

## Experimental Determinations of the First Townsend Ionization Coefficient in Helium

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The first Townsend ionization coefficient  $\alpha/p_0$  ( $\text{cm} \times \text{Torr}$ )<sup>-1</sup> has been experimentally determined for an  $E/p_0$  (volts/cm  $\times$  Torr) range from 3 to 300 in cataphoretically cleaned helium. The results obtained have been compared with several experimental and theoretical determinations of  $\alpha/p_0$ . The present values are in agreement within the combined experimental errors with those of Davies, Jones, and Morgan. Comparison is also made with the experimental results of Townsend and MacCallum where good agreement is found except at their lowest  $E/p_0$  values, where the present results lie below their corresponding data. At low  $E/p_0$ , the present results agree reasonably well with the theoretical values of Phelps and Dunlop except at very low  $E/p_0$  where Dunlop's calculated values are considerably smaller than the present experimental results. The significance of the experimental method at very high  $E/p_0$  where equilibrium conditions do not prevail is also discussed.

### I. INTRODUCTION

DURING the past thirty years, numerous studies of the Townsend electron ionization coefficient have been reported in the literature.<sup>1</sup> While the majority of these investigations have been experimental in nature, there have been a number of attempts to calculate the ionization coefficient  $\alpha/p_0$  using basic data of other studies. Unfortunately, in the case of helium at low  $E/p_0$  where more is known about the behavior of the electrons, ions, and excited atoms than in any other gas and consequently theoretical calculations should be most reliable, there have been few experimental measurements<sup>2-4</sup> extending down to the energy range most amenable to theoretical estimates. There is also reason to suspect that the gas samples employed in some of these studies were contaminated. For these reasons the present studies were undertaken in an attempt to obtain reliable  $\alpha/p_0$  measurements in helium which had been carefully purified, and over as wide a range of  $E/p_0$  as possible. The present article describes the results of this investigation and compares the available theoretical values with the experimental data.

From the theory developed originally by Townsend,<sup>5</sup> and since elaborated on by a number of workers, the growth of prebreakdown currents in uniform fields as the result of ionization by electron collision and secondary electron processes is given by

$$\frac{I(x)}{I(0)} = \frac{\exp \alpha(x-x_0)}{1 - \gamma \{ \exp [\alpha(x-x_0)] - 1 \}}, \quad (1)$$

for  $x \geq x_0$ . Here  $x$  is the interelectrode spacing,  $x_0$  is related to the distance which the electrons must travel before

an equilibrium velocity distribution characteristic of the applied  $E/p_0$  (electric field to pressure ratio,  $p_0 = 273 p/T$ ) is obtained.  $I(0)$  is a small externally generated photoelectric current and  $\gamma$  is a generalized coefficient referring to electron production due to secondary processes. The above equation assumes that electron losses due to diffusion and attachment are negligible;  $\alpha$  and  $\gamma$  are independent of  $x$  for  $x \geq x_0$ ; and the electrons are in equilibrium with the applied electric field (for  $x \geq x_0$ ).

For a given value of  $E/p_0$  a plot of  $\ln I(x)$  as a function of  $x$  appears to be initially linear. In this linear range, measurements of the current as a function of  $x$  (maintaining  $E/p_0$  constant) yield, from the slope, a value of  $\alpha$  which assumes the  $\gamma$  term is negligible. This method of evaluating  $\alpha$  has often been used in the past but it has been shown<sup>6,7</sup> that erroneous values of  $\alpha/p_0$  are obtained unless the value of  $\gamma$  is extremely small. With increasing values of  $x$ , the  $\gamma$  term in the denominator becomes appreciable thus resulting in an upcurving in the  $\ln I$  curve and finally results in gas breakdown. In addition to measuring  $\alpha$  by the above method, Townsend and MacCallum have shown that  $\alpha$  may be calculated from any portion of the  $\ln I$  versus  $x$  curves without any assumptions concerning the magnitude of  $\gamma$ ,  $x_0$ , or  $I(0)$  by measuring three currents  $I_1, I_2, I_3$  corresponding to distances  $x_1, x_2, x_3$ . In the present study  $\alpha$  was evaluated from the expression

$$\exp(\alpha b) = \frac{I_3(I_2 - I_1)}{I_1(I_3 - I_2)}, \quad (2)$$

where  $b = x_3 - x_2 = x_2 - x_1$ . This is equivalent to the expression used by Kruithof and Penning.<sup>8</sup>

In the present studies  $\alpha/p_0$  values were calculated in both the linear and nonlinear portions of the  $\ln I$  versus  $x$  curves using the three-point method given by Eq.

