The Decay Scheme of Ag^{108}†

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The radiations of 2.3-minute Ag^{108} have been examined with scintillation and proportional counter spectrometers and with a coincidence spectrometer. The transitions and their abundances were found to be as follows: β^- 1.77 Mev (97.3 percent); β^- 1.15 Mev accompanied by γ of energy 0.62 Mev (0.8 percent); β^+ (0.14 percent); and K capture to ground and excited states of Pd^{108} (1.6 percent). Gamma rays of energies 0.60 Mev and 0.43 Mev were found to accompany K capture; the abundances of the different capture transitions were measured. Spin and parity assignments were made.

WITH the exception of one study made with coincidence circuitry and counter spectrometry,[1] there has been little reported concerning the radiations of Ag^{108}. Because of the general interest in electron-capture decaying nuclides and because of recent improvements in the techniques of coincidence spectrometry, it was considered reasonable to re-examine the radiations of Ag^{108}.

The Ag^{108} activity was made by the activation in the Brookhaven nuclear reactor of small samples of metallic silver, less than 0.1 mg per cm^2 in thickness, deposited on cellophane by evaporation. For most experiments an irradiation time of two minutes was chosen to give a favorable ratio for the yield of the 2.3-minute Ag^{108} to the yields of 39-second Ag^{109m}, 24-second Ag^{110}, and 44-second Ag^{107m}.[2]

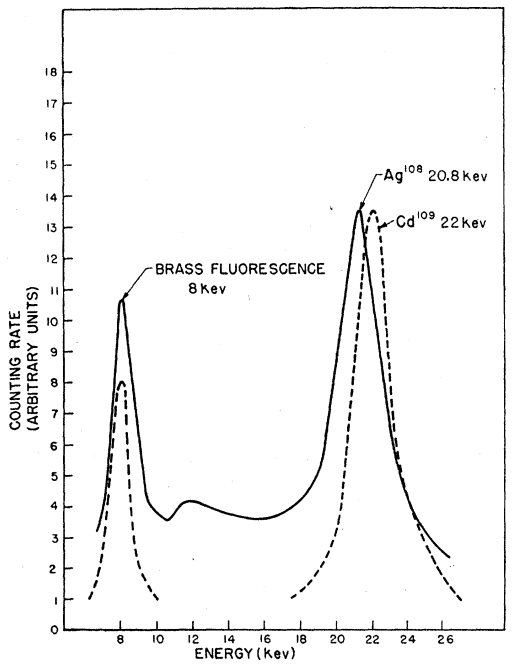

FIG. 1. Spectral distribution of x-radiations from Ag^{108} and Cd^{109}. The peaks at 8 kev represent fluorescence radiation from the brass counter wall.

X-RAY AND BETA-RAY MEASUREMENTS

The x-rays were detected by a brass-walled proportional counter, 12 inches long and 4 inches in diameter; the gas filling consisted of 95 percent argon and 5 percent methane at a pressure of 2 atmospheres. Beryllium absorber, 937 mg per cm^2, was used to prevent β radiations from entering the counter. Amplified counter pulses were examined with a gray-wedge spectrometer[3] and with a single channel analyzer. In Fig. 1 are shown the gray-wedge spectrum of Ag^{108} and, for comparison, that of Cd^{109}, a source of Ag K x-rays. It is seen that the energy of the Ag^{108} radiation, about 4 percent less than that of the Ag K x-ray, corresponds to that of Pd K x-radiation, which would accompany K capture. The x-ray intensity was found to decay with the characteristic period of Ag^{108}. No change in the pulse-height distribution could be observed when a source 10 times as thick as the standard ones was used. It was thus established that the contribution of Ag fluorescence from the beta activity to the x-ray intensity from the thin samples could not have been more than 5 percent. It was assumed that a negligible contribution to the x-ray intensity was made by internal conversion of gamma rays. This assumption will be considered in a following section. The absolute x-ray disintegration rate was determined by measurement of the x-ray pulse-height distribution with the single channel analyzer. Points on the pulse-height distribution curve were corrected for decay occurring during the measurements. From the area under the curve, after the bremsstrahlung contribution had been subtracted, the absolute K electron-capture rate was derived by a method previously described.[4] For Pd K x-rays the transmission of the beryllium absorber and window was 80 percent and the absorption of the counter gas was 21 percent; the fluorescence yield of Pd was taken to be 0.81.[5] Immediately after the x-ray pulse-height distri-

