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Collective Modes and Dynamic Structure Factor of a Two-Dimensional Electron Fluid

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The electrons bound to the surface of liquid dielectrics by image forces are described as a two-dimensional, classical, one-component plasma with inverse distance interactions. Exact expressions for the collective modes and the dynamic structure factor are obtained from first principles in the limit of long wavelengths. The differences and analogies with uncharged particle fluids and with the three-dimensional one-component plasma are explicitly displayed. The previously used mean-field approximation is shown not to describe weakly coupled systems and to be inadequate in the long-wavelength region.

KEY WORDS: Coulomb systems; plasma oscillations; dynamic structure factor; two-dimensional electron fluid, electron surface layer; mean-field approximation.

1. INTRODUCTION

The study of the static and dynamic properties of Coulomb systems has recently become a very active field.[3] Detailed computer experiments on the simplest Coulomb system, the one-component plasma (OCP), have been performed by Hansen *et al.*[1,2] A number of computer experiments on two-component plasmas, molten salts, and ionic mixtures are underway,[1,3] while the number of theoretical studies is also currently increasing.[1,4,5]

Recently, an interesting novel type of Coulomb system has emerged. It is obtained experimentally by fixing extra electrons on the exterior of dielectric surfaces by means of image-binding.[6,7] These electrons can move freely along the surface but find their motion perpendicular to the surface

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[3] For a general review of the present state of the art, consult the proceedings of a summer school held in Orléans in July 1977 (see Ref. 1).

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extremely restricted. In most experimental situations these electrons behave classically. The conditions of density and temperature can be varied rather easily so as to cover the whole fluid phase, ranging from weak to strong coupling situations. As done by previous authors,[8] we will describe this (charge-compensated) electron surface layer as a classical, two-dimensional, one-component plasma (OCP). The interaction energy between two electrons of the layer a distance r apart will be written $V(r) = e^2/r$, with the understanding that here e denotes an effective charge related to the electronic charge, say e_0, by $e = e_0/\epsilon^{1/2}$, where ϵ is the arithmetic mean of the dielectric constants of the adjacent dielectrics [$\epsilon = (\epsilon_1 + \epsilon_2)/2$]. The fact that here the interaction potential $V(r)$ is the three-dimensional ($\sim 1/r$) and not the two-dimensional ($\sim \ln r$) Coulomb potential finds its origin in the surface layer being only approximately two-dimensional. In this treatment, we will neglect the presence of impurity atoms from the surface, which are known to play an important role in the laboratory experiments.[9] The present model is nevertheless directly accessible to computer experiments.[10]

In Section 2, we recall the basic ingredients of the microscopic theory of Coulomb systems, which was developed elsewhere,[5] and adapt them to the present case. The long-wavelength limit of the collective modes and the dynamic structure factor are obtained in Section 3. The results for the electron surface layer are compared there with the earlier results[5] for the three-dimensional OCP and with the Landau–Placzek result for uncharged fluids. In Section 4, we compare our results with the mean-field results obtained by various authors[8] and conclude that the mean-field approximation is inadequate for describing the long-wavelength behavior of the electron surface layer, even if the latter is weakly coupled.[4] Finally, our conclusions are summarized in Section 5.

2. MICROSCOPIC THEORY OF COULOMB SYSTEMS

The charge-compensated electron surface layer will be described here as a classical, 2D (two-dimensional) OCP (one-component plasma). We will start from the microscopic theory developed elsewhere[5a] for the 3D OCP. This theory can be easily adapted to the present case. The interest of starting with a theory that does not rely on any assumption concerning the strength of the system's coupling is twofold. First, most of the experimental systems are known to be fairly strongly coupled.[8–10] More important, however, is the fact that we expect difficulties to show up in the limit of vanishing coupling and in the mean-field approximation. Indeed, as was shown by Totsuji,[8c]

[4] A short report was presented at the recent IUPAP conference on Statistical Physics (Haifa, August 1977) and will be published in the proceedings of this conference.

the Debye–Hückel theory, which is the static version of Vlassov's mean-field theory, does not yield correctly the static equilibrium correlations of a weakly coupled 2D OCP, whereas it does in the 3D case. As the enhancement of the collisional effects noticed by Totsuji for the 2D statics is likely to show up also in the dynamics, we feel that it is important to be able to analyze the mean-field approximation starting from a more general setting.

