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SECONDARY PHOTON-ATOM CONTRIBUTIONS TO NONRESONANT EXCITATION OF ^{115}In ISOMERIC LEVEL

MILICA KRČMAR, ANTE LJUBIČIĆ and KRUNOSLAV PISK

Ruđer Bošković Institute, 41000 Zagreb, Yugoslavia

BRIAN A. LOGAN

Physics Department, University of Ottawa, Ottawa, Ontario, Canada

and

MATIJA BISTROVIĆ

Central Institute for Tumors and Allied Diseases, 41000 Zagreb, Yugoslavia

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A ^{60}Co source has been used to study the photoactivation of the 336 keV ($T_{1/2}$ = = 4.3 h) isomeric level of ^{115}In in the indium-lead alloys. We have investigated the contribution to the nonresonant excitation from photon-atom interactions, involving Compton scattering, photoelectric effect and pair production. Our results show very clearly that the nonresonant excitations of the isomeric level are due to direct interactions between the photons and ^{115}In atoms.

1. *Introduction*

The photoactivation of isomeric levels with ^{60}Co sources has generally been assumed to be due to the excitation, via resonance fluorescence, of higher nuclear levels with photons in the low energy tail of the intense ^{60}Co beams necessary for photoactivation. These higher levels then decay to populate the isomeric levels.

However, recent investigations of the photoactivation of isomeric levels of ^{115}In$^{1)}$ ^{111}Cd$^{2)}$ and ^{113}In and ^{87}Sr$^{3)}$, have shown that, even though the cross sections are only $\approx 10^{-32}$ cm^2, nonresonant contributions can be important. Possible mechanisms for the nonresonant excitations, such as nuclear Raman scattering, off-resonance fluorescence and the inelastic photoelectric effect have been considered$^{1,2)}$, but the cross sections are far too low to explain the experimental results. These nonresonant excitations in which the isomeric level has been excited by direct photon-isomeric atom interactions, we call primary photon-atom contributions.

Secondary contributions to nonresonant excitations can be produced in such photon-atom interactions as the Compton effect, the photoelectric effect and pair production. Free electrons and positrons are produced in irradiated samples and these can excite nuclear levels of isomeric atoms. Nuclear levels can also be excited during positron annihilation.

Although it is possible to make theoretical estimates of these contributions much more confidence can be placed in an experimental investigation and this paper reports an experimental estimate of secondary photon-atom contributions to the photoactivation of isomeric levels.

2. *The experimental technique*

We have investigated the contributions from secondary photon-atom interactions in an experimental arrangement involving ^{115}In and a collimated beam from a 1.12×10^{14} Bq ^{60}Co source which, according to the manufacturer's specifications, had a low energy tail intensity k which was 0.19 of the full energy photon intensity. It is well-known that photoactivation of the 336 keV ($T_{1/2} = 4.3$ h) isomeric level of ^{115}In can proceed via resonance fluorescence of the 1078 keV level. Different composition indium-lead alloys (containing 4—5% antimony) have been irradiated and we have used our knowledge of the atomic number ($Z_{In} = 49$, $Z_{Sb} = 50$, $Z_{Pb} = 82$) dependence of the various photon-atom interactions to estimate the contributions from the different atomic phenomena. We have assumed a Compton contribution which depends linearly on Z, a photoelectric effect contribution which varies as $Z^{4.4}$, and a pair production contribution which depends on Z^2.

Before investigating the different secondary photon-atom contributions, we measured σ_R, the integrated cross section for resonant excitation of the isomeric level via 1078 keV level, and σ_{NR}^T the total cross section for nonresonant excitation of the isomeric level. The probability P of exciting the isomeric level per unit time is given by

$$P = \sigma_R \Phi_R (E_R) + \sigma_{NR}^T \Phi_{NR}, \qquad (1)$$

where $\Phi_R (E_R)$ is the flux of photons per unit area, energy and time at the resonant energy. In this case the resonant energy is 1078 keV. Φ_{NR} is the flux of nonresonant photons per unit area and time. $\sigma_R = g^2 \lambda^2 \Gamma_0 \Gamma_{iso} / 4\Gamma$ represents an integrated cross section for the resonant contribution. The parameters g, Γ_0, Γ_{iso} and Γ are, respectively, the statistical weight, the ground state transition width of the resonance level, the partial width for decay to the isomeric level and the total width of the resonance level. λ is the wavelength of the photons which excite the resonance level at the energy E_R. σ^T_{NR} is the total cross section for the nonresonant excitation of the isomeric level and is given by

$$\sigma^T_{NR} = \sigma_{iso}(E_{iso}) + \sum_i \sigma_{NR}(E_i) \frac{\Gamma_{iso}(i)}{\Gamma_i} \tag{2}$$

