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Reliability of $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ cross-section data on ^{159}Tb

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The majority of partial and total photoneutron cross-section data were obtained using beams of quasimonoenergetic photons produced by annihilation in the flight of fast positrons and the method of neutron multiplicity-sorting procedures at Lawrence Livermore National Laboratory (California) and Saclay (France). Significant systematic disagreements between the two sets of data were obtained by employing the new objective physical data reliability criteria. It was found that many reaction cross sections are not reliable. As an example, a significant systematic uncertainty of the ^{159}Tb$(\gamma,2n)$ cross-section data measured at Livermore is presented. The $(\gamma,2n)$ reaction cross section was obtained as erroneous, whereas the $(\gamma,3n)$ reaction cross section was not obtained at all. The detailed discussion of this analysis is presented. The newly unmeasured before $(\gamma,3n)$ cross section is obtained from the experimental $(\gamma,2n)$ cross section using simple equations based on the physical criteria.

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I. INTRODUCTION

The majority of partial photonuclear reaction cross sections was obtained by using quasimonoenergetic annihilation photon beams [1,2] at Lawrence Livermore National Laboratory (L) (California) and Centre d'Etudes Nucleaires of Saclay (S) (France) and are included in the international nuclear reaction database [3]. The method of photoneutron multiplicity sorting was used in both laboratories to separate reactions with different numbers of neutrons. It is based on the assumption that the energy spectra of neutrons from partial photoneutron reactions $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ are radically different. Note that the thresholds (denoted below as $B1n$, $B2n$, and $B3n$, correspondingly) of the reactions mentioned above are relatively close to each other. Therefore there are wide ranges of the photon energies with competition of two or three partial reactions.

The cross sections of the reactions $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ were measured in the experiments mentioned, and then the total photoneutron reaction cross section,

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

and the neutron yield reaction cross section,

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

were obtained by summing the partial cross sections with different multiplicity factors.

The quite different methods to measure the kinetic energy of neutrons to determine the neutron multiplicity were used. Those are the so-called "ring-ratio" method at Livermore and the specifically calibrated large Gd-loaded liquid scintillator at Saclay. The well-known significant systematic data discrepancies were obtained for 19 nuclei from ^{51}V to ^{232}Th investigated in both laboratories. As a rule the $(\gamma,1n)$ reaction cross sections are noticeably larger at Saclay, and the $(\gamma,2n)$ cross sections vice versa at Livermore (up to 60%–100%). The examples of correspondent comparisons of Livermore and Saclay $(\gamma,2n)$ cross-section values obtained at various photon energies for four nuclei are presented in Table I.

It was shown [11] that for the 19 nuclei mentioned above for the integrated cross section,

$$\sigma^{\text{int}} = \int_B^{E^{\text{int}}} \sigma(E)dE, \qquad (3)$$

the average Saclay/Livermore ratios are $\langle\sigma_S^{\text{int}}/\sigma_L^{\text{int}}(\gamma,1n)\rangle = 1.08$ and $\langle\sigma_\text{S}^{\text{int}}/\sigma_\text{L}^{\text{int}}(\gamma,2n)\rangle = 0.83$. At the same time the average disagreements in the neutron yield reaction cross sections (2) are noticeably smaller (about 10%).

It was shown [11,12] that $\sigma(\gamma,Sn)'$s obtained at S and L for ^{159}Tb are very close to each other. The integrated cross-section ratio for $E^{\text{int}} = 27.4\,\text{MeV}$ is equal to $\sigma_\text{S}^{\text{int}}/\sigma_\text{L}^{\text{int}} = 3200/3170 = 1.01$. At the same time partial reaction cross sections are noticeably different: for $E^{\text{int}} = 27.4\,\text{MeV}, \sigma_\text{S}^{\text{int}}/\sigma_\text{L}^{\text{int}}(\gamma,1n) = 1950/1390 = 1.40$ but $\sigma_\text{S}^{\text{int}}/\sigma_\text{L}^{\text{int}}(\gamma,2n) = 610/870 = 0.70$. For ^{159}Tb the data under discussion are presented in Fig. 1.

