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The Boltzman Equation Theory of Charged Particle Transport

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Abstract

It is shown how a formally exact Kubo-like response theory equivalent to the Boltzmann equation theory of charged particle transport can be constructed. Our response theory gives the general wavevector and time-dependent velocity distribution at any time in terms of an initial distribution function, to which is added the 'response' induced by a generalized 'perturbation' over the intervening time. The usual Kubo linear response result for the distribution function is recovered by choosing the initial velocity distribution to be Maxwellian. For completeness the response theory introduces an exponential convergence function into the 'response' time integral. This is equivalent to using a modified Boltzmann equation but the general form of the transport theory is not changed. The modified transport theory can be used to advantage where possible convergence difficulties occur in numerical solutions of the Boltzmann equation. This paper gives a systematic development of the modified transport theory and shows how our response theory fits into the broader scheme of solving the Boltzmann equation. Our discussion extends both the work of Kumar *et al.* (1980), where the distribution function is expanded out in terms of tensor functions $f^{(J)}$, and the propagator description where the non-hydrodynamic time development of the distribution function is related to the wavevector dependent Green function of the Boltzmann equation.

1. Introduction

This paper examines the formal structure of the Boltzmann equation (BE) theory of charged particle transport in a neutral medium. A number of features are introduced here which are believed to be new. Firstly, our analysis is applied to a slightly broader class of integro-differential equations we refer to here as the modified Boltzmann equation (MBE). The MBE is defined so as to leave the form of the transport theory unchanged. This is not an entirely esoteric development but can be used as a practical means of investigating and minimizing possible numerical convergence difficulties when solving the BE in the analysis of electron and ion swarm data. Secondly, we develop expressions for the MBE distribution function in terms of the BE Green function or propagator and show how the propagator expressions can be used to derive the nonlinear extension of linear response theory (for brief surveys see Chester 1963; Zwanzig 1965; Kubo 1966; McQuarrie 1976). Our nonlinear response theory is applicable to the non-hydrodynamic regime and takes into account particle losses or gains (non-conservative processes). Non-hydrodynamic, non-conservative and nonzero wavevector analogues of the well-known Kubo–Green type time correlation expressions for the drift velocity and diffusion tensor are derived without the usual restriction to a stationary distribution function.

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The paper is organized as follows. Section 2 introduces the MBE and develops the corresponding modified transport theory along lines which are a generalization of the hydrodynamic method of expanding the distribution function out in powers of the spatial gradient of the number density (Kumar *et al.* 1980). This gives non-hydrodynamic modified transport coefficients written in terms of velocity moments of tensor functions $f^{(j)}$. In Section 3, it is shown how formal expressions for the MBE tensors $f^{(j)}$ can be derived in terms of the BE propagator. The non-hydrodynamic extension of the Kubo–Green time correlation relation for the diffusion tensor is derived. In Section 4 it is shown that a special case of the propagator relation for the distribution function is the natural extension of linear response theory to arbitrary field strengths and arbitrary initial distributions. In this nonlinear response theory the collision integral becomes part of a generalized 'perturbation' if the initial distribution is not the Maxwell–Boltzmann distribution. This section also shows that the time-dependent correlation relation for the drift velocity is formally different from that for the diffusion tensor (and other higher order transport coefficients) except in the linear response regime. Section 5 develops the path integral equivalent of the MBE (see Braglia 1980, and references therein for the BE case). Finally, a modified iteration procedure of solving the BE (but using the nonlinear response solution of the MBE) is described in Section 6.

2. Modified Boltzmann Equation and Transport Theory

To avoid later repetition we introduce the new rate parameter s and the MBE from the beginning. We also take particular care to distinguish what are really assumptions or hypotheses about the form of the solution to the MBE from mere definitions. Except for the MBE itself and a few assumptions about the solution at large velocities $\boldsymbol{c}$ and large distances $\boldsymbol{r}$, the transport theory can be constructed using a set of judiciously chosen definitions guided by the review of Kumar *et al.* (1980).

We write the MBE as

$$\{\partial_t+\boldsymbol{c}\cdot\partial_{\boldsymbol{r}}+\boldsymbol{a}(t)\cdot\partial_{\boldsymbol{c}}\}f(\boldsymbol{r},\boldsymbol{c},t;s)+J(f)+s\{f(\boldsymbol{r},\boldsymbol{c},t;s)-g(\boldsymbol{r},\boldsymbol{c},t;s)\}=0\,, \tag{1}$$

where $J(f)$ is the collision integral and $\boldsymbol{a}(t)$ is the acceleration. Equation (1) is not a disguised BE because $g(\boldsymbol{r},\boldsymbol{c},t;s)$ can be chosen independent of $f(\boldsymbol{r},\boldsymbol{c},t;s)$. If it happens that $g(\boldsymbol{r},\boldsymbol{c},t;s)$ is related to $f(\boldsymbol{r},\boldsymbol{c},t;s)$ through a kernel $A(\boldsymbol{c}\,|\,\boldsymbol{c}')$ in a relation of the form

$$g(\boldsymbol{r},\boldsymbol{c},t;s)=\int A(\boldsymbol{c}\,|\,\boldsymbol{c}')f(\boldsymbol{r},\boldsymbol{c}',t;s)\,\mathrm{d}\boldsymbol{c}'\,,$$

then equation (1) reverts to an ordinary BE for all s. Otherwise the BE only corresponds to $s=0$. The modified number density is

$$n(\boldsymbol{r},t;s)=\int f(\boldsymbol{r},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,. \tag{2a}$$

As will be seen below, in order that the form of the transport or continuity equation be the same as for the ordinary BE theory we also require

$$n(\boldsymbol{r},t;s)=\int g(\boldsymbol{r},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,. \tag{2b}$$

The most convenient development of the transport theory employs wavevector $\boldsymbol{k}$ dependent distribution functions and transport coefficients (Kumar and Robson 1973). Defining therefore

$$f(\boldsymbol{k},\boldsymbol{c},t;s) = \int \exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\,f(\boldsymbol{r},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{r}\,, \tag{3a}$$

$$g(\boldsymbol{k},\boldsymbol{c},t;s) = \int \exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\,g(\boldsymbol{r},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{r}\,, \tag{3b}$$

we have

$$\{\partial_t + L(\boldsymbol{k},\boldsymbol{c},t)\}f(\boldsymbol{k},\boldsymbol{c},t;s) + s\{f(\boldsymbol{k},\boldsymbol{c},t;s) - g(\boldsymbol{k},\boldsymbol{c},t;s)\} = 0\,, \tag{4a}$$

where

$$L(\boldsymbol{k},\boldsymbol{c},t)\,f(\boldsymbol{k},\boldsymbol{c},t;s) = \{-\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{c} + \boldsymbol{a}(t)\,.\,\partial_{\boldsymbol{c}}\}f(\boldsymbol{k},\boldsymbol{c},t;s) + J(f)\,. \tag{4b}$$

The definitions (3) assume that all $\boldsymbol{r}^n$ moments of f and g are finite so that $f(\boldsymbol{k},\boldsymbol{c},t;s)$, $g(\boldsymbol{k},\boldsymbol{c},t;s)$ and their velocity moments can be expanded as Taylor series in $\mathrm{i}\boldsymbol{k}$. From equations (2) and (3) we also have

$$N(\boldsymbol{k},t;s) = \int \exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\,n(\boldsymbol{r},t;s)\,\mathrm{d}\boldsymbol{r} \tag{5a}$$

$$= \int f(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} \tag{5b}$$

$$= \int g(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,. \tag{5c}$$

A hierarchy of functions $f^{(j)}$ and $g^{(j)}$ are now introduced by the definitions

$$f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = f(\boldsymbol{k},\boldsymbol{c},t;s)/N(\boldsymbol{k},t;s)\,, \tag{6a}$$

$$g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = g(\boldsymbol{k},\boldsymbol{c},t;s)/N(\boldsymbol{k},t;s)\,, \tag{6b}$$

and for $j \geqslant 1$

$$\boldsymbol{f}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s) = (1/j!)\,\partial^{j}_{\mathrm{i}\boldsymbol{k}}\,f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,, \tag{6c}$$

$$\boldsymbol{g}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s) = (1/j!)\,\partial^{j}_{\mathrm{i}\boldsymbol{k}}\,g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,. \tag{6d}$$

It easily follows from definitions (5) and (6) that

$$\int f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} = \int g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} = 1\,, \tag{7a}$$

$$\int \boldsymbol{f}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} = \int \boldsymbol{g}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} = \boldsymbol{0};\quad j = 1,2,\ldots. \tag{7b}$$

Finally to set up our transport theory we need to define transport coefficients by

$$\omega^{(0)}(\boldsymbol{k},t;s) = -\int J(f^{(0)})\,\mathrm{d}\boldsymbol{c}\,, \tag{8a}$$

$$\boldsymbol{\omega}^{(1)}(\boldsymbol{k},t;s) - \partial_{\mathrm{i}\boldsymbol{k}}\,\omega^{(0)}(\boldsymbol{k},t;s) = \int \boldsymbol{c}\,f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,, \tag{8b}$$

and for $j \geqslant 1$

$$\boldsymbol{\omega}^{(j)}(\boldsymbol{k},t;s) = \{1/(j-1)!\}\partial_{ik}^{j-1}\{\boldsymbol{\omega}^{(1)}(\boldsymbol{k},t;s)-(1-j^{-1})\partial_{ik}\omega^{(0)}(\boldsymbol{k},t;s)\}. \qquad (8c)$$

Integrating equation (4a) over all $\boldsymbol{c}$ and using definitions (5b), (5c), (6a), (6b), (8a) and (8b), we find

$$\{\partial_t - \omega^{(0)*}(\boldsymbol{k},t;s)\}N(\boldsymbol{k},t;s) = 0, \qquad (9a)$$

where

$$\omega^{(0)*}(\boldsymbol{k},t;s) = \omega^{(0)}(\boldsymbol{k},t;s) + i\boldsymbol{k}.\{\boldsymbol{\omega}^{(1)}(\boldsymbol{k},t;s) - \partial_{ik}\omega^{(0)}(\boldsymbol{k},t;s)\}. \qquad (9b)$$