<sup>6</sup> E. Jones and F. Llewellyn Jones, Proc. Phys. Soc. (London) **72**, 363 (1958).

<sup>7</sup> D. E. Davies and J. G. C. Milne, Brit. J. Appl. Phys. **10**, 301 (1959).

<sup>8</sup> A. A. Kruithof and F. M. Penning, Physica **3**, 515 (1936).

<sup>1</sup> An excellent summary of work in this field is given in L. B. Loeb, *Basic Processes of Gaseous Electronics* (University of California Press, Berkeley 1955), Chap. VIII.

<sup>2</sup> E. W. B. Gill and F. B. Pidduck, Phil. Mag. **23**, 837 (1912).

<sup>3</sup> J. S. Townsend and S. P. MacCallum, Phil. Mag. **17**, 678 (1934).

<sup>4</sup> D. Kenneth Davies, F. Llewellyn Jones, and C. G. Morgan, Proc. Phys. Soc. (London) **80**, 898 (1962).

<sup>5</sup> J. S. Townsend, *Electricity in Gases* (Oxford University Press, London, 1915).

(2). In order to determine for a given  $\ln I$  versus  $x$  measurement the approximate value of  $x_0$  the method of Gosseries<sup>9</sup> was often used in the present work. A more reliable method of determining if the electrons are in equilibrium with the field involves calculations of  $\alpha/p_0$  as a function of the number of collisions which they undergo in passing from the cathode to anode. These measurements are discussed in Sec. III.

For  $E/p_0 < 10$  V/cm $\times$ Torr, since the differences in current values are very small, considerable errors can result using the normal three-point method. Under these conditions Kruthof and Penning have shown that Eq. (1) becomes

$$d \ln I(x)/dx = \alpha(1+\gamma). \quad (3)$$

In the present measurements,  $\alpha/p_0$  values were calculated using Eq. (3) at very low  $E/p_0$ . Values of  $\gamma$  which were used were calculated at higher  $E/p_0$  using formula given by Davies and Milne,<sup>7</sup> then extrapolated to the  $E/p_0$  range of interest. Corrections to  $\alpha/p_0$  for the secondary emission coefficient were approximately 10%. In the course of the present study a number of  $\ln I$  versus  $x$  measurements were fitted using Eq. (1) and the calculated values of  $\alpha/p_0$  and  $\gamma$ . This procedure, however, was not considered reliable in view of the considerable errors associated with the determination of the value of  $\gamma$ , thus, a fit to the observed curve within several percent was not considered meaningful when  $\gamma$  could not be determined to within 50%.

In addition to experimental determinations  $\alpha/p_0$  may be evaluated from theoretical considerations. Dunlop<sup>10</sup> and Abdelnabi and Massey<sup>11</sup> have employed the method originally developed by Emeleus, Lunt, and Meek<sup>12</sup> to calculate values of  $\alpha/p_0$  corresponding to several values of  $E/p_0$  for the case of helium. From a consideration of the roles played by atomic and molecular ions, metastable atoms, and photons in the breakdown of noble gases, Phelps<sup>13</sup> has calculated their various contributions to  $\alpha/p_0$  for the case of helium. In Sec. III the present experimental values are compared with the various theoretical values.

## II. APPARATUS

A schematic drawing of the apparatus used in the present studies is shown in Fig. 1. The experimental tube is a modern version of the type which has been used previously by numerous investigators in studies of this nature. The tube has been described in detail elsewhere<sup>14</sup>; therefore, only a brief description will be given here. The tube was constructed from stainless steel and has two demountable gold wire gasket flanges

