Response functions for multicomponent plasmas. I. Perturbation calculation for weak coupling

Hong Zhang and G. Kalman
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02167
(Received 11 March 1991)

We calculate the rigorous perturbation-theoretic result to first order in the plasma parameter γ ($=\kappa^3/4\pi n$, κ being the Debye wave number and n the density) for the partial density response functions and for the dielectric functions of a multicomponent plasma. The model used allows for the modification of the bare Coulomb potential due to short-range quantum effects into a general set of pseudopotentials. The derivation is based on applying the Vlasov-Dupree operator formalism in the first two equations of Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy. The result is given in terms of a compact expression which is valid for arbitrary $\mathbf{k}$ and ω values.

PACS number(s): 52.25.Dg, 51.10.+y, 52.25.Mq, 05.20.Dd

I. INTRODUCTION

The purpose of this and the following paper II is to analyze the dielectric and related response functions of a correlated classical multicomponent plasma. [The strength of the correlations can be characterized by the coupling parameter which for one-component plasma is $\Gamma=\beta Z^2e^2/a$ or $\gamma=\kappa^3/4\pi n$, where $\beta=(k_BT)^{-1}$, a is the interparticle distance, $(4\pi/3)a^3n=1$, κ is the Debye wave number and $\kappa=(4\pi Z^2e^2n\beta)^{1/2}$; the former is appropriate for strong, the latter for weak coupling; for a multicomponent plasma there is no unique definition, but a similar definition can be adopted with appropriate average $\overline{Z^2},\overline{n}$ values.] The present paper concerns itself with the weak-coupling ($\gamma<1$) situation, where the perturbation approach is legitimate. This problem has a long-standing history, going back to the 1960's. It was Perel and Eliashberg [1] who first went beyond the Vlasov random-phase approximation (RPA) $O(\gamma^0)$ calculation of the wave-number- and frequency-dependent dielectric function $\epsilon(\mathbf{k}\omega)$ for an electron-ion plasma and pointed out that for long wavelengths the $O(\gamma)$ contribution dominates, even for $\gamma\ll1$. Subsequently, following the work of Dawson and Oberman [2], Oberman, Ron, and Dawson [3] applied a systematic expansion procedure to the first two equations of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy and calculated the high-frequency conductivity in the $\mathbf{k}\to0$ limit. Based on Guernsey's [4] γ expansion of the BBGKY hierarchy, Coste [5] presented the most accurate solution of the problem, valid, albeit only formally, over the entire range of ω and $\mathbf{k}$ values. Kivelson and DuBois [6] and Tzoar and collaborators [7] approached the problem with the aid of the quantum many-body Green's-function formalism. More recent approaches based on the Green's-function method are due to Kraeft *et al.* [8] who mostly calculate self-energy corrections to one- and two-particle Green functions. The application of the equilibrium nodal expansion to equilibrium two-component plasmas is discussed by Deutsch, Furutani, and Gombert [9].

The renewed interest in the problem of the calculation of the correlational dielectric function stems from a number of sources. Foremost among them is the progress made in the understanding of the properties of strongly coupled ($\gamma>1$) Coulomb systems [10]. Such an understanding is not complete as long as it does not provide a link to the weakly coupled domain and a comprehensive description of the effect of the increasing correlations. From a formal point of view the rigorous perturbation calculational results contribute a paradigm against which the limits of the various strongly coupled approximations can be tested.

As to the physical systems studied, early works focused exclusively on the electron-ion plasma; since then, the importance of the binary ionic mixture (BIM)—two ion species in a neutralizing background— and its distinct characteristics have been identified. There is also increasing interest in three-component electron-ion plasmas and in the semiclassical description of three-component electron-hole systems. Another aspect of the weak-coupling calculation is the analysis of the correlation effects on collective modes. While the early works concentrated on the high-frequency conductivity and on the collisional damping of the plasmon mode, more recent interest focuses on the correlational shift of the plasmon frequency [11] and on the ion-acoustic [12] and other possible low-frequency modes.

In this paper we develop a systematic perturbation expansion and a comprehensive expression for the partial density response and for the dielectric-response function of a multicomponent plasma [13]. The perturbation expansion is carried out in γ, the plasma parameter of coupling, in the first and the second equations of the BBGKY hierarchy. This expansion technique implies that the formal expansion parameter is the "uncompensated" e^2; i.e. "compensated" terms of $O((e^2n)^m)$ (n is the density) are retained to arbitrary order but others only to $O(e^2)$.

There are three distinctive elements in our approach. The first is the adoption of a set of generalized pseudopotentials [14–20], rather than the bare Coulomb potential, to represent the interaction between the particles. Pseudopotentials, in general, are introduced either to phenomenologically describe short-range quantum-mechanical

effects within the classical formalism [14–16], or because the interacting "particles" have a structure that causes the short-range potential to deviate from its Coulomb form [16–20]. Thus, this method allows one to treat a great variety of physical systems with the correctly adapted pseudopotentials [21]. For example, for an ionized alkali-atom plasma, the interactions between the electrons and the ions, or between the ions are affected by the inner-shell electrons, and are different from the bare Coulomb interaction. The same situation prevails in liquid metals, where pseudopotentials are routinely used [22]. Similarly, in electrolyte plasmas and in molten salts [23], where the charged particles are large composite ions, pseudopotentials have been used successfully. The interaction between the electrons at short range is modified by diffraction, exchange, and spin-spin-interaction effects, while the interaction between the electrons and ions is modified by exchange repulsion, diffraction, and formation of bound and virtual bound states. The ion-ion interaction is affected by exchange-enhanced ion-ion repulsion and the modification of the Coulomb interaction due to ion penetration and screening. Properly chosen pseudopotentials can reasonably well characterize such features within the framework of a purely classical description. There is a large body of literature on the choice and construction of pseudopotentials. Simple analytic models with adjustable parameters were suggested by Hellmann [14], Dunn and Broyles [15], and Deutsch and Gombert [16]. More elaborate pseudopotentials for electron-ion interaction based on the Slater sum over one-particle wave functions were constructed by Barker [17], Storrer [18], Rogers [19], and Kraeft *et al.* [20]. However, the precise form of the pseudopotentials is not of concern to us in the present paper. Rather, it is important to point out one of their common features. It follows from the nature of the physical effects that give rise to the pseudopotential that the pseudopotentials between particle pairs belonging to different species are quite different from each other. For example, in a hydrogenic plasma, the characteristic distance within which the electron-electron pseudopotential deviates from the Coulomb potential is given by the de Broglie wavelength; for the electron-ion pseudopotential the characteristic distance is determined by both the de Broglie wavelength and by the Bohr radius, while the ion-ion pseudopotential is practically identical to the Coulomb potential. In general, in a multispecies plasma, ψ_{AA} and ψ_{BB}, the pseudopotentials within species A and B, respectively, do not determine the ψ_{AB}, the pseudopotential between particles A and B. This is, of course, in contrast to the case of the Coulomb potential (or of any other potential derivable from a Hamiltonian) where $\psi_{AB}=\sqrt{\psi_{AA}\psi_{BB}}$, i.e., where the potential is factorizable, while the pseudopotential, in general, is not. The more precise mathematical statement corresponding to this fact is that $\det\psi\neq 0$. This formal difference between pseudopotentials and the Coulomb potential has to be carefully observed in the development of the multispecies formalism. While the justification of the use of pseudopotentials for the analysis of static properties is relatively straightforward, it is more difficult to be satisfied with their role in the description of dynamical processes. Nevertheless, there is a fair amount of evidence, both from computer simulations [21] and from the experimental study of liquid metals [22] and of molten salts [23], that the approach provides a reasonable description of both static and dynamical properties, provided either the quantum-mechanical expansion parameter $r_s=a/a_B$ (a_B is the Bohr radius) for the electrons satisfies the $r_s \ll 1$ condition, or the electronic Fermi energy ε_F is small compared to the thermal energy, $\varepsilon_{F/kT} \ll 1$. If this is not the case, exchange and related effects become sufficiently dominant to invalidate the naive pseudopotential approach.

