

The Radiations of Tantalum¹⁸², Rhenium^{186,188}, and Gold^{199*}

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A survey of the radiations of Ta¹⁸², Re^{186,188} and Au¹⁹⁹ has been made down to energies of the order of 5 kev with a small 180 degree spectrometer.

The disintegration scheme of Ta¹⁸² is complex. The twenty or more conversion lines found superimposed on the beta-ray spectrum and the numerous peaks found in the photoelectron spectrum are analysed into seventeen gammas in the energy region 80 to 330 kev. In addition, three gamma-rays of energies 1.13, 1.22, and 1.24 Mev are found. The continuous spectrum has an end point at 0.53 Mev.

The radiations of Re¹⁸⁶ consist of a single beta-ray group of maximum energy 1.07 Mev and two gamma-rays of energies 0.138 and 0.212 Mev. The 0.138 Mev gamma-ray is converted. Coincidence experiments strongly suggest that the mode of decay is one involving the emission of a single beta-ray group, followed by the cascade emission of the two gamma-rays. The disintegration of Re¹⁸⁸ is more complicated. A single group of beta-rays is found with the end point at 2.10 Mev. Five gamma-rays with energies essentially the same as those reported by Miller and Curtiss are present.

All the radiations of Au¹⁹⁹ lie below 0.32 Mev. Superimposed on the beta-ray spectrum are eight conversion lines that may be attributed to 4 gamma-rays having energies of 0.024, 0.051, 0.156 and 0.207 Mev. The beta-rays have a maximum energy of 0.32 Mev. A possible decay scheme is suggested.

I. INTRODUCTION

THE disintegration schemes of the heavier unstable isotopes are in many cases complex and the radiation is characterized by the presence of several beta-ray groups and numerous gamma-rays, especially in the low energy region. As a consequence, there generally appears a number of conversion lines superimposed on the beta-ray spectra of these elements with the *K*, *L*, and *M* components so disposed as to make analysis difficult. The radiations of Ta¹⁸², Re^{186,188}, and Au¹⁹⁹ which are discussed in this paper are typical examples. Since the low energy region is of particular importance in these cases, most of the previous work has been done with spectrometers using photographic plate detection.¹ This gives a rather accurate determination of the energies of the individual conversion lines but does not give sufficient information about the relative intensities or the photoelectron spectrum to make analysis into corresponding gamma-rays definite. In this work an attempt has been made to determine the energies of the gamma-rays with greater certainty by studying both the photoelectrons and beta-rays with thin window G-M tube detection.

II. APPARATUS

A small 180° spectrometer, previously described, was used for the measurements.² The range of the instrument has been extended down to about 5 kev by the use of very thin window G-M tube detection. The windows consist of five to seven layers of zapon film 0.005 mg/cm² in thickness supported on a grid. During the filling process the spectrometer chamber and tube are evacuated simultaneously, thus eliminating excessive pressure on the window. The tube is filled from a

reservoir containing the proper mixture of argon and alcohol. In the event of slow diffusion of the mixture through the window and consequent threshold change, the counter can be stabilized by leaving it connected to the reservoir. In most of the work the diffusion was negligible during the period of operation. Since several hours were sometimes required for the counter to come

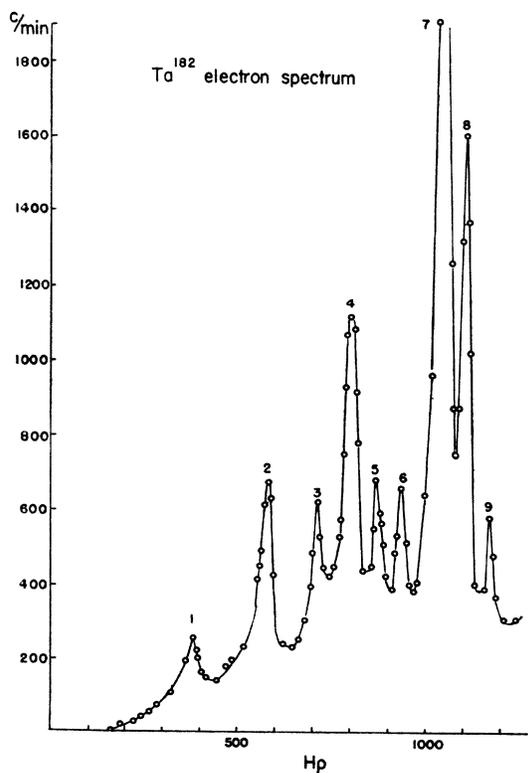


FIG. 1. The beta-ray spectrum of Ta¹⁸² in the low energy region obtained with a G-M tube detector having a window 0.03 mg/cm² thick. See Tables I and III for the energies and identification.

* Assisted by the Joint Program of the ONR and the AEC.

