Microwave Investigation of the Transition from Ambipolar to Free Diffusion in Afterglow Plasmas

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The time decay of electron density in pulsed helium afterglow discharges is studied experimentally employing both conventional and modified microwave-cavity techniques. The two methods permit density measurements from $n_e \simeq 10^{11}$ cm^{-3} to as low as $n_e \simeq 2\times10^{4}$ cm^{-3} in helium at pressures of 0.4 and 4.0 Torr. For Debye lengths less than $\sim$1% of the characteristic diffusion length Λ, the electron loss rate is through ambipolar diffusion controlled by the atomic ion He^+ at 0.4 Torr and by the molecular ion He_2^+ at 4.0 Torr. At lower electron densities, the electrons diffuse more rapidly than ambipolar diffusion, and in the limit of Debye lengths much greater than the characteristic diffusion length, the electrons diffuse freely to the walls. The experimentally observed effective diffusion coefficient D_a in the transition region is compared with a mathematical expression proposed by Phelps relating D_a to n_e. Satisfactory agreement is obtained at 4.0 Torr where the electron mean free path $\lambda \ll \Lambda$. By using this expression to calculate theoretically expected decays of electron density, a computer optimization procedure produced an even better fit to experimental data by slightly altering a numerical parameter in the formula. At 0.4 Torr, where $\lambda \approx \Lambda$ and the theoretical treatment is not expected to be valid, diffusion in the transition region is observed to occur at a rate substantially less than that predicted theoretically.

I. INTRODUCTION

THE diffusion loss of electrons in a gaseous discharge is dependent upon the relative influence of concentration gradients and space-charge effects.[1] When the Debye length is much less than container dimensions (high electron density), a space-charge field is created by unequal electron and ion free diffusion rates. This field tends to pull the slower ions towards the container wall and correspondingly retards the electron flow, with the result that both proceed to the wall at the same ambipolar diffusion rate.[2] When the Debye length substantially exceeds the container dimensions (low electron densities) the space-charge fields are negligible and both electrons and ions diffuse freely to the walls. Thus in an afterglow plasma the electron density is initially controlled by the ambipolar diffusion rate, whereas late in the afterglow the electron loss rate is substantially increased as electron diffusion tends toward the free limit. In the transition region between ambipolar and free diffusion the loss rate depends nonlinearly upon electron concentration; this behavior has been described in detail by several theoretical analyses.[3–8] Recently, Phelps[9] has suggested a closed mathematical expression relating the effective diffusion coefficient D_a to electron density, based in particular upon the theoretical treatments of Allis and Rose[3] and Cohen and Kruskal.[4]

In the present study conventional microwave-cavity techniques[10] are employed to measure the electron-density decay during the ambipolar diffusion regime of helium afterglows at 0.4 and 4.0 Torr. As the electron density decreases below $\sim10^{8}$ cm^{-3} in the transition region, a modified microwave cavity and synchronous detection scheme[11] is used to measure the electron-density decay to as low as $n_e \simeq 2\times10^{4}$ cm^{-3}, only a few orders of magnitude above the concentration at which free diffusion occurs. The combined diagnostic system, capable of measuring electron densities over some seven orders of magnitude, provides an experimental comparison of electron diffusion rates from the ambipolar region to well into the transition regime. The experimental tehcnique is described in Sec. II, and in Sec. III the electron-density decay in helium afterglows is discussed. Measured ionic mobilities are compared to those determined by other investigators, and the observed behavior in the transition diffusion regime is compared with theoretical descriptions as summarized by the mathematical relationship of Phelps. The implications of these comparisons are discussed in Sec. IV.

II. EXPERIMENTAL PROCEDURE

The helium discharge was produced in an 8-mm-i.d., 35-mm-long quartz tube by a voltage pulse of 7.5-μsec duration repeated at a 90-Hz rate. These tube dimen-

