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Abstract. There are well-known systematic disagreements in partial photoneutron reaction cross sections obtained using quasimonoenergetic annihilation photons in experiments based on neutron multiplicity sorting methods. Using newly proposed criteria we demonstrate that a large part of the systematic uncertainty comes from certain shortcomings of experimental methods of neutron multiplicity sorting. To develop methods of correction of data obtained in experiments a new approach to data evaluation was developed in which a combined model of photonuclear reactions is used to decompose experimental total neutron yield reaction cross sections into partial reaction contributions. Evaluated cross sections of partial photoneutron reactions obtained using this method show a good agreement with results of alternative experiments.

1 Introduction

Measurements of cross sections of partial photoneutron reactions with different number of outgoing particles — primarily (γ, 1n), (γ, 2n), and (γ, 3n) — form an important body of experimental data that is widely used in both fundamental and applied research, including traditional studies of the Giant Dipole Resonance (GDR) and mechanisms of its excitation and decay (configurational and isospin splitting, competition between statistical and direct processes in GDR decay channels, sum rule exhaustion, etc.) as well as in various applications, such as beam luminosity monitoring in ultra-relativistic heavy-ion colliders [1]. Since the energy thresholds of the (γ, 1n), (γ, 2n), (γ, 3n), … reactions B₁ₙ, B₂ₙ, B₃ₙ, … are relatively close, there is a competition of two or three simultaneously open reaction channels in certain ranges of the incident photon energy. The majority of experiments in this field involved direct detection of outgoing neutrons and summed photoneutron yield cross sections were obtained (we use the notation from refs. [2,3] throughout the paper)

\[ \sigma(\gamma, Sn) = \sigma(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n) + \ldots, \]

where the multiplicity factors before the partial photoneutron reaction terms correspond to the number of detected neutrons. If some means to identify the reaction that produced the detected neutron is available, then \( \sigma(\gamma, Sn) \) can be decomposed into corresponding partial reaction cross sections. This is the well-known problem of neutron multiplicity sorting.

The photoneutron yield cross section is then used to obtain the total photoneutron cross section (combined cross section of all partial photoneutron reactions)

\[ \sigma(\gamma, \text{tot}) = \sigma(\gamma, 1n) + \sigma(\gamma, 2n) + \sigma(\gamma, 3n) + \ldots \]

\[ = \sigma(\gamma, Sn) - \sigma(\gamma, 2n) - 2\sigma(\gamma, 3n) - \ldots \] (2)

For medium and heavy nuclei the cross sections of reactions with outgoing protons are small and the total photoneutron cross section is close to the total photoabsorption cross section

\[ \sigma(\gamma, \text{abs}) = \sigma(\gamma, \text{tot}) + \sigma(\gamma, 1p) + \sigma(\gamma, 2p) + \ldots, \] (3)

which can be estimated with the help of the Thomas-Reiche-Kuhn sum rule:

\[ \sigma^{\text{int}}(\gamma, \text{tot}) \approx \sigma^{\text{int}}(\gamma, \text{abs}) = \int_0^\infty \sigma(E) dE \]

\[ = 60NZ/A \text{MeV} \cdot \text{mb}, \] (4)

where \( Z \) and \( N \) are, respectively, the numbers of protons and neutrons in the nucleus, and \( A = Z + N \) is the mass number.

Large-scale measurements involving direct neutron detection were performed in the 1960–80s at the National Lawrence Livermore Laboratory (USA) and France Centre...
d’Études Nucléaires de Saclay using quasimonoenergetic annihilation photon beams. Most of the neutron yield, total photoneutron and partial photonuclear reaction cross sections published in numerous reviews [2], atlases [3, 4], and databases [5] were obtained as part of these two research programs.

Both laboratories employed similar methods of determination of reaction multiplicities, based on the assumption that neutrons from ($\gamma$, 1n) and ($\gamma$, 2n) reactions can be identified by their spectra. However the methods of measurement of kinetic energies of neutrons were entirely different, which led to the well-known complex systematic discrepancies in partial photoneutron reaction cross sections: in many cases for the same nuclei the ($\gamma$, 1n) reaction cross sections were noticeably larger at Saclay, while the ($\gamma$, 2n) cross sections were in turn noticeably larger at Livermore. Figure 1 shows disagreements of this kind for 159Tb as a typical example.

