On Nonlocal Constitutive Relations, Continued Fraction Expansion for the Wave Vector Dependent Diffusion Coefficient

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Nonlocal constitutive relations which involve wave vector dependent transport coefficients can be derived from the Boltzmann equation. Diffusion of a Lorentzian gas is treated as an illustrative example. Transport-relaxation equations obtained from the Boltzmann equation with the help of the moment method lead to a continued fraction expansion for the wave vector dependent diffusion coefficient $D(\boldsymbol{k})$. Rapidly converging upper and lower bounds on $D(\boldsymbol{k})/D(0)$ are found which are meaningful for all values of lk where l is a mean free path and k is the magnitude of the wave vektor $\boldsymbol{k}$. Also some remarks on a frequency and wave vector dependent diffusion coefficient are made.

The constitutive laws which link the "thermodynamic fluxes" with their pertaining "forces" are nonlocal, in general. Thus these laws formulated in terms of the spatial Fourier transforms of the relevant macroscopic variables involve transport coefficients which depend on the wave vector $\boldsymbol{k}$. In the limit $\boldsymbol{k} \rightarrow 0$ they reduce to the usual local transport coefficients of hydrodynamics. For dilute gases, such nonlocal constitutive laws can be obtained from an approximate solution of the Boltzmann[1] equation or a generalization thereof[2, 3]. It is the purpose of this article to show that the moment method[4, 5] yields well defined upper and lower bounds on a wave vector dependent transport coefficient. Diffusion of a Lorentzian gas is treated as an illustrative example. Here, a continued fraction expansion for the diffusion coefficient $D(\boldsymbol{k})$ is found. Compared with the higher order Chapman-Enskog solution precedure[4, 6], which essentially aims for an expansion of the wave vector dependent transport coefficients in terms of powers of the wave vector, the results obtained from the moment method enjoy all the advantages of a rather rapidly converging continued fraction expansion over a power series expansion with finite radius of convergence.

This paper is divided into 4 sections. In Sect. 1, some general remarks are made on nonlocal constitutive relations for a medium which is translationally invariant and isotropic in the absence of a transport process. Sections 2–4 are devoted to treatment of a simple example, viz. diffusion of a Lorentzian gas. In Sect. 2, the Boltzmann equation is stated and it is outlined how the "transport-relaxation equations" are derived with the help of the moment method. Then, in Sect. 3, the subset of the transport-relaxation equations needed for the calculation of the wave vector dependent diffusion coefficient $D(\boldsymbol{k})$ is considered in more detail. The case where the time derivatives can be disregarded is treated first. Truncation of the infinite set of equations at the n-th moment and successive elimination of moments leads to the n-th approximant of a continued fraction expansion for $D(\boldsymbol{k})$. It turns out that the diffusion coefficient $D(\boldsymbol{k})$ depends on $\boldsymbol{k}$ via the dimensionless variable lk where l is a mean free path and k the magnitude of the wave vector $\boldsymbol{k}$. Alternating orders n of the continued fraction expansion are upper and lower bounds on the true $D(\boldsymbol{k})$ which improve with increasing n. To demonstrate this point, $D(\boldsymbol{k})/D(0)$ is plotted as function of lk or $(lk)^2$ for two specific collision models. The corresponding power series approximation is also displayed. It can be used for $lk \lesssim 1$ only. This is in contradistinction to the continued fraction expansion which is well defined for all values of lk. In Sect. 4, some supplementary remarks are made. Firstly, it is indicated how a noninstantaneous constitutive relation is derived from the transport-relaxation equations. Then the diffusion coefficient depends on the frequency and the wave vector. It is mentioned that this coefficient can also be inferred from the spectral functions associated with the density autocorrelation function. Secondly, results analogous to those obtained in Sect. 3 are stated for a 2-dimensional model gas.

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1. General Remarks on Nonlocal Constitutive Relations

The local conservation laws contain thermodynamic fluxes $j(\boldsymbol{r})$ (e.g. heat flow, diffusion flow). Hydrodynamics is based on the use of local constitutive relations of the form

$$j(\boldsymbol{r}) = L_0 F(\boldsymbol{r}) \tag{1.1}$$

where $F(\boldsymbol{r})$ is the pertaining thermodynamic force (for heat conduction and diffusion, e.g. $-F$ is proportional to the gradient of the temperature and of the concentration, respectively). The quantity $L_0 > 0$ is a transport coefficient. In general, the interrelation between $j(\boldsymbol{r})$ and $F(\boldsymbol{r})$ is nonlocal, viz.

$$j(\boldsymbol{r}) = \int \mathrm{d}^3 r' \, \mathcal{L}(\boldsymbol{r}-\boldsymbol{r}') \, F(\boldsymbol{r}') = \int \mathrm{d}^3 r' \, \mathcal{L}(\boldsymbol{r}') F(\boldsymbol{r}-\boldsymbol{r}') \tag{1.2}$$

where $\mathcal{L}(\boldsymbol{r})$ is a "transport coefficient density". In Eq. (1.2), the system under consideration has been assumed to be translational invariant [in the absence of the "force" $F(\boldsymbol{r})$].

