surfaces of both electrodes. Rutherford[12] found in 1901 that unequal currents resulted from intense, unsymmetrical ionization of dry air. It is rather interesting that with the arrangement he employed, Rutherford actually collected greater negative than positive ion currents. With the ionization confined to the immediate neighborhood of the high potential electrode, he found for certain impressed voltages that the ratio of the negative to the positive ion currents received at the collecting electrode equalled the ratio of the mobilities of the corresponding ions in air. At both higher and lower voltages the ratio approached unity. The difference between currents of the two signs was also considerably decreased by the introduction of various vapors, particularly alcohol. In our observations the numerical difference between the weak currents of the two signs does not appear to depend much upon the pressure or intensity of the collecting field. The "positive" ion current values of Fig. 5 collected with a very low impressed P.D. show the effect of contact potentials, of course.

[12] E. Rutherford, Phil. Mag. 2, 210 (1901).

JULY 1, 1938 PHYSICAL REVIEW VOLUME 54

The Analysis of High Gradient, High Pressure, Gamma-Ray, Air Ion Current Measurements, by Zanstra's Adaptation of Jaffé's Columnar Theory

JAMES W. BROXON AND GEORGE T. MERIDETH
University of Colorado, Boulder, Colorado

(Received April 27, 1938)

A brief survey of theories advanced to explain the chief characteristics of high pressure ionization current measurements is given. The equation developed by Zanstra from Jaffé's theory of recombination of the ions formed by a single particle, yielding a linear relation between the reciprocal of the measured current and a function of the collecting field intensity and pressure, was applied to our high pressure, gamma-ray, ionization current measurements. The curves yielded were not linear, but departure from linearity was rather small in the region of high gradients. By extrapolation, "saturation" current values were obtained. These "saturation" currents were found to vary linearly with the air density over certain pressure ranges, as found by Clay and van Tijn, but changes in slope at very high pressures would indicate a modification of Clay's "wall" radiation theory if the analysis is regarded as reliable.

THAT measured gamma-ray ionization currents in air at high pressures depend upon the pressure in much the same manner as do cosmic-ray ionization currents has been demonstrated by Compton, Bennett and Stearns[1] and by Broxon.[2] Tarrant[3] made a similar observation consequent to his gamma-ray measurements. Gross[4] pointed out that Florance's[5] beta-ray ionization pressure curves also closely resemble the gamma-ray curves of Bowen[6] and the cosmic-ray curves of Broxon,[7] over Florance's range of 70 atmospheres. This situation has necessitated theoretical treatment designed to clarify those ionization phenomena which result in measurements of such striking similarity associated with these radiations of different nature and vastly different penetrating power. It has also provided us with the means of investigating and interpreting high pressure cosmic-ray ionization measurements by conducting gamma-ray ionization current measurements under conveniently controllable conditions.

Attempted explanations of the characteristics of recent measurements of cosmic-ray and gamma-ray ionization currents at high pressures are interestingly similar to initial attempts at explaining alpha-ray and gamma-ray currents measured a quarter of a century earlier. Many of the same characteristics are outstanding in

[1] A. H. Compton, R. D. Bennett and J. C. Stearns, Phys. Rev. 39, 873 (1932).
[2] J. W. Broxon, Phys. Rev. 42, 321 (1932).
[3] G. T. P. Tarrant, Proc. Roy. Soc. A135, 223 (1932).
[4] B. Gross, Zeits. f. Physik 80, 125 (1933).
[5] D. C. H. Florance, Phil. Mag. 25, 172 (1913).
[6] I. S. Bowen, Phys. Rev. 41, 24 (1932).
[7] J. W. Broxon, Phys. Rev. 37, 1320 (1931).

both instances, and the fundamental points of view have been duplicated.

To explain relations between current and intensity of collecting field observed in connection with work on recombination of alpha-ray ions, Bragg and Kleeman,[8] in 1906, used a suggestion made by Rutherford[9] in 1904, assuming a "process of recombination of newly-formed ions with the atoms from which they have just been separated. The effects of it are proportional to the number of ions formed in a cubic cm in unit time, not to the product of the existing numbers of positives and negatives. They are independent of the shape of the ionization chamber, and in this they differ from those of general recombination. They depend directly on pressure, and vary greatly from gas to gas." They did some quantitative work, deciding that fields of the order of 1000 volts/cm would be necessary to affect this "initial" recombination considerably.

In 1908 Erikson[10] used this "initial" (sometimes called "preferential" or "selective") recombination hypothesis in explaining his extensive measurements of high pressure gamma-ray ionization currents.

