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Reliability of Photonuclear Data: Various Experiments and Evaluations

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Abstract—The well-known problem of reliability of partial photoneutron cross-section data obtained using beams of quasimonoenergetic annihilation photons was discussed. Noticeable disagreements between data from various experiments were analyzed using physical data reliability criteria. The experimentally-theoretical method for evaluating of reaction cross sections satisfied physical criteria was used and new reliable data were obtained for many nuclei. The disagreements between newly evaluated cross sections and data from the IAEA Digital Photonuclear Data Library were discussed.

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INTRODUCTION

Cross sections of partial photoneutron reactions with different numbers of outgoing particles, primarily (γ, 1*n*), (γ, 2*n*), and (γ, 3*n*), are widely used in both fundamental and applied research in various branches of science, such as nuclear physics, astrophysics, geology, chemistry, medicine, etc. The majority of those data was obtained using quasimonoenergetic annihilation photon beams and the method of photoneutron multiplicity sorting at Livermore (USA) and Saclay (France) [1–4].

The significant systematic data disagreements between data of both laboratories for partial reaction cross sections were obtained for 19 nuclei (^{51}V, ^{75}As, ^{89}Y, ^{90}Zr, ^{115}In, ^{116,117,118,120,124}Sn, ^{127}I, ^{133}Cs, ^{159}Tb, ^{165}Ho, ^{181}Ta, ^{197}Au, ^{208}Pb, ^{232}Th, ^{238}U) [5–9]. It was found that although the disagreements between photoneutron yield cross sections,

$$\sigma(\gamma,\mathrm{S}n) = \sigma(\gamma,1n) + 2\sigma(\gamma,2n) + 3\sigma(\gamma,3n) + \ldots \quad (1)$$

obtained in various laboratories are about 10%, as a rule the (γ, 1*n*) reaction cross sections are larger at Saclay, but the (γ, 2*n*) cross sections vice versa are larger at Livermore (up to 100%). The average ratio of integrated cross sections for Saclay data to those for Livermore data $\sigma_\mathrm{S}^\mathrm{int}/\sigma_\mathrm{L}^\mathrm{int}$ is equal to 1.07 in case of (γ, 1*n*) reaction but 0.84 in case of (γ, 2*n*) reaction. It means that there are noticeable systematic uncertainties in partial reaction cross sections and therefore nobody knows which data are reliable or not. The correspondent systematics is presented in Fig. 1.

Because of that the International Atomic Energy Agency Coordinated Research Project [10] was realized with the aim to evaluate the partial and total photoneutron reaction cross sections which are free from the problems of experimental neutron multiplicity sorting. Evaluations were carried out for 164 isotopes of 48 elements from ^{2}H to ^{241}Pu using various nuclear modeling codes. All data were included into the digital IAEA Photonuclear Data Library (PDL).

But when the new experimental-theoretical method for evaluating the partial reaction cross sections on the base of using objective physical criteria of

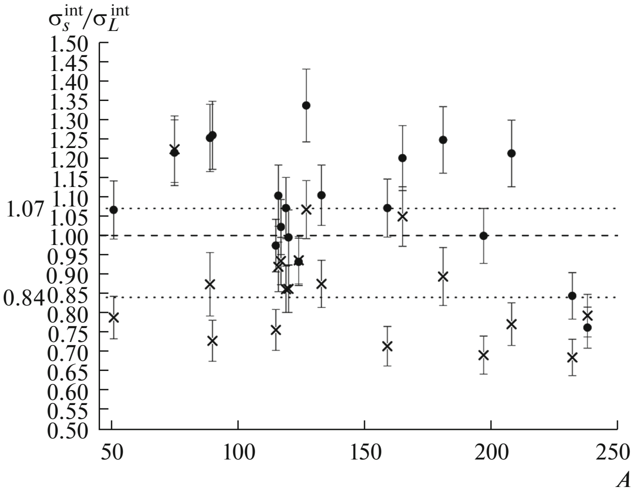

Fig. 1. The systematics of the values of the ratios $\sigma_\mathrm{S}^\mathrm{int}(\gamma, 1n)/\sigma_\mathrm{L}^\mathrm{int}(\gamma, 1n)$ (circles) and $\sigma_\mathrm{S}^\mathrm{int}(\gamma, 2n)/\sigma_\mathrm{L}^\mathrm{int}(\gamma, 2n)$ (crosses) obtained using Saclay and Livermore experiments data.

