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The reliability of photoneutron cross sections for ^{90,91,92,94}Zr

V.V. Varlamov[1,a], A.I. Davydov[2], B.S. Ishkhanov[1,2], and V.N. Orlin[1]

[1] Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, 119991 Moscow, Russia
[2] Physics Faculty, Lomonosov Moscow State University, 119991 Moscow, Russia

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Abstract. Data on partial photoneutron reaction cross sections $(\gamma, 1n)$ and $(\gamma, 2n)$ for ^{90,91,92,94}Zr obtained at Livermore (USA) and for ^{90}Zr obtained at Saclay (France) were analyzed. Experimental data were obtained using quasimonoenergetic photon beams from the annihilation in flight of relativistic positrons. The method of photoneutron multiplicity sorting based on the neutron energy measuring was used to separate partial reactions. The research carried out is based on the objective of using the physical criteria of data reliability. The large systematic uncertainties were found in partial cross sections, since they do not satisfy those criteria. To obtain the reliable cross sections of the partial $(\gamma, 1n)$ and $(\gamma, 2n)$ and total $(\gamma, 1n)$ + $(\gamma, 2n)$ reactions on ^{90,91,92,94}Zr and $(\gamma, 3n)$ reaction on ^{94}Zr, the experimental-theoretical method was used. It is based on the experimental data for neutron yield cross section rather independent from the neutron multiplicity and theoretical equations of the combined photonucleon reaction model (CPNRM). Newly evaluated data are compared with experimental ones. The reasons of noticeable disagreements between those are discussed.

1 Introduction

Data on cross sections for both total and partial photoneutron reactions with different numbers of outgoing particles, primarily $(\gamma, 1n)$, $(\gamma, 2n)$, and $(\gamma, 3n)$, are important for basic research mainly for energies of giant dipole resonance (GDR). At the same time those data are widely used in many applications for atomic energy, high-energy physics, safety, geology, chemistry, medicine, etc.

Experimental data on the photodisintegration of a large number of medium- and heavy-mass nuclei (such as ^{59}Co, ^{63,65}Cu, ^{80}Se, ^{115}In, $^{112-124}$Sn, ^{133}Cs, ^{138}Ba, ^{159}Tb, ^{181}Ta, $^{186-192}$Os, ^{197}Au, ^{208}Pb, ^{209}Bi) obtained using beams of quasimonoenergetic annihilation photons by the method of photoneutron multiplicity sorting at Livermore (USA) and Saclay (France) [1,2] were analyzed [3–12]. It was found that, as a rule, experimental cross sections for the $(\gamma, 1n)$, $(\gamma, 2n)$ and $(\gamma, 3n)$ partial reactions were determined with large systematic uncertainties, so that their reliability may be questioned.

The main circumstances that are indicative of the presence of such uncertainties in many photon energy ranges are the following:

i) there are many unreliable forbidden negative values in various, primarily the $(\gamma, 1n)$ reaction, cross sections;

ii) the specially introduced as objective physical data reliability criteria neutron-multiplicity transition functions, ratios

$$F_i^{\exp} = \sigma^{\exp}(\gamma, in)/\sigma^{\exp}(\gamma, Sn), \tag{1}$$

where

$$\sigma^{\exp}(\gamma, Sn) = \sigma^{\exp}(\gamma, 1n) + 2\sigma^{\exp}(\gamma, 2n) + 3\sigma^{\exp}(\gamma, 3n) + \ldots, \tag{2}$$

of definite partial reaction cross section $\sigma^{\exp}(\gamma, in)$ to that of neutron yield cross section $\sigma^{\exp}(\gamma, Sn)$, exceed the limits physically allowed for them by definition (1.0, 0.5, 0.33, ... for $i = 1, 2, 3, \ldots$) because F_1 is the ratio of $\sigma(\gamma, 1n)$ to the sum $[\sigma(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n) + \ldots]$, F_2 is the ratio of $\sigma(\gamma, 2n)$ to the sum $[\sigma(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n) + \ldots]$, etc.

It means that many experimental data do not satisfy the physical data reliability criteria.

The experimental-theoretical method for evaluating the partial reaction cross sections satisfied data reliability criteria was developed [4,5]. It is based on using the experimental neutron yield cross section $\sigma^{\exp}(\gamma, Sn)$, expressed as the sum of the partial cross sections $\sigma^{\exp}(\gamma, in)$, where i (equal to $1, 2, 3, \ldots$) denotes the neutron multiplicity, which is rather independent of the problem regarding the neutron multiplicity sorting because all detected neutrons are included. The experimental neutron yield cross

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[a] e-mail: `vvvarlamov@gmail.com`

section $\sigma^{\text{exp}}(\gamma, Sn)$ was decomposed into the partial reaction cross sections,

$$\sigma^{\text{eval}}(\gamma, in) = F_i^{\text{th}}\sigma^{\text{exp}}(\gamma, Sn) = [\sigma^{\text{th}}(\gamma, in)/\sigma^{\text{th}}(\gamma, Sn)]\sigma^{\text{exp}}(\gamma, Sn), \qquad (3)$$

using the transitional neutron multiplicity functions $F_i^{\text{th}} = \sigma^{\text{th}}(\gamma, in)/\sigma^{\text{th}}(\gamma, Sn)$, calculated within the framework of the combined photonucleon reaction model CPNRM [13–15]. This preequilibrium exciton model employs nuclear level densities calculated on the basis of the Fermi gas model and takes into account the deformation of the nucleus being considered and the isospin splitting of its giant resonance. The photoneutron yield cross sections calculated in the frame of the model are in agreement with experimental data for many medium and heavy nuclei. Such evaluation method means that competitions between partial cross sections $\sigma^{\text{eval}}(\gamma, in)$ are keeping in accordance with the theoretically calculated ratios F_i^{th} and $\sigma^{\text{eval}}(\gamma, Sn)$ is keeping close to $\sigma^{\text{exp}}(\gamma, Sn)$.

It was found that new data evaluated using experimental-theoretical method in accordance with eq. (3) disagree with data obtained neutron multiplicity sorting method but agree with the experimental results obtained by using activation method in which the identification of definite partial reaction is based not on outgoing neutrons but on final nuclei. It was found [3–12] that the main reason of the disagreements between the partial reaction cross sections mentioned above is originated from the definite shortcomings of the neutron multiplicity sorting procedures. It was shown that those individual shortcomings of methods explored at Livermore and Saclay are the reasons of well-known systematic disagreements between data obtained in the two laboratories. As a rule values of $(\gamma, 1n)$ reaction cross sections are larger at Saclay but those of $(\gamma, 2n)$ reaction vice versa at Livermore because of unreliable sorting of various energy neutrons between partial reactions.

