with a carbon nucleus "picked up" a proton and escaped as a deuteron. The present observation of 1/14 as many deuterons as neutrons and protons may be the result of such a "pick-up" process at low energies. If this interpretation were true, one might expect to observe a few tritons also emerging from copper.

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be small.

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# The Radiations from Hafnium

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The 70-day, 45-day, 5.5-hour, and 19-second activities of Hf have been investigated by spectrometric, spectrographic, absorption, and coincidence methods. From activation experiments on the separated Hf isotopes, the following assignments have been made: 70-day-72Hf175; 19-second-72Hf179\*; 5.5-hour-72Hf180\*; 45-day-72Hf181.

70-day period.-The photographic spectrograms of the internal conversion electrons associated with the 70-day period in 72Hf175 indicated the presence of four gamma-rays. The association of Lu work functions with the various lines confirmed the hypothesis of K electron capture in  $_{72}$ Hf<sup>175</sup> leading to an excited state of  $_{71}$ Lu<sup>175</sup>.

45-day period.—An investigation of the internal conversion electron spectrum in the Argonne 180° beta-ray spectrometer indicated the presence of one beta-ray of  $0.42 \pm 0.01$  Mev and five gamma-rays. An upper limit of 1 percent may be placed on any more energetic beta-ray. Single and coincidence absorptions in lead and aluminum showed the three strongest gamma-rays and the single beta-ray. Delayed coincidence absorption indicated that the 0.42-Mev beta-ray leads directly to the 22-microsecond metastable state of 73 Ta<sup>181</sup>. No delayed gamma-coincidences were

#### INTRODUCTION

 $\mathbf{E}^{\text{XTENSIVE}}$  experiments have been carried out by several investigators<sup>1-15</sup> on the 45-day activity of 72Hf181. A longer activity of 70 days has also been reported<sup>16</sup> and assigned to 72Hf<sup>175</sup>. The results of the spectrographic measurements are generally in good agreement; but many of the coincidence measurements

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    <sup>10</sup> K. Y. Chu and M. L. Wiedenbeck, Phys. Rev. 75, 226 (1949).

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found. Thus, all gamma-rays appear to follow the 22-microsecond state. Additional coincidence measurements showed internal conversion electron groups corresponding to three of the gamma-rays. The decay scheme proposed by Chu and Wiedenbeck, as modified by Cork et al. was confirmed.

However, the binding energy of a triton to its residual

nucleus is large, so that its probability of escape would

Some of the initial photographic techniques and the

nuclear emulsion camera used in this work were

developed by Miss M. Elaine Toms.

5.5-hour period.-Activation of extremely pure Hf samples showed an activity of 5.5 hours in addition to the 45-day period. The assignment of this activity to Hf was confirmed by chemical methods. No delayed coincidences were observed. Internal conversion electron groups corresponding to five gamma-rays were resolved; absorption in lead indicated three of the photon components. Aluminum absorptions indicated coincidences between internal conversion electron groups from at least two of the gamma-rays. Activation of separated isotopes shows that this activity is one of an isomeric state of 72Hf180.

19-second period.-A photographic spectrogram of the internal conversion electrons showed the presence of a single gamma-ray; the association of Hf work functions with the four lines observed verified the hypothesis of an isomeric state in 72Hf179.

seem to be in conflict and at times have led to anomalous conclusions. The high intensity of the internal conversion electron groups from the many gamma-rays present make the interpretation of both single and coincidence absorption curves extremely difficult. We have taken advantage of the 22-microsecond isomeric state of 73Ta<sup>181</sup> to separate the primary beta-ray from the internal conversion electron spectrum, thereby facilitating careful inspection of each independently.

After the completion of many of these experiments using normal Hf, the separated isotopes were obtained from the Isotopes Division of the Oak Ridge National Laboratory. In the subsequent discussion, all references to Hf indicate normal Hf, the separated isotopes being referred to explicitly. These isotopes made possible the definite isotopic assignment of the activities. Accurate gamma-ray energy determinations were made by measurement of the internal conversion electron groups associated with each isotope. The details of the techniques and instrumentation employed in these measurements will be discussed in a subsequent publication.<sup>17</sup>

<sup>&</sup>lt;sup>1</sup> H. Neuert, Z. Naturforsch. 2a, 432 (1947). <sup>2</sup> L. Madansky and M. L. Wiedenbeck, Phys. Rev. 72, 185 (1947)

<sup>&</sup>lt;sup>17</sup> H. B. Keller and J. M. Cork, to be published.

