Decay of Ag^{108m}†

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The decay of the long-lived isomer (>5 yr) of Ag^{108} has been investigated with scintillation and proportional-counter spectrometers employing coincidence techniques. Conversion-electron measurements with a high-transmission "orange"-type spectrometer and a high-resolution $\pi\sqrt{2}$ double-focusing spectrometer together with gamma-gamma angular-correlation measurements served to establish the multipolarities of most of the observed transitions. Ag^{108m} has been found to decay 8.5% of the time via a two-step isomeric cascade consisting of a 30.4-keV $M4$ transition followed by a 79.4-keV $E1$ transition. The remaining 91.5% of the decays proceed by an electron-capture transition to a 1770-keV level in Pd^{108}. This level then decays to the ground state of Pd^{108} via a 722.7±1.0 keV, 613.9±0.6 keV, 433.6±0.5 keV triple gamma cascade. Two excited states at 79.4±0.5 and 109.8±0.5 keV are established in Ag^{108}, with proposed spins and parities of 2- and 6+, respectively. The half-life of the 79-keV level has been shown to be $\leq 10^{-9}$ sec. The spins and parities of the 1770- and 1048-keV levels in Pd^{108} are assigned as 6+ (possibly 5+) and 4+, respectively.

I. INTRODUCTION

THE existence of a long-lived isomer of Ag^{108} was first reported by Wahlgren and Meinke.[1] These investigators reported that this isomer decayed with a half-life of 5 or more years. The principal mode of decay was by electron capture to a state at 1772 keV in Pd^{108} which deexcited via a triple cascade of γ rays to the ground state. An 81-keV γ ray was tentatively assigned to be in cascade with an unobserved isomeric transition in Ag^{108}. These early measurements were confirmed and extended by several workers.[2–4] In particular, γ-γ angular correlation measurements were carried out[2–4] on all γ-ray pairs in the electron capture branch, and a 30-keV isomeric transition in coincidence with the 81-keV γ ray was established.[4] We present here the details of our earlier measurements as well as the results of recent magnetic-spectrometer studies of the conversion-electron spectra in both the electron-capture and the isomeric transition branches of the Ag^{108m} decay. These studies establish the multipolarities of most of the known transitions.

II. APPARATUS, SOURCES, AND EXPERIMENTAL PROCEDURES

The early low-energy electron measurements were made with a gas-flow proportional counter. With this device it was possible to work with the very low-activity-electron sources which were available. The counter had an aluminum body with an inside diameter of 2 in. and a length of 7 in. The center wire was 0.002 in. diam tungsten. A 1-in.-diam, 0.010-in.-thick beryllium window was sealed into the side wall of the counter by means of "O" rings and was easily removable for insertion of the source. In practice, the source was glued to the inner surface of the window, which made up part of the cathode. A commercially available mixture of 90% argon and 10% methane gas was flowed through the counter at an absolute pressure of ~125 cm of mercury.

Two magnetic β-ray spectrometers were used in conversion-electron studies when a mass-separated source of considerably higher activity was available. The six-gap "orange" spectrometer at Brookhaven[5] was used to measure the weaker conversion-electron lines which required the high transmission of this spectrometer. The electron detector of this instrument was an anthracene scintillation counter. For those measurements which required high resolution, a 50-cm radius, $\pi\sqrt{2}$, iron, double-focusing instrument[6] was used. A gas-flow proportional counter at a pressure of 25 mm of mercury of butene 2, with a 100 μg/cm² window of VYNS served as the electron detector in this instrument.

Two sources of Ag^{108m} were used for the conversion electron measurements. The initial measurements were carried out with very low specific activity sources originally prepared by activation of natural silver in the Brookhaven reactor in 1951. The old age of this source made it largely free of the contaminating 250-day Ag^{110m} activity. Electron sources for the proportional counter were prepared by displacement plating from $AgNO_3$ solutions of this activity onto thin copper foil. The resulting sources had a surface density of ≈40 μg/cm². The later magnetic-spectrometer measurements were carried out with sources of much higher specific activity. These sources were prepared by mass separation in the Brookhaven Chemistry Department 60 in. radius, 90° sector, electromagnetic isotope separator manufactured by High Voltage Engineering Corporation of a neutron-activated sample of silver enriched in mass 107.

