

Radiations from Hf¹⁸¹

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The radiations from Hf¹⁸¹ have been examined with a magnetic lens spectrometer. Four gamma-rays were found having energies of 0.087, 0.134, 0.347, and 0.485 Mev. There is evidence for the existence of a fifth gamma-ray. The maximum energy of the beta-rays was estimated to be 0.41 Mev. The three higher energy gamma-rays were found to have intensities proportional to 11, 29, and 100, respectively. The total internal conversion coefficient for the 0.134-Mev transition was calculated to be 11. Calculations on the lifetime and the ratio of the conversion electrons in the *K* and *L* shells indicate that in the transition from the metastable state the nucleus undergoes a spin change of three units.

I. INTRODUCTION

A NUMBER of authors¹⁻⁶ have examined the radiations from Hf¹⁸¹ by means of absorption and coincidence techniques. In addition, Cork, Shreffler, and Fowler⁷ have examined the internal conversion spectrum with a 180° spectrograph. They also looked for higher energy gammas by absorption in copper and aluminum. Benes, Ghosh, Hedgran, and Hole⁸ and Chu and Wiedenbeck⁹ have reported on the radiations as found with a magnetic lens and semicircular spectrometer, respectively.

The results reported in this paper are those obtained by examining the radiations from Hf¹⁸¹ by means of a

thin magnetic lens spectrometer previously described,¹⁰ and provide evidence for an additional gamma-ray that has not been reported previously. In addition, calculations have been made on the intensity ratios of the gamma-rays, the total internal conversion coefficient for one of the gamma-rays, and the spin change of the nucleus in the transition from the metastable state as predicted from the lifetime and the ratio of the conversion electrons in the *K* and *L* shells. The spectrometer was calibrated by means of the *F* conversion line of ThB (1385Hρ) and the annihilation radiation from Zn⁶⁵. The resolution of the spectrometer was 2.1 percent (half-width).

II. PHOTOELECTRON SPECTRUM

The composite spectrum of photoelectrons and Compton electrons ejected from a lead foil of surface density 37.0 mg/cm² is shown in Fig. 1. The 46-day¹¹ Hf¹⁸¹ was obtained by neutron bombardment of HfO₂ in the Clinton pile. The Hf¹⁸¹ was placed in a Lucite holder and covered with an aluminum cap of sufficient thickness to absorb all electrons emitted. A lead foil having a surface density of 37.0 mg/cm² was fastened to the aluminum cap. Curve 1 of Fig. 1 was obtained with a Geiger counter having a mica window of surface density 4.0 mg/cm², while curve 2 was obtained with a mica counter window of surface density 1.1 mg/cm² and was run 49 days later than curve 1. The curves have not been corrected for window absorption.

The gamma-ray energies calculated from the data shown in Fig. 1 are given in Table I as conversions in lead.

III. INTERNAL CONVERSION SPECTRUM

The composite spectrum of internal conversion electrons and beta-rays from Hf¹⁸¹ is shown in Fig. 2. This curve has not been corrected for window absorption. The source had a surface density of about 15 mg/cm² and was mounted on mica of surface density 2.8 mg/cm². This is too large a surface density for accurate beta-ray work, but is of no particular disadvantage in

TABLE I. Gamma-ray energies of Hf¹⁸¹.