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[1] M. Goodrich and E. C. Campbell, Phys. Rev. **85**, 742 (1952).

[2] *Nuclear Data*, National Bureau of Standards Circular 499 (U. S. Government Printing Office, Washington, D. C., 1950).

[3] Bernstein, Chase, and Schardt, Rev. Sci. Instr. **24**, 437 (1953).

[4] Friedlander, Perlman, Alburger, and Sunyar, Phys. Rev. **80**, 30 (1950).

[5] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).

bution had been scanned, the beryllium absorber was removed and the total rate in the proportional counter was measured. This rate, corrected for decay, for backscattering, and for window absorption in accordance with the beta energy determination to be described, was taken to be the product of the beta disintegration rate and the geometry; and it was directly compared with the K-capture rate obtained in the same geometry. The ratio of the K-capture rate to the beta rate was calculated to be 0.016 ± 0.001. The ratio of L capture to K capture is expected to be 0.11.[6]

The beta spectrum was obtained by gray-wedge analysis of the pulse-height distribution from a scintillation counter; the anthracene crystal used was 1.5 cm in thickness. Energy and resolution calibrations were made with the Cs^{137} internal conversion electron line (624 kev). The thickness of the Ag^{108} source was less than 0.1 mg per cm^2; there was no backing other than the 3.5 mg per cm^2 Cellophane. Figure 2 shows the Fermi plot derived from the observed pulse-height distribution corrected for the variation in resolution with pulse height.[7] Within the accuracy of the measurement the spectrum appears to be simple with a maximum energy of 1.77 ± 0.06 Mev. The deviation from the line of points below 0.8 Mev is attributed to scattering of betas out of the crystal.

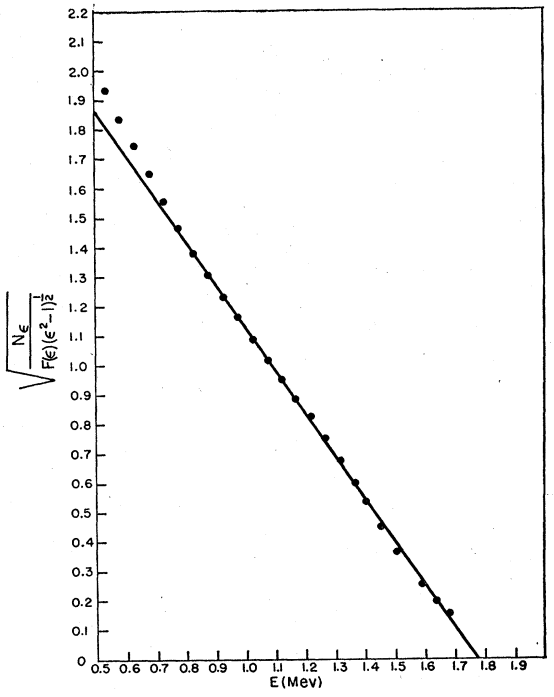

FIG. 2. Fermi plot for the β radiations of Ag^{108}.

GAMMA-RAY MEASUREMENTS

The gamma rays from Ag^{108} were examined with the gray-wedge spectrometer and scintillation detector. A DuMont Type K1186 photomultiplier tube was used together with a NaI(Tl) crystal 3 cm in diameter and 2 cm high; this combination had a resolution of 8 percent for the 662-kev gamma ray of Cs^{137}. The spectrum, as displayed on the instrument, is shown in Fig. 3. Beryllium absorbers served to screen beta rays from the crystal. Gamma rays of energies 616 ± 20 kev, 510 ± 20 kev, and 435 ± 20 kev may be identified. The indication of a peak in the vicinity of 200 kev is probably attributable to backscattering; and the pulse continuum, which increases in intensity at low energies, is produced by bremsstrahlung from beta rays. The character of the spectrum did not change as the sources decayed.

