

Neutron-deficient isotopes  $^{64}\text{Ge}$  and  $^{65}\text{Ge}^\dagger$ 

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The  $\beta^+$  decays of  $^{65}\text{Ge}$  and the new isotope  $^{64}\text{Ge}$  have been studied by  $\gamma$ -ray spectrometry with chemically separated sources. Measured half-lives are  $30.0 \pm 1.2$  sec for  $^{65}\text{Ge}$  and  $63.7 \pm 2.5$  sec for  $^{64}\text{Ge}$ . The decay-scheme studies have been supplemented by investigation of the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  and  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  reactions, and the mass excess of  $^{64}\text{Ga}$  was found to be  $-58.819 \pm 0.008$  MeV. Excitation functions for the  $(^3\text{He}, 2n)$ ,  $(^3\text{He}, 3n)$  and  $(^3\text{He}, p2n)$  reactions induced on  $^{64}\text{Zn}$  are presented for comparison with other work. The question of the role of  $^{64}\text{Ge}$  in nucleosynthesis is considered.

<p>RADIOACTIVITY <math>^{64,65}\text{Ge}</math> [by <math>^{64}\text{Zn}(^3\text{He}, xn)</math>]; measured <math>T_{1/2}</math>, <math>E_\gamma</math>, <math>I_\gamma</math>; deduced <math>I_\beta</math>, <math>\log ft</math>. <math>^{64,65}\text{Ga}</math>, deduced levels, <math>J</math>, <math>\pi</math>; Ge(Li) detector.</p> <p>NUCLEAR REACTIONS <math>^{64}\text{Zn}(p, n\gamma)</math>, <math>E = 7.8-9.5</math> MeV; measured <math>E_\gamma</math>. <math>^{64}\text{Ga}</math> deduced levels. <math>^{64}\text{Zn}(^3\text{He}, t)</math>, <math>E = 37.6</math> MeV, measured <math>Q</math>, <math>^{64}\text{Ga}</math> levels; magnetic spectrograph. <math>^{64}\text{Zn}(^3\text{He}, 2n)</math>, <math>(^3\text{He}, 3n)</math>, <math>(^3\text{He}, p2n)</math>, <math>E = 20-70</math> MeV, measured <math>\sigma(E)</math>.</p>
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## I. INTRODUCTION

Recently, Arnett, Truran, and Woosley (ATW)<sup>1</sup> pointed out that certain elements on the high-mass side of the iron peak were synthesized in a supernova explosion when the temperature and density were such that significant amounts of  $^4\text{He}$  were present. In this circumstance a quasiequilibrium is established between  $^4\text{He}$  and relatively stable  $\alpha$ -particle nuclei such as  $^{56}\text{Ni}$ ,  $^{60}\text{Zn}$ ,  $^{64}\text{Ge}$ , etc., and the synthesis process is dominated by a sequence of  $(\alpha, \gamma)$  reactions. The final abundances of the nuclei involved then depend primarily on the  $\alpha$ -particle binding energies and on the freeze-out temperature (the temperature at which nuclear reactions effectively cease). If a nucleus is weakly bound, its abundance will be low and the chain of  $(\alpha, \gamma)$  reactions will be broken at that point.

At the time of their initial calculations,  $^{64}\text{Ge}$  was unknown and ATW used for its mass the prediction of Garvey *et al.*<sup>2</sup> With this mass,  $^{64}\text{Ge}$  turned out to be the weak link in the chain, its synthesis leading (after two  $\beta^+$  decays) to about 1% of the  $^{64}\text{Zn}$  seen in nature. However, the possibility existed that  $^{64}\text{Ge}$  was in fact more tightly bound than the theoretical prediction. In the hope of clarifying this point a number of searches for  $^{64}\text{Ge}$  were undertaken,<sup>3-5</sup> but without success. This raised speculation that  $^{64}\text{Ge}$  was not detected because it had unusual properties including, perhaps, an unusual mass.

This paper reports the detection and identification of  $^{64}\text{Ge}$ , enlarging on the description contained in a preliminary letter.<sup>6</sup> Since the publication of that letter, Davids and Goosman<sup>7</sup> have also ob-

served  $^{64}\text{Ge}$  and have obtained a value for its mass with an uncertainty of  $\pm 0.25$  MeV.

The experimental work described herein comprises five sections. First, a necessary preliminary to the search for  $^{64}\text{Ge}$  was an investigation of the levels of the daughter  $^{64}\text{Ga}$ , and the reaction chosen for this purpose was  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$ . Second, the methods used to produce  $^{64}\text{Ge}$  and measure its decay are described. Third, as a check on the decay scheme (which is of key importance in the mass measurement of Davids and Goosman) the  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  reaction was investigated. Fourth, it was possible to obtain, concurrently with the measurements on  $^{64}\text{Ge}$  decay, new results on the decay of  $^{65}\text{Ge}$  which are in disagreement with early work. Finally, the excitation functions for  $^{64}\text{Zn}(^3\text{He}, 2n)^{65}\text{Ge}$ ,  $^{64}\text{Zn}(^3\text{He}, 3n)^{64}\text{Ge}$ , and  $^{64}\text{Ge}(^3\text{He}, p2n)^{64}\text{Ga}$  were measured, chiefly to confirm the identification of  $^{64}\text{Ge}$  but also to compare with results obtained by Crisler *et al.*<sup>3</sup> for other reactions induced by  $^3\text{He}$  on  $^{64}\text{Zn}$ .

II. IDENTIFICATION OF  $^{64}\text{Ge}$ A.  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  experiments

The  $\beta^+$  decay of even-even  $^{64}\text{Ge}$  populates states in the daughter  $^{64}\text{Ga}$  and the identification of  $^{64}\text{Ge}$  is based primarily on the observation of  $\gamma$  rays from the decay of  $^{64}\text{Ga}$  levels. In order to find what  $\gamma$  rays might be expected in the  $^{64}\text{Ge}$  decay, a preliminary experiment was carried out in which the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  reaction was studied on line.

A self-supporting 1-mg  $\text{cm}^{-2}$  target of metallic  $^{64}\text{Zn}$ , enriched to 99.85%, was bombarded with

proton beams from the Michigan State University (MSU) cyclotron.  $\gamma$  rays were observed at  $90^\circ$  with a Ge(Li) detector having a resolution of 1.2 keV at 122 keV, and an efficiency 7% of that of a  $7.6 \times 7.6$ -cm NaI(Tl) detector at 1333 keV. Singles spectra were taken at a number of beam energies both above and below the threshold for production of  $^{64}\text{Ga}$  by the  $(p, n)$  reaction. The use of a thin target and a thin "Kapton" (polyimide foil) window on the scattering chamber allowed observation of low-energy  $\gamma$  rays. The energy calibration of the system was checked before and after the experiment using sources of  $^{241}\text{Am}$ ,  $^{139}\text{Ce}$ ,  $^{22}\text{Na}$ , and  $^{60}\text{Co}$ .

Spectra were obtained at eight bombarding energies as listed in Table I. Those  $\gamma$  rays appearing for the first time at energies above the calculated threshold for production in the  $^{64}\text{Zn}(p, n)^{64}\text{Ga}$  reaction are considered probably attributable to that reaction. Most others were due to the  $^{64}\text{Zn}(p, p'\gamma)^{64}\text{Zn}$  reaction and reactions induced on aluminum,

TABLE I. Proton beam energies used in the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  experiments.

	$E_{\text{lab}}^a$ (MeV)	Excitation energy in $^{64}\text{Ga}^b$ (MeV)
1	7.830	-0.251
2	8.193	0.107
3	8.225	0.138
4	8.251	0.164
5	8.305	0.217
6	8.418	0.328
7	8.577	0.485
8	9.530	1.423

<sup>a</sup> The proton energies are determined from the calibration of the beam transport system and have an uncertainty of  $\pm 15$  keV.

