Journal of Statistical Physics, Vol. 10, No. 2, 1974

Approximate Solutions of the Liouville Equation. IV. The Two-Body Additive Approximation

Kenneth D. Bergeron,[1] Eugene P. Gross,[1] and Rodney L. Varley[1]

Received August 28, 1973

The two-body additive approximation on the time-dependent Liouville distribution, first introduced in part I of this series, is put into the conventional form of a self-contained kinetic equation for the doublet distribution. From this point of view the approximation consists in truncating the BBGKY chain by expressing the triplet distribution as a functional of lower distributions at the same value of the time variable. To accomplish this, it is necessary to study two associated purely spatial integral equations. The doublet kinetic equation can then be written in terms of solutions of these integral equations and comparison with conventional methods of truncating the BBGKY chain can then be made. For the purpose of comparison a method of truncating the chain based on the Kirkwood superposition approximation is introduced and discussed briefly. The momentum structure of the resulting doublet kinetic equation is similar, but the nonlocality in space of our truncation introduces distinct differences in the spatial structure. The inconsistency between conventional truncations and the exact initial conditions used for the calculation of time-dependent correlation functions is pointed out. This inconsistency is not shared by the two-body additive approximation.

KEY WORDS: BBGKY hierarchy; truncation; kinetic equations; Kirkwood superposition approximation.

Work supported in part by the National Science Foundation under Contract No. NSF GH35691.

[1] Department of Physics, Brandeis University, Waltham, Massachusetts.

1. INTRODUCTION

In a previous article,[1] which we refer to as I, one of us presented an approach to the solution of the classical Liouville equation in the linear response domain. The approach was based on assuming an explicit functional form for the N-particle Liouville distribution. Here the notation was

$$f_N = \Phi(1 + F_N), \qquad \int F_N \Phi \, d\Gamma = 0$$

where Φ is the equilibrium distribution.

The one-body approximation consisted in putting

$$F_N = \sum_{i=1}^{N} \psi(\mathbf{p}_i, \mathbf{q}_i, t) - \langle \psi \rangle = \{N(\bar{1}) - \langle N(\bar{1}) \rangle\} \psi(\bar{1})$$

where the function $\psi(\mathbf{p}, \mathbf{q}, t)$ is determined by the theory. Such a restriction of the functional form of F_N is equivalent to a truncation of the hierarchy of reduced distribution functions. In the one-body additive case $\psi(1)$ determines the time-dependent singlet distribution for the same time. However, the approximate doublet distribution and higher-order distributions depend only on $\psi(1)$. Hence the doublet distribution is a linear functional of the singlet, i.e., we have a truncation. One can then use the first equation of the time-dependent BBGKY hierarchy to obtain a self-contained kinetic equation for the singlet distribution $N(1)$. This procedure is very simple, but there is one remaining subtle point. The resulting kinetic equation contains both the bare interparticle potential and the equilibrium position correlation functions. However, the theory is of a type that assumes the exact static correlation functions to be known, and is concerned with computing time-dependent correlations based on that knowledge. Therefore one can utilize the exact equilibrium hierarchy to transform the kinetic equations to a form in which the potential does not appear. The result is a simple, well-known generalization of the Vlasov equations.[2–5] It retains meaning even for strong short-range forces since the bare interparticle potential has been replaced by the Ornstein–Zernike direct correlation function.

The main new result of our previous work was to show that this method of attack can be generalized and leads at the next level of approximation to a self-contained doublet kinetic equation. Again, the bare potential is completely eliminated so that the equation retains meaning in the two extremes of strong short-range forces and long-range Coulomb forces. The differences between the two types of system are reflected in the properties of the static position correlation functions.

The purpose of this paper is to put the equations of the two-body additive approximation into a conventional form of a self-contained equation for the time-dependent doublet distribution function. We will furthermore

exhibit the relation of this unified theory to other equations, i.e., the Boltzmann–Enskog equation[6] for strong short-range forces and the Lenard–Balescu–Guernsey[7] equation for long-range Coulomb forces.

The two-body additive approximation works with a function $\Psi(\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}_1, \mathbf{q}_2, t)$. The Liouville distribution is taken to be of the form[2]

$$F_N = \tfrac{1}{2}\{\hat{N}(\bar{1}\bar{2}) - \langle N(\bar{1}\bar{2})\rangle\}\Psi(\bar{1}\bar{2}) \tag{1}$$

In this paper we simplify the presentation by not splitting off one-body additive parts explicitly. The generality of the theory is not affected, because the class of one-body additive functions is contained in the class of two-body additive functions. (For a further elaboration of this point see Appendix A.) The theory determines the form of $\Psi(12, t)$ by the observation that the function $\Psi(12)$ is directly connected to the time-dependent doublet distribution by the definition of the doublet:

$$\tilde{N}(12) \equiv \langle \hat{N}(12)F_N\rangle = \tfrac{1}{2}\{\langle \hat{N}(12)\hat{N}(\bar{3}\bar{4})\rangle - \langle N(12)\rangle\langle N(\bar{3}\bar{4})\rangle\}\Psi(\bar{3}\bar{4}) \tag{2}$$

We can consider this as an integral equation for $\Psi(12)$, with $\tilde{N}(12)$ as an inhomogeneous term. If we can solve the integral equation, we express $\Psi(12)$ as a linear functional of $\tilde{N}(12)$. This is referred to as the inversion problem. In the case of the one-body additive approximation the corresponding integral equation can be solved exactly by making a Fourier transformation in the spatial variables and obtaining a degenerate integral equation in the momentum variables. In the two-body additive approximation, however, one must introduce resolvent kernels for two associated spatial integral equations. If this is done, the momentum dependence can be handled exactly. The inversion problem is treated in Sections 2 and 3 of this paper. The analysis is somewhat tedious but must be done with care to handle the thermodynamic limit properly.

Once the inversion problem has been put into a convenient form one can immediately find the triplet distribution expressed as a functional of the lower-order distributions, by substituting in the definition

$$\tilde{N}(123) \equiv \{\langle \hat{N}(123)\hat{N}(\bar{4}\bar{5})\rangle - \langle \hat{N}(123)\rangle\langle N(\bar{4}\bar{5})\rangle\}\Psi(\bar{4}\bar{5})/2 \tag{3}$$

The triplet distribution can then be written as a linear, spatially nonlocal functional of lower-order distributions at the same value of the time variable. This is exhibited explicitly in Appendix B.

Actually, as shown in I, the theory does not require an explicit knowledge of the form of $\tilde{N}(123)$. Thus we found the doublet equation

$$\begin{aligned}[\partial\tilde{N}(12)/\partial t] + \{L(12) + L(21)\}\langle \hat{N}(12)\rangle\Psi(12) \\ + \langle \hat{N}(12\bar{3})\rangle M(12|\bar{3})\{\Psi(1\bar{3}) + \Psi(2\bar{3})\} = 0\end{aligned} \tag{4}$$

[2] We use the notation of I. In particular, $F(1\bar{2}) \equiv \int dx_2\, dp_2\, F(p_1 p_2 x_1 x_2)$.

where

$$L(12) = \frac{\mathbf{p}_1}{m}\cdot\frac{\partial}{\partial \mathbf{x}_1} + \frac{1}{\theta}\frac{\partial \log\langle \hat{N}(12)\rangle}{\partial \mathbf{x}_1}\cdot\frac{\partial}{\partial \mathbf{p}_1}$$

$$M(12|3) = \frac{\mathbf{p}_1}{m}\cdot\frac{\partial}{\partial \mathbf{x}_1} + \frac{\mathbf{p}_2}{m}\cdot\frac{\partial}{\partial \mathbf{x}_2} + \frac{1}{\theta}\frac{\partial \log\langle \hat{N}(123)\rangle}{\partial \mathbf{x}_1}\cdot\frac{\partial}{\partial \mathbf{p}_1}$$

$$+ \frac{1}{\theta}\frac{\partial \log\langle \hat{N}(123)\rangle}{\partial \mathbf{x}_2}\cdot\frac{\partial}{\partial \mathbf{p}_2} \tag{5}$$

and $\theta = 1/K_B T$. With the formal solution of the inversion problem in terms of spatial resolvent kernels one can eliminate $\Psi(12)$ and obtain a self-contained doublet kinetic equation. This is Eq. (88) of Section 4.

Section 5 is devoted to a discussion of some of the more conventional methods of truncating the hierarchy and a comparison with the present truncation.

In summary, in this paper the equations of the second approximation are put in the conventional form of a self-contained equation for the time-dependent doublet distribution function. In the streaming part of the equation the bare two-body potential is replaced by $-\theta^{-1}\log[\rho_2(\mathbf{x}_1\mathbf{x}_2)]$, where ρ_2 is the equilibrium pair distribution. The integral terms of the equation represent additional effects of the medium. In the simplest conventional approximation (neglect of the third-order cumulant) the integral terms have a convolution character, i.e., the kernels depend on the difference of spatial arguments. In previous work this fact has made possible a solution of the doublet equation using the theory of singular integral equations, provided the two-body potential in the streaming term is neglected (Lenard, Balescu, Guernsey).[7] The Boltzmann–Enskog approximation, on the other hand, handles the doublet equation by treating the direct two-body interaction term exactly, but entirely neglects the integral terms.

We show, using a conventional approach based on the superposition approximation, that in any minimally adequate unified theory of fluids the integral terms do not have a purely convolution character. (Actually, we are able to find the convolution part of the integral terms exactly for our theory.) In addition, the bare two-body interaction is modified by medium effects. Our equations exhibit precisely these features, with the fortunate circumstance that the momentum variable structure of the integral terms is tractable. The present theory has the advantage of satisfying exact requirements such as normalization conditions on the distribution functions, correct short-time behavior, etc.

It should be stressed that the natural procedure in our approach is to work directly with approximations to the Liouville distribution. The goal of the theory is the calculation of time-dependent correlation functions in terms

of equilibrium correlation functions. From this point of view the route through kinetic equations is not the most direct. Nevertheless, there is value in finding the explicit doublet kinetic equation implied by the two-body additive approximation. It is the clearest way to exhibit the relationship among various quantities of direct physical interest. For example, the hydrodynamic limit is most easily studied in the kinetic equation approach. The truncation of the triplet distribution in terms of lower-order distributions has a spatially nonlocal character that is unnatural from the point of view of conventional truncations. Yet this nonlocal character is needed to remedy deficiencies of standard truncation such as errors at $t = 0$ and for short time. To a certain extent this was already apparent in the one-body additive approximation of Zwanzig. Conventional theories fail to obtain the simple result that the Vlasov equation should be replaced by an equation involving the Ornstein–Zernike direct correlation. Our theory is not unique in this respect. For example, the theory of Lebowitz *et al.*[5] pays careful attention to the short-time behavior and is an improvement over conventional theories.

In other papers in this series we develop the analytical tools such as variational methods and projection operator techniques required to find reliable results for time-dependent correlation functions.[2,3] These methods are needed irrespective of whether one works directly with the two-body additive function or with a doublet kinetic equation.

2. STRUCTURE OF THE INVERSION PROBLEM

We start by defining

$$\chi(12) \equiv \tfrac{1}{2}\langle N(12)\rangle[\Psi(12) + \Psi(21)] \tag{6}$$

and writing the time-dependent doublet distribution in the form

$$\tilde{N}(12) = \chi(12) + \langle N(12\bar{3})\rangle\left(\frac{\chi(1\bar{3})}{\langle N(1\bar{3})\rangle} + \frac{\chi(2\bar{3})}{\langle N(2\bar{3})\rangle}\right) + \tfrac{1}{2}\langle N_{4R}(12\bar{3}\bar{4})\rangle \frac{\chi(\bar{3}\bar{4})}{\langle N(\bar{3}\bar{4})\rangle} \tag{7}$$

Here

$$\begin{aligned} \langle N(12)\rangle &= \phi_1\phi_2\rho_2(\mathbf{x}_1\mathbf{x}_2), \qquad \phi_1 \equiv \phi(\mathbf{p}_1) \\ \langle N(123)\rangle &= \phi_1\phi_2\phi_3\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ \langle N_{4R}(1234)\rangle &= \phi_1\phi_2\phi_3\phi_4\{\rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4) - \rho_2(\mathbf{x}_1\mathbf{x}_2)\rho_2(\mathbf{x}_3\mathbf{x}_4)\} \end{aligned} \tag{8}$$

The ρ's are equilibrium position correlation functions.

