Aust. J. Phys., 1974, **27**, 195–209

On the Theory of Electron Diffusion in Electrostatic Fields in Gases

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Abstract

The motion of electrons in a gas in the presence of large electron density gradients has been studied theoretically, starting from the two-term expansion of the Boltzmann equation. The effects of material boundaries have not been considered. An electron swarm released as a δ-function in space and with an equilibrium energy distribution is found initially to develop as a spheroid with dimensions determined by the lateral diffusion coefficient. It subsequently passes through a stage involving a slowly decaying pear-shaped deformation, before ultimately becoming an ellipsoid with dimensions determined by the longitudinal and lateral components of the diffusion tensor. Numerical values cited in the literature for the long-term deviations from the mean square widths predicted by the diffusion equation have been found to be in error by factors of 10 or more.

1. Introduction

The problem of electron motion in a gas in the presence of an electrostatic field and electron density gradients has been treated theoretically by a number of authors over the last five years (e.g. Parker and Lowke 1969; Skullerud 1969; Lucas 1970; Lowke 1971; Huxley 1972; McIntosh 1974; for a comprehensive review see Huxley and Crompton 1974). The interest in the subject has largely been due to the experimental observations by Hurst and Parks (1966) and Wagner *et al.* (1967) that the diffusion tensor describing the spread of an electron swarm is usually strongly anisotropic, in contrast to what had been generally assumed, and to the demand from experimentalists for a theory enabling an accurate description of the regimes with strong relative density gradients occurring in the vicinity of metal boundaries.

Satisfactory explanations of the observed anisotropic diffusion of an electron swarm were arrived at independently by Parker and Lowke (1969) and Skullerud (1969). The latter author obtained an expression for the diffusion tensor by using a perturbation method, originally suggested by Wannier (1953) in a study of ion motion in strong electric fields, while Parker and Lowke constructed a mathematical model of an electron swarm, initially released as a δ-function in space, and found the diffusion tensor from the asymptotic expression obtained for the mean square random displacement of the electrons.

A satisfactory theory, based on the Boltzmann equation, for the description of regimes with *strong* density gradients has apparently not yet been found.

Lucas (1970) tried to include the effects of strong gradients by assuming an energy distribution depending on the logarithmic derivative of the density. He used this implicit space dependence of the energy distribution to determine local values of the drift velocity and the diffusion coefficient for subsequent inclusion in the diffusion

equation. However, the assumed connection between the energy distribution and the logarithmic derivative of the density does not follow from a correct treatment of the Boltzmann equation, and the validity of the suggested (nonlinear) transport equation seems doubtful.

Huxley (1972) attempted a solution of the problem by assuming that the spatial dependence of the electron density distribution could be expressed by means of a series expansion using the gradient operator, acting upon the number density, as a formal expansion parameter. In order to eliminate the time derivatives from his equations, however, he made a further, inadmissible, assumption which erroneously led him to the conclusion that the ordinary (anisotropic) diffusion equation is exact, under rather general conditions.

McIntosh (1974) has performed a numerically exact investigation of the development of an electron swarm initially released as a δ-function in space, by simulating the motion of a large number of electrons on a digital computer. The actual model he used was the one of electrons interacting as hard spheres with the atoms of a zero-temperature gas. He observed a long-lasting pear-shaped deformation of the electron swarm, in qualitative agreement with what one would expect from Parker and Lowke's (1969) asymptotic expressions, and with predictions by Kumar and Robson (1973). Quantitatively, however, his results seem to disagree with those of Parker and Lowke, thus casting doubt upon the validity of their expressions beyond the first-order 'diffusion approximation'.

In the present work, it is first shown how a higher-order transport equation can be obtained by suitably combining elements from Huxley's (1972) work with suggestions made by Kumar and Robson (1973). At best, however, the equation can only be asymptotically correct and cannot be used to describe conditions near material boundaries. The problem of how an electron swarm develops when initially released as a δ-function in space, with a given energy distribution, is then examined. Using a generalized Hermite-polynomial expansion in the spatial coordinates, the Boltzmann equation for this problem can be transformed to a set of rather well-behaved parabolic equations, which can be solved numerically by standard methods. The effects of material boundaries are only discussed qualitatively.

2. Basic Theory

Although the form of the Boltzmann equation used in this work may be found in standard references (e.g. Shkarofsky *et al.* 1966), it will be useful for the discussion of the results to have a sketch of its derivation. A qualitative discussion of the diffusion process in terms of the velocity autocorrelation function (Kubo 1966) will also be given.

(*a*) *Two-term Boltzmann Equation*

The Boltzmann equation for the electron distribution function $f(\boldsymbol{r}, \boldsymbol{v}, t)$ may be written

$$\frac{\partial f}{\partial t} + \boldsymbol{v} \cdot \frac{\partial f}{\partial \boldsymbol{r}} + \boldsymbol{a} \cdot \frac{\partial f}{\partial \boldsymbol{v}} = C(f), \tag{1}$$

$C(f)$ being the collision integral and $\boldsymbol{a}$ the acceleration of an electron in an electrostatic field $\boldsymbol{E}$,

$$\boldsymbol{a} = -e\boldsymbol{E}/m. \tag{2}$$

The distribution function is expanded in spherical harmonics in velocity space,

$$f(\boldsymbol{r}, \boldsymbol{v}, t) = f_0(\boldsymbol{r}, v, t) + \boldsymbol{f}_1(\boldsymbol{r}, v, t) \cdot \boldsymbol{v}/v + \mathbf{f}_2(\boldsymbol{r}, v, t) : (3\boldsymbol{vv}/v^2 - \mathbf{1}) + \ldots . \tag{3}$$

When the expansion (3) is inserted into the Boltzmann equation (1), it is found that the relative magnitudes of the terms will be of the order (for not too large gradients or too small velocities)

$$f_l(v)/f_{l-1}(v) \sim (m/M)^{\frac{1}{2}}, \tag{4}$$

M being the mass of a gas molecule. This justifies the truncation of the expansion after two terms, leading to two coupled equations for f_0 and $\boldsymbol{f}_1$:

$$\frac{\partial f_0}{\partial t} + \tfrac{1}{3} v \frac{\partial}{\partial \boldsymbol{r}} \cdot \boldsymbol{f}_1 + \frac{1}{3v^2} \frac{\partial (v^2 \boldsymbol{a} \cdot \boldsymbol{f}_1)}{\partial v} = C_0(f_0), \tag{5}$$

$$\frac{\partial \boldsymbol{f}_1}{\partial t} + v \frac{\partial f_0}{\partial \boldsymbol{r}} + \boldsymbol{a} \frac{\partial f_0}{\partial v} = C_1(\boldsymbol{f}_1). \tag{6}$$