The second calculational feature is the use of the Vlasov-Dupree (VD) operator technique [24,25]. The VD operator is the time-evolution operator associated with the Vlasov equation. However, its applicability is much more general than that suggested by its origin. This is due to the structural feature of the BBGKY equations, which exhibit the simple two-, three-, etc. particle generalizations of the Vlasov operator. Thus, it is natural to represent the solutions (in particular the solution of the second BBGKY equation) in terms of the VD operator. This allows a great calculational simplification.

The third line of development in the present paper consists of the adoption of the partial response function formalism as developed by Kalman and Golden [26]. The concept of the partial response function was originally suggested by Vashishta, Bhattacharya, and Singwi [27] in order to make it possible to construct response functions in equal number to the structure functions in a multicomponent system. The formal development of the idea is, however, due to Kalman and Golden [26], who introduced matrix algebra in species space. As a result, the analysis of the multispecies problem can be brought into a one-to-one correspondence with that of a simple one-component plasma (OCP), and we can display the calculational results in a transparent and relatively compact form. This is not a negligible advantage in view of the inherent complexity of the perturbation calculation and the traditional proliferation of algebraic errors in such calculations. We also find results for the density response functions $\chi_{AB}(\mathbf{k}\omega)$ that are directly related via the fluctuation-dissipation theorem to the dynamical structure function $S_{AB}(\mathbf{k}\omega)$ of species A and B. An additional benefit of the formalism is that, even though our main interest lies in binary systems, the results also hold for systems of an arbitrary number of components.

Our results are given for arbitrary $\mathbf{k}$ and ω values. For a bare Coulomb potential in the long-wavelength $\mathbf{k}=0$ limit they are identical, although given in a much more concise form, with the earlier results of Oberman, Ron, and Dawson [3]; in the static $\omega=0$ limit, they agree with those of Kalman [28]. The question of the uniformity of the γ expansion should be mentioned. There is a good indication that in the $\omega\to 0$ and the $\mathbf{k}\to 0$ domain the expansion breaks down. However, this does not seem to affect the correctly obtained limiting cases described above, and not even the calculation of the dc conductivity [13]. The problem deserves more detailed analysis, but such a study is not the subject of the present paper.

The results of this paper are formal: no explicit calculations are performed and no numerical results are presented. The computation of the dielectric function based on the formal expressions given in this paper and of the related damping and dispersion will be the subject of a separate publication [29] (see also [13]). The implementation of the numerical program is feasible on the basis of the formulas derived in the present work without any further approximation, although the actual computations are restricted to the small-k domain.

Sections II and III of this paper are devoted to the exposition of the calculational techniques. In Sec. IV, we study the second equation of the BBGKY in the presence of an external perturbation and obtain the perturbed pair-correlation function. In Sec. V, the density response functions and the dielectric-response function are calculated, and the static and long-wavelength limits are also evaluated and contact is made with earlier results.

II. GENERAL FORMALISM

The theory of the partial response functions [26] is a central tool in the formalism of this paper. This section summarizes it and the related definitions.

The density response of species A is related to the external or to the total fields (the words "field" or "potential" are used as a shorthand expression for the potential energy of the particle in the given potential or field) through the partial density response functions as

$$\begin{aligned} n_A &= \hat{\chi}_{AB}\hat{\Phi}_B + \hat{\chi}_{ABC}\hat{\Phi}_B\hat{\Phi}_C + \cdots \\ &= \chi_{AB}\Phi_B + \chi_{ABC}\Phi_B\Phi_C + \cdots , \end{aligned} \tag{1}$$

where the subscripts denote the different species, $\hat{\chi}_{AB}, \chi_{AB}$ are the external and the total partial linear density response functions, respectively, and $\hat{\chi}_{ABC}, \chi_{ABC}$ are their quadratic counterparts; $\hat{\Phi}_B$ represents the external "partial potentials," an artificial construct conceived to act on the specified species only; Φ_B stands for the total partial potential which includes the induced average plasma potential. The notation is highly symbolic: depending on the representation used, integration over space-time variables or integration and summation over frequencies and wave vectors are implied. Unless specified otherwise, summation over the repeated species indices is understood here and throughout this paper, if the index does not appear again on the other side of the equation.

The total field Φ_B is

$$\Phi_B = \hat{\Phi}_B + \Phi_B^{\text{ind}} , \tag{2}$$

where the induced field due to the density fluctuation is related to the density of species C by

$$\Phi_B^{\text{ind}} = \psi_{BC} n_C . \tag{3}$$

ψ_{BC} is the general pseudopotential describing the effective interaction between species B and C, whose properties have been described in the Introduction. Substituting (2) and (3) into (1), and changing to matrix representation in species space, we have

$$\boldsymbol{\epsilon}\boldsymbol{\Phi} = \hat{\boldsymbol{\Phi}} , \tag{4}$$

with

$$\boldsymbol{\epsilon} = 1 - \boldsymbol{\psi}\boldsymbol{\chi} , \tag{5}$$

which is the dielectric matrix in species space. One can also introduce the inverse dielectric matrix $\boldsymbol{\eta}$

$$\boldsymbol{\Phi} = \boldsymbol{\eta}\hat{\boldsymbol{\Phi}} . \tag{6}$$

Then,

$$\boldsymbol{\eta} = \boldsymbol{\epsilon}^{-1} = 1 + \boldsymbol{\psi}\hat{\boldsymbol{\chi}} . \tag{7}$$

The connection between $\hat{\chi}_{AB}$ and $\hat{\chi}_{ABC}$ on the one hand and χ_{AB} and χ_{ABC} on the other is provided by

$$\hat{\chi}_{AB}(\mathbf{k}\omega) = \chi_{AC}(\mathbf{k}\omega)\eta_{CB}(\mathbf{k}\omega) , \tag{8}$$

$$\hat{\chi}_{ABC}(\mathbf{p}\mu;\mathbf{q}\nu) = \eta_{DA}(\mathbf{k}\omega)\chi_{DEF}(\mathbf{p}\mu;\mathbf{q}\nu)\eta_{EB}(\mathbf{p}\mu)\eta_{FC}(\mathbf{q}\nu) ,$$
$$\mathbf{k} = \mathbf{p} + \mathbf{q}, \quad \omega = \mu + \nu . \tag{9}$$

Finally, the conventional dielectric function, a scalar in species space, is given by

$$\begin{aligned} \epsilon(\mathbf{k}\omega) &= \|\boldsymbol{\epsilon}(\mathbf{k}\omega)\| \\ &= 1 - \mathrm{tr}[\boldsymbol{\psi}(k)\boldsymbol{\chi}(\mathbf{k}\omega)] + \|\boldsymbol{\psi}(k)\|\,\|\boldsymbol{\chi}(\mathbf{k}\omega)\| , \end{aligned} \tag{10}$$

the second part of the equation being valid for binary systems only; $\|\ \|$ stands for the determinant and tr for the trace. In the case of a (factorizable) Coulomb potential $\psi_{AB}(k) = Z_A Z_B \varphi(k)$, $\varphi(k) = 4\pi e^2/k^2$, and (10) reduces to the traditional expression for $\epsilon(\mathbf{k}\omega)$:

$$\epsilon(\mathbf{k}\omega) = 1 - \varphi(k)\sum_A Z_A^2 \chi_A(\mathbf{k}\omega) .$$

It is the $\epsilon(\mathbf{k}\omega) = 0$ dispersion relation that determines the longitudinal collective modes of the system.