We now introduce, using a one-dimensional notation

$$\begin{aligned}
\chi^C(\mathbf{x}_1\mathbf{x}_2) &= \int d\mathbf{p}_1\, d\mathbf{p}_2\, \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) \\
\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) &= \int d\mathbf{p}_2\, \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) - \chi^C(\mathbf{x}_1\mathbf{x}_2)\phi_1 \\
\chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \int d\mathbf{p}_1\, \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) - \chi^C(\mathbf{x}_1\mathbf{x}_2)\phi_2 \\
\chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) - \phi_2\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) \\
&\quad - \chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\phi_1 - \phi_1\phi_2\chi^C(\mathbf{x}_1\mathbf{x}_2)
\end{aligned} \tag{9}$$

We then have

$$\begin{aligned}
\chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) + \phi_2\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) \\
&\quad + \phi_1\chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) + \phi_1\phi_2\chi^C(\mathbf{x}_1\mathbf{x}_2)
\end{aligned} \tag{10}$$

We can define $\tilde{N}^A$, $\tilde{N}^B$, and $\tilde{N}^C$ in a similar manner such that

$$\begin{aligned}
\tilde{N}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \tilde{N}^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) + \phi_2 N_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) \\
&\quad + \phi_1\tilde{N}_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) + \phi_1\phi_2\tilde{N}^C(\mathbf{x}_1\mathbf{x}_2)
\end{aligned} \tag{11}$$

The reason we have decomposed our distribution in this manner is that these four types of functions define four invariant subspaces of the linear operator relating N to χ. The structure of the operator within each subspace is different for different subspaces, but it involves only spatial variables.

We note the following normalization properties (which apply to both $\Psi(12)$ and $\tilde{N}(12)$):

$$\begin{aligned}
\int \chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_1 &= \int \chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_2 = 0 \\
\int \chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_1 &= \int \chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_2 = 0 \\
\int \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_2 &= \chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) + \phi_1\chi^C(\mathbf{x}_1\mathbf{x}_2)
\end{aligned} \tag{12}$$

And we observe that a commonly encountered quantity, involving functions from two of the subspaces, is

$$\tilde{\rho}_R(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) = \int \tilde{N}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{p}_2 = \tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) + \phi_1\tilde{N}^C(\mathbf{x}_1\mathbf{x}_2) \tag{13}$$

There are also symmetry properties that follow from the general symmetry of both χ and $\tilde{N}$:

$$\begin{aligned}
\chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \chi(\mathbf{p}_2\mathbf{p}_1|\mathbf{x}_2\mathbf{x}_1), \qquad \chi^C(\mathbf{x}_1\mathbf{x}_2) = \chi^C(\mathbf{x}_2\mathbf{x}_1) \\
\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) &= \chi_L{}^B(\mathbf{p}_1|\mathbf{x}_2\mathbf{x}_1), \qquad \chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) = \chi^A(\mathbf{p}_2\mathbf{p}_1|\mathbf{x}_2\mathbf{x}_1)
\end{aligned} \tag{14}$$

The inversion problem can now be put in the form

$$\begin{aligned}
\tilde{N}^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \chi^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) \\
\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) &= \chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) + \int [\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)/\rho_2(\mathbf{x}_1\mathbf{x}_3)]\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_3)\, d\mathbf{x}_3 \\
\tilde{N}_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) &= \chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) + \int [\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)/\rho_2(\mathbf{x}_2\mathbf{x}_3)]\chi_L{}^B(\mathbf{p}_2|\mathbf{x}_2\mathbf{x}_3)\, d\mathbf{x}_3 \\
\tilde{N}^C(\mathbf{x}_1\mathbf{x}_2) &= \chi^C(\mathbf{x}_1\mathbf{x}_2) + \int \rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\left(\frac{\chi^C(\mathbf{x}_1\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_3)} + \frac{\chi^C(\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_2\mathbf{x}_3)}\right) d\mathbf{x}_3 \\
&\quad + \frac{1}{2}\{\rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4) - \rho_2(\mathbf{x}_1\mathbf{x}_2)\rho_2(\mathbf{x}_3\mathbf{x}_4)\}\frac{\chi^C(\mathbf{x}_3\mathbf{x}_4)}{\rho_2(\mathbf{x}_3\mathbf{x}_4)}\, d\mathbf{x}_3\, d\mathbf{x}_4
\end{aligned} \tag{15}$$

The inversion problem thus amounts to finding the spatial resolvent kernels defined by

$$\begin{aligned}
\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) &= \int K_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_3)\, d\mathbf{x}_3 \\
\chi^C(\mathbf{x}_1\mathbf{x}_2) &= \int K_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4)\tilde{N}^C(\mathbf{x}_3\mathbf{x}_4)\, d\mathbf{x}_3\, d\mathbf{x}_4
\end{aligned} \tag{16}$$

The momentum variable $\mathbf{p}_1$ enters only as a parameter in the first expression. The solution for $\chi_L{}^B(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2)$ follows from the one for $\chi_R{}^B$ by the symmetry relation.

The complete formal solution to the inversion problem can be written as

$$\begin{aligned}
\chi(12) &= \tilde{N}^A(12) + \phi_2 \int K_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\tilde{N}_R{}^B(\mathbf{x}_1\mathbf{x}_3|\mathbf{p}_1)\, d\mathbf{x}_3 \\
&\quad + \phi_1 \int K_3(\mathbf{x}_2\mathbf{x}_1\mathbf{x}_3)\tilde{N}_L{}^B(\mathbf{x}_2\mathbf{x}_3|\mathbf{p}_2)\, d\mathbf{x}_3 \\
&\quad + \phi_1\phi_2 \int\int K(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4)\tilde{N}^C(\mathbf{x}_3\mathbf{x}_4)\, d\mathbf{x}_3\, d\mathbf{x}_4
\end{aligned} \tag{17}$$

It has the property

$$\begin{aligned}
\int \chi(13)\, d\mathbf{p}_3 &= \int K_3(\mathbf{x}_1\mathbf{x}_3\mathbf{x}_4)\tilde{N}_R{}^B(\mathbf{x}_1\mathbf{x}_4|\mathbf{p}_1)\, d\mathbf{x}_4 \\
&\quad + \phi_1 \int K_4(\mathbf{x}_1\mathbf{x}_3\mathbf{x}_4\mathbf{x}_5)\tilde{N}^C(\mathbf{x}_4\mathbf{x}_5)\, d\mathbf{x}_4\, d\mathbf{x}_5
\end{aligned} \tag{18}$$

It is worthwhile making the connection between the present formulation of the two-body additive approximation and the one-body approximation. This is most easily done in the language of "gauge transformations," dis-

cussed in Appendix A. There it is shown that we can break up any $\Psi(12)$ into two pieces:

$$\Psi(12) = \Psi'(12) + \{[\psi(1) + \psi(2)]/(N-1)\} \tag{19}$$

in such a way that $\Psi'(12)$ has the property

$$\tfrac{1}{2}\{\langle N(1)N(\bar{2}\bar{3})\rangle - \langle N(1)\rangle\langle N(\bar{2}\bar{3})\rangle\}\Psi'(\bar{2}\bar{3}) = 0 \tag{20}$$

Equation (2) then becomes

$$\tilde{N}(1\bar{2})/(N-1) \equiv \tilde{f}(1) = \{\langle N(1)N(\bar{2})\rangle - \langle N(1)\rangle\langle N(\bar{2})\rangle\}\psi(\bar{2}) \tag{21}$$

$$\begin{aligned}\pi(12) &\equiv N(12) - \tilde{N}_0(12) \\ &= \tfrac{1}{2}\{\langle N(12)N(\bar{3}\bar{4})\rangle - \langle N(12)\rangle\langle N(\bar{3}\bar{4})\rangle\}\Psi'(\bar{3}\bar{4})\end{aligned} \tag{22}$$

where

$$\tilde{N}_0(12) \equiv \{\langle N(12)N(\bar{3})\rangle - \langle N(1)\rangle\langle N(2\bar{3})\rangle\}\psi(\bar{3}) \tag{23}$$

We first note that (21) is the definition for the $\psi(1)$ which appears in the one-body theory. By Fourier transforming it is easy to invert this exactly, obtaining $\psi(1)$ as a functional of $\tilde{f}(1)$. In (22), therefore, $\tilde{N}_0(12)$ is also a known functional of the singlet; it is the value of $\tilde{N}(12)$ predicted by the one-body theory. Hence $\pi(12)$ and $\Psi'(12)$ represent deviations of F_N from one-body additivity. Furthermore, the kernel of the integral equation (22) is the same as that of (6), so the same solving kernels K_3 and K_4 can be used to obtain the full two-body additive theory. On the other hand, this separation, along with the formulation in terms of sectors of the momentum variable, is useful if one seeks theories intermediate between the one-body and two-body additive theories.

3. SOLUTION OF INVERSION PROBLEM

3.1. Inversion Problem for *B* Sector

We write

$$N_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) \equiv N_R{}^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{x}), \qquad \mathbf{x} = \mathbf{x}_2 - \mathbf{x}_1 \tag{24}$$

and we make use of the translational invariance of the equilibrium system to introduce the notation

$$\begin{aligned}R_2(\mathbf{x}_2 - \mathbf{x}_1) = \rho_2(\mathbf{x}_2\mathbf{x}_1), \qquad R_3(\mathbf{x}_2 - \mathbf{x}_1|\mathbf{x}_3 - \mathbf{x}_1) = \rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ R_4(\mathbf{x}_2 - \mathbf{x}_1|\mathbf{x}_3 - \mathbf{x}_1|\mathbf{x}_4 - \mathbf{x}_3) = \rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4)\end{aligned} \tag{25}$$

Then the integral equation for $\chi_R{}^B$ is

$$N_R{}^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{x}) = \chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{x}) + \int \frac{R_3(\mathbf{x}|\mathbf{y})}{R_2(\mathbf{y})}\,\chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{y})\,dy \tag{26}$$

In this case we can check that the solution may be written as

$$\chi_R^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{x}) = \int K_3(\mathbf{x}|\mathbf{y}) N_R^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{y})\, d\mathbf{y} \tag{27}$$

This involves a single kernel $K_3(\mathbf{x}|\mathbf{y})$ obeying

$$\delta(\mathbf{x} - \mathbf{z}) = K_3(\mathbf{x}|\mathbf{z}) + \int [R_3(\mathbf{x}|\mathbf{y})/R_2(\mathbf{y})] K_3(\mathbf{y}|\mathbf{z})\, d\mathbf{y} \tag{28}$$

We have a simple equation for the quantity

$$\Lambda_3(\mathbf{x}) \equiv \int K_3(\mathbf{x}|\mathbf{z})\, d\mathbf{z} \tag{29}$$

It is

$$1 = \Lambda_3(\mathbf{x}) + \int [R_3(\mathbf{x}|\mathbf{y})/R_2(\mathbf{y})] \Lambda_3(\mathbf{y})\, d\mathbf{y} \tag{30}$$

In addition, the normalization condition on $R_3(\mathbf{x}|\mathbf{y})$ yields

$$1 = (N - 1) \int K_3(\mathbf{x}|\mathbf{z})\, d\mathbf{x} \tag{31}$$

To analyze the spatial structure of the kernel $K_3(\mathbf{x}|\mathbf{y})$, we introduce Fourier transforms by

$$\begin{aligned} \langle \mathbf{k}|K_3|\mathbf{l}\rangle &\equiv (1/\Omega) \int (\exp i\mathbf{k}\cdot\mathbf{x}) K_3(\mathbf{x}|\mathbf{y}) (\exp -i\mathbf{l}\cdot\mathbf{y})\, d\mathbf{y}\, d\mathbf{x} \\ K_3(\mathbf{x}|\mathbf{y}) &= (1/\Omega) \sum \sum (\exp -i\mathbf{k}\cdot\mathbf{x})(\exp i\mathbf{l}\cdot\mathbf{y}) \langle \mathbf{k}|K_3|\mathbf{l}\rangle \\ \Delta(\mathbf{k}|\mathbf{l}) &\equiv (1/\Omega) \int (\exp i\mathbf{k}\cdot\mathbf{x}) [R_3(\mathbf{x}|\mathbf{y})/R_2(\mathbf{y})] (\exp -i\mathbf{l}\cdot\mathbf{y})\, d\mathbf{y}\, d\mathbf{x} \end{aligned} \tag{32}$$

We then obtain

$$\langle 0|K_3|\mathbf{l}\rangle \equiv [1/(N - 1)] \delta_{\mathbf{l},0} \tag{33}$$

For $\mathbf{k} \neq 0$ we have the equations

$$\Omega \delta_{\mathbf{k},\mathbf{l}} = \langle \mathbf{k}|K_3|\mathbf{l}\rangle + \sum_{\mathbf{k}'} \Delta(\mathbf{k}|\mathbf{k}') \langle \mathbf{k}'|K_3|\mathbf{l}\rangle \tag{34}$$

In analyzing these equations, it is important to note some general properties of the matrix elements $\Delta(\mathbf{k}|\mathbf{l})$. We have $\Delta(0|0) = N - 2, \Delta(0|\mathbf{l}) = 0$. The elements $\Delta(\mathbf{k}|\mathbf{k})$ and $\Delta(\mathbf{k}|0)$ are of order unity, while all the other $\Delta(\mathbf{k}|\mathbf{l})$ are of order Ω^{-1}. It is then convenient to distinguish three sectors.

