# Measurements of the electron-photon double decay in <sup>113</sup>In at 35°\*

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Double- and triple-coincidence measurements of the electron-photon double decay of the 392-keV state in <sup>113</sup>In were performed at a relative angle of emission of 35°. The method of three-dimensional analysis was applied to record the amplitudes of pulses from the electron and photon detectors and the time difference in the occurrence of pulses.  $K \gamma$  decay was well resolved from higher-shell decays, but  $L \gamma$ ,  $M \gamma$ , and  $N \gamma$  decays were not resolved. The energy spectrum of photons for  $K \gamma$  and  $(L + M + N) \gamma$  decays was derived from the experimental data. The results are in agreement with the theory of the internal Compton effect of Spruch and Goertzel.

RADIOACTIVITY <sup>113</sup>In<sup>m</sup>; measured  $e\gamma$  double decay at 35°; deduced photon energy distribution in the range 17.6 to 248.4 keV for  $K \gamma$  decay and 37.8 to 163.9 keV for  $(L + M + N) \gamma$  decay.

### INTRODUCTION

The results of measurements of conversion-coefficient ratios for higher electron shells and subshells for the 392-keV isomeric transition in <sup>113</sup>In, when compared with the theoretical values, lead to the conclusion that it is a highly pure M4transition, and that the E5 admixture is most probably absent.<sup>1</sup> This transition is hindered by a factor of about 5.5 when compared with the Weisskopf single-particle estimate. Therefore, this transition is very suitable for studying higherorder processes. Among these processes, which are difficult to observe, a considerable number of experiments have been performed in the search for two-quantum  $(\gamma\gamma, e\gamma, and ee)$  decays. Attempts have so far been made to observe doubleelectron  $(ee)^2$  and double-photon  $(\gamma\gamma)$  emission<sup>3</sup> in the decay of the 392-keV state in <sup>113</sup>In, but only upper limits of transition probabilities have been determined.

The  $e\gamma$  process can proceed via nuclear and via electronic intermediate states. According to theoretical estimates<sup>4</sup> the former mechanism has a transition probability which is very small. The latter mechanism in lowest order is the internal Compton effect (ICE). This process and the internal bremsstrahlung in  $\beta$  decay, which is closely related to it, have been theoretically studied by several authors.<sup>5-7</sup> Transitions have been experimentally investigated in several nuclei.<sup>8-10</sup> However, only measurements of the  $e\gamma$  decay of the 662-keV isomeric state in <sup>137</sup>Ba allowed a detailed check of the theory.<sup>9</sup>

In the theory of Spruch and Goertzel<sup>6</sup> the differential coefficient of the ICE for magnetic transitions is calculated in the Born approximation, assuming a point nucleus. The angular and energy distributions calculated from the Spruch-Goertzel theory are in very good agreement with the experimental data for the transition in <sup>137</sup>Ba. Some deviations were observed at low photon energies, but a remeasurement using new experimental techniques is required to check the experimental results. Such agreement was not expected because the condition for the validity of the Born approximation is not well satisfied for this transition. For these reasons it seemed of interest to extend measurements to the 392-keV *M*4 transition in <sup>113</sup>In.

#### APPARATUS AND MEASUREMENTS

A new system was built for measurements of double-decay processes, including a cold-finger vacuum chamber and a three-dimensional 256  $\times 256 \times 256$ -channel analyzer.<sup>11</sup> A block diagram of the apparatus is shown in Fig. 1. Here we report on three measurements of the  $e\gamma$  decay of the 392-keV state in <sup>113</sup>In, all at a relative angle of emission of electrons and photons of about 35°. For each coincidence event the analyzer recorded the amplitudes of pulses from the electron and photon detectors and the time difference of their occurrence. Electrons and photons from  $e\gamma$  decay have continuous energy distributions because they share the decay energy, but for each electron shell the sum energy is constant, i.e.,

$$E + E_e = W - B_e. \tag{1}$$

Here E and  $E_e$  are the photon and the electron energy, respectively, W is the decay energy, and

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 $B_e$  is the electron binding energy. This relation, together with the condition of simultaneity were used to identify  $e\gamma$  events.

