Radiations from W^{185} and W^{187}†

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The radiations of W^{185} and W^{187} have been examined in a thin-lens magnetic spectrometer, by coincident scintillation spectrometers, and with a gray-wedge analyzer. W^{185} was found to emit no detectable gamma rays. Its beta spectrum is first-forbidden ($\Delta I=1$, yes!) and of maximum energy 440±5 kev. By observation of internal conversion lines and photoelectric peaks, fourteen gamma rays have been resolved in the decay of W^{187}. Three beta-ray spectra are emitted with end-point energies of 340±20, 630±10, and 1325±15 kev with relative intensities of 10%, 70%, and 20%. The most energetic spectrum has the unique shape associated with a spin change of two and a change of parity.

INTRODUCTION

When naturally occurring wolfram is irradiated by slow neutrons, radionuclides W^{181} (140 day), W^{185} (73.2 day), and W^{187} (24 hr) are formed. Although numerous investigations have been performed, their precise decay schemes have remained undecided. In the present studies, certain aspects of the disintegration of two of these elements have been clarified with utilization of such instruments as a thin-lens beta spectrometer, coincident-scintillation spectrometers, etc. In addition isotopically concentrated W^{184} has been employed as the target material in forming W^{185} so that its radiations could be studied with decreased interference from those of W^{181}.

W^{185}

The radiations of W^{185} were first reported to consist of beta rays[1] at 428 kev[2] and a single gamma ray of energy 133.7 kev.[3] Other investigators[4,5] reported no evidence of any gamma radiation from W^{185}. Subsequent to these latter papers in which W^{185} was said to emit no gamma rays, one at 55.6 kev was stated to be present.[6] Still other measurements have led to the conclusion that gamma rays are present at 57 kev,[7] 570 and 770 kev,[8] and 132 kev.[9] The aforecited data are evidently highly contradictory in character. In order to reinvestigate the radiations of W^{185}, targets of wolfram isotopically concentrated in W^{184} were irradiated by slow neutrons in the Brookhaven reactor.

Spectra of the radiations of W^{185} are shown in Fig. 1. In Fig. 1(a) are presented the beta-ray continuum of W^{185} as detected in an anthracene crystal along with the gamma-ray spectrum of the same source, detected in a crystal of NaI(Tl). Naturally occurring wolfram was the target material. Electromagnetically separated W^{184} (isotopic enrichment 95%) was employed as a target in obtaining the W^{185} of Fig. 1(b). The beta spectrum appears as before, but the quantum radiations are greatly reduced in intensity, showing that these quanta are not related to the decay of W^{185}. In obtaining the data of these two sets of curves, the geometries were identical, and the beta-ray intensities equal. The

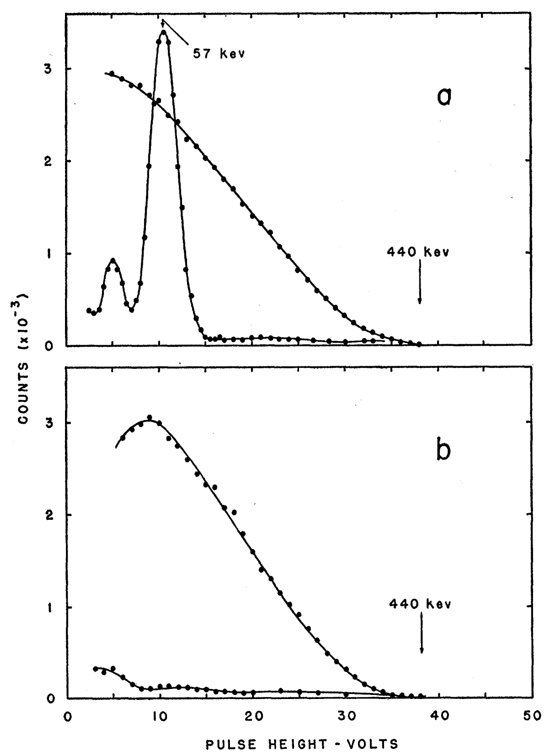

Fig. 1. (a) Beta and gamma spectra of naturally occurring wolfram, aged after irradiation. An energy scale is not indicated for the gamma-ray spectrum. (b) Beta and gamma spectra of an aged sample of wolfram enriched in W^{184}. The material had been irradiated by slow neutrons.