However, Moulin,[11] in 1911, found this inadequate to explain his measurements, particularly with reference to the dependence of experimental currents upon collecting fields, and invoked a suggestion he attributed to Langevin (apparently 1902 or 1903) that the preponderance of recombination among the ions formed by a single ionizing particle is strikingly effective, writing: *Pour interpréter le phénomène de recombinaison initiale, M. Langevin a proposé une autre hypothèse qui fait intervenir une recombinaison générale entre tous les ions produits par une même particule α, sur laquelle un champ extérieur doit avoir une action marqueé puisqu'il peut diminuer le nombre total de rencontres qui ont lieu entre ces différents ions. Cette interprétation, qui a servi de point de départ aux recherches qui vont suivre, est basée sur l'hypothèse que les ions produits par chaque particule α s'éloignent relativement peu de la trajectoire et sont, initialement, distribués en colonnes, . . .*

Jaffé,[12] in 1913, proceeded further with the "columnar" (sometimes called "initial") recombination development of Moulin, deducing formulae relating rate of collection of ions to magnitude and direction of the collecting field and involving the coefficients of ionic recombination, mobility, and diffusion. Jaffé assumed an initial probable cylindrical distribution of the ions, and set up a differential equation for which he was able to obtain only an approximate solution.

Meanwhile, in 1909, Wilson[13] had explained the current-pressure relation he had observed in gamma-ray and natural ionization current measurements extending to 40 and 45 atmospheres, in terms of the supposed considerable contribution to the ionization by secondary radiations excited in the walls of the ionization chamber. He attributed his "wall" radiation hypothesis to McLennan[14] who worked on gamma-ray subsidiaries in 1907. Wilson offered some mathematical development.

After the World War when the study of the penetrating (cosmic) radiation was resumed, certain investigators, notably Professor Swann, appreciated the advantages offered by the high pressure ionization chamber. Measurements of this type again raised many of the old questions.

In 1920 Downey[15] employed the "wall" radiation hypothesis to explain the natural ionization current-pressure relation she observed at pressures extending to 58 atmospheres. In 1926 Broxon[16] extended the pressure range and provided some mathematical development of the hypothesis, later[2] concluding that it was inadequate to explain all the phenomena.

In 1932 Compton, Bennett and Stearns[1] pointed out inadequacies in the "wall" hypothesis, and applied the hypothesis of "initial" recombination of ions with atoms from which they were separated. They were able to explain the cosmic-ray current-pressure relations observed by Broxon,[7]

[8] W. H. Bragg and R. D. Kleeman, Phil. Mag. **11**, 466 (1906).
[9] E. Rutherford, *Radioactivity* (1904), p. 33.
[10] H. A. Erikson, Phys. Rev. **27**, 473 (1908).
[11] M. Moulin, Ann. de chim. et phys. **22**, 26 (1911).

[12] G. Jaffé, Ann. d. Physik 4, **42**, 303 (1913).
[13] W. Wilson, Phil. Mag. **17**, 216 (1909).
[14] J. C. McLennan, Phil. Mag. **14**, 760 (1907).
[15] K. M. Downey, Phys. Rev. **16**, 420 (1920); **20**, 186 (1922).
[16] J. W. Broxon, Phys. Rev. **28**, 1071 (1926). (Also see reference 7.)

as well as dependences upon temperature and upon the nature of the gas. They concluded that in air at 20°C fields of the order of 40,000 volts/cm would be necessary to affect this "initial" recombination.

At about the same time Millikan and Bowen[17] also drew attention to the "initial" recombination hypothesis, but offered no mathematical treatment. Bowen,[6] Gross,[18] Lea,[19] and Harig[20] have all objected to the "initial" recombination treatment by Compton, Bennett and Stearns[1] because of their conclusion relative to the lack of dependence of current upon field intensity at high values less than the critical value they specify.

Gross,[18, 4] by means of certain assumptions relative to dependence of the ion constants upon pressure and temperature, adapted Jaffé's[12] "columnar" theory formulae to high pressure measurements, including consideration of "general" or "volume" recombination not restricted to columns. He succeeded in approximating the high density gamma-ray current-pressure curves of Erikson[10] at the several collecting field intensities he used, Broxon's[7, 21] cosmic-ray curves in air and nitrogen, Bowen's[6] low density gamma-ray curves, and Florance's[5] beta-ray curves, as well as temperature effects observed at high pressures.

Harig[20] also used the "columnar" hypothesis and Jaffé's calculations to explain Bowen's observations. Lea[19] criticized Gross' treatment of the problem, and introduced the assumption that the ion distribution is better represented by "clusters" symmetrical relative to points than by "columns" symmetrical relative to lines, these clusters perhaps being arranged in columns. He concluded that "cluster" recombination may roughly be regarded as accounting for the dependence of currents upon pressure, and "columnar" recombination, for the dependence upon the collecting field.

