THE PHYSICAL REVIEW A Journal of Experimental and Theoretical Physics

Vol. 43, No. 9 MAY 1, 1933 SECOND SERIES

Use of Argon in the Ionization Method of Measuring Cosmic Rays and Gamma-Rays

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The saturation-voltage, current characteristics of argon were studied at two pressures of argon and at four widely different ionization intensities produced by gamma-rays. Pressure-ionization curves for argon and for air were made. A comparison of the argon curve with the nitrogen one shows that argon is about twice as sensitive for use in

rays. Some transient ionization effects are described. A comparison of the intensity of cosmic rays to gamma-rays was made and this ratio was found to be lower for argon than for air in the ionization chamber at the same pressure.

ionization chambers for studying gamma-rays and cosmic

I. INTRODUCTION

HIS paper is chiefly concerned with the study of argon^{1, 2} with reference to its ionization at high pressure by cosmic rays and by gamma-rays from radium. Since argon was to be used by Compton in an extensive geographical survey of cosmic rays, it was important to know its behavior with reference to the above rays. Some rather striking results were obtained with this gas; results that show its superiority over the gases air and nitrogen that had already been used by various investigators of cosmic radiation.

Some of the earliest work on the ionization of gases at high pressures was done by Erikson,³ who investigated air to a pressure of 400 atmospheres, and carbon dioxide to the pressure of liquefaction. Unfortunately my present data on air are comparable with his only for the stronger currents. I deduce from his paper that the ionization he used was about 8 times the greatest

¹ A. H. Compton and J. J. Hopfield, Phys. Rev. 41, 593L (1932).

used by me. His data were carried over a much wider range of pressures and potential gradients than any here recorded. Indeed from his work one can predict the ultimate form of the present curves and it is interesting to note to what extent these forms are developed.

Some of the results of Erikson are: (1) The saturation curves for air rise steeply for the first 50 volts and then swing over and continue to rise gradually for the rest of their course, never becoming horizontal, however, even at gradients of more than 1000 volts per cm. (2) The ionization-pressure curves for air reach flat maxima and descend with increasing pressure even in the case of the greatest potential gradient.

Some recent work of Bowen⁴ on the ionization of air by gamma-rays carried over about the same range of pressure as the present work is valuable for comparison with the present data.

II. SATURATION CURVES FOR AIR AND FOR ARGoN AT HIGH PREssUREs

The saturation data for argon were obtained in the following manner: The radium sample,

« I. S. Bowen, Phys. Rev. 41, 24 (1932).

² J. J. Hopfield, Phys. Rev. 41, 904A (1932).

³ H. A. Erikson, Phys. Rev. 27, 473 (1908).

containing 0.971 mg of radium surrounded by a centimeter of lead, was placed at various distances from the center of a spherical bomb, 10 cm in diameter, which was the ionization chamber.⁵ The bomb was made of steel and served as one electrode of the ionization chamber and a central wire connected to the needle of a Lindemann electrometer served as the other electrode. The "saturation" curves were made with voltages ranging from 4 to 200 volts applied between needle and bomb. This voltage was read from a high-resistance voltmeter and the ionization current was read from the electrometer. The constants of the apparatus were measured in order to reduce this current to ions per cc per sec.

The curves are shown at four ionization intensities (namely, with the radium at 30 cm, 1 m, 3 m, and infinity from the center of the bomb) for each of the two pressures (see Figs. 1, 2, 3, 4). Some of the more obvious things that are shown by these curves may be noted. Curves 1A, 18, and 2A are far from saturation values even at the highest voltages used. This has already been shown by Erikson³ for saturation curves in air. Curve 2B also has an upward trend. On the other hand the data of curves 38 and 4A, 8 show much less upward trend indicating that saturation was more nearly attained.

The data taken at the higher current intensities (Figs. 1 and 2) are much smoother than the data for smaller intensities (Figs. 3 and 4). This is perhaps due to the probability fluctuations of the weak radiation used for the latter data, and to fluctuations in the potential of the bomb due to faulty contacts, etc., which cause a corresponding induced potential on the electrometer. This source of error is more effective for the higher sensitivity of the electrometer used with the smaller currents.

The saturation curve for air 2C is much more nearly horizontal than 28. This shows that it is easier to produce this form of saturation in air than in argon at about the same pressure.

