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Dynamical structure functions for charged particle bilayers and superlattices

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Abstract
A modified Feynman construction with a zero-frequency central peak is used to model the dynamical structure functions for layered charged particle systems. This construction recognizes the affinity between layered and multicomponent systems. It also guarantees the simultaneous satisfaction of all three frequency-moment sum rules. The frequencies and spectral weights of the long-wavelength collective excitations and the strength of the diffusive central peak are calculated for arbitrary degeneracy.

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1. Introduction

Over the past decade, interest in the static and dynamic properties of layered charged particle systems in the strongly coupled Coulomb liquid and solid phases has been stimulated by experimental activities in the areas of strongly coupled plasma physics and condensed matter plasmas. In strongly coupled plasma physics, there are the seminal NIST/Boulder experiments [1] in which laser-cooled classical ions in a cryogenic trap spontaneously organize themselves into layered structures in highly correlated liquid and solid phases. In condensed matter plasmas, advances in semiconductor nanotechnology have made it possible to routinely fabricate multiple quantum well structures of parallel electronic layers in a strongly correlated liquid phase [2].

On the theoretical/computational side, in the case of the classical bilayer, the equilibrium structure of the correlation-dominated Coulomb solid [3] and liquid [4] phases is now well understood. In the quantum domain, information on the intralayer and interlayer static structure functions and pair correlation functions has recently become available for $r_s \leqslant 5$ [5]. For both bilayers and superlattices, the spectrum of collective excitations in the random-phase-approximation (RPA) has been known for a long time [6, 7], but the analysis of the effect

of particle correlations beyond the RPA, both in the weak coupling [5] and strong coupling [8, 9] regimes is more recent. A remarkable effect that emerges in the latter regime is the development of a long-wavelength frequency gap in the out-of-phase collective modes [8, 9]. Recent molecular dynamics simulations of the classical bilayer by Donko *et al* [10] confirm the predicted collective mode dispersion including the frequency gap.

The in-phase and out-of-phase dynamical structure functions should, in principle, provide all the information on the collective excitations and static structure functions in charged particle multilayers over the entire classical to quantum domain. In this paper, we review and extend a novel approach to the construction of the dynamical structure functions that makes it possible to explicitly track its evolution with temperature all the way down to the $T = 0$ quantum limit. Central to this approach is the introduction of a zero-frequency central diffusive peak so that all three principal frequency-moment sum rules can be satisfied.

The paper has three main objectives: first we will formulate compressibility sum rules for charged particle bilayers and superlattices over the entire classical to quantum domain. Second, we will use the compressibility rules and the already established classical/quantum third-frequency-moment and *f*-sum rules to analyse the long-wavelength behaviour of the dynamical structure functions. Finally, by way of the newly constructed dynamical structure functions, we will calculate both the oscillation frequencies and spectral weights of the collective modes along with the strength of the diffusive central peak. Some of the results of the bilayer calculations appearing in this paper are reported by the authors elsewhere [11].

The bilayer and superlattice calculations are displayed side-by-side in this work in order to highlight the similarities and differences between the two configurations.

The paper proceeds according to the following plan. In section 2, we introduce the relevant response functions. Perfect screening and compressibility sum rules are formulated in section 3. In section 4, we postulate Feynman-like representations for the dynamical structure functions. Conclusions are drawn in section 5.

2. Response functions

We consider a charged particle multilayer described by a model that consists of a stack of N equal-density two-dimensional (2D) charged particle layers embedded in a dielectric substrate; d is the distance between adjacent layers. Each layer contains a 2D electron fluid ($Z = -1$) neutralized by a rigid uniform positive background. No restriction is placed on the temperature that can range from the $T = 0$ degenerate limit to the $T \to \infty$ classical limit. No tunnelling is considered. We will address the two configurations that have attracted the most experimental and theoretical interest, namely the charged particle bilayer ($N = 2$) and the superlattice ($N \to \infty$).

We define $\tilde{\Gamma} = [e^2/(a\varepsilon_r E_0)][1 - \exp(-\beta E_0)]$, which turns out to be the measure of the coupling strength for arbitrary degeneracy; $E_0 = \pi n \hbar^2/m$ is the zero-temperature Fermi energy of the non-interacting 2D electron gas, $a = 1/\sqrt{\pi n}$ is the 2D Wigner–Seitz radius and ε_r is the dielectric constant of the substrate. Note that for $T \to \infty$, $\tilde{\Gamma} = \Gamma = \beta e^2/a\varepsilon_r$, the familiar classical coupling constant, and for $T = 0$, $\tilde{\Gamma} = r_s = a/a_B$; $a_B = \varepsilon_r \hbar^2/(me^2)$ is the Bohr radius.

The formalism for the description of the equal-density bilayer is identical to that of a two-component system: the two layers can be regarded as two 'species' (1 and 2), with interaction matrix

$$\phi(k) = \frac{2\pi e^2}{\varepsilon_r k} \begin{bmatrix} 1 & \mathrm{e}^{-kd} \\ \mathrm{e}^{-kd} & 1 \end{bmatrix}.$$

Then with the aid of $\phi(k)$ and the screened (total) density response matrix, $\bar{\chi}(\mathbf{k})$, the dielectric matrix $\varepsilon(\mathbf{k})$, its inverse $\eta(\mathbf{k})$ and the full (external) density response matrix $\chi(\mathbf{k})$ can be constructed from

$$\varepsilon(\mathbf{k}) = \mathbf{I} - \phi(k)\bar{\chi}(\mathbf{k}) \qquad \chi(\mathbf{k}) = \bar{\chi}(\mathbf{k})\eta(\mathbf{k}) \qquad \eta(\mathbf{k}) = \mathbf{I} + \phi(\mathbf{k})\chi(\mathbf{k}) \quad (1)$$

where $\mathbf{I}$ is the (2×2) identity matrix.

