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New Data on Cross Sections for Partial and Total Photoneutron Reactions on the Isotopes ^{91,94}Zr

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Abstract—Experimental data on ^{91,94}Zr photodisintegration that were obtained in a beam of quasi-monoenergetic annihilation photons by the method of neutron multiplicity sorting are analyzed. It is found that the cross sections for the $(\gamma, 1n)$, $(\gamma, 2n)$, and $(\gamma, 3n)$ reactions on both isotopes do not meet the objective data-reliability criteria formulated earlier. Within the experimental—theoretical method for evaluating partial-reaction cross sections that satisfy these criteria, new data on the cross sections for the aforementioned partial reactions, as well as for the $(\gamma, sn) = (\gamma, 1n) + (\gamma, 2n) + (\gamma, 3n) + \ldots$ total photoneutron reaction, are obtained for the isotopes ^{91,94}Zr.

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1. INTRODUCTION

Experimental data on the photodisintegration of a large number of medium-mass and heavy nuclei (such as ^{90}Zr, ^{115}In, ^{112,114,116,117,118,119,120,122,124}Sn, ^{159}Tb, ^{181}Ta, and ^{197}Au) studied in beams of quasi-monoenergetic annihilation photons in [1–5] by the method of photoneutron multiplicity sorting, the multiplicities of photoneutrons being determined from their measured kinetic energies were analyzed. It was found that, as a rule, the cross sections for the $(\gamma, 1n)$, $(\gamma, 2n)$ and $(\gamma, 3n)$ partial reactions were determined with large systematic errors, so that their reliability may be questioned.

The circumstances that are indicative of the presence of such errors are primarily the following:

(i) In many regions of photon energies, the specially introduced neutron-multiplicity transition functions

$$F_i = \sigma(\gamma, in)/\sigma(\gamma, xn) \quad (1)$$
$$= \sigma(\gamma, in)/\sigma[(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n) + \ldots]$$

exceed limits physically allowed for them by definition (1.00, 0.50, 0.33,… for, respectively, $i = 1, 2, 3, \ldots$).

(ii) In the same regions of photon energies, the cross sections for the various reactions—mainly for $(\gamma, 1n)$—take negative values, which are forbidden physically.

It was found that values established for the systematic errors are associated with shortcomings in photoneutron-sorting methods used in the quoted experiments and based on the results of measurements of photoneutron kinetic energies.

In order to obtain reaction cross sections that are free from the aforementioned problems and which satisfy criteria of data reliability, an experimental—theoretical method for evaluating them was proposed in [1, 2]. This method is based on employing, in the evaluation procedure, only experimental neutron-production cross sections

$$\sigma(\gamma, xn) \approx \sigma(\gamma, 1n) + 2\sigma(\gamma, 2n) + 3\sigma(\gamma, 3n) + \ldots, \quad (2)$$

which are independent of problems of neutron multiplicity sorting, and underlying principles of the combined photonuclear-reaction model proposed in [6, 7], which are also independent of these problems. The preequilibrium exciton model that employs nuclear level densities calculated on the basis of the Fermi gas model and which takes into account the deformation of the nucleus being considered and the isospin splitting of its giant resonance (for a detailed description of this model, we refer the interested reader to [8]) makes it possible to obtain successful approximations to the neutron-production cross sections (2) in the region of medium-mass and heavy nuclei.

Within this experimental—theoretical approach, the evaluated partial-reaction cross sections

$$\sigma^{\text{eval}}(\gamma, in) = F_i^{\text{theor}} \sigma^{\text{expt}}(\gamma, xn), \quad (3)$$

are determined by the ratios F_i^{theor} (1) calculated on the basis of the model used, and the respective

