

Disintegration of Ta<sup>180</sup>

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The modes of decay of 8-hour Ta<sup>180</sup> have been studied by means of a magnetic spectrometer and coincidence-absorption techniques. It is found that approximately 21 percent of the decays involve either of two negative beta-particles in transitions to states of W<sup>180</sup>. The remainder of the decays proceed by means of K-capture to at least two different states of Hf<sup>180</sup>. No positrons are observed. Very prominent conversion lines attributed to a 93-keV gamma-transition in Hf<sup>180</sup> are observed, in addition to a weaker line considered to be associated with a 102-keV gamma-ray in W<sup>180</sup>. Auger electrons are also present. The multipolarity of the 93-keV gamma-ray is thought to be electric quadrupole.

## I. INTRODUCTION

EIGHT-HOUR radioactive Ta<sup>180</sup> was investigated by Oldenberg,<sup>1</sup> and more recently by Wilkinson.<sup>2</sup> The first investigator employed absorption techniques upon a sample produced by the reaction Ta<sup>181</sup>(*n,2n*)Ta<sup>180</sup>. K x-radiation was found which was attributed to decay by K-capture. Negatively charged particles of maximum energy 0.48 Mev were also found. The second worker produced the activity by means of (*n,2n*) and (*p,pn*) reactions. Utilizing absorption methods and a magnetic spectrometer, Wilkinson found negative beta-particles of maximum energy 0.7 Mev, L and K x-radiation, and a 1.3-Mev gamma-ray.

Ta<sup>180</sup> has been extensively investigated in this laboratory during the past two years. The work was reported in part at the 1950 meeting of the American Physical Society at Chicago.<sup>3</sup> The activity was obtained in abundance and in pure form by a ( $\gamma,n$ ) reaction on the 100 percent isotope Ta<sup>181</sup>.

## II. APPARATUS

The sources were produced by a probe technique of activation in the 22-Mev betatron at the University of

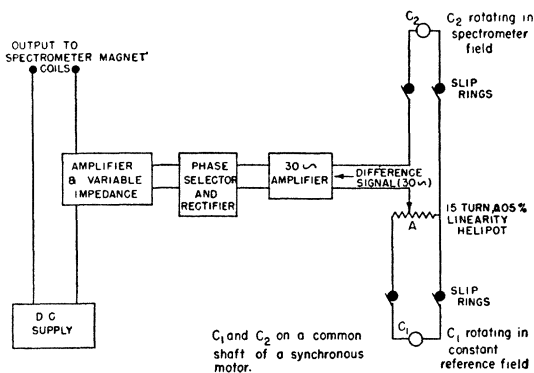


FIG. 1. Block diagram of the field regulator employed with the 180° magnetic spectrometer.

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† Assisted in part by the joint program of the ONR and AEC.

<sup>1</sup> O. Oldenberg, Phys. Rev. **53**, 35 (1938).

<sup>2</sup> G. Wilkinson, Phys. Rev. **80**, 495 (1950).

<sup>3</sup> Bendel, Brown, and Becker, Phys. Rev. **81**, 300 (1951).

Illinois. The details of this method of activation have been reported previously.<sup>4</sup> Metallic tantalum foils of thickness  $\frac{1}{4}$  mil ( $\sim 11$  mg/cm<sup>2</sup>) were largely used in the spectrometric measurements of the particles, while somewhat thicker samples were employed for the gamma-ray measurements. Typical activities for a  $\frac{1}{4}$ -mil foil, 2 mm wide and 24 mm long, were of the order of 20 microcuries of particles.

A 180-degree magnetic spectrometer, a scintillation spectrometer, and absorption and coincidence absorption techniques were employed in the present work. The latter were of a standard variety and need not be discussed here, and the scintillation spectrometer, a multichannel device, will be described elsewhere. The magnetic spectrometer permits the recording of data either manually or automatically. The automatic features provide constant rates of change of the magnetic field, and a recording of counter data at uniform time intervals.

