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Generalized Einstein Relation and Negative Differential Conductivity in Gases

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Abstract

Transport coefficients of charged particles undergoing both elastic and inelastic collisions with a gas of neutral molecules are calculated using momentum-transfer theory. A criterion is obtained for the phenomenon of negative differential conductivity (i.e. the drift velocity decreasing with applied electric field) and the well-known generalized Einstein relation is appropriately modified.

1. Introduction

Consider a tenuous swarm of particles of charge q moving in a neutral gas of number density N under the influence of an applied electrostatic field E. The density n of the swarm is so low that interaction between charged particles can be neglected and only charged particle–neutral molecule collisions need be considered. The companion paper (Petrović *et al.* 1984; referred to hereafter as PCH, see p. 23) considers the average or 'drift' velocity v_{dr} of electrons in gases under these conditions in the absence of spatial inhomogeneity, i.e. no diffusion. The phenomenon of negative differential conductivity (NDC), that is,

$$dv_{dr}/dE < 0, \tag{1}$$

is considered by PCH and a clearer physical picture of the reasons underlying this phenomenon emerges from their discussion in terms of model electron–molecule scattering cross sections. It is not the main aim of this paper to consider NDC *per se*, but rather to consider the next logical step in the discussion, the calculation of diffusion coefficients parallel and transverse to E. However, on the way to finding these coefficients, $D_{\|}$ and $D_{\perp}$ respectively, we derive an NDC criterion which warrants some discussion and comparison with the criterion of PCH.

Wannier's (1953) classic paper on gaseous ion transport contains a conjecture concerning a possible connection between the longitudinal diffusion coefficient and mobility $K = v_{dr}/E$, namely

$$\frac{D_{\|}}{K} = \frac{kT_{\|}}{q}\left(1 + \frac{\partial \ln K}{\partial \ln E}\right), \tag{2}$$

where k is Boltzmann's constant and $T_{\|}$ is the temperature associated with random motion parallel to the field direction. Equation (2) apparently went unnoticed for

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nearly twenty years. The validity of (2) was put on a firm foundation through nonequilibrium thermodynamics by Robson (1972), who at the same time illustrated its potential usefulness as a means of obtaining diffusion coefficients directly from measurements of mobility. Wannier (1973) subsequently obtained (2) via a Langevin equation. Klots and Nelson (1970) also reported an alternative derivation but the details have never been published.

More recently, the emphasis has been on obtaining corrections to (2) and the corresponding transverse relation

$$D_\perp/K = kT_\perp/q, \tag{3}$$

through so-called moment solutions of Boltzmann's equation, in which the ion distribution function is expanded in parametrized Burnett functions (see Viehland and Mason 1975*a*, 1975*b*, 1978; Waldman and Mason 1981, and references therein). In these works equations (2) and (3) have been called the generalized Einstein relations (GER) and we shall adhere to this nomenclature. Apart from the work of Viehland *et al.* (1981), however, all these derivations are based upon *elastic* collisions* and are therefore not appropriate for a description of NDC, which is intimately associated with certain inelastic processes. Viehland *et al.* (1981) did in fact obtain (2) and (3), but only in the context of a low-order truncation approximation scheme. The corrections to the GER are, as we shall see, of utmost importance.

We note in passing that these moment theories, while extremely valuable for providing numerical values of transport coefficients, tend to become rather cumbersome when analytic formulae are sought. There is an underlying fundamental physical basis for the existence of the GER, which the more elaborate theories tend to obscure, but which is evident particularly in the thermodynamic argument (Robson 1972) and in the so-called momentum-transfer theories (Whealton *et al.* 1974; Robson 1976; Robson and Mason 1982; Mason and McDaniel 1983). The observations of Kumar (1977) are also relevant in this regard.

