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Nonlinear Fluctuation–Dissipation Theorem

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Using statistical mechanical perturbation theory, the second-order average current density response is calculated for magnetic field-free classical plasmas. A dynamical fluctuation-dissipation theorem is then derived, thus establishing a connection between triplet microscopic current–current correlations and quadratic response functions; it also leads to a static fluctuation-dissipation theorem which provides a dielectric description of the equilibrium ternary correlation. A comparison of the latter with its expansion in terms of the Mayer pair correlation clusters is discussed.

KEY WORDS: Plasmas; quadratic response functions; external conductivity; dielectric function; triplet correlation function; ternary correlation function; Mayer cluster expansion.

1. INTRODUCTION

The conventional fluctuation-dissipation theorem (FDT) establishes a relation between the equilibrium correlations in and the linear response of the same system to a small external perturbation.[1] In particular, the pertinent correlations are doublet correlations, i.e., correlations connecting physical quantities at two different space–time points. A specialized form of the general dynamical FDT is its static variant, which provides information about the pair correlation function in thermal equilibrium.[2]

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The response of a system, in general, is, however, not restricted to be linear. In the family of nonlinear response functions, second-order ones are the simplest objects and their properties and explicit functional forms have been studied in plasma physics, crystal physics, and nonlinear optics.[3] That such a quadratic response function should satisfy some kind of a fluctuation-dissipation-like theorem is a rather obvious expectation. Indeed, even a cursory reflection over the derivation of the linear FDT should suggest a connection between second-order response and triplet correlations.

The present paper is devoted to the establishment and examination of this *quadratic fluctuation-dissipation* theorem, whose existence, thus, is not unexpected. The relation and its derivation are, however, far from being a trivial exercise in perturbation theory. Of special interest is its static (dc) limit, which leads to a relation for the three-particle (ternary) correlation function in thermal equilibrium in terms of the static quadratic response function. The linear FDT already possesses a noteworthy "order-raising" property: When applied to quantities calculated in the "plasma approximation, i.e., by expanding in the small parameter $(ne^2\beta)^{1/2}\, e^2\beta$ (β is the inverse temperature), the FDT expresses $(n + 1)$th-order (in particular, first-order) pair correlation functions in terms of the nth-order (in particular, Vlasov) response functions. A similar, though enhanced feature prevails in the present case: The "order-raising" now operates by two units, thus providing a connection between the $(n + 2)$th-order (in particular, second-order) triplet correlation function and the nth-order (in particular, Vlasov) quadratic response.

FDT relationships between higher-order correlations and transport coefficients have also been established by Friedman *et al.*[4] for systems (such as partially ionized gases and electrolyte solutions) under homogeneous and stationary perturbations. 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, our analysis is restricted to a consideration of diagonal longitudinal (with respect to the wave vectors) elements only and assumes that the driving perturbations are functions of space and time. We therefore obtain wavevector- and frequency-dependent FDT relations which are model-independent. Our analysis nevertheless shares one common feature with that of Friedman *et al.* : Both their and our approach have independently established that the second-order longitudinal projection of the wavevector- and frequency-independent conductivity is identically zero.

While in this study we restrict ourselves to a consideration of fully ionized plasma, we do this for the sake of concreteness and in order to deal with an easily tractable model; it should be understood that our conductivity-equilibrium correlation FDT relations and the ensuing dielectric formulation of the ternary correlation function are entirely valid for weakly ionized plasmas and related systems. The collisional model dependence arises only when one seeks more detailed descriptions of the correlations, obtained from an evaluation of the transport coefficients from specific kinetic equations. The order-raising property of the so-called static FDT, for example, is model-dependent; so is the lowest-order Mayer cluster expansion for the ternary correlations function obtained by evaluating the dielectric functions from the Vlasov equation; both of these approaches are based on an explicit expansion of the collision operator in e^2, the plasma coupling parameter.

There is no external magnetic field in our model and we consider a longitudinal (Coulomb) field only as the perturbation. Therefore there is no magnetic field at all in the system and that leaves two independent elements in the three-dimensional $3 \times 3 \times 3$ matrices of the conductivity tensor $\hat{\sigma}_{\alpha\beta\gamma}$ [see Eq. (26)] and related objects.

This remainder of this paper is divided into two main parts. Section 2 concerns itself with the development of nonlinear dynamical fluctuation-dissipation theorems, while Section 3 considers their static limit and subsequent application to equilibrium statistical-mechanical calculations of the ternary correlation functions. More precisely, Section 2.1 describes the unperturbed state of the magnetic field-free plasma. In Section 2.2, we let the equilibrium plasma be driven by a small time-dependent scalar potential, and we determine the subsequent average second-order current density response using the well-known statistical-mechanical perturbation-theoretic method of Kubo.[1c,d,e] In this method, one essentially follows the evolution of the first- and second-order Liouville distribution functions; the average second-order response, which is of interest here, is then calculated by ensemble-averaging the microscopic current density over the second-order distribution function. This result and Ohm's law for the *external* conductivity (response function which connects the induced current to products of the driving electric field) are then used in Sections 2.3 and 2.4 to establish dynamical FDT relationships between the quadratic external conductivity and triplet correlation of the microscopic current densities in the equilibrium state. Our results are presented in a variety of forms. For example, the FDT's (33) and (48) each relate a single current correlation to a combination of three conductivities. Such time-domain and frequency-domain representations are seen to be manifestly symmetric under interchange of their two space–time (or equivalently $\mathbf{k}\omega$) variables. Beyond this, we shall see that FDT relations like (48) and (66) exhibit a higher triangle antisymmetry for three ($\mathbf{k}\omega$) defined by Eqs. (40). We exploit this symmetry in Section 2.4 to extricate the current correlation function from under the integral equation it originally is associated with, thus enabling us to derive explicit relations for it. In Section 2.4, we introduce new internal response functions, paving the way for Section 3, where, in Section 3.1, a simple relationship is then established between the equal-time ternary charge density correlation function and a single static (zero-frequency) internal response function. This result is then used in Section 3.2 to establish the FDT for the ternary correlation of equilibrium statistical mechanics. Finally, in Section 3.3, we compare these results with the relations provided by the Mayer-type cluster expansion in equilibrium statistical mechanics and by BBGKY-type kinetic equations for the connection between ternary and binary correlation functions.

2. NONLINEAR RESPONSE AND THE DYNAMICAL FLUCTUATION-DISSIPATION THEOREM

2.1. Description of the Unperturbed System

Consider a classical electron plasma in the large but bounded volume V. For the present, only the N electrons (each of mass m and charge $e = -|e|$) are assumed to be dynamical; the N/Z positive ions (each of mass M and charge $Z|e|$), which are present to provide a stabilizing background, are considered to be nailed down to fixed spatial points.[3]

The microscopic charge and current densities at the space–time point $(\mathbf{r}, t)$ are respectively given by

$$\rho(\mathbf{r}, t) = e \sum_{i=1}^{N} \delta(\mathbf{r} - \mathbf{x}_i(t)) \tag{1}$$

$$\mathbf{j}(\mathbf{r}, t) = e \sum_{i=1}^{N} \mathbf{v}_i(t)\, \delta(\mathbf{r} - \mathbf{x}_i(t)) \tag{2}$$

with spatial Fourier transforms[4]

$$\rho_{\mathbf{k}}(t) = e \sum_{i=1}^{N} \exp[-i\mathbf{k} \cdot \mathbf{x}_i(t)] \tag{3}$$

[3] Our theory can be easily extended to take account of dynamical ion motions [e.g., see Eq. (78)]. In this more realistic plasma model, the ions and electrons are then assumed to coexist in thermal equilibrium ($T_i = T_e$).

[4] We adopt here the Fourier transform convention:

$$f(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} dt \int_V d^3\mathbf{r}\{\exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})]\} f(\mathbf{r}, t)$$

$$f(\mathbf{r}, t) = (1/V) \sum_{\mathbf{k}} \int_{-\infty}^{\infty} (d\omega/2\pi)\{\exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)]\} f(\mathbf{k}, \omega)$$

$$\mathbf{j}_{\mathbf{k}}(t) = e \sum_{i=1}^{N} \mathbf{v}_i(t) \exp[-i\mathbf{k} \cdot \mathbf{x}_i(t)] \tag{4}$$

where the $\mathbf{x}_i$ and $\mathbf{v}_i$ are the particle coordinates and velocities, respectively.

The unperturbed state of the electron plasma in the infinite past is characterized by the macrocanonical distribution function (normalized to unity):

$$\Omega^{(0)} = (\exp -\beta H^{(0)}) \Big/ \int dx\, dp \exp -\beta H^{(0)} \tag{5}$$

where $\mathbf{p}_i$ is the ith particle momentum,

$$dx\, dp = \prod_{i=1}^{N} \prod_{j=1}^{N+(N/Z)} d^3\mathbf{p}_i\, d^3\mathbf{x}_j$$

β^{-1} is the temperature in energy units, and

$$H^{(0)} = \sum_{i=1}^{N} (p_i^2/2m) + \tfrac{1}{2}e^2 \sum_{\substack{i,j=1\\ i\neq j}} \phi_{ij}(\mathbf{x}_i - \mathbf{x}_j) + Ze^2 \sum_{i=1}^{N} \sum_{j=1}^{N/Z} \phi_{ij}(\mathbf{x}_i - \mathbf{x}_j)$$
$$+ \tfrac{1}{2}Z^2e^2 \sum_{\substack{i,j=1\\ i\neq j}}^{N/Z} \phi_{ij}(\mathbf{x}_i - \mathbf{x}_j) \tag{6}$$

is the unperturbed Hamiltonian including electron–electron, electron–ion, and ion–ion interactions. For a partially ionized plasma (which is not of interest in this paper), $H^{(0)}$ should of course be extended to include both the kinetic energy contributions from the neutral constituents and neutral–neutral and neutral–charged-particle interactions. In the sequel, we shall see that the average current density response is independent of any such modifications in $H^{(0)}$ (in so far as $H^{(0)}$ is explicitly independent of the perturbing field) and depends explicitly on the form of the Hamiltonian $H^{(1)}$ for the interaction between the plasma and the external electric field perturbation. Since $H^{(1)}$ is unaffected by the presence of neutrals [see Eq. (11) below], it is clear that the conductivity-equilibrium correlation FDT relations of this paper will have the same form, whatever be the (nonzero) degree of ionization.

2.2. Nonlinear Response Theory

Following the well-known statistical-mechanical perturbation-theoretic method of Kubo, we analyze the second-order response of the plasma to the weak time-dependent external scalar potential

$$\hat{\phi}(\mathbf{r}, t) = (1/V) \sum_{\mathbf{k}'} \hat{\phi}_{\mathbf{k}'}(t) \exp(i\mathbf{k}' \cdot \mathbf{r}) \tag{7}$$

We assume that the perturbation is sufficiently weak to allow one to expand the relevant quantities in powers of the perturbing field.

The Hamiltonian H and the Liouville operator

$$L = -i[H,...] = i \sum_{i=1}^{N} \left(\frac{\partial H}{\partial \mathbf{x}_i} \cdot \frac{\partial}{\partial \mathbf{p}_i} - \frac{\partial H}{\partial \mathbf{p}_i} \cdot \frac{\partial}{\partial \mathbf{x}_i} \right) \tag{8}$$

are, at most, perturbed to first order in $\hat{\phi}$, i.e.,

$$H = H^{(0)} + H^{(1)} \tag{9}$$

$$L = L^{(0)} + L^{(1)} \tag{10}$$

where

$$H^{(1)}(t) = (1/V) \sum_{\mathbf{k}} \hat{\phi}_{\mathbf{k}}(t)\, \rho_{-\mathbf{k}}\,, \tag{11}$$

and

$$L^{(1)}(t) = -(i/V) \sum_{\mathbf{k}} \hat{\phi}_{\mathbf{k}}(t)[\rho_{-\mathbf{k}}\,,...] \tag{12}$$

The subsequent perturbation of the Liouville equation

$$\partial \Omega / \partial t = -iL\Omega \tag{13}$$

for the distribution function Ω results in the formal solution

$$\Omega = \Omega^{(0)} + \Omega^{(1)} + \Omega^{(2)} + \cdots \tag{14}$$

where

$$\Omega^{(1)}(t) = -i \int_0^{\infty} d\tau\, (\exp -i\tau L^{(0)})\, L^{(1)}(t-\tau)\, \Omega^{(0)} \tag{15}$$

$$\Omega^{(2)}(t) = - \int_0^{\infty} d\tau \int_0^{\infty} d\tau'\, (\exp -i\tau L^{(0)})\, L^{(1)}(t-\tau) \times (\exp -i\tau' L^{(0)})\, L^{(1)}(t-\tau-\tau')\, \Omega^{(0)} \tag{16}$$

Further development of the expression (15) for $\Omega^{(1)}$ and subsequent ensemble-averaging of the microscopic current density (4) over $\Omega^{(1)}$ leads to the well-known average first-order current density response and linear conductivity.[5] It is the development of $\Omega^{(2)}$ with is of interest in this paper.