| Gamma-ray | Electron energy (corrected) Mev | Conversion shell | Gamma-ray energy Mev | Relative weight | Average energy Mev |
|-----------|---------------------------------|------------------|----------------------|-----------------|--------------------|
| 1 | 0.0708 | Pb(<i>L</i>) | 0.0866 | 5 | 0.087 |
| 2 | 0.0458 | Pb(<i>K</i>) | 0.1334 | 10 | 0.134 |
| | 0.1206 | Pb(<i>L</i>) | 0.1364 | 10 | |
| | 0.1310 | Pb(<i>M</i>) | 0.1348 | 5 | |
| | 0.0641 | Ta(<i>K</i>) | 0.1315 | 10 | |
| | 0.1227 | Ta(<i>L</i>) | 0.1344 | 10 | |
| 4 | 0.2600 | Pb(<i>K</i>) | 0.3476 | 10 | 0.347 |
| | 0.3335 | Pb(<i>L</i>) | 0.3493 | 5 | |
| | 0.2794 | Ta(<i>K</i>) | 0.3468 | 10 | |
| | 0.3300 | Ta(<i>L</i>) | 0.3417 | 1 | |
| 5 | 0.3975 | Pb(<i>K</i>) | 0.4851 | 10 | 0.485 |
| | 0.4706 | Pb(<i>L</i>) | 0.4864 | 5 | |
| | 0.4158 | Ta(<i>K</i>) | 0.4832 | 10 | |
| | 0.4730 | Ta(<i>L</i>) | 0.4847 | 5 | |

* Work was performed at the Ames Laboratory of the AEC.

¹ H. Neuert, *Zeits. f. Naturforschung* **2a**, 432 (1947).² L. Madansky and M. L. Wiedenbeck, *Phys. Rev.* **72**, 185 (1947).³ A. F. Voigt and B. J. Thamer, *AECD 2083 and Phys. Rev.* **74**, 1264 (1948).⁴ S. DeBenedetti and F. K. McGowan, *Phys. Rev.* **74**, 728 (1948).⁵ Bunyan, Lundby, Ward, and Walker, *Proc. Phys. Soc. London* **61**, 300 (1948).⁶ Mandeville, Scherb, and Keighton, *Phys. Rev.* **75**, 221 (1949).⁷ Cork, Shreffler, and Fowler, *Phys. Rev.* **72**, 1209 (1947).⁸ Benes, Ghosh, Hedgran, and Hole, *Nature* **162**, 261 (1948).⁹ K. Y. Chu and M. L. Wiedenbeck, *Phys. Rev.* **75**, 226 (1949).¹⁰ Jensen, Laslett, and Pratt, *Phys. Rev.* **75**, 458 (1949).¹¹ Seren, Friedlander, and Turkel, *Phys. Rev.* **72**, 888 (1947).

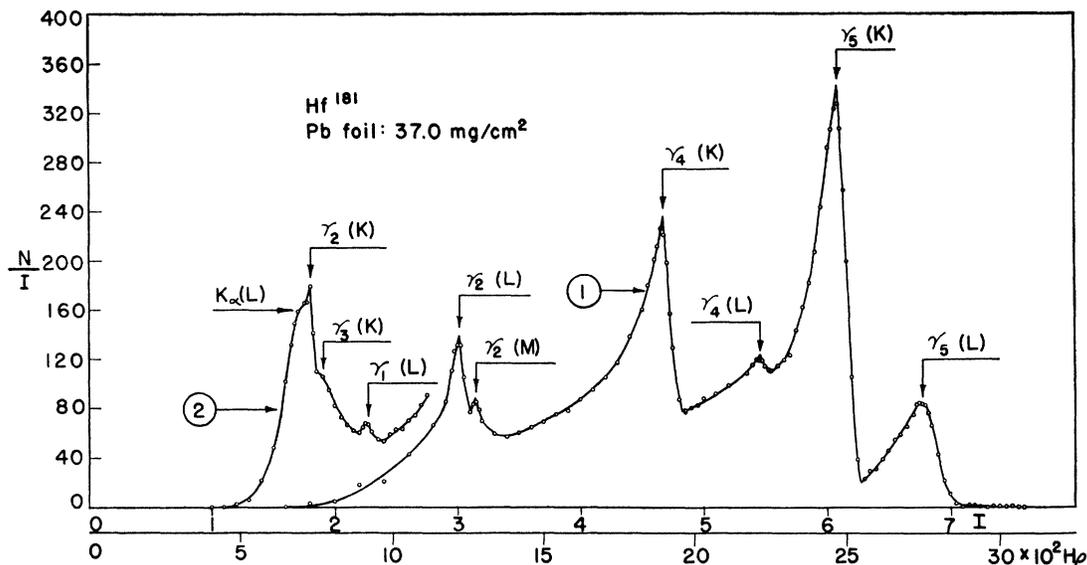


Fig. 1. Composite spectrum of photoelectrons and Compton electrons ejected from 37.0 mg/cm² lead foil by the gamma-rays from Hf^{181} . N is the counting rate in counts/min. and I is the current in the spectrometer coil.

