

Parkinson,¹⁵ however, has reported configuration mixing to be present in the shell model description of the ground state of P^{31} , and such mixing can be used to explain the presence of the branch in the decay of S^{31} . In the unified model description, the configurations are also expected to be mixed,¹⁴ and again this would allow a transition to the first excited state of P^{31} in the decay of S^{31} .

¹⁵ W. C. Parkinson, *Phys. Rev.* **110**, 485 (1958).

ACKNOWLEDGMENTS

We wish to thank Dr. D. J. Zaffarano for the initial suggestion of the experiment and for his constant interest and encouragement, and Dr. B. C. Carlson for the helpful discussions concerning the interpretation of the results. We are also indebted to Dr. A. J. Bureau and the staff of the synchrotron for their assistance and cooperation during the experimental portions of this investigation.

Decay of Ga^{66} and $Cu^{66}\dagger$

ARTHUR SCHWARZSCHILD AND LEE GRODZINS*
Brookhaven National Laboratory, Upton, New York

(Received February 11, 1960)

The 9.5-hour decay of Ga^{66} and the 5.1-minute decay of Cu^{66} have been studied by a variety of techniques including gamma-ray spectroscopy, internal conversion measurements, and angular correlation studies. All but one of the 18 gamma rays observed have been ordered into a structure of 11 levels and the spins and parities of all but one of these levels have been determined. It is shown that Ga^{66} has spin zero and even parity and that its 4.166-Mev β spectrum is a pure Fermi transition. The energy of the internal conversion line of the 4.300 ± 0.005 Mev transition was measured with great care and this line may be useful for spectrometer calibration. The second excited state of Zn^{66} , at 1.875 Mev, has $2+$ spin and parity. The stopover transition from this state contains at least 10% $M1$ radiation: the stopover to crossover ratio is greater than 100 to 1.

I. INTRODUCTION

THE decay of Ga^{66} to Zn^{66} was investigated in great detail by Mann, Meyerhof, and West.¹ Earlier work, mainly magnetic spectrometer measurements of the positron spectra, was performed by Langer and Moffat² and by Mukerji and Preiswerk.³ The decay of Cu^{66} was studied by Friedlander and Alburger.⁴ Recently, Horen and Meyerhof,⁵ in a new study of the decay of Cu^{66} to Zn^{66} , found a level at 1.865 Mev (the second excited state) which strongly suggested that some of the gamma rays in the decay scheme of Ga^{66} had been misplaced. This conclusion was strengthened by the (p, p') reaction data of Beurtey et al.,⁶ who were able to resolve inelastic proton groups corresponding to excitation of the lowest levels in Zn^{66} .

Magnetic moment measurements of Ga^{66} indicate that its spin is zero,⁷ while shell-model considerations

imply that the parity of this state is positive. The 4.17-Mev ground-state positron spectrum in the Ga^{66} decay is thus expected to be a pure Fermi transition. Measurements of the polarization of these positrons⁸ have figured significantly in the determination of the beta-decay invariants of the Fermi interaction. It therefore seemed valuable to check that the $0+$ assignment to Ga^{66} was in agreement with the Ga^{66} decay scheme, and to determine the purity of the higher energy portion of the positron spectrum.

The second excited state of Zn^{66} at 1.865 Mev, while similar in many respects to the second excited states of Zn^{64} and Zn^{68} , has a very much smaller crossover to stopover transition ratio.⁹ Its behavior was, therefore, studied in some detail.

We have been able to place all but one of the 18 gamma rays we observed into an ordered scheme containing 11 levels and to determine unambiguously the spins and parities of all but one of these levels. The measurements which led to our decay schemes of Ga^{66} and Cu^{66} are described in the following sections and are summarized as follows:

(1) The energies and intensities of the gamma rays fed in the decay of Ga^{66} were determined by single-crystal scintillation spectrometry, by three-crystal pair

[†] Work performed under the auspices of the U. S. Atomic Energy Commission.

* Present address: Massachusetts Institute of Technology, Cambridge, Massachusetts.

¹ L. G. Mann, W. E. Meyerhof, and H. I. West, Jr., *Phys. Rev.* **92**, 1481 (1953).

² L. M. Langer and R. J. D. Moffat, *Phys. Rev.* **80**, 651 (1950).

³ A. Mukerji and P. Preiswerk, *Helv. Phys. Acta* **25**, 387 (1952).

⁴ G. Friedlander and D. E. Alburger, *Phys. Rev.* **84**, 231 (1951).

⁵ D. J. Horen and W. E. Meyerhof, *Phys. Rev.* **111**, 559 (1958).

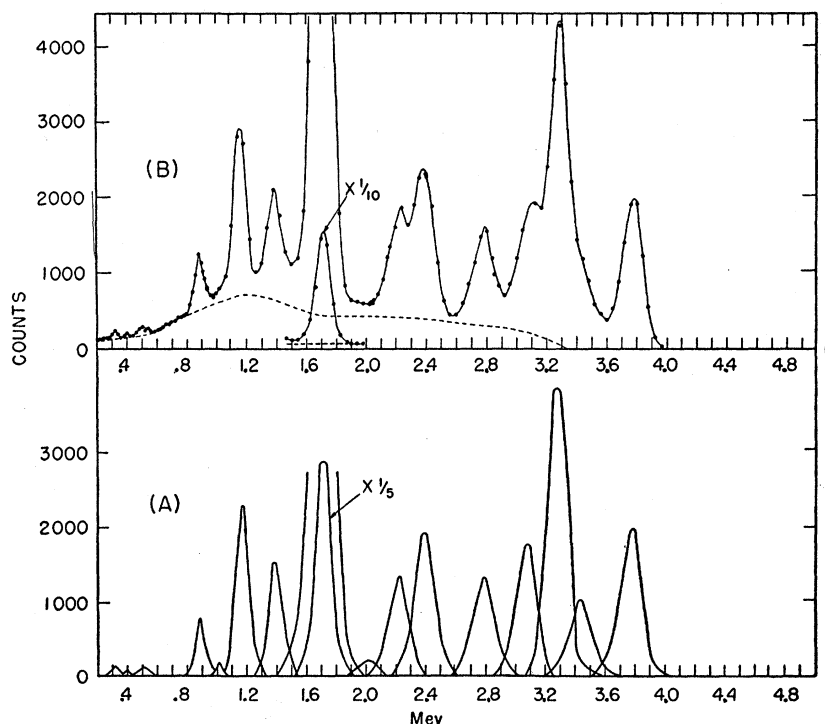
⁶ R. Beurtey, P. Catillon, R. Chaminade, H. Faraggi, A. Papineau, and J. Thirion, *Nuclear Phys.* **13**, 397 (1959).

⁷ J. C. Hubbs, W. A. Nierenberg, H. A. Shugart, and J. L. Worcester, *Phys. Rev.* **105**, 1928 (1957).

⁸ M. Deutsch, B. Gittelman, R. Bauer, L. Grodzins, and A. Sunyar, *Phys. Rev.* **107**, 1733 (1957); S. Frankel, P. Hansen, O. Nathan, and G. Temmer, *Phys. Rev.* **108**, 1099 (1957).

⁹ D. J. Horen, *Phys. Rev.* **113**, 572 (1959).

FIG. 1. Three-crystal pair spectrum of γ rays in the Ga^{66} decay. The curve (A) is the resolution of the experimental curve (B) into its separate peaks. The dotted curve in (B) is the assumed continuous background. The energy scale refers to the pair energy, i.e., $(E_\gamma - 2m_0c^2)$.



spectrometry, and by internal conversion spectrometry using an intermediate image spectrometer.

