Comment on "Resonant and nonresonant contributions to the photoactivation of ^{111}Cd"

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(Received 2 January 1991)

The most recent investigation of the photoactivation of isomeric nuclei reported by Krčmar *et al.* continues to contain a strong contribution from nonresonant channels. In this Comment we report that integrated cross sections for resonant photoactivation of isomeric levels in ^{87}Sr, ^{111}Cd, and ^{113,115}In have been calculated from recent nuclear structure data and compared to those experimental results which assert that nonresonant contributions are important. The latter have shown systematically smaller values and the amount of missing resonant strength can be correlated to the magnitude of nonresonant cross section found in these investigations. Monte Carlo simulations of realistic experimental geometries display important components in the photon fields, resulting from environmental Compton scattering, which have been omitted in previous analyses of experimental data. The strength and shape of this contribution as a function of the experimentally important parameters suggest that the data can be explained entirely on the basis of a resonant excitation mechanism without any need to introduce a nonresonant contribution.

Photoactivation cross sections for the reaction $^{111}\mathrm{Cd}(\gamma,\gamma')^{111}\mathrm{Cd}^m$ have been inferred by Krčmar *et al.* [1] to proceed through *nonresonant* channels of excitation. The study of the photoactivation of isomeric states has a strong tradition in nuclear structure studies, and it has been generally accepted that the isomer population occurs via *resonant* excitation of higher-lying intermediate states which show finite branching ratios for a γ decay (or a cascade) to the isomer. Nonresonant cross sections from higher-order photonuclear interactions are typically many orders of magnitude smaller and have never been considered important for these reactions. However, in the last few years a series of experiments has been performed [2–4] on the photoactivation of $^{115,113}\mathrm{In}^m$, $^{111}\mathrm{Cd}^m$, and $^{87}\mathrm{Sr}^m$ in order to search for possible nonresonant contributions. The paper [1] upon which we comment is the most recent example in this series.

The experiments designed to show nonresonant excitation typically involved strong ^{60}Co sources. The methodology depended upon the distortion of the source spectrum by interposing lead scatterers of varying thickness. In this way the ratio of photon fluxes at energies of resonant intermediate states and at source-line energies were changed in a definite way. The intensities at the energies of the source lines were believed to dominate the nonresonant interaction. The excitation probability per unit time of the isomeric level, P, was then fitted to a sum of resonant and nonresonant contributions:

$$P=\Phi_R(E_R)\sigma\Gamma_R+\Phi_{\mathrm{NR}}\sigma_{\mathrm{NR}}\ \mathrm{s}^{-1}\ . \tag{1}$$

In this expression Φ_R was the photon flux at resonance energy E_R, $\sigma\Gamma_R$ represented the integrated isomeric cross section from resonance fluorescence, Φ_{NR} was the nonresonant photon flux integrated over energy, and σ_{NR} was a total nonresonant cross section.

The authors of Refs. [1–4] (called the Zagreb group hereafter) claim that the nonresonant contribution to the population of isomers is significant and even dominates in experiments using radioactive sources. Because of its potential importance, these findings stimulated a series of experimental and theoretical work over the last years. Hot atom chemical studies [5] and investigations using bremsstrahlungs sources [6,7] found no evidence for nonresonant cross sections in ^{115}In and ^{111}Cd. Bikit *et al.* [8] repeated the ^{111}Cd experiment of Ref. [3], but used iron instead of lead scatterers. They, too, report a null result within experimental limits. All attempts to explain theoretically such a strong nonresonant excitation mechanism have failed so far [2,9,10].

Most recently, the Zagreb group has repeated the ^{111}Cd experiment including iron scatterers also [1]. They report large nonresonant cross sections independent of the absorber type or excitation energy of the dominant resonant intermediate state, which is not well established in ^{111}Cd. A comprehensive discussion of their views of the validity of the experiments described in Refs. [5–8] is additionally presented.

We would like to discuss several aspects which raise serious doubts about the results presented in Refs. [1–4]. In particular, we will compare their results to resonant cross sections calculated from recent nuclear structure data and show that their results deviate significantly in those cases where the largest nonresonant contribution is obtained with their method. We will further show that an accurate knowledge of the impinging photon field is crucial for the data analysis. The incident spectrum depends strongly upon the experimental environment, and the approximation used for the unknown photon field in

those experiments [1–4] turns out to be poor. The neglect of forward Compton scattering in collimators and other environmental material could have led to severe errors.