Zanstra[22] has written formula (32′) of the paper by Jaffé[12] in the form

$$\frac{1}{i}=\frac{1}{I}+\frac{q}{I}f(x). \tag{A}$$

Here i is the measured ionization current and I the supposed "saturation" current which could be obtained by an infinite collecting field, so that i/I is the same as Jaffé's N_∞'/N_0, both "initial" and "general" recombination being neglected and only "columnar" recombination considered.

In this equation, $q=\alpha N_0/8\pi D$, where α is the "coefficient of recombination" among ions of a column, N_0 is the initial line density of ions in a column, and D is the ionic "diffusion coefficient." Also,

$$f(x)=e^x(j\pi/2)H_0^{(1)}(jx),$$

where $j=\sqrt{(-1)}$ and $H_0^{(1)}$ is designated by Gross[18] *eine Hankelsche Funktion nullter Ordnung*, values of $jH_0^{(1)}(jx)$ being given in *Table of Functions* by Janke and Emde. In this,

$$x=(buE\sin\phi/2D)^2,$$

where b is a "column parameter" proportional to the initial mean radial displacement of the ions in a column, u is the ionic "mobility" assumed the same for ions of both signs, E is the intensity of the collecting field, and ϕ is the angle between column axis and collecting field (or $\sin^2\phi$ may be replaced by an "average" value unless ϕ is predominantly small).

Assuming u/D independent of the pressure, and the average $(b\sin\phi)$ inversely proportional to the pressure, Zanstra decided that for high pressure gamma-ray ionization currents in air collected with strong fields, $x=1.24(10)^{-4}(E/p)^2$ to a sufficient degree of approximation, p being the pressure in atmospheres and E being measured in volts/cm. Zanstra pointed out that as E approaches ∞, $f(x)$ approaches 0. Hence the $(1/i)$ intercept of the line represented by Eq. (A), linear in $f(x)$, must represent the reciprocal of the "saturation" current collected at infinite field intensity if the relation is correct. Graphs representing $f(x)$ for values of x from 10^{-8} to 12 are given by Zanstra.

Zanstra emphasized that the relation should be obeyed only at high field intensities where "general" recombination is eliminated. He ap-

[17] R. A. Millikan and I. S. Bowen, Nature **128**, 582 (1931).
[18] B. Gross, Zeits. f. Physik **78**, 271 (1932).
[19] D. E. Lea, Proc. Camb. Phil. Soc. **30**, 80 (1933–34).
[20] G. Harig, Physik. Zeits. Sowjetunion **5**, 637 (1934).
[21] J. W. Broxon, Phys. Rev. **38**, 1704 (1931).
[22] H. Zanstra, Physica **2**, 817 (1935).

