Response function and plasmon dispersion for strongly coupled Coulomb liquids

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We present a new approach for the calculation of the dielectric response functions for various strongly coupled Coulomb systems, whose main common feature is that the charges are quasilocalized. Such a model is expected to be a valid description of a strongly coupled plasma in the $\Gamma \gg 1$ limit. The dielectric function $\epsilon(\mathbf{k}\omega)$ and the longitudinal plasmon dispersion $\omega(k)$ appear as functionals of the equilibrium pair-correlation functions. We derive the longitudinal-plasmon dispersions for arbitrary $\mathbf{k}$ values for the three- and two-dimensional one-component plasmas; both of them saturate after an oscillatory behavior substantially below their random-phase approximation value for large $\mathbf{k}$ values. In two-component systems correlational effects bring about marked upward and downward shifts in the plasma frequency $\omega(k=0)$. We find good agreement with molecular-dynamics data for H^+-H^{2+} mixtures and predict a more significant shift for H^+–high-Z mixtures.

I. INTRODUCTION

Most approximation schemes for many-body systems, including classical plasmas, adopt the model of noninteracting particles as a point of departure and add the effect of interaction as a perturbation. For intermediate or strong coupling, this is obviously not an expedient approach. Various nonperturbative mean-field-like[1,2] and semiphenomenological[3] theories have attempted, with varying degrees of success, to attack the problem in this latter domain. In this work we present a novel view and method based upon approaching the problem from the strong coupling side, i.e., from the $\Gamma \gg 1$ side, where $\Gamma = Z^2 e^2 / k_B T a$ is the characteristic coupling parameter for Coulomb systems [a is the interparticle distance; $(4\pi/3)a^3 n = 1$ for three-dimensional systems and $\pi a^2 n = 1$ for two-dimensional systems]. Our general formalism will be applied to a variety of classical plasmas. In the present paper, the following are of interest: (i) uniform-background three-dimensional one-component plasmas (3D OCP)—a single species of classical ions moving in a neutralizing uniform background of rigid (degenerate) electrons; (ii) two-dimensional (2D) electron plasma—classical electrons trapped in surface-bound states at the interface of dielectric materials, e.g., a 2D electron layer confined to the free surface of liquid helium;[4] here the compensating uniform positive background is provided by an electrode placed just below the surface; (iii) uniform-background binary ionic mixtures (BIM); and (iv) electron-ion two-component plasmas (TCP).

The physical observation that serves as the basis of the formal development presented in this paper is that the dominating feature of the physical state of a plasma with $\Gamma \gg 1$ is the quasilocalization of the particles. Data available from Monte Carlo (MC) simulations[5–8] and hypernetted-chain (HNC) calculations[9,10] for the OCP corroborate this physical picture: (i) for high coupling, the pair-correlation function $g(r)$ establishes strong peaks at points separated by the order of the interparticle distance; (ii) the thermodynamic properties (e.g., energy) of the OCP change very little as the system passes from the liquid to crystalline state around $\Gamma_m = 178$ for the 3D OCP and $\Gamma_m = 137 \pm 15$ for the 2D OCP.

This physical picture immediately leads to a model which resembles that of a disordered solid, where the particles occupy randomly located sites and undergo oscillations around them. At the same time, however, the site positions also change and a continuous rearrangement of the underlying quasiequilibrium configuration takes place. Nevertheless, inherent in the model is the assumption that the two time scales are well separated and that for the description of the fast oscillating motion, the time average (converted, of course, into ensemble average) of the drifting quasiequilibrium configuration is sufficient. A requirement certainly necessary for this condition to be satisfied is that the amplitude of the excursion of the oscillations, say s, is much smaller than the interparticle distance a. That this is indeed so, provided $\Gamma \gg 1$, can be seen by considering the paradigm of a strongly coupled one-component plasma, in which the equilibrium level of the energy density of the electric field associated with plasma oscillations can be estimated to be $\lambda(\Gamma) k_B T n$, where $\lambda(\Gamma)$ is a slowly rising function of Γ, saturating around 0.1.[11] Then, by virtue of the equipartition between kinetic and potential energy,

$$\tfrac{1}{2} m \omega_p^2 s^2 = \lambda(\Gamma) k_B T \ ,$$

leading to

$$\frac{s}{a}=\left[\frac{2\lambda(\Gamma)}{3\Gamma}\right]^{1/2}\ll 1 .$$

In the presence of an external perturbation, further deviations from the quasiequilibrium positions result, whose analysis provides the basis for the determination of the response functions. In the spirit of the conventional harmonic approximation for phonons, it will be assumed that the amplitudes of the total excursions are small and can be described in terms of a linear analysis. Since the equilibrium excursions have already been shown to be small,this assumption is consistent with the standard formalism of linear response theory. However, we will further assume that the equilibrium oscillations are negligible and that the excursion amplitude is proportional to the perturbation. This amounts to neglecting direct thermal effects against correlation effects in the response: for strong coupling this is a good approximation since the former is of $O(\Gamma^{-1})$ times the latter.[6] The distinction between the "direct" and "indirect" thermal effects inherent in the model should be emphasized. The indirect thermal effects refer to the accessibility of the possible configurations of the random sites and to the temperature dependence of the probability of a particular configuration: this aspect is well represented in the present model through the Γ dependence of $g(r)$. On the other hand, direct thermal effects are responsible for the actual motion and migration of the particles and give rise, e.g., to the $3k^2\langle v^2\rangle$ Bohm-Gross term in the ω^{-4} sum rule coefficient: this effect is not taken into account in the present model, although subsequent papers will explore the phenomenological inclusion of the direct thermal effects in $\epsilon(\mathbf{k}\omega)$.

It should be further realized that even though the approximation is expected to provide a good description of the dispersion of the collective modes, it is incapable of handling their damping or dissipation in general. Plasmon damping, in general, originates from plasmon decay into particle pairs (a collisional effect) or from plasmon-plasmon (or plasmon-sonon, etc.) interaction.[12] The former is absent from the model because particles on different sites are virtually isolated from each other. The latter is due to the inherent nonlinearity of the Coulomb interaction which is not taken into account in the harmonic approximation. That the plasmon damping decreases with increasing coupling strength for $\Gamma > 10$, as the plasmon-particle interaction loses importance, is known from 3D molecular-dynamics (MD) simulations[13] and is corroborated by theoretical calculations.[14] Thus damping does not appear to be a decisive factor for strong coupling: this is indeed what one would expect on the basis of the model of quasilocalized particles. Its calculation, in any case, is beyond the scope of the present paper and has to await further work and clarification.

Based on the physical picture of quasilocalization, we will derive relatively simple expressions for the longitudinal dielectric function $\epsilon(\mathbf{k}\omega)$ of a strongly coupled multispecies Coulomb system of arbitrary dimensionality, and we will show that the resulting dispersion relations reproduce in the known cases the strong coupling limit of dispersion relations calculated by nonperturbative methods. The present formalism, however, goes much beyond the earlier approaches. While earlier calculations[14–17] for intermediate or strong coupling were restricted to the $k\rightarrow 0$ domain, here we derive dispersion relations for arbitrary k values and can predict the lower bound of the plasmon frequency and an unexpected oscillatory behavior. For multicomponent systems, we can establish new relationships for the shift of the plasma frequency $[\omega(k=0)]$ and its random-phase approximation (RPA) values, in more satisfactory agreement with MD data[18] than earlier sum rule analysis data. The predicted shift can be quite substantial and the parameter ranges where large shifts occur correspond to experimentally realizable and interesting situations.

The plan of the paper is as follows. In Sec. II, starting from a Hamiltonian formulation, we calculate the dielectric response function for a plasma system based on the model we have described. From this parent calculation, we formulate in Secs. III–V the plasmon dispersion relations for the 3D ion OCP, the 2D electron OCP, and the binary ionic mixture. In Secs. III and IV, we calculate the OCP plasmon dispersion at finite wave numbers; in Sec. V, we calculate the ionic mixture plasma mode frequency at zero wave number. A dispersion relation for the two-component electron-ion plasma is briefly analyzed in Sec. VI. Conclusions are drawn in Sec. VII.