Gamma-gamma coincidence measurements were performed with 3 in.×3 in. NaI(Tl) scintillators using a conventional fast-slow coincidence system ($2\tau \simeq 10^{-7}$

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] M. A. Wahlgren and W. W. Meinke, Phys. Rev. **118**, 181 (1960).

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[3] E. G. Funk, Jr., G. C. Martin, R. C. Pilger, Jr., and J. W. Mihelich, Bull. Am. Phys. Soc. **6**, 428 (1961).

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[6] G. T. Emery, W. R. Kane, M. McKeown, M. L. Perlman, and G. Scharff-Goldhaber, Phys. Rev. **129**, 2597 (1963).

sec). In addition, a γ-ray spectrum was obtained using a Li-drifted Ge detector of $\simeq 6$ cm^2 area and $\simeq 6$ mm depth.

III. PROPORTIONAL-COUNTER STUDIES AND ELECTRON-GAMMA COINCIDENCE MEASUREMENTS

The electron spectrum obtained with our proportional counter using a source of only 3 mμCi of Ag^{108m} is shown in curve (a) of Fig. 1. Prominent lines consist of *KLL* and *KLM* Auger electron groups, the *L* and *M* conversion electrons of a 30-keV transition and a *K* conversion line of a 79-keV transition. In addition, there is a broad peak which consists of *L* and *M* conversion electrons of the 79-keV transition as well as several sum lines. The prominent Cu *K* x ray is caused by fluorescence of the source backing. Curve (b) of Fig. 1 exhibits the proportional counter electron spectrum taken in coincidence with 79-keV photons detected by a 2 mm$\times 1\frac{1}{2}$ in. NaI(Tl) scintillation counter which viewed the source through the beryllium window. This

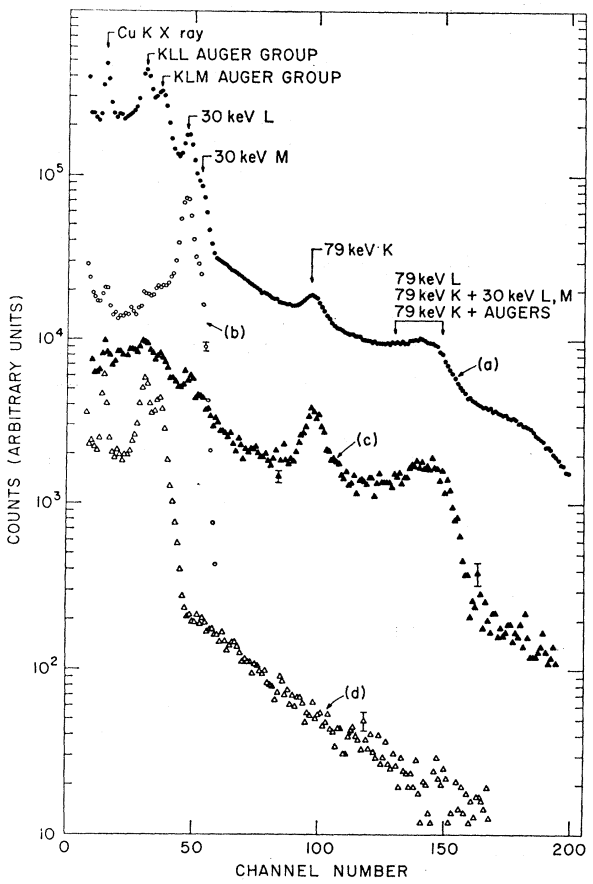

FIG. 1. Ag^{108m} electron spectra as measured with the argon-methane proportional counter. (a) Singles spectrum. (b) Spectrum recorded in coincidence with 79-keV γ rays. (c) Spectrum recorded in coincidence with *K* x rays. (d) Spectrum recorded in coincidence with 434-, 614-, and 723-keV γ rays.