When a perturbation expansion in the coupling γ is contemplated, both n and Φ (but not $\hat{\Phi}$) and $\chi_{AB}, \chi_{ABC}, \hat{\chi}_{AB}, \hat{\chi}_{ABC}$ can be expressed as sums over terms of order γ^n: $\chi_{AB} = \sum_{n=0}^{\infty} \chi_{AB}^{(n)}$, etc. The lowest-order term is the RPA (Vlasov) contribution: to this order both χ_{AB} and χ_{ABC} (but not $\hat{\chi}_{AB}$ and $\hat{\chi}_{ABC}$) are diagonal:

$$\chi_{AB}^{(0)}(\mathbf{k}\omega) \equiv \delta_{AB}\chi_A^{(0)}(\mathbf{k}\omega) , \tag{11}$$

$$\chi_{ABC}^{(0)}(\mathbf{p}\mu;\mathbf{q}\nu) \equiv \delta_{AB}\delta_{AC}\chi_A^{(0)}(\mathbf{p}\mu;\mathbf{q}\nu) . \tag{12}$$

To next order

$$\mathbf{n}^{(1)} = \boldsymbol{\chi}^{(1)}\boldsymbol{\Phi}^{(0)} + \boldsymbol{\chi}^{(0)}\boldsymbol{\Phi}^{(1)} = \boldsymbol{\eta}^T\boldsymbol{\chi}^{(1)}\boldsymbol{\Phi}^{(0)} = \hat{\boldsymbol{\chi}}^{(1)}\hat{\boldsymbol{\Phi}} , \tag{13}$$

which then implies

$$\hat{\boldsymbol{\chi}}^{(1)} = \boldsymbol{\eta}^T\boldsymbol{\chi}^{(1)}\boldsymbol{\eta} . \tag{14}$$

Here and in the sequel $\boldsymbol{\eta}$ and $\boldsymbol{\epsilon}$ stand for their Vlasov values: $\boldsymbol{\eta} \equiv \boldsymbol{\eta}^{(0)}$, $\boldsymbol{\epsilon} \equiv \boldsymbol{\epsilon}^{(0)}$.

III. THE VLASOV-DUPREE-OPERATOR TECHNIQUE

The Vlasov-Dupree-operator technique was originally introduced for the OCP [24,25], but its generalization to the multicomponent situation is straightforward. The Vlasov-Dupree operator is defined through the Vlasov

equation as its time-evolution operator: its importance lies in the fact that its two-particle, etc. generalizations reappear in the second- and higher-order equations of the BBGKY hierarchy.

The linearized Vlasov equation for a multicomponent plasma is

$$-i[\omega\delta_{AB}-V_{AB}(\mathbf{k};\mathbf{v}\mathbf{v}')]F_B^{(10)}(\mathbf{v}';\mathbf{k}\omega) = i\frac{1}{m_A}\mathbf{k}\cdot\frac{\partial F_A^{(00)}(v)}{\partial\mathbf{v}}\hat{\Phi}_A(\mathbf{k}\omega) , \quad (15)$$

where here and in the sequel the $F_A^{(mn)}$ are the one-particle distributions function in the mth order in the external field and in the nth order in the coupling. ψ_{AB} is the general pseudopotential; $\hat{\Phi}_A$ is the external partial field acting on the species A only. The velocity arguments $\mathbf{v}$ and $\mathbf{v}'$ are treated as matrix indices and summation (integration) is implied over dummy velocity arguments. The Vlasov operator is defined through Eq. (15); for any "well-behaved" $f(\mathbf{v})$,

$$V_{AB}(\mathbf{k};\mathbf{v}\mathbf{v}')f_B(\mathbf{v}')=\int d^3v'\left[\mathbf{k}\cdot\mathbf{v}'\delta(\mathbf{v}-\mathbf{v}')\delta_{AB}-\frac{1}{m_A}\psi_{AB}(k)\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}}F_A^{(00)}(v)\right]\times f_B(\mathbf{v}') . \quad (16)$$

Equation (15) is solved formally to yield

$$F_A^{(10)}(\mathbf{v};\mathbf{k}\omega)=iW_{AB}(\mathbf{k}\omega;\mathbf{v}\mathbf{v}')\frac{1}{m_B}\hat{\Phi}_B(\mathbf{k}\omega)\mathbf{k}\cdot\frac{\partial F_B^{(00)}(v')}{\partial\mathbf{v}'} , \quad (17)$$

where W now is the Vlasov-Dupree operator, the Fourier transform of the time-evolution operator (resolvent operator) associated with V:

$$W(\mathbf{k}\omega;\mathbf{v}\mathbf{v}')=i[\omega-V(\mathbf{k};\mathbf{v}\mathbf{v}')]^{-1} . \quad (18)$$

The solution of Eq. (15) is well known:

$$F_A^{(10)}(\mathbf{v};\mathbf{k}\omega)=-\frac{1}{m_A}\frac{1}{\omega-\mathbf{k}\cdot\mathbf{v}}\mathbf{k}\cdot\frac{\partial}{\partial v}F_A^{(00)}(v)\times\eta_{AB}(\mathbf{k}\omega)\hat{\Phi}_B(\mathbf{k}\omega) , \quad (19)$$

where η is constructed with the aid of the Vlasov (RPA) density response

$$\chi_A^{(0)}(\mathbf{k}\omega)=-\frac{1}{m_A}\int d^3v\frac{\mathbf{k}\cdot\frac{\partial F_A(v)}{\partial\mathbf{v}}}{\omega-\mathbf{k}\cdot\mathbf{v}} .$$

Comparison of (17) and (19) immediately yields the W operator:

$$W_{AB}(\mathbf{k}\omega;\mathbf{v}\mathbf{v}')F_B(\mathbf{v}')=\frac{i}{\omega-\mathbf{k}\cdot\mathbf{v}+io}\int\left[\delta(\mathbf{v}-\mathbf{v}')\delta_{AB}-\frac{1}{m_A}\frac{\psi_{AC}(k)}{\omega-\mathbf{k}\cdot\mathbf{v}'+io}\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}}F_A^{(00)}(v)\eta_{BC}(\mathbf{k}\omega)\right]F_B(\mathbf{v}')d^3v' . \quad (20)$$

The correct treatment of the singular denominators is by adding a small, positive imaginary part ($+io$) as indicated, which then ensures that $W_{AB}(\omega)$ possesses proper causal behavior. This interpretation of the frequency denominators will be implicitly understood throughout the paper, without the io term being explicitly displayed.

IV. THE PERTURBED PAIR-CORRELATION FUNCTION

The second equation of BBGKY hierarchy to the first order in coupling is [30]

$$\left[\frac{\partial}{\partial t}+\mathbf{v}_1\cdot\frac{\partial}{\partial\mathbf{x}_1}-\frac{1}{m_A}\frac{\partial}{\partial\mathbf{x}_1}\hat{\Phi}_A(\mathbf{x}_1,t)\cdot\frac{\partial}{\partial\mathbf{v}_1}+\mathbf{v}_2\cdot\frac{\partial}{\partial\mathbf{x}_2}-\frac{1}{m_B}\frac{\partial}{\partial\mathbf{x}_2}\hat{\Phi}_B(\mathbf{x}_2,t)\cdot\frac{\partial}{\partial\mathbf{v}_2}\right.$$
$$-\left[\int\frac{1}{m_A}\frac{\partial}{\partial\mathbf{x}_1}\psi_{AC}(\mathbf{x}_1-\mathbf{x}_1')\cdot\frac{\partial}{\partial\mathbf{v}_1}F_C(\mathbf{x}_1',\mathbf{v}_1';t)d^3x_1'd^3v_1'\right.$$
$$\left.\left.+\int\frac{1}{m_B}\frac{\partial}{\partial\mathbf{x}_2}\psi_{BD}(\mathbf{x}_2-\mathbf{x}_2')\cdot\frac{\partial}{\partial\mathbf{v}_2}F_D(\mathbf{x}_2',\mathbf{v}_2';t)d^3x_2'd^3v_2'\right]\right]G_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t)$$
$$-\int\frac{1}{m_A}\frac{\partial}{\partial\mathbf{x}_1}\psi_{AC}(\mathbf{x}_1-\mathbf{x}_1')\cdot\frac{\partial}{\partial\mathbf{v}_1}F_A(\mathbf{x}_1,\mathbf{v}_1;t)G_{BC}(\mathbf{x}_2,\mathbf{v}_2;\mathbf{x}_1',\mathbf{v}_1';t)d^3x_1'd^3v_1'$$
$$-\int\frac{1}{m_B}\frac{\partial}{\partial\mathbf{x}_2}\psi_{BD}(\mathbf{x}_2-\mathbf{x}_2')\cdot\frac{\partial}{\partial\mathbf{v}_2}F_B(\mathbf{x}_2,\mathbf{v}_2;t)G_{DA}(\mathbf{x}_2',\mathbf{v}_2';\mathbf{x}_1,\mathbf{v}_1;t)d^3x_2'd^3v_2'$$
$$=\left[\frac{1}{m_A}\frac{\partial}{\partial\mathbf{x}_1}\psi_{AB}(\mathbf{x}_1-\mathbf{x}_2)\cdot\frac{\partial}{\partial\mathbf{v}_1}+\frac{1}{m_B}\frac{\partial}{\partial\mathbf{x}_2}\psi_{AB}(\mathbf{x}_1-\mathbf{x}_2)\cdot\frac{\partial}{\partial\mathbf{v}_2}\right]F_A(\mathbf{x}_1,\mathbf{v}_1;t)F_B(\mathbf{x}_2,\mathbf{v}_2;t) . \quad (21)$$