II. FORMALISM

Our principal purpose is to determine the response of the system to a small perturbing external potential. In order to do this, we develop a Hamiltonian for the small displacements (as dynamical coordinates) around the quasiequilibrium sites (playing the role of parameters only). Our notation is as follows: i,j,k enumerate particles; A,B,C designate different species; α,β,γ are the three-dimensional vector indices. Einstein summation convention for the repeated vector indices is understood, but repeated (or unrepeated) species indices are summed over only when barred ($\bar{A}$, etc). Now, let

$$X_{i,\alpha}^{A}(t)=x_{i,\alpha}^{A}+\xi_{i,\alpha}^{A}(t) \tag{1}$$

be the momentary position of the ith particle of species A, $\mathbf{x}_i^A$ its average quasiequilibrium position, and $\xi_i^A(t)$ the amplitude of its small excursion. The (partial) Hamiltonian appropriate for the description of the dynamical processes is

$$H=\frac{1}{2m^{\bar{A}}}\sum_{i}\pi_{i,\alpha}^{\bar{A}}\pi_{i,\alpha}^{\bar{A}}+\frac{1}{2}\sum_{i,j}K_{ij,\alpha\beta}^{\bar{A}\bar{B}}\xi_{i,\alpha}^{\bar{A}}\xi_{j,\beta}^{\bar{B}}+\sum_{i}\xi_{i,\alpha}^{\bar{A}}\frac{\partial}{\partial x_{i,\alpha}^{\bar{A}}}\hat{\Phi}^{\bar{A}}(\mathbf{x}_i^{\bar{A}},t) , \tag{2}$$

where $K_{ij,\alpha\beta}^{AB}$ is given by

$$K_{ij,\alpha\beta}^{AB}=(1-\delta^{AB}\delta_{ij})\frac{\partial^2}{\partial x_{i,\alpha}^{A}\partial x_{j,\beta}^{B}}\phi^{AB}(|\mathbf{x}_i^A-\mathbf{x}_j^B|)$$
$$-\delta^{AB}\delta_{ij}\left[\sum_l(1-\delta^{A\bar{C}}\delta_{il})\frac{\partial^2}{\partial x_{i,\alpha}^{A}\partial x_{l\beta}^{\bar{C}}}\phi^{A\bar{C}}(|\mathbf{x}_i^A-\mathbf{x}_l^{\bar{C}}|)+\frac{N_b}{V_d}\int d^d y^b\frac{\partial^2}{\partial x_{i,\alpha}^{A}\partial y_{\beta}^{b}}\phi^{Ab}(|\mathbf{x}_i^A-\mathbf{y}^b|)\right],$$
$$\phi^{AB}(r)=\frac{Z^A Z^B e^2}{r}. \tag{3}$$

This Hamiltonian, formally similar to that of a multicomponent crystalline solid, describes a system of charged particles of one or more species ($A,B,C=1,2,\ldots$) embedded, if needed, in a uniform background (b) of opposite charge which ensures overall electrical neutrality. [$\hat{\Phi}^A$ is the potential energy, in the external potential, of a particle of species A; $\pi_{i,\alpha}^A=m_A\dot{\xi}_{i,\alpha}^A$ is the momentum conjugate to $\xi_{i,\alpha}^A$; N_b is the total number of background particles; V_d is the large but bounded volume of the d-dimensional ($d=2,3$) system. The charge neutrality condition requires $N_b Z^b=-\sum_A N_A Z^A$.] Note the characteristic separation of the Hamiltonian into off-diagonal ($1-\delta^{AB}\delta_{ij}$) and diagonal ($\delta^{AB}\delta_{ij}$) contributions (δ^{AB} and δ_{ij} are Kronecker deltas); this latter originates from the displacement of a particle in a fixed environment of the other particles, while the former originates from the fluctuating environment.

We now introduce the collective coordinates in Fourier representation, $\xi_{\mathbf{k},\alpha}^A$ and $\pi_{\mathbf{k},\alpha}^A$, in terms of which

$$\xi_{i,\alpha}^A=\frac{1}{(N_A m_A)^{1/2}}\sum_{\mathbf{k}}\xi_{\mathbf{k},\alpha}^A e^{i\mathbf{k}\cdot\mathbf{x}_i^A},$$
$$\pi_{i,\alpha}^A=\left[\frac{m_A}{N_A}\right]^{1/2}\sum_{\mathbf{k}}\pi_{\mathbf{k},\alpha}^A e^{i\mathbf{k}\cdot\mathbf{x}_i^A}. \tag{4}$$

These are formally similar to the coordinates used in the harmonic approximation of lattice vibration. Nevertheless, in view of the lack of periodicity in the background in the present situation, the question of the transformation leading from $(\xi_{i,\alpha}^A,\pi_{i,\alpha}^A)$ to $(\xi_{\mathbf{k},\alpha}^A,\pi_{\mathbf{k},\alpha}^A)$ being a good canonical transformation needs further clarification and is discussed in the Appendix. Keeping this in mind, the Hamiltonian (2) then can be rewritten as

$$H=\frac{1}{2N_{\bar{A}}}\sum_{\mathbf{p},\mathbf{q}}\pi_{\mathbf{p},\alpha}^{\bar{A}}\pi_{\mathbf{q},\alpha}^{\bar{A}}n_{\mathbf{p}-\mathbf{q}}^{\bar{A}}$$
$$+\frac{1}{2N_{\bar{A}}N_{\bar{B}}}\sum_{\mathbf{k},\mathbf{p},\mathbf{q}}\frac{k_\alpha k_\beta}{k^2}\omega^{\bar{A}}(k)\omega^{\bar{B}}(k)\left[n_{\mathbf{k}-\mathbf{p}}^{\bar{A}}n_{\mathbf{k}-\mathbf{q}}^{\bar{B}}-\delta^{\bar{A}\bar{B}}\left[\frac{Z^{\bar{C}}}{Z^{\bar{A}}}n_{\mathbf{k}-\mathbf{p}-\mathbf{q}}^{\bar{A}}n_{\mathbf{k}}^{\bar{C}}+\frac{Z^b}{Z^{\bar{A}}}N_b\delta_{\mathbf{k}}n_{\mathbf{p}-\mathbf{q}}^{\bar{A}}\right]\right]\xi_{\mathbf{p},\alpha}^{\bar{A}}\xi_{\mathbf{q},\beta}^{\bar{B}}$$
$$+\frac{i}{V_d}\frac{1}{(N_{\bar{A}}m_{\bar{A}})^{1/2}}\sum_{\mathbf{p},\mathbf{q}}q_\alpha\hat{\Phi}^{\bar{A}}(\mathbf{q},t)n_{\mathbf{p}-\mathbf{q}}^{\bar{A}}\xi_{\mathbf{p},\alpha}^{\bar{A}}, \tag{5}$$

where

$$\omega^A=\begin{cases}Z^A e\left[\dfrac{4\pi N_A}{m_A V_3}\right]^{1/2} & \text{for 3D}\\ Z^A e\left[\dfrac{2\pi N_A k}{m_A V_2}\right]^{1/2} & \text{for 2D}\end{cases}$$

is the (signed) d-dimensional plasma frequency of species A; the unperturbed ("base") density

$$n_{\mathbf{k}}^A=\sum_{i=1}^{N_A}e^{i\mathbf{k}\cdot\mathbf{x}_i^A} \tag{6}$$

depends on the $\mathbf{x}_i,\mathbf{x}_j$, etc.—coordinates which are not dynamical variables. Our principal approximation now consists of replacing this random base by its ensemble average. This latter is evaluated through

$$\langle n_{\mathbf{k}}^A\rangle=N_A\delta_{\mathbf{k}},$$
$$\langle n_{\mathbf{p}}^A n_{\mathbf{q}}^B\rangle=N_A N_B\delta_{\mathbf{p}+\mathbf{q}}\left[\delta_{\mathbf{q}}+\frac{1}{V_d}g^{AB}(\mathbf{q})\right]$$
$$+N_A\delta^{AB}\delta_{\mathbf{p}+\mathbf{q}}, \tag{7}$$

where $g^{AB}(q)$ is the Fourier transform of the equilibrium pair-correlation function $g^{AB}(r)$ for an (A,B) pair. The resulting new Hamilton is

$$\mathcal{H}=\tfrac{1}{2}\sum_{\mathbf{k}}\pi_{\mathbf{k},\alpha}^{\bar{A}}\pi_{-\mathbf{k},\alpha}^{\bar{A}}$$
$$+\tfrac{1}{2}\sum_{\mathbf{k}}\left[D_{\alpha\beta}^{\bar{A}\bar{B}}(\mathbf{k})+\frac{k_\alpha k_\beta}{k^2}\omega^{\bar{A}}(k)\omega^{\bar{B}}(k)\right]\xi_{\mathbf{k},\alpha}^{\bar{A}}\xi_{-\mathbf{k},\beta}^{\bar{B}}$$
$$-\frac{i}{V_d}\left[\frac{N_{\bar{A}}}{m_{\bar{A}}}\right]^{1/2}\sum_{\mathbf{k}}k_\alpha\hat{\Phi}^{\bar{A}}(-\mathbf{k},t)\xi_{\mathbf{k}\alpha}^{\bar{A}}, \tag{8}$$

where

$$D_{\alpha\beta}^{AB}(\mathbf{k})=\frac{1}{V_d}\sum_{\mathbf{q}}\frac{q_\alpha q_\beta}{q^2}\omega^A(q)\omega^B(q) \times\left[g^{AB}(\mathbf{k}-\mathbf{q})-\delta^{AB}\frac{N_{\bar{C}}Z^{\bar{C}}}{N_A Z^A}g^{A\bar{C}}(\mathbf{q})\right] . \tag{9}$$

Note that the background term has been canceled by a contribution originating from the $\delta_{\mathbf{q}}$ term in (7) due to overall charge neutrality.