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[2] W. Schottky, Physik Z. **25**, 635 (1924).
[3] W. P. Allis and D. J. Rose, Phys. Rev. **93**, 84 (1954).
[4] I. M. Cohen and M. D. Kruskal, Phys. Fluids **8**, 920 (1965).
[5] L. E. Belousova, Zh. Tekhn. Fiz. **35**, 475 (1965) [English transl.: Soviet Phys.—Tech. Phys. **10**, 369 (1965)].
[6] D. Ecker, Proc. Phys. Soc. (London) **67**, 485 (1954).
[7] R. G. Fowler, Proc. Phys. Soc. (London) **80**, 620 (1962).
[8] T. Holstein, Phys. Rev. **75**, 1323 (1949).
[9] A. V. Phelps, in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Co., New York, 1966), p. 538; see also, A. V. Phelps and R. D. Hake (unpublished).
[10] D. J. Rose and S. C. Brown, J. Appl. Phys. **23**, 1028 (1952).
[11] L. A. Weaver and R. J. Freiberg (unpublished).

sions yield a characteristic diffusion length

$$\Lambda=[(2.405/r)^2+(\pi/l)^2]^{-1/2} \quad (1)$$

of $\Lambda=0.166$ cm, where r and l are the tube radius and length, respectively. The discharge tube was exhausted to $\sim 10^{-7}$ Torr with a conventional bakeable high-vacuum system and filled with spectroscopically pure helium. No attempt was made to further purify the helium gas by using cataphoretic separation techniques.[12] Thus the absolute determination of helium ionic diffusion rates is expected to be slightly in error due to neon impurities in the gas samples.[13] However, the nature of the transition from ambipolar to free electronic diffusion should be unaffected by the ionic species present, providing the ionic identity does not change with time and accurate experimental ambipolar rates are used to compare theory and experiment.

Electron number density was measured by microwave-cavity techniques with the discharge tube located along the axis of a demountable cylindrical C-band microwave cavity excited to the TM_{010} mode, as shown schematically in Fig. 1. The loaded cavity Q was ~ 5500, and the resonant frequency without a discharge was 4950 MHz. For electron densities greater than $n_e \simeq 8\times 10^8$ cm^{-3} the shift in cavity resonant frequency due to the presence of the plasma was measured by operating the klystron cw and allowing the decaying electron density to sweep through the loaded cavity resonance. A square-law microwave crystal was used as a detector, and the microwave power level was kept low enough ($\lesssim 10\,\mu$W) so that the plasma was not disturbed by the probing signal. To measure lower densities, a sensitive pulse-stretching and synchronous detection scheme was employed while sampling the microwave-cavity fields twice during each afterglow period.[11] This experimental arrangement is illustrated in Fig. 1. For a given microwave frequency near the cavity resonance the crystal detector voltage is dependent upon the complex conductivity of the plasma along the cavity axis. As the electron density decays with time, this Lorentzian cavity resonance shifts to lower frequencies and the Q increases slightly, with a consequent change in the crystal detector output. The microwave signal is sampled electronically for 0.5 μsec once during the afterglow period of interest and once during the no-plasma period at a 180-Hz rate. Thus the sampled signal for a constant probing frequency is a pulse train of alternating amplitudes whose difference in successive heights is related to the complex conductivity of the afterglow plasma. For a high-Q cavity this difference is primarily due to resonant frequency shifts, so that Q changes may be neglected by comparison.[14] By electronically stretching the pulse lengths of this train to a full 50% duty cycle,

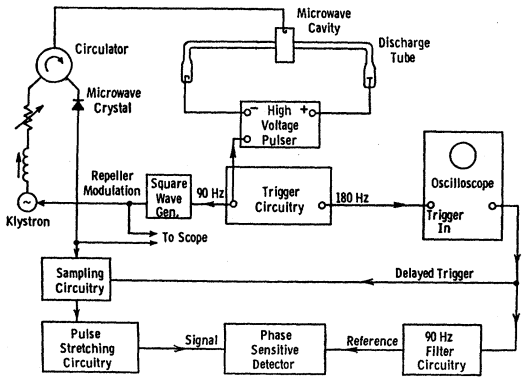

FIG. 1. Schematic diagram of the conventional microwave-cavity diagnostic system and the modified cavity apparatus used for measuring electron-density decay with time.