For this calculation, we first observe from (12) and (5) that

$$\begin{aligned} L^{(1)}(t-\tau-\tau')\, \Omega^{(0)} &= -(i/V) \sum_{\mathbf{k}''} \hat{\phi}_{\mathbf{k}''}(t-\tau-\tau')[\rho_{-\mathbf{k}''}(t), \Omega^{(0)}] \\ &= (-i\beta\Omega^{(0)}/V) \sum_{\mathbf{k}''} \hat{\phi}_{\mathbf{k}''}(t-\tau-\tau')[H^{(0)}, \rho_{-\mathbf{k}''}(t)] \\ &= (i\beta\Omega^{(0)}/V) \sum_{\mathbf{k}''} \mathrm{k}''_{\beta} \mathrm{k}''_{\nu} \hat{F}_{\mathbf{k}''\nu}(t-\tau-\tau')\, \mathbf{j}_{-\mathbf{k}''\beta}(t) \end{aligned} \tag{17}$$

[5] See, for example, Ref. 1(c), p. 580, Eq. (5.11) with $h = 0$.

where k″ is the unit wave vector and $\hat{\mathbf{E}}_{\mathbf{k}''} = -i\mathbf{k}''\hat{\phi}_{\mathbf{k}''}$. Since

$$\mathbf{j}_{\mathbf{k}}(t) = (\exp i\tau L^{(0)})\, \mathbf{j}_{\mathbf{k}}(0) \tag{18}$$

satisfies the "Heisenberg" equation

$$d\mathbf{j}_{\mathbf{k}}(t)/dt = iL^{(0)}\mathbf{j}_{\mathbf{k}}(t)$$

it follows that

$$\begin{aligned}(\exp -i\tau' L^{(0)})\, &L^{(1)}(t - \tau - \tau')\, \Omega^{(0)} \\ &= (i\beta\Omega^{(0)}/V) \sum_{\mathbf{k}''} \mathrm{k}''_\beta \mathrm{k}''_\nu \hat{E}_{\mathbf{k}''\nu}(t - \tau - \tau')\, j_{-\mathbf{k}''\beta}(t - \tau')\end{aligned} \tag{19}$$

Continuing the calculation according to (16) and using (12), one has that

$$\begin{aligned}&L^{(1)}(t - \tau)(\exp -i\tau' L^{(0)})\, L^{(1)}(t - \tau - \tau')\, \Omega^{(0)} \\ &= \frac{\beta}{V^2} \sum_{\mathbf{k}',\mathbf{k}''} \mathrm{k}''_\beta \mathrm{k}''_\nu \hat{E}_{\mathbf{k}''\nu}(t - \tau - \tau')\, \hat{\phi}_{\mathbf{k}'}(t - \tau')[\rho_{-\mathbf{k}'}(t), \Omega^{(0)} j_{-\mathbf{k}''\beta}(t - \tau')] \\ &= \frac{i\beta\Omega^{(0)}}{V^2} \sum_{\mathbf{k}',\mathbf{k}''} \frac{\mathrm{k}''_\beta \mathrm{k}_\mu' \mathrm{k}''_\nu}{k'} \hat{E}_{\mathbf{k}'\mu}(t - \tau)\, \hat{E}_{\mathbf{k}''\nu}(t - \tau - \tau') \\ &\quad \times \{i\beta k_\alpha' j_{-\mathbf{k}'\alpha}(t)\, j_{-\mathbf{k}''\beta}(t - \tau') + [\rho_{-\mathbf{k}}(t), j_{-\mathbf{k}''\beta}(t - \tau')]\}\end{aligned} \tag{20}$$

so that use of the Heisenberg time-shifting property (18) in the subsequent operation of $\exp(-i\tau L^{(0)})$ on (20) ultimately leads to

$$\begin{aligned}\Omega^{(2)}(t) = \frac{\beta\Omega^{(0)}}{V^2} \sum_{\mathbf{k}',\mathbf{k}''} \frac{\mathrm{k}_\mu' \mathrm{k}''_\beta \mathrm{k}''_\nu}{k'} \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt''\, \hat{E}_{\mathbf{k}'\mu}(t')\, \hat{E}_{\mathbf{k}''\nu}(t'') \\ \times \{\beta k_\alpha' j_{-\mathbf{k}'\alpha}(t')\, j_{-\mathbf{k}''\beta}(t'') - i[\rho_{-\mathbf{k}'}(t'), j_{-\mathbf{k}''\beta}(t'')]\}\end{aligned} \tag{21}$$

where $t' = t - \tau$, $t'' = t' - \tau'$. The two times t' and t'' are physically equivalent: therefore a symmetry with respect to prime–double-prime interchange should prevail. In order to make the symmetry manifest, the order of integration should first be reversed as follows:

$$\begin{aligned}\Omega^{(2)}(t) = \frac{\beta\Omega^{(0)}}{V^2} \sum_{\mathbf{k}',\mathbf{k}''} \frac{\mathrm{k}_\mu' \mathrm{k}''_\beta \mathrm{k}''_\nu}{k'} \int_{-\infty}^{t} dt'' \int_{t''}^{t} dt'\, \hat{E}_{\mathbf{k}'\mu}(t')\, \hat{E}_{\mathbf{k}''\nu}(t'') \\ \times \{\beta k_\alpha' j_{-\mathbf{k}'\alpha}(t')\, j_{-\mathbf{k}''\beta}(t'') - i[\rho_{-\mathbf{k}'}(t'), j_{-\mathbf{k}''\beta}(t'')]\}\end{aligned} \tag{22}$$

Then, upon interchanging the t' and t'' variables, making the appropriate interchanges among the (dummy) tensor indices, and combining the resulting expressions with (21), one obtains

$$\begin{aligned}
\Omega^{(2)}(t)& \\
&= \frac{\beta^2\Omega^{(0)}}{2V^2}\sum_{\mathbf{k}',\mathbf{k}''} \mathrm{k}_\alpha'\mathrm{k}_\mu'\mathrm{k}_\beta''\mathrm{k}_\nu'' \int_{-\infty}^{t} dt' \int_{-\infty}^{t} dt''\, \hat{E}_{\mathbf{k}'\mu}(t')\, \hat{E}_{\mathbf{k}''\nu}(t'')\, j_{-\mathbf{k}'\alpha}(t')\, j_{-\mathbf{k}''\beta}(t'') \\
&- \frac{i\beta\Omega^{(0)}}{2V^2}\sum_{\mathbf{k}',\mathbf{k}''} \frac{\mathrm{k}_\mu'\mathrm{k}_\beta''\mathrm{k}_\nu''}{k'} \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt''\, \hat{E}_{\mathbf{k}'\mu}(t')\, \hat{E}_{\mathbf{k}''\nu}(t'')[\rho_{-\mathbf{k}'}(t'), j_{-\mathbf{k}''\beta}(t'')] \\
&- \frac{i\beta\Omega^{(0)}}{2V^2}\sum_{\mathbf{k}',\mathbf{k}''} \frac{\mathrm{k}_\mu'\mathrm{k}_\beta'\mathrm{k}_\nu''}{k''} \int_{-\infty}^{t} dt' \int_{t'}^{t} dt''\, \hat{E}_{\mathbf{k}'\mu}(t')\, \hat{E}_{\mathbf{k}''\nu}(t'')[\rho_{-\mathbf{k}''}(t''), j_{-\mathbf{k}'\beta}(t')]
\end{aligned} \tag{23}$$

We are now ready to calculate the average second-order current density response according to

$$\langle j_{\mathbf{k}\gamma}(t)\rangle^{(2)} = \int dx\, dp\, \Omega^{(2)} j_{\mathbf{k}\gamma} \tag{24}$$

It is convenient to rewrite (23) in terms of the relative time variables $\tau' = t - t'$, $\tau'' = t - t''$. Then, upon putting this latter into (24) and exploiting the stationarity of the equilibrium system, one obtains an expression which is most conveniently formulated as an integration over the entire time domain by introducing appropriate step (Θ) functions. Thus the average second-order current density response becomes

$$\begin{aligned}
\langle j_{\mathbf{k}\gamma}(t)\rangle^{(2)}& \\
&= \frac{\beta}{2V^2}\sum_{\mathbf{k}'} \mathrm{k}_\mu'\mathrm{k}_\nu'' \int_{-\infty}^{\infty} d\tau' \int_{-\infty}^{\infty} d\tau''\, \hat{E}_{\mathbf{k}'\mu}(t-\tau')\, \hat{E}_{\mathbf{k}''\nu}(t-\tau'')\, \Theta(\tau')\, \Theta(\tau'') \\
&\quad \times \{\beta \mathrm{k}_\alpha'\mathrm{k}_\beta'' \langle j_{-\mathbf{k}'\alpha}(0)\, j_{-\mathbf{k}''\beta}(\tau'-\tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)} \\
&\quad - \frac{i\mathrm{k}_\beta''}{k'} \langle [\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau'-\tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}\, \Theta(\tau''-\tau') \\
&\quad - \frac{i\mathrm{k}_\beta'}{k''} \langle [\rho_{-\mathbf{k}''}(0), j_{-\mathbf{k}'\beta}(\tau''-\tau')]\, j_{\mathbf{k}\gamma}(\tau'')\rangle^{(0)}\, \Theta(\tau'-\tau'')\}
\end{aligned} \tag{25}$$

where the $\langle\cdots\rangle^{(0)}$ brackets denote ensemble-averaging with respect to $\Omega^{(0)}$ and $\mathbf{k}'' = \mathbf{k} - \mathbf{k}'$ in virtue of the homogeneity of the system.

2.3. Dynamical Fluctuation-Dissipation Theorems

The average second-order current density response is connected to the external driving fields $\hat{\mathbf{E}}$ through the quadratic *external* conductivity

$$\hat{\sigma}^{(2)}_{\gamma\mu\nu}(\mathbf{k}', \mathbf{k}''; \tau', \tau'')$$

defined by Ohm's law[5]:

$$\langle j_{\mathbf{k}\gamma}(t)\rangle^{(2)}$$
$$= (1/V)\sum_{\mathbf{k}'}\int_{-\infty}^{\infty} d\tau' \int_{-\infty}^{\infty} d\tau''\, \hat{\sigma}^{(2)}_{\gamma\mu\nu}(\mathbf{k}', \mathbf{k}''; \tau', \tau'')\, \hat{E}_{\mathbf{k}'\mu}(t - \tau')\, \hat{E}_{\mathbf{k}''\nu}(t - \tau''),$$
$$\mathbf{k}'' = \mathbf{k} - \mathbf{k}' \quad (26)$$

Comparison of Eqs. (25) and (26) then leads to the primitive form of the quadratic theorem (FDT):

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'')$$
$$= (\beta/2V)\,\Theta(\tau')\,\Theta(\tau'')\,\mathrm{k}_\gamma\{\beta \mathrm{k}_\alpha'\mathrm{k}_\beta''\langle j_{-\mathbf{k}'\alpha}(0)\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}$$
$$- (i\mathrm{k}_\beta''/k')\langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau' - \tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}\,\Theta(\tau'' - \tau')$$
$$- (i\mathrm{k}_\beta'/k'')\langle[\rho_{-\mathbf{k}''}(0), j_{-\mathbf{k}'\beta}(\tau'' - \tau')]\, j_{\mathbf{k}\gamma}(\tau'')\rangle^{(0)}\,\Theta(\tau' - \tau'')\} \quad (27a)$$

where again $\mathbf{k}'' = \mathbf{k} - \mathbf{k}'$ and where the scalar conductivity

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'') = \mathrm{k}_\gamma \mathrm{k}_\mu' \mathrm{k}_\nu'' \hat{\sigma}^{(2)}_{\gamma\mu\nu}(\mathbf{k}', \mathbf{k}''; \tau', \tau'') \quad (27b)$$

obviously projects out the longitudinal (with respect to $\mathbf{k}$) response to the longitudinal excitations $\hat{E}_{\mathbf{k}'\mu}\hat{E}_{\mathbf{k}''\nu}$. The l.h.s. of (27a) is manifestly symmetric in prime-double-prime interchange [were it not, only the symmetric projection would enter in (26), which thus could be redefined as $\hat{\sigma}^{(2)}$]. So is the rhs thanks to the manipulation performed earlier.

