The Disintegration of Ruthenium 103 and Palladium 103*

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The beta- and gamma-ray spectra of Ru^{103} (43 days), Pd^{103} (17 days), and Rh^{103m} (56 min.) have been investigated in a magnetic lens spectrograph. Ru^{103} disintegrates with the emission of two beta-ray groups of end-point energies 0.684 Mev (six percent) and 0.204 Mev (94 percent) together with a gamma-ray of 0.494 Mev and an internally converted gamma-ray of 40.4-kev energy. Pd^{103} disintegrates by orbital electron capture. The spectrum consists of an electron line at 36.9 kev, shown to be due to L electrons from a highly converted gamma-ray of 40.4-kev energy, together with Auger electrons. The spectrum of the metastable Rh^{103m}, whose half-life has been determined as 56 min., consists of an L conversion line for a gamma-ray of 40.4 kev, with some indication of the presence of the K-line. Ru^{97} (2.8 days) was also measured and found to emit a gamma-ray of 0.217 Mev.

I. INTRODUCTION

THE two elements Ru^{103} and Pd^{103} both disintegrate to form Rh^{103}, the first by the emission of beta-rays, accompanied by gamma-rays and internal conversion electrons, and the second by K-electron capture. In both cases there is evidence that the metastable state of Rh^{103}, approximately 50-min. half-life, is formed. As will be shown below, Pd^{103} gives no radiation except that associated with the metastable state of Rh^{103}, together with x-rays and Auger electrons. Since the half-life of Pd^{103} is 17 days and since it can be produced with high specific activity, this element provides an excellent means of studying the radiations from Rh^{103m}.

Sullivan, Sleight, and Gladrow[1] have investigated two ruthenium activities produced by neutron and deuteron bombardment. Activities of half-life 42 days,[2] ascribed to Ru^{103}, and 2.8 days ascribed to Ru^{97}, were reported. The absorption experiments of Sullivan, Sleight, and Gladrow suggest that the 42-day Ru^{103} emits a gamma-ray of approximately 0.56 Mev energy and two beta-ray groups of approximately 0.3 (95 percent) and 0.8 (five percent) Mev. They report that Ru^{97} decays by orbital electron capture followed by a gamma-ray of approximately 0.23 Mev energy.

Hole[3] has measured the beta-ray spectrum of Ru^{103} in a spectrometer. The results of his investigation show that the spectrum is complex and that the energy of the highest energy component has an end point of 0.665 Mev. In addition, he finds three internal conversion lines of energies 34 kev, ascribed to Rh^{103m}, 217 kev and 290 kev. Mandeville and Shapiro[4] have made coincidence studies of the disintegration of Ru^{103} and find results essentially similar to those of Sullivan, Sleight, and Gladrow.

The radiations from Pd^{103} have been investigated by Matthews and Pool[5] and Brosi[6] who have shown that this element decays by K-electron capture with a half-life of 17 days. The former authors state that the decay takes place entirely through x-ray emission. They also observed the radiations from Rh^{103m} associated with the decay.

The metastable state Rh^{103m}, half-life 45 to 57 min., has been studied by various authors. Glendenin and Steinberg,[7] Flammersfeld and Bruna,[8] Wiedenbeck,[9]

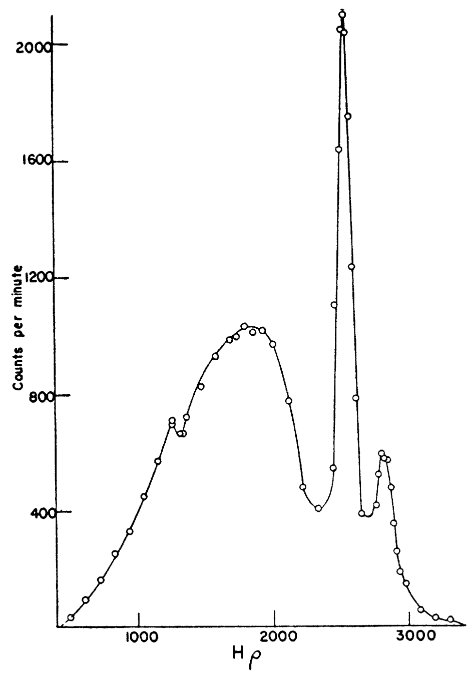

FIG. 1. Distribution of photo-electrons produced by gamma-rays of ruthenium.

