the molybdenum data to give a yield of 5.50 at mass 99 determines all the yields (except one) in the light group between mass 89 and 100 inclusive. The data are also presented in Fig. 2. It is seen that the "spike" at mass 99 to 100, as well as the "shoulder" below mass 93 is confirmed by the new results. Most of the absolute yields are, however, significantly lower than the curve of Glendenin et al. Since even this curve integrated to less than 100 percent, higher yields than the curve of Fig. 2 are indicated for the mass region below 89 and above 100.

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#### PHYSICAL REVIEW

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# Decay of Ga<sup>66</sup><sup>†</sup>

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Scintillation equipment consisting of a three-crystal pair spectrometer, two-crystal Compton spectrometer, and single NaI(Tl) and anthracene crystals used in coincidence have been used to investigate the decay of Ga<sup>66</sup>. Gamma-rays of 4.83 (2), 4.33 (4.5), 4.12 (1.5), 3.78 (2), 3.41 (3), 3.24<sup>1</sup>(2), 2.75 (22), 2.40 (2), 2.18 (6), 1.93 (3.5), 1.58 (<0.4), 1.37 (3), 1.04 (30), and 0.83 (1) Mev energy have been observed. (Intensities, given parentheses, indicate percent of Ga<sup>66</sup> decay.) These are fitted into a consistent decay scheme in which Ga<sup>66</sup> decays to levels in Zn<sup>66</sup> at 4.83 (2), 4.33 (8), 4.12 (1.5), 3.78 (21), 3.41 (4), 3.24 (10), 2.75 (2), 2.40 (~0.5), 1.04 (<0.5) and 0 (56) Mev. (Branching intensities, given in parentheses, indicate percentages.) The betaand gamma-ray branchings have been used to assign possible spins and parities to the states involved.

### INTRODUCTION

HE decay of Ga<sup>66</sup> has been investigated using scintillation detection techniques for gamma- and beta-ray spectroscopy. At the time this work was started the known features of the decay were<sup>1-3</sup> (1) a complex positron spectrum containing components of 4.15, 1.4, 0.9, and 0.4 Mev, and (2) gamma rays of 1.04, 2.74, 4.2, and 4.8 Mev. Since that time a more complete investigation of this problem has been carried out by Mukerji and Preiswerk,<sup>4</sup> more or less concurrently with our work but using for the most part different techniques. They confirmed the positron spectra reported by Langer and Moffat1 and observed new gamma rays of 1.7, 2.2 and 3.3 Mev, and they showed that the 1.04- and 2.75-Mev gamma rays are in coincidence. They also proposed a level scheme consisting of excited states in Zn<sup>66</sup> at 1.05, 2.75, 3.30, 3.80, 4.25, and 4.80 Mev to fit all the data.

The present work has improved the accuracy of the known gamma-ray energies and has shown new gamma rays requiring additional energy levels in Zn<sup>66</sup>. Because of this, and because different techniques have been used, it seems worth while to describe the work in some detail. Our proposed decay scheme is shown in Fig. 1.

#### SOURCE PREPARATION

The Ga<sup>66</sup> was produced by the Zn(p,n) reaction in the University of California, Berkeley, 60-in. cyclotron.<sup>5</sup> Carrier free separation of the Ga was accomplished by the ether extraction process.<sup>6</sup> Sources were prepared either by evaporating a drop of the ether solution on a source holder (thin, weak sources for coincidence work) or by evaporating the entire ether solution and then picking up the Ga<sup>66</sup> activity in a drop of HCl (for strong gamma-ray sources  $\sim 1$  mc).

### EQUIPMENT

Initial investigations using single iodide crystals<sup>7</sup> showed that better gamma-ray spectroscopy was meeded. A three-crystal pair spectrometer<sup>8</sup> of the type originally suggested by Hofstadter<sup>9</sup> and subsequently built by several workers<sup>10</sup> was constructed for this purpose. Excellent rejection of background was achieved by the use of lead shielding and differential pulse height

<sup>†</sup> Supported in part by the joint program of the U. S. Office of Naval Research and U. S. Atomic Energy Commission. \* Now at the University of California Radiation Laboratory,

<sup>Livermore, California.
<sup>1</sup> L. M. Langer and R. D. Moffat, Phys. Rev. 80, 651 (1950).
<sup>2</sup> R. Hofstadter and J. A. McIntyre, Phys. Rev. 80, 636 (1950).
<sup>3</sup> A. Mukerji and P. Preiswerk, Helv. Phys. Acta 23, 516 (1950).
<sup>4</sup> A. Mukerji and P. Preiswerk, Helv. Phys. Acta 25, 387 (1952).</sup> 

<sup>&</sup>lt;sup>5</sup> The staff of the Crocker Radiation Laboratory very kindly produced a number of these sources for us.

<sup>&</sup>lt;sup>6</sup> See, for example, E. Bleuler and G. J. Goldsmith, Experimental Nucleonics (Rinehart and Company, Inc., New York, 1952), p. 196.

<sup>&</sup>lt;sup>7</sup> All of our crystals were rough ground and mounted under magnesium oxide smoked aluminum covers. See, for example, W. H. Jordan, Ann. Rev. Nuc. Sci. 1, 207 (1952).

