

GAMMA RAY IONIZATION IN SEVERAL GASES AS A  
FUNCTION OF PRESSURE AND COLLECTING FIELD.

Thesis by  
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## SYNOPSIS

Ionization by low intensity  $\gamma$  radiation in  $N_2$ ,  $CO_2$ , A, and He has been measured with pressures from 0.98 to 93 atm. of  $N_2$ , 61 atm. of  $CO_2$ , 25 atm. of A, and 98 atm. of He and with uniform collecting fields of from 1.57 to 1065 volts per cm., direct comparison being made with the results of Bowen for air. The marked increase in current with increased collecting gradient and independence of the shape of the current-field curves on intensity of radiation up to the intensity used demands rejection of the wall emission theory of Broxon and adaption of some recombination theory. Junction of Kaye and Laby's results to those of Bowen show a gradient of  $4 \times 10^3$  volts per cm. is saturation value for air at 10 atm. pressure. A field of only 100 volts per cm. is saturation value for argon up to 25 atm., and with this field the current from the inert gases examined is a linear function of the pressure up to 25 atm. Less recombination was observed in  $N_2$  than in air, and investigations of the effect of  $O_2$  impurity in  $N_2$  are now in progress.

## INTRODUCTION

In the first edition of his book "Radio-Activity", published in 1904, Rutherford makes the following statement<sup>1)</sup> with respect to the ionization produced in a gas as a function of the collecting field: "There is no evidence of complete saturation, although the current increases very slowly for large increases in voltage. .... Taking into consideration the early part of the curves, the current does not reach a practical maximum as soon as would be expected on the simple ionization theory. ... It is possible that the presence of a strong electric field may assist in the separation of ions which otherwise would not initially escape from the sphere of one another's attraction." Bragg and Kleeman<sup>2)</sup> in 1906 produced additional experimental evidence of this effect in air and heavier gases up to one atmosphere pressure and found the phenomenon even more pronounced in the heavier gases. This work, as well as that mentioned previously by Rutherford, was done with the radioactive source within the ionization chamber, alpha rays therefore producing the major part of the ionization.

The next notable account bearing on this thesis is that of H. A. Erikson<sup>3)</sup> on research in which he used a cylindrical ionization chamber and produced the ionization with gamma rays, probably very intense. With this apparatus he measured the currents up to 400 atmospheres for air and to 64 atmospheres for carbon dioxide, and studied the action of the collecting field for voltages from 6

to 1040 on air with pressures of 20, 40, 140, and 400 atmospheres and from 8.5 to 1040 volts on carbon dioxide at pressures of 6 to 64 atmospheres. He observed that every current-pressure curve at constant voltage had a maximum value which moved in the direction of increasing pressure as well as in the direction of increasing current as the voltage was increased. The author has been unable to find any other reference in which the curves show a maximum value.

The first experiments in which the ionization chamber was designed for a constant field and appreciable pressure were conducted by T. H. Laby and G. W. C. Kaye.<sup>4)</sup> With a parallel plate condenser chamber they measured the ionization in hydrogen, carbon dioxide, and air as a function of the pressure and collecting field for pressures of from one to 17 atmospheres and fields up to 3500 volts. The intensity of the gamma rays used by them is comparable with that used by the author.

Other works of this period deserving mention are those of W. Wilson<sup>5)</sup> on air up to 40 atmospheres and very doubtful "saturation" field strength; and D. C. H. Florance<sup>6)</sup>, who uses very intense gamma rays with a cylindrical chamber in measuring the pressure-voltage variation of current in air up to 80 atmospheres and 2200 volts.

For the next seven years the investigations bearing on the subject of this thesis are absent from the literature, and in 1920 the students of W. F. G. Swann<sup>7)</sup> took up the work as subsidiary to

investigations of the "residual" ionization caused by "penetrating" radiation. The work was begun by Miss K. M. Downey<sup>8)</sup> who measured residual ionization in a spherical chamber. In her investigations on the voltage effect in air at 40 atmospheres she found a change of only eleven percent when the voltage was increased from 600 to 1500 volts. In a later paper<sup>9)</sup> she presented another current-voltage curve at 52 atmospheres with voltages from 120 to 800 volts. It is of interest to note that Miss Downey's paper of 1920 contains the last reference to the work of Kaye and Laby<sup>4)</sup>, and consequently to that of Erikson<sup>3)</sup>, in the subject literature until 1932. H. F. Fruth<sup>10)</sup> extended the experiments on air up to 80 atmospheres, and investigated nitrogen, oxygen, and carbon dioxide.

James W. Broxon took up the work following Fruth and has published many results<sup>11)</sup>, in very few of which he includes data on variation of the current with respect to the voltage applied to his various spherical chambers. His treatise<sup>12)</sup> attempting to explain the observed results makes no mention of preferential or initial recombination.

By this time, historically speaking, nearly all observers in the field of cosmic ray research had begun to improve their accuracy (and the author is of the opinion that a parallel gain was undoubtedly made in their dispositions) by using higher pressures in the sensitive electroscopes. Although the work in which gamma rays were employed as the ionizing energy had clearly indicated that for the collecting fields used the number of ions collected in

a given time was not a linear function of the pressure, yet some of the pioneers in the field<sup>13),14)</sup> were of the opinion that for the hard cosmic rays the measured ionization would be directly proportional to the pressure. By direct comparison they found the results the same as for the gamma rays.

Apparently the earlier research had been nearly forgotten, and it remained for Millikan and Bowen<sup>15)</sup> and A. H. Compton, Bennett, and Stearns<sup>16)</sup> to point out the cause of this departure from linearity, their independent suggestions differing not in the least from those of Rutherford<sup>1)</sup>, Bragg and Kleeman<sup>2)</sup>, Erikson<sup>3)</sup>, and Laby and Kaye<sup>4)</sup>. The author wishes to show no disrespect for the men working in the subject, for the pioneer work seems to have been generally forgotten. As a particular example the following quotation<sup>17)</sup> will serve well: "At a pressure of 110 atmospheres the ionization is fifty percent lower than would be expected (proportionality to pressure), while the figures given in table I (current against voltage from 6 to 400 volts with a cylindrical ionization chamber !) make it quite clear that the failure to obtain complete electrical saturation cannot account for more than a few percent of this difference."

