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MEASUREMENT OF $\alpha(E_n) = \sigma_c(E_n)/\sigma_f(E_n)$

IN THE RESONANCE ENERGY REGION OF INTERACTING NEUTRONS

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ABSTRACT

The proposed improvement in the technique for measuring $\alpha(E_n)$ with the help of low-efficiency γ-ray and prompt fission neutron detectors makes it possible to obtain more accurate and reliable results. The possibilities of the method have been illustrated by time-of-flight measurements of $\alpha(E_n)$ for ^{235}U made at CEN Saclay in France.

Experimental determination of the ratio of the radioactive capture cross-section to the fission cross-section, $\alpha(E) = \sigma_c(E_n)/\sigma_f(E_n)$, for trans-uranium isotopes over a large neutron energy range is important both for reactor design and for nuclear physics. The methods of relative measurement of $\alpha(E_n)$ used during the last few years [1-8] have so far not delivered the requisite accuracy of 3-5% [9] owing to the large uncertainties associated with (i) separation of radiative capture γ-rays from the total γ-spectrum, (ii) the possible dependence of the capture and fission γ-ray spectra on the quantum properties of the compound nucleus and on the energy of the interacting neutrons, (iii) the need to determine a minimum of two normalizing "constants" in calibrating $\alpha(E_n)$, and so forth [10]. These defects are found to a very great degree in the most commonly used method of measuring $\alpha(E_n)$, which uses low-efficiency detectors to count prompt fission neutrons and capture and fission γ-rays. The present paper suggests a way of improving the method and shows that a new experimental approach will make it possible to avoid many of the uncertainties and to enhance the accuracy and reliability of the results.

Measurement method

At present two substantially different methods of measuring $\alpha(E_n)$ are being used. In the first [3, 5] γ-rays are recorded with high efficiency by a large liquid scintillation detector, an arrangement which makes the method comparatively insensitive to variations in the γ-spectrum and to possible changes in the average number of prompt neutrons per fission. However, the overall background of such a detector is high and can lead to large uncertainties which reduce the accuracy of the measurements. Furthermore, equipment of this kind is expensive and complex in use.

In the second method [1, 7], which we propose to go into more thoroughly, "low-efficiency" scintillation counters are used to record radiative capture and fission γ-rays (γ-channel) and prompt fission neutrons (fission channel). The count rate of the prompt neutron detector in the thin specimen approximation ($n\sigma_o \ll 1$) is then

$$n_f = N_f \{\sum_\nu [1-(1-\varepsilon_{nf})^\nu] P(\nu)\} + N_c \int_{E_a}^{E_b} \varepsilon_\gamma^{cf}(E) \cdot \nu_\gamma^c(E)\, dE \ , \qquad (1)$$

where: N_f is the number of fissions taking place in the sample in 1 second;

N_c is the number of capture events in the sample in 1 second;

ν is the number of prompt neutrons emitted per fission event;

$P(\nu)$ is the probability of ν neutrons being emitted in a given fission event;

ε_{nf} is the efficiency with which the neutron counter records a prompt fission spectrum neutron;

$\nu_\gamma^c(E)dE$ is the number of radiative capture γ-rays with energies between E and E + dE;

$\varepsilon_\gamma^{cf}(E)$ is the efficiency with which the neutron counter records γ-rays with energies between E and E + dE; and

E_a, E_b is the energy range in which the neutron detector records γ-rays.

In the γ-channel, with low efficiency of γ-ray recording and negligibly small sensitivity to prompt neutrons, the count rate in the energy interval from E_1 to E_2 is

$$n_\gamma = n_\gamma^c + n_\gamma^f = N_c \int_{E_1}^{E_2} \varepsilon_\gamma^c(E) \cdot \nu_\gamma^c(E) dE + N_f \int_{E_1}^{E_2} \varepsilon_\gamma^f(E) \cdot \nu_\gamma^f(E) dE, \tag{2}$$

where $\nu_\gamma^c(E)dE$ and $\nu_\gamma^f(E)dE$ are respectively the number of capture γ-quanta and the number of fission γ-quanta with energies between E and E + dE, and $\varepsilon_\gamma^c(E)$ and $\varepsilon_\gamma^f(E)$ are the efficiencies with which these γ-quanta are recorded by the γ-detector. The remaining notation is the same as in Eq. (1).