The newly evaluated partial photoneutron reaction cross sections for ^{181}Ta, ^{197}Au, and ^{209}Bi [16–20] were compared 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 found that evaluated partial photoneutron reaction cross sections really are reliable because they agree with data obtained using activation method although they contradict data obtained using the neutron multiplicity sorting method. Therefore it was concluded that if F_i^{exp} noticeably differs from F_i^{theor} one has definite doubts in experimental data reliability. Therefore the noticeable differences between F_i^{exp} and F_i^{th}, using in the experimental-theoretical method of reliable partial reaction cross sections evaluation, was added to two circumstances indicative of the presence systematic uncertainties mentioned above.

This work is devoted to the analysis of reliability of the experimental data for the isotopes ^{90,91,92,94}Zr [21,22] in which cases the inconsistency of the experimental data to the reliability criteria (1) is quite distinct.

Isotopes of zirconium are a subject of widespread scientific interest because they are the nuclei near the closed neutron shell at $N = 50$. They illustrate the effect of adding several numbers of neutrons to the closed neutron shell. From this point of view the reliable information on partial photoneutron reaction cross sections is of great interest.

The new reliable partial $(\gamma, 1n)$, $(\gamma, 2n)$ reaction cross sections for ^{90,91,92,94}Zr and additionally $(\gamma, 3n)$ for ^{94}Zr free from the aforementioned shortcomings of experimental data were evaluated and used to obtain the reliable cross sections also for total photoneutron reaction,

$$(\gamma, \text{tot}) = (\gamma, 1n) + (\gamma, 2n) + (\gamma, 3n). \qquad (4)$$

2 Analysis of partial reaction cross sections reliability using objective physical criteria

The data for F_i^{exp}, obtained for experimental data for ^{90}Zr [21,22] and ^{91,92,94}Zr [21] under discussion are presented in figs. 1–4, respectively, together with the results of calculations F_i^{th} in the frame of CPNRM [13–15].

In fig. 1 one can see the typical disagreements between Livermore [21] and Saclay [22] data for ^{90}Zr and clear disagreements between both of them and the results of calculations in the model:

- Livermore $\sigma(\gamma, 1n)$ data are noticeably smaller in comparison of Saclay data and include physically forbidden negative values;
- Saclay $F_{1,2}^{\text{exp}}$ data do not contradict to data reliability criteria (1) but noticeably differ from the theoretical values F_i^{th};
- Livermore $F_{1,2}^{\text{exp}}$ data are close to the theoretical values F_i^{th} only for energies up to ~ 24 MeV; at higher energies there is noticeable difference between experimental and theoretical values; in the energy range $\sim$ 24–28 MeV there are clear correlation of physically forbidden negative values in $\sigma(\gamma, 1n)$ and unreliable $F_2 > 0.5$ values.

One can see in fig. 1(a) the Livermore negative F_1^{exp} values at energies $\sim$ 24–26 MeV corresponding to the negative values in the reaction $^{90}\text{Zr}(\gamma, 1n)$ [21].

It is important to point out that those negative F_1^{exp} values correlate in energy with $F_2^{\text{exp}} > 0.5$ values (fig. 1(b)). Though those negative F_1^{exp} values and $F_2^{\text{exp}} > 0.5$ values are within the limits of uncertainties one can see their definite systematic behavior. At the same time the disagreements between experimental F_i^{exp} and theoretical F_i^{th} values are significant. Therefore it can be concluded that for ^{90}Zr the Saclay data [22] at all energies and the Livermore data [21] at energies above ~ 24 MeV are not reliable.

In figs. 2–4 one can see the analogous clear disagreements between the Livermore data [21] and the results of calculations in the model for all energies investigated,

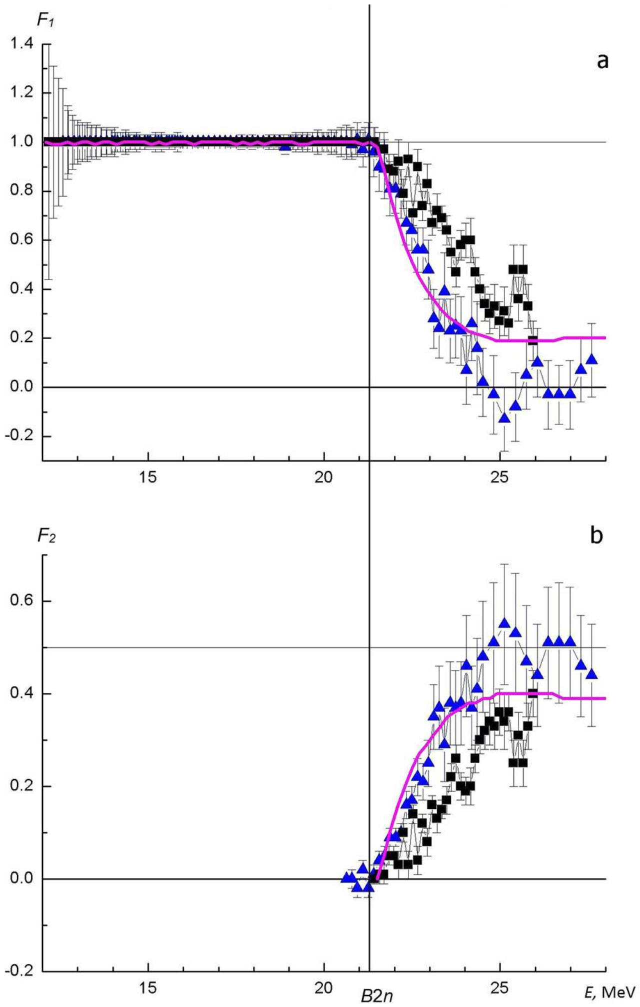

Fig. 1. The comparison of F_1 (a) and F_2 (b) ratios for ^{90}Zr (Livermore [21] triangles and Saclay [22] squares, CPNRM [13–15] lines).

for energies higher than $\sim 23\,\mathrm{MeV}$ for ^{91}Zr, for energies higher than $\sim 16\,\mathrm{MeV}$ for ^{92}Zr, and for energies higher than $\sim 15\,\mathrm{MeV}$ for ^{94}Zr. At the same time one can see clear negative F_1^{exp} values (correlated with the $F_2^{\mathrm{exp}} > 0.5$ values) at energies $\sim$ 25–30 MeV for ^{91}Zr, at energies $\sim$ 20–22 MeV for ^{92}Zr, and at energies $\sim$ 23–27 MeV for ^{94}Zr.

It can be concluded that the evaluation of reliable photoneutron reaction cross sections using the experimental-theoretical method (3) described above is of interest.

