Decay of Ru^{105}, Pd^{109}, and Rb^{86}†

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A decay scheme for Ru^{105} has been proposed with levels at 130, 395 (or 530), 475, 685, 726, 795, 960, 1350, and 1720 kev. These levels are excited by seven beta groups of maximum energies of 1875, 1185, 1150, 1080, 915, 525, and 155 kev. Beta-gamma coincidence studies have confirmed a decay scheme for Pd^{109} having levels at 88, 217, 258, 307, 412, 719, and 860 kev, while existence of a 260- and a 400-kev beta group in addition to the 1030-kev beta ray has been established. The half-lives of Ru^{105}, Rh^{105}, and Pd^{109} have been determined as 4.43±0.02 hours, 35.88±0.02 hours, and 13.47±0.01 hours, respectively. The branching ratio of Rb^{86} has been experimentally measured as (8.79±0.09)%.

INTRODUCTION

IN the course of determination of absolute fission product cross sections in this laboratory, it became important to have more accurate and extensive decay scheme information on certain nuclides. Of particular importance were certain key isotopes, such as Ru^{105} and Pd^{109}, which are usually located near the valley region in the mass yield curves of heavy elements.

The decay of Ru^{105} has been studied by several investigators[1–3] in the last ten years. These studies led to a simple decay scheme which had a 1150-kev beta ray terminating in an excited level 856 kev above the ground state. This state de-excited by the emission of a 726-kev gamma ray to a metastable level at 130 kev, which in turn decayed with a 30-second half-life to the 35-hour ground state of Rh^{105}. Preliminary measurements in this laboratory showed the 726-kev gamma transition to be complex. Since this work was started, Heath and Vegors[4] have also shown that the gamma-ray spectrum of Ru^{105} contained at least seven peaks in addition to the previously reported 130- and 726-kev transitions.

Recently, several investigators[5–7] have worked on the decay scheme of Pd^{109} and have demonstrated that it was not the reported[8,9] simple 1030-kev beta ray leading to an 88-kev metastable state of Ag^{109}. For the most part, however, these studies have been concerned only with gamma analyses and very little accurate beta-gamma coincidence data has been taken.

The study of Rb^{86} was undertaken to confirm the branching ratio of (8.7±0.2)% reported by Campion and Taylor[10] since several other workers had previously given (8.5±0.5)%,[11] (8.9±0.5)%,[12] and 8.4%.[13]

SOURCE PREPARATION AND APPARATUS

Ruthenium metal which had been isotopically enriched to 98.2% Ru^{104} was obtained from the Oak Ridge National Laboratory for use in the irradiations. The major isotopic impurity was Ru^{102}, in an abundance of 1.3%. Ru^{99-101} accounted for the remaining 0.5%, and therefore, less than 0.01% of the prepared Ru^{105} activity was due to impurities. Samples weighing 1 mg. were irradiated for 11 hours at a thermal neutron flux of $2\times10^{12}/cm^2$ sec in the CP-5 reactor at the Argonne National Laboratory. The ruthenium was dissolved in aqueous KClO and then purified by distillation of the tetroxide from a perchloric acid solution.[14] The tetroxide was collected in 6*N* NaOH and reduced with ethanol; the resulting precipitate was dissolved in a minimum volume of 6*N* HCl. Samples for gamma studies were placed in glass test tubes, while the samples (5–10 μg) used for beta studies were deposited on thin Zapon films (7–25 $\mu g/cm^2$).

The purity of the enriched Pd^{108} obtained from the Oak Ridge National Laboratory was 94.7%, the major impurities being Pd^{106} and Pd^{110} (3.2% and 1.1%, respectively). $Pd^{104-105}$ constituted the remaining 1%. Since the half lives of the Pd^{107} and Pd^{111} formed by neutron capture of the impurities are 7×10^6 years and 22 minutes, respectively, their presence is not significant. To insure against the presence of unknown impurities, however, the decay of all peaks in the gamma spectrum was checked. Again, 1-mg samples of Pd^{108} were bombarded with a thermal neutron flux of $3\times10^{13}/cm^2$ sec for four hours in the CP-5 facility at the Argonne National Laboratory. The palladium was dissolved in HCl and H_2O_2 and the resulting solution was purified

