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THE DETERMINATION OF (γ, n) AND $(\gamma, 2n)$ CROSS SECTIONS
IN ^{70}Ge , ^{72}Ge , ^{74}Ge , AND ^{76}Ge

Ph.D. Thesis Submitted to Iowa State University,
November, 1972

H. J. Vander Molen

Ames Laboratory, USAEC
Iowa State University
Ames, Iowa 50010

Date of Manuscript: November, 1972

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION
DIVISION OF RESEARCH UNDER CONTRACT NO. W-7405-eng-82

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cross sections in ^{70}Ge , ^{72}Ge , ^{74}Ge , and ^{76}Ge

by

Harold John Vander Molen

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of
The Requirements of the Degree of
DOCTOR OF PHILOSOPHY

Department: Physics

Major: Nuclear Physics

Approved:

____ R. C. Morrison _____

____ D. J. Zaffarano _____
In Charge of Major Work

For the Major Department

For the Graduate College

Iowa State University
Ames, Iowa

1972

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The determination of (γ, n) and $(\gamma, 2n)$
cross sections in ^{70}Ge , ^{72}Ge , ^{74}Ge , and ^{76}Ge

Harold John Vander Molen
Under the supervision of R. C. Morrison and D. J. Zaffarano
From the Department of Physics
Iowa State University

Cross section for the (γ, n) and $(\gamma, 2n)$ reactions of ^{70}Ge , ^{72}Ge , ^{74}Ge , ^{76}Ge , have been measured from threshold to 40 MeV. One gram separated isotope targets of the four nuclides were bombarded with bremsstrahlung from the Iowa State University 70 MeV electron synchrotron. The photo-neutrons emitted during bombardment were counted by a high-efficiency 4π neutron-sensitive scintillation detector. The neutron multiplicities were separated by an on-line event-by-event recording system. Reaction yields were obtained at bremsstrahlung end point energies between 10 and 40 MeV at 2 MeV intervals, and cross sections were extracted from the resulting yield curves. Resonances were observed at about 17 MeV in the (γ, n) reactions, and at about 26 MeV in the $(\gamma, 2n)$ reaction, for all four nuclides. In addition, a secondary resonance in the (γ, n) reaction was observed at 35 MeV in

^{70}Ge , 34 MeV in ^{72}Ge , 31 MeV in ^{74}Ge , and 25 MeV in ^{76}Ge .

These results, along with the (γ, p) and (γ, np) cross sections measured in parallel experiments, will be used to study the systematics of the giant resonance structure of germanium, as a function of neutron excess.

CHAPTER 1. INTRODUCTION

The electromagnetic interaction is a unique means of probing the structure of nuclear states. While Coulomb excitation and gamma de-excitation have provided much of our knowledge of discrete particle stable energy levels, at excitations above the nucleon separation energy, a continuum region is entered. This continuum region is still imperfectly understood.

A dominant feature of the continuum region is the giant dipole resonance. In heavier nuclei, the giant resonance is understood as a collective oscillation, with the resonance frequency determined by the dimensions, shape, and mass of the nucleus. In light nuclei, the giant resonance may be accounted for as the absorption of a photon to excite a single nucleon from its ground state configuration to a highly excited state prior to ejection.

Photonuclear experiments usually fall into three general classes. The methods are total absorption, activation, and measurement of particle energy spectra.

Total absorption work is limited by the fact that nuclear absorption cross sections are at most 10% of the total absorption. The atomic absorption cross section must be very well known in order to extract good nuclear data from such experiments.

In activation work, the partial cross section for production of a particular daughter nucleus is measured by counting the radioactive decays of the product nuclei. This method gives information on both the photon absorption cross section and the decay of the dipole state. It is often possible to sort out several reactions simultaneously from a multi-isotopic target by separating the various activities by half life or some other characteristic of the decay radiation. However, this method will not work on reactions which lead to stable daughter nuclei or to nuclei whose half lives are very short or very long.

Measurements may be made of the energy spectra of the emitted particles. This type of work is very useful for light target nuclei, where proton emission channels are strong, and the density of states in the residual nuclei are small. For heavier nuclei, neutron emission dominates and the final state density is high. Such experiments become difficult to perform and difficult to interpret.

Relatively little work has been done on (γ, n) reactions by directly counting the emitted neutrons. The work that has been done usually has no experimental means for separating the $(\gamma, 2n)$ reactions from the (γ, n) reactions. Moreover, there has been almost no work on $(\gamma, 2n)$ reactions, even by means of the activation technique. This experiment is a study of (γ, n) and $(\gamma, 2n)$ reactions in four isotopes of

germanium using a large neutron sensitive scintillation detector to count the photoneutrons directly, and isotopically separated material for targets.

Motivation

The selection rules for electric dipole absorption are $\Delta\pi=1$, $\Delta J=0,1$ (0-0 forbidden), and $\Delta T=0,1$ (0-0 forbidden). The isobaric spin quantum number has only recently become of interest in photoabsorption. The isobaric spin formalism, also called isotopic spin or simply isospin, has been used successfully in light ($A < 20$) nucleus calculations. It has always been assumed that the large Coulomb potentials in medium and heavy nuclei would result in sufficient configuration mixing to destroy the isospin identity of the various levels.

In 1961, isobaric analog resonances were identified in (p,n) reactions for target nuclei as heavy as mass 93 (1). Such results suggest that the isobaric spin formalism has a much greater range of validity than was expected, and confirmation has been found in a large variety of nuclear excitations.

However, the validity of the isospin formalism has not yet been firmly established for photonuclear reactions. Electric dipole absorption can excite two giant resonance states in the compound nucleus. If the target nucleus has isospin T_0 ($=T_{03} = (N-Z)/2$), then the lower giant resonance

state has $T=T_0$ and can decay by proton emission (to $T=T_0+\frac{1}{2}$) or by neutron emission (to $T=T_0-\frac{1}{2}$). The upper giant resonance state has $T=T_0+1$. Proton emission is allowed, but neutron emission (to $T_0-\frac{1}{2}$) is isospin forbidden because the decay involves $\Delta T=3/2$. (It should be noted, however, that neutron emission to an excited state with $T=T_0+\frac{1}{2}$ is allowed.) Therefore, both giant resonance states should decay by proton emission, but the lower $T=T_0$ state should dominate the $T=T_0+1$ state in neutron emission.

Efforts have been made to verify the splitting of the giant resonance predicted by the isospin formalism. The most direct approach is to compare (γ, p) or (p, γ) cross sections with (γ, n) cross sections in the same nuclei (2). However, such comparisons are difficult because the absolute normalization of photonuclear cross sections is seldom very accurate.

Attempts have also been made to measure the T_0 component of the proton channel directly (3, 4). While the splitting was indeed observed, the strengths were found to be less than predicted. A possible reason for the lack of strength in the proton channel is the availability of other isospin allowed channels, e.g. by neutron emission to an excited state in the daughter nucleus.

Schamber (5) measured the (γ, n) , (γ, np) , and $(\gamma, 2n)$ cross sections of ^{64}Zn by means of the activation technique.

The proton channel has been investigated by Paul et al. (6), who studied the $^{63}\text{Cu}(p,\gamma)^{64}\text{Zn}$ reaction and used the principle of detailed balance to infer the (γ,p) cross section, and recently by Clark (7), who studied the (γ,p) reaction by measuring the spectra of the photoprotons at several bombarding energies. The results seemed to confirm the general predictions of the isospin theory.

Another way to examine the isospin of the dipole state would be to study several nuclides with varying N or Z, and look for systematics in the giant resonance structure. The isospin splitting of the resonance is sensitive to the neutron excess, and systematic splitting consonant with the prediction of the isospin formalism would constitute strong evidence of the validity of the isospin theory. The measurement reported here is one of a series of three experiments designed to measure the n, p, np, and 2n exit channels for four isotopes of germanium. By including all of the major exit channels, it should be possible to identify conclusively and measure the strengths of the two resonances considerably more definitively than could be done in previous experiments.

In addition to the possibility of observing isospin effects, there are further reasons for examining photonuclear reactions in the general mass region of germanium. The anomalously large (γ,n) cross section of ^{60}Ni , relative to that of ^{58}Ni , is well known, as is also the structure in the

giant resonance (8, 9). Copper (10) and zinc (5) have also been studied, and there is evidence of structure in these elements also. There is very little data for elements heavier than zinc. ^{75}As is the only other nucleus in this region which has been studied extensively, since it is the only monoisotopic element in the region. Moreover, there is evidence of structure in ^{75}As also (11). In summary, because of the difficulty of experiments on separated isotopes, the mass region around germanium has not been very well studied, but the available data suggest that structure may exist which current theories do not explain (8, 9, 10, 11).

Plan of Experiment

The overall plan of the series of experiments is shown in Table 1. In the activation method, targets of natural germanium are used and spectra of the residual activity of the daughter nuclei are measured with Ge(Li) detectors. The various reactions are then sorted out by means of the gamma spectra of the radioactive daughter nuclei. The np channels of all four isotopes, the proton channel of ^{74}Ge (and possibly of ^{76}Ge), and the neutron channels of ^{70}Ge and ^{72}Ge can all be studied by this technique.

