

Determination of the Photo-Nuclear Cross Sections $S^{32}(\gamma, d)P^{30}$, $S^{32}(\gamma, np)P^{30}$, and $P^{31}(\gamma, n)P^{30}$

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The photo-nuclear cross sections for the gamma-deuteron and gamma-neutron proton, reactions of the parent isotope S^{32} were measured. An attempt was made to discover whether these reactions are sufficiently strong to offer enough competition to the gamma-neutron reaction to account for the shape of the latter cross section. Sulfur samples were irradiated in the photon beam from the University of Saskatchewan's 22-Mev betatron and an activity curve was obtained. The activity curve was analyzed to give a cross-section curve that had a peak value at 26 Mev and an indication of the possible existence of another peak at 24 Mev. The observed threshold energy was 19.15 ± 0.20 Mev, a value that is consistent with the calculated value for a gamma-deuteron reaction. The cross-section curve can be interpreted to be the result of a superposition of a gamma-deuteron cross section and a

gamma-neutron proton, cross section with peak values of 0.7 ± 0.1 and 1.5 ± 0.2 millibarns, respectively. The half-width energy of the combined cross-section curve was roughly 3 Mev. The integrated cross section gave a value of 4 millibarns-Mev and it is estimated that this value is too low to offer appreciable competition to the gamma-neutron reaction.

The gamma-neutron cross section from the parent isotope P^{31} was also measured. This curve had a threshold at 12.4 ± 0.2 Mev and the peak occurred at 19 Mev. The peak value was 17 ± 2 millibarn and a half-width energy of 7.6 Mev.

It is observed that all cross sections for photo-nuclear disintegrations so far measured in this laboratory show an energy interval of about 6 Mev between threshold and peak.

I. INTRODUCTION

RECENT measurements¹⁻³ of (γ, n) cross sections of nuclides have shown that the cross section *versus* energy curves have shapes similar to resonance curves; that is, they are peaked. In general, the half-width energy of a gamma-neutron cross section can be taken to be of the order of 6 Mev.

In the light of the present conceptions of nuclear level spacing, an explanation based on a simple resonance theory is not possible. If the observed cross section is due to resonance over a single nuclear energy level, the separation of the levels would need to be of the order of 36 Mev. Experiments³ have shown the existence of only one peak for the $Cu^{63}(\gamma, n)$ reaction over the energy range from 10 to 100 Mev.

According to the continuum theory of nuclear reactions, one assumes that contributions to the reaction come from a great many nuclear levels. This theory is based on the Bohr assumption that a nucleus absorbs an incident particle a to form a compound nucleus and the compound nucleus subsequently decays with the emission of a particle b . The mode of decay is assumed to be independent of the mode of formation. The cross section for the reaction can then be expressed, following Blatt and Weisskopf,⁴ as:

$$\sigma(a, b) = \sigma_c(a)G_c(b), \quad (1)$$

where σ_c is the probability of forming the compound nucleus and is equal to the product of the probability that the particle will reach the nuclear surface and the sticking functions; G_c is the probability that the nucleus

decays with the emission of a particle b .

$$G_c = \frac{\text{probability for the emission of } b}{\Sigma \text{ probabilities for decay by all modes}} \\ = F_b / \Sigma_c F_c. \quad (2)$$

According to the calculations of Weisskopf and Ewing,⁵ the F_c functions increase monotonically with energy.

Thus, a cross-section curve for a particular reaction can exhibit a peak only if the denominator of Eq. (2) becomes large beyond a certain energy owing to the effect of strongly competing reactions or $\sigma_c(a)$ exhibit this energy dependence.

It was suggested by Wäffler and Hirzel⁶ that a $(\gamma, 2n)$ reaction might supply sufficient competition to the (γ, n) process to account for the shape of the latter cross section. Measurements in our laboratory indicate that at least for Cu^{63} this reaction is insignificant in the required energy range.

It was therefore of some interest to determine whether sufficient competition could be supplied by (γ, d) and (γ, np) reactions.

