PHYSICAL REVIEW VOLUME 118, NUMBER 6 JUNE 15, 1960

Spins and Decay Modes of Certain Neutron-Deficient Silver Isotopes*

O. AMES, A. M. BERNSTEIN, M. H. BRENNAN, R. A. HABERSTROH, AND D. R. HAMILTON
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey

(Received January 22, 1960)

Isotopically enriched Pd foils were bombarded with protons to determine the origin of the 1.2-hr activity which was previously discovered in an even-A Ag isotope. The only appropriate activity was found in Ag^{104} which was observed to have a (69 ± 3)-min predominantly K-capture activity with intense γ rays of 550, 764, and 920 kev. Using the atomic beam magnetic resonance method it was verified that the 69-min activity has $I=5$. The $I=2$ resonance in Ag^{104}, when counted in an x-ray detector, was found to decay with a 69-min half-life with a small admixture of a 27-min component. When viewed in a β counter, only the 27-min component is observed. These characteristics of the $I=2$ resonance can be explained by placing the $I=2$ level above the $I=5$ level with an appreciable amount of isomeric transition. This interpretation is supported by the observation of feeding in the 920-kev γ ray which only occurs in the decay of the $I=5$ state.

Further work with isotopically-enriched Pd foils showed γ rays of 120 and 150 kev, and, tentatively, 260 kev which have been assigned to the decay of 59-min Ag^{103}. A positive identification of a (15 ± 2)-min activity in Ag^{102} has been made.

I. INTRODUCTION

THE assignments of half-lives, spins, and γ rays to the neutron-deficient silver isotopes with mass numbers 102, 103, and 104 has proved to be very difficult. Following the early work of Enns[1] in 1939, various investigations[2–7] have not been able to provide complete and consistent assignments.

Particular difficulty has been experienced in identifying the origin of approximately 1.1-hour γ rays and spins in this region. Gamma rays of 554 kev and 764 kev decaying with half-lives of 66 minutes were found by Bendel and co-workers[2] and assigned by Haldar and Wiig[4] to the decay of Ag^{103}. Also, a γ ray of 555 kev following the positron decay of 27-min Ag^{104} was found by Johnson.[3] The possibility that the 554-kev γ ray found by Bendel *et al.* resulted from the decay of a 1.1-hour level in Ag^{104} was increased following the results of spin measurements made by the Princeton atomic beam group.[5] They found a strong 1.2-hour activity with a nuclear spin, I, of 2. However, the assignment of this activity to Ag^{104} would have required the occurrence of isomerism between two states having the same nuclear spin, since this group also positively assigned a spin of 2 to the 27-min level in Ag^{104}. Also, recent work of the Berkeley atomic beam group has shown that there is a 1.2-hour, $I=5$ level in Ag^{104}.[6] On the other hand, the assignment of the 1.2-hour, $I=2$ activity to Ag^{102} would require an abnormally high cross section for the reaction $Pd^{102}(p,n)Ag^{102}$.[5] The possibility that this activity came from another silver isotope produced by bombarding palladium with the 18.5-Mev protons from the Princeton cyclotron could not be definitely excluded, although these isotopes appear to have been studied[7] in sufficient detail to make this unlikely.

These results clearly indicated the need for further investigations of the neutron-deficient silver isotopes. The present work was therefore directed towards obtaining positive identification of half-lives, associated γ rays, and, where possible, nuclear spins of the ground states and isomeric states for these nuclei. The assignments of the half-lives and γ rays of the various isotopes were made following an analysis of data obtained after bombarding foils containing varying enrichments of the stable palladium isotopes. This work is described in Secs. II and IV. In addition, the atomic beam magnetic resonance technique was used to investigate in some detail the characteristics of the 1.2-hour spin-2 activity seen by Reynolds *et al.*[5] This work is described in Sec. III.