Two of the remaining channels can be studied using ^{69}Ga and ^{71}Ga targets in a (p, γ) reaction. The (γ, p) cross sections can then be calculated by using the principle of detailed balance.

Table 1. Plan of experiments^a

Reaction	⁷⁰ Ge	⁷² Ge	⁷⁴ Ge	⁷⁶ Ge
γ, n	A, N	N	N	A, N
γ, p	P	P	A	A?
γ, np	A	A	A	A
$\gamma, 2n$	N	N	N	N

^a Reactions which are studied in this experiment by direct counting of neutrons are designated by the letter N. Reactions which will be studied by the activation method in the companion experiment are designated by the letter A, and those which can be studied by (p, γ) reactions by the letter P.

The remaining (γ, n) and $(\gamma, 2n)$ cross sections are the subject of the present experiment. Measurement of these cross sections is not possible by activation techniques, and can be accomplished only by direct neutron counting. The reactions were studied using targets fabricated from isotopically separated germanium. The targets were exposed to bremsstrahlung produced by the Iowa State University 70 MeV electron synchrotron, and the photoneutrons were counted directly by a high efficiency neutron sensitive scintillation detector.

In Chapter 2, we shall examine the predictions of the isobaric spin formalism. Chapter 3 deals with the fabrication of the targets from isotopically separated germanium. Chapter 4 is concerned with the use of the accelerator; Chapter 5 describes the neutron detector and its associated circuitry. The data-taking phase is described in Chapter 6, and the method of analysis in Chapter 7. Chapter 8 presents the results of the experiment and discusses possible errors. The implications of the results for photonuclear physics are discussed in Chapter 9.

Previous Work on Germanium

The photoneutron cross sections of ^{70}Ge and ^{76}Ge have been studied before by means of activation techniques. Borello et al. (12) measured the cross sections of $^{70}\text{Ge}(\gamma, n)^{69}\text{Ge}$ and of $^{76}\text{Ge}(\gamma, n)^{75}\text{Ge}$ to 21 MeV. Ferrero et al.

(13) extended the study of $^{70}\text{Ge}(\gamma, n)$ to 31 MeV, also by means of activation techniques. Both experiments stop at too low an energy to show the structure of interest here. These older experiments do constitute a very valuable check on the present experiment.

CHAPTER 2. THEORY OF ISOSPIN

Before proceeding with the experiment, it is necessary to investigate specific consequences of the isobaric spin formalism for the particular case of germanium.

The electric dipole operator can be expanded in isospin space as:

$$\begin{aligned}\vec{D} &= \sum_{i=1}^Z \vec{r}(i) \\ &= \sum_{i=1}^A \vec{r}(i) [\frac{1}{2} - t_3(i)] \\ &= \frac{1}{2} \sum_{i=1}^A \vec{r}(i) - \sum_{i=1}^A \vec{r}(i) t_3(i)\end{aligned}$$

where t_3 equals $-1/2$ for protons and $+1/2$ for neutrons. The first term corresponds to Thomson scattering from the entire nuclear charge. The second term is the operator of interest for photonuclear excitations. The cross section for forming an excited state with total isospin T from the ground state $|T_0 T_0\rangle$ is:

$$\sigma(T_0 \rightarrow T) = \frac{4\pi^2 e^2}{3 \hbar c} E \left| \langle TT_0 | D_3 | T_0 T_0 \rangle \right|^2$$

The electric dipole operator D is a spherical tensor of rank one in isospin space. If the Wigner-Eckart theorem is applied to the matrix element, the result is:

$$\langle T T_0 | D_3 | T_0 T_0 \rangle = (-1)^{T-T_0} \begin{pmatrix} T & 1 & T_0 \\ -T_0 & 0 & T_0 \end{pmatrix} \langle T || D || T_0 \rangle$$

If explicit expressions for the 3-j symbol are substituted in, the geometrical factor can be seen in the cross sections.

$$\sigma(T_0 \rightarrow T_0 - 1) = 0$$

$$\sigma(T_0 \rightarrow T_0) = \frac{T_0}{T_0 + 1} \frac{4\pi^2 e^2}{3\hbar c} E |\langle T_0 || D || T_0 \rangle|^2$$

$$\sigma(T_0 \rightarrow T_0 + 1) = \frac{1}{T_0 + 1} \frac{4\pi^2 e^2}{3\hbar c} E |\langle T_0 + 1 || D || T_0 \rangle|^2$$

The ratio of the strengths is then given by:

$$\frac{\sigma(T_0 \rightarrow T_0 + 1)}{\sigma(T_0 \rightarrow T_0)} = \frac{1}{T_0} \frac{|\langle T_0 + 1 || D || T_0 \rangle|^2 E_{T_0 + 1}}{|\langle T_0 || D || T_0 \rangle|^2 E_{T_0}}$$

In our specific case, T_0 ranges from 3 in ^{70}Ge to 6 in ^{76}Ge . However, until cross sections are available in the (γ, p) channels, strength ratios will not be very meaningful, since transitions from the $T_0 + 1$ state to the neutron residual ground state are forbidden by the isospin selection rules. In other words, one expects to see very little of the $T_0 + 1$ strength in the (γ, n) or $(\gamma, 2n)$ channels.

A more useful quantity for calculation is the bremsstrahlung weighted cross section.

$$\sigma_{-1} \equiv \int \sigma/E \, dE$$

Hayward et al. (14) have derived sum rules for the bremsstrahlung weighted cross section by starting with the identity:

$$\begin{aligned} \langle T_0 | [[D \times D]^v] | T_0 \rangle &= \sqrt{2v+1} \, (-1)^{2T_0+v} \times \\ &\times \sum_{T'} \langle T_0 | D | T' \rangle \langle T' | D | T_0 \rangle \begin{Bmatrix} 1 & 1 & v \\ T_0 & T_0 & T' \end{Bmatrix} \end{aligned}$$

which is equation 7.1.1 of Edmonds (15). The bremsstrahlung weighted cross section is calculated from:

$$\sigma_{-1} = \frac{4\pi^2 e^2}{3\hbar c} \sum_k \langle 0 | D_3 | k \rangle \langle k | D_3 | 0 \rangle$$

The sum over k indicates a sum over all quantum numbers. If the sum is performed over all quantum numbers except T , then it is possible to define a bremsstrahlung weighted cross section for each isospin resonance.

$$\sigma_{-1}(T) = \frac{4\pi^2 e^2}{3\hbar c} |\langle TT_0 | D_3 | T_0 T_0 \rangle|^2$$

If the Wigner-Eckart theorem is applied, the result in terms of reduced matrix elements becomes:

$$\sigma_{-1}(T_0-1) = 0$$

$$\sigma_{-1}(T_0) = \frac{4\pi^2}{3} \alpha \frac{T_0}{(T_0+1)(2T_0+1)} |\langle T_0 || D || T_0 \rangle|^2$$

$$\sigma_{-1}(T_0+1) = \frac{4\pi^2}{3} \alpha \frac{1}{(T_0+1)(2T_0+3)} |\langle T_0+1 || D || T_0 \rangle|^2$$

where α is the fine structure constant. If this is substituted into the identity and explicit expressions are used for the 6-j symbol, three relations emerge:

$$\begin{aligned} \sigma_{-1}(T_0) - T_0 \sigma_{-1}(T_0+1) &= \frac{2}{3} \pi^2 \alpha \{ \sqrt{2} \langle T_0 T_0 || [D \times D]_0^1 || T_0 T_0 \rangle \\ &\quad - \sqrt{6} \langle T_0 T_0 || [D \times D]_0^2 || T_0 T_0 \rangle \} \end{aligned}$$

$$\begin{aligned} \sigma_{-1}(T_0) + \sigma_{-1}(T_0+1) &= \frac{4}{9} \pi^2 \alpha \{ \sqrt{3} \langle T_0 T_0 || [D \times D]_0^0 || T_0 T_0 \rangle \\ &\quad - \sqrt{6} \langle T_0 T_0 || [D \times D]_0^2 || T_0 T_0 \rangle \} \end{aligned}$$

$$\begin{aligned} \sigma_{-1}(T_0) + (2T_0+3) \sigma_{-1}(T_0+1) \\ &= \frac{4}{3} \pi^2 \alpha \{ \sqrt{3} \langle T_0 T_0 || [D \times D]_0^0 || T_0 T_0 \rangle \\ &\quad - \sqrt{2} \langle T_0 T_0 || [D \times D]_0^1 || T_0 T_0 \rangle \} \end{aligned}$$

It is then convenient to define isoscaler, isovector, and isotensor radii as follows:

$$\langle R_s^2 \rangle \equiv \langle T_0 T_z | \sum_{ij} (\vec{r}_i \cdot \vec{r}_j) (\vec{t}_i \cdot \vec{t}_j) | T_0 T_z \rangle$$

$$= \sqrt{3} \langle T_0 T_z | [D \times D]_0^2 | T_0 T_z \rangle$$

$$\langle R_v^2 \rangle \equiv \frac{1}{2T_z} \langle T_0 T_z | \sum_i r_i^2 t_{iz} | T_0 T_z \rangle$$

$$= \frac{\sqrt{2}}{2T_z} \langle T_0 T_z | [D \times D]_0^1 | T_0 T_z \rangle$$

$$\langle R_t^2 \rangle \equiv \frac{1}{3T_z - T_0(T_0+1)} \times \\ \times \langle T_0 T_z | \sum_{ij} (\vec{r}_i \cdot \vec{r}_j) (3t_{iz} t_{jz} - \vec{t}_i \cdot \vec{t}_j) | T_0 T_z \rangle$$

$$= \frac{\sqrt{6}}{3T_z - T_0(T_0+1)} \langle T_0 T_z | [D \times D]_0^2 | T_0 T_z \rangle$$

Combining these definitions with the previous set of equations gives:

$$\sigma_{-1}(T_0) = \frac{4\pi^2\alpha}{3(T_0+1)} \left[\frac{1}{3} T_0 \langle R_s^2 \rangle + T_0 \langle R_v^2 \rangle + \right. \\ \left. + \frac{1}{6} T_0 (2T_0-1) (2T_0+3) \langle R_t^2 \rangle \right]$$

$$\sigma_{-1}(T_0+1) = \frac{4\pi^2\alpha}{3(T_0+1)} \left[\frac{1}{3} \langle R_s^2 \rangle - T_0 \langle R_v^2 \rangle - \frac{1}{6} T_0 (2T_0-1) \langle R_t^2 \rangle \right]$$

The three relations which emerged from the tensor product identity are not linearly independent, and therefore the three have reduced to only two equations. These two expressions are the basis for calculations of the relative

strengths of the two resonances.

