

## Statistical mechanics of dense ionized matter. V. Hydrodynamic limit and transport coefficients of the classical one-component plasma

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We study the static and dynamical correlations of density fluctuations in a classical one-component plasma (OCP), in the framework of thermodynamic fluctuation theory and linearized hydrodynamics. First we show that the fluctuations of the local electric field stabilize the OCP against density fluctuations, even when the compressibility becomes negative for  $\Gamma \gtrsim 3$ , where  $\Gamma$  is the usual plasma coupling parameter. Then, following closely the work of Mountain, we compute the dynamical structure factor in the hydrodynamic limit and show that the thermal Rayleigh peak vanishes in the long-wavelength limit, in agreement with recent molecular dynamics results. The shear and bulk viscosity coefficients  $\eta$  and  $\zeta$  are calculated for large  $\Gamma$  in the framework of the "generalized hydrodynamics" formalism, using the known short-time expansion of the correlation functions. The coefficient  $\eta$  is found to exhibit a minimum as a function of  $\Gamma$  and  $\zeta$  is found to be negligible compared to  $\eta$  in the OCP.

### I. INTRODUCTION

In a recent paper of this series<sup>1</sup> the spectra of the longitudinal and transverse current-current correlation functions of a classical one-component system of point ions, immersed in a uniform background of opposite charge (the OCP), have been computed by the molecular dynamics (MD) method<sup>2</sup> over a wide range of the dimensionless parameter  $\Gamma = (Ze)^2/\bar{v}k_B T$ , which entirely determines the thermodynamic state of the OCP;  $Ze$  is the ionic charge,  $T$  the temperature,  $k_B$  is Boltzmann's constant, and  $\bar{v}$  is the ion-sphere radius, defined as

$$\bar{v} = (3/4\pi\rho)^{1/3},$$

where  $\rho = N/V$  is the number density.  $\Gamma$  is related to the familiar plasma parameter  $\Lambda$  by

$$\Lambda = [4\pi\rho(e^2/k_B T)^3]^{1/2} = \sqrt{3}\Gamma^{3/2}.$$

The Debye-Hückel limit is reached for  $\Gamma \ll 1$ , whereas at  $\Gamma \simeq 155$ , the OCP undergoes a fluid-solid phase transition.<sup>3</sup>

The computations in Ref. 1 covered the range  $1 \leq \Gamma \leq 155$ . The main findings were the following:

(a) The velocity autocorrelation function exhibits marked oscillations in time for  $\Gamma \gtrsim 10$ ; the period of these oscillations is close to  $2\pi/\omega_p$ , where  $\omega_p$  is the plasma frequency

$$\omega_p = [4\pi\rho(Ze)^2/m]^{1/2}$$

and  $m$  is the ionic mass. The resulting diffusion constant decreases faster than  $1/\Gamma$ .

(b) Near crystallization ( $\Gamma \simeq 155$ ), the spectrum of the transverse-current correlation function,  $C_t(q, \omega)$ , exhibits a well-defined peak at nonzero frequency, for reduced wave numbers  $q = \bar{v}k > 1$ ;

this peak is characteristic of propagating "shear modes." A memory-function analysis of these correlation functions yields an estimate of the reduced kinematic shear viscosity:

$$\eta^* = \eta/\rho m \omega_p \bar{v}^2. \quad (1)$$

(c) The spectrum of the longitudinal density (or charge) fluctuations, as measured by the dynamical structure factor  $S(q, \omega)$ , exhibits sharp "plasmon" peaks up to reduced wave numbers  $q$  of the order of 2. In the limit of small  $q$ , the dispersion relation of this collective mode was obtained from a simple moments analysis, as

$$\omega(q) = \omega_p(1 + \frac{1}{2}dq^2) + O(q^4), \quad (2)$$

where  $d$  is related to the coefficient of the  $q^4$  term in the small- $q$  expansion of the structure factor  $S(q)$ . Use of the compressibility sum rule<sup>4</sup> immediately allows  $d$  to be expressed in terms of the isothermal compressibility,  $K_T = (1/\rho)(\partial\rho/\partial P)_T$ , of the OCP:

$$d = \frac{1}{3\Gamma} \frac{K_T^0}{K_T}, \quad (3)$$

where  $K_T^0$  is the ideal-gas compressibility,  $K_T^0 = \beta/\rho$ .  $K_T$  is given explicitly as a function of  $\Gamma$  in Ref. 5, where it was found that the compressibility becomes negative beyond  $\Gamma \simeq 3$ . It follows that the dispersion  $d\omega/dq$ , calculated from (2), also becomes negative beyond  $\Gamma \simeq 3$ , in agreement with the negative shift of the plasmon peak position in  $S(q, \omega)$  obtained by the MD computations. This behavior contrasts with the predictions of mean-field theory. In particular, the collisionless Vlasov equation,<sup>6</sup> or the equivalent random-phase approximation<sup>7</sup> (RPA) predicts the following dispersion relation:

$$\omega(q) = \omega_p(1 + q^2/2\Gamma) + O(q^4). \quad (4)$$

Clearly the difference between (2) and (4) is due to the interionic collisions which are neglected in the mean-field approximation.

To characterize the importance of collisions, we estimate a mean time  $\tau$  between two collisions as follows. A crude effective cross section is calculated from the classical "distance of closest approach,"  $r_c = \beta e^2$ , yielding  $\sigma^2 \simeq (\beta e^2)^2$ ; from this, the mean free path  $\lambda \simeq 1/\rho\sigma$  and the relaxation time  $\tau \simeq \lambda/\langle v^2 \rangle^{1/2} \simeq \lambda\beta^{1/2}m^{1/2}$  can be derived. Finally, the product  $\omega_p\tau$  is obtained as

$$\omega_p\tau \simeq \Gamma^{-3/2}. \quad (5)$$

$\Gamma \ll 1$  corresponds to the "kinetic" regime, where the relaxation time  $\tau$  is much longer than the period of a plasma oscillation, and hence collisions do not sensibly alter the predictions of mean-field theory. The Landau<sup>8</sup> and Balescu-Lenard<sup>9</sup> equations are well-known examples of theories which generalize the Vlasov equation to include the effects of collisions in the kinetic regime. However, when  $\Gamma > 1$ , the relaxation time  $\tau$  becomes short compared to  $\omega_p^{-1}$ ; this means that many collisions take place during a plasma oscillation, and the kinetic theories cannot be expected to remain valid. This range of  $\Gamma$  values corresponds rather to the "hydrodynamic" regime of fluids, characterized by a time scale which is long compared to  $\tau$ .

The purpose of this paper is to study the density fluctuations of a very dense plasma ( $\Gamma > 1$ ) in the limit of large wavelengths. Because we are in the regime where each ion experiences many collisions during the period of a plasma oscillation, we shall assume local thermodynamic equilibrium.

In Sec. II we calculate the probability of appearance of a density fluctuation, and show that the fluctuation of the electric field ensures that the system is thermodynamically stable against such density fluctuations, even when the compressibility becomes negative.

Following closely the work of Mountain,<sup>10</sup> we obtain in Sec. III an explicit expression for the dynamical structure factor  $S(q, \omega)$  from the linearized hydrodynamic equations with proper inclusion of the electric field. The presence of the electric field completely modifies the structure of  $S(q, \omega)$ , in agreement with the MD results at small  $q$ .