(2) These gamma rays were ordered into a decay scheme by the requirements placed by their energies and intensities as well as by γ - γ coincidence measurements using both conventional methods (i.e., multi-channel analyzer and single-channel analyzer) and an XYZ coincidence analyzer.¹⁰ The level scheme resulting from these studies was further confirmed by positron-gamma coincidence studies, and, very critically, by a study of the gamma-ray intensities in coincidence with 511-511 kev annihilation radiation.

(3) The spins and parities of the levels were deduced from (a) angular correlation measurements on cascades through the known $2+$ first excited state at 1.037 Mev, (b) the internal conversion and gamma-ray intensity measurements, and (c) from the allowed character of the beta feedings to the levels. In Sec. IX the experimental evidence leading to decay scheme of Fig. 6 is summarized. The final section contains a discussion of the results.

II. SOURCE PREPARATION

All the Ga^{66} activity was prepared by 40-Mev α bombardments of natural Cu targets. For many of the γ -ray measurements the Cu target was allowed to decay for several hours and then used without chemical separation. For the β spectra, conversion electron spectra, and γ - γ correlation measurements, the Ga

activity was separated from the Cu target and from Zn activities produced in the bombardment by the ether extraction process. The Ga activity was separated from the ether solution by back-extracting into water.

Cu^{66} sources were produced by neutron activation of Cu^{65} . Preliminary runs were performed using natural Cu targets and also isotopically separated (90%) Cu^{65} obtained from the Oak Ridge National Laboratory. Because of difficulties with even small amounts of Cu^{64} activity, the final experiments were performed using 99.4% Cu^{65} targets obtained from the Atomic Energy Research Establishment at Harwell.

III. γ -RAY INTENSITIES

A. Three-Crystal Pair Spectra

A single NaI scintillation spectrometer of presently available resolution cannot resolve the complex spectrum of γ rays between 0.8 and 4.8 Mev. For this reason, Mann, Meyerhof, and West¹ investigated the γ -ray spectrum with the use of a three-crystal pair spectrometer using an oscilloscope display of pulse heights. We have repeated this measurement using a 100-channel analyzer in place of their oscilloscope. Our spectrometer was constructed using a 2 in. \times 2 in. NaI center (display) crystal and two 3 in. \times 3 in. NaI side crystals for detection of the annihilation radiation. The pulses displayed on the multichannel analyzer were those in coincidence ($2\tau \sim 10^{-7}$ sec) with 511 ± 25 kev radiations in both side crystals. Energy calibration of the center crystal was determined using the photo-

¹⁰ Lee Grodzins, Rev. Sci. Instr. 26, 1208 (1955).

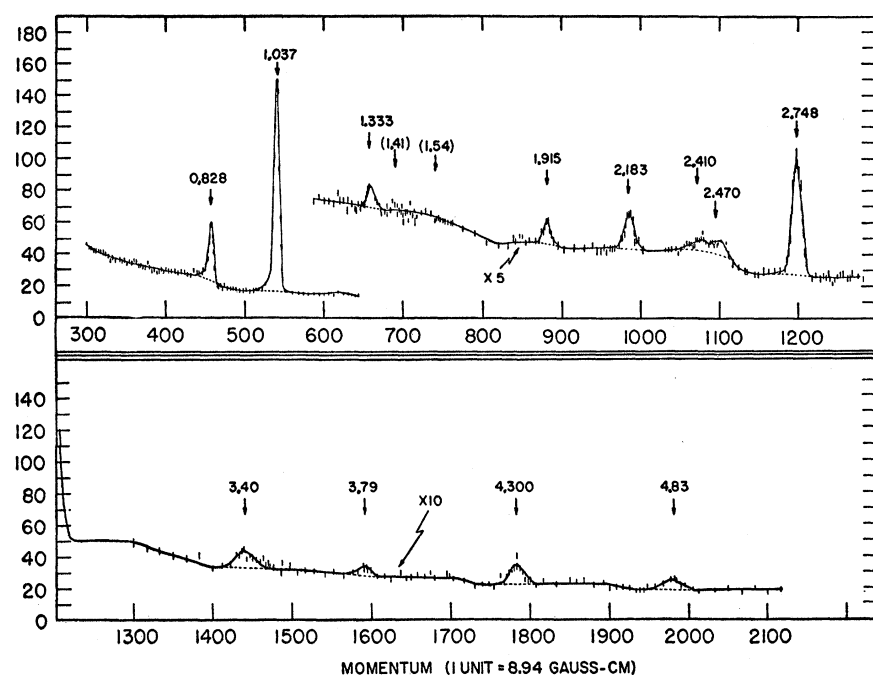


FIG. 2. Internal conversion spectrum. The energies of all observed lines are given in Mev and refer to the corresponding γ -ray transitions. The K and L lines are unresolved in all cases.

peak positions (in the single spectrum of the center crystal) of the 0.511-, 1.037-, and 2.748-Mev peaks. The pair spectrum is shown in Fig. 1(B). A continuous background indicated in this figure, due to brems-

strahlung escape, is subtracted before analysis of the spectrum into the separate peaks as indicated in Fig. 1(A). The relative intensities of the γ rays were obtained using the relative pair efficiency curves given by Mann et al.,¹ and are given in the second column of Table I.

TABLE I. Gamma-ray energies and intensities.

E_γ (Mev)	Measured relative γ -ray intensities ^a	Intensities adjusted for conversion data ^b (percentage per ground state β^+)	Intensities adopted in decay scheme (percent per decay Ga^{66})	Intensities ^c by Mann et al.
0.828 ± 0.002^d	12 ± 2	12 ± 1	5.4	2.6
1.037 ± 0.002^d	76 ± 7	81 ± 5	37	36
1.333 ± 0.003^d	4.1 ± 1.1	3.0 ± 0.3	1.2	3.5
1.41 ± 0.03^e	1.4 ± 0.8	1.4 ± 0.8	0.6	
1.54 ± 0.04^e	2.1 ± 0.5	2.1 ± 0.5	1.0	
1.915 ± 0.010^d	5.4 ± 0.7	6.0 ± 1.0	2.5	4.2
2.183 ± 0.004^d	13.1 ± 1.2	12 ± 1	5.0	6.9
$2.410 \pm 0.020^d, f$	7.2 ± 0.8	3.5 ± 1.0	2	2.8
$2.470 \pm 0.020^d, g$		4.5 ± 1.0	2	
2.748 ± 0.004^d	60	60 ± 6	25.5	26
$3.03 \pm 0.05^e, g$	0.7 ± 0.4	0.7 ± 0.4	0.3	
$3.24 \pm 0.04^e, h$	4.8 ± 0.9	4.8 ± 0.9	2.1	2.3
$3.400 \pm 0.20^d, i$	6.8 ± 1.1	5.7 ± 1.0	1.0 ± 1.6^j	3.4
3.790 ± 0.030^d	4.3 ± 0.6	4.1 ± 0.6	2.0	2.1
4.10 ± 0.04^e	5.0 ± 1.8	5.0 ± 1.8	2.2	1.8
4.300 ± 0.005^d	10.8 ± 2.2	10.8 ± 2.2	4.8	5.3
4.45 ± 0.06^e	2.9 ± 0.8	2.9 ± 0.8	1.3	
4.833 ± 0.030^d	5.4 ± 0.4	5.4 ± 0.4	2.4	2.5
Annihilation (0.511)	233 ± 28	229 ± 10	101	

^a Intensities normalized to an arbitrary value of 60 for the 2.748-Mev γ . (Sixty percent is the approximate ratio of 2.748-Mev γ rays per ground-state position transition.) All intensities are derived from three-crystal pair spectrometer data except those of the 0.511-, 0.828-, and 1.037-Mev transitions whose intensities are derived from singles and coincidence spectra as described.

^b Adjustments made on γ rays whose internal conversion intensity measurement is more precise than the γ -ray intensity measurement. The gamma intensities are computed from conversion intensity using assuming $E2-M1$ multipolarity.

^c Normalized to 26 for 2.748-Mev γ ray.