a 90-Hz square wave locked in time phase to the afterglow discharge is generated. Phase-sensitive detection of this square wave then yields a signal directly related to the plasma-induced resonant frequency shift. This shift may be measured by synchronously upshifting the microwave probing frequency by an equal amount to exactly compensate for the upshift in cavity resonant frequency caused by the plasma. A null in the phase-sensitive detector reading indicates complete source frequency compensation. The klystron source was frequency modulated by applying a small-amplitude 90-Hz square wave to the repeller, with care being taken that no amplitude modulation of the microwave level was produced. Calibration of the repeller modulation signal against the klystron frequency shift provided a direct measurement of the plasma-induced cavity resonance shift under compensated conditions. The combination of phase-sensitive detection and signal nulling techniques provides a greatly improved sensitivity in the determination of cavity resonance shifts, and permits the measurements of afterglow densities in helium to as low as $n_e \simeq 2\times 10^4$ cm^{-3}.[15] In the electron density region between $\sim 8\times 10^8$ and $\sim 3\times 10^9$ cm^{-3} where both conventional and modified microwave-cavity techniques were applicable, good agreement between the two methods of measurement was obtained.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The measurement of electron-density decay rates in helium afterglow discharges has been employed by several investigators[13,16–18] to determine diffusion rates and ionic mobilities in the ambipolar regime. Ion-transit-time experiments have provided independent

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[13] H. J. Oskam and V. R. Mittelstadt, Phys. Rev. **132**, 1435 (1963).
[14] L. A. Weaver and R. J. Freiberg, J. Appl. Phys. **39**, 1550 (1968).
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measurements of these quantities,[19–22] and there now exists general agreement on the mechanism and rate of ambipolar diffusion in low-pressure helium. Above ~ 3.5 Torr the ambipolar loss rate is controlled by diffusion of the molecular ion He_2^+, whereas at lower pressures the atomic ion He^+ determines the ambipolar diffusion rate.[13] Using ion-transit-time techniques, Madson, Oskam, and Chanin[22] have reported the reduced ionic mobilities as $\mu_0(He^+)=10.7$ cm^2/V sec and $\mu_0(He_2^+)=16.6$ cm^2/V sec, in good agreement with values of 10.4 and 16.7 cm^2/V sec reported previously be Beaty and Patterson[21] from ion-transit-time experiments. Electron-density decay-rate measurements by Kerr and Leffel[18] and Oskam and Mittelstadt[13] have both yielded values of $\mu_0(He^+)=10.7$ cm^2/V sec and $\mu_0(He_2^+)=16.2$ cm^2/V sec, in close agreement with ion-transit-time results. In each of these experiments cataphoretically purified helium samples were used.

In the present work electron-density decay with time was measured using a C-band microwave cavity. The experimental points in Fig. 2 illustrate this decay for a pulsed afterglow discharge in helium at 0.4 Torr. At this pressure the decay is expected to be an exponential function of time for $\lambda_D \ll \Lambda$ whose rate is determined by the atomic helium ion ambipolar diffusion coefficient $D_a(He^+)$. For electron densities greater than $n_e \sim 10^9$ cm^{-3} the experimental points lie along a straight line on the semilogarithmic plot, yielding a decay constant $\tau = 22.6$ μsec. The ambipolar diffusion coefficient D_a is given by

$$D_a = \Lambda^2/\tau, \qquad (2)$$

or $D_a = 1.23\times 10^3$ cm^2/sec over the linear portion of the curve. This corresponds to a reduced mobility $\mu_0(He^+)$ $=11.4$ cm^2/V sec, which is slightly higher ($\sim 8\%$) than the generally accepted value of 10.2 to 10.7 cm^2/V sec for the atomic helium ion.[13,22] Since the helium was not cataphoretically purified, a more rapid ambipolar rate would be expected due to the presence of impurity ions, particularly neon.[13] This result is consistent with earlier measurements by other investigators[16,17] in helium of similar purity, where even higher values were obtained.

