Spins of Certain Short-Lived Cu and Ag Isotopes*

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Spins of several short-lived isotopes of Cu and Ag have been measured by the atomic-beam method. Spin values are as follows: Cu^{60}, $I=2$; Cu^{61}, $I=\frac{3}{2}$; 27-minute Ag^{104}, $I=2$; 24-minute Ag^{106}, $I=1$; and $I=2$ for a 1.2-hr Ag activity, the identity of which is not certain, but which may be attributed to Ag^{102} or Ag^{104}.

INTRODUCTION

BOMBARDMENT of Pd and Ni by approximately 19-Mev protons from the Princeton cyclotron produces several short-lived isotopes of Ag and Cu in quantities sufficient to perform atomic-beam measurements of spins. The half-lives involved are shorter than is that of any isotope that has previously been studied in the Princeton atomic-beam machine; however, the close proximity of the cyclotron to the beam machine together with some streamlining of ordinary operating techniques for the machine made spin measurements possible. Measurements of magnetic moments of the short activities were not attempted in this experiment. However, indications are that such measurements are possible and may be done in the future.

EXPERIMENTAL PROCEDURE

Copper isotopes were prepared by bombarding thick targets of "A" nickel (manufacturer's analysis: 99.40% Ni, 0.10% Cu; 0.15% Fe; 0.10% C; 0.20% Mn; 0.05% Si; 0.005% S) with 18.8-Mev protons. The resulting isotopes were identified by half-lives[1] as 25-min Cu^{60}, 3.3-hour Cu^{61}, and 12.8-hr Cu^{64}. It was estimated that a 30-minute bombardment at about 1 microampere beam current yielded between 10 and 40 mC of Cu^{60}. Considerably less Cu^{61} and Cu^{64} isotopes were produced although sufficient Cu^{61} was made to allow spin determination. There were indications of some Cu^{62} being produced. However, the amount was small and would have been impossible to work with in the presence of the much more abundant Cu^{60}.

Silver isotopes were prepared by bombarding thick targets of Pd with 18.8-Mev protons. Spectroscopic analysis of the Pd foil gave the following impurity content: about one part in 10^4 each of C, Ag, and Mg and traces of Au, Fe, B, and Si. Silver isotopes produced[1] were 27-min Ag^{104}, and 24-min Ag^{106}, and a 1.2-hr activity which has been variously attributed to Ag^{102}, Ag^{103}, and Ag^{104} (see below).

The cyclotron bombardments lasted for approximately one-half hour for the shorter activities and for about one half-life of the Cu^{61} and Ag 1.2-hr activities. Immediately after bombardment, the active material was trimmed from the target tip and placed in the atomic-beam oven, which in turn was inserted in the oven chamber of the atomic-beam machine. This procedure took about 10 minutes. Evacuating the oven chamber and aligning and heating the oven took an additional 10 minutes, so that the run was usually under way within 20–25 min of completion of bombardment. The atomic-beam machine used was the focusing apparatus described by Lemonick, Pipkin, and Hamilton.[2] It had, however, at this stage been modified to a flop-out apparatus for use in the measurement of the spin of As^{76}.[3] The flop-out nature of the machine was maintained in the present experiment since certain features of it seemed appropriate for experiments with short-lived isotopes. Conversion to flop-out consisted of changing the stop system and detector so that the detector collected only atoms which passed through a circular region of $\frac{1}{4}$ in. diameter centered on the machine axis and located at the detector position. The stop system was such that only unflopped atoms could do this. When hyperfine transitions were induced in the C field the focussed beam was reduced and one observed a flop-out resonance. In this arrangement the detector consisted of a $1\frac{3}{4}$-in. diameter disk divided into as many as eight sectors. The disk was placed at the detector position (but with its center somewhat off the beam axis) by insertion through an air lock; an external indexing system was supplied so that each of the sectors in turn could be rotated to the proper position to accept the beam.