In order to reduce the complexity of this work, it will be assumed that only elastic collisions occur; C_0 can then be transformed to a differential form (Davydov 1935) and the collision integrals written as

$$C_0(f_0) = (m/M) v^{-2} \frac{\partial}{\partial v} \left\{ v^3 \nu \left(f_0 + \frac{kT}{mv} \frac{\partial f_0}{\partial v} \right) \right\}, \tag{7}$$

$$C_1(\boldsymbol{f}_1) = -\nu \boldsymbol{f}_1, \tag{8}$$

ν being the collision frequency for momentum transfer and T the gas temperature. It should be remarked that the Davydov expression for C_0 is only valid for speeds $v \gg (kT/M)^{\frac{1}{2}}$.

We now assume that the relative rate of change of $\boldsymbol{f}_1$ is small compared with the collision frequency,

$$|\partial \boldsymbol{f}_1/\partial t| \ll \nu |\boldsymbol{f}_1|, \tag{9}$$

and hence that the time derivative in equation (6) can be neglected. Equations (5) and (6) then give

$$\boldsymbol{f}_1 = -\frac{v}{\nu} \frac{\partial f_0}{\partial \boldsymbol{r}} - \frac{\boldsymbol{a}}{\nu} \frac{\partial f_0}{\partial v}, \tag{10}$$

$$\frac{\partial f_0}{\partial t} - \frac{v^2 \nabla^2 f_0}{3\nu} - \tfrac{2}{3} v \frac{\boldsymbol{a}}{\nu} \cdot \frac{\partial}{\partial \boldsymbol{r}} \left(\frac{\partial f_0}{\partial v} \right) - \frac{\partial f_0}{\partial \boldsymbol{r}} \cdot \left(\frac{1}{3v^2} \frac{\mathrm{d}(v^3 \boldsymbol{a}/\nu)}{\mathrm{d}v} \right)$$

$$= \frac{1}{3v^2} \frac{\partial}{\partial v} \left[v^2 \nu \left\{ \frac{3mv f_0}{M} + \left(\frac{a^2}{v^2} + \frac{3kT}{M} \right) \frac{\partial f_0}{\partial v} \right\} \right]. \tag{11}$$

Equation (11) is the starting point for the present work. Its range of validity might be loosely indicated as follows. The time intervals Δt, position intervals Δr and speed intervals Δv over which significant changes in f_0 occur must fulfill the conditions:

for equation (9) to be valid, $\qquad \Delta t \gg \nu^{-1} \equiv \tau_{\mathrm{m}}$, (12a)

where τ_m is the free time for momentum transfer;

for f_1 to be $\ll f_0$ (equation (10)),

$$\Delta r \gg v\tau_m = \lambda_m, \tag{12b}$$

$$\Delta v \gg a\tau_m, \tag{12c}$$

where λ_m is the free path for momentum transfer. Further,

for valid use of the Davydov term, $$v \gg (kT/M)^{\frac{1}{2}} \tag{12d}$$

are the only speeds that can be handled.

(*b*) *Diffusion and Autocorrelation*

Consider a system in a steady state, i.e. where the velocity distribution does not depend on time. Writing $\boldsymbol{c}$ for random velocity and $\boldsymbol{s}$ for random displacement,

$$\boldsymbol{c} = \boldsymbol{v} - \langle \boldsymbol{v} \rangle, \qquad \boldsymbol{s} = \boldsymbol{r} - \langle \boldsymbol{v} \rangle t = \int_0^t \boldsymbol{c}(t)\, \mathrm{d}t, \tag{13}$$

we obtain, after some manipulations involving the steady-state assumption

$$\langle \boldsymbol{c}(0)\, \boldsymbol{c}(t) \rangle = \langle \boldsymbol{c}(0)\, \boldsymbol{c}(-t) \rangle, \tag{14}$$

the following expression for the mean square random displacement in a time t (see e.g. Egelstaff 1967)

$$\langle \boldsymbol{ss} \rangle = 2t \int_0^t \langle \boldsymbol{c}(0)\, \boldsymbol{c}(\tau) \rangle (1 - \tau/t)\, \mathrm{d}\tau. \tag{15}$$

At sufficiently large times, the random velocity autocorrelation function will be zero, and the mean square displacement will be given by the expression

$$\langle \boldsymbol{ss} \rangle \underset{t}{\to} 2(\mathbf{D}t + \mathbf{A}). \tag{16}$$

The diffusion tensor $\mathbf{D}$ and constant $\mathbf{A}$ are obtained as

$$\mathbf{D} = \int_0^\infty \langle \boldsymbol{c}(0)\, \boldsymbol{c}(\tau) \rangle\, \mathrm{d}\tau, \qquad \mathbf{A} = -\int_0^\infty \langle \boldsymbol{c}(0)\, \boldsymbol{c}(\tau) \rangle\, \tau\, \mathrm{d}\tau. \tag{17a, b}$$

For electrons moving in a gas under the influence of an electrostatic field, the correlation functions will behave differently according to whether one considers motion normal to the field or in the field direction. In a direction normal to the field, say the x direction, the 'correlation time' will be of the order of a free time for momentum transfer τ_m, that is,

$$\langle c_x(0)\, c_x(t) \rangle = \langle c_x^2 \exp(-t/\tau_m) \rangle, \tag{18}$$

and the corresponding components of $\mathbf{D}$ and $\mathbf{A}$ will be

$$D_\perp \equiv D_{xx} = \langle c_x^2 \tau_m \rangle = \langle c^2/3\nu \rangle, \tag{19a}$$

$$A_\perp \equiv A_{xx} = -\langle c_x^2\, \tau_m^2 \rangle = -\langle c^2/3\nu^2 \rangle. \tag{19b}$$

In the field direction, say the z direction, the correlation function will in addition to the term (18) contain a term due to the coupling between energy and 'instantaneous drift velocity' (see equation (29b) below); this term has a correlation time of the order of a free time for energy transfer τ_e,

$$\tau_e = (M/2m)\tau_m, \tag{20}$$

and is the cause of the difference between the longitudinal diffusion coefficient $D_\parallel \equiv D_{zz}$ and the lateral diffusion coefficient $D_\perp$. In the A value, this term would be expected to give a contribution of the order of magnitude

$$A_\parallel \equiv A_{zz} \sim (D_\perp - D_\parallel)\tau_e \sim (M/m)A_\perp. \tag{21}$$

Owing to the limitations (12), the components $A_\perp$ would be expected to be lost when using the two-term expansion (10) and (11) of the Boltzmann equation, while $A_\parallel$ should be obtainable from a theory based on this expansion.

