schmidt [16] obtains:

$$\langle \Gamma_{f} \rangle_{J=0} = 1.3 \text{ eV} \quad \langle \Gamma_{f} \rangle_{J=1} = 65 \text{ meV}.$$

He compares this $\langle \Gamma_f \rangle_{J=0}$ value with 2.8 eV, which follows from the channel theory of fission. With their spin assignments for 15 resonances at low energies (3 J = 0; 12 J = 1) Bowman and Sauter [37] find from the fission widths of the Saclay measurements [31] the following averages:

 $\langle \Gamma_f \rangle_{J=0} = 403 \text{ meV}; \quad \langle \Gamma_f \rangle_{J=1} = 41.7 \text{ meV}.$

Based on these experimental results Yiftah et al. [38] have tried a new splitting of the Uttley fission widths using a boundary of 150 meV. In this way they find $\langle \Gamma_f \rangle_{J=0} = 1 \text{ eV}$. Pitterle [39] has made some general Doppler effect calculations for

Pitterle [39] has made some general Doppler effect calculations for 239 Pu/ 238 U mixtures using different distributions for the fission widths in 239 Pu. His results show that for a 238 U/ 239 Pu = 5:1 mixture the Pu Doppler effect may be reduced to one half, using very broad $\langle \Gamma_f \rangle_{J=0}$ values (1.5 eV); compared to those calculated with the assumption $\langle \Gamma_f \rangle_{J=0} = \langle \Gamma_f \rangle_{J=1}$. He concludes that the broad fission widths will reduce the Doppler effects for the heated Pu samples in the ANL criticals quite substantially, but that even $\langle \Gamma_f \rangle_{J=0} = 1.5$ eV will not be sufficient to reach agreement with experimental results.

3.4. ²⁴¹Pu

3.4.1. Cross-sections

Very few measurements of the fission cross-section of 241 Pu have been made BNL-325 [4] contains the measurements of Moore et al. [40] up to 100 eV and those of James [41] up to 2.5 keV. In addition, the results of the measurements with a nuclear explosion (PETREL) were available. Results from this experiment in the energy range 20-200 eV were recently published by Simpson et al. [42], while at higher energies preliminary results were received from Hemmendinger [22]. The recommended curve of BNL-325 [4] and the results from the PETREL event [42, 22] were weighted with a 1/E spectrum. Partial resonance integrals found in this manner are given in Table XIII. Integrals calculated from the Bondarenko [23] group constants are given for comparison. New measurements have been performed at Harwell by James and Endacott [43] between 1 eV and 30 keV. But so far only linearly averaged data for broad energy groups have been published [43].

Experimental results for the total fission resonance integral are presented in Table XIV. No differential measurements exist for the capture cross-section, nor has α been measured as a function of energy.

Based on this information recommended resonance integrals and group constants were chosen, as listed in Table XV. For fission the data from BNL-325 [4] have been used up to 2.15 keV. These partial fission resonance integrals result in an integral of 603 b for the energy range 0.5 eV-10 keV. Adding 14 b for energies above 10 keV (from Bondarenko), one gets 617 b for the total resonance integral. This value is in reasonable agreement with the most recent measured value of Hardy et al. [46] of 581 ± 33 b. As no experimental results for the capture cross-section are available, the results of a TRIX-1 calculation using the parameters given in section 3.4.2 are taken as recommended integrals and group constants, respectively (Table XV). The energy range, 0.5 eV to 10 keV, gives a capture resonance integral of 181 b.

3.4.2. Resonance parameters

A TRIX-1 calculation with the recommended parameters presented in Table XVI was performed. The results are given in Tables XIII and XV.

Parameters for 23 resolved resonances up to 35 eV have been taken from BNL-325 [4]. The fission resonance integrals at low energies be-

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	Integ	70.07 1		
Energy range (eV)	Bondarenko	BNL-325	Petrel	calculation BNL parameters
Ref.:	[23]	[4]	[42, 22]	
0.5-1	34.65 ⁸	29.07		79.07
1-2.15	26,95	23.39		37.54
2.15-4.65	61.60	89.80		88.55
4.65-10	169.40	193.60	·	181.78
10-21.5	100.10	106.34		120.16
21.5-46.5	46.20	52.85	52.44	59.93
46.5-100	30.80	41.32	39.93	37.41
100-215	23,10	20,65	21, 55 ^b	25,14
215-465	16.17	19.12	20.10	17.24
465-1000	12.32	9. 98	9.49	11.39
1000-2150	9.24	7.15	6.21	7.61
2150-4650	6,93		4.65	5.06
4650-10000	5.00		3.60	3.31
	1			[

TABLE XIII. PARTIAL FISSION RESONANCE INTEGRALS FOR ²⁴¹ Pu. INTEGRATED CROSS-SECTIONS AND INTEGRALS CALCULATED WITH THE TRIX-1 CODE.

a 0.465 - 1 eV group

^b 100-194 eV only

tween 0.5 and 2.15 eV are much higher than those found by integration of the BNL-325 cross-sections. This is due to the strong influence of the resonance at 0.26 eV.

The average $\langle D \rangle = 1.17 \text{ eV}$ for the unresolved resonances was calculated from the 43 resonances which are given in BNL-325 [4] below 50.4 eV. This value is in agreement with $D = 1.3 \pm 0.2 \text{ eV}$ given by James and Endacott [33]. $\Gamma_{\gamma} = 40 \text{ meV}$ is based on the information contained in BNL-325 for the resolved resonances. $\langle \Gamma_n^0 \rangle$ and $\langle \Gamma_f \rangle$ as a function of energy were chosen, so that the TRIX-1 results gave good agreement with the experimental cross-sections.

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Reference	Cadmium cutoff (eV)	Fission resonance integral (b)
Bigham [44]	0.45	532±16
Hardy et al. [45]	0.45	565 ± 33
Hardy et al. [46]	0.5	581 ± 33

TABLE XIV. EXPERIMENTAL RESULTS FOR THE FISSION RESONANCE INTEGRAL OF $^{241}\mathrm{Pu}$

4. CONCLUSIONS

An evaluation of the available experimental data for the fission and capture cross-sections of the fissile nuclei in the resonance region shows that there still exist large uncertainties. Even for the most intensively studied nucleus, ²³⁵U, the uncertainties in the fission cross-section are about $\pm 5\%$ and in the capture cross-section $\pm 20\%$. For ²³⁹Pu, $\sigma_{\rm f}$ is known to $\pm 10\%$ and $\sigma_{\rm c}$ to $\pm 25\%$. For ²³³U and ²⁴¹Pu, the uncertainty in $\sigma_{\rm f}$ is $\pm 20\%$ and in $\sigma_{\rm c} \pm 30\%$. It is anticipated that when the measured data of recent experiments have been more fully analysed, the accuracy to which present data is known will be considerably improved.

The resolved energy region for the fissile nuclides considered in the present analysis is in general below 300 eV. Thus, the analysis of fast reactor Doppler coefficients must still make use of averaged parameters for the unresolved resonance region.

In the resolved energy region, the infinitely dilute resonance fission cross-sections calculated by TRIX-1, using the single-level Breit-Wigner model, were 10-20% smaller than the corresponding measured data. In the unresolved region the average parameters were so chosen that the calculated fission cross-sections were in satisfactory agreement with measurements.

From similar calculations, it was determined that the infinitely dilute capture cross-sections in the resolved energy region agree more closely with measured data than do the fission cross-sections. In the unresolved energy region, there is a singular lack of measured capture cross-section data; therefore, the recommended cross-section values quoted in this report were obtained from TRIX-1 calculations.

Although there is reasonably good agreement between the measured cross-section data and the calculated cross-section data, using the singlelevel formula, one must not assume that similar agreement exists for the calculated Doppler effects, particularly if the calculation is performed using temperature-dependent cross-sections obtained from the singlelevel analysis.

Lynn [47] has analysed the resonance structure of these fissile nuclei and his results clearly show the strong influence of interference

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TABLE XV. RECOMMENDED PARTIAL FISSION AND CAPTURE RESONANCE INTEGRALS AND DERIVED GROUP CONSTANTS FOR ²⁴¹ Pu, AND RESONANCE INTEGRALS CALCULATED BY TRIX-1 WITH THE RECOMMENDED PARAMETERS

Energy range (eV)	Recommended resonance integrals and group constants (b)				Resonance integrals from TRIX-1 with recommended parameters	
	(RI _f)	<or f="">g</or>	(RI _c) _i	<0°, >g	(RI _f) _i	(RI _c) _i
0.5-1	29.07	42.13	27.04	39.19	79.07	27.04
1-2.15	23.39	30.38	9.15	11.88	37.54	9.15
2.15-4.65	89.80	116.62	47.93	62.25	88.55	47.93
4.65-10	193.60	251.42	26.09	33.88	181.78	26.09
10-21.5	106.34	138.10	39.47	51,26	120.16	39.47
21.5-46.5	52.85	68.64	9.99	12.97	59.93	9.99
46.5-100	41.32	53.66	7.92	10.29	37.41	7.92
100-215	20.65	26,82	5.17	6. 71	25.14	5.17
215-465	19.12	24.83	3.33	4.32	17.24	3.33
465 -1 000	9.98	12.96	2.07	2.69	11.39	2,07
1000-2150	7.15	9.29	1.33	1.73	7.61	1.33
2150-4650	5.20	6.75	0.85	1.10	5.06	0.85
4650-10000	4.10	5.32	0.52	0.68	3.31	0.52
	602.57		180,86		674.19	180,86

effects between neighbouring levels of the same isotope. This interference between levels modifies the temperature-dependent crosssections and very different results may be obtained for calculated Doppler 'effects.

Our knowledge of the resonance cross-sections of the fissile nuclei is not good enough to make reliable calculations of the Doppler effect component of these isotopes in fast reactors. Unfortunately, even better experimental resolution will not extend the upper limit of the resolved resonance region far enough because of the Doppler-broadening of the resonances. Therefore we have to rely on measurements of effective (self-shielded) cross-sections, as for instance performed on bulk Pu by Bramblett and Czirr [48].

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TABLE XVI.RECOMMENDED RESONANCE PARAMETERSFOR 241Pu

Number of resolved resonances: 23 . (parameters as given by BNL-325 [4])

Negative energy resonance parameters:

/E/ = 0.16 eV g = 0.5 $\Gamma_{n}^{0} = 0.0725 \times 10^{-3} \text{ eV}^{\frac{1}{2}}$ $\Gamma_{\gamma} = 40.0 \text{ meV}$ $\Gamma_{f} = 60 \text{ meV}$

Average parameters for unresolved resonances:

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DISCUSSION

J.J. SCHMIDT: I would like to comment on the large ²³⁹Pu alpha values found recently at Harwell for energies between 1 and 10 keV. Discussions during this Conference with Dr. Sowerby and others have shown that, if the Harwell experimentalists had taken the statistical fluctuation factors to be ≤ 3 in estimating the average resonance scattering cross-section, instead of assuming them to be equal to unity, the alpha values would have been much lower, agreeing with the generally accepted values based on the earlier KAPL (Knolls Atomic Power Laboratory) measurements in this range.

CONTRIBUTIONS OF NEGATIVE ENERGY AND DISTANT RESONANCES IN THE RESOLVED RESONANCE REGION

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Abstract

CONTRIBUTIONS OF NEGATIVE ENERGY AND DISTANT RESONANCES IN THE RESOLVED RESONANCE REGION. Extensive sets of resonance parameters are becoming available for the neutron cross-sections of many nuclides, from which the partial cross-sections can be computed, over a wide energy range, by using a multilevel resonance formula. To obtain reliable results it is necessary to allow for the effects of negative energy resonances and of unresolved positive energy resonances.

The first part of this paper describes briefly the theory of such effects. It is possible to calculate numerically the expected contributions to the capture cross-section and the effective channel radii from unresolved resonances, assuming that the statistical distributions and mean values of the resonance parameters are known. Approximate formulae for these contributions are given. The standard deviations of the contributions may also be calculated; these are large if the difference between the energy at which the contributions are calculated and the energy at which the unresolved region is assumed to begin is much less than the mean spacing between resonances. All negative energy resonances could be treated in this statistical manner, but because both the expected contributions and their standard deviations at low energies (e.g. around 0.0253 eV) will then be very large, it is preferable to fix the parameters of the first negative energy resonances, and to treat only the relatively small contributions from the more distant negative energy resonances statistically.

Methods are therefore discussed for the estimation of the widths and energy of the first negative energy resonance. (If two spin-states of the compound nucleus may be excited by s-wave neutrons, two such resonances need to be considered.) The parameters may be determined quite closely by the values and energy dependence of the low-energy cross-sections. However, for some nuclides, particularly if this resonance is small or relatively distant, it is not possible to calculate them so definitely; additional arguments based on the statistical distributions of widths and spacings are then used to select reasonable values for the required parameters.

The second part of the paper applies these methods to the low-energy cross-sections of the isotopes of nickel.

1. Introduction

The physicist who has to evaluate nuclear cross-section data for reactor calculations must frequently extrapolate from measured information. He often has to consider the following situation: the total cross-section has been measured over a particular energy range, and resonances have been resolved and analysed in that range. He now wishes to use the resonance parameters to generate the total cross-section at low energies (particularly around thermal energies), where measurements may have been sparse, and also to compute the (n,γ) cross-section, which may only have been measured at one energy (usually 0.025 eV), or over a thermal spectrum. The difficulty in doing this in a straightforward way arises from the effects of the unresolved resonances, which contribute a background that is energy dependent, albeit weakly so. These unresolved resonances are at high positive energies, and at negative energies; of which the nearby energy resonances are likely to have the greatest effect on low energy cross-sections. In particular, the topmost negative energy resonance may

well have a greater effect on the thermal cross-section than the first positive resolved resonance.

If sufficient data are available, it may be possible to allow for the contributions from unresolved resonances by an empirical formulae fitted to the data; this has been done by for example Firk et al. [1] and Coté et al. [2]. Often, however, data are too sparse for this to be done, and then it is necessary to try to estimate the contributions. Such estimates must rely mainly on statistical methods, since the only information that is available about the unresolved resources is of a statistical nature; that is, the distributions and means of the widths and spacings The plan that is outlined in this paper consists in of resonances. trying to choose reasonable parameters for the first negative energy resonance (because of its pre-eminent importance) and then making a statistical estimate of the contributions from the remaining unresolved resonances. Of course we must recognise that for any real nuclide our estimates may be far from the truth, if the nuclide is not statistically "typical" - if, for example, the first few negative resonances have abnormally large or small widths and spacings - but this is a disadvantage from which any statistical treatment must suffer when applied to single members of an ensemble - in this case to a small isolated energy region of a particular nuclide. We try whenever possible to indicate the amount of confidence to be placed in the results; but because of this uncertainty it is important to use what data are available to determine as many parameters as possible, and only to rely on statistical arguments when there is no alternative.

2. Contributions from unresolved resonances

For brevity a number of simplifying assumptions will be made; these will not be very restrictive, and it is not difficult to consider more general cases. First, the target nucleus will be assumed to be nonfissile. Secondly, the multilevel Breit-Wigner formulae will be used, and only s-wave neutrons will be considered. Thirdly, the reduced neutron widths I_n^{O} of the resonances and the spacings S between neighbouring resonances of the same spin and parity are assumed to be independent random variables and to have respectively a Porter-Thomas [3] and a Wigner [4] distribution:

	Ρ(η) dr) =	[exp(-ŋ/2)]/√(2πŋ)	}	/~ · · ·
where	T) =	$r_n^{\circ}/\bar{r}_n^{\circ}$,	J	(2.1)
and	\$(\$) d	5 =	$\frac{1}{2}\pi\xi \exp(-\pi\xi^2/4) d\xi$	7	(a a)
where	Ę	. =	s/D .	}	(2.2)

In these equations, $\overline{\Gamma_n}^o$ is the mean reduced neutron width, and $D = \overline{S}$ is the mean spacing.

The capture widths Γ_v of all resonances are assumed to be constant.