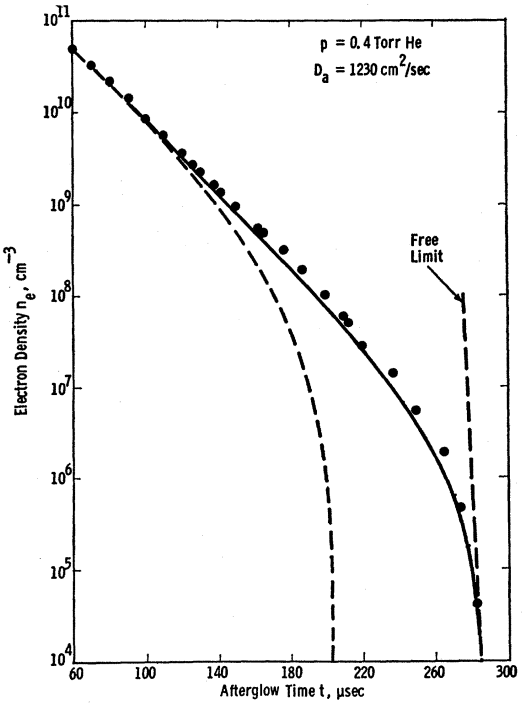

FIG. 2. Electron-density decay with time in helium at $p=0.4$ Torr. Experimental points describe the observed decay, and the dotted curve shows the theoretically predicted decay for $T_-=300$°K. The solid curve is the theoretically predicted decay for $T_-=12$°K and the power 0.92 used in the mathematical expression describing D_a.

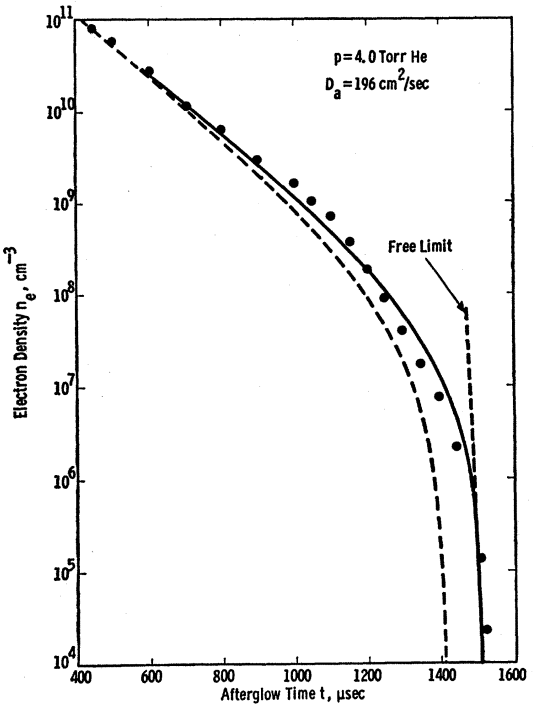

FIG. 3. Electron-density decay with time in helium at $p=4.0$ Torr. Experimental points describe the observed decay, and the dotted curve shows the theoretically predicted decay for $T_-=300$°K using the power 0.78 in the mathematical expression describing D_a. The solid curve is the theoretically predicted decay for $T_-=300$°K and the power 0.78 changed to 0.92.

The experimental points in Fig. 3 describe the measured electron-density decay with time in helium at 4.0 Torr. In this pressure regime the ambipolar rate is determined by the molecular ion He_2^+, and the slope of the linear portion of the curve above $n_e \sim 10^9$ cm^{-3} yields $D_a = 1.96\times 10^2$ cm^2/sec, which corresponds to a reduced

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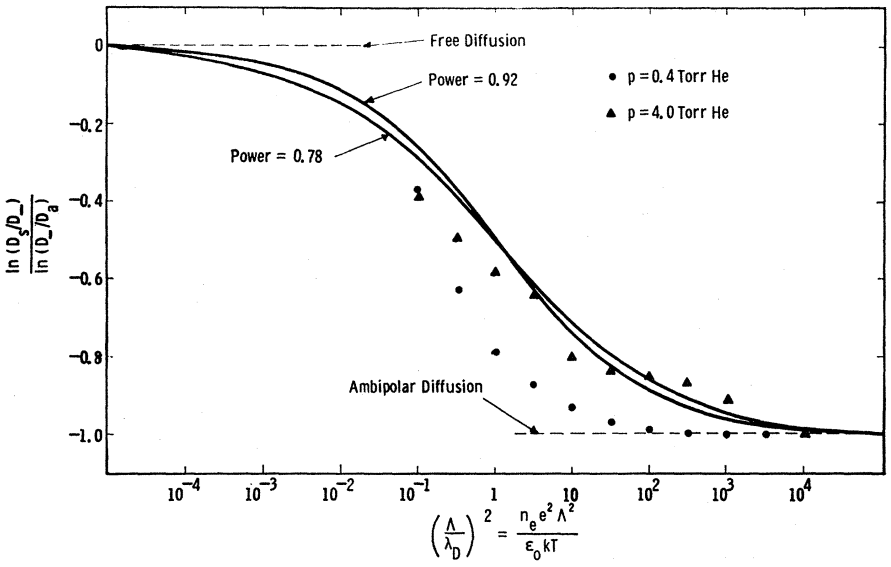