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1 W. H. Sullivan, Phys. Rev. **68**, 277 (1945); D. Saxon, Phys. Rev. **74**, 1264(A) (1948).
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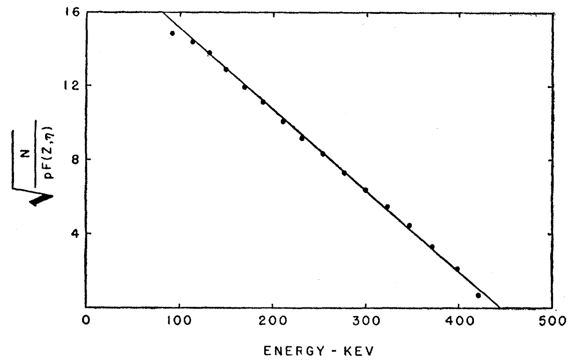

FIG. 2. Fermi-Kurie plot of the beta spectrum of W^{185}.

electromagnetic radiation of Fig. 1(a) is interpreted as x-radiation of Ta, emitted following *K*-capture in W^{181}. A search was made for any gamma rays emitted in the disintegration of W^{185}, and a series of measurements were performed to place an upper limit upon the intensity of any gamma rays present. The source was located before an anthracene beta-ray counter and subsequently before a sodium iodide counter of a scintillation spectrometer. The number of gamma rays per disintegration emitted in any given energy interval could thus be determined. The upper limits of gamma-ray intensities so obtained are given in Table I. Smaller values of these upper limits were unobtainable by counting singles because of the presence of the complex gamma-ray spectrum of Ta^{182} which gave rise to a background widely distributed in energy. Attempted chemical separations of Ta from W did not better background conditions. It is probable that the more part of the counting rates, from which the upper limits of Table I have been calculated, actually arise almost entirely from the presence of Ta^{182} so that the intensities of any gamma rays from W^{185} are in fact much lower. This conclusion is confirmed by the fact that the gamma-ray intensity per disintegration could be estimated in various portions of the gamma-ray spectrum by measuring the beta-gamma coincidence rate. By this latter method it was concluded that the upper limit of intensity of any 57-kev gamma-ray coincident with the beta rays of W^{185} is $(2.4\pm1.2)\times10^{-4}$ γ/disintegration. A conversion coefficient[7] of 3.3 was assumed in making this estimate.

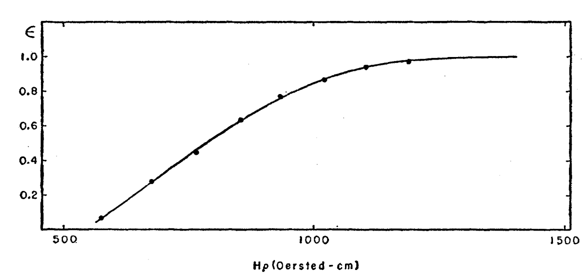

FIG. 3. Detection efficiency of the anthracene beta-ray detector of the lens spectrometer as a function of momentum.

In a similar measurement, it was concluded that any gamma rays coincident with beta rays and of energy greater than this amount could have an intensity of at most $(4.6\pm1.3)\times10^{-4}$ γ/disintegration.

A Fermi-Kurie plot of the beta spectrum of W^{185} is given in Fig. 2. These data were obtained in a thin-lens magnetic spectrometer of resolution 1.8% in momentum, as measured by observing the conversion electrons of the 662-kev gamma ray of Cs^{137}. The end-point energy calculated from the data of Fig. 2 is 440±5 kev. A beta-ray component of end-point energy 370 kev has been previously reported[6,7] with an intensity of 0.10–0.15 per disintegration. No spectrum of this energy or intensity can be inferred from the data of Fig. 2.