We summarize now the main steps of the microscopic approach, referring to the literature[5a] for the details. We start from the microscopic phase-space density $f(\mathbf{r}\mathbf{p}t)$ of the N particles:

$$f(\mathbf{r}\mathbf{p}t) = \sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{x}_j(t))\, \delta(\mathbf{p} - \mathbf{p}_j(t)) \tag{1}$$

where $\mathbf{x}_j(t)$ and $\mathbf{p}_j(t)$ denote, respectively, the position and momentum of particle j at time t. Next, we consider the space–time correlation function S of the equilibrium fluctuations $\delta f = f - \langle f \rangle$ of f:

$$S(\mathbf{r} - \mathbf{r}', t - t'; \mathbf{p}\mathbf{p}') = \langle \delta f(\mathbf{r}\mathbf{p}t)\, \delta f(\mathbf{r}'\mathbf{p}'t') \rangle \tag{2}$$

where, as usual, $\langle A \rangle$ denotes the canonical equilibrium average of A over the initial phase $\{\mathbf{x}_j(0), \mathbf{p}_j(0)\}$. From the Liouville equation obeyed by $f(\mathbf{r}\mathbf{p}t)$ an exact kinetic equation for S is then derived.[5] This equation reads in Fourier–Laplace transform

$$zS(\mathbf{k}z; \mathbf{p}\mathbf{p}') - \int d\mathbf{p}''\, \Sigma(\mathbf{k}z; \mathbf{p}\mathbf{p}'')S(\mathbf{k}z; \mathbf{p}''\mathbf{p}') = iS(\mathbf{k}, t = 0; \mathbf{p}\mathbf{p}') \tag{3}$$

where Σ is the so-called memory function. The kinetic equation (3) is then finally transformed into an algebraic set of transport equations:

$$\sum_{i=1}^{5} [z\, \delta_{ji} - \Omega_{ji}(\mathbf{k}z)]G_{ij'}(\mathbf{k}z) = iG_{jj'}(\mathbf{k}, t = 0) \tag{4}$$

for the hydrodynamic space–time correlation functions G_{ij}:

$$G_{ij}(\mathbf{k}z) = \int d\mathbf{p}\, d\mathbf{p}'\, \frac{u_i(\mathbf{p})}{a_i}\, S(\mathbf{k}z; \mathbf{p}\mathbf{p}')\, \frac{u_j(\mathbf{p}')}{a_j} \tag{5}$$

where the $u_i(\mathbf{p})$ ($i = 1$–5) correspond to the density ($i = n$), the longitudinal ($i = l$), and transverse ($i = t_1, t_2$) momentum and the excess kinetic energy ($i = \epsilon$) states, and the a_i are normalization constants. The transport matrix Ω_{ij} appearing in Eq. (4) can be further related to the memory function Σ of Eq. (3) [see, for instance, Eq. (44)]. Using the system's invariance and

[5] We follow here very closely a theory first developed by Forster and Martin[15] for uncharged particle fluids.

conservation laws, we arrive at the following form for the matrix elements Ω_{ij} of interest to us:

$$\Omega_{nl}(\mathbf{k}z) = kv_0 \tag{6a}$$

$$\Omega_{ln}(\mathbf{k}z) = kv_0[1 - c(k)]; \qquad \Omega_{ll}(\mathbf{k}z) = -ik^2 D_l(\mathbf{k}z); \qquad \Omega_{l\epsilon}(\mathbf{k}z) = kD_{l\epsilon}(\mathbf{k}z) \tag{6b}$$

$$\Omega_{\epsilon l}(\mathbf{k}z) = kD_{\epsilon l}(\mathbf{k}z); \qquad \Omega_{\epsilon\epsilon}(\mathbf{k}z) = -ik^2 D_\epsilon(\mathbf{k}z) + zB_\epsilon(z) \tag{6c}$$

These are in fact the only nonvanishing matrix elements Ω_{ij} for $(i, j) = (n, l, \epsilon)$. The k and z factors appearing in Eq. (6) have been pulled out from Ω_{ij} by using the microscopic conservation laws, while the i-factors have been introduced for later convenience. In Eqs. (6) we have also introduced the thermal velocity v_0 [$v_0^2 = (m\beta)^{-1}$, m being the mass of the particles and β the inverse temperature in energy units] and the (dimensionless) direct correlation function $c(k)$, which is related to the static structure factor $S(k)$ by

$$S(k) = [1 - c(k)]^{-1} \tag{7}$$

With these ingredients, the density–density correlation function G_{nn}, which is obtained from Eq. (5) with $u_n(\mathbf{p}) = 1$ and a_n^2 equal to the number density n, i.e.,

$$G_{nn}(\mathbf{k}z) = (1/n)\int d\mathbf{p}\, d\mathbf{p}'\, S(\mathbf{k}z; \mathbf{p}\mathbf{p}') \tag{8}$$

can now be further resolved exactly in terms of the transport matrix elements of Eqs. (6) as

$$G_{nn}(\mathbf{k}z) = iS(k)\frac{[z + ik^2 D_l(kz)][z + ik^2\alpha_\epsilon(kz)] - k^2\gamma_{l\epsilon}(kz)}{[z^2 - \omega^2(k) + zik^2 D_l(kz)][z + ik^2\alpha_\epsilon(kz)] - zk^2\gamma_{l\epsilon}(kz)} \tag{9}$$

where $S(k) = G_{nn}(k, t = 0)$, while the following abbreviations have been introduced:

$$\omega^2(k) = (kv_0)^2[1 - c(k)]; \qquad \alpha_\epsilon(kz) = D_\epsilon(kz)/[1 - B_\epsilon(z)];$$
$$\gamma_{l\epsilon}(kz) = D_{l\epsilon}(kz)D_{\epsilon l}(kz)/[1 - B_\epsilon(z)] \tag{10}$$

In the following we will be concerned with the behavior of $G_{nn}(kz)$, as given by Eq. (9), but only for small k values. In order to perform such an analysis we need to know the small-k behavior of $c(k)$ and the amount of analyticity satisfied by $D_l(kz)$, $\alpha_\epsilon(kz)$, and $\gamma_{l\epsilon}(kz)$. Leaving the discussion of the dynamical quantities such as $D_l(kz)$ for next section, we close this summary of the microscopic approach with a discussion of the small-k statics.