An alternative, more compact form of (27a) can be derived with the aid of a Poisson bracket relation which is exhibited in Appendix A:

$$\langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau' - \tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}$$
$$= -i\beta k_\alpha'\langle j_{-\mathbf{k}'\beta}(0)\, j_{-\mathbf{k}''\gamma}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}$$
$$- \langle[\rho_{-\mathbf{k}'}(0), j_{\mathbf{k}\gamma}(\tau')]\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\rangle^{(0)} \quad (28)$$

Noting also the counterpart of (28) formed from prime–double-prime interchange, the transformed form of (27a) becomes

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'')$$
$$= \Theta(\tau')\Theta(\tau'')(i\beta/2V)\mathrm{k}_\gamma\{(\mathrm{k}_\beta''/k')\langle[\rho_{-\mathbf{k}'}(0), j_{\mathbf{k}\gamma}(\tau')]\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\rangle^{(0)}\,\Theta(\tau'' - \tau')$$
$$+ (\mathrm{k}_\beta'/k'')\langle[\rho_{-\mathbf{k}''}(0), j_{\mathbf{k}\gamma}(\tau'')]\, j_{-\mathbf{k}'\beta}(\tau'' - \tau')\rangle^{(0)}\,\Theta(\tau' - \tau'')\} \quad (29)$$

Invariance with respect to space reflection requires that

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'') = \hat{\sigma}^{(2)}(-\mathbf{k}', -\mathbf{k}'', \tau', \tau'') \quad (30)$$

with similar relations for the longitudinal projections of the correlations of (27a) and (29). The usefulness of this symmetry rule will soon become evident.

Our next objective is to eliminate the unwieldy Poisson bracket terms entirely from the theorem and derive a relation for the triplet current correlation alone. In order to accomplish this, we first evaluate FDT relations for $\hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; \tau'' - \tau', \tau'')$ and $\hat{\sigma}^{(2)}(-\mathbf{k}, \mathbf{k}'', \tau', \tau' - \tau'')$. Thus upon noting that a typical correlation term containing ρ undergoes a change in sign under time reversal, e.g.,

$$\langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau'' - \tau')]\, j_{\mathbf{k}\gamma}(-\tau')\rangle^{(0)} = -\langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau' - \tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}$$

one can show that

$$\begin{aligned}
&\hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; \tau'' - \tau', \tau'') + \hat{\sigma}^{(2)}(-\mathbf{k}, \mathbf{k}''; \tau', \tau' - \tau'') \\
&= -\frac{i\beta}{2V}\, \mathrm{k}_\gamma \left\{ \frac{\mathrm{k}_\beta''}{k'} \langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau' - \tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}\, \Theta(\tau')\, \Theta(\tau'')\, \Theta(\tau'' - \tau') \right. \\
&\quad \left. + \frac{\mathrm{k}_\beta'}{k''} \langle[\rho_{-\mathbf{k}''}(0), j_{-\mathbf{k}'\beta}(\tau'' - \tau')]\, j_{\mathbf{k}\gamma}(\tau'')\rangle^{(0)}\, \Theta(\tau')\, \Theta(\tau'')\, \Theta(\tau' - \tau'') \right\} \\
&\quad - \frac{i\beta}{2V} \frac{\mathrm{k}_\beta' \mathrm{k}_\gamma''}{k} \{\langle[\rho_{\mathbf{k}}(0), j_{-\mathbf{k}''\gamma}(\tau'')]\, j_{-\mathbf{k}'\beta}(\tau')\rangle^{(0)}\, \Theta(-\tau')\, \Theta(\tau'')\, \Theta(\tau'' - \tau') \\
&\quad + \langle[\rho_{\mathbf{k}}(0), j_{-\mathbf{k}'\beta}(\tau')]\, j_{-\mathbf{k}''\gamma}(\tau'')\rangle^{(0)}\, \Theta(\tau')\, \Theta(-\tau'')\, \Theta(\tau' - \tau'')\}
\end{aligned} \tag{31}$$

so that the combination of Eqs. (27a) and (31) gives

$$\begin{aligned}
&\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'') - \hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; \tau'' - \tau', \tau'') - \hat{\sigma}^{(2)}(-\mathbf{k}, \mathbf{k}''; \tau', \tau' - \tau'') \\
&= \frac{\beta^2}{2V}\, \mathrm{k}_\alpha' \mathrm{k}_\beta'' \mathrm{k}_\gamma \langle j_{-\mathbf{k}'\alpha}(0)\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}\, \Theta(\tau')\, \Theta(\tau'') \\
&\quad + \frac{i\beta}{2V} \frac{\mathrm{k}_\beta' \mathrm{k}_\gamma''}{k} \{\langle[\rho_{\mathbf{k}}(0), j_{-\mathbf{k}''\gamma}(\tau'')]\, j_{-\mathbf{k}'\beta}(\tau')\rangle^{(0)}\, \Theta(-\tau')\, \Theta(\tau'')\, \Theta(\tau'' - \tau') \\
&\quad + \langle[\rho_{\mathbf{k}}(0), j_{-\mathbf{k}'\beta}(\tau')]\, j_{-\mathbf{k}''\gamma}(\tau'')\rangle^{(0)}\, \Theta(\tau')\, \Theta(-\tau'')\, \Theta(\tau' - \tau'')\}
\end{aligned} \tag{32}$$

Successive multiplication of (32) by $\Theta(\tau')$ and $\Theta(\tau'')$ then serves to project out only those terms which are nonzero for $\tau' > 0$, $\tau'' > 0$, namely

$$\begin{aligned}
&\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \tau', \tau'') - \hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; \tau'' - \tau', \tau'')\, \Theta(\tau') \\
&\quad - \hat{\sigma}^{(2)}(-\mathbf{k}, \mathbf{k}''; \tau', \tau' - \tau'')\, \Theta(\tau'') \\
&= -\frac{\beta^2}{2V}\, \mathrm{k}_\alpha \mathrm{k}_\beta' \mathrm{k}_\gamma'' \langle j_{\mathbf{k}\alpha}(0)\, j_{-\mathbf{k}'\beta}(\tau')\, j_{-\mathbf{k}''\gamma}(\tau'')\rangle^{(0)} \Theta(\tau')\, \Theta(\tau''), \\
&\qquad \mathbf{k}'' = \mathbf{k} - \mathbf{k}'
\end{aligned} \tag{33}$$

This is now the desired result which, however, can be cast into a more useful form by taking the temporal Fourier transform and introducing

$$\begin{aligned}\Psi(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') &- \int_{-\infty}^{\infty} d\mu\, \delta_+(\omega' + \mu)\, \hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; \mu, \omega'' - \mu) \\ &- \int_{-\infty}^{\infty} d\mu\, \delta_+(\omega'' + \mu)\, \hat{\sigma}^{(2)}(-\mathbf{k}, \mathbf{k}''; \omega' - \mu, \mu)\end{aligned} \tag{34}$$

$$Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \mathrm{k}_\alpha \mathrm{k}_\beta' \mathrm{k}_\gamma'' Q_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{35}$$

$$\begin{aligned}&Q_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')\, \delta(\omega' + \omega'' - \omega) \\ &\quad = (1/4\pi V)\langle j_{\mathbf{k}\alpha}(-\omega)\, j_{-\mathbf{k}'\beta}(\omega')\, j_{-\mathbf{k}''\gamma}(\omega'')\rangle^{(0)}\end{aligned} \tag{36}$$

with $\delta_\pm(\omega) = \frac{1}{2}\delta(\omega) \pm (i/2\pi)\, P(1/\omega)$, allowing us to write

$$\begin{aligned}&\Psi(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ &\quad = -\beta^2 \int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu\, \delta_+(\omega' - \mu)\, \delta_+(\omega'' - \nu)\, Q(\mathbf{k}', \mathbf{k}''; \mu, \nu)\end{aligned} \tag{37}$$

We observe that (27b), (30), and the reality condition

$$\hat{\sigma}^{(2)}_{\gamma\mu\nu}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \hat{\sigma}^{(2)*}_{\gamma\mu\nu}(-\mathbf{k}', -\mathbf{k}''; -\omega', -\omega'')$$

combine to give

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = -\hat{\sigma}^{(2)*}(\mathbf{k}', \mathbf{k}''; -\omega', -\omega'') \tag{38}$$

Clearly, the dynamical FDT (37) satisfies this last parity rule.

We note here that the real part of $\hat{\sigma}^{(2)}$ has odd parity with respect to simultaneous sign reversal of ω' and ω''. This, together with the fact that the average current density is a bounded response to the bounded driving $\hat{\mathbf{E}}$ field, assures us that the real part of $\hat{\sigma}^{(2)}$ is identically zero in the dc limit $\omega' = \omega'' = 0$. It follows from symmetry considerations [see Ref. 4(b), e.g.] that the corresponding wavevector- and frequency-independent quadratic conductivity is identically zero. Their result is compatible with ours if one exercises care by going first to the zero-freqeuncy limit and then to the zero-wavevector limit [see Eqs. (61) and (B.15) for the explicit k-dependence].

In the sequel, the superscript$^{(2)}$ will be omitted most of the time without any danger of confusion.

A final transformation which justified the nomenclature in the FDT is obtained by taking the real (dissipative) part of Ψ. Then, we observe from (36) that the current correlation tensor $Q_{\alpha\beta\gamma}$ undergoes no net change in sign under simultaneous microscopic time reversal ($\omega' \to -\omega'$, $\omega'' \to -\omega''$, $\omega \to -\omega$) and space inversion ($\mathbf{k}' \to -\mathbf{k}'$, $\mathbf{k}'' \to -\mathbf{k}''$, $\mathbf{k} \to -\mathbf{k}$), so that $Q_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')$ and, consequently, $Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'')$ must be real. The real part of (37) is therefore[6]

$$\Psi'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = -\frac{\beta^2}{4} Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'') + \frac{\beta^2}{4\pi^2} PP \int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu \frac{Q(\mathbf{k}', \mathbf{k}''; \mu, \nu)}{(\omega' - \mu)(\omega'' - \nu)} \tag{39}$$

and it is hereafter understood that $\mathbf{k}'' = \mathbf{k} - \mathbf{k}'$.

2.4. Solution of the Integral Equation

Equation (39) constitutes the closest analog of the linear FDT: both relate the dissipative part of a response function to the dynamical equilibrium correlations of microscopic current densities.[7] The important difference, however, is that in the nonlinear case, Q appears in combination with its double Hilbert transform. Therefore, in order to obtain an explicit expression for Q in terms of the response function, the resulting integral equation has to be solved. This is the purpose of the present section. First, we note that the current correlation function is invariant—up to a sign change—with respect to rotation on the triangle formed by the "four-vectors" $(\mathbf{k}, \omega)$, $(\mathbf{k}', \omega')$, $(\mathbf{k}'', \omega'')$, i.e.,

$$Q(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = -Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{40a}$$

$$= Q(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') \tag{40b}$$

The Hilbert transform operation, however, violates this invariance; thus the function Ψ' does not satisfy a similar symmetry rule.