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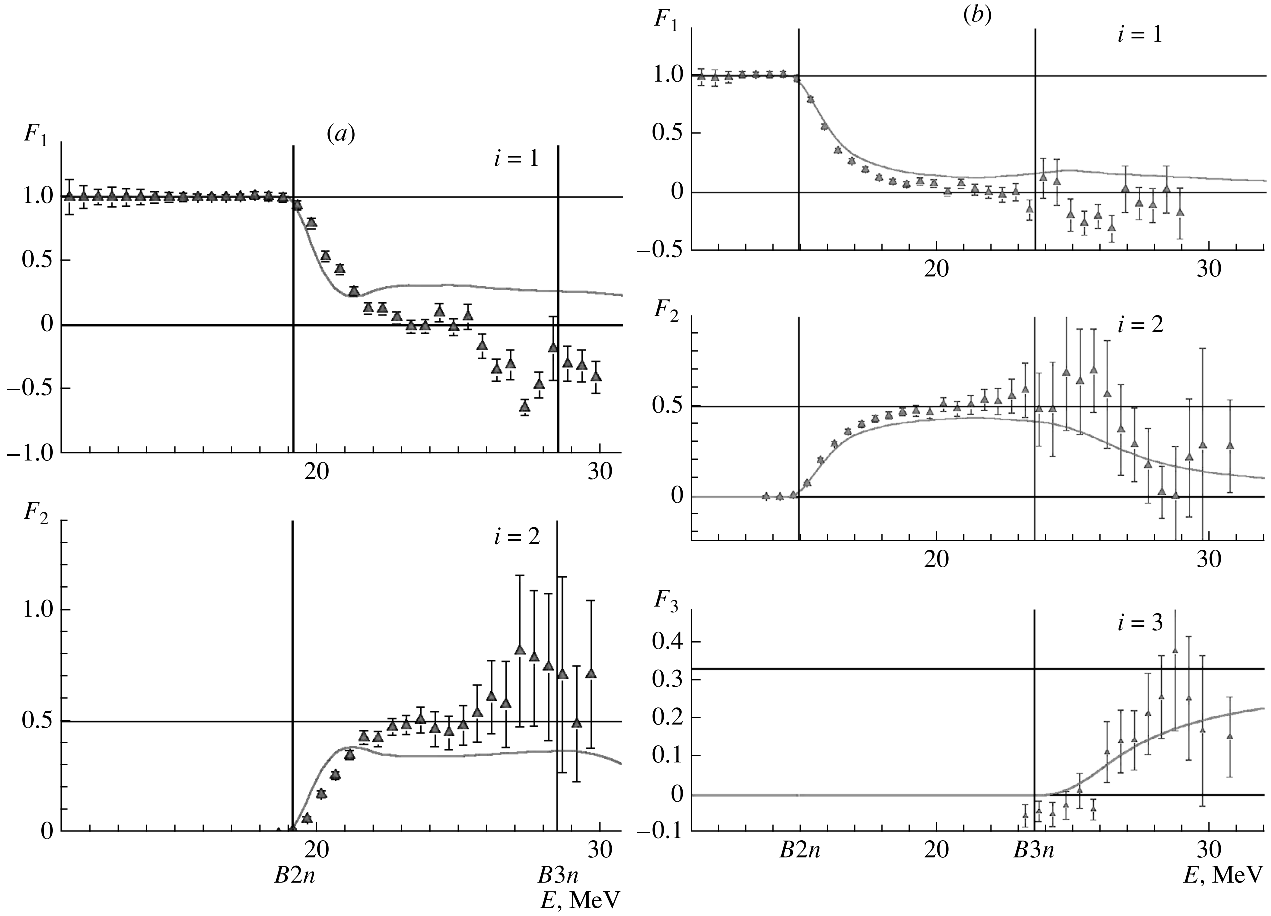

Fig. 1. Multiplicity transition functions $F_i^{\rm expt}$ (for $i = 1, 2, 3$) obtained from experimental data reported in [9] (triangles) along with the functions $F_i^{\rm theor}$ obtained on the basis of the results of theoretical calculations performed in [6–8] (curves) for the isotopes (*a*) ^{91}Zr and (*b*) ^{94}Zr.

sum in (2) agrees with the experimental cross section $\sigma^{\rm expt}(\gamma, xn)$

In the present study, this approach is used to evaluate the cross sections for the $(\gamma, 1n)$, $(\gamma, 2n)$, and $(\gamma, 3n)$ partial and the $(\gamma, sn) = (\gamma, 1n) + (\gamma, 2n) + (\gamma, 3n) + \ldots$ total photoneutron reactions for the isotopes ^{91,94}Zr, in which case the inconsistency of the experimental data from [9] with the reliability criterion in (1) is quite distinct.

2. ANALYSIS OF RELIABILITY OF EXPERIMENTAL DATA ON THE BASIS OF NEUTRON-MULTIPLICITY TRANSITION FUNCTIONS F_i

Figure 1 clearly demonstrates the inconsistency of experimental data for the isotopes ^{91,94}Zr with the physical reliability criteria. For the two zirconium isotopes discussed in the present study, this figure illustrates a comparison of the energy dependences of the neutron-multiplicity transition functions $F_i^{\rm theor}$ (1) calculated in [6–8] on the basis of the combined photonuclear-reaction model (curves) with the energy dependences of the functions $F_i^{\rm expt}$ (1) obtained from data in [9] (triangles with error bars).

From Fig. 1, one can clearly see that, according to the definition in (1), the energy dependences of the functions $F_i^{\rm theor}$ have the following form:

(i) Only the $(\gamma, 1n)$ reaction is possible at photon energies below the threshold $B2n$ for the $(\gamma, 2n)$ reaction; therefore, $F_1^{\rm theor} = 1$, $F_2^{\rm theor} = F_3^{\rm theor} = 0$.