II. EVEN-A 1.2-HOUR ACTIVITY

In order to identify the 1.2-hour activity seen by Reynolds et al.,[5] isotopically-enriched and natural palladium foils were bombarded with protons of 11-Mev energy. This energy is below the $(p,2n)$ threshold, insuring that Ag^{103} would not be produced. (Pd^{103} is not stable.) The 18.5-Mev protons of the Princeton cyclotron were reduced to 11 Mev by 0.014 inch of copper foil.

Four palladium foils enriched in isotopes with mass numbers of 102, 104, 106, and 108 were bombarded in turn. Each enriched foil was placed in front of a natural foil cut to have approximately the same shape as the enriched foil. The relative enrichments of the two foils which proved most useful in the identification of the 1.2-hour activity are given in Table I.

The decays of the various activities produced were followed in an x-ray counter consisting of a 1-mm thick NaI crystal with a 0.001-inch aluminum window viewed

* This work was supported by the U. S. Atomic Energy Commission and the Higgins Scientific Trust Fund.
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 F. A. Johnson, Can. J. Phys. **33**, 841 (1955).
4 B. C. Haldar and E. O. Wiig, Phys. Rev. **94**, 1713 (1954).
5 J. B. Reynolds, R. L. Christensen, D. R. Hamilton, W. M. 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 D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

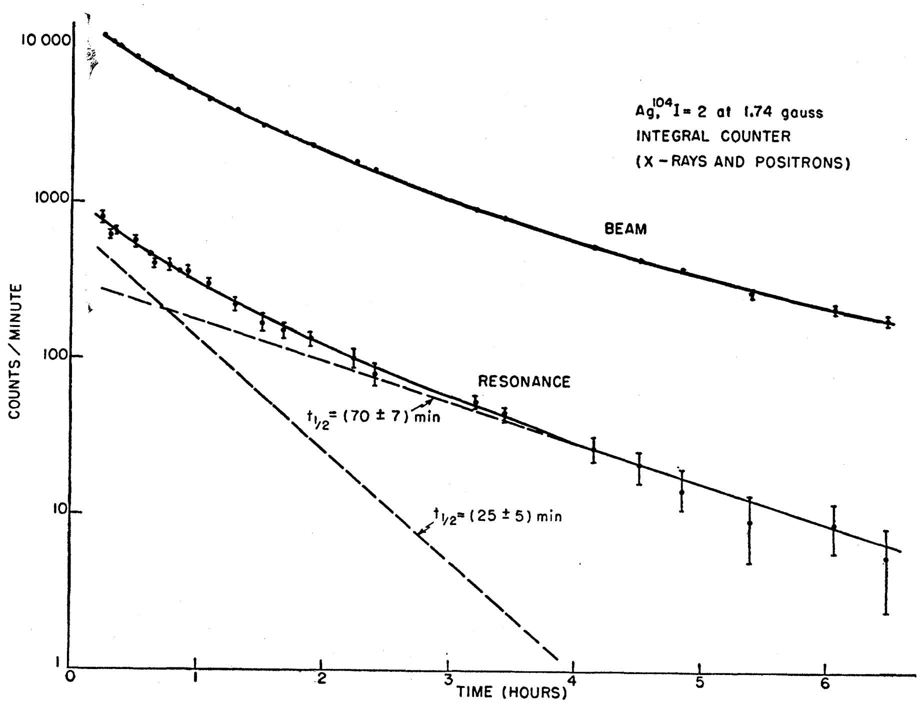

FIG. 1. Decay of beam and resonance activity for the spin-2 resonance in Ag^{104}. Counter integral bias set below the palladium x-ray peak. Errors shown are standard deviations. The solid curve drawn for the resonance data is the sum of the two dashed curves.

by a 2-inch photomultiplier. The counter was set to record x-rays of energies 10 to 30 kev. An x-ray counter was chosen because the 1.2-hour component of the spin-2 activity was not observed when a β counter was used,[5] indicating the presence of K x-rays in its decay.

