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Disintegration of Ge^{68}†

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The isotope Ge^{68} has been prepared by the bombardment of zinc with 37-Mev alpha particles and separated chemically. The half-life of Ge^{68} is 275 ± 20 days, as determined by comparison with a Co^{60} standard over 10 months. The positron spectrum was measured with a thick-lens magnetic spectrometer. Positron groups from Ga^{68} have maximum energies of 1.94 and 0.92 Mev and relative intensities of 1 and 0.04, respectively; no positrons from Ge^{68} were observed above 0.3 Mev. The scintillation spectrum shows annihilation radiation and a 1.02-Mev gamma ray from Ga^{68}. Comparison of areas under these two peaks, corrected for crystal efficiency, indicates that there are 14.4 ± 1.7 positrons per 1-Mev quantum. Within the probable error, this result is compatible with the decay of Ge^{68} by electron capture alone.

I. INTRODUCTION

THE radioactive isotope Ge^{68} probably was first produced in 1938 by Mann[1] who described, but did not definitively assign, a long-lived (~195 day) germanium activity obtained in the bombardment of zinc with 17-Mev alpha particles. Hopkins[2,3] obtained Ge^{68} among 38 nuclear species formed through spallation reactions in the bombardment of arsenic with 190-Mev deuterons. He reported a half-life of 250 days and decay by electron capture. Batzel *et al.*[4] found a long-lived activity in the germanium fraction from the high-energy spallation products of copper, which was comparable with that expected for Ge^{68}.

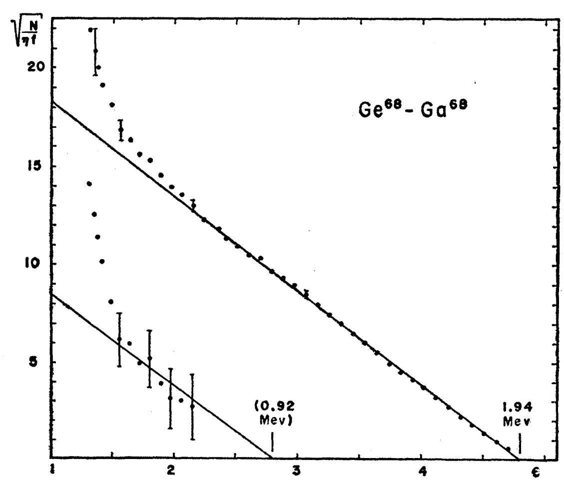

FIG. 1. Fermi plot of the positron spectrum from $Ge^{68}-Ga^{68}$.

Since so little information was available on Ge^{68}, it was considered worth while to produce a sample of this isotope directly, to redetermine its half-life, and to verify the absence of gamma rays and positrons.

II. SOURCE PREPARATION

The Ge^{68} sample was prepared by bombarding a zinc-coated copper probe with 220 microampere-hours of 37-Mev alpha particles in the Crocker Laboratory cyclotron of the University of California, leading to the reaction $Zn^{66}(\alpha,2n)Ge^{68}$. The zinc layer on the probe was dissolved in cold concentrated HCl containing Ge carrier. $GeCl_4$ was distilled into dilute H_2SO_4 and GeS_2 precipitated by bubbling H_2S through the solution. The germanium sulfide was washed and dissolved in NH_4OH, then transferred to thin Tygon foils mounted on Lucite source holders.

III. HALF-LIFE DETERMINATION

Beginning 330 days after bombardment, the activity from a sample of Ge^{68} was determined weekly with a well-shielded Victoreen 1B67 Geiger tube in fixed geometry. A standard source of Co^{60} was counted immediately after every Ge^{68} count. The ratio of the two activities was plotted, thus eliminating the effect of any slow drift in the efficiency of the counting device. The measurements extended over 300 days. Using Brosi and Ketelle's[5] value of 5.38 ± 0.03 years for the half-life of Co^{60}, a half-life of 275 ± 20 days was obtained for Ge^{68}.

