Journal of Statistical Physics, Vol. 61, Nos. 1/2, 1990

Linear Quantum Enskog Equation. II. Inhomogeneous Quantum Fluids

D. Loss[1,2]

Received December 18, 1989

This is the second part of a work concerned with the quantum-statistical generalization of classical Enskog theory, whereby the first part is extended to spatially inhomogeneous fluids. In particular, working with Liouville operators and using cluster expansions and projection operators, we derive the inhomogeneous linear quantum Enskog equation and express the dynamic structure factor and the nonlocal mobility tensor in terms of the corresponding quantum Enskog collision operator. Thereby static correlations due to excluded volume effects and quantum-statistical correlations due to the fermionic (bosonic) character of the pairwise strongly interacting particles are treated exactly. When static correlations are neglected, this Enskog equation reduces to the inhomogeneous linear quantum Boltzmann equation (containing an exchange-modified t-matrix). In the classical limit, the well-known linear revised Enskog theory is recovered for hard spheres.

KEY WORDS: Quantum kinetic theory, linear; dynamic structure factor; nonlocal mobility; Liouville operators; cluster expansions; static, dynamic, and quantum-statistical correlations; inhomogeneous quantum Enskog equation, linear.

1. INTRODUCTION

In a preceding paper[1] (hereafter referred to as I) we derived the quantum-statistical generalization of the linear revised Enskog equation by making use of a recently developed superoperator formalism[2-4] (See I for a more general introduction). This generalized kinetic equation is an extension of the linear quantum Boltzmann equation and is applicable to dense normal

[1] Institut für Theoretische Physik der Universität Zürich, CH-8001 Zürich, Switzerland.
[2] Present address: Department of Physics, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801.

0022-4715/90/1000-0467$06.00/0

quantum fluids consisting of fermions (bosons) which interact via an arbitrary short-range central pair potential. Thereby, as an extension of the usual Enskog theory[5-12] of classical hard-sphere systems, effects of finite interaction range are taken into account.

However, the formalism in I (as well as in refs. 2–4) was restricted to spatially homogeneous systems. The objective of the present paper, therefore, is to extend this formalism to spatially inhomogeneous quantum fluids.

The central quantity of interest here is a *space-dependent* one-particle equilibrium time correlation function from which physically important functions, such as the dynamic structure factor and the external nonlocal mobility tensor, are obtained. The microscopic evaluation of this quantum time correlation function is performed in the Liouville space formalism and makes use of cluster expansions and projection operators. The main advantage of this approach over other formalisms, such as Green's function techniques, is that *dynamic* and *static* correlations are always clearly separated. This is crucial in order to perform an Enskog-type approximation because in this approximation the dynamic and static parts are treated differently.

In analogy to the classical hard-sphere case, it is reasonable to expect this different treatment of statics and dynamics to be consistent (which, however, we do not demonstrate here) and to lead to good agreement with experimental data. We shall elaborate on this point in future work by applying the results obtained here to a specific example.

The derivation presented here runs largely parallel to that given before for homogeneous fluids.[1-4] Therefore, besides some unavoidable repetitions, we shall discuss only those points of the derivation in detail which are new and due to the spatial inhomogeneity.

The paper is organized as follows. In Section 2, the problem to be considered is defined and some important relations between our time correlation function, the dynamic structure factor, and the external nonlocal mobility tensor are given. In Section 3, we derive a Dyson-equation-like formula (containing a generalized collision operator) for the time correlation function. Here we introduce projection operators and cluster expansion techniques and prove a factorization theorem which allows us to obtain a closed equation. The Enskog approximation is explained and performed in Section 4, leading to the inhomogeneous linear quantum Enskog equation. There we also express the dynamic structure factor and the mobility tensor in terms of the linear quantum Enskog collision operator $B_1^{QE}(z)$. In Section 5, we show that $B_1^{QE}(z)$ reduces to the linear quantum Boltzmann collision operator (with exchange-modified scattering cross section) when static correlations are neglected. In Section 6, we consider the classical limit and show that $B_1^{\mathrm{QE}}(z)$ reduces to the classical linear revised Enskog collision operator for hard spheres as given by van Beijeren and Ernst.[10,11] This demonstration is based on functional derivative techniques. Some conclusions are gathered in Section 7. Most of the technical details are deferred to the Appendices A–D.

2. CORRELATION FUNCTIONS

We consider a normal quantum fluid of N identical pairwise interacting fermions (bosons) of mass m enclosed in a periodic box of volume Ω at temperature $\beta^{-1} = k_{\mathrm{B}} T$ (k_{B} is Boltzmann's constant). The Hamiltonian H of this system is given as

$$H = H_0 + V = \sum_{i=1}^{N} H_0(i) + \sum_{1 \leqslant i < j \leqslant N} V_{ij} \tag{2.1}$$

with

$$H_0(i) = \frac{\hat{\mathbf{p}}_i^2}{2m} \tag{2.2}$$

and

$$V_{ij} = V(|\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|) \tag{2.3}$$

$\mathbf{p}_i$ and $\mathbf{r}_i$ are the momentum and position operators, respectively, of the ith particle. V is a short-range pair-interaction potential that depends only on the relative coordinates of the particles i and j.

The main focus of this work will be an evaluation of equilibrium one-particle time correlation functions of microscopic densities in this quantum fluid. Such correlation functions can be expressed in terms of a basic space- and time-dependent correlation function $C_{ab}(\mathbf{r}, \mathbf{r}'; t)$ given by

$$C_{ab}(\mathbf{r}, \mathbf{r}'; t) = \langle a(\mathbf{r}; t)\, b(\mathbf{r}') \rangle = \operatorname{Tr} \rho\, a(\mathbf{r}; t)\, b(\mathbf{r}') \tag{2.4}$$

where ρ is the canonical density matrix,

$$\rho = Z^{-1} e^{-\beta H}, \qquad Z = \operatorname{Tr} e^{-\beta H} \tag{2.5}$$

The microscopic densities $a(\mathbf{r})$ and $b(\mathbf{r}')$ are given by sums of one-particle operators of the form

$$a(\mathbf{r}) = \sum_{i=1}^{N} a_i(\mathbf{r}), \qquad b(\mathbf{r}') = \sum_{i=1}^{N} b_i(\mathbf{r}') \tag{2.6}$$

with

$$a_i(\mathbf{r}) = \tfrac{1}{2}\{\bar{a}(\hat{\mathbf{p}}_i), \delta(\mathbf{r} - \hat{\mathbf{r}}_i)\}$$
$$b_i(\mathbf{r}') = \tfrac{1}{2}\{\bar{b}(\hat{\mathbf{p}}_i), \delta(\mathbf{r}' - \hat{\mathbf{r}}_i)\} \tag{2.7}$$

where the braces denote anticommutators. $\bar{a}(\hat{\mathbf{p}}_i)$ and $\bar{b}(\hat{\mathbf{p}}_i)$ are arbitrary functions of the momentum operator $\hat{\mathbf{p}}_i$ only (examples are given below).

The time dependence of $a(\mathbf{r}; t)$ is given in the Heisenberg picture and reads explicitly (we set $\hbar = 1$)

$$a(\mathbf{r}; t) = e^{iHt} a(\mathbf{r})\, e^{-iHt} = e^{iLt} a(\mathbf{r}) \tag{2.8}$$

Here we have introduced the Liouville operator $L = L(1 \cdots N)$ defined by $La = [H, a]$, with

$$L = L_0 + L_V = \sum_{i=1}^{N} L_0(i) + \sum_{1 \leqslant i < j \leqslant N} L_{ij} \tag{2.9}$$

where

$$L_0(i)\, a = [H_0(i), a] \tag{2.10}$$

$$L_{ij} a = [V_{ij}, a] \tag{2.11}$$

The Liouville operator belongs to the class of superoperators[14,15] which are formally defined as linear operators acting on ordinary Hilbert-space operators. Further superoperators will be introduced below.

Due to the translational invariance of H, we can replace the computation of $C_{ab}(\mathbf{r}, \mathbf{r}'; t)$ by an equivalent computation of its Fourier and Laplace transform $C_{ab}(\mathbf{q}; z)$ defined by

$$C_{ab}(\mathbf{q}; z) = \int_0^\infty dt\, e^{-zt} \int d(\mathbf{r} - \mathbf{r}')\{\exp[-i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')]\}\, C_{ab}(\mathbf{r} - \mathbf{r}', 0; t)$$
$$\operatorname{Re} z > 0 \tag{2.12}$$

Explicitly, we have

$$C_{ab}(\mathbf{q}; z) = \frac{1}{\Omega} \int_0^\infty dt\, e^{-zt} \langle a(\mathbf{q}; t)\, b(-\mathbf{q}) \rangle \tag{2.13}$$

with

$$a(\mathbf{q}) = \sum_{i=1}^{N} a_i(\mathbf{q})$$
$$a_i(\mathbf{q}) = \tfrac{1}{2}\{\bar{a}(\hat{\mathbf{p}}_i), \exp(-i\mathbf{q} \cdot \hat{\mathbf{r}}_i)\} \tag{2.14}$$

and analogously for $b(-\mathbf{q})$. The Fourier wave vector $\mathbf{q}$ is a measure of the spatial imhomogeneity of our quantum fluid. In the following we consider only $\mathbf{q} \neq 0$, with the consequence that in this case $\langle a(\mathbf{q}; t)\rangle = \langle b(-\mathbf{q})\rangle = 0$. The homogeneous case $\mathbf{q} = 0$ was treated in *I*.

As a first important example, let us consider the dynamic structure factor $S(\mathbf{q}; \omega)$. This quantity is of interest because it can be measured by means of neutron scattering experiments (see, e.g., refs. 12 and 14; for the interpretation of neutron scattering data on classical liquids with the help of Enskog's theory, see ref. 16 and the references given therein).

Usually $S(\mathbf{q}; \omega)$ is defined as

$$S(\mathbf{q}; \omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} dt\, e^{i\omega t} \langle n(\mathbf{q}; t)\, n(-\mathbf{q})\rangle \tag{2.15}$$

where

$$n(\mathbf{q}) = \sum_{i=1}^{N} \exp(-i\mathbf{q} \cdot \hat{\mathbf{r}}_i) \tag{2.16}$$

The dynamic structure factor can now be expressed in terms of our basic correlation function $C_{ab}(\mathbf{q}; z)$:

$$S(\mathbf{q}; \omega) = \frac{\Omega}{\pi} \lim_{\varepsilon \to 0^+} \operatorname{Re} C_{ab}(\mathbf{q}; \varepsilon - i\omega) \tag{2.17}$$

with $a(\mathbf{q}) = b^+(\mathbf{q}) = n(\mathbf{q})$. Here Re denotes the real part.

