fluidity and superconductivity. They have been explicitly evaluated for the *ideal* Bose gas only. We have shown that above the transition temperature the off-diagonal element of the single-particle density matrix vanishes in the limit of large R, that is there is no off-diagonal long-range order in the single-particle density matrix. We find also that the off-diagonal element of the two-particle density matrix vanishes in the region of large distances. In the degenerate region both in the vicinity of the transition temperature and near $T=0$ we find that the off-diagonal element of the single-particle density matrix does not vanish as R becomes large, that is, there is off-diagonal long-range order. Similarly we find that the off-diagonal element of the two-particle density matrix does not vanish in the limit of large distances.

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First Townsend Ionization Coefficient in Hydrogen*

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Rose's measurements of the first Townsend ionization coefficient η in hydrogen for $E/p_0 \gtrsim 100$ V (cm Torr)$^{-1}$ have been questioned by a number of authors principally either because of a supposed neglect of the second Townsend ionization coefficient γ, or because electrons allegedly do not have an energy distribution which is only characteristic of E/p_0, or because of gas purity. In order to resolve these questions, new measurements of η in hydrogen have been carried out for $50 \leq E/p_0 \leq 800$ in an ultrahigh-vacuum system. The present work takes γ into account, and the values of η obtained are in agreement with those of Rose over the whole range of E/p_0 studied. Various methods of evaluating η from prebreakdown ionization currents are critically discussed. The present results show that prebreakdown ionization-current data in hydrogen can be represented in the usual way, at least up to $E/p_0=800$.

INTRODUCTION

The prebreakdown ionization current I in a uniform electric field E with gas at pressure[1] p_t may be written as

$$I/I_0=\exp[\eta(V-V^*)]/[1-\gamma\{\exp[\eta(V-V^*)]-1\}], \quad (1)$$

where η is the average number of electrons created in the gas by an electron per unit of potential difference applied across the gap, γ is the average number of secondary electrons liberated from the cathode (by any mechanism) which escape the cathode per positive ion produced in the gas, I_0 is the electron current liberated from the cathode by external means which escapes the cathode, V is the gap voltage, and V^* is related to the voltage through which the electrons must move in order that the electrons have a velocity distribution characteristic of the applied value of E/p_t. Equation (1) is valid only for $V>V^*$.

In order to be able to use Eq. (1) to obtain the value of η at a particular value of E/p_t, it is necessary to measure currents at various values of V at that value of E/p_t. Keeping E/p_t constant insures that I_0, V^*, and γ are constant providing the illumination of the cathode is constant and that the cathode surface is stable. A voltage variation at constant E/p_t implies a prescribed variation in the product p_td where d is the electrode separation. The most common method which has been used to determine η has been to measure prebreakdown ionization currents keeping E/p_t and p_t constant by varying d. However, it is often more convenient to keep E/p_t and d constant by varying p_t.

Despite the fact that values of η have been measured for many years, there has been little agreement between different experimental values over an extended range of E/p_t for any gas. It seemed that the work of Rose[2] had finally made available reliable values of η in hydrogen[3] for $15 \leqslant E/p_0 \leqslant 1000$.[4] Subsequent measurements of η were carried out by Blevin, Haydon, and Somerville[5] for $50<E/p_0<190$, by Jones and Llewellyn Jones[6] for

* Supported by the Lockheed Independent Research Funds.

[1] The subscript t refers to the gas temperature in degrees centigrade.

[2] D. J. Rose, Phys. Rev. **104**, 273 (1956). Rose summarizes earlier measurements of η in hydrogen.

[3] All subsequent discussion in the paper refers to hydrogen.

[4] Units of E, p, and d are V/cm, Torr, and cm, respectively throughout.

[5] H. A. Blevin, S. C. Haydon, and J. M. Somerville, Nature **179**, 38 (1957).

[6] E. Jones and F. Llewellyn Jones, Proc. Phys. Soc. (London) **72**, 362 (1958).

