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MEASUREMENT OF THE CAPTURE-FISSION CROSS-SECTION RATIO (α) OF ^{239}Pu IN THE 0.007 eV - 12 keV NEUTRON ENERGY RANGE

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ABSTRACT

The author measured the radiative capture-fission cross-section ratio (α) for ^{239}Pu with a resolution $\frac{\Delta t}{L} = 0.23$ µsec/m.

The experimental method consisted in comparing the scintillation-detector count of gamma quanta and prompt fission neutrons from a sample with pulse separation based on scintillation decay time. Values are presented for $\alpha(E)$ of ^{239}Pu in the 0.007 eV-12 keV reacting neutron energy range.

Detailed information on the energy dependence of the radiative capture-fission cross-section ratio $\alpha(E) = \sigma_c(E)/\sigma_f(E)$ has been the subject of particular interest in recent years because of its implications for the choice of optimum lines of development in nuclear power engineering. In the neutron energy range which is of interest from the point of view of practical applications, important experimental results have been obtained by the time-of-flight method. However, these findings not only are at variance with the data of integral experiments but they also fail to agree satisfactorily with each other [1]. As a result it is still not possible to make a reliable selection of the nuclear data needed for reactor calculations. In the near-thermal range of neutron energies the results of differential and integral measurements of nuclear data by various methods are in agreement to a high degree of accuracy [2]. With the existing neutron spectrometers using the time-of-flight method it is difficult, by means of a single method, to make measurements of partial cross-sections and their relationships from the thermal energy range up to an energy of several tens of keV. For this reason it is difficult to check the applicability, effectiveness and reliability of a particular method of measurement, for example, of $\alpha(E)$ in the range of hundreds of eV, with simultaneous measurement in the near-thermal energy range.

The present study, which was made in 1972, represents an effort to obtain, in a single experiment and by a single method, values of $\alpha(E)$ in the neutron energy range from 0.007 eV to 12 keV, i.e. in a range which is important both for reactors based on thermal and intermediate neutrons and for fast reactors with a dilute core and correspondingly softened neutron spectrum.

The measurements

The measurements of $\alpha(E)$ for ^{239}Pu were made by the time-of-flight method at a flight distance $L = 251.6$ m with nominal resolution $(\sqrt{\Sigma \Delta t_i^2}/L) = 0.23$ µsec/m. In order to cover the largest possible energy range in one measurement the author used as his neutron source the JINR pulsed fast reactor operating at an infrequent pulse regime (1 pulse per 3.8 sec) and at a capacity of 10 kW. The experimental method consisted in comparing the count of gamma quanta and prompt fission neutrons from a sample of ^{239}Pu as a function of time-of-flight. The detector of fission neutrons and gamma quanta was a stilbene crystal (70 mm in diameter and in height) and a FEU-82 photomultiplier. The prompt fission neutrons (f channel) were recorded as the basis of scintillation flashes from recoil protons with gamma background discrimination based on scintillation decay time [3]. The energy threshold for the recording of neutrons was ~0.9 MeV; suppression of gamma quanta: ~3×10^4. The threshold for recording gamma quanta from fission and radiative capture (γ channel) was fixed at a level of ~0.5 MeV.

The detector was positioned outside a carefully collimated (up to a diameter of 30 mm) neutron beam and enclosed in a shielding of B_4C and Pb about 15 cm thick. A metallic sample of ^{239}Pu 28 mm in diameter and containing 1.75% ^{240}Pu was placed in the neutron beam at an angle of 45° directly opposite the detector window. The surface density of the sample was 2.13×10^{21} nuclei per cm^2. The entry window of the detector was shielded from scattered resonance neutrons of the beam by means of a ^{10}B filter 25 mm thick and from the natural gamma activity of the sample and the gamma quanta from the capture of scattered neutrons on the boron filter by means of lead 5 mm thick.

The use of a well-shielded scintillation detector of small volume in combination with an infrequently pulsed neutron source of large pulsing power enabled us to make measurements with a constant background which was extremely low for the method chosen (less than 1% of the count in the channel at an energy $E_n = 0.0253$ eV for the two spectra) and which was determined only by spontaneous fission in the sample. The background of fast neutrons and gamma rays from the pulsed reactor became considerable at energies above 100 eV and was determined by means of a set of "black" filters made of Cd, Ag, Co, Mn and Na [4]. The sensitivity of the detector to scattered neutrons was determined by using an equivalent sample of a lead scattering material. Subsequent measurement is particularly important in the energy region below 5 eV: this is the only means by which account can be taken experimentally of the contribution of satellites to the f- and γ-channels [5]. The background characteristics of the experiment are given in Table I.