From Eqs (1) and (2) we can derive the customary expression for $\alpha(E_n)$ [1, 4]:

$$\alpha(E_n) = \frac{A\left[\dfrac{n_\gamma(E_n)}{n_f(E_n)}\right] - 1}{B - \left[\dfrac{n_\gamma(E_n)}{n_f(E_n)}\right] C}, \tag{3}$$

where

$$A = \frac{\sum\limits_{\nu}[1-(1-\varepsilon_{nf})^{\nu}]P(\nu)}{\int\limits_{E_1}^{E_2}\varepsilon_{\gamma}^{f}(E)\cdot\nu_{\gamma}^{f}(E)\,dE} \qquad B = \frac{\int\limits_{E_1}^{E_2}\varepsilon_{\gamma}^{c}(E)\,\nu_{\gamma}^{c}(E)\,dE}{\int\limits_{E_1}^{E_2}\varepsilon_{\gamma}^{f}(E)\,\nu_{\gamma}^{f}(E)\,dE}$$

$$C = \frac{\int\limits_{E_a}^{E_b}\varepsilon_{\gamma}^{cf}(E)\,\nu_{\gamma}^{c}(E)\,dE}{\int\limits_{E_1}^{E_2}\varepsilon_{\gamma}^{f}(E)\,\nu_{\gamma}^{f}(E)\,dE}$$

The quantities A, B and C are considered "constants" and are determined from normalization of the experimental data to known values of α_0 for a few well isolated resonances or for a particular energy region [1, 4, 6]. In doing this one assumes that the spectrum and multiplicity of capture and fission γ-radiation are uncorrelated, independent of the neutron resonance characteristics in the energy interval from E_1 to E_2, and, in general, not dependent on the energy of the interacting neutrons.

Similar assumptions are made with respect to the characteristics of the neutron emission accompanying fission. Furthermore, it is assumed that the number of counts from fission γ-rays in the γ-detector is strictly proportional to the number of counts from fission neutrons in the neutron detector for each narrow energy interval over the whole range of interacting neutrons, and that each of these individually is strictly proportional to the number of counts from fission fragments, i.e. to the fission cross-section. This enables us, as has been demonstrated experimentally, to use a convenient procedure in allowing for the contribution of fission γ-rays: to a substantial extent (up to 80-90%) [10] the number of counts from fission γ-rays, whose dependence on the energy of the interacting neutrons is not measured experimentally, can be replaced by the number of counts from neutrons or fission fragments, as follows from expression (3). In the first differential measurements of $\alpha(E_n)$ for ^{235}U [3, 5] satisfactory agreement of the results was observed at the accuracy of 20-30% indicated by the authors. However, measurements of $\alpha(E_n)$ for ^{239}Pu with the same accuracy showed large discrepancies in certain energy regions which could not be explained merely by possible systematic errors associated with the experimental conditions [10].

