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STATISTICAL APPROACH TO THE KINETICS OF NONUNIFORM FLUIDS

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We present a new formalism in Fourier space for the study of spatially nonuniform fluids in nonequilibrium states which generalizes previous work on uniform fluids. Starting from the Liouville equation we obtain a hierarchy of equations for the reduced distribution functions which gives their rate of change at any given order of the system mean density as a sum of a *finite* number of terms. Using a finite-ranged repulsive interaction potential we derive, as a first application of the formalism, the Boltzmann integrodifferential equation for an infinite system which is initially in a "weakly" inhomogeneous state. This is accomplished introducing an *initial* statistical assumption, namely initial molecular chaos; this condition is seen to hold during the time evolution described by the resulting kinetic equation.

1. Introduction

The statistical analysis of fluid dynamics away from equilibrium has pointed out during the last decade or so some outstanding features of present-day kinetic theory, mainly the "long-time tail" effects and the divergences in the nonequilibrium virial expansions[1,2]) which have attracted much attention leading to a better understanding of the subject. The attention has also been concentrated on a more general behavior of fluids through the analysis of soluble models[3]) and computer experiments[4]), and on semiphenomenological theories[5]); the comparison between these approaches seems to indicate that kinetic theory is in good shape. It lasts however a fundamental interest in general microscopic formalisms allowing the study of some basic problems of theory[2,6–8]). In this spirit we present in this paper the generalization to spatially inhomogeneous (or nonuniform) fluids of previous work[9–11]) which

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was restricted to uniform systems and hence prevented of most interesting applications. We also deal here with the concrete problem of a controllable derivation of the familiar integrodifferential Boltzmann equation from the Liouville equation. This is expected to shed some light on the conditions of validity of Boltzmann's equation and on the connection between a reversible microscopic description and an irreversible macroscopic model, i.e., on the precise way in which irreversibility is introduced in the formalism.

The formalism we develop here is concerned with the time evolution of the reduced distribution functions $f_n(\boldsymbol{R}_n, p^n; t)$ where p^n denotes the set of momenta and $\boldsymbol{R}_n$ the position of the center of mass of a subsystem of n particles. By means of an appropriate projection of the BBGKY hierarchy[12]) for the rate of change of the functions $F_n(q^n, p^n; t)$ we obtain a new hierarchy for the functions $f_n(\boldsymbol{R}_n, p^n; t)$ which presents the same formal structure as the one previously obtained[9]) and investigated[9-11]) in connection with spatially homogeneous systems. In the form suitable for the applications in this paper, the rate of change of $f_n(\boldsymbol{R}_n, p^n; t)$ is thus expressed as a sum of two types of terms besides the characteristic streaming term: terms which at finite times are of the order of the mean system (number-) density, D, and which also involve the initial conditions $F_n(q^n, p^n; 0)$, and a term which at finite times is of the order of the square of the mean density. Iterating this equation one is led to familiar non-equilibrium virial expansions; we can write the resulting equations, however, as a sum of a *finite* number of terms (for any order in the density) as an exact consequence of the Liouville equation in the thermodynamic limit for a system with a finite-ranged repulsive interparticle potential.

The formalism accounts for the systems nonuniformity through the dependence of $f_n(\boldsymbol{R}_n, p^n; t)$ on $\boldsymbol{R}_n$. Thus, one may deal with the Fourier components $\hat{f}_n(\boldsymbol{k}, p^n; t)$ where the wave vector $\boldsymbol{k}$ plays the role of an "inhomogeneity parameter". In the case $k = 0$ we recover the functions $f_n(p^n; t)$ relevant in the analysis of uniform systems. The formalism trivially reduces then to the one previously considered[9,10]) which allowed a controllable derivation of kinetic equations. For $\boldsymbol{k}$ "small" the formalism can be applied to the much more interesting spatially inhomogeneous systems where the distances over which the system changes significantly are large compared to the range of the interparticle potential.

In this paper we apply the equations corresponding to the latter case to the investigation of a controllable derivation of Boltzmann's integrodifferential equation. As is well known, the Boltzmann equation constitutes a great step towards understanding the temporal evolution of infinite systems but its original derivation involves uncontrolled approximations and raises serious conceptual problems. The familiar BBGKY approach, on the other hand, fails

to derive that equation in a consistent way, and the alternative statistical approaches developed by several authors present similar difficulties. The situation is especially discouraging in the case of nonuniform systems where one encounters extremely difficult problems related to the existence and uniqueness of the nonlinear inhomogeneous Boltzmann equation[13]). Accordingly, given that this equation is known to be valid as an empirical formula, and given that there is no proof of it being completely correct, the interest of new derivations allowing the analysis of its validity is clear. In particular, beyond the zeroth-order approximation corresponding to homogeneous systems[2,8,9,13]), of primary interest is the situation in which (a) the system is initially "weakly" inhomogeneous and (b) there is initially a condition of "molecular chaos". Under these conditions, we show that the terms of the hierarchy for $f_n(\boldsymbol{R}_n, p^n; t)$ which for finite times are of the order of the mean density, D, together with the streaming term, give rise to Boltzmann's equation in the limit of low mean density and for times longer than the collision time. The term containing the initial conditions vanishes in this limit.

The persistence of conditions (a) and (b) during the kinetic evolution of the system is also investigated. This is intimately related with two interesting questions which are proved in this paper, namely the factorization at any time of the n-body reduced distribution functions in terms of one-body reduced distribution functions and the existence of the corresponding kinetic equation as a consequence of the Liouville equation. Concerning the latter, we can see that the relative importance of spatial correlations at a given time, $t > 0$, is measured in a sense by the relative magnitude of the term of order D^2 (at finite times), i.e. the "remainder" term in our hierarchy, which is presented in a closed form. This question, which was already analysed in connection with the homogeneous case, was translated there into the more transparent one of the existence of a proper limit for a well-defined function when the relative distance of two particles tends to infinity[9]). The problem can be formulated in similar terms in the case of inhomogeneous systems. We mention that an alternative procedure in the investigation of kinetic equations, in particular a proof of the existence of the Boltzmann equation for homogeneous systems which avoids the above restriction, was given elsewhere[11]); this, however, was restricted to a limited class of potential functions.

Section 2 contains the general formalism in phase space while section 3 is devoted to the analysis of the integral kernels appearing in the time evolution equations. In section 4 we translate the formalism to Fourier space. Section 5 contains the analysis of initial conditions; in particular, we introduce the condition of initial weak inhomogeneity in order to state the relevant equations for the applications in this paper. We also show in section 5 that, as a consequence of the initial finite range of the correlations in the infinite

system, the functions $\hat{f}_n(\boldsymbol{k}, p^n; 0)$ factorize into a product of n one-body distribution functions, and the term in our equations containing information about the initial state vanishes in the limit of low density. The latter result is necessary in order to prove the existence of a kinetic equation. It is also proved in section 5 that the relevant collision operator for weakly inhomogeneous systems essentially reduces to the relevant one in the case of homogeneous systems. In section 6 we obtain from our formalism some previously derived results[9,10]) for comparison and completeness; in particular we derive the Boltzmann equation for homogeneous systems. Section 7 contains a derivation of the familiar nonlinear Boltzmann equation for nonuniform systems; we also prove the persistence in time of the factorization of the reduced distribution functions in a certain familiar sense (molecular chaos). The problem of the existence and correctness of Boltzmann's equation can be formulated in simple terms; the conditions for its derivation are analysed in section 8.

