The Decay of Rh¹⁰⁶[†]

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Beta-rays with maximum energies of 3.53 ± 0.01 , 3.1 ± 0.1 , 2.44 ± 0.07 , and 2.0 ± 0.1 Mev and relative intensities of 68, 11, 12, and 3 percent, respectively, have been found with a lens spectrometer in the decay of Rh¹⁰⁶. Internal conversion lines of 0.513 and 0.624 Mev transitions occur with intensities of 7.5×10^{-4} and 2.3×10^{-4} relative to total beta-emission. The first of these has a K/L ratio of 7.8 ± 1 , indicating an electric quadrupole radiation. Lens and scintillation spectrometer measurements show that the gamma-ray spectrum consists of lines at 0.511, 0.621, 0.87, 1.045, 1.55, and 2.41 Mev with relative intensities of 100:53:3:8:2.5:1, respectively. The results are consistent with beta-decay to states in Pd¹⁰⁶ at 0, 0.513, 1.137, 1.55, and 2.42 Mev having even parities and spins of 0, 2, 0, 2, and 2, respectively. Rh¹⁰⁶ is assigned spin 1 and even parity. The possibility of explaining the gamma-gamma angular correlation of Rh¹⁰⁶ based on this decay scheme is mentioned.

I. INTRODUCTION

MEASUREMENT of the angular correlation of succession radiations as a means of determining certain properties of nuclear energy levels has been developed extensively during the past few years. Rh¹⁰⁶, a 30-sec beta-activity¹ which is generally used in an equilibrium condition with 1-yr Ru¹⁰⁶, was one of the early cases studied. Brady and Deutsch found² that the angular correlation of the gamma-rays emitted in the decay of Rh¹⁰⁶ exhibited a strong anisotropy while at the same time Deutsch and Metzger showed³ that an anisotropic effect existed in the direction-polarization correlation.

Prior to the experiments of Brady, Deutsch, and Metzger and some preliminary results reported⁴ by the author, the only complex decay scheme for Rh¹⁰⁶ was proposed⁵ by Peacock. He found two beta-ray groups with end points at 3.55 Mev (82 percent) and 2.30 Mev (18 percent) and gamma-rays of 0.51, 0.73, and 1.25 Mev by means of a lens spectrometer. The data were interpreted as beta-decay to the ground state of Pd¹⁰⁶ and to a level at 1.25 Mev, the latter being followed by either a 0.51–0.73-Mev gamma-ray cascade (17 percent) or a 1.25-Mev crossover (1 percent).

On the basis of this decay scheme Brady, Deutsch, and Metzger deduced the following from the correlation measurements: (1) the first excited state of Pd¹⁰⁶ has a spin $J \ge 2$; (2) the parities of the ground and second excited states are the same; and (3) if both cascade transitions are quadrupole, they are electric. They also showed that the general shape of the angular correlation most closely corresponds to a 0-2-0 cascade but that the magnitudes of the anisotropy coefficients are smaller than expected by a factor of two.

Ling and Falkoff examined⁶ this case theoretically and proved that no combination of quadrupole and dipole radiations could account for the angular correlation if only a single two-step cascade were involved. The analysis was extended⁷ to higher multipole orders by Spiers, but the correlation could still not be explained successfully without introducing additional cascade processes. Williams and Wiedenbeck suggested⁸ that the first excited state is 2+ and that two higher levels might be present, one being 0+ and the other $1\pm$ or 3+.

The first experimental evidence for greater complexity of the Rh¹⁰⁶ decay scheme was obtained⁹ by Goldhaber and der Mateosian who discovered that this activity could produce photoneutrons in Be and D_2O The fraction of gamma-rays above 2.23 Mev was estimated to be approximately 1 percent of the total gamma-ray intensity and appeared to be too great to be accounted for as bremsstrahlung from the energetic beta-rays.

With this new information and the unsatisfactory situation regarding the angular and direction-polarization correlations, it was considered desirable to reinvestigate the Rh¹⁰⁶ decay scheme. The results reported here show that altogether four excited states in Pd¹⁰⁶ are involved in the decay of Rh¹⁰⁶. In addition to a strong 0.624-0.513-Mev gamma-ray cascade several other cascade processes are present which might conceivably contribute sufficiently large enough effects to help explain the observed correlations.

