Journal of Statistical Physics, Vol. 29, No. 2, 1982

Dynamical Three-Point Correlations and Quadratic Response Functions in Binary Ionic Mixture Plasmas[1]

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Received January 4, 1982

Nonlinear equilibrium fluctuation-dissipation relations are established for magnetic field-free binary ionic mixtures. These relations are derived from calculations of the second order partial current density response to perturbing fields which act on type A ions or on type B ions only. Our principal result connects a *single* three-point dynamical structure function to a combination of quadratic partial density response functions. This kind of formulation makes it possible to obtain a more detailed description of three-point spectral correlations by evaluating the response functions from model-dependent kinetic equations. We carry out such an evaluation in the random phase approximation.

KEY WORDS: Current correlation functions; dynamical structure functions; partial conductivities; partial density response functions; linear and quadratic polarizabilities; dielectric response function; constitutive relations; species charge; random phase approximation.

1. INTRODUCTION

In this paper, we derive new dynamical nonlinear fluctuation-dissipation relations for binary ionic mixtures and from these relations we evaluate the

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[1] Supported in part by National Science Foundation Grant No. ECS-8107449 and by Air Force Office of Scientific Research Grant No. 76-2960.

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0022-4715/82/1000-0281$03.00/0

three-point dynamical structure functions in the random phase approximation (RPA).

Model-independent fluctuation-dissipation theorems (FDT's) link transport coefficients (electric susceptibilities, conductivities, etc.) to equilibrium n-point spectral correlation functions. Linear ($n = 2$) FDT's, established some time ago[(1)] for one- and two-component plasmas without and with magnetic fields, are by now textbook examples. Useful formulations of their dynamical nonlinear counterparts are, however, still lacking in all but the simplest kind of plasma configuration, namely, the magnetic field-free classical one-component plasma (OCP).

If one is attempting an explicit calculation of the dynamical structure function in some model-dependent approximation—and that is the stated second goal of the present paper—then what is needed is a nonlinear FDT which connects one and only one n-point ($n \geqslant 3$) structure function to its transport coefficient relatives since it is these latter which are customarily evaluated from model-dependent kinetic equations. A number of investigators have succeeded in formulating the OCP nonlinear FDT's in this way: Golden *et al.*[(2)] and Sitenko[(3)] established the FDT connecting a *single* three-point dynamical structure function to a combination of three quadratic polarizabilities [see Eq. (65) below].[5,6] Their work has only very recently been extended by Kargin,[(6)] who demonstrates, via the Sitenko formalism, how one can generate $n \geqslant 3$ nonlinear FDT tensor relations for classical OCP's under the influence of both scalar and vector potential perturbations.[7]

The nonlinear FDT established by Golden *et al.* and Sitenko has proved to be a valuable tool in OCP many-body theory. It has made possible (i) the formulation of test particle energy loss entirely in terms of

[5] Efremov's earlier formulation[(4)] is considered to be less general since it leaves out spatial dispersion.

[6] FDT relationships between higher order correlations and transport coefficients have been established by Friedman *et al.*[(5)] for systems (such as partially ionized gases and electrolyte solutions) under homogeneous and stationary perturbations. As it was explained in Ref. 2, the role of the higher-order correlations under these circumstances is either to modify the dynamics of simple collisional models or to alter the relationships between diagonal and off-diagonal (Hall components) matrix elements of the conductivity. In contrast, the model-independent theories in Refs. 2 and 3 and the present paper are restricted to a consideration of diagonal longitudinal (with respect to the wave vectors) elements and assumes that the driving perturbations are functions of space and time. The FDT relations of Refs. 2 and 3 and the present paper do, however, share one common feature with those of Friedman *et al.*: all the approaches independently establish that the longitudinal projection of the wave-vector- and frequency-independent conductivity is identically zero.

[7] The corresponding ($n = 3, 4$) static FDT's established via a well-known functional derivative procedure, are reported in Ref. 7.

polarizability functions,[8] (ii) the formulation of useful frequency moment sum rules for nonlinear response functions,[3c,6,9] (iii) the explicit k-space calculations of higher-order OCP equilibrium correlations,[10] and most importantly, (iv) the formulation of a novel dynamical theory [the Golden–Kalman–Silevitch (GKS) approximation scheme[11a,b]] of strongly coupled classical electron liquids[8]; to date, this is the only theory which is exact in the static ($\omega = 0$) limit[7] and at high frequencies ($\omega \gg \omega_p$ = plasma frequency)[11b] and which, at the same time provides a reliable description of long wavelength plasmons ($\omega \cong \omega_p$) for all values of the coupling strength up to crystallization of the OCP liquid.[11c]

Over the past several years, a great deal of attention has been directed at binary ionic mixture (BIM) plasmas, e.g., pressure-ionized classical H^+–He^{2+} particles in a neutralizing background of degenerate electrons. A considerable effort—both computer experimental and theoretical[12,13,14] —has already been expanded in analyzing the collective mode behavior of such systems. Our own program of research, which enlarges on this effort, is based on the previously described GKS formalism.[8] The formulation of a dynamical theory of BIM plasmas along the lines of Ref. 11b, however, first calls for the formulation of their dynamical quadratic ($n = 3$) FDT's. *The latter is the central task of the present paper.* The resulting new fluctuation-dissipation relations—each expressing a single three-point dynamical structure function in terms of nonlinear partial response functions —will also contribute in other fundamental ways to the foundations of plasma many body theory, e.g., they will provide useful new sum rules and the corresponding nonlinear static FDT's open the way to calculating a variety of higher order BIM equilibrium correlation functions well beyond the Debye–Hückel approximation.

Our derivation will be carried out more or less along the lines of the Ref. 2 OCP derivation except for one notable difference: rather than work in terms of physical conductivities and polarizabilities, we shall instead

[8] There are two principal building blocks in the GKS theory. The first is the so-called "velocity-average approximation" (VAA), which consists of replacing the "irreducible" part of the nonequilibrium two-particle distribution function in the first BBGKY kinetic equation by its velocity average. The formal advantage of this step is that it allows one to replace the nonequilibrium two-particle distribution function by a nonequilibrium two-point density correlation function which, in turn, can be traded for an equilibrium three-point dynamical structure function. The second building block, *the application of the nonlinear FDT*, introduces quadratic response functions as the basic objects whose approximation is required. There seems to be a more natural way, especially at the dynamical level, to generate approximate structures for these quadratic response functions [see, e.g., the RPA expression Eq. (81)] than for any other quantity that might be a candidate for occupying a central place in the theory.

work in terms of the partial response function formalism suggested by Vashishta *et al.*[15] and by Tosi *et al.*[16] and further clarified by Kalman.[17] Indeed, it is Kalman's "species charge" concept[17] (described in the next paragraph) which gives credibility to the application of the partial response function formalism to normal plasma mixtures and it is this concept which we adopt in the present paper.

Partial response functions describe the response of the plasma mixture to perturbing fields which act on type-A ions or on type-B ions only. Such fields never actually occur in normal plasma mixtures, of course, but the concept of them is physically 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 with (i) its corresponding perturbing field and (ii) its companion species charges. The use of partial response functions makes it possible to formulate fluctuation-dissipation relations in such a way that each involves one and only one dynamical structure function.

The plan of the paper is as follows. In Section 2, we define linear and quadratic partial response functions and we formulate useful symmetry rules for the latter. In Section 3, current correlation and dynamical structure functions are introduced. Descriptions of the unperturbed and perturbed binary ionic mixture are given in Sections 4 and 5. Nonlinear response calculations and the subsequent derivations of new quadratic FDT's follow in Section 6; the new theorems are expressed first in terms of particle current conductivities in the time domain and then in terms of conductivity and density response functions in the frequency domain. In Section 7, we show, via the FDT's, how one can evalute the three-point dynamical structure functions in the RPA by calculating their nonlinear response function relatives from Vlassov equations; these latter results are new. Conclusions are drawn in Section 8.

2. PARTIAL RESPONSE FUNCTIONS

Consider a mixture of N_A and N_B classical point ions of like charge in a uniform neutralizing background of N_e degenerate rigid electrons; the entire system occupies the large but bounded volume Λ. Let m_σ and $Z_\sigma e$ ($\sigma = A, B$) denote the mass and electrical charge of an ion belonging to the σ species; m_e and $-e$ are the mass and charge of an electron. Microscopic number and particle current densities are given by

$$\rho_{\sigma,\mathbf{k}}(t) = \sum_{i=1}^{N_\sigma} \exp[-i\mathbf{k}\cdot\mathbf{x}_{\sigma,i}(t)] \tag{1}$$

$$\dot{\rho}_{\sigma,\mathbf{k}}(t) = -i\sum_{i=1}^{N_\sigma} \mathbf{k}\cdot\mathbf{v}_{\sigma,i}(t)\exp[-i\mathbf{k}\cdot\mathbf{x}_{\sigma,i}(t)] \qquad (\sigma = A, B) \tag{2}$$

with Fourier transforms

$$\rho_{\sigma\mathbf{k}\omega} = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \rho_{\sigma,\mathbf{k}}(t) \tag{3}$$

$$\dot{\rho}_{\sigma\mathbf{k}\omega} = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \dot{\rho}_{\sigma,\mathbf{k}}(t) = -i\omega\rho_{\sigma,\mathbf{k}}\omega \tag{4}$$

$\mathbf{x}_{\sigma,i}$ and $\mathbf{v}_{\sigma,i}$ are the position and velocity of the ith ion of species σ. As to the uniform background, $\rho_{e,\mathbf{k}} = N_e \delta_{\mathbf{k}}$.

We shall suppose that each particle, in addition to its actual electrical charge $Z_\sigma e$, is endowed with a weak fictitious "species charge" $X_\sigma e$ ($X_\sigma \ll Z_\sigma$) which can, by definition, interact only with (i) its corresponding electric field perturbation $\hat{E}^\sigma(\mathbf{k},t) = -ik\hat{\phi}^\sigma(\mathbf{k}t)$ and (ii) its companion species charges. From (i), the corresponding potential of the *external* force acting on the particle is

$$\hat{V}^\sigma(\mathbf{k}t) = X^\sigma e \hat{\phi}^\sigma(\mathbf{k}t) \qquad (\sigma = A, B) \tag{5}$$

A single such partial driving potential produces density excitations (to all orders in $\hat{V}^\sigma$) in each ionic species. The latter are linked to the former by linear and nonlinear wave-vector- and frequency-dependent response functions defined through the constitutive relations[9,10,(15–17)]

$$\langle \rho_{\sigma,\mathbf{k}\omega} \rangle^{(1)} = \sum_{\sigma'=A,B} \hat{\chi}_{\sigma\sigma'}(\mathbf{k}\omega)\, \hat{V}^{\sigma'}(\mathbf{k}\omega) \tag{6}$$

$$\langle \rho_{\sigma,\mathbf{k}\omega} \rangle^{(2)} = \frac{1}{2\pi\Lambda} \sum_{\sigma',\sigma''=A,B} \sum_{\mathbf{pq}} \int d\mu \int d\nu\, \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu)\, \hat{V}^{\sigma'}(\mathbf{p}\mu)\, \hat{V}^{\sigma''}(\mathbf{q}\nu) \times \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}} \delta(\omega - \mu - \nu) \tag{7}$$

Note that the density response of type-σ' ions

$$\langle \rho_{\sigma',\mathbf{k}}(t) \rangle = \langle \rho_{\sigma',\mathbf{k}}(t) \rangle^{(1)} + \langle \rho_{\sigma',\mathbf{k}}(t) \rangle^{(2)} + \cdots \tag{8}$$

enters into the expression

$$V^\sigma(kt) = \hat{V}^\sigma(\mathbf{k}t) + \sum_{\sigma'} \phi^{\sigma\sigma'}(k) \langle \rho_{\sigma'\mathbf{k}}(t) \rangle \tag{9}$$

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

$$\phi^{\sigma\sigma'}(k) = \frac{4\pi e^2}{k^2} (Z^\sigma Z^{\sigma'} + \delta^{\sigma\sigma'} X^\sigma X^{\sigma'}) \tag{10}$$

[9] The angular brackets denote an ensemble averaged quantity and $\langle \cdots \rangle^{(n)} = O[(\hat{V}^\sigma)^n]$ refers to ensemble averaging over the perturbed system for $n \geq 1$ and over the unperturbed system for $n = 0$.

