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Decay of In^{114m} and In^{114}†

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Scintillation counter experiments have been performed on the 72-second activity of In^{114} produced by (p,n) and $(d,2n)$ cyclotron bombardment of enriched Cd^{114} and on the 49-day activity of In^{114m} produced by neutron activation. The well-known 556–722-kev cascade radiation of Cd^{114} is shown to be excited by a K-capture transition from the 49-day isomeric level. The weak 1300-kev gamma ray previously assigned to Cd^{114} is shown to come from the first excited state of Sn^{114} and is in coincidence with a low-energy beta spectrum having an end point of 675±40 kev. No gamma rays in Cd^{114} other than the 556- and 722-kev lines have been observed from the decay of In^{114m} or In^{114}. A 1.9% K-capture branch from In^{114} to the ground state of Cd^{114} has been found. A positron branch of ~0.004% between these two states is shown to have an end point of 400±25 kev.

I. INTRODUCTION

In recent years several investigations have been conducted on the radiations from the 72-second ground state and the 49-day isomeric state of In^{114}.[1–10] The results of Brazos and Steffen[7–9] on the polarization-directional correlation of the well-known 556–722-kev cascade in Cd^{114} indicate that the spin of the state at 1278 kev is probably 4^+ and that of the 556-kev state 2^+. It is known that the isomeric state of In^{114} has a spin 5,[10] and thus it would be expected that this cascade would be excited by electron capture from this state only and not from the 72-second ground state of spin 1 as has been reported.[2] Gamma rays of weak intensity having energies of 576±3, 1271±6, 1300±3 have been reported and were also assigned to the K-capture branch.[4–6] The reported observation of 556–1300 kev coincidences indicated the existence of a state in Cd^{114} at 1856 kev.[4,6]

Kinsey and Bartholomew[11] did not observe a high-energy gamma ray from the $Cd^{113}(n\gamma)Cd^{114}$ reaction which would correspond to the reported level at 1856 kev, although their data do suggest the existence of levels in Cd^{114} at 563±8, 1205±14, 1320±11 and 1381±11 kev. Preliminary coincidence measurements by Bernstein[12] at Brookhaven National Laboratory failed to verify the existence of 556–1300 kev coincidences. The 556–1300 kev coincidences were also reported to be much weaker than previously suggested by the work of Lu *et al.*[13] Because of these discrepancies and the necessity of knowing the energy level scheme of Cd^{114} in arranging the many gamma rays from the reaction $Cd^{113}(n\gamma)Cd^{114}$ [14] into a reliable decay scheme, the present work on In^{114} was undertaken. Scintillation counter experiments were performed on the 72-second activity produced by (p,n) and $(d,2n)$ cyclotron bombardment of 83% enriched Cd^{114} and on the 49-day In^{114m} produced by neutron capture in 65% enriched

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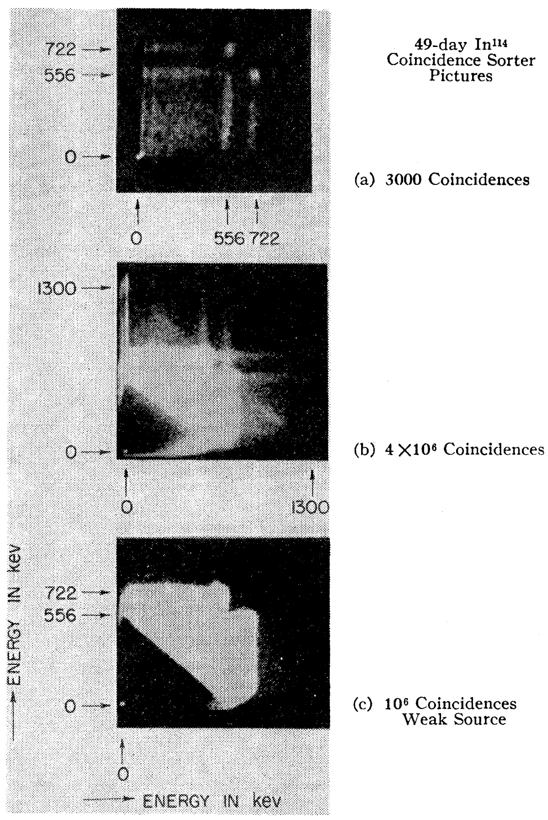

FIG. 1. In^{114m} gamma-gamma coincidences pulse photographed with the coincidence sorter (see text).