In the detailed investigations of recent years carried out by numerous authors with a view to increasing the accuracy of $\alpha(E_n)$ measurements, attention has been concentrated mainly on improving the experimental technique, and the basic concept underlying the measurements has remained unchanged. The results obtained revealed even larger discrepancies which were not due to systematic errors, both for ^{239}Pu and (to a lesser extent) for ^{235}U. In the situation that arose it was difficult to compare the data of different authors without knowing the details of their experiments, because, owing to the non-linear relationship between $\alpha(E_n)$ and the measured quantities $(n_\gamma(E_n)/n_f(E_n))$, it was impossible to normalize the known data by a uniform method for purposes of comparison, as is usually done in the case of effective cross-sections [11]. For the same reason it was impossible to take account in a uniform manner of the uncertainties associated with A, B and C. All this limits the possibilities of an objective evaluation of known experimental data and of obtaining a recommended set of $\alpha(E_n)$ values with a realistic accuracy [12, 13]. In these circumstances, further improvement of the measurement accuracy without clarification of certain physical processes basic to the experimental methods used made little sense. As far back as 1966 it was noted [14] that the values of resonance integrals and average fission cross-sections calculated with high accuracy depend on whether they are derived from measurements in which fission fragments or the radiations accompanying fission are recorded. Moreover, even measurements carried out by both methods on the same time-of-flight spectrometer showed discrepancies in certain energy regions which were several times greater than the measurement errors [14]. It emerged, too, that when fission cross-sections were measured on the basis of secondary neutrons (or γ-rays) by methods sensitive to the multiplicity of the secondary radiation, widely differing results were obtained [15, 16]. Substantial fluctuations (up to 14%) were detected in $\bar{\nu}$, $\bar{E}_{\gamma\ total}$ and $\bar{\nu}_\gamma$ for some fission resonances of ^{239}Pu [17].

It is natural to suppose that these effects which influence the accuracy of $\sigma_f(E_n)$ and $\alpha(E_n)$ measurements are governed by the individual properties of the neutron resonances, primarily by total momentum, its projection on the axis of symmetry of the nucleus, the corresponding characteristics of the fission barriers and the sub-barrier fluctuations in the fission cross-sections. Therefore, even at the measurement accuracies that have been achieved for $\alpha(E_n)$ there would appear to be no justification for assuming A, B and C in expression (3) to be rigorously constant; and accordingly, if there is to be any further improvement in accuracy, it will be necessary either to develop methods which are largely insensitive to changes in A, B and C or to work out new methods which allow, experimentally, for possible changes in these quantities.

One of the possible approaches is to perfect the measurement methodology described in detail above. Thus expression (3) can, with an accuracy to within terms of the second order in C ($C \lesssim 10^{-2}$), be rewritten as follows:

$$\alpha(E_n) = [\int_{E_1}^{E_2} \varepsilon_\gamma^c(E)\nu_\gamma^c(E)dE]^{-1}\{\frac{n_\gamma(E_n)}{n_f(E_n)}[\sum_\nu [1-(1-\varepsilon_{nf})^\nu]P(\nu) - k\int_{E_a}^{E_b} \varepsilon_\gamma^{cf}(E)\nu_\gamma^c(E)dE] - \frac{n_\gamma^f(E_n)}{n_f(E_n)}\} \qquad (4)$$

where $n_\gamma^f(E_n) = N_f \int_{E_1}^{E_2} \varepsilon_\gamma^f(E)\nu_\gamma^f(E)dE[\sum_\nu \{1-(1-\varepsilon_{nf})^\nu\}P(\nu)]$ is the count rate for γ-quanta in coincidences with fission events in the same energy recording ranges as the total γ-spectrum, $k \approx 1$. From expression (4) it is quite apparent that by now our only source of uncertainty is the quantity $\int_{E_1}^{E_2} \varepsilon_\gamma^c(E)\nu_\gamma^c(E)dE = f(E_n)$, on which $\alpha(E_n)$ depends linearly. This expression is also useful for comparing different sets of data, provided the requisite renormalization is carried out. Then, using the notation of correlated efficiencies, we can write expression (4) in a form convenient for the processing of experimental data:

$$\langle\alpha(E_n)\rangle = \frac{1}{\varepsilon_c}\{\frac{\langle n_\gamma(E_n)\rangle}{\langle n_f(E_n)\rangle}[\overline{\varepsilon_{nf}\nu_o}\langle R(E_n)\rangle - \bar{\varepsilon}_{cf}k] - \frac{\langle n_\gamma^f(E_n)\rangle}{\langle n_f(E_n)\rangle}\}, \qquad (5)$$

where $\langle R(E_n)\rangle = \langle\frac{\bar{\nu}(E_n)}{\bar{\nu}_o}\rangle$ can likewise be measured in the same experiment by the method of Trochon and Lucas [18], the brackets < > signify averaging over the selected energy interval, and $\bar{\nu}_o$ is the value (for an individual resonance or energy region) selected for normalizing the quantity $\overline{\varepsilon_{nf}\nu_o}\langle R(E_n)\rangle$.