One can, however, form the symmetrized combination

$$\tilde{\Psi}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \tfrac{1}{3}\{\Psi'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') - \Psi'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) - \Psi'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')\} \tag{41}$$

which does possess the triangle antisymmetry exhibited by Q.

[6] Prime and double-prime symbols are used (for example, as follows: $\Psi' = \mathrm{Re}\,\Psi$, $\Psi'' = \mathrm{Im}\,\Psi$) to denote real and imaginary parts.

[7] For the linear result, see, for example, Ref. 2(e), Eq. (117).

Proceeding, we have from (34) that

$$\begin{aligned}\Psi'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = {} & \hat{\sigma}'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') - \frac{1}{2}\,\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) \\ & - \frac{1}{2}\,\hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\omega' + \mu}\,\hat{\sigma}''(\mathbf{k}', -\mathbf{k}; \mu, \omega'' - \mu) \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\omega'' + \mu}\,\hat{\sigma}''(-\mathbf{k}, \mathbf{k}''; \omega' - \mu, \mu)\end{aligned} \tag{42}$$

whence and from (42) it can be shown that

$$\begin{aligned}\Psi'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = {} & \hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) - \frac{1}{2}\,\hat{\sigma}''(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ & + \frac{1}{2}\,\hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\mu - \omega'}\,\hat{\sigma}''(\mathbf{k}', \mathbf{k}''; \mu, \omega - \mu) \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\omega'' + \mu}\,\hat{\sigma}''(-\mathbf{k}, \mathbf{k}''; \omega' - \mu, \mu)\end{aligned} \tag{43}$$

and

$$\begin{aligned}\Psi'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') = {} & \hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') + \frac{1}{2}\,\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; \omega, -\omega') \\ & - \frac{1}{2}\,\hat{\sigma}'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\omega' + \mu}\,\hat{\sigma}''(\mathbf{k}', -\mathbf{k}; \mu, \omega'' - \mu) \\ & + \frac{1}{2\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\mu - \omega''}\,\hat{\sigma}''(\mathbf{k}', \mathbf{k}''; \omega - \mu, \mu)\end{aligned} \tag{44}$$

Then, upon combining (42)–(44) according to (41), one readily obtains

$$\begin{aligned}& \tilde{\Psi}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ & \quad = \tfrac{2}{3}\{\hat{\sigma}'(\mathbf{k}', \mathbf{k}''; \omega', \omega'') - \hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) - \hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')\}\end{aligned} \tag{45}$$

Now, a triangle-antisymmetrized FDT can be derived from Eq. (39) by first observing from (39) and (40a) that

$$\begin{aligned}\Psi'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) &= -\frac{\beta^2}{4} Q(\mathbf{k}', -\mathbf{k}; -\omega', \omega)\\ &\quad - \frac{\beta^2}{4\pi^2} PP \int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu\, \frac{Q(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu)}{(\omega' - \mu)(\omega - \mu - \nu)}\\ &= -\frac{\beta^2}{4} Q(\mathbf{k}', -\mathbf{k}; -\omega', \omega)\\ &\quad + \frac{\beta^2}{4\pi^2} PP \int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu\, \frac{Q(\mathbf{k}', \mathbf{k}''; \mu, \nu)}{(\omega' - \mu)(\omega - \mu - \nu)}\end{aligned} \tag{46}$$

Similarly, one has from (39) and (40b) that

$$\begin{aligned}\Psi'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') &= -\frac{\beta^2}{4} Q(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')\\ &\quad + \frac{\beta^2}{4\pi^2} PP \int_{-\infty}^{\infty} d\nu \int_{-\infty}^{\infty} d\mu\, \frac{Q(\mathbf{k}', \mathbf{k}''; \mu, \nu)}{(\omega'' - \nu)(\omega - \mu - \nu)}\end{aligned} \tag{47}$$

In order to bring (47) to a form comparable with (46), the Poincaré–Bertrand theorem[6] has to be invoked, leading to

$$\begin{aligned}\Psi'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') &= -\frac{\beta^2}{4} Q(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'') + \frac{\beta^2}{4} Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'')\\ &\quad + \frac{\beta^2}{4\pi^2} PP \int_{-\infty}^{\infty} d\mu \int_{-\infty}^{\infty} d\nu\, \frac{Q(\mathbf{k}', \mathbf{k}''; \mu, \nu)}{(\omega'' - \nu)(\omega - \mu - \nu)}\end{aligned} \tag{47a}$$

Thus the combination of Eqs. (39), (46), and (47) according to (41), with subsequent use of the parity relations (40), yields the desired relation:

$$\tilde{\Psi}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = -\tfrac{1}{3}\beta^2 Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{48}$$

in a form in which both sides are manifestly triangle-antisymmetric. [We note, however, that expressions figuring in Eq. (48), unlike those in (39), are not causal with respect to (ω', ω'').]

The path we followed in the above derivation circumvents, by exploiting the appropriate symmetry properties, the problem of actually solving the integral equation (39). We now present an alternative method for the derivation of (48), based on the explicit inversion of (39). Consider the "minus" and "plus" projections of $\hat{\sigma}^{(2)}(\mathbf{k}', -\mathbf{k}; -\omega', \omega)$ with respect to ω':

$$\hat{\sigma}_{-}(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \int_{-\infty}^{\infty} d\mu\, \delta_{-}(\omega' - \mu)\, \hat{\sigma}(\mathbf{k}', -\mathbf{k}; -\mu, \omega'' + \mu)$$

$$\hat{\sigma}_{+}(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \int_{-\infty}^{\infty} d\mu\, \delta_{+}(\omega' - \mu)\, \hat{\sigma}(\mathbf{k}', -\mathbf{k}; -\mu, \omega'' + \mu)$$

so that

$$\hat{\sigma}_{-}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \tfrac{1}{2}\{\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) + \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega)\} \quad (49)$$

where

$$\textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \dfrac{1}{\pi} P \displaystyle\int_{-\infty}^{\infty} \frac{d\mu}{\omega' + \mu}\, \hat{\sigma}''(\mathbf{k}', -\mathbf{k}; \mu, \omega'' - \mu) \quad (50)$$

Similarly,

$$\hat{\sigma}_{+}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \tfrac{1}{2}\{\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) - \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega)\} \quad (51)$$

Since $\hat{\sigma}_{-}(\mathbf{k}', -\mathbf{k}; -\omega', \omega)$ is a minus function of ω' and a plus function of ω'', it follows that

$$\hat{\sigma}_{-}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \mathscr{H}[\omega', \mu]\, \mathscr{H}[\omega'', \nu]\, \hat{\sigma}_{-}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu) \quad (52)$$

where, e.g.,

$$\mathscr{H}[\omega', \mu] = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{d\mu}{\omega' - \mu} (\cdots)$$

is the Hilbert transform operator. On the other hand, $\hat{\sigma}_{+}(\mathbf{k}', -\mathbf{k}; -\omega', \omega)$ is a plus function of both ω' and ω'', so that

$$\hat{\sigma}_{+}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) = -\mathscr{H}[\omega', \mu]\, \mathscr{H}[\omega'', \nu]\, \hat{\sigma}_{+}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu) \quad (53)$$

From (49), (51)–(53), one then obtains

$$\begin{aligned}&\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) + \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega) \\ &\quad = \mathscr{H}[\omega', \mu]\, \mathscr{H}[\omega'', \nu]\{\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu) + \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu)\}\end{aligned} \quad (54)$$

$$\begin{aligned}&\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) - \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega) \\ &\quad = -\mathscr{H}[\omega', \mu]\, \mathscr{H}[\omega'', \nu]\{\hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu) - \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu)\}\end{aligned} \quad (55)$$

Thus

$$\textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega) = \mathscr{H}[\omega', \mu]\, \mathscr{H}[\omega'', \nu]\, \hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu) \quad (56)$$

which, of course, follows directly from (50) since

$$\hat{\sigma}''(\mathbf{k}', -\mathbf{k}; -\mu, \omega'' + \mu) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{d\nu}{\omega'' - \nu}\, \hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\mu, \mu + \nu)$$

One then derives similar relations for $\hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')$. Then, let

$$J = \hat{\sigma}'(\mathbf{k}', -\mathbf{k}; -\omega', \omega) + \hat{\sigma}'(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')$$

$$K = \textstyle\sum' (\mathbf{k}', -\mathbf{k}; -\omega', \omega) + \sum' (-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')$$

so that in a short-hand notation

$$(1 - \mathscr{H}\mathscr{H})(J + K) = 0 \quad (57)$$

$$(1 + \mathscr{H}\mathscr{H})(J - K) = 0 \quad (58)$$

Exploiting the fact that Ψ is a plus function of both ω' and ω'', we also have

$$(1 - \mathscr{H}\mathscr{H})\, \Psi' = 2\Psi'$$

Thus the general solution of Eq. (39),

$$-\tfrac{1}{4}\beta^2(1 - \mathscr{H}\mathscr{H})\, Q = \Psi'$$

certainly can be cast in the form

$$\beta^2 Q = -2\Psi' + a(J + K) \quad (59)$$

where a is an undetermined constant. This choice, however, becomes unambiguous by demanding the triangle antisymmetry of Q to be satisfied by the solution. Thus Ψ' should appear in its symmetrized form, $\tilde{\Psi}$, only. However, it is readily demonstrated that

$$\tilde{\Psi} = \tfrac{2}{3}\{\Psi' - \tfrac{1}{2}(J + K)\} \quad (60)$$

which renders $a = 1$. Then, (59) in conjunction with (60) is identical to (48).

2.5. FDT in Terms of Nonlinear Polarizabilities

The FDT (48) can be cast in yet another form which features *internal* response functions on the l.h.s. rather than the inconvenient *external* conductivities. Thus, we introduce here the linear and nonlinear dielectric tensors $\epsilon_{\alpha\beta}(\mathbf{k}, \omega)$ and $\epsilon^{(2)}_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')$ defined (for a homogeneous and stationary system) by the constitutive relation

$$\begin{aligned} D_\alpha(\mathbf{k}, \omega) &= \epsilon_{\alpha\beta}(\mathbf{k}, \omega)\langle E_\beta(\mathbf{k}, \omega)\rangle^{(1)} + \epsilon_{\alpha\beta}(\mathbf{k}, \omega)\langle E_\beta(\mathbf{k}, \omega)\rangle^{(2)} \\ &+ \frac{1}{2\pi V}\sum_{\mathbf{k}'}\int_{-\infty}^{\infty} d\omega' \, \epsilon^{(2)}_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ &\times \langle E_\beta(\mathbf{k}', \omega')\rangle^{(1)} \langle E_\gamma(\mathbf{k}'', \omega'')\rangle^{(1)} + \cdots \\ \mathbf{k}'' &= \mathbf{k} - \mathbf{k}', \qquad \omega'' = \omega - \omega' \end{aligned}$$

which connects the electric induction $\mathbf{D}(\mathbf{k}, \omega)$ to the *total* average electric field $\langle \mathbf{E}(\mathbf{k}, \omega)\rangle$. The appropriate longitudinal projections are then given by

$$\epsilon(\mathbf{k}, \omega) = \mathrm{k}_\alpha \mathrm{k}_\beta \epsilon_{\alpha\beta}(\mathbf{k}, \omega)$$

$$\epsilon^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \mathrm{k}_\alpha \mathrm{k}_\beta' \mathrm{k}_\gamma'' \epsilon^{(2)}_{\alpha\beta\gamma}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')$$