COINCIDENCE MEASUREMENTS

Coincidence measurements were made with pairs of scintillation detectors appropriate to the various radiations and a coincidence gray-wedge spectrometer.[8] With this instrument it is possible to display all events in one detector which occur in time coincidence with events of selected amplitude in a second detector. The effective coincidence time is 10^{-7} sec. Accidental coincidence effects are evaluated by the introduction of a delay line, 5×10^{-7} sec, between the selecting detector and the coincidence circuit. Data were recorded with a Polaroid Land Camera. No information could be obtained earlier than 2 minutes after irradiation because of high accidental rates produced by the 24-second Ag^{110} activity.

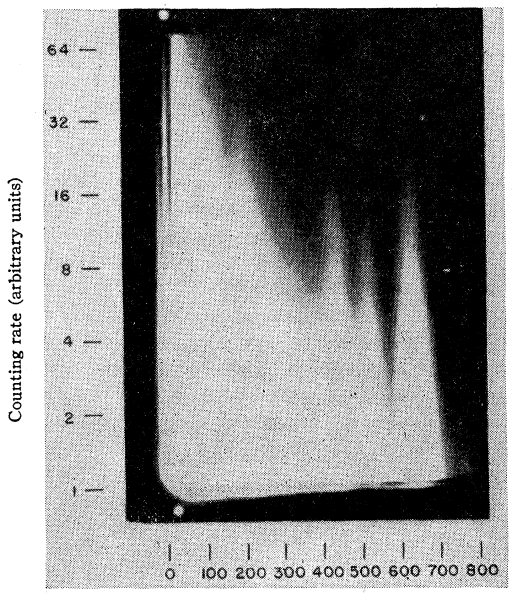

FIG. 3. Gamma-ray spectrum of Ag^{108}.

[6] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).
[7] G. E. Owen and H. Primakoff, Phys. Rev. **74**, 1406 (1948).
[8] Chase, Bernstein, and Schardt, Phys. Rev. **90**, 353 (1953); Chase, Bernstein, and Schardt, Brookhaven National Laboratory Report BNL-237 (T37) (unpublished).

Gamma-gamma coincidence measurements were made with two 3-cm×2-cm NaI(Tl) detectors. Sufficient beryllium absorber was interposed between sample and detectors to screen out β rays. In coincidence with the 435-kev gamma ray there was observed only a gamma ray of energy 602 kev, approximately three percent lower in energy than the 616-kev line which appeared in the "singles" gamma spectrum. That the 510-kev gamma line represents a ground-state positron transition is shown by the facts that it was in coincidence only with a 510-kev gamma ray and that these coincidences disappeared when the counter-source-counter geometry was other than 180 degrees. The presence of the annihilation line as an internal reference made possible the definite detection of the small energy shift from the 616-kev line in the "singles" spectrum to the 602-kev line in the coincidence spectrum.

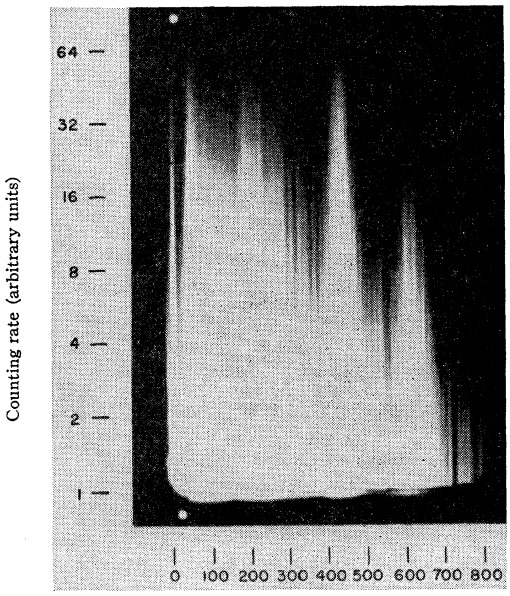

FIG. 4. Spectrum of gamma rays in coincidence with x-rays from Ag^{108}.

