Relative Intensities of the Radiations from Hf^{175}†

A. O. BURFORD, J. F. PERKINS,* AND S. K. HAYNES
Vanderbilt University, Nashville, Tennessee

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The intensities of the electronic and electromagnetic radiations of Hf^{175} have been measured with a magnetic-lens spectrometer. The existence of transitions of 89.1, 113.4, 228, 318, 342.3, and 431 kev was confirmed. For the 89.1-kev transition $\alpha_K=3.5_{-2.5}^{+1.5}$, $\alpha_K/\alpha_L=6.0\pm1$, $\alpha_L/\alpha_N=3.5\pm0.9$, and the transition is predominantly $M1$. For the 228-kev transition $\alpha_K/\alpha_{L+M}=2.0\pm0.5$ and the transition is of the type $E2$. For the 342.3-kev transition $\alpha_K/\alpha_{L+M}=4.94\pm0.5$, and the transition is a mixture of $M1$ and $E2$. A decay scheme is given in which the ground state of Lu^{175} is characterized as $\Omega=7/2$, $I=7/2$, parity even; the 113.4-kev state is the first rotational excited state with $\Omega=7/2$, $I=9/2$, parity even; the 342.3-kev state is a single-particle excited state with $\Omega=5/2$, $I=5/2$, parity even; and the 431-kev state is the first rotational excitation of the 342.3-kev state with $\Omega=5/2$, $I=7/2$, parity even.

I. INTRODUCTION

HAFNIUM-175 was discovered in 1949 by Wilkinson and Hicks[1] as a product of the reactions $Lu^{175}(p,n)Hf^{175}$ and $Lu^{175}(d,2n)Hf^{175}$. The half-life was measured and found to be 70 ± 2 days. They showed the presence of strong Lu K x-rays which indicated K capture and found a 350-kev gamma ray with a conversion coefficient of approximately 0.4. A 1.5-Mev gamma ray reported by these observers has not been found by others including ourselves. The assignment of the activity to Hf^{175} was confirmed by Hedgran and Thulin[2] and by Burson *et al.*[3] who produced the activity by neutron capture in separated isotopes of Hf. The conversion lines have been studied by Cork *et al.*[4] who discovered the K, L, and M lines of 89.1- and 342.3-kev gamma rays, by Burson *et al.*[3] who confirmed these lines and found conversion lines from gamma rays of 113.4 and 228.4 kev, and by Burson and Rutledge[5,6] who found lines from gamma rays of 318 and 431 kev and who measured the K/L conversion ratios of the four strongest lines to be ~3 for 89.1 kev, ~10 for 113.4 kev, ~2 for 228.4 kev, and 4.93 ± 0.20 for 342.3 kev. Later Bashilev *et al.*[7] obtained an $\alpha_K:\alpha_L:\alpha_M$ of 30:15:1.5 for the 89.1-kev transition and 100:20:5 for the 342.3-kev transition.

The studies reported here were designed as far as possible to provide more information on the relative intensities (a) of the electronic radiations by use of a magnetic-lens spectrometer, and (b) of the gamma radiations by measurement of the photoelectrons in the magnetic-lens spectrometer.

II. SOURCES

The Hf used was 20 mg of separated Hf^{174} obtained from the Isotopes Division, Oak Ridge, Tennessee, in the form of HfO_2. The analysis showed Hf^{174}, 7.85 percent; Hf^{176}, 16.70 percent; Hf^{177}, 27.29 percent; Hf^{178}, 24.07 percent; Hf^{179}, 8.24 percent; Hf^{180}, 15.84 percent, with impurities of a few hundredths of a percent of Cu, Mg, Na, and Si.

The sample was irradiated for one month in the Oak Ridge reactor during January 1953. No chemical purification was carried out with the result that certain weak impurity effects were encountered in the spectra. The material was dissolved in HF and a drop was placed on a tight 20-mil tungsten-wire coil in the evaporation apparatus previously described.[8] The wire was heated to a bright white heat for about 30 minutes. This operation was carried out three times. The resulting source was about 8 mm in diameter on a 120-$\mu g/cm^2$ aluminum foil and had a thickness of less than 50 $\mu g/cm^2$. This source was used for the electron spectrum in the magnetic spectrometer.

