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EVALUATION OF THE  $(n, 2n)$  AND  $(n, 3n)$  CROSS-SECTIONS

FOR HEAVY NUCLEI WITH ALLOWANCE FOR NON-EQUILIBRIUM PROCESSES

V.M. Bychkov, V.I. Plyaskin, Eh.F. Toshinskaya

Translation from Nuclear Constants 3(42) 26 (1981)

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Problems related to the reprocessing of nuclear fuel for the external fuel cycle and to a number of other aspects of nuclear power require for their solution a knowledge of the cross-sections for the (n,2n) and (n,3n) reactions in fissile isotopes. The experimental study of these reactions is a very complex task and as a result there is insufficient information available for evaluating the excitation functions of these reactions in the case of practically all fissile nuclei (with the exception of  $^{238}\text{U}$  and  $^{232}\text{Th}$ ).

The main difficulty in the theoretical prediction of the cross-sections of interest lies in making correct allowance for fission competition. In the majority of papers this difficulty is overcome by adjustment of the calculated values to experimental data through variation of the parameters in the theoretical models used. This probably explains why the predictions made by different methods [1-6] agree in describing the experimental data for the  $^{238}\text{U}$  nucleus but show wide differences for other nuclei where experimental information is not available.

Another important condition for getting a proper description of fissile nuclei cross-sections is that allowance should be made for neutron emission resulting from direct and pre-equilibrium processes. This question has been discussed in detail by Kornilov and co-workers [7] in an analysis of the spectra for inelastic scattering of neutrons by the  $^{238}\text{U}$  nucleus.

In the present paper, we use a theoretical method [8] for evaluating the (n,2n) and (n,3n) excitation functions, based on simplified versions of the statistical and exciton models. With this method it is quite simple to calculate absolute cross-section values for nuclei with relative atomic masses  $A \geq 100$  without adjustment to experimental data for the (n,2n) and (n,3n) reactions. The method is used to evaluate the excitation functions of the given reactions in 20 fissile isotopes. Comparison is made with the results of other evaluations and it is shown to be important to allow for non-equilibrium processes in the neutron channel.

Derivation of equations and description of the method. As Bychkov and co-workers [9] have shown, the neutron emission spectra and the (n,2n) cross-sections for non-fissile nuclei can quite accurately be described in terms of the statistical theory of nuclear reactions and the exciton model of pre-equilibrium decay. This can be done for a wide range of nuclear masses and incident neutron energies with a single set of model parameters.

For calculating the cross-sections of the (n,2n) and (n,3n) reactions in heavy nuclei ( $A \gtrsim 100$ ), in which charged-particle emission can be neglected, the relationships used in Ref. [9] can be greatly simplified. As before, we shall distinguish between equilibrium and non-equilibrium nuclear reaction mechanisms. As non-equilibrium processes we shall take all interactions which do not lead to an equilibrium stage of the compound nucleus. The total contribution of these processes can be described with sufficient accuracy by the exciton model of pre-equilibrium decay [10]. Assuming that the compound and non-equilibrium processes are well separated in time (and therefore independent), we can write the nuclear reaction cross-section as the sum of the equilibrium and pre-equilibrium components:

$$\sigma_{n,2n}(E_n) = \sigma_{n,2n}^{eq}(E_n) + \sigma_{n,2n}^{pre}(E_n), \quad (1)$$

where  $E_n$  is the incident neutron energy and  $\sigma_{n,2n}^{eq,pre}$  are the equilibrium and pre-equilibrium components of the (n,2n) cross-section.

The pre-equilibrium component of the (n,2n) reaction is taken to be the probability of occurrence of a process in which the inelastic scattering of the neutron incident on the target nucleus occurs as a result of a direct interaction and a second neutron is evaporated from a residual nucleus which is in thermodynamic equilibrium. We can evaluate this component by summing all cases of pre-equilibrium neutron emission after which the escape of a second neutron is energetically possible:

$$\sigma_{n,2n}^{pre}(E_n) = \sigma_a(E_n) \int_0^{E_n + Q_{2n}} P^{pre}(E_n, E) dE, \quad (2)$$

where  $Q_{2n}$  is the energy of the (n,2n) reaction;  $\sigma_a(E_n)$  is the neutron absorption cross-section; and  $P^{pre}(E_n, E)dE$  is the probability of pre-equilibrium emission of a neutron with energy between  $E$  and  $E+dE$ .

Approximating the shape of the pre-equilibrium emission spectrum by a rectangle, we can simplify Eq. (2) to

$$\sigma_{n,2n}^{pze}(E_n) \approx \sigma_\alpha(E_n) P^{pze}(E_n)(E_n + Q_{2n}). \quad (3)$$

In writing the equilibrium component of the cross-section, we allow for the effective reduction in the probability of compound nucleus formation as a result of pre-equilibrium decay. As an approximation we shall assume constant temperature and a constant inverse-reaction cross-section for the neutron:

$$\sigma_{n,2n}^{eq}(2n) = \sigma_\alpha(E_n) \left[ 1 - P^{pze}(E_n)E_n \right] \frac{1}{T_1^2} \int_0^{E_n + Q_{2n}} E \exp(-E/T_1) dE. \quad (4)$$

Here,  $T_1$  is the thermodynamic temperature of the nucleus, related to the level density parameter  $a$  in the Fermi gas model [2] by the relationship  $T_1 \approx \sqrt{\frac{E_n}{a}}$ .

Equations (3) and (4) are valid for neutron energies below the threshold of the  $(n,3n)$  reaction. The equilibrium and pre-equilibrium components of the  $(n,3n)$  cross-section can be written as follows:

$$\left. \begin{aligned} \sigma_{n,3n}^{eq}(E_n) &= \sigma_\alpha(E_n) \left[ 1 - P^{pze}(E_n)E_n \right] \frac{1}{T_1^2} \int_0^{E_n + Q_{3n}} E_1 \exp\left(-\frac{E_1}{T_1}\right) \frac{\int_0^{E_n + Q_{3n} - E_1} E_2 \exp(-E_2/T_2) dE_2}{\int_0^{E_n + Q_{2n} - E_1} E_2 \exp(-E_2/T_2) dE_2} dE_1; \\ \sigma_{n,3n}^{pze}(E_n) &= \sigma_\alpha(E_n) P^{pze}(E_n)(E_n + Q_{3n}) \frac{\int_0^{E_n + Q_{3n}} E_2 \exp(-E_2/T_2) dE_2}{\int_0^{E_n + Q_{2n}} E_2 \exp(-E_2/T_2) dE_2}, \end{aligned} \right\} \quad (5)$$

where  $Q_{3n}$  is the energy of the  $(n,3n)$  reaction and  $T_2 = T_1 \sqrt{1 + Q_{2n}/E_n}$ .