Carrier-free <sup>113</sup>Sn (NEN Chemical Company, Boston, Massachusetts) of a specific activity of about 7.35 mCi/mg in a 4 N HCl solution was used to prepare the sources. In each preparation a small drop of the solution was micropipetted onto a thin Zapon foil about 100  $\mu$ g/cm<sup>2</sup> thick and about 20 mm ×20 mm in area mounted on a holder made of Plexiglass. After drying, the spot was about 3 mm in diameter and the average thickness of SnCl<sub>2</sub> was about 8  $\mu$ g/cm<sup>2</sup>. To reduce effects of electrostatic charges on the foil, a strip of aluminium 5 mm wide and of about 15- $\mu$ g/cm<sup>2</sup> thickness was previously evaporated in vacuum onto the Zapon foil. The source was mounted inside the



FIG. 1. Block diagram of the apparatus (PA, preamplifier; FA, fast amplifier; FD, fast discriminator; CFTD, constant fraction timing discriminator; DL, delay line; FC, fast coincidence; LA, linear amplifier; TAC, time-to-amplitude converter; ADC, analog-todigital converter).

vacuum chamber on a cold-finger cooled plate in order to reduce the evaporation of SnCl<sub>2</sub>.

A surface-barrier detector of a sensitive area of 12 mm in diameter was used to detect electrons. It was mounted inside the vacuum chamber and also cooled by a cold finger to improve its performance. The energy resolution of the detector was about 6 keV for the 364-keV K conversion electrons emitted from the source. In the first measurement the detector was at a distance of 62 mm from the source, and in the second and third measurements it was at a distance of 88 mm. In order to prevent the scattering from one detector into another, lead shields were placed between the electron detector and the other detectors used in the measurements.

In the first measurement a true coaxial Ge(Li) detector (made in the "Boris Kidrič" Institute, Belgrade) of a sensitive volume of 25 cm<sup>3</sup> was used to detect photons. It had a resolution of 2.5 keV for the 122-keV  $\gamma$  rays. It was placed outside the vacuum chamber at a distance of 104 mm from the source. To reduce the absorption of low-energy photons, a vacuum window made of an aluminium foil 0.2 mm thick was positioned in front of the detector. Double coincidences of electron and photon pulses were measured.

In the second measurement we made an attempt to determine the low-energy part of the photon spectrum. A closed-end coaxial Ge(Li) detector of a sensitive volume of 8  $cm^3$  (Canberra model 7221) and a resolution of 1 keV for the 122-keV  $\gamma$  rays was used. It had a thin beryllium window and a detection efficiency of 20% for the 6.45-keV x rays emitted in the decay of <sup>57</sup>Co. A new upper part of the vacuum chamber was made which had a vacuum port into which the Canberra Ge(Li) detector was placed, sealed by vacuum O rings. In this way the absorption of low-energy photons was minimized. The detector was at a distance of 100 mm from the source. However, the aim of this measurement was not achieved because the background due to a large number of real coincidences of K conversion electrons and K x rays prevented the determination of  $e\gamma$  events below the x-ray line of 27 keV.

Therefore, the third measurement was performed. An x-ray scintillation counter was introduced to the arrangement of the second measurement and triple-coincidence events were recorded. It was a packed NaI(T1) crystal (38 mm diam  $\times$ 3 mm thick) mounted on an XP-1020 photomultiplier tube and placed close to a recessed vacuum window made of an aluminium foil 0.2 mm thick. The NaI(T1) crystal was placed 25 mm above the source.

The time spectra obtained from a time-to-amplitude converter were fairly asymmetrical. The relatively steep side of the peaks was due to the electron detector. The relatively long tails were mainly due to the coincident detection of low-energy photons (K x rays) in both Ge(Li) detectors or in the NaI(T1) scintillation detector. The time resolution  $(2\tau)$  of the electron and 25-cm<sup>3</sup> Ge(Li) detectors, the electron and 8-cm<sup>3</sup> Ge(Li) detectors, and the electron and NaI(T1) detectors was 35, 50, and 15 ns, respectively.

