Ionization of Gases by Collisions of Their Own Accelerated Molecules

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The ionization of argon, nitrogen, helium, and hydrogen by fast neutrals in their own gases has been found to occur in the 1000- to 8000-electron-volt range. The efficiencies of ionization are represented in terms of an approximate cross section. The values of the cross sections at 5000 electron volts are, respectively, for A, N_2, H_2, and He, 1.5, 0.9, 0.2, and 0.05 cm^2/cm^3 at 1-mm pressure. In this range, A shows a continuously decreasing efficiency with energy of the particle, while H_2 and He both show increasing functions. N_2 shows both a maximum and a minimum. The apparatus for production of the neutral beam and measurement of the ionization is described, as are possible defects in the measurements.

INTRODUCTION

THE ionization of gases by their own neutral atoms has been definitely established for the noble gases. Previous work[1–4] has shown the onset of ionization to occur when the impinging atom has an energy of only 30 to 70 ev. This energy, just sufficient to detach an electron from another atom, is found in each case to be roughly three times the ionization potential of the particular gas, but for other than noble gases ionization by neutrals is not known to occur, nor has the ionization function of the noble gases been measured above 1000 ev. Since the earlier work on other than noble gases was confined to less than 1000-ev energy of the impinging particle, there is no assurance that ionization will not occur in an energy range higher than this but still below the range characteristic of the alpha-particle. Experimental information in this range is necessary to an understanding of the ionizing process and its connection with that of such high energy particles as the alpha.

A preliminary report[5] of this work gives a qualitative description of the ionization of argon and nitrogen by their own neutral molecules in the 1000- to 8000-ev range. Since then these measurements have been refined and extended to more gases.

[1] R. N. Varney, Phys. Rev. **50**, 159 (1936).
[2] A. Rostagni, Nuovo Cimento **11**, 34 (1934).
[3] H. Wayland, Phys. Rev. **52**, 31 (1937).
[4] O. Beeck and H. Wayland, Ann. d. Physik **19**, 129 (1934).
[5] H. W. Berry, R. N. Varney, and S. Newberry, Phys. Rev. **61**, 63 (1942).

EXPERIMENTAL PROCEDURE

Since the discovery by Rosen and Kallmann[6] of the high efficiency of charge exchange or neutralization of positive ions as they pass through their own gas, the formation of high velocity molecular beams has been possible. This charge exchange occurs at cross sections many times the kinetic theory values for the gas molecules themselves; hence a classical collision is not necessary for the process to take place. There need be then no deviation of the positive ion or reduction in its velocity in order that the neutralization occur.

To form such a neutral beam which would have energies up to 8000 ev it is necessary first to form a beam of positive ions with the desired energy. These ions are formed in the ion gun (see Fig. 1)

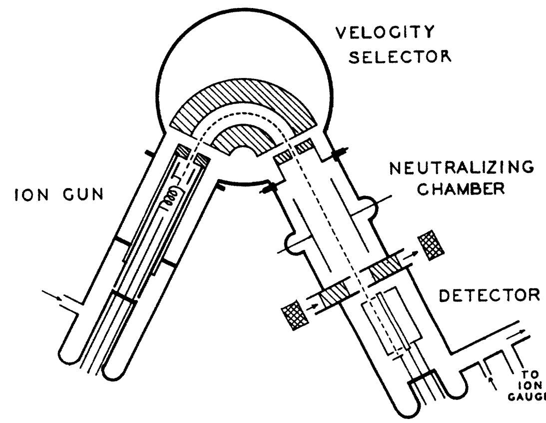

FIG. 1. Diagram of apparatus.

[6] H. Kallmann and B. Rosen, Zeits. f. Physik **61**, 61 (1930).

by electron bombardment. The electrons emitted by a spiral filament are drawn to a cylindrical electrode by a potential of about 300 volts. After passing through the cylinder they are retarded by a reverse voltage sufficient to stop and reverse their direction. The positive ions formed from the collisions of the electrons and gas atoms are drawn by this reverse field to the deep slit. The filament is coiled coaxially with the gun so that positive ions formed between it and the cylinder will be drawn through the filament thus neutralizing the negative space charge and effectively increasing the emission.

The ions are accelerated by a voltage between the deep slit and the slit in the end plate of the electrostatic "velocity" selector[7] which homogenizes the beam. From the velocity selector, the ion beam enters the neutralizing chamber through a second deep slit. This chamber is about 10 cm long and is kept field free by two parallel plates at the same potential as the deep slit. Positive ions remaining in the beam at the end of the neutralizing chamber are removed by a reverse field between the parallel plates and the plate at the end of the chamber. This plate is at a potential 90 volts more positive than the cylinder from which the ions start, and hence is capable of removing all the ions from the beam. Those which are neutralized go through a hole in this and a second plate which is 45 volts above the potential of the cylinder, thus stopping low voltage secondary electrons removed from the edges of the holes by the beam. To remove any secondaries formed in the gas in the neutralizing chamber and hence having the energy of the retarding field for the positive ions, a magnetic field is set up across the region between the plates.

