Decay of the Radioisotopes Ge^{68} and Ga^{68}†

DANIEL J. HOREN
Stanford University, Stanford, California
(Received August 18, 1958)

The decays of Ge^{68} (275 day) and Ga^{68} (68 minute) have been reinvestigated with single and coincidence scintillation spectrometers. Comparison of the gamma-ray singles spectra of the two radioisotopes indicates that Ge^{68} decays only to the ground state of Ga^{68} by electron capture. In addition to the known annihilation quanta and a gamma ray of 1.07 Mev in the decay of Ga^{68}, gamma rays of 0.81±0.02, 1.24±0.02, and 1.88±0.02 Mev are observed. A level scheme of Zn^{68} with levels at 1.07, 1.88, and 2.31 Mev is proposed and is compared with level schemes of neighboring even-even nuclei.

I. INTRODUCTION

THE half-life and the positron spectrum of the radioactive isotope Ge^{68} (275 day) have recently been remeasured by Crasemann *et al.*[1] Using a scintillation spectrometer and a single-channel analyzer, they also investigated the gamma-ray spectrum up to an energy of 1.2 Mev and found only a 1.07-Mev gamma ray in addition to the annihilation radiation. This gamma ray was assumed to arise from the decay of the first excited state of Zn^{68}, as surmised by Mukerji and Preiswerk[2] who had measured the beta spectrum of Ga^{68} (68 minute). These authors[2] had found two positron groups with end-point energies of 1.88±0.02 and 0.77 ±0.02 Mev, and a gamma ray of 1.10-Mev with a magnetic lens spectrometer.

More recently, Sinclair[3] has observed a 0.82-Mev gamma ray in the inelastic neutron scattering on Zn^{68}, which he attributed to the decay of a 1.91-Mev level in this isotope, as predicted by Way *et al.*[4] on the basis of the capture gamma-ray work of Kinsey and Bartholomew.[5] From the $Zn^{67}(d,p)Zn^{68}$ reaction, Elwyn has observed levels in Zn^{68} at 1.11, 1.88, and 3.49 Mev.[6]

Since the even-even nuclei in this region of mass number seem to be little understood on a detailed theoretical basis at present, it was considered desirable to investigate more thoroughly a number of radioisotopes which decay to even-even nuclei in the neutron shell $28<N<50$. The present work represents part of such a program.

II. SOURCE PREPARATION

The preliminary work was performed with the Ge^{68} source used by Crasemann *et al.*[1,7] Three subsequent sources were made by bomardment of zinc with 43-Mev alpha particles in the 60-in. cyclotron of the Crocker Radiation Laboratory of the University of California, utilizing the reaction $Zn^{66}(\alpha,2n)Ge^{68}$. For the first source, zinc dust was bombarded for 25 microampere-hours, and for the other two sources, zinc-plated copper probes were irradiated for 200 and 580 microampere-hours, respectively. The chemical separation was similar to that used by Crasemann *et al.*[1] with the following addition: After distillation, the distillate was made 5*M* with HCl, passed through an ion exchange column of Dowex-1 (200 mesh), eluted with 4*M* HCl, precipitated as GeS_2 with H_2S, washed and dried. This additional procedure removed any Ga which might have come over in the distillation. No measurement was made with a new Ge^{68} source for at least one month after bombardment in order to allow for the decay of 40-hour Ge^{69}.

A Ga^{68} source was made by the (γ,n) reaction on natural gallium in the form of Ga_2O_3, using the Stanford Mark II electron linear accelerator at about 26 Mev. The Ga_2O_3 was irradiated for approximately two hours, and measurements begun seventy-five minutes later.

Deuterons of approximately 2.8 Mev from the Stanford cyclotron were used to make a number of auxiliary O^{15} sources by the reaction $N^{14}(d,n)O^{15}$. The nitrogen was in the form of CrN.

III. APPARATUS

The detectors consisted of $1\frac{1}{2}$-in. diameter×$1\frac{1}{2}$-in. long NaI(Tl) cylindrical crystals mounted on DuMont 6292 photomultipliers. The electronic equipment was composed of a conventional fast-slow coincidence circuit with a resolving time of about 0.2-μsec, used in conjunction with a 256-channel pulse-height analyzer.

IV. GAMMA-RAY MEASUREMENTS

A. Comparison of Ge^{68} and Ga^{68} Singles Gamma-ray Spectra

Ga^{68} and Ge^{68} sources were successively sandwiched between $\frac{3}{8}$-in. Lucite absorbers to ensure complete

† Supported in part by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] Crasemann, Rehfuss, and Easterday, Phys. Rev. **102**, 1344 (1956). [The gamma-ray energy was originally measured as 1.02 Mev, but recently remeasured as 1.07 Mev (private communication from B. Crasemann).]

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

[3] Rolf M. Sinclair, Phys. Rev. **107**, 1306 (1957).

[4] Way, King, McGinnis, and van Lieshout, *Nuclear Level Schemes, A=40–A=92*, U. S. Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).

[5] B. Kinsey and G. Bartholomew, Phys. Rev. **89**, 375 (1953).