3 The new evaluated partial and total photoneutron reaction cross sections

3.1 Isotope ^{90}Zr

The experimental-theoretical method (3) for partial photoneutron reaction cross sections evaluation is based on using only the experimental data for neutron yield cross section (2). For ^{90}Zr we have both Livermore [21] and Saclay [22] experiments results obtained using quasimonoenergetic annihilation photons and the method for neutron multiplicity sorting.

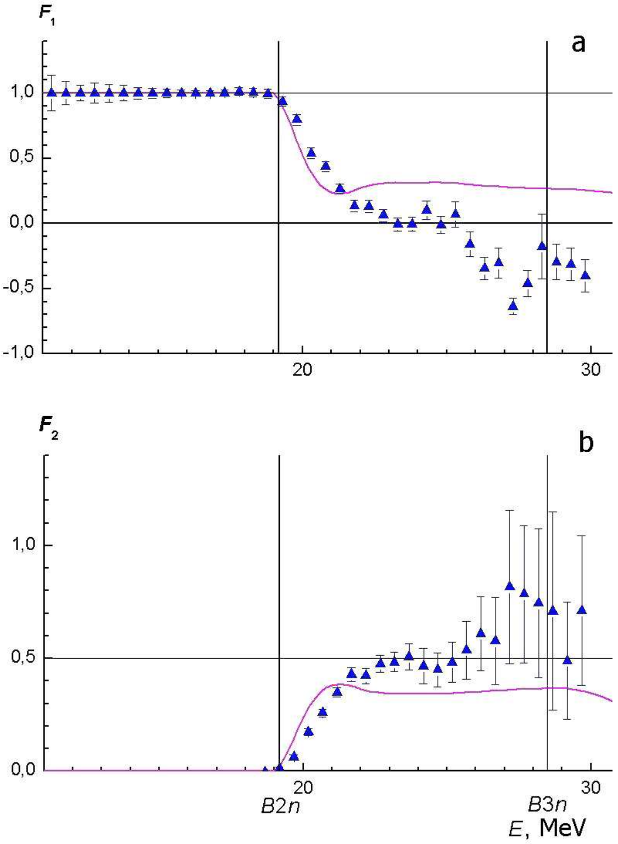

Fig. 2. The comparison of F_1 (a) and F_2 (b) ratios [10] for ^{91}Zr (Livermore [21] triangles, CPNRM [13–15] lines).

Figure 5 illustrates the choice of the $\sigma(\gamma, Sn)$ cross section to use in the evaluation procedure (3). One can see that the initial $\sigma^{\mathrm{th}}(\gamma, Sn)$ ([13–15], dotted line) is much more close to the Saclay $\sigma^{\mathrm{exp}}(\gamma, Sn)$ [22]. Therefore, the $\sigma^{\mathrm{exp}}(\gamma, Sn)$ [22] was chosen as a basis for the evaluation. Preliminarily for better agreement between $\sigma^{\mathrm{exp}}(\gamma, Sn)$ and $\sigma^{\mathrm{th}}(\gamma, Sn)$ for energies up to the energy threshold of $\sigma(\gamma, 2n)$ reaction $E^{\mathrm{int}} = B_{2n} = 21.3\,\mathrm{MeV}$ the latter one was slightly corrected (shifted to higher energies by 0.31 (17.09–16.78) MeV and multiplied by 1.04 (1011.86/970.80). The data for $E^{\mathrm{int}} = 21.3\,\mathrm{MeV}$ were used because at higher energies there is noticeable disagreement between $\sigma^{\mathrm{exp}}(\gamma, Sn)$ and $\sigma^{\mathrm{th}}(\gamma, Sn)$. Centers of gravity $E^{\mathrm{c.g.}}$ and integrated cross sections σ^{int} obtained for two values $E^{\mathrm{int}} = 21.3$ and $27.6\,\mathrm{MeV}$ are presented in table 1.

The ratios F_i^{th} (1) were obtained using the corrected $\sigma^{\mathrm{th\text{-}corr}}(\gamma, Sn)$ and corresponding to it $\sigma^{\mathrm{th\text{-}corr}}(\gamma, in)$. Evaluated cross sections $\sigma^{\mathrm{eval}}(\gamma, 1n)$, $\sigma^{\mathrm{eval}}(\gamma, 2n)$, and $\sigma^{\mathrm{eval}}(\gamma, \mathrm{tot})$ for ^{90}Zr are presented in fig. 6. The corresponding integrated cross section data are presented in table 2. One can see that evaluated cross sections disagree significantly with data of both experiments [21,22]. Those disagreements are clear for all photon energies in the case

Table 1. The centers of gravity $E^{\text{c.g.}}$ (MeV) and the integrated cross sections σ^{int} (MeV mb) for the experimental [21,22] and the calculated [13–15] data for the reaction $^{90}\text{Zr}(\gamma, Sn)$.

	$E^{\text{c.g.}}$	σ^{int}	$E^{\text{c.g.}}$	σ^{int}
Energy range	$E^{\text{int}} = B_{2n} = 21.3\,\text{MeV}$		$E^{\text{int}} = 27.6\,\text{MeV}$	
Experiment [21]	17.21 ± 0.16	858.65 ± 1.95	18.55 ± 0.38	1098.68 ± 4.48
Experiment [22]	17.09 ± 0.14	1011.86 ± 2.08	18.50 ± 0.20	1308.75 ± 3.15
Theory, initial	16.78 ± 0.83	970.80 ± 11.70	19.53 ± 0.81	1156.41 ± 11.84
Theory, corrected	17.09 ± 0.84	1011.00 ± 12.18	19.68 ± 0.81	1194.58 ± 12.32

