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Transport Coefficients and Velocity Distribution Function of an Ion Swarm in an A.C. Electric Field Obtained from the BGK Kinetic Equation

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Abstract

The transition to a periodic steady state for an ion swarm in a gas is investigated using the BGK model kinetic equation. Exact expressions for transport coefficients and the velocity distribution function are obtained and the latter is compared with experimental observations of ions in their parent gases undergoing predominantly charge-transfer collisions.

1. Introduction

The term 'hydrodynamic regime' is usually employed in the kinetic theory of gaseous swarms (Kumar 1981, 1984; Kumar *et al.* 1980; Mason and McDaniel 1988) to describe the situation where the space–time dependence of the swarm phase-space distribution function $f(\boldsymbol{r}, \boldsymbol{v}, t)$ is carried by the number density

$$n(\boldsymbol{r},t) = \int \mathrm{d}\boldsymbol{v}\ f(\boldsymbol{r},\boldsymbol{v},t)\,, \tag{1}$$

and all memory of initial conditions, save those for $n(\boldsymbol{r},t)$ itself, is lost. This, at least, is the case for swarms in d.c. fields, and a definitive study of the transition from an arbitrary state to the hydrodynamic regime has been given by Kumar (1981). He showed that the relaxation from the initial state to the hydrodynamic regime is controlled by a single time constant, the same for all transport quantities. In doing so, however, he made certain assumptions about the spectral properties of the operator,

$$M = \boldsymbol{a}\,.\,\partial_{\boldsymbol{v}} + J\,,$$

which still await proof. In this equation, J denotes the swarm particle–neutral molecule Boltzmann collision operator and $\boldsymbol{a} = e\boldsymbol{E}/m$ is the acceleration undergone by a swarm particle of mass m, charge e, in the presence of the electric field $\boldsymbol{E}$. The Boltzmann equation for the swarm phase-space distribution function is thus

$$(\partial_t + \boldsymbol{v}\,.\,\nabla + M)f = 0\,. \tag{2}$$

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In the present paper, we consider the situation where the field varies harmonically in time,

$$\boldsymbol{E}(t) = \boldsymbol{E}_0 \cos\omega t\,,$$

and consider the transition to the 'periodic steady state', where all memory of initial conditions has been lost. In this state, all transport properties oscillate at the frequency ω of the applied field (or at harmonics thereof) about respective mean values which do not change in the course of time. Any *spatial* (but not time) variation is carried entirely by the density $n(\boldsymbol{r},t)$, but whether or not the term 'hydrodynamic regime' should be applied is a moot point.

There are certain symmetry properties that can be deduced from (2), e.g. in the absence of spatial variations

$$f(-\boldsymbol{v},\, t+\pi/\omega) = f(\boldsymbol{v},t), \qquad f(\boldsymbol{v},\, t+2\pi/\omega) = f(\boldsymbol{v},t)\,, \tag{3}$$

but otherwise the mathematical problems posed by time-varying fields are far more onerous than for the d.c. case and hence a detailed investigation, along the lines of Kumar (1981), seems out of the question at present. For this reason, we have opted in the first instance to present the results of a model kinetic equation calculation, based on the well-known BGK expression (Bhatnagar *et al.* 1954) for J. The main advantage of this model lies in its mathematical tractability. However, the BGK model also represents a reasonable description of ions and their parent gas undergoing charge-transfer collisions (Kumar *et al.* 1980), an important type of interaction in radio frequency plasmas. We show here that the peculiar structure observed in the ion energy distribution (Kuypers and Hopman 1990; Lie *et al.* 1990; Manenschijn *et al.* 1991) can be understood, at least qualitatively, on the basis of this model.

The format of this paper is as follows: in Section 2 we review briefly Kumar's formal theory of time-dependent transport coefficients, and indicate how it can be extended to include time-dependent fields. We then perform explicit calculations using the BGK collision operator, showing the transition to the periodic steady state and providing asymptotic expressions for transport coefficients. In Section 3, we examine the ion velocity distribution function and, in particular, the positions of peaks and other peculiar phenomena.