(ii) At photon energies between $B2n$ and $B3n$, $F_1^{\rm theor}$ decreases in accordance with the competition between decreasing $\sigma(\gamma, 1n)$ and increasing $\sigma(\gamma, 2n)$, while $F_2^{\rm theor}$ increases in accordance with the same competition between $\sigma(\gamma, 2n)$ and $\sigma(\gamma, 1n)$ and approaches from below the limit of 0.50 motivated physically [see Eq. (1)], reaching it nowhere.

(iii) At photon energies above the energy threshold $B3n$, the function $F_2^{\rm theor}$ decreases in view of the appearance of the term $3\sigma(\gamma, 3n)$ in expression (1).

Figure 1 also clearly shows that the energy dependences of the functions F_i^{expt} and F_i^{theor} are markedly different. In the region of energies above about 25.5 MeV, the ratios F_1^{expt} for the isotope ^{91}Zr are in the region of physically forbidden negative values, obviously going beyond the errors, while the ratios F_2^{expt} take physically unreliable values in excess of the limit of 0.50. This shows compellingly that the experimental sorting of multiplicity-1 and multiplicity-2 neutrons was performed incorrectly in [9].

The situation around the isotope ^{94}Zr is more complicated in view of larger errors, but the conclusion that the experimental neutron multiplicity sorting was performed incorrectly can drawn definitively in that case inclusive.

By way of example, we indicate that, since, at energies higher than some 22.2 MeV, there are physically forbidden negative value among the ratios F_1^{expt} (with the exception of two values at energies of about 24.0 to 24.5 MeV), a negative area of some part under the curve of $F_1^{\text{expt}}(E)$,

$$\int_{22.2}^{29.0} F_1(E)dE = -0.55 \pm 0.26 \text{ MeV}, \quad (4)$$

along with a negative value of the integrated cross section for the respective $(\gamma, 1n)$ reaction in the same energy region,

$$\int_{22.2}^{29.0} \sigma(E)dE = -19.05 \pm 14.94 \text{ MeV mb}, \quad (5)$$

casts some doubt on the reliability of this determination of the $(\gamma, 1n)$ cross section.

In the energy range between about 20.3 and 26.0 MeV, the ratio F_2^{expt} exhibits values exceeding the limit of 0.50; in the case of the experimental data from [9], the calculated area under the curve $F_2^{\text{expt}}(E)$ for the region in question is

$$\int_{20.3}^{26.0} F_2(E)dE = 3.20 \pm 0.38 \text{ MeV}, \quad (6)$$

while, in the case of the calculated data, it is 2.30 ± 0.08 MeV, the physically reliable limit being $0.5 \times [26.0 - 20.3] = 2.85$ MeV according to the definition in Eq. (1). Since the function F_2^{expt} exceeds substantially this limit, a determination of the $(\gamma, 2n)$ cross section seems questionable.

In addition, it is noteworthy that, over the entire energy region, the function F_2^{expt} increases, but, according to the definition in (1), it should decrease above the threshold of $B3n = 23.6$ MeV.

It should also be noted that, for both of the isotopes ^{91,94}Zr considered here, the reliability of data in the region of lower energies is not beyond doubt either since there are significant discrepancies between the values of F_1^{expt} and F_1^{theor}, as well as between the values of F_2^{expt} and F_2^{theor}.

All of the foregoing indicates that the experimental neutron mutiplicity sorting was performed incorrectly in [9] for the multiplicities of one, two, and three.

3. EVALUATING PARTIAL-PHOTONEUTRON-REACTION CROSS SECTIONS FREE FROM PROBLEMS OF NEUTRON MULTIPLICITY SORTING

Data for the Isotope ^{91}Zr

As was indicated above, an evaluation of partial-reaction cross sections that are free from the drawbacks of the experimental methods of neutron multiplicity sorting relies on respective relations of the theoretical model proposed in [6–8] that determine the breakdown of the experimental neutron-production cross section (2) into contributions corresponding different numbers of neutrons. At the preliminary stage of evaluating partial-reaction cross sections, the experimental and theoretical neutron-production cross sections should therefore be matched with each other to the maximum possible degree.

For the ^{91}Zr nucleus, the experimental neutron yield cross section $\sigma^{\text{expt}}(\gamma, xn)$ agrees reasonably well with the theoretical cross section $\sigma^{\text{theor}}(\gamma, xn)$ calculated on the basis of the model proposed in [6–8]. It should be noted, however, that, prior to employing the functions F_i^{theor} in the evaluation procedure based on Eq. (3), the theoretical cross section should be slightly corrected by shifting it toward higher energies by 0.30 MeV and by multiplying the result by a factor of 0.84 in order to reach closest agreement with the experimental cross section. A comparison of the original and corrected theoretical cross sections with the experimental cross section from [9] is illustrated in Fig. 2. The correspondent integrated cross sections are given in Table 1.