[10] The significance of the super and subscripted species indices is as follows: repeated indices occurring in a product, one as a superscript the other as a subscript, underscore $\sum_{\sigma'=A,B}$-type summation. This kind of illuminating tensor notation is used throughout the paper.

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

$\hat{\eta}$ "conductivity" functions can be similarly defined through the constitutive relations

$$\langle \dot{\rho}_{\sigma,\mathbf{k}\omega} \rangle^{(1)} = \sum_{\sigma'} \hat{\eta}_{\sigma\sigma'}(\mathbf{k}\omega)\, \hat{V}^{\sigma'}(\mathbf{k}\omega) \tag{11}$$

$$\langle \dot{\rho}_{\sigma,\mathbf{k}\omega} \rangle^{(2)} = \frac{1}{2\pi\Lambda} \sum_{\sigma',\sigma''} \sum_{\mathbf{pq}} \int d\mu \int d\nu\, \hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu)\, \hat{V}^{\sigma'}(\mathbf{p}\mu)\, \hat{V}^{\sigma''}(\mathbf{q}\nu)$$

$$\times\, \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\, \delta(\omega - \mu - \nu) \tag{12}$$

whence

$$i\hat{\eta}_{\sigma\sigma'}(\mathbf{k}\omega) = \omega \hat{\chi}_{\sigma\sigma'}(\mathbf{k}\omega) \tag{13}$$

$$i\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = (\mu + \nu) \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) \tag{14}$$

in virtue of Eqs. (4), (6), (7), (11), and (12).

The above nonlinear response functions obey fundamental symmetry rules, some of which will be of use in the sequel. We list them here.

Interchange Symmetry:

$$\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = \hat{\eta}_{\sigma\sigma''\sigma'}(\mathbf{q}\nu; \mathbf{p}\mu) \tag{15}$$

Reality Condition:

$$\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = \hat{\eta}^{*}_{\sigma\sigma'\sigma''}(-\mathbf{p} - \mu; -\mathbf{q} - \nu) \tag{16}$$

Invariance under Spatial Reflection:

$$\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = \hat{\eta}_{\sigma\sigma'\sigma''}(-\mathbf{p}\mu; -\mathbf{q}\nu) \tag{17}$$

Note from (16) and (17) that

$$\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = \hat{\eta}^{*}_{\sigma\sigma'\sigma''}(\mathbf{p}, -\mu; \mathbf{q}, -\nu) \tag{18}$$

As to the $\hat{\chi}_{\sigma\sigma'\sigma''}$ response functions, we see from (14) that they satisfy the symmetry rules (15)–(18) as well.

3. CURRENT CORRELATION AND STRUCTURE FUNCTIONS

We next define relevant two- and three-point current correlation functions:

$$(N_\sigma N_{\sigma'})^{1/2} Q_{\sigma\sigma'}(\mathbf{k}t)\delta_{\mathbf{k}-\mathbf{p}} \equiv \langle \dot{\rho}_{\sigma,\mathbf{k}}(0) \dot{\rho}_{\sigma',-\mathbf{p}}(-t) \rangle^{(0)} \tag{19}$$

$$(N_\sigma N_{\sigma'} N_{\sigma''})^{1/2} Q_{\sigma\sigma'\sigma''}(\mathbf{p}t'; \mathbf{q}t'')\delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}} \equiv -i\langle \dot{\rho}_{\sigma,\mathbf{k}}(0) \dot{\rho}_{\sigma',-\mathbf{p}}(-t') \dot{\rho}_{\sigma'',-\mathbf{q}}(-t'') \rangle^{(0)} \tag{20}$$

where the (0) superscript indicates that ensemble averaging is carried out over the equilibrium sytem. The corresponding wave-vector- and frequency-dependent correlations are then

$$(N_\sigma N_{\sigma'})^{1/2} Q_{\sigma\sigma'}(\mathbf{k}\omega)\delta_{\mathbf{k}-\mathbf{p}}\delta(\omega-\mu) \equiv \frac{\omega\mu}{2\pi}\langle \rho_{\sigma,\mathbf{k}\omega}\rho^*_{\sigma,\mathbf{p}\mu}\rangle^{(0)} \tag{21}$$

$$(N_\sigma N_{\sigma'} N_{\sigma''})^{1/3} Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)\delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\delta(\omega-\mu-\nu) \equiv \frac{\omega\mu\nu}{2\pi}\langle \rho_{\sigma,\mathbf{k}\omega}\rho^*_{\sigma',\mathbf{p}\mu}\rho^*_{\sigma'',\mathbf{q}\nu}\rangle^{(0)} \tag{22}$$

Two- and three- point dynamical structure functions are similarly defined as follows:

$$\frac{1}{2\pi}\langle \rho_{\sigma,\mathbf{k}\omega}\rho^*_{\sigma',\mathbf{p}\mu}\rangle^{(0)} \equiv (N_\sigma N_{\sigma'})^{1/2} S_{\sigma\sigma'}(\mathbf{k}\omega)\delta_{\mathbf{k}-\mathbf{p}}\delta(\omega-\mu) + 2\pi N_\sigma N_{\sigma'}\delta_{\mathbf{k}}\delta_{\mathbf{p}}\delta(\omega)\delta(\mu) \tag{23}$$

$$\begin{aligned}\frac{1}{2\pi}\langle \rho_{\sigma,\mathbf{k}\omega}\rho^*_{\sigma',\mathbf{p}\mu}\rho^*_{\sigma'',\mathbf{q}\nu}\rangle^{(0)} \equiv{}& (N_\sigma N_{\sigma'} N_{\sigma''})^{1/3} S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)\delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\delta(\omega-\mu-\nu)\\ &+ 2\pi N_\sigma (N_{\sigma'}N_{\sigma''})^{1/2}\delta_{\mathbf{k}}\delta_{\mathbf{p}+\mathbf{q}}\delta(\omega)\delta(\mu+\nu)S_{\sigma'\sigma''}(\mathbf{p}\mu)\\ &+ 2\pi N_{\sigma'} (N_{\sigma''}N_{\sigma})^{1/2}\delta_{\mathbf{p}}\delta_{\mathbf{k}-\mathbf{q}}\delta(\mu)\delta(\omega-\nu)S_{\sigma''\sigma}(q\nu)\\ &+ 2\pi N_{\sigma''} (N_{\sigma}N_{\sigma'})^{1/2}\delta_{\mathbf{q}}\delta_{\mathbf{k}-\mathbf{p}}\delta(\nu)\delta(\omega-\mu)S_{\sigma\sigma'}(\mathbf{k}\omega)\\ &+ (2\pi)^2 N_\sigma N_{\sigma'} N_{\sigma''}\delta_{\mathbf{p}}\delta_{\mathbf{q}}\delta_{\mathbf{k}}\delta(\mu)\delta(\nu)\delta(\omega)\end{aligned} \tag{24}$$

Then from (21) and (23),

$$Q_{\sigma\sigma'}(\mathbf{k}\omega) = \omega^2 S_{\sigma\sigma'}(\mathbf{k}\omega) \tag{25}$$

and from (22) and (24),

$$Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) = \mu\nu(\mu+\nu)S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \tag{26}$$

The three-point Q and S functions are, by definition, invariant with respect to rotation on the triangle formed by the four vectors $(\mathbf{p}\mu)$, $(\mathbf{q}\nu)$, $(\mathbf{k}=\mathbf{p}+\mathbf{q},\ \omega=\mu+\nu)$, i.e.,

$$Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) = Q_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) = Q_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega) \tag{27}$$

and

$$S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) = S_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) = S_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega) \tag{28}$$

To verify that these three-point functions are real, we observe that the ternary correlation appearing on the left-hand side of (24) undergoes no change in sign either under microscopic time reversal ($\mu \to -\mu$, $\nu \to -\nu$, $\omega \to -\omega$) or under space inversion ($\mathbf{p} \to -\mathbf{p}$, $\mathbf{q} \to -\mathbf{q}$, $\mathbf{k} \to -\mathbf{k}$). Consequently, the correlation is real, whence from Eqs. (22) and (26), $Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)$ and $S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)$ must also be *real*.

Finally, note that (23) and (24) readily generate corresponding relations which define the well-known two- and three-point static structure functions

$$S_{\sigma\sigma'}(\mathbf{k}) = \int \frac{d\omega}{2\pi} S_{\sigma\sigma'}(\mathbf{k}\omega) \tag{29}$$

$$S_{\sigma\sigma'\sigma''}(\mathbf{pq}) = \int \frac{d\mu}{2\pi} \int \frac{d\nu}{2\pi} S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) \tag{30}$$

in terms of *equal-time* equilibrium correlations.

4. DESCRIPTION OF THE UNPERTURBED SYSTEM

The state of the magnetic field-free unperturbed BIM in the infinite past is characterized by the macrocanonical distribution function (normality to unity):

$$\Omega^{(0)}(\Gamma) = e^{-\beta H^{(0)}(\Gamma)} \Big/ \int d\Gamma\, e^{-\beta H^{(0)}} \tag{31}$$

where

$$d\Gamma = \prod_{i=1}^{N_A} \prod_{j=1}^{N_B} d^3x_{A,i}\, d^3p_{A,i}\, d^3x_{B,j}\, d^3p_{B,j}$$

is a differential volume element in the phase space spanned by the $6(N_A + N_B)$ ion coordinates and momenta; $\mathbf{x}_{A,i}$ and $\mathbf{p}_{A,i}$ are the ith position and momentum of an ion belonging to species A, β^{-1} is the temperature in energy units, and

$$H^{(0)}(\Gamma) = \sum_{\sigma=A,B} \sum_{i=1}^{N_\sigma} \frac{p_{\sigma,i}^2}{2m_\sigma} + V^{(0)} \tag{32}$$

is the unperturbed Hamiltonian including A–A, A–B, B–B, A–background, B–background, and background–background interactions. Letting $\mathbf{y}$ and $\mathbf{z}$ denote the coordinates of the uniform neutralizing (electron) background, the potential energy is given by

