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# GROUP CROSS-SECTIONS AND RESONANCE SELF-SHIELDING FACTORS FOR $^{239}$Pu IN THE UNRESOLVED RESONANCE REGION

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## ABSTRACT

The authors analyse experimental data on the transmission and fission self-indication functions for $^{239}$Pu in the unresolved resonance region. Use is made of the method of generating a cross-section structure based on the multi-level R-matrix formalism (stochastic K-matrix method). Evaluations of the average resonance parameters and group constants for $^{239}$Pu are made.

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Plutonium-239 is the main component of breeder reactor fuel and therefore very high requirements are imposed with regard to the accuracy of nuclear data for this isotope. A special role is played by the region of unresolved resonances. For example, for neutron energy below 20 keV about half of all events of neutron radiative capture by $^{239}$Pu nuclei and about 20% of the fission events occur in the core of a high-power breeder. If the calculation accuracy required for $K_{eff}$ and the breeding ratio are 1% and 2%, respectively, we obtain maximum permissible errors of about 7% for the radiative capture cross-section and about 2% for the fission cross-section. Although the methodological accuracies of measurement of the above types of average cross-sections achieved so far are close to what is required, at neutron energies above 20 keV the true accuracy of the evaluated data in the unresolved resonance region is appreciably poorer. Thus, for this region, according to Ref. [1], the difference between the evaluated data for $\sigma_f$ ($^{239}$Pu) is as much as 10-12%. A similar situation with respect to the $\sigma_c$ ($^{239}$Pu) data is attributed to the greater difficulty of measuring cross-sections in the unresolved resonance region.

Particular attention has been paid lately to the importance of knowing not only the average cross-sections but also the resonance self-shielding factors. For example, in Ref. [2], Rowlands pointed out the need for determining them with an accuracy of about 1%. Calculation on the basis of average resonance parameters does not unambiguously yield resonance self-shielding factors of the same accuracy even if these average resonance parameters are well "fitted" to the average cross-sections. It is essential to check the average resonance parameters against the measured transmissions (and self-indication functions). In a more general formulation of the problem we have to test the average resonance parameters against a wide set of microscopic experimental data in order to ensure that the group constants derived on their basis have an accuracy which satisfies the requirements of reactor calculations. Thus, the evaluations given in Ref. [3] show that a 10% uncertainty in the average resonance parameters for $^{239}$Pu gives rise to the following errors: about 0.5% for $K_{eff}$ and about 1% for the breeding ratio, owing to the uncertainty of parameters $\bar{\Gamma}_\gamma$ and $\bar{\Gamma}_f$, and errors approximately twice as great owing to the uncertainty of parameters $g\Gamma_n$ and $\bar{D}$ (or strength functions).

Another pressing problem concerns the reliability attained in calculating the Doppler coefficients, which depends entirely on knowing the resonance structure of the neutron cross-sections in the unresolved resonance region and on our ability to describe parametrically the corresponding physical phenomena for fissile nuclei. Unfortunately, the presence of many spin and parity states, strong inter-resonance interference effects, the potential effects of intermediate structure in neutron cross-sections and the specific nature of the fission reaction lead to difficulties or doubts when we try to use a simple theoretical scheme to interpret experimental data on the neutron cross-sections of fissile isotopes and to extend it to the region of high temperatures. Evaluators are therefore compelled to resort to "subjective" solutions when faced with contradictory experimental data and sometimes choose a "speculative" calculation model.

The purpose of the present work is to analyse experimental data on average cross-sections and transmission functions of the type $T(n) = 1/\Delta u \int_{\Delta u} \exp[-\sigma_t(u)n]\,du$ and $T_f(n) = 1/\langle\sigma_f\rangle \int_{\Delta u} \sigma_f(u)\exp[-\sigma_t(u)n]\,du$ for the case of $^{239}$Pu in order to evaluate its average resonance parameters and, on their basis, derive the group constants (average cross-sections and resonance self-shielding factors) within the framework of the conventional R-matrix formalism. For this purpose, we had to develop a multi-level method of calculating such substantially non-linear functionals as transmission functions and resonance self-shielding factors, together with an optimization technique based on that of sensitivity coefficients. This is the way of deriving self-consistent constants with estimation of the confidence interval (covariance matrix).