Thus from our earlier nonlinear electrodynamic study,[5] it was found that

$$\hat{\sigma}^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = i\epsilon_0(\omega' + \omega'')\, \eta^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{61}$$

where

$$\eta^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \frac{\epsilon^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\epsilon(\mathbf{k}', \omega')\, \epsilon(\mathbf{k}'', \omega'')\, \epsilon(\mathbf{k}' + \mathbf{k}'', \omega' + \omega'')} \tag{62}$$

and $(k/k'k'')\,\eta^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')$ is the response function which connects the average second-order induced charge density response $\langle \rho(\mathbf{k}, \omega)\rangle^{(2)}$ to the external charge perturbations $\hat{\rho}(\mathbf{k}', \omega')\,\hat{\rho}(\mathbf{k}'', \omega'')$, namely

$$\langle \rho(\mathbf{k}, \omega)\rangle^{(2)} = -\frac{i}{\epsilon_0 V}\sum_{\mathbf{k}'}\left(\frac{k}{k'k''}\right)\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\, \eta^{(2)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')\, \hat{\rho}(\mathbf{k}', \omega')\, \hat{\rho}(\mathbf{k}'', \omega'') \tag{63}$$

Concerning the r.h.s. of (48), we introduce here the dynamical charge density correlation function P defined by

$$P(\mathbf{k}', \mathbf{k}''; \omega', \omega'')\, \delta(\omega' + \omega'' - \omega) = (1/4\pi V)\langle \rho_{\mathbf{k}}(-\omega)\, \rho_{-\mathbf{k}'}(\omega')\, \rho_{-\mathbf{k}''}(\omega'')\rangle^{(0)} \tag{64}$$

One can then show that

$$Q(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = -(\omega\omega'\omega''/kk'k'')\, P(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{65}$$

Thus Eqs. (45), (48), (61), and (65) combine to yield the following alternative form of the FDT:

$$\begin{aligned} &\frac{\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} - \frac{\eta''(\mathbf{k}', -\mathbf{k}; -\omega', \omega)}{\omega\omega'} - \frac{\eta''(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')}{\omega\omega''} \\ &\quad = \frac{\beta^2}{2\epsilon_0 kk'k''}\, P(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \end{aligned} \tag{66}$$

3. STATIC FLUCTUATION-DISSIPATION THEOREM AND EQUILIBRIUM STATISTICAL MECHANICS

3.1. Static FDT

One of the most important ramifications of the conventional FDT is its static form, which ultimately links the equilibrium pair correlation function with the static dielectric function. In the present context, a similar relation will provide a connection between the equilibrium three-particle correlation function and the static quadratic response function. This section is devoted to the derivation of this relation.

Our starting point is Eq. (66) which, upon integration over ω' and ω'',

$$\begin{aligned}
&\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega''}{2\pi} \frac{\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} \\
&\quad - \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega''}{2\pi} \frac{\eta''(\mathbf{k}', -\mathbf{k}; -\omega', \omega)}{\omega\omega'} \\
&\quad - \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega''}{2\pi} \frac{\eta''(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')}{\omega\omega''} \\
&\quad = \frac{\beta^2}{2\epsilon_0 k k' k''} P(\mathbf{k}', \mathbf{k}''; t = 0, t = 0)
\end{aligned} \tag{67}$$

yields an expression for the equal-time charge density correlation function. However, the lhs of (67) can be considerably simplified. To begin, we let

$$\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \eta''(\mathbf{k}', \mathbf{k}'') + \bar{\eta}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{68}$$

where $\eta''(\mathbf{k}', \mathbf{k}'') = \eta''(\mathbf{k}', \mathbf{k}''; \omega' = 0, \omega'' = 0)$. Putting (68) back into the dynamical FDT (66) and going to the static limit ($\omega' = \omega'' = 0$), one obtains

$$\begin{aligned}
&\lim_{\omega'\to 0} \lim_{\omega''\to 0} \left\{ \frac{\eta''(\mathbf{k}', \mathbf{k}'') - \eta''(\mathbf{k}', -\mathbf{k})}{\omega'} + \frac{\eta''(\mathbf{k}', \mathbf{k}'') - \eta''(-\mathbf{k}, \mathbf{k}'')}{\omega''} \right\} \\
&\quad = - \lim_{\omega'\to 0} \lim_{\omega''\to 0} \omega \left\{ \frac{\bar{\eta}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} \right. \\
&\qquad \left. - \frac{\bar{\eta}(\mathbf{k}', -\mathbf{k}; -\omega', \omega)}{\omega\omega'} - \frac{\bar{\eta}(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')}{\omega\omega''} \right\} \\
&\quad + \lim_{\omega\to 0} \frac{\omega\beta^2}{4\epsilon_0 k k' k''} P(\mathbf{k}', \mathbf{k}''; \omega' = 0, \omega'' = 0) = 0
\end{aligned} \tag{69}$$

The vanishing of the r.h.s. of (69) is assured by noting that for ω', ω'' small, one has, for symmetry reasons,[8]

$$\begin{aligned}\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'') &= \eta''(\mathbf{k}', \mathbf{k}'') + (\partial^2\eta''/\partial\omega'\,\partial\omega'')_{0,0}\,\omega', \omega'' \\ &\quad + \tfrac{1}{2}(\partial^2\eta''/\partial\omega'^2)_{0,0}\,\omega'^2 \\ &\quad + \tfrac{1}{2}(\partial^2\eta''/\partial\omega''^2)_{0,0}\,\omega''^2 + \cdots \qquad (70)\end{aligned}$$

so that the limit

$$\begin{aligned}&\lim_{\omega'\to 0}\lim_{\omega''\to 0} \frac{\bar{\eta}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega', \omega''} \\ &= \left(\frac{\partial^2\eta}{\partial\omega'\,\partial\omega''}\right)_{0,0} + \lim_{\omega'\to 0}\lim_{\omega''\to 0} \frac{1}{2}\left\{\left(\frac{\partial^2\eta''}{\partial\omega'^2}\right)_{0,0}\frac{\omega'}{\omega''} + \left(\frac{\partial^2\eta''}{\partial\omega''^2}\right)_{0,0}\frac{\omega''}{\omega'}\right\}\end{aligned}$$

must be bounded. $P(\mathbf{k}', \mathbf{k}''; \omega' = 0, \omega'' = 0)$ is also bounded for physical reasons. Thus, except for the trivial case $\eta'' = 0$, the vanishing of the lhs is assured if, and only if,

$$\eta''(\mathbf{k}', \mathbf{k}'') = \eta''(\mathbf{k}', -\mathbf{k}) = \eta''(-\mathbf{k}, \mathbf{k}'') \qquad (71)$$

We note that the Kalman–Pomeau expression for η'' in the Vlasov approximation,[7] which is reproduced in Appendix B [see Eq. (B.15)], indeed satisfies this last symmetry rule.

Next, we turn to the calculation of the l.h.s. of (67). Let

$$\begin{aligned}&PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} \\ &- PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(\mathbf{k}', -\mathbf{k}; -\omega', \omega)}{\omega\omega'} \\ &- PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')}{\omega\omega''} = I \qquad (72)\end{aligned}$$

By repeated application of the Kramers-Kronig formula, one obtains

$$PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} = -\frac{1}{4}\,\eta''(\mathbf{k}', \mathbf{k}'') \qquad (73a)$$

and

$$\begin{aligned}&PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(\mathbf{k}', -\mathbf{k}; -\omega', \omega''+\omega')}{\omega'(\omega''+\omega')} \\ &- PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\,\frac{\eta''(\mathbf{k}', -\mathbf{k}; \omega', \omega''-\omega')}{\omega'(\omega''-\omega')} \\ &= \frac{1}{4}\,\eta''(\mathbf{k}', -\mathbf{k}) + \frac{1}{4}\,\eta''(\mathbf{k}', \mathbf{k}'') \qquad (73b)\end{aligned}$$

[8] Since $\eta''(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = \eta''(\mathbf{k}', \mathbf{k}''; -\omega', -\omega'')$ [see Eq. (38)], the expression (70) cannot contain terms like $\omega'(\partial\eta''/\partial\omega')_{0,0}$ and $\omega''(\partial\eta''/\partial\eta'')_{0,0}$.

Note, however, that the third integral of (72), as it stands, is not amenable to similar simplification. However, by first invoking the Poincaré-Bertrand theorem, one derives

$$PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\frac{\eta''(-\mathbf{k},\mathbf{k}'';\omega'+\omega'',-\omega'')}{\omega''(\omega'+\omega'')}$$
$$= -PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\frac{\eta''(-\mathbf{k},\mathbf{k}'';\omega'-\omega'',\omega'')}{\omega''(\omega'-\omega'')}$$
$$= -PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\frac{\eta''(-\mathbf{k},\mathbf{k}'';\omega'-\omega'',\omega'')}{\omega''(\omega'-\omega'')}$$
$$-\frac{1}{4}\eta''(-\mathbf{k},\mathbf{k}'') = 0 \tag{73c}$$

in virtue of the Kramers–Kronig formula it satisfies. Consequently, the combination of (73a)–(73c) according to (72) yields

$$I = -\tfrac{1}{2}\eta''(\mathbf{k}',\mathbf{k}'') \tag{74}$$

On the other hand, from (68), (71), and the definition (72) of I, one can write

$$I = PP\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\eta''(\mathbf{k}',\mathbf{k}'')\left\{\frac{1}{\omega'\omega''}-\frac{1}{\omega\omega'}-\frac{1}{\omega\omega''}\right\}$$
$$+\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\left\{\frac{\bar{\eta}(\mathbf{k}',\mathbf{k}'';\omega',\omega'')}{\omega'\omega''}\right.$$
$$\left.-\frac{\bar{\eta}(\mathbf{k}',-\mathbf{k};-\omega',\omega)}{\omega\omega'}-\frac{\bar{\eta}(-\mathbf{k},\mathbf{k}'';\omega,-\omega'')}{\omega\omega''}\right\}$$
$$=\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\eta''(\mathbf{k}',\mathbf{k}'')\left\{\frac{1}{\omega'\omega''}-\frac{1}{\omega\omega'}-\frac{1}{\omega\omega''}\right\}$$
$$+\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\left\{\frac{\bar{\eta}(\mathbf{k}',\mathbf{k}'';\omega',\omega'')}{\omega'\omega''}\right.$$
$$\left.-\frac{\bar{\eta}(\mathbf{k}',-\mathbf{k};-\omega',\omega)}{\omega\omega'}-\frac{\bar{\eta}(-\mathbf{k},\mathbf{k}'';\omega,-\omega'')}{\omega\omega''}\right\}$$
$$=\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega''}{2\pi}\left\{\frac{\eta''(\mathbf{k}',\mathbf{k}'';\omega',\omega'')}{\omega'\omega''}\right.$$
$$\left.-\frac{\eta''(\mathbf{k}',-\mathbf{k};-\omega',\omega)}{\omega\omega'}-\frac{\eta''(-\mathbf{k},\mathbf{k}'';\omega,-\omega'')}{\omega\omega''}\right\} \tag{75}$$

since, in any case,[9]

$$\frac{1}{\omega'\omega''} - \frac{1}{\omega\omega'} - \frac{1}{\omega\omega''} = 0$$

and terms like $\bar{\eta}(\omega', \omega'')/\omega'\omega''$ are nonsingular.