The two-body distribution function $F_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t)$ has been expressed as

$$F_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t)=F_A(\mathbf{x}_1,\mathbf{v}_1;t)F_B(\mathbf{x}_2,\mathbf{v}_2;t) +G_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t) , \quad (22)$$

where $G_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t)$ is the nonequilibrium two-body correlation function, and one has the normalization conditions

$$\sum_A \int F_A(\mathbf{x},\mathbf{v};t)d^3v=\sum_A n_A=n . \quad (23)$$

Now, we further take the expansion up to the first order both in the external perturbation $\hat{\Phi}$ and in the coupling constant γ:

$$F_A(\mathbf{x},\mathbf{v};t)= F_A^{(00)}(v)+F_A^{(10)}(\mathbf{x},\mathbf{v};t)+F_A^{(11)}(\mathbf{x},\mathbf{v};t) , \quad (24)$$

$$G_{AB}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t)= F_A^{(00)}(v_1)F_B^{(00)}(v_2)g_{AB}^{(01)}(\mathbf{x}_1-\mathbf{x}_2) +G_{AB}^{(11)}(\mathbf{x}_1,\mathbf{v}_1;\mathbf{x}_2,\mathbf{v}_2;t) , \quad (25)$$

where we have set $F^{(01)}=0$, for correlations do not affect the equilibrium distribution. Substituting (24) and (25) into Eq. (21), and keeping the first-order terms both in the external field and in the coupling, and taking the Fourier transform, one finally obtains [13]

$$-i[\omega\delta_{AC}\delta_{BD}-\delta_{AC}V_{BD}(\mathbf{q};\mathbf{v}_2\mathbf{v}_2') -\delta_{BD}V_{AC}(\mathbf{p};\mathbf{v}_1\mathbf{v}_1')]G_{CD}^{(11)}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1'\mathbf{v}_2') =L_{AB}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1\mathbf{v}_2) , \quad (26)$$

where

$$\begin{aligned}L_{AB}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1\mathbf{v}_2)=& \, i\frac{1}{m_A}\hat{\Phi}_A(\mathbf{k}\omega)\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}_1}F_A(v_1)F_B(v_2)g_{AB}(q)\\ &+i\frac{1}{m_A}\int d^3v_1'\psi_{AC}(k)F_C^{(10)}(\mathbf{v}_1',\mathbf{k}\omega)g_{AB}(q)\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}_1}F_A(v_1)F_B(v_2)\\ &-i\frac{1}{m_A}\int d^3v_1'\psi_{AC}(q)\mathbf{q}\cdot\frac{\partial}{\partial\mathbf{v}_1}F_A^{(10)}(\mathbf{v}_1;\mathbf{k}\omega)g_{BC}(q)F_B(v_2)F_C(v_1')\\ &-i\frac{1}{m_A}\psi_{AB}(q)\mathbf{q}\cdot\frac{\partial}{\partial\mathbf{v}_1}F_A^{(10)}(\mathbf{v}_1;\mathbf{k}\omega)F_B(\mathbf{v}_2)\\ &+i\frac{1}{m_B}\psi_{AB}(q)\mathbf{q}\cdot\frac{\partial}{\partial\mathbf{v}_2}F_B(v_2)F_A^{(10)}(\mathbf{v}_1;\mathbf{k}\omega)+[1\leftrightarrow2, A\leftrightarrow B,\mathbf{p}\leftrightarrow\mathbf{q}], \quad \mathbf{k}=\mathbf{p}+\mathbf{q} . \end{aligned} \quad (27)$$

Note that the γ expansion implies that while e^2 is of the order of the coupling constant, the product e^2n is of zeroth order in the coupling, that is, e^2 is formally compensated by the density. $[1\leftrightarrow2, A\leftrightarrow B,\mathbf{p}\leftrightarrow\mathbf{q}]$ signifies the preceding terms subject to the designated exchange operation. L_{AB} is a known function and can be simplified to [13]

$$\begin{aligned}L_{AB}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1\mathbf{v}_2)=-i&\left[\frac{\beta^2\psi_{AB}(q)\mathbf{q}\cdot\mathbf{v}_2\mathbf{k}\cdot\mathbf{v}_1}{\omega-\mathbf{k}\cdot\mathbf{v}_1}+\frac{1}{m_A}g_{AB}(q)\left[\beta m_A\mathbf{k}\cdot\mathbf{v}_1+\frac{m_A\beta\mathbf{k}\cdot\mathbf{v}_1\mathbf{q}\cdot\mathbf{v}_1}{\omega-\mathbf{k}\cdot\mathbf{v}_1}-\frac{\omega\mathbf{k}\cdot\mathbf{q}}{(\omega-\mathbf{k}\cdot\mathbf{v}_1)^2}\right]\right]\\ &\times F_A(v_1)F_B(v_2)\Phi_A(\mathbf{k}\omega)+[1\leftrightarrow2, A\leftrightarrow B,\mathbf{p}\leftrightarrow\mathbf{q}] . \end{aligned} \quad (28)$$

We have dropped the superscripts from $g^{(01)}$ and $F^{(00)}$: $g(k)$ and $F(v)$ signify the equilibrium Debye pair-correlation function and the equilibrium Maxwell velocity distribution, respectively. Equation (26) is easily solved by applying the Vlasov-Dupree-operator method to yield

$$G_{AB}^{(11)}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1\mathbf{v}_2)=\frac{1}{2\pi}\int d\nu\, W_{AC}(\mathbf{p}\nu;\mathbf{v}_1\mathbf{v}_1') \times W_{BD}(\mathbf{q}\mu;\mathbf{v}_2\mathbf{v}_2') \times L_{CD}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1'\mathbf{v}_2') , \quad \omega=\mu+\nu . \quad (29)$$

This is an exact formal solution for the perturbed two-body correlation function. By substituting it into the first equation of the BBGKY hierarchy, one is able to obtain the density response to the first order in the coupling.

V. THE DENSITY RESPONSE FUNCTIONS AND THE DIELECTRIC FUNCTION

The Fourier transform of the first equation of BBGKY [30] in the first order in both of the external field and of the coupling is

$$-i[\omega\delta_{AC}-V_{AC}(\mathbf{k};\mathbf{v}_1\mathbf{v}_1')]F_A^{(11)}(\mathbf{v}_1';\mathbf{k}\omega)$$
$$=\frac{i}{m_A}\frac{1}{V}\sum_{\mathbf{p}}\psi_{AC}(p)\mathbf{p}\cdot\frac{\partial}{\partial\mathbf{v}_1}\int d^3v_2 G_{AC}^{(11)}(\mathbf{p},\mathbf{q},\omega;\mathbf{v}_1\mathbf{v}_2)\ . \tag{30}$$

Equation (30) is solved by using the Vlasov-Dupree operator again to yield

$$F_A^{(11)}(\mathbf{k}\omega;\mathbf{v}_1)=i\,W_{AB}(\mathbf{k}\omega;\mathbf{v}_1\mathbf{v}_1')\frac{1}{m_B}\frac{1}{V}\times\sum_{\mathbf{p}}\psi_{BC}(p)\mathbf{p}\cdot\frac{\partial}{\partial\mathbf{v}_1}\times\int d^3v_2 G_{BC}^{(11)}(\mathbf{q},\mathbf{p},\omega;\mathbf{v}_1'\mathbf{v}_2)\ . \tag{31}$$