[6] A. J. Elwyn, Ph.D. dissertation, Washington University, 1956 (unpublished).

[7] The author would like to thank Dr. Crasemann for kindly making this source available to him.

annihilation of the positrons and mounted $1\frac{1}{2}$ in. above a $1\frac{1}{2}$-in. diameter×$1\frac{1}{2}$-in. long NaI crystal. As Ga^{68} decays with a 68-minute half-life,[4] a number of short runs (about ten minutes) representing various integral counting rates were taken with this source. The run whose average integral counting rate was nearest that of the Ge^{68} source, taken under identical conditions for the same time, was used in a comparison of the two decays. This precaution was taken to compensate for possible chance addition of the annihilation quanta. The two spectra were normalized by the integral counts above 0.8 Mev since the decay of Ge^{68} is not expected[4] to contain gamma rays above this energy. It was found that the two spectra overlapped each other (within statistics) up to 1.2 Mev, as shown in Fig. 1. The points represent the Ge^{68} spectrum and the full line represents the Ga^{68} spectrum.

From the data of Fig. 1, it is possible to place a conservative upper limit of 0.01 for the ratio of the number of positrons emitted in the decay of Ge^{68} to the number emitted in the decay of Ga^{68}. Since the total positron branching in Ga^{68} was found to be 89% (see Sec. V, B), this leads to an upper limit of 0.9% for the decay of Ge^{68} by positron emission. It was further established that no gamma ray between 0.60 and 1.10 Mev could be present in the Ge^{68} decay to more than 0.1% and between 0.10 and 0.50 Mev to more than 1% of the intensity of the total positrons emitted in the Ga^{68} decay.

B. Ga^{68} Gamma-Ray Singles Spectrum and Analysis

Having shown the absence of gamma rays in the decay of Ge^{68}, sources of the latter in secular equilibrium with Ga^{68} were used for the study of the decay of Ga^{68}. The singles spectrum was measured using different sources, crystals, and geometries with consistent results. In Fig. 2, the points show the spectrum obtained with the source placed between two $\frac{3}{8}$-in. Lucite absorbers and $2\frac{1}{4}$ in. above a $1\frac{1}{2}$-in. diameter×$1\frac{1}{2}$-in. long NaI crystal. The spectrum above 0.51 Mev was repeated after placing $\frac{1}{4}$ in. of lead between the source and crystal. Comparison of the spectra with and without lead showed that the peaks at 1.07, 1.24, and 1.88 Mev were due to single gamma rays and not addition peaks, while the peak at about 0.68 Mev was predominantly caused by addition of a 0.51-Mev annihilation quantum and its partner's backscattered quantum of about 0.17 Mev.

In order to analyze the singles spectrum, the annihilation-in-flight and bremsstrahlung spectrum arising mostly from the 1.88-Mev positron branch (87%) in the Ga^{68} decay first had to be subtracted. This was accomplished as follows: A number of O^{15} sources were made and data recorded under essentially the same conditions as used for the Ga^{68} singles. The result is shown by the points in Fig. 3. O^{15} is known to be a pure positron emitter with an end-point energy of 1.72 Mev.[8]

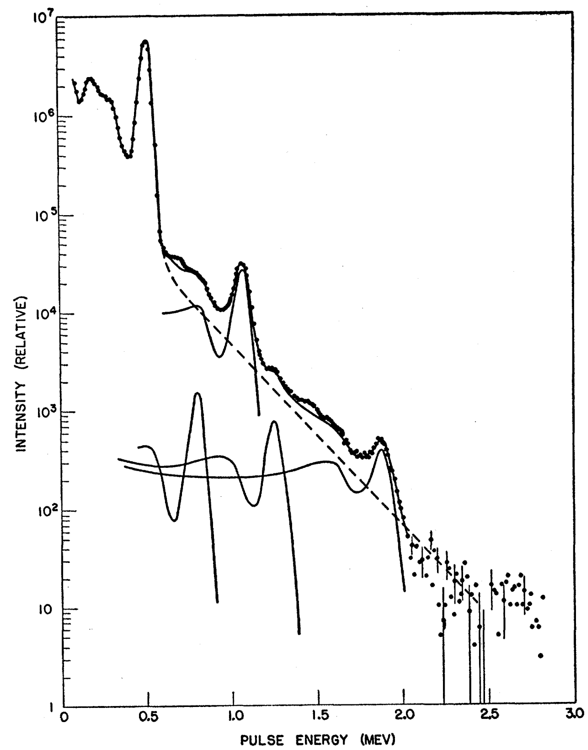

FIG. 2. Ge^{68} gamma-ray spectrum and analysis. The points represent the raw data; the dashed curve represents annihilation in flight and bremsstrahlung arising from the positrons, as determined from Fig. 3. (See text.) The single gamma rays yielded by analysis of the spectrum are shown. The solid curve through the points corresponds to the sum of the single gamma-ray distributions and the continuous spectrum. The excesses in the vicinity of 0.7, 1.45, and 1.60 Mev are discussed in the text.