As second example, let us mention the external mobility tensor $\mu_{\alpha\beta}(\mathbf{q}; \omega)$, which describes the change of the particle current in linear response to an externally applied potential. From this transport coefficient one immediately obtains, e.g., the electrical conductivity, the dielectric constant, or the density susceptibility (for the explicit forms of these relations see chapter 4.5 of ref. 13). The mobility reads explicitly[13]

$$\mu_{\alpha\beta}(\mathbf{q}; \omega) = \frac{1}{\Omega} \int_0^\infty dt\, e^{i\omega t} \int_0^\beta d\lambda \langle j_\alpha(\mathbf{q}; t - i\lambda)\, j_\beta(-\mathbf{q})\rangle \tag{2.18}$$

with the current density operator

$$\mathbf{j}(\mathbf{q}) = \sum_{i=1}^{N} \mathbf{j}_i(\mathbf{q}) = \frac{1}{2m} \sum_{i=1}^{N} \{\hat{\mathbf{p}}_i, \exp(-i\mathbf{q} \cdot \hat{\mathbf{r}}_i)\} \tag{2.19}$$

Due to spatial isotropy we have, by choosing $\mathbf{q} = (0, 0, \mathrm{q})$,

$$\mu_{\alpha\beta}(\mathbf{q}; \omega) = \delta_{\alpha\beta}\mu_{\alpha\alpha}(\mathbf{q}; \omega) \tag{2.20}$$

with $\mu_{11} = \mu_{22}$ being the transverse (to $\mathbf{q}$) and μ_{33} the longitudinal (to $\mathbf{q}$) components. These components can also be written in terms of our basic correlation function $C_{ab}(\mathbf{q}; z)$. Indeed, expressing $\mu_{\alpha\alpha}$ in the eigenstates of the full Hamiltonian H (Lehmann representation) and using that $j_\alpha(-\mathbf{q})^+ = j_\alpha(\mathbf{q})$, we find

$$\mu_{\alpha\alpha}(\mathbf{q}; \omega) = \frac{i}{\pi} \lim_{\varepsilon \to 0^+} \int_{-\infty}^{+\infty} d\omega' \, \frac{\operatorname{Re} \mu_{\alpha\alpha}(\mathbf{q}; \omega')}{\omega - \omega' + i\varepsilon} \tag{2.21}$$

where the real part of $\mu_{\alpha\alpha}$ is given by

$$\operatorname{Re} \mu_{\alpha\alpha}(\mathbf{q}; \omega) = \frac{1 - e^{-\beta\omega}}{\omega} \lim_{\varepsilon \to 0^+} \operatorname{Re} C_{ab}(\mathbf{q}; \varepsilon - i\omega) \tag{2.22}$$

with $a(\mathbf{q}) = b^+(\mathbf{q}) = j_\alpha(\mathbf{q})$. Taking the imaginary part of Eq. (2.21), one obtains a Kramers–Kronig relation[13–15] between real and imaginary parts of $\mu_{\alpha\alpha}$. We note that $(m/n) \operatorname{Re} \mu_{11}(\mathbf{q}; \omega)$, where $n = N/\Omega$ is the density, represents the quantum version of the transverse-momentum autocorrelation function.[12,17] The longitudinal part, $\operatorname{Re} \mu_{33}$, on the other hand, is connected with the dynamic structure factor via the relation

$$S(\mathbf{q}; \omega) = \frac{\Omega}{\pi\omega} \frac{q^2}{e^{-\beta\omega} - 1} \operatorname{Re} \mu_{33}(\mathbf{q}; \omega) \tag{2.23}$$

as is easily shown by performing partial integrations and by making use of the continuity equation

$$\dot{n}(\mathbf{q}; t) + i\mathbf{q} \cdot \mathbf{j}(\mathbf{q}; t) = 0 \tag{2.24}$$

3. CLUSTER EXPANSION AND FACTORIZATION THEOREM

Having motivated the consideration of the correlation function $C_{ab}(\mathbf{q}; z)$ in the last section, we shall now turn to the evaluation of this quantity in the Enskog approximation. The procedure will be very similar to the one discussed in I. In particular, this means that we make use of cluster expansion techniques in combination with projection operators. The aim thereby is to derive a Dyson-equation-like formula for superoperators with a generalized collision operator which will be evaluated in the Enskog approximation. In order to clearly exhibit the main ideas of this procedure, we defer most of the technical steps to the Appendices.

To begin with, let us first rewrite the correlation function $C_{ab}(\mathbf{q}; z)$, given in Eq. (2.13), in a form where the restriction in the trace operation

Tr due to Fermi–Dirac (FD) or Bose–Einstein (BE) statistics is removed [see ref. 3, Eqs. (2.11-17)]:

$$C_{ab}(\mathbf{q}; z) = \mathrm{Tr}_1\, b_1(-\mathbf{q})\, h_1(\mathbf{q}; z) \tag{3.1}$$

where $h_1(\mathbf{q}; z)$ is a one-particle correlation operator given by

$$h_1(\mathbf{q}; z) = n\, \mathrm{Tr}_{2 \cdots N} \frac{1}{z - iL} fa(\mathbf{q}) \tag{3.2}$$

The quantum-statistical correlations are now absorbed in f:

$$f = \rho\pi = \pi\rho \tag{3.3}$$

where the projector π (anti-) symmetrizes the direct product of single-particle momentum eigenstates (we work exclusively in these states here):

$$\pi = \frac{1}{N!}\pi^{1 \cdots N} = \frac{1}{N!} \sum_{\sigma \in S_N} \pi_\sigma \tag{3.4}$$

$$\pi_\sigma\, |\mathbf{k}_1 \cdots \mathbf{k}_N\rangle = \eta^{|\sigma|}\, |\mathbf{k}_{\sigma(1)} \cdots \mathbf{k}_{\sigma(N)}\rangle \tag{3.5}$$

with

$$|\mathbf{k}_1 \cdots \mathbf{k}_N\rangle = |\mathbf{k}_1\rangle \times \cdots \times |\mathbf{k}_N\rangle \equiv |k\rangle \tag{3.6}$$

The sum in Eq. (3.4) runs over all permutations σ of N particles, and $\eta^{|\sigma|}$ equals 1 for bosons, whereas for fermions it equals $1(-1)$ for even (odd) permutations σ. Finally, $\mathrm{Tr}_{i \cdots j} = \mathrm{Tr}_i \cdots \mathrm{Tr}_j$ denotes the trace for Boltzmann (i.e., classical) statistics. Note that in deriving Eq. (3.1) we have used that $f \cdot La = L(fa)$.

We shall now evaluate the operator $h_1(\mathbf{q}; z)$ with the help of cluster expansions and the projection superoperator $\bar{P}^{1 \cdots s}_{-\mathbf{q}}$ (and its complement $\bar{Q}^{1 \cdots s}_{-\mathbf{q}}$) defined by

$$\bar{P}^{1 \cdots s}_{-\mathbf{q}} = \sum_{\sigma \in S_s} \pi_\sigma P^{1 \cdots s}_{-\mathbf{q}} \pi_\sigma^{-1}, \qquad \bar{Q}^{1 \cdots s}_{-\mathbf{q}} = 1 - \bar{P}^{1 \cdots s}_{-\mathbf{q}}, \qquad \mathbf{q} \neq 0 \tag{3.7}$$

with

$$P^{1 \cdots s}_{-\mathbf{q}} = \sum_{i=1}^{s} P^i_{-\mathbf{q}} P^{1 \cdots \not{i} \cdots s} \tag{3.8}$$

$$(P^i_{-\mathbf{q}} a)_{kk'} = \langle k|\, P^i_{-\mathbf{q}} a\, |k'\rangle = a_{kk'} \delta_{\mathbf{k}'_i, \mathbf{k}_i + \mathbf{q}} \tag{3.9}$$

and

$$P^{i \cdots j} = P^i \cdots P^j, \qquad P^i = P^i_{\mathbf{q}=0} \tag{3.10}$$

π_σ^{-1} denotes the inverse of the permutation operator π_σ defined in Eq. (3.5). The use of $\bar{P}^{1\cdots s}_{-\mathbf{q}}$ is motivated by the fact that due to translational invariance of the Hamiltonian, one has (cf. Appendix B of ref. 2)

$$h_1(\mathbf{q}; z) = P^1_{-\mathbf{q}} h_1(\mathbf{q}; z) = n \,\mathrm{Tr}_{2\cdots N}\, P^1_{-\mathbf{q}} P^{2\cdots N} \frac{1}{z - iL} fa(\mathbf{q}) \tag{3.11}$$

$\bar{P}^{1\cdots s}_{-\mathbf{q}}$ represents then the completely symmetrized version of $P^1_{-\mathbf{q}} P^{2\cdots s}$, which takes the effect of the FD (BE) statistics properly into account (see also the discussion given in ref. 3, pp. 771–772).

Now, with the aid of some formal manipulations, we derive in Appendix A the following result:

$$\begin{aligned}
&[z - iL_0(1)]\, h_1(\mathbf{q}; z) \\
&\quad = \sum_{s=2}^{N} \mathrm{Tr}_{2\cdots s}\, G^{\mathbf{q}}_{1\cdots s}(z)[z - iL_0(1\cdots s)] \\
&\quad \times \bar{P}^{1\cdots s}_{-\mathbf{q}} h_{1\cdots s}(\mathbf{q}; z) + n_1(\mathbf{q}; z)
\end{aligned} \tag{3.12}$$

Here the cluster superoperator $G^{\mathbf{q}}_{1\cdots s}(z)$ is given by

$$\begin{aligned}
G^{\mathbf{q}}_{1\cdots s}(z) = {} & iL_{12} \frac{1}{z - iL(12)} i\bar{Q}^{12}_{-\mathbf{q}} (L_{13} + L_{23}) \frac{1}{z - iL(123)} i\bar{Q}^{123}_{-\mathbf{q}} \\
& \times \cdots \frac{1}{z - iL(1\cdots s-1)} i\bar{Q}^{1\cdots s-1}_{-\mathbf{q}} \\
& \times (L_{1s} + \cdots + L_{s-1s}) \frac{1}{z - iL(1\cdots s)}
\end{aligned} \tag{3.13}$$

The operator $h_{1\cdots s}(\mathbf{q}; z)$ denotes the s-particle generalization of $h_1(\mathbf{q}; z)$:

$$h_{1\cdots s}(\mathbf{q}; z) = \frac{N!}{\Omega(N-s)!} \mathrm{Tr}_{s+1\cdots N} \frac{1}{z - iL} fa(\mathbf{q}) \tag{3.14}$$

The remaining one-particle operator $n_1(\mathbf{q}; z)$ reads

$$n_1(\mathbf{q}; z) = \frac{1}{\Omega} [1 + N_1(\mathbf{q}; z)]\, U_1 a_1(\mathbf{q}) \tag{3.15}$$

with

$$N_1(\mathbf{q}; z) = \mathrm{Tr}_{2\cdots N} \sum_{s=2}^{N} \frac{N!}{(N-s)!} G^{\mathbf{q}}_{1\cdots s}(z)\, \bar{Q}^{1\cdots s}_{-\mathbf{q}} \mathscr{f} \sum_{i=1}^{N} \sigma_{1i} U_1^{-1} \tag{3.16}$$

and

$$U_1 = N\,\mathrm{Tr}_{2\cdots N} f \sum_{i=1}^{N} \sigma_{1i} \tag{3.17}$$

σ_{ij} is a permutation superoperator which interchanges the indices i and j. Note that $N_1(\mathbf{q}; z)$ and the purely static factor U_1 are one-particle superoperators acting on everything to their right. Some properties of U_1 (and of its inverse U_1^{-1}) can be found in Section 3 of I (see also Appendix B of ref. 18).