^d Energy determined from internal conversion measurements.

^e Energy determined from 3-crystal pair spectrometer measurements.

^f Energy given as 2.43 Mev in decay scheme.

^g Gamma rays not shown in decay scheme.

^h Energy given as 3.22 Mev in decay scheme.

ⁱ Double line—shown as two transitions in decay scheme.

B. Intensity of γ Rays Below 1.2 Mev

The only γ rays observed are at 0.511, 0.83, and 1.037 Mev. The intensities of the 0.511- and 1.037-Mev γ rays relative to the 2.75-Mev γ ray can be obtained from the singles γ -ray spectrum in a 3 in. \times 3 in. NaI spectrometer. We have been able to obtain the relative intensity of the weak 0.83-Mev γ ray only by comparison of the 0.83-Mev and 2.75-Mev γ rays in coincidence with 1.037 Mev.¹¹ Since we believe both of these γ rays are completely in coincidence with the 1.037 γ ray, this measurement seems justified. The results are given in the second column of Table I.

IV. INTERNAL CONVERSION

Measurements of internal conversion electrons from Ga^{66} were performed using an intermediate image magnetic lens spectrometer.¹² The spectrometer was fitted with spiral baffles to separate electrons and positrons,¹³ and the resolution was adjusted to $\sim 1.5\%$ yielding a transmission of $\sim 2\%$ of 4π . Sources of ~ 3 mC of Ga^{66} were prepared by evaporation of a water

¹¹ These coincidence measurements have been corrected for the angular correlation between 1.037- and 1.87-Mev γ rays and the 1.037- and 0.83-Mev γ rays.

¹² D. E. Alburger, Rev. Sci. Instr. **27**, 991 (1956).

¹³ D. E. Alburger, S. Ofer, and M. Goldhaber, Phys. Rev. **112**, 1998 (1958).

drop containing the activity on 1 mg/cm² Mylar film resulting in sources estimated to be less than 1 mg/cm² in thickness. Three separate runs of the conversion spectra were made one of which is shown in Fig. 2. This curve has been corrected for both room background and source decay. The remaining background is due to Compton electrons produced in the source itself and in the spectrometer baffles and to a small component of positrons scattered through the spiral baffles. The rather abrupt breaks in the background correspond to the Compton edges of the intense γ rays. The K and L conversion lines are unresolved in all cases. The energies of γ rays corresponding to the conversion lines are given in Fig. 2 and in Table I. The spectrometer was calibrated with the 0.976-Mev conversion line of Bi^{207} . Particular care was taken in the measurement of the energy of the 4.3-Mev conversion line energy so that it might be used as a calibration for spectrometers containing iron.

The β^+ spectrum of one of the sources was measured several days after the conversion measurements. At

TABLE II. Internal conversion coefficients.

E_γ (Mev)	$\frac{e(K+L)}{\beta^+(4.16 \text{ Mev})} \times 10^4$		$\frac{\gamma}{\beta^+(4.16 \text{ Mev})} \times 10^2$		$(\alpha_K + \alpha_L) \times 10^4$
0.828	0.52	$\pm 10\%$	12		4.3 ± 0.6
1.037	2.08	$\pm 5\%$	76		2.74 ± 0.36
1.333	0.045	$\pm 10\%$	4.1		1.1 ± 0.4
1.915	0.057	$\pm 25\%$	5.4		1.05 ± 0.35
2.183	0.064	$\pm 11\%$	13.1		0.49 ± 0.08
2.410	0.020	$\pm 55\%$	} 7.2 ^a		0.65 $\pm 40^a$
2.470	0.027	$\pm 40\%$			
2.748	0.227	$\pm 8\%$	60		0.38 ± 0.05
3.400 ^b	0.014	$\pm 16\%$	6.8 ^a		0.21 $\pm 0.04^a$
3.786	0.0081	$\pm 35\%$	4.3		0.19 ± 0.08
4.300	0.020	$\pm 23\%$	10.8		0.185 ± 0.060
4.823	0.0095	$\pm 25\%$	5.4		0.175 ± 0.050

^a Unresolved γ -ray intensity. Conversion coefficient is just sum of electron intensities divided by unresolved γ intensity.

^b Probably double.

that time the 1.037-Mev conversion line was still quite easily observed, thus enabling us to check the source decay over about seven half-lives. Averages of the conversion line intensity from the three runs relative to the β^+ intensity in the ground-state β^+ transition (4.166 Mev) assumed to be of allowed shape are given in Table II. The γ -ray intensities relative to total β^+ emission is determined from the relative intensity of γ rays to annihilation radiation. If we assume, in accordance with our final decay scheme of Fig. 6 that 88% of the β^+ decays are in the 4.166-Mev group, then the numerical values for the γ intensities in the second column of Table I are the measured γ intensities per 100 ground state β^+ . From these intensity measurements and the measured conversion line intensities, we obtain the conversion coefficients given in Table II, and plotted in Fig. 3. The theoretical $K+L$ conversion coefficients are obtained by graphical extrapolation to

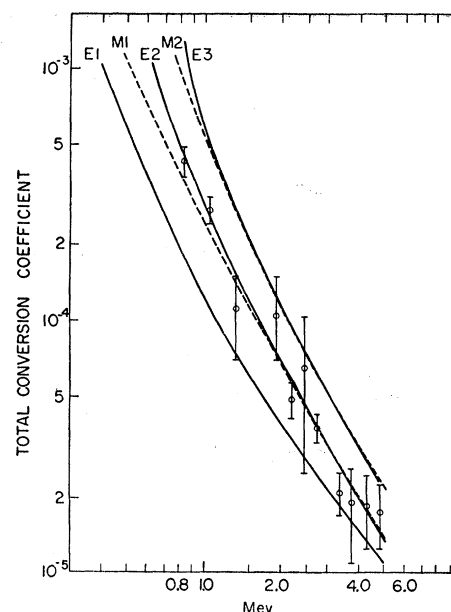


FIG. 3. Theoretical total conversion coefficients for different multipolarities in Zn as a function of energy. The points indicate the measured coefficients of the observed transitions (see Table II).

high energies of the lower energy coefficients of Sliv.¹⁴ The errors indicated in the figure and table are relative errors and do not reflect any of the uncertainty in the determination of the β^+ branching. However, the excellent agreement between the measured and theoretical conversion coefficient for the 1.037-Mev γ ray, which is undoubtedly $E2$, confirms the β^+ branching ratio to the ground state.

We conclude that the conversion coefficients of the 1.037-, 0.83-, and 2.748-Mev transitions are indicative of $E2$ or $M1$ character and that those of all the other measured transitions are consistent with $E2$ or $M1$ character (or mixture). Also, the theoretical $M1$ and $E2$ conversion coefficients differ only slightly from one another and are well known. We have, therefore, adjusted some of the relative γ intensities to those obtained from the conversion data and the theoretical conversion coefficients assuming $E2$ or $M1$ multipolarity, since the relative intensities of the stronger conversion lines can be measured more accurately than the corresponding γ rays. The adjusted intensities are given in the third column of Table I.

A careful search for conversion electron lines between 0.30-Mev and 0.82-Mev was performed. The absence of any conversion line indicates that no gamma-ray transition in this energy region can occur in the Ga^{66} decay with more than 1% intensity even if its internal

¹⁴ L. A. Sliv and J. M. Band, Tables of Gamma-Ray Internal Conversion Coefficients, Leningrad Physico-Technical Institute Report, 1956 [translation: Report 571CC K1, issued by Physics Department, University of Illinois, Urbana, Illinois, (unpublished)].