#### THE SOURCES

The samples of normal  $HfO_2$  were prepared through the courtesy of J. J. Katz and the Chemistry Division of the Argonne Laboratory. All sources used in these experiments were activated by neutron bombardment in the heavy water moderated nuclear reactor of this laboratory with the exception of the 45-day Hf source used in the beta-ray spectrometer, which was activated in the Oak Ridge reactor. The activated powder was placed on Scotch tape previously arranged on the source holders.

#### COINCIDENCE CIRCUITS

Two concidence circuits were used. The first circuit measured only direct coincidences and had a resolving time of 0.48  $\mu$ sec. The second circuit had a resolving time variable from 1.25 to 10  $\mu$ sec in five steps. In addition, this circuit was equipped with a trigger-pair type delay circuit in one of the channels. This allowed a variable delay from 0 to 30  $\mu$ sec with six intervening positions. The counters used were of the end window type having mica windows of 2 mg/cm<sup>2</sup>.

A lead shield  $\frac{1}{4}$ -in. thick not only served as a support for the source and holder, but also reduced the probability of spurious coincidences due to either beta- or Compton scattering.

#### 70-DAY ACTIVITY

By proton and deuteron bombardment of Lu, Wilkinson, and Hicks<sup>16</sup> have produced an activity of  $70\pm 2$  days which they have attributed to  $_{72}Hf^{175}$ . From our decay curves of each of the separated Hf isotopes, we are able to confirm their conclusion as to the assignment of this activity. The spectrographic data on the gamma-ray energies for this period are set forth in Table I.

The best agreement among the gamma-ray energies computed from the various conversion electron lines was obtained when fitted with Lu work functions, confirming the hypothesis of K electron capture. This conclusion was further born out by the relatively high intensity of the Auger lines, being due to the K x-rays of high intensity. As this type of photographic data does not lend itself readily to intensity determinations, no effort was made to determine relative intensities of the lines.

Four gamma-rays have been resolved. A tentative level scheme is presented (Fig. 1). Placing the 0.0891-Mev gamma-ray above rather than below the 0.3423-Mev gamma-ray is arbitrary; none of our experiments yielded evidence which would resolve this question.

## 45-DAY ACTIVITY

### **Photon Radiations**

The values of the energies of the gamma-rays were determined by an examination of the internal conversion electron spectrum of  $_{72}$ Hf<sup>181</sup> in both the 180°

TABLE	I.	Gamma-ray	energies	from	internal	conversion
		electron s	pectrogra	m of	72Hf <sup>175</sup> .	

Electron energy	Proposed interpretation	Energy sum (Mev)	Gamma-energy (Mev)
0.0257	K	0.0891	
0.0782	$L_{I}$	0.0891	
0.0864	M	0.0889	
0.0885	N	0.0890	0.0891
0.0500	K	0.1134	
0.1025	L	0.1134	
0.1110	M	0.1135	0.1134
0.1650	K	0.2284	0.2284
0.2789	K	0.3423	
0.3314	$L_{I}$	0.3423	
0.3400	$\overline{M}$	0.3425	0.3423
0.0427	Auger L11		<i>α</i> 1
0.0440			a1: a2
0.0500	M		<i>a</i> 1
0.0511	M. N		an: an
0.0532	M'		$\alpha_2$

beta-ray spectrometer and the  $180^{\circ}$  internal conversion electron spectrograph. The values are given in Tables II and III, respectively. The energy values have been assigned from the more accurate photographic measurements on the activated enriched  $_{72}Hf^{180}$ .

Absorption of the photon radiations in Pb indicated three components (Fig. 2) that agree reasonably well with the spectrographic determinations of the three strongest groups. Finally, the photon spectrum was examined by covering the spectrometer source with an Al cap and a 40-mg Pb/cm<sup>2</sup> foil and observing the photoelectron spectrum. The results were in substantial



FIG. 1. Proposed decay scheme for 72Hf<sup>175</sup> (70-day).

