Quadratic fluctuation-dissipation theorem for multilayer plasmas

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The author establishes the dynamical and static quadratic fluctuation-dissipation theorems (QFDTs) for multilayer classical one-component plasmas in the absence of external magnetic fields. Areal densities and spacings between layers need not be equal. The static QFDT is used to derive the lowest-order (in coupling parameter) Mayer cluster expansion for the layer-space matrix elements of the equilibrium three-point correlation function. [S1063-651X(99)08301-4]

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I. INTRODUCTION

Interest in correlated multilayer plasmas over the past decade has been stimulated by activities in two different areas. In the area of strongly coupled plasma physics, there are the compelling experiments of Bollinger and co-workers [1] where laser-cooled ions in a trap form a realistic model of a layered classical one-component plasma (OCP) in its strongly coupled liquid and crystalline phases (see Ref. [2] for a summary and theoretical description). In the area of condensed matter plasmas, advances in modern semiconductor nanotechnology have made it possible to routinely fabricate multiple-quantum-well structures of parallel electron layers in a strongly correlated liquid phase [3].

Theoretical efforts pertaining to static properties have been primarily directed at questions of liquid-solid phase boundaries in multiple-quantum-well structures [4], at identification of structural phases in the classical bilayer liquid [5] and Wigner crystal phases [6], and at adapting the classical hypernetted-chain (HNC) approximation to the computation of pair correlation and static structure functions for the bilayer electron liquid [5]. Efforts pertaining to dynamic properties have concentrated on frequency-moment-sum rules [7] and on the dielectric response tensor and collective mode behavior in strongly correlated bilayer and superlattice plasmas [8–13].

Central to the description of the static and dynamic properties of multilayer plasmas are the density-response and structure function layer-space matrices and the hierarchy of fluctuation-dissipation theorems (FDTs) which link these quantities. The linear conventional FDT has already been established [7,13] for the infinite, type-I superlattice model consisting of a large stack of N_L equally spaced two-dimensional (2D) electron plasma monolayers of equal areal density $n_e = N_e/A$. It is a relatively easy task to recast the linear FDT in a compact matrix form that applies to more general classical OCP multilayers where N_L is arbitrary and where the areal densities and spacings between adjacent layers need not be the same. This is done in the present paper en route to pursuing the main goal described below.

Next in the FDT hierarchy is the quadratic fluctuation-dissipation theorem (QFDT) which, in its most useful frequency-domain form, connects a single three-point dynamical structure function to a triangle-symmetric combination of three quadratic-density-response functions [see Eq. (29) below]. Such QFDT relations have been formulated [14–18] and implemented in novel kinetic theory approaches [19–21] to the calculation of the dielectric response function and plasmon structure in three-dimensional OCP [19] and binary-ionic-mixture (BIM) plasmas [20,21].

The main goal of the present paper is to establish the dynamical and static QFDT relations for the unmagnetized multilayer OCP; areal densities and spacings between adjacent layers need not be equal. It is also of interest to apply the static QFDT to the calculation of the lowest-order (in the coupling parameter) Mayer expansion linking the matrix elements of the k-space equilibrium ternary and pair correlation functions.

The plan of the paper is as follows. Matrix elements of two- and three-point current correlation (C) and structure (S) functions are defined in Sec. II. In Sec. III two kinds of linear and quadratic response functions are introduced: external response functions which portray the system response to external potential perturbations and total (screened) response functions which portray the response to total (polarization plus external) perturbing potentials. The dynamical QFDTs are established in Sec. IV, first in the time domain and then in the frequency domain with the results displayed as C-$\hat{\sigma}$ and S-$\hat{\chi}$ relations ($\hat{\sigma}$ and $\hat{\chi}$ are symbols for the external conductivity and density-response function matrices). In Sec. V the static form of the QFDT is established. This is followed by a derivation of the lowest-order Mayer cluster expansion for the matrix elements of the equilibrium three-point correlation function. Conclusions are drawn in Sec. VI.