Unknown transport coefficients (the shear and bulk viscosities  $\eta$  and  $\zeta$ , and the thermal conductivity  $\kappa$ ) appear in the hydrodynamic equations. In Secs. IV and V we calculate  $\eta$  and  $\zeta$ , using the "generalized hydrodynamics" formalism of Kadannoff and Martin<sup>11</sup> and the short-time expansions of the current-current correlation functions. The

reduced shear viscosity  $\eta^*$  is found to exhibit a minimum as a function of  $\Gamma$ , whereas the reduced bulk viscosity  $\zeta^*$  is found to be very small compared to  $\eta^*$ . In absolute units,  $\eta$  increases monotonically with density along any isotherm, whereas along any isochore,  $\eta$  exhibits a minimum as a function of temperature, not unlike the behavior in simple fluids.<sup>12</sup>

## II. THERMODYNAMIC FLUCTUATIONS AND STABILITY

We first generalize the standard thermodynamic fluctuation theory<sup>13</sup> to the case of the OCP, by taking into account the local electric field. Let  $\delta f(\vec{r})$  denote the local deviation of a thermodynamic quantity  $f$  from its equilibrium value  $f_0$ :

$$f(\vec{r}) = f_0 + \delta f(\vec{r}).$$

$\delta f(\vec{r})$  can be expressed in terms of its Fourier components  $\delta f_{\vec{k}}$ :

$$\delta f(\vec{r}) = \frac{1}{V} \sum_{\vec{k}} \delta f_{\vec{k}} \exp(i\vec{k} \cdot \vec{r}) \quad (6)$$

where

$$\delta f_{\vec{k}} = \int_V \delta f(\vec{r}) \exp(-i\vec{k} \cdot \vec{r}) d^3r. \quad (7)$$

The local thermodynamic quantities which will appear in the following are the entropy per ion  $s(\vec{r})$ , the number density  $\rho(\vec{r})$ , the internal energy per ion  $\epsilon(\vec{r})$ , the temperature  $T(\vec{r})$ , and the pressure  $P(\vec{r})$ .  $\vec{v}(\vec{r})$  will represent the local fluid velocity, and  $\vec{E}(\vec{r})$  the local electric field; the equilibrium values of these two quantities vanish.

If we choose  $\rho$  and  $T$  as the independent thermodynamic variables, the entropy per unit volume,  $\rho s$ , is, to second order in  $\delta\rho$  and  $\delta T$ ,

$$\begin{aligned} \rho s = \rho_0 s_0 + \frac{1}{T} \delta(\rho\epsilon) - \frac{\mu_0}{T_0} \delta\rho - \frac{1}{2} \frac{\rho_0 c_v}{T_0^2} (\delta T)^2 \\ - \frac{1}{2\rho_0 T_0} \left( \frac{\partial P_0}{\partial \rho_0} \right)_{T_0} (\delta\rho)^2, \end{aligned} \quad (8)$$

where  $\mu_0$  and  $c_v$  are the equilibrium values of the chemical potential and the specific heat at constant volume.  $\delta(\rho\epsilon)$  is the variation of the internal energy per unit volume; this variation corresponds to the transformation of internal energy into local kinetic energy and into potential energy associated with the local electric field:

$$\delta(\rho\epsilon) = -\left[ \frac{1}{2}\rho_0 m \vec{v}^2 + (1/8\pi)\vec{E}^2 \right]. \quad (9)$$

$\vec{E}(\vec{r})$  is related to  $\delta\rho(\vec{r})$  through Poisson's equation

$$\vec{\nabla} \cdot \vec{E}(\vec{r}) = 4\pi(Ze)\delta\rho(\vec{r}). \quad (10)$$

We now express the various functions in (8) in terms of their Fourier components (7), make use of Eqs. (9) and (10), and integrate both sides of

(8) over the volume  $V$  of the system; this yields

$$S = S_0 - \frac{\rho_0 m}{2T_0 V} \sum_{\vec{k}}' \vec{v}_{\vec{k}} \cdot \vec{v}_{-\vec{k}} - \frac{\rho_0 c_v}{2T_0^2 V} \sum_{\vec{k}}' \delta T_{\vec{k}} \delta T_{-\vec{k}} - \frac{1}{2T_0 V} \sum_{\vec{k}}' \left[ \frac{4\pi(Ze)^2}{k^2} + \frac{1}{\rho} \left( \frac{\partial P_0}{\partial \rho_0} \right)_{T_0} \right] \delta \rho_{\vec{k}} \delta \rho_{-\vec{k}}, \quad (11)$$

where  $S$  is the total entropy of the system,  $S = \int \rho s d^3r$ , and the primes indicate that the terms  $\vec{k} = 0$  are to be left out in the summations. The probability of having a fluctuation described by  $\vec{v}_{\vec{k}}$ ,  $\delta T_{\vec{k}}$ , and  $\delta \rho_{\vec{k}}$  is proportional to  $e^{S/k_B}$ .<sup>13</sup> It is seen from (11) that, to second order, the velocity, temperature, and density fluctuations are not coupled. For the system to be stable against these fluctuations, the coefficients of  $\vec{v}_{\vec{k}} \cdot \vec{v}_{-\vec{k}}$ ,  $|\delta T_{\vec{k}}|^2$ , and  $|\delta \rho_{\vec{k}}|^2$  must be positive. This implies the well-known condition  $c_v > 0$ , which is satisfied by the OCP for all  $\Gamma$ .<sup>5</sup> However, the usual condition that the compressibility must be positive is replaced in the case of the OCP by the condition

$$\frac{4\pi(Ze)^2}{k^2} + \frac{1}{\rho} \left( \frac{\partial P}{\partial \rho} \right)_T > 0.$$

Consequently, the system can sustain density fluctuations even when the compressibility becomes negative, i.e., for  $\Gamma \gtrsim 3$ . It is the local electric field created by the density fluctuation which contributes the term  $4\pi(Ze)^2/k^2$  and hence stabilizes the system, i.e., prevents the density fluctuation from building up indefinitely.

Expression (11) for the entropy allows us to calculate the long-wavelength limit of the structure factor,

$$S(k) = (1/N) \langle \delta \rho_{\vec{k}} \delta \rho_{-\vec{k}} \rangle,$$

the normalized average being calculated with the weight function  $e^{S/k_B}$ . The result is

$$S(k) = \frac{1}{4\pi(Ze)^2 \beta \rho / k^2 + \beta (\partial P / \partial \rho)_T},$$

i.e., with  $q = \bar{r}k$ ,

$$S(q) = \frac{q^2}{3\Gamma + q^2 (K_T^0 / K_T)}. \quad (12)$$

Obviously, (12) can only be expected to be valid in the small- $q$  (or hydrodynamic) limit, and must break down when the reduced wavelength  $\lambda = 2\pi/q$  becomes of the order of the interionic spacing. In fact,  $S(q)$  given by (12) has the correct small- $q$  expansion, to order  $q^4$ , provided the exact compressibility  $K_T$  is used.<sup>4,14</sup> Equation (12) also reduces to the Debye-Hückel limit when  $\Gamma \rightarrow 0$  and  $K_T^0 / K_T \rightarrow 1$ . However, for  $\Gamma \gtrsim 3$ ,  $S(q)$  has a singularity, and becomes negative for sufficiently large  $q$  because of the negative compressibility.

