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IONIZATION BY PENETRATING RADIATION AS A FUNCTION OF PRESSURE AND TEMPERATURE

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ABSTRACT

To account for the experimental fact that the ionization of a gas exposed to γ-rays or cosmic rays is not proportional to the pressure, but approaches a limiting value for pressures of about 140 atmospheres, the hypothesis is suggested that at these high pressures the initially formed ions may remain so close together that they frequently reunite under their mutual electrostatic attractions. The probability is calculated for the ions to become separated by diffusion, and formulas are thus obtained for the saturation ionization current as a function of pressure. The most satisfactory formula is based upon an arbitrary but reasonable assumption regarding the ranges of the secondary electrons ejected by ionizing beta particles. Knowing the variation of ionization with pressure, this diffusion theory predicts a definite variation of ionization with temperature. Such a temperature variation is experimentally discovered and is in good accord with the theoretical prediction. The temperature coefficient is negligible for pressures less than 10 atmospheres, but at pressures over 100 atmospheres the ionization approaches proportionality to the absolute temperature.

IT HAS been shown by Swann[1] and his students[2] that the ionization in an ionization chamber, due to either gamma rays or cosmic rays, is not proportional to the pressure, but approaches a maximum value for pressures of about 140 atmospheres. Miss Downey[3] proposed the explanation that the ions are formed by high speed beta particles ejected from the walls of the ionization chamber, and that these beta particles are completely absorbed by the gas in the chamber if the pressure is sufficiently high. Broxon, in his 1931 paper, was able to show that this hypothesis leads to an exponential formula for the relation between pressure and ionization which agrees well with his experimental measurements. Yet there has seemed to be no explanation on this hypothesis for such facts as the following: 1. The variation of ionization with pressure when gamma rays are used is approximately the same

[1] W. F. G. Swann, J. Frank. Inst. **209,** 151 (1930).

[2] K. M. Downey, Phys. Rev. **20,** 186 (1922); H. F. Fruth, Phys. Rev. **22,** 109 (1923); J. W. Broxon, Phys. Rev. **27,** 542 (1926); **37,** 1320, (1931).

[3] Reference 2.

as when cosmic rays are used,[4] whereas on the average the beta rays ejected by gamma rays have a much shorter range than those associated with cosmic rays. 2. The ionization-pressure relation is nearly independent of the diameter of the chamber,[5] contrary to expectation. 3. In pure nitrogen the ionization is more nearly proportional to the pressure than in air.[6]

An alternative explanation of the phenomenon is that a kind of recombination may occur at high pressures, due to the fact that the electron ejected from a molecule by the ionizing beta ray may lose its initial energy through molecular collisions before it has moved far enough from the parent positive ion to escape from the effect of its electrostatic attraction.[7] In accord with the ideas underlying Thomson's theory of recombination[8] we may suppose that if the initial energy of the electron carries it beyond a critical distance, molecular diffusion will probably carry it away, and a permanent ion will be formed. If i_1 is the ionization per unit pressure when all ions remain permanent, p is the pressure, and P is the probability that an ion will remain permanent, the measured ionization may be written as

$$i = i_1 p P. \tag{1}$$

The probability P will approach unity for low pressures. It will also have a greater value at high temperatures than at low temperatures, since diffusion will be more rapid. Experiments described below show that the ionization at high temperatures is indeed greater than at low temperatures, as this statement would imply. The hypothesis of the absorption of the beta rays from the walls does not account for such a temperature variation. Moreover, the difficulties enumerated above in connection with the beta ray absorption hypothesis disappear, as we shall see, when the phenomenon is considered as one of recombination. It remains to develop a quantitative theory of ionization as a function of pressure and temperature, and to compare its predictions with experiment.

Theory of Ionization as a Function of Pressure

Let us suppose that when a beta particle passing through gas ionizes an atom, the ejected electron moves a distance r before it loses its initial energy and comes to thermal equilibrium with the surrounding molecules. The average energy spent by the beta particle in producing an ion pair is found to be 25 or 30 electron volts, so that the energy with which the electron escapes is usually much greater than the equilibrium thermal energy. Since most of the electron collisions are elastic, or nearly so, we may expect the distance r to be much larger than a molecular mean free path. As a result of molecular diffu-

[4] H. F. Fruth, reference 2, supported by later measurements.

[5] J. W. Broxon, reference 2 (1926).

[6] H. F. Fruth, reference 2.

[7] This explanation has been proposed independently by R. A. Millikan and I. S. Bowen, Nature **128**, 582 (Oct. 3, 1931) and A. H. Compton, R. D. Bennett and J. C. Stearns, Phys. Rev. **38**, 1865 (Oct. 15, 1931).

