Nonlinear-response-function approach to binary ionic mixtures: Dynamical theory

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(Received 29 April 1985)

The authors generalize the Golden-Kalman one-component-plasma (OCP) nonlinear-response-function approach to the formulation of a dynamical theory for binary-ionic-mixture plasmas. The principal result of the new dynamical theory is a self-consistent approximation scheme for the calculation of linear ionic polarizabilities and collective-mode structure at long wavelengths and arbitrary coupling strengths. The approximation scheme is constructed from the dynamical nonlinear fluctuation-dissipation theorem and the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) kinetic equation prepared in the velocity-average approximation (VAA). Equilibrium three-point correlations, quadratic response functions, and the dynamical superposition approximation are all central elements of the theory. The theory is exact at zero frequency and exactly reproduces the coefficients of the high-frequency-moment-sum-rule expansion through order $1/\omega^4$. Collective-mode calculations based on the new approximation scheme indicate that the (positive) shift in the plasma frequency is temperature dependent at weak coupling and temperature independent at very strong coupling. These calculations, moreover, reproduce the qualitative features of the Hansen-McDonald-Vieillefosse molecular-dynamics data for the dispersion of the optical mode in the strong-coupling regime, while at the same time very nearly reproducing the weak-coupling frequency shift predicted by Baus's microscopic theory. The authors conclude that the temperature-dependent broadening and shifting of the plasma made at weak coupling is controlled primarily by ionic interdiffusion transport. At very strong coupling, the dispersion of the optical mode is almost entirely controlled by the static-correlational parts of the third-frequency-moment-sum-rule coefficient. Finally, new OCP-like formulas are presented for the dispersion and damping of the plasma mode in the special "symmetric" ($e_A/m_A = e_B/m_B$) ionic mixtures.

I. INTRODUCTION

Using a nonlinear-response-function approach, we formulate a promising new dynamical theory of binary-ionic-mixture plasmas at arbitrary coupling.[1] The plasma configuration of the present paper consists of two mobile classical ion species immersed in a uniform, inert, and neutralizing background. The extreme conditions of density and temperature for such a configuration are typical of degenerate stellar matter where the electrons are highly degenerate and rigid, and where the positive nuclei are fully pressure ionized. Examples are the interiors of carbon-oxygen stars in their helium shell-burning phase[2,3] and certain type-I presupernova cores.[3]

The recent computer experimental and theoretical efforts of Hansen, McDonald, and Vieillefosse[4] (HMV) and Baus[5] have provided new information about the dynamical properties of the uniform-background—binary-ionic-mixture model in the weak- and strong-coupling regimes. Structure function and longitudinal collective-mode data generated from molecular-dynamics simulations[4,6] of strongly coupled H^+-He^{2+} mixtures and concomitant theoretical calculations[4] suggest (i) that short-range-static-correlational effects bring about a small positive *temperature-independent* shift in the plasma frequency and (ii) that this shift and the dispersion of the optical mode are structured, respectively, by the $O((k/\omega)^0)$-short-range and $O((k/\omega)^2)$-long-range-static-correlational parts of the third-frequency-moment-sum-rule coefficient. The kinetic theory approach pursued by Baus *et al.*,[5,6] on the other hand, suggests (iii) a *temperature-dependent* shifting and broadening of the plasma mode by $O((k/\omega)^0)$ ionic interdiffusion transport resulting from dynamic collisions. In the present work, we demonstrate that ionic interdiffusion transport plays the principal role at weak coupling and that short- and long-range-static-correlational effects play the principal role at very strong coupling.

Our approximation scheme is a generalization of the Golden-Kalman (GK) one-component-plasma (OCP) scheme.[7] The principal building blocks to its construction are (i) the first Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) kinetic equations linking nonequilibrium one- and two-particle distribution functions [labeled $F_\sigma(1)$ and $G_{\sigma\sigma'}(12)$; $\sigma,\sigma'=A,B$] and (ii) dynamical nonlinear fluctuation-dissipation theorems[8] (NLFDT's) linking n-point ($n=3,4,5,\ldots$) structure functions and nonlinear-(quadratic, cubic, quartic, ...) response functions. The central hypothesis of the theory, the VAA [velocity-average approximation: Eq. (25) (Refs. 7, 9, and 10)] supposes that the correlational part of $G_{\sigma\sigma'}(12)$ can be replaced by a suitably chosen velocity average.

Preliminary calculations indicate that the theory will provide a reliable description of the longitudinal collective modes over a range of coupling strengths [characterized by the plasma parameter $\Gamma=\beta e^2/a$, $a=(3/4\pi n)^{1/3}$, $n=\sum_\sigma (N_\sigma/V)$] spanning the entire fluid regime. At long wavelengths [$k \ll \omega(\beta m_\sigma)^{1/2}$], these calculations reproduce the qualitative features of the HMV molecular-dynamics data for the dispersion of the optical mode in the strong-coupling ($\Gamma \gg 1$) regime,[4] while at the same time very nearly reproducing the $k=0$ weak-coupling ($\Gamma \ll 1$) collective-mode results of Baus's microscopic theory.[5] As far as we know, this is the first time a dynamical theory has succeeded in securing the correct collective-mode properties of binary ionic mixtures in these two extreme coupling regimes.

In the GK approach, a chain of polarizability equations can be generated by combining the first BBGKY equations with the NLFDT's (Secs. III and IV). This combination is made possible by the VAA which converts the velocity-dependent collision terms [in Eq. (24)] into more tractable velocity-independent density-density correlation functions [see Eq. (27)]. The first equation in the chain links linear and quadratic polarizabilities, the second links quadratic and cubic polarizabilities, and so on. Depending upon the degree of accuracy desired, one can truncate the chain at any level. If, for example, all equations after the first one are dropped, then closure is effected by approximating the quadratic polarizability in terms of linear ones. This we do in the present paper. A systematic closure procedure called the "dynamical superposition approximation" (DSA), first developed by Golden and Kalman for the OCP,[7] will be formulated (in Sec. V) to handle the binary-ionic-mixture configuration of the present paper.

The binary-ionic-mixture plasma is obviously a more complex system than the OCP, indeed to a substantial degree so, making the task of this paper far from trivial. When the number of ionic species is greater than 1 (say K), the number of linear polarizabilities is K, but the number of dynamical two-point structure functions is $\frac{1}{2}K(K+1)$. Thus, even for $K=2$, the straightforward application of linear fluctuation-dissipation theorems (FDT's) becomes impossible when formulated, as is customary, in the language of "external" polarizabilities for each species. Some time ago, Vashishta, Bhattacaryya, and Singwi[11] suggested a way of handling the multicomponent situation. They introduced the concept of "partial" response functions describing the response of the system to fictitious external fields which act on each of the species independently. In their paper, Vashishta *et al.*[11] derived simple relationships for the linear partial response functions in the framework of the Singwi-Tosi-Land-Sjolander (STLS) mean-field-theory[12] approximation. In subsequent works, Golden and Kalman[9,13] exploited the concept of partial response functions to set up a general framework for various approximation schemes for strongly coupled multicomponent plasmas. Finally, the recent formulation by Golden and Lu[8] of the fundamental dynamical NLFDT relations for ionic mixtures in the language of quadratic partial response functions has made it possible to go ahead and set up the theory of the present paper.

As emphasized in Ref. 13, the concept of partial response functions is a powerful formal tool; however, it should be clearly understood that it is not more than that. Partial response functions are not directly observable quantities. They describe the response of the plasma mixture to perturbing fields that act on type-A ions or type-B ions only. Such fields never actually occur in normal plasma mixtures, of course, but the concept of them is perfectly reasonable. This kind of perturbation requires that each ion, in addition to its actual electrical charge, be endowed with a weak fictitious "species charge" which can interact only (i) with its corresponding perturbing field, or (ii) with its companion species charges. Even though such species charges, in general, do not exist, there is nothing physically inconsistent in adding them to the system and, once they have completed their task, in letting them vanish.

The plan of the paper now can be sketched as follows: In Sec. II we introduce a variety of dynamical response and correlation functions which are to play a role in the development of the theory. Broadly speaking, the former falls into two main categories: (1) "external" response functions, which connect the plasma response to external field perturbations, and (2) "total" response functions, which connect the response to total (external + plasma) field perturbations. The development of the approximation scheme is carried out in three stages in Secs. III—V. In the first stage (Sec. III), we establish the fundamental VAA-BBGKY kinetic equation for binary ionic mixtures. We next linearize and convert its right-hand-side non-equilibrium two-point density correlation functions into equilibrium three-point structure functions via routine statistical mechanical linear-response calculations. The simple linear external polarizability formula

$$\hat{\alpha}_\sigma(\mathbf{k},\omega)=\hat{\alpha}_\sigma^{\mathrm{RPA}}(\mathbf{k},\omega)[1+\hat{\Gamma}(S_{\sigma\sigma'\sigma''})] \quad (\sigma,\sigma',\sigma''=A,B)$$

then follows; here the coupling correction $\hat{\Gamma}$ [to the random-phase approximation (RPA)] is expressed entirely in terms of dynamical three-point structure functions, $S_{\sigma\sigma'\sigma''}$'s. In the second-stage calculations (Sec. IV), we eliminate the $S_{\sigma\sigma'\sigma''}$'s in favor of the more accessible quadratic partial response functions, $\hat{\chi}_{\sigma\sigma'\sigma''}$'s, by application of the NLFDT.[8] The subsequent conversion of the $\hat{\chi}_{\sigma\sigma'\sigma''}$'s into quadratic *total* polarizabilities, α_{2A},α_{2B}, is next accomplished by supposing that $\hat{\chi}_{\sigma\sigma'\sigma''}$ can be reasonably well approximated by its RPA structure at arbitrary coupling; straightforward algebra leads to the fully *total* polarizability expression

$$\alpha_\sigma(\mathbf{k},\omega)=\alpha_\sigma^{\mathrm{RPA}}(\mathbf{k},\omega)[1+\Gamma(\alpha_{2A},\alpha_{2B})] \quad (\sigma=A,B) .$$

Self-consistency at long wavelengths is then guaranteed in the third stage (Sec. V) by approximating the quadratic polarizabilities in terms of linear ones. The self-consistent pair of coupled ionic polarizability equations resulting from the combination of (46), (49), and (50) comprises the approximation scheme of the present paper. In Sec. VI we calculate the collective-mode structure in the $k=0$, $\Gamma \ll 1$, and $k \to 0, \Gamma \gg 1$ parameter domains. New VAA formulas are also established in these extreme coupling regimes for the dispersion of the long-wavelength plasma mode in "symmetric" ($e_A/m_A=e_B/m_B$) ionic mixtures. Conclusions are drawn in Sec. VII.

II. CORRELATION FUNCTIONS AND RESPONSE FUNCTIONS

Two- and three-point correlation functions and a variety of linear- and quadratic response functions are quantities which play a central role in our dynamical theory. In this section, these quantities are defined. Relationships among the various response functions are then derived and relevant fluctuation-dissipation relations are displayed. Useful symmetry rules for some quadratic response functions are also listed.

Consider a mixture of N_A and N_B classical point ions of like charge in a uniform neutralizing background of degenerate rigid electrons; the entire system occupies the large but bounded volume V. Let m_σ and e_σ $(\sigma=A,B)$ denote the mass and electrical charge of an ion belonging to the σ species; $n_\sigma=N_\sigma/V$ is the partial uniform density of the unperturbed system.

We begin by listing the relevant two- and three-point time-dependent structure functions; they are defined in terms of n-point equilibrium correlations as follows:

$$\langle n_\sigma(\mathbf{k},t)n_{\sigma'}(-\mathbf{k},0)\rangle^{(0)}=(N_\sigma N_{\sigma'})^{1/2}S_{\sigma\sigma'}(\mathbf{k},t)+N_\sigma N_{\sigma'}\delta_{\mathbf{k}}\ , \tag{1}$$

$$\begin{aligned}\langle n_\sigma(\mathbf{k},t)n_{\sigma'}(-\mathbf{q},0)n_{\sigma''}(\mathbf{q}-\mathbf{k},0)\rangle^{(0)}=&(N_\sigma N_{\sigma'}N_{\sigma''})^{1/3}S_{\sigma\sigma'\sigma''}(\mathbf{q},t;\mathbf{k}-\mathbf{q},t)\\&+N_\sigma(N_{\sigma'}N_{\sigma''})^{1/2}\delta_{\mathbf{k}}S_{\sigma'\sigma''}(\mathbf{q},0)+N_{\sigma'}(N_{\sigma''}N_\sigma)^{1/2}\delta_{\mathbf{q}}S_{\sigma''\sigma}(\mathbf{k},t)\\&+N_{\sigma''}(N_\sigma N_{\sigma'})^{1/2}\delta_{\mathbf{k}-\mathbf{q}}S_{\sigma\sigma'}(\mathbf{k},t)+N_\sigma N_{\sigma'}N_{\sigma''}\delta_{\mathbf{k}}\delta_{\mathbf{q}}\ \ (\sigma,\sigma',\sigma''=A,B)\ ,\end{aligned} \tag{2}$$

the angular brackets denote ensemble averaging of the microscopic densities

$$n_\sigma(\mathbf{k},t)=\sum_i \exp[-i\mathbf{k}\cdot\mathbf{x}_i(t)]$$

and the zero superscript indicates that the averaging is to be performed over the equilibrium (unperturbed) system.

Turning next to the response functions—and first among these the partial response functions—we shall suppose that each particle, in addition to its actual charge e_σ, is endowed with a weak fictitious "species charge" q_σ. The species charge can, by definition, interact only (i) with its corresponding scalar potential perturbation $\hat{\phi}_\sigma(\mathbf{k},t)$ and (ii) with another particle carrying a similar charge. From (i), the corresponding potential of the *external* force acting on the particle is then $\hat{U}_\sigma(\mathbf{k},t)=q_\sigma\hat{\phi}_\sigma(\mathbf{k},t)$. A single such partial driving potential produces an average density response (to all orders in $\hat{\phi}_\sigma$) in each ionic species; the latter are linked to the former by linear and nonlinear partial response functions defined through the constitutive relations[8]

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(1)}=\hat{\chi}_{\sigma\sigma'}(\mathbf{k},\omega)\hat{U}^{\sigma'}(\mathbf{k},\omega)\ , \tag{3}$$

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(2)}=\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)\hat{U}^{\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{U}^{\sigma'}(\mathbf{q},\mu)\ \ (\sigma,\sigma',\sigma''=A,B)\ . \tag{4}$$

The significance of the superscripted and subscripted species indices is as follows: repeated indices occurring in a product, one as a superscript, the other as a subscript, denote species summation. Note how the density response of type-σ' ions

$$\langle n_{\sigma'}(\mathbf{k},t)\rangle=\langle n_{\sigma'}(\mathbf{k},t)\rangle^{(1)}+\langle n_{\sigma'}(\mathbf{k},t)\rangle^{(2)}+\cdots \tag{5}$$

enters into the expression

$$U_\sigma(\mathbf{k},t)=\hat{U}_\sigma(\mathbf{k},t)+\psi_\sigma^{\sigma'}(k)\langle n_{\sigma'}(\mathbf{k},t)\rangle \tag{6}$$

for the potential of the *total* force acting on a type-σ ion; here, the Coulomb interaction energy

$$\psi_{\sigma\sigma'}(k)=\frac{4\pi}{k^2}(e_\sigma e_{\sigma'}+\delta_{\sigma\sigma'}q_\sigma q_{\sigma'}) \tag{7}$$

takes account of the additional species charges in accordance with (ii) above.