For several years these disagreements were a subject of special studies [8–12]. About 500 photoneutron yield cross sections ($\gamma$, Sn) obtained by different institutions were analyzed in [11, 12], and integrated cross sections

$$\sigma^{\text{int}} = \int_{B_{1n}}^{B_{2n}} \sigma(E) dE,$$

were calculated for nuclei from 3H to 238U. In order to exclude possible effects of neutrons with different multiplicities the integration was performed for the incident photon energies from $B_{1n}$ to $B_{2n}$, effectively selecting only the ($\gamma$, 1n) contribution. Hence, differences of cross sections of the same reactions could be attributed only to systematic errors in normalization factors: detector efficiencies for neutrons with different energies, photon doses, cross sections of monitor reactions, and so on [10]. Uncertainties due to partial reaction separation procedures could not contribute in this energy range. Based on this, ratios

$$R^{\text{int \, syst}} = \sigma^{\text{int \, various}} / \sigma^{\text{int \, Livermore}},$$

of the experimental integrated cross sections measured elsewhere to the corresponding Livermore values were calculated (fig. 2).

Despite the apparent spread of the $R^{\text{int \, syst}}$ values, they generally exceed the 1.0 level: the average value $\langle R^{\text{int \, syst}} \rangle = 1.12 \pm 0.24$, i.e. the Livermore data points are on the whole slightly lower than the corresponding measurements from other experiments. It follows from the irregular behavior of $R^{\text{int \, syst}}$ that individual corrections have to be applied for each isotope before using the photoneutron yield cross sections for evaluation [11, 12].

Similar ratios were calculated for partial reaction cross sections. Figure 3 shows “Saclay/Livermore” ratios of integrated cross sections of partial photoneutron reactions (integrated from $B_{1n}$ to $B_{2n}$ and from $B_{2n}$ to $B_{3n}$, respectively):

$$R^{\text{int \, (1n)}} = \sigma^{\text{int \, S}}(\gamma, 1n)/\sigma^{\text{int \, L}}(\gamma, 1n),$$

$$R^{\text{int \, (2n)}} = \sigma^{\text{int \, S}}(\gamma, 2n)/\sigma^{\text{int \, L}}(\gamma, 2n),$$

calculated in [11] for 19 nuclei ($^{51}$V, $^{75}$As, $^{89}$Y, $^{90}$Zr, $^{115}$In, $^{116,117,118,120,124}$Sn, $^{127}$I, $^{133}$Cs, $^{159}$Tb, $^{165}$Ho, $^{181}$Ta, $^{168}$Tm, $^{197}$Au).
The proposed method of evaluation of partial photoneutron reaction cross sections $\sigma(\gamma, i\nu)$ relies only on the photoneutron yield cross sections (1) which were not contaminated with systematic uncertainties due to neutron multiplicity sorting, in contrast with the photoabsorption cross sections or other secondary quantities. The evaluated partial reaction cross sections are obtained via the following decomposition of the experimental neutron yield cross section $\sigma_{\text{exp}}(\gamma, i\nu)$:

$$\sigma_{\text{eval}}(\gamma, i\nu) = F_{1-\text{th}} \sigma_{\text{exp}}(\gamma, i\nu)$$

$$= \left[ \sigma_{\text{th}}(\gamma, i\nu) / \sigma_{\text{th}}(\gamma, i\nu) \right] \sigma_{\text{exp}}(\gamma, i\nu),$$

$$\sigma_{\text{eval}}(\gamma, 2n) = F_{2-\text{th}} \sigma_{\text{exp}}(\gamma, 2n)$$

$$= \left[ \sigma_{\text{th}}(\gamma, 2n) / \sigma_{\text{th}}(\gamma, 2n) \right] \sigma_{\text{exp}}(\gamma, 2n), \ldots,$$

where $F_{\text{th}}$ are the so-called transitional neutron multiplicity functions