The total entropy production

$$\left(\frac{\delta S}{\delta t}\right)_{\text{irr}} = \int \mathrm{d}^3 r \, j(\boldsymbol{r}) \, F(\boldsymbol{r}) = \iint \mathrm{d}^3 r \, \mathrm{d}^3 r' \, F(\boldsymbol{r}) \mathcal{L}(\boldsymbol{r}-\boldsymbol{r}') \, F(\boldsymbol{r}') \tag{1.3}$$

caused by the transport process is positive provided that $\mathcal{L}(\boldsymbol{r})$ is positive definite.

The local constitutive law (1.1) is recovered from (1.2) if $\mathcal{L}(\boldsymbol{r})$ is approximated by $L_0 \delta(\boldsymbol{r})$. Formally, this local law may be considered as a first term in an expansion of (1.2) with respect to higher spatial derivatives of $F(\boldsymbol{r})$. For a system which is isotropic (in the absence of a transport process), this expansion is

$$j(\boldsymbol{r}) = L_0 F(\boldsymbol{r}) + L_2 \Delta F(\boldsymbol{r}) + L_4 \Delta^2 F(\boldsymbol{r}) + \dots, \tag{1.4}$$

where the L_2, L_4 ... are higher order Burnett coefficients. The L_p, $p = 0, 2, 4, \dots$ are related to the quantity $\mathcal{L}(\boldsymbol{r})$ by

$$L_p = [(p+1)!]^{-1} \int \mathrm{d}^3 r \, r^p \, \mathcal{L}(\boldsymbol{r}) \, . \tag{1.5}$$

It is a crucial question whether the expansion (1.4) which underlies the "higher order hydrodynamics" exists beyond the first term and when it is physically meaningful.

For $j(\boldsymbol{r})$ and $F(\boldsymbol{r})$ of the form $j(\boldsymbol{r}) = j_k e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$ and $F(\boldsymbol{r}) = F_k e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$, the relation (1.2) is equivalent to $j_k = L(\boldsymbol{k}) F_k$ where

$$L(\boldsymbol{k}) = \int \mathrm{d}^3 r \, e^{-i\boldsymbol{k}\cdot\boldsymbol{r}} \mathcal{L}(\boldsymbol{r}) \tag{1.6}$$

is the spatial Fourier transform of $\mathcal{L}(\boldsymbol{r})$. Notice that $L(\boldsymbol{k})$ is real since one has $\mathcal{L}(\boldsymbol{r}) = \mathcal{L}(-\boldsymbol{r})$ due to parity arguments. Positive entropy production requires $L(\boldsymbol{k}) > 0$. Furthermore, the local transport coefficient L_0 is simply related to $L(\boldsymbol{k})$ by $L_0 = L(0)$. The expansion (1.4) corresponds to an expansion of $L(\boldsymbol{k})$ in powers of the wave vector $\boldsymbol{k}$. Hence the limit of applicability of (1.4) is determined by the radius of convergence of the power series expansion of $L(\boldsymbol{k})$.

For dilute gases, in principle the transport coefficient density $\mathcal{L}(\boldsymbol{r})$ or equivalently $L(\boldsymbol{k})$ can be calculated from the Boltzmann equation. In the remainder of this paper, it is pointed out that the transport-relaxation equations derived from the Boltzmann equation lead to a rather rapidly converging continued fraction expansion for $L(\boldsymbol{k})$. The (higher order) Chapman-Enskog solution procedure aims for a power series expansion of $L(\boldsymbol{k})$ equivalent to (1.4). Due to the finite radius of convergence, the power series expansion and consequently the "higher order hydrodynamics" is of limited value only. This is in contradistinction to the "generalized hydrodynamics" based on the transport-relaxation equations.

In the following, diffusion is considered as a specific example for a transport process.

2. Kinetic Equation, Transport-Relaxation Equations

To treat the simplest case, diffusion of a Lorentzian gas of structureless particles is considered. The magnitude c of the particle velocity $\boldsymbol{c}$ is assumed to be constant. The distribution function f then depends on t, $\boldsymbol{r}$ and $\boldsymbol{e}$ where $\boldsymbol{e}$ is a unit vector parallel to $\boldsymbol{c}$ (i.e. $\boldsymbol{c} = c\,\boldsymbol{e}$). The local average of a quantity $\Psi(\boldsymbol{e})$ is then given by

$$n \langle \Psi \rangle = \int \Psi(\boldsymbol{e}) f(t, \boldsymbol{r}, \boldsymbol{e}) \, \mathrm{d}^2 e \tag{2.1}$$

where $n = \int f \, \mathrm{d}^2 e$ is the number density of the Lorentzian particles.