Finally the beam passes through a space charge type of positive ion detector[8] where the ionizing collisions can be measured. Usually the detector is composed of two identical cylinders with tungsten cathodes placed in opposite arms of a Wheatstone bridge and operates so that the fluctuations common to both cancel out.

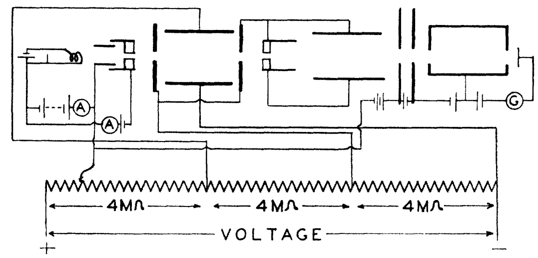

FIG. 2. Electrical circuit.

It was found in an earlier study[9] of such a detector with tungsten filament cathodes that the troublesome fluctuations observed were caused by the presence of residual gas chiefly oxygen and were entirely random in character. Further, the use of an oxide coated cathode instead of the tungsten wire proved satisfactory.[10] Hence a single cylinder with an oxide coated indirectly heated cathode is used in the present experiments, and a fixed resistor substituted for the other cylinder. After passing through the space charge detector, the beam strikes a nickel plate thereby knocking off secondary electrons. These secondaries are collected on the space charge cylinder by a potential of 10 to 45 volts and the resulting current, read on a galvanometer, provides a measure of the neutral atom beam intensity.

The gases used in this experiment are the commercial tank product and it is necessary to purify them partially. The gas is first passed over copper at 450°C for the removal of O_2 and then over phosphorus pentoxide and potassium hydroxide for the removal of H_2O and CO_2. In the case of He, in addition to the above, the gas passes through charcoal at liquid-air temperature. For H_2, the charcoal is at solid CO_2 temperature. The gas enters the system through capillary leaks. A schematic diagram of the apparatus and the electrical circuit is shown in Fig. 2.

RESULTS

The ratio of the two galvanometer readings is used as a measure of the ionization function. The galvanometer in the bridge circuit, since

[7] A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 284 (1929).
[8] R. N. Varney, Phys. Rev. **47**, 483 (1935) and **53**, 732 (1938).
[9] H. W. Berry and R. N. Varney, Phys. Rev. **57**, 1063A (1940).
[10] H. Karr and R. N. Varney, Phys. Rev. **57**, 1064A (1940).

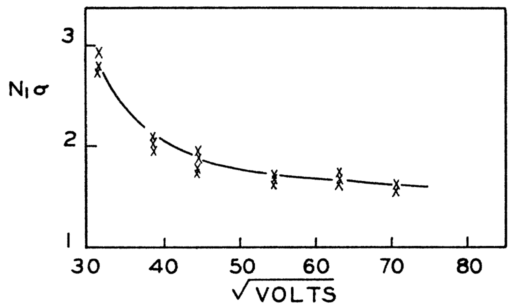

FIG. 3. Ionization of argon neutrals in argon.

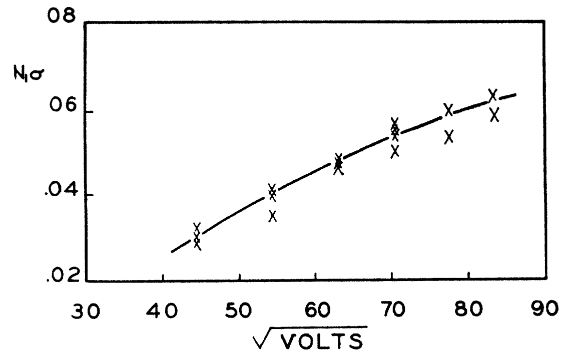

FIG. 4. Ionization of helium neutrals in helium.

it measures the unbalance of the bridge, is an indication of the change in the space charge limited current caused by the presence of positive ions. The other galvanometer reads directly the secondary electron current produced by the neutral beam on striking the plate at the end of the beam path, or indirectly the size of the neutral beam producing the ionization. The ratio of the space charge galvanometer reading to the secondary electron current then measures the relative ionization. Both galvanometer readings are taken at the same time, hence such slow fluctuations of the beam strength that the galvanometers can follow the variation will have no effect on the results.

