Two Radioactive Substances from Magnesium after Deuteron Bombardment*

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Under bombardment by 3.3 MEV deuterons, magnesium becomes radioactive with the emission of negative electrons and gamma-rays, and with two decay periods: $10\frac{1}{4}\pm\frac{1}{4}$ min. and 15.8 ± 0.5 hours. These periods are characteristic of Mg^{27} (Fermi) and Na^{24} (Lawrence). The reactions producing these bodies are:

$_{12}Mg^{26}+{}_1H^2={}_{12}Mg^{27}+{}_1H^1$; $\quad {}_{12}Mg^{27}={}_{13}Al^{27}+\beta+\gamma$.

$_{12}Mg^{26}+{}_1H^2={}_{11}Na^{24}+{}_2He^4$; $\quad {}_{11}Na^{24}={}_{12}Mg^{24}+\beta+\gamma$.

The protons have not been looked for. The alpha-particles have a range of about 7 cm. The Mg^{27} beta-rays have 2.0 MEV maximum energy, and the accompanying hard gamma-ray has about 1.3 MEV energy. The voltage excitation functions of these two activities have been measured, and have been found to follow different laws. The Mg^{27} following the Oppenheimer and Phillips function and the Na^{24} the older Gamow function. At 3 MEV one atom becomes Na^{24} for each 9.6 atoms going to Mg^{27}. The thick target yield of Mg^{27} atoms is about 5 per 10^7 deuterons. The nuclear cross section (Mg^{26}) at 3 MEV is about 2.4×10^{-26}.

1. INTRODUCTION

IMMEDIATELY after the discovery by I. Curie and F. Joliot[1] that artificial radioactivity could be produced by alpha-particle bombardment, several laboratories followed up their suggestion that it might be produced by deuteron and proton bombardment as well. The first communication from this laboratory on this subject reported[2] the results of preliminary work with elements in the first two rows of the periodic table. It was found at that time that several of them showed strong radioactive properties after bombardment with 3-MEV deuterons. The ionizing radiations emitted were "particles of electronic mass," but were not identified as to sign. For magnesium the half-life obtained was about nine minutes.

Since these first observations the apparatus has been modified[3] so that it is now possible to bombard the target with the desired ion, either in vacuum or in air, and to remove it immediately for measurement without disturbing the vacuum in the ion production chamber. Last spring work with magnesium was resumed, with the object of measuring the half-life more accurately and investigating the activity more in detail.

2. APPARATUS

The source of deuterons was the large cyclotron in the Radiation Laboratory. At the time the magnesium work was carried on, it was adjusted to furnish deuterons of energy of about 3.3 MEV. The energy of the ions produced by a cyclotron can be approximately calculated from the magnetic field or from the wave-length employed, but for experimental purposes it is always measured by determining their range in air, after they have passed out of the apparatus through a thin aluminum window. During most of the runs this range was 11.5 cm, taken as corresponding to deuterons of about 3.36 MEV.

The target was in all cases a piece of magnesium ribbon of 99.97 percent purity. Before bombardment the surface was cleaned with emery paper and washed with petroleum ether to remove oil film. The first sample was bombarded in air, but all subsequent ones were bombarded in vacuum to avoid oxidation and contamination of the surface by radioactive oxygen.[4]

All the measurements of activity were carried out on the Lauritsen-type quartz fiber electroscope of simple design that has been used for nearly all the electroscope measurements in this laboratory. A thin aluminum window, against which the radioactive source was placed, admitted the beta and gamma-rays to the ionization chamber, and the absorption measurements

* Reported at the Los Angeles Meeting, June, 1935. Abstract 5.

[1] I. Curie and F. Joliot, Nature **133**, 201 (1934).

[2] M. C. Henderson, M. S. Livingston and E. O. Lawrence Phys. Rev. **45**, 428 (1934).

[3] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608 (1934). See also diagram in Phys. Rev. **47**, 453 (1935).