Table I

Ratio of total count to background count

ΔE, keV / Recording channel	0.1–0.2	1–1.5	10–12
f–channel	125.0	26.3	7.1
γ–channel	19.2	5.0	1.8

Ultimately, information was collected in the memories of two 4096-channel analysers with channel widths of 16 μsec (the first thousand) and 64 μsec. Figure 1 presents recorded spectra obtained by using the measurement method described.

Results and discussion

The number of counts in the f- and γ-channels after elimination of background may be represented in the form

$$\begin{aligned} N_n &= \varepsilon_n n_f + \varepsilon_{cn} n_c , \\ N_\gamma &= \varepsilon_\gamma n_f + \varepsilon_c n_c , \end{aligned} \qquad (1)$$

where n_f and n_c are the number of fissions and captures, respectively, in the sample during 1 sec; ε_n, ε_{cn} are, respectively, the recording efficiencies for fission neutrons and radiative-capture gamma rays in the f-channel; ε_γ, ε_c are the recording efficiencies for gamma quanta from fission and capture, respectively, in the γ-channel. Making allowance for multiple scattering, we can obtain from expression (1) the following well-known expression for α(E) [6]:

$$\alpha(E) = \frac{A\frac{N_\gamma}{N_n} - 1}{B - C\frac{N_\gamma}{N_n}} \left\{ \frac{1 + \sigma_{nn} < \frac{(1 - T^1)}{\sigma_{T^1}} \cdot \frac{\sigma_f^1}{\sigma_f} > + \cdots}{1 + \sigma_{nn} < \frac{1 - T^1}{\sigma_{T^1}} \cdot \frac{\alpha^1}{\alpha} \cdot \frac{\sigma_f^1}{\sigma_f} > + \cdots} \right\} \qquad (2)$$

in which $\alpha = \sigma_c/\sigma_f$, $\alpha^1 = \sigma_c^1/\sigma_f^1$ and σ_T, σ_c, σ_f, σ_{nn} are the total cross-section, the cross-sections for radiative capture, fission and scattering of neutrons in the first interaction with target nuclei, T is the transmission of the sample, the super-scripts 1, 2 etc. denote the cross-sections after the scattering of neutrons in the sample one time, two times etc.; $A = \varepsilon_n/\varepsilon_\gamma$, $B = \varepsilon_c/\varepsilon_\gamma$ and $C = \varepsilon_{cn}/\varepsilon_\gamma$. The second multiplier in expression (2) is a correction for multiple neutron interaction in the sample: it is a function mainly of the ratio $\alpha^1(E)/\alpha(E)$ and so forth, i.e. of the actual value of α(E) and its variation with the energy of the interacting neutrons. The neutron can undergo a number of scatterings, losing in each of them approximately 1% of its energy before being captured by a nucleus and this will result in fission or in the emission of γ-quanta. However, the energy spread in scattering is 10 times less than the experimental energy resolution and this results in some degree of "smoothing" of the abrupt variations in $\alpha(E)$ and for the measurement accuracy achieved we may consider that $\alpha(E) \approx \alpha^1(E) = \alpha^2(E) = \ldots$.

Then, using expression (2) we can obtain $\alpha(E)$, determining the constants A, B and C by the method of least squares by normalizing to known values of α_o for low-energy resonances [4]. In the normalization we introduced into the experimental data corrections for the energy dependence of the mean number of prompt neutrons on the basis of the data of Trochon and Lukas [7], who used a similar measurement method. The values for the constants in expression (2) obtained by normalization were: $A = 0.26$, $B = 0.66$ and $C = 0.0054$. The calculated values of $\alpha(E)$ in the neutron energy range 100 eV-12 keV are compared in Fig. 2 with data from Refs [4, 6, 8, 9, 10], which were obtained by various methods with the same or higher energy resolutions. Better agreement is observed with the data of Sowerby et al. [6] which were obtained by a similar method of measurement. However, the results of the present study, like those of Kurov et al. [4], where a time-of-flight energy resolution was used, do not clearly confirm a pronounced structure in $\alpha(E)$ below an energy of 1 keV. The experimental errors of $\alpha(E)$ depend, as pointed out by Ryabov [1], on the method of normalization, i.e. on the errors in the determination of A, B and C, the accuracy in allowing for background and the amount of background. Counting statistics have practically no effect on the error, except for the inter-resonance region in the case of low neutron energies. The increase in $\alpha(E)$ errors in the energy range above 3 keV is due to the fact that the last energy point for determining background is at $E_n = 2.85$ keV (resonance of Na), and least-squares fitting of the background curve leads to greater uncertainties than in the energy region below 3 keV.