Substituting Eq. (29) into Eq. (31), one finds

$$F_A^{(11)}(\mathbf{k}\omega;\mathbf{v}_1)=i\,W_{AB}(\mathbf{k}\omega;\mathbf{v}_1\mathbf{v}_1')\frac{1}{m_B}\frac{1}{V}\sum_{\mathbf{p}}\psi_{BC}(p)\mathbf{p}\cdot\frac{\partial}{\partial\mathbf{v}_1}\left[\frac{1}{2\pi}\int d\nu\int d^3v_2 W_{BE}(\mathbf{q}\nu;\mathbf{v}_1'\mathbf{v}_1'')W_{CF}(\mathbf{p}\mu;\mathbf{v}_2'\mathbf{v}_2'')L_{EF}(\mathbf{q},\mathbf{p},\omega;\mathbf{v}_1''\mathbf{v}_2'')\right], \tag{32}$$

and the density response is given by

$$n_A^{(11)}(\mathbf{k}\omega)=\int d^3v_1 F_A^{(11)}(\mathbf{k}\omega;\mathbf{v}_1)\ . \tag{33}$$

By using the explicit representation of W as given in (20), one can see that the integrals over $\mathbf{v}_1$, $\mathbf{v}_2$, and $\mathbf{v}_1'$ in Eq. (33) can be carried out by defining the dimensionless function

$$T_B(\mathbf{k}\omega;\mathbf{q}\nu)\equiv\frac{1}{n_B(\beta m_B)^2}\int d^3v\frac{\mathbf{k}\cdot\mathbf{p}\,\mathbf{q}\cdot\frac{\partial}{\partial\mathbf{v}}F_B(v)}{(\omega-\mathbf{k}\cdot\mathbf{v})^2(\nu-\mathbf{q}\cdot\mathbf{v})}\ , \tag{34}$$

which is related to the RPA quadratic density response function:

$$\chi_B^{(0)}(\mathbf{q}\nu;\mathbf{p}\mu)=-\frac{\beta^2 n_B}{2}[T_B(\mathbf{k}\omega;\mathbf{q}\nu)+T_B(\mathbf{k}\omega;\mathbf{p}\mu)]\ . \tag{35}$$

One can finally obtain

$$n_A^{(11)}(\mathbf{k}\omega)=-\frac{1}{m_B V}\sum_{\mathbf{p}}\frac{1}{2\pi}\int d\nu\int d^3v_3\int d^3v_4\eta_{BA}(\mathbf{k}\omega)\psi_{BC}(p)\times\left[\frac{\mathbf{k}\cdot\mathbf{p}}{(\omega-\mathbf{k}\cdot\mathbf{v}_3)^2(\nu-\mathbf{q}\cdot\mathbf{v}_3)}\delta_{BE}-\beta^2 n_B m_B\frac{T_B(\mathbf{k}\omega;\mathbf{q}\nu)}{\nu-\mathbf{q}\cdot\mathbf{v}_3}\psi_{BG}(q)\eta_{EG}(\mathbf{q}\nu)\right]\times\frac{1}{\mu-\mathbf{p}\cdot\mathbf{v}_4}\eta_{FC}(\mathbf{p}\mu)L_{EF}(\mathbf{q},\mathbf{p},\omega;\mathbf{v}_3\mathbf{v}_4)\ , \tag{36}$$

where L_{EF} is given by (28).

By comparing Eqs. (36) and (13), one obtains the first-order correlational part of the partial density response function,

$$\chi_{BD}^{(1)}(\mathbf{k}\omega)=-\frac{\beta^2 n_B}{V}\sum_{\mathbf{p}}\frac{1}{2\pi}\int d\nu\int d^3v_3\int d^3v_4\psi_{BC}(p)\times\left[\frac{\mathbf{k}\cdot\mathbf{p}}{(\omega-\mathbf{k}\cdot\mathbf{v}_3)^2(\nu-\mathbf{q}\cdot\mathbf{v}_3)\beta^2 m_B n_B}\delta_{BD}-\frac{T_B(\mathbf{k}\omega;\mathbf{q}\nu)}{\nu-\mathbf{q}\cdot\mathbf{v}_3}\psi_{BF}(q)\eta_{DF}(\mathbf{q}\nu)\right]\times\frac{1}{\mu-\mathbf{p}\cdot\mathbf{v}_4}\eta_{EC}(\mathbf{p}\mu)F_D(v_3)F_E(v_4)[H_{DE}(\mathbf{k},\mathbf{p},\omega;\mathbf{v}_3\mathbf{v}_4)+H_{ED}(\mathbf{k},\mathbf{q},\omega;\mathbf{v}_4\mathbf{v}_3)]\ , \tag{37}$$

where

$$H_{DE}(\mathbf{k},\mathbf{p},\omega;\mathbf{v}_3\mathbf{v}_4)=-\frac{i}{m_D}g_{DE}(p)\left[\beta m_D\mathbf{k}\cdot\mathbf{v}_3+\frac{1}{\omega-\mathbf{k}\cdot\mathbf{v}_3}\left[m_D\beta\mathbf{k}\cdot\mathbf{v}_3\mathbf{p}\cdot\mathbf{v}_3-\frac{\omega\mathbf{k}\cdot\mathbf{p}}{\omega-\mathbf{k}\cdot\mathbf{v}_3}\right]\right]-i\beta^2\psi_{DE}(p)\frac{\mathbf{k}\cdot\mathbf{v}_3\,\mathbf{p}\cdot\mathbf{v}_4}{\omega-\mathbf{k}\cdot\mathbf{v}_3}\ . \tag{38}$$

The velocity integrals in (37) can be carried out in terms of the Vlasov response function $\chi_A^{(0)}$, and the $T_A(\mathbf{k}\omega;\mathbf{p}\nu)$ function defined in (34) and its ω frequency derivatives [13].

After some cumbersome but straightforward algebraic calculations, the partial response function can be finally expressed as

$$\chi^{(1)}_{AD}(\mathbf{k}\omega)=-\frac{4}{\beta V}\sum_{\mathbf{p}}\int d\nu\,\delta_-(\nu)\Bigg[\psi_{AC}(q)\eta_{DC}(\mathbf{q}\nu)\psi_{AE}(p)\eta_{DE}(\mathbf{p}\mu)\chi^{(0)}_A(\mathbf{p}\mu;\mathbf{q}\nu)\chi^{(0)}_D(\mathbf{p}\mu;\mathbf{q}\nu) - \left[\frac{\delta_{AD}}{24}\frac{\beta^2 n_A}{m_A}\mathbf{k}\cdot\mathbf{p}\psi_{AC}(p)\frac{\partial^2}{\partial\omega^2}\right]_{\nu,\mu} \times[\eta_{AC}(\mathbf{p}\mu)T_A(\mathbf{k}\omega;\mathbf{q}\nu)+\eta_{AC}(\mathbf{p}\nu)T_A(\mathbf{k}\omega;\mathbf{q}\mu)-2\eta_{AC}(\mathbf{p}0)T_A(\mathbf{k}\omega;\mathbf{q}0)]\Bigg], \tag{39}$$

where $\delta_-(\nu)$ has its conventional definition

$$\delta_-(\nu)=\lim_{o\to 0}\frac{-i}{2\pi}\frac{1}{\nu-io}.$$

The dielectric function ϵ can be calculated through (10).

The result (39) is a relatively simple and compact expression for arbitrary $\mathbf{k}$ and ω values; it possesses the requisite symmetry $\chi^{(1)}_{AB}=\chi^{(1)}_{BA}$, which can be seen by observing the identity

$$\sum_C \psi_{AC}\eta_{DC}=\sum_C \psi_{DC}\eta_{AC}. \tag{40}$$

It also satisfies the ω^{-4} high-frequency sum rule. As we show in the following, in the $\mathbf{k}=0$ and $\omega=0$ limits the general expression (39) for the response function can be reduced to much simpler forms.