However, as was mentioned before, it will not be possible to compare calculated strength ratios with experimental data until the results of all three photonuclear experiments are available. The energy splitting of the resonances is a more meaningful parameter at this stage. The energy of a resonance can be calculated from:

$$\bar{E} = \frac{\sigma_{\text{int}}}{\sigma_{-1}} = \frac{\int \sigma \, dE}{\int \sigma/E \, dE}$$

Leonardi and Rosa-Clot (16) have worked out sum rules, analogous to the Thomas-Reiche-Kuhn sum rule of atomic physics, for the integrated cross section. Leonardi (17) has used all these sum rules to calculate the energy splitting in a modified harmonic oscillator model. His results for germanium are summarized in Table 2.

In summary, in this experiment the T_0+1 resonance is expected to be about 5-6 MeV above the T_0 resonance in the extreme case of ^{76}Ge , and should show at most 30% of the strength of the main resonance in the extreme case of ^{70}Ge . At this stage, the relative strengths are not decisive, but the second resonance, if observed, can be expected to show systematics as predicted in Table 2.

The total strength of all the resonances can be estimated using the electric dipole sum rule. This sum rule is completely analogous to the Thomas-Reiche-Kuhn sum rule of

Table 2. Theoretical predictions for germanium

Nucleus	T_0^a	$\bar{E}^b$	ΔE^c
^{70}Ge	3	17	3.8
^{72}Ge	4	17	4.0
^{74}Ge	5	17	5.4
^{76}Ge	6	17	5.6

^a T_0 is the ground state isospin

^b $\bar{E}$ is the main resonance energy (in MeV).

^c ΔE is the splitting between the two resonances as calculated by Leonardi.

atomic physics. The formula is:

$$\int \sigma(E) dE = (0.06 \text{ MeV-barns}) \frac{NZ}{A}$$

This formula has long been used as an estimate of total photonuclear cross section. In the mass region of germanium, the experimental cross sections generally exceed the predictions of the dipole sum rule by about 30%. A complete treatment of the dipole sum rule is given in Reference 18.

CHAPTER 3. TARGET PREPARATION

Since germanium has five stable isotopes, measurement of (γ, n) and $(\gamma, 2n)$ cross sections required the fabrication of targets of isotopically pure germanium.

Isotopically separated germanium was available from the Oak Ridge National Laboratory, but only in the form of GeO_2 . Because of the high cost of isotopically separated material, samples of GeO_2 containing only one gram of germanium metal were obtained from the Oak Ridge isotope pool.

Oxygen has strong resonances in the energy range under study. It was therefore very desirable to reduce the germanium dioxide samples to metallic germanium and eliminate a serious background problem. However, germanium dioxide is a very stable compound. Because of the high cost of the material, a high efficiency reduction process was necessary.

The method finally decided upon was based on Szekely's (19) industrial process. This process uses an atmosphere of hot hydrogen to reduce the dioxide to powdered germanium. Since the application to this experiment was concerned more with high efficiency of metal recovery than with maintaining extremely high purity as in an industrial situation, three loss mechanisms had to be dealt with. First, germanium forms hydrides which are gases at reduction temperatures. Second, germanium forms a monoxide which sublimes quite readily above about 700°C . Finally, the powdered germanium produced is ex-

tremely fine, and is very difficult to transfer quantitatively from one container to another.

After considerable trial and error, using practice samples of natural germanium, an adequate method was devised. The sample of dioxide (about 1.44g) was placed in a graphite boat and inserted into a quartz reaction tube and tube furnace. Hydrogen was passed through the tube continuously, and the furnace was slowly heated to 640°C. The reduction proceeded very slowly at this temperature, but monoxide sublimation became significant when higher temperatures were tried. After 12 hours the reaction was about 95% complete. In order to salvage as much of the remaining germanium as possible, without an unreasonably long reaction time, the temperature was then raised slowly to 710°C. Some germanium sublimed off as the monoxide at this point, but of course very little unreacted dioxide was left, and the reaction was effectively complete after five minutes at this temperature.

The reduced germanium at this point was a very finely divided powder. In order to put the metal in a form more easily removed from the reaction boat, the reaction tube was flushed with helium and the temperature then quickly raised to 1000°C. This temperature is well above the melting point (937.2°C) of germanium. The surface tension of the liquid was so high, the molten metal collected into a spherical droplet. When the furnace was cooled and the boat removed,

the solidified droplet could be removed with no detectable germanium remaining in the boat.

Total losses of germanium averaged about 8 mg for this process, implying a recovery efficiency of better than 99%. The hydrides of germanium are quite unstable (19) and probably dissociated spontaneously at the temperatures used here. Certainly most of the losses can be explained by monoxide sublimation. The sublimation was quite obvious because the monoxide formed a visible deposit on the reaction tube walls as the hydrogen flow carried it out of the hot region.

The end products of the reduction described above were small spheres of germanium about 5 mm in diameter and weighing about one gram. Such a small target mass makes it imperative to intercept as much beam flux as possible. Therefore it was necessary to flatten the samples into wafers. Germanium is extremely brittle in metallic form, so the samples were re-melted and flattened while molten. Tungsten-weighted graphite presses and a graphite boat were used once again for this process. The surface tension of liquid germanium is fairly high, and a press weighing 15 grams was necessary to flatten the samples to a thickness of one millimeter. This gave rectangular wafers with a beam interception area of about 1.5 cm². The samples were bombarded in this form. A quantitative description of the targets is given in Table 3.

Table 3. Target parameters

ISOTOPE	MASS (g)	% ⁷⁰ Ge	% ⁷² Ge	% ⁷³ Ge	% ⁷⁴ Ge	% ⁷⁶ Ge
⁷⁰ Ge	0.9774	98.8	0.71	0.10	0.29	0.10
⁷² Ge	0.9766	2.70	90.88	1.27	4.23	0.93
⁷⁴ Ge	0.9860	1.27	1.61	0.64	95.98	0.51
⁷⁶ Ge	0.9893	7.69	6.65	1.69	10.08	73.89

CHAPTER 4. PHOTON BEAM

The Iowa State University Number Two Synchrotron was an accelerator capable of accelerating electrons to a maximum energy of 70 MeV. The electrons struck a 10-mil tungsten target mounted on the inner radius of the donut, producing a bremsstrahlung beam which was then used to bombard the germanium targets.

A large, extremely sensitive detector such as the one used in this experiment naturally required a very tightly collimated beam. However, the heavy elements usually used for collimators have large (γ, n) cross sections, and therefore produce considerable neutron background during an experiment. A compromise solution was to use two collimators. The first collimator was molded out of Chemtree, a very dense neutron absorbing tungsten compound designed for reactor shielding. The second or "post-" collimator was a six inch hollow cylinder of nickel fitted into the iron shielding between the synchrotron and the neutron detector. Nickel has an unusually low (γ, n) cross section, and the use of the nickel post-collimator reduced the background by 30%.

The beam was monitored for total dose by a small thick ionization chamber calibrated in terms of a larger "P2" chamber built to the standards of Pruitt and Domen (20). The ionization produced within this chamber was integrated on a

.001 μ fd glass capacitor and the charge was measured by a vibrating reed electrometer.

Because of the method used to extract reaction rates, it was highly desirable to produce as stable a beam as possible, i.e. to cause all beam bursts to have exactly the same intensity. Unfortunately, there was always some intensity jitter, and the beam quality went down as the mean intensity was reduced. Long-term (about one second) variations in beam intensity were compensated for by placing a six-inch liquid scintillation detector in the beam path, integrating the output pulse to compute the burst intensity, and feeding this information into a servo system governing the synchrotron's injection timing. Fast, burst-to-burst jitter was alleviated somewhat by monitoring the magnetic field risetime and tailoring the injection timing on a burst-to-burst basis. When coupled with a considerable amount of skill on the part of the operator, beam pulse intensity distributions with widths as low as 8% were achieved.

