Ionization, Attachment, and Breakdown Studies in Oxygen*

JOHN B. FREELY†
Department of Physics, New York University, New York, New York and Roberts College, Istanbul, Turkey

AND

L. H. FISHER‡
Department of Physics, New York University, New York, New York and Research Laboratories, Lockheed Missiles and Space Company, Palo Alto, California

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Uniform field steady state pre-breakdown currents have been measured in oxygen as a function of pressure corrected to 0°C, p_0, at constant E/p_0 (E is field strength) and at constant electrode separation d for $32<E/p_0<100$ V (cm Torr)$^{-1}$. Similar currents have been measured in oxygen-helium mixtures for $16<E/p_0<36$ (here p_0 is the sum of oxygen and helium pressures). Uniform field breakdown potentials have been measured in oxygen for a p_0d range corresponding to $46<E/p_0<100$ at breakdown. The currents have been used to evaluate the Townsend ionization coefficients α/p_0 (or F_0) and γ, and the electron attachment coefficient η/p_0 (or G_0). From the currents measured in oxygen, values of F_0 and G_0 were obtained for $32<E/p_0<60$. For $60<E/p_0<100$, it was not possible to evaluate F_0 and G_0 separately; however, for this E/p_0 range, values were obtained for F_0-G_0. It was possible to obtain values of F_0 and G_0 for oxygen for $16<E/p_0<30$ from the currents in the oxygen-helium mixtures. Values of γ for oxygen were obtained from pre-breakdown currents for $46<E/p_0<50$ (nickel cathode) and for $50<E/p_0<60$ (molybdenum cathode). In addition, values of γ for oxygen were obtained from sparking potential measurements for $46<E/p_0<100$ (nickel cathode) and for $50<E/p_0<100$ (molybdenum cathode). The values of γ obtained by the two methods are in agreement and are independent of cathode material. $\gamma\cong10^{-5}$ at $E/p_0=100$ and decreases to about 10^{-6} at $E/p_0=46$. The values of F_0 and G_0 obtained in the present work were used in conjunction with previous sparking potential measurements of Kachickas in oxygen (brass cathode) to evaluate γ for $39<E/p_0<44$. The results of this analysis indicate that in this E/p_0 range γ increases very sharply with decreasing E/p_0, reaching a value of about 10^{-3} at $E/p_0=39$.

INTRODUCTION

STUDIES of electron attachment by means of swarm experiments depend on some difference in property between electrons and negative ions, e.g., diffusion (Bailey[1]) and mobility (Bradbury[2] and Herreng[3]). Such methods assume that there is no ionization produced by electrons and hence are only properly applicable to conditions where no such ionization is produced.

Geballe and Harrison[4] have devised a method for studying electron attachment based on the difference in ability of electrons and negative ions to produce ionization.[5] Their method involves the measurement of steady-state Townsend ionization currents and is applicable to higher ratios of electric field strength to pressure than are the above methods. Schlumbohm[6] has developed a method also based on the difference in ionizing ability between the two kinds of particles. This method involves the measurement of the statistical distribution of the multiplication factor of avalanches produced by small numbers of initial electrons. The present paper is essentially an extension of the method of HG in which ionization and attachment in oxygen are studied over a wider range of field strength to pressure ratio. Furthermore, the method has been extended to include a study of secondary ionization in oxygen.

HG have derived an expression for the steady state uniform field pre-breakdown current I in a directly or dissociatively electron-attaching gas with no secondary ionization. For this case, they give

$$I/I_0=\{F_t\exp[(F_t-G_t)p_td]-G_t\}/(F_t-G_t), \quad (1)$$

where I_0 is the electron current produced at the cathode by external means, p_t is the pressure referred to some arbitrary temperature t, F_t is the number of ionizations produced per electron per unit distance of electron travel in the field direction per unit pressure referred to temperature t, G_t is the number of attachments per electron per unit distance of electron travel in the field direction per unit pressure referred to temperature t,

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† Present address: Roberts College, Istanbul, Turkey.

‡ Present address: Research Laboratories, Lockheed Missiles and Space Company, Palo Alto, California.

[1] V. A. Bailey, Phil. Mag. **50**, 825 (1925).

[2] N. E. Bradbury, Phys. Rev. **44**, 883 (1933).

[3] P. Herreng, Cahiers Phys. **38**, 7 (1952).

[4] R. Geballe and M. A. Harrison, Phys. Rev. **85**, 372 (1952); M. A. Harrison and R. Geballe, Phys. Rev. **91**, 1 (1953). This work will be referred to as HG throughout the paper.