data reliability was proposed [11] it was found that many newly evaluated data are noticeably disagree not only with experimental data obtained using the method of photoneutron multiplicity sorting but with previously evaluated data included into the PDL.

It meant that the IAEA PDL needs to be revised and updated. Therefore the new special IAEA Coordinated Research Program "Updating the Photonuclear Data Library and generating a reference database for Photon Strength Functions" was adopted for period 2016–2019 [12].

1. THE PROBLEM OF PARTIAL PHOTONUCLEAR REACTION CROSS SECTIONS RELIABILITY

The experimental-theoretical method for evaluating the partial reaction cross sections was developed [11] in order to resolve the problems of significant disagreements between results of various experiments. Its main idea is to decompose the experimental neutron yield cross-section (1) rather independent from the neutron multiplicity sorting problem into the partial reaction cross sections using the ratios,

$$\begin{aligned} F_i^{\text{theor}} &= \sigma^{\text{theor}}(\gamma, in)/\left[\sigma^{\text{theor}}(\gamma, 1n) + \sigma(\gamma, 1n1p)\right. \\ &\quad \left. + 2\sigma^{\text{theor}}(\gamma, 2n) + 3\sigma^{\text{theor}}(\gamma, 3n) + \ldots\right], \end{aligned} \tag{2}$$

calculated within the framework of the combined photonucleon reaction model (CPNRM) [13, 14] for the partial reactions (γ, *in*) by the way,

$$\begin{aligned} \sigma^{\text{eval}}(\gamma, in) &= F_i^{\text{theor}} \sigma^{\text{exp}}(\gamma, \mathrm{S}n) \\ &= \left[\sigma^{\text{theor}}(\gamma, in)/\sigma^{\text{theor}}(\gamma, \mathrm{S}n)\right] \sigma^{\text{exp}}(\gamma, \mathrm{S}n). \end{aligned} \tag{3}$$

The CPNRM is based on the statistical approach and uses a combination of the preequilibrium exciton model and particle evaporation process to calculate probabilities of formation of specific final nuclei after absorption of a photon. It takes into account the deformation of nucleus and isospin splitting of its giant dipole resonance and gives to one the opportunity for description and is well tested for many medium and heavy nuclei.

According to the definitions (2) $F_1 > 1.00$, $F_2 > 0.50$, $F_3 > 0.33$, etc never can be. Larger F_i^{exp} values mean that partial reaction cross sections obtained are not reliable because experimental sorting of neutrons between those reactions was carried out with large systematic uncertainties. Therefore ratios F_{exp} of definite partial reaction cross sections to that of neutron yield reaction were proposed [11] as objective physical criteria partial of reliability of photoneutron reaction cross-section experimental data.

Additionally it should be pointed out that ratios F_{exp} include only the cross-section terms and therefore should be definitely positive.

The newly evaluated partial photoneutron reaction cross sections for ^{181}Ta [15], ^{197}Au [11] and ^{209}Bi [16] were compared [17–19] with the results of measurements of reaction yields using bremsstrahlung beams and activation method. In this method of various partial reactions separation alternative to the method of neutron multiplicity sorting, the direct identification of each partial reaction is based on the final nuclei. It was concluded that evaluated partial photoneutron reaction cross sections really are reliable because they agree with data obtained using activation method although contradict to data obtained using neutron multiplicity sorting method. Therefore it was concluded that if F_i^{exp} noticeably differ from F_i^{theor} one has definite doubts in experimental data reliability.