One can see several negative cross-section values in the energy range from ~18.5 up to ~26.0 MeV. The doubts concerning $(\gamma,1n)$ reaction cross sections lead certainly to the doubts concerning the $(\gamma,2n)$ reaction cross sections.

The Livermore-Saclay disagreements were the subject of special studies, for example, Refs. [13–17]. The results produced by the neutron multiplicity-sorting method for ^{181}Ta were compared [13,14] with those obtained using the activation method, which allows one to identify partial reactions directly by detecting final nucleus deexcitation γ quanta. It was found that the $\sigma(\gamma,2n)$ reaction cross section obtained at Livermore agreed with the induced activity measurements, but the Saclay data were significantly underestimated [and correspondingly those for the $\sigma(\gamma,1n)$ were overestimated]. It has been suggested [13,14] that the difference in the partial reaction cross sections originated from the procedures used to separate counts into $1n$ and $2n$ events. It was concluded that the main reason was that some $(\gamma,2n)$ events were interpreted mistakenly as two $(\gamma,1n)$ events at Saclay. Therefore the

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TABLE I. Comparison of the $(\gamma,2n)$ cross-section values [3] obtained by L and S at various photon energies.

Nucleus	Cross-section value (mb) at photon energy (MeV, in brackets)		Reference
^{89}Y	19.38 (23.344)	19.68 (25.821)	[4], L
	10.60 (23.343)	18.00 (25.794)	[5], S
^{159}Tb	145.00 (17.614)	57.00 (26.750)	[6], L
	100.00 (17.600)	16.80 (26.840)	[7], S
^{181}Ta	196.00 (15.911)	86.00 (21.176)	[8], L
	156.00 (15.960)	50.70 (21.130)	[7], S
^{208}Pb	127.00 (16.995)	111.00 (25.202)	[9], L
	90.00 (17.050)	17.00 (25.260)	[10], S

Saclay data were declared as unreliable, but the Livermore data vice versa were reliable.

Unfortunately after more detailed investigations [11,12,18–20] the doubts concerning the Livermore data reliability appeared because of the very strange energy dependencies of the $\sigma(\gamma,1n)$ for many nuclei. Many negative experimental cross-section values in wide energy ranges were found.

These doubts contradict the conclusions that the Livermore data are reliable for both the $\sigma(\gamma,1n)$ and the $\sigma(\gamma,2n)$ mentioned above. Therefore we need objective criteria of data reliability.

II. ANALYSIS OF THE PARTIAL PHOTONEUTRON REACTION CROSS-SECTION DATA RELIABILITY USING THE PHYSICAL CRITERIA

The ratios of definite partial reaction cross sections to that of the neutron yield reaction,

$$F_i = \sigma(\gamma,in)/\sigma(\gamma,Sn) = \sigma(\gamma,in)/[\sigma(\gamma,1n) + 2\sigma(\gamma,2n) + 3\sigma(\gamma,3n) + \cdots] \quad (4)$$

were proposed [18,19] as the objective physical criteria of partial photoneutron reaction cross-section reliability. According to the definitions (4) $F_1 > 1.0$, $F_2 > 0.50$, $F_3 > 0.33$, etc., never can be. F_i values larger than the top limits mentioned mean that experimental sorting of neutrons between partial reactions has been carried out with large systematic uncertainties. Therefore obtained reaction cross sections are not reliable. Additionally it should be underlined that ratios F_i include only the cross-section terms and therefore should be definitely positive.