If the lowest and highest resolved resonances of a given spin and parity are at E_L and E_H , the energies of the unresolved resonances of the same spin and parity below E_L and above E_H form renewal processes [5], and the expected density of resonances at an energy xD below E_L or above E_H is $\rho(\mathbf{x})$, which satisfies the renewal equation:-

$$\rho(x) = \phi(x) + \int_{0}^{x} \phi(x - y) \rho(y) \, dy \qquad (2.3)$$

where $\phi(\mathbf{x})$ is the Wigner distribution (2.2).

Equation (2.3) may be solved numerically quite easily, and a table and graph may be found in, for example, a paper by Reichel and Wilkins [6]. The property of $\rho(\mathbf{x})$ that is of most importance to us is that

$$\rho(\mathbf{x}) \rightarrow 1 \text{ as } \mathbf{x} \rightarrow \infty;$$

and in fact, $\rho(\mathbf{x}) \approx 1$ for $\mathbf{x} \ge 2$. An approximation that is sufficiently accurate for many purposes is:

 $\rho(\mathbf{x}) \approx \rho_{a}(\mathbf{x})$ where $\rho_{a}(\mathbf{x}) = \mathbf{x}/c$ for $\mathbf{x} \le c$ (2.4) = 1 for $\mathbf{x} > c$, and $c = 2 - 4/\pi = 0.7268$.

With these preliminaries completed, we may write down the elastic scattering and capture cross-sections at a neutron energy E eV:-

$$\sigma_{nn}(\mathbf{E}) = \begin{cases} \xi_{\mathbf{J}} \sigma_{nn}^{\mathbf{J}}(\mathbf{E}) \\ \sigma_{\mathbf{n}\gamma}(\mathbf{E}) = \begin{cases} \xi_{\mathbf{J}} \sigma_{\mathbf{n}\gamma}^{\mathbf{J}}(\mathbf{E}) \\ \xi_{\mathbf{J}} \sigma_{\mathbf{n}\gamma}^{\mathbf{J}}(\mathbf{E}) \end{cases} \\ g_{\mathbf{I}} = (2J+1)/2(2I+1) \end{cases}$$
(2.5)

I being the spin of the target nucleus, and J that of the compound nucleus. In Eqs. (2.5)

$$\sigma_{nn}^{J}(E) \approx 4\pi \left| a_{eff}(E) - \frac{1}{2k_{o}} \sum_{\mathbf{r}} \frac{\Gamma_{\mathbf{r}n}}{E_{\mathbf{r}} - E - i\Gamma_{\mathbf{r}}/2} \right|^{2}$$
(2.6)

$$\sqrt{E} \sigma_{n\gamma}^{J}(E) \approx \frac{\pi}{k_{o}^{2}} \left[\frac{\Gamma}{r} \frac{\Gamma_{n\gamma}^{o} \Gamma_{n\gamma}^{o}}{(E_{r} - E)^{2} + \Gamma_{r}^{2}/4} + \frac{\overline{\Gamma}_{r} \overline{\Gamma}_{n}^{o}}{D^{2}} \mathfrak{I}(E) \right]. \quad (2.7)$$

In Eqs. (2.6) and (2.7) all the quantities are those appropriate to the particular spin J. E is the neutron energy (lab. system) in electron-volts, and k_0 is the wavenumber in the centre-of-mass system of a 1 eV neutron:-

$$k_0 = 2.19685 \frac{A}{A+1.008665} \times 10^{-4} \text{ fm}^{-1}$$

where A is the atomic weight of the target nucleus (on the $c^{12} = 12$ scale).

 Γ_{rrn}° , Γ_{rr}° , and $\Gamma_{r}^{\circ} = \Gamma_{rrn}^{\circ} \sqrt{E} + \Gamma_{rr}^{\circ}$ are the reduced neutron width, the capture width, and the total width, of the resonance at E_{r} . All these energies are assumed to be referred to the laboratory system.

The sums over r are over all the resolved resonances of the given spin. The contributions from the unresolved resonances are included in $a_{eff}(E)$, the effective channel radius, and X(E).

$$\mathbf{a}_{eff}(\mathbf{E}) = \mathbf{a} - \frac{1}{2\mathbf{k}_{o}} \quad \overline{\mathbf{F}_{n}}^{o} \quad \mathbf{Y}(\mathbf{E}) , \qquad (2.8)$$

,

where a fm is the constant channel radius, and Y(E) is a sum over unresolved resonances.

To evaluate X(E) and Y(E), it is assumed that they are equal to the expected values of the contributions from unresolved levels; use of the approximation (2.4) then gives:-

$$X(E) = \alpha \left(\frac{E_{H} - E}{D} \right) + \alpha \left(\frac{E - E_{L}}{D} \right)$$
(2.9)

with

$$\alpha(u) \approx 1.38 \ln(1 + 0.727/u)$$
, (2.10)

$$I Y(E) = \beta \left(\frac{E_H - E}{D} \right) - \beta \left(\frac{E - E_L}{D} \right) (2.11)$$

with
$$\beta(u) \approx 1.38 [(0.727 + u) \ln(0.727 + u) - u \ln u]. (2.12)$$

In the right hand sides of Eqs. (2.9) and (2.11), the first terms give contributions from resonances above $E_{\rm H}$, while the second give contributions from those below $E_{\rm L}$. It has been assumed that $(E_{\rm H} - E)$ and $(E - E_{\rm L})$ are both much greater than the mean total width $(\Gamma_{\rm Y} + \overline{\Gamma_{\rm n}}^{\circ} \sqrt{E})$.

Two further assumptions are implicit in these equations: it is assumed that the neutron energy E is low, so that $k_0 \sqrt{E} \ a <<1$, and any variation in the strength function $(\overline{\Gamma_n}^0/D)$ over the energy range EL to E_H is neglected. More general results have been obtained for cases in which these assumptions are not made.

The standard deviations of the contributions are obviously needed and may be found from general formulae given in [6] and [7]. For the contribution $\alpha(u)$ to the capture cross-section, the fractional standard deviation $\approx 1/\sqrt{u}$ for u >> 1. Thus, even at several mean spacings from EL and EH, there is considerable inherent uncertainty in the estimation of the contribution (e.g. a standard deviation of about 30% of the expected value at 9 mean spacings from either of the end points). If $u \leq 1$, numerical quadrature must be used: for the particular case u = 0.34 a calculation gave a standard deviation about 25% greater than the expected value. This result indicates that not only is the uncertainty in this range very great, but also the probability distribution is markedly skew, so that the expected value (given approximately by Eq. (2.10)) is not equal to the most probable value. Ideally, the most probable value is a better estimate than the expected value, but it is also a much more difficult quantity to compute. Formulae given elsewhere [7] could be used to calculate it (and indeed the whole probability distribution) but no numerical computations have yet been made. Qualitatively, the most probable value of $\alpha(u)$ should be less than the expected value, but should approach it asymptotically for large u.

The standard deviation of the contribution to the effective nuclear radius has not yet been calculated; but there is no reason to suppose that it is any smaller than that for the contribution to σ_{nY} .

Because of this large statistical uncertainty, it is desirable to increase u; that is, to make EL as small, and EH as large, as possible. For low energy neutrons, the predominant contribution comes from negative energy resonances, and so EL is the important quantity. If the crosssections are being calculated below the lowest positive energy resonance, this means that it is important to calculate the parameters of the first negative energy resonance as accurately as possible, and to treat statistically the contributions from the remaining bound levels. Note that E_L and E_H are here the energies of actual resonances: the theory may be extended to cover the case where they are arbitrary energies, but the results are more complicated, and no more useful.

Finally, it is worth pointing out some general properties of the contributions Y(E) and X(E) to the effective radius and capture cross-sections. First, the effective radius is a monotonically decreasing function of neutron energy and, if the target nucleus has non-zero spin, this radius will be different for the two possible values of J. Secondly, provided $|E - E_L| >> \overline{\Gamma_{Y}} + \overline{\Gamma_n} \sqrt{E}$ and $|E_H - E| >> \overline{\Gamma_{Y}} + \overline{\Gamma_n} \sqrt{E}$,

$$X(E) = -dY(E)/dE$$
(2.13)

3. The calculation of the parameters of the first negative energy resonance

For simplicity we first consider the case of a target nucleus of zero spin, so that only one value of J is involved.

Suppose that the first negative energy resonance is at $E_L \in V$ ($E_L < 0$), and that its reduced neutron and capture widths are Γ_{Ln}^{0} and $\Gamma_{L\gamma}^{r}$ respectively. Usually we shall assume $\Gamma_{L\gamma} = \overline{\Gamma_{\gamma}}$, unless there is enough data to enable some other choice to be made; frequently, too, we shall have to assume for want of any more detailed information that all resonances have the same capture width $\overline{\Gamma_{\gamma}}$, and indeed this mean value may itself be one of the quantities that need to be determined.

The procedure is, at least in principle, iterative. A value is chosen for E_L , by (for example) assuming that the interval between it and the lowest positive energy resonance is equal to the mean spacing, D, or (better) the most probable spacing 0.8D (provided the chosen estimate gives a negative value for E_L). The contributions from negative energy resonances beyond E_L may then be estimated from the formulae of the previous Section. The contributions from resolved and unresolved positive energy resonances may also be computed, so that finally, by subtracting these from measured quantities, the contributions of the resonance at E_L may be found:-

$$\Gamma_{Ln}^{\circ} \Gamma_{L\gamma} / [(E_{L} - E)^{2} + \Gamma_{L}^{2}/4] = k_{o}^{2} \sigma_{n\gamma}(E) \sqrt{E/\pi} - \overline{\Gamma_{\gamma n}} \overline{\Gamma_{0}} X(E)/D^{2} - \sum_{r} \Gamma_{rn}^{\circ} \Gamma_{r\gamma} / [(E_{r} - E)^{2} + \Gamma_{r}^{2}/4]$$
(3.1)

and $\Gamma_{Ln}^{o}/(E_{L} - E) \approx 2k_{o} a_{eff} - \frac{\sum}{r} \Gamma_{rn}^{o}/(E_{r} - E) \pm 2k_{o} \sigma_{nn}(E)/4\pi$ (3.2)

if $|E_r - E| >> \Gamma_r$ for all resonances (which usually holds for $E \sim 0.025$ eV).

In the last member of the right hand side of Eq. (3.2) the sign should be the same as that of $d\sigma_{nn}/dE$ at E. The term $\Gamma_L^{2/4}$ in the denominator of the left hand side of Eq. (3.1) may often be omitted.

The next step depends on the data available.

(i) If there are data for σ_{nn} at many values of E, a plot of the reciprocal of the right hand side of Eq. (3.2) against E should give a straight line of slope (- $1/\Gamma_{Ln}$) and intercept (E_L/T_{Ln}) on the (E=0) axis. The value of a, the channel radius, may

need to be adjusted slightly to give a good fit. If this fails, then probably the parameters of two negative energy resonances need to be determined and this is best done by a computer programme. Once Γ_{In}° and E_L are determined, a single measured value of $\sigma_{n\gamma}$ suffices to fix $\Gamma_{L\gamma}$, if the capture widths for the positive resonances are known, or $\overline{\Gamma}_{\gamma}$ if they are not, and it is assumed that all $\Gamma_{r\gamma} = \overline{\Gamma}_{\gamma}$.

(ii) If the data for $\sigma_{nn}(E)$ are sparse, or if the line obtained in (i) is nearly horizontal, only the ratio $\Gamma_{Ln}\circ/(E_L-E)$ will be obtainable. A value of $\sigma_{n\gamma}$ will give $\Gamma_{Ln}\circ\Gamma_{L\gamma}/(E_L-E)^2$, and so $\Gamma_{L\gamma}/(E_L-E)$ may be found. One then must guess one of the three quantities $\Gamma_{Ln}, \Gamma_{L\gamma}$, and E_L . If information on $\overline{\Gamma}_{\gamma}$ is available, one can put $\Gamma_{L\gamma} = \overline{\Gamma}_{\gamma}$. Otherwise, one can keep the previously selected value of E_L ; notice that the most probable value of $\Gamma_{Ln}\circ$ (and hence of $(E_L - E)$) is zero, which is not very helpful.

If it is thought that Γ_{γ} should be within a certain range, the Porter-Thomas and Wigner distributions can be used to find the most probable values of Γ_{Ln}^{o} and E_L compatible with a Γ_{γ} in that range.

Finally, one should return to the first step and recalculate the contributions from unresolved resonances using the improved value for EL just obtained. However, this is rarely necessary, because the contributions are usually small and, as has already been pointed out, are not accurately known.

If there are two spin states of the compound nucleus to be considered, the process outlined above may be generalised, and made the basis of a least-squares fitting programme. Other data, such as the value and sign of the coherent scattering, can be included.

This completes the discussion of the methods available for the calculation of the effects of negative energy and unresolved resonances on low energy neutron cross-sections.

4. The Low Energy Cross-Sections of the Isotopes of Nickel

In this Section, these methods are applied to the analysis of the low energy cross-sections of the isotopes of nickel. Lack of space prevents a detailed discussion, which will be found in a forthcoming publication [8] on an evaluation of data for nickel over a much greater energy range. The numbers quoted here are preliminary estimates and may be altered in the final report, but they will illustrate the method.

The data on resolved resonances is derived chiefly from refs. [9], [10], and [11]. The values suggested for the channel radius for 5^{6} Ni and 60 Ni are those quoted in [9]; they are here taken to be the values for aerr at about 100 keV, which is the low energy end of the region analysed in the references quoted. The values for the other isotopes are just reasonable values which we have taken to be the mean of those for 5^{6} Ni and 60 Ni; better values of a are clearly needed.

No measured values of Γ_{γ} are available. We therefore rely on Cameron's semi-empirical formula [12], and note that comparison with other nuclides in the same mass region suggests a value of 0.75 eV.

$$D = 28.5 \pm 3 \text{ keV}; \Gamma_0 = 8.4 \pm 2.6 \text{ eV}$$

The two lowest resonances, at 13.8 keV and 33.2 keV, are both large, and these help to depress σ_{nn} at low energies: note that the measured value is only 1.45 b.

The highest resonance is at $E_H = 583$ keV; $E_L = 13.8 - 0.8 \times 28.5$ = - 9.0 keV. Using suffices H, L to renote contributions from positive and negative unresolved resonances, and using the notation of Section 2, at E = 0 we find that $\alpha_{\rm H}$ = 0.048, $\alpha_{\rm L}$ = 1.65, $\beta_{\rm H}$ = 4.05 and $\beta_{\rm L}$ = 0.561. At E = 100 keV, $\beta_{\rm H}$ = 3.86 and $\beta_{\rm L}$ = 2.44. Hence, if aeff (100 keV) = 7.0 fm, a_{eff} (0 eV) = (7.0 + 0.13 + 1.28) fm = 8.41 fm, where the second term is due to positive, and the third to negative, unresolved resonances.

at low energies yield: - $\Gamma_{T-}/R_T = -1.611 \times 10^{-3}$

and
$$\overline{r}_{\gamma} r_{\text{In}}^{o} / E_{\text{I}}^{2} = (0.1664 - 0.2017 \overline{r}_{\gamma}) \times 10^{-6}$$
,

so that $\overline{\Gamma}_{\gamma} < 0.83 \text{ eV}$.

If we assume E_L has its most probable value of -9 keV, these give:- $\Gamma_{Ln}^{o} = 14.5 \text{ eV} (= 1.7 \overline{\Gamma}_{n}^{o}) \text{ and } \overline{\Gamma}_{r} = 0.437 \text{ eV}.$ If we assume that $\overline{\Gamma}_{r} = 0.145 \text{ eV}$, from Cameron's [12] formula, we get instead: $\Gamma_{n}^{o} = 2.70 \text{ eV} (= 0.32 \overline{\Gamma}_{n}^{o}) \text{ and } E_{L} = -1.67 \text{ keV}$ (giving a spacing of 0.54D between this and the resonance at 13.8 keV).

The latter set of values of Γ_n^{o} and E_L is four times more probable than the former; so it is to be preferred lacking better information on Γ_{γ} . We should now iterate, since EL has been altered; but because of the considerable uncertainties involved, especially that in α_L and β_L (~100%), we stop at this stage, and note that the contributions from negative unresolved resonances is calculated as if EL = - 9.0 keV, giving an underestimate.