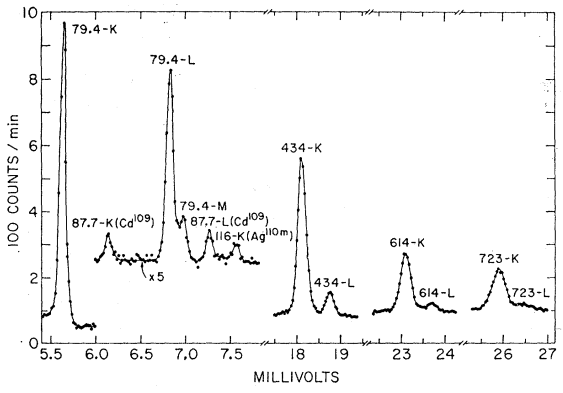

FIG. 2. Conversion-electron spectrum from an isotope-separated Ag^{108m} source measured with the "orange" spectrometer.

spectrum clearly demonstrates the coincidence of the 79-keV transition with the *L* and *M* conversion electrons from the transition of ≈ 30 keV. Curve (c), the proportional counter spectrum gated by coincident *K* x rays, shows only an enhancement of the 79 *K* line and its sum lines. Curve (d), the proportional counter spectrum gated by coincident high-energy (434, 614, 723 keV) gamma rays detected with a 3 in.$\times$3 in. NaI(Tl) scintillation counter, shows enhancement of the Auger electron groups arising from the *K* holes created by the preceding electron capture transitions.

IV. MAGNETIC-SPECTROMETER MEASUREMENTS

The magnetic spectrometer measurements were made using the mass-separated source which consisted of $\approx\frac{3}{4}$ μCi of Ag^{108m} deposited on 2.5×10^{-4} in. thick Al foil. The accelerating voltage in the mass-separator drives the ions to a depth equivalent to $\cong 8$ μg/cm^2 into the Al backing. Measurements were made using 3 gaps of the orange spectrometer, with a transmission of $\sim 4\%$ and a resolution of $\sim 1.25\%$. The parts of the spectrum which are of interest are shown in Fig. 2. The conversion lines from a small amount of Cd^{109} present as a contaminant in the source furnished a convenient check on the energy calibration of the orange spectrometer near the conversion lines of the 79-keV transition. In particular, the measured K-L_{2+3} separation of the 88-keV Ag^{109} isomeric transition allowed us to fix the K-L_1 separation of the 79-keV transition as 22.1 ± 0.4 keV, thereby establishing the transition as one which converts in silver. The conversion line intensities of the 79-, 434-, 614-, and 723-keV transitions as determined from these measurements are listed in Table I, normalized with respect to the 434-keV *K*-conversion line.

The *L* conversion lines of the 30-keV isomeric transition were measured with the 50 cm, $\pi\sqrt{2}$, double-focusing spectrometer operated at a resolution of $\approx 0.3\%$. This spectrum is shown in Fig. 3. The resolved *L* structure permits an unambiguous assignment of the multipolarity as *M*4 for this transition. The relative *L*-conversion

TABLE I. Transition energies and results of conversion-electron (c.e.) measurements for Ag^{108m}.

Transition (keV)	Relative gamma intensity	Conversion shell	Relative c.e. intensities	Conversion[a] coefficient	Theoretical conversion coefficients[b] $E1$	$M1$	$E2$
79.4±0.5	0.073±0.008	K	2.04 ±0.10	0.22 ±0.003	0.27	0.71	2.40
		L	0.25 ±0.02	0.028±0.003	0.034	0.091	0.81
		$M, N, \cdots$	0.061±0.013	0.007±0.002	...	...	...
433.6±0.5	1.00	K	1.00	...	...	...	7.8×10^{-3}
		$L, M, \cdots$	0.148±0.023	$(1.16\pm0.18)\times10^{-3}$	...	...	...
613.9±0.6	1.03 ±0.03	K	0.37 ±0.03	$(2.85\pm0.22)\times10^{-3}$	1.04	3.00	2.92
		$L, M, \cdots$	0.051±0.016	$(0.40\pm0.13)\times10^{-3}$	...	...	...
722.7±1.0	1.02 ±0.03	K	0.250±0.012	$(1.95\pm0.10)\times10^{-3}$	0.73×10^{-3}	2.05×10^{-3}	1.91×10^{-3}
		$L, M, \cdots$	0.046±0.008	$(0.36\pm0.06)\times10^{-3}$	...	...	...