Upon the suggestion of the cause of the effect many directly related papers have ensued. Compton, Bennett, and Stearns presented a theoretical treatment of the problem of preferential recombination<sup>18)</sup>; it does not seem out of place to state here that if their

theory were correct the present investigation would have been fruitless, for according to their predictions a field of the order of  $4 \times 10^4$  volts would be required to appreciably effect the permanence of an ion pair. Their treatment also predicted a temperature variation which has led to further research.<sup>19), 20)</sup>

Experimental work dealing more directly with the present problem is that of Bowen<sup>21)</sup>, A. H. Compton and J. J. Hopfield<sup>22)</sup>, and Sievert.<sup>23)</sup> As the other papers will be considered more in detail later, it will suffice here to say that Sievert finds a 7000 volt field is not saturation value for air at 160 atmospheres.

Additional theoretical treatments have been presented by Harper<sup>24)</sup> and Gross<sup>25)</sup>. These will also be treated more in detail. At present the experimental work is far in advance of a complete theory for these "... experiments so simple to the experimentalist and so complex to the theoretician".<sup>24)</sup>

#### THE PROBLEM

Practically all of the ionization in a gas irradiated by  $\gamma$  rays is due not to the action of the  $\gamma$  rays directly but to the  $\beta$  rays ejected by them in Compton encounters between the photons and neighboring atoms. In the research here described the radiation was nearly monochromatic, the photons having an energy of  $2.65 \times 10^6$  electron volts; this energy completely transferred to an electron would give it a range of between 10 and 25 meters in air at N.T.P.

If any gas is contained in a thick walled vessel, and the gas pressure is low, then most of the ionizing  $\beta$  rays are ejected from the walls. According to the explanation propounded by Miss Downey<sup>8),9)</sup> and Broxton<sup>11)</sup>, as the pressure of the gas is increased, more and more of the  $\beta$  ray tracks are made to end within the gas; and thus when the pressure reaches a certain value, all the  $\beta$  rays are completely stopped within the gas, and no further increase in ionization would be observed by raising the pressure even higher. In contradiction to this simple explanation of the decrease of current per atmosphere as the pressure is increased, a fact observed by all the workers, Millikan and Bowen<sup>15)</sup> and Compton, Bennett and Stearns<sup>16)</sup> have pointed out that the number of ions formed within the chamber should be directly proportional to the pressure. The observed decrease of the current per atmosphere with increasing pressure is attributed to the inability of the ionization chambers to collect all, indeed even a small fraction, of those formed.

The suggestion of the two last mentioned papers comes about as follows:

Consider the ionization  $I$  in one cc. of gas located at the origin of coordinates. Let  $b$  be the number of ions formed per cm. of path by one  $\beta$  ray in the gas at unit pressure, and  $n$  the number of paths passing thru a square cm. arbitrarily taken normal to the  $x$  axis. Then in general  $I = b n p$  when  $p$  is the pressure in atmospheres. To evaluate  $n$  we shall consider two simplified cases:



Case I. An infinite gas chamber.

Let  $J$  be the intensity of  $\gamma$  rays and assume it a constant over the range  $\lambda$  of the  $\beta$  particles. If  $\alpha$  is the coefficient of absorption of the  $\gamma$  rays, then

$$n = \int_0^{\lambda} \alpha J dx = \alpha J \lambda .$$

Since  $\alpha$  is directly proportional to the pressure, and  $\lambda$  inversely proportional,

$$I \propto p$$

Case II. A finite chamber.

Let the distance from the cc. of gas considered to the wall be  $d$ . The number of paths  $n$  will now be due to the contributions from the gas and from the wall if  $d < \lambda$ , otherwise we return to case I. To a close approximation the coefficient of absorption for the gamma rays in the walls will be  $\alpha \rho_w / \rho_g$ , where  $\rho_w$  and  $\rho_g$  are the wall and gas densities. Also to a lesser degree of accuracy, the range of a  $\beta$  particle in the wall will be  $\lambda \rho_g / \rho_w$ .

$$\begin{aligned} \therefore n &= \int_0^d \alpha J dx + \int_d^{d + (\lambda - d) \rho_g / \rho_w} \alpha \rho_w / \rho_g \cdot J dx \\ &= \alpha \lambda J, \text{ identical with case I.} \end{aligned}$$

The primary theories of ionization all include an account of the decrease of the number of collected ions as the collecting field is lowered, and the explanation fits the facts fairly well. It is due to "volume" recombination, the occasional collision of the positive and negative ions formed. But when the ionization per unit volume is sufficiently small this simple theory does not explain the effect. Two additional recombination theories have been added. "Preferential recombination" is caused at high pressures by the formation of a negative ion by the ejected electron ( $\delta$  ray) within such a small distance from the parent positive ion that the pair are recombined by the action of their mutual attraction. Since  $\alpha$  or  $\beta$  rays ionize a gas along their paths, the ions in one of these columns may be in such propinquity that the electrostatic attraction between those of unlike signs causes many unions. This is termed "initial recombination". As in the case of "volume" recombination, the "preferential" and "initial" recombinations should be reduced by an increased collecting field.

All of the workers in the field of cosmic ray research have found it much more convenient to use ionization chambers of the cylindrical type, concentric spheres, or a fine wire in a large sphere, rather than a chamber allowing a constant collecting field. Defining average in the usual manner,  $\bar{E} = \frac{\int E dr}{\int dr}$ , we have for cylinders of radii  $b > a$

$$\bar{E} = \frac{2V}{(b+a) \log \frac{b}{a}} \quad \text{and for spheres}$$

$$\bar{E} = \frac{3 a b V}{(b^3 - a^3)}$$

where in each case  $V$  is the collecting voltage applied. Bowen<sup>21)</sup> has computed that the effective gradient used in nearly all recent measurements is of the order of 5 to 10 volts per cm.

As the author has mentioned in the introduction, the early experiments seem to have been generally overlooked from 1920 until 1932. (See for instance the letter of H. Zanstra and J. Clay, *Phys. Rev.*, 41, 679; 1932). So between the publications<sup>15), 16)</sup> indicating a possible cause of the observed decrease in current per atmosphere with increasing pressure and that<sup>18)</sup> predicting a field of the order of  $4 \times 10^4$  volts as necessary to observe an appreciable change, Bowen had constructed a parallel plate condenser ionization chamber and began an investigation on air. His results<sup>21)</sup> show without a doubt the theory of Compton, Bennett and Stearns to be far from correct, altho the preferential recombination which they treated is very likely the cause.