Figure 3 shows the fission cross-section $\sigma_f(E)$ obtained from normalizing the relative trend of $N_n(E)$ in the area of individual resonances with allowance for sample thickness and the relative trend of the neutron flux, as was done on the study of Kurov and Ryabov [4]. Figure 3 also includes data on $\sigma_f(E)$ from Refs [4, 6, 8, 11 and 12], obtained in measurements made with ionization chambers and detectors of prompt fission neutrons. The cross-section $\sigma_f(E)$ obtained in the present study is in good agreement with the measurements reported in Ref. [4], which were made with a fission ionization chamber and with the same time-of-flight energy resolution. Satisfactory agreement is also observed in the entire interacting neutron energy region investigated and with other data averaged over the 0.1 and 1 keV energy intervals.

In Fig. 4, for the first time, detailed and very complete data on $\alpha(E)$ in the 0.007–5 eV energy range are presented; in the 1.4–5 eV region the experimental count was summed in 10–20 channels in order to eliminate as far as possible the possibility of insufficient counting statistics in this region affecting the results. Table II gives the average values of $\langle\alpha(E)\rangle$ in individual energy intervals from 0.007 to 100 eV, where the energy resolution still permits unlimited use of the averaging procedure. The results obtained are for the most part in agreement – within the limits of error – with the data on this energy range which are known at the present time [8, 13].

Table II

Average values of $\langle\alpha(E)\rangle$ in the 0.007–100 eV energy range

Δ E, eV	<α(E)>		Δ E, eV	<α (E)>	
	This study	Ref. [8]		This study	Refs [8] and [13]
0,007-0,008	0,332±0,036		0,5-0,6	0,446±0,055	0,46±0,05
0,008-0,009	0,335±0,036		0,6-0,7	0,394±0,048	0,38±0,081
0,009-0,010	0,338±0,036		0,7-0,8	0,363±0,046	0,31±0,11
0,010-0,015	0,346±0,038		0,8-0,9	0,348±0,046	0,31±0,14
0,015-0,020	0,350±0,039		1,5-2,0	0,218±0,096	
0,02 -0,03	0,357±0,041	0,37±0,03	2,0-3,0	0,221±0,107	
0,03 -0,04	0,368±0,042	0,38±0,03	3,0-4,0	0,231±0,085	
0,04 -0,05	0,386±0,044	0,40±0,03	4,0-5,0	0,233±0,078	
0,05 -0,06	0,414±0,044	0,42±0,03	5,0-10,0	0,482±0,067	0,64±0,07
0,06 -0,07	0,439±0,045	0,44±0,03	10,0-20,0	0,579±0,069	0,52±0,06
0,07 -0,08	0,451±0,046	0,46±0,03	20,0-30,0	0,787±0,089	0,61±0,07
0,08 -0,09	0,465±0,046	0,48±0,03	30,0-40,0	0,840±0,093	0,40±0,04
0,09 -0,10	0,485±0,049	0,50±0,03	40,0-50,0	3,261±0,192	1,40±0,15
0,10 -0,15	0,538±0,054	0,61±0,04	50,0-60,0	1,082±0,087	0,74±0,08
0,15 -0,20	0,636±0,059		60,0-70,0	0,398±0,058	0,47±0,06
0,2 -0,3	0,675±0,065	0,69±0,04	70,0-80,0	0,412±0,061	0,47±0,09
0,3 -0,4	0,637±0,058	0,66±0,04	80,0-90,0	0,237±0,047	0,23±0,03
0,4 -0,5	0,534±0,056	0,56±0,03	90,0-100,0	0,682±0,076	1,16±0,12

In Table III, for purposes of comparison, we give α_0 for low-energy resonances, which are calculated from expression (2), with the above-mentioned values of A, B and C and which are in good agreement with known values [4, 6, 13].