II. DISCUSSION OF THE EXPERIMENT

The isotope S^{32} was chosen for a target. The (γ, n) reaction gives the short-lived S^{31} isotope, whose activity dies away quickly, while the (γ, d) reaction gives P^{30} , with the convenient half-life of 2.55 min. Cadmium shielding was used to eliminate the 5-min sulfur activity formed by slow neutron absorption. All other unwanted contributions to the activity were found to be negligible.

Sulfur flowers were heated to the liquid state and poured into a one-inch diameter glass tube. The result-

¹ Johns, Katz, Douglas, and Haslam, *Phys. Rev.* **80**, 1062 (1950).

² B. C. Diven and G. M. Almy, *Phys. Rev.* **80**, 407 (1950).

³ G. C. Baldwin and G. S. Klaiber, *Phys. Rev.* **73**, 1156 (1948).

⁴ J. M. Blatt and V. F. Weisskopf, *The Theory of Nuclear Reactions*, Technical Report No. 42, Laboratory for Nuclear Science and Engineering, M.I.T.

⁵ V. F. Weisskopf and D. H. Ewing, *Phys. Rev.* **57**, 472 (1940).

⁶ H. Wäffler and O. Hirzel, *Helv. Phys. Acta* **21**, 200 (1948).

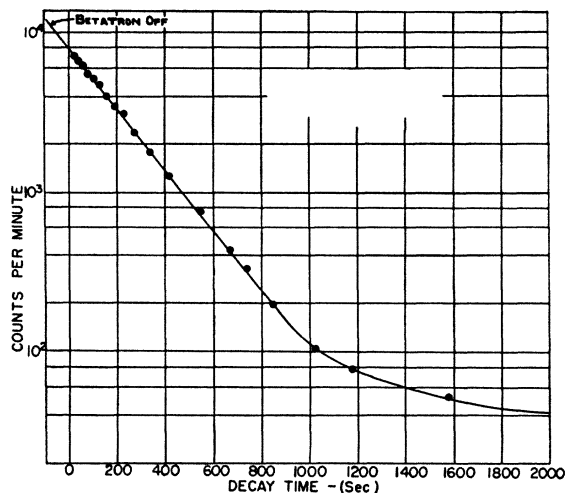


FIG. 1. Decay curve of P^{30} . The maximum energy of the betatron was 24 Mev. The dose rate was 538 r/min and the irradiation time was 8 min. The measured half-life is 2.62 min. Subtraction of the slight amount of unidentified long-lived activity gives a half-life in good agreement with the listed value of 2.55 min.

ing cylinder was cut into samples of thickness equal to the diameter of a Victoreen r-meter. The samples weighed about nine grams each.

The samples were irradiated in the photon beam from the University of Saskatchewan's 22-Mev betatron.

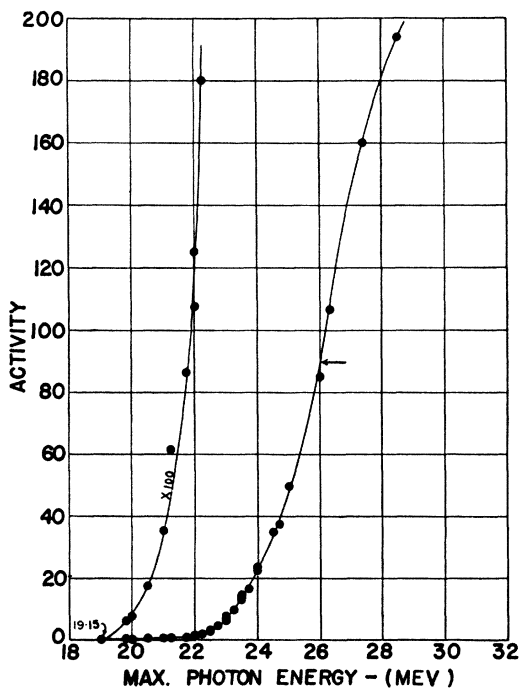


FIG. 2. Activation curve for the reaction $S^{32}(\gamma, n_d^p)P^{30}$ as obtained with the betatron. The ordinates of the figure are in arbitrary units, but the point at 26 Mev corresponds to 22×10^{-19} events/100r/atom. The region immediately above the threshold is re-drawn to a 100 \times scale. The threshold energy is 19.15 ± 0.20 Mev.