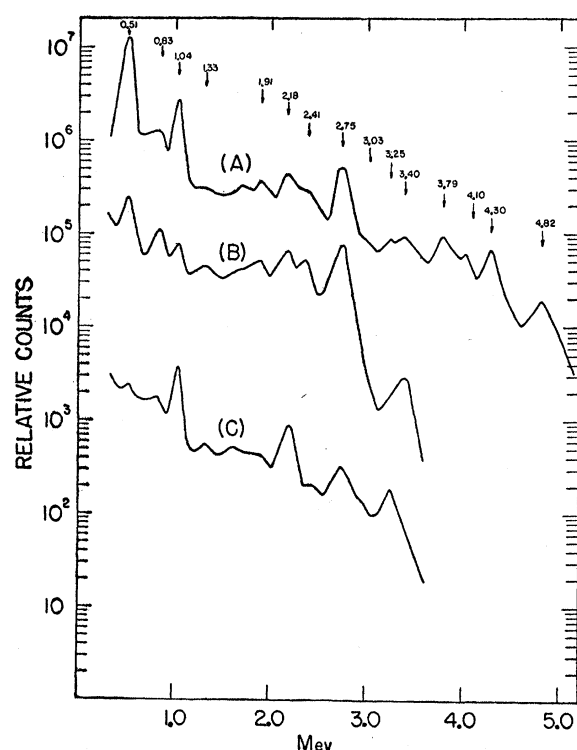


FIG. 4. NaI pulse-height spectra. (A) Singles observed in a 3 in. \times 3 in. crystal, (B) γ rays in coincidence with the 1.04-Mev γ ray, (C) γ rays in triple coincidence with two annihilation quanta from the source. The arrows indicate the expected positions of peaks corresponding to γ rays observed in the three-crystal pair spectrum and the conversion electron spectrum.

conversion coefficient were as low as that of an $E1$ transition.

V. COINCIDENCE EXPERIMENTS

The singles NaI spectrum of Ga^{66} γ rays obtained with a 3 in. \times 3 in. crystal is shown in Fig. 4(A). It is

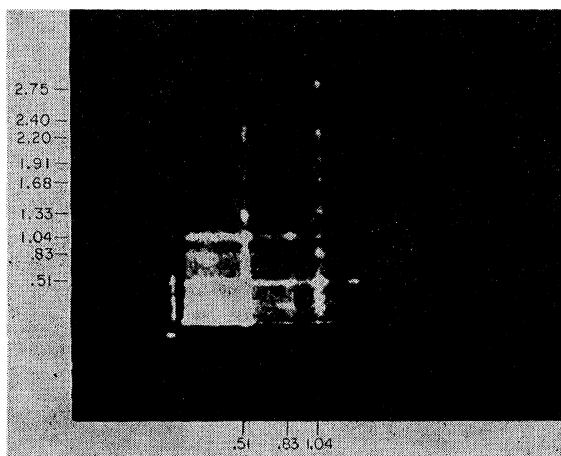


FIG. 5. X-Y-Z analyzer spectrum of Ga^{66} coincidences. The camera field cuts off slightly above 1 Mev in the horizontal direction leading to the asymmetry of the display.

clear that the complexity of this spectrum precludes the possibility of quantitative coincidence spectroscopy except for the most intense γ rays.

A. Gamma-Gamma Coincidence

Gamma rays in coincidence with the 1.037-Mev transitions were measured with conventional fast-slow coincidence circuitry employing two 3 in. \times 3 in. NaI detectors and a 100-channel analyzer for the display. The spectrum of coincidences is shown in Fig. 4(B). A list of the coincident γ rays and some of their relative intensities are given in Table III(A). It was not considered possible to obtain more quantitative data than is given in the table.

TABLE III. Results of coincidence experiments.

(A) γ rays in coincidence with 1.037 Mev: γ (Fig. 4)	
(a)	0.83, 1.30, 2.90, 2.18, 2.40, 2.75, 3.4 Mev γ .
(b)	$I_{0.83}/I_{2.75} = 0.20 (\pm 10\%)$ corrected for angular correlation).
(c)	$I_{2.75}/I_{3.4} \sim$ twice ratio in singles.
(B) Coincidence sorter: (Fig. 5)	
(a)	in coincidence with 0.51: 0.83, 1.04, 1.30, (1.68), 2.18, 2.75, 3.24, 3.79
(b)	in coincidence with 0.83: 0.51, 1.04, 1.90, 2.40
(c)	in coincidence with 1.05: 0.51, (0.68), 0.83, 1.30, (1.68), 1.91, 2.18, (2.40), 2.75, 3.40
(d)	in coincidence with 1.30: very weak 1.4
(e)	except for very weak 1.4-1.30 coincidence no γ ray of energy > 1.1 Mev in coincidence with any γ ray of energy > 1.1 Mev.
(C) Coincidences between γ rays and 2 annihilation quanta: (Fig. 4)	
(a)	relative intensities in coincidence: $I_{1.04}:I_{1.33}:I_{2.18}:I_{2.75}:I_{3.25} = 1.1:0.08:0.6:0.25:0.26$ (intensities expected from decay scheme: 1.1:0.12:0.65:0.23:0.26.)
(D) Coincidences between γ rays and β^+	
β channel	Most intense γ rays
$0.22 < E_\beta < 0.62$	1.04, 1.33, 2.18, 3.24, 2.75
$0.6 < E_\beta < 1.2$	1.04, 1.33, 2.18
$1.3 < E_\beta < 2.2$	1.04, 1.33, (2.18)

The X-Y-Z coincidence sorter is described in detail by Grodzins.¹⁰ As used, the oscilloscope X and Y deflections correspond to pulse-height amplitude in two 3 in. \times 3 in. NaI crystals, respectively. The oscilloscope spot is intensified only if a coincidence occurs between the two detectors. The detectors were shielded from one another by a $\sim 1\frac{1}{2}$ -in. Pb absorber and each crystal was shielded from the source by a 3 g/cm² Pb absorber. Photographic exposures of the scope screen were made, one of which is shown in Fig. 5. The results of these measurements are given in Table III(B). Some of the coincidences recorded in Table III(B) are enclosed in parentheses since they probably correspond to escape peaks of higher energy γ rays. It should be noted that even in very long photographic exposures no coincidences were observed between two γ rays of energy

