Properties of the Low-Density Memory Function*

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In an earlier paper an expression was derived for the memory function associated with the classical phase-space fluctuation function. In this paper, we investigate further the properties of this memory function. We dicuss the region of validity of our approximation, the low-wave-number and -frequency limit, as well as general properties of the memory function. We also discuss the hydrodynamics predicted by the memory function and the associated kinetic equation. We see, for example, that the sound velocity is given by the adiabatic speed of sound calculated thermodynamically from that static energy and pressure evaluated to lowest order in the density. Finally, we discuss two techniques for handling the two-particle Liouville operator that appears in the memory function. One technique is to introduce the Koopman operator which serves as the classical time propagator. The second technique is to introduce eigenfunctions of the two-particle Liouville operator. In an appendix the exact form of these eigenfunctions is given in terms of Hamilton's characteristic function. These eigenfunctions, to my knowledge, were not available previously.

INTRODUCTION

In a previous paper (I),[1] we discussed a technique for determining the memory function associated with the equilibrium phase-space fluctuation function. In particular, the memory function appropriate for a low-density system was derived and the classical limit of this memory function was investigated. Finally, a short discussion of the properties of this memory function was given. In this paper, we want to discuss further the properties of this low-density memory function and indicate the range of validity of our approximation. We shall limit ourselves to a study of the classical memory function. We begin with a review of our previous work.

I. SUMMARY

In I we discussed the kinetic equation

$$\left(z-\frac{\vec{k}\cdot\vec{p}}{m}\right)S(\vec{k},\vec{p}\vec{p}\,',z)-\int d^3\bar{p}\;\phi(\vec{k},\vec{p}\vec{\bar{p}},z)S(\vec{k},\vec{\bar{p}}\vec{p}\,',z)=-\tilde{S}(\vec{k},\vec{p}\vec{p}\,') \qquad (1.1)$$

satisfied by the Fourier-Laplace transform of the equilibrium phase-space fluctuation function

$$S(\vec{r}-\vec{r}\,',\vec{p}\vec{p}\,',t-t')=\langle[f(\vec{r}\vec{p}t)-\langle f(\vec{r}\vec{p}t)\rangle][f(\vec{r}\,'\vec{p}\,'t')-\langle f(\vec{r}\,'\vec{p}\,'t')\rangle]\rangle, \qquad (1.2)$$

where

$$f(\vec{r}\vec{p}t)=\sum_{i=1}^{N}\delta(\vec{r}-\vec{r}_i(t))\,\delta(\vec{p}-\vec{p}_i(t)) \quad . \qquad (1.3)$$

ϕ is the memory function associated with S and $\tilde{S}$ is the spatial Fourier transform of the static correlation function $S(\vec{k},\vec{p}\vec{p}\,',t-t'=0)$. We found in I that in the low-density limit

$$\phi(\vec{k},\vec{p}\vec{p}\,',z)=\phi^{(s)}(\vec{k},\vec{p})+\phi^{(c)}(\vec{k},\vec{p}\vec{p}\,',z) \quad , \qquad (1.4a)$$

where

$$\phi^{(s)}(\vec{k},\vec{p})=-(\vec{k}\cdot\vec{p}/m)C(k)f_0(p) \quad , \qquad (1.4b)$$

$$\phi^{(c)}(\vec{k},\vec{p}\vec{p}\,',z)f_0(p')=n^2(\beta/\pi m)^3\nabla_p{}^l\nabla_{p'}{}^j$$
$$\times\int d^3\alpha\, d^3r\, d^3\bar{p}\; e^{-\beta(\alpha^2+\bar{p}^2)/m}g(r)\nabla_r{}^jV(r)$$
$$\times[e^{+i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{\alpha}-\vec{p}\,'+\vec{\bar{p}})-e^{-i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{\alpha}-\vec{p}\,'-\vec{\bar{p}})]$$
$$\times[z-\vec{k}\cdot\vec{\alpha}/m+L(\vec{r},\vec{\bar{p}})]^{-1}\nabla_r{}^lV(r)\,e^{-i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}}), \qquad (1.4c)$$

and

$$\tilde{S}(\vec{k},\vec{p}\vec{p}\,')=f_0(p)\delta(\vec{p}-\vec{p}\,')+h(k)f_0(p)f_0(p') . \qquad (1.5)$$

In these equations, $f_0(p)$ is the Maxwellian

$$f_0(p)=n(\beta/2\pi m)^{3/2}\,e^{-\beta p^2/2m} \quad , \qquad (1.6)$$

$C(k)$ is the direct-static correlation function and is related to the hole function $h(k)$ and the pair-correlation function

$$n^2g(|\vec{r}-\vec{r}\,'|)=\langle\sum_{i\neq j}^{N}\delta(\vec{r}-\vec{r}_i)\delta(\vec{r}\,'-\vec{r}_j)\rangle \qquad (1.7)$$

by

$$h(k)=C(k)/[1-nC(k)]=\int d^3r[g(r)-1]e^{+i\vec{k}\cdot\vec{r}} \quad . \qquad (1.8)$$

In the low-density limit, $g(r)=e^{-\beta V(r)}$, where $V(r)$ is the interparticle potential. Finally, we note that $L(\vec{r},\vec{p})$ is the relative two-particle Liouville operator

$$L(\vec{r},\vec{p})=L_0(\vec{r},\vec{p})+L_I(\vec{r},\vec{p}) \quad , \qquad (1.9a)$$

$$L_0(\vec{r},\vec{p})=-2i\,\vec{p}\cdot\nabla_r/m \quad , \qquad (1.9b)$$

$$L_I(\vec{r},\vec{p})=i\,\nabla_rV(r)\cdot\nabla_p \quad . \qquad (1.9c)$$

The kinetic part of this Liouville operator has a more familiar form if we introduce the reduced mass $\mu = \frac{1}{2}m$.

II. TWO-PARTICLE LIOUVILLE EIGENFUNCTIONS

As a first step in developing a practical method for use in numerical calculations, we must develop techniques for treating the two-particle Liouville operator in the memory function. In this section we develop the eigenfunction method. In Sec. IV we develop the orbit method. Since the eigenfunction method is particularly useful in discussing the general properties of the memory function, we introduce it here.

We introduce the exact two-particle Liouville eigenfunctions which satisfy

$$L(\vec{r},\vec{p})\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0) = \vec{\bar{k}}\cdot\vec{v}_0\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0) \quad . \tag{2.1}$$

We show in Appendix A that we can find such a set of eigenfunctions and that they can be constructed to be complete and orthogonal:

$$\int d^3\bar{k}\,d^3v_0\psi^*(\vec{r}\,'\vec{p}\,'\,|\,\vec{\bar{k}}\vec{v}_0)\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0) = \delta(\vec{r}-\vec{r}\,')\delta(\vec{p}-\vec{p}\,'), \tag{2.2}$$

$$\int d^3r\,d^3p\psi^*(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0)\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\,'\vec{v}_0\,') = \delta(\vec{\bar{k}}-\vec{\bar{k}}\,')\delta(\vec{v}_0-\vec{v}_0\,'). \tag{2.3}$$

We can then introduce the identity (2.2) into (1.4c), and use (2.1) to find

$$\phi^{(c)}(\vec{k},\vec{p}\,\vec{p}\,',z)f_0(p') = \tfrac{1}{2}n^2(\beta/\pi m)^3\nabla_p{}^i\nabla_{p'}{}^j\int d^3\alpha\,d^3\bar{k}\,d^3v_0\,e^{-\beta\alpha^2/m} \times F^*_{k,\alpha}(\vec{\bar{k}}\vec{v}_0\,|\,\vec{p})_i(z-\vec{k}\cdot\vec{\alpha}/m+\vec{\bar{k}}\cdot\vec{v}_0)^{-1} \times F_{k,\alpha}(\vec{\bar{k}}\vec{v}_0\,|\,\vec{p}\,')_j \quad , \tag{2.4}$$

where

$$F_{k,\alpha}(\vec{\bar{k}}\vec{v}_0\,|\,\vec{p})_l = \int d^3r\,d^3\bar{p}\,e^{-\beta H(\vec{r},\vec{\bar{p}})/2}\psi(\vec{r}\,\vec{\bar{p}}\,|\,\vec{\bar{k}}\vec{v}_0) \times\nabla_r{}^l V(r)\rho_k{}^\alpha(\vec{r},\vec{\bar{p}},\vec{p}) \quad , \tag{2.5}$$

$$\rho_k{}^\alpha(\vec{r},\vec{\bar{p}},\vec{p}) = e^{+i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{\alpha}-\vec{p}+\vec{\bar{p}}) - e^{-i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{\alpha}-\vec{p}-\vec{\bar{p}}) . \tag{2.6}$$

We now have a well-defined expression for the memory function. We note that the dynamics are now included in the eigenfunctions. In our further analysis, it will be useful to write $\phi^{(c)}$ in a spectral form. Since the collision operator is analytic in z for $\mathrm{Im}z \neq 0$, and vanishes as $z\to\infty$, we can write

$$\phi^{(c)}(\vec{k},\vec{p}\,\vec{p}\,',z)f_0(p') = \int \frac{d\omega}{\pi}\,\frac{\phi''(\vec{k},\vec{p}\,\vec{p}\,',\omega)f_0(p')}{\omega-z} \quad , \tag{2.7}$$

where

$$\phi''(\vec{k},\vec{p}\,\vec{p}\,',\omega) = \mathrm{Im}\phi^{(c)}(\vec{k},\vec{p}\,\vec{p}\,',\omega+i\,0^+) \quad . \tag{2.8}$$