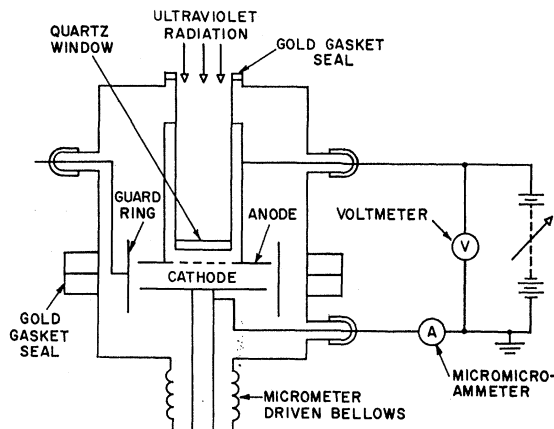


FIG. 1. Schematic diagram of the experimental tube and part of the associated circuitry.

to facilitate cleaning and assembly operations. The anode and cathode were made of nickel; the center portion of the anode consists of a fine nickel mesh (100 lines per inch) with an approximate 70% transparency. The electrode separation was adjustable from 0–2.5 cm, by means of a micrometer-driven bellows assembly with an accuracy of  $10^{-3}$  cm. Light from the ultraviolet source (Osram 100-W lamp) was introduced into the tube through an ultrapure silica window. Typical initial photoelectric current densities were  $\sim 10^{-11}$  A/cm<sup>2</sup>; reduction by a factor of 50 had no detectable effect on the measured values of  $\alpha/p_0$ . In the measurements  $I(0)$  found to be constant within 2%.

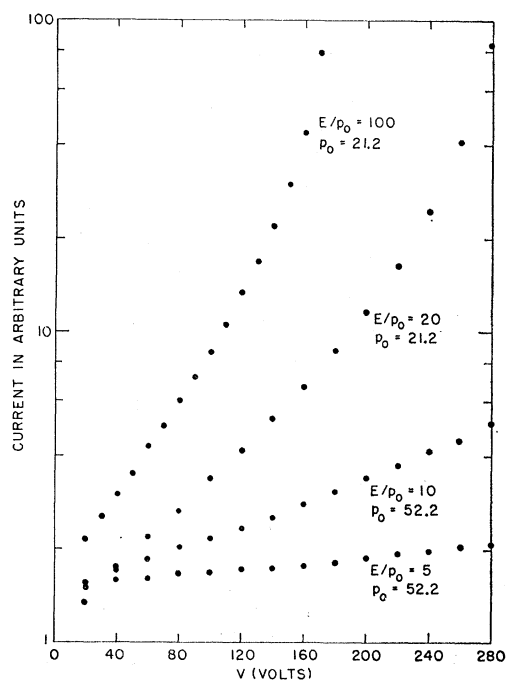


FIG. 2. Examples of  $\ln I$  versus  $V$  measurements at the indicated  $E/p_0$  values.

<sup>9</sup> A. Gosseries, *Physica* **6**, 458 (1939).

<sup>10</sup> S. H. Dunlop, *Nature* **164**, 452 (1949).

<sup>11</sup> I. Abdelnabi and H. S. W. Massey, *Proc. Roy. Soc. (London)* **A66**, 288 (1953).

<sup>12</sup> K. G. Emeleus, R. W. Lunt, and C. A. Meek, *Proc. Roy. Soc. (London)* **A156**, 394 (1936).

<sup>13</sup> A. V. Phelps, *Phys. Rev.* **117**, 619 (1960).

<sup>14</sup> L. M. Chanin and G. D. Rork, *Phys. Rev.* **132**, 2547 (1963).

The experimental tube was mounted on an ultrahigh vacuum gas handling system of the Alpert<sup>15</sup> type, which utilized bakeable vacuum valves and a capacitance type manometer.<sup>16</sup> The system was baked at temperatures of approximately 400°C for 15 h, following which the residual pressure was  $\sim 10^{-9}$  Torr with rates of rise of residual pressure  $\sim 10^{-10}$  Torr/min. The gas samples used in the study were Airco Reagent Grade helium. In measurements of this type, gas purity is of great importance. Of all of the gases, helium is perhaps the most sensitive to minor impurity contamination since, due to its high ionization potential, any impurity other than Ne will form a Penning mixture which has a strong influence on  $\alpha/p_0$  at low  $E/p_0$  values. In the present studies cataphoretic gas purification methods<sup>17,18</sup> were employed to remove minor impurity constituents. Prior to making measurements on a given gas sample, the helium was purified at pressures  $> 50$  Torr with currents  $> 70$  mA for periods of time exceeding 24 h. Since the cataphoretic efficiency is also apparently a function of cathode temperature, precautions were taken to prevent the cathode from achieving excessive temperatures. Evidence that purification resulted from the cataphoresis discharge (which was maintained during the measurements) could be easily demonstrated at low  $E/p_0$  values. Thus inadequate purification as the result of either too low a pressure, current, or too short a time, resulted in  $\alpha/p_0$  values considerably higher than those values obtained from gas samples purified using maximum cataphoretic efficiency.