2. General time evolution of reduced distribution functions: expansions with a finite number of terms

Let us consider a classical conservative system of N identical point-particles of mass m confined in a bounded region Ω of volume V. The state of the system at any time t can be specified by the N-body distribution function or Gibbs ensemble density $\mu_N(q^N, p^N; t)$ which we choose normalized to unity. Here $q^N = (\boldsymbol{q}_1, \ldots, \boldsymbol{q}_N)$ and $p^N = (\boldsymbol{p}_1, \ldots, \boldsymbol{p}_N)$ denote respectively the coordinates and conjugate momenta of the N particles or, alternatively, the representative point in the associated phase space. Relevant information about kinetics is also contained in the n-body reduced distribution functions $F_n(q^n, p^n; t)$, $n = 1, 2, \ldots$, defined by

$$F_n(q^n, p^n; t) = V^n \int\int \mathrm{d}q^{(N-n)}\, \mathrm{d}p^{(N-n)} \mu_N(q^N, p^N; t), \tag{2.1}$$

where $\mathrm{d}q^{(N-n)} = \mathrm{d}\boldsymbol{q}_{n+1} \ldots \mathrm{d}\boldsymbol{q}_N$ and $\mathrm{d}p^{(N-n)} = \mathrm{d}\boldsymbol{p}_{n+1} \ldots \mathrm{d}\boldsymbol{p}_N$. Integrations with respect to the particle coordinates are restricted to the region Ω; integrations with respect to the momenta are extended to all possible values. We now introduce

$$\boldsymbol{R}_n = \frac{1}{n} \sum_{j=1}^{n} \boldsymbol{q}_j, \quad \boldsymbol{r}_n^{(l)} = \boldsymbol{q}_l - \boldsymbol{q}_n, \quad (l = 1, \ldots, n-1), \tag{2.2a}$$

$$\boldsymbol{P}_n = \sum_{j=1}^{n} \boldsymbol{p}_j, \quad \pi_n^{(l)} = (\boldsymbol{p}_l - \boldsymbol{p}_n)/n^{1/(n-1)}, \quad (l = 1, \ldots, n-1), \tag{2.2b}$$

which correspond respectively to the position of the center of mass, the relative positions, and the corresponding momenta; all of them refer to the subsystem of n particles. Denoting $r_n^{n-1} = (r_n^{(1)}, \ldots, r_n^{(n-1)})$, the change of variables (2.2a) transforms the reduced distribution functions $F_n(q^n, p^n; t)$ into $F_n[q^n(R_n, r_n^{n-1}), p^n; t]$ which we may write as $F_n(R_n, r_n^{n-1}, p^n; t)$. Next we define a new set of n-body reduced distribution functions $f_n(R_n, p^n; t)$ through the relations

$$\begin{aligned} f_n(R_n, p^n; t) &= V^{-(n-1)} \int \mathrm{d}r_n^{n-1} F_n(R_n, r_n^{n-1}, p^n; t) \\ &= V^{-(n-1)} \int \mathrm{d}q^n \, \delta\left(\frac{1}{n}\sum_{j=1}^{n} q_j - R_n\right) F_n(q^n, p^n; t) \\ &= \Delta_n F_n(q^n, p^n; t), \end{aligned} \tag{2.3}$$

with δ the Dirac delta function. The last equality in eq. (2.3) defines the idempotent operator $\Delta_n (\Delta_n^2 = \Delta_n)$. We also introduce the operator Γ_n such that $\Delta_n + \Gamma_n = 1$. The evolution with time of the functions $f_n(R_n, p^n; t)$, which no longer depend on the relative coordinates, is the main subject of the present paper.

The Gibbs ensemble density satisfies the Liouville equation,

$$\partial_t \mu_N = -\mathrm{i} L_N \mu_N, \tag{2.4}$$

where $\partial_t \equiv \partial/\partial t$ and L_N denotes the Liouvillian associated with the set of N particles. To be specific, we consider the Hamiltonian function

$$H_N(q^N, p^N) = \sum_{j=1}^{N} (p_j^2/2m) + \sum_{j>l=1}^{N}\sum \varphi_{jl}, \tag{2.5}$$

where $\varphi_{jl} = \varphi(|r_{jl}|)$ stands for the finite-range repulsive interaction which is assumed to act between particles j and l; $r_{jl} \equiv q_j - q_l$. Thus, the Liouvillian L_n associated with any set of $n = 2, 3, \ldots, N$ particles assumes the explicit form

$$L_n = -\mathrm{i}\sum_{j=1}^{n} (p_j/m) \cdot \nabla_j + \mathrm{i}\sum_{j>l=1}^{n}\sum \nabla_j \varphi_{jl} \cdot \partial_{jl}, \tag{2.6}$$

where we have used the notation $\nabla_j \equiv \partial/\partial q_j$, $\partial_{jl} \equiv \partial/\partial p_j - \partial/\partial p_l$; also $\partial_j \equiv \partial/\partial p_j$, $\tilde{\nabla}_n \equiv \partial/\partial R_n$ will be used.

By integrating eq. (2.4), discarding surface integrals at infinity in phase space and using (2.1) one obtains the set of coupled equations [BBGKY hierarchy[12])]

$$\partial_t F_n = -\mathrm{i} L_n F_n - \mathrm{i}(N-n) V^{-1} \mathscr{L}_{n,n+1} F_{n+1}, \tag{2.7}$$

for the reduced distribution functions $F_n(q^n, p^n; t)$. Here

$$\mathscr{L}_{n,n+1} \equiv \mathrm{i}\sum_{j=1}^{n} \int \mathrm{d}p_{n+1} \int \mathrm{d}q_{n+1} \nabla_j \varphi_{j,n+1} \cdot \partial_j. \tag{2.8}$$

We now apply to the hierarchy (2.7) the projection operators Δ_n and Γ_n defined through eq. (2.3); the resulting equations respectively reduce in the thermodynamic limit ($N \to \infty$, $V \to \infty$, $N/V = D$ finite constant; this limit will hereafter be denoted T-lim) to the two coupled sets of equations

$$\partial_t f_n = -\mathrm{i}\tilde{L}_n f_n - \mathrm{i}D\ \text{T-lim}\ \Delta_n \mathscr{L}_{n,n+1} h_{n+1}, \tag{2.9}$$

$$\partial_t h_n = -\mathrm{i}(L_n - \tilde{L}_n) f_n - \mathrm{i}L_n h_n - \mathrm{i}D\ \text{T-lim}\ \Gamma_n \mathscr{L}_{n,n+1} h_{n+1}, \tag{2.10}$$

where

$$h_n(q^n, p^n; t) \equiv \Gamma_n F_n(q^n, p^n; t) = F_n(q^n, p^n; t) - f_n(\boldsymbol{R}_n, p^n; t). \tag{2.11}$$

Here use has been made of the properties

$$\text{T-lim}\ \Delta_n \mathscr{L}_{n,n+1} f_{n+1} = 0, \quad \text{T-lim}\ \Delta_n L_n F_n = \tilde{L}_n f_n, \tag{2.12}$$

with

$$\tilde{L}_n = -\mathrm{i}(nm)^{-1} \boldsymbol{P}_n \cdot \boldsymbol{\nabla}_n, \tag{2.13}$$

and

$$\mathscr{L}_{n,n+1} f_{n+1} = 0. \tag{2.14}$$

Properties (2.12) are a direct consequence of the finite range of the considered interaction potential; (2.14) holds whenever the sets of n particles are sufficiently far (i.e., at a distance larger than the range of the interparticle forces) from the boundaries of the region Ω. The formal solution of eq. (2.10) obtained by Laplace transforms is

$$h_n(t) = \exp(-\mathrm{i}tL_n) h_n(0) - \mathrm{i} \int_0^t \mathrm{d}\tau \exp(-\mathrm{i}\tau L_n) \times \{(L_n - \tilde{L}_n) f_n(t-\tau) + D\ \text{T-lim}\ \Gamma_n \mathscr{L}_{n,n+1} h_{n+1}(t-\tau)\}. \tag{2.15}$$

Using the same iterative procedure in Laplace space which we used in ref. 10, one finally obtains

$$\begin{aligned} &\partial_t f_n(t) + \mathrm{i}\tilde{L}_n f_n(t) \\ &= \sum_{m=1}^{\infty} D^m \left\{ \int_0^t \mathrm{d}\tau [\partial_\tau K_{n,n+m}(\tau)] f_{n+m}(t+\tau) + K_{n,n+m}(t) h_{n+m}(0) \right. \\ &\quad \left. + \mathrm{i} \int_0^t \mathrm{d}\tau\, K_{n,n+m}(\tau) \tilde{L}_{n+m} f_{n+m}(t-\tau) \right\} \\ &= \sum_{m=1}^{l} D^m \left\{ \int_0^t \mathrm{d}\tau [\partial_\tau K_{n,n+m}(\tau)] f_{n+m}(t+\tau) \right. \end{aligned}$$

$$+ K_{n,n+m}(t)h_{n+m}(0) + \mathrm{i}\int_0^t \mathrm{d}\tau\, K_{n,n+m}(\tau)\tilde{L}_{n+m}f_{n+m}(t-\tau)\Big\}$$

$$- \mathrm{i}D^{l+1}\int_0^t \mathrm{d}\tau\, K_{n,n+l}(\tau)\Gamma_{n+l}\mathscr{L}_{n+l,n+l+1}h_{n+l+1}(t-\tau), \tag{2.16}$$

where we write

$$K_{n,n+m}(t) = (-\mathrm{i})^m\ \text{T-lim}\ \Delta_n\mathscr{L}_{n,n+1} \times \Big\{\prod_{k=1}^{m-1}{}'\Big[\int_{\tau_{k-1}}^t \mathrm{d}\tau_k \exp(-\mathrm{i}\tau_k L_{n+k})\Gamma_{n+k}\mathscr{L}_{n+k,n+k+1}\exp(\mathrm{i}\tau_k L_{n+k+1})\Big]\Big\} \times \exp(-\mathrm{i}tL_{n+m}). \tag{2.17}$$

The product denoted by Π', which is by definition equal to 1 for $m = 1$, is ordered from left to right according to increasing values of the index k; also $\tau_0 = 0$.

