

Radiations from Ho<sup>164</sup>†\*

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(Received May 3, 1954)

The decay modes of 36.7-min Ho<sup>164</sup> have been studied with a magnetic spectrometer, scintillation spectrometer, and proportional counter. Decay to the two even-even daughters, Dy<sup>164</sup> by orbital electron capture, and Er<sup>164</sup> by beta decay, is indicated. The beta spectrum appears to be complex, with maximum energy  $0.99 \pm 0.03$  Mev. Gamma rays of energy 90.5, 72.8, and 37.3 kev, together with x-rays at 45.7 kev, are found. There is also evidence for a gamma ray at approximately 46 kev. Photon-photon coincidences are found for 46-73 and 37-46 kev approximate energies. Photon-beta coincidences are found to involve photons of approximate energy 91 and 50 kev. The data are interpreted in terms of first excited levels at 72.8 kev in Dy<sup>164</sup>, 90.5 kev in Er<sup>164</sup>, with probably a second level in Dy<sup>164</sup> at 110 kev. Delayed photon-beta and photon-photon coincidences were found with a half-life of  $1.4 \times 10^{-9}$  sec. The former are associated with the 90.5-kev level of Er<sup>164</sup>; the assignment of the photon-photon coincidences is uncertain. A possible isomeric transition in Ho<sup>164</sup> is also suggested. Spin and parity assignments, on the basis of conversion coefficients and  $\log(f_0 t)$  value, are made for certain of the levels.

## I. INTRODUCTION

HALF-LIVES for the beta decay of Ho<sup>164</sup> of 47, 38.6, 41.5, and 35 and 34.0 minutes have been reported, respectively, by Pool and Quill,<sup>1</sup> Wäffler and Hürzel,<sup>2</sup> Wäffler,<sup>3</sup> and Wilkinson and Hicks.<sup>4,5</sup> Wilkinson and Hicks<sup>5</sup> reported a negative beta particle of 0.95-Mev maximum energy. The Al absorption curve indicated a simple spectrum. It was stated by the latter workers that if photons were present there would be fewer than 0.05 photon per beta particle. Moreover, no conversion electrons were found in their work.

The present investigation was undertaken because of several factors. At the present time, rare earth samples of a fairly high degree of purity are available in both the oxide form and the metal. Also, the activity can be produced readily in a rather pure state by means of a gamma-neutron reaction on the 100-percent isotope Ho<sup>165</sup>. These considerations gave promise that the study of the Ho<sup>164</sup> radiations might be a fruitful one to carry out by means of the probe technique<sup>6</sup> employed in this laboratory with the 22-Mev betatron. Further, it was expected that the decay might be more complicated than had been indicated by the work of previous observers, and that two even-even daughter nuclei might be involved. Previous work in this laboratory on the Ta<sup>180</sup> decay scheme<sup>7</sup> and the discussion of

even-even nuclei in general by Scharff-Goldhaber<sup>8</sup> suggested that gamma rays in the energy range 50 to 100 kev might be found, together with the possible occurrence of K-capture transitions to Dy<sup>164</sup>, as well as beta minus transitions to states in Er<sup>164</sup>.

## II. APPARATUS

Electron momentum spectra were measured in the double-focusing 255° magnetic spectrometer constructed by Bendel.<sup>9</sup> Gamma radiation energies were measured with scintillation counters employing RCA 5819 and Du Mont 6292 photomultipliers and NaI(Tl) crystals and with an argon-filled proportional counter of the type described by Cockcroft and Curran.<sup>10</sup> The pulse height distributions were analyzed by means of a 10-channel differential discriminator built by Shore<sup>11</sup> and a single-channel analyzer similar to that described by Frankel.<sup>12</sup> A conventional coincidence circuit with a resolving time of 0.25 μsec and having a Rossi type crystal diode mixer was utilized. A fast coincidence circuit was also built for delayed coincidence measurements. The resolving time was  $2\tau \approx 5 \times 10^{-9}$  sec. Again, a Rossi type mixer with 1N56 diodes was employed. The detectors were trans-stilbene crystals employed in combination with RCA 6199 phototubes. Hewlett-Packard distributed amplifiers and RG-62/U delay lines were used.

## III. DATA

The half-life of Ho<sup>164</sup> was determined to be  $36.7 \pm 0.5$  min from measurements of the particles by means of an end-window Geiger counter. This agrees well with

† This work was supported in part by the U. S. Atomic Energy Commission and the U. S. Office of Naval Research. The former is to be thanked for providing Sn<sup>113</sup>, Cs<sup>137</sup>, and Hg<sup>203</sup>, the radiations from which were used to calibrate various pieces of equipment. In particular, thanks are due Professor F. H. Spedding and the Ames Laboratory of the U. S. Atomic Energy Commission for kindly providing pure samples of metallic holmium and holmium oxide, without which this work could not have been done.

\* Part of the doctoral thesis of Hugh Needham Brown.

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<sup>1</sup> M. L. Pool and L. L. Quill, *Phys. Rev.* **53**, 437 (1938).

<sup>2</sup> H. Wäffler and O. Hürzel, *Helv. Phys. Acta* **21**, 200 (1948).