where E_{iso} is the energy of the isomeric level and i^{th} nuclear level has an energy E_i $(E_i > E_{iso})$, a width Γ_i and a partial width $\Gamma_{iso}(i)$ for decay to the isomeric level. Our experimental technique was the same as in the previous measurements[1,2,3], and involved interposing the lead absorbers with thicknesses of 0.4, 0.8, 1.2 and 1.6 cm between the ^{60}Co and indium sample to vary the relative values of $\Phi_R(E_R)$ and Φ_{NR}, and allows an estimate of the resonant and nonresonant contributions. The relative values of $\Phi_R(E_R)$ and Φ_{NR} depend on the tail of low energy photons emerging from the source and the scattering and absorption in the lead absorbers and samples. The effects of the absorbers and the sample on the photon energy distribution are investigated using a small ^{60}Co source, which in contrast to the source used for photoactivation, has a negligible low energy tail. In our case Φ_{NR} is dominated by the flux of 1173 keV and 1332 keV photons and has been taken as the sum of the fluxes associated with these gamma rays. The natural indium disc had a diameter of 2.54 cm and a thickness of 0.4 cm; it was placed 22 cm from the source. Irradiations were made about 13 h and the subsequent decays of the isomeric levels were measured with a Ge(Li) detector. A χ^2 analysis of the experimental data was made with k, σ_R and σ^T_{NR} as variable parameters. The best χ^2 value coresponded to a k value of 0.19, which is in excellent agreement with the source specifications, and cross sections of $\sigma_R = (9.6 \pm \pm 2.0) \times 10^{-29}$ cm^2 keV and $\sigma^T_{NR} = (4.5 \pm 0.9) \times 10^{-32}$ cm^2 were obtained.

In our investigations of the secondary photon-atom contributions to the nonresonant isomeric level excitation, we have used four alloys which contained, respectively, 28%, 42%, 46% and 71% of ^{115}In atoms. The samples were 0.4 cm thick and had a diameter of 2.54 cm, and were placed 22 cm from the source. The number of nonresonant excitations of the ^{115}In isomeric level per unit time N^{iso}_{NR}, in the irradiated alloys, was estimated from

$$N^{iso}_{NR} = N^{iso}_T - N^{iso}_R, \tag{3}$$

where N^{iso}_T is the observed number of isomeric level excitations per unit time and N^{iso}_R is the number of excitations produced via resonance fluorescence of the 1078 keV level. N^{iso}_R can be estimated from σ_R:

$$N^{iso}_R = \Phi_R(E_R)\,\sigma_R\,N_{In}. \tag{4}$$

N_{In} is the number of ^{115}In atoms in the alloys. Irradiations were made for about 13 h and the subsequent decays of the isomeric levels were measured with a Ge (Li) detector.

3. Secondary photon-atom contributions

a) *Contributions involving the Compton effect*

It has been assumed that these contributions produced via excitation of the 1078 keV level by free electrons from the Compton scattering. The number of isomeric level excitations per unit time due to the Compton effect is given by

$$N_C^{iso} \simeq \Phi_{NR}\, n_C\, \mathrm{d}\sigma_C\, l_C\, n_{In}\, \sigma_{ee'}\, \frac{\Gamma_{iso}}{\Gamma}. \tag{5}$$

$\mathrm{d}\sigma_C$ is the differential cross section[4)] corresponding to scattering angles which produce Compton recoil electron with energies $\geqslant 1078$ keV. l_C is the average sample thickness corresponding to the Compton electrons slowing down to 1078 keV (electron energy loss[5)] for different alloys is in the range of 9—11.5 MeV/cm). n_{In} is the number of ^{115}In atoms per unit volume. $\sigma_{ee'}$ is the cross section for the excitation of the 1078 keV level by Compton electron; a value of 10^{-32} cm^2 has been taken[6)]. n_C is given by

$$n_C = N\,(p_{In}\, Z_{In} + p_{Sb}\, Z_{Sb} + p_{Pb}\, Z_{Pb}), \tag{6}$$

where N is the number of atoms in the sample and the p values represent the fractions of different elements in the sample. Γ_{iso}/Γ is the ratio of the partial width for decay to the isomeric level to the total width of the 1078 keV level. A value of 0.19 was assumed.