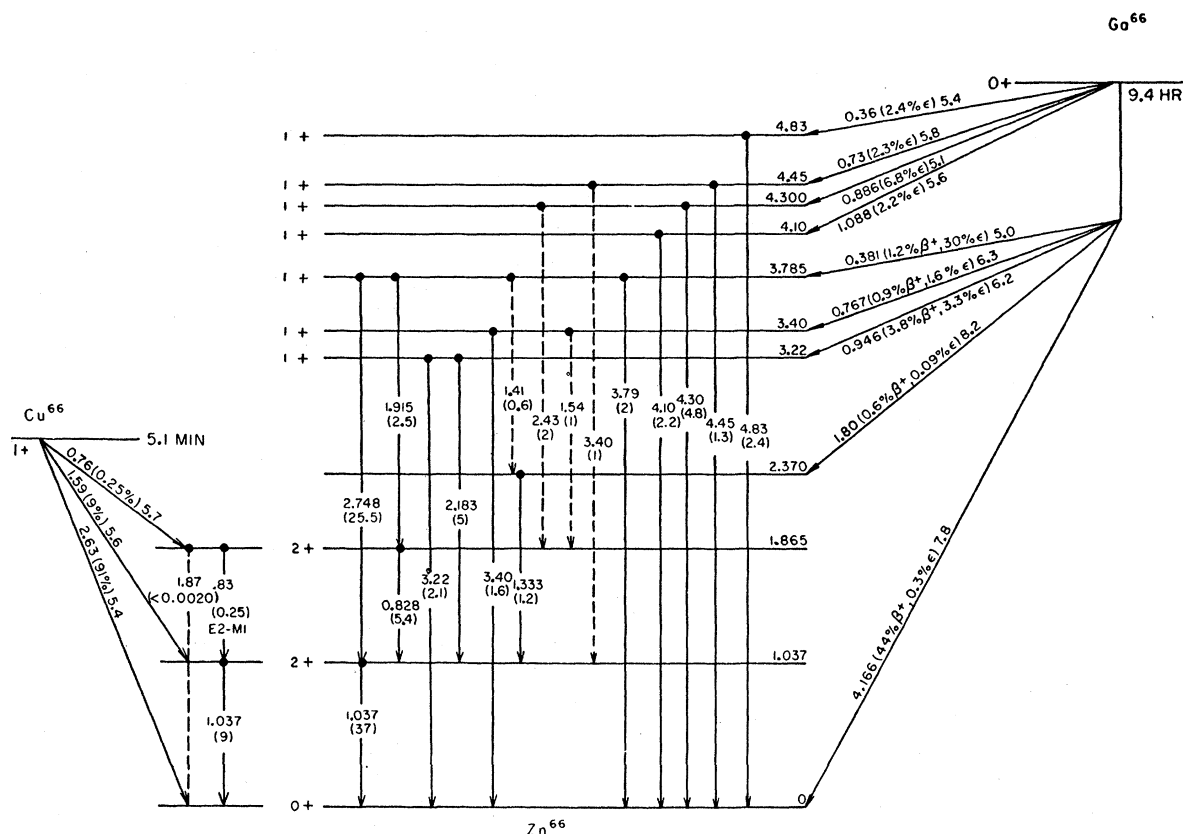


FIG. 6. Proposed decay scheme of Ga^{66} and Cu^{66} . All intensities, given in parentheses, refer to percentage of decays of the parent activity. $\text{Log } ft$ values for the β -decay transitions are indicated.

greater than 1.1 Mev except for the weak 1.3–1.4 coincidence.

The knowledge of the precise energies of many of the γ rays of Ga^{66} , the available information on the levels of Zn^{66} from the decay of Cu^{66} and the qualitative results of the coincidence sorter experiments provide sufficient information to determine most of the level scheme of Zn^{66} as given in Fig. 6. Further confirmation of this level scheme was obtained from the coincidence experiments described below.

B. γ Rays in Coincidence with Annihilation Radiation

The spectrum of γ rays in coincidence with two annihilation quanta, indicating β^+ decay in the source, is a sensitive test of some of the features of the assumed decay scheme since the K capture to β^+ ratios to particular states depends critically on the β -decay energies. The coincidence experiment was performed using three 3 in. \times 3 in. NaI crystals. The resulting spectrum is shown in Fig. 4(c). The measured relative intensities of γ rays in coincidence with positrons as deduced from this spectrum are given in Table III(C) and are in good agreement with the values expected from the decay scheme.

It should be noted that this experiment fixes an

upper limit of 1% for the ground-state transition from the 2.37-Mev state to be compared to the 1.2% intensity of the 1.33-Mev cascade transition from this state.

C. Positron-Gamma Coincidences

The thick anthracene crystal necessary for this experiment has, unfortunately, an appreciable efficiency for gamma detection. Gamma-gamma coincidences accounted for about one-half the measured rate when the anthracene crystal pulses were selected for high β^+ energies. First order measurement of this effect was done by interposing an absorber for β^+ rays. The qualitative information obtained is listed in Table III and is in agreement with the decay scheme.

VI. POSITRON SPECTRUM

The positron spectrum from the decay of Ga^{66} was measured with the intermediate image spectrometer. The observed spectrum is shown in Fig. 7. Kurie analysis of the upper group by fitting in the region $5 < p < 9m_{0c}$ yields a straight Kurie plot with end point of 4.166 ± 0.030 Mev. All the points below 1.8 Mev¹⁵

¹⁵ The source used for these measurements had decayed for more than 48 hours thus assuring that the 1.8-Mev β^+ spectrum of 1-hr Ga^{66} was no longer present.

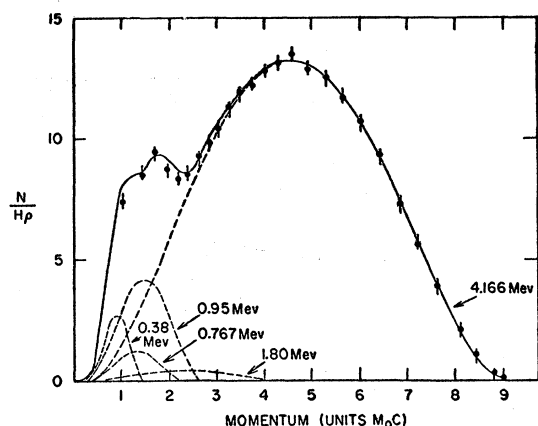


FIG. 7. Positron spectrum of Ga^{66} observed with intermediate image spectrometer. The solid curve is the calculated sum of the separate groups with intensities and end points indicated in the decay scheme.

lie above the straight line extrapolation of the high-energy Kurie plot. It is, however, clear that more than 75% of the total β^+ intensity belongs to the ground-state transition.

Considerations of the precision of β -spectral measurements¹⁶ indicate that one cannot generally expect a single group allowed β spectrum to yield a Kurie plot linear to better than several percent at $\sim \frac{1}{5}$ of the maximum energy unless extreme care is taken with the source and spectrometer. Since small and unknown instrumental deviations from the expected allowed shape of the intense high-energy branch have a serious effect upon the apparent intensities and end points of the lower energy groups, it is not considered particularly significant to perform Kurie analysis with successive subtractions in order to analyze the Ga^{66} β^+ spectrum.

The solid curve drawn in Fig. 7 is a theoretical spectrum determined by adding all the β groups with end points and relative intensities as given in the decay scheme and is normalized to the total measured spectrum intensity. The agreement between the measured spectrum and the curve is probably within the possible systematic errors of the measurement.

In the decay scheme of Fig. 6 the intensities of the β^+ group are determined from the γ -ray and internal conversion data, in such a manner as to account for the measured internal conversion intensity of the 1.037-Mev transition relative to the intensity of the ground-state positron branch and for the intensities of the other γ rays. All the capture to positron branching ratios are determined theoretically¹⁷ from the values of K to β^+

ratios of Zweifel as corrected by Perlman and Wolfsberg and a value of $\sim 10\%$ for the K to L capture ratio.

Table IV gives the β group intensities as determined from the decay scheme, together with a comparison with the results of Mukerji and Preiswerk,³ and Langer and Moffat.² The different intensities of the inner groups as determined by these authors may well be due to systematic errors in the measurement (scattering effect, etc.) and in the analysis. It is, however, interesting and gratifying to point out that the ratio of the sum of the intensities of the inner groups to the ground-state group intensity is $\sim 15\%$ in the work of these authors and in our decay scheme.

VII. DECAY OF Cu^{66}

During the course of our investigation of this decay, the detailed work of Horen and Meyerhof⁵ was reported. Our results are in agreement with the decay scheme they propose. We shall discuss in detail only those measurements which extend their work.

The intensity ratio between the 0.83-Mev γ ray and the 1.037-Mev γ ray was measured by comparing the 1.037-Mev intensity with the intensity of the (true coincident) sum peak at 1.87 Mev as obtained with a single 3 in. \times 3 in. NaI detector. These measurements indicated a relative intensity of 0.028 ± 0.003 for the ratio of 0.83- to 1.037-Mev γ -ray intensities. This method is reliable only because the 1.87-Mev crossover is extremely weak.

An attempt was made to observe the very weak crossover transition. Isotopically pure Cu^{66} , bombarded for several minutes in the Brookhaven reactor to produce ~ 1.5 mC of the Cu^{66} activity, was placed in a Lucite container thick enough to stop all the electrons. An absorber consisting of 1.5 inches of Pb and $\frac{3}{8}$ -in. of Cu was placed between the source and the 3 in. \times 3 in. NaI crystal in order to reduce the pile-up of bremsstrahlung pulses, to enhance the crossover relative to the intense 1.037-Mev γ ray, and to suppress the 1.037-0.83 Mev sum peaks. Figure 8 shows the spectrum

TABLE IV. β decay intensities.

