In order to determine  $\Gamma_0$  and  $\delta$  separately, a selfabsorption experiment was attempted. However, the resonant absorption was too small to make statistically significant measurements. Using a nickel absorber (natural abundance) of thickness 1.25 cm, the upper limit of the self-absorption ratio was estimated to be about 2%. In the experiment of Ref. 3, the temperature variation of the scattering cross section was measured to determine  $\delta = 11$  eV. Combining this value with their elastic-scattering cross section  $\langle \sigma_{\gamma\gamma} \rangle = 375$  mb, Giannini et al. obtained  $\Gamma_0 = (150 \pm 20)$  mV. On the other hand, if  $\delta = 11$  eV is used together with our measured cross section,  $\langle \sigma_{\gamma\gamma} \rangle = 190$  mb,  $\Gamma_0 = 74$  mV is obtained. This value of the radiation width is consistent with the fact that the value of  $\langle \sigma_{\gamma\gamma} \rangle$  of this experiment is smaller than the value of Ref. 3 by a factor of 2, while the two values of the branching ratio,  $\Gamma_0/\Gamma$  are in good agreement. (See Secs. III D2 and III D3.) With  $\delta = 11 \text{ eV}$ and  $\langle \sigma_{\gamma\gamma} \rangle = 190$  mb of this experiment, one would expect a self-absorption ratio of less than 1%.

## III. DISCUSSION

The results have shown the feasibility of studying the spectrum of the cascade photons coincident with inelastically scattered photons from a highly excited nuclear level. By studying the spectrum of the cascade photons, the isotope responsible for the resonant

scattering can be identified even when the separated isotopes are not used as targets. Furthermore, the coincidence spectrum yields information about the structure of lower lying levels. In this work, the resonant scattering of the iron capture gamma radiation is attributed to the 7.64-MeV J=1 level in Ni<sup>62</sup>. In addition to the dominant elastic scattering, two inelastically scattered photons populate the 1.17-MeV and 2.05-MeV levels. The 2.05-MeV level is known to be a member of the two phonon vibrational triplet. It is interesting to note that the other two members of the triplet, the 2<sup>+</sup> and 4<sup>+</sup> levels observed at 2.302 MeV and 2.336 MeV, respectively in other reactions,<sup>6</sup> are not observed in this experiment, even though the energies are well separated from the 2.05-MeV level. It is not difficult to understand why the 4<sup>+</sup> level was not seen. Since the 7.64-MeV resonant level has J=1, the transition to the  $4^+$  level requires either E3 or M3, while all three observed transitions are most likely dipole. Therefore, it would have been difficult to observe the transition to the 4<sup>+</sup> level. However, no such restriction exists for the transition to the 2<sup>+</sup> level. The absence of this transition seems to indicate that the wave functions of the 2.05-MeV and 2.302-MeV levels have different admixtures of the component which can couple to the 7.64-MeV J=1 level through the dipole operator.

PHYSICAL REVIEW

VOLUME 154, NUMBER 4

20 FEBRUARY 1967

# Activation Cross Sections of Germanium for 14.4-MeV Neutrons\*

R. E. WOOD, † W. S. COOK, ‡ J. R. GOODGAME, ‡ AND R. W. FINK School of Chemistry, Georgia Institute of Technology, Atlanta, Georgia (Received 4 October 1966)