II. CURRENT CORRELATION AND STRUCTURE FUNCTIONS

Consider a multilayer plasma model consisting of a stack of two or more electron-plasma monolayers, each of large but bounded area A and parallel to the xy plane. The two-dimensional OCP in a typical monolayer A ($A=1,2,3,...$) is comprised of N_A classical point electrons in a neutralizing uniform positive background. Fourier components of the microscopic charge and current densities are given by

$$\rho_{\mathbf{k}}^{A}(t) = -e n_{\mathbf{k}}^{A}(t) = -e \sum_{i=1}^{N_A} e^{-i\mathbf{k}\cdot\mathbf{x}_i^{A}(t)}, \tag{1a}$$

$$\mathbf{j}_{\mathbf{k}}^{A}(t)=-e\sum_{i=1}^{N_A}\mathbf{v}_i^A(t)e^{-i\mathbf{k}\cdot\mathbf{x}_i^A(t)}, \tag{1b}$$

where $\mathbf{x}_i^A$ and $\mathbf{v}_i^A$ are the position and velocity of the ith electron in layer A; $\mathbf{k}$ is a wave vector in the xy plane.

We first define longitudinal (with respect to in-plane wave vectors $\mathbf{k},\mathbf{k}',\mathbf{k}''$) two- and three-point current correlation functions C_{AB} and C_{ABC}:

$$C_{AB}(\mathbf{k},t)\delta_{\mathbf{k}-\mathbf{k}'}=\frac{1}{Akk'}\langle[\mathbf{k}\cdot\mathbf{j}_{\mathbf{k}}^A(0)][\mathbf{k}'\cdot\mathbf{j}_{-\mathbf{k}'}^B(-t)]\rangle^{(0)}, \tag{2}$$

$$C_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\delta_{\mathbf{k}-\mathbf{k}'-\mathbf{k}''}=\frac{1}{2Akk'k''}\times\langle[\mathbf{k}\cdot\mathbf{j}_k^A(0)][\mathbf{k}'\cdot\mathbf{j}_{-\mathbf{k}'}^B(-t')][\mathbf{k}''\cdot\mathbf{j}_{-\mathbf{k}''}^C(-t'')]\rangle^{(0)}. \tag{3}$$

The $\langle\ \rangle^{(0)}$ denote averaging over the equilibrium ensemble characterized by the macrocanonical distribution $\Omega^{(0)}\propto\exp(-\beta H^{(0)})$; β is the inverse temperature (in energy units) and $H^{(0)}$ is the Hamiltonian of the equilibrium system. Two- and three-point layer-space structure functions are next introduced in the customary way in terms of microscopic fluctuating densities $\delta n_{\mathbf{k}}^A=n_{\mathbf{k}}^A-N_A\delta_{\mathbf{k}}$, $\delta n_{\mathbf{k}}^B=n_{\mathbf{k}}^B-N_B\delta_{\mathbf{k}},\ldots$:

$$\begin{aligned}(N_AN_B)^{1/2}S_{AB}(\mathbf{k},t)\delta_{\mathbf{k}-\mathbf{k}'}&=\langle\delta n_{\mathbf{k}}^A(0)\delta n_{-\mathbf{k}'}^B(-t)\rangle^{(0)}\\&=\langle n_{\mathbf{k}}^A(0)n_{-\mathbf{k}'}^B(-t)\rangle^{(0)}\\&\quad-N_AN_B\delta_{\mathbf{k}}\delta_{\mathbf{k}'},\end{aligned} \tag{4}$$

$$\begin{aligned}&(N_AN_BN_C)^{1/3}S_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\delta_{\mathbf{k}-\mathbf{k}'-\mathbf{k}''}\\&=\langle\delta n_{\mathbf{k}}^A(0)\delta n_{-\mathbf{k}'}^B(-t')\delta n_{-\mathbf{k}''}^C(-t'')\rangle^{(0)}\\&=\langle n_{\mathbf{k}}^A(0)n_{-\mathbf{k}'}^B(-t')n_{-\mathbf{k}''}^C(-t'')\rangle^{(0)}\\&\quad-N_A(N_BN_C)^{1/2}\delta_{\mathbf{k}}\delta_{\mathbf{k}'+\mathbf{k}''}S_{BC}(\mathbf{k}',t''-t')\\&\quad-N_B(N_AN_C)^{1/2}\delta_{\mathbf{k}'}\delta_{\mathbf{k}-\mathbf{k}''}S_{AC}(\mathbf{k},t'')\\&\quad-N_C(N_AN_B)^{1/2}\delta_{\mathbf{k}''}\delta_{\mathbf{k}-\mathbf{k}'}S_{AB}(\mathbf{k},t')\\&\quad-N_AN_BN_C\delta_{\mathbf{k}}\delta_{\mathbf{k}'}\delta_{\mathbf{k}''}.\end{aligned} \tag{5}$$