From (2.4) we have immediately

$$\phi''(\vec{k},\vec{p}\,\vec{p}\,',\omega)f_0(p') = -\tfrac{1}{2}\pi n^2\nabla_p{}^i\nabla_{p'}{}^j\int d^3\alpha\,d^3\bar{k}\,d^3v_0\,F_{k,\alpha}(\vec{\bar{k}}\vec{v}_0\,|\,\vec{p}\,')_j \times F^*_{k,\alpha}(\vec{\bar{k}}\vec{v}_0\,|\,\vec{p})_i(\beta/\pi m)^3\delta(\omega-\vec{k}\cdot\vec{\alpha}/m+\vec{\bar{k}}\cdot\vec{v}_0). \tag{2.9}$$

We then want to take matrix elements of ϕ'' with respect to the complete set of momentum states $H_i(p)$,

$$\phi''_{ij}(\vec{k},\omega) = \int d^3p\,d^3p'\,H_i^*(p)\phi''(\vec{k},\vec{p}\,\vec{p}\,',\omega)f_0(p')H_j(p') = -\tfrac{1}{2}\pi n^2(\beta/\pi m)^3\int d^3\alpha\,d^3\bar{k}\,d^3v_0\,e^{-\beta\alpha^2/m} \times\delta(\omega-\vec{k}\cdot\vec{\alpha}/m+\vec{\bar{k}}\cdot\vec{v}_0)B_i^*(\vec{\bar{k}}\vec{v}_0|\vec{k}\,\vec{\alpha})B_j(\vec{\bar{k}}\vec{v}_0|\vec{k}\,\alpha), \tag{2.10}$$

where

$$B_i(\vec{\bar{k}}\vec{v}_0|\vec{k}\,\vec{\alpha}) = \int d^3p\,d^3\bar{p}\,d^3r\,e^{-\beta H(\vec{r},\vec{\bar{p}})/2} \times\nabla_p H_i(p)\cdot\nabla_r V(r)\psi(\vec{r}\,\vec{\bar{p}}\,|\,\vec{\bar{k}}\vec{v}_0)\rho_k{}^\alpha(\vec{r},\vec{\bar{p}},\vec{p}) \quad . \tag{2.11}$$

If this expression is to have more than formal significance, we need an explicit form for the eigenfunction $\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0)$. We give an explicit expression for these eigenfunctions in Appendix A. The form for the eigenfunctions given by (A2) can be inferred through the use of the eikonel approximation in taking the classical limit of the quantum-mechanical equation:

$$L\,|\,E_i\rangle\langle E_j\,| = (i/\hbar)[H,\,|\,E_i\rangle\langle E_j\,|\,] = (i/\hbar)(E_i-E_j)\,|\,E_i\rangle\langle E_j\,| \quad , \tag{2.12}$$

where the $|E_i\rangle$ are relative two-particle energy eigenstates. While we have seen related approaches in the literature,[2] we have not seen this form (A2) previously identified as the exact classical two-particle eigenfunctions.

III. SOME GENERAL PROPERTIES OF THE MEMORY FUNCTION

In this section we summarize some of the general properties of our low-density memory function.

A. Symmetry Properties

First we note, as can easily be seen from the symmetry properties of the Liouville eigenfunctions, that the collisional part of our memory function satisfies the symmetry conditions[1]

$$\phi^{(c)}(\vec{k},\vec{p}\,\vec{p}\,',z) = -\phi^{(c)}(-\vec{k},\vec{p}\,\vec{p}\,',-z) = -\phi^{(c)}(\vec{k},-\vec{p}-\vec{p}\,',-z) = [\phi^{(c)}(\vec{k},\vec{p}\,\vec{p}\,',z^*)]^* \quad , \tag{3.1}$$

$$\phi^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p')=\phi^{(c)}(\vec{k},\vec{p}'\vec{p},z)f_0(p) \quad . \quad (3.2)$$

These symmetries guarantee, with the kinetic equation (1.1), that $S(\vec{k},\vec{p}\vec{p}',\omega)$ is invariant under translations, rotations, parity, and time reversals.

B. Sum Rules

For short times $S(\vec{k},\vec{p}\vec{p}',z)$ can be calculated from sum rules.[3] These rum rules imply large-z conditions on ϕ^4:

$$\lim_{z\to\infty}\phi(\vec{k},\vec{p}\vec{p}',z)=\phi^{(s)}(\vec{k},\vec{p}) \quad , \quad (3.3)$$

$$\lim_{z\to\infty} z\phi^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p')=n\beta^{-1}\int d^3r\, g(r)[\nabla_r^i\nabla_r^j V(r)]\nabla_p^i\nabla_{p'}^j[f_0(p)\delta(\vec{p}-\vec{p}')]$$
$$-\beta^{-1}\int d^3r\cos(\vec{k}\cdot\vec{r})[g(r)\nabla_r^i\nabla_r^j V(r)+\beta^{-1}\nabla_r^i\nabla_r^j C(r)]\nabla_p^i\nabla_{p'}^j[f_0(p)f_0(p')] \quad . \quad (3.4)$$

In the low-density limit, where $g(r)=e^{-\beta V(r)}$, we see that our memory function satisfies both sum rules and gives the correct short-time behavior for S.

Note, because we have used a kinetic description where the memory function depends on the continuous indices p and p', the single expression (3.4) summarizes in a symmetric form the many sum rules one obtains in a finite-component description.

C. Weakly Coupled Limit

If we expand our approximate memory function to lowest order in the potential and introduce the Fourier transform of the potential, we obtain precisely the form for the memory function obtained by Forster and Martin.[4,5] Forster has shown that in the small-k and -z limit their memory function reduces to the Fokker-Planck operator that is well known in the theory of Brownian motion.

D. Dynamical Stability

We note from (2.10) that if we set $i=j$ we have the inequality

$$\phi_{ii}''(\vec{k},\omega)\le 0 \quad . \quad (3.5)$$

This inequality property is related to the dynamical stability of the system and guarantees the required[4,5] positivity for S:

$$\int d^3p\, d^3p'\, H_i^*(p)S(\vec{k},\vec{p}\vec{p}',\omega)H_i(p')\ge 0 \quad . \quad (3.6)$$

Of course, this positivity requirement of S follows physically from the fluctuation-dissipation theorem (I 2.11)

$$\tfrac{1}{2}\beta\omega S(\vec{k},\vec{p}\vec{p}',\omega)=\chi''(\vec{k},\vec{p}\vec{p}',\omega) \quad (3.7)$$

and the identification of the dissipation with the work done on the system which implies[6]

$$\int d^3p\, d^3p'\, H_i^*(p)\omega\chi''(\vec{k},\vec{p}\vec{p}',\omega)H_i(p')\ge 0 \quad . \quad (3.8)$$

This inequality holds for response functions describing decaying systems. It is easily seen that (3.5) is related to the well-known theorem that eigenvalues of the linearized Boltzmann equation are negative and serve as the inverse relaxation times associated with the decaying modes in a system near equilibrium.

E. Conservation Laws

It is also well known from the Boltzmann theory that the eigenvalues corresponding to the five conserved momentum eigenfunctions

$$\begin{aligned} &H_1(p)=n(p)=1 \quad ,\\ &H_2(p)=g_l(p)=p_3 \quad ,\\ &H_3(p)=\epsilon(p)=(1/\sqrt{6})(p^2-3) \;,\\ &H_4(p)=g_t(p)=p_1 \quad , \quad H_5(p)=g_t'(p)=p_2 \end{aligned} \quad (3.9)$$

are zero. In our case, where the collision operator depends on k and z, Forster and Martin[5] have shown that the matrix elements of the memory function with respect to H_1, H_2, H_4, and H_5 form the potential contributions to the currents associated with the particle and momentum densities multiplied by k. The matrix elements taken with respect to $H_3(p)$ have two parts, the potential contribution to the energy density times z, and the potential contribution to the energy current times k. We have shown these statements hold for our low-density memory function. The explicit forms for these currents are not very illuminating. We shall be satisfied with showing that as k and z go to zero the matrix elements with respect to the five conserved eigenfunctions do vanish, thus guaranteeing the total conservation of particles, momentum, and energy.

1. Conservation of Particles

If $H_1=1$ in (2.11), then

$$B_1(\vec{k}\vec{v}_0\,|\,\vec{k}\vec{\alpha})=\nabla_\alpha\cdot\int d^3r\, d^3\bar{p}\,\psi(\vec{r}\bar{\vec{p}}\,|\,\bar{\vec{k}}\vec{v}_0)\times\nabla_r V(r)2i\sin(\tfrac{1}{2}\vec{k}\cdot\vec{r})=0, \quad (3.10)$$

$$\phi_{1j}''(\vec{k},\omega)=\phi_{i1}''(\vec{k},\omega)=0$$

for any k and ω, which guarantees the conservation of particles.