The time evolution of f is governed by the Boltzmann equation

$$\partial f / \partial t + \boldsymbol{c} \cdot \boldsymbol{\nabla} f + \omega(f) = 0 \, . \tag{2.2}$$

Here $\omega(f)$ stands for the collision term. It is proportional to the number density n_s of the randomly distributed scattering centers. Conservation of the number of particles implies $\omega(1)=0$ and $\int \omega(f)\,\mathrm{d}^2e = 0$. Hence integration of (2.2) over d^2e yields the continuity equation

$$\partial n/\partial t + \nabla \cdot \boldsymbol{j} = 0 . \tag{2.3}$$

where $\boldsymbol{j} = n\,c\,\langle \boldsymbol{e} \rangle$ is the particle flux density.

Next, the moment method is used to derive an infinite set of transport-relaxation equations which is equivalent to the Boltzmann equation (2.2). The particle flux density $\boldsymbol{j}$ and the nonlocal diffusion coefficient can then be inferred from a truncated set of these equations.

The distribution function $f(t, \boldsymbol{r}, \boldsymbol{e})$ is written as

$$f = f_0(1+\varphi)\,, \quad f_0 = n_0\,(4\,\pi)^{-1} . \tag{2.4}$$

Here f_0 is the equilibrium distribution function with the equilibrium number density n_0. The quantity $\varphi = \varphi(t, \boldsymbol{r}, \boldsymbol{e})$ is a measure for the deviation of f from f_0. Following the procedure used in Ref. [5], φ is expanded with respect to cartesian tensors $\Phi_{\mu_1\ldots\mu_l}$ constructed from the unit vector $\boldsymbol{e}$, viz:

$$\varphi = a + a_\mu\,\Phi_\mu + a_{\mu\nu}\,\Phi_{\mu\nu} + \ldots . \tag{2.5}$$

Cartesian components are denoted by Greek subscripts; the summation convention is used. The $\Phi_{\mu_1\ldots\mu_l}$ $(l=1, 2, \ldots)$ are defined by

$$\Phi_{\mu_1\ldots\mu_l} \left[\frac{(2\,l+1)\,!!}{l!}\right]^{1/2} \overline{e_{\mu_1}\,e_{\mu_2}\ldots e_{\mu_l}} \tag{2.6}$$

where the symbol $\overline{\ldots}$ refers to the symmetric traceless part of a tensor. The expansion tensors are orthogonal and normalized in the sense

$$\begin{aligned}&\langle \Phi_{\mu_1\ldots\mu_l}\,\Phi_{\nu_1\ldots\nu_k} \rangle_0 = 0 \quad \text{for } l \neq k \text{ and}\\ &\langle \Phi_{\mu_1\ldots\mu_l}\,\Phi_{\nu_1\ldots\nu_l} \rangle_0 = \Delta^{(l)}_{\mu_1\ldots\mu_l,\,\nu_1\ldots\nu_l}\end{aligned} \tag{2.7}$$

where $\langle \ldots \rangle_0 = (4\,\pi)^{-1} \int \mathrm{d}^2e \ldots$ denotes an equilibrium average. In (2.7), $\Delta^{(l)}_{\ldots,\ldots}$ stands for an isotropic tensor of rank $2\,l$ which is symmetric and traceless both in its front and back sets of indices [7]. Applied on an arbitrary tensors $c_{\mu_1\ldots\mu_l}$ of rank l, it projects out the symmetric traceless part $\overline{c_{\mu_1\ldots\mu_l}}$ i.e. one has $\overline{c_{\mu_1\ldots\mu_l}} = \Delta^{(l)}_{\mu_1\ldots\mu_l,\,\nu_1\ldots\nu_l}\,c_{\nu_1\ldots\nu_l}$. Furthermore, it has the property $\Delta^{(l)}_{\mu_1\ldots\mu_l,\,\nu_1\ldots\nu_l} = 2\,l+1$.

The expansion coefficients occurring in (2.5) are moments of the distribution function which depend on t and $\boldsymbol{r}$. In particular, the properties (2.7) imply

$$a = (n-n_0)/n_0\,, \quad a_{\mu_1\ldots\mu_l} = (n/n_0)\,\langle \Phi_{\mu_1\ldots\mu_l} \rangle\,, \quad l = 1, 2, \ldots . \tag{2.8}$$

In the following, the factor n/n_0 in front of $\langle \Phi \ldots \rangle$ in (2.8) is replaced by 1, i.e. terms nonlinear in quantities which characterize a deviation from equilibrium are disregarded. Notice that the expansion (2.5) is equivalent to an expansion with respect to spherical harmonics depending on the polar angles of $\boldsymbol{e}$.