Eq. (2.16) for the time evolution of the reduced distribution functions $f_n(\boldsymbol{R}_n, p^n; t)$ will later appear to have a more useful structure than the BBGKY hierarchy. Indeed, while the first expression in (2.16) is equivalent to the usual virial expansions, the second expression in (2.16) presents the advantage of being expressable for a given value of l as a sum of a finite number of terms. An equation for the n-body momentum distribution function with a structure formally similar to (2.16) allowed an interesting treatment of spatially uniform fluids[9-11]). Of particular interest is the case $l = 1$ in eq. (2.16) or, alternatively, the equation which follows upon substitution of an expression of the form (2.15) for $h_{n+1}(t)$ into the last term of eq. (2.9):

$$\partial_t f_n(t) = -\mathrm{i}\tilde{L}_n f_n(t) - D\ \text{T-lim}\int_0^t \mathrm{d}\tau\, \Delta_n\mathscr{L}_{n,n+1}\exp(-\mathrm{i}\tau L_{n+1})(L_{n+1} - \tilde{L}_{n+1})f_{n+1}(t-\tau)$$

$$- D^2\,\text{T-lim}\int_0^t \mathrm{d}\tau\, \Delta_n\mathscr{L}_{n,n+1}\exp(-\mathrm{i}\tau L_{n+1})\Gamma_{n+1}\mathscr{L}_{n+1,n+2}h_{n+2}(t-\tau)$$

$$-\mathrm{i}D\ \text{T-lim}\ \Delta_n\mathscr{L}_{n,n+1}\exp(-\mathrm{i}tL_{n+1})h_{n+1}(0). \tag{2.18}$$

If we write

$$J_{n,n+1,n+2}(t) = \mathrm{i}K_{n,n+1}(t)\Gamma_{n+1}\mathscr{L}_{n+1,n+2}, \tag{2.19}$$

eq. (2.18) can be expressed as

$$\partial_t f_n(\boldsymbol{R}_n; t) = -\mathrm{i}\tilde{L}_n f_n(\boldsymbol{R}_n; t) + \mathrm{i}D \int_0^t \mathrm{d}\tau\, K_{n,n+1}(\tau)(L_{n+1} - \tilde{L}_{n+1}) f_{n+1}(\boldsymbol{R}_{n+1}; t-\tau)$$

$$- D^2 \int_0^t \mathrm{d}\tau\, J_{n,n+1,n+2}(\tau) h_{n+2}(t-\tau) + DK_{n,n+1}(t) h_{n+1}(0). \tag{2.20}$$

Here we have explicitly indicated the dependence of the functions f_n on the center-of-mass coordinates and on time, and omitted the dependence on other variables for simplicity in the notation. Eq. (2.20) gives the time evolution of the reduced distribution functions $f_n(\boldsymbol{R}_n, p^n; t)$ in terms of $f_{n+1}(\boldsymbol{R}_{n+1}, p^{n+1}; t)$, $h_{n+2}(q^{n+2}, p^{n+2}; t) \equiv h_{n+2}(\boldsymbol{R}_{n+2}, r_{n+2}^{n+1}, p^{n+2}; t)$ and certain information about the initial state which is contained in $h_{n+1}(0)$.

3. Formal properties of integral kernels

Before proceeding further in the analysis of the explicit temporal evolution, let us establish certain formal properties of the operator $K_{n,n+1}(t)$ in eq. (2.20). According to (2.17), (2.8) and (2.3)

$$K_{n,n+1}(t) \equiv K_{n,n+1}(\boldsymbol{R}_n, p^n; t) = -\mathrm{i}\,\text{T-lim}\, \Delta_n \mathscr{L}_{n,n+1} \exp(-\mathrm{i}tL_{n+1})$$

$$= \text{T-lim}\, V^{-(n-1)} \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}q^{n+1}\, \delta\left(\frac{1}{n}\sum_{j=1}^{n} \boldsymbol{q}_j - \boldsymbol{R}_n\right)$$

$$\times \sum_{j=1}^{n} \nabla_j \varphi_{j,n+1} \cdot \partial_j \exp(-\mathrm{i}tL_{n+1}). \tag{3.1}$$

We first note that the canonical transformation defined by (2.2) is inverted by the relations

$$\boldsymbol{q}_n = \boldsymbol{R}_n - n^{-1} \sum_{l=1}^{n-1} \boldsymbol{r}_n^{(l)}, \quad \boldsymbol{q}_j = \boldsymbol{r}_n^{(j)} + \boldsymbol{q}_n, \tag{3.2a}$$

$$\boldsymbol{p}_n = n^{-1}\boldsymbol{P}_n - n^{1/(n-1)} n^{-1} \sum_{l=1}^{n-1} \pi_n^{(l)},$$

$$\boldsymbol{p}_j = n^{1/(n-1)} \pi_n^{(j)} + \boldsymbol{p}_n, \tag{3.2b}$$

with $j = 1, \ldots, n-1$. For simplicity we shall write hereafter

$$\boldsymbol{P}_n' = (nm)^{-1} \boldsymbol{P}_n. \tag{3.2c}$$

Using (3.2), the n-body Hamiltonian function (2.5) can be separated

$$H_n(q^n, p^n) = \frac{\boldsymbol{P}_n^2}{2nm} + H_n'(\pi_n^{n-1}) + H_n''(r_n^{n-1}), \tag{3.3}$$

with $\pi_n^{n-1} = (\pi_n^{(1)}, \ldots, \pi_n^{(n-1)})$. Accordingly, the corresponding Liouvillian can be written as a sum,

$$L_n = \tilde{L}_n + L_{r,n}, \tag{3.4}$$

of a part which only depends on the center-of-mass variables, $\tilde{L}_n$ as defined in (2.13), and another part which only depends on the relative variables,

$$L_{r,n} = \mathrm{i} \sum_{j=1}^{n-1} \left(\frac{\partial H_n''}{\partial \boldsymbol{r}^{(j)}} \cdot \frac{\partial}{\partial \boldsymbol{\pi}_n^{(j)}} - \frac{\partial H_n'}{\partial \boldsymbol{\pi}_n^{(j)}} \cdot \frac{\partial}{\partial \boldsymbol{r}_n^{(j)}} \right). \tag{3.5}$$

Given that $\tilde{L}_n$ commutes with $L_{r,n}$, one may also write

$$\exp(-\mathrm{i}tL_n) = \exp(-\mathrm{i}tL_{r,n}) \exp(-\mathrm{i}t\tilde{L}_n). \tag{3.6}$$

Now, using the canonical transformation (2.2) we write the operator $K_{n,n+1}(t)$ in the form

$$\begin{aligned} K_{n,n+1}(t) = \text{T-lim } V^{-(n-1)} \int \mathrm{d}\boldsymbol{p}_{n+1} \iint \mathrm{d}\boldsymbol{R}_{n+1}\, \mathrm{d}r_{n+1}^n \\ \times \delta\left(\boldsymbol{R}_{n+1} + \frac{1}{n(n+1)} \sum_{j=1}^{n} \boldsymbol{r}_{n+1}^{(j)} - \boldsymbol{R}_n \right) \\ \times K'_{n,n+1}(r_{n+1}^n, \pi_{n+1}^n; t) \exp(-\mathrm{i}t\tilde{L}_{n+1}), \end{aligned} \tag{3.7}$$

where $K'_{n,n+1}$ only depends on the relative variables.

The operator (3.7) was already analyzed in ref. 9 in connection with uniform systems; we shall write $K^0_{n,n+1}(t)$ for that operator. It was shown[9]) that $K^0_{n,n+1}(t)$ can be written, due to the finite range of the interaction potential, as a sum

$$K^0_{n,n+1} = \sum_{j=1}^{n} K^{(j,n+1)} \Delta_{(n-1)}, \tag{3.8}$$

where each term contains the two-body operator

$$K^{(j,l)} = \text{T-lim } V^{-1} \int \mathrm{d}\boldsymbol{p}_l \int \mathrm{d}\boldsymbol{q}_l \int \mathrm{d}\boldsymbol{q}_j\, \nabla_j \varphi_{j,l} \cdot \partial_j \exp[-\mathrm{i}tL^{(j,l)}], \tag{3.9}$$

with

$$L^{(j,l)} = -\mathrm{i}[m^{-1}(\boldsymbol{p}_j \cdot \nabla_j + \boldsymbol{p}_l \cdot \nabla_l) - \nabla_j \varphi_{j,l} \cdot \partial_{jl}], \tag{3.10}$$

the corresponding Liouvillian operator. The notation

$$\Delta_{(n-1)} = V^{-(n-1)} \int \cdots \int \mathrm{d}\mathbf{q}_1 \ldots \mathrm{d}\boldsymbol{q}_{j-1}\, \mathrm{d}\boldsymbol{q}_{j+1} \ldots \mathrm{d}\boldsymbol{q}_n, \tag{3.11}$$

is used in (3.8).