¹ J. M. Cork, Phys. Rev. **72**, 581 (1947).

² C. L. Peacock and R. G. Wilkinson, Phys. Rev. **74**, 297 (1948).

TABLE I. The conversion lines of Ta¹⁸².

Line* number	Energy (Mev)	Identification	Line* number	Energy (Mev)	Identification
1	0.0129	K_1	11	0.1395	$L_7 + M_6$
2	0.0286	K_2	12	0.1541	$K_{11} + L_8 + M_7$
3	0.0428	K_3	13	0.1595	$L_9 + M_8$
4	0.0527	K_4	14	0.1699	M_9
5	0.0625	K_5	15	0.1795	K_{12}
6	0.0690	$K_6 + L_1$	16	0.1882	$K_{13} + L_{10}$
7	0.0877	$K_7 + L_2$	17	0.1963	$K_{14} + L_{11} + M_{10}$
8	0.0957	$K_8 + L_3 + M_2$	18	0.2189	$K_{15} + M_{11}$
9	0.1105	$K_9 + L_4 + M_3$	19	0.2294	$K_{16} + L_{12}$
10	0.1286	$K_{10} + L_5 + M_5$	20	0.2539	K_{17}

* The number in this column refers to the legend of Fig. 2.

to equilibrium after filling, the data above 60 kev was taken with a conventional counter having a mica window 2 mg/cm² in thickness.

III. TANTALUM¹⁸²

The radiations of Ta¹⁸² have been studied by several investigators in various energy regions. Cork¹ has surveyed the conversion electrons in the low energy region with a 180° spectrometer using photographic plate detection. Some thirty-four conversion lines are reported which he has attributed to sixteen gamma-rays on the basis of the characteristic differences of the K , L , and M binding energies of tungsten. Rall and Wilkinson³ have reported gamma-rays of energies 0.15, 0.22, 1.13, and 1.22 Mev from a study of the photoelectron spectrum. No attempt was made to examine the low energy part of the spectrum. The beta-ray end point was found to be 0.53 Mev. The coincidence studies of Mandeville and Scherb⁴ suggest the presence of only one group of beta-rays. They concluded that

TABLE II. The photoelectron lines of Ta¹⁸².

Line number*	Energy (Mev)	Identification	Line number*	Energy (Mev)	Identification
1	0.0435	K_5	11	0.1760	K_{13}
2	0.0530	K_6	12	0.2055	$K_{15} + L_{11}$
3	0.0656	L_1	13	0.2370	$K_{17} + L_{13}$
4	0.0698	K_7	14	0.2480	L_{14}
5	0.0840	K_9	15	1.045	K_{18}
6	0.0975	L_3	16	1.131	K_{19}
7	0.1140	$K_{10} + L_5$	17	1.149	K_{20}
8	0.1340	K_{11}	18	1.201	L_{19}
9	0.1550	$K_{12} + L_9$	19	1.234	L_{20}
10	0.1640	K_{13}			

* The number in this column refers to the legend in Fig. 3.

the high energy gamma-rays are not in cascade since no appreciable gamma-gamma coincidences were found.

Sources for the present investigation were prepared from a sample of TaO₂ irradiated by slow neutrons in the Oak Ridge pile. Chemical separations were performed to insure purity, with special attention given to the separation of calcium, iron, and tungsten. Sources were made from Ta₂O₅ precipitated from K₈Ta₃O₁₉. For the low energy work, the beta-ray source consisted of less than 0.2 mg/cm² of Ta₂O₅ on a backing of zapon film about 0.003 mg/cm² thick.

Figure 1 shows the beta-ray spectrum obtained with the very thin window G-M tube detector in the region 7–125 kev. The remainder of the beta-ray spectrum was taken with a conventional mica window tube (Fig. 2). In all, twenty conversion lines are found superimposed upon the beta-ray spectrum. Undoubtedly, greater resolution would reveal further L and M components reported by Cork. Line No. 7 is by far the most intense. Assignment of the gamma-ray energies from these data

