# Photoneutron reaction cross-section data for <sup>75</sup>As: Experiments and evaluation

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The problems of reliability of partial photoneutron cross-section data for <sup>75</sup>As obtained using beams of quasimonoenergetic photons produced by annihilation in flight of relativistic positrons and the method of neutron multiplicity sorting at Lawrence Livermore National Laboratory (USA) and Centre d'Etudes Nucleaires of Saclay (France) were discussed using the objective physical data reliability criteria. New data for photoneutron reaction cross sections for <sup>75</sup>As, satisfying those criteria, were obtained using the experimental-theoretical method for partial reaction cross-section evaluating. Evaluated data for <sup>75</sup>As were compared with experimental data and the problems of significant disagreements between Livermore and Saclay data were discussed in detail. It was shown that experimental data for the ( $\gamma$ , 1n) and ( $\gamma$ , 2n) reactions' cross sections obtained at Livermore are not reliable because of significant systematic uncertainties of different nature.

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### I. INTRODUCTION

Cross sections of partial photoneutron reactions with different number of outgoing particles, primarily  $(\gamma, 1n)$ ,  $(\gamma, 2n)$ , and  $(\gamma, 3n)$ , form an important body of experimental data [1–3] that are widely used in both fundamental and applied research in various branches of science, such as nuclear physics, astrophysics, geology, chemistry, and medicine. The majority of those data was obtained in the experiments, carried out using quasimonoenergetic annihilation photon beams and the method of photoneutron multiplicity sorting at Lawrence Livermore National Laboratory (USA) and Centre d'Etudes Nucleaires of Saclay (France) [1,2,4].

The significant systematic data disagreements between data of both laboratories for the cross sections of the  $(\gamma, 1n)$  and  $(\gamma, 2n)$  reactions were obtained for 19 nuclei: <sup>51</sup>V, <sup>75</sup>As, <sup>89</sup>Y, <sup>90</sup>Zr, <sup>115</sup>In, <sup>116,117,118,120,124</sup>Sn, <sup>127</sup>I, <sup>133</sup>Cs, <sup>159</sup>Tb, <sup>165</sup>Ho, <sup>181</sup>Ta, <sup>197</sup>Au, <sup>208</sup>Pb, <sup>232</sup>Th, and <sup>238</sup>U [5–9]. It was found that as a rule the  $(\gamma, 1n)$  reaction cross sections are larger at Saclay, but the  $(\gamma, 2n)$  cross sections are larger at Livermore, up to 60–100%. For the nuclei mentioned above the average ratio  $\sigma_{\rm S}^{\rm int}/\sigma_{\rm L}^{\rm int}$  of integrated cross sections for Saclay data to those for Livermore data is equal to 1.08 in the case of the  $(\gamma, 1n)$  reaction.

At the same time the average disagreement between neutron yield cross sections,

$$\sigma(\gamma, Sn) = \sigma(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n), \quad (1)$$

obtained in various laboratories is about 10% [7]. This means that one has noticeable systematic uncertainties in partial reaction cross sections. The reasons are the definite shortcomings of the neutron multiplicity-sorting method. So nobody knows which data are reliable or not.

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The experimental-theoretical method for evaluating the partial reaction cross sections was developed in order to resolve these problems [10]. In this method an experimental neutron yield cross section  $\sigma(\gamma, Sn)$  [Eq. (1)], which is rather independent from the neutron multiplicity-sorting problems because all outgoing neutrons are included, was decomposed into partial reaction cross sections,

$$\sigma^{\text{eval}}(\gamma, in) = F_i^{\text{theor}} \sigma^{\exp}(\gamma, Sn)$$
$$= [\sigma^{\text{theor}}(\gamma, in) / \sigma^{\text{theor}}(\gamma, Sn)] \sigma^{\exp}(\gamma, Sn), \quad (2)$$

using transitional neutron multiplicity functions:

$$F_i^{\text{theor}} = \sigma^{\text{theor}}(\gamma, in) / [\sigma^{\text{theor}}(\gamma, 1n) + \sigma(\gamma, 1n1p) + 2\sigma^{\text{theor}}(\gamma, 2n) + 3\sigma^{\text{theor}}(\gamma, 3n) + \cdots ].$$
(3)

Those were calculated for partial reactions ( $\gamma$ , *in*) with definite neutron multiplicity factors *i* = 1, 2, 3, ... within the framework of the combined photonucleon reaction model (CPNRM) [11,12]. 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. Additionally the model considers deformation of nucleus and isospin splitting of its giant dipole resonance. The CPNRM was well tested for many medium and heavy nuclei.

The ratios  $F_i$  [Eq. (3)] of specific partial reaction cross sections to that of the neutron yield reaction were proposed as the objective physical criteria of reliability of experimental partial photoneutron reaction cross-section data [10]. According to the definitions (3),  $F_1 > 1.00$ ,  $F_2 > 0.50$ ,  $F_3 >$ 0.33, etc., never can be reliable. So  $F_i^{exp}$  values larger than those upper limits mean that partial reaction cross sections are not reliable because experimental sorting of neutrons between those reactions was carried out with large systematic

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uncertainties. Additionally it should be underlined that ratios  $F_i$  include only the cross-section terms and therefore they should be definitely positive.