Zn^{66} state fed	Positron energy	Our results			Mukerji/Prieswerk ^a and Langer/Moffat ^b	
		Percent intensity per decay Ga^{66}	$K+L$ capture	$\log_{10} f t$	Positron energy	Intensity normalized to 44% for ground-state transition
0	4.166	44	0.3	7.8	4.15	44
2.37	1.80	0.6	0.09	8.2
2.75?		<1.0			1.38	2.1
3.22	0.946	3.8	3.3	6.2	0.90	3.5
3.40	0.767	0.9	1.6	6.3
3.785	0.381	1.2	30	5.0	0.40	0.91
	Electron capture energy					
4.10	1.088		2.2	5.6		
4.300	0.886		6.8	5.1		
4.45	0.73		2.3	5.8		
4.83	0.36		2.4	5.4		

^a See reference 3.

^b See reference 2.

¹⁶ A. Schwarzschild, thesis, Pupin Cyclotron Laboratory Report CU-167, AT30-1-GEN-72, Columbia University, 1957 (unpublished).

¹⁷ M. L. Perlman and M. Wolfsberg, Brookhaven National Laboratory Report No. 485 (T110) (unpublished); P. F. Zweifel, Phys. Rev. **107**, 329 (1957); A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

obtained in this manner. The background pulses obtained in the high-energy region of Fig. 8 are presumably due to summing of the intense bremsstrahlung and the 1.037-Mev γ ray and to high-energy bremsstrahlung. The 1.87-Mev transition was not observed; an upper limit of 2.2×10^{-4} per 1.037-Mev γ -ray for the crossover intensity, corresponding to a limit of 0.8×10^{-2} for the ratio of the ground state to cascade intensity in the decay of the 1.865-Mev level is deduced.

VIII. ANGULAR CORRELATION MEASUREMENTS USING Ga^{66} AND Cu^{66} SOURCES

The angular correlation measurements were performed using two 3 in. \times 3 in. NaI detectors each placed at about 15 cm from the source. The detectors were unshielded except for an absorber of ~ 2.6 g/cm² Pb over the face of each crystal and a μ -metal shield surrounding the motor driven movable detector. The electronics consisted of a fast-slow coincidence system employing a 6BN6 coincidence tube and having a fast resolving time of $2\tau \sim 2 \times 10^{-8}$ sec.

A. 1.04–2.75 Mev γ Rays in the Ga^{66} Decay

The angular correlation of the 1.04–2.75 Mev cascade is shown in Fig. 9. The sources used were surrounded by ~ 2 g/cm² brass to stop electrons. The decay scheme of Ga^{66} as well as the coincidence curve of Fig. 4(B) indicate that there is $< 5\%$ contribution to the counting rate in coincidence from other cascades. Corrections of about 6% for these other contributions including a 3%

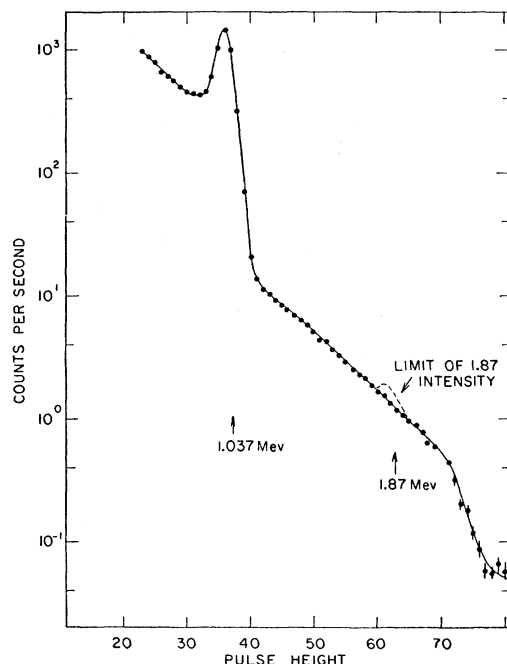


FIG. 8. NaI scintillation spectrum of Cu^{66} γ rays. The dashed curve at 1.87 Mev corresponds to the intensity limit given in the decay scheme for a transition of this energy.

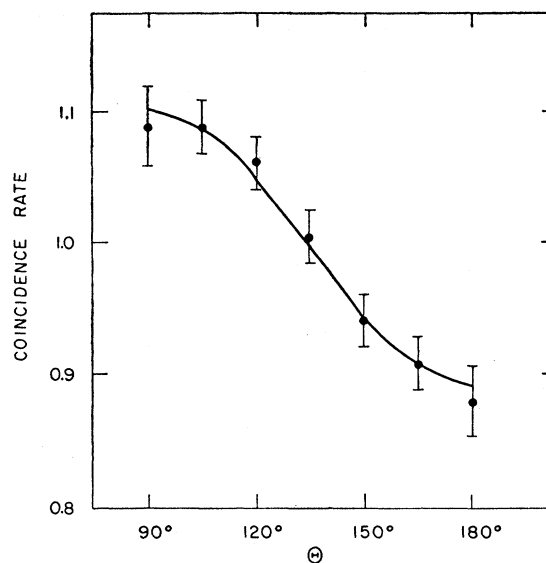


FIG. 9. Angular correlation between 1.04- and 2.75-Mev γ rays in the decay of Ga^{66} .

random coincidence rate have been made to the data of Fig. 9. A least squares fit according to the method of Rose¹⁸ yields the correlation

$$N'(\theta) = (1.03 \pm 0.01) - (0.14 \pm 0.03)P_2(\cos\theta).$$

A curve of this function is drawn in Fig. 9. The correlation is consistent with no $P_4(\cos\theta)$ term. Correction of the correlation function for the finite solid angles of the detectors yield the correlation function

$$W(\theta) = 1 - (0.15 \pm 0.03)P_2(\cos\theta).$$

This correlation is consistent with that expected for a 1–2–0 cascade with a mixing ratio of quadrupole to dipole intensities $0.01 < \delta^2 < 0.1$ (δ negative) in the $1 \rightarrow 2$ transition. These mixing ratios would give a small $P_4(\cos\theta)$ term in the correlation which would, however, not have been detected.

B. 0.83–1.04 Mev γ - γ Correlation in the Decay of Cu^{66}

Only two nuclear γ rays of 0.83 and 1.04 Mev are present in the decay of Cu^{66} . However, the 5-minute half-life of the Cu^{66} activity, the low intensities of the 0.83-Mev γ rays per decay, and the large intensity ratio of the two γ rays create special problems for the γ - γ correlation measurement.

In order to obtain a sufficient number of coincidences from each irradiated sample, the intensity of the source had to be extremely high at the beginning of each run. Sources of ~ 0.5 mC resulting in counting rates of $\sim 5 \times 10^3$ cps in the 1.04-Mev photopeak at the beginning of the measurement, yielded a coincidence rate of ~ 0.8 cps one quarter of which were accidentals. The

¹⁸ M. E. Rose, Phys. Rev. **91**, 610 (1953).

