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Equilibrium Time Correlation Functions in the Low-Density Limit

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We consider a system of hard spheres in thermal equilibrium. Using Lanford's result about the convergence of the solutions of the BBGKY hierarchy to the solutions of the Boltzmann hierarchy, we show that in the low-density limit (Boltzmann–Grad limit): (i) the total time correlation function is governed by the linearized Boltzmann equation (proved to be valid for short times), (ii) the self time correlation function, equivalently the distribution of a tagged particle in an equilibrium fluid, is governed by the Rayleigh–Boltzmann equation (proved to be valid for all times). In the latter case the fluid (not including the tagged particle) is to zeroth order in thermal equilibrium and to first order its distribution is governed by a combination of the Rayleigh–Boltzmann equation and the linearized Boltzmann equation (proved to be valid for short times).

KEY WORDS: Time correlation functions; low-density limit; linearized Boltzmann equation ; Boltzmann–Grad limit.

1. INTRODUCTION

In order to motivate the limits studied in this paper we consider first a fluid of hard spheres of diameter one and unit mass at low densities $\rho_\epsilon = \epsilon\rho$, $\epsilon \to 0$. In many cases of physical interest one expects in this regime typical spatial variations of the fluid to be of the order of a mean free path, $\sim 1/\epsilon$, and typical time variations to be of the order of a mean free time, $\sim 1/\epsilon$. Therefore, in order to study the dynamics of the fluid on its proper time and space scale, it is convenient to rescale time and space as

$$t' = \epsilon t, \qquad q' = \epsilon q \tag{1.1}$$

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Here t and q are the dynamical variables appearing in the equations of motion and t' and q' are the rescaled variables. The velocities and the mass of the particles remain unscaled.

In the rescaled t', q' variables typical time and space variations are of order one. On this scale a particle has diameter ϵ and the number of particles per unit volume increases as ϵ^{-2} in three dimensions. The $\epsilon \to 0$ limit is called the Boltzmann–Grad limit, since Grad[1] first wrote down and discussed this limit as the appropriate one for the exact validity of the Boltzmann equation. Subsequently, Lanford indeed proved[2,3] that, at least for short times, the nonlinear Boltzmann equation becomes exact in the Boltzmann–Grad limit for a rather general class of initial conditions on the n-particle correlation functions. The purpose of this paper is to study in the same limit equilibrium time correlation functions.

The self time correlation function can be regarded as describing the dynamics of a test particle in the fluid; e.g., imagine particle one painted red. Therefore this correlation function is governed, in the low-density limit, by the Rayleigh–Boltzmann equation, which is obtained from the nonlinear Boltzmann equation by replacing in the quadratic collision term the distribution function that is integrated over by the Maxwellian equilibrium distribution. The total time correlation function describes the time-dependent fluctuations of the fluid in thermal equilibrium. It is therefore governed, in the low-density limit, by the linearized Boltzmann equation which is obtained by linearizing the collision term at the Maxwellian.[4–6]

Our results are quite analogous to the fluctuation results obtained for the Vlasov equation by Braun and Hepp[7] (and for its quantum counterparts, the mean field models as studied by Hepp and Lieb[8]). They are only less complete in the sense that we can prove convergence only for short times and that, instead of proving a central limit theorem, we can show only convergence of the covariance.

2. THE LOW-DENSITY LIMIT. LANFORD'S THEOREM

We describe Lanford's result[2] about the convergence of the solutions of the BBGKY hierarchy to the solutions of the Boltzmann hierarchy. Since we will use an iteration argument later, we state the theorem as in King's thesis.[9]

We consider a system of hard spheres of diameter ϵ and unit mass inside a bounded region Λ with smooth boundary $\partial\Lambda$. The spheres (particles) are elastically reflected among themselves and at the boundary $\partial\Lambda$. Let the state of the system be specified by the absolutely continuous probabilities of finding exactly n particles at $dx_1 \cdots dx_n$

$$\left\{ f_n(x_1, \dots, x_n) \frac{1}{n!} dx_1 \cdots dx_n \middle| n \geqslant 0 \right\}$$

Here $x_i = (q_i, p_i) \in \Lambda \times R^3$ stands for the position of the center and the momentum of the ith particle. Then the distribution functions $\{\rho_n{}^\epsilon | n \geqslant 0\}$ corresponding to this state are defined by

$$\rho_n{}^\epsilon(x_1,\dots, x_n) = \sum_{m=0}^{\infty} \frac{1}{m!} \int_{(\Lambda \times R^3)^m} dy_1 \cdots dy_m f_{n+m}(x_1,\dots, x_n, y_1,\dots, y_m) \tag{2.1}$$

The time evolution of a state of the hard-sphere system is studied by means of the time evolution of the corresponding distribution functions. A straightforward computation, which is, however, nontrivial to justify rigorously,[10,11] leads to the following evolution equation:

$$\begin{aligned}\frac{\partial}{\partial t} \rho_n{}^\epsilon(x_1,\dots, x_n, t) &= H_n{}^\epsilon \rho_n{}^\epsilon(x_1,\dots, x_n, t) \\ &\quad + \epsilon^2 \sum_{j=1}^{n} \int_{R^3} dp_{n+1} \int_{S^2} d\omega\, \omega \cdot (p_{n+1} - p_j) \rho_{n+1}^\epsilon(x_1,\dots, x_n, q_j + \epsilon\omega, p_{n+1}, t)\end{aligned} \tag{2.2}$$

Here ω is a unit vector in R^3 and $d\omega$ is the surface measure of the unit sphere S^2 in three dimensions. $H_n{}^\epsilon$ describes the evolution of n hard spheres of diameter ϵ inside Λ. Equation (2.2) is the *BBGKY hierarchy* for hard spheres. The solutions of the BBGKY hierarchy are denoted by

$$\rho_n{}^\epsilon(x_1,\dots, x_n, t) = (V_t{}^\epsilon \rho^\epsilon)_n(x_1,\dots, x_n) \tag{2.3}$$

for the initial vector of distribution functions $\rho^\epsilon = (\rho_1{}^\epsilon, \rho_2{}^\epsilon,\dots)$.

Remark. The phase space of n hard spheres in Λ is

$$\begin{aligned}\mathscr{X}(n, \epsilon) = \{(q_1, p_1,\dots, q_n, p_n) \in (\Lambda \times R^3)^n \mid\, &|q_i - q_j| \geqslant \epsilon \text{ for } i \neq j, \\ &\operatorname{dist}(q_i, \partial\Lambda) \geqslant \epsilon/2\}\end{aligned}$$

In this space, boundary points of $\mathscr{X}(n, \epsilon)$ corresponding to a collision with the wall $\partial\Lambda$ and to a collision between two spheres are identified. E.g., if $q_j = q_i + \epsilon\omega$, $i \neq j$, and with incoming momenta p_i, p_j going over to p_i', p_j' in a collision, then $(q_1, p_1,\dots, q_i, p_i,\dots, q_i + \epsilon\omega, p_j,\dots, q_n, p_n)$ is identified with $(q_1, p_1,\dots, q_i, p_i',\dots, q_i + \epsilon\omega, p_j',\dots, q_n, p_n)$. There remains a set of "bad" points in $\partial\mathscr{X}(n, \epsilon)$ corresponding to triple and grazing collisions. In the interior of $\mathscr{X}(n, \epsilon)$ the time evolution is defined by free motion with infinitesimal generator $-\sum_{j=1}^n p_j\, \partial/\partial q_j$. This prescription extends smoothly through the points of $\partial\mathscr{X}(n, \epsilon)$ corresponding to pair collisions and to collisions with the wall $\partial\Lambda$. Points lying on trajectories leading to the bad points of $\mathscr{X}(n, \epsilon)$ form a set of Lebesgue measure zero. On this set the time evolution remains undefined. (Cf. the thesis of Alexander[12] for a detailed treatment of the time evolution of hard spheres.)

At this stage we can formally lift the restriction that Λ has to be a bounded region. So Λ may be, for example, a slab or the whole three-dimensional space. It is also clear that specular reflection at $\partial\Lambda$ is only one choice out of many possible boundary conditions: we could consider, for example, a stochastic boundary condition at $\partial\Lambda$ corresponding to a wall with a certain temperature. All these boundary conditions would be included in the definition of H_n^ϵ.

