# Short-Lived Isomers\* of Ta<sup>181</sup>

W. C. BARBER Stanford University, Stanford, California (Received July 3, 1950)

Excited states of Ta<sup>181</sup> which result from the 46-day  $\beta$ -decay of Hf<sup>181</sup> were investigated by the method of delayed coincidences. Using trans-stilbene scintillation counters and a coincidence system of about  $1.3 \times 10^{-8}$ sec. resolving time, a state with a half-life of  $(1.08 \pm 0.05) \times 10^{-8}$ -sec. was observed. Delayed coincidence absorption experiments indicated that the radiation following the delayed state arises chiefly from a transition of about 0.5-Mev energy.

The radiations preceding and following the  $2.2 \times 10^{-5}$ -sec. state were also investigated and, in agreement with other recent results, no  $\gamma$ -rays were observed in the preceding radiation.

Possible assignments of spin and parity to the excited states of the Ta<sup>181</sup> nucleus are discussed.

The scintillation counter system was also used to search for delayed coincidences in the radiations following the decay of other isotopes. Sources of 5.3-yr. Co<sup>60</sup>, 225-day Ag<sup>110</sup>, 2.7-day Au<sup>198</sup>, 2.6-hr. Mn<sup>56</sup>, and 34-hr. Br<sup>82</sup> gave negative results. The 8×10<sup>-8</sup>-sec. state of Cd<sup>111</sup> was observed in the decay of 2.8-day In<sup>111</sup>.

### I. INTRODUCTION

HE measurement of short-time intervals by the delayed coincidence method provides, in certain cases, a direct method for determining the lifetimes of excited states of nuclei.<sup>1</sup> In case a state of measurable lifetime is observed, the delayed coincidence system is a valuable tool to be used in connection with absorption or spectrometer studies to give information on the position in the decay scheme of the nuclear energy levels involved. Knowledge of the lifetime and positions of the energy levels is of importance in obtaining further valuable information about the nuclear states (e.g., spins and parities).

This paper is chiefly a report of a delayed coincidence investigation of the radiations emitted in the decay of Hf<sup>181</sup>. Evidence is presented for the existence of a new short-lived isomer of Ta<sup>181</sup>, and the sequence of the transitions of this latter nucleus is discussed in the light of delayed coincidence absorption experiments.



FIG. 1. Level diagram showing the major radiations produced by Hf<sup>181</sup>. Transition energies are given in Mev. The  $\gamma$ -rays are labeled in order of increasing energy.

\* Assisted by the joint program of the ONR and AEC. <sup>1</sup>S. DeBenedetti and F. K. McGowan, Phys. Rev. 74, 728

The radiations of Hf<sup>181</sup> have been studied by a number of observers, and there is agreement among recent investigators as to the general features of the decay scheme.<sup>2-5</sup> However, there are several details of the level scheme which are still in doubt, and a recent investigation<sup>5</sup> indicates that the radiation spectrum may be more complicated than was previously supposed.\* The facts generally agreed upon can be summed up in the level scheme shown in Fig. 1. In this figure the  $\gamma$ -ray energies have been taken from reference 5. The branching ratio for parallel transitions has been calculated from data given in reference 2. The assignment of the  $1.08 \times 10^{-8}$ -sec. level is made on the basis of investigation herein reported.

#### **II. APPARATUS**

Figure 2 shows a block diagram of the delayed coincidence apparatus as used in studying the very shortlived states.

The source was placed in position "S" and various absorption foils were inserted between the source and either counter.

The particle detectors were scintillation counters employing anthracene or trans-stilbene crystals mounted on 931-A photo-multiplier tubes.<sup>6</sup> The multiplier tubes were usually operated at room temperature, but in a few experiments, where multiplier noise counts were troublesome, the tubes were cooled with dry ice.

Cathode follower tubes sent the output pulses from

<sup>5</sup> Cork, Stoddard, Rutledge, Branyan, and Le Blanc, Phys. Rev.

78, 299 (1950).