We shall prove (see section 5) that the operator $K_{n,n+1}$ defined in eq. (3.7) essentially reduces, in the limit of weakly inhomogeneous systems, to $K^0_{n,n+1}$, i.e. to the relevant operator in the analysis of uniform systems. Given the relation (3.8), it follows that $K_{n,n+1}$ can be essentially written in this limit as a sum of operators which only involve pairs of particles $(j, n+1)$. Thus the interaction between the set of n particles and particle $n+1$, as well as the complete dynamics of the set of $n+1$ particles, can be reduced under these conditions to a two-body problem. Accordingly, we only have to consider binary collisions among the particles in the system as long as we are concerned with the first order in the mean density.

One also notes the property

$$K^{(j,l)}(0)G(\boldsymbol{p}_j, \boldsymbol{p}_l) = 0, \tag{3.12}$$

as an immediate consequence of definition (3.9); here $G(\boldsymbol{p}_j, \boldsymbol{p}_l)$ denotes an arbitrary function of the momenta of particles j and l. Property (3.12) at $t = 0$ will be needed later. Otherwise, we shall be mainly concerned with the asymptotic behavior of the operator $K^{(j,l)}(t)$ for large times (see section 7); this fact further simplifies our dynamical problem. Indeed, one has[9]) for large times (in fact, for times larger than the duration of a collision)

$$\lim_{t\to\infty} K^{(j,l)}(t)G(\boldsymbol{p}_j, \boldsymbol{p}_l) = K_B^{(j,l)}G(\boldsymbol{p}_j, \boldsymbol{p}_l), \tag{3.13}$$

with $K_B^{(j,l)}$ the corresponding Boltzmann collision operator[9,8]):

$$K_B^{(j,l)}G(\boldsymbol{p}_j, \boldsymbol{p}_l) = \int d\boldsymbol{p}_l \iint dx\, dy\, 2m^{-1}|\boldsymbol{p}_j - \boldsymbol{p}_l|[G(\boldsymbol{p}_j^0, \boldsymbol{p}_l^0) - G(\boldsymbol{p}_j, \boldsymbol{p}_l)], \tag{3.14}$$

where $\boldsymbol{p}_j^0$ denotes the momentum of particle j before the collision, and x, y refer to the relative coordinate in a reference frame for which the positive z axis points in the direction $(\boldsymbol{p}_j - \boldsymbol{p}_l)$. In short, the initial $(n+1)$-body problem may be ignored here: only the values of the momenta of particles j and l before and after a collision happen to be relevant quantities in this case.

4. Time evolution of Fourier components

We now consider the Fourier expansion of $F_n(\boldsymbol{R}_n, r_n^{n-1}, p^n; t)$ with respect to the coordinate of the center of mass:

$$F_n(\boldsymbol{R}_n, r_n^{n-1}, p^n; t) = V(2\pi)^{-3} \int d\boldsymbol{k} \exp(i\boldsymbol{k}\cdot\boldsymbol{R}_n)\hat{F}_n(\boldsymbol{k}, r_n^{n-1}, p^n; t), \tag{4.1}$$

where the corresponding Fourier components are given by

$$\hat{F}_n(\boldsymbol{k}, r_n^{n-1}, p^n; t) = V^{-1} \int d\boldsymbol{R}_n \exp(-i\boldsymbol{k}\cdot\boldsymbol{R}_n)F_n(\boldsymbol{R}_n, r_n^{n-1}, p^n; t). \tag{4.2}$$

Note that for $\boldsymbol{k}=0$ we have $\hat{F}_n(0, r_n^{n-1}, p^n; t) \equiv F_n(r_n^{n-1}, p^n; t)$ which are the appropriate distribution functions for the analysis of spatially uniform fluids. We also introduce according to (4.2) the Fourier components

$$\hat{f}_n(\boldsymbol{k}, p^n; t) = V^{-1} \int \mathrm{d}\boldsymbol{R}_n \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_n) f_n(\boldsymbol{R}_n, p^n; t)$$

$$= V^{-n} \int \mathrm{d}q^n \exp\left(-\mathrm{i}n^{-1}\boldsymbol{k} \cdot \sum_{j=1}^{n} \boldsymbol{q}_j\right) F_n(q^n, p^n; t), \tag{4.3}$$

where the second equality follows from (2.3); $\hat{f}_n(0, p^n; t) \equiv f_n(p^n; t)$ coincides with the n-body momentum distribution function. From (2.11), (4.2) and (4.3) we have

$$\hat{h}_n(\boldsymbol{k}, r_n^{n-1}, p^n; t) = \hat{F}_n(\boldsymbol{k}, r_n^{n-1}, p^n; t) - \hat{f}_n(\boldsymbol{k}, p^n; t). \tag{4.4}$$

The term affected by a factor D^2 in eq. (2.20) happens to give no contribution to the main result in this paper. This term will be analysed in sections 6 and 8 and we shall omit it for the moment in our equations. Thus, the Fourier transform of eq. (2.20) and the use of (4.1), the inverse transformation to (4.3) for f_n and f_{n+1}, and the relation between h_{n+1} and $\hat{h}_{n+1}$, leads to the expression

$$\partial_t \hat{f}_n(\boldsymbol{k}; t) = -\mathrm{i}(nm)^{-1} \boldsymbol{P}_n \cdot \boldsymbol{k} \hat{f}_n(\boldsymbol{k}; t)$$

$$- \mathrm{i}D \int_0^t \mathrm{d}\tau \int \mathrm{d}\boldsymbol{k}' \, K''_{n,n+1}(\tau)(L_{n-1} - \tilde{L}_{n+1}) \hat{f}_{n+1}(\boldsymbol{k}'; t-\tau)$$

$$+ D \int \mathrm{d}\boldsymbol{k}' \, K''_{n,n+1}(\tau) \hat{h}_{n+1}(\boldsymbol{k}'; 0). \tag{4.5}$$

Here we have written

$$K''_{n,n+1}(t) \equiv K''_{n,n+1}(\boldsymbol{k}, \boldsymbol{k}', p^n; t)$$

$$= (2\pi)^{-3} \int \mathrm{d}\boldsymbol{R}_n \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_n) K_{n,n+1}(t) \exp(\mathrm{i}\boldsymbol{k}' \cdot \boldsymbol{R}_{n+1}) \tag{4.6}$$

and used the result (3.4), i.e. that $L_{n+1} - \tilde{L}_{n+1} = L_{r,n+1}$ has no dependence on $\boldsymbol{R}_{n+1}$. Using expression (3.7) and the identity

$$\delta(\boldsymbol{x}) = (2\pi)^{-3} \int_{-\infty}^{\infty} \mathrm{d}\boldsymbol{k} \, \mathrm{e}^{-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{x}}, \tag{4.7}$$

one has

$$K''_{n,n+1}(t) = \text{T-lim}(2\pi)^{-6} V^{-(n-1)} \int \mathrm{d}\boldsymbol{p}_{n+1} \iint \mathrm{d}\boldsymbol{R}_{n+1}\, \mathrm{d}r^n_{n+1}$$
$$\times \int \mathrm{d}\boldsymbol{R}_n \int \mathrm{d}\boldsymbol{k}'' \exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_n)$$
$$\times \exp\left\{-\mathrm{i}\boldsymbol{k}''\cdot\left(\boldsymbol{R}_{n+1} + [n(n+1)]^{-1}\sum_{j=1}^{n} \boldsymbol{r}^{(j)}_{n+1} - \boldsymbol{R}_n\right)\right\}$$
$$\times K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; t)\exp\{\mathrm{i}\boldsymbol{k}'\cdot(\boldsymbol{R}_{n+1} - \boldsymbol{P}'_{n+1}t)\}, \tag{4.8}$$

where we have also applied the general property

$$\exp\{-t\boldsymbol{P}'_{n+1}\cdot\tilde{\nabla}_{n+1}\}\phi(\boldsymbol{R}_{n+1}) = \phi(\boldsymbol{R}_{n+1} - \boldsymbol{P}'_{n+1}t). \tag{4.9}$$