The detecting device of this spectrometer was an anthracene crystal of thickness 2 mm, covered by a silver foil of thickness 0.18 mg/cm^2. The phototube was a DuMont 6292 photomultiplier, enclosed in a solenoid designed to provide a compensating field to counteract the effects of the magnetic field of the thin lens itself upon photomultiplier operation. The current of the solenoid was in parallel with the current of the lens so that as the lens current was changed, compensation was automatic. A curve of the detection efficiency of the anthracene counter as a function of $H\rho$ is shown in Fig. 3. This curve was obtained by measuring the known beta spectrum of Co^{60}.

TABLE I. Upper limits of intensities of possible gamma rays from W^{185}.

Energy interval	Upper limit of radiation intensity (gamma ray per beta ray)
33 kev$\leq E_\gamma \leq$ 88 kev	3.5×10^{-3}
88 kev$\leq E_\gamma \leq$154 kev	1.3×10^{-3}
154 kev$\leq E_\gamma \leq \infty$	2.4×10^{-3}

The beta-ray sources were mounted upon thin backings of Nu-skin. Color fringes were often observable across the faces of the sources; so it was estimated that the beta-ray sources were in general of thickness only a few micrograms/cm^2.

W^{187}

The radiations of W^{187} have been investigated in extensive detail over a period of many years by numerous authors. Recent relatively complete studies have been carried out by Cork *et al.*,[10] using a photomagnetic semicircular focusing beta-ray spectrometer, and by Sunyar.[11] Gamma rays have been detected[10] with quantum energies of 72, 106, 113, 134, 206, 239, 246, 480, 513, 619, 625, 686, 774, and 866 kev. An additional gamma ray at 552 kev was also reported.[11] Coincidence studies, determining lifetimes, multipolarities, and cascade relationships, have been made of the gamma-

[10] Cork, Brice, Nester, LeBlanc, and Martin, Phys. Rev. **89**, 1291 (1953).
[11] A. W. Sunyar, Phys. Rev. **90**, 387(A) (1953).

ray spectrum.[11,12] Scintillation counters were employed in these latter measurements. Several measurements[10,13,14] of the beta-ray spectrum have indicated as many as three groups of beta rays, having maximum energies of approximately 1330 kev, 630 kev, and 330 kev.

Fermi-Kurie plots of the presently observed beta-ray spectra of W^{187} are shown in Fig. 4, where the high-energy portion of the curve has been replotted at the upper right as $\{N/[pF(Z,\eta)a_1]\}^{\frac{1}{2}}$, where a_n is given by[15]

$$a_n=\sum_{\nu=0}^{\nu=n}\frac{(2n+1)!(2\nu+1)!}{2^{2\nu}(\nu!)^2(2n-2\nu+1)!}q^{2(n-\nu)}L_\nu(p,\mp Z).$$

The linearity of this plot of the spectrum of highest energy (1325±15 kev) shows it to be in the category of the "unique" forbidden transitions ($\Delta I=2$; yes). The 1225-kev beta spectrum of Mo^{99} was also examined in

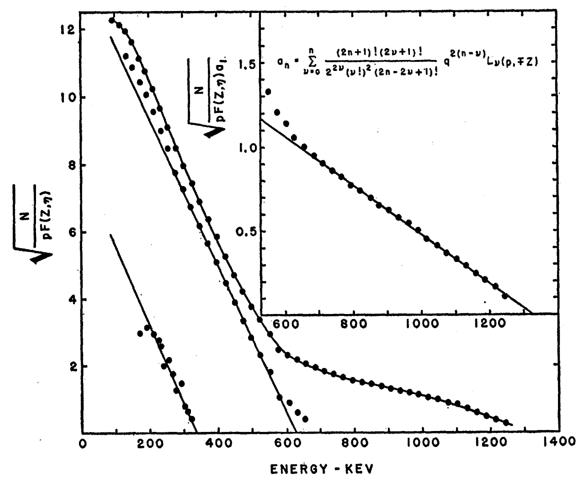

FIG. 4. Fermi-Kurie plots of the beta spectra of W^{187}.

the thin lens spectrometer and was found to have the allowed or more usual first forbidden shape, yielding a straight line on a Fermi-Kurie plot, without introduction of any correction factor, to beta-ray energies as low as 550 kev. This measurement was taken as further evidence of unique shape of the high-energy spectrum of W^{187}. From the data of Fig. 4, it is clear that two inner spectra of lower energies are also present having maximum energies of 630±10 kev and 340±20 kev. Other data pertinent to these spectra are shown in Table II.