The routine calculation of $\ddot{\xi}_{\mathbf{k},\alpha}^A(t)$ from Hamilton's equation yields

$$\ddot{\xi}_{\mathbf{k},\alpha}^A=\dot{\pi}_{\mathbf{k},\alpha}^A=-\frac{\partial\mathcal{H}}{\partial\xi_{-\mathbf{k},\alpha}^A} =\left[-D_{\alpha\beta}^{A\bar{B}}(\mathbf{k})+\omega^A(k)\omega^{\bar{B}}(k)\frac{k_\alpha k_\beta}{k^2}\right]\xi_{\mathbf{k},\beta}^{\bar{B}} -\frac{i}{V_d}\left[\frac{N_A}{m_A}\right]^{1/2}k_\alpha\hat{\Phi}^A(\mathbf{k},t) . \tag{10}$$

$\xi_{\mathbf{k},\alpha}^A(\omega)$, the Fourier transform of $\xi_{\mathbf{k},\alpha}^A$, can then be expressed as

$$\xi_{\mathbf{k},\alpha}^A(\omega)=\frac{i}{V_d}\left[\frac{N_{\bar{B}}}{m_{\bar{B}}}\right]^{1/2}k_\beta(\Gamma^{-1})_{\alpha\beta}^{A\bar{B}}(\mathbf{k}\omega)\hat{\Phi}^{\bar{B}}(\mathbf{k}\omega) , \tag{11}$$

where the propagator Γ^{-1} for the external perturbation is the inverse of the matrix

$$\Gamma_{\alpha\beta}^{AB}(\mathbf{k}\omega)=\omega^2\delta^{AB}\delta_{\alpha\beta}-\omega^A(k)\omega^B(k)\frac{k_\alpha k_\beta}{k^2}-D_{\alpha\beta}^{AB}(\mathbf{k}) . \tag{12}$$

We now consider the Fourier component of the perturbed microscopic density

$$\rho_{\mathbf{k}}^A=-ik_\alpha\frac{1}{(N_A m_A)^{1/2}}\sum_{\mathbf{q}}n_{\mathbf{k}-\mathbf{q}}^A\xi_{\mathbf{q},\alpha}^A . \tag{13}$$

Its equilibrium average $\rho^A(\mathbf{k}\omega)=\langle\rho_{\mathbf{k}}^A\rangle$ can be calculated by averaging only over $n_{\mathbf{k}-\mathbf{q}}^A$ in (13), since the $\xi_{\mathbf{q},\alpha}^A$, by virtue of the assumption used in deriving $\mathcal{H}$, is independent of the $\mathbf{x}_i$ coordinates. Substituting (10) into (13) and averaging, we obtain

$$\rho^A(\mathbf{k}\omega)=\frac{1}{V_d}\left[\frac{N_A N_{\bar{B}}}{m_A m_{\bar{B}}}\right]^{1/2}k_\alpha k_\beta(\Gamma^{-1})_{\alpha\beta}^{A\bar{B}}\hat{\Phi}^{\bar{B}}(\mathbf{k}\omega) . \tag{14}$$

We now use the formalism of partial response functions[19] to calculate partial density response functions $\hat{\chi}^{AB}$ (to the external potential) and χ^{AB} (to the total potential), the dielectric matrix ϵ^{AB} and the dielectric function ϵ. The definition of $\hat{\chi}^{AB}$

$$\rho^A(\mathbf{k}\omega)=\hat{\chi}^{A\bar{B}}(\mathbf{k}\omega)\hat{\Phi}^{\bar{B}}(\mathbf{k}\omega) , \tag{15}$$

compared with (14), provides an expression for $\hat{\chi}^{AB}$:

$$\hat{\chi}^{AB}(\mathbf{k}\omega)=\frac{1}{V_d}\left[\frac{N_A N_B}{m_A m_B}\right]^{1/2}k_\alpha k_\beta(\Gamma^{-1})_{\alpha\beta}^{AB}(\mathbf{k}\omega) . \tag{16}$$

χ^{AB}, on the other hand, with

$$\rho^A(\mathbf{k}\omega)=\chi^{A\bar{B}}(\mathbf{k}\omega)\Phi^{\bar{B}}(\mathbf{k}\omega) , \tag{17}$$

can be obtained from (16) as

$$\chi^{AB}(\mathbf{k}\omega)=\frac{1}{V_d}\left[\frac{N_A N_B}{m_A m_B}\right]^{1/2}k^2\Delta^{-1\,AB}(\mathbf{k}\omega) . \tag{18}$$

Here the Δ^{-1} propagator for the total longitudinal field perturbation is the inverse of the matrix

$$\Delta^{AB}(\mathbf{k}\omega)=\omega^2\delta^{AB}-D^{AB}(\mathbf{k}) , \qquad D^{AB}(\mathbf{k})=\frac{k_\alpha D_{\alpha\beta}^{AB}(\mathbf{k})k_\beta}{k^2} . \tag{19}$$

The standard procedure[19] now provides ϵ^{AB} as

$$\epsilon^{AB}(\mathbf{k}\omega)=\delta^{AB}-\phi^{A\bar{C}}(\mathbf{k})\chi^{\bar{C}B}(\mathbf{k}\omega) , \tag{20}$$

where $\phi^{AB}(\mathbf{k})$ is the interaction potential between species A and B. Here

$$\phi^{AB}(\mathbf{k})=\begin{cases}Z^A Z^B\dfrac{4\pi e^2}{k^2} & \text{for 3D}\\ Z^A Z^B\dfrac{2\pi e^2}{k} & \text{for 2D}\end{cases}$$

and thus (19) becomes

$$\epsilon^{AB}(\mathbf{k}\omega)=\delta^{AB}-\omega^A(k)\omega^{\bar{C}}(k)\Delta^{-1\,\bar{C}B}(\mathbf{k}\omega) . \tag{21}$$

The physical dielectric function $\epsilon(\mathbf{k}\omega)$ is given by[19]

$$\epsilon(\mathbf{k}\omega)=1-\phi^{\bar{A}\bar{B}}(\mathbf{k})\chi^{\bar{B}\bar{A}}(\mathbf{k}\omega) =1-\omega^{\bar{A}}(k)\Delta^{-1\,\bar{A}\bar{B}}(\mathbf{k}\omega)\omega^{\bar{B}}(k) . \tag{22}$$

The dispersion relation now can be obtained in general from

$$\det\epsilon^{AB}(\mathbf{k}\omega)=0 , \tag{23}$$

or for the Coulomb potential, from the more familiar

$$\epsilon(\mathbf{k}\omega)=0 \tag{24}$$

relation. Equation (22) is a new and general result for the dielectric function of a strongly coupled Coulomb liquid of an arbitrary number of species and of any dimensionality. Application to particular systems will be discussed in the sequel.

It is clear from Eq. (22) that $\epsilon(\mathbf{k}\omega)$ is real and thus so are the frequencies satisfying the dispersion relation. In other words, only the dispersion, but not the damping of the collective modes, can be determined from the present theory. Nevertheless, some speculation on the way damping would appear in a more extended description is in order. As we have already noted, plasmon damping, in general, originates from plasmon decay into particle pairs and from plasmon-plasmon (or plasmon-sonon, etc.) interaction.[12] In the physical situation in which the present model is appropriate, the main mechanism seems to be the latter. More precisely, plasmon splitting, al-

lowed by energy-momentum conservation, is probably the most dominant process. Thus we expect a small, but finite damping: its calculation, however, is not part of the present work.

We will apply formulas (21) and (19) to three uniform-background situations: the 3D positive ion OCP, the 2D electron OCP, and the binary ionic mixture. These formulas can also be applied o the two-component electron-ion plasma with the stipulation that the bare Coulomb potential is to be modified to incorporate softening of the Coulomb interaction as a result of quantum-mechanical effects, and to eliminate the divergence of $g(r)$ or $r \to 0$.

III. IONIC OCP

The classical (three-dimensional) one-component plasma (OCP: particles of charge Ze embedded in an inert neutralizing background) is a widely used model for the study of the fundamental properties of Coulomb systems. There is a strong indication[6] that the OCP crystallizes at $\Gamma_m = 178 \pm 1$. Specifying the charges to be ions and the background to be that of rigid degenerate electrons, the OCP provides as well a workable model for certain astrophysical situations (planetary interiors, white dwarfs, neutron star crusts) and it also faithfully describes recent experimental situations[20] consisting of $^9Be^+$ ions trapped in a Penning trap. The present model should be an adequate description for the strongly coupled liquid state ($15 < \Gamma < 178$; see below for the lower bound estimate) and for the possible supercooled[21] ($178 < \Gamma < 400$) and conjectured amorphous glassy[22] ($\Gamma > 300$) states.

The OCP dispersion relation for intermediate and strong coupling has been calculated by Carini, Kalman, and Golden[14] for small $\mathbf{k}$ values. Good agreement with the MD data of Hansen and co-workers[13,23] was found; in particular, the critical Γ value for the onset of the negative dispersion ($\Gamma \simeq 9$) was accurately predicted. There is no information, though, in these calculations on the manner in which the descending portion of the dispersion curve terminates (for an estimate, see, however Ref. 24). An asymptotic analysis of the dispersion relation in the present work provides a prediction for this; even though refinement of the model[25] somewhat alters the predicted value, it certainly can be accepted as a reliable lower bound.