Continuity of charge then requires that the temporal Fourier transforms of Eqs. (2)–(5) satisfy

$$k^2C_{AB}(\mathbf{k},\omega)=e^2\frac{(N_AN_B)^{1/2}}{A}\omega^2S_{AB}(\mathbf{k},\omega), \tag{6}$$

$$\begin{aligned}&k'k''|\mathbf{k}'+\mathbf{k}''|C_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&=-e^3\frac{(N_AN_BN_C)^{1/3}}{2A}\omega'\omega''(\omega'+\omega'')\\&\quad\times S_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'').\end{aligned} \tag{7}$$

III. RESPONSE FUNCTIONS

We begin with Ohm's law for the longitudinal external conductivity matrices linking the first- and second-order average current density responses in layer A to driving external scalar potentials acting at layers $B=1,2,\ldots$:

$$j_A^{(1)}(\mathbf{k},t)=-ik\sum_B\int_{-\infty}^{\infty}dt'\,\hat\sigma_{AB}(\mathbf{k},t')\hat\Phi_B(\mathbf{k},t-t'), \tag{8}$$

$$\begin{aligned}j_A^{(2)}(\mathbf{k},t)=&-\frac{1}{A}\sum_{\mathbf{k}'}k'k''\sum_{B,C}\int_{-\infty}^{\infty}dt'\int_{-\infty}^{\infty}dt''\\&\times\hat\sigma_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\hat\Phi_B(\mathbf{k}',t-t')\\&\times\hat\Phi_C(\mathbf{k}'',t-t'')\quad(\mathbf{k}''=\mathbf{k}-\mathbf{k}').\end{aligned} \tag{9}$$

We next introduce external response matrices linking the first- and second-order average particle density responses in layer A to the $\hat\Phi_B,\hat\Phi_C,\ldots$ driving external potentials:

$$n_A^{(1)}(\mathbf{k},t)=-e\sum_B\int_{-\infty}^{\infty}dt'\,\hat\chi_{AB}(\mathbf{k},t')\hat\Phi_B(\mathbf{k},t-t'), \tag{10}$$

$$\begin{aligned}n_A^{(2)}(\mathbf{k},t)=&\frac{e^2}{A}\sum_{\mathbf{k}'}\sum_{B,C}\int_{-\infty}^{\infty}dt'\int_{-\infty}^{\infty}dt''\,\hat\chi_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\\&\times\hat\Phi_B(\mathbf{k}',t-t')\hat\Phi_C(\mathbf{k}'',t-t'')\quad(\mathbf{k}''=\mathbf{k}-\mathbf{k}').\end{aligned} \tag{11}$$

Comparison of the temporal Fourier-transformed Eqs. (8) and (9) with their respective Fourier-transformed counterpart Eqs. (10) and (11) then yields the useful matrix relations

$$\omega e^2\hat\chi_{AB}(\mathbf{k},\omega)=-ik^2\hat\sigma_{AB}(\mathbf{k},\omega), \tag{12}$$

$$\begin{aligned}&(\omega'+\omega'')e^3\hat\chi_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&=k'k''|\mathbf{k}'+\mathbf{k}''|\hat\sigma_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'').\end{aligned} \tag{13}$$