2. Transition to Periodic Steady State: Model Calculations

The theory of Kumar (1981) for time-dependent transport coefficients can be taken over to a large extent even if the electric field is time-dependent. Thus Kumar's equation (12) adapted to a.c. fields becomes

$$(\partial_t + \boldsymbol{a}_0 \cos\omega t\,.\,\partial_{\boldsymbol{v}} + J)\, F^{(n)} = \boldsymbol{v}\, F^{(n-1)} \qquad (n = 0,1,2,\ldots)\,, \tag{4}$$

where $\boldsymbol{a}_0 = e\boldsymbol{E}_0/m$. This gives a hierarchy of equations for spatial moments,

$$F^{(n)}(\boldsymbol{v},t) = \int \mathrm{d}\boldsymbol{r}\, \frac{\boldsymbol{r}^n}{n!}\, f(\boldsymbol{r},\boldsymbol{v},t)\,, \tag{5}$$

and generalised transport coefficients $\omega^{(n)}(t)$ are defined by the equations

$$\partial_t N^{(n)} = \sum_{r=0}^{n} \omega^{(r)}(t)\, N^{(n-r)}\,, \tag{6}$$

where

$$N^{(n)}(t) \equiv \int \mathrm{d}\boldsymbol{v}\, F^{(n)}(\boldsymbol{v},t) \tag{7}$$

are found after the $F^{(n)}$ are calculated from (4).

In the limit of long times, $t \gg \tau_{\mathrm{B}}$ (a timescale defined below),

$$\omega_{\infty}^{(n)}(t) \equiv \{\omega^{(n)}(t)\}_{t\gg\tau_B} \tag{8}$$

determines conventional transport coefficients defined as coefficients of $\nabla^r n$ in the density-gradient expansion of the phase-space distribution function (Kumar *et al.* 1980) and, in particular,

$$\begin{aligned} \alpha_{\mathrm{R}} &= -\,\omega_{\infty}^{(0)}(t) && \text{(net loss rate)},\\ \boldsymbol{W} &= \omega_{\infty}^{(1)}(t) && \text{(drift velocity)},\\ \mathbf{D} &= \omega_{\infty}^{(2)}(t) && \text{(diffusion tensor)}\,. \end{aligned} \tag{9}$$

The proof for time-dependent fields follows along similar lines to that given in Sections 2.2 and 3 of Kumar (1981), and will not be repeated here.

It is, however, a rather more difficult proposition to establish just *how* this transition takes place. While Kumar (1981) was able to demonstrate for d.c. fields that *all* transport properties will relax to their hydrodynamic values with the same time constant, a similar result for a.c. fields would seem very difficult to establish. For this reason, we have opted to use a BGK model collision operator (Bhatnagar *et al.* 1954) as a first step in trying to understand the relaxation process. As mentioned previously, this model perhaps best describes ion motion in the parent gas, where charge-transfer collisions are dominant (Kumar *et al.* 1980), but it is nevertheless intended to offer a qualitative picture of other types of swarms in gases.

To make things as simple as possible, we shall consider only particle-conserving collisions and limit the discussion to one dimension, with variations in space only along the z axis of a system of coordinates, which also defines the direction of $\boldsymbol{a}$. Integration of (4) over tranverse components v_x, v_y of velocity then furnishes the one-dimensional BGK model equation

$$(\partial_t + a_0 \cos\omega t + \nu)\, F^{(n)} = v_z\, F^{(n-1)} + \nu w(\alpha, v_z) \int_{-\infty}^{\infty} \mathrm{d}v'_z\, F^{(n)}(v'_z, t)\,, \tag{10}$$

where ν is a constant, represenative of the ion–gas atom collision frequency,

$$w(\alpha, v_z) = \left(\frac{\alpha^2}{2\pi}\right)^{\frac{1}{2}} \exp(-\alpha^2 v_z^2/2) \tag{11}$$

is a Maxwellian distribution at gas temperature T_g, and

$$\alpha^2 \equiv m/kT_g\,. \tag{12}$$

It is convenient to take a Fourier transform in velocity space,

$$\overline{F}^{(n)}(s,t) = \int_{-\infty}^{\infty} \mathrm{d}v_z\, \mathrm{e}^{\mathrm{i}sv_z}\, F^{(n)}(v_z,t)\,, \tag{13}$$

and hence, by (7),

$$N^{(n)}(t) \equiv \int_{-\infty}^{\infty} \mathrm{d}v_z\, F^{(n)}(v_z,t) = \overline{F}^{(n)}(0,t)\,. \tag{14}$$