$$\begin{aligned} V^{(0)} = {} & \frac{1}{2} \sum_{\sigma'=A,B} \sum_{\sigma''=A,B} \sum_{\substack{i=1 \\ i\neq j}}^{N_{\sigma'}} \sum_{j=1}^{N_{\sigma''}} \frac{(Z^{\sigma'}Z^{\sigma''} + \delta^{\sigma'\sigma''}X^{\sigma'}X^{\sigma''})e^2}{|\mathbf{x}_{\sigma',i} - \mathbf{x}_{\sigma'',j}|} \\ & - \frac{N_e}{\Lambda} \sum_{\sigma'=A,B} \sum_{i=1}^{N_{\sigma'}} \int d^3y \frac{Z^{\sigma'}e^2}{|\mathbf{x}_{\sigma',i} - \mathbf{y}|} \\ & + \frac{N_e^2}{2\Lambda^2} \int d^3y \int d^3z \frac{(1 + X^{e^2})e^2}{|\mathbf{y} - \mathbf{z}|} \end{aligned} \tag{33}$$

or equivalently,

$$V^{(0)} = \frac{1}{2\Lambda} \sum_{\sigma', \sigma'' = A, B} \sum_{\mathbf{k} \neq 0} \phi^{\sigma'\sigma''}(k) \{ \rho_{\sigma', -\mathbf{k}} \rho_{\sigma'', \mathbf{k}} - \delta_{\sigma'\sigma''} N_{\sigma'} \} + C \tag{34}$$

where $\phi^{\sigma'\sigma''}(k) = \phi_{\mathbf{k}}(Z^{\sigma'}Z^{\sigma''} + \delta^{\sigma'\sigma''}X^{\sigma'}X^{\sigma''})$ is the Fourier transform of the Coulomb energy defined earlier [see Eq. (10)]; the omission of the $\mathbf{k} = 0$ contribution takes account of the uniform background and we note that the divergent constant

$$C = -\frac{1}{2\Lambda} \sum_{\sigma' = A, B} \phi^{\sigma'\sigma'}(k = 0) N_{\sigma'} + \frac{1}{2\Lambda} \phi_{\mathbf{k}=0} \left[\sum_{\sigma' = A, B} (X^{\sigma'} N_{\sigma'})^2 + (X^e N_e)^2 \right] \tag{35}$$

is customarily left out of the collective coordinate formulation (34). In deriving (34) and (35) from (33), we have exploited the charge neutrality requirement

$$\sum_{\sigma' = A, B} Z^{\sigma'} N_{\sigma'} = N_e \tag{36}$$

As to the species charges, they also satisfy their own neutrality condition

$$\sum_{\sigma' = A, B} X^{\sigma'} N_{\sigma'} = -X^e N_e \tag{37}$$

where $X_A \ll Z_A$, $X_B \ll Z_B$, and $|X_e| \ll 1$. Thus if the BIM is driven by $\hat{\phi}^A$ and $\hat{\phi}^B$ perturbations, the uniform background is still left unaffected since its species charge cannot interact with the perturbations.

Now, it is not $H^{(0)}$ which enters directly into the calculation of the partial ion density response (inasmuch as $H^{(0)}$ is explicitly independent of the perturbing field) but rather the Hamiltonian $\hat{H}$ for the interactions between the plasma ions and the external field perturbations. We therefore turn next to the formulation of $\hat{H}$ and the ensuing calculations of the linear and nonlinear ion density responses from perturbed Liouville distribution functions.

5. DESCRIPTION OF THE PERTURBED SYSTEM

Let the time-dependent scalar potentials

$$\hat{\phi}^{\sigma}(\mathbf{r}t) = \frac{1}{\Lambda} \sum_{\mathbf{k}} \hat{\phi}^{\sigma}(\mathbf{k}t) e^{i\mathbf{k}\cdot\mathbf{r}}, \qquad \sigma = A, B \tag{38}$$

be introduced into the system. The equilibrium Hamiltonian $H^{(0)}$ and Liouville operator

$$\mathfrak{L}^{(0)} \equiv -i[H^{(0)}, \ldots] = i \sum_{\sigma = A, B} \sum_{i=1}^{N_\sigma} \left[\frac{\partial H^{(0)}}{\partial \mathbf{x}_{\sigma,i}} \cdot \frac{\partial}{\partial \mathbf{p}_{\sigma,i}} - \frac{\partial H^{(0)}}{\partial \mathbf{p}_{\sigma,i}} \cdot \frac{\partial}{\partial \mathbf{x}_{\sigma,i}} \right] \tag{39}$$

are accordingly perturbed by amounts

$$\hat{H}(t) = \sum_{\sigma=A,B} X^{\sigma} e \sum_{i=1}^{N_\sigma} \hat{\phi}^{\sigma}(\mathbf{x}_{\sigma,i}, t)$$

$$= \frac{1}{\Lambda} \sum_{\sigma} \sum_{\mathbf{k}} \hat{V}^{\sigma}(\mathbf{k}t) \rho_{\sigma,-k} \tag{40}$$

$$\hat{\mathfrak{L}}(t) = -\frac{i}{\Lambda} \sum_{\sigma} \sum_{\mathbf{k}} \hat{V}^{\sigma}(\mathbf{k}t) [\rho_{\sigma,-\mathbf{k}}, \cdots] \tag{41}$$

The subsequent perturbation of the Liouville equation

$$\frac{\partial}{\partial t} \Omega(\mathbf{\Gamma}t) + i\mathfrak{L}\Omega(\mathbf{\Gamma}t) = 0 \tag{42}$$

for the distribution function then results in the formal solution

$$\Omega(\mathbf{\Gamma}t) = \Omega^{(0)}(\mathbf{\Gamma}) + \Omega^{(1)}(\mathbf{\Gamma}t) + \Omega^{(2)}(\mathbf{\Gamma}t) + \cdots \tag{43}$$

$$\Omega^{(1)}(\mathbf{\Gamma}t) = -i \sum_{\sigma'=A,B} \int_0^{\infty} d\tau\, U(\tau) \hat{\mathfrak{L}}^{\sigma'}(t-\tau) \Omega^{(0)} \tag{44}$$

$$\Omega^{(2)}(\mathbf{\Gamma}t) = -\sum_{\sigma',\sigma''=A,B} \int_0^{\infty} d\tau \int_0^{\infty} d\tau'\, U(\tau) \hat{\mathfrak{L}}^{\sigma'}(t-\tau) U(\tau') \hat{\mathfrak{L}}^{\sigma''}(t-\tau-\tau') \Omega^{(0)} \tag{45}$$

where $U(\tau) = e^{-i\tau\mathfrak{L}^{(0)}}$ is the time evolution operator pertaining to the equilibrium system. Our derivation of the quadratic partial FDT's from (45) will follow the procedure of the Ref. 2 derivation for the OCP. For the sake of completeness and because it is instructive, we have displayed in Appendix A the key steps leading to the establishment of the by-now-classic linear FDT's (quoted, e.g., in Refs. 15 and 16).

6. NONLINEAR RESPONSE THEORY

We come now to the central task of this paper—the derivation of the nonlinear FDT relations. From Appendix A [Eq. (A1)],

$$\hat{\mathfrak{L}}^{\sigma'}(t-\tau) U(\tau') \hat{\mathfrak{L}}^{\sigma''}(t-\tau-\tau') \Omega^{(0)}$$

$$= -\frac{\beta \Omega^{(0)}}{\Lambda^2} \sum_{\mathbf{pq}} \hat{V}^{\sigma'}(\mathbf{p}t-\tau) \hat{V}^{\sigma''}(\mathbf{q}t-\tau-\tau')$$

$$\times \left\{ \beta \dot{\rho}_{\sigma',-\mathbf{p}} U(\tau') \dot{\rho}_{\sigma'',-\mathbf{q}} + \left[\rho_{\sigma',-\mathbf{p}}, U(\tau') \dot{\rho}_{\sigma'',-\mathbf{q}} \right] \right\} \tag{46}$$

Substitution of (46) into (45) then gives

$$\Omega^{(2)}(\boldsymbol{\Gamma}t) = \frac{1}{2}\sum_{\sigma'\sigma''}\left\{\overline{\Omega}(\boldsymbol{\Gamma}t;\sigma',\sigma'') + \overline{\Omega}(\boldsymbol{\Gamma}t;\sigma'',\sigma')\right\} \tag{47}$$

$$\begin{aligned}\overline{\Omega}(\boldsymbol{\Gamma}t;\sigma',\sigma'') = \frac{\beta\Omega^{(0)}}{\Lambda^2}\sum_{\mathbf{pq}}\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''\,\hat{V}^{\sigma'}(\mathbf{p}t')\hat{V}^{\sigma''}(\mathbf{q}t'')\\ \times U(t-t')\left\{\beta\dot{\rho}_{\sigma',-\mathbf{p}}U(t'-t'')\dot{\rho}_{\sigma'',-\mathbf{q}}\right.\\ \left.+\left[\rho_{\sigma',-\mathbf{p}},U(t'-t'')\dot{\rho}_{\sigma'',-\mathbf{q}}\right]\right\}\end{aligned} \tag{48}$$

where $t' = t - \tau$, $t'' = t' - \tau'$. The two times t' and t'' are physically equivalent so that a symmetry with respect to prime–double-prime interchange should prevail. To make the symmetry manifest, consider the contribution $\overline{\Omega}(\Gamma\tau;\sigma'',\sigma')$: reversal of its order of integration accompanied by the interchanges $\mathbf{p}\leftrightarrow\mathbf{q}$, $t'\leftrightarrow t''$ gives

$$\begin{aligned}\overline{\Omega}(\boldsymbol{\Gamma}t;\sigma'',\sigma') = \frac{\beta\Omega^{(0)}}{\Lambda^2}\sum_{\mathbf{pq}}\int_{-\infty}^{t}dt'\int_{t'}^{t}dt''\,\hat{V}^{\sigma'}(\mathbf{p}t')\hat{V}^{\sigma''}(\mathbf{q}t'')\\ \times U(t-t'')\left\{\beta\dot{\rho}_{\sigma'',-\mathbf{q}}U(t''-t')\dot{\rho}_{\sigma',-\mathbf{p}}\right.\\ \left.+\left[\rho_{\sigma'',-\mathbf{q}},U(t''-t')\dot{\rho}_{\sigma',-\mathbf{p}}\right]\right\}\end{aligned} \tag{49}$$

whence the manifestly interchange symmetric expression

$$\begin{aligned}\Omega^{(2)}(\Gamma t) = \frac{\beta\Omega^{(0)}}{2\Lambda^2}\sum_{\sigma',\sigma''}\sum_{\mathbf{pq}}\int_{-\infty}^{t}dt'\\ \times\int_{-\infty}^{t}dt''\,\hat{V}^{\sigma'}(\mathbf{p}t')\hat{V}^{\sigma''}(\mathbf{q}t'')I_{\sigma''}(\mathbf{pq};t't'';t)\\ = \theta(t'-t'')U(t-t')\left\{\beta\dot{\rho}_{\sigma',-\mathbf{p}}U(t'-t'')\dot{\rho}_{\sigma'',-\mathbf{q}}\right.\\ \left.+\left[\rho_{\sigma',-\mathbf{p}},U(t'-t'')\dot{\rho}_{\sigma'',-\mathbf{q}}\right]\right\}\\ +\theta(t''-t')U(t-t'')\left\{\beta\dot{\rho}_{\sigma'',-\mathbf{q}}U(t''-t')\dot{\rho}_{\sigma',-\mathbf{p}}\right.\\ \left.+\left[\rho_{\sigma'',-\mathbf{q}},U(t''-t')\dot{\rho}_{\sigma',-\mathbf{p}}\right]\right\}\end{aligned} \tag{50}$$

results from Eqs. (47)–(49) [θ is the unit step function]. As a consequence, second-order ensemble-averaged quantities, e.g., the partial current density response

$$\langle \dot{\rho}_{\sigma,\mathbf{k}} \rangle^{(2)}(t) = \int d\Gamma\, \Omega^{(2)}(\Gamma t) \dot{\rho}_{\sigma,\mathbf{k}}$$

$$= \frac{\beta}{2\Lambda^2} \sum_{\sigma'\sigma''} \sum_{\mathbf{pq}} \int_{-\infty}^{t} dt' \times \int_{-\infty}^{t} dt'' \, \hat{V}^{\sigma'}(\mathbf{p}t') \hat{V}^{\sigma''}(\mathbf{q}t'') \langle \dot{\rho}_{\sigma,\mathbf{k}} I_{\sigma'\sigma''}(\mathbf{pq}; t't''; t) \rangle^{(0)} \quad (51)$$

are now guaranteed to exhibit the symmetry

$$\langle \dot{\rho}_{\sigma,\mathbf{k}} I_{\sigma'\sigma''}(\mathbf{pq}; t't''; t) \rangle^{(0)} = \langle \dot{\rho}_{\sigma,\mathbf{k}} I_{\sigma''\sigma'}(\mathbf{qp}; t''t'; t) \rangle^{(0)} \quad (52)$$

required for the derivation of the nonlinear FDT in the sequel.