[5] The referee of this paper pointed out that L. Frommhold in an unpublished paper presented at the Third International Conference on the Physics of Electronic and Atomic Collisions presented evidence that negative ions formed in oxygen under some of the conditions of the present experiment are unstable and undergo collisional detachment. An indication that detachment occurs in oxygen under some of the conditions of the present experiment has been given earlier by L. Frommhold, Z. Physik **160**, 554 (1960). If this is correct, reanalysis of the data in the present paper as well as all earlier data including an additional parameter, the detachment coefficient, may be called for.

[6] H. Schlumbohm, Z. Angew. Physik **11**, 156 (1959), in *Proceedings of the Fourth International Conference on Ionization Phenomena in Gases*, edited by N. R. Messon (North-Holland Publishing Company, Amsterdam, 1960); Z. Physik **166**, 192 (1962).

and d is the electrode separation. If Eq. (1) is valid, and if F_t and G_t are functions of E/p_t only, then pre-breakdown currents measured with various values of p_t, and d at constant E/p_t and I_0 can be analyzed to yield values of F_t and G_t. F_t is generally accepted to be a function of E/p_t only[7]; G_t will be a function of E/p_t only, providing the attachment is dissociative or if it is directly attaching at pressures high enough that saturation occurs. Geballe and Reeves[8] subsequently generalized Eq. (1) to take into account the release of secondary electrons at the cathode, i.e.,

$$I/I_0=A(1-[\gamma F_t/(F_t-G_t)] \times\{\exp[(F_t-G_t)p_td]-1\})^{-1}, \quad (2)$$

where γ is the number of secondary electrons released at the cathode per ion pair produced in the gas, and A is the expression on the right-hand side of Eq. (1). They also suggested a generalized Townsend breakdown condition for a gas whose pre-breakdown current is described by Eq. (2), i.e.,

$$1-[\gamma F_t/(F_t-G_t)]\{\exp[(F_t-G_t)p_td]-1\}=0. \quad (3)$$

HG obtained values of F_0 and G_0 in oxygen for $27<E/p_0<82$ and for $27<E/p_0<72$, respectively,[9] by fitting Eq. (1) to uniform field pre-breakdown currents measured as a function of d at constant p_0 and at constant E/p_0. (E is field strength.) Subsequently, DeBitetto and Fisher[10] studying pre-breakdown currents used a simplified form of Eq. (2) to obtain values of F_0, G_0, and γ for oxygen from pre-breakdown currents for $E/p_0=38.2$. From a study of pre-breakdown currents, Prasad and Craggs[11] obtained values of F_0 and G_0 in oxygen for $32<E/p_0<54$ and of γ for $48<E/p_0<54$. They also obtained values of γ in oxygen for $43<E/p_0<54$ from breakdown potentials using Eq. (3) together with their values of F_0 and G_0. Dutton *et al.*[12] have recently obtained values of F_0 and G_0 in oxygen for $37<E/p_0<42$ in the same manner. The values of F_0 and G_0 of HG and PC are in fair agreement while DF's single value and those of Dutton *et al.* are lower than their values by about a factor of two. Schlumbohm's[6] values of F_0 and G_0 are in reasonable agreement with those of HG and PC although his values of G_0 show considerable scatter. DF obtained a value of γ of 0.045 for $E/p_0=38.2$. This value was considered to be surprisingly high since the highest value of γ which had been measured in air was of the order of 10^{-4}.[13] In contrast to the large value of γ found by DF in oxygen, PC obtained values of γ in oxygen ranging from 10^{-7} to about 10^{-5}.

In addition, there exists a large number of determinations of G_t in oxygen at lower values of E/p_t.[1–3,14–18] These experiments which are based on entirely different principles than the one used in the present work apply to $E/p_0<10$ and have recently been reviewed extensively.[19] Furthermore, Burch and Geballe[20] using a pulsed Townsend discharge were able to obtain values of G_0 in the range of E/p_0 from 10 to 30.

The present work using steady-state Townsend discharges was undertaken to obtain values of F_0 and G_0 in oxygen over the same E/p_0 range as studied by HG, PC, and DF as well as for lower values of E/p_0 to bridge the gap between the low E/p_0 measurements,[1–3,14–18] and the measurements at high E/p_0.

Due to lack of current multiplication, steady-state pre-breakdown currents in oxygen cannot be used below $E/p_0=30$ to evaluate G_0. However, it was found possible to infer values of G_0 for oxygen for $16<E/p_0<30$ by measuring ionization currents in this E/p_0 region in oxygen-helium mixtures.