If, instead of the partial perturbing field $\hat{\phi}_\sigma$, one contemplates a scalar field $\hat{\phi}$ which simultaneously drives both species, then it is customary to define the linear and quadratic *external* polarizabilities through the constitutive relations

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(1)}=-\frac{\hat{\alpha}_\sigma(\mathbf{k},\omega)}{\phi_{\sigma\sigma}(k)}e_\sigma\hat{\phi}(\mathbf{k},\omega)\ , \tag{8}$$

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(1)}=-\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\frac{\hat{\alpha}_\sigma(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)}{\phi_{\sigma\sigma\sigma}(q,|\mathbf{k}-\mathbf{q}|)}e_\sigma\hat{\phi}(\mathbf{q},\mu)e_\sigma\hat{\phi}(\mathbf{k}-\mathbf{q},\omega-\mu)\ , \tag{9}$$

where

$$\phi_{\sigma\sigma'}(k)=\frac{4\pi e_\sigma e_{\sigma'}}{k^2}\ ,$$

$$\phi_{\sigma\sigma'\sigma''}(q,|\mathbf{k}-\mathbf{q}|)=\frac{4\pi i e_\sigma e_{\sigma'} e_{\sigma''}}{q\,|\mathbf{k}-\mathbf{q}|\,k}\quad(\sigma,\sigma',\sigma''=A,B)\ .$$

This eliminates at the same time any further need to make a distinction between species charge and normal charge. In other words, the stipulation that $\hat{\phi}_\sigma=\hat{\phi}_{\sigma'}=\hat{\phi}_{\sigma''}=\cdots=\hat{\phi}$ is tantamount to identifying $q_\sigma\hat{\phi}_\sigma$ as being $e_\sigma\hat{\phi}$, whence

$$\hat{\alpha}_\sigma(\mathbf{k},\omega)=-\hat{\chi}_{\sigma\sigma'}(\mathbf{k},\omega)\phi_\sigma^{\sigma'}(k)\ , \tag{10}$$

$$\hat{\alpha}_\sigma(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)=-\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)\phi_\sigma^{\sigma''\sigma'}(q,|\mathbf{k}-\mathbf{q}|)\quad(\sigma=A,B)\ . \tag{11}$$

Or, one can contemplate a *total* scalar field $\Phi=\hat{\phi}+\sum_\sigma\langle\phi_\sigma\rangle$ which simultaneously drives both species, in which case the constitutive relations

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(1)}=-\frac{\alpha_\sigma(\mathbf{k},\omega)}{\phi_{\sigma\sigma}(k)}e_\sigma\Phi^{(1)}(\mathbf{k},\omega)\ , \tag{12}$$

$$\begin{aligned}\langle n_\sigma(\mathbf{k},\omega)\rangle^{(2)}=&-\frac{\alpha_\sigma(\mathbf{k},\omega)}{\phi_{\sigma\sigma}(k)}e_\sigma\Phi^{(2)}(\mathbf{k},\omega)\\&-\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\frac{\alpha_\sigma(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)}{\phi_{\sigma\sigma\sigma}(q,|\mathbf{k}-\mathbf{q}|)}e_\sigma\Phi^{(1)}(\mathbf{q},\mu)e_\sigma\Phi^{(1)}(\mathbf{k}-\mathbf{q},\omega-\mu)\quad(\sigma=A,B)\end{aligned} \tag{13}$$

define the linear and quadratic total polarizabilities. The important polarizability relations

$$\hat{\alpha}_\sigma(\mathbf{k},\omega)=\frac{\alpha_\sigma(\mathbf{k},\omega)}{\varepsilon(\mathbf{k},\omega)}\ , \tag{14}$$

$$\hat{\alpha}_\sigma(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)=\frac{[1+\alpha_\eta(\mathbf{k},\omega)]\alpha_\sigma(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)-\alpha_\sigma(\mathbf{k},\omega)\alpha_\eta(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)}{\varepsilon(\mathbf{q},\mu)\varepsilon(\mathbf{k}-\mathbf{q},\omega-\mu)\varepsilon(\mathbf{k},\omega)}\quad(\sigma,\eta=A,B:\ \eta\neq\sigma) \tag{15}$$

then follow from (8), (9), (12), (13), and the fact that $\Phi^{(1)}=\hat{\phi}/\varepsilon$; here $\varepsilon=1+\sum_\sigma\alpha_\sigma$ is the dielectric response function.

The link between the response and correlation functions is provided by the linear and nonlinear fluctuation-dissipation relations[8]

$$\mathrm{Im}\left[\frac{\hat{\chi}_{\sigma\sigma'}(\mathbf{k},\omega)}{\omega}\right]=-\frac{\beta}{2}(n_\sigma n_{\sigma'})^{1/2}S_{\sigma\sigma'}(\mathbf{k},\omega)\ , \tag{16}$$

$$\begin{aligned}\mathrm{Re}&\left[\frac{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{p},\nu)}{\mu\nu}-\frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k},-\omega;\mathbf{q},\mu)}{\mu\omega}-\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{p},\nu;-\mathbf{k},-\omega)}{\omega\nu}\right]\\&=-\frac{\beta^2}{4}(n_\sigma n_{\sigma'}n_{\sigma''})^{1/3}S_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{p},\nu)\quad(\sigma,\sigma',\sigma''=A,B;\ \mathbf{k}=\mathbf{p}+\mathbf{q};\ \omega=\mu+\nu)\ .\end{aligned} \tag{17}$$

Finally, the quadratic partial response functions obey fundamental symmetry rules, some of which will be of use in the sequel. We list them here.

Interchange symmetry:

$$\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{p},\nu)=\hat{\chi}_{\sigma\sigma''\sigma'}(\mathbf{p},\nu;\mathbf{q},\mu)\ . \tag{18}$$

Reality condition and spatial reflection invariance:

$$\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{p},\nu)=\hat{\chi}^{*}_{\sigma\sigma'\sigma''}(\mathbf{q},-\mu;\mathbf{p},-\nu)\ . \tag{19}$$

Triangle symmetry:

$$\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},0;\mathbf{p},\omega)=\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k},\omega;\mathbf{q},0)\ , \tag{20a}$$

$$\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{q},\omega;\mathbf{p},0)=\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{p},0;-\mathbf{k},\omega)\ . \tag{20b}$$

Equations (20a) and (20b) are a direct consequence of the nonlinear FDT (17).

The nonlinear FDT relation (17) is one of the two principal building blocks to the construction of the approximation scheme in this paper. The other principal building block is the VAA kinetic equation which we formulate in the next section.

III. VAA RESPONSE-FUNCTION RELATIONS: STAGE-ONE CALCULATIONS

The stage-one calculations of this section are directed primarily at establishing the fundamental VAA relationship between the external linear polarizability of type-σ particles and the time-dependent three-point structure functions defined in the Sec. II.

Let $F_\sigma(\mathbf{x},\mathbf{v};t)$ and $G_{\sigma\sigma'}(\mathbf{x},\mathbf{v};\mathbf{x}',\mathbf{v}';t)$ be one- and two-particle velocity distribution functions normalized to N_σ and $N_\sigma(N_{\sigma'}-\delta_{\sigma\sigma'})$, respectively. The unperturbed state of the ionic mixture is characterized by the equilibrium distributions

$$F_\sigma^{(0)}(v)=\left[\frac{\beta m_\sigma}{2\pi}\right]^{3/2}\exp(-\beta m_\sigma v^2/2)\,, \tag{21}$$

$$G_{\sigma\sigma'}^{(0)}(\mathbf{x},\mathbf{v};\mathbf{x}',\mathbf{v}')=F_\sigma^{(0)}(v)F_{\sigma'}^{(0)}(v')[1+g_{\sigma\sigma'}(|\mathbf{x}-\mathbf{x}'|)]\,, \tag{22}$$

where the static pair correlation function

$$\begin{aligned}g_{\sigma\sigma'}(|\mathbf{x}-\mathbf{x}'|)&=\frac{1}{V}\sum_{\mathbf{q}}g_{\sigma\sigma'}(q)\exp[i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')]\\&=\frac{1}{(N_\sigma N_{\sigma'})^{1/2}}\sum_{\mathbf{q}}(S_{\sigma\sigma'}(\mathbf{q},t=0)-\delta_{\sigma\sigma'})\exp[i\mathbf{q}\cdot(\mathbf{x}-\mathbf{x}')]\quad(\sigma,\sigma'=A,B)\end{aligned} \tag{23}$$

is known from hypernetted-chain (HNC) calculations or from Monte Carlo computer experiments.[14]

The calculation of the average density response of each species to a single driving potential $\hat{\phi}_{\bar\sigma}$ proceeds from the first BBGKY kinetic equation

$$\left[\frac{\partial}{\partial t}+\mathbf{v}\cdot\frac{\partial}{\partial\mathbf{x}}-\delta_{\sigma\bar\sigma}\frac{1}{m_\sigma}\frac{\partial\hat{U}_{\bar\sigma}(\mathbf{x},t)}{\partial\mathbf{x}}\cdot\frac{\partial}{\partial\mathbf{v}}\right]F_\sigma(\mathbf{x},\mathbf{v};t)=-\frac{1}{m_\sigma}\frac{\partial}{\partial\mathbf{v}}\cdot\int d^3v'\int d^3x'\,\mathbf{K}_\sigma^{\sigma'}(|\mathbf{x}-\mathbf{x}'|)G_{\sigma'\sigma}(\mathbf{x}',\mathbf{v}';\mathbf{x},\mathbf{v};t)\quad(\sigma,\bar\sigma=A,B)\,, \tag{24}$$

where $K_{\sigma\sigma'}(|\mathbf{x}-\mathbf{x}'|)=-\nabla\psi_{\sigma\sigma'}(|\mathbf{x}-\mathbf{x}'|)$ is the interaction force between the type σ field ion (at $\mathbf{x}$) and a typical σ' source ion (at $\mathbf{x}'$) [cf. Eq. (7)].

In order to be able to express the right-hand side of (24) in terms of nonequilibrium two-point functions—binary correlations of microscopic densities, we suppose that $G_{\sigma\sigma'}$ is well described by its velocity average in the restricted sense, where only one of the velocity arguments is averaged out, viz.,

$$\begin{aligned}G_{\sigma\sigma'}(\mathbf{x},\mathbf{v};\mathbf{x}',\mathbf{v}';t)&=\tfrac{1}{2}f_\sigma(\mathbf{x},\mathbf{v};t)\int d^3\bar{v}\,G_{\sigma\sigma'}(\mathbf{x},\bar{\mathbf{v}};\mathbf{x}',\mathbf{v}';t)+\tfrac{1}{2}f_{\sigma'}(\mathbf{x}',\mathbf{v}';t)\int d^3\bar{v}'G_{\sigma\sigma'}(\mathbf{x},\mathbf{v};\mathbf{x}',\bar{\mathbf{v}}';t)\,,\\f_\sigma(\mathbf{x},\mathbf{v};t)&\equiv F_\sigma(\mathbf{x},\mathbf{v};t)/\langle n_\sigma(\mathbf{x},t)\rangle\quad(\sigma,\sigma'=A,B)\,.\end{aligned} \tag{25}$$

The VAA ansatz (25) is the central hypothesis of this paper. While (25) is exact in equilibrium and for the case when the ionic plasma is driven by a static perturbation,[10] it is certainly an approximation when a dynamical perturbation is contemplated. The resulting double-velocity space-integral term

$$f_\sigma(\mathbf{x},\mathbf{v};t)\int d^3v\int d^3v'\,G_{\sigma'\sigma}(\mathbf{x}',\mathbf{v}';\mathbf{x},\mathbf{v};t)$$

which replaces $\int d^3v'\,G_{\sigma'\sigma}(\mathbf{x}',\mathbf{v}';\mathbf{x},\mathbf{v};t)$ in (24) can now be expressed in terms of the nonequilibrium two-point function $\langle n_{\sigma'}(\mathbf{x}')n_\sigma(\mathbf{x})\rangle(t)$ since

$$\int d^3v\int d^3v'\,G_{\sigma'\sigma}(\mathbf{x}',\mathbf{v}';\mathbf{x},\mathbf{v};t)=\langle n_{\sigma'}(\mathbf{x}')n_\sigma(\mathbf{x})\rangle(t)-\delta_{\sigma\sigma'}\delta(\mathbf{x}-\mathbf{x}')\langle n_\sigma(\mathbf{x})\rangle(t)\quad(\sigma,\sigma'=A,B)\,. \tag{26}$$

$n_\sigma(\mathbf{x})$ and $n_{\sigma'}(\mathbf{x}')$ are equal-time microscopic densities and the notation $\langle\ \rangle(t)$ refers to the time evolution carried by the Liouville distribution function.

Upon combining (24) to (26) and Fourier transforming, one obtains the VAA kinetic equation,

$$\begin{aligned}&(\omega-\mathbf{k}\cdot\mathbf{v})F_\sigma(\mathbf{k},\mathbf{v};\omega)+\delta_{\sigma\bar\sigma}\frac{1}{m_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\mathbf{q}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\hat{U}_{\bar\sigma}(\mathbf{q},\mu)\cdot\frac{\partial}{\partial\mathbf{v}}F_\sigma(\mathbf{k}-\mathbf{q},\mathbf{v};\omega-\mu)\\&\quad=-\frac{1}{m_\sigma}\frac{1}{V^2}\sum_{\mathbf{q}}\sum_{\mathbf{p}}\mathbf{q}\psi_\sigma^{\sigma'}(q)\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\langle n_{\sigma'}(\mathbf{q})n_\sigma(\mathbf{k}-\mathbf{p}-\mathbf{q})\rangle(\omega-\mu)\cdot\frac{\partial}{\partial\mathbf{v}}f_\sigma(\mathbf{p},\mathbf{v};\mu)\quad(\sigma,\bar\sigma=A,B)\,.\end{aligned} \tag{27}$$

Equation (27) is valid to all orders in $\hat{\phi}_{\bar\sigma}$. The introduction of $\hat{\phi}_{\bar\sigma}$ into the equilibrium system perturbs $F_\sigma^{(0)}$, $\langle n_{\sigma'}n_\sigma\rangle^{(0)}$, etc., by amounts $\Delta F_\sigma=\sum_{n\geq1}F_\sigma^{(n)}$, $\Delta\langle n_{\sigma'}n_\sigma\rangle=\sum_{n\geq1}\langle n_{\sigma'}n_\sigma\rangle^{(n)}$, etc. These perturbation expansions generate from (27) a chain of coupled VAA kinetic equations. Only the first of these,

$$(\omega-\mathbf{k}\cdot\mathbf{v})F_\sigma^{(1)}(\mathbf{k},\mathbf{v};\omega)+\delta_{\sigma\bar\sigma}\frac{1}{m_\sigma}\hat{U}_{\bar\sigma}(\mathbf{k},\omega)\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}}F_\sigma^{(0)}(v)=-\frac{1}{m_\sigma}\frac{\partial F_\sigma^{(0)}(v)}{\partial\mathbf{v}}\cdot\frac{1}{N_\sigma}\sum_{\mathbf{q}}\mathbf{q}\psi_\sigma^{\sigma'}(q)\langle n_{\sigma'}(\mathbf{q})n_\sigma(\mathbf{k}-\mathbf{q})\rangle^{(1)}(\omega)\quad(\sigma,\bar\sigma=A,B) \tag{28}$$

will be of interest in this paper.

The subsequent conversion of the nonequilibrium two-point function into equilibrium three-point functions is effected by performing routine statistical mechanical linear-response calculations.[7,10] We obtain

$$\langle n_{\sigma'}(\mathbf{q})n_\sigma(\mathbf{k}-\mathbf{q})\rangle^{(1)}(\omega)=-\beta\hat{U}_{\bar\sigma}(\mathbf{k},\omega)(n_{\bar\sigma}n_\sigma n_{\sigma'})^{1/3}\left[\frac{i\omega}{2\pi}\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\,\delta_+(\omega-\mu-\nu)S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)\right.$$
$$\left.+S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},t=0;\mathbf{q},t=0)\right]$$
$$-i\beta\hat{U}_{\bar\sigma}(\mathbf{k},\omega)\delta_{\mathbf{q}}(n_{\bar\sigma}n_\sigma)^{1/2}N_{\sigma'}\int_{-\infty}^{\infty}d\mu\,\delta_+(\omega-\mu)\mu S_{\bar\sigma\sigma}(\mathbf{k},\mu)$$
$$-i\beta\hat{U}_{\bar\sigma}(\mathbf{k},\omega)\delta_{\mathbf{k}-\mathbf{q}}(n_{\bar\sigma}n_{\sigma'})^{1/2}N_\sigma\int_{-\infty}^{\infty}d\mu\,\delta_+(\omega-\mu)\mu S_{\bar\sigma\sigma'}(\mathbf{k},\mu) \tag{29a}$$
$$=-\beta\hat{U}_{\bar\sigma}(\mathbf{k},\omega)(n_{\bar\sigma}n_\sigma n_{\sigma'})^{1/3}\left[i\omega\int_0^\infty dt\,e^{i\omega t}S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},t;\mathbf{q},t)+S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},t=0;\mathbf{q},t=0)\right]$$
$$+\hat{U}_{\bar\sigma}(\mathbf{k},\omega)[\delta_{\mathbf{q}}N_{\sigma'}\hat{\chi}_{\bar\sigma\sigma}(\mathbf{k},\omega)+\delta_{\mathbf{k}-\mathbf{q}}N_\sigma\hat{\chi}_{\bar\sigma\sigma'}(\mathbf{k},\omega)]\quad(\sigma,\bar\sigma=A,B)\ . \tag{29b}$$

The conversion of the two-point structure functions into linear $\hat{\chi}$'s in the last step follows from the FDT Eq. (16). The ionic density response

$$\langle n_\sigma(\mathbf{k},\omega)\rangle^{(1)}=\int d^3v\,F_\sigma^{(1)}(\mathbf{k},\mathbf{v};\omega)=\hat{U}_{\bar\sigma}(\mathbf{k},\omega)\frac{\alpha_{\sigma 0}(\mathbf{k},\omega)}{\phi_{\sigma\bar\sigma}(k)}\left[-\delta_{\sigma\bar\sigma}+\hat{\alpha}_{\bar\sigma}(\mathbf{k},\omega)+\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega)\right], \tag{30}$$

in turn, follows from (28), (29b), and (10); here $\phi_{\sigma\bar\sigma}(k)=4\pi e_\sigma e_{\bar\sigma}/k^2$,

$$\alpha_{\sigma 0}(\mathbf{k},\omega)=\frac{\phi_{\sigma\sigma}(k)}{m_\sigma}\int d^3v\frac{\mathbf{k}\cdot\frac{\partial}{\partial\mathbf{v}}F_\sigma^{(0)}(v)}{\omega-\mathbf{k}\cdot\mathbf{v}} \tag{31}$$

is the Vlasov polarizability, and

$$\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega)=\frac{\beta}{N_\sigma}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}(n_{\bar\sigma}n_\sigma n_{\sigma'})^{1/3}$$
$$\times\left[\frac{i\omega}{2\pi}\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\,\delta_+(\omega-\mu-\nu)\right.$$
$$\left.\times S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)+S_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},t=0;\mathbf{q},t=0)\right]\phi_{\bar\sigma}^{\sigma'}(k)\quad(\sigma,\bar\sigma=A,B) \tag{32}$$

is the Γ-dependent dynamical coupling correction.[1] Comparison of (30) with the constitutive relation (3) gives

$$\hat{\chi}_{[\sigma\bar\sigma]}(\mathbf{k},\omega)=\frac{\alpha_{\sigma 0}(\mathbf{k},\omega)}{\phi_{\sigma\bar\sigma}(k)}[-\delta_{\sigma\bar\sigma}+\hat{\alpha}_{\bar\sigma}(\mathbf{k},\omega)+\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega)]\quad(\sigma,\bar\sigma=A,B)\ . \tag{33}$$