TABLE II. Gamma-ray energies from internal conversion electron lines measured in the 180° beta-ray spectrometer for  $_{72}$ Hf<sup>181</sup> (normal).

Electron energy	Proposed interpretation	Energy sum (Mev)	Gamma-energy (Mev)
0.0730	L	0.084	0.084ª
0.0662	K	0.133	0.133
0.1228	L	0.134	
0.1316	M	0.134	
0.693	K	0.136	0.136
0.1240	L	0.135	
0.1334	$\overline{M}$	0.136	
0.275	K	0.342	0.342ª
0.279	K	0.346	0.346
0.333	$\overline{L}$	0.345	
0.420	K	0.487	0.488
0.478	$\hat{L}$	0.489	51200
0.554	К	0.621	0.621
0.611	$\overline{L}$	0.622	

<sup>a</sup> To be identified with 72Hf<sup>175</sup> (70 day).

agreement with those of the internal conversion electron spectrum.

## **Delayed Coincidence Measurements**

Since the counting rate for delayed coincidences increases with resolving time, it is desirable to increase the latter to a point as high as possible consistent with a tolerable accidental background. To this end a value of 7.25  $\mu$ sec was selected in conjunction with a delay of 8.5  $\mu$ sec. This is sufficiently greater than the resolving time to prevent the recording of any prompt coincidences.

Figure 3 shows the results of an aluminum absorption on the delayed side; the counter whose response was not delayed received all radiations not stopped by the counter window (2 mg mica/cm<sup>2</sup>). The presence of but a single component of 10.5 mg Al/cm<sup>2</sup> is evident. A check absorption in Be as well as the notably high coincidence rate eliminated the possibility of this being an uncharged radiation. Several points taken with



FIG. 2. Absorption in Pb of Hf gamma-rays.

TABLE III. Gamma-ray energies from internal conversion electron spectrogram for 72Hf<sup>181</sup> (activated enriched 72Hf<sup>180</sup>).

Electron energy	Proposed interpretation	Energy sum (Mev)	Gamma-energy (Mev)
0.0655	K	0.1329	
0.1217	$L_{\rm I, II}$	0.1329	
0.1230	$L_{III}$	0.1329	
0.1303	M	0.1330	
0.1322	N	0.1328	0.1329
0.0685	K	0.1359	
0.1245	$L_{\rm L,II}$	0.1357	
0.1335	M	0.1362	0.1358
0.2773	K	0.3447	
0.3330	$L_{\rm LH}$	0.3442	
0.3408	M	0.3435	0.3441
0.4135	K	0.4809	
0.4696	$L_{1 II}$	0.4808	
0.4780	M	0.4807	0.4808
0.5438	K	0.6112	0.6112

thicker absorbers than are shown indicated no harder component and apparently a complete absence of coincidences involving delayed gamma-rays. The conclusions from these experiments are that the 22- $\mu$ sec metastable state follows directly upon a beta-decay from  $_{72}$ Hf<sup>181</sup> to  $_{73}$ Ta<sup>181</sup>, and that the energy of the beta-ray is about 0.4 Mev. Thus, all gamma-ray transitions follow the 22- $\mu$ sec metastable state to lower states in  $_{73}$ Ta<sup>181</sup>.

Figure 4 shows the results of a similar absorption on the undelayed side. The thinness of the source and counter windows accentuated the characteristic shape of the monoenergetic electron groups, permitting a relatively good estimation of their ranges. The electron group energies as assigned in Table IV agree quite well with the three strongest gamma-rays previously discussed. In obtaining the energy values of the electron groups from our calibration curves the 2-mg mica/cm<sup>2</sup> counter window was assumed to have the same absorption characteristics as aluminum.



FIG. 3. Absorption in Al of  $_{72}$ Hf<sup>181</sup> (45-day)  $\beta$ ,  $e^-$  coincidences with  $\beta$ -counter delayed 8.5 microseconds. Absorption on delayed side,  $\tau = 7.25$  microseconds.