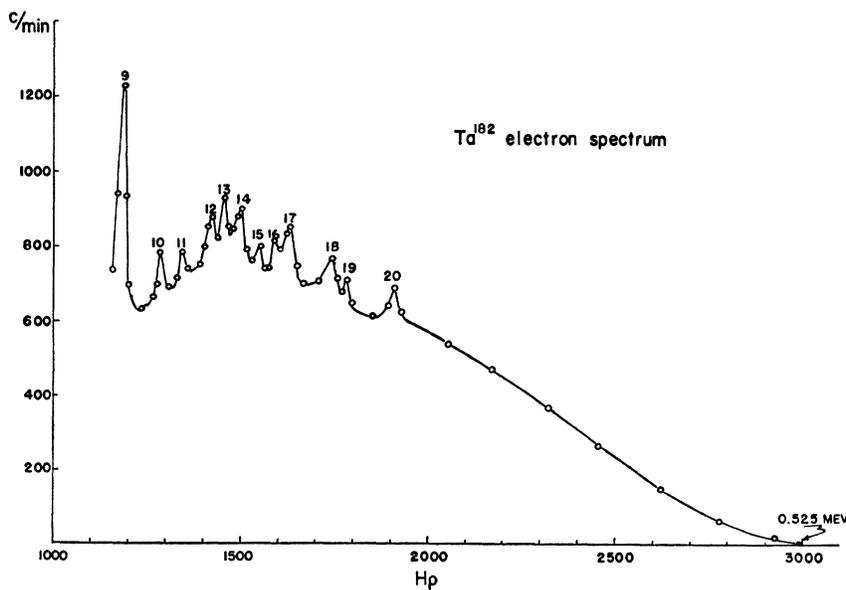


FIG. 2. The higher energy portion of the beta-ray spectrum obtained with a G-M tube detector having a mica window 2 mg/cm² thick. See Tables I and III for the energies and identification.

³ W. Rall and R. G. Wilkinson, Phys. Rev. **71**, 321 (1947).

⁴ C. E. Mandeville and M. V. Scherb, Phys. Rev. **73**, 340 (1948).

alone is difficult and uncertain, since the lines are closely spaced and since it is possible for *L* and *M* lines from certain gamma-rays to be masked by stronger *K* lines of others. Furthermore, it is possible for the *L* line in some cases to be stronger than the *K*, so that relative intensities do not furnish a sufficient criterion. Table I summarizes the conversion electron data and gives the most probable gamma-ray assignments consistent with the photoelectron data. Although four significant figures are given for convenience in analysis, no claim for accuracy of the measurements to this extent is made.

In order to make gamma-ray assignments more positive, the photoelectrons ejected from a 30 mg/cm² lead radiator were studied. Figure 3 shows 16 of the 19 lines so obtained, together with the strong Compton electron distribution associated with the high energy gamma-rays. Three lines measured with the thin window counter are not shown. The high energy lines are readily resolved into *K* and *L* components due to three gamma-rays at 1.13, 1.22, and 1.24 Mev. Rall and Wilkinson³ have previously reported only two gamma-rays in this region, the latter two apparently being unresolved. Table II gives the energies of the photoelectron lines and the most probable gamma-ray assignments.

The analysis of all data is given in Table III. In at least four cases no photoelectron lines were found. The *K* photoelectron lines associated with the first four

TABLE III. Analysis and summary of the conversion line and photoelectron line data.

Gamma-ray number*	Gamma-energy (Mev) from conversion in the:			Gamma-energy (Mev) from photoelectrons from:		Average gamma-energy (Mev)
	<i>K</i> shell	<i>L</i> shell	<i>M</i> shell	<i>K</i> shell	<i>L</i> shell	
1	0.0822	0.0811	—	—	0.0816	0.0820
2	0.0979	0.0998	0.0985	—	—	0.0980
3	0.1121	0.1078	0.1133	—	0.1135	0.1120
4	0.1220	0.1226	—	—	—	0.1220
5	0.1318	—	0.1314	0.1315	0.1300	0.1317
6	0.1383	0.1407	0.1423	0.1410	—	0.1410
7	0.1570	—	0.1569	0.1578	—	0.1570
8	0.1650	0.1662	0.1623	—	—	0.1650
9	0.1798	0.1716	0.1727	0.1720	0.1710	0.1720
10	0.1979	0.2003	0.1991	0.2020	0.1920	0.1980
11	0.2234	—	0.2217	0.2220	0.2215	0.2220
12	0.2488	0.2415	—	0.2430	—	0.2430
13	0.2575	—	—	0.2520	0.2530	0.2550
14	0.2656	0.2660	—	0.2640	0.2640	0.2640
15	0.2882	—	—	0.2935	—	0.2900?
16	0.2987	—	—	—	—	0.2990?
17	0.3232	—	—	0.3250	—	0.3240
18	—	—	—	1.1330	—	1.1330
19	—	—	—	1.2190	1.2170	1.2190
20	—	—	—	1.2370	1.2500	1.2370

* The number in this column refers to the identification number in column three of Tables I and II.

gamma-rays are too low in energy to be observed. In other cases, e.g., gamma-ray No. 8, the gamma-ray is probably highly converted. The overlapping of several components of the conversion electron data makes it