$$F_{\gamma-\text{th}} = \sigma_{\text{th}}(\gamma, i\nu) / \sigma_{\text{th}}(\gamma, i\nu)$$

$$= \sigma_{\text{th}}(\gamma, 1n) / \sigma_{\text{th}}(\gamma, 1n) + 2(\gamma, 2n) + 3(\gamma, 3n) + \ldots,$$

which are calculated numerically using the combined photoneutron reaction model code [15–17]. The calculation is based on a semi-microscopic description of photon absorption and uses a combination of the Hauser-Feshbach evaporation model and pre-equilibrium mechanisms of nucleon emission to compute theoretical cross sections $\sigma_{\text{th}}$ of photoneutron reactions. In addition, characteristically photoneutron effects such as nuclear deformation and isospin splitting of the GDR are properly accounted for by this calculation.

The reliability of the photoneutron yield cross sections $\sigma(\gamma, i\nu)$ is essential for the quality of evaluation. Within the described approach the available experimental photoneutron yield cross sections are jointly evaluated before splitting into partial cross sections in order to prepare a single input dataset. This step is described with more detail in ref. [12]. The initial data preparation is performed on an individual basis, depending on the available experimental measurements on the specific isotope, and it generally includes normalization by a discrepancy factor, energy grid rescaling, and comparison with model calculations. Typically, after this evaluation the closest matching experimental photoneutron yield cross section is selected. When the only available measurement is the Livermore one, the average discrepancy factor $\langle R_{\text{int} \text{sys}} \rangle = 1.12$ is applied.

The results of initial evaluation and a comparison with the theoretically calculated photoneutron yield cross sections for $^{150}$Tb are shown in fig. 4. Saclay data [7] are
presented without correction, Livermore data [6] were multiplied by 1.06, Moscow data [18] multiplied by 0.83, theoretical data [15–17] multiplied by 0.93. The multipliers were obtained for this nucleus similarly to (6), i.e. from the comparison of the integrated cross sections with each other. Based on this evaluation the Saclay data were used for the following partial cross section evaluation procedures.

2.1 Criteria of the systematic uncertainties

In a series of works [16,17,19–22] on the example of the photoneutron reaction cross sections on the $^{90,91,94}$Zr [23], $^{115}$In, $^{112,114,116,117,118,119,120,122,124}$Sn [24], $^{159}$Tb [6], $^{181}$Ta [25], $^{188,189}$Os [26], $^{208}$Pb [27] isotopes it was shown that the theoretically calculated functions $F_{1,2,3}$ can be used to detect systematic uncertainties in partial reaction cross sections. In this work we further illustrate this approach for $^{94}$Zr, $^{159}$Tb, $^{181}$Ta, and $^{118}$Sn.

According to the definition (11) $F_{1\text{-th}}$ can never be greater than 1.00 since all cross sections $\sigma_{\text{exp}}(\gamma, i\text{n}) > 0$; similar inequalities hold for other multiplicity functions: $0 \leq F_{2\text{-th}} \leq 1/2 = 0.50$, $0 \leq F_{3\text{-th}} \leq 1/3 = 0.33$, and so on. Figures 5(a),(b) show theoretical functions $F_{1\text{-th}}$ and $F_{2\text{-th}}$ calculated for $^{94}$Zr in comparison with the multiplicity functions calculated from the Livermore data [23].

One would expect from definition (11) that the energy dependence of $F_{1,2\text{-th}}$ should have the following properties:

- below the $(\gamma, 2n)$ reaction threshold $B_{2n}$ the only open reaction channel is $(\gamma, 1\text{n})$ and therefore $F_{1\text{-th}} = 1$, $F_{2\text{-th}} = 0$;
- above $B_{2n}$ both $(\gamma, 1\text{n})$ and $(\gamma, 2n)$ reactions are possible. $F_{1\text{-th}}$ increases approaching the theoretical limit of 0.50, but never reaching it because of a high-energy part in $\sigma_{\text{th}}(\gamma, 1\text{n})$;
- above the $B_{3n}$ threshold the $(\gamma, 3\text{n})$ reaction is possible, $F_{2\text{-th}}$ decreases due to the $3\sigma_{\text{th}}(\gamma, 3\text{n})$ term in the denominator of (11).