The remainder of the activity was enclosed in an Al capsule $\frac{3}{16}$ in. in diameter with 0.0158 in. of Al covering the active material as an electron absorber. This source was much stronger than the other and was used for the photoelectron spectrum of Hf^{175}. The converters were stuck on the end of the capsule with a small amount of very dilute cement.

III. ELECTRON SPECTRUM OF Hf^{175}

The electron spectrum of the thin source of Hf^{175} was run completely with the magnetic-lens spectrometer[8] with a window of 26 μg cm^{-2} from 10 to 500 kev in June, 1953 and again in September, 1953. The average interval between the spectra was 105 days. Certain peaks were run again in April, 1954, about 180 days after the September run. By comparison it was possible to identify and subtract off the continuous beta-ray spectrum of the weak Hf^{181} impurity. The remainder of the electron spectrum as of September, 1953 is shown in Fig. 1. The identification of the various lines shown in

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* U. S. Atomic Energy Commission Predoctoral Fellow. Present address: Lockheed Aircraft Corporation, Smyrna, Georgia.

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[7] Bashilev, Anton'eva, Dzhelepov, and Dolgentseva, Isvest. Akad. Naut. Ser. Fiz. U.S.S.R. **17**, 437 (1953).
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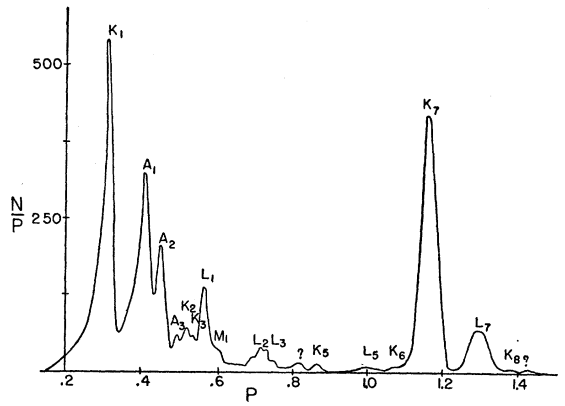

FIG. 1. Electron spectrum of Hf^{175} after subtraction of the continuous beta-ray spectrum of the Hf^{181} impurity.

the figure together with their relative intensities is given on the left-hand side of Table I.

All of the peaks assigned to Lu^{175} had a half-life of 70 days within experimental error except the Auger peaks. The peaks assigned to Hf^{181} had a half-life which agreed with Hf^{181} within experimental error. Two small unidentified peaks were also found. The one at $p=0.8$ did not appear in the June data. Further evidence of impurities was the failure of the Auger peaks to decay with the proper half-life. Between June and September these peaks decayed too fast, while between September and April they decayed too slowly. This difficulty combined with the fact that the K-conversion line of the 113-kev gamma ray should be under the A_2 peak makes the error in Auger intensity considerably larger than the errors in the determination of the areas under the peaks which are shown in the table.

From the third column of the table, the following important ratios can be obtained. For the 342.3-kev transition $\alpha_K/\alpha_{L+M}=4.94\pm0.5$; for the 228.4-kev transition, it is 2 ± 0.5; and for the 89.1-kev transition, α_K/α_L is 6.0 ± 1 and α_L/α_M is 3.5 ± 0.9.

The first of these ratios is about the same as that which Burson and Rutledge[5] and Bashilov[7] give for α_K/α_L. Although the difference is not great, we feel that it is greater than our experimental error. We estimate that our value of α_K/α_{L+M} is consistent with a value of about 6 for α_K/α_L.

The value of α_K/α_{L+M} for the 228.4-kev transition is in excellent agreement with Burson and Rutledge but the α_K/α_L ratio for the 89.1-kev transition is double their value and triple the value of Bashilov *et al.*[7] Both because Burson's value was obtained photographically over an energy range of 3 to 1 and because most of our errors would tend to make our value too small, we shall assume that the α_K/α_L ratio of 6 is substantially correct.

IV. PHOTOELECTRON SPECTRUM OF Hf^{175}

The spectrum of the strong source enclosed in an aluminum capsule was observed five times during the summer of 1953 in the following order; no converter, 1.53-mg cm^{-2} Au converter, no converter, 0.88-mg cm^{-2} Au converter, no converter. The no converter runs combined with the other runs between photoelectron lines made it possible to determine the Compton electron intensity under the photoelectron peaks. The three no-converter runs also helped identify parts of the spectrum which did not have the 70-day half-life of Hf^{175}. Figure 2 shows the spectrum obtained with the 0.88-mg cm^{-2} converter together with the Compton background under certain peaks. The 0.88 mg cm^{-2} was preferable for analysis both because, occurring later, there was less short lived impurity and also because it was desirable to have a converter which was reasonably thin for L and M photoelectrons from the 89.1- and 113-kev gamma rays.