Discussion of nuclear structure data.—The excitation probability of isomeric levels through resonant photon absorption to intermediate states can be calculated easily when the relevant quantum numbers of the intermediate states are known. The integrated cross section is given by

$$\sigma\Gamma_R = \frac{\lambda^2}{4}\,\frac{2J_f+1}{2J_i+1} b_0 b_{\rm iso}\Gamma \ , \tag{2}$$

with $b_0=\Gamma_0/\Gamma$, $b_{\rm iso}=\Gamma_{\rm iso}/\Gamma$, and $\lambda=2\pi\hbar c/E_R$. The quantity E_R denotes the energy of the intermediate state, $J_{i,f}$ are the spins of the initial and final states, Γ and Γ_0 represent the total and partial widths for direct decay to the ground state, and $\Gamma_{\rm iso}$ is the effective partial decay width to the isomer (including possible cascades). The branching ratios b_0, $b_{\rm iso}$, and Γ (or the lifetime $\tau=\hbar/\Gamma$) or Γ_0 have been measured with a variety of experimental methods for the nuclei studied by the Zagreb group. Table I presents adopted values from the most recent Nuclear Data Sheets plus additional results from new experiments. It has been argued in Ref. [2] that such a comparison shows variations of the different experimental data much too wide to allow for any conclusion. Thus this question will be discussed in detail for the nuclides studied.

^{115}In. Two resonant states are known in the energy region accessible by ^{60}Co sources. The lower level at 0.941 MeV contributes roughly an order of magnitude less to the isomer cross section than the state at 1.078 MeV. The branching ratios have been measured by various methods including Coulomb excitation, β decay, and $(n,n'\gamma)$ reactions [11]. There is, however, a discrepancy of the $(n,n'\gamma)$ results adopted in Ref. [11] with two $(n,n'\gamma)$ studies [12, 13] quoting significantly larger $b_{\rm iso}$ values (35–40 %) for the 1.078-MeV state. At first sight this is surprising, since $(n,n'\gamma)$ results were generally found to agree well with each other, as well as with other experiments. A closer inspection of the original γ spectra resolves the problem. The corresponding γ transition is placed on the tail of a much stronger line with a separation in energy close to the resolution of the detectors used. Accordingly, results are very sensitive to small variations of the line shape, and a systematic error must be assumed for the integration, which might easily reach a factor 2 or more.

The lifetime has been measured by Coulomb excitation [14] and nuclear resonance fluorescence [15] (NRF) with reasonable agreement of both results. Some older NRF experiments quote a larger total width [16,17], but Ref. [16] gives a very large error. The most recent NRF experiment is considered superior to Ref. [17] because the critical evaluation of the bremsstrahlung flux and spectral distribution was achieved from a consistent set of self-absorption measurements. In any case the integrated cross section calculated is at least 2 times bigger than the result of Ref. [2]. The agreement with recent experimental work [5,6] and with the older survey of Booth and Brownson [18] is acceptable. Allowing for the Γ values of Refs. [16,17] would only enlarge the discrepancy with Ref. [2].

^{113}In. The result of Ref. [4] is roughly 40% smaller than the calculation, if one includes the weaker 1.021-MeV level. Again, the branching ratios are well defined by different experiments [19], but the lifetimes are given only by one measurement [14]. While this allows the possibility for some systematic error, the agreement might still be regarded as acceptable. The only other experimental result [18] on ^{113}In^m shows an error compatible with both the calculation and Ref. [4].

^{111}Cd. No calculations can be performed for ^{111}Cd, since the branching ratios are not known and since there are conflicting results about the excitation energy of the main resonant state [1,7,18,20]. Recent studies have re-

TABLE I. Energies, spins, half-lives, and branching ratios of resonant intermediate states populating ^{87}Sr^m, ^{111}Cd^m, and ^{113,115}In^m that could be excited in ^{60}Co source experiments. The calculated integrated cross sections are compared to values for the resonant and nonresonant contributions derived by the Zagreb group [1–4].