\*Note added in proof: It has recently been shown [M. Deutsch and A. Hedgran, Phys. Rev. 79, 400 (1950)] that most of the radiation which had previously been attributed to  $\gamma_3$  is due to a longer-lived activity than that of Hf181. However, the investigation of Cork et al. indicates the presence of two different  $\gamma$ -rays of for the task in indicates the presence of two uniterim priors of about 0.345-Mev energy and coincidence spectrometer studies [C. M. Fowler and R. G. Shreffler, Rev. Sci. Inst. 21, 740 (1950)] indicate that  $\gamma_3$  has a place in the Hf<sup>181</sup> decay scheme. • The stilbene crystals used in the investigation were kindly when the proceeding the proceeding the process of the stille science of the stille science of the science of

supplied by Professor Robert Hofstadter of Princeton University.

<sup>(1948).</sup> 

<sup>&</sup>lt;sup>2</sup> K. Y. Chu and M. L. Wiedenbeck, Phys. Rev. 75, 226 (1949).
<sup>8</sup> E. N. Jensen, Phys. Rev. 76, 958 (1949).
<sup>4</sup> Arne Lundby, Phys. Rev. 76, 1809 (1949).



FIG. 2. Block diagram of the delayed coincidence apparatus. Scintillation counters are used. The amplifiers (amp.) are of the distributed type (reference 7).

the multiplier tubes into the coaxial cables which served as delay lines. The cables were Type RG-7/U which have a characteristic impedance of 97.5 ohms and a propagation velocity of 0.84 c. The amplifiers were of the distributed type,<sup>7</sup> having a voltage gain of about 25 and a band width (to the half-power point) of about 60 Mc/sec.

The coincidence system was a Rossi-type circuit which operated reliably for input pulses greater than  $2 \text{ volts.}^8$ 

The  $2.2 \times 10^{-5}$ -sec. state of Ta<sup>181</sup> was investigated using vacuum-tube univibrator circuits to shape the pulses and produce the delays. Pulse widths and delay intervals were measured on a synchroscope. Pulses from the counters were amplified by conventional high gain amplifiers.<sup>9</sup> Except for the fact that both scintillation counters and Geiger counters were used as particle detectors, the arrangement of the apparatus into a delayed coincidence system was the same as that described by DeBenedetti and McGowan.<sup>1</sup>

### III. SOURCES

The sources of Br<sup>82</sup>, Au<sup>198</sup>, and Mn<sup>56</sup> were prepared by irradiation of the stable isotopes of these elements with neutrons produced by the Stanford cyclotron. The Co<sup>60</sup>, Ag<sup>110</sup>, and Hf<sup>181</sup> sources were produced at Oak Ridge by pile irradiation. The In<sup>111</sup> source was produced at the University of California at Berkeley and kindly supplied to the author by Professor A. C. Helmholz.

The first investigation of the Hf radioactivity was

made with a source which was more than one year old. This source showed the presence of an electron component of energy slightly greater than that of any of the electrons reported<sup>2</sup> from the 46-day Hf<sup>181</sup>. These energetic electrons were observed to be in immediate coincidence with  $\gamma$ -rays capable of penetrating several g/cm<sup>2</sup> of lead,<sup>10</sup> a situation which is at variance with the decay scheme shown in Fig. 1. A second source of Hf<sup>181</sup>, which had been produced shortly prior to the time measurements were made, showed no evidence for an appreciable number of energetic electrons in coincidence with hard  $\gamma$ -rays, so the early results must be due either to a longer-lived<sup>5</sup> Hf or an impurity.

The radiation spectrum of the second Hf source was kindly observed in a  $\beta$ -ray spectrograph by Mr. R. W. Hayward of the University of California. This investigation showed the spectrum to be essentially the same as that reported for Hf<sup>181</sup> by other experimenters.<sup>2,3</sup> The continuous  $\beta$ -spectrum and the conversion lines of  $\gamma_1$ ,  $\gamma_3$ , and  $\gamma_4$  were observed in one experiment. In a second experiment Compton electrons from an aluminum radiator were measured. Those produced by  $\gamma_4$  were



FIG. 3. Curves of coincidence rate as a function of delay, taken with the apparatus of Fig. 2. The points (+++) were taken with "simultaneous" events in each counter. The points (0 o 0) represent electron- $\gamma$ -coincidences produced by Hf<sup>181</sup>, where the electrons are admitted into counter 2 only. The two curves have been normalized to include the same area on a linear plot. The vertical lines indicate statistical standard errors.

<sup>10</sup> W. C. Barber, Phys. Rev. 78, 82 (1950).

