INSTITUTE OF THEORETICAL AND EXPERIMENTAL PHYSICS

USSR STATE COMMITTEE ON THE UTILIZATION OF ATOMIC ENERGY

MEASUREMENT OF η AND PARTIAL CROSS-SECTIONS OF U^{235} AND Pu^{239} FOR RESONANCE NEUTRONS

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Moscow 1963

No. 147

63-5316

published in Atomnaja Energija
no.1 6(1964) p. 110

ABSTRACT

Use was made of a pulsed-cyclotron neutron spectrometer (1) to carry out measurements of η and the total cross-sections of U^{235} and Pu^{239}. Data were obtained on the energy dependence of the fission and radiative-capture cross-sections of these isotopes for neutron energies ranging from thermal to 20 eV for U^{235} and from 5 to ~ 90 eV for Pu^{239}.

This is the first time that measurements have been made of the η of U^{235} and Pu (for neutron energies over 40 eV). Spectrometer resolution was high enough to enable parameters to be determined for a considerable number of resonances.

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Apart from their purely practical significance, measurements of the cross-sections involved in the interaction of slow neutrons with fissioning nuclei are valuable inasmuch as they can throw light on the properties of the nucleus at medium excitation energies (~ 5-6 MeV) and provide direct information on the fission process itself. Thanks to the high energy resolution of spectrometers neutron-spectrometry methods are in a way unique and can be used to investigate extremely subtle effects which are beyond the reach of other nuclear-spectroscopy techniques.

Interesting information can be obtained by studying the interference effects in fission cross-sections to which attention was first drawn in (2). Cross-section interference analyses have already been made for a number of isotopes (3-6) but, owing to insufficient experimental data, they have been confined to the first resonance levels. For a reliable analysis two basic partial cross-sections have to be known, viz. the fission and the radiative-capture sections. However, while the former has been the subject of a large number of investigations (7-12), no measurements have so far been made of radiative-capture cross-sections. In the case of low neutron energies (~ up to 2-3 eV), the radiative cross-section can be worked out as the difference between the total and the fission cross-sections. At higher energies, however, this is not possible because of the fact that installations with different resolutions are generally used to measure these cross-sections.

With a view to undertaking a detailed study of the energy dependence of the partial cross-sections of fissioning nuclei over a wider energy range, use has been made of a pulsed-cyclotron neutron spectrometer (1) to obtain measurements of η and the total cross-sections of U^{235} and Pu^{239}. Once these parameters are known, it is possible to obtain the fission and radiative-capture cross-sections since at low neutron energies resonance scattering can be disregarded. Measurements were made over a neutron-energy range of 0.03 eV - 20 eV for U^{235} and 5 eV - 100 eV for Pu^{239}, and it was possible to carry out an interference analysis of the fission cross-sections for a considerable number of resonances (13, 14). The measurement methods employed and the results obtained are quoted and briefly discussed below.

Measurement Methods

For the various energy ranges investigated the resolution of the neutron spectrometer varied from 0.02 µsec/m to 0.8 µsec/m. For measurement work use was made of a 2048-channel time analyser with electrostatic memory tube; channel width was 0.2 µsec (12).

The value of η was determined by recording the secondary neutrons; in the case of the total cross-sections, transmission measurements were carried out on models, with good geometrical arrangement. Recording of the slow neutrons was performed by means of a scintillation counter with $B_{10}H_{14}$ + ZnS(Ag) (15) used as crystal. For secondary-neutron detection use was made of a similar counter with a paraffin-zinc-sulphide alloy employed as crystal, and also a scintillation counter with pulse-shape particle differentiation.

The total cross-section was determined by means of the expression:

$$\sigma_t = -\frac{1}{n}\ln T \qquad (1)$$

where T is the transmission of the sample

n is the thickness of the sample (atoms/barn).