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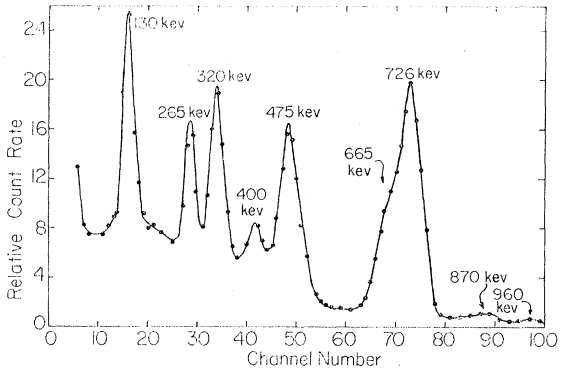

FIG. 1. Low-energy gamma spectrum of Ru^{105}.

by an ion exchange technique described by Buchanan *et al.*[15]

Enriched $Rb^{85}Cl$ was obtained from the Oak Ridge National Laboratory. Isotopic analysis showed 99.69% Rb^{85} while the major chemical impurity was Cs^{133}. Rubidium chloride was converted to the nitrate by prolonged boiling with concentrated HNO_3 until no traces of chloride were present. Samples containing 1 mg Rb^{85} were irradiated at the Argonne National Laboratory for twenty hours in a thermal neutron flux of $3\times10^{13}/cm^2$ sec. The $Rb^{86}NO_3$ was dissolved in water and separated from any Cs^{134} activity by purification on a zirconium tungstate ion exchange column as described by Crouch *et al.*[16] Ammonium chloride in the eluent was destroyed by boiling with aqua regia. Aliquots for the branching ratio determinations were taken from this solution. Samples were placed on aluminized Zapon films and the total beta disintegration rate was determined using a calibrated[17] $4\pi\ \beta$ proportional counter.

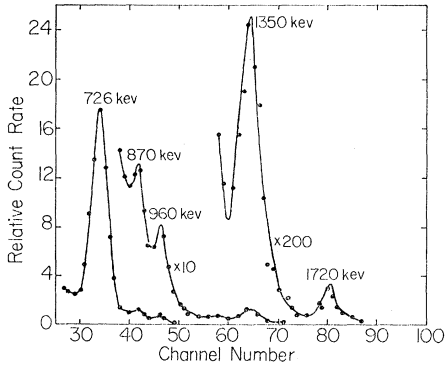

FIG. 2. High-energy gamma spectrum of Ru^{105}.

Rb^{86} samples were gamma counted in a calibrated 5-in. NaI(Tl) crystal[18] while gamma spectral studies on the other isotopes were made with two 3-in.×3-in. NaI(Tl) crystals. Both of the latter spectrometers had a resolution of 7.5% (full width at half maximum counting rate) for the 662-kev gamma ray of Cs^{137}.

The beta spectra were obtained using a hollow-well plastic scintillation crystal designed after Bell[19] and Gardner and Meinke.[20] The crystal used in these studies had a resolution of 15% for the 624-kev conversion electrons of Cs^{137}.

Spectra were displayed on multichannel analyzers. In all coincidence studies, the analyzers were used as the coincidence gate, activated by an output pulse from a single-channel analyzer. The spectrum pulse was delayed an amount equal to the time lag in the single-channel analyzer and the coincident spectrum displayed in the normal manner.

EXPERIMENTAL RESULTS

Ru^{105}

In a typical half-life determination of Ru^{105} and Rh^{105}, the decay was followed from 10^5 counts per minute until it could be shown that the activity due to 39.8-day Ru^{103} was less than 25 counts per minute. This is taken as a further confirmation that the total activity not due to Ru^{105} was on the order of 0.01%. A total of six samples from different irradiations gave 4.43±0.02 hours for the half-life of Ru^{105} and 35.88±0.02 hours for the half-life of Rh^{105}.

TABLE I. Gamma-ray relative intensities for Ru^{105}.

Energy (kev)	Relative intensity	Energy (kev)	Relative intensity
130	11.6±0.2	875	4.7±0.3
265	14.0±0.8	960	3.1±0.2
320	21.3±1.1	1350	0.6±0.04
400	12.2±1.3	1720	0.1±0.002
475	30.4±2.3[a]	485	4.4[b]
665	38.2±2.5	210	(≤1)
726	100	...	...