<sup>&</sup>lt;sup>7</sup> Ginzton, Hewlett, Jasberg, and Noe, Proc. I.R.E. 36, 956 (1948).

<sup>&</sup>lt;sup>8</sup> Interested persons may secure a diagram of the coincidence circuit by writing to the author.

<sup>&</sup>lt;sup>9</sup> Some of the equipment used in this study was kindly lent to the Stanford Physics Department by the Department of Physics of the University of Illinois.



FIG. 4. Absorption in lead of the  $\gamma$ -radiation which is in delayed coincidence with electrons. In taking this curve the pulses from the electron counter were electrically delayed by  $4 \times 10^{-8}$  sec. more than the pulses from the  $\gamma$ -counter. The hard  $\gamma$ -component has an energy of about 0.5 Mev.

easily detected, but no electrons corresponding to  $\gamma$ -rays of energy greater than  $\gamma_4$  were observed.

The experiments on the radioactivity of Hf<sup>181</sup> which are described in the remainder of this paper were all made with the second source of Hf<sup>181</sup>. This source may also contain the longer-lived activity, but the spectrometer investigation shows that the amount of such activity must be less than one or two percent of the activity due to Hf181.

### IV. THE $1.08 \times 10^{-8}$ -SEC. STATE OF Ta<sup>181</sup>

Figure 3 shows data taken with the apparatus of Fig. 2 using stilbene scintillation counters. The normalized coincidence rate (logarithmic scale) is plotted as a function of the relative delay in channel 2. All data have been corrected for accidental coincidences. The points (+++) in Fig. 3 were taken with the counters in their usual geometry, but with a strong  $\gamma$ -ray source located far away from both counters. The distance from the source to either counter was at least ten times the distance between counters, so that double Compton scattering was the only process which contributed appreciably to the coincidence rate. This process is not so improbable as the word "double" suggests, because almost all of the counts produced by  $\gamma$ -rays in a stilbene crystal are due to Compton scattering, and since the two counters are close together, there is a good chance of detecting the scattered photon in the second counter. Since it is likely that any delays in the Compton process are small compared with the resolving time of this apparatus,<sup>11</sup> the curve (+++) in Fig. 3 is taken as a

measure of the resolving time of the system. This interpretation is supported by the observation that  $\beta - \gamma$ and  $\gamma - \gamma$ -coincidences produced by Co<sup>60</sup> sources yield curves similar to that of the double Compton process. The results of Bell and Petch<sup>12</sup> show that the transitions in Ni<sup>60</sup> proceed with half-lives less than  $3 \times 10^{-9}$  sec.

The data plotted as circles in Fig. 3 were taken using sources of Hf<sup>181</sup>. The source was shielded from the crystal of counter 2 by less than 0.4 mg/cm<sup>2</sup> of Nylon and from counter 1 by 3.2 g/cm<sup>2</sup> of lead plus 350 mg/cm<sup>2</sup> of aluminum. Thus only hard  $\gamma$ -rays are detected in counter 1, whereas very soft  $\beta$ -rays are detected in counter 2. The presence of the metastable state is indicated by the exponential decrease in the coincidence rate as the pulses from counter 2 are delayed. The asymmetry of the curve shows that the particles detected in counter 1 have suffered a nuclear delay which compensates for the electrical delay in channel 2.

The solid line in Fig. 3 represents the curve to be expected if all of the coincidences arise from events where the particles detected in counters 2 and 1, respectively, precede and follow a nuclear state of halflife  $1.08 \times 10^{-8}$  sec. Computation of this curve is as follows: Let  $f(\delta)$  represent the curve (+++) in Fig. 3 (i.e.,  $f(\delta)$  is the experimental delay curve when the two particles are simultaneous). Here  $\delta$  stands for the electrical delay introduced in channel 2. If the particles are separated by a nuclear state of decay constant  $\lambda$ the frequency distribution of pairs of particles separated by a time between t and t+dt is given by  $\lambda e^{-\lambda t}$ . The probability of detecting this pair of particles in the coincidence circuit with delay setting  $\delta$  is proportional to  $f(\delta - t)$  and hence the over-all delay curve  $F(\delta, \lambda)$  is given by

$$F(\delta, \lambda) = \int_0^\infty f(\delta - t) \lambda e^{-\lambda t} dt.$$
 (1)

Differentiation of Eq. (1) with respect to  $\delta$  and then integration of the resulting equation by parts gives the result

$$\partial F(\delta, \lambda) / \partial \delta = \lambda [f(\delta) - F(\delta, \lambda)],$$
 (2)

which means that  $F(\delta, \lambda)$  has its maximum where it intersects  $f(\delta)$ .<sup>13</sup>

The solid curve of Fig. 3 was computed by numerical integration of Eq. (1), using a value of  $\lambda$  determined by the slope of the experimental delay curve at large values of  $\delta$ .