For 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 [8-10,13-21] that in many cases experimental partial reaction cross sections do not satisfy the proposed data reliability criteria because of many negative cross-section values and, correspondingly,  $F_1^{exp}$  values and/or  $F_i^{exp}$  values larger than the upper limits mentioned above, and that evaluated cross sections are noticeably different from those obtained in experiments using the neutron multiplicitysorting method.

The newly evaluated partial photoneutron reaction cross sections for <sup>181</sup>Ta [14], <sup>197</sup>Au [10], and <sup>209</sup>Bi [18] were compared with the results of measurements of multineutron reaction yields using bremsstrahlung beams and the activation method [19–21]. In this method direct identification of a specific partial reaction is based on final nucleus, but not outgoing neutrons', features. It was concluded that evaluated partial reaction cross sections are reliable because they agree with data obtained using the activation method although they contradict data obtained using the neutron multiplicity-sorting method. Therefore it was concluded that if  $F_i^{exp}$  are noticeably different from  $F_{I}^{\text{theor}}$  one has definite doubts in experimental data reliability.

So one could have three partial photoneutron reaction cross-section data reliability criteria.

- (1) Ratios  $F_i^{exp}$  should not have values larger than the upper limits mentioned above.
- (2) Cross sections  $\sigma^{\exp}(\gamma, in)$  and correspondingly the ratios  $F_i^{exp}$  should not have negative values. (3) Differences between  $F_i^{exp}$  and  $F_I^{theor}$  should not be
- noticeable.

It was shown, in general, that the main reason for noticeable disagreements between the partial reaction cross sections obtained at Livermore and Saclay is the difference between procedures used to separate counts into 1n and 2n events [8–10,13–21]. The same photoneutron multiplicity-sorting method, based on its kinetic-energy measuring, was used at Saclay and Livermore, but the types of so-called slowingdown neutron detectors were quite different.

The specifically calibrated large Gd-loaded liquid scintillator was used at Saclay. It is important to point out that there are definite technical reasons for some overestimation of  $\sigma(\gamma, 1n)$  in comparison to  $\sigma(\gamma, 2n)$ . As was written in [4], the Saclay detector "...suffers from a much higher background rate, made up largely of single-neutron events, which introduces larger uncertainties in the background subtractions and pile-up corrections ...." It could be concluded that there was an opportunity for some overstating in counting 1n events and correspondingly understating in 2n events and therefore for unreliable (erroneous) transmission of neutrons from the  $(\gamma, 2n)$  reaction into the  $(\gamma, 1n)$  reaction.

Many BF3 counters in several concentric counter rings embedded in a paraffin moderator were used at Livermore, as the so-called ring-ratio method. It could be supposed that

there were definite technical reasons for possible overestimation of  $\sigma(\gamma, 2n)$  in comparison to  $\sigma(\gamma, 1n)$  in the case of this detector. Low-energy neutrons, originating primarily from a  $(\gamma, 2n)$  reaction, must have time to be moderated to thermal energy on their path to one of the inner rings, while high-energy neutrons originating, primarily, from the reaction  $(\gamma, 1n)$  must traverse those rings and undergo moderation on their path to one of the outer rings. However, because of multiple scattering, it was not mandatory that the path of a fast neutron was rectilinear; such a neutron could return to the inner rings upon traveling along a curvilinear trajectory. So there was a reason for some overstating in counting 2n events and correspondingly understating in 1n events and therefore for unreliable (erroneous) transmission of neutrons from the  $(\gamma, 1n)$  reaction into the  $(\gamma, 2n)$  reaction.

The position of partial reaction cross-section data for <sup>75</sup>As in the systematics of integrated cross-section ratios mentioned above is very specific and complicated. The point is that for this nucleus the ratios  $\sigma_S^{int}/\sigma_L^{int}$  are near identical for both  $(\gamma, 1n)$  and  $(\gamma, 2n)$  reactions, but at the same time both are extremely large. Namely,  $\sigma_{\rm S}^{\rm int}(\gamma, 1n)/\sigma_{\rm L}^{\rm int}(\gamma, 1n)$  is equal to 1.22 and  $\sigma_{\rm S}^{\rm int}(\gamma, 2n)/\sigma_{\rm L}^{\rm int}(\gamma, 2n)$  is equal to 1.21. Thus, the disagreements between cross sections of the reactions  $(\gamma, 1n)$ and  $(\gamma, 2n)$  obtained at Livermore and Saclay are noticeable [1-3]. This means that some additional uncertainties of unknown nature are presented in data in addition to systematic uncertainties resulting from the neutron multiplicity-sorting method used.