The counting rate in the three measurements



FIG. 2. Two-dimensional spectra of photon versus electron energy for four consecutive (out of seven) time intervals each 32 nsec wide. The peaks above the dotted lines (upper end of each diagram) correspond to constant energies of electrons of 364 and 388 keV and are due to accidental coincidences of conversion electrons and  $\gamma$ rays. The  $K \gamma$  data appear above the line of constant sum energy of 364 keV (dashed line) and the  $(L + M + N) \gamma$  data above the line of constant sum energy of 388 keV (dashdotted line). These data are prominent in the diagram (b), corresponding to true coincidence events.

was about 10, 140, and 2 coincidences per minute, respectively.

### ANALYSIS OF THE RESULTS

The data were recorded on punched paper tape and analyzed off line in a CAE-9040 computer. One-dimensional projections were used to recheck the stability and energy scales. The number of counts due to  $e\gamma$  events was deduced from twodimensional diagrams, each corresponding to an interval of time difference. Time intervals of 32 ns were usually chosen, and seven two-dimensional diagrams were obtained from the computer. Four such diagrams of photon energy versus electron energy are shown in Fig. 2. In the first measurement the system was adjusted to have the same energy interval of 1.5 keV per channel in both the electron and photon dimension, while in the second and third measurements the energy interval was four times larger in the electron (1.6 keV) than in the photon dimension (0.4 keV).

To simplify the analysis of data, two-dimensional diagrams of photon energy versus sum energy were made in the computer, each corresponding again to a time interval of 32 ns. In these diagrams  $K \gamma$  events appeared along a constant sum energy of 364 keV, while events due to the L, M, and N electron shells, which could not be resolved, appeared along the line of constant sum energy of 388 keV. The lines were quite distinct and well separated. The number of counts for some value of the photon channel was obtained by adding the contents in six channels of the sum-energy line and by subtracting the contents in three adjacent channels to the left (lower sum energy) and three to the right. The data were concentrated in one time interval (see Fig. 2), but some appeared also in adjacent intervals. Therefore, the results from three or four time intervals were added. When these numbers were plotted against the photon channel number, the diagram of photon energy distribution was obtained.

The external bremsstrahlung of conversion electrons in the source and in surrounding materials "viewed" by both the electron and photon detectors would yield coincident electron and photon pulses which could not be distinguished from  $e\gamma$  events. Seykora<sup>10</sup> measured the contribution of bremsstrahlung to the continuous photon spectrum in the decay of the 662-keV state in <sup>137</sup>Ba and of the 279-keV state in <sup>203</sup>Tl by placing aluminium foils of 30 to 200 mg/cm<sup>2</sup> at the source. He showed that bremsstrahlung was negligible. In this experiment, the bremsstrahlung in the source and backing was completely negligible because of the very small thickness. The contribution from the

scattering in the walls of the scattering chamber was greatly reduced by the shielding and the conical collimator around the Ge(Li) detector. Owing to the degradation of energy of electrons in the walls a continuous sum-energy distribution up to the limiting value of 364 keV would be obtained. From the counting rates at sum energies immediately below the peaks at 364 keV, taking into account the response curves of the detectors and the procedure of calculation of the numbers of counts in the peaks (preceding paragraph), we estimated that the contribution of the external bremsstrahlung to the photon distribution of our measurements is less than 3%.

The differential coefficient of  $e\gamma$  double decay, defined as the ratio of the differential  $e\gamma$  transition probability  $dT_{e\gamma}(E,\Theta_0)/dEd\Omega$  to the electronconversion transition probability  $T_e$ , was calculated from the data obtained in double-coincidence measurements by the formula

$$dB_{e\gamma}(E,\Theta_0)/dEd\Omega = N_{e\gamma}(E,\Theta_0)/N_e\Omega(E)\Delta E\epsilon_{e\gamma}C(E,\Theta_0).$$
(2)

Here the index e stands for K or L+M+N;  $N_{e\gamma}(E, \Theta_0)$  is the number of  $e\gamma$  events obtained as described above, calculated per unit time, at the photon energy E and at the nominal angle  $\Theta_0$  of  $35^{\circ}$ .  $N_e$  is the number of K or L+M+N conversion electrons detected per unit time in the silicon surface-barrier detector,  $\Omega(E)$  is the effective solid angle of the photon detector (including the geometrical, absorption, and detection-efficiency factors),  $\Delta E$  is the width of the photon energy interval,  $\epsilon_{e\gamma}$  is the coincidence efficiency, and  $C(E, \Theta_0)$  is the correction factor that accounts for the finite geometry of the electron and photon