II. BETA-RAY SPECTRUM

The beta-ray spectrum of Rh¹⁰⁶ was examined with a lens spectrometer set for a resolution of 2.5 percent. Sources of Ru¹⁰⁶ aged for approximately two years, were mounted on 0.09 mg/cm² Nylon and the deposits were estimated to be less than 0.5 mg/cm^2 thick.

[†] Under contract with the AEC. ¹ Way, Thano, Scott, and Thew, *Nuclear Data*, National Bureau of Standards Circular No. 499 (1950).

² E. L. Brady and M. Deutsch, Phys. Rev. 74, 1541 (1948); 78, 558 (1950).
⁸ M. Deutsch and F. Metzger, Phys. Rev. 74, 1542 (1948); 78,

^{551 (1950).}

⁴ D. E. Alburger, Phys. Rev. 85, 734 (1952). ⁵ W. C. Peacock, Phys. Rev. 72, 1049 (1947).

 ⁶ D. S. Ling and D. L. Falkoff, Phys. Rev. 76, 431 (1949).
⁷ J. A. Spiers, Phys. Rev. 78, 75 (1950).
⁸ A. H. Williams and M. L. Wiedenbeck, Phys. Rev. 78, 822 (1950).

⁹ M. Goldhaber and E. der Mateosian, Brookhaven National Laboratory Report BNL 51 (S-5) (1950) (unpublished).



FIG. 1. Kurie plot analysis of the Rh¹⁰⁶ beta-ray spectrum. An enlargement of the total curve is shown at the right.

Because of the indications of complexity found in preliminary runs, considerable care was exercised in taking and analyzing the data. The statistical accuracy was high, such that \sim 500,000 counts per point were recorded at the top of the curve, the points were interlaced in a random manner, and the results of a number of complete runs were averaged together in order to minimize effects of drift in the counting system. Corrections for counter dead-time and background were applied. In spite of the smooth appearance of the total curve data were taken at approximately 85 momentum settings. Calculation and analysis of the Kurie plot was carried out with the aid of a table of relativistically accurate Fermi functions kindly supplied by I. Feister of the National Bureau of Standards. An extrapolation of the table above 3.1 Mev was necessary in order to cover the full range of beta-ray energies.

Figure 1 shows a Kurie plot analysis of the Rh¹⁰⁶ beta-ray spectrum. Subtraction of the various components was done in the usual manner by least squares



superposed on the beta-spectrum.

fitting of a straight line to points near the end of each curve. In this way four beta-ray groups could be resolved consistently assuming that the allowed shape applied to each. Owing to the statistical scatter of the points below 2 Mev and the possible influence of source thickness effects, it was not considered advisable to extend the analysis further. The end-point energies of the various groups are 3.53 ± 0.01 , 3.1 ± 0.1 , 2.44 ± 0.07 , and 2.0 ± 0.1 Mev. The relative intensities of 68, 11, 12, and 3 percent, respectively, were determined by reconstructing the momentum distributions of each component and comparing their areas as measured with a planimeter. The errors in their intensity determinations are about 15 percent for the most energetic group and 25 percent for the inner groups. The 6 percent excess intensity at low energies is not considered significant in view of the errors involved and the number of subtractions made.

III. INTERNAL CONVERSION LINES

Internal conversion lines were found superposed on the Rh¹⁰⁶ beta-ray spectrum as shown in Fig. 2. The energies of the transitions are 513 and 624 kev in agreement with internal conversion lines also observed¹⁰ by Hill. The intensities of the K-lines relative to total beta-radiation are 7.5×10^{-4} and 2.3×10^{-4} , respectively. No evidence for other conversion lines was found in spite of the high statistical accuracy of the points.

Measurement of the K/L ratio of the 513-kev transition was carried out at a spectrometer resolution of 1.6 percent. Good statistics were necessary since the *L*-line was only 2 percent above the beta-ray yield at this point. When the beta-background is subtracted, the curve in Fig. 3 is obtained from which the K/L ratio is 7.8 \pm 1. Because of higher background and lower net yield a K/L measurement was not attempted on the 624-kev transition.