We want to study the low-density limit of the solutions of the BBGKY hierarchy. The low-density (Boltzmann–Grad) limit is obtained by letting the fraction of volume occupied by the particles $\sim \rho\epsilon^3$, with ρ the average density, go to zero while keeping the mean free path of the hard spheres, $\sim 1/\epsilon^2\rho$, constant. This requires that, as $\epsilon \to 0$, the density is increased as ϵ^{-2}. Therefore for each hard-sphere diameter ϵ one chooses an initial state with distribution functions ρ_n^ϵ such that $\rho_n^\epsilon \sim \epsilon^{-2n}$. With this in mind we define the *rescaled distribution functions*

$$r_n^\epsilon(x_1,..., x_n) = \epsilon^{2n}\rho_n^\epsilon(x_1,..., x_n) \tag{2.4}$$

Then (2.2) reads

$$\frac{d}{dt} r_n^\epsilon(t) = H_n^\epsilon r_n^\epsilon(t) + C_{n,n+1}^\varepsilon r_{n+1}^\varepsilon(t) \tag{2.5}$$

where the collision term in that equation is abbreviated as $C_{n,n+1}^\varepsilon$. Regarding the sequence $\{r_n^\epsilon | n \geq 0\}$ as the vector r^ϵ, one can write (2.5) compactly as

$$\frac{d}{dt} r^\epsilon(t) = H^\epsilon r^\epsilon(t) + C^\epsilon r^\epsilon(t) \tag{2.6}$$

where H^ϵ is a diagonal matrix with entries H_n^ϵ, and C^ϵ is a matrix with entries $C_{n,n+1}^\varepsilon$ and zero otherwise.

Let us now consider H^ϵ as the unperturbed part of the operator $H^\epsilon + C^\epsilon$ and C^ϵ as the perturbation. The time-dependent (Dyson) perturbation series for the solution of (2.6) then reads

$$r^\epsilon(t) = \sum_{m=0}^{\infty} \int_{0 \leq t_1 \leq \cdots \leq t_m \leq t} dt_m \cdots dt_1 \, S^\epsilon(t - t_m)C^\epsilon \cdots C^\epsilon S^\epsilon(t_1)r^\epsilon \tag{2.7}$$

where r^ϵ stands for $r^\epsilon(0)$, and where $(S^\epsilon(t)r^\epsilon)_n = ([\exp(H^\epsilon t)]r^\epsilon)_n = [\exp(H_n^\epsilon t)]r_n^\epsilon$ gives the evolution of n hard spheres of diameter ϵ inside Λ, always including the specular reflection at $\partial\Lambda$. Solutions of the BBGKY hierarchy are always understood in the sense of (2.7). Of course, one has to say in what sense (2.7) converges.

For $t \geqslant 0$ the time evolution of $r_n^\epsilon(t)$ is determined by backward streaming. Therefore it seems natural to replace, for a collision, the phase point $(x_1,..., q_j, p_j,..., q_j + \epsilon\omega, p_{n+1})$ with outgoing momenta by the phase point $(x_1,..., q_j, p_j',..., q_j + \epsilon\omega, p_{n+1}')$ with incoming momenta. (As explained before, these are just two different representations of the same phase point.) This leads to

$$\begin{aligned}
&\frac{\partial}{\partial t} r_n^\epsilon(x_1,..., x_n, t) \\
&\quad = H_n^\epsilon r_n^\epsilon(x_1,..., x_n, t) \\
&\qquad + \sum_{j=1}^{n} \int_+ dp_{n+1}\, d\omega\, \omega\cdot(p_j - p_{n+1}) \\
&\qquad \times \{r_{n+1}^\varepsilon(x_1,..., q_j, p_j',..., q_j - \epsilon\omega, p_{n+1}', t) \\
&\qquad - r_{n+1}^\varepsilon(x_1,..., q_j, p_j,..., q_j + \epsilon\omega, p_{n+1}, t)\}
\end{aligned} \tag{2.8}$$

where $\int_+$ indicates that the integration over ω is restricted to the upper hemisphere $\omega\cdot(p_j - p_{n+1}) \geqslant 0$. Formally, the limiting form of (2.8), which the limiting distribution functions $r(t) = \lim_{\epsilon\to 0} r^\epsilon(t)$ might satisfy, for $t \geqslant 0$, is

$$\begin{aligned}
&\frac{\partial}{\partial t} r_n(x_1,..., x_n, t) \\
&\quad = -\sum_{j=1}^{n} p_j \frac{\partial}{\partial q_j} r_n(x_1,..., x_n, t) \\
&\qquad + \sum_{j=1}^{n} \int_+ dp_{n+1}\, d\omega\, \omega\cdot(p_j - p_{n+1}) \\
&\qquad \times \{r_{n+1}(x_1,..., q_j, p_j',..., q_j, p_{n+1}', t) \\
&\qquad - r_{n+1}(x_1,..., q_j, p_j,..., q_j, p_{n+1}, t)\}
\end{aligned} \tag{2.9}$$

(Implicitly, the free motion $-\sum_{j=1}^{n} p_j\, \partial/\partial q_j$ includes the specular reflection at $\partial\Lambda$.)

For $t \leqslant 0$ the time evolution of $r_n^\epsilon(t)$ is determined by forward streaming. In that case, for a collision, the phase point $(x_1,..., q_j, p_j,..., q_j + \epsilon\omega, p_{n+1})$ with incoming momenta should be replaced by the phase point $(x_1,..., q_j, p_j',..., q_j + \epsilon\omega, p_{n+1}')$ with outgoing momenta. The formal limit of the resulting equation is then again (2.9) but with the sign of the collision term reversed.

Equation (2.9) for $t \geqslant 0$ and Eq. (2.9) with the sign of the collision term reversed for $t \leqslant 0$ is called the *Boltzmann hierarchy*, which can be written in the form

$$\frac{d}{dt} r_n(t) = H_n r_n(t) + C_{n,n+1} r_{n+1}(t)$$

or compactly

$$\frac{d}{dt} r(t) = Hr(t) + Cr(t) \tag{2.10}$$

Letting $(S(t)r)_n = (e^{Ht}r)_n = e^{H_n t}r_n$ denote the free motion of n particles inside Λ, the time-dependent perturbation series for the Boltzmann hierarchy reads

$$r(t) = \sum_{m=0}^{\infty} \int_{0 \leqslant t_1 \leqslant \cdots \leqslant t_m \leqslant t} dt_m \cdots dt_1 \, S(t - t_m)C \cdots CS(t_1)r \tag{2.11}$$

To prove that $r^\epsilon(t)$ defined by (2.7) indeed converges to $r(t)$ defined by (2.11) as $\epsilon \to 0$, we need two conditions.

First, the initial distributions r^ϵ have to be uniformly bounded in ϵ. This guarantees the uniform convergence of the perturbation series (2.7) for some interval $|t| \leqslant t_0$. If h_β denotes the normalized Maxwellian at inverse temperature β, then a suitable choice for this bound is as follows:

(C1) There exist a pair (z, β) such that

$$r_n^\epsilon(x_1,..., x_n) \leqslant Mz^n \prod_{j=1}^{n} h_\beta(p_j) \tag{2.12}$$

for all $\epsilon < \epsilon_0$ with a positive constant M independent of ϵ.

Second, r_n^ϵ has to converge to r_n in such a way that the series (2.7) converges term by term to the series (2.11). For the initial phase point $x^{(n)} = (x_1,..., x_n) \in (\Lambda \times R^3)^n$ let $q_j(t, x^{(n)})$, $j = 1,..., n$, be the position of the jth point particle at time t under the free motion. Then

$$\Gamma_n(t) = \{x^{(n)} = (x_1,..., x_n) \in (\Lambda \times R^3)^n | q_i(s, x^{(n)}) \neq q_j(s, x^{(n)}) \\ \text{for } i \neq j = 1,..., n \text{ and } -t \leqslant s \leqslant 0 \text{ if } t \geqslant 0, 0 \leqslant s \leqslant -t \text{ if } t \leqslant 0\}$$

In words, $\Gamma_n(t)$ is the restriction of the n-particle phase space to the set of phase points that under free backward streaming over a time t, if t is positive (or free forward streaming over a time $|t|$, if t is negative) do not lead to a collision between any pair of particles, regarded as point particles. By this restriction only a set of Lebesgue measure zero is excluded from $(\Lambda \times R^3)^n$.