Insertion of (2.5) into the Boltzmann equation (2.2), multiplication of this equation by 1, Φ_μ, $\Phi_{\mu\nu}$, ..., integration over d^2e, use of (2.7), (2.8) and of

$$c\,\langle \Phi_{\mu_1\ldots\mu_{l-1}}\,e_{\mu_l}\,\Phi_{\nu_1\ldots\nu_l} \rangle_0 = c_l\,\Delta^{(l)}_{\mu_1\ldots\mu_l,\,\nu_1\ldots\nu_l}\,, \quad c_l = c\left(\frac{l}{2\,l+1}\right)^{1/2} \tag{2.9}$$

leads to a set of coupled linear differential equations for the moments a, a_μ, $a_{\mu\nu}$, ... These equations are referred to as transport-relaxation equations. For the spatial Fourier transform $a_{\mu_1\ldots\mu_l}(k)$ of the moments $a_{\mu_1\ldots\mu_l}(\boldsymbol{r})$, the first three of these equations are

$$\frac{\partial a}{\partial t} + i\,c_1\,k_\mu\,a_\mu = 0\,, \tag{2.10}$$

$$\frac{\partial a_\mu}{\partial t} + i\,c_1\,k_\mu\,a + i\,c_2\,k_\nu\,a_{\nu\mu} + \omega_1\,a_\mu = 0\,, \tag{2.11}$$

$$\frac{\partial a_{\mu\nu}}{\partial t} + i\,c_2\,\overline{k_\mu\,a_\nu} + i\,c_3\,k_\lambda\,a_{\lambda\mu\nu} + \omega_2\,a_{\mu\nu} = 0\,. \tag{2.12}$$

The l-th equation of this infinite set of equations is

$$\partial a_{\mu_1\ldots\mu_l}/\partial t + i\,c_l\,\overline{k_{\mu_l}\,a_{\mu_1\ldots\mu_{l-1}}} + i\,c_{l+1}\,k_{\mu_{l+1}}\,a_{\mu_{l+1}\mu_1\ldots\mu_l} + \omega_l\,a_{\mu_1\ldots\mu_l} = 0\,. \tag{2.13}$$

The collision frequencies ω_l, $l = 1, 2, 3$ which stem from the collision term of the Boltzmann equation (2.2) are defined by

$$\omega_l = (2\,l+1)^{-1}\,\langle \Phi_{\mu_1\ldots\mu_l}\,\omega\,(\Phi_{\mu_1\ldots\mu_l}) \rangle_0\,. \tag{2.14}$$

Notice that Eq. (2.10) (where no relaxation frequency shows up) is equivalent to the continuity equation (2.3). The coupling between the equations of the various moments is caused by the flow term of the Boltzmann equation. Clearly, this coupling is such that the "l"-moment is linked with the "$l \pm 1$"-moments (dipolar selection rule).

In the following section, it is shown how a continued fraction expression for the wave vector dependent diffusion coefficient $D(\boldsymbol{k})$ (corresponding to the quantity $L(\boldsymbol{k})$ of Sect. 1) can be inferred from the transport relaxation equations.

3. Continued Fraction Expansion for the Wave Vector Dependent Diffusion Coefficient

In terms of the variables used in Sect. 2, the nonlocal relation between the diffusion flow density and the gradient of the number density corresponding to (1.6) is

$$c_1\, a_\mu(\boldsymbol{k}) = -D(\boldsymbol{k})\, i\, k_\mu\, a(\boldsymbol{k})\,. \tag{3.1}$$

Here $D(\boldsymbol{k})$ is the wave vector dependent diffusion coefficient corresponding to $L(\boldsymbol{k})$. For $\boldsymbol{k} \rightarrow 0$, $D(\boldsymbol{k})$ reduces to the ordinary (local) diffusion coefficient D_0. Before a relation of the form (3.1) between a_μ and a is inferred from the transport-relaxation equations (2.11) – (2.13), it is noticed that $\boldsymbol{a}$ as given by (3.1) is parallel to $\boldsymbol{k}$. Hence it suffices to consider the equation for $\hat{\boldsymbol{k}} \cdot \boldsymbol{a}$ (where $\hat{\boldsymbol{k}}$ is a unit vector parallel to the wave vector $\boldsymbol{k}$) which is obtained from (2.11) by scalar multiplication with $\hat{k}_\mu$. The resulting equation then contains the component $\hat{k}_\mu \hat{k}_\nu a_{\mu\nu}$ of the second rank tensor $a_{\mu\nu}$. An equation for this component is obtained by multiplication of (2.12) with $\hat{k}_\mu \hat{k}_\nu$. Thus for the diffusion problem, it is sufficient to consider the set of transport-relaxation equations for the scalars

$$a_j = \hat{k}_{\mu_1} \dots \hat{k}_{\mu_j} a_{\mu_1 \dots \mu_j}\,, \quad j = 1, 2, \dots \tag{3.2}$$