The operation of the system proceeded as follows: each of the sectors in use (most commonly five in number) was rotated into the beam-accepting position and received an accurately timed interval of exposure (usually 20 or 30 seconds) after which the next sector was rotated into position and exposed. A number of complete revolutions of the detector disk were made in this fashion until the total exposure on the sectors was found to be sufficient.[4] At the completion of a disk's

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[1] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953). More detailed references to specific work on these isotopes are also given in subsequent sections of the present paper.

[2] Lemonick, Pipkin, and Hamilton. Rev. Sci. Instr. **26**, 1112 (1955).

[3] Christensen, Bennewitz, Hamilton, Reynolds, and Stroke, Phys. Rev. **107**, 633 (1957).

[4] The total exposure time was usually about one mean life of the

exposure, it was retracted from the vacuum system. The sectors were separated from one another, sealed in a matrix of moisture-absorbing paper with "Scotch" tape, and mounted on holders for insertion into the counters.

The advantages of the flop-out, rotating-disk procedure in dealing with short-lived isotopes are: (1) The use of the small collecting area makes it possible to obtain up to 8 different exposures without breaking vacuum whereas the old system, in which the collector had to cover a $1\frac{1}{2}$-in. diameter annulus, required insertion of a probe (with attendant delay required for pumping out air locks, etc.) for each exposure. This reduction in "turn around" time made possible the accumulation of data in a time sufficiently small compared to the half-lives involved. (2) The rotation procedure had an averaging effect which reduced the difficulty associated with any inconstancy of the evaporated beam. This second point was particularly helpful in the case of the Ag isotopes. Here it was found that satisfactory data could be obtained despite the fact that with constant oven temperature the beam intensity dropped monotonically at a rapid rate. This decrease was in addition to that resulting from radioactive decay of the material in the oven; it was presumably associated with driving Ag atoms out of the carrier Pd. The old flop-in procedure would have been difficult, if not impossible, to use with the Ag isotopes.

The molybdenum oven was usually heated to about 1550°C by electron bombardment (about $\frac{1}{2}$ amp at 2500 v). The detector surface was cooled by liquid nitrogen as had been required for arsenic, although this surface cooling was not known to be necessary in these experiments.

Two counting setups were used. One of these was mainly β sensitive and consisted of a 1-mm thick organic phosphor viewed by a photomultiplier tube with which were associated conventional circuits. The other setup, which was γ sensitive and discriminated somewhat against β rays, used a NaI(Tl) scintillator.

To make maximum use of the activity on all sectors, each one was counted in turn for 2 minutes with a $\frac{1}{2}$-minute "turn-around" interval. The time of starting each interval was accurately noted for precision in making half-life corrections. Two counting cycles took about one half-life of the shorter-lived activities.

Analysis of the data was accomplished as follows for a five-sector cycle (two sectors with rf applied, spaced among three having no rf): There results a set of five counting rates, $C_1, C_2, \cdots C_5$, for sectors 1, 2, $\cdots$ 5, respectively, taken at times spaced quite accurately by equal intervals. Factors which would cause $C_1, \cdots C_5$ to differ from one another are (1) radioactive decay during counting, (2) variations in beam intensity due to changing oven conditions, and (3) the radio-frequency diminution of beam intensity which corresponds to the existence of a resonance. The three no-rf counting rates, C_1, C_3, and C_5, determine a quadratically time-dependent counting rate, Σ. The values of this function at the times of counting sectors 2 and 4, Σ_2 and Σ_4, respectively, are taken as being the beam background with no rf present. Σ_2 and Σ_4 are given by

$$\Sigma_2=0.375C_1+0.75C_3-0.125C_5$$

and

$$\Sigma_4=-0.125C_1+0.75C_3+0.375C_5.$$

The rf effect on sector 2 is then characterized by $\Delta_2=\Sigma_2-C_2$ and the fractional "flop" by Δ_2/Σ_2, with similar expressions for sector 4. The validity of this procedure was verified by running several disks for which all five sectors had no rf applied. The average Δ/Σ for several such disks was 0.006 ± 0.005. When sectors 2 or 4 had rf applied at a resonant frequency, Δ/Σ ranged from 0.08 to 0.15 with a statistical probable error of 0.01 or less. Typical counting rates per sector (for five sectors exposed to the beam a total time of four or five minutes each) were 5000 to 30 000 counts per two-minute interval, at the start of counting.