Nitrogen is known to be an "electronegative" gas, behaving much like the inert gases with respect to electrons. In fact the number of molecules which an electron will probably strike before becoming attached varies widely for all different gases.<sup>26)</sup> It was thus valuable to extend Bowen's work to other gases; especially to argon, which is now being extensively used in the cosmic ray electroscopes<sup>27)</sup>. There are also the theoretical predictions to check, altho at present only one<sup>25)</sup> allows calculation. These investigations have been conducted by the writer under the supervision of Professor

Bowen, and the results are herewith presented.

#### APPARATUS

The ionization chamber with which the problem was attacked is in many respects like that used by Laby and Kaye<sup>4</sup>), the number of plates being increased. It is a steel cylinder 12.5 cm. in diameter and 20 cm. in length designed to hold at least 100 atmospheres pressure (left in the picture); the lid is held in place by 15 tungsten-steel bolts. Inside are placed 8 plates of 10 cm. diameter which can be connected to a source of high potential; alternating with these are 7 collecting plates 7.5 cm. in diameter. These collecting plates are surrounded by grounded guard rings from which they are insulated by small amber lugs in grounded metal shields. The inside diameter of the guard rings is 7.7 cm. and the outside 10 cm. The distance between the potential and collecting plates is one centimeter, and we can thus collect the ions from a definitely known volume with a uniform field numerically equal to the applied voltage. Because of the large capacity of this condenser and the very weak source of ionization used, it was necessary to compensate for any changes in the battery potential. In the case of helium gas the number of ions collected from one atmosphere pressure changed the potential of the collecting plates at the rate of approximately 0.03 volts per hour! Any variation in the high potential because of battery fluctuations would induce potential fluctuations in the collecting plates completely overshadowing these extremely small measured changes. Hence a duplicate of the ionization chamber was built, except that it was enclosed in a light brass case; the high potential plates of the

Picture of Apparatus

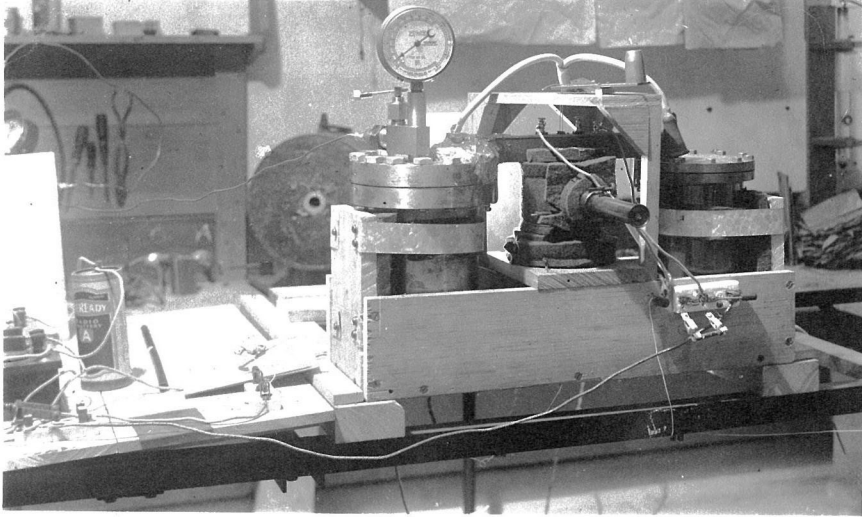


Diagram of Apparatus

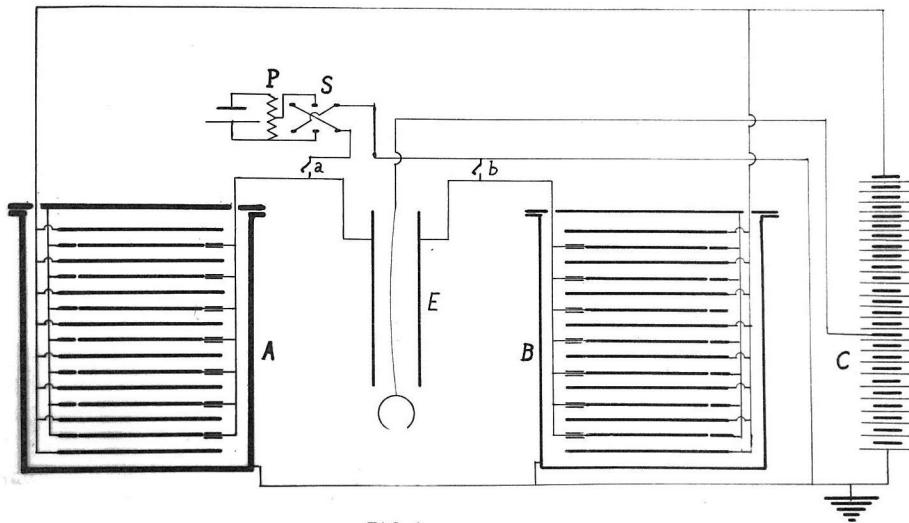


FIG. 1

ionization chamber A and the compensating chamber B were connected to the same battery as shown in Fig. 1, and the electrometer E was fixed to measure the difference in potential of the two sets of collecting plates. The two plates of the single string electrometer used were connected respectively to the two sets of collecting plates, the leads being completely electrostatically shielded, while a positive potential, changed from time to time throughout the experiment, was applied to the string. During an ordinary run the sensitivity was adjusted to about 500 divisions per volt. The interiors of the static shielding of the electrometer leads and the electrometer itself were kept dry by open contact with small containers of anhydrous  $P_2O_5$ .

A lead cylinder 30 cm. in diameter and 50 cm. long with a conical opening leading to one end was placed about two meters away from the ionization chamber. At the rear of the opening was placed a small quantity of radio-thorium, and the  $\gamma$  rays from this substance, filtered thru 13 mm. of lead, were used as the ionizing agent. The conical hole subtended a solid angle sufficient to envelope the ionization chamber. This  $\gamma$  ray gun was on a swivel so that the rays could be directed at the ionization chamber or turned to one side of it. By measuring the ionization with the  $\gamma$  rays passing thru the chamber and again when they were missing it, a determination of the number of ions which are due to the  $\gamma$  rays alone is possible and independent of the ionization produced by cosmic and  $\alpha$  rays.

