BRIEF REPORTS

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

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A ^{60}Co source has been used to photoactivate the 396 keV isomeric level ($T_{1/2} \sim 48.6$ min) of ^{111}Cd. Our results confirm the existence of nonresonant contributions.

The photoactivation of isomeric levels has been studied extensively by using experimental arrangements that involve either γ-ray sources or bremsstrahlung from electron beams. The general assumption has been that resonant energy photons excite higher nuclear levels via nuclear resonance fluorescence, and that these levels decay to populate the isomeric levels. However, some investigations have produced evidence for nonresonant γ-ray contributions.[1-4] Possible contributions due to secondary electrons produced in photon-atom interactions have been shown to be negligible.[5]

If an allowance is made for the possibility of nonresonant excitations, the probability P of exciting an isomeric level per unit time is given by

$$P=\phi_R\sigma_R(E_R)+\phi_{\rm NR}\sigma_{\rm NR}^T , \quad (1)$$

where $\sigma_R=\pi^2 g\lambda\!\!\!^{-2}\Gamma_0\Gamma_{\rm iso}/\Gamma$ represents the resonance fluorescence contribution. The parameters g, Γ_0, $\Gamma_{\rm iso}$, and Γ are, respectively, the statistical weight, the ground-state transition width, the partial width for decay to the isomeric level, and the total width of the level that is involved in the resonance fluorescence.

$\phi_R(E_R)$ is the flux of photons per unit area, energy, and time at the resonance energy. $\sigma_{\rm NR}^T$ is the total nonresonant cross section and $\phi_{\rm NR}$ is the flux of nonresonant photons per unit area and time,

$$\sigma_{\rm NR}^T=\sigma_{\rm NR}(E_{\rm iso})+\sum_i \sigma_{\rm NR}(E_i)\Gamma_{\rm iso}(i)/\Gamma_i , \quad (2)$$

where $E_{\rm iso}$ is the energy of the isomeric level and the ith level has an energy E_i ($E_i > E_{\rm iso}$), a width Γ_i, and a partial width $\Gamma_{\rm iso}(i)$ for decay to the isomeric level. The details of Eq. (2) are not known, as the mechanism for the nonresonant contribution is not understood, although it would seem reasonable to assume some interaction via an intermediate level of energy E_i that decays to the isomeric level.

The intrinsic probability of nonresonant excitation of an isomeric level is much lower than the probability of excitation via resonance fluorescence, and it is only in experimental arrangements involving intense radioactive sources that nonresonant contributions are expected to be significant. With such sources the relative intensity of higher-energy nonresonant photons is very high; in contrast, when investigations involve electron bremsstrahlung the intensity of higher-energy photons is relatively low and the nonresonant contributions will usually be small.

Several groups have also searched for nonresonant contributions. Yoshihara *et al.*[6] investigated the photoactivation of ^{115}In with a ^{60}Co source and used the Szilard-Chalmers process as a physical basis for the search for nonresonant contributions. The resonant process for ^{115}In is due to absorption by a level at 1078 keV and a photon satisfying this energy requirement imports less recoil than, for example, a 1332 keV photon involved in a nonresonant interaction.

They irradiated *tris* (acetylacetonato) indium (III) with a ^{60}Co source and searched for liberated indium atoms that they assumed would be ejected from the molecule by the nonresonant 1332 keV photons. Although there is some uncertainty in the energy necessary for rupture of the molecule, it was assumed that necessary recoil energy could be as high as 5 eV.[6,7] It is well known (the Suess effect) that because of conservation of momentum, when the indium atom is in a molecule, all of the energy imparted to the indium is not available for bond rupture.

TABLE I. A summary of the search for nonresonant contributions to the photoactivation of ^{111}Cd.

Absorber	Assumed resonance level (keV)	σ_R (10^{-29} cm^2 keV)	σ_{NR}^T (10^{-32} cm^2)	Reference
Lead	1330	5.8±0.8	1.57±0.22	2
Iron	1330	14 ±1	0.1 ±0.2	10
Lead	1190	7.8±0.8	1.0 ±0.2	This work
Iron	1190	9.7±0.6	0.7 ±0.2	
Lead	1330	6.2±0.5	1.2 ±0.2	
Iron	1330	8.3±0.5	0.8 ±0.2	