The method through which the changing magnetic field is produced is summarized in the block diagram shown in Fig. 1. The field is monitored by means of two small rotating coils, C<sub>1</sub> and C<sub>2</sub>. C<sub>1</sub> rotates in the field of a reference permanent magnet (magnetron magnet) and C<sub>2</sub> rotates in the gap of the spectrometer. The two coils are on a common shaft of a 30-cycle synchronous motor in order to minimize frequency and phasing difficulties. The output emf of each is taken off by means of slip rings. C<sub>1</sub> is connected to a 15 turn, 0.05 percent linearity Beckman Helipot, A as shown, the output of which is connected in series with the output of C<sub>2</sub>. Any small difference signal is amplified by a 30-cycle amplifier of the Sturtevant<sup>5</sup> type and compared by means of a phase selector with the reference voltage (output of C<sub>1</sub>). Following this the amplified difference signal is rectified. Depending upon the phase of the difference signal relative to that of the emf from C<sub>1</sub>, a positive or negative dc voltage is produced by the rectifier. After a stage of amplification, this control voltage is applied to the grids of 6AS7 triodes to correct

<sup>4</sup> R. A. Becker and F. S. Kirn, Phys. Rev. **76**, 182 (1949); Becker, Kirn, and Buck, Phys. Rev. **76**, 1406 (1949); R. A. Becker, Rev. Sci. Instr., to be published.

<sup>5</sup> J. M. Sturtevant, Rev. Sci. Instr. **18**, 124 (1947).

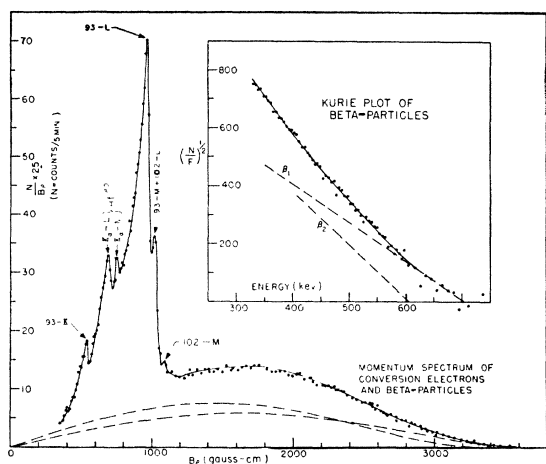


FIG. 2.  $B\rho$  curve showing conversion lines and the continuous beta-spectrum. The Kurie plot of the continuum, depicted in the insert, is consistent with the presence of two components as indicated.

the portion of the magnet current controlled by them. The latter circuit is represented in the diagram as a variable impedance. The result is a field-regulated device. Such a technique has been employed by others.<sup>6</sup>

The dial of the helipot is calibrated directly in terms of  $B\rho$  values by means of the conversion line found in the decay of Cs<sup>137</sup>, and the gamma-rays found in the decay of Co<sup>60</sup>. A linear variation of the spectrometer field with time is realized by driving the helipot shaft with one of an assortment of synchronous timing motors. The counter data are recorded by a Streeter-Amet-type<sup>7</sup> recorder which prints accumulated counts at specified intervals.

### III. DATA

The half-life was measured with five different samples, three of which were followed for at least 10 half-lives. One of the latter was followed for a time greater than 100 half-lives, and showed that the amount of Ta<sup>182</sup> activity present (formed from  $n$  capture by Ta<sup>181</sup>) was of the order of only  $10^{-5}$  of the initial Ta<sup>180</sup> activity. The data were obtained continuously (save for the long run), employing a Streeter-Amet recorder<sup>7</sup> in conjunction with an Amperex type 200C (1.4-mg/cm<sup>2</sup> mica window) halogen-filled counter. The measurements were corrected for dead-time, the presence of the weak Ta<sup>182</sup> activity, and other background. On the basis of six such determinations the half-life was found to be  $8.15 \pm 0.02$  hours.

Figure 2 shows a conventional momentum spectrum of the negative particles present, together with a Kurie plot of the continuous portion of the spectrum. The analysis of the beta-particles is consistent with the existence of two components,  $\beta_1$  and  $\beta_2$ , with end points

<sup>6</sup> E. P. Tomlinson, Phys. Rev. 74, 1231 (1948); E. M. Lyman, private communication.

<sup>7</sup> Streeter-Amet Company, 4101 North Ravenswood Avenue, Chicago, Illinois.

separated by about 100 keV, and with the upper endpoint at  $705 \pm 15$  keV. Presumably the betas involve transitions to states of W<sup>180</sup>. The source thickness was 11 mg/cm<sup>2</sup>, and the counter window thickness was 75  $\mu$ gm/cm<sup>2</sup>. The slight break at approximately 450 keV on the Kurie plot was shown, by employing still thicker sources, to be the result of the unfavorable source thicknesses which were necessarily employed for reasons of intensity. The evidence consisted in the fact that when the foil thickness was changed, the location of the break near 600 keV remained unaltered, while the other shifted in position. The dashed lines on the momentum plot represent the decomposition of the continuum into the two components  $\beta_1$  and  $\beta_2$ . The ratio  $\beta_2/(\beta_1 + \beta_2)$  was found to be  $0.53 \pm 0.09$ . It will be shown below that the disintegration of the parent Ta<sup>180</sup> state proceeds predominantly by  $K$ -capture, there being about four  $K$ -captures to every beta-particle. The conversion lines associated with a 93-keV gamma-ray