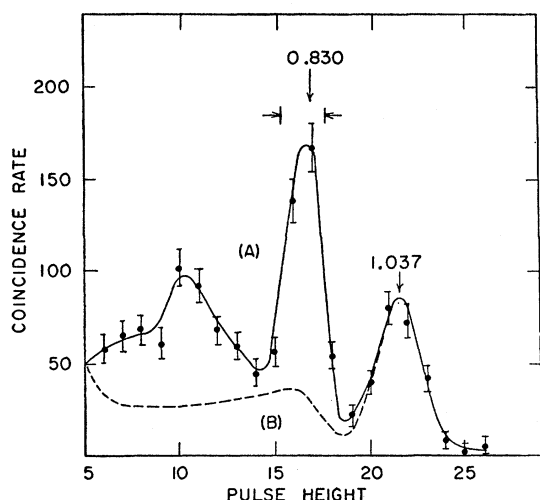


FIG. 10. Pulse-height spectrum of γ rays in coincidence with the 1.037-Mev γ ray in the decay of Cu^{66} . The solid curve (A) is the observed spectrum and the dashed curve (B) is the inferred accidental coincidence contribution. The horizontal arrows refer to the edges of the channel used for the 0.83-Mev detector in the angular correlation experiment.

high singles counting rate and rapid decay produces large gain shifts in the DuMont 6363 photomultiplier, so that it was necessary to use gain stabilizing circuits of the type described by DeWaard¹⁹ on each of the detectors. The stabilizers changed the effective phototube high voltage by about 100 volts (out of 1500 v) during the decay of a single source in order to maintain constant gain. Tests showed that the effect of this change in phototube voltage on the fast coincidence efficiency was negligible.

The detector angle was changed every minute for about 4 half-lives of each source. Accidental coincidences had to be subtracted individually from each 1-minute count. The starting angular position was changed for each separate source. Preliminary runs with 90% separated Cu^{65} showed that a large contribution of 0.511-0.511 Mev coincidences at 180° due to the Cu^{64} activity were piling up into the 0.83- and 1.04-Mev channels giving an anomalously high value of the correlation at 180° . For this reason the highly separated Cu^{66} (99.4%) targets were used for the final neutron bombardments.

Figure 10 shows a spectrum of γ rays in coincidence with the 1.04-Mev γ ray. Beside the 0.83-Mev peak, some 1.04-Mev γ rays are detected which correspond to accidental coincidences. Contributing to the true coincidence rate are some inner and outer bremsstrahlung-1.04 Mev coincidences and some pile-up effects of the annihilation radiation from the small Cu^{64} activity. However, in the region of the 0.83-Mev channel used in the correlation experiment, indicated in Fig. 10, no significant contribution which might distort the correlation other than accidental coin-

cidences are present in the coincidence spectrum. It should be noted that the efficiency for detecting annihilation coincidences at our 165° position is minute.

The correlation measurements were performed with 25 separate sources. Coincidence rates at each angle were normalized to the singles rate of the 1.04-Mev radiation which was detected in the movable counter, in order to correct for decay of the source and possible small geometric misalignment. The results of this measurement are shown in Fig. 11. This correlation was fit by the least squares procedure of Rose¹⁸ to the function

$$N'(\theta) = (0.976 \pm 0.019) + (0.236 \pm 0.037)P_2(\cos\theta) + (0.218 \pm 0.041)P_4(\cos\theta).$$

After correction for the finite solid angle of the detectors the correlation function becomes

$$W(\theta) = 1 + (0.26 \pm 0.05)P_2(\cos\theta) + (0.29 \pm 0.06)P_4(\cos\theta).$$

In view of the allowed $\log ft$ for the β decay to the 1.87-Mev level in the Cu^{66} decay, the possible spins for this level are 0, 1, or 2. The observed correlation doesn't fit any of the possible combinations of spin and multipolarity mixture except $2-2-0$ where the $2 \rightarrow 2$ transition is a mixture of dipole and quadrupole radiations. The mixing ratio of the quadrupole to dipole intensities lies between the limits $2.4 < \delta^2 < 9.0$ and the sign of δ is positive. The parity of the 1.87-Mev level is obviously positive from the β -decay data, therefore the 0.83-Mev transition is of mixed $E2-M1$ character.

IX. SUMMARY OF EXPERIMENTAL RESULTS

The following is a summary of the experimental evidence leading to the decay scheme in Fig. 6.

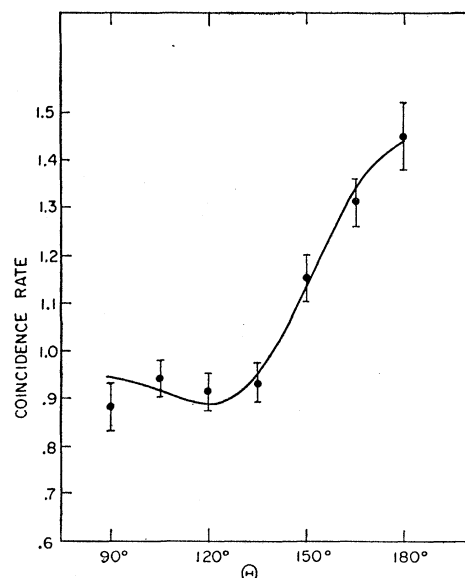


FIG. 11. Angular correlation between the 1.04- and 0.83-Mev γ rays in the Cu^{66} decay.

¹⁹ H. DeWaard, *Nucleonics* 13, No. 7, 36 (1955).

A. Level Structure

The existence of the first two levels of Zn^{66} at 1.037 and 1.865 Mev is clearly implied by the decay of Cu^{66} ; the first level has been observed in Coulomb excitation studies by several workers. The 2.370-Mev level is simply inferred from the evidence that the 1.333-Mev transition is in coincidence with the 1.037-Mev gamma ray, and is also in coincidence with positrons of energy greater than 1 Mev, and at least half its intensity is in coincidence with annihilation radiation. The 2.183-Mev gamma ray is in coincidence with the 1.037-Mev transition and also with annihilation radiation, thus requiring a level at 3.22 Mev. The existence of the 3.40-Mev level is implied by the large fraction of the 3.4-Mev radiation not in coincidence with gamma rays. The precise energy determination of the 2.748-, 1.037-, 0.828-, 1.915-, and 3.79-Mev gamma rays, as well as the observed coincidences between members of this group, assures the cascade relations between them and fixes the 3.785-Mev level. The upper states at 4.1, 4.300, 4.45, and 4.83 Mev are attested to by the ground-state transitions from these levels.

B. Intensity of Beta Groups

The intensity of the ground-state beta transition relative to the 1.037-Mev gamma rays is determined from the observed intensity ratios of internal conversion of this gamma ray to the ground-state positron group and the assumed theoretical $E2$ conversion coefficient. The intensities of the other positron groups are deduced from the required feeding of the various levels to account for the gamma-ray intensities. These intensities are strongly corroborated by the shape of the measured positron spectrum as well as the ratio of total annihilation radiation to gamma-ray intensities and the γ -511-511 coincidence experiments. All of the capture to positron ratios given in the decay scheme are theoretically deduced.

C. Spins and Parities

The zero spin of Ga^{66} is known from magnetic moment measurements and the unit spin of Cu^{66} has been inferred from the electron branching to the first two states of Zn^{66} . The $2+$ spin-parity of the 1.865-Mev level is determined from our angular correlation measurements together with the allowed character of the electron decay from Cu^{66} to this level. The unit spin of the 3.785-Mev state is deduced from angular correlation measurements. The even parity of this state is determined by the $E2$ - $M1$ character of the conversion coefficient of the 2.748-Mev transition. The even parity of Ga^{66} is thus assured since it decays via an allowed beta transition to the 3.785-Mev level. All of the other observed states of Zn^{66} , except the one at 2.370 Mev, must then have spin 1 and positive parity since they are fed by allowed positron or K -capture decay from Ga^{66} and all decay directly to the $0+$

ground state of Zn^{66} . The spin and parity of the level at 2.370 Mev is not determined by this work. The $\log ft$ of 8.2 for the positron transition to this state as well as the apparent absence of any ground-state transition are consistent with either $0+$ or $2-$ assignments—the $2+$ assignment that might be given in analogy to the similar state in Zn^{68} is excluded by the $\log ft$ value of 8.2.