If $\mathrm{d}v_{\mathrm{dr}}/\mathrm{d}E$ is negative, then so is

$$\frac{\mathrm{d}\ln v_{\mathrm{dr}}}{\mathrm{d}\ln E} = 1 + \frac{\partial \ln K}{\partial \ln E},$$

and equation (2) implies a negative and therefore unphysical diffusion coefficient. Thus, when NDC occurs, the traditional GER (2) is not merely inaccurate, but fails hopelessly, violating the Second Law of Thermodynamics! In Section 2 we derive a modified form of (2) on the basis of momentum-transfer theory analysis applied to a swarm of structureless charged particles undergoing inelastic as well as elastic collisions. The corresponding analysis of Robson (1976) in the absence of inelastic collisions yielded

$$\frac{D_\parallel}{K} = \frac{kT_\parallel}{q}\left(1+(1+\Delta_\parallel)\frac{\partial \ln K}{\partial \ln E}\right), \tag{4}$$

* This qualification was overlooked by Kleban and Davis (1978); it is not surprising that the unmodified GER does not work for electrons in CH_4, for in that case inelastic processes are important. The thermodynamic analysis (Robson 1972) explicitly includes such circumstances.

where the 'correction factor' $\Delta_{\|}$ is defined by

$$\Delta_{\|} = Q/(2kT_{\|}\, v_{\mathrm{dr}}), \tag{5}$$

Q being the heat flux per particle associated with charged particles in the spatially uniform state. The transverse relation (3), however, remained intact.* Precisely the same *form* of equation is found as a result of the more general analysis of Section 2, but now $\Delta_{\|}$ assumes the role of a fundamental parameter, not merely a small correction factor. [The general validity of (4) has already been assumed by Lin *et al.* (1979) for purposes of approximate calculation of $D_{\|}$ for electrons in methane gas, without any attempt at justification.]

In Section 3, we compare values of the ratio $D_{\|}/D_{\perp}$ computed from (i) the modified GER and (ii) the Boltzmann equation (Lin *et al.* 1979) for electrons in one of the model gases used by PCH. Even though significant inelastic collisions do occur, the electron velocity distribution function remains very nearly isotropic for the case chosen, i.e.

$$T_{\|} \approx T_{\perp}, \tag{6}$$

and hence $D_{\|}/D_{\perp}$ involves only K and $\Delta_{\|}$, as can be seen from (3) and (4). Indications are that the modified GER provides at least a good qualitative representation of $D_{\|}/D_{\perp}$ in the presence of NDC.

2. Theory

(a) *Momentum and Energy Balance Equations*

We employ approximate forms of momentum and energy balance equations for the ion swarm based upon the following assumptions:

(i) The hydrodynamic state has been reached and the ion density gradient is weak.

(ii) Collisional momentum and energy exchange between ions and neutrals can be represented by expressions of the same *form* as those in the special case where all relevant collision frequencies are constants, independent of energy.

The second assumption is the basis of the so-called 'momentum-transfer' theory, and although this is well known, some brief discussion is warranted, especially since we intend to apply it when inelastic processes are operative. The moment equations below are formed by integration of the Boltzmann equation with the Wang Chang–Uhlenbeck–de Boer (1964) collision term, as discussed in Section 14 of Kumar *et al.* (1980). Thus, for example, multiplication by $m\boldsymbol{v}$, where m and $\boldsymbol{v}$ are the ion mass and velocity respectively, followed by integration over all $\boldsymbol{v}$ yields

$$k\mathbf{T}\,.\,\nabla n - nq\boldsymbol{E} = -n\mu\langle \boldsymbol{g}\, \nu_{\mathrm{m}}(\varepsilon)\rangle \tag{7}$$

for the momentum balance equation, linearized in ∇n according to assumption (i), but otherwise exact. The average $\langle ... \rangle$ is over all relative velocities $\boldsymbol{g}$ of an ion and a molecule, μ is the reduced mass, $\varepsilon = \frac{1}{2}\mu g^2$ is the energy associated with relative ion–molecule motion, and ν_{m} denotes the momentum-transfer collision frequency.

* The more elaborate theories (Waldman and Mason 1981) yielded a nonzero transverse correction factor $\Delta_{\perp}$.