We come now to the essential step in the derivation: We show in Appendix B that the projected part of $h_{1\cdots s}(\mathbf{q}; z)$ [i.e., $\bar{P}_{-\mathbf{q}}^{1\cdots s} h_{1\cdots s}(\mathbf{q}; z)$] occurring in Eq. (3.12) becomes a linear functional of $h_1(\mathbf{q}; z)$ in the thermodynamic limit (i.e., Ω, $N \to \infty$, with finite density $n = N/\Omega$). Explicitly, we have the following *factorization theorem*:

$$\bar{P}_{-\mathbf{q}}^{1\cdots s} h_{1\cdots s}(\mathbf{q}; z) = \pi^{1\cdots s} \sum_{i=1}^{s} \sigma_{1i} h_1(\mathbf{q}; z) f_2 \cdots f_s \quad \text{for } \Omega \to \infty \tag{3.18}$$

where f_i is the reduced distribution operator, generally defined by

$$f_{1\cdots s} = \frac{N!}{(N-s)!} \mathrm{Tr}_{s+1\cdots N} f \tag{3.19}$$

The permutation operator $\pi^{1\cdots s}$ is given in Eq. (3.4). The proof of formula (3.18) (see Appendix B) runs completely parallel to those given in refs. 2–4, where similar factorization theorems (in homogeneous systems) were used. We note that Eq. (3.18) is exact in the thermodynamic limit (the neglected terms are of relative order Ω^{-1}) and holds there for all z with $\mathrm{Re}\, z > 0$.

Next we insert formula (3.18) into Eq. (3.12), thereby obtaining a closed equation for $h_1(\mathbf{q}; z)$. After some transformations (for details see Appendix C), we then finally arrive at the following Dyson-equation-like expression for the one-particle correlation operator $h_1(\mathbf{q}; z)$:

$$h_1(\mathbf{q}; z) = \frac{1}{\Omega} \frac{1}{z - iL_0(1) - B_1(\mathbf{q}; z)} U_1 a_1(\mathbf{q}) \tag{3.20}$$

Here $B_1(\mathbf{q}; z)$ is a generalized collision (super-) operator given by

$$B_1(\mathbf{q}; z) = \frac{1}{1 + N_1(\mathbf{q}; z)} D_1(\mathbf{q}; z)[z - iL_0(1)] \tag{3.21}$$

where $N_1(\mathbf{q}; z)$ is defined in Eq. (3.16) and the superoperator $D_1(\mathbf{q}; z)$ reads

$$D_1(\mathbf{q}; z) = \mathrm{Tr}_{2\cdots N} \sum_{s=2}^{N} \frac{N!}{(N-s)!} G_{1\cdots s}^{\mathbf{q}}(z) f \sum_{i=1}^{N} \sigma_{1i} U_1^{-1} \tag{3.22}$$

Let us conclude this section with some remarks. First we note that Eq. (3.20), which forms the main result of this section, is exact in the thermodynamic limit and holds for all z with $\operatorname{Re} z > 0$. It represents the generalization of the corresponding homogeneous expression obtained in Eq. (3.6) of I. As in that work, the main feature of our formalism is that in the collision operator $B_1(\mathbf{q}; z)$ the *dynamic* correlations [originating from the z-dependent parts in $D_1(\mathbf{q}; z)$ and $N_1(\mathbf{q}; z)$] and the *static* correlations (originating from the equilibrium distribution f) are clearly separated. This fact immediately allows us to identify those terms in $B_1(\mathbf{q}; z)$ that are relevant for obtaining the quantum-statistical generalization of the classical Enskog theory.

Finally, we note that Eq. (3.20) can be transformed into a non-Markovian linear kinetic equation for $h_1(\mathbf{q}; t)$ by going back to time space:

$$\left[\frac{\partial}{\partial t} - iL_0(1)\right] h_1(\mathbf{q}; t) = \int_0^t dt'\, B_1(\mathbf{q}; t')\, h_1(\mathbf{q}; t - t') \tag{3.23}$$

with initial condition

$$h_1(\mathbf{q}; t = 0) = \frac{1}{\Omega}\, U_1 a_1(\mathbf{q}) \tag{3.24}$$

The memory kernel $B_1(\mathbf{q}; t)$ is the inverse Laplace transform of $B_1(\mathbf{q}; z)$ given in Eq. (3.21).

4. LINEAR QUANTUM ENSKOG EQUATION

In this section we evaluate the generalized collision operator $B_1(\mathbf{q}; z)$ given in Eq. (3.21), in the Enskog approximation, which will lead us to the linear quantum Enskog equation for inhomogeneous quantum fluids. We closely follow the discussion presented in Sections 4 and 5 of I. (Some repetition is therefore unavoidable.) As in that work, we mean by this approximation that in the dynamic part of $B_1(\mathbf{q}; z)$ only the binary collision contribution (combined with a certain short-time limit) is retained, whereas the static factor f is treated exactly. Thereby the approximations will be such that in the limit of no static correlations the linear quantum Boltzmann equation is recovered (see Section 5), whereas in the classical limit the linear revised Enskog equation results (see Section 6).

Now, in a first approximation step we replace in $B_1(\mathbf{q}; z)$ the factor $[1 + N_1(\mathbf{q}; z)]^{-1}$ by 1 (the neglected terms contain at least three-particle dynamic processes) and furthermore $z - iL_0(1)$ by z, being the leading term for large z (i.e., short times). Thus, we are left with

$$B_1(\mathbf{q}; z) \doteqdot zD_1(\mathbf{q}; z) \tag{4.1}$$

Next we replace $D_1(\mathbf{q}; z)$ by its binary collision approximation. For that purpose we have to take the many-body effects due to FD (BE) statistics properly into account. This means we have to bring $D_1(\mathbf{q}; z)$ into a renormalized form in which the dynamic s-particle contributions are explicitly grouped together. [For instance, the $s=3$ term in the unrenormalized form, Eq. (3.22), contains also a binary collision contribution due to the effect of the (anti) symmetrizer π occurring in f (see refs. 3 and 4).] This goal is achieved with the help of the resummation procedure discussed in Appendix A of I, which also holds for the projector $\bar{P}^{1\cdots s}_{-\mathbf{q}}$ considered here.

As result we find [cf. Eq. (I.4.12)]

$$zD_1(\mathbf{q}; z)\, h_1(\mathbf{q}; z) = -\frac{\partial}{\partial\lambda}\bigg|_{\lambda=0} \frac{1}{2} \operatorname{Tr}_2 \hat{T}^{\lambda}_{12}(z) \frac{z}{z - iL_0(12)} f^{\lambda}_{12} + \text{DTC} \tag{4.2}$$

where we have introduced the following notations: $\hat{T}^{\lambda}_{12}(z)$ is the renormalized Liouville t-matrix given as

$$\hat{T}^{\lambda}_{12}(z) = -\pi^{12} iL_{12} \frac{1}{z - i\hat{L}(12; \lambda)} [z - iL_0(12)] \tag{4.3}$$

Here, $\pi^{12} = 1 + \pi_{12}$, and the renormalized Liouville operator $\hat{L}(12; \lambda)$ is defined by

$$\hat{L}(12; \lambda) = L_0(12) + \hat{L}^{\lambda}_{12} \tag{4.4}$$

$$\hat{L}^{\lambda}_{12} a = S^{\lambda}_{12} V_{12} a - a V_{12} S^{\lambda}_{12} \tag{4.5}$$

S^{λ}_{12} is the quantum-statistical weighting operator[4,18–22] of the form

$$S^{\lambda}_{12} = 1 + \eta f^{\lambda}_1 + \eta f^{\lambda}_2 \tag{4.6}$$

where $\eta = -1(1)$ for FD (BE) statistics. The generalized distribution operator f^{λ}_{12} is defined by

$$f^{\lambda}_{1\cdots s} = \frac{N!}{(N-S)!} \operatorname{Tr}_{s+1\cdots N} f^{\lambda} \tag{4.7}$$

with

$$f^{\lambda} = Z^{-1}(\lambda)\, e^{-\beta H} e^{\lambda \bar{h}(\mathbf{q}; z)} \pi, \qquad Z(\lambda) = \operatorname{Tr} e^{-\beta H} e^{\lambda \bar{h}(\mathbf{q}; z)} \tag{4.8}$$

$$\bar{h}(\mathbf{q}; z) = \sum_{i=1}^{N} \sigma_{1i} U_1^{-1} h_1(\mathbf{q}; z) \tag{4.9}$$

Finally, DTC denotes dynamic triple (or higher) collision terms.