The gamma-ray spectrum is depicted by way of internal conversion lines in Figs. 5 and 6. Low-energy

TABLE II. Characteristics of the beta-spectrum of W^{187}.

E_β(max)	Rel. int.	logft
340±20	~10%	6.4
630±10	70%	6.3
1325±15	20%	8.0

conversion lines are shown in Fig. 5, and the high-energy region is depicted in Fig. 6 where a somewhat thicker source was employed. In all, fourteen gamma rays were detected. The data of Figs. 5 and 6 are summarized in

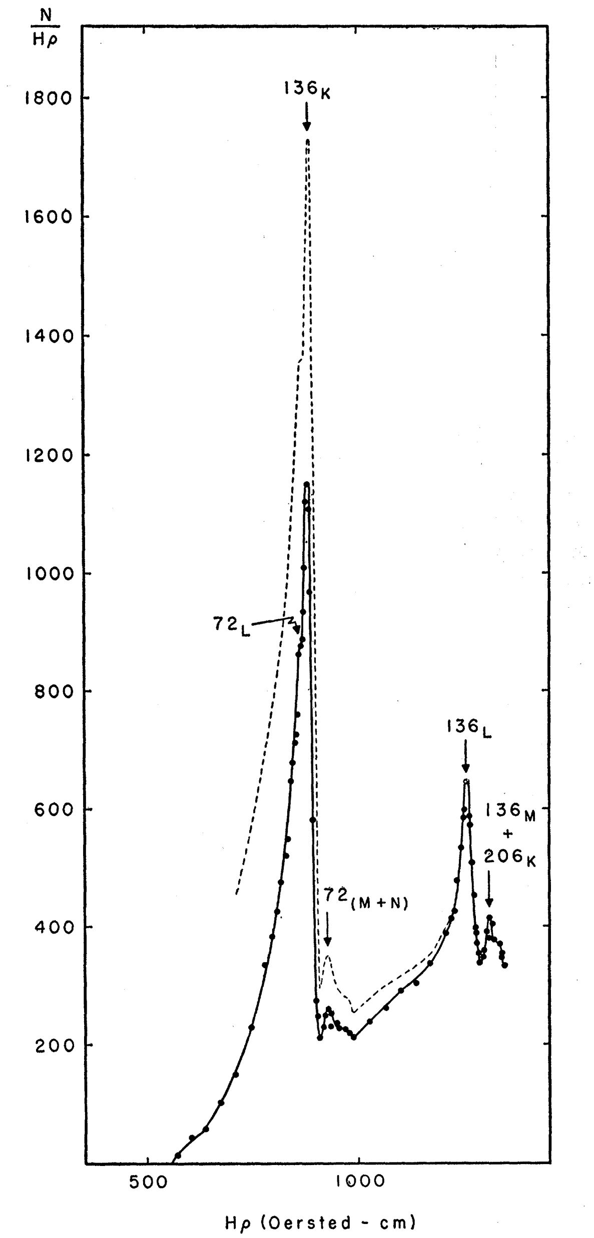

FIG. 5. Internal conversion lines of the soft gamma rays of W^{187}. The correction for the detection efficiency is indicated by the broken line.

[12] Germagnoli, Malvini, and Zappa, Nuovo cimento 10, 1388 (1953).
[13] C. L. Peacock and R. G. Wilkinson, Phys. Rev. 74, 297 (1948).
[14] Hole, Benes, and Hedgran, Arkiv Mat., Astron. Fysik 35A, No. 35 (1948).
[15] E. J. Knopinski, in *Beta- and Gamma-Ray Spectroscopy*, edited by Kai Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. X, p. 292.