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[8] A. Flammersfeld and O. Bruna, Zeits. f. Naturforschung **2a**, 241 (1947).
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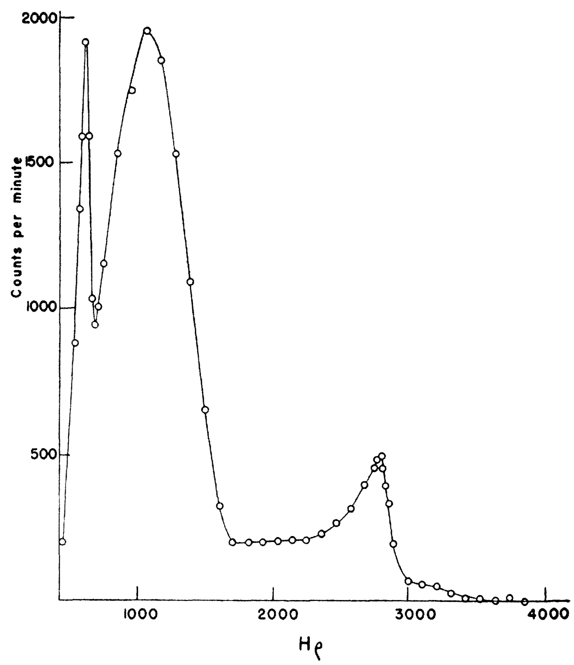

FIG. 2. Beta-rays and internal conversion electrons from Ru^{103}.

Hole,[10] and Gunlock and Pool[11] have all made observations on this element. The metastable state has been formed from normal Rh^{103} by fast neutrons[8] (n,n), by deuterons[8, 10, 11] (d,d^1) and by x-rays.[9] Gunlock and Pool found that x-rays of Rh were emitted in the disintegration and also found electron lines at 39.9 and 42.7 kev. Hole[10] measured the energy of the internal conversion lines in a magnetic spectrograph and found one line of energy 34 kev. He was not able to determine whether the electrons come from the K or L shell.

II. THE SPECTRUM OF Ru^{103} AND Ru^{97}

The present experiments on ruthenium were performed with a source of normal ruthenium prepared by neutron bombardment in the Oak Ridge Pile. The ruthenium was purified by distilling it as RuO_4 from a strongly acid solution, and was converted to the metallic state for use.

The spectrum of the photoelectrons ejected from a lead radiator of 16 mg/cm^2 surface density was measured in a magnetic lens spectrometer. The results are shown in Fig. 1. The K- and L-photoelectric lines corresponding to a gamma-ray of energy 0.494±0.002 Mev and a K-line for a gamma-ray of energy 0.217±0.002 Mev are to be seen. The line at 0.494 Mev decays with a period of 42 days and is to be ascribed to Ru^{103}. The line at 0.217 Mev decays with a period of 2.8 days and belongs to Ru^{97}.

The particle spectrum was next investigated. In the source which was used for this purpose the 2.8-day Ru^{97} had been allowed to die out. The first observations were made with a thin source, the activity of which was quite low. The measurements showed that there is a main group of beta-particles with a maximum energy of 0.204 Mev and a much weaker group of considerably higher energy. In addition, an internal conversion line corresponding to the gamma-ray at 0.494 Mev was found and also an internal conversion line of energy approximately 0.035 Mev, presumably emitted from the daughter substance Rh^{103m}.

In order to determine the end point of the higher energy group, and to get some information on the relative intensities of the two groups, a thicker source was measured. The results of the measurement are shown in Fig. 2. The internal conversion lines corresponding to the 0.494 Mev gamma-ray and the low energy line from Rh^{103m} are clearly seen. A Fermi plot of the beta-ray data was made and two beta-ray groups were found; one with an end point at 0.204±0.010 Mev (94 percent) and the other at 0.684±0.010 Mev (six percent). The ft-values for the two groups are $ft_{0.204} = 3.9\times10^5$ and $ft_{0.684} = 1.8\times10^8$. The lines at 0.217 and 0.290 Mev, mentioned by Hole, were not observed. It is believed that the lines which he reported were probably due to impurities.

Owing to the low specific activity of the source and the presence of the strong group of beta-rays of low energy, it is not possible to tell whether the line at 35 kev occurs as a result of electrons being ejected from the K or L shell. It was therefore decided to attempt to produce this line under circumstances in which there would be no beta-rays and under which the source would be as free as possible of inert material. This can be done best through an investigation of Pd^{103}.

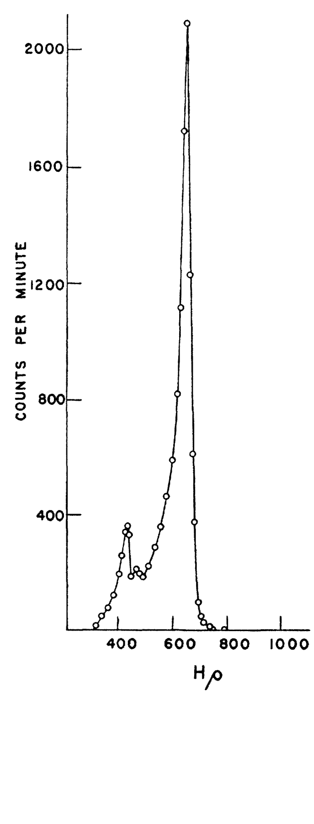

FIG. 3. Internal conversion electrons and Auger electrons from Pd^{103}.