The good agreement between the computed and the experimental delay curves permits the following conclusions:

(1) A metastable state is present and its half-life is about  $1.08 \times 10^{-8}$  sec. A probable error of  $0.05 \times 10^{-8}$  sec. is assigned to

<sup>&</sup>lt;sup>11</sup> R. Hofstadter and J. A. McIntyre, Phys. Rev. 78, 24 (1950).

<sup>&</sup>lt;sup>12</sup> R. E. Bell and H. E. Petch, Phys. Rev. **76**, 1409 (1949). <sup>13</sup> This result has been derived independently by T. D. Newton who has given a general discussion of the evaluation of delayed coincidence experiments, Phys. Rev. 78, 490 (1950).

the result from consideration of the statistical errors in the determination of the delay curves as well as the uncertainty in the propagation velocity of the cable (two or three percent).

(2) The decay of the Hf<sup>181</sup> source produces few electrons in immediate coincidence with hard  $\gamma$ -rays. If such coincidences were present they would produce a delay curve like  $f(\delta)$  and the experimental delay curve would not agree with  $F(\delta, \lambda)$ .

In order to investigate the position of the metastable state in the level scheme of Ta<sup>181</sup>, absorption studies were made of the delayed and preceding radiation.

The presence of  $\gamma$ -radiation preceding the metastable state was demonstrated by detecting  $\gamma - \gamma$ -delayed coincidences. With counter 1 shielded by 3.2 g/cm<sup>2</sup> of lead and counter 2 shielded by 450 mg/cm<sup>2</sup> of aluminum a delay curve similar to that of the circles in Fig. 3 was obtained. An absorption study of the preceding  $\gamma$ -rays is difficult because of the high ratio of accidental to true delayed coincidences. However, data were taken which indicated that the preceding  $\gamma$ -rays are strongly absorbed by lead (half-value thickness less than 0.5 g/cm<sup>2</sup>) and hence consist mostly of photons of energy less than 200 kev.

Figure 4 represents the absorption in lead of the electromagnetic radiation which follows the metastable state. Statistical errors are less than or equal to the size of the plotted circles. In taking this curve the delay in channel 2 was fixed at  $4 \times 10^{-8}$  sec. and the lead absorbers were placed between the source and counter 1. An absorber of 180 mg/cm<sup>2</sup> of Celluloid was kept directly in front of counter 1 to prevent  $\beta$ -rays or secondary electrons from entering this counter. The half-value thickness of the delayed hard component is 5.3 g/cm<sup>2</sup>, which indicates a quantum energy of about 0.5 Mev. The absorption of the delayed soft component is consistent with the interpretation that it consists of  $\gamma$ -rays of about 140 kev with perhaps some K x-rays of Ta as well. The absorption curve shows the amount of the soft component to be between 10 and 15 percent of the total, but since the efficiency of the counters for soft radiation is much less than for hard, the actual amount of soft component is more than this.

Figure 5 shows the absorption in Celluloid of the electrons which precede and follow the metastable state. Counts caused by  $\gamma$ -rays have not been subtracted, but the data were taken using thin stilbene crystals which had low efficiency for detecting  $\gamma$ -rays. Both of these curves were taken with an electrical delay of  $4 \times 10^{-8}$  sec. Statistical standard deviations are indicated by the vertical lines.

Absorption of the particles following the metastable state (shown by the circles in Fig. 5) indicates the presence of energetic electrons not completely absorbed by 85 mg/cm<sup>2</sup> of Celluloid as well as a softer electronic component. These two electron components are presumably due to the conversion of the transitions which produce the two  $\gamma$ -components indicated by Fig. 4. The data are consistent with the level scheme in Fig. 1 where  $\gamma_2$  is responsible for the delayed soft component and  $\gamma_4$  (and perhaps  $\gamma_3$  as well) for the delayed hard component.