3. A Higher-order Transport Equation

When spatial derivatives of order higher than two are neglected, the transport equation describing the evolution of the electron density can be written (assuming a homogeneous gas and a homogeneous electric field)

$$\partial n/\partial t = -\boldsymbol{v}_d \cdot \nabla n + \mathbf{D} : \nabla\nabla n, \tag{22}$$

$\boldsymbol{v}_d$ being the drift velocity and $\mathbf{D}$ the diffusion tensor. Kumar and Robson (1973) point out that this can be looked upon as part of a more general multipole expansion

$$\partial n/\partial t = \sum_{i,j=0}^{\infty} \omega_{2i,j}(\nabla_\perp^2)^i(-\partial/\partial z)^j n \qquad \text{with} \quad \omega_{00} = 0, \tag{23}$$

where we have written $\partial/\partial z$ for the gradient operator in the direction of the acceleration $\boldsymbol{a} = -e\boldsymbol{E}/m$, and $\nabla_\perp$ for $\partial/\partial x + \partial/\partial y$. The expansion (23) is equivalent to a Fourier expansion of the density, assuming a single frequency $\omega(\boldsymbol{k})$ belonging to each wave number $\boldsymbol{k}$, and a power series expansion for the relation $\omega = \omega(\boldsymbol{k})$:

$$n(\boldsymbol{r}, t) = (2\pi)^{-3} \int \tilde{n}(\boldsymbol{k}) \exp\{\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r} - \omega(\boldsymbol{k})\, t\}\, \mathrm{d}^3k, \tag{24}$$

$$\omega(\boldsymbol{k}) = -\sum_{i,j} \omega_{2i,j}(-k_\perp^2)^i(-\mathrm{i}k_z)^j. \tag{25}$$

Huxley (1972) assumed an expansion for the isotropic part $f_0(v)$ of the distribution function similar to the expansion (23):

$$f_0(\boldsymbol{r}, v, t) = \sum_{i,j=0}^{\infty} f_{2i,j}(v)(\nabla_\perp^2)^i(-\partial/\partial z)^j n \tag{26}$$

with the normalization conditions

$$\langle f_{ij} \rangle \equiv \int f_{ij}(v)\, \mathrm{d}^3v = 1 \qquad \text{if} \qquad i = j = 0, \tag{27a}$$

$$= 0 \qquad \text{otherwise}. \tag{27b}$$

Inserting the expansion (26) into the Boltzmann equation (11), he arrived at the result that the diffusion equation (22) is exact, under rather general conditions. However, this result arose from an artificial splitting-up of the Boltzmann equation and the equating of the first terms individually to zero. If equation (22) is then inserted in the remaining terms, it is easily seen that those cannot generally be equal to zero, and the splitting-up procedure is therefore not admissible. This inconsistency can be removed by using equation (23) to eliminate the time derivatives in the Boltzmann equation.

We insert the expansions (23) and (25) into equation (11), equate the coefficients of $(\nabla_\perp^2)^i(\partial/\partial z)^j$ individually to zero and obtain the system of equations

$$\sum_{k=0}^{i}\sum_{l=0}^{j}\omega_{2k,l}f_{2i-2k,j-l}-d(v)[f_{2i-2,j}+f_{2i,j-2}]+\left(w(v)+\frac{2av}{3v}\frac{\mathrm{d}}{\mathrm{d}v}\right)f_{2i,j-1}$$
$$=\frac{1}{3v^2}\frac{\mathrm{d}}{\mathrm{d}v}\left[v^2\nu\left\{\frac{3mvf_{2i,j}}{M}+\left(\frac{a^2}{v^2}+\frac{3kT}{M}\right)\frac{\mathrm{d}f_{2i,j}}{\mathrm{d}v}\right\}\right], \qquad (28)$$

where we have written

$$d(v)\equiv\frac{v^2}{3\nu(v)}, \qquad w(v)\equiv\frac{1}{3v^2}\frac{\mathrm{d}}{\mathrm{d}v}\left(\frac{v^3a}{\nu(v)}\right). \qquad (29a,b)$$

The first of the system of equations (28), $i=j=0$, has zero on the left-hand side and gives for $f_{00}(v)$ the Davydov distribution

$$f_{00}(v)=N\exp\left(-\frac{3m}{M}\int_0^v\frac{c\,\mathrm{d}c}{\{a^2/\nu^2(c)+3kT/M\}}\right), \qquad (30)$$

where N is determined by the normalizing condition

$$\int f_{00}(v)\,\mathrm{d}^3v=4\pi\int f_{00}(v)\,v^2\,\mathrm{d}v=1. \qquad (31)$$

The right-hand side of equations (28), designated R(28), can now be simplified by introducing the function $f_{2i,j}(v)/f_{00}(v)$:

$$\mathrm{R}(28)=\frac{1}{3v^2}\frac{\mathrm{d}}{\mathrm{d}v}\left[v^2\nu\left\{\left(\frac{a}{\nu}\right)^2+\frac{3kT}{M}\right\}\frac{\mathrm{d}}{\mathrm{d}v}\left(\frac{f_{2i,j}}{f_{00}}\right)\right]. \qquad (32)$$