Using the Taylor expansions of $\omega^{(0)}$ and $\boldsymbol{\omega}^{(1)}$ into powers of $i\boldsymbol{k}$ and using the definition (8c), equation (9a) becomes

$$\left(\partial_t - \omega^{(0)}(\boldsymbol{0},t;s) - \sum_{j=1}^{\infty} (i\boldsymbol{k})^j.\boldsymbol{\omega}^{(j)}(\boldsymbol{0},t;s)\right)N(\boldsymbol{k},t;s) = 0. \qquad (9c)$$

By Fourier inversion we find the modified transport equation or continuity equation

$$\{\partial_t - \omega^{(0)}(\boldsymbol{0},t;s)\}n(\boldsymbol{r},t;s) + \partial_{\boldsymbol{r}}.\boldsymbol{\Gamma}(\boldsymbol{r},t;s) = 0, \qquad (10a)$$

where the modified flux is

$$\boldsymbol{\Gamma}(\boldsymbol{r},t;s) = \sum_{j=1}^{\infty} \boldsymbol{\omega}^{(j)}(\boldsymbol{0},t;s).(-\partial_{\boldsymbol{r}})^{j-1}n(\boldsymbol{r},t;s). \qquad (10b)$$

An integro-differential equation for $f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)$ is obtained by combining equations (4a) and (9a) with definitions (6a) and (6b) to give

$$\{\partial_t + \mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) + s\{f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) - g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\} = 0, \qquad (11a)$$

where

$$\mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = \{\boldsymbol{a}(t).\partial_{\boldsymbol{c}} + \omega^{(0)*}(\boldsymbol{k},t;s) - i\boldsymbol{k}.\boldsymbol{c}\}f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) + J(f^{(0)}). \qquad (11b)$$

The hierarchy of integro-differential equations for the $\boldsymbol{f}^{(j)}$ is obtained from successive $i\boldsymbol{k}$ derivatives of equation (11a) using definitions (6c), (6d) and (8c). This gives for $j \geqslant 1$

$$\{\partial_t + \mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}\boldsymbol{f}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s) + s\{\boldsymbol{f}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s) - \boldsymbol{g}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;s)\}$$
$$= \boldsymbol{c}\boldsymbol{f}^{(j-1)}(\boldsymbol{k},\boldsymbol{c},t;s) - \sum_{l=1}^{j} \boldsymbol{\omega}^{(l)*}(\boldsymbol{k},t;s)\boldsymbol{f}^{(j-l)}(\boldsymbol{k},\boldsymbol{c},t;s), \qquad (12a)$$

where

$$\boldsymbol{\omega}^{(l)*}(\boldsymbol{k},t;s) = \boldsymbol{\omega}^{(l)}(\boldsymbol{k},t;s) + i\boldsymbol{k}.\boldsymbol{\omega}^{(l+1)\dagger}(\boldsymbol{k},t;s); \qquad (12b)$$

$$\boldsymbol{\omega}^{(l)\dagger}(\boldsymbol{k},t;s) = \boldsymbol{\omega}^{(l)}(\boldsymbol{k},t;s) - (1/l!)\partial_{ik}^{l}\omega^{(0)}(\boldsymbol{k},t;s). \qquad (12c)$$

By integrating both sides of equations (11a) and (12a) over all $\boldsymbol{c}$ and using the definitions (7) a self-consistent identification of the transport coefficients is found to be

$$\boldsymbol{\omega}^{(j)}(\boldsymbol{k},t;s) = \int \boldsymbol{c}\boldsymbol{f}^{(j-1)}(\boldsymbol{k},\boldsymbol{c},t;s)\,d\boldsymbol{c} - \int J(\boldsymbol{f}^{(j)})\,d\boldsymbol{c}, \qquad (13a)$$

or equivalently by combining relations (6c), (6d), (8a) and (12c)

$$\boldsymbol{\omega}^{(j)\dagger}(\boldsymbol{k},t;s) = \int \boldsymbol{c} f^{(j-1)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,. \tag{13b}$$

Equations (10) show that for practical purposes only the zero wavevector transport coefficients are needed. Further, the transport coefficients measured experimentally are those of the hydrodynamic limit (Kumar *et al.* 1980; equation 10),

$$\boldsymbol{\omega}^{(j)} = \lim_{t\to\infty} \boldsymbol{\omega}^{(j)}(\boldsymbol{0},t;0)\,, \tag{14}$$

and assumed to be time independent. Definition (6a) can be rewritten in the form

$$\begin{aligned} f(\boldsymbol{k},\boldsymbol{c},t;s) &= f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,N(\boldsymbol{k},t;s) \\ &= \sum_{j=0}^{\infty} \boldsymbol{f}^{(j)}(\boldsymbol{0},\boldsymbol{c},t;s)\,.\,(\mathrm{i}\,\boldsymbol{k})^j N(\boldsymbol{k},t;s)\,, \end{aligned} \tag{15a}$$

which can be Fourier inverted to give

$$f(\boldsymbol{r},\boldsymbol{c},t;s) = \sum_{j=0}^{\infty} \boldsymbol{f}^{(j)}(\boldsymbol{0},\boldsymbol{c},t;s)\,.\,(-\partial_{\boldsymbol{r}})^j n(\boldsymbol{r},t;s)\,. \tag{15b}$$

The usual hypothesis of the charged particle Boltzmann equation transport theory with constant acceleration $\boldsymbol{a}$ is that for the long-time or hydrodynamic limit (and $s = 0$) all the $\boldsymbol{f}^{(j)}(\boldsymbol{k},\boldsymbol{c},t;0)$ become time independent. A further condition for stationary solutions when $s \neq 0$ is that all the $\boldsymbol{g}^{(j)}$ are time independent when $t \to \infty$. Stationary $\boldsymbol{f}^{(j)}$ are needed in order to give time-independent transport coefficients. It follows that all the residual time dependence in the hydrodynamic regime is contained in $n(\boldsymbol{r},t;s)$ in equations (10b) and (15b). There are situations where this hypothesis fails because the applied electric field strength is sufficiently great and the energy dependence of the momentum transfer cross section is such that the momentum gained from the field cannot be dissipated by collisions (Cavalleri and Paveri-Fontana 1972); however, we avoid this problem in our analysis.

For completeness and later applications we show how the $\boldsymbol{\omega}^{(j)}(\boldsymbol{k},t;s)$ can be related to various $\boldsymbol{k}$, t and s dependent $\boldsymbol{r}^n$ moments. The case $\boldsymbol{k} = \boldsymbol{0}$ and $s = 0$ has been discussed by Kumar *et al.* (1980). The first moment relation is just equation (9a):

$$\omega^{(0)*}(\boldsymbol{k},t;s) = \{1/N(\boldsymbol{k},t;s)\}\partial_t N(\boldsymbol{k},t;s)\,. \tag{9a$'$}$$

A convenient notation is introduced by rewriting equation (5a) as

$$N(\boldsymbol{k},t;s) = \langle \exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\rangle N(\boldsymbol{0},t;s)\,,$$

so that

$$\partial_{\mathrm{i}\boldsymbol{k}} N(\boldsymbol{k},t;s) = \langle \boldsymbol{r}\exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\rangle N(\boldsymbol{0},t;s)\,.$$

We may then define the vectors

$$\bar{\boldsymbol{r}}(\boldsymbol{k},t;s) = \frac{\langle \boldsymbol{r}\exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\rangle}{\langle \exp(\mathrm{i}\,\boldsymbol{k}\,.\,\boldsymbol{r})\rangle} = \frac{1}{N(\boldsymbol{k},t;s)}\partial_{\mathrm{i}\boldsymbol{k}} N(\boldsymbol{k},t;s)\,, \tag{16a}$$

$$\boldsymbol{r}^* = \boldsymbol{r} - \bar{\boldsymbol{r}}(\boldsymbol{k},t;s)\,. \tag{16b}$$

Then by repeated $\mathrm{i}\boldsymbol{k}$ derivatives of equation (16a) we find for $j \geqslant 2$

$$\overline{\boldsymbol{r}^{*j}}(\boldsymbol{k},t;s) = \partial_{\mathrm{i}\boldsymbol{k}}^{j-1}\bar{\boldsymbol{r}}(\boldsymbol{k},t;s). \tag{16c}$$

By successive $\mathrm{i}\boldsymbol{k}$ derivatives of equation (9a′) using equations (16) we find

$$\boldsymbol{\omega}^{(1)*}(\boldsymbol{k},t;s) = \partial_t\bar{\boldsymbol{r}}(\boldsymbol{k},t;s), \tag{17a}$$

and for $j \geqslant 2$

$$\boldsymbol{\omega}^{(j)*}(\boldsymbol{k},t;s) = (1/j!)\partial_t\overline{\boldsymbol{r}^{*j}}(\boldsymbol{k},t;s). \tag{17b}$$

3. Solutions of MBE in Terms of BE Propagator

In this section we show that solutions for the MBE can be written formally in terms of the BE propagator and three functions that within limits can be chosen arbitrarily. Our discussion here is confined to the case of time-independent charged particle accelerations $\boldsymbol{a}$. Time-dependent accelerations are discussed in Section 4 for the special case referred to as the response solution.

