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EVALUATION OF NUCLEAR CONSTANTS FOR ^{239}Pu IN THE NEUTRON ENERGY RANGE 10^{-3} eV-15 MeV

V.A. Konshin, G.V. Antsipov, L.A. Bakhanovich,
G.B. Morogovskij, Ya.Sh. Sukhovitskij

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ABSTRACT

The paper contains a brief description of the results of evaluating nuclear constants for ^{239}Pu in the energy range 10^{-3} eV - - 15 Mev. More detailed results will be presented in the Byuleten Tsentra po Yadernym Dannym.

In the thermal energy range (10^{-3} - 1 eV) the authors analyse existing experimental data for σ_a, σ_f, α and η, evaluate these values, and compare the values of η obtained by the direct method as well as from the cross-section ratio.

In the resonance energy range the authors propose a multilevel formalism allowing for interference, develop and run a programme on the "Minsk-22" computer, and analyse simultaneously three types of cross-sections σ_t, σ_f and σ_c. In the unresolved resonance energy range an algorithm is developed and the corresponding parameters for describing the cross-sections are derived.

An evaluation is performed of the total cross-section, the fission cross-section, the value $\bar{\nu}$ and the value α, along with an analysis of existing experimental data. The neutron inelastic scattering cross-section is calculated from a statistical nuclear model. A model is developed for calculating the cross-sections of the reactions (n,2n) and (n,3n). The angular distributions of elastically scattered neutrons are expanded in terms of Legendre polynomials.

1. Introduction

The whole set of nuclear constants for ^{239}Pu is evaluated for the purpose of establishing a complete file on this element for the whole energy range from 10^{-3} eV to 15 MeV, bearing in mind the nuclear data requirements of fast and thermal reactor designers.

2. Thermal neutron energy range (10^{-3} to 1 eV)

As early as the First Geneva Conference on the Peaceful Uses of Atomic Energy it was shown that there is a discrepancy between $\eta(E)$ measured by the direct method and $\eta(E)$ obtained from the ratio of the fission to the absorption cross-section. Since the detailed behaviour of $\eta(E)$ is particularly important for thermal reactors, we have paid special attention to this value.

Four quantities have been considered: the total cross-section, from which the absorption cross-section was obtained by subtracting the elastic scattering cross-section, the fission cross-section, and the quantities η and α in the energy range 10^{-3} to 1 eV.

A large amount of experimental data is available for the first three quantities, and their accuracy varies widely.

All existing information on these four parameters was collected, a detailed analysis of the experimental data was carried out and, using the PREDA program [1], the "best" curves were obtained for σ_f, σ_a, α and η.

The experimental data used in the analysis were renormalized where necessary, to thermal constants at 2200 m/sec [2].

The experimental data for σ_a, which were used as the basis of this evaluation, are the data of Gwin et al. [3], Leonard [4], Safford and Havens [5] and Bollinger et al. [6] (except for the range 0.25–0.30 eV).

The following data were considered at a lower priority: the data of Bollinger et al. [6] in the range 0.25–0.30 eV, in order to allow for possible systematic errors caused by the shape of the resolution function; the data of Palevsky [7] in the range 0.23–0.40 eV, since the background in these measurements was two times greater than the effect; the data of Havens et al. [8], which were replaced by newer data [5]; the data of Pattenden [9] (since no corrections were made for the ^{240}Pu and ^{241}Pu contained in the sample, there is a systematic uncertainty in the thickness of the sample, and a marked scatter of the points is observed in the range 0.003–0.024 eV); and the data of Nikitin et al. [10], which do not give the detailed shape of the curve.

The experimental data for σ_f, on which this evaluation is based, are the data of Gwin et al. [3], Deruytter et al. [11] and Leonard et al. [12]. The data of Bollinger et al. [6] at energies below 0.032 eV were omitted, because they have low statistical accuracy and exhibit a certain amount of scatter about a relatively smooth curve. At higher energies these data were used with a weighting factor of 1. The data of Richmond and Price [13] were taken with a slightly lower weighting factor (0.9) because the data are scattered and the cross-section at the resonance peak is reduced. The data of Adamchuk et al. [14], Tunnicliffe [15] and Auclair [16] were not taken into account.

The only direct measurements of α (^{239}Pu) in the energy range below 1 eV, performed with sufficiently high accuracy, are those of Gwin et al. [3].

We consider the most reliable direct measurements of η to be those of Seppi et al. [17], Skarsgard et al. [18] and Bollinger et al. [6], which were used with a weighting of unity. These data, which were obtained independently of each other, agree within 2%, with some odd deviations of up to 4%.

Comparison of the curve for η, obtained by direct methods, with the curve of η derived from the ratio of the cross-sections σ_f and σ_a and from α, show that good agreement exists (better than 1%) between the three curves in the range 0.02–0.24 eV. In the range 0.24–0.50 eV the curve of η, derived from the cross-section ratio and α, is systematically 3-5% higher than the curve for η obtained by direct measurements.

The evaluated curve of σ_a has an accuracy varying from 1.5 to 3% in the range 0.001–0.80 eV and from 10 to 15% in the range 0.8-1.0 eV. Below 0.02 eV there is only one set of data having sufficient accuracy, and therefore additional measurements are needed to determine the shape of the curve in this energy range.

The fission cross-section was described by a smooth curve in the range 0.001-1.0 eV. The accuracy of the fission cross-section in the range 0.02–0.2 eV is about 1.5 to 2.0%, and in the range 0.2-1.0 eV it is around 3 to 4%. In the energy range below 0.02 eV the shape of the fission cross-section curve is largely determined by the data of Leonard et al. [12] which exhibit a scatter from the smooth curve of about 3% in this range. It is necessary to refine the shape of the curve in this neutron energy range.

The value of α is determined mainly by the data of Gwin et al. [3] and has an accuracy of $\sim 6\%$ in the range 0.02–0.5 eV and 30 to 50% in the range 0.7–1.0 eV.

The accuracy of the value η is 2% at 0.02–0.24 eV, 3% at 0.24–0.5 eV, 5% at 0.5–0.8 eV and 10 to 15% in the range 0.8–1.0 eV.

3. Resonance neutron energy range

The single-level Breit-Wigner formula is a good approximation for analysing the cross-sections σ_f and σ_t only if the interference effect between resonances is weak. For ^{239}Pu this condition is fulfilled for 1^+ resonances but is not fulfilled for 0^+ resonances.

Therefore, in order to describe the cross-sections in the resonance energy range, we have used a slightly different form of the multilevel formalism and have provided simultaneous description of all three types of cross-sections σ_f, σ_t and σ_c.

It is well known that none of the existing formalisms for representing cross-sections in the resonance energy range enable a self-consistent set of parameters to be obtained for describing the reactions (n,T), (n,f), (n,γ) and (n,n). For this reason we have used an approach derived from the analysis of the Breit-Wigner and Adler-Adler formalisms.

In the case of widely-spaced levels the cross-sections described by the two formalisms should coincide, since the inter-level interference is negligible. Thus, the first term in the Adler-Adler formalism is pure Breit-Wigner. Consequently, the Breit-Wigner formalism with interference in the Adler-Adler form can be used to describe the reactions.

On the other hand, since the reaction cross-section can be written in the form [19]:

$$\sigma_{n,r} \sim \left| \sum_{\lambda} \frac{\sqrt{\Gamma_{\lambda n}\,\Gamma_{\lambda r}}}{E_\lambda - E - \frac{i}{2}\Gamma_\lambda} \right|^2, \qquad (1)$$

it is not difficult to obtain an expression allowing for interference between levels

$$\sigma_{n,r} = \pi \lambda^2 \Bigg[\sum_{\lambda} \frac{g_\lambda \Gamma_{\lambda n} \Gamma_{\lambda r}}{(E_\lambda - E)^2 + \frac{\Gamma^2}{4}} + \qquad (2)$$

$$+ \sum_{\lambda_1} g_{\lambda_1} \sum_{\lambda_2 \neq \lambda_1} \frac{\sqrt{\Gamma_{\lambda_1 n}\Gamma_{\lambda_2 n}\Gamma_{\lambda_1 r}\Gamma_{\lambda_2 r}}\left((E_{\lambda_1}-E)(E_{\lambda_2}-E) + \frac{\Gamma_{\lambda_1}\Gamma_{\lambda_2}}{4}\right)}{\left[(E_{\lambda_1}-E)(E_{\lambda_2}-E)+\frac{\Gamma_{\lambda_1}\Gamma_{\lambda_2}}{4}\right]^2 + \left[(E_{\lambda_1}-E)(E_{\lambda_2}-E)\frac{\Gamma_{\lambda_2}}{2} + (E_{\lambda_2}-E)\frac{\Gamma_{\lambda_1}}{2}\right]^2} \Bigg]$$

That is to say the second term in the Adler-Adler formalism can be expressed by Breit-Wigner parameters. However, to use this expression, it is necessary to have the interference sign for each pair of levels and therefore it is more convenient to take the second term in the Adler-Adler form which allows for the total interference effect.