Following the bombardment of the enriched Pd^{104} foil, an enhanced (69 ± 3)-min activity was found. The uncorrected enhancement ratio for this activity was 5.2. This ratio has to be corrected for the difference in proton energy in the enriched and natural foils and for a small difference in their masses. After applying these corrections the enhancement ratio becomes 5.8 ± 1.2, where the error is an estimate based on the uncertainty in the variation of excitation as a function of energy. This enhancement agrees well with the Pd^{104} enrichment factor of 5.7 (see Table I). This evidence alone is sufficient to assign the 69-min activity to Ag^{104}.[8]

TABLE I. Percentage abundances[a] of the isotopes in natural palladium and in foils enriched in Pd^{102} and Pd^{104}.

Mass number	Natural foil	Enriched Pd^{102}	Enriched Pd^{104}
102	0.96	35.2 ± 0.9	...
104	10.97	26.3 ± 0.3	63.20 ± 2.43
105	22.2	22.0 ± 0.5	23.22 ± 0.60
106	27.3	11.9 ± 0.2	9.51 ± 0.74
108	26.7	3.9 ± 0.1	2.78 ± 0.71
110	11.8	0.7 ± 0.1	1.38 ± 0.59

[a] The natural abundances are taken from the review article of Strominger, Hollander, and Seaborg.[7] The abundances for the enriched foils were supplied, with the foils, by Oak Ridge National Laboratory.

[8] The value obtained in this work for the half-life is in agreement with the value of 70 min obtained by Girgis and van Lieshout and with the value of 1.2 hours obtained by Ewbank *et al.*[6] These determinations were made following bombardments of rhodium with α particles.

Following the bombardment of the enriched Pd^{102} foil a strong (15 ± 2)-min activity was observed. No quantitative enhancement ratio could be obtained in this case since this activity was not detected in the decay of the natural foil, owing to the low natural abundance of Pd^{102}. No evidence for an approximately one-hour activity in Ag^{102} was found. The present work appears to provide the first direct experimental evidence for the existence of a 15-min activity in Ag^{102}.[9]

Bombardments of the other foils, enriched in Pd^{106} and Pd^{108}, provided additional confirmation of the assignment of the 69-min activity to Ag^{104} and indicated that there were no other even-A silver isotopes in the range $102\leq A\leq108$ with half-lives of approximately one hour.

The question of the association of a one-hour activity with Ag^{103} is coupled with the identification of some of the γ rays in its decay and will be discussed in Sec. IV.

III. ATOMIC BEAM EXPERIMENTS

The radioactive silver for the beams experiments was produced by bombarding natural palladium foils[10] with 18.5-Mev protons. After removal from the cyclotron, the foils were placed in a molybdenum oven in

[9] This activity should not be confused with the 16-min activity seen by Enns.[1] The reassignment by Strominger, Hollander, and Seaborg,[7] of this activity to Ag^{102} would appear to require an abnormally large $Pd^{102}(p,n)Ag^{102}$ cross section, which is not suggested by our present work.

[10] These foils were spectroscopically analyzed by Dr. Morris Slavin of the Chemistry Department of Brookhaven National Laboratory and were found to contain no impurities in excess of 0.1%.

