THE INFLUENCE OF PRESSURE AND COLLECTING FIELD ON GAMMA RAY IONIZATION IN GASES

Thesis by

Dwight Olcott North

In Partial Fulfillment of the Requirements

for the Degree of Doctor of Philosophy

California Institute of Technology

Pasadena, California

1933

ABSTRACT

The first part of this paper presents and discusses the conflicting theories which have been proposed to account for the observed dependence on pressure and collecting field of the ionization current in high pressure chamber electroscopes irradiated with γ -rays. The three types of recombination of ions in gases - preferential, initial, and volume - are considered in detail. The conclusion is drawn that, when low intensities are used, preferential recombination assumes a dominant role. The second part of the paper is devoted to an extension of the Compton, Bennett, and Stearns theory to include dependence on collecting field. Satisfactory agreement with Bowen's observations on air is obtained. The approximate, and partially empirical nature of the expressions developed prohibits a decisive test of the theory. The agreement found <u>does demonstrate its fundamental correctness and superiority</u>.

The weak ionization produced in the usual types of gamma- and cosmic ray ionization chambers has lead to usage of increasingly higher pressures therein in an attempt to obtain stronger and thus more accurately measured currents. But, as Harper[1)] and many others have pointed out, although it was expected that the ionization should increase in proportion to the pressure, and that consequently the ionization current should behave in like manner, actually it was found that with increasing pressure the number of collected ions falls off rapidly from this linear relationship. In fact, with pressures of around 100 atmospheres, despite the use of correspondingly higher collecting fields, it was found impossible to bring the ion current back to the expected magnitude. Sievert[2)] remarks that he has found no indications of saturation at 160 atmospheres with fields as high as 7000 volts/cm.

At first it was supposed that this failure to obtain "full saturation" could be traced to a flaw in the premise that the actual ionization held a strict linear relationship to the pressure, that as the pressure increased the fast β-particles, from the walls, which produced the measured ionization in the chamber eventually ceased their traversal of the chamber from wall to wall and found their terminus within the chamber. Pushing the pressure beyond this point could then be expected to provide little increase in the ionization. Miss Downey[3)] proposed such a theory, and Broxon[4)] deduced from the hypothesis an exponential current-pressure relationship which agrees well with his observations. But it was unable to withstand such criticisms as the following, directed at it by Compton, Bennett, and Stearns[5]. "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 as when cosmic rays are used, 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 relationship is nearly independent of the diameter of the chamber, contrary to expectation. 3) In pure nitrogen the ionization is more nearly proportional to the pressure than in air." That the hypothesis is also theoretically untenable was indicated by Bowen[6] who showed that, inasmuch as the mass absorption coefficients for light substances are very little different from those for the metals from which ionization chambers are made, any β -rays arising in the walls that are completely absorbed by the gas in the chamber are replaced by an equal number of β -rays ejected from the gas. Consequently one should expect the initial ionization in the chamber to bear a direct proportion to the pressure. (A simple mathematical formulation and proof of this idea will be found elsewhere[7].) The Downey-Broxon theory proving inadequate, one must look to a recombination of a portion of the initially generated ion cloud for an explanation of the phenomenon.

There are three more or less distinct types of recombination recognized. When an electron is torn away from its parent it may have insufficient velocity, or it may suffer a sufficient number of energy absorbing collisions in the immediate vicinity that it is never able to escape (despite the presence of a high collecting field) the electrostatic attraction of its parent positive and returns to it. Employing the clarifying terminology of Harper we shall call this type preferential recombination. Again, whether the ionization be produced by α - or β - particles, the initial distribution of the ions is bound to be far from homogeneous, the pairs being distributed along the path of the ray at distances which are small compared to the separation of pairs generated by two distinct rays. This is true to a certain extent in the case of β -ray tracks; one can see in the original C. T. R. Wilson[8)] cloud chamber photographs distinct aggregates of a dozen or so pairs with comparatively large distances between the clumps. It is more especially true of α-ray tracks, for in this case the track is in nearly every instance rectilinear, and with a specific ionization of around 2×10^4, even at one atmosphere there is obviously a marked inhomogeneity in the initial distribution. Thus it may happen that, before sufficient time has elapsed for diffusion to render the distribution uniform, an ion may find itself close to several ions in its own aggregate, and may be captured by one of them under the influence of the combined electrostatic fields of the whole group. We shall term this initial recombination. (It should be pointed out here that the probability of such a recapture occurring, although enormously influenced by the <u>specific</u> distribution of the ions in the aggregate in each instance, should on the average be less than would be the case were the negative left along with a single positive at the same separation. The presence of several ions in a group will generally tend to annul the powerful electrostatic fields which are of so much importance in this type of recombination, and which would be displayed to their best advantage had we only a single pair to reckon with.) Finally, after diffusion has served to produce an effectively random distribution of the positives and negatives, chance encounters may bring two ions of opposite sign close enough that a recapture will ensue. This, the only type of the three which has so far lent itself to experimental investigation (with results all too frequently pitifully at variance) is the well-known <u>volume recombination</u>. To a good approximation this process follows the mass-action law, $\frac{dn}{dt} = -\alpha n_{+}n_{-}$, where n is the density of ions of either sign and α is the coefficient of volume recombination.

Now it is of the utmost importance to be able to say to what extent each of these three possible processes plays a rôle in the measurement of the ionization produced in a chamber by penetrating radiation. Let us consider them in turn in an endeavor to prescribe certain bounds to the magnitude of each effect, certain criteria by which we shall, in a rough way, be able to judge their relative importance in any particular instance.

VOLUME RECOMBINATION

Suppose that the collecting device is a parallel plate condenser and that the final steady state of ionization and collected current has been reached. For convenience we shall also assume that the ion density is constant throughout the volume. This is not strictly true inasmuch as the difference of ionic mobilities for the two signs of ion means the building up of a space charge or concentration gradient. However in the case of air this difference is slight. We shall employ the following symbols:

E_o = potential difference of the plates

l = separation of the plates

u = mean ionic mobility $\frac{u_+ + u_-}{2}$

α = coefficient of volume recombination

n = # ions of one sign per c.c.