Yoshihara *et al.*[6] claim that the energy needed to break the chemical bond is reduced in molecular environments. This is a trivial error, as the rest of the article makes it clear that they do not believe this, but for the sake of clarity we should emphasize that this is not true; the energy available for chemical bond rupture is reduced because of the Suess effect. On the basis of the Suess model they calculate that nonresonant 1332 keV photons give a recoil of 5.98 eV, while a 1078 keV resonant photon gives a recoil of 3.92 eV. If it is assumed that 5 eV is necessary for bond rupture then nonresonant photons eject ^{115}In atoms and resonant photons do not. However, the Suess model is too simplistic for complex molecules. More realistic models also allow for the possibility of internal molecular vibrations and rotations.[8] Typically, this leads to 25% reductions in the energy available for bond rupture.[9] For example the 5.98 eV recoil energy calculated for 1332 keV nonresonant photons would be reduced to a value below 5 eV and the indium atom may not be ejected. Of course, there is considerable uncertainty in both the values for the energy necessary for chemical bond rupture and in the percentage reduction in the available energy due to internal molecular excitations. Values can vary by big factors, and it seems reasonable to conclude that the results of Yoshihara *et al.*[6] cannot be considered as conclusive evidence for the absence of nonresonant contributions to the photoactivation of isomeric levels.

Bikit *et al.*[10] searched for nonresonant effects in the photoactivation of ^{111}Cd with a ^{60}Co source. Their technique was based on the method used in the earlier successful searches; however, they used iron, rather than lead absorbers, to vary the relative numbers of resonant and nonresonant photons. They found no evidence for nonresonant contributions. However, more recently this group made a careful investigation of the photoactivation of ^{115}In and, in agreement with the results of Ljubičić *et al.*[1] and Krčmar *et al.*,[5] they have found clear evidence for nonresonant contributions.[11]

Anderson *et al.* have developed an intense bremsstrahlung source and have investigated the photoactivation of several isomeric levels.[12–16] Recently, they studied the photoactivation of ^{111}Cd and found no evidence for the necessity of including nonresonant contributions.[16] This is not surprising, as their technique is excellent for studying resonant excitations, but the linear falloff of the bremsstrahlung spectrum results in there only being a relatively low intensity of high-energy photons, and the contribution of nonresonant effects cannot be observed. Their statistical accuracy is also rather limited. It should be emphasized that they made no specific investigations of nonresonant effects, and their conclusions are simply based on their result for the photoactivation cross section being in general agreement with previous results.

One very interesting aspect of their work is that they show that a resonance level at 1190 keV in ^{111}Cd makes a strong contribution to the photoactivation of the isomeric level. Before this analysis it had been assumed that a resonance level at 1330 keV was dominant, and earlier searches for nonresonant contributions were analyzed on this assumption.[2,10]

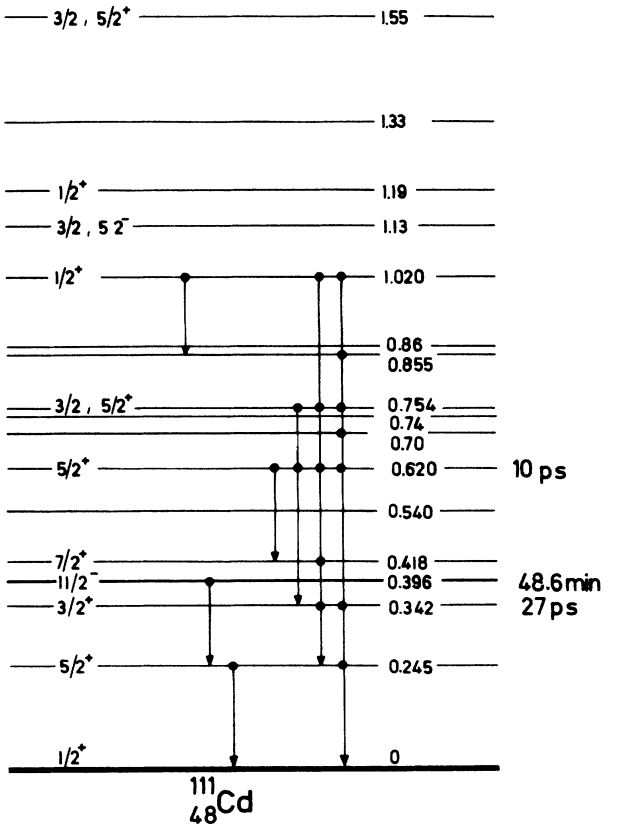

FIG. 1. The level structure and γ-ray branching scheme for ^{111}Cd.

We have repeated an investigation of the photoactivation of the 396 keV isomeric level ($T_{1/2} \sim 48.6$ min) of ^{111}Cd and have made analyses that assumed resonant interactions with both the 1190 and the 1330 keV levels. We also used both iron and lead absorbers in the technique we developed in our earlier investigations.