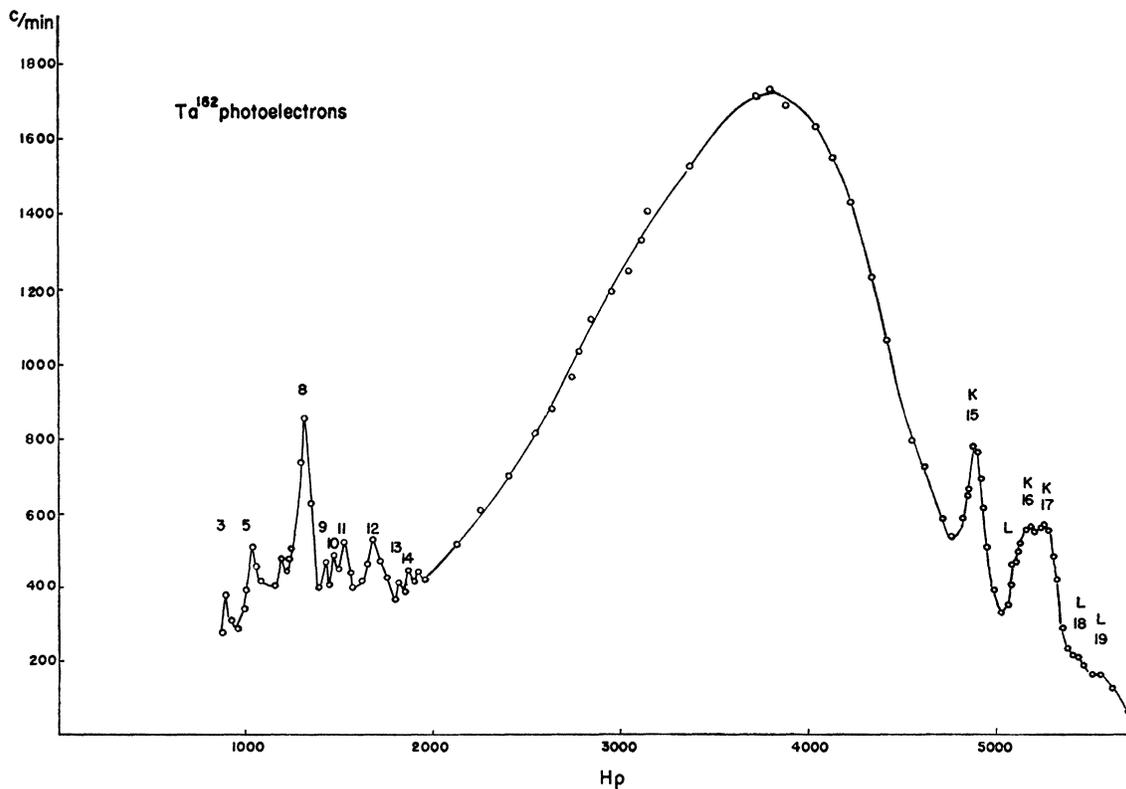


FIG. 3. Compton- and photoelectrons ejected from a 30 mg/cm² Pb radiator by the gamma-rays of Ta¹⁶². See Tables II and III for the energies and identification.

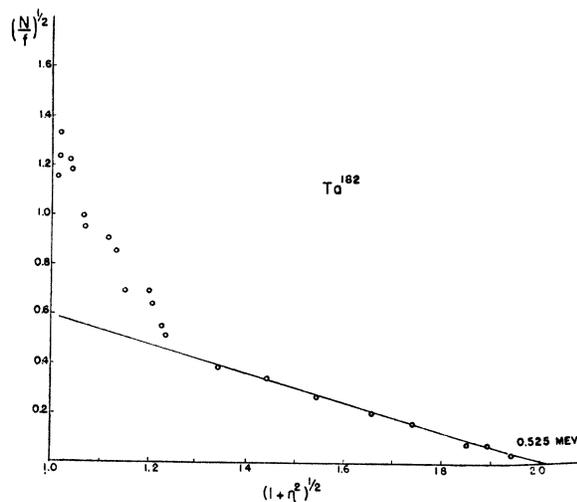


FIG. 4. Fermi plot of the beta-rays of Ta^{182} . The deviation from the straight line at low energy is possibly due to a second group having a maximum energy at about 0.25 Mev.

impossible in some instances to make a definite assignment. For example, the L conversion line of gamma-ray No. 5 is masked by the K line of No. 10. If the identification is correct, gamma-rays Nos. 7, 8, 9, 11, 18, 19, 20 are the most intense. Gamma-ray No. 7, assigned as 0.157 Mev, is the most intense of the low energy group and is highly converted. The corresponding conversion line has been previously reported by Cork¹ as the L line due to a gamma-ray at 0.996. However, the photoelectron spectrum suggests that the assignment as a K line is more probable. Gamma-ray No. 11, assigned as 0.222 Mev, exhibits the strongest photo-peak in the low energy region and is weakly converted. Of all the gamma-

rays, those of highest energy are the most intense (Nos. 18, 19, 20) and are about equally strong. Even with the additional information afforded by the photoelectron spectrum, some uncertainty persists in the identification. However, at least seven of the suggested gamma-rays of Table III agree with Cork's assignment.