Thus from Eqs. (67), (74), and (75), one readily obtains the static FDT:

$$\eta''(\mathbf{k}', \mathbf{k}'') = (-\beta^2/\epsilon_0 kk'k'')\, P(\mathbf{k}', \mathbf{k}''; t = 0, t = 0) \qquad (76)$$

or

$$\langle \rho_{\mathbf{k}}\rho_{-\mathbf{k}'}\rho_{-\mathbf{k}''}\rangle_{t=0}^{(0)} = -(2V\epsilon_0 kk'k''/\beta^2)\, \eta''(\mathbf{k}', \mathbf{k}'')$$

It is an easy matter to generalize the previous study to a two-component plasma having N dynamical electrons (each of mass m and charge e) and N/Z dynamical ions (each of mess M and charge $-Ze$). Letting

$$R(\mathbf{r}, t) = -Ze \sum_{i=1}^{N/Z} \delta(\mathbf{r} - \mathbf{X}_i(t)) \qquad (77)$$

where $R(\mathbf{r}, t)$ is the ion microscopic charge density, one can show that the full static FDT is

$$\langle(\rho_{\mathbf{k}} + R_{\mathbf{k}})(\rho_{-\mathbf{k}'} + R_{-\mathbf{k}'})(\rho_{-\mathbf{k}''} + R_{-\mathbf{k}''})\rangle_{\tau=0}^{(0)} = -(2V\epsilon_0 kk'k''/\beta^2)\, \eta''(\mathbf{k}', \mathbf{k}'') \qquad (78)$$

where

$$\eta''(\mathbf{k}', \mathbf{k}'') = \eta_e''(\mathbf{k}', \mathbf{k}'') + \eta_i''(\mathbf{k}', \mathbf{k}'')$$

3.2. Dielectric Description of Pair and Triplet Correlation Functions

Our final task consists of expressing the lhs of (78) in terms of the well-known pair and triplet correlations [defined by (80) below] of equilibrium

[9] In the light of (71), Eq. (69) can also be written in the form

$$\eta''(\mathbf{k}', \mathbf{k}'') \lim_{\omega'\to 0} \lim_{\omega''\to 0} \left\{\frac{1}{\omega'\omega''} - \frac{1}{\omega\omega'} - \frac{1}{\omega\omega''}\right\}$$
$$= -\lim_{\omega'\to 0} \lim_{\omega''\to 0} \left\{\left(\frac{\bar{\eta}(\mathbf{k}', \mathbf{k}''; \omega', \omega'')}{\omega'\omega''} - \frac{\bar{\eta}(\mathbf{k}', -\mathbf{k}; -\omega', \omega)}{\omega\omega'} - \frac{\bar{\eta}(-\mathbf{k}, \mathbf{k}''; \omega, -\omega'')}{\omega\omega''}\right)\right.$$
$$\left. + \frac{\beta^2}{2V\epsilon_0 kk'k''} P(\mathbf{k}', \mathbf{k}''; \omega' = 0, \omega'' = 0)\right\}$$

Clearly, the boundedness of the rhs requires that the lhs vanish identically.

statistical mechanics. We introduce here a variety of one-, two-, and three-particle distribution functions, e.g.,

$$F_e(\mathbf{p}_1) = N \underset{N-1+(N/Z)}{\int \cdots \int} \Omega^{(0)}\, d2 \cdots dN\, dI \cdots dN/Z$$

$$F_i(\mathbf{P}_1) = (N/Z) \underset{N+(N/Z)-1}{\int \cdots \int} \Omega^{(0)}\, d1 \cdots dN\, dII \cdots dN/Z$$

$$G_{ee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2) = N(N-1) \underset{N-2+(N/Z)}{\int \cdots \int} \Omega^{(0)}\, d3 \cdots dN\, dI \cdots dN/Z$$

$$G_{ei}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{X}_1, \mathbf{P}_1) = (N^2/Z) \underset{N-2+(N/Z)}{\int \cdots \int} \Omega^{(0)}\, d2 \cdots dN\, dII \cdots dN/Z,$$

$$H_{eee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2; \mathbf{x}_3, \mathbf{p}_3) = N(N-1)(N-2) \underset{N-3+(N/Z)}{\int \cdots \int} \Omega^{(0)}\, d4 \cdots dN\, dI \cdots dN/Z \tag{79}$$

etc., where, in an obvious notation,

$$d2 = d^3\mathbf{x}_2\, d^3\mathbf{p}_2, \qquad dIII = d^3\mathbf{X}_3\, d^3\mathbf{P}_3, \qquad \text{etc.}$$

so that, e.g.,

$$\underset{N-1+(N/Z)}{\int \cdots \int}$$

actually implies $6[N - 1 + (N/Z)]$ integrations. The pair and triplet correlations (Ursell functions) $g_{ee}(12)[\equiv g_{ee}(|\mathbf{x}_1 - \mathbf{x}_2|)]$, $g_{ei}(1, I)$, $h_{eee}(1, 2, 3)$, $h_{eei}(1, 2, I)$, etc., which are of interest here, are then defined through relations like

$$G_{ee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2) = F_e(\mathbf{p}_1)\, F_e(\mathbf{p}_2)\{1 + g_{ee}(12)\}$$

$$G_{ei}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{X}_1, \mathbf{P}_1) = F_e(\mathbf{p}_1)\, F_i(\mathbf{P}_1)\{1 + g_{ei}(1, I)\}$$

$$H_{eee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2; \mathbf{x}_3, \mathbf{p}_3) = F_e(\mathbf{p}_1)\, F_e(\mathbf{p}_2)\, F_e(\mathbf{p}_3)\{1 + g_{ee}(12) + g_{ee}(13) + g_{ee}(23) + h_{eee}(123)\} \tag{80}$$

$$H_{eei}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2; \mathbf{X}_1, \mathbf{P}_1) = F_e(\mathbf{p}_1)\, F_e(\mathbf{p}_2)\, F_i(\mathbf{P}_1)\{1 + g_{ee}(1, 2) + g_{ei}(1, I) + g_{ei}(2, I) + h_{eei}(1, 2, I)\}$$

etc. We now proceed to the calculation of the lhs of (78). Considering first the pure electron correlation, we have

$$
\begin{aligned}
&\langle \rho_{\mathbf{k}} \rho_{-\mathbf{k}'} \rho_{-\mathbf{k}''} \rangle_{t\,0}^{(0)} \\
&\quad = e^3 \sum_{i,j,s} \langle \exp(-i\mathbf{k}\cdot\mathbf{x}_i) \exp(i\mathbf{k}'\cdot\mathbf{x}_j) \exp(i\mathbf{k}''\cdot\mathbf{x}_s)\rangle^{(0)} \\
&\quad = Ne^3 + e^3 \sum_{\substack{i,j\\ i\neq j}} \{\langle \exp[i\mathbf{k}\cdot(\mathbf{x}_i - \mathbf{x}_j)]\rangle^{(0)} + \langle \exp[-i\mathbf{k}'\cdot(\mathbf{x}_i - \mathbf{x}_j)]\rangle^{(0)} \\
&\qquad + \langle \exp[-i\mathbf{k}''\cdot(\mathbf{x}_i - \mathbf{x}_j)]\rangle^{(0)}\} \\
&\qquad + e^3 \sum_{\substack{i,j,s\\ i\neq j\neq s\neq i}} \langle \exp(-i\mathbf{k}\cdot\mathbf{x}_i) \exp(i\mathbf{k}'\cdot\mathbf{x}_j) \exp(i\mathbf{k}''\cdot\mathbf{x}_s)\rangle^{(0)} \\
&\quad = Ne^2 + e^3 \iint d1\, d2\, G_{ee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2)\{\exp[i\mathbf{k}\cdot(\mathbf{x}_1 - \mathbf{x}_2)] \\
&\qquad + \exp[-i\mathbf{k}'\cdot(\mathbf{x}_1 - \mathbf{x}_2)] + \exp[-i\mathbf{k}''\cdot(\mathbf{x}_1 - \mathbf{x}_2)]\} \\
&\qquad + e^3 \iiint d1\, d2\, d3\, H_{eee}(\mathbf{x}_1, \mathbf{p}_1; \mathbf{x}_2, \mathbf{p}_2; \mathbf{x}_3, \mathbf{p}_3) \\
&\qquad \times \exp(-i\mathbf{k}\cdot\mathbf{x}_1) \exp(i\mathbf{k}'\cdot\mathbf{x}_2) \exp(i\mathbf{k}''\cdot\mathbf{x}_3)
\end{aligned} \tag{81}
$$

Then, upon eliminating G_{ee} and H_{eee} in favor of g_{ee} and h_{eee} through (80), Eq. (81) ultimately integrates to

$$
\begin{aligned}
\langle \rho_{\mathbf{k}} \rho_{-\mathbf{k}'} \rho_{-\mathbf{k}''} \rangle^{(0)} = {} & Ne^3 + N^2e^3(\delta_{\mathbf{k},0} + \delta_{\mathbf{k}',0} + \delta_{\mathbf{k}'',0}) + N^3e^3\delta_{\mathbf{k},0}\delta_{\mathbf{k}',0}\delta_{\mathbf{k}'',0} \\
& + nNe^3[g_{ee}(k) + g_{ee}(k') + g_{ee}(k'')] + nN^2e^3[g_{ee}(k')\,\delta_{\mathbf{k}',\mathbf{k}} \\
& + g_{ee}(k'')\,\delta_{\mathbf{k}',0} + g_{ee}(k')\,\delta_{\mathbf{k}',0}] + n^2Ne^3h_{eee}(\mathbf{k}', \mathbf{k}'')
\end{aligned} \tag{82a}
$$

where $g_{ee}(k)$ is the Fourier transform of $g_{ee}(|\mathbf{x}_1 - \mathbf{x}_2|)$ and n is the electron number density. Similarly, one finds that

$$
\begin{aligned}
&\langle R_{\mathbf{k}} R_{-\mathbf{k}'} R_{-\mathbf{k}''} \rangle^{(0)} \\
&\quad = -Z^2Ne^3 - ZN^2e^3(\delta_{\mathbf{k},0} + \delta_{\mathbf{k}',0} + \delta_{\mathbf{k}'',0}) \\
&\qquad - N^3e^3\delta_{\mathbf{k},0}\delta_{\mathbf{k}',0}\delta_{\mathbf{k}'',0} - ZnNe^3[g_{ii}(k) + g_{ii}(k') + g_{ii}(k'')] \\
&\qquad - nN^2e^3[g_{ii}(k')\,\delta_{k'',0} + g_{ii}(k'')\,\delta_{k',0} + g_{ii}(k'')\,\delta_{k,0}] - n^2Ne^3h_{iii}(\mathbf{k}', \mathbf{k}'')
\end{aligned} \tag{82b}
$$

and similar relations resulting from the interchange of $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{k}''$. Thus, the full ion-electron three-body density correlation function is obtained by adding equations like (82).

One ultimately obtains

$$\langle(\rho_{\mathbf{k}} + R_{\mathbf{k}})(\rho_{-\mathbf{k}'} + R_{-\mathbf{k}'})(\rho_{-\mathbf{k}''} + R_{-\mathbf{k}''})\rangle^{(0)}$$

$$= Ne^3(1 - Z^2) + nNe^3 \sum_{l=k,k',k''} \{[g_{ee}(l) - g_{ie}(l)] - Z[g_{ii}(l) - g_{ei}(l)]\}$$

$$+ n^2Ne^3h(\mathbf{k}', \mathbf{k}'') \tag{83}$$

where

$$h(\mathbf{k}', \mathbf{k}'') = h_{eee}(\mathbf{k}', \mathbf{k}'') - h_{eei}(\mathbf{k}', \mathbf{k}'') - h_{eie}(\mathbf{k}', \mathbf{k}'') - h_{iee}(\mathbf{k}', \mathbf{k}'')$$
$$+ h_{eii}(\mathbf{k}', \mathbf{k}'') + h_{iei}(\mathbf{k}', \mathbf{k}'') + h_{iie}(\mathbf{k}', \mathbf{k}'') - h_{iii}(\mathbf{k}', \mathbf{k}'') \tag{83a}$$

Now, from our earlier linear FDT study, it was found that the pair correlation function satisfies[2c]

$$n[g_{ee}(l) - g_{ei}(l)] - nZ[g_{ii}(l) - g_{ei}(l)]$$
$$= (\epsilon_0 l^2/\beta ne^2)\,\mathrm{Re}\{[\alpha_e(l) - Z\alpha_i(l)]/\epsilon(l)\} - (1 - Z^2) \tag{84}$$

where α_e and α_i are the electron and ion linear polarizabilities, and

$$\epsilon(l) = 1 + \alpha_e(l) + \alpha_i(l)$$

Then Eqs. (78), (83), and (84) can be contracted into the relation for the ternary correlation function:

$$n^2h(\mathbf{k}', \mathbf{k}'') = -\frac{2\epsilon_0 kk'k''}{ne^3\beta^2}\,\eta''(\mathbf{k}', \mathbf{k}'')$$
$$- \mathrm{Re} \sum_{l=k,k',k''} \frac{\epsilon_0 l^2}{\beta ne^2} \left\{\frac{\alpha_e(l) - Z\alpha_i(l)}{\epsilon(l)}\right\} + 2(1 - Z^2) \tag{85}$$

Equation (85) is the main statement of the present section. Its importance lies in that the high-order ternary correlation function can be inferred from the easily accessible quadratic polarization, whose evaluation can be restricted to the consideration of processes of much lower order than the others contributing to the ternary correlation.