Brief description of experimental data

We measured the transmission function T(n) and the fission-reaction self-indication function $T_f(n)$ with the time-of-flight spectrometer belong to the IBR reactor at the JINR Neutron Physics Laboratory (Dubna). We used the pulsed source in two operating regimes - a reactor regime (path length $\ell$ = 1000 m, burst duration $\tau \approx 100$ μs with the repetition frequency f = 5 Hz, reactor power W = 30 kW, and a booster regime in the linear electron accelerator ($\ell \approx 100$ m, $\tau \approx 3$ μs, f = 50–100 Hz, W ≈ 5–10 kW). The neutron spectrum of the scource was close to the Fermi slowing-down spectrum. An array of $^{3}He$-counters was used as the neutron detector. The fission process was recorded by a multi-layer fission ionization chamber with about 600 mg of $^{239}Pu$ and a fission fragment recording efficiency of about 50%. Alpha-pulse discrimination was made possible by means of high-speed electronics. The filter-samples of metallic plutonium were 50 mm in diameter and had a high-purity and ultra-high-purity (set of small thicknesses) chemical and isotopic composition. The samples were enclosed in a thin-walled steel shell. The measurements were made relative to equivalent empty shells, and the background was determined by the resonance filter method. In the measurements with $^{3}He$-counters the background was low (1–2% in the reactor regime) and depended hardly at all on the thickness of the filter sample. In the fission chamber measurements (mainly in the booster regime) the background amounted to 10–20% and a considerable part of the measurement time was spent on determining it. The experiment is described in Ref. [4].

A special feature of the measurements made with samples of fissile material is that the latter must have a small diameter in order to satisfy the requirements of operational safety. The diameter of the collimating holes in the gap in the neutron guide and near the fission chamber was about 40 mm, which substantially reduced the counting rate. The errors in measuring the function $T_f(n)$ were due to the counting statistics when measuring each cycle: open beam, sample and resonance filter. The reliability of the measurement of function T(n) was greater on account of the higher efficiency of the neutron detector and better background conditions. The experimental points on the T(n) and $T_f(n)$ curves, which we analysed in our study, are shown in Fig. 1, where the points obtained in the experiments carried out by Czirr and Bramblett [5] are also given.

The paper by the two mentioned authors [5], apart from ours [4], is the only one where $T_f(n)$ was measured as a function of neutron energy for $^{239}Pu$. However, from the standpoint of analysing the data in the unresolved resonance region, the results given in Ref. [5] (see Fig. 1) provide virtually no information. In fact, the authors only measured the self-indication function $T_f(n)$, but without measurements of transmission T(n) under the same conditions the evaluation of the resonance self-shielding factors is unreliable. As we know, this factor is defined as the ratio of the areas under the curves: $f_f(\sigma_0) = \int_0^\infty T_f(n)\exp(-\sigma_0 n)dn / \int_0^\infty T(n)\exp(-\sigma_0 n)dn$. Consequently, we need to try to measure both types of curve, but if we