It was mentioned above that the average disagreement between neutron yield cross sections [Eq. (1)] for many nuclei in general is about 10% [7]. But in the case of  $^{75}$ As this disagreement is much larger. In accordance with the results of systematic investigations [7–9,13], the ratio of integrated neutron yield cross sections  $\sigma_{\rm L}^{\rm int}(\gamma, Sn)/\sigma_{\rm S}^{\rm int}(\gamma, Sn)$ , calculated for energies up to  $E^{\text{int}} = 26.2 \text{ MeV}$ , is equal to 1.22. The correspondent cross sections  $\sigma(\gamma, Sn)$  obtained at Livermore [22] and Saclay [23] are presented in Fig. 1 in comparison with the results of calculation in the CPNRM [11,12]. The significant disagreements could mean that in addition to unreliable sorting of neutrons between 1n and 2n channels many neutrons in both of them could be lost.

It is important to point out that analogous extreme  $\sigma_{\rm s}^{\rm int}/\sigma_{\rm I}^{\rm int}$ values for  $(\gamma, 1n)$  and  $(\gamma, 2n)$  reactions were found before for <sup>181</sup>Ta [14]. For this nucleus  $\sigma_{\text{eval}}^{\text{int}}(\gamma, Sn)/\sigma_{\text{L}}^{\text{int}}(\gamma, Sn) = 1.24$ ,  $\sigma_{\text{eval}}^{\text{int}}(\gamma, \text{tot})/\sigma_{\text{L}}^{\text{int}}(\gamma, \text{tot}) = 1.30$ ,  $\sigma_{\text{eval}}^{\text{int}}(\gamma, 1n)\sigma_{\text{L}}^{\text{int}}(\gamma, 1n) = 1.46$ , and  $\sigma_{\text{eval}}^{\text{int}}(\gamma, 2n)/\sigma_{\text{L}}^{\text{int}}(\gamma, 2n) = 1.05$ . This means that the greater the fraction of the  $(\gamma, 1n)$  partial reaction cross section in the definite other reaction cross sections the higher the degree of disagreement:  $1.24 \rightarrow 1.30 \rightarrow 1.46$ . Upon the subsequent transition to the cross section  $\sigma(\gamma, 2n)$ , in which the fraction of  $\sigma(\gamma, 1n)$  is naturally equal to zero, the ratio under discussion decreases sharply to 1.05. This means definitely that at Livermore many neutrons from the 1n channel were lost and therefore for  $^{181}$ Ta the ratio  $\sigma_{\rm S}^{\rm int}(\gamma, 1n)/\sigma_{\rm L}^{\rm int}(\gamma, 1n)$  is equal to the extremely small value 0.89, but the ratio  $\sigma_{\rm S}^{\rm int}(\gamma,2n)/\sigma_{\rm L}^{\rm int}(\gamma,2n)$  is equal to the extremely large value 1.25. At the same time the ratio  $\sigma_{\rm S}^{\rm int}(\gamma, Sn)/\sigma_{\rm L}^{\rm int}(\gamma, Sn)$  has also the extremely large value 1.24.



FIG. 1. Comparison of the experimental ([22], triangles; [23], squares) neutron yield cross sections  $\sigma(\gamma, Sn)$  with the cross sections calculated before (dotted line) and after (solid line) correction (see further) in the CMPNR ([11,12].

Because differences between evaluated and experimental cross sections obtained for many nuclei are not absolutely identical for reactions ( $\gamma$ , 1n) and ( $\gamma$ , 2n) [8–10,13–21], neutron losses are possible in 1n, or in 2n, or in both channels. Therefore the detailed analysis of <sup>75</sup>As data reliability using the objective physical criteria in addition to the results of previous analysis for <sup>181</sup>Ta is of large interest.

In Fig. 1 the energy thresholds B1n1p = 17.1 MeV and B2n = 18.2 MeV of the reactions  $(\gamma, 1n1p)$  and  $(\gamma, 2n)$  for <sup>75</sup>As, correspondingly, are indicated. These values mean that in wide energy regions  $\sigma(\gamma, 1n)$ , presented in atlases and databases [1,2], in reality is  $[\sigma(\gamma, 1n) + \sigma(\gamma, 1n1p)]$ .

## II. ANALYSIS OF THE RELIABILITY OF PARTIAL PHOTONEUTRON REACTION CROSS SECTIONS BASED ON THE OBJECTIVE PHYSICAL CRITERIA

As was mentioned above, the ratios [Eq. (3)] of specific partial reaction cross sections to that of neutron yield reaction were proposed as the objective physical criteria of partial photoneutron reaction cross-section reliability [10]. It was found that for many nuclei experimental partial photoneutron reaction cross sections obtained using quasimonoenergetic annihilation photons and the photoneutron multiplicity-sorting method are not reliable because in many photon energy regions they do not satisfy proposed data reliability criteria [8–10,13–21]. It was shown that many  $F_1^{exp}$  values are negative, and/or  $F_2^{exp}$  values are negative or larger than 0.50, and/or  $F_3^{exp}$  values are negative or larger than 0.33, and/or there are noticeable differences between  $F_i^{exp}$  and  $F_I^{theor}$  values.