The propagator $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)$ of the BE connecting the initial velocity $\boldsymbol{c}_0$ with final velocity $\boldsymbol{c}$ is defined by the relations

$$P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t) = 0; \qquad t<0, \tag{18a}$$

$$P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},0) = \delta(\boldsymbol{c}-\boldsymbol{c}_0), \tag{18b}$$

$$\{\partial_t + L(\boldsymbol{k},\boldsymbol{c})\}P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t) = 0. \tag{18c}$$

There is no obvious *a priori* method of constructing formal solutions to equations (11a) and (12a). However, with experience the following components of $f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)$ have been useful:

$$f_{\mathrm{A}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = \mathrm{e}^{-st}\frac{N(\boldsymbol{k},0;s)}{N(\boldsymbol{k},t;s)}\int P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)\,f^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,0;s)\,\mathrm{d}\boldsymbol{c}_0, \tag{19a}$$

$$\begin{aligned} f_{\mathrm{B}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = \int\!\!\int_0^t \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}\{L(\boldsymbol{k},\boldsymbol{c})\,P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t') \\ -P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\,L(\boldsymbol{k},\boldsymbol{c}_0)\}\,h^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t', \end{aligned} \tag{19b}$$

$$\begin{aligned} f_{\mathrm{C}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = s\int\!\!\int_0^t \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t') \\ \times\, g^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t', \end{aligned} \tag{19c}$$

where $f^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,0;s)$, $h^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)$ and $g^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)$ are unspecified functions and

$$f^{(0)} = f_{\mathrm{A}}^{(0)} + f_{\mathrm{B}}^{(0)} + f_{\mathrm{C}}^{(0)}. \tag{19d}$$

The components of $f^{(0)}$ satisfy the equations

$$\{\partial_t + \mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}f_X^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) + s f_X^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = 0, \tag{20a}$$

for $X = \mathrm{A},\mathrm{B}$, and

$$\{\partial_t + \mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}f_{\mathrm{C}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) + s\{f_{\mathrm{C}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) - g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\} = 0. \tag{20b}$$

It will be noticed that only $f_A^{(0)}$ relates to $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)$ in the usual way of a propagator which acts on the initial distribution. However, due to s and $\boldsymbol{k}$ nonzero and non-conservative processes, it has been found necessary to introduce $f_B^{(0)}$ and $f_C^{(0)}$ as well. The reasoning that led to the expressions (19a)–(19c) will be made clearer in Section 4. Note how s enters equations (19a)–(19c) through the simple exponential factor e^{-st}.

Extending the definition (6c) to the components of $f^{(0)}$ we define

$$\partial_{\mathrm{i}\boldsymbol{k}} f_X^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = \boldsymbol{f}_X^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s), \tag{21a}$$

where an explicit evaluation of the $\mathrm{i}\boldsymbol{k}$ derivatives shows a natural decomposition into

$$\boldsymbol{f}_X^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \boldsymbol{f}_{X\mathrm{H}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)+\boldsymbol{f}_{X\mathrm{I}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s). \tag{21b}$$

Equation (21b) introduces a 'homogeneous' contribution $\boldsymbol{f}_{X\mathrm{H}}^{(1)}$ and an 'inhomogeneous' contribution $\boldsymbol{f}_{X\mathrm{I}}^{(1)}$ to $\boldsymbol{f}_X^{(1)}$. The integro-differential equations satisfied by the $\boldsymbol{f}_X^{(1)}$ can be deduced from the $\mathrm{i}\boldsymbol{k}$ derivatives of equations (20). Splitting these equations into homogeneous and inhomogeneous parts we have

$$\{\partial_t+\mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}\boldsymbol{f}_{X\mathrm{H}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)+s\boldsymbol{f}_{X\mathrm{H}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = 0, \tag{22a}$$

for X = A, B, and

$$\{\partial_t+\mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}\boldsymbol{f}_{\mathrm{CH}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)+s\{\boldsymbol{f}_{\mathrm{CH}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)-\boldsymbol{g}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)\} = 0, \tag{22b}$$

while for all X the inhomogeneous equations are

$$\{\partial_t+\mathscr{L}(\boldsymbol{k},\boldsymbol{c},t;s)\}\boldsymbol{f}_{X\mathrm{I}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)+s\boldsymbol{f}_{X\mathrm{I}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \{\boldsymbol{c}-\boldsymbol{\omega}^{(1)*}(\boldsymbol{k},t;s)\}f_X^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s). \tag{22c}$$

The general expression for $\boldsymbol{f}^{(1)}$ is

$$\boldsymbol{f}^{(1)} = \boldsymbol{f}_{\mathrm{A}}^{(1)}+\boldsymbol{f}_{\mathrm{B}}^{(1)}+\boldsymbol{f}_{\mathrm{C}}^{(1)}, \tag{22d}$$

whereupon the sum of equations (22a)–(22c) is just equation (12a) for $j = 1$.

Explicit expressions for the $\boldsymbol{f}_{XY}^{(1)}$ are obtainable from the $\mathrm{i}\boldsymbol{k}$ derivative of equations (19a)–(19c). This requires the $\mathrm{i}\boldsymbol{k}$ derivative of $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)$ which, as shown in Appendix 1, can be reduced to a time and velocity integral of the products of two propagators. Also required is the $\mathrm{i}\boldsymbol{k}$ derivative of $N(\boldsymbol{k},t;s)$ which mixes in $\bar{\boldsymbol{r}}(\boldsymbol{k},t;s)$ by equation (16a). Here $\bar{\boldsymbol{r}}(\boldsymbol{k},t;s)$ can be rewritten as a time integral of $\boldsymbol{\omega}^{(1)*}$ (see equation 17a). The details of the calculation are given in Appendix 1 and the results are

$$\boldsymbol{f}_{\mathrm{AH}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \mathrm{e}^{-st}\frac{N(\boldsymbol{k},0;s)}{N(\boldsymbol{k},t;s)}\int P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)\boldsymbol{f}^{(1)}(\boldsymbol{k},\boldsymbol{c}_0,0;s)\,\mathrm{d}\boldsymbol{c}_0, \tag{23a}$$

$$\begin{aligned}\boldsymbol{f}_{\mathrm{BH}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \int\int_0^t \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}\{&L(\boldsymbol{k},\boldsymbol{c})\,P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\\ &-P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\,L(\boldsymbol{k},\boldsymbol{c}_0)\}\boldsymbol{h}^{(1)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t',\end{aligned} \tag{23b}$$

$$\begin{aligned}\boldsymbol{f}_{\mathrm{CH}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = s\int\int_0^t \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}&P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\\ &\times \boldsymbol{g}^{(1)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t',\end{aligned} \tag{23c}$$

$$f_{XI}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \int\int_0^t \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')$$
$$\times\{\boldsymbol{c}_0-\boldsymbol{\omega}^{(1)*}(\boldsymbol{k},t';s)\}f_X^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \tag{23d}$$

Direct time derivatives of both sides of equations (23) can be used to show via equations (9a) and (18) that equations (23) satisfy relations (22a)–(22c). This procedure of building up functions $f_X^{(j)}$ can be followed for higher j values; however, for our discussion of the drift velocity and the diffusion tensor this is unnecessary.

To get physically acceptable results for $f^{(0)}$ and $f^{(1)}$, constraints need to be imposed upon the otherwise free choices of $f^{(0)}$ (at $t = 0$), $g^{(0)}$, $h^{(0)}$ and their $\mathrm{i}\boldsymbol{k}$ derivatives. In particular we require the integrals in equations (19a)–(19c) and (23) to be convergent. For instance the arbitrary functions should go to zero for $|\boldsymbol{c}| \to \infty$. Also, choices such that all $f^{(j)}$ become time independent in the long-time hydrodynamic limit are needed. Finally, the constraints (7) must be satisfied.

To proceed further and derive more specific constraints for the velocity integrals of $h^{(0)}$ and $h^{(1)}$ the two properties derived below are required. Consider the conservative case $\omega^{(0)} = 0$. By integrating equation (18c) over all $\boldsymbol{c}$ we find

$$\int \mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{c}\,P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)\,\mathrm{d}\boldsymbol{c} = \partial_t\int P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)\,\mathrm{d}\boldsymbol{c}. \tag{24}$$

For $\boldsymbol{k} = \boldsymbol{0}$ equation (24) is recognized as the unitarity of $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},t)$, which is then just a conditional probability distribution function. Another relationship which is needed in the hydrodynamic limit is

$$\frac{N(\boldsymbol{k},t-t';s)}{N(\boldsymbol{k},t;s)} = 1 - t'\frac{1}{N(\boldsymbol{k},t;s)}\partial_t N(\boldsymbol{k},t;s) + \frac{t'^2}{2!}\frac{1}{N(\boldsymbol{k},t;s)}\partial_t^2 N(\boldsymbol{k},t;s) + \dots$$
$$+\frac{(-t')^j}{j!}\frac{1}{N(\boldsymbol{k},t;s)}\partial_t^j N(\boldsymbol{k},t;s) + \dots, \tag{25a}$$

which by repeated use of equation (9a′) on the RHS gives

$$\lim_{t\to\infty}\frac{N(\boldsymbol{k},t-t';s)}{N(\boldsymbol{k},t;s)} = \exp\{-\omega^{(0)*}(\boldsymbol{k},\infty;s)t'\}. \tag{25b}$$

Now consider the evaluation for $\omega^{(0)} = 0$ of the RHS of

$$\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{\omega}^{(1)}(\boldsymbol{k},\infty;s) = \lim_{t\to\infty}\int \mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{c}\,f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}. \tag{26}$$

This is easily carried out using equations (24) and (25b) and the explicit expressions (19a)–(19c) for the $f^{(0)}$. For instance for $X = \mathrm{A}$ we find

$$\lim_{t\to\infty}\int \mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{c}\,f_{\mathrm{A}}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}$$
$$= \mathrm{e}^{-st}\frac{N(\boldsymbol{k},0;s)}{N(\boldsymbol{k},t;s)}\partial_t\int\int P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t)\,f^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,0;t)\,\mathrm{d}\boldsymbol{c}\,\mathrm{d}\boldsymbol{c}_0$$
$$= \partial_t\int f_{\mathrm{A}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c} + \{s+\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{\omega}^{(1)}(\boldsymbol{k},\infty;s)\}\int f_{\mathrm{A}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}. \tag{27}$$