2. Conservation of Momentum

If $H_i=p_\nu$, $\nu=2$, 4, or 5, then from (2.11)

$$B_\nu(\bar{\vec{k}}\vec{v}_0|\vec{k}\vec{\alpha}) = \int d^3r\, d^3\bar{p}\, \psi(\vec{r}\bar{\vec{p}}|\bar{\vec{k}}\vec{v}_0) \times \nabla_r{}^\nu V(r) 2i \sin(\tfrac{1}{2}\vec{k}\cdot\vec{r}) , \quad (3.11)$$

and as k goes to zero

$$\lim_{k\to 0} B_\nu(\bar{\vec{k}}\vec{v}_0|\vec{k}\vec{\alpha}) = 0$$

for $\nu = 2$, 4, and 5, and the total momentum will be conserved since

$$\lim_{k\to 0}\phi''_{\nu j}(\vec{k},\omega) = \lim_{k\to 0}\phi''_{i\nu}(\vec{k},\omega) = 0 \quad (3.12)$$

for $\nu = 2$, 4, and 5.

3. *Conservation of Energy*

If $H_i = H_3$, then

$$B_3(\bar{\vec{k}}\vec{v}_0|\vec{k}\vec{\alpha}) = \int d^3r\, d^3\bar{p}\, \psi(\vec{r}\bar{\vec{p}}|\vec{k}\vec{v}_0)[2\vec{\alpha}\cdot\nabla_r V(r) 2i \sin(\tfrac{1}{2}\bar{\vec{k}}\cdot\vec{r}) + 2\bar{\vec{p}}\cdot\nabla_r V(r) 2\cos(\tfrac{1}{2}\bar{\vec{k}}\cdot\vec{r})] .$$

If we first set $k=0$ and insert the explicit form for ψ given by (A2), we find

$$B_3(\bar{\vec{k}}\vec{v}_0|\vec{0}\vec{\alpha}) = 4\int d^3r\, n(\vec{r},\vec{v}_0) \times \nabla_r W(\vec{r},\vec{v}_0)\cdot\nabla_r V(r)\, e^{-i\bar{k}\cdot\nabla_{v_0}W(\vec{r},\vec{v}_0)} . \quad (3.13)$$

If we integrate this by parts and use the "microscopic conservation law" (A9) satisfied by the density of orbits, we find

$$B_3(\bar{\vec{k}}\vec{v}_0|\vec{0}\vec{\alpha}) = \int d^3r\, V(r) n(\vec{r},\vec{v}_0)\, e^{-i\bar{k}\cdot\nabla_{v_0}W(\vec{r},\vec{v}_0)} \times 4i \sum_{l,j} \bar{k}_l \nabla_r{}^j W(\vec{r},\vec{v}_0) \nabla_{v_0}{}^l \nabla_r{}^j W(\vec{r},\vec{v}_0) .$$

We can then use (A7) to find

$$B_3(\bar{\vec{k}}\vec{v}_0|\vec{0}\vec{\alpha}) = 4i\,\mu^2(\bar{\vec{k}}\cdot\vec{v}_0)\int d^3r\, d^3\bar{p}\, V(r)\psi(\vec{r}\bar{\vec{p}}|\bar{\vec{k}}\vec{v}_0) . \quad (3.14)$$

Since B_3 is multiplied by the δ function $\delta(\bar{\vec{k}}\cdot\vec{v}_0+\omega)$ in the matrix elements ϕ''_{3j}, we find as we let $\omega\to 0$ that

$$\lim_{\omega\to 0}\lim_{k\to 0}\phi''_{3j}(\vec{k},\omega) = \lim_{\omega\to 0}\lim_{k\to 0}\phi''_{i3}(\vec{k},\omega) = 0 . \quad (3.15)$$

We should point out that this alone does not show that the matrix element of the complex memory function vanishes as $\bar{\vec{k}}\to 0$ and $z\to i0^+$ or

$$\lim_{z\to i0^+}\lim_{k\to 0}\phi_{3j}(\vec{k},z) = \lim_{z\to i0^+}\lim_{k\to 0}\int (d\omega/\pi) \times [\phi''_{3j}(\vec{k},\omega)/(\omega - z)] = 0 . \quad (3.16)$$

As a first step in showing that (3.16) holds, we can use the result (3.15) with the Plemelj relations to find

$$\lim_{z\to i0^+}\lim_{k\to 0}\phi_{3j}(\vec{k},z) = \lim_{k\to 0} P\int (d\omega/\pi)[\phi''_{3j}(\vec{k},\omega)/\omega] . \quad (3.17)$$

Inserting $\phi''_{3j}(\vec{k},\omega)$ using (3.14) and (2.10), we see that the integration over ω can be easily performed:

$$\int d\omega\,[\delta(\omega + \bar{\vec{k}}\cdot\vec{v}_0)/\omega] = -1/(\bar{\vec{k}}\cdot\vec{v}_0) .$$

This $\bar{\vec{k}}\cdot\vec{v}_0$ cancels the factor in B_3. We can then use the completeness of the Liouville eigenfunctions to rewrite (3.17) as

$$\lim_{z\to i0^+}\lim_{k\to 0}\phi_{3j}(\vec{k},z) = \tfrac{1}{2}n^2(4i\mu)(\beta/\pi m)^3 \int d^3r\, d^3p\, d^3\alpha\, e^{-\beta(\alpha^2+p^2)/m} e^{-\beta V(r)} \times V(r)\nabla_r V(r)\cdot\nabla_\alpha[H_j(\vec{\alpha}+\vec{p}) - H_j(\vec{\alpha}-\vec{p})] ,$$

and we note that this vanishes due to the oddness of the integrand in r, thus proving (3.15). Therefore, we have ensured over-all conservation of energy in the system. We note that energy conservation requires some attention to the detailed dynamics of the system. Consequently, one must be very careful when modeling memory functions to be sure this property is preserved.

IV. ORBIT METHOD

The traditional method used in dealing with the Liouville operator is to use the time-propagation property for Koopman's operator e^{+iLt}. This operator has the property that for some function $F(\vec{r},\bar{\vec{p}})$,

$$e^{+iL(\vec{r},\bar{\vec{p}})t} F(\vec{r},\bar{\vec{p}}) = F(\vec{r}(t),\bar{\vec{p}}(t)) , \quad (4.1)$$

where $\vec{r}(t)$ and $\bar{\vec{p}}(t)$ are the phase-space coordinates for the relative motion of two particles as a function of time, and $\vec{r}$ and $\bar{\vec{p}}$ are the initial coordinates. Using this property we show in Appendix B that the memory function can be written in the form

$$\phi^{(c)}(\vec{k},\vec{p}\vec{p}',z) = \phi_1^{(c)}(\vec{k},\vec{p}\vec{p}',z) + \phi_2^{(c)}(\vec{k},\vec{p}\vec{p}',z) , \quad (4.2)$$

where

$$\phi_1^{(c)}(\vec{k},\vec{p}\vec{p}',z) f_0(p') = -n^2(\beta/\pi m)^3 \int d^3\alpha\, d^3r\, d^3\bar{p}\,[e^{+i\vec{k}\cdot\vec{r}/2}\delta(\vec{\alpha}-\vec{p}+\bar{\vec{p}}) + e^{i\vec{k}\cdot\vec{r}/2}\delta(\vec{\alpha}-\vec{p}-\bar{\vec{p}})] \times e^{-\beta(\alpha^2+\bar{p}^2)/m}[z - \vec{k}\cdot\vec{\alpha}/m + L_0(\vec{r},\bar{\vec{p}})]\{g(r)[i(z-\vec{k}\cdot\vec{p}'/m)\int_0^{+\infty} dt\, e^{+i(z-\vec{k}\cdot\vec{\alpha}/m)t}$$

$$\times e^{-i\vec{k}\cdot\vec{r}(t)/2}\,\delta(\vec{p}'-\vec{\alpha}-\vec{\bar{p}}(t))+e^{-i\vec{k}\cdot\vec{r}/2}\,\delta(\vec{p}'-\vec{\alpha}-\vec{\bar{p}})]\} \;, \tag{4.3}$$

$$\phi_2^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p')=(\vec{k}\cdot\vec{p}'/m)C(k)f_0(p)f_0(p') \;. \tag{4.4}$$

Therefore, to obtain numerical results, we must specify the form for the interparticle potential and then solve Newton's equations for the relative trajectory and momenta of the "colliding particles" as a function of time given the phase-space initial conditions $(\vec{r},\vec{\bar{p}})$. We then have several integrals to perform; one of which is a time integration. This form is particularly useful if we choose a potential that has a step discontinuity in space; then the force is a δ function in space and the relative momenta of the particles are constant in time except for a finite change over an infinitesimal time element at the point of force. We can perform the time integration in the memory function explicitly, because the momenta, and, therefore, the coordinates have such a simple time dependence in this case. We intend to discuss the case of discontinuous potentials in a companion paper.