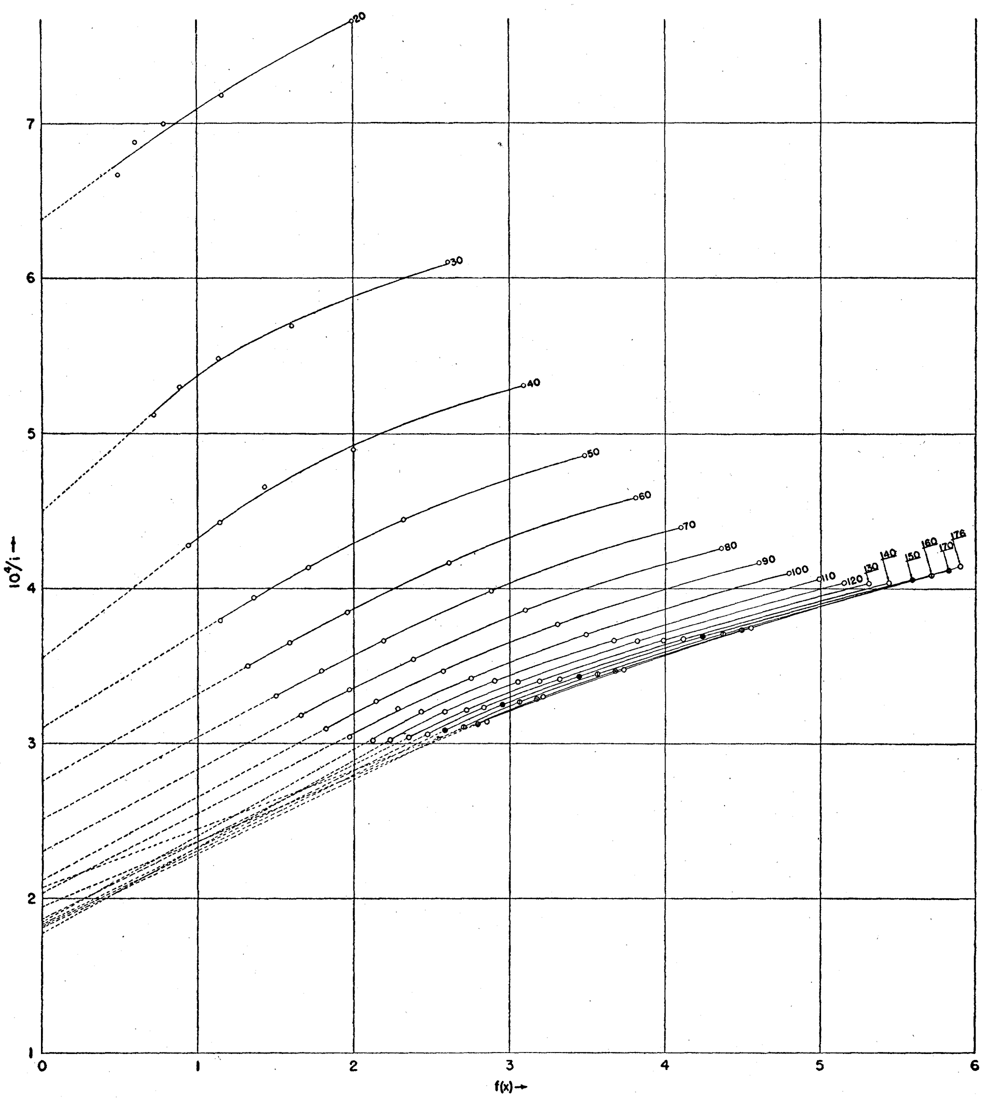

FIG. 1. Jaffé-Zanstra function curves corresponding to our measurements with central source. Air at 28°C. Central source of gamma-radiation. Numbers at upper ends of curves represent specific gravity relative to air at N.T.P.

plied the theory to Erikson's[10] measurements, finding the linear relation obeyed by those data, and extrapolated to find the "saturation" currents. He also pointed out that for specified field intensities the accuracy of determination of I decreases with increase of P and the consequent greater extrapolation necessitated.

Clay[23] and Clay and van Tijn[24] applied Zanstra's deductions to their measurements of gamma-ray and cosmic-ray ionization currents in various gases at high pressures, and found the linear relation between $1/i$ and $f(x)$ obeyed for large collecting field intensities. Clay[23] found the "saturation" current to increase linearly with pressure at different rates in different pressure ranges, and explained the situation in terms of secondary radiations produced in the gas and in the walls of the vessel.

Bowen and Cox[25] have criticized the work of

[23] J. Clay, Physica **2**, 811 (1935).
[24] J. Clay and M. A. van Tijn, Physica **2**, 825 (1935).
[25] I. S. Bowen and E. F. Cox, Phys. Rev. **51**, 232 (1937).