In^{113}.[15] A summary of the results was presented at the 1955 Chicago American Physical Society.[16]

II. UPPER LIMIT FOR THE 556–1300 kev TRANSITIONS

In order to determine if this cascade transition exists, a coincidence sorter[17] was used which records all coincidences on a single photographic image. In using this instrument the source is placed between two NaI(Tl) crystals each mounted on a photomultiplier as in the conventional coincidence scintillation spectrometer. When a coincidence occurs the pulses from each of the two crystals are stretched at their maxima and applied to the X and Y axes of an oscilloscope. The intensifier of the oscilloscope is activated for a few microseconds after both pulses have reached their maxima, thus producing a spot on the oscilloscope face whose $X-Y$ position corresponds to the pulse heights in the NaI crystals due to the gamma rays producing the coincidence. A long-time exposure on polaroid film is taken in order to obtain the pattern of dots. Figure 1 shows the results of three exposures taken with weak 49-day In^{114m} sources and $1\frac{1}{2}\times 1$ in. NaI(Tl) crystals. Figure 1(a) was produced by 3000 coincidences. The two white spots correspond to the coincidence of photopeaks of 556 kev and 722 kev; the picture is symmetrical about 45° corresponding to $X=556$ kev, $Y=722$ kev and *vice versa.* The horizontal and vertical lines are the result of coincidences between the 556-kev photopeak and the 722 Compton distribution and vice versa. Figure 1(b) is an exposure of 4×10^6 coincidences. The fact that the 1300-kev gamma ray is in coincidence with both the 556- and 722-kev gammas shows that the coincidences are probably accidental. This conclusion is substantiated by the lack of such coincidences in Fig. 1(c) which contains 10^6 coincidences from a weaker source in which the true to chance rate is approximately ten times greater. An estimate of the minimum number of coincidences observable in such a picture allows the determination of an upper limit for the 556–1300 kev coincidences. The value obtained is $(556-1300)/(556-722)\leq 0.001$, which is 2% of the value reported by Johns *et al.*[6]

A confirmation of the absence of 556–1300 kev coincidences was made with the use of a 20-channel analyzer. Figure 2 shows the resulting gamma-ray distribution in coincidence with 1300- and 722-kev gamma rays as well as the singles spectrum for the same geometry. Assuming that all of the 722–1300 kev coincidences observed are random, it is found that $(556-1300)/(556-722)\leq 0.0004$, in agreement with the coincidence sorter result.

Since the above work was completed, Johns[18] has communicated to us results of a re-examination of the decay of 49-day In^{114m}. From external conversion measurements he finds no evidence for the 576-kev gamma ray previously assigned to the K-capture branch. He also finds no 1300–556 kev coincidences, in agreement with the results described above. We, therefore, conclude that there is no evidence for a level at 1856 kev in Cd^{114} from the decay of In^{114m}.

III. THE 1300-KEV STATE IN Sn^{114}

The intensity of the 1300-kev gamma ray in the singles spectrum of In^{114m} was determined by means of

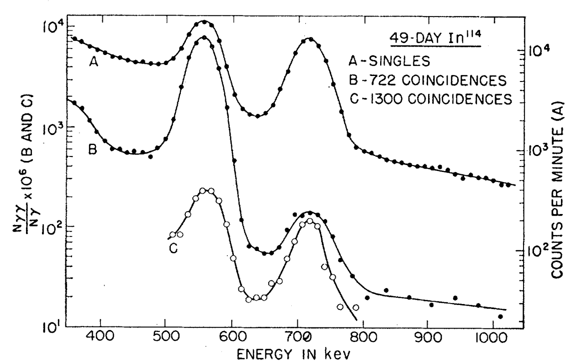

FIG. 2. Twenty-channel analyzer data showing presence of 556–722 kev coincidences and absence of 1300–556 kev coincidences.

[15] Enriched isotopes obtained from the Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

[16] L. Grodzins and H. T. Motz, Phys. Rev. **100**, 1236(A) (1955).

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[18] Johns, Williams, and Brodie, Can. J. Phys. **34**, 147 (1955).

