PHYSICAL REVIEW A VOLUME 17, NUMBER 1 JANUARY 1978

Kinetic theory for the coherent scattering function $S(q, \omega)$ of classical liquids

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(Received 6 July 1977)

Within the formalism of Mori and Zwanzig a kinetic equation for the phase-space density of simple classical liquids is derived whose relaxation kernel is obtained as the solution of a second kinetic equation. The latter equation is closed by introducing a phenomenological relaxation rate. The solution of the resulting equations can be reduced to the evaluation of the self-correlation function for a tagged particle, studied earlier by the present authors. The calculated scattering functions $S(q,\omega)$ for liquid argon and rubidium are compared with neutron scattering experiments as well as molecular dynamics calculations. For liquid argon the earlier results of Duderstadt and Akcasu are improved, and it is furthermore shown that the present theory adequately accounts for the persistence of propagating density fluctuations in liquid rubidium.

I. INTRODUCTION

Van Hove's dynamical structure factor $S(q, \omega)$ is the most relevant function characterizing the dynamics of liquids. It has been measured by neutron scattering experiments, for example, for liquid argon[1,2] and for liquid rubidium.[3,4] Extensive molecular dynamics calculations are also available for these liquids.[5,6,7] A remarkable difference between the two liquids has been found in the propagation of density fluctuations. In the case of liquid rubidium the experimental $S(q, \omega)$ shows a well-defined propagating mode for values of the wave vector q up to 1.2 Å^{-1}. For liquid argon a well-defined side-peak ceases to exist for $q \geq 0.3$ Å^{-1}.

Many attempts have been made in the past to achieve a theoretical understanding of $S(q, \omega)$.[8] Here we are concerned with those theories aiming to derive and to solve generalizations of Boltzmann's equation for the phase-space density; the correlation of the phase-space density determines $S(q, \omega)$ after averaging over the momentum distribution for the particles.

The simplest improvement of the dilute-gas theory is obtained by taking care of an effective particle interaction by complementing the Boltzmann equation with a Vlassov term[9]; but this approach cannot account for the spectra observed in liquid argon. The Vlassov equation yields better results if the effective interaction is fitted to the fourth sum rule for $S(q, \omega)$ or if retardation effects are taken into account.[10,11] Along these lines a self-consistent microscopic theory has been proposed by Sjögren and Sjölander.[12]

Systematic improvements in Boltzmann's equation require nonlocal retarded collision kernels[13] reducing to a standard collision integral in the long-wavelength zero-frequency limit. In the high-frequency limit the kernel should reduce to the Fokker-Planck operator supplemented by several mean-field terms, which can be calculated exactly from the static properties of the liquid. Thus Duderstadt and Akcasu[14] have proposed a phenomenological collision kernel interpolating between the limits mentioned. Their theory yielded acceptable results for $S(q, \omega)$ for argon, especially when the coupling to heat fluctuations was incorporated.[15]

In a preceding paper[16] (referred to as I hereafter) a kinetic equation was discussed for the phase-space distribution of a tagged particle. For the integral kernel entering the equation of motion a second kinetic equation was derived with a phenomenological collision-time approximation for its memory kernel. This theory yielded a qualitative improvement of the theory of Akcasu *et al.*[17] for the incoherent scattering function. In this paper, therefore, it is our aim to extend the theory of Duderstadt and Akcasu in the spirit of I in order to obtain improved results for $S(q, \omega)$. We apply our theory to liquid argon and liquid rubidium. In particular, we show that it adequately accounts for the persistence of propagating density fluctuations in rubidium for $q \leq 1.2$ Å^{-1}.

In Sec. II the basic two kinetic equations are derived and in Sec. III the collision-time approximation for the second kinetic equation is formulated. Then, in Sec. IV, the equations are solved making use of the results derived earlier.[16] In the last section, V, the results for the dynamical structure factor for liquid argon and liquid rubidium are compared with experiment.

II. EXACT KINETIC EQUATIONS

We consider a classical system of N identical particles of mass m enclosed in a volume V at temperature T interacting via the pair potential $v(r)$. Let $\vec{r}_n$ and $\vec{p}_n$ ($n = 1, 2, \ldots, N$) denote the position and momentum of the nth particle. The basic quantity we are interested in is the one-particle phase-space density, defined as

$$f_{\vec{p}}(\vec{q})=\frac{1}{[\varphi(\vec{p})]^{1/2}}\sum_{n=1}^{N}\delta(\vec{p}-\vec{p}_n(t))\exp[i\vec{q}\cdot\vec{r}_n(t)]\,. \tag{1}$$

For the sake of mathematical convenience a normalization factor has been included in the definition (1) by means of the Maxwell distribution

$$\varphi(\vec{p})=(2\pi mT)^{-3/2}\exp(-\vec{p}^{\,2}/2mT)\,. \tag{2}$$

Dynamical variables like the density and the current density are given as moments of the phase-space density $f_{\vec{p}}(\vec{q})$:

$$\begin{aligned}\rho(\vec{q})&=\int d^3p[\varphi(\vec{p})]^{1/2}f_{\vec{p}}(\vec{q})=\sum_{n=1}^{N}\exp(i\vec{q}\cdot\vec{r}_n)\\ j^\alpha(\vec{q})&=\int d^3p[\varphi(\vec{p})]^{1/2}\frac{p^\alpha}{m}f_{\vec{p}}(\vec{q})\\ &=\sum_{n=1}^{N}\frac{p_n^\alpha}{m}\exp(i\vec{q}\cdot\vec{r}_n)\,,\quad(\alpha=x,y,z)\,.\end{aligned} \tag{3}$$

The information that can be gathered in neutron scattering experiments is summarized in the Fourier transform of the density correlation,

$$\begin{aligned}S(q,\omega)&=\int dt\,e^{i\omega t}S(q,t)\,,\\ S(q,t)&=\frac{1}{N}\left\langle\sum_{n,m=1}^{N}\exp[-i\vec{q}\cdot\vec{r}_n(t)]\exp[i\vec{q}\cdot\vec{r}_m(0)]\right\rangle.\end{aligned} \tag{4}$$

Here and in the following $|\vec{r}|=r$, $|\vec{q}|=q$, etc. and $\langle\cdots\rangle$ denotes the thermal average.

We prefer to consider first the phase-space correlation function,

$$\begin{aligned}S_{\vec{p}\vec{k}}(q,t)&=\left(\frac{1}{\varphi(\vec{p})\varphi(\vec{k})}\right)^{1/2}\\ &\quad\times\left\langle\sum_{n,m=1}^{N}\delta[\vec{p}-\vec{p}_n(t)]\delta[\vec{k}-\vec{p}_m(0)]\right.\\ &\qquad\left.\times\exp[-i\vec{q}\cdot\vec{r}_n(t)]\exp[i\vec{q}\cdot\vec{r}_m(0)]\right\rangle,\end{aligned} \tag{5}$$

for the same reason as in the case of self-diffusion.[16] Integration of its Fourier transform $S_{\vec{p}\vec{k}}(q,\omega)$ over momentum variables, reproduces the van Hove scattering function $S(q,\omega)$

$$S(q,\omega)=\frac{1}{N}\int d^3p\,d^3k[\varphi(\vec{p})]^{1/2}S_{\vec{p}\vec{k}}(q,\omega)[\varphi(\vec{k})]^{1/2}\,. \tag{6}$$

