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Decay of Ag^{104} and Levels in Pd^{104} ^{†*}

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The radioactive decay of the isomers of Ag^{104} has been investigated using scintillation spectrometers and a magnetic beta-ray spectrometer. Half-lives of (66 ± 1) and (29.8 ± 0.5) minutes were determined for the spin 5 and spin 2 isomers, respectively. Conversion electrons corresponding to 15 gamma rays assigned to Pd^{104} were observed. Two allowed positron transitions were detected: one of end-point energy (990 ± 10) kev from the spin 5 isomer of Ag^{104} ; and the other of end-point energy (2705 ± 15) kev from the spin 2 isomer. Gamma-gamma and beta-gamma coincidences were observed. The experimental data together with data obtained by other investigators is used to determine the spins and parities of the low-lying excited states of Pd^{104} .

SINCE 1939 the neutron-deficient isotopes of silver have been the subject of many experimental investigations.¹⁻⁸ Because similar radiations and similar half-lives occur in the decays of these isotopes it has been difficult to assign the observed gamma and beta rays unambiguously to the silver isotopes of mass numbers 102, 103, and 104. The tentative assignment by Reynolds *et al.*⁵ of two spin 2 activities to Ag^{104} with half-lives approximately one hour and one-half hour, respectively, stimulated the present work and also concurrent investigations in other laboratories. Ewbank *et al.*,⁶ combining experimental excitation curves with atomic beam measurements, showed that one of the spin 2 activities assigned by Reynolds *et al.*⁵ to Ag^{104} should be assigned instead to Ag^{102} and that Ag^{104} has

spin 5 and spin 2 isomers. Girgis and van Lieshout,⁷ using scintillation spectrometers, identified 10 gamma rays following the decay of the Ag^{104} spin 5 isomer and positrons from the decay of the Ag^{104} spin 2 isomer. Using these data they have proposed a level scheme for Pd^{104} . Ames *et al.*⁸ demonstrated that the spin 2 level in Ag^{104} lies above the spin 5 level.

In the work described here Ag^{104} was studied using both scintillation spectrometers and a magnetic beta-ray spectrometer. We observed conversion electrons corresponding to all 10 gamma rays observed by Girgis and van Lieshout⁷ and in addition, conversion electrons corresponding to several other gamma rays in Pd^{104} were observed. The 2.7-Mev positron transition from the spin 2 isomer of Ag^{104} detected previously by Johnson⁴ and by Girgis and van Lieshout⁷ was observed. A previously unknown 990-kev positron transition from the spin 5 isomer was discovered. Beta-gamma and gamma-gamma coincidences were obtained to confirm certain features of the Pd^{104} level scheme proposed by Girgis and van Lieshout.⁷ By combining our data with those from other investigations we have confirmed the level order proposed by Girgis and van Lieshout and are able to make specific spin and parity assignments to the levels of Pd^{104} . The latter assignments differ somewhat from those suggested by Girgis and van Lieshout.

EXPERIMENTAL MEASUREMENTS

Ag^{104} was produced by the reaction $Rh^{108}(\alpha, 3n)Ag^{104}$ using 38-Mev alpha particles (degraded from the 42-Mev alpha beam of the University of Washington cyclotron.) We chose this bombarding energy to obtain

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¹ T. Enns, Phys. Rev. **56**, 872 (1939).

² W. L. Bendel, F. J. Shore, H. N. Brown, and R. A. Becker, Phys. Rev. **90**, 888 (1953).

³ B. C. Halder and E. O. Wiig, Phys. Rev. **94**, 1713 (1954).

⁴ F. A. Johnson, Can. J. Phys. **33**, 841 (1955).

⁵ J. B. Reynolds, R. E. Christensen, D. R. Hamilton, W. H. Hooke, and H. H. Stroke, Phys. Rev. **109**, 465 (1958).

⁶ W. B. Ewbank, L. L. Marino, W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Phys. Rev. **115**, 614 (1959).

⁷ R. K. Girgis and R. van Lieshout, Nuclear Phys. **13**, 493 (1959); **13**, 509 (1959).