<sup>b</sup> Calculated assuming a mass excess for  $^{64}\text{Ga}$  of  $-58.830$  MeV. See Sec. III.

TABLE II.  $\gamma$  rays observed during proton bombardment of enriched  $^{64}\text{Zn}$ .

Energy (keV)	Identification	Threshold number <sup>a</sup>	Energy (keV)	Identification	Threshold number <sup>a</sup>
1	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	3	31	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
2	$^{66}\text{Zn}(p, n\gamma)^{66}\text{Ga}$		32	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
3	$^{64}\text{Zn}(p, \alpha)^{61}\text{Cu}(\beta^+)^{61}\text{Ni}$		33	Ann. rad.	
4	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	3	34	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
5	$^{19}\text{F}(p, p'\gamma)^{19}\text{F}$		35	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
6	$^{65}\text{Ga}(\beta^+)^{65}\text{Zn}$		36	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
7	$^{57}\text{Co}(\beta^+)^{57}\text{Fe}$		37	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
8	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8	38	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
9	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	3	39	$^{64}\text{Zn}(p, \alpha)^{61}\text{Cu}(\beta^+)^{61}\text{Ni}$	
10	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	7	40	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
11	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$		41	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
12	$^{27}\text{Al}(p, p'\gamma)^{27}\text{Al}$		42	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
13	$^{19}\text{F}(p, p'\gamma)^{19}\text{F}$		43	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
14	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$		44	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
15	$^{19}\text{F}(p, n\gamma)^{19}\text{Ne}$		45	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
16	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$		46	$^{64}\text{Zn}(p, p'\gamma)^{64}\text{Zn}$	
17	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	7	47	$^{27}\text{Al}(p, p'\gamma)^{27}\text{Al}$	
18	$^{64}\text{Zn}(p, \alpha)^{61}\text{Cu}(\beta^+)^{61}\text{Ni}$		48	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
19	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8	49	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8
20	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	7	50	$^{64}\text{Zn}(p, p'\gamma)^{64}\text{Zn}$	
21	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$		51	$^{64}\text{Zn}(p, \alpha\gamma)^{61}\text{Cu}$	
22	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	7	52	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	
23	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8	53	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	
24	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8	54	$^{64}\text{Zn}(p, \alpha\gamma)^{61}\text{Cu}$	
25	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8	55	$^{64}\text{Zn}(p, p'\gamma)^{64}\text{Zn}$	
26	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$		56 <sup>l</sup>	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	
27	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	7	57	$^{27}\text{Al}(p, p'\gamma)^{27}\text{Al}$	
28	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$				
29	$^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$	8			
30	$^{64}\text{Zn}(p, \alpha\gamma)^{61}\text{Cu}$				

<sup>a</sup> See Table I. Where not noted, the  $\gamma$  rays were seen at  $E_p = 7.830$  MeV, below the threshold for production of  $^{64}\text{Ga}$ .

the beam-pipe material. A few lines remain unidentified. A complete list of  $\gamma$ -ray energies and production thresholds is given in Table II, together with their suggested origin. (Here, and throughout this paper, the uncertainty in the last digit of a number is placed in parentheses; that is, 42.8(3) keV means  $42.8 \pm 0.3$  keV, etc.) The threshold numbers correspond to the list in Table I. Figure 1 shows two spectra, one at a beam energy below threshold for all  $^{64}\text{Ga}$   $\gamma$  rays but the 42.8-keV line, and the other at the highest energy used. All energies marked above the upper spectrum are presumed to be from the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  reaction, while the other transitions are from competing processes. Those not identified are shown in parentheses. The identification of the stronger transitions as belonging to the  $^{64}\text{Ga}$  level scheme has been confirmed by the recent neutron- $\gamma$  and  $\gamma$ - $\gamma$  coincidence experiments on the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  reaction by King, Draper, and McDonald,<sup>8</sup> by Davids, Matthews, and Whitmire,<sup>9</sup> by Hansen, Gregory, and Dietrich,<sup>10</sup> and by Gosselaar.<sup>11</sup>

#### B. Preparation of Ge sources

Neutron-deficient Ge isotopes were produced by  $^3\text{He}$  bombardment of natural and enriched  $^{64}\text{Zn}$  targets. Chemical separations were necessary, not only to identify the activities as isotopes of germanium, but also to remove other radioelements produced in far greater abundance. In particular, the cross section for production of 159-sec  $^{64}\text{Ga}$  by the  $^{64}\text{Zn}(^3\text{He}, p2n)^{64}\text{Ga}$  reaction is three orders of magnitude larger than that for the  $^{64}\text{Zn}(^3\text{He}, 3n)^{64}\text{Ge}$  re-

action. Observation of the growing in of the  $^{64}\text{Ga}$  daughter activity resulting from the decay of  $^{64}\text{Ge}$  is very desirable confirmation of the isotopic identification, but requires extremely high radiochemical rejection of reaction-produced  $^{64}\text{Ga}$ .

Targets of 99.66% enriched  $^{64}\text{Zn}$  were prepared by reduction of  $^{64}\text{ZnO}$  with Zr powder and simultaneous evaporation of the  $^{64}\text{Zn}$  onto thick copper backings. These targets, 5–10  $\text{mg cm}^{-2}$  thick and with an area of approximately 5  $\text{mm}^2$ , were irradiated in a pneumatic rabbit facility<sup>12</sup> for periods of 2 min each. A 1- $\mu\text{A}$  70-MeV  $^3\text{He}$  beam from the MSU cyclotron passed through a vacuum window and an energy degrader consisting of 300  $\text{mg cm}^{-2}$  of zinc foil before impinging on the target with an energy of 49 MeV.

The chemical separation of Ge activities from the zinc targets took advantage of the high volatility of  $\text{GeCl}_4$  relative to the chlorides of other elements. A schematic diagram of the apparatus used is shown in Fig. 2. The metallic Zn of the irradiated targets was dissolved in concentrated HCl which contained  $\text{KClO}_3$  as an oxidizing agent, as recommended by Porile.<sup>13</sup> No carrier was used, and the  $\text{GeCl}_4$  was vacuum distilled at room temperature by pumping. The activity which recondensed in a cold trap cooled by dry ice in alcohol was found to comprise only Ge isotopes and their daughters together with a little  $^{10}\text{C}$ . The total time from the end of irradiation to the start of counting was approximately 25 sec, consisting of ~10 sec for transport and demounting of the target and ~15 sec for the chemical separation. After each run,