In this paper we apply the formalism of Zwanzig and Mori[18] to the phase-space correlation function $S_{\vec{p}\vec{k}}(q,\omega)$. The formal framework will be very similar to I. It is convenient to introduce the Laplace transform of the correlation function $S_{\vec{p}\vec{k}}(q,t)$

$$\phi_{\vec{p}\vec{k}}(q,z)=\mp\frac{i}{T}\int dt\,\Theta(\pm t)e^{izt}S_{\vec{p}\vec{k}}(q,t)\,,\quad \operatorname{Im}z\gtrless0\,. \tag{7}$$

This function is analytic for nonreal z and is discontinuous across the real axis,

$$\phi_{\vec{p}\vec{k}}(q,\omega\pm i\epsilon)=\phi'_{\vec{p}\vec{k}}(q,\omega)\pm i\phi''_{\vec{p}\vec{k}}(q,\omega)\,, \tag{8}$$

with $\phi'_{\vec{p}\vec{k}}(q,\omega)$ and $\phi''_{\vec{p}\vec{k}}(q,\omega)$ being real. As $\phi_{\vec{p}\vec{k}}(q,z)$ decreases for large z as $1/z$, the spectral function $\phi''_{\vec{p}\vec{k}}(q,\omega)$ determines $\phi_{\vec{p}\vec{k}}(q,z)$ by a Kramers-Kronig relation,

$$\phi_{\vec{p}\vec{k}}(q,z)=\int\frac{d\omega}{\pi}\frac{\phi''_{\vec{p}\vec{k}}(q,\omega)}{\omega-z}\,. \tag{9}$$

The time evolution of a dynamical variable like the phase-space density is given by the Liouville operator

$$\mathcal{L}f_{\vec{p}}(\vec{q})=-i\partial_t f_{\vec{p}}(\vec{q})\,. \tag{10}$$

To evaluate the right-hand side of (10) we have to use Newton's equations of motion

$$\begin{aligned}&\dot{\vec{r}}_n=\vec{p}_n/m\,,\\ &\dot{\vec{p}}_n=-\frac{\partial U}{\partial\vec{r}_n}\,,\quad U=\frac{1}{2}\sum_{n\neq m}V(\vec{r}_n-\vec{r}_m)\end{aligned} \tag{11}$$

and get

$$\begin{aligned}\mathcal{L}f_{\vec{p}}(\vec{q})&=\frac{\vec{p}\cdot\vec{q}}{m}f_{\vec{p}}(\vec{q})-\frac{i}{[\varphi(\vec{p})]^{1/2}}\frac{\partial}{\partial p^\alpha}\\ &\quad\times\sum_{n=1}^{N}\exp(i\vec{q}\cdot\vec{r}_n)\frac{\partial U}{\partial\vec{r}_n^{\,\alpha}}\delta(\vec{p}-\vec{p}_n)\,.\end{aligned} \tag{12}$$

Introducing a scalar product

$$(f_{\vec{p}}(\vec{q})\,|\,f_{\vec{k}}(\vec{q}))=(1/T)\langle f^*_{\vec{p}}(\vec{q})f_{\vec{k}}(\vec{q})\rangle\,, \tag{13}$$

the Liouville operator turns out to be Hermitian and $\phi_{\vec{p}\vec{k}}(q,z)$ can be written as a resolvent matrix element of $\mathcal{L}$,

$$\phi_{\vec{p}\vec{k}}(q,z)=(f_{\vec{p}}(\vec{q})\,|[1/(z-\mathcal{L})]|\,f_{\vec{k}}(\vec{q}))\,. \tag{14}$$

We define a projector $\mathcal{P}_0$ onto the subspace spanned by the phase-space density $f_{\vec{p}}(\vec{q})$

$$\mathcal{P}_0=\sum_{\vec{p},\vec{k}}|f_{\vec{p}}(\vec{q}))(\chi_0^{-1})_{\vec{p}\vec{k}}(f_{\vec{k}}(\vec{q})|\,. \tag{15}$$

Here $\vec{p}$ and $\vec{k}$ are used as matrix subscripts and $(\chi_0)_{\vec{p}\vec{k}}$ denotes the normalization matrix

$$\begin{aligned}(\chi_0)_{\vec{p}\vec{k}}&=(f_{\vec{p}}(\vec{q})\,|\,f_{\vec{k}}(\vec{q}))\\ &=(N/T)[\delta(\vec{p}-\vec{k})+b_{\vec{p}}b_{\vec{k}}ng(q)]\,,\quad b_{\vec{p}}=[\varphi(\vec{p})]^{1/2}\,,\end{aligned} \tag{16}$$

with the pair correlation

$$ng(q)=\frac{1}{N}\sum_{n\neq m}\langle\exp(i\vec{q}\cdot\vec{r}_{nm})\rangle$$

and the density $n=N/V$. The normalization matrix can be easily inverted[14] to yield

$$(\chi_0^{-1})_{\vec{p}\vec{k}} = (T/N)[\delta(\vec{p}-\vec{k}) - nc(q) b_{\vec{p}} b_{\vec{k}}] , \qquad (17)$$

with the direct correlation function $nc(q) = ng(q)/[1+ng(q)]$. Application of the Zwanzig-Mori formalism[18] leads to the following representation of $\phi_{\vec{p}\vec{k}}(q,z)$ in matrix notation:

$$\phi(q,z) = \chi_0(z\chi_0 - \Omega_0 - M_0)^{-1}\chi_0 . \qquad (18)$$

The restoring force matrix Ω_0 abbreviates to

$$\begin{aligned}(\Omega_0)_{\vec{p}\vec{k}} &= (f_{\vec{p}}(\vec{q})\,|\,\mathcal{L} f_{\vec{k}}(\vec{q}))\\ &= (N/T)(\vec{p}\cdot\vec{q}/m)\delta(\vec{p}-\vec{k}) . \qquad (19)\end{aligned}$$

With $\mathcal{Q}_0 = 1 - \mathcal{P}_0$ the relaxation kernel M_0 is another resolvent matrix element of the reduced Liouville operator $\mathcal{Q}_0\mathcal{L}\mathcal{Q}_0$ taken between the fluctuating forces $\mathcal{Q}_0\mathcal{L} f_{\vec{p}}(\vec{q})$,

$$(M_0)_{\vec{p}\vec{k}} = (\mathcal{Q}_0\mathcal{L} f_{\vec{p}}(\vec{q})\,|\,\frac{1}{z-\mathcal{Q}_0\mathcal{L}\mathcal{Q}_0}\,|\,\mathcal{Q}_0\mathcal{L} f_{\vec{k}}(\vec{q})) . \qquad (20)$$

Equation (20) can be rewritten in the standard form

$$\begin{aligned}&\left(z - \frac{\vec{p}\cdot\vec{q}}{m}\right)\phi_{\vec{p}\vec{p}'}(q,z) + nc(q)\frac{\vec{p}\cdot\vec{q}}{m} b_{\vec{p}}\\ &\quad\times\int d^3k\, b_{\vec{k}}\phi_{\vec{k}\vec{p}'}(q,z) - \frac{N}{T}[\delta(\vec{p}-\vec{p}\,') + ng(q) b_{\vec{p}} b_{\vec{p}'}]\\ &\qquad = -\frac{T}{N}\int d^3k\,(M_0)_{\vec{p}\vec{k}}(q,z)\phi_{\vec{k}\vec{p}'}(q,z) . \qquad (21)\end{aligned}$$

The correlation function at $t=0$,

$$\phi_{\vec{p}\vec{k}}(q, t=0) = (\chi_0)_{\vec{p}\vec{k}} , \qquad (22)$$

is determined by the Fourier transform $g(q)$ of the pair-correlation $g(r)$ [compare Eq. (16)]. The correlation function changes in time, first of all because of the free flow of the particles. This gives rise to a streaming term $\vec{p}\cdot\vec{q}/m$ in the kinetic equation for ϕ, ensuring the correct free-gas limit for the present theory. Interaction effects are taken into account by the memory kernel M_0 and by a mean-field term

$$nc(q)\frac{\vec{p}\cdot\vec{q}}{m} b_{\vec{p}} \int d^3k\, b_{\vec{k}}\phi_{\vec{k}\vec{p}'}(q,z), \qquad (23)$$

whose effective potential is given by the direct correlation function. Neglecting memory effects ($M_0 = 0$) in (21), one gets the well-known Vlassov equation, which was discussed in the literature extensively in connection with the existence of high frequency collective modes.[9] Retardation effects as well as nonlocalities are represented by the z and q dependences of the memory kernel M_0.