[4] E. M. McMillan and M. S. Livingston, Phys. Rev. **47**, 452 (1935).

were carried out by placing aluminum or lead sheets between the source and window. Unfortunately the intensity of the radiations was not great enough to allow the source to be more than a few centimeters from the window, and consequently the geometry of the arrangement left something to be desired.

3. The Decay Periods

A thick target of magnesium was bombarded in air for about 10 minutes by an ion beam of 3 MEV energy and about 0.9 microampere intensity. At the end of the exposure the target was at once placed at the window of the electroscope. The measured activity, as a function of time, is plotted on a logarithmic scale in Fig. 1, upper curve. The half-life determined from the linear portion, after due allowance for the natural leak and background activity of longer life, is 10.25 ± 0.25 min. The points taken in the first eight minutes show a more rapid decay as a result of the presence of radio-oxygen, which was deposited from the air around the target during the bombardment.

After about an hour and a half the successive readings showed definitely the presence of some substance with a longer life: in fact the activity two hours after exposure was still 20 times the natural leak. The target was therefore replaced in the cyclotron and the bombardment resumed with a view to intensifying the long-lived activity. After waiting two hours for the ten-minute substance to decay completely, a reading of the residual activity was taken and the decay was then followed for the next 24 hours. The half-life proved to be 15.8 ± 0.5 hours. These two periods were checked several times in the course of later runs that were made for other purposes. The errors are conservatively estimated.

To determine the sign of the ionizing particles they were observed visually in a Wilson cloud chamber that was placed in a magnetic field of a few hundred gauss.[5] All the particles, both those from the ten-minute and those from the sixteen-hour activity, proved to be negative electrons. No positrons could be detected. This fact rules out a carbon contamination as a possible source of the ten-minute activity, since after deuteron bombardment carbon becomes radioactive with the emission of positrons.

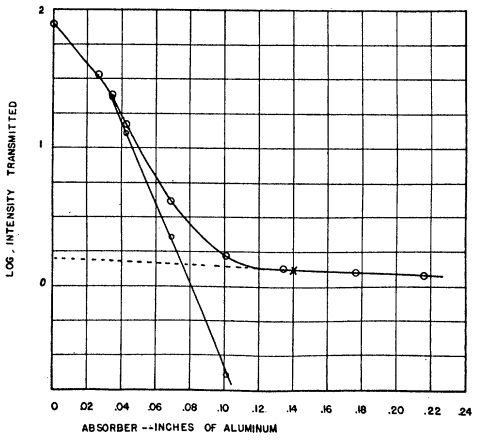

Fig. 1. Upper curve: decay of the activity of a magnesium target after deuteron bombardment. Lower curve: absorption of the beta-rays from Mg^{27} in Al.

4. The Active Substances

It is thus apparent that magnesium combines with deuterons to produce at least two radioactive substances. Their identity is at once apparent from the work of others, since the periods are characteristic of two substances already discovered: radio-sodium, Na^{24}, described by Lawrence,[6] having a half-life of 15.5 hours and produced by the action of deuterons on sodium; and radio-magnesium, Mg^{27}, discovered by Fermi,[7] half-life ten minutes and produced—among other ways—by the action of slow neutrons on magnesium.

Known types of reaction, when applied to the Mg^{26} isotope, lead at once to Mg^{27} and Na^{24}. Accordingly, we have:

$$_{12}Mg^{26} + {}_{1}H^{2} = {}_{12}Mg^{27} + {}_{1}H^{1};$$

$$_{12}Mg^{27} = {}_{13}Al^{27} + \beta + \gamma;$$

$$_{12}Mg^{26} + {}_{1}H^{2} = {}_{11}Na^{24} + {}_{2}He^{4};$$

$$_{11}Na^{24} = {}_{12}Mg^{24} + \beta + \gamma.$$

[5] I am indebted to Dr. Kurie for the use of the cloud chamber.

[6] E. O. Lawrence, Phys. Rev. **47**, 17 (1935).

[7] Fermi, Amaldi and others, Proc. Roy. Soc. **A146**, 483 (1934); Proc. Roy. Soc. **A149**, 522 (1935).