In the case of self-consistent treatment of the experimental data it is necessary to introduce the following condition: the interference for the reaction (n,T) must be the sum of the interference of the reactions (n,f), (n,γ) and (n,n).

Thus the final expression for the reaction cross-section allowing for Doppler broadening takes the form

$$\sigma_{n,r} = \pi \lambda^2 \sum_{\lambda} \frac{g_\lambda \Gamma_{\lambda n} \Gamma_{\lambda r}}{\Gamma_\lambda^2} \psi(x,\theta) + \sum_{\lambda} \frac{H_{\lambda r}}{E} \chi(x,\theta) \qquad (2a)$$

The above formalism was used for simultaneous description of the following series of experimental data: for σ_f in Refs [3, 20-23], for σ_t in Refs [2, 24-27], and for σ_c in Ref. [3].

The standard energy scale was based on the resonance energies measured in experiments at Saclay [28]. The remaining experimental data were energy adjusted where necessary. The data for σ_f were renormalized with respect to the fission integral from 9 to 20 eV, which was reported in Ref. [29].

The resonance parameters obtained here are self-consistent and describe well the experimental data in the resolved resonance energy range (up to 500 eV). For purposes of comparison the Adler-Adler parameters for σ_f, σ_t and σ_c were also obtained.

4. Unresolved resonance energy range

The unresolved resonance range of neutron energies for ^{239}Pu extends from 600 eV to 25 keV. In this energy range the contribution from other states, apart from s^- and p^-, is negligible. The average resonance parameters in this energy range can be obtained by averaging the resonance parameters in the resolved resonance range and by fitting the theoretical cross-sections to the existing experimental data.

We have used both these approaches in this work. The statistical parameters which were assumed to be independent of energy were taken from data in the literature. The energy-dependent parameters were determined by the authors by choosing $\langle \Gamma_n^0 \rangle$ for $\ell = 0$, J = 0 and J = 1, and $\langle \Gamma_f \rangle$ for $\ell = 0$ and J = 1, which were determined from the experimental data for σ_t, σ_f and α.

For calculating the cross-sections in the unresolved resonance range we used the formula:

$$\overline{\sigma_x} = \frac{K}{E_0} \sum_s \frac{g_s}{\langle D \rangle_s} \cdot \frac{\langle \Gamma_n \rangle_s \langle \Gamma_x \rangle_s}{\langle \Gamma \rangle_s} \langle S_x \rangle_s \qquad (2b)$$

where s is the state with given (ℓ, J);

$g_s = \dfrac{2J+1}{2(2I+1)}$ is the statistical coefficient for a given (J, ℓ) state.

K is a constant equal to 4.09×10^6 (barn eV)

E_0 is the neutron energy (eV).

$\langle S_x \rangle_f = \dfrac{\left\langle \dfrac{\Gamma_n \Gamma_x}{\Gamma} \right\rangle_f}{\dfrac{\langle \Gamma_n \rangle \langle \Gamma_x \rangle_f}{\langle \Gamma \rangle_f}}$ is the average value of the s-factor allowing for the statistical distributions of $\langle \Gamma_x \rangle_f$ and $\langle \Gamma_n \rangle_f$

The s-factors were calculated using the so-called representative parameters [30, 31].

The statistical parameters which were assumed independent of energy in the range 600 eV - 25 keV were taken from published data. These parameters were as follwos:

$\langle D \rangle$ = 9.0 eV for ℓ = 0 and J = 0; 3.1 eV for ℓ = 0 and J = 1; 9.0 eV for ℓ = 1 and J = 0; 3.1 eV for ℓ = 1 and J = 1; 2.1 eV for ℓ = 1 and J = 2; $\langle \Gamma_\gamma \rangle$ = 41.5 MeV for s- and p-waves; $\langle \Gamma_f \rangle_{J=0}^{\ell=0}$ = 2.25 eV,

$$\langle \Gamma_f \rangle_{J=0}^{\ell=1} = 0 \qquad \langle \Gamma_n^0 \rangle_{J=0}^{\ell=1} = 1.8 \times 10^{-3} (eV)^{1/2}$$

$$\langle \Gamma_f \rangle_{J=1}^{\ell=1} = 1.0 \text{ eV} \qquad \langle \Gamma_n^0 \rangle_{J=1}^{\ell=1} = 0.62 \times 10^{-3} (eV)^{1/2}$$

$$\langle \Gamma_f \rangle_{J=2}^{\ell=1} = 1.0 \text{ eV} \qquad \langle \Gamma_n^0 \rangle_{J=2}^{\ell=1} = 0.42 \times 10^{-3} (eV)^{1/2}$$

and $f_1 = 2.0 \times 10^{-4} (eV)^{1/2}$

The neutron widths $\left\langle \Gamma_n^0 \right\rangle_{J=0}^{\ell=0}$ and $\left\langle \Gamma_n^0 \right\rangle_{J=1}^{\ell=0}$ depend on neutron energy and were determined from existing experimental data on the total cross-section for ^{239}Pu. The evaluated data for σ_t were used to obtain the energy-dependent strength function f_0 for the s-wave (allowing for the contribution of the p-wave).

Using the above parameters the widths $\left\langle \Gamma_f \right\rangle_{J=1}^{l=0}$ were determined from the experimental data for σ_f and α. Forty points were selected in the range from 300 eV to 25 keV which give a reasonably good description of the cross-sections in this range. In the energy range above 8 keV no allowance was made in the analysis for the inelastic scattering width $\Gamma_{nn'}$, which means that $\left\langle \Gamma_\gamma \right\rangle_{J=1}^{l=0}$ is obviously reduced in this range.

The parameters obtained enabled us to calculate σ_f, α and σ_t with sufficient accuracy in the unresolved resonance range.

5. The quantity α (^{239}Pu) in the range 100 eV - 15 MeV

The evaluation of α (^{239}Pu) was based on the work of Sowerby and Konshin [32], but taking into account new measurements which were reported after completion of this work - the new measurements of Gwin et al. [33], Bergman et al. [34], Kononov et al. [35], and Dvukhsherstnov et al. [36].

The results of existing measurements are not in good agreement and the only acceptable way of evaluating α in our opinion is to make a detailed analysis of the measuring technique.

On carrying out this analysis we came to the conclusion that none of the existing detectors is good enough. Ideally one needs a detector which would record 100% of fission events, otherwise the gamma ray and fission detectors are sensitive to possible variations in the characteristics of the fission process as a function of neutron energy. We believe that, until a fission detector with very high efficiency is developed, it will be impossible to perform very accurate measurements of α, unless detailed measurements are made of fission gamma rays and angular distributions of fission fragments as a function of neutron energy.

The disagreements in the results of various experiments to measure α are not caused by a difference in normalization or by variation of $\bar{\nu}$ with neutron energy. It seems to us that the main reason for the scatter of the data for α lies in errors arising in determining the background. Unfortunately, in almost all experiments which we have examined, there is not sufficient information for us to tell which experiment is less sensitive to background errors.

Delayed gamma rays from fission products are not a factor determining the accuracy of measurements at the present time. However, knowledge of these will be much more important when more accurate measurements of α become possible in the future.

Our evaluated curve for α agrees with all the data on total and partial cross-sections for ^{239}Pu, including measurements at 2 keV. We also carried out a comparison with integral measurements of α in various reactors. Within the error limits of the measurements and our evaluation, the calculated and measured integral data agree, although there are certain indications that the evaluated data should be 10% higher in the range above 30 keV, and 3-5% higher in the range below 30 keV. However, since this conclusion may be wrong due to errors in the adapted reactor spectrum as well as in the ^{239}Pu fission cross-section, we have not changed our evaluated curve.

The accuracy of α is approximately $\pm$ 10% in the range of 100 eV-400 keV. At higher energies the error increases to $\pm$ 25% at 1 MeV. Measurements of α at energies below 30 keV give quite a large degree of scatter but this is not particularly surprising, since σ_f (^{239}Pu), σ_c(^{238}U), and σ_c (^{197}Au) are known to an accuracy of no better than $\pm$ 5%, and measurements of α are much more difficult than measurements of these cross-sections.

The evaluated data for α (^{239}Pu) are not accurate enough to satisfy the requirements of reactor designers, and further measurements are clearly necessary. However, these measurements should not be performed until new measuring techniques have been developed or the old ones have been considerably improved.