The transform of (10) is thus

$$(\partial_t + \mathrm{i}sa_0 \cos\omega t + \nu)\,\overline{F}^{(n)} = \mathrm{i}\frac{\partial}{\partial s}\,\overline{F}^{(n-1)} + \nu\overline{w}(\alpha,s)\,N^{(n)} \qquad (n = 0,1,2,...)\,, \tag{15}$$

where

$$\overline{w}(\alpha,s) = \exp(-s^2/2\alpha^2)\,. \tag{16}$$

By setting $s = 0$ in (15) and using (14), we have

$$\partial_t\,N^{(n)} = (\mathrm{i}\,\partial_s\,\overline{F}^{(n-1)})_{s=0} \qquad (n = 0,1,2,...)\,, \tag{17}$$

and hence

$$\partial_t\,N^{(0)} = 0\,. \tag{18}$$

This last equation merely expresses the conservation of total swarm particle number $N^{(0)}$, under the model conditions assumed. By (6) and (9) then

$$\alpha_R = -\omega^{(0)} = -\frac{1}{N^{(0)}}\,\partial_t\,N^{(0)} \equiv 0\,. \tag{19}$$

Equation (15) can be integrated to give

$$\begin{aligned}\overline{F}^{(n)}(s,t) \;=\; & \overline{F}^{(n)}(s,0)\exp[-\nu t \;-\; (\mathrm{i}sa_0/\omega)\sin\omega t] \\ & + \int_0^t \mathrm{d}\tau \exp[-\nu\tau \;-\; \mathrm{i}sv_\tau(\omega,t)] \\ & \times\; [\nu\overline{w}(\alpha,s)\, N^{(n)}(t-\tau) \\ & + \mathrm{i}\,\partial_s\, \overline{F}^{(n-1)}(s,t-\tau)] \quad (n=0,1,2,...)\,, \end{aligned} \tag{20}$$

where

$$v_\tau(\omega,t) \equiv \frac{a_0}{\omega}[\sin\,\omega t - \sin\omega(t-\tau)] \tag{21}$$

is an important quantity which we shall refer to as the 'spectral velocity'.

Notice that in the asymptotic limit $\nu t \gg 1$, equation (20) yields

$$\begin{aligned}\overline{F}^{(n)}_\infty(s,t) \;=\; & \int_0^\infty \mathrm{d}\tau \exp[-\nu\tau \;-\; \mathrm{i}sv_\tau(\omega,t)] \\ & \times\; [\nu\overline{w}(\alpha,s)N^{(n)}_\infty(t-\tau) \\ & + \mathrm{i}\,\partial_s\, \overline{F}^{(n-1)}_\infty(s,t-\tau)] \quad (n=0,1,2,...)\,. \end{aligned} \tag{22}$$

It is clear that all distribution functions, and therefore all transport coefficients, relax to the periodic steady state described by (22) with the same time constant, $\tau_\mathrm{B} \equiv \nu^{-1}$. This is hardly surprising, since there is only a single relaxation time in the BGK model: it is quite a different matter to establish that this situation pertains for the full Boltzmann collision operator, as Kumar has done for d.c. fields.

We now give explicit expressions for periodic steady-state transport coefficients obtained from (22). Firstly, we set $n=0$ in (22) and make use of the fact that $N^{(0)}$ is a constant by (18), i.e.

$$N^{(0)} = \text{const.} = N^{(0)}_\infty\,. \tag{23}$$

Thus we obtain

$$\overline{F}^{(0)}_\infty(s,t) = \overline{w}(\alpha,s)\nu N^{(0)}_\infty \int_0^\infty \mathrm{d}\tau \exp[-\nu\tau \;-\; \mathrm{i}sv_\tau(\omega,\,t)]\,. \tag{24}$$

