FIG. 7. Probable decay scheme of  $Ba^{133}$ .

integration and that the counting efficiency of these x-rays is one percent, this cross section for normal barium is at least 0.006 barn (corresponding to the minimum half-life of 20 years). If assignment is made to mass 129, then it could only have been formed from stable  $Ba^{130}$ , whose abundance is 0.10 percent, by an  $(n, 2n)$  reaction. This would mean that the isotopic cross section for this process is at least 6 barns, which is ex-

tremely improbable since most of the pile neutrons are of low energy. Furthermore,  $Ba^{129}$  would decay to an active  $Cs^{129}$ , and no such activity could be detected. Assignment of the long-lived Ba to mass 131 is ruled out by the fact that it does not decay to the 10.2d  $Cs^{131}$ . The only possible remaining assignment is to mass 133. In this case the isotope could be formed from stable  $Ba^{132}$ , whose abundance is also 0.10 percent, by an  $(n, \gamma)$  reaction. The isotopic cross section for this process is then a minimum of about 6 barns. Thus an assignment to mass 133 is fairly certain. The 37.8h Ba which is also assigned to mass 133 is known to decay by isomeric transition. The lower state can now be identified as this  $>20$ y  $Ba^{133}$ . The decay scheme is probably as shown in Fig. 7.

This paper is based on work performed at the Metallurgical Laboratory of the University of Chicago under Contract W-7401-eng-37 for the Manhattan Project, and the information contained therein will appear in Division IV of the Manhattan Project Technical Series. Counting of the long-lived  $Ba^{133}$  is continuing at Los Alamos.

## Coincidence Measurements. Part I. Beta Spectra

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The necessary and sufficient conditions for a simple  $\beta$  spectrum (not accompanied by internally converted  $\gamma$  radiation) are discussed. It is generally considered that the  $\beta$ - $\gamma$  coincidence rate per recorded  $\beta$  particle decreases with an increase of absorber thickness for a complex  $\beta$  spectrum. Although this is the most common condition, it is possible for the ratio to remain constant or even rise as the  $\beta$  absorber thickness is increased.

The ratio,  $N_{\beta\gamma}/N_{\beta}$ , is also calculated for simple spectra followed by non-delayed conversion electrons as well as for conversion in metastable levels of lifetime long compared with the resolving time of the coincidence circuit.

Data have been obtained for several nuclei including antimony<sup>124</sup> (60 days), cesium<sup>134</sup> (1.7 years), europium<sup>154</sup> (5-8 years), gold<sup>198</sup> (2.7 days), hafnium<sup>181</sup> (55 days), iridium<sup>192,194</sup> (60 days), mercury<sup>203,205</sup> (51.5 days), osmium<sup>193</sup> (17 days), and potassium<sup>42</sup> (12.4 hours).

### 1. INTRODUCTION

**I**N Part II of this set of articles we shall indicate a simple and accurate method for determining the total absolute conversion coefficients of nuclear levels. This method demands little knowledge of the  $\gamma$  radiation involved in

the transition if the  $\beta$  spectrum is simple but a considerable amount of information must be available if the  $\beta$  spectrum is complex. Since little is known about the structure of most complex  $\beta$  emitters we have limited our investigation to nuclei which have simple  $\beta$  spectra.

It was therefore thought advisable to consider first the nature of the  $\beta$  radiation of several nuclei to which this method seems applicable.

Two methods have commonly been used to determine whether a  $\beta$  spectrum is simple or complex. The first can be obtained from the energy distribution of the ejected  $\beta$  particles as obtained with a  $\beta$  spectrograph. The symmetry of this distribution as well as the linearity of the Fermi plot are then used as the criteria of a single  $\beta$  transition. This method is somewhat difficult and cannot be used for energies below  $\sim 0.2$  or  $0.3$  Mev due to the distortion of the  $\beta$  distribution by scattering and window absorption. The spectrograph method also becomes difficult for short-lived materials.

The second method involves the study of the number of  $\beta$ - $\gamma$  coincidences per observed  $\beta$  particle as a function of the  $\beta$  absorber thickness. This is a simple procedure which can be carried out with nuclei which emit low energy  $\beta$  radiation. It can also be applied to short lived nuclei (possibly to lifetimes as short as several seconds). Such coincidence measurements were carried out by Langer, Mitchell, and McDaniel<sup>1</sup> as early as 1939. Since then a considerable amount of work has been carried out by the Indiana group on the level structure of nuclei by these methods.