In case of the bare Coulomb potential, (39) becomes

$$\chi^{(1)}_{AD}(\mathbf{k}\omega)=-\frac{4}{\beta V}\sum_{\mathbf{p}}\int d\nu\,\delta_-(\nu)\Bigg[Z_A^2Z_D^2\frac{\varphi(q)\varphi(p)}{\epsilon(\mathbf{q}\nu)\epsilon(\mathbf{p}\mu)}\chi^{(0)}_A(\mathbf{p}\mu;\mathbf{q}\nu)\chi^{(0)}_D(\mathbf{p}\mu;\mathbf{q}\nu) - \left[\frac{\delta_{AD}}{24}\frac{\beta^2 n_A Z_A^2}{m_A}\mathbf{k}\cdot\mathbf{p}\varphi(p)\frac{\partial^2}{\partial\omega^2}\right]_{\nu,\mu}\left[\frac{T_A(\mathbf{k}\omega;\mathbf{q}\nu)}{\epsilon(\mathbf{p}\mu)}+\frac{T_A(\mathbf{k}\omega;\mathbf{q}\mu)}{\epsilon(\mathbf{p}\nu)}-2\frac{T_A(\mathbf{k}\omega;\mathbf{q}0)}{\epsilon(\mathbf{p}0)}\right]\Bigg]. \tag{41}$$

The partial response functions $\chi^{(1)}_{AB}(\mathbf{k}\omega)$ can be combined into an expression for the first-order correlational correction $\epsilon^{(1)}(\mathbf{k}\omega)$ to the centrally important dielectric-response function by using Eq. (10). In addition, the partial response functions possess their own physical significance, for $\chi^{(1)}_{AB}(\mathbf{k}\omega)$ given by Eqs. (39) and (41) relates directly to the dynamical structure function $S_{AB}(\mathbf{k}\omega)$ via the conventional fluctuation-dissipation theorem. The structure of Eq. (41) contains important physical clues: this point will be elaborated upon in Sec. VI.

A. The static limit

In order to evaluate the static ($\omega=0$) limit result, we effect the interchange of the variables $\mathbf{p}\leftrightarrow\mathbf{q}$, $\nu\leftrightarrow\mu$ in (39); this leaves the integrand invariant, except for the δ_- function. But

$$\lim_{\omega\to 0}[\delta_-(\nu)+\delta_-(\omega-\nu)]=\delta(\nu), \tag{42}$$

and thus the ν integral can immediately be done, which eliminates all but the first term in the integrand. Observing that

$$\chi^{(0)}_A(\mathbf{k}0)=-\beta n_A, \tag{43}$$

$$\chi^{(0)}_A(\mathbf{p}0;\mathbf{q}0)=\tfrac{1}{2}\beta^2 n_A, \tag{44}$$

one then obtains

$$\chi^{(1)}_{AD}(\mathbf{k}0)=-\frac{n_A n_D\beta^3}{2}\frac{1}{V}\sum_{\mathbf{p}}\psi_{AC}(q)\eta_{DC}(\mathbf{q}0)\times\psi_{AE}(p)\eta_{DE}(\mathbf{p}0). \tag{45}$$

This result can be shown to be equivalent to the results of Kalman and Golden [26], Golden and Lu [31], and Kalman [28], which are obtained by applying the linear and quadratic fluctuation dissipation theorems (FDT and QFDT) [31,32] to the first equations of the BBGKY hierarchy at the static limit. Their result is

$$\chi^{(1)}_{AD}(\mathbf{k}0)=\frac{n_D\beta^2}{k^2}\frac{1}{V}\sum_{\mathbf{p}}\mathbf{k}\cdot\mathbf{p}\psi_{AC}(p)\eta_{DC}(\mathbf{p}0)\eta_{DA}(\mathbf{q}0). \tag{46}$$

Using the relation

$$\eta_{DA}=\delta_{DA}+\psi_{DC}\hat{\chi}_{AC}=\delta_{DA}+\psi_{DC}\chi_A\eta_{AC}=\delta_{DA}+\chi_A\psi_{AC}\eta_{DC}, \tag{47}$$

the δ_{DA} term in (46) will vanish because it is an odd function of $\mathbf{p}$:

$$\chi^{(1)}_{AD}(\mathbf{k}0)=-\frac{n_A n_D\beta^3}{k^2}\frac{1}{V}\sum_{\mathbf{p}}\mathbf{k}\cdot\mathbf{p}\psi_{AC}(p)\eta_{DC}(\mathbf{p}0)\times\psi_{AE}(q)\eta_{DE}(\mathbf{q}0)\ . \tag{48}$$

By combining the above expression after the variable change $\mathbf{p}\rightarrow\mathbf{q}$ with its original form, one can immediately recover the result Eq. (45).

In the important two-component case (electron-ion plasma or binary ionic mixture) $\chi^{(1)}_{11}$ and $\chi^{(1)}_{12}$ become

$$\chi^{(1)}_{11}(\mathbf{k}0)=-\frac{n_1^2\beta^3}{2}\frac{1}{V}\sum_{\mathbf{p}}\frac{[\psi_{11}(p)+\beta n_2\|\psi(p)\|][\psi_{11}(q)+\beta n_2\|\psi(q)\|]}{\epsilon(\mathbf{p}0)\epsilon(\mathbf{q}0)}\ ,$$
$$\chi^{(1)}_{12}(\mathbf{k}0)=-\frac{n_1 n_2\beta^3}{2}\frac{1}{V}\sum_{\mathbf{p}}\frac{\psi_{12}(p)\psi_{12}(q)}{\epsilon(\mathbf{p}0)\epsilon(\mathbf{q}0)}\ . \tag{49}$$

For the bare Coulomb interaction (49) reduces to

$$\chi_{11}(\mathbf{k}0)=-Z_1^4 n_1^2\frac{\beta^3}{2}P(\mathbf{k})\ ,$$
$$\chi_{12}(\mathbf{k}0)=-Z_1^2 Z_2^2 n_1 n_1\frac{\beta^3}{2}P(\mathbf{k})\ , \tag{50}$$

where

$$P(\mathbf{k})=\frac{1}{V}\sum_{\mathbf{q}}\frac{\varphi(q)\varphi(p)}{\epsilon(\mathbf{q})\epsilon(\mathbf{p})}\ . \tag{51}$$

More interesting are, actually, the $\hat{\chi}_{11}(\mathbf{k}0)$ and $\hat{\chi}_{12}(\mathbf{k}0)$ expressions which relate directly, via the fluctuation-dissipation theorem, to the partial structure functions $S_{11}(k)$ and $S_{12}(k)$:

$$\hat{\chi}^{(1)}_{11}(\mathbf{k}0)=-Z_1^4 n_1^2\frac{\beta^3}{2}\left[1+\frac{Z_1-Z_2}{Z_1}\frac{Z_2^2 n_2}{Z_1^2 n_1+Z_2^2 n_2}\frac{\kappa^2}{k^2}\right]^2\times\frac{1}{\epsilon^2(\mathbf{k}0)}P(\mathbf{k})\ ,$$
$$\hat{\chi}^{(1)}_{12}(\mathbf{k}0)=-Z_1^2 Z_2^2 n_1 n_2\frac{\beta^3}{2}\times\left[1+\frac{Z_1-Z_2}{Z_1}\frac{Z_2^2 n_2}{Z_1^2 n_1+Z_2^2 n_2}\frac{\kappa^2}{k^2}\right]\times\left[1+\frac{Z_2-Z_1}{Z_2}\frac{Z_1^2 n_1}{Z_1^2 n_1+Z_2^2 n_2}\frac{\kappa^2}{k^2}\right]\times\frac{1}{\epsilon^2(\mathbf{k}0)}P(\mathbf{k})\ , \tag{52}$$

with

$$\kappa^2=4\pi e^2\beta(Z_1^2 n_1+Z_2^2 n_2)\ .$$

These expressions are equivalent to those given earlier by Kalman [28] for the calculation of the pair-correlation functions g_{AB}. Finally, we note that expressions (51) and (52) satisfy, as they should, the generalized multicomponent "compressibility" sum rules given by Totsuji [33].

