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Equilibration distance of ions in the cathode fall

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The motion of atomic ions in a rare-gas cathode fall is limited by symmetric charge exchange. Nonequilibrium solutions of the Boltzmann equation for this problem are presented. The distance required for the average ion velocity to approach within 10% of the equilibrium drift velocity is calculated for constant and for linearly increasing fields, with a plane ionization source and with a uniform source. The equilibration distances range from two-thirds to six mean free paths. A rare-gas cathode fall is typically 50—100 mean free paths thick; hence the ion motion may be accurately described by the equilibrium drift velocity throughout most of the cathode fall.

The cathode fall region of glow discharges is the least understood region and yet the most important region for many discharge applications. The primary difficulty in modeling the cathode fall is the failure of the electron distribution function to be in hydrodynamic equilibrium with the local E/N (electric field to gas density ratio). The lack of hydrodynamic equilibrium is caused by the large and rapidly changing E/N and by the proximity of the boundary. New theoretical approaches and recently developed optogalvanic diagnostics should soon lead to a more quantitative understanding of the cathode fall.[1-3] Accurate spatially resolved electric field and gas density (temperature) measurements are used to map the cathode fall region of rare-gas discharges.[2,3] The space-charge density, which is dominated by the positive-ion density, is determined from the spatial derivative of the electric field. The ions are assumed to be in hydrodynamic equilibrium with local E/N, and thus the drift velocity of the ions is determined from known ion mobilities and the local E/N. The product of the ion density and the ion drift velocity determines the ion-current density. The difference between the total discharge-current density and the ion-current density is the electron-current density. The spatial derivative of the electron-current density provides a map of the ionization rate in the cathode fall. This simple analysis is dependent on the assumptions: (1) that singly charged atomic ions are the dominant species and (2) that the ions are in hydrodynamic equilibrium. The first assumption must be experimentally verified, but it is very likely correct for low pressures (~ 1.0 Torr). Rare-gas molecular ions are likely to be dominant only at rather high pressures ($\gtrsim 100$ Torr). The distance required for the average velocity of atomic ions to approach within 10% of the equilibrium drift velocity is derived for four idealized cases in the following paragraphs. The equilibration distance for the ion velocity is two-thirds of a mean free path for a constant field with a plane ionization source, 4.5 mean free paths for a constant field with a uniform source, 1.7 mean free paths for a linearly increasing field with a plane source, and 5.7 mean free paths for a linearly increasing field with a uniform source. The latter two cases provide a lower and upper bound for the fraction of the cathode fall where the ion velocity is less than the equilibrium drift velocity. A normal rare-gas cathode fall has a thickness of approximately 50 to 100 mean free paths for symmetric charge exchange. The average ion velocity can be approximated as the equilibrium drift velocity throughout most all of the cathode fall.

The electric field in the normal and abnormal cathode fall is very definitely a function of position, but for the purpose of this initial discussion it will be assumed that the field is constant and is in the z direction. The mobility at high E/N of atomic ions in their parent gas is largely determined by symmetric charge exchange. Little momentum is transferred in these charge-exchange collisions. The symmetric charge-exchange-collision cross section is only weakly energy dependent. Wannier presented an elegant expression for the collision term of the Boltzmann equation in the approximation that (1) no momentum is transferred in the charge-exchange collision and (2) the cross section is independent of energy.[4] The Boltzmann equation including this collision term is

$$\frac{\partial f}{\partial t}+\frac{eE}{M}\frac{\partial f}{\partial v_z}+\mathbf{v}\cdot\nabla f = -N\sigma v f + N\sigma\delta(\mathbf{v})\int\int\int f v\, dv_z dv_x dv_y + P(z)\delta(\mathbf{v}) , \tag{1}$$

where e is the electric charge, M is the ion mass, σ is the charge-exchange cross section, and other symbols have their usual meanings. The last term which involves $P(z)$ is a source term which represents the production of "new" ions via electron impact ionization. Consider, as case 1, a plane source at the origin. The source function $P(z)$ is therefore $j\delta(z)$ where j is a constant. The time-independent nonequilibrium solution in which the ions start from rest at $z=0$ is

$$f_1 = j\delta(v_x)\delta(v_y)\{\exp(-\sigma N z)s(v_z)\delta(v_z^2/2-az) + (\sigma N/a)[s(v_z)-s(v_z-\sqrt{2az})] \times\exp[-v_z^2 N\sigma/(2a)]\} , \tag{2}$$

where a is the ion acceleration eE/M, and $s(v_z)$ is the step function of v_z. This solution for large z approaches