62_{Ni} (16.3 ± 2,4%)

Experimental data: $\sigma_{m\gamma} = (16.3 \pm 2.4)b$ at 0.0253 eV; $\sigma_{mn} = 9.21 \pm 0.4$ at about 1 eV; seff (100 keV) \approx 7 fm; $\Gamma_{\gamma} = 0.192$ eV from [12], or 0.75 eV by comparison with neighbouring muclides.

The parameters of the lowest positive energy resonance are taken from [13], instead of [10]; its energy is 4.6 keV. D = 18.9 keV and $T_n = 5.44$ eV.

The boundaries of the unresolved regions are at $E_L = -10.5$ keV and $E_{H} = 590.5 \text{ keV}.$ $a_{eff} (0 \text{ eV}) = (a_{eff} (100 \text{ keV}) + 0.12 + 1.29) \text{fm} = 8.41 \text{ fm}.$ Assuming Γ_{γ} to be unknown, the low energy cross-sections are fitted if:- $\Gamma_{\text{Ln}}^{0}/E_{\text{L}} = -4.919 \times 10^{-3}$ and $\Gamma_{\text{Ln}}^{0}/E_{\text{L}}^{2} = (3.85794/\overline{\Gamma}_{\gamma} - 0.92295) \times 10^{-6}$, so that $\overline{\Gamma}_{\gamma} < 4.2 \text{ eV}$.

If $E_L = -10.5 \text{ keV}$, $\Gamma_{Ln}^{\circ} = 51.8 \text{ eV}$ and $\overline{\Gamma}_{\gamma} = 2.77 \text{ eV}$, while if $\Gamma_{\gamma} = 0.192 \text{ eV}$ (from Cameron's formula) $E_L = -0.26 \text{ keV}$ and $\Gamma_{Ln}^{\circ} = 1.26 \text{ eV}$.

Again, the latter pair of values for E_L and $\Gamma_{\text{Ln}^{O}}$ is more probable, and so this set is adopted. The scattering cross-section has been assumed to be a decreasing function of energy at the energies considered (~ 0.02 eV).

$\frac{61}{Ni}$ (Abundance = 1.19 ± 0.07%; spin = $\frac{3}{2}$)

Experimental data: $\sigma_{n\gamma} = 2.5 \pm 1.1$ b at 0.025 eV; $\sigma_{nn} = 9.6 \pm 2$ b at about 1 eV; aeff \approx 7 fm in the centre of the resolved resonance region (~20 keV); $\Gamma_{\gamma} = 0.433$ eV from [12], but see below.

Parameters for 19 resonances are given in [13]; these were observed by Good et al. [14].

The resonances may of course be of either of the two spins 1 or 2; no experimental allocation of spins has been made, and the parameters given in [13] and [14] are just $E_{\rm T}$ and $2g\Gamma_{\rm N}^{0}$. The parameters give D = 2.3 keV for both spins combined, and $2g\Gamma_{\rm N}^{0} = 1.2$ eV. The first quoted resonance is at 6.97 keV, so it is very likely that there is at least one between this energy and 0 eV. In fact, Kapchigashev and Popov [15] have detected a resonance at 2.3 keV in the capture cross-section for natural nickel, which may be attributed to this isotope. From the quoted area of this resonance [15], one obtains $\Gamma_{\gamma} \approx 3.0 \pm 1.0$ eV, which is much higher than the estimate from [12].

There are therefore many uncertainties in the data for this isotope; fortunately its abundance is low, so that the effect on the cross-sections for natural nickel of any incorrect assumptions that we make will be small.

If we assume that the 2.3 keV resonance belongs to ⁶¹Ni, and has a value of $2g\Gamma_n^{\circ}$ equal to the mean, then, just considering the known resonances (including this one), $\Gamma_{\gamma} < 3.6$ eV.

Assume that there is a resonance of each spin state near an energy E_L , and that $2g\Gamma_n^0$ for each resonance is equal to the mean (1.2 eV). Then taking $\Gamma_{\gamma} = 3.0$ eV, and assuming $\alpha_L \approx -\frac{D}{E_L}$, we find that $E_L = -13.2$ keV; this is very distant, since the positive resonances account for most of the capture. (This would not be so if Γ_{γ} is in fact much less than 3 eV).

Since no spins are known for the positive resonances, we shall just assume here that the cross-section near thermal is due entirely to potential scattering, giving eff ≈ 8.7 fm.

Obviously much more information is needed about this isotope, and in particular about the spins of the resolved resonances, and about the existence of low positive energy resonances.

⁶⁰Ni (26.21 ± 0.51%)

Experimental data: $\sigma_{n\gamma} \approx 2.84 \pm 0.23$ b at 0.0253 eV; $\sigma_{nn} = 1.07$ b at low energies; aeff = 6.5 ± 0.5 fm at about 100 keV [9]; $T_{\gamma} = 0.311$ eV[12].

The parameters for the positive resolved resonances are taken from [13] and [10]. D = 17.6 keV, and $\overline{\Gamma_n}^\circ = 4.91 \text{ eV}$. Hence, $E_L = -1.58 \text{ keV}$ for an initial guess; $E_H = 594.8 \text{ keV}$. The low energy radius is aeff (OeV) = 6.5 + 0.12 + 1.76 = 8.38 fm.

Fitting the thermal cross-sections gives:- Γ_{Ln}^{o} / E_L = - 2.29 x 10⁻³

and $\Gamma_{In}^{0} / E_{L}^{2} = (0.672/T_{\gamma} - 0.206) \times 10^{-6}$, so that $\overline{T}_{\gamma} < 3.3 \text{ eV}$.

with $\overline{\Gamma}_{\gamma} = 0.311 \text{ eV} [12]$, $E_L = -1.2 \text{ keV}$ and $\Gamma_{Ln}^{0} = 2.7 \text{ eV}$.

⁵⁸Ni (67.86 ± 0.22%)

Experimental data: $\sigma_{n\gamma} = 4.69 \pm 0.386$ at 0.0253 eV; $\sigma_{nn} = 24.584 \pm 0.093$ b at about 1 eV. $a_{eff} = 7.5 \pm 0.5$ fm at about 100 keV [9]. From [12], we expect $\Gamma_{\gamma} \approx 0.568$ eV.

This is the most abundant isotope, and also perhaps the hardest to fit. The main reason for the difficulty is the combination of a very large scattering cross-section, indicating a large value for $\Gamma_{Ln}/|E_L|$, and a moderate value for the capture cross-section, indicating a moderate value for Γ_{Ln}^{o}/E_L^2 . Hence EL, and consequently Γ_{Ln}^{o} , must be very large, if only one "resolved" negative energy resonance is considered.

The parameters are taken from [10] and [13]. The lowest positive resonance is at 15.5 keV; D = 20.9 keV and $\Gamma_n^0 = 6.4$ eV. We assume to start with that $E_L = -5.4$ keV. This gives aeff (0eV) = 8.9 fm and the experimental data are fitted if:-

$$\Gamma_{\text{Ln}^{0}} / E_{\text{L}} = -3.88 \times 10^{-3}$$

 $\Gamma_{\text{Ln}^{0}} / E_{\text{L}}^{2} = 1.11 / \Gamma_{\text{Y}} - 0.0846$, so that $\overline{\Gamma}_{\text{Y}} < 13.1 \text{ eV}$.

We also attempt to fit the scattering cross-section at 40 keV, where $\sigma_{nn} \approx 8$ b (reading from a graph in [9]). At 40 keV, $e_{eff} = 7.7$ fm, and to fit σ_{nn} we need

 Γ_{Ln}^{o} (- E_{L} + 4 x 10⁴) = 1.09 x 10⁻³ Hence, E_{L} = - 14.3 keV, Γ_{Ln}^{o} = 55.6 eV, and $\overline{\Gamma}_{Y}$ = 3.1 eV.

This result is only a first approximation, and a more detailed fit will be attempted; this may need two negative energy resonances.

It will be realised that many of the results we have obtained are only intelligent guesses: better results must await more experimental information. It is clear, in particular, that effective channel radii need to be better known, and that some more information on capture widths is needed (perhaps from resonance integral measurements). However, it is hoped that this detailed discussion of the nickel isotopes will have illustrated, and shown the usefulness of, the methods.

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MECHANIZED EVALUATION OF NEUTRON CROSS-SECTIONS

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Abstract

MECHANIZED EVALUATION OF NEUTRON CROSS-SECTIONS. The evaluation work to provide accurate and consistent neutron cross-section data for multigroup neutronics calculations is not fully exploiting the available theoretical and experimental results; this has been so particularly since the introduction of on-line data handling techniques enabled experimenters to turn out vast quantities of numbers. This situation can be radically improved only by mechanizing the evaluation processes. Systems such as the SCISRS tape will not only largely overcome the task of collecting data but will provide speedy access to it; by using computers and graph-plotting machines to tabulate and display this data, the labour of evaluation can be very greatly reduced.

With some types of cross-section there is hope that by using modern curve-fitting techniques the actual evaluation and statistical accounting of the data can be performed automatically. Some areas where automatic evaluation would seem likely to succeed are specified and a discussion of the mathematical difficulties incurred, such as the elimination of anomalous data, is given.

Particularly promising is the use of splines in the mechanized evaluation of data. Splines are the mathematical analogues of the draughtsman's spline used in drawing smooth curves. Their principal properties are the excellent approximations they give to the derivatives of a function; in contrast to conventional polynomial fitting, this feature ensures good interpolation and, when required, stable extrapolation. Various methods of using splines in data graduation and the problem of marrying these methods to standard statistical procedures are examined.

The results of work done at AWRE with cubic splines on the mechanized evaluation of neutron scattering total cross-section and angular distribution data are presented.

DISCUSSION

K. PARKER: This is the only paper dealing specifically with the need to develop and use computer techniques for handling the ever-growing volume of experimental data with which the evaluator must struggle. Believers in mechanized evaluation do not claim that computers can make the physics judgements essential in evaluation, but that much of the tedious routine work can be done on computers (for example, the least-squares fit of the 2200-m/s cross-sections of fissile nuclides). As SCISRS tapes become more widely available one can envisage programmes that will produce best-fit curves to many sets of experimental measurements having different statistical errors and different weights. The physics consists in assigning weights to experiments that give values differing by more than the combined stated statistical errors, but the remainder of the process can be automated, leading to an evaluated curve at each point of which one can calculate a probable error in the evaluated cross-section.

R. TASCHEK: I should like to warn against the assumption that all one requires for solving an evaluation problem are the right kind of computer code and the correct calculational approach. One often needs to determine where a complex experiment has gone wrong or whether experimental values really are correct within the error given. This is a matter

for personal judgement and a highly professional part of evaluating which cannot be done by computer.

K. PARKER: I was careful to say that computers cannot make physics decisions. However, they can be used to compute best curves through conflicting experimental measurements once certain decisions on the physics have been made. This is what is done with the least-squares analysis of 2200-m/s cross-sections of fissile nuclides. Drawing a best smooth curve as a function of energy is a more difficult, but still solvable, problem for a computer, and it is with this kind of problem that mechanized evaluation in the sense of paper CN-23/24 is concerned.

J.J. SCHMIDT: In my paper (CN-23/124) I stressed that discrepancies between sets of different experimental data for one and the same quantity constitute the basic bottle-neck in evaluation. The difficulties of the evaluator are often increased by the fact that experimentalists themselves do not make a sufficiently thorough comparison of their results with those of other groups.

THE ROLE OF ISOTOPIC COMPOSITION MEASUREMENTS IN CROSS-SECTION EVALUATION

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Abstract

THE ROLE OF ISOTOPIC COMPOSITION MEASUREMENTS IN CROSS-SECTION EVALUATION. Isotopic composition measurements, made on fuel irradiated in operating power reactors, provide a valuable input to select the most useful set of evaluated cross-sections for reactor design application. Evaluators often establish the validity of a cross-section evaluation by analysis of resonance integral measurements and critical experiments. Experimental data obtained from operating reactors can provide a more sensitive test of the adequacy of cross-section sets.

Critical experiments test evaluated data for eta, while isotopic ratio measurements test relative reaction rates of all isotopes present. A vast amount of data for a wide range of conditions provides a statistically significant sample for cross-section ratio evaluations. This statistical sample provides a more sensitive test of relative reaction rates than is normally obtained from precise laboratory measurements of a limited range for conditions occurring in critical experiments.

If cross-section evaluating is to be kept in proper perspective, the evaluation process cannot be considered independently of the cross-section applications. The use of only critical experiments or resonance integrals, to test an evaluated set of data and models, is not sufficient to arrive at the best set for general application. Ratios of reaction rates, inferred from isotopic composition measurements, should be included. For light-water power-reactor design application, as for other reactor types, the interaction between crosssection data uncertainties and approximations used in the models is sufficient to affect significantly final results and conclusions. For these reasons it is important to make use of all sources of data, including data from operating power reactors, to select evaluated cross-section sets for general use in reactor design calculations.

EVALUATION OF THE ELASTIC AND INELASTIC SCATTERING CROSS-SECTIONS OF 14-MeV NEUTRONS FOR EVEN-EVEN NUCLEI

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Abstract

EVALUATION OF THE ELASTIC AND INELASTIC SCATTERING CROSS-SECTIONS OF 14-MeV NEUTRONS FOR EVEN-EVEN NUCLEI. A method for the evaluation of elastic and inelastic scattering cross-sections for the first collective state of even-even nuclei is described. Some theoretical results are compared with the experimental ones; comparisons are also made with DWBA.

TABLE DES INTEGRALES DE RESONANCE

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Abstract — Résumé

TABLE OF RESONANCE INTEGRALS. The authors' purpose is to regroup the measured resonance integrals for a number of materials, after correcting them with a view to reducing them to a common definition, and to compare them with the value calculated from resonance parameters, when these parameters are known with a sufficient degree of accuracy. These comparisons are made for use with the infinite dilution method.

The resonance integral above the 1/V part, which corresponds to infinite dilution, is defined by the relationship:

$$I = \int_{E_{C}}^{\infty} \left[\sigma(E) - \sigma_{0} \sqrt{\frac{E}{E_{0}}} \right] \frac{dE}{E}$$

From this we derive the total integral I_r by selecting the lower limit of integration E_c equal to 0.55 eV. Only those materials are studied whose lowest resonance wing is extinguished at this energy.

The resonance integrals calculated from the parameters are determined for infinite dilution by means of the ZUT computation programme and the complementary TUZ programme for the contribution of unresolved resonances.

The authors have made certain corrections in the measured resonance integrals to standardize and thereby facilitate comparisons. These consisted, in particular, of the following: calculation of the 1/V part to obtain I from I_r ; modification of quenching to reduce them to energy $E_c = 0.55$ eV; standardization relative to the resonance integral for gold, taken as equal to I = 1540 b and to the cross-section at 2200 m/s of natural boron, taken as equal to 761 b or 771 b, depending on the isotopic concentrations; extrapolation to infinite dilution; adoption, if necessary, of the final thermal cross-sections when they enter into the calculation of I.

For each material the authors mention all the measurements with which they are acquainted, the calculated value and the recommended values for I and I_{τ} that can be deduced from this compilation.

TABLE DES INTEGRALES DE RESONANCE. Les auteurs se proposent de regrouper, pour différents corps, les valeurs des intégrales de résonance mesurées, après les avoir corrigées pour les ramener à une définition commune, et de les comparer à la valeur calculée à partir des paramètres de résonance lorsque ceux-ci sont connus avec une précision suffisante. Ces comparaisons se font pour la dilution infinie.

L'intégrale de résonance, au-dessus de la partie en 1/V, correspondant à la dilution infinie, est définie par la relation

$$I = \int_{E_0}^{\infty} \left[\sigma(E) - \sigma_0 \sqrt{\frac{E}{E_0}} \right] \frac{dE}{E}$$

On en déduit l'intégrale totale I_r en choisissant la limite inférieure d'intégration E_c égale à 0,55 eV. On n'étudie que les corps dont l'aile de la résonance la plus basse est éteinte à cette énergie.

Les valeurs des intégrales de résonance calculées à partir des paramètres sont établies pour la dilution infinie à l'aide du programme de calcul ZUT et de son complément TUZ pour la contribution des résonances non résolues.