N' = " " " " " " " per atmos. which escape initial and preferential recombination

N = " " " " sign per c.c. per atmos. actually collected

N_o' = $N'p$ = # ions of one sign per c.c. per sec. which escape initial and preferential recombination.

N_o = Np = # ions of one sign per c.c. per sec. actually collected.

Then the expression for conservation of particles is:

$$o = N_o' - \alpha n^2 - N_o$$

One has also: $$n = N_o \frac{l^2}{2uE_o}$$

Combining these to eliminate n:

$$o = N_o' - \alpha N_o^2 \left(\frac{l^2}{2uE_o}\right)^2 - N_o$$

Now if we write $u = \frac{u_o}{p}$ where u_o = mobility at one atmosphere;
and define $\gamma = \alpha p\left(\frac{l^2 p}{u_o E_o}\right)^2$, we have

$$o = N_o' - \frac{\gamma}{4p} N_o^2 - N_o$$

Or

$$o = N' - \frac{\gamma}{4} N^2 - N$$

This yields finally:

$$N = \frac{2}{\gamma}\left[\sqrt{1 + \gamma N'} - 1\right]$$

as a good approximation to the number of collected ions under the above circumstances. However, since we are looking for a limit below which we can reject the possibility of this type of recombination, we can assume that in the expression just obtained N is very nearly equal to N'. In this event the formula reduces to:

$$N = N'\left[1 - \frac{N'\gamma}{4}\right]$$

Hence if we agree that volume recombination effects are negligible when they influence the collected current by less than one percent, our criterion for rejection of this phenomenon from consideration must be the following --- we can expect volume recombination to influence the collection of ions in the chamber described to an extent of more than one percent only when:

$$\frac{N'\gamma}{4} > \frac{1}{100}$$

It is well established that for air $u_0 = 1.6 \frac{\text{cm}}{\text{sec}}$ per $\frac{\text{volt}}{\text{cm}} = \frac{1.4_+ + 1.8_-}{2}$. And at one atmosphere[9] $\alpha = 1.7 \times 10^{-6}$. The behavior of this coefficient at pressures of more than one atmosphere has long been a subject of much controversy. More will be said of this later. It suffices at the present to note only that if there is any marked deviation from the one atmosphere value it is probably a decrease. This serves only to make our criterion more effective. We have, then, finally:

$$N\left(\frac{1}{E_0}\right)^2 l^2 p^3 > \frac{u_0^2}{25\,\alpha} \sim 5 \times 10^4$$

INITIAL RECOMBINATION

The essential characteristic of this phenomenon, namely that it depends wholly upon the specific peculiarities of an inhomogenoeus distribution of the ions, makes our quest for a rigorous criterion this time a much more difficult one. One knows for a certainty that the ionization of α-rays is rectilinear, the ion pairs being formed in a dense grouping along the path of the particle. Upon this fact as a foundation, Jaffé[10] in 1913 constructed a theory to explain the voltage-pressure curves obtained by Moulin for α-ionization in air and CO_2 at one atmosphere pressure. The recombination was obtained by applying the ordinary volume recombination to the column as it expanded under diffusion and the influence of the collecting field. From the start it seems hardly justifiable to apply such a coefficient - founded upon an assumption of a completely random and therefore homogeneous distribution of the ions - to the case in hand where it is a basic hypothesis of the theory that the ion pairs are laid out regularly along a straight line. However there must be an element of correctness here, for the results can be seen to give a fairly good agreement with Moulin's data, particularly in that they explain the differences in the collected current when the field is respectively parallel with and at right angles to the columns. Furthermore the constants Jaffé inserts into his equations in order to produce this agreement are of a reasonable magnitude. But, it seems to me, one has to be very careful in attempting to extend this same analysis to the case of ionization by γ - or cosmic radiation. First of all, since the majority of the ionization is here produced by fast β -particles, and a few of their secondaries, and since at pressures of one atmosphere and above (in which range most of the work has been done) their trajectories are wholly irregular and broken lines, the fundamental hypothesis of this theory bears no longer any semblance to actuality. Secondly, one must appreciate that, whereas the specific ionization N_0 of α -rays is of the order 10^4, this quantity in the case of all penetrating radiations is around 30 - 50 ion pairs/cm/atmosphere. Jaffé, in testing his theory in such instances, remarked upon both of these differences. He was able to explain away the first by adapting his theory to fit any arbitrary angle between field and column, but he found himself forced to use values of N_0 around 800 in order to get the desired agreement. He attempted to justify this high figure by the photographs of C. T. R. Wilson, who, he maintained, found counts ranging from 150 to 2160. But one must realize that these were total counts, including all of the branch tracks, and that in no sense could it be said that 2160 ion pairs lay along a cm. of rectilinear path.

After the recent desire for greater efficiency in cosmic ray electroscopes had lead to the use of higher pressures and the publishing of the Downey-Broxon theory had reawakened the whole question which had lain dormant since, and supposedly satisfactorily answered by the work of Jaffé, Gross[11)] exhumed this theory and applied it practically in toto to ionization by γ - and cosmic rays. However, in applying the formula to the excellent γ -ray work of Erikson[12)] performed with a cylindrical condenser in 1908, he seems to have completely overlooked the two pitfalls referred to in the previous paragraph. His formula is of the form:

$$J = C_1 p \frac{1}{1 + C_2 p \log \frac{C_3 p}{E}}$$

(I have left off a correction factor he employed to care for the spatial recombination.) J is the current, p the pressure, E the field strength, and C_1, C_2, C_3 are constants. It is easily seen that C_2 and C_3 are the only ones which are of significance in determining the behavior of the curves, particularly C_2 since the other is the argument of a logarithm; hence large variations of C_3 will change the character of the formula but little. Now according to the Jaffé theory C_2 is expected to have the value