More complicated expressions are found for X = B and C, however all have a term similar to the last one on the RHS of equation (27). Then using equation (7) we see that $\mathrm{i}\boldsymbol{k}.\boldsymbol{\omega}^{(1)}(\boldsymbol{k},\infty;s)$ cancels on both sides of equation (26), which leads to the final self-consistency constraint

$$\lim_{t\to\infty}\left(\partial_t\int f_{\mathrm{A}}^{(0)}(f^{(0)}-h^{(0)})\,\mathrm{d}\boldsymbol{c}+s\int f_{\mathrm{A}}^{(0)}(g^{(0)}-h^{(0)})\,\mathrm{d}\boldsymbol{c}\right.$$

$$+\mathrm{i}\boldsymbol{k}.\int\{\boldsymbol{c}\,h^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)-\boldsymbol{\omega}^{(1)}(\boldsymbol{k},\infty;s)f_{\mathrm{A}}^{(0)}(h^{(0)})\}\,\mathrm{d}\boldsymbol{c}$$

$$\left.+\int L(\boldsymbol{k},\boldsymbol{c})\,h^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}-\int f_{\mathrm{A}}^{(0)}(Lh^{(0)})\,\mathrm{d}\boldsymbol{c}\right)=0. \tag{28}$$

The new arguments of $f_{\mathrm{A}}^{(0)}$ here indicate that they replace $f^{(0)}$ at $t=0$ in equation (19a). For $\boldsymbol{k}=\mathbf{0}$ equation (28) for $s\neq 0$ reduces to

$$\int g^{(0)}(\mathbf{0},\boldsymbol{c},0;s)\,\mathrm{d}\boldsymbol{c}=\int h^{(0)}(\mathbf{0},\boldsymbol{c},0;s)\,\mathrm{d}\boldsymbol{c}=1. \tag{29a}$$

The same approach can be applied to $\mathrm{i}\boldsymbol{k}.\boldsymbol{\omega}^{(2)}(\boldsymbol{k},\infty;s)$ using equation (13b) for $j=2$ and equations (23). For $\boldsymbol{k}=\mathbf{0}$ and $s\neq 0$ we find the weak constraint on $\boldsymbol{h}^{(1)}$ of

$$\int \boldsymbol{g}^{(1)}(\mathbf{0},\boldsymbol{c},0;s)\,\mathrm{d}\boldsymbol{c}=\int \boldsymbol{h}^{(1)}(\mathbf{0},\boldsymbol{c},0;s)\,\mathrm{d}\boldsymbol{c}=0. \tag{29b}$$

There is a high degree of freedom in choosing solutions for $\boldsymbol{f}^{(1)}$ because, for instance even with $s\neq 0$, we can put $\boldsymbol{f}^{(1)}=\mathbf{0}$ (at $t=0$), $\boldsymbol{g}^{(1)}=\mathbf{0}$ and $\boldsymbol{h}^{(1)}=\mathbf{0}$ without necessarily violating any of the consistency constraints so far found or equation (12a). Such a choice corresponds to a particular set of transport coefficients $\boldsymbol{\omega}^{(j)}(\boldsymbol{k},t;s)$ in the non-hydrodynamic regime. For $s=0$, or alternatively $g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)=f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)$, and $t\to\infty$ we hypothesize that all the choices give the same transport coefficients. The particular solution obtained with the assumptions above is, from equation (23d),

$$\boldsymbol{f}_{\mathrm{I}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s)=\int\int_0^t \mathrm{e}^{-st'}\frac{N(\boldsymbol{k},t-t';s)}{N(\boldsymbol{k},t;s)}P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t')\{\boldsymbol{c}_0-\boldsymbol{\omega}^{(1)*}(\boldsymbol{k},t-t';s)\}$$

$$\times f^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t-t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \tag{30}$$

Multiplying $\boldsymbol{f}_{\mathrm{I}}^{(1)}$ by $\boldsymbol{c}$ and integrating over all $\boldsymbol{c}$ we obtain an extension of the time correlation expression for the diffusion tensor, except now s and $\boldsymbol{k}$ are arbitrary, non-conservative processes may be included and $f^{(0)}$ is not required to be a stationary time-independent distribution.

To see how the usual form of the Kubo–Green type relation (Green 1954; Kubo 1966, and other reviews; see also Kumar *et al.* 1980, equation 18) for $\boldsymbol{\omega}^{(2)}$ is recovered we set $\omega^{(0)}=0$ and $\boldsymbol{k}=\mathbf{0}$ so that by equations (13b) and (30)

$$\boldsymbol{\omega}^{(2)}(\mathbf{0},t;s)=\int_0^t \mathrm{e}^{-st'}\langle\boldsymbol{c}^*(t)\,\boldsymbol{c}^*(t-t')\rangle_s\,\mathrm{d}t', \tag{31a}$$

where

$$\langle c^*(t)\, c^*(t-t')\rangle_s = \int\!\!\int P(c\,|\,c_0;\mathbf{0},t')\{c-\omega^{(1)}(\mathbf{0},t;s)\}$$

$$\times\, \{c_0-\omega^{(1)}(\mathbf{0},t-t';s)\} f^{(0)}(\mathbf{0},c_0,t-t';s)\,\mathrm{d}c\,\mathrm{d}c_0\,. \tag{31b}$$

It is possible to replace c by $\{c-(\text{any vector})\}$ here as the extra term contributes zero by the integral property (7b) for $f_1^{(1)}$. Equations (31) still have s arbitrary and a non-stationary distribution $f^{(0)}$. The 'correlation' is between c^* at time t and c^* at the earlier time $t-t'$. For a stationary $f^{(0)}$ and $s=0$, $\omega^{(1)}$ is time independent and equations (31) agree with the usual result (Kumar *et al.* 1980). Even if $f^{(0)}$ is not stationary, taking $t\to\infty$ recovers the standard time correlation result anyway as the only important t' dependent term is $P(c\,|\,c_0;\mathbf{0},t')$, provided that this term decays fast enough. This is just an example of the usual hypothesis that all solutions give the same hydrodynamic limit. We should perhaps mention that the agreement in the appropriate limits of equations (31) with the usual time correlation expression for $\omega^{(2)}$ [which is traditionally derived from equation (17b) for $j=2$] further verifies that the velocity moment expressions (13) are mathematically equivalent to the spatial moment equations (17).

4. Generalized Response Theory for Charged Particle Transport

For a time-independent acceleration a, the 'response' solution corresponds to $f^{(0)}=g^{(0)}$ at $t=0$ and $g^{(0)}=h^{(0)}$ for all times in equations (19a)–(19c) and their $\mathrm{i}k$ derivatives. Our discussion is extended here initially to general time-dependent accelerations. Also, a slightly different approach to constructing the response solution to the MBE is developed. Originally, the response solution was encountered for the hydrodynamic limit of the mobility equation (11a), with $k=\mathbf{0}$ and $\omega^{(0)}=0$, when the author observed how the Laplace transform of equation (18c) for $P(c\,|\,c_0;\mathbf{0},t)$ can be used to reconstruct the mobility equation (McMahon 1982). This early result is now generalized here to the non-hydrodynamic regime, arbitrary k vectors and non-conservative processes. The connection between our response solution to the MBE and the Kubo linear response theory is demonstrated. Later in Section 6 the particular advantages of the response expressions for an iterative evaluation of the corrections to any first approximation for the BE distribution function is discussed.

Guided by the early results (McMahon 1982) we define the response solution to equation (4a) as

$$f_{\mathrm{R}}(k,c,t;s) = g(k,c,t;s) - \int\!\!\int_0^t P(c\,|\,c_0;k,t,t')\,\mathrm{e}^{-s(t-t')}$$

$$\times\, \{\partial_{t'}+L(k,c_0,t')\}\, g(k,c_0,t';s)\,\mathrm{d}c_0\,\mathrm{d}t'\,, \tag{32}$$

where $P(c\,|\,c_0;k,t,t')$ is the propagator defined by

$$P(c\,|\,c_0;k,t,t') = 0;\qquad t<t'\,, \tag{33a}$$

$$\lim_{t\to t'} P(c\,|\,c_0;k,t,t') = \delta(c-c_0);\qquad t\geqslant t'\,, \tag{33b}$$

$$\{\partial_t+L(k,c,t)\}P(c\,|\,c_0;k,t,t') = 0\,. \tag{33c}$$

Because $L(\boldsymbol{k},\boldsymbol{c},t)$ is now time dependent the propagator cannot be a function of the time interval $t-t'$ as in Section 3. By differentiating equation (32) on both sides with respect to t and using relations (33) we verify that $f_{\mathrm{R}}(\boldsymbol{k},\boldsymbol{c},t;s)$ satisfies (4a).

In practical calculations with equation (32) it is necessary to be able to calculate the propagator. One method of calculation is to note that $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t')$ itself can be written in the response form

$$P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t') = Q(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t') - \int\int_{t'}^{t} P(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{k},t,t'')$$

$$\times\ \{\partial_{t''}+L(\boldsymbol{k},\boldsymbol{c}',t'')\}Q(\boldsymbol{c}'\,|\,\boldsymbol{c}_0;\boldsymbol{k},t'',t')\,\mathrm{d}t''\,\mathrm{d}\boldsymbol{c}', \tag{34}$$

where $Q(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t')$ is an arbitrary propagator chosen in practice for its convenient properties. For instance Q might be chosen as that for the system with zero electric field. The propagator P on the RHS of (34) can be expressed itself in the form of (34), and so on, to give P as an infinite expansion in powers of Q. Because Q is arbitrary in equation (34) one could use a different propagator Q_j at each stage. We include this possibility for generality whereupon our expansion of P becomes

$$P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t_0) = Q_0(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t_0) + \sum_{l=1}^{\infty}(-)^l\int\ldots\int\int_{t_{l-1}}^{t}\ldots\int_{t_0}^{t} Q_l(\boldsymbol{c}\,|\,\boldsymbol{c}_l;\boldsymbol{k},t,t_l)$$

$$\times\prod_{j=1}^{l}\Big(\{\partial_{t_j}+L(\boldsymbol{k},\boldsymbol{c}_j,t_j)\}Q_{j-1}(\boldsymbol{c}_j\,|\,\boldsymbol{c}_{j-1};\boldsymbol{k},t_j,t_{j-1})\,\mathrm{d}\boldsymbol{c}_j\,\mathrm{d}t_j\Big). \tag{35}$$

The RHS of equation (35) with $t_0=t'$ can be substituted into (32) to give our final and most general response theory expression for the distribution function. For reasons made clearer by our discussion of linear response theory we refer to $\partial_t+L(\boldsymbol{k},\boldsymbol{c},t)$ as a generalized perturbation operator. Equations (32) and (35) can be thought of as generalized responses of the system under the influence of the generalized perturbation operator.