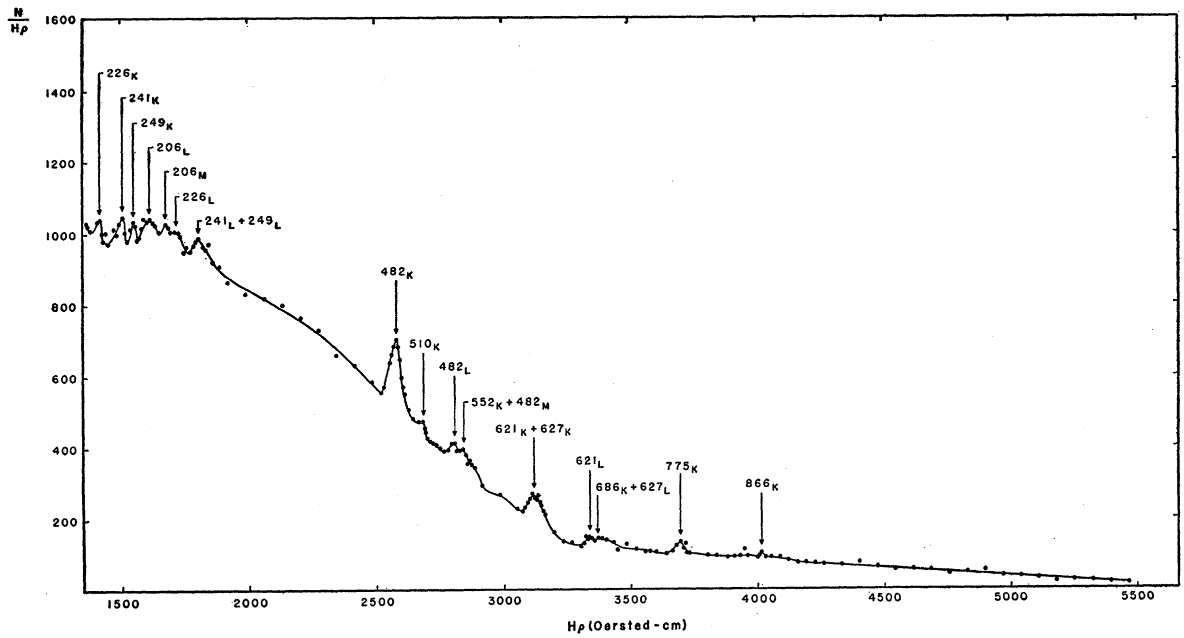

FIG. 6. Internal conversion lines of the harder gamma rays of W^{187}.

Table III. The probable error in kilovolts of the gamma rays can be taken as that of the K-shell conversion line in each case.

Photoelectric lines of the gamma rays of W^{187} were also observed. The source was located inside of a brass capsule of sufficient thickness to absorb the primary beta rays of W^{187}. This capsule was in turn encased in an aluminum cylinder on the bottom of which was placed a radiator of thickness varying from 5 to 10 mg/cm^2 of gold. The spectrum of photoelectron lines is shown in Fig. 7. The data of Fig. 7 are interpreted in Table IV. The photoelectron energies of Table IV have been translated into the gamma-ray energies of Table V. Moreover, from the areas under the photoelectric peaks of Fig. 7, the relative intensities of the unconverted quantum radiations have been calculated. For reasons of energy or intensity or the masking effects of other more intense radiations the relative intensities of the unconverted quanta of the 72- and 206-kev gamma rays have not been calculated. At low energies (e.g., 72 and 136 kev) absorption in the

TABLE III. Conversion lines of the gamma rays of W^{187}.

$H\rho$	Electron energy in kev	Interpretation (gamma-ray energy in kev)	Conversion coefficient ratio
857	60±2	72K	
878	64±2	136K	$K/L=5.0\pm0.5$
928	70	72(M+N)	
1257	124	136L	$L/M=3.5\pm0.5$
1310	134	136M	
1331	137±3	206K	$K/L=4.5\pm0.5$
1412	153±3	226K	
1501	170±3	241K	
1542	177±3	249K	
1607	191	206L	
1670	204	206M	
1712	213	226L	
1798	238	241L+249L	
2570	414±4	482K	$K/L=4.0\pm0.5$
2676	440±4	510K	
2796	471	482L	
2832	480±5	552K+482M	
3110	552±6	621K+627K	
3223	609	621L	$K/L=3.8\pm1$
3346	614±6	686K+627L	
3682	703±5	775K	
4000	794±5	866K	

TABLE IV. Photoelectron lines of the gamma rays of W^{187}.