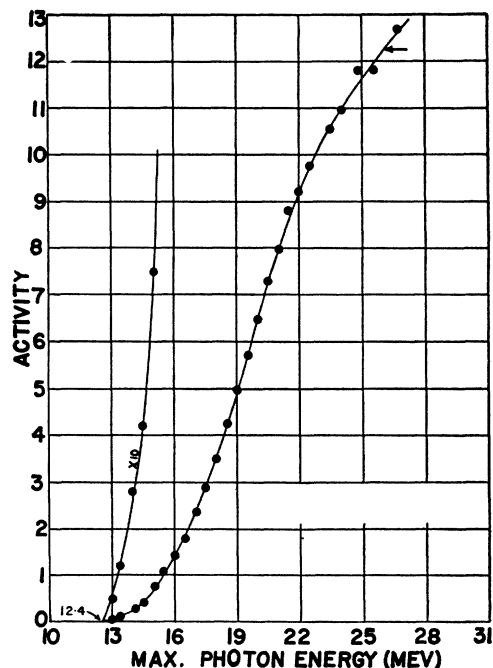


FIG. 3. Activation curve for the reaction $P^{31}(\gamma, n)P^{30}$ as obtained with the betatron. The units of the ordinates are arbitrary but the point at 26 Mev corresponds to 63×10^{-18} events/100r/atom. The region immediately above the threshold is re-drawn to a 10 \times scale. The threshold energy is 12.4 ± 0.2 Mev.

The experimental arrangement was similar to that described in a previous paper.¹

Horizontally, the samples subtended the same angle to the x-ray source as did the r-meter used for monitor calibration. In the vertical direction they subtended about twice the angle of the r-meter. This arrangement is suitable if it is assumed that the x-ray beam swings a negligible amount (with temperature, pressure, etc.) in the vertical plane during irradiation.

The maximum energy of the betatron bremsstrahlung was held constant to within 0.1 Mev through the use of an integrator-expander circuit⁷ for energies up to 25 Mev. The energy calibration was based on the thresholds for (γ, n) reactions in Cu^{63} and C^{11} as reported by McElhinney.⁸ Points above 25 Mev were taken at night, when the line frequency was particularly stable, the energy being determined from the voltage induced in a leg turn on the betatron. The betatron monitor was calibrated against the r-meter, both before and after each irradiation, and the samples were counted on thin plastic films in front of the window of an end-on Geiger counter.

Decay curves were taken with the use of an automatic time-stamping device and the saturated 2.55-min activity per gram per 100 r (measured in Lucite) was obtained at each energy. A sample decay curve is shown

⁷ Katz, McNamara, Forsyth, Haslam, and Johns, Can. J. Research 28, 93 (1950).

⁸ McElhinney, Hanson, Becker, Duffield, and Diven, Phys. Rev. 75, 542 (1949).

in Fig. 1. The resulting activity curve is shown in Fig. 2. A threshold energy of 19.15 ± 0.20 Mev was obtained.

Smaller sulfur disks were cast to determine the absolute ordinates of Fig. 1. Corrections for self-absorption, backscattering, etc., were made and the counting geometry factor was determined by comparison with a standard RaE source.

Similar experimental arrangements, but powdered samples, were used to determine the activations curve for the reaction $P^{31}(\gamma, n)P^{30}$. This curve is shown in Fig. 3.

The measured gamma-neutron threshold is 12.4 ± 0.2 Mev, a value in excellent agreement with a recent measurement by McElhinney, *et al.*⁸

The two cross-section curves were obtained from the pure β^+ activity curves through the use of a progressive-smoothing method previously described.¹ This analysis yielded points on the cross-section curve one Mev apart.

III. DISCUSSION OF THE (γ, n) CROSS SECTION

The (γ, n) curve for P^{31} has the same general features, with one exception, as had been observed previously for other (γ, n) reactions. The exception is the steep rise and large tail of the cross-section curve (Fig. 4). It has a peak value of 17 ± 2 millibarns. The half-width is 7.5 Mev, and the peak occurs 6.6 Mev above the threshold. The value of the cross section rises very steeply near the threshold. The integrated cross section is 120 mb-Mev.