The equations in expression (5) are valid up to the threshold energy of the  $(n,4n)$  reaction. For neutron energies  $E_n > Q_{3n}$ , the  $(n,2n)$  cross-section defined above should be written as  $\sigma'_{n,2n}(E_n > Q_{3n}) = \sigma_{n,2n}(E_n) - \sigma_{n,3n}(E_n)$ . To get a correct

description of the (n,2n) cross-section near the threshold in heavy nuclei we must allow for the competition of gamma-photons from the (n,n'γ) reaction [9]. In this paper, we make approximate allowance for this by means of an effective increase in the reaction threshold,

$$E_{\text{thr}} = -Q_{2n} + \Delta Q. \quad (6)$$

We calculated the shift ΔQ (in megaelectron volts) from the semi-empirical equation

$$\Delta Q = \frac{T^2}{4S_n} \left[ \ln \frac{\sigma_a A \exp(-2S_n/T) 10^8}{4S_n^2} \right]^2, \quad (7)$$

where  $S_n$  is the separation energy of the second neutron ( $S_n = -Q_{2n}$ );  $T = \sqrt{\frac{10E_n}{A}}$  is the nuclear temperature at excitation energy  $E_n$ ; and  $\sigma_a$  is the neutron absorption cross-section in barns [to get an approximate value of which we used the simple expression  $\sigma_a = (1 + 7.5 \cdot A \cdot 10^{-3})$ ].

An analysis of Eq. (7) shows that the shift in the reaction threshold decreases with increasing neutron energy and increases as the values of A and  $S_n$  rise. The effect of the corrections for gamma-photon competition near the threshold of the  $^{197}\text{Au}(n,2n)^{196}\text{Au}$  reaction can be seen from Fig. 1.

Allowance for the competition of the fission channel for fissile nuclei can be made by means of the ratios of neutron to fission widths  $\Gamma_n/\Gamma_f$  obtained from experiment. To check the correctness of the allowance for the fission channel it is useful to calculate the fission cross-section as well. We shall write the relationships for the cross-sections of the reactions (n,f), (n,nf) and (n,2nf) assuming that the fission occurs only from the equilibrium state of the compound nucleus:

$$\left. \begin{aligned} \sigma_{n,f}(E_n) &= [\sigma_a(E_n) - \sigma_a(E_n) P^{pze}(E_n) E_n] K_{A+1}; \\ \sigma_{n,nf}(E_n) &= [\sigma_a(E_n) - \sigma_{nf}(E_n) - \sigma_a(E_n) P^{pze}(E_n) E_n K_A^f] K_A [1 - \exp(-\gamma_A)]; \\ \sigma_{n,2nf}(E_n) &= [\sigma_a(E_n) - \sigma_{nf}(E_n) - \sigma_{n,nf}(E_n) - \sigma_a(E_n) P^{pze}(E_n) E_n K_{A-1}^f] K_{A-1} [1 - \exp(-\gamma_{A-1})], \end{aligned} \right\} \quad (8)$$

where  $K_A^f$  is a coefficient defining the fraction of pre-equilibrium emission events after which fission of the nucleus A is energetically impossible; and  $K_A = 1/[1 + (\Gamma_n/\Gamma_f)_A]$  is the probability of fission of A. The factor  $[1 - \exp(-\gamma_A)]$  is introduced to describe the threshold dependence of the fission cross-section;



$\gamma_A = (E_n - B_A^f)C$ , where  $B_A^f$  is the energy threshold for fission of nucleus A and C is the diffusion coefficient. The fission threshold is found from the semi-empirical equations of Ref. [11]. For the (n,nf) and (n,2nf) reactions, the quantity  $B_A^f$  increases by  $T_n$  and  $2T_n$ , respectively, where  $T_n$  is the temperature of the neutrons emitted before fission. It is assumed that the ratio  $\Gamma_n/\Gamma_f$  depends little on the nucleus excitation energy. Taking this ratio to be constant for a given nucleus, we can obtain from the relationships in expressions (1) to (5) the following analytical equations for calculating the (n,2n) and (n,3n) cross-sections with allowance for the fission channel:

$$\left. \begin{aligned}
 \sigma_{n,2n}(E_n) &= \sigma_a(E_n) F_1 \left\{ B_2 P^{pze}(E_n) + \left[ 1 - P^{pze}(E_n) E_n - \frac{\sigma_{nf}(E_n)}{\sigma_a(E_n)} \right] \times \right. \\
 &\quad \left. \times \left[ 1 - (1 + B_2/T_1) \exp(-B_2/T_1) \right] \right\} - \sigma_{n,2nf} - \sigma_{n,3n}; \\
 \sigma_{n,3n} &= \sigma_a(E_n) F_2 A_1 \frac{A_2 - A_3}{A_2}; \\
 A_1 &= B_3 P^{pze}(E_n) + \left[ 1 - P^{pze}(E_n) E_n - \frac{\sigma_{nf}(E_n)}{\sigma_a} - \frac{\sigma_{n,nf}(E_n)}{\sigma_a} \right] \left[ 1 - (1 + B_3/T_1) \exp(-B_3/T_1) \right]; \\
 A_2 &= 1 - (B_3/T_1 + 1) \exp(-B_3/T_1); \\
 A_3 &= (T_3/T_1)^2 \exp(-B_3/T_1) \left\langle (B_3/T_2 + 1) \left[ (B_3/T_3 - 1) \exp(B_3/T_3) + 1 \right] - \right. \\
 &\quad \left. - \left\{ \left[ \frac{B_3}{T_3} \frac{B_3}{T_2} - 2 \frac{T_3}{T_2} (B_3/T_3 - 1) \right] \exp(B_3/T_3) - 2 \frac{T_3}{T_2} \right\} \right\rangle.
 \end{aligned} \right\} \quad (9)$$

Here,  $B_2$  and  $B_3$  are the thresholds of the (n,2n) and (n,3n) reactions, respectively;  $T_3 = (T_1 - T_2)/T_1 T_2$ ;  $F_1 = \beta_1/(1 + \beta_1)$ ;  $F_2 = \beta_2/(1 + \beta_2)$ ;  $\beta_1 = (\Gamma_n/\Gamma_f)_A$  and  $\beta_2 = (\Gamma_n/\Gamma_f)_{A-1}$ .

The quantity  $P^{\text{pre}}(E_n)$  can be approximated [8] by the expression  $P^{\text{pre}}(E_n) = 5 \times 10^{-2} \sigma_a / AE_0 (E_n/E_0)^2$ , where  $E_0 = E_n + S_n$  is the excitation energy of the compound nucleus.