We will assume that, if $V(r)$ is the interaction potential, we can write in position space

$$c(r) = -n\beta V(r) \qquad \text{for} \quad r \to \infty \tag{11}$$

This relation states that distant particles can only be weakly coupled. Indeed, the rhs of Eq. (11) is also the weak-coupling limiting value of $c(r)$. In three dimensions, Eq. (11) is very well satisfied by the computer experiments[11] on the OCP. A formal proof of Eq. (11) for a general fluid appears to be underway.[12] We also notice that relation (11) is very sensitive to the nature of the interaction potential. For instance, for the 3D OCP with $V(r) = e_0^2/r$ we have in wave-vector space

$$c(k) = -k_3^2/k^2 \qquad \text{for} \quad k \to 0 \tag{12}$$

where k_3 is the 3D Debye wave vector ($k_3^2 = 4\pi e_0^2 n\beta$). The singular nature of $c(k)$ for the OCP, as displayed in Eq. (12), was shown elsewhere[5a] to explain the profound differences between the small-k behavior of the OCP and of ordinary fluids. For the 2D OCP with $V(r) = e^2/r$, where e is the effective charge, we obtain instead of Eq. (12)

$$c(k) = -k_2/k \qquad \text{for} \quad k \to 0 \tag{13}$$

where $k_2 = 2\pi e^2 n_2 \beta$ is the 2D analog of the Debye wave vector, n_2 being the average number of particles per unit area. From Eq. (13) we see that in two-dimensions $c(k)$ is still singular for small k, but this singularity, which is characteristic of Coulomb systems, has been weakened. As will be seen below, it is this weakening of the Coulomb singularity that is responsible for the fact that in two dimensions the plasma mode is a low-frequency mode whereas the Debye screening is algebraic rather than exponential.[8] We find it illuminating to be able to follow explicitly the modifications brought about by the change of dimensionality d between $d = 2$ and $d = 3$. We therefore introduce a dimensionality index d so that all the relevant information about $c(k)$ can be summarized in the following expression:

$$c(k) = -(k_d/k)^{d-1} + \hat{c}(k) \tag{14}$$

The d-D OCP ($d = 2, 3$) with $1/r$ interactions can then be characterized by the coupling constant $\lambda_d = (k_d)^d/n_d$ measuring the inverse of the number of particles in a d-D cube constructed with the Debye length k_d^{-1} as unit length. Here and in Eq. (14), k_d denotes the Debye wave vector, $k_d = (2^{d-1}\pi e_d^2 n_d \beta)^{1/d-1}$, of a system of temperature β^{-1}, of particles of charge e_d ($e_2 = e$, $e_3 = e_0$), and of average number of particles per unit d-D volume given by n_d. In Eq. (14), $\hat{c}(k)$ represents a remainder, which is assumed regular as $k \to 0$ and which can incorporate any short-range interactions eventually

present together with the Coulomb interactions. It is convenient to define the isothermal compressibility χ_T through the relation

$$\hat{c}(k = 0) = 1 - \chi_T{}^0/\chi_T \tag{15}$$

where $\chi_T{}^0 = \beta/n_d$ is the perfect-gas value of χ_T. With these definitions, Eq. (14) reduces to the Debye–Hückel approximation for a pure Coulomb case if we put $\hat{c}(k) = 0$, whereas it reduces to the neutral fluid result if we put e_d, and hence also k_d, equal to zero. Substitution of Eqs. (14)–(15) into Eq. (7) leads then immediately to

$$S(k) = (k/k_d)^{d-1} \qquad \text{for} \quad k \to 0 \tag{16}$$

for the OCP ($e_d \neq 0$), whereas for a neutral fluid ($e_d = 0$) we recover the compressibility sum-rule

$$S(k = 0) = \chi_T/\chi_T{}^0 \tag{17}$$

This is as far as we need to go for the static properties.

3. THE COLLECTIVE MODES AND DYNAMIC STRUCTURE FACTOR

From Eq. (9) we see that the collective modes building up $G_{nn}(\mathbf{k}z)$ are given by the solutions of the following dispersion equation:

$$[z^2 - \omega^2(k) + zik^2 D_l(kz)][z + ik^2\alpha_\epsilon(kz)] = zk^2\gamma_{l\epsilon}(kz) \tag{18}$$

This equation has the typical structure of a coupled mode spectrum where the density modes, corresponding to the zeros of $z^2 - \omega^2 + zik^2 D_l$, are coupled to the energy modes, $z + ik^2\alpha_\epsilon = 0$, by the rhs of Eq. (18), i.e., by $\gamma_{l\epsilon}$.