D. Comparison with Other Investigations

With the possible exception of a level at 2.75 Mev, the level structure presented in Fig. 5 is in accord with that given by Mann et al.,¹ as corrected by Horen and Meyerhof, though the relative intensities we obtained for the gamma rays as well as the beta-decay branching ratios differ from theirs. (The precise energy determinations of a number of the gamma rays definitely exclude some of the cascades they suggest.) Moreover, the positron spectrum observed by Mukerji and Preiswerk is probably not in disagreement with the decay scheme presented although their analysis of the spectrum differs from ours. The existence of a 2.75-Mev level suggested by Mukerji and Preiswerk³ and seen in (pp') reaction (at 2.87 ± 0.05) by Beurtey et al., is not observed though, of course, it is not excluded by this work. The level is not populated by more than 1% of the Ga^{66} decays,²⁰ corresponding to less than $\frac{1}{3}$ that suggested by other workers.

IX. CONCLUSIONS AND DISCUSSION

1. The parity of Ga^{66} is even and the Fermi type positron spectrum, above 2 Mev, contains less than 1% of other beta groups.
2. The internal conversion line of the 4.300-Mev gamma ray has been observed and may be useful for higher energy calibration of beta-ray spectrometers.
3. With the possible exception of the 2.370 level, all states above 2 Mev are shown to have even parity and unit spin.
4. The 2.370 level is, most probably, a $0+$ or $2-$ state.
5. The second excited state at 1.865 Mev does not appear to fit well into any present theory of vibrational or rotational excitations; though systematics of even-even nuclei strongly suggest that it is a collective state. A large effort was made to understand the nature of this level. The angular correlation measurements on the de-excitation gamma rays, together with the fact that the beta transition from Cu^{66} to this level is allowed, fixes its spin parity as $2+$. The crossover to

²⁰ This experimental limit is obtained as follows. The intensity of the 2.75 peak as observed in the γ -511-511 coincidence spectrum allows us to set a limit $<0.5\%$ per Ga^{66} decay on any transition in the region of 2.75 Mev which is fed completely by positrons. The limit of intensity of a 1.7-Mev transition is set by the 3-crystal pair spectrometer measurements as less than 0.5% per Ga^{66} decay. Since γ rays at 2.75 Mev and 1.7 Mev would be the major modes of de-excitation of a level at 2.75 Mev, the limit of the β^+ intensity to this level is $<1\%$.

stopover ratio is less than 1/100. This ratio is consistent with the predictions of the Scharff-Goldhaber and Weneser model of vibrational excitations of single phonon character. However, such a model predicts that the stopover transition must be pure electric quadrupole, in contradiction with the angular correlation measurements which require at least 10% $M1$ intensity. Moreover, this level is not explained by the recent theory of asymmetric rotors elucidated by Davidov and Filippov. This theory does not allow for a second excited state having less than twice the energy of the first excited state; the energy ratio in this case is 1.8. Even if we assume that this ratio will be lowered by interactions, we are still faced with the anomalous behavior of this level as compared to the similar states in Zn^{64} and Zn^{68} . These latter two levels at 1.79 and 1.60 Mev have crossover to stopover ratios which are close to 1. These nuclei, $\text{Zn}^{64,66,68}$, have been studied by (p, p') interactions by Beurtey et al.,⁶ who examined

both the elastic and inelastic distributions. They find similar behavior for the ground state as well as the first two excited states. Thus, the puzzle remains as to why the crossover transition in Zn^{66} should be so strongly suppressed. One can, of course, argue that only a small change in the wave function describing this state could cause such a suppression since even in Zn^{64} and Zn^{68} the crossover transition is much less than would be expected on a single-particle model.

X. ACKNOWLEDGMENTS

We would like to thank Dr. D. Alburger for the use of his intermediate image spectrometer. Several discussions with Dr. M. Goldhaber, Dr. G. Scharff-Goldhaber, and Dr. A. W. Sunyar were most helpful. We also thank Mr. F. Truman for his technical assistance. Dr. Baker's aid in arranging the cyclotron bombardments was most valuable.

PHYSICAL REVIEW

VOLUME 119, NUMBER 1

JULY 1, 1960

Accurate Method for Measuring Internal Conversion Coefficients*

D. C. LU

Institute for Atomic Research and Department of Physics, Iowa State University, Ames, Iowa

(Received February 10, 1960)

To reveal the effect on internal conversion due to the nuclear structure and extension, measurements are needed which have higher accuracy than is attainable from currently used methods. This article describes how, under certain conditions, the absolute value of the total internal conversion coefficient can be measured to $\pm 0.5\%$ by the use of a large NaI(Tl) detector with a thin well-type window. Complications encountered in the comparison between experimental and computed values are mentioned.

THE influence on internal conversion coefficients (hereafter denoted by I. C. C.) due to the finite extension of the nucleus and its internal structure has led to many recent theoretical discussions¹⁻⁶ and to extensive revisions^{1,2} of the calculated I. C. C. tables which were heavily relied upon in the past. It has been mentioned³ that although internal conversion is no longer the clean cut tool it was thought to be in determining many nuclear properties, its dependence on the nuclear wave function may reveal additional information about nuclear structure, provided that it can be measured experimentally with sufficient accuracy. At

the present, only a few I. C. C. measurements are as accurate as $\pm 5\%$. Even such measurements are not precise enough to reveal the nuclear effects which are generally small. It is the purpose of this paper to demonstrate a new method whereby the I. C. C. of many nuclear transitions can be measured with much improved accuracy. This method is a refinement based upon the same principle that the author described some years ago.⁷ However, since it is not well known, the method and its capability will be described in more detail here.

As an illustration, let us consider Cd^{111m} , whose decay scheme consists of a simple cascade of two transitions as shown in Fig. 1. Each 150-keV $E3$ transition is in coincidence with a 247-keV $E2$ transition, and vice versa. To measure the I. C. C. of these transitions, the sample is placed at the center of a large NaI(Tl) scintillator which has a small diameter well, as shown in Fig. 2. The well is lined with a plastic test tube of 50 mg/cm² wall thickness which is thick enough to cut off all the

* Work was performed in the Ames Laboratory of the U. S. Atomic Energy Commission.

¹ L. A. Sliv and I. M. Band, Leningrad Physico-Technical Institute Reports 1956 and 1958 [translation: Reports 57ICCK1 and 58ICCL1, by Physics Department, University of Illinois, Urbana, Illinois].

² M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

³ E. L. Church and J. Weneser, Phys. Rev. **104**, 1382 (1956).

⁴ T. A. Green and M. E. Rose, Phys. Rev. **110**, 105 (1958).

⁵ L. S. Kisslinger, Phys. Rev. **114**, 292 (1959).

⁶ J. Weneser, Bull. Am. Phys. Soc. **5**, 52 (1960).

⁷ D. C. Lu, W. H. Kelly, and M. L. Wiedenbeck, Phys. Rev. **97**, 139 (1955).

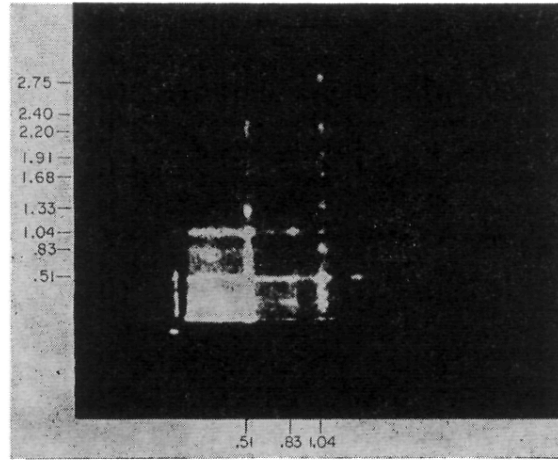


FIG. 5. X-Y-Z analyzer spectrum of Ga^{66} coincidences. The camera field cuts off slightly above 1 Mev in the horizontal direction leading to the asymmetry of the display.