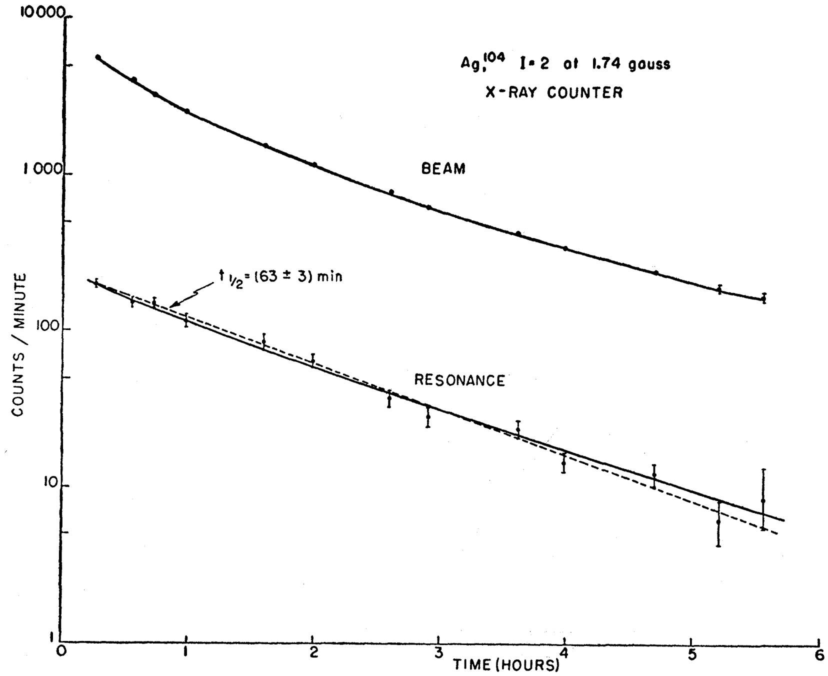

FIG. 2. Decay of beam and resonance activity for the spin-2 resonance in Ag^{104}. Counted in the palladium x-ray region (see text). The solid curve drawn for the resonance is the sum of a 69-min and a 27-min decay.

the beams apparatus[11] and a beam of silver atoms was obtained by heating the oven to approximately 1600°C. Resonances were obtained in "flop-in" so that they appeared as an increase in intensity over the normal background signal. The detector and stop system described by Lemonick, Pipkin, and Hamilton[11] was modified to allow the unflopped beam to be observed at all times, thus providing a method for monitoring the beam intensity. The radioactive atoms were collected on carefully cleaned copper disks and the counting system described in the previous section was used.

The $I=2$ resonance was the first one to be studied. It was observed at several values of the homogeneous field from 1.74 to 71.4 gauss. Figure 1 shows the decay of this resonance where the counters were set to record all pulses greater than about 10 kev. With this setting, positrons and x-rays are detected with high efficiency, and γ rays with considerably reduced efficiency. The short half-life component of this decay has previously been identified as Ag^{104}.[3,5] The long half-life component is that seen by Reynolds *et al.*[5] Figure 2 shows the decay of the same resonance with the counters biased to record those pulses in the energy range from 10 to 30 kev. Using this setting of the counters, only the palladium x-rays arising in the decay of the silver isotopes are detected with high efficiency. The shape of this curve is consistent with a single activity of (63 ± 3) min or with a 69-min activity plus a 20% admixture of the 27-min activity. From the two figures it follows that the ratio of the long-lived resonance activity as counted integrally from 10 kev to that counted in the interval from 10 to 30 kev is approximately 1.8 to 1.

Further spin searches revealed an $I=5$ resonance. This was observed at 1.34, 3.99, and 8.06 gauss. It should be noted here that the full width at half maximum for resonances obtained in this apparatus is about 25 kc/sec, so that reliable spin searches could be made at low values of the homogeneous field. Figure 3 shows the decay of an $I=5$ resonance. This curve was obtained in the same manner as that shown in Fig. 1. Further runs showed that the ratio of the 72-min $I=5$ activity counted integrally to that counted in the range from 10 to 30 kev was about 1.8 to 1.

The following conclusions may now be drawn by combining the enriched foil data with those from the beams apparatus work.

First, Ag^{104} is the only neutron-deficient even-A silver isotope which shows the presence of any approximately one hour activity. The $I=5$ (72 ± 4)-min activity therefore must be in Ag^{104}. This result is in agreement with the work of Ewbank *et al.*[6]

Second, there is a strong (70 ± 7)-min component in the $I=2$ resonance.[12] Finally, the 27-min $I=2$ activity is known to be in Ag^{104}.