In order to approximate (7) with a more useful form, we assume $\nu_m(\varepsilon)$ to be in some sense a slowly varying function of ε. If the relative energy distribution is sharply peaked at ε_b, then the main contribution to the average in (7) comes from energies $\varepsilon \sim \varepsilon_b$, and we represent ν_m by the Taylor series expansion

$$\nu_m(\varepsilon) = \nu_m(\varepsilon_b) + (\varepsilon - \varepsilon_b)\nu_m'(\varepsilon_b) + \ldots, \tag{8}$$

for purposes of evaluating the right-hand side of (7). Momentum-transfer theory consists of retaining only the first term in the expansion and assuming that $\varepsilon_b = \langle\varepsilon\rangle$, that is

$$\langle \boldsymbol{g}\, \nu_m(\varepsilon)\rangle \approx \langle \boldsymbol{g}\rangle\, \nu_m(\langle\varepsilon\rangle). \tag{9}$$

It is to be emphasized, however, that ε_b could be left as a free parameter, to be subsequently adjusted to optimize the approximation. Similar reasoning applies in the choice of an unconstrained 'basis temperature' parameter T_b in the more rigorous Burnett function expansion method of solution of Boltzmann's equation (Lin *et al.* 1979). Notice that corrections to the momentum-transfer approximation involve $d\nu_m/d\varepsilon$, a point which we discuss later.

With the additional assumption that the average neutral molecule velocity vanishes, i.e. $\langle \boldsymbol{V}\rangle = 0$, we have $\langle \boldsymbol{g}\rangle = \langle \boldsymbol{v}\rangle$ and (7) is then approximated by

$$k\mathbf{T}.\nabla n - nq\boldsymbol{E} = -n\mu\, \nu_m(\langle\varepsilon\rangle)\langle \boldsymbol{v}\rangle. \tag{10}$$

This is exact if ν_m is a constant, independent of energy.

Similarly, we obtain an approximate energy balance equation, which after some manipulation takes the form

$$-\boldsymbol{Q}.\nabla n/\nu_e n = \langle\varepsilon\rangle - \tfrac{1}{2}M\langle V^2\rangle - \tfrac{1}{2}M\langle \boldsymbol{v}\rangle^2 + \Omega, \tag{11}$$

where $\boldsymbol{Q}$ is the heat flux per ion,

$$\nu_e = \frac{2m}{m+M}\nu_m \tag{12}$$

can be thought of as a collision frequency for elastic energy transfer, and

$$\Omega \equiv \frac{M}{m+M}\sum_i \varepsilon_i^*\{\langle \vec{\nu}_i(\varepsilon)\rangle - \langle \overleftarrow{\nu}_i(\varepsilon)\rangle\}/\nu_e(\langle\varepsilon\rangle), \tag{13}$$

the sum being over all inelastic processes i characterized by an energy loss ε_i^* and collision frequency $\vec{\nu}_i$. Superelastic processes have been accounted for through the collision frequency $\overleftarrow{\nu}_i$.

In this case the approximation $\langle \vec{\nu}_i(\varepsilon)\rangle \approx \vec{\nu}_i(\langle\varepsilon\rangle)$ may be a poor one, since inelastic processes are characterized by rapidly varying cross sections in the neighbourhood of thresholds, i.e. $\vec{\nu}_i(\varepsilon)$ is *not* a slowly varying function of ε and (8) is inappropriate. Another means of averaging must be sought; for example, one might assume that the energy distribution is Maxwellian and in that case a straightforward calculation indicates that

$$\langle \vec{\nu}_i(\varepsilon)\rangle \approx \vec{\nu}_i(\langle\varepsilon\rangle)(1+\xi)\exp(-\xi), \tag{14}$$

where $\xi \equiv -\frac{3}{2}\varepsilon_i^*/\langle\varepsilon\rangle$. The exponential factor varies between 0 and 1 for $\langle\varepsilon\rangle \ll \varepsilon_i^*$ and $\langle\varepsilon\rangle \gg \varepsilon_i^*$ respectively, and has the effect of 'smoothing' the rapid jump in $\nu_i(\langle\varepsilon\rangle)$ in the vicinity of the threshold.

The main thrust of this paper is, however, independent of the prescription for averaging and we really need only specify that $\Omega = \Omega(\langle\varepsilon\rangle)$ for present purposes.