As in I [see the remarks after Eq. (I.4.16)], we finally replace the free resolvent $z/[z - iL_0(12)]$ occurring on the rhs of Eq. (4.2) by its large-z (i.e., short-time) limit 1. As before, corrections of the form L_0/z are omitted. Therefore, the generalized collision operator $B_1(\mathbf{q}; z)$ becomes in the Enskog approximation

$$B_1(\mathbf{q}; z)\, h_1(\mathbf{q}; z) \rightleftarrows B_1^{\mathrm{QE}}(z)\, h_1(\mathbf{q}; z) \tag{4.10}$$

where

$$B_1^{\mathrm{QE}}(z)\, h_1(\mathbf{q}; z) = -\frac{\partial}{\partial\lambda}\bigg|_{\lambda=0} \frac{1}{2} \mathrm{Tr}_2\, \hat{T}_{12}^{\lambda}(z)\, f_{12}^{\lambda} \tag{4.11}$$

$B_1^{\mathrm{QE}}(z)$ is the linear quantum Enskog collision operator for inhomogeneous quantum fluids in its most compact form. A more explicit representation is obtained by carrying out the λ-differentiation in Eq. (4.11), which leads to two different contributions:

$$B_1^{\mathrm{QE}}(z) = B_1^{\mathrm{QE,cl}}(z) + B_1^{\mathrm{QE},\eta}(z) \tag{4.12}$$

Here, $B_1^{\mathrm{QE,cl}}(z)$ represents the quantum analog of the revised linear classical Enskog collision operator (see Section 6) and reads explicitly

$$B_1^{\mathrm{QE,cl}}(z) = -\tfrac{1}{2} \mathrm{Tr}_2\, \hat{T}_{12}(z)[f_{12}(1+\sigma_{12}) + \mathrm{Tr}_3\, f_{123}\sigma_{13}]\, U_1^{-1} \tag{4.13}$$

where $\hat{T}_{12}(z) = \hat{T}_{12}^{\lambda=0}(z)$.

On the other hand, $B_1^{\mathrm{QE},\eta}(z)$ results from purely quantum-statistical effects and therefore has no classical counterpart (it vanishes for $\eta = 0$):

$$\begin{aligned} B_1^{\mathrm{QE},\eta}(z)\, y_1 = {} & -\frac{1}{2} \mathrm{Tr}_2\, \hat{T}_{12}(z) \frac{1}{z - iL_0(12)}\, iL_{12}^{\eta}[y_1] \\ & \times \frac{1}{z - i\hat{L}(12)}\, [z - iL_0(12)]\, f_{12} \end{aligned} \tag{4.14}$$

where $\hat{L}(12) = \hat{L}(12; \lambda = 0)$ and

$$L_{12}^{\eta}[y_1]\, a = \eta(y_1 + y_2)\, V_{12} a - \eta a V_{12}(y_1 + y_2) \tag{4.15}$$

y_i and a are arbitrary operators.

A further interesting representation of $B_1^{\mathrm{QE}}(z)$ is obtained by expressing $\hat{T}_{12}^{\lambda}(z)$ in terms of the exchange-modified Hilbert-space t-matrices[3,18,19] defined by

$$\hat{t}_{12}^{+}(E^+; \lambda) = \left\{\frac{z}{2} + i[H_0(12) - E]\right\} \frac{1}{z/2 + i[\hat{H}'(12; \lambda) - E]}\, V_{12} \tag{4.16}$$

and

$$\hat{t}_{12}^{-}(E^{-};\lambda)=V_{12}\frac{1}{z/2-i[\hat{H}(12;\lambda)-E]}\left\{\frac{z}{2}-i[H_0(12)-E]\right\} \tag{4.17}$$

where $E^{\pm}=E\pm iz/2$ and

$$\hat{H}(12;\lambda)=H_0(12)+S_{12}^{\lambda}V_{12} \tag{4.18}$$

$$\hat{H}'(12;\lambda)=H_0(12)+V_{12}S_{12}^{\lambda} \tag{4.19}$$

are renormalized Hamiltonians [note that H and H' are not self-adjoint and that $H^{+}\neq H'$, since $a(\mathbf{q})^{+}=a(-\mathbf{q})$]. Making use of Eq. (D.13) derived in Appendix D of ref. 3, we find

$$\begin{aligned}
&B_1^{\mathrm{QE}}(z)\,h_1(\mathbf{q};z)\\
&\quad=\frac{\partial}{\partial\lambda}\bigg|_{\lambda=0}\frac{i}{2\pi}\int_{-\infty}^{+\infty}dE\,\mathrm{Tr}_2\{\hat{t}_{12}^{-}(E^{-};\lambda)\,g_{12}^{\lambda}(E;z)\\
&\quad-g_{12}^{\lambda}(E;z)\,\hat{t}_{12}^{+}(E^{+};\lambda)\\
&\quad-i\hat{t}_{12}^{-}(E^{-};\lambda)\,g_{12}^{\lambda}(E;z)\,\hat{t}_{12}^{+}(E^{+};\lambda)\,S_{12}^{\lambda}\,g_{12}^{0}(E^{+})\\
&\quad-ig_{12}^{0}(E^{-})\,S_{12}^{\lambda}\hat{t}_{12}^{-}(E^{-};\lambda)\,g_{12}^{\lambda}(E;z)\,\hat{t}_{12}^{+}(E^{+};\lambda)\}
\end{aligned} \tag{4.20}$$

with the following abbreviations:

$$g_{12}^{\lambda}(E;z)=f_{12}^{\lambda}\,g_{12}^{0}(E^{+})+g_{12}^{0}(E^{-})\,f_{12}^{\lambda} \tag{4.21}$$

$$g_{12}^{0}(E^{\pm})=\frac{1}{z/2\pm i[H_0(12)-E]} \tag{4.22}$$

Note that Eq. (4.20) differs slightly from the corresponding expression obtained in I [cf. Eq. (5.8) there] due to the inhomogeneity of the system. A further evaluation of the representation (4.20) by scattering length expansion of $\hat{t}_{12}^{\pm}$ shall be discussed elsewhere.

Having determined the quantum Enskog collision operator, we are now in the position to write down its associated kinetic equation, which is obtained from Eq. (3.23) in the Markovian limit (see Section 5 of I). For this we have to assume that the kernel $B_1^{\mathrm{QE}}(t)$ in Eq. (3.23) decays rapidly to zero for times t on the order of the mean free time τ_m and that $\tau_m \gg \tau_c$, where τ_c is a typical binary collision time. This means we now use $B_1^{\mathrm{QE}}(t)$ beyond the time regime for which it has been derived [$B_1^{\mathrm{QE}}(t)$ resulted from $B_1(\mathbf{q};t)$ in a short-time limit]. Note that the same situation is met in classical hard-sphere systems[8] and that there the Enskog theory leads to very good agreement with experimental data.

Now, the Markovian limit of Eq. (3.23) becomes in the Enskog approximation

$$\left[\frac{\partial}{\partial t} - iL_0(1)\right] h_1(\mathbf{q}; t) = B_1^{\mathrm{QE}} h_1(\mathbf{q}; t) \tag{4.23}$$

where

$$B_1^{\mathrm{QE}} = \lim_{z \to 0^+} B_1^{\mathrm{QE}}(z) \tag{4.24}$$

The initial condition is given in Eq. (3.24). Equation (4.23) is the linear quantum Enskog equation governing the time evolution of the equilibrium-time correlation operator $h_1(\mathbf{q}; t)$ of an inhomogeneous quantum fluid.

We conclude this section by expressing the dynamic structure factor and the external mobility in terms of the Enskog collision operator. Using Eqs. (2.17) [(2.22), respectively], (3.1), (3.20), and (4.10), we find

$$S(\mathbf{q}; \omega) = \frac{1}{\pi} \lim_{\varepsilon \to 0^+} \operatorname{Re} \operatorname{Tr}_1 \exp(i\mathbf{q} \cdot \hat{\mathbf{r}}_1) \frac{1}{\varepsilon - i\omega - iL_0(1) - B_1^{\mathrm{QE}}(\varepsilon - i\omega)} \times U_1 \exp(-i\mathbf{q} \cdot \hat{\mathbf{r}}_1) \tag{4.25}$$

and

$$\operatorname{Re} \mu_{\alpha\alpha}(\mathbf{q}; \omega) = \frac{1 - e^{-\beta\omega}}{\Omega\omega} \lim_{\varepsilon \to 0^+} \operatorname{Re} \operatorname{Tr}_1 j_{1\alpha}(-\mathbf{q}) \times \frac{1}{\varepsilon - i\omega - iL_0(1) - B_1^{\mathrm{QE}}(\varepsilon - i\omega)} U_1 j_{1\alpha}(\mathbf{q}) \tag{4.26}$$

where the current density operator $\mathbf{j}_1(\mathbf{q})$ is given in Eq. (2.19). From the dynamic many-body point of view, the problem of the evaluation of, e.g., $S(\mathbf{q}; \mathrm{w})$ and $\mu_{\alpha\alpha}(\mathbf{q}; \omega)$ is solved—it has been converted to a complicated one-body problem, the further evaluation of which will be investigated elsewhere.

5. BOLTZMANN LIMIT OF $B_1^{\mathrm{QE}}(z)$

In this section we show that Eq. (4.23) reduces to the inhomogeneous linear Boltzmann equation in the limit of no static correlations (see also Section 6 of I).

To begin with, we note that our basic function $C_{ab}(\mathbf{q}; t)$ given in

Eq. (3.1) can be expressed in terms of a "Wigner correlation function" $h(\mathbf{q}, \mathbf{p}; t)$:

$$C_{ab}(\mathbf{q}; t) = \sum_{\mathbf{p}} \langle \mathbf{p} + \mathbf{q}/2 |\, b_1(-\mathbf{q}) \,| \mathbf{p} - \mathbf{q}/2 \rangle \, h(\mathbf{q}, \mathbf{p}; t) \tag{5.1}$$

where

$$h(\mathbf{q}, \mathbf{p}; t) = \langle \mathbf{p} - \mathbf{q}/2 |\, h_1(\mathbf{q}; t) \,| \mathbf{p} + \mathbf{q}/2 \rangle \tag{5.2}$$

We are now interested in the linear Boltzmann equation for $h(\mathbf{q}, \mathbf{p}; t)$. For this we consider the matrix element $\langle \mathbf{p} - \mathbf{q}/2 | \cdot | \mathbf{p} + \mathbf{q}/2 \rangle$ of Eq. (4.23) and find

$$\left(\frac{\partial}{\partial t} + \frac{i}{m} \mathbf{p} \cdot \mathbf{q} \right) h(\mathbf{q}, \mathbf{p}; t) = \left\langle \mathbf{p} - \frac{\mathbf{q}}{2} \right|\, B_1^{\mathrm{QE}} h_1(\mathbf{q}; t) \left| \mathbf{p} + \frac{\mathbf{q}}{2} \right\rangle \tag{5.3}$$