For the latter we can derive a second kinetic equation. Let us define a projector $\mathcal{P}_1$ and its orthogonal complement $\mathcal{Q}_1$:

$$\mathcal{P}_1 = \sum_{\vec{p},\vec{k}} |\mathcal{Q}_0\mathcal{L} f_{\vec{p}}(\vec{q}))[\chi_1^{-1}]_{\vec{p}\vec{k}}(\mathcal{Q}_0\mathcal{L} f_{\vec{k}}(\vec{q})| , \qquad (24)$$

$$\mathcal{Q}_1 = 1 - \mathcal{P}_1 ,$$

where χ_1 denotes the normalization matrix of the fluctuating forces,[14]

$$\begin{aligned}(\chi_1)_{\vec{p}\vec{k}} &= (\mathcal{Q}_0\mathcal{L} f_{\vec{p}}(\vec{q})\,|\,\mathcal{Q}_0\mathcal{L} f_{\vec{k}}(\vec{q}))\\ &= Nm\Omega_E^2 F_{\vec{p}\vec{k}} + \frac{N}{T}\left(nc(q)\frac{q^\alpha q^\beta}{m^2} - \frac{\omega_{\alpha\beta}^2(\vec{q})}{Tm}\right)p^\alpha k^\beta b_{\vec{p}} b_{\vec{k}} . \qquad (25)\end{aligned}$$

$F_{\vec{p}\vec{k}}$ is the Fokker-Planck operator

$$F_{\vec{p}\vec{k}} = \frac{1}{b_{\vec{p}} b_{\vec{k}}}\frac{\partial}{\partial p^\alpha}\frac{\partial}{\partial p^\beta}\varphi(\vec{p})\delta(\vec{p}-\vec{k}) , \qquad (26)$$

and Ω_E^2 and $\omega_{\alpha\beta}^2(\vec{q})$ are defined as

$$\begin{aligned}\Omega_E^2 &= \frac{n}{3m}\int d^3r\, g(r)\Delta V(r) ,\\ \omega_{\alpha\beta}^2(\vec{q}) &= \frac{n}{m}\int d^3r\, g(r) e^{-i\vec{q}\cdot\vec{r}}\frac{\partial}{\partial r^\alpha}\frac{\partial}{\partial r^\beta} V(r) . \qquad (27)\end{aligned}$$

In analogy to (18) one gets the following representation for M_0,

$$M_0 = \chi_1(z\chi_1 - \Omega_1 - M_1)^{-1}\chi_1 \qquad (28)$$

with the restoring-force matrix[19]

$$\begin{aligned}(\Omega_1)_{\vec{p}\vec{k}} &= (\mathcal{Q}_0\mathcal{L} f_{\vec{p}}(\vec{q})\,|\,\mathcal{L}\mathcal{Q}_0\mathcal{L} f_{\vec{k}}(\vec{q}))\\ &= Nm\Omega_E^2 P_{\vec{p}\vec{k}}(q) - NmT[q^\gamma\omega_{\alpha\beta}^2(\vec{q}) - q\nu_{\alpha\beta\gamma}^2(\vec{q})]\frac{1}{b_{\vec{p}} b_{\vec{k}}}\\ &\quad\times\left(\frac{\partial}{\partial p^\gamma}\frac{\partial}{\partial p^\beta}\frac{\partial}{\partial k^\alpha} + \frac{\partial}{\partial k^\gamma}\frac{\partial}{\partial k^\beta}\frac{\partial}{\partial p^\alpha}\right)\varphi(\vec{p})\varphi(\vec{k}) . \qquad (29)\end{aligned}$$

P is an operator similar to F:

$$P_{\vec{p}\vec{k}}(q) = \frac{1}{b_{\vec{p}} b_{\vec{k}}}\frac{\partial}{\partial p^\alpha}\frac{\partial}{\partial k^\alpha}\varphi(\vec{p})\frac{\vec{p}\cdot\vec{q}}{m}\delta(\vec{p}-\vec{k}) , \qquad (30)$$

and $\nu_{\alpha\beta\gamma}^2(\vec{q})$ is defined as

$$\nu_{\alpha\beta\gamma}^2(\vec{q}) = iq\frac{n}{m}\int d^3r\, e^{i\vec{q}\cdot\vec{r}} g(r)\frac{\partial}{\partial r^\alpha}\frac{\partial}{\partial r^\beta}\frac{\partial}{\partial r^\gamma} V(r) . \qquad (31)$$

The relaxation kernel M_1 reads

$$\begin{aligned}(M_1)_{\vec{p}\vec{k}}(q,z) &= (\mathcal{Q}_1\mathcal{Q}_0\mathcal{L}\mathcal{Q}_0 f_{\vec{p}}(\vec{q})\,|\,\frac{1}{z-\mathcal{Q}_1\mathcal{Q}_0\mathcal{L}\mathcal{Q}_0\mathcal{Q}_1}\\ &\quad\times|\,\mathcal{Q}_1\mathcal{Q}_0\mathcal{L}\mathcal{Q}_0 f_{\vec{k}}(\vec{q})) . \qquad (32)\end{aligned}$$

In analogy to (21) the kinetic equation for M_0 contains a streaming term PF^{-1}, a relaxation kernel M_1 and several mean-field-type expressions.

