JANUARY 1 AND 15, 1943 PHYSICAL REVIEW VOLUME 63

Saturation Characteristics for Alpha-Particles in Purified Gases

E. D. KLEMA* AND H. H. BARSCHALL
Department of Physics, University of Kansas, Lawrence, Kansas

(Received October 29, 1942)

Saturation curves were taken in nitrogen, argon, and an argon-hydrogen mixture. The effect of purification on the saturation properties of argon and the argon-hydrogen mixture was studied. It was found that even unpurified tank argon and nitrogen show saturation at very much smaller field strengths than, for instance, air. Upon purification an appreciable lowering of the field necessary for saturation was observed.

INTRODUCTION

IN many studies of high speed particles it is necessary to fill ionization chambers with gases at high pressure, particularly in investigations of cosmic rays and in determinations of the energy of fast neutrons by use of measurements on the recoiling protons.[1] The use of gases at high pressure introduces several difficulties, the most important of which is caused by lack of saturation. Jaffé's theory[2] of saturation shows that the field strength necessary for saturation increases very rapidly with increasing gas pressure. Another difficulty arises when a linear amplifier is used to amplify ionization pulses. Ortner and Stetter[3] pointed out that for proper operation of the amplifier all the time constants in the coupling circuits between stages should be long compared to the collecting time of the ions in the chamber. In practice it is very inconvenient to make the time constants of an amplifier longer than about 1/50 sec. Thus extremely high voltages have to be used to collect the ions in a chamber filled to high pressures. Both of these difficulties could be overcome if it were possible to increase the mobility of the ions in the chamber.

It was first suggested by Ortner and Stetter[4] that one could take advantage of the high mobilities of the free electrons in very pure gases which do not form negative ions, such as hydrogen, nitrogen, and the inert gases. These authors carried out experiments in nitrogen at atmospheric pressure. The nitrogen was carefully purified before it was admitted to the chamber, and it was shown to give saturation at much lower voltages than air at the same pressure. More recently Jentschke and Prankl[5] used purified argon in experiments on the ionization produced by fission fragments, and obtained saturation for α-particles at field strengths as low as three volts/cm in argon at atmospheric pressure. In spite of these successes very few other workers have used purified gases. Probably the main reason for this was the belief that it would be almost impossible, without unusual precautions, to obtain sufficiently high purities to observe free electrons, since early work on the mobility of negative ions had shown that minute traces of electronegative gases decreased the mobility appreciably. It was the object of the present work to investigate the purity necessary to obtain improved saturation characteristics and to study methods of purification which could be used at high pressures and which would not require outgassing the chamber. The measurements of Helbig[6] on the saturation characteristics of unpurified argon seemed to indicate that it might not be necessary to attain such high purities as had been believed earlier.

EXPERIMENTAL METHOD

As a source of ionization the α-particles from polonium were used. The polonium was deposited in a thin layer on a silver foil. The α-particles were collimated by means of a Lucite sleeve 5 mm long. It had three cylindrical holes 2.2 mm in diameter, the axes of the cylinders being perpendicular to the silver foil. The α-particles leaving this sleeve

---

* Now at Palmer Physical Laboratory, Princeton University, Princeton, New Jersey.

[1] H. H. Barschall and M. H. Kanner, Phys. Rev. **58**, 590 (1940).

[2] G. Jaffé, Ann. d. Physik **42**, 303 (1913); **1**, 977 (1929).

[3] G. Ortner and G. Stetter, Wien. Ber. **142**, 485 (1933).

[4] G. Ortner and G. Stetter, Wien. Anz. **70**, 341 (1933); Physik. Zeits. **35**, 563 (1934).

[5] W. Jentschke and F. Prankl, Physik. Zeits. **40**, 706 (1939).

[6] E. Helbig, Zeits. f. Physik **116**, 444 (1940).

could have a maximum angle of 24° with respect to the axis of the cylinder; their average angle was about 8°.

To collect the ions an ionization chamber with parallel circular brass plates without guard rings was used. The plates were 5 cm in diameter and 2.9 cm apart. The leads to the plates were brought in through the top and bottom of the chamber and were waxed into Lucite insulators. The chamber proper was made of iron and sealed by means of a rubber gasket. The Lucite insulators were waxed into the chamber.

In all the experiments (unless otherwise stated) α-particles whose paths were nearly parallel to the electric field were investigated, since Jaffé's theory of columnar ionization shows that it is most difficult to obtain saturation in this case. To produce a beam of α-particles approximately parallel to the direction of the electric field, the radioactive source with its collimator was placed behind the top plate of the chamber, so that the α-particles could enter the collecting volume through a hole in the center of the plate. The hole in the plate was covered with an aluminum foil of about one-mm air equivalent stopping power to prevent ionization produced in the collimator from being collected.