⁸ O. Ames, A. M. Bernstein, M. H. Brennen, R. A. Haberstroh, and D. R. Hamilton, Phys. Rev. **118**, 1599 (1960).

TABLE I. Data on gamma rays from Ag^{104} .

Gamma-ray energies		Relative conversion electron intensity	Relative gamma-ray intensity	Calculated gamma-ray intensity ^a
Present work	Girgis and van Lieshout ^b	Present work	Girgis and van Lieshout ^b	
116 \pm 3 ^c		10		
167 \pm 4		6 \pm 1.8		
262 \pm 3		19 \pm 2.9		2.5 \pm 0.4
355 \pm 3		6.0 \pm 0.6		1.9 \pm 0.2
443 \pm 3		7.3 \pm 0.7		3.4 \pm 0.3
478 \pm 8		2.7 \pm 0.8		1.9 \pm 0.6
556 \pm 3	555 \pm 5	100	100	100
621 \pm 4 ^c		4.3 \pm 0.4		
747 \pm 7	745 \pm 10	4 \pm 2	20 \pm 6	2.2 \pm 1.1
767 \pm 4	780 \pm 10	36.7 \pm 1.0	57 \pm 8	82 \pm 3
854 \pm 5	860 \pm 10	6.7 \pm 0.3	15 \pm 2	18 \pm 0.7
938 \pm 5	935 \pm 5	10.3 \pm 0.4	35 \pm 5	37 \pm 2
1026 \pm 7 ^c		1.2 \pm 0.2		
1074 \pm 7 ^c		0.9 \pm 0.2		
1263 \pm 7	1260 \pm 10	0.9 \pm 0.2	4.0 \pm 1.0	7.2 \pm 1.4
1343 \pm 8	1340 \pm 10	1.6 \pm 0.2	10.0 \pm 1.5	14.3 \pm 2.0
1529 \pm 8	1540 \pm 10	1.0 \pm 0.2	8.3 \pm 1.0	7.5 \pm 2.3
1623 \pm 8	1640 \pm 15	0.73 \pm 0.14	9.0 \pm 1.5	5.5 \pm 1.1
1806 \pm 10	1810 \pm 15	0.63 \pm 0.13	8 \pm 1	9.0 \pm 1.8

^a Calculated from the observed conversion electron intensities, the spin and parity assignments of our proposed Pd^{104} level scheme, and theoretical conversion coefficients.

^b See reference 7.

^c Not assigned to the decay of Ag^{104} .

the maximum yield of the Ag^{104} activity.^{6,9} Most of our data were taken using a 0.0002-inch rhodium target foil as a source without any separation of the Ag^{104} activity from the foil. However, chemical separations were performed to identify the activity, and a few measurements were made with thin sources evaporated from the target foils. We estimate that when equilibrium is established with the bombarding alpha particle beam this method of Ag^{104} production populates the spin 5 and 2 states approximately in the ratio 12 to 1. This is in agreement with the results of Ewbank *et al.*⁶

Conversion electrons from the decay of Ag^{104} were observed using a uniform-field solenoidal beta-ray spectrometer¹⁰ operated with momentum resolution 2.5%. Table I summarizes these data. The decay of the conversion lines was used to determine a half-life of (66 \pm 1) minutes for the Ag^{104} spin 5 isomer.

A single positron spectrum was observed in the magnetic spectrometer corresponding to the decay of the Ag^{104} spin 2 isomer (see Fig. 1). This spectrum, whose end-point energy was found to be (2705 \pm 15) keV, had an allowed shape in the positron energy range above 1.6 Mev. Using positrons of energy 1.8 Mev the half-life of the spin 2 isomer was determined to be (29.8 \pm 0.5) minutes. For this transition we obtain $\log ft=4.7$. We found no evidence for a 1.9-Mev positron branch reported earlier,⁷ our data being consistent with an intensity for such a branch of less than 5% of the principal branch.