Many cases are now known in which the reaction between two nuclei leads to several pairs of end products. The two reactions just given are, however, the first in which two radioactive substances have been produced from a single substance by the use of a charged particle as a projectile. It seems reasonable to believe that this type of branching should be quite general and that not only Mg^{26} but also O^{18}, Ne^{22}, Si^{30} and other atoms may also yield more than one radioactivity under deuteron bombardment.

The protons emitted in the first reaction have not been looked for as yet. The alpha-particles, on the other hand, have already been observed, although their number was too small for accurate measurement.[8]

5. The Radioactive Radiations

The beta and gamma-rays from Na^{24} have been studied in detail by Lawrence (reference 6 and also unpublished work in this laboratory) and there seemed to be no point in repeating his experiments with an intrinsically weaker source. On the other hand, the beta-rays from Mg^{27} were measured by Fermi using a source obtained by neutron bombardment, and therefore necessarily much weaker than one obtained by the direct deuteron-magnesium reaction in the cyclotron.

An absorption curve of the radiation from a Mg^{27} source is shown in Fig. 1, lower curve. All points are corrected for decay and for natural leak. It will be seen that there is a component of hard radiation present along with the easily absorbed beta-rays. When this component is subtracted from the measured total intensity the steep linear curve results. Extrapolating this linear curve to the level of the natural leak gives for an "end point"—the maximum beta-ray range—the point marked with a cross. This end point falls at 0.95 g/cm² of Al. Using Feather's empirical rule—$E=(R+0.091)/0.511$, where E is in MEV and R in g/cm²—the maximum beta-ray energy is 2.05 MEV. Fermi found a half-value absorption thickness of 0.07 g/cm² in Al, in fair agreement with the data in Fig. 2.

The point corresponding to 2.05 MEV and 10.25 minutes, when placed on Sargent's curves of beta-ray periods and energies, lies midway between two branches. If the maximum energy were 3.8 or 1.2 MEV it would lie on one or the other of them, but the latter values seem to lie well outside the limits of experimental error.

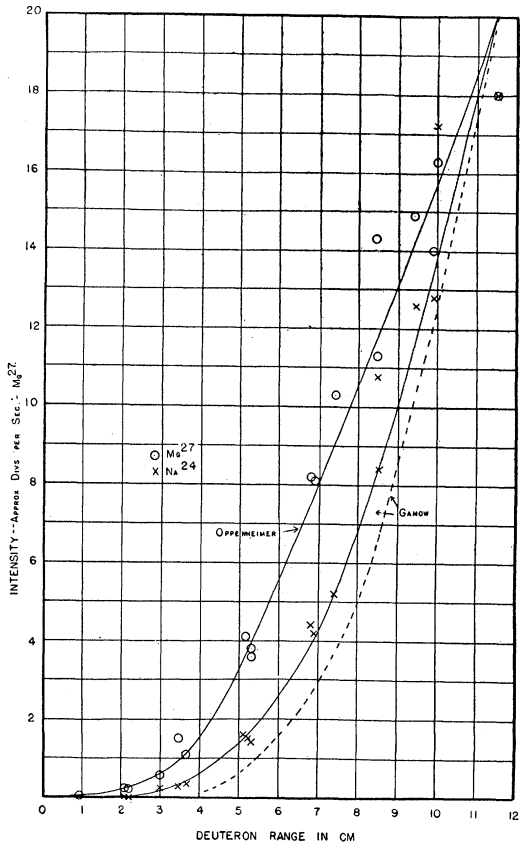

FIG. 2. The voltage transmutation functions of Mg^{27} and Na^{24}.

The absorption coefficient of the gamma-rays was determined in a separate experiment. A value of $\mu=0.74$ (cm^{-1}) of lead was obtained. The geometrical conditions were poor, but this value probably corresponds to an energy of about 1.3 MEV. From the intensity of the ionization produced by the gamma-rays, nearly 2 percent of that produced by the beta-rays, it appears that one gamma-ray is emitted for each beta-ray.