So there are three following partial photoneutron reaction cross-section data reliability criteria:

(1) the ratios F_i^{exp} must not be larger than the upper limits mentioned above;

(2) $\sigma^{\text{exp}}(\gamma,\ \text{in})$ and correspondingly F_i^{exp} must not be negative;

(3) the differences between F_i^{exp} and F_i^{theor} must not be noticeable.

For many nuclei (^{63,65}Cu, ^{80}Se, ^{91,94}Zr, ^{115}In, $^{112–124}$Sn, ^{133}Cs, ^{138}Ba, ^{159}Tb, ^{181}Ta, $^{186–192}$Os, ^{197}Au, ^{208}Pb, ^{209}Bi and some others) it was shown [8, 9, 11, 15–25] that in many cases the experimental partial reaction cross sections do not satisfy the proposed data reliability criteria because there are many negative cross-section (and correspondingly F_i^{exp}) values and/or F_i^{exp} values larger than top limits mentioned above and/or large differences among and F_i^{theor}).

In addition to the examples presented in previous review [24] the typical results of data do not satisfied physical reliability criteria are presented in Fig. 2 for ^{94}Zr and ^{116}Sn. One can see that in wide energy ranges there are many forbidden negative values and values exceeded upper limits mentioned above. At the same time energy dependencies of all F_i^{exp} are noticeably different from F_i^{theor}.

It was found [8, 9, 11, 15–26] that the cross sections obtained using the experimental-theoretical method for evaluating the partial reaction cross sections [10] in many cases are noticeably different from those obtained in experiments using neutron multiplicity sorting method. Again in addition to the examples presented before [24] the typical results of noticeable disagreements between evaluated and data from various experiments are presented in Fig. 3 for ^{59}Co and ^{208}Pb.

It was shown [8, 9, 11, 15–26] that the main reason of noticeable disagreements between the partial reaction cross sections obtained at Livermore and Saclay is