(*b*) *Spatial Homogeneity, Drift Velocity and NDC Criterion*

For convenience, we shall omit the averaging brackets below, i.e.

$$\langle \boldsymbol{v}\rangle \to \boldsymbol{v}, \qquad \langle\varepsilon\rangle \to \varepsilon .$$

For spatial uniformity $\nabla n = 0$ and equations (10) and (11) yield

$$\boldsymbol{v}_{dr} = q\boldsymbol{E}/\mu\nu_m(\varepsilon), \tag{15}$$

$$\varepsilon = \tfrac{3}{2}kT + \tfrac{1}{2}Mv_{dr}^2 - \Omega(\varepsilon), \tag{16}$$

respectively, where we have assumed a Maxwellian distribution for the neutrals,

$$\tfrac{1}{2}M\langle V^2\rangle = \tfrac{3}{2}kT,$$

and we have written v_{dr} for v. Following convention, we call v_{dr} the 'drift velocity'.

Equations (15) and (16) are two equations for the two unknowns v_{dr} and ε. In special cases, analytic solutions are obtainable; for example, if inelastic processes are absent so that $\Omega = 0$, then

(i) for constant collision frequency ν_m,

$$v_{dr} = qE/\mu\nu_m \propto E;$$

(ii) for constant cross section, $\nu_m \propto \varepsilon^{\frac{1}{2}}$ and at high fields ($\frac{1}{2}Mv_{dr}^2 \gg \frac{3}{2}kT$)

$$v_{dr} \propto E^{\frac{1}{2}}.$$

Both these results are well known. In more general circumstances (15) and (16) have to be solved numerically. However, a knowledge of the explicit field dependence is not necessary to achieve the result we desire. The above examples are given for purposes of illustration only.

We note in passing that (16) is essentially the same as equation (77) of Viehland *et al.* (1981), if we identify $\frac{3}{2}kT_{eff}$ with ε and $\xi(\varepsilon)$ with $(m/M)(\Omega/\varepsilon)$. Their equation derives from a low-order truncation approximation in a polynomial expansion method of solution of Boltzmann's equation. The connection between momentum-transfer theory and such low-order approximations has been established, at least in the absence of inelastic processes (see e.g. Viehland and Mason 1975*b*, 1978).

It is useful at this point to establish the identity

$$\frac{\partial \ln K/\partial \ln E}{1 + \partial \ln K/\partial \ln E} = -\frac{Mv_{dr}^2\, \mathrm{d}\ln \nu_m/\mathrm{d}\varepsilon}{1 + \mathrm{d}\Omega/\mathrm{d}\varepsilon}. \tag{17}$$

This is used in Section 2*c*.

Proof: By (15), we have for the mobility coefficient

$$K = v_{dr}/E = q/\mu\nu_m,$$

and hence

$$\frac{\partial \ln K}{\partial \ln E} = -\frac{\partial \ln \nu_m}{\partial \ln E} = -\frac{\mathrm{d} \ln \nu_m}{\mathrm{d}\varepsilon}\frac{\partial \varepsilon}{\partial \ln E}.$$

By (16), we have upon differentiating with respect to $\ln E$ and regrouping terms

$$\frac{\partial \varepsilon}{\partial \ln E} = \frac{M v_{dr}\, \partial v_{dr}/\partial \ln E}{1 + \mathrm{d}\Omega/\mathrm{d}\varepsilon}, \tag{18}$$

and hence

$$\frac{\partial \ln K}{\partial \ln E} = -\frac{\mathrm{d} \ln \nu_m}{\mathrm{d}\varepsilon}\frac{M v_{dr}^2}{1 + \mathrm{d}\Omega/\mathrm{d}\varepsilon}\left(1 + \frac{\partial \ln K}{\partial \ln E}\right).$$

Equation (17) then follows immediately.

From equation (18), we see that the criterion for NDC, i.e. for $\partial v_{dr}/\partial E < 0$, is

$$1 + \mathrm{d}\Omega/\mathrm{d}\varepsilon < 0, \tag{19}$$

if it is assumed that the mean energy monotonically increases with increasing field.

Our NDC criterion is different from equation (3) of PCH, whose energy balance equation (2) is different* from ours and does not produce (19).