FIG. 3. Detection efficiency  $\Omega(E)$  of Ge(Li) detectors including the solid-angle, absorption, and intrinsic efficiency factors, determined by means of calibrated sources of <sup>113</sup>Sn, <sup>57</sup>Co, <sup>241</sup>Am, <sup>22</sup>Na, and <sup>137</sup>Cs and uncalibrated sources of <sup>75</sup>Se and <sup>169</sup>Yb.

detectors.

The effective solid angle of the photon detector  $\Omega(E)$  was determined by a direct calibration of each Ge(Li) detector in the same geometry as in the experiment, using a set of calibrated  $\gamma$ -ray sources of <sup>113</sup>Sn, <sup>57</sup>Co, <sup>241</sup>Am, <sup>22</sup>Na, and <sup>137</sup>Cs

TABLE I.  $K \gamma$  and  $(L + M + N) \gamma$  differential coefficients in the  $e\gamma$  decay of the 392-keV state in <sup>113</sup>In [in units of  $10^{-3} (mc^2 \text{ sr})^{-1}$ ] obtained in the first measurement (double coincidences, detector A).

E	$dB_{K\gamma}(E,\Theta_0=35^\circ)$	$dB_{(L+M+N)\gamma}(W,\Theta_0=35^\circ)$
(keV)	$dEd\Omega$	dEdΩ
33.2	$3.21 \pm 0.34$	
37.8	$2.59 \pm 0.28$	$3.32 \pm 0.72$
42.3	$2.62 \pm 0.28$	$3.00 \pm 0.70$
46.8	$2.16 \pm 0.25$	$1.56 \pm 0.59$
51.3	$\textbf{2.14} \pm \textbf{0.25}$	$1.91 \pm 0.58$
55.9	$\textbf{1.88} \pm \textbf{0.25}$	$1.79 \pm 0.47$
60.4	$1.45 \pm 0.23$	$2.96 \pm 0.55$
64.9	$1.26 \pm 0.21$	$1.84 \pm 0.46$
69.5	$1.59 \pm 0.23$	$1.66 \pm 0.46$
74.0	$\textbf{1.59} \pm \textbf{0.23}$	$1.48 \pm 0.46$
78.5	$\textbf{0.63} \pm \textbf{0.19}$	$1.38 \pm 0.46$
83.1	$1.11 \pm 0.20$	$0.83 \pm 0.28$
87.6	$1.02 \pm 0.20$	$0.92 \pm 0.37$
92.1	$1.11 \pm 0.20$	$1.57 \pm 0.37$
99.6	$1.22 \pm 0.20$	$0.56 \pm 0.29$
104.2	$1.18 \pm 0.21$	$0.43 \pm 0.22$
109.5	$0.52 \pm 0.12$	$0.30 \pm 0.15$
115.6	$0.51 \pm 0.11$	$0.00 \pm 0.16$
121.6	$0.23 \pm 0.09$	$0.41 \pm 0.24$
127.6	$0.46 \pm 0.12$	$0.59 \pm 0.26$
133.7	$0.25 \pm 0.10$	$0.81 \pm 0.36$
139.7	$0.41 \pm 0.11$	$0.37 \pm 0.37$
145.8	$0.41 \pm 0.11$	$0.10 \pm 0.29$
151.8	$0.46 \pm 0.12$	$0.20 \pm 0.30$
157.8	$0.33 \pm 0.14$	$0.31 \pm 0.31$
163.9	$0.15 \pm 0.12$	$0.43 \pm 0.32$
169.9	$0.44 \pm 0.13$	
175.9	$0.00 \pm 0.13$	
181.9	$0.17 \pm 0.11$	
188.0	$0.34 \pm 0.12$	
194.1	$0.21 \pm 0.12$	
200.1	$0.18 \pm 0.12$	e e
206.1	$0.06 \pm 0.12$	
212.2	$0.06 \pm 0.12$	
218.2	$0.17 \pm 0.13$	
224.3	$0.03 \pm 0.17$	
230.3	$0.03 \pm 0.14$	
236.4	$0.15 \pm 0.11$	
242.4	$0.04 \pm 0.11$	
248.4	$0.04 \pm 0.11$	

(supplied by the International Atomic Energy Agency, Vienna) and uncalibrated sources of <sup>75</sup>Se and <sup>169</sup>Yb. The results are shown in Fig. 3.