It was calculated that the quadratic correction procedure described above corrected for radioactive decay effects during counting (effect [1]) to better than 0.25%. The negligible value of Δ/Σ for the "no rf" test disks is taken to mean that any random fluctuatious in the beam were adequately smoothed out by the rotating disk method. In the case of the Ag isotopes, the amount of material coming from the oven was observed to decrease monotonically during a 20-minute run. Various considerations, particularly the low value of Δ/Σ on the test disks which had no rf on any sector, show that the quadratic correction procedure also takes this oven effect into account satisfactorily.

RESULTS

The approximately 25-min isotope resulting from bombardment of Ni was observed to have spin 2. The assignment of this activity to Cu^{60} seems quite certain[1,5,6] so that one may conclude $I=2$ for Cu^{60}. The resonance was observed at four values of the static field from 1.6 to 9.6 gauss.

The 3.3-hour Cu^{61} was found to have $I=\frac{3}{2}$. This resonance was observed twice at 6.9 gauss and once at 9.6 gauss. The result is in agreement with that of Nierenberg, Shugart, and Silsbee.[7]

The short-lived activity in Ag (hereafter referred to as the "26-min" activity; as previously mentioned,

activity involved. This is based on the fact that the optimum exposure time (that which gives maximum radioactivity on detector disk at completion of exposure) for a single disk placed in a beam of atoms is one mean life. This assumes that decrease of beam intensity is due only to radioactive decay of the material in the oven.

[5] van Lieshout, Nussbaum, Nijgh, and Wapstra, Phys. Rev. **93**, 255 (1954).

[6] Nussbaum, van Lieshout, Wapstra, Verster, Ter Haaf, Nijgh, and Ornstein, Physica **20**, 555 (1954).

[7] Nierenberg, Shugart, and Silsbee, Bull. Am. Phys. Soc. Ser. II **2**, 200 (1957).