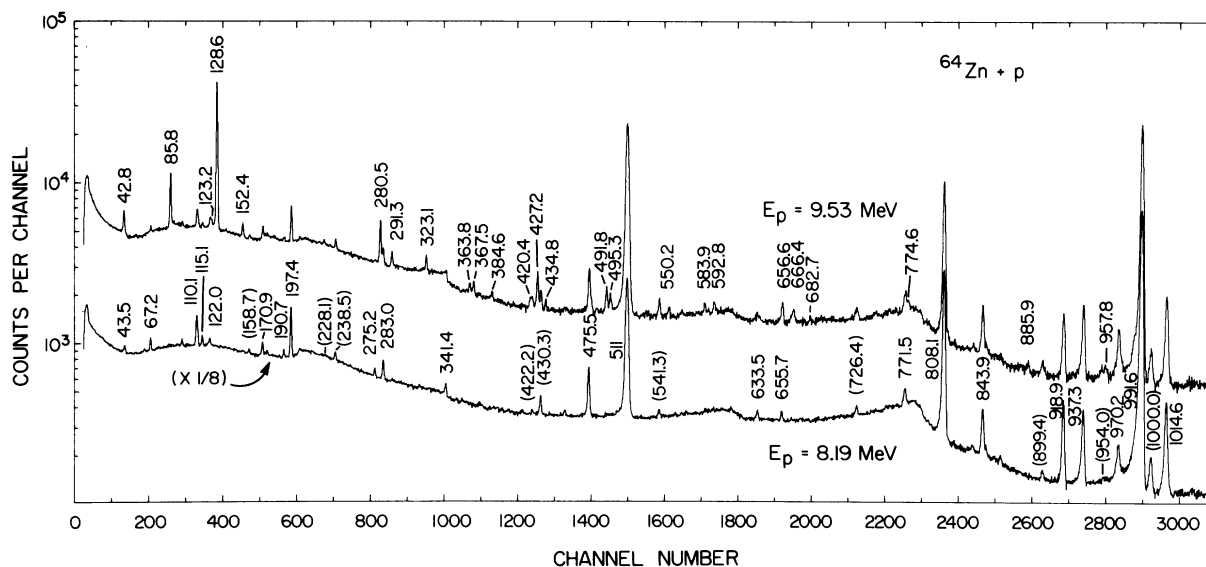


FIG. 1. In-beam  $\gamma$ -ray spectra resulting from proton bombardment of  $^{64}\text{Zn}$ . The lower spectrum is at an energy too low to excite transitions in  $^{64}\text{Ga}$ , while the upper is at an energy 1.4 MeV above the  $(p, n)$  threshold. Only transitions marked above the upper graph are attributable to  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$ ; those in parentheses have not been identified.

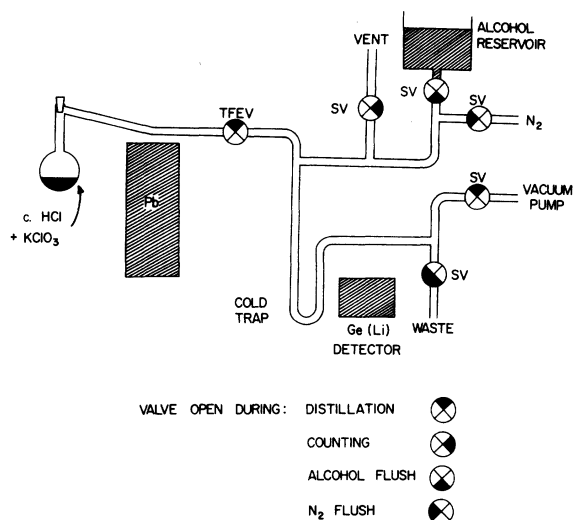


FIG. 2. Schematic diagram of apparatus used for distillation of  $\text{GeCl}_4$ . SV denotes solenoid valve, TFEV denotes a manually-operated Teflon valve.

remaining activities were removed from the cold trap by flushing, first with alcohol and then with dry  $\text{N}_2$  gas.

The cold trap was located inside a lead cave and  $\gamma$  rays were detected with the  $\text{Ge}(\text{Li})$  detector described above. On each run, spectra of 4096 channels were accumulated for eight successive 50.0-sec intervals and routed into separate locations in the memory of an XDS Sigma-7 computer. The routing signals were obtained by digital division of a highly stable oscillator signal. A fixed-rate pulser signal was also recorded for correction of

dead-time losses. For activities that decay with time such a method is only approximate, but by estimating the relevant corrections and maintaining small dead times, adequate accuracy can be obtained. Two normalization procedures were compared, one using the pulser information and the second using lines from long-lived (2.4-h)  $^{66}\text{Ge}$ , and were found to agree well except for a small but systematic discrepancy in the first time group. This was traced to the motion of a small amount of alcohol which remained inside the cold trap after flushing and which was disturbed by pumping. Activities trapped in the alcohol thus moved slightly during the first few seconds of counting but the effect was small (<15%) and could be corrected by normalizing that group to the  $^{66}\text{Ge}$  lines. For subsequent groups, no difference between pulser normalization and  $^{66}\text{Ge}$  normalization could be detected.

The experimental spectrum resulting from a total of 12 irradiations is shown in Fig. 3 for the first 50.0-sec time group. For each  $\gamma$  ray observed, the fitting program SAMPO<sup>14</sup> was used to find peak positions and areas, and a decay curve for each line was then derived. The energies of the  $\gamma$  rays were determined from a calibration with standard sources. The relative intensities were obtained by substituting a natural Zn target for the  $^{64}\text{Zn}$  one, carrying out the irradiation and chemical separation as before, and using the many strong lines from  $^{66}\text{Ge}$ <sup>15</sup> and  $^{67}\text{Ge}$ <sup>16</sup> to calibrate the detector efficiency. A complete list of all  $\gamma$  rays seen, with their assigned parentage, individually fitted half-lives (where applicable), and intensities, is given in Table III. The line at 1779 keV has not been assigned.

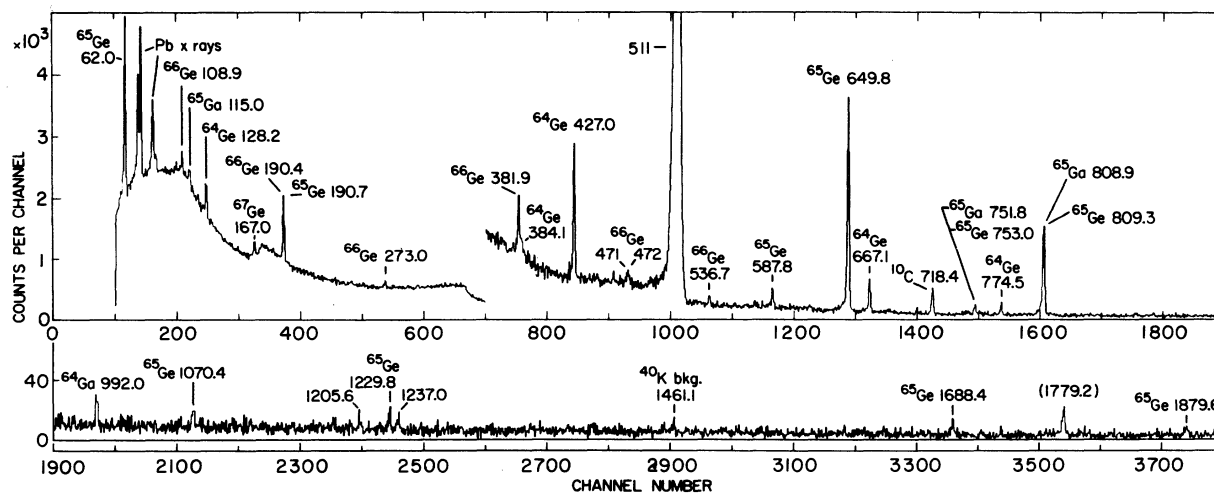


FIG. 3.  $\gamma$ -ray spectrum accumulated in first 50.0-sec interval following chemical separation of Ge from the  $^{64}\text{Zn}$  targets. A spectrum taken 100 sec later is shown in Ref. 6.

TABLE III.  $\gamma$  rays observed following  $^3\text{He}$  bombardment of enriched  $^{64}\text{Zn}$ .