Now neglecting static correlations in the collision term in Eq. (5.3), we can replace the two-particle distribution operator f_{12}^{λ} occurring in B_1^{QE} [see Eq. (4.11)] by $\pi^{12} f_1^{0,\lambda} f_2^{0,\lambda}$, where $f_i^{0,\lambda}$ is equal to f_i^{λ} given in Eq. (4.7), but with the interaction V put equal to zero. The f's occurring in $\hat{T}_{12}^{\lambda}$ are also to be replaced by f^0. Furthermore, we neglect inhomogeneity corrections of order $\mathbf{q}$ (and higher) in the collision term which result from the fact that $h_1(\mathbf{q}; t)$ is not diagonal, i.e., $\langle \mathbf{p} |\, h_1(\mathbf{q}; t) \,| \mathbf{p}' \rangle \sim \delta_{\mathbf{p}', \mathbf{p}+\mathbf{q}}$. It is not difficult to see that this amounts to replacing $h_1(\mathbf{q}; t)$ (occurring in $f_i^{0,\lambda}$) by the diagonal operator $\hat{h}_1(\mathbf{q}; t)$ defined as

$$\langle \mathbf{p} |\, \hat{h}_1(\mathbf{q}; t) \,| \mathbf{p}' \rangle = \delta_{\mathbf{p}\mathbf{p}'} h(\mathbf{q}, \mathbf{p}; t) \tag{5.4}$$

and neglecting all other $\mathbf{q}$ dependences. Thus, in the limit of no static correlations we have

$$\begin{aligned} &\langle \mathbf{p} - \mathbf{q}/2 |\, B_1^{\mathrm{QE}} h_1(\mathbf{q}; t) \,| \mathbf{p} + \mathbf{q}/2 \rangle \\ &\quad \rightleftarrows - \frac{\partial}{\partial \lambda}\bigg|_{\lambda=0} \lim_{z \to 0^+} [\mathrm{Tr}_2\, \hat{T}_{12}^{\lambda}(z) f_1^{0,\lambda} f_2^{0,\lambda}]_{\mathbf{p}\mathbf{p}} \cdot [1 + \mathcal{O}(q)] \end{aligned} \tag{5.5}$$

Since $f_i^{0,\lambda}$ is now diagonal, the rhs of the foregoing equation is formally equivalent to the Boltzmann limit obtained in the homogeneous case in I [see Eq. (I.6.1)]. Therefore, we can use the result given there to obtain finally the inhomogeneous linear quantum Boltzmann equation from Eqs. (5.3) and (5.5):

$$\begin{aligned} &\left(\frac{\partial}{\partial t} + \frac{i}{m} \mathbf{p}_1 \cdot \mathbf{q} \right) h(\mathbf{q}, \mathbf{p}_1; t) \\ &\quad = -4\pi \sum_{\mathbf{p}_2, \mathbf{p}_1', \mathbf{q}_2'} \hat{\tau}(\mathbf{p}_1 \mathbf{p}_2; \mathbf{p}_1' \mathbf{p}_2') \\ &\qquad \times (1 + \eta f_{\mathbf{p}_1}^0)(1 + \eta f_{\mathbf{p}_2}^0) f_{\mathbf{p}_1'}^0 f_{\mathbf{p}_2'}^0 [\tilde{h}(\mathbf{q}, \mathbf{p}_1, t) + \tilde{h}(\mathbf{q}, \mathbf{p}_2; t) \\ &\qquad - \tilde{h}(\mathbf{q}, \mathbf{p}_1'; t) - \tilde{h}(\mathbf{q}, \mathbf{p}_2'; t)] \end{aligned} \tag{5.6}$$

Here the exchange-modified scattering cross section $\hat{\tau}$ is given by

$$\hat{\tau}(\mathbf{p}_1\mathbf{p}_2;\mathbf{p}_1'\mathbf{p}_2') = |\langle \mathbf{p}_1\mathbf{p}_2|\ \hat{t}_{12}^-(\varepsilon_{\mathbf{p}_1}+\varepsilon_{\mathbf{p}_2})\ \tfrac{1}{2}(1+\pi_{12})\ |\mathbf{p}_1'\mathbf{p}_2'\rangle|^2 \\ \times \delta(\varepsilon_{\mathbf{p}_1}+\varepsilon_{\mathbf{p}_2}-\varepsilon_{\mathbf{p}_1'}-\varepsilon_{\mathbf{p}_2'}) \tag{5.7}$$

where $\hat{t}_{12}^-(E) = \lim_{z\to 0^+} \hat{t}_{12}^-(E-iz;\lambda=0)$ [see Eq. (4.17)] depends functionally on the Fermi (Bose) distribution $f_{\mathbf{p}}^0 = \langle \mathbf{p}|\ f_1^0\ |\mathbf{p}\rangle$ due to many-body exchange effects. Here $\varepsilon_{\mathbf{p}} = \mathbf{p}^2/2m$ is the kinetic energy and

$$\tilde{h}(\mathbf{q},\mathbf{p};t) = [(1+\eta f_{\mathbf{p}}^0)\, f_{\mathbf{p}}^0]^{-1}\, h(\mathbf{q},\mathbf{p};t)$$

Finally, neglecting static correlations, we find for the initial condition of $h(\mathbf{q},\mathbf{p};t)$

$$h(\mathbf{q},\mathbf{p};t=0) \,\underline{\overrightarrow{=}}\, \frac{1}{\Omega} f_{\mathbf{p}}^0(1+\eta f_{\mathbf{p}}^0)\,\bar{a}(\mathbf{p})\cdot[1+\mathcal{O}(q)] \tag{5.8}$$

where we have used Eq. (3.24) and the relation $U_1 a_1 = f_1^0 a_1(1+\eta f_1^0) +$ static correlations, derived in I [see Eq. (I.3.14)].

For a derivation of the *nonlinear* quantum Boltzmann equation (with exchange-modified cross section) based on the Green's function formalism, see Danielewicz.[23]

6. CLASSICAL LIMIT OF $B_1^{QE}(z)$

Let us now consider the classical limit of $B_1^{QE}(z)$ given in Eq. (4.12). Replacing the quantum operators by their classical counterparts and the trace operation by a phase space integral, we see that $B_1^{QE}(z)$ reduces to

$$B^{E,cl}(x_1;z)\,h^{cl}(x_1,\mathbf{q};z) = -\int ds_2\, T_{12}^{cl}(z)\, F_{12} h^{cl}(x_1,\mathbf{q};z) \tag{6.1}$$

where

$$F_{12} = \left[\rho_2^{cl}(x_1,x_2)(1+\sigma_{12}) + \int dx_3\, \rho_3^{cl}(x_1,x_2,x_3)\,\sigma_{13}\right][U_1^{cl}]^{-1} \tag{6.2}$$

with the following notations: $x_i = (\mathbf{r}_i, \mathbf{p}_i)$ denotes position and momentum of the ith particle. $T_{12}^{cl}(z)$ is the classical Liouville t-matrix[2,24–28] given by

$$T_{12}^{cl}(z) = -iL_{12}^{cl}\frac{1}{z-iL^{cl}(12)}[z-iL_0^{cl}(12)] \tag{6.3}$$

where $L^{\mathrm{cl}}(12) = L_0^{\mathrm{cl}}(12) + L_{12}^{\mathrm{cl}}$ with

$$L_0^{\mathrm{cl}}(12) = -\frac{i}{m}\left(\mathbf{p}_1 \cdot \frac{\partial}{\partial \mathbf{r}_1} + \mathbf{p}_2 \cdot \frac{\partial}{\partial \mathbf{r}_2}\right) \tag{6.4}$$

$$L_{12}^{\mathrm{cl}} = i\frac{\partial V_{12}}{\partial \mathbf{r}_1} \cdot \left(\frac{\partial}{\partial \mathbf{p}_1} - \frac{\partial}{\partial \mathbf{p}_2}\right) \tag{6.5}$$

The canonical distribution function $\rho^{\mathrm{cl}}(X)$ reads

$$\rho^{\mathrm{cl}}(X) = \varphi_0(P)\,\rho_V(R) \tag{6.6}$$

with $X = (x_1, ..., x_N)$, etc., and where $\varphi_0(P) = \varphi_0(\mathbf{p}_1) \cdots \varphi_0(\mathbf{p}_N)$ is the Maxwell–Boltzmann distribution,

$$\varphi_0(\mathbf{p}) = \left(\frac{\beta}{2\pi m}\right)^{3/2} e^{-(\beta/2m)\,\mathbf{p}^2} \tag{6.7}$$

and $\rho_V(R)$ the configurational part,

$$\rho_V(R) = Q^{-1} e^{-\beta V}, \qquad Q = \int d\mathbf{r}_1 \cdots d\mathbf{r}_N\, e^{-\beta V} \tag{6.8}$$

ρ_s^{cl} denotes the reduced distribution function:

$$\rho_s^{\mathrm{cl}}(x_1, ..., x_s) = \varphi_0(\mathbf{p}_1, ..., \mathbf{p}_s)\, n_s(\mathbf{r}_1, ..., \mathbf{r}_s) \tag{6.9}$$

with

$$n_s(\mathbf{r}_1, ..., \mathbf{r}_s) = \frac{N!}{(N-s)!} \int d\mathbf{r}_{s+1} \cdots d\mathbf{r}_N\, \rho_V(R) \tag{6.10}$$

The classical version of U_1 reads

$$U_1^{\mathrm{cl}} = n\varphi_0(\mathbf{p}_1) + \int dx_2\, \varphi_0(\mathbf{p}_1, \mathbf{p}_2)\, n_2(\mathbf{r}_1, \mathbf{r}_2)\, \sigma_{12} \tag{6.11}$$

Finally, the function $h^{\mathrm{cl}}(x_1, \mathbf{q}; z)$ is the classical limit of $h_1(\mathbf{q}; z)$ defined in Eq. (3.2) and is given by

$$h^{\mathrm{cl}}(x_1, \mathbf{q}; z) = n \int dx_2 \cdots dx_N \frac{1}{z - iL^{\mathrm{cl}}}\, \rho^{\mathrm{cl}}(X)\, a^{\mathrm{cl}}(\mathbf{q}) \tag{6.12}$$

with

$$a^{\mathrm{cl}}(\mathbf{q}) = \sum_{i=1}^{N} \bar{a}^{\mathrm{cl}}(\mathbf{p}_i) \exp(-i\mathbf{q} \cdot \mathbf{r}_i) \tag{6.13}$$

being the classical limit of Eq. (2.14). In the notation commonly used in classical kinetic theory (see, e.g., refs. 10 and 11), Eq. (6.12) becomes (for $\mathbf{q} \neq 0$)

$$h^{\mathrm{cl}}(x, \mathbf{q}; z) = \frac{1}{\Omega} \int_0^\infty dt\, [\exp(-zt)] \int dx'\, [\exp(-i\mathbf{q} \cdot \mathbf{r}')\, \bar{a}^{\mathrm{cl}}(\mathbf{q}')\, F(x, x'; t) \tag{6.14}$$

with the one-particle, one-particle correlation function[10,11]

$$F(x, x'; t) = \left\langle \sum_{i=1}^{N} \delta(x - x_i) \left\{ \sum_{j=1}^{N} \delta(x' - x_j(t)) - n\varphi_0(\mathbf{p}') \right\} \right\rangle_{\mathrm{cl}} \tag{6.15}$$

where $\langle \cdot \rangle_{\mathrm{cl}}$ denotes the classical phase average in the canonical ensemble.