The procedure in taking observations is as follows: Connect the collecting plates of the compensating chamber B to ground while

holding the collecting plates of the chamber A at some potential, usually 0.05 volts above ground, by means of the potentiometer P, and read the electrometer. Reverse the switch S, thereby putting the collecting plates of A at 0.05 volts below ground, and again read the electrometer. Now increase the negative potential a very small amount, and open the switches a and b simultaneously, leaving both sets of collectors floating. Start the stop watch when the electrometer string indicates the potential of the collecting plates of A to be 0.05 volts negative and stop the watch when the string indicates approximately 0.05 volts above ground. Now close the switches a and b and apply this positive potential to the collectors of A, recording the error made. Since the electrometer was adjusted to give a linear displacement with applied voltage on its plates, this error due to zero shift or change of sensitivity could be corrected. This then gives quite accurately the rate of change of potential difference between the collecting plates of A and B.

The relation of this measured rate of potential change to the ionization per cc. per second is given by Bowen<sup>21)</sup> as follows: "Let  $C_1$  be the electrical capacity between the collecting plates and the high potential plates of the ionization chamber,  $C_2$  the capacity to ground of the collecting plates, leads and electrometer plates. By symmetry these capacities are the same for compensating chamber. Let  $C_3$  be the capacity between the plates of the electrometer. It is then easy to show that if a charge  $Q_1$  is accumulated on the collecting plates of the ionization chamber and  $Q_2$  on these

plates of the compensating chamber the potential difference in volts across the electrometer is  $W$  where

$$W = (300(Q_1 - Q_2))/(C_1 + C_2 + 2C_3). \quad (1)$$

Likewise it may be shown that if the potential plates of the compensating cylinder are grounded while the potential of these plates in the ionization chamber is varied by an amount  $v$ , then the charges induced on the collecting plates (left floating) are such as to change the potential between the collecting plates of the two cylinders by an amount  $w$ , where

$$w = vC_1/(C_1 + C_2 + 2C_3). \quad (2)$$

$w$  is obviously the change in reading of the electrometer when this variation takes place and hence can be determined directly.

From (1) it is seen that the difference of the ionization currents in the two chambers is

$$\frac{dQ_1}{dt} - \frac{dQ_2}{dt} = \frac{dW}{dt} \frac{C_1 + C_2 + 2C_3}{300} \quad (3)$$

where  $dW/dt$  is the rate of change of potential across the electrometer, in volts per sec. But substituting from (2)

$$\frac{dQ_1}{dt} - \frac{dQ_2}{dt} = \frac{dW}{dt} \frac{vC_1}{300w} \quad (4)$$

Since, however, the distance between the potential and collecting plates is 1 cm.

$$C_1 = A/4\pi$$



where A is the total effective area of the collecting plates in each cylinder

$$\frac{dQ_1}{dt} - \frac{dQ_2}{dt} = \frac{dW}{dt} \frac{v}{300w} \frac{A}{4\pi}$$

The difference in the number of ions ( $N_1 - N_2$ ) collected per cc. per sec. in the two chambers is then, since the volume from which the ions are obtained is A cc and the charge on each ion is e,

$$N_1 - N_2 = \frac{1}{eA} \frac{dQ_1}{dt} - \frac{dQ_2}{dt} = \frac{dW}{dt} \frac{v}{w300 \times 4\pi e} \quad ."$$

A more careful consideration will show that the first assumption made is not strictly true, for altho the compensator is commensurate with the ionization chamber, their capacities are only equal if the gas pressures within are the same. Letting k be the variation from unity of the specific inductive capacity of the gas in chamber A, a quantity exactly proportional to the pressure<sup>28)</sup>, the expression is more correctly given by

$$N_1 - N_2 = \frac{dW}{dt} \frac{1}{300 \times 4\pi e} \left( \frac{v}{w} + k \right) ,$$

where  $\frac{v}{w}$  is determined with atmospheric pressure in both chambers.

An interesting by-product comes from the measurement of  $\frac{v}{w}$  at higher pressures; signifying these by primes we get the relation with that determined at one atmosphere to be

$$\frac{v'}{w'} = \frac{v}{w} (1 - k) + k$$

as a first approximation. Thus we might determine the quantity k experimentally.

When the  $\gamma$  rays are directed at the chamber A the number  $N_1$  consists of the ions due to  $\gamma$  rays,  $N_{1\gamma}$ , and those due to radiations from the walls of the chamber, the walls of the room, and cosmic rays,  $N_{1R}$ ; when the gun is turned aside it is only  $N_{1R}$ . Always  $N_2$  consists entirely of ions formed by uncontrolled radiations. Hence if we measure  $\frac{dW}{dt}$  when the gun is turned toward the chamber and again when it is turned away, the difference in the values will give us directly  $N_{1\gamma}$ . At very high pressures the second observed value  $N_{1R} - N_2$  is taken as  $N_{1R}$ , since the number collected from the compensator is nearly negligible.

A new U. S. Test Gauge, face diameter 6 inches, was used to measure pressures above one and below 50 atmospheres. This was calibrated on a gauge tester, and hence these values are not in error by more than 0.1 atmospheres. At one atmosphere the laboratory barometer reading was corrected by an open tube manometer attached to the gas inlet, but because of the very small tubing used, these values may be as much as 2% in error. Two gauges reading high pressures were connected to the apparatus, neither calibrated, and as a result these pressures may be as much as 10% off, probably too high if the new gauges of the cryogenic laboratory are assumed correct.

In all investigations a given collecting field was maintained on the system for several hours before any readings were taken to allow any charges accumulating on the insulators to take up a steady value.

Since the  $\gamma$  rays used gave a current about 20 times as large as the residual ionization current, the form of the ionization-voltage

curves was examined for an intermediate intensity of  $\gamma$  rays by filtering them thru an added 4.78 centimeters of steel. These investigations were made with the top pressures of argon and  $\text{CO}_2$ .

#### RESULTS

In order to fix the curves obtained by Bowen<sup>21)</sup> for air with those of the writer for other gases, data was taken for air at pressures of 0.98, 24.95, and 51 atmospheres. Multiplication of Bowen's values by the factor 0.69 placed the curves identical well within experimental error. A factor 0.805 multiplying Bowen's values for room radiation also gave nearly perfect agreement. These transposed values with those of the author for air are given in Fig. 2 and 6.