For the fission cross-section the calculations were as follows. N_f - the number of secondary neutrons recorded by the detector - can be linked with the cross-section values by means of the following equation:

$$N_f \sim v\cdot N\cdot\sqrt{E}\cdot(1-T)\left\{\frac{\sigma_f}{\sigma_t} + \frac{\sigma_{sc}}{\sigma_t}(1-K_1)\left[\frac{\sigma'_f}{\sigma'_t} + \frac{\sigma'_{sc}}{\sigma'_t}(1-K_2)\dots\right]\right\} \qquad (2)$$

where $N\sqrt{E}$ is the slow-neutron flux,

v is the number of secondary neutrons per fission event,

$\left.\begin{matrix}\sigma_{sc}, & \sigma'_{sc}\ldots \\ \sigma_f, & \sigma'_f\ \ldots \\ \sigma_t, & \sigma'_t\ \ldots\end{matrix}\right\}$ are the scattering, fission and total cross-sections for the first and for repeated neutron interactions, and

$K_1, K_2\ldots$ is the fraction of neutrons escaping from the sample after the i^{th} scattering.

The values inside the braces take account of the contribution of neutrons which undergo repeated interactions with the target nuclei. Allowance for these terms is only of importance on the wings of resonances where the cross-sections are small and vary relatively slowly as a function of energy. It can therefore be assumed that

$$\frac{\sigma_f}{\sigma_t} = \frac{\sigma'_f}{\sigma'_t} = \frac{\sigma''_f}{\sigma''_t} = \ldots. \qquad \text{and} \qquad \frac{\sigma_{sc}}{\sigma_t} = \frac{\sigma'_{sc}}{\sigma'_t} = \frac{\sigma''_{sc}}{\sigma''_t} = \ldots.$$

and equation (2) can be rewritten in the following form:

$$N_f \sim v \cdot N \cdot \sqrt{E}\ (1-T)\, \frac{\sigma_f}{\sigma_t} \left\{1 + \frac{\sigma_{sc}}{\sigma_t}(1-K_1) + \left(\frac{\sigma_{sc}}{\sigma_t}\right)^2 (1-K_1)(1-K_2) + \ldots.\right\} \qquad (3)$$

Curves showing the functions of K_1 and K_2 - which were obtained by numerical integration - are given in Fig. 1. Limitation to the first two terms of the expansion (3) ensures an accuracy to within 2-3% where $\frac{\sigma_{sc}}{\sigma_t} < 0.5$. The radiation cross-section was obtained by subtracting the fission and scattering cross-sections from the total cross-section.

<u>U^{235}</u>. Measurements of η and the total cross-section of U^{235} were performed on a sample having a thickness of 5.6 g/cm^2 in the following, partly overlapping, neutron-energy intervals:

(1) From 0.025 to 2.3 eV with a resolution of 0.8 µsec/m;

(2) From 0.8 to 4.0 eV with a resolution of 0.2 µsec/m;

(3) From 2.1 to 20.0 eV with a resolution of 0.3 µsec/m.

The value of η was normalized directly to a thermal point. The value adopted for η thermal (= 2.07) was taken from (7). The scattering cross-section was approximated by the expression $11 + \frac{22}{(E + 2.3)^2}$ barn over the whole range of neutron energies (7). The correction for scattering was greatest in the case of parts of cross-sections between resonances; the values were 15% in the 2.5-eV region, 23% at energies around 4.5 eV and 25% at energies of 7.5-8 eV. For small cross-sections the statistical error of the measurements was not higher than 10%; it was considerably lower for large cross-sections. The background of the fast-neutron counter was between 5 and 10% inside and about 30-40% between resonances. To ensure reliable background elimination, all measurements in the energy range up to 4 eV were done with the help of an 0.1 mm Ag filter placed in the beam of neutrons. The size of the effect in the resonance at E ≈ 5 eV coincides with the background because of the fact that neutrons of this energy are completely absorbed by the filter. Cross-sections for neutron energies of 2-20 eV were measured without filter; the background was assumed to be equivalent to the effect where the U^{234} resonance was situated at 5 eV. In the case of the sample-transmission measurements, the background was determined on the basis of the absorption in the resonances of U^{235} (0.3 and 8.8 eV) and U^{234} (5 eV); the maximum background/effect ratio did not exceed 5-7%.

$\underline{Pu^{239}}$. Measurements were carried out in the neutron-energy range of 5-100 eV with a resolution of 0.02 μsec/m. For the basic measurements use was made of a sample with a thickness of 0.5 g/cm^2; for the cross-sections between resonances a sample with a thickness of 4.5 g/cm^2 was employed.

In this series of measurements, no attempt was made to obtain data in the neutron-energy range below about 5 eV. The fission cross-section was normalized on the basis of resonance 7.84 eV, for which it was assumed - from an analysis of earlier measurement data (9, 10, 11) - that $\frac{\Gamma_f}{\Gamma} = 0.465$.