The constitutive relations for the total particle-density-response functions are

$$n_A^{(1)}(\mathbf{k},t)=-e\sum_B\int_{-\infty}^{\infty}dt'\,\chi_{AB}(\mathbf{k},t')\Phi_B^{(1)}(\mathbf{k},t-t'), \tag{14}$$

$$\begin{aligned}n_A^{(2)}(\mathbf{k},t)=&-e\sum_B\int_{-\infty}^{\infty}dt'\,\chi_{AB}(\mathbf{k},t')\Phi_B^{(2)}(\mathbf{k},t-t')\\&+\frac{e^2}{A}\sum_{\mathbf{k}'}\sum_{B,C}\int_{-\infty}^{\infty}dt'\int_{-\infty}^{\infty}dt''\,\chi_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\\&\times\Phi_B^{(1)}(\mathbf{k}',t-t')\Phi_C^{(1)}(\mathbf{k}'',t-t'')\quad(\mathbf{k}''=\mathbf{k}-\mathbf{k}'),\end{aligned} \tag{15}$$

where Φ_B and Φ_C are total (screened) potentials. For the case of an equal-density ($n_e=N_1/A=N_2/A$) two-layer system, a physically transparent relationship (from the point of view of dynamical screening) between the external and total particle-density-response matrix elements can be readily established. Introducing the dielectric matrix

$$\epsilon_{AB}(\mathbf{k},\omega)=\delta_{AB}-\sum_{C}\chi_{AC}(\mathbf{k},\omega)\phi_{CB}(k), \tag{16}$$

where $\phi_{11}(k)=\phi_{22}(k)=\phi_{2D}(k)=2\pi e^2/k$ and $\phi_{12}(k)=\phi_{21}(k)=\phi_{2D}(k)e^{-kd}$ (d is the separation distance), and observing that

$$\Phi_A^{(1)}(\mathbf{k},\omega)=\sum_{B}[\boldsymbol{\epsilon}^{-1}(\mathbf{k},\omega)]_{AB}\hat{\Phi}_B(\mathbf{k},\omega), \tag{17}$$

comparison of the temporal Fourier-transformed Eqs. (10) and (11) with their respective counterpart Eqs. (14) and (15) yields

$$\hat{\chi}_{AB}(\mathbf{k},\omega)=\sum_{C}[\boldsymbol{\epsilon}^{-1}(\mathbf{k},\omega)]_{AC}\chi_{CB}(\mathbf{k},\omega), \tag{18}$$

$$\begin{aligned}\hat{\chi}_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')&=\sum_{A',B',C'}[\boldsymbol{\epsilon}^{-1}(\mathbf{k}'+\mathbf{k}'',\omega'+\omega'')]_{AA'}\\&\quad\times\chi_{A'B'C'}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&\quad\times[\boldsymbol{\epsilon}^{-1}(\mathbf{k}',\omega')]_{B'B}[\boldsymbol{\epsilon}^{-1}(\mathbf{k}'',\omega'')]_{C'C}.\end{aligned} \tag{19}$$

Equation (18), in fact, holds for multilayer OCPs, where areal densities and spacings between adjacent layers need not be equal. The matrix dielectric screening depicted by Eq. (19) for the equal-density bilayer plasma mirrors the scalar dielectric screening structure reported for the one-component plasma [14,19].

In establishing the relations between the external and total-density-response-matrix elements, the matrix formulation becomes unwieldy for more than two layers. However, for a large number N_L of equally spaced OCP layers of equal areal density, the periodic structure (for $N_L\to\infty$) of the configuration allows one to introduce a Fourier transformation along the superlattice axis, e.g.,

$$\epsilon(\mathbf{k},k_z,\omega)=\sum_{A}\epsilon_{AA'}(\mathbf{k},\omega)e^{-ik_zd(A-A')}, \tag{20}$$

where d is the spacing between layers. For the infinite superlattice, comparison is made between the temporal- and layer-space Fourier-transformed Eqs. (10), (11) and their respective Fourier transformed counterpart Eqs. (14), (15). With the stipulation that the external charge perturbation is confined to the lattice planes, the desired relations

$$\hat{\chi}(\mathbf{k},k_z,\omega)=\frac{\chi(\mathbf{k},k_z,\omega)}{\epsilon(\mathbf{k},k_z,\omega)}, \tag{21}$$

$$\hat{\chi}(\mathbf{k}',k_z',\omega';\mathbf{k}'',k_z'',\omega'')=\frac{\chi(\mathbf{k}',k_z',\omega';\mathbf{k}'',k_z'',\omega'')}{\epsilon(\mathbf{k}'+\mathbf{k}'',k_z'+k_z'',\omega'+\omega'')\epsilon(\mathbf{k}',k_z',\omega')\epsilon(\mathbf{k}'',k_z'',\omega'')} \tag{22}$$

then follow from Poisson's equation, the constitutive relation (40) of Ref. [22] linking external and induced charge densities, and the χ-ϵ relation (18) of Ref. [7].