Thus to calculate the (n,2n) and (n,3n) cross-sections from the equations in expression (9) and the (n,f), (n,nf) and (n,2nf) cross-sections from expression (8) we need to know only three parameters: the neutron absorption cross-section  $\sigma_a(E_n)$ , the level density parameter  $a$  and the ratio  $\Gamma_n/\Gamma_f$  for the nuclei  $A + 1$ ,  $A$  and  $A - 1$ . Either experimental data obtained in independent measurements or theoretical evaluations may be used for these parameters. Consequently, the equations in expressions (8) and (9) can also be used for predicting cross-sections in the absence of experimental data on the (n,f) and (n,2n) reactions. Although the three parameters can be chosen individually for each nucleus, it is better to use systematized sets of data for large-scale calculations. In this paper we have used the following scheme:

- (1) The neutron absorption cross-section (in millibarns) was approximated by the equation  $\sigma_a = (1000 + 7.5A)$ . Calculations from this equation agree well with results from the optical model;
- (2) The level density parameter is chosen to provide the best possible description of the excitation function of the (n,2n) and (n,3n) reactions over a wide range of nuclei ( $100 \leq A \leq 200$ ) and is determined from the relationship

$$a = \frac{A}{12} - \frac{400}{36 + (A - 208)^2}.$$

The second term in this equation has been introduced to describe the sudden drop in the parameter  $a$  in the region close to the doubly magic lead nucleus. The relationship agrees well with systematized values of the level density parameter allowing for collective effects in heavy nuclei [12];

- (3) The parameters  $\Gamma_n/\Gamma_f$  are taken from the systematized data in Ref. [13], which uses both experimental and calculated values corresponding to incident neutron energies of 3-4 MeV or composite nucleus excitation energies of 8-10 MeV. The ratio of the widths was assumed to be independent of the excitation energy and was approximated by an exponential function of the fissionability parameter  $Z^2/A$ :

$$\Gamma_n/\Gamma_f = \exp[-\alpha(Z^2/A - \beta)]. \quad (10)$$

The coefficients  $\alpha$  and  $\beta$  for different elements are shown in Table 1.

It is shown in Ref. [13] that a simple relationship like Eq. (10), reflecting the properties of the liquid drop model, can be used only for a limited group of nuclei for which shell effects in the fission barrier structure are small. For nuclei with proton and neutron numbers ( $Z$  and  $N$ ) outside the region  $90 \leq Z \leq 95$  and  $140 \leq N \leq 146$ , the dependence of  $\Gamma_n/\Gamma_f$  on  $Z$  and  $N$  may be quite different from that given by Eq. (10) as a result of the shell correction. In these nuclei we may also expect a more marked dependence on energy in connection with rearrangement of the shell structure of the nucleus. It should be noted, however, that these effects can be taken into account quite simply here by giving each nucleus the appropriate value of  $\Gamma_n/\Gamma_f$ . The method used in this paper also allows us to take into account the energy dependence of  $\Gamma_n/\Gamma_f$ ; this leads to more complicated cross-section expressions which have to be evaluated by numerical methods.

Discussion of the results. A FORTRAN-IV computer program (SIMPL) was written to calculate the cross-sections from the relationships given above. The results for the cross-section of the reaction  $^{197}\text{Au}(n,2n)^{196}\text{Au}$  are shown together with the available experimental data in Fig. 1. The dashed line denotes the results of a calculation which does not allow for gamma-photon competition [ $\Delta Q = 0$  in Eq. (6)]. A comparison between calculation and experiment for nuclei with  $100 \leq A \leq 200$  shows that the error in the predictions of the  $(n,2n)$  cross-sections by this method is less than 15%.

Allowance for the fission channel increases the possible error in the predicted  $(n,2n)$  cross-section values to a maximum of about 30% for nuclei with a fissionability parameter  $\Gamma_f/(\Gamma_n + \Gamma_f) \lesssim 0.5$ ; for isotopes with a higher value of this parameter, the accuracy is further reduced. The reliability of the evaluations may be checked by comparing the calculated fission cross-sections with experimental values. It can be seen from Fig. 2 that the agreement between calculation and the ENDF/B library evaluation [14] (based on experimental data) is on average better than 10%. The monotonic decrease in the partial contribution of  $(n,f)$  can be explained by the effect of pre-equilibrium emission. Figure 3 shows the  $(n,2n)$  and  $(n,3n)$  excitation functions for the isotopes  $^{232}\text{Th}$  and  $^{238}\text{U}$ . The cross-sections of these isotopes have been studied experimentally in more detail than those of others. The agreement between the calculated results and experiment is good, both the absolute values and the shape of the excitation function curves being very similar.

For the  $^{238}\text{U}$  nucleus, we also show the results of a calculation without allowance for pre-equilibrium neutron emission, using the statistical model alone.

It can be seen from Fig. 3 that allowance for pre-equilibrium emission reduces the (n,2n) cross-section at the maximum of the excitation function and increases it for incident neutron energies above the threshold of the (n,3n) reaction. There is a corresponding and quite significant reduction in the cross-section for the (n,3n) reaction.

Any attempt to describe the experimental data on (n,2n) and (n,3n) cross-sections within the framework of statistical theory alone involves introducing a considerable reduction in the level density parameter  $a$  and the neutron absorption cross-section  $\sigma_a$ . Pearlstein [1] and Segev and Caner [6], for example, had to choose a parameter  $a$  equal to  $A/22$  to describe the (n,2n) cross-sections, and this is about one half of the actual value of the parameter obtained from other data.

The (n,2n) and (n,3n) excitation functions calculated for  $^{239}\text{Pu}$  are shown in Fig. 3c. The agreement between the calculated curve and the direct measurements made by Mather (taken from Ref. [6]) is not satisfactory. The figure also shows the data from Ref. [15], evaluated from measurements of the cross-sections for the (d,f), (d,2n) and (t,f), (t,2n) reactions. The target nuclei in this paper were chosen so that the composite nucleus obtained was the same as that in the neutron-induced reaction. The (n,2n) cross-sections were determined from known values of  $\sigma_{n,f}$ ; it was assumed that the probability of decay of the compound nucleus was independent of its method of formation. In other words, the data in Ref. [15] were obtained on the assumption that no direct inelastic scattering reaction mechanism exists. This is also clear from a comparison of the data from Ref. [15] with the dashed curve in Fig. 3c, which was calculated on the basis of a statistical model without allowance for pre-equilibrium emission. The agreement between the dashed curve and the data from Ref. [15] provides indirect confirmation that the statistical theory parameters have been correctly chosen. The inclusion of non-equilibrium processes in the inelastic scattering mechanism on the other hand, leads to the excitation function shown by the continuous curve in the figure. Additional measurements of the (n,2n) cross-section for this nucleus would be very useful.