The beam burst duration was an extremely important parameter. Efficiency considerations required that the counting gate start no more than two microseconds after the beginning of the beam burst. Therefore, the beam burst had to be shorter than two microseconds. This requirement was met by increasing the accelerator's radio frequency from its usual 165 MHz to 169.05 MHz. This change increased the

synchrotron orbit radius and produced a better geometrical relationship between the electron orbit and the knockout coils. With this arrangement, the beam burst lasted about one microsecond. However, the beam direction would swing out of line with the collimators at energies below 15 MeV. A compromise was reached by lowering the knockout coil current pulse at these energies, but the price was paid in reduced beam intensities.

Stability Considerations

The beam intensity required when bombarding these small samples was about a factor of 10 larger than the low intensities used by Jones (21) and Kocinski (22) on much larger targets, but was still about a factor of 100 below the minimum intensities needed for activation-type experiments. It was very difficult to get a controllable beam at such intensities; most machine controls would drop the intensity to zero very rapidly if detuned. Moreover, the eight magnetic focusing coil settings were not only interdependent, but also were functions of the electron current within the donut. The solution was to inject at a lower energy (50 KeV rather than the usual 70 KeV), which reduced the phase space window for capture in the betatron orbit, and then inject sooner in the machine cycle to bring the intensity down to the value

desired. The 50 kilovolt injection pulse required several microseconds to reach its peak, and this gave the injection timing profile a lower "Q" than the other timing controls.

CHAPTER 5. NEUTRON DETECTOR

The neutron detector used in this experiment was a cylindrical tank, 40" in diameter and 43" in length, containing approximately 240 gallons of liquid scintillator loaded with 5 kg of gadolinium. This detector has been described in detail by C. C. Jones (21). The layout of the detector is shown in Figure 1.

The method of detection used the high thermal neutron cross sections of ^{157}Gd ($\sigma = 240\,000\text{ b}$) and ^{155}Gd ($\sigma = 58\,000\text{ b}$). Photoneutrons produced in the germanium targets were thermalized in the scintillator fluid and most were then captured by a ^{157}Gd nucleus, releasing 8 MeV as neutron capture gamma rays. These gammas were detected by scintillation in the usual manner, and it was this capture gamma pulse which was counted.

Naturally, a detector with such a large sensitive volume, almost a cubic meter, presented a considerable background problem. It is useful to distinguish two types of background, natural and beam-induced. Natural background was always present, and was produced primarily by cosmic sources and by residual radioactivity in the accelerator room. Beam-induced background was present only during bombardments and was produced by sources such as scattered gammas from the beam and photoneutrons from the collimation system. These effects made heavy shielding essential. The innermost layer

Figure 1. Detector layout

consisted of a four inch wall of lead. The next layer was a quarter inch wall of "Boral" neutron shielding. This entire assembly was placed in a concrete counting house with 18 inch solid concrete walls and a four inch steel roof. The shielding in the direction of the accelerator consisted of 26 inches of iron and four inches of boron loaded paraffin. Shielding was also needed in the beam exit wall because of backscattered photoneutrons from the beam monitors.

Detector Electronics

The counting system electronics are shown in Figure 2. The scintillation flashes were collected by 28 EMI-9583B photomultipliers. The photomultiplier outputs were summed in two banks such that physically adjacent tubes were members of different banks. These banks were fed into discriminators and the logic pulses fed into a fast (50 nanosecond) coincidence circuit. This fast coincidence virtually eliminated noise pulses from photomultiplier dark current.

The lower and upper level discriminators were necessary to reduce natural background. The energy spectra of the data pulses, measured using several sources, are illustrated in Figure 3. Upper level discrimination, set at the equivalent of 15 MeV, cut out the saturation level pulses from cosmic rays. The lower level discriminators were set at a compromise energy equivalent of 2 MeV. This still allowed some low energy noise to come through, but any higher setting

Figure 2. Detector electronics

Figure 3. Energy spectra of the detector

would have cut out too many data pulses.

The fourth "strobe" input to the fast coincidence gate was connected to a timing pulse which governed the whole system. A plot of data counts versus time is given in Figure 4. The gate was turned on two microseconds after the start of the beam burst. Photons scattered out of the beam, plus neutron thermalization pulses, saturated the analog electronics and thus made counting impractical until two microseconds had passed. Once initiated, the counting interval lasted for 20 microseconds. It was discovered that counting intervals longer than 20 microseconds resulted in a reduction of foreground to background ratio because of beam induced neutron background.

The entire timing cycle is given in Figure 5. Counts detected during the "data" interval were accumulated in scaler 1, counts detected during any "background" interval were accumulated in scaler 2. These background counting intervals were used to monitor natural background during a bombardment. In this way, the contribution of sample radioactivity was included in corrections for natural background.

Scalers 3 and 4 were diagnostic in purpose. Scaler 3 counted the number of background intervals (nominally 100) and scaler 4 merely indicated that a beam burst had lasted too long. All four scalers were read and zeroed after each

Figure 4. Neutron detector time spectrum
Time zero corresponds to
surface barrier detectors
registering a fission in the
 ^{252}Cf source

Figure 5. Detector timing cycle

synchrotron cycle by the SDS 910 computer, and data collected during any bad beam bursts were automatically rejected.

Some difficulties were encountered in starting the detector timing circuitry. The first two microseconds had to include the entire beam burst. If this critical condition were not met, either the data would be buried in beam flash or else the efficiency would be seriously reduced. Therefore, the fast beam monitor was used to tie the timing circuitry directly to the beam. However, background in the fast monitor started the cycle early about 10% of the time. For this reason, a delayed coincidence with the synchrotron "start knockout" control pulse was included. This left the system exposed to spurious monitor pulses for only a few microseconds and reduced the number of bad bursts described above almost to zero.

Detector Efficiency

The counting efficiency was calibrated with a weak ^{252}Cf fission source. This source was mounted in a small brass container between two surface barrier fission fragment detectors, and the container placed in the center of the detector's beam tube. The detector timing cycle was initiated by the surface barrier detectors, and the number of counts per fission measured. Since the average number of neutrons per fission was measured by DeVolpi and Porges (23) to be 3.725 ± 0.015 , the calculation of the efficiency was very

straightforward. The result of the calibration runs was an efficiency of $73.6 \pm 0.5\%$.

Detector Stability

Detector stability was a problem at first. This was dealt with in two ways: First, all analog circuitry was installed in the temperature-controlled environment of the counting house. Second, a light emitting diode was used to simulate a monoenergetic source in the intervals between beam pulses, and these monoenergetic pulses were used in a feedback loop to the phototube power supplies to keep the overall gain of the system constant. As a result, efficiency varied less than a half per cent during the entire experiment.

CHAPTER 6. DATA COLLECTION

The entire data collection process was automated by means of an SDS 910 on-line computer. After every beam burst, the computer read all four scalers and rejected the data if scaler four indicated trouble. The contents of scaler two (background counts) and scaler three (background intervals) were simply added into accumulators. Scaler one (data counts) was analysed into channels called multiplicity bins. If a beam burst produced no neutron counts at all, bin zero was incremented. If one neutron was detected, bin one was incremented, if two were detected, bin two was incremented, and so on. Forty bins (zero through nine for four beam intensity regions) were accumulated in this experiment. Bin nine actually was incremented not only for a scaler reading of nine, but also for any scaler reading greater than nine. However, the count rate in the ninth bin was less than one per day, so this inclusion of overflows had little practical significance.

The computer also monitored the intensity of each beam burst by means of an ADC connected to the fast beam monitor. The burst intensity pulse was analysed and accumulated into a peak height spectrum in the conventional manner. The pulse height spectrum was thus a beam intensity histogram, which was necessary in corrections for beam jitter.

Each data-taking bombardment was preceded by a one minute "pre-run" bombardment. This short pre-run was used not only to obtain some diagnostic information on the equipment, but also to estimate the distribution of burst intensity. Burst intensities varied up to 30% in this experiment, although it was usually possible to narrow the width of the distribution to as low as 8%. The pre-run distribution was used to classify bursts as low, medium, or high intensity. A "medium burst" was defined as having an intensity within one half of a standard deviation of the mean pre-run intensity. "High" and "low" bursts were defined as being greater than or less than the "medium" region, as illustrated in Figure 6. Four sets of multiplicity bins were accumulated, one for low bursts, one for medium bursts, one for high bursts, and one for all bursts. This method of detailed event by event recording was essential for the separation of reaction multiplicities, as will be demonstrated in the analysis of the data.

Data-taking runs lasted one hour, not including the pre-run, and were taken at end-point energies from 10 MeV to 40 MeV in 2 MeV steps. The computer had a cathode-ray tube display which was used by the synchrotron operator to tune the accelerator for optimum parameters for any particular energy. The beam histogram described above was continuously displayed to allow the operator to tune for a narrow and

Figure 6. Layout of burst intensity regions

Beam bursts were classified as low, medium, or high on the basis of an intensity histogram. A set of 10 multiplicity bins was accumulated for each region, as well as a set for all bursts. The three beam-classified sets were used to correct the inclusive set for distortion caused by the intensity jitter.

symmetrical distribution. The count rate, averaged over sixteen beam bursts, was also displayed. Reaction multiplicity extraction can be done most accurately when the count rate is 0.3 to 0.5 neutrons per burst, and the operator adjusted the intensity accordingly. The screen also displayed some diagnostic information, such as excessively long burst duration or wrong target in the beam, and thus prevented unnecessary lost runs.