Continuing the calculation of (51) according to the procedure of Appendix A, the passage from Eulerian to Lagrangian representation is effected by letting the time evolution operators act on $\dot{\rho}_{\sigma',-\mathbf{p}}$ and $\dot{\rho}_{\sigma'',-\mathbf{q}}$. One obtains

$$\begin{aligned}\langle \dot{\rho}_{\sigma,\mathbf{k}}(t) \rangle^{(2)} = \frac{\beta}{2\Lambda^2} \sum_{\sigma',\sigma''} \sum_{\mathbf{pq}} \int_0^{\infty} d\tau' \int_0^{\infty} d\tau'' \, \hat{V}^{\sigma'}(\mathbf{p}t - \tau') \hat{V}^{\sigma''}(\mathbf{q}t - \tau'') \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}} \\ \times \Big\{ \beta \langle \dot{\rho}_{\sigma,\mathbf{k}}(0) \dot{\rho}_{\sigma',-\mathbf{p}}(-\tau') \dot{\rho}_{\sigma'',-\mathbf{q}}(-\tau'') \rangle^{(0)} \\ + \theta(\tau'' - \tau') \langle \dot{\rho}_{\sigma,\mathbf{k}}(0) \big[\rho_{\sigma',-\mathbf{p}}(-\tau'), \dot{\rho}_{\sigma'',-\mathbf{q}}(-\tau'') \big] \rangle^{(0)} \\ + \theta(\tau' - \tau'') \langle \dot{\rho}_{\sigma,\mathbf{k}}(0) \big[\rho_{\sigma'',-\mathbf{q}}(-\tau''), \dot{\rho}_{\sigma',-\mathbf{p}}(-\tau') \big] \rangle^{(0)} \Big\}\end{aligned} \quad (53)$$

where $\tau' = t - t'$, $\tau'' = t - t''$.

Dynamical Fluctuation-Dissipation Relations: Time and Frequency Domain Formulations

The average second-order partial current density response is connected to the external driving potentials through quadratic "conductivities" defined by the constitutive relation [see Eq. (12)]:

$$\langle \dot{\rho}_{\sigma,\mathbf{k}}(t) \rangle^{(2)} = \frac{1}{\Lambda} \sum_{\sigma',\sigma''} \sum_{\mathbf{pq}} \int_{-\infty}^{\infty} d\tau' \times \int_{-\infty}^{\infty} d\tau'' \, \hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\tau'; \mathbf{q}\tau'') \hat{V}^{\sigma'}(\mathbf{p}t - \tau') \hat{V}^{\sigma''}(\mathbf{q}t - \tau'') \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}} \quad (54)$$

The quadratic FDT

$$\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\tau';\mathbf{q}\tau'') = \frac{\beta}{2\Lambda}\theta(\tau')\theta(\tau'')$$
$$\times\Big\{\beta\langle\dot{\rho}_{\sigma,\mathbf{k}}(0)\dot{\rho}_{\sigma',-\mathbf{p}}(-\tau')\dot{\rho}_{\sigma'',-\mathbf{q}}(-\tau'')\rangle^{(0)}$$
$$+\theta(\tau''-\tau')\langle\dot{\rho}_{\sigma,\mathbf{k}}(0)\big[\rho_{\sigma',-\mathbf{p}}(-\tau'),\dot{\rho}_{\sigma'',-\mathbf{q}}(-\tau'')\big]\rangle^{(0)}$$
$$+\theta(\tau'-\tau'')\rangle\dot{\rho}_{\sigma,\mathbf{k}}(0)\big[\rho_{\sigma'',-\mathbf{q}}(-\tau''),\dot{\rho}_{\sigma',-\mathbf{p}}(-\tau')\big]\rangle^{(0)}\Big\}$$
$$(\mathbf{k}=\mathbf{p}+\mathbf{q}) \quad (55)$$

then immediately follows from Eqs. (53) and (54). Thanks to the above manipulations, the right-hand side of (55) is manifestly symmetric under $\mathbf{p}\leftrightarrow\mathbf{q}$ and prime–double-prime interchange; so is the left-hand side [were it not, only the symmetric projections would enter in (54), which could then be redefined as $\hat{\eta}_{\sigma\sigma'\sigma''}$].

The more useful time domain expression

$$\hat{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\tau';\mathbf{q}\tau'')$$
$$\equiv \hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\tau';\mathbf{q}\tau'') - \hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}\tau''-\tau';-\mathbf{k}\tau'')\theta(\tau')$$
$$-\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\tau';\mathbf{q}\tau'-\tau'')\theta(\tau'')$$
$$=\frac{\beta^2}{2\Lambda}\theta(\tau')\theta(\tau'')\langle\dot{\rho}_{\sigma,\mathbf{k}}(0)\dot{\rho}_{\sigma',-\mathbf{p}}(-\tau')\dot{\rho}_{\sigma'',-\mathbf{q}}(-\tau'')\rangle^{(0)} \qquad (\mathbf{k}=\mathbf{p}+\mathbf{q}) \quad (56)$$

which instead relates a single dynamical correlation to three nonlinear conductivities, can be defined from (55) according to the procedure of Ref. 2. The corresponding frequency domain FDT

$$\mathrm{Im}\{\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) + \hat{\eta}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) + \hat{\eta}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)\}$$
$$=\frac{\beta^2}{4}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)$$
$$(n_\sigma = N_\sigma/\Lambda) \quad (\mathbf{k}=\mathbf{p}+\mathbf{q},\ \omega=\mu+\nu) \quad (57)$$

then follows from (56) (see Appendix B for details). Finally, the conductivities and current correlation function can be eliminated in favor of the more popular partial density response and structure functions [cf. Eqs. (14) and (26)]. The dynamical FDT

$$\mathrm{Re}\left\{\frac{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)}{\mu\nu} - \frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu)}{\mu\omega} - \frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)}{\omega\nu}\right\}$$
$$=-\frac{\beta^2}{4}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \qquad (\mathbf{k}=\mathbf{p}+\mathbf{q},\ \omega=\mu+\nu) \quad (58)$$

which results is the central statement of the present paper. There are eight such relations ($\sigma,\sigma',\sigma''=A,B$).

It is now a simple matter to derive the nonlinear FDT for the spectral correlation of *combined* microscopic charge densities,[11]

$$2P(\mathbf{p}\mu;\mathbf{q}\nu) = \sum_{\substack{\sigma,\sigma',\sigma''\\=A,B}} Z_\sigma Z^{\sigma'} Z^{\sigma''} e^3 (n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \tag{59}$$

[cf. Eqs. (23) and (24)]. First, observe from (58) and (59) that

$$\operatorname{Re} \sum_{\sigma,\sigma',\sigma''} Z_\sigma Z^{\sigma'} Z^{\sigma''} e^3 \times \left\{ \frac{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)}{\mu\nu} - \frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu)}{\mu\omega} - \frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)}{\omega\nu} \right\} = -\frac{\beta^2}{2} P(\mathbf{p}\mu;\mathbf{q}\nu) \tag{60}$$

We next express the left-hand side of (60) in terms of external polarizabilities, $\hat{\alpha}$'s, defined by the constitutive relation

$$\begin{aligned}\langle q_{\mathbf{k}\omega}\rangle^{(2)} &= \sum_{\sigma=A,B} Z_\sigma e \langle \rho_{\sigma,\mathbf{k}\omega}\rangle^{(2)} \\ &= \frac{i}{8\pi^2\Lambda} \sum_{\mathbf{pq}} pqk \int d\mu \int d\nu\, \hat{\alpha}(\mathbf{p}\mu;\mathbf{q}\nu)\hat{\phi}(\mathbf{p}\mu)\hat{\phi}(\mathbf{q}\nu)\delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\delta(\omega-\mu-\nu)\end{aligned} \tag{61}$$

On the other hand, we have from (7) that

$$\langle q_{\mathbf{k}\omega}\rangle^{(2)} = \frac{1}{2\pi\Lambda} \sum_{\sigma,\sigma',\sigma''} \sum_{\mathbf{pq}} \int d\mu \int d\nu\, \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) Z_\sigma X^{\sigma'} X^{\sigma''} e^3 \hat{\phi}^{\sigma'}(\mathbf{p}\mu)\hat{\phi}^{\sigma''}(\mathbf{q}\nu) \times \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\delta(\omega-\mu-\nu) \tag{62}$$

In order to reconcile Eq. (61) with (62), we must now entirely discard the notion of partial perturbing fields in favor of a field $\hat{\phi}$ which simultaneously drives both species [see Eq. (61)]. This eliminates at the same time any further need to make a distinction between "species charge" and actual charge. To summarize: the stipulation that $\hat{\phi}^{\sigma'} = \hat{\phi}^{\sigma''} = \hat{\phi}$ is tantamount to identifying $X^\sigma e\hat{\phi}^\sigma$ as being $Z^\sigma e\hat{\phi}$, whence

$$\langle q_{\mathbf{k}\omega}\rangle^{(2)} = \frac{1}{2\pi\Lambda} \sum_{\sigma,\sigma',\sigma''} \sum_{\mathbf{pq}} \int d\mu \int d\nu\, \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) Z_\sigma Z^{\sigma'} Z^{\sigma''} e^3 \hat{\phi}(\mathbf{p}\mu)\hat{\phi}(\mathbf{q}\nu) \times \delta_{\mathbf{k}-\mathbf{p}-\mathbf{q}}\delta(\omega-\mu-\nu) \tag{63}$$

Comparison of (61) and (63) then gives for the external quadratic polarizability

$$\hat{\alpha}(\mathbf{p}\mu;\mathbf{q}\nu) = \frac{-4\pi i}{pqk} \sum_{\sigma,\sigma',\sigma''} Z_\sigma Z^{\sigma'} Z^{\sigma''} e^3 \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \tag{64}$$

[11] The factor 2 has been introduced into (59) to facilitate comparison with the Ref. 2 OCP result.

leading, in turn, to the *total* FDT

$$\operatorname{Im}\left\{\frac{\hat{\alpha}(\mathbf{p}\mu;\mathbf{q}\nu)}{\mu\nu}-\frac{\hat{\alpha}(-\mathbf{k}-\omega;\mathbf{p}\mu)}{\mu\omega}-\frac{\hat{\alpha}(\mathbf{q}\nu;-\mathbf{k}-\omega)}{\omega\nu}\right\}=\frac{2\pi\beta^2}{pqk}P(\mathbf{p}\mu;\mathbf{q}\nu) \tag{65}$$

Equation (65) is identical to its OCP counterpart [Ref. 2: Eq. (66)], as it should be.