<sup>W. H. Jordan, Ann. Rev. Nuc. Sci. 1, 207 (1952).
<sup>8</sup> H. I. West and L. G. Mann, Rev. Sci. Instr. (to be published).
<sup>9</sup> R. Hofstadter and J. A. McIntyre, Phys. Rev. 79, 389 (1950).
<sup>10</sup> J. K. Bair and F. C. Maienscheim, Rev. Sci. Instr. 22, 343 (1951); S. A. E. Johansson, Nature 166, 794 (1950); R. S. Foote and G. Kamm, Phys. Rev. 87, 193 (1952); G. M. Griffiths and J. B. Warren, Proc. Phys. Soc. (London) 65, 1050 (1952).</sup> 



FIG. 1. Proposed decay scheme of Ga<sup>66</sup>. The transitions indicated by dashed lines have energies which could not be resolved from the energies of some of the other transitions indicated. In the text all of the observed intensities have been ascribed to the solid-line transitions. The observed gamma-ray energies are shown on each transition. An order number is also given for reference to Table II. Gamma intensities, and uncertainties in energies and intensities are given in Table II. Electron capture and positron intensities are given in Table IV.

discrimination on the side crystal annihilation radiation pulses (0.51 Mev). Figure 2 shows a block diagram of the apparatus as it was used in the pair spectrometer. The discriminator is a cathode-ray tube with a mask such that only the 0.511-Mev annihilation pulses can be picked up by the 931A photomultiplier. The components were also used in double coincidence experiments (0.15  $\mu$ sec resolving time) involving pulse-height discrimination on one or both crystals. The recording cathode-ray tube constitutes a multichannel discriminator, in which the height of each pulse is displayed as a dot which is photographed. The number of dots falling in a given 1-mm channel were counted and plotted as a function of the height of the channel.

The pair spectrometer was tested with the  $Na^{24}$  gamma rays from a 1-mc source collimated with a  $\frac{1}{4}$ -in. diameter by 6-in. hole in lead. Figure 3 shows the gamma-ray spectrum. The tail of low-energy pulses is the result of escape of bremsstrahlung and electrons from the center crystal.<sup>8</sup> It must be taken into account in the interpretation of all data observed with the apparatus.

For gamma rays of less than 2 Mev, the pair spectrometer efficiency was too low, and a two-crystal Compton spectrometer of the type suggested by Hof-



FIG. 2. Block diagram of the three-crystal pair spectrometer.

stadter<sup>11</sup> was constructed. An anthracene crystal was used as the Compton scatterer, to avoid pair production. A  $1\frac{1}{2}$ -in. by  $1\frac{1}{2}$ -in. sodium iodide cylinder was used for detection of the scattered radiation and was biased to reject all pulses greater than 0.3 Mev. The anthracene crystal was  $\frac{1}{2}$  in. thick, capable of stopping 3-Mev electrons, and gave a resolution of 15 to 17 percent for the internal conversion electrons of Cs<sup>137</sup> (0.62 Mev).

The detector arrangement for  $\gamma$ - $\gamma$  coincidence experiments consisted of two sodium iodide crystals placed back to back with the source in between. Copper  $(\frac{3}{32}$  in.) was used to absorb the beta particles. For  $\beta$ - $\gamma$  work an anthracene crystal was used in place of one of the NaI crystals. Pulse-height discrimination, either differential or integral, was used on one crystal while the pulseheight distribution in the other crystal was photographed. Poor geometry was necessary in order to obtain sufficient detection efficiency for coincidence work on weak branches without excessive chance coincidences from the other intense radiations. As a result of this arrangement there is a (calculated) probability of about 10 percent that in the  $\beta$ - $\gamma$  experiments an annihilation quantum was detected in coincidence with a gamma-ray.<sup>12</sup> These addition pulses were subtracted from all of the data (Figs. 4-6). A similar addition effect can occur due to Compton scattering between the two crystals. The copper shielding used between counters was sufficient to reduce this effect by 50 percent. However, it had to be taken into account in the  $\gamma$ - $\gamma$  experiments (Fig. 6); in  $\beta$ - $\gamma$  work the method of subtracting  $\gamma$ - $\gamma$  coincidences also corrected for the Compton scattering coincidences.

#### **EXPERIMENTS**

An outline of the experiments which support the decay scheme of Fig. 1 is given in Table I.

<sup>11</sup> R. Hofstadter and J. A. McIntyre, Phys. Rev. 78, 619 (1950).
 <sup>12</sup> Jastram, Whalen, and Zinke, Rev. Sci. Instr. 23, 648 (1952).

## A. Spectrometer Measurements

The pair spectrometer results are shown in Fig. 7. A  $\sim$ 1-mc source was used with a  $\frac{1}{4}$ -in. diameter by 5-in. lead collimator hole. About six hours of running time were needed to record these data. Similar curves were taken with two other sources and confirmed all the gamma rays shown here.

The method of analysis to take account of bremsstrahlung and electron escape<sup>8</sup> is indicated for the two highest energy lines in the figure. A Gaussian curve has been fitted to the 4.83-Mev line and the calculated tail is indicated. Then the next highest energy line is fitted in the same way, using a Gaussian those width is reduced by the square root of the energy ratio.<sup>13</sup> This rule seems to be completely valid for all our data<sup>8</sup> (see for example the agreement in our Na<sup>24</sup> results, Fig. 3, fitted in the same way). The true



FIG. 3. The Na<sup>24</sup> gamma-ray spectrum (2.76 and 1.38 Mev) observed with the three-crystal pair spectrometer. Note the change by a factor of five in the ordinate scale for the solid points. The beginning of the tail of the 2.76-Mev gamma ray is shown on both scales. The lines through the peaks are Gaussians. Elsewhere the solid line is the calculated tail of the 2.76-Mev peak, which is caused by electron and bremsstrahlung escape from the center crystal.

Gaussian widths were increased by  $\sim 5$  percent at 4.83 Mev and 10 percent at 1.9 Mev to take into account the finite channel width. The curve shown in Fig. 8 was obtained by the above procedure. Of the excess counts observed in the region below 1.93 Mev, 35 percent can be accounted for by pair production of the 1.58- and 1.37-Mev gamma rays, the peaks of which are not resolved because of poor statistics. We believe that the remainder is the result of insufficient lead shielding, which caused low energy background in addition to the bremsstrahlung tail. (When more shielding was used, as in Fig. 3, the same calculational procedure gave excellent agreement with the data.)



FIG. 4. Beta-gamma coincidences in Ga<sup>66</sup>. The gamma-ray pulse-height distribution in a single sodium iodide crystal is shown for the pulses in coincidence with beta rays of <0.5-Mev energy. (An anthracene crystal was used as the beta spectrometer.) The points were fitted by photo, Compton, and pair distributions of the type shown for the 3.41-Mev gamma ray (dotted curve). The intensities which give the resultant curve shown are given in Table III, third column. The data for the solid points were taken with a source seven times the intensity of that used for the circled points. Experimental gamma-gamma and chance coincidences have been subtracted.