In what follows we will only be interested in the solutions $z = z(k)$ of Eq. (18) for small k values. In this microscopic region ($k \to 0$) we expect *weakly damped* modes to show up, which for large enough times t will dominate the Van Hove function $G_{nn}(\mathbf{k}, t)$, i.e., the time-image of $G_{nn}(\mathbf{k}z)$. The small-k, large-t behavior of $G_{nn}(\mathbf{k}, t)$ will be analyzed below. First, we will concentrate on the collective modes given by the small-k solutions of Eq. (18). The small-k behavior of $\omega(k)$ appearing in Eq. (18) follows immediately from Eqs. (10), (14)–(15):

$$\omega^2(k) = (kv_0)^2[(k_d/k)^{d-1} + \chi_T{}^0/\chi_T] \qquad \text{for} \quad k \to 0 \tag{19}$$

For uncharged particles ($e_d = 0 = k_d$) Eq. (19) yields the isothermal sound wave frequency, $\omega^2(k) = (kc)^2$, with $c^2/v_0{}^2 = \chi_T{}^0/\chi_T$, whereas for the d-D OCP we get $\omega^2(k) = \omega_d{}^2(k/k_d)^{3-d}$, with $\omega_d = v_0 k_d$, i.e., a high-frequency plasma mode in three dimensions and a low-frequency plasma mode in two dimensions. In order to obtain the small-k solutions of Eq. (18) we also need

some information about the behavior of $D_l(\mathbf{k}z)$, $\alpha_\epsilon(\mathbf{k}z)$, and $\gamma_{l\epsilon}(\mathbf{k}z)$. In three dimensions $D_l(\mathbf{0}0)$ and $\alpha_\epsilon(\mathbf{0}0)$ can be related to the transport coefficients[5a] (respectively, the longitudinal viscosity and the thermal conductivity), whereas $\gamma_{l\epsilon}(\mathbf{0}0)$ is a thermodynamic coefficient given by

$$\gamma_{l\epsilon}(\mathbf{0}0) = c^2(c_p/c_v - 1) \tag{20}$$

where c_p/c_v is the specific heat ratio. In two dimensions, the situation is not so favorable, as $D_l(\mathbf{0}, z)$ and $\alpha_\epsilon(\mathbf{0}, z)$ presumably[13,14] exhibit a weak logarithmic singularity for small z. As our intention here is not to tackle this difficult problem, we will content ourselves with the following assumptions, compatible with our present knowledge of 2D transport. We will assume that the transport quantities D_l and α_ϵ are such that $kD_l(\mathbf{0}, z(k))$ and $k\alpha_\epsilon(\mathbf{0}, z(k)$ vanish as $k \to 0$ for $z(k)$ at most of order $k^{1/2}$, whereas $\gamma_{l\epsilon}(\mathbf{k}z)$, not being a transport quantity, will be assumed to exist at $k = 0$, $z = 0$. Then one can formally prove that $\gamma_{l\epsilon}(\mathbf{0}0)$ is still given by Eq. (20) with c_v interpreted as the specific heat at constant d-D volume. With these assumptions the dispersion equation (18) can be shown to possess small-k solutions corresponding to *weakly damped* modes, which will now be displayed.

3.1. The Diffusive Mode

Dividing Eq. (18) by the first factor in its lhs and rearranging terms, we obtain the dispersion equation in the form

$$z = -ik^2\alpha_\epsilon(\mathbf{k}z)\left[1 - \frac{k^2\gamma_{l\epsilon}(\mathbf{k}z)}{z^2 - \omega^2(k) + zik^2D_l(kz)}\right]^{-1} \tag{21}$$

which we solve for $z = z(k)$. Using the assumptions stated above, we can reduce Eq. (21) to

$$z(k) = -ik^2\alpha_\epsilon(\mathbf{k}, z(k))\left[1 + \frac{k^2\gamma_{l\epsilon}(\mathbf{k}, z(k))}{\omega^2(k)}\right]^{-1} \tag{22}$$

which takes the form of a diffusive mode, $z_T(k) = -ik^2D_T(k)$, with a thermal diffusitivity $D_T(k)$ given to dominant order in k by

$$D_T(k) = \alpha_\epsilon(\mathbf{0}, z_T(k))\left[1 + \left(\frac{c_p}{c_v} - 1\right)\frac{\chi_T{}^0}{\chi_T}S(k)\right]^{-1} \tag{23}$$

where we have used Eq. (20) and $\omega^2(k) = (kv_0)^2/S(k)$ [see Eqs. (7), (10)]. For a neutral fluid we obtain from Eqs. (7) and (23)

$$D_T(k) = \alpha_\epsilon(\mathbf{0}, -ik^2D_T(k))c_v/c_p \qquad \text{for} \quad e_d = 0 \tag{24}$$

whereas for the d-D OCP Eqs. (16) and (23) lead to

$$D_T(k) = \alpha_\epsilon(\mathbf{0}, -ik^2D_T(k)) \qquad \text{for} \quad e_d \neq 0 \tag{25}$$