determining gamma-ray energies from internal conversion lines if a correction is applied for the thickness of the source.¹⁰ There is a distinct advantage in a thick conversion source since the intensity is increased appreciably. The counter window was a thin film of Formvar of surface density about 0.3 mg/cm².

The gamma-ray energies calculated from the data shown in Fig. 2 are given in Table I as conversions in Ta, since Hf^{181} decays by beta-emission.

In calculating the gamma-ray energies given in Table I, the data were corrected for the earth's magnetic field, the surface density of the source and the resolution of the spectrometer as explained in reference 10. The probable error is estimated to be 1 percent.

The maximum energy of the beta-rays, as determined from Fig. 2, is estimated to be 0.41 ± 0.02 Mev.

IV. DISCUSSION

A. Energies of Gamma-Rays

The gamma-ray energies listed for γ_2 , γ_4 , and γ_5 in Table I are in fairly good agreement with those given by Benes, Ghosh, Hedgran, and Hole⁸ (0.128, 0.342, and 0.472 Mev), Cork, Shreffler, and Fowler⁷ (0.132, 0.344, and 0.479 Mev), and three of the four gamma-rays reported by Chu and Wiedenbeck⁹ (0.130, 0.337 and 0.471 Mev). Chu and Wiedenbeck report a fourth gamma-ray with an energy of 0.134 Mev, which was observed in their internal conversion spectrum. This gamma-ray was not observed in the internal conversion spectrum of this investigation, but the hump labeled $\gamma_3(K)$ in the photoelectron spectrum of Fig. 1 is presumably due to this gamma-ray. Cork, Shreffler, and Fowler⁷ report that absorption measurements in copper and aluminum give evidence for the existence of an

additional gamma-ray at an energy of about 0.60 Mev. A search was made for a gamma-ray of about this energy, but no evidence was obtained for its existence.

Gamma-ray number 1 has not been reported by other investigators. There is some uncertainty in determining the conversion shell for this gamma-ray, due to the low intensity of the conversion electrons. The assignment given in Table I seems to be consistent with the data from both the internal conversion and photoelectron spectra. An impurity of Zr^{95} was found in the sample by Voigt and Thamer³ of this laboratory, but the intensity of the radiations was too low to account for the observed line. Furthermore, no gamma-ray with an energy of 0.087 Mev has been reported for Zr^{95} or its daughter Cb^{95} .

The peculiar shape of the $\gamma_2(K)$ photoelectron line is due to the photoelectrons [$K_\alpha(L)$] produced by the characteristic x-rays from Ta following the internal conversion of the gamma-rays, and also to the photoelectrons [$\gamma_3(K)$] produced by the gamma-ray of energy 4 kev higher than γ_2 , which has been reported by Chu and Wiedenbeck.⁹ Chu and Wiedenbeck⁹ did not observe the photoelectron lines of γ_2 and cite this as further evidence that γ_2 is highly converted.

B. Intensities of Gamma-Rays

For Fig. 1 the spread in momentum due to the surface density of the lead foil was greater than the momentum spread which the spectrometer could accept.¹⁰ In this case, neglecting scattering, the maximum counting rate for any line is proportional to the number of such quanta emitted from the atom, the photoelectric absorption coefficient, the transmission of the counter window and inversely proportional to the spread in momentum produced by the lead foil. Using the K