A direct comparison between the predictions of

(12) and the "exact" computer data<sup>5</sup> is made in Table I for  $\Gamma = 1$  and  $\Gamma = 155$ . The agreement is seen to be remarkable up to  $q \approx 2$ , which corresponds to a wavelength of the order of the interionic spacing! It should be noted that at this wave number the  $q^2$  and  $q^4$  terms are of the same order of magnitude. This remarkable agreement is another illustration of the predominantly collective behavior of the OCP over a wide range of wave numbers. Finally, it is interesting to note that Eq. (12) indicates a direct link between the change in sign of the compressibility, and the onset of "short-range order," i.e., the appearance of oscillations in the radial distribution function  $g(r)$ . Both phenomena happen at about the same value of  $\Gamma$  ( $\approx 3$ ), according to the computer-simulation results. And indeed, according to (12),  $S(q)$  has two poles on the imaginary axis as long as the compressibility stays positive, implying an exponentially decaying  $g(r)$ , whereas for  $\Gamma \approx 3$ , the negative compressibility entails two poles on the real axis for  $S(q)$ , i.e., an undamped oscillatory behavior of  $g(r)$ . Previous studies of the onset of short-range order in the OCP were based on diagrammatic techniques and predicted a somewhat lower value of  $\Gamma$  for the onset of oscillations in  $g(r)$  ( $\Gamma \approx 1$ ).<sup>15</sup> A very recent and more complete diagram summation predicts  $\Gamma = 2.61$  for the onset of oscillations,<sup>16</sup> in excellent agreement with our own findings.

### III. DYNAMICAL STRUCTURE FACTOR IN THE HYDRODYNAMIC LIMIT

Following the work of Mountain<sup>10</sup> for neutral fluids, we now derive the dynamical structure factor  $S(q, \omega)$  of the OCP from the linearized hydrodynamic equations and the assumption of local thermodynamic equilibrium. The three standard hydrodynamic equations express the conservation

TABLE I. Theoretical structure factor,  $S^{\text{th}}(q)$ , compared to the "exact"  $S(q)$ , for several values of the reduced wave number  $q = k\bar{r}$ , at  $\Gamma = 1$  and  $\Gamma = 155$ .

| $q = k\bar{r}$ | $\Gamma = 1, \beta \frac{\partial p}{\partial \rho} \Big _T = 0.7265$ |         | $\Gamma = 155, \beta \frac{\partial p}{\partial \rho} \Big _T = -59.82$ |          |
|----------------|-----------------------------------------------------------------------|---------|-------------------------------------------------------------------------|----------|
|                | $S^{\text{th}}(q)$                                                    | $S(q)$  | $S^{\text{th}}(q)$                                                      | $S(q)$   |
| 0.4            | 0.05134                                                               | 0.05136 | 0.000351                                                                | 0.000352 |
| 0.6            | 0.1104                                                                | 0.1104  | 0.000812                                                                | 0.000817 |
| 0.8            | 0.1847                                                                | 0.1840  | 0.001500                                                                | 0.001513 |
| 1.0            | 0.2683                                                                | 0.2678  | 0.002468                                                                | 0.002492 |
| 1.2            | 0.3559                                                                | 0.3540  | 0.003801                                                                | 0.003827 |
| 1.4            | 0.4430                                                                | 0.4386  | 0.005636                                                                | 0.005627 |
| 1.8            | 0.6052                                                                | 0.5908  | 0.01195                                                                 | 0.01133  |
| 2.2            | 0.7428                                                                | 0.7115  | 0.02758                                                                 | 0.02215  |
| 2.6            | 0.8545                                                                | 0.8007  | 0.11152                                                                 | 0.04483  |
| 3.0            | 0.9435                                                                | 0.8638  | ...                                                                     | 0.1003   |

of mass, momentum, and energy. We assume that these three equations remain valid in the case of the OCP, provided a term proportional to the local electric field is added in the momentum flux tensor of the Navier-Stokes equation. Extending the notations of Sec. II to the deviations at time  $t$  of the various quantities from their equilibrium value, the linearized equations read

$$\frac{\partial}{\partial t} \delta \rho(\vec{r}, t) + \rho_0 \vec{\nabla} \cdot \vec{v}(\vec{r}, t) = 0, \quad (13)$$

$$m \rho_0 \frac{\partial}{\partial t} \vec{v}(\vec{r}, t) = -\vec{\nabla} \delta P(\vec{r}, t) + \rho_0 (Ze) \vec{E}(\vec{r}, t) + \eta \Delta \vec{v}(\vec{r}, t) + (\zeta + \frac{1}{3} \eta) \vec{\nabla} [\vec{\nabla} \cdot \vec{v}(\vec{r}, t)], \quad (14)$$

$$\rho_0 T_0 \frac{\partial}{\partial t} \delta s(\vec{r}, t) = \kappa \vec{\nabla} \cdot [\vec{\nabla} \delta T(\vec{r}, t)]. \quad (15)$$

Here,  $\eta$  and  $\zeta$  are the shear and bulk viscosities, and  $\kappa$  is the thermal conductivity. Equations (13)–(15) must be supplemented by Eq. (10) relating  $\vec{E}(\vec{r}, t)$  to  $\delta \rho(\vec{r}, t)$ . Moreover, the electric field is purely longitudinal, since  $\text{rot} \vec{E} = 0$ .  $\vec{E}$  and  $\vec{v}$  are eliminated by taking the divergence of (14) and combining with (10) and (13).  $\delta P$  and  $\delta s$  are expressed in terms of  $\delta \rho$  and  $\delta T$  through the usual thermodynamic relations. The set of equations (13)–(15) then reduces to two coupled partial differential equations for  $\delta \rho$  and  $\delta T$ :

$$\left( \frac{\partial^2}{\partial t^2} + \omega_p^2 - b \frac{\partial}{\partial t} \Delta - \frac{\kappa}{\gamma} \Delta \right) \delta \rho(\vec{r}, t) = \frac{1}{m} \left( \frac{\partial P}{\partial T} \right)_\rho \Delta \delta T(\vec{r}, t), \quad (16)$$

$$\left( \frac{\partial}{\partial t} - a \Delta \right) \delta T(\vec{r}, t) = -\frac{T}{c_v} \left( \frac{\partial s}{\partial \rho} \right)_T \frac{\partial}{\partial t} \delta \rho(\vec{r}, t), \quad (17)$$

where we have dropped the subscript for the equilibrium values, and we have defined the following quantities:

$$\begin{aligned} & \left[ \left( \omega_p^2 - \omega^2 - i \omega b k^2 + \frac{K}{\gamma} k^2 \right) (a k^2 - i \omega) - i \omega k^2 \frac{\gamma - 1}{\gamma} K \right] \delta \bar{\rho}_{\vec{k}}(\omega) \\ & = \frac{k^2}{m} \left( \frac{\partial P}{\partial T} \right)_\rho \delta T_{\vec{k}} + i \rho (a k^2 - i \omega) \vec{k} \cdot \vec{v}_{\vec{k}} + \left( k^2 \frac{\gamma - 1}{\gamma} K + (a k^2 - i \omega) (b k^2 - i \omega) \right) \delta \rho_{\vec{k}}, \end{aligned} \quad (22)$$

where use was made of the thermodynamic identity

$$\frac{T}{m c_v} \left( \frac{\partial S}{\partial P} \right)_T \left( \frac{\partial P}{\partial T} \right)_\rho = -\frac{\gamma - 1}{\gamma} K.$$

We now multiply both sides of (22) by  $\delta \rho_{-\vec{k}}$  and take the average with the weight function  $e^{s/K_B}$  in order to obtain  $S(k, \omega)$ . Since  $\delta \rho_{\vec{k}}$ ,  $\delta T_{\vec{k}}$ , and  $\vec{v}_{\vec{k}}$

$$a = \frac{\kappa}{\rho c_v}, \quad b = \frac{\zeta + 4\eta/3}{m \rho}, \quad (18)$$

$$\gamma = \frac{c_p}{c_v}, \quad K = \frac{1}{m} \left( \frac{\partial P}{\partial \rho} \right)_s = \frac{1}{m \rho K_s}.$$

Equation (16) differs from the corresponding equation for neutral fluids only by the term  $\omega_p^2$  stemming from the local electric field. The presence of the plasma frequency will completely modify the structure of  $S(k, \omega)$ , as compared to the neutral fluid case.