While the detector gating and coincidence circuit were quite effective in reducing natural background, there was no way to eliminate beam-induced background counts with the circuitry. It was necessary to measure this background separately and subtract it numerically from the yield data. Every data run was preceded and followed by a 30 minute background run. Background runs had their own pre-runs and were identical to data runs except a sample holder with no germanium was placed in the detector's beam tube. It was soon discovered, despite the careful shielding and collimation, that the neutron count rate from the germanium target was never more than about 7% of the total.

Stability checks were made after every four sets of runs. These checks included a "standardization" run, a "natural background" run, and a normal background-data-background set at 40 MeV. The 40 MeV set was simply to check for reproducibility at the energy where the synchrotron was

easiest to control. The natural background run used a pulser instead of beam bursts to drive the timing electronics, and checked the natural background count rate and the drift of the beam monitoring ionization chamber with the beam off. The standardizing runs checked the detector efficiency and the ionization chamber sensitivity. The ^{252}Cf source was placed in the detector's beam tube and a standard ^{90}Sr source was placed in a port in the side of the ionization chamber. Both the natural background and the standardizing runs lasted ten minutes and were completely computer controlled.

The various energies were run in random order until at least three yields in reasonable agreement were obtained at each energy. Six weeks of continuous operation were required for the four isotopes.

CHAPTER 7. ANALYSIS OF THE YIELDS

The data analysis involved three distinct stages. The first problem was the separation of single and double neutron events. Secondly, the individual reaction yields were averaged together and normalized to the energy collected on the ionization chamber. Finally, reaction cross sections were extracted from the reduced yields.

Separation of Multiplicities

The multiplicity-bin analysis of the neutron counts for each individual beam burst allowed the statistical extraction of the reaction multiplicities. First it was necessary to calculate the form of the multiplicity spectrum. Since this experiment covered an energy range up to and including 40 MeV, it was energetically possible to have photoabsorption leading to emission of up to four neutrons. If R_k is defined to be the average number of disintegrations of the form (Y, kn) per burst, then the average number of neutrons emitted per burst by reaction (Y, kn) is then $k R_k$. If one divides the 20 μ sec counting interval into b sub-intervals, where b is so large that the possibility of two events being detected in the same sub-interval can be neglected, then the probability of detecting an event in any particular sub-interval is R_k/b . The probability that j events will be detected in a particular set of the sub-intervals, and not in the remaining $b-j$ sub-intervals, is:

$$(R_k/b)^j (1 - R_k/b)^{b-j}$$

However, there are many possible sets of j sub-intervals. The first detection can fall into b intervals, the second into $b-1$, and the last into $b-j+1$ sub-intervals. Therefore the number of possible sets of sub-intervals is

$$b(b-1)(b-2)\dots(b-j+1) = b!/j!$$

Because the events are indistinguishable, this expression must be divided by the number $j!$ of equivalent permutations. The number of non-equivalent sets of j sub-intervals is then

$$b!/(j!)^2$$

Define $P(j,k)$ to be the probability that j sub-intervals will contain an event caused by the reaction (γ, kn) .

$$P(j,k) = [b!/(j!)^2] (R_k/b)^j (1 - R_k/b)^{b-j}$$

Assume $b \gg j$. This permits the approximation

$$b!/(j!)^2 = \frac{b(b-1)\dots(b-j+1)}{j!} \approx b^j/j!$$

Substituting this in the equation for $P(j,k)$, one obtains:

$$\begin{aligned} P(j,k) &= (b^j/j!) (R_k/b)^j (1 - R_k/b)^{b-j} \\ &= \frac{R_k^j}{j!} \left(1 - \frac{R_k}{b}\right)^{b-j} \end{aligned}$$

Using the binomial theorem to expand the last factor, this becomes:

$$P(j,k) = \frac{R_k^j}{j!} \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \frac{(b-j)!}{(b-j-n)!} \left(\frac{R_k}{b}\right)^n$$

Then as b approaches infinity:

$$\begin{aligned} P(j,k) &= \lim_{b \rightarrow \infty} \frac{R_k^j}{j!} \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} b^n \left(\frac{R_k}{b}\right)^n \\ &= \frac{R_k^j}{j!} \sum_{n=0}^{\infty} (-1)^n \frac{R_k^n}{n!} \end{aligned}$$

But this series is just the expansion of the reciprocal exponential function.

$$P(j,k) = \frac{R_k^j}{j!} \exp(-R_k)$$

This is, of course, the Poisson distribution. We now define r to be the probability that m neutrons are produced by a single beam burst. Since the $(\gamma, 4n)$ reaction has the highest multiplicity that is energetically allowed, the r_m are given by:

$$r_0 = P(0,1) P(0,2) P(0,3) P(0,4)$$

$$r_1 = P(1,1) P(0,2) P(0,3) P(0,4)$$

$$r_2 = P(2,1) P(0,2) P(0,3) P(0,4) \\ + P(0,1) P(1,2) P(0,3) P(0,4)$$

$$r_3 = P(3,1) P(0,2) P(0,3) P(0,4) \\ + P(2,1) P(1,2) P(0,3) P(0,4) \\ + P(0,1) P(0,2) P(1,3) P(0,4)$$

etc.

or in general:

$$r_m = \sum_{\substack{j_1 j_2 \\ j_3 j_4}} P(j_1,1) P(j_2,2) P(j_3,3) P(j_4,4) \delta(j_1 + 2j_2 + 3j_3 + 4j_4, m)$$

where the delta function is the usual Kronecker delta.

If the neutron detector were 100% efficient, these r_m

would give the multiplicity spectrum directly. To take the efficiency into account, let y_n be the probability of observing n neutrons, and let ϵ be the counting efficiency.

$$y_0 = r_1 + (1-\epsilon)r_2 + (1-\epsilon)^2 r_2 + \dots$$

$$y_1 = \epsilon r_1 + 2(1-\epsilon)r_2 + 3\epsilon(1-\epsilon)^2 r_3 + 4\epsilon(1-\epsilon)^3 r_3 + \dots$$

$$y_2 = \epsilon^2 r_2 + 3\epsilon^2(1-\epsilon)r_3 + 6\epsilon^2(1-\epsilon)^2 r_4 + \\ + 10\epsilon^2(1-\epsilon)^3 r_5 + \dots$$

and in general:

$$y_n = \sum_{m=n}^{\infty} \frac{m!}{n! (m-n)!} \epsilon^n (1-\epsilon)^{m-n} r_m$$

This is the desired result. If the symbols defined along the way are substituted in, the result is:

$$\begin{aligned}
Y_n = & \sum_{m=n}^{\infty} \frac{m!}{n!(m-n)!} \epsilon^n (1-\epsilon)^{m-n} \times \\
& \times \sum_{\substack{j_1 j_2 \\ j_3 j_4}} \frac{R_1^{j_1} R_2^{j_2} R_3^{j_3} R_4^{j_4}}{j_1! j_2! j_3! j_4!} \exp-(R_1+R_2+R_3+R_4) \times \\
& \times \delta(j_1+2j_2+3j_3+4j_4, m)
\end{aligned}$$

where R_1 is the rate of the reaction (γ, n) , R_2 of $(\gamma, 2n)$, etc. and ϵ is the counting efficiency. In practice, since only 10 bins were accumulated and there was very seldom a count above bin 6, the sum on m was truncated at $m=9$.

Unfortunately, the experiment provided the multiplicity bins y_n , but the reaction rates R are the desired quantity. Ten bins were accumulated, each one of which followed the usual Poisson counting distribution, and the determination of the R 's became a nonlinear regression problem in statistics. The algorithm used to extract the R 's was the gradient-expansion method of Marquardt (24) and Bevington (25). The method will be sketched briefly here; a more complete description can be found in Reference 25. χ^2 is defined in the usual manner:

$$\chi^2 = \sum_{n=1}^9 (y'_n - y_n)^2$$

where y'_n is the experimental multiplicity bin, normalized to a single beam burst, and y_n is the calculated value from the above equation. It is desired to vary the R 's so as to minimize χ^2 . To do this, we expand the y_n in a Taylor series about some point R_k^0 :

$$y_n(R_k) = y_n(R_k^0) + \sum_{k=1}^4 \frac{\partial y_n}{\partial R_k} \Delta R_k$$

where $\Delta R_k = R_k - R_k^0$. To minimize χ^2 , its derivatives with respect to the ΔR_k are set to zero:

$$\frac{\partial \chi^2}{\partial \Delta R_k} = 0$$

$$-2 \sum_{n=1}^9 (y'_n - y_n - \sum_{i=1}^4 \frac{\partial y_n}{\partial R_i} \Delta R_i) \frac{\partial y_n}{\partial R_k} = 0$$

$$\sum_{i=1}^4 \sum_{n=1}^9 \frac{\partial y_n}{\partial R_k} \frac{\partial y_n}{\partial R_i} \Delta R_i = \sum_{n=1}^9 (y'_n - y_n) \frac{\partial y_n}{\partial R_k}$$