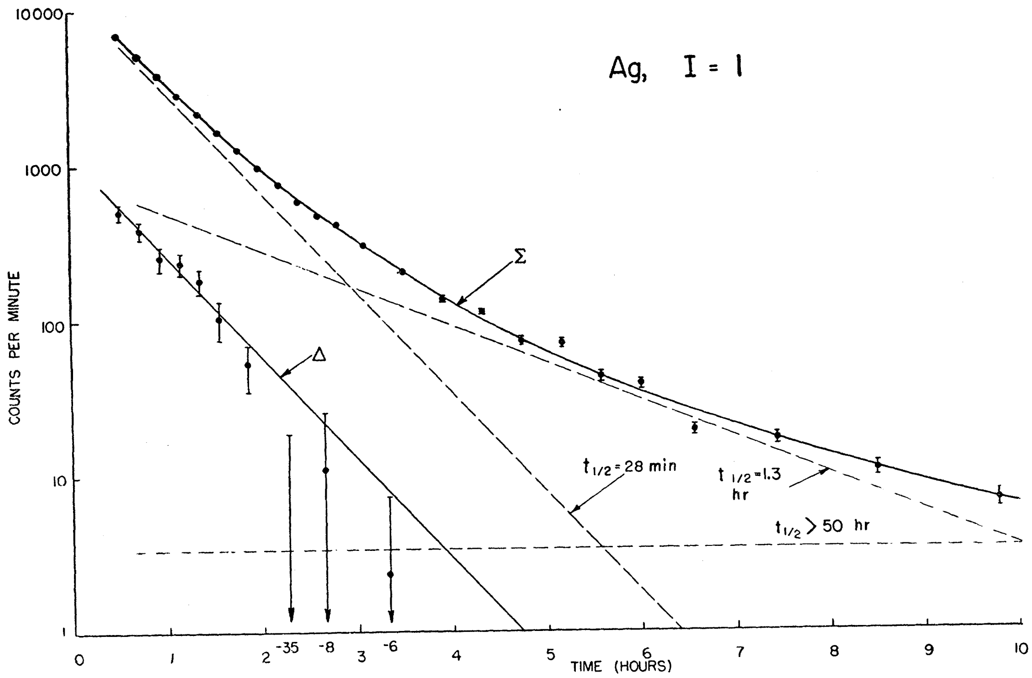

FIG. 1. Decay of unflopped background, Σ, and the resonance depth, Δ, for the spin-1 resonance of the silver isotopes. ½-hour bombardment of Pd by 18.8-Mev protons. β counter used as detector. Dashed lines give decomposition of Σ and Δ into components. Numbers −35, −8, −6 refer to lower bound of corresponding standard deviation flags.

this may be attributed to Ag^{104}, Ag^{106}, or both) was found to be primarily a β emission, while the 1.2-hour activity involves emission of a marked amount of penetrating radiation. As an aid in discrimination, observations of decay curves and resonances were made with both the β counter and γ-ray counter. No resonances were observed at frequencies corresponding to spins 0, 3, and 4—several attempts were made at each spin. Resonances were observed for spins of 1 and 2 as described in detail below.

Figure 1 shows data from a run at spin 1. The data were taken on the β counter which emphasizes the "26-min" activity and it is seen that a spin-1 resonance is present. Figure 1 is a plot of Δ and Σ as a function of time. It will be noted that Δ/Σ is about 0.09 ± 0.01 at the beginning and that Δ decays with about a 26-min half-life. The indication is that there is no spin-1 resonance in the 1.2-hr activity; however, the statistics in such a run are not good enough to assure this conclusion. Data taken in a different type of run clarify this point as shown in Fig. 2.

In this other run the cyclotron bombardment lasted 1 hour to emphasize the 1.2-hr activity. Here again the resonant frequency for spin 1 was applied when the rf sectors were exposed to the beam. The counting, however, was done in two stages: first in the β counter and then, after about 45 min, in the γ counter. The difference in sensitivity of the two counters to the two activities may be seen from Fig. 2. Analysis of the data indicates a change by a factor of roughly 9 in the relative efficiency of detection of the two activities when detectors are interchanged.

From Fig. 2 one sees that when counted in the β counter, the depth of the observed resonance is $12\frac{1}{2}\%$ and that Δ decays with a (27 ± 3)-min half-life.

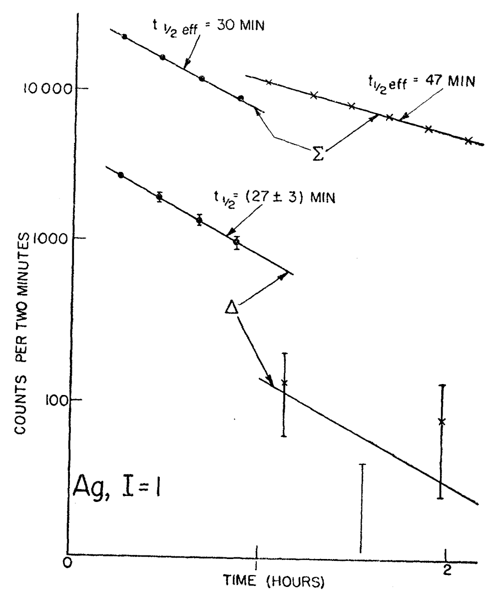

FIG. 2. Decay of Σ and Δ for spin-1 resonance in silver isotopes. One-hour bombardment. Initial and subsequent counting done with β and γ counters (circles and ×'s), respectively.