The correction factor  $C(E, \Theta_0)$  was calculated from the window function of the electron- and photon-detector areas, as viewed from the source at the nominal angle  $\Theta_0$  of 35° and assuming the theoretical angular distribution of Spruch and Goertzel.<sup>6</sup> Values of 0.97 and 0.98 were obtained for this factor in the first and in the second and third measurement, respectively.

From the triple-coincidence data the  $e\gamma$  differential coefficient (for the K shell) was obtained using the formula

$$dB_{K\gamma}(E,\Theta_0)/dEd\Omega$$
  
=  $N_{K\gamma x}(E,\Theta_0)\epsilon_{Kx}/N_{Kx\gamma}(E)\Delta E\epsilon_{K\gamma x}C(E,\Theta_0),$   
(3)

where  $N_{K\gamma x}(E, \Theta_0)$  is the number of measured triple-coincidence  $K \gamma$  events per unit time and  $N_{Kx}$  is the number of double coincidences of K xrays and K conversion electrons per unit time. The value of  $N_{Kx}$  was measured during regular checks of the apparatus and it amounted to about eight coincidences per second.

In the measurements the coincidence unit was adjusted to a relatively long resolving time of about 200 nsec. For this reason, taking into account the resolving times of the detectors, which are much smaller, it was assumed that the ef-

TABLE II. K  $\gamma$  and  $(L + M + N) \gamma$  differential coefficients in the  $e\gamma$  decay of the 392-keV state in <sup>113</sup>In [in units of  $10^{-3}(mc^2 \text{ sr})^{-1}$ ] obtained in the second measurement (double coincidences, detector B).

E (keV)	$\frac{dB_{K\gamma}(E,\Theta_0=35^\circ)}{dEd\Omega}$	$\frac{dB_{(L+M+N)\gamma}(E,\Theta_0=35^\circ)}{dEd\Omega}$
32.0	$4.31 \pm 0.42$	
36.8	$3.72 \pm 0.34$	
41.6	$3.65 \pm 0.35$	
<b>46.4</b>	$2.76 \pm 0.30$	$5.57 \pm 1.24$
51.2	$2.63 \pm 0.29$	$3.27 \pm 1.00$
56.0	$2.41 \pm 0.27$	$4.08 \pm 0.87$
60.8	$1.53 \pm 0.23$	$2.28 \pm 0.87$
65.6	$1.41 \pm 0.22$	$1.47 \pm 0.82$
70.4	$1.28 \pm 0.23$	$2.43 \pm 0.89$
75.2	$1.50 \pm 0.26$	$2.85 \pm 0.82$
80.0	$\textbf{1.12}\pm\textbf{0.20}$	$1.61 \pm 0.67$
84.8	$1.14 \pm 0.20$	$1.20 \pm 0.60$
89.6	$1.04 \pm 0.19$	$1.04 \pm 0.58$
94.4	$\textbf{1.04} \pm \textbf{0.19}$	$0.21 \pm 0.55$
99.2	$0.84 \pm 0.19$	$1.16 \pm 0.51$
104.0	$0.83 \pm 0.18$	$0.49 \pm 0.51$

ficiencies of the coincidence unit in double- and triple-coincidence measurements ( $\epsilon_{e\gamma}$ ,  $\epsilon_{Kx}$  and  $\epsilon_{K\gamma x}$ ) were equal to one.

## **RESULTS AND DISCUSSION**

The results for  $e\gamma$  decay on K electrons are shown in Tables I, II, and III and in Fig. 4. Figures 4(a) and 4(b) show averages taken over three channels ( $\Delta E = 4.5 \text{ keV}$ ) for photon energies up to 100 keV and over four channels ( $\Delta E = 6.0 \text{ keV}$ ) for higher energies. Figure 4(c) shows averages over 20 channels ( $\Delta E = 8.0 \text{ keV}$ ) except for the lowest-energy point, which is an average over 18 channels ( $\Delta E = 7.2 \text{ keV}$ ). This point for the photon energy interval from 14 to 21.2 keV is the only result that could be derived below the x-ray lines from triple-coincidence measurements. In the region from 21.2 to 28.8 keV the background due to accidental coincidences of x rays and of coincident events of x rays and conversion electrons was too high.