The neutron yield/mole/r for the gamma-neutron reaction at 18 and 22 Mev has been calculated. These calculations gave values of 1.1×10^5 and 2.92×10^5 , respectively. This is in very good agreement with the values of 1.1×10^5 and 3.2×10^5 obtained by Price and Kerst.¹⁰

IV. DISCUSSION OF THE (γ, n, d, p) CROSS SECTION

An attempt was made to calculate the value of the (γ, d) threshold energy using known values of reaction energies and decay energies. The values were taken from Mattauch and Flammersfeld's tables,¹¹ Seaborg's tables,¹² and the paper on thresholds by McElhinney, *et al.*⁸

Table I gives the reactions, reaction energies, and decay energies used in the calculations. Four separate paths of calculation were chosen as indicated in Fig. 1 of a recent letter to the editor by one of the authors.¹³ of 272.12 Mev.

The four paths of calculation yielded values of 18.98, 19.27, 19.17, and 18.89 Mev. The mean value of these is 19.08 Mev, and it is in excellent agreement with our experimentally determined value of 19.15 Mev.*

⁹ C. Magnan, Ann. de physique 15, 5 (1941).

¹⁰ G. A. Price and D. W. Kerst, Phys. Rev. 77, 806 (1950).

¹¹ J. Mattauch and A. Flammersfeld, "Isotopic report," special issue of the Z. Naturfor., 1949.

¹² G. T. Seaborg and I. Pearlman, Revs. Modern Phys. 20, 585 (1948).

¹³ A. S. Penfold, Phys. Rev. 80, 116 (1950).

* The measured threshold for the (γd) reaction would normally be expected to be above that calculated from mass differ-

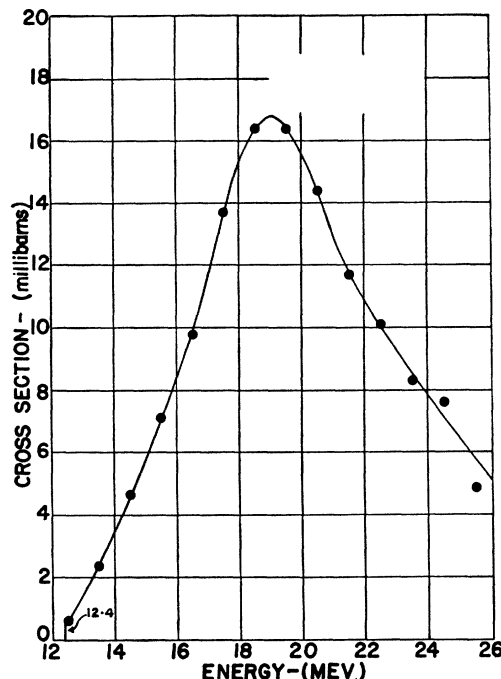


FIG. 4. Cross-section curve for the reaction $P^{31}(\gamma, n)P^{30}$. The curve shows one peak with a maximum value of 16.6 mb and a half-width of 7.6 Mev. A finite beginning of 0.5 mb is evident. The peak occurs 6.6 Mev above the threshold.

The (γ, np) threshold is higher than the (γ, d) threshold by the amount of the binding energy¹⁴ of the

TABLE I. Nuclear data used in the calculations.

| Reaction | Reaction energy (Mev) | Decay energy (Mev) |
|------------------------------|-----------------------|--------------------|
| $S^{31} \rightarrow P^{31}$ | | 3.85 |
| β^+ | | |
| $P^{30} \rightarrow Si^{30}$ | | 3.50 |
| β^+ | | |
| $S^{32}(\gamma, n)S^{31}$ | -14.8 | |
| $P^{31}(\gamma, n)P^{30}$ | -12.35 | |
| $Si^{29}(d, n)P^{30}$ | 3.38 | |
| $Al^{27}(\alpha, n)P^{30}$ | -2.93 | |
| $Si^{30}(d, n)P^{31}$ | 4.56 | |
| $Al^{27}(\alpha, p)Si^{30}$ | 2.26 | |
| $S^{32}(n, \alpha)Si^{29}$ | 1.2 | |

ences because of barrier effects. Recent discussions between one of the authors (L. K.) and Mr. A. G. W. Cameron of this laboratory has led them to propose the following process to account for the good agreement between the measured and calculated thresholds. It may also account for the large $(\gamma d)/(\gamma p)$ ratio in copper found by P. R. Byerly and W. E. Stephens [Phys. Rev. 81, 473 (1951)]