6. Fission cross-section σ_f (^{239}Pu) and cross-section ratio σ_f (^{239}Pu)/σ_f (^{235}U) in the energy range 1 keV-15 MeV

The evaluation of σ_f (^{239}Pu) and of the ratio σ_f (^{239}Pu)/σ_f (^{235}U) was based on the work of Byer and Konshin [37, 38] and on the work of Byer [39]. In it we used the evaluated data for σ_f (^{235}U), published by Konshin and Nikolaev [40]. Our method of evaluation involved analysing the ratio of σ_f (^{239}Pu)/σ_f (^{235}U) and making an independent analysis of σ_f (^{239}Pu), in order to obtain

σ_f (^{235}U) from these values in the required energy range. The derived values of the ^{235}U fission cross-section were then compared with the evaluated value of σ_f (^{235}U) [40], which leads in some cases to a slight variation, within the error limits of the initial values of σ_f (^{239}Pu) and σ_f (^{239}Pu)/σ_f (^{235}U). In this way it was possible to obtain agreement between the values of σ_f (^{239}Pu), σ_f (^{239}Pu)/σ_f (^{235}U) and σ_f (^{235}U). This method was used to obtain σ_f (^{239}Pu) in the energy range 10 keV-1 MeV. In the range 1-10 keV this method is not applicable because the cross-section ratio is not known with sufficient accuracy, and it is not applicable in the range 1-15 MeV because there are no accurate absolute measurements of the ^{239}Pu fission cross-section.

The estimated systematic errors in the evaluations of σ_f (^{239}Pu) are 6% for the range 1-10 keV, 5% for 10-30 keV, 4% for 30-600 keV, 5% for 600-1000 keV, 8% for 1-2 MeV and 8-10% for 2-15 MeV. For the σ_f (^{239}Pu)/σ_f (^{235}U) ratio these errors are 8% for the range 1-10 keV, 4% for 10-30 keV, 3% for 30-100 keV, 2.5% for 100-600 keV, 3.5% for 600-1000 keV, 4% for 1-2 MeV and 6-7% for 2-15 MeV.

We concluded that it is necessary for the following measurements to be performed:

(a) In the range from 20 keV to 1 MeV at least one absolute measurement of σ_f (^{239}Pu) should be performed with an accuracy of 2-3%, especially in the range from 30-80 keV and from 200 to 600 keV, in order to confirm the data of Szabo et al. [41];

(b) In the range 1-50 keV it is necessary to carry out direct measurements of the ratio σ_f (^{239}Pu)/σ_f (^{235}U) with an accuracy of 3-4%;

(c) In the range 600-1000 keV further measurements of the fission cross-section ratio are necessary with an accuracy of 2-2.5%, in order to confirm the structure reported by Pfletschinger and Kappeler [42].

(d) From the point of view of the fast reactor programme the energy range above 1 MeV is less important and therefore it is desirable to carry out absolute measurements of σ_f (^{239}Pu) in the range 1-15 MeV with an accuracy of 4%, and to carry out measurements of the fission cross-section ratio in the range 1-2 MeV with an accuracy of 2-3%, and in the range 2-15 MeV with an accuracy of 4%;

(e) In the energy range 300-700 keV the fission cross-section of ^{235}U, derived from the ratios of σ_c (^{238}U) and σ_c (^{238}U)/ /σ_f (^{235}U) or from σ_c (^{197}Au) and σ_c (^{197}Au)/σ_f (^{235}U), has a low value and confirms the data of Poenitz for σ_f (^{235}U). This conclusion is in contradiction with the data for σ_f (^{239}Pu) and σ_f (^{239}Pu)/σ_f (^{235}U) which results in a fundamental disagreement between these six types of cross-sections. Direct measurement of the ratio σ_c (^{197}Au)/σ_f (^{239}Pu) could help to resolve this disagreement.

7. Evaluation of $\bar{\nu}$ (E) for ^{239}Pu in the energy range from thermal to 15 MeV

All existing experimental data on the energy dependence of ν (^{239}Pu) and an evaluation thereof are presented in Ref. [43]. We have used the main results of this work without alteration.

Despite the great efforts made in many laboratories around the world in recent years, and the large number of accurate measurements carried out, the situation regarding the values of $\bar{\nu}$ for the main fissile nuclei is still unsatisfactory. Among the problems awaiting solution we have singled out the following:

The dependence of $\bar{\nu}$ on energy for fissile nuclei is closely connected with the absolute value of $\bar{\nu}$ (^{252}Cf). An uncertainty of the order of 1.2% remains in $\bar{\nu}$ (^{252}Cf), and consequently also in the measurements of $\bar{\nu}$ (E) for fissile nuclei. This uncertainty is due to discrepancies between measurements with liquid scintillators and measurements performed by other methods. The two possible sources of errors, namely the yield from delayed gamma rays during fission of ^{252}Cf and the dependence of recording efficiency on the number of neutrons recorded, do not explain this discrepancy.

It is necessary to make new efforts to establish conclusively the cause of the disagreement in the values of $\bar{\nu}$ (^{252}Cf) obtained by different methods, also between direct measurements and values derived from measurements at thermal energy of η and α for ^{235}U, as well as on the ratio $\bar{\nu}$ (^{235}U)/ $\bar{\nu}$ (^{252}Cf). The "derived" value of $\bar{\nu}$ (^{252}Cf) is 1.5% higher than the latest direct measurement. The cause of this disagreement may lie in the underestimation of the experimental errors of η. Accurate and reliable measurements of $\bar{\nu}$ (^{239}Pu) in the energy range from thermal to several hundred keV do not exist. Several measurements performed in the eV-range disagree both in respect of the existence of structure in $\bar{\nu}$ and in the correlation of $\bar{\nu}$ with resonance spin. There are no measurements at all of $\bar{\nu}$ in the region above resonance and up to 200 keV. The estimated error in the values of $\bar{\nu}_p$ (^{239}Pu) is 1.2% in the range 0.01-1.2 MeV, 1.6% in the range 1.2-6.0 MeV and 1.2% in the range 6-15 MeV.

8. Evaluation of the total cross-section σ_t (^{239}Pu)

We have evaluated all existing experimental data for σ_t (^{239}Pu) in the energy range from thermal to 15 MeV. In the energy range 1 keV-1MeV the total cross-section is known to an accuracy of about 5%. The evaluated curve in the range 1-100 keV follows the data of Uttley [44]. The values of the total cross-sections calculated from the resonance parameters obtained by us agree well with Uttley's data, providing a further argument in support of the latter.

The trend of the curve of σ_t in the range 0.1-1.0 MeV is largely determined by the measurements of Cabe et al. [45], and in the range 1-15 MeV by the data of Cabe et al. [45], Foster et al. [46] and Bratenahl et al. [47]. To increase the accuracy of σ_t in the range 1 keV - 1 MeV, further measurements are necessary with an accuracy of $\sim$2%, so as to confirm the data of Uttley [44] in the range 1-130 keV and the data of Cabe et al. [45] in the range 0.1-1.0 MeV. In order to establish the shape of the curve in the range around 100 keV, it is necessary in one experiment to perform measurements of σ_t with an accuracy of $\sim$2% in the range from 70-300 keV.

9. Evaluation of the inelastic scattering cross-section $\sigma_{nn'}$

A detailed analysis was made of existing evaluations and experimental data on $\sigma_{nn'}$ (E). A calculation of the statistical nuclear model of the inelastic scattering cross-section was performed for all resolved levels of ^{239}Pu from 8 keV to 850 keV, correcting for fluctuation of the neutron widths and allowing for fission competition. In order to select the best parameters for the nuclear model, the calculated data were normalized to the experimental data of Cavanagh et al. [48] for the (57 + 76) keV, 286 keV and 331 keV levels. The data obtained for $\sigma_{nn'}$ for excitation of the individual levels are systematically lower than Prince's data [49].

10. Angular distributions of elastically scattered neutrons

We determined the coefficients for the Legendre polynomial expansion of the experimental angular distributions of elastically scattered neutrons with the aid of a special programme which performed the calculation by a scheme differing somewhat from that used in Ref. [50]. The experimental data from Refs. [51-56] were at 17 energy points. The B_i coefficients were compared with the data in Ref. [50].

11. Evaluation of the cross-sections of the reactions (n,2n) and (n,3n)

Experimental data on the cross-sections of the reactions (n,2n) and (n,3n) are completely lacking. In order to evaluate the cross-sections of these reactions, a special model was developed based on a statistical approach to nuclear reactions. The need to develop such a model resulted from the impossibility of using any of the existing models [57, 58, 49], since with practically the same premises they give cross-sections which differ from each other by a factor of 3-4. Analysis of the most frequently cited work [57] shows that the authors do not allow for competition of other processes after formation of the compound nucleus. Moreover, in Pearlstein's model [57] the inelastic scattering process makes a contribution to the (n,2n) and (n,3n) cross-sections the effect of which was not allowed for at all. Testing of this model on ^{238}U, for which sufficiently accurate experimental data exist, shows that this treatment actually leads to the results being $\sim$20% too high. Calculation of $\sigma_{n,2n}$ and $\sigma_{n,3n}$ for ^{238}U using the model we propose gives good agreement with experiment.

12. Fission neutron spectrum

We have described the fission neutron spectrum by a Maxwellian distribution with a temperature T = 1.38 MeV for zero-energy incident neutrons. The calculation allowed for variation of the spectrum with the bombarding neutron energy in accordance with Ref. [59].