IV. GAMMA-RAY MEASUREMENTS

The lens spectrometer was used to examine the photoelectron spectrum due to the gamma-rays of Rh¹⁰⁶. The results obtained with a 25-mC source and converters of lead (22 mg/cm²) and uranium (44 mg/cm²) are shown in Figs. 4 and 5. Assignments of the lines are given in each figure. It may be noted that the lead converter permits the *L*-0.511 and *K*-0.621 lines to be separated, while the corresponding conversion lines from uranium have very nearly the same energy because of binding effects. On the other hand, the uranium allows clearer observation of the *K*-0.87 line and in addition shows the presence of a weak 1.55-Mev gamma-ray.

Examination of Peacock's lead conversion curve⁵ suggests that the L-0.511 and K-0.621 lines were not resolved and that this resulted in the misassignment of the L-0.621 peak as being the K conversion line of a 0.72 Mev gamma-ray.

¹⁰ R. D. Hill (private communication).

The energies of the various gamma-rays were determined from the positions of the K peaks and in some cases checks were made using the extrapolated high energy edge of the line. The energies thus obtained are 0.511, 0.621, 0.87, 1.045, and 1.55 Mev with errors of not more than 1 percent.

The relative intensities were determined from the momentum plots of K conversion lines by comparing areas and correcting for photoelectric cross section according to the empirical formula of Gray.¹¹ The applicability of Gray's formula to the source and converter geometry used in this work was studied by examining in a similar geometry the photoelectron peaks due to sources of Na²² and Na²⁴. The disintegration schemes of these isotopes are known and the range of energies covered is from 0.51 to 2.76 Mev. It is found that in the energy range of the lens measurements on Rh¹⁰⁶,



FIG. 3. K and L internal conversion lines of the 513-kev transition after subtraction of the beta-ray background.

0.51 to 1.55 Mev, Gray's formula may give results in error by as much as 50 percent at the extreme ends of this interval, while for gamma-rays between 0.5 and 1.0 Mev the errors are probably not more than 25 percent. The intensities of the Rh¹⁰⁶ gamma-rays relative to the 0.511-Mev transition, as given in Fig. 7, must be regarded as having errors which increase from about 20 percent for the 0.621-Mev line to 50 percent for the 1.55-Mev gamma-ray.

As a check on the above gamma-ray measurements a 100-mC Rh¹⁰⁶ source was examined recently using a uranium converter and slightly better resolution than previously. A curve closely similar to that of Fig. 5 was obtained except that the *L* lines of both the 0.87-and 1.55-Mev gamma-rays were visible. From this result it is more certain that all of the gamma-rays are

¹¹ L. H. Gray, Proc. Cambridge Phil. Soc. 27, 103 (1931).



due to Rh¹⁰⁶ and not to impurities. With this same source the region above 1.55 Mev was examined carefully because of the photoneutron evidence previously mentioned. No photoelectron peaks were found but there appeared to be a weak Compton electron group ending at about 2.2 Mev. In spite of this result it was not felt that the presence of an additional gamma-ray had been established definitely. The evidence for a gamma-ray of 2.9 Mev previously suspected⁴ was not confirmed.

In collaboration with Dr. W. F. Hornyak, a scintillation crystal spectrometer was put in operation using a 5819 photomultiplier tube together with a NaI(Tl) crystal 4 cm in diameter and 3 cm thick specially prepared by Dr. A. Schardt. The resolution of this device at 661-kev gamma-ray energy is 10.0 percent. The gamma-rays of Rh¹⁰⁶ previously measured with the lens spectrometer were all observed with this instrument. To examine the region above 1.55 Mev the gamma-rays from a 200-mC Rh¹⁰⁶ source were filtered through 5 cm of lead before reaching the NaI crystal. In this arrangement a high discrimination against the lower energy and more intense components was achieved. Without the lead filter the pile-up of small pulses prevented a clear examination at high energies with at the same time reasonable counting rates.

The spectrum obtained with a differential pulse-height analyser and covering energies between 1.0 and 3.2 Mev



FIG. 5. Rh¹⁰⁶ photoelectron spectrum using a U converter 44 mg/cm² thick.