The general response expression for $f_{\mathrm{R}}^{(0)}$ can be obtained from the definitions (6a) and (32) and is

$$f_{\mathrm{R}}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) = g^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) - \int\int_0^t P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t')\,\mathrm{e}^{-s(t-t')}$$

$$\times\ \frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}\{\partial_{t'}+\mathscr{L}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\}g^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \tag{36}$$

That equation (36) for $\boldsymbol{a}$ time independent is a special case of equation (19d) is demonstrated in Appendix 2. One can think of the second term on the RHS of equation (36) as the 'response' to the 'perturbation' arising from the fact that $g^{(0)}$ is not necessarily the exact solution of equation (11a). If $g^{(0)}$ is an exact solution ($g^{(0)}=f^{(0)}$) of (11a) then the response term is identically zero and equation (36) reverts to a tautology.

To see how equation (36) recovers the well-known linear response theory (reviews of the basic ideas may be found in Kubo 1957, 1966; Chester 1963; Zwanzig 1965; McQuarrie 1976) consider $\boldsymbol{k}=\boldsymbol{0}$ and $\omega^{(0)}=0$, and set $g^{(0)}=f_{\mathrm{MB}}$, the time-independent Maxwell–Boltzmann distribution. By detailed balance we have

$$J(f_{\mathrm{MB}}) = 0, \tag{37a}$$

and also

$$\partial_{c0} f_{\mathrm{MB}}(\boldsymbol{c}_0) = -(m/k_{\mathrm{B}}T)\boldsymbol{c}_0 f_{\mathrm{MB}}(\boldsymbol{c}_0), \tag{37b}$$

where m is the mass of the charged particle, T the temperature and k_{B} Boltzmann's constant. For the linear response case P can be approximated by P_{MB}, which is the propagator for the undisturbed thermal system. The linear response distribution function is then just

$$f_{\mathrm{R}}^{(0)}(\mathbf{0}, \boldsymbol{c}, t; s) = f_{\mathrm{MB}}(\boldsymbol{c}) + \frac{m}{k_{\mathrm{B}}T}\int\int_0^t \mathrm{e}^{-st'} P_{\mathrm{MB}}(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \mathbf{0}, t')\, \boldsymbol{a}(t-t') \,.\, \boldsymbol{c}_0 f_{\mathrm{MB}}(\boldsymbol{c}_0)\, \mathrm{d}\boldsymbol{c}_0\, \mathrm{d}t', \tag{38}$$

which agrees with the usual approach. A restriction on the choice of $g^{(0)}$ is that the response term integrals converge. For $t' \to \infty$, $P_{\mathrm{MB}}(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \mathbf{0}, t')$ tends to $f_{\mathrm{MB}}(\boldsymbol{c})$ so that no divergence in the t' integral should occur, even for $s = 0$, because the $\boldsymbol{c}_0$ integral is zero in this limit. A nonzero $\boldsymbol{c}_0$ integral would give unacceptable results for $s = 0$ as has been discussed elsewhere (McMahon 1982).

Consider a time-independent acceleration $\boldsymbol{a}$ and again choose f_{MB} for $g^{(0)}$. To get the exact nonlinear response theory we simply replace P_{MB} by P in equation (38). Then multiplying by $\boldsymbol{c}$ and integrating we have an exact nonlinear and non-hydrodynamic extension of the fluctuation–dissipation theorem (Callen and Walton 1951; Nakano 1956; Kubo 1956, 1957, 1966; Chester 1963):

$$\boldsymbol{\omega}^{(1)}(\mathbf{0}, t; s) = \frac{q\boldsymbol{E}}{k_{\mathrm{B}}T} \cdot \int_0^t \mathrm{e}^{-st'} \langle \boldsymbol{c}(0)\, \boldsymbol{c}(t') \rangle^* \,\mathrm{d}t', \tag{39a}$$

where we have written $\boldsymbol{a} = q\boldsymbol{E}/m$ in terms of the charge q and electric field $\boldsymbol{E}$, and

$$\langle \boldsymbol{c}(0)\, \boldsymbol{c}(t') \rangle^* = \int\int \boldsymbol{c}_0\, \boldsymbol{c}\, P(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \mathbf{0}, t') f_{\mathrm{MB}}(\boldsymbol{c}_0)\, \mathrm{d}\boldsymbol{c}\, \mathrm{d}\boldsymbol{c}_0 . \tag{39b}$$

Notice that for the nonlinear regime the 'correlation function' defined by (39b) is fundamentally different from equation (31b) for the diffusion tensor because of the latter's different weight function $f^{(0)}$, which coincides with f_{MB} only in the linear response limit. There is no difficulty going further and defining a nonlinear response solution for the general N-body system described by the Liouville equation.

Instead of f_{MB} one can choose $g^{(0)}$ as almost any function $f_0^{(0)}$ of velocity. Then for $\boldsymbol{a}$ time independent this response solution is

$$f_{\mathrm{R}}^{(0)}(\mathbf{0}, \boldsymbol{c}, t; s) = f_0^{(0)}(\boldsymbol{c}) - \int\int_0^t \mathrm{e}^{-st'} P(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \mathbf{0}, t') \times \{\boldsymbol{a} \,.\, \partial_{c0} f_0^{(0)}(\boldsymbol{c}_0) + J(f_0^{(0)})\}\, \mathrm{d}\boldsymbol{c}_0\, \mathrm{d}t'. \tag{40}$$

The great variety of response solutions is due to the unlimited choice of initial distributions. The hypothesis is that all solutions agree in the hydrodynamic limit. Particular care is needed with the integration over $\boldsymbol{c}_0$ otherwise a nonzero $\boldsymbol{c}_0$ integral for $t' \to \infty$, where $P(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \mathbf{0}, t') \to f^{(0)}(\mathbf{0}, \boldsymbol{c}, \infty; 0)$, leads to a divergent t' integral for the case $s = 0$. Taking $t \to \infty$, one can regard $f_0^{(0)}$ as the first approximation to $f^{(0)}$ so that the response term represents a formal expression for the correction needed to get the exact distribution. Note that the collision integral contributes to the 'perturbation' in equation (40) in contrast to the more familiar Kubo-type response theory where the collision part does not appear by equation (37a).

The response solution for $\boldsymbol{f}^{(1)}$ can be written generally as

$$f_{\mathrm{R}}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) = \boldsymbol{g}^{(1)}(\boldsymbol{k},\boldsymbol{c},t;s) - \int\int_0^t P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t,t')\,\mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k},t';s)}{N(\boldsymbol{k},t;s)}$$

$$\times\,[\{\partial_{t'}+\mathscr{L}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\}\,\boldsymbol{g}^{(1)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)$$

$$+\{\boldsymbol{\omega}^{(1)*}(\boldsymbol{k},t';s)-\boldsymbol{c}_0\}\,f_{\mathrm{R}}^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)]\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \qquad (41)$$

For $\boldsymbol{a}$ time independent, equation (41) is just (22d) with $\boldsymbol{f}^{(1)} = \boldsymbol{g}^{(1)}$ at $t' = 0$ and $\boldsymbol{g}^{(1)} = \boldsymbol{h}^{(1)}$ for all t'. It is also the $\mathrm{i}\boldsymbol{k}$ derivative of $f_{\mathrm{R}}^{(0)}$. From equation (41) $\boldsymbol{g}^{(1)}$ can be seen to induce a response contribution by virtue of it not being in general the exact solution to equation (12a) for $j = 1$. As with equation (40), the response solution for $\boldsymbol{f}^{(1)}$ can be used to get the correction to a first approximation $\boldsymbol{f}_0^{(1)}$. The result required for our discussion in Section 6 is $\boldsymbol{k} = \boldsymbol{0}$, $\omega^{(0)} = 0$ and $\boldsymbol{a}$ time independent so that

$$f_{\mathrm{R}}^{(1)}(\boldsymbol{0},\boldsymbol{c},t;s) = \boldsymbol{f}_0^{(1)}(\boldsymbol{c}) - \int\int_0^t \mathrm{e}^{-st'}P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},t')[\boldsymbol{a}\,.\,\partial_{\boldsymbol{c}_0}\boldsymbol{f}_0^{(1)}(\boldsymbol{c}_0)+J(\boldsymbol{f}_0^{(1)})$$

$$+\{\boldsymbol{\omega}^{(1)}(\boldsymbol{k},t-t';s)-\boldsymbol{c}_0\}f_{\mathrm{R}}^{(0)}(\boldsymbol{0},\boldsymbol{c}_0,t-t';s)]\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \qquad (42)$$

5. Path Integral Equivalent Equation to MBE

One way of writing down the solution of the MBE at time t is as an infinite sum of terms, the nth term representing the contribution of $n-1$ collisions in the interval $(0,t)$ to the evolution from some initial state (McMahon 1982). Such a sum is an explicit representation of an iterative solution. To construct the iteration it is necessary to break the collision integral up in the form

$$J(f) = \frac{1}{\tau_0(c)}f(\boldsymbol{c}) - \int\frac{K(\boldsymbol{c}\,|\,\boldsymbol{c}')}{\tau_0'(c')}f(\boldsymbol{c}')\,\mathrm{d}\boldsymbol{c}', \qquad (43)$$

where $K(\boldsymbol{c}\,|\,\boldsymbol{c}')$ is a kernel connecting an initial velocity $\boldsymbol{c}'$ to final velocity $\boldsymbol{c}$. The kernel may be thought of as a conditional probability distribution and so must be positive definite and normalized according to

$$\int K(\boldsymbol{c}\,|\,\boldsymbol{c}')\,\mathrm{d}\boldsymbol{c} = \int K(\boldsymbol{c}\,|\,\boldsymbol{c}')\,\mathrm{d}\boldsymbol{c}' = 1\,.$$