The curve of the ionization of argon by argon neutrals as a function of the square root of the energy is shown in Fig. 3. The ordinates are the ratios of the detector galvanometer reading to the secondary electron current corrected to give an approximate value of the cross section per cm^3 of gas at 1 mm Hg pressure. This cross section is obtained by first observing the sensitivity of the space charge detector to argon ions produced by electron ionization. Since the efficiency for ionization by electrons is already known, the number of ions formed by the known electron current can be calculated. The ratio of the detector galvanometer reading to this calculated ion current is the sensitivity. This value of the sensitivity is then used to calculate the ion current produced by the neutral atom beam. Knowing the size of the neutral beam, one may readily find the cross section in units of $N_1\sigma$ in cm^2/cm^3 at 1-mm pressure.

The ionization function for A in A is seen to fall with increasing velocity. Although the various curves obtained at different times do not fall off at quite the same rate, they all have the same general shape. Figure 4 shows the curve of the ionization efficiency of helium neutrals in helium. The cross sections denoted by the ordinates are calculated in the same way as for argon. The curve shows a continuous rise but with a cross section much less than that for A.

Figure 5 shows a similar curve for the ionization of H_2 by hydrogen neutrals. Since in the ionization of hydrogen by electrons there are obtained three kinds of ions, it should be expected that the neutral beam have several kinds of neutrals. Hogness and Lunn[11] show that chiefly H_2^+ and H_3^+ are produced, hence H_2 and H_3 would be the components of the neutral beam. As the gun pressure is changed, the relative amounts of the two ions change. However, no significant change in the shape of the ionization function is observed for the different gun pressures.

For nitrogen, as in the case of hydrogen, there is more than one kind of particle in the neutral beam. The yield of N_2^+ and N^+ for different pressures has also been investigated by Hogness and Lunn.[12] They show that as the gas pressure increases, the ratio of N^+/N_2^+ increases. Figure 6 shows the efficiency of ionization of nitrogen by nitrogen neutrals. It is to be noted that the curve has both a maximum and a minimum in the range studied. Since the ionization is caused by two kinds of particles, it should be expected that the experimental curve be composite of the ionization of nitrogen by N_2 and N. When

[11] T. R. Hogness and E. G. Lunn, Phys. Rev. **26**, 44 (1925).
[12] T. R. Hogness and E. G. Lunn, Phys. Rev. **26**, 786 (1925).

different gun pressures are used, the following is found: As the pressure is lowered, the maximum of the curve becomes less pronounced with a slight shift toward the higher energy, and there is a faster rise at the high voltage end of the curve. This would indicate that the maximum belongs to ionization by N but no definite conclusions can be drawn without knowing how the neutralization process affects the relative composition of the beam.

It should be noted that the change in ionization over the whole voltage range for N_2 is slight and therefore more subject to the masking effects of the errors inherent in the experimental method. However, of the fifty-odd sets of data for nitrogen, all those which may be considered reliable—that is, those which show no change in sensitivity during the run, show a linearity in response of detector to various amounts of ionization, and have well-grouped readings—have a maximum and minimum. The position of the maximum varies from 2000 to 3000 ev, which can be accounted for in the variation of the gun pressure, as can be the different rates of rise at the high energy end of the curve.

DISCUSSION OF RESULTS

Although the ionization of argon by argon neutrals was previously known, this is the first definite evidence of the ionization of nitrogen, helium, and hydrogen by neutrals of their own gas. A previous work on argon by Varney[13] shows a continuous rise from onset to 400 ev, the limit of his experiment, Since the ionization observed in the present experiments is decreasing continuously with voltage in the range of 1000 to 8000 volts, a maximum must lie somewhere between 400 and 1000 volts. The newly obtained curve for ionization of nitrogen actually shows such a maximum.

The question of the sensitivity of the space charge detector to various ions traveling at different speeds is an important one in that it is conceivable that the drop of the ionization function for A at higher energies may be a spurious apparatus defect. The following evidence on the matter exists.[14,15] In vacuum the sensitivity of the space charge detector is as high as 50,000 for very slow ions (less than 1 volt). But the sensitivity falls off to a value as low as 10 when the ions have several hundred volts energy. With gas in the detector the sensitivity to slow ions is less, having a value for the ions formed by electron bombardment in this experiment of 5000 to 10,000. This is at a pressure of about 5×10^{-3} mm. The sensitivity to faster ions with gas in the tube is not so well known. The high efficiency of the detector for the argon ions produced by neutral atoms in the high energy range of the present experiments implies either that little kinetic energy is transferred in ionizing impacts or that the detector is retaining its sensitivity to faster ions. The questionable drop in the ionization function for A begins at neutral atom energies of about 1000 volts. It seems highly unlikely that a transfer of kinetic energy from the beam particle to the newly formed ion should set in only at these high energies. Until definite measurements of the sensitivity of the detector to fast ions in the

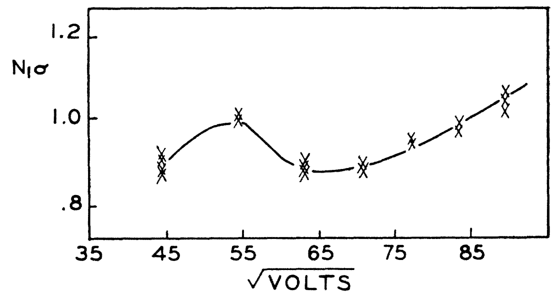

FIG. 6. Ionization of nitrogen neutrals in nitrogen.