However, the actual behavior (fig. 5(a),(b)) of the experimental functions $F_{1\text{-exp}} = \sigma_{\text{exp}}(\gamma, i\text{n})/\sigma_{\text{exp}}(\gamma, \text{Sn})$ obtained using the measured cross sections of reactions on $^{94}$Zr [23], exhibits obvious differences from the expected theoretical behavior of $F_{1\text{-th}}$: in the energy range 21.5–28.0 MeV $F_{1\text{-exp}}$ takes physically forbidden negative values and $F_{2\text{-exp}} > 0.50$. Although the errors in the energy range of multiple neutron emission are large, there is an apparent systematic difference between the theoretical and experimental values. The observed correlation can be a result of incorrect multiplicity sorting which was the reason of an excess of reported multiplicity-two neutrons and a corresponding lack of multiplicity-one neutrons.

2.2 Evaluated partial photoneutron reaction cross sections

Figure 6 shows evaluated cross sections of $(\gamma, 1\text{n})$ and $(\gamma, 2n)$ on $^{159}$Tb, obtained using procedures (9)–(11), in comparison with experimental data from refs. [6,7]. The evaluated cross sections were calculated using the Saclay experimental $\sigma_{\text{exp}}(\gamma, \text{Sn})$ [7], which was selected after the initial step of photoneutron yield cross section evaluation [19].

In the energy range between $B_{2n}$ and $B_{3n}$ there are large differences between experimental results and evaluation: $\sigma_{\text{eval}}(\gamma, 1\text{n})$ is about 20% smaller than the Saclay measurements [7] and 20% bigger than the Livermore points [6], whereas $\sigma_{\text{eval}}(\gamma, 2n)$ is 15% bigger than [7] and 20% smaller as compared to [6]. The difference between the evaluated and experimental values [6,7] of the important cross section ratio $\sigma_{\text{eval}}(\gamma, 2n)/\sigma_{\text{eval}}(\gamma, 1\text{n})$ is about 30%. Above the $B_{3n}$ threshold the differences between experimental and evaluated cross sections are even larger.

It should be noted that experimental partial photoneutron reaction cross sections are eventually used to obtain total photoneutron reaction cross sections (2) and photoabsorption cross sections (3), and, therefore, their values are also affected by the described discrepancies.
To make the comparison special measurements were performed using the bremsstrahlung photon beam produced by the 67.7 MeV race-track microtron of the Skobeltsyn Institute of Nuclear Physics [28]. Reaction yields

\[ Y(E_{\text{max}}) = k \int_{E}^{E_{\text{max}}} W(E_{\text{max}}, E) \sigma(E) dE, \]  \tag{12}  

(where \( W(E_{\text{max}}, E) \) is the energy spectrum of bremsstrahlung radiation with end-point energy \( E_{\text{max}} = 67.7 \text{ MeV} \) and \( B \) is the corresponding reaction threshold) were measured for reactions \((\gamma, \text{1n})-(\gamma, \text{7n})\) on \(^{181}\text{Ta}\). In table 1 corresponding experimental ratios of yields of pairs of reactions \(Y(\gamma, \text{2n})/Y(\gamma, \text{1n})\) and ratios of the integrated cross sections \(\sigma^{\text{int}}(\gamma, \text{2n})/\sigma^{\text{int}}(\gamma, \text{1n})\) are compared to the results of evaluation, published in preprint [20]. For this evaluation the Saclay data were selected during the initial photoneutron yield cross section analysis step. To obtain the reaction yields corresponding to the experiments with quasimonochromatic photons the cross sections were folded with a simulated bremsstrahlung spectrum calculated using GEANT4 [29].

The presented results clearly show that the Saclay (0.34) and the Livermore ratios are overestimated (0.67) in comparison with the evaluation (0.49). The same inconsistency can be seen for the ratios of yields: 0.24 and 0.42 versus 0.33. The values obtained using the proposed evaluation method agree well with the results of the activation experiment (0.33 versus 0.34).