The system of equations (28) are solved in two steps, for successively larger i,j values. Firstly, the whole equation is multiplied by $4\pi v^2\,\mathrm{d}v$ and integrated. For reasonably well-behaved collision frequencies $\nu(v)$ and physically acceptable $f_{2i,j}$'s, the right-hand side then averages out to zero, and an equation is obtained for $\omega_{2i,j}$,

$$\omega_{2i,j}=\langle d(v)[f_{2i-2,j}(v)+f_{2i,j-2}(v)]\rangle+\langle w(v)f_{2i,j-1}(v)\rangle. \qquad (33)$$

(The expression involving $f_{2i,j-1}$ has been transformed by a partial integration.) Now, the whole left-hand side of (28) (L(28)) is known and, using equation (32) for R(28), $f_{2i,j}(v)$ can be found by two integrations:

$$f_{2i,j}(v)=f_{2i,j}(0)+4\pi f_{00}(v)B(v), \qquad (34)$$

where

$$B(v) = 3\int_0^v [c^2 \nu(c)\{(a/v)^2+3kT/M\}]^{-1}\,\mathrm{d}c \int_0^c x^2 \mathrm{L}(28)\,\mathrm{d}x. \tag{34a}$$

The constant $f_{2i,j}(0)$ is found by using the normalization conditions (27).

The coefficients $\omega_{2i,j}$ have been calculated numerically for $(2i+j) \leqslant 4$ for several models of interaction (that is, $\nu(v)$), assuming zero gas temperature $kT = 0$. The results are tabulated in Section 4. Here we will only consider the hard-sphere zero-temperature results, and use them to discuss the possible range of validity of the transport equation (23).

Introducing a 'mean free path for energy transfer', λ_e,

$$\lambda_e = (M/2m)^{\frac{1}{2}}\lambda_m, \tag{35}$$

λ_m being the mean free path for momentum transfer, the transport coefficients can be written

$$\left.\begin{aligned}
&\omega_{01} = v_d = 0{\cdot}7546(2m/M)^{\frac{1}{2}}(a\lambda_e)^{\frac{1}{2}}, && \omega_{20} = D_\perp = 0{\cdot}4607\,\lambda_e\omega_{01},\\
&\omega_{02} = D_\parallel = 0{\cdot}2262\,\lambda_e\omega_{01} = 0{\cdot}4910\,D_\perp, && \omega_{21} = 0{\cdot}164\,\lambda_e\omega_{20},\\
&\omega_{03} = 0{\cdot}163\,\lambda_e\omega_{20}, && \omega_{40} = (0{\cdot}121\,\lambda_e)^2\omega_{20},\\
&\omega_{22} = (0{\cdot}248\,\lambda_e)^2\omega_{20}, && \omega_{04} = (0{\cdot}234\,\lambda_e)^2\omega_{20}.
\end{aligned}\right\} \tag{36}$$

The relative magnitudes of the coefficients seem to indicate that the expansion (25) will diverge for wave numbers $k \gtrsim \lambda_e^{-1}$, showing that the transport equation (23) can, at most, only be valid for gradients small enough to fulfill the condition

$$k \sim |\nabla n|/n \lesssim \lambda_e^{-1}. \tag{37}$$

It is easily seen that, when the electron energy is large compared with thermal energies, the maximum length an electron can diffuse *against* the accelerating force is of the order λ_e. The quantity λ_e is thus a measure of the thickness of the 'boundary layers' where the influence of electrodes will be felt by the electron swarm. The condition (37) thus means that realistic boundary conditions cannot possibly be imposed upon equation (23); this should also apply to its truncated form (22).

Obviously, the condition (37) also means that a δ-function initial condition cannot be used. However, if the divergence of the expansion (25) for large wave numbers is neglected, it is possible to arrive at an expression for $n(\boldsymbol{r}, t)$ for this case—although the validity of the expression may be doubtful. As the expression obtained in this way gives a suitable starting point for a more general theory, we will sketch its derivation, following Kumar and Robson (1973).

With $n(\boldsymbol{r}, t = 0) = \delta(\boldsymbol{r})$, the Fourier spectrum of n becomes white, $\tilde{n}(\boldsymbol{k}) = 1$ in equation (24), and this equation, with the expansion (25) inserted for $\omega(\boldsymbol{k})$, can then be written

$$n(\boldsymbol{r}, t) = (2\pi)^{-3}\int \exp\{\mathrm{i}(\boldsymbol{k}.\boldsymbol{r} - \omega_{01}k_z t) - (\omega_{20}k_\perp^2 + \omega_{02}k_z^2)t\} \times \exp\left(\sum_{2i+j>2} \omega_{2i,j}\,t(-k_\perp^2)^i(-\mathrm{i}k_z)^j\right)\mathrm{d}^3k. \tag{38}$$

Without the second exponential present, this would have given an elliptically distorted Gaussian, the fundamental solution $P(\boldsymbol{r}, t)$ of equation (22):

$$P(\boldsymbol{r}, t) = (2\pi)^{-3} \int \exp\{\mathrm{i}(\boldsymbol{k}\cdot\boldsymbol{r} - \omega_{01} k_z t) - (\omega_{20} k_\perp^2 + \omega_{02} k_z^2)t\}\, \mathrm{d}^3k$$

$$= (4\pi t)^{-3/2} D_\parallel^{-1/2} D_\perp^{-1} \exp\left(-\frac{x^2+y^2}{4D_\perp t}\right) \exp\left(-\frac{(z-v_\mathrm{d} t)^2}{4D_\parallel t}\right) \tag{39}$$

(noting that $\omega_{01} = v_\mathrm{d}$, $\omega_{20} = D_\perp$ and $\omega_{02} = D_\parallel$ from (36)). We observe that the wave number $\boldsymbol{k}$ can be obtained by taking the gradient of $P(\boldsymbol{r}, t)$,

$$\nabla P(\boldsymbol{r}, t) = \mathrm{i}\boldsymbol{k} P(\boldsymbol{r}, t), \tag{40}$$

and use this to eliminate the second exponential in equation (38):

$$n(\boldsymbol{r}, t) = \exp\left(\sum_{2i+j>2} \omega_{2i,j}\, t (\nabla_\perp^2)^i (-\partial/\partial z)^j\right) P(\boldsymbol{r}, t). \tag{41}$$