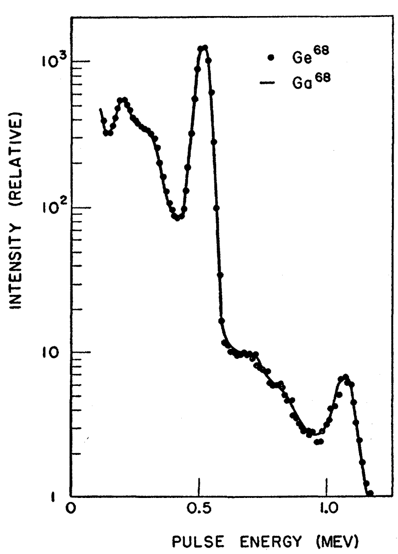

FIG. 1. Ge^{68} (points) and Ga^{68} (solid curve) normalized singles gamma-ray spectra taken under identical conditions.

[8] Kistner, Schwarzschild, Rustad, and Alburger, Phys. Rev. **105**, 1339 (1957).

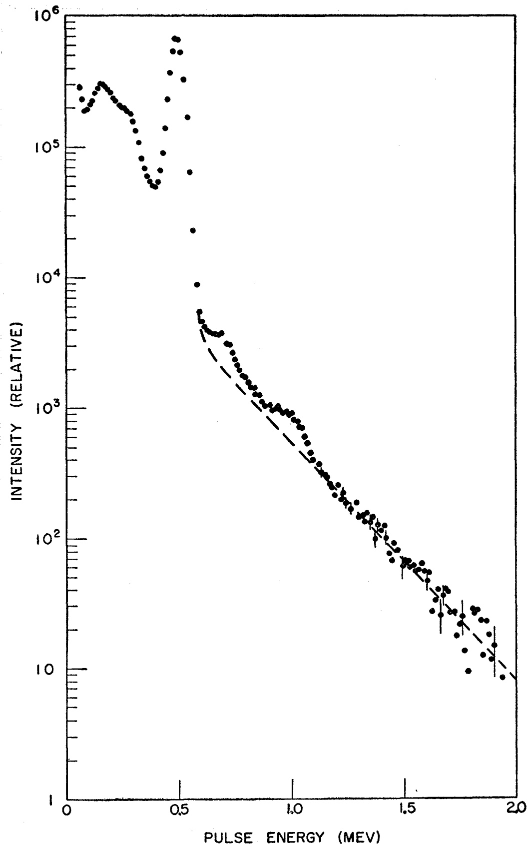

FIG. 3. Annihilation-in-flight and bremsstrahlung spectrum of the 1.72-Mev positrons from the decay of O^{15}. The geometry used here was identical to that used for the data of Fig. 2. The peak at 0.68 Mev is caused by addition of annihilation quanta and their backscattered partners; the peak at 1.02 Mev by chance pile-up of annihilation quanta. The dashed curve was used in the analysis of the Ge^{68} singles. (See text.)

Hence, the peak at about 0.68 Mev is presumed to be due to addition of annihilation quanta and their backscattered partners as mentioned above; the peak at about 1.02 Mev is due to chance pile up of annihilation quanta.

Use was then made of the fact that on a semilogarithmic plot of relative intensity *versus* pulse-height the bremsstrahlung and annihilation-in-flight spectrum is practically a straight line above 0.51 Mev.[9] A straight line was therefore fitted to the points above the peak at 1.02 Mev and extrapolated toward the lower energies (dashed curve in Fig. 3). The joining of this line to the tail of the 0.51-Mev peak is a little uncertain. This, coupled with the addition peak in the vicinity of 0.68 Mev, makes the analysis in this region quite difficult, and this is reflected in our results and quoted errors. The bremsstrahlung and annihilation-in-flight spectrum was normalized to the Ge^{68} spectrum by plotting the two curves on semilogarithm paper and sliding the 0.51-Mev peaks together (dashed curve in Fig. 2). No attempt was made to correct the bremsstrahlung and annihilation-in-flight spectrum for the energy difference between the O^{15} and Ga^{68} positrons. Reference to the work of Gerhart *et al.*[9] leads one to expect this correction to be at most 20% in the region of interest. This was taken into account in computing the errors of the relative intensities.

After subtraction of the bremsstrahlung and annihilation-in-flight contribution, the remainder of the analysis of the spectrum in Fig. 2 was carried out in the customary way of fitting previously determined single gamma-ray spectra (taken under identical conditions) to the resultant curve, starting at the high-energy end. The photopeaks were measured and corrected for absorption, for the ratio of photopeak to total area, and for the detection efficiency of the NaI crystal.[10] Table I lists the results of this analysis. The 1.07-Mev peak has been corrected for chance pile up of annihilation quanta, which amounted to about 5%.

As can be seen in Fig. 2, the sum of the individual contributions to the spectrum (solid curve) does not quite overlap the experimental points in the regions of 0.68 Mev and about 1.2 to 1.7 Mev. The reason for the former has been discussed above. A preliminary analysis of the gamma-ray spectrum had indicated peaks at 1.46 and 1.58 Mev. However, there was a 1.46-Mev peak in the room background, and a slight gain shift between the Ge^{68} and background runs could easily account for its presence in the analysis. Furthermore, calculation showed that the 1.58-Mev peak could be attributed to solid angle addition of annihilation radiation and coincident 1.07-Mev gamma rays (see Sec. C). However, we cannot completely rule out the presence of gamma rays at these energies. These would have an intensity of less than one percent of the intensity of the 1.07-Mev gamma ray.