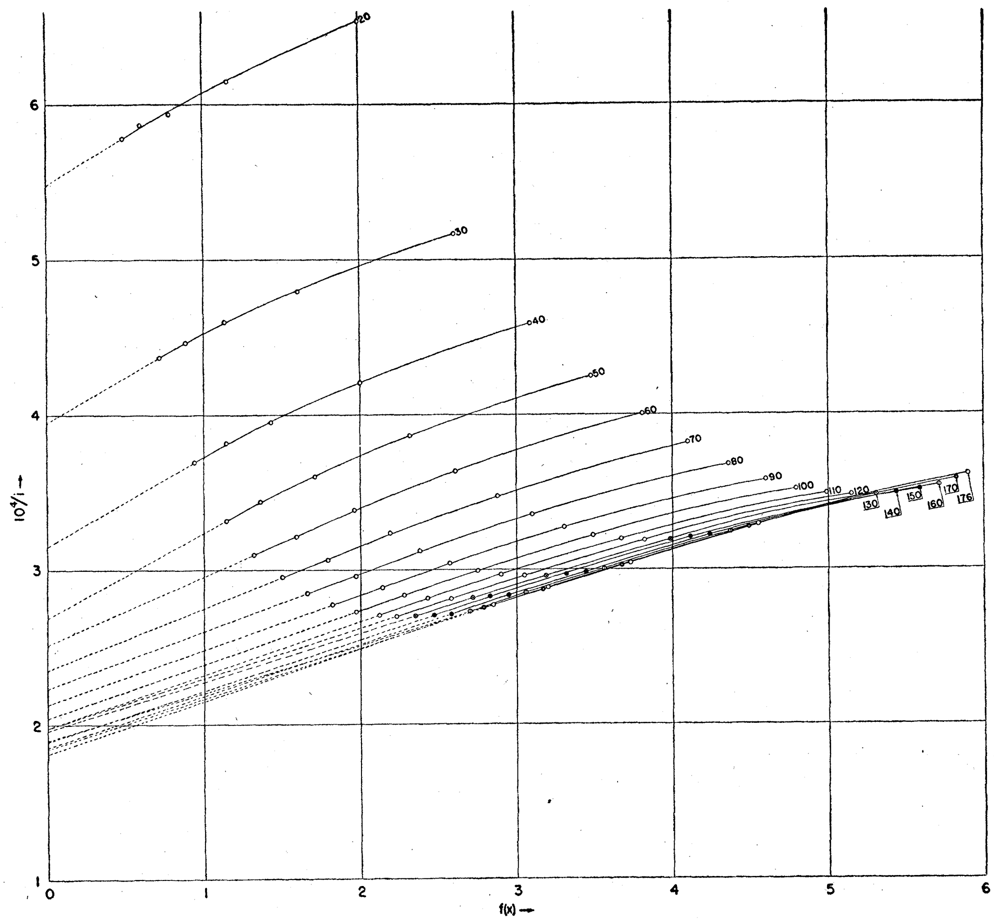

FIG. 2. Jaffé-Zanstra function curves corresponding to our measurements with ring-source. Air at 27°C. Ring-source of gamma-radiation beneath chamber. Numbers at upper ends of curves represent specific gravity relative to air at N.T.P.

Zanstra and Clay and van Tijn. Applying Zanstra's equation to their own measurements, they obtained a curve concave toward the $f(x)$ axis rather than a straight line. They emphasized the necessity of very low ion densities such as employed in their own measurements, in order to assure freedom from "general" or "volume" recombination.

Clay[26] then showed that "volume" recombination could not have affected his measurements, and pointed out that the collecting fields employed by Bowen and Cox were too weak to provide a proper test of the Zanstra equation. Clay and van Tijn[27] have provided striking evidence in favor of this method of analysis by deducing Eve's constant from measurements of gamma-ray ionization currents in air at pressures extending to 147 atmospheres.

Our recent measurements[28] of gamma-ray ionization currents in air provide a desirable test of this theory which presumes to evaluate actual "saturation" currents (or rates of *production* of ions?) in gases at high pressures. We employed the same low ion densities advocated by Bowen and Cox, and the range of collecting field intensities and pressures found satisfactory by Clay and van Tijn.

In Fig. 1 are the curves obtained by substituting in Zanstra's equation our experimental values observed with the central source of

[26] J. Clay, Phys. Rev. **52**, 143 (1937).

[27] J. Clay and M. A. van Tijn, Physica **4**, 648 (1937).

[28] J. W. Broxon and G. T. Merideth, Phys. Rev. **54**, 1 (1938).

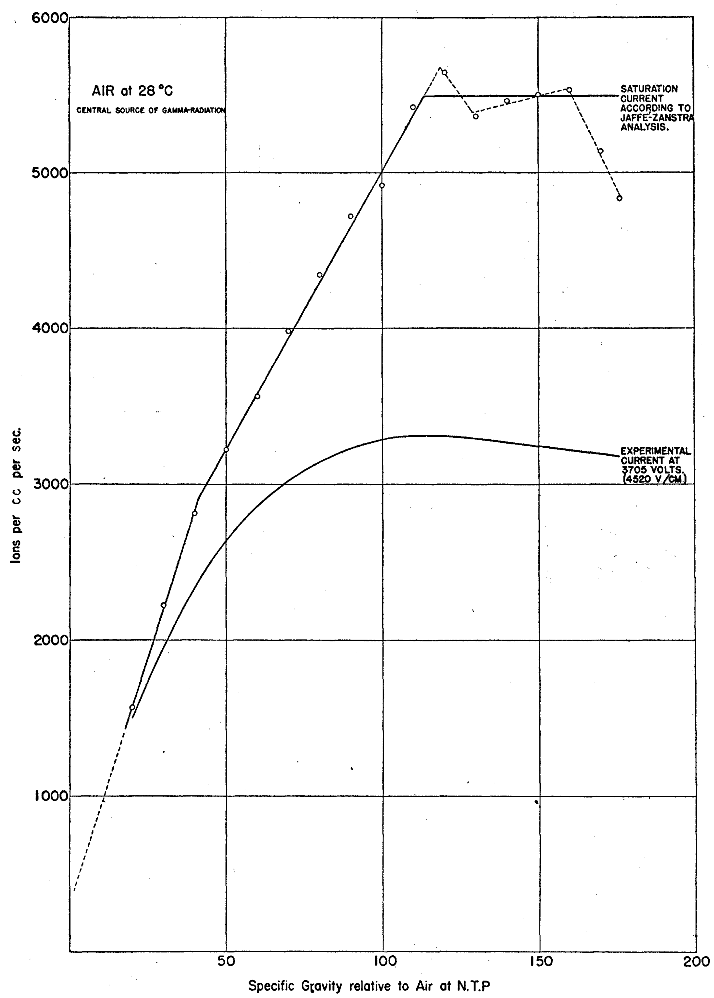

FIG. 3. "Saturation" current curve corresponding to our observations with central source, together with experimental curve obtained with strongest collecting field.