The crosses in Fig. 5 show the absorption of the particles preceding the metastable state. Ninety percent of these particles are absorbed by 13 mg/cm<sup>2</sup> of Celluloid, indicating that the particles are electrons of about 100-kev energy. This result is also consistent with the level scheme of Fig. 1, where the preceding electrons are the result of the K and L conversion of the  $\gamma_1$ -transition.

## V. THE 2.2 $\times$ 10<sup>-5</sup>-SEC. STATE OF Ta<sup>181</sup>

The radiations preceding and following the  $2.2 \times 10^{-5}$ sec. state were also investigated by absorption measurements. The delayed coincidences were produced by the system of univibrators mentioned earlier.

The delayed radiation showed the presence of  $\gamma$ -rays as well as electrons. The  $\gamma$ -radiation contained a component with a half-value thickness greater than 3 g/cm<sup>2</sup> of lead and is thus probably due to  $\gamma_4$ ,  $\gamma_3$ , or both. The half-life of the state was observed to be about 20  $\mu$ sec., but an accurate determination was not attempted.

All attempts to detect  $\gamma$ -rays or x-rays in the radiation preceding the  $2.2 \times 10^{-5}$ -sec. state yielded negative results. Experiments to detect  $\gamma - \gamma$ -delayed coincidences were made at first, using anthracene scintillation counters and later using Geiger counters with plated silver cathodes. The counters were shielded from  $\beta$ -rays by 200 mg/cm<sup>2</sup> of Celluloid and the immediate as well as the delayed coincidences were recorded. The sta-



FIG. 5. Absorption in Celluloid of electron-electron delayed coincidences. The points (x-x-x) were obtained when the absorber was placed in front of the counter whose pulses were electrically delayed. In taking each curve the electrical delay was fixed at  $4 \times 10^{-8}$  sec.

TABLE I. Calculated  $\gamma$ -radiation decay constants for the transitions in Ta<sup>181</sup>.

<i>l</i> = 2	l=3	1-4	1=5
1.1×10 <sup>8</sup>	4×10 <sup>2</sup>	8×10-4	
$1.3 \times 10^{8}$	$5 \times 10^2$	1×10-3	•••
$1.6 \times 10^{10}$	3×10 <sup>5</sup>	•••	
8×1010	$3 \times 10^{6}$	$8 \times 10$	
	1×107	$7 \times 10^2$	2.5×10 <sup>-2</sup>
	$\begin{array}{c} l=2\\ \hline 1.1 \times 10^8\\ 1.3 \times 10^8\\ 1.6 \times 10^{10}\\ 8 \times 10^{10}\\ & \ddots \end{array}$	$\begin{array}{c c} & & & & \lambda_{\gamma}(g_{1}) \\ \hline l=2 & l=3 \\ \hline 1.1 \times 10^{8} & 4 \times 10^{2} \\ 1.3 \times 10^{8} & 5 \times 10^{2} \\ 1.6 \times 10^{10} & 3 \times 10^{5} \\ 8 \times 10^{10} & 3 \times 10^{6} \\ \cdots & 1 \times 10^{7} \end{array}$	$\begin{array}{c ccccc} & & & & & & & & \\ \hline l=2 & l=3 & l=4 \\ \hline 1.1 \times 10^8 & 4 \times 10^2 & 8 \times 10^{-4} \\ 1.3 \times 10^8 & 5 \times 10^2 & 1 \times 10^{-3} \\ 1.6 \times 10^{10} & 3 \times 10^5 & \cdots \\ 8 \times 10^{10} & 3 \times 10^6 & 8 \times 10 \\ \cdots & 1 \times 10^7 & 7 \times 10^2 \end{array}$

TABLE II. Half-lives of the states of Ta<sup>181</sup>, calculated from Table I but taking into account experimental values of internal conversion and  $\gamma$ -ray branching. Experimental half-lives are given for comparison.