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MEASUREMENT OF ALPHA FOR ^{239}Pu IN THE 3-200 keV ENERGY RANGE

P.E. Vorotnikov, V.A. Vukolov, E.A. Koltypin,
Yu.D. Molchanov, G.B. Yankov

I.V. Kurchatov Institute of Atomic Energy, Moscow, USSR

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Abstract

The energy dependence of the capture-to-fission ratio for ^{239}Pu was measured in the 3-200 keV neutron energy range. The work was performed on a pulsed electrostatic accelerator. The neutron energy was determined by the time-of-flight method. The capture gamma rays as well as the fission gamma rays and neutrons were recorded with stilbene detectors. The results obtained are presented in the table.

Introduction

The capture-to-fission (α) for ^{239}Pu is one of the most important parameters in fast reactor design. Up to now the uncertainty of this quantity in the keV neutron energy region has been quite considerable. To some extent this is due to the fact that this is a region in which the methods of measurement converge and for this reason it is important to lower the energy limit of the measurement by using methods generally employed for higher neutron energies.

Method of Measurement

The method used to measure $\alpha(E_n)$, which is based on the simultaneous recording of the gamma rays and fast secondary fission neutrons formed in a ^{239}Pu sample on absorption of primary neutrons, was similar to that used earlier for investigating $\alpha(E_n)$ of ^{235}U [1]. The source of the primary neutrons was the reaction $^{7}Li(p,n)^{7}Be$. The protons were accelerated by an electrostatic pulsed accelerator [2]. The accelerator target was a layer of metallic lithium deposited by evaporation on a tantalum base. In the 3-90 keV neutron energy range the measurements were performed with a continuous neutron spectrum and a target thickness of 20 keV. The neutron energy was measured by the time-of-flight method over a path length of about 40 cm with a total time resolution of 8 nsec. The energy scale was normalized to the resonance in aluminium at a neutron energy of 35 keV. Monoenergetic neutrons were used at neutron energies of 120 and 200 keV. In this case the target thickness was 9 keV and the time-of-flight method was used for discriminating against the background.

The sample of metallic ^{239}Pu was the same as that used by Kononov and Poletaev [3]. The total weight of the sample was 14.5 g, the thickness 2.9×10^{-3} atoms/barn and the ^{240}Pu content was 0.2%.

The fission neutrons and the capture and fission gamma rays were detected by three stilbene crystals measuring 5 x 5 cm, connected to a FEU-30 photo-multiplier. The signals from neutrons and gamma rays were discriminated by a pulse shape circuit, a check on which showed that the gamma pulses passing into the neutron recording channel did not exceed 0.01% at 3×10^4 pulses/sec. The pulses produced by neutrons with energies of 1.2-8 MeV and gamma rays with energies of 0.6-2.6 MeV served to control the time-to-pulse height converter, the signals from which were recorded in two different channel groups of an AI-4096 analyser.

The following relation is valid for a sufficiently thin sample of fissionable material ($n\sigma_{tot} \ll 1$) or for a weak energy dependence of α energy

$$\alpha = \frac{n_c}{n_f} = \left(\frac{N_\gamma}{N_n} - \frac{\mathcal{E}_{\gamma f}}{\mathcal{E}_{nf}}\right) \Big/ \frac{\mathcal{E}_{\gamma c}}{\mathcal{E}_{nf}} \qquad (1)$$

where n_c and n_f are the numbers of capture and fission events in the sample, N_γ and N_n the numbers of gamma rays and neutrons recorded, $\mathcal{E}_{\gamma c}$ the recording efficiency of neutron capture events with respect to gamma rays, and $\mathcal{E}_{\gamma f}$ and $\mathcal{E}_{nf}$ the recording efficiencies of fission events with respect to the accompanying gamma rays and neutrons. The ratio $\mathcal{E}_{\gamma f}/\mathcal{E}_{nf}$ was determined by means of an additional scintillation fission chamber containing two thin (2.5×10^{-6} atoms/barn) layers of ^{239}Pu. This chamber was placed in front of the sample and was used together with the crystal detectors to record the coincidences of fragments with gamma rays ($n_f \mathcal{E}_f \mathcal{E}_{\gamma f}$) and of fragments with fission neutrons ($n_f \mathcal{E}_f \mathcal{E}_{nf}$). Measurements of $\mathcal{E}_{\gamma f}/\mathcal{E}_{nf}$ were performed simultaneously with the other measurements throughout the experiment.

The quantity $\mathcal{E}_{\gamma n}/\mathcal{E}_{nf}$, which is a normalizing factor, was determined by comparing the experimental data obtained with a known value of α close to $E_n = 30$ keV. The reference value adopted was $\alpha_{30} = 0.277$, which is equal to the mean of the values obtained by Kononov and Poletaev [3] and by Lottin et al. [4].

Two independent methods were used for recording capture and fission gamma rays. In the first method the gamma rays were recorded in all three detectors (summation of gamma pulses from the individual detectors). In the second method the coincidences of gamma signals from two out of three detectors were recorded (coincidence method). Measurements by the summation method were performed with an accelerator pulse repetition rate of 2 MHz in the 9-60 keV neutron energy range and at E_n = 120 and 200 keV. The ratio $\varepsilon_{\gamma f}/\varepsilon_{nf}$ was 1.30 ± 0.04 and $\varepsilon_{\gamma c}/\varepsilon_{\gamma f}$ was about 1.2. With the coincidence method measurements were performed in the 3-90 keV neutron energy range with a pulse repetition rate of 1 MHz. In this case $\varepsilon_{\gamma f}/\varepsilon_{nf}$ = 0.097 ± 0.009 and $\varepsilon_{\gamma c}/\varepsilon_{\gamma f} \simeq 0.8$.

The background was measured by replacing the plutonium sample with a lead sample with equivalent primary neutron scattering. In the summation regime the background in the gamma channel was 35% at $E_n \sim 45$ keV and rose to 80% at a primary neutron energy of 10 keV. In the coincidence regime the background in this channel was 6.44 and 78% in the 40-50, 9-10 and 3-4 keV energy ranges respectively.

In the neutron channel the background was approximately the same in both regimes, amounting to 2.5, 30 and 72% in the above mentioned energy ranges.

Results and discussion

The results were corrected for fragment collimation in the layers of plutonium in the chamber and for anisotropic escape of neutrons relative to the direction of fragment divergence. This correction amounted to 10% at E_n = 75 keV, passed through zero at 30 keV, was 6% at 10 keV and reached 12% at 3 keV. Corrections for multiple neutron scattering, absorption of gamma rays and the presence of ^{240}Pu in the sample were insignificant.

The results of the measurements of $\alpha(E_n)$ are presented in the table. The errors indicated in the table correspond to measuring errors and do not include the uncertainty of the reference value of α_{30}.

Use of the coincidence method made it possible to reduce the background at low E_n and to measure α in the neutron energy ranges above and below 10 keV with one method instead of the differing methods used hitherto. In the 9-60 keV energy range, where the measurements were performed by the summation and coincidence methods, the root-mean-square values of α have been entered in the table. The agreement of the results obtained by the two methods is good in all cases. This indicates that we have allowed correctly for the background, which is essentially different in these methods.

At neutron energies above 5 keV the relative trend of our results agrees with the estimated curve of $\alpha(E_n)$ given by Sowerby and Konshin [5]. The difference in the absolute values is fully explained by the difference in α_{30} which these authors assumed to be 0.326. Below 5 keV our results rise more steeply than the curve of Sowerby and Konshin [5] but appear to agree with the absolute measurement of α at E_n = 2 keV given by Dvukhsherstnov et al. [6].

The authors wish to thank Mr. G.A. Otroshchenko for his assistance.

Table

Averaging range keV	$\alpha \pm \alpha$	Averaging range keV	$\alpha \pm \alpha$
3 - 4	I,20±0,28	I8 - 2I	0,37±0,02
4 - 5	0,96±0,I8	2I - 24	0,36±0,02
5 - 6	0,8I±0,I4	24 - 28	0,32±0,0I
6 - 7	0,69±0,II	28 - 32	0,297±0,0II
7 - 8	0,54±0,08	32 - 38	0,263±0,0II
8 - 9	0,54±0,07	38 - 45	0,25±0,0I
9 - I0	0,43±0,03	45 - 54	0,23±0,0I
I0 - I2	0,45±0,03	54 - 66	0,20±0,0I
I2 - I4	0,5I±0,03	66 - 84	0,I4±0,04
I4 - I6	0,43±0,02	II8±I3	0,I4±0,02
I6 - I8	0,39±0,02	2I8±33	0,09±0,03

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URANIUM-238 NEUTRON DATA EVALUATED FOR THE SOKRATOR LIBRARY

L.P. Abagyan, N.O. Bazazyants, L.V. Gorbacheva, A.S. Zabrodskaya, Zh.A. Korchagina, M.N. Nikolaev, L.V. Petrova, V.V. Sinitsa

Institute of Physics and Power Engineering, Obninsk.

The uranium-238 neutron data which we evaluated have now been put into the SOKRATOR library format [1]. The ^{238}U file contains 42 sections, giving data for 14 reactions in the range 0.001 eV to 15 MeV (cross-sections, angular and energy distributions). A short description of the evaluation is given below.