$H\rho$	Electron energy in kev	Interpretation (gamma-ray energy, kev)
745	52	Auger electrons from Au
822	56	136K+72L
914	69	72M
1242	121	136L+206K
1310	133	136M
1400	150	226K
1460	160	241K
1490	168	249K
1610	192	206L
1710	213	227L
1770	226	241L
1808	234	249L
2520	399	482K
2620	426	510K
2765	463	482L
2800	471	552K
2900	498	510L
3070	542	552L+621K+627K
3310	605	686K+621L
3335	613	627L
3560	672	686L
3640	694	775K
3885	761	775L
3975	785	866K
4215	852	866L

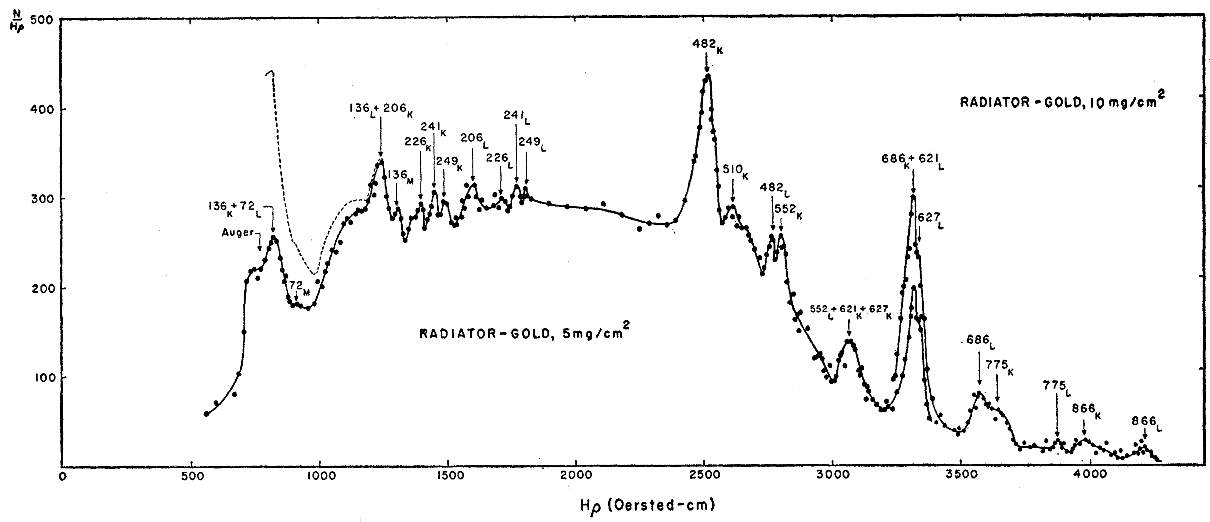

FIG. 7. Photoelectric lines generated in gold by the gamma rays of W^{187}. The correction for the detection efficiency is indicated by the broken line.

converter itself becomes important, and no corrections have been made for this effect in this paper.

Cascade relationships among the various gamma rays were investigated with the use of coincident scintillation spectrometers. The resolving time of the circuit was 0.2 microsecond. Some observed gamma-gamma coincidence rates are shown in Fig. 8. In Fig. 8(a), one channel of the coincidence arrangement was placed so as to count only pulses of the 866-kev gamma ray, while the other channel was permitted to pass through the region of lower energies. The coincidences of Fig. 8(a) show the 866-kev gamma ray to be in cascade with the 136-kev radiation. In this case, gamma-(x-ray) coincidences are also observed, because the 136-kev gamma ray is to some extent converted. In Fig. 8(b) are recorded coincidences between the 482-kev gamma ray and the 136-kev gamma ray, the 72-kev gamma, and x-rays. That the 866-kev gamma ray is noncoincident with the 72-kev gamma ray is indicated by the smaller ratio of the peak at ~60 kev to the 136-kev peak, in Fig. 8(a). Coincidences were also noted between the 482-kev gamma ray and radiation near 210 kev. The 686-kev gamma ray was found to be noncoincident with gamma radiation of all energies. This gamma ray was found to be coincident with a beta spectrum of maximum energy about 630 kev. Several supplementary experiments with a gray-wedge analyzer were carried out. Gamma radiation of a mixture of energies of 750 kev or greater was found to be coincident with the 72- and 134-kev quanta.