For the x-ray–gamma-ray coincidence measurements one of the thick NaI(Tl) detectors was replaced by a 2 mm deep NaI(Tl) crystal in order to provide a more favorable x to gamma sensitivity ratio. Beryllium beta-ray absorbers were used. Figure 4 shows the spectrum of gamma rays in coincidence with the x-rays; the lines at 435 kev and at 602 kev appear. The inverse experiments confirmed the existence of these x-γ coincidences. It is seen in the x-γ coincidence photograph that the 602-kev photopeak is much less intense than the 435-kev peak, whereas in the "singles" photograph the 616-kev line is the more intense. The 616-kev "singles" line is therefore composed of two lines not resolved from each other, only one of which, the 602-kev, is in coincidence with the x-rays. Examination of the "singles" spectrum in the region of 1 Mev gave no evidence for the possible cross-over transition. From the areas under the 435-kev and 602-kev photopeaks, the relative intensities of the two gamma transitions were calculated to be 1.00 and 0.79±0.12, respectively; correction was made for variation of photopeak efficiency in the scintillator[9] with energy. These intensities are interpreted to mean that of the capture events which produce the excited states of Pd^{108}, 79 percent give rise to the 602, 435 cascade and 21 percent give rise only to the 435-kev lower state. The area of the 435-kev photopeak was derived from the coincidence spectrum after subtraction of the Compton contribution from the 602-kev gamma ray and of a small bremsstrahlung contribution.

All coincidence spectra observed with the 180-degree geometry show a broad peak in the vicinity of 200 kev, which disappeared when the displaying detector was so positioned as not to intercept backscattered radiation from the other detector. This 200-kev peak may be what was previously reported to be a 190-kev gamma ray.[1]

The anthracene detector described above was used in combination with one of the thick NaI(Tl) detectors for the β-γ coincidence measurements. A beryllium beta-ray absorber was placed between the gamma detector and the source; and the discriminator bias on the selecting anthracene detector was set so as to cut off the response to the x-rays. The 616-kev line and annihilation radiation were observed. The 616-kev gamma ray is therefore associated with beta decay; however, the 510-kev line arises from the detection in the NaI crystal of annihilation quanta of the positrons whose kinetic energy is expended in the anthracene detector.

In order to determine the fraction of K electron-capture events which lead to excited states of Pd^{108}, the measured ratio of x-γ coincidences to x-rays in Ag^{108} was compared with the ratio measured in the same geometry for 65-day Sr^{85}. The gamma-ray detector was operated so as to be sensitive to essentially all of the pulse-height distribution of the gamma rays involved. The Sr^{85} activity was chosen as a reference because all capture transitions in it have been found to be followed by one gamma ray of energy 513 kev,[10–12] and because this gamma-ray energy is not greatly different from those associated with electron capture in Ag^{108}. Corrections for the variation with energy of the NaI efficiency are therefore relatively small. Corrections for x-ray fluorescence yield, sensitivity, and absorption are unnecessary because they cancel in each ratio. The

[9] P. R. Bell (unpublished data).
[10] M. Ter-Pogossian and F. T. Porter, Phys. Rev. **81**, 1057 (1951).
[11] Sunyar, Mihelich, Scharff-Goldhaber, Goldhaber, Wall, and Deutsch, Phys. Rev. **86**, 1023 (1952).
[12] W. S. Emmerich and J. D. Kurbatov, Phys. Rev. **85**, 149 (1952).