Up to now we have derived exact kinetic equations, which are equivalent to the first two steps in Mori's continued-fraction representation for the set of variables $f_{\vec{p}}(\vec{q})$. It should be noted, however, that the formulas (18) and (28) imply the inversion of infinite matrices, i.e., the solution of kinetic equations. Furthermore the kernel $M^{1}_{\vec{p}\vec{k}}(q,z)$ is unknown. The representation of $\phi_{\vec{p}\vec{k}}(q,z)$ in terms of the unknown kernel $M^{1}_{\vec{p}\vec{k}}(q,z)$ is nevertheless advantageous. First of all it guarantees four sum rules for the spectral function $\phi''_{\vec{p}\vec{k}}(q,\omega)$, no matter how the kernel $M^{1}_{\vec{p}\vec{k}}(q,z)$ will be approximated. It should be noted, that a knowledge of the pair correlation $ng(r)$ is sufficient to evaluate the first four moments of the spectral function $\phi''_{\vec{p}\vec{k}}(q,\omega)$.[19] Furthermore the representation of ϕ in terms of M_1 ensures the conservation laws for the number of particles and their momentum. This can be seen as follows: Conservation of particle number requires

$$\int d^3p\, b_{\vec{p}}(M_0)_{\vec{p}\vec{k}}(q,z)=0$$

or

$$\int d^3p\, b_{\vec{p}}(\chi_1)_{\vec{p}\vec{k}}=0\,. \tag{33}$$

Inserting the explicit expression for $(\chi_1)_{\vec{p}\vec{k}}$, we get

$$\frac{1}{b_{\vec{k}}}Nm\Omega_E^2\int d^3p\,\frac{\partial}{\partial p^\alpha}\frac{\partial}{\partial k^\alpha}\,\varphi(\vec{p})\delta(\vec{p}-\vec{k}) + \frac{N}{T}\left(nc(q)\frac{q^\alpha q^\beta}{m^2}-\frac{\omega^2_{\alpha\beta}(\vec{q})}{Tm}\right)\int d^3p\, b_{\vec{p}}\,p^\alpha k^\beta b_{\vec{p}}\, b_{\vec{k}}=0\,. \tag{34}$$

To conserve momentum, we have to require[20]

$$\lim_{q\to 0}\int d^3p\, b_{\vec{p}}\,p^\alpha(M_0)_{\vec{p}\vec{k}}(q,z)=0\,,\quad (\alpha=x,y,z)$$

or (35)

$$\lim_{q\to 0}\int d^3p\, b_{\vec{p}}\,p^\alpha(\chi_1)_{\vec{p}\vec{k}}=0\,.$$

Direct computation yields

$$\lim_{q\to 0}\int d^3p\, b_{\vec{p}}\,p^\alpha(\chi_1)_{\vec{p}\vec{k}} = -\frac{1}{b_{\vec{k}}}Nm\Omega_E^2\int d^3p\,\frac{\partial}{\partial k^\alpha}\varphi(\vec{p})\delta(\vec{p}-\vec{k}) - \frac{N}{T}k^\alpha b_{\vec{k}}\lim_{q\to 0}\omega^2_{\alpha\beta}(\vec{q})\,. \tag{36}$$

Evaluating $\omega^2_{\alpha\beta}(\vec{q})$ in the limit of small q,

$$\lim_{q\to 0}\omega^2_{\alpha\beta}(\vec{q})=\delta_{\alpha\beta}\Omega_E^2\,, \tag{37}$$

shows indeed

$$\lim_{q\to 0}\int d^3p\, b_{\vec{p}}\,p^\alpha(\chi_1)_{\vec{p}\vec{k}}=0\,.$$

Since the energy cannot be represented as a moment of the one-particle phase-space density energy conservation is violated within the present theory; this feature is shared with most of the preceding work.[12,14]

III. COLLISION TIME APPROXIMATION FOR M_1

Within the formalism of Zwanzig and Mori, approximations are formulated by modeling the relaxation kernels like M_0 or M_1. In this paper we want to keep the first exact kinetic equation (21) and calculate M_0 by means of Eq. (28). In the latter we propose a collision time ansatz for the kernel M_1

$$[M_1(q,z)\chi_1^{-1}]_{\vec{p}\vec{k}}=-\nu(q,z)\delta(\vec{p}-\vec{k})\,. \tag{38}$$

Essentially the same approximation has been used in I for the self-part of the correlation function. The damping function $\nu(q,z)$ has the analytical properties of ϕ with $\nu''(q,\omega)\geq 0$. Eqs. (21) and (28) will be solved in the next section for a general $\nu(q,z)$. However, to get useful results, $\nu(q,z)$ has to be specified further. One can get some information from the well-known form of the longitudinal current correlation in the hydrodynamic limit[21]

$$\phi_L(q,z)=(j_L(q)|[1/(z-\mathfrak{L})]|j_L(q)) \simeq\frac{N}{m}\frac{1}{z-q^2C_T^2/z+iq^2D_l-q^2(C_p/C_v-1)C_T^2[z+iq^2(\lambda/mC_v)]^{-1}}\,. \tag{39}$$

$D_l=(1/mn)(\frac{4}{3}\eta+\xi)$ denotes the longitudinal viscosity, η is the shear and ξ the bulk viscosity. C_v (C_p) denotes the specific heat at constant volume (pressure), C_T the isothermal speed of sound, and λ the heat conductivity. Neglecting thermal fluctuations, one cannot reproduce the hydrodynamic result (39) unless complicated frequency dependences of $\nu(q,z)$ with new additional parameters are introduced.[22] Ailawadi *et al.*[21] have tried to estimate, how much thermal fluctuations contribute to the relaxation of longitudinal current correlations. Evaluating their formulas for liquid argon, one concludes: For wave numbers $q>0.5$ Å^{-1}—that are available in neutron scattering experiments—the contribution of thermal fluctuations is of minor importance. We therefore ignore frequency variations and replace $\nu(q=0,z)$ by a constant

$$\nu(q=0,z)=\pm i\beta\,,\quad \operatorname{Im} z \gtreqless 0\,. \tag{40}$$

One way to fix the constant β is the $z \to 0$ limit of the transverse current correlation

$$-\frac{\eta}{mn}=\lim_{\omega\to 0}\lim_{q\to 0}\frac{\omega^2}{q^2}\phi''_T(q,\omega)\frac{m}{N}\,, \tag{41}$$

with $\phi_T(q,z)$ defined as

$$\phi_T(q,z)=(j_T(q)|[1/(z-\mathcal{L})]|j_T(q))\,. \tag{42}$$

Eq. (41) has been used by Duderstadt and Akcasu[14] and by Jhon and Forster.[15] Considering the molecular dynamics data of Levesque *et al.*[6] it seems questionable to replace the relaxation frequency $\nu(q,z)$ by its value at $z=0$. Levesque *et al.* have calculated the Kubo integrand for the shear viscosity, i.e., the correlation of the stress tensor, as a function of time. The Fourier transform $\eta(\omega)$ can be represented as a sum of two Lorentzians. If one wants to replace this function by a constant, the approximation for intermediate frequencies will certainly be improved by using a mean value $\bar{\eta}$ instead of the value at $\omega=0$. We therefore replace $\eta(\omega=0)$ in Eq. (41) by a mean value $\bar{\eta}$, which we choose about one-half of the value at $\omega=0$.

Evaluating the right-hand side of Eq. (41) one has to take care of the 0-eigenvalues of the matrices Ω_1 and χ_1. Forster and Martin[20] showed how to handle that problem by projecting out the subspace of the hydrodynamic modes. Choosing the external wave vector q parallel to the z direction the result is

$$\eta/mn=(1/\beta)[\delta_1/2-v^2(1+\delta_2)]+\beta v^2/2\Omega_E^2\,,\quad v=T/m \tag{43a}$$

with

$$\delta_1=\frac{n}{m}\int d^3r\,g(r)z^2\partial_x^2V(r)\,, \tag{43b}$$

and

$$\delta_2=\lim_{q\to 0}[\omega_{xx}^2(q)-2\nu_{xxx}^2(q)]/\Omega_T^2\,. \tag{43c}$$

The q dependence of the damping function will be chosen exactly as in the case of self-diffusion. Since the fluctuating force $\mathfrak{Q}_0\mathcal{L}f_{\vec{p}}(\vec{q})$ and hence $\mathfrak{Q}_0\mathcal{L}\mathfrak{Q}_0\mathcal{L}f_{\vec{p}}(\vec{q})$ is proportional to the interaction, the memory kernel vanishes for a free gas and it is plausible, that M_1 should approach zero, when q tends to infinity. As in the case of self-diffusion, it is very important to take this feature into account. We propose the same simple ansatz as in I,

$$\nu(q,z)=\nu(q=0,z)/[1+(q/q^*)^2]\,, \tag{44}$$

with the same cutoff

$$q^*=\Omega_E/(4mT)^{1/2}\,.$$

Neglecting the restoring force Ω_1 in Eq. (28), one recovers the theory of Duderstadt and Akcasu.[14] The restoring force leads to a streaming term PF^{-1} in the kinetic equation for M_0. This term, not treated appropriately in the earlier theories,[14,15] increases linearly with q and dominates the collision frequency ν for q exceeding 3 or 4 Å^{-1}.