Table III

Experimental values of α_o for resonances of ^{239}Pu, calculated from expression (2)

$E_{res.}$, eV	α_o	$E_{res.}$, eV	α_o
0,0253	0,363 ± 0,040	11,93	1,49 ± 0,09
0,3	0,666 ± 0,059	17,7	1,03 ± 0,08
7,83	0,79 ± 0,06	22,3	0,63 ± 0,06
10,97	0,37 ± 0,04	26,2	0,90 ± 0,07

In conclusion, it should be pointed out that the data on $\alpha(E)$ were not corrected for radiative capture in the ^{240}Pu admixture since the large, systematic errors (~10–20%) characteristic of the method of measurement and normalization used considerably exceed the magnitude of such correction.

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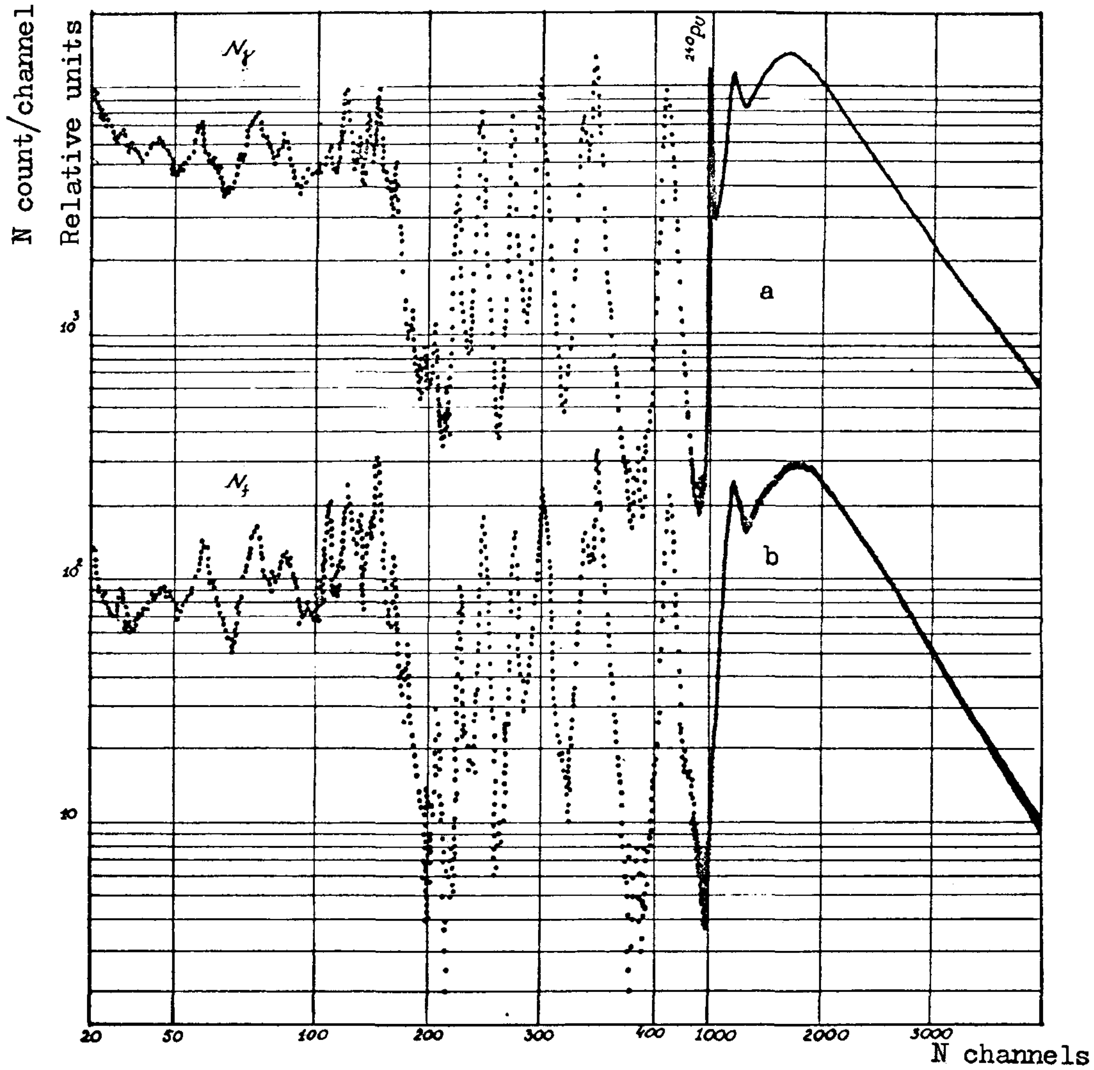

Fig. 1. Experimental time-of-flight spectra:

(a) γ-channel;

(b) f-channel (horizontal scale not uniform).