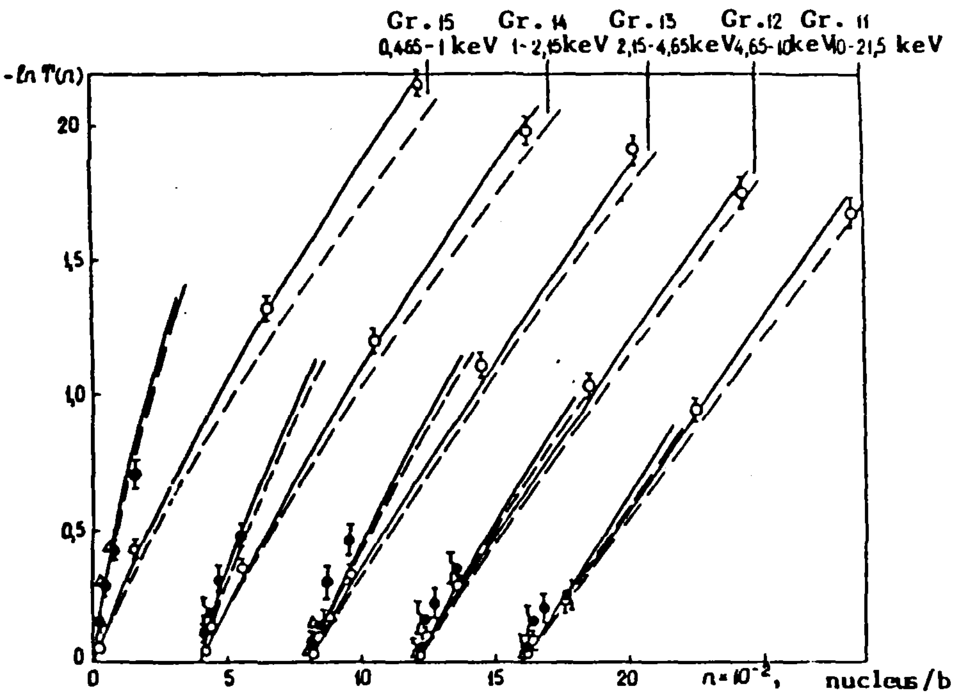


**Fig. 1. Group transmission functions T(n) (O) and $T_f(n)$ (. - [4], Δ - [5]) for $^{239}Pu$. The solid curves (the lower one for T(n) and the upper one for $T_f(n)$ represent calculations based on optimized parameters, and the dashed curves (same notation) calculations based on the parameters recommended in Ref. [6]. The origin is shifted along the n scale of each successive graph.**

measure only one of the functions $T_f(n)$ and T(n), the latter should be preferred since it gives us more information - there is better measurement accuracy and the levels of attenuation achievable in practical measurements are deeper. In Ref. [5] $T_f(n)$ was measured only at the initial section of sample thickness, and results are given for averaging intervals $\Delta E_n$ equal to 1-10, 0.465-1 keV, and for groups with lower energies (unresolved resonance region). In Ref. [4] we attempted to measure $T_f(n)$ for neutron energy below 21 keV. However, the reliability of these measurements declines with an increase in energy because of difficulties in measuring the background. As will be seen from Fig. 1, as the energy rises, the errors in $T_f(n)$ increase; there is then greater spread of the data and in groups 11-13 ($E_n$ > 2 keV) the experimental points tend to shift systematically.

Theoretical method

In the case of heavy fissile nuclei we have to take into account the effect of inter-resonance interference in the energy behaviour of neutron cross-sections. We used the well-known Reich-Moore approximation, in which the elements of the R-matrix for the fixed moment and parity state are expressed in the form

$$R_{cc'}(E) = \sum_{\lambda} \frac{\gamma_{\lambda c}\gamma_{\lambda c'}}{E_\lambda - E - i\bar{\Gamma}_\gamma/2}, \qquad \text{(A)}$$

where $\gamma_{\lambda c}$ is the amplitude of the reduced width in the channel with a set of quantum numbers C, $E_\lambda$ the resonance energy and $\bar{\Gamma}_\gamma$ the average radiative width.

The quantity $\beta_{\lambda c} = \gamma_{\lambda c}/\bar{\gamma}_c$ obeys normal distribution (which corresponds to the Porter-Thomas distribution for the respective reaction widths). We also introduce the quantity $\bar{D}$, which is the average distance between the resonances of the fixed state. In calculations it is convenient to use a K-matrix containing the partial widths $\bar{\Gamma}_{\lambda c}$:

$$K_{cc'} = \frac{\bar{\Gamma}^{1/2}_{\lambda c}\,\bar{\Gamma}^{1/2}_{\lambda c'}}{2\bar{D}} \sum \frac{\beta_{\lambda c}\,\beta_{\lambda c'}}{\left[(E_\lambda - E)/\bar{D}\right] - (i\bar{\Gamma}_\gamma/2D)} \quad \text{(B)}$$