We shall attempt to examine the coincidence conditions for simple and complex spectra which

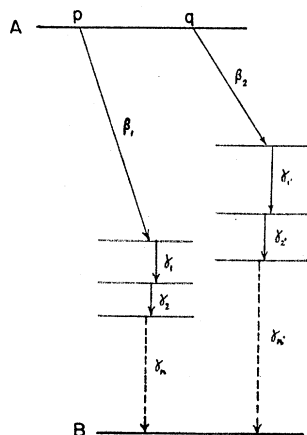


FIG. 1. Decay of nucleus  $A$  to nucleus  $B$  by complex  $\beta$ -spectra.  $p$  is the fraction of nuclei which emit  $\beta$  rays of energy  $\beta_1$ .  $q = 1 - p$ .

<sup>1</sup>L. M. Langer, A. C. G. Mitchell, and P. W. McDaniel, Phys. Rev. 56, 422 (1939).

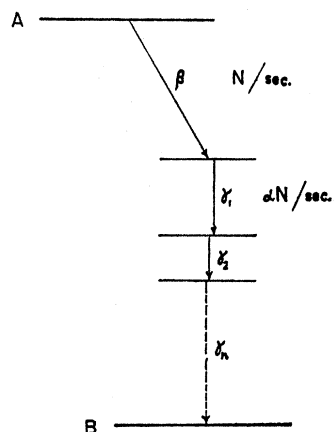


FIG. 2. Simple  $\beta$  decay followed by a series of  $\gamma$  rays.  $\gamma_1$  is internally converted. The conversion coefficient is  $\alpha$ .

do not involve internal conversion, as well as simple spectra followed by converted  $\gamma$  radiation.

### 2. THEORY

Let us assume that nucleus  $A$  decays by  $\beta$  emission to nucleus  $B$  as indicated in Fig. 1. The fraction of nuclei which emit  $\beta$  particles of maximum energy  $\beta_1$  is given by " $p$ ," those decaying with the maximum energy  $\beta_2$  is " $q$ ." A series of  $\gamma$  rays follows each  $\beta$  disintegration. These are designated  $\gamma_1, \gamma_2 \dots \gamma_n$  for the former case and  $\gamma_1', \gamma_2' \dots \gamma_n'$  for the latter case.

An arrangement is now set up with the source of radiation fixed rigidly to a  $\gamma$  ray counter. The source also faces a  $\beta$  counter. Absorbers can be placed between the source and the  $\beta$  counter.

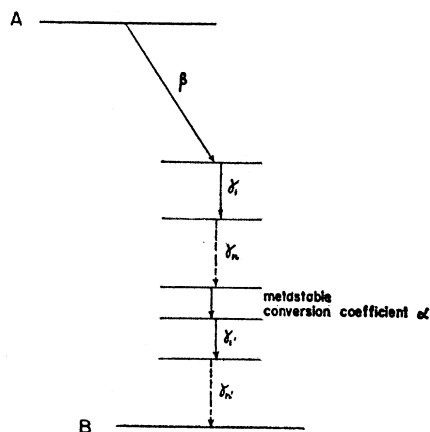


FIG. 3. Simple  $\beta$  spectrum followed by  $\gamma$  rays. One  $\gamma$  ray is internally converted and delayed.