IV. SOLUTION OF THE KINETIC EQUATIONS

In order to obtain the correlation function $\phi_{\vec{p}\vec{k}}(q,z)$ we have to solve two integral equations

$$\phi=\chi_0\frac{1}{z\chi_0-\Omega_0-M_0}\chi_0\,, \tag{45a}$$

$$M_0=\chi_1\frac{1}{(z+\nu)\chi_1-\Omega_1}\chi_1\,. \tag{45b}$$

The matrices χ_1 and Ω_1 consist of differential operators—the Fokker-Planck operator F and the operator for the restoring force P—and of separable terms. The inversion of the differential operators was accomplished in I and the separable terms are easy to handle, so that the solution of the kinetic equation (45) does not offer new problems. When dealing with the differential operators P and F it turned out to be helpful to use creation and annihilation operators. Exactly as in I we introduce a scalar product,

$$\langle f|g\rangle=\int d^3p\,f^*(\vec{p})g(\vec{p})\,, \tag{46}$$

treating the space of $\vec{p}$-dependent functions as a Hilbert space. The operators

$$a_\alpha=\frac{1}{(4mT)^{1/2}}\left(p^\alpha+2mT\frac{\partial}{\partial p^\alpha}\right),$$

$$a_\alpha^\dagger=\frac{1}{(4mT)^{1/2}}\left(p^\alpha-2mT\frac{\partial}{\partial p^\alpha}\right)\quad(\alpha=x,y,z) \tag{47}$$

are conjugate to each other and obey the canonical commutation relation

$$[a_\alpha,a_\beta^\dagger]=\delta_{\alpha\beta}\,,\quad [a_\alpha,a_\beta]=[a_\alpha^\dagger,a_\beta^\dagger]=0\,.$$

The space can be spanned by an orthonormalized set of functions

$$|n_1n_2n_3\rangle=(a_1^\dagger)^{n_1}(a_2^\dagger)^{n_2}(a_3^\dagger)^{n_3}\left(\frac{1}{n_1!\,n_2!\,n_3!}\right)^{1/2}|\vec{0}\rangle\,,$$

$$n_\alpha=0,1,2\ldots \tag{48}$$

with the vacuum state

$$\langle\vec{p}|\vec{0}\rangle=b_{\vec{p}}\,. \tag{49}$$

Functions such as $\phi_{\vec{p}\vec{k}}$ and $(M_0)_{\vec{p}\vec{k}}$ are considered as matrix operators, using the notation

$$(M_0g)(\vec{p})=\int d^3k\,(M_0)_{\vec{p}\vec{k}}\,g(\vec{k})\,. \tag{50}$$

The density correlation is the vacuum expectation value of

$$\phi(q,z) = (\rho(\vec{q})|[1/(z-\mathcal{L})]|\rho(\vec{q})) = \langle\vec{0}|\phi|\vec{0}\rangle, \tag{51}$$

and the current correlation appears as

$$\phi_{\alpha\alpha}(q,z) = (j_\alpha(\vec{q})|[1/(z-\mathcal{L})]|j_\alpha(\vec{q})) = (T/m)\langle 1_\alpha 0_\beta 0_\gamma|\phi|1_\alpha 0_\beta 0_\gamma\rangle. \tag{52}$$

We formally rewrite Eqs. (45) to get a single equation for ϕ,

$$\{\chi_1^{-1}[(z+\nu)\chi_1 - \Omega_1]\chi_1^{-1}(z-\Omega_0\chi_0^{-1}) - \chi_0^{-1}\}\phi = \chi_1^{-1}[(z+\nu)\chi_1 - \Omega_1]\chi_1^{-1}\chi_0. \tag{53}$$

We furthermore try to reduce the problem to a one-dimensional one by taking matrix elements of Eq. (53). More precisely we calculate $\langle 0_x 0_y|(53)|0_x 0_y 1_z\rangle$ to get an equation for the normalized correlation function Θ,

$$\Theta(q,z) = \langle 0_x 0_y|\phi(T/N)|0_x 0_y 1_z\rangle,$$

which only depends on one variable, p_z. Delegating some simple but rather lengthy calculations to an appendix, we just want to state the resulting equation for Θ:

$$\Theta + T^2 G\tilde{N}\left(\sum_{\nu,\mu=1}^{2} s_{\nu\mu}|\nu\rangle\langle\mu|\phi_0^{-1}\Theta + \sum_{\nu=1}^{2} t_\nu|\nu\rangle\langle 0|\Theta\right) = TG|1\rangle + T^2 G\tilde{N}\sum_{\nu=1}^{2} u_\nu|1_\nu\rangle. \tag{54}$$

Concerning the notation: one can write the Hilbert space as a product space and denote by $|\nu\rangle$ the νth eigenfunction with respect to the only nontrivial component of $\vec{p}$, i.e., p_z. G and $\tilde{N}$ are defined as

$$G = \frac{1}{T}\frac{1}{z - qv(a_z + a_z^\dagger) - T\tilde{N}}, \tag{55a}$$

$$\tilde{N} = \frac{1}{T}\Omega_E^2 a_z^\dagger \frac{1}{z + \nu - qv(a_z + a_z^\dagger)} a_z. \tag{55b}$$

These functions can be identified with the self-part of the correlation function and its memory kernel according to Eqs. 41(a) and 41(b) of I. ϕ_0 denotes the free-gas function

$$\phi_0 = qv/[z - qv(a_z + a_z^\dagger)].$$

The q and z dependence of Θ, ϕ_0, G and $\tilde{N}$ has not been indicated explicitly, the same is true for the q- and z-dependent coefficients

$$t_1 = qv[nc(q)/\Delta^2](z+\nu), \tag{56a}$$

$$\Delta^2 = \Omega_E^2 + q^2v^2 nc(q) - \omega_{zz}^2(q), \tag{56b}$$

$$t_2 = [nc(q)q^2v^2/2\Delta^2\Omega_E^2][\Omega_E^2 + \omega_{zz}^2(q) - \nu_{zzz}^2(q)], \tag{56c}$$

$$s_{11} = qv(z+\nu)(1/\Delta^2 - 1/\Omega_E^2), \quad s_{22} = 0, \tag{57a}$$

$$s_{21} = s_{12} = -(q^2v^2/\sqrt{2}\,\Delta^2\Omega_E^2)[\Omega_E^2 - \Delta^2 + \omega_{zz}^2(q) - \nu_{zzz}^2(q)], \tag{57b}$$

$$u_1 = s_{11}/qv, \tag{58a}$$

$$u_2 = s_{21}/qv. \tag{58b}$$

In the exact equations the restoring-force matrix contains a term that couples the density correlation to a transverse matrix element $\langle 2_x 0_y 0_z|\phi \times |0_x 0_y 1_z\rangle$. This coupling is neglected in deriving Eq. (54) from Eq. (45). We consider this a small error only, since the coupling vanishes in the hydrodynamic as well as in the free-gas limit.