The valleys between peaks fall somewhat below the points in most cases. An effect which can partially explain this is Compton scattering of the annihilation quanta as they leave the center crystal. Recoil electrons of 100-kev energy are produced in the center crystal for scattering angles of 40°, and lower energies for smaller angles. These events can produce coincidences if the side crystal discriminator channels extend down to 0.4 Mev (as they did, to include the entire 0.511-Mev photopeak). The result is to broaden the pair peaks on the high energy side. This effect is most pronounced on low energy peaks and has been verified for Co<sup>60</sup> pair peaks.<sup>14</sup> Using the differential Compton scattering cross



FIG. 5. Beta-gamma coincidences in Ga<sup>66</sup>. Same as Fig. 4 except that the beta detector was biased so that only pulses of >1.0-Mev energy were accepted. Table III gives the intensities.

<sup>&</sup>lt;sup>13</sup> A. W. Schardt and W. Bernstein, Rev. Sci. Instr. **22**, 1020 (1951). We seem to obtain better agreement with the  $E^{\pm}$  law than these authors. It may be that the effect of photomultiplier cathode non-uniformity was reduced because of collimation and the elimination of all secondary processes in the crystal. This tends to localize the source of light in a small region of the crystal.

<sup>&</sup>lt;sup>14</sup> H. I. West, Ph.D. thesis, in preparation, Stanford University, Stanford, California.



FIG. 6. Gamma-gamma coincidences in Ga<sup>66</sup>. The pulse-height distribution in a single NaI(Tl) crystal is shown for the pulses in coincidence with gamma-ray pulses of >2.15 Mev in a second NaI(Tl) crystal. The 1.58-Mev peak may be entirely the result of annihilation radiation addition to the 1.04-Mev pulses. The dotted line is the Gaussian tail of the 1.04-Mev peak, shown on the scale of the 1.58-Mev peak.

section for 0.51-Mev gamma rays and the solid angles of our side crystals, and referring to the 2.75-Mev gamma-ray peak in Fig. 7, one can calculate that the 3.0-Mev point should be increased by a maximum of 40 counts and the 3.1-Mev point by 10 counts. Similarly, up to 50 percent of the discrepancy at 4.6 Mev can be removed. This effect could have been prevented by proper setting of the side crystal discriminators, but it was not expected at the time of the experiments.

In order to take into account the effect of the excess low energy pulses and the valley effect described above, the peak heights indicated in Fig. 7 for the 2.40-, 2.18-, and 1.93-Mev gamma rays were reduced by 50, 11, and 13 percent, respectively, for the intensity calculation (Table II). Although these figures cannot be very



FIG. 7. Gamma-ray spectrum of Ga<sup>66</sup> observed with the threecrystal pair spectrometer. The calculated bremsstrahlung and electron escape tails are shown for the two highest energy peaks. The final curve is a superposition of the Gaussian plus tail distributions of each gamma ray. The measured gamma-ray energy of each Gaussian peak is given on the diagram in Mev. The 2.76-Mev peak is shown with the ordinate scale reduced by a factor of 10 (solid points).

accurate, they are of the correct order of magnitude and are consistent with other experiments (see below).

In order to determine gamma-ray intensities the pair cross section of sodium iodide is needed as a function of gamma-ray energy. We have determined three points on this curve, at 1.17, 1.33, and 1.38 Mev, by analyzing the peaks obtained in the pair spectrometer with Co<sup>60</sup> and Na<sup>24</sup> sources.<sup>14</sup> With the help of these points the Bethe-Heitler formula was extended to low energies as shown in Fig. 8. The points of Yaeger and Hulme,15 based on exact calculation, of Colgate,<sup>16</sup> based on absorption measurements and theoretical Compton, photoelectric, and Rayleigh cross sections, and of Dayton,17 based on measured Z dependence and the Born approximation calculation at low Z, are also shown. (The Born approximation should be valid at low Z. This was veri-

TABLE I. Experimental evidence supporting the proposed decay scheme of  $Ga^{66}$  (Fig. 1).

Level (Mev)	Evidence				
4.83					
4.33	$\gamma$ decay to ground				
4.12					
3.78	$\gamma$ decay to ground 2.75-1.04 Mev $\gamma$ coincidence (Mukerji, <sup>a</sup> present authors) 0.4 Mev $\beta$ -spectrum (Mukerji <sup>a</sup> )				
3.41	$\gamma$ decay to ground $\beta$ - $\gamma$ coincidence: $\beta < 0.5$ Mev $\rightarrow 3.41 \gamma$ $\beta > 1.0$ Mev $\rightarrow$ no 3.41 $\gamma$				
3.24	$\gamma$ decay to ground 0.9 Mev $\beta$ -spectrum (Mukerji <sup>a</sup> ) $\beta$ - $\gamma$ coincidence: $\beta < 0.5$ Mev $\rightarrow 2.18$ , 3.24 $\gamma$ $\beta > 1.0$ Mev $\rightarrow no$ 2.18, 3.24 $\gamma$				
2.75	1.4 Mev $\beta$ -spectrum (Mukerji <sup>a</sup> ) $\beta$ - $\gamma$ coincidence: $\beta > 1.0$ Mev $\rightarrow 2.75 \gamma$				
2.40	$\gamma$ decay to ground $\beta$ - $\gamma$ coincidence: $\beta$ >1.0 Mev $\rightarrow$ 1.37, 1.04 $\gamma$ (cascade) 1.93, 1.37, and 0.83 gammas fit this level				
1.04	2.75–1.04 Mev $\gamma$ coincidence (Murkerji, <sup>a</sup> present authors) Cu <sup>86</sup> decay <sup>b</sup>				

<sup>a</sup> References 3 and 4.
 <sup>b</sup> G. Friedlander and D. E. Alburger, Phys. Rev. 84, 231 (1951); Roderick, Meyerhof, and Mann, reference 30.

fied for anthracene.<sup>14</sup>) Small corrections to the resulting intensities (of the order of 10 percent) were made for counts lost due to positrons which escape from the center crystal or annihilate in flight. The final results of the energy and intensity analysis are given in the second and third columns of Table II.