As stated above, we will not further elaborate on the weak z dependence of $\alpha_\epsilon(\mathbf{0}, z)$ still retained in Eqs. (24), (25). Let us only recall that in the 3D case $\alpha_\epsilon(\mathbf{0}, 0)$ can be related to the thermal conductivity κ by $\alpha_\epsilon(\mathbf{0}, 0) = \kappa/nmc_v$. This then brings us to our main point here, that both the charged ($e_d \neq 0$) and uncharged ($e_d = 0$) fluid exhibit a thermal diffusion mode, but with different thermal diffusitivities, as seen from the absence of the c_v/c_p factor of Eq. (24) from Eq. (25). In the presence of Coulomb interactions the coupling of the energy and density fluctuations has thus been weakened both in two and three dimensions. This effect, which was known previously[5a] for the 3D OCP, has been shown here to persist also for the electron surface layer because the static structure factor $S(k)$ as it appears in Eq. (23) is vanishingly small for small k in both cases. This Coulomb effect is only weakly dependent on the dimensionality [$S(k) = O(k^{d-1})$].

3.2. The Propagating Modes

The situation is quite different for the remaining density modes, which we obtain by rewriting the dispersion equation (18) in the form

$$z^2 = \omega^2(k) - zik^2 D_l(\mathbf{k}z) + zk^2\gamma_{l\epsilon}(\mathbf{k}z)/[z + ik^2\alpha_\epsilon(\mathbf{k}z)] \tag{26}$$

and solving once more for $z = z(k)$. The small-k solution of Eq. (26) is now seen to be controlled by $\omega(k)$, which, according to Eq. (19), is of order k for a neutral fluid, of order k^0 for the 3D OCP, and of order $k^{1/2}$ for the 2D OCP. In all cases we obtain from Eq. (26) two oppositely propagating modes, $z_\pm(k) = \pm\bar{\omega}(k) - \frac{1}{2}ik^2\Gamma(\pm\bar{\omega}(k))$, with $\bar{\omega}(k)$ defined by

$$\bar{\omega}^2(k) = \omega^2(k) + k^2\gamma_{l\epsilon}(\mathbf{0}, 0) \tag{27}$$

and with $\Gamma(\pm\bar{\omega})$ given by

$$\Gamma(\pm\bar{\omega}(k)) = \frac{k^2\gamma_{l\epsilon}(\mathbf{0}, 0)}{\bar{\omega}^2(k)}\alpha_\epsilon(\mathbf{0}, \pm\bar{\omega}(k)) + D_l(\mathbf{0}, \pm\bar{\omega}(k)) + \frac{\gamma_{l\epsilon}(\mathbf{0}, \pm\bar{\omega}(k)) - \gamma_{l\epsilon}(\mathbf{0}, 0)}{\mp i\bar{\omega}(k)} \tag{28}$$

Let us consider now the different cases more explicitly. First, for the neutral case ($e_d = 0$), we obtain from Eqs. (27) and (19)–(20)

$$\bar{\omega}^2(k) = (k\bar{c})^2; \qquad \bar{c}^2 = c^2c_p/c_v \qquad \text{for} \quad e_d = 0 \tag{29}$$

showing that the coupling of the energy and density fluctuations described by $\gamma_{l\epsilon}$ has shifted the sound speed from its isothermal (c) to its isentropic ($\bar{c}$)

value according to Eq. (27). The damping of these sound modes is then further described by Eq. (28), leading to

$$\Gamma(\pm\bar{\omega}) = \left(1 - \frac{c_v}{c_p}\right)\alpha_\epsilon(\mathbf{0}, \pm\bar{\omega}(k)) + D_l(\mathbf{0}, \pm\bar{\omega}(k)) + i\left.\frac{d\gamma_{l\epsilon}(\mathbf{0}, z)}{dz}\right|_{z=0}; \qquad e_d = 0 \tag{30}$$

where in three dimensions, $\alpha_\epsilon(\mathbf{0}, 0)$ and $D_l(\mathbf{0}, 0) + i\, d\gamma_{l\epsilon}(\mathbf{0}, 0)/dz$ can be further identified as, respectively, the damping due to the thermal conductivity (κ/nmc_v) and to the viscosity $[(\frac{4}{3}\eta + \xi)/nm]$ of the fluid. Second, for the 3D OCP, Eqs. (27) and (19) show that $\bar{\omega}(k = 0) = \omega(k = 0)$ is finite and equal to ω_3, the standard plasma frequency. Hence the thermal conductivity contribution to Γ, i.e., the first term in the rhs of Eq. (28), drops out for small k, whereas the last term in the rhs of Eq. (28), which is part of the bulk viscosity contribution, recombines with the second term in the rhs of Eq. (27), yielding finally