Further, because the energy threshold B3n of the reaction  $^{75}\text{As}(\gamma, 3n)^{72}\text{As}$  is equal to 29.02 MeV, only  $(\gamma, 1n)$  and  $(\gamma, 2n)$  reaction cross sections will be used for obtaining



FIG. 2.  $F_1^{exp}$  (a) and  $F_2^{exp}$  (b) data obtained for <sup>75</sup>As using experimental data of Livermore ([22], triangles) and Saclay ([23], squares) in comparison with results of calculated  $F_{1,2}^{\text{theor}}$  (model from [11,12], lines).

functions:

$$F_i^{\exp} = \sigma^{\exp}(\gamma, in) / \sigma^{\exp}(\gamma, Sn)$$
  
=  $\sigma^{\exp}(\gamma, in) / [\sigma^{\exp}(\gamma, 1n) + 2s^{\exp}(\gamma, 2n)].$  (4)

The comparisons of  $F_1^{exp}$  and  $F_2^{exp}$  data obtained for <sup>75</sup>As using Livermore [22] and Saclay [23] experimental data with calculated  $F_1^{theor}$  [11,12] are presented in Fig. 2.

It is important to underline that all  $F_I^{\text{theor}}$  values were obtained taking into account contributions of  $\sigma(\gamma, 1n1p)$ . But in accordance with the CPNRM calculations [11,12] it was found that at all photon energies under discussion values of  $\sigma(\gamma, 1n1p)$  are about ten times smaller in comparison to the  $\sigma(\gamma, 2n)$  values.

In Fig. 2 one can see that the Saclay experimental cross sections [23] satisfy in general the three above-mentioned physical data reliability criteria at all energies. Therefore there are not serious reliability problems with Saclay data.

At the same time in Fig. 2 one can see that the Livermore cross sections [22] satisfy data reliability criteria at energies only up to about 22 MeV. At larger energies there are noticeable differences between  $F_i^{exp}$  and  $F_1^{theor}$ . Moreover at energies 25–29 MeV there are many  $F_1^{exp}$  values very close to zero, several of which are negative. Correspondent  $F_2^{exp}$  values are very close to the upper limit 0.50, and several of

	$E^{\text{int}} = 22.4 \text{ MeV}$		$E^{\text{int}} = 26.2 \text{ MeV}$	
	$\sigma^{ m int}$	$E^{\mathrm{c.g.}}$	$\sigma^{ m int}$	$E^{\mathrm{c.g.}}$
Experiment [22]	$829.41 \pm 2.49$	$17.40 \pm 0.22$	$1018.07 \pm 3.39$	$18.64 \pm 0.29$
Experiment [23]	$1035.20 \pm 5.22$	$17.41 \pm 0.37$	$1308.77 \pm 6.61$	$18.81\pm0.42$
Theory [11,12]	$974.91 \pm 2.49$	$17.51 \pm 0.90$	$1264.45 \pm 13.38$	$19.02\pm0.81$
Theory corrected	$1044.87 \pm 13.06$	$17.46\pm0.90$	$1351.30 \pm 14.24$	$18.97\pm0.81$

TABLE I. Experimental [22,23] and calculated [11,12] integrated (up to energy  $E^{int}$ ) cross sections  $\sigma^{int}$  (in MeV mb) and centers of gravity  $E^{c.g.}$  (in MeV) for neutron yield cross section  $\sigma^{exp}(\gamma, Sn)$ .

those are larger than the limit. Livermore data are clearly underestimated for  $\sigma(\gamma, 1n)$  and overestimated for  $\sigma(\gamma, 2n)$ in comparison with the calculated data. Therefore, reliability of experimental cross sections [22] of both partial reactions  $(\gamma, 1n)$  and  $(\gamma, 2n)$  could be seriously called into question.

This phenomenon is connected definitely with shortcomings of the experimental method for neutron multiplicity sorting based on its energy measurement. In Refs. [19,24-26] photoneutron energy spectra for <sup>116</sup>Sn, <sup>141</sup>Pr, <sup>181</sup>Ta, <sup>186</sup>W, <sup>208</sup>Pb, and <sup>209</sup>Bi were calculated using the CPNRM [11,12]. It was shown that similarity between energies of neutrons from different partial reactions greatly complicates the procedure of determining neutron multiplicity from this energy and makes this procedure ambiguous. Noticeable differences between data obtained at Saclay and Livermore (and between data obtained at the same laboratory but for different nuclei) [8-10,13-21,24-26] could result from complicated and not direct connection between the energy of the neutron and its multiplicity. For example, neutrons from the  $(\gamma, 1n)$  reaction could have large energy in transitions from excited states of the target nucleus to the ground state of the final nucleus. But in the case of transitions to excited states of the final nucleus, neutrons could have energy noticeably smaller than and near those of neutrons from the  $(\gamma, 2n)$  reaction. Therefore reliability of neutron multiplicity determination based on its measured energy strongly directly depends on neutron energy spectra.