The operator $K''_{n,n+1}(t)$ can also be expressed as follows,

$$K''_{n,n+1}(t) = \text{T-lim}\ V^{-(n-1)} \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}r^n_{n+1}\, \delta(\boldsymbol{k}-\boldsymbol{k}')$$
$$\times \exp\left\{-\mathrm{i}\boldsymbol{k}\cdot[n(n+1)]^{-1}\sum_{j=1}^{n} \boldsymbol{r}^{(j)}_{n+1} - \mathrm{i}\boldsymbol{k}'\cdot\boldsymbol{P}'_{n+1}t\right\}$$
$$\times K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; t), \tag{4.10}$$

as a consequence of delta function properties. The use of this expression in eq. (4.5) leads to

$$\partial_t \hat{f}_n(\boldsymbol{k}; t) = -\mathrm{i}\boldsymbol{P}'_n\cdot\boldsymbol{k}\hat{f}_n(\boldsymbol{k}; t)$$
$$-\mathrm{i}D\ \text{T-lim}\ V^{-(n-1)} \int_0^t \mathrm{d}\tau \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}r^n_{n+1}$$
$$\times \exp\left\{-\mathrm{i}\boldsymbol{k}\cdot\left([n(n+1)]^{-1}\sum_{j=1}^{n} \boldsymbol{r}^{(j)}_{n+1} + \boldsymbol{P}'_{n+1}\tau\right)\right\}$$
$$\times K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; \tau) L_{r,n+1}\hat{f}_{n+1}(\boldsymbol{k}; t-\tau) + D\ \text{T-lim}\ V^{-(n-1)}$$
$$\times \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}r^n_{n+1} \exp\left\{-\mathrm{i}\boldsymbol{k}\cdot\left([n(n+1)^{-1}\sum_{j=1}^{n} \boldsymbol{r}^{(j)}_{n+1} + \boldsymbol{P}'_{n+1}t\right)\right\}$$
$$\times K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; t)\hat{h}_{n+1}(\boldsymbol{k}; 0). \tag{4.11}$$

This equation gives the rate of change of the Fourier components, $\hat{f}_n(\boldsymbol{k}, p^n; t)$, of the reduced distribution functions $f_n(\boldsymbol{R}_n, p^n; t)$ in terms of $\hat{f}_{n+1}(\boldsymbol{k}, p^{n+1}; t)$ and the functions $\hat{h}_{n+1}(\boldsymbol{k}, r^n_{n+1}, \boldsymbol{P}_{n+1}, \pi^n_{n+1}; 0)$ which refer to the initial state of the system. No approximation has been introduced up to here. Eq. (4.11) has been derived from the Liouville equation in the thermodynamic limit assuming a finite-ranged interaction potential. We have restricted ourselves, however, to the first order (at finite times) in the number density D in order to

focus attention to the applications in the following sections. In any case, it is to be noted that eq. (2.16) can be easily submitted to the same transformations; this would lead to time evolution equations with a general interest in the theory of fluids.

5. Nonuniform systems; initial conditions

In this paper we are mainly concerned with spatially nonuniform systems which initially present "weak" inhomogeneities; let us first characterize these systems. A homogeneous system would present the property of translational invariance of the n-body distribution functions:

$$F_n(\boldsymbol{q}_1, \ldots, \boldsymbol{q}_n, p^n; 0) = F_n(\boldsymbol{q}_1 + \boldsymbol{a}, \ldots, \boldsymbol{q}_n + \boldsymbol{a}, p^n; 0),$$

with $\boldsymbol{a}$ an arbitrary vector in Ω. Accordingly, the functions $F_n(\boldsymbol{R}_n, r_n^{n-1}, p^n; 0)$ will not depend on $\boldsymbol{R}_n$ and we have $\boldsymbol{k} = 0$ in $\hat{f}_n(\boldsymbol{k}, p^n; 0)$ for a homogeneous system. Thus, a "weakly" inhomogeneous initial state can be roughly characterized by a "soft" dependence of $F_n(0)$ on $\boldsymbol{R}_n$, i.e. "small" derivatives with respect to $\boldsymbol{R}_n$. Indeed, for such a state one expects the difference between F_n at two points to be small even in the case that the points are separated by a macroscopic distance. Then we can associate a characteristic length, λ, to the gradient $\tilde{\nabla}_n F_n(0)$ such that $L/\lambda \approx L|\tilde{\nabla}_n F_n(0)|$, with L a macroscopic length, is small. From eq. (4.1) it follows that for a weakly inhomogeneous system the magnitude of the vector $\boldsymbol{k}$ at $t = 0$ is small, and of the order of $1/\lambda$.

We can introduce now a new wave vector scale defined by

$$\boldsymbol{k} = D\boldsymbol{\kappa}. \tag{5.1}$$

This can be seen as an initial property of our system which is made up by particles not influenced by external forces; indeed, one can relate the characteristic length λ in these cases to the free distance between particles. Eq. (4.11) shows that relation (5.1) will persist during the subsequent evolution. The utility of change (5.1) shows up in the limit of low mean density and large time,

$$D \to 0, \quad t \to \infty, \quad Dt = s \text{ finite}, \tag{5.2}$$

which shall be introduced later in our equations. For the moment we note that using (5.1) the term $\exp\{-\mathrm{i}[D\boldsymbol{\kappa}/n(n+1)] \cdot \Sigma_j \boldsymbol{r}_{n+1}^{(j)}\}$ in eq. (4.11), where the range of the relative coordinates $\boldsymbol{r}_{n+1}^{(j)}$ is limited by the finite range of the interparticle potential contained in the operator $K'_{n,n+1}$ [compare eqs. (3.7) and (3.1)], reduces to unity in the limit (5.2). If we drop this term, still considering the original time scale, eq. (4.11) can be written with the change (5.1), using

the same notation for $\hat{h}_n$ and $\hat{f}_n$, in the form

$$\partial_t \hat{f}_n = -\mathrm{i}D\boldsymbol{P}'_n \cdot \boldsymbol{\kappa}\hat{f}_n(\boldsymbol{\kappa}, t) - \mathrm{i}D\ \text{T-lim}\ V^{-n} \int_0^t \mathrm{d}\tau \int \mathrm{d}\boldsymbol{p}_{n+1} \iint \mathrm{d}\boldsymbol{R}_{n+1}\, \mathrm{d}r^n_{n+1}$$
$$\times K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; \tau) \exp(-\mathrm{i}\tau\tilde{L}_{n+1})$$
$$\times L_{r,n+1} \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}\tau\}\hat{f}_{n+1}(\boldsymbol{\kappa}; t-\tau)$$
$$+ D\ \text{T-lim}\ V^{-n} \int \mathrm{d}\boldsymbol{p}_{n+1} \iint \mathrm{d}\boldsymbol{R}_{n+1}\, \mathrm{d}r^n_{n+1}\, K'_{n,n+1}(r^n_{n+1}, \pi^n_{n+1}; t)$$
$$\times \exp(-\mathrm{i}t\tilde{L}_{n+1}) \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}t\}\hat{h}_{n+1}(\boldsymbol{\kappa}; 0). \tag{5.3}$$

The actual form of the last two terms in eq. (5.3) is obtained after introducing $V^{-1}\int \mathrm{d}\boldsymbol{R}_{n+1} \exp(-\mathrm{i}t\tilde{L}_{n+1})$ which only acts as a factor given that the dependence on $\boldsymbol{R}_{n+1}$ was already removed. Using the canonical change of the variables (3.2) we obtain [cf. eqs. (3.1) and (3.7)]

$$\partial_t \hat{f}_n(\boldsymbol{\kappa}; t) = -\mathrm{i}D\boldsymbol{P}'_n \cdot \boldsymbol{\kappa}\hat{f}_n(\boldsymbol{\kappa}, t) - \mathrm{i}D\ \text{T-lim}\ V^{-n} \int_0^t \mathrm{d}\tau \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}q^{n+1}$$
$$\times \sum_{j=1}^{n} \nabla_j \varphi_{j,n+1} \cdot \partial_j \exp(-\mathrm{i}\tau L_{n+1}) L_{n+1}$$
$$\times \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}\tau\}\hat{f}_{n+1}(\boldsymbol{\kappa}; t-\tau)$$
$$+ D\ \text{T-lim}\ V^{-n} \int \mathrm{d}\boldsymbol{p}_{n+1} \int \mathrm{d}q^{n+1} \sum_{j=1}^{n} \nabla_j \varphi_{j,n+1} \cdot \partial_j \exp(-\mathrm{i}tL_{n+1})$$
$$\times \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}t\}\hat{h}_{n+1}(\boldsymbol{\kappa}; 0), \tag{5.4}$$