	Energy (keV)	Present work		Photon intensity <sup>b</sup>	Jongsma <i>et al.</i> <sup>a</sup>	
		Assigned parentage	Half-life (sec)		Energy (keV)	Photon intensity
1	62.0(2)	$^{65}\text{Ge}$	30.1(24)	24.0(40)	62.1(5)	26.0(50)
2	108.9(2)	$^{66}\text{Ge}$	Long			
3	115.0(2)	$^{65}\text{Ga}$	Compound			
4	128.2(2)	$^{64}\text{Ge}$	62.0(100)	10.7(7)		
5	167.0(2)	$^{67}\text{Ge}$	~1000			
6	190.4(3)	$^{66}\text{Ge}$	Long			
7	190.7(3)	$^{65}\text{Ge}$	31.1(20)	9.9(4)	190.8(2)	10.2(6)
8	273.1(3)	$^{66}\text{Ge}$	Long			
9	381.9(3)	$^{66}\text{Ge}$	Long			
10	384.1(3)	$^{64}\text{Ge}$	85.0(180)	4.7(5)		
11	427.0(3)	$^{64}\text{Ge}$	63.7(24)	37.4(10)		
12		$^{65}\text{Ge}$			459.1(5)	2.0(3)
13	471	$^{66}\text{Ge}$	Long			
14	472	$^{66}\text{Ge}$	Long			
15	536.7(3)	$^{66}\text{Ge}$	Long			
16	587.8(3)	$^{65}\text{Ge}$	30.3(47)	2.5(3)	587.7(2)	2.6(4)
17		$^{65}\text{Ge}$			618.7(4)	1.5(2)
18	649.8(3)	$^{65}\text{Ge}$	29.1(11)	33.0(13)	649.7(2)	32.6
19	667.1(3)	$^{64}\text{Ge}$	58.0(40)	16.9(10)		
20	718.4(3)	$^{10}\text{C}$	20.9(46)			
21	751.8(3)	$^{65}\text{Ga}$	Compound			
22	753.0(3)	( $^{65}\text{Ge}$ )	Short	1.3(2)		
23	774.5(3)	$^{64}\text{Ge}$	85.4(240)	7.0(6)		
24	808.4(3)	$^{64}\text{Ga}$	Compound			
25	809.3(3)	$^{65}\text{Ge}$	37.2(23)	21.0(10)	809.1(2)	21.2(13)
26		$^{65}\text{Ge}$			826.8(15)	0.36(13)
27		$^{65}\text{Ge}$			884.9(3)	0.33(13)
28		$^{65}\text{Ge}$			970.7(15)	0.23(10)
29	992.0(3)	$^{64}\text{Ga}$	Compound			
30	1070.4(3)	$^{65}\text{Ge}$	20.0(130)	1.3(2)	1070.2(3)	0.91(10)
31		$^{65}\text{Ge}$			1075.9(3)	0.82(10)
32		$^{65}\text{Ge}$			1150.7(15)	0.13(7)
33		$^{65}\text{Ge}$			1183.6(3)	0.46(10)
34	1205.6(5)	$^{65}\text{Ge}$	Short	1.4(4)	1205.7(4)	1.21(13)
35	1229.8(3)	$^{65}\text{Ge}$	Short	2.2(3)		
36	1237.0(5)	$^{65}\text{Ge}$	Short	1.9(4)	1237.1(3)	1.24(10)
37	1387.7(5)	$^{64}\text{Ga}$	Compound			
38		$^{65}\text{Ge}$			1511.9(10)	0.33(7)
39		$^{65}\text{Ge}$			1600.8(5)	0.67(10)
40		$^{65}\text{Ge}$			1616.6(5)	0.72(10)
41	1688.4(5)	$^{65}\text{Ge}$	32.0(90)	3.6(5)	1688.5(5)	2.18(18)
42	1779.2(5)		89.0(80)			
43		$^{65}\text{Ge}$			1816.3(15)	0.39(10)
44	1879.6(6)	$^{65}\text{Ge}$	28.0(50)	3.6(5)	1879.2(5)	0.95(23)

<sup>a</sup> Reference 25.  $\gamma$  rays above 1900 keV not listed.<sup>b</sup> Intensity per 100 decays of assigned parent. Over-all normalization:  $100 \pm 10$  for  $^{64}\text{Ge}$ ,  $100^{+30}_{-10}$  for  $^{65}\text{Ge}$ .

C. Decay of  $^{64}\text{Ge}$ 

Five of the  $\gamma$  rays observed in the chemically separated sources, the 128-, 384-, 427-, 667-, and 775-keV lines, correspond to transitions observed in the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  reaction. They decay with the same half-life, within experimental uncertainty, and are all assigned to the decay of  $^{64}\text{Ge}$ . A sixth line, at 1779 keV, has a half-life similar to the others but is present in bombardments at  $E_{^3\text{He}} = 20$  MeV [below the ( $^3\text{He}, 3n$ ) threshold] and cannot therefore be assigned to  $^{64}\text{Ge}$ . At the time of the initial report of this work<sup>6</sup> it was known that a weak transition of 427.83 keV existed in the decay of 2.4-h  $^{66}\text{Ge}$  (1.9% of the 382-keV intensity<sup>15</sup>), and that a  $\gamma$  ray of 427.1 keV was present in the decay of  $^{64}\text{Ga}$  (2% of the 992-keV intensity<sup>17</sup>). Subsequently, however, Davids *et al.*<sup>18</sup> searched carefully for the latter transition and found no evidence for it (<0.018% of the 992-keV intensity). This leads to a slight revision in the  $^{64}\text{Ge}$  half-life from the previously reported value of 62.3(20) sec to 63.7(25) sec. Davids and Goosman<sup>7</sup> find from the  $^{54}\text{Fe}(^{12}\text{C}, 2n)^{64}\text{Ge}$  reaction a half-life of 70(7) sec, in agreement with this result. Using the measured half-life of  $^{64}\text{Ge}$  (63.7 sec) and the known half-life of  $^{64}\text{Ga}$  (159 sec)<sup>19</sup> the growth and decay of the 992-keV line from  $^{64}\text{Ga}$  can be accurately fitted, which is further confirmation of the isotopic identification of  $^{64}\text{Ge}$  (see Fig. 2 of Ref. 6). Furthermore, the initial activity of  $^{64}\text{Ga}$  found in such a fit is, within experimental error, zero, showing that all the observed  $^{64}\text{Ga}$  results from  $^{64}\text{Ge}$  decay, and that the radiochemical rejection of reaction-produced  $^{64}\text{Ga}$  is in excess of  $10^4$ .

A decay scheme for  $^{64}\text{Ge}$  constructed with the aid

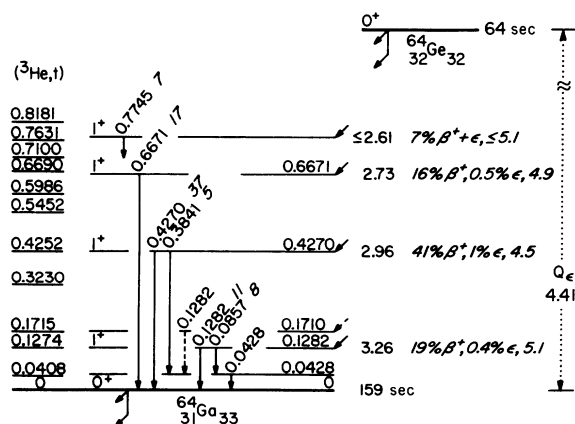


FIG. 4. Decay scheme for  $^{64}\text{Ge}$ . The notation and conventions of Nuclear Data Sheets have been adopted. The 42.8- and 85.7-keV transitions have not been experimentally observed in the decay of  $^{64}\text{Ge}$ ; their energies and intensities are derived from other data (see text).