The time-dependent density-density correlation function is defined as

$$F(k, t) = (1/N) \langle \delta \rho_{\vec{k}}(t) \delta \rho_{-\vec{k}}(0) \rangle, \quad (19)$$

where

$$\delta \rho_{\vec{k}}(t) = \int \exp(-i \vec{k} \cdot \vec{r}) \delta \rho(\vec{r}, t). \quad (20)$$

The dynamical structure factor  $S(k, \omega)$  is given by the Fourier transform of  $F(k, t)$ , or, since this function is even in  $t$ , by twice the real part of its Laplace transform

$$S(k, \omega) = \int_{-\infty}^{+\infty} F(k, t) e^{i \omega t} dt = 2R\bar{S}(k, \omega)$$

with

$$\begin{aligned} \bar{S}(k, \omega) &= \int_0^{\infty} e^{i \omega t} F(k, t) dt \\ &= (1/N) \langle \delta \bar{\rho}_{\vec{k}}(\omega) \delta \rho_{-\vec{k}} \rangle, \end{aligned} \quad (21)$$

where  $\delta \bar{\rho}_{\vec{k}}(\omega)$  is the Laplace transform of (20). Taking Fourier-Laplace transforms of (16) and (17), we obtain algebraic equations relating  $\delta \bar{\rho}_{\vec{k}}(\omega)$  and  $\delta \bar{T}_{\vec{k}}(\omega)$  to  $\delta \rho_{\vec{k}}$ ,  $\delta T_{\vec{k}}$ , and  $\vec{v}_{\vec{k}}$ , which are the values at time  $t=0$  of  $\delta \rho_{\vec{k}}(t)$ ,  $\delta T_{\vec{k}}(t)$ , and  $\vec{v}_{\vec{k}}(t)$ . Eliminating  $\delta \bar{T}_{\vec{k}}(\omega)$ , we obtain for  $\delta \bar{\rho}_{\vec{k}}(\omega)$  the relation

are statistically independent, the two first terms on the right-hand side of (22) will not contribute to  $S(k, \omega)$ . Defining  $\bar{\sigma}(k, \omega)$  by the relation

$$\bar{S}(k, \omega) = S(k) \bar{\sigma}(k, \omega), \quad (23)$$

we finally obtain  $\bar{\sigma}(k, \omega)$  as a rational fraction, with real coefficients, in the variable  $s = i \omega$ :

$$\bar{\sigma}(k, \omega) = \frac{s^2 + (a+b)k^2s + abk^4 + k^2(\gamma-1)\gamma^{-1}K}{s^3 + (a+b)k^2s^2 + (\omega_p^2 + Kk^2 + abk^4)s + ak^2(\omega_p^2 + \gamma^{-1}Kk^2)}. \quad (24)$$

In terms of the roots of the denominator,  $\bar{\sigma}(k, \omega)$  can be rewritten as

$$\bar{\sigma}(k, \omega) = \frac{A}{s-\alpha} + \frac{B}{s-\beta} + \frac{B^*}{s-\beta^*}. \quad (25)$$

The roots are easily determined to order  $k^2$ :

$$\alpha = -ak^2, \quad \beta = -\frac{1}{2}bk^2 + i\omega_p[1 + (K/2\omega_p^2)k^2]. \quad (26)$$

It is interesting to compare these results with those obtained for a neutral fluid<sup>10</sup>; in the latter case, one finds

$$\alpha = -(a/\gamma)k^2, \quad \beta = -\frac{1}{2}(b+a-a/\gamma)k^2 + i\sqrt{K}k. \quad (27)$$

The first root, which gives rise to the thermal Rayleigh peak, only differs by a factor  $\gamma$  in the OCP case. On the other hand, the two roots  $\beta$  and  $\beta^*$ , which give rise to the plasmon or Brillouin peaks, are essentially different in two respects. The first difference lies in the imaginary part of the roots, which determines the oscillation frequency. In the neutral-fluid case, we have a sound wave the frequency of which goes to zero as  $k \rightarrow 0$ ; the corresponding group velocity, equal to  $\sqrt{K}$ , is independent of  $k$  in that limit. In contrast to this, the frequency stays finite ( $\omega \rightarrow \omega_p$ ) as  $k \rightarrow 0$  in the OCP case; the group velocity is linear in  $k$ , and is thus practically zero for small  $k$ . The second important difference lies in the real part of the roots, which governs the damping of the oscillations (remember that  $s = i\omega$ ). In the neutral case the sound damping is due both to thermal conduction and to viscosity, whereas in the OCP case the plasmon damping is only due to the viscosity; to order  $k^2$  there is no coupling between thermal and mechanical modes.

It is also interesting to compare the relative importance of the Rayleigh and plasmon or Brillouin peaks. In the OCP case, we find for  $A$  and  $B$  in (25)

$$A = \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2}, \quad (28)$$

$$B = \frac{1}{2} \left( 1 - \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2} \right) - ib \frac{k^2}{4\omega_p},$$

whereas for the neutral fluid

$$A' = \frac{\gamma-1}{\gamma}, \quad B' = \frac{1}{2\gamma}. \quad (29)$$

Finally, we obtain for the dynamical structure factor the expression

$$S(k, \omega) = S(k)\sigma(k, \omega),$$

$$\sigma(k, \omega) = \frac{-2A\alpha}{\omega^2 + \alpha^2} - 2 \frac{B_1\beta_1 + B_2(\omega + \beta_2)}{(\omega + \beta_2)^2 + \beta_1^2} - 2 \frac{B_1\beta_1 - B_2(\omega - \beta_2)}{(\omega - \beta_2)^2 + \beta_1^2}. \quad (30)$$

The Rayleigh peak is a Lorentzian centered around  $\omega = 0$ , and of height  $2[(\gamma-1)/\gamma]K/a\omega_p^2$ , which is independent of  $k$ . The "plasmon doublet" is the superposition of two pseudo-Lorentzian curves centered around  $\omega = \pm\omega_p(1 + Kk^2/2\omega_p^2)$ , of height

$$\frac{2}{bk^2} \left( 1 - \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2} \right),$$

which is proportional to  $1/k^2$  in the long-wavelength limit. The total integrated intensities of the Rayleigh peak and of the plasmon doublet are, to order  $k^2$ ,

$$\frac{I_R}{S(k)} = \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2}, \quad (31)$$

$$\frac{I_P}{S(k)} = 1 - \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2}.$$

Hence the equivalent of the Landau-Placzek ratio,<sup>17</sup> which in the neutral fluid case is  $\gamma-1$ , is in the OCP case

$$\frac{I_R}{I_P} = \frac{\gamma-1}{\gamma} \frac{Kk^2}{\omega_p^2} + O(k^4). \quad (32)$$

In the limit  $k \rightarrow 0$  this ratio vanishes, showing the clear predominance of the "mechanical" plasmon mode over the thermal mode in the long-wavelength limit. In this limit (30) reduces to

$$\lim_{k \rightarrow 0} \sigma(k, \omega) = \pi[\delta(\omega + \omega_p) + \delta(\omega - \omega_p)].$$

These results are in qualitative agreement with the MD data<sup>1</sup> which showed no trace of a Rayleigh peak for the smallest accessible wave numbers. It should be noted that  $(\gamma-1)/\gamma$  and  $K$  are always of the same sign, and, in particular, change sign for the same value of  $\Gamma$  ( $\approx 3$ ).<sup>5</sup>

Finally, it is interesting to compare the dispersion relation (2), obtained from the exact moments of  $S(q, \omega)$ ,<sup>1</sup> with the relation derived here from the hydrodynamic equations,

$$\omega(q) = \omega_p(1 + \frac{1}{2}\gamma dq^2) + O(q^4), \quad (33)$$

where  $d$  is given by (3). The two dispersion relations (2) and (33) are seen to differ by a factor  $\gamma = c_p/c_v$  in the  $q^2$  term. However, for  $\Gamma \gtrsim 20$ ,  $\gamma$  differs from 1 only by a few percent, and the dif-

ference decreases with increasing  $\Gamma$ ; e.g., for  $\Gamma=40$  and  $100$ ,  $\gamma=0.95$  and  $0.98$ , respectively. Consequently, the two dispersion relations are practically identical for large  $\Gamma$  values.