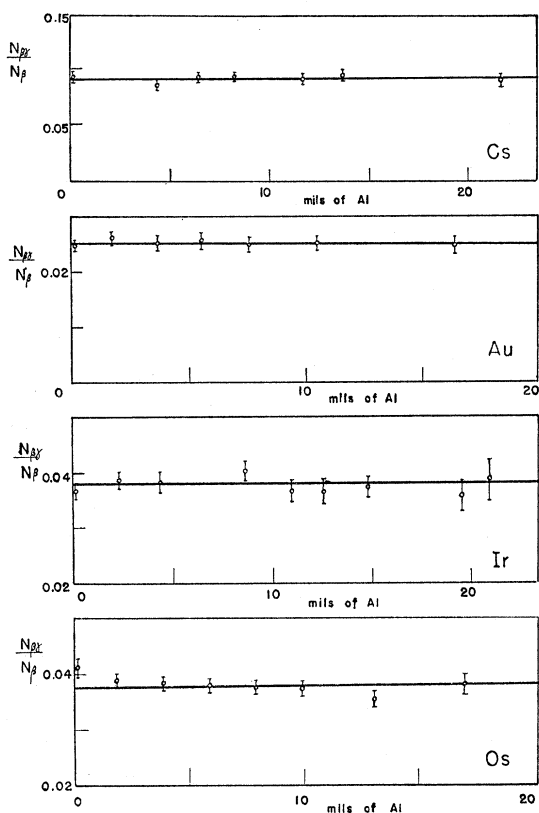


FIG. 4.  $\beta$ - $\gamma$  coincidence rate per observed  $\beta$  particle as a function of absorber thickness. These four nuclei appear to have simple  $\beta$  spectra.

The fraction of  $\beta_1$  and  $\beta_2$  particles transmitted by the absorber is given by  $F_1$  and  $F_2$ , respectively. In addition, it is assumed that the efficiency of the  $\gamma$  counter is small so that the number of  $\gamma$  rays recorded is directly propor-

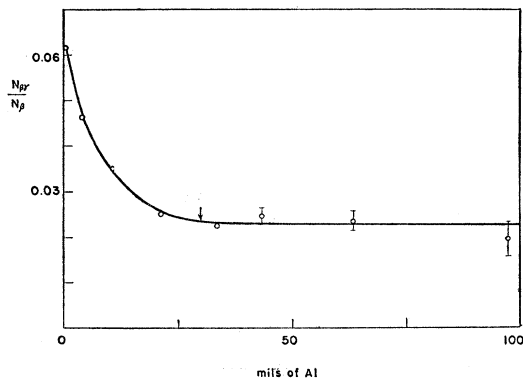


FIG. 5. Coincidence rate for  $Sb^{124}$  showing a complex  $\beta$  spectrum.

tional to the number passing through the counter. (In practice the efficiency is  $\sim 1$  percent or less.)

Further, the fraction of the sphere subtended by the  $\beta$  and  $\gamma$  counters is  $\sigma_\beta$  and  $\sigma_\gamma$ , respectively. Their respective efficiencies being  $\epsilon_\beta$  and  $\epsilon_{\gamma n}$ . (The efficiency of the  $\gamma$  counter depends upon the energy of the  $\gamma$  radiation.)

The ratio

$$R_1 = N_{\beta\gamma} / N_\beta = \left[ pF_1 \left( \sum_1^n \epsilon_n \right) + qF_2 \left( \sum_1^{n'} \epsilon_{n'} \right) \right] \sigma_\gamma / [pF_1 + qF_2].$$

It is clear that for a simple spectrum, i.e.,  $p$  or  $q=0$ , without internal conversion,  $R_1 = \text{constant}$ , since the ratio is independent of  $F$ .

**Complex Spectra**

For the complex spectrum in Fig. 1 we must consider several cases. First, if

$$\left( \sum_1^{n'} \epsilon_{n'} \right) > \left( \sum_1^n \epsilon_n \right)$$

the ratio,  $R_1$ , will decrease until  $F_2$  becomes 0. Thereafter it will remain constant at a value,

$$R_1 = \left( \sum_1^n \epsilon_n \right) \sigma_\gamma.$$

The point at which the curve reaches a constant value gives the energy of  $\beta_2$ , the lower energy  $\beta$  ray. This is by far the most common case.

On the other hand, if

$$\left( \sum_1^{n'} \epsilon_{n'} \right) < \left( \sum_1^n \epsilon_n \right),$$

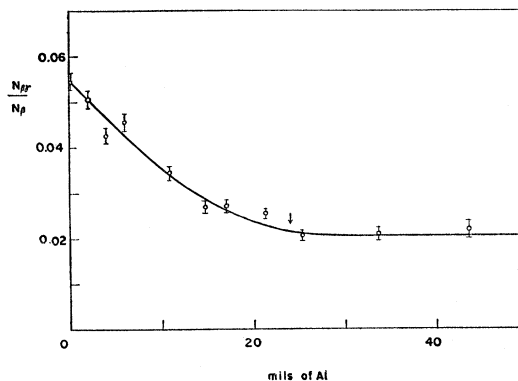


FIG. 6. Complex  $\beta$  spectrum of  $Eu^{154}$ .