Note that the left-hand-side species indices are enclosed in square brackets to indicate that the required $\sigma\leftrightarrow\bar\sigma$ interchange symmetry of the VAA expression (33) is not manifest.[15] This puzzling feature, which was reported some time ago by Golden and Kalman,[9] has recently been clarified at the static level by the same authors:[10,13] they demonstrated (i) that the VAA ansatz (25) is *exact* at $\omega=0$,[10] and (ii) that the matrix elements of $\hat{\boldsymbol{\chi}}(\mathbf{k},\omega=0)$, when explicitly calculated through $O(\gamma)$ in the VAA, are indeed interchange symmetric.[13] Equation (33) is also interchange symmetric at high frequencies $\omega\gg\Omega_\sigma$—at least through $O((\Omega_\sigma/\omega)^4)$. To see this, we note that the coupling correction (32) collapses into the two-point expression[7]

$$\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega\to\infty)=-\frac{\Omega_{\bar\sigma}^2}{\omega^2}\frac{1}{V}\sum_{\mathbf{q}}\chi^2\left[g_{\sigma\bar\sigma}(|\mathbf{k}-\mathbf{q}|)-\delta_{\sigma\bar\sigma}\sum_s\frac{e_sN_s}{e_\sigma N_\sigma}g_{\sigma s}(q)\right],$$
$$\chi\equiv(\mathbf{k}\cdot\mathbf{q})/(kq)\quad(\sigma,\bar\sigma=A,B) \tag{34}$$

for $\omega\gg\Omega_\sigma\equiv(4\pi n_\sigma e_\sigma^2/m_\sigma)^{1/2}$, whence

$$\hat{\chi}'_{[\sigma\bar{\sigma}]}(\mathbf{k},\omega\to\infty)\equiv\mathrm{Re}\hat{\chi}_{[\sigma\bar{\sigma}]}(\mathbf{k},\omega\to\infty)=\left.\frac{\Omega^{(2)}_{\sigma\bar{\sigma}}(\mathbf{k})}{\omega^2}+\frac{\Omega^{(4)}_{\sigma\bar{\sigma}}(\mathbf{k})}{\omega^4}\right|_{\mathrm{VAA}}+\cdots , \tag{35a}$$

$$\Omega^{(2)}_{\sigma\bar{\sigma}}(\mathbf{k})=\Omega^2_\sigma\frac{\delta_{\sigma\bar{\sigma}}}{\phi_{\sigma\bar{\sigma}}(k)} , \tag{35b}$$

$$\Omega^{(4)}_{\sigma\bar{\sigma}}(\mathbf{k})\,|_{\mathrm{VAA}}=\frac{(\Omega_\sigma\Omega_{\bar{\sigma}})^2}{\phi_{\sigma\bar{\sigma}}(k)}\left[1+3\frac{k^2}{\kappa^2_\sigma}\delta_{\sigma\bar{\sigma}}+\frac{1}{V}\sum_{\mathbf{q}}\chi^2 g_{\sigma\bar{\sigma}}(|\mathbf{k}-\mathbf{q}|)-\delta_{\sigma\bar{\sigma}}\sum_s\frac{e_sN_s}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\chi^2 g_{\sigma s}(q)\right]\quad(\sigma,\bar{\sigma}=A,B) , \tag{35c}$$

where $\kappa^2_\sigma=4\pi\beta n_\sigma e^2_\sigma$. We then observe that (35) is *exact* through $O((\Omega_\sigma/\omega)^4)$ since (35c) is identical to the model-independent third-frequency-moment-sum-rule coefficient

$$\Omega^{(4)}_{\sigma\bar{\sigma}}(\mathbf{k})\,|_{\mathrm{exact}}=-\frac{1}{\pi}\int_{-\infty}^{\infty}d\omega\,\omega^3\hat{\chi}''_{\sigma\bar{\sigma}}(\mathbf{k},\omega)=\beta(n_\sigma n_{\bar{\sigma}})^{1/2}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\omega^4 S(\mathbf{k},\omega) \tag{36}$$

calculated from the linear FDT (16). All of the above considerations notwithstanding, any judgment about the symmetry of (33) in the intermediate-frequency range remains speculative and, in any case, is academic since one can always replace (33) with the expression

$$\begin{aligned}\hat{\chi}_{\sigma\bar{\sigma}}(\mathbf{k},\omega)&=\tfrac{1}{2}[\hat{\chi}_{[\sigma\bar{\sigma}]}(\mathbf{k},\omega)+\hat{\chi}_{[\bar{\sigma}\sigma]}(\mathbf{k},\omega)]\\&=\frac{1}{\phi_{\sigma\bar{\sigma}}(k)}\left[-\alpha_{\sigma0}(\mathbf{k},\omega)\delta_{\sigma\bar{\sigma}}+\frac{\alpha_{\sigma0}(\mathbf{k},\omega)\hat{\alpha}_{\bar{\sigma}}(\mathbf{k},\omega)+\alpha_{\bar{\sigma}0}(\mathbf{k},\omega)\hat{\alpha}_{\sigma}(\mathbf{k},\omega)}{2}\right.\\&\qquad\left.+\frac{\alpha_{\sigma0}(\mathbf{k},\omega)\hat{v}_{\bar{\sigma}\sigma}(\mathbf{k},\omega)+\alpha_{\bar{\sigma}0}(\mathbf{k},\omega)\hat{v}_{\sigma\bar{\sigma}}(\mathbf{k},\omega)}{2}\right]\quad(\sigma,\bar{\sigma}=A,B) ,\end{aligned} \tag{37}$$

which guarantees symmetry while leaving unaffected the sum rule conserving feature of the VAA formalism. Summation over species indices according to (10) then leads to the desired relation[1]

$$\begin{aligned}\hat{\alpha}_\sigma(\mathbf{k},\omega)=&\,\hat{\alpha}_{\sigma0}(\mathbf{k},\omega)[1-\hat{v}_{\sigma\sigma}(\mathbf{k},\omega)]-\frac{\hat{\alpha}_{\sigma0}(\mathbf{k},\omega)\alpha_{\eta0}(\mathbf{k},\omega)}{1+\varepsilon_0(\mathbf{k},\omega)}[\hat{v}_{\sigma\sigma}(\mathbf{k},\omega)-\hat{v}_{\eta\eta}(\mathbf{k},\omega)]\\&-\frac{1+\alpha_{\eta0}(\mathbf{k},\omega)}{1+\varepsilon_0(\mathbf{k},\omega)}[\hat{\alpha}_{\eta0}(\mathbf{k},\omega)\hat{v}_{\sigma\eta}(\mathbf{k},\omega)+\hat{\alpha}_{\sigma0}(\mathbf{k},\omega)\hat{v}_{\eta\sigma}(\mathbf{k},\omega)]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\end{aligned} \tag{38}$$

linking the external linear polarizability to the dynamical three-point structure functions. The VAA equations (38) and (32) are valid for arbitrary k values and over the entire frequency domain. The exactness of (38) at $\omega=0$ is especially evident at this stage since the static projection of (37) is identical to the first equations of the BGY hierarchy

$$\begin{aligned}S_{\sigma\bar{\sigma}}(\mathbf{k},t=0)&+\tfrac{1}{2}\beta(n_\sigma n_{\sigma'})^{1/2}S_{\bar{\sigma}\sigma'}(\mathbf{k},t=0)\phi^{\sigma'}_{\sigma}(k)+\tfrac{1}{2}\beta(n_{\bar{\sigma}}n_{\sigma'})^{1/2}S_{\sigma\sigma'}(\mathbf{k},t=0)\phi^{\sigma'}_{\bar{\sigma}}(k)\\&=\delta_{\sigma\bar{\sigma}}-\frac{1}{2}\left[\frac{n_\sigma}{n_{\bar{\sigma}}}\right]^{1/2}\frac{e_\sigma}{e_{\bar{\sigma}}}\hat{v}_{\bar{\sigma}\sigma}(\mathbf{k},0)-\frac{1}{2}\left[\frac{n_{\bar{\sigma}}}{n_\sigma}\right]^{1/2}\frac{e_{\bar{\sigma}}}{e_\sigma}\hat{v}_{\sigma\bar{\sigma}}(\mathbf{k},0) ,\end{aligned} \tag{39a}$$

$$\hat{v}_{\bar{\sigma}\sigma}(\mathbf{k},0)=\frac{\beta}{N_\sigma}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}(n_{\bar{\sigma}}n_\sigma n_{\sigma'})^{1/3}S_{\bar{\sigma}\sigma\sigma'}(\mathbf{k}-\mathbf{q},t=0;\mathbf{q},t=0)\phi^{\sigma'}_{\bar{\sigma}}(k)\quad(\sigma,\bar{\sigma}=A,B) , \tag{39b}$$

linking the static two- and three-point structure functions.[9] As to the exactness of the VAA polarizability at high frequencies $\omega\gg\Omega_\sigma$, the following expression for its real part

$$\hat{\alpha}'_\sigma(\mathbf{k},\omega\to\infty)=-\frac{\Omega^2_\sigma}{\omega^2}-\frac{\Omega^{(4)}_\sigma(\mathbf{k})\,|_{\mathrm{VAA}}}{\omega^4}-\cdots , \tag{40a}$$

$$\Omega^{(4)}_\sigma(\mathbf{k})\,|_{\mathrm{VAA}}=(\Omega_\sigma\Omega)^2+\Omega^4_\sigma\left[3\frac{k^2}{\kappa^2_\sigma}+\sum_s\frac{e_sN_s}{e_\sigma N_\sigma}\frac{1}{V}\sum_q\chi^2\left[\frac{e_s m_\sigma}{e_\sigma m_s}g_{\sigma s}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma s}(q)\right]\right] , \tag{40b}$$

$$\Omega^2=\sum_s\Omega^2_s=\sum_s 4\pi n_s e^2_s/m_s\quad(\sigma=A,B) ,$$

which results from Eqs. (38) and (32) [or from Eqs. (35) and (10)], is seen to be *identical* to the known $\omega\to\infty$ sum-rule expansion through $O((\Omega_\sigma/\omega)^4)$.

IV. STAGE-TWO CALCULATIONS

The stage-two calculations of this section are directed at transforming Eqs. (38) and (32) into relations which involve only linear and quadratic *total* polarizabilities. To accomplish this, we first eliminate the Eq. (32) three-point functions in favor of the more accessible quadratic partial response functions by application of the nonlinear FDT (17). After some algebra (see Ref. 7 and Appendix A for the details), one obtains

$$\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega)=\frac{2}{\beta N_\sigma}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\chi}_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)+\hat{\chi}_{\bar\sigma\sigma\sigma'}(\mathbf{k}-\mathbf{q},\mu;\mathbf{q},\omega-\mu)]\phi_{\bar\sigma}^{\sigma'}(k)\quad(\sigma,\bar\sigma=A,B)\ . \tag{41}$$

The conversion of the Eq. (41) partial response functions into total quadratic polarizabilities is best accomplished at this time by supposing that the triangle-symmetric RPA structure[8]

$$\begin{aligned}i\hat{\chi}_{\bar\sigma\sigma\sigma'}(\mathbf{p},\nu;\mathbf{q},\mu)=\frac{1}{\varepsilon(\mathbf{p},\nu)\varepsilon(\mathbf{q},\mu)\varepsilon(\mathbf{k},\omega)}\Bigg[&\frac{\beta^2 n_{\bar\sigma}}{2}a_{\bar\sigma}(\mathbf{p},\nu;\mathbf{q},\mu)[1+\alpha_{\bar\eta}(\mathbf{k},\omega)]\{\delta_{\bar\sigma\sigma}[1+\alpha_{\bar\eta}(\mathbf{p},\nu)]-\delta_{\bar\eta\sigma}(e_{\bar\sigma}/e_{\bar\eta})\alpha_{\bar\eta}(\mathbf{p},\nu)\}\\&\times\{\delta_{\bar\sigma\sigma'}[1+\alpha_{\bar\eta}(\mathbf{q},\mu)]-\delta_{\bar\eta\sigma'}(e_{\bar\sigma}/e_{\bar\eta})\alpha_{\bar\eta}(\mathbf{q},\mu)\}-\frac{\beta^2 n_{\bar\eta}}{2}a_{\bar\eta}(\mathbf{p},\nu;\mathbf{q},\mu)(e_{\bar\eta}/e_{\bar\sigma})\alpha_{\bar\sigma}(\mathbf{k},\omega)\\&\times\{\delta_{\bar\eta\sigma}[1+\alpha_{\bar\sigma}(\mathbf{p},\nu)]-\delta_{\bar\sigma\sigma}(e_{\bar\eta}/e_{\bar\sigma})\alpha_{\bar\sigma}(\mathbf{p},\nu)\}\\&\times\{\delta_{\bar\eta\sigma'}[1+\alpha_{\bar\sigma}(\mathbf{q},\mu)]-\delta_{\bar\sigma\sigma'}(e_{\bar\eta}/e_{\bar\sigma})\alpha_{\bar\sigma}(\mathbf{q},\mu)\}\Bigg]\end{aligned}$$

$$(\bar\sigma,\sigma,\sigma',\bar\eta=A,B:\ \bar\eta\neq\bar\sigma;\mathbf{k}=\mathbf{p}+\mathbf{q},\omega=\mu+\nu)\ , \tag{42}$$

which prevails in the weak-coupling limit, reliably describes arbitrary coupling situations. (Note the introduction of the "reduced" polarizability $a_{\bar\sigma}(\mathbf{p},\nu;\mathbf{q},\mu)=-2i\alpha_{\bar\sigma}(\mathbf{p},\nu;\mathbf{q},\mu)/[\beta^2 n_{\bar\sigma}\phi_{\bar\sigma\bar\sigma\bar\sigma}(p,q)]$ for notational convenience in the sequel.) The reader can readily verify that the model-independent electrodynamic relation (15) is recovered from the Eq. (42) structure when the latter is summed over species space according to (11); this particular test is crucially important. Substitution of (42) into (41) gives[1]

$$\begin{aligned}\hat{v}_{\bar\sigma\sigma}(\mathbf{k},\omega)=&-\frac{[1+\alpha_{\bar\eta}(\mathbf{k},\omega)]}{\varepsilon(\mathbf{k},\omega)}\left[\delta_{\bar\sigma\sigma}u_{\bar\sigma}(\mathbf{k},\omega)+[\delta_{\bar\sigma\sigma}-(e_{\bar\sigma}/e_{\bar\eta})\delta_{\bar\eta\sigma}]\frac{N_{\bar\sigma}}{N_\sigma}w_{\bar\sigma}(\mathbf{k},\omega)\right]\\&+\frac{\alpha_{\bar\sigma}(\mathbf{k},\omega)}{\varepsilon(\mathbf{k},\omega)}\left[\delta_{\bar\eta\sigma}u_{\bar\eta}(\mathbf{k},\omega)+[\delta_{\bar\eta\sigma}-(e_{\bar\eta}/e_{\bar\sigma})\delta_{\bar\sigma\sigma}]\frac{N_{\bar\eta}}{N_\sigma}w_{\bar\eta}(\mathbf{k},\omega)\right]\quad(\bar\sigma,\sigma,\bar\eta=A,B:\ \bar\eta\neq\bar\sigma)\ ,\end{aligned} \tag{43}$$

where

$$u_{\bar\sigma}(\mathbf{k},\omega)=i\beta n_{\bar\sigma}\phi_{\bar\sigma\bar\sigma}(k)\frac{1}{N_{\bar\sigma}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{\mathbf{q}^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\left[\frac{a_{\bar\sigma}(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)}{\varepsilon(\mathbf{q},\mu)\varepsilon(\mathbf{k}-\mathbf{q},\omega-\mu)}+\frac{a_{\bar\sigma}(\mathbf{q},\omega-\mu;\mathbf{k}-\mathbf{q},\mu)}{\varepsilon(\mathbf{q},\omega-\mu)\varepsilon(\mathbf{k}-\mathbf{q},\mu)}\right] \tag{44}$$

and

$$\begin{aligned}w_{\bar\sigma}(\mathbf{k},\omega)=i\beta n_{\bar\sigma}\phi_{\bar\sigma\bar\sigma}(k)\frac{1}{N_{\bar\sigma}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\Bigg[&\frac{\alpha_{\bar\eta}(\mathbf{k}-\mathbf{q},\omega-\mu)a_{\bar\sigma}(\mathbf{q},\mu;\mathbf{k}-\mathbf{q},\omega-\mu)}{\varepsilon(\mathbf{q},\mu)\varepsilon(\mathbf{k}-\mathbf{q},\omega-\mu)}\\&+\frac{\alpha_{\bar\eta}(\mathbf{k}-\mathbf{q},\mu)a_{\bar\sigma}(\mathbf{q},\omega-\mu;\mathbf{k}-\mathbf{q},\mu)}{\varepsilon(\mathbf{q},\omega-\mu)\varepsilon(\mathbf{k}-\mathbf{q},\mu)}\Bigg]\quad(\bar\sigma,\bar\eta=A,B:\ \bar\eta\neq\bar\sigma)\ .\end{aligned} \tag{45}$$

The polarizability formulas

$$\alpha_\sigma(\mathbf{k},\omega)=\alpha_{\sigma0}(\mathbf{k},\omega)[1+u_\sigma(\mathbf{k},\omega)]+\bar{w}_\sigma(\mathbf{k},\omega)-\frac{\varepsilon_0(\mathbf{k},\omega)}{A(\mathbf{k},\omega)}\left[\frac{e_\eta N_\eta}{e_\sigma N_\sigma}\alpha_{\sigma0}(\mathbf{k},\omega)w_\eta(\mathbf{k},\omega)-\frac{e_\sigma N_\sigma}{e_\eta N_\eta}\alpha_{\eta0}(\mathbf{k},\omega)w_\sigma(\mathbf{k},\omega)\right]\ , \tag{46a}$$

$$\bar{w}_\sigma(\mathbf{k},\omega)=\left[\alpha_{\sigma0}(\mathbf{k},\omega)-\frac{e_\sigma N_\sigma}{e_\eta N_\eta}\alpha_{\eta0}(\mathbf{k},\omega)\right]w_\sigma(\mathbf{k},\omega)\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ , \tag{46b}$$

$$A(\mathbf{k},\omega)=1+\varepsilon_0(\mathbf{k},\omega)+\sum_{\sigma=A,B}[\alpha_{\sigma0}(\mathbf{k},\omega)u_\sigma(\mathbf{k},\omega)+\bar{w}_\sigma(\mathbf{k},\omega)]=1+\varepsilon(\mathbf{k},\omega) \tag{46c}$$

then follow from (38) and (43). This completes the second-stage derivation. The formal operations which transform (32) into (43) do not entail any restrictions on the range of $(\mathbf{k},\omega)$. Consequently, Eqs. (43) to (46) are valid for arbitrary wave numbers and over the entire frequency domain.