Laby and Kaye<sup>4)</sup> give tables for the variation of current with collecting field from 500 to 3500 volts at pressures of 8 and 15 atmospheres for air. If their results are fixed to Bowen's 10 atmosphere curve at the 1000 volt value, the curves match beautifully; and at the field value of 3500 volts, the curve has turned asymptotically to within 3% of the value at one atmosphere, showing definitely that with a sufficiently large gradient the ions can be practically all collected. This value is still much less than the  $4 \times 10^4$  volt gradient required by the Compton, Bennett, and Stearns theory.

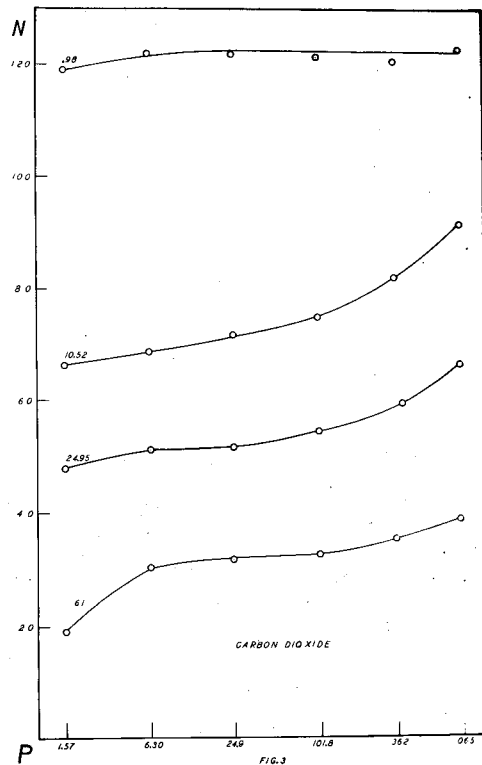
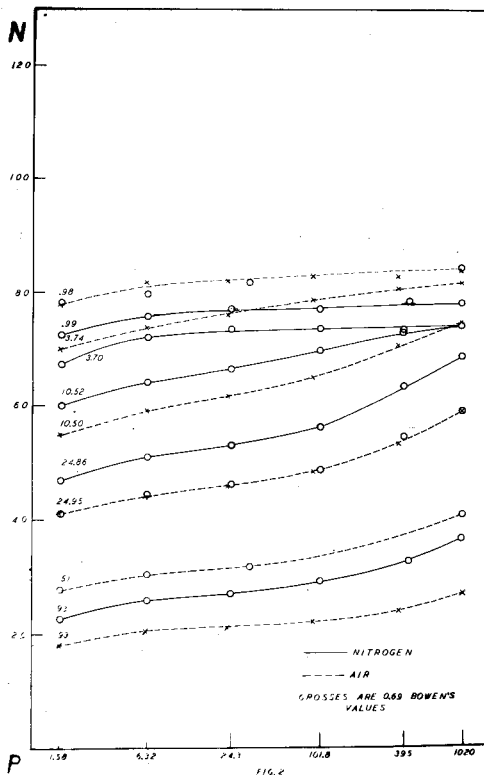
Commercial nitrogen bought from the Linde Air Products Co. was the next gas to be admitted into the chamber. A hy-vac pump was connected to the gas inlet and the chamber evacuated to less

than 3 mm. pressure before admitting the gas. In the case of the cheaper gases the chamber was filled to atmospheric pressure and re-evacuated several times before any observations were made. Mr. W. M. Zaikowsky analysed the nitrogen chemically for  $\text{CO}_2$  and  $\text{O}_2$ , finding no trace of  $\text{CO}_2$ , and  $0.93 \pm .025\%$  of  $\text{O}_2$  present. The accompanying table gives the number of ions collected per cc. per second per atmosphere for the different pressures and collecting fields. The results are shown graphically in Fig. 2 whereon the number of ions per cc. per sec. per atmosphere is plotted against a logarithmic scale of the field.

TABLE I

No. of Ions per cc. per sec. per atm. from nitrogen

Field in volts per cm.	Pressure in atm.	0.99	3.70	10.52	24.86	93
	1.58		72.3	67.1	60.0	46.7
6.32		76.3	71.9	64.0	51.1	26.0
24.3		76.9	73.4	66.4	53.1	27.1
101.8		77.2	73.4	69.7	56.2	29.3
395.		78.2	73.1	72.7	63.2	32.6
1020		78.0	74.0	73.9	68.6	36.7



These results are not in agreement with the results of Broxon<sup>11)</sup>: "At all pressures at which observations were made, from 0.82 to 167 atmospheres, the ionization in  $N_2$  was greater than that in air at corresponding pressures." It is directly evident that less recombination takes place in  $N_2$  than in air as we might predict from Loeb's statement.<sup>26)</sup> However, Loeb has proven that the number of collisions which a free electron makes with the molecules before it becomes permanently attached depends to a large extent on the purity of the gas. Measurements of various authors on ionization in  $N_2$  differ widely. The author

had planned to investigate the dependence of the shape of the curves on the purity of the  $N_2$  and had constructed an oxygen remover containing copper turnings for the purpose. But the valve on the tank developed a leak and the gas practically all escaped before this research could be carried out.

No good explanation can be found for the coincidence of the 3.70 and 10.52 atmosphere curves at the highest voltage; the higher pressure curve is more reliable, and the 3.70 curve should undoubtedly approach more rapidly to the 0.99 atmosphere curve.

According to Loeb,  $CO_2$  is more electropositive than  $N_2$  and more electronegative than air. We should thus expect less recombination to take place in  $CO_2$  than in air. The number of ions collected per cc. per sec. per atmosphere from  $CO_2$  is given in Table II and shown graphically in Fig. 3. The gas used was commercial  $CO_2$  purchased from the Pure Carbonic Company of Los Angeles; no chemical analysis was made of its purity.