The neutron cross-sections are expressed in terms of the S-matrix

$$S_{cc'} = exp(-2i\varphi_\ell)(1 + iK_{cc'})/(1 - iK_{cc'});$$

$$\sigma_t = 2\pi\lambda^2 \sum_{J,\pi} g(J) \sum_{\ell,j} (1 - Re\, S^{J\pi}_{n\ell j,\,n\ell j});$$

$$\sigma_f = \pi\lambda^2 \sum_{J,\pi} g(J) \sum_{\ell,j} |S_{n\ell j,\,f\ell j}|^2; \quad \text{(C)}$$

$$\sigma_e = \pi\lambda^2 \sum_{J,\pi} g(J) \sum_{\ell,j} |1 - S_{n\ell j,\,n\ell j}|^2 .$$

The radiative-capture cross-section was determined in a separate calculation using the single-level Breit-Wigner formalism. In accordance with the recommendations of evaluators in Ref. [6], we took the (n,γf) process into account. In order to maintain accurately the balance of average neutron cross-sections, we determined the elastic scattering cross-section $\sigma_e$ as the difference between the total cross-section and the sum of the fission and radiative capture cross-sections.

In the calculations we took into account the contributions of the s- and p-neutrons to the neutron cross-sections. This was legitimate for the neutron energy range considered $E_n < 21$ keV. The parameters $\Gamma_n$ and $\Phi_\ell$ dependent on $\ell$ are expressed normally as:

$$\varphi_0 = kR_0; \quad \varphi_1 = kR_1 - arctg(kR_1); \quad \Gamma_n(\ell=0) = \Gamma^0_{n_0}\sqrt{E}\,v_0; \quad \Gamma_n(\ell=1) = \Gamma^0_{n_1}\sqrt{E}\,v_1, \quad \text{(D)}$$

where $v_0$, $v_1$ are transmission coefficients for s- and p-neutrons, while $R_0$, $R_1$ are the scattering radii of s- and p-neutrons, which were assumed to be identical.

The novelty of the method developed for calculating the average functionals (transmission functions, moments of cross-sections, including average cross-sections and, lastly, resonance self-shielding factors) lies in the concept of stochastic modelling of the resonance structure of the neutron cross-sections for fissile nuclei on the basis of the multi-level formalism described here. In the early studies there was similar modelling within the framework of simpler models allowing the application of the method of Ψ-, X- functions (for example, the two-level Breit-Wigner formula with approximate consideration of their resonance interference [7]). The validity of such approximations for fissile nuclei of the $^{235}U$ and $^{239}Pu$ type is doubtful. Instead of the modelling method, one can also use numerical integration of multiple integrals over the statistical distributions of the nuclear levels (distances and widths) for each system of levels (for example, as in Ref. [8]). However, this is done within the framework of the same approximate formalisms; moreover, there arise problems of optimum selection of points of the calculation grid and quadrature formulae (as a function of the functional) and also the problem of the method of convolution of integrals for different level systems (when we go over to the functionals from the observed cross-sections).

In the present approach these difficulties are overcome by applying the method of statistical tests in the form of multiple neutron "emissions" at the points on a uniform lethargic grid (usually 200 measurements at one point) with a step $\Delta u$ (usually $u$ = 20 meV) over a wide interval $\Delta U$ (usually $\Delta U$ = 100 eV). In each test the Doppler energy shift is randomly selected (for several temperatures at once). One run denotes a set of statistics for random tests over the whole interval $\Delta U$, in which the random realization of neutron cross-sections with a set of resonances of all the required states is determined. This picture is built up from a set of random numbers from the "generators" of position and level width of the given system in the 10-level approximation, with random selection of the sign of the pair interference. The formulae for cross-sections are expressions using the inverted K-matrix (in this case, of the third rank - according to the number of channels: one neutron channel and two fission channels) for 10 interacting random resonances of the given system. Therefore, the method described here can be called the stochastic K-matrix method. Realization of the sequence of the resonances in the wide interval $\Delta U$ is performed by successive addition of a new random resonance on the right with simultaneous elimination of resonances on the extreme left. The whole of the information on this realization is retained in the computer's operational memory.