For the energy region below 2 Mev the two-crystal Compton spectrometer was more useful. Figure 9 shows the gamma-ray spectrum up to the 2.75-Mev gammaray as observed with this instrument. Energies were obtained from a continuous photograph of the pulses, a photometer curve of which is shown in Fig. 10. The important results from these curves are that gamma rays of 0.83-, 1.37-, and 1.96-Mev energy exist in addi-

<sup>16</sup> S. A. Colgate, Phys. Rev. 87, 592 (1952).
 <sup>17</sup> I. E. Dayton, Phys. Rev. 89, 544 (1953).

<sup>&</sup>lt;sup>15</sup> J. C. Yaeger and H. R. Hulme, Nature 137, 781 (1936); 148, 86 (1941).

	Pair spectrometer		· Compton sp	ectrometer		Final result		
Gamma- ray	Energy (Mev)	<b>Relative</b> intensity	Energy (Mev)	<b>Relative</b> intensity	Energy (Mev)	Intensity (percent of Ga <sup>66</sup> decay)		
1 2 3 4 5 6, 7 8, 9 10 11 11 12 13 14 15	$\begin{array}{c} 1.90 \pm 0.04\\ 2.17 \pm 0.03\\ 2.40 \pm 0.04\\ 2.75^{a}\\ 3.24 \pm 0.04\\ 3.41 \pm 0.04\\ 3.78 \pm 0.03\\ 4.12 \pm 0.05\\ 4.33 \pm 0.05\\ 4.83 \pm 0.05\\ \end{array}$	$16.0\pm3.0 \\ 26.9\pm2.7 \\ 6\pm4 \\ 100 \\ 8.6\pm1.8 \\ 13.5\pm2.5 \\ 8.5\pm1.3 \\ 6.9\pm1.5 \\ 20.7\pm2.0 \\ 9.7\pm1.0 \\ 1000$	$\begin{array}{c} 0.511^{a} \\ 0.83 \pm 0.01 \\ 1.044^{a} \\ 1.37 \pm 0.02 \\ 1.94 \pm 0.02 \\ 2.19 \pm 0.02 \\ 2.75 \end{array}$	$ \begin{array}{c} 10\pm6\\ 138^{b}\\ 14.0\pm6\\ 17.8\pm6\\ 26.9\\ <9\\ 100 \end{array} $	$\begin{array}{c} 0.83\\ 1.044\\ 1.37\\ 1.93\\ 2.18\\ 2.40\\ 2.75\\ 3.24\\ 3.41\\ 3.78\\ 4.12\\ 4.33\\ 4.83\end{array}$	$\begin{array}{c} 2.2 \pm 1.4 \\ 30.0 \\ 3.0 \pm 1.1 \\ 3.5 \pm 1.2 \\ 5.8 \pm 0.6 \\ 2.4 \pm 1.6^{\rm e} \\ 21.6^{\rm d} \\ 1.9 \pm 0.4 \\ 2.9 \pm 0.6 \\ 1.8 \pm 0.3 \\ 1.5 \pm 0.3 \\ 4.5 \pm 0.5 \\ 2.1 \pm 0.2 \end{array}$		

TABLE II. Ga<sup>66</sup> gamma rays measured with the pair and Compton spectrometers.

Gamma-ray energies used for calibration are given without errors. The 1.044-Mev energy was measured by G. Friedlander and D. E. Alburger, Phys. <sup>a</sup> Gamma-ray energies used for calibration are given without errors. The 1.044-MeV energy was measured by G. Friedlander and D. E. Alburger, Phys. Rev. 84, 231 (1951). <sup>b</sup> The intensities given without errors were used for intensity normalization. The value of 138 is obtained from our decay scheme by adding up all the transitions feeding the 1.04-MeV level.  $\circ \gamma_{s} = 1.6_{-1} \epsilon^{+0}; \gamma_{\tau} = 0.8_{-0} \epsilon^{+0}$ . These figures are consistent with the upper limits found in the  $\beta - \gamma$  coincidence experiments.  $d \gamma_{8} = 19.4 \pm 2; \gamma_{9} = 2.2 \pm 1$ .

tion to others known from the pair spectrometer and single crystal data. No 2.40-Mev gamma ray is detectable, but we can only assign an upper limit of  $\sim \frac{1}{3}$ the intensity of the 2.18-Mev gamma ray. This is the same order of magnitude that was observed in the pair spectrometer.

their Compton recoil data and private communication with Professor Preiswerk, we believe that this was a misinterpretation of a peak due to the 1.93-Mev gamma-ray.

### **B.** Coincidence Measurements

The Compton spectrometer curve was extended down to the 0.39- and 0.30-Mev gamma rays of Ga<sup>67</sup> (78 hr half-life)<sup>18</sup> in order to be sure that the peak at 0.83 Mev is not caused by the 0.88-Mev transition in Ga<sup>67</sup>. This transition is less than 3 percent of the 0.30-Mev intensity, whereas our observed peak was 40 percent. It is disturbing that Mukerji and Preiswerk<sup>4</sup> could not see this gamma-ray in their beta-ray spectrometer experiments using a lead radiator. However, the intensity that we observe is near the limit of their resolution, and it does not seem to be excluded entirely by their data.<sup>19</sup>

An intensity analysis was carried out using the differential Compton cross section at 135°.20 The 2.75and 2.18-Mev peaks were matched to the pair spectrometer results for normalization. Because of greater amounts of electron escape from the low density anthracene crystal there is considerably more background than in the pair spectrometer, making the quantitative analysis less accurate. The fourth and fifth columns in Table II show the energies and intensities obtained from the Compton spectrometer, and the last two columns show our final results expressed in terms of the total number of Ga<sup>66</sup> decays (assuming our decay scheme).