Notice that $a_j \sim \langle P_j(\boldsymbol{e} \cdot \hat{\boldsymbol{k}}) \rangle$ where P_j is a Legendre polynomial. From (2.11) – (2.13), one readily infers

$$\frac{\partial a_1}{\partial t} + i\, k\, c_2\, a_2 + \omega_1\, a_1 = -i\, c_1\, \gamma_1\, k\, a\,, \tag{3.3}$$

$$\frac{\partial a_2}{\partial t} + i\, k\, c_2\, \gamma_2\, a_1 \;+ i\, k\, c_3\, a_3 + \omega_2\, a_2 = 0\,, \tag{3.4}$$

$$\frac{\partial a_j}{\partial t} + i\, k\, c_j\, \gamma_j\, a_{j-1} + i\, k\, c_{j+1} \;+ \omega_j\, a_j \;= 0\,. \tag{3.5}$$

Here, use has been made of

$$\hat{k}_{\mu_1} \dots \hat{k}_{\mu_j} \overline{k_{\mu_j}\; a_{\mu_1 \dots \mu_{j-1}}} = \gamma_j\, k\, a_{j-1}\,, \tag{3.6}$$

with

$$\gamma_j = j(2\,j-1)^{-1}\,. \tag{3.7}$$

Equations (3.3) – (3.5) for $a_1, a_2, \dots a_j, \dots$ have to be solved for a_1 where the term involving $i k a$ on the right hand side of Eq. (3.3) which corresponds to $\boldsymbol{\nabla}\, n$) is the inhomogeneity. This solution is now discussed for the case where all time derivatives in (3.3) – (3.5) can be disregarded, i. e. for the case where the time variation of the moments a_j is slow compared with the relaxation times $\tau_j = \omega_j^{-1}$, $j = 1, 2, \dots$ If this approximation does not apply, the resulting constitutive relation becomes not only nonlocal but also noninstantaneous. Some remarks on this point are made in Section 4.

Next, it is shown how a continued fraction expansion for $D(\boldsymbol{k})$ is obtained from a truncated version of the infinite set of transport-relaxation equations. Let a_n be the highest moment that is included. Then the equation for a_n, with $\partial a_n/\partial t = 0$, yields.

$$a_n = -i\, k\, c_n\, \gamma_n\, \omega_n^{-1}\, a_{n-1}\,. \tag{3.8}$$

Insertion of (3.8) into the equation for a_{n-1} then, with $\partial a_{n-1}/\partial t = 0$, leads to

$$a_{n-1} = -i\, k\, c_{n-1}\, \gamma_{n-1} \\ (\omega_{n-1} + k^2\, c_n^2\, \gamma_n\, \omega_n^{-1})^{-1}\, a_{n-2}\,. \tag{3.9}$$

Continuation of this procedure finally leads to

$$a_1 = -i\, c_1\, \gamma_1\, k\, a\, \frac{1}{\omega_1 +}\, \frac{c_2^2\, \gamma_2\, k^2}{\omega_2 +} \dots \frac{c_n^2\, \gamma_n\, k^2}{\omega_n} \tag{3.10}$$

where $\dfrac{x_1}{y_1 +}\, \dfrac{x_2}{y_2 +} \dots$ stands for the continued fraction $\dfrac{x_1}{y_1 + \dfrac{x_2}{y_2 + \dots}}$.

To obtain the desired expression for the wave vector dependent diffusion coefficient $D(\boldsymbol{k})$, Eq. (3.10) has to be compared with

$$c_1\, a_1 = -D(\boldsymbol{k})\, i\, k\, a \tag{3.11}$$

which follows from (3.1) after multiplication with $\hat{k}_\mu$. The hydrodynamic approximation (corresponding to $n = 1$) for $D(\boldsymbol{k})$ is

$$D_0 = D(0) = c_1^2\, \gamma_1\, \omega_1^{-1} = \frac{1}{3}\, \frac{c^2}{\omega_1}\,, \tag{3.12}$$

cf. Eqs. (2.9), (3.7). The n-th approximation for $D(\boldsymbol{k})$ as it can be inferred from (3.10) is determined by

$$\mathrm{D}(\boldsymbol{k})/D(0) = \vartheta^{(n)}(\boldsymbol{k}) \equiv \\ \frac{1}{1 +}\, \frac{\alpha_2\, k^2\, l^2}{1 +}\, \frac{\alpha_3\, k^2\, l^2}{1 +} \dots \frac{\alpha_n\, k^2\, l^2}{1}\,, \tag{3.13}$$

where the mean free path l and the coefficients α_j, $j = 2, 3, \dots$, are defined by

$$l = c\, \omega_1^{-1} = (n_{\mathrm{s}}\, \sigma_1)^{-1}\,, \tag{3.14}$$

and

$$\alpha_j = \frac{j^2}{(2\,j+1)\,(2\,j-1)}\, \frac{\omega_1^2}{\omega_{j-1}\, \omega_j}\,. \tag{3.15}$$