V. BOLTZMANN LIMIT

In I the relationship of our memory function to the Boltzmann collision operator was discussed. Here it is to be demonstrated explicitly that

$$\lim_{z\to i0^+}\lim_{k\to 0}\phi^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p')=iK_B(\vec{p}\vec{p}')f_0(p') \;, \tag{5.1}$$

where $K_B(\vec{p}\vec{p}')$ is the linearized Boltzmann collision operator.[7] First we set $k=0$ in (4.2) to find

$$\phi^{(c)}(\vec{0},\vec{p}\vec{p}',z)f_0(p')=-n^2(\beta/\pi m)^3\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-\beta(\alpha^2+\bar{p}^2)/m}[\delta(\vec{\alpha}-\vec{p}'+\vec{\bar{p}})+\delta(\vec{\alpha}-\vec{p}'-\vec{\bar{p}})]$$
$$\times[z+L_0(\vec{r},\vec{\bar{p}})]\{g(r)[iz\int_0^{+\infty}dt\,e^{+izt}\,\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}}(t))+\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}})]\} \;. \tag{5.2}$$

In the limit as $z\to i0^+$, we can use the identity derived in Appendix C to find

$$\phi^{(c)}(\vec{p}\vec{p}')f_0(p')=in^2(\beta/\pi m)^3\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-\beta(\alpha^2+\bar{p}^2)/m}[\delta(\vec{\alpha}-\vec{p}'+\vec{\bar{p}})+\delta(\vec{\alpha}-\vec{p}'-\vec{\bar{p}})]$$
$$\times(2\vec{\bar{p}}\cdot\nabla_r/m)\{g(r)[\Theta(-\hat{\vec{r}}\cdot\vec{\bar{p}})(\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}})-\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}}^*))]\} \;, \tag{5.3}$$

where the step function $\Theta(-\vec{r}\cdot\vec{\bar{p}})$ is necessary since there will be a collision only if $\vec{r}\cdot\vec{\bar{p}}<0$ (for $\vec{r}\cdot\vec{\bar{p}}>0$, $\vec{\bar{p}}^*=\vec{\bar{p}}$). If we fix $\vec{\bar{p}}$ along the z axis, change integration variables from d^3r to $d\psi b\, db\, dz$ (where ψ is the azimuthal angle and b is the impact parameter), and express the momentum "after the collision" $(\vec{\bar{p}}^*)$ in terms of $\bar{p}$, b, and ψ, we can easily perform the integration over z evaluating the integrand at the surface $z=\pm\infty$. Because of the step function the $z=+\infty$ term does not contribute, and we find

$$\phi^{(c)}(\vec{p}\vec{p}')f_0(p')=-in^2(\beta/\pi m)^3\int d^3\alpha\, d^3\bar{p}\, e^{-\beta(\alpha^2+\bar{p}^2)/m}(2|\bar{p}|/m)[\delta(\vec{\alpha}-\vec{p}'+\vec{\bar{p}})+\delta(\vec{\alpha}-\vec{p}'-\vec{\bar{p}})]$$
$$\times\int_0^{+\infty}b\,db\int_0^{2\pi}d\psi\,[\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}})-\delta(\vec{p}-\vec{\alpha}-\vec{\bar{p}}^*)] \;. \tag{5.4}$$

After doing the integration over α, and performing a change of variables, we find

$$\phi^{(c)}(0,\vec{p}\vec{p}',i0^+)f_0(p')=i\int d^3p_1\, d\Omega\,\sigma(\vec{p}-\vec{p}_1,\Omega)(|\vec{p}-\vec{p}_1|/m)f_0(p)f_0(p_1)$$
$$\times[\delta(\vec{p}'-\vec{p}_1^*)-\delta(\vec{p}'-\vec{p}_1)+\delta(\vec{p}'-\vec{p}^*)-\delta(\vec{p}'-\vec{p})] \;, \tag{5.5}$$

where σ is the differential scattering cross section, $d\Omega$ is the differential solid angle, and the p's are related by

$$\vec{p}+\vec{p}_1=\vec{p}^*+\vec{p}_1^* \;, \tag{5.6}$$

$$\frac{p^2}{2m}+\frac{p_1^2}{2m}=\frac{p^{*2}}{2m}+\frac{p_1^{*2}}{2m} \;. \tag{5.7}$$

When we compare this result with the linearized collision operator $K_B(\vec{p}\vec{p}')$ we see that they differ only be a factor of i.

There are several points to note here. First, since (1.4c) was derived via a density expansion for the self-energy associated with the single-particle Green's function, it has not been necessary to truncate a hierarchy of equations. Consequently, we have not used the molecular chaos assumption. Second, we see that the standard coarse graining arguments are equivalent to looking at the small-frequency and wave-number behavior of the memory function. This, of course, means that we average over the region of two-particle interactions in space and time. Finally, we comment on the fact that we have derived the "irreversible" Boltzmann collision operator from the reversible memory function. The answer to this apparent paradox is that we "prejudiced" the time variable to be used in connection with K_B by requiring that Im$z > 0$. By choosing Im$z > 0$, we were able to use the identity (B9) which meant that we were to propagate quantities forward in time. If we had chosen Im$z < 0$, then the propagator

$$[z - \vec{k}\cdot\vec{\alpha}/m + L(\vec{r},\bar{\vec{p}})]^{-1} = +i\int_0^{-\infty} dt\; e^{-i(z-\vec{k}\cdot\vec{\alpha}/m+L(\vec{r},\bar{\vec{p}}))t}$$

would enter the calculation. Simple manipulations then show that the memory function in the limit $z \to i0^-$, $k \to 0$ is given by (5.5) with $\bar{\vec{p}}^*$ replaced with the time-reversed momentum and $\Theta(-\vec{r}\cdot\vec{p}) \to \Theta(\vec{r}\cdot\bar{\vec{p}})$. If we let $\vec{p} \to -\vec{p}$, $\vec{p}\,' \to -\vec{p}\,'$ and $\bar{\vec{p}} \to -\bar{\vec{p}}$, $\vec{\alpha} \to -\vec{\alpha}$ in the integrand and note $\bar{\vec{p}}^*_{tr}(-\vec{p}) = -\vec{p}^*$, we find

$$\phi^{(c)}(\vec{0}, -\vec{p}-\vec{p}\,', i0^-) = -\phi^{(c)}(\vec{0}, \vec{p}\vec{p}\,', i0^+) \quad ,$$

which is precisely the requirement for time-reversal invariance. Consequently, even in the Boltzmann limit our equation is reversible if we keep track of our position in the complex plane.

VI. HYDRODYNAMICS

It was indicated in I and in Sec. III of this paper, that our memory function is in agreement with the conservation laws governing the system. More specifically, in this section, it is to be demonstrated that our memory function is in agreement with the long-wavelength and small-frequency limit of these conservation laws where there is the hydrodynamical "contraction" of the description. Our kinetic equation in this case should reduce to the five equations forming the Navier-Stokes theory. A complete analysis of the relationship between our memory-function formalism and hydrodynamics would be rather lengthy. The reader is referred to the paper by Forster and Martin (FM)[5] where such an analysis is carried out for their weak-coupling memory function. We note that much of their analysis is not tied to their particular choice for the memory function, but depends on the general symmetry properties satisfied by the memory function and the form of the kinetic equation. Thus, many of their results are applicable to our low-density case if we replace their "mass operator" with our low-density memory function. Here we only want to outline the technique they have developed for extracting the hydrodynamical behavior from the kinetic equation. This is done by showing how one can obtain microscopic expressions for the transport coefficients and by demonstrating that the density-density correlation function does have a sound and diffusive mode in the hydrodynamical limit. In particular, we shall want to show that the sound velocity is equal to the adiabatic speed of sound as predicted by hydrodynamics.

In order to simplify the discussion, let us introduce dimensionless momentum variables. We let $\vec{p} \to mv_0\vec{p}$ for all momenta where $mv_0^2 = \beta^{-1}$, and we define

$$W_0(p) = e^{-p^2/2}/(2\pi)^{3/2} \quad . \tag{6.1}$$

We then introduce a bracket notation for integrals in momentum space. The scalar product for two functions $H(\vec{p})$, $G(\vec{p})$ is denoted by

$$\langle H \,|\, G\rangle = \int d^3p\, H^*(\vec{p}) W_0(p) G(\vec{p}) \quad . \tag{6.2}$$

The matrix elements of some "operator" $R(\vec{p}, \vec{p}\,')$ are defined by

$$\langle H | R \,|\, G\rangle = \int d^3p\, d^3p\,' H^*(\vec{p}) R(\vec{p}\vec{p}\,') W_0(p') G(\vec{p}\,') \quad . \tag{6.3}$$

We then observe that the subspace in momentum space on which K_B vanishes is very important in the hydrodynamic limit. This subspace is spanned by the five orthonormal states $H_i(p)$, defined by (3.9). We will take k in the three direction so that H_i, $i = 1, 2, 3$ are longitudinal modes and H_i, $i = 4, 5$, are transverse modes. These five states can be understood as part of a complete orthonormal set $\{H_i(\vec{p})\}$ such that

$$\langle H_i \,|\, H_j\rangle = \delta_{ij} \quad , \tag{6.4}$$

$$\sum_i |H_i\rangle\langle H_i| = 1 \quad . \tag{6.5}$$

We will not need to specify the form of these functions for $i > 5$ in this section.