<sup>3</sup> H. Wäffler, *Helv. Phys. Acta* **23**, 239 (1950).

<sup>4</sup> G. Wilkinson and H. G. Hicks, *Phys. Rev.* **74**, 1733 (1948).

<sup>5</sup> G. Wilkinson and H. G. Hicks, *Phys. Rev.* **79**, 815 (1950).

<sup>6</sup> R. A. Becker, *Rev. Sci. Instr.* **22**, 773 (1951).

<sup>7</sup> Brown, Bendel, Shore, and Becker, *Phys. Rev.* **84**, 292 (1951).

<sup>8</sup> G. Scharff-Goldhaber, *Phys. Rev.* **90**, 587 (1953).

<sup>9</sup> W. L. Bendel, Ph.D. thesis, University of Illinois, 1953 (unpublished).

<sup>10</sup> A. L. Cockcroft and S. C. Curran, *Rev. Sci. Instr.* **22**, 37 (1951).

<sup>11</sup> F. J. Shore, Jr., Ph.D. thesis, University of Illinois, 1952 (unpublished); Shore, Bendel, Brown, and Becker, *Phys. Rev.* **91**, 1203 (1953).

<sup>12</sup> S. Frankel, Ph.D. thesis, University of Illinois, 1949 (unpublished).

certain of the previous measurements mentioned above. Weak long-lived components were noted, but their origins were not ascertained.

### 1. Particle Spectra

Sources for the magnetic spectrometer were prepared by depositing a few milligrams of irradiated Ho<sub>2</sub>O<sub>3</sub> on a 0.6-mg/cm<sup>2</sup> rubber acetate film. These sources were necessarily rather thick, having an average surface density of about 15 mg/cm<sup>2</sup>.

The momentum spectrum of the electrons emitted from Ho<sup>164</sup> is shown in Fig. 1. The inset is a Kurie plot of the continuous spectrum, constructed with the aid of tables.<sup>13</sup> This analysis indicates that the maximum disintegration energy for the negatron decay is  $990 \pm 30$  kev. The nonlinearity of the Kurie plot near the end point is consistent with a complex beta spectrum consisting of two groups separated by about 90 kev. Subsequent experiments, to be mentioned later, will be seen to urge this interpretation. The departure from a straight line at 550 kev and below is assumed to arise from source thickness.

No positrons could be detected in the magnetic spectrometer, even with an intense source consisting of an irradiated chunk of Ho metal. Provided the kinetic energy available for positron emission is greater than 200 kev, there are less than  $5 \times 10^{-4}$  positron per negatron emitted by Ho<sup>164</sup>.

The nine conversion electron lines in Fig. 1 were detected using a 130- $\mu$ g/cm<sup>2</sup> zapon window on the Geiger counter. Table I lists the electron energies, interpretations, and intensities, relative to the betas, of these conversion lines. A small correction for counter window absorption (<20 percent) was applied to the data, but, because of the thick sources utilized, the intensities given are inaccurate. They were employed only as crude estimates or as lower limits.

The photon energies listed in the table are mean values derived from several determinations. The *K*, *L*, and *M* binding energies for the appropriate elements

TABLE I. Energies, interpretations, and relative intensities of conversion electrons.

Line	<i>T</i> (kev)	Interpretation	Intensity ( <i>e</i> / $\beta$ )
A	19.5	72.8- <i>K</i>	0.04
B	27.8	37.3- <i>L</i>	0.067
C	33.6	37.3- <i>M</i> , 90.5- <i>K</i>	0.017
D	37.1	46- <i>L</i>	0.20
E	44.6	46- <i>M</i>	0.10
F	64.5	72.8- <i>L</i>	0.24
G	71.2	72.8- <i>M</i>	0.036
H	82.1	90.5- <i>L</i>	0.19
J	88.2	90.5- <i>M</i>	0.024

<sup>13</sup> National Bureau of Standards, *Tables for the Analysis of Beta Spectra* (U. S. Government Printing Office, Washington, D. C., 1952).

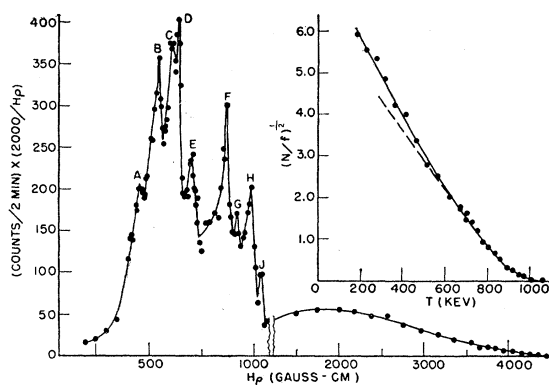


Fig. 1. Electron momentum spectrum and Kurie plot of the continuum.

(to be assigned below) were taken from the table of Hill, Church, and Mihelich.<sup>14</sup>

### 2. Photon Spectra

The most prominent electromagnetic radiation from Ho<sup>164</sup> has an energy of about 46 kev. Within experimental error, the energy is the same as the mean of the *K* $\alpha$  x rays from Dy, 45.7 kev. The evidence for this is contained in Fig. 2 which shows photon spectra observed with the argon-filled proportional counter. The 46-kev radiation from Ho<sup>164</sup> is compared with the *K* $\alpha$  x rays from Hf and Gd. It is estimated that Ho or Er x rays, if present, contribute less than 30 percent or 20 percent, respectively, to the observed intensity.