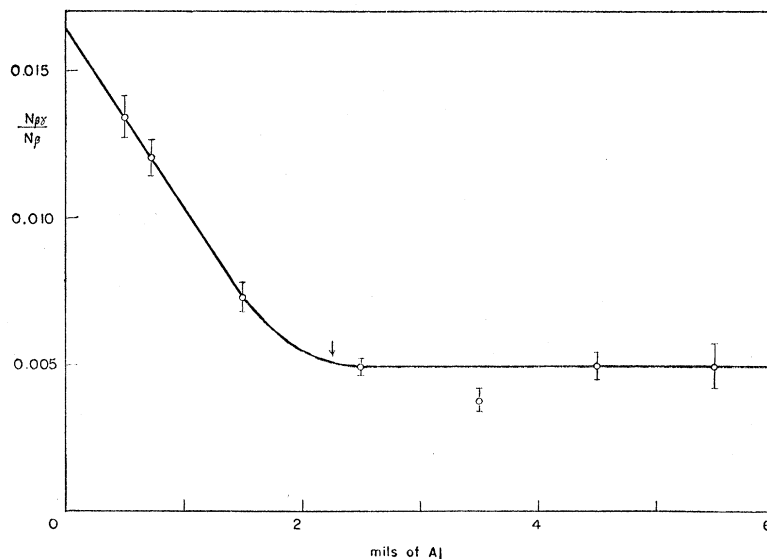


FIG. 7. Coincidence rate curve for  $Hg^{203,205}$ .

the ratio  $R_1$  will rise with an increase in absorber thickness becoming

$$R_1 = \left( \sum_1^n \epsilon_n \right) \sigma_{\gamma}$$

at the point where no  $\beta_2$  are transmitted through the absorber.

The third case to consider occurs if

$$\left( \sum_{1'}^{n'} \epsilon_{n'} \right) = \left( \sum_1^n \epsilon_n \right).$$

This would give rise to  $R_1 = \text{constant}$  and as such

$$R_2 = N_{\beta\gamma}/N_{\beta} = \left\{ F_1 \left[ (1-\alpha)\epsilon_1 + \sum_2^n \epsilon_n \right] + F_2 \alpha \sum_2^n \epsilon_n \right\} \sigma_{\gamma} - \frac{1}{2} \alpha F_1 F_2 \sigma_{\beta} \epsilon_{\beta} \sigma_{\gamma} \left( \sum_2^n \epsilon_n \right) / \left[ F_1 + \alpha F_2 \right] - \alpha F_1 F_2 \sigma_{\beta} \epsilon_{\beta} / 2.$$

The last term in the numerator as well as in the denominator arises from the possibility that both the  $\beta$  particle and the conversion electron enter the  $\beta$  counter which, however, records only one particle. In practice  $\sigma_{\beta} \epsilon_{\beta} / 2$  may be  $\sim 0.05$  or less and as such these terms can be neglected. Thus  $R_2$  would appear similar to  $R_1$  (a complex spectrum) except for the cases (a)  $\alpha \sim 1$ , (b)  $\alpha \ll 1$  or (c) if  $n \gg 1$ . In the above-mentioned cases  $R_2 \simeq \text{constant}$ . Several converted lines each having a conversion coefficient much less than unity would also appear as a true simple spectrum.

would appear to be a simple spectrum. Therefore, if no internal conversion exists, the constancy of  $R_1$  is a necessary condition for a simple  $\beta$  spectrum but not a sufficient condition.

### Effect of Internal Conversion

For a simple  $\beta$  spectrum followed by a series of  $\gamma$  rays, one of which is internally converted (but not delayed) with a conversion coefficient  $\alpha$  (Fig. 2), we have a similar relationship.  $F_1$  is again the fraction of the  $\beta$  radiation transmitted through the absorber and  $F_2$  the corresponding fraction of the conversion electrons.

### Metastable States

In Fig. 3 we have indicated a simple  $\beta$  spectrum followed by a series of  $\gamma$  rays, one of which has a lifetime greater than the resolving time of the coincidence circuit. The conversion coefficient is  $\alpha$ ,  $F_1$  is the fraction of the  $\beta$  rays transmitted through the absorber and  $F_2$  the corresponding fraction of the conversion electron.