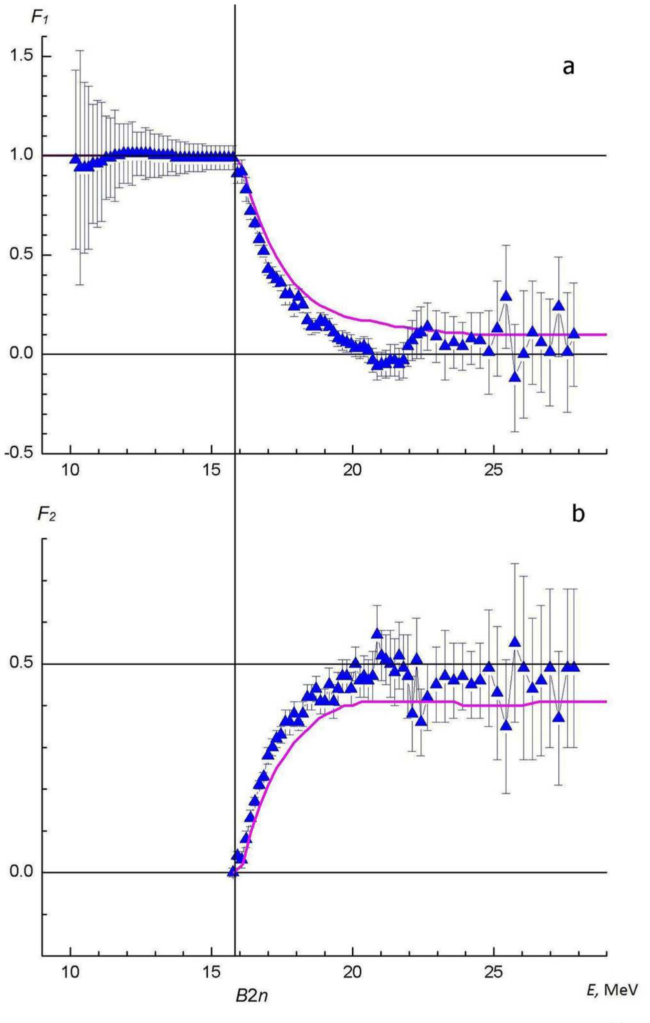

Fig. 3. The comparison of F_1 (a) and F_2 (b) ratios for ^{92}Zr (Livermore [21] triangles, CPNRM [13–15] lines).

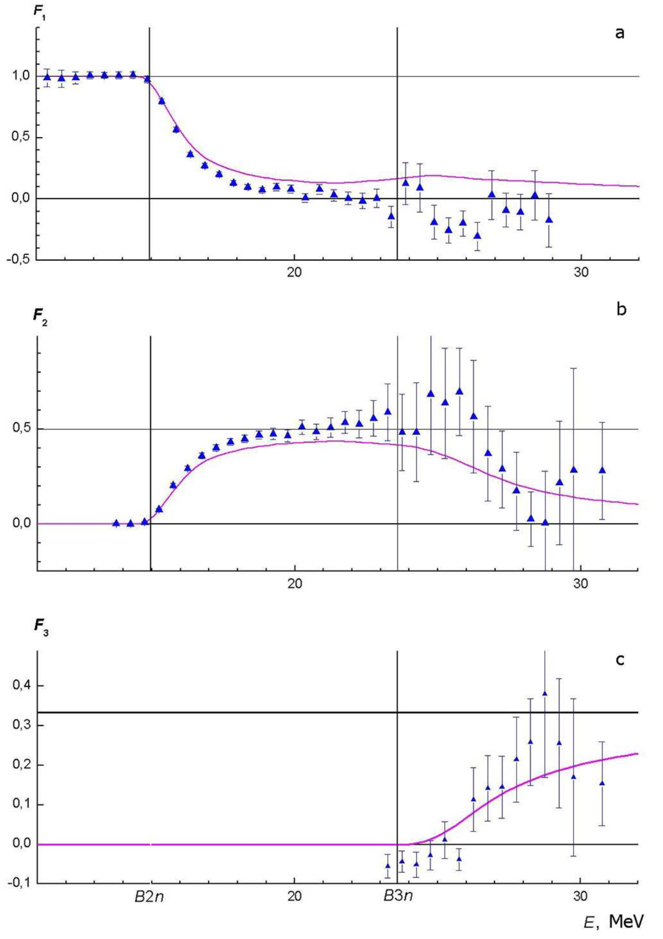

Fig. 4. The comparison of F_1 (a), F_2 (b) and F_3 (c) ratios [10] for ^{94}Zr (Livermore [21] triangles, CPNRM [13–15] lines).

of data from ref. [21]. This is natural because in accordance with data from fig. 5 $\sigma^{\text{exp}}(\gamma, Sn)$ [21] significantly differs from $\sigma^{\text{th}}(\gamma, Sn)$.

The disagreements between evaluated cross sections and experimental data [22] are much more interesting. In accordance with the data from fig. 1 evaluated and experimental data are identical in the energy range below $B_{2n} = 21.3\,\text{MeV}$. For energy range from 21.3 to 27.6 MeV evaluated $\sigma^{\text{eval}}(\gamma, 1n)$ is 35% ($198.44 - 146.88/146.88$) smaller in comparison with $\sigma^{\text{exp}}(\gamma, 1n)$ [17] but evaluated $\sigma^{\text{eval}}(\gamma, 2n)$ is 44% ($70.85 - 49.23/49.23$) larger in comparison with $\sigma^{\text{exp}}(\gamma, 2n)$ [17].

Such disagreements between Livermore and Saclay data and data evaluated using the experimental-theoretical method for many nuclei were investigated in many studies (for example, [3–12,23]). It has been shown that the main reason of those is the difference of procedures used to separate counts into $1n$ and $2n$ events. The main result of that was unreliable transmission of many neutrons from one partial reaction to another. It

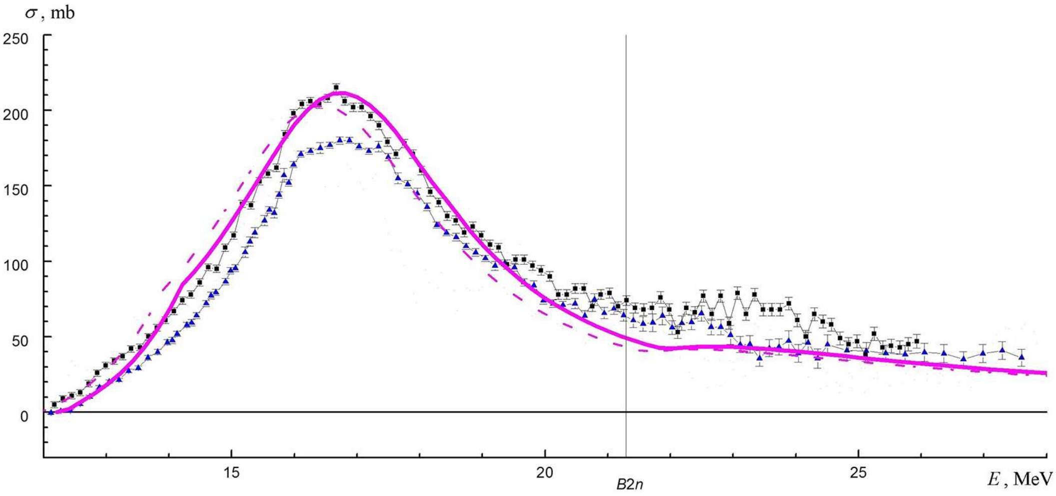

Fig. 5. The comparison of the experimental cross sections $\sigma(\gamma, Sn)$, obtained for ^{90}Zr at Livermore ([21] triangles) and Saclay ([22] squares) with the result of calculation (dotted line before and solid line after the correction) in the frame of the CPNRM [13–15].

Table 2. The integrated cross sections σ^{int} (MeV mb) of the evaluated partial reaction cross sections in comparison with the experimental data [21,22] for various reactions on ^{90}Zr.