### III. NEW RELIABLE CROSS SECTIONS EVALUATED USING THE EXPERIMENTAL-THEORETICAL METHOD

To overcome the problems described above the experimental-theoretical method for evaluating partial reaction cross sections, not dependent on the systematic uncertainties of the experimental neutron multiplicity sorting, was used and the newly reliable data for many nuclei mentioned above were obtained [8-10,13-21,24-26].

In the experimental-theoretical method reliable partial reaction cross sections for each multiplicity (i = 1, 2) were evaluated using Eqs. (2), where  $\sigma^{\exp}(\gamma, Sn)$  is the experimental neutron yield cross-section and  $F_I^{\text{theor}}$  are the ratios calculated in the CPNRM [11,12]. This evaluating method means that competitions of partial reactions are specified in accordance with equations of the model and that the correspondent sum,

$$\sigma^{\text{eval}}(\gamma, Sn) = \sigma^{\text{eval}}(\gamma, 1n) + 2\sigma^{\text{eval}}(\gamma, 2n), \quad (5)$$

is equal to the experimental neutron yield cross section  $\sigma^{\exp}(\gamma, Sn)$ , which is rather independent on the experimentally determined neutron multiplicity.

From Fig. 1 one can see that  $\sigma^{\text{theor}}(\gamma, Sn)$ , calculated in the CPNRM, is much closer to the Saclay [23] than to the Livermore [22] cross section and therefore Saclay data were used in the evaluation procedure [Eq. (2)]. For better agreement with the experimental  $\sigma^{\exp}(\gamma, Sn)$  the calculated  $\sigma^{\text{theor}}(\gamma, Sn)$  was slightly corrected. It was shifted to low energies for 0.10 MeV and multiplied by 1.06. The correspondent integrated cross section and center of gravity values are presented in Table I.

The total photoneutron reaction cross section,

$$\sigma(\gamma, \text{tot}) = \sigma(\gamma, 1n) + \sigma(\gamma, 2n), \tag{5}$$

and the partial reaction cross sections  $\sigma(\gamma, 1n)$  and  $\sigma(\gamma, 2n)$  for <sup>75</sup>As evaluated using the experimental-theoretical method, described above, are presented in Fig. 3. The correspondent integrated cross-section values are presented in Table II.

The differences,

$$\Delta \sigma = \sigma^{\text{eval}} - \sigma^{\text{exp}},\tag{6}$$

between the evaluated and the experimental cross sections obtained separately for both partial reactions are presented in Fig. 4(a).

From Figs. 3 and 4(a) and Table II one can see that at energies up to B2n = 18.2 MeV evaluated cross sections are very close to the Saclay data [23]. At higher energies those data are also near to each other although some disagreements exist. For the energy range 18.2–26.2 MeV  $\sigma^{\text{eval}}(\gamma, 1n) = 293.34$  (890.14–596.80) MeV mb is 5% larger than  $\sigma^{\exp}(\gamma, 1n) = 278.06$  (873.82–595.78) MeV mb but  $\sigma^{\text{eval}}(\gamma, 2n) = 200.27$  MeV mb is 8% smaller than  $\sigma^{\exp}(\gamma, 2n) = 216.85$  (217.43–0.58) MeV mb. It is interesting to point out that this means that an arbitrarily small part of neutrons was moved not from the  $(\gamma, 2n)$  reaction into the  $(\gamma, 1n)$  one as in many cases investigated before but, vice versa, from the  $(\gamma, 1n)$  reaction into the  $(\gamma, 2n)$  one.

The differences  $\Delta\sigma$  between experimental and evaluated cross sections [Fig. 4(a)] obtained for partial reactions appear "reflected in a mirror." Almost all values of the  $\Delta\sigma(\gamma, 1n)$ are positive but those of the  $\Delta\sigma(\gamma, 2n)$  are negative. Such deviations clearly demonstrate the reason for systematic uncertainties in sorting of a certain number of neutrons between 1n and 2n channels because of indirect dependence of measured neutron kinetic energy and its determined multiplicity. As was mentioned above, it was shown that this kind of error

Reaction	[22]	[23]	Evaluation	[22]	[23]	Evaluation
	$E^{\text{int}} = B2n = 18.2 \text{ MeV}$			$E^{\text{int}} = 26.2 \text{ MeV}$		
$(\gamma, Sn)^{a}$	$485.03 \pm 1.63$	$596.92 \pm 3.63$	$596.80 \pm 9.47$	$1018.07 \pm 3.39$	$1308.77 \pm 6.61$	$1290.68 \pm 12.04$
$(\gamma, tot)$	$484.57 \pm 1.66$	$596.36 \pm 3.63$	$596.80 \pm 9.47$	$841.44 \pm 4.1$	$1091.25 \pm 6.61$	$1090.40 \pm 11.57$
$(\gamma, 1n)$	$484.46 \pm 1.65$	$595.78\pm3.57$	$596.80 \pm 9.47$	$666.33 \pm 3.73$	$873.82 \pm 5.60$	$890.14 \pm 10.98$
$(\gamma, 2n)$	$0.11\pm0.19$	$0.58\pm0.64$	-	$175.10\pm1.70$	$217.43\pm3.51$	$200.27\pm3.66$

TABLE II. Integrated cross sections  $\sigma^{\text{int}}$  (in MeV mb) of the evaluated cross sections of the total and partial photoneutron reactions on <sup>75</sup>As, compared with the experimental data [22,23].