Fig. 5 shows that any two of the above curves are not convertible into one another by

a constant factor. This indicates quantitatively that the percentage of saturation at a given applied voltage is not constant with changing intensity of ionization but is a function of that intensity. This is at variance with some of the data of Bowen⁴ taken in air. It is seen that the two smallest currents are nearest to coincidence. The other two show no semblance of coincidence. In Bowen's case the superposition of curves was good for large potential gradients and not so good for smaller ones. These data would correspond to his test with smaller gradients (see paragraph on potential distribution in the ionization chamber) and hence are to be compared only in such cases.

To account for the general shape of the curves and for their behavior when compared with one another one can picture the physical process to be the following: the gas in the bomb is traversed by many thin, nearly parallel tracks of intense ionization due to secondary swift electrons ejected by the gamma-rays, A few of the ions are pulled away from these tracks by the applied electric field and commingle in the bomb while they drift toward the electrodes. Recombination takes place in both the region of the parent tracks and in the intervening region. However, with a given distribution of potential gradient, the percentage of the ions lost by recombination in the parent tracks (since the tracks are too far apart to inhuence one another) is constant, i.e., independent of the number of tracks, or what amounts to the same thing, of the intensity of ionization. The ionization current, which is due to the ions outside the tracks, would then be proportional to the number of tracks and any two of the above curves should be exactly superposable by a constant factor. The fact that they are not, but drop lower for the higher intensities indicates that ions are lost by recombination in the interspaces, and moreover, the number thus lost (proportional decrease in current) is not proportional to the number of tracks but to some higher power or function of that number. The ordinary n^2 relation for recombination is expected to hold in this region.

This result, as already mentioned, seems to be at variance with the work of Bowen, who, however, experimented with small ionization

 $* A$ complete description of the apparatus will be published shortly in the Review of Scientific Instruments. See also A. H, Compton, Phys. Rev. 43, 387 (1933).

FIG. 1. Saturation current in argon, radium distance 30 cm.

FIG. 2. Saturation current in argon and air, radium at 1 m.

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ARGON, 31 A.

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FIG. 4. Saturation current in argon, radium at infinity.

Ftc. 5. Argon at 71 atm. Figs. 1-4 reduced to a common scale.

intensities. Also it does not agree with the conclusion of Compton' and others that recombination takes place only in the tracks of ionization.

The fact of columnar ionization lends itself well to explaining qualitatively the form of the ionization curves at different intensities as well as to pointing out some of the characteristics of the I—P curves discussed in a subsequent paragraph. The saturation curve at high pressure can be considered as farmed by adding two ideal curves, the one rising rapidly with the voltage and bending over sharply to the horizontal at the saturation value even at comparatively low voltage, and the other starting from the origin and rising slowly with the slope of the actual ionization curve at high field intensities. The resultant curve would be the sum of these. The first part of the experimental curve (showing the rapid rise at low voltage) is chiefly due to the loose ions in the interspaces; the slowly

rising second part is due to ions pulled from the tracks.

The design of the ionization chamber affects the distribution of the applied potential. By making the following simplifying assumptions: (a) that the central wire is very thin in comparison with the radius of the sphere; (b) that the charge is distributed uniformly over the surface of the sphere; (c) that the linear density of the charge on the wire is constant, it can be shown that the distribution of potential in the equatorial plane of the bomb normal to the central electrode has the following approximate values: the drop in potential for the outside centimeter of distance is 6 volts, if 144 volts is applied to the chambers; over the next centimeter 8.7 volts; the third centimeter 13.3; the fourth 24.0 and over the last centimeter 92 volts. The calculation shows that most of the potential drop occurs in a small volume near the center of the bomb and that the remainder of the volume of the bomb has a small potential gradient.