The total normalization error probably does not exceed 5%.

The spontaneous fission of the Pu^{240} admixture in the sample was responsible for a relatively high background in the secondary-neutron counter (between 20 and 40% of the effect in the resonances). The calculation of the background is particularly important in the small cross-section range. In determining the inter-resonance cross-sections, therefore, special measurements were made at a higher neutron-flux intensity (approximately tenfold increase) but with a worse resolution (0.2 μsec/m). The background level for these measurements was determined by means of a 0.1-mm Ag filter. As in the case of the absorption cross-section measurements of U^{235}, the background in the slow-neutron counter did not exceed a few per cent.

The scattering cross-section was taken to be equal to 10.5 barn. In the neutron-energy range of up to 30 eV, this involved a maximum error of approximately 3% (resonance 22.2 eV). At higher neutron energies (50-100 eV) resonance scattering accounted for a considerable part of the total cross-section and it is not possible to obtain the radiation cross-section from measurements of η and σ_t.

The use of thick samples, necessary for the measurement of cross-sections by means of secondary-neutron recording, makes it difficult to determine the values of cross-sections in the resonance peaks. In order to isolate the interference effects, however, one has to know the exact trend of the cross-sections on the wings of resonances, and here the secondary-neutron recording method offers considerable advantages. The values for the cross-sections in the peaks were obtained by analysing the cross-sections by the areas method.

Results of Measurements

Figs. 2-5 show measurement data for the cross-sections and η of U^{235} for neutrons with energies of up to 20 eV. Table 1 gives values for $\frac{\Gamma_f}{\Gamma}$ as well as for the fission and radiation widths. To obtain the parameters for the first twelve levels we relied on cross-section interference analysis (14). For the other cases Γ_f was determined via the energy dependence, it being assumed that Γ_γ = 40 MeV.

Table 1

E_o, eV	$\frac{\eta}{\nu} = \frac{\Gamma_f}{\Gamma}$	Γ_f MeV	Γ_γ MeV	Notes
- 2.2		187		
0.30 ± 0.01		99	36	I
1.14 ± 0.01		129	43	I
2.04 ± 0.01		10	38	I
2.84 ± 0.04	0.80 ± 0.15	160		I
3.14 ± 0.02		79	44	I
3.60 ± 0.02		43	40	I
4.84 ± 0.02	0.095± 0.01	4		
5.45 ± 0.02	0.36 ± 0.04	23		
6.20 ± 0.08		270 ± 70		I
6.39 ± 0.03	0.21 ± 0.02	11		
7.07 ± 0.03	0.38 ± 0.03	25		
8.80 ± 0.04	0.55 ± 0.03	49		
9.26 ± 0.05	0.38 ± 0.10	25		
10.16 ± 0.05	0.15 ± 0.05	7		
11.68 ± 0.04	0.13 ± 0.04	6		
12.40 ± 0.04	0.27 ± 0.04	15		
13.00 ± 0.08	0.25 ± 0.12	13		
13.40 ± 0.05	0.1	4		
14.1 ± 0.1	0.65 ± 0.10	74		
14.5 ± 0.1	0.50 ± 0.20	40		
15.5 ± 0.1	0.45 ± 0.04	33		
16.2 ± 0.1	0.30 ± 0.03	17		
16.7 ± 0.1	0.48 ± 0.05	37		
18.2 ± 0.1	0.48 ± 0.05	37		
19.4 ± 0.1	0.38 ± 0.04	24		

Notes: I denotes that values for Γ_f and Γ_γ were obtained by cross-section interference analysis. In all other cases it was assumed that $\Gamma_\gamma = 40$ MeV and $\Gamma_f = \frac{40}{\frac{\nu}{\eta} - 1}$ MeV.

In a number of cases, taking account of inter-resonance interference has a noticeable effect on the fission widths of the levels. The nature of the width distribution is not, however, affected, and this latter can be approximated by means of Porter-Thomas curves (16) for various degrees of freedom (Fig. 6). If one takes into account the fact that the number of U^{235} levels of each momentum ($J = 3$ and 4) is approximately the same and if one takes as average values $\bar{\Gamma}_f = 20$ MeV and 116 MeV (14) for various J, it is possible to obtain an even better fit between computed distributions and experimental data.