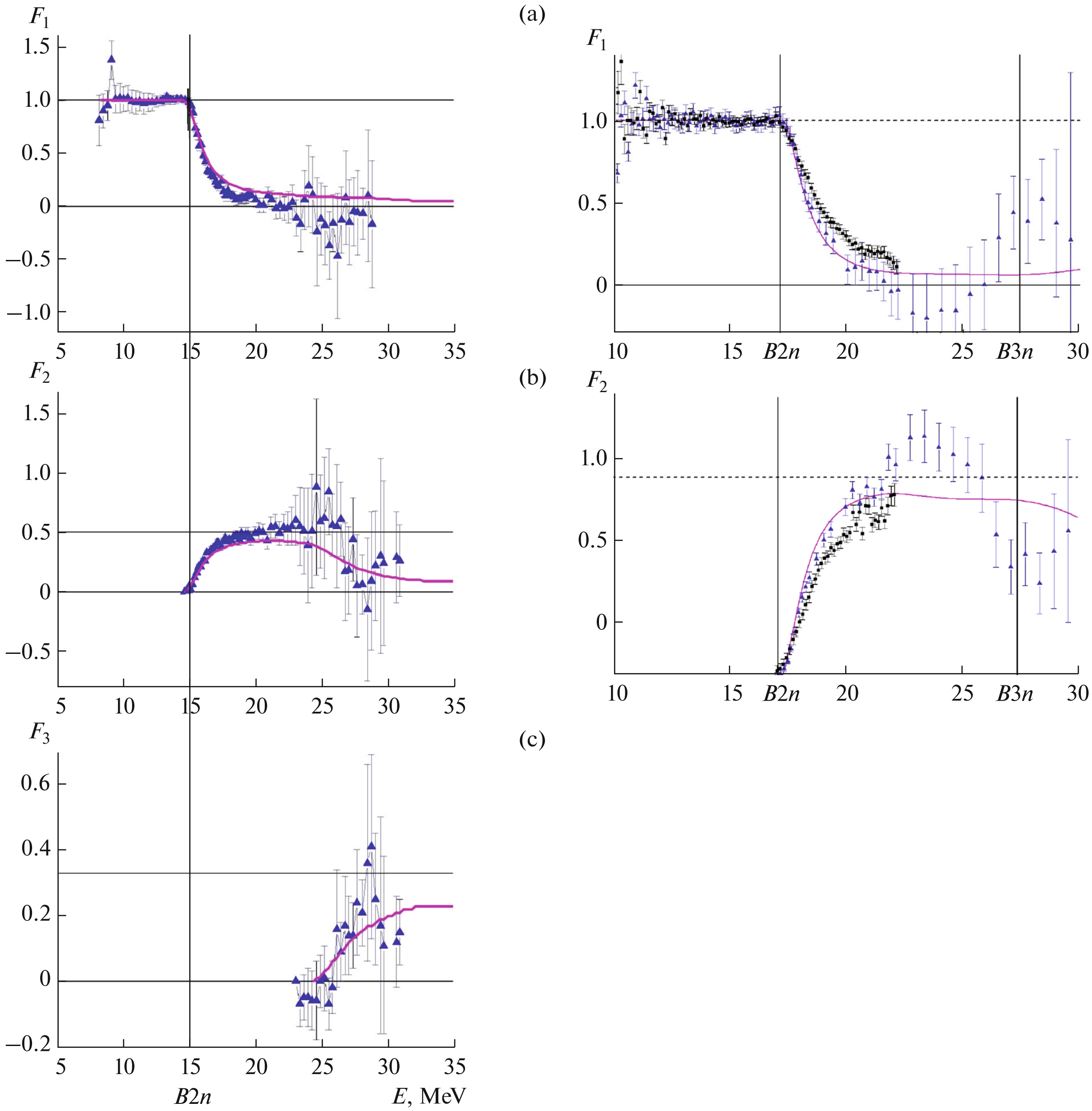

Fig. 2. Comparison of ratios F_i^{exp} ((a) $i = 1$, (b) $i = 2$, (c) $i = 3$) obtained for results of various experiments with those F_i^{theor} calculated in combined model ([13, 14]—lines). Left: ^{94}Zr (data obtained [21] for Livermore data [27], triangles), right: ^{116}Sn (data obtained [28] for Livermore data [29], triangles and Saclay data [30], squares).

the difference of procedures used to separate counts into 1*n* and 2*n* events. The large systematic uncertainties of the procedure of determination neutron multiplicity on the base of measurement its energy resulted in unreliable (erroneous) transmission of many neutrons from one partial reaction to another. One can see those unreliable transmissions in Fig. 4 where the examples of differences,

$$\Delta\sigma = \sigma^{\text{eval}} - \sigma^{\text{exp}}, \qquad (4)$$

between the evaluated and the experimental cross sections obtained separately for both partial reactions $(\gamma, 1n)$ and $(\gamma, 2n)$ are presented for ^{40}Zr and ^{188}Os. The unreliable distortions of each partial reaction cross-section have values about tens mb.

Because of noticeable disagreements between new data evaluated using objective physical criteria and experimental data obtained using the method of photoneutron multiplicity sorting and included into the international database [1] the competition of those