Sur les valeurs des intégrales de résonance mesurées, les auteurs ont effectué certaines corrections afin de les normaliser pour faciliter les comparaisons, en particulier: calcul de la partie en 1/V pour obtenir l'à partir de I_r; modification de la coupure pour les rapporter à l'énergie $E_c = 0, 55 \text{ eV}$; normalisation par rapport à la valeur de l'intégrale de résonance de l'or prise égale à I = 1540 b et à la section efficace à 2200 m/s du bore naturel prise égale à 761 b ou 771 b suivant les concentrations isotopiques; extrapolation à la dilution infinie; éventuellement, adoption des dernières valeurs des sections efficaces thermiques lorsqu'elles interviennent dans le calcul de I.

Pour chaque matériau, les auteurs citent l'ensemble des mesures dont ils ont eu connaissance, la valeur calculée et les valeurs recommandées pour l et l_r qu'ils ont pu déduire de cette compilation.

ACTIVITIES OF THE CROSS-SECTION COMPILATION AND EVALUATION CENTERS AT THE BROOKHAVEN NATIONAL LABORATORY*

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Abstract

ACTIVITIES OF THE CROSS-SECTION COMPILATION AND EVALUATION CENTERS AT THE BROOKHAVEN NATIONAL LABORATORY. The growth of the compilation and evaluation efforts at the Brookhaven National Laboratory are reviewed. The current work of the Sigma Center is discussed, including the status of the publication of supplements to BNL-325 and the current state of the SCISRS-1 tape. Future needs for BNL-325 type publications and SCISRS-11 cross-section tapes are outlined.

The history of the Cross-Section Evaluation Center at the Brookhaven National Laboratory is similarly reviewed. The status of current work is discussed, including the growth of the ENDF/A tape. The status of US efforts to produce a cross-section tape (ENDF/B) at an early date to satisfy the needs of US reactor designers is discussed. The continued importance of integral experiments and their accurate analysis to provide checks of the cross-section tapes is pointed out.

The role of the Brookhaven National Laboratory in collaboration on an international basis is reviewed, including its current relationship to the ENEA Neutron Data Compilation Centre, the International Atomic Energy Agency and other nuclear centres.

1. INTRODUCTION

The Brookhaven National Laboratory has traditionally played a key role in the measurement, compilation and evaluation of neutron cross-sections. At a time when new information centers are being proposed and when "centers of excellence" are supposed to spring up Minerva-like into instant maturity it may be well to reflect on the major reasons for these past accomplishments; the presence of strong staffs of both nuclear and reactor physicists, of the required tools such as advanced research reactors, accelerators and computers, and last but not least the steady support of the USAEC and its advisory bodies.

We last reported on Brookhaven's activities in cross-section compilation and evaluation at the AEC-ENEA sponsored seminar of last May [1]. We assume that the papers which were presented at this Conference are generally available and shall therefore concentrate on developments since that time.

2. THE SIGMA CENTER

During the past year, the compilation and evaluation efforts at the Brookhaven National Laboratory have been accelerated. We shall discuss first the status of the publications of supplements to BNL-325 since these are of worldwide interest. In May, 1965 we were able to report that a comprehensive three-volume second supplement was in preparation. Volume I, Z=1-20 had already appeared and Volume III, Z=88-96 was distributed during

* Work performed under the auspices of the United States Atomic Energy Commission.

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the summer of 1965. The rapid increase in cross-section data for medium weight elements has made it necessary to publish Volume II in three parts, each consisting of about 400 pages, in our new book-size format. The formal work on this volume was completed in August, 1966. At that time, Volume II-A, Z=21-40 had been published and distribution had been started. Volume II=B, Z=41-60 was in press and distribution is anticipated in October. The final volume of the series, Volume II-C, Z=61-87 is expected to be in print about the end of this year.

With the completion of these supplements there will be a lull in formal BNL publication activities. As M. Goldberg has pointed out [2], the next edition of BNL-325 will certainly run to several volumes. We are therefore anxious to learn whether a new ten-volume set of BNL-325's will be as well-thumbed as the older editions. Some speed-up in our methods of processing and publishing data will also be required and we are concerned lest the quality of the work suffer on this account. During the next year we shall be gaining experience with fast computer-based, graphical plotters. A modern Cal-Comp plotter and associated equipment has recently been acquired by the BNL Applied Mathematics Department. As these facilities become operative, an attempt will be made to supplement BNL-400, the angular distribution compilation, entirely by computer.

The storage and transmittal of cross-section data at the ENL Cross-Section Compilation Center has for some time been accomplished by use of a magnetic tape data library known as SCISRS [3], the Sigma Center Information Storage and Retrieval System. The rapidity with which the SCISRS tape can grow is indicated by the fact that it contained only 230 000 individual data points when Goldberg reported on its use in March, 1966 at the Conference on Neutron Cross-Section Technology [2]. In August 1966, it contained over 600 000 data points. Much of this growth is attributable to the digestion of data contributed by the Nevis group at Columbia and to the transfer of all data contained in BNL-400, the "Angular Distribution" compilation, onto tape. A similar large increase in data storage is expected as the recent Los Alamos bomb test cross-section data arrives and is digested.

To date no problems have arisen in the rapid storage and transmittal of data via SCISRS and relatively modest blocks of machine time are required for this purpose. As an example, the bulk of the recent Columbia data was rapidly transferred to tape by arranging for block time on the IEM-7094 computer. A block time of two hours per night (during which up to 20 000 data points can be processed) is found to be more than ample to keep up with the current rate of data acquisition and no requirement for an order of magnitude increase in machine time is anticipated in the future.

While SCISRS is therefore satisfactory for the internal needs of the Center, it is realized that it cannot, in its present form, satisfy the needs of all users. Recommendations for the specifications of a second generation SCISRS has therefore been undertaken by a subcommittee of the NCSAG (Neutron Cross Section Advisory Group) for the USAEC Division of Research. As of this date, the required SCISRS-II specifications have not yet been defined but every effort will be made to coordinate the Sigma Center compilation work with that of its many diverse groups of users.

3. THE CROSS-SECTION EVALUATION CENTER

An important group of users of the Sigma Center compilations is the USAEC Division of Reactor Development and Technology (DRDT). This Division maintains the Cross-Section Evaluation Center at Brookhaven. In contrast to the work of the Sigma Center, the work of the Cross-Section Evaluation Center is primarily that of systematic evaluation of neutron cross-section data. However, the operations of the two Centers are linked and their personnel have always worked in close proximity which results in considerable mutual benefits. The work of this group has been previously described [1] and the recent individual evaluation efforts of its members will be discussed in a companion paper by D. Goldman and K. Parker.

We shall therefore confine our discussion of the Cross-Section Evaluation Center's work to group efforts. First, it is worth mentioning that the optical model code ABACUS-2 of E. H. Auerbach has received worldwide interest and distribution. Recognition of the rapidity and flexibility of this code has led to a continuing series of requests for its listings, master deck or other components from universities and major nuclear research laboratories both in the United States and abroad. The international appeal of such codes as ABACUS-2, THERMOS, etc. points out the importance of this work. It is clear that the field badly needs an improved optical model code for neutron cross-sections of similar flexibility and convenience. In the same vein, we badly need satisfactory calculational models for multilevel fits of neutron cross-sections both for compilation and for evaluation work. For some time the Cross-Section Evaluation Center directly supported the application of the Adler and Adler multilevel formalism to fissionable isotopes. Some work of this type is now supported directly by the DRDT Division of the USAEC.

In addition to the work of its Centers, members of the Reactor Physics Division at Brookhaven have frequently published reports which contain correlations of nuclear data with integral experiments [1]. Discrepancies that have been noted between differential cross-section measurements and integral measurements have frequently led to the correction and improvement of the techniques for both types of experiments. As an example, the discrepancy in the epithermal value of alpha of 236 U which was the subject of a special panel discussion at Gatlinburg in 1965 [4] has now been considerably clarified. Similarly, the analysis of critical experiment data for thermal reactor assemblies in the U. K. had led G. Tyror and J. R. Askew to recommend a substantial reduction in resonance integrals for 238 U, a recommendation that requires serious further investigation of these supposedly firm numbers.

Cross-section accuracies of about $\frac{1}{2}\%$ are frequently requested and can be justified for several nuclides. However many cross-section measurers feel that accuracies of 1-3% are probably ultimate limits. On the other hand, criticality data (and associated material tolerances and quality control) to $\frac{1}{2}\%$ appear feasible. With techniques for integral experiments improving to the point where measurements of detailed neutron spectra are becoming feasible, it appears that integral and critical experiments for thermal as well as fast spectra should continue to be important for many years.

4. THE ENDF FORMAT

Various systems for storing and retrieving evaluated nuclear data have sprung into being at individual laboratories in response to their CHERNICK

specific requirements. The need for a flexible standard format was recognized some years ago by H. Honeck at Brookhaven. Under the sponsorship of the Reactor Mathematics and Computation Division of the American Nuclear Society, he developed the punched card/magnetic tape system called the ENDF or Evaluated Nuclear Data File. Details of the system are contained in various BNL reports [5,6] and Cross-Section Evaluation Center Newsletters. The latter are issued whenever major changes or additions to the ENDF Master Library are made.

As shall shortly be made clear the ENDF system has recently been split into two systems ENDF/A (Evaluated Nuclear Data File, Version A) and ENDF/B. Version A contains evaluated point data and is intended for use by individuals actively engaged in evaluation work. Thus it may contain multiple evaluations of the same data. It may be fragmentary in the sense that the entire energy range may not be covered or a complete set of partial cross sections may not be given. On the other hand, it uses the original, highly flexible format.

Version B is intended as direct input into multigroup and Monte Carlo codes. It will contain one complete set of data for each material and a relatively simple format. The data on Version B will serve as a reference cross-section set for user groups. Where Version A is a continuously growing library, it is expected that ENDF/B will be changed only occasionally, possibly on an annual basis.

The library ENDF/A presently contains an updated version of the Aldermaston/Winfrith Data File. An entirely tape-to-tape Fortran II program was written to convert the data to the ENDF/A format. About 15 minutes of IEM-7094 time were required to convert the first such tape received, dated July 1, 1964, which contained 23,964 BCD card images in the A/W format. In addition, ENDF/A contains a recent KAPL evaluation of fast neutron cross-sections of oxygen and recent GA evaluations of thermal neutron scattering kernels for graphite, beryllium and water. As similar careful evaluation efforts are completed and sent to S. Pearlstein at the Cross-Section Evaluation Center they will be placed in the ENDF/A Library at the Brookhaven National Laboratory.

5. THE CROSS-SECTION EVALUATION WORKING GROUP (CSEWG)

The limitation of the original ENDF system was that it was openended. There remained the need to close the loop for reactor designers by putting the data into a format of immediate use to the largest or the smallest user. The splitting of the ENDF system into two parts was a step in this direction. Additional links are however still required. There are shown schematically in Fig. 1.

The circles in Fig. 1 may be thought of as data libraries, while the boxes are computer codes designed to process the data. The links designated as SCØRE and ENCØRE are complex and do not yet exist. Since SCØRE forms the link between SCISRS and ENDF/A, it is part of the problem currently under consideration by the NCSAG. Some steps in the development of SCØRE and ENCØRE have also been taken by the DRDT Division of the USAEC, but it is clear that it will take considerable time before these steps are fully implemented.

For these reasons, the DRDT Division of the USAEC on the advice of its Advisory Committee on Reactor Physics (ACRP), has decided to concentrate on the early completion of an ENDF/B library and its associated programs. The ENDF/B tapes can serve as direct input to existing Monte



FIG.1. Schematic of the flow of nuclear data from compilation to reactor calculation

Carlo codes or the MC² code which is being designed by the Argonne National Laboratory for the design of fast breeders. For other applications, further processing of ENDF/B data is required. The code ETØM (ENDF to MUFT) is currently being developed to process the data into multigroup sets for thermal codes such as MUFT. Finally, FLANGE has been developed to obtain thermal neutron scattering kernels from the scattering laws contained on ENDF/B. All of these codes are expected to be operational during the Fall of 1966.

The evaluation of the data required for the first ENDF/B tape is being carried out by a working committee (CSEWG) composed of representatives of USAEC laboratories and contractors having direct responsibilities to the DRDT Division of the USAEC, plus representatives from the AEC, the Advisory Committee on Reactor Physics and laboratories in the Naval Reactors Branch. The effort is focused in the BNL Cross-Section Evaluation Center with S. Pearlstein as its Chairman. The goal is a complete set of cross-section data for some fifty reactor materials and isotopes from hydrogen to curium-244 by November, 1966. Once the data has been placed on ENDF/B, distributed to reactor designers and processed into multigroup forms, it will be tested against integral measurements obtained from critical and exponential experiments. The total experience gained from using the data plus new cross-section data can then be combined in an attempt to generate an improved ENDF/B tape. With time it is expected that formats and codes will also be improved and the list of isotopes expanded.

At the CSEWG meeting of June 9-10 at Brookhaven, members of the commitee agreed to the following standards in their evaluation of data for individual isotopes: Data submitted for inclusion in the ENDF/B library should be documented even if in minimal form. The source of the data, whether by report or personal communication should be indicated. In addition comments about the methods, experimental or theoretical, used in deriving the data are requested. Participants are urged to provide as complete a documentation as possible. The CSEWG committee itself is composed of a strong group of nuclear and reactor physicists most of whom have had

CHERNICK

experience in reactor cross-section evaluation. Many previous data evaluations are available to them. Thus, in spite of time limitations, it is hoped that the first ENDF/B library, after thorough checking, will become a widely used tool.

6. INTERNATIONAL COOPERATION

By international agreement, the Sigma Center at ENL services the United States and Canada in the compilation of measured neutron data. Similar Centers now exist in Saclay, Vienna and Obninsk. By agreement with the ENEA, the SCISRS data are routinely sent to the Neutron Data Compilation Center (CCDN) at Saclay. The second CCDN newletter (CCDN-NW2) contains a complete listing of their SCISRS master tape as of June, 1966. During the first eight months of 1966 some 450,000 data points were sent from the Sigma Center to the Saclay Center. In addition, the Sigma Center cooperated in the training of personnel for both the Saclay center and the IAEA Nuclear Data Unit at Vienna although formal cooperative agreement for regular exchange of large blocks of data.with the latter Center has not yet been established. However, some fission cross-section data, published in BNL-325, were sent to the IAEA Center for their use at this Conference.

The return flow of cross-section data from Saclay to Brookhaven is still sporadic. In part, this is due to computer problems and to the strong effort being made there on the CINDA System. We have found the CINDA system useful, particularly in cross-section evaluation. The system still has some weaknesses, chiefly the lack of completeness of its references and lack of documentation behind some of its cited references. The latter problem could be avoided if minimum standards were adopted for cross-section evaluation, while the former problem can be overcome with time and a strong staff of CINDA readers.

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DISCUSSION

J.S. STORY: I should like to ask whether the ENDF systems are available to users outside the United States. Many of the European files are already available from the ENEA Neutron Data Compilation Centre (CCDN) at Saclay, from where I believe Honeck obtained them for inclusion in the ENDF/A system.

J. CHERNICK: Many of the evaluations already on the ENDF/A tape are immediately available. The ENDF/B system still requires a certain amount of testing. We hope that the scope of our agreements with the ENEA Neutron Data Compilation Centre at Saclay and with similar centres will be enlarged to include the ENDF/B system.

J.S. STORY: I should like to raise the same question in relation to the associated computer programmes. I think this poses a somewhat more complex organizational problem, because computer programmes always need up-dates, and of course an adequate operating manual. Most of the computer programmes associated with the UKAEA Nuclear Data Library are available from the ENEA Computer Programme Library at Ispra. A review of nuclear theory programmes useful for data evaluation was prepared by Mike James earlier this year. This review was published in Newsletter No.5 from the ENEA Computer Porgramme Library. Later another member of the Winfrith evaluation staff spent two weeks at Ispra ensuring that the programmes associated with the UKAEA Nuclear Data Library would operate on the computer at the ENEA centre.

J. CHERNICK: Certainly the special programmes will be available at Brookhaven. In addition, Argonne National Laboratory will have an arrangement with the ENEA Computer Programme Library at Ispra.