From this we have two results:

$$
\begin{aligned}
F^{(0)}_{\infty}(v_z, t) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}s\, \mathrm{e}^{\mathrm{i}sv_z}\, \overline{F}^{(0)}_{\infty}(s, t) \\
&= \nu N^{(0)}_{\infty} \int_{0}^{\infty} \mathrm{d}\tau\, \mathrm{e}^{-\nu\tau}\, w(\alpha, |\, v_z - v_\tau(\omega, t)\,|)\,, \qquad (25)
\end{aligned}
$$

$$
\begin{aligned}
\partial_t\, N^{(1)}_{\infty} &= \mathrm{i}\, \partial_s\, \overline{F}^{(0)}_{\infty}(0, t) \\
&= \nu N^{(0)}_{\infty} \int_{0}^{\infty} \mathrm{d}\tau\; e^{-\nu\tau}\, v_\tau(\omega, t) \\
&= N^{(0)}_{\infty} \frac{a_0}{(\omega^2 + \nu^2)^{\frac{1}{2}}} \cos\,(\omega t - \phi)\,, \qquad (26)
\end{aligned}
$$

where the phase lag ϕ is given by

$$
\tan\phi = \omega/\nu\,. \qquad (27)
$$

We shall examine the distribution function (25) in the cold gas limit in Section 3.
Next set $n = 1$ in (6) to get

$$
\partial_t\, N^{(1)}_{\infty} = \omega^{(0)}_{\infty}\, N^{(1)}_{\infty} + \omega^{(1)}\, N^{(0)}_{\infty}\,,
$$

and hence with (19) and (9)

$$
\begin{aligned}
W = \omega^{(1)}_{\infty} &= \frac{1}{N^{(0)}_{\infty}}\, \partial_t\, N^{(1)}_{\infty} \\
&= \frac{a_0}{(\omega^2 + \nu^2)^{\frac{1}{2}}} \cos\,(\omega t - \phi)\,. \qquad (28)
\end{aligned}
$$

From (26) it follows that

$$
N^{(1)}_{\infty}(t) = \frac{N^{(0)}_{\infty} a_0/\omega}{(\omega^2 + \nu^2)^{\frac{1}{2}}} \sin\,(\omega t - \phi)\,, \qquad (29)
$$

and by setting $n = 1$ in (22), we find

$$
\begin{aligned}
\overline{F}^{(1)}_{\infty}(s, t) &= \int_{0}^{\infty} \mathrm{d}\tau\, \exp[-\nu\tau \;-\; \mathrm{i}sv_\tau(\omega,\, t)] \\
&\quad \times\; [\nu N^{(1)}_{\infty}(t - \tau)\, \overline{w}(\alpha, s) + \mathrm{i}\, \partial_s\, \overline{F}^{(0)}_{\infty}(s, t - \tau)]\,, \qquad (30)
\end{aligned}
$$

from which we can calculate

$$\partial_t N_\infty^{(2)} = \mathrm{i}\,\partial_s \overline{F}_\infty^{(1)}(0,t)\,. \tag{31}$$

For simplicity in what follows, we take a cold gas, $T_{\mathrm{g}} = 0$. Thus, by (16) $\overline{w}(\alpha, s) = 1$ in the above formulas. It then follows by setting $n = 2$ in (6) and using (9) that the longitudinal diffusion coefficient is given by

$$\begin{aligned} D_{||}(t) &= \omega_\infty^{(2)} = \frac{1}{N_\infty^{(0)}}[\partial_t N_\infty^{(2)} - \omega_\infty^{(1)} N_\infty^{(1)}] \\ &= \frac{(a_0/\sqrt{2}\nu)^2}{\nu(1+\omega^2/\nu^2)}\left(1 + \frac{\cos(2\omega t - \phi_{\mathrm{D}})}{1+4\omega^2/\nu^2}\right), \end{aligned} \tag{32}$$

where

$$\phi_{\mathrm{D}} \equiv 2\phi + 2\psi$$

$$\tan\psi \equiv 2\omega/\nu$$

We also note in passing that the ion mean random energy associated with motion in the longitudinal direction has the form

$$kT_{||}(t) = \frac{m(a_0/\sqrt{2}\nu)^2}{1+\omega^2/\nu^2}\left(1 + \frac{\cos(2\omega t - \phi_{\mathrm{e}})}{(1+4\omega^2/\nu^2)^{\frac{1}{2}}.}\right), \tag{33}$$

with

$$\phi_{\mathrm{e}} \equiv 2\phi + \psi\,. \tag{34}$$

Notice also that the useful identity,

$$D_{||}(t) = \int_0^\infty \mathrm{e}^{-\nu\tau}\,\frac{kT_{||}}{m}(t - \tau)\,d\tau\,, \tag{35}$$

may also be readily established.