To investigate the low-energy region, the gain was expanded and the spectrum taken through a 2-in. lead collimator with a $\frac{1}{2}$-in. diameter hole. The source was mounted between $\frac{1}{4}$-mil Mylar, and a $\frac{3}{8}$-in. Lucite plug was placed midway in the collimator to minimize bremsstrahlung and annihilation-in-flight events. No gamma rays between 0.1 and 0.5 Mev were detected,

TABLE I. Ge^{68} gamma-ray data[a] from singles spectrum.

Energy (Mev)	Intensity (relative)[b]
0.81±0.04	~3.7
1.07±0.02	100
1.24±0.02	3.4±1.5
1.88±0.02	4.6±0.8
2.31	≤0.2
0.51 (annihilation quanta)	5460±600

[a] These gamma rays are all from the decay of Ga^{68} to Zn^{68}. (See text, Sec. IV, A.)
[b] The intensities are relative to 100 1.07-Mev gamma rays.

[9] Gerhart, Carlson, and Sherr, Phys. Rev. **94**, 917 (1954).

[10] Use was made in these computations of the detection efficiency calculations of Wolicki, Jastrow, and Brooks, Naval Research Laboratory Report NRL-4833, 1956 (unpublished).

which shows that their intensity could not be greater than 1.3 relative to 100 positrons.

C. Gamma-Gamma Coincidence Spectra

The gamma-gamma coincidence measurements were performed with and without total positron annihilation. For these experiments, the counters were set 110 degrees apart and shielded from one another with a $\frac{1}{2}$-in. lead absorber. The source was placed at the intersection of the counter axes. The detectors were two $1\frac{1}{2}$-in. diameter $\times 1\frac{1}{2}$-in. long NaI crystals, and a conventional fast-slow coincidence circuit of 0.2-μsec resolving time was used in conjunction with a 256-channel pulse-height analyzer.

Figure 4 shows the results obtained by suspending the source between $\frac{1}{4}$-mil Mylar, and setting the discriminating gamma-ray counter differentially at (*a*) 0.81, (*b*) 1.07, (*c*) 1.24, and (*d*) 1.46 Mev. The channel width was approximately 100 kev at each position. In analyzing these curves, it was necessary to include a continuous spectrum which was due in part to coincidences between two photons arising from positron annihilation in flight as well as to 1.07-Mev gamma rays in coincidence with bremsstrahlung and annihilation-in-flight quanta associated with the 0.8-Mev positron group feeding the 1.07-Mev state in Zn^{68}. Curve (*b*) illustrates the result of such an analysis.

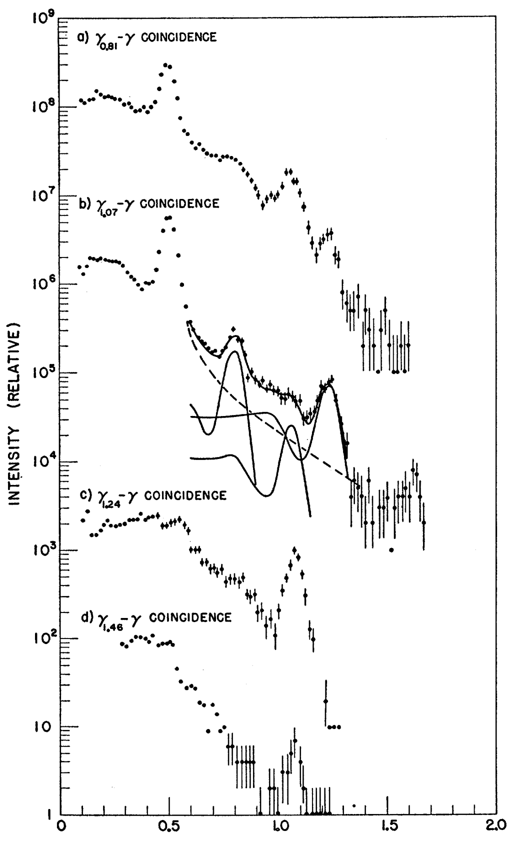

FIG. 4. Ge^{68} gamma-gamma coincidence spectra (incomplete annihilation of positrons). The curves correspond to settings of the gamma-ray discriminator at (*a*) 0.81, (*b*) 1.07, (*c*) 1.24, and (*d*) 1.46 Mev. (See text for method of analysis.)