Resonance region. An analysis was made of the data of 21 groups of authors who determined ^{238}U resolved resonance parameters during the period 1955-1972. A comparison of the resonance energies given by the different authors showed that most had systematic errors in the energy determination. In cases where these errors were not too great, corrections were made on the basis of the high resolution resonance energy data given in Refs [3-5], which are in good agreement. Systematic errors were also found in the case of neutron and radiation widths, and these data were similarly corrected or rejected. In particular, the data of Glass [5] were disregarded in averaging Γ_γ .

The s and p level resonances were separated on the basis of the magnitude of the neutron widths and adjustments were made so that the distribution of level spacings and corrected widths of s-resonances could be brought into agreement with the theoretical values. The division adopted is in good agreement with that obtained by Lion and co-workers [6] on the basis of Dyson's $\mathcal{F}$-statistics.

It should be noted that the distribution of the corrected neutron widths of p-resonances differs greatly from the Porter-Thomas distribution (Fig. 1), most likely owing to experimental errors.

Averaging of the resonance parameter values obtained by different authors, with weighting inverse to the square of the error indicated by the authors, was performed only when the data of at least three studies were in agreement within the limits of these errors. In other cases the errors increased, thus explaining the observed scatter in the data. In practice, this meant arithmetic averaging of the results of analyses which were not contradictory and elimination of results that differed greatly from the data of the remaining authors.

The bounds of the resolved resonance regions (0.8 keV for p-levels, and 2 keV for s-levels) were taken as the limiting energies up to which the number of observed resonances formed by neutrons with given ℓ, increases linearly with energy. In the fully resolved regions there are 97 s-resonances and 104 p-resonances.

Various evaluations were used to determine the mean resonance parameters [7]. The errors in the evaluated mean resonance parameters were determined as a function of the extent of agreement between different evaluations, taking into account their accuracy.

In determining the mean parameters for p-neutrons, special attention was given to evaluating of the effect of omitting weak levels (see Fig. 2). The evaluated mean resonance parameters ($\bar{D}_{\frac{1}{2}} = 2\bar{D}_{3/2} = 20.8 \pm 0.5$ eV; $\Gamma_\gamma = 23.5 \pm 0.7$ meV;

$\overline{\Gamma}_n^{\circ} = 0.0022 \pm 0.0002$ meV$^{3/2}$; $g\overline{\Gamma}_n^{(1)} = 0.004 \pm 0.001$ meV$^{3/2}$)

allow a good description of direct cross-section evaluations in the unresolved resonance region up to 100 keV (e.g. Ref. [8]). The transmission curves measured for thick samples [9] are also well described; the mean total cross-sections obtained from these curves, however, are systematically higher than the results of the present evaluation [10]. The mean characteristics of the resonance structure of the ^{238}U cross-sections - moments of type $\langle \sigma_{tot}^{n} \; \sigma_r^{m} \rangle$ - in the range 465 eV (where practically all resonances are narrow compared with the energy losses in elastic scattering) are also described in the sub-group presentation. In this presentation the total cross-section distribution function is approximated in a narrow energy interval which nevertheless contains a few resonances by the sum of the weighted δ functions: $P(\sigma_{tot}) = \sum a_i \cdot \delta(\sigma_{tot} - \sigma_i)$.

The probabilities a_i are called "sub-group" fractions. σ_i represents the sub-group total cross-sections. Averaged reaction cross-sections σ_r are also determined for each sub-group with allowance for the correlation between the energy dependences of σ_{tot} and σ_r. The sub-group parameters were calculated from the moments $\langle \sigma_{tot} \; \sigma_r \rangle$ so that the values of these moments can be described with an accuracy not less than that given for $-5 \leq m \leq 3$ [11]. The moments of the cross-sections were calculated from the evaluated magnitudes of the resolved resonance parameters and/or from the mean resonance parameters (in the latter case with allowance for fluctuations in level width and spacing). The dependence of the sub-group parameters on energy and temperature is shown in Fig. 3.

Non-resonance region. The ^{238}U fission cross-section was taken as evaluated in Ref. [12], but corrected for the difference between the ^{235}U reference cross-section σ_f adopted there and the evaluation given in Ref. [13]. The mean fission cross-section of ^{238}U for the ^{235}U fission spectrum is 0.292 barns, i.e., considerably lower than indicated by the results of integral experiments (0.310 $\pm$ 0.005 barns). To clarify this discrepancy it will be necessary to repeat the measurements of (^{238}U) σ_f for the fission spectrum and to improve the data on σ_f of ^{235}U in the 1-3 MeV range.

The evaluated ^{238}U inelastic scattering data are given in Fig.4. We did not consider the data of Barnard and co-workers [14] sufficient justification to accept the rather inexplicable "hump" in the inelastic scattering total cross-section at 1.5 MeV, especially as one would then have no agreement between the cross-sections measured by different methods below the ^{238}U fission threshold. The recent evaluation of Smith [15] is closer to our evaluation than to that described in Refs [16 and 17]. The spectra of neutrons inelastically scattered on all the unresolved levels and the spectra of neutrons from (n,2n) and (n,3n) reactions were calculated by means of a statistical model. Fig. 4 shows nuclear temperature as a function of excitation energy.

The radiative capture cross-section was taken from the evaluation performed by Tolstikov [18]. The angular distributions of elastically scattered neutrons were taken from the data in Ref. [19]. The angular distribution of inelastically scattered neutrons was assumed to be isotropic.

A full report on the evaluation, including all the numerical data used, a description of the evaluation method, will soon be published in "Nuclear Constants".

Figure 1. Distribution of Corrected neutron widths of p-resonances in the fully resolved region.

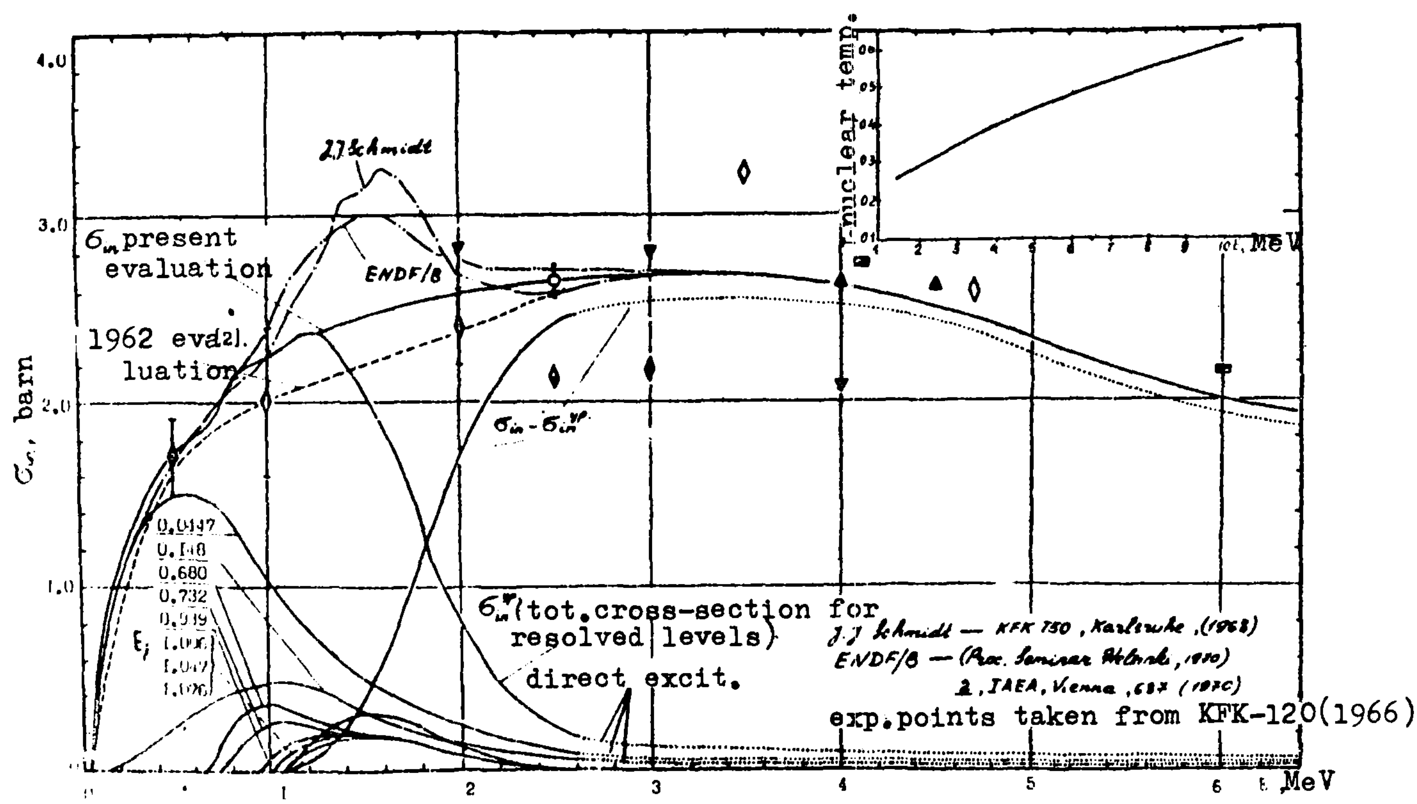
Graph not reproducible because of poor original.