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Compo- nent	Range (mg Al/cm²) (corrected)	Electron energy (Mev)	Shell in Ta	Energy sum (Mev)	Probable $\gamma$ -ray (Mev)
A	14	0.090	K	0.157	0.1329
B	31	0.150	L	0.161	0.1329
C	75	0.300	L K	0.367	0.3441
D	96	0.350		0.361	0.3441
$\tilde{E}$	115	0.395	K	0.467	0.4808

TABLE IV. Analysis of electron group energies.

### **Prompt Coincidences**

With no filter before the 2-mg mica/cm<sup>2</sup> window of one counter, an absorption in aluminum on the second counter yielded a curve for prompt coincidences (resolving time 0.48  $\mu$ sec) that shows three electron groups, which from their ranges probably correspond to the gamma-rays of 0.4808, 0.3441, and 0.1329, and/or 0.1358 Mev. The structure of this curve (Fig. 5) indicates that most of the prompt coincidences are between internal conversion electrons.

When a filter of 15.6 mg Al/cm<sup>2</sup> (which with the Scotch tape source backing gave an effective filter thickness of about 23.4 mg Al/cm<sup>2</sup>) was placed before the first counter, the absorption curve showed the intensities of the two harder components to be reduced to  $\sim$ 5 percent relative to the softer group, which alone remained clearly distinguishable (Fig. 6). From this one may conclude that relatively few of the coincidences are ones between two electrons of high energy.

Furthermore, if an undelayed beta-ray branch of energy comparable to that of the delayed branch represented any appreciable fraction of the disintegrations, it would have stood out clearly when the filter was inserted, for the low resolving time of the coincidence circuit suppressed the delayed coincidence rate to approximately 0.8 percent of the integrated rate.

One further experiment was done in an attempt to find an undelayed branch. A source was placed in coincidence geometry with a filter of 1.885 g Pb/cm<sup>2</sup> between the source and one counter. This filter reduced to a negligible intensity the soft gamma-rays



FIG. 4. Absorption in Al of  $_{72}$ Hf<sup>181</sup> (45-day)  $\beta$ ,  $e^-$  coincidences with  $\beta$ -counter delayed 8.5 microseconds. Absorption on undelayed side,  $\tau = 7.25 \,\mu$ sec.

FIG. 5. Absorption in Al of  $_{72}$ Hf<sup>181</sup> (45-day)  $e^--e^-$  coincidences.

and x-rays and attenuated the 0.3441-Mev gammaray to 50 percent and the 0.4808-Mev gamma-ray to 77 percent. Between the source and the other counter a composite filter was placed. This filter consisted of the Scotch tape source mounting,  $10.8 \text{ mg Ta/cm}^2$ , and  $6.9 \text{ mg Al/cm}^2$ . The entire filter was approximately equivalent to 30 mg Al/cm<sup>2</sup>, which was sufficient to eliminate the soft conversion electrons from the 0.1329and 0.1358-Mev gamma-rays. The Ta was included to preferentially remove the L x-rays. This arrangement would permit coincidences to be recorded only if they were between one of the two harder gamma-rays and beta-rays or electrons with sufficient energy to penetrate the filter. The filter reduced the beta-ray associated with the delayed state to about 12 percent. Since a beta-ray associated with the undelayed branch would be probably expected to have as much energy as the delayed beta-ray, this arrangement was quite sensitive to prompt beta-gamma-coincidences, especially considering that the short resolving time of the circuit  $(1.25 \ \mu sec)$  already suppressed the delayed coincidences to 4 percent. After determining the coincidence rate with no delay, the beta-counter was delayed 1.75  $\mu$ sec and the coincidence counting rate due to the delayed branch determined. Correction for X-gamma- and gamma-gamma-coincidences (determined with a thick Al filter before the beta-counter) allowed all betagamma-coincidences observed in the nondelayed condi-



FIG. 6. Absorption in Al of  $_{72}$ Hf<sup>181</sup> (45-day)  $e^- - e^-$  coincidences.

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FIG. 7. Absorption in Al of 72Hf<sup>181</sup> (45-day) β-rays.

tion to be attributed to the delayed branch. The low counting rate did not permit good statistics to be obtained; hence the experiment permits us only to place an upper limit of about 5 percent on a possible undelayed branch.