In (3.14), n_s is the number density of the scattering centers and the effective cross section σ_1 is defined by $\omega_1 = n_s \, c \, \sigma_1$. The continued fraction (3.13) converges for all values of $l k$ provided that the series $\Sigma \, a_j^{-1/2}$ diverges [8]. Thus (3.13) converges if the collision frequencies ω_j do not decrease stronger than j^{-1} for increasing j. Consequently, for the relaxation frequencies ω_j encountered in kinetic theory [4] which do not decrease with increasing j, the convergence of the continued fraction expansion is certainly guaranteed.

It is a particular useful property of a converging continued fraction expansion [8], that the approximants $\vartheta^{(n)}(k)$ with even n and odd n are lower and upper bounds, respectively, on $D(\boldsymbol{k})/D(0)$. These bounds improve with increasing n.

A power series expansion for $D(\boldsymbol{k})$ which corresponds to an expansion with respect to higher order gradients of the form (1.4) can now also be inferred from (3.13). Up to 4-th order, this expansion is

$$D(\boldsymbol{k}) = D(0)\,[1 - a_2 k^2 l^2 + a_2 (a_2 + a_3) k^4 l^4 \ldots] \,. \qquad (3.16)$$

In general, the radius of convergence of such an expansion depends on the a_j and it will be finite, i. e. (3.16) is only meaningful if $l k$ is smaller than a certain finite value. The continued fraction expansion (3.13), on the other hand, is well defined for all values of $l k$.

To demonstrate the essential features of the approximations (3.13) and (3.16) for the wave vector dependent diffusion coefficient $D(\boldsymbol{k})$, two specific model collision terms are considered. Firstly, $\omega_j = \omega_1$ is assumed [9] (single relaxation time). Then, for $n \to \infty$, $D(\boldsymbol{k})$ becomes

$$D(\boldsymbol{k}) = D(0)\, 3\, (l\,k)^{-2}\, [l\,k\, (\operatorname{arc\,tg} l\,k)^{-1} - 1] \,. \qquad (3.17)$$

Incidentally, for this special case the exact $D(\boldsymbol{k})$ could also have been inferred from the solution of the kinetic equation as discussed in Reference [10]. In Fig. 1, $\vartheta(k) = D(\boldsymbol{k})/D(0)$ is plotted as function of $l k$. The unlabelled curve is the exact result given by (3.17). Curves calculated from the approximants $\vartheta^{(n)}$, cf. Eq. (3.13) which are based on the use of n moments are labelled by $M(n)$. Clearly, the approximations $M(2)$ and $M(3)$ are lower and upper bounds on the exact function $D(\boldsymbol{k})/D(0)$. Curves obtained from an power series expansion of the form (3.16) with the highest order term proportional to $(k\,l)^{2(n-1)}$ are denoted by $P(n)$. With this notation, both $M(1)$ and $P(1)$ correspond to the straight line $D(\boldsymbol{k})/D(0) = 1$. For the specific example under consideration, the power series converges for $l k < 1$ only. Thus it is not surprising that the curves $P(2)$ and $P(3)$ are rather useless for $l k > 1$. In Fig. 2, $D(\boldsymbol{k})/D(0)$ is plotted as function of $(l\,k)^2$ over a somewhat larger range of $(l\,k)$-values. Besides the exact curve (unlabelled) the moment approximations $M(2) - M(5)$ are displayed. This demonstrates how the upper and lower bounds improve with increasing n. For comparison, the power series approximations $P(2)$ and $P(3)$ are shown again.

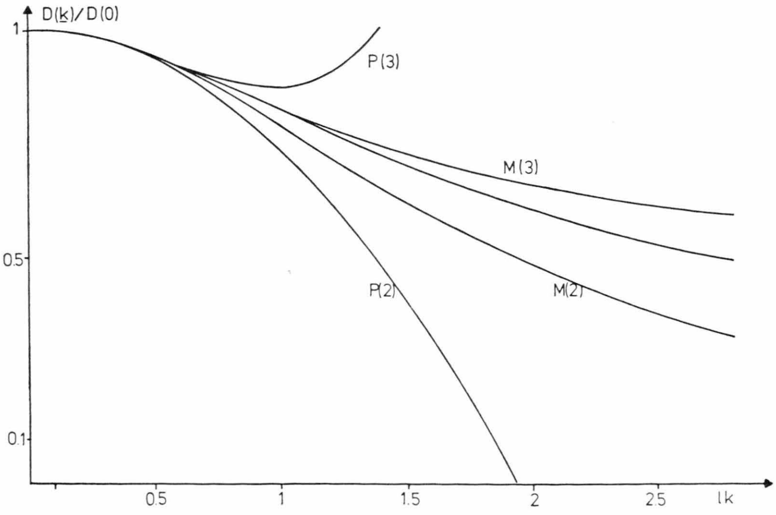

Fig. 1. The ratio $D(\boldsymbol{k})/D(0)$ as function of $l k$ for a collision term with a single relaxation frequency. Curves calculated from the truncated continued fraction expansion (3.13) based on the use of n moment equations are labelled by $M(n)$. Curves obtained from the power series expansion (3.16) with the highest order term $\sim (k l)^{2(n-1)}$ are denoted by $P(n)$. The unlabelled curve is the exact result as given by (3.17).