[a] Corrected for the 485-kev component.
[b] Calculated from the decay scheme and other relative intensities.

The Ru^{105} gamma spectrum up to 1 Mev is shown in Fig. 1. This spectrum was taken immediately after distillation of the tetroxide in order to minimize the grow-in contribution of Rh^{105}. Separate peaks can be observed at 130, 265, 320, 400, 475, 665, 726, 875, and 960 kev. Figure 2 shows the gamma spectrum from 600 kev to 2200 kev in which additional gamma rays at 1350 and 1720 kev are observed. The peak at 475 kev was later found to be complex, consisting of two gamma rays of

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475- and 485-kev energy. Gamma-gamma coincidence studies also confirmed the presence of a 210-kev transition. Relative intensities of these gamma rays are given in Table I. After waiting several days, the residual spectrum showed only the presence of Rh^{105} and Ru^{103} by the 320- and 498-kev gamma rays respectively. This also gave a check of possible source contamination which agreed with those previously mentioned.

Part of Fig. 3 is the coincidence spectrum obtained gating on the 320-kev gamma ray; it indicates a strong coincidence with the 475-kev transition. The slight coincidences at 265 and 320 kev are due to coincidences of Compton-scattered gamma rays from the 400- and 475-kev transitions. Similarly, Fig. 3 also indicates those gamma rays coincident with the 265-kev gamma, demonstrating one strong coincidence at 400 kev. The other peaks are due to coincidences with Compton-scattered gamma rays of 475 and 320 kev, and chance coincidences with 400-kev gammas.

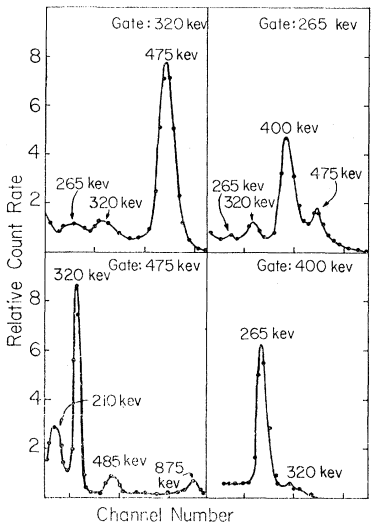

FIG. 3. Gamma-gamma coincidence spectra of Ru^{105}.

The third part of Fig. 3 gives the coincidence spectrum associated with the gamma peak at 475 kev. There are strong coincidences at 210, 320, 485, and 875 kev. Since a coincidence study using the 875-kev gamma as the gating peak showed the presence of only a 475-kev gamma ray, it has been inferred that these four gamma rays feed the 475-kev level in parallel. There was also a possibility that the 210-kev gamma ray feeds the 485-kev transition, but this was ruled out because of the intensity of the 210-kev peak. Finally, Fig. 3 shows the coincidence spectrum gated by the 400-kev gamma ray. The main coincidence is with a 265-kev gamma ray, the small peak at 320 kev being caused by chance coincidences with 475-kev gammas due to the window width used on the single-channel analyzer.

Figure 4 is a Kurie plot of the beta spectrum of Ru^{105} demonstrating a high-energy component whose maximum energy is 1870 kev, and present to an abundance

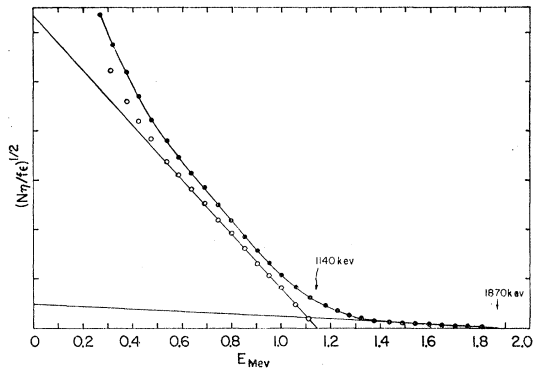

FIG. 4. Kurie plot of the beta spectrum of Ru^{105}.

of $(10.6\pm0.2)\%$. The main portion has a maximum energy of 1140 ± 10 kev.