Pre-breakdown currents were measured in oxygen for large enough values of p_0d to determine γ for $50<E/p_0<60$. Values of γ in oxygen were also obtained from sparking potential measurements for $46<E/p_0<100$.

APPARATUS AND EXPERIMENTAL PROCEDURE

The discharge chamber used in most of the work has been described previously[21] and will be referred to as chamber A. A 0–5 kV supply with a voltage stability of 0.01% was used with chamber A. This chamber was connected to an ultrahigh vacuum system in which base pressures of about 10^{-9} Torr could be attained after a bake-out at 300°C. The pressure was measured with a null reading differential diaphragm manometer. Unless otherwise noted, spectroscopically pure tank oxygen obtained from Mathieson was used in chamber A.

A much larger discharge chamber designed for much higher voltage than chamber A (chamber B) was used to measure some breakdown potentials. Chamber B is described elsewhere.[22] The voltage source used with

[7] D. E. Golden and L. H. Fisher, Phys. Rev. **123**, 1079 (1961) indicate that under certain conditions F_t in argon may not be a function of E/p_t only.

[8] R. Geballe and M. L. Reeves, Phys. Rev. **92**, 867 (1953).

[9] Units of E/p are to be understood as being V (cm Torr)$^{-1}$ throughout. Subscripts of zero on p, F, and G indicate that these quantities are referred to 0°C.

[10] D. J. DeBitetto and L. H. Fisher, Phys. Rev. **111**, 390 (1958). This paper will be referred to as DF.

[11] A. N. Prasad and J. D. Craggs, Proc. Phys. Soc. (London) **77**, 385 (1961). This work will be referred to as PC.

[12] J. Dutton, F. Llewellyn Jones, and G. B. Morgan, Nature **198**, 680 (1963).

[13] F. Llewellyn Jones and A. B. Parker, Proc. Roy. Soc. (London) **A213**, 185 (1952).

[14] R. H. Healey and J. W. Reed, *The Behavior of Slow Electrons in Gases* (Amalgamated Wireless, Sydney, 1941), p. 94.

[15] A. Doehring, Z. Naturforsch. **7A**, 253 (1952).

[16] L. G. Huxley, R. W. Crompton, and C. H. Bagot, Australian J. Phys. **12**, 303 (1959).

[17] L. M. Chanin, A. V. Phelps, and M. A. Biondi, Phys. Rev. Letters **2**, 344 (1959).

[18] E. Kuffel, Proc. Phys. Soc. (London) **74**, 297 (1959).

[19] A. N. Prasad and J. D. Craggs, *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York and London, 1962).

[20] D. S. Burch and R. Geballe, Phys. Rev. **106**, 183 (1957).

[21] D. J. Rose, Phys. Rev. **104**, 273 (1956).

[22] L. H. Fisher, Phys. Rev. **72**, 423 (1947); D. J. DeBitetto and L. H. Fisher, *ibid.* **104**, 1213 (1956).

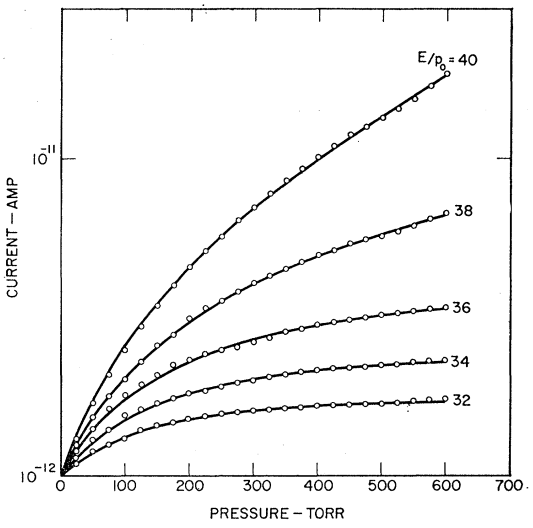

FIG. 1. I versus p_0 for oxygen at $d=0.200$ cm at various constant values of E/p_0.

chamber B was a 0–30.1 kV dc power supply with a voltage stability of 0.001%. With chamber B, a conventional unbaked vacuum system was used, with a base pressure of about 10^{-5} Torr. The pressure in chamber B was measured by a mercury manometer isolated from the chamber by a liquid-nitrogen trap. Ordinary tank oxygen was used in the experiments conducted in chamber B.

The procedure followed to obtain reproducible pre-breakdown currents is described elsewhere.[23] Pre-breakdown currents and breakdown potentials were measured with values of I_0 between 10^{-12} and 10^{-11} A.