Note that: (i) $\Gamma_n(t)$ depends only on the free motion, (ii) $\Gamma_n(t) \subset \Gamma_n(t')$ for $t' = \alpha t$, $\alpha \geqslant 1$, (iii) $\Gamma_n(t) \neq \Gamma_n(-t)$, and (iv) $x^{(n)} \in \Gamma_n(t)$ is equivalent to $\bar{x}^{(n)} \in \Gamma_n(-t)$, where $\bar{x}^{(n)}$ is the phase point obtained from $x^{(n)}$ under the reversal $p_j \mapsto -p_j$. In particular $\Gamma_n(t)$ is not invariant under reversal of velocities.

The suitable choice of convergence is then as follows:

(C2) There exists a continuous function r_n on $(\Lambda \times R^3)^n$ such that

$$\lim_{\epsilon \to 0} \epsilon^{2n} \rho_n^\epsilon = \lim_{\epsilon \to 0} r_n^\epsilon = r_n \tag{2.13}$$

uniformly on all compact sets of $\Gamma_n(s)$ for some $s \geqslant 0$.

Theorem (Lanford). Let $\{\rho_n^\epsilon | n \geqslant 0\}$ be a sequence of initial distribution functions of a fluid of hard spheres of diameter ϵ inside a region Λ and let the sequence $\{r_n^\epsilon | n \geqslant 0\}$ of rescaled distribution functions satisfy (C1) and (C2). Let $r_n^\epsilon(t)$ be the solution of the BBGKY hierarchy with initial conditions r_n^ϵ, and let $r_n(t)$ be the solution of the Boltzmann hierarchy with initial conditions r_n.

Then there exists a $t_0(z, \beta) > 0$ such that for $0 \leqslant t \leqslant t_0(z, \beta)$ the series (2.7) and (2.11) converge and such that $r_n^\epsilon(t)$ satisfies a bound of the form (C1) with $z' > z$ and $\beta' < \beta$. Furthermore,

$$\lim_{\epsilon \to 0} r_n^\epsilon(t) = r_n(t) \tag{2.14}$$

uniformly on compact sets of $\Gamma_n(s + t)$.

For $-t_0(z, \beta) \leqslant t \leqslant 0$, (2.14) holds provided that $s \leqslant 0$ and that in the Boltzmann hierarchy the collision term $C_{n,n+1}$ is replaced by $-C_{n,n+1}$.

Remark. It is the conditions for the validity of the limit (2.14) that make the irreversible nature of the Boltzmann hierarchy consistent with the reversibility of the BBGKY hierarchy (cf. Appendix A).

We now describe three interesting properties of the Boltzmann hierarchy. The first one is the well-known "propagation of chaos."

Property 1. If the initial conditions of the Boltzmann hierarchy factorize,

$$r_n(x_1, \ldots, x_n) = \prod_{j=1}^{n} f(x_j) \tag{2.15}$$

then the solutions with this initial condition stay factorized,

$$r_n(x_1, \ldots, x_n, t) = \prod_{j=1}^{n} f(x_j, t). \tag{2.16}$$

$f(x, t)$ is the solution of the *Boltzmann equation*

$$\frac{\partial}{\partial t} f(q, p, t) = -p \frac{\partial}{\partial q} f(q, p, t) + \int_+ dp_1\, d\omega\, \omega \cdot (p - p_1) \times \{f(q, p', t) f(q, p_1', t) - f(q, p, t) f(q, p_1, t)\} \tag{2.17}$$

with initial condition $f(q, p)$.

The second property comes from considering one of the fluid particles as a test particle, e.g., imagine particle one painted red.

Property 2. If the initial conditions of the Boltzmann hierarchy are of the form

$$r_n(x_1, \ldots, x_n) = f(x_1) \prod_{j=1}^{n} \{z h_\beta(p_j)\} \tag{2.18}$$

corresponding to an initial test particle distribution of the form $f(x_1)zh_\beta(x_1)$, then its solutions are

$$r_n(x_1,\ldots, x_n, t) = f(x_1, t)\prod_{j=1}^{n} \{zh_\beta(x_j)\} \tag{2.19}$$

$f(x, t)$ is the solution of the *Rayleigh–Boltzmann equation*

$$\begin{aligned}\frac{\partial}{\partial t}f(q, p, t) &= -p\frac{\partial}{\partial q}f(q, p, t) + z\int_{+} dp_1\, d\omega\, \omega\cdot(p - p_1)h_\beta(p_1)\\ &\quad\times \{f(q, p', t) - f(q, p, t)\} = (Af(t))(q, p)\end{aligned} \tag{2.20}$$

with initial condition $f(q, p)$. Equation (2.20) is also known as Lorentz–Boltzmann equation or linear Boltzmann equation.

Finally, we have the following property:

Property 3. If the initial conditions of the Boltzmann hierarchy are of the form

$$r_n(x_1,\ldots, x_n) = \left[\sum_{j=1}^{n} f(x_j)\right]\prod_{j=1}^{n} \{zh_\beta(x_j)\} \tag{2.21}$$

then its solutions are

$$r_n(x_1,\ldots, x_n, t) = \left[\sum_{j=1}^{n} f(x_j, t)\right]\prod_{j=1}^{n} \{zh_\beta(x_j)\} \tag{2.22}$$

$f(x, t)$ is the solution of the *linearized Boltzmann equation*

$$\begin{aligned}\frac{\partial}{\partial t}f(q, p, t) &= -p\frac{\partial}{\partial q}f(q, p, t) + z\int_{+} dp_1\, d\omega\, \omega\cdot(p - p_1)h_\beta(p_1)\\ &\times \{f(q, p_1', t) + f(q, p', t) - f(q, p_1, t) - f(q, p, t)\} = (Lf(t))(q, p)\end{aligned} \tag{2.23}$$

with initial condition $f(q, p)$.

Property 3 is proved by inserting the Ansatz (2.22) in the Boltzmann hierarchy and then by using repeatedly the fact that the collision operator acting on the Maxwellian h_β vanishes.

Properties 2 and 3 remain valid for $\prod_{j=1}^{n} \{zh(x_j)\}$ replaced by $\prod_{j=1}^{n} g(x_j)$, i.e., when the fluid is not in thermal equilibrium. In that case the analogs of A and L are time-dependent through the fluid distribution evolving according to the Boltzmann equation.

It should be understood that properties 1–3 are subject to the conditions of the theorem; in particular, the initial conditions have to satisfy the bound (C1) and the results are valid only up to $t_0(z, \beta)$. However, in contrast to the nonlinear Boltzmann equation, existence and uniqueness of the solutions of the Rayleigh–Boltzmann equation and the linearized Boltzmann equation in suitable spaces of functions have been proved for all times.[13] In particular, $\{e^{At}|t \geqslant 0\}$ and $\{e^{Lt}|t \geqslant 0\}$ are contraction semigroups on the Hilbert space $\mathscr{H} = L^2(\Lambda \times R^3, h_\beta(p)\, dq\, dp)$.[14]

3. EQUILIBRIUM TIME CORRELATION FUNCTIONS

We consider the fluid of hard spheres of diameter ϵ to be in thermal equilibrium with fugacity z_ϵ and inverse temperature β; grand canonical ensemble.

The Boltzmann–Grad limit corresponds to letting $\epsilon \to 0$ while *increasing* the fugacity as $z_\epsilon = \epsilon^{-2}z$. Since the equilibrium distribution functions have the form

$$\rho^{\varepsilon}_{eq,n}(x_1,\dots, x_n) = \prod_{j=1}^{n} \{z_\epsilon h_\beta(p_j)\} G_n(q_1,\dots, q_n, z_\epsilon \epsilon^3) \tag{3.1}$$

with $G_n \to 1$ as $z_\epsilon \epsilon^3 \to 0$ for all $q_1,\dots, q_n$ in which no two positions coincide, it is clear that as $\epsilon \to 0$ the system will resemble an ideal gas at infinite density. In particular, the rescaled distribution functions converge to

$$\lim_{\varepsilon\to 0} \epsilon^{2n}\rho^{\varepsilon}_{eq,n}(x_1,\dots, x_n) = \prod_{j=1}^{n} \{z h_\beta(p_j)\} \tag{3.2}$$

uniformly on compact sets of $\Gamma_n(0)$. [As mentioned in the introduction, this limit can be viewed alternatively by considering a spatial scale on which the diameter of a sphere equals one while $q_j' = \epsilon^{-1}q_j$. On this scale the fugacity decreases as ϵz and as $\epsilon \to 0$ the fluid reaches an ideal gas at zero density. To discuss time-dependent properties on this scale we would have to let $t' = \epsilon^{-1}t$.]