The formal mathematical necessity of a positive kernel is to guarantee the convergence of the iteration, as has been emphasized by Vassel (1970) and Braglia (1980). This usually requires the introduction of null collisions (Vassel 1970). With null collisions any desired speed dependence of the collision rate $1/\tau_0(c)$ can be introduced. In equation (43) τ_0' only differs from τ_0 whenever there are non-conservative processes which have the rate

$$\omega^{(0)}(\boldsymbol{k},t;s) = \int\left(\frac{1}{\tau_0'(c)}-\frac{1}{\tau_0(c)}\right)f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)\,\mathrm{d}\boldsymbol{c}\,.$$

To construct the iteration (and hence path integral equation) in the presence of number density gradients it is simplest to proceed from the Green function solution to equation (1) for $s = 0$. The Green function is defined to have the properties

$$G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t) = 0; \qquad t < 0, \tag{44a}$$

$$G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; 0) = \delta(\boldsymbol{r}-\boldsymbol{r}_0)\,\delta(\boldsymbol{c}-\boldsymbol{c}_0), \tag{44b}$$

$$G(\boldsymbol{r}+\Delta\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0+\Delta\boldsymbol{r}, \boldsymbol{c}_0; t) = G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t). \tag{44c}$$

The property (44c) is the requirement that the neutral particle background be spatially uniform. This means that G is actually a function of $\boldsymbol{R} = \boldsymbol{r}-\boldsymbol{r}_0$. The functions G and P are related by

$$P(\boldsymbol{c} \,|\, \boldsymbol{c}_0; \boldsymbol{k}, t) = \int \exp(\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R})\, G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t)\, \mathrm{d}\boldsymbol{R}. \tag{45}$$

The first contribution to G in the interval $(0, t)$ is that of zero collisions. The probability of zero collisions is

$$p_0(t; \boldsymbol{c}) = \exp\left(-\int_0^t \frac{\mathrm{d}t'}{\tau_0(|\boldsymbol{c}+\boldsymbol{a}t'|)}\right), \tag{46}$$

where a time-independent acceleration has been assumed. Then $p_0(t; \boldsymbol{c})$ is a weight function for the zero collision component of G, which is

$$\delta(\boldsymbol{r}-\boldsymbol{r}_0 - \boldsymbol{c}_0 t - \tfrac{1}{2}\boldsymbol{a}t^2)\,\delta(\boldsymbol{c}-\boldsymbol{c}_0-\boldsymbol{a}t)\, p_0(t; \boldsymbol{c}).$$

The contribution of one collision is just the two zero collision parts on the intervals $(0, t_1)$ and (t_1, t) appropriately weighted by zero collision probabilities and connected by the kernel. Written explicitly the first two terms for G are

$$\begin{aligned} G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t) &= \delta(\boldsymbol{r}-\boldsymbol{r}_0 - \boldsymbol{c}_0 t - \tfrac{1}{2}\boldsymbol{a}t^2)\,\delta(\boldsymbol{c}-\boldsymbol{c}_0-\boldsymbol{a}t)\, p_0(t; \boldsymbol{c}_0) \\ &\quad + \int_0^t \mathrm{d}t_1 \int\!\!\int\!\!\int \delta\big(\boldsymbol{r}-\boldsymbol{r}_1-\boldsymbol{c}_1(t-t_1)-\tfrac{1}{2}\boldsymbol{a}(t-t_1)^2\big)\,\delta\big(\boldsymbol{c}-\boldsymbol{c}_1-\boldsymbol{a}(t-t_1)\big) \\ &\qquad \times p_0(t-t_1; \boldsymbol{c}_1)\{K(\boldsymbol{c}_1 \,|\, \boldsymbol{c}_0')/\tau_0'(\boldsymbol{c}_0')\}\delta(\boldsymbol{r}_1-\boldsymbol{r}_0-\boldsymbol{c}_0 t_1 - \tfrac{1}{2}\boldsymbol{a}t_1^2) \\ &\qquad \times \delta(\boldsymbol{c}_0'-\boldsymbol{c}_0-\boldsymbol{a}t_1)\, p_0(t_1; \boldsymbol{c}_0)\, \mathrm{d}\boldsymbol{r}_1\, \mathrm{d}\boldsymbol{c}_1\, \mathrm{d}\boldsymbol{c}_0' + \dots . \end{aligned} \tag{47}$$

Such a series can be formally summed by noticing that G can be re-identified in the RHS of equation (47). A simpler example is given elsewhere (McMahon 1982). The result is the path integral equation

$$\begin{aligned} G(\boldsymbol{r}, \boldsymbol{c} \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t) &= \delta(\boldsymbol{r}-\boldsymbol{r}_0 - \boldsymbol{c}_0 t - \tfrac{1}{2}\boldsymbol{a}t^2)\,\delta(\boldsymbol{c}-\boldsymbol{c}_0-\boldsymbol{a}t)\, p_0(t; \boldsymbol{c}_0) \\ &\quad + \int_0^t \int \frac{K\big(\boldsymbol{c}-\boldsymbol{a}(t-t') | \boldsymbol{c}'\big)}{\tau_0'(\boldsymbol{c}')}\, p_0(t-t'; \boldsymbol{c}-\boldsymbol{a}t) \\ &\qquad \times G\big(\boldsymbol{r}-\boldsymbol{c}(t-t')+\tfrac{1}{2}\boldsymbol{a}(t-t')^2, \boldsymbol{c}' \,|\, \boldsymbol{r}_0, \boldsymbol{c}_0; t'\big)\, \mathrm{d}\boldsymbol{c}'\, \mathrm{d}t'. \end{aligned} \tag{48}$$

That equation (48) is equivalent to the integro-differential equation for G is easily shown by taking the time derivative of both sides and using the identities

$$\partial_t A(\boldsymbol{c}-\boldsymbol{a}t) = -\boldsymbol{a}\,.\,\partial_c A(\boldsymbol{c}-\boldsymbol{a}t),$$

$$\partial_t B(\boldsymbol{r}-\boldsymbol{c}t+\tfrac{1}{2}\boldsymbol{a}t^2) = -(\boldsymbol{a}\,.\,\partial_c+\boldsymbol{c}\,.\,\partial_r)B(\boldsymbol{r}-\boldsymbol{c}t+\tfrac{1}{2}\boldsymbol{a}t^2).$$

Applying definition (45) to equation (48) the path integral equation for P is

$$P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t) = P_0(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t) + \int_0^t\!\!\int M(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{k},t')P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\,\mathrm{d}t'\,\mathrm{d}\boldsymbol{c}', \qquad (49\mathrm{a})$$

where the zero collision propagator is

$$P_0(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t) = \delta(\boldsymbol{c}-\boldsymbol{c}_0-\boldsymbol{a}t)\exp\{\mathrm{i}\boldsymbol{k}\,.\,(\boldsymbol{c}_0 t+\tfrac{1}{2}\boldsymbol{a}t^2)\}\,p_0(t;\boldsymbol{c}_0), \qquad (49\mathrm{b})$$

and the memory kernel M is

$$M(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{k},t') = \int P_0(\boldsymbol{c}\,|\,\boldsymbol{c}_1;\boldsymbol{k},t')\{K(\boldsymbol{c}_1\,|\,\boldsymbol{c}')/\tau_0'(\boldsymbol{c}')\}\,\mathrm{d}\boldsymbol{c}_1. \qquad (49\mathrm{c})$$

The formal summation of expansions like (47) to obtain path integral equations with attendant memory kernel (and related memory functions) is a well-known method employed in the theory of time correlation functions and general relaxation processes (see McMahon 1976, and references therein).

Path integral expressions for $f_{\mathrm{A}}^{(0)}$, $f_{\mathrm{B}}^{(0)}$ and $f_{\mathrm{C}}^{(0)}$ can be obtained by substituting the RHS of equation (49a) into equations (19a)–(19c) in place of P. After some straightforward algebra the $f_X^{(0)}$ can be re-identified in the RHS. We only need the result for the full $f^{(0)}$, which is

$$\begin{aligned} f^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) &= \mathrm{e}^{-st}\exp\{\mathrm{i}\boldsymbol{k}\,.\,(\boldsymbol{c}t-\tfrac{1}{2}\boldsymbol{a}t^2)\}\,p_0(t;\boldsymbol{c}-\boldsymbol{a}t)f^{(0)}(\boldsymbol{k},\boldsymbol{c}-\boldsymbol{a}t,0;s) \\ &\quad + s\int_0^t \mathrm{e}^{-st'}\exp\{\mathrm{i}\boldsymbol{k}\,.\,(\boldsymbol{c}t'-\tfrac{1}{2}\boldsymbol{a}t'^2)\}\,p_0(t';\boldsymbol{c}-\boldsymbol{a}t')\frac{N(\boldsymbol{k},t-t';s)}{N(\boldsymbol{k},t;s)} \\ &\qquad\qquad \times g^{(0)}(\boldsymbol{k},\boldsymbol{c}-\boldsymbol{a}t',t-t';s)\,\mathrm{d}t' \\ &\quad + \int_0^t\!\!\int \mathrm{e}^{-st'}\frac{N(\boldsymbol{k},t-t';s)}{N(\boldsymbol{k},t;s)}M(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{k},t')f^{(0)}(\boldsymbol{k},\boldsymbol{c}',t-t';s)\,\mathrm{d}t'\,\mathrm{d}\boldsymbol{c}'. \end{aligned} \qquad (50)$$

In the special case $s = 0$, $\boldsymbol{k} = \boldsymbol{0}$ and $\omega^{(0)} = 0$, equation (50) is seen to be equivalent to that obtained by other authors (see Braglia 1980, and references therein). Taking $t \to \infty$ the first RHS term goes to zero and the result can be regarded as a detailed balancing relation for $f^{(0)}(\boldsymbol{k},\boldsymbol{c},\infty;s)$.