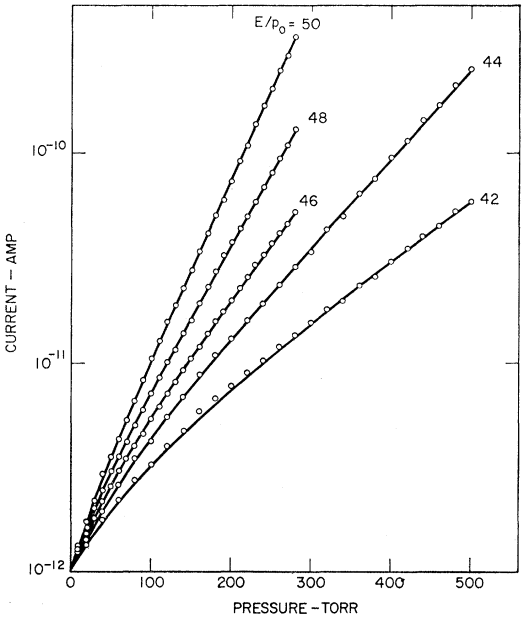

FIG. 2. I versus p_0 for oxygen at $d=0.200$ cm at various constant values of E/p_0.

[23] D. E. Golden and L. H. Fisher, Phys. Rev. **123**, 1079 (1961).

In obtaining the curves of I versus p_0d at constant E/p_0, p_0 rather than d was varied.

The potential of the conducting wall coating of chamber A was set midway between the anode and cathode potentials. Preliminary experiments had indicated that the pre-breakdown currents were unaffected by the value of the wall potential.

EXPERIMENTAL DATA AND DISCUSSION

A. Measurements of F_0 and G_0

Pre-breakdown currents were measured in oxygen for $32<E/p_0<100$ using chamber A. For $32<E/p_0<50$ the value of d was 0.200 cm; for $50<E/p_0<100$ the value of d was 0.136 cm. The data are shown as points in Figs. 1–4 where all points have been normalized to the same value of I_0 (10^{-12} A). (The method of obtaining I_0 is described below.) There is no evidence of up bending in these figures due to the presence of secondary emission, and it was assumed that secondary effects can be neglected in analyzing such data. For $E/p_0<32$, $F_0 \ll G_0$ and the current amplification is small. Therefore $E/p_0=32$ represents the practical lower limit at which steady-state pre-breakdown currents in oxygen can be studied conveniently to give accurate values of F_0 and G_0. For $E/p_0>60$, $F_0 \gg G_0$, and as has been pointed out by HG, the pre-breakdown currents resemble those in a nonattaching gas. Therefore, for $E/p_0>60$, the coefficients F_0 and G_0 cannot be determined separately; only an effective ionization coefficient F_0-G_0 has been determined.

The coefficients F_0 and G_0 were evaluated for $32<E/p_0<60$ by fitting the experimental points to Eq. (1) using an iterative least-squares analysis programmed for an IBM 650 computer. A value of I_0 was inserted into the computer program which was obtained in the following way. DF have shown that in oxygen, I varies linearly with p_0d at that value of E/p_0 (~ 38) for which $F_t=G_t$, and for every set of current-voltage curves taken in oxygen, several values of I were measured at this value of E/p_0 at very low values of p_0d. The resulting linear plot of I versus p_0d was then extrapolated to determine I_0. The curves for $32<E/p_0<60$ in Figs. 1–3 were obtained by using the values of F_0 and G_0 resulting from the least-squares analysis. In most cases not a single point lies more than 1% off the curves. Inserting other values of I_0 into the computer resulted in values of F_0 and G_0 which led to curves which deviate more from the experimental points. This indicates that the method of determining I_0 described above is not affected by back diffusion for $32<E/p_0<60$.[24] Values of F_0-G_0 for $60<E/p_0<100$ were obtained by a simple graphical method and were

[24] Measurements of back diffusion in oxygen by J. K. Theobald, J. Appl. Phys. **24**, 123 (1953) were limited to $E/p \leq 20$ (temperature not specified).

used to obtain the curves for this E/p_0 range in Figs. 3 and 4.

Measurements carried out with values of d different from those above have shown that the values of the coefficients obtained are independent of the value of d used.