We also introduce the projection operator

$$P = \sum_{i=1}^{5} |H_i\rangle\langle H_i| = 1 - Q \quad , \tag{6.6}$$

which projects onto the subspace where K_B vanishes,

$$K_B P = P K_B = 0 \quad . \tag{6.7}$$

We now want to evaluate the transverse-current correlation function

$$S_t(\vec{k}, z) = (mv_0)^2 \langle g_t \,|\, S(\vec{k}, z) \,|\, g_t\rangle = -n(mv_0)^2 \langle g_t \,|\, [z - z^0 - \phi(\vec{k}, z)]^{-1} \,|\, g_t\rangle \tag{6.8}$$

and the density-density fluctuation function

$$S_{nn}(\vec{k}, z) = -n(1 - nC(k))^{-1}$$

$$\times\langle n|[z-z^0(k)-\phi(\vec{k},z)]^{-1}|n\rangle \quad , \qquad (6.9)$$

where the operator in momentum space $z^0(k)$ has the explicit form

$$z^0(\vec{k},\vec{p}\vec{p}\,')=v_0\vec{k}\cdot\vec{p}\,\delta(\vec{p}-\vec{p}\,'). \qquad (6.10)$$

We consider these quantities since one can identify the shear viscosity with the relaxation rate for the transverse excitations, and the sound pole is associated with the excitations in the longitudinal modes. We consider the transverse excitations first and omit the k, z arguments where possible. Forster and Martin have shown that the transverse fluctuation function can be written in the form

$$S_t(\vec{k},z)=-n(mv_0)^2[z-D_t(\vec{k},z)]^{-1} \quad , \qquad (6.11)$$

where

$$D_t(\vec{k},z)=\langle g_t|\overline{\phi}|g_t\rangle+\langle g_t|\overline{\phi}Q[z-Q\overline{\phi}Q]^{-1}Q\overline{\phi}|g_t\rangle \quad , \qquad (6.12)$$

$$\overline{\phi}(\vec{k},z)=z^0(k)+\phi(\vec{k},z) \quad . \qquad (6.13)$$

If we compare (6.11) with the expression from hydrodynamics, we see that the shear viscosity is given by

$$\eta=\lim_{z\to i0^+}\lim_{k\to 0}\frac{imn}{k^2}D_t(\vec{k},z) \quad . \qquad (6.14)$$

We can go further if we note from (3.11), (2.7), and (2.10) that

$$\phi^{(c)}(\vec{k},z)|g_t\rangle=v_0k\tau_t(\vec{k},z)|g_t\rangle \quad , \qquad (6.15)$$

$$\langle g_t|\phi^{(c)}(\vec{k},z)=v_0k\langle g_t|\tau_t(\vec{k},z) \quad , \qquad (6.16)$$

where we expect that

$$\tau_t(0,i0^+)=\lim_{z\to i0^+}\lim_{k\to 0}\tau_t(\vec{k},z)$$

is a well-defined quantity. Then we can easily show, using symmetry arguments, that

$$\langle g_t|z^0+\phi^{(s)}+\phi^{(c)}|g_t\rangle=\langle g_t|\phi^{(c)}(k,z)|g_t\rangle \quad , \qquad (6.17)$$

and since $\phi^{(s)}Q=0$, we can write

$$\langle g_t|(z^0+\phi^{(s)}+\phi^{(c)})Q=v_0k\langle g_t|(p_3+\tau_t(\vec{k},z))Q \quad . \qquad (6.18)$$

We can calculate the matrix element $\phi_{44}(kz)$ for small k and z explicitly. We find to lowest order in k and z that

$$\langle g_t|\phi^{(c)}(\vec{k},z)|g_t\rangle=-\tfrac{1}{4}ik^2v_0\int_0^{+\infty}dt\int d^3r\,d^3p \times W_0(p)g(r)I_V(\vec{r})I_V(\vec{r}(t)) \quad , \qquad (6.19)$$

where $I_V(r)$ is the potential contribution to the dynamical flux for the "shear viscosity of a two-particle system":

$$I_V(\vec{r})=-r_3\nabla_{r_2}V(r) \quad . \qquad (6.20)$$

We then have the expression for the viscosity

$$\eta=\eta_1+\eta_2 \quad , \qquad (6.21a)$$

where

$$\eta_1=\tfrac{1}{4}mn\int_0^{+\infty}dt\int d^3r\,d^3p\,W_0(p)g(r)I_V(\vec{r})I_V(\vec{r}(t)) \quad , \qquad (6.21b)$$

$$\eta_2=mnv_0^2\langle g_t|(p_3+\tau_t(0,i0^+))Q(-K_3^{-1})Q \times(p_3+\tau_t(0,i0^+))|g_t\rangle \quad . \qquad (6.21c)$$

We understand the need for the minus sign multiplying K_B^{-1} since the eigenvalues of K_B are negative and η should come out positive. Thus we see how one can extract expressions for transport coefficients from our microscopic equation of motion. We note that if we had started with the Boltzmann equation, we would have derived the expression

$$\eta_B=mnv_0^2\langle g_t|p_3Q(-K_B^{-1})Qp_3|g_t\rangle \qquad (6.22)$$

for the viscosity. Comparing this result with the result we have derived above, we see that by setting $k=0$ before solving the kinetic equation we lose contributions from the potential to the viscosity.

We now want to consider the existence of sound modes in the system. These modes are longitudinal, and in this case, the density, longitudinal momentum and energy correlation functions are coupled. Because of this coupling, Forster and Martin considered the matrix correlation function

$$G_{ij}(\vec{k},z)=\langle i|[z-z^0(k)-\phi(\vec{k},z)]^{-1}|j\rangle \quad , \qquad (6.23)$$

where i, $j=1$, 2, 3 are the three longitudinal states n, g_l, and ϵ. We note that

$$S_{nn}(\vec{k},z)=-n[1-nC(k)]^{-1}G_{11}(\vec{k},z) \quad . \qquad (6.24)$$

Forster and Martin show, in complete analogy to the transverse case, that we can write

$$[z\delta_{ij}-D_{ij}(\vec{k},z)]G_{jk}=\delta_{ik} \quad , \qquad (6.25)$$

where

$$D_{ij}(\vec{k},z)=\langle i|z^0+\phi|j\rangle +\langle i|(z^0+\phi^{(c)})Q[z-Q(z^0+\phi^{(c)})Q]^{-1}Q \times(z^0+\phi^{(c)})|j\rangle \quad . \qquad (6.26)$$

Forster and Martin give many details about the treatment of such an equation in the hydrodynamical limit. We will not go through the entire analysis, but we will simply show that the position of the sound pole is correct. To find the poles of $S_{nn}(kz)$ we must examine the zeros of the 3×3 matrix $z-D(kz)$ by solving the determinantal equation

$$\det[z-D(\vec{k},z)]=0 \quad . \qquad (6.27)$$

To solve this equation to lowest order in k, z we note that we need D_{ij} to first order in k, z. Since $\langle i|\phi^{(c)}(kz)$ is explicitly of first order in k, we see that the second term in D_{ij} is of second order in k and

$$D_{ij}(\vec{k},z)=\langle i|z^0+\phi^{(s)}+\phi^{(c)}|j\rangle+0(k^2)$$
$$=v_0kD^0_{ij}(x)+0(k^2) \quad , \tag{6.28}$$

where $x=z/v_0k$. The matrix elements of $z^0(k)$ and $\phi^{(s)}$ are straightforward. We discuss the evaluation of the matrix elements $\langle i|\phi^{(c)}(kz)|j\rangle$ in Appendix D. We obtain the result

$$D^0_{ij}(x)=\begin{pmatrix} 0 & 1 & 0 \\ 1-nC(0) & 0 & \sqrt{\tfrac{2}{3}}\,(1+\tfrac{3}{4}d') \\ 0 & \sqrt{\tfrac{2}{3}}(1+\tfrac{3}{4}d') & -xd \end{pmatrix} \quad , \tag{6.29a}$$

where

$$d=\tfrac{1}{3}n\int d^3r\,[\beta V(r)]^2 g(r) \quad , \tag{6.29b}$$

$$d'=-\tfrac{2}{3}n\int d^3r\,[\beta V(r)g(r)+C(r)] \quad . \tag{6.29c}$$

The associated determinantal equation reads

$$X[x^2(1+d)-\tfrac{2}{3}(1+\tfrac{3}{4}d')^2]-x(1+d)[1-nC(0)]=0 \quad ,$$

with the solutions

$$X_1=0 \quad , \tag{6.30a}$$

$$X^2_{2,3}=1-nC(0)+\tfrac{2}{3}[(1+\tfrac{3}{4}d')^2/(1+d)] \quad . \tag{6.30b}$$