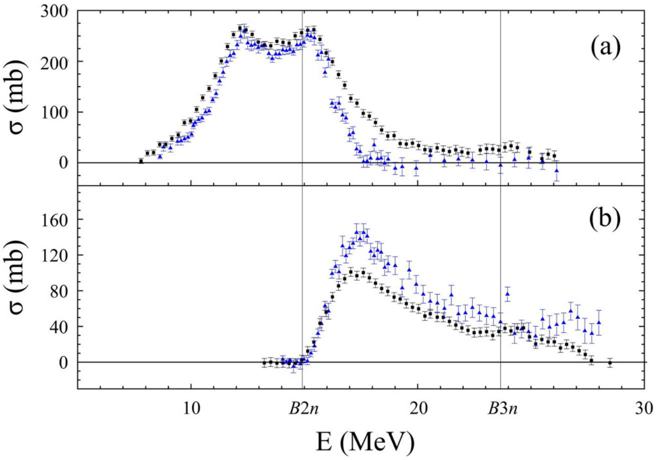

FIG. 1. The comparison of ^{159}Tb cross-section data obtained at Livermore ([6]: triangles) and Saclay ([7]: squares): (a) $\sigma(\gamma,1n)$ and (b) $\sigma(\gamma,2n)$.

In Refs. [11,12,18–20] by using the proposed criteria it was found that the experimental partial photoneutron reaction cross sections obtained using quasimonoenergetic annihilation photons for many nuclei (^{91,94}Zr, ^{115}In, $^{112-124}$Sn, ^{133}Cs, ^{159}Tb, ^{181}Ta, ^{186,188,189,190,192}Os, ^{197}Au, ^{208}Pb, . . .) are not reliable because in many photon energy regions they do not satisfy the proposed data reliability criteria. One can see that many F_1^{exp}'s are negative and/or F_2^{exp}'s are negative or larger than 0.50 and/or F_3^{exp}'s are negative or larger than 0.33, ...

The typical examples for ^{159}Tb are presented in Fig. 2 in comparison with the results of the calculations in the combined photonucleon reaction model (CPNRM) [21,22]. To compute theoretical photonuclear reactions cross sections this model is based on a semimicroscopic description of photon absorption and uses a combination of the Hauser-Feshbach evaporation model and preequilibrium mechanisms of nucleon emission. Additionally nuclear deformation and isospin splitting of the nucleus giant dipole resonance are properly accounted for by this calculation. The model is well tested for data for many medium and heavy nuclei.

In Fig. 2 one can see that energy dependencies of Saclay data $F_{1,2}^{\text{exp}}$ [7] have no values $F_1^{\text{exp}} > 1.0$ and $F_2^{\text{exp}} > 0.5$ correspondingly. But it could be pointed out that in general there are noticeable differences between $F_{1,2}^{\text{exp}}$ [7] and $F_{1,2}^{\text{theor}}$. Experimental data are overestimated clearly for $\sigma(\gamma,1n)$ and vice versa are underestimated for $\sigma(\gamma,2n)$ in comparison with the calculated data. Therefore the reliability of the Saclay data is definitely doubtful.

The energy dependencies of the Livermore data $F_{1,2}^{\text{exp}}$ [6] are much more interesting; first of all for the energy range of ∼18.5–26.0 MeV:

(1) here are several physically forbidden F_1^{exp} negative values;
(2) there are several F_2^{exp} values noticeably larger than 0.50 (up to 0.60);
(3) F_1^{exp} increases in correlation with the F_2^{exp} decrease and vice versa.