A Fermi plot of the continuous portion of the beta-ray spectrum is shown in Fig. 4. The graph is a straight line down to 0.15 Mev and yields an end point at 0.525 Mev, in agreement with the previous value given by Rall and Wilkinson.³ There is some evidence for another group at approximately 0.25 Mev but the presence of the conversion lines does not permit positive identification.

The results of this investigation may be summarized as follows. Ta^{182} decays by the emission of a beta-ray of maximum energy 0.53 Mev and possibly by the emission of another group of beta-rays of approximately 0.25 Mev to excited W^{182} , which returns to the ground state by complex gamma-ray emission. The gamma-rays of highest energy (1.13, 1.22, and 1.24 Mev) are the most intense. Of the fifteen or more low energy gamma-rays (Table III) those of energies 0.157, 0.165, and 0.222 Mev are the most prominent. No attempt at formulating a decay scheme is made. Since the high energy gamma-rays are so close in energy, the coincidence data of Mandeville and Scherb⁴ does not eliminate the possibility of cascade emission. This, however, is probably not the case.

IV. RHENIUM^{186, 188}

The previous studies of the radiations of the 90-hour Re^{186} have been made by absorption⁵ and cloud-chamber⁶ methods. A single group of beta-rays with a maxi-

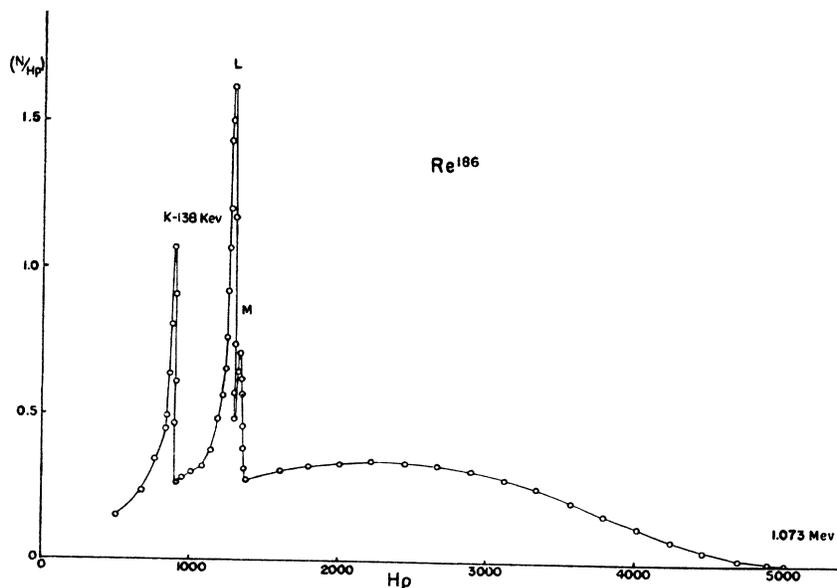


FIG. 5. The beta-ray spectrum of Re^{186} . The value of the energy is for the gamma-ray, not the conversion line.

⁵ L. J. Goodman and M. L. Pool, Phys. Rev. **71**, 288 (1947).

⁶ K. Sinma and H. Yamasaki, Phys. Rev. **55**, 320 (1939).

imum energy of 1.07 Mev is reported. No gamma-rays have been observed. The 18-hour Re^{188} has been investigated by a number of workers. Only the gamma-rays have been studied by spectrometer methods. By means of a thin lens spectrometer, Miller and Curtiss⁷ found five gamma-rays associated with Re^{188} . The coincidence work of Mandeville, Scherb, and Keighton⁸ suggests a complex mode of decay involving two groups of beta-rays and the cascade emission of some of the gamma-rays.

The results of the present work were obtained with neutron activated samples from Oak Ridge. High specific activity permitted the use of very thin sources ($<0.1 \text{ mg/cm}^2$) for the beta-ray studies. Figure 5 shows the beta-ray spectrum of Re^{186} taken after the Re^{188} had decayed. Three conversion lines are present all of which

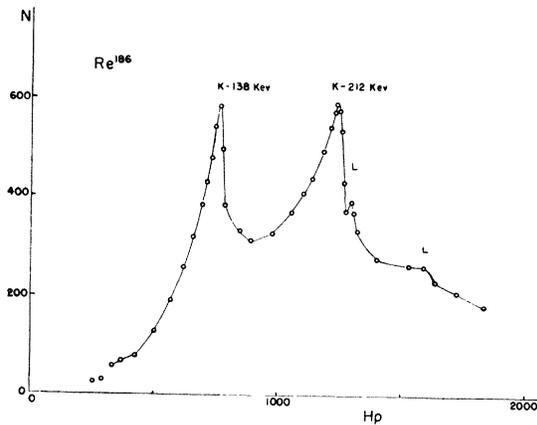


FIG. 6. Photoelectrons ejected from a 32 mg/cm^2 Pb radiator by the gamma-rays of Re^{186} . The values of the energies are for the gamma-rays, not the photoelectron lines.