A. H. Compton, R. D. Bennett and J. C. Stearns, Phys. Rev. 39, 873 (1932).

III. IONIZATION-PRESSURE CURVES FOR AIR AND FOR ARGON

The ionization-pressure data for air and for argon were taken in the following manner: The bomb of the apparatus was pumped out well and then filled with air or with argon to the highest pressure. The gas was allowed to stand unused for about a day after filling. When ionization current readings were taken, the radium was at a distance of 100 cm from the center of the sphere, and the sphere was unshielded. A potential of 144 volts was placed on the sphere and the needle of the electrometer was at first connected to earth, the grounding key opened to allow the charge to accumulate on the needle, and the ionization current was read. The temperature of the gas was taken from a thermometer placed against the bomb. The pressures were read from a gauge connected to the bomb. The readings of current were taken first with $+144$ volts on the sphere and then with —¹⁴⁴ volts on it, in order to reduce the effects of small electrical leaks. Some gas was then allowed to leak out slowly, the cock shut off, and after waiting an hour or more, readings were taken at the new pressure. The range of pressure covered for air was from 105 to atmospheres, and for argon from 95 to 1 atmospheres. These data are plotted in Fig. 6, curves A and B. Curve C for nitrogen was taken from the work on ionization of nitrogen by Broxon.⁷

In explaining the form of these curves, Broxon⁸ using simple assumptions and an ingenious analysis showed that the I—P curve for air fits a cubic equation in which the parameters are all determined from the data. However, his theory' yielded an absorption coefficient for cosmic rays which he concedes is too large. Compton, Bennett and Stearns, on the other hand, find the curves fit equally well when derived on other assumptions.

The I—P function of neither of the above authors has a term which would allow a decrease of ionization with pressure when higher pressures are reached. However, this decrease has been experimentally observed by Erikson' (not by Broxon, δ however) so that the I-P relation is even more complicated than those derived by the above authors.

Obviously the intensity of ionization in a given gas is primarily dependent upon the source of ionization. If the source of ionization, or the gas in the ionization chamber, or the shielding is not changed and the ionizing radiation traverses the entire chamber with practically no diminution of intensity then the ionization per atmosphere at all pressures should be the same, and the I—P curve should be a straight line through the origin. Since this is not so, it is obvious that some of the above assumptions are not warranted, or that some other factor has to be considered. This additional factor is the recombination of ions in the chamber. This factor probably accounts for the departure of the curves from straight lines. If the slope of these curves at zero pressure and with saturation voltage could be determined, then the remainder of the experimental curve would be a measure of the incompleteness of saturation.

Practically all observers agree as to the form of the curves. In no case has a saturation current been produced. To show how the I—P curves fall short of saturation one could measure the slope of the curves at zero pressure, and draw a straight line with this slope from the origin. Theoretically these slopes for air, nitrogen and argon, when using gamma-rays, are in the ratios of their respective number of electrons per
molecule.¹⁰ molecule.

This ratio for argon and air is 18:14.4. This should represent the ratio of the slopes of the experimental curves at the origin. Since this slope could not easily be measured in these experiments, the slopes of the chords connecting the point at one atmosphere pressure with the origin was determined instead. The slope of the argon curve is 3.65×10^2 ions/cc/sec. per atmosphere, and that of air is 2.28×10^2 , and the ratio of these is 18:11.²⁵ which is much greater than the theoretical value given above. It is evident then that neither air nor argon shows saturation values, and that the loss of ions by recombination in argon is much less than in air. A comparative study of the gases can be made

⁷ J. W. Broxon, Phys. Rev. 38, 1704 (1932).

J. W. Broxon, Phys. Rev. 37, ¹³²⁰ (1931).

⁹ J. W. Broxon, Phys. Rev. 42, 321 (1932) (abandons the above theory).

¹⁰ Cf. Rutherford, Radioactive Substances and Their Radiations, p. 216, 1913.

1	$\overline{2}$	3	$\overline{4}$	5	6		8	9	10	11	12
Þ, (Atm.)	I , argon ions cm^{-3} sec. ⁻¹	I/p , argon	I, argon (theor.)	Satura- tion	percent I , air ions argon $\rm cm^{-3}$ sec. ⁻¹	I/p , air	I , air (theor.)	Satura- tion air	percent $\frac{I, \text{argon}}{I}$		<i>I</i> , argon I , air nitrogen I , nitrogen
10 20 30 40 50 60 70 80 90 100 110 120 130 140 150 160	3.75×10^{3} 3.75×10^{2} 6.90 9.26 11.20 12.84 14.19 15.30 16.23 17.03 17.73 $*(18.30)$ (18.70) (19.00) (19.14) (19.28) (19.30)	3.45 3.09 2.80 2.57 2.36 2.19 2.03 1.89 1.77	3.61×10^{3} 7.22 10.83 14.44 18.05 21.66 25.37 28.88 32.49 36.10	100.4 95.5 85.5 77.5 71.1 65.5 60.5 56.1 54.1 49.1	1.80×10^{3} 2.88 3.55 4.10 4.51 4.80 5.01 5.21 5.35 5.49	1.80×10^{2} 1.44 1.18 1.02 0.90 0.80 0.71 0.65 0.59 0.549	2.89×10^{3} 5.78 8.66 11.56 14.44 17.34 20.2 23.1 26.0 28.9	62.3 49.9 41.0 35.5 31.2 27.7 24.8 22.6 20.6 19.0	2.08 2.40 2.61 2.73 2.84 2.95 3.05 3.12 3.18 3.22	2.1×10^{3} 3.50 4.55 5.44 6.18 6.75 7.19 7.56 7.90 8.22 8.46 8.66 8.80 8.86 8.92 8.94	1.87 1.97 2.04 2.06 2.08 2.10 2.13 2.14 2.16 2.16