Radiation-width values were determined for five resonances. Attention has been drawn by various investigators to the considerable variations - which cannot be accounted for in terms of experimental error - that are possible for Γ_γ for various levels of U^{235} (12). The values we have obtained for Γ_γ vary much less and the fluctuations from level to level do not exceed the limits of error to which this type of determination is subject. The constancy of the radiation width can be inferred from the nature of the spectra of the gamma rays emitted following neutron capture for the resonance levels of heavy nuclei (17). In view of the complexity of the spectra, the possibility of appreciable fluctuations in the total radiation widths is slight.

The average value of the radiation width for the five levels was $\bar{\Gamma}_\gamma = 40 \pm 2$ MeV.

The fission and radiative-capture cross-sections for Pu^{239} in the 5-27 eV range are given in Figs. 7 and 8. Figs. 9 and 10 show the total and fission cross-sections for plutonium at neutron energies of 40-100 eV. The graphs show the combined results of various series of determinations carried out with different resolutions and with samples of different thicknesses.

Table 2 gives the values of $\frac{\eta}{\nu} = \frac{\Gamma_f}{\Gamma_a}$ and $2g\Gamma_n$ for the levels resolved. The values for the fission widths were obtained, on the assumption that $\Gamma_\gamma = 40$ MeV, from the formula $\Gamma_f = \frac{40}{\frac{\nu}{\eta} - 1}$ MeV; for the resonance where $E_o = 15.5$ eV, Γ_f was obtained directly from an analysis of the fission cross-section. Comparison with the data in (11) shows a satisfactory fit with the measured results of $\frac{\Gamma_f}{\Gamma_a}$ and $2g\Gamma_\eta$ for resonances up to 30 eV.

Table 2

E_o, eV	$\frac{\eta}{\nu} = \frac{\Gamma_f}{\Gamma_a}$	$2g\Gamma_n$ MeV	Γ_f, MeV
7.84	0.465	1.33 ± 0.03	35
10.95	0.67 ± 0.03	2.15 ± 0.20	81
11.95	0.37 ± 0.03	1.35 ± 0.35	23
14.25	0.46 ± 0.10		34
14.7	0.37 ± 0.08		23
15.5	0.9 ± 0.1	1.34 ± 0.20	(1000)
17.7	0.42 ± 0.03	2.2 ± 0.3	29
22.4	0.55 ± 0.02	3.3 ± 0.3	49
26.5	0.39 ± 0.02	2.2 ± 0.4	25
41.5	0.14 ± 0.03	5.6 ± 2.0	6.5
44.5	0.09 ± 0.05	7.5 ± 1.5	4.5
47.7	0.65 ± 0.20	3.0 ± 1.0	74
50.0	0.40 ± 0.10	5.1 ± 1.5	27
52.3	0.17 ± 0.05	10.0 ± 5.0	8
57.5	0.80 ± 0.15		160
~~59.5~~	~~0.80 ± 0.15~~		160
62.5	0.90 ± 0.10		360
65.7	0.52 ± 0.10	14 ± 7	43
74.5	0.55 ± 0.10	29 ± 14	49
82.5	0.90 ± 0.10		360
85.0	0.62 ± 0.20	23 ± 10	65
90.5	0.2 ± 0.1	21 ± 10	10
95.5	0.3 ± 0.1	8 ± 4	17

Note: $f = \frac{40}{\frac{\nu}{\eta} - 1}$ MeV

Consideration of the interference effects only slightly affects the parameters for the levels of Pu^{239}. This can be accounted for by the relatively large distances between interfering levels; the only exception is the group of levels around 14-15 eV. With the help of data given in (18) and interference-analysis results it was possible to make spin identifications for most of the levels of Pu^{239} below 30 eV. The difference in the average values for the fission widths (Γ_f = 40 where J = 0 and Γ_f = 160 MeV where J = 1 (14)) is mainly attributable to the presence of two very broad levels in this energy range. Hence considerable interest attaches to cross-section measurement data in the 50-100 eV range, where there is a substantial number of strong resonances.