The order-raising property of the static FDT can be most readily visualized by considering η in the lowest-order (Vlasov) approximation (for details, see Appendix B):

$$-\frac{2\epsilon_0 kk'k''}{ne^3\beta^2}\,\eta''(\mathbf{k}', \mathbf{k}'') = \frac{1 - Z^2}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')}$$

$$\alpha_e(k) = \beta ne^2/k^2, \qquad \alpha_i(k) = Z\beta ne^2/k^2, \qquad \epsilon(k) = 1 + (1 + Z)\,\beta ne^2/k^2$$

so that (85) becomes

$$n^2 h(\mathbf{k}', \mathbf{k}'') = (1 - Z^2) \left\{ \frac{1}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')} \sum_{l=k,k',k''} \frac{1}{\epsilon(l)} + 2 \right\}$$

$$= (1 - Z^2) \frac{1}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')} \{2\alpha(k)\,\alpha(k')\,\alpha(k'') + \alpha(k)\,\alpha(k') + \alpha(k')\,\alpha(k'') + \alpha(k'')\,\alpha(k)\} \quad (86)$$

Since, to this order, $\alpha = O(e^2 n)$, it then follows that $h = O(e^4)$, which is the expected result. If now α and η are calculated beyond the Vlasov approximation to order $O\{\sum_p (e^2 n)^p e^{2q}\}$, then h will evidently be determined to order $O\{\sum_p (e^2 n)^p e^{2(2+q)}\}$.

For the case of a pure electron plasma where the N/Z ions are assumed to be nailed down, Eq. (85) is altered insofar as one sets

$$Z = 0,\ \alpha_i(k) = 0 = \eta_i(k', k''),\ g(k) = g_{ee}(k), \text{ and } h(k', k'') = h_{eee}(k', k'').$$

The case of the electron–proton plasma is rather special: for $Z = 1$, Eq. (86) reveals that $h(k', k'') = 0$ and there are no triplet correlations [in the sense defined by (83a)] to $O(e^4)$.

3.3. Ternary Correlation Function: Conclusions and Comparisons

Equations (85) and (86) can be regarded in two different ways: First, if the response functions are given, then these equations are used for the calculation of the ternary correlation function. The second interpretation follows by eliminating ϵ with the aid of the linear FDT in favor of g and then constructing a *cluster expansion* for h in terms of the pair correlation function g.

Consider first Eq. (86) for an electron plasma. Invoking the relation

$$1/\epsilon(k) = 1 + ng(k) \quad (87)$$

valid in the Vlasov approximation, one immediately verifies that

$$h(k', k'') = g(k')\,g(k'') + g(k'')\,g(k) + g(k)\,g(k') + ng(k')\,g(k'')\,g(k) \quad (88)$$

This is the Fourier transform of the lowest-order Mayer cluster expansion for the ternary correlation function derived by Salpeter[8] by equilibrium statistical mechanics and by O'Neil and Rostoker[9], and by Lie and Ichikawa[10] solving the BBGKY hierarchy equations.

With ion dynamics included, the situation is somewhat more complicated. The four correlation functions appearing in (84) can be expressed as

$$g_{ee} = g_0 , \qquad g_{ei} = g_{ie} = -Zg_0 , \qquad g_{ii} = Z^2 g_0 , \tag{89}$$

and

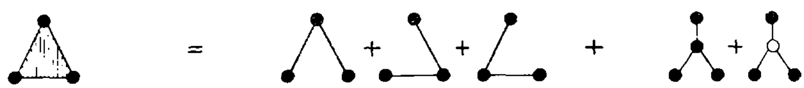
$$1/\epsilon(k) = 1 + (1 - Z)\, ng_0(k) \tag{90}$$

Then the relation equivalent to (88) becomes

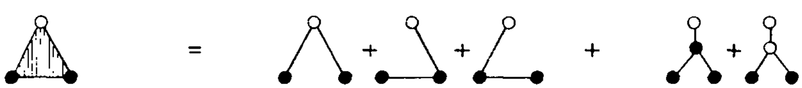
$$\begin{aligned} h(\mathbf{k}', \mathbf{k}'') = (1 - Z^2)(1 + Z)^2 \{ & g_0(k')\, g_0(k'') + g_0(k'')\, g_0(k) + g_0(k)\, g_0(k') \\ & + (1 + Z)\, ng_0(k')\, g_0(k'')\, g_0(k)\} \end{aligned} \tag{91}$$

However, $h(\mathbf{k}', \mathbf{k}'')$ does not have an immediate physical meaning: it is related to the physical correlations by (83a). On the other hand, the triplet correlations between specific groups of particles like h_{eee} and h_{eei} are expandable in Mayer-type clusters, similar to (88). These expansions, illustrated by their corresponding cluster diagrams,[10] are obvious generalizations of Salpeter's[(8)] formulation and are listed below:

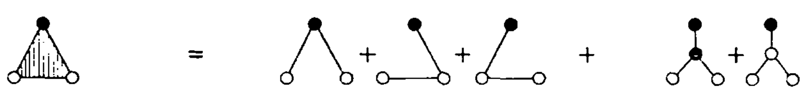
$$h_{eee}(\mathbf{k}', \mathbf{k}'') = \sum_{l,l' = k,k',k''} g(l)\, g(l') + n(1 - Z^2)\, g(k')\, g(k'')\, g(k)$$



$$h_{eei}(\mathbf{k}', \mathbf{k}'') + h_{eie}(\mathbf{k}', \mathbf{k}'') + h_{iee}(\mathbf{k}', \mathbf{k}'')$$
$$= (Z^2 - 2Z) \sum_{l,l' = k,k',k''} g(l)\, g(l') + 3(Z^3 - Z)\, ng(k')\, g(k'')\, g(k)$$

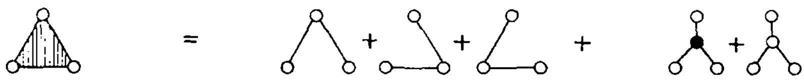


$$h_{iie}(\mathbf{k}', \mathbf{k}'') + h_{iei}(\mathbf{k}', \mathbf{k}'') + h_{eii}(\mathbf{k}', \mathbf{k}'')$$
$$= (Z^2 - 2Z^3) \sum_{l,l'} g(l)\, g(l') + 3(Z - Z^4)\, ng(k')\, g(k'')\, g(k)$$



[10] The diagrams are conventional Mayer cluster diagrams in configuration space. Filled vertices represent electrons and hollow vertices represent ions. Bonds between vertices 1 and 2 are associated with $g(12)$. Triple products of Fourier transforms appear as integrals over a dummy particle carrying a density factor n (if it is an electron) or N/Z (if it is an ion).

$$h_{iii}(\mathbf{k}', \mathbf{k}'') = Z^4 \sum_{l,l'} g(l)\, g(l') + (Z^5 - Z^2)\, ng(k')\, g(k'')\, g(k)$$



When these terms are combined according to the rules of (83a), the result is identical to (91).

In the rest of this section, we confine our attention to an electron plasma. If one goes beyond the lowest order approximation, higher-order contributions to h emerge both from higher-order corrections to g and η. The form assumed by η in the Vlasov approximation suggests the introduction of the collisional correction $\delta(\mathbf{k}', \mathbf{k}'')$ as follows:

$$\eta''(\mathbf{k}', \mathbf{k}'') = -\frac{\beta^2 ne^3}{2\epsilon_0 kk'k''} \frac{1}{\epsilon(k)\, \epsilon(k')\, \epsilon(k'')} [1 + \delta(\mathbf{k}', \mathbf{k}'')]$$

We also set

$$g(k) = g_0(k) + \bar{g}(k), \qquad \Gamma(k) = \bar{g}(k)/[1 + ng_0(k)]$$

where g_0 is the Debye pair correlation function and $\bar{g}$ (or Γ) is a collisional correction. Then, with the aid of (87), one can rewrite (85) (with $Z = 0$) as

$$\begin{aligned} h(\mathbf{k}', \mathbf{k}'') = {} & \sum_{l,l'} g(l)\, g(l') + ng(k)\, g(k')\, g(k'') + \sum_{l,l',l''} ng(l)\, g(l')\, \Gamma(l'') \\ & + \sum_{l,l'} g(l)\, \Gamma(l') - \sum_{l,l',l''} ng(l)\, \Gamma(l')\, \Gamma(l'') + \sum_{l} (1/n)\, \Gamma(l) \\ & - \sum_{l,l'} \Gamma(l)\, \Gamma(l') - n\Gamma(k)\, \Gamma(k')\, \Gamma(k'') \\ & + \delta(\mathbf{k}', \mathbf{k}'')\{1 + ng(k') - n\Gamma(k')\}\{1 + ng(k'') - n\Gamma(k'')\} \\ & \cdot \{1 + ng(k) - n\Gamma(k)\} \end{aligned}$$

displaying modifications both in the cluster structure and due to the new units building up individual clusters.

4. CONCLUSIONS AND REMARKS

In this paper, we have established what relation prevails between the quadratic plasma conductivities, dielectric functions, and the response functions on the one hand, and equilibrium three-point density and current correlations on the other. In addition to the inference drawn concerning the triplet correlation function, discussed in the previous section, there are further implications of the theorem. Whenever the calculation of scattering cross sections, Fokker-Planck coefficients, and like quantities describing the interaction of incident electromagnetic waves or particle beams with an equilibrium plasma is contemplated beyond the lowest-order Born-type approximation,[11,13] an expansion in the perturbing electric field (due to the wave or to the particles) ensues with two-, three-, n-point equilibrium correlation functions appearing as expansion coefficients. That the lowest-order coefficients can be explained entirely in terms of the dielectric function is by now well-established; the present theorem tells us how a similar procedure can be performed for the next term in the expansion (for the details of the test-particle problem, see Ref. 12).

The above observation indicates in which direction one could look for direct experimental verification of the results derived in this paper. Evidently, independent measurements of the two physical quantities linked by the theorem–the quadratic conductivity and the three-point correlation function–would be required. The first one has been a long-standing experimental program for semiconductor plasmas.[3] In the case of gaseous plasmas, the difficulty of generating genuine equilibrium plasmas, especially under the influence of strong perturbations, is, at the present time, a major experimental obstacle. As far as the second objective is concerned, the most promising method to gain information about the triplet correlation function, seems to be—in view of the discussion presented in the above paragraph—the measurement of incoherent scattering crosssections for high-intensity electromagnetic waves. No detailed theoretical understanding or experimental observation of this process seems to exist, however, at the present time.