In the  $\Gamma \rightarrow 0$  limit,  $\gamma$  and  $d$  can be calculated in the Debye-Hückel limit:

$$d \simeq \frac{1}{3\Gamma} \left( 1 - \frac{\sqrt{3}}{4} \Gamma^{3/2} \right) \quad \text{and} \quad \gamma \simeq \frac{5}{3} + \frac{\Gamma^{3/2}}{2\sqrt{3}}.$$

Retaining only the dominant ideal-gas terms, (33) reduces to

$$\omega(q) \simeq \omega_p \left( 1 + \frac{1}{2} \frac{5}{3} \frac{q^2}{3\Gamma} \right),$$

which differs from the Vlasov dispersion relation (4) by a factor  $\frac{1}{3}\gamma$ . This difference clearly illustrates the limits of the hydrodynamic dispersion relation: in the  $\Gamma \rightarrow 0$  limit, the relaxation time  $\tau$  is very long compared to the period of the plasma oscillation, and the fundamental hypothesis of local thermal equilibrium is no longer valid. In this limit, a kinetic theory taking into account the local anisotropy of the velocity distribution function is expected to be more adequate.

#### IV. TRANSVERSE CURRENT CORRELATIONS AND SHEAR VISCOSITY

In this section we shall apply the generalized hydrodynamics formalism of Kadanoff and Martin<sup>11</sup> to the OCP in order to determine the coefficient of shear viscosity from the knowledge of the short-time expansion of the transverse-current correlation function. A similar calculation has been made by Forster *et al.* in the case of a neutral fluid (argon).<sup>18</sup> Using notations similar to those in Ref. 1, we define the current operator

$$j_{\vec{k}}(t) = \sum_{i=1}^N \vec{v}_i(t) \exp[i\vec{k} \cdot \vec{r}_i(t)] \quad (34)$$

and the transverse-current correlation function

$$C_t(\vec{k}, t) = (1/2N) \text{Tr} \langle [i\vec{k} \wedge \vec{j}_{\vec{k}}(t)] \cdot [\vec{k} \wedge \vec{j}_{-\vec{k}}(0)] \rangle.$$

In the hydrodynamic limit,  $j_{\vec{k}}(t) = \rho \vec{v}_{\vec{k}}(t)$ , which satisfies the projection perpendicular to  $\vec{k}$  of the Fourier transform of Eq. (14); the Laplace transform of the corresponding correlation function is

$$\tilde{C}_t(k, \omega) = \frac{\omega_0^2}{-i\omega + \eta k^2/m\rho}, \quad (35)$$

where  $\omega_0^2 = k^2/\beta m$ . The shear viscosity  $\eta$  is then obtained through the familiar Kubo relation<sup>11,18</sup>:

$$\eta = \lim_{\omega \rightarrow 0} \lim_{k \rightarrow 0} \frac{m\rho}{k^2} \left( i\omega + \frac{\omega_0^2}{\tilde{C}_t(k, \omega)} \right). \quad (36)$$

We proceed by writing the rate of decay of  $C_t(k, t)$  as

$$\frac{\partial}{\partial t} C_t(k, t) + \int_0^t dt' M_t(k, t-t') C_t(k, t') = 0, \quad (37)$$

which defines the memory function  $M_t$ ; taking the Laplace transform of (37), we obtain

$$\tilde{M}_t(k, \omega) = i\omega + \frac{\omega_0^2}{\tilde{C}_t(k, \omega)}. \quad (38)$$

$\tilde{C}_t(k, \omega)$ , and hence  $\tilde{M}_t(k, \omega)$ , are related to the transverse-current-response function through the fluctuation-dissipation theorem.<sup>19</sup> From this relation, a certain number of important properties of  $\tilde{M}_t(k, \omega)$  can be derived.<sup>11</sup> In particular,  $\tilde{M}_t(k, \omega)$  is an analytic function of  $\omega = \omega_1 + i\omega_2$  in the complex upper half-plane ( $\omega_2 \geq 0$ ), and its real part  $\tilde{M}_{t1}$  is positive for real  $\omega$  ( $\omega_2 = 0$ ). The asymptotic behavior of  $\tilde{M}_t(k, \omega)$  is governed by the short-time behavior of  $M_t(k, t)$ , and hence of  $C_t(k, t)$ , which can be expanded in powers of  $t^2$ :

$$C_t(k, t) = \omega_0^2 \sum_{p=0}^{\infty} (-1)^p \omega_{pt}^{2p} \frac{t^{2p}}{(2p)!}. \quad (39)$$

The expressions of the three lowest moments  $\omega_{pt}$  in terms of the equilibrium pair and triplet distribution functions  $g$  and  $g_3$  have already been calculated in Ref. 1:

$$\omega_{0t} = 1, \quad \omega_{1t}^2 = \omega_0^2 - \omega_p^2 G_{5/2}(q), \quad (40)$$

$$\begin{aligned} \omega_{2t}^4 = & 3\omega_0^4 + \frac{17}{15} \omega_0^2 \omega_p^2 - 18\omega_0^2 \omega_p^2 G_{7/2}(q) + \omega_p^4 [G_{5/2}(q)]^2 + \frac{2}{3} \omega_p^4 r^3 \int_0^\infty \frac{dr}{r^4} \left( 2 - \frac{\sin kr}{kr} + \frac{3 \cos kr}{k^2 r^2} - \frac{3 \sin kr}{k^3 r^3} \right) \\ & + \frac{\omega_p^4}{2(4\pi)^2} \int \frac{d^3 r}{r^3} \int \frac{d^3 r'}{r'^3} [g_3(\vec{r}, \vec{r}') - g(r)g(r')] [9\vec{r}_0 \cdot \vec{r}'_0 (\vec{r}_0 \cdot \vec{r}'_0 - z_0 z'_0) + 3z_0^2 + 3z_0'^2 - 4] \\ & \times (e^{-ik(z-z')} + 1 - e^{-ikz} - e^{ikhz'}). \end{aligned} \quad (41)$$

In (40) and (41),  $q = k\bar{r}$ , and  $G_\nu(q)$  represents the following integral over  $g(r)$ :

$$G_\nu(q) = \left( \frac{\pi}{2} \right)^{1/2} q^2 \int_0^\infty x dx [g(x) - 1] \frac{J_\nu(qx)}{(qx)^\nu},$$

where  $J_\nu(qx)$  is a Bessel function of the first kind, and  $x=r/\bar{r}$ . In (41),  $\bar{r}_0$  and  $z_0$  are the unit vector and its  $z$  component in the direction of  $\bar{r}$ . Note that the term (+1) in the last set of parentheses of (41) is missing in formulas (A1) and (A2) of Ref. 1.