Sector 1

$$\langle \mathbf{k}|K_3|0\rangle \{1 + \Delta(\mathbf{k}|\mathbf{k})\} = -[\Delta(\mathbf{k}|0)/(N - 1)] - \sum_{\substack{\mathbf{k}' \neq \mathbf{k} \\ \mathbf{k}' \neq 0}} \Delta(\mathbf{k}|\mathbf{k}') \langle \mathbf{k}'|K_3|0\rangle \tag{35}$$

This shows that $\langle \mathbf{k}|K_3|0\rangle$ is of order $1/(N - 1)$.

Sector 2

$$\langle \mathbf{k}|K_3|\mathbf{k}\rangle = \frac{1}{1+\Delta(\mathbf{k}|\mathbf{k})} - \frac{1}{1+\Delta(\mathbf{k}|\mathbf{k})} \sum_{\mathbf{k}'\neq\mathbf{k}} \Delta(\mathbf{k}|\mathbf{k}')\langle \mathbf{k}'|K_3|\mathbf{k}\rangle \tag{36}$$

Sector 3

$$\langle \mathbf{k}|K_3|\mathbf{l}\rangle = \frac{\Delta(\mathbf{k}|\mathbf{l})}{[1+\Delta(\mathbf{k}|\mathbf{k})][1+\Delta(\mathbf{l}|\mathbf{l})]} - \sum_{\substack{\mathbf{k}'\neq\mathbf{k}\\ \mathbf{k}'\neq\mathbf{l}}} \frac{\Delta(\mathbf{k}|\mathbf{k}')}{1+\Delta(\mathbf{k}|\mathbf{k})} \langle \mathbf{k}'|K_3|\mathbf{l}\rangle \tag{37}$$

It follows that $\langle \mathbf{k}|K_3|\mathbf{l}\rangle$ are of order Ω^{-1}. From Eq. (2) we see that in the infinite-volume limit

$$\langle \mathbf{k}|K_3|\mathbf{k}\rangle \to \{1+\Delta(\mathbf{k}|\mathbf{k})\}^{-1} \tag{38}$$

which is of order unity. We thus have the spatial structure of $K_3(\mathbf{x}|\mathbf{y})$

$$\begin{aligned} K_3(\mathbf{x}|\mathbf{y}) = {} & [1/(N-1)\Omega] + (1/\Omega)\sum_{\mathbf{k}\neq 0}\langle \mathbf{k}|K_3|0\rangle(\exp -i\mathbf{k}\cdot\mathbf{x}) \\ & + (1/\Omega)\sum_{\mathbf{k}\neq 0}\langle \mathbf{k}|K_3|\mathbf{k}\rangle[\exp i\mathbf{k}\cdot(\mathbf{y}-\mathbf{x}) \\ & + (1/\Omega)\sum_{\substack{\mathbf{k}\neq\mathbf{l}\\ \mathbf{k}\neq 0}}\sum\langle \mathbf{k}|K_3|\mathbf{l}\rangle \exp i(\mathbf{l}\cdot\mathbf{y}-\mathbf{k}\cdot\mathbf{x}) \end{aligned} \tag{39}$$

$$\Lambda_3(\mathbf{x}) = \int K_3(\mathbf{x}|\mathbf{y})\,d\mathbf{y} = [1/(N-1)] + \sum_{\mathbf{k}\neq 0}\langle \mathbf{k}|K_3|0\rangle \exp -i\mathbf{k}\cdot\mathbf{y} \tag{40}$$

To have a feeling for the values of the matrix elements, we note that the Kirkwood superposition approximation for the equilibrium distribution functions, namely $R_3(\mathbf{x}|\mathbf{z}) = \rho_0^{-3}R_2(\mathbf{x})R_2(\mathbf{z})R_2(\mathbf{x}-\mathbf{z})$, gives, for $\mathbf{k}\neq 0$, $\mathbf{l}\neq 0$, and $\mathbf{k}\neq\mathbf{l}$

$$\Delta(\mathbf{k}|0) = \Delta(\mathbf{k}|\mathbf{k}) = [1-(1/N)]\bar{R}_2(\mathbf{k})/\rho_0 \tag{41}$$

$$\Delta(\mathbf{k}|\mathbf{l}) = (1/\Omega\rho_0{}^3)\bar{R}_2(\mathbf{l})\bar{R}_2(\mathbf{k}-\mathbf{l}) \tag{42}$$

3.2. Inversion Problem for Sector *C*

In this case it is desirable to go to center-of-mass and relative coordinates, with $(\mathbf{x}_2-\mathbf{x}_1)=\mathbf{x}$ and $\frac{1}{2}(\mathbf{x}_1+\mathbf{x}_2)=\mathbf{x}_1+\frac{1}{2}\mathbf{x}$:

$$\begin{aligned} \chi^C(\mathbf{x}_1\mathbf{x}_2) &= \sum_{\boldsymbol{\lambda}} \chi_{\boldsymbol{\lambda}}{}^C(\mathbf{x})[\exp -i\boldsymbol{\lambda}\cdot(\mathbf{x}_1+\tfrac{1}{2}\mathbf{x})]/\Omega \\ \tilde{N}^C(\mathbf{x}_1\mathbf{x}_2) &= \sum [N_{\boldsymbol{\lambda}}{}^C(\mathbf{x})/\Omega] \exp -i\boldsymbol{\lambda}\cdot(\mathbf{x}_1+\tfrac{1}{2}\mathbf{x}) \end{aligned} \tag{43}$$

It suffices to consider each value of $\boldsymbol{\lambda}$ separately. From (15) we have the integral equation

$$(\exp -\tfrac{1}{2}i\boldsymbol{\lambda}\cdot\mathbf{x})N_{\lambda}{}^{C}(\mathbf{x}) = \{\delta(\mathbf{x}-\bar{\mathbf{z}}) + \frac{R_3(\mathbf{x}|\bar{\mathbf{z}})}{R_2(\bar{\mathbf{z}})} + (\exp -i\boldsymbol{\lambda}\cdot\mathbf{x})\frac{R_3(-\mathbf{x}|\bar{\mathbf{z}})}{R_2(\bar{\mathbf{z}})}$$
$$+ \frac{1}{2}(\exp -i\boldsymbol{\lambda}\cdot\bar{\mathbf{y}})\left[\frac{R_4(\mathbf{x}|\bar{\mathbf{y}}|\bar{\mathbf{z}})}{R_2(\bar{\mathbf{z}})} - R_2(\mathbf{x})\right]\}$$
$$\times\left\{\chi_{\lambda}{}^{C}(\bar{\mathbf{z}})\exp -\frac{1}{2}i\boldsymbol{\lambda}\cdot\bar{\mathbf{z}}\right\} \tag{44}$$

3.2.1. Spatially Homogeneous Case. It is convenient to first analyze the spatially homogeneous case, when $\boldsymbol{\lambda} = 0$. We introduce the matrix element

$$W_0(\mathbf{k}|\mathbf{k}') \equiv \frac{1}{\Omega}\int(\exp i\mathbf{k}\cdot\mathbf{x})(\exp -i\mathbf{k}'\cdot\mathbf{z})\left\{\frac{R_4(\mathbf{x}|\mathbf{y}|\mathbf{z})}{R_2(\mathbf{z})} - R_2(\mathbf{x})\right\}d\mathbf{x}\,d\mathbf{y}\,d\mathbf{z} \tag{45}$$

When we take the Fourier components, we find for $\mathbf{k} = 0$ that $\int N_0{}^{C}(\mathbf{x})\,d\mathbf{x} = 0$. In addition, as shown in Appendix A we are free to set $\int \chi_0{}^{C}(\mathbf{x})\,d\mathbf{x} = 0$. Hence we are only concerned with nonzero wave vectors. We write

$$N_0{}^{C}(\mathbf{k})/D_0(\mathbf{k}) = \bar{\chi}_0{}^{C}(\mathbf{k}) + \sum_{\substack{\mathbf{k}'\neq 0\\ \mathbf{k}'\neq\mathbf{k}}}\langle\mathbf{k}|P_0|\mathbf{k}'\rangle\bar{\chi}_0{}^{C}(\mathbf{k}') \tag{46}$$

where

$$D_0(\mathbf{k}) = 1 + 2\Delta(\mathbf{k}|\mathbf{k}) + 2W_0(\mathbf{k}|\mathbf{k})$$
$$\langle\mathbf{k}|P_0|\mathbf{k}'\rangle = [2\Delta(\mathbf{k}|\mathbf{k}') + \tfrac{1}{2}W_0(\mathbf{k}|\mathbf{k}')]/D(\mathbf{k}) \tag{47}$$

Here we have used the symmetry $\bar{\chi}_0{}^{C}(\mathbf{k}) = \bar{\chi}_0{}^{C}(-\mathbf{k})$.

Analysis of the matrix elements $W_0(\mathbf{k}|\mathbf{k}')$ by decomposition into Mayer clusters shows that when $\mathbf{k}' \neq \mathbf{k}$ the matrix element is of order unity. Otherwise it is of order Ω^{-1}. If we now introduce a resolvent kernel

$$\bar{\chi}_0{}^{C}(\mathbf{k}) = \sum_{\mathbf{l}}\langle\mathbf{k}|\mathbf{K}_{40}|\mathbf{l}\rangle\bar{N}_0{}^{C}(\mathbf{l}) \tag{48}$$

we find that in the thermodynamic limit the diagonal element is

$$\langle\mathbf{k}|\mathbf{K}_{40}|\mathbf{k}\rangle \to D_0{}^{-1}(\mathbf{k}) \tag{49}$$

The matrix element $\langle\mathbf{k}|\mathbf{K}_{40}|-\mathbf{k}\rangle$ is zero. The other off-diagonal elements obey the equation

$$\mathbf{k} \neq \pm\mathbf{l}, \qquad \langle\mathbf{k}|K_{40}|\mathbf{l}\rangle + \sum_{\mathbf{k}'\neq\pm\mathbf{k}}\langle\mathbf{k}|P_0|\mathbf{k}'\rangle\langle\mathbf{k}'|K_{40}|\mathbf{l}\rangle = 0 \tag{50}$$

To estimate the matrix elements occurring, we make use of the generalized superposition principle

$$\rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\mathbf{x}_4) = \rho_0^{-8}\rho_2(\mathbf{x}_1\mathbf{x}_2)\rho_2(\mathbf{x}_1\mathbf{x}_3)\rho_2(\mathbf{x}_1\mathbf{x}_4)\rho_2(\mathbf{x}_2\mathbf{x}_3)\rho_2(\mathbf{x}_2\mathbf{x}_4)\rho_2(\mathbf{x}_3\mathbf{x}_4) \tag{51}$$

We find

$$W_0(\mathbf{k}|\mathbf{k}') = (\rho_0^{-8}/\Omega)\int d\mathbf{x}\, F_{\mathbf{k}'}(\mathbf{x})F_{-\mathbf{k}'}(\mathbf{x})R_2(\mathbf{x})(\exp i\mathbf{k}\cdot\mathbf{x})\, d\mathbf{x} \tag{52}$$

with

$$F_{\mathbf{k}'}(\mathbf{x}) = \int (\exp -\mathbf{k}'\cdot\boldsymbol{\xi})R_2(\boldsymbol{\xi} + \mathbf{x})R_2(\boldsymbol{\xi})\, d\boldsymbol{\xi} \tag{53}$$

The $F_{\mathbf{k}}(\mathbf{x})$ have the properties

$$F_{\mathbf{k}}(\mathbf{x}) = F_{-\mathbf{k}}(-\mathbf{x}) = (\exp i\mathbf{k}\cdot\mathbf{x})F_{-\mathbf{k}}(\mathbf{x}) \tag{54}$$

It is easy to verify the assertions concerning the order of magnitude of the $W_0(\mathbf{k}|\mathbf{k}')$. A rigorous demonstration can be made using a cluster development of the four-body correlation function.