These results confirm the gamma rays reported by Mukerji and Preiswerk<sup>4</sup> with the exception of their 1.7-Mev transition. On the basis of a re-examination of

In coincidence work the procedure was to isolate as far as possible certain parts of the beta- or gamma-ray spectrum, using a single crystal as a discriminator, and to observe the coincident radiations in a second crystal. This is difficult to do for a decay scheme as complicated as that shown in Fig. 1, except for favorable regions of the spectra. We have performed  $\beta$ - $\gamma$  coincidence experiments as indicated in Table I, using beta rays >1.0Mev, and <0.5 Mev, and analyzing the coincident gamma-ray pulse-height distributions. The gamma-ray distribution in coincidence with gamma rays of >2.15Mev has also been analyzed.

Figure 4 shows the gamma-ray pulse heights observed in coincidence with beta rays of < 0.5 Mev. This experiment was expected to enhance the radiations following the decay of the 3.78-Mey level and in particular to show the intensity of the ground-state transition from that level. However, it was not possible to put a very small upper limit on this transition because of the complexity of the spectra.

Figure 5 shows the gamma-ray pulse heights observed in coincidence with beta rays of >1.0 Mev. This eliminates completely the strong 2.75- to 1.04-Mev gamma-ray cascade and enables one to observe the decay of the 2.75- and 2.40-Mev levels (if they are fed by beta decay).

Figure 6 shows the gamma-ray pulse heights observed in coincidence with gamma-ray pulses of >2.15 Mev. In this experiment the discriminated crystal receives mainly the 2.75-Mev gamma ray, so the strong 2.75- to 1.04-Mev gamma cascade is emphasized. Any gamma

 <sup>&</sup>lt;sup>18</sup> Ketelle, Brosi, and Porter, Phys. Rev. 90, 567 (1953);
 Meyerhof, Mann, and West, Phys. Rev. 92, 756 (1953).
 <sup>19</sup> Private communication from Professor Preiswerk.

<sup>20</sup> C. M. Davisson and R. D. Evans, Revs. Modern Phys. 24, 79 (1952).

rays feeding the 2.75-Mev or higher levels may also be detected.

The quantitative analysis of the coincidence experiments are illustrated in Fig. 4. In this analysis the aforementioned addition of annihilation radiation must be corrected for. Also, parts of the higher energy beta spectra are detected in the beta counter. This effect is appreciable because, while only a fraction of the higher energy spectra are detected, the ratio of  $\beta^+$  to K-capture decay increases with increasing energy.

The experimental points were fitted by means of Gaussian photo and pair distributions and a flat Compton distribution for each gamma ray. The three distributions are shown for the 3.41-Mev gamma ray. The number of counts in each group was adjusted according to the relative (experimental) cross sections. In order to compare these data with a given decay scheme, the absolute beta and gamma detection efficiencies are needed. We have determined our gamma detector efficiency accurately for  $Co^{60}$  gamma rays (1.33 and 1.17 Mev) by coincidence experiments. The



FIG. 8. Pair-production cross section in iodine as a function of gamma-ray energy above threshold. (1) West thesis (pair spectrometer); (2) Dayton (measured Z dependence and Born approximation at low Z limit); (3) Colgate (measured total cross section minus theoretical Compton, photoelectric, and Rayleigh cross sections); (4) Yaeger and Hulme (exact calculation). Experimental accuracy is given by the size of the circles. The dashed line is the Born approximation calculation. The cross section of sodium is found from Dayton's data to be 4 percent of that for iodine at 1.33 and 2.62 Mev, and the Born-approximation calculation at higher energies gives the same ratio.



FIG. 9. Gamma-ray spectrum of  $Ga^{66}$  observed with a two-crystal Compton spectrometer. The abscissa is electron recoil energy at 135° scattering. The corresponding gamma-ray energies are indicated for the peaks. For the 2.75- and 1.04-Mev gamma-ray peaks the tails caused by electron escape from the anthracene crystal are shown.

other gamma-ray efficiencies, for energies up to 3.78 Mev, can then be calculated with sufficient accuracy because of the relatively small dependence of cross section on energy in this region. The beta-ray detection efficiency was assumed to be  $[1 \times (\Omega/4\pi)]$  for energies within the discriminator channel, where  $\Omega$  is the solid angle subtended by the detector at the source.

The number of coincidences is given by

$$n_{\beta\gamma_j} = n_{\gamma_j} \sum_{i} \left[ \alpha_i^{j} E_{\beta} \frac{\beta_i}{\beta_i + K_i} \times \delta_i \right],$$

where

- $n_{\gamma j} = N_{\gamma j} E_{\gamma j} =$  number of counts in gamma-ray counter due to a particular gamma ray,
- $\alpha_i^{j}$  = probability that  $\gamma_j$  is in coincidence with beta transition  $\beta_i$  (or the corresponding K-capture),
- $E_{\beta} = 1 \times (\Omega_{\beta}/4\pi) = \text{beta-detector efficiency},$
- $\beta_i$  number of positrons
- $\frac{\beta_i}{\beta_i + K_i} = \frac{\beta_i}{\text{number of decays}} \text{ for beta transition } \beta_i,^{21}$ 
  - $\delta_i$  = fraction of  $\beta_i$  spectrum accepted by the integral discriminator (energy <0.5 Mev),
  - $N_{\gamma_j}$  = number of  $\gamma_j$  emitted by the source,
  - $E_{\gamma j} =$  efficiency of the gamma-ray crystal for  $\gamma_j$ .

The  $\alpha_i{}^i$  depend on the decay scheme, and  $\beta_i{}/(\beta_i{+}K_i)$ and  $\delta_i$  were obtained from theoretical curves assuming allowed beta spectra. In order to show whether or not a particular decay scheme was consistent with all the coincidence data, we calculated each  $N_{\gamma}$ , using the observed  $n_{\beta\gamma}$  and the  $\alpha_i$  for the particular scheme. For the correct scheme the  $N_{\gamma}$  must agree with the spectrometer results.

Table III shows the results for each of the coincidence experiments, using our final decay scheme (Fig. 1). The agreement with the spectrometer measurements is excellent, considering the limited accuracy with which

<sup>21</sup> E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).