We now write

$$\boldsymbol{\rho} = (\boldsymbol{x}+\boldsymbol{y})/(2D_\perp t)^{\frac{1}{2}} + (z - v_\mathrm{d} t)/(2D_\parallel t)^{\frac{1}{2}} \tag{42}$$

and introduce generalized Hermite polynomials $\mathrm{He}_{i,j}(\boldsymbol{\rho})$ (Grad 1949),

$$\mathrm{He}_{2i,j}(\boldsymbol{\rho}) = \exp(\tfrac{1}{2}\rho^2)\,(\partial/\partial\rho_\perp)^{2i}(-\partial/\partial\rho_z)^j \exp(-\tfrac{1}{2}\rho^2). \tag{43}$$

Expanding the exponential in (41) in a power series and writing $(\partial/\partial\rho_\perp)^2 = 2D_\perp t\,\nabla_\perp^2$ and $\partial/\partial\rho_z = (2D_\parallel t)^{\frac{1}{2}}\partial/\partial z$, the density can then be written

$$n(\boldsymbol{r}, t) = P(\boldsymbol{r}, t)\,[1 + t^{-\frac{1}{2}}\{\Omega_{21}\,\mathrm{He}_{21}(\boldsymbol{\rho}) + \Omega_{03}\,\mathrm{He}_{03}(\boldsymbol{\rho})\} + t^{-1}\{\Omega_{40}\,\mathrm{He}_{40}(\boldsymbol{\rho}) + \ldots\} + \ldots], \tag{44}$$

$$\Omega_{21} = \omega_{21}/2D_\perp(2D_\parallel)^{\frac{1}{2}}, \quad \Omega_{03} = \omega_{03}/(2D_\parallel)^{3/2}, \quad \Omega_{40} = \omega_{40}/(2D_\perp)^2, \quad \text{etc.} \tag{44a}$$

The Hermite polynomials $\mathrm{He}_{2i,j}(\boldsymbol{\rho})$ are 'scaled' with the Gaussian $P(\boldsymbol{r}, t)$ and thus give the *relative* shape corrections to the latter. The first correction involves third-order polynomials, and will give rise to a pear-shaped distortion that decays as $t^{-\frac{1}{2}}$. Note, however, that the lack of corrections of second order means that the mean square width of the expanding swarm is given by $P(\boldsymbol{r}, t)$ alone, i.e.

$$\langle (\boldsymbol{r} - v_\mathrm{d} t)^2 \rangle = 2\mathbf{D}t. \tag{45}$$

The expected correction term **A** (equations (16) and (17b)) has thus been lost in the present treatment, and one would expect the same to have happened to other terms of order τ_e/t.

4. Hermite Polynomial Expansion of Boltzmann Equation

The expression (44) for the electron density suggests that the spatial dependence of the distribution function could be expressed, from the beginning, by a generalized Hermite polynomial expansion, instead of the conjecture (26). This possibility has been explored and the approach has, in fact, been found to be a fruitful one,

enabling, for example, the handling of problems with a δ-function initial density distribution. In the following, the method of approach to the problem is described in more detail and some numerical results are then presented.

(*a*) *Expansion and Method of Solution*

We define generalized coordinates $\boldsymbol{\rho}$ as

$$\boldsymbol{\rho} = (x+y)/\sigma_x + (z-z_0)/\sigma_z, \tag{46}$$

σ_x, σ_z and z_0 for the moment being arbitrarily chosen position-independent quantities, and introduce the polynomials $H_{2i,j}(\boldsymbol{\rho})$,

$$H_{2i,j}(\boldsymbol{\rho}) = (\sigma_x^{2i}\sigma_z^j)^{-1}\,\mathrm{He}_{2i,j}(\boldsymbol{\rho}) = \exp(\tfrac{1}{2}\rho^2)(\nabla_\perp^2)^i(-\partial/\partial z)^j\exp(-\tfrac{1}{2}\rho^2). \tag{47}$$

The isotropic part of the distribution function can now be written, quite generally, in the form

$$f_0(\boldsymbol{r}, v, t) = (2\pi)^{-3/2}(\sigma_x^2\sigma_z)^{-1}\exp(-\tfrac{1}{2}\rho^2)\sum_{i,j=0}^{\infty} g_{2i,j}(v,t)\,H_{2i,j}(\boldsymbol{\rho}), \tag{48}$$

if we restrict ourselves to problems with cylindrical symmetry. Insertion of the expansion (48) into the Boltzmann equation (11) yields, when the coefficients of the $H_{2i,j}$'s are equated individually to zero, a set of parabolic equations for the functions $g_{2i,j}(v,t)$:

$$\frac{\partial g_{ij}}{\partial t} - \frac{1}{3v^2}\frac{\partial}{\partial v}\left[v^2\nu\left\{\frac{3mv\,g_{ij}}{M} + \left(\frac{a^2}{\nu^2} + \frac{3kT}{M}\right)\frac{\partial g_{ij}}{\partial v}\right\}\right]$$

$$= g_{i-2,j}(d(v)-d_x) + g_{i,j-2}(d(v)-d_z) + g_{i,j-1}(w(v)-v_0) - \frac{2}{3v^2}\frac{\partial}{\partial v}\left(\frac{v^3 a\,g_{i,j-1}}{\nu}\right). \tag{49}$$

Here $d(v)$ and $w(v)$ are as defined by equations (29) while d_x, d_z and v_0 are connected with the quantities σ_x, σ_z and z_0 by

$$d_x = \tfrac{1}{2}\mathrm{d}\sigma_x^2/\mathrm{d}t, \qquad d_z = \tfrac{1}{2}\mathrm{d}\sigma_z^2/\mathrm{d}t, \qquad v_0 = \mathrm{d}z_0/\mathrm{d}t. \tag{50}$$

We will solve the equations (49) directly by numerical methods. It may be useful, however, to note that their homogenous form is easily transformed to a Sturm–Liouville equation, with all eigenfunctions except the one corresponding to the Davydov distribution decaying exponentially with time. This observation makes it possible to draw conclusions concerning the asymptotic behaviour of the g_{ij}'s without really solving the equations.