The $(\gamma, 1n)$, $(\gamma, 2n)$ and (γ, sn) cross sections for the isotope ^{91}Zr that were evaluated after the above correction within the experimental–theoretical approach based on Eq. (3) are presented in Fig. 3 along with the experimental (γ, xn) cross section from [9],

Table 1. Centers of gravity $E^{c.g.}$ and integrated cross sections σ^{int} for the reaction ^{91}Zr(γ, xn)

Energy region	$E^{c.g.}$, MeV	σ^{int}, MeV mb	$E^{c.g.}$, MeV	σ^{int}, MeV mb	$E^{c.g.}$, MeV	σ^{int}, MeV mb
	$E^{int} = B2n = 19.2$ MeV		$E^{int} = B3n = 28.5$ MeV		$E^{int} = 30.0$ MeV	
Experiment [9]	16.1	782.3 ± 4.8	18.5	1235.8 ± 12.5	18.9	1276.0 ± 17.2
Theory—original [6–8]	15.8	937.4 ± 19.2	18.4	1497.1 ± 21.7	18.8	1548.0 ± 21.9
Theory— corrected	16.0	782.3 ± 19.6	18.6	1249.4 ± 22.2	19.0	1291.8 ± 22.3

which was used as input information for the evaluation procedure based on Eq. (3).

The data presented in Fig. 3 and in Table 2 give a clear idea of those sizable systematic errors that were mentioned earlier in analyzing the energy dependences of the functions F_1^{expt} and F_2^{expt} and which were the cause of incorrectness in sorting multiplicity-1 and multiplicity-2 neutrons. In the region below the threshold of $B2n = 19.2$ MeV, the experimental and evaluated integrated cross sections for the respective $(\gamma, 1n)$ reaction are very close (780.8 and 782.6 MeV mb, respectively), but, in the energy region where the $(\gamma, 2n)$ reaction is also possible, the experimental integrated cross section for the $(\gamma, 1n)$ reaction proves to be 8% smaller than its evaluated counterpart (881.8 and 947.5 MeV mb, respectively), while the cross section for the $(\gamma, 2n)$ reaction is 22% larger (174.8 and 143.4 MeV mb).

So distinct an asymmetry of the difference in the experimental and evaluated cross sections for the $(\gamma, 1n)$ and $(\gamma, 2n)$ reactions in the energy region where the emission of three neutrons is forbidden in energy indicates clearly that the experimental method of neutron multiplicity sorting on the basis of measured neutron kinetic energies has yet another shortcoming. It stems from the disregard of proton-reaction contributions in the experiments under discussion. For example, the reaction denoted in [9] by $(\gamma, 1n)$ is in fact the $(\gamma, 1n) + (\gamma, 1n1p)$ reaction. The $(\gamma, 1n1p)$ reaction on the ^{91}Zr nucleus has a relatively low energy threshold of 15.6 MeV and a cross section commensurate with the $(\gamma, 2n)$ cross section. Obviously, the distribution of the excited-nucleus energy between the emitted neutron and proton in the $(\gamma, 1n1p)$ reaction is expected to be very close to the distribution of this energy between the two neutrons in the $(\gamma, 2n)$ reaction. However, the neutron multiplicity is one in the $(\gamma, 1n1p)$ reaction and two in the $(\gamma, 2n)$ reaction. In view of this, the procedure used in [9] to perform neutron multiplicity sorting and based on measurements of neutron kinetic energies is not quite correct, since the neutrons from the $1n$ and $2n$ channels are entangled within this approach: low-energy neutrons from the $(\gamma, 1n1p)$ reaction are misinterpreted as those that are associated with the $(\gamma, 2n)$ rather than with the $(\gamma, 1n)$ reaction.

It is noteworthy that the real discrepancies between the cross sections for the partial reactions in question because of the errors in neutron multiplicity sorting are even greater. This is because the difference between the integrated cross sections that were calculated within the above limits are somewhat distorted by the discrepancies between the experimental and evaluated reaction cross sections in the energy region extending to about 22.5 MeV, where they show a trend opposite to that at higher energies [for the $(\gamma, 1n)$ reaction, the experimental cross section is larger than its evaluated counterpart, while, for the $(\gamma, 2n)$ reaction, it is on the contrary smaller].

Table 2. Integrated cross sections σ^{int} for total and partial photoneutron reactions on the isotope ^{91}Zr along with experimental data from [9]

Reaction	σ^{int}, MeV mb	
	Evaluated data	Experimental data
$E^{int} = B2n = 19.2$ MeV		
$(\gamma, xn)^*$	782.3 ± 4.8	$782.3 \pm 4.8^*$
(γ, sn)	781.4 ± 4.8	782.8 ± 4.8
$(\gamma, 1n)$	782.6 ± 4.8	780.8 ± 22.0
$E^{int} = B3n = 28.5$ MeV		
$(\gamma, xn)^*$	$1276.0 \pm 17.2^*$	1235.8 ± 12.5
(γ, sn)	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

* Experimental cross section [9] used as an input in the evaluation procedure.