1-2	7 _ 5	Experi- mental		
1-2	1-3	1-4	1-3	1 g (sec.)
4×10 <sup>-10</sup>	1 ×10 <sup>-4</sup>	6×10	•••	$2.2 \times 10^{-5}$
8×10 <sup>-10</sup>	$2 \times 10^{-4}$			
$1 \times 10^{-11}$	$7 \times 10^{-7}$			
$6 \times 10^{-12}$	$1.5 \times 10^{-7}$	$6 \times 10^{-3}$		$1.08 \times 10^{-8}$
	$7 \times 10^{-10}$	$1 \times 10^{-5}$	2.7×10 <sup>-1</sup>	2.2 ×10 <sup>-5</sup>
	$l=2$ $4 \times 10^{-10}$ $8 \times 10^{-10}$ $1 \times 10^{-11}$ $6 \times 10^{-12}$	$\begin{array}{c} \mbox{Calculated}\\ l=2 & l=3 \\ 4\times10^{-10} & 1 & \times10^{-4} \\ 8\times10^{-10} & 2 & \times10^{-4} \\ 1\times10^{-11} & 7 & \times10^{-7} \\ 6\times10^{-12} & 1.5\times10^{-7} \\ \cdots & 7 & \times10^{-10} \end{array}$	$\begin{array}{c} \begin{array}{c} \mbox{Calculated } T_{\frac{1}{2}} \ (\text{sec.}) \\ l=2 & l=3 & l=4 \end{array} \\ \hline 4\times10^{-10} & 1 \ \times10^{-4} & 6\times10 \\ 8\times10^{-10} & 2 \ \times10^{-4} & \cdots \\ 1\times10^{-11} & 7 \ \times10^{-7} & \cdots \\ 6\times10^{-12} & 1.5\times10^{-7} & 6\times10^{-3} \\ \cdots & 7 \ \times10^{-10} & 1\times10^{-5} \end{array}$	$\begin{array}{c c} & \begin{array}{c} \text{Calculated } T_{i} \ (\text{sec.}) \\ l=2 & l=3 & l=4 & l=5 \end{array} \\ \hline 4 \times 10^{-10} & 1 \ \times 10^{-4} & 6 \times 10 & \cdots \\ 8 \times 10^{-10} & 2 \ \times 10^{-4} & \cdots & \cdots \\ 1 \times 10^{-11} & 7 \ \times 10^{-7} & \cdots & \cdots \\ 6 \times 10^{-12} & 1.5 \times 10^{-7} & 6 \times 10^{-3} & \cdots \\ \cdots & 7 \ \times 10^{-10} & 1 \times 10^{-5} & 2.7 \times 10^{-1} \end{array}$

tistical significance of the negative result is indicated by the ratio of total delayed to total immediate coincidences which (after correction for accidental coincidences and for the finite gate widths of the delayed coincidence system) was  $0.013\pm0.020$ . The  $\pm$  figure gives the statistical standard deviation.

The negative result of the attempts to detect  $\gamma - \gamma$ delayed coincidences is in agreement with the results of other recent investigations4, 14, 15 and indicates that the  $\beta$ -decay of Hf<sup>181</sup> must, in almost all cases, lead directly to the  $2.2 \times 10^{-5}$ -sec. state of Ta<sup>181</sup>. Additional support for this conclusion is provided by the absence of immediate coincidences between energetic electrons and hard  $\gamma$ -rays. Coincidences of this sort were sought, using the scintillation counter system with  $1.3 \times 10^{-8}$ sec. resolving time. The  $\gamma$ -counter was shielded by 2.3  $g/cm^2$  of lead. When the  $\beta$ -counter was shielded by 7 mg/cm<sup>2</sup> of aluminum the net  $\beta - \gamma$ -coincidence rate was 28 counts/min. Increasing the aluminum absorber to 33 mg/cm<sup>2</sup> reduced the net  $\beta - \gamma$ -coincidence rate to  $(0.8\pm0.3)$  counts/min. The coincidences with only 7 mg/cm<sup>2</sup> of absorber are chiefly due to the conversion electrons of  $\gamma_1$  in coincidence with  $\gamma_4$ . An absorber of  $33 \text{ mg/cm}^2$  of aluminum is enough to absorb all of the conversion electrons of  $\gamma_1$  and  $\gamma_2$ , and therefore according to Fig. 1 no coincidences between hard  $\gamma$ -rays and electrons should be observed. The experiment indicates a small positive effect which may be due to the presence of a longer-lived activity in the source. (Refer to discussion under Section III.) Comparison of this small effect with the coincidence rate when only 7 mg/cm<sup>2</sup> of aluminum shields the  $\beta$ -counter shows that the amount of such activity must be small. Because of the presence of the  $1.08 \times 10^{-8}$ -sec. state, only about