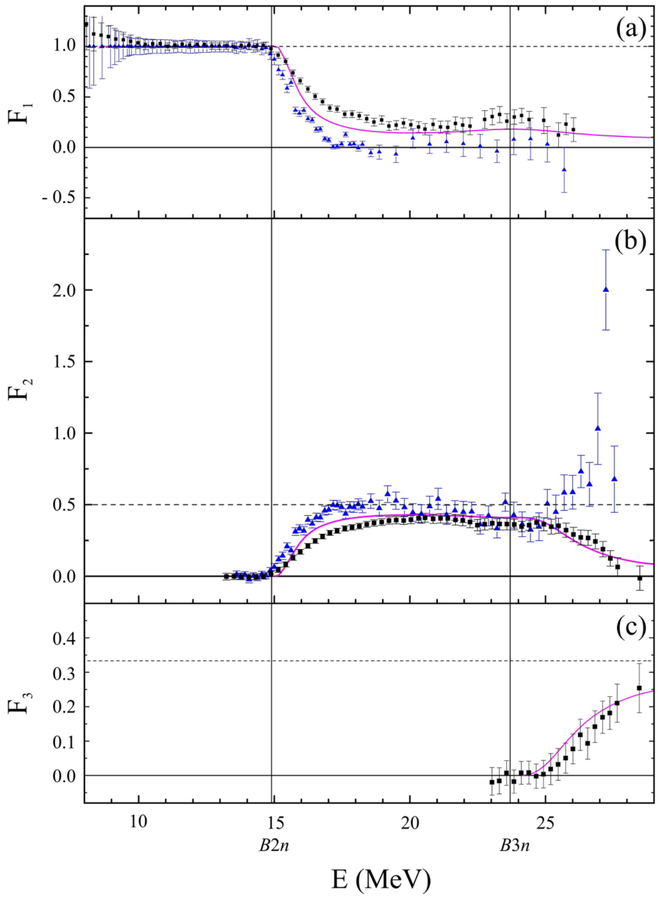

FIG. 2. (a) F_1^{exp}, (b) F_2^{exp}, and (c) F_3^{exp} data obtained for ^{159}Tb using the experimental data of Livermore ([6]: triangles) and Saclay ([7]: squares) in comparison with the results of calculated $F_{1,2}^{\mathrm{theor}}$ (model [21,22], lines).

But the most interesting are the significant disagreements among F_2^{exp} [6], F_2^{exp} [7], and F_2^{theor} [21,22] in the energy range higher than $E \sim 25$ MeV. In Fig. 1(b) one can see that in this energy range $\sigma_{\mathrm{L}}(\gamma,2n)$ is significantly larger versus $\sigma_{\mathrm{S}}(\gamma,2n)$. The astonishing feature is that the $\sigma(\gamma,3n)$ was determined at Saclay [7] but not at all at Livermore [6].

It should be pointed out that F_2^{exp} [6] does not decrease at energies higher than ∼24 MeV because of the appearance of the $\sigma(\gamma,3n)$ term in the denominator of (4). Moreover it conversely increases and comes into the region of physically unreliable values of $F_2 > 0.50$. The maximal value in this energy region $F_2^{\mathrm{exp}} = 2.0$ is exotic. It means that partial contribution $\sigma(\gamma,2n)$ is twice as large as the yield reaction cross-section $\sigma(\gamma,Sn)$! This indicates unambiguously that the sorting of neutrons with multiplicities 2 and 3 [6] was definitely incorrect. The only natural explanation [12] is that the $(\gamma,3n)$ reaction cross section was not determined [6] because all neutrons from it were attributed erroneously to the reaction $(\gamma,2n)$.

III. THE EXPERIMENTAL-THEORETICAL METHOD FOR THE PARTIAL REACTION CROSS-SECTION EVALUATING

The method for evaluating partial reaction cross sections not dependent on experimental neutron multiplicity sorting was proposed [18,19] to overcome the problems described above. It was used for obtaining the reliable data for the many nuclei mentioned above [11,12,18–20].

In this method the neutron yield cross-section $\sigma^{\mathrm{exp}}(\gamma,Sn)$ (1) is used only as initial experimental data. To separate it into the partial reaction cross sections the CPNRM [21,22] equations are used. Evaluated cross sections for each multiplicity ($i = 1{-}3,\ldots$) are evaluated using the following equation:

$$\sigma^{\mathrm{eval}}(\gamma,in) = F_i^{\mathrm{theor}}(\gamma,in)\sigma^{\mathrm{exp}}(\gamma,Sn). \tag{5}$$

This evaluating method means that the competitions of partial reactions are specified in accordance with equations of the model and the correspondent sum,

$$\sigma^{\mathrm{eval}}(\gamma,Sn) = \sigma^{\mathrm{eval}}(\gamma,1n) + 2\sigma^{\mathrm{eval}}(\gamma,2n) + 3\sigma^{\mathrm{eval}}(\gamma,3n) \tag{6}$$

is equal to the experimental cross-section $\sigma^{\mathrm{exp}}(\gamma,Sn)$.