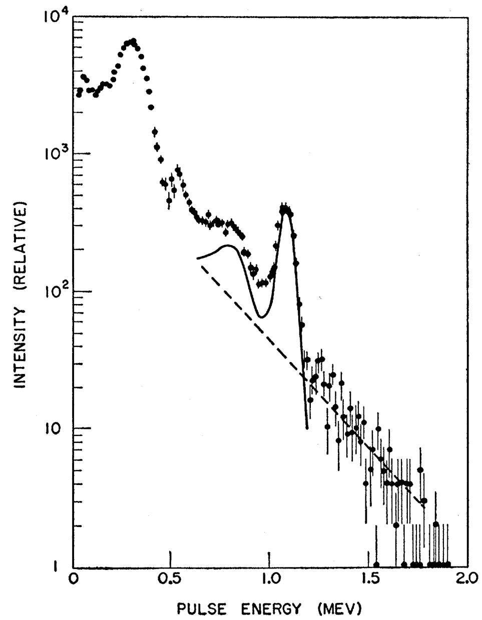

FIG. 5. Ge^{68} gamma-ray spectrum in coincidence with annihilation quanta (with total positron annihilation). The dashed curve represents the assumed annihilation-in-flight and bremsstrahlung background. (See text for discussion.)

After analyzing and correcting the data for the pulses at each discriminator setting caused by Compton events from the higher energy gamma rays, it was found that within the experimental error the data could be explained by 1.07-Mev gamma rays being in coincidence with annihilation quanta and with 0.81- and 1.24-Mev gamma rays. The possibility of a weak 0.81–0.81 Mev gamma-gamma coincidence branch cannot be excluded. The branching ratios obtained from these data were consistent with those determined in the directional correlation experiments, to be discussed in the next section.

For the total-annihilation coincidence measurements, the Ge^{68} source was placed in a copper cylinder whose sides were greater than the range of the high-energy positron group. The coincidence spectrum obtained with the discriminator set to cover the 0.51-Mev photopeak is shown in Fig. 5. The broad peak in the vicinity of 0.3 Mev is undoubtedly caused by coincidences involving annihilation quanta and their partners which have been Compton-scattering by about 70°. The continuous spectrum upon which the 1.07-Mev gamma ray is superimposed is caused by annihilation-in-flight photons. It was assumed to be a straight line[11] on a semilog plot in the region above about 0.65 Mev (dashed curve in Fig. 5). Following this background subtraction, the residue above 0.65 Mev could be com-

[11] See, for example, Hilliard Roderick, Ph.D. dissertation, Stanford University, 1956 (unpublished).

TABLE II. Relative intensities of coincident branches.

Energy (Mev)—(Mev)	Intensity (relative)[a]
(0.81±0.02)—(1.07)	3.5±0.9[c]
(1.24±0.02)—(1.07)	3.4±0.6[d]
(0.51, annihilation quanta)[b]—(1.07)	82.4±13[e]

[a] Intensity relative to 100 1.07-Mev gamma rays.
[b] Positron group to the 1.07-Mev level in Zn^{68}.
[c] Corrected for anistropy as noted in the text.
[d] Corrected for anistropy.
[e] Obtained by combining the relative positron to 1.07-Mev gamma-ray intensity from Table I with the $\beta^+_{0.8}/\beta^+_{total}$ value found in the coincidence work.

pletely accounted for by a 1.07-Mev gamma-ray distribution as determined by a comparison with the 1.11-Mev gamma ray of Zn^{65} taken under similar conditions. To determine the absolute branching of the 0.8-Mev positron branch relative to the total number of positrons, a comparison was made with Na^{22} taken in the same geometry. After making the necessary corrections, a value of 0.015±0.002 was found for the ratio $\beta^+_{0.8}/\beta^+_{total}$. The error includes a twenty percent allowance for the uncertainty in the magnitude of the continuous spectrum under the 1.07-Mev photopeak.

D. Directional Correlation Measurements

For the angular correlation measurements, the source in the form of GeS_2 was suspended between $\frac{1}{4}$-mil Mylar to minimize the positron annihilation in flight. The crystals were shielded with tapered lead collimators, and $\frac{3}{8}$-in. thick Lucite was placed in front of the crystals. The circuitry was the same as that described in the previous section. With the differential discriminator set at 1.07 Mev, it was determined that the singles counting rates were independent of the movable counter position. Measurements were taken at 90°, 130°, and 150°. Interference from annihilation in flight and bremsstrahlung was small for the 1.07 Mev—1.24 Mev correlation. However, it was quite significant in the 1.07 Mev—0.81 Mev correlation, and we were only able to conclude that this correlation is similar to the 1.07 Mev—1.24 Mev correlation.

For the 1.07 Mev—1.24 Mev correlation, values of $A_2=0.33\pm0.11$ and $A_4=0.17\pm0.21$ (not corrected for finite angular resolution of the counters) were obtained in the expression $\bar{W}(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)$. This correlation was used also to determine the 1.24-Mev gamma-ray branching ratio relative to that of the 1.07-Mev gamma-ray branch. The resultant value, as well as a summary of the coincidence measurements, is given in Table II.

From the measurements, the branching of the 0.81-Mev gamma ray relative to that of the 1.24-Mev gamma ray was determined at each angle. Since the three values so obtained were nearly equal, the 0.81 Mev—1.07 Mev gamma-gamma angular anisotropy was assumed to be the same as that for the 1.24 Mev—1.07 Mev correlation in calculating the branching ratio of the 0.81-Mev gamma ray relative to that of the 1.07-Mev gamma ray. This value has been used in Table II.