Thus all the quantities figuring in expression (5) except $\bar{\varepsilon}_c$ and $\bar{\varepsilon}_{cf}$ can be measured simultaneously in the same experimental conditions. Since $\overline{\varepsilon_{nf}\nu_o} < R(E_n) > \approx 100\,k\,\bar{\varepsilon}_{cf}$, the absence of data on the energy dependence of $\bar{\varepsilon}_{cf}$ cannot have an effect on $< \alpha(E_n) >$, comparable to the uncertainty introduced by $\bar{\varepsilon}_c$. Even though at the present time there are no experimental techniques which enable us to determine the dependence of $\bar{\varepsilon}_c$ on E_n, the method of measuring $\alpha(E_n)$ proposed here does make it possible to find the energy dependence of the quantity $< \alpha(E_n) > \overline{\varepsilon_c(E_n)}$ as distinct from the usual relationship $\alpha(E_n)[< \alpha(E_n) > + b(E_n)$. It is quite obvious, then, that in equivalent experimental conditions the accuracy of the proposed method will be considerably greater than that of the methods used earlier.

<u>Experimental checking of the method of measuring $\alpha(E_n)$</u>

The proposed method has been tested experimentally; for this purpose the measurement results of Trochon and Ryabov [19] were used. The experimental data were obtained by the time-of-flight method, the 65-MeV linear electron accelerator of the Nuclear Research Centre at Saclay (France) being used as a pulsed neutron source for the purpose. The flight path was 12.5 m, giving a nominal resolution of 5.6 nsec/m for a neutron pulse and analyser channel width of 50 nsec. A "shadow" shield of lead was used to reduce the influence of γ-ray pulses from the neutron source. The neutron beam was covered by ^{10}B, Mn and Co filters to exclude recycle neutrons and control the background.

Fission events were recorded by two liquid scintillation counters (NE-213) with pulse-form gamma discrimination ($\bar{\varepsilon}_{cf} = 1 \times 10^{-3}$) [18]. To record γ-rays from fission and radiative capture, two NaI crystals were used, connected to a coincidence circuit with a recording amplitude window equal to (0.55-2.0) MeV. Gamma radiation in coincidences with fission events was recorded simultaneously.

The metallic ^{235}U sample (93.36%) and all four detectors were arranged in a plane perpendicular to the direction of the neutron beam. The detectors were covered with 5 mm of lead to protect them against natural radioactivity and with amorphous ^{10}B filters to shield them from scattered neutrons. The parameters of the background curve were determined from the background points of the resonance filters at E_n = 132 eV (Co); 0.337 and 2.38 keV (Mn) and the procedure used in allowing for the background was the same as in Refs [3] and [4]. The background conditions are shown in Table 1 below.

TABLE 1

Signal to Background Ratio

Measurement	Energy Interval, keV		
	0.2–0.3	1–2	8–9
Channel n_f	8.4	6.3	5.4
Channel n_γ	5.1	3.6	3.0
Channel n_γ^f	28.2	26.8	21.4