6. The Voltage Transmutation Function

The variation in the yield of transmutations as a function of the energy of the incident particle

[8] G. N. Lewis, M. S. Livingston and E. O. Lawrence, Phys. Rev. **44**, 55 (1933).

has been called the excitation function, or more properly the voltage transmutation function. The first theoretical treatment of this variation and a method for calculating it in any given case was first given by Gamow in *Atomic Nuclei and Radioactivity*. The formulas are obtained by considering, among other things, the variation with energy in the probability that the projectile as a whole penetrate the nucleus. For most of the reactions thus far studied, in which protons and alpha-particles are the projectiles, the formulas thus obtained fit the data well. However, it has been found in this laboratory[9] that the yield of transmutations caused by deuterons does not fit the Gamow curve at the higher values of deuteron energy, at least in the reactions in which a proton is emitted.

To meet this difficulty, Professor Oppenheimer and M. Phillips[10] have recently developed a theory for reactions of this type, one of the underlying assumptions of the theory being that the deuteron as a whole does not penetrate the target nucleus. Only the neutron in the deuteron enters the nucleus, the proton being "rejected." According to their calculations the transmutations should begin at a lower voltage than on the Gamow theory and should not increase as rapidly in number with increasing deuteron energy.[11]

Transmutations in which a radioactive body is produced are particularly convenient for studies of the transmutation function. Consequently, as soon as it was established that two radioactive bodies were obtained from magnesium, the attempt was made to measure the transmutation functions of both reactions. The measurement is particularly important because one of the bodies, Na^{24}, is produced by the penetration of the deuteron as a whole and the subsequent expulsion of an alpha-particle, while the other, Mg^{27}, is produced by the "rejection" of a proton and the absorption of a neutron. Thus the two activities should be different in their dependence upon voltage, the one following the older Gamow formula and the other the Oppenheimer theory.

Several runs were made to determine the transmutation functions by using the now standard method of exposing a pile of thin magnesium foils to the action of the deuteron beam and then separating the foils and measuring their activity separately. This method has several advantages: It gives the "differential" transmutation function directly, which is easier to compare with theory than the integral function; and it also ensures that all foils receive exactly the same exposure, since the deuteron beam passes through them all and since the number of deuterons that react in any given foil is a minute fraction of the total number passing through. The top foils in a pile thus act as absorbers to reduce the energy of the deuterons that reach the lower foils.

Magnesium is sufficiently malleable to be reduced with ease to foils a few ten-thousandths of an inch thick. The thickness was measured directly with a micrometer, and also more accurately, and for these experiments, more significantly by measuring their stopping power for polonium alpha-particles. For magnesium 0.001 inch is the equivalent of 2.5 cm of normal air (760 mm and 20°C). The thickness found convenient was about 1.4 cm of air, 0.00056 inch, or 0.0014 cm.

Before making an exposure each foil was carefully measured for thickness and was then stacked in a pile with a "mask" of thin copper sheet. This mask was placed on the pile to protect the edges of the foils from the deuterons and to prevent irregularities or cracks in the edges from allowing deuterons to leak through to lower members of the pile. After exposure in vacuum to about 0.8 microampere for $1\frac{1}{2}$ hours, the pile was removed, separated and the activity of each foil measured; the weakest first. After a lapse of two hours, sufficient to permit the Mg^{27} to decay completely, the foils were again measured for the Na^{24} activity. Since the Na^{24} activity was never more than about 1 percent of the Mg^{27} activity at the time the latter was measured, no correction has been applied for it. The activity of each foil, of each kind, was then corrected for decay since the end of the exposure and also corrected for any difference in thickness there might be between foils, since a thicker foil is proportionately more active.

[9] E. O. Lawrence, E. M. McMillan and R. L. Thornton—abstract in Science **81**, 421 (1935); Phys. Rev. **48**, 493 (1935).

[10] Oppenheimer and Phillips, Phys. Rev. **48**, 500 (1935).

[11] I am indebted to Professor Oppenheimer for calculating the yield curve according to his theory.