Figure 5 shows the Kurie plots of beta spectra coincident with gamma rays of 265, 320, 726, and 875–960 kev. Gamma-beta coincidence studies were also made on the other gammas and similar results were obtained. It can be noted that the 265- and 320-kev gamma rays are coincident with a 1080-kev beta and the 726-kev transition is initiated by a 1150-kev beta ray. The 875- and 960-kev gamma rays are associated with beta rays of 525 and 915 kev, respectively. This is consistent with a mass difference of 1875 kev. The difference in end-point energies of the 1080- and 1150- kev beta rays was quite reproducible and there is little doubt that two different beta groups are involved.

DISCUSSION

From the gross gamma spectrum and the gamma-gamma coincidence studies, a tentative level scheme can be proposed. The strong 320–475 kev coincidence suggests a level at 795 kev which is confirmed by the 400–265 kev transition which terminates at the 130-kev metastable level of Rh^{105}. Furthermore, the 400–265 kev transition also suggests that the 665-kev gamma ray is a crossover to the metastable state from the 795-kev level. Unfortunately, it has not yet been possible to determine the order of emission of the 400–265 kev gamma rays, so the energy of the intermediate level

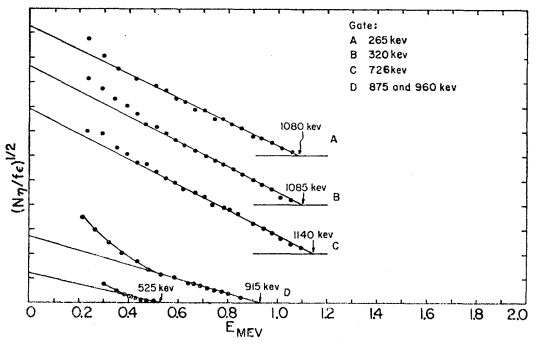

FIG. 5. Kurie plots of gamma-beta coincidence spectra for Ru^{105}.

TABLE II. Absolute beta intensities and log ft values for Ru^{105}.

Energy (kev)	Beta transition probability (%)	Log ft
1875	10.6	7.7
1150	48.2	6.1
1080	35.0	6.1
915	3.6	6.9
525	2.5	6.0
155	0.1	5.4
1185	≤ 0.5	~ 8.2

is not certain. Coincidence spectra also imply that the 485- and 875-kev gamma rays are parallel paths leading to the 475-kev gamma which suggests levels at 475, 960, and 1350 kev. These levels are confirmed by the 960- and 1350-kev crossover transitions. Finally, it is assumed that the 1720- and 726-kev gamma rays feed the ground state directly. These results suggest a scheme with the following levels: 130, 395 (or 530), 475, 726, 795, 960, 1350, and 1720 kev. There is also the possibility of a level at 685 kev due to the 210-kev transition terminating at the 475-kev level.

The 1870-kev beta obtained from the gross beta spectrum is assumed to go directly to the ground state of Rh^{105} since this beta is not in coincidence with any observable gamma ray. This is confirmed by the 1150–726 kev beta-gamma coincidence, which results in a mass difference of approximately 1875 kev. Since the 265- and 320-kev gamma rays both show coincidences with a 1080-kev beta, the assignment of a level at 795 kev and all the associated transitions is confirmed. Similarly, the 915-kev beta excites the 960-kev level, and the 525-kev beta transition terminates at the 1350-kev state. No coincidences were obtained with the 1720-kev gamma ray due to its low abundance, nor could any be obtained with the 210-kev gamma, since it could not be observed in the gross gamma spectrum.

The absolute intensities and log ft values for these beta rays as calculated from the level scheme, beta spectrum, and gamma relative intensities are given in Table II.