The collecting voltages were obtained from "B" batteries up to 300 volts and from a transformer-rectifier power supply with three RC filter stages for potentials up to 4000 volts. The values of the collecting field were obtained by dividing the voltages applied to the chamber by the distance between the plates. The effective field may have been somewhat smaller, since no account was taken of the edge effect.

In all the measurements the ionization currents were measured by means of a vacuum tube electrometer, in which a type FP-54 tube was used in the balanced circuit described by DuBridge and Brown.[7] In order to check the sensitivity of the apparatus, a constant potential could be applied to the grid of the tube. This was done by applying a part of the voltage from a dry cell, obtained through use of a high resistance potential divider, to a fraction of the grid leak resistor.

Since the design of the ionization chamber excluded the possibility of outgassing it, a continuous method for purifying the gas in the chamber had to be used, i.e., a method which would remove impurities given off by the inside surfaces of the chamber. For the purification of argon, which was tried first, a method similar to that described by Jentschke and Prankl[5] was used. Pieces of $\frac{3}{8}''$ copper tubing led from the bottom and the top of the ionization chamber to the two ends of a cylindrical purification chamber made of brass. The axes of the two chambers were vertical. The purification chamber was sealed by means of a water-cooled rubber gasket. In order to remove electronegative gases the purification chamber could be filled with about 50 g of calcium turnings. The calcium was supported in the chamber on copper screens so that the gas could pass through the chamber readily. When the argon was to be purified, the purification chamber was maintained at a temperature of about 250°C. The temperature gradients in the system were then sufficient to make the argon circulate.

This method of purification was satisfactory for argon and would undoubtedly be equally applicable to other inert gases, but could not be used for hydrogen or nitrogen, since these gases react with the hot calcium. To remove any oxygen from hydrogen, or mixtures of hydrogen and argon or nitrogen, the calcium was replaced by platinized asbestos which acts as a catalyst in the combination of oxygen and hydrogen to form water vapor. The platinized asbestos was kept at 300°C both to increase the rate of the catalytic reaction and to provide the necessary temperature gradients. To remove the water vapor so formed, or any other traces of water vapor, a second purification chamber containing potassium hydroxide was placed between the chamber containing the platinized asbestos and the ionization chamber. The desiccating agent, in stick form, was supported by spirals of iron wire and cooled with circulating ice water.

RESULTS

Some preliminary experiments were carried out in *air* at one, two, and three atmos. pressure. The saturation curves under these conditions agree reasonably well with those obtained by Jaffé,[8]

---

[7] L. A. DuBridge and H. Brown, Rev. Sci. Inst. **4**, 532 (1933).

[8] G. Jaffé, Physik. Zeits. **30**, 849 (1929).

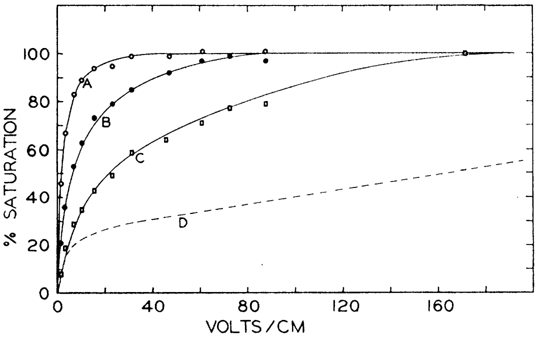

FIG. 1. Saturation curves obtained for α-particles parallel to the electric field in tank nitrogen. Curve A represents the results at one atmos., curve B at two atmos., curve C at three atmos. pressure. Curve D is the saturation curve in three atmos. of air.

although he gives slightly higher values for the ratio of ionization current to saturation current. The difference is undoubtedly due to the better collimation of the α-particles used by Jaffé. Our measurements showed about 90 percent saturation for air at two atmos. for a field strength of 1000 volts/cm. In this case the saturation value was obtained by extrapolation according to Jaffé's theory, and by using a graphical method similar to the one described by Zanstra.[9]

Further experiments were carried out in one, two, and three atmos. of *nitrogen*. The nitrogen[10] was oil pumped, and according to the manufacturer had a purity of 99.8 percent. Before the ionization chamber was filled for the measurements, it was flushed several times; but no further purification was attempted. Figure 1 shows the results obtained for nitrogen. The percentage saturation is plotted against the field strength in volts/cm. The saturation current for the curve at three atmos. was obtained from experimental points at high field strengths. For comparison, the saturation curve for air at three atmos. with the same experimental arrangement is also plotted. The figure shows the striking difference between the saturation characteristics of nitrogen and air. No change in the saturation curves for nitrogen was observed when the gas was allowed to enter the chamber through a trap maintained at the temperature of solid carbon dioxide.