6. Iterative Technique employing Response Solution

Taking $t \to \infty$ in equation (40) we have for conservative processes

$$f^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s) = f_0^{(0)}(\boldsymbol{c}) - \int \tilde{P}(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},s)\{\boldsymbol{a}\,.\,\partial_{c0}f_0^{(0)}(\boldsymbol{c}_0)+J(f_0^{(0)})\}\,\mathrm{d}\boldsymbol{c}_0, \qquad (51)$$

where $\tilde{P}$ is the Laplace transform of P. This transform diverges for $s = 0$ because for $t \to \infty$, $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},t) \to f^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;0)$. However, with any realistic choice of

$f_0^{(0)}(\boldsymbol{c})$ the overall response term should still converge. The path integral equation for $\tilde{P}$ from equation (49a) is

$$\tilde{P}(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},s) = \tilde{P}_0(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{0},s) + \int \tilde{M}(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{0},s)\,\tilde{P}(\boldsymbol{c}'\,|\,\boldsymbol{c}_0;\boldsymbol{0},s)\,\mathrm{d}\boldsymbol{c}', \tag{52}$$

which can be used to evaluate $f^{(0)}-f_0^{(0)}$ by iteration. Thus we can write formally

$$f^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s)-f_0^{(0)}(\boldsymbol{c}) = \lim_{n\to\infty} \Delta f_n^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s), \tag{53a}$$

where

$$\Delta f_n^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s) = -\int \tilde{P}_n(\boldsymbol{c}\,|\,\boldsymbol{c}_0;0,s)\{\boldsymbol{a}\,.\,\partial_{\boldsymbol{c}_0} f_0^{(0)}(\boldsymbol{c}_0) + J(f_0^{(0)})\}\,\mathrm{d}\boldsymbol{c}_0, \tag{53b}$$

which can be evaluated by the iterative sequence

$$\Delta f_n^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s) = \Delta f_0^{(0)}(\boldsymbol{0},\boldsymbol{c},\infty;s) + \int \tilde{M}(\boldsymbol{c}\,|\,\boldsymbol{c}';\boldsymbol{0},s)\,\Delta f_{n-1}^{(0)}(\boldsymbol{0},\boldsymbol{c}',\infty;s)\,\mathrm{d}\boldsymbol{c}'. \tag{53c}$$

This method is equivalent to using the same $Q_l = P_0$ in equation (35).

Equation (51) is equivalent to (50) for $t\to\infty$, $\boldsymbol{k}=\boldsymbol{0}$ and $\omega^{(0)}=0$ and can be converted to (50) by substituting for $\tilde{P}$ using the RHS of equation (52). Use of the response form of the solution gives rise to an iteration of the inhomogeneous equation (53c) for the correction to the first approximation of $f^{(0)}$, whereas for $s=0$ equation (50) is homogeneous for the full distribution. Iteration of the homogeneous equation has been attempted by Kleban and Davis (1977, 1978) and Kleban *et al.* (1980) and convergence to $f^{(0)}$ with the correct eigenvalue $\lambda = 1$ is guaranteed by a positive definite initial solution (Vassel 1970). The fact that Kleban and coworkers obtained incorrect results may be due to the interaction of errors with the finite grid of velocity points as suggested by Braglia (1980, 1981*a*). We now discuss a number of technical reasons for performing calculations with the new convergence parameter s nonzero.

In any iteration method numerical inaccuracies propagate and affect higher iterations. Consequently, for convergence difficulties to be avoided, a means of adjusting the iteration periodically throughout the calculation is needed. How to do this using the response formulation and $s\neq 0$ and yet obtain convergence to the desired solution for the BE ($s=0$) is discussed in detail in the next paragraph. We note here that $s\neq 0$ can be used to suppress numerical inaccuracies by reducing the effective number of iterations. A simple example of this has been discussed elsewhere (McMahon 1982). This article also suggested $s\neq 0$ as a diagnostic probe to test the sensitivity of any given method of calculation to numerical errors. To see how $s\neq 0$ may suppress numerical errors, consider the iteration of equation (49a) with a speed-independent collision rate $1/\tau_0$. Each iteration adds the contribution of one more collision and so extends the interval $(0,t)$ where the approximation P_n is accurate. For instance, the probability of n collisions in $(0,t)$ takes the Poisson form and is given by

$$p_n(t) = (1/n!)(t/\tau_0)^n \exp(-t/\tau_0).$$

This becomes a maximum at $t_n = n\tau_0$ and so the nth iteration of equation (49a) is adequate up to a time $\approx t_n$. But equation (51) only uses the Laplace tranform of

P so that iterations where $n\tau_0 \gg 1/s$ do not contribute. Hence, the largest iterations with the greatest accumulated inaccuracies are eliminated. A more formal mathematical reason for having $s \neq 0$ also exists. Suppose that τ_0 and τ_0' are speed independent and consider equation (52) in the low field limit $\boldsymbol{a} \approx \mathbf{0}$. The memory kernel is

$$\tilde{M}(\mathbf{0}\,|\,\boldsymbol{c}';\mathbf{0},s) \approx \left(\frac{1}{s\tau_0' + \tau_0'/\tau_0}\right) K(\boldsymbol{c}\,|\,\boldsymbol{c}').$$

Provided that the factor in front of K is less than the lowest eigenvalue λ of K (which is $\lambda = 1$ with eigenvector f_{MB}), then the iteration of equations (52) and (53) converges (Buckner 1962; see also Braglia 1980). Thus either $\tau_0' > \tau_0$ if $s = 0$ (overall charged particle losses) or $s \neq 0$ is needed.

To get convergence to the desired $s = 0$ solution of the BE, yet take advantage of $s \neq 0$ in the calculations, one only needs a hierarchy of calculations with $n \approx (s\tau_0)^{-1}$ iterations each. For instance, the first member of the hierarchy leads to the new approximation for the $g^{(0)}$ function

$$f_1^{(0)}(\mathbf{0},\boldsymbol{c},\infty;s) = f_0^{(0)}(\boldsymbol{c}) + \Delta f_n^{(0)}(\mathbf{0},\boldsymbol{c},\infty;s), \tag{54a}$$

which is then substituted in place of $f_0^{(0)}$ in equation (51) and the process repeated. If convergence occurs by performing sufficiently accurate calculations then $f_m^{(0)}$ approaches the desired $f^{(0)}$ in the limit $m \to \infty$. This means that the effective $g^{(0)}$ used in equation (11a) is made to approach $f^{(0)}$, so that the converged solution is actually independent of the s value used and is just the desired solution to the BE. Convergence may be considered to have occurred when the response correction to $f_m^{(0)}$ is below some preset fraction of $f_m^{(0)}$. In practice equation (54a) is not quite adequate because ideally the integral of $\Delta f_n^{(0)}$ over all $\boldsymbol{c}$ must be exactly zero, and this cannot necessarily be achieved for the calculated $\Delta f_n^{(0)\prime}$. It is necessary to modify $f_n^{(0)\prime}$ to ensure that $f_1^{(0)}$ is properly normalized. Thus, if it happens that

$$\varepsilon_n^{(0)} = \int \Delta f_n^{(0)\prime}\, \mathrm{d}\boldsymbol{c},$$

then equation (54a) should be modified to

$$f_1^{(0)}(\mathbf{0},\boldsymbol{c},\infty;s) = (1+\varepsilon_n^{(0)})^{-1}\{f_0^{(0)}(\boldsymbol{c}) + \Delta f_n^{(0)\prime}(\mathbf{0},\boldsymbol{c},\infty;s)\}. \tag{54b}$$

The same iteration technique with nonzero s can be applied to the calculation of $f^{(1)}$ using equation (43) with $t \to \infty$. Only $\boldsymbol{\omega}^{(1)}(\mathbf{0},\infty;0)$ and $f^{(0)}(\mathbf{0},\boldsymbol{c},\infty;0)$ are available; however, these can be used in place of the nonzero s values in equation (42). In the limit $f_m^{(1)}$ approaches $f^{(1)}$ and the response contribution in equation (42) approaches zero independent of s, so that this choice of $\boldsymbol{\omega}^{(1)}$ and $f^{(0)}$ is acceptable. In theory the velocity integral of each $f_m^{(1)}$ and $\Delta f_n^{(1)}$ is zero. In practice, to eliminate unwanted propagating errors, an adjustment to a calculated $\Delta f_n^{(1)\prime}$ to give a zero velocity integral is needed. A reasonable procedure is for instance

$$\Delta f_n^{(1)} = \Delta f_n^{(1)\prime} - \boldsymbol{\varepsilon}_n^{(1)} f^{(0)}, \tag{55}$$

where $\boldsymbol{\varepsilon}_n^{(1)}$ is the calculated velocity integral of $\Delta f_n^{(1)\prime}$. Finally, it is essential to explicitly impose for $c \to \infty$ the constraints $\Delta f_n^{(j)} \to \mathbf{0}$ and $\partial_c \Delta f_n^{(j)} \to \mathbf{0}$.

As a result of accumulated numerical errors a given sequence of iterations may not produce a solution of the desired MBE. However, using the procedures (54b) and (55), the calculation is periodically 'reset' to formally exact solutions (40) and (42) for $t \to \infty$ and so should further minimize convergence problems. A hierarchy of iterations which employs the path integral equation (50) rather than the response solution can still be used with $s \neq 0$ and get convergence to the BE solution. However, it is more advantageous to use the response method and keep the iterated quantity as small as possible so that the effect of errors is more readily gauged in comparison with the quantity $\Delta f_n^{(0)}$ being calculated.

7. Discussion

Several new aspects of charged particle transport theory have been introduced:

(1) The transport theory has been extended with the introduction of the modified Boltzmann equation and associated modified transport equation. The equivalent path integral equation with arbitrary s and $\boldsymbol{k}$ parameters and including non-conservative effects has been derived.

(2) The degree of arbitrariness in constructing non-hydrodynamic solutions $f_X^{(j)}$ has been demonstrated.