It was found possible to obtain measurable values for the current multiplication in oxygen-helium mixtures for $E/p_0<32$. It was thus possible to obtain ionization and attachment coefficients for oxygen-helium mixtures for $E/p_0<32$. It was hoped that an attachment coefficient could be obtained for oxygen alone for $E/p_0<32$ by extrapolating to zero helium concentration. Therefore, steady-state pre-breakdown currents in oxygen-helium mixtures were measured at a number of oxygen to helium mole fraction ratios at each of a number of

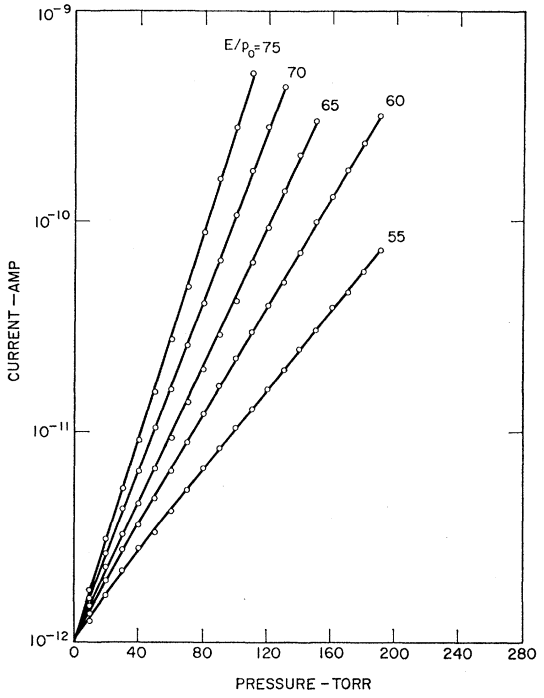

FIG. 3. I versus p_0 for oxygen at $d=0.136$ cm at various constant values of E/p_0.

values of $E/p_0<32$ (at $d=0.200$ cm), p_0 being the sum of the partial pressures of the two components. (The helium was spectroscopically pure.) An effective value of the attachment coefficient for oxygen alone G_0 was calculated from the expression $G_0=G_0^*/(1-f_{\mathrm{He}})$, where G_0^* is the attachment coefficient measured for the mixture and f_{He} is the mole fraction of helium. As mentioned above, it was thought that G_0 versus f_{He} could be extrapolated to $f_{\mathrm{He}}=0$ so as to give the value representative of oxygen alone at that value of E/p_0. To test this procedure, currents were measured as a function of p_0 in oxygen-helium mixtures (in chamber A) as a function of f_{He} at $E/p_0=32$, 34, and 36 where

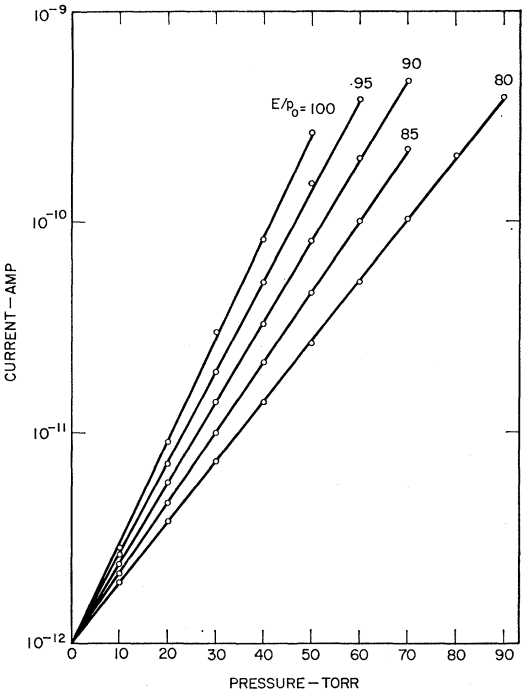

FIG. 4. I versus p_0 for oxygen at $d=0.136$ cm at various constant values of E/p_0.

measurements of G_0 had already been made in oxygen alone. Figure 5 shows the results obtained at $E/p_0=36$ where the points are once again the measured values and the curves were obtained by the same least squares analysis mentioned earlier. In the case of mixtures, the value of I_0 was obtained by extrapolation of measured currents to $p_0d=0$. For all the mixture measurements to be described, the experimental points deviate from the resulting curves by only 1 or 2% on the average. Again

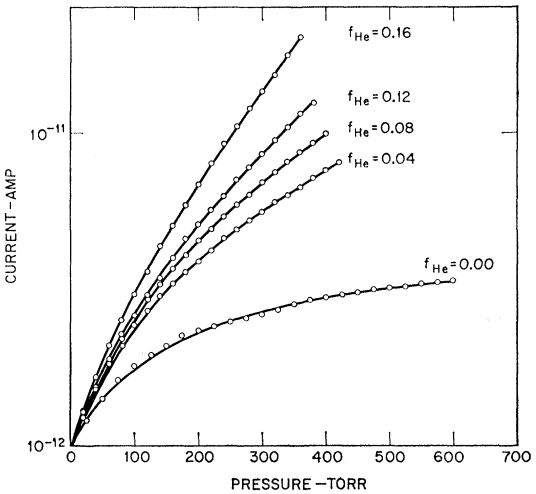

FIG. 5. I versus p_0 at $d=0.200$ cm and at $E/p_0=36$ for various oxygen-helium mixtures. f_{He} represents the mole fraction of helium.