Transition (keV)	Conversion ratio	Experimental value	Theoretical value $M4$	$E3$	$M3$
30.38±0.06	$L_I/(L_{II}+L_{III})$	0.20 ±0.05	0.155	0.009	0.373
	$\alpha_K/(1+\alpha_T)$	0.037±0.005			

[a] These conversion coefficients are based on the theoretical value of α_K for the 434-keV transition. The 434-, 614-, and 723-keV transition intensities are assumed to be equal.
[b] See Refs. 8 and 9.

coefficients as determined from these measurements are also listed in Table I.

V. GAMMA-RAY INTENSITY AND GAMMA-GAMMA COINCIDENCE MEASUREMENTS

Relative intensities of the 79-, 434-, 614-, and 723-keV gamma rays were derived from NaI(Tl) scintillation-counter spectra and are listed in Table I. The 434-keV γ intensity has been set equal to unity. Internal-conversion coefficients for all transitions have been determined by combining the results of the relative-electron-intensity measurements, and by assuming that the 434-keV $2+\rightarrow0+$ $E2$ transition[7] has the theoretical value[8,9] 7.8×10^{-3} for its K-shell internal conversion coefficient. The internal conversion coefficients arrived at in this manner are also shown in Table I.

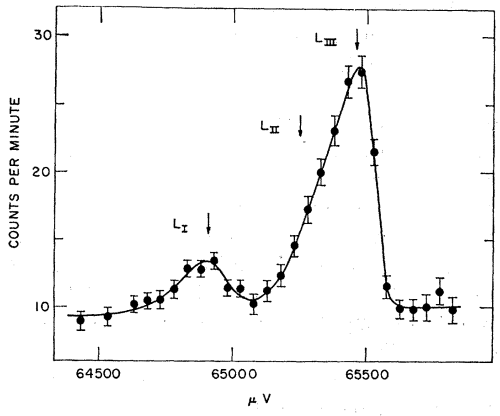

FIG. 3. L-conversion lines of the 30-keV isomeric transition in Ag^{108m} as measured with the 50-cm, $\pi\sqrt{2}$, double-focusing spectrometer.

[7] F. K. McGowan and P. H. Stelson, Phys. Rev. **99**, 127 (1955).
[8] L. A. Sliv and I. M. Band, Physics Department, University of Illinois, Urbana, Illinois, Reports Nos. 57 ICC K and 58 ICC L (unpublished).
[9] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

Once the total conversion of the 79-keV transition is known, the fraction of Ag^{108m} decays which proceed via the isomeric branch may be determined from the 79 to 434-keV γ-ray ratio. The value of this branch is (8.5±0.8)%.

The high-energy portion of the γ-ray spectrum was measured with a Li-drifted Ge detector. In order to improve the accuracy of our energy measurements, this spectrum was recorded simultaneously with the 411.795 ±0.009-keV γ ray[10] of Au^{198}, the 661.595±0.076-keV γ ray[10] of Cs^{137} and the 569.62±0.06- and 1063.44±0.09-keV γ rays[11,12] of Bi^{207}. The amplifier response was calibrated with a precision pulser. The gamma-ray energies derived from these measurements are those listed in Table I. A Ag^{108m} gamma spectrum measured with the Li-drifted Ge detector is shown in Fig. 4.

The following gamma-gamma coincidence measure-

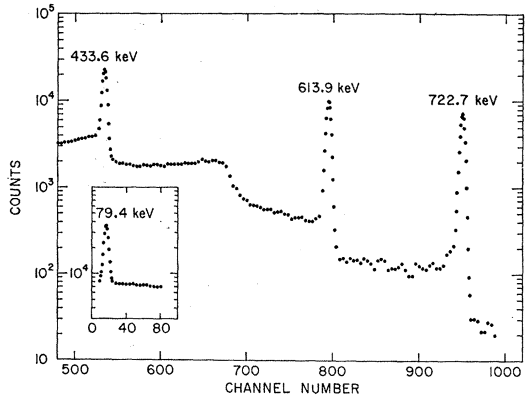

FIG. 4. Ag^{108m} γ-ray spectrum as measured with an ≈3 cc Li-drifted Ge detector.