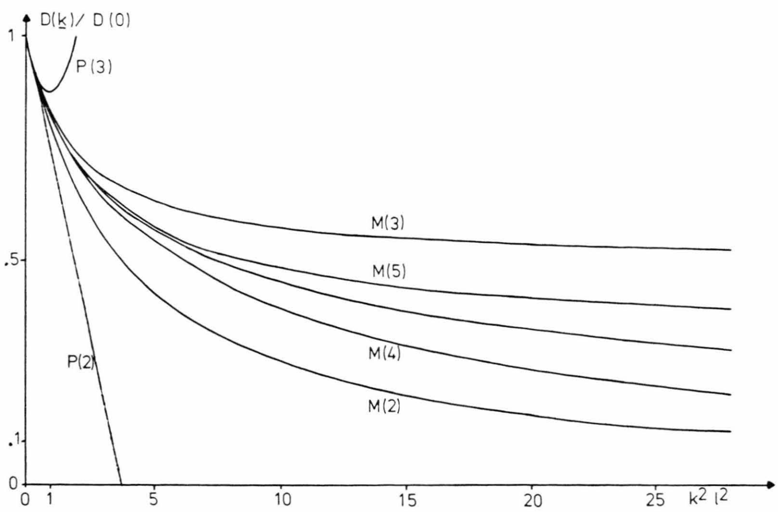

Fig. 2. The ratio $D(\boldsymbol{k})/D(0)$ as function of $(lk)^2$ for a collision term with a single relaxation frequency. The labelling of the curves has the same meaning as in Fig. 1, the unlabelled curve is the exact result.

Next, the case $\omega_j = \frac{1}{2} j(j+1)\omega_1$ is considered as it results from a Fokker-Planck type collision term (diffusion in velocity space). In Fig. 3 $D(\boldsymbol{k})/D(0)$ is plotted as function of $(lk)^2$ over the same range as in Figure 2. Clearly, the convergence of the continued fraction approximants is much better if the ω_j increase with increasing j. For this reason $M(2)$ and $M(3)$ only are shown. Also the power series approximations are somewhat better, yet still useless for $lk \gtrsim 2$.

In the following section, some supplementary remarks are made on the case where a frequency dependence of the diffusion coefficient has also to be taken into account. Furthermore, it is pointed out that a result similar to (3.13) is also obtained for a 2-dimensional model gas.

4. Some Supplementary Remarks

a) Frequency and Wave Vector Dependent Diffusion Coefficient

In the preceding section, it has been assumed that the time derivatives in the transport-relaxation equations (3.3) – (3.5) can be disregarded. If this approximation cannot be made, the resulting constitutive relation is not only nonlocal but also noninstantaneous. In this case it is advantageous to use Fourier transformations both with respect to time and space. Then the diffusion coefficient defined in (3.1) also depends on the frequency variable ω. A continued fraction expansion analogous to (3.13) for this diffusion coefficient $D(\omega, \boldsymbol{k})$ can be ob-

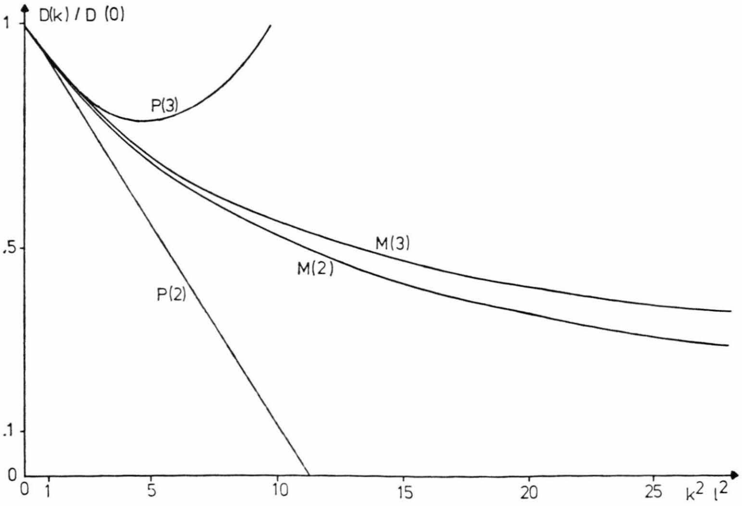

Fig. 3. The ratio $D(\boldsymbol{k})/D(0)$ as function of $(lk)^2$ for a collision term with relaxation frequencies $\omega_j = \frac{1}{2} j(j+1)\omega_1$. The labelling of the curves by $M(n)$ and $P(n)$ has the same meaning as in Figure 1.