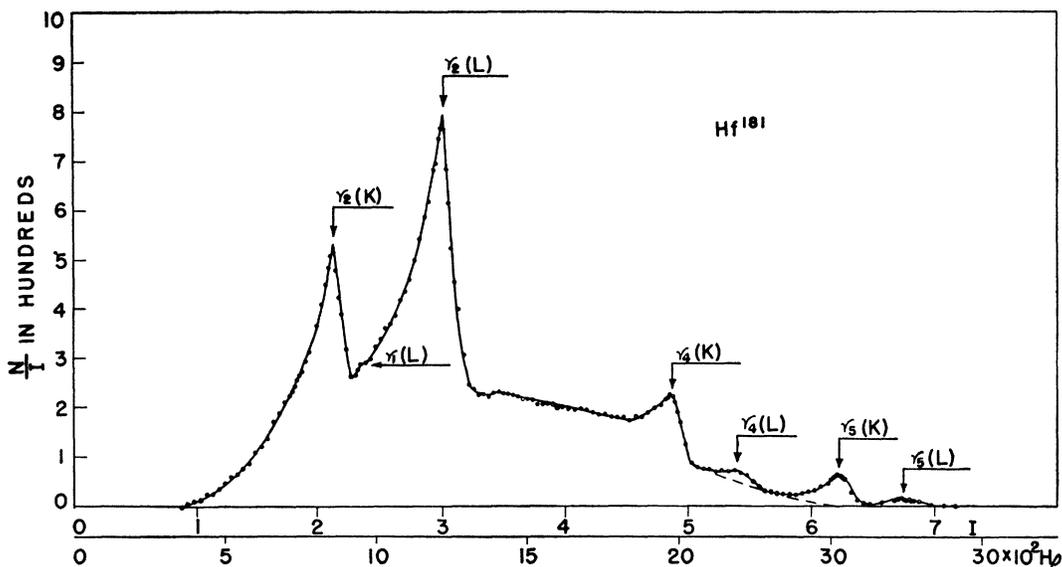


Fig. 2. Composite spectrum of internal conversion electrons and beta-rays from Hf^{181} . N is the counting rate in counts/min. and I is the current in the spectrometer coil.

photo-lines of γ_4 and γ_6 and the L photo-line of γ_2 , after subtracting the Compton background, the gamma-ray intensities were found to be proportional to 11, 29, and 100 for γ_2 , γ_4 , and γ_6 , respectively. In these calculations it was assumed that the photoelectric absorption coefficient for the L shell of γ_2 was 0.23 of that for the K shell as is the case for γ_6 . The photoelectric absorption coefficients were calculated by means of Gray's¹² empirical formula and the spread in momentum produced by the lead foil by means of a formula given by Heitler.¹³

Chu and Wiedenbeck⁹ have proposed a decay scheme, based on coincidence measurements, which is reproduced in Fig. 3. The gamma-ray energies are those determined in this investigation with the exception of γ_3 which is taken as 4 keV greater than γ_2 as determined

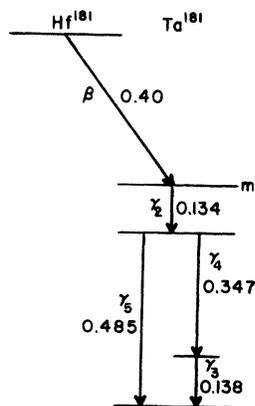


FIG. 3. Proposed decay scheme by Chu and Wiedenbeck (see reference 9). The energies are given in Mev.

by Chu and Wiedenbeck.⁹ In this decay scheme, the number of transitions for γ_3 and γ_4 should be equal. From the spectra determined in this investigation and those of Chu and Wiedenbeck, this does not seem to be the case. It is possible that there are other weak radiations which together with γ_1 are in parallel with γ_3 . For this to be a single gamma-ray, the energy should be 51 keV and the internal conversion line from the L shell should be at 1.7 amp. As Fig. 2 indicates, it was not possible to resolve a line at this current value. If the gamma-ray exists, it undoubtedly has a low intensity.

C. Internal Conversion Coefficient of γ_2

The total internal conversion coefficient is defined as the ratio Ne/Nq , where Ne is the total number of conversion electrons and Nq the number of quanta emitted from the atom for this transition. Assuming the decay scheme shown in Fig. 3, Ne/Nq can be calculated from the ratios of the intensities of the gamma-rays.