<sup>a</sup>Experimental neutron yield cross section  $\sigma^{\exp}(\gamma, Sn)$  [23] used as the initial one for the evaluation procedure [Eq. (2)].

arises from using kinetic energy of neutrons to classify neutrons from different reaction channels [8–10,13–21,24–26]. Because neutron energy spectra are overlapping, this leads to incorrect classification of some of the detected neutrons. Therefore many neutrons originating from the 1*n* reaction could be assigned to the 2*n* channel, and vice versa. It should be noted that differences  $\Delta \sigma$  between evaluated and experimental data of Saclay [23] are arbitrarily small. Generally  $\Delta \sigma$  are about 2–3 mb with only three narrow "ejections" with amplitudes about 6–8 mb at energies near 18.5, 22.5, and 25.5 MeV. Therefore it can be concluded that the doubts in reliability of Saclay [23] data are arbitrarily poor.



FIG. 3. The comparison of the evaluated (circles) and the experimental ([22], triangles; [23], squares) cross sections of the reactions on <sup>75</sup>As: (a)  $\sigma(\gamma, \text{tot})$ , (b)  $\sigma(\gamma, 1n)$ , and (c)  $\sigma(\gamma, 2n)$ .

At the same time for the Livermore data [22] the situation is completely different. The differences  $\Delta\sigma$  between evaluated and experimental cross sections are significantly larger [Fig. 4(b)] in comparison with those for



FIG. 4. Comparison of the differences  $\Delta \sigma$  [Eq. (6)] between the evaluated and the experimental cross sections for <sup>75</sup>As: (a) for data from [23] [squares for reaction ( $\gamma$ , 1*n*); circles for ( $\gamma$ , 2*n*)] and (b) for data from [22] [triangles for reaction ( $\gamma$ , 1*n*); circles for ( $\gamma$ , 2*n*)].

	$E^{\text{int}} = 22.4 \text{ MeV}$		$E^{\text{int}} = 26.2 \text{ MeV}$	
	$\sigma^{ m int}$	$E^{ ext{c.g.}}$	$\sigma^{ m int}$	$E^{\mathrm{c.g.}}$
Experiment [23]	$1035.20 \pm 5.22$	$17.41 \pm 0.37$	$1308.77 \pm 6.61$	$18.81 \pm 0.42$
Experiment [22]	$829.41 \pm 2.49$	$17.40 \pm 0.22$	$1018.07 \pm 3.39$	$18.64\pm0.29$
Corrected [22]	$1034.89 \pm 3.11$	$17.41 \pm 0.22$	$1270.52 \pm 4.23$	$18.65\pm0.29$

TABLE III. Experimental [22,23] and corrected [22] integrated (up to energy  $E^{int}$ ) cross sections  $\sigma^{int}$  (in MeV mb) and centers of gravity  $E^{c.g.}$  (in MeV) for <sup>75</sup>As neutron yield cross section  $\sigma^{exp}(\gamma, Sn)$ .

Saclay data [Fig. 4(a)]. From Figs. 3 and 4(b) one can see that at energies higher than B2n = 18.2 MeV the Livermore data [22] are noticeably different from the evaluated data:  $\sigma^{\text{eval}}(\gamma, 1n) = 293.34$  MeV mb is 61% larger in comparison with  $\sigma^{\text{exp}}(\gamma, 1n) = 181.87$  (666.33–484.46) MeV mb, and  $\sigma^{\text{eval}}(\gamma, 2n) = 200.27$  MeV mb is 15% larger in comparison with  $\sigma^{\text{exp}}(\gamma, 2n) = 174.99$  (175.10–0.11) MeV mb. Moreover for both reactions the differences  $\Delta\sigma$  are positive. This is absolutely atypical for uncertainties under discussion [8–10,13–21,24–26]. Therefore one is forced to conclude that Livermore experimental data [22] are in general absolutely unreliable. So the special investigation of the situation with those data is of great interest.

## IV. SPECIAL INVESTIGATION OF THE LIVERMORE DATA

As was mentioned above, the values of both total and partial reactions cross sections obtained at Livermore [22] are noticeably smaller in comparison with the correspondent data obtained at Saclay [23]. But at the same time in Fig. 1 one can see that at all energies the shapes of neutron yield cross sections  $\sigma^{\exp}(\gamma, Sn)$  (1) obtained at Livermore and Saclay are very similar. So there is a question: Is not the simple difference in the normalization of data the reason for all discrepancies discussed above?



FIG. 5. Comparison of the experimental ([22], triangles; [23], squares) neutron yield cross sections  $\sigma(\gamma, Sn)$  with the corrected (open triangles) cross section [22].