of the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  results (present work and Refs. 8–11) and the  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  spectrum (to be discussed below) is shown in Fig. 4. The mass of  $^{64}\text{Ge}$  has recently been measured by Davids and Goosman<sup>7</sup> who observed positrons in coincidence with 427-keV  $\gamma$  rays and found the endpoint of that  $\beta^+$  transition to be 2.96(25) MeV. The  $\log ft$  values shown in Fig. 4 are based on that result. Absolute  $\gamma$ -ray intensities have been obtained by normalization to the measured growth of  $^{64}\text{Ga}$  activity. A total of 85(10)% of the  $^{64}\text{Ge}$  decay is accounted for, provided none of the observed  $\gamma$  rays is in cascade or appreciably converted. Spin and parity assignments of  $1^+$  have been made where  $\log ft$  values indicate an allowed transition. Placement of the 774.5-keV  $\gamma$  transition is uncertain, but the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  excitation function requires it to originate below 1.4 MeV excitation in  $^{64}\text{Ga}$ . The 85.7-keV transition was not observed directly in the experiments on  $^{64}\text{Ge}$  decay because of the presence of Pb x rays, but its intensity relative to the 128.2-keV line may be inferred from the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  experiments. However, the intensity reported here differs from that previously reported<sup>6</sup> because it is now known that the 128-keV line is an unresolved doublet (see below). The present result for the 86-keV line is taken from the  $(p, n\gamma)$  spectra at  $E_p = 8.225$  and 8.251 MeV because those energies are below threshold for exciting the second 128-keV line (from the level at 171 keV). The intensity obtained for the 86-keV line must still be regarded as rather uncertain, because angular-correlation effects have been neglected. The 42.8-keV  $\gamma$  ray was also not observed, but its presence is implied by the 384- and 86-keV decays. Its intensity (including internal conversion) must be between 13 and 28% per decay of  $^{64}\text{Ge}$ . (However, it must be remarked that since neither the 86- nor the 43-keV  $\gamma$  rays have been actually observed in  $^{64}\text{Ge}$  decay, their presence is only conjectural. The experimental results would also support a scheme in which allowed  $\beta^+$  decay populated the 171-keV level in  $^{64}\text{Ga}$ . On the other hand, the fact that the 171-keV level does not undergo  $\gamma$  decay to the ground state<sup>8–11</sup> weakly suggests that its spin is not 1.)

III.  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  REACTION

Although  $^{64}\text{Ga}$  has been extensively studied by the  $^{64}\text{Zn}(p, n\gamma)^{64}\text{Ga}$  reaction, seemingly conflicting evidence has come from the threshold studies of Davids, Matthews, and Whitmire<sup>9</sup> and from the  $n\text{-}\gamma$  and  $\gamma\text{-}\gamma$  coincidence studies of Davids, Matthews, and Whitmire,<sup>9</sup> King, Draper, and McDonald,<sup>8</sup> and Hansen, Gregory, and Dietrich.<sup>10</sup> For example, the threshold measurements of Davids *et al.* showed clearly that the 43-keV line was ground-

state transition, while the 128- and 86-keV  $\gamma$  rays had a common threshold at 128 keV excitation. On the other hand, the  $\gamma$ - $\gamma$  coincidence experiments showed a number of transitions in coincidence with both the 128- and the 86-keV line, but others coincident only with the 128-keV  $\gamma$  ray, implying that the 128- and 86-keV transitions do not originate from the same level.

In hopes of resolving the discrepancies and at the same time measuring the mass of  $^{64}\text{Ga}$ , the  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  reaction has been examined at 37.6 MeV using an Enge split-pole spectrograph to analyze the tritons. Targets of approximately  $100 \mu\text{g cm}^{-2}$  of  $^{64}\text{Zn}$  enriched to 99.66% were prepared by vacuum evaporation onto  $20\text{-}\mu\text{g cm}^{-2}$  carbon foils. Spectra were taken at laboratory angles of  $9^\circ$  and  $12^\circ$  with an energy resolution of 35 keV. The spectrum obtained at  $9^\circ$  using a nuclear emulsion is shown in Fig. 5. A recent high-resolution study<sup>20</sup> of the  $^{64}\text{Zn}(p, n)^{64}\text{Ga}$  reaction by neutron time of flight has shown the existence of previously unknown states at 171 and 323 keV excitation in  $^{64}\text{Ga}$ , and this has facilitated an unambiguous interpretation of the complex ( $^3\text{He}, t$ ) spectrum. At both  $9^\circ$  and  $12^\circ$  the population of the ground state is very weak, but fitting the peaks with the program SAMPO<sup>14</sup> yields its position with a relative uncertainty of 4 keV. A complete list of energy levels

observed in the  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  reaction is given in Table IV and the low-lying levels are also shown in Fig. 4. For the purpose of calculating excitation energies, the 323-keV state has been used as a reference rather than the ground state. Identification of the isobaric analog of the  $^{64}\text{Zn}$  ground state (IAS) is based on its dominant strength in the  $12^\circ$  spectrum. A more complete angular distribution for the IAS has been obtained by Hinrichs *et al.*<sup>21</sup>

The observation of the 171-keV state in  $^{64}\text{Ga}$  in the ( $^3\text{He}, t$ ) spectrum and the ( $p, n$ ) time-of-flight spectrum<sup>20</sup> provides an explanation for the discrepancy mentioned earlier. It appears that there are two transitions of 128 keV, one deexciting the 128.2-keV state and the other deexciting the 171-keV state. This interpretation is supported by the most recent work of the authors of Refs. 8–11.

The  $Q$  value of the 323-keV level was determined by comparison with the  $^{27}\text{Al}(^3\text{He}, t)^{27}\text{Si}$  reaction. The 2646.8-keV level<sup>22</sup> in  $^{27}\text{Si}$  provides a calibration peak which lies very close to the 323-keV state of  $^{64}\text{Ga}$  in the focal plane of the spectrograph. The resulting mass excess for  $^{64}\text{Ga}$  is  $-58.819(8)$  MeV. There is perhaps a minor disagreement between the present work and the ( $p, n$ ) threshold measurement of Davids, Matthews, and Whitmire<sup>9</sup> [ $-58.836(6)$  MeV], but a substantial deviation from earlier work<sup>23</sup> [ $-58.934(30)$  MeV].

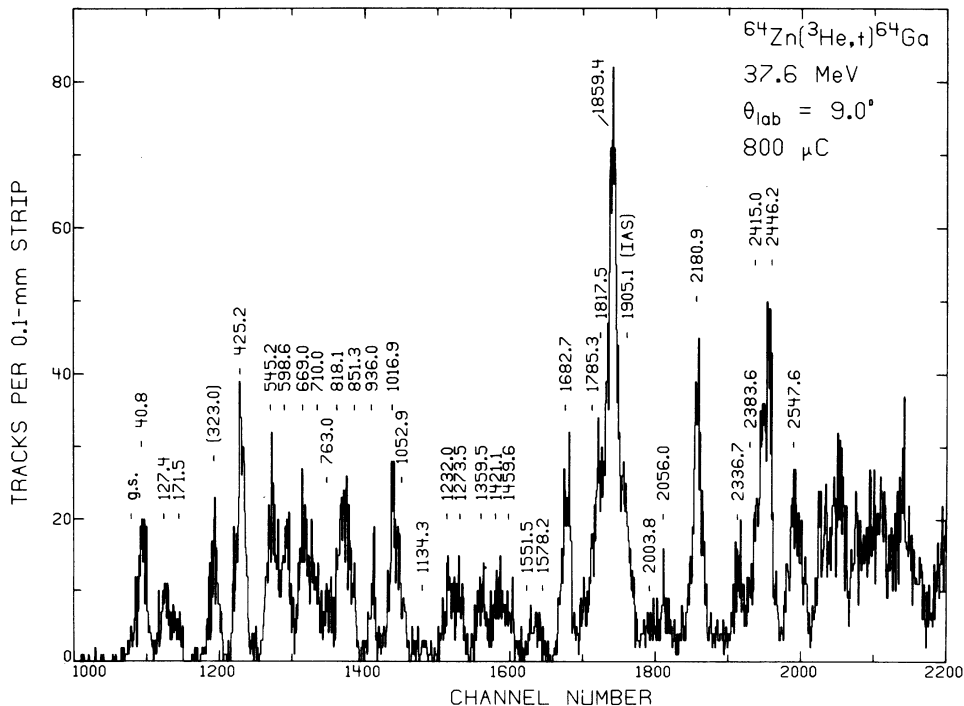


FIG. 5. Triton spectrum from  $^{64}\text{Zn}(^3\text{He}, t)^{64}\text{Ga}$  observed at  $\theta_{\text{lab}} = 9^\circ$  in the focal plane of a magnetic spectrograph.