3.2.2. Spatially Inhomogeneous Case. The most difficult part of the inversion problem is the spatially inhomogeneous case for sector C. We have already written $\chi^C(\mathbf{x}_1\mathbf{x}_2)$ in the form of Eq. (44). We will be concerned with the Fourier transform

$$\begin{aligned}\bar{M}_{\boldsymbol{\lambda}}{}^C(\mathbf{k}) &\equiv \int (\exp i\mathbf{k}\cdot\mathbf{x})(\exp -\tfrac{1}{2}i\boldsymbol{\lambda}\cdot\mathbf{x})N_{\boldsymbol{\lambda}}{}^C(\mathbf{x})\, d\mathbf{x}\\ \bar{\chi}_{\boldsymbol{\lambda}}{}^C(\mathbf{k}) &\equiv \int (\exp i\mathbf{k}\cdot\mathbf{x})(\exp -\tfrac{1}{2}i\boldsymbol{\lambda}\cdot\mathbf{x})\chi_{\boldsymbol{\lambda}}{}^C(\mathbf{x})\, d\mathbf{x}\end{aligned} \tag{55}$$

Another useful form is the double Fourier transform representation

$$\chi^C(\mathbf{x}_1\mathbf{x}_2) = (1/\Omega^2)\sum_{\mathbf{k}_1\mathbf{k}_2}\bar{\chi}^C(\mathbf{k}_1|\mathbf{k}_2)\exp i(\mathbf{k}_1\cdot\mathbf{x}_1 + \mathbf{k}_2\cdot\mathbf{x}_2) \tag{56}$$

The connection between the two descriptions is given by

$$\bar{\chi}^C(\boldsymbol{\lambda} - \mathbf{k}|\mathbf{k}) \equiv \bar{\chi}_{\boldsymbol{\lambda}}{}^C(\mathbf{k}) \tag{57}$$

The singlet contributions to $\bar{N}^C(\mathbf{k}_1\mathbf{k}_2)$ come from $\mathbf{k}_1 = 0$ or $\mathbf{k}_2 = 0$. Thus in $\bar{M}_{\boldsymbol{\lambda}}{}^C(\mathbf{k})$ these contributions are contained in the components $\mathbf{k} = \boldsymbol{\lambda}$ and $\mathbf{k} = 0$. The permutation symmetry $\bar{\chi}^C(\mathbf{k}_1|\mathbf{k}_2) = \bar{\chi}^C(\mathbf{k}_2|\mathbf{k}_1)$ has its counterpart in the relation

$$\bar{\chi}_{\boldsymbol{\lambda}}(\mathbf{k}) = \bar{\chi}_{\boldsymbol{\lambda}}(\boldsymbol{\lambda} - \mathbf{k}) \tag{58}$$

and in particular $\bar{\chi}_{\boldsymbol{\lambda}}(0) = \bar{\chi}_{\boldsymbol{\lambda}}(\boldsymbol{\lambda})$.

The Fourier transformation of the inversion equation is

$$\bar{M}_\lambda{}^C(\mathbf{k}) = \bar{\chi}_\lambda{}^C(\mathbf{k}) + \sum \{\Delta(\mathbf{k}|\mathbf{k}') + \Delta(\boldsymbol{\lambda} - \mathbf{k}|\mathbf{k}') + \tfrac{1}{2}W_\lambda(\mathbf{k}|\mathbf{k}')\}\bar{\chi}_\lambda{}^C(\mathbf{k}') \tag{59}$$

where

$$W_\lambda(\mathbf{k}|\mathbf{k}') = \frac{1}{\Omega}\int (\exp i\mathbf{k}\cdot\mathbf{x})(\exp -i\boldsymbol{\lambda}\cdot\mathbf{y})(\exp -i\mathbf{k}'\cdot\mathbf{z})\,\frac{R_4(\mathbf{x}|\mathbf{y}|\mathbf{z})}{R_2(\mathbf{z})}\,d\mathbf{x}\,d\mathbf{y}\,d\mathbf{z} \tag{60}$$

We have made use of the gauge freedom to set $\int \chi^C(\mathbf{x}, \mathbf{y})\,d\mathbf{x}\,d\mathbf{y} = 0$ (see Appendix A). Our object now is to find a solution in the form

$$\bar{\chi}_\lambda{}^C(\mathbf{k}) = \sum \langle \mathbf{k}|K_{4\lambda}|\mathbf{l}\rangle \bar{M}_\lambda{}^C(\mathbf{l}) \tag{61}$$

and thus to find the solution of the inversion problem as

$$\begin{aligned} \chi_\lambda{}^C(\mathbf{x}) &= \int K_{4\lambda}(\mathbf{x}|\mathbf{y})N_\lambda{}^C(\mathbf{y})\,d\mathbf{x} \\ K_{4\lambda}(\mathbf{x}|\mathbf{y}) &= (1/\Omega)\sum (\exp i\mathbf{l}\cdot\mathbf{y})(\exp -i\mathbf{k}\cdot\mathbf{x})(\exp \tfrac{1}{2}i(\mathbf{x} - \mathbf{y})\cdot\boldsymbol{\lambda})\langle \mathbf{k}|K_{4\lambda}|\mathbf{l}\rangle \end{aligned} \tag{62}$$

The complete inversion is then given by (43).

Let us first isolate the singlet contribution by taking $\mathbf{k} = 0$. We use the results

$$W_\lambda(0|\mathbf{k}') = (N - 3)\,\Delta(\boldsymbol{\lambda}|\mathbf{k}'), \qquad \Delta(0|\mathbf{k}') = (N - 2)\,\delta_{\mathbf{k}',0} \tag{63}$$

Then

$$\bar{M}_\lambda{}^C(0)/(N - 1) = [1 + \Delta(\boldsymbol{\lambda}|0)]\bar{\chi}_\lambda{}^C(0) + \tfrac{1}{2}\sum_{\substack{\mathbf{k}'\neq 0\\ \mathbf{k}'\neq\boldsymbol{\lambda}}} \Delta(\boldsymbol{\lambda}|\mathbf{k}')\bar{\chi}_\lambda{}^C(\mathbf{k}') \tag{64}$$

Next we write the equation for $\mathbf{k} \neq 0$ (and $\mathbf{k} \neq \boldsymbol{\lambda}$). We isolate the terms involving $\bar{\chi}_\lambda{}^C(0)$ and $\bar{\chi}_\lambda{}^C(\boldsymbol{\lambda})$ and eliminate them with the help of the preceding equation. To simplify writing, introduce

$$E_\lambda(\mathbf{k}) \equiv \frac{\Delta(\boldsymbol{\lambda} - \mathbf{k}|0) + \Delta(\mathbf{k}|0) + \tfrac{1}{2}[W_\lambda(\mathbf{k}|0) + W_\lambda(\mathbf{k}|\boldsymbol{\lambda})]}{1 + \tfrac{1}{2}[\Delta(\boldsymbol{\lambda}|0) + \Delta(\boldsymbol{\lambda}|\boldsymbol{\lambda})]} \tag{65}$$

$$\begin{aligned} H_\lambda(\mathbf{k}) = {} & \Delta(\mathbf{k}|\mathbf{k}) + \Delta(\boldsymbol{\lambda} - \mathbf{k}|\boldsymbol{\lambda} - \mathbf{k}) \\ & + \tfrac{1}{2}[W_\lambda(\mathbf{k}|\mathbf{k}) + W_\lambda(\mathbf{k}|\boldsymbol{\lambda} - \mathbf{k})] \end{aligned} \tag{66}$$

$$\begin{aligned} P_\lambda(\mathbf{k}|\mathbf{k}') = {} & [1 + H_\lambda(\mathbf{k})]^{-1}\,[\Delta(\mathbf{k}|\mathbf{k}') + \Delta(\boldsymbol{\lambda} - \mathbf{k}|\mathbf{k}') \\ & + \tfrac{1}{2}W_\lambda(\mathbf{k}|\mathbf{k}') - \tfrac{1}{2}E_\lambda(\mathbf{k})\,\Delta(\boldsymbol{\lambda}|\mathbf{k}')] \end{aligned} \tag{67}$$

Then for $\mathbf{k} \neq 0$ or $\boldsymbol{\lambda}$,

$$\chi_{\lambda}{}^{C}(\mathbf{k}) + \sum_{\substack{\mathbf{k}' \neq 0 \\ \mathbf{k}' \neq \boldsymbol{\lambda}}} \bar{\chi}_{\lambda}(\mathbf{k}') P_{\lambda}(\mathbf{k}|\mathbf{k}') = \frac{\bar{M}_{\lambda}{}^{C}(\mathbf{k}) - [E_{\lambda}(\mathbf{k})/(N-1)]\bar{M}_{\lambda}{}^{C}(0)}{1 + H_{\lambda}(\mathbf{k})} \tag{68}$$

The $P_{\lambda}(\mathbf{k}|\mathbf{k}')$ are of order $1/\Omega$, and the sum is over only one index. The situation is now the same as for our previous considerations. In the thermodynamic limit we have for $\mathbf{k} \neq 0$

$$\langle \mathbf{k}|\bar{K}_{4\lambda}|\mathbf{k}\rangle = 1/[1 + H_{\lambda}(\mathbf{k})] \tag{69}$$

The off-diagonal matric elements $\langle \mathbf{k}|\bar{K}_{4\lambda}|0\rangle$ and $\langle \mathbf{k}|\bar{K}_{4\lambda}|\mathbf{l}\rangle$ for $\mathbf{l} \neq 0$ require the solution of the nontrivial integral equation (68). This is a nontrivial integral equation but it is now in a form amenable to a variety of iterative solutions. However, we will not at this time investigate further the details of such solutions.

When these matrix elements have been obtained we can revert to the singlet equation (64) and write

$$\chi_{\lambda}{}^{C}(0) = \left\{1 + \frac{\Delta(\boldsymbol{\lambda}|0) + \Delta(\boldsymbol{\lambda}|\boldsymbol{\lambda})}{2}\right\}^{-1} \times \left\{\frac{\bar{M}_{\lambda}{}^{C}(0)}{N-1} - \frac{1}{2}\sum_{\mathbf{l}'}\sum_{\substack{\mathbf{k}' \neq 0 \\ \mathbf{k}' \neq \boldsymbol{\lambda}}} \langle \mathbf{k}'|\bar{K}_{4\lambda}|\mathbf{l}'\rangle \Delta(\boldsymbol{\lambda}|\mathbf{k}')\bar{M}_{\lambda}{}^{C}(\mathbf{l}')\right\} \tag{70}$$

from which we obtain $\langle 0|K_{4\lambda}|\mathbf{k}'\rangle$ directly.

4. THE DOUBLET KINETIC EQUATION

We now proceed to use the analysis of the inversion problem together with Eq. (4) to derive a self-contained kinetic equation for the time-dependent doublet distribution. We begin by defining

$$S(12) \equiv L(12) + L(21) \tag{71}$$

$$M'(12|3) \equiv M(12|3) - S(12) \tag{72}$$

where $M'(12|3)$ does not contain free streaming operators. Writing Eq. (4) in terms of these new operators and using equations (6) and (7) one obtains

$$\{(\partial/\partial t) + S(12)\}\tilde{N}(12) + \mu_1 + \mu_2 = 0 \tag{73}$$

where the medium terms are

$$\mu_1 \equiv M'(12|\bar{3})\langle \hat{N}(12\bar{3})\rangle\{[\chi(1\bar{3})/\langle N(1\bar{3})\rangle] + [\chi(2\bar{3})/\langle N(2\bar{3})\rangle]\} \tag{74}$$

$$\mu_2 \equiv -\tfrac{1}{2}S(12)\langle \hat{N}_{4R}(12\bar{3}\bar{4})\rangle\chi(\bar{3}\bar{4})/\langle N(\bar{3}\bar{4})\rangle \tag{75}$$

The doublet kinetic equation has the feature that the two-body streaming operator $S(12)$ depends on $-\theta^{-1}\log R_2(x)$ rather than on the bare two-body potential found in the BBGKY chain.

4.1. Spatially Homogeneous Disturbances

For the spatially homogeneous nonequilibrium problem we have $\chi(12) = \chi(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_2 - \mathbf{x}_1)$ and $\tilde{N}(12) = \tilde{N}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_2 - \mathbf{x}_1)$ in addition to the translational invariance of the equilibrium distribution functions indicated in Eq. (25).