These results lead to the decay scheme in Fig. 6. This decay scheme as shown is similar to that of Saraf *et al.*[21] except for the additional level at 685 kev. This same level has also been reported by Ricci *et al.*[22] From the log ft values and the spectral shape it can be deduced that the 1875-kev beta transition is almost certainly allowed (l-forbidden). Since the ground and first excited state of Rh^{105} are known to be $\frac{7}{2}+$ and $\frac{1}{2}-$, respectively[23] (indicated by underscoring in the figures), the ground state of Ru^{105} must be $\frac{5}{2}+$ to satisfy the beta systematics. Tentative spin and parity assignments can be made for most of the other levels. The 395-kev state is probably $\frac{3}{2}-$ as it decays only to the $\frac{1}{2}-$ level. From log ft values, the 725-kev level is either $\frac{7}{2}+$ or $\frac{5}{2}+$. However, since this level does not decay to any negative-parity levels, $\frac{7}{2}+$ is most reasonable. The 795-kev level may be a $\frac{5}{2}-$ state, since it would then show the proper decay characteristics to be associated with a $K=\frac{1}{2}$ rotational band. This also makes the $\frac{3}{2}-$ assignment for the 395-kev level quite consistent. It should also be mentioned that these assignments are consistent with level schemes of several neighboring isotopes (Ru^{103}, Pd^{103}, Pd^{109}, Ag^{107}).

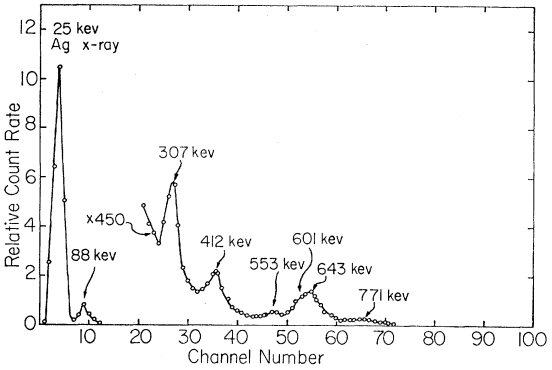

FIG. 7. Gamma spectrum of Pd^{109}.

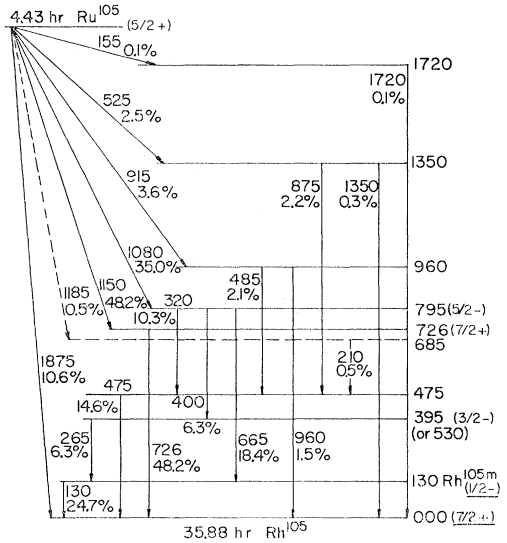

FIG. 6. Proposed decay scheme of Ru^{105}.

EXPERIMENTAL RESULTS

Pd^{109}

Half-life determinations were made on six purified samples from different irradiations and they followed a 13.47- $\pm$0.01-hr decay. The half-life was followed from approximately 10^5 counts per minute down to less than

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TABLE III Gamma-ray relative intensities for Pd^{109}.

Relative (kev)	Relative intensity	Energy (kev)	Relative intensity
88	6×10^4	553	5
307	100	601	30
412	100	643	100
448	5	772	40

10 counts per minute as a further check on source purity. Figure 7 shows the gamma spectrum associated with Pd^{109}, and Table III lists the observed gamma rays and their relative intensities. Two additional gamma rays (41 and 129 kev) were observed from coincidence data, and they will be discussed later. Again, the half-life of these peaks was followed to insure that they belonged to Pd^{107}.

Figure 8 summarizes the gamma-gamma coincidence studies in which it can be seen that the 307-kev gamma is in strong coincidence with both the 412- and 553-kev gamma rays. The slight peak at 307 kev is due to coincidences with the Compton distribution of the 412-kev gamma. Similarly, the 412-kev gamma ray is coincident with the 307- and 448-kev gammas, the small peak at 412 kev being due to chance coincidences.

No coincidences were observed for the 772-kev gamma ray, but it can be seen from Fig. 8 that the 601-kev gamma is coincident with both 41- and 129-kev gamma rays, while the 643-kev gamma shows coincidences with only the 129-kev gamma ray. The prominent peak at 88 kev in the 601-kev coincidence spectrum is due to chance coincidences with that most abundant gamma ray. Since all coincidence spectra showed this effect, it has been omitted in the other diagrams for clarity.