[10] N. Hole, Arkiv. f. Mat. Astr. o. Fys. **34B**, No. 5 (1947).
[11] H. F. Gunlock and M. L. Pool, Phys. Rev. **74**, 1264(A) (1948).

III. THE SPECTRUM OF Pd^{103}

Pd^{103} was made by bombarding rhodium metal with 23-Mev alpha-particles in the cyclotron. The rhodium was hard-soldered to a probe and given a bombardment of several hundred microampere-hours of alpha-particles. The rhodium was put into solution by fusing. A few milligrams of palladium carrier were added and the palladium was precipitated as palladium-dimethylglyoxime. The material was then converted to the oxide for use as a gamma-ray source. The oxide was dissolved in *aqua regia* and a drop of this solution was evaporated and used as a beta-ray source.

The gamma-ray source, using a lead radiator, was measured in the spectrometer. No photo-electrons at all were seen. The line of energy 0.494 Mev was entirely absent. In an attempt to find the gamma-ray which gives rise to the internally converted line at 35 kev, a thin copper radiator, instead of the lead radiator was used, and a search was made for photo-electrons. If the line at 35 kev results from conversion in the K shell the gamma-ray energy should be 59 kev, and if it results from conversion in the L shell the gamma-ray energy should be 40 kev. No photo-electrons were found corresponding to a gamma-ray of either energy. This suggests that the gamma-ray giving rise to the line is very highly internally converted.

The beta-ray spectrum of Pd^{103} was investigated using a counter fitted with a thin zapon window. The result of the investigation is shown in Fig. 3. It will be noted that only three low energy lines appear in the spectrum. There are no positrons.

The most intense line of the spectrum corresponds to that observed in Ru^{103}. The energy of the line is 36.9 ± 0.5 kev. If it be assumed that this line can be attributed to the ejection of electrons from the K shell by a gamma-ray of energy 60 kev, the L-line should easily be seen. Since no line is seen corresponding to the ejection of electrons from the L shell by a gamma-ray of 60 kev it must be assumed that the line in question is either an L-line or that the K/L ratio is at least 100:1. In order to explain the half-life of the Rh^{103m} state, it is necessary to assume a spin change of approximately 4 units. This would entail, at these energies, a $K/L<1$. This suggests that the line in question arises from the conversion in the L shell of a 40.4 kev gamma-ray.

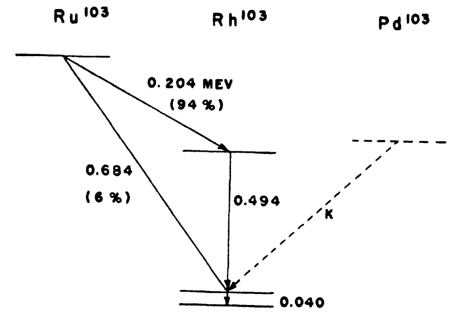

FIG. 5. Energy levels of Rh^{103}.

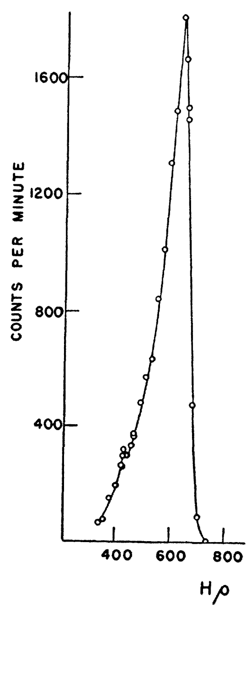

FIG. 4. Internal conversion electrons from Rh^{103m}.

It will be noted also that there are two lower energy lines in the spectrum. These come at a setting of 0.22 and 0.2025 amp. for the current in the field coils. If the 40.4 kev gamma-ray is internally converted in the K shell, the electrons should have an energy of 16.89 kev and should be focused at 0.2041 amp. On the other hand, the Auger electrons ($K-2L_I$), arising from orbital electron capture will have a energy of 16.40 kev and will be focused at 0.2075 amp. It is impossible to tell what fraction of the line at 0.2025 amp. is due to Auger electrons and to electrons internally converted in the K shell. The line at 0.22 amp. is clearly an Auger line corresponding to $K-L_I-M$, which should be focused at 0.2185 amp.