We note that Eq. (6.2) has the same form as obtained in I for the homogeneous case [see Eq. (6.5)], with the only difference that now the time correlation function $h(x_1, \mathbf{q}; t)$ depends on the position $\mathbf{r}_1$ due to the inhomogeneity of the fluid.

Equation (6.1) is not yet of the form known from classical Enskog theory. To arrive at this form, some transformations are necessary. Making use of functional derivative techniques, we show in Appendix D that F_{12} given in Eq. (6.2) reduces to the following form:

$$F_{12} = \left\{ \varphi_0(\mathbf{p}_1, \mathbf{p}_2)[1 + v_2(\mathbf{r}_1, \mathbf{r}_2)](1 + \sigma_{12}) \right.$$
$$\left. + \varphi_0(\mathbf{p}_1, \mathbf{p}_2) \int dx_3\, \varphi_0(\mathbf{p}_3)\, H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3)\, \sigma_{13} \right\} \frac{n}{\varphi_0(\mathbf{p}_1)} \tag{6.16}$$

where $v_2(\mathbf{r}_1, \mathbf{r}_2)$ is the pair correlation function given as

$$v_2(\mathbf{r}_1, \mathbf{r}_2) = \frac{n_2(\mathbf{r}_1, \mathbf{r}_2)}{n^2} - 1 \tag{6.17}$$

and $H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3)$ represents the sets of all Mayer graphs that can be obtained by replacing one field point by a root-point in the Mayer graph representation of $v_2(\mathbf{r}_1, \mathbf{r}_2)$.[10,11,29–31] In terms of functional derivatives, this can be expressed as

$$H(\mathbf{r}_1, \mathbf{r}_2 | \mathbf{r}_3) = n \frac{\delta}{\delta n_1(\mathbf{r}_3)} v_2(\mathbf{r}_1, \mathbf{r}_2; n_1(\mathbf{r})) \Big|_{n_1(\mathbf{r}) = n} \tag{6.18}$$

where $v_2(\mathbf{r}_1, \mathbf{r}_2; n_1(\mathbf{r}))$ is a functional of the local density $n_1(\mathbf{r})$ (for details see Appendix D).

Inserting this result into Eq. (6.1), we finally obtain from Eq. (4.23)

$$\left[\frac{\partial}{\partial t} - iL_0^{\rm cl}(1)\right] h^{\rm cl}(x_1, \mathbf{q}; t) = B^{\rm E,cl}(x_1)\, h^{\rm cl}(x_1, \mathbf{q}; t) \tag{6.19}$$

with

$$B^{\rm E,cl}(x_1) = \lim_{z \to 0^+} B^{\rm E,cl}(x_1; z) \tag{6.20}$$

$$B^{\rm E,cl}(x_1; z) = -\int dx_2\, T_{12}^{\rm cl}(z)\, F_{12} \tag{6.21}$$

where F_{12} is given in Eq. (6.16). The initial condition reads [see Eq. (3.24)]

$$h^{\rm cl}(x_1, \mathbf{q}; t=0) = \frac{1}{\Omega} U_1^{\rm cl} a_1^{\rm cl}(\mathbf{q}) \tag{6.22}$$

with $U_1^{\rm cl}$ given in Eq. (6.11).

Redefining our quantities $h^{\rm cl}(x_1, \mathbf{q}; t)$ and $B^{\rm E,cl}(x_1)$ as $\varphi_0^{-1}(\mathbf{p}_1)\, h^{\rm cl}(x_1, \mathbf{q}; t)$ and $\varphi_0^{-1}(\mathbf{p}_1)\, B^{\rm E,cl}(x_1)$, we see that Eq. (6.19) formally agrees with the revised linear classical Enskog equation as obtained by van Beijeren[10] [see his Eqs. (8.16) and (11.3)] and van Beijeren and Ernst.[11] As in I (see Section 6 there), the only difference is that here we consider more general short-range potentials than the hard-sphere interaction, with the consequence that in our Enskog collision operator the z-dependent t-matrix, $T_{12}^{\rm cl}(z)$, for continuous potentials occurs instead of the binary hard-sphere collision operator. For a discussion of $T_{12}^{\rm cl}(z)$ and its hard-sphere limit see McLennan (ref. 28, p. 272). Note that a similar form of the classical linear Enskog equation [containing also $T_{12}^{\rm cl}(z)$] was obtained by Mazenko (see Section 6 of ref. 27) in his fully renormalized kinetic theory. For an extension of the hard-sphere Enskog theory to a dense classical square-well fluid see also Karkheck *et al.*[32] and Leegwater *et al.*[33] (this extension, however, does not reduce to the Boltzmann equation in the low-density limit, which is a serious drawback of this theory).

7. CONCLUSIONS

The linear revised Enskog equation for classical hard spheres is generalized to normal quantum fluids. Thereby static correlations coming from the equilibrium distribution and quantum-statistical correlations due to FD (BE) statistics are fully retained, whereas in the dynamic part only uncorrelated binary collisions are taken into account (as in the Boltzmann equation). This generalized Enskog theory is applicable to systems with continuous short-range central interaction potentials.

We express physically important quantities such as the dynamic structure factor (measured in neutron or light scattering experiments) and the nonlocal mobility tensor in terms of a linear quantum Enskog collision operator. Thereby we go beyond the time regime for which this Enskog collision operator has been derived. However, exactly the same situation already occurs in the classical hard-sphere Enskog theory (as pointed out by van Beijeren and Ernst in Section 2 of ref. 8), which, nevertheless, leads to very good agreement with experimental data, in particular for Ar.[8,12] It is therefore to be expected that the quantum Enskog theory derived here when applied to real systems such as normal ^{3}He and ^{4}He, spin-polarized hydrogen, Ne, nuclear matter, etc., leads also to very good results in comparison with experiments.

In the limit when static correlations are neglected, the inhomogeneous linear quantum Boltzmann equation is recovered, and in the classical limit the linear revised Enskog equation results.

Concluding, we remark that it would be interesting to know whether the linear (quantum) Enskog equation obtained here fulfills an H-theorem similar to that proven by Résibois[36] in the classical hard-sphere case. For an H-theorem applied to the square-well fluid see also ref. 37.

APPENDIX A

In this Appendix, starting from Eq. (3.11), we derive Eq. (3.12) with the help of cluster expansions and the relation $\bar{P}^{1\cdots s}_{-\mathbf{q}} + \bar{Q}^{1\cdots s}_{-\mathbf{q}} = 1$.

By using the identity

$$\frac{1}{X-Y} = \frac{1}{X} + \frac{1}{X} Y \frac{1}{X-Y} \tag{A.1}$$

we obtain in a first step with $L = L_0 + L_v$ and $P^i L_0(i) = 0$

$$\begin{aligned} z h_1(\mathbf{q}; z) &= n \operatorname{Tr}_{2\ldots N} f a(\mathbf{q}) + n \operatorname{Tr}_{2\ldots N} iL \frac{1}{z - iL} f a(\mathbf{q}) \\ &= \frac{1}{\Omega} U_1 a_1(\mathbf{q}) + iL_0(1)\, h_1(\mathbf{q}; z) + c_1 \end{aligned} \tag{A.2}$$

with

$$c_1 = \frac{N!}{\Omega(N-2)!} \operatorname{Tr}_{2\ldots N} iL_{12} \frac{1}{z - iL} f a(\mathbf{q}) \tag{A.3}$$

Consider now c_1 and replace there L_V by

$$L_V = \bar{Q}^{12}_{-\mathbf{q}} L_V(12) + [(\bar{P}^{12}_{-\mathbf{q}} + \bar{Q}^{12}_{-\mathbf{q}})\, L_V - \bar{Q}^{12}_{-\mathbf{q}} L_V(12)] \tag{A.4}$$

with $L_V(1 \cdots s) = \sum_{1 \leqslant i < j \leqslant s} L_{ij}$. Using Eq. (A.1) and $\bar{P}^{12}_{-\mathbf{q}} + \bar{Q}^{12}_{-\mathbf{q}} = 1$, we then obtain, with $\mathrm{Tr}_{ij} L_{ij} \cdots = 0$ and $P^i L_0(i) = 0$,

$$\begin{aligned} c_1 = \frac{N!}{\Omega(N-2)!} \mathrm{Tr}_{2 \ldots N}\, G^{\mathbf{q}}_{12}(z) \Big\{ \bar{P}^{12}_{-\mathbf{q}} \Big[1 + iL_V \frac{1}{z - iL} \Big] + \bar{Q}^{12}_{-\mathbf{q}} \\ + (N-3)\, \bar{Q}^{12}_{-\mathbf{q}} i(L_{13} + L_{23}) \frac{1}{z - iL} \Big\} fa(\mathbf{q}) \end{aligned} \tag{A.5}$$

Consider now the last term in Eq. (A.5) and replace there L_V by

$$L_V = \bar{Q}^{123}_{-\mathbf{q}} L_V(123) + [(\bar{P}^{123}_{-\mathbf{q}} + \bar{Q}^{123}_{-\mathbf{q}})\, L_V - \bar{Q}^{123}_{-\mathbf{q}} L_V(123)] \tag{A.6}$$