Session XI

INTERNATIONAL CO-OPERATION IN THE FIELD OF NUCLEAR DATA

INTERNATIONAL CO-OPERATION IN THE FIELD OF NUCLEAR DATA

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I see from the programme of this Conference that I am supposed to be giving a talk, the title of which would be announced. Perhaps the organizers of this Conference showed some wisdom in arranging that the title was not in fact known since a session heading of "International Co-operation in the Field of Nuclear Data" would normally be more than sufficient to ensure a limited audience. However, now that I have a captive audience, and, I see, a very large one, inveigled by the lack of title, if not by the speaker, I am fairly safe in giving the title of my talk as "International Co-operation in the Field of Nuclear Data in as far as it is Influenced by Recent Trends in the Measurement and Storage of Such Data and the Effects on this of Grandiose Schemes for a World Locked in Permanent Embrace by Computers".

Now, before you all leave, perhaps I can tempt you by changing the title to that of "The Effects of Progeny on Legalizing Illicit Unions" and also remark that, boring as it may be, it is perhaps correct that the majority of you who were thinking of leaving are of course professional scientists who should perhaps ponder at least a few of the serious remarks which I hope to make this afternoon.

There are several levels of international co-operation in the field of nuclear data, if not in many other fields, not all of which I should wish to expatiate. They may be **con**veniently classified as follows:

1. Co-operation at government level which manifests itself mainly in conducted visits of scientists under official auspices, the exchanges of personnel, bi-lateral (and occasionally multi-lateral) agreements, exchanges of samples, experimental equipment, etc. These cooperative ventures are arranged principally by politicians, lawyers, scientific lawyers and politically motivated scientists, and, except as they influence each and every one of us in some way, even if only indirectly, are not perhaps of great interest at this meeting.

2. International committees of which the best known example is the European-American Nuclear Data Committee (EANDC), who exist in a somewhat autonomous manner and succeed in settling what can be done by co-operation without coercion; these I would like to return to;

3. International centres, such as EURATOM, Dubna, the Trieste Centre and the European Neutron Data Compilation Centre, which I had better return to, if only because I have a fair working knowledge of how international co-operation can be affected, or can go sour, in this particular milieu; 4. International secretariats, such as the United Nations, the International Atomic Energy Agency (IAEA) in Vienna, and the European Nuclear Energy Agency (ENEA) in Paris, who exist to further scientific co-operation in an international manner, whether or not they carry out particular projects, as some of them of course do. Here the emphasis is on creating a formal framework within which scientists from many countries may discuss and set up international collaborative exchanges and projects;

5. International scientific meetings which, like the present one, are a specific co-operative activity of the organizations of the previous group which I mentioned above, and whose main benefits are provided by their facilitating my final level of international co-operation.

6. The personal contacts between individual scientists.

Let me just make three points about the last two levels of international co-operation. First, they are undoubtedly the most fruitful form of co-operation in their effect on the advances of science. Secondly, one of the unfortunate facts of life is that most of you who come here earn your trip by writing a paper. This is a hardship arising largely from the dealings between your directors and their suppliers of money. The inevitable effect, as we all know, is the presentation in many instances of slightly revised versions of papers already given elsewhere. The effect of this on the multiplicity of reports and the resulting confusion has to be seen to be believed, but its worst features are felt most severely by the groups who compile references and data. I therefore make two pleas which have been made ever more frequently recently: if you must give a follow-up paper then please, if only in the written version, give us the relationship between your new paper and its multitudinous predecessors and tell us what supersedes what. My second plea which will fall on more barren ground is that those of you who are directors of research groups try sending some of your personnel to conferences without their presenting a paper. Let them come just to talk to their colleagues.

And the last point, which necessitates a jump ahead in this paper, is that I should like to give an assurance on the part of the Compilation Centres, which I hope can also be given by national atomic energy commissions, that the old-established personal contacts of scientists with each other will not, and should not, be destroyed by the existence of committees, centres, and agencies whose chief prerogative is that of collecting data and collecting knowledge about data.

I don't consider that it is part of my duty this afternoon to describe the activities of my level 4 of international co-operation, i.e. the international agencies, who are well able to look after their own selfadvertisment, mainly by the issue of excellent annual reports of their activities.

I come now to the international committees and international centres and I will concentrate on the field of neutron data. Tedious as these matters may be to many of you at this meeting, it should be evident to all of us by now that the hard facts of life have to be faced. Nowadays, almost every government has a limited amount of money which it will allow to be spent on research, and we are fast approaching the point
where there is a limited number of highly qualified professional scientists available to supply all the scientific disciplines. The neutron scientist no longer has the romantic appeal of twenty years ago, which limits the intake into the field, and perhaps he has also been so successful in his work that governments must now of necessity question the usefulness, if not the interest, of further spending of vast quantities of money in this field.

So it behoves us, if we wish to continue in what can still be an exciting field, to attempt the maximum amount of co-operation to avoid duplication and to improve the process of decision making for the future by pooling our brains rather than charging off in all directions for reasons of national prestige, or simple inability to talk to each other.

Luckily, as it turns out, or perhaps it is not so surprising, there are broad areas of agreement between the requestors of data, which are quite independent of national boundaries. At the same time it is obvious that work carried out in one research laboratory can be of great interest thousands of miles away and that this information not only should get from one of these laboratories to another but should be widely available to anyone who can make the slightest use of it.

Co-operation in these matters has been attempted over the last six years through the medium of the European-American Nuclear Data Committee. This committee, which carefully inspects requests for data, bearing in mind the present status of available nuclear data, allocates a system of priorities which depend to some extent on the requestors providing adequate reasons for their demand. Discussion within this committee enables measurements on nuclear data to be carried out in a rational manner and avoids, where possible, unnecessary duplication of effort.

The EANDC, which is an organization of the OECD countries, was based on the original Tripartite Nuclear Cross Sections Committee, the TNCC and has been a fruitful example of the effective international committee [1]. Perhaps its autonomous nature and the fact that scientists themselves run it have in no small way contributed to its success.

We can now look forward hopefully to a world-wide organization of this type which could be promoted under the aegis of the International Atomic Energy Agency and which might succeed the International Nuclear Data Scientific Working Group (INDSWG).

I return now to a point hinted at earlier; results of experimental research are unnecessarily difficult to find and do not reach the various users of data — other research workers, evaluators, reactor designers. There is only one solution to this problem — the formation of information centres, whose main objectives should be that of efficient collection and dissemination of the material of relevance in any particular field. Where possible, it appears more sensible to organize these on an international scale. The results which they collect are produced all over the world and are needed all over the world. In the case of neutron data the very quantity of the material and therefore the expense of its collection and dissemination make it less than reasonable for each and every country to set up its own information unit. On the other hand, the efficient operation of such a centre will probably limit its geographical coverage to that area which it can most effectively service. This brings me then to level three of international co-operation. I am referring to neutron data compilation centres. The four principal ones which I shall discuss are the Centre de compilation de données neutroniques serving those European countries and Japan which are members of the Organisation for Economic Co-operation and Development, (OECD), the Obninsk Centre serving the USSR, the Sigma Center in Brookhaven serving North America, and the Vienna Nuclear Data Unit at the moment serving other areas.

The common services which are provided by neutron data centres, although all do not necessarily take place in any one unit, may be classified quite easily into collection of references, collection of data, assessment of data which I have liked to call in the past "physics evaluation", collection of evaluations, and finally evaluation activities as the word is now understood today, in their entirety.

The American activities, which I take up first for historical reasons, have been concentrated until recently in the collection of references which began in the United Nuclear Corporation in 1956. This collection of references, which is called CINDA (Computer Index of Neutron Data), was conceived by Professor Herbert Goldstein and his collaborators. From obscure beginnings, this index has achieved world-wide recognition despite some undoubted faults. It is now concentrated at the Division of Technical Information Extension, of the United States Atomic Energy Commission situated at Oak Ridge.

The collection of data has taken place at the Sigma Center in Brookhaven from the early days of Don Hughes. The "physics evaluation" which I referred to above is also carried out at the Sigma Center and takes the form of "barn books", the well-known BNL 325 and its many volumes and supplements. Collection of evaluations and evaluation activities themselves are carried out at the evaluation centre at Brookhaven National Laboratory.

The activities in the USA have been very fully described by Goldberg at the 1966 Washington Conference [2], and by Chernick in a paper to this Conference [3]; therefore, I need not go into greater detail.

The United States' efforts to compile the more widely used neutronic data, in the face of the rapidly increasing quantities of experimental data being produced, pointed out inadequacies in existing compilation facilities. The Americans had undertaken it alone and the rest of us had made no great contribution in the compilation field. But now the situation had become critical and this is where we see the effects of the different levels of international co-operation which I mentioned above.

Informal contacts, some taking place at international meetings, resulted in the EANDC recognizing that a crisis was fast approaching. It was a very common complaint that the end results of research were becoming more and more difficult to obtain and were not reaching the users. The EANDC, which had itself introduced co-ordination into measurement programmes, recommended the creation in Europe of a centre for compiling and classifying data, making their existence known, and distributing them on request. This proposal was studied within the framework of the European Nuclear Energy Agency and its implementation took the form of the Neutron Data Compilation Centre (better known under its French abbreviation - CCDN, Centre de compilation de données neutroniques) established in May 1964 at the Centre d'études nucléaires at Saclay, France. Finally, in 1965 a co-operative arrangement was agreed between the European Nuclear Energy Agency and the United States Atomic Energy Commission for the exchange of information. We can see here in this very brief summary of the course of events, actions taking place at all the levels of co-operation which I mentioned above.

The activities of the CCDN are concentrated on the collection of references, the collection of data, the study of the data, and the collection of evaluations; the CCDN does not itself carry out evaluation.

At about the same time as the West was trying to improve the data situation the International Atomic Energy Agency had of course recognized the same problem but on a world-wide scale. The Nuclear Data Unit in Vienna was formed to aid compilation in those parts of the world not already served by previous organizations. Here activities have covered collection of references, and more recently of data, but a physics evaluation of the 2200 m/s fission data is perhaps the Unit's most widely known activity to date [4].

In the meantime the Information Centre for Nuclear Data set up in the USSR at Obninsk by the State Committee on the Utilization of Atomic Energy had made perhaps the most overall attack of any of the Centres on the problem. This Centre produces bulletins, the first of which was issued in 1964 by the Publishing House for Atomic Literature, Atomizdat [5]. This book of more than 400 pages was an up-to-date compilation covering much of the information on the interactions of neutrons, microscopic constants, empirical formulae and integral parameters used in calculations for reactors and reactor shielding. Most of the data, which had not been referred to in previous publications by Atomizdat, were obtained in 1963 to 1964, or became available at that time. A second bulletin appeared in 1965 [6] and Dr. Usachev, who is the director of this Centre along with Dr. Abramov, informed me that the third bulletin should be out in about one month's time.

The Obninsk centre produces progress reports on its activities. It centralizes in Obninsk all the CINDA reference-type activities for the USSR; it is collecting data on punched cards and shortly on magnetic tape; it produces evaluations and multigroup cross-sections, the latest result being an up-dating of the multigroup set originally produced by Abagyan et al.[7]. The Obninsk Centre is also in contact with the International Centre at Dubna and arrangements are being made for centralized collection of data.

There are further connotations in the short title of this talk. The vast quantity of data resulting from the nuclear effort and the simultaneous development and improvement of computers and long distance telecommunications have met in the recent recognition that computers are as effective in collecting information as they are in carrying out complex or long and tedious scientific calculations. The implications of this were most recently described in an issue of the Scientific American [8] which was devoted entirely to this subject. It has taken some time to recognize that computer languages used in calculation are not necessarily convenient ones for compilation but inevitably, for historical reasons, somewhat limited languages have been used in the data field.

The Sigma Center at Brookhaven foresaw this problem of the quantity of material in 1961 when they commenced to write their socalled SCISRS programme (Sigma Center Information Storage and Retrieval System) [9]. This programme, in which was invested more than two man-years of effort, was the first major programme used for handling compiled neutron data in computers. At the same time, Goldstein at Columbia University converted his CINDA index into a computeroriented system. I don't wish to go into any detail whatsoever on these systems or programmes or the interchange activities which are all fully described in the first Newsletter [10] issued by our Centre at Saclay. For those of you unfamiliar with this work Figs. 1 and 2 show a typical page of CINDA.

There are now 1400 pages like this in CINDA '66 [11], which was printed on 1 July, and the first Supplement to CINDA '66 [12] went to press today. This index is contributed to by so-called "CINDA readers" whose efforts in the USA go to DTIE, whose efforts in Europe and Japan are collected at Saclay, whose efforts in the USSR are collected at Obninsk, and whose efforts in Poland, India, Australia and recently South Africa are collected by the Vienna Data Unit. Exchange tapes between the USA and Europe regularly fly the Atlantic and the process is completely automatic up to the point where, on one occasion, we even sent the exchange material across the Atlantic and across the channel by telecommunication links straight off magnetic tape.

I should like to pay a special tribute here to our colleagues in Russia and Japan who have joined this English language activity, not only in a foreign language but especially in a different alphabet. Here is an excellent example of spontaneous international collaboration.

A page of SCISRS output is shown in Figs. 3 and 4.

The contents of the data tapes are of course never published in their entirety but are consulted by machine requests.

At this point I can refer back to computer language problems by remarking that CINDA was written in FORTRAN which makes it flexible in its exchange with other data centres, but inefficient in actual machine usage. SCISRS, on the other hand, was written in FAP for efficient use of 7094s but with the resultant inability of easy transfer to other machines. Furthermore, being written in FAP, a time-consuming, skilled and difficult process, it is difficult to alter. The analogy here is between houses built of prefabricated units and those built in brick and concrete. The former can be taken down and rebuilt at will and are perhaps cheaper to build in the first place. The latter if they need major changes must either be blown up or sunk without trace. The upstart centres, at least in Europe and Vienna, hope to benefit from this experience and from inevitable developments in languages suitable to handling data. As an example, some programmers now use the appropriate utility programmes supplied by the software services of computer manufacturers and alternative high-level languages were used to operate on the 'bits' contained in the master SCISRS library to produce an even more condensed packing of the SCISRS information and a different output arrangement from that shown in Fig. 3, as shown in the third Newsletter of the CCDN [13] (see Fig. 5). SCISRS exchange tapes too flow across the Atlantic in ever increasing quantities.

Where the amount of the material has not been great, the Vienna Unit and the Obninsk Unit have been able to join in to CINDA activities by using the facilities already set up in the USA and in Europe. The quantity of data on the other hand, which as we have agreed can only be handled by computers, poses much more serious problems of machine compatibility. Dr. Usachev has told me of the Russian attempts to solve such difficulties in close collaboration with the Vienna Data Unit who have been very successful in handling data from the extensive range of computers in use today.

And now I should like to close without forgetting about my alternative title for this talk. I am going to quote, if I may, an analogy made by Professor Goldstein in his talk "Neutrons, Nuclei and Oyster Creek" given at the Washington Conference [14].

"Perhaps an analogy will be useful (if not stretched too far). Let's say that over the past 20-25 years the two groups, those who produce cross-sections and those who use them, have been carrying on an affair with each other. And like all affairs it's had its ups and downs. Sometimes it's reached peaks of intensity, at other times the partners may not be on speaking terms, or are just bored with each other. On occasion one partner has been deeply involved, while the other has been uninterested or more concerned in a flirtation with a third party. But now the time has come to put an end to the affair and to settle down to steady respectable matrimony, each sharing a continuous and clear responsibility for the common good. And if matrimony bespeaks much that is humdrum and routine, it also implies a lasting and constructive attachment.

The change to domesticity always requires some alterations in attitude. For example, the in-laws (you know who they are) will have to give their whole-hearted, sincere, blessing to the union. They may have to decrease a bit of their spending on glamorous status symbols and help set up the couple with the more down-to-earth (but still expensive) equipment with which they need to furnish house. An they 'd better not be stingy - many a marriage has gone on the rocks for such a cause."

Professor Goldstein ended by modestly remarking that it was not for him "to stand on the platform and pronounce our couple "man and wife", but here and now may be as good a time as any to get accustomed to the idea".