This figure also shows the presence of a 37.3-kev gamma ray. After correction for counter detection efficiency, the intensity of this gamma ray,  $\gamma_3$ , relative to the x ray is  $\gamma_3/x = 0.040 \pm 0.003$ .

Figure 3 illustrates the photon spectrum obtained with a NaI(Tl) crystal ( $1\frac{1}{2}$  in. diam  $\times$  1 in.) and a Du Mont 6292 photomultiplier.  $\gamma_3$  is not resolved, but the 72.8- and 90.5-kev gammas,  $\gamma_2$  and  $\gamma_1$  respectively, show up clearly. (Energies indicated on this and other figures were derived from a calibration curve for the crystal, but may differ slightly from the "best" values given by the conversion electron data.) Relative to the x ray, the intensities of these two gammas are  $\gamma_1/x = 0.039 \pm 0.007$  and  $\gamma_2/x = 0.037 \pm 0.007$ . Corrections were made in all cases for the "escape peak" intensities employing calculated values in those instances in which the escape peak was obscured by another line.

The ratio  $x/\beta$  was determined by comparison of the x-ray intensity detected by a NaI crystal and the  $\beta$ -ray intensity detected by an N. Wood end-window Geiger counter. The result, after correction for the variation in sensitivity of the Geiger counter across the window, was  $x/\beta = 0.90 \pm 0.20$ , the uncertainty quoted arising mainly from the corrections to the beta intensity for absorption in the air and counter window.

<sup>14</sup> Hill, Church, and Mihelich, *Rev. Sci. Instr.* **23**, 523 (1952).

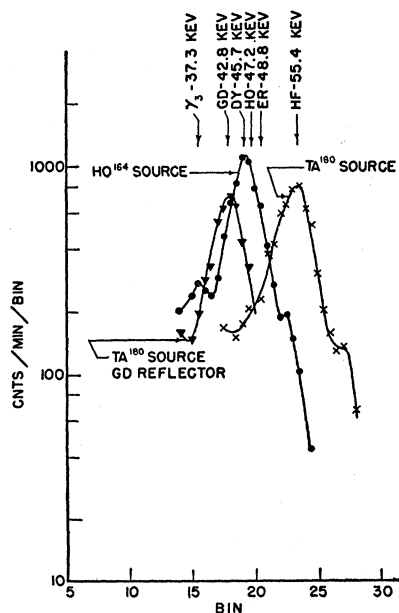


FIG. 2. Comparison of  $\text{Ho}^{164}$  photons with Hf and Gd  $K\alpha$  x-rays in an argon filled proportional counter.

Possible discrepancies owing to the use of two separate counters have not been taken into account in the uncertainty quoted.

The photon spectrum at higher energies was also examined with the NaI crystal-photomultiplier combination. No radiation in the vicinity of 500 keV, other than bremsstrahlung, was found with a very active Ho metal source. The resulting upper limit on positrons from  $\text{Ho}^{164}$  is  $8 \times 10^{-4}$  positron per negative beta.

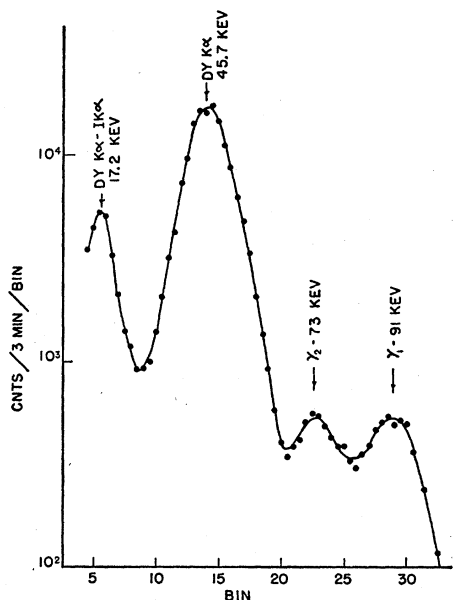


FIG. 3. Pulse height spectrum of x-rays and  $\gamma$  rays from  $\text{Ho}^{164}$  with a NaI(Tl) crystal.

Table II summarizes the data on photon energies and intensities.

### 3. Coincidence Experiments

Gamma-gamma coincidences in the vicinity of 500 keV were found to have a sharp maximum at  $180^\circ$ , indicating the presence of positrons. The decay of these coincidences did not, however, exhibit the proper lifetime. A half-life of about 93 minutes was measured. It is assumed that some impurity was responsible.