As noted above, the $\text{Rh}^{103}(\alpha,3n)\text{Ag}^{104}$ reaction popu-

lates the spin 5 isomer of Ag^{104} more strongly than the spin 2 isomer. The spin 5 isomer, however, decays primarily by K capture, so that the positrons from this decay have an intensity in freshly prepared sources which is only comparable with the intensity of the more energetic positrons from the spin 2 isomer. To observe the positrons from the 66-minute spin 5 isomer without interference from positrons from the 29.8-minute spin 2 isomer thin evaporated sources were prepared and aged 5 or more hours before their positron spectra were observed. When 38-Mev α particles were used to produce such sources, two positron spectra were observed with end points (990 \pm 10) keV and (1345 \pm 70) keV, respectively. Both had half-lives of about one hour. We suspected that the higher energy transition was the same as the 1.3-Mev transition observed by Wiig and Halder⁸ and assigned by them to the decay of Ag^{103} [which could be produced in our source by the $(\alpha,4n)$ reaction on Rh^{103}]. To confirm this we prepared sources with 32-Mev α particles whose energy is less than the threshold for the $(\alpha,4n)$ reaction. These sources exhibited only a single positron spectrum (see Fig. 2) with allowed shape and end-point energy (990 \pm 10) keV. This transition, which we assign to Ag^{104} , has $\log ft=5.25$.

NaI(Tl) scintillation spectrometers and a conventional fast-slow coincidence circuit were used to obtain coincidences between the more intense Ag^{104} gamma rays. We found the 767-, 854-, and 938-keV gamma rays to be in coincidence with the 556-keV gamma ray; the 556-, 854-, and 938-keV gamma rays with the 767-keV gamma ray; and the 556- and 767-keV gamma rays with the 938-keV gamma ray. With a stilbene scintilla-

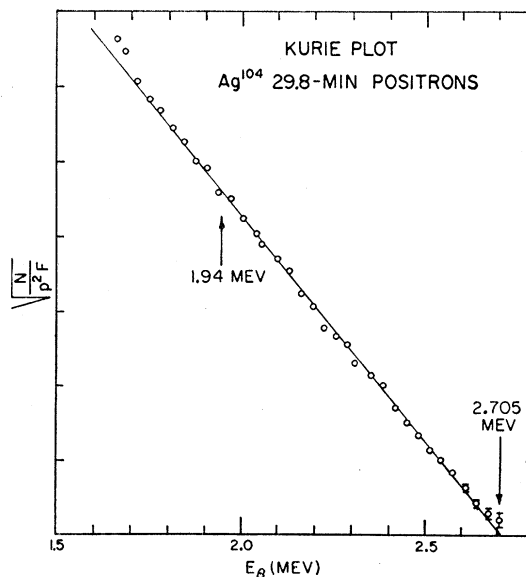


Fig. 1. Kurie plot of the positron spectrum of 29.8-min Ag^{104} . The 1.94-Mev end-point energy corresponding to the transition to the second excited state of Pd^{104} is shown.

⁹ Some measurements were made using sources produced with other alpha particle bombarding energies.

¹⁰ F. H. Schmidt, Rev. Sci. Instr. **23**, 361 (1952).

tion spectrometer to detect positrons we observed coincidences between positrons of greater than 1.5-Mev energy from the spin 2 isomer of Ag^{104} and 556-keV gamma rays, but not with the 767-, 854-, or 938-keV gamma rays.

LEVELS IN Pd^{104}

Conversion electrons corresponding to all of the 10 gamma rays assigned by Girgis and van Lieshout⁷ to the decay of Ag^{104} were observed, though slightly different gamma-ray energies were obtained in some instances (see Table I). Conversion electrons corresponding to 167- and 262-keV gamma rays tentatively assigned to the decay of Ag^{103} by Ames *et al.* were observed but we assign these to crossover transitions in the Pd^{104} level scheme. In addition, we assign conversion electrons corresponding to gamma rays of 355, 443, and 478 keV to the Pd^{104} level scheme. The 116-keV gamma ray we observed we assign to the decay of Ag^{103} as do other investigators.^{7,8} We cannot fit the weak 621-, 1026-, and 1074-keV gamma rays into the Pd^{104} scheme and they may arise from the decay of Ag^{103} though we could not confirm this supposition.