Nucl.	E_x (MeV)	J^π	$T_{1/2}$ (ps)	b_0 (%)	$b_{\rm iso}$ (%)	Calc. $\sigma\Gamma_R$ (μb keV)	Zagreb group[a] $\sigma\Gamma_R$ (μb keV)	$\sigma_{\rm NR}$ (nb)
^{115}In	0.941	$\frac{5}{2}^+$	15.1(1.4)	88(2)	12(2)	8(1)		
	1.078	$\frac{5}{2}^+$	0.99(10)	83(2)	16(2)	120(15)	54(7)	380(40)
^{113}In	1.021	$\frac{5}{2}^+$	3.6(3)	89(2)	11(2)	23(2)		
	1.131	$\frac{5}{2}^+$	0.97(7)	85(2)	14(2)	101(9)	81(1)	20(3)
^{111}Cd	1.190[b]						78(8)[c]	10(3)[c]
							97(6)[d]	7(3)[d]
	1.330[b]						62(5)[c]	12(2)[c]
							83(5)[d]	10(2)[d]
^{87}Sr	1.229	$\frac{5}{2}^+$	0.97(35)	86(3)	14(3)	86(33)	47(7)	32(4)

[a]For other experimental $\sigma\Gamma_R$ results see Table I in Ref. [6] (^{115}In), Ref. [18] (^{113}In), Table I in Ref. [7] (^{111}Cd), and Table I in Ref. [4] (^{87}Sr). [b]Assumed energy of the main intermediate state. [c]Results obtained with lead scatterer/absorber. [d]Results obtained with iron scatterer/absorber.

vealed a low-lying state which is probably the last step of cascades towards the isomer [21], but the intermediate states for photoabsorption were not discovered. Németh and Veres [22] have constructed a possible scheme of levels assuming a similarity to the nuclear structure of the lowest intermediate states in ^{87}Sr and ^{113,115}In. It should be noted, however, that the ground state and isomeric spins completely differ ($\frac{1}{2}^+ \rightarrow \frac{11}{2}^-$ for ^{111}Cd and $\frac{9}{2}^+ \rightarrow \frac{1}{2}^-$ for the others). A similar structure of the last three is suggested by the single neutron (Sr) or proton (In) hole relative to the shell closure $N=50$. A microscopic analysis within the unified model [23,24] shows indeed that the wave functions of all three main intermediate states are dominated by amplitudes of $[(A+1,2_1^+)\otimes g_{9/2}^{-1}]_{5/2+}$ character, while the structure of a corresponding state in ^{111}Cd depends on a completely different region of valence single-particle and hole states.

Certainly, further experimental clarification of the ^{111}Cd low-energy spectrum is needed. The resonant cross sections for the population of the isomer in Ref. [1] are within the bandwidth of other experimental results, and no further conclusions are possible. However, it seems remarkable that the $\sigma\Gamma_R$ values presented in Ref. [1] disagree with each other, depending on the scatterer type (iron or lead).

^{87}Sr. The branching ratios are averages of three different experimental methods [25–27], which agree with each other within 3%. A half-life measurement obtained from the Doppler shift attenuation method is given by Ref. [26]. The calculated value agrees with Ref. [18], but the result of Ref. [4] is significantly smaller, even regarding the error quoted for the half-life value.

The discussion shows that the integrated cross sections for resonant isomer excitation given by the Zagreb group are generally smaller than values calculated from nuclear structure data characterized and established by a variety of independent experimental methods. A clear correlation can be found between the missing resonant cross section and the magnitude of the nonresonant cross section derived in Refs. [1–4]. This points toward problems related to the data analysis of their experimental method, which we address next.

Monte Carlo simulations.—A crucial point of all photoactivation experiments is the characterization of the incident photon field. The spectra resulting from strong radioactive sources (of the order of kCi) cannot be measured easily. Thus the following approximation has been used [1–4,8] for the unknown variation with the thickness of the scatterer of Φ at the energies of the resonant states and of full-energy γ rays (defining $\Phi_{\rm NR}$): The function $f(E_R,d)$, describing the flux ratio, was taken from data measured with a weak ^{60}Co source which emits full-energy photons only. Because of the finite volume and additional shielding, typical for a strong source of the type used in the reported experiments, a certain fraction k is scattered out of the full-energy lines to form a low-energy tail even at $d=0$. Assuming that the distribution of k is constant in the vicinity of the resonance energies, k can be treated as a free parameter which is fixed by the data.