Equations (44) to (46) constitute the central relations of our approximation scheme. They determine the linear polarizabilities in terms of the quadratic ones. As such, they are evidently not self-consistent. However, they open up avenues to further approximation methods that lead to self-consistency. To accomplish closure in the preset work, we follow the guidelines set forth by Golden and Kalman for the OCP:[7] we postulate a decomposition of $a_\sigma(\mathbf{p},\nu;\mathbf{q},\mu)$ in terms of linear α's. The more ambitious approach of relegating the closure to a higher level, i.e., expressing $a_\sigma(\mathbf{p},\nu;\mathbf{q},\mu)$ in terms of cubic polarizabilities with the aid of the VAA kinetic equation (27) perturbed to higher order in $\hat{\phi}_{\bar{\sigma}}$, is well beyond the scope of this paper. Indeed, this latter approach has yet to be pursued for the less complicated OCP configuration. In Sec. V we will analyze the RPA quadratic polarizability in the parameter domain $(k/\kappa_\sigma)\ll(\omega/\Omega_\sigma)$ and will show that it has a tractable decomposition in terms of linear polarizabilities. This relationship will then be postulated to serve as the basis of the self-consistency scheme for arbitrary Γ values.

V. STAGE THREE: DYNAMICAL SUPERPOSITION APPROXIMATION AT LONG WAVELENGTHS

The main physical interest lies in the dynamical behavior of binary-ionic-mixture plasmas at long wavelengths $(k\to 0)$, especially the behavior of the long-wavelength plasma mode. We therefore turn now to the (third-stage) derivation of the formulas for $u_\sigma(\mathbf{k}\to\mathbf{0},\omega)$ and $w_\sigma(\mathbf{k}\to\mathbf{0},\omega)$. We begin by supposing that the quadratic polarizability can be described by the RPA structure[8]

$$a_{\sigma 0}(\mathbf{q},\mu;\mathbf{p},\nu)=\frac{i}{\beta m_\sigma n_\sigma}\int d^3v\frac{F_\sigma^{(0)}(v)}{(\omega-\mathbf{k}\cdot\mathbf{v})^2}\left[\mathbf{k}\cdot\mathbf{q}\frac{\mathbf{p}\cdot\mathbf{v}}{\nu-\mathbf{p}\cdot\mathbf{v}}+\mathbf{k}\cdot\mathbf{p}\frac{\mathbf{q}\cdot\mathbf{v}}{\mu-\mathbf{q}\cdot\mathbf{v}}\right]\quad(\mathbf{k}=\mathbf{q}+\mathbf{p};\omega=\mu+\nu)\ . \tag{47}$$

Following the procedure of Ref. 7, we next develop (47) at long wavelengths $|\mathbf{k}\cdot\mathbf{v}|\ll|\omega|$ and introduce the resulting expression into (44). After some algebra (see Ref. 7 for some of the details), one obtains

$$u_{\sigma 0}(\mathbf{k}\to\mathbf{0},\omega)=\left[\frac{\Omega_\sigma}{\omega}\right]^2[u_{\sigma 0}^{\text{stat}}(\mathbf{k})+u_{\sigma 0}^{\text{dyn}}(\mathbf{k}\to\mathbf{0},\omega)]\ , \tag{48a}$$

$$u_{\sigma 0}^{\text{stat}}(\mathbf{k})=\sum_{\sigma'=A,B}\frac{e_{\sigma'}N_{\sigma'}}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma'_0}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma'_0}(q)]\ , \tag{48b}$$

$$\begin{aligned}u_{\sigma 0}^{\text{dyn}}(\mathbf{k}\to\mathbf{0},\omega)=&-\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}(1-6\chi^2+8\chi^4)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}_0(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\\&-\frac{k^2}{2\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}(1-4\chi^2+4\chi^4)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)-\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)]\\&-\lim_{k\to 0}R_{\sigma\eta_0}(\mathbf{k},\omega)\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ ,\end{aligned} \tag{48c}$$

where

$$\begin{aligned}R_{\sigma\eta_0}(\mathbf{k},\omega)=\frac{k}{2\kappa_\sigma^2 N_\sigma}\sum_{\mathbf{q}}q(\chi-2\chi^3)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[&\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)-\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\\&+\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)-\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)]\\&(\sigma,\eta=A,B:\ \eta\neq\sigma)\end{aligned} \tag{48d}$$

and $\hat{\alpha}_0=\sum_\sigma\hat{\alpha}_{\sigma 0}$. The dynamical superposition formula for $u_\sigma(\mathbf{k}\to\mathbf{0},\omega)$,

$$u_\sigma(\mathbf{k}\to\mathbf{0},\omega)=\left[\frac{\Omega_\sigma}{\omega}\right]^2[u_\sigma^{\text{stat}}(\mathbf{k})+u_\sigma^{\text{dyn}}(\mathbf{k}\to\mathbf{0},\omega)]\ , \tag{49a}$$

$$u_\sigma^{\text{stat}}(\mathbf{k})=\sum_{\sigma'=A,B}\frac{e_{\sigma'}N_{\sigma'}}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma'}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma'}(q)]\ , \tag{49b}$$

$$u_\sigma^{\rm dyn}(\mathbf{k}\to\mathbf{0},\omega)=-\frac{3k^2}{5\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}(\mathbf{q},\mu)\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)$$
$$-\frac{7k^2}{30\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)-\hat{\alpha}_\eta(\mathbf{q},\mu)\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)]$$
$$-\lim_{k\to 0}R_{\sigma\eta}(\mathbf{k},\omega)\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,, \tag{49c}$$

which results from (48) when the zero subscripts are dropped, we now propose to be valid for arbitrary coupling. The Ref. 7 procedure also can be adapted to the more involved calculation of $w_{\sigma 0}(\mathbf{k}\to\mathbf{0},\omega)$; details are given in Appendix B. Here too, we suppose that the RPA structure of (B8) can be taken over to describe arbitrary coupling states, viz.,

$$w_\sigma(\mathbf{k}\to\mathbf{0},\omega)=\left[\frac{\Omega_\sigma}{\omega}\right]^2[w_\sigma^{\rm stat}(\mathbf{k})+w_\sigma^{\rm dyn}(\mathbf{k}\to\mathbf{0},\omega)]\,, \tag{50a}$$

$$w_\sigma^{\rm stat}(\mathbf{k})=-\frac{e_\eta N_\eta}{e_\sigma n_\sigma}\left[\frac{1}{V}\sum_{\mathbf{q}}\chi^2 g_{\sigma\eta}(|\mathbf{k}-\mathbf{q}|)+\frac{2}{5}\left[\frac{\Omega_\sigma}{\omega}\right]^2\frac{k^2}{\kappa_\sigma^2}g_{\sigma\eta}(r=0)\right]\,, \tag{50b}$$

$$w_\sigma^{\rm dyn}(\mathbf{k}\to\mathbf{0},\omega)=\frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}q\chi(k-q\chi)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{k}-\mathbf{q},\omega-\mu)+\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)\hat{\alpha}_\eta(\mathbf{k}-\mathbf{q},\mu)]$$
$$-\frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2|\mathbf{k}-\mathbf{q}|^2\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_\sigma(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_\eta(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}(\mathbf{q},\mu)$$
$$+\hat{\alpha}_\sigma(\mathbf{k}-\mathbf{q},\mu)\alpha_\eta(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}(\mathbf{q},\omega-\mu)]$$
$$+\frac{2k}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2(k-q\chi)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)\hat{\alpha}_\eta(\mathbf{k}-\mathbf{q},\mu)$$
$$-\hat{\alpha}_\sigma(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_\eta(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}(\mathbf{q},\mu)]$$
$$+\frac{6}{5}\left[\frac{\Omega_\sigma}{\omega}\right]^2\frac{k^2}{\kappa_\sigma^2}w_\sigma^{\rm dyn}(0,\omega)$$
$$-\frac{3}{5}\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma\omega^2}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\mu^2\hat{\alpha}_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)+(\omega-\mu)^2\hat{\alpha}_\eta(\mathbf{q},\mu)\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)$$
$$+\mu^2\hat{\alpha}_\sigma(\mathbf{q},\mu)\alpha_\eta(\mathbf{q},\mu)\hat{\alpha}(\mathbf{q},\omega-\mu)$$
$$+(\omega-\mu)^2\hat{\alpha}_\sigma(\mathbf{q},\omega-\mu)\alpha_\eta(\mathbf{q},\omega-\mu)\hat{\alpha}(\mathbf{q},\mu)]$$
$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\,, \tag{50c}$$

where $\hat{\alpha}=\sum_\sigma\hat{\alpha}_\sigma$.

The equations (49) are a natural generalization of the Ref. 7 dynamical coupling coefficient $u_{\rm OCP}(\mathbf{k}\to\mathbf{0},\omega)$; as such, $u_\sigma(\mathbf{k}\to\mathbf{0},\omega)$ provides information only about $O(k^2)$ long-range-correlational effects. Equations (50) go much further; they provide information about $O(k^0)$ ionic interdiffusion and short-range-static-correlational effects on the collective-mode structure.

The self-consistent pair of coupled ionic polarizability equations which results from the combination of (46), (49), and (50) comprises the approximation scheme of the present paper. Since the parent VAA Eqs. (44)–(46) are exact at $\omega=0$, the dynamical coupling coefficients $u_\sigma(\mathbf{k}\to\mathbf{0},\omega)$ and $w_\sigma(\mathbf{k}\to\mathbf{0},\omega)$ can be inputted with static pair correlation function data which are assumed to be determined by Monte Carlo simulations or by an independent theoretical approach. At high frequencies $\omega\gg\Omega_\sigma$ *and* Γ *arbitrary*, the correct small-k limit of the sum-rule coefficient $\Omega_\sigma^{(4)}(\mathbf{k})$ [see Eq. (40)] is readily recovered from (46), (49), and (50); thus, internal consistency between the third-stage construction of our approximation scheme and the (exact) VAA expression (40) is guaranteed. Note that the $\hat{\alpha}$-pair- and triple-cluster structures of the *dynamical superposition* formulas (49c) and (50c) have an obvious resemblance to various well-established approximations for the static three-particle correlation function in terms of a superposition of pair correlation function clusters. Because of the inherent RPA-like character of the dynamical polarizability clusters, the Eqs. (49c) and (50c) **q** summations are cut off at the customary $q_{\max}\sim 1/a$ for strong-coupling situations. The nature of the cutoff at weak coupling is discussed at some length in Sec. VI.

Finally, a much simpler approximation scheme can be derived from Eqs. (46b), (46c), and (49) for the exceptional *symmetric* case where $(e_A/m_A)=(e_B/m_B)$, e.g., D^+-He^{2+} mixtures,

$$\sum_{\sigma=A,B}\overline{w}_\sigma(\mathbf{k}\to 0,\omega)=0\ ,$$

$$\sum_{\sigma=A,B}\alpha_{\sigma 0}(\mathbf{k}\to 0,\omega)u_\sigma(\mathbf{k}\to 0,\omega)=-\sum_{\sigma,\sigma'=A,B}\frac{\Omega_\sigma^2\Omega_{\sigma'}^2}{\omega^4}\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma'}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma'}(q)]+\frac{3k^2\Omega^2}{5\beta\rho\omega^4}\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}(\mathbf{q},\mu)\hat{\alpha}(\mathbf{q},\omega-\mu)\ ,$$

where $\Omega^2=\sum_\sigma\Omega_\sigma^2$ and $\rho=\sum_\sigma m_\sigma n_\sigma$.

The self-consistent equations for the *combined* polarizability α

$$\alpha(\mathbf{k}\to 0,\omega)\,|_{\substack{\text{sym.}\\\text{bim}}}=\alpha_0(\mathbf{k}\to 0,\omega)[1+u_{\substack{\text{sym.}\\\text{bim}}}(\mathbf{k}\to 0,\omega)]\ , \tag{51a}$$

$$u_{\substack{\text{sym.}\\\text{bim}}}(\mathbf{k}\to 0,\omega)=\sum_{\sigma,\sigma'=A,B}\left[\frac{\Omega_\sigma\Omega_{\sigma'}}{\omega\Omega}\right]^2\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma'}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma'}(q)]-\frac{3k^2}{5\beta\rho\omega^2}\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}(\mathbf{q},\mu)\hat{\alpha}(\mathbf{q},\omega-\mu) \tag{51b}$$

then follows from (46c). Comparison with their OCP counterparts[7] (obtained by setting $e_A=e_B$ and $m_A=m_B$ with the concentrations left arbitrary)

$$\alpha(\mathbf{k}\to 0,\omega)\,|_{\text{OCP}}=\alpha_0(\mathbf{k}\to 0,\omega)[1+u_{\text{OCP}}(\mathbf{k}\to 0,\omega)]\ , \tag{52a}$$

$$u_{\text{OCP}}(\mathbf{k}\to 0,\omega)=\left[\frac{\Omega}{\omega}\right]^2\left[\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g(|\mathbf{k}-\mathbf{q}|)-g(q)]-\frac{3k^2}{5\kappa^2}\frac{1}{N}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}(\mathbf{q},\mu)\hat{\alpha}(\mathbf{q},\omega-\mu)\right]$$

$$\left[N=\textstyle\sum_\sigma N_\sigma,\ \ \kappa^2=\sum_\sigma\kappa_\sigma^2\right] \tag{52b}$$

illustrates the extent to which such systems are OCP-like: the $O(k^0)$ ionic interdiffusion and short-range-static-correlational effects are now entirely absent. This salient feature of symmetric ionic plasmas has been observed in molecular-dynamics experiments on D^+-He^{2+} mixtures,[4(b)] and it is confirmed at $k=0$ on the basis of an independent microscopic theory.[5]

VI. COLLECTIVE-MODE BEHAVIOR

In this section we analyze the collective-mode structure of binary ionic mixtures in the $k=0$, $\Gamma\ll 1$ and $k\to 0$, $\Gamma\gg 1$ parameter domains.[1] New VAA formulas are also established in these extreme coupling regimes for the dispersion of the long-wavelength plasma mode in symmetric $[(e_A/m_A)=(e_B/m_B)]$ ionic mixtures.