We use the Crank–Nicolson finite difference method (see e.g. Smith 1965) to solve equations (49), calculating for each time step the functions with the smallest $(i+j)$ values first. For the solution we need two boundary conditions, which deserve some comments. Physically, neither the regions $v \to 0$ nor $v \to \infty$ should be of significance for the problem, and the boundary conditions to be used should therefore not be given, as is most usual, at the ends of the available speed interval $v \in (0, \infty)$. Furthermore, in the region $v \to 0$ the equations (49) are, for most realistic collision

frequencies, not well behaved, some coefficients tending to infinity. This is in line with the limitations (12) to the two-term Boltzmann equation used here.

At high v values, the functions $g_{ij}(v)$ must go to zero, and the boundary condition problem turns out to be easy. When the condition

$$g_{ij}(v_{\max}) = 0, \tag{51}$$

was tried, with $v_{\max}$ chosen to be a speed at which the Davydov distribution function has fallen to less than 10^{-4} of its maximum value, it was found that the solutions of the equations were insensitive to the actual choice of the cutoff speed $v_{\max}$, as one would expect.

To avoid the use of a boundary condition at $v \to 0$, we use the observation that when integrating the equations (49) over all velocity space the $\partial/\partial v$ terms average out to zero, leaving the relation

$$\frac{\mathrm{d}\langle g_{ij}\rangle}{\mathrm{d}t} \equiv \frac{\mathrm{d}}{\mathrm{d}t}\left(4\pi \int_0^\infty g_{ij}(v)\, v^2\, \mathrm{d}v\right)$$

$$= \langle g_{i-2,j}(d-d_x)\rangle + \langle g_{i,j-2}(d-d_z)\rangle + \langle g_{i,j-1}(w-v_0)\rangle\,. \tag{52}$$

The average $\langle g_{ij}\rangle$ can thus be obtained as the time integral over the right-hand side of (52) and can be calculated before the function $g_{ij}(v)$ is calculated (at the actual time). The remaining boundary condition can therefore be obtained by writing

$$4\pi \int g_{ij}(v)\, v^2\, \mathrm{d}v \to \sum \alpha_r g_{ij}(v_r) = \langle g_{ij}\rangle\,, \tag{53}$$

the α_r being the coefficients of the applied quadrature formulae. At each time step, each g_{ij} is thus found by solving a linear equation described by a matrix composed of one full row (equation (53)) plus a tridiagonal submatrix.

(b) Results for $\delta(r)$ Initial Conditions

We now specialize to the case of a δ-function initial distribution in space, corresponding to the initial conditions

$$\langle g_{00}(v, t = 0)\rangle = 1\,, \tag{54a}$$

$$g_{ij}(v, t = 0) = 0, \qquad \text{if} \quad i \text{ or } j \neq 0\,, \tag{54b}$$

$$\sigma_x(0) = \sigma_z(0) = z_0(0) = 0\,. \tag{54c}$$

The initial speed distribution is given by specifying $g_{00}(v, 0)$.

Inspection of equations (49) and (52) and a comparison with (28) and (33) shows that asymptotically $g_{00}(v)$ will always be a Davydov distribution, and further that asymptotically $(\partial/\partial t)g_{01}$, $(\partial/\partial t)g_{20}$ and $(\partial/\partial t)g_{02}$ will all be zero, if as $t \to \infty$ we let $d_x \to D_\perp$, $d_z \to D_\parallel$ and $v_0 \to v_{\mathrm{d}}$. We choose for simplicity d_x, d_z and v_{d} to have these values at all times, i.e.

$$d_x = D_\perp\,, \qquad d_z = D_\parallel\,, \qquad v_0 = v_{\mathrm{d}}\,. \tag{55}$$

Numerical calculations have been performed assuming zero gas temperature $kT = 0$, a collision frequency $\nu \propto v^{\gamma}$ $(-0{\cdot}5 \leqslant \gamma \leqslant 2)$ and either a steady-state (equation (30)) or a zero-energy initial speed distribution. The results are presented in terms of the ratios $D_{\perp}/v_{\mathrm{d}}$ as a length scale and $D_{\perp}/v_{\mathrm{d}}^2$ as a time scale:

$$l_0 = D_{\perp}/v_{\mathrm{d}} \quad \mathrm{m}, \qquad t_0 = D_{\perp}/v_{\mathrm{d}}^2 \quad \mathrm{s}. \tag{56}$$

The actual values of $D_{\perp}$ and v_{d} are easily calculated as

$$D_{\perp} = 4\pi \int_0^{\infty} \{v^2/3\nu(v)\} f_{00}(v)\, v^2 \, \mathrm{d}v, \tag{57}$$

$$v_{\mathrm{d}} = 4\pi \int_0^{\infty} \left(\frac{1}{3v^2} \frac{\mathrm{d}(v^3 a/\nu)}{\mathrm{d}v}\right) f_{00}(v)\, v^2 \, \mathrm{d}v, \tag{58}$$

where $f_{00}(v)$ is given by equation (30) and can for the chosen $\nu(v)$'s be expressed by means of Γ functions.

Table 1. Asymptotic ($t \to \infty$) values of the coefficients $\langle g_{ij} \rangle$

The results are expressed in terms of $l_0 = D_{\perp}/v_{\mathrm{d}}$ and $\tau = t/(D_{\perp}/v_{\mathrm{d}}^2)$. The values in parentheses in (*b*) are from Parker and Lowke (1969)