From our experimentally determined gamma-ray energies we can construct a number of possible level schemes for Pd^{104} . Figure 3 shows the level scheme which we adopt. It is the same as that proposed by Girgis and van Lieshout⁷ except that the energy values assigned to the various levels are based on our data and that the spin and parity assignments for several levels are different. Our observed gamma-ray energies agree within

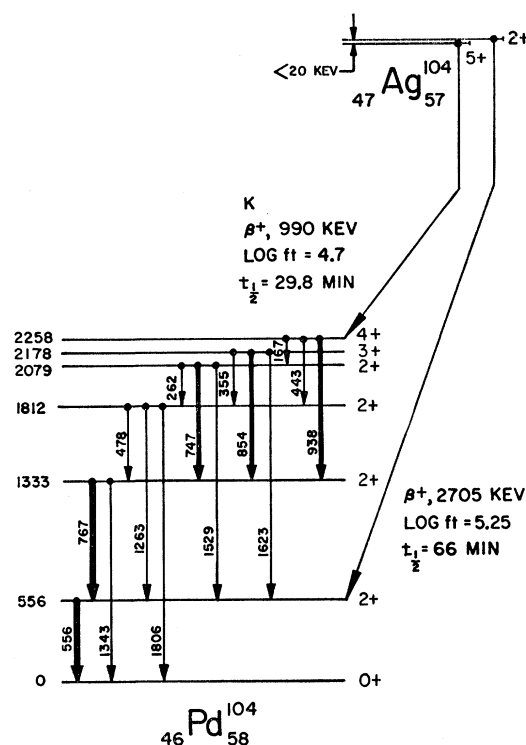


FIG. 3. Proposed decay scheme of Ag^{104} .

experimental uncertainty with the values predicted from this level scheme. Any other satisfactory level scheme based on our data would require additional levels. In the absence of further data we prefer to choose this most economic scheme.

The assignment of the 2705-keV positron transition from the Ag^{104} spin 2 isomer to the 556-keV state of Pd^{104} is suggested by the fact that the spin of the 556-keV level is known^{11,12} to be 2. Consequently this would be the normal decay route. Our beta-gamma coincidence data confirm this supposition.

The assignment of the 990-keV positron transition from the spin 5 isomer of Ag^{104} to the 2258-keV level in Pd^{104} is necessary since, as Ames *et al.*⁸ have shown, this isomer lies below the spin 2 isomer. Its assignment to a possible higher state in Pd^{104} would require reversal of the Ag^{104} level order. Its assignment to a lower level in Pd^{104} would make the energy separation of the two Ag^{104} levels so great that the spin 2 level of Ag^{104} would decay predominantly by a gamma transition to the spin 5 level instead of by positron emission. The fact that the 990-keV positron transition to the 2258-keV level of Pd^{104} is allowed in character, and that there is no evidence for higher energy positron transitions to the 2079- and 2178-keV levels in Pd^{104} makes it very unlikely that these three levels have the same spin as was

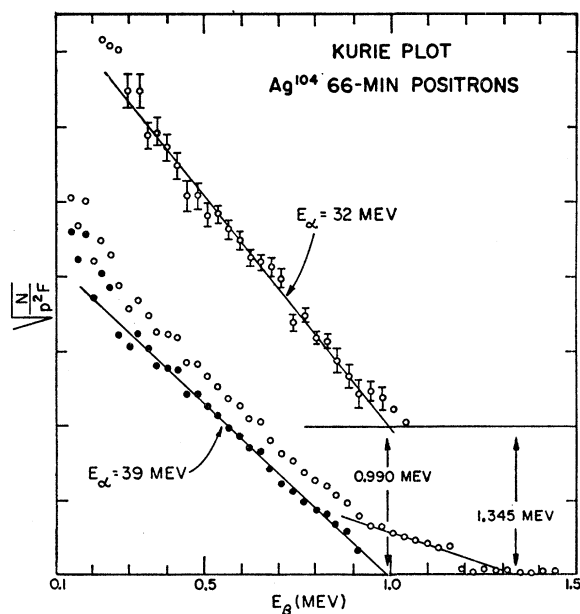


FIG. 2. Kurie plots of the positron spectrum of 66-min Ag^{104} . The upper curve was made with a source produced with 32-MeV α particles and does not exhibit the 1.345-MeV positron spectrum of Ag^{103} .