[8] J. J. Thomson, Phil. Mag. **47**, 337 (1924).

sion the positive and negative ions will now tend to move farther apart, while their mutual electrostatic attraction will tend to draw them together.[9]

To calculate the rate at which diffusion separates the ions, let us suppose that when the ejected electron comes to thermal equilibrium it forms a negative molecular ion of the same mass as the parent positive ion. If we assume the electron to remain free, the final result is unaltered. Let λ be the mean free path of either ion, and r their initial distance apart. After one free motion, the probable distance of the second ion from the initial position of the first is

$$r + \delta_2 r = (r^2 + \lambda^2)^{1/2}.$$

Similarly, the probable distance of the first ion from the initial position of the second is

$$r + \delta_1 r = (r^2 + \lambda^2)^{1/2}.$$

Thus, after one mean free time τ, the probable separation of the ions is to a close approximation, since $\lambda^2 \ll r^2$,

$$r + \delta_1 + \delta_2 r = r(1 + \lambda^2/r^2).$$

Thus,

$$\delta r = \delta_1 r + \delta_2 r = \lambda^2/r.$$

The probable rate at which the ions separate by diffusion is accordingly,

$$\frac{dr}{dt} = \frac{\delta r}{\tau} = \frac{\lambda^2}{\tau r}\cdot \tag{2}$$

The rate of approach due to electrostatic attraction may be calculated as follows. The average distance of approach of each ion toward the other during the molecular free time interval τ is $\frac{1}{2}a\tau^2$, whence during this interval r will have changed by

$$\delta r = -2\cdot\frac{1}{2}\,\frac{e^2/r^2}{m}\,\tau^2,$$

and the rate of approach is given by

$$\frac{dr}{dt} = \frac{\delta r}{\tau} = -\frac{e^2\tau}{mr^2}\cdot \tag{4}$$

The condition that the ion pair shall remain permanent is thus that

$$\frac{\lambda^2}{\tau r} > \frac{e^2\tau}{mr^2},$$

or that

$$r > r_0,$$

[9] Cf. E. B. Loeb and L. C. Marshall, J. Franklin Inst. **208**, 371 (1929), who have considered a very similar problem.

where

$$r_0 \equiv \frac{\tau^2 e^2}{m\lambda^2}\cdot \tag{5}$$

Writing $\tau = \lambda/v$, where v is given by the relation

$$\tfrac{1}{2}mv^2 = \frac{3}{2}kT,$$

we have[10]

$$r_0 = e^2/3kT. \tag{6}$$

At 20°C, using the usual values of e and k, r_0 becomes 1.88×10^{-6} cm. At this distance the field due to an electron is about 4×10^4 volts per cm, which means that ordinary electric fields will not appreciably affect the permanence of these ions.

If we are to determine the probability that the ion will remain permanent, we must now calculate the probability that the electron comes to equilibrium at a distance greater than r_0 from its parent ion. This would be possible if we knew the function $F(r)dr$ representing the probability that the electron will stop between r and $r+dr$. Unfortunately the form of this function is not known.

Let us first suppose that the distribution of distances is similar to that which applies to the diffusion of a gas molecule. If R is the probable (root mean square) distance to which the electron goes, the probability that it will stop in the range dr is then,

$$F(r)dr = \frac{4r^2}{\alpha^3(\pi)^{1/2}}\, e^{-r^2/\alpha^2}dr, \tag{7}$$

where

$$\alpha^2 = \tfrac{2}{3}R^2. \tag{8}$$

The probability of going beyond r_0 is then

$$\begin{aligned} P &= \frac{4}{\alpha^3(\pi)^{1/2}}\int_{r_0}^{\infty} r^2 e^{-r^2/\alpha^2}dr \\ &= \frac{2}{(\pi)^{1/2}}\,\frac{r_0}{\alpha}\, e^{-r_0^2/\alpha^2} + \frac{2}{(\pi)^{1/2}}\int_{r_0/\alpha}^{\infty} e^{-x^2}dx. \end{aligned} \tag{9}$$

This is also the probability of permanence of the ion pair. By Eq. (1), the saturation ionization current is however $i = i_1\, p\, P$. P may be expressed as a function of the pressure if we note that α varies as $1/p$. We may thus write

$$z \equiv r_0/\alpha = cp \tag{10}$$

where c is a constant of proportionality. Then

[10] Loeb and Marshall (reference 9) find $r_0 = e^2/6kT$. Their calculation is however for the case where λ is large compared with r_0, whereas in the present case λ is small compared with r.

$$i = i_1 p P = \frac{i}{c} \cdot zP$$
$$= C \left\{ z^2 e^{-z^2} + z \int_z^\infty e^{-z^2} dz \right\}, \tag{11}$$

where C is a new proportionality constant.