From (39), we obtain the following asymptotic expansion for  $\bar{M}_t(k, \omega)$ :

$$\bar{M}_t(k, \omega) \approx i\omega \left( \frac{\omega_{1t}^2}{\omega^2} + \frac{\omega_{2t}^4 - \omega_{1t}^4}{\omega^4} + \dots \right). \quad (42)$$

Now, since  $\bar{M}_t(k, \omega)$  is analytic in the complex upper half-plane,  $\bar{M}_t(k, \omega)$  and its real part  $\bar{M}_{t1}(k, \omega)$  are related by a Kramers-Kronig relation. It follows that the coefficients in expansion (42) are related to the moments of  $\bar{M}_{t1}(k, \omega)$ :

$$\frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega \bar{M}_{t1}(k, \omega) = \omega_{1t}^2, \quad (43)$$

$$\frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega \omega^2 \bar{M}_{t1}(k, \omega) = \omega_{2t}^4 - \omega_{1t}^4. \quad (44)$$

Since  $\bar{M}_{t1}(k, \omega)$  is a positive, even function for real  $\omega$ , and since all its moments exist, it has an exponential character, and it is reasonable to approximate the function by a Gaussian which is entirely determined by the first two moments (43) and (44). In terms of these moments,  $\bar{M}_{t1}(k, \omega)$  reads

$$\bar{M}_{t1}(k, \omega) = \left( \frac{\pi}{2} \right)^{1/2} \frac{\omega_{1t}^3}{(\omega_{2t}^4 - \omega_{1t}^4)^{1/2}} \exp\left(-\frac{\omega_{1t}^2}{\omega_{2t}^4 - \omega_{1t}^4} \frac{\omega^2}{2}\right). \quad (45)$$

In order to determine  $\eta$  through formula (36), we need only the small- $q$  limit of the moments  $\omega_{1t}^2$  and  $\omega_{2t}^4$ . Expanding (40) and (41) to order  $q^2$ , we obtain

$$\omega_{1t}^2 \approx \frac{q^2 \omega_p^2}{15} \left( \frac{5}{\Gamma} - I_1 \right), \quad (46)$$

$$\omega_{2t}^4 \approx \frac{q^2 \omega_p^4}{45} \left( \frac{17}{\Gamma} + 8I_{-2} + \frac{81}{10}H_3 + \frac{39}{10}H_1 \right), \quad (47)$$

where

$$I_n = \int_0^\infty x^n [g(x) - 1] dx, \quad n > -1 \quad (48)$$

$$I_n = \int_0^\infty x^n g(x) dx, \quad n < -1$$

$$H_n = \int_0^\infty dx \int_0^\infty dx' \int_{-1}^{+1} d(\cos \alpha) P_n(\cos \alpha) \times [g_3(r, r', \alpha) - g(r)g(r')]. \quad (49)$$

$P_n$  is the Legendre polynomial of order  $n$ . Substituting (45), (46), and (47) into (36), we obtain the reduced kinematic viscosity:

$$\begin{aligned} \eta^* &= \frac{\eta}{m\rho\omega_p\bar{r}^2} \\ &= \frac{1}{15} \left( \frac{3\pi}{2} \right)^{1/2} \frac{(5/\Gamma - I_1)^{3/2}}{(17/\Gamma + 8I_{-2} + \frac{81}{10}H_3 + \frac{39}{10}H_1)^{1/2}}. \end{aligned} \quad (50)$$

Formula (50) expresses  $\eta^*$  in terms of the integrals (48) and (49), which can be calculated for each value of  $\Gamma$  from the equilibrium pair and triplet distribution functions.  $I_1$  is simply related to the excess internal energy per particle<sup>5</sup>

$$I_1 = \frac{2}{3\Gamma} \frac{U}{Nk_B T}.$$

$I_{-2}$  has been calculated using the radial distribution functions of Ref. 5. For large  $\Gamma$ ,  $I_{-2}$  is very well approximated by the corresponding bcc lattice sum:

$$I_{-2}^\infty = 0.788466.$$

The three-body integrals (49) are more difficult to calculate. Fortunately,  $H_1$  can be related to  $I_{-2}$  through the Born-Green-Yvon hierarchy linking  $g$  to  $g_3$ . The relation is

$$H_1 = \frac{2}{3}(1/\Gamma - I_{-2}), \quad (51)$$

and is only valid in the case of the OCP. Unfortunately, no such relation exists for  $H_3$ , which we have estimated using the superposition approximation (SA) for  $g_3$ . This was also done for  $H_1$ , so that comparison with the result from (51) gave us an estimate of the error due to the SA. We found that  $H_1$  and  $H_3$  are both slowly varying functions of  $\Gamma$ , and that the error on  $H_1$  due to the SA does not exceed 4%. In the perfect-lattice limit ( $\Gamma \rightarrow \infty$ ), the cubic symmetry implies that  $H_1$  and  $H_3$  are equal, and

$$H_1^\infty = H_3^\infty = -\frac{2}{3}I_{-2}^\infty = -0.525644.$$

The results for the various integrals are shown, as a function of  $\Gamma$ , in Table II. In Table III we give the results for  $\eta^*$ . The values listed have been calculated with the "exact"  $H_1$ , obtained from (51). Unfortunately, at high  $\Gamma$ , the values of  $\eta^*$  are very sensitive to small variations in  $H_3$ . For this reason we have also tabulated the values obtained by taking into account the estimated uncertainty on  $H_3$ . Finally, we indicate the value of  $\eta^*$  obtained when all the integrals are replaced by the corresponding lattice sums ( $\Gamma \rightarrow \infty$ ). The most striking feature which can be seen from Table III is the fact that  $\eta^*$  exhibits a *minimum* as a function of  $\Gamma$ ;  $\eta^*$  decreases rapidly for  $\Gamma < 10$ , goes through a minimum around  $\Gamma = 20$ , and then increases slowly with  $\Gamma$ . This general feature is independent of the uncertainty affecting the value of  $H_3$ . The situation, including the error bars,

TABLE II. Various thermodynamic quantities and integrals appearing in expressions (50) and (63) of  $\eta^*$  and  $b^*$ , as a function of  $\Gamma$ . The integrals  $I_1$  and  $I_2$  are defined by (48).  $H_1$  is calculated, using the exact relation (51). The two last columns list the integrals  $H_1$  and  $H_3$ , defined by (49), and are calculated with the help of the superposition approximation. The last line lists the bcc lattice sums corresponding to the various integrals.

| $\Gamma$ | $\beta \frac{\partial p}{\partial \rho} \Big _T$ | $\gamma - 1$       | $I_1$     | $I_2$     | $H_1$   | $H_1$<br>Super. | $H_3$<br>Super. |
|----------|--------------------------------------------------|--------------------|-----------|-----------|---------|-----------------|-----------------|
| 1        | 0.726                                            | 0.919              | -0.3879   | 1.814     | -0.543  | -0.529          | -0.315          |
| 2        | 0.384                                            | 1.743              | -0.4393   | 1.3496    | -0.5664 | -0.550          | -0.363          |
| 4        | -0.339                                           | -1.986             | -0.4877   | 1.108     | -0.5720 | -0.554          | -0.407          |
| 10       | -2.611                                           | -0.2624            | -0.5331   | 0.9491    | -0.5661 | -0.547          | -0.458          |
| 20       | -6.493                                           | -0.10775           | -0.55557  | 0.8866    | -0.5577 | -0.543          | -0.491          |
| 40       | -14.340                                          | -0.050026          | -0.57053  | 0.8494    | -0.5496 | -0.543          | -0.515          |
| 70       | -26.175                                          | -0.028051          | -0.57911  | 0.8291    | -0.5432 | -0.548          | -0.526          |
| 100      | -38.037                                          | -0.019620          | -0.58320  | 0.8200    | -0.5400 | -0.550          | -0.530          |
| 120      | -45.954                                          | -0.016385          | -0.58491  | 0.8160    | -0.5384 | -0.554          | -0.528          |
| 140      | -53.875                                          | -0.014086          | -0.58617  | 0.8132    | -0.5374 | -0.556          | -0.525          |
| 160      | -61.800                                          | -0.012367          | -0.58704  | 0.8110    | -0.5365 | -0.560          | -0.528          |
| $\infty$ | -0.398191 $\Gamma$                               | -1.67424/ $\Gamma$ | -0.597286 | 0.7884666 |         | -0.5256444      |                 |

is pictured in Fig. 1. The MD computations of  $C_t(q, t)$  at finite values of  $q$  yielded the estimate  $\eta^* \approx 0.25$  near the fluid-solid transition.<sup>1</sup> Bearing in mind the large uncertainties of both calculations at high  $\Gamma$ , this is in semiquantitative agreement with the present value,  $\eta^* \approx 0.14$ . Note that if the perfect-lattice values of the various integrals are used, we find  $\eta^* = 0.22$ , which is remarkably close to the MD estimate at  $\Gamma \approx 155$ , although the agreement may just be accidental.