radiation. σ, the specific gravity of the air relative to air at 0°C and standard atmospheric pressure, was substituted for p in the expression for x. E was obtained by dividing the voltage across the chamber by 0.82 cm, as was done in obtaining the gradients of Figs. 7 and 8 of the preceding paper. Values of i for a particular density of air were read from the family of curves of Fig. 3 of the preceding paper, the smoothed-out curves being regarded as more reliable than individual observations. Such series of values were determined at pressures corresponding to intervals of 10 in the specific gravity, and a Zanstra curve constructed for each set. Only values corresponding to the five highest gradients we employed, within the range suggested by Clay, are included in the diagram.

The curves of Fig. 2 correspond precisely to those of Fig. 1, but are obtained from the ring-source curves of Fig. 5 of the preceding paper.

It will be observed that all the curves of Figs. 1 and 2 are somewhat concave toward the $f(x)$ axis, in the sense noted by Bowen and Cox[25] but in many cases the curvature is very slight. The curvature was found much more pronounced in the same sense in the region of low field intensities, not shown. Since the curves were not straight, extrapolations were attempted by producing tangents to the curves at their high gradient termini where the ideal should have been most nearly approached.

Reciprocals of the intercepts on the $1/i$ axis of the extrapolations of the curves in Figs. 1 and 2 are plotted as "saturation" currents in

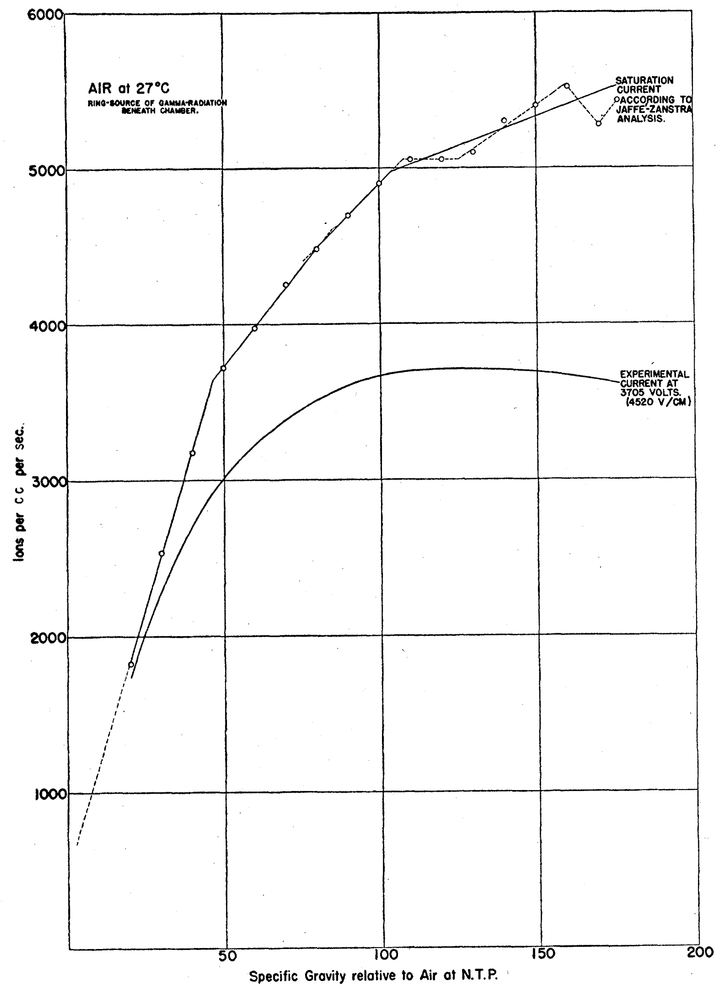

FIG. 4. "Saturation" current curve corresponding to our observations with ring-source, together with experimental curve obtained with strongest collecting field.