IV. DYNAMICAL FLUCTUATION-DISSIPATION RELATIONS

We come now to the main point of the paper: the formulation of the dynamical QFDT for the unmagnetized multilayer OCP; areal densities and spacings between adjacent layers need not be equal.

The unperturbed state of the multilayer system is characterized by the macrocanonical distribution function $\Omega^{(0)}\propto\exp[-\beta H^{(0)}]$. Then following the well-known statistical-mechanical perturbation-theoretic method of Kubo [23], I calculate the first- and second-order current density response in layer A to the weak perturbing Hamiltonian

$$\hat{H}(t)=\frac{1}{A}\sum_{B}\sum_{\mathbf{k}}\hat{\Phi}_B(\mathbf{k},t)\rho_{-\mathbf{k}}^B \tag{23}$$

by ensemble averaging over the perturbed first- and second-order Liouville distribution functions. The lengthy procedure parallels the one carried out some time ago by Lu and myself [18] for binary ionic mixture plasmas and it suffices here to proceed directly to the principal results. Though the focus is primarily on the QFDT relations, their linear companion relations are also displayed to better elucidate the structure of the FDT hierarchy. The results are presented in the order in which they have been derived beginning with the external conductivity-current correlation relations: *time domain:*

$$\hat{\sigma}_{AB}(\mathbf{k},t)=\beta C_{AB}(\mathbf{k},t)\theta(t), \tag{24}$$

$$\begin{aligned}&\hat{\sigma}_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')-\hat{\sigma}_{CBA}(\mathbf{k}',t''-t';-\mathbf{k},t'')\\&\quad\times\theta(t')-\hat{\sigma}_{BAC}(-\mathbf{k},t';\mathbf{k}'',t-t'')\theta(t'')\\&\quad=\beta^2C_{ABC}(\mathbf{k}',t';\mathbf{k}'',t'')\theta(t')\theta(t'')\quad(\mathbf{k}''=\mathbf{k}-\mathbf{k}'),\end{aligned} \tag{25}$$

frequency domain:

$$\mathrm{Re}\,\hat{\sigma}_{AB}(\mathbf{k},\omega)=\tfrac{1}{2}\beta C_{AB}(\mathbf{k},\omega), \tag{26}$$

$$\begin{aligned}&\mathrm{Re}\{\hat{\sigma}_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')+\hat{\sigma}_{CAB}(-\mathbf{k},-\omega;\mathbf{k}',\omega')\\&\quad+\hat{\sigma}_{BCA}(\mathbf{k}'',\omega'';-\mathbf{k},-\omega)\}\\&\quad=\tfrac{1}{2}\beta^2C_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&\qquad(\mathbf{k}''=\mathbf{k}-\mathbf{k}',\ \omega''=\omega-\omega').\end{aligned} \tag{27}$$

$\theta(t),\theta(t'),\theta(t'')$ are unit step functions. In Eqs. (26) and (27) only the (physically meaningful) dissipative parts of the conductivity matrix elements are displayed. We observe that the current correlation matrix element $C_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')$ undergoes no net change in sign under simultaneous microscopic time reversal and 2D space inversion so that $C_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega''{}'')$ must be real.