B. The long-wavelength limit

The long-wavelength ($\mathbf{k}=0$) limit result can also be easily extracted from the result (39). At this limit, one has

$$T_B(\mathbf{k}\rightarrow 0\omega;\mathbf{q}\nu)=\frac{\mathbf{k}\cdot\mathbf{p}}{n_B m_B\beta^2\omega^2}\chi^{(0)}_B(\mathbf{p}\nu) \tag{53}$$

and

$$\lim_{\mathbf{k}\rightarrow 0}\chi^{(0)}_A(\mathbf{p}\mu;\mathbf{q}\nu)=\frac{\mathbf{k}\cdot\mathbf{p}}{2m_A\omega^2}[\chi^{(0)}_A(\mathbf{p}\mu)-\chi^{(0)}_A(\mathbf{p}\nu)]\ . \tag{54}$$

Thus we find the partial response function up to order (k^2),

$$\chi^{(1)}_{AD}(\mathbf{k}\rightarrow 0\omega)=-\frac{1}{V}\sum_{\mathbf{p}}\frac{(\mathbf{k}\cdot\mathbf{p})^2}{m_A m_D\omega^4\beta}\left[\int d\nu\,\delta_-(\nu)[\psi_{DC}\psi_{DE}\eta_{AC}\tilde{\eta}_{AE}(\chi^{(0)}_A-\tilde{\chi}^{(0)}_A)(\chi^{(0)}_D-\tilde{\chi}^{(0)}_D)+\delta_{AD}\psi_{AC}(\tilde{\eta}_{DC}\chi^{(0)}_A+\eta_{DC}\tilde{\chi}^{(0)}_A)]-\delta_{AD}\psi_{AC}\eta_{AC}(\mathbf{p}0)n_A\beta\right]\ , \tag{55}$$

where we have dropped the wave-vector and frequency arguments with the understanding that

$$\psi_{AB}\equiv\psi_{AB}(p),\quad \chi^{(0)}_{AB}\equiv\chi^{(0)}_{AB}(\mathbf{p}\nu),\quad \tilde{\chi}^{(0)}_{AB}\equiv\chi^{(0)}_{AB}(\mathbf{p}\omega-\nu)\ ,$$

etc. Applying the relation

$$\psi_{DE}\eta_{AE}\chi_A^{(0)}=\psi_{DE}\hat{\chi}_{AE}^{(0)}=\psi_{DE}\hat{\chi}_{EA}^{(0)}=\eta_{DA}-\delta_{DA}\ , \tag{56}$$

Eq. (55) becomes

$$\chi_{AD}^{(1)}(\mathbf{k}\to 0\omega)=-\frac{1}{V}\sum_{\mathbf{p}}\frac{(\mathbf{k}\cdot\mathbf{p})^2}{m_A m_D\omega^4\beta}\left[\int d\nu\,\delta_-(\nu)[\,\psi_{DC}\tilde{\eta}_{AC}(\chi_D^{(0)}-\tilde{\chi}_D^{(0)})(\eta_{DA}-\delta_{DA})-\psi_{DC}\eta_{AC}(\chi_D^{(0)}-\tilde{\chi}_D^{(0)})(\tilde{\eta}_{DA}-\delta_{DA})\right.$$
$$\left.+\delta_{AD}\psi_{AC}(\tilde{\eta}_{DC}\chi_A^{(0)}+\eta_{DC}\tilde{\chi}_A^{(0)})]-\delta_{AD}\psi_{AC}\eta_{AC}(\mathbf{p}0)n_A\beta\right]\ . \tag{57}$$

Exploiting the properties of the plus and minus functions, one has

$$\int d\nu\,\delta_-(\nu)\eta_{AC}\chi_D=\eta_{AC}(\mathbf{p}0)\chi_D^{(0)}(\mathbf{p}0)\ ,$$
$$\int d\nu\,\delta_-(\nu)\tilde{\eta}_{AC}\tilde{\chi}_D^{(0)}=0\ , \tag{58}$$

and thus the final result for the partial response function in the $\mathbf{k}=0$ limit can be expressed in the more compact form

$$\chi_{AD}^{(1)}(\mathbf{k}\to 0\omega)=\frac{1}{m_A m_D\omega^4\beta V}\times\sum_{\mathbf{p}}(\mathbf{k}\cdot\mathbf{p})^2\psi_{AC}\times\int d\nu\,\delta_-(\nu)(\eta_{DA}\tilde{\eta}_{DC}-\tilde{\eta}_{DA}\eta_{DC})\times(\chi_D^{(0)}-\tilde{\chi}_D^{(0)})\ . \tag{59}$$

Although the symmetry of the above expression is not manifest, it can be proven with the aid of (40).

The most interesting cases to which (59) applies are again the two-component electron-ion plasma and the binary ionic mixture. For these situations, $\chi_{11}^{(1)}$, $\chi_{12}^{(1)}$, and the dielectric function $\epsilon^{(1)}$ can be displayed concisely as

$$\chi_{11}^{(1)}(\mathbf{k}\to 0\omega)=\frac{k^2}{6\pi^2\beta m_1^2\omega^4}A(\omega)\ , \tag{60}$$

$$\chi_{12}^{(1)}(\mathbf{k}\to 0\omega)=-\frac{k^2}{6\pi^2\beta m_1 m_2\omega^4}A(\omega)\ , \tag{61}$$

and

$$\epsilon^{(1)}(\omega)=-\frac{2e^2}{3\pi\beta\omega^4}\left[\frac{Z_1}{m_1}-\frac{Z_2}{m_2}\right]^2 A(\omega)\ , \tag{62}$$

with

$$A(\omega)=\int_0^\infty dp\,p^4\psi_{12}^2(p)\times\int d\mu\,\delta_-(\mu)\frac{(\chi_1^{(0)}-\tilde{\chi}_1^{(0)})(\chi_2^{(0)}-\tilde{\chi}_2^{(0)})}{\epsilon\tilde{\epsilon}}\ , \tag{63}$$

where we have exploited that $\psi_{AB}(k)\to Z_A Z_B\varphi(k)$ for $k\to 0$.

Specializing further to the Coulomb potential, we have the more explicit expressions for the partial response functions:

$$\chi_{AB}^{(1)}(\mathbf{k}\to 0\omega)=\frac{k^2 Z_A Z_B e^2}{3\omega^4 m_A m_B\beta}\frac{1}{V}\times\sum_{\mathbf{p}}\int d\nu\,\delta_-(\nu)\left[2\delta_{AB}\beta n_B\frac{p^2}{p^2+\kappa^2}+\delta_{AB}\left[\frac{\tilde{\chi}_B^{(0)}}{\epsilon}+\frac{\chi_B^{(0)}}{\tilde{\epsilon}}\right]+\varphi_{AB}\frac{(\chi_A^{(0)}-\tilde{\chi}_A^{(0)})(\chi_B^{(0)}-\tilde{\chi}_B^{(0)})}{\epsilon\tilde{\epsilon}}\right]\ , \tag{64}$$

and the expression for the dielectric function is the same as in Eq. (62), except that the interaction potential is replaced by the bare Coulomb potential, resulting in

$$A(\omega)=(4\pi Z_1 Z_2 e^2)^2\times\int_0^\infty dp\int d\mu\,\delta_-(\mu)\frac{(\chi_1^{(0)}-\tilde{\chi}_1^{(0)})(\chi_2^{(0)}-\tilde{\chi}_2^{(0)})}{\epsilon\tilde{\epsilon}}\ . \tag{65}$$

VI. CONCLUSIONS

In this paper we have presented the most complete perturbation theoretic result for the response functions of a classical multicomponent plasma. The expansion parameter is the plasma parameter (or coupling constant) γ, or in other words, the expansion is in the "uncompensated" e^2, while terms "compensated" by a density factor n, i.e., being of the order $O((e^2 n)^m)$, are included to all orders. Even though the treatment is classical, quantum effects

due to virtual bound-state formation, diffraction, and exchange can phenomenologically be accounted for through a properly chosen pseudopotential. Our derivation is valid for any physically reasonable set of pseudopotentials and for an arbitrary number of components.