Coincidences involving the low-energy gammas were obtained. Beta-photon and photon-photon coincidences were observed. The pulse height spectra of the gammas involved are reproduced in Fig. 4. The gamma spectrum concerned with the beta-gamma coincidences was measured with a NaI crystal detector shielded from the betas by  $940 \text{ mg/cm}^2$  of Be. The beta counter was an anthracene scintillation detector which was made to be insensitive below 150 keV in order to exclude low-energy photons and conversion electrons. The beta single counting rate was  $8.81 \times 10^5$  per minute and the  $\gamma$  counter subtended 1.66 steradians at the source. A NaI detector, sensitive down to 15 keV, replaced the

TABLE II. Photon energies and relative intensities.

Photon	$E(\text{keV})$	Int. ( $\gamma/\beta$ )
$\gamma_1$	$37.3 \pm 0.5$	$0.036 \pm 0.01$
x	$45.7 \pm 0.5$	$0.90 \pm 0.20$
$\gamma_2$	$72.8 \pm 0.5$	$0.033 \pm 0.007$
$\gamma_3$	$90.5 \pm 0.5$	$0.035 \pm 0.007$

anthracene counter in the photon-photon coincidence experiment. The calculated accidental coincidences for the  $\gamma$ - $\gamma$  curve indicate the shape of the single-count gamma-ray spectrum for comparison purposes. The accidentals in the  $\beta$ - $\gamma$  case were slightly less in number.

Evidently, there are photons of about 50- and 91-keV energy in coincidence with the betas, and 46- and 70-keV photons coinciding with one another. It is to be noted that the low-energy end of the gamma-gamma spectrum does not fall away as sharply as either the beta-gamma or randoms spectra. One suspects a possible influence here of the 37.3-keV gamma ray. The gamma-gamma coincidences were therefore examined in more detail with pulse height selection in both channels. That is, spectra were recorded, with the 10-channel analyzer attached to counter No. 1, which were in coincidence with pulses from counter No. 2 in a narrow range selected by the single-channel analyzer. NaI crystals ( $1\frac{1}{2}$  in. diam  $\times$  1 in.) and Du Mont 6292 tubes were utilized.

Figure 5 contains three spectra taken with counter No. 2 set to select three different energy bands. The range covered by counter No. 2 for each curve is indicated by the correspondingly numbered shaded region. Accidental coincidences were negligible. These

curves show clearly that 46–73 and 37–46-keV coincidences occur. No noticeable evidence is given by this figure for 37–73-keV coincidences. The total number of coincidences observed are plotted in Figs. 4 and 5 and, hence, the statistical probable errors are very nearly equal to the square roots of the ordinates.

Beta—conversion-electron coincidences were looked for with the 255°, double-focusing magnetic spectrometer by placing an anthracene crystal near the source to detect beta particles and an anthracene crystal at the exit slit to detect conversion electrons. Definite evidence was obtained for such coincidences involving the conversion electrons associated with the 90.5-keV gamma ray. However, no evidence for coincidences of this kind was found for conversion electrons associated with photons of energy near 46 keV.

Another coincidence experiment measured the total energy of events coinciding within the resolving time of the 10 channel analyzer, about 3  $\mu\text{sec}$ . A  $\text{Ho}_2\text{O}_3$  source was placed between two NaI crystals, on a single RCA 5819, which absorbed completely all x rays and particles from the source. Since the 90.5-keV gamma and its conversion electrons are known from the preceding discussion to be in coincidence with a beta, pulses arising from these transitions were conveniently removed from the low-energy line spectrum and spread out over the beta continuum. Hence, the low-energy lines remaining were caused only by other decay branches or by decay of isomeric states produced by the beta decay. The ratio of such low-energy counts to the total beta continuum was found to be  $1.3 \pm 0.15$ .

#### 4. Delayed Coincidences

Beta-gamma and gamma-gamma coincidences were examined, with the fast-coincidence equipment already described, for possible measurable lifetimes. The resolving time was not low enough ( $\sim 5 \times 10^{-9}$  sec) for a precise determination, but an analysis of the delayed coincidence curves employing the method of moments<sup>15</sup> yielded a half-life for both  $\beta$ - $\gamma$  and  $\gamma$ - $\gamma$  coincidences of  $(1.4 \pm 0.5) \times 10^{-9}$  sec (assuming a single period in each case). The  $\beta$ - $\gamma$  result is deemed to be the more reliable since it is derived from a clearly discernable shift in the centroid of the delay curve. The  $\gamma$ - $\gamma$  lifetime was derived from a broadening of the delay curve using the second moment.

#### IV. DISCUSSION

The obvious interpretations of the conversion electron lines were given in Table I. Lines *D* and *E* at 37.1 and 44.6 keV demand further attention. Their positions are, within experimental error, just what one would expect for Auger electrons owing to the conversion of the Dy x rays. However, the fluorescent yield of the *K* shell,  $\omega_K$ , is about 0.92 for the elements of importance here.<sup>16</sup> Hence, the Auger intensity should

<sup>15</sup> Z. Bay, Phys. Rev. **77**, 491 (1950).

<sup>16</sup> Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).