TABLE I. Comparative ionisations in argon, air and nitrogen.

with the slope of the argon I–P curve as a basis. The straight line A' representing this is drawn. The straight line for air B' is drawn with a slope 14.4/18 times this. Table I gives the results of this comparative study.

These curves and the table indicate the following things: (1) Curve A being more nearly linear than that of air B shows that a given voltage applied to the chamber produces a greater degree of saturation in argon than in air under the ionization of gamma-rays. (2) The greater height of the argon curve above either the air or nitrogen curves shows how much greater sensitivity can be expected from the use of argon in high pressure ionization chambers. (3) It is remarkable that the curves for argon and for nitrogen for the most part are similar (column 12). This makes it possible to extrapolate the argon curve by using the factor 2.16 to 150 atmospheres on the basis of Broxon's' data. These extrapolated values in the table are in parentheses. (4) The curves for air and argon are not similar (column 10).

The above data are offered with qualifications, namely: it was shown in a former section devoted to saturation current data that the same voltage does not produce the same degree of saturation at different pressures. Furthermore, although reasonable care was taken in filling the bomb with argon, nevertheless it was rated by the

producers as containing 0.15 percent oxygen and 2.2 percent nitrogen as the chief impurities. The curve for pure argon should be slightly higher than this one.

IV. CURRENT LAG ON FILLING IONIZATION BOMB

The bomb was newly filled with argon to 70.5 atmospheres pressure and the readings of ionization current with the radium at 1 meter were taken at intervals. Fig. 7 shows the results.

FIG. 7. Current change after filling the ionization chamber.

The current at first rises rapidly with the time. After three hours it is only 13.8×10^3 ions/cc/sec., whereas the end-current for this pressure is

^{*}The values in parentheses represent an extrapolation of the argon values based on Rroxon's data for nitrogen $f = 2.16$ was used.

 14.3×10^{3} ions/cc/sec. It takes about a day for the current to reach a constant value. It is for this reason that the data for the I—P curve with argon and with air were taken in the direction of decreasing pressure. The rise of current with time agrees with the results of Steinke and time agrees with the results of Steinke and
Schindler,¹¹ in CO₂, but is the reverse of the result of Broxon' in air, who found a decrease of current with time.

Evidently the cause of this lowered current is the dust from the walls and connection tubes and stopcock grease vapors tom loose by the rapidly moving gas. Until these particles have had time to settle out or move to the walls of the chamber they act as centers for clustering of ions where recombination of ions may readily take place. They also form ions of low mobility. Compton in a private communication states that a similar effect can be observed when the apparatus is first set up after being knocked about in transportation.

V. RECOMBINATION OF IONS

Recombination of ions in air and in argon at low pressures have been extensively studied. (Erikson.³) Argon behaves in this respect like air. Figs. 8 and 9 show these data. The manner of obtaining them is as follows: the radium capsule was placed in contact with the unshielded bomb and was allowed to rest there until a large concentration of ions developed in the bomb, the field inside of the bomb being zero. The radium was then taken to a distance of two meters and placed behind 5 cm of lead. Since no 6eld was applied at first, the concentration of the ions diminished chiefly by recombination. At a given time after removal of the radium, $10, 20, 30,$ etc., seconds, the 144 volt field was suddenly switched on and a fraction of a second later the key was opened so that the current to the electrometer could be measured. The data obtained in this manner are represented by the series of short curves of Figs. 8 and 9.