activity was produced by the irradiation in the Brookhaven nuclear reactor of $SrSO_4$ enriched[13] in Sr^{84}. No gamma radiation of energy other than 513 kev was observed at the time when the Sr^{85} measurements were made. The contribution of radiations other than x-rays to the count-rate in the thin NaI detector was determined, for the Sr^{85}, by the interposition of a thin copper absorber between source and detector. In the case of the Ag^{108} the fraction of the measured count-rate attributable to Pd K x-rays in a pulse-height interval which included the Pd K x-rays was evaluated from a gray-wedge photograph; the absorber method could not be used because it would remove some bremsstrahlung in addition to x-rays. The fraction of Pd x-rays was 0.70 ± 0.05. A difficulty associated with the use of Sr^{85} in this experiment is that the 513-kev gamma ray has a half-life of $(0.9\pm0.2)\times10^{-6}$ sec.[11] This means that there is recorded only a fraction of the coincidences, the fraction being a function of the ratio of the coincidence resolution time to the half-life. Because of the uncertainty in the lifetime, measurements on the Sr^{85} were made at two resolution times, 0.12×10^{-6} sec and $(1.5\pm0.2)\times10^{-6}$ sec. The two values for the ratio (x-γ coinc)/x calculated for infinite resolution time agreed within 10 percent; and their average, 0.066 ± 0.006, was used. In the case of the Ag^{108} the ratio (x-γ coinc)/x was 0.0183 ± 0.0015. The individual x, γ, and coincidence count-rates decayed with the characteristic 2.3 to 2.4-minute half-life. The value, corrected for total NaI gamma efficiencies,[9] of the ratio

$$\frac{(\text{x-}\gamma)\ \text{coinc/x Ag}^{108}}{(\text{x-}\gamma)\ \text{coinc/x Sr}^{85}},$$

was calculated to be 0.155 ± 0.021. In the Ag^{108}, therefore, 15 percent of the capture transitions are to excited states of Pd^{108}, and the remaining 85 percent are to the ground state.

From the photopeak areas in the "singles" gamma-ray spectrum, the abundances of the annihilation radiation and of the sum of the 602- and 616-kev transitions were determined relative to the abundance of the 435-kev transition. The appropriate corrections for efficiency and backgrounds were made. Because the abundance of the 602-kev gamma relative to that of the 435-kev gamma was already determined and because the abundance of the 435-kev gamma transition in the scheme was known, the abundances of all radiations of Ag^{108} could be evaluated. The final results are given in the decay scheme shown in Fig. 5. It may be noted that the positron abundance is less than the upper limit established by Barber.[14]

[13] Supplied by the Y-12 plant, Carbide & Carbon Corporation, through the Isotopes Division, U. S. Atomic Energy Commission, Oak Ridge, Tennessee.

[14] W. C. Barber, Phys. Rev. **72**, 1156 (1947).

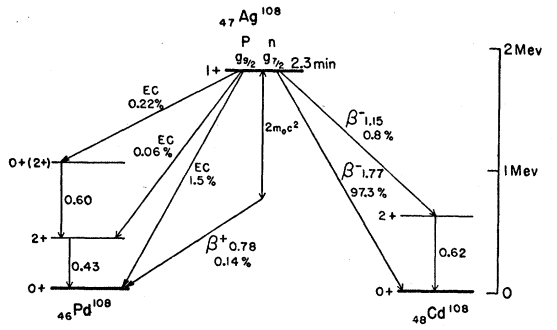

FIG. 5. Decay scheme of Ag^{108}.