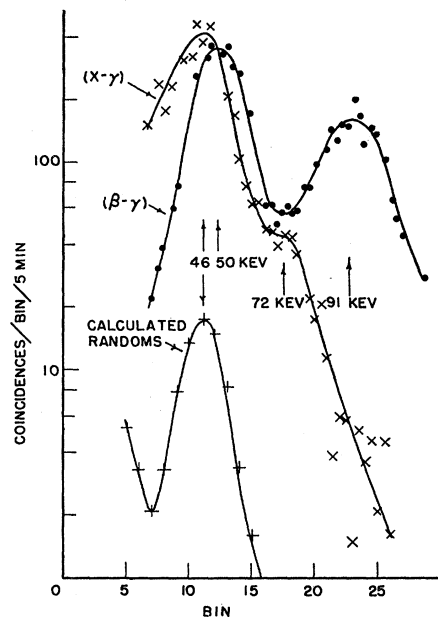


FIG. 4. Pulse height spectra of photons concerned with  $\beta$ - $\gamma$  and  $\gamma$ - $\gamma$  coincidences.

be less than 10 percent of the x-ray intensity or less than 9 percent of the beta intensity. The observed strength of these lines, from Table I, is 30 percent of that of the betas. Moreover, this value is a minimum because of the thick source employed. Hence, it is concluded that a part of the electrons at 37.1 and 44.6 keV arises from the conversion of a nuclear gamma ray of about 46-keV energy.

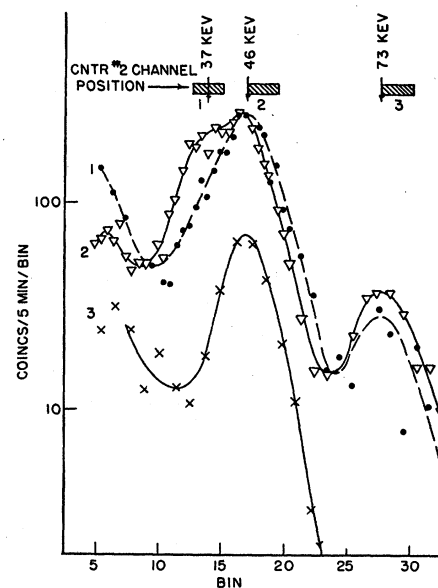


FIG. 5. Pulse height spectra of photons reaching counter No. 1 in coincidence with photons of selected energies detected by counter No. 2.

### 1. Negative Beta Branches

The possible complexity of the beta spectrum suggested by the Kurie plot and the beta—90.5-keV gamma coincidences support the assumption of a 90.5-keV level in  $\text{Er}^{164}$  which is reached by the beta decay. The coincidences between betas and photons near 50 keV is interpreted to be caused by the  $K$ -shell conversion of  $\gamma_1$ , which leads to an x ray at 48.8 keV. The spectrum of Fig. 4 can then be employed to find the  $K$  conversion coefficient of  $\gamma_1$  from the ratio of x rays to  $\gamma_1$  rays. This ratio is  $(x/\gamma_1)_{\text{obs}} = \omega_K \alpha_{1K} = 1.8$  where  $\alpha_{1K}$  is the conversion coefficient sought. Hence,  $\alpha_{1K} = 1.9 \pm 0.2$ . An extrapolation for the calculation of  $K$ -conversion coefficients at low energies is described by Axel and Goodrich.<sup>17</sup> The results of this extrapolation compare with the measured value as shown by Table III which applies for  $Z=68$  and  $E_\gamma=90$  keV. If  $\gamma_1$  goes to the ground state of  $\text{Er}^{164}$ , assumed to be  $0+$ , it must be a pure multipole and, hence,  $\alpha_{1K}=1.9$  establishes that  $\gamma_1$  is  $E2$  and the 90.5-keV level is  $2+$ .

The empirical  $K/L$  ratios of Goldhaber and Sunyar<sup>18</sup> and the observed  $L/M$  ratio, combined with  $\alpha_{1K}$ , yield a total conversion coefficient of  $\alpha_1 = 11 \pm 3$  for  $\gamma_1$ . The relative number of betas leading to the excited level in  $\text{Er}^{164}$ ,  $\beta_1/\beta$ , then may be calculated from the observed  $\gamma_1/\beta$  ratio and, independently, from the  $\beta$ - $\gamma$  coincidence experiment. The result is  $\beta_1/\beta = 0.38 \pm 0.08$ . Since this ratio is significantly less than one, the assumption of a ground state transition is justified. Hence, the  $\text{Ho}^{164} - \text{Er}^{164}$  atomic mass difference is taken to be  $0.99 \pm 0.03$  MeV.

The delayed beta-gamma coincidences yield  $(1.4 \pm 0.5) \times 10^{-9}$  sec as the half-life of the 90.5-keV excited state. The  $\gamma$ -ray transition probability, corrected for conversion, is then  $T_{\text{exptl}} = 4.0 \times 10^7 \text{ sec}^{-1}$ . Now the transition rate for a single nucleon pair changing from a state  $(j^2)_{J=2}$  to a state  $(j^2)_{J=0}$  is  $T_{\text{calc}} = 9.65 \times 10^5 \text{ sec}^{-1}$ , as calculated from an expression given by Bohr and Mottelson.<sup>19</sup> Hence,  $F = T_{\text{exptl}}/T_{\text{calc}} = 41 \pm 13$ . This is appreciably smaller than the  $F$  values ( $\sim 150$ ) given by Bohr and Mottelson for similar nuclei. The difference is evidently caused by the use of a larger conversion coefficient here, since the observed lifetimes are approximately the same. Conversion data published by

TABLE III. Calculated  $K$ -conversion coefficients for  $Z=68$  and  $E=90$  keV for various multiplicities compared with the experimental value.