Neglecting small differences in the attenuation coefficients $\mu \simeq \mu(E_R) \simeq \mu(E_0)$, the reaction rate $P(d)$ is then given by [1–4,8]

$$P(d)=\frac{A}{4\pi r^2(1+k)} \times e^{-\mu d}\left\{\sigma_{\rm NR}+\left[\frac{k}{\Delta E}+f(E_R,d)\right]\sigma\Gamma_R\right\}. \quad (3)$$

Here ΔE is the interval of energies over which k is distributed, and A and r represent the activity of the source and distance to the target, respectively. In Refs. [1–4,8] values for $\sigma\Gamma_R$, $\sigma_{\rm NR}$, and k resulted from simultaneous least-squares fits to the experimental $P(d)$ values.

For the preparation of these comments, the validity of this approach was investigated in three aspects: (i) the approximations included in the term in square brackets of Eq. (3), (ii) the importance of the experimental environment as a source of additional Compton-scattered photons, and (iii) the influence of a collimator as used in Ref. [2]. The discussion is based on Monte Carlo simulations performed with the code GEANT [28]. The characteristic quantity

$$F(E_R,d)=\frac{N(E_R)}{\Delta E' N(E_0)} \quad (4)$$

is calculated for a variety of geometries and may be compared to $f(E_R,d)$ described above. The number $N(E_0)$ is determined by the sum of the contents of the intervals containing the 1.173- and 1.332-MeV lines, and $N(E_R)$ is averaged over $\Delta E'=50$ keV in order to improve statistics. In the case of ^{87}Sr, where the main intermediate state lies above the 1.173-MeV level, only the 1.332-MeV line is counted for $N(E_0)$.

(i) As an example, results for the 1.229-MeV state in ^{87}Sr are discussed. The characteristic ratio $F(E_R,d)$ is compared in Fig. 1 for a pointlike source, an extended cylindrical source of 1 cm^3 volume, and an extended source surrounded by 1 and 2 cm of lead shielding, respectively. The function obtained for the pointlike source, simulating $f(E_R,d)$ in Eq. (3), is shifted such that $k/\Delta E=F(E_R,0)$ for the extended source geometries. The agreement is acceptable, although a slightly slower increase with d is systematically indicated for the pointlike case. Such an effect would significantly alter the results of the Zagreb group, if source shielding material equivalent to 2 cm Pb or more was present in the experiments.

(ii) An important contribution not considered in Eq. (3) is the presence of *additional* photons at the resonance energies from scattering in the experimental environment. At energies around 1 MeV, the Compton cross sections are large and the Klein-Nishina formula is only moderately peaked in the forward direction. The schematic view of the experimental setup of the ^{87}Sr and ^{113}In measurements [4], presented in Fig. 4 of Ref. [29], indicates the presence of enough scattering material to provide a substantial contribution of this type. The geometry is simplified in the present calculations by replacing these scatterers with a lead cone having an open-

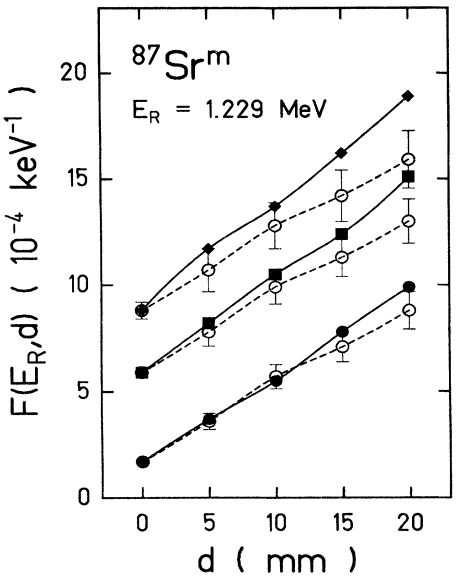

FIG. 1. Comparison of the photon flux at resonance energy E_R to the 1.332-MeV line flux for different geometries of a ^{60}Co source. The characteristic ratio $F(E_R,d)$ is shown as a function of the Pb scatterer thickness for a pointlike source (open circles), an extended source (solid circles), an extended source plus 1-cm Pb shielding (squares), plus 2-cm Pb shielding (diamonds). For details of the extended source geometry, see text. Statistical errors of the extended source calculations (not shown) are of the order of the symbol size. The straight and dashed lines are drawn to guide the eye.