For ^{159}Tb the evaluated $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ reaction cross sections were obtained and discussed [11,12] in detail. The Saclay $\sigma^{\mathrm{exp}}(\gamma,Sn)$ [7] was used as the initial experimental data for the evaluation procedure (5). The Saclay data were chosen because all three $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ reaction cross sections were obtained. As was mentioned above the $(\gamma,3n)$ reaction cross section was not determined at Livermore.

In full agreement with the noticeable differences between F_i^{exp} and F_i^{theor} (Fig. 2) the large differences between the experimental and the evaluated cross sections were found [11,12] in the energy range between $B2n$ and $B3n$. The integrated cross-section $\sigma^{\mathrm{int-eval}}(\gamma,1n)$ is about 20% smaller than the Saclay points [7] and 20% larger than the Livermore points [6], whereas $\sigma^{\mathrm{int-eval}}(\gamma,2n)$ is 15% larger than in Ref. [7] and 20% smaller as compared to Ref. [6]. So the difference between the evaluated and the experimental values [6,7] of the cross-section ratio $\sigma^{\mathrm{int-eval}}(\gamma,2n)/\sigma^{\mathrm{int-eval}}(\gamma,1n)$, important for various physical effect estimations, is about 30%.

IV. ESTIMATION OF NONMEASURED $\sigma(\gamma,3n)$ USING THE EXPERIMENTAL $\sigma(\gamma,2n)$

It was mentioned above that in the energy range above $B3n$, F_2^{exp}'s [6] have the exotic large values (up to 2.0!). This could be explained by only one natural assumption [12] that the cross section of the $(\gamma,3n)$ reaction was not determined because all neutrons from this reaction were attributed erroneously to the reaction $(\gamma,2n)$. For this case it is possible to estimate the correspondent $\sigma^{\mathrm{est}}(\gamma,3n)$ on the base of the experimental $\sigma^{\mathrm{exp}}(\gamma,2n)$ using the natural expression for the neutron yield reaction cross section,

$$\begin{aligned}\sigma^{\mathrm{exp}}(\gamma,Sn) = 2\sigma^{\mathrm{exp}}(\gamma,2n) &= \sigma^{\mathrm{eval}}(\gamma,1n) + 2\sigma^{\mathrm{eval}}(\gamma,2n)\\ &\quad + 3\sigma^{\mathrm{est}}(\gamma,3n),\end{aligned} \tag{7}$$

where $\sigma^{\mathrm{eval}}(\gamma,1n)$ and $\sigma^{\mathrm{eval}}(\gamma,2n)$ mean cross sections evaluated [11,12] using the experimental-theoretical method and data reliability criteria F_{1-3}.

After the natural transformation of (7),

$$\sigma^{\mathrm{est}}(\gamma,3n) = 2/3[\sigma^{\mathrm{exp}}(\gamma,2n) - \sigma^{\mathrm{eval}}(\gamma,2n)] - 1/3\sigma^{\mathrm{theor}}(\gamma,1n), \tag{8}$$

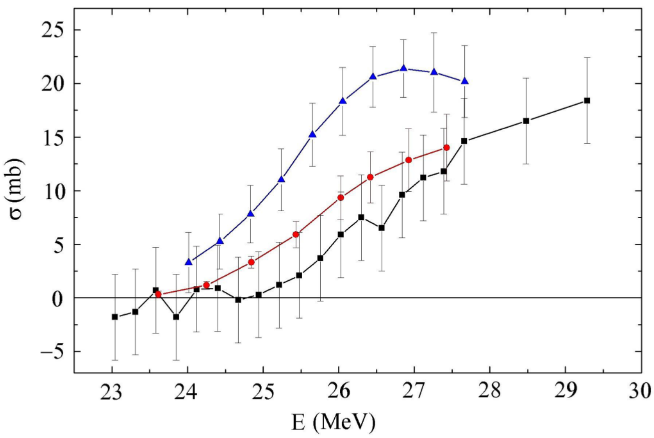

FIG. 3. Comparison of ^{159}Tb(γ,3n) ^{156}Tb reaction cross sections obtained by different methods: (1) experimental $\sigma^{\text{exp}}(\gamma,3n)$ Saclay data ([7]: squares); (2) evaluated $\sigma^{\text{eval}}(\gamma,3n)$ data ([11,12]: circles); (3) estimated $\sigma^{\text{est}}(\gamma,3n) = \sigma^{\text{exp}}(\gamma,3n)$ [6] data [(8): triangles].