The first mode corresponds to a diffusive mode which gives rise to the Rayleigh peak observed in light-scattering experiments and the second and third solutions correspond to the Brillouin peaks with the associated speed of sound

$$C_0^2=V_0^2X^2_{2,3} \quad . \tag{6.31}$$

We can check this expression for the speed of sound against that calculated from hydrodynamics. According to hydrodynamics, the speed of sound is equal to the adiabatic speed of sound

$$C_A^2=\left(\frac{\partial P}{\partial mn}\right)_s \quad . \tag{6.32}$$

We can express this thermodynamic derivative in terms of the n and β variables as

$$\left(\frac{\partial P}{\partial mn}\right)_s=\left(\frac{\partial P}{\partial mn}\right)_\beta+\frac{1}{mn}\left(\frac{\partial P}{\partial \beta}\right)_n\frac{\epsilon+p-n(\partial\epsilon/\partial n)_\beta}{(\partial\epsilon/\partial\beta)_n} \quad , \tag{6.33}$$

where for a low-density system

$$\epsilon=\tfrac{3}{2}nkT+\tfrac{1}{2}n^2\int d^3r\,V(r)e^{-\beta V(r)} \quad , \tag{6.34}$$

and the static pressure is given by

$$P=nkT-\tfrac{1}{2}n^2kTC(0) \quad . \tag{6.35}$$

This follows from the exact equation

$$P=nkT-\tfrac{1}{6}n^2\int d^3r\,\vec{r}\cdot\nabla_r V(r)g(r)$$

and the low-density result

$$\int d^3r\,\vec{r}\cdot\nabla_r V(r)e^{-\beta V(r)}=(3/\beta)\int d^3r\,(e^{-\beta V(r)}-1)$$
$$=(3/\beta)C(0) \quad .$$

We easily compute the thermodynamical derivatives to find

$$C_A^2=V_0^2\left(1-nC(0)+\frac{2}{3}\frac{\{1-\frac{1}{2}n\int d^3r\,[\beta V(r)\,e^{-\beta V(r)}+e^{-\beta V(r)}-1]\}^2}{1+\frac{1}{3}n\int d^3r\,[\beta V(r)]^2e^{-\beta V(r)}}\right) \quad . \tag{6.36}$$

Comparing this with the expression derived from dynamical considerations, we see that the results are identical. This is, of course, an important test for our theory, and we have agreement. We will not investigate the damping of these sound waves here since the analysis follows that for the viscosity and we do not anticipate any unexpected results.

In summary, we see that (1.4) gives us considerably more information about the dynamics of a low-density system than the Boltzmann equation. For example, if we had used the Boltzmann equation we would replace (6.36) with $C_A^2=\frac{5}{3}v_0^2$. This is, of course, because the Boltzmann equation is only compatible with free-particle thermodynamics.

VII. RANGE OF VALIDITY OF THE THEORY

Before going further we want to discuss the range of densities for which our theory is applicable. It is plausible that our approximation for the memory function breaks down at approximately the same densities that our approximation for the static correlation function breaks down. This is another way of saying, at least for short times, that we believe that higher-order corrections in the densities become important through mean-field effects before they enter through multiple-scattering effects, where we consider the density corrections to the time propagator e^{iLt}. We can, therefore, estimate the density at which our approximation fails by analyzing where $C(r)$ deviates from its low-density value. In this regard the solution of the Percus-Yevick equation for hard spheres should be adequate.[8] In that approximation we find that the direct correlation function deviates from its low-density value when

$$\tfrac{4}{3}\pi r_0^3 n\approx 0.10$$

(r_0 = hard-core diameter) or $r_0/n^{1/3}\sim 0.5$. We expect, therefore, that our "low-density" approximation is valid for moderately dense gases. In contrast, our approximation is clearly inadequate for describing liquid systems where $\frac{4}{3}\pi r_0^3 n \sim 3$.

In discussing an experimental test of this theory, we note that light-scattering experiments do not probe with wave numbers large enough to observe deviations from the Boltzmann equation. This is supported by the fact that the linearized Boltzmann equation[9] adequately describes the light-scattering experiments of Greytak and Benedek[10] which probed nonhydrodynamical behavior in gases.

The need for a theory beyond the Boltzmann equation develops only for systems where either $kr_0 \sim 0.1$ or $n4\pi r_0^3/3 \sim 0.1$ where r_0 is the effective hard-core radius. The first condition indicates that the probe samples the region of interaction and the second indicates that the density is large enough for static correlations to be important $[C(k=0) \approx -\frac{4}{3}\pi r_0^3]$. These two conditions also demand that $kl \sim 1$ where $l = \frac{1}{4}(\frac{1}{2}\pi)^{1/2}(n\pi r_0^2)^{-1}$ is the mean free path.

Greytak and Benedek performed their experiments on xenon at 24.8 °C and a pressure of 780 mm Hg, reaching wave numbers on the order of 2×10^5 cm^{-1}. If we note that the effective hard-core radius for xenon is about 5.2 Å, we have $kr_0 \sim 0.01$. Thus, even for these relatively large wave numbers, one does not really need the detailed structure of the memory function. We also note that these experiments were performed on a low-density system where the density times the direct correlation function is a small number,

$$nC(0) \approx -n\tfrac{4}{3}\pi r_0^3 \approx -10^{-2} \quad .$$

Consequently, the static part of the memory function can be neglected and one can use the free-particle initial condition

$$\tilde{S}(\vec{k}, \vec{p}\vec{p}\,') = \delta(\vec{p} - \vec{p}\,') f_0(p') \quad .$$

At present, the only experiments that measure S for wave numbers such that $kr_0 \sim 0.1$ are neutron scattering experiments on simple liquids. The densities involved in these experiments are, from a rigorous point of view, beyond the range of our "low-density" approximation. Thus the results we can derive from our memory function which show definite deviations from predictions of the Boltzmann equation cannot as yet be checked against experiment. Consequently, it would be very interesting to see the results of scattering experiments performed in the intermediate-density region $n4\pi/3r_0^3 \sim 0.1$.

VIII. DISCUSSION

In this paper we have discussed some of the properties of our low-density memory function. We have shown that our kinetic equation is consistent with all of the independent checks (sum rules, hydrodynamics, etc.) that we have at our disposal. We are, of course, more interested in solving our kinetic equation to determine the momentum moments of S for intermediate k and ω where we have no information. Techniques for carrying through this calculation will be discussed elsewhere.

Even before carrying out such an explicit calculation, we feel that we have gained some insight into the structure of the memory function associated with the phase-space fluctuation function. Furthermore, we believe that much of this information will be very useful in understanding memory functions associated with more complicated systems.

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APPENDIX A: TWO-PARTICLE LIOUVILLE EIGENFUNCTIONS

The solutions of the eigenfunction equation

$$L(\vec{r}, \vec{p})\psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\,\vec{v}_0) = \vec{\bar{k}} \cdot \vec{v}_0 \psi(\vec{r}\,\vec{p}\,|\,\vec{\bar{k}}\vec{v}_0) \quad , \tag{A1}$$

where $L(r,p)$ is given by (1.9), are given by

$$\psi(\vec{r}\,\vec{p}\,|\vec{\bar{k}}\,\vec{v}_0) = n(\vec{r}, \vec{v}_0)\delta(\vec{p} - \nabla_r W(\vec{r}, v_0))\, e^{-i\vec{\bar{k}}\cdot\nabla_{v_0} W(\vec{r}, \vec{v}_0)} \,, \tag{A2}$$

where

$$n(\vec{r}, \vec{v}_0) = (2\pi)^{-3/2} \det(\nabla_{v_0}{}^i \nabla_r{}^j W(\vec{r}, \vec{v}_0)) \tag{A3}$$

is what we call the density of orbits.[11] $W(r, v_0)$ is Hamilton's characteristic function satisfying the equation

$$[\nabla_r W(\vec{r}, \vec{v}_0)]^2/2\mu + V(r) = \tfrac{1}{2}\mu v_0^2 \quad , \tag{A4}$$

which leads to the expressions, via differentiation,

$$\nabla_r{}^i V(r) + (1/\mu)\textstyle\sum_j \nabla_r{}^j W(\vec{r}, \vec{v}_0)\nabla_r{}^i \nabla_r{}^j W(\vec{r}, \vec{v}_0) = 0 \,, \tag{A5}$$

$$\mu v_0^i = (1/\mu)\textstyle\sum_j \nabla_r{}^j W(\vec{r}, \vec{v}_0)\nabla_r{}^j \nabla_{v_0}{}^i W(\vec{r}, \vec{v}_0) \quad , \tag{A6}$$

$$\nabla_r{}^i \textstyle\sum_j \nabla_r{}^j W(\vec{r}, \vec{v}_0)\nabla_r{}^j \nabla_{v_0}{}^i W(\vec{r}, \vec{v}_0) = 0 \,. \tag{A7}$$

The quantity $n(r, v_0)$ satisfies the conservation law

$$\textstyle\sum_i \nabla_r{}^i [\nabla_r{}^i W(\vec{r}, \vec{v}_0) n(\vec{r}, \vec{v}_0)] = 0 \quad . \tag{A8}$$

Proof of (A8)

If we define the quantity

$$Q = \textstyle\sum_i \nabla_r{}^i W(\vec{r}, \vec{v}_0)\nabla_r{}^i n(\vec{r}, \vec{v}_0)$$

and note the identity

$$\delta(\det A) = \det A \sum_{i,j} A_{ij}^{-1}\,\delta A_{ji} \quad ,$$

we have then

$$Q=\sum_l \nabla_r{}^l W(\vec r,\vec v_0)\sum_{i,j}[\nabla_r{}^i\nabla_{v_0}{}^j W(\vec r,\vec v_0)]^{-1}[\nabla_r{}^l\nabla_r{}^j\nabla_{v_0}{}^i W(\vec r,\vec v_0)]n(\vec r,\vec v_0)$$

$$=-n(\vec r,\vec v_0)\sum_{i,j,l}\nabla_r{}^l\nabla_{v_0}{}^i W(\vec r,\vec v_0)[\nabla_r{}^i\nabla_{v_0}{}^j W(\vec r,\vec v_0)]^{-1}\nabla_r{}^l\nabla_r{}^j W(\vec r,\vec v_0)\ ,$$

where we have used (A8) to obtain the last line. Then, since $\sum_i X_{1i}X_{ij}^{-1}=\delta_{1j}$, we have

$$Q=-n(\vec r,\vec v_0)\textstyle\sum_l \nabla_r{}^l\nabla_r{}^l W(\vec r,\vec v_0)\ .$$