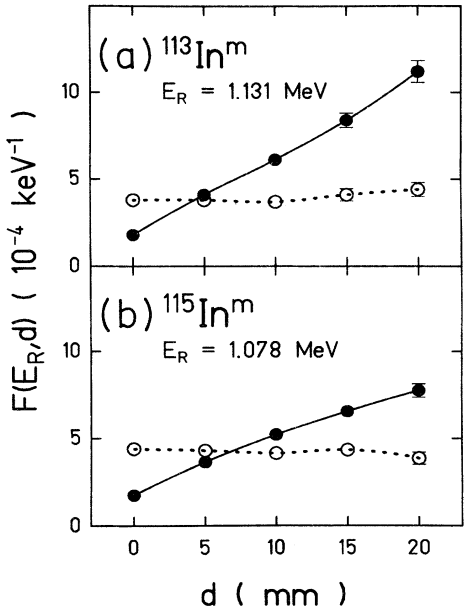

FIG. 2. Direct (solid circles) and scattered (open circles) contributions to $F(E_R,d)$ from the simulation of the experiments described (a) in Ref. [4] and (b) in Ref. [2]. For details, see text. The straight and dashed lines are drawn to guide the eye.

ing of 4 cm at the source, widening to 11 cm at a target distance of 22 cm. The source itself is assumed to be extended as described above and without additional lead shielding. Using ^{113}In as example, the contributions of the direct and indirect (i.e., scattered) parts are shown in Fig. 2(a) as a function of d by solid and open symbols, respectively. *The flux at resonance produced by scattered photons reveals a completely different functional behavior and even dominates at* $d=0$.

From Eq. (3) it is obvious that a roughly constant additional contribution directly competes with the σ_{NR} term. Because of the strength indicated by the calculations, any need for a nonresonant cross section to describe the $P(d)$ data in Ref. [4] is completely destroyed. This statement would still hold even if the geometry used were to overestimate the indirect contribution in the experiment up to a factor of about 4. Note that the additional photon flux cannot be accounted for by an increase of the parameter k in Eq. (3) because of the presence of the $(1+k)^{-1}$ factor.

(iii) An even more striking example of the role of Compton-scattered photons in the experimental analysis is found in the anomalously large nonresonant cross section reported [2] in ^{115}In (see Table I). In contrast to the other experiments, a collimator was used. While no further details of the collimator were given, we tried to follow the experiment description given in Ref. [2] as close as possible. A distance source target of 28 cm was chosen, and the target diameter and collimator width were set to 2.54 cm. The source description corresponds to case (ii). Figure 2(b) shows again that the shape of $F(E_R,d)$ differs completely for the direct and indirect contributions, and that the indirect flux even dominates in the absence of the lead/iron scatterers. The strength of scattered photons in such a geometrical arrangement is absolutely sufficient to explain the experimental $P(d)$ values in Ref. [2] without a nonresonant cross section. We also ascertained that this result is valid independent of a variation of the collimator radius within reasonable limits.

In conclusion, a comparison of experimental integrated cross sections for resonant photoexcitation obtained by the Zagreb group with results calculated from independent nuclear structure data shows systematic deviations. The amount of resonant cross section missing from the Zagreb measurements is closely correlated to the magnitude of the nonresonant cross sections they report.

We have shown with Monte Carlo simulations of typical experimental geometries that important parts of the photon spectra are missed in the approximation used by the Zagreb group for their data analysis. The computational results demonstrate the importance of additional photon flux resulting from Compton scattering in the experimental environment. In particular, the results derived for ^{115}In are severely affected by the use of a collimator, and the data analysis reported in those experiments must be reconsidered. This neglected contribution seems to be sufficient to explain all published experimental data without any need to introduce a nonresonant

cross section.

Finally, we remark that the calculations presented do not attempt to provide a detailed characterization of the experiments in Refs. [1–4], since not enough details were given there. A measurable nonresonant contribution to the photoexcitation of isomers cannot be definitely excluded by our arguments, but any claim for its existence from the experimental methods reported to date will have to be based on an accurate description of the photon field in the respective experimental geometry.

We are indebted to N. Huxel for his assistance with the Monte Carlo calculations. This work has been supported by the Bundesministerium für Forschung und Technologie of the Federal Republic of Germany under Contract No. 06DA184I.

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