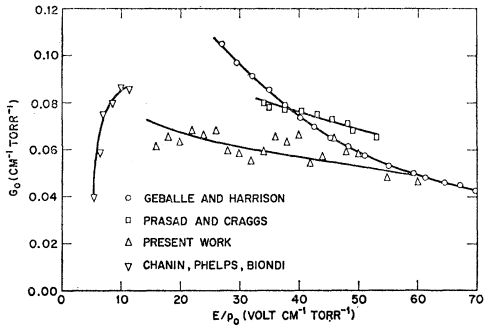

FIG. 6. G_0 versus E/p_0 for oxygen obtained in the present work as well as the results of some other workers.

calculations with slightly varying values of I_0 led to larger deviations.

Values of G_0 obtained with mixtures and with pure oxygen at $E/p_0=32$, 34, and 36 are given in Table I. Here $\langle G_0\rangle_{av}$ is the average value of G_0 for all of the values of f_{He} (excluding $f_{He}=0.00$) at a particular value of E/p_0. G_0 seems to be independent of f_{He} and is in good agreement with the value for oxygen alone. It was concluded from these experiments that values of $\langle G_0\rangle_{av}$ obtained in oxygen-helium mixtures could be used with confidence for the determination of the attachment coefficient in oxygen at low values of E/p_0 where measurements in oxygen alone are not possible.

Measurements of pre-breakdown currents were therefore carried out for $16<E/p_0<30$ using four or five different relative concentrations at each value of E/p_0. At each value of E/p_0 the total spread in values of G_0 was less than 20%. The measurements were not extended below $E/p_0=16$ because in that region the current multiplication is negligible even for mixtures containing very high percentages of helium. Furthermore, when very high concentrations of helium are used, G_0* becomes too small to evaluate conveniently.

Figure 6 presents the values of G_0 obtained in the present work along with the results of HG and two of the most recent studies, one at low and the other at high E/p_0. (For a complete summary of all studies of G_0 in oxygen up to 1961, one may refer to Fig. 5 of PC.) The curve due to HG is their published curve based on values of G_0 obtained by a graphical curve-fitting procedure. However, we have subjected their original data to the iterative least-squares analysis mentioned earlier and find values of G_0 which are different from theirs by as much as 50%. Also, the recalculated points scatter to such an extent as to make it difficult to decide on the best curve to fit the data.

The values of F_0 for oxygen obtained in the present work are shown in Fig. 7, together with the published curve of HG and the values given by PC. Figure 8 gives the values of F_0-G_0 obtained in the present experiment.

TABLE I. Comparison of attachment coefficients in oxygen-helium mixtures with those in oxygen alone.

E/p_0 (V cm^{-1} Torr^{-1})	f_{He}	G_0 (cm Torr)$^{-1}$	$\langle G_0\rangle_{av}$ (cm Torr)$^{-1}$
32	0.00	0.055	
	0.04	0.058	
	0.08	0.064	
	0.12	0.066	
	0.16	0.063	0.063
34	0.00	0.059	
	0.04	0.065	
	0.08	0.062	
	0.12	0.062	
	0.16	0.067	
	0.20	0.067	0.067
36	0.00	0.065	
	0.04	0.068	
	0.08	0.070	
	0.12	0.063	
	0.16	0.064	0.065

B. Measurements of γ

Values of γ were determined in oxygen both by studies of pre-breakdown currents extended to large values of p_0d and also from sparking potential measurements. Values of γ were obtained from pre-breakdown currents for $46<E/p_0<50$ (nickel cathode, chamber B) and for $50<E/p_0<60$ (molybdenum cathode, chamber A). Values of γ were obtained from sparking potential measurements for $46<E/p_0<100$ (nickel cathode, chamber B) and for $50<E/p_0<100$ (molybdenum cathode, chamber A).

