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MEASUREMENT OF ABSORPTION CROSS-SECTION AND RESONANCE PARAMETERS OF Pu<sup>239</sup>

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ABSTRACT\*

Direct count of the  $\gamma$ -rays emitted under slow neutron bombardment of Pu<sup>239</sup> was made between incident neutron energies of 7 and 550 eV. Measurements were carried out also of the sample transmission and number of fast neutrons per captured slow neutron.

The results of the measurements confirm the possibility of obtaining level parameters for  $\text{Pu}^{239}$  by the simultaneous count of secondary  $\gamma\text{-rays}$  and fast neutrons.

Values of  $\Gamma$  f/ $\Gamma$  are given for a number of levels. Spin assignments were made for five levels.

## SUMMARY

The method developed earlier for measuring  $\eta$  and the cross-sections of fissionable isotopes (1-3) was supplemented by equipment for detecting the  $\gamma$ -rays emitted by the nucleus after capture of a neutron. This made it possible to make independent measurements of the absorption cross-section, as well as measuring the total and fission cross-sections.

The cross-sections of the isotope  $Pu^{239}$  were measured in the neutron energy range from 7 eV to 550 eV; the resolution of the spectrometer was about 0.02  $\mu$ s/m.

It is concluded, on the basis of the results, that the radiative capture  $\gamma$ -ray yield and the radiation widths vary little from resonance to resonance. This justifies use of the method consisting in measuring the  $\gamma$ -ray yield to determine the parameters of the levels of Fu<sup>239</sup> at high neutron energies.

In the energy range up to 100 eV, most of the levels are resolved. Definite spin values are ascribed to a number of resonances. Further evidence is given to support the view that the average fission width of the resonances is strongly dependent on spin.

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The normal method of determining the parameters of the resonance levels of fissionable nuclei is to use the results of measurements of the total and fission cross-sections. Considerably more complete information may be obtained by carrying out at the same time independent measurements of the radiative capture cross-section or the absorption cross-section. With this aim in mind, the method developed earlier for measuring  $\eta$  and the cross-sections of figsionable isotopes (1-3) was supplemented by equipment for detecting the  $\gamma$ -rays emitted by a nucleus after capture of a neutron. Measurements of the  $\gamma$ -ray yield of the sample have been used in a number of works (4,5) to obtain the radiative capture cross-sections. The attempt to use this method for measuring the cross-sections of fissionable isotopes comes up against a number of difficulties, resulting from the fact that the total  $\gamma$ -ray yield on capture of a neutron by a fissionable nucleus consists both of radiative capture  $\gamma$ -rays and of those accompanying fission.

The relative size of the effects due to the detection of the fission and capture  $\gamma$ -rays will depend on a number of factors which it is difficult to take into account (shape of spectrum, counting efficiency, geometry of equipment), and must be determined experimentally. All that can be said is that the average number of  $\gamma$ -quanta emitted on fission is apparently a little higher than during radiative capture (6). The efficiency of detection must therefore be higher for cases of fission than for cases of radiative capture.

Moreover, the spectra of the fission and capture  $\gamma$ -rays must exceed a certain degree of complexity before the size of the effect detected can be related with certainty to particular cross-section values. In this case the variations in intensity of individual lines in the  $\gamma$ -spectra (in the transition from resonance to resonance) are averaged out by the large number of transitions, and the total  $\gamma$ -ray yield will not be subject to chance fluctuations. The spectra of the  $\gamma$ -rays of resonance radiative capture of a neutron by fissionable nuclei are unknown, but it may be assumed that, for heavy nuclei far from magic (with closed shells), they are close to evaporative (7). In this case the effect from the detection of radiative capture  $\gamma$ -rays will be proportional to the capture cross-section.

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The fission  $\gamma$ -ray spectrum has been obtained for the fission of  $U^{235}$  by al neutrons (6). It is practically uninterrupted; the average energy . the  $\gamma$ -quanta is about 1 MeV. The complexity of the spectrum makes any marked change in its shape on passing from resonance to resonance unlikely. There is every justification for assuming that the fission  $\gamma$ -ray spectra will be of a similar type for other fissionable nuclei as well.

If these ideas are correct, the size of the effect recorded by the  $\dot{\gamma}$ -detector may be written in the form (ignoring corrections for the scattering)

$$\mathbb{N}_{\gamma} \sim \frac{\sigma_{nf} + a\sigma_{n\gamma}}{\sigma_{t}} \cdot \mathbb{N} \cdot \sqrt{\mathbb{E}} \cdot (1 - T)$$

where  $\sigma_{nf}$ ,  $\sigma_{\gamma}$ ,  $\sigma_{t}$  are the cross-sections of fission and radiative capture and the total cross-section,

- N is the density of the resonance neutron flux,
- T is the transmission of the sample, and
- a is the ratio of the officiency of detection of cases of radiative capture to that of cases of fission.

Simultaneous measurements of the yield of secondary fission neutrons  $(N_f)$  make it possible to calculate  $\Gamma_f/\Gamma_a$ , while when the transmission of the sample is also known, other parameters of the resonance levels may also be obtained.