In order to illustrate the meaning of (19), we take the case of electrons for which $m \ll M$ and assume only one inelastic process with threshold energy ε^* and cross section $\sigma_i(\varepsilon)$. The momentum-transfer cross section is denoted by $\sigma_m(\varepsilon)$. We assume a cold gas ($T = 0$) for simplicity, enabling us to neglect superelastic collisions. Thus, we get

$$\vec{\nu}_i(\varepsilon) = N(2\varepsilon/m)^{\frac{1}{2}}\sigma_i(\varepsilon)\, S(\varepsilon), \qquad \nu_m(\varepsilon) = N(2\varepsilon/m)^{\frac{1}{2}}\sigma_m(\varepsilon),$$

and from the definitions (13) and (2), we have

$$\Omega(\varepsilon) = \frac{M}{2m}\varepsilon^*\frac{\sigma_i(\varepsilon)}{\sigma_m(\varepsilon)}S(\varepsilon), \tag{20}$$

where $S(\varepsilon)$ is a smoothing factor (cf. the approximation 14).

It is now clear what combinations of elastic and inelastic processes will satisfy (19): two possibilities are shown in Figs 1*a* and 1*b*. A comprehensive discussion is to be found in PCH. Qualitatively speaking, the shape of the curve of Ω versus ε is the same in both cases and is shown in Fig. 2. Points 1 and 2 mark the boundaries of the NDC region and correspond to the zeros of

$$1 + \Omega'(\varepsilon) = 0. \tag{21}$$

The drift velocity takes on a maximum and a minimum respectively at these two points (see Fig. 3). When (21) has only one real positive zero, the curve of drift velocity versus E merely exhibits a point of inflection, there being no NDC.

* PCH do not consider elastic collisional energy transfer. Their momentum balance equation is, on the other hand, in agreement with ours.

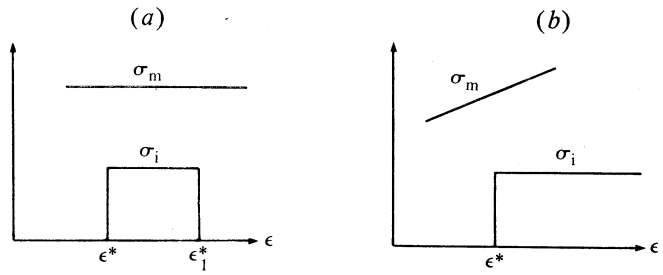

Fig. 1. Schematic representation of two types of elastic–inelastic cross section combinations favourable for NDC.

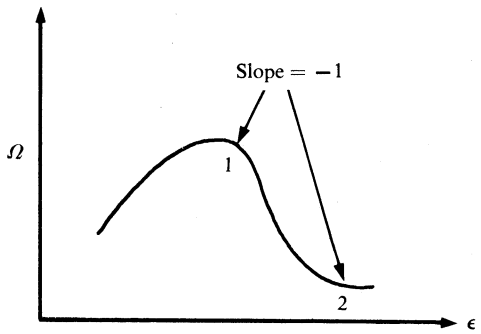

Fig. 2. Schematic representation of the function Ω defined by equation (20). As in the text ε, now stands for the mean energy. The region between the two points 1 and 2 corresponds to NDC, where the inequality (19) holds.

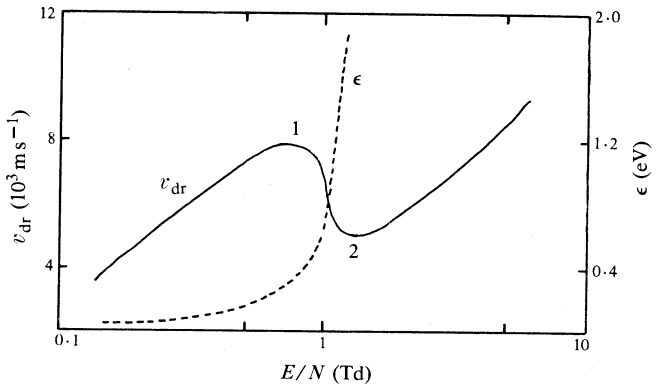

Fig. 3. Drift velocity and mean energy as functions of field as computed from equations (15) and (16) for cross sections of the form shown in Fig. 1*a*, with $\sigma_i = 0{\cdot}03$ Å^2, $\sigma_m = 5{\cdot}0$ Å^2, $\varepsilon^* = 0{\cdot}1$ eV, $\varepsilon^* = 1{\cdot}0$ eV, $M = 28$ a.m.u. and $T = 0$. The turning points in v_{dr} correspond to energies satisfying (21), i.e. to points 1 and 2 in Fig. 2. (1 Td $\equiv 10^{-21}$ V m^2.)