	$E1$	$E2$	$E3$	$M1$	$E4$	$M2$	$E5$
$\alpha_K$ (calc.)	0.37	1.6	5.4	6.4	18	54	54
$\alpha_{1K}$ (exptl.)		$1.9 \pm 0.2$					

<sup>17</sup> P. Axel and R. F. Goodrich, University of Illinois, U. S. Office of Naval Research Report (unpublished).

<sup>18</sup> M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 956 (1910).

<sup>19</sup> A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

McGowan<sup>20</sup> and by Mihelich and Church<sup>21</sup> for  $\text{Dy}^{160}$ ,  $\text{Er}^{166}$ , and  $\text{Yb}^{170}$  also lead to  $F$  values on the order of 40 to 50.

After subtracting the x-ray contribution associated with the conversion of  $\gamma_1$  from the total x-ray intensity, the remainder may be assumed to represent decay by other branches. Making use of the  $\beta_1/\beta$  ratio, the  $f_{0\pm}$  values for the two transitions may be found by employing the tables of Feenberg and Trigg.<sup>22</sup> The results are  $\log(f_{0\pm}) = 5.76$  and  $5.56$ , respectively, for  $\beta_1$  and  $\beta_2$ . These values are compatible with either allowed or first forbidden transitions. Assignment of  $0-$  or  $2-$  to the 36.7-min level in  $\text{Ho}^{164}$  would be permitted by selection rules, but in that event the  $\Delta I=2$  transition to the  $2+$  or  $0+$  state in  $\text{Er}^{164}$  would be expected to have  $\log(f_{0\pm}) \approx 8$ . A  $0+$  or  $2+$  state in  $\text{Ho}^{164}$ , as well as spins higher than 2, require more highly forbidden transitions, with an estimated  $\log(f_{0\pm}) \lesssim 10$ . (See Blatt and Weisskopf<sup>23</sup> for a discussion of these points.) Hence,  $1\pm$  seems to be the most likely assignment for the parent level. Coupling rules proposed by Nordheim<sup>24</sup> for odd-odd nuclei give odd parity for  $\text{Ho}^{164}$  and favor either a zero-spin or a high-spin state. A  $1-$  level is excluded by these rules.

The conclusions reached above are contained in the proposed decay scheme of Fig. 6.

### 2. $K$ Capture and Positron Branches

The 72.8-keV gamma ray  $\gamma_2$  can be ascribed to  $\text{Dy}^{164}$  with a high degree of certainty. The energy sums for the  $K$ ,  $L$ , and  $M$  conversion lines give the best agreement if Dy binding energies are used. In addition, the  $\gamma$ - $\gamma$  coincidences prove that  $\gamma_2$  coincides with a 46-keV photon, presumably Dy  $K$  x-rays following  $K$  capture to a 72.8-keV excited state in  $\text{Dy}^{164}$ . Figure 5 also shows that  $\gamma_3$ , 37.3 keV, is in coincidence with the x rays. Simple calculations verify that the 37.3-keV pulses do not arise from x-x coincidences degraded by Compton scattering in the beta absorbers, or by the escape of iodine x-rays from one crystal to the other. A consideration of the coincidence data makes the placement of  $\gamma_2$  and  $\gamma_3$  as shown in the decay scheme of Fig. 6 seem the most probable. The fact that no  $\gamma_2$ - $\gamma_3$  coincidences were observed does not invalidate this scheme since the conversion of  $\gamma_2$  reduces the number of such events by a factor of  $(1+\alpha)$ , which is on the order of 20.

The  $K$ -conversion coefficient of  $\gamma_2$  may be estimated from Fig. 5, corrections being made for contributions from  $\gamma_3$  by using the known efficiencies of the single-

<sup>20</sup> F. K. McGowan, Phys. Rev. **85**, 151 (1952).

<sup>21</sup> J. W. Mihelich and E. L. Church, Phys. Rev. **85**, 690 (1952); **87**, 1144 (1952).

<sup>22</sup> E. Feenberg and G. L. Trigg, Revs. Modern Phys. **22**, 399 (1950).

<sup>23</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

<sup>24</sup> L. W. Nordheim, Revs. Modern Phys. **23**, 322 (1951).

channel counter for various energies. The many possible combinations make the exact expression for the coincidence spectrum rather lengthy and it is not given here. The type of result obtained is illustrated more clearly by the simple case of *K*-capture decay to a single excited level in the daughter which then decays with the emission of a gamma ray. If it is assumed that the x-rays and  $\gamma$  rays are totally absorbed in the NaI crystals, that counter No. 2 is equally sensitive to all photon energies, and that the spectrum of coincident photons is observed in counter No. 1, then the ratio of x-rays to  $\gamma$  rays is  $(x/\gamma)_{\text{coinc}} = 1 + 2\alpha_{K\omega_K}$ . The first term originates from coincidences between *K*-capture x-rays and the  $\gamma$  rays, while the second is caused by x-x coincidences arising from *K* capture followed by *K*-shell conversion of the  $\gamma$ . A factor of 2 appears since the x-rays may enter either counter.