Then

$$R_3 = N_{\beta\gamma}/N_{\beta} = \left[ F_1 \left( \sum_1^n \epsilon_n \right) + \alpha F_2 \left( \sum_{1'}^{n'} \epsilon_{n'} \right) \right] \sigma_{\gamma} / F_1 + \alpha F_2$$

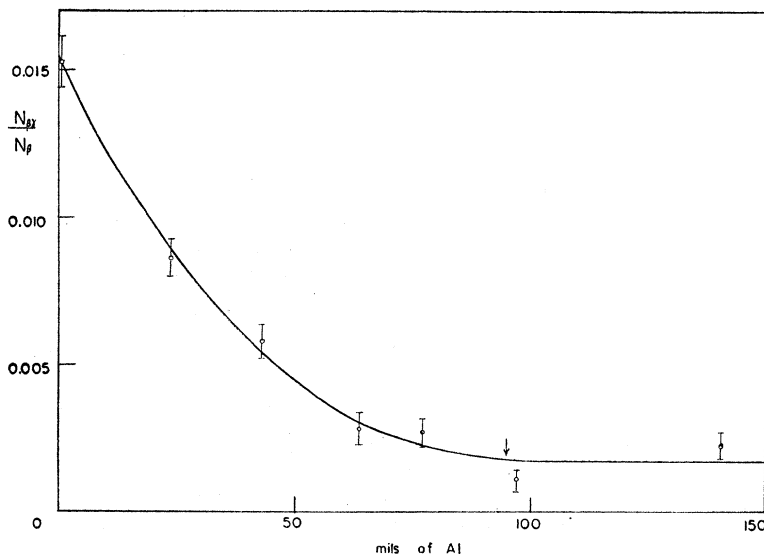


FIG. 8.  $\beta$ - $\gamma$  coincidences of  $K^{42}$ . The end point of the lower energy component of  $\beta$  radiation is not sharp but appears to be  $\sim 1.5$  Mev.

similar to the case of a complex  $\beta$  spectrum. Again the shape of the curve is determined by the values of

$$\left( \sum_1^n \epsilon_n \right) \text{ and } \left( \sum_{1'}^{n'} \epsilon_{n'} \right).$$

One considerable difference exists, however, most generally for a complex  $\beta$  spectrum: the sum,

$$\left( \sum_{1'}^{n'} \epsilon_{n'} \right),$$

for the set of  $\gamma$  rays associated with the lower energy  $\beta$  particle is greater than the corresponding sum for the higher energy particle, thus causing a decrease in  $R_1$  with an increase in absorber thickness. For delayed radiation, on the other hand, the converted electrons should be expected to be of low energy and may be followed by few  $\gamma$  rays. As such it may often be found that  $R_3$  will rise as the absorber thickness is increased.

We see, therefore, that a simple  $\beta$  spectrum gives rise to a value  $N_{\beta\gamma}/N_{\beta} = \text{constant}$ , while a complex spectrum may give rise to any shape of curve. This method can then be used to identify many of the complex spectra and to indicate which are possibly simple. Whenever possible it is desirable to have additional proof of the simplicity from the spectrograph method.

### 3. EXPERIMENTAL

The nature of the  $\beta$  spectra has been investigated for a group of nuclei having relatively long half-lives. Many of these were produced by slow neutron bombardment at Oak Ridge, while some were produced by deuteron bombardment.

For the coincidence measurements, self-quenching argon-ether counters of brass construction were used. The radiation was admitted through an end window of  $\frac{1}{2}$  to 1 mil Cellophane. When it was desired to count only  $\gamma$  radiation, a small plug could be fitted into the window frame. The background of these counters without shielding was normally  $\sim 30/100$  seconds.

Each counter was attached to a fast scaling unit so that the individual counting rate was at all times available. Sharp pulses (rise time  $\sim 0.1$  microsecond) from a pulse former before the scaler were fed to a coincidence circuit. The resolving time of this circuit could be varied between  $\sim 0.25$  to 2.5 microseconds. Since the variation in the time of the initiation of a discharge in counters may be  $\sim 0.3 \rightarrow 0.4$  microsecond, the resolving time ordinarily used was  $\sim 0.6$  microsecond.