Figs. 3 and 4, respectively, against the specific gravity of the air. As the theory predicts, the "saturation" curves in each case lie above the experimental curves obtained with the highest field intensity employed, throughout the pressure range. As Clay and his associates found, the "saturation" current curves are made up of straight line segments, at least at the lower pressures. As in their curves, extrapolations of the low pressure segments to zero density cut the current axes above the origins. The first breaks in the curves occur at about $\sigma = 41$ and $\sigma = 46$, or pressures of about 45 and 50 atmospheres at 28°C. They correspond to breaks in the curves of Clay and van Tijn at about 40 atmospheres, the slope being less steep above the break than below in each case. In this connection it may be noted that their electrode spacing was slightly less than ours.

The "saturation" currents show a new feature, however, in that a second break occurs at about $\sigma = 114$ or 126 atmospheres in Fig. 3, and at a somewhat lower pressure in Fig. 4. A very slight break is indicated at $\sigma = 80$ or 87 atmospheres, also, in the second case. Whereas in Fig. 4, corresponding to the ring source, the "saturation" current continues to increase with pressure at a further decreased rate above the high pressure break, the central-source curve of Fig. 3 indicates the attainment of a maximum "saturation" current or even a possible decrease, although it should be remembered that the extent of extrapolation (with accompanying decrease of accuracy) increases rapidly with pressure. In this connection Zanstra's[22] calculations from Erikson's data, represented in Fig. 5, are interesting in that they also show a high-pressure break in the "saturation" current curve, which may be significant in spite of the wide limits of error indicated. However, Clay and van Tijn[27] attained pressures of 147 atmospheres without the appearance of the additional break.

If Clay's explanation of the breaks in terms of the absorption of "wall" radiations is to be applied, it would seem to be necessary to invoke a dual type of "wall" radiation to explain our new breaks in the neighborhood of 130 atmospheres. The possibly horizontal portion of the "saturation" curve of Fig. 3 may call to mind attempts by Broxon to explain the pressure effect entirely in terms of "wall" radiation.

Relative to the similarities of the experimental and theoretical curves corresponding to the two sources of radiation, the diversity of directions of excited beta-rays relative to exciting gamma-rays should be borne in mind, together with the fact brought out by Jaffé that except in case of very accurate collimation in the direction of the collecting field, there is little dependence of experimental current upon angle between columns and field, even in the case of alpha-rays.

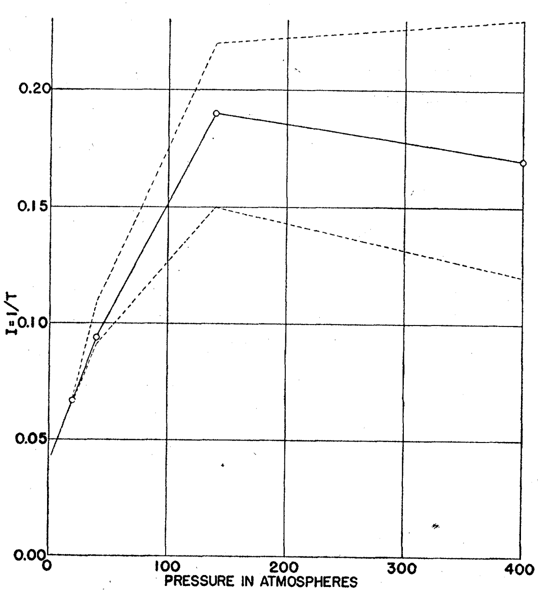

Fig. 5. "Saturation" current curve corresponding to Erikson's measurements, as calculated by Zanstra. Dotted lines represent limits of error estimated by Zanstra.

In Fig. 6 are given the Zanstra curves for the currents obtained upon collection of negative and positive ions, respectively, at the three pressures at which both currents are shown in Fig. 6 of the preceding paper. Intercept values for ions of the two signs at a particular pressure do not appear to be identical as in the case of Clay and van Tijn.[24]

We have also measured ionization currents produced in air at 100 atmospheres by a larger supply of radium located about 65 cm beneath the ionization chamber. The negative-ion currents measured at the five highest gradients were almost exactly 9.7 times as great as the corresponding currents obtained with the central source, throughout this range of gradients. Again negative-ion currents were larger than the positives. The Zanstra curves for both negatives and positives (not shown) had the small curvature toward the $f(x)$ axis, but in this instance the intercept values for the extrapolated curves might well be considered identical.