$40 \leqslant E/p_0 \leqslant 375$, by Davies and Milne[7] for $40 \leqslant E/p_0 \leqslant 500$, by Haydon and Robertson[8] for $50 \leqslant E/p_0 \leqslant 500$, by Chanin and Rork[9] for $20 \leqslant E/p_0 \leqslant 500$, and by Barna, Edelson, and McAfee[10] for $17 \leqslant E/p_0 \leqslant 100$. There seems to be general agreement for the values of η for $E/p_0 \lesssim 100$. For $E/p_0 \gtrsim 100$, Jones and Llewellyn Jones[6] and Haydon and Robertson[8] obtained values which are in good agreement with each other but which are in some cases only half as large as those of Rose.[2] However, for the same E/p_0 range, Davies and Milne[7] obtained values only 10 to 15% lower than those of Rose.[2] Chanin and Rork[9] quote results in good agreement with those of Rose over the entire range of E/p_0 studied but arrive at the conclusion that values of η are meaningless for $E/p_0 \gtrsim 100$ because the electrons do not have enough collisions to obtain a velocity distribution characteristic of the applied values of E/p_0 only, up to and including the limiting values of V determined by breakdown. Jones and Llewellyn Jones[6] criticized Rose's values of η on the basis that Rose neglected γ when evaluating η. Haydon and Robertson[8] suggested that Davies and Milne's[7] gas was not pure but do not discuss the work of Rose.[2] Thus, for values of $E/p_0 \gtrsim 100$ the situation seemed sufficiently confused to warrent further study.

We have been analyzing ionization currents in nonuniform electric fields in hydrogen, and have required values of η for large values of E/p_0.[11] We have therefore measured values of η in hydrogen over an extended range ($50 \leqslant E/p_0 \leqslant 800$) in order to provide ourselves with reliable values of η taking special account of all previous work.

APPARATUS AND PROCEDURE

The tube used in this study was the same one described by Rose[2] and was part of an ultrahigh-vacuum system capable of an ultimate pressure of less than 10^{-9} Torr. The rate of rise of pressure in the system when blanked off from the pump was about 2×10^{-9} Torr/min over the first 5 min. The cathode surface was stabilized as has been previously described.[12] The data were obtained at constant values of E/p_0 and d by varying p_0 as has also been previously described.[12] The pressure was measured by mercury manometer, Dubrovin gauge, or McLeod gauge, all connected to the system by means of a capacitance manometer. The gas was reagent-grade hydrogen[13] in Pyrex flasks.

RESULTS

Measurements of prebreakdown ionization currents were made for $E/p_0 = 50$, 100, 150, 200, 300, 400, 500, 600, and 800 at various constant values of d. For $E/p_0 \leqslant 200$, the value of d was 0.200; for $E/p_0 = 300$ and 400 two values of d were used: 0.200 and 0.067. For $E/p_0 \geqslant 500$, d was 0.067. At each value of E/p_0 and d, at least two and in most cases three runs were made.

The data were analyzed in the following way. For a set of data at a given E/p_0 guesses were first made for values of I_0, η, γ, and V^*. Then a computer was used to calculate currents for $V > V^*$ from these values and for ten other values of each of these parameters using Eq. (1). The parameters were varied by adding and subtracting 18, 36, 54, 72, and 90% to the original guesses for each parameter. The best fit within this range of variables was determined by choosing the set of parameters which yielded the minimum sum of the squares of the percentage differences between calculated and measured currents. If the best result did not include a value of any one of the parameters either 90% above or below the original choices, the process was repeated using the best values of the four parameters as new guesses with the same number of intervals but each interval being 0.7 of the previous one. This process was repeated ten times (assuming that the extreme values of the parameters were never among the best values). If at any time an extreme value of one of the parameters was a best value, the program stopped, and new guesses were in-