Wannier's equilibrium solution.[4] The distribution function given in Eq. (2) is not normalized in the usual sense because it is a nonequilibrium distribution. The integral

$$\int\int\int f_1 \mathbf{v}\, dv_z dv_x dv_y$$

is the particle flux density $j\hat{\mathbf{z}}$, which must be independent of position by conservation of particles. The integral

$$\int\int\int f_1 dv_z dv_x dv_y$$

is the particle density which is dependent on position because the ions are accelerating. The average velocity of the ions in the z direction is

$$\langle v_z \rangle = \frac{\int\int\int f_1 v_z dv_z dv_x dv_y}{\int\int\int f_1 dv_z dv_x dv_y} = \frac{1}{\exp(-\sigma N z)/\sqrt{2az} + \sqrt{\pi\sigma N/(2a)}\,\mathrm{erf}(\sqrt{\sigma N z})}, \quad (3)$$

where erf(x) is the error function.[5] The average velocity has the small-z limit of $\sqrt{2az}$ which is expected from kinematics, and it has the large-z limit of $\sqrt{2a/(\sigma N\pi)}$, in agreement with Wannier's equilibrium drift velocity. The average velocity reaches 90% of the equilibrium drift velocity in a distance $0.65/(\sigma N)$, or about two-thirds of a mean free path. Higher moments of the distribution function also approach their equilibrium values in short distances, but not as quickly as the average velocity.

The distribution function of Eq. (2) has been presented by several authors in discussion of ions in the cathode fall.[6–9] The distribution function has previously been misidentified as an energy distribution function; probably because $\int f_1 M v_z dv_z$ is position independent. The distribution function of Eq. (2) is here identified as a solution to the Boltzmann equation and, thus, is a velocity distribution function. An integral expression which can be used to derive a velocity distribution function for an arbitrary position-dependent field with a plane ionization source is given in Ref. 8. Nonequilibrium double-humped distribution functions for Ar^+ ions in Ar have been observed in a low-pressure drift-chamber experiment.[10]

A solution of the Boltzmann equation for a constant field with an arbitrary source function $P(z)$ is constructed using the plane-source solution as a Green's function. The distribution function for constant field with an arbitrary $P(z)$ is

$$f = \delta(v_x)\delta(v_y)\int_{-\infty}^{z} P(z_0)[\exp[-\sigma N(z-z_0)]s(v_z)\delta(v_z^2/2 - a(z-z_0)) + (\sigma N/a)\{s(v_z) - s[v_z - \sqrt{2a(z-z_0)}]\}\exp[-v_z^2 N\sigma/(2a)]]dz_0 \,. \quad (4)$$

Consider, as case 2, a uniform source for non-negative z. The source function $P(z_0)$ in Eq. (4) is replaced by $rs(z_0)$. The particle flux density

$$\int\int\int f_2 \mathbf{v}\, dv_z dv_x dv_z$$

is equal to $rz\hat{\mathbf{z}}$; it grows linearly with distance from the origin. The average velocity of the ions in the z direction is

$$\langle v_z \rangle = \frac{\int\int\int f_2 v_z dv_z dv_x dv_y}{\int\int\int f_2 dv_z dv_x dv_y} = \frac{z}{\sqrt{\pi/(2aN\sigma)}[\mathrm{erf}(\sqrt{\sigma N z})(\sigma N z + 1/2) + \sqrt{\sigma N z/\pi}\exp(-\sigma N z)]} \,. \quad (5)$$

The average velocity in case 2 approaches the equilibrium drift velocity of $\sqrt{2a/(\sigma N\pi)}$, but approaches more slowly than in case 1. The slower convergence is due to the production of new ions at rest via electron impact ionization at all $z \geq 0$. The distance required for the average ion velocity to reach 90% of the equilibrium drift velocity is 4.5 mean free paths in case 2.

Experimental studies of the electric field in the cathode fall indicate that the field increases with distance from the cathode-fall—negative-glow boundary. The field reaches a maximum at or near the cathode surface. The spatial dependence of the field has long been approximated as directly proportional to z, the distance from the cathode-fall—negative-glow boundary. Recent accurate electric field measurements using optogalvanic detection of Rydberg atoms support this simple spatial dependence of the field.[11] The Boltzmann equation with a linearly increasing field is

$$\frac{\partial f}{\partial t} + kz\frac{\partial f}{\partial v_z} + \mathbf{v}\cdot\nabla f = -N\sigma v f + N\sigma\delta(\mathbf{v})\int\int\int f v\, dv_z dv_x dv_y + P(z)\delta(\mathbf{v}) \,, \quad (6)$$

where kz is the ion acceleration. The source function $P(z)$ for case 3 is a plane source $j\delta(z-z_0)$ where $z_0 > 0$. The distribution function for $z \geq z_0$ is

$$f_3 = j\delta(v_x)\delta(v_y)[\exp[-\sigma N(z-z_0)]s(v_z)2\delta(v_z^2 - kz^2 + kz_0^2) + (\sigma N/\sqrt{k})\{s(v_z) - s[v_z - (kz^2 - kz_0^2)^{1/2}]\} \times \exp(\sigma N z\{[1 - v_z^2/(kz^2)]^{1/2} - 1\})/(kz^2 - v_z^2)^{1/2}] \,. \quad (7)$$