### **Beta-Spectrum**

The presence of internal conversion electrons in the neighborhood of the beta-end point prevented an exact determination of the beta-ray energy in the  $180^{\circ}$  spectrometer. However, an extrapolation of a Fermi-Kurie plot of the beta-ray continuum measured in the spectrometer gave a beta-ray end point of  $0.420\pm0.010$  Mev. No harder beta-ray was observed. The sensitivity of the spectrometer would have allowed the detection



FIG. 8. Proposed decay scheme for 72Hf<sup>181</sup> (45-day).

of a harder beta-ray component of intensity > 1 percent of the 0.420-Mev beta-ray.

A careful single beta-ray absorption in aluminum was made (Fig. 7). By attributing about 5 percent of the radiation to internal conversion electrons from the combined 0.3441- and 0.4808-Mev gamma-rays and by superimposing the  $e^--e^-$  coincidence curve to obtain an approximation to the shape of the electron groups, the absorption reduced to a single beta-ray of 11 mg Al/cm<sup>2</sup> half-thickness, which corresponds to the delayed 0.4-Mev beta-ray.

### **Decay Scheme**

The decay scheme for  $_{72}$ Hf<sup>181</sup> as originally proposed by Chu and Wiedenbeck<sup>10</sup> and modified by Cork *et al.*<sup>13</sup> and by Barber<sup>15</sup> is confirmed. It is shown in Fig. 8. The energy levels assigned to  $_{73}$ Ta<sup>181</sup> show excellent

TABLE V. Gamma-ray energies from internal conversion electron spectrogram for 72Hf<sup>180</sup> (5.5-hr) (activated enriched 72Hf<sup>179</sup>).

Electron energy	Proposed interpretation	Energy sum (Mev)	Gamma-energy (Mev)
0.0460	L	0.0568	
0.0473		0.0569	
0.0542	M	0.0568	0.0568
0.0277	K	0.0931	
0.0824	LTI	0.0932	
0.0837		0.0933	
0.0906	$\overline{M}$	0.0932	
0.0926	$\widetilde{N}$	0.0931	0.0932
0.1485	K	0.2139	
0.2032	LTT	0.2140	
0.2116	$\overline{M}$	0.2142	0.2140
0.2649	K	0.3303	
0.3196	$\overline{L}_{I,II}$	0.3304	0.3304
0.3763	K	0.4417	
0.4317	$\overline{L}_{I,II}$	0.4425	0.4420

internal consistency, the maximum deviation of any sum being about 0.3 percent.

## 5.5-HOUR ACTIVITY

A period of  $5.5\pm0.1$  hours was identified in our sources.<sup>18</sup> No period comparable to 5.5 hours has been reported in Zr, the only spectrographically detected impurity in the sample. We further excluded this possibility by activating a pure Zr sample; no such activity was observed. Because of its analogous chemistry, Ti was similarly examined, again with negative results.

To establish the Z assignment more definitely, part of a bombarded  $HfO_2$  sample was submitted to several Hf chemical separations and the ratio of the 5.5-hour to 45-day activity in the separated fraction determined from its decay curve.<sup>19</sup> A similar comparison was made

<sup>19</sup> See Appendix.

<sup>&</sup>lt;sup>18</sup> This activity, earlier designated  $\sim 6$  hours, was first observed in this Laboratory by C. O. Muchlhause.

on the remaining portion of the originally activated sample. The observed 5.5 hr/45 day ratio for the separated fraction was  $1.57\pm0.05$  and for the fraction of the original  $1.55\pm0.03$ . The chemical procedure was highly specific for Hf, so that it would be very unlikely that the ratios of the activities would remain constant unless both periods were in the same element. Finally, upon activation of the various separated isotopes the ratio of the 5.5-hr to 45-day periods was found to be greatly increased upon bombardment of enriched  $_{72}$ Hf<sup>179</sup>; the period was thus assigned to  $_{72}$ Hf<sup>180</sup>.