IV. DECAY OF  $^{65}\text{Ge}$ 

The majority of the  $\gamma$  rays seen in the chemically separated sources with enriched  $^{64}\text{Zn}$  targets are assigned to the decay of  $^{65}\text{Ge}$ . The weighted average half-life for the 62.0-, 190.7-, 587.8-, and 649.8-keV lines is 30.0(12) sec. While that value is a factor of 3 shorter than the early result of Porile,<sup>13</sup> the assignment to  $^{65}\text{Ge}$  is unequivocal in the light of chemical identification, agreement of energies with levels seen in  $^{64}\text{Zn}(^3\text{He}, d)^{65}\text{Ga}$ ,<sup>24</sup>

TABLE IV. Energy levels in  $^{64}\text{Ga}$  from the  $^{64}\text{Zn}(^3\text{He}, t)-^{64}\text{Ga}$  reaction.

	Excitation energy (keV)
1	0.4(42) <sup>a</sup>
2	40.8(27)
3	127.4(21)
4	171.5(27)
5	[323.0] <sup>b</sup>
6	425.2(17)
7	545.2(20)
8	598.6(23)
9	669.0(20)
10	710.0(26)
11	763.1(33)
12	818.1(19)
13	851.3(29)
14	936.0(21)
15	1016.9(19)
16	1052.9(35)
17	1134.3(58)
18	1232.0(20)
19	1273.5(22)
20	1359.5(21)
21	1421.1(29)
22	1459.6(32)
23	1551.5(51)
24	1578.2(59)
25	1682.7(18)
26	1785.3(42)
27	1817.5(37)
28	1859.4(17)
29	1905.1(23) <sup>c</sup>
30	2003.8(42)
31	2056.0(29)
32	2180.9(17)
33	2336.7(18)
34	2383.6(42)
35	2415.0(33)
36	2446.2(19)
37	2547.6(17)

<sup>a</sup> Ground state.

<sup>b</sup> Reference peak for calculation of excitation energies.

<sup>c</sup> IAS.

and the observation of the rapid growth of the  $^{65}\text{Ga}$  daughter activity. The decay of the 650-keV line and the growth and decay of the 115-keV line from the daughter are shown in Fig. 6. Because the 62.0-keV line lies near the threshold used in the experiments with the enriched targets, its energy, intensity, and decay were measured in a separate experiment using natural Zn targets. The half-life of the 809.3-keV line (Table III) is anomalously long, which may reflect difficulties in correcting for the presence of the 808.4-keV line from  $^{64}\text{Ga}$ .

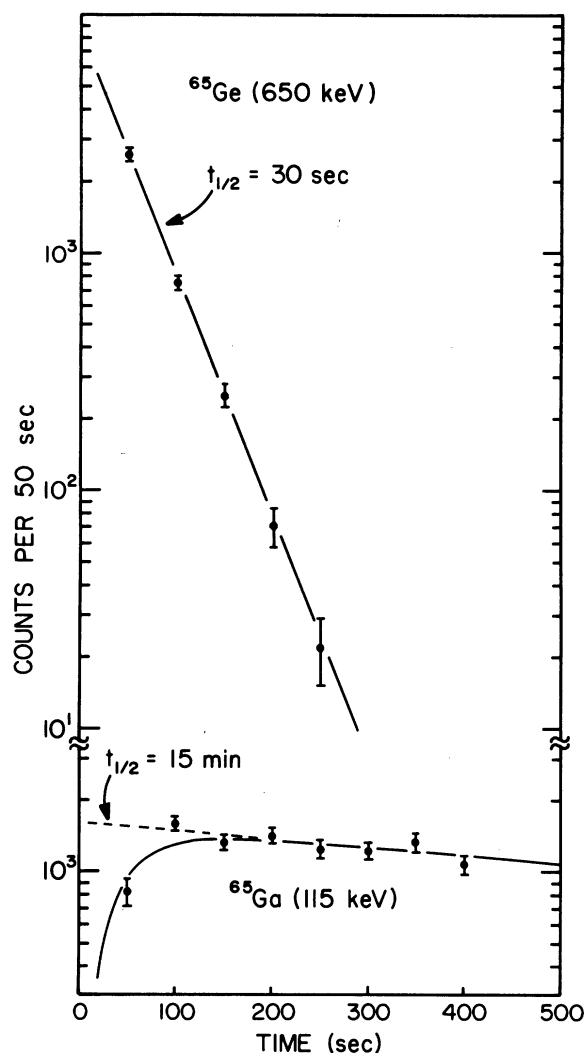


FIG. 6. Upper graph: decay of the 650-keV  $\gamma$  ray from  $^{65}\text{Ge}$ . The straight line represents the adopted half-life (30.0 sec) fitted by least squares for intensity. Lower graph: growth and decay of the 115-keV line from the daughter activity  $^{65}\text{Ga}$ . The solid curve is a calculation in which only the initial activities of the two isotopes, and not their half-lives, are fitted by least squares to the experimental points.



A total of 12  $\gamma$  rays are attributed here to  $^{65}\text{Ge}$  decay. With the exception of the 753- and 1230-keV lines, these lines and many weaker transitions (see Table III) have been observed in recently published experiments on the decay of  $^{65}\text{Ge}$  by Jongsma *et al.*<sup>25</sup> In that work, where chemical separation was not used, the 753- and 1230-keV lines are obscured by contaminants. They find, in agreement with the present result, a half-life of 31.2(7) sec for  $^{65}\text{Ge}$ .

Figure 7 shows the  $^{65}\text{Ge}$  decay scheme from the present work compared with the  $^{64}\text{Zn}(^3\text{He}, d)^{65}\text{Ga}$  spectrum. Three weak transitions observed by Jongsma *et al.*<sup>25</sup> have also been incorporated into this scheme. Absolute  $\gamma$ -ray intensities (shown in the decay scheme corrected for internal conversion assuming  $M1$  multipolarity) have been obtained by normalizing to the measured growth of  $^{65}\text{Ga}$  daughter activity. They are in good agreement with the results of Jongsma *et al.*, where normalization to the intensity of annihilation radiation was used, except that the highest-energy  $\gamma$  rays appear somewhat more intense in the present work. Because of the relatively long half-life of  $^{65}\text{Ga}$  and the low energy of its principal  $\gamma$  ray, there is somewhat greater uncertainty about the over-all normalization than in the case of  $^{64}\text{Ge}$ . In contrast to Porile,<sup>13</sup> who found  $\sim 95\%$  feed to the

ground state, the present results indicate little or no direct feed ( $< 30\%$ ) to the ground state.