tained from Eqs. (3.3) – (3.5) with $\partial a_j/\partial t$ replaced by $-i\,\omega\,a_j$. Now

$$D(\omega,\boldsymbol{k}) = D(\omega,0)\,\vartheta^{(n)}(\omega,\boldsymbol{k}) \qquad (4.1)$$

is found with $D(\omega,0) = D_0[1-i(\omega/\omega_1)]^{-1}$, for D_0 see (3.12). The quantity $\vartheta^{(n)}(\omega,\boldsymbol{k})$ is the n-th approximation of a continued fraction as given by (3.13) but with $a_i(j=2,3,\dots)$ replaced by

$$a_j\left(1-i\frac{\omega}{\omega_{j-1}}\right)^{-1}\left(1-i\frac{\omega}{\omega_j}\right)^{-1}, \qquad (4.2)$$

for a_j see (3.15). Notice that $D(\omega,\boldsymbol{k})$ is complex, in general, whereas $D(0,\boldsymbol{k}) = D(\boldsymbol{k})$ is real.

Insertion of $c_1\,a_1 = -D(\omega,\boldsymbol{k})\;i\,k\,a$ into the Fourier transformed continuity equation (2.1) yields

$$S(\omega,\boldsymbol{k}) \sim \mathrm{Re}\,[-i\,\omega + D(\omega,\boldsymbol{k})\,k^2]^{-1} \qquad (4.3)$$

for the spectral function $S(\omega,\boldsymbol{k})$ associated with the density autocorrelation function. This spectral function e. g. occurs in connection with incoherent neutron scattering [10, 11] and Raman scattering [10]. For a model kinetic equation with a single relaxation frequency ω_1, $S(\omega,\boldsymbol{k})$ can be calculated exactly. Comparison with (4.3) then yields [10]

$$D(\omega,\boldsymbol{k})\,k^2 = [\langle(-i\,\omega+\omega_1 + i\,\boldsymbol{c}\cdot\boldsymbol{k})^{-1}\rangle_0]^{-1} - \omega_1\,. \qquad (4.4)$$

The expression for $D(0,\boldsymbol{k})$ obtained from (4.4) agrees with (3.17).

b) Two-Dimensional System

Moment equations corresponding to Eqs. (2.10) to (2.13) and (3.3) – (3.5) can also be derived for a two-dimensional model gas. The scalar variables a_j occurring in the equations analogous to (3.3) to (3.5) then are proportional to $\langle\cos j\,\varphi\rangle$ where φ is the angle between $\boldsymbol{e}$ and $\boldsymbol{k}$, i. e. between the direction of the velocity and the wave vector. In this case, $D(0) = D_0 = \frac{1}{2}\,c/\omega_1$ is found and $D(\boldsymbol{k})$ is again given by a continued fraction expansion of the form (3.13) with

$$a_j = \frac{1}{4}\,\frac{\omega_1{}^2}{\omega_{j-1}\,\omega_j}\,, \quad j=2,3,\dots\,. \qquad (4.5)$$

For the special case $\omega_j = \omega_1$, $j=2,3,\dots$ (single relaxation time), the expression analogous to (3.17) is

$$D(\boldsymbol{k}) = D(0)\,2\,(l\,k)^{-2}\,[(1-l^2\,k^2)^{1/2}-1]\,. \qquad (4.6)$$

This result can also be inferred from (4.4). Clearly, in the 2-dimensional case, $D(\boldsymbol{k})$ as given by the continued fraction expansion or by (4.4) is just as well defined for all values of $l\,k$ as in the 3-dimensional case.

Concluding Remarks

In this article, it has been demonstrated in connection with the diffusion of a Lorentzian gas that the moment solution procedure of the Boltzmann equation leads to a very useful continued fraction expansion of a wave vector (and frequency) dependent transport coefficient. The specific results presented in Sect. 3 (and 4) are restricted to the special case of a constant magnitude of the particle velocity. For the general case (Maxwellian velocity distribution in thermal equilibrium) Sonine polynomials show up in the expansion tensors (cf. Sect. 2) and an enlarged set of transport-relaxation equations has to be studied. Still, it should be possible to derive meaningful upper and lower bounds on $D(\boldsymbol{k})$.

Furthermore, it seems desirable to extend the present approach to the heat conduction and viscosity problems. Applications for wave vector and frequency dependent transport coefficients are found in connection with the absorption and dispersion of sound waves and with the Rayleigh and Brillouin scattering.

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