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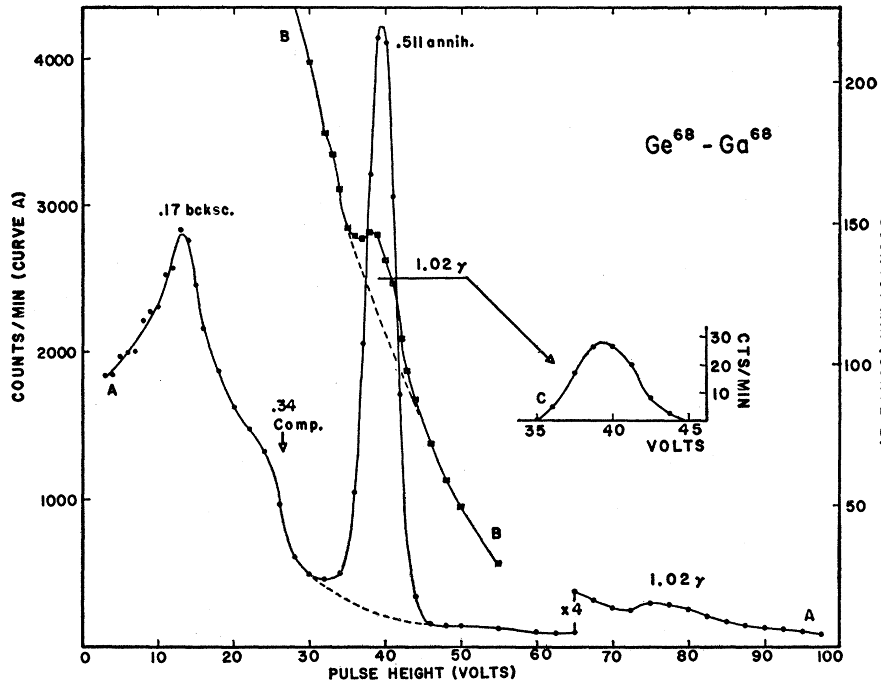

FIG. 2. Scintillation spectrum of the gamma-rays from $Ge^{68}-Ga^{68}$.

IV. POSITRON SPECTRUM

The positron spectrum of Ge^{68} and its daughter, Ga^{68}, was determined with a thick-lens magnetic spectrometer that has been described previously.[6] The detector was a Geiger tube with a 0.67-mg/cm^2 Mylar window. Figure 1 is a Fermi plot of the data, which shows the two known positron branches from the decay of Ga^{68}.[7] The end point of the high-energy positron group is at 1.94 ± 0.05 Mev. The counting rate for the low-energy group is not sufficient to fix its end point from the Fermi plot alone. The measured energy (see below) of the Ga^{68} gamma ray, however, determines the maximum energy of the second positron branch as 0.92 Mev. The intensity of the low-energy group is $(4.1\pm1.4)\times10^{-2}$ times the intensity of the 1.9-Mev group. The excess of positrons below 0.3 Mev presumably is due to source thickness, made necessary by low specific activity of the germanium samples. No positron group that may be ascribed to Ge^{68} is observed above 0.3 Mev, and no conversion electron peaks were found when the spectrometer was set to detect electrons.

V. GAMMA-RAY SPECTRUM

A scintillation spectrometer was used to examine the spectrum of gamma rays emitted by the sample of Ge^{68}. The detector consisted of a cylindrical NaI(Tl) crystal ($1\frac{1}{2}$-in. diameter$\times$1-in. height), and a Du Mont type 6292 multiplier phototube. The distance from source to crystal face was 4.9 cm.

The pulse-height distribution obtained with this equipment is reproduced in Fig. 2. Curve *A* covers the energy range up to 1.2 Mev. In addition to the 1-Mev gamma ray known to occur in the decay of Ga^{68},[7] only annihilation radiation is observed, with its Compton edge at 0.34 Mev and backscattering peak at 0.17 Mev. The energy of the Ga^{68} gamma ray was found to be 1.02 ± 0.02 Mev. The gamma rays from Co^{60} and Na^{22} were used to produce calibration peaks in the immediate vicinity of the unknown. The germanium and reference sources were counted simultaneously in order to maintain the total counting rate at a constant level, since the Du Mont 6292 tube may display a shift of pulse height with counting rate.[8]

In order to compare the intensities of the 1-Mev gamma ray and of the annihilation radiation, the scintillation spectrum of the former was redetermined with the gain of the linear amplifier reduced to one-half of its previous value. In this way, the two peaks appear superimposed and any errors due to a variation of window width with pulse height are eliminated.[9] Counts were accumulated for one hour at each setting of the pulse-height selector. Curve *B* (Fig. 2) shows the results. Curve *C* represents the counts due to the 1-Mev gamma ray alone, after background has been subtracted. The ratio of the area under the annihilation radiation peak to the area under the 1-Mev gamma peak is 142 ± 15. In order to obtain a true intensity ratio, the area ratio has to be corrected for crystal efficiency.