We now want to study the *self* and the *total* equilibrium time correlation functions in the low-density limit.

3.1. The Self Correlation Function

We consider a bounded region Λ, but will later drop this restriction. Let $f, g\colon \Lambda \times R^3 \to R$ be bounded and continuous functions of compact support. On $(\Lambda \times R^3)^n$ let us consider the functions $f_j(x_1,\dots, x_n) = f(x_j)$, $g_j(x_1,\dots, x_n) = g(x_j)$, $j \leqslant n$. Then the time-dependent self correlation $C(g, f; t)$ is defined as the grand canonical average of $\sum_{j=1}^{n} g(x_j) f_j(x_1,\dots, x_n, t)$,

$$C(g, f; t) = \left\langle \sum_{j=1}^{n} g_j f_j(t) \right\rangle \tag{3.3}$$

where $f_j(t)$ is f_j time-evolved under the dynamics of n hard spheres in Λ.

We transform (3.3) into a somewhat more manageable form. Let $e^{\epsilon}(q_1,\ldots, q_n)$ be the characteristic function, which is zero whenever $|q_i - q_j| \leqslant \epsilon$, $i \neq j$, or $d(q_i, \partial\Lambda) \leqslant \epsilon/2$, $i, j = 1,\ldots, n$, and which is one otherwise and let $S_n^{\epsilon}(t)$ denote, as before, the time evolution of n hard spheres of diameter ϵ inside Λ. Then, using the symmetry of the equilibrium distributions, we find

$$C(g,f;\, t) = \int dx_1\, f(x_1) \sum_{m=1}^{\infty} \frac{1}{(m-1)!} \int dx_2 \cdots dx_m \times (g_1 \circ S_m^{\epsilon})(-t)(x_1,\ldots, x_m) e_m^{\epsilon}(q_1,\ldots, q_m) \prod_{j=1}^{m} \{z_\epsilon h_\beta(p_j)\} Z^{-1} \tag{3.4}$$

where Z is the grand canonical partition function. Defining the signed initial distribution functions

$$(\rho_{s,g}^{\varepsilon})_n(x_1,\ldots, x_n) = g(x_1)\rho_{\mathrm{eq},n}^{\varepsilon}(x_1,\ldots, x_n) \tag{3.5}$$

one can rewrite (3.4) as

$$C(g,f;\, t) = \int dx_1\, f(x_1)(V_t^{\epsilon}\rho_{s,g}^{\varepsilon})_1(x_1) \tag{3.6}$$

where we have used the notation (2.3).

One may interpret the quantity $(V_t^{\epsilon}\rho_{s,g}^{\varepsilon})_1(x_1)$ as the test particle distribution function at time t resulting from an initial distribution $g(x_1)\rho_{\mathrm{eq},1}^{\varepsilon}(x_1)$ (cf. Section 4). Strictly speaking, this interpretation is allowed only if $g \geqslant 0$ and if $\int dx_1\, g(x_1)\rho_{\mathrm{eq},1}^{\varepsilon}(x_1) = 1$.

Remark. In (3.6), V_t^{ϵ} depends on the bounded region Λ. To obtain the result for an unbounded Λ, we choose a sequence of bounded regions Λ_m such that $\Lambda_m \to \Lambda$. The infinite-volume limit

$$\lim_{m\to\infty} V_t^{\epsilon}(\Lambda_m) = V_t^{\epsilon}(\Lambda)$$

can then be taken in the perturbation series (2.7) *before* taking the low-density limit $\epsilon \to 0$. The infinite-volume limit causes no difficulty, since all estimates in the proof of the theorem are uniform in Λ. With this prescription in mind, we drop the restriction of Λ being bounded.

Remark. For the computation of transport coefficients one has to consider such quantities as the velocity autocorrelation function $\langle p(t)\cdot p\rangle$ in the infinite-volume limit. In that case one has to show first the existence of

$$\lim_{\Lambda\to R^3} (1/|\Lambda|)C(g_\Lambda, f_\Lambda;\, t) = \langle p(t)\cdot p\rangle$$

with $f_\Lambda(q, p) = g_\Lambda(q, p) = \chi_\Lambda(q)p$, where χ_Λ is the characteristic function of the bounded region Λ. We have not studied this limit. The subsequent low-density limit follows by the argument used in the proof of Theorem 3.4. In the low-density limit $\langle p(t)\cdot p\rangle$ is governed by the spatially homogeneous Rayleigh–Boltzmann equation.

To obtain the low-density limit of (3.6), Lanford's theorem has to be applied to $V_t^\epsilon \rho_{s,g}^\varepsilon$. Therefore one has to check the conditions (C1) and (C2). Condition (C2) follows from (3.2) and (C1) from the following result:

Lemma 3.1. If $\sup_x |g(x)| < \infty$, then $\epsilon^{2n}(V_t^\epsilon \rho_{s,g}^\varepsilon)_n$ satisfies the bound (C1) for all t.

Proof. For a bounded region Λ we clearly have, by the invariance of $\rho_{\text{eq}}^\varepsilon$,

$$(V_t^\epsilon \rho_{s,g}^\varepsilon)_n(x_1,\ldots,x_n) \leqslant \sup_x |g(x)| \prod_{j=1}^n \{h_\beta(p_j)\}\bar{\rho}_{\text{eq},n}^\varepsilon(q_1,\ldots,q_n) \tag{3.7}$$

Here $\bar{\rho}_{\text{eq},n}^\varepsilon$ are the spatial parts of the equilibrium distribution functions at fugacity z_ϵ, for which it is known[15] that

$$\bar{\rho}_{\text{eq},n}^\varepsilon(q_1,\ldots,q_n) \leqslant (z_\epsilon)^n \tag{3.8}$$

independent of Λ. ■

The fact that $C(g,f;t) \sim \epsilon^{-2}$ can also be seen directly. Consider as a typical example the case where f and g are of the form $\chi_\Delta\phi$ with $\Delta \subset \Lambda$, and ϕ some function of the momentum. Then $C(g,f;t) \sim \langle N\rangle/|\Lambda|$, with $\langle N\rangle$ the average number of particles in Λ, because the probability of finding a given particle initially within Δ is proportional to $1/|\Lambda|$ and the average number of particles contributing to this correlation is $\langle N\rangle$. In the limit as $\epsilon \to 0$, $\langle N\rangle/|\Lambda| \sim z_\epsilon$; hence $C(g,f;t) \sim \epsilon^{-2}$.

Theorem 3.2. Let $f, g \in \mathscr{H} = L^2(\Lambda \times R^3, h_\beta(p)\,dq\,dp)$. Then, for $t \geqslant 0$

$$\lim_{\epsilon\to 0}(z_\epsilon)^{-1}\left\langle \sum_i g_i f_i(t)\right\rangle_{z_\epsilon,\beta} = \int dx\, h_\beta(p)f(x)(e^{At}g)(x) \tag{3.9}$$

Proof. Let f, g be continuous and of compact support. By Lanford's theorem, Property 2, (3.2), and Lemma 3.1,

$$\lim_{\epsilon\to 0} \epsilon^{2n}(V_t^\epsilon \rho_{s,g}^\varepsilon)_n(x_1,\ldots,x_n) = (e^{At}g)(x_1)\prod_{j=1}^n \{zh_\beta(p_j)\} \tag{3.10}$$

uniformly on compact sets of $\Gamma_n(t)$ for $0 \leqslant t \leqslant t_0(z,\beta)$. At $t = t_0 = t_0(z,\beta)$ the uniform bound (C1) is still valid by Lemma 3.1. Therefore, using (3.10), Lanford's theorem can be applied again to conclude that

$$\lim_{\epsilon\to 0} \epsilon^{2n}(V_t^\epsilon(V_{t_0}^\varepsilon \rho_{s,g}^\varepsilon))_n(x_1,\ldots,x_n) = (e^{A(t+t_0)}g)(x_1)\prod_{j=1}^n \{zh_\beta(p_j)\} \tag{3.11}$$

uniformly on compact sets of $\Gamma_n(t_0(z, \beta) + t)$ for $0 \leqslant t \leqslant t_0(z, \beta)$. Iterating, the result is valid for all times. In particular

$$\lim_{\epsilon \to 0} \epsilon^2 (V_t^{\epsilon} \rho_{s,g}^{\varepsilon})_1(x_1) = (e^{At} g)(x_1) z h_\beta(p_1) \tag{3.12}$$

uniformly on compact sets for all $t \geqslant 0$, which proves (3.9) for continuous f, g of compact support.