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FIG. 10. Densitometer curve of a photograph of the Ga<sup>66</sup> gamma-ray pulses in the two-crystal Compton spectrometer. Abscissa is recoil electron energy at 135° scattering. The corresponding gamma-ray energies are indicated on the peaks. There are about twice as many total counts as in Fig. 9. The region below 0.51 Mev consists of gamma rays in the Ga<sup>67</sup> decay (unresolved by the densitometer). Note ordinate scale change by an undetermined factor.

values could be assigned to the  $n\beta_{\gamma j}$  in Figs. 4-6. Because of this limited accuracy, questions about the existence of certain weak branches (indicated by dashed lines in our decay scheme) cannot be decided. On the other hand, the 0.83-Mev gamma ray is a good example of a significant result found in the  $\beta < 0.5 - \gamma$  experiment. Without this transition feeding the 2.40-Mev level, the intensity of the 1.37-Mev gamma ray would have to be six times greater in order to explain the 1.37-Mev peak observed in this experiment. The 1.58-Mev gamma ray was observed only in the  $(\gamma > 2.15 - \gamma)$  experiment and its actual existence must be questioned because of the amount of annihilation radiation addition that had to be corrected for. Forty percent of the 1.58-Mev peak in Fig. 6 was calculated to be due to addition to the 1.04-Mev peak of annihilation quanta from positrons feeding the 2.75-1.04 Mev gamma-ray cascade.

## DECAY SCHEME

Our final decay scheme of Ga<sup>66</sup> is shown in Fig. 1. The 2.75- and 2.40-Mev levels of Zn<sup>66</sup> and their associated radiations require some discussion. The 2.75-Mev level was reported by Mukerji and Preiswerk<sup>4</sup> on the basis of an observed 1.4-Mev beta spectrum. Our  $(\beta > 1.0 - \gamma)$  coincidence experiments confirm this conclusion. This level also offers our most satisfactory explanation for the 1.58-Mev gamma ray. There seems to be no evidence either in our work or in Mukerji's<sup>19</sup> for a 1.7-Mev gamma ray from this level.

The 2.40-Mev level fits the 0.83-, 1.37-, 1.93- and 2.40-Mev gamma rays observed in the spectrometers. Also, the 0.83-1.37-1.04-Mev cascade from the 3.24-Mev level proves to be the main source of the 1.37-Mev gamma ray appearing in the ( $\beta < 0.5-\gamma$ ) experiment. Without this cascade some other way is needed to explain the 1.37-Mev peak in that experiment. The 2.40-Mev level is fed primarily by the 1.93- and 0.83-Mev gamma rays. A small amount of beta decay to

that level is needed to account for the 1.37- and 1.04-Mev gamma rays seen in the  $(\beta > 1.0 - \gamma)$  experiment.

The transitions indicated by dashed lines in Fig. 1 have energies which duplicate closely those for transitions appearing elsewhere in the decay scheme. Because of the uncertainty in assigning values to the coincidence rates we cannot say to what extent these transitions occur; they have been excluded from the scheme merely because they would change intensities in the wrong direction. The 2.75-Mev gamma ray was split between two different transitions (8 and 9) in order to agree with the results in Figs. 5 and 6 (Table II gives the amount of splitting). The 2.40-Mev gamma ray was also split between two transitions (6 and 7), because the coincidence experiments indicated an upper limit for these transitions which was too small to account for the total 2.40 intensity.

Table IV shows the intensities of the transitions from Ga<sup>66</sup> to each level of Zn<sup>66</sup> as calculated for our decay scheme. In this calculation we have used our observed relative gamma-ray intensities and the ratio measured by Mukerji for the ground state (4.14 Mev) beta spectrum intensity to total beta intensity. The theoretical  $K/\beta^+$  ratios of Trigg<sup>21</sup> have been used to obtain the relative beta and *K*-capture intensities, as was done by Mukerji. In addition, 9 percent of the electron capture was assumed to be from the *L* shell.<sup>22</sup> Log *ft* values<sup>21,23</sup> are given in the tenth column of Table IV.

Our results are compared in Table IV with those of Mukerji (which are nearly identical with Langer and Moffat's results). Mukerji's quoted intensities are based on an assumed 4 percent electron capture to the three highest levels. We have recalculated these intensities in columns 8 and 9 of Table IV using our measured

TABLE III. Gamma-ray intensities deduced from the coincidence experiments.

	Gamma		Total $\gamma$ intensity (percent of Ga <sup>66</sup> decays)			
Experiment	energy (Mev)	Number of coincidences	Coincidence	Spec- trometer		
$\beta < 0.5 \text{ Mev} - \gamma$	1.04	7500	25.5	30.0		
	1.37	1600	3.7	3.0		
(Fig. 4)	2.18	4600	6.3	5.8		
	2.40	<700	<2.3	2.4		
	2.75	1700	21.6ª	21.6		
	3.24	1600	2.4	1.9		
	3.41	1500	3.0	2.9		
	3.78	<300	<4.1	1.8		
$\beta > 1.0 \text{ Mev} - \gamma$	1.04	1000	31.3 <sup>b</sup>	30.0		
,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	1.37	960	3.9b	3.0		
(Fig. 5)	2.40	<150	<2.3 <sup>b</sup>	2.4		
(	2.75	820	21.6ª	21.6		
$\gamma > 2.15 - \gamma$	1.04	12 700	30.0ª	30.0		
(Fig. 6)	1.58	740	<0.4			

<sup>a</sup> These intensities were matched to the spectrometer results for normalization.

<sup>b</sup> These coincidence results are based on the decay scheme of Fig. 1, with a 1.7-Mev beta spectrum intensity of 0.5 percent.