3.2 Comparison with results of contemporary experiments with quasimonochromatic laser-Compton scattering photons

New advanced gamma-ray sources using laser-Compton scattering have recently become available to nuclear physics. Quasimonochromatic photons are used in studies of photonuclear reactions in different energy ranges, and, in particular, to measure accurate \((\gamma, \text{1n})\) cross sections near threshold. A large amount of data was obtained with this technique at the National Institute of Advanced Industrial Science and Technology (Japan) [30], including reaction cross sections on the nuclei that were previously studied in our evaluations.

Figure 7 shows the comparison of the \(^{118}\text{Sn}(\gamma, \text{1n})^{117}\text{Sn} \) reaction cross section measured using laser-Compton scattering [30] with quasimonochromatic annihilation photon cross sections [24,31] and our evaluation [13]. The initial evaluated photoneutron yield cross section \(\sigma^{\text{exp}}(\gamma, \text{Sn})\) in this case was calculated as a weighted average of experimental measurements [24,31] (quasimonochromatic photons) and [32] (bremssstrahlung). The results of evaluation are in agreement with the results of the laser-Compton scattering experiment while there is a clear difference in the case of the quasimonochromatic annihilation photon experiments: the Saclay [31] points are significantly underestimated, and the Livermore [24] points are slightly below the evaluation. The

3 Experimental verification of evaluated data

The proposed evaluation procedure based on criteria (11) and corrections (9), (10) can be used to reduce systematic uncertainties of the partial reaction cross sections obtained using neutron multiplicity sorting methods. In the following section we compare the evaluated cross sections with results of experiments that are presumably free from the systematic errors of the neutron multiplicity sorting method.

3.1 Comparison with results of photon activation experiment

One of the alternative experimental techniques is the photon activation method in which the studied reaction is identified by the residual radioactivity of the produced nucleus. In the photon activation technique gamma-ray spectra of final nuclei are measured with a high resolution spectrometer and thus yields of partial reactions can be obtained directly.

Fig. 6. Comparison of evaluated ([19], dots) and experimental (Livermore [6], triangles and Saclay [7], squares) photoneutron reaction cross sections for \(^{159}\text{Tb}\): (a) \(\sigma(\gamma, \text{1n})\); (b) \(\sigma(\gamma, \text{2n})\). See also fig. 1(b),(c).
Table 1. Comparison of ratios of reaction yields \(Y\) and integrated cross sections \(\sigma^{\text{int}}\) obtained for experimental (according to [3]) and evaluated data for \(^{181}\text{Ta}\) for \(E_{\gamma}^{\text{int}} = 67.7\text{ MeV}\).

<table>
<thead>
<tr>
<th>Ratio</th>
<th>Experiment</th>
<th>Evaluation</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\sigma^{\text{int}}(\gamma, 2n)/\sigma^{\text{int}}(\gamma, 1n))</td>
<td>Saclay [7]</td>
<td>Livermore [6]</td>
</tr>
<tr>
<td>(0.36 (4))</td>
<td>0.67 (7)</td>
<td>0.49 (5)</td>
</tr>
<tr>
<td>(Y(\gamma, 2n)/Y(\gamma, 1n))</td>
<td>0.24 (3)</td>
<td>0.42 (4)</td>
</tr>
</tbody>
</table>

above made point about the difference between the Livermore and Saclay data in the energy range below \(E_{2n}\) is still valid in this case, but now it is most probably due to overestimation of Saclay data, and not to underestimation of Livermore’s.

4 Possible reasons of difference between experimental data and evaluations

As it has been noted above, the neutron multiplicity sorting methods employed by large-scale photodisintegration experiments were based on the assumption that the energy of the single neutron originating from the \((\gamma, 1n)\) reaction is noticeably greater than the energies of the \((\gamma, 2n)\) reaction neutrons.

It can be seen from figs. 4–6, however, that the degree of discrepancies between partial photoneutron cross-sections obtained in quasimonoenergetic annihilation photon experiments in the \(1n\), \(2n\), and \(3n\) channels depends on the photon energy, as do the energy spectra of outgoing neutrons. Thus, the relationship between the energy of a neutron and its multiplicity is more complex than a simple threshold.