V. CONCLUSIONS

A. $Ge^{68}-Ga^{68}$ Decay

No evidence has been found for transitions from Ge^{68} to the known[12] excited states of Ga^{68} at 0.19, 0.34, 0.57, and 0.85 Mev.[13] When considered with our small upper limit for positron emission, it appears as though Ge^{68} decays to the ground state of Ga^{68} by about 100% electron-capture. From β-decay energy systematics, Way *et al.*[4] predict a value of about 0.7 Mev for the $Ge^{68}-Ga^{68}$ decay energy. Applying the above branching and decay energy to Moszkowski's nomogram,[14] one finds a $\log ft$ of about 6.8 for the ground-state transition. Since the ground-state spin of Ga^{68} has been determined[15] to be 1, and its parity is expected to be even (on the basis of the shell model), this transition is undoubtedly allowed. Such a relatively large $\log ft$ value for an allowed transition does not appear to be exceptional for the even mass nuclei in this region. In a similar manner, using an upper limit of one percent for gamma-ray branching in the Ge^{68} decay, one finds for the transitions to the first, second, and third excited levels of Ga^{68} lower limits for $\log ft$ of 8.5, 8.2, and 7.3, respectively. About all that one might conclude from these limits is that they are indicative of spins ≥ 2 for these levels if they have even parity. Such spins would be predicted on a zero-range model[16,17] if the proton configuration of Ga^{68} were $[(p_{\frac{3}{2}})^3]_{\frac{3}{2}}$ and the neutron configuration were $[(p_{\frac{3}{2}})^4(f_{\frac{5}{2}})^5]_{\frac{5}{2}}$. Such inference should not be taken too seriously, since the $\log ft$ values are rather sensitive to the $Ge^{68}-Ga^{68}$ decay energy.

B. $Ga^{68}-Zn^{68}$ Decay Scheme

From Table I, the number of positrons to 1.07-Mev gamma rays ($\beta^+_{total}/\gamma_{1.07}$) in the Ga^{68} decay is seen to be 27.3±3.0. This value is considerably larger than that determinered by other workers, as can be seen from Table III. As noted in Table II, combining our $\beta^+_{total}/\gamma_{1.07}$ from the single gamma-ray work with the $\beta^+_{0.8}/\beta^+_{total}$ obtained in the coincidence measurements, we find $\beta^+_{0.8}/\gamma_{1.07}=0.41\pm0.07$. When due account is taken of the 0.81- and 1.24-Mev gamma-ray branches, this value yields an electron-capture to positron ratio (ϵ/β^+) of 1.3±0.3 for the transition to the first excited state of Zn^{68}. An end-point energy of 1.88 Mev[18] for

[12] R. A. Chapman and J. C. Slattery, Phys. Rev. **105**, 633 (1957).
[13] M. K. Ramaswamy has also failed to observe gamma rays in the Ge^{68} decay. He performed x-ray–x-ray and x-ray–gamma coincidence measurements (private communication).
[14] S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).
[15] Hubbs, Marrus, and Worcester, Phys. Rev. **110**, 534 (1958).
[16] C. L. Schwartz, Phys. Rev. **94**, 95 (1954).
[17] A. de-Shalit, Phys. Rev. **91**, 1479 (1953).
[18] A value of 1.88±0.02 Mev has recently been obtained by H. Daniel, Z. Naturforsch. **12a**, 363 (1957).

the ground-state positron transition in conjunction with our measured value of 1.07 Mev for the first excited-state to ground-state transition in Zn^{68} predicts an end-point energy of 0.81 Mev for the positron branch to the first excited level. Assuming this value, a theoretical ϵ/β^+ of 1.6 is determined from the curves of Feenberg and Trigg[19] corrected for L capture with the curves of Rose and Jackson.[20] It should be noted that the K/β^+ ratio is very sensitive to the positron end-point energy in this region.

The decay scheme for the radioactive series Ge^{68}—Ga^{68}—Zn^{68} is shown in Fig. 6, where an ϵ/β^+ ratio[19,20] of 0.11 has been assumed for the ground-state transition. The $\log ft$ values for the ground-state and first excited-state transitions have been taken from the curves of Feenberg and Trigg,[19] and those for the transitions leading to the two upper levels in Zn^{68} from Moszkowski.[14] The spin and parity of the 1.07-Mev level are known[4] to be 2^+. The allowed $\log ft$ values in conjunction with the Ga^{68} ground-state assignment of 1^+ should restrict the spins and parities of the second and third levels to 0^+, 1^+, or 2^+. Of these, the 1.88-Mev crossover gamma ray rules out 0^+ for the 1.88-Mev level. The apparent absence of a crossover gamma ray makes 1^+ unlikely for the 2.31-Mev state. Although the large errors on the angular correlation coefficients do not allow a precise determination of the spin of the 2.31-Mev level, they are consistent with a spin 2, but no limits for the $E2/M1$ mixing ratio of the 1.24-Mev gamma ray can be set. The value of A_4 (0.17±0.21) for the 1.24 Mev—1.07 Mev correlation is inconsistent with a 0—2—0 sequence. Therefore, spin and parity 2^+ are tentatively assigned the 2.31-Mev level in Zn^{68}.