The disagreements between Livermore and Saclay data for the reaction  $\sigma^{\exp}(\gamma, Sn)$  were specially investigated in detail for <sup>nat</sup>Zr, <sup>127</sup>I, <sup>141</sup>Pr, <sup>197</sup>Au, and <sup>nat</sup>Pb [27]. It was noted that Livermore and Saclay cross-section comparison "…implies an error in the flux determination or in the neutron detection efficiency or in both …" in the Livermore experiment. To overcome the disagreements it was recommended that the Saclay cross sections for various nuclei "be reduced by about  $18 \pm 4\%$ ." But in [7–9] about 500 photoneutron yield cross sections  $\sigma^{\exp}(\gamma, Sn)$  obtained for nuclei from <sup>3</sup>H to <sup>238</sup>U by different institutions were analyzed. It was shown that better agreement between various data is achieved if not Saclay data are multiplied by about 0.82 but if Livermore cross sections



FIG. 6. Comparison of the specially evaluated using normalized data [22] cross sections (open circles) and the normalized experimental [22] (open triangles) cross sections of the reactions on <sup>75</sup>As: (a)  $\sigma(\gamma, \text{tot})$ , (b)  $\sigma(\gamma, 1n)$ , and (c)  $\sigma(\gamma, 2n)$ .

Reaction	Normalized data [22]	Special evaluation	Normalized data [22]	Special evaluation
	$E^{\text{int}} = B2n = 18.2 \text{ MeV}$		$E^{\text{int}} = 26.2 \text{ MeV}$	
$(\gamma, Sn)^{a}$	$604.53 \pm 2.03$	$604.45 \pm 8.59$	$1370.40 \pm 5.21$	$1250.03 \pm 11.52$
$(\gamma, \text{tot})$	$603.95 \pm 2.07$	$604.45 \pm 8.59$	$1049.80 \pm 5.10$	$1067.67 \pm 10.23$
$(\gamma, 1n)$	$603.74 \pm 2.06$	$604.45 \pm 8.59$	$830.91 \pm 4.64$	$885.30 \pm 9.76$
$(\gamma, 2n)$	$0.13\pm0.23$		$218.69 \pm 2.12$	$182.36 \pm 3.06$

TABLE IV. Integrated cross sections  $\sigma^{\text{int}}$  (in MeV mb) of the photoneutron reaction cross sections for the <sup>75</sup>As specially evaluated using normalized  $\sigma^{\exp}(\gamma, Sn)$  [22] compared to the normalized experimental data [22].

<sup>a</sup>Experimental neutron yield cross section  $\sigma^{\exp}(\gamma, Sn)$  [23] used as the initial one for the evaluation procedure [Eq. (2)].

are multiplied by about 1.12. At the same time it was found that individual normalization factors for various nuclei have values from about 0.60 to about 1.80. For <sup>75</sup>As this factor is equal to 1.22.

The comparison of the initial experimental [22,23] neutron yield cross-section data for <sup>75</sup>As with corrected Livermore data [22] is presented in Fig. 5. The correspondent integrated cross-section and center of gravity values are presented in Table III. The energy range up to  $E^{\text{int}} = 22.4$  MeV was used for normalization.

One can see that after slightly shifting to high energies for 0.01 (17.41–17.40) MeV and multiplying by 1.25 (1035.20/829.41) the corrected Livermore cross section [22] became very close to the Saclay cross section [23] for all energies. Further, because the Livermore data correction energy shift was very small, the designation "normalized" will be used for corrected [22] data.

It is evident that using in the evaluation procedure [Eq. (2)] the normalized Livermore  $\sigma^{\exp}(\gamma, Sn)$  [22], which is very close to the Saclay one [23], will result in evaluated partial reaction cross sections very close to those evaluated before (Fig. 3 and Table II) using Saclay  $\sigma^{\exp}(\gamma, Sn)$ . But the normal-



ization  $\sigma^{\exp}(\gamma, Sn)$  [22] and correspondingly both  $\sigma^{\exp}(\gamma, 1n)$ and  $\sigma^{\exp}(\gamma, 2n)$  do not change the values of ratios  $F_i^{\exp}(\gamma, 2n)$  because this means multiplying both the numerator and the denominator in Eq. (4). Therefore after using the normalized Livermore  $\sigma^{\exp}(\gamma, Sn)$  [22] in the procedure [Eq. (2)] we obtained again noticeable differences between experimental and specially evaluated cross sections. In Fig. 6 the results of new evaluation are compared with the normalized experimental [22] cross sections  $\sigma^{\exp}(\gamma, 1n)$  and  $\sigma^{\exp}(\gamma, 2n)$ .

In Figs. 3 and 6 and Tables II and IV one can see that the cross sections evaluated using the normalized Livermore data [22] are naturally very close, within only several percent, to those obtained before using Saclay data [23].