Using known values for the parameters, we calculated N_C^{iso} for each alloy. The calculated values were six orders of magnitude below the experimental results. This result shows that secondary contributions involving the Compton effect are negligible.

b) *Contributions involving the photoelectric effect*

It has been assumed that this contribution also proceeds via excitation of the 1078 keV level and the number of isomeric level excitation per unit time due to the photoelectric effect is given by

$$N_{ph}^{iso} \simeq \Phi_{NR}\, n_{ph}\, \sigma_{ph}^{o}\, l_{ph}\, \sigma_{ce'}\, \frac{\Gamma_{iso}}{\Gamma}, \tag{7}$$

where l_{ph} is the average sample thickness corresponding to photoelectrons ejected from K-shells slowing down to 1078 keV (electron energy loss for different alloys is in the range of 9—11.5 MeV/cm). σ_{ph}^{o} is the photoelectric cross section[4)] for a hydrogen atom and n_{ph} is given by

$$n_{ph} = N\,(p_{In}\, Z_{In}^{4.4} + p_{Sb}\, Z_{Sb}^{4.4} + p_{Pb}\, Z_{Pb}^{4.4}). \tag{8}$$

The calculated values from Eq. (7) for various composition of indium-lead alloys are four orders of magnitude below the experimental results. These values show that secondary contributions involving the photoelectric effect are negligible.

c) *Contributions involving the pair production*

It has been assumed that this contribution proceeds via resonant excitation of the 1078 keV level during positron-K electron annihilation. The number of isomeric level excitation per unit time due to the pair production is given by

$$N_{pp}^{iso} \simeq \Phi_{NR}\, n_{pp}\, \sigma_{pp}^{o}\, l_{pp}\, n_{In}\, \sigma_{ee^+}^{e} \frac{\Gamma_{iso}}{\Gamma}. \tag{9}$$

σ_{pp}^{o} is the integrated pair production cross section$^{7)}$ for a hydrogen atom and n_{pp} is given by

$$n_{pp} = N(p_{In} Z_{In}^2 + p_{Sb} Z_{Sb}^2 + p_{Pb} Z_{Pb}^2). \tag{10}$$

l_{pp} is the average sample thickness for positrons to slow down to an energy of 83.9 keV (positron energy loss$^{5)}$ for different alloys is in the range of 18.0—20.5 MeV/cm). We have assumed an effective cross section $\sigma_{ec^+}^{e} = \sigma_{ee^+} \Gamma/\langle T_1 - T_0\rangle$ for the excitation of the 1078 keV level during positron-K electron annihilation. Free positrons are produced mainly in the interactions of the 1332 keV and 1173 keV energy photons with atoms of a sample (pair production). σ_{ee^+} is the cross section for a resonant excitation of the 1078 keV level during positron-K electron annihilation, and Γ is a total width of that level. Values of $\sigma_{ee^+} = 3.9 \times 10^{-24}$ cm^2, $\Gamma = 5.8 \times 10^{-4}$ eV $^{8)}$ and $\langle T_1 - T_0\rangle = 150$ keV have been assumed, where T_1 is the initial kinetic positron energy, and T_0 is the kinetic positron energy for the resonant excitation. The calculated values from Eq. (9) are six orders of magnitude below our experimental results.

4. *Primary photon-atom contributions*

If the nonresonant contributions are produced by direct photon- ^{115}In atom interactions, the number of the nonresonant isomeric level excitation is given by

$$N_{p}^{iso} = \Phi_{NR}\, n_{NR}\, \sigma_{NR}^{T}, \tag{11}$$

where

$$n_{NR} = N p_{In}. \tag{12}$$

Using the experimental value of the nonresonant cross section $\sigma_{NR}^{T} = 4.5 \times 10^{-32}$ cm^2 (Section 2) and known values for Φ_{NR}, N and p_{In}, we calculated N_{p}^{iso} for each alloy. The χ^2 analysis compared the experimental values N_{NR}^{iso} from Eq. (3) with the calculated values N_{p}^{iso} for five values of p_{In}. The parameter K_p was varied to minimize

$$\chi^2 = \sum \frac{[N_{NR}^{iso}(p_{In}) - K_p N_{p}^{iso}(p_{In})]^2}{N_{NR}^{iso}(p_{In})}. \tag{13}$$