We are unable to chose between assignments 1^+ and 2^+ for the 1.88-Mev state, although the decay from this level has characteristics[21] found in nuclei with 2^+—2^+—0^+ assignments (second, first, and ground states). It might be pointed out that recent work[22] on even-even nuclei in this region is consistent with the assignment 2^+ for the second excited state.

TABLE III. Number of positrons per 1.07-Mev gamma ray in Ga^{68}.

Experimenters	$\beta^+_{total}/\gamma_{1.07}$
Mukerji and Preiswerk[a]	8.8
Crasemann *et al.*[b,c]	14.4±1.7[f]
Ramaswamy[b,d]	19.5±2.1
Present work[e]	27.3±3.0[g]

[a] See reference 2.
[b] Ge^{68} source in equilibrium with Ga^{68}.
[c] See reference 1.
[d] M. K. Ramaswamy (private communication).
[e] Ge^{68} source in equilibrium with Ga^{68}. Same result obtained for pure Ga^{68} source.
[f] Crasemann has repeated this measurement using one of our Ge^{68} sources and different instruments and 1½-in.×1-in. NaI crystals, and has obtained results consistent with his earlier work (reference 1) (private communications).
[g] Measurement made with 1½-in.×1½-in. crystal; same results obtained with 1-in.×1-in. crystal.

[19] E. Feenberg and G. Trigg, Revs. Modern Phys. **22**, 406 (1950).
[20] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).
[21] Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. **28**, 432 (1956).

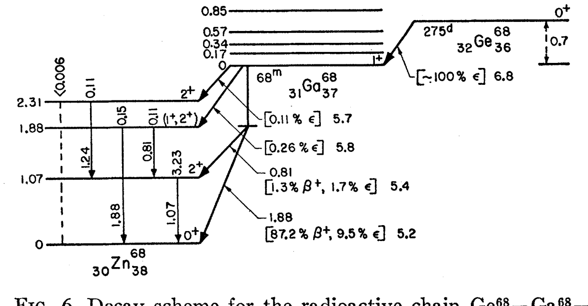

FIG. 6. Decay scheme for the radioactive chain Ge^{68}—Ga^{68}—Zn^{68}. Gamma rays shown in dashed lines have not been detected, and only upper limits for their intensities are given. Positron end-point, gamma-ray, and level energies are in Mev; intensities, in percent decay. Logft values are given to the right of the boxed branching percentages. The Ge^{68}—Ga^{68} decay energy of 0.7 Mev is adopted from reference 4. The levels in Ga^{68} determined by the $Zn^{68}(p,n)Ga^{68}$ reaction (reference 12) are shown. Spins and parities are discussed in the text. *Note added in proof.*—The energy of the first excited state of Ga^{68} should be 0.19 Mev instead of 0.17 Mev, as shown.

C. Comparison with Neutron-Capture Gamma Rays

The agreement between the neutron-capture gamma rays from natural zinc found by Kinsey and Bartholomew[5] and the level scheme proposed in Fig. 6 is quite good. As predicted by Way *et al.*,[4] the 9.12- and 8.31-Mev capture gamma rays undoubtedly lead to the levels at 1.07 and 1.88 Mev in Zn^{68}. Kinsey and Bartholomew[5] tentatively assigned a 7.88-Mev gamma ray to neutron capture in Zn^{64}, but noted that its intensity was rather strong for such a transition, and suggested it might partially be associated with the $Zn^{67}(n,\gamma)Zn^{68}$ reaction. The level at 2.31 Mev in Zn^{68} certainly offers support for the latter assumption. These authors also saw evidence for weak capture gamma rays of 8.58, 8.98, and 9.51 Mev which they assigned to Zn^{68} on the basis of neutron binding energy considerations. A 8.58-Mev gamma ray would predict a level at 1.61 Mev. We are unable to offer evidence for or against such a state, although as previously noted, a gamma ray of nearly this energy could be present in the decay. A 8.78-Mev gamma ray would predict a level at 1.21 Mev, which is close to our observed gamma ray of 1.24 Mev, but for which we can offer no further evidence. Finally, a 9.51-Mev gamma ray, which Kinsey and Bartholomew claim is indicated but not well established, would predict a state at 0.68 Mev. If such a level actually exists, one would expect its spin and parity to be either 0^+ or 2^+, on the basis of the first excited-state systematics of even-even nuclei. The assignment 2^+ would

[22] See, for example, D. J. Horen and W. E. Meyerhof, Phys. Rev. **111**, 559 (1958); Levine, Frauenfelder, and Rossi, Z. Physik **151**, 241 (1958); J. W. Butler and C. R. Gossett, Phys. Rev. **112**, 1257 (1958); T. Jacobi has informed us that the second excited state in Zn^{64} is also indicative of spin and parity 2^+ (private communications).