The test procedure is as follows. The neutron "emitted" from point u' is moved as a result of the Doppler shift to position u", where the values of cross-sections for the given realization are accurately determined. To speed up the counting, the Doppler shift is randomly selected on the given grid consisting of points with the step $\Delta u$ (not more than 20 meV). The tests are carried within the interval $\Delta U$ with the addition of five Doppler widths at the ends. In this manner, in one typical run we perform 200 measurements at each of $5 \times 10^3$ points approximately, and this ensures good averaging over the Doppler function (error not exceeding 1% for the cross-section moments) and over the selection of resonances for $\Delta U$ = 100 eV. As information accumulators we used the evaluations of the distribution functions for the total cross-section $P(\sigma_t)$ and partial cross-sections $\sigma_x(\sigma_t)$ with division of the variation range of $\sigma_t$ into a large number of intervals (250 or 500), which are uniform on the logarithmic scale. This enables us to find an evaluation for the arbitrary functional $F(\sigma_t, \sigma_x)$: $\langle F \rangle = \int_0^\infty F(\sigma_t, \sigma_x) P(\sigma_t) d\sigma_t$ with evaluation of the dispersion $\mathcal{D}_F = \langle F^2 \rangle - \langle F \rangle^2$.

Obviously, any functional can also be evaluated directly, without the operation of convolution in terms of function $P(\sigma_t)$. However, storage of information in the form of $P(\sigma_t)$, $\sigma_x(\sigma_t)$ is convenient in practice, since it subsequently makes possible accurate averaging of the functionals over the isotope mixture (the problem of preparing macroscopic constants). As we know, this problem has so far been solved in the approximate formalism of the dilution cross-section $\sigma_0$, which has no mathematical justification.

In the version described above, the BEhSM-6 computer time spent on one run is about 5 min. For better averaging over the resonance statistics, we have to carry out several runs, changing the resonance selection (realization) each time. Evidently, five runs correspond to the averaging of functionals over the statistics for the resonances contained in the 0.5 keV energy interval; this is quite sufficient for the analysis of the transmission functions and the average cross-sections in the unresolved resonance region in the case of heavy nuclei.

Before we used to make a preliminary analysis of experimental data on transmission and fission self-indication for $^{239}Pu$. In Ref. [9] the processing was carried out by the sub-group method for evaluation of the resonance self-shielding factors (area method). In Refs [10, 11] we made an attempt to interpret experimental data by the stochastic K-matrix method, but with the use of the procedure by which the Doppler broadening is calculated by numerical integration. A more careful analysis showed that such a procedure did not have sufficient accuracy. Hence the present study is a valid extension of the Monte Carlo method for correct consideration of the Doppler effect in addition.