#### Simple Spectra

Of the group of nuclei studied, cesium<sup>134</sup>, gold<sup>198</sup>, iridium<sup>192,194</sup>, and osmium<sup>193</sup> appear to be simple, i.e., the ratio  $N_{\beta\gamma}/N_{\beta} = \text{constant}$ . These curves are shown in Fig. 4. Samples of

these four elements were obtained from Oak Ridge. Some samples of gold were also produced by deuteron bombardment with the Michigan cyclotron.

The upper limit of the  $\beta$  spectrum of  $\text{Cs}^{134}$  has been found by Kalbfell and Cooley<sup>2</sup> to be 0.9 Mev. Using Feather's rule, we obtain a value of 0.8 Mev for the upper limit.

The  $\beta$ - $\gamma$  coincidence rate of gold<sup>198</sup> has been studied by Clark<sup>3</sup> who found that the coincidence rate did not decrease with an increase in absorber thickness. More recently Siegbahn<sup>4</sup> has made a study of the shape of the  $\beta$  spectrum and has found that it is simple. As is seen in Fig. 4, we find that the  $\beta$ - $\gamma$  rate is not dependent upon absorber thickness. The upper limit of the spectrum from the absorption end point of 0.332 g/cm<sup>2</sup> is 0.91 Mev.

Both the iridium<sup>192,194</sup> (60 days) and osmium<sup>193</sup> (17 days)  $\beta$ - $\gamma$  rates appear to be independent of the absorber thickness within the limit of error. However, it may be of significance that the first

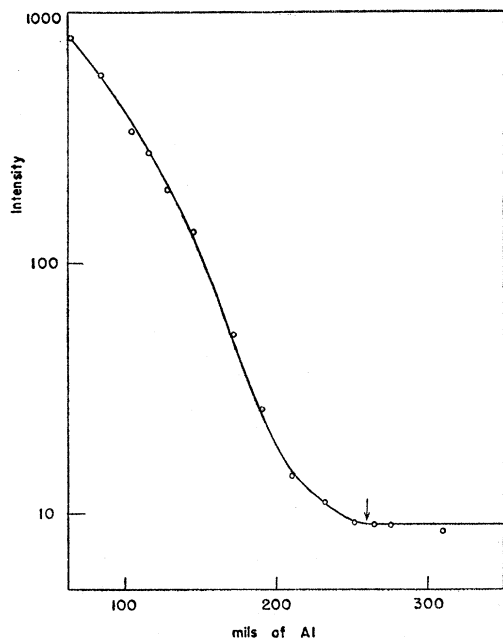


FIG. 9.  $\beta$  absorption curve for  $\text{K}^{42}$ . The end point for the high energy component is 3.57 Mev according to Feather's rule.

<sup>2</sup> D. C. Kalbfell and R. A. Cooley, Phys. Rev. 58, 91 (1940).

<sup>3</sup> A. F. Clark, Phys. Rev. 61, 242 (1942).

<sup>4</sup> K. Siegbahn, Proc. Roy. Soc. 187, 527 (1947).

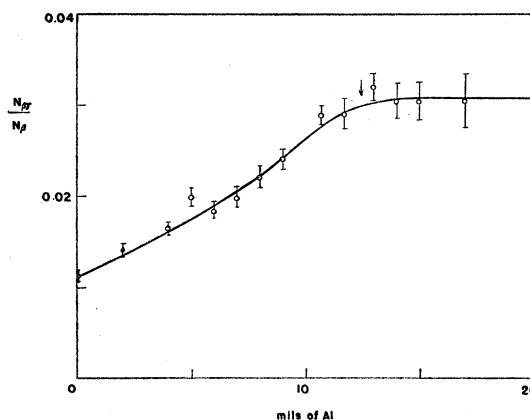


FIG. 10.  $N_{\beta\gamma}/N_{\beta}$  as a function of absorber thickness for  $\text{Hf}^{181}$ . The rise may be due to the presence of the metastable state of half-life 22 microseconds.

few points of the osmium curve fall slightly higher than the following points. This seems to be a real effect and may possibly be attributable to internal conversion.

Absorption measurements give an upper limit of 0.68 Mev for the spectrum of iridium.