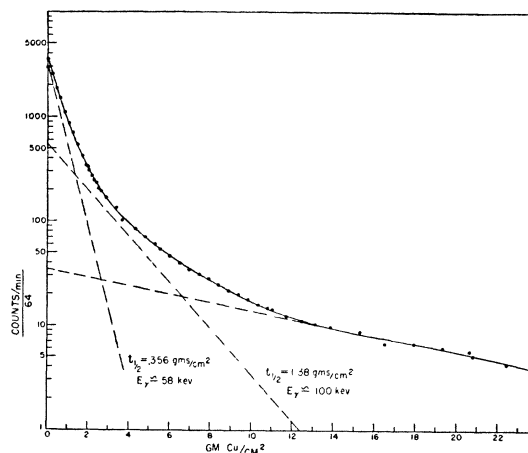


FIG. 3. Absorption, in copper, of x-rays and gamma-rays.

are too intense to be attributed to a transition in W<sup>180</sup>, following the  $\beta_2$ -decay, and are consequently assigned to a transition in Hf<sup>180</sup> following a  $K$ -capture transition. This is consistent with other evidence cited below. Likewise, the two lines between 93-K and 93-L are interpreted as Auger electrons arising from the conversion of the Hf<sup>180</sup> x-rays of energy 54.7 to 55.9 keV.

The weak line near 1100  $B\rho$  was interpreted as an  $M$  conversion of a 102-keV gamma-ray and, for reasons given below, was associated with a transition in W<sup>180</sup>, following  $\beta_2$ . The  $L$  line of the 102-keV gamma-ray is superimposed upon the  $M$  line of the converted 93-keV gamma-ray.

An absorption curve of the electromagnetic radiation, using copper absorbers, is presented in Fig. 3. A scintillation counter detector composed of a 5819 photo-multiplier tube and a 5-mm thick crystal of NaI were employed. The results show that the radiation consists mainly of a portion in the range 55 to 60 keV, interpreted as x-rays, and another at about 100 keV, the

latter being present to the extent of  $0.13 \pm 0.05$  of the lower energy fraction. The symbol  $N_\gamma/N_x$  will be employed below for the ratio. Additional absorption data in the range of higher energies, and using both copper and lead absorbers, indicated the possible presence of one or more weak components in the range 175 to 450 keV. Evidence for this was obtained also with a scintillation spectrometer which confirmed the strong components near 55 keV and 100 keV, and revealed weak lines at approximately 0.2 MeV and 0.4 MeV. The intensity of the weaker radiation was less than 1 percent of the 55-keV component.

The supposition stated above, and by Oldenberg,<sup>1</sup> that the soft component is  $K_\alpha$  x-radiation following a  $K$ -capture transition to  $\text{Hf}^{180}$ , was supported by critical absorption measurements performed with absorbers made from the rare earth oxides  $\text{Nd}_2\text{O}_3$ ,  $\text{Er}_2\text{O}_3$ , and  $\text{Yb}_2\text{O}_3$ . This point of view is supported also by measurements of the relative intensities of the various radiations. These indicated far too many 55-keV quanta (as well as too many conversion electrons) for these to be associated exclusively with the beta-particles going to  $\text{W}^{180}$ .

The relative intensities of the conversion electrons and beta-particles was found to be  $N_e/N_\beta = 2.2 \pm 0.7$ .