Ge<sup>89,75</sup>, Ga<sup>72,73,74</sup>, and Zn<sup>69m,71m</sup> were produced by irradiating natural germanium with 14.4±0.3-MeV neutrons. Two mixtures of powders containing GeO2+Al+Fe and GeO2+Al+Cu also were irradiated. The activities were identified from gamma spectra taken with a Ge(Li) detector and the cross sections were measured using the improved technique described by Rao and Fink. The reactions for which cross sections were measured are:  $\hat{\operatorname{Ge}}^{70}(n,2n)\operatorname{Ge}^{69}$ ,  $\hat{4}47\pm45$  mb;  $\hat{\operatorname{Ge}}^{76}(n,2n)\operatorname{Ge}^{75}$ ,  $1236\pm120$  mb;  $\operatorname{Ge}^{72}(n,p)\operatorname{Ga}^{72}$ ,  $47.4\pm4.7$ mb;  $\operatorname{Ge}^{73}(n,p)\operatorname{Ga}^{73}$ , 26.4 $\pm$ 2.6 mb;  $\operatorname{Ge}^{74}(n,p)\operatorname{Ga}^{74}$ , 13.2 $\pm$ 1.3 mb;  $\operatorname{Ge}^{72}(n,\alpha)\operatorname{Zn}^{69m+g}$ , 15.2 $\pm$ 1.5 mb; and  $\operatorname{Ge}^{74}(n,\alpha)\operatorname{Zn}^{71m+g}$ , 13.3±1.3 mb. For all cases where isomers exist, the cross-section value is total for m+g. The  $Ge^{76}(n,2n)Ge^{75}$  (82-min) cross section also was checked by beta counting, which is independent of decay-scheme assumptions. The results are compared with the systematics of (n,2n), (n,p), and  $(n,\alpha)$  cross sections at 14-15 MeV.

## INTRODUCTION

**TEUTRON-ACTIVATION** cross sections of germanium have been measured at  $14.4\pm0.3$  MeV as part of a systematic study being carried out for comparision with cross-section systematics for 14-15-MeV neutron reactions. In view of the increasing use of Ge(Li) detectors for prompt-gamma studies in fast-neutron reactions, a knowledge of the cross sections for neutron reactions in germanium is also of interest. A comparison of the present results with earlier studies appears in the discussion section.

#### EXPERIMENTAL

Monoenergetic neutrons are produced by the H<sup>3</sup> (d,n)He<sup>4</sup> reaction in thick titanium-tritium targets on the Georgia Tech 200 kV accelerator. Cross sections were measured using an improved technique developed

<sup>\*</sup> Supported in part by the U. S. Atomic Energy Commission. † Present address: Emory University, Atlanta, Georgia. ‡ Supported in part by the U. S. Public Health Service through a grant to Emory University.

by Rao and Fink.<sup>1</sup> One-gram samples of spectroscopically pure (99.9999+%) natural germanium were irradiated with an exponentially decaying flux of about  $10^9-10^{10}$  neutrons/cm<sup>2</sup> sec for 50 min, typically. Activities of Ge<sup>69,75</sup>, Ga<sup>72,73,74</sup>, and Zn<sup>69m,71m</sup> were identified from the gamma spectra taken with a 2 mm×1 cm<sup>2</sup> Ge(Li) detector having a resolution of 4 keV FWHM (full width at half-maximum). The photopeak efficiency of the detector was calibrated with standard gammas relative to a 3×3-in. NaI (Tl) crystal. The energies were calibrated with gammas from Na<sup>22</sup>, Mn<sup>54</sup>, Co<sup>60</sup>, Se<sup>75</sup>, and Cs<sup>137</sup>.

The relative yield A of a given product activity was measured from the equation

$$A = A'/\epsilon f_s f_d, \qquad (1)$$

where  $\epsilon$  is the photopeak detection efficiency in the Ge(Li) detector,  $f_s$  is the correction factor for source self-absorption, and  $f_d$  is the number of gammas of given energy emitted per disintegration of the product nucleus. Values of  $f_d$  used in this work are listed in Table I. The activity extrapolated to the end of bombardment  $A^0$  is obtained by following the decay of the gamma spectra. The cross section is then given by

$$\frac{\sigma}{\sigma_m} = \frac{A^0 n_m \lambda_m (\lambda - \Delta) (e^{-\Delta T} - e^{-\lambda m T})}{A_m^0 n \lambda (\lambda_m - \Delta) (e^{-\Delta T} - e^{-\lambda T})}, \qquad (2)$$

where the subscript *m* stands for the monitor, *n* is the total number of target nuclei in the irradiated sample,  $\lambda$  is the decay constant of the activity produced, and  $\Delta$  is the *measured* decay constant of the neutron flux during the irradiation time *T*. (In these experiments,  $\Delta$  was found to vary with different Ti-T targets, with beam current, and with the degree of focusing. The target half-lives were found to be as short as 35 min or as long as 4.5 h, indicating the necessity for determining flux half-life in each individual irradiation.)