### Complex Spectra

A beta spectrograph study of  $\text{Sb}^{124}$  (60 days) has been carried out by Hales and Jordan.<sup>5</sup> This work indicated that the  $\beta$  spectrum is complex with end points at 0.78 and 2.48 Mev. Previously Mitchell, Langer, and McDaniel<sup>6</sup> had found that the  $\beta$ - $\gamma$  coincidence rate was independent of the  $\beta$  energy and the end point which they obtained by absorption was given as 1.53 Mev.

Our coincidence curve is shown in Fig. 5. The  $\beta$ - $\gamma$  rate per  $\beta$  particle clearly decreases by a factor of 2.5 in the initial portion. From this curve a value of 0.67 Mev is obtained for the low energy component of the  $\beta$  spectrum. An absorption curve of the  $\beta$  radiation gives an upper limit of the high energy  $\beta$  radiation at 2.45 Mev. These values are in good agreement with the data of Hales and Jordan.

Coincidence measurements have also been made with europium<sup>154</sup> (5-8 years). The curve, shown in Fig. 6, indicates the complexity of the  $\beta$  spectrum. The low energy spectrum has an

<sup>5</sup> E. B. Hales and E. B. Jordan, Phys. Rev. 64, 202 (1943).

<sup>6</sup> A. C. G. Mitchell, L. M. Langer, and P. W. McDaniel, Phys. Rev. 57, 1107 (1940).

TABLE I. Beta ray energies.

Isotope	$\beta$ spectrum	End point		Energies	
		mg/cm <sup>2</sup> of Al			
Sb <sup>124</sup>	Complex	204	1160	0.67	2.45
Cs <sup>134</sup>	Simple		269		0.80
Eu <sup>154</sup>	Complex	177	388	0.62	1.0
Au <sup>198</sup>	Simple		332		0.91
Hf <sup>181</sup>	?	98	184	0.34	0.64
Ir <sup>192, 194</sup>	Simple		208		0.68
Hg <sup>203, 205</sup>	Complex	15.3	77.5	0.11	0.44
Os <sup>193</sup>	Simple		184		0.64
K <sup>42</sup>	Complex	~660	1770	~1.52	3.57

upper limit of 0.62 Mev. Absorption of the total  $\beta$  radiation gives a value of 1.0 Mev for the higher energy  $\beta$  radiation.

The corresponding curve is shown for Hg<sup>203, 205</sup> (51.5 days) in Fig. 7. The low energy  $\beta$  radiation corresponds to an energy of 0.11 Mev, while the total absorption curve gives a value of 0.44 Mev for the high energy component.

Potassium<sup>42</sup> (12.4 hours) prepared by deuteron bombardment also shows a complex spectrum (Fig. 8). It was very difficult to obtain the end point of the low energy spectrum, but an estimate of 1.5 Mev can be given. The high energy radiation was found to be completely absorbed by 1.77 g/cm<sup>2</sup> of aluminum, i.e.,  $E_{\max} = 3.6$  Mev (Fig. 9). This seems to be in agreement with the cloud-chamber work of Kurie, Richardson, and

Paxton.<sup>7</sup> It appears that the higher energy  $\beta$  transition is not followed by  $\gamma$  radiation.

### Hafnium

The decay of hafnium<sup>181</sup> (55 days) has been shown by De Benedetti and McGowan<sup>8</sup> to involve a metastable state of lifetime 22 microseconds. This is long compared to the resolving time ( $\sim 0.5$  microsecond) of the coincidence circuit used. It is interesting to note that the curve, Fig. 10, rises as the  $\beta$  absorber thickness is increased. A constant value is obtained when the thickness is  $\sim 12$  to 13 mils of aluminum. It is not known whether or not the  $\beta$  spectrum itself is simple. From the data it would seem that the spectrum may really be simple, the rise being brought about by the delayed conversion electrons of lower energy.

### 4. SUMMARY OF RESULTS

In Table I, we have tabulated the  $\beta$  energies which we have found for these nine nuclei. All  $\beta$  energies except the low energy component of Hf and Hg are calculated from Feather's rule, i.e.,  $M(\text{g/cm}^2) = 0.54E_{\max} - 0.16$ .

<sup>7</sup> F. N. D. Kurie, J. R. Richardson, and H. C. Paxton, Phys. Rev. **49**, 368 (1936).

<sup>8</sup> S. De Benedetti and F. K. McGowan, Phys. Rev. **70**, 569 (1946).