The next gas investigated was *argon*.[10] According to the manufacturer, the argon had a purity of 99.6 percent. Preliminary measurements showed that the saturation curves for argon depended greatly on how many times the chamber had been flushed with argon and on how long the argon had been in the chamber before the measurements were started; these results indicate a strong dependence on purity. After argon had been purified for about one hour by allowing it to pass over hot calcium, the measurements became quite reproducible. The results in purified argon are shown in Fig. 2. Even at pressures as high as seven atmos., 70 volts/cm were sufficient to obtain saturation, while at one atmos. seven volts/cm gave saturation. In order to study the effect of impurities, air was added to five atmos. of argon. Figure 3 gives the results when two percent and five percent of air, respectively, were mixed with the argon. The effectiveness of the method of purification was tested by purifying for one hour five atmos. of argon to which two percent air had been added. The resulting saturation curve, which is also shown in Fig. 3, agrees with the curve obtained when the tank argon was purified.

Since in some experiments with ionization chambers it might be objectionable to have the gas circulate through the chamber, measurements were taken after the purification chamber was allowed to cool to room temperature. A change in the saturation characteristic was observed after a few hours. The saturation voltage in two atmospheres of argon increased by less than a

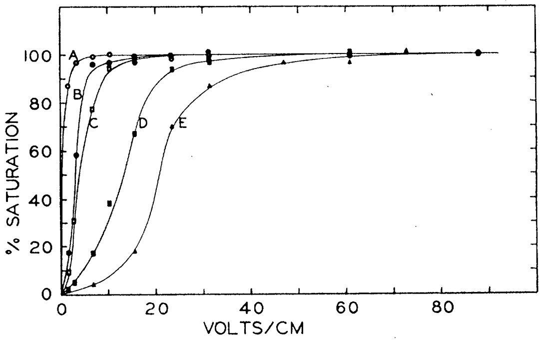

FIG. 2. Saturation curves obtained in argon which was purified by circulating it over hot calcium. The curves were taken at the following pressures: A, one atmos.; B, three atmos.; C, four atmos.; D, five atmos.; E, seven atmos.

---

[9] H. Zanstra, Physica 2, 817 (1935).

[10] Purchased from the Ohio Chemical and Manufacturing Company, Cleveland, Ohio.

factor of two in a period of twelve hours during which the purification chamber was not operating.

A *mixture of argon and hydrogen* seemed particularly worth investigating, since it might be useful for the measurement of the energies of fast neutrons which produce recoiling protons, the argon serving to stop the protons. According to the manufacturer, the hydrogen had a purity of 99.5 percent. A mixture of three atmos. of argon and one atmos. of hydrogen was used. The saturation curve for this mixture is shown in Fig. 4. In the same figure the curve is given which was obtained after the mixture had been purified for one hour by the use of hot platinized asbestos and potassium hydroxide. Although the effect of the purification is very noticeable, higher field strengths were necessary for saturation in this case than in pure argon. This may be due either to the ineffectiveness of KOH in removing water vapor or to the presence of electronegative gases other than oxygen and water vapor.

Measurements were also carried out for a mixture of nitrogen and hydrogen. The results were very similar to the ones obtained for the argon-hydrogen mixture.

Hydrogen by itself is known[8] to saturate relatively easily. The effect of purification by the use of platinized asbestos and KOH was investigated. It was found that the saturation characteristics were improved by purification, but not as much as in the case of argon.

When the polarity of the collecting voltage was reversed, no change in the voltage necessary for saturation was observed. In the case of purified argon, however, the shape of the lower part of the saturation curve depended on the polarity of the collecting voltage. When the top electrode, near which the ionization took place, was made positive the saturation curves rose more steeply in the initial portion than is shown in Fig. 2. This finding agrees with the observations of Ortner and Stetter[4] in purified nitrogen.

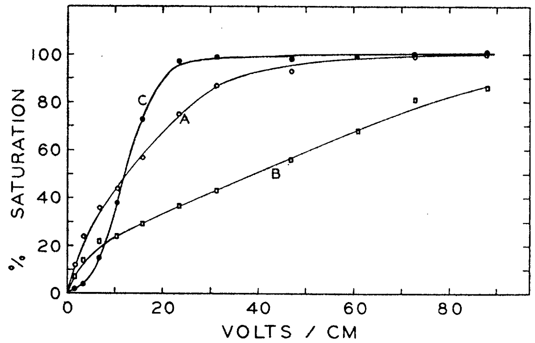

FIG. 3. Curve A: Saturation curve in five atmos. of argon plus two percent air. Curve B: five atmos. of argon plus five percent air. Curve C: Same as A, but purified by circulating mixture over hot calcium.