FIG. 4. Graph showing the relationship between the effective diffusion coefficient D_s and the electron number density n_e. The solid curves represent Phelps's expression in Eq. (4) using the mathematical parameters 0.78 and 0.92. The experimental points represent values of D_s determined graphically from the experimentally observed electron-density decay in helium at 0.4 and 4.0 Torr.

mobility of $\mu_0(He_2^+)=18.0$ cm^2/V sec. This is slightly higher ($\sim 8\%$) than the value 16.7 cm^2/V sec reported by other workers, and is evidence of the presence of impurity ions in the afterglow discharge. However, it is the electron-density range below $n_e \sim 10^9$ cm^{-3} in Figs. 2 and 3 which is of particular interest, for here the decay departs markedly from a pure exponential and approaches free-electron diffusive decay at densities below $n_e \sim 10^3$ cm^{-3}. The free-electron diffusion rate D_- in helium has been measured[23] as $D_- = 5.17 \times 10^5$ cm^2/sec at $p=0.4$ Torr and $D_- = 5.17 \times 10^4$ cm^2/sec at $p=4.0$ Torr; the asymptotic values of electron-density decay corresponding to these free diffusion rates are shown in Figs. 2 and 3 for the lowest measured densities $n_e = 2 \times 10^4$ cm^{-3}. It is seen that the experimental points describe a smooth transition from the ambipolar to the free diffusion limit as the electron density falls below $n_e \sim 10^8$ cm^{-3}. As space-charge effects become less important later in the afterglow the electron diffusion rate increases by a factor of 420 above the ambipolar rate at 0.4 Torr in approaching the free limit (a factor of 264 at 4.0 Torr). The manner in which this transition occurs may be described by defining an effective diffusion coefficient D_s such that for a given electron density n_e the instantaneous decay is exponential with a time constant

$$\tau_s = \Lambda^2/D_s. \tag{3}$$

Since the diffusion rate is itself a function of n_e, the exponential decay rate changes with n_e; thus D_s describes the instantaneous slope of the n_e-versus-t semilogarithmic plot.

The relationship between the effective diffusion coefficient D_s and the electron density n_e in the transition region has been studied theoretically by many authors.[3–8] Phelps[9] has recently fitted a closed mathematical expression to these theoretical predictions which agrees particularly well with the theories of Allis and Rose[3] and Cohen and Kruskal[4] over the transition region. The expression, valid for the electron mean free path much less than the container dimensions, is[24]

$$\frac{D_s}{D_-} = \left(\frac{D_a}{D_-}\right)^{[1+(\lambda_D/\Lambda)^{0.78}]^{-1}}, \tag{4}$$

where D_s, D_a, and D_- are the effective, ambipolar, and free diffusion rates, respectively, Λ is the characteristic diffusion length, and λ_D is the electron Debye shielding length

$$\lambda_D = (\epsilon_0 k T_-/n_e e^2)^{1/2}. \tag{5}$$

Here T_- is the electron temperature, n_e is the electron density, ϵ_0 is the permittivity of free space, k is Boltzmann's constant, and e is the electronic charge. Near the ambipolar limit where $(\lambda_D/\Lambda) \ll 1$, Eq. (4) may be approximated by

$$D_s \approx D_a[1+(\lambda_D/\Lambda)^{0.78} \ln(D_-/D_a)]. \tag{6}$$

For $(\Lambda/\lambda_D) \ll 1$ near the free diffusion limit, Eq. (4) may be expressed as

$$D_s \approx D_-[1-(\Lambda/\lambda_D)^{0.78} \ln(D_-/D_a)]. \tag{7}$$

Thus the mathematical description of D_s approaches D_a at high electron densities and D_- at low densities, yielding a value of

$$D_s = (D_a D_-)^{1/2} \tag{8}$$

midway through the transition regime at $\lambda_D = \Lambda$. For the conditions of this experiment, this occurs at $n_e = 5.19 \times 10^5$ cm^{-3} for $T_- = 300$°K. Equation (4) is plotted as the solid curve in Fig. 4 against the dimensionless variable $(\Lambda/\lambda_D)^2$.