Two possible schemes are consistent with this information. Either there are t o $I=2$ levels and an $I=5$

[11] A. Lemonick F. M. Pipkin, and D. R. Hamilton, Rev. Sci. Instr. **26**, 1112 (1955).

[12] The reason for the failure of Ewbank *et al.* to see this component of the $I=2$ resonance is not clear. It may, however, be due to the fact that the spin-2 resonance decay curves could only be followed using positron counters which, as found previously by Reynolds et al.,[5] show a pure 27-min decay for this resonance.

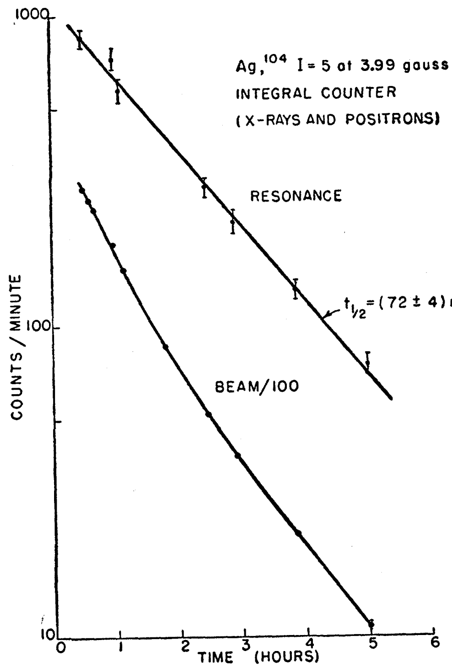

FIG. 3. Decay of beam and resonance activity for the spin-5 resonance in Ag^{104}. Counter integral bias set below the palladium x-ray peak.

level in Ag^{104}, one or both of the spin-2 levels being isomeric; or the ground state of Ag^{104} is the $I=5$ state, and there is one $I=2$, 27-min isomeric state.

Preliminary results from measurements at 71.4 gauss indicate that if there are two $I=2$ levels in Ag^{104}, they both must have positive magnetic moments of several nuclear magnetons. The necessary similarity in configurations to give two states of $I=2$ and positive magnetic moment makes it extremely unlikely that one state would be isomeric with respect to the other.

We therefore consider the second scheme. Here it is assumed that the $I=2$, 27-min level is the isomeric state in Ag^{104}. From the work of Reynolds et al.,[5] it is known that the decay of this level to Pd^{104} goes primarily by β^+ emission. (Using the β^+ end-point energy of 2.7 Mev,[3] we would expect a K/β^+ ratio of 20% for this allowed transition.) If in addition to this decay mode the $I=2$ level decays to the $I=5$ level, then the resonance decay should be a composite of a 27-min activity and a 69-min activity as seen in Fig. 1. If the counter used to observe the $I=2$ resonance decay is set to look only at the Pd K x-ray, the picture will be somewhat different. If one were to assume that the $I=2$ level decays to Pd^{104} purely by β^+ emission and to the $I=5$ level by gamma emission only, then the $I=2$ resonance decay curve would show a build-up followed by the 69-min decay, i.e., a characteristic feeding curve would be seen.

It is clear, however, that this shape will be modified by the detection of the 27-min K x-rays in the decay of the $I=2$ level to Pd^{104} and of any K x-rays resulting from internal conversion of the isomeric transition. The decay curves shown in Figs. 1 and 2 would be obtained if the branching ratio for the isomeric transition in the decay of the spin-2 level were between 20% and 40%, with a corresponding range of zero to 0.5 for the number of K x-rays emitted per isomeric transition.

One further point should be mentioned. It was observed earlier that the ratio of the 69-min activity as counted integrally to that counted in the K x-ray region was about 1.8 to 1 for both the decay of the $I=2$ resonance and the $I=5$ resonance. This strongly suggests the identity of the $I=5$ activity and the 69-min activity observed in the $I=2$ resonance decay.