Figure 2. Effect of transmission of low-lying levels on mean p-resonance spacing.

Graph not reproducible because of poor original.

Figure 3. Energies and Temperature dependence of sub-group parameters.

Graphs not reproducible because of poor original.



<u>Fig. 4.</u> Inelastic scattering cross-section for ^{238}U.

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EVALUATION OF GROUP CROSS-SECTIONS FOR PLUTONIUM-238, AMERICIUM-243 AND CURIUM-244

L.P. Abagyan, A.G. Dovbenko, S.M. Zakharova,
V.F. Kapustina, M.N. Nikolaev, L.V. Petrova

I. The accumulation of ^{238}Pu, ^{243}Am and ^{244}Cm in fuel elements of fast power reactors is interesting both from the point of view of the production of these valuable radioisotopes and in the assessment of their radiations during the transport and processing of irradiated fuel (the (α,n) reaction on oxygen in fast reactor oxide fuel, induced by the alpha activity of the above-mentioned isotopes, and the spontaneous fission of these isotopes determine the neutron activity of the irradiated fuel).

It therefore seemed important to evaluate group cross-sections for these transuranium isotopes, taking into account the new experimental information which has become available in recent years. This evaluation is described here and its results are analyzed. It should be noted that most of the experimental data used in the evaluation are preliminary, so that the evaluated results must also be regarded as preliminary.

II. In the resolved-resonance region, the detailed cross-section curves were constructed from experimental data on resonance level parameters. The calculation was performed on a computer using the URAN programme [1]; this programme employs the Breit-Wigner formula for the isolated level allowing for Doppler broadening of the resonance lines, for interference between potential and resonance scattering and (approximately) for interference between resonances. The calculated cross-sections were then compared with the experimental data we have on thermal cross-sections, the energy dependence of the cross-sections and the resonance capture intervals, and were then appropriately corrected. A preliminary evaluation was made of the experimental data and the results we considered most reliable were selected.

In the unresolved resonance region there was enough experimental information on the energy dependence of the fission cross-sections to construct a detailed $\sigma_f(E)$ curve for all the isotopes up to 10.5 MeV. The detailed capture cross-section curve for ^{238}Pu was constructed from experimental data on $\sigma_c(E)$; for the other isotopes, where such data were not available, σ_c was calculated from statistical theory. The calculation was performed on a computer with a programme using the Hauser-Feshbach statistical theory formula with the nuclear-surface penetrations taken from the optical model and the level-density formula from the Fermi gas model. The method of calculation is described in detail in Ref. [2]. The parameters of the optical potential (Woods-Saxon type) were based on the σ_{tot} calculations performed by Kolesov et al. for a wide range of energies and mass numbers [3]. The capture cross-section σ_c was calculated only up to $\sim$ 1 MeV, where the characteristics of the inelastic scattering levels are known. Above 1 MeV the relative capture cross-section curve for all the isotopes considered was taken to be the same as for ^{238}U [4] with absolute normalization to the values at $\sim$1 MeV. The σ_c given in this paper for the energy region above 1 MeV are therefore estimated values.

Table 1 gives, for each isotope, a list of references containing the experimental information used in this report, and various comments are added relating to the construction of the detailed cross-section curve for the particular isotope.

III. Table 2 gives the group constants. The method of averaging the group constants is the same as in Ref. [48] and is therefore not discussed here in detail.

Table 1

ISOTOPE		Thermal cross-sections and resonance integrals	Resonance parameters	Fission cross-sections in the region above 1 keV	Radiative capture cross-sections above 1 keV
^{238}Pu	Data from	[5 - 8]	[5,6,9-19]	[8, 19-26]	[15]
	Remarks	Values calculated from resonance parameters: σ_c=545 barn RI_c = 165 barn σ_f = 17 barn RI_f = 23 barn (not in conflict with direct measurements).	E_o values from [17] values of other parameters averaged from data in [6], [10], [17] and [19].	Evaluated curve based mainly on data in [19], which are in agreement with results in [21], [22], [24], [25] and [8].	Above $\sim$100 keV $\sigma_c(E)$ is taken to be the same as for ^{238}U, with normalization to the data of [15] for $\sim$100 keV.
^{243}Am	Data from	[5, 26-28, 36]	[5, 29-33]	[22, 34, 35]	Data available only on the energy dependence of the reaction $^{243}Am\ (n,\gamma)^{244m}Am$.
	Remarks	Values calculated from resonance parameters: σ_c = 75 barn RI_c = 1800 barn (not in conflict with direct measurements)	E_o values from [33]. Values of other parameters averaged from data in [29], [31] and [33].	Evaluated curve based on data in [34], which are in agreement with [22] and [35].	Up to $\sim$1 MeV $\sigma_c(E)$ is calculated from statistical theory. Above 1 MeV $\sigma_c(E)$ is taken to be the same as for ^{238}U with normalization to the calculated value for $\sim$1 MeV.
^{244}Cm	Data from	[27, 37, 38]	[5, 39-44]	[22,42,45-47]	No data available
	Remarks	Values calculated from resonance parameters: σ_c=12.6barn RI_c = 635 barn σ_f = 1.1 barn RI_f = 18 barn (not in conflict with direct measurements)	E_o values from [42]. Values of other parameters averaged from data in [39], [42] and [44].	Evaluated curve based on data in [45] and [46].	Up to 1 MeV $\sigma_c(E)$ is calculated from statistical theory. Above 1 MeV $\sigma_c(c)$ is taken to be the same as for ^{238}U with normalization to the calculated value for $\sim$1 MeV.

Table 2

Isotope / Group	Pu - 238 σ_f	Pu - 238 σ_c	Am - 243 σ_f	Am - 243 σ_c	Cm - 244 σ_f	Cm - 244 σ_c
1	2.5	0.009	2.0	0.013	2.2	0.003
2	2.2	0.015	1.4	0.028	2.0	0.007
3	2.2	0.026	1.4	0.064	2.0	0.015
4	2.2	0.054	1.4	0.17	1.9	0.04
5	2.1	0.10	0.98	0.38	2.0	0.09
6	1.6	0.21	0.09	0.59	0.8	0.23
7	0.98	0.37	0.01	0.70	0.08	0.35
8	0.70	0.68	0	0.96	0.04	0.45
9	0.66	1.2		1.4	0.04	0.7
10	0.73	1.7		2.0	0.05	1.1
11	0.81	2.6		2.7	0.06	1.4
12	0.97	3.7		3.9	0.07	2.1
13	1.2	4.7		6.0	0.09	3.3
14	1.6	5.8		9.2	0.12	5.4
15	2.5	8.2		13	0.2	8.5
16	4.6	14.4		20	0.3	12
17	5.0	29.8		32	0.3	11
18	4.6	10.7		45	0.4	15
19	0.1	0.5		63	1.6	22
20	2.5	52		100	0.92	34
21	1.5	6.4		140	19.1	701
22	1.6	47.5		66	0.07	2.7
23	0.2	5.5		1750	0.05	2.0
24	0.6	19		80	0.06	2.3
25	2.1	58		42	0.08	3.0
26	17	545		75	1.1	12.6

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STUDY OF PHOTONEUTRON REACTIONS CLOSE TO THE THRESHOLD

A. I. ABRAMOV

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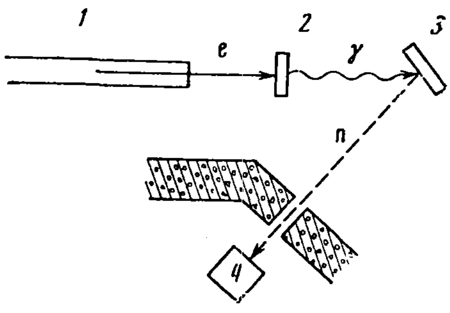
The last few years have seen the development of a new method for studying the properties of atomic nuclei, based on the measurement of neutron spectra and photoneutron reaction cross-sections close to the threshold. A considerable improvement in the time-of-flight method employed for experiments of this kind has allowed for good enough energy resolution to obtain detailed information on the parameters of individual nuclear levels at excitation energies several hundred keV higher than the neutron binding energy. This paper reviews the data available on photonuclear reactions close to the threshold and points out some of the possibilities for future application of the method.

One of the first papers dealing with the analysis of photoneutron spectra close to the threshold by the time-of-flight method was published in 1963 by a team at the Massachusetts Institute of Technology [1]. This was followed by a long cycle of experiments at the Lawrence Radiation Laboratory [2-11], and similar studies were also begun at the Argonne National Laboratory [12,13]. So far the experiments of these laboratories have remained the only ones of their kind that have been extensively discussed in the scientific literature. Similar experiments are known to have been started at a number of other laboratories abroad, but detailed accounts of these experiments have not yet been published. In the Soviet Union measurements of photoneutron reaction cross-sections close to the threshold recently got under way at the Institute of Physics and Power Engineering (FEI). The results of the first series of measurements were reported at the Kiev Neutron Physics Conference [14]. In the last few months a second series of measurements was carried out, and the results are at present being processed.