We conclude this section now with a brief derivation of the *static* partial FDT relation from (58). Since $S_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)$ is expected to be nonsingular, the $\mu=0$, $\nu=0$, $\omega=0$ singularities in (58) are spurious and the nonlinear FDT remains unchanged if one stipulates that each frequency denominator in (58) is a double principal value denominator. With this understanding, integrations over μ and ν provide

$$\begin{aligned}&\mathrm{PP}\int\frac{d\mu}{2\pi}\int\frac{d\nu}{2\pi}\\&\qquad\times\operatorname{Re}\left\{\frac{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)}{\mu\nu}-\frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu)}{\mu\omega}-\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)}{\omega\nu}\right\}\\&=-\frac{\beta^2}{4}(n_\sigma n_{\sigma'}n_{\sigma''})^{1/3}S_{\sigma\sigma'\sigma''}(\mathbf{pq}),\end{aligned} \tag{66}$$

where $S_{\sigma\sigma'\sigma''}(\mathbf{pq})$ is defined by Eq. (30). One then finds that

$$\mathrm{PP}\int\frac{d\mu}{2\pi}\int\frac{d\nu}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)}{\mu\nu}=-\frac{1}{4}\operatorname{Re}\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}0;\mathbf{q}0) \tag{67}$$

$$\begin{aligned}&\mathrm{PP}\int\frac{d\mu}{2\pi}\int\frac{d\nu}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu)}{\mu\omega}\\&\qquad=-\mathrm{PP}\int\frac{d\mu}{2\pi}\int\frac{d\bar{\nu}}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}\bar{\nu}-\mu;\mathbf{p}\mu)}{\mu(\bar{\nu}-\mu)}\\&\qquad=\frac{1}{4}\operatorname{Re}\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}0;\mathbf{p}0)\end{aligned} \tag{68}$$

$$\begin{aligned}&\mathrm{PP}\int\frac{d\mu}{2\pi}\int\frac{d\nu}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)}{\omega\nu}\\&=-\mathrm{PP}\int\frac{d\bar{\mu}}{2\pi}\int\frac{d\nu}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}\bar{\mu}-\nu)}{\nu(\bar{\mu}-\nu)}\\&=-\mathrm{PP}\int\frac{d\nu}{2\pi}\int\frac{d\bar{\mu}}{2\pi}\operatorname{Re}\frac{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}\bar{\mu}-\nu)}{\nu(\bar{\mu}-\nu)}-\frac{1}{4}\operatorname{Re}\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}0;-\mathbf{k}0)=0\end{aligned} \tag{69}$$

in virtue of the double Hilbert transform formula

$$\hat{\chi}(\mu\nu) = -\frac{1}{\pi^2}\,\mathrm{PP}\int\frac{d\mu'}{\mu-\mu'}\int\frac{d\nu'}{\nu-\nu'}\,\hat{\chi}(\mu'\nu')$$

satisfied by the $\hat{\chi}$'s. We note that the evaluation of the third left-hand side member of (66) [Eq. (69)] relies on the Poincaré–Bertrand theorem[18] cited in Appendix B. Substitution of (67) to (69) into (66) readily yields

$$\mathrm{Re}\{\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}0;\mathbf{q}0) + \hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}0;\mathbf{p}0)\} = \beta^2(n_\sigma n_{\sigma'} n_{\sigma''})^{1/2} S_{\sigma\sigma'\sigma''}(\mathbf{pq}) \tag{70}$$

But Eq. (70), in turn, generates the FDT relations

$$\mathrm{Re}\{\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}0;\mathbf{p}0) + \hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}0;-\mathbf{k}0)\} = \beta^2(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma''\sigma\sigma'}(-\mathbf{kp}) \tag{71}$$

$$\mathrm{Re}\{\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}0;-\mathbf{k}0) + \hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}0;\mathbf{q}0)\} = \beta^2(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma'\sigma''\sigma}(\mathbf{q},-\mathbf{k}) \tag{72}$$

whence

$$\mathrm{Re}\,\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}0;\mathbf{q}0) = \mathrm{Re}\,\hat{\chi}_{\sigma''\sigma\sigma'}(-\mathbf{k}0;\mathbf{p}0) = \mathrm{Re}\,\hat{\chi}_{\sigma'\sigma''\sigma}(\mathbf{q}0;-\mathbf{k}0) \tag{73}$$

in virtue of the triangle symmetry rule (28) obeyed by the S's. The desired static FDT result

$$\mathrm{Re}\,\hat{\chi}_{\sigma\sigma'\sigma''}(\mathbf{p}0;\mathbf{q}0) = \tfrac{1}{2}\,\beta^2(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma\sigma'\sigma''}(\mathbf{pq}) \tag{74}$$

follows from (70) and (73). Confidence in its dynamical counterpart (58) is further enhanced by the fact that Eq. (74) exactly reproduces the static FDT's (independently formulated from a functional derivative procedure and) quoted in Refs. 17 and 19.

7. NONLINEAR RESPONSE IN THE RPA

This paper has as its second objective the explicit calculation of the dynamical three-point structure function. Thanks to the new FDT relation (58), this can be accomplished by calculating the nonlinear partial $\hat{\chi}$'s from model-dependent plasma kinetic equations. For binary ionic mixtures in the intermediate and strong coupling regimes, such a task is indeed formidable and is, in any case, well beyond the scope of the present paper. Rather, we shall confine ourselves to calculations made in the RPA. This is, of course, the appropriate first step: it paves the way for future perturbation theoretic calculations of dynamical three-point structure functions in weakly coupled systems.

The introduction of driving potentials $\hat{\phi}^{\sigma}$ $(\sigma = A, B)$ into a mixture of noninteracting ions results in the following Vlassov expressions for the first- and second-order (in $\hat{\phi}^{\sigma}$) one-particle distribution functions

$$F^{(1)}_{\sigma,\mathbf{k}\omega}(\mathbf{v}) = -\frac{1}{m_\sigma}\frac{\mathbf{k}\cdot\partial F^{(0)}_\sigma(v)/\partial\mathbf{v}}{\omega-\mathbf{k}\cdot\mathbf{v}}\sum_{s'}\left[\delta_{\sigma s'}\hat{V}^{s'}(\mathbf{k}\omega) + \phi^{s'}_\sigma(k)\langle\rho_{s',\mathbf{k}\omega}\rangle^{(1)}\right]$$

$$= -\frac{1}{m_\sigma}\frac{\mathbf{k}\cdot\partial F^{(0)}_\sigma(v)/\partial\mathbf{v}}{\omega-\mathbf{k}\cdot\mathbf{v}}\sum_{\sigma',s'}\left[\delta_{\sigma s'}\delta^{s'}_{\sigma'} + \phi^{s'}_\sigma(k)\hat{\chi}^{\mathrm{RPA}}_{s'\sigma'}(\mathbf{k}\omega)\right]\hat{V}^{\sigma'}(\mathbf{k}\omega)$$

(75a, b)

$$F^{(2)}_{\sigma,\mathbf{k}\omega}(v) = -\frac{1}{m_\sigma}\frac{\mathbf{k}\cdot\partial F^{(0)}_\sigma(v)/\partial\mathbf{v}}{\omega-\mathbf{k}\cdot\mathbf{v}}\sum_{s'}\phi^{s'}_\sigma(k)\langle\rho_{s',\mathbf{k}\omega}\rangle^{(2)}$$

$$-\frac{\beta}{m_\sigma}\frac{1}{\Lambda}\sum_{\substack{\sigma',\sigma''\\ s',s''}}\sum_{\mathbf{p}}\int\frac{d\mu}{2\pi}\frac{1}{\omega-\mathbf{k}\cdot\mathbf{v}}\mathbf{q}\cdot\frac{\partial}{\partial\mathbf{v}}\frac{\mathbf{p}\cdot\mathbf{v}F^{(0)}_\sigma(v)}{\mu-\mathbf{p}\cdot\mathbf{v}}$$

$$\times\left[\delta_{\sigma s'}\delta^{s'}_{\sigma'} + \phi^{s'}_\sigma(p)\hat{\chi}^{\mathrm{RPA}}_{s'\sigma'}(\mathbf{p}\mu)\right]$$

$$\times\left[\delta_{\sigma s''}\delta^{s''}_{\sigma''} + \phi^{s''}_\sigma(q)\hat{\chi}^{\mathrm{RPA}}_{s''\sigma''}(\mathbf{q}\nu)\right]\hat{V}^{\sigma'}(\mathbf{p}\mu)\hat{V}^{\sigma''}(\mathbf{q}\nu)$$

$$(\mathbf{k} = \mathbf{p} + \mathbf{q},\ \omega = \mu + \nu) \quad (76)$$

where

$$\langle\rho_{s',\mathbf{k}\omega}\rangle = \int d^3v\, F_{s',\mathbf{k}\omega}(\mathbf{v}) \quad (77)$$

$$\phi^{s'}_\sigma(k) = \phi_{\mathbf{k}}\left(Z^{s'}Z_\sigma + \delta^{s'}_\sigma X^2_\sigma\right) \quad (78)$$

and

$$F^{(0)}_\sigma(v) = n_\sigma\left(\frac{\beta m_\sigma}{2\pi}\right)^{3/2}\exp\left(\frac{-\beta m_\sigma v^2}{2}\right) \quad (79)$$

The derivation of the RPA $\hat{\chi}$ density response functions from (75) and (76) is routine. We first generate first- and second-order manifestly interchange symmetric expressions for $\langle\rho_{\sigma,\mathbf{k}\omega}\rangle$ by integrating (75a) and (76) over velocity space. We next drop all terms proportional to $(Xe/Ze)^2$ since, by definition, they are small compared with order unity terms. From the $\hat{\chi}$

constitutive relations (6) and (7), one then obtains the desired RPA expressions:

$$\hat{\chi}^{\mathrm{RPA}}_{\sigma\sigma'}(\mathbf{k}\omega) = \frac{\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{k}\omega)}{Z_\sigma Z_{\sigma'}\phi_{\mathbf{k}}}\left[\frac{\alpha^{\mathrm{RPA}}_{\sigma'}(\mathbf{k}\omega)}{\epsilon^{\mathrm{RPA}}(\mathbf{k}\omega)} - \delta_{\sigma\sigma'}\right] \tag{80}$$