For the case at hand, the final result, after making corrections using the complete expression for the coincidence spectrum, is  $\alpha_{2K} = 2.7 \pm 0.5$ . This agrees well with the extrapolated theoretical value for an *E2* transition which is 2.6. Hence, again assuming 0+ for the Dy<sup>164</sup> ground state, the 72.8-keV level is 2+. Electron-gamma coincidences furnished qualitative support for the assumed cascade of  $\gamma_2$  and  $\gamma_3$  but, owing to poor electron energy resolution and calibration in the thin anthracene crystal, no reliable quantitative information was obtained from these data.

A very rough estimate of the *L* conversion coefficient of  $\gamma_3$  may be found from the electron momentum spectrum, Fig. 1, and the measured ratio  $\gamma_3/\beta$ . The result is that  $\alpha_{3L}$  is at least 2 and, guessing at the effect of source self-absorption, is probably not greater than 10 or 15. Gellman, Griffith, and Stanley<sup>25</sup> have calculated *L* conversion coefficients. Interpolated to *Z*=66 and *E<sub>γ</sub>*=37 keV, it is found from their calculations that  $\alpha_L = 200$  for an *E2* transition. Hence, a pure *E2* transition would seem to be excluded for  $\gamma_3$ , although pure *E1* or *M1* could occur since the calculated *L* coefficients are about 0.8 and 9.0, respectively. Of course, *M1*+*E2* is also possible.

With  $\gamma_3$  placed as in Fig. 6, the second excited state of Dy<sup>164</sup> therefore has a spin of 1, 2, or 3 and lies at 110 keV. This would not be consistent with the pure rotational spectrum predicted by the strong coupling approximation of the collective model<sup>19</sup> which would give a spin 4+ and an energy of 240 keV (assuming 73 keV for the first excited state). It is thought that the 110-keV level is probably 2+ since Scharff-Goldhaber<sup>8</sup> finds that about 50 percent of the second excited levels in even-even nuclei are 2+. This would also allow the transition to this level from the Ho<sup>164</sup> level, which is thought to be 1±, to compete favorably with those to the ground and 72.8-keV states whereas a spin of 3 probably would not. The absence of a crossover gamma

<sup>25</sup> Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).

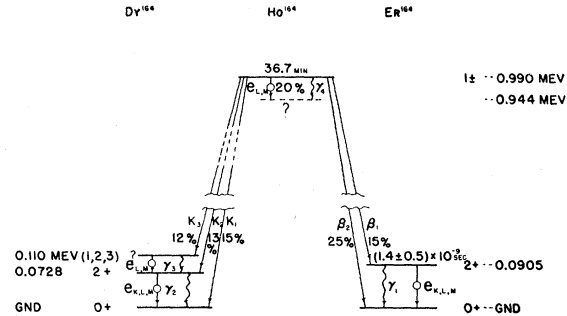


FIG. 6. A proposed decay scheme for Ho<sup>164</sup>. The Ho<sup>164</sup>-Dy<sup>164</sup> mass difference is not specified in the figure.

to the ground state is not surprising in view of the work of Kraushaar and Goldhaber.<sup>26</sup>

It would be satisfying to attribute the 1.4 × 10<sup>-9</sup>-sec half-life of the delayed  $\gamma$ - $\gamma$  coincidences to the 72.8-keV gamma, but because of  $\gamma_3$  this aspect of the problem remains undecided.

The *K* capture to positron ratio,  $f_K/f_+ > 2 \times 10^3$ , indicates that the Ho<sup>164</sup>-Dy<sup>164</sup> atomic mass separation is less than 1.4 MeV,<sup>22</sup> if one assumes an allowed transition. The result should not be greatly different for a first forbidden decay. Mass spectrographic measurements by Hogg and Duckworth,<sup>27</sup> combined with 0.99 MeV as the Ho<sup>164</sup>-Er<sup>164</sup> separation, give 2.1 ± 0.9 MeV for the Ho<sup>164</sup>-Dy<sup>164</sup> difference. (The error quoted was not given explicitly for these nuclei but is the minimum uncertainty for their measurements in general.)

### 3. Long-Lived Isomer

The total-absorption experiment described in III suggests the possible existence of a long-lived isomeric transition. Entirely independent of the detailed decay scheme, if all multiple-step decay branches had mean lives less than 3 × 10<sup>-6</sup> sec (the analyzer's resolving time), then in this experiment all *K*-capture processes would produce discrete, low-energy lines whereas all beta emission processes would produce a continuum. Now, as determined by separate measurements,  $x/\beta = 0.9$ . When one corrects for conversion x rays and fluorescent yield, the maximum possible relative *K*-capture intensity is  $(K/\beta)_{\text{max}} = 0.9 \pm 0.2$ . However, the ratio of the total intensity of the low-energy line structure to the beta continuum in the total absorption experiment was 1.3 ± 0.15. Hence, the excess was attributed to the existence of an isomeric level with  $\tau > 3 \times 10^{-6}$  sec. No further specific information was obtained on this point. The assignment of the remaining 46-keV gamma ray  $\gamma_4$  to an isomeric transition in Ho<sup>164</sup> is, at most, only a reasonable suggestion and therefore is indicated by broken lines in the figure.