The results of the last three runs are shown in Fig. 2. To bring out the difference in transmutation function, all the activities have been made comparable by making the points at highest energy coincide. There are thus six experimental points at 11.5 cm and 18 div/sec. The ordinates for Na^{24} have been multiplied by approximately 200 to make this fit. The two solid curves are drawn in accordance with the two theories of the transmutation function, and have been adjusted to pass approximately through the mean of the points around 9 cm. The reason for using these points instead of the 11.5-cm point is that the top foil in a pile loses active atoms by recoil to the foil next beneath it, while the lower foils gain by this mechanism approximately as much as they lose.

It is readily seen that the transmutation function of the reaction producing Mg^{27} is quite different from the corresponding Na^{24} function, and the goodness of fit furnishes strong support to both theories.

The Gamow formula for the variation in number of transmutation, N, with voltage, V, is the following: $N=kV^{-1}e^{-S}$, where k is a constant of proportionality and S is defined by:

$$S=\left(\frac{4\pi e^2}{h}\right)\left\{Z_{Mg}\left(\frac{M_D}{2E_D}\right)^{\frac{1}{2}}+Z_\alpha Z_{Na}\left(\frac{M_\alpha\cdot M_{Na}}{2(M_\alpha+M_{Na})(Q+E_D^*)}\right)^{\frac{1}{2}}\right\}.$$

Z and M are the atomic numbers and masses, in grams, of the atoms indicated by the subscripts; Q, E_D and E_D^* are the energies, in ergs, of the transmutation, of the deuteron, and of the deuteron with respect to the center of mass of the system: i.e.,

$$E_D^*=E_D\cdot M_{Mg}/(M_D+M_{Mg}).$$

The first term inside the bracket takes account of the penetration of the deuteron through the nuclear barrier, the second term the penetration of the alpha-particle outwards through the sodium nuclear barrier. In actual numbers,

$$S=17.0/V^{\frac{1}{2}}+42.5/((14/13)Q+V)^{\frac{1}{2}},\quad V \text{ in MEV}.$$

In the calculation it is necessary to assume some value of Q, the energy of transmutation. An accurate knowledge of the range of the alpha-particles produced would of course lead at once to the value of Q. However, at the time the alpha-particles were found[8] the energy of the deuterons was only 1.2 MEV and the currents of the order of 10^{-8} ampere. A calculation from the Gamow formula and the yield found at higher energies shows at once that the number of alpha-particles to be expected under the old conditions was just on the limit of measurement, 2 or 3 a minute. Consequently no great accuracy is to be ascribed to the value found: 6 cm. A reconsideration of the data leads to 7 cm as the most probable value, corresponding to a Q of 8 MEV. More data are needed on this point.

In Fig. 2 two curves have been drawn for different Q's: the solid curve for $Q=10$ MEV, the dotted for 5 MEV. From the fit $Q=10\pm2$ MEV. Imperfections in the magnesium foils will lead to too high a value of Q.

One may also estimate Q from the masses and energies concerned. By writing the complete equations of the Na^{24} reaction and adding them:

$$_{12}Mg^{26}+{}_1H^2={}_{11}Na^{24}+{}_2He^4+Q.$$

$$_{11}Na^{24}={}_{12}Mg^{24}+\beta+\gamma.$$

$$(_{12}Mg^{26}-{}_{12}Mg^{24})+{}_1H^2-{}_2He^4=Q+\beta+\gamma.$$

The difference in mass between the two magnesium isotopes has not been accurately measured. In the parallel case of neon 20 and 22 the difference in mass is 1.998 ± 0.001.[12] One would offhand expect the Mg^{26} isotope to be about 2.000 heavier than Mg^{24}, since there are two more neutrons in the nucleus than are needed to form complete alpha-particles; but on the other hand, both the number of protons and the number of neutrons in both nuclei are even, so the general trend of the mass defect curve would lead to about 1.998 for the difference. Taking 2.000 ± 0.002 for this difference and Oliphant's values for H^2 and He^4, one gets $2.000+2.014-4.003=Q+\beta+\gamma\cdot=0.011\pm0.002$. From Lawrence's work[6] and from unpublished work in this laboratory, $\beta+\gamma$ is about 4 MEV, and hence Q is about 7 ± 2 MEV, in rough agreement with the value given by the Gamow curve.