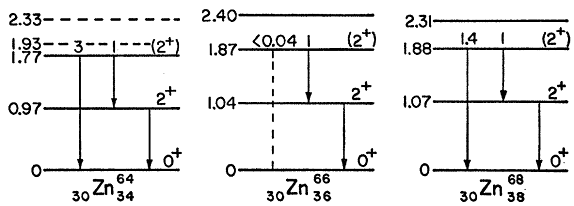

FIG. 7. Comparison of the first few excited states in Zn^{64}, Zn^{66}, and Zn^{68}. The tentative spin and parity assignments for the second excited levels are discussed in the text. The crossover-to-cascade gamma-ray intensities are relative to each other. (The Zn^{64} data were kindly provided by T. Jacobi.) (At the time of this writing, it was not certain whether the third excited level of Zn^{64} occurs at 1.93 or 2.33 Mev; hence, both levels are shown dashed.) *Notes added in proof.*—For Zn^{64} the intensity of the crossover gamma ray should be 0.34 instead of 3. Dr. H. Howe has informed us that the third excited state in Zn^{64} lies at 2.35 Mev (private communication).

be highly unlikely in the light of Heydenburg and Temmer's[23] Coulomb excitation work, as well as the inelastic neutron scattering performed by Sinclair[3] and the inelastic proton scattering experiments of Van Patter *et al.*,[24] which did not reveal the presence of such a level.

D. Comparison of the Level Structure of Zn^{64}, Zn^{66}, and Zn^{68}

In Fig. 7 are shown the first few levels of Zn^{64}, Zn^{66}, and Zn^{68}, as well as the decay characteristics of the first two excited states. The similarity of the level energies is evident. Why the crossover-to-cascade ratio for Zn^{66} is so different from that for Zn^{64} and Zn^{68} is puzzling in view of the energy and probable spin similarities.

In Table IV the first column lists those even-even nuclei in the range $28<N<50$ for which the spin sequence is, or is compatable with, 0^+—2^+—2^+ (ground, first and second levels). (We ignore the 0^+ first excited state of Ge^{72}.) The second column lists the ratio of the second to first excited-state energies (E_2/E_1), the third column the spin and parity of the second excited state, and the fourth column the crossover-to-cascade intensity ratio for radiations originating from the second excited state. It would be of interest to extend the data of Table IV to other even-even nuclei in this region in order to determine whether there is a break in E_2/E_1 *versus* N between $N=40$, 42, and whether the crossover-to-cascade ratio varies in a systematic manner.

[23] Private communication by N. Heydenburg and G. Temmer in reference 4.

[24] Van Patter, Rothman, Porter, and Mandeville, Phys. Rev. **107**, 171 (1957).

VI. ACKNOWLEDGMENTS

The author wishes to express his appreciation to Professor W. E. Meyerhof for suggesting and encouraging this work, as well as for many valuable discussions and for reading the manuscript. Thanks are due Dr. L. F. Chase for producing the O^{15} sources, Dr. W. C. Barber for making the Ga^{68} source, and Mr. Bart Jones and the crew of the 60-in. cyclotron of the Crocker Radiation Laboratory of the University of California for the Ge^{68} sources. Dr. E. K. Hyde generously provided information pertaining to the Ge^{68} chemistry. Thanks are also due Mr. W. Landgraf for two of the Ge^{68} chemical separations, and Dr. J. O. Rasmussen of the University of California Radiation Laboratory for allowing this chemistry to be performed in his laboratory. The assistance of B. Hoop, D. Yount, and K. Kerwin in reducing the data is appreciated.

TABLE IV. Comparison of the first two level characteristics of even-even nuclei in the range $28<N<50$ whose spin and parity sequence is, or is compatible with, 0^+—2^+—2^+ (ground, first, and second levels).

Nuclide	E_2/E_1	I_2	Crossover/cascade	Reference
Fe^{58}	2.00	2^+	1/3.2	a
Ni^{58}	1.70	—	?	b
Ni^{60}	1.64	2^+	1/15	Levine *et al.*[c]
Ni^{62}	1.75	$(0,2)^+$	<1/5	Butler and Gossett[c]
Zn^{64}	1.82	(2^+)	1/2.9	Jacobi[c]
Zn^{66}	1.80	$(0,1,2)^+$	<1/25	Horen and Meyerhof[c]
Zn^{68}	1.76	$(1,2)^+$	1.4/1	Present work
Ge^{72}	1.76	2^+	1/36	d, e, f
Ge^{74}	2.0	—	~3/1	g
Se^{76}	2.06	2^+	~1	d
Se^{78}	2.15	—	?	d
Kr^{82}	1.91	(2^+)	~1/2	d
Kr^{84}	2.16	$(1,2)^+$	1.7/1	h

[a] Frauenfelder, Levine, Rossi, and Singer, Phys. Rev. **103**, 352 (1956).
[b] Spencer, Phillips, and Young, Phys. Rev. **108**, 69 (1957); Rolf Sinclair, Phys. Rev. **102**, 461 (1956).
[c] See reference 22.
[d] See reference 4.
[e] C. F. Coleman, Nuclear Phys. **7**, 488 (1958).
[f] R. G. Arns and M. L. Wiedenbeck, Phys. Rev. **112**, 229 (1958).
[g] Horen, Meyerhof, Kraushaar, Wells, Brun, and Neighbor, Phys. Rev. (to be published).
[h] N. R. Johnson and G. D. O'Kelley, Phys. Rev. **108**, 82 (1957).