A Kurie plot of the gross beta spectrum is given in Fig. 9. The spectrum has an allowed shape with a maximum energy of 1028 ± 2 kev.

Kurie plots of the gamma-beta coincidence studies are shown in Fig. 10. There is only slight indication of the expected coincidences, because of large chance coincidences with the main beta group (1028 kev), would tend to obscure any other results. To be certain that the lower energy coincidences obtained were meaningful, a "control" experiment was run on the 307- and 412-kev gamma rays. From the level scheme already devised,[6,7] these gammas should show coincidences with beta groups having maximum energies of 260 and 400 kev. It can be seen that this is approximately the case, that both groups can be somewhat resolved from each other, and that they can be observed in the presence of the large chance spectrum. Similarly, from Fig. 10 it can be seen that the 772-kev gamma ray is coincident with a 260-kev beta group, as would also be expected from the level scheme. When the 601–643 kev gamma group was studied, only one beta ray was observed in

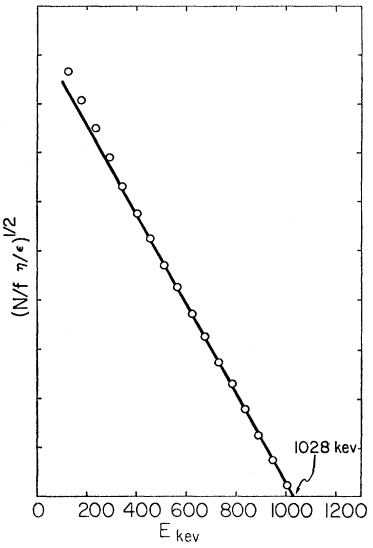

FIG. 9. Kurie plot of the beta spectrum of Pd^{109}.

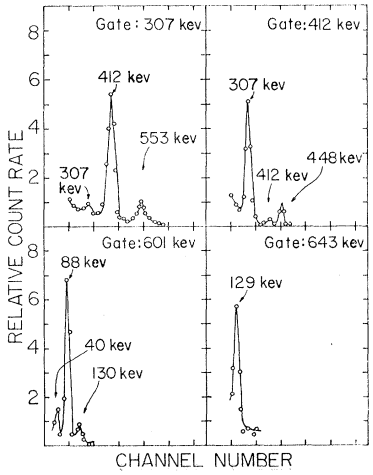

FIG. 8. Gamma-gamma coincidence spectra of Pd^{109}.

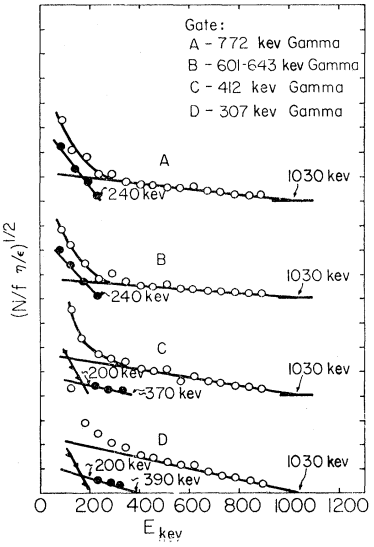

FIG. 10. Kurie plots of gamma-beta coincidence spectra for Pd^{109}.

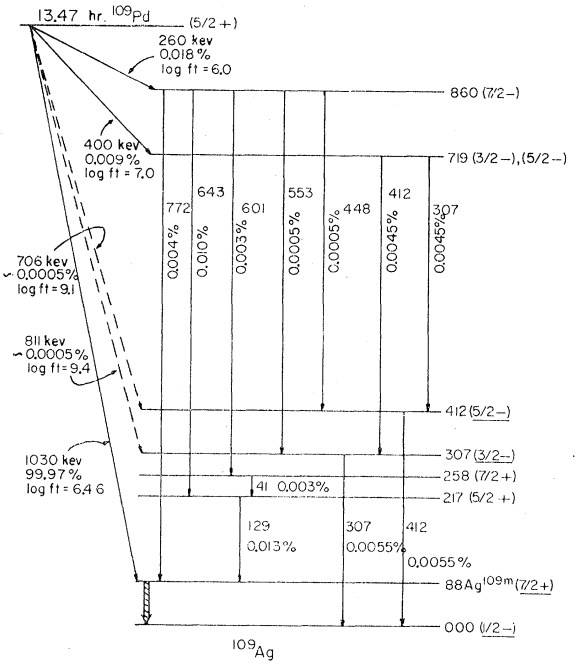

FIG. 11. Proposed decay scheme of Pd^{109}.

coincidence, that being the 260-kev beta leading to the 860-kev level.