Reaction	Evaluation	Experiment	
		[22]	[21]
$E^{\text{int}} = B_{2n} = 21.3\,\text{MeV}$			
$(\gamma, Sn)^{(\text{a})}$	1011.86±2.08	1011.86±2.08$^{(\text{a})}$	858.65±1.95
(γ, tot)	1011.30±11.86	1011.84±2.08	858.45±1.95
$(\gamma, 1n)$	1011.30±11.86	1011.84±2.08	858.42±1.93
$E^{\text{int}} = 27.6\,\text{MeV}$			
$(\gamma, Sn)^{(\text{a})}$	1308.75±3.15	1308.75±3.15$^{(\text{a})}$	1098.68±4.48
(γ, tot)	1228.76±12.45	1259.51±3.15	1029.73±5.09
$(\gamma, 1n)$	1158.18±12.24	1210.28±2.95	960.94±4.43
$(\gamma, 2n)$	70.85±1.67	49.23±1.09	68.79±2.51

(a) The experimental cross section [22] is the initial data for the evaluation procedure (3).

was concluded that some $(\gamma, 2n)$ events were mistakenly interpreted as two $(\gamma, 1n)$ events at Saclay. Therefore at Saclay data were overestimated for $(\gamma, 1n)$ and underestimated for $(\gamma, 2n)$ and the situation is vice versa at Livermore.

The possible reasons for that could be the following specific features of Saclay and Livermore neutron detectors.

At Livermore [21] the so-called "ring-ratio" method was used. The detector consisted of a 2-ft cube of paraffin in which were inserted 48 BF$_3$ tubes arranged in four concentric rings of 12 tubes each, at radial distances of 2.50, 4.25, 5.75, and 7.00 in. Low-energy neutrons (from reaction $(\gamma, 2n)$) should have enough time for moderation in the way to inner rings but high-energy neutrons (from reaction $(\gamma, 1n)$) should go to the outer rings passing inner rings. But due to multiple scattering some high-energy neutrons could return to inner rings made up largely of double-neutron events: "… the ratio of the number of neutrons detected on the outermost to hat in the innermost ring, the "ring-ratio" varies with the average neutron energy" [16].

At Saclay [22] the large Gd-loaded liquid scintillator calibrated by means of the ^{252}Cf source was used. There was a near constant value for the efficiency in the photoneutron data reduction procedure only up to neutron energy $\sim 5\,\text{MeV}$. In [1] it was written that such detector "suffered from a high background rate, made up largely of single-neutron events, which introduces larger uncertainties in the background subtraction and pile-up corrections".

The differences between the experimental and the evaluated cross sections for $(\gamma, 1n)$ and $(\gamma, 2n)$ reactions, correspondingly,

$$\Delta\sigma_1(\gamma, 1n) = \sigma^{\text{exp}}(\gamma, 1n) - \sigma^{\text{eval}}(\gamma, 1n), \tag{5}$$

$$\Delta\sigma_2(\gamma, 2n) = \sigma^{\text{exp}}(\gamma, 2n) - \sigma^{\text{eval}}(\gamma, 2n), \tag{6}$$

presented in fig. 7, directly confirm that in Saclay experiment [22] many neutrons were unreliably transmitted from the reaction $(\gamma, 2n)$ to the reaction $(\gamma, 1n)$.

3.2 Isotope ^{91}Zr

For isotope ^{91}Zr experimental photoneutron partial reaction cross section data were obtained only at Saclay [22]. Using physical criteria of data reliability those were analyzed in detail in ref. [10].

The newly evaluated cross sections were obtained using procedure (3) after a slight correction of the theoretical cross section $\sigma^{\text{th}}(\gamma, Sn)$. In order to reach the closest agreement with the experimental cross section $\sigma^{\text{exp}}(\gamma, Sn)$ it was shifted toward higher energies by 0.30 MeV and the result was multiplied by a factor of 0.84.

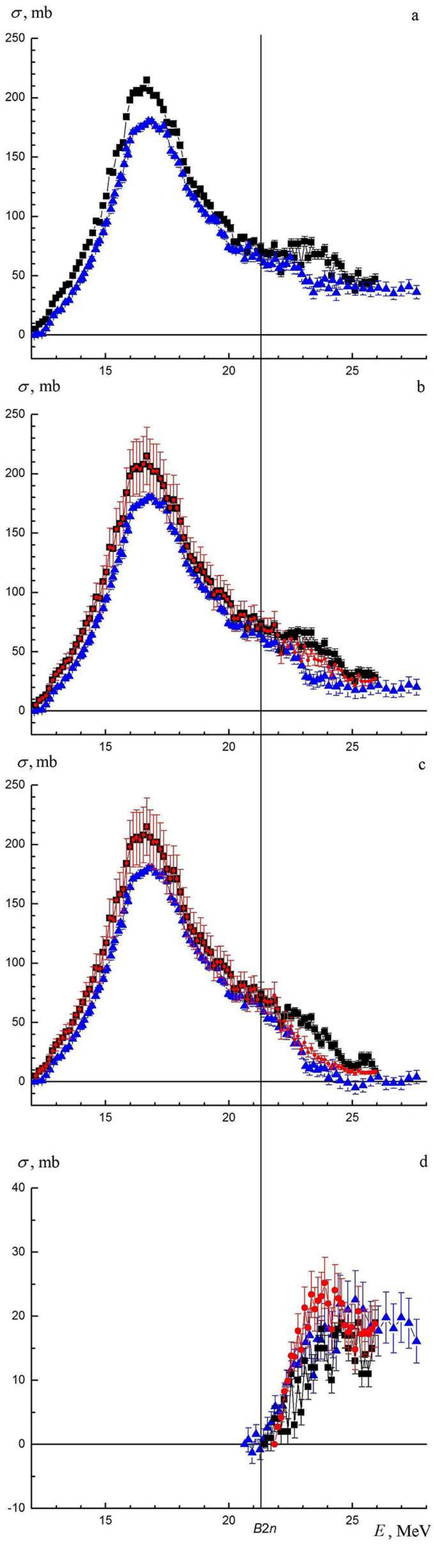

Fig. 6. The comparison of the evaluated (dots) and the experimental ([21] triangles and [22] squares) photoneutron reaction cross sections for ^{90}Zr: (a) $\sigma(\gamma, Sn)$, (b) $\sigma(\gamma, \mathrm{tot})$, (c) $\sigma(\gamma, 1n)$, (d) $\sigma(\gamma, 2n)$.