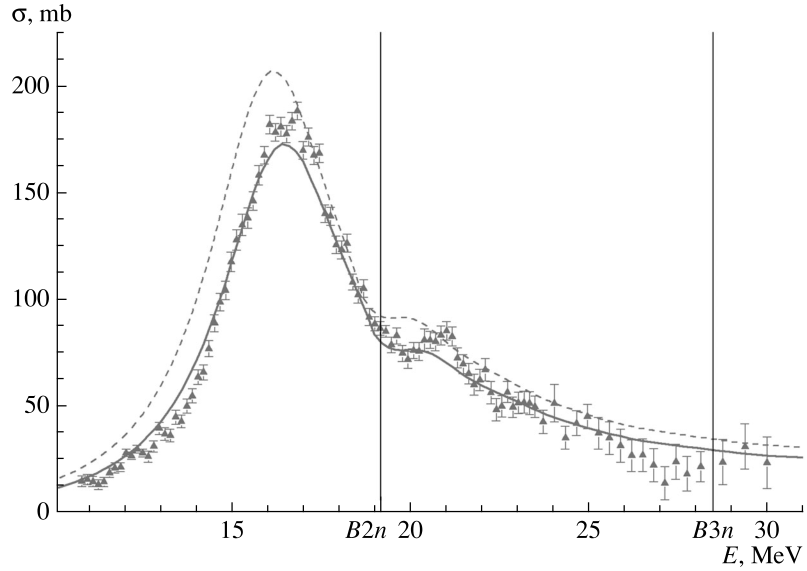

Fig. 2. Comparison of the (dashed curve) original and (solid curve) corrected theoretical cross sections for the reaction ^{91}Zr(γ, xn) with (triangles) experimental data from [9].

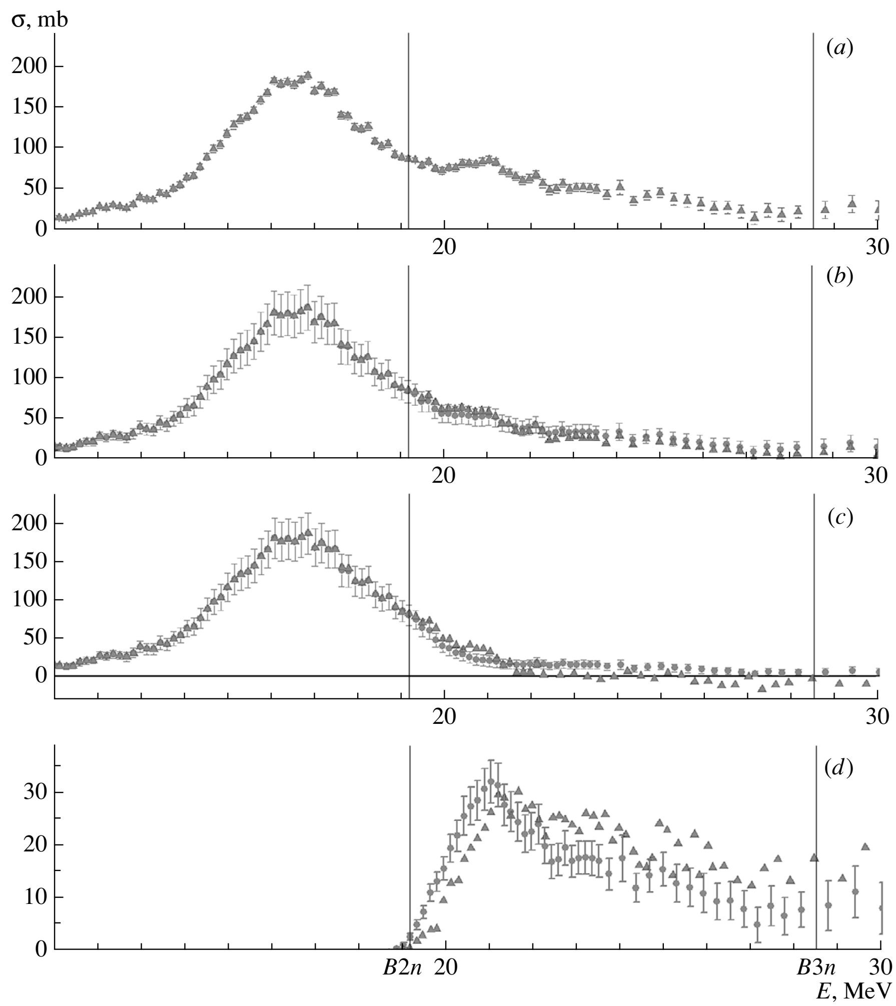

Fig. 3. Comparison of (circles) evaluated and (triangles) experimental [9] cross sections for the total and partial photoneutron reactions on ^{91}Zr nuclei: (a) $\sigma(\gamma, xn)$, (b) $\sigma(\gamma, sn)$, (c) $\sigma(\gamma, 1n)$, and (d) $\sigma(\gamma, 2n)$.