¹¹ G. M. Temmer and N. P. Heydenburg, Phys. Rev. **104**, 967 (1956).

¹² P. H. Stelson and F. K. McGowan, Phys. Rev. **110**, 489 (1958).

suggested by Girgis and van Lieshout.⁷ This evidence limits the choice of spin for the 2258-kev level to 4, 5 or 6 and for the 2079- and 2178-kev levels to less than 4 or more than 6. On the basis of the decay scheme adopted our data indicates that the spin 5 and spin 2 levels in Ag^{104} differ in energy by less than 20 kev.

The ground state of Pd^{104} is 0^+ since this is an even-even nucleus. The 556-kev excited state was shown to be 2^+ by Coulomb excitation.^{11,12} Since the 556-kev gamma ray is known to be from an $E2$ transition, we used our relative conversion electron intensities and Girgis and van Lieshout's⁷ relative gamma-ray intensities (in combination with the theoretical conversion coefficient for the 556-kev gamma ray) to determine conversion coefficients for the other gamma rays. From these conversion coefficients and the relative gamma-ray intensities we were able to make the spin and parity assignments indicated in Fig. 3. These assignments are the only ones compatible with all the available data and with the energy level scheme adopted.

Very recently Bunker and Starnier¹³ have reported the results of their angular correlation measurements on the 1.24- and 0.56-Mev Pd^{104} gamma rays following the decay of Rh^{104} . There is no doubt that the 0.56-Mev gamma ray is the same as the 556-kev gamma ray observed in the Ag^{104} decay. The angular correlation measurements indicate, however, that the 1.24-Mev gamma ray originates in a 0^+ state of Pd^{104} . This conclusion is also confirmed by the absence of a 1.8-Mev cross-over transition in the Rh^{104} decay. On the other hand, both our data and that of Girgis and van Lieshout⁷ on the Ag^{104} decay exhibit a 1.8-Mev gamma ray of greater intensity than the 1.2-Mev gamma ray. These different observations are compatible if we assume there are two levels of Pd^{104} at about 1.8 Mev. The level populated in the decay of Rh^{104} is 0^+ . The level populated in the Ag^{104} decay is 2^+ . The fact that the 0^+ level is not observed in the Ag^{104} decay is compatible with the decay scheme shown in Fig. 3.

¹³ M. E. Bunker and J. W. Starnier, *Bull. Am. Phys. Soc.* **5**, 253 (1960).

DISCUSSION

There have been numerous attempts to account for the frequent occurrence of the spin and parity sequence $0^+, 2^+, 2^+$ for the lowest levels of even-even nuclei. The most recent study of these levels, a survey by Van Patter¹⁴ of 55 known examples in nuclei with $A > 30$, concludes that the asymmetric rotor model developed by Davydov and Filippov¹⁵ provides the best description of these nonspherical nuclei. What is known of the 3 lowest levels of Pd^{104} is in good agreement with this conclusion. Experimentally, less is known of the higher levels in even-even nuclei. Often the third excited state is a 4^+ level and most theoretical treatments have predicted the existence of 4^+ levels in this region. In this respect Pd^{104} appears to be somewhat unusual, since any 4^+ level of less than 2-Mev excitation should have been populated by beta decay from the 5^+ isomer of Ag^{104} .

As discussed earlier, there apparently is a 0^+ level in Pd^{104} at about 1.8 Mev. There could also be 0^+ levels in the range from 2 to 2.2 Mev near the $2^+, 3^+$, and 4^+ levels. Such levels would not be strongly populated by the Ag^{104} decay and consequently would remain undetected. The absence of low-lying spin 1 levels, and the fact that the lowest spin 3 level lies so high are in good agreement with theoretical predictions for even-even nuclei.¹⁶

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¹⁴ D. M. Van Patter, *Nuclear Phys.* **14**, 42 (1959).

¹⁵ A. S. Davydov and G. F. Filippov, *Nuclear Phys.* **8**, 237 (1958); **10**, 654 (1959).

¹⁶ R. K. Sheline, *Revs Modern Phys.* **32**, 1 (1960).