Values of γ obtained from pre-breakdown current studies were limited to $50<E/p_0<60$ for chamber A and to $46<E/p_0<50$ for chamber B. It is impossible to use pre-breakdown currents to evaluate γ for $E/p_0>60$, because the value of p_0d at sparking is too low to allow a sufficiently large variation in p_0 (even for small d) to obtain accurate values of γ. Measurements of γ in chamber A could not be made for $E/p_0<50$ because chamber A was not designed for the high voltage required. For this reason chamber B was used to extend the pre-breakdown (and sparking potential) studies to

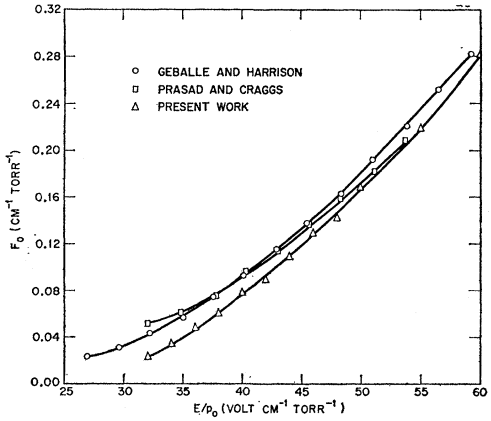

FIG. 7. F_0 versus E/p_0 for oxygen obtained in the present work as well as the results of other workers.

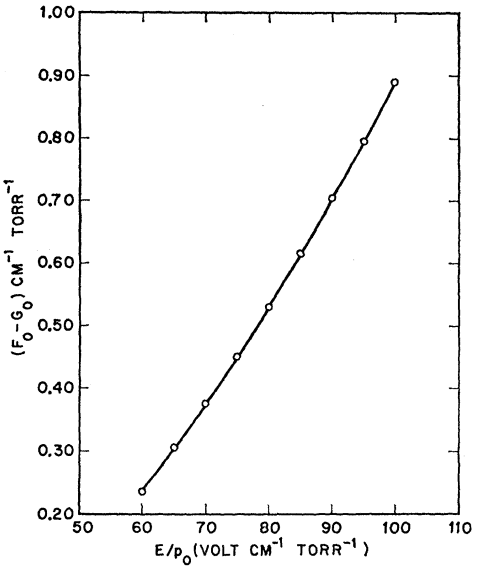

FIG. 8. F_0-G_0 versus E/p_0 for oxygen.

$E/p_0<46$. The available power supply did not permit studies for $E/p_0<46$.

The values of γ from the pre-breakdown currents were evaluated in the following way. For large values of p_td Eq. (2) assumes the form

$$I/I_0=\{[F_t/(F_t-G_t)]\exp[(F_t-G_t)p_td]\}/\{1-[\gamma F_t/(F_t-G_t)]\exp[(F_t-G_t)p_td]\}. \quad (4)$$

Under these conditions the semilogarithmic plot of I versus p_t resembles that of a nonattaching gas with an effective primary ionization coefficient F_t-G_t. F_0-G_0 was first determined from the linear section of the semilogarithmic plot of I versus p_0d. This linear portion was extrapolated to $p_0d=0$ to give $I_0F_0/(F_0-G_0)$. From a determination of I_0 in the manner described earlier, the quantity $F_0/(F_0-G_0)$ was then obtained. Using these quantities, values of γ were obtained from the up-bending part of the curve using Eq. (4).

Values of γ were obtained from the sparking potential measurements by use of the measured values of F_0 and G_0 as functions of E/p_0 in conjunction with Eq. (3). For $p_0d>50$, the determination of the sparking potential (for both nickel and molybdenum cathodes) was complicated by the fact that the sparking potential does not appear to have a unique value. Sparking occurs over a wide range of voltage (e.g., 200 V in 5000 V) depending on how long one is willing to wait for a spark to occur. This time increases as the voltage is decreased and may be as long as many minutes. This effect has been observed previously in oxygen.[25] The value of the sparking potentials used in the calculation are those for which a spark occurred within a fraction of a second after the application of the voltage. This definition of the sparking potential leads to reproducible values. (The values of γ would not be drastically different had lower voltages in the breakdown range been used.) For $p_0d<50$, this effect does not occur and the sparking potential is uniquely defined. It is interesting to note that in this p_0d region, the gas breaks down into a diffuse glow whereas for $p_0d>50$ the breakdown occurs as a filamentary spark.

The values of γ as determined from the study of the pre-breakdown currents are in good agreement with those obtained from sparking potential measurements. Furthermore, they seem to be the same for molybdenum and nickel cathodes. Figure 9 shows the values of γ found in the present work from sparking potentials. Although γ could not be measured directly for $E/p_0<46$ in the present experiments, it was possible to compute γ (brass cathode) from Eq. (3) for $39<E/p_0<45$ using the sparking potentials measured by Kachickas[25] together with values of F_0 and G_0 determined in the present experiments. The values of γ obtained by this calculation are also shown in Fig. 9 where it is seen that γ rises very sharply with decreasing E/p_0 below about $E/p_0=45$. The value of γ obtained by DF at $E/p_0=38.2$ is also shown in Fig. 9. It appears that the low values of γ found in the present experiments for $E/p_0>46$ may not be inconsistent with the large value of γ found by DF at $E/p_0=38.2$. Also shown in Fig. 9 are the values obtained by PC for several kinds of electrodes in oxygen by pre-breakdown current measurements (I) and from sparking potential measurements (V_s). It seems reasonable to assume that the spectacular increase in γ with decreasing E/p_0 is associated with a change in the nature of the secondary mechanism from a photoelectric one at low E/p_0 to a positive ion one at high E/p_0.