Layout of an experiment on photoneutron reactions close to the threshold (explained in text).

Fig. 1

The layout for all the experiments mentioned above was essentially the same (Fig. 1). The electron beam from a pulsed accelerator (1) is aimed at the target (2), giving rise to Bremsstrahlung which causes a (γ,n) reaction in the specimen under study (3). The generated neutrons are recorded by a detector (4) located at a certain distance from the specimen. The time distribution of the pulses N(t) can easily be converted into a neutron energy spectrum $N_n(E_n)$ or into differential (γ,n) reaction cross-sections as a function of the neutron energy (or photon energy, since there is a unique correspondence between E_n and E_γ).

In most of the experiments described in Refs [1-13] linear accelerators were used as the electron source, and it was only in our own work [14] that a microtron was employed. Although the microtron is inferior to the linear accelerator in current strength and pulse length, it produces a beam with a much smaller spread energy which is of particular importance for measurements close to the threshold when one wishes to separate the transitions to the different levels of the final nucleus. In order to attain high resolution it is necessary not only to cut down the length of the accelerator pulse, but also to select detectors with the best time characteristics. The ^{10}B liquid scintillation counters used in some experiments [2,3,5,14] are quite good, but their time characteristics are such that it is impossible to obtain resolutions better than 15-30 nsec. m^{-1}. This has limited the area of research to neutron energies of the order of 20-50 keV. Thanks to the ^{235}U plate detector developed since by the Lawrence Radiation Laboratory, which correlates neutron capture events to the gamma radiation produced, and to the recoil proton detector, it has proved possible to improve the resolution considerably, in some cases to 0.6 nsec. m^{-1} [10]. This has improved the potential of the method to a considerable extent, and in fact it has been possible to raise the upper limit of the neutron energy range to 1.5-2 MeV.

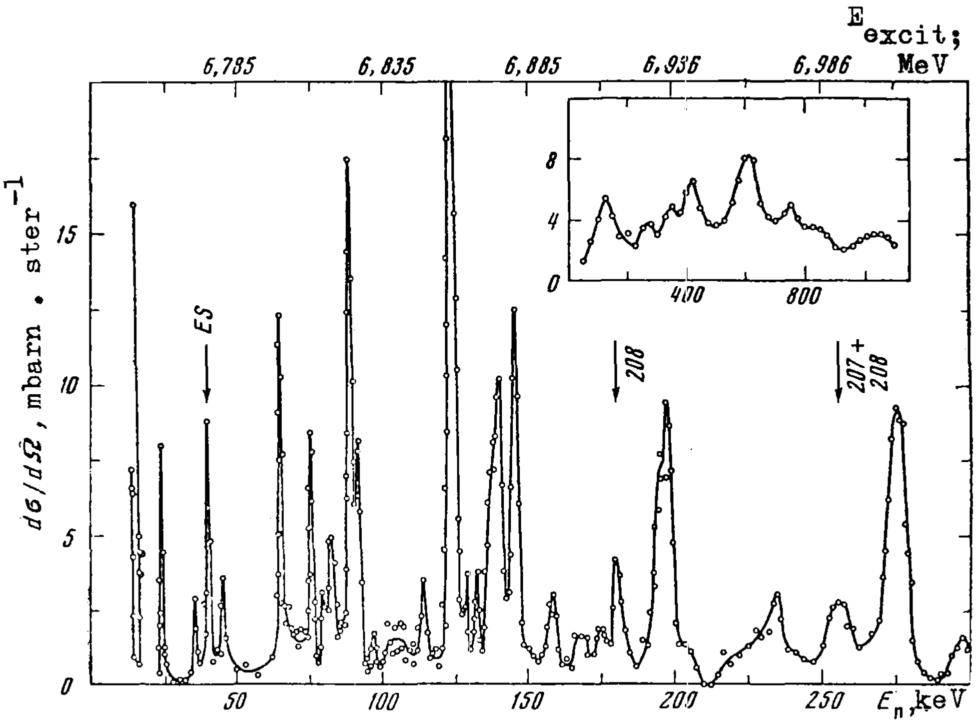


Fig. 2. Differential cross-sections for the reaction $^{207}Pb(\gamma,n)^{206}Pb$ /11/

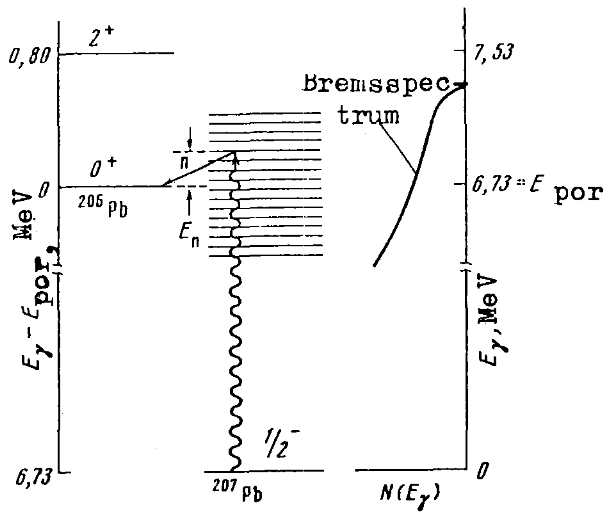


Fig. 3. Diagram showing the reaction $^{207}Pb(\gamma,n)^{206}Pb$ /Editor's note: the figure 6.73 on the left should be -6.73/.

Typical of the curves for $\sigma_{\gamma,n}$ as a function of E_n obtained by this method is the presence of sharp peaks (Fig. 2), the origin of which is explained in the following way (Fig. 3). If in the parent nucleus the levels are not closed at excitation energies higher than the neutron binding energy, the excited nuclei formed after absorption of the photon will be in discrete energy states. As a result, the energy spectrum of the neutrons emitted during their decay will also be discrete. The $\sigma_{\gamma,n}(E_n)$ curve shows moreover the same resonances that can be observed in studying the inverse neutron radiative capture reaction. The principle of detailed balance enables us to calculate, from the measured cross-sections for the (γ,n) reaction the partial (n,γ) reaction cross-section for direct radiative transition to the ground state:

$$\sigma_{n\gamma}^{o} = \sigma_{n\gamma} \frac{(\hbar k)^2}{p^2} \frac{2 I_A + 1}{2 I_{A-1} + 1}$$

The cross-sections $\sigma_{n\gamma}^{o}$ derived in this way can add much to the information obtained from direct measurements. For example, if in the reaction $A(\gamma,n)B$, the B nucleus is radioactive, then direct measurement of the $B(n,\gamma)A$ cross-section may prove very difficult or even impossible. An example of this is provided by the familiar reaction $^{56}Fe(\gamma,n)^{55}Fe$ ($T_{\frac{1}{2}}$ = 2.6 y). In such cases, measurement of the cross-sections for the (γ,n) reaction is the only source of information on $\sigma_{n\gamma}$, a point which may have both theoretical and practical importance.

Just as interesting is the study of transitions to the excited levels of the final nucleus. These transitions can be observed if the maximum bremsstrahlung energy exceeds the sum of the neutron binding energy B_n and the excitation energy for the level of interest in the final nucleus, that is, if $E_\gamma^{max} > B_n + E_{level}$. The spectral peaks for these transitions can be detected by comparing spectra obtained for $E_\gamma^{max} > B_n + E_{level}$ and $E_\gamma^{max} < B_n + E_{level}$ (in Fig. 2 one such peak is marked ES). It is easy to see that the transitions to the excited levels of the final nucleus correspond to an inverse reaction of neutron capture by the excited nucleus, although it is virtually impossible to observe this directly.

Furthermore, if we manage to separate the peaks corresponding to decays of a given state of the parent nucleus, then the ratio of their areas should yield a ratio of the probabilities to different levels of the final nucleus. These ratios are dependent on the transition energies and the spin characteristics of the initial and final states. More particularly, if I^{π} for the initial state proves closer to I^{π} for the excited level of the nucleus A = 1 than to I^{π} for its ground state, then transitions to the excited level may prove useful for comparison with the theory.

Since the transitions to excited levels are associated with relatively small neutron energies - energies at which the time-of-flight method enables us to obtain a particularly good energy resolution - we have the unique possibility of a precision measurement of the parameters of those levels lying hundreds of keV higher than the neutron binding energy (studying these levels by the traditional method of neutron spectrometry we would require considerably higher neutron energies, and the energy resolution would be much worse). It should be noted that high energy resolution is one of the fundamental advantages of the method under consideration. With a resolution of the order of 1 nsec m^{-1} the uncertainty in measuring the neutron energy in the 100 keV range is about $\pm$ 1 keV. Since in this case the nuclear excitation energy is at the $\sim$10 MeV level, to obtain the same resolution by the conventional methods of studying photonuclear reactions we would have to have $\Delta E_{\gamma}/E_{\gamma} \approx 10^{-4}$, which is approximately two orders of magnitude better than what has been obtained in practice.