The full curves in Fig. 4 represent the distributions calculated from the Spruch and Goertzel theory of the ICE on K electrons for the same transition at a relative angle of emission of 35°, averaged over the same energy intervals as the experimental data. The results of all three measurements of  $K \gamma$  decay at photon energies above 29 keV are in good agreement with the theory of Spruch and Goertzel. Therefore, we consider the  $K \gamma$  decay of the 392-keV state in <sup>113</sup>In to proceed mainly as the ICE.

The condition for the validity of the Born approximation applied in the theory of Spruch and Goertzel to the low-energy region of photons  $(m\alpha Z/P \ll 1)$  is satisfied to a lesser degree in the  $K \gamma$  decay of <sup>113</sup>In  $(m\alpha Z/P \approx 0.28)$  than in the

TABLE III.  $K\gamma$  differential coefficients in the decay of the 392-keV state in <sup>113</sup>In [in units of  $10^{-3} (mc^2 \text{ sr})^{-1}$ ] obtained in the third measurement (triple coincidences, detector B).

<i>E</i> (keV)	$\frac{dB_{K\gamma}(E,\Theta_0=35^\circ)}{dEd\Omega}$
17.6	15.8±3.7
30.4	$6.92 \pm 1.34$
38.4	$2.72 \pm 0.65$
46.4	$3.97 \pm 0.75$
54.4	$2.17 \pm 0.60$
62.4	$2.12 \pm 0.59$
70.4	$2.33 \pm 0.58$
78.4	$1.40 \pm 0.47$
86.4	$1.05 \pm 0.47$
94.4	$1.41 \pm 0.47$
102.4	$1.20 \pm 0.48$



FIG. 4. Differential coefficients of  $K \gamma$  decay in <sup>113</sup>In as a function of the photon energy E obtained in the three measurements at 35°. The solid lines were calculated from the Spruch and Goertzel theory which was modified by taking averages over energy intervals of the same width as the presented experimental data.

662-keV K  $\gamma$  decay of <sup>137</sup>Ba ( $m\alpha Z/P \approx 0.22$ ). This indicates that the theory is valid in a broader range than originally expected (at least for M4 transitions). The single point at 17.6 keV lies above the theoretical curve by a factor of 1.82  $\pm 0.43$ . This is not a significant deviation, but it may be interpreted as an indication that the theory is valid in this energy range, or even as an indication of a higher number of low-energy photons than predicted by the theory. This may be due to the emission of two (or more) soft photons by the outgoing electron (e.g.,  $K \gamma \gamma$  decay).

Results for  $e\gamma$  decay on higher-shell electrons were also obtained in double-coincidence measurements. The shells were not resolved and therefore they are indicated as  $(L + M + N) \gamma$  decay. The *L*-shell electrons are estimated to give a major contribution to the decay. The  $(L + M + N) \gamma$ differential coefficient was calculated from the experimental data in the same manner as for  $K \gamma$ 



FIG. 5. Differential coefficients of  $(L + M + N) \gamma$  decay in <sup>113</sup>In as a function of the photon energy *E* obtained in double-coincidence measurements at 35°. The solid lines were calculated from the Spruch and Goertzel theory using formula (4) and modified by taking averages over energy intervals of the same width as the presented experimental data.

decay. The results from the first and second measurements are shown in Tables I and II (last columns) and in Fig. 5. To the knowledge of the authors, a detailed theoretical calculation of the ICE on higher-shell electrons has not been made. Spruch and Goertzel have estimated the differential coefficient for the ICE for the L shell to be approximately equal to the coefficient for the K shell, i.e.,

$$dB_L(E\Theta)/dEd\Omega \approx dB_K(E,\Theta)/dEd\Omega . \tag{4}$$

The full curves in Fig. 5 show the results of the calculation. The experimental results are somewhat higher than predicted by the theory.

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