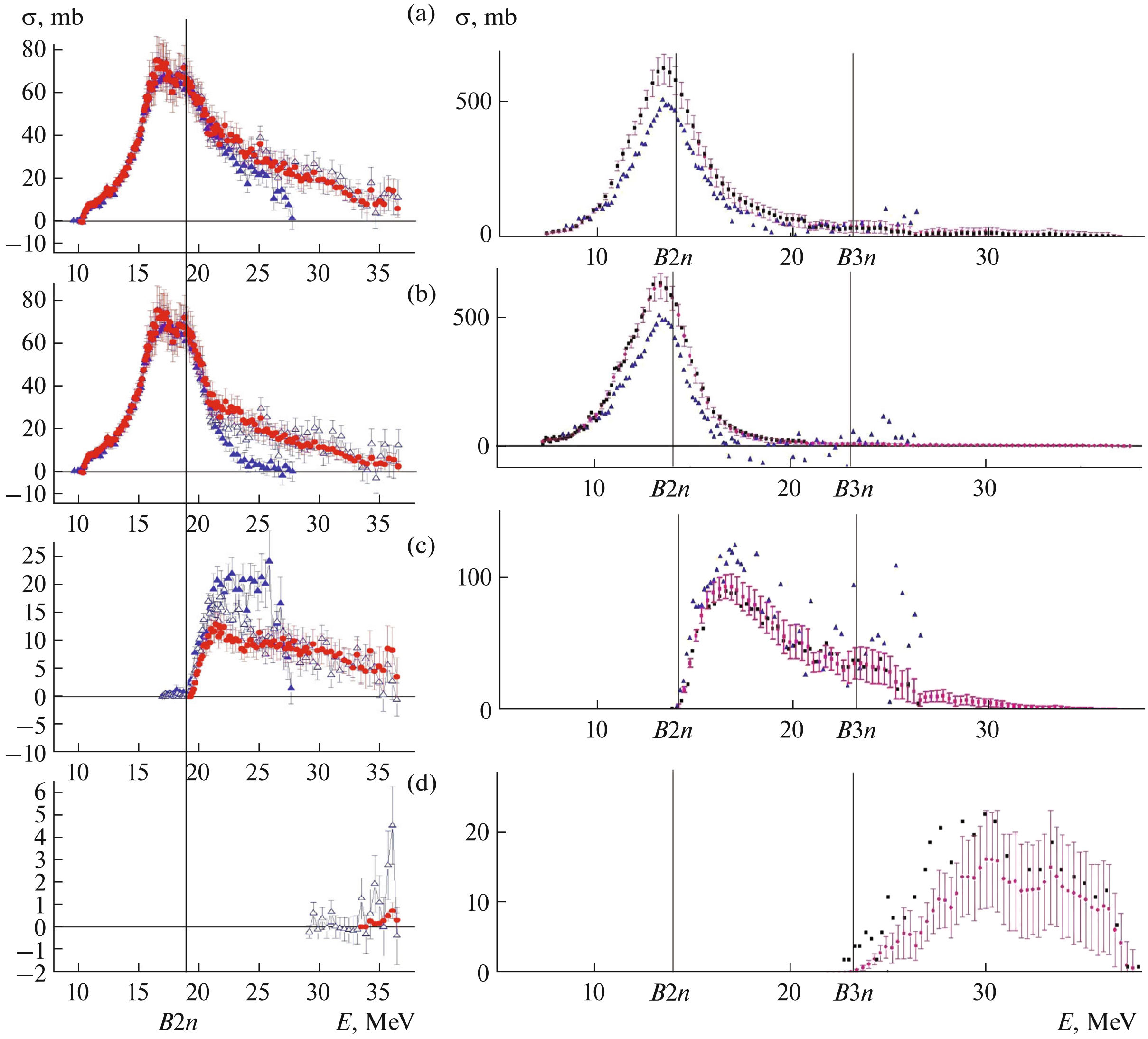

Fig. 3. Comparison of evaluated (circles) and experimental photonuclear reaction cross sections: (a) σ(γ, tot); (b) σ(γ, 1*n*), (c) σ(γ, 2*n*); (d) σ(γ, 3*n*). Left—^{59}Co ([31]—filled triangles and [32]—open triangles), right—^{208}Pb ([33]—triangles, [34]—squares).

new data with previously evaluated data from the IAEA PDL [10] is of interest.

3. COMPARISON OF NEWLY EVALUATED PARTIAL PHOTONEUTRON REACTION CROSS SECTIONS WITH THOSE FROM THE IAEA PDL

As was mentioned above the new evaluated partial photonuclear reaction cross sections were obtained using together experimental neutron yield cross sections σ(γ, *Sn*) (1) and F_i functions (2) calculated in the frame of CPNRM. The σ(γ, *Sn*) is rather independent on experimental neutron multiplicity sorting problems because it includes all outgoing neutrons.