To extend (3.9) to all of $\mathscr{H}$ we use Schwarz's inequality and the invariance of the grand canonical equilibrium probability densities $\{f_{\mathrm{eq},n}^{\varepsilon} | n \geqslant 0\}$ to show

$$\begin{aligned}
&\left| \left\langle \sum_i g_i f_i(t) \right\rangle_{z_\epsilon, \beta} \right| \\
&\quad \leqslant \sum_{n=0}^{\infty} \sum_{i=1}^{n} \left| \int dx_1 \cdots dx_n f_{\mathrm{eq},n}^{\varepsilon}(x_1, \ldots, x_n) g(x_i) f_i(x_1, \ldots, x_n, t) \right| \\
&\quad \leqslant \sum_{n=0}^{\infty} n \left[\int dx_1 \cdots dx_n f_{\mathrm{eq},n}^{\varepsilon}(x_1, \ldots, x_n) g(x_1)^2 \right]^{1/2} \\
&\qquad \times \left[\int dx_1 \cdots dx_n f_{\mathrm{eq},n}^{\varepsilon}(x_1, \ldots, x_n) f(x_1)^2 \right]^{1/2} \\
&\quad \leqslant \left[\int dx_1\, \rho_{\mathrm{eq},1}^{\varepsilon}(x_1) g(x_1)^2 \right]^{1/2} \left[\int dx_1\, \rho_{\mathrm{eq},1}^{\epsilon}(x_1) f(x_1)^2 \right]^{1/2}
\end{aligned} \tag{3.13}$$

where we used (2.1) in the last step. Therefore $(z_\epsilon)^{-1} \langle \sum_i g_i f_i(t) \rangle_{z_\epsilon, \beta}$ is a bounded bilinear form on $\mathscr{H}$ and, since continuous functions of compact support are dense in $\mathscr{H}$, (3.9) extends by continuity. ■

3.2. The Total Correlation Function

We proceed with the total equilibrium time correlation functions. Let us define the sum $\sum g$ of one-particle functions g, which are assumed to be continuous and of compact support, as

$$\left(\sum g \right)(x_1, \ldots, x_n) = \sum_{j=1}^{n} g(x_j) \tag{3.14}$$

We define the total correlation functions of f and g as the grand canonical equilibrium average of

$$\left(\sum g \right)(x_1, \ldots, x_n) \left[\left(\sum f \right) \circ S_n^{\epsilon}(t) \right](x_1, \ldots, x_n) \tag{3.15}$$

In condensed form we write this average as

$$\left\langle \sum g \left(\sum f \right)(t) \right\rangle_{z_\epsilon, \beta} \tag{3.16}$$

It is not difficult to see that in the low-density limit

$$\lim_{\varepsilon\to 0}(z_\epsilon)^{-2}\left\langle \sum g\left(\sum f\right)(t)\right\rangle_{z_\epsilon,\beta} = \int dx_1\, h_\beta(p_1)g(x_1)\int dx_1\, h_\beta(p_1)f(x_1) \tag{3.17}$$

Therefore, a nontrivial result is only obtained upon subtracting out this limit, and the quantity to be considered is

$$\begin{aligned}(z_\epsilon)^{-1}&\left(\left\langle \sum g\left(\sum f\right)(t)\right\rangle_{z_\epsilon,\beta} - \left\langle \sum g\right\rangle_{z_\epsilon,\beta}\left\langle \sum f\right\rangle_{z_\epsilon,\beta}\right)\\ &= (z_\epsilon)^{-1}\left\langle (\delta g)(-t)\left(\sum f\right)\right\rangle_{z_\epsilon,\beta}; \qquad \delta g = \sum g - \left\langle \sum g\right\rangle_{z_\epsilon,\beta}\end{aligned} \tag{3.18}$$

where we have used the time invariance of the equilibrium measure.

We may think of (3.18) as giving the expectation value of $\sum f$ at time t when we start with a signed initial distribution obtained by multiplying the equilibrium density by δg. Equivalently, if at $t = 0$ the distribution functions $\rho_g{}^\epsilon$ are given by

$$\begin{aligned}\rho^\varepsilon_{g,n}(x_1,\dots,x_n) = &\left[\sum_{j=1}^n g(x_j)\right]\rho^\varepsilon_{\mathrm{eq},n}(x_1,\dots,x_n)\\ &+\int dx_{n+1}\, g(x_{n+1})\{\rho^\varepsilon_{\mathrm{eq},n+1}(x_1,\dots,x_{n+1})\\ &-\rho^\varepsilon_{\mathrm{eq},n}(x_1,\dots,x_n)\rho^\varepsilon_{\mathrm{eq},1}(x_{n+1})\}\end{aligned} \tag{3.19}$$

then

$$\left\langle (\delta g)(-t)\left(\sum f\right)\right\rangle_{z_\epsilon,\beta} = \int dx_1\, f(x_1)(V_t^\epsilon \rho_g{}^\epsilon)_1(x_1) \tag{3.20}$$

To apply Lanford's theorem, the conditions (C1) and (C2) have to be verified for $\rho_g{}^\epsilon$. By (3.2) and (3.8) the first term in (3.19) clearly causes no problem. The second term is estimated by

$$\begin{aligned}&\left|\int dx_{n+1}\, g(x_{n+1})\{\rho^\varepsilon_{\mathrm{eq},n+1}(x_1,\dots,x_{n+1}) - \rho^\varepsilon_{\mathrm{eq},n}(x_1,\dots,x_n)\rho^\varepsilon_{\mathrm{eq},1}(x_{n+1})\}\right|\\ &\times \leqslant \sup_x|g(x)|\prod_{j=1}^n\{h_\beta(p_j)\}\\ &\qquad\times\int_\Lambda dq\,|\bar\rho^\varepsilon_{\mathrm{eq},n+1}(q,q_1,\dots,q_n) - \bar\rho^\varepsilon_{\mathrm{eq},1}(q)\bar\rho^\varepsilon_{\mathrm{eq},n}(q_1,\dots,q_n)|\end{aligned} \tag{3.21}$$

Then the uniform bound (C1) follows from the next result:

Lemma 3.3. Let $z' > ez$. Then there exists a constant $c > 0$ and

$\epsilon(z') > 0$ such that

$$\sup_{q_1,\ldots,q_n \in \Lambda} \epsilon^{2n} \int dq \, |\bar{\rho}^{\varepsilon}_{\mathrm{eq},n+1}(q, q_1, \ldots, q_n) - \bar{\rho}^{\varepsilon}_{\mathrm{eq},1}(q)\bar{\rho}^{\varepsilon}_{\mathrm{eq},n}(q_1, \ldots, q_n)| \leqslant \epsilon c(z')^n \tag{3.22}$$

for all $\epsilon \leqslant \epsilon(z')$ independent of Λ.

Proof. Cf. Appendix B.