$$z_\pm(k) = \pm\omega_3\left[1 + \frac{k^2}{2}\frac{c^2 + \gamma_{l\epsilon}(\mathbf{0}, \pm\omega_3)}{{\omega_3}^2}\right] - i\frac{k^2}{2}D_l(\mathbf{0}, \pm\omega_3); \qquad e_3 \neq 0 \tag{31}$$

where the c^2 contribution stems from Eq. (19). As observed elsewhere,[5a] the fact that here $\gamma_{l\epsilon}$ and D_l have to be evaluated at the finite frequency ω_3 makes them complex quantities and prevents us from relating them further to the standard thermodynamic and transport coefficients. Finally, for the 2D OCP we get an intermediate behavior between the two previous cases. From Eqs. (27) and (19) we get now

$$\bar{\omega}^2(k) = {\omega_2}^2 k/k_2 + k^2\bar{c}^2; \qquad e_2 \neq 0 \tag{32}$$

or

$$\bar{\omega}(k) = \pm\left(\frac{k}{k_2}\right)^{1/2}\omega_2\left(1 + \frac{1}{2}\frac{k}{k_2}\frac{\bar{c}^2}{{v_0}^2}\right) \tag{33}$$

which when substituted into Eq. (28) yields

$$\Gamma(\pm\bar{\omega}(k)) = D_l(\mathbf{0}, \pm\bar{\omega}(k)) + i\, d\gamma_{l\epsilon}(\mathbf{0}, 0)/dz; \qquad e_2 \neq 0 \tag{34}$$

From Eq. (33) we see that the plasma mode has now become a low-frequency mode $[\bar{\omega}(k) = O(k^{1/2})]$, and hence the dispersive corrections to the plasma frequency can again be expressed in terms of thermodynamic quantities $(\bar{c})$. The plasma frequency, Eq. (33), is, however, still too high for the thermal conductivity contribution to Eq. (28) to remain, so that the damping of this plasma mode has the same form as the sound absorption coefficient, but without the thermal conductivity contribution [compare Eqs. (34) and (30)].

3.3. The Dynamic Structure Factor

Besides the nature of the collective modes, it is also of interest to know the strength with which they contribute to the dynamic structure factor or the Van Hove function $G_{nn}(\mathbf{k}, t)$. Returning to Eq. (9), it is easily established with the aid of the results of the preceding sections that for small k we can write

$$\frac{G_{nn}(\mathbf{k}z)}{S(k)} = \frac{i}{z - z_T(k)} \frac{k^2\gamma_{l\epsilon}(\mathbf{0}, z_T(k))}{\bar{\omega}^2(k)} + \frac{1}{2}\sum_{\pm} \frac{i}{z - z_{\pm}(k)} \left(1 - \frac{k^2\gamma_{l\epsilon}(\mathbf{0}, \pm\bar{\omega}(k))}{\bar{\omega}^2(k)}\right) \tag{35}$$

where the diffusive mode $z_T(k)$ and the propagating modes $z_{\pm}(k)$ have been discussed in Sections 3.1 and 3.2, respectively. The inverse Laplace transform of Eq. (35) yields then the Van Hove function $G_{nn}(\mathbf{k}, t)$ whose Fourier transform is the dynamic structure factor. We can thus continue our discussion on the basis of Eq. (35). Let us first consider the strength, say $a_T(k)$, of the thermal mode, which, according to Eq. (35), is given by $a_T = k^2\gamma_{l\epsilon}/\bar{\omega}^2$. For the neutral fluid we obtain from Eqs. (20), (29)

$$a_T(k = 0) = 1 - c_v/c_p; \qquad e_d = 0 \tag{36}$$

whereas for the d-D OCP we get from Eqs. (19), (20)

$$a_T(k) = \left(\frac{k}{k_d}\right)^{d-1} \frac{\bar{c}^2 - c^2}{{v_0}^2}; \qquad e_d \neq 0 \tag{37}$$

For the strength of the propagating modes $2a_{\pm}(k) = 1 - k^2\gamma_{l\epsilon}(\mathbf{0}, \pm\bar{\omega})/\bar{\omega}^2$ we get, similarly,

$$2a_{\pm}(k = 0) = c_v/c_p; \qquad e_d = 0 \tag{38}$$

for the neutral fluid and

$$2a_{\pm}(k) = 1 - \left(\frac{k}{k_d}\right)^{d-1} \frac{\gamma_{l\epsilon}(\mathbf{0}, \pm\bar{\omega}(k))}{{v_0}^2}; \qquad e_d \neq 0 \tag{39}$$

for the charged fluid. In all cases we find that $a_T + \sum_{\pm} a_{\pm} = 1$ for small k, i.e., the collective modes we have found exhaust the small-k portion of the zeroth-order sum rule of the dynamic structure factor. There is, however, a definite difference between the neutral and charged fluid, in that the propagating plasma modes of the d-D OCP exhaust the sum rule by themselves, the diffusive thermal mode contributing only an order k^{d-1} term.