TABLE V. Energies and relative intensities of some of the unconverted quanta of W^{187}.

Gamma-ray energy in kev	Relative intensity[a] of the unconverted quanta
136	45[b]
224	2
241	4
249	2
482	100
510	12
552	40
621	30
627	15
686	185
775	20
866	12

[a] Obtained from areas under the photopeaks of Fig. 7, and from photoelectric absorption coefficients associated with article by C. M. Davisson, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. II, p. 24.
[b] Taken from reference 11.

DISCUSSION OF RESULTS

The foregoing measurements can be summarized in decay schemes of W^{185} and W^{187} shown in Figs. 9 and 10. Since no gamma rays were actually detected in the decay of W^{185}, only upper limits of intensities having been assigned, its decay scheme can be best represented by a single beta transition (see Fig. 9). The ground-state spin of Re^{185} has been measured[16] as $\frac{5}{2}$, and the

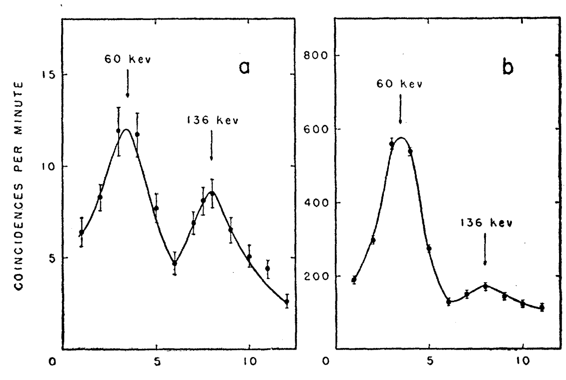

FIG. 8. (a) Coincidences between the 860-kev gamma ray and the 136-kev gamma ray. (b) Coincidences between the 480-kev gamma ray and gamma rays at 136 and 72 kev and x-rays.

[16] H. Schuler and H. Korsching, Z. Physik **104**, 468 (1937).

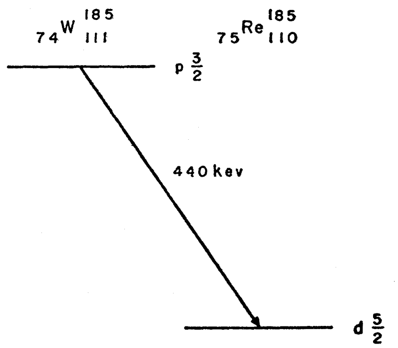

FIG. 9. Disintegration scheme of W^{185}.

beta spectrum of W^{185} is first-forbidden, suggesting a spin of $\frac{3}{2}$ for the ground state of W^{185}. Shell-model indications are that the ground-state orbitals of W^{185} and Re^{185} are respectively ($p_{\frac{1}{2}}, p_{\frac{3}{2}}$) and $d_{\frac{5}{2}}$. A first-forbidden spectrum is usually interpreted as being associated with a spin change of zero or one unit, unless it exhibits the unique shape. Such a shape is not found in the Fermi-Kurie plot of Fig. 2. When these data were distorted by the factor $(a_1)^{\frac{1}{2}}$, a curve differing markedly from the observed plot of Fig. 2 was obtained. Thus, it can be concluded that a spin change of at most one unit is involved so that an orbital value of $p_{\frac{3}{2}}$ is predicted for the ground state of W^{185}.

The ground state spin of Re^{187} has also been measured[16] as $\frac{5}{2}$, and the nature of the ground state transition is in accord with a spin assignment of $\frac{1}{2}$ for the ground state of W^{187}. Possibilities suggested by shell model considerations indicate orbitals of $p_{\frac{1}{2}}$ and $d_{\frac{5}{2}}$ for the ground states of W^{187} and Re^{187}. The measured[17] ground-state spin of W^{183} is $\frac{1}{2}$. That the spin of W^{187} is also $\frac{1}{2}$ is consistent with this measurement. However, the spin value of $\frac{3}{2}$ assigned to W^{185} is somewhat at variance with the simplest possibility presented by shell-model considerations.