In contrast with the Na^{24} reaction, no sensible

[12] Bainbridge, Phys. Rev. **43**, 424 (1933).

adjustment of the Gamow formula gives a fit with the Mg^{27} data. Even taking Q as infinite, tantamount to neglecting the second term in S, still gives a curve rising too steeply. The Gamow formula after making appropriate changes in the Z's and M's, becomes $S=17.0/V^{\frac{1}{2}}+12.3/((14/13)Q+V)^{\frac{1}{2}}$. The solid curve drawn through the Mg^{27} points has been calculated from the theory of Oppenheimer and Phillips[10] according to the equation (Eq. (12)). $N=kV^{-1}e^{-S'}$, where S' is defined as

$$S'=\frac{4Ze^2}{\hbar}\left(\frac{M}{I}\right)^{\frac{1}{2}}\overline{F(W/I)}.$$

In this equation, $\overline{F(W/I)}$ is the minimum value of $F(W/I)$, the values of which are taken from their plot of this function. I is the binding energy of the deuteron, $2M$ its mass, and W its kinetic energy. The only adjustable constant is the binding energy of the deuteron. This has been taken to be 2.0 MEV.

Although Q for the reaction in which Mg^{27} is produced does not enter the formula for the transmutation function, it is of interest to estimate it. As before, writing the equations of production and decay, one has:

$$_{12}Mg^{26}+{}_1H^2={}_{12}Mg^{27}+{}_1H^1+Q,$$

$$_{12}Mg^{27}={}_{13}Al^{27}+\beta+\gamma.$$

and adding them:

$$(_{12}Mg^{26}-{}_{13}Al^{27})+{}_1H^2-{}_1H^1=Q+\beta+\gamma,$$

Al^{27} should be at least 1.000 heavier than Mg^{26} from the mass defect curve. Thus one has $-1.000+2.014-1.008=0.006$. $\beta+\gamma$ is about 3.3 MEV. Q is therefore about 3 MEV. This consideration supplies an upper limit to the mass of Al^{27}, since if $Q=0$, $Al^{27}-Mg^{26}=1.003$.

7. The Branching Ratios and the Absolute Yields

When a target is exposed to a constant beam of deuterons, any radioactive atoms that may be formed increase in number with the expression $(1-e^{-T/t_{av}})$. Consequently, after an exposure of two hours the number of Mg^{27} atoms in a magnesium target has practically reached a constant maximum value, while the number of Na^{24} atoms is only 8.5 percent of the maximum number. This maximum number represents, for each activity, a state of affairs in which as many atoms are being formed per second as are decaying. Since the number decaying per second is what is measured as "activity," the ratio of the final maximum activities is the ratio of the probability of formation of the two kinds of atom.

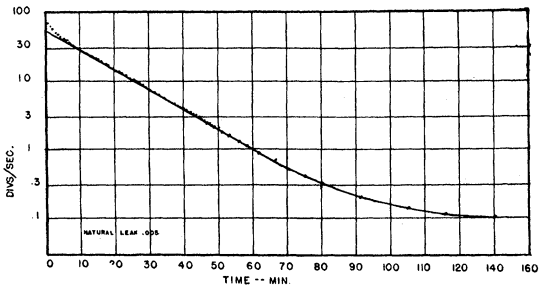

FIG. 3.

In one experiment the activities in the top foil, 1.4-cm air equivalent, at the end of a two hour exposure to 0.8 microampere were 19.5 div/sec. due to Mg^{27} and 0.18 due to Na^{24}. The saturation values of these activities, allowing for the finite time of exposure, are therefore 19.5 and 2.1. Hence for each atom of Na^{24}, $19.5/2.1=9.3$ atoms of Mg^{27} are produced. While this ratio is different at lower voltages, its variation with voltage may be readily calculated from Fig. 3.[13]

The absolute number of radioactive atoms produced is much more difficult to estimate accurately, depending as it does on the constancy of the deuteron beam, the transmission of the window, the calibration of the electroscope, and other factors. However, assuming a transmission of $\frac{1}{2}$—certainly too high—and the electroscope calibration of 2×10^4 beta-particles/sec. for 1 div/sec.; one finds that there are 1.6 Mg^{27} atoms produced for each 10^7 deuterons. The integrated yield, for a thick target, is about 3 times this, or 5 per 10^7.