Figure 4 shows the values calculated in this paper for the fission cross-sections and (n,2n) and (n,3n) cross-sections of  $^{237}\text{Np}$  compared with available experimental data and the ENDF/B and ENDL library evaluation [16]. The (n,2n) excitation function values calculated in this paper agree well with the experimental data from Refs [17] and [18]. The fact that the data from Ref. [15] lie somewhat above the calculated curve is due, as in the case of  $^{239}\text{Pu}$ , to neglect of non-equilibrium processes in that paper. The difference in the (n,2n) cross-sections

recommended in the present paper and in the ENDF/B library seems to be caused by lack of agreement between the evaluated fission cross-sections (the ENDF/B evaluation curve is much higher than our recommendations and those in the ENDL library, which agree quite well with each other).

Figure 5 shows the fission cross-sections and (n,2n) and (n,3n) excitation functions for the isotopes  $^{233}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{242}\text{Pu}$ . Of the (n,2n) excitation functions, experimental data are available only for  $^{235}\text{U}$ . The curve calculated in this paper shows satisfactory (within 30%) agreement with these data (except for the points from Ref. [6] corresponding to  $E = 14$  MeV). Measurements in the fission neutron spectrum for the  $^{233}\text{U}(n,2n)^{232}\text{U}$  reaction have been published in Ref. [21]. The value of the cross-section obtained in that paper ( $\langle \sigma_{n,2n} \rangle^E = 4.08 \pm 0.3$  mb) agrees satisfactorily with the result of integration over the spectrum of the excitation function calculated in the present paper ( $\langle \sigma_{n,2n} \rangle^T = 4.48$  mb; see Table 2). For the remaining two isotopes considered in Fig. 5,  $^{234}\text{U}$  and  $^{242}\text{Pu}$ , there is no experimental information from which the reliability of the (n,2n), (n,3n) excitation function evaluations can be assessed. The calculated value of the  $^{234}\text{U}$  fission cross-section agrees satisfactorily with the ENDF/B evaluation while the (n,2n) cross-section is greater than the library data. For  $^{242}\text{Pu}$ , the calculated fission cross-section is slightly smaller than the result given in Ref. [19], the ENDF/B library value and the value recommended on the basis of the experimental data. This may possibly be explained by an incorrect approximation of the quantity  $\Gamma_n/\Gamma_f$  for the  $^{242}\text{Pu}$  nucleus in the present paper. In evaluating the excitation function of the  $^{242}\text{Pu}(n,2n)^{241}\text{Pu}$  reaction, we therefore use a fission cross-section renormalized to the ENDF/B library recommendation.

Figure 6 gives the results obtained for the isotopes  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{231}\text{Pa}$  and  $^{233}\text{Pa}$  compared with other evaluations. The calculation of the  $^{241}\text{Pu}(n,2n)^{240}\text{Pu}$  reaction carried out in Ref. [22] for energies below 11 MeV produces a considerably higher value than the present paper; the (n,3n) cross-section is also higher than our evaluation.

The  $^{231}\text{Pa}$  fission cross-section we have calculated is in fairly good agreement with the ENDF/B evaluation but the (n,2n) cross-section is considerably smaller. There are no experimental data on the  $^{233}\text{Pa}$  fission cross-section in the energy range of interest and so all the evaluations quoted are based on calculations. It should be noted that there is a large discrepancy between the curves in Fig. 6.

The (n,2n) cross-section evaluated in Ref. [22] is close to that recommended here, although the evaluated cross-section is considerably higher. This is probably due to the fact that non-equilibrium effects are not taken into account in Ref. [22].

Figures 7a and 7b show the results of various evaluations for curium isotopes. It can be seen that there is a considerable difference between the results; this is due in particular to the absence of sufficiently reliable experimental data. Another important reason for the discrepancy between the evaluated cross-sections for fission and the (n,2n) and (n,3n) reactions could be the difference in the methods used. Thus, allowance for non-equilibrium processes in nuclei with a large fissionability parameter strongly affects the result of the cross-section calculation. A large contribution to the (n,2n) cross-section in the isotopes  $^{241}\text{Cm}$  and  $^{242}\text{Cm}$ , for example, comes from the non-equilibrium component and the neutron channel is therefore not completely suppressed by fission as it would be in a purely statistical reaction mechanism. This is probably the reason for the difference in the (n,f), (n,2n) and (n,3n) cross-sections recommended in this paper and in the ENDF/B library. Our calculation of the (n,2n) and (n,3n) cross-sections averaged over the fission neutron spectrum are shown in Table 2 in comparison with the results of the evaluation in Ref. [1].

Thus, allowance for non-statistical effects in the neutron channel during the interaction of fast neutrons with fissile nuclei has a considerable effect on the absolute value and the energy dependence of the neutron cross-sections. In the method we propose here for calculating the cross-sections, unified sets of parameters are used and there is no fitting to any particular experimental result. The good agreement with the experimental data for nuclei with a relative atomic mass  $10 \lesssim A \lesssim 200$  gives reason to hope that predictions made by this method for fissile nuclei in the region where no experimental data exist will also be fairly reliable.

To evaluate the (n,2n) and (n,3n) excitation functions in fissile nuclei we have used a general approach with a systematized set of calculation parameters so that we have been able to predict the cross-section to within about 30% for nuclei with a fission parameter  $\Gamma_f/\Gamma \lesssim 0.5$ .

The accuracy of the excitation function evaluations made by this method can be improved by, firstly, choosing accurate values of  $\Gamma_n/\Gamma_f$  for individual nuclei and, secondly, allowing for the possible dependence of this ratio on the compound nucleus excitation energy.

Table 1. Parameters used in Eq. (10) for the ratio of the neutron and fission widths of various isotopes

Isotope	Parameter		Isotope	Parameter	
	$\alpha$	$\beta$		$\alpha$	$\beta$
$^{91}\text{Pa}$	4.12	35.82	$^{94}\text{Pu}$	2.0	36.6
$^{92}\text{U}$	2.37	36	$^{95}\text{Am}$	1.47	37
$^{93}\text{Np}$	2.44	36.34	$^{96}\text{Cm}$	1.12	37.3

Table 2. Cross-sections for (n,2n) and (n,3n) reactions, averaged over the fission spectrum