[23] J. L. Pack and A. V. Phelps, Phys. Rev. **121**, 798 (1961).

[24] This expression was printed incorrectly in Ref. 9, and has been expressed here in terms of diffusion coefficients.

In order to compare the experimentally observed behavior with that predicted by Eq. (4), the measured instantaneous slopes of the electron-density decay curves on semilogarithmic paper were converted into effective diffusion coefficients for various densities in the transition region. These are shown in Fig. 4 as experimental points for $p=0.4$ Torr and $p=4.0$ Torr for an assumed electron temperature of 300°K. It is seen that for $p=0.4$ Torr the points lie reasonably close to the theoretical curve, although there is considerable scatter due to the difficulty of accurately determining slopes by graphical means. However, this deviation is no more than the spread of points corresponding to the various theoretical treatments to which the mathematical relationship was fitted. Thus the agreement can be considered quite satisfactory, particularly near the ambipolar limit. The 0.4-Torr data, however, show a significant deviation from the theoretical curve which is well beyond the experimental error in the measurements. The electron mean free path λ computed from momentum-transfer cross section data[25] is 0.14 cm at $p=0.4$ Torr and 0.014 cm at $p=4.0$ Torr, as compared to the characteristic diffusion length $\Lambda=0.166$ cm. In the transition diffusion theories considered here[3–9] it is assumed that the electron mean free path λ is much shorter than container dimensions, so that average charged-particle motions are determined by mobility and diffusion. This permits use of the boundary condition that charged-particle densities vanish at the confining walls. Thus the transition diffusion theories would not be expected to be strictly valid for 0.4-Torr data where $\lambda\approx\Lambda$, and is the likely explanation for the observed disagreement. The effects of diffusion cooling of the electrons[26] can also account for a part of this discrepancy. According to the mobility measurements of Oskam and Mittelstadt[13] this effect is quite evident in helium at pressures below 1.2 Torr. Estimates based upon these measurements indicate that $T_-\simeq 190$°K, and the arguments presented by Biondi[26] reveal that T_- could be as low as 60°K. By incorporating such lower electron temperatures[27] into the Debye length it is seen that the experimental data is moved towards closer agreement with the theoretical curve of Fig. 4. However, even for $T_-=60$°K, the experimental points do not lie close to the theoretical curve, indicating that the long electron mean free path at 0.4 Torr is largely responsible for the lack of agreement.

An alternate comparison between theory and experiment is obtained by using the transition diffusion relationship of Eq. (4) to generate the expected electron-density decay with time from a given set of initial conditions. A computer program was written to approximate the decay by a pure exponential over incrementally small time intervals. The effective diffusion coefficient D_a was recomputed for each new value of n_e, and the interval was reduced until only a negligible error was introduced by the exponential approximation. Using $D_a=1.96\times10^2$ cm²/sec, $D_-=5.17\times10^4$ cm²/sec, and $T_-=300$°K, the dotted curve of Fig. 3 for $p=4.0$ Torr was computed using Eq. (4). It is seen that the general behavior in the transition region is similar to that observed experimentally, except that the predicted density decays too rapidly, being some two orders of magnitude below experimental values at low densities. To obtain better agreement the power 0.78 was adjusted to provide the best fit to the experimentally observed decay. It was found that for a power of 0.92 in Eq. (4) good agreement was obtained, as shown by the solid curve of Fig. 3. The mathematical relationship of Eq. (4) is replotted in Fig. 4 for a power of 0.92, and it is noted that the curve deviates only slightly from the 0.78 curve. Thus it seems that the effective diffusion relationship of Eq. (4) tends to overestimate the diffusion coefficient slightly near the ambipolar limit. The adjustment of this expression to include the power 0.92 rather than 0.78 is questionable, however, since several theoretical treatments[4,8] indicate that this power should be more like $\frac{2}{3}$ near the ambipolar limit. Nonetheless the experimental evidence does indicate an upward adjustment of this power, or perhaps a more refined treatment of the theory is necessary to include cases where the electron mean free path begins to become comparable to container dimensions.[28–31]