To get a closed system of equations we multiply (54) successively by $\langle 0|$, $\langle 1|\phi_0^{-1}$, and $\langle 2|\phi_0^{-1}$. The resulting equations include several matrix elements of the functions G and $\tilde{N}$. These have been calculated in I for an arbitrary relaxation frequency $\nu(q,z)$. It remains the solution of a 3 × 3 matrix equation.

Let's make a brief comment on Eq. (54) for Θ. Neglecting all projector terms, one recovers the results, we got for the case of self-diffusion.[16] In this approximation all the information about the collective motion of the particles is hidden in the relaxation frequency $\nu(q,z)$. Comparing these frequencies for the correlation function and its self-part, we find that the two quantities are of the same order of magnitude providing some justification for some preceding work.[10,11] The main difference between the motion of a tagged particle and the collective motion of many particles is reflected in the kinetic equation for Θ by the presence of several projector terms, which guarantee the conservation laws and take care of the static correlations.

V. RESULTS

The preceding theory has been used to calculate the van Hove function for argon at $T = 85\,^\circ\mathrm{K}$ and $n = 2.14\times10^{22}\ \mathrm{cm}^{-3}$ (particle mass $m = 66.28\times10^{-24}$ g) and rubidium $T = 319\,^\circ\mathrm{K}$ and $n = 1.058\times10^{22}\ \mathrm{cm}^{-3}$ (particle mass $m = 142\times10^{-24}$ g).

For argon the pair-correlation function of Yarnell *et al.*[23] and a 12-6 potential with $\epsilon = 120\,^\circ\mathrm{K}$, $\sigma = 3.4$ Å have been used to calculate the various static averages like $c(q)$, $\omega_{zz}^2(q)$, $\nu_{zzz}^2(q)$, etc. The characteristic frequency Ω_E is $7.4\times10^{12}\ \mathrm{sec}^{-1}$ (parameter as in I). The phenomenological damping parameter β in Eq. (40) is taken as $4.17\times10^{12}\ \mathrm{sec}^{-1}$. Substituting this number into Eq. (43a) one gets $\bar{\eta}$ about half the hydrodynamic shear viscosity η.

Since the plots of $S(q,\omega)$ are not sensitive enough for a comparison between theory and experiment,

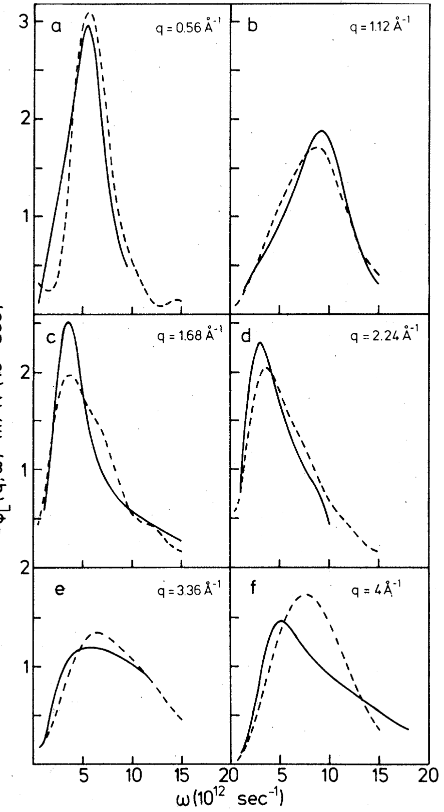

FIG. 1. Normalized longitudinal current-fluctuation spectrum of liquid argon according to the present theory (full curves) in comparison with the molecular dynamics data of Rahman (Ref. 5) [dashed curves (a)–(e)] and in comparison with the free-gas result (dashed curve (f)].

we compare the longitudinal current fluctuation spectrum with Rahman's molecular dynamics calculations for argon [Fig. 1(a)–(e)]. The agreement between theory and experiment is improved compared with the original work of Duderstadt and Akcasu.[14] In Fig. 1(f) the current-fluctuation spectrum is compared with that for a noninteracting system. We consider it remarkable that both curves differ so much for frequencies exceeding 1.5×10^{13} sec^{-1}. The theoretical curve does not change very much in the high-frequency regime if the damping parameter is changed and thus we consider the mentioned effect a realistic feature of liquid dynamics. In Fig. 2 the peak positions of $\phi_L''(q, \omega)$ are compared with Rahman's data. Except for $q \sim 2$ Å^{-1}, where our results yield too sharp resonances, the present theory reproduces the experiment.

For liquid rubidium the interaction potential and the pair-correlation function have been taken from Rahman.[7] The characteristic frequency Ω_E is 6.1×10^{12} sec^{-1}; the damping parameter β is chosen

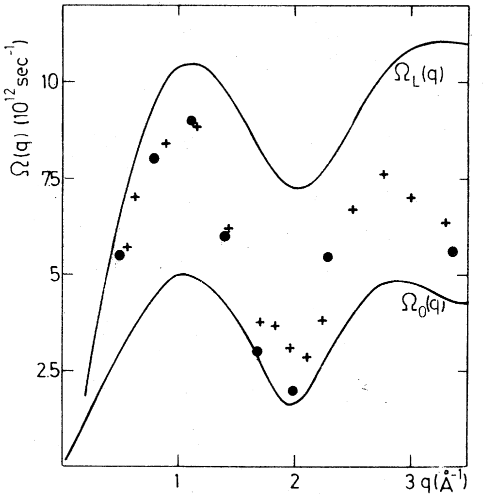

FIG. 2. Peak position of $\phi_L''(q, \omega)$ (dots) compared with Rahman's results (Ref. 5) for liquid argon (crosses).

as 1.59×10^{12} sec^{-1}; in Eq. (43a) one gets an $\overline{\eta}$ about 10% smaller than $\frac{1}{2}\eta$.

In Fig. 3 good agreement between the theoretical results for the current fluctuation spectra and the experimental results of Copley and Rowe[4] is demonstrated. The theoretical results for the peak positions compare well with the experimental ones (Fig. 4). The dynamical structure factor $S(q, \omega)$ of rubidium exhibits a pronounced side peak for wave numbers up to 1.2 Å^{-1}. According to Fig. 5

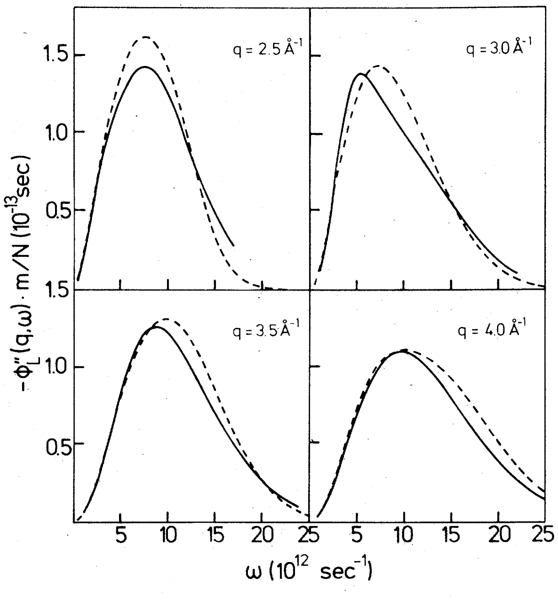

FIG. 3. Normalized longitudinal current-fluctuation spectra of liquid rubidium according to the present theory (full curves) in comparison with the experiments of Copley and Rowe (Ref. 4) (dashed curves).