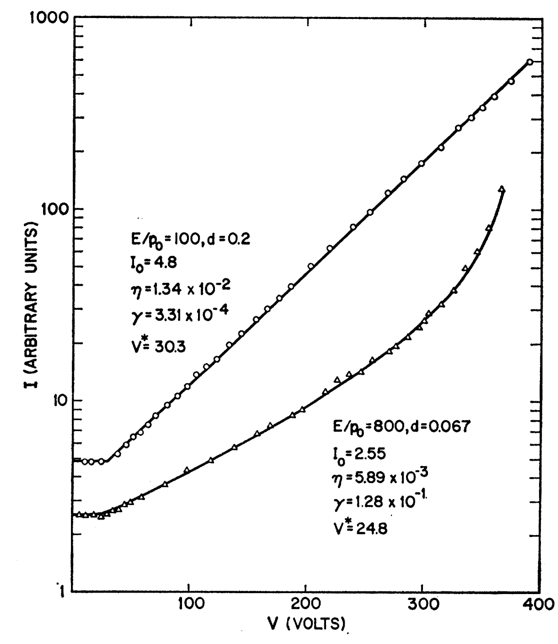

FIG. 1. Prebreakdown ionization currents as a function of voltage for $E/p_0 = 100$, $d = 0.2$ and $E/p_0 = 800$, $d = 0.067$. Points are measured values and curves for $V > V^*$ are calculated from Eq. (1) with values of I_0, η, γ, and V^* obtained by the least-squares analysis described in text and listed in figure. For $V < V^*$ a horizontal line was drawn for each curve with $I = I_0$.

[7] D. E. Davies and J. G. C. Milne, Brit. J. Appl. Phys. **10**, 301 (1959).

[8] S. C. Haydon and A. G. Robertson, Proc. Phys. Soc. (London) **78**, 92 (1961).

[9] L. M. Chanin and G. D. Rork, Phys. Rev. **132**, A2547 (1963).

[10] S. F. Barna, Jr., D. Edelson, and K. B. McAfee, Jr., J. Appl. Phys. **35**, 2781 (1964).

[11] L. H. Fisher and D. E. Golden, Bull. Am. Phys. Soc. **10**, 188 (1965).

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

[13] An analysis of the hydrogen supplied by the manufacturer showed the following maximum values of gas impurities in ppm V/V: $A=1$, $CO_2=1$, $CO=1$, $CH_4=1$, Hydrocarbons $=1$, $N_2=5$, $O_2=5$.

serted. The maximum difference between measured and calculated currents obtained in this way is about 3%.

Values of η obtained from run to run at a particular value of E/p_0 showed a spread of about $\pm 5\%$. The value of η was observed to be independent of d for the two values of E/p_0 for which different values of d were used. Calculated values of I_0 were between 10^{-12} and 10^{-11} A and measured currents were always kept below 10^{-9} A. The initial currents and multiplications were always small enough to assure that space-charge effects were negligible. Calculated values of V^* were between about 18 and 30 V. Calculated values of I_0 and V^* were consistent with rough values of these parameters estimated from the data for low voltages. Values of γ increase with increasing values of E/p_0 from 2×10^{-4} to about 0.1.

Figure 1 shows two runs of measured current-voltage characteristics (one for $E/p_0=100$ and the other for $E/p_0=800$). The points are the experimental data and the curves are plots of Eq. (1) for $V>V^*$ using values of I_0, η, γ, and V^* determined in the manner described above. For $V<V^*$ the calculated values of I_0 are shown by horizontal lines. The points in Fig. 2 show the resulting values of η obtained for the various values of E/p_0. The upper curve represents the data of Rose[2] to $\pm 2\%$. The other curves shown are the results of Davies and Milne,[7] Haydon and Robertson,[8] and Jones and Llewellyn Jones.[6] Within the precision of the experiment, the values of η obtained in the present work are in good agreement with those of Rose.[2,14]

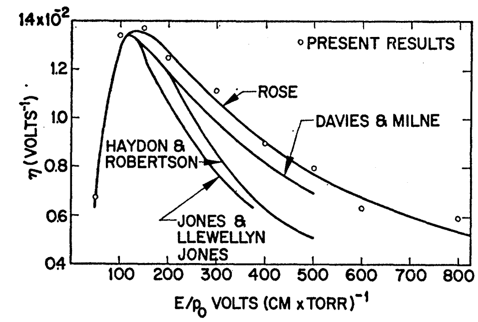

FIG. 2. Values of the first Townsend ionization coefficient η versus E/p_0. The points are values obtained in the present study and the curves represent the results of other workers.