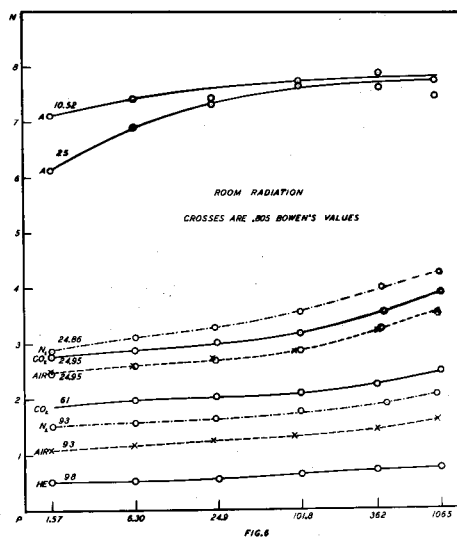
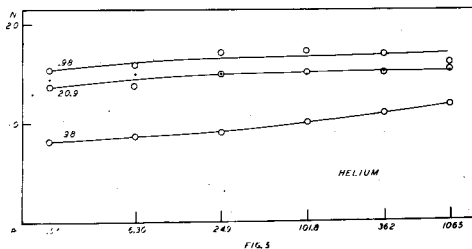
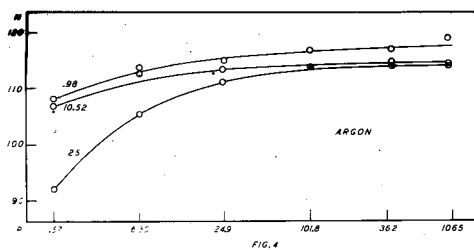
TABLE II

No. of Ions per cc. per sec. per atm. from carbon dioxide.

Field in volts per cm.	Press. in atm.			
	0.98	10.52	24.95	61
1.57	118.8	66.2	47.8	18.8
6.30	121.7	68.5	51.1	30.2
24.9	121.4	71.5	51.5	31.5
101.8	120.9	74.6	54.3	32.4
362	120.1	81.6	59.1	35.1
1065	122.3	91.1	66.0	38.4

No remarkable differences are found among the curves for air,  $N_2$ , and  $CO_2$  of the purity used. They all show high recombination effects which are noticeably changed by increasing the field collecting the ions. Conclusions which may be drawn from these curves are identical with those presented by Bowen<sup>21</sup>); they will be presented after a discussion of the results obtained by the author for two noble gases.

The argon examined was obtained from General Electric. A chemical analysis made by Mr. C. E. Hablutzel, Jr. revealed no trace of  $CO_2$  and less than 0.2% of  $O_2$ ; a weight determination gave no evidence of  $N_2$ , but the method was not very accurate. The ionization chamber was more completely evacuated before introducing the argon than it had been for the  $N_2$  or  $CO_2$ , and was re-evacuated once after filling to atmospheric pressure. Because of the size of the chamber and limited quantity of argon the



highest pressure curve is 25 atmospheres. Data was taken at that pressure with the  $\gamma$  rays at full strength and when filtered thru 4.78 cm. of iron. The column of Table III marked  $4.55 I_2$  is the result of multiplying the data obtained with the smaller intensity  $I_2$  by the factor 4.55. It is observed to give values well within the experimental error with the possible exception of the point at the lowest gradient, an effect also observed by Bowen for air. The ionization was studied for four different pressures and the results all presented in Table III; curves for three of the pressures are given in Fig. 4, the black dots thereon being the values for 3.72 atmospheres pressure.

TABLE III

No. of Ions per cc. per sec. per atm. from argon.

Field in volts per cm.	Press. in atm.			25	
	0.98	3.72	10.52	I	$4.55 I_2$
1.57	108.2	105.9	106.9	92.1	95.1
6.30	113.8	112.3	112.7	105.5	105.5
24.9	115.1	112.8	113.5	111.2	111.2
101.8	117.0	114.0	114.0	114.0	112.6
362.	117.2	114.1	114.9	114.1	113.8
1065.	119.2	114.2	114.6	114.2	111.9

The gradual sloping of the one atmosphere curve beyond the 100 volt field value may be attributed to ionization by posi-



tives, which, according to Dr. Otto Beeck, sets in at 100 volts in argon; but the accuracy of these readings is not sufficient for vindication.

Table IV gives the data obtained for helium; these results are shown graphically in Fig. 5, the black dots representing the results for 10.52 atmospheres pressure. The helium used was kindly furnished by Professor A. Goetz from the supply of the cryogenic laboratory; no analysis of its purity was made.

TABLE IV

No. of Ions per cc. per sec. per atm. from helium.

Field in volts per cm.	Press. in atm.	0.98	10.52	20.9	98
	1.57		13.4	14.4	13.7
6.30		15.9	14.9	13.8	8.77
24.9		17.2	14.9	15.0	9.12
101.8		17.3	14.9	15.1	10.0
362.		17.0	15.2	15.1	11.0
1065.		16.1	15.2	15.4	11.8

In Table V are given the values of the difference  $N_{IR} - N_2$  which for the pressures used is nearly the number of ions collected per cc. per second per atmosphere when the ionizing radiations are only  $\alpha$  rays from the chamber walls and cosmic rays. It is obvious from these results that even when the intensity of

ionization is as low as one twentieth that of the  $\gamma$  rays, the pressure and field dependence, shown in Fig. 6, is of the same form as for the greater intensities.

TABLE V

No. of Ions per cc. per sec. per atm. by room radiation

Gas	Press. in atm.	Field in volts per cm.					
		1.57	6.30	24.9	101.8	362	1065
Air	24.95	2.45	2.60	2.70	2.87	3.26	3.53
	93	1.08	1.17	1.25	1.32	1.42	1.60
N <sub>2</sub>	24.86	2.86	3.10	3.28	3.56	4.00	4.26
	93	1.51	1.57	1.65	1.76	1.89	2.05
CO <sub>2</sub>	24.95	2.76	2.88	3.01	3.19	3.56	3.92
	61	(2.76)	1.98	2.05	2.10	2.24	2.49
A	10.52	7.10	7.41	7.43	7.74	7.90	7.48
	25.0	6.12	6.91	7.52	7.66	7.64	7.77
He	98	.502	.530	.565	.635	.702	.742

The subsidiary experiment performed prior to the absolute evaluation of the number of ions collected, that is a determination of the ratio of  $v$  to  $w$  as discussed previously, gave values of this quotient of the order of 2.72 when both chambers held one atmosphere pressure. Several times during the period of research it was necessary to dismantle the electrometer leads of their shield-

ing, and once the electrometer had to be removed for repair. These interruptions affected the second and third decimal places of the quotient. Attempted determinations of the departure from unity of the specific inductive capacity were not very successful in absolute value, for when the pressure was large enough to offer perceptible variation, the ionization by uncontrolled radiations discharged the collecting plates at such a rate that an accurate measurement of  $w$  could not be made. Results of these measurements are hardly worth presenting, but in all cases the order of magnitude was correct.