(a) $\Gamma\ll 1$, $\mathbf{k}=0$. First note from Eqs. (49) that $u_\sigma(\mathbf{0},\omega)=0$. The calculation of $w_\sigma(\mathbf{0},\omega)$ at weak coupling is facilitated by rewriting (50a) in a form which makes the distinction not between static and dynamical contributions, but rather between double- and triple-polarizability cluster contributions. Therefore, let

$$w_{\sigma 0}(\mathbf{0},\omega)=\left[\frac{\Omega_\sigma}{\omega}\right]^2[I_\sigma(\omega)+J_\sigma(\omega)]\ , \tag{53a}$$

where

$$I_\sigma(\omega)=\frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\left[\frac{\kappa_\eta^2}{\kappa^2+q^2}-\left[\frac{q}{\kappa_\sigma}\right]^2\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)+\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)]\right] \tag{53b}$$

and

$$J_\sigma(\omega)=-\frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\left[\frac{q}{\kappa_\sigma}\right]^2\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)+\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)]$$

$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \tag{53c}$$

We then observe from (50b) and (50c) that both $w_\sigma^{\rm stat}(0)$ and $w_\sigma^{\rm dyn}(\mathbf{0},\omega)$ [and, consequently, $I_\sigma(\omega)$] exhibit large-q divergences when evaluated in the Debye-Hückel [Eq. (B5b)] and RPA [Eq. (B8c)] limits, respectively. These divergences, however, exactly cancel each other under addition (with or without the screening) leaving us with the expression (see Appendix C for the details)

$$I_\sigma(\omega)=\frac{\gamma n}{6n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\int_0^{x_{\max}}dx\frac{x^4}{(1+x^2)^2}yZ(y)$$
$$=\frac{\gamma n}{6n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\eta^5\int_{\eta/x_{\max}}^{\infty}dy\frac{Z(y)}{y(y^2+\eta^2)^2}\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ ,\tag{54}$$

where the plasma dispersion function $Z(y)$ is given by (C3), $y=\eta/x$, $x=q/\kappa$, $\eta=\omega/\omega_0$, and $\omega_0=\{[4\pi(n_Ae_A^2+n_Be_B^2)]/[m_Am_B/(m_A+m_B)]\}^{1/2}$; the rather involved expressions for the real and imaginary parts of $J_\sigma(\omega)$ are displayed in Appendix C. The plasma parameter $\gamma=\kappa^3/(4\pi n)$ appearing in (54) more appropriately characterizes weak-coupling situations. In deriving (54) from (53b), we have assumed that $\hat\alpha_\sigma(\mathbf{q},\mu)$ and $\hat\alpha_\eta(\mathbf{q},\omega-\mu)$ can be replaced by $\alpha_\sigma(\mathbf{q},\mu)/\varepsilon(\mathbf{q},0)$ and $\alpha_\eta(\mathbf{q},\omega-\mu)/\varepsilon(\mathbf{q},0)$. This static approximation has been used extensively[16] and has been instrumental in earlier calculations[7,17] of the VAA and exact OCP dynamical coupling function $u_{\rm OCP}(\mathbf{k},\omega)$.

The integral $I_\sigma(\omega)$ still exhibits divergences in its real and imaginary parts. The divergence in the latter is the well-known logarithmic one which is handled by the usual $\gamma\ll1$ cutoff $q_{\max}=1/\beta e^2$. The familiar $\gamma\ln\gamma^{-1}$ expression

$$\mathrm{Im}\varepsilon(\mathbf{0},\omega)\,|_{\Gamma\ll1}\cong\left[\frac{2}{\pi}\right]^{1/2}\frac{n}{3}\{(m_A+m_B)[(n_Ae_A^2/m_A)+(n_Be_B^2/m_B)](n_Ae_A^2+n_Be_B^2)\}^{-1/2}$$
$$\times\frac{\kappa_A\kappa_B}{\kappa^2}\left[e_A\left[\frac{m_B}{m_A}\right]^{1/2}-e_B\left[\frac{m_A}{m_B}\right]^{1/2}\right]^2\frac{\Omega\Omega_A\Omega_B}{\omega^3}\gamma\ln\gamma^{-1}\tag{55}$$

then follows from the subsequent evaluation of $\mathrm{Im}w(\mathbf{0},\omega)$ in the domain $(\gamma\omega/\omega_0)\ll1$; again, see Appendix C for the details. The divergence in the real part appears only at high frequencies $(\gamma\omega/\omega_0>1)$ and would adversely affect the $(1/\omega^4)$ structure of $\alpha'_\sigma(\omega\to\infty)\,|_{\rm VAA}$ were it not for the imposition of the cutoff: indeed the Appendix C Eqs. (C15)–(C19) rigorously demonstrate that the $q_{\max}$ cutoff is required to maintain internal consistency with the VAA sum-rule coefficient $\Omega_\sigma^{(4)}(\mathbf{k})\,|_{\rm VAA}$ [cf. Eq. (40b)].

At the lower frequencies $\omega\ll\Omega/\gamma$, the divergence in $\mathrm{Re}w_{\sigma0}(\mathbf{0},\omega)\equiv w'_{\sigma0}(\mathbf{0},\omega)$ disappears so that the VAA collective-mode frequency shift

$$\Delta\omega'(k=0;\gamma\ll1)\,|_{\rm VAA}=\mathrm{Re}\omega(k=0;\gamma\ll1)\,|_{\rm VAA}-\Omega=-\frac{\gamma\Omega}{2}\left[\frac{\partial}{\partial\gamma}\sum_{\sigma=A,B}\bar w'_{\sigma0}(\mathbf{0},\Omega)\right]_{\gamma=0}\tag{56}$$

is cutoff independent as it should be. Numerical calculations for the H^+-He^{2+} mixture with $N_+=N_{2+}$ lead to [see Eqs. (C25)]

$$w_{2+}(\mathbf{0},\Omega)=\tfrac14 w'_+(\mathbf{0},\Omega),\quad w'_+(\mathbf{0},\Omega)\cong0.125\gamma\ ,$$

whence

$$\Delta\omega'(k=0;\gamma\ll1)\,|_{\rm VAA}\cong0.008\gamma\Omega=0.053\Gamma^{3/2}\Omega\ .\tag{57}$$

Our real frequency shift compares favorably with Baus's predicted[5] $\Delta\omega'(k=0;\gamma\ll1)\,|_{\rm Baus}=0.08\Gamma^{3/2}\Omega$ for the same mixture. At weak coupling, our calculations therefore support his contention that the (positive) shift in the plasma frequency is temperature dependent.

(b) *$\Gamma\ll1$, $k\to0$; $(e_A/m_A)=(e_B/m_B)$.* In the previous section the OCP-like character of symmetric ionic mixtures was firmly established by Eqs. (51). The complete absence of the $0(k^0)$ ionic interdiffusion and short-range-static-correlational effects indicates that the collective-mode frequency $\omega(k\to0)$ has the general structure

$$\omega(k\to0)\,|_{(e_A/m_A)=(e_B/m_B)}=\Omega\left[1+[\tfrac32-A(\Gamma)-iB(\Gamma)]\frac{k^2}{\kappa^2}\right]\tag{58}$$

for arbitrary Γ values. For $\Gamma\ll1$, the dispersion and damping coefficient formulas

$$A(\gamma)=-\frac{\gamma\kappa^2}{2k^2}\left[\frac{\partial}{\partial\gamma}u'(\mathbf{k},\Omega)\right]_{\gamma=0},\tag{59a}$$

$$B(\gamma)=-\frac{\gamma\kappa^2}{2k^2}\left[\frac{\partial}{\partial\gamma}u''(\mathbf{k},\Omega)\right]_{\gamma=0}\quad(u_{\rm sym.\,bim}\equiv u=u'+iu'')\tag{59b}$$

can be readily inferred from (58) and the VAA dispersion relation [cf. (51a)]

$$0=\varepsilon(\mathbf{k},\omega(k);\gamma\ll 1)$$
$$\cong \varepsilon_0(\mathbf{k},\omega(k))+\gamma\alpha_0(\mathbf{k},\Omega)\left[\frac{\partial}{\partial\gamma}u(\mathbf{k},\Omega)\right]_{\gamma=0} . \tag{60}$$

The explicit calculation of $u(\mathbf{k}\to\mathbf{0},\omega)$ starting from (51b) follows the procedure of Ref. 7. Again, many of the details are relegated to Appendix C, and we display here only some key steps in the calculations. They are as follows:

$$\begin{aligned}\frac{1}{V}\sum_{\mathbf{q}}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}_0(\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu) &= \sum_{\sigma,\sigma'=A,B}\frac{1}{V}\sum_{\mathbf{q}}\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\alpha}''_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma' 0}(\mathbf{q},\omega-\mu)\\ &\cong \sum_{\sigma,\sigma'=A,B}\frac{1}{V}\sum_{\mathbf{q}}\frac{x^4}{(1+x^2)^2}\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\alpha''_{\sigma 0}(\mathbf{q},\mu)\alpha_{\sigma' 0}(\mathbf{q},\omega-\mu)\\ &= \sum_{\sigma,\sigma'=A,B}\frac{\kappa_\sigma^2\kappa_{\sigma'}^2}{\kappa^4}\left[\frac{m_\sigma}{m_\sigma+m_{\sigma'}}\right]\frac{1}{V}\sum_{\mathbf{q}}\frac{1}{(1+x^2)^2}[1+yZ(y)]\\ &= \frac{1}{2V}\sum_{\mathbf{q}}\frac{1}{(1+x^2)^2}[1+yZ(y)]\\ &= \frac{\gamma n}{4}\left[P(\eta)+i\left[\frac{2}{\pi}\right]^{1/2}Q(\eta)\right],\end{aligned} \tag{61a}$$

where

$$P(\eta)=1-\eta^2\left[1-\left[\frac{\pi}{2}\right]^{1/2}\eta\exp\left[\frac{\eta^2}{2}\right]\mathrm{erfc}\left[\frac{\eta}{\sqrt{2}}\right]\right] \tag{61b}$$

and

$$Q(\eta)=\frac{\eta}{\sqrt{2}}\left[1-\frac{\eta^2}{2}\exp\left[\frac{\eta^2}{2}\right]E_1\left[\frac{\eta^2}{2}\right]\right] \tag{61c}$$

are positive for $0<\eta<1$; $E_1(\eta^2/2)$ is the exponential integral [given in Appendix C, just below (C22)]. Substituting (61a) and

$$\lim_{k\to 0}\left[\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma_0'}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma_0'}(q)]\right]=\frac{-2\gamma}{15}\frac{k^2}{\kappa^2}\frac{\kappa_\sigma\kappa_{\sigma'}}{\kappa^2}\frac{n}{(n_\sigma n_{\sigma'})^{1/2}} \tag{62}$$

into (51b) then gives

$$u_{\substack{\text{sym.}\\ \text{bim}}}(k\to 0,\omega)|_{\gamma\ll 1}=-\frac{n(n_Ae_A^2+n_Be_B^2)}{(n_Ae_A+n_Be_B)^2}\gamma\frac{k^2}{\kappa^2}\left[\frac{\Omega}{\omega}\right]^2\left\{\frac{2}{15}+\frac{3}{20}\left[P(\eta(\omega))+i\left[\frac{2}{\pi}\right]^{1/2}Q(\eta(\omega))\right]\right\}, \tag{63}$$

whence it follows that

$$A(\gamma)=\frac{\gamma}{2}\frac{n(n_Ae_A^2+n_Be_B^2)}{(n_Ae_A+n_Be_B)^2}\left[\frac{2}{15}+\frac{3}{20}P(\eta(\Omega))\right]>0\,, \tag{64a}$$

$$B(\gamma)=\frac{3\gamma}{40}\frac{n(n_Ae_A^2+n_Be_B^2)}{(n_Ae_A+n_Be_B)^2}\left[\frac{2}{\pi}\right]^{1/2}Q(\eta(\Omega))>0\,, \tag{64b}$$

since $\eta(\Omega)=\Omega/\omega_0<1$ *always*. The likeness of our VAA results (63) and (64) to their OCP counterparts[7,17]

$$u_{\mathrm{OCP}}(k\to 0,\omega)|_{\gamma\ll 1}=-\gamma\frac{k^2}{\kappa^2}\left[\frac{\Omega}{\omega}\right]^2\left\{\frac{2}{15}+\frac{3}{20}\left[P(\eta(\omega))+i\left[\frac{2}{\pi}\right]^{1/2}Q(\eta(\omega))\right]\right\}, \tag{65}$$

$$A_{\mathrm{OCP}}(\gamma)=\frac{\gamma}{2}\left[\frac{2}{15}+\frac{3}{20}P\left[\frac{1}{\sqrt{2}}\right]\right], \tag{66a}$$

$$B_{\mathrm{OCP}}(\gamma)=\frac{3\gamma}{40}\left[\frac{2}{\pi}\right]^{1/2}Q\left[\frac{1}{\sqrt{2}}\right], \tag{66b}$$

is remarkable. The lack of an imaginary $O(k^2\gamma\ln\gamma^{-1})$ damping contribution in (63), (64b), (65), and (66b) is to be noted. This defect—which, incidentally, is more significant for small-γ than for large-γ values[7,18]—is a well known but not yet well-understood feature of the VAA approach.[7] Apart from this, it has been recently demonstrated[17] that the OCP coupling function (65) reproduces reasonably well all other important correlational and long-time effects over the entire frequency domain. Our formula (64a) is therefore expected to provide a reliable description of the long-wavelength plasmon dispersion in symmetric ionic mixtures.

(c) *$\Gamma\gg 1$, $k\to 0$.* The dynamical $\hat{\alpha}\hat{\alpha}$ and $\hat{\alpha}\alpha\hat{\alpha}$ cluster terms, while they play an important role in the weak- and intermediate-coupling regimes, contribute only negligibly to the structure of the optical mode at very strong coupling ($\Gamma\gg 1$). To see this, we recall from the analysis of Sec. V that the GK OCP approximation scheme Eqs. (52) (Ref. 7) can be exactly recovered from the present theory simply by setting $m_A=m_B$ and $e_A=e_B$ with the concentrations left arbitrary. We then examine the Carini-Kalman-Golden $\Gamma=110.4$ OCP dispersion curve[18] [which originated from the GK dynamical theory[7] and, consequently, from Eqs. (52)] and observe that this high-Γ curve can be quite accurately reproduced solely from the static pair correlation function part of (52b). Thus, in binary ionic mixtures, $\mathrm{Re}\,u_\sigma^{\mathrm{dyn}}(\mathbf{k}\to 0,\omega)$ should not significantly affect the dispersion of the optical mode for $\Gamma\gg 1$. The same holds true for $\mathrm{Re}\,w_\sigma^{\mathrm{dyn}}(\mathbf{k}\to 0,\omega)$. Indeed, our compressibility-sum-rule-based estimates indicate that both $\mathrm{Re}\,u_\sigma^{\mathrm{dyn}}$ and $\mathrm{Re}\,w_\sigma^{\mathrm{dyn}}$ drop off like $1/\Gamma$ as $\Gamma\to\infty$. It therefore follows that the correlational correction to the dielectric response function:

$$\begin{aligned}\mathrm{Re}[\varepsilon(\mathbf{k}\to\mathbf{0},\omega)\,|_{\Gamma\gg 1}-\varepsilon_0(\mathbf{k}\to\mathbf{0},\omega)]={}&\left[\frac{e_A m_B}{e_B m_A}+\frac{e_B m_A}{e_A m_B}-2\right]\frac{\Omega_A^2\Omega_B^2}{\omega^4}\frac{1}{V}\sum_{\mathbf{q}}\chi^2 g_{AB}(|\mathbf{k}-\mathbf{q}|)\\&-\sum_{\sigma,\sigma'=A,B}\frac{e_\sigma m_{\sigma'}}{e_{\sigma'}m_\sigma}\frac{\Omega_\sigma^2\Omega_{\sigma'}^2}{\omega^4}\frac{1}{V}\sum_{\mathbf{q}}\chi^2[g_{\sigma\sigma'}(|\mathbf{k}-\mathbf{q}|)-g_{\sigma\sigma'}(q)]\\&+\frac{7}{5}\left[\frac{e_A}{e_B}\left[\frac{m_B}{m_A}\right]^{3/2}+\frac{e_B}{e_A}\left[\frac{m_A}{m_B}\right]^{3/2}-\left[\frac{m_B}{m_A}\right]^{1/2}-\left[\frac{m_A}{m_B}\right]^{1/2}\right]\\&\times\frac{\Omega_A^3\Omega_B^3}{\omega^6}\frac{k^2}{\kappa_A\kappa_B}g_{AB}(r=0)\end{aligned}\tag{67}$$

can be constructed solely from (49b) and (50b). Since $(1/V)\sum_{\mathbf{q}}g_{AB}(q)=g_{AB}(r=0)=-1$ for $\Gamma\neq 0$,[14,19,20] the $k=0$ collective-mode frequency

$$\mathrm{Re}\,\omega(k=0)\,|_{\Gamma\gg 1}=\frac{\Omega}{\sqrt{2}}\left\{1+\left[1+\frac{4\Omega_A^2\Omega_B^2}{3\Omega^4}\left[\frac{e_A m_B}{e_B m_A}+\frac{e_B m_A}{e_A m_B}-2\right]\right]^{1/2}\right\}^{1/2}\tag{68}$$

follows from (67). For the H^+-He^{2+} mixture with $N_+=N_{2+}$, Eq. (68) provides $\mathrm{Re}\,\omega(k=0)=1.0198\Omega$ in exact agreement with the result of the Hansen-McDonald-Vieillefosse (HMV) memory-function analysis.[4(b)] Our result (68) certainly supports HMV's contention that, at strong coupling ($\Gamma\gg 1$), the positive shift in Ω is Γ independent. For $0\neq kv_\sigma\ll|\omega|$, the last right-hand-side group ($\propto 1/\omega^6$) in (67) contributes only negligibly to the dispersion. This leaves us with a dispersion relation which is almost entirely controlled by the correlational parts of the third-frequency-moment-sum-rule coefficient $\Omega_\sigma^{(4)}(\mathbf{k})$. Inputting with static pair correlation function data from Ref. 14 would therefore result in a dispersion curve for the optical mode which should coincide with HMV's Fig. 6 sum-rule-moment-based theory curve,[4(b)] and which therefore reproduces the qualitative features of their molecular-dynamics data[4(b)] for $\Gamma\gg 1$. Indeed our high-Γ formula for the collective-mode frequency

$$\mathrm{Re}\,\omega(k\to 0)\,|_{\Gamma\gg 1}\approx\frac{\Omega}{\sqrt{2}}[1+(\tfrac{7}{6}-0.3088k^2a^2)^{1/2}]^{1/2}\,,\tag{69}$$

obtained by inputting (67) with the more accessible Refs. 14 and 21 correlation energy-density formulas, rigorously demonstrates near-coincidence of the two curves for $ka\lesssim 1$.