A special study showed that the mean energy of the first neutron from the reaction \(^{181}\text{Ta}(\gamma, 2n)\) is positively larger than that of the second neutron \((\text{e.g., when the photon energy is } 25\text{ MeV})\) the mean energy of the 1st neutron is 4.0 MeV, and the one of the 2nd neutron is 1.4 MeV) [28]. Theoretical calculations show that the shapes of the energy spectra of neutrons in the \((\gamma, 1n)\) reaction at \(E_{\gamma} = 12.2\text{ MeV}\) and \((\gamma, 2n)\) at \(E_{\gamma} = 19.2\text{ MeV}\) are close in the case of \(^{159}\text{Tb}\) and \(^{181}\text{Ta}\).

In addition it must be stressed that due to the method of direct detection of outgoing neutrons in past experiments with quasimonoenergetic annihilation photons the proton channels were not considered at all. In many cases the reported value \(\sigma^{\text{exp}}(\gamma, 1n)\) in these experiments was in fact the sum \(\sigma^{\text{exp}}(\gamma, 1n) + \sigma^{\text{exp}}(\gamma, 1np)\), which represented the sum \(\sigma^{\text{exp}}(\gamma, 2n) + \sigma^{\text{exp}}(\gamma, 2np)\), and so on. That could seriously distort the dependence of neutron multiplicity on its kinetic energy. One possible source of ambiguity in this case comes from the distribution of reaction energy among the outgoing particles: if in the \((\gamma, np)\) reaction the energy is shared between the nucleons similarly to \((\gamma, 2n)\), there remains only one way to distinguish between them—by the number of detected neutrons. Due to the latter complex detector geometries, multiple scattering of neutrons and resulting variations of their energy, background conditions, and other factors there were rather large uncertainties of the numbers of detected neutrons. Reference [2] notes “a much higher background rate [of the Saclay detector], made up largely of single-neutron events (from our analysis—evidently overestimated), which introduces larger uncertainties in the background subtraction and pile-up corrections.”

Our approach to evaluation of partial photoneutron reaction cross sections properly takes into account the proton decay channels since the \(\sigma^{\text{th}}\) value used for determination of the \(F_{1,2,3}\) functions explicitly includes \(\sigma(\gamma, 1np)\):

\[
\sigma^{\text{th}}(\gamma, Sn) = \sigma^{\text{th}}(\gamma, 1n) + \sigma^{\text{th}}(\gamma, 1np) + 2\sigma^{\text{th}}(\gamma, 2n) \\
+ 3\sigma^{\text{th}}(\gamma, 3n) + \ldots
\]

This leads to the conclusion that one of the main reasons of disagreements between experimental cross sections and our evaluations is a very complex and indirect relationship between neutron kinetic energies and reaction multiplicities.

5 Summary and conclusions

New criteria \(F_{i} = \sigma(\gamma, in)/\sigma(\gamma, Sn)\) of systematic uncertainties in photoneutron partial reaction cross sections...
were proposed and a new method was developed for evaluation of partial reaction cross sections. Experimental neutron yield reaction cross sections $\sigma_{exp}(\gamma,\text{Sn})$ that do not depend on shortcomings of the neutron multiplicity sorting methods are decomposed into partial reaction cross sections using the combined photonuclear reaction model [15–17].

On the example of results obtained for the isotopes $^{94}\text{Zr}$, $^{118}\text{Sn}$, $^{159}\text{Tb}$, and $^{181}\text{Ta}$ it is shown that evaluated cross sections do not agree with the data obtained in quasi-monoenergetic annihilation photon experiments with neutron multiplicity sorting methods but agree with results obtained using the activation technique.

The results suggest that a significant part of experimental data on partial photoneutron reaction cross sections has to be re-analyzed and/or re-evaluated.

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References

11. V.V. Varlamov, N.N. Peskov, D.S. Rudenko, M.E. Stepanov, Consistent Evaluation of Photoneutron Reaction Cross-sections Using Data Obtained in Experiments with Quasimonoenergetic Annihilation Photon Beams at Livermore (USA) and Saclay (France) (IAEA NDS, Vienna, Austria, 2004) p. 37 INDC(CCP)-440.