Isotope	$\langle \sigma_{n,2n} \rangle, \text{Mb}$		$\langle \sigma_{n,3n} \rangle, \text{Mb}$	
	Present paper	Ref. [1]	Present paper	Ref. [1]
$^{232}\text{Th}$	15,4	I6	II8	210
$^{233}\text{U}$	4,48	3,3	2,04	6
$^{234}\text{U}$	3,30	7,0	4,92	42
$^{235}\text{U}$	16,2	I5	12,8	45
$^{238}\text{U}$	14,1	I5	71,3	I40
$^{231}\text{Pa}$	5,44	-	7,68	-
$^{233}\text{Pa}$	II,0	-	27,7	-
$^{237}\text{Np}$	3,5	I,3	6,7	9,0
$^{239}\text{Pu}$	5,72	I,9	3,9	5,5
$^{240}\text{Pu}$	4,25	-	9,4	-
$^{241}\text{Pu}$	12,0	-	I4	-
$^{242}\text{Pu}$	9,0	-	22,8	-
$^{241}\text{Cm}$	2,16	-	I,0	-
$^{242}\text{Cm}$	I,65	-	2,1	-
$^{243}\text{Cm}$	5,45	-	4,4	-
$^{244}\text{Cm}$	3,02	-	7,0	-
$^{245}\text{Cm}$	7,32	-	7,2	-
$^{246}\text{Cm}$	6,57	-	13,0	-
$^{247}\text{Cm}$	I7,6	-	28,0	-
$^{248}\text{Cm}$	8,55	-	35,4	-

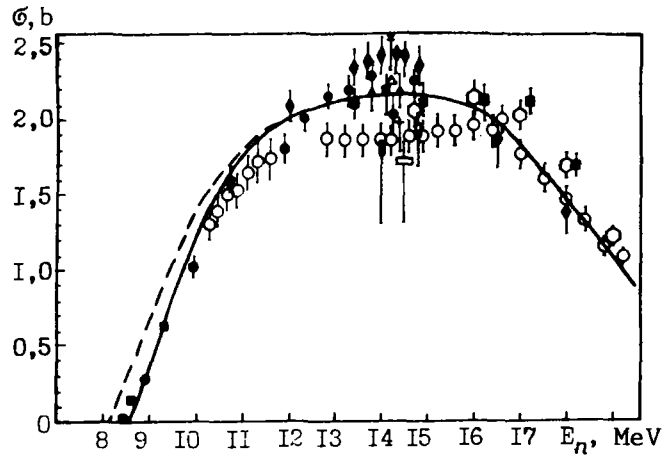


Fig. 1. Excitation function of the  $^{197}\text{Au}(n,2n)^{196}\text{Au}$  reaction calculated in the present paper (continuous curve) compared to the available experimental data from various sources (points). The dashed curve is derived from a calculation which does not allow for gamma-photon competition.

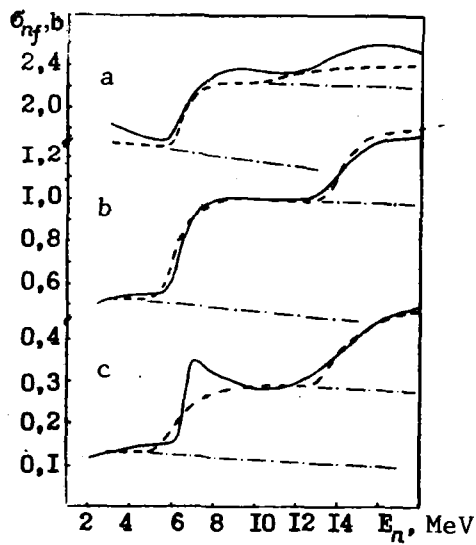


Fig. 2. Cross-sections for the  $(n,f)$  reaction in  $^{239}\text{Pu}$  (a),  $^{238}\text{U}$  (b) and  $^{232}\text{Th}$  (c): - - - calculation from Eq. (8) in the present paper; — ENDF/B library evaluation; — · — partial contributions of the  $(n,f)$  and  $(n,nf)$  processes calculated in this paper.