one-half of the soft electron-hard  $\gamma$ -events are close enough in time to be detected by the coincidence system. Furthermore, the efficiency of the scintillation counters is much less for the soft electrons than for the hard ones. Taking these corrections into account, it can be concluded that the number of coincidences between hard  $\gamma$ -rays and penetrating electrons is fewer than one or two percent of the number of Ta<sup>181</sup> disintegrations.

#### VI. DISCUSSION OF THE Ta<sup>181</sup> ENERGY LEVELS

The delayed coincidence absorption experiments are in good agreement with the level scheme of Fig. 1, but attempts to make complete spin and parity assignments to the levels of  $Ta^{181}$  meet with difficulty.

The assignment of the multipole character of  $\gamma$ -ray transitions on the basis of lifetime measurements is somewhat uncertain, because the calculation of the radiative transition probability requires a detailed knowledge of the nucleus. However, various nuclear models have been used successfully to estimate transition probabilities. A recent survey of nuclear isomers<sup>16</sup> indicates that the formula

$$\lambda_{\gamma} = \frac{1}{3} \left( \rho^{2l} / (l!)^2 \right) (W/137)^{2l+1} (mc^2/\hbar)$$

is a good approximation for the average decay constant of  $\gamma$ -ray transitions, but that individual cases may be expected to vary by factors of 10 to 100 from this value. In this formula l is the multipole order for electric radiation, W is the transition energy in units of  $mc^2$ , and  $\rho$  is the nuclear radius in units of  $e^2/mc^2$ . Application of the formula to the  $\gamma$ -rays of Ta<sup>181</sup> yields the values which are given in Table I.

The entries in Table I are the decay constants for  $\gamma$ -emission only. Any competing processes, such as internal conversion or  $\gamma$ -ray branching, make the total decay constant of a state larger. Table II gives the expected half-lives of the states of Ta<sup>181</sup> as computed from the  $\gamma$ -decay constants of Table I, taking into account the experimentally determined correction for internal conversion and branching. The half-life is calculated from the relation

$$T_{\frac{1}{2}}=0.693f/\lambda_{\gamma}(1+\alpha),$$

where  $\alpha = \text{conversion}$  coefficient and f = fractionalbranching. Except for  $\gamma_5$ , the data necessary to calculate the conversion coefficients and branching ratios have been taken from the paper of Chu and Wiedenbeck.<sup>2</sup> The relative intensity of  $\gamma_5$  has not been measured, but it is certainly small. In order to give a halflife in Table II, a value of one percent has been assumed for the branching of  $\gamma_5$ .

As has been pointed out by Lundby,<sup>4</sup> the observed lifetime and conversion coefficients of  $\gamma_1$  lead to the conclusion that this transition is mixed electric octopole and magnetic quadrupole. The transition thus involves a parity change and a spin change of 2 units.

<sup>&</sup>lt;sup>14</sup> D. Walker and E. W. Fuller, Nature 164, 226 (1949).

<sup>&</sup>lt;sup>15</sup> S. B. Burson and K. W. Blair, Phys. Rev. 78, 89 (1950).

<sup>&</sup>lt;sup>16</sup> P. Axel and S. M. Dancoff, Phys. Rev. 76, 892 (1949).

Gamma-5 also arises from the 22-µsec. state, and to account for the fact that this transition is not observed with appreciable intensity, an l value of at least 4 is required.

Gamma-4 is the most prominent of the radiations following the  $10^{-8}$ -sec. state. An l value of 3 for this transition predicts a lifetime of  $1.5 \times 10^{-7}$  sec., which is 14 times longer than the experimental value. However, any other l assignment gives discrepancies by factors greater than 1000. The value l=3 corresponds to electric octopole or magnetic quadrupole radiation. This assignment can be checked by the measurements of the conversion coefficient. The data of Chu and Wiedenbeck give  $\alpha_k = 0.031$ , whereas the calculations of Rose *et al.*<sup>17</sup> predict 0.04 if the transition is electric octopole and 0.13 if magnetic quadrupole. The experimental value is uncertain because the branching ratio is uncertain. For this reason the discrepancy is not serious, provided the transition is mostly electric.