#### V. LONGITUDINAL CURRENT CORRELATION FUNCTION AND BULK VISCOSITY

We now present a calculation of the bulk viscosity, which is in all points similar to the treatment of the shear viscosity in the previous sec-

tion. We start from the longitudinal current correlation function

$$C_t(k, t) = (1/N) \langle [\vec{k} \cdot \vec{j}_{\vec{k}}(t)] [\vec{k} \cdot \vec{j}_{-\vec{k}}(0)] \rangle. \quad (52)$$

Its Laplace transform is directly related to the Laplace transform (21) of the density-density correlation function by

$$\tilde{C}_t(k, \omega) = -i\omega[S(k) + i\omega\tilde{S}(k, \omega)]. \quad (53)$$

Introducing the associated memory function  $\tilde{M}_t(k, \omega)$ , it follows from (24) that, in the hydrodynamic limit,

$$\tilde{M}_t(k, \omega) = \frac{\omega_p^2 + K\gamma^{-1}k^2}{-i\omega} + bk^2 + \frac{\gamma - 1}{\gamma} \frac{Kk^2}{-i\omega + ak^2}. \quad (54)$$

TABLE III. Reduced viscosities  $b^*$  and  $\eta^*$  as a function of  $\Gamma$ , calculated with the help of the superposition approximation for the three-body integral  $H_3$ . Columns 2, 3, 5, and 6 list the upper and lower limits of  $b^*$  and  $\eta^*$ , if the estimated uncertainty on the values of  $H_3$  is taken into account.

| $\Gamma$ | $b^*$        |                    |                    | $\eta^*$     |                    |                    |
|----------|--------------|--------------------|--------------------|--------------|--------------------|--------------------|
|          | $H_3$ super. | $H_3$ super. -0.02 | $H_3$ super. +0.02 | $H_3$ super. | $H_3$ super. -0.02 | $H_3$ super. +0.02 |
| 1        | 0.462        | 0.463              | 0.460              | 0.349        | 0.350              | 0.348              |
| 2        | 0.260        | 0.261              | 0.258              | 0.194        | 0.195              | 0.193              |
| 4        | 0.161        | 0.163              | 0.159              | 0.120        | 0.1215             | 0.119              |
| 10       | 0.1105       | 0.113              | 0.108              | 0.1827       | 0.0847             | 0.0808             |
| 20       | 0.104        | 0.109              | 0.100              | 0.0781       | 0.0821             | 0.0750             |
| 40       | 0.117        | 0.129              | 0.108              | 0.0881       | 0.0972             | 0.0812             |
| 70       | 0.142        | 0.172              | 0.124              | 0.1075       | 0.131              | 0.0934             |
| 100      | 0.159        | 0.211              | 0.133              | 0.121        | 0.162              | 0.1005             |
| 120      | 0.170        | 0.2405             | 0.138              | 0.129        | 0.1855             | 0.104              |
| 140      | 0.179        | 0.274              | 0.143              | 0.136        | 0.213              | 0.108              |
| 160      | 0.188        | 0.315              | 0.147              | 0.1425       | 0.246              | 0.1105             |
| $\infty$ |              | 0.335              |                    |              | 0.221              |                    |



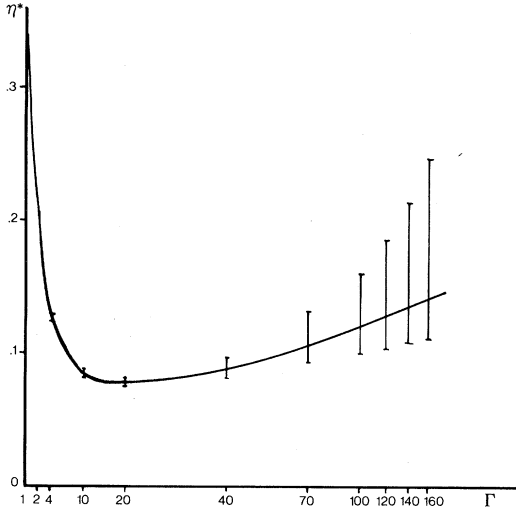


FIG. 1. Reduced shear viscosity  $\eta^*$  as a function of  $\Gamma$ . The error bars reflect the uncertainties affecting the values of the three-body integral  $H_3$  calculated with the superposition approximation.

$\tilde{M}_l(k, \omega)$  has simple poles at  $\omega = 0$  and  $\omega = -iak^2$ . In the limit  $k \rightarrow 0$ , the second pole contributes to the

residue of  $\tilde{M}_l(k, \omega)$  at  $\omega = 0$ , which is hence equal to  $i(\omega_p^2 + Kk^2)$  in this limit. This residue must be included in the Kramers-Kronig relation between  $\tilde{M}_l(k, \omega)$  and its real part,  $\tilde{M}_{1l}(k, \omega)$ , in the limit of small  $k$ . Note that the contribution of the last term in  $\tilde{M}_l(k, \omega)$  is small at large  $\Gamma$ , since  $\gamma$  is then close to 1.<sup>5</sup>

From (54) we immediately derive the Kubo relation for  $b$ :

$$b^* = \frac{b}{\omega_p \bar{r}^2} = \lim_{\omega \rightarrow 0} \lim_{k \rightarrow 0} \frac{\tilde{M}_{1l}(k, \omega)}{\omega_p \bar{r}^2 k^2}. \quad (55)$$

Except for the simple pole at  $\omega = 0$ ,  $\tilde{M}_l(k, \omega)$  has the same properties as  $\tilde{M}_l(k, \omega)$ . Similarly,  $\tilde{M}_{1l}(k, \omega)$  is a positive, even function of  $\omega$  which has all its moments, and hence is exponential in character. Consequently, it seems again reasonable to choose a simple Gaussian form satisfying the two lowest-moment sum rules:

$$\frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega' \tilde{M}_{1l}(k, \omega') = \omega_{1l}^2 - (\omega_p^2 + Kk^2), \quad (56)$$

$$\frac{1}{\pi} \int_{-\infty}^{+\infty} d\omega' \omega'^2 \tilde{M}_{1l}(k, \omega') = \omega_{2l}^4 - \omega_{1l}^4, \quad (57)$$

with

$$\omega_{1l}^2 = \omega_p^2 + 3\omega_0^2 + 2\omega_p^2 G_{5/2}(q), \quad (58)$$

$$\begin{aligned} \omega_{2l}^4 &= \omega_p^4 + 15\omega_0^2 + \frac{37}{5}\omega_0^2\omega_p^2 + 36\omega_0^2\omega_p^2 G_{7/2}(q) + 4\omega_p^4 G_{5/2}(q) + 4\omega_p^4 [G_{5/2}(q)]^2 \\ &+ \frac{4}{3}\omega_p^4 \bar{r}^3 \int_0^\infty dr \frac{g(r)}{r^4} \left( 1 - \frac{2 \sin kr}{kr} - \frac{3 \cos kr}{(kr)^2} + \frac{3 \sin kr}{(kr)^3} \right) \\ &+ \frac{\omega_p^4}{(4\pi)^2} \int \frac{d^3 r}{r^3} \int \frac{d^3 r'}{r'^3} [\mathcal{G}_3(\vec{r}, \vec{r}') - g(r)g(r')] (9\vec{r}_0 \cdot \vec{r}'_0 z_0 z'_0 - 3z_0^2 - 3z_0'^2 + 1)(e^{-ik(z-z')} + 1 - e^{-ikz} - e^{ikz'}). \end{aligned} \quad (59)$$