It is now easy to prove the following result:

Theorem 3.4. Let $f, g \in \mathscr{H}$. Then for $0 \leqslant t \leqslant t_0(ez, \beta)$

$$\lim_{\epsilon \to 0} (z_\epsilon)^{-1} \left[\left\langle \left(\sum g\right)\left(\sum f\right)(t) \right\rangle_{z_\epsilon,\beta} - \left\langle \sum g \right\rangle_{z_\epsilon,\beta} \left\langle \sum f \right\rangle_{z_\epsilon,\beta} \right] = \int dx \, h_\beta(p) f(x)(e^{Lt}g)(x) \tag{3.23}$$

Proof. Let f, g be continuous and of compact support. By Lemma 3.3, $\epsilon^{2n}\rho^{\varepsilon}_{g,n}$ satisfies the uniform bound (C1) for the pair (ez, β). By (3.2), (3.19), and Lemma 3.3

$$\lim_{\epsilon \to 0} \epsilon^{2n}\rho^{\varepsilon}_{g,n}(x_1, \ldots, x_n) = \left[\sum_{j=1}^{n} g(x_j)\right] \prod_{j=1}^{n} \{z h_\beta(p_j)\} \tag{3.24}$$

uniformly on compact sets of $\Gamma_n(0)$. Therefore by Lanford's theorem and by Property 3 [Eqs. (2.21) and (2.22)]

$$\lim_{\epsilon \to 0} \epsilon^{2n}(V_t^{\epsilon}\rho_g^{\epsilon})_n(x_1, \ldots, x_n) = \sum_{j=1}^{n} (e^{Lt}g)(x_j) \prod_{j=1}^{n} \{z h_\beta(x_j)\} \tag{3.25}$$

uniformly on compact sets of $\Gamma_n(t)$ for $0 \leqslant t \leqslant t_0(ez, \beta)$. Hence it follows from (3.20) that the left-hand side of (3.23) converges to $\int dx \, h_\beta(x) f(x)(e^{Lt})(x)$.

To extend (3.23) to all of $\mathscr{H}$, we use again Schwarz' inequality

$$\left| \left\langle \left(\sum g - \left\langle \sum g \right\rangle_{z_\epsilon,\beta}\right)\left[\left(\sum f\right)(t) - \left\langle \sum f \right\rangle_{z_\epsilon,\beta}\right] \right\rangle_{z_\epsilon,\beta} \right| \leqslant \left\langle \left(\sum g - \left\langle \sum g \right\rangle_{z_\epsilon,\beta}\right)^2 \right\rangle_{z_\epsilon,\beta}^{1/2} \left\langle \left(\sum f - \left\langle \sum f \right\rangle_{z_\epsilon,\beta}\right)^2 \right\rangle_{z_\epsilon,\beta}^{1/2} \tag{3.26}$$

Therefore for ϵ small enough

$$(z_\epsilon)^{-1} \left\langle \left(\sum g - \left\langle \sum g \right\rangle_{z_\epsilon,\beta}\right)\left[\left(\sum f\right)(t) - \left\langle \sum f \right\rangle_{z_\epsilon,\beta}\right] \right\rangle_{z_\epsilon,\beta}$$

is a bounded bilinear form on $\mathscr{H}$ and (3.23) follows by continuity.

Remark. Although $e^{Lt}g$ is known to exist for all $t \geq 0$, we have been unable to extend Theorem 3.4 beyond $t_0(ez, \beta)$.

Remark. The result of Theorem 3.4 can be viewed in a somewhat different way, which we feel to be rather instructive. Let us define the random variables

$$X_f^\epsilon = \sum f \tag{3.27}$$

on the phase space equipped with the equilibrium measure at fugacity z_ϵ and inverse temperature β. For the particular choice $f = \chi_\Delta$, X_f^ϵ is the number of particles in the region $\Delta \subset \Lambda \times R^3$. A straightforward equilibrium estimate shows that

$$\begin{aligned} \lim_{\epsilon\to 0}\langle \epsilon^2 X_f^\epsilon\rangle_{z_\epsilon,\beta} &= \int dx\, zh_\beta(p)f(x) \\ \lim_{\epsilon\to 0}\langle (\epsilon^2 X_f^\epsilon)^2\rangle_{z_\epsilon,\beta} - \langle \epsilon^2 X_f^\epsilon\rangle^2_{z_\epsilon,\beta} &= 0 \end{aligned} \tag{3.28}$$

for all $f \in \mathscr{H}$. This means that the distribution of $\epsilon^2 X_f$ converges to a δ-function as $\epsilon \to 0$. In particular, the relative number of particles in Δ has a sharp value in this limit.

Let us now consider the fluctuations of X_f around its average value, i.e., the *fluctuation observables*

$$\xi_f^\epsilon = \epsilon(X_f^\epsilon - \langle X_f^\epsilon\rangle_{z_\epsilon,\beta}) \tag{3.29}$$

and also their time evolution $\xi_f^\epsilon(t)$. One expects and can prove[16] that $\xi_f^\epsilon(t)$ has a Gaussian distribution as $\epsilon \to 0$ with mean zero and variance $z\int dx\, h_\beta(p)f(x)^2$. In other words, the central limit theorem holds for the sequence of random variables $\xi_f^\epsilon(t)$. But one also expects that $\{\xi_f^\epsilon(t) | t \in R, f \in \mathscr{H}\}$ become *jointly* Gaussian. Now Theorem 3.4 tells us that at least their covariance exists in the limit $\epsilon \to 0$ for short times, i.e.,

$$\lim_{\epsilon\to 0}\langle \xi_f^\epsilon(t)\xi_g^\epsilon(s)\rangle_{z_\epsilon,\beta} = \int dx\, zh_\beta(p)f(x)(e^{L(t-s)}g)(x) \tag{3.30}$$

for $t \geq s$, $t - s \leq t_0(ez, \beta)$. So we conjecture that $\{\xi_f^\epsilon(t) | t \in R, f \in \mathscr{H}\}$ converges as $\epsilon \to 0$ to a Gaussian stochastic process indexed by $\mathscr{H}$ with mean zero and covariance (3.30).

4. A TAGGED PARTICLE IN AN EQUILIBRIUM HARD-SPHERE FLUID

As is well known, the self time correlation function can equally well be interpreted as describing the distribution of a tagged particle in a fluid. Thinking of this tagged particle as an external probe of the fluid, it is then of interest to consider also the response of the fluid to the perturbation caused by the test particle. This in turn is related to the total time correlation function. But there are some new insights to be gained by looking at the problem from this point of view.

We consider a tagged particle in a fluid of hard spheres of diameter ϵ and mass one. The tagged particle is assumed to have the same properties. (We could allow the tagged particle to have a different mass and diameter. However, it is necessary that its diameter also decrease in proportion to ϵ.) The fluid plus tagged particle is enclosed in the region Λ. Initially, the fluid is assumed to be in thermal equilibrium at fugacity $z_\epsilon = \epsilon^{-2}z$ and inverse temperature β *conditioned* on the tagged particle being located at q_1 while the tagged particle has the distribution $f(x_1)\,dx_1$. Here $x_1 = (q_1, p_1)$ stands for the position and momentum of the tagged particle and $x_i = (q_i, p_i)$, $i \geqslant 2$, stands for the position and momentum of the $(i-1)$th fluid particle. Therefore the initial probability density of the joint system is proportional to

$$\left\{\frac{1}{n!} f(x_1)[\rho^\varepsilon_{\mathrm{eq},1}(x_1)]^{-1} e^\varepsilon_{n+1}(q_1,\dots, q_{n+1}) \prod_{j=1}^{n+1} \{z_\epsilon h_\beta(p_j)\} \middle| n \geqslant 0\right\} \tag{4.1}$$

with $f(x_1) \geqslant 0$ and $\int dx_1\, f(x_1) = 1$; $e^\varepsilon_n = 1$ if $|q_i - q_j| \geqslant \varepsilon$, zero otherwise.