I would like to stretch this analogy just a little further and to indicate how we might at least get to the point of publishing the bans. He referred earlier in his talk to two new types of professional activity, the old activities were those of the measurers and the users; the new types of middle-men as he called them were of course the compilers and the evaluators and perhaps now you will see my point. They of course are the progeny, spawned by the illicit and protracted union of their erstwhile respectable parents. Like all bastards of course we are looked down upon by the respectable members of the community, including our parents. But we all know that despite their unruly and often ungracious treatment of their parents the appeal of young children, bastard or otherwise, has on many occasions been the causative factor in bringing their parents home and dry if a little tired and dishevilled to the bonds of matrimonial co-existence.

One final note: children accomplish little of significance in their early years but when they do begin to grow, and we all know how poisonous they are in adolescence, their output in terms of contribution to the community rapidly reaches parity with that of their parents. Most parents are indulgent in these early years but also admonitory. An analysis of

EXAMPLE OF A CINDA ENTRY

The extract from Supplement 2 to CINDA '65, given on page xxiopposite, demonstrates the kind of information which CINDA provides. As a particular example, we can discover from the final entry on the page (last two lines) that:

- an article in the Journal
- 2) Physical Review, Volume 139, page B 331
- published in July 1965
- 4) reports on an experiment, with some theoretical interpretation
- 5) carried out in a laboratory in Yugoslavia
- 6) on the reaction $\frac{48}{22}$ Ti (n,d)
- 7) at 14 MeV $(1.4 \times 10^7 \text{ eV})$. From the comment we note further that:
- this particular experiment measured the angular distribution of the deuteron group leaving the residual nucleus in the ground state,
- 9) using a counter telescope to detect the deuterons, and
- 10) a Cockroft-Walton accelerator as the neutron saurce. The second line of this entry indicates that:
- the article in Physical Review supersedes a paper given as Contribution 22 at a Conference held in Antwerp in July 1965.

If we should be interested in the actual history of the CINDA entry itself we can observe that:

- 12) a second ('supersedes') card was entered by the CCDN (Serial no. 507,517 is >500,000) referring to an entry added to the master tape
- 13) on 15th October 1965 as a result of a CINDA operation originating in
- 14) the US CINDA Center (Serial no. 24,566 is <500,000, and US Center denoted by "U") on an entry contributed by its CINDA reader for the Physical Review
- 15) Professor H. Goldstein (Reader Symbol "G").

In point of fact the CCDN had also made an entry for this paper as an Antwerp Conference contribution (the entries appear together on page 56 of the 1st Supplement to CINDA '65 dated October 15th, 1965), later deleted it and entered their second card.

					MARCH 1 1966 PAG	GE 91	TITANIUM
ELEMENT SZA	QUANTETY	ENERGY MIN MAX		E DATE TYPE LAB	REMARKS OR VALUES	ENTRY Date	ND.
TI 22 046	N. PROTON	TR 20 7	EUR 119E SUPPL	5765 COMP-REPT GEL	LISKIEN+PAULSEN.ALL DATA.XCIT FNCTN	●66D208EE	506145 C
TI 22 D46	N. PROTON	147157	NUC 23 8 112	8/65 COMP-JOUR IND	CHATTERJEE TABLE WITH REFS	651015UH	25325
					Entry appearing	for first t	ime
TI 22 047	TOTAL XSECT	25 3 70 4	ORNL3778 P69	5/65 EXPT-PROG ORL	GODD, 20 RES, CRV	651015U+	24562
TI 22 047	TOTAL XSECT	25 3 70 4	65ANTWRP 99	7/65 EXPT-CONF ORL	GODO+. PULSED VDG. GRAPH.	+660210E9	505370
T1 22 047	RESON PARAMS	30 3 57 4	ORNL3778 P69	5/65 EXPT-PRCG ORL	GOOD, WN, REDUCED WN, AVG D FOR 20 RES	651015U+	24563
TI 22 047	STRNTH FNCTN	0 57 4	ORNL3778 P69	5/65 EXPT-PRCG CRL	GOOD, REDUCED STF 5.8+2	651015U+	24564 reference
TI 22 047	STRNTH FNCTN	2 3 60 4	65ANTWRP 99	7/65 EXPT-CONF ORL	GOOD+. 2.6+9 SWAVE.	+660210E9	505371 into com-
T1 22 047	SPECT NGAMMA	THR	ORN13425 60	63 EXPT-PROG ORL	REL INTENSITY VERSUS CHANNEL NUMBER	+651202E8	504472 library
TI 22 047	N, PROTON	PILE	KE 6 37	1/63 EXPT-JOUR ROS	G SPECTR ABS METHD.NIESE+.AECTR-618	6 +660210E 8	505372
TI 22 047	N. PROTON	FISS	EANDC (E) 57 U	2/65 EXPT-PROG MUN	13.2+-1.0M8 REL TO \$32(NP)=66MB	650915EE	502738 SAC

20 7 EUR 119E SUPPL 5/65 COMP-REPT GEL LISKIEN+PAULSEN.ALL DATA.XCIT FNCTN +66020BEE 506146

7/65 EXPT-CONF ORL GOOD+. 2.6+-.4 AVERAGE LVL SPACING

8/65 COMP-JCUR IND CHATTERJEE TABLE WITH REFS

TI 22 047 N. PROTON

TI 22 047 LVL DEN LAW

N, PROTON

T1 22 047

TR

14 7

2 3 60

7

15

NUC 23 8 112

65ANTHRP 99

1

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c

Entry has appeared in a previous CINDA issue but has since been <u>changed</u>

+660210E9 505373

25324

651015UH



FIG.2. Page of a CINDA entry

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AN EXAMPLE FROM SCISRS OUTPUT

The printout of a section of the library tape, given on page 31 opposite, demonstrates the kind of information provided. The first line on the page shows that:

- 1) for a measurement in an Italian laboratory
- 2) the data is taken from a journal, augmented by a private communication,
- 3) (Physics Letters 5, page 205
- 4) June 1963).
- 5) For He⁴
- 6) the differential elastic scattering cross section
- 7) at a neutron energy 14.9 MeV
- 8) and an angle $\cos^{-1} 0.6584$
- 9) has the value 169.1 mb/steradian.
- 10) This is the most recent measurement.
- 11) A recoil counter was used
- 12) and values are normalised to the total cross section.
- 13) The comment at the foot of the page gives the reference for the value used in normalisation.

The asterisked entries for the next reference show an energy resolution of 0.44 MeV and an error in the cross section of \pm 0.030 barns.

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FIG. 4. Page from SCISRS output

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200	-3					2.00	10 L	-
210	-3					2.30	10 L	-
220	-3					2.60	10 L	-
230	- 3					2.75	11 L	-
245	-3					2+92	12 L	-
253	-3					2.99	11 L	-
262	-3					3.00	10 L	*
272	-3					2.87	12 L	-
282	-3					2.75	12 L	-

FIG. 5. Example of SCISRS output arrangement from CCDN Newsletter 3

activities in CINDA demonstrates very clearly this type of growth. In the data, which is a more difficult accomplishment, the apprenticeship has been harder and longer. The present number of data lines in the SCISRS main library is approximately 675 000. The Sigma Center in Brookhaven has collected some 575 000 of these data lines and its upstart younger brother the CCDN has collected some 100 000. Perhaps this was the reason why in Dr. Chernick's paper [3], he took the part, was it that of the admonishing parent or the in-law, and referred somewhat unkindly to the sporadic efforts here in Europe. However, I feel that if Dr. Chernick had time to look more closely at the different types of material being collected and remembered that the Brookhaven collection stems from many years of collecting activities and the vast mountains of paper which have resulted, and that the Saclay centre started from absolute scratch to the point two and a half years later where it engages fully in all its proper activities, has a full staff of professional physicists, and its own computer and programmers, that he will indulge us just a little longer, and I hope, be surprised at the final results.

So finally it comes to a plea, first to you professional physicists here who need us, to give us all the help you possibly can, and to the agencies, governments, and committees, in fact Goldstein's in-laws, to be forbearing and give us a chance to get the work done. International co-operation although more difficult is not basically different from the

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fundamental rules of treating your neighbour as you would wish to be treated yourself, or for that matter your children.

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PANEL DISCUSSION

Moderator: R. Taschek (United States of America)

Panel members: H. Goldstein (United States of America) - Session I

A. Michaudon (France) - Sessions II and III

R. Batchelor (United Kingdom) - Sessions IV and V

A. B. Smith (United States of America) - Session VI

N. Starfelt (Sweden) - Session VII

G.C. Hanna (Canada) and N.J. Pattenden (United Kingdom) - Session VIII

S. I. Sukhoruchkin (USSR) - Session IX

G.H. Kinchin (United Kingdom) - Session X.

Session I: Nuclear data requirements

H. GOLDSTEIN: Session I was concerned primarily with nuclear data requirements. Three applications were considered: thermal reactors, in paper CN-23/117 by G. H. Kinchin; fast reactors, in paper CN-23/52 by R. D. Smith and paper CN-23/14 by M. Segev, S. Yiftah and L. Gitter; and reactor shielding, in paper CN-23/118 by myself.

All papers agreed that calculation techniques had improved to the stage where basic microscopic cross-sections could soon be utilized directly; no longer would it be necessary to "adjust" the cross-sections in order to cover up inadequate calculation techniques. There is, therefore, a reason for measuring cross-sections: they will be of direct use.

All the papers dealing with reactors pointed out that there was a limit beyond which variations associated with the composition and dimensions of actual reactors could not be reduced. Accordingly, there was a limit beyond which there was no point in demanding greater accuracy of cross-section measurements; and we were rapidly approaching this limit.

In discussing thermal reactors, Dr. Kinchin concluded that reactivity was the most important single parameter. In this connection, he felt that existing data and experiments in progress came close to satisfying foreseeable reactivity data requirements; while one might continue to worry about certain problems (such as 241 Pu in the thermal region, or whether the discrepancy in $\bar{\nu}$ conceals some - as yet unknown - physical phenomenon, such as a variation in $\bar{\nu}$ with energy over small energy ranges, the general state of affairs was satisfactory.

I doubt whether Dr. Kinchin's opinion is shared by everyone involved in work with thermal or near-thermal reactors; uncertainties in the data used for burn-up calculations can have a very significant effect on the economics of such reactors. In undermoderated reactors, moreover, emphasis shifts from the thermal region to energies of the order of several electron-volts, where there are gaps and discrepancies (e.g. for ²³⁹Pu).

However, Dr. Kinchin's main conclusion – namely, that we are closer to satisfying requirements in this field than in any other – does, I believe, hold good.

PANEL DISCUSSION

Dr. Smith, surveying fast reactors, also emphasized the calculation of reactivity, which naturally directs the spotlight to the properties of the fissile and fertile materials. Here, the accuracy requirements are much more stringent than for thermal reactors, in relation both to existing data and to the measurement capabilities. The energy range is, of course, much greater, with emphasis perhaps on energies of the order of 2 MeV.

In addition to reactivity, fast reactors pose a number of serious questions, relating to safety (for example, Doppler coefficients and sodium void coefficients) and to the economics of breeding; indeed, some fast reactor study groups may consider that more emphasis should be placed on such questions than on the calculation of reactivity in cold systems. One would then encounter questions ranging from the resonance parameters of the fissionable and coolant materials, through the capture cross-sections in the keV region of most of the materials present, to the elastic and inelastic scattering of 238 U, oxygen, carbon and other nuclei present in fuel elements. Even when the requirements cannot be met by existing data, they at least appear to be within present measuring capabilities.

Reactor shielding, on the other hand, presents a different picture. The dominant energies of interest (5 MeV and above) are difficult of access, while the emphasis shifts from the familiar fission and capture cross-section to transport and scattering properties, and from the heavy to the lighter elements. The inadequacies of the data are correspondingly worse in shielding than in other nuclear applications. The economic incentives are perhaps less strong; on the other hand, the accuracy requirements are usually much less stringent than for any of the reactor core calculations.

The question naturally arises as to how far the new data presented at this Conference will help in satisfying the nuclear data requirements discussed in Session I. I hope that the other panel members will address themselves to this question in their summaries. I myself cannot help noticing the lack of new data for energies above 5 MeV.

Discussion

R. BATCHELOR: With regard to the question of making measurements at energies above 5 MeV, the techniques are available; it is simply a matter of getting down to the job. The more publicity Dr. Goldstein can give to his shielding requirements, therefore, the better the chances are of getting something done in this energy region.

Session II: Cross-sections and resonance parameters of non-fissile nuclides in the resonance energy region

Session III: Special Session: Statistical properties of resonance parameters

A. MICHAUDON: Session II opened with an account of measurements with mechanical choppers – slow choppers for low-energy neutrons and fast choppers with better resolution for neutrons of higher energy and for small samples. The latter point was illustrated by Z.K. Karalova and co-workers (CN-23/104) in studies carried out on 150 mg of 230 Th.

Several papers dealt with high-resolution measurements on structural materials and coolant fluids. For example, Rohr and coworkers (CN-23/9) have carried out measurements on 51 V, 55 Mn and 56 Fe, while groups at Harwell (CN-23/27) and Saclay (CN-23/72) have done similar work with sodium. The Harwell group's measurements indicate that the resonance at 2860 ± 15 eV has 1 = 0 and J = 1, while the resonance at 54 ± 1 keV has 1 = 1 and J = 2. However, agreement on these results has not yet been reached by the different groups of experimentalists. The Saclay measurements show in transmission narrow resonances that are also found in the capture cross-section by other groups.

There were also several papers relating to the strength function S_0 . These may be summarized by considering different aspects of this parameter.

(a) Studies of S_0 as a function of A show that S_0 values are generally in good agreement with the predictions of the optical model, except in the range 100 < A < 120, where the experimental values are consistently lower;

(b) Studies on local variations in S_0 as a function of A are being carried out by the Kurchatov group with separated isotopes of cadmium (CN-23/108) and tin (Ref.[11] of CN-23/107). For the latter element the S_0 values agree with those obtained at Oak Ridge, although they are consistently higher (the Kurchatov group makes no correction for p-resonances). They are also in agreement with the three-quasiparticle intermediate structure calculations (except perhaps for ¹²²Sn);

(c) According to results presented by the Kurchatov group (CN-23/107), the strength function S_0 is different for ^{107}Ag and ^{109}Ag , although these have the same spin. The disagreement with the results of the Saclay group (which has found the two S_0 values to be equal) may only be an apparent one, due to the fact that the Saclay measurements were made with natural silver;

(d) Variations in S_0 as a function of the spin of the compound nucleus have been found at Saclay in a systematic study (CN-23/64) on a group of six nuclei having spin $\frac{3}{2}$ ($S_{0,G=2} \simeq 2 S_{0,G=1}$). Variations have also been found for spins other than $\frac{3}{2}$ (for example, ⁷⁷Se having spin $\frac{1}{2}$), but these have still to be confirmed;

(e) Fluctuation in S_0 as a function of energy and with a period of about 70 keV has been found in cobalt at energies up to 120 keV (CN-23/65). It is also found in the total cross-section at energies up to approximately 500 keV. This result should be compared with statistical studies carried out at the Institute for Theoretical and Experimental Physics, Moscow, on a large number of isotopes ranging from ¹³C to ²⁰⁹Bi (CN-23/105), in which the period was found to be 60-70 keV. A similar period is observed in the first excited states of the nuclei (with the exception of collective levels). The same statistical study, on a large number of nuclei with smaller resonance spacing, gives a period of about 100 eV. This should be compared with measurements on ²³⁹Pu presented in Session VIII. Many of these results cannot be explained, even by means of the intermediate structure concept.

The strength function S_1 for p waves

A systematic study of S_1 has been carried out at Harwell (CN-23/36) by the high-energy average cross-section method; some approximate values for S_2 (strength function for d waves) are also given in this paper. The S_1 values for 93 Nb and 103 Rh are in good agreement with the Saclay results obtained by resonance analysis.

Distribution of level spacings

The results of measurements made by the Kurchatov group on 150 g of 107 Ag and 109 Ag (CN-23/107) are in agreement with theory (for random merging of two Wigner distributions) and conflict with the results obtained at Columbia University with natural silver. Results obtained at Saclay with 2 g of 155 Gd and 157 Gd also bring out the importance of having adequate amounts of separated isotopes and the difficulty of analysing measurements made with natural elements. No correlation is found in the Kurchatov measurements between the 107 Ag and the 109 Ag levels.