### III. RESULTS

Figure 2 shows examples of measurements of  $\ln I$  versus  $V$  for various values of  $E/p_0$ . Measurements of the prebreakdown currents for a relatively large number

TABLE I. Comparison of experimental measurements.

$E/p_0$	Present pressure range investigated	$\alpha/p_0$		
		Present	Townsend and MacCallum	Davies <i>et al.</i>
3	52.2 - 75.8	0.00080	...	...
4	52.2 - 74.5	0.00298	...	...
5	29.0 - 52.2	0.00597	...	...
7	9.1 - 75.8	0.0161	...	0.017
10	9.1 - 74.5	0.0513	...	0.049
15	9.1 - 29.0	0.113	0.15	0.095
20	6.25 - 29.0	0.189	0.22	0.14
30	4.25 - 29.0	0.342	0.40	0.26
50	2.06 - 21.2	0.630	0.74	0.58
70	1.25 - 21.2	0.917	0.95	0.81
100	1.25 - 21.2	1.31	1.3	1.3
150	1.25 - 9.1	1.83	...	...
200	1.25 - 9.1	2.08	...	...
300	1.25 - 9.1	2.43	...	...

<sup>15</sup> D. Alpert, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 12.

<sup>16</sup> D. Alpert, *Rev. Sci. Instr.* **22**, 370 (1951).

<sup>17</sup> D. Riesz and C. H. Dieke, *J. Appl. Phys.* **25**, 196 (1954).

<sup>18</sup> A. M. Smeltkopf, Doctoral dissertation, University of Texas, Austin, Texas, 1962 (unpublished).

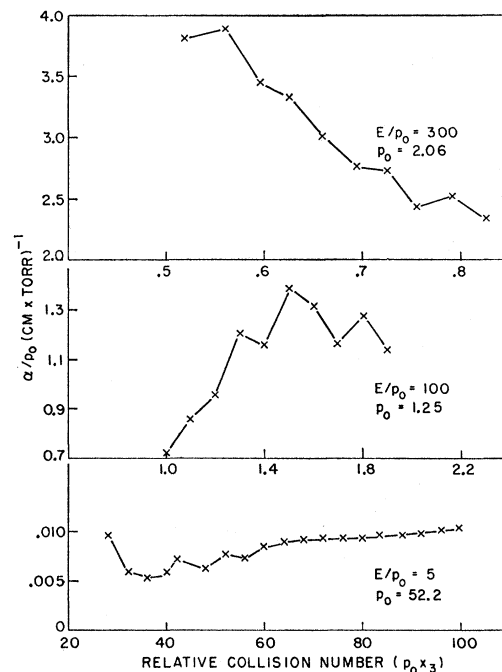


FIG. 3. Examples of the variation of  $\alpha/p_0$  with the relative electron collision number for the case of  $E/p_0=5, 100$ , and  $300$ .

of electrode separations permitted a comparison of  $\alpha/p_0$  values in the various regions of the  $\ln I$  versus  $x$  curves. In addition, such measurements permit  $\Delta x$  to be chosen as large as possible, such that the errors in the final evaluation of  $\alpha/p_0$  could be minimized. Maximum currents in the present studies were restricted to less than  $10^{-8}$  A to prevent serious alteration of the cathode surface due to ion bombardment and distortion of the electric field due to space charge effects. In the present measurements, for each  $E/p_0$  values investigated, a number of measurements of  $\alpha/p_0$  were made as a function of gas pressure. The range of pressures studied for a given  $E/p_0$  value is given in Table I.