where the operator $\tilde{L}_{n+1}$ contained [see eq. (3.4)] in $L_{r,n+1}$ appearing in the second term of the right-hand side of eq. (5.3), which acted on a function independent of $\boldsymbol{R}_{n+1}$, has been omitted. Simplifying the notation, one may write eq. (5.4) in the form

$$\partial_t \hat{f}_n(\boldsymbol{\kappa}, p^n; t) = -\mathrm{i}D\boldsymbol{P}'_n \cdot \boldsymbol{\kappa}\hat{f}_n(\boldsymbol{\kappa}, p^n; t)$$
$$+ D \int_0^t \mathrm{d}\tau[\partial_\tau K^0_{n,n+1}(\tau)] \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}\tau\}\hat{f}_{n+1}(\boldsymbol{\kappa}, p^{n+1}; t-\tau)$$
$$+ DK^0_{n,n+1}(t) \exp\{-\mathrm{i}D\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1}t\}\hat{h}_{n+1}(\boldsymbol{\kappa}, r^n_{n+1}, p^{n+1}; 0), \tag{5.5}$$

where the operator $K^0_{n,n+1}(t)$ was defined and analysed in section 3. This

equation can be finally written:

$$\partial_t \hat{f}_n(\boldsymbol{\kappa}, p^n; t) = -\mathrm{i}D\boldsymbol{P}'_n \cdot \boldsymbol{\kappa}\hat{f}_n(\boldsymbol{\kappa}, p^n; t) + D\sum_{j=1}^{n}\Big[\int_0^t \mathrm{d}\tau(\partial_\tau K^{(j,n+1)}(\tau))\exp\{-\mathrm{i}D\boldsymbol{\kappa}\cdot\boldsymbol{P}'_{n+1}\tau\} \times \hat{f}_{n+1}(\boldsymbol{\kappa}, p^{n+1};\ t-\tau) + K^{(j,n+1)}(t)\exp\{-\mathrm{i}D\boldsymbol{\kappa}\cdot\boldsymbol{P}'_{n+1}t\} \times \Delta_{(n-1)}\hat{h}_{n+1}(\boldsymbol{\kappa}, r^n_{n+1}, p^{n+1}; 0)\Big], \tag{5.6}$$

where use has been made of property (3.8).

Before extracting any more consequences from eq. (5.6) let us consider in Fourier formalism the initial factorization of the reduced distribution functions.

We may assume that all correlations in the infinite system (i.e. in the T-limit in our formulation) are finite ranged at time $t = 0$. In particular, we can write the functions $F_n(0)$ in the form

$$F_n(q^n, p^n; 0) = \prod_{j=1}^{n} F_1(\boldsymbol{q}_j, \boldsymbol{p}_j; 0)[1 + g_n(q^n, p^n; 0)], \tag{5.7}$$

which defines the correlation functions $g_n(q^n, p^n; 0)$, where

$$g_n(q^n, p^n; 0) = 0, \quad \text{if } |\boldsymbol{r}_{jl}| \geqslant \xi, \quad \text{for all pairs } (j, l), \tag{5.8}$$

here ξ is a characteristic length of the order of the (finite) range of the interparticle forces. Applying the transformation (4.3) to (5.7) we have

$$\hat{f}_n(\boldsymbol{k}, p^n; 0) = V^{-n}\int \mathrm{d}q^n \prod_{j=1}^{n}\exp(-\mathrm{i}k\cdot\boldsymbol{q}_j/n)F_1(\boldsymbol{q}_j, \boldsymbol{p}_j; 0)[1 + g_n(q^n, p^n; 0)]. \tag{5.9}$$

We note that the second term in the right hand side of this expression vanishes in the T-limit in view of property (5.8); thus, using (4.3) for $n = 1$, (5.9) reduces to

$$\hat{f}_n(\boldsymbol{k}, p^n; 0) = \prod_{j=1}^{n}\hat{F}_1(\boldsymbol{k}/n, \boldsymbol{p}_j; 0). \tag{5.10}$$

That is, as a consequence of the assumption (5.8) for the initial state, the Fourier components $\hat{f}_n$ referring to the infinite system are factorized at $t = 0$.

Another interesting question which can be analysed at this stage concerns the influence of initial conditions on the time evolution of the system; this is relevant in order to obtain a true kinetic equation. The explicit character of the present formalism allows us to prove that the last term on the right-hand

side of eq. (5.6), the one which contains the contribution from initial conditions, vanishes in the limit (5.2) when (5.8) is assumed. To this end we first note the relation

$$\begin{aligned}
&\Delta_{(n-1)}\hat{F}_{n+1}(\boldsymbol{\kappa}, r_{n+1}^{n}, p^{n+1}; 0) \\
&\quad = V(2\pi)^{-3(n+1)} \int \mathrm{d}\boldsymbol{R}_{n+1} \exp(-\mathrm{i}\boldsymbol{\kappa}\cdot\boldsymbol{R}_{n+1}) \\
&\qquad \times \int \mathrm{d}\boldsymbol{k}_j \int \mathrm{d}\boldsymbol{k}_{n+1} \hat{F}_1(\boldsymbol{k}_j, \boldsymbol{p}_j; 0)\hat{F}_1(\boldsymbol{k}_{n+1}, \boldsymbol{p}_{n+1}; 0) \\
&\qquad \times \prod_{\substack{l=1 \\ l\neq j}}^{n} \int \mathrm{d}\boldsymbol{k}_l \int \mathrm{d}\boldsymbol{r}_{n+1}^{(l)} \exp(\mathrm{i}\boldsymbol{k}_l\cdot\boldsymbol{q}_l)\hat{F}_1(\boldsymbol{k}_l, \boldsymbol{q}_l; 0) \exp\{\mathrm{i}(\boldsymbol{k}_j\cdot\boldsymbol{q}_j + \boldsymbol{k}_{n+1}\cdot\boldsymbol{q}_{n+1})\} \\
&\qquad \times [1 + g_{n+1}(q^{n+1}, p^{n+1}; 0)],
\end{aligned} \tag{5.11}$$

which follows from definition (5.7) when one applies the transformation (4.2) and the operator (3.11), and uses expression (4.1) for $n = 1$. We shall assume, in addition to (5.8), that the correlation functions $g_{n+1}(0)$ have no dependence on $\boldsymbol{R}_{n+1}$, and that the integrations to which they are subject in (5.11) do not modify their fundamental character, mainly (5.8). Then, using delta function properties, we have after some manipulations

$$\begin{aligned}
&\lim_{|r_{j,n+1}|\to\infty} \Delta_{(n-1)}\hat{F}_{n+1}(\boldsymbol{\kappa}, r_{n+1}^{n}, p^{n+1}; 0) \\
&\quad = \lim_{|r_{j,n+1}|\to\infty} V(2\pi)^{-3} \prod_{\substack{l=1 \\ l\neq j}}^{n} \hat{F}_1(\boldsymbol{\kappa}/(n+1), \boldsymbol{p}_l; 0) \\
&\qquad \times \int \mathrm{d}\boldsymbol{k}_j\, \hat{F}_1(\boldsymbol{k}_j; \boldsymbol{p}_j; 0)\hat{F}_1(2\boldsymbol{\kappa}/(n+1) - \boldsymbol{k}_j, \boldsymbol{p}_j; 0) \\
&\qquad \times \exp\{\mathrm{i}\boldsymbol{r}_{j,n+1}\cdot[\boldsymbol{k}_j - \boldsymbol{\kappa}/(n+1)]\}.
\end{aligned} \tag{5.12}$$

Now we recall[9]) (see section 3) that one of the main effects of the operator $\boldsymbol{K}^{(j,n+1)}(t)$ in the limit (5.2) is the introduction of the limit $|\boldsymbol{r}_{j,n+1}|\to\infty$ into the functions to which it is applied. Realizing that the latter limit has no effect on $\hat{f}_n(\boldsymbol{k}, p^n; 0)$ and that we have $\Delta_{(n-1)}\hat{f}_n(0) = \hat{f}_n(0)$, it follows from (4.4) and (5.12) that

$$\lim_{|r_{j,n+1}|\to\infty} \Delta_{(n-1)}\hat{h}_{n+1}(\boldsymbol{\kappa}, 0) = \prod_{l=1}^{n+1} \hat{F}_1(\boldsymbol{\kappa}/(n+1); 0) - \hat{f}_n(\boldsymbol{\kappa}; 0) = 0, \tag{5.13}$$

where use has been made of the result (5.10).