Define two matrices α and β :

$$\alpha_{ki} = \sum_{n=1}^9 \frac{\partial y_n}{\partial R_k} \frac{\partial y_n}{\partial R_i}$$

$$\beta_k = \sum_{n=1}^9 (y'_n - y_n) \frac{\partial y_n}{\partial R_k}$$

In terms of α and β , the equation for the ΔR_i is:

$$\sum_i \alpha_{ki} \Delta R_i = \beta_k$$

In matrix form,

$$\alpha \Delta R = \beta$$

In this equation, if initial estimates R_1^0 are available, α and β can be calculated and α can be inverted to solve for the ΔR_i .

$$\Delta R = \alpha^{-1} \beta$$

If the functional behavior of the y_n were close to linear with respect to the R_i , this simple approach would be sufficient. The ΔR_i could be added to the R_1^0 and the whole process repeated until solutions of the desired accuracy were obtained. Unfortunately, the higher-order terms in the Taylor series can be neglected only if the initial estimates R_1^0 are very close to the ultimate solution. The gradient-

expansion algorithm provides a way out of this difficulty by defining a new matrix α' :

$$\alpha'_{ki} = \alpha_{ki} (1 + \lambda \delta_{ki})$$

where the delta is a Kronecker delta. α' is used in place of α in the iteration equations:

$$\alpha' \Delta R = \beta$$

If λ is small, the ΔR_i produced in each iteration are very close to the values obtained in the simple expansion algorithm. However, if λ is large, the diagonal terms of α' dominate, and the equations decouple:

$$\lambda \sum_{n=1}^9 \left(\frac{\partial y_n}{\partial R_k} \right)^2 \Delta R_k = \sum_{n=1}^9 (y'_n - y_n) \frac{\partial y_n}{\partial R_k}$$

$$\Delta R_k = \left[\lambda \sum_{n=1}^9 \left(\frac{\partial y_n}{\partial R_k} \right)^2 \right]^{-1} \sum_{n=1}^9 (y'_n - y_n) \frac{\partial y_n}{\partial R_k}$$

However,

$$(\vec{\nabla} \chi^2)_k = -2 \sum_{n=1}^9 (y'_n - y_n) \frac{\partial y_n}{\partial R_k}$$

$$\Rightarrow \Delta R_k = -\frac{1}{2} \left[\lambda \sum_{n=1}^9 \left(\frac{\partial y_n}{\partial R_k} \right)^2 \right]^{-1} (\vec{\nabla} \chi^2)_k$$

In the case of large λ , the increments ΔR_k are opposite to the direction of the gradient of the χ^2 hypersurface and are scaled down by α_{kk} and λ . The gradient vector points in the direction in which χ^2 increases most rapidly, so the increments ΔR_k must make χ^2 decrease if λ is large enough.

In summary, when λ is small, the algorithm is ideally suited to locating the minimum χ^2 from a point close by, and when λ is large, the algorithm is suitable for finding the approximate minimum from far away. The procedure suggested by Marquardt is:

1. Start with $\lambda = 0.001$.
2. Compute the ΔR_k and the new χ^2 .
3. If χ^2 increased, multiply λ by 10 and repeat step 2.
4. If χ^2 decreased, divide λ by 10, add the ΔR_k to the R_k^0 , and go back to step 2.

Two problems arose when this standard statistical method was applied to this particular analysis. The first problem arose from a very simple physical reason: reaction rates must be zero when the bombarding energy is below the reaction

threshold. When $R_k^0 = 0$ for more than one reaction, the partial derivatives with respect to these rates become equal and the matrix α' becomes singular. The solution to this difficulty was to reduce the dimensions of α' and β so that only one zero R_k^0 was included. Higher-order reaction rates were then fixed at zero for every iteration in which a lower order R_k was zero. Since it can be argued from physical grounds that

$$R_k \geq R_{k+1} \geq 0$$

the fitting method gave physically acceptable solutions.

The second problem which arose in application was that the equations

$$\alpha' \Delta R = \beta$$

were ill-conditioned for the numbers in this particular experiment. "Ill-conditioned" equations have small determinants and therefore the solution is very sensitive to any errors in the input values. Examples were encountered where changes in α' on the order of 1% produced changes of two orders of magnitude in the solutions ΔR_k . To overcome this difficulty, the computer program had to re-normalize the equations whenever this ill-conditioning appeared.

Except for the two problems in application mentioned above, the method worked very well. The initial estimates for R_1 and R_2 were computed using Goryachev's (26) method, R_3 and R_4 were simply started at zero. Typically, after only six iterations the relative change $\Delta R_k / R_k$ was less than one part in ten thousand and the fitting procedure was terminated. The accuracy of this fitting method was estimated using a Monte Carlo simulation technique. The results are illustrated in Figure 7. It is characteristic of this procedure that, as the average number of counts per beam burst increases, the accuracy of the fitting procedure decreases. However, if the count rate is too low, the total statistics introduce significant error. The optimum count rate was found to be about 0.3 counts per beam burst, and the experiment was run at carefully regulated beam intensities for this reason.

One last correction used in the fitting procedure must still be taken into account. The derivations above all assume a photon beam with no variations in intensity. In practice, the intensity jitter was often as high as 10% of the mean intensity. This jitter had the effect of spreading out the distribution of counts over the multiplicity bins, and therefore introduced considerable error into the fitting procedure. To calculate the effect of beam intensity jitter, first expand the multiplicity bin rates in a Taylor series

Figure 7. Accuracy of fitting procedure
 The accuracy of the statistical fitting procedure was estimated by means of a Monte Carlo technique. The accuracy with which reaction rates were recovered from multiplicity spectra is plotted against the average count rate.

about the mean intensity:

$$y_n(I) = y_n(\bar{I}) + (I - \bar{I}) y'_n(\bar{I}) + (I - \bar{I})^2 y''_n(\bar{I}) + \dots$$

where I is the beam burst intensity. The experiment measures the average of this quantity over all intensities in the distribution.

$$\bar{y}_n = \frac{\int_0^\infty N(I) y_n(I) dI}{\int_0^\infty N(I) dI}$$

where $N(I)$ is the intensity distribution.

Let

$$N_t \equiv \int_0^\infty N(I) dI$$

$$\begin{aligned} \bar{y}_n = \frac{1}{N_t} \int_0^\infty N(I) y_n(\bar{I}) dI &+ \frac{1}{N_t} \int_0^\infty N(I) y'_n(\bar{I}) (I - \bar{I}) dI + \\ &+ \frac{1}{N_t} \int_0^\infty N(I) y''_n(\bar{I}) (I - \bar{I})^2 dI + \dots \end{aligned}$$

Truncating the series after the third term, and considering the second term:

$$\begin{aligned}
\int_0^{\infty} N(I) y_n'(\bar{I}) (I - \bar{I}) dI &= y_n'(\bar{I}) \int_0^{\infty} I N(I) dI \\
&- y_n'(\bar{I}) \bar{I} \int_0^{\infty} N(I) dI \\
&= y_n'(\bar{I}) (\bar{I} N_t) - y_n'(\bar{I}) \bar{I} (N_t) \\
&= 0
\end{aligned}$$

Let σ_I be the standard deviation of the beam intensity histogram. Then:

$$\bar{y}_n = \frac{1}{N_t} y_n(\bar{I}) N_t + \frac{1}{N_t} y_n''(\bar{I}) \sigma_I$$

$$\bar{y}_n = y_n(\bar{I}) + \frac{\sigma_I}{N_t} y_n''(\bar{I})$$

$$y_n(\bar{I}) = \bar{y}_n - \frac{\sigma_I}{N_t} y_n''(\bar{I})$$

The experiment measured $\bar{y}_n$ and the fitting procedure needed $y_n(\bar{I})$. σ_I and N_t were easily calculated from the beam histogram. The only quantity necessary to complete the correction was the second derivative $y_n''(\bar{I})$. This is the reason multiplicity bins were accumulated for low, medium, and high bursts as well as for all bursts. The $y_n''(\bar{I})$ were calculated numerically, and the $\bar{y}_n$ were corrected for jitter just before fitting. The coefficient σ_I/N_t was on the order of 0.1, so higher-order terms in the Taylor expansion were not signifi-

cant, and no further corrections were made for intensity jitter.