The (γ, d) process near the threshold may be visualized as similar to an inverse Oppenheimer-Phillips process. The photon energy is assumed to be largely absorbed by the proton of a "deuteron" in the nucleus. The proton thus excited may penetrate the coulomb barrier relatively easily and more radially outward. A redistribution of energy may then take place in which the neutron remaining inside the nucleus is raised far enough to escape from the remaining potential well. The deuteron would thus escape in two stages, and the product of the separate probabilities for each stage may well be much greater than the probability for the penetration of the coulomb barrier by the deuteron as a unit.

¹⁴ R. E. Bell and L. G. Elliot, Phys. Rev. 79, 282 (1950).

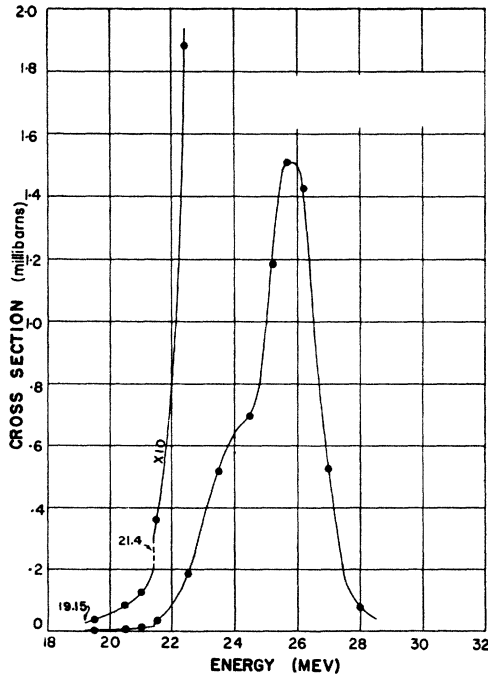


FIG. 5. Cross-section curve for the reaction $S^{32}(\gamma, ndp)P^{30}$. The curve shows two peaks and is interpreted to be the superposition of a (γ, d) and a (γ, np) cross section. The enlarged plot of the region near the threshold shows a discontinuity. This is believed to be the result of a small finite beginning of the (γ, np) cross section, similar to that for the (γ, d) cross section. The maximum value of the (γ, np) cross section is 1.5 mb and the maximum value of the (γ, d) cross section is somewhat smaller than 0.7 mb.

deuteron; 2.230 Mev. Thus the (γ, np) threshold occurs at 21.4 Mev.

It is to be noted that the curve of Fig. 5 exhibits a small "finite" beginning of 0.03 mb. Such a rapid rise of cross section near the threshold has been observed by us in all (γ, n) reactions investigated and is to be expected for these reactions from theoretical considerations.

Since the (γ, d) cross-section curve has a "finite" beginning, it is reasonable to assume that a (γ, np) curve would have a finite value also. In fact, one can imagine that a discontinuity, corresponding to this quantity, occurs between 21 and 21.5 Mev in Fig. 5.

It is also noted that the cross-section curve exhibits a bump in the neighborhood of 24 Mev. None of the (γ, n) cross-section curves examined by the authors have exhibited this shape; and they take it to indicate the presence of two reactions, (γ, d) and (γ, np) . The peak value for the cross sections may be estimated at about 0.7 ± 0.1 mb for (γ, d) and 1.5 ± 0.2 mb for (γ, np) .