In the broken curve of Fig. 1, Eq. (11), with suitably chosen constants C and $c = z/p$, is compared with a typical set of Broxon's data[11] for the ionization by cosmic rays in air for various pressures. The evident differences between the theoretical curve and the experimental data indicate that our assumed function $F(r)$, as given by Eq. (7), does not allow as wide a range of r values as actually occur. Experiments, such as cloud expansion photographs, indicate that the electrons ejected by a fast beta ray frequently have

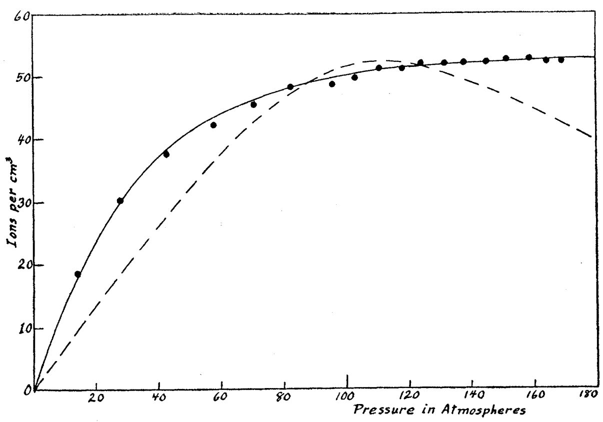

Fig. 1. Ionization as function of pressure. Data, Broxon. Broken line, Eq. 11. Solid line, Eq. 14.

ranges comparable with a millimeter at atmospheric pressure. According to Eq. (9), however, using the value of α which works best in comparing Eq. (11) with experiment, the probability that the electron should have a range greater than 0.1 mm at atmospheric pressure is only about 10^{-400}. If an arbitrary function $F(r)$ is chosen which will give a theoretical relation between i and p in accord with the experiments, the chance of observing an electron with a large range is found to be much greater.

A suitable function is found to be,

$$F(r)dr = \frac{ar}{(a^2 + r^2)^{3/2}} dr, \tag{12}$$

where a is an arbitrary constant. Writing

$$P = a \int_{r_0}^\infty \frac{rdr}{(a^2 + r^2)^{3/2}},$$

[11] J. W. Broxon, Phys. Rev. **37**, 1325 (1931), Curve II, Fig. 5.

we find on integration,

$$P = \left(1 + \frac{r_0^2}{a^2}\right)^{-1/2}. \tag{13}$$

The constant a may be interpreted as an average range of the electron, which will vary inversely as the pressure. Since r_0 is a constant at a particular temperature, we may write

$$r_0/a = hp,$$

where h is a new constant of proportionality, and we get for the ionization current,

$$i = i_1 p(1 + h^2p^2)^{-1/2}. \tag{14}$$

To fit Broxon's data of Fig. 1 we choose $i_1 = 1.3$ and $h = 0.0241$, which gives the solid curve of this figure. It will be seen that the agreement is satisfactory.

The derivation of a suitable formula, such as Eq. (14), to represent the relation between ionization and pressure is thus to a large extent arbitrary, since we have no independent knowledge of the range distribution of the ejected electrons. It is however possible thus to formulate a satisfactory relation between the ionization and the temperature.

Theory of Ionization as a Function of Temperature

In the last section we have considered the temperature as constant. Let us now remove this restriction, and consider the ionization current as a function of the density of the gas ρ and the absolute temperature T. From Eqs. (9) and (13) it becomes evident that the probability that an ion pair will remain permanent is a function of r_0/a, where a is an average range of the ejected electron and r_0 is the critical distance for forming a permanent ion pair. We note that

$$a = b/\rho, \tag{15}$$

where ρ is the density of the gas, and the proportionality constant b is presumably independent of temperature and density, From Eqs. (6) and (15) we then have,

$$\frac{r_0}{a} = A\frac{\rho}{T} \tag{16}$$

where $A = e^2/3kb$ is also independent of temperature and density. Thus the probability of permanence is

$$P = f_1\left(\frac{r_0}{a}\right) = f_2\left(\frac{\rho}{T}\right). \tag{17}$$

If I is the number of initial ions produced for unit density of the gas, for density ρ the initial ionization is $I\rho$, and the ionization current is

$$i = I\rho f_2(\rho/T). \tag{18}$$

Thus

$$i/I\rho = f_2(\rho/T). \tag{19}$$

If ρ is expressed in units of ρ_1 = density of the gas at $T=293°$ and $p=1$ atmosphere, then ρ is numerically equal to the pressure as observed under ordinary conditions. Then,

$$f_2(\rho/T) = f_2(p/T) = i/Ip, \tag{20}$$

where p is the pressure at 293°K. Using experimental data such as those of Broxon, the form of the function f_2 may be determined from Eq. (20), and by substitution in Eq. (18) the variation of i with T may be calculated.