This distribution function for large z approaches Wannier's equilibrium distribution function[4]

$$f_{\mathrm{eq}} = j\delta(v_x)\delta(v_y)[\sigma N/(kz)]s(v_z)\exp[-v_z^2\sigma N/(2kz)] \,. \quad (8)$$

We need to specify z_0 in order to compute an equilibration distance for the average velocity. If $z_0 \gg (\sigma N)^{-1}$, then the field changes only slightly in a mean free path and the equilibration distance will be that of the constant field problem. The interesting case is for z_0 near zero. The average velocity in the z direction for this case is

$$\langle v_z \rangle = \frac{\int\int\int f_3 v_z dv_z dv_x dv_y}{\int\int\int f_3 dv_z dv_x dv_y} = \frac{1}{\exp(-\sigma Nz)/(\sqrt{k}z) + \pi\sigma N \exp(-\sigma Nz)[I_0(\sigma Nz) + L_0(\sigma Nz)]/(2\sqrt{k})}, \quad (9)$$

where $I_0(z)$ is a modified Bessel function of order 0 and $L_0(z)$ is a modified Struve function of order 0 as defined and tabulated in Refs. 12 and 13. The average velocity for this case reaches 90% of the equilibrium drift velocity $\sqrt{2kz/(\pi\sigma N)}$ in 1.7 mean free paths.

The plane-source distribution function for the linearly increasing field can be used as a Green's function to construct an ion distribution function for an arbitrary source function $P(z)$. The distribution function for a linearly increasing field with an arbitrary source function is

$$f = \delta(v_x)\delta(v_y) \int_0^z P(z_0)[\exp[-\sigma N(z - z_0)]s(v_z) 2\delta(v_z^2 - kz^2 + kz_0^2) + (\sigma N/\sqrt{k})\{s(v_z) - s[v_z - (kz^2 - kz_0^2)^{1/2}]\}$$
$$\times \exp(\sigma Nz\{[1 - v_z^2/(kz^2)]^{1/2} - 1\})/(kz^2 - v_z^2)^{1/2}] dz_0 . \quad (10)$$

If the source function of the cathode fall is known, then it is possible to calculate a fairly realistic distribution function and equilibration distance for the ions. Unfortunately, it is the source function we are proposing to map from accurate field and gas density (temperature) measurements by assuming that the ions are equilibrated. This seems to suggest that the Boltzmann equation for ions, the Boltzmann equation for electrons, and Poisson's equation must be solved simultaneously. Fortunately, a simultaneous self-consistent solution of these three equations is not necessary. The source function $P(z)$ is a decreasing function of z throughout the cathode fall. An electron avalanche starts from one electron emitted at the cathode and grows as z decreases. Thus we can compute an upper limit for the equilibration distance of the ions in the cathode fall by using a uniform source function for positive z. This is case 4. The average ion velocity for a linearly increasing field with a uniform source of ions at all positive z is

$$\langle v_z \rangle = \frac{\int\int\int f_4 v_z dv_z dv_x dv_y}{\int\int\int f_4 dv_z dv_x dv_y} = \frac{z}{\exp(-\sigma Nz)\pi\{I_0(\sigma Nz) + L_0(\sigma Nz) + \sigma Nz[I_1(\sigma Nz) + L_1(\sigma Nz) + 2/\pi]\}/(2\sqrt{k})} \quad (11)$$

where $I_1(z)$ and $L_1(z)$ are modified Bessel and modified Struve functions of order 1. The distance required for the average velocity to reach 90% of the equilibrium drift velocity $\sqrt{2kz/(\pi\sigma N)}$ is 5.7 mean free paths.

A normal rare-gas cathode fall is 50 to 100 mean free paths thick.[4,14] The ratio of 50 or 100 is independent of pressure because the product in Torr cm of pressure and thickness is constant for a normal cathode fall. The assertion that the ion velocity in a rare-gas cathode fall can be approximated by the equilibrium drift velocity is justified. The approximation fails within the first 6 mean free paths of the cathode-fall—negative-glow boundary. The approximation should be reliable for positions more than 6 mean free paths from the cathode-fall—negative-glow boundary. The collision term of the Boltzmann equation used in the preceding calculations is valid only for high field, but this should lead to an overestimate of the equilibration distance. Recent nonhydrodynamic calculations of electron kinetics suggest that the source function is fairly uniform near the cathode-fall—negative-glow boundary.[15]

It should be emphasized that the short equilibration distance is unique to the ions because of the symmetric charge exchange. The electrons in the cathode fall are not in hydrodynamic equilibrium. The nonequilibrium distribution of the electrons is a topic of high current interest for theoretical and experimental researchers.

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