$\frac{\alpha}{4} \frac{N_0}{\pi D}$, where α is the volume coefficient at one atmosphere, N_0 is the specific ionization, and D is the coefficient of diffusion for ions at one atmosphere. As we have seen, N_0 should be around 50, whereas $D \sim 4 \times 10^{-2}$. Using the value for $C_2 = 1.75 \times 10^{-2}$ which Gross assumes, one finds that he must then have a value $\alpha \sim 1.7 \times 10^{-4}$ which is exactly 100 times the accepted value. Later Gross adapted the same formula to Broxon's curves for air and nitrogen, using a value of C_2 which would require α to be again some 30 times too large, provided one is going to take N_0 literally. In view of what has been said, of course this can not be done. But if one is required to replace the quantities which enter into the original expression by others whose significance is vague and whose magnitudes are wholly arbitrary, it seems to me that the theory is no longer adequate despite the good agreement one might obtain by use of the formulized conclusions. Erickson used very high intensities; in fact, he inserted his radium source into the very center of his ionization chamber, cutting off the α-radiation by means of a metallic shield. The current-pressure curves he thus obtained show, for all voltages, a maximum at a pressure which steadily increases with the voltage. The Gross calculations show the same characteristic, but it is the writer's belief that this cannot be maintained as a justification of the theory; for the criterion previously developed for the appearance of a volume recombination effect has also <u>the same properties</u>, provided the ionization is strong enough. Unfortunately, (like so many other workers in this field) Erikson omitted publication of the actual intensities obtained, or field strengths employed. It is certain only that the currents were enormous compared with those obtained by workers using cosmic rays only, or γ -rays from a source at a considerable distance from the chamber.

The writer is tempted here to remark upon another point in the theory under discussion. Both Jaffé and Gross have insisted upon employing the relationship, $\alpha \propto 1/p$. This comes from the Langevin expression, α_L . The Thomson expression, α_T , on the other hand, yields such a relationship only at low pressures, while at high pressures (above five atmospheres) it becomes independent of the pressure. The non-existence of accurate experimental determinations of this coefficient at pressures of more than three atmospheres hampers an attempt to make a rejection of the one and acceptance of the other. But Loeb, in his treatise "The Kinetic Theory of Gases", p. 488, gives a comparison of α_L and α_T as regards their dependence both upon pressure and temperature at pressures of one atmosphere and below. It is too lengthy a discussion to include here; I wish only to remark that α_T was found to fit the facts better upon every count. One can still believe whatever he likes about the behavior of α at high pressures, but one is bound to concede that the Thomson theory is more likely to be right*.

Altogether it is difficult to see the applicability of the Jaffé theory to ionization by γ - or cosmic radiation, particularly when the ionization is very weak, as it nearly always is. Bowen[6] exhibits in his paper a typical set of γ - ray data, taken with a parallel plate condenser. If we insert into the previously developed formula, viz: $n = \frac{N_0 l^2}{2uE_0}$, the most unfavorable values from his observations, so as to obtain the greatest possible ion density ($N_0 = 1.2 \times 10^4$, at a $p = 100$ atmos.; $u = \frac{1.6}{100}$; $l = 1$ cm; $E_0 = 1.55$ volts), we find a density of ions of one sign equal to 2.4×10^5 ions/cc. This makes the smallest possible average separation of the ions approximately 0.013 cm. With a broad γ -ray beam directed so as to include the whole chamber and provide ionizing β -particles emerging from the walls at all angles, it is difficult for the author to imagine how, with a density as small as this, enough of these ion pairs could originate in an array at all comparable to the columnar distribution that is necessary if one is to apply to it the Jaffé theory.

Now it is well enough, perhaps, to discard this particular theory as unsuitable in the present circumstances, but at the same time it <u>must be admitted</u> that initial recombination does

---

* On the other hand, Harper, Proc. Camb. Phil. Soc., 28, 219, 1932 has produced a new derivation of this coefficient which approximates that of Langevin.

most certainly occur. However enough has been said, I think, to make it clear that when such a recombination does obtain, it will be limited to groups of two or three ion pairs which by chance are generated in close proximity to one another. For this reason the effect will be weak, and for the same reason one is unable to say exactly how weak. We have arrived at no definite criterion for discarding the effect, but it seems reasonable to suppose that in treating observations such as Bowen's this type of reunion will play no greater role than does volume recombination.

PREFERENTIAL RECOMBINATION

In 1906, Bragg and Kleeman[12)] first suggested this mode of recombination to explain the fact that, in the case of α -rays, saturation currents were much more difficult to obtain than was to be expected from the well-known Thomson parabola formula, and also attempted to account, by this hypothesis, for the fact that as weaker ionizing agents were used the saturation currents obtained became more and more nearly independent of the intensity and of the form of the ionization chamber. Moulin undertook experimental investigation of this hypothesis and succeeded in convincing the workers of the time that the Bragg concept was not wholly adequate in the case of α -radiation, but that the effects were more nearly accounted for by an hypothesis of Langevin's based upon the interaction of, not merely pairs of ions, but of all the ion pairs in a "column". Consequently the Bragg and Kleeman idea was forgotten, the Langevin theory carried forward to receive its final treatment at the hands of Jaffé, as we have seen.

It lay dormant until the publishing of the Downey-Broxon theory invoked the criticism given above. Rejection of this theory demanded something to take its place. Simultaneously there appeared letters by Millikan and Bowen[13)] and by Compton, Bennett, and Stearns[14)] suggesting anew 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." By this hypothesis one could already account qualitatively for those phenomena for which the Downey-Broxon theory could offer no support, namely the independence of the form of the pressure-ionization curves on "hardness" of the ionizing beta-particles, and on the diameter of the chamber; and the wide variation in intensity and form of the curves when different gases were used. Shortly thereafter Compton, Bennett, and Stearns[5)] brought out a theory based upon the assumption that all of the effect in the case of γ - or cosmic radiation was due to preferential recombination. Their results, when applied to a curve of Broxon's gave very good agreement for the pressure-ionization relationship. They developed a critical radius, r_0 , around the parent ion equal to $\frac{e^2}{3kT}$, where the constants have the usual significance. Any electron, ejected from its parent and brought to equilibrium with the molecules of the gas by energy absorbing collisions before it had wandered outside this critical pale would be expected to undergo a preferential recombination.