The term μ_2 has a contribution only from the C sector. Using the properties $\chi^C(-\mathbf{x}) = \chi^C(\mathbf{x})$ and $\chi^C(\bar{\mathbf{x}}) = 0$, we find

$$\mu_2 = -\tfrac{1}{2}\phi_1\phi_2[(\mathbf{p}_2 - \mathbf{p}_1)/m]\cdot\mathbf{D}_\mathbf{x}R_4(\mathbf{x}|\bar{\mathbf{y}}|\bar{\mathbf{w}})\chi^C(\bar{\mathbf{w}})/R_2(\bar{\mathbf{w}}) \tag{76}$$

On the other hand, μ_1 has contributions from the B and C sectors, and we write $\mu_1 = \mu_1{}^B + \mu_1{}^C$. We first introduce the vector operator

$$\mathbf{D}_\mathbf{x} = \frac{\partial}{\partial\mathbf{x}} - \frac{\partial \log R_2(\mathbf{x})}{\partial\mathbf{x}} \tag{77}$$

$$\mu_1 = -\frac{1}{\theta}\phi_2\frac{\partial}{\partial\mathbf{p}_1}\cdot\left[D_\mathbf{x}R_3(\mathbf{x}|\bar{\mathbf{y}}) + \frac{\partial R_3(\mathbf{x}|\bar{\mathbf{y}})}{\partial\mathbf{y}}\right]\frac{\chi(\mathbf{p}_1\bar{\mathbf{p}}_3|\bar{\mathbf{y}})}{R_2(\bar{\mathbf{y}})}$$
$$+ (1\leftrightarrow 2) + \frac{1}{\theta}\frac{\partial\phi_2}{\partial\mathbf{p}_2}\cdot D_\mathbf{x}R_3(\mathbf{x}|\bar{\mathbf{y}})\frac{\chi(\mathbf{p}_1\mathbf{p}_3|\bar{\mathbf{y}})}{R_2(\bar{\mathbf{y}})} + (1\leftrightarrow 2) \tag{78}$$

And we note that in the homogeneous case (13) becomes

$$\int\chi(\mathbf{p}_1\mathbf{p}_3|\mathbf{y})\,d\mathbf{p}_3 = \phi_1\chi^C(\mathbf{y}) + \chi_R{}^B(\mathbf{p}_1|\mathbf{y}) \tag{79}$$

so that

$$\mu_1{}^C = -\phi_1\phi_2\frac{\mathbf{p}_2 - \mathbf{p}_1}{m}\cdot\frac{\partial R_3(\mathbf{x}|\bar{\mathbf{w}})}{\partial\mathbf{w}}\frac{\chi^C(\bar{\mathbf{w}})}{R_2(\bar{\mathbf{w}})} \tag{80}$$

We now use the inversion for χ^C in Eq. (16):

$$\mu_2 + \mu_1{}^C = [(\mathbf{p}_1 - \mathbf{p}_2)/m]\cdot\phi_1\phi_2\mathbf{F}(\mathbf{x}|\bar{\mathbf{z}})\tilde{N}^C(\bar{\mathbf{z}}) \tag{81}$$

where

$$\mathbf{F}(\mathbf{x}|\mathbf{z}) = \{\tfrac{1}{2}\mathbf{D}_\mathbf{x}R_4(\mathbf{x}|\bar{\mathbf{y}}|\bar{\mathbf{w}}) + [\partial R_3(\mathbf{x}|\bar{\mathbf{w}})/\partial\mathbf{w}]\}K_{40}(\bar{\mathbf{w}}|\mathbf{z})/R_2(\bar{\mathbf{w}}) \tag{82}$$

To find $\mu_1{}^B$ we use the inversion with (78) and (79)

$$\mu_1{}^B = -(1/\theta)\phi_2(\partial/\partial\mathbf{p}_1)\cdot G(\mathbf{x}|\bar{\mathbf{z}})\tilde{N}_R{}^B(\mathbf{p}_1|\bar{\mathbf{z}}) + (1\leftrightarrow 2)$$
$$+ (1/\theta)(\partial\phi_2/\partial\mathbf{p}_2)\mathbf{H}(\mathbf{x}|\bar{\mathbf{z}})\tilde{N}_R{}^B(\mathbf{p}_1|\bar{\mathbf{z}}) + (1\leftrightarrow 2) \tag{83}$$

where

$$\mathbf{H}(\mathbf{x}|\mathbf{z}) = \mathbf{D}_{\mathbf{x}}\{\delta(\mathbf{x}-\mathbf{z}) - K_3(\mathbf{x}|\mathbf{z})\} = \mathbf{D}_{\mathbf{x}}\{[R_3(\mathbf{x}|\bar{\mathbf{y}})/R_2(\bar{\mathbf{y}})]K_3(\bar{\mathbf{y}}|\mathbf{z})\} \quad (84)$$

by Eq. (27), and

$$\mathbf{G}(\mathbf{x}|\mathbf{z}) = \mathbf{H}(\mathbf{x}|\mathbf{z}) + [\partial R_3(\mathbf{x}|\bar{\mathbf{y}})/\partial \mathbf{y}][K_3(\bar{\mathbf{y}}|\mathbf{z})/R_2(\bar{\mathbf{y}})] \quad (85)$$

To isolate the singlet terms, we express the pair distribution in terms of its cumulant and singlet parts

$$\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{y}) = \tilde{P}^B(\mathbf{p}_1|\mathbf{y}) + \rho_0 \tilde{f}(\mathbf{p}_1) \quad (86)$$

There is no singlet from the C sector in the homogeneous case. We then define

$$\mathbf{H}_s(\mathbf{x}) = \mathbf{H}(\mathbf{x}|\bar{\mathbf{z}}) = [-\partial \log R_2(\mathbf{x})/\partial \mathbf{x}] - \mathbf{D}_{\mathbf{x}}\Lambda_3(\mathbf{x}), \qquad \mathbf{G}_s(\mathbf{x}) = \mathbf{G}(\mathbf{x}|\bar{\mathbf{z}}) \quad (87)$$

to obtain the final form of the doublet equation for a homogeneous system:

$$\left[\frac{\partial}{\partial t} + S(12)\right]\tilde{N}(12) + \left\{\frac{\rho_0}{\theta}\frac{\partial \phi_2}{\partial \mathbf{p}_2}\cdot\mathbf{H}_s(\mathbf{x})\tilde{f}(\mathbf{p}_1) - \frac{\rho_0}{\theta}\phi_2\mathbf{G}_s(\mathbf{x})\frac{\partial f(\mathbf{p}_1)}{\partial \mathbf{p}_1}\right.$$
$$\left. + \frac{1}{\theta}\frac{\partial \phi_2}{\partial \mathbf{p}_2}\mathbf{H}(\mathbf{x}|\bar{\mathbf{z}})\tilde{P}_R{}^B(\mathbf{p}_1|\bar{\mathbf{z}}) - \frac{1}{\theta}\phi_2\mathbf{G}(\mathbf{x}|\bar{\mathbf{z}})\frac{\partial \tilde{P}_R{}^B(\mathbf{p}_1|\bar{\mathbf{z}})}{\partial \mathbf{p}_1}\right\}$$
$$+ \{1 \leftrightarrow 2\} + \frac{\mathbf{p}_1 - \mathbf{p}_2}{m}\phi_1\phi_2\mathbf{F}(\mathbf{x}|\bar{\mathbf{z}})\tilde{N}^C(\bar{\mathbf{z}}) = 0 \quad (88)$$

The terms indicated by $\{1 \leftrightarrow 2\}$ are obtained by exchanging $\mathbf{x} \leftrightarrow -\mathbf{x}$, $\mathbf{p}_1 \leftrightarrow \mathbf{p}_2$ in the entire preceding bracketed expression.

The momentum structure of this equation is quite simple. It is the nonlocal spatial structure of the medium terms which complicates the equation. Note that the kinetic equation requires equilibrium correlation functions instead of interparticle potentials as in the usual theories.

4.2. Spatially Inhomogeneous Doublet Equation

The term $\mu_1{}^B$ is very similar to that for the homogeneous case. The quantity $\chi^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \mathbf{z})$ replaces $\chi^B(\mathbf{p}_1|\mathbf{z})$. Thus

$$\mu_1{}^B = -\frac{1}{\theta}\left\{\phi_2 G(\mathbf{x}|\mathbf{z})\cdot\frac{\partial}{\partial \mathbf{p}_1} - \frac{\partial}{\partial \mathbf{p}_2}\cdot\mathbf{H}(\mathbf{x}|\mathbf{z})\right\}\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1|\mathbf{x}_1 + \bar{\mathbf{z}}) + (1 \leftrightarrow 2) \quad (89)$$

Using (61),

$$\mu_1{}^C = \frac{1}{\theta\Omega}\sum_{\boldsymbol{\lambda}}(\exp -i\boldsymbol{\lambda}\cdot\mathbf{x}_1)\left\{\phi_1\frac{\partial \phi_2}{\partial \mathbf{p}_2}\cdot\mathbf{J}_{\boldsymbol{\lambda}}(\mathbf{x}|\bar{\mathbf{y}}) - \phi_2\frac{\partial \phi_1}{\partial \mathbf{p}_1}\mathbf{I}_{\boldsymbol{\lambda}}(\mathbf{x}|\bar{\mathbf{y}})\right\} \quad (90)$$
$$\times \tilde{N}_{\boldsymbol{\lambda}}{}^C(\bar{\mathbf{y}}) + (1 \leftrightarrow 2)$$

where

$$\mathbf{I}_\lambda(\mathbf{x}|\mathbf{y}) = \left\{\mathbf{D}_\mathbf{x} R_3(\mathbf{x}|\bar{\mathbf{z}}) + \frac{\partial R_3(\mathbf{x}|\bar{\mathbf{z}})}{\partial \bar{\mathbf{z}}}\right\}(\exp -\tfrac{1}{2}i\boldsymbol{\lambda}\cdot\mathbf{z})\,\frac{K_{4\lambda}(\bar{\mathbf{z}}|\mathbf{y})}{R_2(\bar{\mathbf{z}})} \tag{91}$$

$$\mathbf{J}_\lambda(\mathbf{x}|\mathbf{y}) = \mathbf{D}_\mathbf{x} R_3(\mathbf{x}|\bar{\mathbf{z}})\,\frac{K_{4\lambda}(\bar{\mathbf{z}}|\mathbf{y})}{R_2(\bar{\mathbf{z}})}\,(\exp -\tfrac{1}{2}i\boldsymbol{\lambda}\cdot\bar{\mathbf{z}}) \tag{92}$$

We also have

$$\mu_2 = \frac{\phi_1\phi_2}{2}\,\mathbf{p}_1\cdot\left[\frac{\partial}{\partial \mathbf{x}_1} - \frac{\partial \log \rho_2(\mathbf{x}_1\mathbf{x}_2)}{\partial \mathbf{x}_1}\right]\frac{R_4(\mathbf{x}_2 - \mathbf{x}_1|\bar{\mathbf{x}}_3 - \mathbf{x}_1|\bar{\mathbf{z}})}{R_2(\bar{\mathbf{z}})}$$

$$\times\,\frac{1}{\Omega}\sum_{\boldsymbol{\lambda}} K_{4\lambda}(\bar{\mathbf{z}}|\bar{\mathbf{y}})\left[\exp -i\boldsymbol{\lambda}\cdot\left(\bar{\mathbf{x}}_3 + \frac{\bar{\mathbf{z}}}{2}\right)\right]\tilde{N}_\lambda(\bar{\mathbf{y}}) + (1 \leftrightarrow 2) \tag{93}$$

Each of these terms has a singlet contribution which can be identified by the decomposition

$$\begin{aligned}\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) &= \tilde{P}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) + \rho_0\tilde{f}^B(\mathbf{p}_1|\mathbf{x}_1)\\ \tilde{N}_\lambda{}^C(\mathbf{y}) &= 2\rho_0\tilde{f}^C(\boldsymbol{\lambda}/2) + \tilde{P}_\lambda{}^B(\mathbf{y})\end{aligned} \tag{94}$$

where

$$\tilde{f}^C(\boldsymbol{\lambda}/2) = \int [\exp\,(i\boldsymbol{\lambda}\cdot\mathbf{x}/2)]\tilde{f}(\mathbf{p}_1|\mathbf{x})\,d\mathbf{p}_1\,d\mathbf{x} \tag{95}$$

Then the analog of (88) for the general inhomogeneous problem can now be written explicitly, but we shall not do so.

We now comment on the singlet equation. From our derivation of the two-body additive approximation the first equation of the BBGKY hierarchy holds exactly. Thus we have

$$\frac{\partial \tilde{f}(1)}{\partial t} + \frac{\mathbf{p}_1}{m}\frac{\partial}{\partial \mathbf{x}_1}\tilde{f}(1) = \frac{\partial V(\mathbf{x}_1 - \bar{\mathbf{x}}_2)}{\partial \mathbf{x}_2}\frac{\partial}{\partial \mathbf{p}_1}\rho_R(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_2) \tag{96}$$

This involves the bare potential. On the other hand, integration of (73) over $\mathbf{x}_2$ and $\mathbf{p}_2$ yields

$$\frac{\partial \tilde{f}(1)}{\partial t} + \frac{\mathbf{p}_1}{m}\frac{\partial \tilde{f}(1)}{\partial \mathbf{x}} + \frac{\boldsymbol{\Lambda}_1(\mathbf{x}_1\bar{\mathbf{x}}_2)}{\theta}\frac{\partial}{\partial \mathbf{p}_1}\rho_R(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_2) = 0 \tag{97}$$

where

$$\boldsymbol{\Lambda}_1(\mathbf{x}_1\mathbf{x}_2) = \frac{\partial \log \rho_2(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial \mathbf{x}_1}\,K_3(\bar{\mathbf{x}}_3 - \mathbf{x}_1, \mathbf{x}_2 - \mathbf{x}_1) \tag{98}$$

The connection is given by the equilibrium equation

$$\frac{1}{\theta}\frac{\partial \rho_2(\mathbf{x}_1\mathbf{x}_2)}{\partial \mathbf{x}_1} = -\rho_2(\mathbf{x}_1\mathbf{x}_2)\frac{\partial V(\mathbf{x}_1\mathbf{x}_2)}{\partial \mathbf{x}_1} + \rho_3(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_3)\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial \mathbf{x}_1} \tag{99}$$

together with the integral equation for K_3. One easily finds

$$\Lambda_1(\mathbf{x}_1\mathbf{x}_2) = -\theta\,\partial V(\mathbf{x}_1\mathbf{x}_2)/\partial\mathbf{x}_1 \tag{100}$$

an exact property of the inversion kernel K_3.