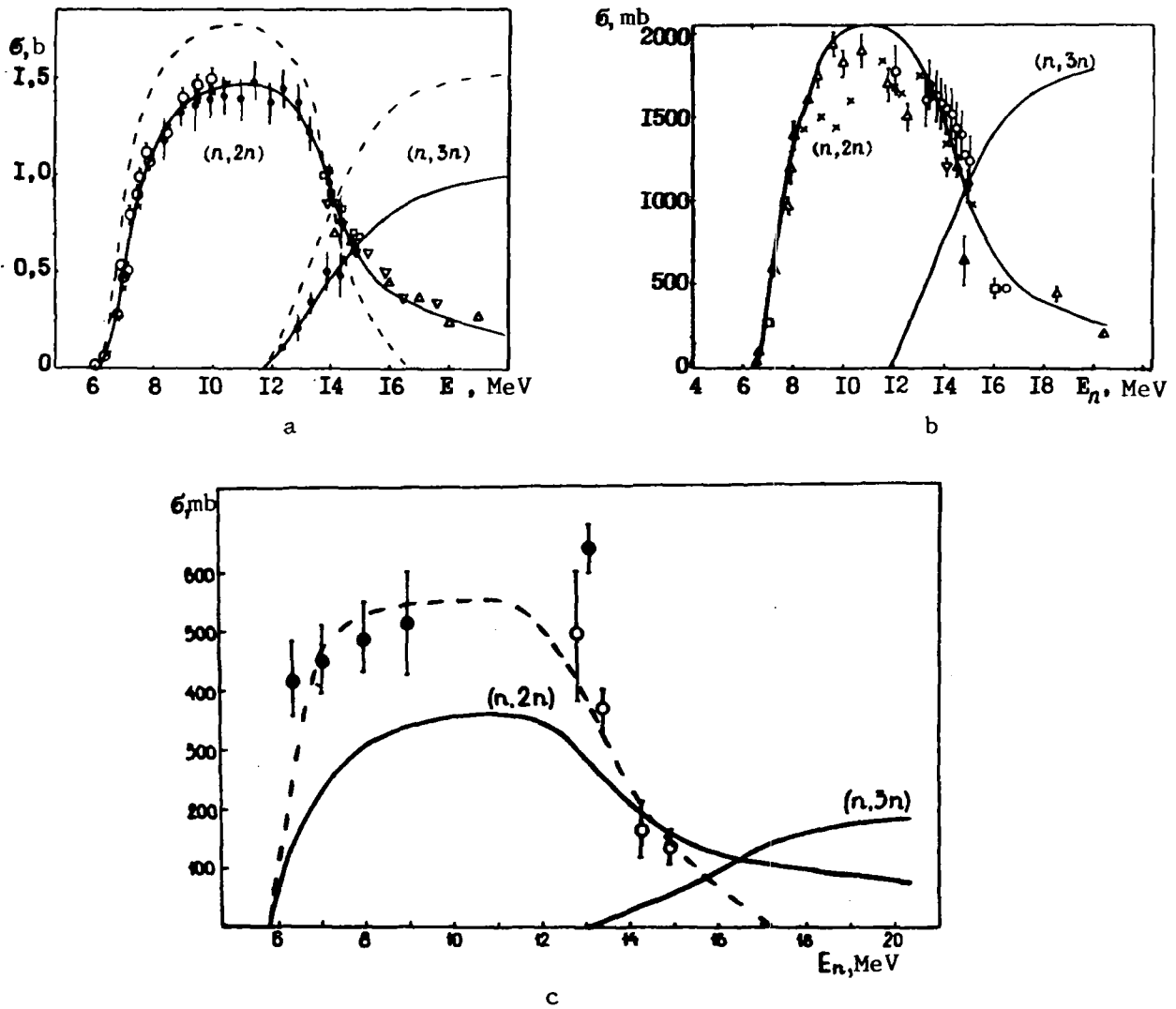


Fig. 3. Excitation functions of the (n,2n) and (n,3n) reactions in  $^{238}\text{U}$  (a),  $^{232}\text{Th}$  (b) and  $^{239}\text{Pu}$  (c): — result of calculation by the method proposed in the present paper; - - - calculation ignoring pre-equilibrium neutron emission; the points are experimental data from various sources. In Fig. 3c:  $\bullet$  - data taken from Ref. [6];  $\circ$  - data from Ref. [15].

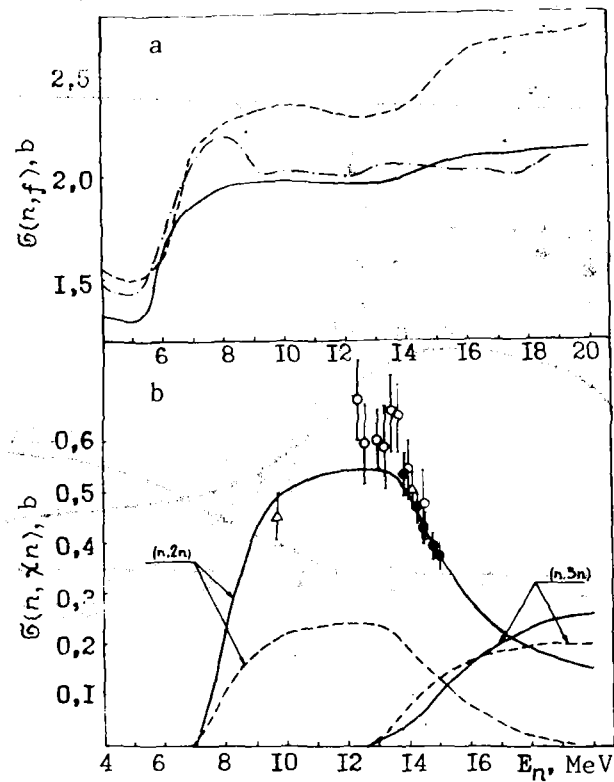


Fig. 4. Fission cross-sections (a) and  $(n,2n)$  and  $(n,3n)$  cross-sections (b) for  $^{237}\text{Np}$ : — calculation in the present paper; - - - ENDF/B library evaluation; — · — ENDL library evaluation;  $\odot$  - data from Ref. [15];  $\triangle$  - from Ref. [18];  $\blacksquare$  - from Ref. [17].

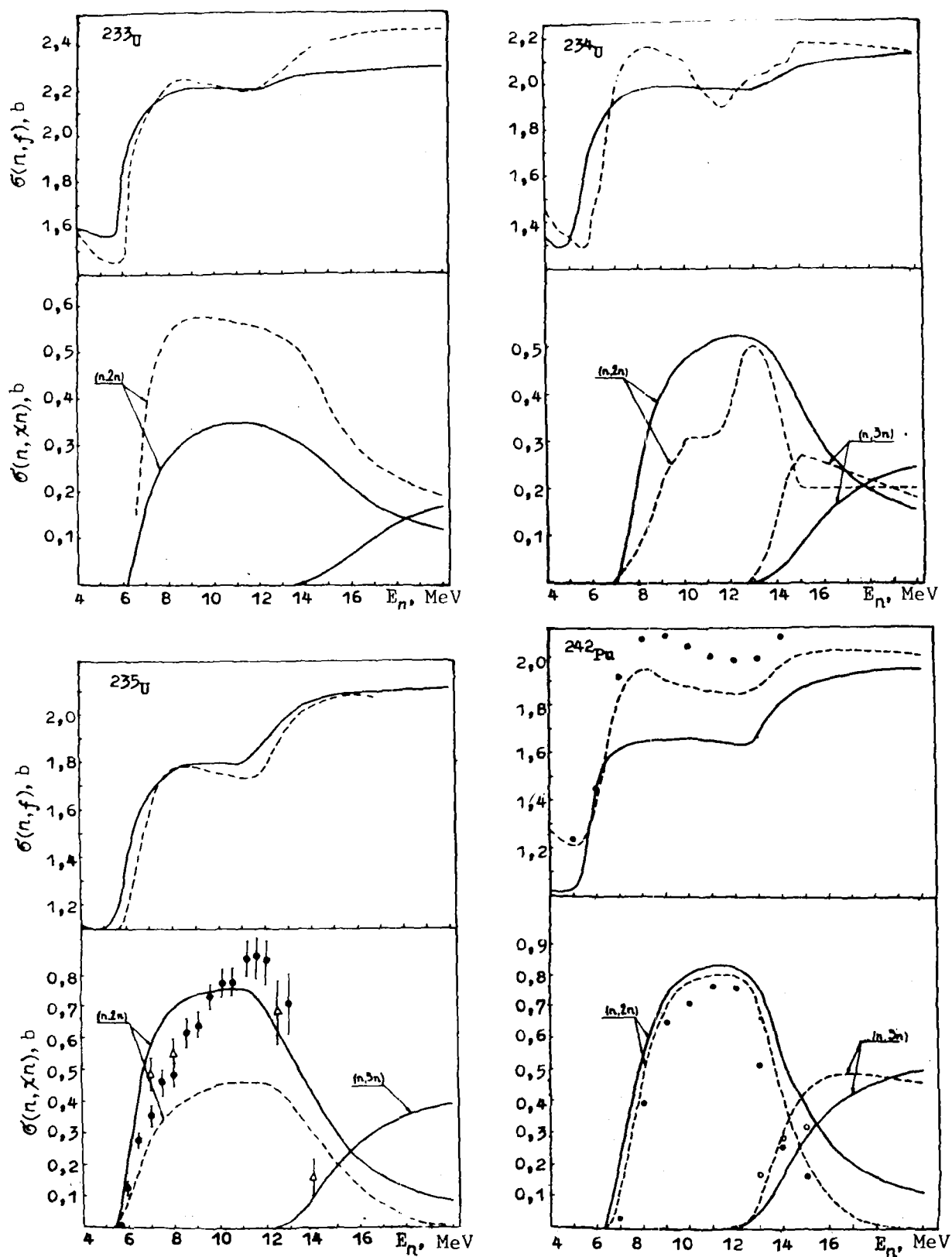


Fig. 5. Fission cross-sections and (n,2n) and (n,3n) excitation functions for the isotopes  $^{233}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{242}\text{Pu}$ : — calculation in the present paper; - - - ENDF/B library evaluation; ●, ○ - evaluations in Ref. [19]; ▲ - Mather's experimental data, taken from Ref. [6]; ■ - experimental data from Ref. [20].