To estimate the total plot cross section in S^{32} including all reactions, the following computations were made. A recent paper by Waffler and Hirzel⁶ lists the value of the cross section for the reaction $S^{32}(\gamma, n)S^{31}$ as 0.041 that of the value for the reaction $Cu^{63}(\gamma, n)Cu^{62}$ at 17.5 Mev. According to measurements made in our labora-

tory¹ the peak of the $Cu^{63}(\gamma, n)Cu^{62}$ cross section occurs at about 17.5 Mev and has the value of 110 mb. Since the threshold for the $S^{32}(\gamma, n)S^{31}$ reaction occurs at 12.35 Mev and by analogy to a typical (γ, n) curve, the value of the $S^{32}(\gamma, n)S^{31}$ cross-section curve at the peak may be estimated to be of the order of $110 \times 0.041 \times 2 = 9$ mb (the factor of 2 being included to correct for the position of the peak). If we assume that the half-width for this reaction is approximately 6 Mev, then the integrated cross section for the (γ, n) reaction is 54 mb-Mev. From Fig. 5 the value of the integrated $(\gamma, d) + (\gamma, np)$ cross sections is 4.1 mb-Mev.

For the integrated (γ, p) cross section from S^{32} one can make an estimate of 1/10 that of the gamma-neutron value.[†] This is probably a liberal estimate¹⁵ (Curtis and Hornbostel¹⁶ find (γ, p) for rhodium to be ≈ 0.4 mb).

Recent measurements at Chalk River¹⁷ indicate that the integrated (γ, α) cross section can be estimated to be around 3 mb-Mev. The (γ, γ') reaction is believed to be small. Thus adding these values we find

$$\sum_b \int \sigma(\gamma, b) dE = 66 \text{ mb-Mev.}$$

According to the theory of Levinger and Bethe¹⁸ the "resonance" shape of the cross-section curve is due to the resonance nature of $\sigma_c(a)$ of Eq. (1) and the value of the integrated cross section summed over all possible decay modes is

$$\sum_b \int \sigma(\gamma, b) dE = 0.015(1 + 0.8x)A, \quad (3)$$

where x is the fraction of attractive exchange forces contributing. With $x=0$, and for our case of $A=32$, Eq. (3) gives 480 mb-Mev, a value which is not in agreement with our estimates.

We conclude that the combined effects of the (γ, d) and (γ, np) reactions are not so great as to offer an appreciable amount of competition to the (γ, n) reaction, nor does the measured integrated cross section summed over all photon reactions agree with the theory of Levinger and Bethe. Perhaps the mode of decay of the compound nucleus cannot be assumed to be independent of the mode of formation. Arguments in support of this conclusion were recently advanced by Weisskopf.¹⁹

Workers in our laboratory have measured the (γ, n) cross sections for Cu^{63} , Cu^{65} , Ta^{181} , Sb^{123} , Sb^{121} , P^{31} , and Zn^{64} (unpublished). These cross sections, along with the (γ, ndp) cross sections of this paper all show one notice-

[†] Recent measurements indicate that the integrated (γ, p) and (γ, n) cross sections are about equal in this region of the periodic table. Our conclusion regarding the relative magnitudes of the measured and theoretical integrated cross sections will not be altered by this change.

¹⁵ L. I. Schiff, Phys. Rev. **73**, 1311 (1948).

¹⁶ Curtis, Hornbostel, Lee, and Salant, Phys. Rev. **77**, 290 (1950).

¹⁷ C. H. Millar and A. G. W. Cameron (to be published).

¹⁸ J. S. Levinger and H. A. Bethe, Phys. Rev. **78**, 115 (1950).

¹⁹ V. F. Weisskopf, Helv. Phys. Acta **XXIII**, 187 (1950).

able similarity. The energy interval between the threshold and the peak value is always about 6 Mev.

The arguments which have been presented might be summed up as follows.

(a) The idea of competing reactions cannot account for the experimentally observed shape of photo-nuclear cross-section curve of S³².

(b) A resonance theory might explain the results, but it would have to differ from present theories.