# **Photon Radiations**

All of the internal conversion electron lines found to be associated with the 5.5-hr activity yielded the best energy agreement when fitted with Hf work functions. Five gamma-rays were identified, and the spectrographic results are tabulated in Table V. The associa-



FIG. 9. Decay of Hf  $e^- -\beta$ -coincidences (with and without 10  $\mu$ sec delay).

tion of Hf work functions with these gamma-rays leads to the conclusion that the 5.5-hr activity is an isomeric state of  $_{72}$ Hf<sup>180</sup>. In the lead absorption curve, Fig. 2, three of the gamma-rays have been resolved. Prior to the time when the enriched isotopes became available, the striking similarity between this curve and the corresponding lead absorption for the 45-day photon radiations led to considerable difficulty regarding the isotopic assignment of the activity.

## Coincidences

The decay of the beta- $e^-$  coincidences (no filters between the source and counters) was followed under two conditions: with neither channel delayed and with one channel delayed 10  $\mu$ sec. Figure 9 shows that only prompt coincidences were found to be associated with the 5.5-hour activity.

An absorption in Al of the prompt gamma- $e^-$  coin-

AI ASSOREER / 1/2 = 7 FILTER HI SOURCE HI SOURCE HI SOURCE D 40 60 60 100 120 10 160 100 200 220 240 260 260 500

FIG. 10. Absorption in Al of  $_{72}$ Hf<sup>180</sup> (5.5-hr)  $\gamma - e^-$  coincidences.

cidences was made using a fixed filter of 32 mg Pb/cm<sup>2</sup> between the source and one of the counters. This filter not only suppressed all internal conversion electrons except those from the hardest gamma-ray, but also served to further accentuate the counter sensitivity to it by acting as a radiator for the unconverted gammarays. The filter was thin enough so that, aside from Lx-rays, none of the other uncharged radiations was appreciably attenuated. The resulting absorption curve in Fig. 10 is resolved into two components. The more energetic component appears to be an electron group with an approximate range of 110 mg Al/cm<sup>2</sup>, which corresponds to the K shell internal conversion electrons from the 0.442-Mev gamma-ray. Note the excellent agreement between the range of this component as observed in the coincidence curve and the corresponding component as observed in the single aluminum absorption (Fig. 11). This component evidently represents coincidences of the K conversion electrons from the 0.442-Mev gamma-ray with related K x-rays or other gamma-rays associated with the same disintegration. The low relative intensity of the component (about 10 percent) relative to the softer group is consistent with this view, especially when the much lower sensitivity of the counter for uncharged radiation is considered. The accuracy of the points does not permit further detailed analysis of the curve. It is clear, how-



FIG. 11. Absorption in Al of 72Hf180 (5.5-hr) radiations.

Electron energy	Proposed interpretation	Energy sum (Mev)	Gamma energy (Mev)
0.0957	K	0.1611	
0.1500	$L_{\rm LII}$	0.1608	
0.1512		0.1608	
0.1587	M	0.1613	0.1610

TABLE VI. Gamma-ray energy from internal conversionelectron spectrogram of 72Hf179 (19-sec).

ever, from the apparent range of the softer group that conversion electrons from the 0.330-Mev gamma-ray are present. Although there may be contributions to this group from the other gamma-rays of lower energy, their resolution would not be possible. It is fairly safe to conclude that the 0.442 and 0.330 gamma-rays are in coincidence.

#### Single Absorption in Al

A careful absorption in Al (Fig. 11) of the charged component of the radiations using the enriched 72Hf<sup>179</sup> resolved into three components. The statistical errors of all points were less than 0.5 percent and the counter sensitivity was frequently checked with a standard during the run. The source was mounted on a backing of 0.7 mg/cm<sup>2</sup> to minimize back-scattering. The accuracy and large number of points permit a reasonably reliable analysis. The range ( $\sim 108 \text{ mg Al/cm}^2 - 0.36$ -Mev electron group) of the most energetic electron group, distinguishable as such by its characteristic shape, is in good agreement both with the corresponding portion of the coincidence curve (Fig. 10), and with the range one would expect to observe for the K shell conversion of the 0.442-Mev gamma-ray. Because of the extrapolations necessary, the succeeding analysis is, of course, not as reliable. The two groups which have been resolved (Fig. 11) are in good agreement with the groups identified in the spectrographic plates which correspond to the K shell conversion lines from the 0.330-Mev and the 0.214-Mev gamma-rays.

This analysis would not of itself present a convincing argument in support of the hypothesis of an isomeric transition, but the fact that an analysis is possible which is consistent with the remaining body of information gives the curve substantiative merit.