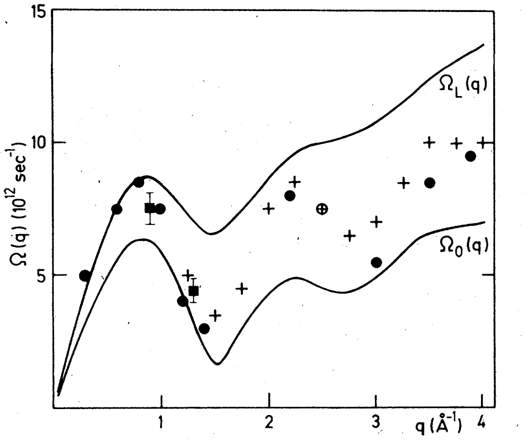

FIG. 4. Peak positions of $\phi''_L(q,\omega)$ (dots) compared with the neutron scattering results of Copley and Rowe (Ref. 4) (crosses) and Suck (Ref. 24) (squares) for liquid rubidium.

this feature is reflected by the present theory, but the resonance is overemphasized to some extent.

For the memory kernel entering the general equation of motion for the phase-space density a kinetic equation has been derived improving the earlier theory of Duderstadt and Akcasu[14] such that the first four frequency moments of the phase-space density-fluctuation spectrum are reproduced correctly. The particle correlations are then described [compare Eqs. (54) and (55)] by the correlations of a tagged particle modified by a number of frequency and wave-number-dependent molecular field terms. The latter terms dominate the motion, and thus the anomalies of the self correlations, discussed earlier,[16] do not yield remarkable qualitative features of $S(q,\omega)$.

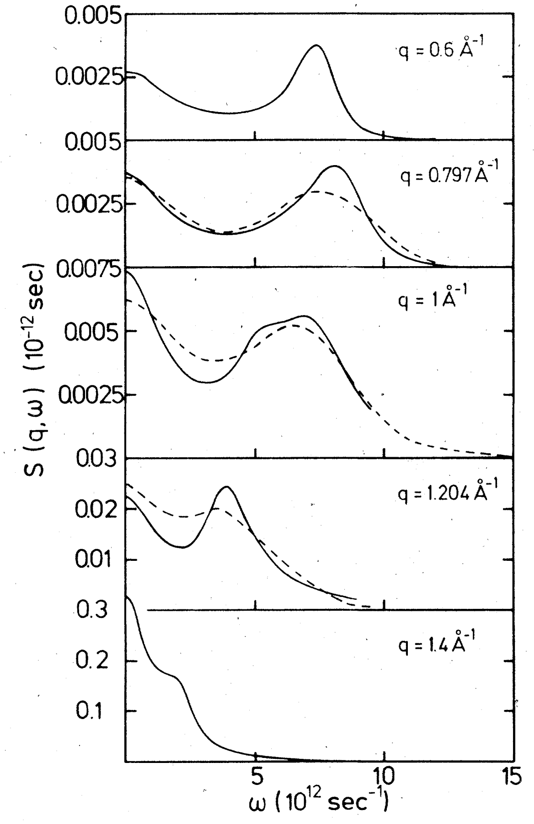

FIG. 5. Dynamical structure factor $S(q,\omega)$ of liquid rubidium. Full curves represent the present theory and dashed curves are Rahman's (Ref. 7) molecular dynamics results.

APPENDIX

In this appendix we want to give some of the detailed calculations entering the derivation of Eq. (54). Starting from Eq. (53) we first have to consider the matrix χ_1, which is given as the sum of a diagonal matrix

$$\chi_1^D = \frac{N}{T}\Omega_E^2 \sum_\alpha a_\alpha^\dagger a_\alpha ,$$

and a separable term,

$$\chi_1^{ND} = \frac{N}{T} nc(q) q^2 v^2 a_z^\dagger |\vec{0}\rangle\langle\vec{0}| a_z - \frac{N}{T}\sum_\alpha \omega_{\alpha\alpha}^2(q) a_\alpha^\dagger |\vec{0}\rangle\langle\vec{0}| a_\alpha .$$

The inverse matrix reads

$$\frac{N}{T}\chi_1^{-1} = (\chi_1^D + \chi_1^{ND})^{-1} = \frac{1}{\Omega_E^2}\left(\sum_{\vec{n}} |\vec{n}\rangle \frac{1}{N}\langle\vec{n}| + \sum_{\alpha=1}^{3} f_\alpha |1_\alpha\rangle\langle 1_\alpha|\right),$$

with the abbreviations

$$|\vec{n}\rangle = |n_1 n_2 n_3\rangle, \quad N = n_1 + n_2 + n_3, \quad |1_\alpha\rangle = a_\alpha^\dagger |\vec{0}\rangle$$

and

$$f_\alpha = [\omega_{\alpha\alpha}^2(q) - nc(q) q^2 v^2 \delta_{\alpha z}] \times [\Omega_E^2 - \omega_{\alpha\alpha}^2(q) + nc(q) q^2 v^2 \delta_{\alpha z}]^{-1} .$$

Inserting the explicit expression for χ_1^{-1} into Eq. (53), we can proceed in reducing the integral equation to a one-dimensional one by integrating over all transverse components. We furthermore multiply the resulting equation for $\phi(p_z, k_z)$ by the first Hermite polynomial in k_z and integrate over k_z. In short-hand notation, we take matrix elements of Eq. (53) between the "states" $\langle 0_x 0_y|$ and $|0_x 0_y 1_z\rangle$. Since the formulas are rather lengthy, we consider the various terms separately,

$$\langle 0_x 0_y | (\chi_1^{-1}(z+\nu)(z - \Omega_0 \chi_0^{-1})\phi | 0_x 0_y 1_z\rangle = \frac{(z+\nu)qv}{\Omega_E^2}\left(\sum_n |n\rangle \frac{1}{n}\langle n| \phi_0^{-1} + nc(q)(1+f_z)|1\rangle\langle 0| + f_z |1\rangle\langle 1|\phi_0^{-1}\right)\Theta .$$

The notation is the same as in Eqs. (53)–(58) with $|n\rangle$ denoting the nth eigenfunction with respect to p_z.

$$\langle 0_x 0_y | \chi_1^{-1}\Omega_1\chi_1^{-1}(z-\Omega_0\chi_0^{-1})\phi\,|0_x 0_y 1_z\rangle\cdot(\Omega_E^2/q^2v^2)$$

$$=\sum_n |n\rangle\frac{1}{n}\langle n|a^\dagger(a+a^\dagger)a\sum_m |m\rangle\frac{1}{m}\langle m|\phi_0^{-1}\Theta+f_z\left[\left(\frac{1}{\sqrt{2}}|2\rangle+|0\rangle\right)\langle 1|+|1\rangle\left(\frac{1}{\sqrt{2}}\langle 2|+\langle 0|\right)\right]\phi_0^{-1}\Theta$$

$$+(\omega_{zz}^2-\nu_{zzz}^2)\frac{(1+f_z)}{\sqrt{2}\,\Omega_E^2}(|2\rangle\langle 1|+|1\rangle\langle 2|)\phi_0^{-1}\Theta+nc(q)(1+f_z)\left[\left(\frac{1}{\sqrt{2}}|2\rangle+|0\rangle\right)\langle 0|+\frac{(\omega_{zz}^2-\nu_{zzz}^2)}{\sqrt{2}\,\Omega_E^2}|2\rangle\langle 0|\right]\Theta$$

$$+(\omega_{zz}^2-\nu_{zzz}^2)\frac{(1+f_z)}{\sqrt{2}\,\Omega_E^2}|1_z\rangle\langle 2_x 0_y 0_z|\phi_0^{-1}\phi\,|0_x 0_y 1_z\rangle\,.$$