At the same time old evaluated data [10] were obtained using various computer codes (GNASH, Los Alamos (USA), ALICE-F and MCPHOTO, Tokai (Japan), GUNF and GLUNF, Beijing China), XCFISS, Obninsk (Russia) based on using not neutron yield cross sections σ(γ, *Sn*) (1) but total photoneutron cross sections,

$$\sigma(\gamma,\mathrm{tot}) = \sigma(\gamma,1n) + \sigma(\gamma,2n) + \sigma(\gamma,3n) + \ldots \quad (5)$$

In many cases old evaluations were carried out in order to model accurately σ(γ, tot) data obtained at Saclay.

For energies up to the threshold *B*3*n* of reaction (γ, 3*n*),

$$\sigma(\gamma,\mathrm{tot}) = \sigma(\gamma,Sn) - \sigma(\gamma,2n), \quad (6)$$

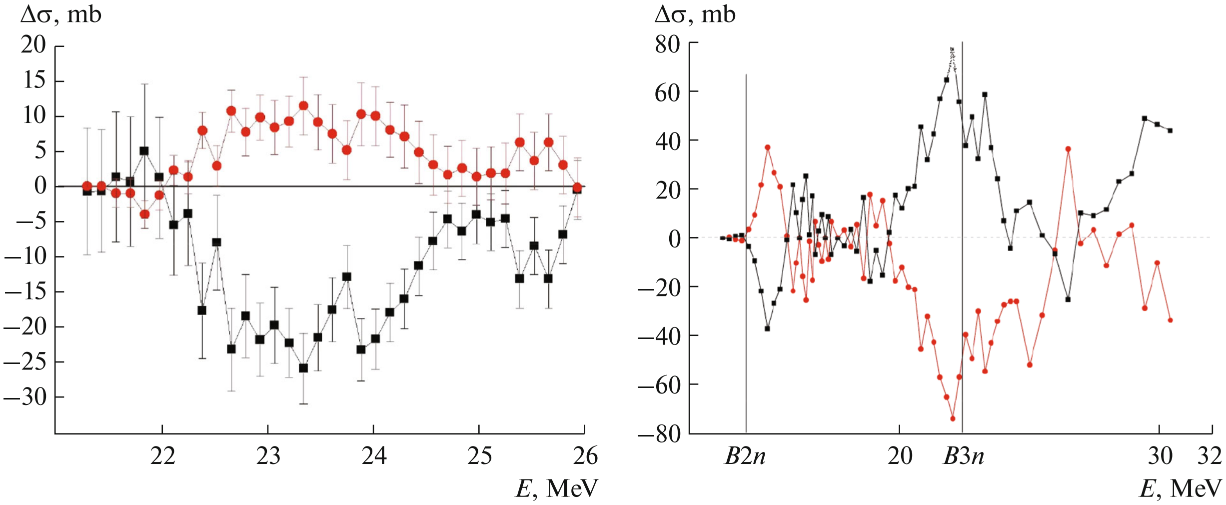

Fig. 4. Comparison of the differences Δσ (4) for between the experimental and the evaluated data for (γ, 1*n*) reaction ($\Delta\sigma_1$, circles) and (γ, 2*n*) reaction ($\Delta\sigma_2$, squares). Left—^{90}Zr [25], right ^{188}Os [26].

and therefore possible mistakes in determination σ(γ, 2*n*) could be the reason of mistakes in σ(γ, tot).

As was mentioned above in many cases experimental cross sections σ(γ, 2*n*) are underestimated at Saclay and vice versa overestimated in Livermore. It was shown that unreliable (erroneous) transportation of many neutrons from one reaction to another is dependent on photon energy and therefore the competition between experimental and evaluated cross sections as between various evaluated cross sections could be dependent on photon energy.

The comparison of newly evaluated cross sections with data evaluated before [10] was carried out in detail for three nuclei—^{91}Zr, ^{159}Tb, and ^{197}Au [36]. It was found that for all partial reaction cross sections there are noticeable disagreements between old and new evaluated data. The results of such comparison for (γ, 2*n*) reaction cross sections are presented in Fig. 5. One can see very clear disagreements dependent on the energy of photons. In the case of ^{91}Zr those disagreements are at all investigated energies. In the cases of ^{159}Tb and ^{197}Au the results of new and all evaluations are in serious disagreements at low energies up to about 20 MeV and are close to each other at higher energies.