5. CONVENTIONAL TRUNCATION THEORIES

We will now compare our equations with those of other theories which aim to deal with the Coulomb force problem and with particles interacting via strong short-range forces. The theories in question are at the level of the two-particle distribution function. For the general nonlinear case the second equation of the time-dependent hierarchy is

$$\left\{\frac{\partial}{\partial t} + S_0(12)\right\}N(12) = \left\{\frac{\partial V(1\bar{3})}{\partial \mathbf{x}_1}\cdot\frac{\partial}{\partial \mathbf{p}_1} + \frac{\partial V(2\bar{3})}{\partial \mathbf{x}_2}\cdot\frac{\partial}{\partial \mathbf{p}_2}\right\}N(12\bar{3})$$
$$S_0(12) = \frac{\mathbf{p}_1}{m}\cdot\frac{\partial}{\partial \mathbf{x}_1} + \frac{\partial V(\mathbf{x}_1\mathbf{x}_2)}{\partial \mathbf{x}_1}\frac{\partial}{\partial \mathbf{p}_1} + (1 \leftrightarrow 2) \tag{101}$$

This equation is converted into a self-determined equation when a functional form (holding at all times) for the dependence of $N(123)$ on the lower-order distributions is assumed.

It is useful to introduce cumulant distributions at this point. The standard definitions in the $N \to \infty$ limit are

$$N(1) = f(1), \qquad N(12) = P(12) + f(1)f(2) \tag{102}$$
$$N(123) = T(123) + P(12)f(3) + P(23)f(1) + P(31)f(2) + f(1)f(2)f(3)$$

The simplest example of a useful truncation is the one on which much of plasma physics is based. That is, $T(123) = 0$. This implies a nonlinear truncation,

$$N(123) = N(12)f(3) + N(23)f(1) + N(31)f(2) - 2f(1)f(2)f(3)$$

This is clearly a normalization-preserving truncation. We will refer to this as the time-dependent cumulant approximation. The equation for the development of $N(12)$ is, with this approximation, nonlinear but closed. To make comparison with our equations more direct, we can write the distributions as a sum of an equilibrium part and a deviation from equilibrium, e.g.,

$$N(12) = \phi(p_1)\phi(p_2)\rho_2(12) + \tilde{N}(12); \qquad \tilde{N}(12) = \tilde{N}(\mathbf{x}, \mathbf{p}_1, \mathbf{p}_2) \tag{103}$$

The equilibrium pair distribution is determined by the equation

$$\theta^{-1}\frac{\partial \rho_2(\mathbf{x})}{\partial \mathbf{x}} - \frac{\partial V}{\partial \mathbf{x}}\rho_2(\mathbf{x}) = -\rho_0\frac{\partial}{\partial \mathbf{x}}\int V(|\mathbf{x} - \mathbf{y}|)\rho_2(\mathbf{y})\,d\mathbf{y} \tag{104}$$

To first order in the deviations from equilibrium we have

$$\left[\frac{\partial}{\partial t} + S_0(12)\right]\tilde{N}(12)$$
$$= \frac{\partial\phi_1}{\partial\mathbf{p}_1}\cdot\phi_2\rho_2(\mathbf{x}_1\mathbf{x}_2)\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial\mathbf{x}_1}\tilde{\rho}(\bar{\mathbf{x}}_1)$$
$$- \phi_2\frac{\partial\tilde{f}(1)}{\partial\mathbf{p}_1}\cdot\frac{\partial V(\bar{\mathbf{y}} + \mathbf{x}_2 - \mathbf{x}_1)}{\partial\mathbf{y}}\rho_2(\bar{\mathbf{y}}) + \rho_0\frac{\partial\phi_1}{\partial\mathbf{p}_1}\cdot\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial\mathbf{x}_1}\tilde{\rho}_R(\mathbf{p}_2|\mathbf{x}_2\bar{\mathbf{x}}_3)$$
$$+ \rho_0\phi_2\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial\mathbf{x}_1}\cdot\frac{\partial}{\partial\mathbf{p}_1}\tilde{\rho}_R(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_3) + (1\leftrightarrow 2) \tag{105}$$

Where we have defined $\tilde{\rho}(\mathbf{x}_3) = \int\tilde{N}(3)\,d\mathbf{p}_3$, and $\tilde{\rho}_R(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_3)$ is defined by (13). One of the most obvious differences between this equation and (72) is that the streaming term $S_0(12)$ in (105) involves the bare interparticle potential $V(12)$, whereas the theory of this paper involves a two-body encounter with an effective potential $-\theta^{-1}\log\rho_2(\mathbf{x})$, where $\rho_2(\mathbf{x})$ is the exact pair correlation function. We find the equation for the doublet cumulant

$$\left[\frac{\partial}{\partial t} + S_0(12)\right]\tilde{P}(12) = \frac{\partial\phi_1}{\partial\mathbf{p}_1}\cdot\phi_2[\rho_2(\mathbf{x}_1\mathbf{x}_2) + \rho_0{}^2]\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial\mathbf{x}_1}\tilde{\rho}(\mathbf{x}_3)$$
$$+ \rho_0\frac{\partial V(\mathbf{x}_1\mathbf{x}_2)}{\partial\mathbf{x}_2}\cdot\frac{\partial\phi_2}{\partial\mathbf{p}_2}\tilde{f}(\mathbf{p}_1|\mathbf{x}_1) - \phi_2\frac{\partial\tilde{f}(\mathbf{p}_1|\mathbf{x}_1)}{\partial\mathbf{p}_1}$$
$$\times\left[\frac{\partial V(\bar{\mathbf{y}} - \mathbf{x}_2 - \mathbf{x}_1)}{\partial\mathbf{y}}\rho_2(\bar{\mathbf{y}}) - \rho_0\frac{\partial V(\mathbf{x}_1\mathbf{x}_2)}{\partial\mathbf{x}_1}\right]$$
$$+ \rho_0\frac{\partial\phi_1}{\partial\mathbf{p}_1}\frac{\partial V(\mathbf{x}_1\bar{\mathbf{x}}_3)}{\partial\mathbf{x}_1}\tilde{P}(2\bar{3}) + (1\leftrightarrow 2) \tag{106}$$

Equation (106) cannot be solved for arbitrary initial conditions. For this one would at least need a detailed knowledge of the Green's function for the operator $\{(\partial/\partial t) + S_0(12)\}$; that is, for the interacting two-particle system. For this reason additional approximations are made in the standard theories. In the case of long-range Coulomb forces the term $[\partial V(12)/\partial\mathbf{x}_1]\times(\partial/\partial\mathbf{p}_1)\tilde{P}(12) + (1\leftrightarrow 2)$ is generally neglected since it is assumed to be of higher order in the expansion parameter $e^2\theta$ than the other terms. This is true except at small interparticle distances. Then one only needs the noninteracting two-particle Green's function. The solution of the resulting equation for a system at equilibrium is the Debye–Hückel pair distribution. For the time-dependent case a Fourier–Laplace transform yields a singular integral equation in the momentum variables which is solvable by Hilbert transform techniques.[8]

The crucial point is that the right-hand side of Eq. (106) takes on a convolution form in the spatial variables when one makes the cumulant approximation. Thus after a spatial Fourier transform the distinct wave vectors are uncoupled. Additional assumptions for the time scales for the relaxation of $\tilde{P}(12)$ and $f(1)$ (adiabatic switching assumptions) yield a closed singlet equation known as the Lenard–Balescu equation. This is a step that does not concern us directly in this paper. We are interested in the structure of the doublet equation. The neglect of the direct two-body force in the streaming term leads to divergences at small distances of the order of $e^2\theta$. The Debye–Hückel distribution is also negative in this region.

For the case of the low-density gas of particles interacting via short-range forces one asserts that the right-hand side of (106) is of higher order in density than the rest of the equation, and can be neglected in the first approximation. Then one does not need the full interacting two-particle Green's function, but only its asymptotic behavior, which can be expressed in terms of the scattering cross section. For a system in equilibrium we obtain the static pair distribution:

$$c_2(r) = -\rho_0{}^2(1 - e^{-\theta v(r)}) \tag{107}$$

The nonequilibrium case is treated by neglecting all integral (medium) terms in the integral equation obeyed by the time-dependent cumulant. The The singlet terms in this equation are treated as inhomogeneous terms. When the solution of the cumulant equation is inserted in the singlet equation, one obtains a non-Markovian singlet kinetic equation. Under suitable circumstances this is equivalent to the Boltzmann–Enskog equation.[6] However, it would be a mistake to try to improve the theory by solving the equation for the doublet including the integral terms. The cumulant approximation is deficient and does not treat properly the strong short-range forces between a member of a given pair and a particle from the medium. Thus we would expect divergences to appear in higher orders in the density.

We have shown that both these kinetic equations can be said to arise from further approximations which result from the same cumulant truncation of the hierarchy. We now consider another method of truncation of the nonequilibrium hierarchy.

In the study of the equilibrium properties of dense gases and liquids use has often been made of the Kirkwood superposition approximation (KSA)[9]

$$\langle \hat{N}(123)\rangle \simeq \langle \hat{N}(12)\rangle\langle \hat{N}(23)\rangle\langle \hat{N}(31)\rangle/\langle \hat{N}(1)\rangle\langle \hat{N}(2)\rangle\langle \hat{N}(3)\rangle \tag{108}$$

This is motivated by the fact that for strong short-range forces the triplet distribution must vanish when any two particles are close to each other. The KSA is a minimal means of taking this into account. It is clear, for

example, that the equilibrium cumulant $c_3(123)$ cannot vanish, and it is only in a low-density treatment that the error of neglecting it causes no trouble. Since the KSA has correct asymptotic behavior at both large and small distances and agrees with the time-dependent cumulant approximation to lowest meaningful order, it is a good candidate for a unified theory of fluids. We now sketch such a theory based on the time-dependent KSA not so much because we are interested in it per se, but rather to demonstrate that the structure and degree of complexity of the resulting doublet kinetic equation show marked similarities to those of the doublet equation derived in Section 4. Approximations similar[3] to the KSA presented here have been considered by other authors.[13,14] In particular, Stillinger and Suplinskas use a Kirkwood truncation as part of a program for practical calculations of the self-diffusion coefficient.