As we have said, the method under consideration opens up broad prospects for measuring the parameters of individual resonances, a point which has been analyzed by Bollinger [15]. Measurements of neutron spectra at different angles to the photon beam (for example at 135° and 90° [7]) enable us to determine from the nature of the angular distribution, the spin and parity of the excited state of the parent nucleus. Along with the most probable E1 transitions, transitions of other types have been found, such as M1 and E2, in a number of cases. Furthermore, since $\Gamma_n \gg \Gamma_{\gamma_0}$ in the energy range characteristic of this type of research, direct measurement of the spectral peak width gives us Γ_n, and from analysis of the areas we get the partial radiation widths Γ_{γ_0} for direct transitions to the ground state.

Information of particular interest can be obtained by comparing measurements of (γ,n) cross-sections with measurements of σ_t and $\sigma_{n\gamma}$ by neutron spectrometry. Differences in the spin characteristics of the parent nuclei may result in large differences in the resonance structures observed. For example, in elastic scattering of neutrons on ^{56}Fe nuclei (which determine the shape of the $\sigma_t(E_n)$ curve) s-resonances of the ^{57}Fe nucleus with $I^{\pi} = \frac{1}{2}^{+}$ predominate, whereas in the $^{57}Fe\ (\gamma,n)^{56}Fe$ reaction we find that the levels excited with maximum probability are those corresponding to E1 transitions from the ground state of the ^{57}Fe nucleus; hence levels with $I^{\pi} = \frac{1}{2}^{+}$ and $3/2^{+}$ are equally probably, and the structure of the curve is considerably finer.

Averaging of the measured parameters for individual resonances makes it possible to find the radiation strength functions $S_{\gamma o} = \frac{\langle \Gamma_{\gamma_0} \rangle}{\langle D \rangle}$ for transitions with different multipolarity (E1, M1, E2 etc.). As an example we can mention the work of Baglan, and co-workers [10], who found $S_{\gamma o}$ values for ^{56}Fe, ^{57}Fe, ^{52}Cr and ^{53}Cr.

A number of publications summarize data on measurements of the principal resonance parameters E_o, I^{π}, Γ_{γ_0}, g, and so forth, for a fairly large number of levels in different nuclei (^{25}Mg, ^{52}Cr, ^{53}Cr, ^{56}Fe, ^{57}Fe, ^{206}Pb, ^{207}Pb, ^{208}Pb, etc.) in the neutron energy range up to 0.5-1 MeV. By comparing these results with neutron studies we can refine and broaden the information already existing on the parameters of the nuclei mentioned.

Measurements of the cross-sections for (γ,n) reactions close to the threshold have been used to detect non-resonance processes (direct and semi-direct) in the resonance energy region. If such processes exist, then interference by their amplitudes with the amplitude of the normal statistical mechanism should result in an asymmetry of the resonance peaks similar to that observed in the case of interference between potential and resonance neutron scattering. Bowman and co-workers [7] study in detail the shape of the resonance for the reaction $^{208}Pb\ (\gamma,n)\ ^{207}Pb$ at $E_n = 41$ keV. The asymmetry of this resonance (Fig. 4) can be explained by assuming a non-resonance process with a cross-section $0.8 \leq \sigma_{NR} \leq 1.2$ mbarn ster^{-1}. It should be noted that asymmetrical resonance peaks may be due to other factors (neutron scattering in the specimen, asymmetry of the detector response function, or inter-resonance interference), but a special study has demonstrated that in our case they cannot play a large role. We know of isolated attempts to detect non-resonance processes in other nuclei, but the significance of the information obtained has not been so clear.

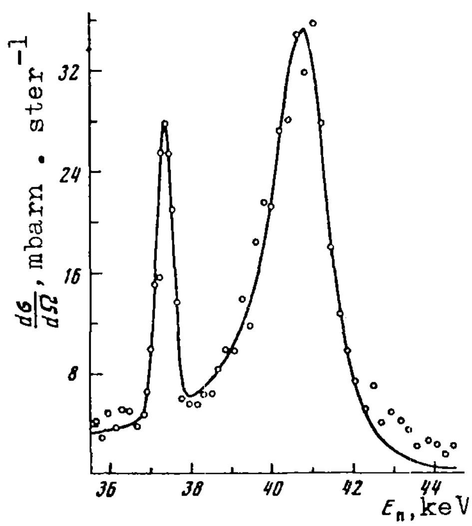


Fig. 4. Resonance for the reaction $^{208}Pb(\gamma,n)^{207}Pb$. The smooth curve corresponds to the parameters $\sigma_{NR} = 1.2$ mbarn . ster^{-1} and $\Gamma_n = 1520$ eV.

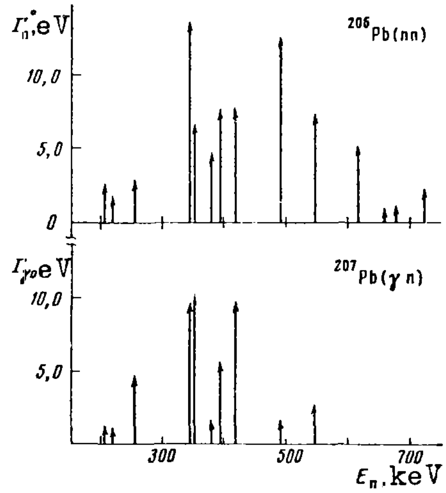


Fig. 5. Comparison of Γ_n^0 (top) and $\Gamma_{\gamma 0}$ (bottom) for the same levels of the ^{207}Pb nucleus.

Also very interesting is the possibility of using the data obtained in (γ,n) reaction studies to detect and analyze so-called doorway states. As we know, the existence of such states should mean the appearance of groups of close levels in the compound nucleus with the same I values and correlated partial widths for the various channels. Baglan and co-workers [11] quote measurements of cross-sections for the reaction $^{207}Pb(\gamma,n)^{206}Pb$ in the energy range 200-700 keV. Among other resonances, the authors discovered ten with values of $I^{\pi} = \frac{1}{2}^{+}$. The partial widths Γ_{γ_0} found for these resonances are compared in Fig. 5 with the values of the reduced neutron widths Γ_n^0 obtained by measuring total cross-sections. Correlation analysis has demonstrated that with a probability of 0.964 there definitely is a correlation between Γ_{γ_0} and Γ_n^0, and that the relevant correlation factor is +0.44. It was found from this that the radiation width of the doorway state $\Gamma_{\gamma_0}^{do}$ is contained within the limits 32 eV $\leq \Gamma_{\gamma_0}^{do} \leq$ 40 eV. Similarly studied were groups of resonances for (γ,n) reactions in the nuclei ^{53}Cr, ^{57}Fe and ^{208}Pb [9-11]. Although it has not been possible to detect a strong correlation between the partial widths for various reaction channels in all the cases studied, the occurrence of close groups of "strong" levels is considered in itself to be an indirect indication of the presence of doorway states.

One further possibility of making use of cross-section measurements for (γ,n) reactions is associated with the study of analogue states. If an analogue level lies above the neutron binding energy in the parent nucleus, it may decay with the emission of neutrons, in which case there will be a corresponding peak in the photoneutron spectrum, the position of which is easy to determine by means of the following equation:

$$E_n = Q_\beta + m_0 C^2 + E_{coul} - \delta_{np} - B_n ,$$

where Q_β is the beta decay energy for the neighbouring isobar, δ_{np} is the difference in masses between the neutron and proton, and E_{coul} is the coulomb shift of levels in the parent nucleus. Disintegration of the analogue state with the emission of neutrons occurs without conservation of isospin, of course, so generally speaking it is less likely than disintegration with the emission of a proton. However, in cases where the proton channel is closed, the probability of neutron emission may be extremely high, and the spectral peak of the analogue level may accordingly be comparable with the other spectral peaks. It is just this situation that is observed in the case of the reaction $^{25}Mg(\gamma,n)^{24}Mg$. The peak of a neutron energy of 438.5 keV corresponds to the analogue state of the ground level for the ^{25}Na nucleus with an excitation energy of (7.793 ± 0.004) MeV [8,10], which tallies well with results obtained in other studies. The peak at 514.8 keV corresponds to the analogue of the first excited level for ^{25}Na. Similar studies have been made of analogue states for ^{52}Cr and ^{56}Fe [10]. A point to bear in mind is that the possibility of determining the position of the analogue levels very accurately means that we can obtain the information needed to decide whether to introduce corrections to the formula for the Coulomb shift.

From what has been said above it will be apparent that in the few years since the publication of the first paper on the (γ,n) reaction close to the threshold there has been an extensive cycle of research from which much has been learned. The possibilities of the method are clearly promising, and a wealth of very interesting information has already been obtained, even though the range of nuclei investigated so far has been small. It would be most useful to extend the range of nuclei so as to obtain information permitting fuller and more reliable conclusions regarding the properties of nuclei and nuclear interactions.

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