## Analysis of experiments and results

At first, we calculated the transmission functions using the parameters recommended in Ref. [6], with averaging equivalent to averaging over the energy groups of the Bondarenko-Nikolaev-Abagyan-Bazazyants (BNAB) system of constants [1]. The averaging function was made uniform on the lethargy scale and the sensitivity coefficients were calculated with respect to the calculation parameters. The main calculation parameters were: scattering radius R' = 9.075 fm, observed distance between the s-resonances 2.38 eV, mean radiation width $\bar{\Gamma}_\gamma$ = 40.7 meV, p-strength function $S_1 = 2.00 \times 10^{-4}$, fission width $\bar{\Gamma}_f$ and s-neutron strength function $S_o$ dependent on the group number, $S_o$ on an average decreasing from the value of 1.13 in the 15th group (0.465-1 keV) to 0.86 in the 11th group (10-21.5 keV). The deviations of experiment from calculation (see dashed curves in Fig. 1) were significant at small energies. In the region of $E_n$>5 keV the agreement with experiment was more satisfactory. It was discovered that the transmission function was most sensitive to the scattering radius R' (in parameters common to the s- and p-neutrons). Trial minimization on the basis of the statistical algorithm [12] showed the need, first of all, to vary the scattering radius and strength functions, and also to make some change in the distance $\bar{D}$. For the average fission-reaction and radiative-capture cross-sections to remain unchanged simultaneously, the parameters $\bar{\Gamma}_\gamma$ and $\bar{\Gamma}_f$ had also to be small. The latter parameters have no appreciable influence on T(n) and $T_f(n)$. It was concluded that the main "dominant" parameters in the fitting of the experimental data were the scattering radius R' ("strongest" parameter) and the neutron strength functions $S_o$ and $S_1$, whose influence depends on neutron energy. Consequently, the greatest refinement is attained with the parameter R'. For the T(n) and $T_f(n)$ measurement error varying from 2-4% (for small sample thicknesses) to 10-20% (for large thicknesses, see Fig. 1), the a posteriori error in R' is of the order of 2.5%. The errors indicate a 95% confidence interval (2σ).

We also carried out calculations by the optical model with a non-spherical potential [13], using the parameters recommended in Ref. [14] on the basis of an analysis of extensive experimental material on neutron cross-sections for heavy nuclei. These calculations show that for neutron energies between 0.5 and 20 keV the neutron strength functions depend hardly at all on the neutron energy, while the variation in the scattering radius is quite appreciable (4-5%). When considering these results we came to the conclusion that a different strategy was desirable for optimizing experimental data - variation of the scattering radius, which is a function of the neutron energy and behaves in approximately the same way as in the optical model, but at constant optimum values of the neutron strength functions. At the same time, the requirement was imposed that the recommended group-averaged neutron cross-sections should remain within 10%. Since the energy group width is

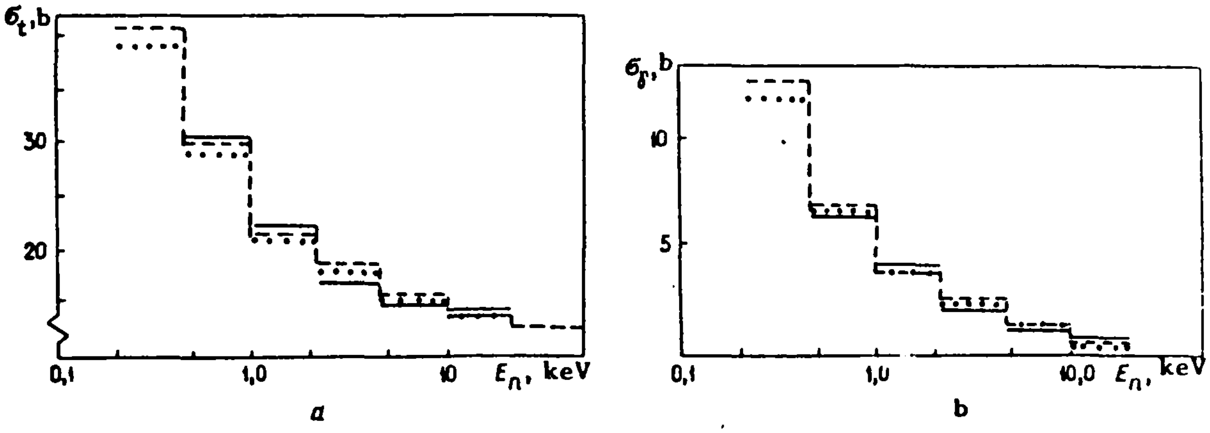


**Fig. 2. Group cross-sections $\sigma_t$ (a) and $\sigma_\gamma$ (b) for $^{239}Pu$: —— - calculation using the optimized parameters; —— - ENDF-B-V data [15]; .... - BNAB-78 data [1].**

sufficient for good averaging over the resonance statistics, it was hoped (and the hope was in fact realized) that, unlike the case of averaging in the narrower groups, here local changes would not be required in the neutron strength functions in order to describe the total group cross-sections and the radiative-capture cross-sections.