Our experimental technique has been described in some detail,[1-5] and it allows us to distinguish between activation via resonance fluorescence and by nonresonant processes by interposing absorbers between the source and sample during the photoactivation exposures. The Compton scattering and absorption in the absorbers distorts the photon spectrum and changes the relative values of $\phi_R(E_R)$ and ϕ_{NR}. In the photoactivation exposures the relative number of resonant excitations increases with the thickness of the absorber, and it is possible to distinguish between resonant and nonresonant contributions. The ratio of the number of resonant energy photons per unit energy to the number of full energy photons, as a function of absorber thickness, were obtained using a relatively weak ($\sim 10^5$ Bq) ^{60}Co source that only emits full energy photons and by observing the variation in the energy spectrum with a Ge(Li) detector as the absorber thickness is varied. The relative values of $\phi_R(E_R)$ and ϕ_{NR} also depend on the tail of low-energy photons emerging from the intense ^{60}Co source. We used a 2.05×10^{14} Bq source that was specified to have a low-energy intensity k that was 0.12 of the full energy intensity.

The decay scheme of ^{111}Cd is shown in Fig. 1. A cadmium disk 0.45 cm thick and 3 cm in diameter, was irradiated for 13 h at a distance of 34 cm from the source. We used iron absorbers with total thicknesses of 0.5, 1.5, 2.5, and 4 cm. For the lead we used thicknesses of 0.4, 0.8, 1.2, and 1.6 cm. Before monitoring the induced photoactivation the disks were "cooled" for 15 min. The induced photoactivity was determined from the intensity of the 245 keV γ ray measured in a Ge(Li) detector. In the case of both the iron and lead absorbers we did two analyses; one in which we assumed the resonance fluorescence proceeded via the 1190 keV level, and another in which we assumed the fluorescence was via the 1330 keV level.

A χ^2 analysis of the experimental data was made with k, σ_R, and σ_{NR}^T as variable parameters. The comparisons were relatively insensitive to k, and the best k value of 0.12 was in agreement with the source specifications. The sensitivity of the analyses is illustrated in Fig. 2.

The results of searches for nonresonant contributions to the photoactivation of ^{111}Cd are summarized in Table I. It can be seen that our results are reasonably consistent and suggest a σ_{NR}^T value of about 10^{-32} cm^2. We have no explanation of the result of Bikit *et al.*[10]

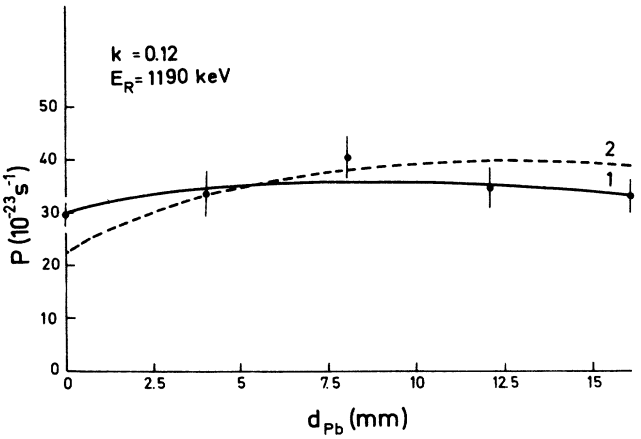

FIG. 2. A comparison of the experimental and calculated values of the ^{111}Cd isomeric level excitation probability, assuming a resonance level at 1190 keV, as a function of d_{Pb}, the lead absorber thickness. The values obtained for χ^2, σ_R, and σ_{NR} are the following. (1) $\chi^2=1.1$, $\sigma_R=7.8\times10^{-29}$ cm^2 keV, $\sigma_{NR}^T=1.0\times10^{-32}$ cm^2; (2) $\chi^2=8.0$, $\sigma_R=11\times10^{-29}$ cm^2 keV, $\sigma_{NR}^T=0$.

The recent work of Collins *et al.*[16] gave a $\sigma_R=(9.8\pm2.5)\times10^{-29}$ cm^2 keV with no evidence for nonresonant contributions. As already discussed, this system is not sensitive to nonresonant contributions. Collins *et al.* assume the 1190 keV level is the dominant fluorescent level. Our values for σ_R, based on fluorescence via the 1190 keV level, are in excellent agreement with the results of Ref. 16 but achieve a superior statistical accuracy. Our results, assuming fluorescence of the 1330 keV level, are also in general agreement. This is to be expected as our earlier investigations[2] showed that the relative value of the lower-energy flux to ϕ_{NR} only varied slightly over the energy range of 1196–1309 keV. Most importantly, our experimental arrangement is also sensitive to the nonresonant contributions that show their presence very clearly.

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