In order to make estimations of the fraction of the total number of ions ejected which would suffer this realliance, it was necessary to know the probability that an ion would come to equilibrium at a distance r from its parent. To date such a function has not been determined. It will depend upon several factors; (1) the initial velocity of ejection, (2) the average fractional energy absorption upon collision with a molecule, and (3) the manner in which the direction of path is changed upon collision. There is every reason to suppose that quantum conditions will control the absorption of energy. In some cases, where the molecules are electropositive it may happen that, with a head-on collision, the electron is captured, yielding up all of its kinetic energy at once to fall into an allowed level of the molecule, the energy being radiated as a quantum of light. In the majority of cases, where the molecule is diatomic, one could expect many collisions to occur, small quants of energy being transferred to the vibrational or rotational energies of the diatomic complex, until finally the electron, coming nearly into equilibrium with the gas, would attach itself to a neutral molecule to form a negative ion. The observed superior efficiency of argon over nitrogen, and of the latter over air, is immediately accounted for. Nitrogen has no affinity for electrons, while the oxygen molecule is ready to attach one to itself; that is to say, there is no allowed quantum state of the normal nitrogen molecule into which the electron can fall, converting its energy into a radiated quant, while such states do exist in the case of oxygen. The only way in which an electron which finds itself in an environment of pure nitrogen can lose its velocity is by the second method, namely, transferring its energy a fraction of a volt at a time to the vibrational or rotational states. But the argon molecule, being monatomic, foregoes even this possibility of energy absorption, so that (discarding as negligibly small the fraction $2\frac{m}{M}$ of energy always lost in elastic impacts) here the electron can be expected to wander on indefinitely in the absence of a collecting field, attaching perhaps to a foreign molecule possessing electron affinity, but never to an argon molecule. (Harper, loc. cit., has criticised the preferential recombination theory on the grounds that it is even qualitatively unable to clear up the difference between nitrogen and argon, but it seems to me that the foregoing argument is sufficient.)

Harper has also adduced the cloud chamber photographs of C. T. R. Wilson as proof that preferential recombination is seriously complicated by initial recombination. The pictures show, he asserts, that on the average the separation of ion pairs is of the same order of magnitude as the separation of the components of a pair. However it seems to me that this observation carries little significance, for in order to produce pictures in which the pairs could be resolved and the intensity sufficiently low for an accurate count to be made low pressures had to be used. Now, at low pressures preferential recombination is admittedly slight. But in any case a picture which can resolve the components of a pair must have been taken at a considerable time interval after the electron had accepted or rejected its chance at recombination. There is also the serious doubt that ion pair components caught by the camera at a separation (around 10^{-6} cm.) favorable to this type of reunion would be sufficiently separated to present to the water vapour the requisite positive and negative charges necessary to condensation.

The critical radius of capture, r_0, having a value at room temperature of 1.88×10^{-6} cm., which means that the electrostatic field at the distance of 50/50 probability of return is approximately 4×10^4 volts/cm., Compton, Bennett, and Stearns concluded that only extremely high collecting fields would materially influence this type of recombination. But it is a well-known fact that changes in the strength of the collecting potential materially alter the collected current, particularly when the voltage is small. (It was precisely for this reason that investigators have always sought a "saturation current" so that their data would be free from the influence of this dependence.) Even at large voltages, Bowen (loc. cit.) has shown, if the field be uniform and the pressure high, the current-voltage curves possess a definite upward slope. He points out that the conclusion of most investigators that they had obtained "saturation" when a variation of plate potential of two or three times the original revealed no appreciable increase in current was erroneous, because of the fact that they were using inverse first power collecting fields, so that most of the potential drop was near the collecting rod and the greater part of the chamber was subject to a field that was only a small fraction of the average potential drop. His experiments were performed with a chamber consisting of eight plates alternating with seven collecting plates, each of them protected by a guard ring, so that a highly uniform field was obtained throughout the apparatus. The observed persistent dependence upon field strength led him to suggest that a modification of the theory of Compton, Bennett, and Stearns was called for, that "given two ions in a gas, there is always a certain probability that one will diffuse close enough to the other, regardless of their original distance apart, so that recombination takes place. Of course this probability falls off rapidly as the distance increases. Furthermore this probability, particularly for an ion at a considerable distance from the parent ion, is materially changed as the strength of an external collecting field is varied". It is this modification which I propose to consider in the following pages.

It will be noted that as yet no criterion has been reached for the acceptance or rejection of this mode of recombination. However, since the other two modes have been shown to be weak, and since this mode is the only one which presents a qualitative explanation of all of the characteristics peculiar to ionization in high pressure chambers by means of γ - or cosmic radiation, it will be assumed in the succeeding discussion that, except in instances where the criterion for volume recombination shows that effect to be of importance, neither volume nor initial recombination plays a significant role.

INVESTIGATION

It has already been pointed out that there are two distinct parts to any theory of perferential recombination; a) the determination of that distribution function which gives the probability that an electron ejected from a molecule finds itself brought into thermal equilibrium with its surroundings at a specified distance r from its parent positive, b) the determination of the probability that this electron will subsequently escape to the plates of the chamber under the combined influence of the field of its parent, the applied collecting field, and Brownian motion. We shall confine our attention to the second of these problems for the moment.

§ 1. The Field.

Figure 1 shows the distribution of lines of force for a charge $-e$ at the origin and a uniform field E_0 in the Y-direction. The potential is

$$V = -E_0 \, r \sin\theta - \frac{e}{r} ,$$

from which the equation of any line of force is readily seen to be

$$C \text{ (constant)} = \frac{E_0}{2e} r^2 \cos^2\theta + \sin\theta$$

It will be useful here to introduce the dimensionless quantity,

$$\xi = r\sqrt{\frac{E_0}{e}}$$

So that $$C = \frac{1}{2}\xi^2 \cos^2\theta + \sin\theta$$

The various values of C, $-1 \leqslant C < \infty$, give the various field lines in Fig. 1, which retains the same properties for all values of E_0. When $C = 1$ the equation gives the envelope (T) of lines of force which end on the charge. When C lies in the range, $-1 \leqslant C \leqslant 1$, $C = \text{Lim} \sin\theta$. The coordinates of the points P, Q on T are found to be $P(\xi = 1, \theta = \pi/2)$, $Q(\xi = \sqrt{2}, \theta = 0)$; and as an exploring point moves off toward infinity along T its coordinates approach the limiting values $(\xi \cos\theta = 2, \theta = -\pi/2)$. This gives an idea of the proportions of the envelope.