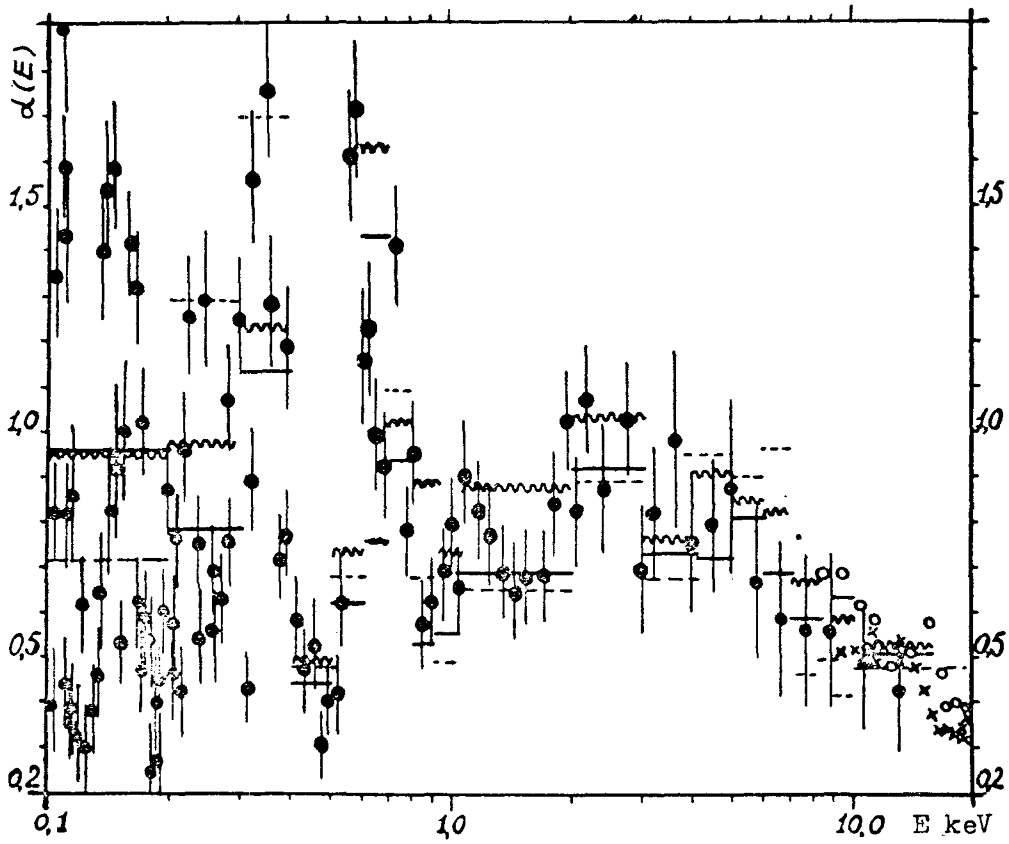

Fig. 2. Results of measurements of α(E) in the 0.1-20 keV energy range.

⧫ = the present study;

----, ——, ∿∿∿ = data from Refs [4, 6, 8] respectively;

x, o = data obtained from electrostatic generators [9, 10].