The Saclay results for ¹⁶⁹Tm $(J = \frac{1}{2})$ and ¹⁹⁷Au $(J = \frac{3}{2})$, where the number of undetected levels is probably very small, are in agreement with theory (CN-23/63). For other measurements, the effect of undetected levels may be greater; in the case of ²³⁸U, for example, it may give rise to a very considerable apparent disagreement with theory. Owing to the consistent non-detection of small spacings and the contribution of p-resonances, it is difficult to make a comparison with the single-population Wigner distribution as it should be in an even-even isotope.

Individual radiative capture transitions

Accurate measurements with Ge-Li detectors have been reported by the Saclay group (CN-23/60) for ¹⁹⁵Pt and by the Harwell group for ¹⁹⁹Hg. The two sets of results are in agreement with a distribution having one degree of freedom.

Session IV: (n, p), (n, α) , (n, 2n) reactions etc. and inverse reactions

Session V: Cross-sections and constants used as standards

R. BATCHELOR: Because of the time limitation I propose to confine my remarks to those results presented in these two sessions, which have a direct bearing on important reactor requests.

First, a few remarks on (n, p), (n, 2n) and (n, α) reactions. The Swedish group (paper CN-23/18) seem to be obtaining good data on the shape of the ⁹Be (n, 2n) reaction near threshold. This is of significance since, although the cross-section is small in this region, it has considerable weight when averaged over a reactor spectrum.

At this Conference we have been presented with more data on the so-called "threshold reactions" - (n, p), (n, α) etc. - which lead to radioactive nuclei. These reactions are of importance in dosimetry work and in crude spectral measurements. It should be noted that the absolute accuracy we have for these cross-sections is at best 5%, and in many cases it is probably worse. We are often pressed to improve the accuracy of data relating to these reactions, but I am sure that it will be a very difficult and time-consuming task to improve on the present 5%. I therefore feel that we should enquire further into how these data are going to be used before embarking on new and more accurate measurements. It is quite possible that what the user really wants is not the actual cross-section data, but the accurate calibration of the detector he uses to detect the β or γ radiation from the product nucleus. If this is the case, several steps, each of which involves an error and all of which are normally required in an absolute cross-section measurement, can be by-passed, the overall error thereby being considerably reduced. I admit that this does not provide a universal answer to the problem, but it at least provides a local solution. Before leaving this topic, it should be pointed out that there is still a lack of data in the 9-14 MeV region, mainly because, as far as I know, only one group, that at Chalk River, has made measurements on these cross-sections, using a Tandem accelerator.

I now come to the question of standards, which lies at the root of the problem of requests for precision data. In his excellent review article, J. Spaepen (paper CN-23/119) clearly outlined the present status of work and showed how far short of the requirements we were. Outside the thermal region, the area in which we are most advanced is certainly $\overline{\nu}$ -measurements. However, the standard ²⁵²Cf $\overline{\nu}$ -value is still not quite satisfactory, since the important Harwell results are a little lower than most other measurements. However, a weighted average of all the values now available clearly gets very close to the requested accuracy, and new measurements are not going to affect the situation very much unless they are extremely accurate (i.e. to within about $\frac{1}{4}$ %). I think our discussion on the ⁶Li (n, α), ¹⁰B (n, α) and ¹⁹⁷Au (n, γ) reactions made it quite clear what further measurements were required, so I will not dwell on these now. Instead, I would like to comment briefly on the requests for really precise data relating to fast reactors which both R.D. Smith (paper CN-23/52, session I) and J. Spaepen mentioned: namely the $\frac{1}{2}$ % fission cross-sections of 235 U and ²³⁹Pu and the 1% (n, γ) cross-section of ²³⁸U. We probably know the latter to between 5 and 10% and the former to about 5% once certain large discrepancies between new and older data have been cleared up. I should like to stress that the clarification of these discrepancies is an urgent problem. Having done this, however, how are we going to face up to improving the accuracy by an order of magnitude? Or are we just going to pay it lip service? Personally, I feel that big improvements could be made - probably not by a factor of 10, but maybe by a factor of 5 - but it will take a vast amount of effort, unless we achieve a technological breakthrough. How much effort - and hence money - will probably be required can be seen if we consider how much has already gone into obtaining a fission crosssection to 5%? I am not going to make any estimates here. Instead, I will conclude by raising the following points for your consideration:

- (1) I think we have an obligation to ourselves, to our sponsors and to the requesters to point out the difficulties of the task, so that its economic worth can be assessed and compared with that of, say, a set of integral experiments that would fulfil the same need;
- (2) Potential measurers should be quite clear that this is a full-time professional task which cannot be undertaken as a spare-time programmatic activity by nuclear physicists engaged mainly in fundamental research;
- (3) Because of the magnitude of the task it will be important to lighten it where possible by international exchange or collaboration.

I should like to summarize my overall impression regarding accurate cross-section constants. A curve describing the data requirements would have two thresholds: the first for thermal reactors, the second for fast reactors. The indications are that we are now close to satisfying the precision data requests for thermal reactors, but far short of the requirements for fast reactors, and, if we keep on our present course, we may never reach the target. The questions to be asked are: (i) Is it an economic proposition to try to satisfy the requirements by microscopic data measurements? (ii) If so, how are we going to set about this task?

Discussion

H. GOLDSTEIN: I should like to make two comments. The first refers to the possibility of by-passing requests for accurate measurements of threshold detector cross-sections. When threshold detectors are used for measuring fast spectra, it is possible to calibrate the entire detecting arrangement by reference to a known monoenergetic flux from a Van de Graaff generator. This is certainly the most accurate calibration method, but unfortunately a Van de Graaff generator is not always available, so that it would still be useful to know the cross-sections.

My second comment relates to the economic worth of some fast cross-section measurements. The estimated cost of the very elaborate test facility for fast reactors which has been proposed in the United States is over \$100 million. I am not saying that the fulfillment of cross-section requirements would obviate the need for such a test facility, but it might very well bring about substantial savings. It might even be better to spend the money on an Intense Neutron Generator.

H. STARFELT: I disagree with Dr. Batchelor's view that we may never satisfy the data requirements for fast reactors. I am convinced that, when we have expensive reactors producing power economically, there will be less money spent on detailed nuclear physics calculations. This means that nuclear data requirements will be reduced, so that we shall eventually be able to meet them.

J. CHERNICK: Nuclear physicists should consider the possibility of obtaining an absolute value of $\bar{\nu}$ for ²³⁵U from a multiplicative assembly. We know α for ²³⁵U quite well, and with the aid of modern computing techniques, Monte Carlo calculations and so forth we might be surprised at the smallness of the error in measuring η for fully enriched solutions and for multiplicative assemblies of this type. This problem deserves to be considered by nuclear physicists, who would introduce techniques not yet available to reactor physicists for making quantitative measurements.

R. TASCHEK (Moderator): I should like to take up Dr. Batchelor's reference to technical breakthroughs. The measurement of α was an important problem even in 1943. It was solved because a group of people wanted to detect neutrinos and measure to neutrino interaction, work which had no application in the atomic power field. The detection techniques developed by them resulted in a major breakthrough: the measurement of $\overline{\nu}$ and α for fast neutrons. Hence a development in basic physics was soon followed by application in the field of nuclear data, with the result that the situation regarding these measurements changed radically within a short period.

A. DIVATIA: I disagree with Dr. Batchelor's view that nuclear data work is a full-time activity. Practically all the nuclear data available at present were obtained by nuclear physicists as a sideline of their main work.

R. BATCHELOR: There may be nuclear data work that the nuclear physicist can do in his spare time, but not the measurement of fission cross-sections to within, say, 1%.

H. GOLDSTEIN: I should like to point out a historical parallel. The first absolute determination of the Ohm to within 1% was achieved by Maxwell at Cambridge. Other physicists, in their spare time, then improved on Maxwell's results by an order of magnitude. However, the extremely accurate determinations that we have today were performed by physicists who have specialized in such precise measurements. Perhaps developments in the nuclear data field are following a similar historical pattern.

R. TASCHEK (Moderator): Perhaps I could give a more recent example. The problem of determining in detail the nucleon-nucleon force is not now generally considered to be one in which high accuracy is required. However, recent work in connection with this problem has involved measuring an energy of about 300 kV with an accuracy of 20-30 V. This is precision measurement of the highest quality. It was not, however, directed towards some application, but towards a better understanding of nature; it therefore differs somewhat from the highaccuracy measurement of fission cross-sections. This does not mean that the high-accuracy measurement of fission cross-sections will not, at some time in the future, be required for an entirely different purpose.

Session VI: Neutron cross-sections above the resonance energy region

A.B. SMITH: Two types of problem were dealt with in this Session: (a) the provision of general, relatively qualitative nuclear data; (b) the provision of specific, high-precision data.

The acquisition of general, qualitative data, where uncertainties greater than 5% are acceptable, is largely an engineering problem; success depends on development and efficiency, rather than on research. This is particularly true at incident energies of about 1.5 MeV and less, where requests are being met rapidly and suitable techniques are being used with a high degree of efficiency. At these low energies nuclear structure is well known, and theory can and should serve to guide the experimenter as well as the evaluator.

The relative need for broad resolution data at energies of 3 MeV and above has recently increased. Here the non-elastic components form a continuum. The experiments become more difficult with increasing energy and the facilities fewer. However, the acquisition of much of the information is within the capabilities of present techniques, and again theory should satisfy many qualitative requests, particularly for elastic scattering information.

The intermediate energy region (1-3 MeV) remains a difficult one, both experimentally and theoretically. However, nuclear structure physicists are doing much to provide the information necessary for good calculations, and, as has been seen at this Conference, experimental results of significance can be obtained. Progress here would be far faster if the best of present technology were applied to the problem.

Turning to specific, high-precision data, we find a more discouraging situation. Engineering techniques are not enough; one requires all the skills of the nuclear structure theorist. For example, a detailed knowledge of inelastic scattering from ²³⁵U or of the multi-particle breakup of a light nucleus can only be acquired by full use of theory and experiment. Indeed, some of these problems are so difficult from an experimental point of view that calculation appears to be the most feasible approach for the immediate future. In some cases the difficulty resides solely in the degree of precision required (a few per cent). Such accuracies would be much more readily obtained if good neutron scattering and gamma production standards were available.

I should like to conclude my remarks with the following four points:

(1) People making requests should give far more precise information. It is not sufficient to state the angular resolution required; we also need to know the resolution required for the energy of the incident and the emerging particle;

(2) More must be done to ensure that theoretical and experimental information is made available to the user as rapidly as possible;

(3) Standard laboratories should aim to provide scattering and gamma production standards that are valid for a wide energy range;

(4) We must pay more attention to the storage, evaluation and use of the information we provide. This is essentially a problem of distribution and advertising, and unless we give considerable thought to it we may find that supply outstrips demand.

Discussion

N. STARFELT: In addition to the technical difficulties at medium energies there is the fact that no, or very few, high-priority requests are made for measurements in this energy region. Hence we do not make many measurements at these energies.

A.B. SMITH: That may be true with regard to reactors, but not with regard to shielding.

R. BATCHELOR: I should like to make two general comments. First, this particular field is likely to appeal to the fundamental nuclear physicist simply because it is one in which he can extract a fair amount of nuclear structure information, especially if he has very good resolution. Second, those interested in the high-energy region, where most data requests relate to shielding problems, are concerned mainly with practical information about the heavier elements rather than with scientific studies of their nuclear structure, of which the statistical model provides a reasonably good understanding.

Session VII: Neutron radiative capture

N. STARFELT: The main purpose of my summary is to consider how well capture data requirements for reactor calculations are being met. However, I shall also refer to radiative capture from the point of view of basic nuclear physics, if only because it will make my task a little more pleasant.

The most striking features of the present situation as I see it are:

(1) The convergence of the many different gold cross-section values;

(2) The convergence, or even merging, of the methods used at low neutron energies and the earlier methods used only at high energies.

At the Symposium on the Absolute Determination of Neutron Flux in the Energy Range 1-100 keV, held at Oxford in September 1963, there was a definite discrepancy (by a factor of about two) between different measurements of the gold cross-section in the 100-keV region, and very few of us dared to hazard an opinion as to the right answer. Today, W. P. Pönitz maintains (paper CN-23/6, session V) that we know the cross-section at 30 keV with an accuracy of about 5%, or perhaps even 2%. I am less optimistic, however. The best value deduced by Grench is some 20% above Pönitz's values, and Abramov mentioned 20% as a lower limit for the present discrepancy.

The uncertainty about the shape of the cross-section in the 1-1000 keV region is still considerable, about 15%, even if the 30-keV value should happen to be exact. I agree with Abramov that the problem is not yet solved, even if the situation has improved immensely in the last two years. The different experimental methods should be studied further and systematic errors looked for, as in the sphere transmission studies presented by D. Bogart (paper CN-23/83). This might be a task for international co-operation, perhaps through the International Atomic Energy Agency.

The second feature I mentioned earlier has resulted from the downward movement of the lower limit for Van de Graaff experiments (for instance, in the liquid scintillator work reported by D. Kompe in paper CN-23/10), and the large upward shift in the upper limit for linac experiments (for example, those reported by Block et al. in paper CN-23/126), which is now approaching 1 MeV.

I expect a large amount of useful data to emerge from the linac experiments: not only beautiful counting rate curves, but also crosssections and resonance parameters.

At this Conference, we have had keV capture data from only one linac laboratory. I should like to see greater competition between the different laboratories.

The detailed study of Γ_{γ} carried out at Saclay (paper CN-23/62) should be mentioned in this connection, as well as in relation to radioactive

capture. Here, the high intensity of the linac, together with the gammaray resolution of germanium detectors, has opened up a large field for refined measurements.

How then are capture cross-section requirements being met? I do not know all the European-American Nuclear Data Committee (EANDC) requests by heart, but I know of several cases, in addition to gold cross-sections, where the required accuracy has not been attained. Many of these cross-sections are now being measured by sufficiently accurate methods (for instance, on the Rensselaer Polytechnic Institute's linac), but others, especially those for the uranium and plutonium isotopes, are difficult to measure, while the requested accuracy is high. Much hard work remains to be done. Perhaps the experiments of Diven, Hemmendinger and others with bomb sources will help, especially in the case of radioactive isotopes.

Another problem relates to the cross-sections of fission products. Here cross-section calculations have been performed for a large number of isotopes, with considerable success. Comparisons with thermal values and resonance integrals have lent support to the results, but more experiments are needed.

For neutron energies above 1 MeV little progress has been reported. Here the interest from the point of view of reactors is not so great. We have heard of several fits of calculated cross-sections to experiments, but I doubt whether theoretical extrapolation is useful in obtaining crosssections at higher energies, where the reaction mechanism is complicated by direct and semi-direct capture. I would like to see more experiments and theoretical work relating to the region about 1 MeV. There are theories for estimating cross-sections in this region, but they are of little use in practice.

The last three years have seen great progress. My guess is that, in three years' time, most of the easy or moderately easy problems will have been solved, while a few difficult ones of high priority and entailing high accuracy will still be far from solved.

I should like to conclude by posing a number of general questions. First, how can we solve these difficult problems? I do not know the answer, but I am sure that the solutions will be found in the most advanced and well-equipped laboratories already in the front line.

'Second, what can those of us do who are working in small laboratories with limited budgets? Could international collaboration help us to solve the difficult problems? I ask these questions at this Conference in the hope of obtaining an answer.

Finally, what shall we do when we have met the nuclear data requests for reactors? Work in atomic energy laboratories has contributed much to basic science, while basic science has contributed much to the development of atomic energy. We must find a way of continuing with basic physics, including nuclear physics, in the atomic energy laboratories. One possibility is to move in the direction of material sciences; it is a field where basic research is needed and where important improvements in reactor technology could result. Moreover, it is one where many of the techniques and methods developed in nuclear physics are applicable. Perhaps you can suggest other ways of ensuring that basic physics has a place in atomic energy laboratories.