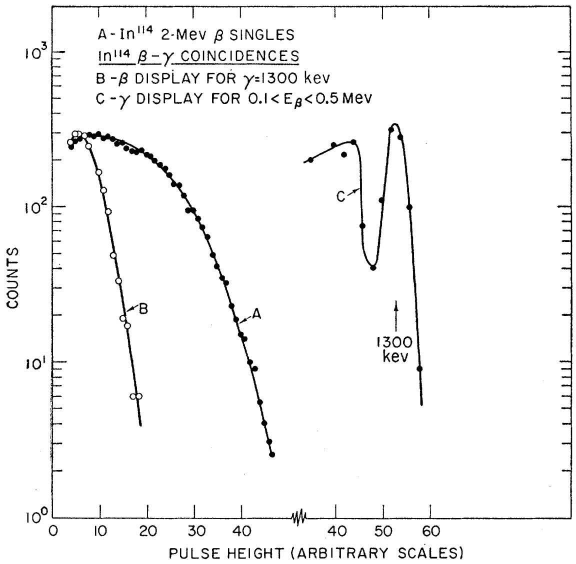

FIG. 3. β-γ coincidence spectra obtained with 49-day In^{114m} source.

a $1\frac{1}{2}\times1$ in. NaI(Tl) crystal, to be $0.09\pm0.01\%$ per disintegration. K x-ray, gamma-ray coincidence measurements were performed on In^{114m} to find out whether this gamma ray is a transition in Cd^{114}. The gamma detection crystal was placed 8 in. from the source to reduce the sum peak (722+556 kev) to a negligible contribution and the relative intensities of the 556-kev, 722-kev, and 1300-kev gamma rays in coincidence with K x-rays were compared with the singles intensity distribution in the same geometry. No 1300-kev gamma rays were found in prompt coincidence with K x-rays.

Beta-gamma coincidences were then investigated by using very weak sources such that the true coincidence to random coincidence rate was 20 to 1. Figure 3, curve C, shows the resulting gamma-ray spectrum in coincidence with low-energy (100–500 kev) betas; the energy of the photopeak is 1300 ± 40 kev. The channel gate was then set on the 1300-kev gamma-ray photopeak and the spectrum of betas in coincidence with it determined with an anthracene crystal covered with 2 mils of Al. The result is shown in Fig. 3, curve B, together with the singles beta-ray spectrum from In^{114m} (curve A). A Fermi plot of the coincidence data yields a beta-ray end point of 674 ± 40 kev. Johns[18] has independently assigned the 1300-kev gamma ray to Sn^{114}.

The branching ratio for the 1300-kev transition was determined by comparing, in the same geometry, the coincidence rate of beta—1300 kev gamma rays for In^{114m} with the positron—1270 kev gamma-ray coincidence rate of Na^{22}. The experimental result is 0.09%, in agreement with the intensity of the 1300-kev gamma ray in the singles spectrum.

As a further check, beta-gamma coincidences were investigated in a 72-sec In^{114} source. The 1300-kev gamma ray was again found to be in coincidence with 100–500 kev betas.

The log ft value calculated for the beta transition is 5.73, indicating an allowed transition. It is consistent with a 2^+ assignment for the 1300-kev level in Sn^{114}, in agreement with the systematics of even-even nuclei.[19] The energy of this level is consistent with the energies of first excited states of nuclei with $Z=50$.

IV. K-CAPTURE BRANCH OF 72-SECOND In^{114}

The 72-sec In^{114} activity was produced by (p,n) and $(d,2n)$ reactions on 83% enriched Cd^{114} in oxide form using the Brookhaven cyclotron which furnished 20-Mev deuterons. A 72-sec cadmium x-ray activity was found and its intensity per beta was measured. The source was placed between an anthracene crystal and a Lucite-covered $2\times15\times15$ mm NaI(Tl) crystal and the pulse distribution from the latter was observed with a 20-channel pulse-height analyzer. Figure 4 shows the resulting decay curve for the x-rays and for the beta rays of energy 1 to 2 Mev. The contribution of the 70-sec F^{17} positrons from bombardment of the oxygen in the target was found to be negligible by the use of a PbO_2 target. Using the same arrangement the corresponding rates for 49-day In^{114m} were obtained for calibration. Dividing the two ratios, we have

$$\frac{(K/\beta)_{72\text{-sec}}}{(K/\beta)_{49\text{-day}}}=\frac{f}{f+0.41},$$

where f is the 72-sec K-capture branching ratio and the value of 0.41 represents the 37.6% K x-ray per disintegration of 49-day In^{114m} due to the internal conversion of the 190-kev transition and the 3.5% K