§ 2. An Initial Attempt at Solving the Problem.

What seems to the writer as an ideal method of searching for a solution to the problem in hand is the following, which is included in this thesis not because it has proved forceful in the hands of the author, but rather because its exposition may prove useful to a later, more ingenious investigator. One desires to know how many charged particles (e), let loose at various points of the field of Fig. 1 will go to the origin, and how many will go to the plates. More explicitly, it is desired to know the probability that a charge (e) set free at a point (r,θ) will move off to the collecting plates. The situation is actually a little less demanding, for the probability function referred to in (a) will naturally possess radial symmetry; consequently we can generate a flock of non-interacting identical particles uniformly and constantly from the surface of a sphere (r). Their density (n) will be determined after a steady state has been reached by the appropriate solution of

$$0 = D\nabla^2 n + u\nabla\cdot(n\nabla V)$$

where u is the mobility of a particle, D the coefficient of diffusion. From this solution could be calculated the fractional flow of particles outward from the generating surface. This, a function of r, would be the desired probability.

The boundary conditions are easily obtained, but the V of § 1 robs the equation of its simplicity. No success was attained by this author in attempting to separate its variables. The method of successive approximations was tried but was found ineffective because of a bad singularity at the point P, Fig. 1, which caused difficulty when one attempted to match the solutions for the inner and outer regions. Perhaps someone will be able to surmount these obstacles and gain a rigorous solution to the problem by this means. The present investigator has achieved no success with it, perhaps due to the scarcity of both time and the requisite patience.

§ 3. Motion of an Ion in the Field.

The alternative to the method of § 2 is an investigation of individual behavior. If we place a charge e at a specific point (r,θ) in the field of Fig. 1, what can we say about its motion? If it starts from rest, its initial motion will be in the direction of the field, but as soon as it gathers momentum, and if the field is sufficiently divergent, its trajectory will soon deviate from the field lines because of the inertia of the associated mass. Brownian motion will also play a part, but this paragraph will be devoted to an investigation of the deviation arising from inertia alone, the Brownian effect appearing only as a viscous term in the equations. The object will be simply to discover whether or not one can expect large inertia-deviations for charges which move through fields whose curvature is comparable to that of the section PQ of the envelope (T).

For our purpose it will be sufficient to suppose the charge (-e) fixed at the origin. The charge (+e) will be allowed to move; it is assumed to have the mass (m) of a molecule of the gas. In the actual case both ions move; this will later be considered. Here we seek only an order of magnitude. The equations of motion are two-dimensional in this case,

$$m\ddot{x} = -e\,\frac{\partial V}{\partial x} - \eta\dot{x}$$
$$m\dot{y} = -e\,\frac{\partial V}{\partial y} - \eta\dot{y}$$

where $\eta \equiv \frac{e}{u}$ and u is the mobility of the ion. If $Z \equiv x + iy$,

$$m\ddot{Z} = -\frac{e^2 Z}{r^3} + iE_o e - \eta\dot{Z}$$

For a first approximation we take $\ddot{Z} = 0$; this is equivalent to postulating an infinite viscosity, so that the trajectory is given by

C = constant

that is, the ion moves along a line of force. This gives,

$$\dot{Z} = i\,\frac{E_o e}{\eta} - \frac{e^2 Z}{\eta r^3}$$

For a second approximation we substitute this value of $\dot{Z}$ together with the derived $\ddot{Z}$ into the original equation. After some transformations into polar coordinates we find the two equations:

$$\dot{r}\left[\frac{2me^2}{\eta^2 r^3} + 1\right] = \frac{E_o e \sin\theta}{\eta} - \frac{e^2}{\eta r^2}$$
$$r\,\dot{\theta}\left[-\frac{me^2}{\eta^2 r^3} + 1\right] = \frac{E_o e \cos\theta}{\eta}$$

Dividing and rearranging,

$$d\left[\frac{E_o}{2e} r^2\cos^2\theta + \sin\theta\right] + \frac{me}{\eta^2}\,\frac{\cos\theta}{r^2}\left[2E_o\cos\theta\,dr + \left(E_o\sin\theta - \frac{e}{r^2}\right)r\,d\theta\right] = 0$$

or $$\left[1 - \frac{me^2}{\eta^2 r^3}\right]\cdot d\left[\frac{E_o}{2e} r^2\cos^2\theta + \sin\theta\right] + \frac{3meE_o}{\eta^2}\,\frac{\cos^2\theta\,dr}{r^2} = 0$$

The factor $\left[1 - \frac{me^2}{\eta^2 r^3}\right]$ will be neglected; it can be shown that for fields $\sim 1000 \frac{\text{volts}}{\text{cm.}}$, $\frac{me^2}{\eta^2 r^3} \ll 1$ on T. To make the second term integrable we require the trajectory to start in the neighborhood of T, so that

$$1 = \frac{E_0}{2e} r^2 \cos^2\theta + \sin\theta$$

or

$$\cos^2\theta \approx \frac{4e}{E_0 r^2}\left(1 - \frac{e}{E_0 r^2}\right)$$

Substitution and integration gives:

$$\left[\frac{E_0}{2e}(r\cos\theta)^2 + \sin\theta - A\right] - \frac{4me^2}{\eta^2 r^3}\left[1 - \frac{3e}{5E_0 r^2}\right] = 0$$

where A is an arbitrary constant of integration. It will be determined by requiring that the trajectory include the point Q. One obtains finally

$$\frac{1}{2}\xi^2\cos^2\theta + \sin\theta - 1 = \frac{4me^2}{\eta^2}\left(\frac{E_0}{e}\right)^{3/2}\left\{\frac{1}{\xi^3} - \frac{3}{5\xi^5} - \frac{7}{20\sqrt{2}}\right\}$$

The right hand side expresses, within the limits of accuracy of our approximations, the deviation of the trajectory of a particle starting at Q from the line of force which runs through Q. The left hand side will be recognized as the equation of the envelope T. Since ξ is O(1) on T, the magnitude of the inertia-deviation is obtained from the factor $\frac{4me^2}{\eta^2}\left(\frac{E_0}{e}\right)^{3/2} \equiv 4mu^2\left(\frac{E_0}{e}\right)^{3/2}$