$$\begin{aligned}
&\hat{\chi}^{\mathrm{RPA}}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)\\
&= \frac{1}{\epsilon^{\mathrm{RPA}}(\mathbf{p}\mu)\epsilon^{\mathrm{RPA}}(\mathbf{q}\nu)\epsilon^{\mathrm{RPA}}(\mathbf{k}\omega)}\left(\frac{\beta^2}{2}n_\sigma\frac{\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{p}\mu;\mathbf{q}\nu)}{\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{p}0;\mathbf{q}0)}\right]\left[1+\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{k}\omega)\right]\\
&\quad\times\left\{\delta_{\sigma\sigma'}\left[1+\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{p}\mu)\right] - \delta_{\bar{\sigma}\sigma'}\frac{Z_\sigma}{Z_{\bar{\sigma}}}\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{p}\mu)\right\}\\
&\quad\times\left\{\delta_{\sigma\sigma''}\left[1+\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{q}\nu)\right] - \delta_{\bar{\sigma}\sigma''}\frac{Z_\sigma}{Z_{\bar{\sigma}}}\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{q}\nu)\right\}\\
&\quad-\frac{\beta^2}{2}n_{\bar{\sigma}}\frac{\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{p}\mu;\mathbf{q}\nu)}{\alpha^{\mathrm{RPA}}_{\bar{\sigma}}(\mathbf{p}0;\mathbf{q}0)}\frac{Z_{\bar{\sigma}}}{Z_\sigma}\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{k}\omega)\\
&\quad\times\left\{\delta_{\bar{\sigma}\sigma'}\left[1+\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{p}\mu)\right] - \delta_{\sigma\sigma'}\frac{Z_{\bar{\sigma}}}{Z_\sigma}\alpha^{\mathrm{RPA}}_{\alpha}(\mathbf{p}\mu)\right\}\\
&\quad\times\left\{\delta_{\bar{\sigma}\sigma''}\left[1+\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{q}\nu)\right] - \delta_{\sigma\sigma''}\frac{Z_{\bar{\sigma}}}{Z_\sigma}\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{q}\nu)\right\}\Bigg)
\end{aligned}$$

$$(\sigma,\bar{\sigma}=A,B;\ \sigma\neq\bar{\sigma}),\ (\mathbf{k}=\mathbf{p}+\mathbf{q},\ \omega=\mu+\nu) \tag{81}$$

where $\epsilon^{\mathrm{RPA}}(\mathbf{k}\omega) = 1 + \alpha^{\mathrm{RPA}}_{A}(\mathbf{k}\omega) + \alpha^{\mathrm{RPA}}_{B}(\mathbf{k}\omega)$ is the wave-vector- and frequency-dependent dielectric response function and

$$\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{k}\omega) = \alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{k}0)\frac{1}{\beta m_\sigma n_\sigma}\int d^3v\,\frac{\mathbf{k}\cdot\partial F^{(0)}_{\sigma}(v)/\partial\mathbf{v}}{\omega-\mathbf{k}\cdot\mathbf{v}} \tag{82}$$

$$\begin{aligned}
\alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{p}\mu;\mathbf{q}\nu) &= \alpha^{\mathrm{RPA}}_{\sigma}(\mathbf{p}0;\mathbf{q}0)\frac{1}{\beta m_\sigma n_\sigma}\int d^3v\,\frac{F^{(0)}_{\sigma}(v)}{(\omega-\mathbf{k}\cdot\mathbf{v})^2}\\
&\quad\times\left(\mathbf{k}\cdot\mathbf{p}\frac{\mathbf{q}\cdot\mathbf{v}}{\nu-\mathbf{q}\cdot\mathbf{v}} + \mathbf{k}\cdot\mathbf{q}\frac{\mathbf{p}\cdot\mathbf{v}}{\mu-\mathbf{p}\cdot v}\right)
\end{aligned} \tag{83}$$

are dynamical linear and quadratic polarizabilities with static values given by

$$\alpha_\sigma^{\mathrm{RPA}}(\mathbf{k}0) = \frac{4\pi\beta n_\sigma Z_\sigma^2 e^2}{k^2} \tag{84}$$

$$\alpha_\sigma^{\mathrm{RPA}}(\mathbf{p}0;\mathbf{q}0) = -\frac{2\pi i\beta^2 n_\sigma Z_\sigma^3 e^3}{pqk} \tag{85}$$

Our main task is completed when (81) and the formulas for $\hat{\chi}^{\mathrm{RPA}}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega; \mathbf{p}\mu)$ and $\hat{\chi}^{\mathrm{RPA}}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu; -\mathbf{k}-\omega)$ that (81) generates are substituted into the FDT (58). The resulting expression for the dynamical three-point structure function will be entirely in terms of familiar RPA linear and quadratic polarizability response functions; these latter can be routinely evaluated from (82) and (83).

It is instructive to examine the above results in the static limit where the quadratic polarizability effects are absent. The $\mu = \nu = 0$ version of (81) when combined with the FDT (74), gives the Debye–Hückel (DH) expression

$$\begin{aligned} S_{\sigma\sigma'\sigma''}(\mathbf{pq}) &= \frac{1}{\epsilon^{\mathrm{RPA}}(\mathbf{p}0)\epsilon^{\mathrm{RPA}}(\mathbf{q}0)\epsilon^{\mathrm{RPA}}(\mathbf{k}0)} \\ &\times \Bigg\{ \frac{n_\sigma}{(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}} \left[1 + \alpha_{\bar\sigma}^{\mathrm{RPA}}(\mathbf{k}0)\right] \\ &\times \left\{ \delta_{\sigma\sigma'}\left[1 + \alpha_{\bar\sigma}^{\mathrm{RPA}}(\mathbf{p}0)\right] - \delta_{\bar\sigma\sigma'} \frac{Z_\sigma}{Z_{\bar\sigma}} \alpha_{\bar\sigma}^{\mathrm{RPA}}(\mathbf{p}0) \right\} \\ &\times \left\{ \delta_{\sigma\sigma''}\left[1 + \alpha_{\bar\sigma}^{\mathrm{RPA}}(\mathbf{q}0)\right] - \delta_{\bar\sigma\sigma''} \frac{Z_\sigma}{Z_{\bar\sigma}} \alpha_{\bar\sigma}^{\mathrm{RPA}}(\mathbf{q}0) \right\} \\ &\quad - \frac{n_{\bar\sigma}}{(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}} \frac{Z_{\bar\sigma}}{Z_\sigma} \alpha_\sigma^{\mathrm{RPA}}(\mathbf{k}0) \\ &\quad \times \left\{ \delta_{\bar\sigma\sigma'}\left[1 + \alpha_\sigma^{\mathrm{RPA}}(\mathbf{p}0)\right] - \delta_{\sigma\sigma'} \frac{Z_{\bar\sigma}}{Z_\sigma} \alpha_\sigma^{\mathrm{RPA}}(\mathbf{p}0) \right\} \\ &\quad \times \left\{ \delta_{\bar\sigma\sigma''}\left[1 + \alpha_\sigma^{\mathrm{RPA}}(\mathbf{q}0)\right] - \delta_{\sigma\sigma''} \frac{Z_{\bar\sigma}}{Z_\sigma} \alpha_\sigma^{\mathrm{RPA}}(\mathbf{q}0) \right\} \Bigg\} \end{aligned}$$

$$(\sigma, \bar\sigma = A, B;\ \sigma \neq \bar\sigma)\ (\mathbf{k} = \mathbf{p} + \mathbf{q}) \tag{86}$$

The triple ϵ denominator is certainly a key element in (86) and in the dynamical $S_{\sigma\sigma'\sigma''}$ generated by (81). This can be seen by setting $Z_{\bar{\sigma}} = 0$ and $\sigma = \sigma' = \sigma''$ (so that $\delta_{\sigma\sigma'} = 1 = \delta_{\sigma\sigma''}$, $\delta_{\bar{\sigma}\sigma'} = 0 = \delta_{\bar{\sigma}\sigma''}$); one then recovers from (86) the well-known DH result

$$S(\mathbf{pq}) = \frac{1}{\epsilon^{\mathrm{RPA}}(\mathbf{p}0)\epsilon^{\mathrm{RPA}}(\mathbf{q}0)\epsilon^{\mathrm{RPA}}(\mathbf{k}0)}$$

for the OCP three-point static structure function.

As to the total correlation function, $2P$, one can show from (86) that

$$\begin{aligned} 2P(\mathbf{pq}) &= \sum_{\sigma,\sigma',\sigma''} Z_\sigma Z^{\sigma'} Z^{\sigma''} e^3 (n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} S_{\sigma\sigma'\sigma''}(\mathbf{pq}) \\ &= \frac{e^3}{\epsilon^{\mathrm{RPA}}(\mathbf{p}0)\epsilon^{\mathrm{RPA}}(\mathbf{q}0)\epsilon^{\mathrm{RPA}}(\mathbf{k}0)} (Z_A^3 n_A + Z_B^3 n_B) \\ &= \frac{ikpq}{2\pi\beta^2} \frac{\alpha^{\mathrm{RPA}}(\mathbf{p}0; \mathbf{q}0)}{\epsilon^{\mathrm{RPA}}(\mathbf{p}0)\epsilon^{\mathrm{RPA}}(\mathbf{q}0)\epsilon^{\mathrm{RPA}}(\mathbf{k}0)} \end{aligned} \tag{87}$$

which is precisely the static FDT counterpart of (65) in the RPA [cf. Eq. (85)].

8. CONCLUSIONS

In this paper, we have established nonlinear fluctuation-dissipation relations for magnetic field-free binary ionic mixtures. These new relations are derived from calculations of the longitudinal second-order response of the equilibrium system to partial longitudinal driving fields.

Our principal time domain result (56) links a single equilibrium three-point current correlation function to a combination of three quadratic partial conductivities. A variety of fluctuation-dissipation relations are exhibited in the frequency domain: FDT's (B1) and (B2) are causal with respect to the frequency arguments μ and ν; FDT's (57) and (58) are not; the latter relations are, however, manifestly triangle symmetric under simultaneous rotation of their species indices and four-wave vector arguments $(\mathbf{p}\mu)$, $(\mathbf{q}\nu)$, $(\mathbf{p} + \mathbf{q}, \mu + \nu)$. Equation (58) is the central statement of the present paper. It relates a *single* three-point dynamical structure function to a combination of quadratic partial density response functions. Therefore, it is now possible to obtain a more detailed description of the BIM spectral correlations by evaluating their density response function relatives from model-dependent kinetic equations. We have carried out such an evaluation [Eq. (81)] in the RPA as an appropriate first step in a systematic perturbation theoretic treatment.

One might well ask if the FDT's reported in this paper also apply to electron–ion plasmas. One physical problem which arises in such systems relates to the negatively divergent Coulomb energy of an electron–ion pair at a small separation distance r. As a consequence, the electron–ion pair correlation function diverges as $r \to 0$. This unphysical behavior, which is a defect of the classical theory, can be removed by properly taking into account the atomic bound states. To avoid the rather difficult quantum mechanical treatment which this entails, Dunn and Broyles[20] suggested that the interaction potential $\phi_{ei}(r) = -Z_i e^2/r$ might be suitably softened by multiplying it by a factor $B_{ei}(r) = 1 - \exp(-\mu_{ei} r)$, where μ_{ei}^{-1} is of the order of the Bohr radius. Following the suggestion of Gombert and Deutsch,[21] one might effect similar modifications in the ion–ion and electron–electron interaction potentials to take account of quantum diffraction effects; for the *like* particle interactions, μ^{-1} is of the order of the de Broglie wavelength. The FDT's (56)–(58), (B1), (B2), (B11), and (65), however, are left unaffected by the ensuing modifications in Eqs. (8), (9), (34) and (35). Hence within the context of such phenomenological formulations of the interaction potentials, all of the fluctuation-dissipation relations reported in this paper are valid for electron–ion plasma systems as well.

ACKNOWLEDGMENT

One of the authors (K. I. G.) would like to thank Gabor Kalman for illuminating and clarifying discussions.