Reduction of the Yields

Reaction rates for the four energetically allowed reactions were extracted from the data using the above procedure. To calculate the reaction yields, these rates were divided by the dose per beam, as measured by the ionization chamber.

$$\alpha_n(E) \equiv \frac{y_n(E)}{\text{Dose per Beam}}$$

This "raw yield" must now be related to the reaction cross section. The notation developed by Penfold and Leiss (27) will be used in the following equations. For simplicity, the subscript "n" will be dropped, since the analysis was the same for $n=1$ and $n=2$. The $(\gamma, 3n)$ and $(\gamma, 4n)$ reactions were of interest only as corrections to the (γ, n) and $(\gamma, 2n)$ yields, and will not be considered further. The raw yield is related to the photonuclear cross section by:

$$\alpha(E) = n_s \int_0^\infty N(E, k) \sigma(k) dk$$

$\alpha(E)$ is the number of reactions per unit of monitor response for the reaction under study. n_s is the number of target nuclei divided by the cross-sectional area of the beam, $N(E, k)$ is the number of photons of energy k per unit range of

k which enter the sample per unit of monitor response, and $\sigma(k)$ is the photonuclear cross section. The incident photon spectrum, $N(E,k)$, can be expressed as:

$$N(E,k) = \left[\frac{\phi(E,k)}{k} \right] \frac{f_s(k)}{F(E)}$$

The factor in brackets is a renormalized form of the bremsstrahlung cross section as calculated by Schiff (28). It is written in this form to emphasize the dominant $1/k$ dependence.

$$\phi(E,k) \equiv \frac{1}{16} \frac{137}{2Z^2} \left(\frac{mc^2}{e^2} \right)^2 k \sigma_{\text{Brem}}(E,k)$$

$f_s(k)$ is the collective transmission function for all material between the bremsstrahlung radiator and the sample. $F(E)$ is the monitor response function which normalizes $N(E,k)$ to unit monitor response. The yield equation is reduced by substituting these functions for $N(E,k)$:

$$\alpha(E) = n_s \int_0^\infty \frac{\phi(E,k)}{k} \frac{f_s(k)}{F(E)} \sigma(k) dk$$

$$F(E) \alpha(E) = \int_0^\infty \frac{\phi(E,k)}{k} n_s f_s(k) \sigma(k) dk$$

Define $Y(E) \equiv F(E) \alpha(E)$

and $S(k) \equiv n_s f_s(k) \sigma(k)$

$$Y(E) = \int_0^{\infty} \frac{\Phi(E,k)}{k} S(k) dk$$

It is this reduced equation which is solved for $S(k)$ and the $\sigma(k)$ are then calculated by a simple multiplication.

"Reducing" the yields was simply a matter of calculating $F(E)$ and multiplying by $\alpha(E)$.

To calculate $F(E)$, it is necessary to examine the photon spectrum incident on the ionization chamber monitor. Let $N_m(E,k)$ be this spectrum.

$$N_m(E,k) = \frac{\Phi(E,k)}{k} \frac{f_m(k)}{F(E)}$$

Where $f_m(k)$ is the collective transmission function for all materials in front of the monitor. The energy in this spectrum is:

$$E_m(E) = \int_0^{\infty} N_m(E,k) k dk = \frac{1}{F(E)} \int_0^{\infty} \Phi(E,k) f_m(k) dk$$

The response of the monitor to each Mev of energy collected is defined as $R(E)$ response units (in this case, volts on the integrating capacitor) per Mev.

$$R(E) = \frac{1}{E_m(E)}$$

$$F(E) = R(E) \int_0^{\infty} \phi(E,k) f_m(k) dk$$

The evaluation of $R(E)$ was straightforward. The voltage on the capacitor gave the total charge collected, and the collected charge was related to the energy collected by Pruitt and Domen's calorimeter calibrations (20).

After the reduced yield from each bombardment was calculated, the beam-induced background yields were averaged together and subtracted from the data yields. The differences, which were the yields from the germanium target, were averaged together to give one reduced yield curve for each of the eight reactions. Error estimates for the reduced yields were calculated from the spread of the unaveraged yields at each energy.

Extraction of the Cross Sections

To extract the cross sections, it was necessary to solve the reduced yield equation:

$$Y(E) = \int_0^{\infty} \frac{\phi(E,k)}{k} S(k) dk$$

Solution of this equation for $S(k)$ is complicated by the

effect of statistical errors in the $Y(E)$, which cause calculated solutions to oscillate, especially at high values of k . Cook (29) has developed a special method, called the "Least Structure Solution," to solve this equation. The Least Structure procedure calculates the smoothest numerical solution statistically consistent with the reduced yields $Y(E)$ and their estimated errors. In this particular application, the smoothest solution was that solution which minimized the second difference function:

$$S_1 = \sum_1 \frac{1}{2} (\sigma_{i-1} - 2\sigma_i + \sigma_{i+1})^2$$

and also satisfied:

$$\bar{\chi}^2 \equiv \sum_1 \frac{(\bar{Y}_i - Y_i)^2}{(\Delta Y_i)^2} \leq 16$$

where the index i is the index of the 16 energies, the Y_i are the yields calculated from the solution σ_i , the ΔY_i are the estimated errors in the reduced yields Y_i , and 16 is the number of data points. A detailed analysis of this method has been presented in reference 29, and the method has been critically evaluated by Bramanis et al. (30).

After the Least Structures procedure produced the $S(k)$, the final cross sections were calculated from:

$$\sigma(k) = \frac{S(k)}{n_s \cdot f_s(k)} \quad .$$

CHAPTER 8. EXPERIMENTAL RESULTS

The cross sections for the eight reactions are presented in Figures 8 through 17. Each of the (γ, n) cross sections shows the familiar giant resonance peaking at about 17 MeV with a width of six to seven MeV and a peak cross section of the order of 125 mb. The more spread out appearance of the ^{70}Ge resonance is certainly at least in part the result of smoothing of the relatively poor quality data obtained for this nucleus. The main giant resonance peaks observed in ^{72}Ge , ^{74}Ge , and ^{76}Ge are identical to within the accuracy of this measurement. This result is, of course, that expected from the usual collective model treatment of the giant resonance.

The second (γ, n) resonance however is more unusual. First, its existence is not predicted by the usual theory. It shows definite systematics in that it loses both strength and energy as the neutron excess increases, going from 71% of the main giant resonance at 35 MeV in ^{70}Ge to 18% of the main peak at 26 MeV in ^{76}Ge . The $(\gamma, 2n)$ resonance peaks at about 26 MeV in all four nuclei, but gains strength as the neutron excess is increased.

The vertical error bars shown in the figures are calculated from the reproducibility of the reduced yields. The horizontal error bars are not uncertainties in the energies. Instead, they are a measure of the degree of smoothing

Figure 8. (γ, n) reactions

The (γ, n) channels of the four isotopes are shown superimposed.

Figure 9. Cross section of $^{70}\text{Ge}(\gamma, n)^{69}\text{Ge}$

The results of this measurement are shown together with the results of the two earlier experiments.

Figure 10. Cross section of $^{72}\text{Ge}(\gamma, n)^{71}\text{Ge}$

Figure 11. Cross section of $^{74}\text{Ge}(\gamma, n)^{73}\text{Ge}$

Figure 12. Cross section of $^{76}\text{Ge}(\gamma, n)^{75}\text{Ge}$

The results of the earlier experiment are shown at half scale to facilitate comparison.

Figure 13. Cross section of $^{70}\text{Ge}(\gamma, 2n)^{68}\text{Ge}$

Figure 14. Cross section of $^{72}\text{Ge}(\gamma, 2n)^{70}\text{Ge}$

Figure 15. Cross section of $^{74}\text{Ge}(\gamma, 2n)^{72}\text{Ge}$

Figure 16. Cross section of $^{76}\text{Ge}(\gamma, 2n)^{74}\text{Ge}$

performed by the Least Structures procedure. If the physical cross section contained a delta function resonance, the Least Structures procedure would show a smoothed resonance with a base width equal to the length of the horizontal error bar. Consequently, the positions of the peaks and the areas included under the peaks are accurate, but the widths of the resonances are not nearly as quantitative.

Sources of Experimental Error

Smoothing is necessary because of random statistical errors in the yields. The Least Structures procedure produces smooth solutions at the expense of resolution. By far the dominant cause of the random errors was the beam induced background. This background exhibited characteristic yield-curve behavior with maximum slope between 14 and 20 MeV. Consequently, the worst foreground to background ratio existed around 16 to 18 MeV, where the sample yield was still low and the background already high. This energy range is just where the (γ, n) giant resonance peaks, and this is one reason why the Least Structures solutions tended to overshoot on the high energy side of the resonances.

The normalization between isotopes is accurate to better than 0.5%, much less than statistical error, since this normalization is affected primarily by the target masses, and to a much lesser degree by the target thicknesses. Both of these were easily measured to good accuracy. The absolute

normalization was much more complex. The estimated error in the absolute normalization is $\pm 50\%$. This large figure was caused by the very large number of calibration factors used in calculating the number of incident photons.

Another source of error was the presence of significant amounts of ^{73}Ge in the target material, especially in the ^{76}Ge target. The other isotopic impurities were corrected by a simple linear matrix inversion on the cross sections. However, the cross sections for ^{72}Ge and ^{76}Ge may be as much as 3% too large due to the presence of ^{73}Ge .

The transition from betatron to synchrotron action within the accelerator cycle was a source of systematic error as high as 5% at the lower energies. Some electrons within the donut were not captured by the synchrotron orbit, and caused a secondary bremsstrahlung beam with endpoint energy of about 4 MeV. This secondary burst caused extraneous ionization in the ionization chamber, and therefore an overestimate of the beam intensity. The size of this secondary beam was measured experimentally with the scintillation monitor as a function of energy and of total ionization. The dose measured by the chamber was adjusted downwards by means of this calibration.

Deadtime was still another source of systematic error. The deadtime of the counting system was measured to be on the order of 200 nanoseconds, or about one per cent of the

20 μ sec counting interval. The correction in absolute counts was insignificant, since the count rate was limited to about 0.4 per burst. Instead, the major effect was to cause some two-neutron bursts to fall in multiplicity bin one instead of bin two, and so on for all the bins. This leakage from higher to lower bins was a source of error for the separation of neutron multiplicities. The calculated adjustment for the $(\gamma, 2n)$ rate was only 0.9%. The effect of deadtime on the $3n$ and $4n$ rates was more serious, but was not significant for the purposes of this experiment.