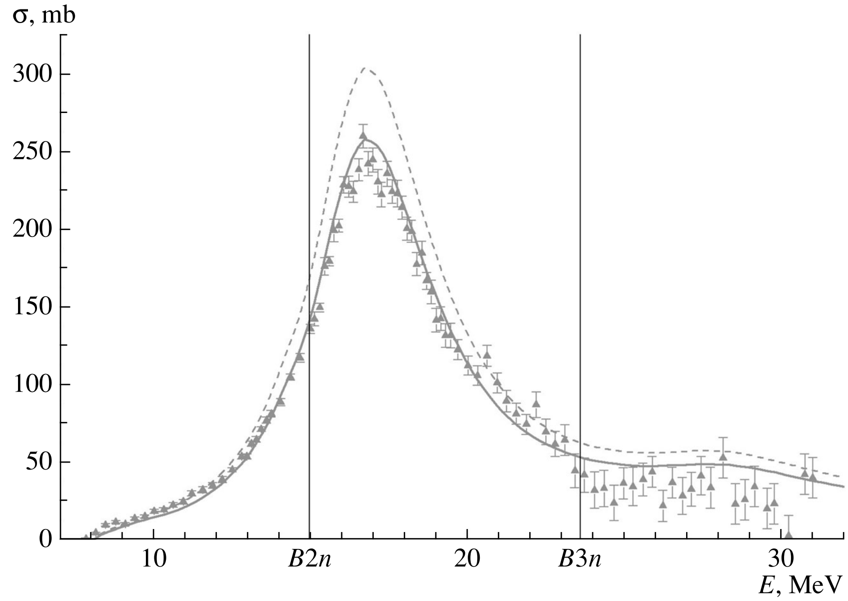

Fig. 4. Comparison of the (dashed curve) original and (solid curve) corrected theoretical cross sections for the reaction ^{94}Zr(γ, xn) with (triangles) experimental data from [9].

3.1. Data for the Isotope ^{94}Zr

By analogy with what was done in the case of the isotope ^{91}Zr, the theoretical neutron-production cross section in the case of the isotope ^{94}Zr was also slightly corrected prior to applying the evaluation procedure based on Eq. (3)—it was shifted toward higher energies by 0.02 MeV and multiplied by the a factor of 0.85. The correspondent data are given in Fig. 4 and in Table 3.

The cross sections evaluated for the $(\gamma, 1n)$, $(\gamma, 2n)$, $(\gamma, 3n)$, and (γ, sn) reactions on the isotope ^{94}Zr within the experimental–theoretical approach based on Eq. (3) are given in Fig. 5 along with the respective experimental (γ, xn) cross sections. Table 4 gives the correspondent integrated cross sections.

Up to the threshold of $B2n = 15.0$ MeV, the experimental and evaluated integrated cross sections for the $(\gamma, 1n)$ reaction are very close (260.6 and 261.8 MeV mb, respectively), in just the same way as in the case of the isotope ^{91}Zr, but, in the energy region where the $(\gamma, 2n)$ reaction is also possible, the experimental integrated cross section is 19% smaller than its evaluated counterpart for the reaction $(\gamma, 1n)$ (546.1 and 652.4 MeV mb, respectively) and is 15% larger for the $(\gamma, 2n)$ reaction (494.5 and 429.0 MeV mb). The asymmetry of the difference between the experimental and evaluated reaction cross sections is substantially smaller in the case of the isotope ^{94}Zr (19% and 15%) than in the case of the isotope ^{91}Zr (8% and 22%). We have shown above that this asymmetry may be due to the contribution of the $(\gamma, 1n1p)$ reaction involving a photoproton. In this case, the distinction between the two isotopes under study may be due to a substantially smaller contribution of this reaction in the case of the isotope ^{94}Zr in relation to the isotope ^{91}Zr—the reaction threshold is 2.2 MeV higher in the former than in the latter case, while the cross section is substantially smaller than the $(\gamma, 2n)$ cross section [6, 7].

In the region of energies above $B3n = 23.6$ MeV, the experimental integrated cross section already proves to be smaller than its evaluated counterpart by 60% for the $(\gamma, 1n)$ reaction (434.7 and 694.9 MeV mb, respectively), larger than that by 23% for the $(\gamma, 2n)$ reaction (662.6 and 539.4 MeV mb), and larger than that by 52% for the $(\gamma, 3n)$ reaction (85.4 and 56.1 MeV mb).