decay with the 90-hour period. The energies of the conversion lines correspond to the *K*, *L*, and *M* components associated with a single gamma-ray of energy 0.138 Mev. The photoelectron spectrum, measured after most of the 18-hour activity had died away, is shown in

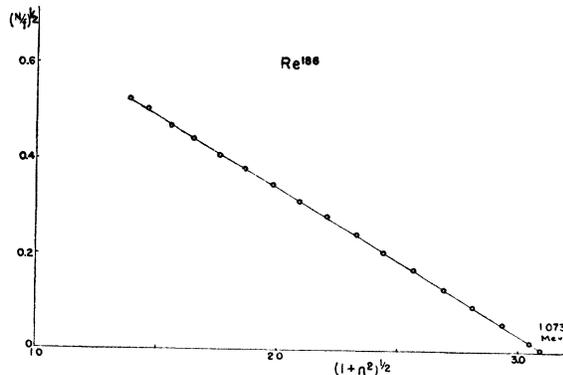


FIG. 7. Fermi plot of the beta-rays of Re^{186} .

⁷ L. C. Miller and L. F. Curtiss, Phys. Rev. **70**, 983 (1946).
⁸ Mandeville, Scherb, and Keighton, Phys. Rev. **74**, 888 (1948).

TABLE IV. The gamma-rays of Au^{199} .

Conversion line energy (Mev)	Photo-line energy (Mev)	Gamma-ray energy (Mev)
0.0090(L)	—	0.024
0.0360(L)	—	0.051
0.0473(M)	—	
—	0.054(L)	0.070
0.0735(K)	0.068(K)	0.156
0.1470(L)	—	
0.1590(M)	—	
0.1243(K)	0.119(K)	0.207
0.1970(L)	0.189(L)	
0.1470(K)	0.143(K)	0.230

Fig. 6. Two lines are present which correspond to gamma-rays having energies 0.138 and 0.212 Mev. If the 0.212-Mev gamma-ray is converted, the conversion must be weak. Unfortunately, the *K* conversion line would be superimposed on the *M* line due to the 0.138-Mev gamma-ray so that no definite decision can be made. A conventional Fermi-plot of the continuous spectrum (Fig. 7) shows conclusively that only one group of beta-rays is present with a maximum energy of 1.07 Mev.

The interpretation of these results is made clear by the recent coincidence measurements of Cuffey and Journey⁹ in this laboratory. They find a substantial number of beta-gamma- and gamma-gamma coincidences which are associated with the 90-hour activity. It is therefore natural to assume that the gamma-rays are in cascade and that the decay scheme of Re^{186} (Fig. 8) involves the emission of beta-rays of a single group, which leaves the resulting Os^{186} in an excited state 0.350 Mev above ground. On this assumption, the conversion coefficients of the 0.138-Mev gamma-ray are $\alpha_K = 3$ percent, and $\alpha_L = 7$ percent. The *K* to *L* ratio is found to be 0.40 and suggests a spin change of 2 or 3 for this gamma-ray transition, according to the curves of Hebb and Nelson.¹⁰ The conversion coefficient of the third line shown in Fig. 5 is 1.2 percent and represents the sum of the *K* conversion of the 0.212-Mev gamma-ray and the *M* conversion of the 0.138-Mev gamma-ray. Com-

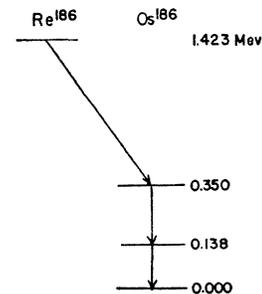


FIG. 8. Probable decay scheme of Re^{186} .

⁹ To be published.
¹⁰ M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).