In contrast to earlier works [2,3,6,7,28] on the subject, which were restricted to the $\mathbf{k}\to 0$ or $\omega=0$ limits, we present results $\chi_{AB}(\mathbf{k}\omega)$ for the full range of $\mathbf{k}$ and ω values [Eqs. (39) and (41)]. A judicious use of the powerful partial response function formalism allows us to display these results not only in a more compact form than given in earlier works, but also to identify the physical origin of the various contributions. A glance at Eq. (41) [or Eq. (39)] shows that the two terms in it have different physical characteristics. The first term can be characterized as a "collective," while the second as a "scattering" contribution. The scattering term apart from its ϵ denominator is proportional to e^2: it is the result of a direct (dynamically screened) two-particle interaction. The collective term, in turn, is proportional to $e^4 n$, and the presence of the quadratic response functions in it indicates that it is akin to the "triangle" vertex term in the quantum-mechanical calculations. The collective nature of this term is most evident in the double-ϵ denominator, which gives rise to mode-mode interaction and is responsible for the appearance of the peaks at $\omega=\omega_p$ and $\omega=2\omega_p$ in the high-frequency conductivity [2,34]. It is also clear that this "collective" term is the source of the correct static behavior. From $\chi_{AB}(\mathbf{k}\omega)$, we can derive the centrally important $\epsilon(\mathbf{k}\omega)$, and we also display results for the external partial density response functions $\hat{\chi}_{AB}(\mathbf{k}\omega)$ which link to the partial dynamical structure functions $S_{AB}(\mathbf{k}\omega)$, and to their combinations into charge and mass density fluctuation spectra.

In the long-wavelength ($\mathbf{k}\to 0$) limit our general result is given by Eq. (59), while the same limit for the case of bare Coulomb interaction reduces to the relations (64) and (65). This latter result is equivalent (although given in a more compact and physically meaningful notation) to the earlier results of Oberman, Ron, and Dawson [3] and Coste [5] and to the classical limit of the quantum-mechanical calculations by Tzoar and Platzman [35]. In the static ($\omega=0$) limit the result can be translated into relationships for the pair-correlation function or for the static structure function. In this language they are equivalent to those derived by Kalman [28].

Some of the physical characteristics of the two- or multicomponent system are quite distinct from those of the OCP. The main difference is that, as dictated by momentum conservation, the OCP conductivity and related response functions vanish in the $\mathbf{k}\to 0$ limit, while those of the multicomponent system do not. This behavior is easily demonstrated by going to the OCP limit (by setting $Z_1=Z_2$, $m_1=m_2$) in Eq. (62). It is also clear that the symmetric two-component mixture with $Z_1/m_1=Z_2/m_2$ has properties in the $\mathbf{k}\to 0$ limit similar to that of the OCP. Finally, the special role of the hydrogenic plasma ($Z_1=-Z_2$, $n_1=n_2$) is worth pointing out: in the static limit, as pointed out by Coste [5], the lowest-order correlational correction vanishes for such a system.

We contend that our results represent the most complete and most systematic calculations available for the lowest-order correlational contribution to the response functions for any Coulomb system. Similar calculations done for the degenerate electron gas are either restricted to what is equivalent to the "scattering" term [36], or in calculating the "collective" contributions include certain terms arbitrarily while discarding others of comparable significance [37]. Only the formal results are presented in this paper. Concrete calculations for various physical quantities, based on the formulas given here, will be the topic of a separate publication [29,13].

ACKNOWLEDGMENTS

We are grateful to Dr. Jack Jasperse for useful conversations. This work was partially supported by NSF Grant No. ECS-87-13337.

[1] V. I. Perel and G. M. Eliashberg, Zh. Eksp. Teor. Fiz. **41**, 886 (1961) [Sov. Phys.—JETP **14**, 633 (1962)].
[2] J. Dawson and C. Oberman, Phys. Fluids **5**, 517 (1962); **6**, 394 (1962).
[3] C. Oberman, A. Ron, and J. Dawson, *ibid.* **5**, 1514 (1962).
[4] R. Guernsey, Phys. Fluids **5**, 322 (1962).
[5] J. Coste, Nucl. Fusion **5**, 284 (1965); **5**, 293 (1965).
[6] M. G. Kivelson and D. F. DuBois, Phys. Fluids **7**, 1578 (1964).
[7] A. Ron and N. Tzoar, Phys. Rev. **131**, 12 (1963).
[8] W. D. Kraeft, D. Kremp, K. Kilimann, and H. E. DeWitt, Phys. Rev. A **42**, 2340 (1990); G. Ropke, K. Kilimann, D. Kremp, and W. D. Kraeft, Phys. Lett. **68A**, 329 (1978); W. Ebeling and K. Kilimann, Z. Naturforsch. Teil A **44**, 519 (1989); see also Ref. [17].
[9] C. Deutsch, Y. Furutani, and M. M. Gombert, Phys. Rep. **69**, 86 (1981); C. Deutsch, Ann. Phys. (N.Y.) **115**, 404 (1978).
[10] *Strongly Coupled Plasmas,* edited by G. Kalman (Plenum, New York, 1978); *Strongly Coupled Plasma Physics,* edited by F. J. Rogers and H. E. DeWitt (Plenum, New York, 1986).
[11] K. I. Golden, F. Green, and D. Neilson, Phys. Rev. A **31**, 3529 (1985); **32**, 1669 (1985); see also K. I. Golden and G. Kalman, *ibid.* **14**, 1802 (1976).
[12] J. R. Jasperse and B. Basu, Phys. Rev. Lett. **58**, 1423 (1987); B. Basu and J. R. Jasperse, Phys. Rev. A **38**, 3785 (1988).
[13] H. Zhang, Ph.D. thesis, Boston College, 1991.
[14] H. Hellmann, J. Chem. Phys. **3**, 61 (1935).
[15] T. Dunn and A. A. Broyles, Phys. Rev. **157**, 156 (1967).
[16] C. Deutch and M. M. Gombert, J. Math. Phys. **17**, 1077 (1976).
[17] A. A. Barker, J. Chem. Phys. **55**, 751 (1971).
[18] R. G. Storrer, J. Math. Phys. **9**, 964 (1968); Phys. Rev. **176**, 326 (1968).

[19] F. J. Rogers, Phys. Rev. A **29**, 868 (1983).
[20] W. D. Kraeft, D. Kremp, W. Ebeling, and G. Röpke, *Quantum Statistics of Charged Particle System* (Akademie-Verlag, Berlin, 1986).
[21] J. P. Hansen and I. R. MacDonald, Phys. Rev. A **11**, 211 (1975).
[22] See, e.g., T. Iida and R. I. L. Guthrie, *The Physical Properties of Liquid Metals* (Clarendon, Oxford, 1988).
[23] See, e.g., *Ionic Liquids,* edited by K. H. Bennemann *et al.* (Springer-Verlag, New York, 1982).
[24] T. H. Dupree, Phys. Fluids **6**, 1714 (1963).
[25] G. Kalman, Proceedings of the Les Houches Summer School of Theoretical Physics, 1972, edited by C. DeWitt (Gordon and Breach, New York, 1975).
[26] G. Kalman and K. I. Golden, Phys. Rev. A **29**, 844 (1984).
[27] P. Vashishta, P. Bhattacharya, and K. S. Singwi, Phys. Rev. B **10**, 5108 (1974).
[28] G. Kalman, Phys. Rev. A **30**, 627 (1984).
[29] H. Zhang, M. Minella, G. Kalman, J. Jasperse, and K. I. Golden (unpublished).
[30] S. Ichimaru, *Basic Principles of Plasma Physics* (Benjamin, Reading, MA, 1973).
[31] K. I. Golden and Lu De-xin, J. Stats. Phys. **29**, 281 (1982).
[32] K. I. Golden, G. Kalman, and M. B. Silevitch, J. Stat. Phys. **6**, 87 (1972); G. Kalman and Xiao-yu Gu, Phys. Rev. A **36**, 3399 (1097).
[33] H. Totsuji, Phys. Rev. A **10**, 5440 (1988).
[34] P. Carini, G. Kalman, and K. I. Golden, Phys. Rev. A **26**, 1686 (1982).
[35] N. Tzoar and P. M. Platzman, in *Linear and Nonlinear Electron Transport in Solids,* edited by J. T. Devreese and V. E. Vandoren (Plenum, New York, 1976).
[36] A. J. Glick, Phys. Rev. **129**, 1399 (1963); A. Holas, P. K. Aravind, and K. S. Singwi, Phys. Rev. B **20**, 4912 (1979).
[37] P. K. Aravind, A. Holas, and K. S. Singwi, Phys. Rev. B **25**, 561 (1982).