Samples of about 1 g or smaller, consisting of mixed powders<sup>1</sup> were packed into plastic source holders 3 mm

TABLE I. Summary of germanium cross sections for  $14.4\pm0.3$  MeV neutrons.

Reaction	$E_{\gamma}$ (keV)	fa	σ (mb)	
$\frac{\text{Ge}^{76}(n,2n)\text{Ge}^{75}}{\text{Ge}^{76}(n,2n)\text{Ge}^{75}} (82.2 \text{ min})$	265 from beta	0.110 1.0(β <sup></sup> )	$\begin{array}{rrr} 1236 & \pm 120 \\ 1444 & \pm 289 \end{array}$	
$Ge^{72}(n,p)Ga^{72}$ (14.69 h) $Ge^{73}(n,p)Ga^{73}$ (4.85 h) $Ge^{74}(n,p)Ga^{74}$ (8 min)	counting 832 297 595	$0.754 \\ 0.970 \\ 0.767$	$47.4 \pm 4.7$ $26.4 \pm 2.6$ $13.2 \pm 1.3$	
$Ge^{72}(n,\alpha)Zn^{69m}$ (13.8 h) $Ge^{72}(n,\alpha)Zn^{69m+g}$	437	0.943	$7.61 \pm 0.76$ $15.2 \pm 1.52$	
${ m Ge}^{74}(n, \alpha) { m Zn}^{71m}$ (4 h) ${ m Ge}^{74}(n, \alpha) { m Zn}^{71m+g}$	384 485	1.0 1.0 }	$3.32 \pm 0.33$ $13.3 \pm 1.33$	

\* See discussion in the text.

<sup>1</sup> P. Venugopala Rao and R. W. Fink, this issue, Phys. Rev. 154, 1023 (1967).

TABLE II. Summary of cross-section measurements with  $14.4\pm0.3$  MeV neutrons in irradiations of mixtures of powders.

Mixture	Reactions	$E_{\gamma}$ (keV)	fa	σ (mb)
Using gamma p	hotopeaks:			
GeO <sub>2</sub> +Al+Fe	$Ge^{70}(n,2n)Ge^{69}$ (35.5 h) Al <sup>27</sup> $(n,\alpha)Na^{24}$ (15 h)	574 1368	0.119 1.0	$448 {\pm} 45$ 113 {\pm} 11
GeO <sub>2</sub> +Al+Cu	$Ge^{70}(n,2n)Ge^{69}$ (35.5 h)	574	0.119	$445{\pm}45$
Using positron a	unnihilation radiation:			
GeO <sub>2</sub> +Al+Cu	$Ge^{70}(n,2n)Ge^{69}$ (35.5 h)	511	0.35	$648 \pm 100$

in depth and 1 cm<sup>2</sup> in area. The samples were placed about 2 mm behind the Ti-T target and subtended an angle of  $\pm 65^{\circ}$  to the beam direction, so that about 92% of the neutrons irradiating the sample had energies within the range 14.4 $\pm$ 0.2 MeV, the remaining 8% being within  $\pm$ 0.3 MeV of 14.4 MeV.