We want to study the time evolution of the distribution functions of this system for $f(x_1) \leqslant ch_\beta(x_1)$. A straightforward computation shows that at $t = 0$ these distribution functions are

$$[\rho^\varepsilon_{\mathrm{eq},1}(x_1)]^{-1} f(x_1) \rho^\varepsilon_{\mathrm{eq},n+1}(x_1,\dots, x_{n+1}) = (\rho^\varepsilon_{s,g})_{n+1}(x_1,\dots, x_{n+1}) \tag{4.2}$$

with $g(x_1) = [\rho^\varepsilon_{\mathrm{eq},1}(x_1)]^{-1} f(x_1)$ in the notation (3.5). As before, $\rho^\varepsilon_{\mathrm{eq},n}$ are the unconditioned equilibrium distribution functions at fugacity z_ϵ and inverse temperature β. Since

$$\lim_{\epsilon\to 0} \epsilon^{-2}[\rho^\varepsilon_{\mathrm{eq},1}(x_1)]^{-1} = [z_\epsilon h_\beta(p_1)]^{-1} \tag{4.3}$$

we conclude from Property 2, (3.2), Lemma 3.1, and the iteration argument used in the proof of Theorem 3.2 that the following result holds:

Theorem 4.1. Let $(\rho^\varepsilon_{s,g})_{n+1}(x_1,\dots, x_{n+1}, t)$ denote the time-evolved distribution functions of the fluid plus tagged particle system with initial distribution functions given by (4.2). Then for all $t \geqslant 0$

$$\lim_{\epsilon\to 0} \epsilon^{2n}(\rho^\varepsilon_{s,g})_{n+1}(x_1,\dots, x_{n+1}, t) = (e^{At} f z^{-1} h_\beta^{-1})(x_1) \prod_{j=1}^{n+1} z h_\beta(p_j) \tag{4.4}$$

uniformly on compact sets of $\Gamma_{n+1}(t)$.

Integrating $(\rho^\varepsilon_{s,g})_{n+1}(x_1,\dots, x_{n+1}, t)$ over x_1 yields the fluid distribution functions $(\rho^\varepsilon_{fl,g})_n(x_2,\dots, x_{n+1}, t)$, which give the expectation of finding n fluid particles at $x_2, \ldots, x_{n+1}$. From Theorem 4.1, by integrating over x_1, one obtains that in the low-density limit

$$\lim_{\epsilon \to 0} \epsilon^{2n}(\rho_{fl,g}^{\varepsilon})_n(x_1, \ldots, x_n, t) = \prod_{j=1}^{n} \{zh_\beta(p_j)\} \tag{4.5}$$

In the limit the fluid is completely undisturbed by the presence of the tagged particle. This is of course to be expected, since in this limit the tagged particle will interact (directly or indirectly) during any fixed time interval only with a vanishing fraction of all particles in any fixed region. Consider now, however, the next order *correction*, i.e., the limiting behavior of

$$(\delta\rho_{fl,g}^{\varepsilon})_n(x_1, \ldots, x_n, t) = \epsilon^{2n-2}\{(\rho_{fl,g}^{\varepsilon})_n(x_1, \ldots, x_n, t) - \rho_{\mathrm{eq},n}^{\varepsilon}(x_1, \ldots, x_n)\} \tag{4.6}$$

Theorem 4.2. For $0 \leqslant t \leqslant t_0(ez, \beta)$

$$\lim_{\epsilon \to 0}(\delta\rho_{fl,g}^{\varepsilon})_n(x_1, \ldots, x_n, t) = \left\{\sum_{j=1}^{n} ([e^{Lt} - e^{At}]fz^{-1}h_\beta^{-1})(x_j)\right\} \prod_{j=1}^{n} \{zh_\beta(p_j)\} \tag{4.7}$$

uniformly on compact sets of $\Gamma_n(t)$.

Proof. With $g(x) = [\rho_{\mathrm{eq},1}^{\varepsilon}(x)]^{-1}f(x)$ and the notations (3.5), (3.19), and (4.6) one checks the identity

$$\rho_{g,n}^{\varepsilon}(x_1, \ldots, x_n) = \sum_{j=1}^{n} (\rho_{s,g}^{\varepsilon})_n(x_j, x_1, \ldots, x_{j-1}, \ldots, x_n) + (\delta\rho_{fl,g}^{\varepsilon})_n(x_1, \ldots, x_n) \tag{4.8}$$

The assertion now follows from Theorem 4.1 and the proof of Theorem 3.4. ■

APPENDIX A

We wish to illustrate here by means of an example how the Lanford theorem, Eq. (2.14), can manage to get the irreversible Boltzmann hierarchy from the reversible BBGKY hierarchy.

For the sake of clarity let us introduce some notation. We denote the velocity reversal operator by R,

$$(R\rho)_n(q_1, p_1, \ldots, q_n, p_n) = \rho_n(q_1, -p_1, \ldots, q_n, -p_n) \tag{A1}$$

As before, V_t^ϵ denotes the solution operator of the BBGKY hierarchy and V_t^0 denotes the solution operator of the Boltzmann hierarchy. [We remind the reader that for $t \leqslant 0$, $V_t^0 r$ is defined as the solution of (2.9) with the sign of the collision term reversed.]

Let us now consider a situation in which the box Λ is divided into two parts Λ_1 and Λ_2 and the initial state, at $t = 0$, corresponds to a canonical equilibrium state of N particles of diameter ϵ all in Λ_1. (We can imagine that there was an impenetrable barrier between Λ_1 and Λ_2 which was removed at $t = 0$.) It is clear that, since the initial state is invariant to reversal of velocities, its distribution functions $\rho^\epsilon = (\rho_1{}^\epsilon, \rho_2{}^\epsilon, \ldots)$ satisfy the equality

$$V_t{}^\epsilon \rho^\epsilon = R V_{-t}^\varepsilon \rho^\epsilon \tag{A2}$$

Furthermore,

$$V_t{}^\epsilon (R V_t{}^\epsilon \rho^\epsilon) = \rho^\epsilon \tag{A3}$$

while

$$V_t{}^\epsilon (V_t{}^\epsilon \rho^\epsilon) = V_{2t}^\varepsilon \rho^\epsilon \tag{A4}$$

This means that if at time t we reverse all velocities, then the system, after another time interval t, will return to its initial state in which all the particles are in Λ_1.

Consider now the sequence of initial states with distribution functions ρ^ϵ in which as $\epsilon \to 0$ the number of particles inside Λ_1 increases with fixed $N\epsilon^2 = z$. Then

$$\begin{aligned} \lim_{\epsilon \to 0} \epsilon^{2n} \rho_n{}^\epsilon (x_1, \ldots, x_n) &= \lim_{\epsilon \to 0} r_n{}^\epsilon (x_1, \ldots, x_n) = r_n(x_1, \ldots, x_n) \\ &= \prod_{j=1}^{n} \{\chi_{\Lambda_1}(q_j) z h_\beta(p_j)\} \end{aligned} \tag{A5}$$

on $\Gamma_n(0)$, where χ_{Λ_1} is the characteristic function of the set Λ_1, and, since (C1) and (C2) are satisfied, by Lanford's theorem

$$\lim_{\epsilon \to 0} \epsilon^{2n} (V_t{}^\epsilon \rho^\epsilon)_n (x_1, \ldots, x_n) = (V_t{}^0 r)_n (x_1, \ldots, x_n) = \prod_{j=1}^{n} \{f(x_j, t)\} \tag{A6}$$

on $\Gamma_n(t)$ for $|t| < t_0(z, \beta)$, where $f(x, t)$ is the solution of the Boltzmann equation with initial conditions $f(q, p) = \chi_{\Lambda_1}(q) z h_\beta(p)$.

Let us now reverse the velocities at time t, $0 \leqslant |t| \leqslant t_0/2$, and let us consider $R V_t{}^\epsilon \rho^\epsilon$ as the new initial state. Clearly

$$V_t{}^0 (R V_t{}^0 r) \neq r \tag{A7}$$

in contrast to (A3), so the limiting r do not have the time reversibility of the r^ϵ. Indeed, the Boltzmann H-function decreases up to t, remains unchanged by R, and continues to decrease as $R V_t{}^0 r$ is evolved for a time interval t.