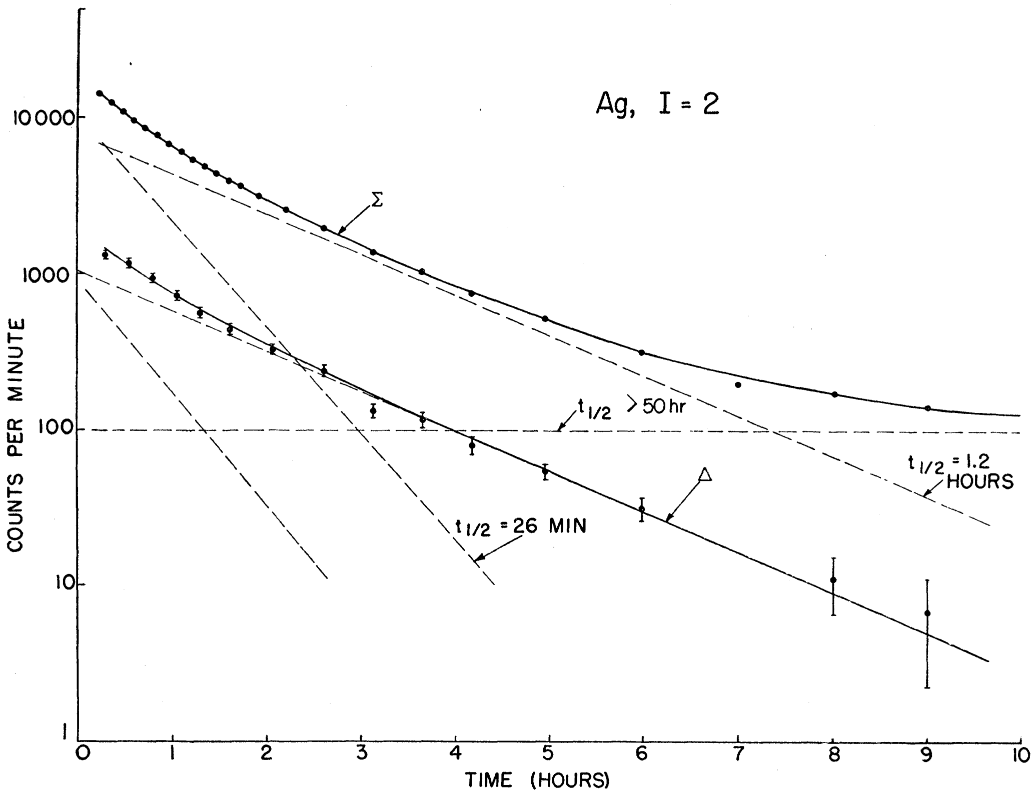

FIG. 3. Decay of Σ and Δ for spin 2 in silver isotopes. ½-hour bombardment. Counted in γ counter.

If a spin-1 resonance were present in the 1.2-hr activity one should still observe a resonance comparable to $12\frac{1}{2}\%$ after changing counters. One observes, however, that changing counters reduces Δ/Σ to about 0.01 or less. This is in fact about the amount one would expect the Δ/Σ for the "26-min" activity to be reduced as a result of the counter efficiency effect mentioned above. We conclude that $I=1$ for one component of the "26-min" activity and not for the 1.2-hr activity.

Figure 3 shows data taken using Ag isotopes, with the proper radio-frequency applied to give a resonance if $I=2$. In this case the cyclotron bombardment lasted for half an hour and the γ counter was used for detection. Analysis of the unflopped intensity (Σ) shows that initially the "26-min" and the 1.2-hour activities were each present in approximately equal amounts. The curve of flopped intensity (Δ) as a function of time in Fig. 3 shows clearly the presence of spin 2 for both of these activities. In addition Fig. 4 shows the results of a spin 2 run counted in the β counter. Here Δ only is plotted and owing to the low efficiency of the counter for the 1.2-hr activity Δ is seen to have only a 26-min half-life (within counting statistics).

In summary, the above results for Ag are: $I=2$ for the 1.2-hr activity and $I=1$ and 2 for the "26-min" activity. From the presence of two spins in the latter we assume that both 27-min Ag^{104} and 24.5-min Ag^{106} were present (in roughly equal amounts as could be estimated from the observed Δ for each spin). The question remains as to which spin goes with which isotope. An experiment with enriched Pd isotopes would have been useful for this. However, at the time of the experiment Pd enriched sufficiently in the correct isotopes was not available in the needed quantities.