<sup>22</sup> M. E. Rose and J. L. Jackson, Phys. Rev. 76, 1540 (1949).
 <sup>23</sup> S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

			β <sup>+</sup> a	and E.C. intensiti	es (percent o	of Ga <sup>66</sup> decay	)			
Zn <sup>66</sup> level (Mev)	Spectrum notation	$\beta^+$ energy (Mev)	Present	authors	Mu	kerji	Mu (correc present	kerji cted by authors)	Lo Present authors	og <i>ft</i> Mukerji
4.83 4.33 4.12 3.78 3.41 3.24 2.75 2.40 1.04 0	$K_{1}' \\ K_{2}' \\ K_{3}' \\ \beta_{1} \\ \beta_{2} \\ \beta_{3} \\ \beta_{4} \\ \beta_{5} \\ \beta_{6} \\ \beta_{7}$	$\begin{array}{c} 0.02\\ 0.36\\ 0.73\\ 0.90\\ 1.39\\ 1.74\\ 3.10\\ 4.14 \end{array}$	$egin{array}{c} eta^+ \ 0.7 \pm 0.1 \ 1.3 \pm 0.3 \ 4.7 \pm 0.7 \ 1.7 \pm 0.9 \ 0.5 \pm 0.3^{\mathrm{a}} \ < 0.5 \ 51.7 \pm 7 \end{array}$	E.C. 2.1 $\pm$ 0.2 7.9 $\pm$ 0.9 1.5 $\pm$ 0.3 20.7 $\pm$ 1.7 2.9 $\pm$ 0.5 4.1 $\pm$ 0.8 0.5 $\pm$ 0.2 0.5	β <sup>+</sup> 1.1 4.5 2.7 56.0	$\begin{cases} K-cap. \\ 4.0 \\ 26.2 \\ 4.5 \\ 0.5 \\ 0.5 \end{cases}$	$\beta^+$ 1.0 4.1 2.5 51.6	K-cap.	$ \begin{array}{r} 4.7 \\ 4.9 \\ 5.9 \\ 5.1 \\ 6.2 \\ 6.0 \\ 7.2 \\ \sim 7.9 \\ > 9.3 \\ 7.8 \\ \end{array} $	4.9 5.7 4.9 5.9 7.1 7.8

TABLE IV. Beta spectra of Ga<sup>66</sup>.

a If our measured gamma-ray intensities are used, we obtain  $-1.4\pm2.0$  for this beta-decay intensity. However, the  $\beta > 1.0 - \gamma$  coincidence seems to indicate that the beta spectrum does exist.

value of 11.5 percent electron capture to these three levels, but preserving Mukerji's relative beta-spectrum intensities. Agreement with our results is then very good. The failure of the beta-spectrometer workers to report the weak 0.73- and 1.74-Mev beta spectra is not a serious objection to our level scheme, because of the difficulty in separating these spectra from the other components in the beta spectrum.

The ratio of total K-capture to total beta decay in Ga<sup>66</sup> has been measured by Langer and Moffat,<sup>1</sup> by comparison of the Auger electron intensity in a beta spectrometer with the beta spectrum intensity. They find the Auger intensity to be 28 percent of the total beta intensity. This indicates that 38 percent of the Ga<sup>66</sup> decay is by orbital electron capture, if one uses the L/K capture ratio of Rose and Jackson<sup>22</sup> (0.09), and the best known values<sup>24</sup> for fluorescence yield (0.48). It is believed that this figure can be as much as 4 percent low because of unknown effects resulting from

TABLE V. Spins and parities of Zn<sup>66</sup> levels.

Zn <sup>66</sup> level (Mev)	Possible spin, parity $(Ga^{66}=1+)$							
	Log ft	From $\beta$ decay	From $\gamma$ -ray branching	Best choice				
4.83 4.33 4.12 3.78 3.41 3.24 2.75 2.40 1.04 0	$ \begin{array}{r} 4.7 \\ 4.9 \\ 5.9 \\ 5.1 \\ 6.2 \\ 6.0 \\ 7.2 \\ \sim 7.9 \\ > 9.3 \\ 7.8 \\ \end{array} $	$\begin{array}{c} 0, 1, 2+\\ 0, 1, 2+\\ 0, 1, 2+\\ 0, 1, 2+\\ 0, 1, 2+\\ 0, 1, 2+\\ 0, 1, 2+\\ 1-\\ 0, 1, 2+\\ 1, 2-\\ 3, 2, 0-\\ 0, 1, 2+\\ 3+\\ 0, 2-\\ 0, 1, 2+\\ 0, 2-\\ 0, 2-\\ \end{array}$	$ \begin{array}{c} 1\pm\\1\pm\\1\pm\\1\pm\\2\pm\\1\pm\\2\pm\\1\pm\\2\pm\\2\pm\\2\pm\\2+\\0+\end{array} $	$ \begin{array}{r} 1+\\ 1+\\ 1+\\ 0,2+\\ 1+\\ 2+\\ 1+\\ 2+\\ 2+\\ 2+\\ 2+\\ 0+\\ \end{array} $				
2.40 1.04 0	~7.9 >9.3 7.8	$\begin{cases} 1, 2-\\ 3, 2, 0-\\ 0, 1, 2+\\ 0, 1, 2+\\ 0, 2-\\ 0, 1, 2+\\ 3-\\ 0, 2-\\ 0, 1, 2+ \end{cases}$	2± 2+ 0+	-				

<sup>24</sup> Broyles, Thomas, and Haynes, Phys. Rev. 89, 715 (1953).

source absorption and scattering.<sup>25</sup> Our results based only on gamma-ray intensities are in excellent agreement (Table IV).

#### CONCLUSIONS

The ground and first excited states of Zn<sup>66</sup> have been assumed to have even parity and spins 0 and 2, respectively, in accordance with the empirical data on eveneven nuclei.26 For the other levels we have used the theoretical<sup>27,28</sup> and empirical <sup>29,30</sup> data for beta decay and gamma-ray lifetimes in order to see if reasonable spin and parity assignments could be made. This is possible, with Ga<sup>66</sup> spins of either 0 or 1, for all the levels except the three lowest, where no set of assignments is found which does not have some apparent discrepancy with the existing knowledge of beta and gamma decay. It must be emphasized, though, that the use of Weisskopf's formula and ft values alone does not make possible a very accurate assignment of spins and parities in this case.