To determine for a given  $\ln I$  versus  $V$  measurement the value of  $x$  such that  $x > x_0$ , a number of Gosseries type of plots were made. A more informative method of determining if the electrons are in equilibrium with the field is that of plotting  $\alpha/p_0$  as a function of the relative collision number. Figure 3 shows examples of this variation for the case of  $E/p_0=5, 100, 300$ . The relative number of collisions<sup>19</sup> which the electrons undergo is proportional to  $p_0 x$ , where  $x$  was taken to be that corresponding to  $x_3$  in the three-point analysis. The  $p_0$

<sup>19</sup> The actual number of collisions which they undergo in crossing the interelectrode separation  $x$  is given by  $\nu_e x / W_e$ , where  $\nu_e$  is the collision frequency and  $W_e$  is the electron drift velocity. In the present discussion we are concerned only with the variation of  $\alpha/p_0$  with the relative collision number. The determination of the actual collision numbers at high  $E/p_0$  values is difficult due to a lack of basic data. The approximate values of the maximum number of actual collisions for the cases shown in Fig. 3 are  $4 \times 10^4$ , 50 and 6 corresponding, respectively, to  $E/p_0=5, 100$ , and  $300$ .

values corresponding to the various  $E/p_0$  values are given in Fig. 3. For  $E/p_0=5$  it will be noted that initially  $\alpha/p_0$  fluctuates with increasing collision number until in the limit of a sufficient number of collisions  $\alpha/p_0$  becomes relatively constant. Variation of  $\alpha/p_0$  with collision number similar to those given in Fig. 3 have previously been observed by Kruthof and Penning<sup>20</sup> for the case of neon-argon mixtures.

From Fig. 3 it will be noted that for the case of  $E/p_0=100$ , initially  $\alpha/p_0$  varies considerably; however, with increasing collision number  $\alpha/p_0$  exhibits a damped type of oscillatory behavior. For  $E/p_0$  values between 5 and 100, the variation of  $\alpha/p_0$  with collision number is intermediate between that depicted in Fig. 3. For the case of  $E/p_0=300$  it is observed that  $\alpha/p_0$  decreases strongly with increasing collision number. The oscillatory behavior<sup>21</sup> may be the result of a fluctuating or changing electron energy distribution. The marked decrease in  $\alpha/p_0$  with collision number as shown for  $E/p_0=300$  in Fig. 3 is interpreted as being due to the electrons not having achieved equilibrium with the applied electric field. From a series of measurements such as shown in Fig. 3 we conclude that the electrons are not in equilibrium with the field for  $E/p_0 > 100$  V/cm  $\times$  Torr. In the course of the studies, measurements on different gas samples at varying pressures for a fixed  $E/p_0$  value produced the same general variation of  $\alpha/p_0$  with collision number. This clearly would not have been the case if the variation had, for example, been the result of field nonuniformities for small electrode separations. The same general variation observed in the present studies has recently been observed for the case of neon and hydrogen.<sup>14</sup> It should be noted that the measurements given in Fig. 3 serve only to indicate qualitatively if the electrons are in equilibrium with the applied field. Thus, since errors in  $\alpha/p_0$  vary inversely with  $\Delta x$ , and since necessarily in plots such as shown in Fig. 3  $\Delta x$  must be chosen to be relatively small, considerable errors may result. In view of this for a given  $\ln I$  versus  $V$  curve, in the calculations,  $\Delta x$  was maintained constant.

From Fig. 3 it will, however, be noted that for the case of  $E/p_0=300$ , where nonequilibrium conditions prevail, the magnitude of the decrease of  $\alpha/p_0$  ( $\sim 60\%$ ) is greater than the estimated error ( $\sim 10\%$ ) for such measurements.

Figure 4 shows a plot of experimental values of  $\alpha/p_0$  as a function of  $E/p_0$ . For comparison with the present results the data of Townsend and MacCallum<sup>22</sup> and

<sup>20</sup> A. A. Kruthof and F. M. Penning, *Physica* 4, 430 (1937).

<sup>21</sup> In principle, a correlation of the oscillatory behavior of  $\alpha/p_0$  with a given voltage or electron energy would be highly desirable. In practice, such a correlation is made difficult due to the fact that in using the three-point analysis  $\alpha/p_0$  is evaluated over a range of voltages.

<sup>22</sup> The data of Townsend and MacCallum used for comparison were corrected to the normalized pressure  $p_0$  assuming a room temperature of 300°K. Their  $\alpha/p_0$  values at given  $E/p_0$  were determined from an average curve drawn through their corrected values.