APPENDIX A: REVIEW OF LINEAR RESPONSE THEORY

Our starting point is Eq. (44) and its explicit evaluation is straightforward. From (41) and (31),

$$\begin{aligned}\hat{\mathfrak{L}}^{\sigma'}(t)\Omega^{(0)} &= -\frac{i}{\Lambda}\sum_{\mathbf{p}} \hat{V}^{\sigma'}(\mathbf{p}t)\left[\rho_{\sigma',-\mathbf{p}}, \Omega^{(0)}\right] \\ &= -\frac{i\beta\Omega^{(0)}}{\Lambda}\sum_{\mathbf{p}} \hat{V}^{\sigma'}(\mathbf{p}t)\left[H^{(0)}, \rho_{\sigma',-\mathbf{p}}\right] \\ &= -\frac{i\beta\Omega^{(0)}}{\Lambda}\sum_{\mathbf{p}} \hat{V}^{\sigma'}(\mathbf{p}t)\dot{\rho}_{\sigma',-\mathbf{p}} \end{aligned} \tag{A1}$$

whence

$$\Omega^{(1)}(\Gamma t) = \frac{-\beta\Omega^{(0)}}{\lambda} \sum_{\sigma'=A,B} \sum_{\mathbf{p}} \int_0^\infty d\tau\, U(\tau)\hat{V}^{\sigma'}(\mathbf{p}t-\tau)\dot{\rho}_{\sigma,-\mathbf{p}} \tag{A2}$$

The first-order partial density is then calculated according to

$$\langle \rho_{\sigma,\mathbf{k}} \rangle^{(1)}(t) = \int d\Gamma\, \Omega^{(1)}(\Gamma t) \rho_{\sigma,\mathbf{k}}$$

$$= -\frac{\beta}{\Lambda} \sum_{\sigma'} \sum_{\mathbf{p}} \int_0^\infty d\tau\, \hat{V}^{\sigma'}(\mathbf{p}t - \tau) \int d\Gamma\, \Omega^{(0)} \rho_{\sigma,\mathbf{k}} U(\tau) \dot{\rho}_{\sigma',-\mathbf{p}} \quad \text{(A3)}$$

In the above Eulerian (Schrödinger-like) picture, the microscopic density and current operators have no explicit time dependence. This time dependence, however, can be generated by shifting the representation to the Lagrangian (Heisenberg-like) picture. Then choosing $t = 0$ as an arbitrary reference time and letting the time evolution operator act on $\dot{\rho}_{\sigma,\mathbf{p}}$, one arrives at the result

$$\langle \rho_{\sigma,\mathbf{k}}(t) \rangle^{(1)} = -\frac{\beta}{\Lambda} \sum_{\sigma'} \sum_{\mathbf{p}} \int_0^\infty d\tau\, \hat{V}^{\sigma'}(\mathbf{p}t - \tau) \langle \rho_{\sigma,\mathbf{k}}(0) \dot{\rho}_{\sigma',-\mathbf{p}}(-\tau) \rangle^{(0)}$$

$$= \frac{\beta}{\Lambda} \sum_{\sigma'} \sum_{\mathbf{p}} \int_0^\infty d\tau\, \hat{V}^{\sigma'}(\mathbf{p}t - \tau) \frac{d}{d\tau} \langle \rho_{\sigma,\mathbf{k}}(0) \rho_{\sigma',-\mathbf{p}}(-\tau) \rangle^{(0)}$$

$$= -\frac{i\beta}{\Lambda} \sum_{\sigma'} \sum_{\mathbf{p}} \int_{-\infty}^\infty \frac{d\nu}{2\pi} \exp(-i\nu t)\, \hat{V}^{\sigma'}(\mathbf{p}\nu)$$

$$\times \int_{-\infty}^\infty d\mu\, \mu \delta_+(\nu - \mu) \langle \rho_{\sigma,\mathbf{k}}(t=0) \rho^*_{\sigma',\mathbf{p}\mu} \rangle^{(0)} \quad \text{(A4)}$$

whence from (23),

$$\langle \rho_{\sigma,\mathbf{k}\omega} \rangle^{(1)} = -\frac{i\beta}{\Lambda} \sum_{\sigma'} \sum_{\mathbf{p}} \hat{V}^{\sigma'}(\mathbf{p}\omega) \int_{-\infty}^\infty d\mu\, \mu \delta_+(\omega - \mu) \langle \rho_{\sigma,\mathbf{k}}(t=0) \rho^*_{\sigma',\mathbf{p}\mu} \rangle^{(0)}$$

$$= -i\beta \sum_{\sigma'} (n_\sigma n_{\sigma'})^{1/2} \hat{V}^{\sigma'}(\mathbf{k}\omega) \int_{-\infty}^\infty d\mu\, \mu \delta_+(\omega - \mu) S_{\sigma\sigma'}(\mathbf{k}\mu) \quad \text{(A5)}$$

$n_\sigma = N_\sigma/\Lambda$. Comparison of Eqs. (A5) and (6) then gives the (three) linear partial FDT's

$$\hat{\chi}_{\sigma\sigma'}(\mathbf{k}\omega) = -i\beta(n_\sigma n_{\sigma'})^{1/2} \int_{-\infty}^\infty d\mu\, \mu \delta_+(\omega - \mu) S_{\sigma\sigma'}(\mathbf{k}\mu) \qquad (\sigma, \sigma' = A, B) \quad \text{(A6)}$$

from which follow the often quoted[16,17] expressions

$$\operatorname{Im} \hat{\chi}_{\sigma\sigma'}(\mathbf{k}\omega) = -\frac{\beta\omega}{2} (n_\sigma n_{\sigma'})^{1/2} S_{\sigma\sigma'}(\mathbf{k}\omega) \qquad (\sigma, \sigma' = A, B) \quad \text{(A7)}$$

APPENDIX B: FREQUENCY DOMAIN FORMULATION OF THE QUADRATIC FDT'S

In this Appendix, we formulate the quadratic partial FDT's in the frequency domain. First, a straightforward Fourier transformation of (56) provides

$$\begin{aligned}\hat{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) &= \hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) - \int_{-\infty}^{\infty} d\lambda\, \delta_+(\mu+\lambda)\hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}\lambda; -\mathbf{k}\nu-\lambda) \\ &\quad - \int_{-\infty}^{\infty} d\lambda\, \delta_+(\nu+\lambda)\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\mu-\lambda;\mathbf{q}\lambda) \\ &= \frac{i\beta^2}{2}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}\int_{-\infty}^{\infty} d\omega' \\ &\quad \times \int_{-\infty}^{\infty} d\omega''\, \delta_+(\mu-\omega')\delta_+(\nu-\omega'')Q_{\sigma\sigma'\sigma''}(\mathbf{p}\omega';\mathbf{q}\omega'')\end{aligned}$$

$$(\mathbf{k}=\mathbf{p}+\mathbf{q}) \quad \text{(B1)}$$

whence

$$\begin{aligned}&\operatorname{Im}\hat{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \\ &\quad = \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\ &\quad\quad \times \left[Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) - \mathrm{PP}\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{\pi}\frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\omega';\mathbf{q}\omega'')}{(\mu-\omega')(\nu-\omega'')}\right] \quad \text{(B2)}\end{aligned}$$

in virtue of the reality of the current correlation function $Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)$ defined by Eq. (22).

In order to obtain an expression for $Q_{\sigma\sigma'\sigma''}$ in terms of the response function $\hat{\psi}_{\sigma\sigma'\sigma''}$, the integral equation (59) has to be solved. To do this, we first recall the invariance of $Q_{\sigma\sigma'\sigma''}$ with respect to rotation on the triangle formed by the four vectors $(\mathbf{p}\mu)$, $(\mathbf{q}\nu)$, $(\mathbf{k}=\mathbf{p}+\mathbf{q},\ \omega=\mu+\nu)$ [cf. Eq. (27)]. The Hilbert transform operation, however, violates this invariance so that $\operatorname{Im}\hat{\psi}_{\sigma\sigma'\sigma''}$ does not satisfy a similar symmetry rule. One can, nevertheless, form the symmetrized combination,

$$\begin{aligned}&\tilde{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) \\ &\quad = \operatorname{Im}\tfrac{1}{3}\left\{\hat{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) + \hat{\psi}_{\sigma'\sigma\sigma''}(-\mathbf{k}-\omega;\mathbf{q}\nu) + \hat{\psi}_{\sigma''\sigma'\sigma}(\mathbf{p}\mu;-\mathbf{k}-\omega)\right\}\end{aligned}$$

$$\text{(B3)}$$

which does possess the triangle symmetry exhibited by $Q_{\sigma\sigma'\sigma''}$. A simple expression for $\tilde{\psi}_{\sigma\sigma'\sigma''}$ in terms of the $\hat{\eta}$'s can be evolved first by expanding each right-hand-side member of (B3) [starting from the definition of $\hat{\psi}_{\sigma\sigma'\sigma''}$

in (B1)] and then recombining the results. Thus,

$$\begin{aligned}
\operatorname{Im}\hat{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)
&= \operatorname{Im}\left\{\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) - \frac{1}{2}\hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}-\mu;-\mathbf{k}\omega) - \frac{1}{2}\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\omega;\mathbf{q}-\nu)\right\}\\
&\quad - \frac{1}{2\pi}P\int\frac{d\lambda}{\mu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}\lambda;-\mathbf{k}\nu-\lambda)\\
&\quad - \frac{1}{2\pi}P\int\frac{d\lambda}{\nu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\mu-\lambda;\mathbf{q}\lambda) \qquad \text{(B4a)}\\
&= \operatorname{Im}\left\{\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) + \frac{1}{2}\hat{\eta}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\nu) + \frac{1}{2}\hat{\eta}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)\right\}\\
&\quad - \frac{1}{2\pi}P\int\frac{d\lambda}{\mu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}\lambda;-\mathbf{k}\nu-\lambda)\\
&\quad - \frac{1}{2\pi}P\int\frac{d\lambda}{\nu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\mu-\lambda;\mathbf{q}\lambda) \qquad \text{(B4b)}
\end{aligned}$$

In deriving (B4b) from (B4a), we have exploited both the interchange symmetry rule (15) and the reality–spatial reflection invariance condition (18). From (B4b) and (15) one can then show that

$$\begin{aligned}
\operatorname{Im}\hat{\psi}_{\sigma'\sigma\sigma''}(-\mathbf{k}-\omega;\mathbf{q}\nu)
&= \operatorname{Im}\left\{\hat{\eta}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega) + \frac{1}{2}\hat{\eta}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) + \frac{1}{2}\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)\right\}\\
&\quad + \frac{1}{2\pi}P\int\frac{d\lambda}{\mu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma''\sigma'\sigma}(\mathbf{p}\lambda;-\mathbf{k}\nu-\lambda)\\
&\quad - \frac{1}{2\pi}P\int\frac{d\lambda}{\nu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}-\omega-\lambda;\mathbf{q}\lambda) \qquad \text{(B5)}
\end{aligned}$$

and

$$\begin{aligned}
\operatorname{Im}\hat{\psi}_{\sigma''\sigma'\sigma}(\mathbf{p}\mu;-\mathbf{k}-\omega)
&= \operatorname{Im}\left\{\hat{\eta}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) + \frac{1}{2}\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) + \frac{1}{2}\hat{\eta}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)\right\}\\
&\quad + \frac{1}{2\pi}P\int\frac{d\lambda}{\nu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}-\omega-\lambda;\mathbf{q}\lambda)\\
&\quad + \frac{1}{2\pi}P\int\frac{d\lambda}{\nu+\lambda}\operatorname{Re}\hat{\eta}_{\sigma'\sigma\sigma''}(-\mathbf{k}\mu-\lambda;\mathbf{q}\lambda) \qquad \text{(B6)}
\end{aligned}$$