Using Eq. (A.1) and $\bar{P}^{123}_{-\mathbf{q}} + \bar{Q}^{123}_{-\mathbf{q}} = 1$, we then obtain

$$\begin{aligned} c_1 = \frac{N!}{\Omega(N-2)!} \mathrm{Tr}_{2 \ldots N}\, G^{\mathbf{q}}_{12}(z) \Big\{ [z - iL_0(12)]\, \bar{P}^{12}_{-\mathbf{q}} \frac{1}{z - iL} + \bar{Q}^{12}_{-\mathbf{q}} \Big\} fa(\mathbf{q}) \\ + \frac{N!}{\Omega(N-3)!} \mathrm{Tr}_{2 \ldots N}\, G^{\mathbf{q}}_{123}(z) \Big\{ \bar{P}^{123}_{-\mathbf{q}} \Big[1 + iL_V \frac{1}{z - iL} \Big] + \bar{Q}^{123}_{-\mathbf{q}} \\ + (N-4)\, \bar{Q}^{123}_{-\mathbf{q}} i(L_{14} + L_{24} + L_{34}) \frac{1}{z - iL} \Big\} fa(\mathbf{q}) \end{aligned} \tag{A.7}$$

Iterating these steps and using Eq. (A.2), we arrive at the desired result, Eq. (3.12). Thereby the $\bar{Q}^{1 \cdots s}_{-\mathbf{q}}$-projectors actually occurring in the *denominators* of $G^{\mathbf{q}}_{1 \ldots s}(z)$ have been omitted in (3.13), since there the $\bar{P}^{1 \cdots s}_{-\mathbf{q}}$-parts in $Q^{1 \cdots}_{\mathbf{q}} = 1 - P^{1 \cdots s}_{-\mathbf{q}}$ lead to vanishing contributions in the thermodynamic limit due to the P_q rule.[2]

APPENDIX B

In this Appendix we sketch the proof of the factorization theorem (3.18). We can be very brief since almost the same result with the diagonal projector $P^{1 \cdots s}$ (instead of $P^{1 \cdots s}_{-\mathbf{q}}$ considered here) has been proven in ref. 3.

We first note that

$$\begin{aligned} \bar{P}^{1 \cdots}_{-\mathbf{q}} h_{1 \ldots s}(\mathbf{q}; z) &= \pi^{1 \cdots s} P^{1 \cdots s}_{-\mathbf{q}} h_{1 \ldots s}(\mathbf{q}; z) \\ &= \pi^{1 \cdots s} \sum_{i=1}^{s} \sigma_{1i} P^{1}_{-\mathbf{q}} P^{2 \cdots s} h_{1 \ldots s}(\mathbf{q}; z) \end{aligned} \tag{B.1}$$

since $\pi_\sigma^{-1} f = f$ and $\sum_{\sigma \in S_s} \pi_\sigma = \pi^{1 \cdots s}$. In the last equation we have used Eq. (3.8) and the fact that $h_{1 \ldots s}(\mathbf{q}; z)$ is symmetric in 1,..., s. Now, we make use of the factorization formula proven in ref. 3 [see Eq. (3.42) there] with the difference that instead of the projector $P^{1 \cdots s}$, we consider here $P^1_{-\mathbf{q}} P^{2 \cdots s}(\mathbf{q} \neq 0)$. This difference, however, has no influence on the factorization formula, since $P^i_{-\mathbf{q}}$ and P^i obey the same P_q rule (cf. ref. 2, Section 4) on which the proof is based. Therefor we have

$$P^1_{-\mathbf{q}} P^{2 \cdots s} h_{1 \ldots s}(\mathbf{q}; z)$$

$$= P^1_{-\mathbf{q}} P^{2 \cdots s} \sum_{i=1}^{s} \sigma_{1i} h_1(\mathbf{q}; z) f_2 \cdots f_s \qquad \text{for} \quad \Omega \to \infty \tag{B.2}$$

Inserting this result into Eq. (B.1) and using that $P^i_{-\mathbf{q}} f_i = 0$ for $\mathbf{q} \neq 0$ (since $P^i f_i = f_i$) and $h_1(\mathbf{q}; z) = P^1_{-\mathbf{q}} h_1(\mathbf{q}; z)$, we arrive at the desired result, Eq. (3.18).

APPENDIX C

The aim of this Appendix is to derive Eq. (3.20). To begin with, we insert the factorization formula (3.18) into Eq. (3.12). Using that $L_0(i) f_i = 0$ and therefore

$$[z - iL_0(1 \cdots s)]\, \pi^{1 \cdots s} \sum_{i=1}^{s} \sigma_{1i} h_1(\mathbf{q}; z) f_2 \cdots f_s$$

$$= \pi^{1 \cdots s} \sum_{i=1}^{s} \sigma_{1i} \tilde{h}_1(\mathbf{q}; z) f_2 \cdots f_s \tag{C.1}$$

with $\tilde{h}_1(\mathbf{q}; z) = [z - iL_0(1)]\, h_1(\mathbf{q}; z)$, we then obtain

$$\tilde{h}_1(\mathbf{q}; z) = \sum_{s=2}^{N} \mathrm{Tr}_{2 \ldots s}\, G^{\mathbf{q}}_{1 \ldots s}(z)\, \pi^{1 \cdots s} \sum_{i=1}^{s} \sigma_{1i} \tilde{h}_1(\mathbf{q}; z) f_2 \cdots f_s + n_1(\mathbf{q}; z) \tag{C.2}$$

Next we note that for $\mathbf{q} \neq 0$ and due to the P_q rule[2] one has

$$\tilde{h}_1(\mathbf{q}; z) f_2 \cdots f_s = P^{1 \cdots s}_{-\mathbf{q}} \tilde{h}_1(\mathbf{q}; z) f_2 \cdots f_s$$

$$= P^{1 \cdots s}_{-\mathbf{q}} f_{1 \ldots s} f_1^{-1} \tilde{h}_1(\mathbf{q}; z)[1 + o(\Omega^{-1})] \tag{C.3}$$

which inserted into (C.2) yields

$$\tilde{h}_1(\mathbf{q}; z) = \sum_{s=2}^{N} \mathrm{Tr}_{2 \ldots s}\, G^{\mathbf{q}}_{1 \ldots s}(z)\, \pi^{1 \cdots s} P^{1 \cdots s}_{-\mathbf{q}} f_{1 \ldots s}$$

$$\times \sum_{i=1}^{s} \sigma_{1i} f_1^{-1} \tilde{h}_1(\mathbf{q}; z) + n_1(\mathbf{q}; z) \tag{C.4}$$

Now, introducing for $h_1(\mathbf{q}; z)$ the identity

$$\tilde{h}_1(\mathbf{q}; z) = U_1 f_1^{-1} \bar{h}_1(\mathbf{q}; z) \tag{C.5}$$

where

$$\bar{h}_1(\mathbf{q}; z) = [U_1 f_1^{-1}]^{-1} \tilde{h}_1(\mathbf{q}; z) \tag{C.6}$$

and following the transformation steps given in Appendix A of ref. 3 (with $P^{1 \cdots s}$ replaced by $P_{-\mathbf{q}}^{1 \cdots s}$), we arrive at

$$\begin{aligned}\tilde{h}_1(\mathbf{q}; z) = \sum_{s=2}^{N} \frac{N!}{(N-S)!} \operatorname{Tr}_{2 \cdots N} G_{1 \cdots s}^{\mathbf{q}}(z) \bar{P}_{-\mathbf{q}}^{1 \cdots s} f \\ \times \sum_{i=1}^{N} \sigma_{1i} U_1^{-1} \tilde{h}_1(\mathbf{q}; z) + n_1(\mathbf{q}; z)\end{aligned} \tag{C.7}$$

Replacing $\bar{P}_{-\mathbf{q}}^{1 \cdots s}$ by $1 - \bar{Q}_{-\mathbf{q}}^{1 \cdots s}$ and using for $n_1(\mathbf{q}; z)$ Eq. (3.15), one finds

$$\begin{aligned}\tilde{h}_1(\mathbf{q}; z) = [D_1(\mathbf{q}; z) - N_1(\mathbf{q}; z)] \tilde{h}_1(\mathbf{q}; z) \\ + [1 + N_1(\mathbf{q}; z)] \frac{1}{\Omega} U_1 a_1(\mathbf{q})\end{aligned} \tag{C.8}$$

where $D_1(\mathbf{q}; z)$ is defined in Eq. (3.22). Simple manipulations lead then to the desired result, Eq. (3.20).

APPENDIX D

Using functional derivative techniques,[34,35] we show here that the static correlation operator F_{12} defined in Eq. (6.2) reduces to the form (6.16). For that purpose let us introduce a generalized distribution function of the form[27]

$$\rho(X; \psi) = Z^{-1}[\psi] \exp\left[-\beta H - \beta \sum_{i=1}^{N} \psi(x_i) \right] \tag{D.1}$$

and its reduced version

$$\rho_s(x_1, ..., x_s; \psi) = \frac{N!}{(N-s)!} \int dx_{s+1} \cdots dx_N \, \rho(X; \psi) \tag{D.2}$$

Here, ρ, ρ_s, and the normalization constant Z depend functionally on the field $\psi(\mathbf{r}_i, \mathbf{p}_i)$, with the consequence that $\rho(X; \psi)$ no longer factorizes into momentum and position parts, in contrast to $\rho(X; \psi \equiv 0) = \varphi_0(P) \cdot \rho_v(R)$.

Now F_{12} is considered as a functional of ψ. For notational simplicity, however, we shall not indicate this ψ dependence explicitly.