The authors have measured the yields of fission  $\gamma$ -rays and secondary fission neutrons from a sample of Pu<sup>239</sup> (thickness 0.001325 atoms/barn) in the range of neutron energies from 7 to 550 eV; for neutrons with energies up to 100 eV the transmission was also measured. The measurements were made on a neutron spectrometer with a "flickering beam" from an ITEF cyclotron (1,2). The resolution of the spectrometer was ~ 0.02  $\mu$ s/m.

Four crystals of NaI (T1) 8 cm in diameter and 8-10 cm high were used as  $\gamma$ -ray detectors. These were placed symmetrically on two sides of the sample of Pu<sup>239</sup>, inside a lead shielding up to 50 cm thick (Figs. 1 and 2). For protection from the scattered neutrons the crystals were covered with 1 cm of boron carbide.

To reduce the background, the multiplicr outputs were connected up in pairs for coincidence; to give maximum efficiency, every possible combination of the photo-multiplier outputs was used (six coincidence schemes).  $\gamma$ -quanta with energies of more than ~ 700 keV were detected; the resolving time of the coincidence schemes was ~ 0.1  $\mu$ s.

The fission neutron counter consisted of two TEU-45 photo-multipliers coated with a paraffin-zinc sulphide mixture (1), the pulses from the photo-multiplier outputs were added together.

Fig. 3 shows some of the experimental curves for  $N_f$  and  $N_\gamma$  (neutron energies 35-600 eV) after subtraction of the background. In spite of the use of double coincidences, the background level in the  $N_\gamma$  curves was fairly high acout 50% of the effect. The background remained constant up to neutron energies of ~ 50 eV, but gradually increased at higher energies (by approximately 30% at 200 eV). The background of the fission curve  $N_f$  was similar.

The curves were normalized according to the first resonances of plutonium  $(E_0 = 7.8 \text{ eV} \text{ and } 15.5 \text{ eV})$ . The value of  $\Gamma_f / \Gamma_a$  for these resonances was set, on the basis of the results of reference (3), at 0.465 and 0.9 respectively. It is interesting to note that the ratio of the efficiency of detection of cases of radiative capture to that of cases of fission was  $a = 0.7 \pm 0.1$ , in accordance with the observations already made.

Table I gives the values of  $\Gamma_f/\Gamma_a$  for resonances in the energy range up to 90 eV; in this range practically all the levels are resolved. In the first column are given the results obtained by comparison of the normalized curves  $N_f$  and  $N_y$ , and in the second the data of reference (3).

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 · <sup>12</sup> 0	Present work	Reference (3)	то	Present work	Reference (3)	
 7.84.	0.46 <u>+</u> 0 05 <sup>*</sup>	0.465	49.9	0.65 <u>+</u> 0.09	0.40 <u>+</u> 0.10	
10.95	0.67 <u>+</u> 0.06	.0.67 <u>+</u> 0.03	52.7	0.33 <u>+</u> 0.04	0.17 <u>+</u> 0.05	
12.00	0.42 <u>+</u> 0.05	0.37 <u>+</u> 0.03	57.8	0.87 <u>+</u> 0.09	0:80 <u>+</u> 0.15	
15.5	0.90 <u>+</u> 0.15 <sup>*</sup>	0.90 <u>+</u> 0.10	59.3	0.82 <u>+</u> 0.08	0.80 <u>+</u> 0.15	
17.8	0.43 <u>+</u> 0.05	0.42 <u>+</u> 0.03	62.1	0.82 <u>+</u> 0.2	0.9 <u>+</u> 0.1	
22.4	0,54 <u>+</u> 0.05	0.55 <u>+</u> 0.02	65.9	0.61 <u>+</u> 0.07	0.52 <u>+</u> 0.10	
26.3	0.59 <u>+</u> 0.12	0.,39 <u>+</u> 0.02	75.0	0.67 <u>+</u> 0.07	0.55 <u>+</u> 0.10	
32.5	0.43 <u>+</u> 0.2	<u></u>	81.5	0.70 <u>+</u> 0.12	0.90 <u>+</u> 0.10	
41.7	0.24 <u>+</u> 0.05	0.14 <u>+</u> 0.03	85.6	0.61 <u>+</u> 0.05	0.62 <u>+</u> 0.20	
44.6	0.11 <u>+</u> 0.03	0.09 <u>+</u> 0.05	90.3	0.29 <u>+</u> 0.06	0.20 <u>+</u> 0.10	
47.8	0.59 <u>+</u> 0.12	0.65 <u>+</u> 0.20			•	
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\* Normalized value.

Comparison of the results of the measurements shows that in the cases when it is possible to disregard neutron width, the  $\Gamma_f/\Gamma_a$  values obtained by the two methods agree (within the limits of measurement error, i.e. with up to 10-15% accuracy). This confirms the validity of the assumption on which the method is based, that there are comparatively small changes in the shape of the fission and capture  $\gamma$ -ray spectra on passing from one level to another. Constancy of the radiation width (within the indicated limits) - a circumstance noted previously (8) - also follows from this.