the $\sigma^{\text{est}}(\gamma,3n)$ cross section can be estimated if the theoretically calculated [21,22] cross-section $\sigma^{\text{theor}}(\gamma,1n)$ would be used instead of $\sigma^{\text{eval}}(\gamma,1n)$. This $\sigma^{\text{est}}(\gamma,3n)$ estimated using the simple physical relations can be interpreted as the "measured at the first time" cross-section $\sigma^{\text{exp}}(\gamma,3n)$ that could be determined in Ref. [6] using the correct neutron multiplicity-sorting procedure.

The estimated cross-section $\sigma^{\text{est}}(\gamma,3n)$, in reality the unmeasured $\sigma^{\text{exp}}(\gamma,3n)$ [6], is presented in Fig. 3 in comparison with both experimental $\sigma^{\text{exp}}(\gamma,3n)$ [7] and evaluated $\sigma^{\text{eval}}(\gamma,3n)$ [11,12]. One can see that all three cross sections in general are similar to each other. The little disagreements are typical for those under discussion. One can see that $\sigma^{\text{est}}(\gamma,3n)$ is only slightly underestimated in comparison with both evaluated cross sections and $\sigma^{\text{exp}}(\gamma,3n)$ [7]. Therefore the result obtained could be the direct confirmation of significant systematic uncertainties of the experimental method of photoneutron multiplicity sorting. Such uncertainties could be the main reasons for well-known disagreements among the results of various experiments.

V. SUMMARY

It was found [11,12,18–20] that many experimental partial photoneutron reaction $(\gamma,1n)$, $(\gamma,2n)$, and $(\gamma,3n)$ cross sections obtained using the neutron multiplicity-sorting method [1–3] are not reliable. In many cases experimental data do not satisfy the objective physical criteria of reliability. One can see that many data contain physically forbidden negative values or values for which ratios $F_i = \sigma(\gamma,in)/\sigma(\gamma,Sn)$ are larger than the physical top limits (1.00,0.50,0.33, ... correspondingly for $i = 1-3, \ldots$). The reason is that many neutrons were transmitted erroneously from one reaction to another because of significant systematic uncertainties of the procedure of determination of neutron multiplicity based on its energy measuring. Using the experimental-theoretical method the reliable reaction cross sections satisfying reliability criteria were evaluated for many nuclei.

One very impressive example of significant systematic uncertainties of the data under discussion obtained at Livermore was found in the case of the data for ^{159}Tb [6]. The $(\gamma,2n)$ reaction cross section was obtained [6] definitely as erroneous because, in the energy range above $B3n$, F_2 has values up to 2. At the same time the cross section for the $(\gamma,3n)$ reaction was not determined at all. This case was investigated in detail. It was supposed that such F_2 values could appear only if all neutrons from the undetermined $(\gamma,3n)$ reaction were attributed erroneously to the reaction $(\gamma,2n)$. The estimation of the $\sigma^{\text{est}}(\gamma,3n)$ reaction was carried out using the simple physical relations. This estimated $\sigma^{\text{est}}(\gamma,3n)$ can be interpreted as the measured at the first time cross-section $\sigma^{\text{exp}}(\gamma,3n)$ [6]. In general it looks near both the correspondent evaluated $\sigma^{\text{eval}}(\gamma,3n)$ [11,12] and the experimental $\sigma^{\text{exp}}(\gamma,3n)$ [7].

The results obtained confirm directly that the main reasons of well-known disagreements among the partial photoneutron reaction cross sections obtained in various experiments under discussion are significant systematic uncertainties of the photoneutron multiplicity-sorting method. Therefore many experimental data obtained using this method should be reanalyzed and reevaluated individually.

Since the newly evaluated (and/or estimated) data noticeably differ from both Livermore and Saclay data [11,12,18–20], a discussion of the physical consequences is needed.

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