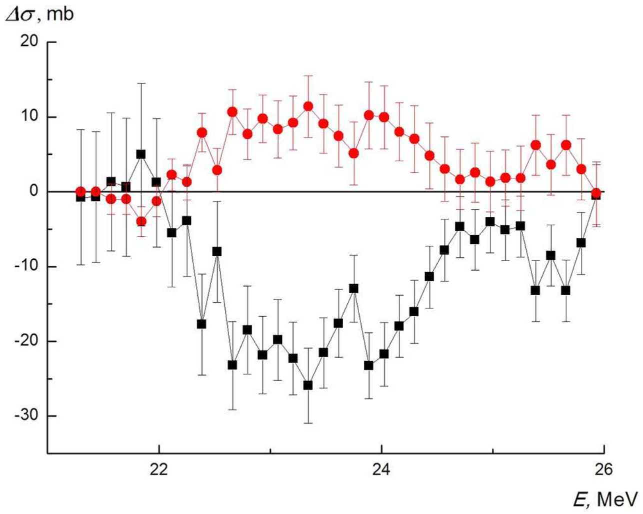

Fig. 7. The comparison of the differences $\Delta\sigma$ ((5), (6)) between the experimental [22] and the evaluated data for $(\gamma, 1n)$ reaction ($\Delta\sigma_1$, circles) and $(\gamma, 2n)$ reaction ($\Delta\sigma_2$, squares) on ^{90}Zr.

Table 3. The integrated cross sections σ^{int} (MeV mb) of the evaluated partial reaction cross sections in comparison with the experimental data [21] for various reactions on ^{91}Zr.

Reaction	Evaluation	Experiment
$E^{\mathrm{int}} = B_{2n} = 19.2\,\mathrm{MeV}$		
$(\gamma, Sn)^{(a)}$	782.3 ± 4.8	$782.3 \pm 4.8^{(a)}$
(γ, tot)	781.4 ± 4.8	782.8 ± 4.8
$(\gamma, 1n)$	782.6 ± 4.8	780.8 ± 22.0
$E^{\mathrm{int}} = 28.5\,\mathrm{MeV}$		
$(\gamma, Sn)^{(a)}$	1276.0 ± 17.2	$1235.8 \pm 12.5^{(a)}$
(γ, tot)	1091.6 ± 27.5	1061.4 ± 11.5
$(\gamma, 1n)$	947.5 ± 24.2	881.8 ± 11.0
$(\gamma, 2n)$	143.4 ± 6.0	174.8 ± 5.2

(a) The experimental cross section [21] is the initial data for the evaluation procedure (3).

Evaluated cross sections $\sigma^{\mathrm{eval}}(\gamma, 1n)$, $\sigma^{\mathrm{eval}}(\gamma, 2n)$, and $\sigma^{\mathrm{eval}}(\gamma, \mathrm{tot})$ for ^{91}Zr are presented in ref. [10]. The corresponding integrated cross section data are presented in table 3.

It is very important to point out that "$\sigma^{\mathrm{eval}} - \sigma^{\mathrm{exp}}$" differences for ^{91}Zr are opposite to those for ^{90}Zr: $\sigma^{\text{int-eval}}(\gamma, 1n) > \sigma^{\text{int-exp}}(\gamma, 1n)$ but $\sigma^{\text{int-eval}}(\gamma, 2n) < \sigma^{\text{int-exp}}(\gamma, 2n)$. The explanation is very clear. One has to have in mind that for ^{90}Zr there was the comparison of evaluated and Saclay [22] data but for ^{91}Zr there is the comparison of evaluated and Livermore [21] data. As was mentioned above on the basis of results obtained for many nuclei [3–12, 23] as a rule Saclay data were overestimated for $(\gamma, 1n)$ reaction and underestimated for $(\gamma, 2n)$ reaction. However, Livermore data were vice versa overes-

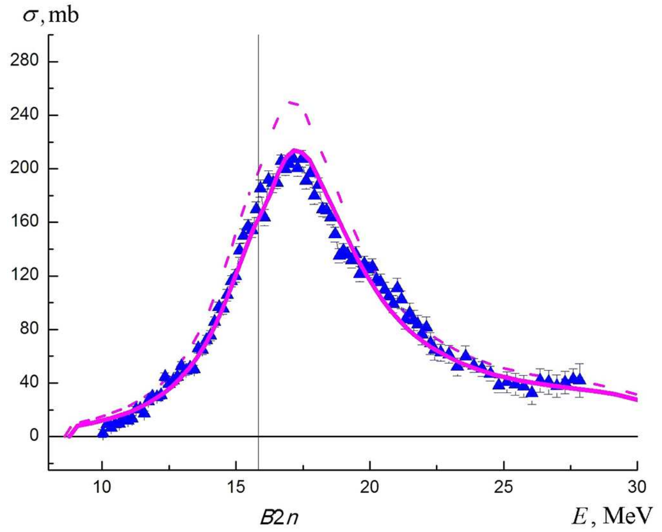

Fig. 8. The comparison of the experimental cross sections $\sigma(\gamma, Sn)$, obtained for ^{92}Zr at Livermore [21] with the result of calculation in the frame of the CPNRM [13–15], dotted line before and solid line after correction.

timated for $(\gamma, 2n)$ reaction and underestimated for $(\gamma, 1n)$ reaction.

3.3 Isotope ^{92}Zr

For the isotope ^{92}Zr the results also were obtained at Livermore [21]. The method of analysis of experimental data reliability and evaluation of new reliable data described above were used in this case and the results analogous to those for isotope ^{90}Zr were obtained.

Figure 8 illustrates the correction of $\sigma^{\rm th}(\gamma, Sn)$ [13–15] for a better agreement with $\sigma^{\rm exp}(\gamma, Sn)$ [21] used as initial data for the evaluating procedure by eq. (3): $\sigma^{\rm th}(\gamma, Sn)$ was shifted to higher energies by 0.17 MeV and multiplied by 0.86.

The ratios $F_i^{\rm th}$ (1) were obtained using the corrected $\sigma^{\text{th-corr}}(\gamma, Sn)$ and the corresponding to it $\sigma^{\text{th-corr}}(\gamma, in)$. Evaluated cross sections $\sigma^{\rm eval}(\gamma, 1n)$, $\sigma^{\rm eval}(\gamma, 2n)$, and $\sigma^{\rm eval}(\gamma, {\rm tot})$ for ^{92}Zr are presented in fig. 9. The corresponding integrated cross section data are presented in table 4.

In accordance with data from fig. 3 evaluated and experimental data are identical in the energy range below $B_{2n} = 15.8$ MeV. For energy range from 15.8 to 27.8 MeV evaluated $\sigma^{\rm eval}(\gamma, 1n)$ is 33% $(406.84 - 304.75/304.75)$ larger in comparison with $\sigma^{\rm exp}(\gamma, 1n)$ [21] but evaluated $\sigma^{\rm eval}(\gamma, 2n)$ is 18% $(447.54 - 379.47/379.47)$ smaller in comparison with $\sigma^{\rm exp}(\gamma, 2n)$ [21].