We are going to neglect the last term, an approximation that becomes exact within the theory of Duderstadt and Akcasu.[14] The last term on the left-hand side of Eq. (53) is easily evaluated,

$$\langle 0_x 0_y|\chi_0^{-1}\phi\,|0_x 0_y 1_z\rangle=[1-nc(q)\,|0\rangle\langle 0|\,]\Theta\,,$$

as well as the right-hand side,

$$\langle 0_x 0_y|\chi_1^{-1}[(z+\nu)\chi_1-\Omega_1]\chi_1^{-1}\chi_0|0_x 0_y 1_z\rangle\frac{\Omega_E^2}{(1+f_z)}$$

$$=(z+\nu)|1\rangle-qv\left(|0\rangle+\frac{1}{\sqrt{2}}|2\rangle\right)$$

$$-\frac{qv}{\sqrt{2}\,\Omega_E^2}(\omega_{zz}^2-\nu_{zzz}^2)|2\rangle\,.$$

Collecting the various terms we arrive at the final equation for Θ, which is of the general form $(A+B)\Theta=C$. A denotes a diagonal matrix, B is the sum of all separable terms, and C denotes the inhomogeneity. Given the explicit form of A,

$$A=\frac{(z+\nu)qv}{\Omega_E^2}\sum_n |n\rangle\frac{1}{n}\langle n|\phi_0^{-1}$$

$$-\frac{qv}{\Omega_E^2}\sum_{n,m}|n\rangle\frac{1}{n}\langle n|a^\dagger(a+a^\dagger)a|m\rangle\frac{1}{m}\langle m|\cdot qv\phi_0^{-1}-1$$

$$=(1/T)\{(\chi_1^s)^{-1}[(z+\nu)\chi_1^s-\Omega_1^s](\chi_1^s)^{-1}[z-\Omega_0^s(\chi_0^s)^{-1}]$$

$$-(\chi_0^s)^{-1}\}\,,$$

we recognize the corresponding susceptibility and restoring forces for the incoherent function, denoted by the index s. Using the results of I the inverse matrix can be written down

$$A^{-1}=TG\Omega_E^2a^\dagger\frac{1}{z+\nu-qv(a+a^\dagger)}a=T^2G\tilde{N}\,,$$

or, inserted in the equation for Θ,

$$\Theta+A^{-1}B\Theta=A^{-1}C\,,$$

we finally arrive at Eq. (54):

$$\Theta+T^2G\tilde{N}\left(\sum_{\nu,\mu=1}^{2}s_{\nu\mu}|\nu\rangle\langle\mu|\phi_0^{-1}+t_\nu\sum_{\nu=1}^{2}|\nu\rangle\langle 0|\right)\Theta$$

$$=TG|1\rangle+T^2G\tilde{N}\sum_{\nu=1}^{2}u_\nu|\nu\rangle\,,$$

with the coefficients given in Eqs. (57)–(58).

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[1] N. Kroó, G. Borgonovi, K. Sköld, and K. E. Larsson, Phys. Rev. Lett. 12, 721 (1964); S. H. Chen, O. J. Eder, P. A. Egelstaff, B. C. G. Haywood, and F. J. Webb, Phys. Lett. 19, 269 (1965); B. A. Dasannacharya and K. R. Rao, Phys. Rev. 137, A417 (1965); K. Sköld and K. E. Larson, Phys. Rev. 161, 102 (1967).

[2] K. Sköld, J. M. Rowe, G. Ostrowki, and P. D. Randolph, Phys. Rev. A 6, 1107 (1972).

[3] J. B. Suck and W. Gläser, in *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1972), p. 435; W. Gläser, S. Hagen, U. Löffler, J. B. Suck, and W. Schommers, in *The Properties of Liquid Metals*, edited by S. Takeudi (Taylor and Francis, London, 1973), p. 111.

[4] J. R. D. Copley and J. M. Rowe, Phys. Rev. Lett. 32, 49 (1974), and Phys. Rev. A 9, 1656 (1974).

[5] A. Rahman, Phys. Rev. Lett. 19, 420 (1967); and in *Neutron Inelastic Scattering* (International Atomic Energy Agency, Vienna, 1968), Vol. 1, p. 561.

[6] D. Levesque, L. Verlet, and J. Kurkijärvi, Phys. Rev. A 7, 1690 (1973).

[7] A. Rahman, Phys. Rev. Lett. 32, 52 (1974); Phys. Rev. A 9, 1667 (1974).

[8] Compare the review of J. R. D. Copley and S. W. Lovesey, Rep. Prog. Phys. (to be published).

[9] J. K. Percus, in *The Equilibrium Theory of Classical Fluids*, edited by H. L. Frisch and J. L. Lebowitz (Benjamin, New York, 1964); R. Zwanzig, Phys. Rev. 144, 170 (1966); M. Nelkin and S. Ranganathan, Phys. Rev. 164, 222 (1967).

[10] W. C. Kerr, Phys. Rev. 174, 316 (1968).

[11] K. S. Singwi, K. Sköld, and M. P. Tosi, Phys. Rev. Lett. 21, 881 (1968), Phys. Rev. A 1, 454 (1970); K. N. Pathak and K. S. Singwi, Phys. Rev. A 2, 2427 (1974).

[12] L. Sjögren and A. Sjölander (unpublished).

[13] J. M. J. van Leeuwen and S. Yip, Phys. Rev. 139,

A1138 (1965); J. L. Lebowitz, J. K. Percus, and J. Sykes, Phys. Rev. 188, 487 (1969).

[14] J. J. Duderstadt and A. Z. Akcasu, Phys. Rev. A 1, 905 (1970); 2, 1097 (1970).

[15] M. S. Jhon and D. Forster, Phys. Rev. A 12, 254 (1975).

[16] W. Götze and Annette Zippelius, Phys. Rev. A 14, 1842 (1976).

[17] A. Z. Akcasu, N. Corngold, J. J. Duderstadt, Phys. Fluids 13, 2213 (1970).

[18] R. Zwanzig, in *Lectures in Theoretical Physics*, edited by W. Brittin and L. Dunham (Wiley, New York, 1961), Vol. 3, p. 135; H. Mori, Prog. Theor. Phys. 33, 423 (1965); 34, 399 (1965).

[19] D. Forster, Phys. Lett. 46A, 9 (1973).

[20] D. Forster and P. C. Martin, Phys. Rev. A 2, 1575 (1970).

[21] N. K. Ailawadi, A. Rahman, and R. Zwanzig, Phys. Rev. A 4, 1616 (1971).

[22] C. H. Chung, S. Yip, Phys. Rev. 182, 323 (1969).

[23] J. L. Yarnell, M. J. Katz, R. C. Wenzel, and S. H. König, Phys. Rev. A 7, 2130 (1973).

[24] J. B. Suck, Ph.D.thesis (Karlsruhe, 1977) (unpublished).