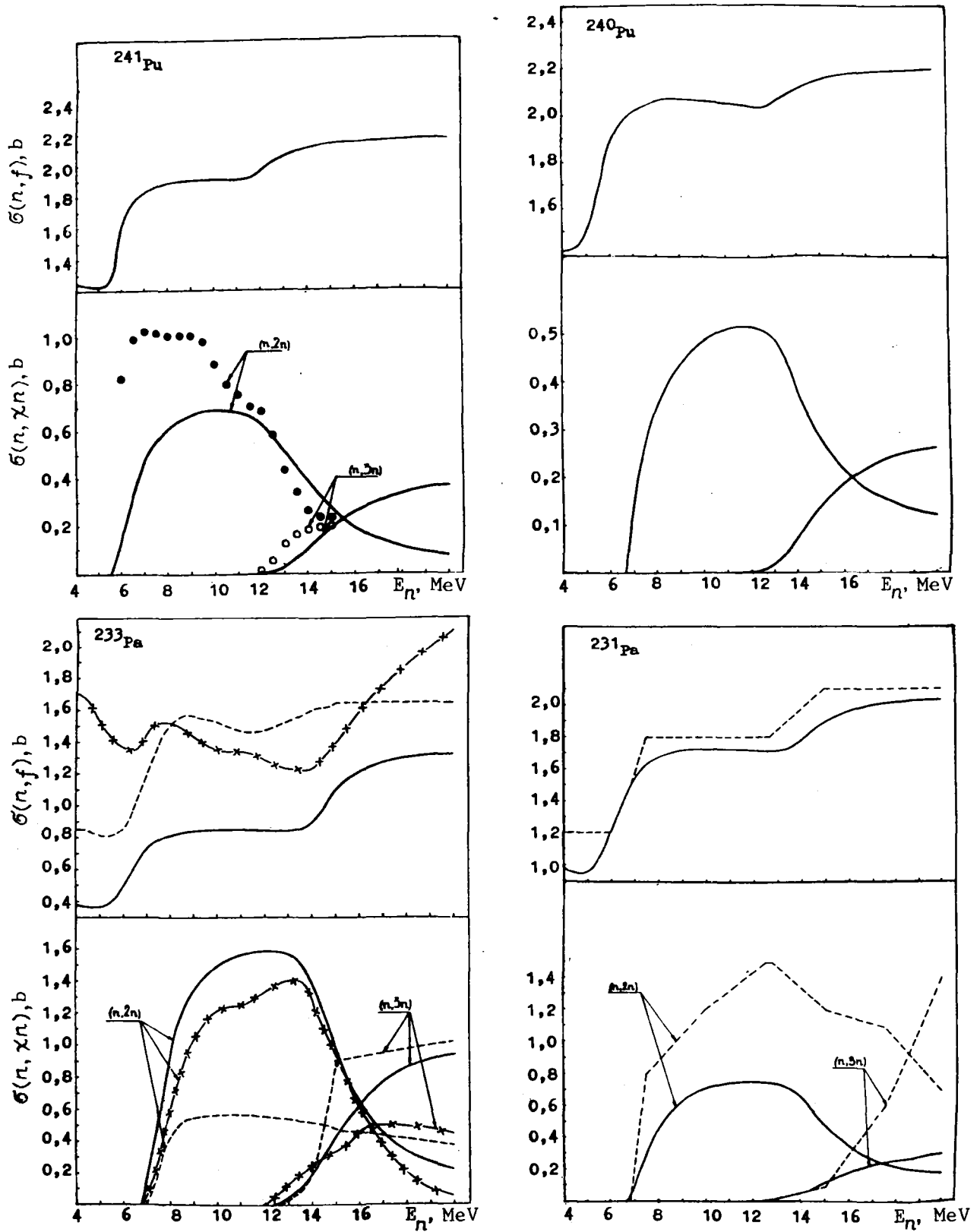


Fig. 6. Fission cross-sections and  $(n,2n)$  and  $(n,3n)$  excitation functions for the isotopes  $^{241}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{233}\text{Pa}$  and  $^{231}\text{Pa}$ : — calculation in the present paper; - - - ENDF/B library evaluation; ●, ○ - evaluation in Ref. [19]; —x—x result of the evaluation in Ref. [22].