By way of criticism of the theory applied herein and of other theories, it should be emphasized that assumptions relative to the ionic "coefficients" at high pressures have been necessary in a region where little and in some instances no experimental work has been done. Moreover, because of the complexity of the problem, over-simplification is likely. It is interesting that in order to obtain an approximate solution to his differential equation, Jaffé[12] made the simplifying assumption that the radial distribution of ions in a column is determined entirely by diffusion and not at all by recombination, the latter supposedly affecting only the gross ion content of the widening column. As Jaffé points out, this amounts to assuming that the rate of recombination is not proportional to the square of the ion density at a point in the column, or to assuming that the "coefficient of recombination" is variable, being least at the axis where the ion density is greatest, and increasing exponentially with the square of the radial distance. Thus at a distance a little less than the mean distance of the ions from the axis the "coefficient of recombination" is presumed to be twice as great as at the axis, and at a distance twice as great as the mean radial distance

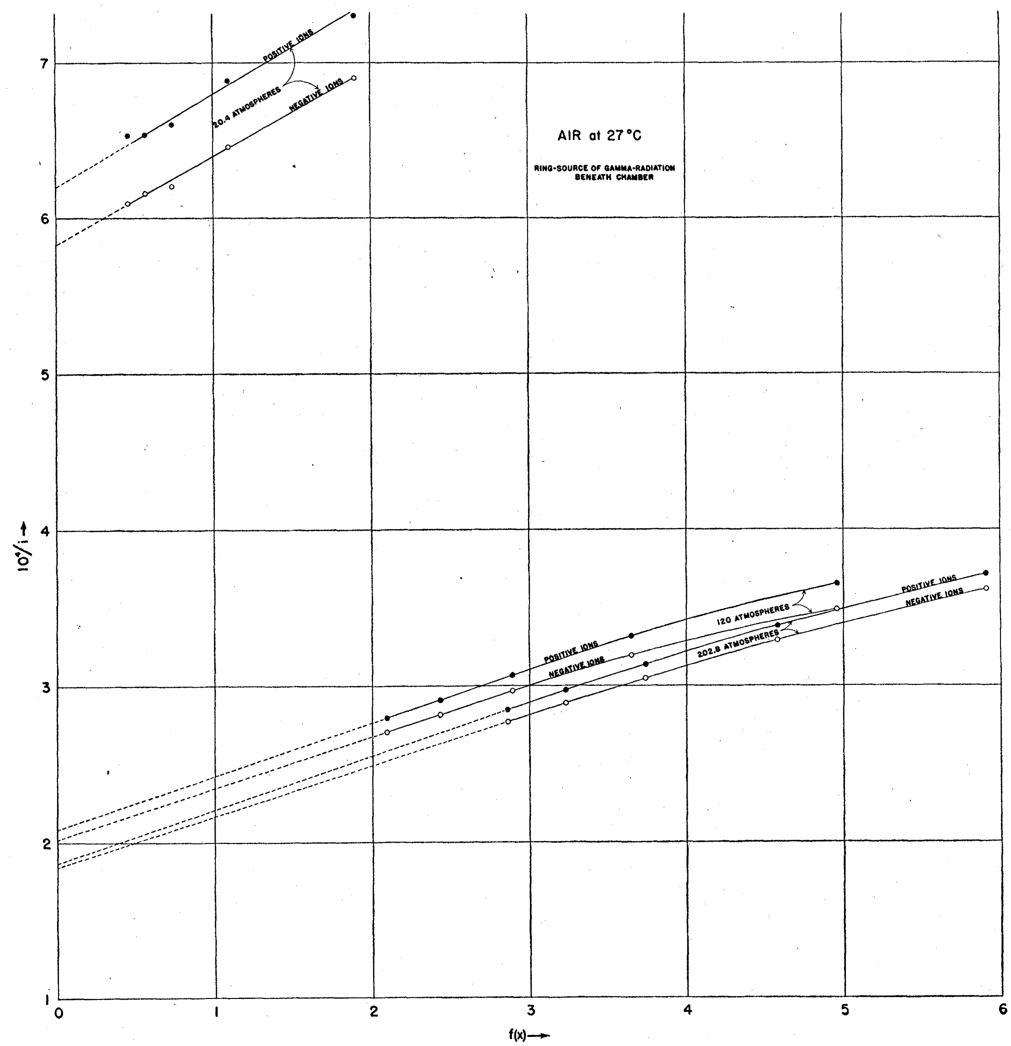

FIG. 6. Jaffé-Zanstra function curves corresponding to our measurements of "negative" and "positive" ion currents at three pressures.

of the ions, the "coefficient of recombination" is about 23 times as great as at the axis. This is inherent in all theories which begin with Jaffé's final equation.

It appears practically universally to be assumed that for a given intensity of a penetrating radiation passing through a specified volume of gas, the rate of production of ions and hence the theoretical "saturation" current which may or may not be capable of measurement, must be directly proportional to the amount of the gas present in that volume. It may possibly be worth mentioning again[2] that if it is conceivable that radiation energy may eventually, through the agency of subsidiary radiations or otherwise, be dissipated in any manner other than by the production of ions, then this universal assumption may not be justified.