Rather than trying to measure the half-life of the two resonances to a precision sufficient to allow discrimination between the two possibilities, recourse was had to an absorption measurement. The Ag^{106} β end point is 1.96±0.02 Mev[8] for the high-energy component and the Ag^{104} end point is 2.70±0.01 Mev[9] corresponding to effective ranges in Al of 950 and 1250 mg/cm², respectively.[10] Hence measurement was made of the fraction of the activity detected when 1, 2, 3, and 4 sheets of 0.025-in. Al (168 mg/cm²) were interposed between the activity and the β counter used for the previous data. To maximize counts obtained, and since relative energy discrimination rather than absolute energy measurement was desired, a "poor geometry" setup was used which involved minimum source-detector distance. Since many electrons traverse the absorber at glancing angles, the absorbers are effectively thicker than with good geometry.

[8] Bendel, Shore, Brown, and Becker, Phys. Rev. **90**, 888 (1953). This paper gives a rather complete bibliography on Ag^{106}.
[9] F. A. Johnson, Can. J. Phys. **33**, 841 (1955).
[10] W. Paul and H. Steinwedel, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. 1.

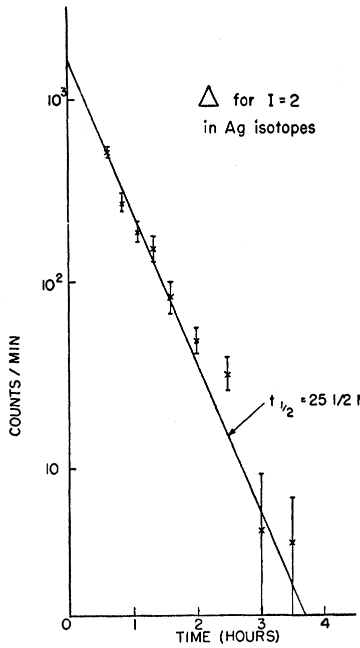

FIG. 4. Decay of Δ for spin 2 in silver isotopes. ½-hour bombardment. Counted in β counter.

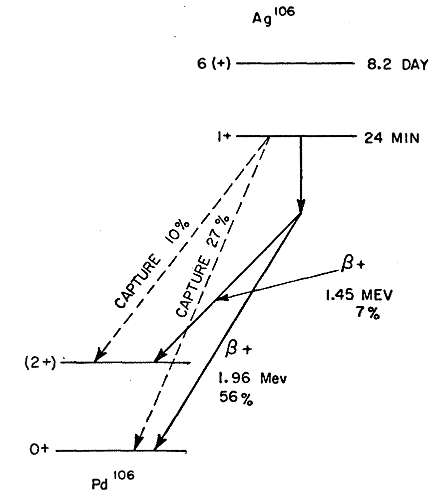

FIG. 6. Decay scheme for Ag^{106} taken from reference 8.

One must make an initial comment concerning any measurement of the absorption of the flopped activity in a flop-out experiment, since the directly measured absorption is that of radiation from unflopped atoms. But consideration of the absorption behavior of the flopped beam and the complementary unflopped beam leads easily to the conclusion that the diminution of Δ by an absorber is the same as would be observed for the diminution of a flop-in resonance signal by the same absorber.

In measuring in this way the relative absorption properties of the β radiations associated with the spin-1 and spin-2 resonances in the "26-min" activity, several complications arise. A relatively minor one is the fact that both of these positron activities have associated nuclear γ rays as well as annihilation γ's (see Figs. 5 and 6). These will give a tail on the absorption curve which is recognizable in itself, will not affect the initial slope of the absorption curve in any case, and will be small because the counter is insensitive to γ rays.