Adding Q to $(\nabla_r{}^2W)n(r,v_0)$, we easily prove (A8). As a first step to showing that ψ given by (A3) and (A4) is a solution to (A1), we consider the effect of the kinetic part of the Liouville operator on the momentum δ function in ψ. We have

$$L_0(\vec r,\vec p)\delta(\vec p-\nabla_r W(\vec r,\vec v_0))=i\sum_{i,j}\frac{p_i}{\mu}[\nabla_r{}^i\nabla_r{}^j W(\vec r,\vec v_0)]\nabla_p{}^j\delta(\vec p-\nabla_r W(\vec r,\vec v_0))$$

$$=i\sum_{i,j}\frac{\nabla_r{}^i W(\vec r,\vec v_0)}{\mu}[\nabla_r{}^i\nabla_r{}^j W(\vec r,\vec v_0)]\nabla_p{}^j\delta(\vec p-\nabla_r W(\vec r,\vec v_0))$$

$$-i\sum_i\frac{[\nabla_r{}^i\nabla_r{}^i W(\vec r,\vec v_0)]}{\mu}\delta(\vec p-\nabla_r W(\vec r,\vec v_0))\ .$$

Using (A6) with the line above, we find that

$$L(\vec r,\vec p)\delta(\vec p-\nabla_r W(\vec r,\vec v_0))=-i[\nabla_r{}^2W(\vec r,\vec v_0)/\mu]\delta(\vec p-\nabla_r W(\vec r,\vec v_0))\ .$$

Combining this expression with the conservation law (A9), we can easily show

$$L(\vec r,\vec p)[\delta(\vec p-\nabla_r W(\vec r,\vec v_0))n(\vec r,\vec v_0)]=0\ .$$

Then we have

$$L(\vec r,\vec p)\psi(\vec r,\vec p\,|\,\bar{\vec k}\vec v_0)$$

$$=n(\vec r,\vec v_0)\delta(\vec p-\nabla_r W(\vec r,\vec v_0))L(\vec r,\vec p)\,e^{+i\bar k\cdot\nabla_{v_0}W(\vec r,\vec v_0)/\mu}$$

$$=n(\vec r,\vec v_0)\delta(\vec p-\nabla_r W(\vec r,\vec v_0))\sum_{j,l}\frac{p_j\nabla_r{}^j\bar k_l\nabla_{v_0}{}^l W(\vec r,\vec v_0)}{\mu}\times e^{+i\bar k\cdot\nabla_{v_0}W(\vec r,\vec v_0)/\mu}$$

$$=\psi(\vec r\,\vec p\,|\,\bar{\vec k}\vec v_0)\sum_{j,l}\frac{\bar k_l}{\mu^2}\nabla_r{}^j W(\vec r,\vec v_0)\nabla_r{}^j\nabla_{v_0}{}^l W(\vec r,\vec v_0)\ ,$$

and, on using (A7), we recover (A1).

Finally, we want to show that these eigenfunctions are complete. We consider the integral

$$\int d^3\bar k\,d^3v_0\,\psi^*(\vec r\,\vec p\,|\,\bar{\vec k}\vec v_0)\psi(\vec r\,'\vec p\,'|\,\bar{\vec k}\vec v_0)$$

$$=\int d^3v_0\,\delta(\vec p-\nabla_r W(\vec r,\vec v_0))\delta(\vec p\,'-\nabla_{r'}W(\vec r\,',\vec v_0))$$

$$\times\delta(\nabla_{v_0}W(\vec r,\vec v_0)-\nabla_{v_0}W(\vec r\,',\vec v_0))(2\pi)^3n(\vec r,\vec v_0)n(\vec r\,',\vec v_0)\,.$$

If we note the identity

$$\delta(\nabla_{v_0}W(\vec r,\vec v_0)-\nabla_{v_0}W(\vec r\,',\vec v_0))=\frac{\delta(\vec r-\vec r\,')}{\det(\nabla_{v_0}{}^i\nabla_r{}^j W(\vec r,\vec v_0))}\,,$$

we can write

$$\int d^3\bar k\,d^3v_0\,\psi^*(\vec r\,\vec p\,|\,\bar{\vec k}\vec v_0)\psi(\vec r\,'\vec p\,'|\,\bar{\vec k}\vec v_0)$$

$$=\delta(\vec r-\vec r\,')\delta(\vec p-\vec p\,')\int d^3v_0\,\delta(p-\nabla_r W(\vec r,\vec v_0))\times\det(\nabla_{v_0}{}^i\nabla_r{}^j W(\vec r,\vec v_0))\ .$$

Then, after setting $\bar p_i=\nabla_r{}^i W(\vec r,\vec v_0)$ and noting that

$$\det(\nabla_{v_0}{}^i\nabla_r{}^j W(\vec r,\vec v_0))\,d^3v_0=d^3\bar p\ ,$$

we recover (2.8). Using quite similar arguments, we can show the orthogonality property (2.9).

APPENDIX B: ORBIT MEMORY FUNCTION

If we rewrite (1.4c) by reintroducing the "interaction" Liouville operators and the definitions

$$\rho_k(\bar{\vec p})=e^{-i\vec k\cdot\vec r/2}\,\delta(\vec p-\vec\alpha-\bar{\vec p})\ ,\qquad\text{(B1)}$$

$$\rho_k'(\bar{\vec p})=e^{+i\vec k\cdot\vec r/2}\,\delta(\vec\alpha-\vec p\,'+\bar{\vec p})+e^{-i\vec k\cdot\vec r/2}\,\delta(\vec\alpha-\vec p\,'-\bar{\vec p})\ ,\qquad\text{(B2)}$$

we have

$$\phi^{(c)}(\vec k,\vec p\,\vec p\,',z)f_0(p')$$

$$=-n^2(\beta/\pi m)^3\int d^3\alpha\,d^3r\,d^3\bar p\,e^{-\beta(\alpha^2+\bar p^2)/m}g(r)$$

$$\times(L_I(\vec r,\bar{\vec p})\rho_k'(\bar{\vec p}))[z-(\vec k\cdot\vec\alpha/m)+L(\vec r,\bar{\vec p})]^{-1}\times L_I(\vec r,\bar{\vec p})\rho_k(\bar{\vec p})\ .\qquad\text{(B3)}$$

If we then integrate by parts over $\bar p$ and use the identities

$$L_I(\vec r,\bar{\vec p})\,e^{-\beta\bar p^2/m}g(r)=-L_0(\vec r,\bar{\vec p})\,e^{-\beta\bar p^2/m}g(r)\ ,\qquad\text{(B4)}$$

$$L_I(z-\vec k\cdot\vec\alpha/m+L)^{-1}L_I=L_I-(z-\vec k\cdot\vec\alpha/m+L_0)\times(z-\vec k\cdot\vec\alpha/m+L)^{-1}L_I\ ,\qquad\text{(B5)}$$

we can break $\phi^{(c)}$ into two parts

$$\phi_1^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p') = -n^2(\beta/\pi m)^3 \int d^3\alpha\, d^3r\, d^3\bar{p}\, \rho_k'(\vec{\bar{p}})\, e^{-\beta(\alpha^2+\bar{p}^2)/m}(z-\vec{k}\cdot\vec{\alpha}/m + L_0(\vec{r},\vec{\bar{p}})) \times [g(r)(z-\vec{k}\cdot\vec{\alpha}/m + L(\vec{r},\vec{\bar{p}}))^{-1}L_I(\vec{r},\vec{\bar{p}})\rho_k(\vec{\bar{p}})] \quad , \tag{B6}$$

$$\phi_2^{(c)}(\vec{k},\vec{p}\vec{p}',z)f_0(p') = n^2(\beta/\pi m)^3 \int d^3\alpha\, d^3r\, d^3\bar{p}\, \rho_k(\vec{\bar{p}})g(r)\, e^{-\beta(\alpha^2+\bar{p}^2)/m}\, L_I\, \rho_k(\vec{\bar{p}}) \quad . \tag{B7}$$

We not concentrate on the first term. If we use the integral representation (for Im$z>0$)

$$[z-(\vec{k}\cdot\vec{\alpha}/m)+L(\vec{r},\vec{\bar{p}})]^{-1} = -i\int_0^{+\infty} dt\, e^{+i[z-\vec{k}\cdot\vec{\alpha}/m + L(\vec{r},\vec{\bar{p}})]t} \tag{B8}$$

and the time-propagation property of e^{iLt} (4.1), we find after simple manipulations the form for $\phi_1^{(c)}$ given by (4.3). $\phi_2^{(c)}$ is relatively easy to evaluate in the form (4.4), if we note the identity

$$\nabla_r V(r)\, e^{-\beta V(r)} = -\beta^{-1}\nabla_r(e^{-\beta V(r)}-1) \quad , \tag{B9}$$

which allows one to integrate by parts over r.