The manner of plotting these is best explained by taking as an example the data for the "40 seconds curve." The successive deflection readings on closing the key were 39, 49, 54, 59, 64. The average times elapsed between these respective deflection readings were 4.6, 4.45, 5.63 and 9.5 seconds. The sensitivity was 0.0615 volts/division. The successive currents calculated from these data are: 2.95, 1.53, 1.205 and 0.806×10^4 ions/cc/sec. The middle point of each of the above time intervals was chosen, so that the times to be plotted against the above currents are 42.3, 46.8, 51.8 and 59.4 seconds. The 40 seconds that has been added in each case is arbitrary. In like manner 30 seconds were added to the "30 seconds" readings, etc., so that these small curves are properly dispersed on a time scale. The heavy curves simply join like points of time elapsed after current readings were started on these smaller curves. The 5, 10 and 15 seconds points respectively are joined to make the larger graphs of curve 8, and the 10 seconds points to form the one of curve 9.

The striking thing shown by Fig. 9 is that this ourve does not dip down to the line of constant current caused by ionization of the radium in its sheltered position but remains definitely above it. Take for example the last set of data which was taken after the ions had recombined for 10 minutes in zero field. These data can also be obtained from the reverse direction, i.e., starting with practically zero concentration of ions and allowing the radium to remain in its shelter for 20 or 30 minutes in the absence of a field. It is found that in this case the ionic concentration builds $u\phi$ to the values measured for the "10 minute curve. " Thus, the current is about 10 times as great as the steady one with the radium in this position. This would seem to be a new method for temporary amplification of small currents. This phenomenon is obviously related to the long life of ions in argon.

VI. MoBILITY QF IQNs IN ARGQN

Fig. 10 shows clearly that it takes some of the ions at least 50 seconds to traverse the ionization chamber, i.e., some of them move not faster than 1 mm/sec. These data were taken by first placing the radium against the ionization chamber but with the 144 volt field switched on, and the electrometer key closed. The current readings were taken by opening the key after the elapsed intervals of time indicated in the data.

¹¹ E. G. Steinke and Schindler, Naturwiss. 20, 15 (1932).

FIG. 9. Equilibrium concentration of ions in zero field.

FIG. 10. Speed of the ions in the ionization chamber.

VII. INVERSE SQUARE LAW

After seeing what difficulty one has in measuring the real value of intensity of ionization or even of intensity ratios, one might think that the inverse square law of intensity could be used as an instrument for ironing out these difficulties. The study of this law simply showed up the more formidable difhculties already described in this paper. Moreover an additional variable was introduced, namely, the scattering of the rays from material near the apparatus, so that one cannot hope with these data alone to solve the problem which now involves intensity of radiation, lack of saturation voltage for the currents measured and the new factor of scattering bodies.

If one should make these experiments at low pressure of argon, where saturation voltages are easily possible, then any discrepancy in the inverse square law could be attributed to scattered radiation, and the percentage of this could be determined. If then a second experiment were made under the same conditions except with high pressure in the bomb, any additional discrepancy could be ascribed to nonsaturation voltage and the degree of this determined. Unfortunately, this could not be done in the present case. Compton suggested in a private communication that the effect of scattering could be largely eliminated by surrounding the bomb with lead. This has not been tried.

Many data were taken at several distances between 3 meters and 60 cm, but all values of n , the inverse square law exponent, were less than 2, and the greatest discrepancies occurred from the data taken at the smaller distances. The effect of scattering was easily observed by placing a plate of lead Hat on the Hoor between the radium and the ionization chamber. The ionization current was increased perceptibly thereby.

VIII. MEAsURING CosMIc RAYs

In using the instrument for measuring cosmic rays relative to gamma-rays from radium, three measurements are necessary besides a determination of the constants of the instrument. The three measurements are: (1) Current due to cosmic rays plus local radiation through two shields of lead and one of bronze.¹ (2) Current due to gamma-rays from radium in a standard position plus cosmic rays plus local rays through two shields of lead and one of bronze. (3) Current due to cosmic rays plus local rays through one lead shield and the bronze shield.