It is noteworthy that, by and large, the discrepancy between the experimental and evaluated cross sections are substantially larger in the case of the isotope ^{94}Zr than in the case of the isotope ^{91}Zr. This is because the energy threshold of $B3n = 23.6$ MeV for ^{94}Zr is much lower than that for ^{91}Zr ($B3n = 28.5$ MeV). Therefore, the errors of experimental neutron multiplicity sorting in the giant-dipole-resonance energy region for the former case receives contributions not only from the sorting of multiplicity-1 and multiplicity-2 neutrons but also

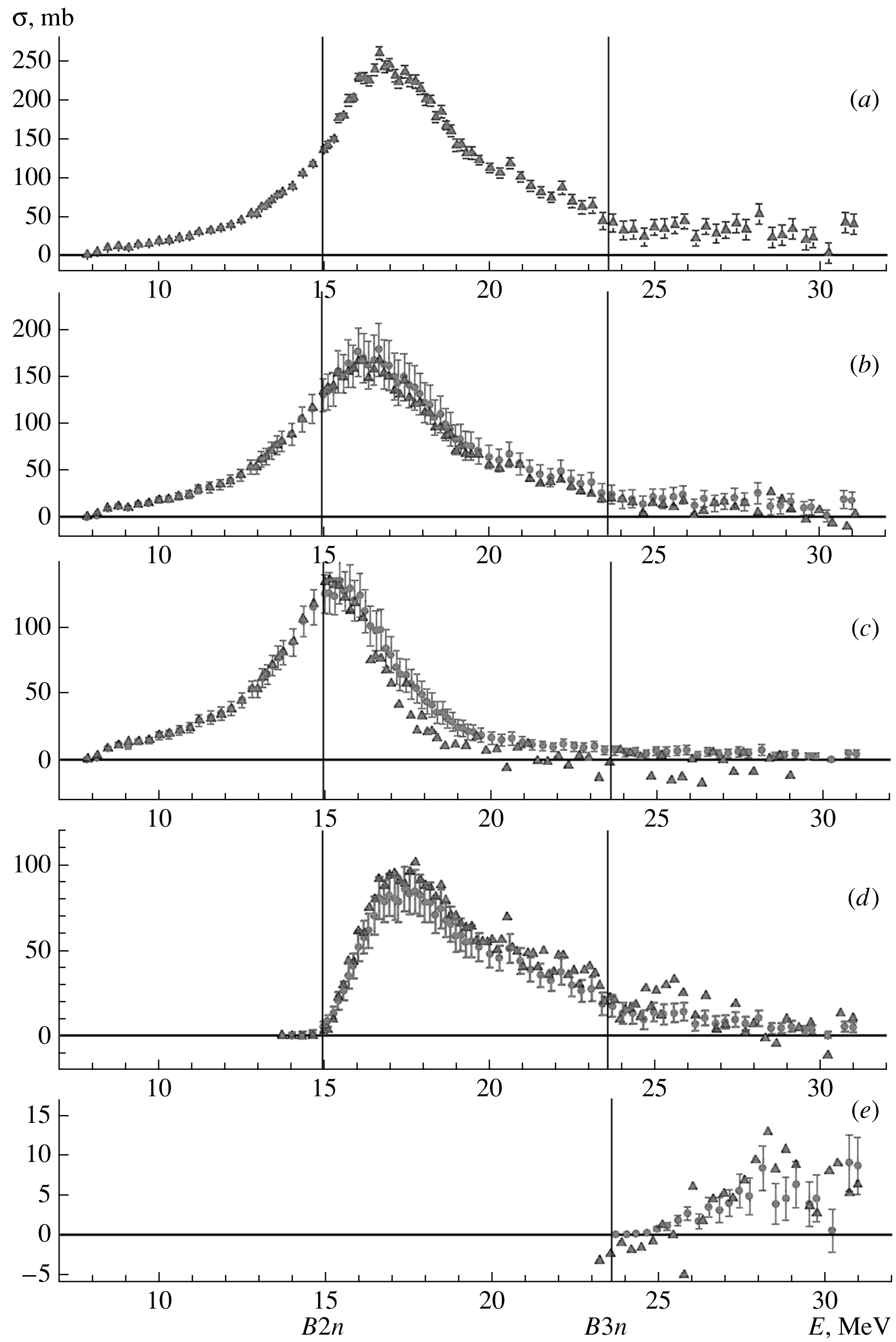

Fig. 5. Comparison of (circles) evaluated and (triangles) experimental [9] cross sections for total and partial photoneutron reactions on ^{94}Zr nuclei: (a) $\sigma(\gamma, xn)$, (b) $\sigma(\gamma, sn)$, (c) $\sigma(\gamma, 1n)$, (d) $\sigma(\gamma, 2n)$, and (e) $\sigma(\gamma, 3n)$.

from the sorting of multiplicity-1 and multiplicity-3, as well as multiplicity 2 and multiplicity-3, neutrons.