The general expression (22) for the dielectric function $\epsilon(\mathbf{k}\omega)$, in the case of the 3D OCP, simplifies to

$$\epsilon(\mathbf{k}\omega) = 1 - \frac{\omega_p^2}{\omega^2 - \omega_p^2 \mathcal{D}(\mathbf{k})} , \tag{25}$$

where $\omega_p = (4\pi n Z^2 e^2 / m_i)^{1/2}$ is the 3D plasma frequency ($n = N_i / V$) and

$$\mathcal{D}(\mathbf{k}) = \frac{1}{V} \sum_{\mathbf{q}} \left[\frac{\mathbf{k}\cdot\mathbf{q}}{kq} \right]^2 [g(\mathbf{k}-\mathbf{q}) - g(\mathbf{q})] . \tag{26}$$

We contend that Eq. (25), in conjunction with Eq. (26), provides the most reliable description in the $\omega \approx \omega_p$ domain of the dielectric function of a strongly coupled OCP. The expressions (25) obviously satisfies the third-moment high-frequency sum rule for $\Gamma \gg 1$, since $-\omega_p^4 \mathcal{D}(\mathbf{k})$ is precisely the required correlational contribution to the coefficient of ω^{-4} in the high-frequency expansion of $\epsilon(\mathbf{k}\omega)$.

The dispersion relation $\epsilon(\mathbf{k}\omega) = 0$ gives

$$\omega^2(\mathbf{k}) = \omega_p^2 [1 + \mathcal{D}(\mathbf{k})] . \tag{27}$$

In contrast to earlier calculations[2,14] on 3D plasmon dispersion restricted to long wavelengths ($k \to 0$), Eqs. (25) and (27) are valid for arbitrary values of k.

For $k \to 0$, $\mathcal{D}(k)$ becomes

$$\mathcal{D}(k \to 0) = \frac{4}{45} \frac{\beta E_c}{n\Gamma} k^2 a^2 , \tag{28}$$

where a is the ion sphere radius, $\beta = 1/k_B T$, and E_c is the correlation energy density. Since $E_c < 0$, (28) provides a negative dispersion in agreement with MD data.[23] The theoretical calculations of Carini, Kalman, and Golden[14] have also shown that for $\Gamma \to \infty$ and $k \to 0$, the plasmon dispersion is described by (27) and (28). We can use the results of Ref. 14 also to estimate the Γ value above which (27) constitutes a good approximation. The asymptotic series for $\Gamma \to \infty$ gives

$$\omega = \omega_p \left[1 - \left[0.04 - \frac{0.5}{\Gamma} - \frac{1}{\Gamma^{7/2}} \right] k^2 a^2 \right] . \tag{29}$$

The leading term corresponding to (28) dominates over the next thermal term for about $\Gamma > 15$, suggesting this latter Γ value as a lower bound beyond which the present theory gives a good approximation. Moreover, for $k \ll \kappa = (4\pi n Z^2 e^2 \beta)^{1/2}$, the high-frequency expansion of $\epsilon(\mathbf{k}\omega)$ is manifestly equivalent to the almost exact (25) for arbitrary frequencies: this justifies the heuristic observation[15] that the high-frequency sum-rule expansion provides a good representation of $\epsilon(\mathbf{k}\omega)$ in the $\Gamma \to \infty$ (i.e., $\Gamma > 15$) limit.

For large values, $\mathcal{D}$ reaches an asymptotic limit

$$\mathcal{D}(k \to \infty) = -\tfrac{2}{3} . \tag{30}$$

Thus for $ka \gg 1$, the behavior of $\mathcal{D}(k)$, as also calculated from HNC data,[9] shows a slower than k^2 dependence. Evidently, and as we have already conjectured,[24] for large k, the plasmon dispersion is always dominated by the thermal k^2 term.

The above asymptotic limit is not reached monotonically: a series of oscillations develops for sufficiently high Γ values. Details of this behavior are discussed in a forthcoming paper.[25]

For $k \to 0$,

$$\omega(k \to 0) = \omega_p \left[1 + \frac{2}{45} \frac{\beta E_c}{n\Gamma} k^2 a^2 \right] , \tag{31}$$

which, if E_c is expressed through the fitted MC formula,[6]

$$\frac{\beta E_c}{n} = -0.898\Gamma + 0.950\Gamma^{1/4} + 0.190 ,$$

becomes

$$\omega(k \to 0) = \omega_p [1 - (0.0399 - 0.0422\Gamma^{-3/4} - 0.0084) k^2 a^2 \Gamma^{-1}] . \tag{31'}$$

For $k \to \infty$,

$$\omega(k \to \infty) = 0.577\omega_p \ , \tag{32}$$

in agreement with the already noted behavior of $\mathcal{D}(\mathbf{k})$.

The static $\epsilon(\mathbf{k}) = \epsilon(\mathbf{k}\omega = 0)$ as determined from (25),

$$\epsilon(\mathbf{k}) = 1 + \frac{1}{\mathcal{D}(\mathbf{k})} \ , \tag{33}$$

is quite different from the screened Debye-type $\epsilon(\mathbf{k})$, and it resembles more the $\epsilon(\mathbf{k})$ of a Wigner lattice, calculated by Bragchi.[26] The data of Rogers *et al.*[9] on the static structure factor $S(\mathbf{k})$ allow one to trace $\epsilon(\mathbf{k})$ by using the fluctuation-dissipation relationship

$$S(\mathbf{k}) = \frac{k^2 a^2}{3\Gamma}\left[1 - \frac{1}{\epsilon(\mathbf{k})}\right] . \tag{34}$$

The results show[24] that there exists a $k_*(\Gamma)$ such that $\epsilon(\mathbf{k}) < 0$ for $k < k_*$ in the high-Γ domain. Comparison with (33) in this domain reveals a qualitative agreement (although with an incorrect numerical coefficient, as can be ascertained by comparison with the requirement of the compressibility sum rule). For $k > k_*$, however, $\epsilon(\mathbf{k})$ from (33) remains negative for all values of k while the actual $\epsilon(k)$, from (34), is positive; the $\epsilon(k) > 0$ behavior can be expected to be recoved only if the direct effect of the thermal motion is correctly included in the model.

IV. 2D ELECTRON OCP

The two-dimensional version of the OCP (with $1/r$ interaction) provides a model for a variety of physical systems, consisting of strongly correlated electrons trapped in surface-bound states at the interface of dielectric materials (e.g., bound electrons in the ground state of a potential well formed above the free surface by an attractive image potential and a repulsive surface barrier. The potential energy of an (ij) pair is given by $\phi(r_{ij}) = e^2/r_{ij}$, where e is a renormalized charge which incorporates the effects of the dielectric substrate. With the experimentally realized density values [e.g., $\beta E_F = \beta\pi n/(2m) \leq 0.06$ ($n = N_e/A$ is the electron areal density) in the Grimes-Adams experiments[4]] the system, in fact, is adequately described as a classical 2D OCP.

There is evidence both from MC data[8] and from experiments[4] that the 2D OCP crystallizes at $\Gamma_m = 137 \pm 15$. Dispersion relation calculations for the plasmon mode in the 2D OCP have been carried out both for the liquid and the solid state. An RPA-like model was developed by Platzman and Tzoar;[27] a genuine treatment of the strong correlations via the static mean-field theory[1] is due to Studart and Hipolito,[28] while the more satisfactory dynamical mean-field theory description is given by Golden and Lu:[16] the latter work, however, is restricted to small k values, whereas the former is not. The dispersion in the hexagonally crystallized solid state[29,30] is isotropic to $O(k^2)$ and thus serves as a required limit for the liquid dispersion as $\Gamma \to \Gamma_m$: the calculation of Ref. 26 fails to reproduce this, while that of Ref. 16, as well as the results of the present work, do accurately provide this limit.

For the 2D OCP, Eq. (22) simplifies to

$$\epsilon(\mathbf{k}\omega) = 1 - \frac{\omega_p^2(k)}{\omega^2 - \omega_p^2(k)\mathcal{D}(\mathbf{k})} \ , \tag{35}$$

where $\omega_p(k) = (2\pi n e^2 k/m_e)^{1/2}$ is the 2D (k-dependent) plasma frequency and

$$\mathcal{D}(\mathbf{k}) = \frac{1}{A}\sum_{\mathbf{q}} \frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^3 q}[g(\mathbf{k}-\mathbf{q}) - g(\mathbf{q})] \ . \tag{36}$$

As in the 3D case, Eq. (35) satisfies the ω^{-4} high-frequency sum rule. The dispersion relation $\epsilon(\mathbf{k}\omega) = 0$ gives, similarly to (27),

$$\omega^2 = \omega_p^2(k)[1 + \mathcal{D}(\mathbf{k})] \ . \tag{37}$$

Equations (35)–(37) are valid for arbitrary values of $\mathbf{k}$.

For $k \to 0$,

$$\mathcal{D}(k \to 0) = \frac{5}{16}\frac{\beta E_c}{n\Gamma} ka \ , \tag{38}$$

where a is the electron circle radius; again note the lowering of the frequency compared to its RPA value, in analogy with the 3D case. In the $\Gamma \gg 1$ domain we can use the HNC formula[10]

$$\frac{\beta E_c}{n} = -1.095\Gamma + 0.985 \quad (\Gamma > 30) \tag{39}$$

which, substituted into (38), yields

$$\omega(k \to 0) = \omega_p(k)\left[1 - \left[0.171 - \frac{0.154}{\Gamma}\right] ka\right] . \tag{40}$$

In order to estimate the Γ value, above which (37) constitutes a good approximation, one can compare the leading term of (40) with the $(3/4\Gamma)$ thermal Bohm-Gross term in the RPA dispersion relation. The leading term dominates over the thermal term for about $\Gamma > 5$, suggesting the latter Γ value beyond which the present theory gives a good approximation. For $k \ll \kappa = 2\pi n e^2 \beta$, the high-frequency expansion of $\epsilon(\mathbf{k}\omega)$ is manifestly equivalent to the almost exact (35) for arbitrary frequencies. This is consistent with an earlier observation[16] that it is the correlational contribution to the ω^{-4} sum rule which controls the long-wavelength $k \to 0$ dispersion of the plasmon excitations in the $\Gamma \to \Gamma_m$ (i.e., $\Gamma > 5$) limit.