The equations necessary to evaluate the intensity of cosmic radiation and the ratio of this intensity to the radium standard are the following:

$$
b = x_1 + x_2 + x_3 \quad (2) \qquad e = x_1/x_4 \quad (5) c = x_4 + x_5 \quad (3)
$$

where x_1 =current due to cosmic rays through shields 1, 2 and 3; x_2 = current due to local rays through shields 1, 2 and 3; x_3 =current due to gamma-rays from radium in standard position through shields 1, 2 and 3; x_4 =cosmic rays through shields 1 and 2 (one bronze and one lead shield); $x_5 =$ current due to local rays through shields 1 and 2; a , b and c are the electrometer currents already described in sentences numbered 1, 2 and 3 above; $d=0.94$ and $e=0.312$, the constants of the lead shields for cosmic rays and local rays, respectively. The value of d was deduced from measurements made by Compton. The value of e was determined for gamma-rays from radium. The application of the same constant to the local radiation seems to be reasonable,

The above five equations can be solved simultaneously for the five x's. However we are interested here only in x_1 , x_3 and their ratio. The solution gives:

$$
I_c = x_1 = [e/(e-d)](a-dc)
$$
 (6)

$$
I_r = x_3 = b - a \tag{7}
$$

$$
I_c/I_r = x_1/x_3 = [e/(e-d)](a-dc)/(b-a).
$$
 (8)

Eq. (8) can be transformed into Compton's equation for measuring the ratio of cosmic rays to gamma-rays.

Substituting the numerical values of d and e in Eqs. (6) and (8) gives the following result:

$$
x_1 = 1.497(a - 0.312c)
$$
 (6a)

$$
x_1/x_3 = 1.497(a - 0.312c)/(b - a). \tag{8a}
$$

Substituting the numerical values of a , b and c in Eqs. (6a), (7) and (8a) the values in Table II result. In this table, a, b, c and the x's are in the same arbitrary unit of current.

TABLE II.

No.	\boldsymbol{a}	b	\mathcal{C}		$I_c = x_1$ $I_r = x_3$ I_c/I_r		Place, Material
$\mathbf{1}$	0.1019	0.740	0.122		0.0956 0.6381	0.1499	Eckhart Hall Air. 36 atm.
2	.1338	.790	.1740	.1190	.6562	.1815	5430 Univ. Ave. Air. 36 atm.
3	.405	3.04	.511	.368	2.645	.1390	Eckhart Hall Argon, 73.6 atm.
	.256	2.21	.329	.230	1.954	.1176	Eckhart Hall Argon, 36 atm.

On comparing the data for the two pressures of argon, the x_1 's and the x_3 's, it is found that they do not change in the same ratio. Thus¹² $(3)x_3/(4)x_3 = 1.35$, whereas $(3)x_1/(4)x_1 = 1.60$. These ratios should be the same if a saturation current is obtained, or if the same degree of

 x^2 (3) x_3 , etc., indicates the x_3 of measurement No. 3 of Table II.

saturation is produced in both cases. The ratio is greater for the x_1 's than for the x_3 's, which is in accordance with the expectation that the increase of ionization current is more nearly proportioned to pressure for the weaker source of ionization. Figs. ¹—4, on the other hand, indicate that the ratio of current to pressure passes through a maximum for certain current values indicating a slight residual ionization, probably due to radioactive material in the chamber.⁹

If one compares the results of air and argon at the same pressure and the cosmic-ray intensity at the same place, namely Nos. 1 and 4, then $(4)x_1/(1)x_1 = 2.41$, and $(4)x_3/(1)x_3 = 3.06$. That is the ratio of increase in ionization when the monatomic gas is used in the ionization chamber is much greater for gamma-rays than for cosmic rays. A suggested explanation for this difference is that the air contains more radioactive contamination than argon. Another is in the possibility that the primary attack of cosmic rays is on atomic nuclei, while the gamma-rays act chiefly on extranuclear electrons. The interaction of cosmic rays and atomic nuclei has already been suggested by experiments of Millikan and been suggested by experiments of Millikan and
Anderson.¹³ Since argon has half as many nucle as air at the same pressure a smaller ratio (2.41) for cosmic rays than for gamma-rays (3.06) should be expected for it. It is seen that these data are quite in accord with this view. This, I believe, is the first direct comparison of the action of cosmic rays on monatomic and diatomic gases.

This work was supported by funds from the Carnegie Institution, and carried out at the University of Chicago under the direction of A. H. Compton.

¹³ R. A. Millikan and C. D. Anderson, Phys. Rev. 40, 1056A (1932}.