Summarizing, the work described in Secs. II and III is consistent with the following picture. The ground state of Ag^{104} has spin 5 and decays to Pd^{104} primarily by K capture with a half-life of 69 ± 3 minutes. The isomeric state is low-lying, has a spin of 2 and a half-life of 27 minutes. It decays to Pd^{104} primarily by β^+ emission. There is also an isomeric transition of appreciable intensity.

IV. GAMMA-RAY DATA

In view of the small difference between the half-lives of Ag^{103} and the ground state of Ag^{104}, it was decided to check the assignment of the γ rays in the decay of these nuclei. First, an enriched Pd^{104} foil was bombarded in the (p,n) region, as described in Sec. II. The γ rays were detected by a scintillation counter consisting of a 3 in.×3 in. NaI crystal and photomultiplier. The spectrum was displayed on a 200-channel analyzer and, where possible, the decay of each γ-ray peak was

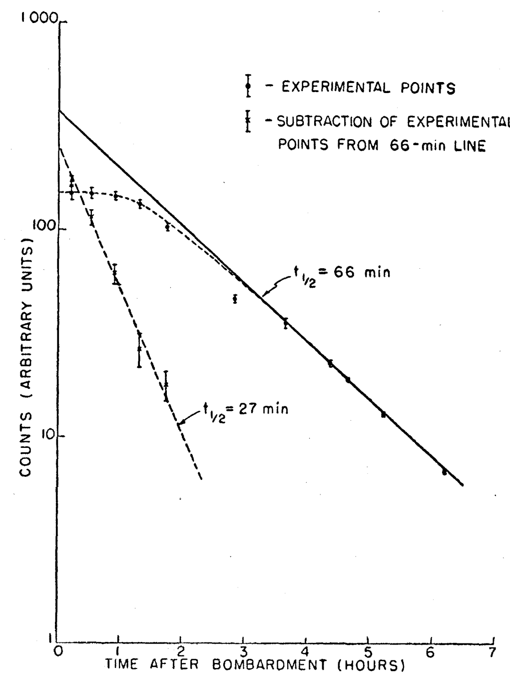

FIG. 4. Feeding in the decay of the 920-kev gamma ray.

followed. Peaks due to the decay of either level in Ag^{104} were easily identified by comparing the spectra obtained with the natural and enriched foils.

Gamma rays of 555, 760, 860, and 920 kev were identified as arising from the decay of Ag^{104}.[13] It was not possible to obtain an accurate enhancement for the positron annihilation peak because of contributions from various positron decays, the 555-kev γ ray, and a 512-kev γ ray in the decay of 24-min Ag^{106}.[7] However, the 2.7-Mev positron in the decay of 27-min Ag^{104} has been previously reported by Johnson.[3]

In following the decay of the 760- and 920-kev γ rays a feeding curve was observed. The decay of the 920-kev peak is shown in Fig. 4. It is seen that there is a 27-min growth and a 66-min decay. The count rate does not begin at zero since both states in Ag^{104} are initially populated by the nuclear reaction. The 555-kev peak shows a decay which is a composite of 27-min and 69-min decay curves. No accurate decay curve for the 860-kev γ ray could be obtained since this peak, which is relatively weak, is partly masked by the 760- and 920-kev peaks.

The γ rays attributed here to the decay of Ag^{104} have also recently been studied by Girgis and van Lieshout,[14]