The interpretation of Fig. 2 and the relative gamma-ray intensities are given on the right-hand side of

TABLE I. Magnetic spectrometer values for the electron and gamma-ray intensities.

Electron spectrum				Photoelectron spectrum		
Peak	Energy of gamma ray kev	Relative intensity	Nuclide	Peak	Energy of gamma ray kev	Relative intensity
K_1	89.1	10.0±5%	Lu^{175}	...	...	...
A_1	Lu $K-LL$	6.26±10%	Lu^{175}	$L_{K\alpha}$	Lu K_α	
A_2	Lu $K-LX$	2.92±10%	Lu^{175}	$L_{K\beta_1}M_{K\alpha}$	Lu $K_\alpha K_\beta$	61.1
A_3	Lu $K-XY$	0.61±25%	Lu^{175}	$M_{K\beta}$	Lu K_β	
K_2	133	...	Ta^{181}	...	...	...
K_3	136	...	Ta^{181}	...	...	...
L_1	89.1	1.67±15%	Lu^{175}	L_1	89.1	2.83±20%
M_1	89.1	0.5±25%	Lu^{175}	M_1	89.1	
L_2	133	...	Ta^{181}	...	...	...
L_3	136	...	Ta^{181}	...	...	...
...	...	...	Lu^{175}	K_4	113.4	...
...	...	...	Lu^{175}	L_4	113.4	0.46±30%
K_5	228.4	0.18±25%	Lu^{175}	...	...	...
L_5	228.4	0.088±50%	Lu^{175}	...	...	...
K_6	318	0.1±100%	Lu^{175}	...	...	...
K_7	342.3	8.5±2.5%	Lu^{175}	K_7	342.3	...
L_7	342.3	1.72±10%	Lu^{175}	L_7	342.3	89.8±8%
K_8	430	0.07±50%	Lu^{175}	...	...	...

Table I. The relative intensities were obtained by dividing the $L+M$ (or L) photoline areas by their photoelectric cross sections *relative* to that of the 342.3-kev gamma ray. These relative cross sections were assumed to be the same as the relative K photoelectron cross sections for the same binding energy. Thus we used the average of Heitler's[9] *relative* values for Al and Sn which agreed closely with each other and whose average K binding energy approximates the L binding energy of Au.

The most surprising result of the photoelectron spectrum is that the 113-kev gamma ray is so strong when it was completely undetectable in the conversion line spectrum. It seems clear that the conversion coefficient of this gamma ray is much smaller than that of the 89.1-kev gamma ray.

V. DISCUSSION AND DECAY SCHEME

A. Relative Gamma-Ray and Electron Intensities

In order to relate the relative electron and gamma-ray intensities, it is necessary to use either the known conversion coefficient of one of the gamma rays or the fluorescence yield of lutecium to relate the x-ray and Auger electron intensities.

The only conversion coefficients available are for the 342.3-kev line. McGowan[10] (0.095 ± 0.015) and Kerr[11] (~0.118) have reported preliminary rough measurements. The experimental α_K/α_L ratios discussed in Sec. III, although not in agreement, seem to lie between 5 and 6. When combined with the theoretical values of α_K[12] and the various α_L,[13] these values seem to indicate

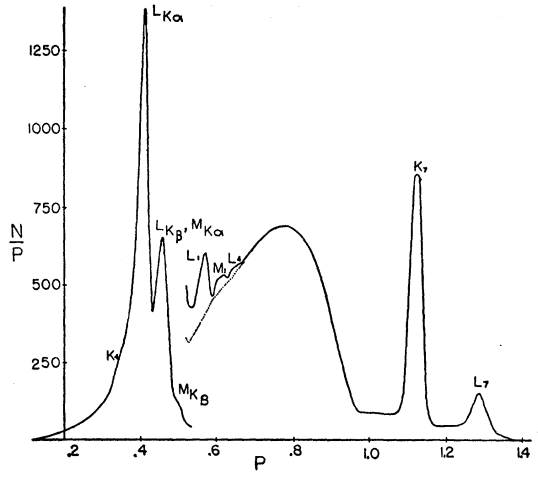

FIG. 2. Photoelectron spectrum of Hf^{175} from a 0.88-mg cm^{-2} radiator. The photoelectron peaks arising from the K x-rays are reduced in ordinate by a factor of 10.