### **Decay Scheme**

Since none of the gamma-ray energies combine to form a significant sum with any of the others, no effort has been made to specify a level scheme for this isomeric state of  $_{72}$ Hf<sup>180</sup>. The prompt gamma- $e^-$  coincidence absorption curve tends to indicate that the two hardest components (0.442 and 0.330 Mev) are in cascade.

Bendel *et al.*<sup>20</sup> have reported observing a K electron capture branch in the 8.15-hr activity in  $_{73}$ Ta<sup>180</sup>. One of the gamma-rays associated with this process was assigned an energy of 0.092 Mev. We suggest the possi-

bility that this radiation is identical with the 0.0932-Mev gamma-ray present in the 5.5-hr activity of  $_{72}$ Hf<sup>180</sup>. Their failure to observe any of the other gamma-rays which we have assigned to this activity may indicate that the  $_{73}$ Ta<sup>180</sup> nucleus decays to one of the lower energy levels of  $_{72}$ Hf<sup>180</sup>, rather than to the 5.5-hr state. If these hypotheses are correct, we would be inclined to place the 0.0932-Mev gamma-ray at or near the bottom of the level scheme.

It is also worthy of note that the assignment of this activity to an isomeric state of  $_{72}$ Hf<sup>180</sup> is in violation of the general rule of Mattauch which states that isomeric states are not in general found in isotopes with both even atomic number and even mass number.

### **19-SECOND ACTIVITY**

In 1940, Flammersfeld<sup>21</sup> reported a 19-sec activity in Hf which he deduced to be an isomeric state of either  $_{72}$ Hf<sup>177</sup> or  $_{72}$ Hf<sup>179</sup>. By absorption measurements, he concluded the energy of the radiation to be about 0.190 Mev.

From the activation experiments on the enriched Hf isotopes, the activity was found to be present in  $_{72}$ Hf<sup>179</sup>.

A spectrogram of the internal conversion electron lines was obtained by 50 successive bombardments and exposures of a source of enriched  $_{72}Hf^{179}$ . Four lines, attributable to a gamma-ray of 0.160 Mev, were found. The work functions used were those of Hf, thus confirming the hypothesis of isomerism. The results of the experiment are shown in Table VI.

Note added in proof:—Through the courtesy of Dr. Bernard Hamermesh (Argonne National Laboratory) an examination of the unconverted photon radiations of the 19-sec activity was made using his NaI (Tl-activated) proportional counter and a 20-channel pulse-height discriminator. Two peaks were resolved. The more intense peak corresponded to a gamma-ray of 0.217 ( $\pm 0.005$ ) Mev, while the less intense peak represented the highly converted 0.160-Mev component. The former value is in excellent agreement with that reported by der Mateosian and Goldhaber.<sup>22</sup> These authors have also recently reported evidence indicating coincidences between the two gamma-rays.

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#### APPENDIX

#### Chemical Procedure

Several mg of very pure activated HfO<sub>2</sub> were dissolved in HF and the excess fumed off with H<sub>2</sub>SO<sub>4</sub>. The solution was diluted to 10 ml, 10-mg inactive Hf added as carrier, Hf(OH)<sub>4</sub> precipitated with 6M NH<sub>4</sub>OH, and the hydroxide washed several times with dilute NH<sub>4</sub>OH. The precipitate was dissolved in 10 ml of 10 percent H<sub>2</sub>SO<sub>4</sub>, 8 ml of 10 percent (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> and 1 ml conc. H<sub>2</sub>SO<sub>4</sub> added, and the solution heated to 40–50°C to effect precipitation of HfO(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub>. The latter was centrifuged, washed with cold 5 percent NH<sub>4</sub>NO<sub>3</sub>, and ignited at 1200°C for a short time.

<sup>&</sup>lt;sup>20</sup> Bendel, Brown, and Becker, Phys. Rev. 81, 300 (1951).

<sup>&</sup>lt;sup>21</sup> A. Flammersfeld, Naturwiss. 32, 68 (1944).

<sup>&</sup>lt;sup>22</sup> E. der Mateosian and M. Goldhaber, Phys. Rev. 82, 115 (1951).