With increasing k, $\omega(k)$ increases to a maximum whose value as $\Gamma \to \Gamma_m$ is

$$\omega_{\max} = 0.874\omega_0 \equiv 0.874\left[\frac{2e^2}{ma^3}\right]^{1/2} \quad \text{at } ka = 1.60 \ . \tag{41}$$

Thereafter, $\omega(k)$ descents through a series of oscillations to an asymptotic value which is determined by

$$\mathcal{D}(k \to \infty) = -1 - \lim_{k\to\infty} \frac{1}{4\pi k}\int_0^\infty dq\, q^2 g(q) \ , \tag{42}$$

whence

$$\omega^2(k \to \infty, \Gamma) = -\frac{1}{2A}\sum_q \omega_p^2(q) g(q)$$
$$= -\tfrac{1}{4}\omega_0^2 \int_0^\infty dx\, x^2 n g(x), \quad x = qa \ . \tag{43}$$

The right-hand-side integral is bounded; we find that

$$-\int_0^\infty dx\, x^2 n g(x) \simeq 1.46 \quad \text{for } \Gamma > 90 \tag{44}$$

leading to the asymptotic value

$$\omega(k \to \infty) = 0.604\omega_0 \ . \tag{45}$$

More detailed analysis of the 2D OCP dispersion relation with the inclusion of direct thermal effects, will be the subject of another forthcoming paper.[31]

V. BINARY IONIC MIXTURE

The binary ionic mixture consisting of two ion species (with different charge, mass, and density values) embedded in a neutralizing background, as a model, is the natural extension of the OCP. The system is conveniently characterized by the total density $n = n_1 + n_2$, the concentrations $c_1 = n_1/n$, $c_2 = n_2/n$, charge averages $\langle Z^r \rangle = c_1 Z_1^r + c_2 Z_2^r$ and $\Gamma^0 = \beta e^2/a$, $(4\pi/3)a^3 n = 1$. It will be also useful to introduce the two asymmetry parameters

$$x = \frac{Z_2 m_1}{Z_1 m_2}, \quad y = \frac{Z_2 c_2}{Z_1 c_1} \ . \tag{46}$$

The precise value of Γ^0 where crytallization occurs is not known, but scaling arguments suggest that

$$\Gamma_m^0 \langle Z^{5/3} \rangle \langle Z \rangle^{1/3} = \Gamma_m^{\rm OCP} \ .$$

For a binary system, $D^{AB}(\mathbf{k})$ is a 2×2 matrix with

$$D^{AB}(\mathbf{k}) = \omega^A \omega^B \mathcal{D}^{AB}(\mathbf{k}) \ . \tag{47}$$

The matrix elements are

$$\begin{aligned} \mathcal{D}^{11}(\mathbf{k}) &= \frac{1}{V}\sum_{\mathbf{q}} \left[\frac{\mathbf{k}\cdot\mathbf{q}}{kq}\right]^2 [g^{11}(\mathbf{k}-\mathbf{q}) - g^{11}(\mathbf{q}) + y g^{12}(\mathbf{q})] \ , \\ \mathcal{D}^{12}(\mathbf{k}) &= \frac{1}{V}\sum_{\mathbf{q}} \left[\frac{\mathbf{k}\cdot\mathbf{q}}{kq}\right]^2 g^{12}(\mathbf{k}-\mathbf{q}) \ , \end{aligned} \tag{48}$$

etc.

From (22) and (48) above, we then find that

$$\epsilon(\mathbf{k}\omega) = 1 - \frac{A\omega^2 - B}{\omega^4 - C\omega^2 + D} \ , \tag{49}$$

$$\begin{aligned} A &= \omega_1^2 + \omega_2^2 = \frac{4\pi n_1 (Z_1 e)^2}{m_1} + \frac{4\pi n_2 (Z_2 e)^2}{m_2} = \omega_p^2 \ , \\ B &= \omega_1^2 \omega_2^2 [\tfrac{1}{3}(y^{1/2} + y^{-1/2})^2 + W_{11}(\mathbf{k}) \\ &\quad - 2W_{12}(\mathbf{k}) + W_{22}(\mathbf{k})] \ , \\ C &= \tfrac{1}{3}\omega_1\omega_2(\sqrt{x/y} + \sqrt{y/x}) + \omega_1^2 W_{11}(\mathbf{k}) + \omega_2^2 W_{22}(\mathbf{k}) \ , \\ D &= \omega_1^2\omega_2^2 \left[\tfrac{1}{3}\left[\frac{1}{y} W_{11}(\mathbf{k}) + 2W_{12}(\mathbf{k}) + y W_{22}(\mathbf{k})\right] \right. \\ &\quad \left. + W_{11}(\mathbf{k}) W_{22}(\mathbf{k}) - W_{12}^2(\mathbf{k})\right] \ , \end{aligned} \tag{50}$$

where

$$W_{AB}(\mathbf{k}) = \frac{1}{V}\sum_{\mathbf{q}} \left[\frac{\mathbf{k}\cdot\mathbf{q}}{kq}\right]^2 [g^{AB}(\mathbf{k}-\mathbf{q}) - g^{AB}(\mathbf{q})] \ , \tag{51}$$

and we have exploited the fact that, in view of the repulsive interaction between the two species, all the pair-distribution functions $1 + g^{AB}(r)$ vanish at $r = 0$, whence

$$\frac{1}{V}\sum_{\mathbf{q}} g^{AB}(\mathbf{q}) = -1 \ . \tag{52}$$

The coefficient of ω^{-4} in the high-frequency expansion of (49) is easily calculated as

$$\omega_p^4 d^2 + \omega_1^4 W_{11}(\mathbf{k}) + 2\omega_1^2\omega_2^2 W_{12}(\mathbf{k}) + \omega_2^4 W_{22}(\mathbf{k}) \ , \tag{53}$$

with

$$d = \sqrt{y/3}\,\frac{1-x}{1+xy} \ . \tag{54}$$

This result is in agreement with those quoted in the literature.[17,18]

In the $k \to 0$ limit, (53) can be expressed in terms of the partial correlation energy densities, E_{AB}, as

$$\omega_p^4 \left[d^2 + \tfrac{4}{45}[(xy)^{1/2} + (xy)^{-1/2}]^{-2} \times \frac{\beta}{\nu n \Gamma^0}(x^{-1}E_{11} + E_{12} + xE_{22}) k^2 a^2 \right] \ , \tag{55}$$

$$\nu = Z_1 Z_2 c_1 c_2 \ ,$$

$$E_{11} = \tfrac{1}{2} n_1^2 \frac{1}{V}\sum_{\mathbf{k}} \phi_{11}(\mathbf{k}) g_{11}(\mathbf{k}) \ ,$$

$$E_{12} = n_1 n_2 \frac{1}{V}\sum_{\mathbf{k}} \phi_{12}(\mathbf{k}) g_{12}(\mathbf{k}) \ ,$$

etc. Note that, in general, the sum-rule coefficient cannot be expressed in terms of the total correlation energy density.

We turn now to the analysis of the dispersion relation. In contrast to the OCP, for a multicomponent sytem, the plasmon frequency deviates from the nominal (RPA) plasma frequency ω_p even for $k = 0$. We focus now on the magnitude of this $\omega(k=0)$ shift, as a function of the asymmetry between the two ion species.

For $k = 0$, the dispersion relation becomes

$$\omega^2 = \frac{\omega_p^2}{2}\{1 + p \pm [(1-p)^2 + 4d^2]^{1/2}\} \ , \tag{56}$$

with

$$p = \frac{1}{3}\left[\frac{x+y}{1+xy}\right] \tag{57}$$

and d given by (54). The binary system possesses two longitudinal modes, corresponding to the plus and minus signs in (56).