In view of the satisfactory agreement of Eq. (14) with experiment, as shown in Fig. 1, we may use the value of P given by Eq. (13). Combining this with Eq. (16) we have

$$f_2\left(\frac{\rho}{T}\right) = \left(1 + A^2\frac{\rho^2}{T^2}\right)^{-1/2},$$

and

$$i = I\rho\left(1 + A^2\frac{\rho^2}{T^2}\right)^{-1/2}. \tag{21}$$

The temperature coefficient of the ionization is then

$$\beta = \frac{di}{idT} = \frac{A^2\rho^2}{T^3}\bigg/\left(1 + \frac{A^2\rho^2}{T^2}\right). \tag{22}$$

If we express ρ in terms of ρ_1 as described above, then $\rho = p$, and $A = e^2/3kb$, whereby comparison with Eq. (14) et seq., $b = r_0/c$. With Broxon's data for air,

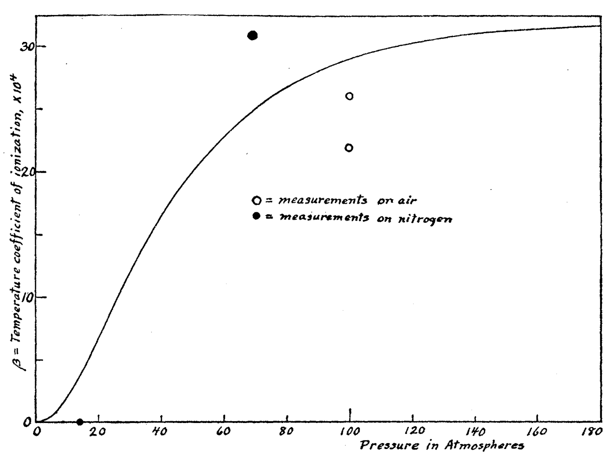

Fig. 2. Temperature coefficient of ionization of air as function of pressure. Data for nitrogen are plotted on same scale by multiplying pressure by A_{N_2}/A_{air}. Curve represents predicted values.

$c = 0.0241$, whence $b = 7.8\times10^{-5}$, and A for air becomes 7.1. For air at 293°K, the values of the temperature coefficient β for different pressures as calculated from Eq. (22) by using this value of A are shown in Fig. 2. At pressures of the order of 1 atmosphere the temperature coefficient is negligible—6×10^{-6} per degree at 1 atmosphere. At high pressures, however, the coefficient approaches the value 1/293, i.e., the ionization becomes proportional to the absolute temperature.

Experimental Tests

We have already shown in the solid curve of Fig. 1 that it is possible to present this recombination theory of the variation of ionization with pressure such a form that it agrees acceptably with the experiments. This is however no distinctive test of the recombination theory, since Broxon has developed a formula on the hypothesis of the absorption of beta rays which is likewise in good agreement with his experiments.

An experiment to measure the temperature coefficient of the ionization affords however a crucial test of the two theories; for the present theory gives a definite prediction of the magnitude of this coefficient, whereas on the beta ray absorption theory there is no reason to suppose that the ionization should depend upon the temperature.

In looking for a possible temperature coefficient, we used a spherical steel ionization chamber of 10 cm internal diameter, which had been built for another purpose. The ionization, due to the gamma rays from a milligram of radium at about 1 meter distance and filtered through a centimeter of lead, was measured by a Lindemann electrometer, operating at about 50 scale divisions per volt. The chamber was immersed in a water bath, which could be cooled with ice or heated by a flame. The measurements consisted merely in recording the temperature and timing the motion of the electrometer needle over 10 divisions. It was necessary to allow ample time, at least 10 minutes, for the temperature and the large ions in the chamber to come to equilibrium.

Two series of readings on air at 100 atmospheres pressure, taken between 0° and 30°C and between 0° and 37°C, showed greater ionization at the higher temperature by 7.8 percent and 8.3 percent respectively. The corresponding values of $\beta=\delta i/\delta T$ are 0.0026 and 0.0022 per degree. For nitrogen at 100 atmospheres between 0° and 31°, which theoretically should have nearly the same temperature coefficient, the increase was 9.5 percent, whence $\beta=0.0031$. The average of these values is $\beta=0.0026\pm0.0002$, which is in good accord with the value 0.0029 predicted for air at this pressure by Eq. (22).