We assume the truncation

$$N(123) \cong N(12)N(23)N(31)/N(1)N(2)N(3) \tag{109}$$

and again put $N(12) = \rho_2(\mathbf{x}_1\mathbf{x}_2)\phi(\mathbf{p}_1)\phi(\mathbf{p}_2) + \tilde{N}(12)$, and develop to the first power of time-dependent quantities. For the time-independent part we obtain the Kirkwood integral equation

$$\theta^{-1}\frac{R_2(\mathbf{x})}{\partial \mathbf{x}} + R_2(\mathbf{x})\frac{\partial V(\mathbf{x})}{\partial \mathbf{x}} + \frac{R_2(\mathbf{x})}{\rho_0{}^3}\frac{\partial V(\bar{\mathbf{y}})}{\partial \mathbf{y}} R_2(\bar{\mathbf{y}} - \mathbf{x})R_2(\bar{\mathbf{y}}) = 0 \tag{110}$$

which relates $R_2(\mathbf{x})$ to the interparticle potential. This equation has sensible solutions both for systems with Coulomb forces and for systems with strong short-range forces at low and moderate densities. For the time-dependent part the truncation is

$$\begin{aligned}\tilde{N}(123) = {} & \{\tilde{N}(12)\rho_2(\mathbf{x}_1\mathbf{x}_2)\rho_2(\mathbf{x}_2\mathbf{x}_3)\phi_3\rho_0^{-3} \\ & - \tilde{N}(1)\phi_2\phi_3\rho_0^{-4}\rho_2(\mathbf{x}_1\mathbf{x}_2)\rho_2(\mathbf{x}_2\mathbf{x}_3)\rho_2(\mathbf{x}_1\mathbf{x}_3)\} \\ & + \{\text{cyclic permutations}\}\end{aligned} \tag{111}$$

We now insert this into the second equation of the hierarchy and for the purpose of comparison with the results of Section 4 we consider the homogeneous problem. (Generalization to the inhomogeneous case is also possible but it does not yield much additional insight.) To facilitate this comparison,

[3] The only difference between our KSA and that of Stillinger and Suplinskas[13] and of Mortimer[14] is that in the latter one ostensibly has the freedom to use the exact, or at least more exact, values of the equilibrium correlation functions, rather than the ones predicted by the KSA on the equilibrium BBGKY hierarchy. However, we strongly suspect this freedom is illusory, because the KSA is not consistently applied to the "steady-state" (i.e., equilibrium) solution as well as to that equation for the deviations from equilibrium, so that secular divergences may appear.

we cast the resulting equation in the form of (88):

$$\left\{\frac{\partial}{\partial t}+\frac{\mathbf{p}_2-\mathbf{p}_1}{m}\frac{\partial}{\partial \mathbf{x}}-\left[\frac{\partial V(\mathbf{x})}{\partial \mathbf{x}}-\rho_0\mathbf{J}(\mathbf{x})\right]\cdot\left[\frac{\partial}{\partial \mathbf{p}_2}-\frac{\partial}{\partial \mathbf{p}_1}\right]\right\}\tilde{N}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x})$$
$$+\left\{\frac{\rho_0}{\theta}\frac{\partial\phi_2}{\partial \mathbf{p}_2}\mathbf{H}_s^K(\mathbf{x})\tilde{f}(\mathbf{p}_1)-\frac{\rho_0}{\theta}\phi_2\mathbf{G}_s^K(\mathbf{x})\frac{\partial\tilde{f}(\mathbf{p}_1)}{\partial \mathbf{p}_1}\right.$$
$$\left.+\frac{1}{\theta}\frac{\partial\phi_2}{\partial \mathbf{p}_2}\mathbf{H}^K(\mathbf{x}|\bar{\mathbf{z}})\tilde{P}_R^B(\mathbf{p}_1|\bar{\mathbf{z}})-\frac{1}{\theta}\phi_2\mathbf{G}^K(\mathbf{x}|\bar{\mathbf{z}})\frac{\partial\tilde{P}_R^B(\mathbf{p}_1|\bar{\mathbf{z}})}{\partial \mathbf{p}_1}\right\}+\{1\leftrightarrow 2\}$$
$$+\frac{\mathbf{p}_1-\mathbf{p}_2}{m}\phi_1\phi_2\mathbf{F}^K(\mathbf{x}|\bar{\mathbf{z}})\tilde{N}^C(\bar{\mathbf{z}})=0 \tag{112}$$

where we have introduced

$$\mathbf{J}(\mathbf{x})=-[\partial V(\mathbf{y})/\partial \mathbf{y}]R_2(\bar{\mathbf{y}}-\mathbf{x})R_2(\bar{\mathbf{y}})\rho_0^{-4} \tag{113}$$

$$\mathbf{H}^K(\mathbf{x}|\mathbf{z})=-\theta R_2(\mathbf{x})R_2(\mathbf{x}+\mathbf{z})[\partial V(\mathbf{x}+\mathbf{z})/\partial \mathbf{z}]\rho_0^{-4} \tag{114}$$

$$\mathbf{G}^K(\mathbf{x}|\mathbf{z})=-\theta R_2(\mathbf{x})R_2(\mathbf{x}-\mathbf{z})[\partial V(\mathbf{z})/\partial \mathbf{z}]\rho_0^{-4} \tag{115}$$

$$\mathbf{F}^K(\mathbf{x}|\mathbf{z})=\mathbf{G}^K(\mathbf{x}|\mathbf{z})+\mathbf{H}^K(\mathbf{x}|\mathbf{z}) \tag{116}$$

$$\mathbf{H}_s^K(\mathbf{x})=\mathbf{G}_s^K(\mathbf{x})=(\theta/\rho_0)R_2(\mathbf{x})c_2(\bar{\mathbf{y}})c_2(\bar{\mathbf{y}}-\mathbf{x})\,\partial V(\bar{\mathbf{y}})/\partial \mathbf{y} \tag{117}$$

We first note that since we assume the superposition approximation on the equilibrium functions, (110) implies

$$[\partial V(\mathbf{x})/\partial \mathbf{x}]-\rho_0\mathbf{J}(\mathbf{x})=-(1/\theta)\,\partial \log R_2(\mathbf{x})/\partial \mathbf{x} \tag{118}$$

Thus, in contrast to the time-dependent cumulant approximation, there is an effective two-body collision term, distinct from the bare potential, and in fact it is the same effective potential as that which appears in the theory of this paper. However, the $R_2(\mathbf{x})$ that appears in the KSA theory is presumed to satisfy (110), whereas the two-body additive approximation imposes no such restriction.

The time-dependent cumulant approximation is obtained from the KSA by substituting $c_2(\mathbf{y})\equiv\rho_0^{-2}R_2(\mathbf{y})-1=0$ in Eqs. (113)–(117). The fact that the resulting $H^C(\mathbf{x}|\mathbf{z})$ is the function only of $(\mathbf{x}+\mathbf{z})$ and that $G^C(\mathbf{x}|\mathbf{z})$ is a function only of $\mathbf{z}$ is the property which gives the spatial convolution character to the RHS of (106). This property is lost in both the KSA and the two-body additive approximation. Hence, even if we assumed knowledge of the two-body interacting Green's function, we would still have a nontrivial integral equation in the spatial variables.

The similarity between the KSA and the two-body additive approximation is more than structural. It is shown in Appendix B that, at least for weakly interacting systems, part of the two-body additive truncation (i.e., those terms that do not involve K_4) agrees approximately with the KSA

result. That is to say we can expect the quantities $G(\mathbf{x}|\mathbf{z})$ and $H(\mathbf{x}|\mathbf{z})$ to be well approximated by the Kirkwood expressions (114) and (114) under these circumstances. However, it is one of the characteristic differences of the two-body additive approximation from conventional theories that the C momentum sector is treated distinctly from the B sector. The consequence is that an equation like (116) does not hold for our theory even for weakly interacting systems.

Another clear difference of the two-body additive theory from other truncation schemes is that it implies a nonlocal truncation. One of the ways that this nonlocality is important is that it makes the truncation exact at $t = 0$ for an important class of initial value problems. The main emphasis of treatments of the time-dependent hierarchy has been the derivation of singlet kinetic equations. However, they can be used to calculate time-dependent correlation functions such as the density autocorrelation function, which is closely related to the inelastic scattering function $S(\mathbf{k}, \omega)$. In that case the microscopic initial condition is

$$F_N(t = 0) = \sum_{\alpha} \exp -i\mathbf{k}\cdot\mathbf{q}_\alpha \tag{119}$$

The initial conditions for the reduced distribution functions are then

$$\tilde{N}(\mathbf{p}_1\mathbf{x}_1|t = 0) = \{\rho_0 + \bar{\rho}_2(\mathbf{k})\}(\exp -i\mathbf{k}\cdot\mathbf{x}_1)\phi(\mathbf{p}_1) \tag{120a}$$

$$\begin{aligned}\tilde{N}(\mathbf{p}_1\mathbf{p}_2\mathbf{x}_1\mathbf{x}_2|t = 0) = {} & \phi(\mathbf{p}_1)\phi(\mathbf{p}_2)\{\rho_3(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_3)(\exp -i\mathbf{k}\cdot\bar{\mathbf{x}}_3) \\ & + \rho_2(\mathbf{x}_1\mathbf{x}_2)[\exp -i\mathbf{k}\cdot\mathbf{x}_1) + \exp -i\mathbf{k}\cdot\mathbf{x}_2]\}\end{aligned} \tag{120b}$$

$$\begin{aligned}\tilde{N}(\mathbf{p}_1\mathbf{p}_2\mathbf{p}_3\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3|t = 0) = {} & \phi(\mathbf{p}_1)\phi(\mathbf{p}_2)\phi(\mathbf{p}_3)\{\rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\bar{\mathbf{x}}_4)(\exp -i\mathbf{k}\cdot\bar{\mathbf{x}}_4) \\ & + \rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ & \times [(\exp -i\mathbf{k}\cdot\mathbf{x}_1) + (\exp -i\mathbf{k}\cdot\mathbf{x}_2) + \exp -i\mathbf{k}\cdot\mathbf{x}_3]\}\end{aligned} \tag{120c}$$

If one wishes to calculate the density autocorrelation function, one must solve the doublet kinetic equation with the initial condition (120b), incorporating the exact values of ρ_2 and ρ_3. However, both the time-dependent cumulant approximation and the KSA violate the initial condition for F_N. The connection assumed between the triplet and doublet distributions is not the same as that between (120c) and (120b). This failure at $t = 0$ is an unavoidable property of any spatially local truncation, including those of Stillinger and Suplinskas and of Mortimer. How serious this violation is depends on the particular situation, i.e., on whether further approximations to the equilibrium correlation functions are adequate. On the contrary, in the type of theory that we have presented, the initial condition on F_N is exactly satisfied. Hence the relation between all reduced distribution functions is exact at $t = 0$. One consequence of this is that the fourth frequency moment

of $S(\mathbf{k}, \omega)$ predicted by the two-body additive theory is exact, as shown in I. The same analysis applied to cumulant and superposition approximations shows that they give an incorrect fourth moment.

APPENDIX A: GAUGE TRANSFORMATIONS

In this appendix we discuss the manner in which an explicit one-body function $\Psi(1)$ can be isolated. In I, the two-body additive approximation was defined by

$$F_N = \{\hat{N}(\bar{1}) - \langle N(\bar{1})\rangle\}\psi(\bar{1}) + \tfrac{1}{2}\{\hat{N}(\bar{1}\bar{2}) - \langle N(12)\rangle\}\Psi(\bar{1}\bar{2})$$

which should be compared with (2). From this we obtain

$$\begin{aligned} \tilde{f}(1) &= M_{11}(1\bar{2})\psi(\bar{2}) + M_{12}(1\bar{2}\bar{3})\Psi(\bar{2}\bar{3}) \\ \tilde{N}(12) &= M_{21}(12\bar{3})\psi(\bar{3}) + M_{22}(12\bar{3}\bar{4})\Psi(\bar{3}\bar{4}) \end{aligned} \tag{A.1}$$

where

$$\begin{aligned} M_{22}(1234) &= \tfrac{1}{2}\{\langle N(12)N(34)\rangle - \langle N(12)\rangle\langle N(34)\rangle\} \\ M_{21}(123) &= \langle N(12)N(3)\rangle - \langle N(1)\rangle\langle N(23)\rangle \\ M_{12}(123) &= \tfrac{1}{2}\{\langle N(1)N(23)\rangle - \langle N(1)\rangle\langle N(23)\rangle, \\ M_{11}(12) &= \langle N(1)N(2)\rangle - \langle N(1)\rangle\langle N(2)\rangle \end{aligned} \tag{A.2}$$

It is clear that there is some redundancy in the description of F_N by $\psi(1)$ and $\Psi(12)$. Indeed, we can make a "gauge transformation" to a different $\psi'(1)$ and $\Psi'(12)$ without affecting the value of F_N. The most general such transformation is

$$\begin{aligned} \Psi(12) &= \Psi'(12) + A(1) + A(2) + C \\ \psi(1) &= \psi'(1) - [A(1)/(N-1)] + D \end{aligned} \tag{A.3}$$

where $A(1) = A(\mathbf{x}_1, \mathbf{p}_1)$ is any one-body function, and C and D are constants. It is not possible to invert Eq. (A.1) uniquely without specifying a gauge—that is, a relation between $\psi(1)$ and $\Psi(1\bar{2})$. The gauge chosen for the bulk of this paper is $\psi(1) = 0$. Since there are two constants in (A.3), we can, in addition, use one of them to impose the condition $\chi(\bar{1}\bar{2}) = 0$. In the notation of Section 3 this is

$$\int \chi^C(\mathbf{x}_1\mathbf{x}_2)\, d\mathbf{x}_1\, d\mathbf{x}_2 = 0 \tag{A.4}$$

However, another gauge is more suitable for explicit comparison to the one-body theory. This gauge is specified by the property

$$M_{12}(1\bar{2}\bar{3})\Psi'(\bar{2}\bar{3}) = 0 \tag{A.5}$$

That an $A(1)$ can be found ensuring this property is easily shown. We define

$$J(1) = M_{12}(1\bar{2}\bar{3})\Psi(\bar{2}\bar{3})$$

Then the requirement is simply

$$J(1)/(N-1) = M_{11}(1\bar{3})A(\bar{3}) \tag{A.6}$$

This equation is structurally identical to (31) and the solution, again, is given by the one-body inversion exhibited in paper I. A solution always exists, so we can always find a (Ψ', ψ') satisfying property (A.5).