The relative efficiency of the scintillation spectrom-

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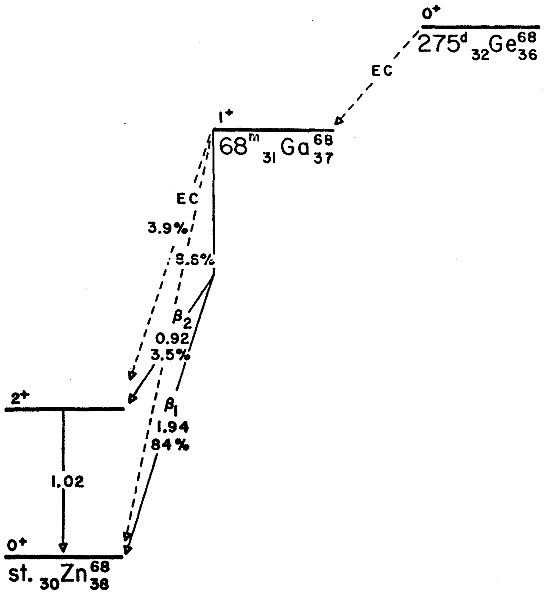

FIG. 3. Decay scheme for $Ge^{68}-Ga^{68}$.

eter at 0.511 and 1.02 Mev was determined in two steps. First, the efficiency for detecting annihilation radiation was found to be 5.91 ± 0.15 times the efficiency for detecting a 1.28-Mev gamma ray, from the respective areas in the scintillation spectrum of Na^{22}. Account was taken of the electron-capture to positron-branching ratio for this isotope.[10] Second, the relative crystal efficiency at 1.28 and 1.02 Mev was obtained from the peak-to-total ratios published by Bell.[11] By applying the proper efficiency correction to the area ratio in the scintillation spectrum and accounting for the fact that each positron will be represented by two annihilation quanta, it was determined that 14.4 ± 1.7 positrons per 1-Mev gamma quantum occur in the decay of Ge^{68} and Ga^{68}.

VI. DISCUSSION

The Fermi plot of Fig. 1 fails to indicate whether low-energy positrons occur in the decay of Ge^{68}. In principle, this fact could be determined by studying whether the decay scheme of Ga^{68} will admit the positron-to-gamma ratio found experimentally. If this ratio is too high, the excess positrons must stem from the decay of Ge^{68}.

A decay scheme for Ga^{68} has been reported by Mukerji and Preiswerk.[7] This is included in Fig. 3. According to the curves of Feenberg and Trigg,[12] the electron-capture to positron ratio is 0.1 for β_1 and 1.1 for β_2. Let N_0 be the total number of Ge^{68} nuclei decaying in unit time, and therefore also the total number of Ga^{68} nuclei that decay in unit time. Now let N_β be the total number of positrons emitted in unit time by both Ge^{68} and Ga^{68}, $N_{\beta1}$ the number of 1.94-Mev positrons and $N_{\beta2}$ the number of 0.92-Mev positrons from Ga^{68}, $N_{\beta0}$ the number, if any, of positrons emitted by Ge^{68}, and N_γ the number of 1-Mev gamma rays. It then follows that the number of positrons emitted by both Ge^{68} and Ga^{68} per 1-Mev gamma quantum is

$$N_\beta/N_\gamma=(N_{\beta1}+N_{\beta2}+N_{\beta0})/(2.1N_{\beta2}), \tag{1}$$

and that the fraction of all Ge^{68} decays that occur by positron emission is

$$N_{\beta0}/N_0=(N_{\beta2}/N_{\beta1})(N_{\beta1}/N_0)\times[2.1(N_\beta/N_\gamma)-1]-N_{\beta1}/N_0, \tag{2}$$

where

$$N_{\beta1}/N_0=0.91/[1+1.91(N_{\beta2}/N_{\beta1})]. \tag{3}$$

The experimentally determined positron branching ratio indicates that the abundance $N_{\beta1}/N_0$ of the high-energy positron group is 0.84 ± 0.03. It then follows from Eq. (2) and from the experimental value for N_β/N_γ that the fraction $N_{\beta0}/N_0$ of all Ge^{68} decays that take place by positron emission is 0.15 ± 0.46.

The large probable error, which makes this result inconclusive, is mostly due to the uncertainty in the positron branching ratio $N_{\beta2}/N_{\beta1}$, which could not be measured more accurately with the low amount of activity available for study. It would appear desirable to produce a strong sample of Ga^{68} directly and to redetermine the branching ratio precisely. Because of the short half-life of Ga^{68} (68 min), this could only be accomplished in the immediate vicinity of an accelerator.

Theoretical considerations make the emission of positrons by Ge^{68} appear unlikely. The curves of Way and Wood[13] for beta-decay energy systematics lead to an expected value of only 0.7 Mev for the energy available in the $Ge^{68}-Ga^{68}$ decay. A further estimate of the decay energy may be gained by considering comparative half-lives. The $Ge^{68}-Ga^{68}$ transition presumably is allowed, since the ground state of Ge^{68} (even-even) is 0+ and the ground state of Ga^{68} must be 1+ because it is linked by allowed transitions to both the 0+ ground state and the 2+ first excited state of Zn^{68}. The logft value for the decay of Ge^{68} by electron capture can therefore be assumed to lie below 6.5.[14] It can then be seen from the nomogram of Moszkowski[15] that theory predicts a decay energy of less than 0.6 Mev. The present experiments, however, do not exclude the possibility of some low-energy positron emission by Ge^{68}.

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