The character of the two solutions (ω_+ and ω_-) represented by (56) depends on whether (i) $\frac{1}{3} < x < 3$, (ii) $x < \frac{1}{3}$, or (iii) $x > 3$, as shown in Fig. 1. In domain (i), $\omega_+ \geq \omega_p$ and approaches ω_p both for $c_2 \to 0$ (species 2 as impurity in a plasma of species 1) and $c_2 \to 1$ (species 1 as impurity in a plasma of species 2); $\omega_- < \omega_p$ and does not approach ω_p in either limit. In domains (ii) and (iii) one solution (say ω_1) approaches ω_p for $c_2 \to 0$, while the oth-

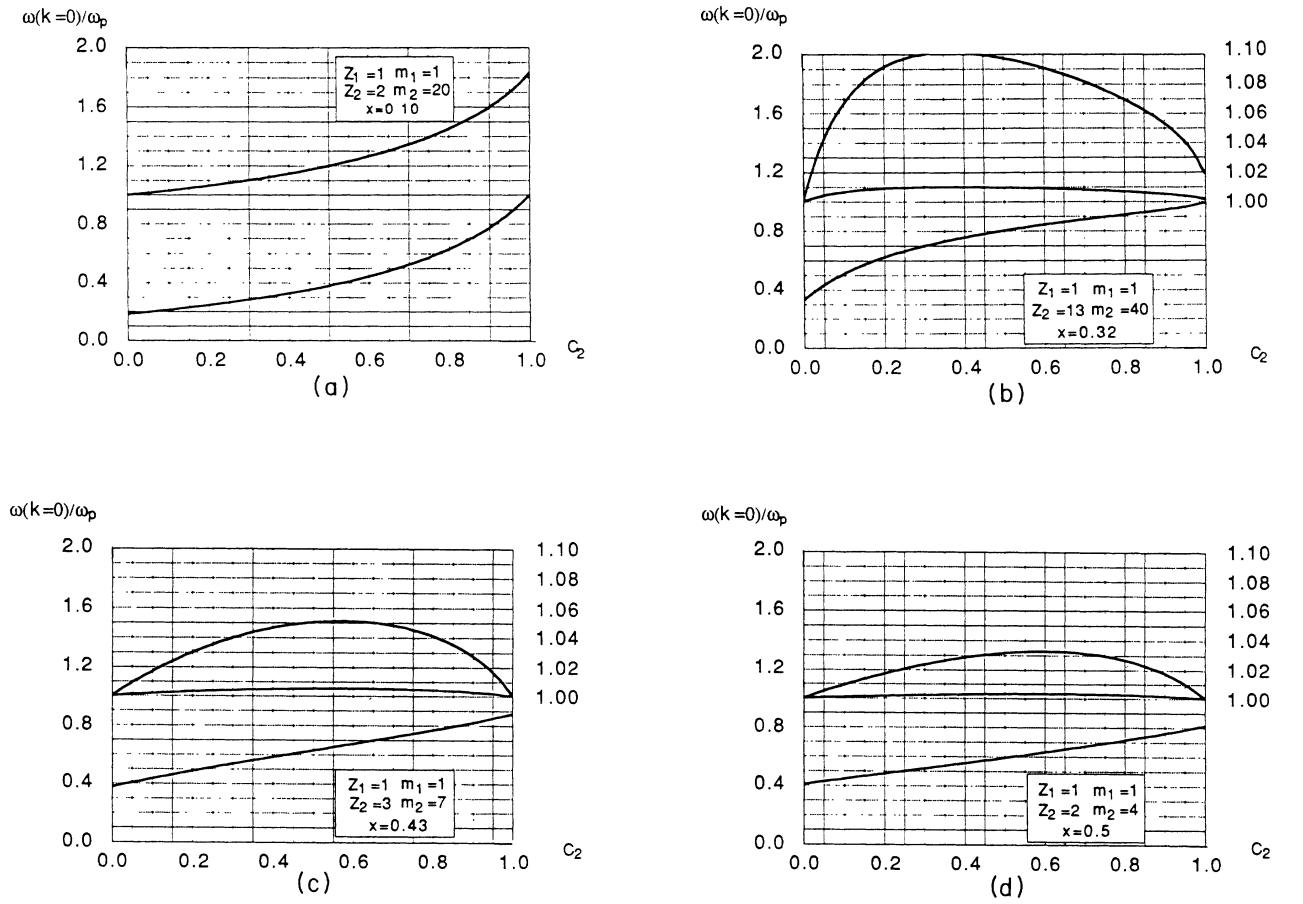

FIG. 1. Typical variations of the shift of the plasma frequency at $k=0$, in terms of the RPA plasma frequency $\omega_p=[4\pi e^2(Z_1^2 n_1/m_1+Z_2^2 n_2/m_2)]^{1/2}$ vs the concentration of the heavier species in hydrogen, c_2. Note that (c) and (d) are in domain (i) (see text), and (a) and (b) are in domain (ii); domain (iii) can be reached by interchanging species 1 and 2, and reading the diagram from the left to the right. For the explanation of the two branches see text. The uppermost curves in (b), (c), and (d) are enlarged versions of the upper branch and are to be read by the right-hand-side scale.

er (say ω_{II}) approaches ω_p for $c_2\to 1$. We have the correspondence

$$\omega_I=\omega_+,\quad \omega_{II}=\omega_- \text{ for domain (ii)}$$
$$\omega_I=\omega_-,\quad \omega_{II}=\omega_+ \text{ for domain (iii)}. \tag{58}$$

The frequencies ω_I and ω_{II} in the opposite limits approach $1/\sqrt{3x}$ and $\sqrt{x/3}$, respectively. These frequencies are independent of the impurity concentration and represent the oscillation frequency of an isolated impurity 2 (1) in the environment of the other species 1 (2).

We expect that in domain (i) ω_+ represents a physically meaningful solution for the plasmon mode for all c_2 values, and so do ω_I for $c_2\ll 1$ and ω_{II} for $c_1\ll 1$ in domains (ii) and (iii). The remaining solutions correspond to the low-frequency longitudinal mode. In the $c_1\to 0$ [for domain (ii)] and in the $c_2\to 0$ [for domain (i)] limits they are expected to be restricted to a very small region of k space and to damp out or to saturate at an extremely low amplitude; these latter effects are, of course, not described by the present theory.

It follows from the above considerations that heavy impurities in a light plasma [$x<3$; domains (i) and (ii)] cause a positive upward shift, $\omega>\omega_p$. The opposite case, light impurities in a heavy plasma ($x>3$), leads to a negative downward shift, $\omega<\omega_p$.

Sizable shifts may occur for large mass ratios ($x\to 0$) when the charge density of the heavy species greatly exceeds that of the light species ($y\gg 1$). Such a situation prevails in the presence of heavy, highly charged scatterers in a hydrogen plasma. An experimental verification of the plasmon frequency shift under these conditions may become feasible. As an illustration, Tables I–III show the plasma frequency as a function of concentration for He^{2+}, Li^{3+}, and Xe^{10+} impurities.

The more general finite k dispersion relation for the binary ionic mixture can be written in the form

$$\omega^2=\frac{\omega_p^2}{2}\{(1+p)+F(\mathbf{k}) \pm[(1-p)^2+4d^2+2(1+p)F(\mathbf{k}) +F^2(\mathbf{k})-4G(\mathbf{k})]^{1/2}\}, \tag{59}$$

TABLE I. Frequency shifts for the H^+-He^{2+} mixture: $Z_1=1$, $m_1=1$, $Z_2=2$, $m_2=4$, $x=0.5$, and $y=2n_2/n_1$, in domain (i).

n_2/n_1	c_2	ω/ω_p
0	0	1
0.01	0.01	1.001
0.10	0.09	0.009
0.50	0.33	0.023
1.00	0.50	1.032
1.50	0.60	1.033[a]

[a]Maximum.

where

$$F(\mathbf{k})=\frac{1}{1+xy}[W_{11}(\mathbf{k})+xyW_{22}(\mathbf{k})]\ ,$$

$$G(\mathbf{k})=\frac{xy}{(1+xy)^2}\left\{[W_{11}(\mathbf{k})-2W_{12}(\mathbf{k})+W_{22}(\mathbf{k})]+\frac{1}{3y}[W_{11}(\mathbf{k})+2yW_{12}(\mathbf{k})+y^2W_{22}(\mathbf{k})]+[W_{11}(\mathbf{k})W_{22}(\mathbf{k})-W_{12}^2(\mathbf{k})]\right\}\ . \quad (60)$$

Further analysis of this dispersion relation will be published in another paper.

It is important to note that, in contrast to what happens in the one-component system, the dispersion relation derived from the sum-rule expansion does not provide, even in the case when the shift from the RPA plasma frequency is small, a good approximation. Special situations, however, still render the sum-rule result equivalent to the more exact (56) or (59). For this to happen, one must have both $p \ll 1$ and $d \ll 1$; in this case, the sum-rule expansion provides a reasonable approach for $\Gamma \gg 1$. This feature has already been conjectured (although without the latter qualification) and used by Golden, Green, and Neilson.[17]

Hansen and his collaborators[18] studied the plasmon behavior for the 50–50 % H^+-He^{2+} BIM system through MD simulation. Results for $\Gamma=40$ show a 3.9% upward shift of the plasma frequency, which is higher than a 1.9–2 % shift predicted[17,18] by the sum-rule expression. Our calculated value (see Table I) 3.2% is quite close to the MD result.[18]

TABLE II. Frequency shifts for the H^+-Li^{3+} mixture: $Z_1=1$, $m_1=1$, $Z_2=3$, $m_2=7$, $x=0.428$, and $y=3n_2/n_1$, in domain (i).

n_2/n_1	c_2	ω/ω_p
0	0	1
0.01	0.01	1.002
0.10	0.09	1.016
1.0	0.50	1.051
1.33	0.57	1.0513[a]

[a]Maximum.