A major complication, on the other hand, is the fact that the $I=2$ resonance involves both the "26-min" and 1.2-hour activities which will have different absorption properties and which are present in relative abundances which vary with time. The relative abundances after passing through the beam and being counted on the β counter are known (see Fig. 3). In an experiment auxiliary to the measurement of the absorption of Δ, the absorption properties of the 1.2-hr activity by itself were deduced by preparing a source containing all of the activities ("26-min," 1.2-hr and a long-lived background) and following its absorption properties with time. From these data it was found that the fraction 0.42±0.03 of the 1.2-hr activity was counted through 1, 2, 3, and 4 Al absorbers. From the abundance data of Fig. 3 and the 1.2-hr absorption information one may now deduce, from the data on the absorption of the total Δ for $I=2$, the absorption of the Δ for the "26-min" $I=2$ resonance. During the first hour of counting from which, because of counting statistics, all relevant data came, three-quarters or more of the activity was in the "26-min" half-life. The correction to the spin-2 absorption to reduce it to pure "26-min" spin-2 absorption was, therefore, not great and in any event was carried out quite accurately as indicated above.

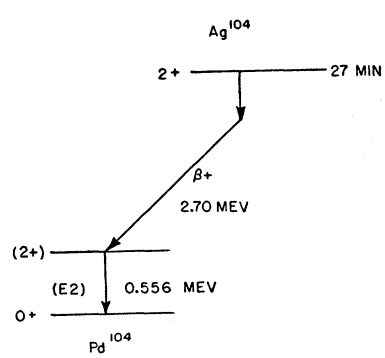

FIG. 5. Decay scheme for Ag^{104} taken from reference 9.

The net result of the absorption measurement is shown in Fig. 7 which is a plot of the fractions of the spin 1 and 2 "26-min" activity counted through 1, 2, 3, and 4 absorbers. Within statistics (which are not good enough to reveal the typical not-quite-exponential behavior[10]) spin 1 and spin 2 are found to absorb exponentially with respective absorption coefficients of 0.82±0.07 (absorber)$^{-1}$ and 1.16±0.07 (absorber)$^{-1}$. In conventional units these are, in the same order, 0.0049±0.0004 cm^2/mg and 0.0069±0.0004 cm^2/mg. These absorption coefficients are the weighted averages of the results with 0, 1, 2, and 3 absorbers; with relatively little counting time invested in four-absorber

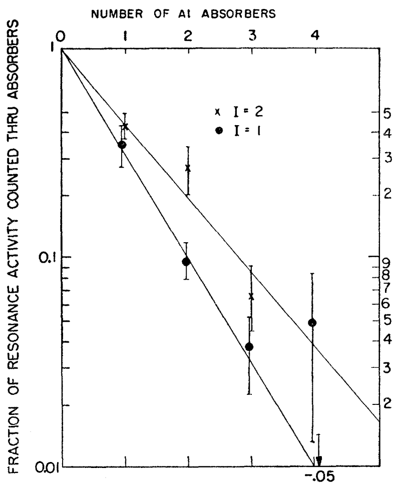

FIG. 7. Fraction of "26-min" resonances for spin 1 and spin 2 penetrating various thicknesses of Al absorbers in "poor geometry" as counted by β counter. The number "-0.05" at the bottom right indicates the ordinate of the 4 absorber, $I=2$ point.

transmission, this transmission is indistinguishable from zero for purposes of calculating an absorption coefficient (i.e., one of the transmissions has a negative value.) Because of the poor geometry these coefficients are, of course, more than one would obtain from published β absorption data.

The auxiliary absorption experiment referred to above showed that not more than $1\frac{1}{2}\%$ of the "26-min" activity counts through four absorbers. This $1\frac{1}{2}\%$ will be predominantly annihilation γ's, of which there will be equal amounts for both spin-1 and spin-2 26-min activities. There are nuclear γ rays in the 0.5-Mev range, but not more than one per positron (see Fig. 5). For any arbitrary assignment of these nuclear γ rays to spin 1 or spin 2, the penetrating γ background still divides so evenly between the two spins that the absorption curves of Fig. 7 are not changed appreciably by the corresponding correction.

We conclude therefore that spin 2 is associated with the 2.7-Mev β-emitting isotope, Ag^{104}, and that spin 1 is associated with the 1.96-Mev β emitter Ag^{106}.