For applied frequencies that are high compared with the collision frequency, i.e. $\omega \gg \nu$, both $D_{||}$ and $T_{||}$ are negligibly modulated and have values corresponding to an effective d.c. field

$$E_{\mathrm{eff}} = \frac{E_0/\sqrt{2}}{(1+\omega^2/\nu^2)^{\frac{1}{2}}}\,. \tag{36}$$

High-frequency behaviour of this type is well-known. [For a recent discussion see Loureiro (1993))]. For $\omega < \nu$, however, all quantities are significantly modulated, with phase lags for drift velocity, diffusion coefficient and temperature of $\phi \approx \omega/\nu$, $\phi_{\mathrm{D}} \approx 6\omega/\nu$ and $\phi_{\mathrm{e}} \approx 4\omega/\nu$ respectively.

The formulas (28), (32) and (33) for transport quantities are all relatively straightforward expressions. In contrast, the velocity distribution function, from which they derive, is surprisingly rich in structure, as well shall see.

3. Ion Velocity Distribution Function

The ion velocity distribution function (25) has a very peculiar structure in the cold gas limit $T_{\mathrm{g}} \to 0$ or $\alpha \to \infty$, for then

$$w(\alpha, |\, v_z - v_T(\omega, t)\,|) \to \delta\big(v_z - v_T(\omega, t)\big)\,,$$

and (25) integrates to

$$F_{\infty}^{(0)}(v_z, t) = \frac{N^{(0)}\nu}{a_0} \sum_{j=-\infty}^{\infty} \mathrm{e}^{-\nu\tau_j\,\theta(\tau_j)} \Big/ [1 - (\sin\omega t - \omega v_z/a_0)^2]^{\frac{1}{2}} \tag{37}$$

for velocities satisfying

$$|\sin\omega t \;-\; \omega v_z/a_0\,| \;< 1\,, \tag{38}$$

while $F_{\infty}^{(0)}(v_z, t) = 0$ otherwise. The times in the exponential are defined by

$$\tau_j = t + \frac{(-)^j}{\omega}\sin^{-1}(v_z\omega/a_0 \;- \sin\omega t) + \frac{j\pi}{\omega}\,, \tag{39}$$

and only those values of j are considered for which $\tau_j \geq 0$, as indicated by the step function $\theta(\tau_j)$ in (37).

There is a resonance phenomenon for velocities v_z for which the denominator on the right side of (37) vanishes, i.e.

$$\sin\omega t \;- \omega v_z/a_0 = \;\pm\, 1\,, \tag{40}$$

for then $F_{\infty}^{(0)}$ becomes infinite. Similar sharply peaked structure has been observed by Kuypers and Hopman (1990), Liu *et al.* (1990) and Manenschijn *et al.* (1991), and is generally attributed to charge-exchange phenomena. Given that the BGK kinetic equation follows from the full Boltzmann equation for the case of idealised charge-transfer collisions (Kumar *et al.* 1980), we should expect to gain at least good qualitative agreement with the other works cited above.

We have plotted $F_{\infty}^{(0)}\ (v_z, t)$ as a function of v_z for several values of ω/ν and at various times through the cycle in Fig. 1. Notice that the general symmetry properties (3) must also apply to $F_{\infty}^{(0)}$ and, indeed, these can be established directly from (37) by using the following identities:

$$\tau_j(-v_z,\, t\, + \pi/\omega) = \tau_{j+1}(v_z,\, t), \qquad \tau_j(v_z,\, t\, + 2\pi/\omega) = \tau_{j+2}(v_z,\, t)\,. \tag{41}$$

These in turn follow from the definition (39).