Now we come back to eq. (5.6) and redefine the time scale through the transformation of variables

$$s = Dt, \quad \sigma = D\tau. \tag{5.14}$$

Using (5.14) in eq. (5.6) one has

$$\partial_s \hat{f}_n(\boldsymbol{\kappa}, p^n; s) = -\mathrm{i}\boldsymbol{P}'_n \cdot \boldsymbol{\kappa} \hat{f}_n(\boldsymbol{\kappa}, p^n; s) + \sum_{j=1}^{n} \Big\{ K^{(j,n+1)}(s/D) \times \exp[-\mathrm{i}\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1} s] \hat{f}_{n+1}(\boldsymbol{\kappa}, p^{n+1}; 0) - \int_0^s \mathrm{d}\sigma\, K^{(j,n+1)}(\sigma/D) \times \partial_\sigma \left(\exp[-\mathrm{i}\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1} \sigma] \hat{f}_{n+1}(\boldsymbol{\kappa}, p^{n+1}; s - \sigma) \right) \Big\} + K^{(j,n+1)}(s/D) \exp\{-\mathrm{i}\boldsymbol{\kappa} \cdot \boldsymbol{P}'_{n+1} s\} \Delta_{(n-1)} \hat{h}_{n+1}(\boldsymbol{\kappa}, r^n_{n+1}, p^{n+1}; 0), \tag{5.15}$$

where we have also performed a partial integration and used property (3.12). Introducing the limit $D \to 0$ in eq. (5.15) [or introducing the limit (5.2) in eqs. (5.6) or (2.20); both procedures are equivalent given (5.14)] we have

$$\partial_s \hat{f}_n(\boldsymbol{\kappa}, p^n; s) = -\mathrm{i}\boldsymbol{P}'_n \cdot \boldsymbol{\kappa} \hat{f}_n(\boldsymbol{\kappa}, p^n; s) + \sum_{j=1}^{n} K_{\mathrm{B}}^{(j,n+1)} \hat{f}_{n+1}(\boldsymbol{\kappa}, p^{n+1}; s), \tag{5.16}$$

after using the asymptotic properties (3.13) and (5.13). The inverse Fourier transform of this equation can be written as

$$\partial_s f_n(\boldsymbol{R}_n, p^n; s) = \sum_{j=1}^{n} [-m^{-1} \boldsymbol{p}_j \cdot \nabla_j f_n(\boldsymbol{R}_n, p^n; s) + K_{\mathrm{B}}^{(j,n+1)} f_{n+1}(\boldsymbol{R}_n, p^{n+1}; s)], \tag{5.17}$$

since $\tilde{\nabla}_n f_n(\boldsymbol{R}_n) = n \nabla_j f_n(\boldsymbol{R}_n)$ when $\boldsymbol{R}_n = n^{-1} \sum_{j=1}^{n} \boldsymbol{q}_j$.

Let us summarize here the conditions which have been introduced at different stages during the preceding derivation:

(i) The thermodynamic limit in the usual sense; it is necessary in order to obtain useful kinetic equations. We have implicitly assumed that the functions involved in our development present nice properties under this and the following conditions.

(ii) The system is spatially inhomogeneous, but the distances over which the system properties change significantly at $t = 0$ are large compared to the range of the interparticle potential (see the beginning of this section).

(iii) The interparticle potential is repulsive and has a finite range in order for the binary collision description to be meaningful. This weakens condition (ii).

(iv) The number density of the system is low enough in the sense of the limit (5.2); this limit also restricts the validity of the resulting equations to times much longer than the collision time, as one would desire in order to obtain irreversible equations[15]).

(v) The spatial correlations have a finite range in the initial state, at $t = 0$, for the infinite system [see eq. (5.7)].

Note that eq. (5.17) is an exact consequence of the Liouville equation under (i)–(v) provided there is no contribution of the term with D^2 in eq. (2.20) under these conditions; the latter point is analysed in section 8.

6. Kinetic equations for initially uniform systems

In spite of the fact that the case of uniform fluids is not very interesting from a strictly physical point of view, given that there is no change at all in the macroscopic variables nor fluid dynamics without spatial variation, it is instructive at this point to consider the consequences of the preceding formalism in that simple case. As already stated, the functions $\hat{f}_n(\boldsymbol{k}, p^n; t)$ for $\boldsymbol{k} = 0$ reduce to the familiar n-body momentum distribution functions $f_n(p^n; t)$ of interest in the theory of uniform fluids. Then eq. (5.16) reduces to the form

$$\partial_s f_n(s) = \sum_{j=1}^{n} K_B^{(j,n+1)} f_{n+1}(s). \tag{6.1}$$

We omit the details of the derivations in this section given that they are essentially similar to the ones in refs. 9–10. Assuming initial factorization of the n-body momentum distribution function, which is a direct consequence of initially finite-ranged correlations in the infinite system [see eq. (5.10)], one readily obtains the factorization at any time,

$$f_n(p^n; s) = \prod_{j=1}^{n} f_1(\boldsymbol{p}_j; s), \tag{6.2}$$

and the Boltzmann equation for homogeneous systems

$$\partial_s f_1(\boldsymbol{p}_j; s) = K_B^{(j,l)} f_1(\boldsymbol{p}_j; s) f_1(\boldsymbol{p}_l; s). \tag{6.3}$$

In order to complete this derivation, however, one has to prove that the term with a factor D^2 in eq. (2.20),

$$R(D, t) \equiv D^2 \int_0^t \mathrm{d}\tau \, J_{n,n+1,n+2}(\tau) h_{n+2}(q^{n+2}, p^{n+2}; t - \tau), \tag{6.4}$$

vanishes under the above conditions leading to (6.3). Once this is accomplished, one would have the interesting result that a low-density kinetic equation referring to a system in an initially uniform state of finite-ranged spatial correlations, is a direct consequence of the Liouville equation. Mazur

and Biel[9]) have shown that this is the case provided the function

$$\zeta(\boldsymbol{r}_{j,n+1}, p^{n+1}; t) \equiv \mathrm{i}\,\mathrm{T\text{-}lim}\, \Delta_{(n-1)} \mathscr{L}_{n+1,n+2} h_{n+2}(q^{n+2}, p^{n+2}; t), \tag{6.5}$$

$\boldsymbol{r}_{j,n+1} = \boldsymbol{q}_j - \boldsymbol{q}_{n+1}$, has a proper limit $\zeta_\infty(p^{n+1}; t)$ for $|\boldsymbol{r}_{j,n+1}| \to \infty$, and that this is equivalent to the (reasonable) assumption that a certain time average,

$$\lim_{t\to\infty} \frac{1}{t} \int_0^t \mathrm{d}\tau\, K^{(j,n+1)}(\tau) \Big[\zeta(\boldsymbol{r}_{j,n+1}; t-\tau) - \mathrm{T\text{-}lim}\, V^{-1} \int \mathrm{d}\boldsymbol{r}_{j,n+1} \zeta(\boldsymbol{r}_{j,n+1}; t-\tau) \Big], \tag{6.6}$$

vanishes. A different approach[11]) to the analysis of (6.4), which avoids the above restriction, is based on the direct estimation of the orders of magnitude involved in $R(D, t)$. In this way one readily shows that

$$R(D, t) = D^2 \int_0^t \mathrm{d}\tau\, N(t, \tau). \tag{6.7}$$

Assuming that $\nabla_j \varphi_{j,n} \cdot \partial_j h_n(q^n, p^n; t)$ is well-behaved, one obtains an upper bound A to the function $N(t, \tau)$. Then $R(D, t) \leq AD^2 t$ which vanishes in the limit (5.2).

We also note, although this paper will not deal with the corresponding problem in nonuniform systems, that the preceding formalism leads correctly[10]) to the Chon-Uhlenbeck triple collisions terms which contain the dominant effects beyond the Boltzmann contribution[14]).