DISCUSSION

If one were to assume the odd-proton state in Cu^{60} to be $2p_{\frac{3}{2}}$ and the odd-neutron state to be $1f_{\frac{5}{2}}$, the Nordheim "strong" rule would be violated by the observed spin 2.[11] On the other hand, the spin of 2 is consistent with Nordheim's "weak" rule if the odd neutron is assumed to be in a $2p_{\frac{3}{2}}$ state which therefore seems the more likely configuration assignment. Nussbaum *et al.*,[6] have studied the decay of 23.4-min Cu^{60} very thoroughly and have assigned spin-parity 2+ to this state in agreement with our result. These workers find that Cu^{60} undergoes β decay to the first excited state (2+) of Ni^{60} but not to the ground state (0+) and not to a higher lying (4+) state. They find $\log ft=7.4$ for the $(2+)\leftrightarrow(2+)$ transition and state that this high value for an allowed transition is probably due to "rearrangement forbiddenness."

Two approximately 25-min activities in Ag have been quite certainly identified. Johnson has shown that Ag^{104}, the daughter of 59-min Cd^{104}, has a half-life of 27 ± 1 min. Johnson also found that Ag^{104} decays principally by β^+ emission with a 2.7-Mev end point to an excited state of Pd^{104} as shown in Fig. 5. The Pd^{104} de-excites with emission of an 0.556-Mev γ ray. The log ft for the Ag^{104} decay is 5.4 (assuming no other modes of decay) and Johnson therefore assumes the transition to be allowed. If the first excited state of the even-even nucleus Pd^{104} has spin-parity of 2+ one may expect Ag^{104} to have spin 1, 2, or 3 and positive parity. In addition, since no β^+ transition is observed to the ground state of Pd^{104}, it is inferred that the 27-min Ag^{104} has spin 2 or 3 and positive parity in agreement with our result.

Ag^{106} has been produced by many workers, most recently by Bendel *et al.*[8] using the (γ,n) reaction on Ag^{107}. The half-life is 24 minutes. Figure 6 shows the level scheme. Decay occurs by β^+ emission as well as by electron capture to two states in Pd^{106}, the β^+ end points being 1.45 Mev and 1.96 Mev. Both of the β^+ decays were found to be allowed ($\log ft=4.90$ for decay to ground state, $\log ft=5.2$ for decay to excited state), from which one infers that the Ag^{106} ground state should have spin-parity of 1+ in agreement with our result.

The assignment of the approximately 1.2-hr half-life to a particular Ag isotope is not certain. Various authors have attributed the activity to Ag^{102}, Ag^{103}, or Ag^{104}. Most recently Haldar and Wiig[12] have definitely associated a 1.1-hr activity with Ag^{103}. This assignment was based partly on β-decay systematics and partly on identification of the daughter of the 1.1-hour decaying material as 17-day Pd^{103}. However, our result of $I=2$ for the approximately 1.2-hr activity rules out its being associated with an odd-A isotope; one must assume it to be another isotope than that studied by Haldar and Wiig. Since we find $I=2$ also for the 27-min Ag^{104}, an assignment of our approximately 1.2-hr activity to Ag^{104} would result in an isomeric state having the same spin as the ground state.

It is possible that the approximately 1.2-hr isotope is Ag^{102}; however Pd^{102}, the only isotope from which 18-Mev protons could produce Ag^{102}, is only 0.8% abundant. The yield of 1.2-hr activity is of the same order as that of Ag^{106} from the 27% abundant isotope Pd^{106}. An abnormally large cross section for $Pd^{102}(p,n)$-Ag^{102} would then be indicated.

ACKNOWLEDGMENTS

We wish to acknowledge the assistance with this experiment which we have received from Mr. Dieter Brill, Mr. Martin Posner, Mr. L. M. Jordan, and Mr. J. L. Snider.

[11] M. G. Mayer and J. H. D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley and Sons, Inc., New York, 1955), p. 196.

[12] B. C. Haldar and E. O. Wiig, Phys. Rev. **94**, 1713 (1954).