In Fig. 1 the solid curves represent the calculation results using the recommended variant for optimization, with the scattering radius varying monotonically as a function of neutron energy from 9.5 fm, in the 15th group, to 9.0 fm, in the 11th group. Here the neutron strength function values are $S_o = 1.10$ and $S_1 = 1.82$, the observed distance between the s-resonances is $\bar{D} = 2.11$ eV, $\bar{\Gamma}_\gamma = 36$ meV, and the fission widths vary. It should be pointed out, however, that the 10% variation in the latter two parameters does not substantially affect the calculation results. Our analysis shows that the problem of optimizing the optical model potential parameters is best formulated on the basis of experimental material on neutron cross-sections, supplemented by experimental data on transmission functions. The incorporation of reaction cross-section data also calls for the use of the statistical neutron cross-section model.

Apart from evaluation of the average resonance parameters by an analysis of the transmission function data, we sought in the present study to derive the group constants (average cross-sections and their resonance self-shielding factors). Figure 2 shows the results of our statistical calculation of the total group cross-sections and the radiative-capture cross-sections on the basis of the optimized set of average resonance parameters. Comparison with the BNAB-78 data in Ref. [1] and ENDF/B-V data in Ref. [15] indicates reasonable agreement between the different evaluations. There is, however, a substantial discrepancy between our

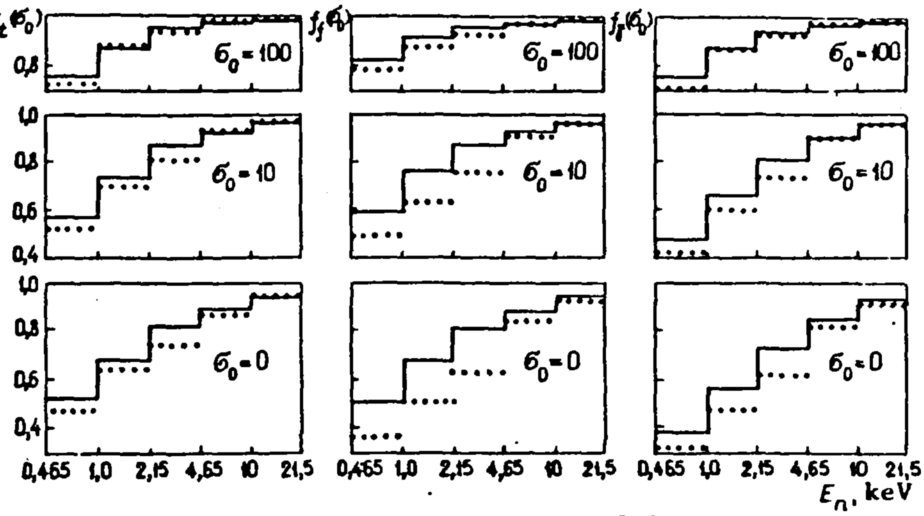


**Fig. 3. Resonance self-shielding factors for $^{239}$Pu neutron cross-sections for different dilution cross-sections $\sigma_0$, b: —— - calculation using the optimized parameters; .... - BNAB-78 data [1].**

evaluations of the resonance self-shielding factors and those of Ref. [1] (Fig. 3). Our own evaluations show a weaker resonance self-shielding effect, at least in the unresolved resonance region $E_n < 5$ keV. For larger dilution cross-sections ($\sigma_0 > 100$), as was to be expected, the differences between the various evaluations of the resonance self-shielding factors become small. The fact that the above differences exist means that the theoretical calculation models and the evaluation parameters contained in them should be studied further. Therefore it is still essential, in our opinion, to carry out new experiments to determine neutron transmission functions in the unresolved resonance region for fissile nuclei.

Our study yielded new evaluations of the group constants for $^{239}$Pu in the unresolved resonance region as a result of the analysis of neutron data, especially the data on transmission functions. A noteworthy feature of the analysis is that a new improved theoretical calculation model has been developed and used to interpret experimental data.

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