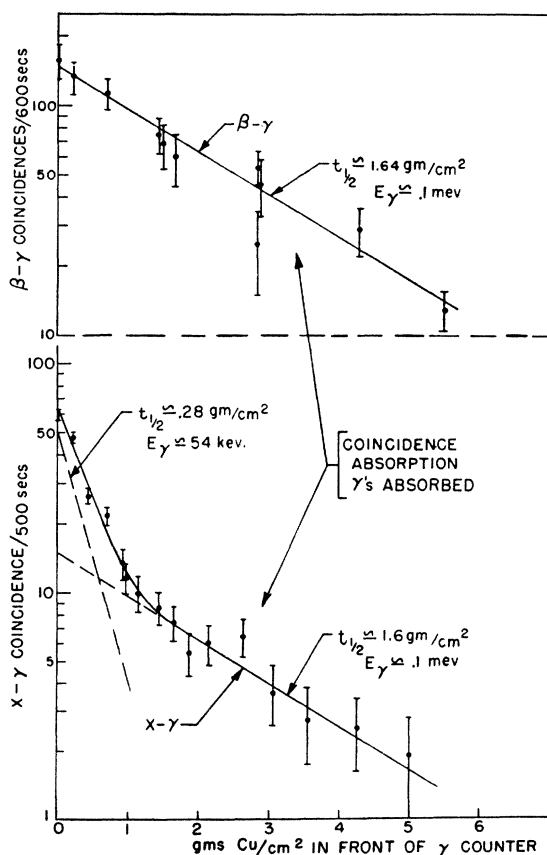


FIG. 4. Beta-gamma and x-gamma coincidences as a function of copper absorber thickness placed in front of the gamma-counter.

The determination was made by absorption of the particles from a somewhat thinner source than used afore-mentioned. Aluminum absorbers and an end-window Geiger counter (1.4-mg/cm<sup>2</sup> window thickness) were employed. The source was prepared chemically from a tantalum foil which had been irradiated with the betatron. The prepared source was estimated to have a thickness of about 1 mg/cm<sup>2</sup> or less.

The possibility was recognized, in these experiments, that the large number of  $L$  x-ray quanta present might affect the numbers  $N_e$  and  $N_\beta$ . Accordingly, absorption curves of the single counting rates from a similar source were taken, in which the radiations were collimated by slits in the field of a permanent magnet. Charged particles of electronic mass were unable, except for scattered radiations, to negotiate the slits in the presence of the field. Data obtained with and without the magnet indicated that the effect of the  $L$  x-rays was not more than 4 percent of that of the charged particles.

Similar measurements were carried out, employing a scintillation counter detector, in order to determine the relative numbers of beta-particles and 55-keV x-rays. The crystal was 5 mm thick NaI covered with 3.4 mg/cm<sup>2</sup> aluminum. The ratio  $N_\beta/N_x$ , after correcting for the thickness of air and aluminum, and assuming 100 percent absorption of the x-rays in the crystal, was found to be  $0.28 \pm 0.08$ .

Figure 4 shows the results of beta-gamma and x-gamma coincidence measurements employing a Woods type end-window counter for the beta-particles, and scintillation counters as detectors for the x-rays and gamma-rays. The data are plotted in terms of numbers of coincidences as a function of copper absorber thickness placed in front of the gamma-ray detector. Particles were prevented from reaching the phosphors by means of aluminum blocks of thickness 1 g/cm<sup>2</sup>. The results indicate that gamma-radiation of approximately 100 keV is concerned in both the beta-gamma coincidences and x-gamma coincidences. The low energy tail of the lower curve owes itself to additional coincidences arising from x-rays being detected in the gamma-counter at small absorber thicknesses. This portion of the absorption curve presents a half-thickness attributable to radiation of about 55-keV energy. These results are consistent with the assignments of the conversion lines previously given in Fig. 2.

A search was made for possible positrons by studying the gamma-gamma coincidences. Scintillation counters were employed at the two angular separations of 90° and 180°. The results indicated that there are not more than 5 positrons in 10<sup>5</sup> disintegrations.

#### IV. DISCUSSION

An estimate of the total conversion coefficient of the 93- and 102-keV gamma-radiation, although subject to large uncertainties, was obtained from  $N_e/N_\gamma$ , the ratio of the total number of electrons to the total gamma-radiation of energy near 100 keV. This was

found to be in the range 2 to 9, with a probable value of 4.6. The determination was made by utilizing the three ratios  $N_e/N_\beta$ ,  $N_\beta/N_x$ ,  $N_x/N_\gamma$  which were mentioned in Sec. III. The quantity  $N_x$  was corrected to include the Auger electrons arising from the conversion of the x-rays in the *L* and *M* shells of Hf<sup>180</sup> (and to some extent in W<sup>180</sup>). The number of these was estimated by comparison with the data of Steffen, Huber, and Humbel<sup>8</sup> for Au<sup>194</sup> and Au<sup>196</sup>. It was thought that a reasonable value was 0.077 for the ratio of Auger electrons to x-rays.

The *K/L* ratio of conversion electrons associated with the 93-keV gamma-ray was estimated from Fig. 2, aided also by an analysis of the type of Richardson's<sup>9</sup> for thick sources. The value found was  $0.15^{+0.15}_{-0.05}$ . Employing Axel's<sup>10</sup> curves, and an empirical curve kindly supplied in advance of publication by Goldhaber,<sup>11</sup> considerations of the total conversion coefficient and of the *K/L* ratio indicated the 93-keV gamma-ray probably to be electric quadrupole radiation.