The corresponding dynamical FDTs relating the external density response and structure function matrix elements readily follow from Eqs. (6), (7), (12), (13), (26), and (27):

$$\mathrm{Im}\ \hat{\chi}_{AB}(\mathbf{k},\omega)=-\beta\,\frac{(N_AN_B)^{1/2}}{2A}\,\omega S_{AB}(\mathbf{k},\omega), \tag{28}$$

$$\begin{aligned}\mathrm{Re}\Big\{&\frac{1}{\omega'\omega''}\,\hat{\chi}_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&-\frac{1}{\omega\omega'}\,\hat{\chi}_{CAB}(-\mathbf{k},-\omega;k',\omega')\\&-\frac{1}{\omega\omega''}\,\hat{\chi}_{BCA}(\mathbf{k}'',\omega'';-\mathbf{k},-\omega)\Big\}\\&=-\beta^2\,\frac{(N_AN_BN_C)^{1/3}}{4A}\,S_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')\\&\qquad(\mathbf{k}''=\mathbf{k}-\mathbf{k}',\quad \omega''=\omega-\omega').\end{aligned} \tag{29}$$

We observe that dynamical QFDTs (27) and (29) respect the invariance of C_{ABC} and S_{ABC} with respect to rotation on the triangle formed by the ''four'' vectors $(\mathbf{k}',z_B,\omega')$, $(\mathbf{k}'',z_C,\omega'')$, $(\mathbf{k},z_A,\omega)$, i.e.,

$$\begin{aligned}C_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')&=C_{CAB}(-\mathbf{k},-\omega;\mathbf{k}',\omega')\\&=C_{BCA}(\mathbf{k}'',\omega'';-\mathbf{k},-\omega),\end{aligned} \tag{30a}$$

$$\begin{aligned}S_{ABC}(\mathbf{k}',\omega';\mathbf{k}'',\omega'')&=S_{CAB}(-\mathbf{k},-\omega;\mathbf{k}',\omega')\\&=S_{BCA}(\mathbf{k}'',\omega'';-\mathbf{k},-\omega)\end{aligned} \tag{30b}$$

(z_A,z_B,z_C locate the layers along the z axis). Note the structural likeness between Eqs. (28) and (29) and their Ref. [18] counterpart Eqs. (58) and (A7) for the 3D binary-ionic-mixture plasmas; this is clearly a consequence of the one-to-one correspondence between the layer-space and species-space density-response-matrix formalisms for multilayer and multispecies plasmas.

For the infinite superlattice comprised of equally spaced 2D layers of equal areal density n_e, a Fourier transformation of Eq. (29) along the superlattice axis [per Eq. (20)] yields the QFDT

$$\begin{aligned}\mathrm{Re}\Big\{&\frac{1}{\omega'\omega''}\,\hat{\chi}(\mathbf{k}',k_z',\omega';\mathbf{k}'',k_z'',\omega'')\\&-\frac{1}{\omega\omega'}\,\hat{\chi}(-\mathbf{k},-k_z,-\omega;\mathbf{k}',k_z',\omega')\\&-\frac{1}{\omega\omega''}\,\hat{\chi}(\mathbf{k}'',k_z'',\omega'';-\mathbf{k},-k_z,-\omega)\Big\}\\&=-\tfrac{1}{4}\beta^2 n_e S(\mathbf{k}',k_z',\omega';\mathbf{k}'',k_z'',\omega'')\\&\qquad\Big(\mathbf{k}''=\mathbf{k}-\mathbf{k}',\quad k_z''=k_z-k_z'+\frac{2\pi}{d}\,s,\\&\qquad\omega''=\omega-\omega';\quad s=0,\pm1,\pm2,\ldots\Big).\end{aligned} \tag{31}$$

Its companion linear FDT (in the quantum domain) is reported in Refs. [7,12(c),13].

V. STATIC FLUCTUATION-DISSIPATION RELATIONS

An important ramification of linear FDT Eq. (28) is its static form

$$\mathrm{Re}\ \hat{\chi}_{AB}(\mathbf{k},\omega=0)=-\beta\,\frac{(N_AN_B)^{1/2}}{A}\,S_{AB}(\mathbf{k},t=0), \tag{32a}$$

or equivalently,

$$\begin{aligned}\delta_{AB}-\mathrm{Re}[\boldsymbol{\epsilon}^{-1}(\mathbf{k},\omega=0)]_{AB}\\=\frac{\beta}{A}\sum_C (N_AN_C)^{1/2}S_{AC}(\mathbf{k},t=0)\,\phi_{CB}(k),\end{aligned} \tag{32b}$$