The duration of the time-of-flight spectrum measurements was 1 hour for $n_\gamma(E_n)$ and 3 hours for $n_f(E_n)$ and $n_\gamma^f(E_n)$. The experimental spectra can be seen in Fig. 1 below. Since no measurements of $R(E_n)$ were made in this work, a correction based on the results of Ref. [22] was made in $\overline{\varepsilon_{nf}\nu_o}$ to obtain $\bar{\varepsilon}_c$ in normalizing to α_o the resonances at E_o = 3.14, 4.84, 6.4, 7.09, 11.66, 15.4, 16.66, 26.5 and 33.58 eV [3, 15, 20, 21], and in the energy range investigated this correction was taken to be constant. Experimental allowance for the contribution of fission γ-quanta to the total spectrum indicates that, since the ratio $n_\gamma^f(E_n)/n_f(E_n)$ is not constant and its fluctuations can amount to 15–20%, there must be significant changes in $\alpha(E_n)$ in the energy ranges 0.1–0.2, 0.6–0.7, 3–4 and 5–6 keV. Another point to be noted is that as the energy of the interacting neutrons increases a small systematic decrease in the ratio $n_\gamma^f(E_n)/n_f(E_n)$ is observed. The values of $< \alpha(E_n) >$ are given in Table 2.

TABLE 2

Mean Values $<\alpha(E_n)> = \frac{<\sigma_c(E_n)>}{<\sigma_f(E_n)>}$

Energy, keV	$<\alpha(E_n)>$	Errors Statistical	Systematic	Total
0.05 -0.06	0.47	0.008	0.046	0.05
0.06 -0.07	0.74	0.020	0.061	0.06
0.07 -0.08	0.50	0.012	0.044	0.05
0.08 -0.09	0.49	0.012	0.043	0.04
0.09 -0.1	0.80	0.019	0.066	0.07
0.1 -0.2	0.55			
0.2 -0.3	0.58	0.008	0.045	0.05
0.3 -0.4	0.54			
0.4 -0.5	0.36	0.009	0.032	0.03
0.5 -0.6	0.41	0.008	0.037	0.04
0.6 -0.7	0.60	0.014	0.051	0.05
0.7 -0.8	0.41	0.010	0.037	0.04
0.8 -0.9	0.45	0.014	0.039	0.04
0.9 -1.0	0.58	0.018	0.047	0.05
1.0 -2.0	0.36	0.004	0.027	0.03
2.0 -3.0	0.41			
3.0 -3.987	0.52	0.011	0.052	0.05
3.987-4.982	0.37	0.011	0.045	0.05
4.982-5.981	0.45	0.012	0.053	0.05
5.981-6.989	0.34	0.011	0.046	0.05
6.989-8.036	0.40	0.012	0.057	0.06
8.036-9.74	0.47	0.017	0.056	0.06

Figure 2 combines the results of Refs [3-8, 23, 24] for purposes of comparison. In the data obtained by the method proposed here it will be seen that there are significant fluctuations in the energy region from 0.1 to 1 keV. In the range 1-10 keV the values of $< \alpha(E_n) >$ are higher than the data from Refs [3-8]. The values of $< \alpha(E_n) >$ averaged over wider energy intervals, $< \alpha(E_n = 0.1\text{-}1\ \text{keV}) > = 0.54 \pm 0.05$, $< \alpha(E_n = 1\text{-}9.7\ \text{keV}) > = 0.43 \pm 0.05$ and $< \alpha(E_n = 0.1\text{-}9.75\ \text{keV}) > = 0.48 \pm 0.05$, are also somewhat higher than the results of the other papers.

Full-scale use of the proposed method of measuring $\alpha(E_n)$ for the transuranics thus seems likely to yield considerably more reliable experimental data. It is particularly important to use a method such as this for ^{239}Pu, an isotope for which there have heretofore been large discrepancies among the measured values of $\alpha(E_n)$. The method can reduce to a minimum the systematic errors associated in particular with the recently observed substantial contribution of the (n, γf)-process to $\sigma_f(E_n)$ and the anti-correlation between total energy (and multiplicity) of prompt γ-rays and the average number of prompt fission neutrons [17], as well as with periodic fluctuations in $\sigma_f(E_n)$ due to the structure of the fission barrier [25].