FIG. 6. Pulse-height spectrum between 1.0 and 3.2 Mev from a NaI crystal due to Rh106 gamma-rays.

is shown in Fig. 6. The highest energy peak is interpreted as the photoline of a 2.41 ± 0.04 Mev gamma-ray, the energy being based on a Na²⁴ calibration. In the Na²⁴ spectrum peaks corresponding to pair production by the 2.76-Mev gamma-ray with the escape of one and two annihilation quanta were present in addition to the photopeak. The one-quantum escape peak was 0.16 as intense and the two-quantum escape peak 0.45 as intense as the photoline.

This information may be used to substantiate the interpretation of the Rh¹⁰⁶ curve in Fig. 6. The two quantum escape peak should be measurable but it is masked by the 1.55-Mev photoline, while the one quantum escape peak should barely be visible. Since no other higher lines seem to be present it would be difficult to interpret the 2.41-Mev line as an annihilation escape peak of a higher energy gamma-ray.

The relatively slow decrease in the yield beyond 2.4 Mev in Fig. 6 is attributed to pulse pile-up. When a Na²⁴ source was examined in the presence of strong fluxes of Cs137 and Co60 gamma-rays incident on the NaI crystal in order to simulate the situation existing in Rh¹⁰⁶ it was possible to obtain a slow decrease in yield beyond the 2.76-Mev photoline.

The relative intensities of the 2.41- and 1.55-Mev gamma-rays of Rh¹⁰⁶ were estimated using the Na²⁴ data on the 1.38- and 2.76-Mev gammas to correct for lead filtering and detection efficiency. It was found that the 2.41-Mev gamma-ray is about 0.4 as intense as the 1.55-Mev component or 1 percent of the 0.513-Mev line. This corresponds to an intensity of about 0.6 percent of all gamma-rays in rough agreement with the estimates of Goldhaber and der Mateosian based on a measurement of the photoneutron yield.9

DISCUSSION

A decay scheme for Rh¹⁰⁶ can be deduced which is consistent with the various measurements described above within their respective errors. This is shown in Fig. 7. All of the radiations were resolved except a beta-ray group, indicated by the dashed line, to a level which is thought to exist in Pd¹⁰⁶ at 2.42 Mev. Such a level would explain both the 0.87- and 2.41-Mev gammaray transitions, the latter being equal within the probable error to the more accurate sum 0.87 + 1.55 = 2.42Mev.

Confirmation of a considerable portion of the Pd¹⁰⁶ level scheme has been obtained¹² by Hayward who has studied the gamma-ray transitions occurring in the K-capture decay of Ag^{106} . He has proposed levels at 0.511 Mev (2+), 1.131 Mev (0+), 1.54 Mev (3+), and 1.76 Mev (4+). The 0.511-Mev transition is found to have a K/L ratio of 8.2 ± 1 in agreement with the present data on Rh¹⁰⁶. If the highest state at 1.76 Mev actually has a spin of 4, its failure to appear in Rh¹⁰⁶ decay would be accounted for by beta-ray selection rules and gamma-ray transition probabilities.

The spin and parity assignments in the present work are based mainly on the measurements described in the foregoing sections. The nature of the 0.513-Mev radiation can be determined from the analysis¹³ of Goldhaber



12 R. H. Hayward, Phys. Rev. 85, 760 (1952) and private communication to M. Goldhaber.

¹³ M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

and Sunyar by extrapolation of the empirical K/L ratio curves. Their data on electric transitions are given in Fig. 8 together with the point corresponding to the 0.513 Mev gamma-ray of Rh¹⁰⁶. It is evident that electric quadrupole radiation is involved, a conclusion which also fits the general rule formulated¹³ by Goldhaber and Sunyar that the first excited states of most even-even nuclei have a spin of 2 and even parity.