All cross sections were measured relative to the internal monitor reaction  $Ge^{70}(n,2n)Ge^{69}$  (35.5 h). Several absolute measurements of this cross section were made. First, by using the technique of Rao and Fink,<sup>1</sup> mixtures of  $GeO_2$ +Al+Fe powders in a weight ratio of 1:1:1 and of GeO<sub>2</sub>+Al+Cu powders in a weight ratio of 3:1:1 were irradiated and studied with the Ge(Li) detector to give the results shown in Table II. For the first mixture, the monitor reaction  $Fe^{56}(n,p)Mn^{56}$ (2.58 h),  $\sigma = 100 \pm 6$  mb, was used as a standard, while for the second mixture the reaction  $Al^{27}(n,\alpha)Na^{24}$  (15 h),  $\sigma = 114 \pm 7$  mb was taken as a standard. The consistency of the results in Table II indicates that the mixing was thorough. The result thus obtained,  $Ge^{70}(n,2n)Ge^{69}$ ,  $\sigma \approx 450$  mb, is not consistent with the literature value of  $610\pm37$  mb.<sup>2</sup> An effort was made therefore to determine this cross section another way. The second mixture was used as a source of annihilation radiation, since Ge<sup>69</sup> and Cu<sup>64</sup> are positron emitters. A 1-mm-thick lead absorber was placed between the source and detector to stop all of the positrons. However, since the  $\beta^+$  endpoints are not identical in these two decays, the annihilation radiation source volume is different for the two cases. This means that the detector efficiency is not the same in the two decays. Taking a literature value<sup>2</sup> for  $Cu^{65}(n,2n)$  $Cu^{64}$  (12.87 h) cross section of 940 $\pm$ 80 mb and assuming a ratio  $\beta^+/(K+\beta^+)$  of 0.19 for Cu<sup>64</sup> and 0.35 for Ge<sup>69</sup>, a value of  $648 \pm 100$  mb for the Ge<sup>70</sup>(n,2n)Ge<sup>69</sup> reaction cross section was obtained, which is in better agreement with the literature.<sup>2</sup> These results are taken to mean that the Ge<sup>69</sup> decay scheme is in need of further study.

In order to check the  $Ge^{76}(n,2n)Ge^{75}$  (82 min) cross section found by gamma spectroscopy, which is dependent upon the absolute intensity of the gammas in the decay scheme and hence upon the intensity assumed for the ground-state beta transition in  $Ge^{75}$  decay, this reaction was also measured by means of beta counting, which is decay scheme independent. An end-window

<sup>2</sup> P. Jesson, M. Bormann, F. Dreyer, and H. Neuert, Nucl. Data 1, 103 (1966).

and

 $(0.9 \text{ mg/cm}^2 \text{ aluminized Mylar})$  methane-flow beta proportional counter was employed to determine the yield of Ge<sup>75</sup> formed in this reaction. Applying the corrections for geometry, saturation backscattering, source self-scattering-self-absorption, window absorption, etc. for absolute beta counting, a value for the  $Ge^{76}(n,2n)$ Ge<sup>75</sup> cross section of  $1444 \pm 289$  mb was obtained.

Since only the metastable states were detected in  $Zn^{69m,71m}$ , the following cross-section ratios, taken from Levkovskii,<sup>3</sup> were used to obtain the total activation cross sections:

$$\sigma \left[ \operatorname{Ge}^{74}(n,\alpha) \operatorname{Zn}^{71m} \right] / \sigma \left[ \operatorname{Ge}^{74}(n,\alpha) \operatorname{Zn}^{71g} \right] = \frac{1}{3},$$

$$\sigma \left[ \operatorname{Ge}^{72}(n,\alpha) \operatorname{Zn}^{69m} \right] / \sigma \left[ \operatorname{Ge}^{72}(n,\alpha) \operatorname{Zn}^{69g} \right] = 1.$$

Conversion coefficients and branching ratios used in calculating the  $f_d$  values were taken in general from Nuclear Data Sheets,<sup>4</sup> except for Ge<sup>69</sup>. Here only the ground-state decay branching (47%) was taken from Nuclear Data Sheets. The decay scheme was constructed from the Ga<sup>69</sup> level scheme of Temperley, McDaniels, and Wells<sup>5</sup> as studied in the decay of Ge<sup>69</sup>.

### DISCUSSION

The  $Ge^{76}(n,2n)Ge^{75}$  cross section (Table I) is significantly higher than would be predicted from the systematics of Bormann.<sup>6</sup> The value is equal approximately to the total nonelastic cross section (Ref. 6). A maximum is expected in this mass-number region due to shell effects on the level density.<sup>6</sup> It is of interest that the  $Ge^{76}(n,2n)Ge^{75}$  cross section (1236 mb) is much larger than the value for the  $Se^{76}(n,2n)Se^{75}$  reaction (808 mb) determined by Rao and Fink.<sup>1</sup> The difference might be attributed to the fact that the neutron excess (A-2Z)in Ge<sup>76</sup> is 12, whereas it is only 8 in Se<sup>76</sup>. Bormann's (n,2n) systematics,<sup>6</sup> in which log  $\sigma$  is plotted against mass number for even-even nuclei, does not take into account possible effects due to pairing-energy differences.