In conclusion I want to express profound gratitude to Professors R. Joli and A. Michaudon for the facilities placed at my disposal for these experiments, to J. Trochon and B. Lucas for assistance in making the measurements, and to V.N. Kononov and E.A. Poletaev for discussing the results with me and offering critical comments.

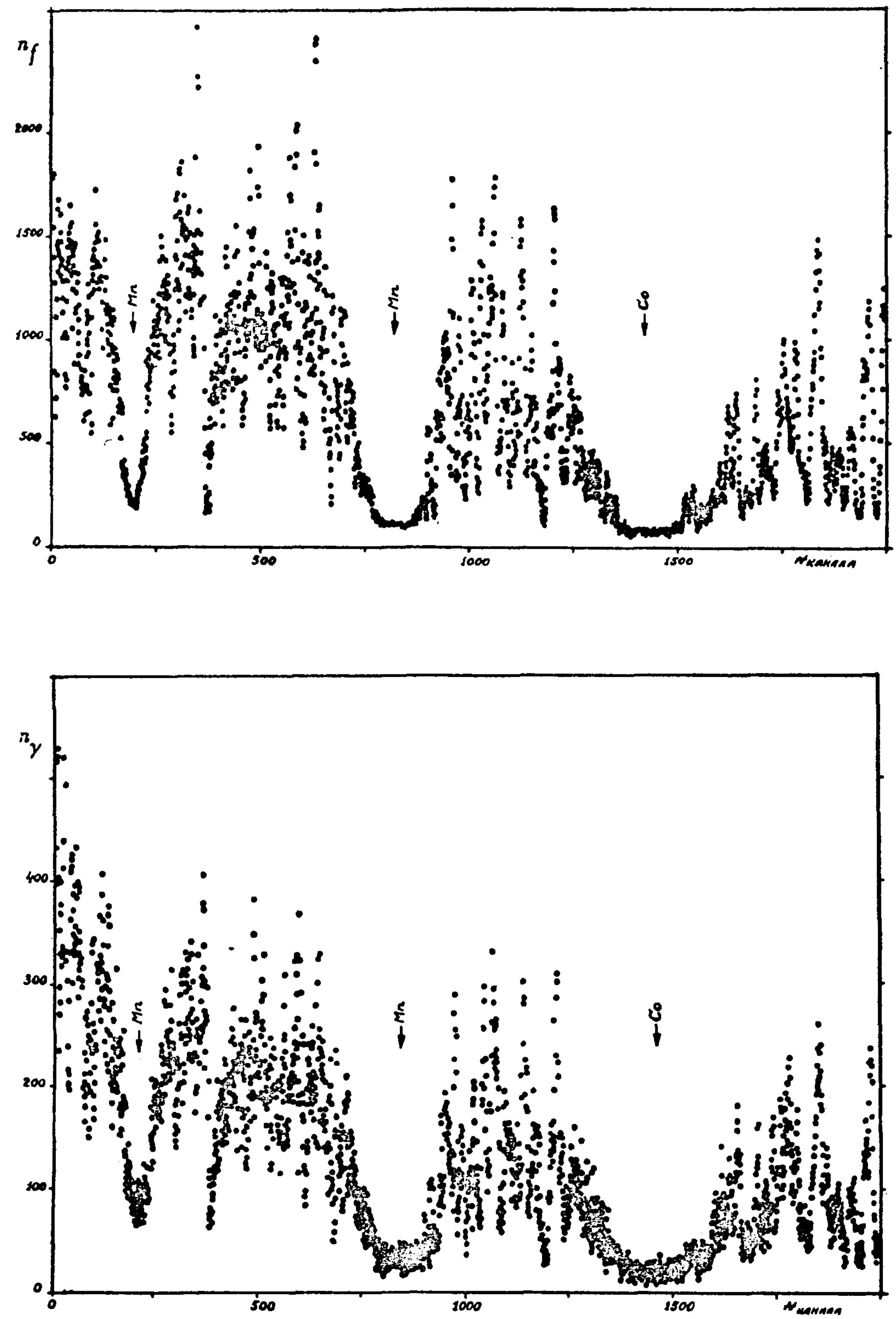

Fig. 1. Experimental time-of-flight spectra.
(a) n_f, (b) n_γ.