[10] R. L. Graham, G. T. Ewan, and J. S. Geiger, Nucl. Instr. Methods **9**, 245 (1960).
[11] G. Bäckström, Arkiv Fysik **10**, 313 (1956).
[12] F. P. Brady, Nucl. Phys. **66**, 365 (1965).

TABLE II. Results γ-γ angular correlation measurements.

Gate	Correlation	Correlation coefficients A_2	A_4
724	724−614	0.099±0.004	0.008±0.005
	724−(614)−434	0.105±0.003	0.012±0.005
614	614−724	0.105±0.003	0.007±0.005
	614−434	0.100±0.003	0.013±0.004
(614+724)	724−614	0.102±0.004	0.010±0.007

ments were performed using NaI(Tl) scintillation detectors.

(a) In a calibrated geometry, the number of K x rays in coincidence with the 434-, 614-, and 723-keV γ rays was measured. The K-shell fluorescent yield[13] in palladium was taken as 0.79. The ratio of K electron capture to total electron capture was determined to be 0.84±0.04. This ratio, together with the computations of Brysk and Rose,[14] implies an energy difference between the isomeric state of Ag^{108} at 110 keV and the Pd^{108} excited state at 1770 keV of 130_{-50}^{+1200} keV. The lower limit of 80 keV for the energy release is the only meaningful interpretation to be placed on this number. A recent positron end-point determination[15] of 880±25 keV for the Ag^{108} ground state to Pd^{108} ground-state positron group leads to a value of 242 keV for the Ag^{108m} to Pd^{108} (1770-keV state) energy difference. Our measurement is in good agreement with this more precise determination.

(b) The number of 79-keV γ rays in coincidence with K x rays was measured in a calibrated geometry in order to derive the fraction of 30-keV isomeric transitions which take place by K conversion. The value we find for this fraction is 0.037±0.005.

(c) Angular correlation measurements have been carried out between all pairs of the 434-, 614-, and 723-keV γ rays. The detectors used in these measurements were 3 in.×3 in. NaI(Tl) scintillators whose front faces were positioned 15 cm from the centered source. The data were corrected for the finite geometry of the experiment according to the method of Rose.[16] The angular correlation coefficients derived from these measurements by least-squares fits to the data are listed in Table II.

(d) An attempt has been made to determine the half-life of the 79-keV state of Ag^{108} in the following way. L and M conversion electrons of the 30 keV isomeric transition were detected in a 0.001-in. plastic scintillator mounted on a 56 AVP photomultiplier. The 79-keV photons were detected in a $1\frac{1}{2}$ in.×1 in. NaI(Tl) scintillator mounted on a second 56 AVP photomultiplier. A fast time to amplitude converter of the overlap type was used to measure the time distribution of coincidences. An upper limit of 1×10^{-9} sec was obtained for the half-life of the 79-keV state.

VI. DISCUSSION AND CONCLUSIONS

The decay of Ag^{108m} has been shown to proceed 8.5% of the time via the isomeric branch, in reasonable agreement with an earlier measurement.[1] Coincidence studies have shown that the 30.4-keV isomeric transition is followed by a 79-keV transition, presumably to the ground state of Ag^{108}. From examination of the conversion electron spectra an upper limit of 0.12% can be placed on the intensity of the cross-over isomeric transition relative to the isomeric cascade. The L subshell conversion electron ratios[8] of the 30.4-keV transition determine its multipolarity as $M4$. Similarly, the experimental K and L conversion coefficients[8,9] of the 79-keV transition determine its multipolarity as $E1$.