The differences (5) and (6) between the experimental and the evaluated cross sections for $(\gamma, 1n)$ and $(\gamma, 2n)$ reactions, correspondingly, presented in fig. 10, directly confirm that in Livermore experiment [21] many neutrons were unreliably transmitted from the reaction $(\gamma, 1n)$ to the reaction $(\gamma, 2n)$.

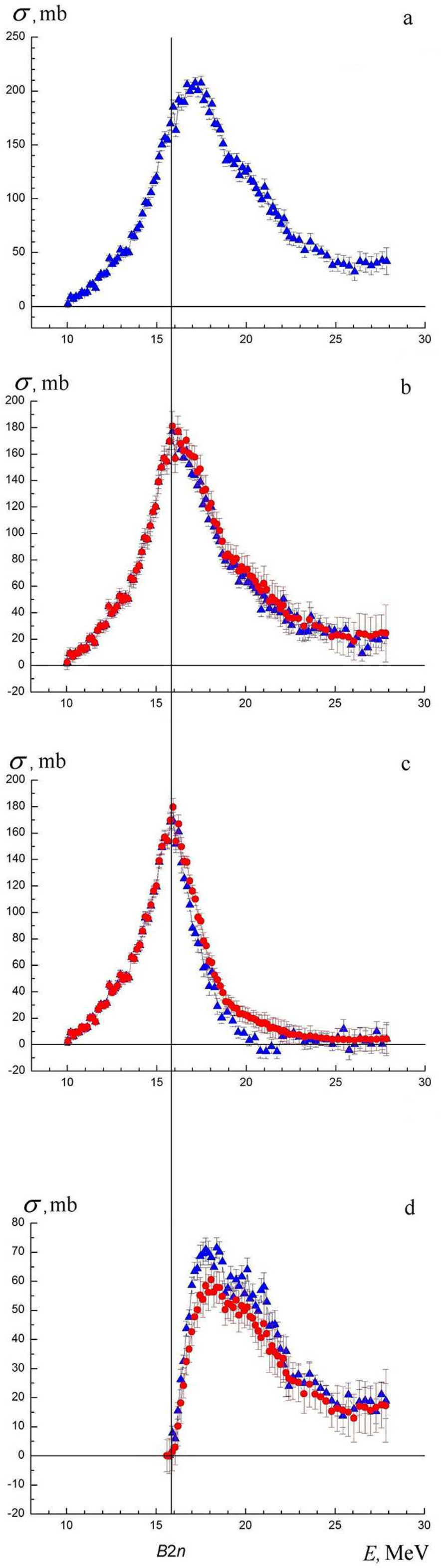

Fig. 9. The comparison of the evaluated (circles) and the experimental ([21] triangles) photoneutron reaction cross sections for ^{92}Zr: (a) $\sigma(\gamma, Sn)$, (b) $\sigma(\gamma, {\rm tot})$, (c) $\sigma(\gamma, 1n)$, (d) $\sigma(\gamma, 2n)$.

Table 4. The integrated cross sections σ^{int} (MeV mb) of the evaluated partial reaction cross sections in comparison with the experimental data [21] for various reactions on ^{92}Zr.

Reaction	Evaluation	Experiment [21]
$E^{\text{int}} = B_{2n} = 15.8\,\text{MeV}$		
$(\gamma, Sn)^{(a)}$	341.40 ± 2.33	$341.40 \pm 2.33^{(a)}$
(γ, tot)	341.40 ± 2.33	341.40 ± 2.33
$(\gamma, 1n)$	341.40 ± 2.34	340.33 ± 2.34
$E^{\text{int}} = 27.8\,\text{MeV}$		
$(\gamma, Sn)^{(a)}$	1548.34 ± 8.45	$1548.34 \pm 8.45^{(a)}$
(γ, tot)	1144.79 ± 14.64	1089.12 ± 6.13
$(\gamma, 1n)$	748.24 ± 8.45	645.08 ± 7.95
$(\gamma, 2n)$	379.47 ± 8.45	447.54 ± 4.22

(a) The experimental cross section [21] is the initial data for the evaluation procedure (3).

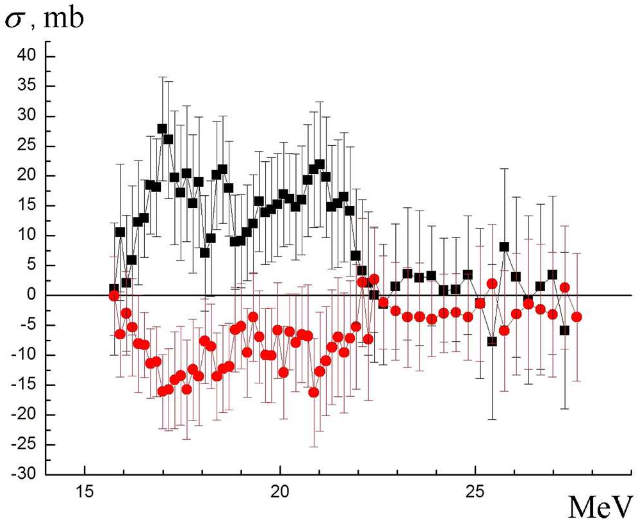

Fig. 10. The comparison of the differences $\Delta\sigma$ ((5), (6)) between the experimental [21] and the evaluated data for $(\gamma, 1n)$ reaction ($\Delta\sigma_1$, circles) and $(\gamma, 2n)$ reaction ($\Delta\sigma_2$, squares) on ^{92}Zr.

One can see that in accordance with the above comments "$\sigma^{\text{eval}} - \sigma^{\text{exp}}$" differences for ^{92}Zr are similar to those for ^{91}Zr and opposite to those for ^{90}Zr.

3.4 Isotope ^{94}Zr

For isotope ^{94}Zr experimental photoneutron partial reaction cross section data were obtained only at Saclay [22]. Using physical criteria of data reliability those were analyzed in detail in ref. [10]. The newly evaluated cross sections were obtained using procedure (3) after a slight correction of the theoretical cross section $\sigma^{\text{th}}(\gamma, Sn)$. In order to reach the closest agreement with the experimental cross section $\sigma^{\text{exp}}(\gamma, Sn)$ it was shifted toward higher energies by 0.02 MeV and the result was multiplied by a factor of 0.85.

Table 5. The integrated cross sections σ^{int} (MeV mb) of the evaluated partial reaction cross sections in comparison with the experimental data [21] for various reactions on ^{94}Zr.