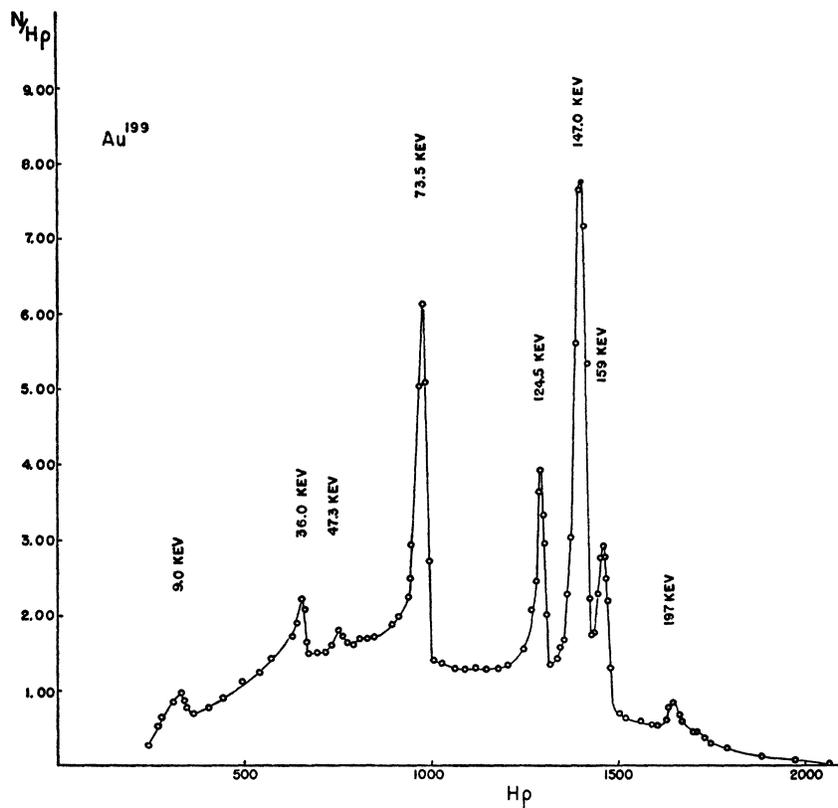


FIG. 9. The beta-ray spectrum of Au¹⁹⁹. The energies given are those of the conversion lines, not the associated gamma-rays.

parison of the values with the tables of Rose and his associates¹¹ shows that both gamma-rays are probably electric dipole radiation. The coincidence work of Cuffey and Jurney further indicates that no metastable state exists in the Os¹⁸⁶.

The data obtained for the 18-hour Re¹⁸⁸ essentially verifies the findings of Miller and Curtiss.⁷ Five gamma-rays were found with energies 0.15, 0.48, 0.64, 0.95, and

1.40 Mev. The 0.15-Mev gamma-ray was found to be converted. The 1.40-Mev gamma-ray is probably quite strong in spite of the fact that the corresponding photoelectron line was observed to be weak. The presence of beta-rays in excess of two Mev necessitated the use of a thick copper absorber, hence the geometry for photoelectron production was poor. A strong Compton elec-

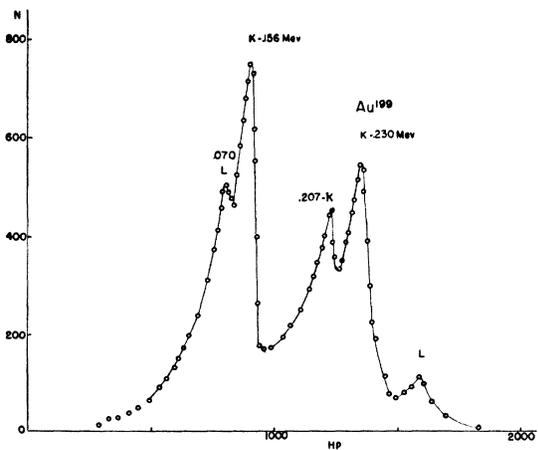


FIG. 10. Photoelectron spectrum of Au¹⁹⁹. The energies are those of the gamma-rays, not the photoelectron lines.

¹¹ Rose, Goertzel, Spinrad, Harr, and Strong, "Tables of K-Shell Internal Conversion Coefficients" (privately distributed prior to publication).

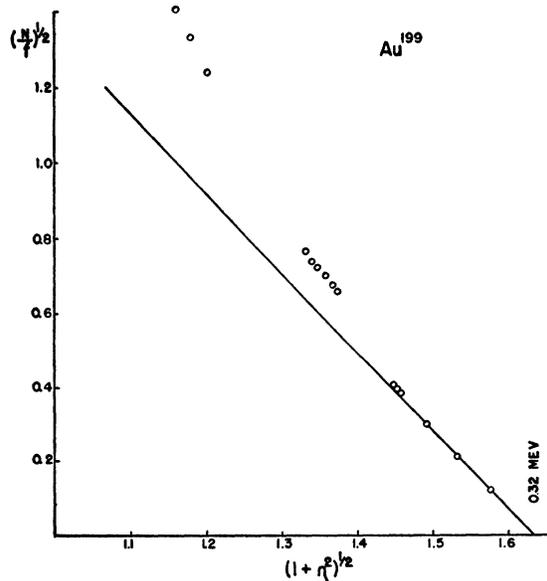


FIG. 11. Fermi plot of the beta-rays of Au¹⁹⁹.

tron distribution due to this gamma-ray was obtained owing to the thick copper absorber. A significant result is the fact that only one group of beta-rays was found. Two groups have been previously reported by Beach *et al.*¹² and by Mandeville, Scherb, and Keighton.⁸ However, subsequent measurements with cyclotron activated samples led to the conclusion that there is a single group of beta-rays with a maximum energy of 2.10 Mev.