DISCUSSION

The assignments, based on the experimental results, of 435 kev and 616 kev as the energies for the first excited states of Pd^{108} and Cd^{108}, respectively, are consistent with expectations based on the observed excited states of neighboring even-even nuclei.[15] The value of logft for the transition to the ground state of Cd^{108} is calculated to be 4.6; for the upper state transition logft is 6.0. The two β^- transitions thus appear to be allowed; and if the Cd^{108} ground and excited state spins and parities are taken to be 0+ and 2+, respectively,[15] the spin-parity assignment for Ag^{108} is restricted to 1+. This assignment is in agreement with the expectation of Nordheim.[16] Because the Pd^{108} ground-state assignment is 0+, the positron and electron-capture ground-state transitions must be allowed; and the K/β^+ ratio calculated from experiment may be used to evaluate the energy of these transitions.[17] The positron energy is thus determined to be 0.78 ± 0.05 Mev. For the transition to the ground state and first and second excited states of Pd^{108} the values of logft are 4.8, 6.0, and 4.8, respectively. The first excited state of Pd^{108} is expected[15] to have spin 2 and even parity, so that the transition to this state should be allowed. If, from its ft value, the transition to the second excited state is taken to be allowed, the choice for spin-parity assignment for this state is limited to either 0+ or 2+. Although 0+ would appear to be the preferred assignment because of the absence of the cross-over transition, the value 2+ cannot be excluded on this basis.[18]

The assignment of spins and parities to the Cd^{108} and Pd^{108} levels makes possible the evaluation of the internal conversion coefficients for the gamma-ray transitions.[19]

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

[16] L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).

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

[18] J. J. Kraushaar and M. Goldhaber, Phys. Rev. **89**, 1081 (1953); also A. de-Shalit and M. Goldhaber, Phys. Rev. **92**, 1211 (1953).

[19] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 906 (1951).

X-rays from conversion are thus calculated to contribute 1/300 of the x-ray intensity from capture.

From the energies of the ground-state transitions, the mass difference Cd^{108} minus Pd^{108} is estimated to be 30±80 kev. This estimate is believed to be more accurate than that obtainable from the mass measurements now available.[20]

[20] R. E. Halsted, Phys. Rev. **88**, 666 (1952).

ACKNOWLEDGMENT

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Nuclear Quadrupole Resonance of the Stable Gallium Isotopes*

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Eight absorption lines have been observed in a sample of polycrystalline $GaCl_3$. Of these, six are ascribed to the Cl isotopes. The remaining two, which are due to the stable gallium isotopes $Ga^{69,71}$, allow us to determine the ratio of their quadrupole moments. $Q_{Ga^{69}}/Q_{Ga^{71}}=1.5867\pm0.0004$, in agreement with earlier atomic beam measurements.

NUCLEAR quadrupole resonance[1,2] in the spectral region from 9.3 to 36.8 Mc/sec has been investigated in a sample of about 10 g $GaCl_3$. A recording superregenerative spectrometer was used, employing 30-cps Zeeman modulation.[3] All together eight absorption lines were observed whose frequencies are given in Table I. As the spins of the gallium isotopes $Ga^{69,71}$ as well as the chlorine isotopes $Cl^{35,37}$ are $\frac{3}{2}$, only the one transition $m=\pm\frac{1}{2}\leftrightarrow\pm\frac{3}{2}$ is expected for each of the four nuclear species. With the help of the known ratio[4,5] of the chlorine quadrupole moments, $Q_{Cl^{35}}/Q_{Cl^{37}}=1.26878\pm0.00015$, six of the observed lines could be grouped into three pairs corresponding to three different lattice sites for the Cl atoms. The two remaining lines, whose frequency ratio is found to be the same as the ratio of the gallium quadrupole moments, $Q_{Ga^{69}}/Q_{Ga^{71}}=1.5868\pm0.0004$, known from atomic beam experiments,[6] were assigned to the gallium isotopes. These lines were 5 kc/sec wide as compared to a width of 2 kc/sec for the Cl lines; also, in contrast to the Cl lines, they showed signs of partial saturation for the same rf power level. Both aspects can be understood in view of the fact that the magnetic moments of the gallium isotopes are two to three times larger than those of the Cl isotopes.

TABLE I. Absorption frequencies in $GaCl_3$.