The data analysis procedure is itself a possible source of error. It is a characteristic of all analyses of photonuclear yield functions that "secondary resonances" may in reality be oscillations of the particular solution of the yield equation. The Least Structure method is generally reliable, but does require considerable care in error estimates (30). The danger of spurious structure is not very great in this particular experiment, since high resolution is not a goal. The second resonances observed in the (γ, n) cross sections could be seen directly before unfolding as sudden increases in the reduced yields. Such "breaks" in the yield curves are more difficult to see at higher energies in the "raw" yields $\alpha(E)$, but can be observed in the unreduced yields if they occur at median energies. The second resonance of ^{74}Ge was in fact observed in the raw yields during

the data-taking phase of the experiment, and extra bombardments were performed to verify the reproducibility of the yields in the neighborhood of the second resonance. This observation and check during the experiment, when coupled with visible breaks in all eight reduced yields, independent confirmation of the $^{76}\text{Ge}(\gamma, n)$ cross section (31), and the systematics discussed in the next chapter, provides compelling evidence for the reality of the structure found in the (γ, n) cross section.

There is one non-physical factor which is introduced into the solutions by the Least Structures procedure. The peaks of the resonances are smoothed into shapes which are not as sharp as the physical resonances. This same smoothing also translates a steep rise or fall in cross section into a less steep slope and an overshoot. This effect can cause negative solutions on either side of a resonance. Such overshoots did indeed occur (e.g. the single neutron cross section of ^{72}Ge) and are a distortion which is characteristic of this type of analysis (29).

Comparison with Other Data

The absolute normalization does not agree very well with that of Borello et al. (12) or Ferrero et al. (13). However, the resonances observed in other nuclei of similar mass are on the order of 100 mb (10, 11), in reasonable agreement with this experiment. The ratio of the strengths of the single

neutron giant resonances of ^{76}Ge to that of ^{70}Ge is about two to one, just as was found by Borello. The ^{70}Ge single neutron resonance peaks at 18 MeV and is about 8 MeV wide at half maximum, in good agreement with the results of Ferrero. Neither of these older measurements extends to a sufficiently high energy to confirm the secondary single neutron resonances. However, some preliminary results are available from the activation-method companion to this experiment. McCarthy (31) has found a definite secondary resonance in the single neutron cross section of ^{76}Ge at 25 MeV. This activation experiment was a completely independent measurement using a different technique, and therefore is a very valuable confirmation of the present experiment.

CHAPTER 9. DISCUSSION AND CONCLUSIONS

The energies and estimated strengths of the resonances are summarized in Table 4. The strengths were calculated by numerically integrating the peaks. Extrapolation was necessary to estimate the strengths of the second resonance in ^{70}Ge , since the cross section is still non-zero at 40 MeV. Also, extrapolation of the main resonance of ^{76}Ge was necessary to separate the doublet. In any case, all the strengths are estimates, and have accuracies of 10% at best.

The energies of the main single neutron resonances are reasonable for this mass range. The giant resonance of ^{70}Ge is not as strong as those of the other isotopes, but ^{70}Ge has the least neutron excess of all the stable germanium isotopes, and there may well be more strength in the proton channel.

In any case, when all strengths are added and are divided by NZ/A , the result is about 90 MeV-mb in each of the four nuclei. The dipole sum rule (18) predicts a value on the order of 60 MeV-mb. In view of the 50% accuracy of the absolute normalization, the values in the table are not unreasonable. The fact that the total strengths are almost constant when divided by NZ/A is even more significant, and the conclusion is that the total strengths show the expected systematics.

Table 4. Resonance energies and strengths^a

Nucleus	T ₃	Last Neutron Shell	Resonance Energies			Resonance Strengths			Total	S ₁ +S ₂	Total	S ₁ +S ₂
			E ₁	E ₁ '	E ₂	S ₁	S ₁ '	S ₂			NZ/A	NZ/A
⁷⁰ Ge	3	5/2 ⁻	18	35	26	854	610	61	1525	671	87.6	38.6
⁷² Ge	4	1/2 ⁻	17.5	34	27.5	849	289	418	1556	707	87.4	39.7
⁷⁴ Ge	5	9/2 ⁺	17	31	24	883	434	364	1681	798	92.3	43.8
⁷⁶ Ge	6	9/2 ⁺	17	26	26	908	159	710	1777	869	96.1	47.0

^a All energies are in MeV; all strengths are in MeV-mb.
E₁ and S₁ refer to the (γ,n) giant resonance, E₁' and S₁' refer to the second resonance, and E₂ and S₂ refer to the (γ,2n) resonance.

The second resonances in the single neutron channels are more puzzling. This is clearly not the isospin splitting discussed in the introduction. The separations between the main and second resonances are more than double the splitting calculated by Leonardi (17). Moreover, the splitting decreases as T_0 increases, the opposite of the systematics predicted by isospin. And, the second resonance in at least three cases is much too large to be the T_1 resonance. The isospin question cannot be settled until the results of the other two experiments are available, but it is evident in any event that some other explanation must be sought for the second resonances observed here.

Ground state deformation of a nucleus will result in splitting of the giant resonance. However, the ground state quadrupole moment has been checked by microwave techniques (32), and was too small to be detectable. Deformation is therefore not a satisfactory explanation.

A third possible explanation is that the second resonance is in the (γ, np) channel, since this experiment had no way of distinguishing single neutron from np decay. Indeed, the np resonances must appear in these results if they are large enough with respect to the single neutron resonances. The second resonances observed are quite large to be np resonances, but the $2n$ resonances are surprisingly large in these nuclei also, and therefore it is well not to place too much

reliance upon estimates. Speculation on this possibility serves no useful purpose; the activation experiment will soon settle the question. Preliminary results have already identified the ^{76}Ge second resonance as a true (γ, n) resonance (31).

The second resonance becomes smaller and comes down in energy as the nuclear neutron number is increased. The $2n$ resonance does not seem to change significantly in energy, but does become stronger as the neutron number increases. A most interesting observation from table 4 is that the sum of the second single neutron resonance strength and the $2n$ resonance strength is almost constant. In fact, the sum seems to vary as NZ/A . Such systematics seem to indicate that the $2n$ and the second single neutron resonance are two competing decay channels from the same giant resonance state. The only measurements in this mass region comparable to this work are the (γ, n) and $(\gamma, 2n)$ cross sections of separated zirconium isotopes, measured by Berman et al. (33). Table 5 summarizes the cross section strength reported in reference 33. Only one resonance is seen in the single neutron channel. Here, the $2n$ channel is gaining strength at the expense of the single (γ, n) giant resonance. The competition between the second resonances and the $2n$ resonances may be an aspect of an effect which could be studied in many nuclei in this region of the periodic table.

Table 5. Cross section strengths of zirconium^a

Nucleus	$\sigma_{\text{int}}(\gamma, n)$	$\sigma_{\text{int}}(\gamma, 2n)$	Total
⁹⁰ Zr	960	100	1060
⁹¹ Zr	880	200	1080
⁹² Zr	650	450	1100
⁹⁴ Zr	430	580	1010

^aAll values are taken from reference 33. All strengths are in MeV-mb.

Suggestions for Future Work

The data collected in this experiment is still incomplete in that the other reaction channels are necessary to completely understand the formation and decay of the giant resonance states in germanium. However, it is very evident that the second resonances in the single neutron cross sections are not easily explained by any current theory. A similar effect seems to take place in the zirconium reactions. This suggests that this mass region deserves further study. ^{75}As is the only mono-isotopic element in the region. Experiments have concentrated on arsenic for this reason, but such experiments cannot show systematics such as were discovered here. It would be desirable to investigate these effects in elements such as gallium, selenium, and bromine. Some higher energy work (up to about 60 MeV) on nickel, copper, and zinc would also be helpful. Experiments at such high energies are rare, and comparison of germanium and zirconium seems to indicate that the second resonance, if present, is probably even higher in energy in these lighter nuclei than it is in germanium.

The medium mass region of the periodic table has not been very well studied. It is the region where nature makes a transition from the single particle effects characteristic of light nuclei to the collective effects observed in heavy nuclei. It can be concluded that this transition region ex-

hibits structures which cannot be easily explained in terms of present theories, but which nevertheless follow simple rules. It would be most interesting to study this mass region more extensively.

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ACKNOWLEDGEMENTS

I am indebted to many people whose participation made this experiment possible. I would especially like to thank the following people for their contributions: Dr. R. C. Morrison, my major professor, whose guidance has contributed greatly to the success of this experiment. Mr. George Holland and Dr. A. J. Bureau, who kept the synchrotron operational and attended to many of the operational details. Mr. Jon McCarthy, for many hours of fruitful discussion and help with the operation of the synchrotron. Mr. Charles Gale, Mr. David Fromme, and Dr. Gary Clark, for their careful work during the data taking. Mr. Stanislaw Kocinski, for his suggestions in the use of the neutron detector. Mr. Bernard Beaudry, for his invaluable suggestions for the reduction of the isotopes. Mr. E. G. Keith, for many suggestions in the design of the computer programs.

I particularly wish to thank Mr. Orien B. Swanson, for his interest and unfailing encouragement during my graduate program.