4. CONCLUSIONS

The investigations reported in the present article lead to specific conclusions on special features of the photodisintegration of the isotopes ^{91,94}Zr studied here.

The experimental data obtained in [9] on the cross sections for the $(\gamma, 1n)$ and $(\gamma, 2n)$ partial photoneutron reactions on ^{91}Zr nuclei and the $(\gamma, 1n)$, $(\gamma, 2n)$, and $(\gamma, 3n)$ partial photoneutron reactions on ^{94}Zr nuclei by the method of photoneutron multiplicity sorting have sizable systematic errors and do not satisfy the proposed criteria of data reliability. These errors stem from an unjustified redistribution of neu-

Table 3. Centers of gravity $E^{\text{c.g.}}$ and integrated cross section σ^{int} for the ^{94}Zr(γ, xn) reactions

Energy region	$E^{\text{c.g.}}$, MeV	σ^{int}, MeV mb	$E^{\text{c.g.}}$, MeV	σ^{int}, MeV mb	$E^{\text{c.g.}}$, MeV	σ^{int}, MeV mb
	$E^{\text{int}} = B2n = 15.0$ MeV		$E^{\text{int}} = B3n = 23.6$ MeV		$E^{\text{int}} = 31.0$ MeV	
Experiment [9]	12.9	299.3 ± 2.7	17.3	1545.8 ± 12.3	18.6	1779.3 ± 25.2
Theory—original [6–8]	13.1	333.4 ± 8.8	17.3	1820.5 ± 30.3	19.1	2228.9 ± 31.6
Theory—corrected	13.1	283.1 ± 9.0	17.3	1545.5 ± 31.1	19.1	1892.2 ± 32.4

trons from the $1n$, $2n$, and $3n$ channels. For example, it is precisely the removal of a significant fraction of neutrons from the $1n$ channel in various photon-energy regions that leads to the decrease in the cross sections for the respective $(\gamma, 1n)$ reactions and even to the appearance in them (and accordingly in the function F_1^{expt}) physically forbidden negative values.

Table 4. Integrated cross sections σ^{int} for total and partial photoneutron reactions on the ^{94}Zr isotope along with experimental data from [9]

Reaction	σ^{int}, MeV mb	
	Evaluated data	Experimental data
$E^{\text{int}} = B2n = 15.0$ MeV		
$(\gamma, xn)^*$	260.5 ± 2.4	$260.5 \pm 2.4^*$
(γ, sn)	262.8 ± 2.4	259.8 ± 2.4
$(\gamma, 1n)$	261.8 ± 9.1	260.6 ± 2.5
$E^{\text{int}} = B3n = 23.6$ MeV		
$(\gamma, xn)^*$	$1506.7 \pm 12.2^*$	1506.7 ± 12.2
(γ, sn)	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
$E^{\text{int}} = 31.0$ MeV		
$(\gamma, xn)^*$	$2067.2 \pm 40.0^*$	2067.2 ± 40.0
(γ, sn)	1011.1 ± 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

* Experimental cross section from [9] used as an input in the evaluation procedure.

At the same time, the cross sections for the $(\gamma, 2n)$ reaction become larger upon misidentifying some neutrons from the $1n$ channel as those that belong to the $2n$ channel. Therefore, the function F_2^{expt} also grows, developing values that exceed the limit of 0.50, which is physically admissible according to the definition in (1).

The systematic errors found in the experimental cross sections for partial photoneutron reactions on nuclei of the isotopes ^{91,94}Zr, as well as on ^{90}Zr, ^{115}In, ^{112,114,116,117,118,119,120,122,124}Sn, ^{159}Tb, ^{181}Ta, and ^{197}Au nuclei studied earlier [1–5], are due to the proximity of the kinetic energies of neutrons from different partial reactions. By considering the example of data on the isotope ^{181}Ta, it was shown in [4] that, as the photon energy grew in the giant-dipole-resonance region, the shape of the photoneutron spectrum did not change strongly—a dominant contribution came from neutrons of energy about 1 MeV. On the other hand, it turned out that, upon the opening of a channel in which more neutrons were emitted (for example, the $2n$ channel), some low-energy neutrons were associated with the $1n$ channel. Moreover, the contribution of reactions where a photoproton (photoprotons) appeared in the final state and which the authors of [9] disregarded aggravated this situation.

We emphasize that the cross sections evaluated for both the $(\gamma, 1n)$, $(\gamma, 2n)$, and $(\gamma, 3n)$ partial reactions and the (γ, sn) total reactions on nuclei of the isotopes ^{91,94}Zr within the theoretical–experimental approach based on Eq. (3) are free from the systematic errors being discussed and differ substantially from the experimental cross sections.

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