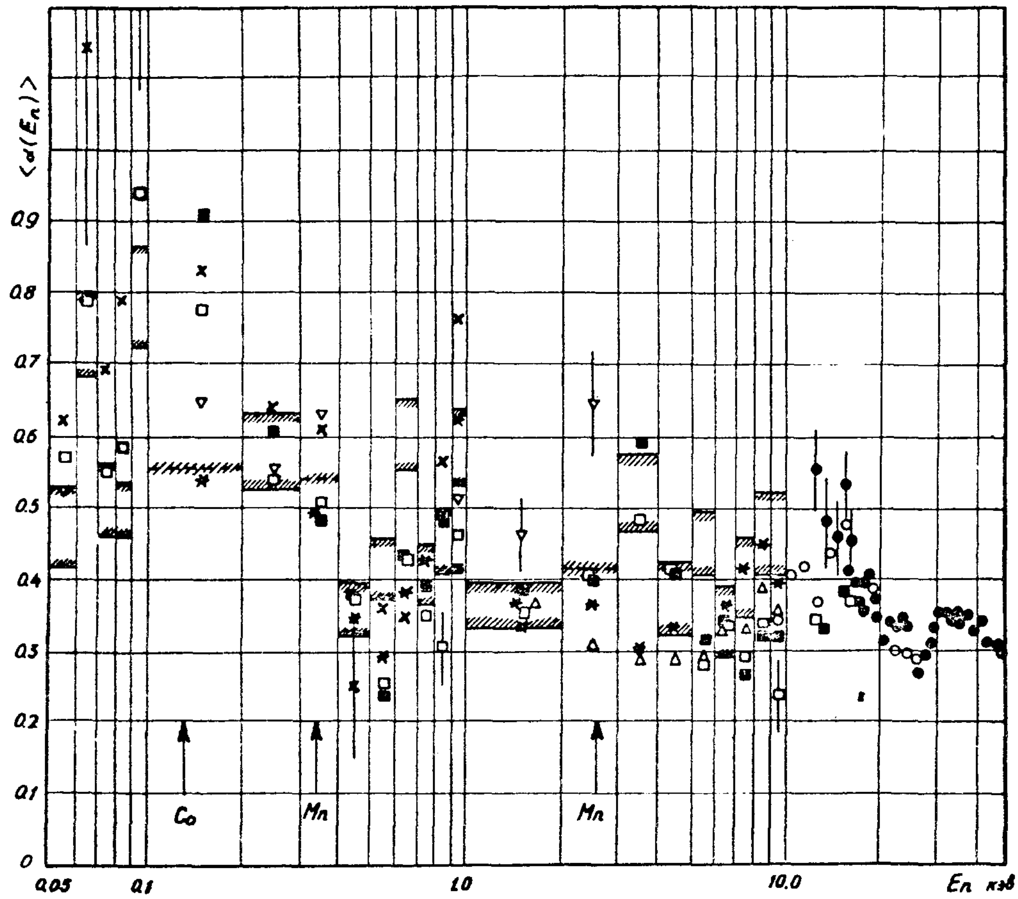

Fig. 2. Comparison of experimental data on < $\alpha(E_n)$ > for ^{235}U:

(hatched bars) – the present work;

■, □ – Joint Institute of Nuclear Research, 1965, 1968 [3, 4];

x, * – Oak Ridge National Laboratory, 1965, 1973 [5, 6];

△ – LRL 1970 [7];

▽ – Institute of Atomic Energy, 1970 [8];

● – Physico-Energetics Institute [23];

o – KFK [24].

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