APPENDIX B: THE STRUCTURE OF THE TRIPLET

In this appendix we exhibit the truncation for $\tilde{N}(123)$ implicit in the two-body approximation by means of the solving kernels K_3 and K_4, and we compare the KSA truncation.

The definition (1) gives for the triplet

$$\tilde{N}(123) = \tfrac{1}{2}\{\langle N(123)N(\bar{4}\bar{5})\rangle - \langle N(123)\rangle\langle N(\bar{4}\bar{5})\rangle\}\Psi(\bar{4}\bar{5}) \tag{B.1}$$

which can be written in terms of reduced equilibrium distribution functions in the form

$$\begin{aligned}\tilde{N}(123) = {} & \langle N(123)\rangle\left\{\frac{\chi(12)}{\langle N(12)\rangle} + \frac{\chi(13)}{\langle N(13)\rangle} + \frac{\chi(23)}{\langle N(23)\rangle}\right\} \\ & + \langle N(123\bar{4})\rangle\left\{\frac{\chi(1\bar{4})}{\langle N(1\bar{4})\rangle} + \frac{\chi(2\bar{4})}{\langle N(2\bar{4})\rangle} + \frac{\chi(3\bar{4})}{\langle N(3\bar{4})\rangle}\right\} \\ & + \frac{1}{2}\left\{\frac{\langle N(123\bar{4}\bar{5})\rangle}{\langle N(\bar{4}\bar{5})\rangle} - \langle N(123)\rangle\right\}\chi(\bar{4}\bar{5})\end{aligned} \tag{B.2}$$

We now divide $N(123)$ into its projections onto different momentum sectors in the manner of Section 2. We define

$$\begin{aligned}\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) &= \int d\mathbf{p}_1\, d\mathbf{p}_2\, d\mathbf{p}_3\, \tilde{N}(123) \\ \tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) &= \int d\mathbf{p}_2\, d\mathbf{p}_3\, \tilde{N}(123) - \phi_1\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ \tilde{N}^{010}(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) &= \int d\mathbf{p}_1\, d\mathbf{p}_3\, \tilde{N}(123) - \phi_2\tilde{N}^{100}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ \tilde{N}^{110}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) &= \int d\mathbf{p}_3\, \tilde{N}(123) - \phi_2\tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ &\quad - \phi_1\tilde{N}^{010}(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) + \phi_1\phi_2\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ \tilde{N}^{111}(\mathbf{p}_1\mathbf{p}_2\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) &= \tilde{N}(123) - \phi_3\tilde{N}^{110}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ &\quad - \phi_2\tilde{N}^{101}(\mathbf{p}_1\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) - \phi_3\tilde{N}^{011}(\mathbf{p}_2\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ &\quad + \phi_1\phi_3\tilde{N}^{010}(\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) + \phi_2\phi_1\tilde{N}^{001}(\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) \\ &\quad - \phi_1\phi_2\phi_3\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) + \phi_2\phi_3\tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\end{aligned} \tag{B.3}$$

We use the formal solution of the inversion problem introduced in (16),

$$\chi^A(12) = \tilde{N}^A(12), \qquad \chi_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) = K_3(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_3)\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_3) \tag{B.4}$$
$$\chi^C(\mathbf{x}_1\mathbf{x}_2) = K_4(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_3\bar{\mathbf{x}}_4)\tilde{N}^C(\bar{\mathbf{x}}_3\bar{\mathbf{x}}_4)$$

to obtain the explicit form of the triplet:

$$\tilde{N}^{111}(\mathbf{p}_1\mathbf{p}_2\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = 0 \tag{B.5a}$$

$$\tilde{N}^{110}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = \rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\tilde{N}^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) \tag{B.5b}$$

$$\begin{aligned}\tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = {} & \left\{\frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_3(\mathbf{x}_1\mathbf{x}_2)} K_3(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_4)\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_4)\right. \\ & \left. + \text{[c.p.(123)]}\right\} \\ & + \rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\bar{\mathbf{x}}_4)K_3(\mathbf{x}_1\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)\frac{\tilde{N}_B{}^R(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_5)}{\rho_2(\mathbf{x}_1\bar{\mathbf{x}}_5)}\end{aligned} \tag{B.5c}$$

$$\begin{aligned}\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = {} & \left\{\frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)} K_4(\mathbf{x}_1\mathbf{x}_2\bar{\mathbf{x}}_3\bar{\mathbf{x}}_4)\tilde{N}^C(\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)\right. \\ & \left. + \text{[c.p.(123)]}\right\} \\ & + \left\{\frac{\rho_4(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\bar{\mathbf{x}}_4)}{\rho_2(\mathbf{x}_1\bar{\mathbf{x}}_4)} K_4(\mathbf{x}_1\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5\bar{\mathbf{x}}_6)\tilde{N}^C(\bar{\mathbf{x}}_5\bar{\mathbf{x}}_6)\right. \\ & \left. + \text{[c.p.(123)]}\right\} \\ & + \frac{1}{2}\left[\frac{\rho_5(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)}{\rho_2(\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)} - \rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)\right]K_4(\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5\bar{\mathbf{x}}_6\bar{\mathbf{x}}_7) \\ & \times \tilde{N}^C(\bar{\mathbf{x}}_6\bar{\mathbf{x}}_7)\end{aligned} \tag{B.5d}$$

Here, the notation [c.p.(123)] indicates the sum over cyclic permutations of the indices 1, 2, and 3.

On the other hand, the linearized KSA (on both time-dependent and equilibrium triplet) gives the following structure:

$$\tilde{N}^{111}(\mathbf{p}_1\mathbf{p}_2\mathbf{p}_3|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = 0 \tag{B.6a}$$

$$\tilde{N}^{110}(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)}\tilde{N}^A(\mathbf{p}_1\mathbf{p}_2|\mathbf{x}_1\mathbf{x}_2) \tag{B.6b}$$

$$\begin{aligned}\tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = {} & \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)}\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2) \\ & + \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)}\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_3) - \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_0(N-1)}\tilde{N}_R{}^B(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_4)\end{aligned} \tag{B.6c}$$

$$\tilde{N}^{000}(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = \left\{\frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)}\,\tilde{N}^{C}(\mathbf{x}_1\mathbf{x}_2) + [\text{c.p.}(123)]\right\}$$
$$- \left\{\frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_0(N-1)}\,\tilde{N}^{C}(\mathbf{x}_1\bar{\mathbf{x}}_4) + [\text{c.p.}(123)]\right\} \tag{B.6d}$$

The two truncations agree in the first (111) sector by virtue of their both being linear truncations. The agreement in the (110) sector is, however, nontrivial. The only difference between the truncations in this sector is that in (B.6b) we must take the KSA form for $\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)$, whereas in (B.5b) we use the exact $\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)$.

There is in addition an interesting similarity between the two truncations in the (100) sector, though it is not as immediately apparent as in the previous two cases. To make the comparison, we need to approximate the ρ_3 and ρ_4 appearing in the (B.5c). Choosing the KSA for ρ_3 and the generalized KSA of (51) for ρ_4, and making use of the integral equation (27) for K_3, as well as property (30), we find that most of the terms involving K_3 explicitly can be eliminated. In particular, (B.5c) becomes

$$\tilde{N}^{100}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3) = \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_2)}\,\tilde{N}_R{}^{B}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_2)$$
$$+ \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_2(\mathbf{x}_1\mathbf{x}_3)}\,\tilde{N}_R{}^{B}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_3) - \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_0}\,\frac{\tilde{N}_R{}^{B}(\mathbf{x}_1\bar{\mathbf{x}}_5)}{N-1}$$
$$+ \frac{\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)}{\rho_0}\,c_2(\mathbf{x}_2 - \bar{\mathbf{x}}_4)c_2(\mathbf{x}_3 - \bar{\mathbf{x}}_4)$$
$$\times K_3(\mathbf{x}_1\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)\tilde{N}_R{}^{B}(\mathbf{p}_1|\mathbf{x}_1\bar{\mathbf{x}}_5) \tag{B.7}$$

where $c_2(\mathbf{x}_1 - \mathbf{x}_2) = [\rho_0^{-2}\rho_2(\mathbf{x}_1\mathbf{x}_2) - 1]$ is the pair correlation function. The first three terms on the right of (B.7) are the KSA result. The last term can be expected to be small for most systems.

We can investigate more precisely how small this term is by assigning an order of magnitude λ to $c_2(\mathbf{r})$ for all $\mathbf{r}$ outside a small volume v. In addition, we decompose

$$\tilde{N}_R{}^{B}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_5) = \rho_0\tilde{f}^{B}(\mathbf{p}_1|\mathbf{x}_1) + \tilde{P}_R{}^{B}(\mathbf{p}_1|\mathbf{x}_1\mathbf{x}_5) \tag{B.8}$$

and we suppose that $\tilde{P}^{B}$ has the same order of magnitude λ for all $|\mathbf{x}_1 - \mathbf{x}_5|$ outside the volume v. The last term of (B.7) then has two parts: a singlet part and a doublet part. The pair part can clearly be expected to be of order v plus order λ^3. The singlet part is

$$\rho_3(\mathbf{x}_1\mathbf{x}_2\mathbf{x}_3)c_2(\mathbf{x}_2 - \bar{\mathbf{x}}_4)c_2(\mathbf{x}_3 - \bar{\mathbf{x}}_4)K_3(\mathbf{x}_1\bar{\mathbf{x}}_4\bar{\mathbf{x}}_5)\tilde{f}^{B}(\mathbf{p}_1|\mathbf{x}_1)$$

This involves only the part of the inversion specified by $\langle\mathbf{k}|K_3|0\rangle$. We refer, then, to (35) and (42) and observe that $K_3(\mathbf{x}_1\mathbf{x}_4\bar{\mathbf{x}}_5)$ should have the same

general spatial structure as $c_2(\mathbf{x}_4 - \mathbf{x}_1)$. Hence the singlet contribution can also be expected to be of order v plus order λ^3. To summarize, in the context of the generalized superposition approximation on the equilibrium distribution the expressions (B.5c) and (B.6c) agree to third order in the pair correlation function.

Finally, when we turn to the lowest sector (000), we are unable to demonstrate such a clear relationship between the two truncations. It should be noted that the failure of the KSA to match the correct initial conditions for the density autocorrelation function problem also occurs in this sector. It is the nonlocal character of the two-body additive truncation which gives the correct initial conditions, and the KSA cannot reproduce this nonlocal behavior.

REFERENCES

1. E. P. Gross, *Ann. Phys. (N.Y.)* **69**:42 (1972).
2. E. P. Gross, *J. Stat. Phys.* **9**:275 (1973).
3. E. P. Gross, *J. Stat. Phys.* **9**:297 (1973).
4. R. Zwanzig, *Phys. Rev.* **144**:170(1966); M. Nelkin and S. Ranganathan, *Phys. Rev.* **164**:222 (1967); P. J. Ortoleva and M. Nelkin, *Phys. Rev.* **181**:429 (1969).
5. J. L. Lebowitz, J. K. Percus, and J. Sykes, *Phys. Rev.* **188**:487 (1969).
6. N. N. Bogolyubov, in *Studies in Statistical Mechanics*, ed. by J. de Boer and G. E. Uhlenbeck, Interscience, New York (1962), Vol. 1.
7. A. Lenard, *Ann. Phys. (N.Y.)* **3**:390 (1960); R. Balescu, *Phys. Fluids* **3**:52 (1960); R. L. Guernsey, *Phys. Fluids* **7**:1600 (1964).
8. R. L. Guernsey, *Phys. Fluids* **5**:322 (1962).
9. I. Z. Fisher, *Statistical Theory of Liquids*, University of Chicago Press, Chicago (1964).
10. B. J. Berne and D. Forster, *Ann. Rev. Phys. Chem.* **22**:563 (1971).
11. T. Y. Wu, *Kinetic Equations of Gases and Plasmas*, Addison-Wesley, Reading, Massachusetts (1966).
12. S. A. Rice and P. Gray, *Statistical Mechanics of Simple Liquids*, Wiley, New York (1964).
13. F. H. Stillinger and R. J. Suplinskas, *J. Chem. Phys.* **44**:2432 (1966).
14. R. G. Mortimer, *J. Chem. Phys.* **48**:1023 (1968).
15. P. Resibois, in *Irreversibility in the Many Body Problem*, ed. by J. Biel and J. Rae, Plenum Press, New York (1972).