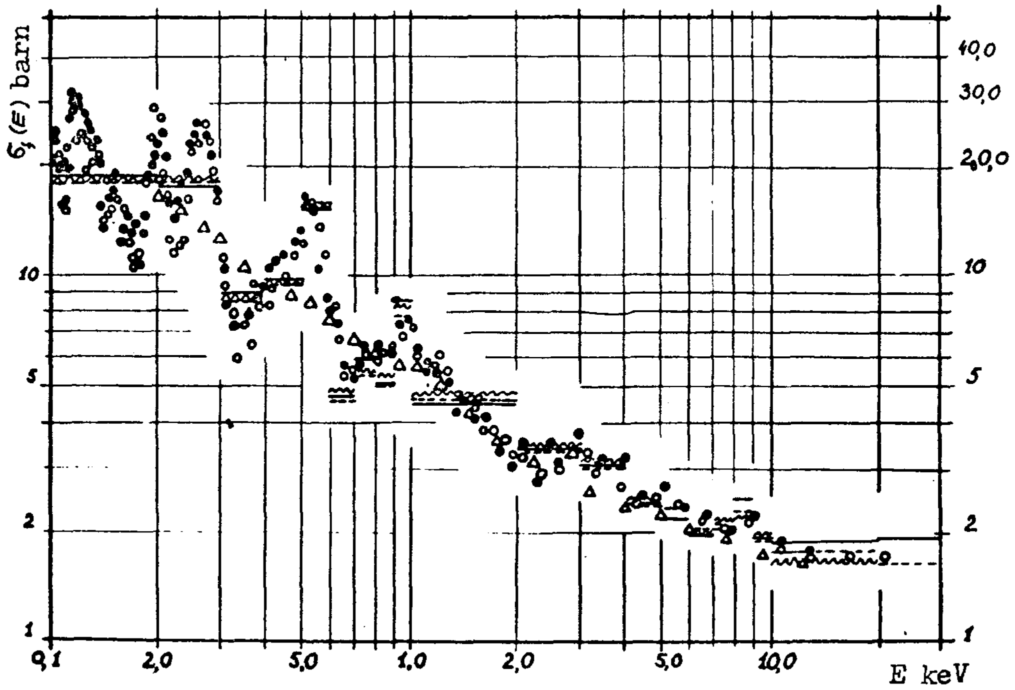

Fig. 3. Results of measurements of $\sigma_f(E)$ in the 0.1-20 keV energy range.

● = the present study;

o, ∿∿∿, ----, ——, Δ = data from Refs [4, 6, 8, 11, 12] respectively.

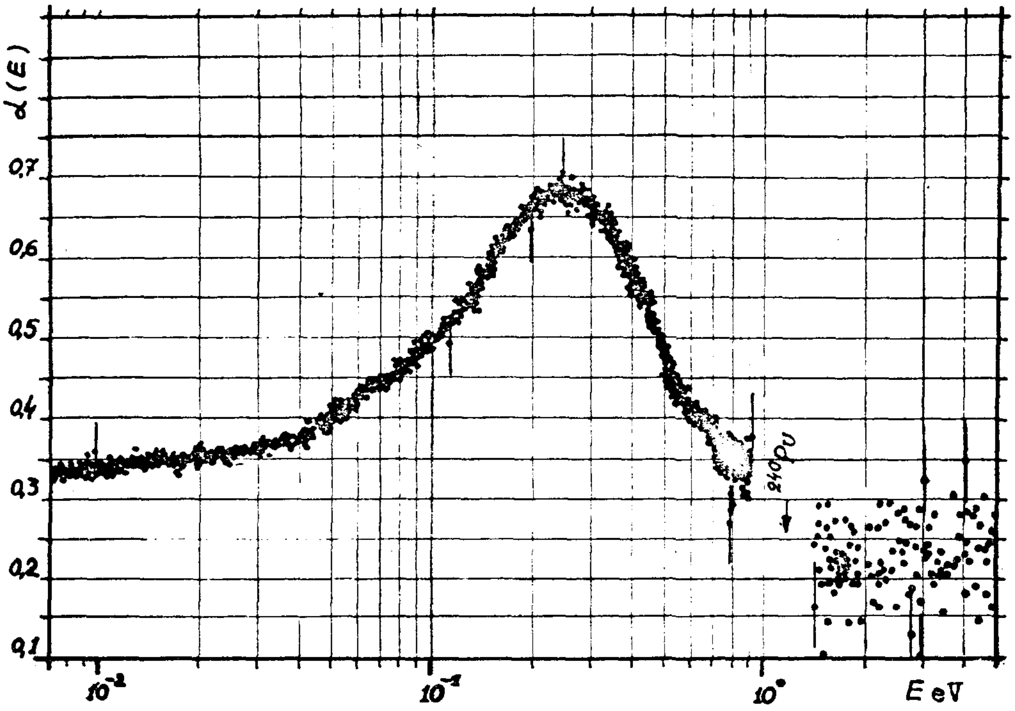

Fig. 4. Results of measurements in the 0.007-5 eV energy range. (The arrow denotes the position of the ^{240}Pu + n resonance.)