(*a*) *Davydov initial speed distribution*

γ	$D_{\parallel}/D_{\perp}$	$\langle g_{02}\rangle/l_0^2$	$\langle g_{21}\rangle/l_0^3$	$\langle g_{03}\rangle/l_0^3$	$\langle g_{40}\rangle/l_0^4$	$\langle g_{22}\rangle/l_0^4$	$\langle g_{04}\rangle/l_0^4$
$-0{\cdot}5$	2·02	$-2{\cdot}05$	$5{\cdot}7\tau - 13$	$7{\cdot}8\tau - 24$	$5{\cdot}0\tau - 11$	$38\tau - 150$	$35\tau - 160$
0	1·00	0	$2{\cdot}0\tau - 2{\cdot}9$	$2{\cdot}0\tau - 2{\cdot}9$	$0{\cdot}98\tau - 15$	$5{\cdot}6\tau - 14$	$4{\cdot}6\tau - 12$
0·5	0·666	0·32	$0{\cdot}82\tau - 0{\cdot}8$	$0{\cdot}74\tau - 0{\cdot}5$	$0{\cdot}25\tau - 0{\cdot}24$	$1{\cdot}1\tau - 1{\cdot}7$	$0{\cdot}90\tau - 1{\cdot}3$
1·0	0·491	0·34	$0{\cdot}35\tau - 0{\cdot}23$	$0{\cdot}36\tau - 0{\cdot}12$	$0{\cdot}068\tau - 0{\cdot}04$	$0{\cdot}29\tau - 0{\cdot}29$	$0{\cdot}26\tau - 0{\cdot}22$
1·5	0·376	0·26	$0{\cdot}12\tau - 0{\cdot}06$	$0{\cdot}19\tau - 0{\cdot}03$	$0{\cdot}011\tau - 0{\cdot}005$	$0{\cdot}065\tau - 0{\cdot}05$	$0{\cdot}097\tau - 0{\cdot}05$
2·0	0·291	0·20	0	$0{\cdot}11\tau - 0{\cdot}010$	0	0	$0{\cdot}039\tau - 0{\cdot}008$

(*b*) *Zero-energy initial distribution*

γ	$\langle g_{01}\rangle/l_0$	$\langle g_{20}\rangle/l_0^2$	$\langle g_{02}\rangle/l_0^2$	γ	$\langle g_{01}\rangle/l_0$	$\langle g_{20}\rangle/l_0^2$	$\langle g_{02}\rangle/l_0^2$
$-0{\cdot}5$	$-0{\cdot}81$ $(-3{\cdot}6)$	$-2{\cdot}80$	$-6{\cdot}4$ (19)	1·0	0·58 $(-2{\cdot}1)$	$-0{\cdot}33$	0·17 (3·5)
0	0 $(-3{\cdot}0)$	$-1{\cdot}44$	$-1{\cdot}44$ (9)	1·5	0·62	$-0{\cdot}12$	0·27
0·5	0·39	$-0{\cdot}71$	$-0{\cdot}22$	2·0	0·60 $(-1{\cdot}4)$	0	0·27 (1·4)

The asymptotic values of the $\langle g_{ij} \rangle$ are given in Table 1*a*, up to order $(i+j) = 4$, for the case of a steady-state initial speed distribution. The coefficients $\langle g_{01} \rangle$ and $\langle g_{20} \rangle$, which give corrections to the drift length and the mean square width in the lateral direction respectively, are in this case both zero. It is interesting to note that the terms linear in time are numerically the same as those obtained from the higher-order transport theory of Section 3, i.e.

$$\langle g_{ij} \rangle / t \underset{\tau}{\to} \omega_{ij}, \qquad (i+j) \leqslant 4. \tag{59}$$

A closer inspection of the relevant equations (namely (28) and (33) with (49) and (52)) shows that this must be the case, for $(i+j) < 5$. In the fifth and higher orders, however, the theory of Section 3 will give qualitatively wrong predictions—a fact that is probably of no practical consequence. Further, one notes that the numerical values of $\langle g_{02} \rangle$ are in fair agreement with the order-of-magnitude estimate, equation (21),

$$\langle g_{02} \rangle = \tfrac{1}{2} A_{\parallel} \sim (D_{\perp} - D_{\parallel}) \tau_{\mathrm{e}}. \tag{60}$$

When a zero-energy initial distribution was used, the terms linear in time in the (asymptotic) $\langle g_{ij} \rangle$ were found to be different from those obtained using a steady-state initial distribution for $(i+j) > 3$. This only reflects the fact, however, that the $\langle g_{01} \rangle$ values are now different from zero, the leading Gaussian in equation (48) thus having its centroid displaced from the actual centroid of the electron swarm. A straightforward transformation revealed that if the leading Gaussian was chosen to have its centroid in the position

$$z_0 = v_d t + \langle g_{01} \rangle, \tag{61}$$

instead of $z_0 = v_d t$, the terms linear in time were independent of the initial distribution, as one would expect.

The asymptotic values of $\langle g_{01} \rangle$, $\langle g_{20} \rangle$ and $\langle g_{02} \rangle$ for the case of zero initial energy are given in Table 1*b*; the values in parentheses are from Parker and Lowke (1969). The asymptotic mean square displacements from the centroid can be obtained from these values as

$$\langle x^2 \rangle = \langle y^2 \rangle = 2(D_\perp t + \langle g_{20} \rangle), \tag{62a}$$

$$\langle (z - \langle z \rangle)^2 \rangle = 2(D_\parallel t + \langle g_{02} \rangle - \tfrac{1}{2}\langle g_{01} \rangle^2). \tag{62b}$$

There is obviously a large discrepancy between the present results and those of Parker and Lowke.* If we take a closer look at their results for $\gamma = 0$, i.e. constant collision frequency ν, these can easily be seen to be wrong. When $\nu = $ const. the average velocity can be found directly from equation (10),

$$\langle \boldsymbol{v} \rangle = \tfrac{4}{3}\pi \int v f_1 v^2 \, \mathrm{d}v \, \mathrm{d}^3 r = a/\nu = v_d \qquad (\nu = \text{const.}) \tag{63}$$

and is independent of $f_0(v)$. Thus $\langle g_{01} \rangle$ must be zero in this case, in contrast to their finding. It is very probable that Parker and Lowke's other $\langle g_{ij} \rangle$ values are also wrong, and it is believed that this may possibly be due either to their use of Sturm–Liouville theory on equations with complex coefficients and eigenvalues or to their use of a series expansion for $\omega(\boldsymbol{k})$ as in equation (25) followed by a transformation similar to the one leading from equation (38) to (41), to include the effects of arbitrarily large gradients, although the series expansion diverges for large wave numbers.

For illustrative purposes, some hard-sphere results ($\gamma = 1$) obtained at times t not very much greater than $D_\perp/v_d^2$ are finally shown. In Fig. 1*a* the mean square displacement in the drift direction is plotted as a function of time. When the electrons have a steady-state (Druyvesteyn) initial distribution they are seen at first to diffuse with an apparent isotropic diffusion coefficient equal to $D_\perp$, the difference between the longitudinal and lateral diffusion first developing on a time scale $t \sim \tau_e \sim D_\perp/v_d^2$. This is in agreement with the behaviour expected from the qualitative considerations of Section 2*b*. With a zero-energy initial distribution, the electrons at first nearly do not move at all. This accounts for the form of the curve at small times. The fact that the asymptotic curve seems to extrapolate through the origin is fortuitous, and does not happen for other γ values.