Experimentally, there exists only a lower limit of 5 yr for the half-life of Ag^{108m}. It is possible, however, to estimate the half-life by comparison to Ag^{110m} which has a similar $M4$ isomeric transition. The initial ratio of Ag^{110m} activity to Ag^{108m} activity in our sample of neutron irradiated natural silver was determined to be 162. If one then assumes the same ratio of neutron cross sections for activation of the isomers in each of the two silver isotopes, the half-life of Ag^{108m} would be approximately 50 yr. Alternatively, if one assumes similar matrix elements for the two $M4$ transitions the half-life of Ag^{108m} would be approximately 200 yr. It seems reasonable, therefore, to expect a half-life of the order of 100 yr for Ag^{108m}.

The Ag^{108} ground-state spin[17] and parity[18] are 1+. The known multipolarities ($M4$ and $E1$) of the transitions in the isomeric cascade, together with the lack of a crossover transition from the isomeric state, make it likely that the 79-keV state and the 110-keV isomeric state have spins and parities 2− and 6+, respectively. If, for example, the spin and parity of the isomeric state were 5+, the unobserved crossover transition would be an $E4$ with an estimated lower γ-ray lifetime limit of $\sim10^8$ yr. This lifetime is about 10^6 times longer than would be expected for a typically slow $E4$ transition similar, for example, to the In^{114m} $E4$ transition. A 6+ assignment is made more plausible by the fact that the Ag^{110m} isomeric state has a measured[19] spin of 6. In addition, a state of spin 2 and odd parity appears as the ground state[20] of Ag^{112}, while the first excited state[21] at 18 keV is 1+.

[13] A. H. Wapstra, G. J. Nijgh, and R. van Lieshout, in *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959), p. 81.
[14] H. Brysk and M. E. Rose, U. S. Atomic Energy Commission Report No. ORNL-1830, 1955 (unpublished).
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[16] M. E. Rose, Phys. Rev. **91**, 610 (1953).
[17] G. K. Rochester and K. F. Smith, Phys. Letters **8**, 266 (1964).
[18] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C.), NRC [5-1-5].
[19] W. B. Ewbank, W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Phys. Rev. **110**, 595 (1958).
[20] Y. W. Chan, W. B. Ewbank, W. A. Nierenberg, and H. A. Shugart, Phys. Rev. **133**, B1138 (1964).
[21] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C.), NRC [60-2-89].

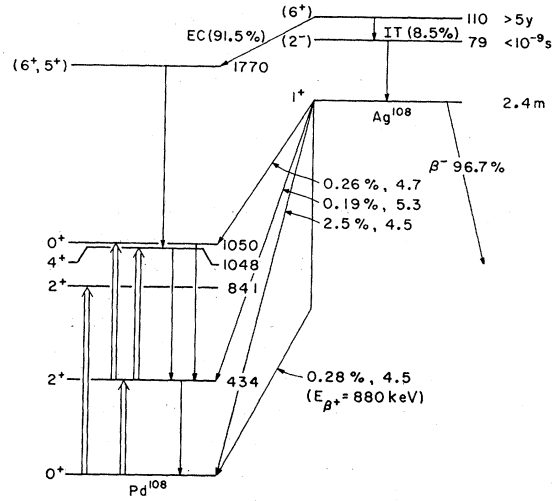

FIG. 5. Decay scheme of Ag^{108m} and Ag^{108}. The intensities and associated logft values of the electron-capture and positron branches from the Ag^{108} ground-state decay are taken from Ref. 15.

Coincidence studies and γ-ray and conversion electron intensity measurements have shown that the electron capture transition from the 6+ isomer of Ag^{108} goes entirely to a 1770 keV level in Pd^{108}. This level then deexcites by a triple cascade to the 0+ ground state. The order of these gamma transitions (723, 614, 434 keV) may be deduced from the following considerations. Coulomb excitation results[7] have established a 2+ first excited state at 434 keV. Double-Coulomb excitation experiments[22] and γ-γ angular correlation studies[23] on the γ rays from the 2.3 min Ag^{108} decay have established the existence of an approximately degenerate 0+ and 4+ doublet at 1050 keV. Since Ag^{108m} most likely has spin and parity 6+, it is reasonable to conclude that the 614-keV γ ray seen in the Ag^{108m} decay is the 4+ $\rightarrow$ 2+ transition seen in the double-Coulomb excitation experiments. This interpretation is supported by our γ-γ angular correlation results, as well as by those of other workers.[2,3] All of these measurements are consistent with the interpretation of the 614–434-keV cascade as a 4+ $\rightarrow$ 2+ $\rightarrow$ 0+ quadrupole-quadrupole cascade which has the theoretical correlation coefficients[24] $A_2=0.1020$ and $A_4=0.0091$.