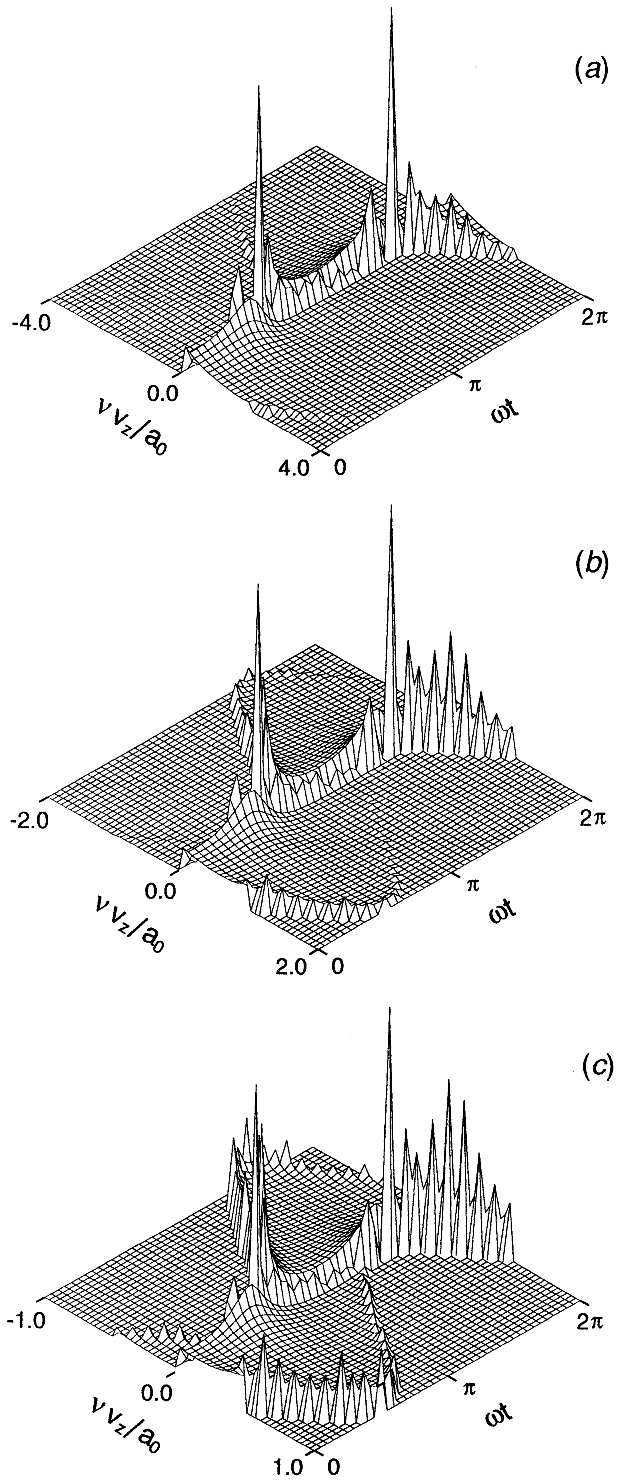

Fig. 1. Asymptotic homogeneous velocity distribution function $F_{\infty}^{(0)}$ (v_z, t) obtained from exact solution of the BGK model kinetic equation assuming a cold gas. Resonance phenomena occur for velocities satisfying (40): (*a*) $\omega/\nu = 0{\cdot}5$, (*b*) $\omega/\nu = 1{\cdot}0$, and (*c*) $\omega/\nu = 2{\cdot}0$.

4. Concluding Remarks

We have extended Kumar's (1981) analysis for a d.c. field to an ion swarm in a radio-frequency field, showing how the relaxation to the periodic steady state takes place by solving exactly the BGK model kinetic equation. This model is best suited to describing ions in their parent gas undergoing predominantly charge-transfer collisions. The velocity distribution function is indeed qualitatively similar to observations reported in the literature, where the charge-transfer collisions are significant.

A more general investigation of the transition to the periodic steady state is warranted, but the mathematical difficulties associated with solving the full Boltzmann equation with a radio-frequency field are daunting, not least because crucial spectral properties of the operators concerned remain largely unexplored. An alternative approach using well-known momentum-transfer theory is currently under investigation.

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