For all the low-lying levels in  $^{65}\text{Ga}$  populated in the  $^{65}\text{Ge}$   $\beta^+$  decay, there exist  $l_p$  values from stripping experiments.<sup>24, 26, 27</sup> The ground, 62-, and 650-keV states are all  $l_p = 1$ . Furthermore, the  $p_{3/2}$  sum-rule limit requires at least one of these states to be  $\frac{1}{2}^-$ . Hence, the observation of allowed  $\beta^+$  decay to these and the 191-keV state ( $l_p = 3$ ) implies that the ground state of  $^{65}\text{Ge}$  is  $\frac{3}{2}^-$  and the 191-keV state in  $^{65}\text{Ga}$  is  $\frac{5}{2}^-$ . The remaining assignments shown in Fig. 7 are based largely on systematics, as discussed by several authors.<sup>24-26</sup>

#### V. $^{64}\text{Zn} + ^3\text{He}$ EXCITATION FUNCTIONS

Measurements of the excitation functions for reactions induced by  $^3\text{He}$  ions on  $^{64}\text{Zn}$  have recently been reported by Crisler *et al.*<sup>3</sup> Omitted from that extensive survey were a number of interesting reactions involving either a low production cross section or a short half-life of the product nucleus, and in particular the  $(^3\text{He}, xn)$  reactions for  $x > 1$ . Both to complement the work of Crisler *et al.* and to provide another check on the identification of  $^{64}\text{Ge}$ , coarse excitation functions for the  $(^3\text{He}, xn)$  reactions were measured

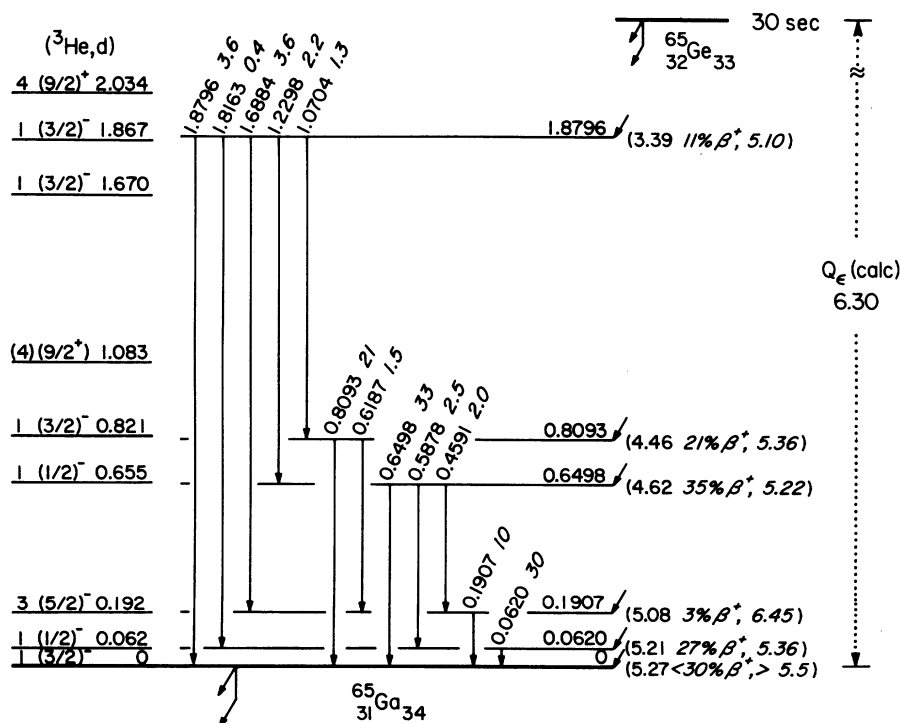


FIG. 7. Decay scheme for  $^{65}\text{Ge}$ , derived from the present work. Also included are three transitions observed by Jongsma *et al.* (Ref. 25). The notation and conventions of Nuclear Data Sheets have been adopted.

using chemical separation and enriched  $^{64}\text{Zn}$  targets. Absolute cross sections for the production of  $^{65}\text{Ge}$  and  $^{64}\text{Ge}$  were obtained at 3 energies, 48.9, 36.7, and 20.2 MeV, by normalizing to the cross sections measured by Crisler *et al.* for  $^{64}\text{Zn}(^3\text{He}, n)^{66}\text{Ge}$ . A small extrapolation of the monotonically decreasing  $(^3\text{He}, n)$  excitation function from 43 to 49 MeV was necessary.

A third reaction not observed by Crisler *et al.* is  $^{64}\text{Zn}(^3\text{He}, p2n)^{64}\text{Ga}$ , due to the short half-life of  $^{64}\text{Ga}$ . The excitation function for this process was therefore measured in a separate experiment in which 35.6-mg  $\text{cm}^{-2}$  natural zinc foils were irradiated with  $^3\text{He}$  beams degraded by zinc absorbers to energies from 68 to 29 MeV.  $\gamma$  rays were then observed with a Ge(Li) detector of calibrated efficiency. The use of natural zinc targets in principle allows the production of  $^{64}\text{Ga}$  by extraneous reactions, such as  $^{66}\text{Zn}(^3\text{He}, p4n)^{64}\text{Ga}$ . However, it is unlikely, especially at the lower energies, that the  $(^3\text{He}, p4n)$  reaction has a cross section comparable to  $(^3\text{He}, p2n)$ , which is elsewhere found to be one of the most prolific  $^3\text{He}$ -

induced reactions.<sup>28</sup> On the other hand, considerable contamination of the  $^{64}\text{Zn}(^3\text{He}, pn)^{65}\text{Ga}$  and  $^{64}\text{Zn}(^3\text{He}, \alpha)^{63}\text{Zn}$  excitation functions due to other zinc isotopes was apparent in a comparison with the data of Crisler *et al.*, and the cross sections quoted here for  $^{64}\text{Zn}(^3\text{He}, p2n)^{64}\text{Ga}$  may, therefore, contain a contribution from  $^{66}\text{Zn}(^3\text{He}, p4n)^{64}\text{Ga}$  above the threshold for that reaction at 36 MeV.

The excitation functions for two- and three-nucleon emission reactions are summarized in Figs. 8 and 9, respectively. The small magnitude of the  $(^3\text{He}, 2n)$  cross section relative to the others is very striking. The fact that  $(^3\text{He}, 2p)$  and  $(^3\text{He}, np)$  reactions can proceed through direct channels while the  $(^3\text{He}, 2n)$  reaction cannot to first order, may explain part of the effect. It should be noted that the small magnitude of the

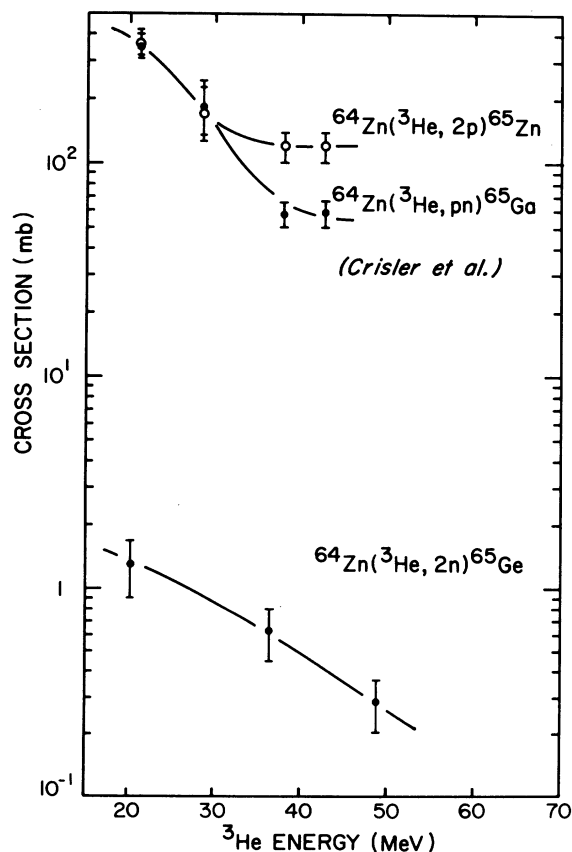


FIG. 8. Excitation functions for  $^3\text{He}$ -induced reactions on  $^{64}\text{Zn}$  resulting in emission of two nucleons. The  $(^3\text{He}, pn)$  and  $(^3\text{He}, 2p)$  results are from Ref. 3.