The Gaussian memory function satisfying (56) and (57) is

$$\begin{aligned} \tilde{M}_{1l}(g, \omega) &= \left( \frac{\pi}{2} \right)^{1/2} \frac{\omega_{1l}^2 - \omega_p^2(1 + \gamma dq^2)}{(\omega_{2l}^4 - \omega_{1l}^4)^{1/2}} \\ &\times \exp\left( -\frac{\omega_{1l}^2 - \omega_p^2(1 + \gamma dq^2)}{\omega_{2l}^4 - \omega_{1l}^4} \frac{\omega^2}{2} \right), \end{aligned} \quad (60)$$

where  $d$  is related to the inverse compressibility through Eq. (3). Expansion of the moments (58) and (59) to order  $q^2$  yields

$$\omega_{1l}^2 \approx \omega_p^2 \left[ 1 + \frac{1}{15} q^2 (15/\Gamma + 2I_1) \right], \quad (61)$$

$$\omega_{2l}^4 \approx \omega_p^4 \left[ 1 + \frac{1}{15} q^2 \left( 37/\Gamma + \frac{14}{3} I_{-2} + 4I_1 + \frac{18}{5} H_3 + \frac{17}{5} H_1 \right) \right]. \quad (62)$$

From (55), using (60), (61), and (62) we obtain the final expression for  $b^*$ :

$$\begin{aligned} b^* &= \frac{\xi + \frac{4}{3}\eta}{m\rho\omega_p \bar{r}^2} \\ &= \frac{1}{15} \left( \frac{\pi}{2} \right)^{1/2} \frac{(15/\Gamma + 2I_1 - 15\gamma d)^{3/2}}{(7/\Gamma + \frac{14}{3} I_{-2} + \frac{18}{5} H_3 + \frac{17}{5} H_1)^{1/2}}. \end{aligned} \quad (63)$$

Equation (63), similar to (50), expresses  $b^*$  in terms of the same integrals  $I_n$  and  $H_n$ , which are tabulated in Table II, together with the values of  $d$  and  $\gamma - 1$  taken from Ref. 5. The values obtained for  $b^*$  are given in Table III. Again,  $b^*$  is very sensitive to the uncertainties affecting  $H_3$  at large  $\Gamma$ , due to the SA. Nevertheless,  $b^*$ , like  $\eta^*$ , clearly exhibits a minimum around  $\Gamma = 20$ .

The most interesting result is the ratio  $b^*/\eta^*$ . We find that, contrarily to  $b^*$  and  $\eta^*$  taken separately, this ratio is rather insensitive to the uncertainty on  $H_3$ . The ratio is, moreover, practic-

ally independent of  $\Gamma$ . Within the small uncertainties due to  $H_3$ , we find that  $b^*/\eta^* \simeq \frac{4}{3}$  for all  $\Gamma$ . Since  $\eta$  and  $\zeta$  are non-negative, the ratio can only be  $\geq \frac{4}{3}$ . Hence our calculations indicate that the bulk viscosity  $\zeta$  is negligible compared to the shear viscosity  $\eta$  for all  $\Gamma$ .

#### VI. CONCLUSION

Starting from the usual conservation equations of macroscopic hydrodynamics and from the thermodynamic theory of fluctuations, we have obtained explicit expressions for the static and dynamical structure factors of the OCP in a uniform background, valid in the long-wavelength limit. The most important result is the disappearance of the thermal Rayleigh peak in that limit, in qualitative agreement with the MD results for small but finite  $q$ . The usual Brillouin doublet of ordinary fluids is replaced by a plasmon doublet; the corresponding dispersion  $d\omega/dq$  becomes negative for  $\Gamma \geq 3$ , due to the negative compressibility. We have shown that the fluctuations of the local electric field renders the OCP stable against density fluctuations in the range where the compressibility becomes negative.

Using a Gaussian approximation for the real part of the Laplace transform of the memory functions associated with the transverse and longitudinal current correlation functions, we have obtained explicit expressions for the reduced shear and bulk viscosity coefficients in terms of integrals involving the equilibrium pair and triplet distribution functions. The resulting  $\eta^*$  and  $b^* = \zeta^* + \frac{4}{3}\eta^*$  plotted versus  $\Gamma$  exhibit a minimum around  $\Gamma = 20$ .  $b^*/\eta^*$  equals  $\frac{4}{3}$  within the uncertainties of this calculation (about 2% for the ratio) for  $1 \leq \Gamma \leq 160$ , indicating that the bulk viscosity is negligible compared to the shear viscosity in the OCP. Unfortunately, this theory does not yield the thermal conductivity, since no relation similar to (36) or (55) can be written down because of the  $k^2$  weighting of the Rayleigh peak in the OCP.

In absolute units we have  $\eta = \eta^* m_p \omega_p \bar{v}^2$ . The conversion factor is independent of temperature, so that the behavior of  $\eta^*$  as a function of  $\Gamma$  re-

flects the behavior of  $\eta$  vs inverse temperature at constant density. As  $T$  is lowered from the melting temperature,  $\eta$  first decreases to about half its value at melting, and then rises sharply. The low-temperature behavior is not unlike that observed in simple fluids by Gosling *et al.*<sup>12</sup> On the other hand, if  $\eta$  is plotted vs density at constant temperature, it is found to increase monotonically. It is interesting to note here that for a "soft-sphere" system of particles interacting through an  $r^{-12}$  potential which has scaling properties similar to that of the OCP, the recent MD simulations of Ashurst and Hoover<sup>20</sup> show no indication of a minimum when the reduced shear viscosity is plotted vs the dimensionless parameter  $\rho T^{-1/4}$ , which is their equivalent of our  $\Gamma$ .

In order to give some feeling for the orders of magnitude, we calculate  $\eta$  under typical white-dwarf conditions. We choose a purely He<sup>4</sup> composition,  $\rho = 10^8$  g/cm<sup>3</sup>,  $T \simeq 10^7$  K; then  $\Gamma \simeq 5.6$ ;  $r_s = \bar{r}/a_0 \simeq 640$  ( $a_0$  is the ionic Bohr radius), the ratio of the thermal de Broglie wavelength  $\lambda$  over the ion-sphere radius  $r$  is  $\lambda/\bar{r} \simeq 0.24$ , so that quantum corrections are weak.<sup>20</sup>  $\omega_p \simeq 5 \times 10^{17}$  Hz; the viscosity unit  $m_p \omega_p \bar{v}^2 = 7.3 \times 10^3$  P;  $\eta \simeq 7.3 \times 10^2$  P, but the kinematic viscosity  $\eta/m_p \simeq 10^{-3}$  cm<sup>2</sup> sec<sup>-1</sup>, which is of the order of magnitude of simple liquids (e.g., Ar).

Our calculation of the transport coefficients can only be expected to be valid as long as the OCP behaves as a dense fluid, so that frequent collisions maintain local thermodynamic equilibrium. For  $\Gamma < 1$ , the collision frequency becomes of the order of magnitude, or smaller than, the plasma frequency, and the generalized hydrodynamics approach cannot be expected to yield reliable results. In that range of  $\Gamma$ , the transport coefficients must be calculated in the framework of kinetic theory.<sup>22,23</sup>

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