* See note added in proof on p. 208.

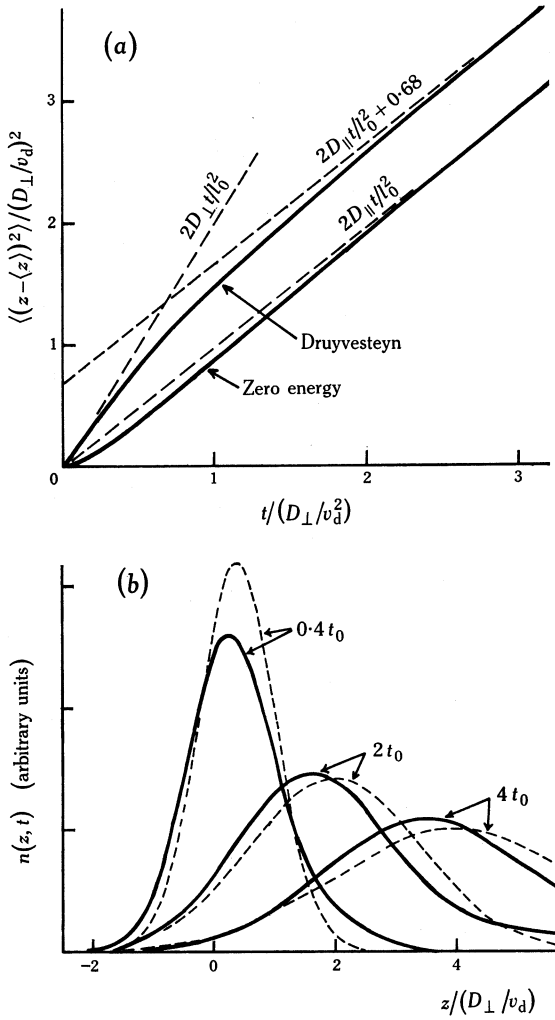

Fig. 1. Results for hard-sphere interaction:

(*a*) Mean square displacement in the drift direction, $\langle(z-\langle z\rangle)^2\rangle$, as a function of time for steady-state (Druyvesteyn) and zero-energy initial speed distributions.

(*b*) Electron density profiles $n(z)$ at times $t = 0{\cdot}4\,t_0$, $2t_0$ and $4t_0$, with $t_0 = D_\perp/v_d^2$, for a Druyvesteyn initial speed distribution. The dashed curves are the density profiles predicted by the diffusion equation (22).

Fig. 1*b* shows density profiles

$$n(z,t) = \int n(\boldsymbol{r},t)\,\mathrm{d}x\,\mathrm{d}y$$

for the case of a Druyvesteyn initial distribution. The density profiles predicted by the diffusion equation (22) are also shown (dashed) for comparison. The $n(z,t)$ profiles in Fig. 1*b* cannot be expected to be correct in detail, as Hermite polynomials of order higher than four have been neglected. However, they illustrate clearly the asymmetric distortion of the electron swarm (disappearing asymptotically as $t^{-\frac{1}{2}}$), which was also noted by McIntosh (1974). Further, they indicate that the thickness of the 'boundary layer', where the presence of a collecting electrode can be felt, is of the order $2D_\perp/v_d$, and illustrate the fact that diffusion theory cannot be used to describe the motion of electrons within this layer.

5. Discussion and Conclusions

The problem of electron motion in an unbounded medium, in the presence of large density gradients, has been attacked in two ways. Firstly, the possibility of using a higher-order transport equation (23) for the electron density $n(\boldsymbol{r}, t)$, instead of the usual diffusion equation (22) has been investigated and expressions for the coefficients of this equation have been found. Secondly, an expansion of the distribution function $f_0(\boldsymbol{r}, v, t)$ in generalized Hermite polynomials (in the space coordinates) has been inserted in the Boltzmann equation. The set of partial differential equations for the 'components' of $f_0(\boldsymbol{r}, v, t)$ that resulted has been solved directly by finite difference methods for various selected cases.

The present work has shown that a 'transport equation' for the density $n(\boldsymbol{r}, t)$, be it the diffusion equation (22) or the higher-order equation (23), can never adequately describe regions with large relative density gradients $|\nabla n|/n \gtrsim v_d D_\perp$. Thus, realistic boundary conditions cannot be imposed upon such equations. However, it has also been found, both from the numerical calculations and from the simpler arguments of Section 2*b*, that the *change with time* of *the mean square width* of an electron pulse outside the boundary regions is given by the diffusion coefficients, i.e. that 'difference measurements' of the mean square width can give experimental values for the diffusion coefficients, even under conditions when the pulse form is far from Gaussian and the total mean square width significantly different from the predictions of the ordinary diffusion equation (22).

Numerical values found in this work for the difference between the actual mean square width and the one predicted by the diffusion equation, disagree strongly with values given by Parker and Lowke (1969). Taking also into account the results of the computer simulations of McIntosh (1974), one would conclude that their values must be wrong, and that their theory cannot be used to correctly predict the effects of the initial development of an electron pulse on its asymptotic form. One would feel tempted also to conclude that reliable theoretical estimates of 'initial effects' and 'boundary effects' in a specific experiment can only (at best) be obtained from detailed, and time-consuming, numerical calculations, such as finite-difference calculations as in this work or direct simulations, and further that a better approach would be to design the experiments as difference experiments to avoid the necessity of such calculations.

Note added in proof

The pressure-dependent correction factors of Parker and Lowke (1969) are in error because of the incorrect assumption made in their paper that $F_1(\varepsilon) = F_2(\varepsilon) = 0$ at $\varepsilon = 0$. It should be noted, however, that this assumption does not affect their calculated values of D_L/μ (Parker and Lowke, personal communication).

Acknowledgments

The author would like to thank Dr R. W. Crompton and Dr M. T. Elford for many stimulating discussions and their continued interest in this work. He is also most grateful to Dr R. O. Watts and Dr K. Kumar for several helpful suggestions and comments.

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Manuscript received 7 September 1973