In the neutron energy range from 100 to 300 eV the positions of the strongest individual resonances have been determined, and the average values  $\overline{\sigma}_{\rm f}/\overline{\sigma}_{\rm a}$  obtained. Strong levels (or possibly groups of levels) occur at the following energies (eV): 95.7, 106.2, 118, 132, 143, 156, 163, 173, 185, 193, 200, 260, 270 (Fig. 3). The values of  $\overline{\sigma}_{\rm f}/\overline{\sigma}_{\rm a}$  are given in Table II.

B	7-40 e <sup>₩</sup>	40-106 e <sup>vr</sup>	106-210 e <sup>V</sup>	7-210 eV
σ <sub>f</sub> /σ <sub>a</sub>	0.57 <u>+</u> 0.05	0.63 <u>+</u> 0.07	0.57 <u>+</u> 0.10	0.59 <u>+</u> 0.04

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The error does not take into account the indeterminacy of the value of  $\Gamma_{\rm f}/\Gamma_{\rm c}$  taken for the 7.84-eV resonance = 0.465 (3).

Simultaneous measurement of the fission and absorption cross-sections, together with the data on transmission of the sample, make it possible to assess in certain cases the spin of the levels. For such a calculation,  $\Gamma_n$ must constitute a considerable part of the total resonance width. Table III gives the results of calculations for some levels.

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Ξo .	F <sub>a</sub> /F .Fresent work	2g <sup>r</sup> n	Г f	$\frac{\pi r}{I=0}$	I taken as 	
41.7	0.86 <u>+</u> 0.09		13	0.87 0.95	(0)?	
44.6	0.92 <u>+</u> 0.09	5.5 <u>+</u> 1.0	5.5	a.80 <u>0.93</u>	. (1)	
52.6	0.69 <u>+</u> 0.07	8.7 <u>+</u> 1.0	23	<u>0.79</u> 0.91	0	
	0.72 <u>+</u> 0.10	19.5 <u>+</u> 4.0	89	0.77 0.90	0	
75.0	0.97 <u>+</u> 0.10	25 <u>+</u> 5	120	0.76 <u>0.90</u>	I	

\* Average values for the present work and reference (3). \*\*  $\Gamma_{f} = \frac{40}{\Gamma_{f}\Gamma_{f} - 1} \frac{\text{MeV}}{\gamma}$  is taken as 40 MeV.

The spin values obtained can of course only be regarded as approximate. It is interesting to compare the assessed values with a consideration of interference effects at these energies. The energy dependence of the fission cross-section in the range 55-80 eV indicates the existence of a level of very great width (of the order of several electron volts) situated at  $E_0 = 62 \text{ eV}$ . The 65.9-eV resonance, on the edge of this level, is of symmetric form, and does not apparently interfere with the level. The next level  $(E_0 = 75 \text{ eV})$  is markedly asymmetrical. The asymmetry may be explained by assuming that this resonance interferes with the wide  $(E_0 = 62 \text{ eV})$  level.

The results of reference (9) show that interference between levels of  $Pu^{239}$  with the same spin value is almost maximal. We should consequently ascribe identical spin to the resonance with  $E_0 = 62$  eV and 75 eV, and opposite spin to the level at 65.9 eV. This conclusion agrees with the results of Table II; thus it is logical to take I = 0 for the resonance at 65.9 eV, and I = 1 for the levels at 62 eV and 75 eV.

The difference in the average fission widths of systems of levels of  $Pu^{239}$  with different spin values has already been noted (9, 10). This conclusion was determined to a considerable extent by the spin of one level ( $E_0 = 15.5 \text{ eV}$ ,  $\Gamma_{e} \approx 1 \text{ eV}$ , I = 1).

If we take into consideration another three resonances ( $E_0 = 65.9 \text{ eV}$ ,  $\Gamma_f \approx 60 \text{ MeV}$ , I = 0;  $E_0 = 75 \text{ eV}$ ,  $\Gamma_f \approx 75 \text{ HeV}$ , I = 1; and  $E_0 = 62 \text{ eV}$ ,  $\Gamma_f \approx 2 \text{ eV}$ , I = 1), we obtain values of  $\overline{\Gamma}_f \approx 350 \text{ MeV}$  and ~ 50 MeV for the systems of levels with I = 1 and 0 respectively.

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Abscissas:Neutron energy (eV)Ordinates: $N_f$  (arbitrary units) $N_\gamma$  (arbitrary units)

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- Fig. 1. Layout of experimental equipment 1 - cyclotron chamber 2 - B<sub>4</sub>C shielding 3 - paraffin retarder 4 - collimator 5 - sample
  - 6 detectors 7 B<sub>4</sub>C shielding 8 lead shielding
  - 9 cyclotron shielding 10 cyclotron magnet



Fig. 2. Arrangement of scintillation counters in relation to sample

Top left: Output cascade of photomultiplier

Middle left (on diagram): FEU-13