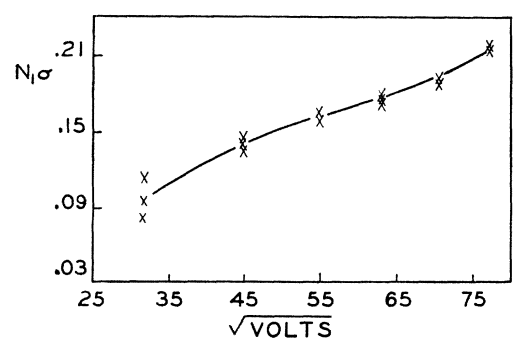

FIG. 5. Ionization of hydrogen neutrals in hydrogen.

[13] R. N. Varney, Phys. Rev. **50**, 159 (1936).

[14] R. N. Varney, Phys. Rev. **47**, 483 (1935).

[15] H. Karr, unpublished M.S. Thesis, Washington University.

presence of gas can be made, such a possibility can only be indicated. It does seem, though, that the observed experimental results are not caused by this variation of the detector sensitivity to the velocity of the ion produced.

Since the size of the neutral beam is measured by the number of secondaries that are produced when it hits a plate after passing through the ionizing chamber, the drop in efficiency may in reality be caused by a rise in the number of secondaries produced per neutral as the energy of the neutral increases. As the number of secondaries produced by a particle as it strikes a metal surface is very much a function of surface conditions, numerical results of other workers are likely to be neither reliable nor applicable. Qualitative measurements made during the present experiments seem to indicate that in the mean velocity range, about one secondary electron is produced for every two positive ions striking the surface. Rostagni[16] has examined the production of secondaries by both positive ions and neutral atoms in the energy range up to 600 volts. His results indicate that as the energy of the particle increases, the number of secondaries per ion and per neutral increases but with a decreasing rate. Judging from a rough extrapolation of the A curve showing secondaries per neutral as a function of voltage, the mean value for the 1000- to 5000-volt range would seem to be about 0.4 secondary electron per impinging particle. For helium, the value is about 0.5. This value of 0.5 was also used for hydrogen and nitrogen in the calculation of their cross sections. If there should be a variation with energy in the number of secondaries produced by the neutrals in the range of this experiment, it would, for the case of H_2 and He, merely increase their rate of rise, and similarly for N_2. It is not conceivable that this change could cause a maximum and minimum in the curves. For A, an increase in secondary emission with voltage would produce an appreciable decrease in the rate of drop, but it would not seem sufficient to remove it entirely.

It should be mentioned again that the calculation of the cross section in absolute units is only approximate. It involves a knowledge both of the number of secondaries liberated by the neutrals in order to determine the beam intensity, and also of the secondaries liberated by electrons from this plate at the end of the beam path. This last is necessary since it is not possible to maintain the plate positive with respect to the space charge cylinder; hence instead of the primary, the secondary electron current is measured. Moreover, in using the apparatus for measurement of ionization by electrons, the gas flow through the gun must be shut off and the voltages reversed. It is possible that these readjustments may affect the sensitivity of the space charge detector, changing it from its value for the ionization by the neutrals, the measurement of which is done just preceding the electron measurement.

Previous work by Rostagni[17] on ionization of A by A neutrals gives a cross section of 3 cm^2/cm^3 at 1-mm pressure at 600 volts. The results here are then in general accord with such a value, indicating that the indirect method of calculating the cross section is essentially correct. It is interesting to note that the ionization by He neutrals occurs well below the range characteristic of the alpha-particle. And also for H_2 neutrals in H_2, ionization is found at an energy much less than that for the proton. For ionization by electrons, it is found that A has the largest cross section and decreasing in order through N_2, H_2, to He. The efficiencies found here for ionization by their own neutrals are similarly arranged.

The writer wishes to express his gratitude for the guidance and help of Professor R. N. Varney, under whose direction the research was done, and also for the valuable suggestions of Professor A. L. Hughes.

[16] A. Rostagni, Zeits. f. Physik **88**, 55 (1934).

[17] A. Rostagni, Nuovo Cimento **11**, 34 (1934).