Only one set of temperature readings was made at a lower pressure. This was for nitrogen at 20 atmospheres. The observed effect was 0.0000 ± 0.0003, whereas the predicted value of β for nitrogen at this pressure is 0.0004.

Thus the ionization in a pressure chamber is found to increase with the temperature, and this increase is at approximately the rate predicted by the recombination theory.

Relative Ionization by Gamma Rays and Cosmic Rays

If the ionization is due to beta rays ejected from the walls, which are absorbed by the gas in the ionization chamber, the variation of the ionization with pressure should differ with different sources of radiation. For the speed of both photoelectrons and recoil electrons is a function of the frequency of the radiation. If the cosmic rays are photons of very high energy, the range of the excited beta rays should be much greater than of those excited by gamma rays, and the ionization by gamma rays should reach its maximum value at lower pressure. On the recombination theory, however, since the speed at which a beta particle ejects electrons from the molecules which it traverses varies very little with the speed of the beta ray, there should be little if any difference between the pressure-ionization curves obtained with cosmic rays and gamma rays.

To test this point, a cylindrical ionization chamber of 14.8 cm internal diameter and 46 cm length, having steel walls 1.2 cm thick, was used. With this chamber measurements were made of the ratio of the ionization due to cosmic rays to that due to cosmic rays plus gamma rays from a radium standard, using pressures up to 50 atmospheres. The average results of the readings taken on Mt. Evans and at Denver are given in Table I. A different

TABLE I. i_c/i_{c+r} *as function of pressure.*

Pressure, atmospheres	10	20	30	40	50
i_c/i_{c+r} elev. 12,700 ft.	0.230	0.230	0.231	0.228	0.231
i_c/i_{c+r} elev. 5,300 ft.	0.490	0.492	0.490	0.492	0.491

radium standard was used at the two locations. The variations in these readings taken for different pressures are not larger than are to be expected from the probable error.

The beta rays ejected from the steel walls by the action of the gamma rays should be almost completely absorbed by 10 cm of air at some 30 atmospheres, so if this absorption were the cause of the limited ionization current, the differences between i_c/i_{c+r} over the range of pressures here used should have been very marked. The results are however in complete accord with the recombination theory.

ANOMALOUS EFFECTS WITH NITROGEN

In comparing the ionization due to cosmic rays in various gases under pressure, Fruth[12] noted that the ionization in nitrogen remained more nearly proportional to the pressure than that in oxygen or air. Thus, though at atmospheric pressure the ionization in nitrogen is less than in oxygen, at high pressures it becomes considerably greater. This result has been qualitatively confirmed by Broxon, though the differences between nitrogen and air which he observed were not as great as those found by Fruth. Our measurements with gamma rays confirm those of Broxon with cosmic rays. We found the ionization in commercial nitrogen at 100 atmospheres to be 25 percent greater than for air, whereas Broxon found 28 percent.

In terms of the limited range of the beta rays from the walls, this phenomenon seems to have no explanation.[13] On the recombination theory, the lower

[12] H. F. Fruth, reference 2.

[13] Cf., however, J. W. Broxon, Phys. Rev. **38**, 1704 (1931).

probability of recombination in nitrogen implied by this phenomenon indicates that the electrons ejected by the fast beta rays have a greater range in nitrogen than in air. This is what might well be expected from Loeb's observation[14] that an electron will make 10^3 times as many molecular collisions in pure nitrogen as in air before attaching itself to form a negative molecular ion. The fact that slight impurities greatly reduce this ratio may perhaps account for the different results with nitrogen obtained by Fruth and Broxon.

Conclusion

It would seem that the existence of a temperature coefficient of ionization of the predicted magnitude is definite evidence that the recombination-diffusion hypothesis is fundamentally sound.

We cannot place any more confidence in the formula (Eq. (14)) for the ionization as a function of pressure than is justified by its agreement with experiment, since the function (12) describing the ranges of the electron ions is entirely empirical. Yet the predicted variation with temperature as given by Eq. (19) is a necessary consequence of the hypothesis, and the formula (22) for the temperature coefficient of ionization should be considerably more precise than are our present experiments.

It is not improbable that this variation of ionization with temperature may have caused apparent diurnal variations in measurements of the intensity of cosmic rays, especially when the ionization chamber has been placed out of doors.

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[14] Cf. K. T. Compton and I. Langmuir, Rev. Mod. Phys. **2,** 193 (1930).