The conversion coefficients of the 614- and 723-keV transitions (relative to the theoretical $E2$ conversion coefficient[8,9] for the 434-keV transition) are consistent with either $M1$ or $E2$ assignments (see Table I). This establishes the parity of the 1770-keV level as positive. The angular correlation measurements (see Table II) are most consistent with a spin assignment of either 4 or 6 for the 1770 keV state. A spin of 5 is not likely but cannot be definitely excluded by the angular correlation measurements. In particular, the experimental value for A_2 in the 723–614 correlation would indicate a dipole/quadrupole mixture in a 5 $\rightarrow$ 4 transition which would require an $A_4\cong-0.0045$ which is more than two standard deviations from the measured value.

The 4+ possibility for the 1770-keV state may be eliminated by the logft value of the electron capture from the 6+ isomeric state of Ag^{108}. This logft is of the order of 8.5, based on the inferred Ag^{108m} half-life of $\sim$100 yr, while the logft for a $\Delta I=2$ "no" K capture would be expected to be of the order of 13. These conclusions are summarized in the decay scheme of Fig. 5.[25]

It has been suggested[17] that the large magnetic moment ($\pm$4.2 nm) of the Ag^{108} 1+ ground state requires a dominant $(g_{9/2}^{-3})_{J=7/2}$ proton configuration coupled to a $d_{5/2}^{-1}$ neutron configuration.

The small magnetic moment of the 2− ground state of Ag^{112} tends to favor[20] a $p_{1/2}$ proton and a $d_{5/2}^{-1}$ neutron configuration. This state is presumably similar to the 79-keV 2− state of Ag^{108}. Since the natural $M4$ transition in this region involves a $g_{9/2}\rightarrow p_{1/2}$ orbital change for a proton, the 6+ isomeric state could be described by a $g_{9/2}$ proton hole and a $d_{5/2}$ neutron hole coupled to a total $J=6$. However, these configurations would very strongly inhibit the 79-keV $E1$ transition, while the 10^{-9}-sec half-life limit for the 79-keV level implies a retardation factor no larger than about 2×10^3. A $g_{7/2}$ neutron component could be reasonably admixed into the ground-state configuration, and the well-known $h_{11/2}$ neutron orbital in this region is a likely component in the 2− state. These configurations still do not permit the $E1$ transition to proceed. It therefore seems necessary that either the 2− or the 1+ Ag^{108} states contain an admixture which relaxes to some extent this strong $E1$ inhibition.

ACKNOWLEDGMENTS

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[22] D. Eccleshall, B. M. Hinds, and M. J. L. Yates, Nucl. Phys. **32**, 190 (1962).

[23] M. E. Bunker and J. W. Starmer, Bull. Am. Phys. Soc. **5**, 253 (1960).

[24] See, for example, L. C. Biedenharn and M. E. Rose, Rev. Mod. Phys. **25**, 729 (1953).

[25] *Note added in proof.* Gamma-ray spectra from sources of Ag^{108m} and Ag^{108} were examined together and separately with a Li-drifted Ge detector in order to accurately measure the energy difference between the nearly degenerate 0+ and 4+ excited states of Pd^{108}. This energy difference was measured by comparing the energy of the 0+ $\rightarrow$ 2+ γ-ray transition following the Ag^{108} decay with that of the 613.9$\pm$0.6 keV 4+ $\rightarrow$ 2+ transition following the Ag^{108m} decay. The 0+ $\rightarrow$ 2+ transition energy was found to be higher by 4.2$\pm$1.1 keV. The excitation energy of the 0+ state in Pd^{108} is therefore 1051.7$\pm$1.4 keV.