TABLE III. Frequency shifts for the H^+-Xe^{10+} mixture: $Z_1=1$, $m_1=1$, $Z_2=10$, $m_2=131$, $x=0.076$, and $y=10n_2/n_1$, in domain (ii).

n_2/n_1	c_2	ω/ω_p
0	0	1
0.01	0.01	1.014
0.1	0.09	1.124
0.25	0.2	1.257

VI. TWO-COMPONENT ELECTRON-ION PLASMA

The two-component electron-ion plasma, a classical charge-neutral system consisting of point ions of charge Ze and electrons of charge $-e$, with densities $n_i=(4\pi a_i^3/3)^{-1}$, $n_e=(4\pi a_e^3/3)^{-1}$, is a realistic model for actual high-temperature plasmas. The classical approach, however, breaks down at low temperatures for at least two reasons. First, full ionization requires temperatures of the order of I_Z, the complete ionization energy of an atom with nuclear charge Z; second, in order for the electrons to be nondegenerate, the temperature should be higher than the Fermi energy of the electrons. These two conditions severely limit the attainable Γ values. Since $I_Z \sim Z^2$ and $a_e \sim Z^{-1/3}$, one finds that $\Gamma_{\text{ion}} < Z^{2/3}/\alpha^{1/2}$, where $\alpha=1/\beta I_Z > \frac{1}{6}$ for almost complete ionization.

The classical model is further limited by the incorrect description of the two-body distribution function at small particle separation. In order to remedy the situation within the classical model, pseudopotentials have been introduced[32] and were used successfully[33] to represent the quantum mechanical softening of the Coulomb interaction as $r \to 0$. The three pseudopotentials $\psi^{e\text{-}e}(k)$, $\psi^{e\text{-}i}(k)$, and $\psi^{i\text{-}i}(k)$ are, in general, different, at least in scale. Therefore, in contrast to the case of the bare Coulomb interaction, the determinant of the interaction matrix $\|\psi^{AB}(k)\|$ does not vanish. This, in turn, invalidates the simple relationship (22) between the response functions $\chi^A(\mathbf{k}\omega)$ and $\epsilon(\mathbf{k}\omega)$. Nevertheless, since the pseudopotentials differ from the Coulomb potential only for very large wave numbers (k^{-1} of the order of the Bohr radius or of the de Broglie wavelength), for $k \to 0$ the formalism we have derived remains valid, with the only stipulation that the replacement

$$\frac{1}{V}\sum_{\mathbf{q}} g^{AB}(\mathbf{q}) \to \frac{1}{V}\sum_{\mathbf{q}} \frac{\psi^{AB}(q)}{\phi^{AB}(q)} g^{AB}(\mathbf{q}) \equiv I \quad (61)$$

in the previously derived formulas in Sec. V should be observed.

The charge neutrality of the system simplifies (54) and (57) to

$$p=-\tfrac{1}{3},\quad d^2=-\tfrac{1}{3}\ . \quad (62)$$

The integral I defined in (61), although not directly related to $g(r=0)$ anymore, still picks up contributions from the close vicinity of $r=0$. This can be seen by rewriting I in r representation:

$$I=-\frac{1}{4\pi e^2 Z_1 Z_2}\int d^3r\, g^{12}(r)\nabla^2\psi^{12}(r)\ . \quad (63)$$

Noting that $\psi(r) \neq \phi(r)$ for small r values only and that $\nabla^2\phi(r)=0$ for $r\neq 0$ proves this assertion. It then also follows that $I>0$. The dispersion relation finally simplifies to[34]

$$\omega^2=\omega_p^2(1+\tfrac{1}{3}I) \ . \tag{64}$$

There is now only implicit dependence on charge and mass ratios through the correlation function and the corresponding integral I. We note the shift is always *positive*. As to the missing ω_- solution, in the charge-neutral system it becomes an *acoustic* mode, and for this reason it does not show up in the $k=0$ analysis.

Molecular-dynamics data by Hansen and co-workers[35] are also available for a hydrogen plasma. The discussed combined conditions of nondegenerate electron gas and of complete ionization, however, limit their Γ values to $\Gamma<\Gamma_{\max}\simeq 2$. Nevertheless, an upward shift of the plasma frequency can be identified. Apart from a qualitative agreement with the prediction of Eq. (64), however, application of the present theory would be appropriate to higher Z systems, with their corresponding higher $\Gamma_{\max}$ values and has to await the evaluation[34] of the corresponding integral (63). This work is in progress[34] by using the pseudopotential determined from the Slater sum with the Planck-Larkin correction.[33]

VII. CONCLUSIONS

In this paper we have developed a new approach to the analysis of collective modes and their dispersion in strongly coupled classical Coulomb systems. The method, based on the quasilocalization of the constituents of a strongly coupled Coulomb liquid, is expected to provide a good description of the dynamical properties of such systems for intermediate and high Γ values ($\Gamma>15$ for 3D and $\Gamma>5$ for 2D plasmas) in the intermediate- and high-frequency domain. The structure of the resulting $\epsilon(\mathbf{k}\omega)$'s not only satisfies the ω^{-4} frequency moment sum rules, but also explains why in many cases the high-frequency sum-rule expansion provides a good approximation even for frequency values which would not justify the use of a high-frequency expansion. The method is not restricted in k, and thus it affords the first analytic glimpse into the high-k behavior of the plasmon mode for strongly coupled systems. The response functions and the ensuing dispersion relations are functionals of the pair-correlation function: in a number of cases, these are readily available either from Monte Carlo computations or from the solution of the hypernetted-chain equations. We have analyzed the overall features of the plasmon dispersion for 3D and 2D one-component plasmas: for the former, we have used the HNC data of Rogers *et al.*[9] and for the latter the HNC data of Lado.[10] The analysis yields the first reliable calculation of the lower bound of the asymptotic value of the plasmon frequency in the 3D and 2D OCP's [$\omega(k\to\infty)=0.577\omega_p$ and $\omega(k\to\infty)=0.604\omega_0$, respectively] and the upper bound ($\omega_{\max}=0.874\omega_0$) in the 2D OCP. We have found an oscillatory behavior in the $\omega(k)$ curve for high-ka values in both systems, reflecting the existence of a well-developed short-range order.

For binary systems, partial correlation function data for the calculation of the full dispersion relation are not immediately available. Here, however, the most interesting effect is the shift of the plasma frequency (at $k=0$), which depends only on $g^{12}(r=0)=(1/V)\sum_q g^{12}(q)$ (modified into $(1/V)\sum_q [\psi^{12}(q)/\phi^{12}(q)]g^{12}(q)$ in the case of a pseudopotential $\psi(q)$ different from the Coulombic $\phi(q)$). Since $g(r=0)=-1$ for a binary ionic mixture, the plasma frequency is easily calculated, providing the first reliable evaluation (going beyond the ω^{-4} sum-rule estimate) of this quantity. For highly asymmetric mixtures, the shift turns out to be quite substantial (12% for H^+ mixed with 10% Xe^{10+}) for parameter ranges of experimental realizability. We also find a low frequency longitudinal mode which does not vanish in the $n_2/n_1\to 0$ limit. The physical significance of such "localized" modes is, however, questionable as long as damping and saturation effects are not included in the description of the system.

For the electron-ion plasma, we demonstrate the existence of an upward shift in the plasma frequency (at $k=0$). Quantitative evaluation of the shift is contingent on the calculation of the pseudopotential-weighted integral of the correlation function (appropriate for the chosen pseudopotential).

Two shortcomings of the method can be identified. One is the absence of damping or dissipative effects. This is a consequence of the harmonic approximation and of the neglect of the slow migration of the particles, considered as quasisites. The second, also stemming from the neglect of the slow thermal migration, is the apparent incorrect structure of $\epsilon(\mathbf{k}\omega)$ for high k values. This is most conspicuous in the behavior of $\epsilon(\mathbf{k}0)$, which remains negative as $k\to\infty$, but manifests itself more importantly in the absence of the thermal (Bohm-Gross–type) term in the plasmon dispersion, which should be the dominant term as $k\to\infty$.

In future papers, we further analyze the collective behavior and dispersion characteristics of strongly coupled Coulomb systems. The inclusion of the "direct" thermal effects is one of the objectives in going beyond the scope of the present work.

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APPENDIX

In this Appendix, we show that the transformation from the $(\xi^A_{i\mu},\pi^A_{i\mu})$ set to the $(\xi^A_{\mathbf{k}\mu},\pi^A_{-\mathbf{k}\mu})$ set is a canonical one. While in the case of a periodic lattice a similar transformation is always canonical within a correctly chosen set of $\mathbf{k}$ vectors, in a random medium the transformation is only approximately canonical. However, the approximate nature of the transformation is consistent with the approach used in deriving the basic Hamiltonian $\mathcal{H}$. The coordinate transformation is defined through [cf. Eq. (4)]

$$\xi^A_{i,\alpha}=\frac{1}{(N_A m_A)^{1/2}}\sum_{\mathbf{k}}\xi^A_{\mathbf{k},\alpha}e^{i\mathbf{k}\cdot\mathbf{x}^A_i},$$
$$\pi^A_{i,\alpha}=\left[\frac{m_A}{N_A}\right]^{1/2}\sum_{\mathbf{k}}\pi^A_{\mathbf{k},\alpha}e^{i\mathbf{k}\cdot\mathbf{x}^A_i}. \tag{A1}$$

Since (A1) represents a nonsingular linear transformation, the transformation matrix certainly possesses an inverse, say $\Theta_{\mathbf{k}_i}$,

$$\xi^A_{\mathbf{k},\alpha}=\frac{1}{(N_A m_A)^{1/2}}\sum_i\Theta^{A\bar{B}}_{\mathbf{k}i,\alpha\beta}\xi^{\bar{B}}_{i,\beta},$$
$$\pi^A_{\mathbf{k},\alpha}=\left[\frac{m_A}{N_A}\right]^{1/2}\sum_i\Theta^{A\bar{B}}_{\mathbf{k}i,\alpha\beta}\pi^{\bar{B}}_{i,\beta}. \tag{A2}$$