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Microwave Spectrum and Molecular Structure of GeF₃Cl

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Measurements of the microwave rotational lines of four different transitions yield B_0 values in Mc/sec as follows: 2168.52 for Ge⁷⁰F₃Cl³⁵, 2167.53 for Ge⁷²F₃Cl³⁵, 2166.60 for Ge⁷⁴F₃Cl³⁵, 2108.13 for Ge⁷⁰F₃Cl³⁷, 2107.04 for Ge⁷²F₃Cl³⁷, and 2105.98 for Ge⁷⁴F₃Cl³⁷. For all these forms $D_J \approx 0.6$ kc/sec and $D_{JK} < |1|$ kc/sec. The molecular dimensions obtained for GeF₃Cl are $\angle \text{FGeF} = 107.7^\circ \pm 1.5^\circ$, $d_{\text{GeF}} = 1.688 \pm 0.017 \text{ \AA}$, and $d_{\text{GeCl}} = 2.067 \pm 0.005 \text{ \AA}$.

I. INTRODUCTION

THE structures of but few germanium compounds are known. Only three germanium halides, GeCl₄, GeBr₄, and GeI₄, have been measured with electron diffraction.¹ Recently, structural determinations of GeH₃Cl and GeH₃Br have been made by microwave spectroscopy.² The present study reports the determination of the structure of GeF₃Cl by the latter method.

II. PROCEDURE

Most of the observations were made with a Stark modulation spectrograph³ employing 100-kc sine wave modulation superimposed on a dc bias. The cell was 20 ft in length and was of brass X band guide with a center electrode of brass supported by Teflon strips. For the most abundant isotopic combinations measurements were also made with a video-type spectrometer⁴ which did not employ Stark modulation. The dipole moment could not be evaluated with the type of modulation employed. It is evidently small, as the intensities of the rotational lines were very weak. We hope to measure the dipole moment later with an improved cell and

square wave modulator. All measurements were made with frequency markers monitored by station WWV. The accuracy was limited by distortions of the Stark modulating system and by the unresolved structure of the lines.

III. RESULTS

Table I lists the frequencies measured for the different transitions. Table II gives the most probable values for B_0 for the different species and the moments of inertia, I_B , calculated from these with $h = 6.62373 \times 10^{-27}$ erg sec. The values of D_J and D_{JK} are too small to be

TABLE I. Observed rotational frequencies of GeF₃Cl.

| Species | Transition $J \rightarrow J'$ | Frequency Mc/sec |
|--|----------------------------------|------------------------|
| Ge ⁷⁰ F ₃ Cl ³⁵ | 6→7 | 30,358.62±0.30 |
| | 7→8 | 34,694.71±0.40 |
| | 8→9 | 39,031.91±0.30 |
| Ge ⁷² F ₃ Cl ³⁵ | 6→7 | 30,344.56±0.30 |
| | 7→8 | 34,679.32±0.30 |
| | 8→9 | 39,014.14±0.30 (Stark) |
| | 8→9 | 39,013.81±0.20 (Video) |
| Ge ⁷⁴ F ₃ Cl ³⁵ | 6→7 | 30,332.58±1.00 |
| | 7→8 | 34,664.55±0.30 |
| | 8→9 | 38,996.79±0.30 (Stark) |
| | 8→9 | 38,996.78±0.20 (Video) |
| Ge ⁷⁰ F ₃ Cl ³⁷ | 6→7 | 29,512.96±0.30 |
| | 7→8 | 33,728.15±0.80 |
| | 8→9 | 37,945.47±0.90 |
| Ge ⁷² F ₃ Cl ³⁷ | 6→7 | 29,497.57±0.30 |
| | 7→8 | 33,711.21±0.30 |
| | 8→9 | 37,925.87±0.90 |
| Ge ⁷⁴ F ₃ Cl ³⁷ | 6→7 | 29,482.88±0.30 |
| | 7→8 | 33,694.43±0.30 |
| | 8→9 | 37,905.91±0.30 |

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¹ P. W. Allen and L. E. Sutton, *Acta Cryst.* **3**, 46 (1950).

² Dailey, Mays, and Townes, *Phys. Rev.* **76**, 137 (1949); Sharbaugh, Pritchard, Thomas, Mays, and Dailey, *Phys. Rev.* **79**, 189 (1950).

³ McAfee, Hughes, and Wilson, *Rev. Sci. Instr.* **20**, 821 (1949); A. H. Sharbaugh, *Rev. Sci. Instr.* **21**, 120 (1950).

⁴ W. Gordy and M. Kessler, *Phys. Rev.* **71**, 640 (1947).