Session VIII: Cross-sections and parameters of fissile nuclides (Part 1)

G. C. HANNA: The first four papers of Session VIII relate to the thermal neutron cross-sections and parameters of fissile nuclei. This is one area where the reactor designers are, or should be, just about satisfied with the accuracy of the data that are available. Some of the results presented in papers (CN-23/21 and CN-23/2 were in a sufficiently final form for inclusion in the Westcott et al. least-squares fit, but the new determination of α for ²⁴¹Pu by Cabell has an important effect on the results for ²⁴¹Pu as Westcott showed in the discussion. One of the more unsatisfactory features of these data so far has been the wide spread in the reported values of the ²³⁹Pu fission cross-section. Although it is probably still true that the most reliable value of this quantity is obtained from the absorption cross-section and the measured values reported by P. H. White et al. in paper CN-23/58 are in such good agreement with the indirect value.

Another, more important, discrepancy has also existed for some time, and unfortunately it shows no signs of disappearing. I refer to the value of $\overline{\nu}$ for the spontaneous fission of 252 Cf and especially to the very accurate and careful measurement carried out by Colvin and Sowerby, which is lower than other values by an amount ($\sim 2\%$) that is not explicable in terms of the errors quoted for the different measurements. This situation was discussed during Session V, but I refer to it again here because of the discrepancy, stressed by D.W. Colvin in paper CN-23/33, between his values of $\bar{\nu}$ for ²³³U and ²³⁵U and those obtained from η -measurements via the α -measurements reported by M. J. Cabell (paper CN-23/21) and C.B. Bigham et al. (paper CN-23/2). Paper CN-23/50, by J.R. Smith and R.G. Fluharty, though not orally presented, represents an important contribution to the problem of getting the most reliable information from measurements carried out in Maxwellian spectra. So far, only preliminary results are reported on the so-called "g factors", but evidently an approach of this type is most valuable.

The rest of the first part of Session VIII was concerned with measurements of prompt and delayed neutrons. Paper CN-23/77 reported new, accurate measurements of the neutron multiplicity distribution, and may represent a first step towards a new, precise measurement of $\bar{\nu}$ for ^{252}Cf .

Paper CN-23/19, on the variation of $\overline{\nu}$ with the energy of the neutrons causing fission in ²⁴¹Pu, has shown that the slope $d\overline{\nu}/dE_n$ over a broad range of energy is very similar to that observed in other nuclei. However, the papers from the Soviet Union presented in this session have drawn attention to the fine structure that exists in the $\overline{\nu}/E_n$ relation. These effects are of course important just as data, but their interpretation, in terms of a better understanding of the fission process, was also discussed in these papers and measurements were made of the mass division and kinetic energy release in order to elucidate the phenomena. Similarly in paper CN-23/102, the changes in delayed neutron emission for (n, nf) have been studied.

It is perhaps appropriate to conclude by stressing that no fully satisfactory data on fission will be provided without a proper detailed understanding of the fission process. In the past we have seen examples of measurements that have gone wrong through an incomplete appreciation of the complexities of fission, for example the angular correlations between neutrons and fragments. Possibly some of the remaining discrepancies are attributable to a similar lack of understanding.

Discussion

R. TASCHEK (Moderator): I would like to second what Dr. Hanna said about fission, which is of course basic to all this activity concerned with reactors. We still do not have a really satisfactory theory of fission, although we keep finding out new things. I must say that the Obninsk paper on the (n, n') fission of delayed neutron distribution (CN-23/102) was a surprise to me, and I am sure it will lead to interesting follow-up experiments. Of course, nuclear theory is hardly better understood than fission: we are still dealing with essentially phenomenological theories, which are not penetrating in their basic approach.

Session VIII: Cross-sections and parameters of fissile nuclides (Part 2)

N. J. PATTENDEN: May I first remind you of the papers presented in the second part of Session VIII before giving my opinions on the more significant points brought out? My views are inevitably coloured by the programme of my own particular laboratory. First, we heard J.E. Lynn, in paper CN-23/122, giving a theoretical interpretation of the fission process in terms of the channel theory. The theory still seems secure, and indeed it seems that it may now assist in the interpretation of detailed structure in the threshold region of the fission cross-sections of thorium-230, thorium-232 and uranium-238.

Next, B. H. Patrick (paper CN-23/30) and H. Derrien (paper CN-23/70) described plutonium-239 fission and total cross-sections and η -measurements over a wide energy range, and I will return to this later.

V. F. Gerasimov (CN-23/112) and C. D. Bowman (CN-23/38) then gave results of some spark chamber measurements of fission crosssections of active samples and demonstrated the real power of this particular technique. K. H. Böckhoff (CN-23/89) described total crosssection measurements on plutonium-240, similar to our work at Harwell as mentioned in paper CN-23/31. The wide energy range now available for resonance analysis with plutonium-240 is really due to the work of the Los Alamos Laboratory in providing large samples of highly enriched materials and also to the intensities of electron linacs.

A. Michaudon, in paper CN-23/123, gave an excellent review talk on the state of spin assignment measurements, and the scattering method of assigning spins was described by M. Asghar (CN-23/31) for plutonium-239 and F. Poortmans (CN-23/79) for uranium-235. Incidentally, in paper CN-23/70, which I mentioned just now, H. Derrien referred to spin assignments by sorting the $\Gamma_{\rm f}$ values into two groups. I think after Michaudon's review there is no need for me to say any more about spin assignments, as it was very comprehensive. Some of the significant points emerging from these papers are as follows:

- (1) The channel theory is vindicated and can be of assistance in describing fission cross-sections by various statistical distributions of parameters. It can also explain why average cross-sections measured over wide energy intervals need not correspond to a calculated average cross-section from a summation of contributions from individually observed resonances. The significance of this is that we must be careful in extrapolating from regions where the individual resonances are available for analysis.
- (2) There is an increasing amount of evidence that the fission cross-section between resonances is much lower than was observed in earlier measurements. The importance of this is that the previous multilevel fits made on the older data may be highly suspect, because the multilevel fits are quite sensitive to the actual cross-section values between resonances, where interference effects may be quite significant.
- (3) In the case of plutonium-239 there is now considerable evidence that the fission cross-section as a function of energy drops rather sharply in the 600-eV region, whereas the total crosssection as a function of energy does not. Now, if we assume that there are no sharp changes in the scattering cross-section as a function of energy over this region, this implies that the value of α as a function of energy becomes very large, of the. order of 2, at about this energy and stays large until about 10 kV, where it converges towards the value given by the high energy direct measurements. This is clearly a point of much significance in fast-breeder reactor design.

In conclusion, we must say that in this region we are certainly well on the way to meeting the more important requests for reactor data, particularly as regards the important plutonium-239 requirements, but the individual measurements are still not really accurate enough and, perhaps even more important, they have not been thoroughly verified by independent measurements in many laboratories.

Discussion

R. TASCHEK (Moderator): Perhaps I should mention the Los Alamos Laboratory's role in connection with the samples of plutonium-240. It was merely the work of conversion into metal and fabrication that was performed at Los Alamos, while the production and separation were carried out at Oak Ridge.

Session IX: Comparison of fission cross-sections in the resonance energy region

S. I. SUKHORUCHKIN: In this special session cross-sections of fissionable nuclei were compared and there was a useful exchange of information on the experimental situation as regards the cross-sections

of the fissionable isotopes ²³⁵U, ²³⁹U and ²³⁹Pu. On the whole there is good agreement between the results obtained in the different laboratories, especially for fission cross-sections. This could be seen also in the integral evaluations made by W. Good's group in the Vienna compilation centre, which were circulated to participants. The discrepancies in fission cross-sections between resonances pointed out by A. Hemmendinger call for further experimental and theoretical study. The number of independent measurements, for instance nine measurements for ²³⁵U, make it possible not only to make compilations, but also to evaluate parameters using multi-level formulae. We may hope that international collaboration in this will increase and that the experimental data will become so accurate that it will even be possible to make a choice between the different approximations in multi-level formalism. At this session we heard three very good experimental papers, and it seems that we are now getting more independent measurements. In a way the situation reminds us of the First Geneva Conference, where it was possible to work out world average standard values for thermal cross-sections eliminating systematic errors and reducing statistical scatter in the data. After the work of compilation, multi-level analysis and comparison of the results I think we will be able to obtain a set of parameters that will satisfy C.R. Lubitz. You will recall that he expressed criticism regarding the scatter of the data on resonance neutron radiation widths and experimenters must agree with him as to the need for better accuracy and further measurements, especially for capture cross-sections. I think that not only reactor designers, but also nuclear physicists, will agree there is a need for further measurements of partial cross-sections of fissionable nuclei in the resonance range.

Discussion

E.G. SILVER: I have a general question addressed to all members of the panel. Dr. Hemmendinger's paper (CN-23/42) on the development of very intense sources has, so to speak, thrown a bomb into the field of measurements with linear accelerators and, perhaps, cyclotrons. It seems to be agreed that there are certain advantages in using a bomb source, as compared with the more conventional sources, for radioactive nuclei, but I believe that there are in fact considerable advantages for a wide range of measurements. It would be interesting to hear comments on this.

A. HEMMENDINGER: This is a difficult matter for me to comment on, because in any specialized branch of nuclear physics it takes a lot of work to learn how to do a job. However, it does appear to me that once we know how to make measurements of this sort it will be possible to save a lot of effort, compared with the man-hours and expenditure involved in making the same measurements with an accelerator, even on stable nuclei. However, the problem of making absolute measurements doesn't disappear just because we have this very intense pulsed source, and this is still one of the very difficult problems in reactor application. Another problem is that experiments in which it is necessary to do what amounts to coincidence measurements are probably not feasible by this method. A. MICHAUDON: For fission cross-section experiments nuclear explosions certainly constitute outstanding sources, but before choosing between a nuclear explosion and, say, a linear accelerator one should know the cost of the explosion. Are there any figures on this?

R. BLOCK: Maybe I shouldn't say this but at the Washington Conference on Neutron Cross Section Technology Iraised the very same question, and the answer was that one of the explosions costs just about as much as the new Oak Ridge linear accelerator.

R. TASCHEK (Moderator): I'm not sure this topic of cost is worth pursuing. Of course one would never use a nuclear explosion to conduct just a few experiments. Being a 4π source it is used to best advantage when there are very many experiments, and one should also remember that the signal-to-noise ratio of this source cannot be bettered by anything else at the moment. These are the advantages that should be taken into consideration.

N.J. PATTENDEN: May I ask what is the present and anticipated time resolution of the nuclear explosion time-of-flight spectrometer?

A. HEMMENDINGER: This is really a variable quantity and it depends on how ambitious you are in designing an experiment. If you insist on covering all energies from 10 eV to 10 MeV, there will be some ranges where the resolution suffers. At low energies there are no neutrons unless a moderator is used; a moderator will give a neutron lifetime of one or more microseconds, while for the neutrons of higher energy the pulse duration is less than a tenth of a microsecond. The resolution thus varies through the spectral region. One could, of course, design an experiment to maximize resolution at some energy. At 50 eV the resolution is defined by a pulse length of, say, four microseconds and a path length of two or three hundred metres depending on the actual experiment. However, at about a kilovolt or a little less there is a cross-over range, where the neutron source is no longer constituted by the moderator but by the actual explosion itself, and from then on the burst length is closer to a tenth of a microsecond.

Session X: Neutron data evaluation

G. H. KINCHIN: In Session X we seem to have reached agreement on what is meant by evaluation - the provision of a complete set of data for several interrelated parameters, usually over a fairly wide energy range. J. Chernick has also stressed the fact that partial evaluations should not be ignored. J.J. Schmidt suggested that an evaluator must be familiar with the experimental techniques used and the data being produced, with nuclear theory, computers and the needs of reactor physicists. In view of the data requirements it is desirable for an evaluator to be concerned with all the parameters for a given isotope even though it may be more convenient or more interesting for him to concentrate on a particular parameter, say the fission cross-section, for all isotopes.

The problems of the evaluator are connected to a great extent with discrepancies and inconsistencies, and we have heard of a number of these during the Conference $-\overline{\nu}$ and the gold cross-section, to mention

only two. J. J. Schmidt dwelt on the data for iron and molybdenum in his paper (CN-23/124), and I think he feels with R. Taschek that these cases are not suitable for automatic evaluation. However, there are cases where these particular problems are not important and where the methods described by A. Horsley and J. B. Parker (CN-23/24) and K. Parker et al. (CN-23/28) can be used to assemble and fit large quantities of broadly consistent data points.

Another problem for the evaluator is that of filling gaps in the experimental data. We have heard a great deal at this Conference about the application of the optical model for extending the range of data, and in paper CN-23/54, by M.F. James and J.S. Story, we heard of the use of negative and unresolved energy levels for filling in data in the thermal region.

The role of integral measurements in data evaluation has evoked considerable interest. It may perhaps be useful to try to subdivide these measurements into three groups. The first type has been accepted by evaluators for a long time, and can be exemplified by the determination of α_0 , where the energy dependence of cross-sections is well known and measurements in a Maxwellian spectrum simply gave an absolute normalization. The second type is exemplified by resonance integral measurements, such as those in paper CN-23/73 by R. Vidal and F. Roullier, and a single cross-section can be adjusted to give agreement with this type of integral measurement. I would argue that this should have been done in the case of the epithermal ²³⁵U data, but this may not now be necessary since there is no longer any discrepancy between the differential measurements and the integral measurements. Finally, there is the most difficult type of integral measurement, as discussed by Y. Reiss (CN-23/15), L.N. Usachev (CN-23/94) and K. Parker (CN-23/28), where one tries to fit many cross-sections for different materials over a wide energy range to the results of reactivity and other measurements made on assemblies containing mixtures of isotopes. If the requested accuracies really are too high for differential measurements one may have to rely on the integral approach which, although unproven, is in principle quite promising.

Of course, an evaluator needs material to work on, and we have heard something of the data compilation centres from J. Chernick and D. W. Colvin. K. Parker gave us an account of available evaluated data together with an impressive list of evaluators and we have heard something of the role of Brookhaven Sigma Center and the ENEA Neutron Data Compilation Centre in exchanging evaluations. The effort required for evaluation has been made very clear at this Conference, and we must hope for increasing international co-ordination and exchange in the evaluation field.

Finally, a general remark. This Conference has been primarily concerned with the basic measurement of quantities needed for reactor design and with the interpretation and compilation of the results of such measurements. Now, although I recollect that Los Alamos did have a gold-plated reactor at one time, I think nobody is frantically interested in another one, and perhaps too much time is devoted to what is at best a secondary standard, whereas too little time is devoted to the measurement and evaluation of data that are in fact of interest in reactor design – for instance, I think a discrepancy in a gold cross-section need not really worry us as much as a discrepancy in, say, the 235 U fission cross-section.

Discussion

R. TASCHEK (Moderator): I have one general remark which possibly runs a little bit contrary to Dr. Kinchin's last comment. There is a mutual stimulus between the interests of those who perform the rather prosaic task of measuring data and the interests of those using the data to improve our understanding of nature by way of nuclear physics. This stimulus can only be beneficial and it should be encouraged. The people in neutron physics have made a really major contribution to out understanding of the nucleus, whilst simultaneously satisfying other interests.

In conclusion, could I ask Dr. Goldberg to give a quick summary of Session XI, on international co-operation in the field of nuclear data?

M. GOLDBERG: I have little to add. I think Dr. Colvin has given us a rather thorough survey of activities pertaining to the various forms of international co-operation. These activities have been greatly intensified during the last five years, and the world of neutron crosssection users is beginning to reap the benefits. CINDA has grown from a part-time activity of one or two physicists to a truly international concern. Experimental data from all over the world have been collected at Brookhaven for the last fifteen years, and we are glad that this labour can now be shared as a result of the founding of the ENEA Neutron Data Compilation Centre, the IAEA data centre in Vienna and the USSR data centre in Obninsk. International co-operation will undoubtedly be extended to the field of evaluator data. As a veteran of many years' experience as data compiler, I should like to assure my good friend and colleague Dr. Colvin, and by inference others who are even newer in the occupation, that the ups and downs of sibling rivalry and outside pressure are very normal and should be tolerated in the interests of progress. The service we are rendering in this no-man's land between experimenters, evaluators and users is an important one, and it is only this conviction that can keep us going.

CONFERENCE ON NUCLEAR DATA MICROSCOPIC CROSS-SECTIONS AND OTHER DATA BASIC FOR REACTORS

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