DISCUSSION

In the past, essentially four methods of analysis have been used to obtain values of η in uniform fields from prebreakdown ionization currents at constant E/p_t. The first is the measurement of the slope of a plot of $\ln I$ versus V for low values of V (but for $V>V^*$). This method is valid only if $\gamma \ll 1$.[12] The second is a three-point method originally due to Townsend and MacCallum[15] which uses prebreakdown currents measured at three voltages $V_1<V_2<V_3$ such that $V_2-V_1=V_3-V_2=\delta V$. This three-point method has been generalized to the case of unequal voltage intervals.[6] The third method is due to Gosseries[16] and is a way of plotting the reciprocal of currents measured at certain values of V against the reciprocal of currents measured at certain other prescribed values of V such that a linear plot results whose slope allows the determination of η. Linearity of the plot is assumed to assure the constancy of I_0 and γ throughout the measurements. The fourth method may be considered to encompass all iterative procedures which involve some criterion to determine all of the various parameters involved simultaneously using all of the current-voltage data.

One of the weaknesses of the first three methods of analysis is that they only give definite recipes for determining η and do not specify a definite procedure for determining the other three parameters. Under suitable circumstances, the first three methods may be used to calculate all four parameters accurately. However, in many cases, these methods have been used where they cannot be applied properly. The second method has been used to determine values of η without using all of the available data. Both the second and third methods have been used to evaluate η without evaluating the other three parameters. Without evaluating all four parameters, there is no guarantee that the data can be well represented using the determined value of η. Furthermore, there always exists the possibility of getting a result for η which does not allow the possibility of obtaining values of one or more of the remaining three parameters of Eq. (1) which are physically meaningful (V^* less than the ionization potential of the gas, $\gamma<0$) although the data may appear to be well represented. As will be discussed later, the three point methods should lead to fluctuations in the calculated values of η depending upon which three points are used.[17] The fourth method is the only method which determines all four parameters simultaneously in a prescribed way using all of the data.

Jones and Llewellyn Jones[6] have used a three-point method to fit their data (obtained in an unbaked system). The only data they present is for $E/p_0 \simeq 375$ for which ten values of $\ln(I/I_0)$ are plotted. The data presented were not obtained for sufficiently low values of V to have observed the constancy of I with V for $V<V^*$. Therefore, their choice of I_0 was not a straightforward matter and their value of I_0 is questionable.

[14] Rose implies that his $\ln I$ versus V curves were linear for current multiplications up to 20 over the entire range of E/p_0 studied. This is contrary to what was observed in the present experiment and must have been due to extraordinarily small values of γ over the entire E/p_0 range in Rose's work. The fact that large values of γ were found in the present study must have been due to the different cathode-conditioning procedure.

[15] J. S. Townsend and S. P. MacCallum, Phil. Mag. **6**, 857 (1928).

[16] A. Gosseries, Physica **6**, 458 (1939).

[17] Breakdown limits the maximum value of δV which can be employed and tends to make the three-point method useful only for extremely small values of E/p_0, a range for which $\gamma \ll 1$, and for which the first method suffices.

They do not state which set or sets of three points they used for analysis. The value of η which they give together with their value of I_0 allowed them to calculate values of γ and V^*. They obtained a value of V^* of less than 1.8 V which is much too small to be physically meaningful. The fact that too small a value of V^* was obtained indicates that the values of η and I_0 are too small and that the value of γ is too large. Although their set of four constants represent their data fairly well, their value of η cannot be accepted. Their data, as read off their graph, were subjected to the same least-squares analysis discussed above. This yielded a value of η in reasonable agreement with the value given by Rose[2] and by the present work for this E/p_0, a value of $V^*\simeq 20$, a value of I_0 about 8% larger and a value of γ about 10% smaller than given by Jones and Llewellyn Jones.[6] The values of the parameters obtained by the least-squares analysis yielded calculated currents in somewhat better agreement with the data than are obtained using the parameters given by Jones and Llewellyn Jones.[6] If they have used similar procedures for evaluating η at other values of E/p_0, their values of η cannot be accepted.