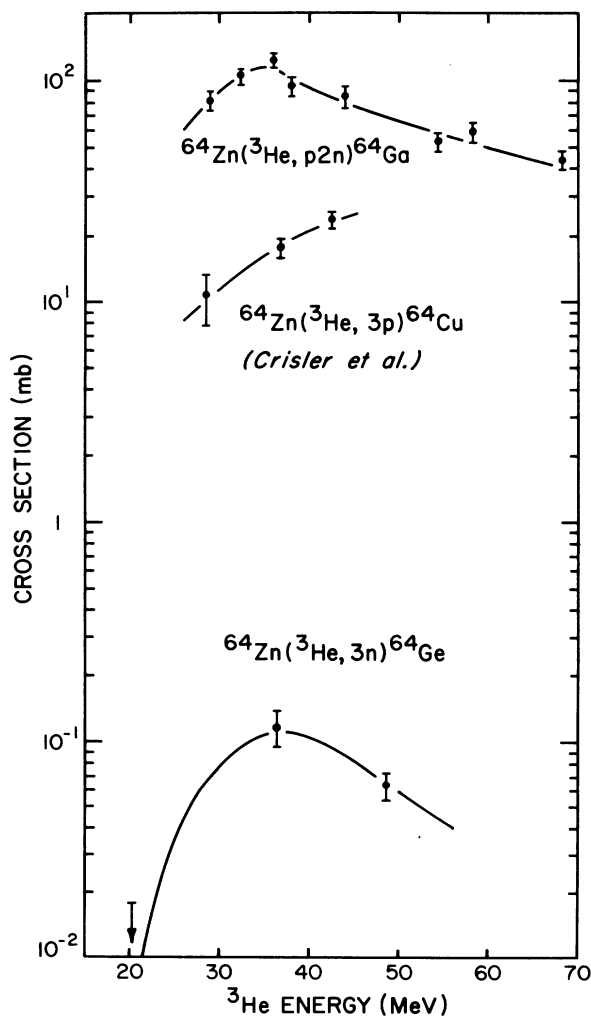


FIG. 9. Excitation functions for  $^3\text{He}$ -induced reactions on  $^{64}\text{Zn}$  resulting in emission of three nucleons. The  $(^3\text{He}, 3p)$  results are from Ref. 3.

( $^3\text{He}, 2n$ ) reaction means that essentially all the  $^{65}\text{Ga}$  detected by Crisler *et al.* resulted from the ( $^3\text{He}, np$ ) process with little contribution from the decay of  $^{65}\text{Ge}$ .

The relative cross sections for the ( $^3\text{He}, 3n$ ), ( $^3\text{He}, p2n$ ), and ( $^3\text{He}, 3p$ ) reactions also show striking differences. In this case, the small magnitude of the ( $^3\text{He}, 3n$ ) cross section may reflect both the small number of bound states in even-even  $^{64}\text{Ge}$  compared to the odd-odd isobars  $^{64}\text{Ga}$  and  $^{64}\text{Cu}$ , and the increasing binding energy of neutrons as one moves toward proton-rich nuclei.

The identification of  $^{64}\text{Ge}$  is further confirmed by the measured excitation function, which is consistent with the calculated threshold of 20 MeV. Production of  $^{64}\text{Ge}$  was observed at 29 MeV, but the absolute cross section was not determined at that energy.

#### VI. CONCLUSION

The principal objective of the work described has been to establish the existence of  $^{64}\text{Ge}$  and ascertain its half-life and decay modes. Conclusive identification of 64-sec  $^{64}\text{Ge}$  and 30-sec  $^{65}\text{Ge}$  has been obtained through (a) chemical separation of germanium, (b) observation of  $\gamma$  rays between known states in the daughter nuclei, (c) observation of the growth of the daughter activities, and (d) measurement of excitation functions. The role of  $^{64}\text{Ge}$  in nucleosynthesis has been extensively discussed elsewhere.<sup>1, 6, 7</sup> The calculations of Arnett, Truran, and Woosley<sup>1</sup> of explosive carbon burning indicate that elements above the iron peak can be synthesized via an  $\alpha$ -capture chain based on  $^{56}\text{Ni}$ . The abundance of mass 60 produced via  $^{56}\text{Ni}(\alpha, \gamma)^{60}\text{Zn}$  is very close to the actual solar-system abundance, whereas the subsequent  $^{60}\text{Zn}(\alpha, \gamma)^{64}\text{Ge}$  process falls short of accounting for the abundance of mass 64 by two orders of magnitude.

However, the final abundance of mass 64 so produced depends sensitively on the binding energy of  $^{64}\text{Ge}$ . The original calculations of ATW made use of the mass excess calculated by Garvey *et al.*<sup>2</sup> (-54.03 MeV). Recently, Davids and Goosman<sup>7</sup> have measured the mass excess of  $^{64}\text{Ge}$  by finding the endpoint of the  $\beta^+$  branch to the 427-keV state in  $^{64}\text{Ga}$ . Their result for the mass excess, -54.43(25) MeV, implies that as much as 10 to 20% of the solar-system abundance of  $^{64}\text{Zn}$  may originate from the process proposed by ATW.

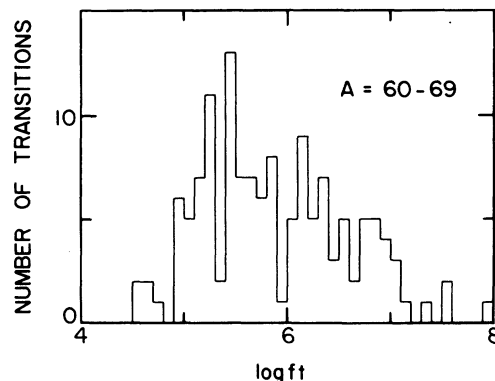


FIG. 10 Distribution of  $\log ft$  values found in  $\beta^+/\epsilon$  decays of neutron-deficient isotopes in the mass-60-69 region.

The uncertainty in the mass measurement of Davids and Goosman still admits an order-of-magnitude range of final abundance of  $^{64}\text{Zn}$ , notwithstanding other uncertainties in the calculation. While such measurements are difficult to carry out accurately, it seems clear that in this case more accuracy is essential before the question can be finally resolved. It appears unlikely that  $^{64}\text{Ge}$  can be much more tightly bound than the median result of Davids and Goosman, because the  $\log ft$  value for the  $\beta^+$  transition to the 427-keV state is (from their measurement) 4.53(17), already a very low value for this region of the nuclidic table. Figure 10 shows the distribution of  $\log ft$  values found in a survey of 139 transitions in the decays of  $^{60-63}\text{Zn}$ ,  $^{64-68}\text{Ga}$ , and  $^{65-69}\text{Ge}$ . Only two other transitions are known in this region with  $\log ft$  as low as 4.5, one in  $^{60}\text{Zn}$  and one in  $^{66}\text{Ge}$ . Such arguments are hardly conclusive, but serve rather to illustrate the need for more definitive measurements. Until they are forthcoming, it would appear that the  $\alpha$ -capture chain proposed by ATW is highly successful in explaining the abundance of mass 60, but that other mechanisms must be invoked to explain most of the mass-64 abundance.

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