Finally, a source of Rh^{103m} was prepared by adding a small amount of rhodium to the Pd^{103} parent and precipitating the palladium away from the rhodium. The period of the rhodium was measured and found to be 56 min. A beta-ray source was prepared and measured using the same counter and window as that used for the investigation of Pd^{103}. The results are shown in Fig. 4. In order to get sufficient intensity the spectrometer had to be used at lower resolution than in the experiments on Pd^{103}. The results show the line at 36.9 kev together with a small line at the correct position to be a K-line. This line, at 0.205 amp., is not as intense, relative to the main group as is the similar line in Pd^{103}. It would appear, therefore, that the low energy line seen in Pd^{103} owes most of its intensity to Auger electrons and only slightly to electrons converted in the K shell.

IV. CONCLUSION

The present experiments on Ru^{103} show that this element disintegrates with the emission of two beta-ray groups of end-point energies 0.684 and 0.204 Mev. In addition, a gamma-ray of energy 0.494 Mev is found together with L conversion electrons from a gamma-ray of 40.4 kev. The energy difference between the two beta-ray groups is 0.480 Mev, somewhat smaller than the energy of the gamma-ray as determined from the distribution of the photo-electrons. While not absolutely certain, it seems reasonably probable that both transitions lead to the metastable state Rh^{103m} 40.4 kev above the ground state.

That the metastable state Rh^{103m}, of 56-min. half-life, decays by the emission of an internally converted gamma-ray of energy 40.4 kev is shown by the work on Pd^{103} and Rh^{103m}. As has been pointed out above, the absence of any L-line attributable to a gamma-ray of 60 kev, definitely shows that the line at 36.9 kev must be an L-conversion line of a 40.4 kev gamma-ray. One can account for the mean life of Rh^{103m} by assuming a spin change $l=4$ and a value of $N_e/N_\gamma \sim 10^3$. In addition the ratio N_K/N_L would be extremely small, in agreement with experiment. From the results of the above experiments a tentative energy level scheme for Rh^{103} is given in Fig. 5.

The authors wish to express their thanks to Dr. Milo B. Sampson and the cyclotron crew for making the bombardments. They are also indebted to Miss Elma Lanterman for preparing the chemical separations.

Note Added in Proof: Since this paper was sent to press, a fission product source of Ru^{103}, also containing Ru^{106} and its daughter Rh^{106}, has been measured. Using a source and counter window 3 mm in diameter, the conversion line at 0.494 Mev has been examined. Under these conditions the breadth of the K-line at half-maximum is 1.3 percent and the K and L lines have been resolved. The ratio $N_K/N_L=6.5$. In addition the low energy beta-ray group has been re-examined. The end point is somewhat higher, 0.222 Mev. The internal conversion coefficient, $(N_e)_K/N_\gamma$, for the line at 0.494 Mev is $5.5\pm 0.5\times 10^3$. The line is either electric quadrupole or magnetic dipole since the conversion coefficient values calculated by Rose *et al.* give $\alpha_2=5.37\times 10^{-3}$ and $\beta_1=5.10\times 10^{-3}$.

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Nuclear Energy Level Argument for a Spheroidal Nuclear Model*

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Recently there has been notable success, particularly by Maria Mayer, in explaining many nuclear phenomena including spins, magnetic moments, isomeric states, etc. on the basis of a single particle model for the separate nucleons in a spherical nucleus. The spherical model, however, seems incapable of explaining the observed large quadrupole moments of nuclei. In this paper it is shown that an extension of the logic of this model leads to the prediction that greater stability is obtained for a spheroidal than for a spherical nucleus of the same volume, when reasonable assumptions are made concerning the variation of the energy terms on distortion. The predicted quadrupole moment variation with odd A is in general agreement with the experimental values as concerns variation with A, but are even larger than the experimental values. Since the true situation probably involves considerable "dilution" of the extreme single particle model, it is encouraging that the present predictions are larger rather than smaller than the experimental results. A solution is given for the energy levels of a particle in a spheroidal box.

I. INTRODUCTION

RECENTLY considerable evidence has been pointed out for the existence of nuclear shell structure on such basis as nuclear spins, magnetic moments, and on the degree of forbiddenness of various β-ray spectra.[1] Probably the most successful scheme is that of Maria Mayer who treats the nuclear energy levels as due to a filling-up of individual particle levels for nucleons in a spherical box. It is assumed that the strong interaction of each nucleon with all other nucleons in the nucleus can be approximated as a (roughly constant) interaction potential extending over the nuclear volume such that the assemblage of nucleons forms a "self consistent" box. When an even number of neutrons or protons are present, it is well known that they pair off to give zero spin and moment, and great success is obtained by attributing the spin and moments to the odd nucleon alone for odd A nuclei. Maria Mayer assumes that strong $L-S$ coupling splits the levels for a given L, but otherwise maintains the normal order nearly intact to explain the closed shell values Z_c or $N_c=2$, 8, 20, 50, 82, and 126 "magic numbers."

Similarly, it has been emphasized[2] that evidence for the nuclear shell structure is also shown by the nuclear quadrupole moments. The material of the following

* Supported by the AEC.

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