V. GOLD¹⁹⁹

The radiations of 3.3-day Au¹⁹⁹ are entirely in the low energy region and therefore have not been previously studied by spectrometer methods. Absorption and coincidence measurements have been made by Krishnan and Nahum,¹³ Mandeville, Scherb, and Keighton¹⁴ and by Meem and Maienschein.¹⁵ The latter group finds evidence for one partially converted gamma-ray at 0.18 Mev and a simple beta-ray spectrum with an end point at 0.38 Mev.

The sources used for this study were prepared by separating Au¹⁹⁹ from its 31-minute parent, Pt¹⁹⁹, produced by neutron bombardment at Oak Ridge. Separation was accomplished by repeated ethyl acetate extraction of AuCl₃. Subsequent examination of the decay showed that the separation was good. Beta-ray sources were made by evaporating a water solution of AuCl₃ on a zapon backing approximately 0.003 mg/cm² thick. Since the specific activity was very high, the sources were less than 0.1 mg/cm² thick. Photoelectron sources were made by depositing the AuCl₃ on a 10-mil copper strip to eliminate the beta-rays. The radiator was a lead strip 32 mg/cm² thick.

The complete beta-ray spectrum is shown in Fig. 9. The continuous spectrum is complicated by the presence of eight conversion lines from 9 to 197 kev in energy. The 9-kev line is probably more intense than it appears to be, since it lies in the region of strong counter window cut-off. Figure 10 shows the photoelectron spectrum produced by the gamma-rays. The five lines are readily identified as the *K* and *L* components due to four gamma-rays. Table IV shows the analysis of the conversion and photoelectron data. Estimation of the relative intensities of the six gamma-rays is made difficult by the overlapping of several of the components. The photoelectron spectrum shows that the 0.156, 0.207, and 0.230-Mev gamma-rays are the most promi-

¹² Beach, Peacock, and Wilkinson, *Phys. Rev.* **76**, 187 (1949).

¹³ R. S. Krishnan and E. A. Nahum, *Proc. Camb. Phil. Soc.* **37**, 422 (1941).

¹⁴ Mandeville, Scherb, and Keighton, *Phys. Rev.* **74**, 601 (1948).

¹⁵ L. Meem and F. Maienschein, *Phys. Rev.* **76**, 328 (1949).

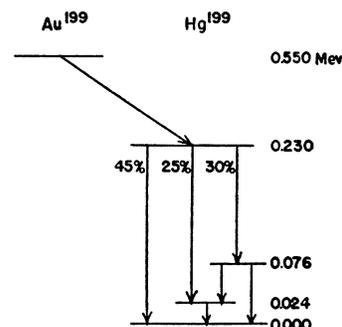


FIG. 12. Possible decay scheme of Au¹⁹⁹.

nent. Estimates based on Gray's¹⁶ empirical curve suggest that the 0.156 and the 0.207-Mev gamma-rays are about the same intensity, while the 0.230-Mev gamma-ray is about twice as strong as these. However, the conversion electron data shows that the 0.156-Mev radiation is most strongly converted, with the *L* conversion being at least as strong as the *K*. The *K* conversion of the 0.230-Mev gamma-ray, if any, overlaps the *L* line of the 0.156-Mev gamma-ray. The fact that no *L* conversion of the 0.230 gamma-ray is detected suggests that conversion in this case is weak. It may be assumed that these three gamma-rays are not in cascade and that they account for 30 percent, 25 percent, and 45 percent of the transitions from the highest excited state of the Hg¹⁹⁹. This assumption is in accord with the results of Mandeville, Scherb, and Keighton,¹⁴ since they found no gamma-gamma coincidences.

A conventional Fermi plot of the continuous portion of the beta-ray spectrum is shown in Fig. 11. No attempt is made to interpret the curvature since the spectrum lies wholly in the low energy region and is perturbed by the presence of many conversion lines. Probably only one group of beta-rays is present with an end point at 0.32 Mev. This conclusion is substantiated by Mandeville *et al.*¹⁴ and Meem¹⁵ since their $N_{\beta-\gamma}/N_{\beta}$ coincidence curve was a straight line, characteristic of a simple spectrum.

Figure 12 summarizes the results and suggests a possible decay scheme. The mode of decay given is very consistent with the measured energy values, coincidence data, and the relative intensities. One apparent inconsistency appears in connection with the 0.024-Mev gamma-ray which is observed to be weak but which must be assumed to be strong since it follows two of the most dominant gamma-rays. However, it lies too low to be observed as a photo-line and the conversion line is in the region of counter tube cut-off.

¹⁶ L. H. Gray, *Proc. Camb. Phil. Soc.* **27**, 103 (1931).