Finally, we derive from (67) the collective-mode frequency

$$\mathrm{Re}\,\omega(k\to 0)\,|_{\Gamma\gg 1}\approx\frac{\Omega}{\sqrt{2}}\left\{1+\left[1-\frac{16}{45}\left[\frac{ne}{n_Ae_A+n_Be_B}\right]^2\left|\frac{\beta E_c(\Gamma)}{n\Gamma}\right|k^2a^2\right]^{1/2}\right\}^{1/2}\tag{70}$$

characterizing the dispersion of the long-wavelength optical mode in symmetric ionic mixtures; here,

$$\begin{aligned}E_c&=E_{AA}+E_{AB}+E_{BB}\\&=\frac{n_A^2}{2}\int d^3r\,\phi_{AA}(r)g_{AA}(r)+n_An_B\int d^3r\,\phi_{AB}(r)g_{AB}(r)+\frac{n_B^2}{2}\int d^3r\,\phi_{BB}(r)g_{BB}(r)\end{aligned}$$

is the total correlation energy density of the system.

A comprehensive numerical program for analyzing the collective-mode structure at intermediate-coupling states and concomitant calculations of $\Gamma_{\rm crit}$ marking the crossover from plasmonlike to optical phononlike dispersion are deferred to a later work.[22]

VII. CONCLUSIONS

The twofold purpose of the present work has been (i) to formulate a self-consistent approximation scheme for the calculation of the dynamical ionic polarizabilities in strongly coupled binary ionic mixtures, and (ii) to infer from (i) the more important aspects of the $k\to 0$ collective mode behavior in the weak- and strong-coupling regimes.

The principal building blocks to the construction of the approximation scheme are the linearized VAA-BBGKY kinetic equation (28) and the nonlinear fluctuation-dissipation theorem (17). Equilibrium three-point correlations, quadratic response functions, and the dynamical superposition approximation are all central elements in the development of the theory.

The development was carried out in three stages. The stage-one and -two calculations led to the important new formulations (44) to (46) of the linear ionic polarizabilities in terms of quadratic ones; these equations are valid at arbitrary values of k, ω, and Γ. In the $\omega=0$ limit, Eqs. (44)—(46) are *exact* [see Eqs. (39)]. For arbitrary values of k and Γ at high frequency ($\omega \gg \Omega_\sigma$), we demonstrated [cf. Eqs. (40)] that $\hat{\alpha}_\sigma(\mathbf{k},\omega)\,|_{\rm VAA}$ reproduces the frequency-moment-sum-rule expansion for $\mathrm{Re}\hat{\alpha}_\sigma(\mathbf{k},\omega\to\infty)\,|_{\rm exact}$ through $O((\Omega_\sigma/\omega)^4)$, i.e., our dynamical theory *exactly* satisfies the third-frequency-moment-sum rule.

In the stage-three calculations, we made Eqs. (44)—(46) self-consistent in the long-wavelength limit $[k \ll \omega(\beta m_\sigma)^{1/2}]$ by postulating the decomposition of dynamical quadratic polarizabilities in terms of linear ones in analogy with the relation which prevails in this limit for $\Gamma \ll 1$. The result is a pair of coupled equations [obtained from (46), (49), and (50)] for the linear ionic polarizabilities $\alpha_A(\mathbf{k},\omega)$ and $\alpha_B(\mathbf{k},\omega)$. Equations (49) are a natural generalization of the Ref. 7 OCP dynamical coupling coefficient $u_{\rm OCP}(\mathbf{k}\to\mathbf{0},\omega)$; as such, $u_\sigma(\mathbf{k}\to\mathbf{0},\omega)$ provides information only about the $O((k/\omega)^2)$ long-range-correlational effects. Equations (50) go much further: they provide information about $O((k/\omega)^0)$ ionic interdiffusion and short-range-static-correlational effects.

We demonstrated the complete absence of these latter effects in the special symmetric $(e_A/m_A=e_B/m_B)$ ionic mixtures, e.g., D^+-He^{2+} mixtures. Their much simpler self-consistent approximation scheme, Eqs. (51), bears a remarkable likeness to its OCP counterpart.[7]

We analyzed the collective-mode structure of binary ionic mixtures in the $k=0$, $\Gamma \ll 1$ and $k\to 0$, $\Gamma \gg 1$ parameter domains. Our calculations indicate that the (positive) shift in the plasma frequency is Γ dependent at weak coupling [Eq. (57)] and Γ independent at very strong coupling [Eq. (68)]. Our calculations, moreover, reproduce the qualitative features of the HMV molecular-dynamics data for the dispersion of the optical mode in the strong-coupling regime,[4] while at the same time very nearly reproducing the $k=0$ weak-coupling frequency shift predicted by Baus's microscopic theory.[5] Evidently, the temperature-dependent broadening and shifting of the plasma mode at $\Gamma \ll 1$ is controlled primarily by ionic interdiffusion transport. For $\Gamma \gg 1$, the dispersion of the optical mode is almost entirely controlled by the static-correlational contributions to the third-frequency-moment-sum-rule coefficient [see Eq. (67) and the discussion following it].

As to the symmetric plasma mixtures, our new dispersion and damping formulas (58), (64), and (70) again indicate the extent to which such mixtures are OCP-like.

The success of the GK nonlinear-response function approach to the binary-ionic-mixture configuration of the present paper, to the OCP,[7] and to the surface plasma[23] attests to the versatility and power of the approach.

ACKNOWLEDGMENTS

We would like to thank Dr. Louise Turtle for useful discussions. This work was partially supported by U.S. National Science Foundation Grants Nos. ECS-83-15801 and INT-83-20531 and by the Australian-American Educational Foundation. One of the authors (K.I.G.) acknowledges support for this research under the Gordon Godfrey Bequest at the School of Physics, University of New South Wales.

APPENDIX A

In this appendix, we show how the three-point structure function expression for the dynamical coupling correction

$$\hat{v}_{\sigma\sigma'}(\mathbf{k},\omega)=\frac{1}{N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}\left\{\frac{i\omega}{2\pi}\int_{-\infty}^{\infty}d\mu\int_{-\infty}^{\infty}d\nu\,\delta_+(\omega-\mu-\nu)S_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)\right.$$

$$\left.+\,S_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},t=0;\mathbf{q},t=0)\right\}\beta\phi_\sigma^{\sigma''}(k)$$

$$=\hat{v}'_{\sigma\sigma'}(\mathbf{k},\omega)+i\hat{v}''_{\sigma\sigma'}(\mathbf{k},\omega)\;, \tag{A1}$$

$$\hat{v}'_{\sigma\sigma'}(\mathbf{k},\omega)\equiv\mathrm{Re}\hat{v}_{\sigma\sigma'}(\mathbf{k},\omega)$$

$$=-\frac{1}{N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}\mathrm{P}\left[\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\frac{\mu+\nu}{\omega-\mu-\nu}S_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)\beta\phi_\sigma^{\sigma''}(k)\right]\;, \tag{A1a}$$

$$\hat{v}''_{\sigma\sigma'}(\mathbf{k},\omega)\equiv \mathrm{Im}\hat{v}_{\sigma\sigma'}(\mathbf{k},\omega)=\frac{\omega}{2N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}\int_{-\infty}^{\infty}\frac{d\mu}{2\pi}S_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)\beta\phi_\sigma^{\sigma''}(k)\quad(\sigma,\sigma'=A,B) \tag{A1b}$$

is converted into the compact and elegant quadratic response function expression

$$\hat{v}_{\sigma\sigma'}(\mathbf{k},\omega)=\frac{2}{\beta N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)+\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\mu;\mathbf{q},\omega-\mu)]\phi_\sigma^{\sigma''}(k)\quad(\sigma,\sigma'=A.B)\ . \tag{A2}$$

The centrally important VAA formula (A2) contains most, if not all, of the information about dynamical and static-correlational effects in multicomponent plasmas. The following calculations leading to it are a generalization of the earlier Golden-Kalman OCP calculation.

We begin by eliminating the three-point functions in favor of the quadratic $\hat{\chi}$'s by application of the NLFDT Eq. (17). Here we note that $S_{\sigma\sigma'\sigma''}(\mathbf{q},\mu;\mathbf{p},\nu)$ is expected to be nonsingular so that the $\mu=0$, $\nu=0$, and $\omega=0$ singularities in (17) are spurious. Consequently, the NLFDT remains unchanged if one stipulates that each frequency denominator in (17) is a principal-value (P) denominator. With this understanding, the substitution of (17) into (A1) readily yields

$$\hat{v}_{\sigma\sigma'}(\mathbf{k},\omega)=\frac{1}{\beta N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\sum_{s=1}^{4}\mathcal{L}_{\sigma\sigma'}(s)\ , \tag{A3}$$

where

$$\mathcal{L}'_{\sigma\sigma'}(1)=\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)}{\nu(\omega-\mu-\nu)}\phi_\sigma^{\sigma''}(k)\ , \tag{A3a}$$

$$\mathcal{L}'_{\sigma\sigma'}(2)=\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)}{\mu(\omega-\mu-\nu)}\phi_\sigma''(k)\ , \tag{A3b}$$

$$\mathcal{L}'_{\sigma\sigma'}(3)=-\phi_\sigma^{\sigma''}(k)\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma''\sigma\sigma'}(-\mathbf{k},-\mu-\nu;\mathbf{k}-\mathbf{q},\nu)}{\nu(\omega-\mu-\nu)}\ , \tag{A3c}$$

$$\mathcal{L}'_{\sigma\sigma'}(4)=-\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma'\sigma\sigma''}(-\mathbf{k},-\mu-\nu;\mathbf{q},\mu)}{\mu(\omega-\mu-\nu)}\phi_\sigma^{\sigma''}(k)\ , \tag{A3d}$$

$$\mathcal{L}''_{\sigma\sigma'}(1)=-\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\frac{\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)}{\mu}\phi_\sigma^{\sigma''}(k)\ , \tag{A3e}$$

$$\mathcal{L}''_{\sigma\sigma'}(2)=-\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\frac{\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\mu;\mathbf{q},\omega-\mu)}{\mu}\phi_\sigma^{\sigma''}(k)\ , \tag{A3f}$$

$$\mathcal{L}''_{\sigma\sigma'}(3)=\phi_\sigma^{\sigma''}(k)\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\frac{\hat{\chi}'_{\sigma''\sigma\sigma'}(-\mathbf{k},-\omega;\mathbf{k}-\mathbf{q},\omega-\mu)}{\omega-\mu}\ , \tag{A3g}$$

$$\mathcal{L}''_{\sigma\sigma'}(4)=\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\frac{\hat{\chi}'_{\sigma'\sigma\sigma''}(-\mathbf{k},-\omega;\mathbf{q},\mu)}{\mu}\phi_\sigma^{\sigma''}(k)\ , \tag{A3h}$$

and

$$\mathcal{L}_{\sigma\sigma'}(s)=\mathcal{L}'_{\sigma\sigma'}(s)+i\mathcal{L}''_{\sigma\sigma'}(s)\ .$$

The evaluation of the $\mathcal{L}_{\sigma\sigma'}$ integrals then proceeds as follows:

$$\mathcal{L}'_{\sigma\sigma'}(1)\overset{(\mathrm{PB})}{=}\left[\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)+\mathrm{P}\frac{1}{\pi^2}\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu-\nu}\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\nu;\mathbf{q},\mu)\right]\phi_\sigma'^{\,''}(k)$$

$$\overset{(\mathrm{HT})}{=}\left[\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)+\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu}\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)\right]\phi_\sigma^{\sigma''}(k)\ , \tag{A4a}$$

$$\mathcal{L}'_{\sigma\sigma'}(2)\overset{(\mathrm{HT})}{=}\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)\phi_\sigma^{\sigma''}(k)\ , \tag{A4b}$$

$$\mathscr{L}'_{\sigma\sigma'}(3)=-\phi_\sigma^{\sigma''}(k)\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma''\sigma\sigma'}(-\mathbf{k},\mu+\nu;\mathbf{k}-\mathbf{q},-\nu)}{\nu(\omega-\mu-\nu)}$$

$$\overset{(\mathrm{PB})}{=}-\phi_\sigma^{\sigma''}(k)\left[\hat{\chi}'_{\sigma''\sigma\sigma'}(-\mathbf{k},\omega;\mathbf{k}-\mathbf{q},0)+\mathrm{P}\frac{1}{\pi^2}\int_{-\infty}^{\infty}\frac{d\nu}{\nu}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\omega-\mu-\nu}\hat{\chi}'_{\sigma''\sigma\sigma'}(-\mathbf{k},\mu+\nu;\mathbf{k}-\mathbf{q},-\nu)\right]$$

$$\overset{(\mathrm{HT})}{=}0\,, \tag{A4c}$$

$$\mathscr{L}'_{\sigma\sigma'}(4)=-\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\nu}{\pi}\frac{\hat{\chi}'_{\sigma'\sigma\sigma''}(-\mathbf{k},\mu+\nu;\mathbf{q},-\mu)}{\mu(\omega-\mu-\nu)}\phi_\sigma^{\sigma''}(k)$$

$$\overset{(\mathrm{HT})}{=}-\hat{\chi}'_{\sigma'\sigma\sigma''}(-\mathbf{k},\omega;\mathbf{q},0)\phi_\sigma^{\sigma''}(k)=\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega;\mathbf{q},0)\phi_\sigma^{\sigma''}(k)\,, \tag{A4d}$$

$$\mathscr{L}''_{\sigma\sigma'}(3)\overset{(\mathrm{HT})}{=}-\phi_\sigma^{\sigma''}(k)\hat{\chi}''_{\sigma''\sigma\sigma'}(-\mathbf{k},-\omega;\mathbf{k}-\mathbf{q},0)=\phi_\sigma^{\sigma''}(k)\hat{\chi}''_{\sigma''\sigma\sigma'}(-\mathbf{k},\omega;\mathbf{k}-\mathbf{q},0)$$

$$=\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)\phi_\sigma^{\sigma''}(k)\,, \tag{A4e}$$

$$\mathscr{L}''_{\sigma\sigma'}(4)\overset{(\mathrm{HT})}{=}-\hat{\chi}''_{\sigma'\sigma\sigma''}(-\mathbf{k},-\omega;\mathbf{q},0)\phi_\sigma^{\sigma''}(k)=\hat{\chi}''_{\sigma'\sigma\sigma''}(-\mathbf{k},\omega;\mathbf{q},0)\phi_\sigma^{\sigma''}(k)=\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega;\mathbf{q},0)\phi_\sigma^{\sigma''}(k)\,. \tag{A4f}$$

The significance of the "(PB)" and "(HT)" notations above the equal signs is as follows: the (PB) symbol indicates application of the Poincaré-Bertrand theorem[24] to reverse the order of integration; the HT symbol indicates a single or double Hilbert transform operation. The last step in (A4d)—(A4f) follows from the "triangle"-symmetry relations

$$\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k},\omega;\mathbf{k}-\mathbf{q},0)=\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)\,, \tag{A5}$$

$$\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q},0;-\mathbf{k},\omega)=\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega;\mathbf{q},0) \tag{A6}$$

which are themselves a direct consequence of the NLFDT relation (17). Substitution of (A4) into (A3) gives

$$\hat{v}'_{\sigma\sigma'}(\mathbf{k},\omega)=\frac{1}{\beta N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\left[\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)+\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\mu;\mathbf{q},\omega-\mu)+\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)+\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega;\mathbf{q},0)\right]\phi_\sigma^{\sigma''}(k)\,, \tag{A7a}$$

$$\hat{v}''_{\sigma\sigma'}(\mathbf{k},\omega)=-\frac{1}{\beta N_{\sigma'}}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\left[\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega-\mu;\mathbf{q},\mu)+\mathrm{P}\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\chi}'_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\mu;\mathbf{q},\omega-\mu)-\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},0;\mathbf{q},\omega)-\hat{\chi}''_{\sigma\sigma'\sigma''}(\mathbf{k}-\mathbf{q},\omega;\mathbf{q},0)\right]\phi_\sigma^{\sigma''}(k)\,. \tag{A7b}$$

Equation (A2) then results from the addition of (A7a) and (A7b).