[9] W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1949), second edition, p. 124.
[10] F. K. McGowan (private communication).
[11] R. J. Kerr, Ph.D. thesis, Vanderbilt University, 1954 (unpublished).
[12] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL, 1023, 1951 (unpublished).
[13] M. E. Rose (privately circulated values).

TABLE II. Summary of data for transitions resulting from the decay of Hf^{175}.

Transition kev	Total intensity percent	α_K measured	α_K/α_L BPH[a]	α_K/α_L BR[b]	α_K/α_L B[c]
89.1	$15.0^{+4}_{-2.5}$	$3.5^{+1.5}_{-2.5}$	6.0 ± 1	~3	2
113.4	~1	<1		~10	
228	~1		2.0 ± 0.5[d]	~2	
318	<1				
342.3	100	0.095 ± 0.015[e]	4.94 ± 0.5[d]	4.93 ± 0.2	5.0
430	~1				

[a] BPH refers to the present paper.
[b] BR refers to references 5 and 6.
[c] B refers to reference 7.
[d] Indicates α_K/α_{L+M} ratio.
[e] Reference 10.

a mixed $M1$, $E2$ transition with the proportion of $M1$ being between 49 and 79 percent and the K-converstion coefficient between 0.079 and 0.104. It would seem that McGowan's value is not far from the mean of these values and that his uncertainty represents pretty well the probable uncertainty of the mean of these values. In the following discussion McGowan's value has been used. The intensity values in Table I have been normalized using this value to make the total intensity of the 342.3-kev transition 100 percent.

In principle, the electron and photoelectron intensities could also be related by means of the K x-ray photoelectrons and the K-Auger electrons, by use of the fluorescence yield for Lu which is calculated to be 0.9326 ± 0.003.[14] In Table I, the total number of empty K shells produced per disintegration comes out 1.45 from the Auger intensity and 0.650 from the x-ray intensity. Even with allowance for the uncertainty of the conversion coefficient of the Auger intensity, and the fluorescence yield, it is evident that the x-ray value is too low by a factor of about two. This discrepancy is probably due to the fact that the 0.88-mg cm^{-2} converter is not really thin for the 40- to 50-kev photoelectrons. Thus a substantial percentage of the photoelectrons produced are scattered out of the acceptance cone of the spectrometer by the converter. Another phenomenon which may contribute to this discrepancy is the change with energy of the mean angle of emission of the photoelectrons.[15] These phenomena undoubtedly occur to a lesser extent for the 76-, 86-, and 100-kev photoelectron peaks of the 89.1- and 113-kev gamma rays. Hence the intensities of these gamma rays given in Table II should be considered as lower limits. The 89.1-kev gamma-ray intensity may be as much as 50 percent greater and the 113-kev intensity as much as 25 percent greater than these given in the table.

Taking into account these uncertainties in gamma-ray intensities and the limits of detection of the photoelec-

[14] E. H. S. Burhop, Colloque du Centre National de la Recherche Scientifique, "Le Rôle du Cortège Electronique dans les Phénomènes Radioactifs," June, 1954. These results are to appear in J. phys. et radium.
[15] J. J. Murray, PhD thesis, California Institute of Technology, 1954 (unpublished).

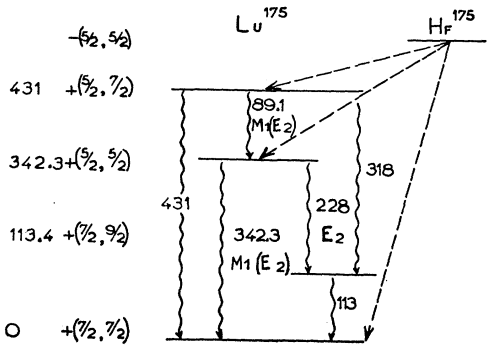

FIG. 3. Decay scheme of Hf^{175}. All energies are in kev. The parentheses opposite the states indicate the values of (Ω,I) where I is the total spin of the state and Ω is the component of the total spin along the axis of symmetry of the nucleus. The existence of capture disintegrations direct to the ground state of Lu^{175} is uncertain.

trons and conversion electrons which were not found, we get the total intensities and conversion coefficients given in Table II. This table also includes the previously discussed conversion ratios.