The (n, p) cross sections exhibit a typical Levkovskii trend.<sup>7</sup> The cross section for  $Ge^{74}(n,p)Ga^{74}$  is a factor of 2 lower than the systematics of Chatterjee,<sup>8</sup> but a factor of 2 higher than the theoretical prediction of Gardner.<sup>9</sup> Paul and Clark<sup>10</sup> previously measured by beta counting a cross section of 65.5 mb for the  $Ge^{72}(n,p)Ga^{72}$  reaction and 136 mb for the  $Ge^{73}(n,p)Ga^{73}$  reaction at 14.5 MeV.



FIG. 1. A replot of Chatterjee's  $(n,\alpha)$  systematics (Ref. 11) in the region  $29 \leq Z_R \leq 35$ . The Zn values are from the present work, and the Ge value is from Rao and Fink (Ref. 1).

These values are higher than the present results and run contrary to the Levkovskii trend.

Figure 1 is a plot of  $(n,\alpha)$  cross sections versus residual nuclear charge  $Z_R$  in the region  $29 \leq Z_R \leq 35$ . Chatterjee<sup>11</sup> suggested that a dip exists in the  $(n,\alpha)$  cross sections in this region because of subshell structure effects. The present results for  $(n,\alpha)$  reactions in this region are shown in Fig. 1 and substantiate the existence of such a minimum, but much more pronounced than Chatterjee projected. Work is continuing in this region to define this effect more fully. The only previous measurement of  $(n,\alpha)$  cross section in germanium is the value of 14.9 mb obtained by beta counting for the reaction Ge<sup>74</sup>- $(n,\alpha)$ Zn<sup>71</sup> by Paul and Clarke.<sup>10</sup> This value agrees with the present result in Table I.

The error limits given in Table I include errors in statistics, detector efficiency, source self-absorption, neutron flux decay during irradiation, half-lives, and the value of the monitor reaction cross section, but do not include those in the decay schemes.

#### ACKNOWLEDGMENTS

The authors wish to thank Mrs. Ann Galli for assistance with the 200-kV accelerator and Robert Harbort for help in processing some of the data at the Emory University Biomedical Data Processing and Analysis Center. We also wish to thank Dr. P. Venugopala Rao for many helpful discussions.

<sup>&</sup>lt;sup>3</sup> V. N. Levkovskii, Zh. Eksperim. i Teor. Fiz. **33**, 1526 (1957) [English transl.: Soviet Phys.—JETP **6**, 1174 (1958)]. <sup>4</sup> Nuclear Data Sheets, compiled by K. Way et al. (Printing and

Publishing Office, National Academy of Sciences—National Re-search Council, Washington 25, D. C.).

<sup>&</sup>lt;sup>6</sup> J. K. Temperley, D. K. McDaniels, and D. O. Wells, Phys. Rev. **139**, B1125 (1965).

<sup>&</sup>lt;sup>6</sup> M. Bormann, Nucl. Phys. **65**, 257 (1965) [(*n*,2*n*) systematics]. <sup>7</sup> V. N. Levkovskii, Zh. Eksperim. i Teor. Fiz. **31**, 360 (1956) Evalish transl. Societ Phys. JETER **4**, 201 (1977)] [English transl.: Soviet Phys.—JETP 4, 291 (1957)]. <sup>8</sup> A. Chatterjee, Nucl. Phys. **60**, 273 (1964) [(n, p) systematics]. <sup>9</sup> D. G. Gardner, Nucl. Phys. **29**, 373 (1962); **35**, 303 (1962). <sup>10</sup> E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).

<sup>&</sup>lt;sup>11</sup> A. Chatterjee, Nucl. Phys. 49, 686 (1963); 47, 511 (1963)  $[(n,\alpha) \text{ systematics}]$