APPENDIX B

In this appendix we derive the long-wavelength ($k\to0$) formula for the RPA dynamical coupling function $w_{\sigma0}(\mathbf{k},\omega)$. Following the procedure of Ref. 7, the $|\mathbf{k}\cdot\mathbf{v}|\ll|\omega|$ development of (47) to order k^2 results in the expression

$$a_{\sigma 0}(\mathbf{q},\mu;\mathbf{p},\nu)\,|_{\,|\mathbf{k}\cdot\mathbf{v}|\ll|\omega|} \cong \left[\frac{\Omega_\sigma}{\omega}\right]^2 [A_\sigma^{(1)}(\mathbf{q},\mu)+A_\sigma^{(1)}(\mathbf{p},\nu)]+2\left[\frac{\Omega_\sigma}{\omega}\right]^3 [A_\sigma^{(2)}(\mathbf{q},\mu)+A_\sigma^{(2)}(\mathbf{p},\nu)] + 3\left[\frac{\Omega_\sigma}{\omega}\right]^4 [A_\sigma^{(3)}(\mathbf{q},\mu)+A_\sigma^{(3)}(\mathbf{p},\nu)]\,, \tag{B1a}$$

$$A_\sigma^{(1)}(\mathbf{q},\mu) = -i\frac{(\mathbf{k}\cdot\mathbf{p})q^2}{\kappa_\sigma^4}\alpha_{\sigma 0}(\mathbf{q},\mu)\,, \tag{B1b}$$

$$A_\sigma^{(2)}(\mathbf{q},\mu) = -i\frac{\mu}{\Omega_\sigma}\frac{(\mathbf{k}\cdot\mathbf{p})(\mathbf{k}\cdot\mathbf{q})}{\kappa_\sigma^4}\alpha_{\sigma 0}(\mathbf{q},\mu)\,, \tag{B1c}$$

$$A_\sigma^{(3)}(\mathbf{q},\mu) = -i\frac{k^2q^2(\mathbf{k}\cdot\mathbf{p})}{\kappa_\sigma^6}\left[1-\chi^2+\frac{\mu^2}{\Omega_\sigma^2}\frac{\kappa_\sigma^2}{q^2}\chi^2\right]\alpha_{\sigma 0}(\mathbf{q},\mu) - i\frac{(\mathbf{k}\cdot\mathbf{q})^2(\mathbf{k}\cdot\mathbf{p})}{\kappa_\sigma^4 q^2}\,, \tag{B1d}$$

where $\kappa_\sigma^2 = 4\pi\beta n_\sigma e_\sigma^2$.

We next write $w_{\sigma 0}(\mathbf{k},\omega)$ in the form

$$w_{\sigma 0}(\mathbf{k},\omega) = \left[\frac{\Omega_\sigma}{\omega}\right]^2 w_{\sigma 0}^{(1)} + 2\left[\frac{\Omega_\sigma}{\omega}\right]^3 w_{\sigma 0}^{(2)} + 3\left[\frac{\Omega_\sigma}{\omega}\right]^4 w_{\sigma 0}^{(3)}\,, \tag{B2}$$

$$w_{\sigma 0}^{(l)} = \frac{i\kappa_\sigma^2}{k^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{q^2}\int_{-\infty}^{\infty} d\mu\,\delta_-(\mu)\left[\frac{\alpha_\eta(\mathbf{k}-\mathbf{q},\omega-\mu)[A_\sigma^{(l)}(\mathbf{q},\mu)+A_\sigma^{(l)}(\mathbf{k}-\mathbf{q},\omega-\mu)]}{\varepsilon_0(\mathbf{q},\mu)\varepsilon_0(\mathbf{k}-\mathbf{q},\omega-\mu)} + \frac{\alpha_\eta(\mathbf{k}-\mathbf{q},\mu)[A_\sigma^{(l)}(\mathbf{q},\omega-\mu)+A_\sigma^{(l)}(\mathbf{k}-\mathbf{q},\mu)]}{\varepsilon_0(\mathbf{q},\omega-\mu)\varepsilon_0(\mathbf{k}-\mathbf{q},\mu)}\right]$$

$$(l=1,2,3;\ \sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{B3}$$

For $w_{\sigma 0}^{(1)}$, we have from (B1b) and (B3),

$$w_{\sigma 0}^{(1)} = \frac{\kappa_\sigma^2}{k^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})[\mathbf{k}\cdot(\mathbf{k}-\mathbf{q})]}{\kappa_\sigma^4}\int_{-\infty}^{\infty} d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)+\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)]$$
$$+\frac{\kappa_\sigma^2}{k^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})^2|\mathbf{k}-\mathbf{q}|^2}{q^2\kappa_\sigma^4}\int_{-\infty}^{\infty} d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)$$
$$-\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q}\mu)$$
$$+\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)-\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)]$$

$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{B4}$$

In virtue of the plus-function character of $\hat{\alpha}_\sigma(\omega)$, Eq. (B4) then splits into

$$w_{\sigma 0}^{(1)} = w_{\sigma 0\,\text{stat}}^{(1)} + w_{\sigma 0\,\text{dyn}}^{(1)}\,, \tag{B5a}$$

where

$$\begin{aligned} w_{\sigma 0\,\text{stat}}^{(1)} &= \frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2|\mathbf{k}-\mathbf{q}|^2\int_{-\infty}^{\infty} d\mu\,\delta_-(\mu)\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\mu) \\ &= \frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2|\mathbf{k}-\mathbf{q}|^2\alpha_{\sigma 0}(\mathbf{k}-\mathbf{q},0)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},0) \\ &= \frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},0) \\ &= \frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\frac{\kappa_\eta^2}{|\mathbf{k}-\mathbf{q}|^2+\kappa^2} \equiv \frac{e_\eta N_\eta}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\chi^2\frac{\kappa_\sigma\kappa_\eta/(n_\sigma n_\eta)^{1/2}}{|\mathbf{k}-\mathbf{q}|^2+\kappa^2} \\ &= -\frac{e_\eta N_\eta}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}\chi^2 g_{\sigma\eta_0}(|\mathbf{k}-\mathbf{q}|)\quad\left[\kappa^2=\sum_\sigma\kappa_\sigma^2\right] \end{aligned} \tag{B5b}$$

and

$$w^{(1)}_{\sigma 0\,\mathrm{dyn}}=\frac{1}{N_\sigma}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})[\mathbf{k}\cdot(\mathbf{k}-\mathbf{q})]}{k^2\kappa_\sigma^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)+\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)]$$
$$-\frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\frac{|\mathbf{k}-\mathbf{q}|^2}{\kappa_\sigma^2}\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)$$
$$+\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{B5c}$$

One can similarly show that to order k^2,

$$w^{(2)}_{\sigma 0}=w^{(2)}_{\sigma 0\,\mathrm{dyn}}=\frac{\omega}{\Omega_\sigma}\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}\chi^2\left[1-\frac{q\chi}{k}\right]\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)$$
$$-\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)] \tag{B6}$$

and

$$w^{(3)}_{\sigma 0}=w^{(3)}_{\sigma 0\,\mathrm{stat}}+w^{(3)}_{\sigma 0\,\mathrm{dyn}}\,, \tag{B7a}$$

where

$$w^{(3)}_{\sigma 0\,\mathrm{stat}}=-\frac{k^2}{\kappa_\sigma^2}\frac{e_\eta N_\eta}{e_\sigma N_\sigma}\frac{1}{V}\sum_{\mathbf{q}}(\chi^2-\chi^4)g_{\sigma\eta_0}(q)\,, \tag{B7b}$$

$$w^{(3)}_{\sigma 0\,\mathrm{dyn}}=-\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma}\sum_{\mathbf{q}}(q/\kappa_\sigma)^2(\chi^2-\chi^4)$$
$$\times\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)+\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)$$
$$+\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)+\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)]$$
$$-\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma\Omega_\sigma^2}\sum_{\mathbf{q}}\chi^4\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\mu^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)+(\omega-\mu)^2\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)$$
$$+\mu^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)$$
$$+(\omega-\mu)^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{B7c}$$

Equations (B2) and (B5)—(B7) then combine into the desired long-wavelength formulas

$$w_{\sigma 0}(\mathbf{k}\to\mathbf{0},\omega)=\left[\frac{\Omega_\sigma}{\omega}\right]^2[w^{\mathrm{stat}}_{\sigma 0}(\mathbf{k})+w^{\mathrm{dyn}}_{\sigma 0}(\mathbf{k}\to\mathbf{0},\omega)]\,, \tag{B8a}$$

$$w^{\mathrm{stat}}_{\sigma 0}(\mathbf{k})=-\frac{e_\eta N_\eta}{e_\sigma N_\sigma}\frac{1}{V}\left[\sum_{\mathbf{q}}\chi^2 g_{\sigma\eta_0}(|\mathbf{k}-\mathbf{q}|)+\frac{2}{5}\left[\frac{\Omega_\sigma}{\omega}\right]^2\frac{k^2}{\kappa_\sigma^2}\sum_{\mathbf{q}}g_{\sigma\eta_0}(q)\right]\,, \tag{B8b}$$

$$w_{\sigma 0}^{\text{dyn}}(\mathbf{k}\to\mathbf{0},\omega)=\frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}} q\chi(k-q\chi)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)+\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)]$$
$$-\frac{1}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2|\mathbf{k}-\mathbf{q}|^2\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)$$
$$+\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)]$$
$$+\frac{2k}{N_\sigma\kappa_\sigma^2}\sum_{\mathbf{q}}\chi^2(k-q\chi)\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_{\eta 0}(\mathbf{k}-\mathbf{q},\mu)$$
$$-\hat{\alpha}_{\sigma 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{k}-\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)]$$
$$+\frac{6}{5}\left[\frac{\Omega_\sigma}{\omega}\right]^2\frac{k^2}{\kappa_\sigma^2}w_{\sigma 0}^{\text{dyn}}(\mathbf{0},\omega)$$
$$-3\frac{k^2}{\kappa_\sigma^2}\frac{1}{N_\sigma\omega^2}\sum_{\mathbf{q}}\chi^4\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)[\mu^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)+(\omega-\mu)^2\hat{\alpha}_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)$$
$$+\mu^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{q},\mu)\hat{\alpha}_0(\mathbf{q},\omega-\mu)$$
$$+(\omega-\mu)^2\hat{\alpha}_{\sigma 0}(\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu)\hat{\alpha}_0(\mathbf{q},\mu)]$$
$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \tag{B8c}$$

APPENDIX C

In this appendix, we evaluate the dynamical coupling function $w_\sigma(\mathbf{k},\omega)$ at $k=0$ and in the weak-coupling limit $\gamma\ll 1$. Equation (53a)—(53c) are the appropriate starting point, and we begin with the calculation of I_σ. Let

$$\hat{H}_{\sigma\eta}(\mathbf{q},\omega)=\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)=2i\int_{-\infty}^{\infty}d\mu\,\delta_-(\mu)\hat{\alpha}''_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)$$
$$=\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\hat{\alpha}''_\sigma(\mathbf{q},\mu)\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)\ . \tag{C1}$$

The second step in (C1) exploits the fact that $\hat{\alpha}_\sigma(\mathbf{q},\mu)=\hat{\alpha}'_\sigma(\mathbf{q},\mu)+i\hat{\alpha}''_\sigma(\mathbf{q},\mu)=\hat{\alpha}^*_\sigma(\mathbf{q},\mu)+2i\hat{\alpha}''_\sigma(\mathbf{q},\mu)$ is a plus function and therefore $\hat{\alpha}^*_\sigma(\mathbf{q},\mu)$ as well as $\hat{\alpha}_\eta(\mathbf{q},\omega-\mu)$ are minus functions of μ. The third step follows from the fact that $\hat{\alpha}''_\sigma(\mathbf{q},0)=0$. We next suppose that $\hat{\alpha}_{\sigma 0}(\mathbf{q},\mu)$ and $\hat{\alpha}_{\eta 0}(\mathbf{q},\omega-\mu)$ can be replaced by $\alpha_{\sigma 0}(\mathbf{q},\mu)/\epsilon_0(\mathbf{q},0)$ and $\alpha_{\eta 0}(\mathbf{q},\omega-\mu)/\epsilon_0(\mathbf{q},0)$, respectively, whence

$$\hat{H}_{\sigma\eta_0}(\mathbf{q},\omega)\cong\frac{x^4}{(1+x^2)^2}H_{\sigma\eta_0}(\mathbf{q},\omega)\ , \tag{C2a}$$

$$H_{\sigma\eta_0}(\mathbf{q},\omega)=\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}\alpha''_{\sigma 0}(\mathbf{q},\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu),\quad x=q/\kappa\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \tag{C2b}$$

This static screening approximation has been used rather extensively[16] and has been instrumental in earlier calculations[7,17] of the VAA and exact OCP dynamical coupling function $u_{\text{OCP}}(\mathbf{k},\omega)$. The RPA expression for $\alpha_\sigma(\mathbf{q},\nu)$ in terms of the plasma dispersion function

$$Z(\bar{\nu})=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}dz\frac{\exp(-z^2/2)}{z-\bar{\nu}-io} \tag{C3}$$

(o is a positive infinitesimal quantity) is

$$\alpha_{\sigma 0}(\mathbf{q},\nu)=\left[\frac{\kappa_\sigma}{q}\right]^2[1+\bar{\nu}_\sigma Z(\bar{\nu}_\sigma)]\ , \tag{C4}$$

where $\bar{\nu}_\sigma=\nu(\beta m_\sigma)^{1/2}/q=(m_\sigma/m_\eta)^{1/2}\bar{\nu}_\eta$. Introducing (C4) into (C2b), one obtains

$$H_{\sigma\eta_0}(\mathbf{q},\omega)=\frac{\kappa_\sigma^2\kappa_\eta^2}{\pi q^4}[\pi+Y_{\text{I}}(\mathbf{q},\omega)+Y_{\text{II}}(\mathbf{q},\omega)]\ , \tag{C5a}$$

$$Y_{\rm I}(\mathbf{q},\omega)=\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\omega_\eta\int_{-\infty}^{\infty}d\bar\mu_\eta Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z(\bar\omega_\eta+\bar\mu_\eta)$$

$$=\frac{1}{2}\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\omega_\eta\int_{-\infty}^{\infty}d\bar\mu_\eta\exp[-(m_\sigma/m_\eta)\bar\mu_\eta^2/2]\int_{-\infty}^{\infty}dt\frac{\exp(-t^2/2)}{t-\bar\mu_\eta-\bar\omega_\eta-io}\ , \tag{C5b}$$

$$Y_{\rm II}(\mathbf{q},\omega)=\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\int_{-\infty}^{\infty}d\bar\mu_\eta\bar\mu_\eta Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z(\bar\omega_\eta+\bar\mu_\eta)$$

$$=\frac{1}{2}\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\int_{-\infty}^{\infty}d\bar\mu_\eta\bar\mu_\eta\exp[-(m_\sigma/m_\eta)\bar\mu_\eta^2/2]\int_{-\infty}^{\infty}dt\frac{\exp(-t^2/2)}{t-\bar\mu_\eta-\bar\omega_\eta-io}\ , \tag{C5c}$$

where $\bar\mu_\eta=\mu(\beta m_\eta)^{1/2}/q$, $\bar\omega_\eta=\omega(\beta m_\eta)^{1/2}/q$, and $Z=Z'+iZ''$. The successive transformations

$$t-\bar\mu_\eta=\frac{1}{2}\left[1+\frac{m_\eta}{m_\sigma}\right]s,\quad dt=\frac{1}{2}\left[1+\frac{m_\eta}{m_\sigma}\right]ds\ , \tag{C6}$$

$$\left[1+\frac{m_\sigma}{m_\eta}\right]^{1/2}\left[2\bar\mu_\eta+\frac{m_\eta}{m_\sigma}s\right]=z,\quad 2\left[1+\frac{m_\sigma}{m_\eta}\right]^{1/2}d\bar\mu_\eta=dz\ , \tag{C7}$$

when applied to (C5b), then give

$$Y_{\rm I}(\mathbf{q},\omega)=\frac{1}{4}\bar\omega_\eta\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\left[1+\frac{m_\eta}{m_\sigma}\right]\int_{-\infty}^{\infty}ds\frac{\exp\{-[1+(m_\eta/m_\sigma)]^2s^2/8\}}{\frac{1}{2}[1+(m_\eta/m_\sigma)]s-\bar\omega_\eta-io}$$

$$\times\int_{-\infty}^{\infty}d\bar\mu_\eta\exp\{-[1+(m_\sigma/m_\eta)]\bar\mu_\eta^2/2-[1+(m_\eta/m_\sigma)]s\bar\mu_\eta/2\}$$

$$=\frac{1}{8}\bar\omega_\eta\left[1+\frac{m_\eta}{m_\sigma}\right]^{1/2}\int_{-\infty}^{\infty}ds\frac{\exp\{-[1+(m_\eta/m_\sigma)]s^2/8\}}{\frac{1}{2}[1+(m_\eta/m_\sigma)]s-\bar\omega_\eta-io}\int_{-\infty}^{\infty}dz\,e^{-z^2/8}=\pi yZ(y)$$

$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\ , \tag{C8}$$

with

$$y=\frac{\bar\omega_\eta}{[1+(m_\eta/m_\sigma)]^{1/2}}=\frac{\bar\omega_\sigma}{[1+(m_\sigma/m_\eta)]^{1/2}}=\frac{\omega}{\omega_0}\frac{1}{x}\ ,$$

$$\omega_0=\left[\frac{4\pi(n_Ae_A^2+n_Be_B^2)}{m_Am_B/(m_A+m_B)}\right]^{1/2}.$$

Similarly,

$$Y_{\rm II}(\mathbf{q},\omega)=-\pi\frac{m_\eta}{m_\sigma+m_\eta}[1+yZ(y)]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \tag{C9}$$

$H_{\sigma\eta_0}(\mathbf{q},\omega)$ is now given by

$$H_{\sigma\eta_0}(\mathbf{q},\omega)=\frac{\kappa_\sigma^2\kappa_\eta^2}{q^4}\left[\frac{m_\sigma}{m_\sigma+m_\eta}\right][1+yZ(y)]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\ , \tag{C10}$$

whence from (C2a) and (53b),

$$I_\sigma(\omega)\cong\left[\frac{\kappa_\eta}{\kappa}\right]^2\frac{1}{N_\sigma}\sum_{x<x_{\rm max}}\chi^2\left[\frac{1}{(1+x^2)^2}-\frac{x^2}{(1+x^2)^2}yZ(y)\right]$$

$$=\frac{\gamma n}{6n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2-\frac{2\gamma n}{3\pi n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\int_0^{x_{\rm max}}dx\frac{x^4}{(1+x^2)^2}yZ(y)$$

$$=\frac{\gamma n}{6n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2-\frac{2\gamma n}{3\pi n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\eta^5\int_{\eta/x_{\rm max}}^{\infty}dy\frac{Z(y)}{y(y^2+\eta^2)^2}\ , \tag{C11}$$

where $\eta=\omega/\omega_0$. Note that in the course of going from (53b) to (C11), the large-q divergences in both $w_{\sigma 0}^{\text{stat}}(0)$ and $w_{\sigma 0}^{\text{dyn}}(0,\omega)$ have exactly canceled each other out. Equation (C11) nevertheless still exhibits divergences in its real and imaginary parts. The divergence in the latter is the well-known logarithmic one which is handled by the usual $\gamma \ll 1$, $q_{\max}=1/\beta e^2$ cutoff.