<sup>26</sup> J. J. Kraushaar and M. Goldhaber, Phys. Rev. **91**, 1081 (1953).

<sup>27</sup> B. C. Hogg and H. E. Duckworth, Phys. Rev. **91**, 1289 (1953).

Isomeric levels with long lifetimes are rare in even-even nuclei; only about three are known and these are highly excited states.<sup>28</sup> All proposed nuclear models predict, in agreement with this fact, that the low-lying levels in such nuclides differ by only one or two units of spin. On the other hand, numerous odd-odd isomers are known.<sup>28</sup> Nordheim's<sup>24</sup> coupling rules, as mentioned before, favor a spin of 0 or a high spin for the ground state of Ho<sup>164</sup>. If the high spin is obtained, then an isomeric level with spin about 1 would be required to permit the 36.7-min decay. Low-lying isomeric states in odd-odd nuclei are, in fact, predicted by the strong-coupling collective model of Bohr and Mottelson.<sup>19</sup> The parity of the isomeric state would be the same as that of the ground state in this case.

Of interest with regard to these considerations is the work of Butement<sup>29</sup> on Ho<sup>166</sup>. This observer reports an activity with half-life greater than 30 years, in addition to the well known 27-hour decay. This indicates the probability of an isomeric pair similar to that hypothesized here for Ho<sup>164</sup>. A long-lived activity decaying to the 90- and the 73-keV  $\gamma$ -ray emitting states was

<sup>28</sup> M. Goldhaber and R. D. Hill, *Revs. Modern Phys.* **24**, 179 (1952).

<sup>29</sup> F. D. S. Butement, *Proc. Phys. Soc. (London)* **A65**, 254 (1952).

searched for in the present work with a source which had been irradiated for nine hours in the betatron. No evidence was found to support such a long-lived decay. However, it is felt that the inefficient method of activation here employed does not preclude its existence. Butement, in the course of his work, irradiated Ho with fast neutrons but found no long-lived activity which could not be attributed to neutron capture and Ho<sup>166</sup>.

In conclusion, the 72.8- and 90.5-keV levels shown in the decay scheme are thought to be reasonably well established, as well as their spins and parities. The 110-keV state in Dy<sup>164</sup> is probably correct but not absolutely forced by the present work. For instance, the existence of a level at about 36.5 keV would give rise to two gammas of almost the same energy. It would be difficult to completely rule out this possibility by the data described above, but the relative intensities in the  $\gamma$ - $\gamma$  coincidence experiment are more consistent with the scheme proposed here. Furthermore, such a low-lying state (36.5 keV) has no precedent in the regular behavior of the many other even-even nuclei. The 36.7-min isomeric transition shown is not at all well established and must be considered to be highly tentative.

## Interaction of Heavy Primary Cosmic Rays in Lead\*

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(Received July 28, 1954)

The energy spectrum of heavy primary cosmic-ray nuclei was measured and found to be in agreement with the assumption that no primaries with energy below the geomagnetic cutoff enter the earth. The mean free path of the primary nuclei in lead was measured. If the transparency of nuclear matter is taken into account, the present results for lead and the measurements by others for lighter elements are in good agreement. If the expression  $R_A = R_0 \times A^{1/3}$  is used for the radius of the nucleus, the data indicate for  $R_0$  the value  $1.3 \times 10^{-13}$  cm. It is also shown that the ratio of the abundances of the charge groups  $Z=12$  to 17 and  $Z=18$  to 28 is  $\sim 2.1$ .

### I. INTRODUCTION

AN "emulsion cloud chamber" consisting of lead plates and photographic emulsions was flown<sup>1</sup> to an altitude of approximately 100 000 feet for about 10 hours. The box was made of aluminum, inside dimensions 4 in.  $\times$  6 in.  $\times$  5  $\frac{3}{4}$  in. In its walls  $\frac{1}{8}$ -in. grooves were cut so that 4 in.  $\times$  6 in.  $\times$   $\frac{1}{8}$  in. lead plates could slide in. The grooves were spaced accurately to within

0.002 in. and the lead plates were machined to the same accuracy.

Number G5 photographic emulsions, 100 $\mu$  thick, mounted on 4-in.  $\times$  6-in. specially cut glass plates, were inserted below each lead plate. The position of a single particle going from plate to plate in the stack could be predicted to within 50–100 $\mu$ . The total number of plates was 22. (In cases of distorted emulsions it was found that the correct angle to be used for the prediction of the position is that given by the part of the track near the air surface. Apparently, in the drying stage, the surface layers dry uniformly, and thus preserve the true angle.)

An ordinary Leitz binocular research microscope

\* Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

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<sup>1</sup> The balloon flight was conducted by the Aero-Medical Field Laboratory, Holloman Air Force Base, New Mexico (41.7°N geomagnetic latitude) on July 13, 1952.