Because of that it became evident that the data included into the digital IAEA Photonuclear Data Library needs to be revised and updated.

4. THE IAEA COORDINATED RESEARCH PROGRAM

To solve the task of revision and updating those data included into the IAEA PDL the new special international Coordinated Research Program F41032 “Updating the Photonuclear Data Library and generating a reference database for Photon Strength Functions” was adopted for period 2016–2019 [12]. The part of that is the MSU SINP Centre for Photonuclear

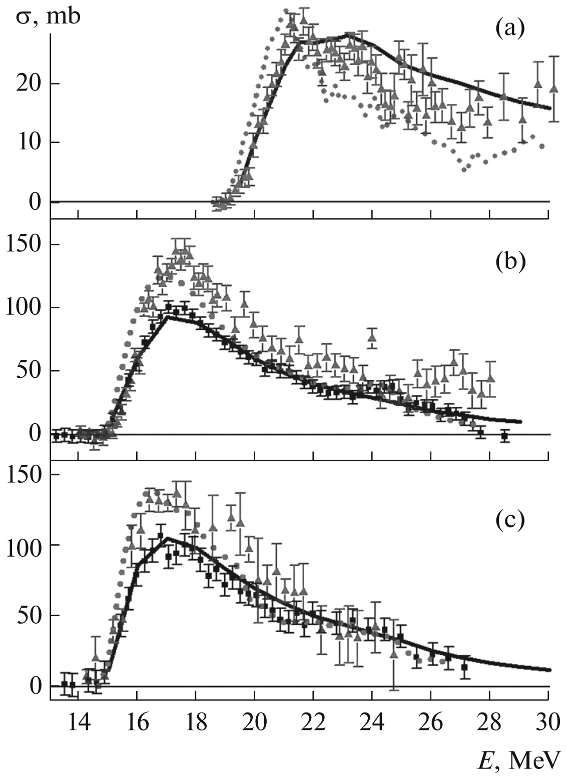

Fig. 5. Comparison of the new ([36, circles) and old ([10], solid lines) evaluated (γ, 2*n*) reaction cross sections with experimental data: (a) ^{91}Zr ([27]—triangles), (b) ^{159}Tb ([37]—triangles, [38]—squares), (c) ^{197}Au ([39]—triangles, [34]—squares).