Now for O_2, $m \doteq 32 \times 1.662 \times 10^{-24}$, $u \sim 1.6 \frac{\text{cm}}{\text{sec}}$ per $\frac{\text{volt}}{\text{cm}}$; if we choose $E = 1000 \frac{\text{volt}}{\text{cm}}$, the maximum field strength used in the Bowen experiments to which this argument is to be applied, we find this factor to be approximately 3×10^{-8}. If pressures higher than one atmosphere are employed, $u \sim \frac{1.6}{p(\text{atmos.})}$, serving to reduce the factor still more. In air, then, one concludes from this analysis that ions which have attained their terminal velocities would possess trajectories almost coinciding with the field-lines for curvatures of the order of the arc PQ of T, were it not for the Brownian motion they undergo in planes normal to the trajectory. The correction term is small enough that the author feels justified in extending the conclusion to trajectories within T possessing larger curvature as shown by Fig. 1, although it is certain that the analysis breaks down in a limited region near P where the field lines undergo very sharp bending.

§ 4. Motion along a Field Line

Now if, on the basis of the preceeding analysis and still disregarding Brownian motion, we premise trajectories coincident with field lines under the law:

$$\frac{d\bar{s}}{dt} = u\bar{E},$$

and if furthermore we suppose the ejected electron to have picked up a neutral molecule so that its mass is the same as that of the positive, they will both move in a symmetrical fashion about their center of gravity along their respective lines of force. The lines of force, now expressed in terms of ξ, θ referred to the center of gravity as an origin, are given by

$$C = 2\,\xi^2 \cos^2\theta + \sin\theta$$

(which is a direct modification of the similar equation of § 1 if you think of the change as amounting only to a diminution of e by a factor 1/4.) The time required for a pair of particles to move into coalition is then

$$t = \frac{1}{u}\int\frac{ds}{E} = \sqrt{\frac{e}{E_0}}\,\frac{4}{uE_0}\int_0^{\xi_0}\frac{\xi^2\,d\xi}{4\xi^3\sin\theta - 1}$$

The desideratum throughout this discussion has been to obtain a critical surface (G) surrounding one particle such that if the other particle is found in thermal equilibrium with the gas within G it will recombine, without G it will go to the plates. A rigorous determination of the equation of G would demand a point to point pursuit of the particle as it moved under the combined influence of Brownian motion and the electric field. Such a procedure, although conceivably a not impossible mathematical task, would yield an equation so complex that its usefulness in conjunction with the distribution function of § 1 a) would be vanishingly small. One must therefore look for an integrable approximation to the actual surface G and try to be content with it. A first step in this approximate method is clearly to equate the above expression for the time required for the particles to move into coalition from points (ξ_0, θ_0) to an appropriate expression for the time allowed by Brownian motion for the particles to separate beyond the limits of G. But here again a glance shows that the equation of the surface would connect θ_0 with ξ_0 by means of an elliptic function, and ones hopes for a manageable expression are once more frustrated.

But one does have a qualitative conception of the kind of surface G should be. It is depicted by the dotted line in Fig. 1. As the collecting field, E_0, is increased more and more of the ions originating within T will be caught, so that for large values of E_0, G must be coincident with T. For lower E_0, G will move inside of T, but will still preserve a somewhat similar shape. The extension of G below the X-axis toward infinity is qualitatively correct. For if an ion originates in this "tail" at a considerable distance from its parent, although there is a chance that its Brownian movement will carry it normal to the field lines and out across the boundary, there is an equal chance for the reverse process to occur. Actually one realizes that the tail must be terminated when its length becomes commensurable with the average separation of ion pairs, that beyond this point a preferential theory must bow and withdraw in deference to the other two types of recombination. But in computation this is a matter of small concern for the fraction of ejected electrons that comes to equilibrium at such distances is very small; most of these ions which escape will do so via the top or "head" of G.

Such a surface as the one depicted above, one having the shape of T, is given by $\xi^2 = \frac{8\ \xi_0^2}{1 + \sin\theta}$, referred to one ion as a center. ξ_0 is the defining parameter which determines its size, $2\xi_0$ being the original separation of pair components on the Y-axis, and ξ_0 itself being determined as follows. The time required for coalition under the influence of electric forces for such a pair of particles symmetrically disposed on the Y-axis is given by the previously developed expression,

$$t = \sqrt{\frac{e}{E_o}}\,\frac{4}{uE_o}\int_0^{\xi_o}\frac{\xi^2 d\xi}{4\xi^2 - 1} \qquad \left(\theta = \pi/2\right)$$

or

$$t = \frac{1}{2}\sqrt{\frac{e}{E_o}}\,\frac{1}{uE_o}\left[\tanh^{-1} 2\xi_o - 2\xi_o\right]$$

During this process Brownian motion will occur in the plane normal to the trajectory, assumed straight. The appropriate Einstein-Smoluchowski expression* is $\overline{r^2} = 4Dt$. On the average the particles will escape if the separation is greater than OQ'. Consequently the parameter ξ_o is defined by these two relationships:

$$8\,\frac{e}{E_o}\,\xi_o^2 = 4D\left\{\frac{1}{2}\sqrt{\frac{e}{E_o}}\,\frac{1}{uE_o}\left[\tanh^{-1} 2\xi_o - 2\xi_o\right]\right\}$$

Rearranging, making use of the well-established relationship[15] $\frac{u}{D} = \frac{e}{kT}$ where k is Boltzmann's constant, T the absolute temperature; and defining

$$\beta \equiv \frac{kT}{4e\sqrt{eE_o}},$$

we have

$$\xi_o^2 = \beta\left[\tanh^{-1} 2\xi_o - 2\xi_o\right]$$

---

* The exact expression is $\overline{r^2} = 4D\left[t + \frac{mu}{e}\left(\epsilon^{-\frac{e}{mu}t} - 1\right)\right]$. A rapid calculation shows the times considered here are sufficiently large to warrant use of the simplified expression.

For each value of $\beta(E_0, T)$ the solution of this transcendental equation, plotted in Fig. 2, determines ξ_0 and the surface G.

§ 5. Equilibrium Distribution Function.