Each value of  $N_{\gamma}$  which is given in the above Tables I, II, and III is the average result obtained from not less than ten separate readings of the time rate of change of potential with the  $\gamma$  rays passing thru chamber A, and ten with the rays turned aside. Many of these points are averages of twenty such observations. With high pressures in the chamber the measured times of discharge either with or without the  $\gamma$  radiation are essentially constant; the zero drift and change in sensitivity of the electrometer make the longer time readings at low pressure more inexact. This last mentioned cause of error affects particularly the values in Table IV, doubly so because of the very low specific ionization in helium. The values for this gas were found by measuring the change of potential over half-hour intervals, instead of the usual measuring of required time for a 0.1 volt change. Only seven readings with and a like number without the  $\gamma$  rays were averaged for

this one gas.

Attention should again be called to the fact that the lowest pressure used could not be accurately measured because of the small tubing between the chamber and the manometer. Also the determination of  $N_{IR} - N_2$  for atmospheric pressure in each tank was never accurate because of zero drift and change of sensitivity.

The tabular values are given as though no variation was observed in the collecting potentials. The graphs show the observed values.

Since the observations were continued over a period of several months, it was of course necessary to correct all observed values to the same intensity of  $\gamma$  radiation. According to Rutherford, Chadwick, and Ellis<sup>29)</sup> the half-life of radio-thorium is 1.90 years, which requires a correction of 0.1% per day. Five months after the first data was collected on air at 25 atmospheres this experiment was performed again, and with the above specified correction for decay, the values determined were identical to within 0.5%.

#### CONCLUSIONS

Bowen's results for ionization in air<sup>21)</sup> supplemented by those of the author, especially for the two noble gases, "provide conclusive evidence that the falling off of ionization per atmosphere at higher pressures is largely due to lack of saturation rather than to the mechanism suggested by Broxon." With an applied

collecting field of more than a few volts per cm below which volume recombination plays an important role, Broxon's simple explanation of the observed decrease in current per atmosphere would require essentially the same form for results taken with air or argon. Furthermore it would not allow the observed pronounced increases of current with raised collecting gradient. In showing the number of ions formed per cc. per sec. in a finite ionization chamber to be proportional to the pressure, the writer mentioned, and Bowen has pointed out that "there may still be a slight variation of the type suggested by Broxon due to a small difference in efficiency of production of  $\beta$  rays by the gas and by the steel walls of the chamber." Workman<sup>30)</sup> has recently shown "that the effect of these materials is at most 30% and is probably in the opposite direction; i.e., it should cause the ionization per atmosphere to increase slightly with the pressure." Compton, Bennett, and Stearns<sup>18)</sup> have called attention to the fact that since both photoelectrons and recoil electrons have ranges which depend on the exciting radiation, the dependence of ionization on pressure would certainly not be the same for  $\gamma$  as for cosmic ray energies if the ionizing  $\beta$  rays were all ejected from the walls. The work of Millikan and Cameron<sup>13)</sup>, and Hoffmann and Lindholm<sup>14)</sup>, checked recently by the three men above, proves that if such a dependence on the radiation frequency exists at all, it is less than their probable error. In his latest publication Broxon<sup>11)</sup> states that he finds

the ratio of the current produced in air by  $\gamma$  radiation to the current from cosmic rays to be a function of the pressure, increasing from 4.21 for a pressure of 0.82 atm. to 5.20 for 80 atm. J. J. Hopfield<sup>22)</sup> finds the pressure effect also in argon, the ratio at 36 atm. being 8.55 and at 74 atm., 7.19; this he frankly attributes to insufficient voltage for saturation.

Reference to any of the curves by Bowen or the author will convince the reader that for effective fields as low as those used in Broxon's or Hopfield's measurements, volume recombination no doubt takes a heavy toll, and Bowen has already pointed out that the shape of the curves for these low gradients is probably dependent on the intensity of ionization. (See however the very recent publication of J. J. Hopfield, Phys. Rev. 43, 675; 1933.)

Beyond qualitative agreement the writer is at a loss to find direct comparison with other authors, for other than Laby and Kaye<sup>4)</sup> and Bowen<sup>21)</sup>, none of them found the advantages of a known uniform potential gradient adequate compensation for the intrinsic difficulties of a parallel plate ionization chamber. Taking the number of ions collected per cc. per sec. with one atmosphere pressure and the highest field value as the number formed per cc. per second per atm., the following agreement of relative ionization is found with the International Critical Tables for  $\alpha$  rays, the values for fast  $\beta$  rays not being given for all the gases used:

Gas	Air	CO <sub>2</sub>	A	N <sub>2</sub>	He
	1	1.47	1.36	0.93	0.21
I.C.T.	1	1.49	1.24	0.94	0.22

Qualitatively the results presented agree with all work related to the subject. Work is now in progress to investigate the effect of the purity of nitrogen, and these results will be included in the publication of this thesis.

Mr. Dwight O. North is presenting a theoretical treatment of preferential recombination as his thesis in partial fulfillment of the requirements for the degree of Doctor of Philosophy, and the reader will find therein a complete resumé of the three mentioned types of recombination. Only a brief treatment of the problem will be given here.

As was stated on page 8, the elementary theory of recombination failed to give agreement with observed values when the ionization was small. However, when the impressed collecting field is small and the pressure relatively high, the time required for the ions to travel to the collecting plates, 0.5 cm. average for that used in this experiment, is so large that kinetic motion may cause many unions. That such is the case may be seen from the agreement of the low voltage region of the curves with those presented by Rutherford<sup>1)</sup> for ionization by  $\alpha$  particles. For higher potential gradients the time allowed for Brownian movements

is not ample to permit enough volume recombination to explain the observed lack of saturation.

By treating ion diffusion in a zero electrostatic field, Compton, Bennett, and Stearns<sup>18)</sup> developed a theory of preferential recombination which qualitatively explained the departure from linearity of the current-pressure curve. Quantitatively it did not allow perceptible increase in current with collecting gradients of less than about  $4 \times 10^4$  volts per cm. Harper<sup>24)</sup> has also made an investigation of preferential recombination and computed the field strength at the boundary of a "critical sphere" about an ion to be one tenth as great as that given previously. Thus he states that his theory allows a practicable field "large enough to reduce preferential recombination considerably." But in reviewing the article North discovered an omission of a factor 3 in Harper's computation of the critical radius, and this brings the value of required gradient back to the Compton, Bennett, and Stearns value. North has borrowed from the Compton, Bennett, and Stearns paper an empirical equation for the probability of finding an electron at a distance from its parent ion greater than the critical radius, and with this one empirical equation has obtained reasonable agreement with Bowen's results for air and those of the author for argon.