For the first-forbidden nature of the beta spectrum of maximum energy 630 kev, the spin of the 686-kev level can be taken as $\frac{1}{2}+$ or $\frac{3}{2}+$. The energy levels indicated for Re^{187} in Fig. 10 have been ordered to best conform to the energy measurements and coincidence studies previously described.

From a study of its L_{III}/L_I ratio, Cork *et al.*[10] have concluded that the 136-kev gamma ray is emitted in a transition which is $M1$ or $M1+E2$ in character, whereas Goldhaber and Sunyar[18] have constructed an empirical curve for K/L ratios of $M2$ transitions which is based upon the assumption that the 136-kev gamma ray is emitted in a magnetic transition of multipole order two. In the present investigation, the measured K/L ratio for the 136-kev gamma ray of 5.0 ± 0.5 has been compared with values theoretically obtained.[19] The theoretically calculated[19] value of the K/L ratio for an $M1$ transition of this energy is 4.8 in close agreement with the experimental value so that the 136-kev transition may be regarded as predominantly $M1$. From similar considerations of K/L ratios, the multipolarities of the 206-, 482-, and 621-kev gamma-ray transitions can be adjudged $M1+E2$, $E2$, and $E2$ respectively. However, as pointed out by Rose,[20] K/L ratios may not be conclusive in determining the multipole orders of transitions in heavy nuclei.

The 136-kev level in Re^{187} has been identified in Coulomb excitation experiments[21] as the first excited rotational level belonging to the ground-state band ($K=\frac{5}{2}$) and therefore may be assigned a spin of $\frac{7}{2}+$. From the systematics[22] of energy levels of isomers of

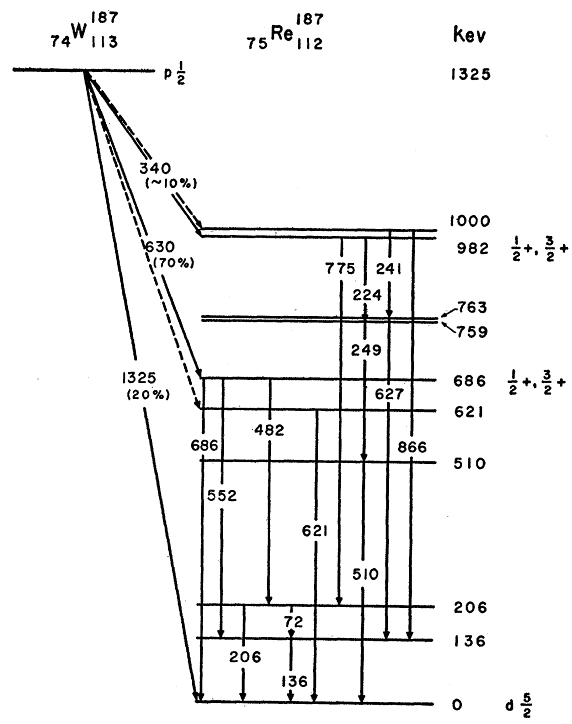

FIG. 10. Disintegration scheme of W^{187}.

odd isotopes of gold, the separation of the levels having orbitals $d_{\frac{5}{2}}$ and $d_{\frac{3}{2}}$ for a neutron number of 112 is expected to be a bit more than 200 kev. Accordingly, the orbital of the 206-kev level in Re^{187} may be taken as $d_{\frac{3}{2}}$.

ACKNOWLEDGMENT

The writers desire to acknowledge the kind interest of Dr. W. F. G. Swann, Director of the Bartol Research Foundation.

[17] H. Kopfermann and D. Meyer, Z. Physik **124**, 685 (1948).
[18] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[19] Rose, Goertzel, and Swift, "Table of Conversion Coefficients" (privately circulated).
[20] M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XIV, p. 396.
[21] Davis, Divatia, Lind, and Moffat, Phys. Rev. **103**, 1801 (1956).
[22] J. W. Mihelich and A. de-Shalit, Phys. Rev. **93**, 135 (1953).