APPENDIX A. DERIVATION OF THE POISSON BRACKET IDENTITY

First, we observe that

$$\langle[\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau'-\tau'')\, j_{\mathbf{k}\gamma}(\tau')]\rangle^{(0)}$$

$$= -\sum_{i=1}^{N}\left\langle\frac{\partial\rho_{-\mathbf{k}'}(0)}{\partial x_{i\alpha}(0)}\frac{\partial}{\partial p_{i\alpha}(0)}\{j_{-\mathbf{k}''\beta}(\tau'-\tau'')\, j_{\mathbf{k}\gamma}(\tau')\}\right\rangle^{(0)}$$

$$= -\sum_{i=1}^{N}\left\langle\frac{\partial}{\partial p_{i\alpha}(0)}\left\{\frac{\partial\rho_{-\mathbf{k}'}(0)}{\partial x_{i\alpha}(0)}\, j_{-\mathbf{k}''\beta}(\tau'-\tau'')\, j_{\mathbf{k}\gamma}(\tau')\right\}\right\rangle^{(0)}$$

$$= -iek_\alpha'\sum_{i}\int dx\, dp\, \Omega^{(0)}\frac{\partial}{\partial p_{i\alpha}(0)}(\{\exp[i\mathbf{k}'\cdot\mathbf{x}_i(0)]\}\, j_{-\mathbf{k}''\beta}(\tau'-\tau'')\, j_{\mathbf{k}\gamma}(\tau'))$$

$$= iek_\alpha' \sum_i \int dx\, dp\, \{\exp[i\mathbf{k}' \cdot \mathbf{x}_i(0)]\}\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\, \frac{\partial \Omega^{(0)}}{\partial p_{i\alpha}(0)}$$

[see Eq. (5)]

$$= -ie\beta k_\alpha' \sum_i \int dx\, dp\, \Omega^{(0)} v_{i\alpha}(0)\, \{\exp[i\mathbf{k}' \cdot \mathbf{x}_i(0)]\}\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')$$

$$= -i\beta k_\alpha' \langle j_{-\mathbf{k}'\alpha}(0)\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)}$$

Hence expansion of the Poisson bracket yields

$$\langle [\rho_{-\mathbf{k}'}(0), j_{-\mathbf{k}''\beta}(\tau' - \tau'')]\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)} = -i\beta k_\alpha' \langle j_{-\mathbf{k}'\alpha}(0)\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\, j_{\mathbf{k}\gamma}(\tau')\rangle^{(0)} - \langle [\rho_{-\mathbf{k}'}(0), j_{\mathbf{k}\gamma}(\tau')]\, j_{-\mathbf{k}''\beta}(\tau' - \tau'')\rangle^{(0)}$$

APPENDIX B. STATIC NONLINEAR DIELECTRIC FUNCTION FOR A VLASOV PLASMA

We consider here an ion–electron Vlasov plasma consisting of N electrons (each of mass m_e and charge $-|e|$) and N/Z ions (each of mass m_i and charge $Z|e|$). The Vlasov equations for the one-particle ion and electron distribution functions are ($\alpha = e, i$)

$$\frac{\partial F_\alpha(\mathbf{r}, t; \mathbf{v})}{\partial t} + \mathbf{v} \cdot \nabla F_\alpha(\mathbf{r}, t; \mathbf{v}) + \frac{Z_\alpha |e|}{m_\alpha} \langle \mathbf{E}(\mathbf{r}, t)\rangle \cdot \frac{\partial F_\alpha(\mathbf{r}, t; \mathbf{v})}{\partial \mathbf{v}} = 0 \qquad \text{(B.1)}$$

where $\langle \mathbf{E}\rangle$ is the average macroscopic field, $Z_e = -1$, and $Z_i = Z$, with the perturbation expansion

$$F_\alpha(\mathbf{r}, t; \mathbf{v}) = F_\alpha^{(0)}(\mathbf{v}) + F_\alpha^{(1)}(\mathbf{r}, t; \mathbf{v}) + F_\alpha^{(2)}(\mathbf{r}, t; \mathbf{v}) + \cdots \qquad \text{(B.2)}$$

$$\langle \mathbf{E}(\mathbf{r}, t)\rangle = \langle \mathbf{E}(\mathbf{r}, t)\rangle^{(1)} + \langle \mathbf{E}(\mathbf{r}, t)\rangle^{(2)} + \cdots \qquad \text{(B.3)}$$

about the Maxwellian state characterized by

$$F_\alpha^{(0)}(\mathbf{v}) = \frac{N/|Z_\alpha|}{V} \left(\frac{\beta}{2\pi m_\alpha}\right)^{3/2} \exp\left(\frac{-\beta m_\alpha v^2}{2}\right) \qquad \text{(B.4)}$$

substituted into (B.1) and one sees that the result can be split into two equations for the first- and second-order distribution functions $F_\alpha^{(1)}$ and $F_\alpha^{(2)}$. Upon taking Fourier transforms of these last two equations, one obtains

$$F_\alpha^{(1)}(\mathbf{k}, \omega; \mathbf{v}) = -\frac{iZ_\alpha |e|}{m_\alpha} \frac{\langle \mathbf{E}(\mathbf{k}\omega)\rangle^{(1)} \cdot \partial F_\alpha^{(0)}(\mathbf{v})/\partial \mathbf{v}}{\omega - \mathbf{k} \cdot \mathbf{v}} \qquad \text{(B.5)}$$

$$F_\alpha^{(2)}(\mathbf{k}, \omega; \mathbf{v}) = -\frac{iZ_\alpha |e|}{m_\alpha} \frac{\langle \mathbf{E}(\mathbf{k}\omega)\rangle^{(2)} \cdot \partial F_\alpha^{(0)}(\mathbf{v})/\partial \mathbf{v}}{\omega - \mathbf{k} \cdot \mathbf{v}} - \frac{iZ_\alpha |e|}{m_\alpha} \frac{1}{2\pi V} \sum_{\mathbf{k}'} \int d\omega' \frac{\langle \mathbf{E}(\mathbf{k}'', \omega'')\rangle^{(1)} \cdot \partial F_\alpha^{(1)}(\mathbf{k}', \omega'; \mathbf{v})/\partial \mathbf{v}}{\omega - \mathbf{k} \cdot \mathbf{v}} \qquad \text{(B.6)}$$

where $\mathbf{k}'' = \mathbf{k} - \mathbf{k}'$.

Let us consider the electron gas. The second-order, average current density, which is of interest here, is readily formed from (B.5) and (B.6) to be:

$$\begin{aligned}\langle j_\alpha^e(\mathbf{k}\omega)\rangle^{(2)} &= -|e| \int d^3\mathbf{v}\, v_\alpha F_e^{(2)}(\mathbf{k}, \omega; \mathbf{v}) \\ &= -\frac{i|e|^2}{m_e^2} \langle E_\beta(\mathbf{k}\omega)\rangle^{(2)} \int d^3\mathbf{v}\, v_\alpha \frac{\partial F_e^{(0)}(\mathbf{v})/\partial v_\beta}{\omega - \mathbf{k}\cdot\mathbf{v}} \\ &+ \frac{|e|^3}{4\pi V m_e^2} \sum_{\mathbf{k}'} \int d\omega' \langle E_\mu(\mathbf{k}', \omega')\rangle^{(1)} \langle E_\nu(\mathbf{k}'', \omega'')\rangle^{(1)} \\ &\times \int d^3\mathbf{v} \frac{v_\alpha}{\omega - \mathbf{k}\cdot\mathbf{v}} \left\{ \frac{\partial}{\partial v_\mu} \frac{\partial F_e^{(0)}(\mathbf{v})/\partial v_\nu}{\omega'' - \mathbf{k}''\cdot\mathbf{v}} + \frac{\partial}{\partial v_\nu} \frac{\partial F_e^{(0)}(\mathbf{v})/\partial \mathbf{v}}{\omega' - \mathbf{k}'\cdot\mathbf{v}} \right\}\end{aligned} \tag{B.7}$$

Upon comparing (B.7) with Ohm's law

$$\begin{aligned}\langle j_\alpha^e(\mathbf{k}\omega)\rangle^{(2)} &= \sigma_{\alpha\beta}^{(e)}(\mathbf{k}, \omega)\langle E_\beta(\mathbf{k}\omega)\rangle^{(2)} + (1/2\pi V) \sum_{\mathbf{k}'} \int d\omega'\, \sigma_{\alpha\mu\nu}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ &\times \langle E_\mu(\mathbf{k}', \omega')\rangle^{(1)} \langle E_\nu(\mathbf{k}'', \omega'')\rangle^{(1)}\end{aligned} \tag{B.8}$$

one finds that

$$\sigma_{\alpha\beta}^{(e)}(\mathbf{k}\omega) = -\frac{i|e|^2}{m} \int d^3\mathbf{v}\, v_\alpha \frac{\partial F_e^{(0)}(\mathbf{v})/\partial v_\beta}{\omega - \mathbf{k}\cdot\mathbf{v}} \tag{B9}$$

and

$$\begin{aligned}&\sigma_{\alpha\mu\nu}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \\ &= \frac{|e|^3}{2m_e^2} \int d^3\mathbf{v} \frac{v_\alpha}{\omega - \mathbf{k}\cdot\mathbf{v}} \left\{ \frac{\partial}{\partial v_\mu} \frac{\partial F_e^{(0)}(\mathbf{v})/\partial v_\nu}{\omega'' - \mathbf{k}''\cdot\mathbf{v}} + \frac{\partial}{\partial v_\nu} \frac{\partial F_e^{(0)}(\mathbf{v})/\partial v_\mu}{\omega' - \mathbf{k}'\cdot\mathbf{v}} \right\}\end{aligned} \tag{B.10}$$

From our earlier electrodynamic study,[5] we found that

$$\sigma_{\alpha\mu\nu}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') = i\epsilon_0(\omega' + \omega'')\, \epsilon_{\alpha\mu\nu}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') \tag{B.11}$$

Then upon eliminating $\sigma_{\alpha\mu\nu}^{(e)}$ between (B.10) and (B.11) in favor of $\epsilon_{\alpha\mu\nu}^{(e)}$, and taking the longitudinal projection of the result, one ultimately obtains the symmetrized (with respect to prime–double-prime interchange) relation

$$\begin{aligned}\epsilon_{NL}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega', \omega'') &= \frac{i|e|^3}{2\epsilon_0 m_e^2 k} \int d^3\mathbf{v} \frac{1}{\omega - \mathbf{k}\cdot\mathbf{v}} \left\{ \mathbf{k}' \cdot \frac{\partial}{\partial \mathbf{v}} \frac{\mathbf{k}''\cdot \partial F_e^{(0)}/\partial \mathbf{v}}{\omega'' - \mathbf{k}''\cdot\mathbf{v}} \right. \\ &\left. + \mathbf{k}'' \cdot \frac{\partial}{\partial \mathbf{v}} \frac{\mathbf{k}'\cdot \partial F_e^{(0)}/\partial \mathbf{v}}{\omega' - \mathbf{k}'\cdot\mathbf{v}} \right\} \\ &= -\frac{i\beta|e|^3}{2\epsilon_0 m_e k k' k''} \int d^3\mathbf{v} \frac{1}{\omega - \mathbf{k}\cdot\mathbf{v}} \left\{ \mathbf{k}' \cdot \frac{\partial}{\partial \mathbf{v}} \frac{F_e^{(0)}(\mathbf{k}''\cdot\mathbf{v})}{\omega'' - \mathbf{k}''\cdot\mathbf{v}} \right. \\ &\left. + \mathbf{k}'' \cdot \frac{\partial}{\partial \mathbf{v}} \frac{F_e^{(0)}(\mathbf{k}'\cdot\mathbf{v})}{\omega' - \mathbf{k}'\cdot\mathbf{v}} \right\}\end{aligned} \tag{B.12}$$

Hence in the static ($\omega' \to 0$, $\omega'' \to 0$) limit,

$$\epsilon_{NL}^{(e)}(\mathbf{k}', \mathbf{k}''; \omega' = 0, \omega'' = 0) = \frac{in\beta^2 \mid e \mid^3}{2\epsilon_0 kk'k''}$$

$$\eta_e''(\mathbf{k}', \mathbf{k}'') = \frac{\beta^2 \mid e \mid^3}{2\epsilon_0 kk'k''} \frac{1}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')} \tag{B.13}$$

For the ion gas, one can similarly show that

$$\eta_i''(\mathbf{k}', \mathbf{k}'') = -\frac{\beta^2 Z^2 \mid e \mid^3 n}{2\epsilon_0 kk'k''} \frac{1}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')} \tag{B.14}$$

Hence the complete expression is

$$\eta''(\mathbf{k}', \mathbf{k}'') = \frac{(1 - Z^2)\,\beta^2 \mid e \mid^3}{2\epsilon_0 kk'k''} \frac{1}{\epsilon(k)\,\epsilon(k')\,\epsilon(k'')} \tag{B.15}$$

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