The ratio of K conversion coefficients of the 0.513 and 0.624 Mev transitions as obtained from the internal and photoelectric conversion measurements is 1.73. This agrees well with the value 1.70 from the tables¹⁴ of Rose et al. assuming both transitions are E2. Furthermore, if the decay scheme is accepted as being correct then the absolute K conversion coefficients of the 0.513and 0.624-Mev transitions may be calculated. These are $(3.5\pm1)\times10^{-3}$ and $(2.1\pm1)\times10^{-3}$, respectively, which are in approximate agreement with the values 4.99 $\times 10^{-3}$ and 2.93×10^{-3} for electric guadrupoles taken from the tables of Rose et al. Although the 1.137-Mev level could then have a spin of 0 or 4 and even parity if the transitions are pure electric, the latter case is ruled out by the beta-ray measurements. The $\log ft$ values of the beta-groups were calculated from the graphs¹⁵ of Moszkowski and are as follows: 3.55-Mev group-5.2, 3.1-Mev group-5.7, 2.44-Mev group-5.3, and 2.0-Mev group-5.6. All of these would appear to be allowed according to the classification¹⁶ of Nordheim. This would not be possible with a spin difference of 4 between the ground and second excited states.

The spins discussed thus far would explain the absence of the crossover transition from the second excited state and would account for the general trend of the angular correlation. The data would also suggest that Rh¹⁰⁶ has spin 1 and even parity. With this assumption and the fact that crossover transitions occur from the levels at 1.55 and 2.42 Mev these states could have a spin of either 1 or 2 and even parity in order for the beta-transitions to be allowed. The allowed nature of a beta-transition to the 2.42-Mev state is inferred from an estimate that the branching to this state is approximately 1.5 percent, based on the gamma-ray intensities, and that the $\log ft$ value is therefore less than 5. In view of the greater intensity of the cascade gamma-rays from both of these states, even though the crossovers are more energetic, it would be more favorable to assign spins of 2 to the 1.55- and 2.42-Mev levels.

After the partial decay scheme up to and including the 1.55-Mev level had been established⁴ several attempts were made to account more completely for the Rh¹⁰⁶ angular correlation. Kraushaar examined¹⁷ this case with NaI detectors in which some energy



FIG. 8. Empirical K/L ratios for electric transitions from Goldhaber and Sunyar (reference 13) showing the point for the 513-kev transition in Pd¹⁰⁶.

discrimination was employed. He was able to show that the effect of the 1.04-0.51 Mev cascade is to lessen the angular correlation anisotropy caused by the 0.624-0.513-Mev main cascade but that contribution of the former is not sufficiently intense to account for the total correlation. The greatest cancellation effect would occur if the 1.55-Mev level were assigned a spin of 2 which would be consistent with the beta-decay analysis. Kraushaar suggested that a loss-of-memory might be involved in the angular correlation of the 0-2-0 cascade. This hypothesis was tested¹⁸ by Steffen who measured the Rh¹⁰⁶ angular correlation after embedding the activity in various metals and ionic crystals. No changes in the correlation could be made and it was concluded that a loss-of-memory effect was not present. Experiments similar to those of Kraushaar have been carried out¹⁹ recently by Arfken, Klema, and McGowan. These authors were also not able to account for the total correlation effect.

Since levels at 1.55 and 2.42 Mev in Pd¹⁰⁸ appear to be excited in Rh¹⁰⁶ decay it is evident that in addition to the 0.624-0.513 Mev pair several weaker cascades occur, one involving three successive gamma-rays. It is possible that the angular correlation could be explained on the basis of the decay scheme presented. However, a more exact knowledge of the relative intensities of Rh¹⁰⁶ gamma-rays would be desirable.

The author is indebted to Dr. M. Goldhaber for many helpful discussions of this problem. Mr. Arthur Schwarzschild gave considerable assistance in taking and analyzing the data, and Miss Elizabeth Wilson and Dr. Seymour Katcoff prepared some of the sources. Thanks are also due to Dr. W. F. Hornyak for collaborating in some of the gamma-ray measurements and to Dr. I. Feister whose Fermi function tables were used. A 200-mC Rh¹⁰⁶ source used in part of this work was obtained on loan from Oak Ridge through the kind cooperation of Mr. John H. Gillette.

¹⁴ Rose, Goertzel, Spinrad, Harr, and Strong (privately circulated).

¹⁵ S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

 ¹⁶ L. W. Nordheim, Phys. Rev. 78, 294 (1950).
¹⁷ J. J. Kraushaar, Phys. Rev. 85, 727 (1952).

¹⁸ R. M. Steffen, Phys. Rev. 86, 632 (1952).

¹⁹ Arfken, Klema, and McGowan, Phys. Rev. 86, 413 (1952).