(3) A special form of solution referred to as the response solution has been defined. The way in which this solution reproduces the well-known linear response theory and the fluctuation–dissipation theorem has been demonstrated. The extension of the time correlation expressions for $\boldsymbol{\omega}^{(1)}$ and $\boldsymbol{\omega}^{(2)}$ for non-stationary distributions has been discussed.

(4) The special properties of the response solution for correcting a first approximation for the distribution function has been pointed out. A hierarchy of iterations which employ the new convergence factor s but where convergence is to the solution of the BE has been proposed as a practical means of doing calculations of the distribution function and transport coefficients.

(5) Because the introduction of s can be made without changing the form of the transport equation, there may be advantages in using the new degree of freedom s as a diagnostic variable for comparing, for instance, the results of different methods of calculation which disagree in the desired case $s = 0$ or for investigating instabilities. This would allow more systematic studies to be made without necessarily changing the model cross sections.

It has become clear in recent years that the traditional two-term approximation for electron transport in gases (Huxley and Crompton 1974) becomes inaccurate when the electron–molecule inelastic cross section is comparable with or larger than the elastic cross section (Reid 1979; Lin *et al.* 1979; Pitchford 1981; Pitchford *et al.* 1981; Pitchford and Phelps 1982; Braglia 1981*a*, 1981*b*; Braglia *et al.* 1982) or when the cross sections are highly anisotropic (Reid 1979; Haddad *et al.* 1981). Lin *et al.* (1979), Pitchford *et al.* (1981) and Pitchford and Phelps (1982) have developed new numerical methods of solving the BE beyond the two-term approximation, whereas Braglia (1981*b*) has developed a new, high accuracy Monte Carlo technique with a computational speed comparable with the numerical solutions of the BE. Braglia *et al.* (1982) have used this Monte Carlo technique to confirm most of the results of the abovementioned BE analysis. The response theory approach described in Section 6 is an additional viable approach which can be used to correct by successive iterations the two-term approximation.

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Appendix 1

To establish the expressions (23) for the $f'^{(1)}_{XY}$ via equations (21) we need to develop two general identities. These are

$$\int_0^{t'} \omega^{(1)*}(\boldsymbol{k}, t-t''; s)\, \mathrm{d}t''\, P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t')$$

$$= \int\int_0^{t'} \omega^{(1)*}(\boldsymbol{k}, t-t''; s)\, P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t'-t'')\, P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t'')\, \mathrm{d}t''\, \mathrm{d}\boldsymbol{c}', \tag{A1}$$

$$\partial_{ik} P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t) = \int\int_0^{t} \boldsymbol{c}'\, P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t-t')\, P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t')\, \mathrm{d}\boldsymbol{c}'\, \mathrm{d}t'. \tag{A2}$$

These identities are proved here in the case of a time-independent acceleration $\boldsymbol{a}$.

Equation (A1) is confirmed by taking the t derivative of both sides:

$$\begin{aligned}\partial_t \mathrm{LHS} &= -\int_0^{t'} \partial_{t''}\boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t-t''; s)\,\mathrm{d}t''\, P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t') \\ &= \{\boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t; s) - \boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t-t'; s)\}\, P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t'), \\ \partial_t \mathrm{RHS} &= -\iint_0^{t'} \partial_{t''}\boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t-t''; s)\, P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t'-t'')\, P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t'')\,\mathrm{d}t''\,\mathrm{d}\boldsymbol{c}' \\ &= \partial_t \mathrm{LHS} + \int_0^{t'} \boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t-t''; s) \\ &\qquad \times \partial_{t''}\left(\int P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t'-t'')\, P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t'')\,\mathrm{d}\boldsymbol{c}'\right)\mathrm{d}t''.\end{aligned}$$

It is easily checked that the term within the large parentheses satisfies

$$\{\partial_{t'} + L(\boldsymbol{k}, \boldsymbol{c})\}\left(\int P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t'-t'')\, P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t'')\,\mathrm{d}\boldsymbol{c}'\right) = 0,$$

if $t' > t''$. At $t' = t''$, the value of the term is $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t')$, and hence it is indistinguishable from $P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t')$ for $t'' \leqslant t'$, and so independent of t''. Thus equation (A1) is established.

The argument applied to the proof of equation (A1) is based on the uniqueness assumption, stating that two functions that have the same initial value and satisfy the same integro-differential equation must be equal. The same assumption can be applied to proving equation (A2). By differentiating equation (18c) with respect to $\mathrm{i}\boldsymbol{k}$ we find for $\boldsymbol{a}$ constant

$$\{\partial_t + L(\boldsymbol{k}, \boldsymbol{c})\}\partial_{\mathrm{i}\boldsymbol{k}} P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t) = \boldsymbol{c}\, P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t). \tag{A3}$$

The initial value of $\partial_{\mathrm{i}\boldsymbol{k}} P$ is zero by equation (18b), which agrees with the RHS of (A2). By taking the t derivative of the RHS one finds that it satisfies the same equation (A3) as $\partial_{\mathrm{i}\boldsymbol{k}} P$. Hence (A2) is established.

Now take the $\mathrm{i}\boldsymbol{k}$ derivative of equation (19a) for $f_{\mathrm{A}}^{(0)}$:

$$\begin{aligned}\boldsymbol{f}_{\mathrm{A}}^{(1)}(\boldsymbol{k}, \boldsymbol{c}, t; s) &= \mathrm{e}^{-st}\frac{N(\boldsymbol{k}, 0; s)}{N(\boldsymbol{k}, t; s)}\int \partial_{\mathrm{i}\boldsymbol{k}} P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t)\, f^{(0)}(\boldsymbol{k}, \boldsymbol{c}_0, \boldsymbol{0}; s)\,\mathrm{d}\boldsymbol{c}_0 \\ &\quad - \mathrm{e}^{-st}\frac{N(\boldsymbol{k}, 0; s)}{N(\boldsymbol{k}, t; s)}\{\bar{\boldsymbol{r}}(\boldsymbol{k}, t; s) - \bar{\boldsymbol{r}}(\boldsymbol{k}, 0; s)\} \\ &\quad \times \int P(\boldsymbol{c}\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t)\, f^{(0)}(\boldsymbol{k}, \boldsymbol{c}_0, 0; s)\,\mathrm{d}\boldsymbol{c}_0 + \boldsymbol{f}_{\mathrm{AH}}^{(1)}(\boldsymbol{k}, \boldsymbol{c}, t; s),\end{aligned} \tag{A4}$$

where $\bar{\boldsymbol{r}}$ has been introduced by equation (16a). The vector $\boldsymbol{r}$ can be converted to a time integral of $\boldsymbol{\omega}^{(1)*}$ by equation (17a). The first two terms on the RHS of equation (A4) can be combined using (A1) and (A2); the result is written as

$$\begin{aligned}\boldsymbol{f}_{\mathrm{A}}^{(1)}(\boldsymbol{k}, \boldsymbol{c}, t; s) &= \boldsymbol{f}_{\mathrm{AH}}^{(1)}(\boldsymbol{k}, \boldsymbol{c}, t; s) + \int_0^t\iint \mathrm{e}^{-s(t-t')}\frac{N(\boldsymbol{k}, t'; s)}{N(\boldsymbol{k}, t; s)} P(\boldsymbol{c}\,|\,\boldsymbol{c}'; \boldsymbol{k}, t-t') \\ &\times \{\boldsymbol{c}' - \boldsymbol{\omega}^{(1)*}(\boldsymbol{k}, t'; s)\}\mathrm{e}^{-st'}\frac{N(\boldsymbol{k}, 0; s)}{N(\boldsymbol{k}, t'; s)} P(\boldsymbol{c}'\,|\,\boldsymbol{c}_0; \boldsymbol{k}, t')\, f^{(0)}(\boldsymbol{k}, \boldsymbol{c}_0, 0; s)\,\mathrm{d}\boldsymbol{c}'\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t',\end{aligned}$$

and $f_{\rm A}^{(0)}(\boldsymbol{k},\boldsymbol{c}',t';s)$ is easily re-identified in the last RHS term. This establishes equation (21b) for $X = {\rm A}$ and the form of $f_{\rm AI}^{(1)}$ equation (23d).

The same approach can be applied to calculating $f_{\rm B}^{(1)}$ and $f_{\rm C}^{(1)}$ from $f_{\rm B}^{(0)}$ and $f_{\rm C}^{(0)}$. The details are a little trickier due to the time integrals in equations (19b) and (19c). These require an interchange of the order of the two time integrations present after equations (A1) and (A2) are used. This interchange is possible due to the causality property (18a).

Appendix 2

We demonstrate that the response solution for the time-independent acceleration case is just the special example of equations (19) when $g^{(0)} = f^{(0)}$ at $t = 0$ and $g^{(0)} = h^{(0)}$ at all t. With $X = {\rm I}$, the $\partial_{t'} g^{(0)}$ term in equation (36) can be integrated by parts and gives immediately

$$\begin{aligned} f_{\rm R}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) &= f_{\rm A}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s)+f_{\rm C}^{(0)}(\boldsymbol{k},\boldsymbol{c},t;s) \\ &\quad+\frac{1}{N(\boldsymbol{k},t;s)}\int_0^t\!\!\int \mathrm{e}^{-s(t-t')}[\partial_{t'}\{P(\boldsymbol{c}\,|\,\boldsymbol{c}_0;\boldsymbol{k},t-t')\,N(\boldsymbol{k},t';s)\} \\ &\quad-N(\boldsymbol{k},t';s)\,\mathscr{L}(\boldsymbol{k},\boldsymbol{c}_0,t';s)]g^{(0)}(\boldsymbol{k},\boldsymbol{c}_0,t';s)\,\mathrm{d}\boldsymbol{c}_0\,\mathrm{d}t'. \end{aligned} \tag{A5}$$

The remaining derivative on the RHS can be carried out using equations (9a) and (18c), whereupon the relation (19b) for $f_{\rm B}^{(0)}$ is obtained. Section 3 makes use of the observation that $g^{(0)}$ neither needs to equal $f^{(0)}$ at $t = 0$ nor equal $h^{(0)}$ at all t as in the response solution. Equations (19) were originally suggested to the author by the response relation.

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