The Poisson bracket of the new coordinates now can be calculated:

$$[\xi^A_{\mathbf{p},\mu},\pi^B_{-\mathbf{q},\nu}]=\sum_i\frac{\partial\xi^A_{\mathbf{p},\mu}}{\partial\xi^{\bar{C}}_{i,\alpha}}\frac{\partial\pi^B_{-\mathbf{q},\nu}}{\partial\pi^{\bar{C}}_{i,\alpha}}$$
$$=\sum_i\frac{1}{(N_A N_B)^{1/2}}\Theta^{A\bar{C}}_{\mathbf{p}i,\mu\alpha}\Theta^{B\bar{C}}_{-\mathbf{q}i,\nu\alpha}. \tag{A3}$$

The transformation matrix Θ can be expressed as

$$\Theta^{AB}_{\mathbf{k}i,\mu\nu}=(e^{-i\mathbf{k}\cdot\mathbf{x}^A_i}+\Delta_{\mathbf{k}i})\delta^{AB}\delta_{\mu\nu},$$

where $\Delta_{\mathbf{k}i}$ represents the deviation, due to the random positions, from the exponential form which would be the correct expression for a periodic lattice. In the spirit of the approximation used to derive (8), we now replace the right-hand side of (A3) by its average

$$\left\langle\sum_i\Theta^{A\bar{C}}_{\mathbf{p}i,\mu\alpha}\Theta^{B\bar{C}}_{-\mathbf{q}i,\nu\alpha}\right\rangle$$
$$=\delta^{AB}\delta_{\mu\nu}\sum_i(\langle e^{-i(\mathbf{p}-\mathbf{q})\cdot\mathbf{x}^A_i}\rangle+\langle e^{-i\mathbf{p}\cdot\mathbf{x}^A_i}\Delta_{-\mathbf{q}i}\rangle$$
$$+\langle e^{i\mathbf{q}\cdot\mathbf{x}^A_i}\Delta_{\mathbf{p}x_i}\rangle+\langle\Delta_{\mathbf{p}i}\Delta_{-\mathbf{q}_i}\rangle). \tag{A4}$$

If we make the plausible assumption that the Δ's are uncorrelated with the parent positions or with each other, then all the averages in (A4) but the first contain the average $\langle\Delta\rangle$, which, in turn, can reasonably be assumed to vanish. Thus one is left with the first term, which, in view of (7), becomes $N_A\delta_{\mathbf{p}-\mathbf{q}}$. Returning now to (A3), the Poisson bracket becomes

$$[\xi^A_{\mathbf{p},\mu}\pi^B_{-\mathbf{q},\nu}]=\delta^{AB}\delta_{\mu\nu}\delta_{\mathbf{p}-\mathbf{q}}, \tag{A5}$$

demonstrating the canonical character of the transformation. Nevertheless, it should be kept in mind that, strictly speaking, the canonical transformation is valid only for $k<k_{\max}$, where $k_{\max}$ is determined by the number of degrees of freedom in the system and therefore is of the order a^{-1} (a is the interparticle distance). Even though in a disordered or fluidlike system, the study of wavelike collective motions for $k>k_{\max}$ is not quite meaningless (in contrast to the situation in a periodic lattice), the resulting relationships should be regarded as relying on more severe approximations than those pertaining to modes with $k<k_{\max}$.

[1]K. S. Singwi, M. P. Tosi, R. H. Land, and A. Sjolander, Phys. Rev. **176**, 589 (1968); K. S. Singwi, A. Sjolander, M. P. Tosi, and R. H. Land, Solid State Commun. **7**, 1503 (1969); Phys. Rev. B **1**, 1044 (1970); H. Totsuji and S. Ichimaru, Prog. Theor. Phys. **50**, 735 (1973); **52**, 42 (1974).

[2]K. I. Golden and G. Kalman, Phys. Rev. **19**, 2112 (1979).

[3]P. Viellefosse and J. P. Hansen, Phys. Rev. A **12**, 1106 (1975); see also Refs. 18, 23, and 28.

[4]C. C. Grimes and G. Adams, Phys. Rev. Lett. **36**, 145 (1976); **42**, 795 (1979).

[5]J.-P. Hansen, Phys. Rev. A **8**, 3096 (1973); E. L. Pollock and J.-P. Hansen, *ibid.* **8**, 3110 (1973).

[6]W. L. Slattery, G. D. Doolen, and H. E. DeWitt, Phys. Rev. A **21**, 2087 (1980); **26**, 2255 (1982).

[7]H. Totsuji, Phys. Rev. A **17**, 399 (1978).

[8]R. C. Gann, S. Chakravarty, and G. V. Chester, Phys. Rev. B **20**, 326 (1979).

[9]H. E. DeWitt (private communication); see also F. J. Rogers, D. A. Young, H. E. DeWitt, and M. Ross, Phys. Rev. A **28**, 2990 (1983).

[10]F. Lado, Phys. Rev. B **17**, 2827 (1978).

[11]P. Bakshi and G. Kalman, Phys. Rev. A **30**, 613 (1984).

[12]See, e.g., V. N. Tsytovich, *Nonlinear Effects in Plasma* (Plenum, New York, 1970).

[13]J.-P. Hansen, J. Phys. (Paris) Lett. **42**, 397 (1981).

[14]P. Carini, G. Kalman, and K. I. Golden, Phys. Lett. **78A**, 450 (1980); P. Carini and G. Kalman, *ibid.* **105A**, 229 (1984).

[15]K. I. Golden, Phys. Lett. **112A**, 3927 (1985).

[16]K. I. Golden and De-xin Lu, Phys. Rev. A **31**, 1763 (1985).

[17]K. I. Golden, F. Green, and D. Neilson, Phys. Rev. A **31**, 3529 (1985); **32**, 1669 (1985).

[18]I. R. McDonald, P. Vieillefosse, and J.-P. Hansen, Phys. Rev. Lett. **39**, 271 (1977); J.-P. Hansen, I. R. McDonald, and P. Vieillefosse, Phys. Rev. A **20**, 2590 (1979); J.-P. Hansen, F. Joly, and I. R. McDonald, Physica **132A**, 472 (1985).

[19]G. Kalman and K. I. Golden, Phys. Rev. A **29**, 844 (1984).

[20]L. R. Brewer, J. D. Prestage, J. J. Bollinger, and D. J. Wineland, in *Strongly Coupled Plasma Physics,* edited by F. H. Rogers and H. E. DeWitt (Plenum, New York, 1987), pp. 53–64; D. J. Larson, J. C. Berqquist, J. J. Bollinger, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 70 (1986); D. J. Wineland and W. M. Itano, Phys. Today **40**, 34 (1987).

[21]H. Totsuji, in *Strongly Coupled Plasma Physics* (Ref. 20), pp. 19–33.

[22]H. Iyetomi and S. Ichimaru, Phys. Rev. A **27**, 1734 (1983); S. Ichimaru, H. Iyetomi, S. Mikake, and N. Itoh, Astrophys. J. Lett. **265**, L83 (1983); S. Ichimaru and S. Tanaka, Phys. Rev. Lett. **56**, 2815 (1986).

[23]J.-P. Hansen, E. L. Pollock, and I. R. McDonald, Phys. Rev. Lett. **32**, 277 (1974); J.-P. Hansen, I. R. McDonald, and E. L. Pollock, Phys. Rev. A **11**, 1025 (1975).

[24]M. Minella and G. Kalman, in *Strongly Coupled Plasma Phys-*

ics (Ref. 20), pp. 483–489.
[25]M. Minella, G. Kalman, and K. I. Golden (unpublished).
[26]A. Bragchi, Phys. Rev. **178**, 707 (1969).
[27]P. M. Platzman and N. Tzoar, Phys. Rev. B **13**, 3197 (1976).
[28]N. Studart and O. Hipolito, Phys. Rev. A **22**, 2860 (1980).
[29]P. M. Platzman and H. Fukuyama, Phys. Rev. B **10**, 3150 (1974).
[30]L. Bonsall and A. A. Maradudin, Phys. Rev. B **15**, 1959 (1977).
[31]K. I. Golden, G. Kalman, and P. Wyns (unpublished).
[32]T. Dunn and A. A. Broylers, Phys. Rev. **157**, 156 (1967); C. Deutsch and M. M. Gombert, J. Math. Phys. **17**, 1077 (1976); H. Minoo, C. Deutsch, and J.-P. Hansen, Phys. Rev. A **14**, 840 (1976); H. Hellmann, J. Chem. Phys. **3**, 61 (1935); Acta Physicachim. USSR **1**, 913 (1935); **4**, 225 (1936); **4**, 324 (1936).
[33]F. J. Rogers, Phys. Rev. A **29**, 868 (1984).
[34]For a preliminary report, see H. Zhang and G. Kalman, Bull. Am. Phys. Soc. **33**, 1888 (1988).
[35]J.-P. Hansen and I. R. McDonald, Phys. Rev. A **23**, 2041 (1981); J.-P. Hansen and L. Sjogren, Phys. Fluids **25**, 617 (1982).