Sparking potentials in oxygen as measured in chambers A and B were compared over a wide range of p_0d and were found to be in excellent agreement despite the difference in gas purity. In addition, some measure-

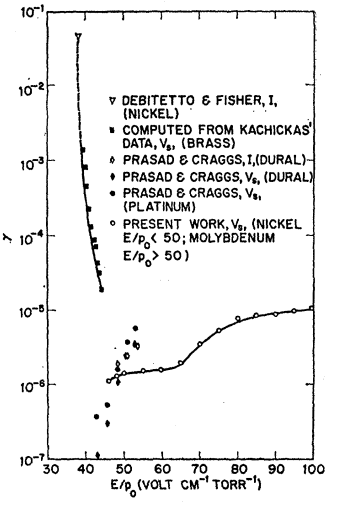

FIG. 9. γ versus E/p_0 for oxygen. Includes present results, those computed from Kachickas' data, point determined by DF, and values obtained by Prasad and Craggs. I and V_s indicate that the values were obtained from pre-breakdown current measurements and from sparking potential measurements, respectively.

[25] G. A. Kachickas, Ph.D. thesis, New York University, 1950, (unpublished).

ments of the sparking potential in oxygen were made in chamber B at a base pressure of 10^{-2} Torr and the values are in excellent agreement with those found in chamber A at a base pressure of 10^{-2} Torr; the values are in excellent agreement with those found in chamber A at a base pressure of 10^{-9} Torr. This indicates that the sparking potential of oxygen is unaffected by the presence of the ordinary background impurities up to partial pressures of about 10^{-2} Torr. Also, in all the measurements it was found that the sparking potential is independent of whether or not the cathode was irradiated with uv light, indicating that the sparking potential is independent of the magnitude of I_0, at least up to values of $I_0=10^{-11}$ A.

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Critical Percolation Probabilities by Series Methods

M. F. SYKES
Wheatstone Physics Laboratory, King's College, London, England
AND
J. W. ESSAM
Westfield College, London, England

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Series estimates of the critical percolation probabilities for the "bond problem" and the "site problem" are presented for two- and three-dimensional lattices. Good agreement with the Monte Carlo estimates of Frisch *et al.* and of Dean is obtained. The series method gives information on the critical behavior of the mean cluster size and it is found that there is a much sharper growth of large clusters in two dimensions than in three dimensions as the critical concentration is approached from below.

1. INTRODUCTION

RECENTLY, numerical estimates for the critical probabilities that arise in two percolation problems of physical interest have been given by a number of authors.[1–5] In the *bond problem*, described by Broadbent and Hammersley,[6] one studies percolation through a "random maze" of paths (bonds) which are "open" with probability p and "blocked" with probability $q=1-p$. We shall treat the case when the "maze" is an infinite crystal lattice. In the *site problem* introduced by Domb,[7] one supposes the sites of the lattice to be occupied with probability p and vacant with probability q. Site problems are more general since every bond problem can be made isomorphic with a site problem on a suitably chosen *covering lattice* by the bond-to-site transformation.[8] A very complete discussion of the underlying physical problems is given in the references quoted above.

Percolation problems on infinite lattices are characterized by the occurrence of a critical probability p_c, above which there is a nonzero probability of a site being a member of an infinite "cluster" of connected sites. Estimates for p_c for the more important crystal lattices have been obtained by Monte Carlo methods[1–4] and from exact series expansions.[5] It is the object of the present paper to develop the series method.

It was suggested by Domb[9] that the method of exact series expansions could be applied to a study of percolation problems. In particular the critical probability could be investigated by expanding the mean size of finite clusters $S(p)$ as a power series in p. A series of positive terms results and hence the radius of convergence of the expansion can be identified with the critical percolation probability p_c. Subsequently, series developments for $S(p)$ were published and examined briefly by Domb and Sykes[5] who also suggested that the mean cluster size in the critical region could be investigated by series methods. Domb and Sykes found the behavior of the coefficients not altogether smooth, particularly in

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