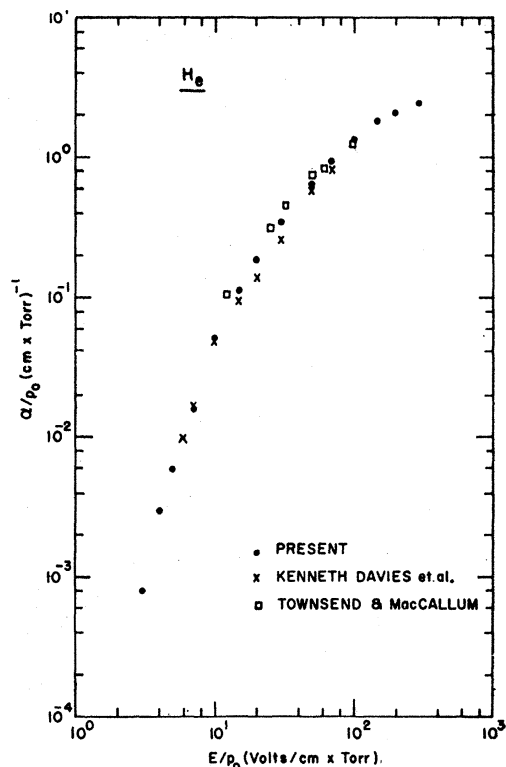


FIG. 4. Comparison of various experimental values of  $\alpha/p_0$  as a function of  $E/p_0$ .

Davies *et al.* are also given. The present values for  $E/p_0 < 100$  shown in Fig. 4 represent average values using the three-point method with  $\Delta x$  chosen to be as large as possible, with the additional restriction that  $x_1 > x_0$  as determined from Gosses type of plots. As previously noted for  $E/p_0 > 100$ , nonequilibrium conditions prevail; thus, the validity of the data is highly questionable. It will be noted that the present results agree within the combined experimental errors with those of Davies *et al.* over the common range of  $E/p_0$ . At low  $E/p_0$  the data of Townsend and MacCallum lie above the present results. It is believed that this is probably due to impurities in their gas samples. Previous measurements by Gill and Pidduck are generally considered to have been contaminated with mercury. Their values (not shown in Fig. 4) lie considerably above any of the previously discussed measurements. As previously pointed out by Loeb, the functional form of  $\alpha/p_0$  versus  $E/p_0$  is complicated due to the fact that the shape of the asymptotic foot of the electron-energy distribution alters radically with  $E/p_0$ . Therefore no effort has been made to express  $\alpha/p_0$  as an analytical function of  $E/p_0$ . The data shown in Fig. 4 are given in tabular form in Table I.

A comparison of the present experimental values with the available theoretical values is given in Fig. 5. The data given in Fig. 5 are presented in tabular form in Table II. The values calculated by Dunlop were ob-

tained using the form of the energy distribution function obtained by Smit,<sup>23</sup> probability of ionization data of Smith,<sup>24</sup> and adopted values of drift velocity approximating experimental values obtained by Nielsen.<sup>25</sup> The values given by Abdelnabi and Massey are those calculated using an extension and refinement of Smit's earlier work. Phelps' values were obtained<sup>26,27</sup> using the theoretical results of Reder and Brown<sup>28</sup> (assuming ac and dc distribution functions identical), unpublished excitation cross section data, and electron energy distributions given by Smit, Abdelnabi and Massey, and Reder and Brown.

It will be noted that the present values are in quite good agreement over the major portion of the  $E/p_0$  range with the various theoretical values. For very low  $E/p_0$  the present values are in better agreement with those of Phelps than Dunlop. The values calculated by Phelps may be more reliable than those given by Dunlop, in view of the fact that more reliable electron drift velocity<sup>29</sup> and excitation cross section data were used in these calculations. At the lowest  $E/p_0$  value investigated there exists a considerable discrepancy between the experimental value and that calculated by Dunlop. At present, the reason for this discrepancy is not clear; however, several possibilities are apparent. In view of the small values of  $\alpha/p_0$  for this range of  $E/p_0$ , the corresponding errors in the measurements are considerably greater than at higher  $E/p_0$ . Moreover, if in correcting  $\alpha/p_0$  for the secondary emission coefficient,<sup>30</sup> there is a

TABLE II. Comparison of present results with theoretical values.