DISCUSSION

From these results, a decay scheme can be proposed with levels definitely existing at 88, 307, 412, 719, and 860 kev. There was uncertainty in the arrangement of the 643–129 and 601–41–129 kev sequences. It is clear that the 643- and 601–41 kev steps are parallel paths either originating at 860 kev and leading to an excited state which decays with the emission of a 129-kev gamma ray to the 88-kev level, or alternatively, being fed by a 129-kev gamma ray (which originates at the 860-kev level) and then decaying to the metastable state. In the first of these possibilities, the 601–643 kev gamma group will show observable coincidences with only a 260-kev beta ray, while in the second case, coincidences should be observed with both a 260- and a 380-kev beta group. The observed beta-gamma coincidence spectrum for the 601–643 kev group was shown to be simple, consisting of only one beta ray whose maximum energy is about 240 kev, consequently confirming the first alternative. The decay scheme which is derived from this information is shown in Fig. 11. The 706- and 811-kev beta rays were not observed directly but were deduced from other similar decay schemes. The abundances indicated as approximate were chosen so as to obtain log ft values consistent with spin and parity assignments. Table IV summarizes the absolute intensities and log ft values for these beta transitions. The only point remaining in doubt is the placement of the 601–41 kev gamma group which has not been resolved. The 707-kev gamma ray reported by Starner[7] was not observed in this study.

TABLE IV. Absolute beta intensities and log ft values for Pd^{109}.

Energy (kev)	Beta transition probability (%)	Log ft
1030	99.97	6.46
811	0.0005	9.4
706	0.0005	9.1
400	0.009	7.0
260	0.018	6.0

The ground state of Pd^{109} is probably $\frac{5}{2}+$ as is predicted from both the de-excitation of[7,24] Pd^{109m} and beta systematics (log $ft_{1028}=6.46$, $\Delta I=1$, No). The spins and pairities of the low-lying levels in Ag^{109} have been taken from published data: the ground state has been experimentally measured as $\frac{1}{2}-$,[25] the 88-kev transition has been shown to be $E3$[5]; consequently the 88-kev level is $\frac{7}{2}+$. The 309- and 412-kev levels were obtained from Coulombic excitation studies[26] which indicated $\frac{3}{2}-$ and $\frac{5}{2}-$ states, respectively.

The 860-kev level is excited by a beta ray with log $ft=6.0$; therefore $\Delta I=0$, or 1, yes. Since the ground state of Pd^{109} is $\frac{5}{2}+$, there is a choice between $\frac{3}{2}$, $\frac{5}{2}$, or $\frac{7}{2}-$ for this level. As this state decays to $\frac{3}{2}-$ and $\frac{5}{2}-$ levels but not to the $\frac{1}{2}-$ ground state, a high negative value is expected, probably $\frac{7}{2}-$. The 719-kev level is in a similar situation so that either $\frac{3}{2}$ or $\frac{5}{2}-$ cannot be excluded. Similarly, the 217- and 258-kev states are fed by a level of high spin and negative parity $(\frac{7}{2}-)$, and they do not decay to the $\frac{1}{2}-$ ground state, so assignments of $\frac{5}{2}$ and $\frac{7}{2}+$ are possible for either level. Because the 860-kev $(\frac{7}{2}-)$ level decays to the 258-kev state, the most probable assignment for the latter is $\frac{7}{2}+$. Therefore, the assignment for the 217-kev level is $\frac{5}{2}+$.

EXPERIMENTAL RESULTS

Rb^{86}

The branching ratio was determined as $(8.79\pm0.09)\%$ on the basis of five independent determinations. It can be seen that this is in agreement with the other reported values but somewhat closer to and more accurate than the previously published data of Campion and Taylor.[10]

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