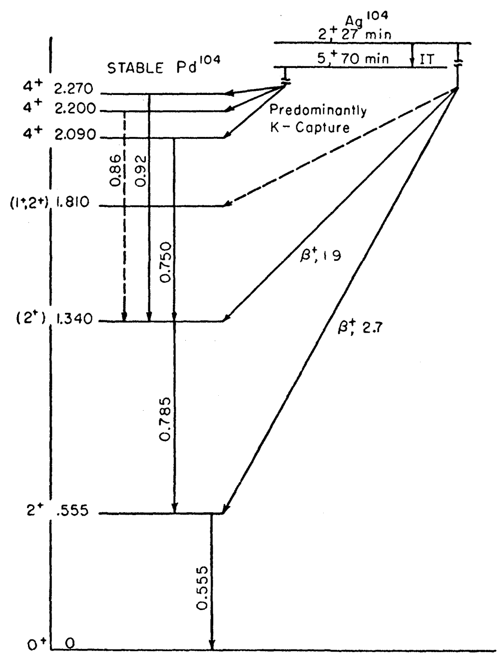

FIG. 5. Level ordering predicted for Ag^{104} and the gamma rays observed in its decay. These results are fitted to the level scheme of Girgis and van Lieshout, reference 14. A dotted gamma transition means that the assignment to Ag^{104} is not firm. All energies are in Mev. *Note.*—The notation "5,+70 min" at the top of the Figure should read "5,+69 min."

[13] Gamma rays of 554 and 764 kev have previously been reported by Bendel *et al.*,[2] and incorrectly assigned to Ag^{103} by Haldar and Wiig.[4]

[14] R. K. Girgis and R. van Lieshout, Nuclear Phys. **13**, 493 and 509 (1959). We are grateful to these authors for supplying us details of their work prior to publication.

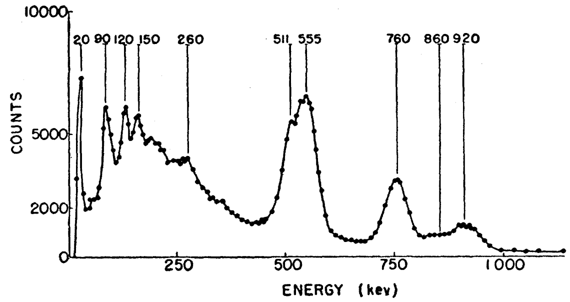

FIG. 6. Gamma-ray spectrum of natural Pd foil taken three hours after bombardment with protons of 18.5-Mev energy.

whose level scheme for Pd^{104} together with the γ rays observed in the present work are shown in Fig. 5. Our 760-kev γ ray is probably a superposition of the (745 ± 10) kev and (780 ± 10) kev γ rays seen by Girgis and van Lieshout. In this figure we have omitted several γ rays with energies greater than 1 Mev, which were observed in the present work but for which positive assignments could not be made. No γ rays were seen which were not consistent with the Pd^{104} level scheme proposed by Girgis and van Lieshout.

It will be noted that our observation of feeding in the 920-kev γ ray provides additional confirmation of the level ordering in Ag^{104} deduced from the work described in Sec. III; we see from Fig. 5, that this γ ray occurs only in the decay of the spin-5 level, so that the presence of feeding in its decay implies that the spin-2 level is above the spin-5 level.

Further confirmation that the $I=5$ level is in Ag^{104} and that its decay scheme includes γ rays of 555, 760, and 920 kev was obtained from an analysis of the γ-ray spectrum of the activity collected during an atomic beam experiment. Despite the very small activity deposited on the copper collectors, it was possible to obtain the spectrum of the spin-5 resonance activity and to compare it with the spectrum of the straight-through, "unflopped" beam. It was found that, as in the experiments with the enriched Pd^{104} foils, the 555-, 760-, and 920-kev γ rays were enhanced in the spin-5 resonance activity, thus identifying this activity with the 69-min level in Ag^{104}.

Having positively identified all the prominent γ rays below 1 Mev in the decay of Ag^{104}, enriched Pd^{104} and natural palladium foils were bombarded with protons of 18.5-Mev energy. This energy is above the threshold for the reaction $Pd^{104}(p,2n)Ag^{103}$, enabling us to investigate the decay of Ag^{103}. A spectrum obtained three hours after bombarding natural palladium with protons of 18.5-Mev energy is shown in Fig. 6.