Other points to note are: (i) NDC is intimately connected with inelastic processes—if only elastic collisions between electrons and neutrals take place, NDC is not possible; (ii) there appears to be no connection with the suggestion of Kleban and Davis (1977, 1978) concerning 'strong anisotropy' in velocity space as being basically responsible for NDC—the phenomenon can occur even when there is no such pronounced anisotropy (see Section 3).

(*c*) *Diffusion and the GER*

Equations (10) and (11) represent momentum and energy balance equations linearized in ∇n. If we write

$$\boldsymbol{v} = \boldsymbol{v}_{\mathrm{dr}} + \delta\boldsymbol{v}, \tag{22}$$

where $\boldsymbol{v}_{\mathrm{dr}}$ is given by (15) and $\delta\boldsymbol{v}$ is a small perturbation of order ∇n accounting for deviations from spatial homogeneity then, after allowing for a similar perturbation $\delta\varepsilon$ in energy,* equations (10) and (11) together yield

$$\delta\boldsymbol{v} = -\mathbf{D}.\nabla n/n, \tag{23}$$

where the *diffusion tensor* is given by

$$\mathbf{D} = D_{\perp}\mathbf{1} + (D_{\parallel} - D_{\perp})\hat{\boldsymbol{E}}\hat{\boldsymbol{E}}, \tag{24}$$

and where

$$D_{\perp} = \frac{kT_{\perp}}{\mu\nu_{\mathrm{m}}}, \tag{25}$$

$$D_{\parallel} = \frac{kT_{\parallel}}{\mu\nu_{\mathrm{m}}}\left(\frac{1-\beta\Delta_{\parallel}}{1+\beta}\right). \tag{26}$$

The quantity $\Delta_{\parallel}$ is defined by (5) and

$$\beta = \frac{M v_{\mathrm{dr}}^2}{1+\Omega'(\varepsilon)}\frac{\mathrm{d}\ln\nu_{\mathrm{m}}}{\mathrm{d}\varepsilon}. \tag{27}$$

It follows from (17) and (27) that

$$\beta = -\frac{\partial\ln K/\partial\ln E}{1+\partial\ln K/\partial\ln E},$$

and substituting this into (26) yields the required modified GER (4) for $D_{\parallel}$. On the other hand, (25) again leads to the traditional form (3) of GER for the transverse diffusion coefficient $D_{\perp}$. (This is basically so because the heat flux $\boldsymbol{Q}$ contains no components transverse to $\boldsymbol{E}$.)

3. Discussion

To illustrate the usefulness of the modified GER we consider electrons in a model gas as in model 5b of PCH (see p. 30). This is a particular case of Fig. 1*a* with $\sigma_{\mathrm{m}} = 5$ Å^2, $\sigma_{\mathrm{i}} = 0{\cdot}03$ Å^2 (1 Å $\equiv 10^{-10}$ m), $\varepsilon^* = 0{\cdot}1$ eV, $\varepsilon_{\mathrm{i}}^* = 1{\cdot}0$ eV and with molecular mass $M = 28$ a.m.u. Using the method of Lin *et al.* (1979) of solving Boltzmann's equation, we have calculated v_{dr}, $D_{\parallel}$ and $D_{\perp}$ over the range of E/N

* This turns out to be

$$\delta\varepsilon = -\left(\frac{M\boldsymbol{v}_{\mathrm{dr}}D_{\parallel} + \boldsymbol{Q}/\nu_{\mathrm{e}}}{1+\Omega'}\right).\nabla n/n.$$

where NDC occurs. It is found that the so-called two-term approximation holds to within about 1% in this case, i.e. the velocity distribution function is only weakly anisotropic, and that equation (6) holds. Equations (3) and (4) may then be combined to give

$$D_{\parallel}/D_{\perp} = 1+(1+\Delta_{\parallel})\partial \ln K/\partial \ln(E/N). \tag{28}$$

The exact value of $D_{\parallel}/D_{\perp}$ obtained directly from solution of Boltzmann's equation is compared with the value obtained from (28) in Fig. 4. [As this is a model calculation, our 'data' consists of values of K and $\Delta_{\parallel}$ calculated from Boltzmann's equation, just as for the CH_4 model treated in Lin *et al.* (1979).] The good semi-quantitative agreement is evident. It is also evident that the momentum-transfer theory values of drift velocity shown in Fig. 3 are in good agreement with the more accurate values shown in Fig. 4.