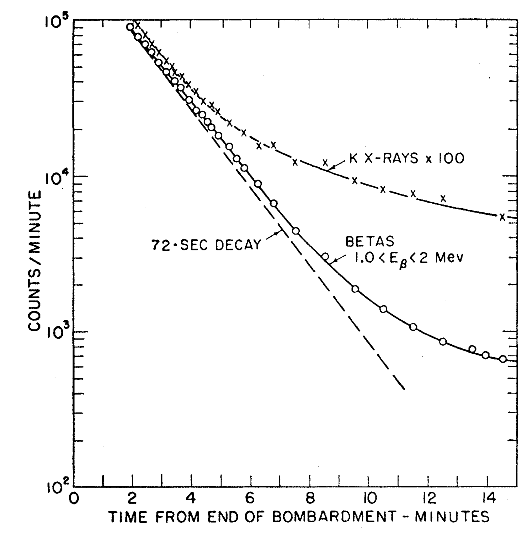

FIG. 4. Decay of cyclotron-irradiated target, indicating presence of 72-second activity.

[19] G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).

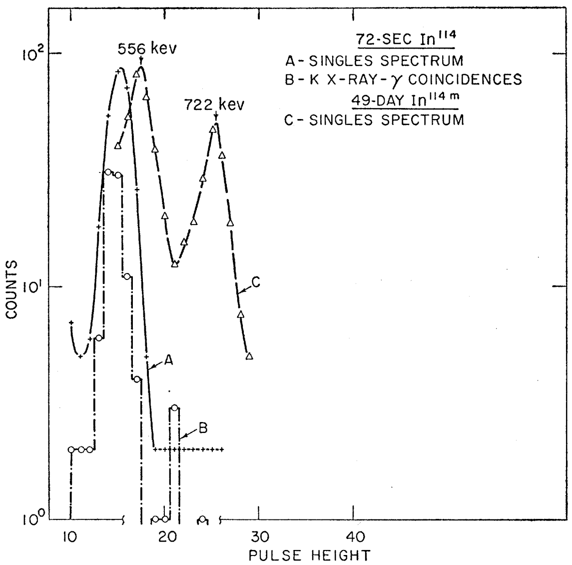

FIG. 5. Twenty-channel analyzer data, indicating lack of 772-kev gamma rays in 72-second activity.

branch to the 1278-kev state. The result for the 72-sec K-capture branch is $f=1.9\%$.

To determine if the 1278-kev level in Cd^{114} is fed by this K-capture branch, the anthracene crystal was replaced by a $1\frac{1}{2}\times 1$ in. NaI(Tl) crystal and K x-ray, gamma coincidences were investigated in a conventional fast ($\tau=10^{-7}$ sec)—slow coincidence spectrometer. The pulses from the $1\frac{1}{2}\times 1$ in. crystal which were in coincidence with K x-rays were displayed on a 20-channel analyzer. The resulting gamma spectrum is shown in Fig. 5. No discernible 722-kev or 556-kev gamma ray is seen. The 511-kev gamma-ray peak is not due to In^{114}. Though it does have a short-lived component with a half-life of approximately 70 sec, its intensity is too high to be accounted for by In^{114} positrons and is believed to be from F^{17}.

In order to obtain an upper limit for the K x-ray—722 kev branch, one can assume that all of the counts in the 722-kev region were real coincidences. Comparing the K x-ray—722 kev coincidences per K x-ray for the 72-sec and 49-day In^{114} gives

$$\frac{[(K-722)/K]_{72\text{-sec}}}{[(K-722)/K]_{49\text{-day}}}=\frac{k}{k_m}\frac{g_m}{g},$$

where k_m and k are the K-capture branching ratios to

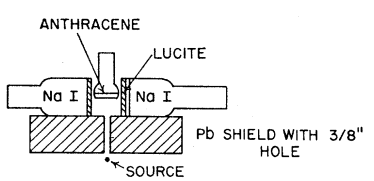

FIG. 6. Three-crystal spectrometer for measuring positron spectra.

the 1278-kev level and g_m and g are the K x-ray per disintegration ratios for the 49-day and 72-sec In^{114} states. The experimental result is $k\leq 0.02\ k_m$. Thus the 1278-kev level must be fed from the isomeric state, In^{114m}, which is known to have a spin of 5^+.[10] This result is consistent with the spin assignment of 4^+ to the 1278-kev level by the polarization-directional correlation data of Brazos and Steffen[7,9] for the 556–722 kev cascade.