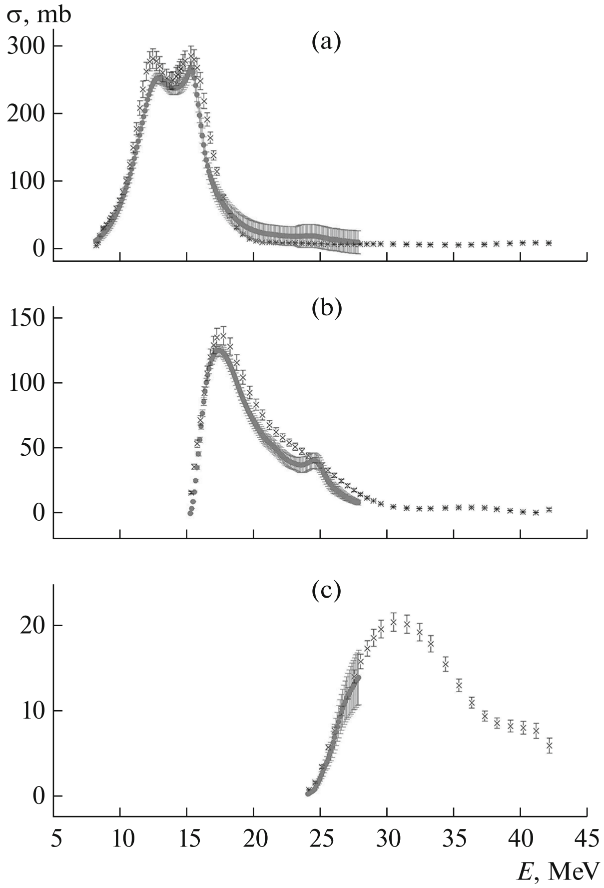

Fig. 6. Comparison of the new experimental (crosses) and evaluated ([8], circles) cross sections for ^{159}Tb: (a) (γ, 1*n*) reaction, (b) (γ, 2*n*) reaction, (c) (γ, 3*n*) reaction.

Experiments Data Research Contract 20501 “Evaluation of Partial and Total Photoneutron Reactions Cross Sections Using New Objective Physical Data Reliability Criteria”. The main tasks of the CDFE Contract are the new evaluations of cross sections for partial photoneutron reactions (γ, 1*n*), (γ, 2*n*), and (γ, 3*n*) and also the total photoneutron reaction using objective physical criteria for isotopes of many elements (Se, Y, Rh, In, Ho, Ba, La, Ce, Os, etc.).

It is evident that new evaluated data must be compared with the experimental data obtained using the methods alternative to the photoneutron multiplicity sorting method. In addition to the results obtained using bremsstrahlung and activation method mentioned above [17–19] it could be the results obtained using quasi-monochromatic laser Compton-scattering (LCS) γ-ray beams and the novel technique of direct neutron-multiplicity sorting with a flat-efficiency detector [40]. A flat response neutron detector with the detection efficiency of 36.5 ± 1.6% over a neutron energy range 0.01–5.00 MeV was developed by optimizing triple-ring configurations of 3He proportional counters embedded in a polyethylene moderator block. Till now photoneutron cross-section measurements were performed for (γ, *xn*) reactions with $x = 1–4$ using LCS γ-ray beams at the NewSUBARU synchrotron radiation facility for several nuclei.

In Fig. 6 the new experimental data obtained for ^{159}Tb are compared with data evaluated using physical data reliability criteria and the experimental-theoretical method described above. One can see that for three partial reactions, (γ, 1*n*), (γ, 2*n*), and (γ, 3*n*), evaluated and new experimental data in good agreement to each other.

5. SUMMARY AND CONCLUSIONS

The experimental data for (γ, 1*n*), (γ, 2*n*) and (γ, 3*n*) reactions for many nuclei (^{63,65}Cu, ^{75}As, ^{80}Se, ^{89}Y, $^{90\text{-}94}$Zr, ^{115}In, $^{112-124}$Sn, ^{133}Cs, ^{138}Ba, ^{139}La, ^{140,142}Ce, ^{141}Pr, ^{145,148}Nd, ^{153}Eu, ^{159}Tb, ^{181}Ta, ^{181}W, $^{186-192}$Os, ^{197}Au, ^{208}Pb, ^{209}Bi and some others) obtained using quasimonoenergetic annihilation photons were analyzed using the objective physical criteria of data reliability. It was found out that many data are not reliable because they are not satisfied the proposed objective physical criteria.

Using the experimental-theoretical method for evaluating of data satisfied reliability criteria new partial and total photoneutron cross section data were obtained. It was shown that the evaluated cross sections contradict to the results of experiments used neutron multiplicity sorting method in experiments with quasimonoenergetic annihilation photons. New evaluated data agree with the data obtained using alternative methods, such as activation method on bremsstrahlung beams and quasi-monochromatic laser Compton-scattering (LCS) γ-ray beams and the novel technique of direct neutron-multiplicity sorting with a flat-efficiency detector.

At the same time there are noticeable disagreements between new data evaluated using physical data reliability criteria and data evaluated before without using such objective criteria and included into the IAEA digital Photonuclear Data Library.

Because of that for many nuclei mentioned above new partial and total photonuclear reaction cross sections were evaluated and are now in detail comparison with previously evaluated data the results of new experiments carried out using the methods alternative the method og photoneutron multiplicity sorting.

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