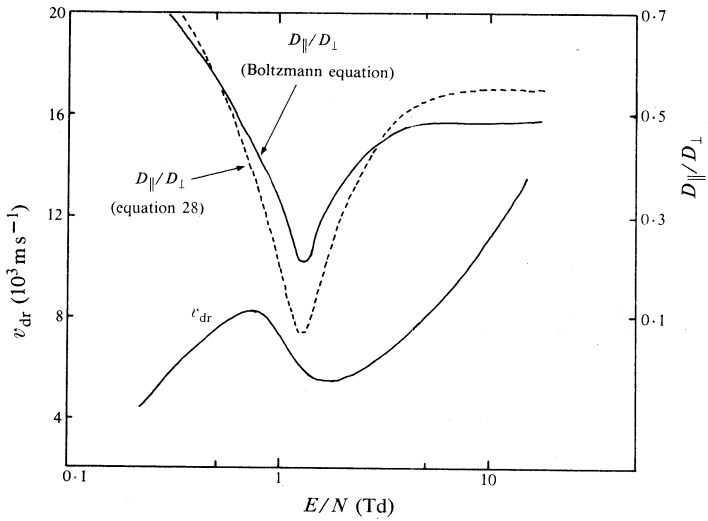

Fig. 4. Ratio of diffusion coefficients as computed from equation (28) and from direct solution of the Boltzmann equation for the same circumstances as in Fig. 3, except that $T = 293$ K. The drift velocity has been computed from the Boltzmann epuation (cf. Petrović *et al.* Fig. 6*b*, curve b; see p. 29).

Notice that at high fields, inelastic collisions may be neglected and $\Delta_{\parallel}$ is small. The field dependence of transport coefficients is thus appropriate to a constant elastic cross section, that is (as observed in Section 2*b*) v_{dr} is proportional to $E^{\frac{1}{2}}$ and by (28), $D_{\parallel}/D_{\perp}$ has the approximate value 0·5.

Representation of $T_{\parallel}$, $T_{\perp}$ and $\Delta_{\parallel}$ in terms of K and its derivatives is essential if (3), (4) and (28) are to be of any practical value. Expressions for these quantities in terms of K could be obtained from higher order moment equations, using the momentum-transfer approximation. However, as Skullerud (1973, 1976) pointed out, the expression for $T_{\parallel}$ obtained from the constant collision frequency model (effectively the momentum-transfer approximation formula) is of little value, and he went on to suggest that a more appropriate value can be obtained from the formula

$$\frac{T_{\parallel}-T}{(T_{\parallel}-T)_{MT}} = 1+\beta_s\frac{\partial \ln K}{\partial \ln E}, \tag{29}$$

where 'MT' denotes the appropriate momentum-transfer theory expression and $\beta_s \sim 0{\cdot}85$ is an adjustable parameter. Waldman and Mason (1981) have pursued this idea further in connection with $\Delta_{\parallel}$ [they also obtained a nonzero correction factor $\Delta_{\perp}$ for (3)] but as they concluded '... a complete restudy is needed in order to proceed to the level of the more refined GER ...' when inelastic collisions occur. Any such study will have to produce formulae which are capable of dealing satisfactorily with the rather extreme circumstances associated with NDC. The present work is offered as a first step in this direction.

Finally, we note that the appearance of correction terms of order $\partial \ln K/\partial \ln E$ for momentum-transfer theory formulae is not unexpected, since terms neglected in making the momentum-transfer approximation are $O(\nu'_m)$ (equation 8) and equation (17) shows clearly that ν'_m is $O(\partial \ln K/\partial \ln E)$.

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