B. Decay Scheme

A proposed decay scheme for Hf^{175} is shown in Fig. 3.[16] The measured spin of the ground state of Lu^{175} is 7/2.[17] Since the 342.3-kev transition is in part $M1$, the 342.3-kev state must have a spin of 5/2, 7/2, or 9/2 with the same parity as the ground state. No 9/2 single-particle state exists in the subshell containing the 71st proton. Therefore if the spin were 9/2 the 342.3-kev state would have to be interpreted as a rotational excited state.[18] It is the 113.4-kev state, however, which is produced by Coulomb excitation[19] and to which the spin of 9/2 should be assigned.[16] Hence, 9/2 is eliminated as a possible value of the spin of the 342.3-kev state.

Our value for the ratio α_K/α_L for the 89.1-kev transition clearly indicates that it is also predominantly $M1$ by comparison with the curves of Goldhaber and Sunyar.[20] The conversion coefficient is also consistent with this interpretation. We estimate by extrapolating the curves of Rose *et al.*[12] from 150 kev to 89.1 kev that the theoretical K-conversion coefficient is between 4 and 5 for a pure $M1$ transition as compared with our value of 3.5 in Table II. The spin of the 431-kev state must therefore be 3/2, 5/2, 7/2, or 9/2.

A spin of 3/2 is excluded[21] because the 318-kev transition would have a spin change of 3 and would be unobservable. A spin of 5/2 is excluded because the 431-kev gamma ray would be more intense than the 89.1-kev gamma ray. Spins of 7/2 for both the 342.3- and the 431-kev states are excluded by the same reasoning applied to both the 431- and the 318-kev gamma rays. A spin of 9/2 is excluded for the same reason as for the 342.3-kev state.

If the spin of the 342.3-kev state is 5/2, the 431-kev state *can* have a spin of 7/2 as the first rotational level ($\Omega=5/2$, $I=7/2$)[18] of the 342.3-kev state. The 89.1-kev transition, being rotational, will have a much higher intensity[16] than the higher-energy 431- and 318-kev single-particle transitions of similar multipolarity. As a check on this interpretation, the energy of the transition between these two rotational states can be computed on the assumption that the effective moment of inertia of the nucleus is the same in the 342.3-kev state as in the ground state. The computed energy is 113.4 kev $(28/36)=88.2$ kev as compared with the experimental value of 89.1 kev.

The above assignment of spins makes the 228-kev radiation virtually pure $E2$ ($M3$ and $E4$ radiation being neglected). The experimental ratio $\alpha_K/\alpha_{L+M}=2.0\pm0.5$ is good agreement with the empirical curve of Goldhaber and Sunyar[20] for $E2$ radiation. The intensity of the K-conversion line of the 228-kev radiation is also about what would be expected for an $E2$ transition if about 20 percent of the 342.3-kev transitions are assumed to be $E2$ as indicated by our value of α_K for the latter. The 113.4-, 318- and 431-kev transitions are probably all mixtures of $M1$ and $E2$ radiation but there is little experimental evidence. Burson and Rutledge's[6] value of 10 for α_K/α_L for the 113.4-kev transition suggests that it may be predominantly $M1$.

Inaccuracies in the Auger peaks as well as in the conversion coefficients make any estimate of the intensity of capture transitions direct to the ground state of little value. It is probable that the spin of the ground state of Hf^{175} is 5/2 in order to have many transitions to states of spin 5/2 and 7/2 but virtually none to the 113/kev state of spin 9/2. A spin of 3/2 for Hf^{175} cannot be excluded however.

VI. ACKNOWLEDGMENTS

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[16] The rotational aspects of this decay scheme were suggested to us by B. R. Mottelson in a private communication.

[17] H. Schuler and T. Schmidt, Naturwiss. **22**, 714 (1934).

[18] A. Bohr, thesis, Institute for Theoretical Physics of the University of Copenhagen, *Rotational States of Atomic Nuclei* (Ejnar Munksgaards Forlag, Copenhagen, 1954).

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[21] This corrects an incorrect assignment made in a preliminary report of part of this work [Burford, Perkins, and Haynes, Phys. Rev. **95**, 303 (1954)].