Harper criticizes all work treating preferential recombination alone and neglecting initial recombination. "It can be seen from the photographs of C. T. R. Wilson (Proc. Roy. Soc. 104 A,



192; 1923) that the average separation of two pairs in a group is comparable with the average separation of the components of a pair; hence in general preferential recombination is seriously complicated by initial recombination, and in fact both occur to a similar extent." Gross<sup>25)</sup> has applied theoretical work done by Jaffé for  $\alpha$  particle ionization to the problem of initial recombination for  $\beta$  particle ionization. Except for North's unpublished work this theory is the only one permitting quantitative comparison with the writer's results. Gross' final equation gives for the number of ions per cc. per sec. per atm. an expression

$$N = \frac{c_1}{1 + c_2 p \log \frac{c_3 p}{E}}$$

where  $E$  is the applied collecting field and  $c_1$ ,  $c_2$ ,  $c_3$  arbitrary constants. To account for volume recombination with low fields and large currents he corrects the above value to

$$N' = N(1 - \exp. -c_4 \frac{E}{\sqrt{p^2 N}}),$$

this of course giving agreement with the large curvature for those fields. Using the reduced values for air at the highest voltage and pressures 3.74, 10.5, and 24.95, the constants of the first equation become

$$c_1 = 82.18 \quad c_2 = 7.16 \times 10^{-3} \text{ atm.}^{-1}$$

$$c_3 = 369 \text{ volts cm.}^{-1} \text{ atm.}^{-1}$$

Plotted to a logarithmic scale as are the figures of this thesis, Gross' equation for a constant pressure becomes a hyperbola. As the voltage is decreased from the highest value, these hyperbolae fall much too rapidly to fit the observed results, especially for the 10.5 and 24.95 atmosphere pressures. The constant  $c_3$  according to Gross is inversely proportional to the mobility of the ions. He candidly concedes "die noch wesentlich höheren Ströme, die Compton in Argon erhalten hat, lassen sich nicht aus einer erhöhten Beweglichkeit allein erklären." The big fault in treating the problem by initial recombination is no doubt the fact that the number of ion pairs generated per cm. of path by a high speed  $\beta$  particle is extremely low compared with the number generated per cm. by an  $\alpha$  particle, the problem to which Jaffé originally applied the equations. As is well known there are only 35 to 60 ion pairs per cm. along the N.T.P. path of a long range  $\beta$  particle granting separations of the order of  $1.5 \times 10^{-2}$  cm. between pairs. This is much too large to be seriously given advantage over attractions at distances of  $1.88 \times 10^{-6}$  cm. as required by preferential recombination.

Without much alteration the present form of apparatus used in these experiments could be used in measuring the effects of known amounts of impurities in the different gases, and perhaps in measuring the temperature effect with sufficient accuracy to decide definitely upon the theory to be accepted. Accurate

temperature effect measurements would be very valuable in cosmic ray work using high altitude balloon flights. In consideration of the very recent paper by Hopfield<sup>32)</sup> it would also be advisable to extend the work on argon to several more intensities of radiation.

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## BIBLIOGRAPHY

1. "Radio-Activity", pp. 32-33; 1904.
2. Bragg and Kleeman, *Phil. Mag.* 11, 466; 1906.
3. H. A. Erikson, *Phys. Rev.* 27, 473; 1908.
4. T. H. Laby and G. W. C. Kaye, *Phil. Mag.* 16, 879; 1908.
5. W. Wilson, *Phil. Mag.* 17, 216; 1909.
6. D. C. H. Florance, *Phil. Mag.* 25, 172; 1913.
7. W. F. G. Swann, *J. Frank. Inst.* 209, 151; 1930.
8. K. M. Downey, *Phys. Rev.* 16, 420; 1920.
9. K. M. Downey, *Phys. Rev.* 20, 186; 1922.
10. H. F. Fruth, *Phys. Rev.* 22, 109; 1923.
11. J. W. Broxon, *Phys. Rev.* 27, 542 (1926); 37, 1320 (1931);  
38, 1704 (1931); 40, 1022 (1932); 42, 321 (1932).
12. J. W. Broxon, *Phys. Rev.* 28, 1071; 1926.
13. Millikan and Cameron, *Phys. Rev.* 31, 922 (1928); 37, 237  
(1930).
14. Hoffmann and Lindholm, *Gerlands Beiträge zur Geophysik* 22,  
23 (1928); Hoffmann, *Zeit. f. Phys.* 69, 704 (1931).
15. R. A. Millikan and I. S. Bowen, *Nature* 128, 582; Oct. 3, 1931.
16. A. H. Compton, R. D. Bennett and J. C. Stearns, *Phys. Rev.* 38,  
1565; Oct. 15, 1931.
17. G. T. P. Tarrant, *Proc. Roy. Soc. A* 135, 223; 1932.
18. A. H. Compton, R. D. Bennett and J. C. Stearns, *Phys. Rev.*  
39, 873; 1932.

19. N. S. Gingrich, Phys. Rev. 41, 679; 1932.
20. Kurt Wolff, Zeit. f. Phys. 75, 570; 1932.
21. I. S. Bowen, Phys. Rev. 41, 24; 1932.
22. A. H. Compton and J. J. Hopfield, Phys. Rev. 41, 539 (1932);  
J. J. Hopfield, Phys. Rev. 42, 904 (1932).
23. R. M. Sievert, Nature 129, 792; 1932.
24. W. R. Harper, Proc. Camb. Phil. Soc. 28, 219 (1932); 29,  
149 (1933).
25. B. Gross, Zeit. f. Phys. 78, 271 (1932); 80, 125 (1933).
26. Loeb, Kinetic Theory of Gases, pp. 507-515.
27. A. H. Compton, Phys. Rev. 43, 337; 1933.
28. Jean's "Electricity and Magnetism" p. 130.
29. Rutherford, Chadwick, and Ellis "Radiations from Radio-  
active Substances", 1930.
30. E. J. Workman, unpublished research at C.I.T., 1932.
31. G. Jaffé, Phys. Z. S. 30, 849 (1939); Ann. d. Phys. 42,  
303 (1913).
32. J. J. Hopfield, Phys. Rev. 43, 675; May 1, 1933.