Reaction	Evaluation	Experiment
$E^{\text{int}} = B_{2n} = 15.0\,\text{MeV}$		
$(\gamma, Sn)^{(a)}$	260.5 ± 2.4	$260.5 \pm 2.4^{(a)}$
(γ, tot)	262.8 ± 2.4	259.8 ± 2.4
$(\gamma, 1n)$	261.8 ± 9.1	260.6 ± 2.5
$E^{\text{int}} = B_{2n} = 23.6\,\text{MeV}$		
$(\gamma, Sn)^{(a)}$	1506.7 ± 12.2	$1506.7 \pm 12.0^{(a)}$
(γ, tot)	1086.3 ± 30.1	1011.1 ± 8.0
$(\gamma, 1n)$	652.4 ± 15.3	546.1 ± 11.5
$(\gamma, 2n)$	429.0 ± 4.3	494.5 ± 7.4
$E^{\text{int}} = 31.0\,\text{MeV}$		
$(\gamma, Sn)^{(a)}$	$2067.2 \pm 40.0^{(a)}$	$2067.2 \pm 40.0^{(a)}$
(γ, tot)	1311.4 ± 8.0	1114.0 ± 13.6
$(\gamma, 1n)$	694.9 ± 14.3	434.7 ± 31.5
$(\gamma, 2n)$	539.4 ± 10.7	662.6 ± 33.8
$(\gamma, 3n)$	56.1 ± 12.3	85.4 ± 18.5

(a) The experimental cross section [21] is the initial data for the evaluation procedure (3).

Evaluated cross sections $\sigma^{\text{eval}}(\gamma, 1n)$, $\sigma^{\text{eval}}(\gamma, 2n)$, $\sigma^{\text{eval}}(\gamma, 3n)$, and $\sigma^{\text{eval}}(\gamma, \text{tot})$ for ^{94}Zr are presented in ref. [10]. The corresponding integrated cross section data are presented in table 5.

One can see that in accordance with above comments "$\sigma^{\text{eval}} - \sigma^{\text{exp}}$" differences for ^{94}Zr are similar to those for ^{91,92}Zr and opposite to those for ^{90}Zr.

The integrated cross sections $\sigma^{\text{int}}(\gamma, \text{tot}) \approx \sigma^{\text{int}}(\gamma, \text{abs})$ obtained for ^{76,78,80,82}Se Giant Dipole Resonance energies [1,21] are presented in table 6 as a comparison of experimental and evaluated data and the classical dipole sum rule of Thomas, Reiche and Kuhn (TRK) estimations.

One can see that the evaluated integrated cross sections of total photoneutron reaction are larger than Livermore data [1,21] and more close to TRK values.

Really the unreliable moving of many neutrons from $(\gamma, 1n)$ to $(\gamma, 2n)$ reaction channel in experiment [21] in agreement with figs. 1–4 could not be the reason of total photoneutron reaction (4) increase. This increase could be connected to the problem of neutron detector efficiency used in experiment [21]. The efficiency of the neutron detector described above was about 40%. It means that many of the neutrons produced in both reactions under discussion were lost. Therefore it could be that the neutron yield cross section (1) used in the evaluation procedure (2) is more reliable in comparison to total photoneutron cross section (4) because it is rather independent from

Table 6. Evaluated and experimental (according to [1,21]) cross sections σ^{int} (in MeV mb) integrated from $B1n$ up to E^{int} in comparison with TRK sum rule estimations.

	^{90}Zr ($E^{\text{int}} = 27.6\,\text{MeV}$)	^{91}Zr ($E^{\text{int}} = 28.5\,\text{MeV}$)	^{92}Zr ($E^{\text{int}} = 27.8\,\text{MeV}$)	^{94}Zr ($E^{\text{int}} = 31.0\,\text{MeV}$)
$60NZ/A$(TRK)	1310	1330	1350	1380
Evaluation	1229	1092	1145	1311
[21]$^{(a)}$	1030	1061	1089	1114
[1]$^{(a)}$	1060	1103	1091	1121

$^{(a)}$ Integrated cross section data are slightly different in refs. [1,21].

the neutron multiplicity sorting problem,

$$\sigma(\gamma,\text{tot}) = \sigma(\gamma,Sn) - \sigma(\gamma,2n). \qquad (7)$$

So the results obtained could directly confirm the conclusion of refs. [3,12] that the main reason of disagreements under discussion is a very complex and indirect relationship between neutron energy and multiplicity. For investigated isotopes ^{90,91,92,94}Zr those relationships are different.

It should be pointed out that the results of new evaluations based on using objective physical criteria of data reliability noticeably differ from previously evaluated data obtained using well-known and widely used GUNF and GNASH codes [24] and included into the IAEA Photonuclear Data Library (PDL). The corresponding differences were found for ^{91}Zr, ^{159}Tb and ^{197}Au [25]. It was shown that the reason could be that previous evaluations [24] were based on the total photoabsorption data really close to the total photoneutron reaction data (4) but not on the photoneutron yield cross sections (2), which are rather independent of neutron multiplicity. It seems that despite the IAEA PDL has been extremely useful to a broad community, it is now evident that it needs to be revised and updated [26].

4 Summary and conclusions

The results of two experiments for the determination of the partial photoneutron reaction cross sections for ^{90,91,92,94}Zr carried out at Livermore [21] and for ^{90}Zr at Saclay [22] using different modifications of the method of photoneutron multiplicity sorting were discussed in detail. New objective physical criteria $F_i = \sigma(\gamma,in)/\sigma(\gamma,Sn)$ based on the experimental neutron yield cross section $\sigma^{\text{exp}}(\gamma,Sn)$ rather independent from the neutron multiplicity sorting problem and the equations of the theoretical CPNRM [13–15] were used to analyze systematic uncertainties present in the experimental cross sections.

It was shown that the definite systematic uncertainties of the neutron multiplicity sorting method were the reason for the noticeable disagreements between the partial photoneutron reactions cross sections obtained. The disagreements between two experimental data for ^{90}Zr under discussion [21,22] were the results of erroneous sorting of neutrons between various partial reactions, primarily reactions $(\gamma,1n)$ and $(\gamma,2n)$.

The experimental-theoretical method for evaluating the partial reaction cross sections was used for the determination of new cross sections for the $(\gamma,1n)$ and $(\gamma,2n)$ reactions on ^{90,91,92,94}Zr and additionally for the reaction $(\gamma,3n)$ for ^{94}Zr satisfied the objective physical criteria of data reliability. The partial reaction cross sections newly evaluated noticeably disagree with the data of both experiments [21,22] under discussion. Therefore a discussion of underlying physics is of great interest.

Since the results of new evaluations based on objective physical criteria noticeably differ from previously evaluated data included into the IAEA Photonuclear Data Library (PDL), therefore it seems that the IAEA PDL needs to be revised and updated. So the IAEA Coordinated Research Project on updating the PDL was adopted for the period 2016–2019 [25].

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