Taking functional derivatives of ρ_s with respect to ψ, we then find

$$\rho_2(x_1, x_2) = -\frac{1}{\beta}\frac{\delta\rho_1(x_1)}{\delta\psi(x_2)} + \rho_1(x_1)\,\rho_1(x_2) - \rho_1(x_1)\,\delta(x_1 - x_2) \quad \text{(D.3)}$$

and

$$\rho_3(x_1, x_2, x_3) = -\frac{1}{\beta}\frac{\delta\rho_2(x_1, x_2)}{\delta\psi(x_3)} + \rho_2(x_1, x_2)\,\rho_1(x_3) - \rho_2(x_1, x_2)[\delta(x_1 - x_3) + \delta(x_2 - x_3)] \quad \text{(D.4)}$$

Using the last relation in the second term of F_{12} given in Eq. (6.2), we obtain [with the abbreviation $h_i = h^{\mathrm{cl}}(x_i, \mathbf{q}; z)$]

$$F_{12} h_1 = -\frac{1}{\beta}\int dx_3\, \frac{\delta\rho_2(x_1, x_2)}{\delta\psi(x_3)}\, U_3^{-1} h_3 \quad \text{(D.5)}$$

where we have used that (for $\mathbf{q} \neq 0$)

$$\begin{aligned}\int dx_1\, \rho_1(x_1)\, U_1^{-1} h_1 &= \int d^N x\, \rho(X) \sum_{i=1}^{N} \sigma_{1i} U_1^{-1} h_1 \\ &= \frac{1}{N}\int dx_1\, U_1 U_1^{-1} h_1 = \frac{1}{N}\int dx_1\, h_1 = \frac{1}{z\Omega}\langle a(\mathbf{q})\rangle_{\mathrm{cl}} = 0 \quad \text{(D.6)}\end{aligned}$$

Next we show that $\bar{h}_1 = U_1^{-1} h_1$ can be expressed in terms of the inverse of $\delta\rho_1(x_1)/\delta\psi(x)$:

$$\bar{h}_1 = -\beta \int dx_2\, \frac{\delta\psi(x_1)}{\delta\rho_1(x_2)}\, \sigma_{12} h_1 \quad \text{(D.7)}$$

For the proof we first note that with Eq. (D.3) one has

$$\begin{aligned}h_1 = U_1 \bar{h}_1 &= \left[\rho_1(x_1) + \int dx_3\, \rho_2(x_1, x_3)\, \sigma_{13}\right] \bar{h}_1 \\ &= -\frac{1}{\beta}\int dx_3\, \frac{\delta\rho_1(x_1)}{\delta\psi(x_3)}\, \sigma_{13} \bar{h}_1 \quad \text{(D.8)}\end{aligned}$$

where we have used again Eq. (D.6). Next, multiplying the foregoing equation by

$$-\beta \int dx_2 \frac{\delta\psi(x_1)}{\delta\rho_1(x_2)} \sigma_{12} \tag{D.9}$$

from the left and making use of the fact that

$$\int dx_2\, dx_3 \frac{\delta\psi(x_1)}{\delta\rho_1(x_2)} \frac{\delta\rho_1(x_2)}{\delta\psi(x_3)} \bar{h}_3 = \int dx_3\, \delta(x_1 - x_3)\, \bar{h}_3 = \bar{h}_1 \tag{D.10}$$

we arrive at Eq. (D.7). Inserting then Eq. (D.7) into Eq. (D.5), we find F_{12} in its most compact form,

$$F_{12} h_1 = \int dx_3 \frac{\delta\rho_2(x_1, x_2)}{\delta\rho_1(x_3)} h_3 \tag{D.11}$$

Upon writing for ρ_2

$$\rho_2(x_1, x_2) = \rho_1(x_1)\, \rho_1(x_2) \frac{\rho_2(x_1, x_2)}{\rho_1(x_1)\, \rho_1(x_2)} \tag{D.12}$$

F_{12} becomes

$$\begin{aligned} F_{12} h_1 &= \rho_2(x_1, x_2)(1 + \sigma_{12}) \frac{1}{\rho_1(x_1)} h_1 \\ &\quad + \rho_1(x_1)\, \rho_1(x_2) \int dx_3 \left[\frac{\delta}{\delta\rho_1(x_3)} \nu_2(\mathbf{r}_1, \mathbf{r}_2) \right] h_3 \end{aligned} \tag{D.13}$$

Here, we have introduced the pair correlation function ν_2, also a functional of ψ [$\rho_1(x; \psi)$, respectively],

$$\nu_2(\mathbf{r}_1, \mathbf{r}_2) = \frac{\rho_2(x_1, x_2)}{\rho_1(x_1)\, \rho_1(x_2)} - 1 = \frac{n_2(\mathbf{r}_1, \mathbf{r}_2)}{n_1(\mathbf{r}_1)\, n_1(\mathbf{r}_2)} - 1 \tag{D.14}$$

where

$$\begin{aligned} n_s(\mathbf{r}_1, \ldots, \mathbf{r}_s) &= \int d\mathbf{p}_1 \cdots d\mathbf{p}_s\, \rho_s(x_1, \ldots, x_s) \\ &= Q^{-1}[\tilde{\psi}] \frac{N!}{(N-s)!} \int d\mathbf{r}_{s+1} \cdots d\mathbf{r}_N \\ &\quad \times \exp\left[-\beta v - \beta \sum_{i=1}^{N} \tilde{\psi}(\mathbf{r}_i) \right] \\ Q[\tilde{\psi}] &= \int d^N\mathbf{r} \exp\left[-\beta v - \beta \sum_{i=1}^{N} \tilde{\psi}(\mathbf{r}_i) \right] \end{aligned} \tag{D.15}$$

with

$$\exp[-\beta\tilde{\psi}(\mathbf{r})] = \int d\mathbf{p}\, \varphi_0(\mathbf{p}) \exp[-\beta\psi(\mathbf{r}, \mathbf{p})] \tag{D.16}$$

$\tilde{\psi}$ depends only on the position $\mathbf{r}$. Therefore, we can consider v_2 also a functional of $\tilde{\psi}$ $[n_1(\mathbf{r}; \tilde{\psi})$, respectively$]$. Representing $v_2[\mathbf{r}_1, \mathbf{r}_2; n_1(\mathbf{r}; \tilde{\psi})]$ as a power series in $n_1(\mathbf{r}; \tilde{\psi})$ (virial expansion[29-31]) and using that

$$\frac{\delta}{\delta\rho_1(x; \psi)} n_1(\mathbf{r}'; \tilde{\psi}) = \frac{\delta}{\delta n_1(\mathbf{r}; \tilde{\psi})} n_1(\mathbf{r}'; \tilde{\psi}) = \delta(\mathbf{r} - \mathbf{r}') \tag{D.17}$$

one obviously has

$$\frac{\delta}{\delta\rho_1(x_3; \psi)} v_2(\mathbf{r}_1, \mathbf{r}_2; n_1(\mathbf{r}; \tilde{\psi})) = \frac{\delta}{\delta n_1(\mathbf{r}_3; \tilde{\psi})} v_2(\mathbf{r}_1, \mathbf{r}_2; n_1(\mathbf{r}; \tilde{\psi})) \tag{D.18}$$

Inserting this into Eq. (D.13) and setting $\psi \equiv 0$ [i.e., $n_1(\mathbf{r}; \tilde{\psi}) = n$], we finally arrive at the desired result, Eq. (6.16).

ACKNOWLEDGMENTS

I am much indebted to A. Thellung for many valuable discussions and for his constant interest and encouragement. I am also very grateful to H. van Beijeren for a helpful correspondence on the problem considered here and to J. Dufty for helpful discussions and generous hospitality during my visit at the University of Florida in Gainesville. This work was supported by the Swiss National Science Foundation.

REFERENCES

1. D. Loss, *J. Stat. Phys.* **59**:691 (1990).
2. D. Loss and H. Schoeller, *Physica A* **150**:199 (1988).
3. D. Loss and H. Schoeller, *J. Stat. Phys.* **54**:765 (1989).
4. D. Loss and H. Schoeller, *J. Stat. Phys.* **56**:175 (1989).
5. J. L. Lebowitz, J. Percus, and J. Sykes, *Phys. Rev.* **188**:487 (1969).
6. G. F. Mazenko, T. Y. C. Wei, and S. Yip, *Phys. Rev. A* **6**:1981 (1972).
7. H. H. U. Konijnendijk and J. M. J. van Leeuwen, *Physica* **64**:342 (1973).
8. H. van Beijeren and M. H. Ernst, *Physica* **68**:437 (1973).
9. P. Résibois and M. De Leener, *Classical Kinetic Theory of Fluids* (Wiley, New York, 1977).
10. H. van Beijeren, Dissertation, Katholieke Universiteit Nijmegen (1974).
11. H. van Beijeren and M. H. Ernst, *J. Stat. Phys.* **21**:125 (1979).
12. J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids*, 2nd ed. (Academic Press, New York, 1986).

13. R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II* (Springer, 1985).
14. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, Reading, Massachusetts, 1975).
15. E. Fick and G. Sauermann, *Quantenstatistik Dynamischer Prozesse*, Vols. I/IIa (Harry Deutsch, Thun-Frankfurt a.M., 1983/1986).
16. I. M. de Schepper, E. G. D. Cohen, and B. Kamgar-Parsi, *J. Stat. Phys.* **54**:273 (1989).
17. J. R. Dorfman and T. R. Kirkpatrick, in *Proceedings of the International School of Physics E. Fermi*, C. XCVII, G. Cicotti and W. G. Hoover, eds. (North-Holland, Amsterdam, 1986).
18. D. B. Boercker and J. W. Dufty, *Phys. Rev. A* **23**:1952 (1981).
19. D. B. Boercker and J. W. Dufty, *Ann. Phys.* (N.Y.) **119**:43 (1979).
20. R. Der and R. Haberlandt, *Physica A* **79**:597 (1975).
21. R. Der and R. Haberlandt, *Physica A* **86**:25 (1977).
22. R. Der, *Ann. Phys.* (N.Y.) **34**:298 (1977).
23. P. Danielewicz, *Ann. Phys.* (N.Y.) **152**:239 (1984).
24. K. Kawasaki and I. Oppenheim, *Phys. Rev.* **139**:A 1763 (1965).
25. J. R. Dorfman and E. G. D. Cohen, *Phys. Rev. A* **6**:776 (1972).
26. R. Zwanzig, *Phys. Rev.* **129**:486 (1963).
27. G. F. Mazenko, *Phys. Rev. A* **9**:360 (1974).
28. J. A. McLennan, *Introduction to Nonequilibrium Statistical Mechanics* (Prentice Hall, Englewood Cliffs, New Jersey, 1989).
29. G. E. Uhlenbeck and G. W. Ford, in *Studies in Statistical Mechanics*, Vol. I, J. de Boer and G. E. Uhlenbeck, eds. (North-Holland, Amsterdam, 1962).
30. G. Stell, in *The Equilibrium Theory of Classical Fluids*, H. L. Frisch and J. L. Lebowitz, eds. (Benjamin, New York, 1964).
31. D. Loss, H. Schoeller, and A. Thellung, *Physica A* **155**:373 (1989).
32. J. Karkheck, H. van Beijeren, I. M. de Schepper, and G. Stell, *Phys. Rev. A* **32**:2517 (1985).
33. J. A. Leegwater, H. van Beijeren, and J. P. J. Michels, *J. Phys. Condens. Matter* **1**:237 (1989).
34. R. Balescu, *Equilibrium and Nonequilibrium Statistical Mechanics* (Wiley, New York, 1975).
35. J. L. Lebowitz and J. K. Percus, *J. Math. Phys.* **4**:116 (1963).
36. P. Résibois, *J. Stat. Phys.* **19**:593 (1978); *Physica* **94A**:1 (1979).
37. J. Blawzdziewicz and G. Stell, *J. Stat. Phys.* **56**:821 (1989).