Earlier in the paper there were enumerated the many obstacles confronting him who would seek to determine the probability of finding an ejected electron brought to equilibrium in a range r, r + dr from its parent. Compton, Bennett, and Stearns(5) have discussed this question. They tried a Maxwell-Boltzmann distribution in a similar theory which essayed to account for dependence of ionization on pressure; but comparison of their findings with data of Broxon's led them to conclude that this distribution did <u>not</u> represent the facts, for it apparently concentrated the electrons too close to the parent. This finding, by the way, defends our hypothesis concerning the mechanism of energy absorption. Were loss of kinetic energy and capture of a neutral molecule a matter of chance alone, a Maxwellian distribution should serve well. But <u>if</u> the process hinges upon <u>quantized</u> exchanges of energy, such distribution could not be expected to obtain. Satisfactory ionization-pressure relationships were secured by these workers with the use of an empirical expression,

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

where F(r)dr is the probability that the ejected electron comes into equilibrium in the range r, r + dr. This formula will be used in the present discussion, its only raison d'être being the good agreement obtained with it by its originators, to whose paper the reader is referred for details, and recently[16] equally good experimental verification of the temperature coefficient derived by these authors from the theory in which this function is embodied.

§ 6. Current-Pressure-Voltage Relationship.

The probability of escape is given by:-

$$P = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \int_{r_0(\xi_0)}^{\infty} F(r)\, dr\, d\theta = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \frac{d\theta}{\sqrt{1 + \frac{r_0^2}{a^2}}}$$

$$= \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \frac{d\theta}{\sqrt{1 + \frac{8e}{E_0 a^2} \frac{\xi_0^2}{1+\sin\theta}}}$$

Carrying out the integration, one has:

$$P = \frac{2}{\pi} \cot^{-1} \left[2\sqrt{\frac{e}{E_0}}\ \frac{\xi_0}{a} \right]$$

The quantity a represents the average range, and varies as $1/p$ where p is the pressure in atmospheres. Writing $a = \frac{1}{Cp}$, where C is a constant, we have finally:

$$P = \frac{2}{\pi} \cot^{-1} \left[2\sqrt{\frac{e}{E_0}}\, \xi_0 C p \right]$$

Then if N = # ions per c.c. per sec. per atmos. which escape preferential recombination,

N_0 = # ions per c.c. per sec. per atmos. initially generated (assumed constant for reasons listed above),

$$N = N_0 \cdot \frac{2}{\pi} \cot^{-1} \left[2\sqrt{\frac{e}{E_0}}\, \xi_0 C p \right]$$

The only two quantities to be obtained from the data, in checking this expression with experiment, are N_o, C. N_o can be found from the current obtained at the lowest pressure and highest potential employed, provided the fields used are sufficient to bring about almost complete saturation and the pressure not so low that the threshold of ionization by collision has been reached.

§ 7. Air.

The formula can be compared with observation only under special, i.e. not ordinarily attained, conditions. The ionization must be so weak that volume recombination plays no significant role; the field must be highly uniform and run through a wide range of values in order that the small changes in the collected ion stream will show up distinctly. Comparison is made in Fig. 3 for Bowen's observations[6] on air irradiated with gamma rays. In these experiments both of the conditions noted above were admirably met. The field strengths are plotted logarithmically. The crosses represent Bowen's observations; the heavy lines, the computed curves. N_o was chosen as 121.1, and $C = 2.08 \times 10^3$ was obtained by fitting to the encircled observation at 25 atmos., 1009 $\frac{\text{volts}}{\text{cm}}$. The inability of the curves to fully match the observations on the high voltage side must be laid to improper choice of equilibrium distribution function, although there is some doubt about the 93 atmos. data. Dr. Bowen has informed the writer that the pressure gauge he was using may have read as much as 5% too high at that pressure. This correction of the data would provide better agreement. It is the slope of the curves that provides a test of the present theory. On the high voltage side the agreement is, in the main, good. But they flatten out altogether too quickly thereafter. The reason is patent. For low voltages G is forced by the present theory to retain the proportions which were shown to be correct only at high voltages. Hence the heavy lines should represent only an upper limit. As the voltage drops to zero, G must manifestly be converted, in an as yet undiscovered manner, from the shape of T to a spherical surface. In order to obtain this lower limit to the variation the writer has assumed G to be spherical for all voltages, with a radius varying according to the law (§ 4) by which ξ_0 varies, and matched to the former calculations at the high-voltage end of each constant pressure curve. These lower limit curves are shown by the dotted lines in Fig. 3. It will be noticed that except for the highest pressure curve, which is obviously in need of shifting vertically although its slope is good, the observed points lie pretty well within these two bounds. The agreement would look much better were it possible to divine the manner in which the transition of the G surface occurs.

Only at the very lowest voltages is there a distinct falling away of the observed current. This may find its explanation in volume recombination; the criterion for this effect shows that in the case of Bowen's measurements this type of current loss should occur, if at all, only at the lowest observed voltage. The magnitude of the effect and its dependence on the pressure are influenced enormously by the way in which the coefficient of volume recombination behaves at high pressures. As was previously observed, practically nothing can yet be said on this subject. On the other hand, from the observer's point of view, no apparent volume effects occurred except at the upper two pressures. Dr. Bowen has, in conversations with the author, repeatedly emphasized the fact that curves for a given intensity when increased by an appropriate factor (approx. 5) coincide almost exactly with the higher intensity observations.

§ 8. Other Gases.