Either  $\gamma_2$  or  $\gamma_3$  must also originate from the  $10^{-8}$ -sec. state. For  $\gamma_3$  the value l=2 predicts a half-life of  $10^{-11}$ sec., and l=3 predicts  $7 \times 10^{-7}$  sec. For  $\gamma_2$  the choice l=2 yields  $8 \times 10^{-10}$  sec., which is in slightly better agreement with the experimental result than either choice for  $\gamma_3$ . For this reason, Fig. 1 has been drawn with  $\gamma_2$  preceding  $\gamma_3$ ; however, the order of these  $\gamma$ -rays is not important in the argument which follows.

On the basis of the level scheme of Fig. 1 and the foregoing classification of the  $\gamma$ -transitions, it is possible to draw some conclusions concerning the spins of the Ta<sup>181</sup> levels. Consideration of  $\gamma_1$ ,  $\gamma_4$ , and  $\gamma_5$  leads to a requirement for a spin change of at least 4 units between the ground state and the  $2.2 \times 10^{-5}$ -sec. state. Since the ground state of Ta<sup>181</sup> has a spin of 7/2, the upper state must have a spin of 15/2 or more. A possible assignment of spins and parities is shown in Fig. 6(a).

A level diagram which avoids the high spin assignments of 15/2 can be made, provided special assumptions about the  $\gamma$ -transitions are introduced. One such scheme is shown in Fig. 6(b). The difficulty here is that electric dipole radiation is allowed for  $\gamma_4$ , and the transition should proceed with a lifetime much shorter than  $10^{-8}$  sec. The scheme is tenable, provided only that some special property of the nucleus reduces the electric dipole matrix element by an exceedingly great amount.

#### VII. OTHER ISOTOPES

The delayed coincidence apparatus has been used to search for short-lived isomers of some other isotopes.



FIG. 6. Possible energy level schemes for Ta<sup>181</sup>. Spins and parities are indicated at the right-hand side of each scheme.

Stilbene scintillation counters were used to measure the coincidences produced by sources of 5.3-yr. Co<sup>60</sup>, 225day Ag<sup>110</sup>, 2.6-hr. Mn<sup>56</sup>, and 2.8-day In<sup>111</sup>. The delay curves produced by the  $\beta - \gamma$ -coincidences of Co<sup>60</sup>, Ag<sup>110</sup>, and Mn<sup>56</sup> were similar to the instantaneous curve (+++) in Fig. 2. It can therefore be concluded that the states of Ni<sup>60</sup>, Cd<sup>110</sup>, and Fe<sup>56</sup>, which give rise to  $\gamma$ -rays of appreciable intensity, do not have lifetimes in the range detectable by this apparatus (from about 3 or 4×10<sup>-9</sup> sec. to 2 or 3×10<sup>-7</sup> sec.).

The delay curve of coincidences produced by the In<sup>111</sup> source showed the presence<sup>18</sup> of the  $8 \times 10^{-8}$ -sec. state of Cd<sup>111</sup>, and provided the first direct evidence that the apparatus was capable of detecting metastable states.

An early version of the apparatus, using anthracene scintillation counters, was used to measure the coincidences produced by 2.7-day Au<sup>198</sup>, and 34-hr. Br<sup>82</sup>, as well as the Co<sup>60</sup>. Due chiefly to the longer decay time of the anthracene, the delay curves were wider (resolving time about  $2.3 \times 10^{-8}$  sec.) than the "no delay" curve of Fig. 3. For each of the three sources, the  $\beta - \gamma$ -delayed coincidence curves were symmetrical about the zero delay axis. Analysis of the curves indicated that no states of lifetime greater than  $10^{-8}$  sec. were playing an appreciable role in the production of the coincidences. The negative results in the case of Au<sup>198</sup> and Co<sup>60</sup> are in agreement with other recent investigations.<sup>12,18</sup>

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<sup>18</sup> M. Deutsch and W. E. Wright, Phys. Rev. 77, 139 (1950).

<sup>&</sup>lt;sup>17</sup> Rose, Goertzel, Spinrod, Harr, and Strong, Phys. Rev. 76, 1883 (1949).