From Fig. 6 and Table IV one can see that at energies up to B2n = 18.2 MeV the cross sections evaluated using normalized Livermore data [22] are very close to the normalized total and partial reaction experimental data [22]. But for the energy range 18.2-26.2 MeV significant disagreements exist.  $\sigma^{\text{eval}}(\gamma, 1n) = 280.85$  (885.30–604.45) MeV mb is 37% larger than  $\sigma^{\text{exp}}(\gamma, 1n) = 227.34$  (830.91–603.74) MeV mb but  $\sigma^{\text{eval}}(\gamma, 2n) = 182.36$  MeV mb is 20% smaller than  $\sigma^{\text{exp}}(\gamma, 2n) = 218.56$  (218.69–0.13) MeV mb.

In Fig. 7 the correspondent differences (6) between specially evaluated and normalized experimental [22] cross sections are presented.

From Fig. 7 one can see that in this case energy dependencies of cross section differences of  $\Delta\sigma(\gamma, 1n)$  and  $\Delta\sigma(\gamma, 2n)$ look absolutely different in comparison with the correspondent differences obtained previously [Fig. 4(b)]. Differences  $\Delta\sigma(\gamma, 1n)$  and  $\Delta\sigma(\gamma, 2n)$  appear reflected in a mirror in analogy to results of many other evaluations [8–10,13–21,24–26] and to differences obtained before using Saclay data [23]. At the same time  $\Delta\sigma(\gamma, 1n)$  and  $\Delta\sigma(\gamma, 2n)$  are not identical: In general values of  $\Delta\sigma(\gamma, 1n)$  are about 12 mb, but those of  $\Delta\sigma(\gamma, 2n)$  are about 7 mb. This could be explained by the loss of a large amount of neutrons, different for reactions  $\sigma(\gamma, 1n)$  and  $\sigma(\gamma, 2n)$ . On the basis of those comparisons one can conclude that even after additional normalization the experimental data [22] remained in general unreliable.

#### **V. SUMMARY**

FIG. 7. Comparison of the differences  $\Delta\sigma$  [Eq. (6)] between the cross sections specially evaluated using normalized [22] data and the normalized experimental [22] cross sections for <sup>75</sup>As [open triangles for reaction ( $\gamma$ , 1n); open circles for ( $\gamma$ , 2n)].

The objective physical data reliability criteria [10] were used to analyze systematic uncertainties presented in the experimental cross sections obtained for <sup>75</sup>As at Livermore [22] and Saclay [23]. The ratios  $F_i = \sigma(\gamma, in)/\sigma(\gamma, Sn)$  of the specific partial reaction cross sections to the neutron yield cross section, which is rather independent from experimental problems of neutron multiplicity sorting, were used as such criteria. The experimental-theoretical method [10] for evaluating the partial reaction cross sections  $\sigma^{\text{eval}}(\gamma, in) =$  $F_i^{\text{theor}}\sigma^{\exp}(\gamma, Sn)$ , based on the experimental neutron yield cross sections  $\sigma^{\exp}(\gamma, Sn)$  [23] and the ratios  $F_{I}^{\text{theor}}$  calculated in the combined photonuclear reactions model CPNRM [11,12], was used for evaluating the new cross sections for the  $(\gamma, 1n)$  and  $(\gamma, 2n)$  reactions on <sup>75</sup>As. It was shown that newly evaluated cross sections for both partial reactions are not so much different from the experimental Saclay data [23] but are significantly different from the experimental Livermore data [22]. It was shown that in analogy to the results of many previous investigations [8-10,13-21,24-26] the main reason for disagreements under discussion is that many neutrons were unreliably, erroneously, transmitted from one partial reaction to another because of significant systematic uncertainties of the procedure of determination neutron multiplicity based on its energy measuring.

Because of significant difference between experimental neutron yield cross sections  $\sigma^{\exp}(\gamma, Sn)$ , obtained at Saclay [23] and Livermore [22], the last one,  $\sigma^{\exp}(\gamma, Sn)$ , was additionally specially normalized before being used in the same evaluation procedure [Eq. (2)]. This  $\sigma^{\exp}(\gamma, Sn)$  obtained at Livermore [22] was slightly shifted in energy and multiplied by 1.25 to put it into consistency with the calculated  $\sigma^{\text{theor}}(\gamma, Sn)$ . The cross sections, specially evaluated using normalized Livermore data, are very close to those evaluated before using Saclay data. At the same time they are very different from the normalized experimental Livermore cross sec-

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tions for reactions  $(\gamma, 1n)$  and  $(\gamma, 2n)$ . One possible conclusion is that experimental Livermore data [22] are absolutely unreliable because of the presence of significant systematic uncertainties from unreliable transportation of many neutrons from one partial channel to another and additionally from the loss of many neutrons.

The results obtained confirm directly that the main reasons for the well-known disagreements between the partial photoneutron reactions cross sections, obtained in experiments under discussion, are definite shortcomings of the photoneutron multiplicity-sorting method. Therefore many experimental data obtained using this method should be reanalyzed and reevaluated individually and compared with new modern experimental data obtained using methods alternative to neutron-multiplicity sorting. Of most interest are the methods similar to the activation method using bremsstrahlung beams [19–21,28] and the novel technique of direct neutron multiplicity sorting with a flat-efficiency detector using monochromatic photon beams from laser-Compton scattering [29].

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