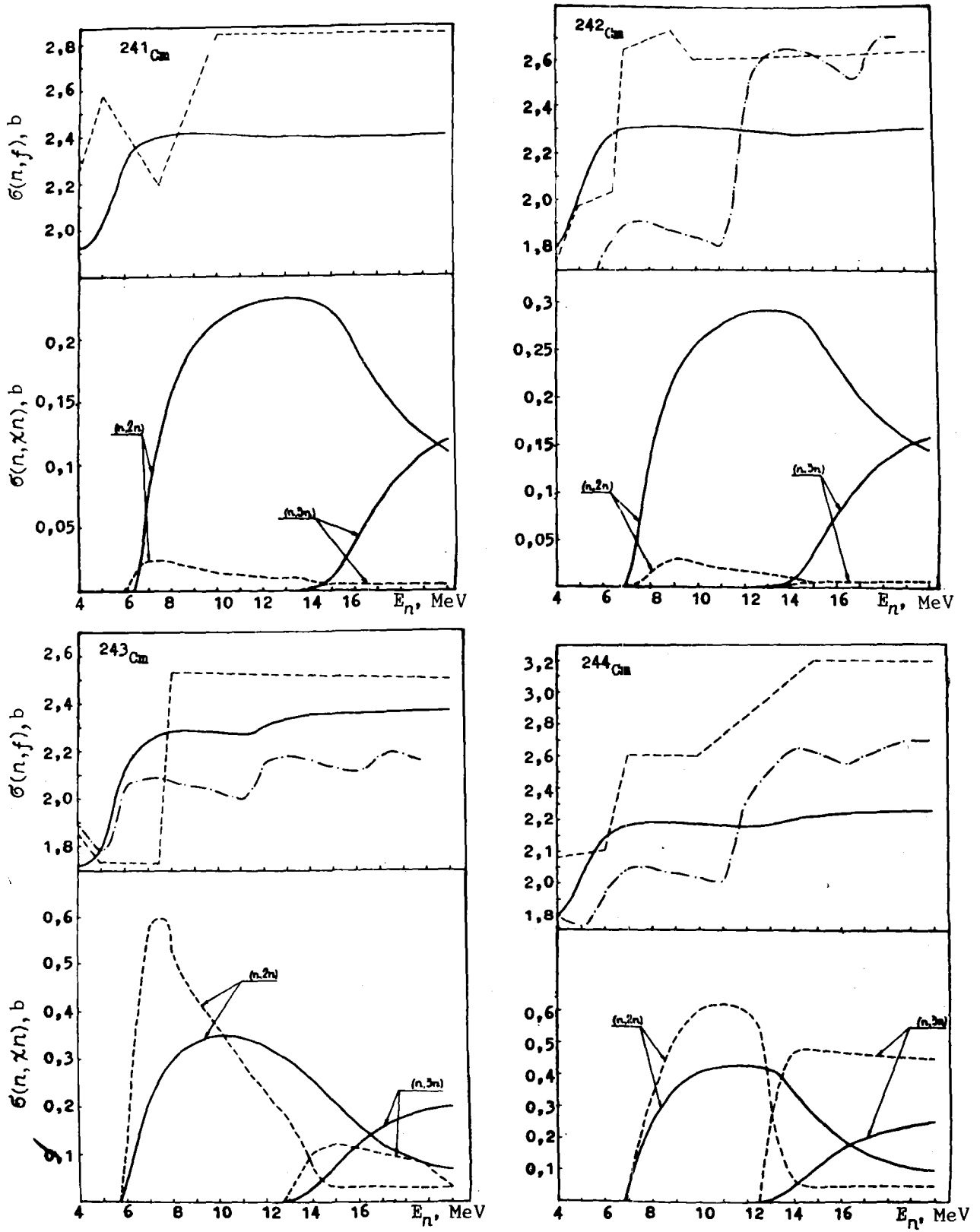


Fig. 7a. Fission cross-sections and  $(n,2n)$  and  $(n,3n)$  excitation functions for the isotopes  $^{241}\text{Cm}$ ,  $^{242}\text{Cm}$ ,  $^{243}\text{Cm}$  and  $^{244}\text{Cm}$ : — calculation in this paper; - - - ENDF/B library evaluation; — · — ENDL library evaluation.

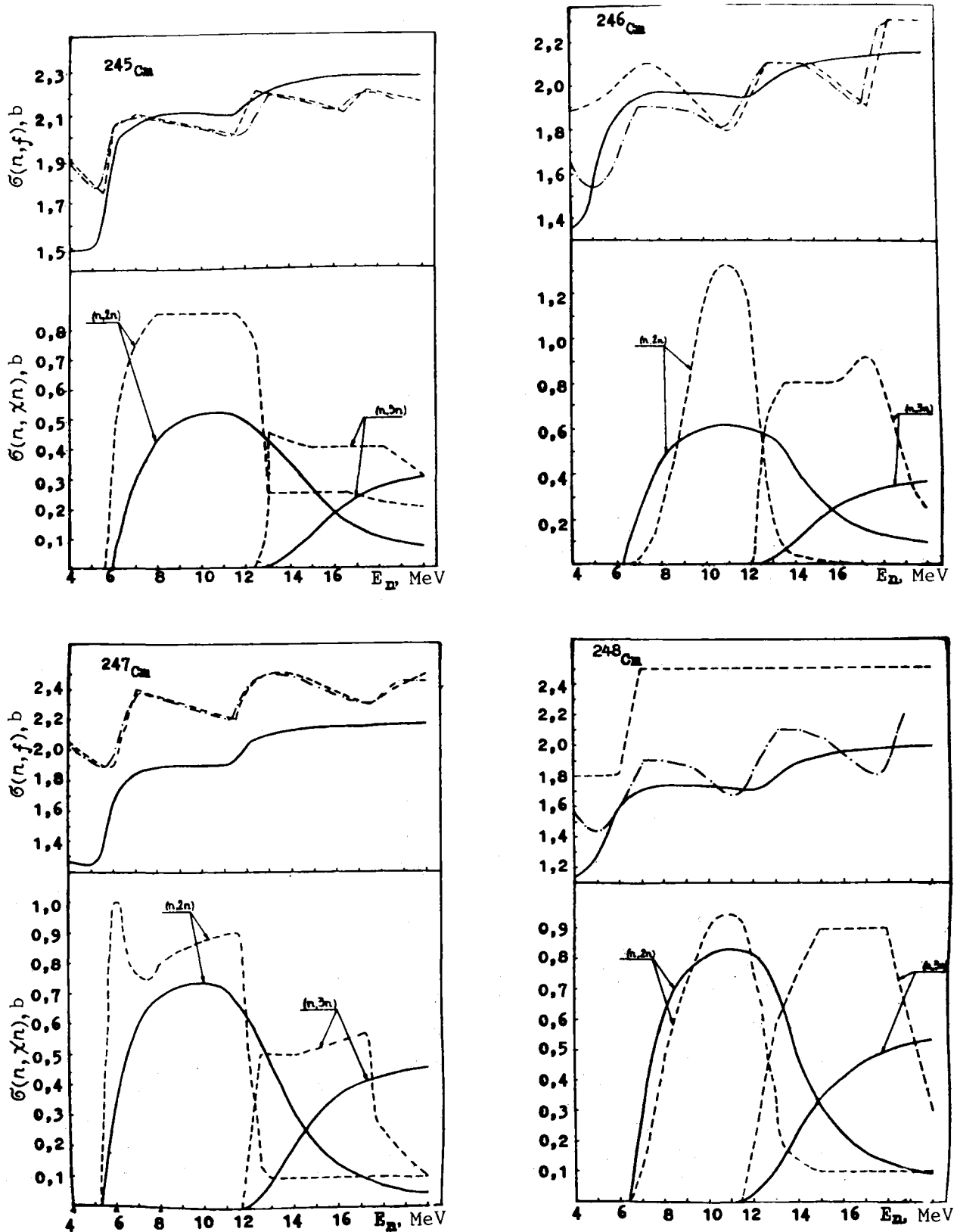


Fig. 7b. Fission cross-sections and  $(n,2n)$  and  $(n,3n)$  excitation functions for the isotopes  $^{245}\text{Cm}$ ,  $^{246}\text{Cm}$ ,  $^{247}\text{Cm}$  and  $^{248}\text{Cm}$ : — calculation in this paper; - - - ENDF/B library evaluation; — · — · ENDL library evaluation.

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