which links the Fourier-transformed equilibrium pair correlation matrix element $g_{AB}(\mathbf{k})$ to the inverse static dielectric matrix via

$$S_{AB}(k)=\delta_{AB}+\frac{(N_AN_B)^{1/2}}{A}\,g_{AB}(\mathbf{k}). \tag{33}$$

The matrix element $\phi_{AB}(k)=(2\pi e^2/k)\exp[-kd(A-B)]$ is the Fourier transform of the layer-A–layer-B Coulomb potential. Equation (32b), which holds for multilayer plasmas having unequal layer populations and unequal spacings between adjacent layers, is reported in Ref. [12(b)] for the equal-density bilayer. Its superlattice linear FDT counterpart is reported in Refs. [12(b),13].

The more involved derivation of the static form of QFDT (29) calls for repeated application of the Kramers-Kronig relations and use of the Poincaré-Bertrand theorem [14,17,18,20,24]. An essential element in the derivation is the triangle symmetry requirement

$$\begin{aligned}\mathrm{Re}\ \hat{\chi}_{ABC}(\mathbf{k}',0;\mathbf{k}'',0)&=\mathrm{Re}\ \hat{\chi}_{CAB}(-\mathbf{k},0;\mathbf{k}',0)\\&=\mathrm{Re}\ \hat{\chi}_{BCA}(\mathbf{k}'',0;-\mathbf{k},0),\end{aligned} \tag{34}$$

paralleling Eq. (30b). Applying the analysis of Ref. [14] to QFDT Eq. (31), I obtain

$$\begin{aligned}\mathrm{Re}\ \hat{\chi}_{ABC}(\mathbf{k}',\omega'=0;\mathbf{k}'',\omega''=0)\\=\beta^2\,\frac{(N_AN_BN_C)^{1/3}}{2A}\\\times S_{ABC}(\mathbf{k}',t'=0;\mathbf{k}'',t''=0).\end{aligned} \tag{35}$$

Again, note the structural likeness between QFDT Eq. (35) and its Ref. [18] binary-ionic-mixture counterpart, Eq. (74).

We turn now to the final task of this paper: the application of Eq. (35) to the formulation of the lowest-order Mayer cluster expansion linking the matrix elements of the equilibrium three-particle (ternary) and pair correlation functions for the equal-density multilayer OCP (the spacings between adjacent monolayers need not be equal, however). The calculation of the total-density-response matrices, which is carried out in the random-phase approximation (RPA), results in the diagonal matrix elements

$$\chi_{AB}(\mathbf{k},0)|_{\mathrm{RPA}}=-\beta n_e\delta_{AB}, \tag{36}$$

$$\chi_{ABC}(\mathbf{k}',0;\mathbf{k}'',0)|_{\mathrm{RPA}}=\tfrac{1}{2}\beta^2 n_e \delta_{AB}\delta_{AC}. \quad (37)$$

Now, the external-total-density-response function relation (19), which is valid for the equal-density bilayer OCP at arbitrary coupling, holds as well for the equal-density multilayer OCP in the RPA. From Eqs. (18), (19), (32a), (36), and (37), one therefore obtains

$$S_{AB}(\mathbf{k},t=0)|_{\mathrm{RPA}}=[\boldsymbol{\epsilon}^{-1}(\mathbf{k},0)]_{AB}, \quad (38)$$

$$\hat{\chi}_{ABC}(\mathbf{k}',0;\mathbf{k}'',0)|_{\mathrm{RPA}}=\tfrac{1}{2}\beta^2 n_e \sum_D [\boldsymbol{\epsilon}^{-1}(\mathbf{k},0)]_{AD} \times[\boldsymbol{\epsilon}^{-1}(\mathbf{k}',0)]_{DB}[\boldsymbol{\epsilon}^{-1}(\mathbf{k}'',0)]_{DC} \quad (\mathbf{k}=\mathbf{k}'+\mathbf{k}''), \quad (39)$$

where

$$\epsilon_{AB}(\mathbf{k},0)|_{\mathrm{RPA}}=\delta_{AB}+\beta n_e \phi_{AB}(k). \quad (40)$$