At first sight, Lanford's theorem seems to assert that

$$\lim_{\varepsilon \to 0} \epsilon^{2n} (V_t{}^\epsilon (R V_t{}^\epsilon \rho^\epsilon))_n = (V_t{}^0 (R V_t{}^0 r))_n \neq r_n$$

However, there is no such contradiction. The answer lies in the fact that while

$$\lim_{\varepsilon \to 0} \epsilon^{2n} (V_t{}^\epsilon \rho^\epsilon)_n = (V_t{}^0 r)_n \quad \text{on } \Gamma_n(t) \tag{A8}$$

it is also true that

$$\lim_{\epsilon\to 0} \epsilon^{2n}(RV_t^{\epsilon}\rho^{\epsilon})_n = (RV_t^0 r)_n = (V_t^0 r)_n \quad \text{on } \Gamma_n(-t) \neq \Gamma_n(t)$$

Therefore, continuing in the same time direction as before the reversal of velocities, $RV_t^{\epsilon}\rho^{\epsilon}$ no longer satisfies the second condition (C2) of Lanford's theorem. The theorem asserts nothing about the convergence of $\epsilon^{2n}(V_t^{\epsilon}(RV_t^{\epsilon}\rho^{\epsilon}))_n$ as $\epsilon \to 0$. [Of course, by (A3), we can say something about this limit. The point is that we cannot conclude from Lanford's theorem that the limit is $(V_t^0(RV_t^0 r))_n$, since condition (C2) is violated.] For the theorem still to be applicable at time t one has only the two choices to consider, either $V_t^{\epsilon}(V_t^{\epsilon}\rho^{\epsilon})$ or $V_{-t}^{\varepsilon}(RV_t^{\epsilon}\rho^{\epsilon})$. In both cases the system evolves further toward equilibrium.

The irreversible Boltzmann hierarchy is consistent with the reversible BBGKY hierarchy, since the approximation by the Boltzmann hierarchy is valid only for a *particular class* of initial states. The condition (C2) excludes highly correlated initial states such as the one just constructed by reversal of velocities.

APPENDIX B. PROOF OF LEMMA 3.3

Relation (3.22) is transformed to a spatial scale on which a sphere has diameter one. Then

$$\begin{aligned} &\sup_{q_1,\ldots,q_n\in\Lambda} \epsilon^{2n}\int_{\Lambda} dq\, |\bar{\rho}^{\epsilon}_{\mathrm{eq},n+1}(q, q_1,\ldots, q_n) - \bar{\rho}^{\epsilon}_{\mathrm{eq},1}(q)\bar{\rho}^{\epsilon}_{\mathrm{eq},n}(q_1,\ldots, q_n)| \\ &\quad = \sup_{q_1,\ldots,q_n\in\varepsilon^{-1}\Lambda} \epsilon\int_{\epsilon^{-1}\Lambda} dq\, \epsilon^{-(n+1)} \\ &\quad \times |\rho^{\epsilon z}_{n+1}(q, q_1,\ldots, q_n; \epsilon^{-1}\Lambda) - \rho^{\epsilon z}_1(q; \epsilon^{-1}\Lambda)\rho^{\epsilon z}_n(q_1,\ldots, q_n; \epsilon^{-1}\Lambda)| \end{aligned} \tag{B1}$$

Here $\rho_n^{\epsilon z}(q_1,\ldots, q_n; \epsilon^{-1}\Lambda)$ represents the spatial part of the grand canonical equilibrium distribution functions of hard spheres of diameter one inside the region $\epsilon^{-1}\Lambda$ at fugacity $\epsilon^3 z_\epsilon = \epsilon z$. Lemma 3.3 follows now from the result:

Lemma B1. There exists an $\epsilon_0 > 0$ such that for all $\epsilon < \epsilon_0$

$$\begin{aligned} &\sup_{q_1,\ldots,q_n\in\varepsilon^{-1}\Lambda} \epsilon^{-(n+1)}\int_{\epsilon^{-1}\Lambda} dq\, |\rho^{\epsilon z}_{n+1}(q, q_1,\ldots, q_n; \epsilon^{-1}\Lambda) \\ &\quad - \rho^{\epsilon z}_1(q; \epsilon^{-1}\Lambda)\rho^{\epsilon z}_n(q_1,\ldots, q_n; \epsilon^{-1}\Lambda)| \leqslant M(z')^n \end{aligned} \tag{B2}$$

where M is a constant and $z' > ez$.

Proof. We consider the particles at $q_1,\ldots, q_n$ as providing an external field and denote the equilibrium distribution functions of this system by

$\rho^z(\cdot\,|q_1,\dots,q_n;\Lambda)$. Then, expanding in z,

$$\rho_1{}^z(q|q_1,\dots,q_n;\Lambda) - \rho_1{}^z(q;\Lambda) = \sum_{j=0}^{\infty} c_j(q|q_1,\dots,q_n)z^{j+1} \tag{B3}$$

In terms of the zero-field Ursell functions U_{j+1} the expansion coefficients are

$$c_j(q|q_1,\dots,q_n) = \frac{1}{j!}\int dq_1{}' \cdots dq_j{}'\; U_{j+1}(q,q_1{}',\dots,q_j{}')\left\{\prod_{k=1}^{n}\left[h(q-q_k)\right.\right.$$
$$\left.\left.\times\prod_{i=1}^{j} h(q_i{}'-q_k)\right] - 1\right\} \tag{B4}$$

where, we let h be the overlap function, $h(q) = 0$ for $|q| \leqslant 1$ and $h(q) = 1$ otherwise. The second factor is negative and, according to Ref. 15, Chapter 4, (5.14), for a positive pair potential $(-1)^{j+1}U_j \geqslant 0$. Therefore

$$(-1)^{j+1}c_j(q|q_1,\dots,q_n) \geqslant 0 \tag{B5}$$

and for $z > 0$

$$\begin{aligned}
|\rho_1{}^z(q|q_1,\dots,q_n;\Lambda) - \rho_1{}^z(q;\Lambda)| &\leqslant \sum_{j=0}^{\infty} |c_j(q|q_1,\dots,q_n)|z^{j+1} \\
&= \sum_{j=0}^{\infty} c_j(q|q_1,\dots,q_n)(-1)^{j+1}z^{j+1} \\
&= \rho_1^{-z}(q|q_1,\dots,q_n;\Lambda) - \rho_1^{-z}(q;\Lambda)
\end{aligned} \tag{B6}$$

For small enough ϵ, $\rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda) \neq 0$ and therefore we have for $z > 0$

$$\begin{aligned}
&\int_{\epsilon^{-1}\Lambda} dq\; |\rho_{n+1}^{\epsilon z}(q,q_1,\dots,q_n;\epsilon^{-1}\Lambda) - \rho_1^{\epsilon z}(q;\epsilon^{-1}\Lambda)\rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)| \\
&\quad\leqslant \rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)\int_{\epsilon^{-1}\Lambda} dq\; |\rho_1^{\epsilon z}(q|q_1,\dots,q_n;\epsilon^{-1}\Lambda) - \rho_1^{\epsilon z}(q;\epsilon^{-1}\Lambda)| \\
&\quad\leqslant \rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)\int_{\epsilon^{-1}\Lambda} dq\; [\rho_1^{-\epsilon z}(q|q_1,\dots,q_n;\epsilon^{-1}\Lambda) - \rho_1^{-\epsilon z}(q;\epsilon^{-1}\Lambda)] \\
&\quad= \rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)[\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)]^{-1} \\
&\qquad\times\int_{\epsilon^{-1}\Lambda} dq\{\rho_{n+1}^{-\epsilon z}(q,q_1,\dots,q_n;\epsilon^{-1}\Lambda) - \rho_1^{-\epsilon z}(q;\epsilon^{-1}\Lambda)\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)\} \\
&\quad= \{\rho_n^{\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)[\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)]^{-1}\} \\
&\qquad\times\left\{-n\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda) + z\frac{d}{dz}\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)\right\}
\end{aligned} \tag{B7}$$

(B7) is estimated using the Mayer expansion. For small enough ϵ the first factor is uniformly bounded. The second factor is bounded by

$$\left| -n\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda) + z\frac{d}{dz}\rho_n^{-\epsilon z}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)\right|$$

$$\leqslant \sum_{m=0}^{\infty} |b_{n,m}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)|\ |-n+n+m|\ |\epsilon z|^{n+m}$$

$$\leqslant \sum_{m=1}^{\infty} n(n+m)^{m-1}m\left(\frac{4\pi}{3}\right)^m \frac{1}{m!}|\epsilon z|^{n+m} < M\frac{1}{1-4\pi|\epsilon z|/3}ne^n|\epsilon z|^{n+1} \tag{B8}$$

where we have used the uniform bound on the coefficients $b_{n,m}(q_1,\dots,q_n;\epsilon^{-1}\Lambda)$ [cf. Ref. 15, Chapter 4, (4.30)]. Relation (B8) together with (B7) proves the lemma. ∎

Remark. Lemma B1 holds for any positive pair potential V with $\int dq\,(1-e^{-\beta V(q)}) < \infty$.

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