7. The Boltzmann integrodifferential equation; persistence in time of the initial molecular chaos

In this section we are directly concerned with a controllable derivation of the conventional Boltzmann integrodifferential equation for nonuniform fluids from the Liouville equation. To this end we note that the set of equations (5.17) is to be solved subject to some initial condition on the reduced distribution functions; one easily convinces oneself that condition (5.7), which simply states finite-ranged correlations in the initial state, is not a sufficient condition to obtain the traditional Boltzmann equation from eq. (5.17). Let us assume initial molecular chaos in the sense

$$F_n(\boldsymbol{q}; p^n; 0) = \prod_{j=1}^{n} F_1(\boldsymbol{q}, \boldsymbol{p}_j; 0), \tag{7.1}$$

where we note that $F_n(\boldsymbol{q}) \equiv f_n(\boldsymbol{q})$. Condition (7.1) is analysed in section 8. The

important point here is that once condition (7.1) is verified at $s = 0$, it will be verified at all times s:

$$F_n(\boldsymbol{q}; p^n; s) = \prod_{j=1}^{n} F_1(\boldsymbol{q}, \boldsymbol{p}_j; s). \tag{7.2}$$

Indeed, the set of equations (5.17) has solutions of the form (7.2), where $F_1(\boldsymbol{q}, \boldsymbol{p}_j; s)$ satisfies the equation

$$\partial_s F_1(\boldsymbol{q}, \boldsymbol{p}_j; s) = -m^{-1}\boldsymbol{p}_j \cdot \nabla_j F_1(\boldsymbol{q}, \boldsymbol{p}_j; s) + K_B^{(j,l)} F_1(\boldsymbol{q}, \boldsymbol{p}_j; s) F_1(\boldsymbol{q}, \boldsymbol{p}_l; s), \tag{7.3}$$

given that eq. (5.17) becomes an identity for any n with (7.2) and (7.3). Thus, the eqs. (5.17) with the initial conditions (7.1) are equivalent to eqs. (7.2) and (7.3). Writing explicitly the symbol $K_B^{(j,l)}$ [see eq. (3.14)] we have

$$\begin{aligned} &\partial_s F_1(\boldsymbol{q}_1, \boldsymbol{p}_1; s) + m^{-1}\boldsymbol{p}_1 \cdot \nabla_1 F_1(\boldsymbol{q}_1, \boldsymbol{p}_1; s) \\ &\quad = \int \mathrm{d}\boldsymbol{p}_2 \iint \mathrm{d}\boldsymbol{x}\, \mathrm{d}\boldsymbol{y}\, 2m^{-1}|\boldsymbol{p}_1 - \boldsymbol{p}_2|[F_1(\boldsymbol{q}_1, \boldsymbol{p}_1^0; s) F_1(\boldsymbol{q}_1, \boldsymbol{p}_2^0; s) \\ &\qquad - F_1(\boldsymbol{q}_1, \boldsymbol{p}_1; s) F_1(\boldsymbol{q}_1, \boldsymbol{p}_2; s)], \end{aligned} \tag{7.4}$$

which corresponds to the conventional Boltzmann integrodifferential equation if one neglects the difference in position of the two colliding molecules.

8. Discussion

We complete our derivation of the Boltzmann equation from microscopic dynamics by analysing in this section: (a) the initial condition (7.1), and (b) the contribution of the term with D^2 in eq. (2.20) to the kinetic evolution of the system.

Concerning (b), it was argued in section 6 that the contribution which (for finite times) is of order D^2 vanishes in the low density limit for initially uniform systems[9]) where the distribution functions are invariant under translations. (Note that once this invariance is verified at $t = 0$, it will be retained by the system under a translational invariant Hamiltonian). Let us show that the proof outlined in section 6 can also be applied to the systems with initial weak inhomogeneities in which we are interested here.

The third term on the right-hand side of eq. (2.18) which we omitted in the subsequent discussion, can be essentially written as

$$\int_0^t \mathrm{d}\tau\, K_{n,n+1}(\tau) \Gamma_{n+1} \mathscr{L}_{n+1,n+2} h_{n+2}(t - \tau). \tag{8.1}$$

Using the results in section 3 and introducing the transformations in sections

4–7, in particular the wave vector scale (5.1) and the time scale (5.14), this is transformed into a sum from $j = 1$ to $j = n$ of terms

$$\int_0^s \mathrm{d}\sigma\, K^{(j,n+1)}(\sigma|D)\theta_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}_{j,n+1}, p^{n+1}; s-\sigma), \tag{8.2}$$

where

$$\theta_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}_{j,n+1}, p^{n+1}; s-\sigma) \equiv \text{T-lim}\, \exp\{-\mathrm{i}\boldsymbol{\kappa}\cdot\boldsymbol{P}'_{n+1}\sigma\}\Gamma^{(j,n+1)}\Delta_{(n-1)}\mathscr{L}_{n+1,n+2}\hat{h}_{n+2}(\boldsymbol{\kappa}, r_{n+2}^{n+1}, p^{n+2}; s-\sigma), \tag{8.3}$$

which would have appeared in eq. (5.15). Here we have used the previous notation and $\Gamma^{(j,n+1)} = (1 - V^{-2}\int\int \mathrm{d}\boldsymbol{q}_j\, \mathrm{d}\boldsymbol{q}_{n+1})$. Thus we also have, as in the case of initially uniform systems, terms of the form (8.2) with a function θ_{n+1} which only depends on $\boldsymbol{r} \equiv \boldsymbol{r}_{j,n-1}$ at all times under a translational invariant Hamiltonian (2.5). We first note[9]) that one can write

$$\lim_{D\to 0}\int_0^s \mathrm{d}\sigma\, K^{(j,n+1)}(\sigma|D)\theta_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}, p^{n+1}; s-\sigma) = s\lim_{t\to\infty}\frac{1}{t}\int_0^t \mathrm{d}\tau\, K^{(j,n+1)}(\tau)\theta_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}, p^{n+1}; t-\tau), \tag{8.4}$$

in the time scale (7.1), i.e. that a proof showing that terms (8.2) vanish in the limit $D\to 0$ (s finite) is equivalent to showing the annihilation of the time average (8.4). In order to state the problem in simpler terms, we now follow step by step the proof in ref. 9 to realize that (8.2) vanishes if

$$\lim_{|\boldsymbol{r}|\to\infty}\theta_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}, p^{n+1}; s-\sigma) = 0. \tag{8.5}$$

One can also realize that the function (8.3) can be written as the difference between a given function $\xi_{n+1}(\boldsymbol{r})$ and its space average,

$$\theta_{n+1}(\boldsymbol{r}) = \xi_{n+1}(\boldsymbol{r}) - \text{T-lim}\, V^{-1}\int \mathrm{d}\boldsymbol{r}\, \xi_{n+1}(\boldsymbol{r}), \tag{8.6}$$

so that (8.5) is equivalent to the existence of the limit

$$\lim_{|\boldsymbol{r}|\to\infty}\xi_{n+1}(\boldsymbol{\kappa}, \boldsymbol{r}, p^{n+1}; s-\sigma) = \xi_\infty(p^{n+1}; s-\sigma). \tag{8.7}$$

By comparison of expressions (8.6) and (8.3) one convinces oneself that, apart from irrelevant factors, the functions ξ_{n+1} in (8.7) are essentially a sum of distribution functions $h_{n+1}(\boldsymbol{\kappa}, r_{n+2}^{n+1}; s-\sigma)$ (see also section 5).

This justifies our assumption neglecting the term (8.1) in the time evolution equations. Accordingly, we have shown that the Boltzmann equation (7.3) is an exact consequence of the Liouville equation under conditions (i)–(v), enumerated in section 5, and initial molecular chaos in the sense of expression (7.1). Furthermore, we have shown that (7.1) is conserved at all times [see eq. (7.2)] during the kinetic evolution of the system when this is described in terms of the time scale (5.14).

It is interesting to compare this derivation of the traditional Boltzmann equation with the corresponding derivation in the case of initial spatially uniform states (section 6). The relevant initial hypothesis for homogeneous systems is (6.2) with $s = 0$; this is a consequence of (5.10) with $\boldsymbol{k} = 0$ which in turn follows from the hypothesis of initial finite-ranged spatial correlations. Thus, only conditions (i) and (iii)–(v) in section 5, and initial spatial uniformity are needed in that case. In the case of initially inhomogeneous systems one also would like to proceed from similar (mild) conditions but we have seen that, in addition to (i)–(v), one has to introduce the condition (7.1). This is not surprising at all because the intuitive original derivation of the Boltzmann equation involves certain uniformity assumptions[16,8]) much more restrictive than our initial hypothesis (ii) and (v); thus our initial condition (7.1) somehow contains those of the original Boltzmann assumptions which are really needed in a derivation from the Liouville equation. In order to understand the contents of (7.1) one may start with the more familiar formal expression of the initial molecular chaos assumption:

$$F_n(q^n, p^n; 0) = \prod_{j=1}^{n} F_1(\boldsymbol{q}_j, \boldsymbol{p}_j; 0), \tag{8.8}$$

which follows from (5.7) when $g_n(q^n, p^n; 0) = 0$, i.e. when all the relative coordinates of the subset of n particles are larger than the correlation length [which is supposed to have initially the same order of magnitude as the range of the interparticle forces; see eq. (5.8)]. Expression (7.1) follows from (8.8) when $|\boldsymbol{q}_j - \boldsymbol{q}|$ is smaller than λ, the characteristic length for the system inhomogeneities (see section 5).

Finally, we mention that from condition (7.1) and the above considerations it follows, given the definition (2.11), that $h_n(0) = 0$, which is consistent with property (5.13). (In the same way, it follows from our result (7.2) that $h_n(s) = 0$; this might seem to imply that the term (8.1) is identically zero but we note that (7.2) was obtained assuming that (8.1) vanishes in the low density limit).

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