The computer plot for 0.4 Torr is shown as the dotted curve in Fig. 2 for a power of 0.78. Here $D_a=1.23\times10^3$ cm²/sec, $D_-=5.17\times10^5$ cm²/sec, and $T_-=300$°K were used. The inadequacy of Eq. (4) in this case is quite evident. Using a power of 0.92 and $T_-=300$°K in Eq. (4) also gave poor agreement; it was found that a good fit could be obtained by assuming the power to be 0.92 and $T_-=12$°K, shown by the solid curve of Fig. 2. However, since $\lambda\approx\Lambda$ at this pressure little significance can be attached to this. It is apparent that the electrons are diffusion-cooled in helium at this pressure, but the inapplicability of the theoretical treatments for $\lambda\approx\Lambda$ undoubtedly accounts for the observed deviation. Thus the mathematical expression of Eq. (4) should be used with caution in this range; however, it adequately describes experimental behavior at higher pressures where $\lambda\ll\Lambda$, especially when the power 0.78 in the relationship is increased slightly, in this case to 0.92.

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[26] M. A. Biondi, Phys. Rev. **93**, 1136 (1954).
[27] Since the electron energy distribution under these circumstances is highly non-Maxwellian, the use of a "temperature" to describe the effects of diffusion cooling is not strictly accurate, but is employed here to illustrate the general effects of a reduced average electron energy upon the electron diffusion rate.
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[29] S. A. Self and H. N. Ewald, Phys. Fluids **9**, 2486 (1966).
[30] H. N. Ewald, F. W. Crawford, and S. A. Self, J. Appl. Phys. **38**, 2753 (1967).
[31] J. R. Forrest and R. N. Franklin, Brit. J. Appl. Phys. **17**, 1569 (1967).

IV. CONCLUSIONS

The decay of electron number density with time in helium afterglows is seen to depart from a simple exponential decrease as the electron Debye shielding length λ_D exceeds $\sim 1\%$ of the characteristic diffusion length Λ. For shorter Debye lengths the electron loss is through ambipolar diffusion with helium ions; at 0.4 Torr the ambipolar rate is controlled by the atomic ion He^+, whereas at 4.0 Torr the molecular ion He_2^+ determines the rate at which electrons diffuse to the walls. The measured values of reduced mobility for these species are slightly higher than recent values quoted in the literature. This discrepancy may be attributed to the fact that the helium used was not cataphoretically purified.

As the Debye length becomes much greater than the characteristic diffusion length it is seen that the electron loss rate approaches that of free-electron diffusion in both pressure ranges. Electron-density measurements in helium down to $n_e \simeq 2\times 10^4$ cm^{-3} ($\lambda_D/\Lambda \simeq 26$) demonstrate that at these low densities the electron loss rate has increased substantially above the ambipolar value and has virtually assumed the form of free-electron diffusion to the walls. In the transition regime between ambipolar and free diffusion the electron loss rate increases nonlinearly with increasing Debye length (decreasing electron density). This behavior has been summarized theoretically by Phelps in a closed mathematical relationship which provides a good fit to several of the more basic theoretical studies of transition diffusion. Experimentally observed effective diffusion coefficients show good agreement with this expression for a helium pressure of 4.0 Torr, where the electron mean free path is much less than container dimensions. At the lower pressure 0.4 Torr, where $\lambda \approx \Lambda$, the observed effective diffusion rate is substantially greater than that predicted by the theoretical relationship, which assumes that $\lambda \ll \Lambda$. Although the electron gas is likely diffusion cooled at this pressure, an assumed electron temperature as low as 60°K does not bring the theoretical diffusion rates into satisfactory agreement with experimental values.

By using the theoretical expression to compute the expected electron-density decay, it was found that better agreement with experimental results was obtained for $\lambda \ll \Lambda$ by adjusting the power 0.78 in this expression to 0.92. Even for $p = 4.0$ Torr the unmodified theory predicts too rapid a diffusion rate near the ambipolar limit, resulting in discrepancies of several orders of magnitude at low electron densities. The revised formulation yields satisfactory agreement with experimentally observed electron-density decay for the case in which $\lambda \ll \Lambda$. However, no reasonable modification of the theoretical expression provided satisfactory agreement for $\lambda \approx \Lambda$. Thus the general behavior of the transition from ambipolar to free diffusion in low-pressure helium afterglows has been established experimentally, and agreement with the form of current theoretical descriptions is good where the electron mean free path is small.

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