The above results are summed up in Fig. 5, which is a partial level scheme thought to be involved in the decay of Ta<sup>180</sup> to Hf<sup>180</sup> and W<sup>180</sup>. The total percentage of *K*-capture transitions was determined to be as shown in the figure from the ratio  $N_\beta/N_x$  by correcting for the presence of Auger electrons, and for the presence of x-rays following *K*-conversion of the two gamma-rays principally involved in various decay processes. The division into the two branches *K*<sub>1</sub> and *K*<sub>2</sub> was necessary in order to account for the excess of x-rays over radiations involved in the 93-keV transition. *K*<sub>1</sub> is assumed to go to the ground state of Hf<sup>180</sup> because of the absence of other radiation of intensity comparable to that of the 93-keV radiations. Although not sufficiently certain to be included in the figure, the ratio of *K*<sub>1</sub>/*K*<sub>2</sub> was estimated to be about 0.7. The branch *K*<sub>3</sub> was included hypothetically in order to explain the apparent presence of weak gamma-ray components in the range 175 to 450 keV. Gamma-rays in this range have been observed in Hf<sup>180</sup> by Burson.<sup>12</sup> The assumption is also made that  $\beta_1$  involves the ground state of W<sup>180</sup>.

<sup>8</sup> Steffen, Huber, and Humbel, *Helv. Phys. Acta* **22**, 167 (1949).

<sup>9</sup> H. O. W. Richardson, *Proc. Phys. Soc. (London)* **A63**, 234 (1950).

<sup>10</sup> P. Axel and R. F. Goodrich, ONR report, unpublished.

<sup>11</sup> M. Goldhaber, private communication.

<sup>12</sup> Burson, Blair, Keller, and Wexler, *Phys. Rev.* **83**, 222 (1951).

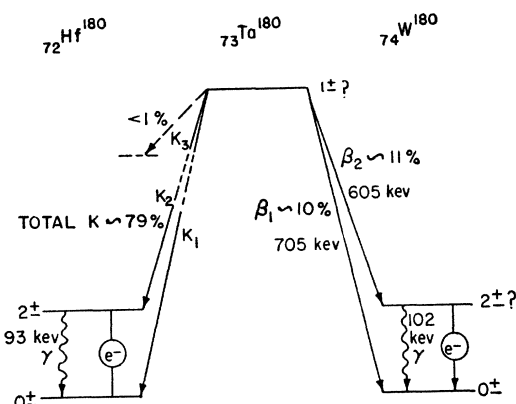


FIG. 5. Tentative partial level scheme involved in the decay of Ta<sup>180</sup> to Hf<sup>180</sup> and W<sup>180</sup>.

Since both Hf<sup>180</sup> and W<sup>180</sup> are even-even nuclei, it is assumed that they both have zero spin in the ground state. Accordingly, since the 93-keV gamma-ray was judged to be electric quadrupole, the spin of the excited state of Hf<sup>180</sup> must be 2 and must have the same parity as that of the ground state. Estimates of the *ft* values of *K*<sub>1</sub> and *K*<sub>2</sub> were made, assuming that at least 400 keV was involved in the *K*<sub>1</sub> transition, and in view of the absence of positrons, that not more than 1400 keV was involved. The values of  $\log ft$  for both *K*<sub>1</sub> and *K*<sub>2</sub> were estimated to be in the range 4.5 to 6.6. The spin of the Ta<sup>180</sup> parent state, consistent with both these *ft* values and with the fact that *K*<sub>1</sub> and *K*<sub>2</sub> are of approximately equal intensity, is chosen to be 1. The magnitudes of  $\log ft$  are consistent with allowed, and possibly with first-forbidden transitions. Thus the parity of Ta<sup>180</sup>, relative to that of the two states of Hf<sup>180</sup>, is subject to some doubt. The parity of Ta<sup>180</sup>, as shown in the figure, is that which is the case if *K*<sub>1</sub> and *K*<sub>2</sub> are both allowed.

The values of  $\log ft$  for  $\beta_1$  and  $\beta_2$  were found to be 6.8 and 6.5, respectively. The spin assignment to the excited state of W<sup>180</sup> is consistent with both these *ft* values and with the fact that  $\beta_1$  and  $\beta_2$  are of approximately equal intensity. It is quite likely that the same type of transition is involved for the major components going to W<sup>180</sup>, as for the major components going to Hf<sup>180</sup>. For the spin and parity assignments shown in W<sup>180</sup> the 102-keV gamma-ray would be electric quadrupole.