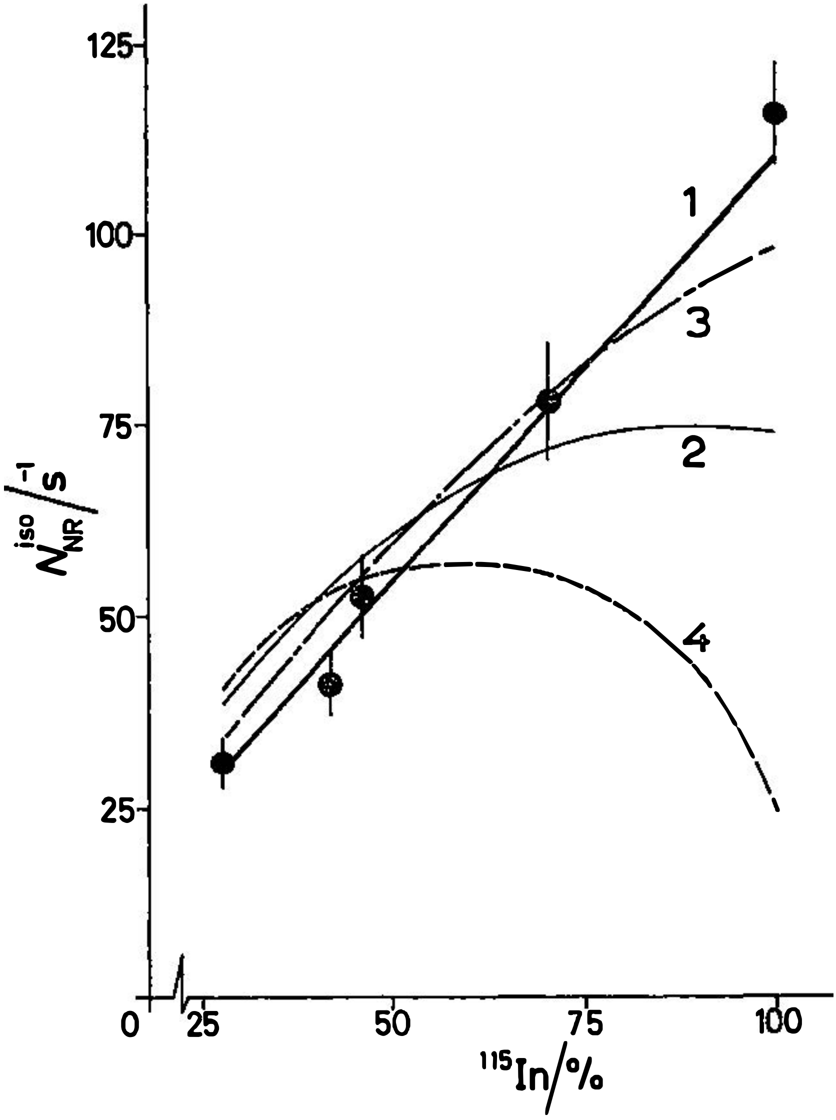

Fig. 1. A comparison of the experimental data for nonresonant contribution to the isomeric level photoactivation and calculated values for the various mechanisms, as a function of ^{115}In percent in the different alloys. The lines for the calculated values are hand drawn through five calculated values.

1. primary photon-atom contribution: $\chi^2 = 2.35$, $K_p = 0.95$;
2. pair production: $N_{pp}^{iso} \times 10^6$;
3. Compton scattering: $N_c^{iso} \times 1.7 \times 10^6$;
4. photoelectric effect: $N_{ph}^{iso} \times 1.5 \times 10^4$.

Factors 10^6, 1.7×10^6 and 1.5×10^4 for pair production, Compton scattering and photoelectric effect, respectively, are obtained by χ^2 analysis.

A value of K_p near unity would show that the nonresonant excitation was dominantly produced by the proposed interaction. The best χ^2 value of 2.35 corresponded to $K_p = 0.95$ or $\sigma_{NR}^T = 4.3 \times 10^{-32}$ cm^2. This result is convincing evidence that nonresonant interactions are produced by direct interactions between photons and ^{115}In atoms.

5. Conclusions

The distributions corresponding to the contributions of the various mechanisms are compared to the experimental results in Fig. 1. The secondary contributions from photon-atom interactions are several orders of magnitude below the value necessary to explain the nonresonant excitation, but primary photon-^{115}In atom contribution is in the excellent agreement with experiment. Additional experimental work and theoretical analyses will be needed if we are to understand the mechanisms for the nonresonant contributions.

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SEKUNDARNI DOPRINOSI NEREZONANTNOM POBUĐENJU IZOMERNOG NIVOA ^{115}In PREKO FOTON-ATOM INTERAKCIJA

MILICA KRČMAR, ANTE LJUBIČIĆ i KRUNOSLAV PISK

Institut »Ruđer Bošković«, 41000 Zagreb, Jugoslavija

BRIAN A. LOGAN

Physics Department, University of Ottawa, Ottawa, Ontario, Canada

MATIJA BISTROVIĆ

Središnji institut za tumore i slične bolesti, 41000 Zagreb, Jugoslavija

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Izvor ^{60}Co korišten je za proučavanje fotopobuđenja izomernog nivoa ^{115}In (336 keV, $T_{1/2} = 4.3$ h) u legurama indij-olovo. Istraživali smo sekundarne doprinose nerezonantnom pobuđenju preko foton-atom interakcija, uključujući Comptonovo raspršenje, fotoelektrični efekt i tvorbu para. Dobiveni rezultati pokazuju da je nerezonantno pobuđenje izomernog nivoa posljedica direktne interakcije fotona i atoma ^{115}In.