The combination of Eqs. (B4b) to (B6) according to (B3) now yields the desired triangle symmetric result:

$$\begin{aligned}
\tilde{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu)
&= \operatorname{Im}\tfrac{2}{3}\left\{\hat{\eta}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu;\mathbf{q}\nu) + \hat{\eta}_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega;\mathbf{p}\mu) + \hat{\eta}_{\sigma'\sigma''\sigma}(\mathbf{q}\nu;-\mathbf{k}-\omega)\right\}
\end{aligned} \qquad \text{(B7)}$$

Now a triangle-symmetrized FDT can be derived from Eqs. (B2), (B3), and (B7) by first observing from (B2) that

$$
\begin{aligned}
\operatorname{Im} \hat{\psi}_{\sigma''\sigma'\sigma}(\mathbf{p}\mu; -\mathbf{k}-\omega)
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma''\sigma'\sigma}(\mathbf{p}\mu; -\mathbf{k}-\omega) + \mathrm{PP}\int \frac{d\omega'}{\pi}\int\frac{d\omega''}{\pi} \frac{Q_{\sigma''\sigma'\sigma}(\mathbf{p}\omega'; -\mathbf{k}\omega'')}{(\mu-\omega')(\omega+\omega'')} \right] \\
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma''\sigma\sigma'}(-\mathbf{k}-\omega; \mathbf{p}\mu) + \mathrm{PP}\int \frac{d\bar{\mu}}{\pi}\int\frac{d\bar{\nu}}{\pi} \frac{Q_{\sigma''\sigma\sigma'}(-\mathbf{k}, -\bar{\mu}-\bar{\nu}; \mathbf{p}\bar{\mu})}{(\mu-\bar{\mu})(\omega-\bar{\mu}-\bar{\nu})} \right] \\
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) + \mathrm{PP}\int \frac{d\bar{\mu}}{\pi}\int\frac{d\bar{\nu}}{\pi} \frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\bar{\mu}; \mathbf{q}\bar{\nu})}{(\mu-\bar{\mu})(\omega-\bar{\mu}-\bar{\nu})} \right]
\end{aligned} \tag{B8}
$$

the second step exploits the interchange symmetry property of $Q_{\sigma\sigma'\sigma''}$, and the third step follows in virture of the triangle symmetry rule (27). Next, from (B2) and (27), we similarly have that

$$
\begin{aligned}
\operatorname{Im} \hat{\psi}_{\sigma'\sigma\sigma''}(-\mathbf{k}-\omega; \mathbf{q}\nu)
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma'\sigma\sigma''}(-\mathbf{k}-\omega; \mathbf{q}\nu) + \mathrm{PP}\int \frac{d\omega'}{\pi}\int\frac{d\omega''}{\pi} \frac{Q_{\sigma'\sigma\sigma''}(-\mathbf{k}\omega'; \mathbf{q}\omega'')}{(\omega+\omega')(\nu-\omega'')} \right] \\
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma'\sigma''\sigma}(\mathbf{q}\nu; -\mathbf{k}-\omega) + \mathrm{PP}\int \frac{d\bar{\nu}}{\pi}\int\frac{d\bar{\mu}}{\pi} \frac{Q_{\sigma'\sigma''\sigma}(\mathbf{q}\bar{\nu}; -\mathbf{k}-\bar{\mu}-\bar{\nu})}{(\nu-\bar{\nu})(\omega-\bar{\mu}-\bar{\nu})} \right] \\
&= \frac{\beta^2}{8}(n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} \\
&\quad \times \left[Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) + \mathrm{PP}\int \frac{d\bar{\nu}}{\pi}\int\frac{d\bar{\mu}}{\pi} \frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\bar{\mu}; \mathbf{q}\bar{\nu})}{(\nu-\bar{\nu})(\omega-\bar{\mu}-\bar{\nu})} \right]
\end{aligned} \tag{B9}
$$

In the second step, the order of integration has been reversed to preserve the separable integrability features of the denominator under the dummy variable transformations $\omega' = -\bar{\mu} - \bar{\nu}$, $\omega'' = \bar{\nu}$. In order to bring (B9) to a form comparable with (B8) the Poincaré–Bertrand theorem[18] has to be invoked providing

$$\mathrm{PP}\int \frac{d\bar{\nu}}{\pi} \int \frac{d\bar{\mu}}{\pi} \frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\bar{\mu}; \mathbf{q}\bar{\nu})}{(\nu - \bar{\nu})(\omega - \bar{\mu} - \bar{\nu})}$$

$$= Q_{\sigma\sigma'\sigma''}(p\mu; q\nu) + \mathrm{PP}\int \frac{d\bar{\mu}}{\pi} \int \frac{d\bar{\nu}}{\pi} \frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\bar{\mu}; \mathbf{q}\bar{\nu})}{(\nu - \bar{\nu})(\omega - \bar{\mu} - \bar{\nu})}$$

whence

$$\operatorname{Im} \hat{\psi}_{\sigma'\sigma\sigma''}(-\mathbf{k} - \omega; \mathbf{q}\nu)$$

$$= \frac{\beta^2}{8} (n_\sigma n_{\sigma'} n_{\sigma''})^{1/3}$$

$$\times \left[2Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) + \mathrm{PP}\int \frac{d\bar{\mu}}{\pi} \int \frac{d\bar{\nu}}{\pi} \frac{Q_{\sigma\sigma'\sigma''}(\mathbf{p}\bar{\mu}; \mathbf{q}\bar{\nu})}{(\nu - \bar{\nu})(\omega - \bar{\mu} - \bar{\nu})} \right] \quad \text{(B10)}$$

We now substitute (B2), (B8), and (B10) into (B3) to obtain the FDT relation

$$\tilde{\psi}_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) = \frac{\beta^2}{6} (n_\sigma n_{\sigma'} n_{\sigma''})^{1/3} Q_{\sigma\sigma'\sigma''}(\mathbf{p}\mu; \mathbf{q}\nu) \quad \text{(B11)}$$

in a form in which both sides are manifestly triangle symmetric. Equations (B7) and (B11) then combine to yield the desired frequency domain FDT result Eq. (57).

REFERENCES

1. (a) H. Nyquist, *Phys. Rev.* **32**:110 (1928); (b) H. B. Callen and T. A. Welton, *Phys. Rev.* **83**:34 (1951); (c) R. Kubo, *J. Phys. Soc. Jpn.* **12**:570 (1957); (d) R. Kubo, in *Lectures in Theoretical Physics*, Vol. I, W. E. Brittin and L. H. Dunham, eds., Interscience, New York (1959), pp. 134–143, 147, 148, 155, 156; (e) R. Kubo, *Rep. Progr. Phys.* **29**:(Part I), 263–283 (1966); (f) E. W. Montroll and J. C. Ward, *Physica* **25**:423 (1959); (g) E. W. Montroll, in *Theory of Neutral and Ionized Gases*, C. DeWitt and J. F. Detoeuff, eds., Université de Grenoble, Ecole d'été de physique théorique, Les Houches (1960), pp. 95–100; (h) G. V. Chester, *Rep. Progr. Phys.* **26**:411 (1963); (i) A. G. Sitenko, *Electromagnetic Fluctuations in Plasma*, Academic Press, New York (1967), p. 256; (j) K. I. Golden and G. Kalman, *J. Stat. Phys.* **1**:415 (1969).
2. K. I. Golden, G. Kalman, and M. B. Silevitch, *J. Stat. Phys.* **6**:87 (1972).
3. (a) A. G. Sitenko, *Phys. Scr.* **7**:190 (1973); (b) A. G. Sitenko, *Fluctuations and Nonlinear Wave Interaction in Plasma*, Pergamon Press, Oxford, (1980); (c) A. G. Sitenko, *Zh. Eksp. Teor. Fiz.* **75**:104 (1978) [*Sov. Phys. JETP* **48**:51 (1978)].

4. G. F. Efremov, *Zh. Eksp. Teor. Fiz.* **55**:2322 (1968) [*Sov. Phys. JETP* **28**:1232 (1969)].
5. (a) H. L. Friedman and A. Ben-Naim, *J. Chem. Phys.* **48**:120 (1968); (b) S. Harris and H. L. Friedman, *J. Chem. Phys.* **50**:765 (1969).
6. A. Yu. Kargin, *Sum Rules and Nonlinear Fluctuation-Dissipation Theorem for the Plasma Kinetic Coefficients*, Academy of Sciences of the Ukrainian SSR Institute for Theoretical Physics, preprint ITP-81-36E (1981).
7. K. I. Golden and G. Kalman, *Plasma Response Functions, Fluctuation-Dissipation Relations and the Velocity-Average-Approximation*, Ann. Phys. (N.Y.) Vol. 143 (1982).
8. M. B. Silevitch and K. I. Golden, *J. Stat. Phys.* **7**:65 (1973).
9. K. I. Golden, G. Kalman, and T. Datta, *Phys. Rev. A* **11**:2147 (1975).
10. Y. Shima, H. Yatom, K. I. Golden, and G. Kalman, *Phys. Rev. A* **22**:2818 (1980).
11. (a) K. I. Golden, G. Kalman, and M. B. Silevitch, *Phys. Rev. Lett.* **33**:1544 (1974); (b) K. I. Golden and G. Kalman, *Phys. Rev. A* **19**:2112 (1979); (c) P. Carini, G. Kalman, and K. I. Golden, *Phys. Lett.* **78A**:450 (1980).
12. (a) I. R. McDonald, P. Vieillefosse, and J. P. Hansen, *Phys. Rev. Lett.* **39**:271 (1977); (b) J. P. Hansen, I. R. McDonald, and P. Vieillefosse, *Phys. Rev. A* **20**:2590 (1979).
13. (a) M. Baus, *Physica* **A88**:319 (1977); (b) **88**:366 (1977); (c) **88**:591 (1977); (d) M. Baus, *Phys. Rev. Lett.* **40**:793 (1978).
14. F. Postogna and M. P. Tosi, *Nuovo Cimento* **55B**:399 (1980).
15. (a) P. Vashishta, P. Bhattacaryya, and K. S. Singwi, *Phys. Rev. B* **10**:5108 (1974); (b) P. Vashishta, P. Bhattacaryya and K. S. Singwi, *Nuovo Cimento* **B23**:172 (1974).
16. M. P. Tosi, M. Parinello, and N. H. March, *Nuovo Cimento* **B23**:135 (1974).
17. G. Kalman, Methods and approximations for strongly coupled plasmas, in *Strongly Coupled Plasmas*, G. Kalman ed., Plenum Press, New York (1978), p. 225.
18. (a) N. I. Muskhelishvili, *Singular Integral Equations*, P. Noordhoff, Groningen, Holland (1953), p. 57, Eq. (23.F); (b) R. Balescu, *Statistical Mechanics of Charged Particles*, Vol. IV, Interscience, London (1963).
19. K. I. Golden and G. Kalman, *Phys. Rev. A* **14**:1802 (1976).
20. T. Dunn and A. A. Broyles, *Phys. Rev. A* **157**:156 (1967).
21. M. M. Gombert and C. Deutsch, *Phys. Lett.* **47A**:473 (1974).