4. RELATION TO MEAN-FIELD THEORY

The results obtained in the previous section do not involve any assumption concerning the strength of the coupling and are restricted only by the

long-wavelength limit and the isotropy of the fluid phase. It is thus of interest to compare them with the results obtained previously by various authors[8] in the mean-field approximation. This approximation can be easily recovered from the kinetic equation (3). Indeed, let us recall that the memory function Σ of Eq. (3) can be split into three terms,[5a] $\Sigma = \Sigma^0 + \Sigma^s + \Sigma^c$, the free-flow term Σ^0, the self-consistent field term Σ^s, and the collision term Σ^c. The mean-field approximation corresponds, then, to neglecting all collisional effects, i.e., $\Sigma^{\text{MFA}} = \Sigma^0 + \Sigma^s$. In this approximation no diffusive heat mode (cf. Section 3.1) is obtained, as the collisions are essential for establishing this transport process. For the plasma modes of the d-D OCP we obtain from Section 2 in a straightforward manner and for small k

$$z_\pm(k) = \pm(k/k_d)^{(3-d)/2}\omega_d[1 + \tfrac{3}{2}(k/k_d)^{d-1}] - i\Gamma_d(k) \tag{40}$$

where $\Gamma_d(k)$ is the Landau damping

$$\Gamma_d(k) = (\pi/8)^{1/2}\omega_d(k_d/k)^{2d-3}\exp[-\tfrac{1}{2}(k_d/k)^{d-1} - \tfrac{3}{2}]$$

As Eq. (40) reduces to the result obtained by previous authors[8] from the random-phase approximation to the dielectric constant, we can skip its derivation here. The contribution from the electron collisions to the damping, say $\Gamma_2{}^c(k)$, was calculated by Totsuji[8c] from the Boltzmann equation. He found

$$\Gamma_2{}^c(k) = (3\pi^{1/2}/16)kv_0 \tag{41}$$

which, when added to the Landau damping $\Gamma_2(k)$ of Eq. (40), is seen to dominate the latter for small k.

For the 3D OCP we have shown elsewhere[5a] that Eq. (40) with $d = 3$ is the weak coupling limit, $\lambda_3 \to 0$, of Eq. (31) except that the Landau damping $\Gamma_3(k)$ is dominated for small k by a small collisional damping term $\Gamma_3{}^c(k)$, which was obtained there as

$$\Gamma_3{}^c(k) = (1/15\pi^{3/2})(k/k_3)^2\omega_3\lambda_3 \ln \lambda_3^{-1} \tag{42}$$

where we recall that $\omega_3 = v_0k_3$ and $\lambda_3 = (k_3)^3/n_3$. Hence, as expected, the mean-field approximation is seen to describe correctly the plasma modes of a weakly coupled 3D OCP in the limit $\lambda_3 \to 0$. In the 2D case, however, this is not the case, i.e., Eq. (40) with $d = 2$ is not equivalent to the weak coupling limit, $\lambda_2 \to 0$, of Eqs. (33)–(34). Indeed, as $\lambda_2 \to 0$, we obtain from Eq. (33)

$$\bar{\omega}(k) = \pm(k/k_2)^{1/2}\omega_2(1 + k/k_2); \qquad \lambda_2 \to 0 \tag{43}$$

because as $\lambda_2 \to 0$, $c^2/v_0{}^2 \to 1$ and $c_p/c_v \to 2$. Comparing Eq. (43) with Eq. (40) for $d = 2$, we see that the dispersive correction to the plasma mode as

computed by the mean-field approximation is 3/2 times the weak coupling limit,[6] Eq. (43), of the exact result given by Eq. (40). Moreover, Eq. (34) can be compared neither with $\Gamma_2(k)$ of Eq. (40) nor with $\Gamma_2{}^c(k)$ of Eq. (41). The reason for this discrepancy in the 2D case is twofold. First there is the fact that a rough estimate of the collision frequency $\omega_c \sim \Sigma^c$ indicates that $\omega_c \sim (\lambda_d)^{d-2}\omega_d$. This enhancement of the collision frequency in two dimensions was first pointed out by Totsuji.[8o] This then implies that the mean-field approximation ($\omega_c \to 0$) is a weak coupling approximation ($\lambda_d \to 0$) for $d = 3$ ($\omega_c \sim \lambda_3\omega_3$), but not for $d = 2$ ($\omega_c \sim \omega_2$). This is in fact already indicated by the fact that $\Gamma_3{}^c$ of Eq. (42) vanishes with λ_3, whereas $\Gamma_2{}^c$ of Eq. (41) is independent of λ_2. The second reason is that in the presence of a low-frequency mode such as the 2D plasma mode (but not the 3D plasma mode) we can not freely commute the long-wavelength limit ($k \to 0$) and the collisionless limit ($\Sigma^c \sim \omega_c \to 0$). This is most clearly seen by recalling the relation which exists[5] between the transport matrix Ω_{ij} of Eq. (4) and the memory function Σ of Eq. (3). For concreteness we focus on $\Omega_{ll}(\mathbf{k}z) = -ik^2 D_l(\mathbf{k}z)$, which appears in Eq. (34). We have then