The fission-width distributions of levels (Fig. 11) in the 0-30 eV and 40-90 eV levels are very similar (no strong levels are observed between 30 and 40 eV). In the 40-90 eV range there are also a few resonances with very large fission widths. The values of Γ_f for broad levels (E_o = 57.5 eV, 59.2 eV, 62.5 eV and 82.5 eV) given in Table 2 may even be somewhat low. For the level at E_o = 62.5 eV, for example, a direct analysis of the energy depndence of the fission cross-section (Fig. 10) yields a value of $\Gamma_f \gtrsim 1$ eV.

Inaccurate determination of the broad-resonance parameters is mainly responsible for the error in determining the average value $\overline{\Gamma}_f$. Averaged out over the whole energy range from 0 to 90 eV, the value of $\overline{\Gamma}_f$ is 114 $\pm$ 25 MeV. The average distance between levels $\overline{D}$ = 2.9 $\pm$ 0.5 eV for neutron energies up to 70 eV; in this interval spectrometer resolution is sufficient to enable most of the resonances to be detected. The value of $\overline{D}$ is in agreement with the data given in (11); for spin states of the nucleus with J = 0 and J = 1, the value of $\overline{D}_J$ is 11.6 eV and 3.9 eV respectively.

The present paper is merely concerned with giving an account of the measurement methods employed and the results obtained; a detailed analysis of the data can be found in (13) and (14). This analysis shows that there is (1) a correlation in the amplitude signs of the reduced resonance widths, and (2) a strong spin dependence on the part of the fission width.

In conclusion, the authors would like to express appreciation for the valuable assistance they have received, in connection with the preparation and carrying out of measurements and the processing of experimental data, from V.V. Pavlov, V.V. Rotman, A.N. Soldatov and A.D. Kharitonov and also, on the mathematics side, from S. Borovlev and L.I. Panova. They would also like to thank the personnel of the cyclotron at the Institute of Theoretical and Experimental Physics for their co-operation in making these determinations possible.

Received by printer: 23 March 1963.

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Diagrams

Fig. 1

Caption: Fraction of neutrons escaping from sample after first and second scattering.

Abscissas: Thickness of samples.

Fig. 2

Caption: Value of η and fission cross-section of U^{235}. Neutron energies: 0.02 - 8 eV.

Abscissas: Neutron energy (eV).

Mid-left: barn. $eV^{\frac{1}{2}}$.

Fig. 3

Caption: Radiative-capture cross-section of U^{235}. Neutron energies: 0.02 - 8 eV.

Abscissas: Neutron energy (eV).

Top left: barn. $eV^{\frac{1}{2}}$.

Fig. 4

Caption: Value of η and fission cross-section of U^{235}. Neutron energies: 8 - 20 eV.

Abscissas: Neutron energy (eV).

Mid-left: barn. $eV^{\frac{1}{2}}$.

Fig. 5

Caption: Radiative-capture cross-section of U^{235}. Neutron energies: 8 - 20 eV.

Abscissas: Neutron energy (eV).

Top left: barn. $eV^{\frac{1}{2}}$.

Fig. 6

Caption: Fission-width distribution for 26 levels of U^{235}. Dotted line indicates computed distribution (allowance is made for difference in mean fission widths of levels of two momenta).

Ordinates: Number of levels for which $(\Gamma_f/\overline{\Gamma}_f)^{\frac{1}{2}}$ larger than indicated value.

Top right: U^{235}
$\overline{\Gamma}_f = 0.055$ eV

Fig. 7

Caption: Fission cross-section of Pu^{239}. Neutron energies: 4 - 37 eV.

Abscissas: Neutron energy (eV).

Top left: barn. $eV^{\frac{1}{2}}$.

Fig. 8	<u>Caption:</u>	Radiative-capture cross-section of Pu^{239}. Neutron energies: 4 - 27 eV.
	<u>Abscissas:</u>	Neutron energy (eV).
	<u>Top left:</u>	barn. $eV^{\frac{1}{2}}$.
Fig. 9	<u>Caption:</u>	Total cross-section of Pu^{239}. Neutron energies: 40 - 110 eV.
	<u>Abscissas:</u>	Neutron energy (eV).
	<u>Top left:</u>	barn.
Fig. 10	<u>Caption:</u>	Fission cross-section of Pu^{239}. Neutron energies: 40 - 100 eV.
	<u>Abscissas:</u>	Neutron energy (eV).
Fig. 11	<u>Caption:</u>	Fission-width distributions of Pu^{239} levels for two energy intervals (0 - 30 eV and 40 - 90 eV).