Haydon and Robertson[8] used the Gosseries[16] method to analyze their data (obtained in an unbaked system). They present no raw data from which one may evaluate their analysis. As previously mentioned, their results for η are in agreement with those of Jones and Llewellyn Jones.[6] After a glow discharge treatment of their cathode for five minutes at $E/p_0=350$, Haydon and Robertson[8] obtained a value of η in good agreement with that of Davies and Milne[7] (and Rose[2]). They consider that the glow discharge contaminated their gas sample and they invoke effects of gas purity to explain why their results are to be preferred to those of Davies and Milne.[7] Since Haydon and Robertson[8] did not use an ultrahigh-vacuum system, their conclusions concerning gas purity cannot be accepted. They also find that the size of holes in anode affects their values of η.

Chanin and Rork[9] present values of η calculated by by the three-point method of equal intervals. They calculated values of η as a function of V_3 at a given value of E/p_0 and found wide fluctuations in the value of η. They state that for low values of E/p_0, η does not exhibit a pronounced variation with V_3 once the electrons have undergone a "sufficient" number of collisions. For moderate values of E/p_0, η was observed to have an oscillatory behavior, while for $E/p_0=500$, η exhibited a strong decrease with increasing V_3. They conclude that the electrons are not in equilibrium with the field for $E/p_0>150$. Chanin and Rork[9] state that to observe the variation of η with V_3, δV must be small with corresponding large errors in the calculated values of η. Their tabulated values of η as a function of E/p_0 were determined by using values of δV "as large as possible" although the values of δV used are not specified.[17] We believe that the fluctuations in η reported by Chanin and Rork[9] may be explained as being simply due to the method of analysis as will now be discussed.

For equal intervals the three-point method may be used to find η from

$$\exp(\eta\delta V)=(I_3/I_1)[(I_2-I_1)/(I_3-I_2)], \qquad (2)$$

where I_1, I_2, and I_3 are the prebreakdown currents appropriate to V_1, V_2, and V_3, respectively. An expression for the error in η may be written as

$$\Delta\eta=(\delta V)^{-1}[\Delta I_3/I_3-\Delta I_1/I_1+\Delta(I_2-I_1)/(I_2-I_1) -\Delta(I_3-I_2)/(I_3-I_2)]. \qquad (3)$$

Equation (3) may be rewritten as

$$|\Delta\eta|\leqslant 6(\Delta I/I)(\delta V)^{-1}, \qquad (4)$$

where $\Delta I/I$ represents the fractional error in any single current measurement. In order to insure that this method gives values of η whose fluctuations are small compared to η one must have

$$\delta V\gg 6(\Delta I/I)/\eta. \qquad (5)$$

The case leading to the smallest acceptable values of δV would be to consider the maximum value of η which is about 0.014 V^{-1}. Assuming that $\Delta I/I$ is about 0.03, this case requires a value of $\delta V\gg 13$ V. In this case, for $\delta V=130$ V there would still be fluctuations in η of about 10%. Smaller values of η would require even larger values of δV. Chanin and Rork[9] present curves of $\eta(E/p_0)$ versus V_3 for $E/p_0=30$, 100, and 500. They do not specify what value or values of δV were used. The maximum values of V_3 used at these three values of E/p_0 were 480, 380, and 200 V, respectively. From the previous discussion, one can see that one cannot obtain more than one or two possibly reliable values of η for any of the above values of E/p_0. To conclude that the value of η at a given E/p_0 is a function of V_3 from the data of Chanin and Rork[9] seems to us unwarranted. The fluctuations in η as a function of V_3 reported by Chanin and Rork[9] are predicted by Eq. (4). Furthermore, although one or two values of η for large δV may be reliable for the cases discussed above, there is no way of determining whether the value obtained is in fact correct. If there is an unusually large error in one of the three widely spaced points, there will be a large error in the value of η obtained. Chanin and Rork[9] did not evaluate I_0, γ, and V^* from their data, and therefore could not have known whether their tabulated values of η are reliable.