Neglecting the small internal conversion coefficients of γ_4 and γ_6 , Ne/Nq for γ_2 is calculated to be 10.8. Taking into account the conversion coefficients determined by Chu and Wiedenbeck⁹ for γ_4 and γ_6 , the total conversion coefficient Ne/Nq is 11.5.

D. Multipole Order of Radiations

DeBenedetti and McGowan¹⁴ report a half-life of 22μ sec. for the short-lived isomer of Ta^{181*} . This has been confirmed by Madansky and Wiedenbeck.² The

¹² L. H. Gray, Proc. Camb. Phil. Soc. 27, 103 (1931).

¹³ W. Heitler, *The Quantum Theory of Radiations* (Oxford University Press, New York 1944), second edition, p. 219.

¹⁴ S. DeBenedetti and F. K. McGowan, Phys. Rev. 70, 569 (1946).

lifetime of a metastable state is given by Bethe¹⁵ as

$$\tau = 5 \times 10^{-21} l!^2 (20/\hbar\omega)^{2l+1} \text{ sec.}, \quad (1)$$

where $\hbar\omega$ is the energy of the emitted radiation in Mev and l is the change in angular momentum of the nucleus. Hebb and Uhlenbeck¹⁶ have suggested that Eq. (1) should be divided by $(1+Ne/Nq)$ in order to allow for the increased decay probability due to internal conversion. Wiedenbeck¹⁷ has shown that this correction improves the agreement between the lifetimes calculated by means of Eq. (1) and those determined experimentally. The corrected half-life, as calculated from Eq. (1), for a metastable state of 0.134 Mev and a spin change of three units is 17μ sec. This is in fairly good agreement with the experimental value of 22μ sec.¹⁴ The corrected half-lives for $l=2$ and 4 are 8.2×10^{-11} sec. and 5.9 sec., respectively.

For the internal conversion spectrum of Fig. 2, the ratio N_K/N_L of the conversion electrons for γ_2 in the K and L shells is 1.19.

Hebb and Uhlenbeck¹⁶ and Dancoff and Morrison¹⁸ have calculated the conversion coefficients for the K shell for electric and magnetic multipole radiation. Hebb and Nelson¹⁹ have made similar calculations for the L shell. From these calculations the ratio N_K/N_L can be determined. These calculations are for elements

with atomic numbers less than about 50. However, it is of interest to apply these formulas to Ta¹⁸¹ for which Z is 73.

The calculated values of N_K/N_L for γ_2 are 0.10 for electric 2^4 radiation and 4.05 for magnetic 2^3 radiation. Since the experimental value of 1.19 is between the two calculated values, the radiation is presumably a mixture of magnetic 2^3 and electric 2^4 radiation. This is consistent with the calculation of the lifetime with a spin change of three units. Hence, the Ta^{181*} nucleus appears to undergo a spin change of three units with the emission of γ_2 (0.134 Mev). Using a formula due to Nelson,²⁰ it is found that the γ_2 -radiation is a little less than half (~ 42 percent) electric and the rest (~ 58 percent) magnetic.

For γ_4 and γ_5 , the experimental values of N_K/N_L are 5.0 and 5.2, respectively, while the calculated values for electric quadrupole radiation ($l=2$) are 6.0 and 6.9, respectively. This suggests that γ_4 and γ_5 are perhaps electric quadrupole radiations.

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¹⁵ H. A. Bethe, Rev. Mod. Phys. **9**, 226 (1937), Eq. (733).

¹⁶ M. H. Hebb and G. E. Uhlenbeck, Physica **5**, 605 (1938).

¹⁷ M. L. Wiedenbeck, Phys. Rev. **69**, 567 (1946).

¹⁸ S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939).

¹⁹ M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940).

²⁰ E. Nelson, cited by A. C. Helmholtz, Phys. Rev. **60**, 415 (1941).