$$\Omega_{ll}(\mathbf{k}z) = \langle l|\Sigma^c|l\rangle + \langle l|(\Sigma^0 + \Sigma^c)Q(z - Q\Sigma^0 Q - Q\Sigma^c Q)^{-1}Q(\Sigma^0 + \Sigma^c)|l\rangle \tag{44}$$

where Q projects out the hydrodynamic states,[5] and where, for simplicity, we did not indicate the k and z dependence of $\Sigma^0(\mathbf{k})$ and $\Sigma^c(\mathbf{k}z)$. For weak collisional effects ($\omega_c \to 0$) we can neglect the first term in the rhs of Eq. (44) and rewrite $\Omega_{ll}(\mathbf{k}, \bar{\omega}(k))$ as needed for Eq. (34) as

$$\Omega_{ll}(\mathbf{k}, \bar{\omega}(k)) = \langle l|\Sigma^0 Q(\bar{\omega}(k) - Q\Sigma^0 Q - Q\Sigma^c Q)^{-1}Q\Sigma^0|l\rangle \tag{45}$$

where $\bar{\omega}(k)$ is given by Eq. (33). From Eq. (45) we see that as $\Sigma^c \to 0$, for given k, there develops a singularity in the propagator $[\bar{\omega}(k) - Q\Sigma^0 Q - i0]^{-1}$ of Eq. (45). This singularity is well known to lead to the Landau damping term of Eq. (40). If, on the contrary, we let $k \to 0$, for given Σ^c however small but different from zero, then as both $\bar{\omega}(k)$ and Σ^0 vanish with k the propagator in Eq. (45) reduces to $(-Q\Sigma^c Q)^{-1}$, i.e., a different result. We can rephrase this somewhat differently. As here $\bar{\omega}(k) = O(k^{1/2})$ we can neglect $Q\Sigma^0 Q$ in front of $\bar{\omega}(k)$ in Eq. (45) and expand $[\bar{\omega}(k) - Q\Sigma^c Q]^{-1}$ for weak Σ^c as $(\bar{\omega})^{-1}(1 + Q\Sigma^c Q/\bar{\omega})$. The contribution of the $\bar{\omega}^{-1}$ term to Eq. (34) can then be shown to operate exactly the transition from Eq. (43) to Eq. (40), while the $Q\Sigma^c Q/\bar{\omega}^2$ term reproduces exactly Eq. (41) if Σ^c is approximated by the linearized Boltzmann collision operator.[7] This then indicates how the mean-

[6] This result, Eq. (43), has also been obtained by Onuki,[16] who applied the method of the hydrodynamical modes[17] to a model-Boltzmann equation. We thank H. Totsuji for calling our attention to this unpublished result.

[7] An equivalent result can also be obtained from the linearized Landau collision operator with an appropriate large-wavevector cutoff.

field result can be obtained from our general formulation. The validity of this collisionless approximation thus rests on an expansion in $Q\Sigma^c Q/\bar{\omega}(k) \sim \omega_c/\bar{\omega}$ and is valid when

$$\omega_c/\omega_2 \ll (k/k_2)^{1/2} \ll 1 \tag{46}$$

while our result of Section 3 will be valid when

$$(k/k_2)^{1/2} \ll \omega_c/\omega_2 \tag{47}$$

Whereas Eq. (47) can always be satisfied for small enough k values, this is not so for Eq. (46). We can rephrase now our objections against the mean-field results as follows. The mean-field or collisionless approximation of the 2D OCP never describes a weakly coupled system ($\lambda_2 \to 0$), because ω_c remains finite as $\lambda_2 \to 0$. When for some as yet unspecified reason the collisional effects are weak, the mean-field approximation can only describe that portion of the k spectrum satisfying Eq. (46), but not the longest wavelengths satisfying Eq. (47).

5. CONCLUSIONS

In an attempt to describe the electrons trapped at the surface of liquid dielectrics, we have considered the collective modes and the dynamical structure factor of a classical, two-dimensional, one-component plasma with $1/r$ interactions. We found it illuminating to treat both the charged and uncharged particle system as well as the two- and three-dimensional system from a unified microscopic viewpoint. As a result of the weak intensity of the coupling of the energy and density fluctuations, the thermal conductivity mode of the two-dimensional electron fluid has been shown to *differ* by a factor c_p/c_v from the corresponding mode of uncharged fluids. For the same reason, the thermal conductivity mode contributes only *weakly* to the dynamical structure factor (Rayleigh's central peak) and to the damping of the plasma oscillations of the electron surface layer. Because of the restricted dimensionality, the plasma modes of the layer are shown to be low-frequency modes. As a consequence of this, the damping of the plasma modes can be expressed in terms of the same quantity, except for the thermal conductivity contribution, as the one giving the absorption of the sound waves of uncharged particle systems. Comparing our results, which only require the long-wavelength approximation, with previous results, which all rely on the mean-field approximation, we found that the mean-field approximation of this *two-dimensional* electron fluid does not describe the long-wavelength region correctly, whereas, contrary to a current statement, the mean-field approximation is, in the present case, not equivalent to a weak coupling approximation.

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