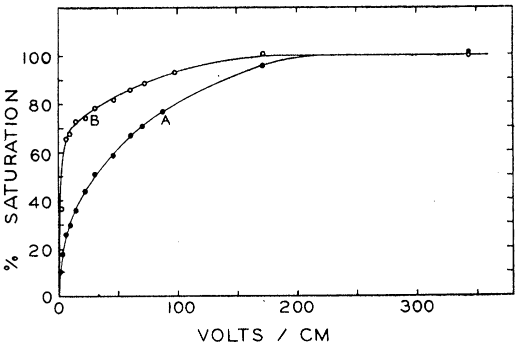

FIG. 4. Saturation curve for mixture of three atmos. of argon plus one atmos. of hydrogen. Curve A was taken in the mixture as obtained from the tanks. For Curve B the mixture was circulated over hot platinum asbestos and KOH.

An attempt was made to obtain a saturation curve while the radioactive source (without collimator) was placed half way between the two electrodes. For this measurement the surface of the source was parallel to the axis of the chamber and in a plane roughly tangential to the two electrodes. It was electrically at ground potential. Whereas with the source in this position ordinary saturation curves were obtained in air, hydrogen, and argon at one atmos., quite anomalous curves were found in purified argon at higher pressures. The curves showed a maximum at relatively low voltages and then dropped to a considerably smaller saturation value. The height of the maximum increased with both increasing purity of the gas and increasing pressure. Any change in the potential at which the source was kept altered the shape of the curve completely. The only explanation which seemed to cover all these phenomena was to assume that electrons formed by the α-particles in the neighborhood of the source were driven by the electric field against the source and its support. These electrons might then eject secondary electrons. This hypothesis was tested by painting the support of the source with colloidal graphite, which is known[11] to give little

---

[11] B. H. Porter, Rev. Sci. Inst. 7, 101 (1936).

secondary emission. It was found that the graphite reduced the height of the peak considerably. This bears out the assumption that the peak was due to secondary emission.

DISCUSSION

While Jaffé's theory of columnar ionization is well established, it seems likely that this theory is not applicable when the ionization is produced in the absence of electronegative gases. In this case the diffusion of the ions is probably so rapid that the columnar arrangement of the ions becomes less important than the general diffusion. Furthermore a comparison with theory is rendered difficult by the lack of sufficient experimental data on the recombination and diffusion coefficients in purified gases.

Since Warburg's[12] discovery of the effect of minute traces of electronegative gases on the conduction of electricity through gases, a number of authors[13] have investigated the effect of impurities on the mobility of negative ions. Franck's[14] experiments in argon showed that the mobility of the negative carriers in argon was decreased from 206.3 to 1.7 cm/sec./volt/cm when 1.2 percent oxygen was added. According to Haines[15] the mobility of the negative ions in nitrogen decreased from 158 to 88 cm/sec./volt/cm when 0.05 percent oxygen was added, and to 11 with 0.7 percent of oxygen.

The effect of the addition of two percent of air (0.4 percent of oxygen) to purified argon in the present measurements (see Fig. 3) seems to be relatively small, whereas measurements on ionic mobilities would lead one to expect a decrease of the mobility by a factor of 10 in this case.

A possible explanation for a smaller effect of impurities in measurements of saturation curves might be that a free electron would have to travel only a very short distance to leave the ion column. It then would be sufficiently separated from the positive ions to avoid recombination, even if it were to form a negative ion before it reached the electrode. In order to be observable in measurements of ion mobilities, the electron would have to travel considerably farther without encountering an electronegative molecule.

The occurrence and size of a peak in some of the saturation curves, which we believe was caused by secondary electron emission from the source, seemed to be more sensitive to the presence of impurities. This finding agrees with the above consideration, since to cause secondary emission the electron actually has to travel from its place of formation to the source.

The relatively small effect of impurities on the saturation characteristics may be understood from a different point of view according to Jaffé.[16] In the theory of columnar ionization[8] it is shown that the saturability depends only on the ratio of the diffusion coefficient (or mobility) to the recombination coefficient. If impurities are added to a very pure gas, both the mobility and the coefficient of recombination will decrease, but the ratio of the two will change less rapidly with added impurities than the mobility.