The more involved calculation of $J_\sigma(\omega)$ starting from its static screening approximation expression

$$J_\sigma(\omega)\cong\left[\frac{\kappa}{\kappa_\sigma}\right]^2\frac{1}{3N_\sigma}\sum_{q<q_{\max}}\frac{x^6}{(1+x^2)^2}\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{d\mu}{\mu}[\alpha''_{\sigma 0}(\mathbf{q},\mu)\alpha'_{\eta 0}(\mathbf{q},\mu)\alpha_0(\mathbf{q},\omega-\mu)$$

$$+\alpha'_{\sigma 0}(\mathbf{q},\mu)\alpha''_{\eta 0}(\mathbf{q},\mu)\alpha_0(\mathbf{q},\omega-\mu)+\alpha_{\sigma 0}(\mathbf{q},\omega-\mu)\alpha_{\eta 0}(\mathbf{q},\omega-\mu)\alpha''_0(\mathbf{q},\mu)]$$

$$(\sigma,\eta=A,B:\ \eta\neq\sigma) \quad \text{(C12)}$$

proceeds along similar lines. Directing our attention primarily at the real part, some rather lengthy algebra leads to

$$\begin{aligned}
J'_\sigma(\omega)\cong &-\frac{\gamma n}{3\pi n_\sigma}\left[\frac{\omega}{\Omega_\sigma}\right]^3\left[2\frac{\kappa_\eta^2\kappa_\sigma^3}{\kappa^5}\left[\frac{m_\eta}{m_\sigma+m_\eta}\right]^{3/2}+\frac{\kappa_\eta^2\kappa_\sigma^5}{\sqrt{2}\kappa^7}+\frac{\kappa_\eta^7}{\sqrt{2}\kappa^7}\left[\frac{\Omega_\sigma}{\Omega_\eta}\right]^3\right]\int_{\eta/x_{\max}}^{\infty}dy\frac{yZ'(y)}{(y^2+\eta^2)^2}\\
&-\frac{\kappa_\eta^2\kappa_\sigma^2}{\kappa^4}\frac{m_\sigma}{m_\eta}\frac{1}{3N_\sigma}\sum_{x<x_{\max}}\frac{1}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^{\infty}d\bar\mu_\eta(\bar\omega_\eta-\bar\mu_\eta)\bar\mu_\eta Z'\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}(\bar\omega_\eta-\bar\mu_\eta)\right]\\
&\qquad\times\left[Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z'(\bar\mu_\eta)+Z'\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z''(\bar\mu_\eta)\right]\\
&-\frac{\kappa_\eta^4}{\kappa^4}\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\frac{1}{3N_\sigma}\sum_{x<x_{\max}}\frac{1}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^{\infty}d\bar\mu_\eta(\bar\omega_\eta-\bar\mu_\eta)\bar\mu_\eta Z'(\bar\omega_\eta-\bar\mu_\eta)\\
&\qquad\times\left[Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z'(\bar\mu_\eta)+Z'\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z''(\bar\mu_\eta)\right]\\
&-\mathrm{Re}\left[\frac{\kappa_\sigma^2\kappa_\eta^2}{\kappa^4}\frac{1}{3N_\sigma}\sum_{x<x_{\max}}\frac{1}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^{\infty}d\bar\mu_\sigma(\bar\omega_\sigma+\bar\mu_\sigma)(\bar\omega_\eta+\bar\mu_\eta)Z(\bar\omega_\sigma+\bar\mu_\sigma)Z(\bar\omega_\eta+\bar\mu_\eta)Z''(\bar\mu_\sigma)\right]\\
&-\mathrm{Re}\left[\frac{\kappa_\eta^4}{\kappa^4}\frac{1}{3N_\sigma}\sum_{x<x_{\max}}\frac{1}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^{\infty}d\bar\mu_\eta(\bar\omega_\sigma+\bar\mu_\sigma)(\bar\omega_\eta+\bar\mu_\eta)Z(\bar\omega_\sigma+\bar\mu_\sigma)Z(\bar\omega_\eta+\bar\mu_\eta)Z''(\bar\mu_\eta)\right]
\end{aligned}$$

$$(\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \quad \text{(C13)}$$

At very high frequencies ($\eta/x_{\max}>1$), Eq. (C13) and the real part of (C11) can be further evaluated by exploiting the asymptotic formula

$$Z'(y\to\infty)\approx-\frac{1}{y}-\frac{1}{y^3}-\cdots\ . \quad \text{(C14)}$$

One readily obtains

$$I'_\sigma(\omega\to\infty)=\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\left[x_{\max}-\frac{\pi}{2}\right], \quad \text{(C15)}$$

$$J'_\sigma(\omega\to\infty)=O(x_{\max}/\eta^2)\ \text{at most}\ (\sigma,\eta=A,B:\ \eta\neq\sigma)\ . \quad \text{(C16)}$$

The result (C16) follows from the fact that the ω-independent amount $(\gamma n/3n_\sigma)(\kappa_\eta/\kappa)^2$ contributed by the first right-hand-side group in (C13) is exactly compensated by the $-(\gamma n/3n_\sigma)(\kappa_\eta/\kappa)^2$ amount contributed by the last four triple Z-cluster groups. Taking $x_{\max}\sim 1/\gamma$, we then have

$$w'_{\sigma 0}(0,\omega\to\infty)=\left[\frac{\Omega_\sigma}{\omega}\right]^2\left[\frac{\kappa_\eta}{\kappa}\right]^2\frac{2n}{3\pi n_\sigma}\left[1-\frac{\pi\gamma}{2}\right]+O\left[\frac{\Omega_\sigma^2\omega_0^2}{\gamma\omega^4}\right]\ (\sigma,\eta=A,B:\ \eta\neq\sigma) \quad \text{(C17)}$$

whence from (46a),

$$\alpha'_\sigma(0,\omega\to\infty)\,|_{\substack{\text{VAA}\\ \gamma\ll 1}}=-\frac{\Omega_\sigma^2}{\omega^2}+\frac{\Omega_\sigma^2\Omega_\eta^2}{\omega^4}\left[1-\frac{e_\sigma m_\eta}{e_\eta m_\sigma}\right]\frac{4\pi\beta n e_\sigma e_\eta}{\kappa^2}\frac{2\gamma}{3\pi}\left[x_{\max}-\frac{\pi}{2}\right]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{C18}$$

Consequently, our internally consistent result (C18) is tantamount to the exact third-frequency-moment-sum-rule coefficient

$$\Omega_\sigma^{(4)}(\mathbf{k}=0)\,|_{\gamma\ll 1}=(\Omega_\sigma\Omega)^2-(\Omega_\sigma\Omega_\eta)^2\left[1-\frac{e_\sigma m_\eta}{e_\eta m_\sigma}\right]\frac{4\pi\beta n e_\sigma e_\eta}{\kappa^2}\frac{2\gamma}{3\pi}\left[x_{\max}-\frac{\pi}{2}\right]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{C19}$$

As to the imaginary parts of $I_\sigma(\omega\to\infty)$ and $J_\sigma(\omega\to\infty)$, they have the expected odd-frequency parity and tend to zero quite rapidly, e.g.,

$$I''_\sigma\left[\frac{\eta}{x_{\max}}>1\right]\approx-\left[\frac{2}{\pi}\right]^{1/2}\frac{n}{3n_\sigma}\frac{\Omega_\sigma}{\omega_0}\frac{\kappa_\eta^2}{\kappa\kappa_\sigma}\left[\frac{m_\sigma+m_\eta}{m_\eta}\right]^{1/2}\left[\frac{x_{\max}}{\eta}\right]\exp\left[-\frac{\eta^2}{2x_{\max}^2}\right]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\,. \tag{C20}$$

The $(\eta/x_{\max})\ll 1$ frequency domain is of interest especially from the point of view of collective-mode behavior. In this domain, replacement of the lower integration limit in (C11) with zero readily gives

$$I'_\sigma(\omega)=\frac{\gamma}{6}\frac{n}{n_\sigma}\left[\frac{\kappa_\eta}{\kappa}\right]^2\left\{1+\sqrt{2\pi}\eta\left[1-e^{\eta^2/2}\text{erfc}\left[\frac{\eta}{\sqrt{2}}\right]\right]+\left[\frac{\pi}{2}\right]^{1/2}\eta^3e^{\eta^2/2}\text{erfc}\left[\frac{\eta}{\sqrt{2}}\right]-\sqrt{2}\eta^4\right\}>0\,, \tag{C21}$$

$$\begin{aligned}I''_\sigma(\omega)=-\frac{\gamma}{\sqrt{2\pi}}\frac{n}{n_\sigma}\frac{\omega}{\Omega_\sigma}\frac{\kappa_\sigma\kappa_\eta^2}{3\kappa^3}\left[\frac{m_\eta}{m_\sigma+m_\eta}\right]^{1/2}&\left[2\ln\left[\frac{x_{\max}}{\eta}\right]-0.884\right.\\&\left.+\left[\frac{\kappa_\sigma^2}{3\kappa^2}\frac{m_\eta}{m_\sigma+m_\eta}-1\right]e^{\eta^2/2}E_1\left[\frac{\eta^2}{2}\right]\right]\quad(\sigma,\eta=A,B:\ \eta\neq\sigma)\end{aligned} \tag{C22}$$

where

$$E_n\left[\frac{\eta^2}{2}\right]=\int_1^\infty dt\frac{\exp(-t\eta^2/2)}{t^n}\quad(n=0,1,\ldots)$$

is the exponential integral. The $O(\gamma\ln(x_{\max}/\eta))=O(\gamma\ln(\gamma^{-1}/\eta))$ term in (C22) dominates all other terms in that equation. We have, moreover, numerically demonstrated that the imaginary part of $J_\sigma(\omega)$,

$$\begin{aligned}J''_\sigma(\omega)=&-\frac{\gamma}{24\sqrt{\pi}}\frac{n}{n_\sigma}\frac{\omega}{\Omega_\sigma}\left\{\frac{\kappa_\eta^2\kappa_\sigma^3}{\kappa^5}\left[\frac{m_\eta}{m_\sigma+m_\eta}\right]^{3/2}e^{\eta^2/2}\left[E_0\left[\frac{\eta^2}{2}\right]-E_1\left[\frac{\eta^2}{2}\right]\right]+\frac{\kappa_\eta^2\kappa_\sigma^5}{\kappa^7}e^{a^2/2}\left[E_0\left[\frac{a^2}{2}\right]-E_1\left[\frac{a^2}{2}\right]\right]\right.\\&\left.+\frac{\kappa_\eta^7}{\kappa^7}\frac{\Omega_\sigma^3}{\Omega_\eta^3}e^{b^2/2}\left[E_0\left[\frac{b^2}{2}\right]-E_1\left[\frac{b^2}{2}\right]\right]\right\}\\&-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\frac{m_\sigma}{m_\eta}\frac{\kappa_\eta^2\kappa_\sigma^2}{\kappa^4}\int_0^\infty dx\frac{x^2}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^\infty d\bar\mu_\eta(\bar\omega_\eta-\bar\mu_\eta)\bar\mu_\eta Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}(\bar\omega_\eta-\bar\mu_\eta)\right]\\&\times\left[Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z'(\bar\mu_\eta)+Z'\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z''(\bar\mu_\eta)\right]\\&-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\frac{\kappa_\eta^4}{\kappa^4}\int_0^\infty dx\frac{x^2}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^\infty d\bar\mu_\eta(\bar\omega_\eta-\bar\mu_\eta)\bar\mu_\eta Z''(\bar\omega_\eta-\bar\mu_\eta)\\&\times\left[Z''\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z'(\bar\mu_\eta)+Z'\left[\left[\frac{m_\sigma}{m_\eta}\right]^{1/2}\bar\mu_\eta\right]Z''(\bar\mu_\eta)\right]\\&-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\frac{\kappa_\eta^2\kappa_\sigma^2}{\kappa^4}\int_0^\infty dx\frac{x^2}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^\infty d\bar\mu_\sigma(\bar\omega_\sigma+\bar\mu_\sigma)(\bar\omega_\eta+\bar\mu_\eta)Z''(\bar\mu_\sigma)\\&\times[Z'(\bar\omega_\sigma+\bar\mu_\sigma)Z''(\bar\omega_\eta+\bar\mu_\eta)+Z''(\bar\omega_\sigma+\bar\mu_\sigma)Z'(\bar\omega_\eta+\bar\mu_\eta)]\end{aligned}$$

$$-\frac{2\gamma}{3\pi}\frac{n}{n_\sigma}\frac{\kappa_\eta^4}{\kappa^4}\int_0^\infty dx\frac{x^2}{(1+x^2)^2}\frac{1}{\pi}\int_{-\infty}^{\infty}d\bar{\mu}_\eta(\bar{\omega}_\sigma+\bar{\mu}_\sigma)(\bar{\omega}_\eta+\bar{\mu}_\eta)Z''(\bar{\mu}_\eta)$$
$$\times[Z'(\bar{\omega}_\sigma+\bar{\mu}_\sigma)Z''(\bar{\omega}_\eta+\bar{\mu}_\eta)+Z''(\bar{\omega}_\sigma+\bar{\mu}_\sigma)Z'(\bar{\omega}_\eta+\bar{\mu}_\eta)]$$
$$[a=\omega(\beta m_\sigma)^{1/2}/\sqrt{2}\kappa,\ \ b=\omega(\beta m_\eta)^{1/2}/\sqrt{2}\kappa]$$
$$(\sigma,\eta=A,B:\ \eta\neq\sigma) \qquad \text{(C23)}$$

is at most of order γ. It then follows that

$$w''_{\sigma 0}(0,\omega)\approx\left[\frac{\Omega_\sigma}{\omega}\right]^2 I''_\sigma(\omega)\approx-\left[\frac{2}{\pi}\right]^{1/2}\frac{n}{n_\sigma}\frac{\kappa_\sigma\kappa_\eta^2}{3\kappa^3}\left[\frac{m_\eta}{m_\sigma+m_\eta}\right]^{1/2}\frac{\Omega_\sigma}{\omega}\gamma\ln(\gamma^{-1}/\eta)\ \ (\sigma,\eta=A,B:\ \eta\neq\sigma) \qquad \text{(C24)}$$

for $(\eta/x_{\max})\ll 1$.

As to $w'_\sigma(0,\omega)$, we have numerically evaluated (C21) and (C13) at $\omega=\Omega$ for the H^+-He^{2+} mixture with $N_+=N_{2+}$. These formulas, when inputted with the parameters

$$(\kappa_+/\kappa)^2=0.2,\ \ (\kappa_{2+}/\kappa)^2=0.8,\ \ (\Omega_+/\Omega)^2=(\Omega_{2+}/\Omega)^2=0.5\ ,$$

give

$$I'_+(\Omega)\approx 0.393\gamma,\ \ J'_+(\Omega)\approx-0.144\gamma\ , \qquad \text{(C25a)}$$

$$w'_{2+}(0,\Omega)=\tfrac{1}{4}w'_+(0,\Omega)\approx\frac{0.125}{4}\gamma\ . \qquad \text{(C25b)}$$

The results (C25b) are needed for the calculation of the collective-mode frequency in Sec. VI A.

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