Gamma rays of 120 and 150 kev were found which had not been present in the experiments in which the proton energy was below the $(p,2n)$ threshold. The enhancement ratio of 6.5 ± 2.0 for these peaks compared well with the value of 7.12 deduced from the Pd^{104}

TABLE II. Summary of available data on three neutron-deficient silver isotopes.[a]

Isotope	Spin	Half-life (min)	Gamma-ray energies (kev)	Positron energies (Mev)
Ag^{102}	...	15		Some K capture
Ag^{103}	7/2[b]	59	120, 150, 260(?)	1.3
Ag^{104}	5	69	555, 745, 780 860, 935, 1260 1340, 1540, 1640 1810.	Predominantly K capture
Ag^{104m}	2	27	555, 780	2.7, 1.9

[a] References are given in the preceding sections of this paper.
[b] W. B. Ewbank, L. L. Marino, W. A. Nierenberg, H. A. Shugart, and H. B. Silsbee, Bull. Am. Phys. Soc. 3, 370 (1958).

enrichment factor for the foils used.[15] A peak was also observed at 260 kev, for which an accurate enhancement ratio could not be obtained. The internal consistency of the data was checked by noting that the 760-kev peak previously ascribed to Ag^{104} was enhanced by the appropriate factor. Accurate half-lives for these peaks could not be obtained. However, their half-lives were consistent with the half-life of 59 min previously reported for Ag^{103}.[4] We may therefore assign the 120- and 150-kev γ rays to the decay of Ag^{103}. The assignment of the 260-kev γ ray to Ag^{103} is less certain. Girgis and van Lieshout have also tentatively assigned a γ ray of 120 kev to the decay of Ag^{103}.

V. DISCUSSION

We are now in a position to add the results of the present work to those obtained in previous investigations on the neutron-deficient silver isotopes. A summary of the presently available data on these isotopes is given in Table II.

We note, at this point, that the γ ray or conversion electron of the isomeric transition in Ag^{104} has not yet been detected. In particular, Johnson[3] did not observe conversion electrons which could be assigned to this transition. This may indicate that the energy of the transition is less than 25 kev. We also note that, contrary to the results presented in the preceding section, Girgis and van Lieshout[14] find no evidence of feeding in the 920-kev γ ray. This may be due to the smallness of this peak and to the relatively low population of the spin-2 level in Ag^{104} when the reaction $Rh^{103}(\alpha,3n)Ag^{104}$ is used. This conjecture follows from an analysis of the resonance data of Ewbank et al.,[6] in which the spin-2 resonance is very weak compared with the spin-5 resonance.

Finally, we comment on the proton and neutron configurations in the even-A silver isotopes. The spins of the odd-even nuclei in this region indicate that there is a competition between $(p_{\frac{1}{2}})^1(g_{9/2})^{-2}$ and $(p_{\frac{1}{2}})^2(g_{9/2})_{7/2}{}^{-3}$ configurations in the case of the protons and that the neutron configuration is $(d_{\frac{5}{2}})^{-1}$. The assignment of even parity to the levels in Ag^{104} and Ag^{106} [7,14] suggests that for these two nuclei the proton configuration is $(g_{9/2})_{7/2}{}^{-3}$. [A $(g_{9/2})_{9/2}{}^{-3}$ configuration is also possible.] This is supported by preliminary measurements at this laboratory indicating that the magnetic moments of the two levels in Ag^{104} are large and positive.

It is of interest that the occurrence of isomerism in the odd-odd nuclei in this "island of isomerism" is due to the recoupling of angular momenta rather than to the single-particle excitation found in neighboring odd-even nuclei. Several interesting features of this angular momentum recoupling will be the subject of a future paper.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge interesting discussions with Dr. R. A. Naumann. We are also indebted to David Harris and J. C. Walker for assistance in taking data.

[15] The enriched foil used here had a higher enrichment than is shown in Table I.