APPENDIX C: SMALL-z THEOREM

We consider a system where two particles interact through a short-ranged potential. By short ranged, we mean that after some large time T_c, the relative momentum of the particles approaches arbitrarily close to some asymptotic value. Therefore, for some function of the momentum $f(\vec{p}(t))$, we have

$$f(\vec{p}(t)) = f^*(\vec{p}^*) \quad \text{for} \quad t > T_c \quad , \tag{C1}$$

where

$$\vec{p}(t) = \vec{p}^* \quad \text{for} \quad t > T_c \quad .$$

We can then use the final value theorem of Laplace transforms[12] to find

$$\lim_{z\to i0^+} iz\int_0^{+\infty} dt\, e^{+izt} f(t) = \lim_{t\to\infty}[-f(t)] = -f^*(\vec{p}^*) \; . \tag{C2}$$

APPENDIX D: MATRIX ELEMENTS FOR SMALL k AND z

First we consider the matrix elements between the longitudinal states $\langle i|\phi^{(c)}|j\rangle$ (i, j = 1, 2, 3) for small k and z. We have immediately that

$$\langle 1|\phi^{(c)}|j\rangle = \langle i|\phi^{(c)}|1\rangle = 0 \quad . \tag{D1}$$

We are interested then in finding the matrix elements $\langle 2|\phi^{(c)}|3\rangle = \langle 3|\phi^{(c)}|2\rangle$, $\langle 2|\phi^{(c)}|2\rangle$, and $\langle 3|\phi^{(c)}|3\rangle$ to first order k and z. From (1.4c) we have

$$\langle 2|\phi^{(c)}|2\rangle = (in/\pi^3)(mv_0)^{-7}\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-(\alpha^2+\bar{p}^2)} \times g(r)\nabla_{r_3}V(r)2i\sin(\tfrac{1}{2}\vec{k}\cdot\vec{v}) \times [z-kv_0\alpha_3+L(\vec{r},\vec{\bar{p}})]^{-1} \times i\,\nabla_{r_3}V(r)\,e^{-i\vec{k}\cdot\vec{r}/2} \; .$$

If we let $r_3\to -r_3$ and $\bar{p}_3\to -\bar{p}_3$ and add $\frac{1}{2}$ times the two resulting expressions for $\langle 2|\phi^{(c)}|2\rangle$, we find

$$\langle 2|\phi^{(c)}|2\rangle = (2in/\pi^3)(mv_0)^{-7}\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-(\alpha^2+\bar{p}^2)} \times g(r)\nabla_{r_3}V(r)\sin(\tfrac{1}{2}\vec{k}\cdot\vec{r}) \times [z-kv_0\alpha_3+L(\vec{r},\vec{\bar{p}})]^{-1} \times \nabla_{r_3}V(r)\sin(\tfrac{1}{2}\vec{k}\cdot\vec{r}) \; , \tag{D2}$$

and $\langle 2|\phi^{(c)}|2\rangle$ is explicitly of second order in k.

We next calculate the matrix element $\langle 3|\phi^{(c)}|3\rangle$. This calculation is facilitated by the use of (1.4c) in terms of dimensionless momentum together with the identities

$$[z-\vec{k}\cdot\vec{\alpha}/m+L(\vec{r},\vec{\bar{p}})]^{-1}L_I(\vec{r},\vec{\bar{p}}) = 1-[z-\vec{k}\cdot\vec{\alpha}/m+L(\vec{r},\vec{\bar{p}})]^{-1}[z-\vec{k}\cdot\vec{\alpha}/m+L_0(\vec{r},\vec{\bar{p}})],$$

(B9), (B10), and (4.1). We have, after performing the integrations over $\vec{p}$ and $\vec{p}'$,

$$\langle 3|\phi^{(c)}|3\rangle = (in/3\beta\pi^3)(mv_0)^{-7}\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-(\alpha^2+\bar{p}^2)}\,\nabla_r C(r)\cdot[e^{+i\vec{k}\cdot\vec{r}/2}(\vec{\alpha}+\vec{\bar{p}}) - e^{-i\vec{k}\cdot\vec{r}/2}(\vec{\alpha}-\vec{\bar{p}})]i \times\int_0^{+\infty} dt\, e^{+i(z-kv_0\alpha_3)t}\{[z-kv_0(\alpha_3+\bar{p}_3(t))]e^{-ikr_3(t)/2}(\vec{\alpha}+\vec{\bar{p}}(t))^2 - [z-kv_0(\alpha_3+\bar{p}_3)]e^{-ikr_3/2}(\vec{\alpha}+\vec{\bar{p}})^2\} \; . \tag{D3}$$

Expanding this expression in powers of k, we see that the corrections to

$$\langle 3|\phi^{(c)}|3\rangle = \langle 3|\phi^{(c)}(0,z)|3\rangle$$

are of order k^2. Therefore, to lowest order we have

$$\langle 3|\phi^{(c)}|3\rangle$$

$$= -(2zn/3\pi^3\beta)(mv_0)^{-7}\int d^3\alpha\, d^3r\, d^3\bar{p}\, e^{-(\alpha^2+\bar{p}^2)}$$

$$\times \vec{\bar{p}}\cdot\nabla_r C(r)\int_0^{+\infty} dt\, e^{+izt}\{[\vec{\alpha}+\vec{\bar{p}}(t)]^2-(\vec{\alpha}+\vec{\bar{p}})^2\} . \quad \text{(D4)}$$

After doing the integration over α, we find

$$\langle 3|\phi^{(c)}|3\rangle = -(2zn/3\pi^{3/2}\beta)(mv_0)^{-4}\int d^3r\, d^3\bar{p}\, e^{-\bar{p}^2}$$

$$\times \vec{\bar{p}}\cdot\nabla_r C(r)\int_0^{+\infty} dt\, e^{+izt}(\bar{p}^2(t)-\bar{p}^2) . \quad \text{(D5)}$$

If we then use conservation of energy

$$mv_0^2\bar{p}^2(t)+V(\vec{r}(t)) = mv_0^2\bar{p}^2+V(r) \quad \text{(D6)}$$

and note that

$$\int d^3r\,[\nabla_r C(r)]\,V(r) = 0 \quad ,$$

we have

$$\langle 3|\phi^{(c)}|3\rangle = (2zmn/3\pi^{3/2}\beta)(mv_0)^{-2}\int d^3r\, d^3\bar{p}\, e^{-\bar{p}^2}$$

$$\times \vec{\bar{p}}\cdot\nabla_r C(r)\int_0^{+\infty} dt\, e^{+izt}V(r(t)) \quad . \text{(D7)}$$

We now concentrate on the integral

$$I = \int d^3r\, d^3\bar{p}\, e^{-\bar{p}^2}\,\vec{\bar{p}}\cdot\nabla_r C(r)\int_0^{+\infty} dt\, e^{+izt}V(\vec{r}(t)) \quad , \quad \text{(D8)}$$

which we can immediately rewrite as

$$I = -\beta\int d^3r\, d^3\bar{p}\,\vec{\bar{p}}\cdot\nabla_r V(r)\, e^{-\beta H(\vec{r},\vec{\bar{p}})}$$

$$\times\int_0^{+\infty} dt\, e^{+izt}\, e^{+iL(\vec{r},\vec{\bar{p}})t}\, V(\vec{r}) \quad . \quad \text{(D9)}$$

After integrating (D9) by parts,

$$I = -\beta\int d^3r\, d^3\bar{p}\, e^{-\beta H}V(r)$$

$$\times\int_0^{+\infty} dt\, e^{+izt}\, e^{-iL(\vec{r},\vec{\bar{p}})t}\,\vec{\bar{p}}\cdot\nabla_r V(r) \quad , \quad \text{(D10)}$$

and after letting $\vec{r}\to-\vec{r}$, we have

$$I = \beta\int d^3r\, d^3\bar{p}\, e^{-\beta H}V(r)\int_0^{+\infty} dt\, e^{+izt}\,\vec{\bar{p}}(t)\cdot\nabla_r V(r(t)) . \quad \text{(D11)}$$

Using Newton's second law, (D11) can be written as

$$I = -\tfrac{1}{2}\beta mv_0\int d^3r\, d^3\bar{p}\, e^{-\beta H}V(r)\int_0^{+\infty} dt\, e^{+izt}\, d/dt\,\bar{p}^2(t) \quad . \quad \text{(D12)}$$

To lowest order in z we have

$$I = -\tfrac{1}{2}\beta mv_0\int d^3r\, d^3\bar{p}\, e^{-\beta H}V(r)[\bar{p}^2(\infty)-\bar{p}^2] \quad \text{(D13)}$$

$$= -(\beta/2v_0)\int d^3r\, d^3\bar{p}\, e^{-\beta H}V^2(r) \quad , \quad \text{(D14)}$$

so that

$$\langle 3|\phi^{(c)}|3\rangle = -\tfrac{1}{3}nz\int d^3r\,[\beta V(r)]^2 g(r) \quad . \quad \text{(D15)}$$

After a similar analysis, we can show

$$\langle 2|\phi^{(c)}|3\rangle = -(nkv_0/\sqrt{6})\int d^3r\,[C(r)+\beta V(r)g(r)] \quad . \quad \text{(D16)}$$

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