A striking feature of the saturation curves in purified argon at high pressure (see Fig. 2) is their increasing slope at low field strengths. An explanation of this unusual shape according to Jaffé[16] can be found in a calculation by J. J. Thomson[17] concerning the current between two parallel plates when the ionization is confined to a thin layer. This calculation is applicable to the present measurements, because at higher pressures the ionization is increasingly limited to a small region near the top plate. In pure argon the diffusion of the negative carriers is probably sufficiently rapid so that the ion columns spread into a layer. Thomson's calculation yields the result that in this case the ionization current is proportional to the square of the potential difference between the plates and directly proportional to the mobility of the ions moving away from the plate near which the ionization is produced. The curves shown in Fig. 2 may be approximated in their initial portion by a parabola according to Thomson's formula. The fact that the rise of the ionization curves is considerably more rapid when the upper plate is made negative is also in agreement with this idea, since in the absence of electronegative gases the mobility of the negative

---

[12] E. Warburg, Ann. d. Physik **2**, 295 (1900).

[13] See J. J. Thomson, *Conduction of Electricity through Gases* (Cambridge University Press, London, 1928), Vol. I, p. 133.

[14] J. Franck, Verh. d. Deutsch. Phys. Ges. **12**, 291 (1910).

[15] W. B. Haines, Phil. Mag. **30**, 503 (1915).

[16] Private communication from Professor G. Jaffé.

[17] J. J. Thomson, *Conduction of Electricity through Gases* (Cambridge University Press, London, 1928), Vol. I, p. 206.

carriers is so much greater than that of the positive ions.

Whereas no measurements are available with which the present saturation curves in purified gases could be compared, a saturation curve at one atmos. of tank argon was taken for α-particles at right angles to the electric field. This can be compared with Helbig's[6] curve taken under similar conditions. While the argon used by us was supplied as 99.6 percent pure, Helbig's measurements were carried out in argon supplied as 99.5 percent pure. The two curves agree closely, both giving saturation at a field strength of about 18 volts/cm. It is interesting to note that this value is more than twice as great as the field strength required for saturation in purified argon at the same pressure when the paths of the α-particles are parallel to the field.

We wish to thank Professor J. D. Stranathan for many interesting discussions, and Professor H. P. Cady for his helpful advice on the purification of gases.

JANUARY 1 AND 15, 1943 PHYSICAL REVIEW VOLUME 63

The Angular Correlation of Successive Gamma-Rays*

ROBERT BERINGER
Sloane Physics Laboratory, Yale University, New Haven, Connecticut

(Received October 22, 1942)

The angular dependence of gamma-gamma coincidences has been studied for Na^{24}, Cl^{38}, and Br^{82} in an attempt to evaluate the predicted angular correlations between successive quanta. The coincidence rates were found to be independent of the relative directions of the quanta to within a few percent except as the counters approached each other closely, where an increase was observed. This increase was attributed to scattered quanta and was measured by an independent method.

INTRODUCTION

It has been pointed out by Pryce and Dunworth[1] that correlations are to be expected between the directions of quanta emitted in cascade from an excited nucleus, due to the coupling between the emitted quantum and the anisotropic radiation field of the preceding quantum. D. R. Hamilton[2] has calculated these spatial correlations for two successive dipole or quadrupole transitions involving three radiation levels. Since this analysis shows that the correlations are determined by the multipole orders of the transitions and the spins of the levels involved, some knowledge of these important quantities should be gained from experimental determinations of the directional correlations.

Several workers,[1, 2] using coincidence counting methods, have failed to observe any angular dependence in the rate of gamma-ray coincidences from such sources of successive quanta. Recently, however, Watase,[3] using Cl^{38} gamma-rays, reported a marked difference between the double coincidence rate from quantum pairs having directions differing by π radians and $\pi/2$ radians. No directional dependence in the coincidence rate was observed with Na^{24} gamma-rays.

The present paper describes angular coincidence experiments of somewhat greater completeness than have heretofore been reported. Several extraneous effects inherent in this kind of coincidence experiment are discussed and evaluated.

THE COINCIDENCE METHOD

If $W(\theta)$ represents the probability that two coincidence quanta 1 and 2 be emitted from a point source with a relative angle of θ between their directions, and if two counters of vanishingly

---

* This paper together with reference 5 is part of a thesis presented to the Graduate School of Yale University for the Degree of Doctor of Philosophy.

[1] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).

[2] D. R. Hamilton, Phys. Rev. **58**, 122 (1940).

[3] Y. Watase, Proc. Phys. Math. Soc. Japan **23**, 618 (1941).