The desired cluster expression then results from substituting Eq. (38) into Eq. (39) and comparing with Eq. (35):

$$S_{ABC}(\mathbf{k}',t'=0;\mathbf{k}'',t''=0)|_{\mathrm{RPA}} = \sum_D S_{AD}(\mathbf{k})S_{DB}(\mathbf{k}')S_{DC}(\mathbf{k}''). \quad (41)$$

As an aside, we observe that the RPA expression (39) exhibits the triangle symmetry (34). The structure functions in Eq. (41) can be traded for equilibrium ternary and pair correlation functions via Eq. (33) and

$$S_{ABC}(\mathbf{k}',t'=0;\mathbf{k}'',t''=0) = \delta_{AB}\delta_{AC}+n_e\delta_{AB}g_{AC}(\mathbf{k}'')+n_e\delta_{AC}g_{AB}(\mathbf{k}') + n_e\delta_{BC}g_{AB}(\mathbf{k})+n_e^2 h_{ABC}(\mathbf{k}',\mathbf{k}''), \quad (42)$$

where h_{ABC} is a layer-space matrix element of the ternary correlation function. Equations (41), (42), and (33) then give

$$h_{ABC}(\mathbf{k}',\mathbf{k}'')=g_{AC}(\mathbf{k})g_{BC}(\mathbf{k}')+g_{AB}(\mathbf{k})g_{CB}(\mathbf{k}'') + g_{BA}(\mathbf{k}')g_{CA}(\mathbf{k}'') + \sum_D n_e g_{AD}(\mathbf{k})g_{BD}(\mathbf{k}')g_{CD}(\mathbf{k}'') \quad (\mathbf{k}=\mathbf{k}'+\mathbf{k}''). \quad (43)$$

This is the equal-density multilayer matrix generalization of the lowest-order (in the coupling parameter) Mayer cluster expansion derived for the 3D OCP by Salpeter [25] using equilibrium statistical mechanics and by O'Neil and Rostoker [26] and Lie and Ichikawa [27] solving the Born-Bogoliubov-Green-Kirkwood-Yvon hierarchy equations.

VI. CONCLUSIONS AND DISCUSSION

This paper establishes quadratic fluctuation-dissipation relations for the unmagnetized, multilayer OCP in the classical domain; layer populations and spacings between adjacent layers need not be equal. The dynamical QFDT relations, similarly to their OCP and BIM counterparts, connect the layer-space matrix elements of a single equilibrium three-point correlation function to triangle-symmetric [see Eqs. (30a), (30b)] combinations of three quadratic-response-function matrix elements. The principal results are displayed as dynamical QFDT relations (25), (27), and (29), static QFDT (35), and k-space Mayer cluster expansions (41) and (43) (valid only for equal-density multilayers). The external density-response-matrix elements in QFDT (29) [and (31)] can be traded for screened density-response-matrix elements; for equal-density bilayer and superlattice plasmas, the trade is made via Eqs. (19) and (22).

Calculations based on the QFDT-VAA (velocity-average approximation) kinetic equation formalism [20,21] for BIM plasmas affirm the existence of a remarkable positive shift in the plasma frequency predicted by Baus [28] at weak coupling and by Hansen, McDonald, and Vieillefosse [29] at strong coupling. The isomorphism between the multicomponent plasma in three dimensions and the multilayer OCP suggests that the latter should exhibit the same kind of coupling-dependent positive shift—a long-wavelength energy gap—in the acoustic excitations. This correlation-induced gap was predicted by Golden, Kalman, and coworkers for type-I classical bilayer and superlattice plasmas [12] using the quasilocalized charge approximation (QLCA) [30]. QFDT Eq. (29) now makes it possible to develop a multilayer QFDT-VAA kinetic-equation-based approximation scheme for the purpose of confirming the existence of the $k=0$ energy gap and providing new information about its dependence on the interlayer and intralayer coupling parameters—a vital detail that is missing in the QLCA description. The development of such an approximation scheme is underway [31].

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