Transmutation probabilities are often ex-

[13] In this and the other similar calculations no allowance has been made for the different energies of the Mg^{27} and Na^{24} beta-rays.

pressed in terms of the cross section of the nucleus, assuming that every deuteron which strikes within that cross section produces a transmutation and that all others do not. For Mg^{26}, 11 percent of the total number of atoms of Mg present, this cross section, from the data given, is 2.4×10^{-26} cm^2 at 3 MEV. Both this cross section and the yield as given above are certainly too low.

I am very greatly indebted to Professor Lawrence for the opportunity of using the cyclotron and for the facilities of the laboratory. To Professor Oppenheimer and Dr. McMillan I owe many thanks for helpful discussions and cooperation with some of the calculations. Thanks for financial assistance to the laboratory are also due the Research Corporation and the Chemical Foundation.

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The Infrared Absorption Spectrum of Germane

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The infrared spectrum of GeH_4 has been measured to determine the fundamental vibration frequencies and to investigate the rotational structure of the bands. Intense absorption regions were found, enumerated in the order of their intensities, at 4.74μ (2110 cm^{-1}), 10.7μ (934 cm^{-1}), 3.4μ (2922.7 cm^{-1} and 3031 cm^{-1}) and 2.3μ (4300 cm^{-1}) which have respectively been identified as ν_3, ν_4, $\nu_1+\nu_4$, $\nu_3+\nu_4$, and $2\nu_3$. A much weaker absorption peak near 5.7μ is identified as $\nu_2+\nu_4$. From the spacings between the principal rotation lines in the fundamentals ν_3 and ν_4 a value 7.0×10^{-40} g-cm^2 for the moment of inertia of the molecule and values 1.37×10^{-8} and 2.06×10^{-8} cm, respectively, for the Ge-H and the H-H distances are indicated.

THE recent measurements on the infrared spectrum of methane[1] and silane[2] have shown that the observed bands conform very well in a general way with the predictions of the theory for molecules of the regular tetrahedral type, but that there exists a definite complexity of the rotational structure in many of the bands which is not readily accounted for. This lack of agreement between observation and theory and the obvious desirability of finding the values of the fundamental vibrations has seemed sufficient to warrant investigating the infrared spectrum also of germane (GeH_4) a molecule which also must certainly belong to the tetrahedral XY_4 type.

A sample of chemically pure GeH_4 was obtained as a gift from Professor Warren C. Johnston of the Department of Chemistry, University of Chicago. Like silane (SiH_4) this gas should be manipulated in the absence of air or oxygen to guard against explosions. Two absorption cells of glass fitted with windows of polished rocksalt were used, one 30 cm long filled with gas to a pressure of 50 cm of Hg for measurements on weaker bands and another 10 cm long filled with gas to 10 cm Hg pressure for measurements on the more intense regions of absorption.

A set of preliminary measurements extending from 1.0μ to 13.0μ was made with a prism spectrometer of the Wadsworth type, to determine the approximate positions of the principal regions of absorption and their relative intensities. Four fairly intense regions were found, enumerated in the order of their intensities at 4.74μ, 10.7μ, 3.4μ, and 2.3μ. A much weaker peak was found near 5.7μ. Of these the three regions at 3.4μ, 4.74μ and 10.7μ have been studied under high dispersion using a prism grating spectrometer described in relation to earlier experiments, and two different echellette gratings ruled by Wood, one with 4800 lines to the inch for the 4.0μ region and another with 800 lines to the inch for the 10μ region.

[1] A. H. Nielsen and H. H. Nielsen, Phys. Rev. **48**, 864 (1935).

[2] W. B. Steward and H. H. Nielsen, Phys. Rev. **47**, 828 (1935).