The extension of this theory to gases other than air should require little modification of the premises. Preferential recombination in gases, such as argon and nitrogen, where the electron, once stopped, attaches itself only to foreign neutrals, means that in many cases it will not be a heavy ion returning to its parent, but rather the electron itself. The larger mobility of the electron influences the analysis only to the extent that it shifts the center of gravity. This introduces no significant change in the defining equation for ξ_0 ; and the relationship $\frac{u}{D} = \frac{e}{kT}$ still holds. For this reason the abnormally high mobilities of new-born ions, recently reported by Loeb's school at Berkeley and others, have no bearing on this problem. Mr. Cox(7), at this Institute, has lately extended Bowen's work to other gases - nitrogen, carbon dioxide, argon, and helium. The behavior of nitrogen is so similar to that of air that there would be little gain in making a comparison. An attempt was made, however, to fit the theory to his data for argon. The agreement was very poor, partly because of certain marked inconsistencies in the data itself, partly because of the resulting indeterminateness of the influential constants, N_0 and c, which have to be obtained from the data, and partly, perhaps, because the same empirical distribution function was employed as was used in the air comparison. The mechanism of energy absorption being so widely different in the two gases, air and argon, there is no reason to suppose the same function would serve both equally well. Hopfield[(17)] has recently published new data for argon, but no comparison can be made with it either; for although the intensity is low the field is widely divergent, so that at the maximum collecting voltage the effective field is probably less than 15 volt/cm. Whenever a comparison of this theory in its present form is made for gases other than air, it will be crucial only when the data is taken for pressures up to at least one hundred atmospheres, with uniform collecting fields preferably up to two or three thousand volts/cm., and intensities sufficiently low to insure freedom from volume recombination.

§ 9. Conclusion.

Argument has been presented in the earlier part of the paper to show good reason for expecting the major part of current loss in γ-ray ionization chambers collecting low intensity currents to be due to preferential recombination alone. The latter portion has consisted of an attempt to justify such a premise, by constructing a function which purports to give the dependence of the collected current upon both pressure and field strength. The investigation has throughout been seriously hampered by the lack of mathematical tools adequate to handle statistical problems of this nature, as well as by the complexity of the quantum physics governing exchanges of energy between low velocity electrons and neutral molecules. In its present form the analysis set forth can in no way be construed to proffer a decisive test of the premise; it is the writer's belief that this paper does indicate the preferential recombination theory to be qualitatively tenable, quantitatively plausible.

The author desires to thank Dr. Harry Bateman for his advice in connection with a mathematical portion of the problem; Dr. Ira S. Bowen, who proposed the investigation, for his many suggestions and information, particularly concerning the significant experimental facts; and Dr. Zwicky, whose ready counsel, encouragement, and guidance have made this work a distinct pleasure.

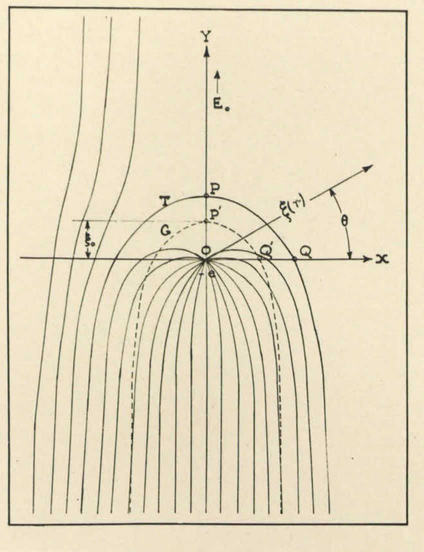

Fig. 1

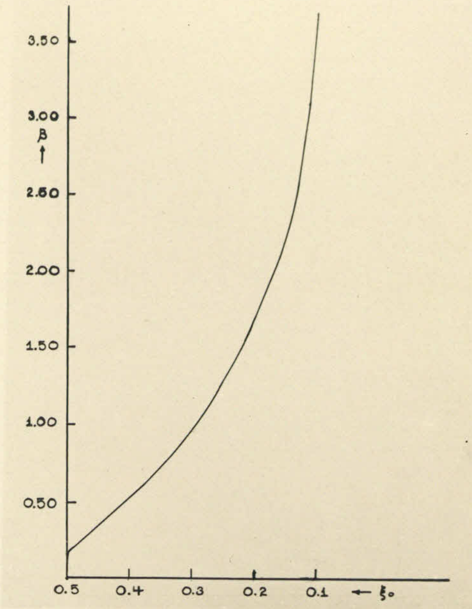

Fig. 2

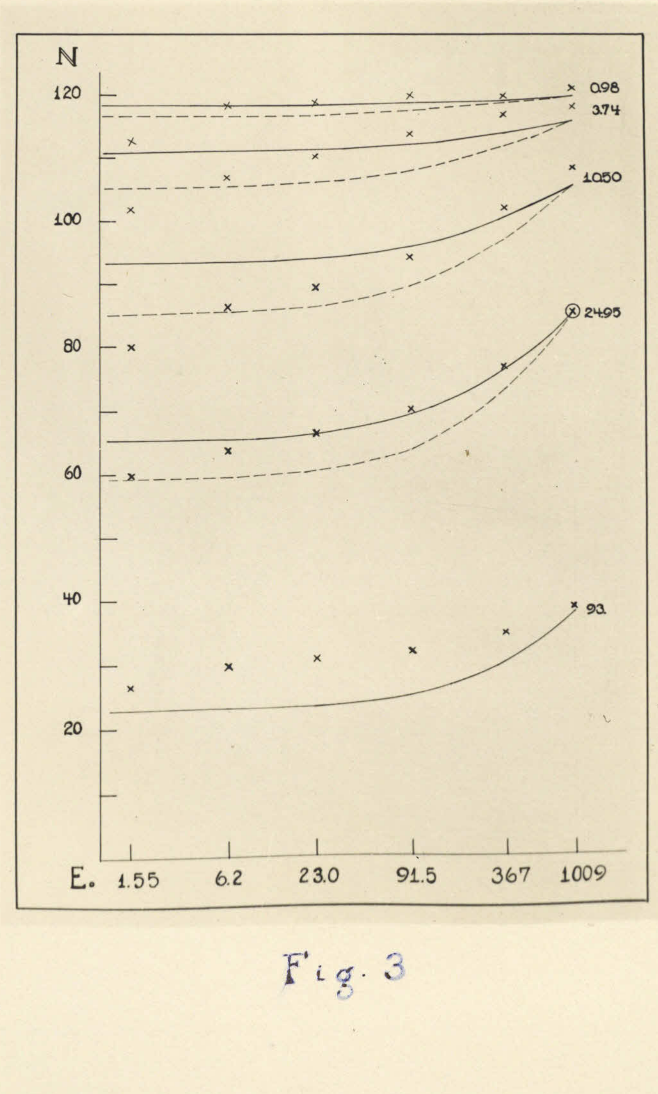

Fig. 3