Further confirmation that the spin of the 1278-kev level is not 2^+ is furnished by Johns[18] who, upon reexamining the external conversion spectrum from In^{114m}, does not find any evidence for a 1278-kev cross-over transition.

The experiment described above also shows that at least 80% of the 1.9% K-capture transition from In^{114} must take place to the ground state of Cd^{114} since the intensities of the 556- and 722-kev gamma rays are equal to within an experimental error of 10%.

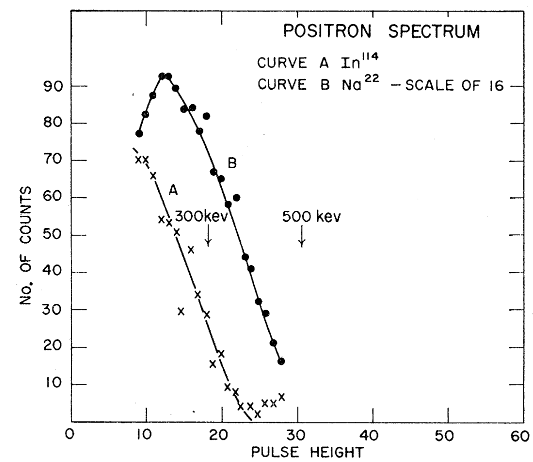

FIG. 7. Positronspectra obtained with the three-crystal spectrometer. Curve A—In^{114} spectrum, and curve B—Na^{22} comparison spectrum.

V. ENERGY DIFFERENCE BETWEEN In^{114} AND Cd^{114} GROUND STATES

The energy difference between the ground states of In^{114} and Cd^{114} was first measured by McGinnis[20] as 2.07 ± 0.2 Mev from the $Cd^{114}(p,n)In^{114m}$ excitation curve. Since he did not measure cross sections for proton energies less than 3.1 Mev and his extrapolation to threshold did not take into account the large centrifugal barrier corresponding to a spin change of 5 units, the threshold value he obtains is subject to considerable doubt. Johns[6] observed a positron branch in In^{114m} of $\sim 0.004\%$ and estimated the positron end point to be 1.2 ± 0.2 Mev by absorption.

We have measured the positron end point by a three-crystal coincidence technique; the experimental arrangement is shown in Fig. 6. Pulses from the side

[20] C. L. McGinnis, Phys. Rev. 81, 734 (1951).

crystals, 3 in.×3 in. NaI(Tl), were selected by single-channel analyzers which accepted quanta of 500±25 kev. The pulses from the center anthracene crystal were displayed on a 20-channel analyzer whenever a triple coincidence ($\tau = 10^{-7}$ sec) occurred. Energy calibration of the anthracene counter was obtained using the 976- and 482-kev internal conversion electrons from a Bi^{207} source. The background counting rate was measured to be less than 5% of the total by covering the source with Lucite; the accidental coincidence rate was negligible as determined by placing delays in the various channels. The resulting positron spectrum is shown in Fig. 7. Also shown on the same figure is the positron spectrum obtained using a Na^{22} source which has a known endpoint of 540±5 kev. Fermi plots of the positron spectra from Na^{22} and In^{114} give end points of

$$E_{\beta+}(Na^{22}) = 560 \pm 15 \text{ kev},$$
$$E_{\beta+}(In^{114}) = 400 \pm 25 \text{ kev}.$$

The positron branching ratio was obtained by comparing the number of triple coincidences per beta in the anthracene crystal for In^{114} with Na^{22} in the same geometry. The result is 0.0035±0.001% in agreement with the value obtained by Johns.[6]

It is possible to calculate the energy difference from the ratio of K capture to positron transitions between ground states of In^{114} and Cd^{114}. Using the value of 1.9% for the K-capture branching ratio and 0.0035% for the positron branching ratio, the ratio of K capture to β^+ emission is then 5.4×10^2. Using the curves of Feenberg and Trigg,[21] one obtains a value of ~1400 kev for the energy available for K capture from the ground state of In^{114}, in agreement with the value obtained from the positron spectrum. This energy difference indicates that it is energetically impossible to excite any level in Cd^{114} with an energy greater than 1650 kev in the decay of In^{114m}. This is an independent indication that the previously reported level at 1856 kev in Cd^{114} is not excited in the decay of indium.