Present experimental values		Theoretical values					
$E/p_0$	$\alpha/p_0$	Phelps		Dunlop		Abdelnabi and Massey	
$E/p_0$	$\alpha/p_0$	$E/p_0$	$\alpha/p_0$	$E/p_0$	$\alpha/p_0$	$E/p_0$	$\alpha/p_0$
3	0.00080	4.4	0.00308	3	0.00007	11	0.036
4	0.00298	6.6	0.0149	4	0.001	22	0.17
5	0.00597	8.8	0.0322	5	0.0042		
7	0.0161	11.0	0.0541	6	0.0077		
10	0.0513	13.2	0.0682	8	0.024		
20	0.189	17.6	0.113				
30	0.342	19.8	0.147				
50	0.630	22.0	0.204				

<sup>23</sup> J. A. Smit, *Physica* **3**, 543 (1936).

<sup>24</sup> P. T. Smith, *Phys. Rev.* **36**, 1293 (1930).

<sup>25</sup> R. A. Nielsen, *Phys. Rev.* **50**, 950 (1936).

<sup>26</sup> A. V. Phelps (private communication).

<sup>27</sup> The values of Phelps and Abdelnabi and Massey have been corrected to the corresponding  $p_0$  values assuming a room temperature of 300°K.

<sup>28</sup> F. H. Reder and S. C. Brown, *Phys. Rev.* **95**, 885 (1954).

<sup>29</sup> A. V. Phelps, J. L. Pack, and L. S. Frost, *Phys. Rev.* **117**, 470 (1960).

<sup>30</sup> The value of  $\gamma$  used in Eq. (3) to determine  $\alpha/p_0$  values for  $E/p_0 < 10$  was 0.08.

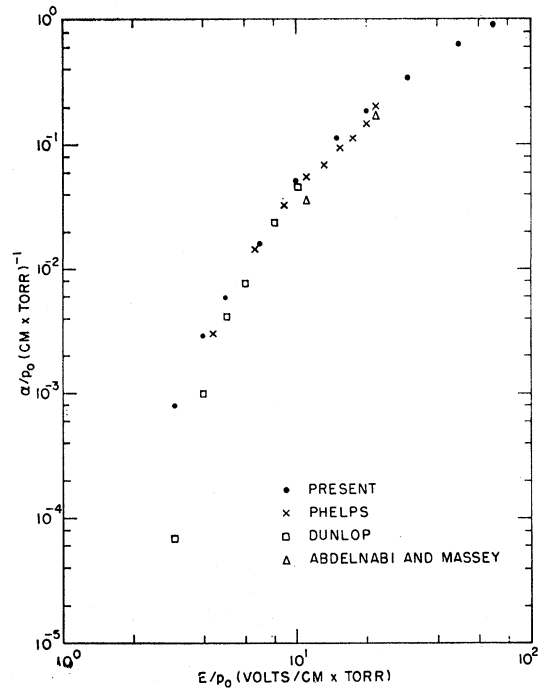


FIG. 5. Comparison of the present values of  $\alpha/p_0$  with the various theoretical results.

pronounced increase in  $\gamma$  going from high to low  $E/p_0$ , thus, invalidating a linear extrapolation (as was performed in the present studies), this would have the effect of reducing the observed discrepancy. A behavior of  $\gamma$  similar to this has been recently reported at low  $E/p_0$  values in the case of argon.<sup>31</sup> Finally, in recent years, it has been suggested<sup>32,33</sup> that Maier-Leibnitz's<sup>34</sup> cross-section data which were used in Smit's analysis, may be subject to errors arising from the complicated geometry of his apparatus. Confirmation of these ideas has very recently resulted from the measurements of Fleming and Higginson.<sup>35</sup>

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<sup>31</sup> D. E. Golden and L. H. Fisher, *Phys. Rev.* **123**, 1079 (1961).

<sup>32</sup> L. W. Kerr, thesis, University of Belfast, 1950 (unpublished).

<sup>33</sup> B. T. McClure (private communication).

<sup>34</sup> H. Maier-Leibnitz, *Z. Physik* **95**, 499 (1933).

<sup>35</sup> R. L. Fleming and G. S. Higginson, Sixth International Conference on Ionization Phenomena in Gases, Paris, June 1963 (unpublished).