Nucleus	Lattice site	T °C	Frequency kc/sec	Frequency ratio of isotopic pairs
Ga^{69}		31.8±0.1	29 060±2	1.5867±0.0002
Ga^{71}			18 315±1	
Cl^{35}	1*a*	33.2±0.6	20 225±3	1.2692±0.0004
Cl^{37}			15 935±2	
Cl^{35}	1*b*		19 084±3	1.2689±0.0004
Cl^{37}			15 040±2	
Cl^{35}	2		14 667±2	1.2686±0.0003
Cl^{37}			11 562±1	

The ratio of the observed gallium frequencies should agree with that of the quadrupole moments at least to the same degree as given by the experimental error of 0.02 percent, since, for molecules which are made up from atoms that heavy, the effects of small isotopic differences in zero-point vibration on the field gradient are proven to be negligible.[5] Therefore, we obtain $Q_{Ga^{69}}/Q_{Ga^{71}}=1.5867\pm0.0004$, in close agreement with the atomic beam value.

Investigations of other gallium compounds are under way.

* Work supported by the U. S. Office of Ordnance Research.
[1] H. G. Dehmelt and H. Krüger, Z. Physik **129**, 401 (1951); H. G. Dehmelt, Am. J. Physics (to be published).
[2] R. V. Pound, Phys. Rev. **79**, 685 (1950).
[3] H. G. Dehmelt, Phys. Rev. **91**, 313 (1953).
[4] R. Livingston, Phys. Rev. **82**, 289 (1951).
[5] Wang, Townes, Schawlow, and Holden, Phys. Rev. **86**, 809 (1952).
[6] G. E. Becker and P. Kusch, Phys. Rev. **73**, 584 (1948).

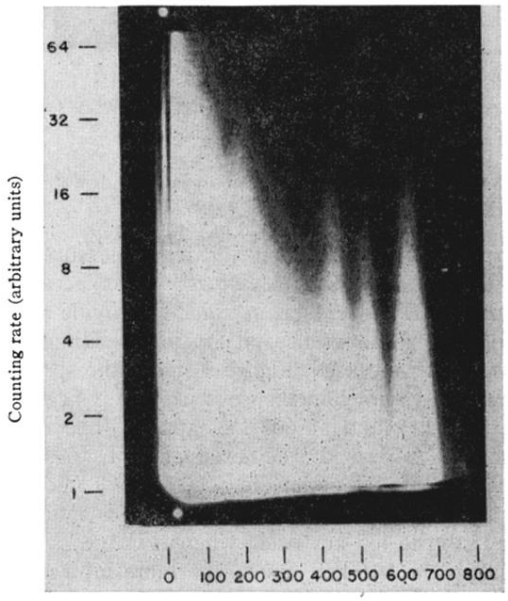

FIG. 3. Gamma-ray spectrum of Ag^{108}.

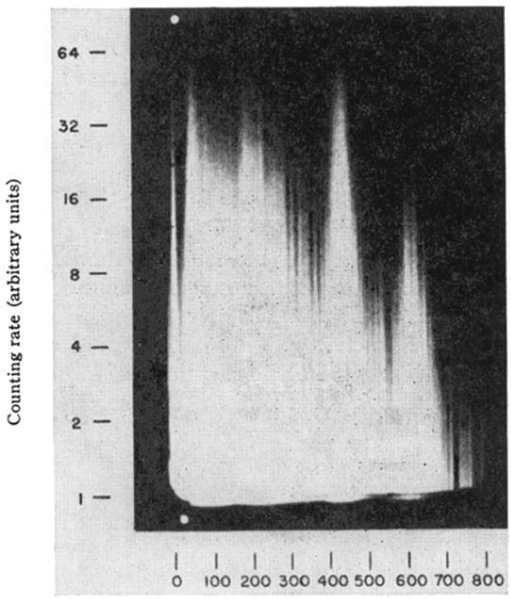

FIG. 4. Spectrum of gamma rays in coincidence with x-rays from Ag^{108}.