The log ft value of the K-capture transition from In^{114} is 4.2 and from In^{114m} is 7.5. The latter value is high for an allowed transition and indicates that it is not a pure one-particle transition.

VI. CONCLUSIONS

The decay scheme which agrees with these results is shown in Fig. 8. The evidence that the spin of the

[21] E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).

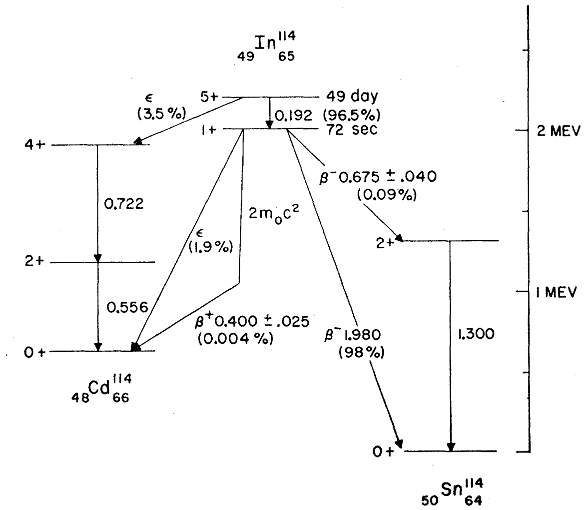

FIG. 8. Decay scheme for In^{114} based on most recent data.

1278-kev level in Cd^{114} is 4^+ is the following: (1) Angular correlation[1–4] measurements on the 556–722 kev transition indicate that the spin of the 1278-kev state is either 2^+ or 4^+; (2) angular correlation with polarization measurements[9] on this transition strongly favor a 4^+ assignment; (3) the lack of a crossover transition from this level,[18] and (4) the fact that it is fed from the 5^+ state of In^{114m}. The energy difference of 1420±25 kev between the ground states of In^{114} and Cd^{114} has been determined by a measurement of the positron spectrum endpoint. The measurements of the K-capture and positron branching ratios between these two ground states are consistent with such an energy difference. The existence of a 1300-kev level in Sn^{114} fed from In^{114} is established from β-γ coincidence experiments on both 49-day and 72-sec sources. A spin assignment of 2^+ for this state is consistent with the allowed ft value of the transition and the systematics of even-even nuclei.[19]

ACKNOWLEDGMENTS

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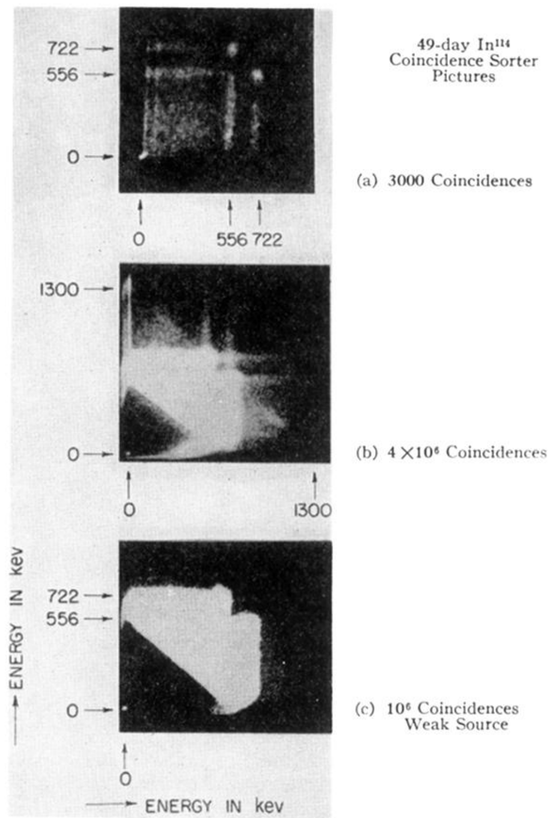

FIG. 1. In^{114m} gamma-gamma coincidences pulse photographed with the coincidence sorter (see text).