Table V shows the possible Zn<sup>66</sup> assignments for Ga<sup>66</sup> having spin 1 and even parity (which seems to be the least objectionable choice). The assignments based on beta decay log ft values and on gamma-ray branchings are listed in order of preference in columns 3 and 4, respectively. In most cases a change of unity in the spin of a level changes the gamma-ray branching ratio by a factor of at least 50. The fifth column gives the values which seem to be the best compromise. It is seen that the three lowest levels give poor agreement with Nordheim's classifications of log ft values.<sup>29</sup> The 1.04-Mev level appears to require a forbidden beta spectrum

<sup>&</sup>lt;sup>25</sup> Professor Langer (private communication).

<sup>&</sup>lt;sup>26</sup> G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).
<sup>27</sup> E. J. Konopinski, Revs. Modern Phys. 15, 209 (1943).
<sup>28</sup> V. F. Weisskopf, Phys. Rev. 83, 1073 (1951). We have used the nomogram from R. Montalbetti, Can. J. Phys. 30, 660 (1952).

 <sup>&</sup>lt;sup>30</sup> L. W. Nordheim, Revs. Modern Phys. 23, 322 (1951).
 <sup>30</sup> Summary Report of the Indiana Conference on Nuclear Spectroscopy and the Shell Model, Indiana University, May, 1953 (unpublished).

with a spin of 3 for the level.<sup>31</sup> The assignment of spin 0<sup>+</sup> for Ga<sup>66</sup> would be more favorable in this case, but then there is a discrepancy in the beta decay to the ground state.

If Ga<sup>66</sup> is 1<sup>-</sup>, then it is possible to assign spins of 1<sup>-</sup> to all of the Zn<sup>66</sup> states above 2.40 Mev, and this would agree with Glaubman's recent proposal<sup>32</sup> that low lying levels of even-even nuclei have odd spin if the parity is odd, and even spin if the parity is even. The greatest objection to this would be the assignment of  $1^-$  to the 3.78- and 3.24-Mev levels which decay predominantly to the 1.04-Mev level. Odd parity for Ga<sup>66</sup> would not agree with the shell model prediction  $P: P_{\frac{3}{2}} N: (p_{\frac{3}{2}})^4 (f_{\frac{5}{2}})^3$ or  $(p_{\frac{3}{2}})^3 (f_{\frac{5}{2}})^4$  (P:proton configuration, N:neutron configuration).

It does not seem possible to say anything conclusive about the states above 1.04 Mev without internal conversion and angular correlation data.<sup>33</sup>

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## PHYSICAL REVIEW

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# Half-Life Determination of Po<sup>210</sup> by Alpha Counting\*

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A half-life of  $138.374 \pm 0.032$  days for polonium-210 was determined by alpha counting a sample of approximately 0.5 millicurie over a period of 328 days.

HE National Bureau of Standards<sup>1</sup> lists two values for the half-life of Po<sup>210</sup>: 140 days as determined by Curie<sup>2</sup> by gamma counting, and 138.3 days as determined by Beamer and Easton<sup>3</sup> by calorimetry. Alpha counting affords an entirely different method of determining the half-life, and requires only a small amount of activity. Therefore, for comparison, a half-life determination by alpha counting was undertaken, with a sample of approximately 0.5 millicurie of Po<sup>210</sup>.

The counting instrument chosen was the Logac,<sup>4</sup> a low-geometry alpha counter chosen for its stability and low-coincidence loss. It consists of a low-geometry attachment used with a methane-flow proportional alpha counter. The sample was kept in the counting chamber throughout the experiment, so that no changes in geometry could occur.

The sample was pipetted from a solution of purified Po<sup>210</sup> in nitric acid onto a glass slide. Mica, weighing  $0.92 \text{ mg/cm}^2$ , was cemented over the sample to prevent migration of activity from the slide. Tests made by adding air to the evacuated counting chamber and counting at varying air pressures showed that the sample was sufficiently thin that no counts were lost by absorption or would be lost by diffusion into the glass.

Over a period of 328 days, 81 measurements were made. Each measurement was of sufficient duration to total at least 500,000 counts, to reduce the statistical probable error to 0.1 percent per measurement. No geometry factor was used, since the decay could be followed from the counting rate.

A least-squares analysis of the data gave a half-life of 138.374 days which compares favorably with the Beamer and Easton value. The probable error in the determination was 0.032 day or 0.02 percent, as compared with 0.1 percent by Beamer and Easton.<sup>3</sup>

<sup>&</sup>lt;sup>31</sup> It would be very unusual if this spin is not 2. Furthermore, the lifetime of this level has been found to be  $<5\times10^{-9}$  sec which makes a spin >2 very unlikely. [Roderick, Meyerhof, and Mann, Phys. Rev. 84, 887 (1951).] <sup>32</sup> M. J. Glaubman, Phys. Rev. 90, 1000 (1953).

<sup>&</sup>lt;sup>33</sup> Mukerji and Preiswerk mention an (unpublished) angular correlation measurement of the 2.75-1.04-Mev gamma cascade which was inconclusive because of unknown contributions to the coincidences due to weaker components. An accurate measurement of this angular correlation could determine whether the 3.78-Mev level has spin 0 or 2. If this spin is 2 then the possibility of spin 0 for Ga<sup>66</sup> must be ruled out because of the allowed beta transition to the 3.78-Mev state of Zn<sup>66</sup>.

<sup>\*</sup> Operated by Monsanto Chemical Company under a U.S. Atomic Energy Commission contract.

<sup>&</sup>lt;sup>1</sup> Nuclear Data, National Bureau of Standards Circular No. 499 (U. S. Government Printing Office, Washington, D. C., 1950),

<sup>p. 247.
<sup>2</sup> M. Curie, J. phys. et radium (6) 1, 12 (1920).
<sup>3</sup> W. H. Beamer and W. E. Easton, J. Chem. Phys. 17, 1298</sup> 

<sup>&</sup>lt;sup>4</sup> Rose, DeBenedetti, Heyd, Pittenger, Powers, Brenneman, and Curtis, Mound Laboratory Final Report No. 47, M-270, 1947 (unpublished).