In the case of the infinite superlattice, the periodic structure of the system allows one to replace the layer-space matrix formalism with a scalar one by introducing a Fourier transformation along the superlattice axis, e.g., $\bar{\chi}(\mathbf{k}, q) = \sum_m \bar{\chi}_{m0}(\mathbf{k}) \exp[-\mathrm{i}q z_m]$; $z_m = md (m = 0, \pm 1, \pm 2, \ldots)$ locates the mth layer above or below the $m = 0$ reference layer. We introduce the interaction potential $\phi(k, q) = [2\pi e^2/\varepsilon_r k]F(k, q)$, where $F(k, q) = \sinh kd \big/ (\cosh kd - \cos qd)$ is the superlattice form factor [6]. The superlattice counterparts of equations (1) are

$$\begin{aligned} &\varepsilon(\mathbf{k}, q) = 1 - \phi(k, q)\bar{\chi}(\mathbf{k}, q) \qquad \chi(\mathbf{k}, q) = \bar{\chi}(\mathbf{k}, q)\eta(\mathbf{k}, q) \\ &\eta(\mathbf{k}, q) = 1 + \phi(k, q)\chi(\mathbf{k}, q). \end{aligned} \quad (2)$$

3. Perfect screening and compressibility rules

The perfect screening sum rule for the bilayer follows from the fact that a charged impurity placed in one of the layers (say layer 1) is screened so that by the combined effect of all the layers, equal but negative charge is generated to surround it. The polarization charges, ρ_1 and ρ_2 surrounding the impurity with charge $Ze = +1$ in layer 1 are, respectively, $\rho_1(\mathbf{k}) = \eta_{11}(\mathbf{k}) - 1$ and $\rho_2(\mathbf{k}) = \eta_{12}(\mathbf{k})$. The total charges in the layers are $\rho_1(k = 0) + 1$ and $\rho_2(k = 0)$. Thus the perfect screening requirement [12]

$$\eta_{11}(k = 0) + \eta_{12}(k = 0) = 0. \quad (3)$$

$\eta_{11}(k = 0)$ and $\eta_{12}(k = 0)$ can assume any positive or negative values compatible with (3).

For the infinite superlattice, a similar such argument leads to

$$\eta(k = 0, q = 0) = \sum_m \eta_{m0}(k = 0) = 0. \quad (4)$$

Perfect screening sum rules (3) and (4) are, of course, statistics independent.

The Stillinger–Lovett (SL) condition that relates directly to the equilibrium pair correlation function imposes quite similar statistics-independent requirements on the static structure functions:

$$S_{11}(k = 0) + S_{12}(k = 0) = 0 \qquad \text{bilayer} \quad (5)$$

$$S(k = 0, q = 0) = \sum_m S_{m0}(k = 0) = 0 \qquad \text{superlattice.} \quad (6)$$

While the derivation of compressibility rules based on the balancing of a screened perturbing field by pressure gradients in the respective layers is straightforward, its formulation in a manner that *explicitly* encompasses both the classical and quantum domains is possible only for the 2D non-interacting electron gas [13]. Here the $k = 0$ value of the static Lindhard function and inverse isothermal compressibility are given by

$$\chi_0 = \bar{\chi}_0(k = 0) = -\frac{n}{E_0}[1 - \exp(-\beta E_0)] \quad (7)$$

$$\left[\frac{\partial P_0}{\partial n}\right]_T = \frac{E_0}{1 - \exp(-\beta E_0)}. \quad (8)$$

P_0 is the pressure of the non-interacting 2D electron gas.

Addressing first the bilayer, the resulting compressibility rule is expressed in terms of the screened (total) density response matrix $\bar{\chi}$ as

$$\bar{\chi}(k=0) = \chi_0 \mathbf{L}^{-1} \tag{9}$$

$$\mathbf{L} = (L_{ij}) = \begin{bmatrix} L & N \\ N & L \end{bmatrix} \qquad L_{ij} = \frac{[\partial P_i/\partial n_j]_T}{[\partial P_0/\partial n]_T}. \tag{10}$$

$L = L_{11} = L_{22}$ and $N = L_{12} = L_{21}$ are the direct- and trans-inverse compressibility coefficients, respectively. The superlattice counterparts of (9) and (10) are

$$\bar{\chi}(k=0,q) = \frac{\chi_0}{L(q)} \tag{11}$$

$$L(q) = \frac{1}{[\partial P_0/\partial n]_T} \sum_m \left[\frac{\partial P_m}{\partial n_0}\right]_T \exp(-\mathrm{i}q z_m). \tag{12}$$

For the bilayer, the information available in equation (9) is sufficient to generate the elements of the dielectric matrix to $O(\bar{k})$, since $\bar{\chi}_{ij}(\mathbf{k})$ is expected to be an analytic function of k to start with an $O(\bar{k}^2)$ term only. With this proviso, we obtain

$$\begin{aligned} \varepsilon_{11}(k\to 0) &= \frac{2\tilde{\Gamma}}{\bar{k}}\frac{1}{L+N} + 1 + 2\tilde{\Gamma}\bar{d}\frac{N}{L^2-N^2} + O(\bar{k}) \\ \varepsilon_{12}(k\to 0) &= \frac{2\tilde{\Gamma}}{\bar{k}}\frac{1}{L+N} - 2\tilde{\Gamma}\bar{d}\frac{L}{L^2-N^2} + O(\bar{k}). \end{aligned} \tag{13}$$

We note that $\varepsilon(\mathbf{k})$ and all other physical quantities can be diagonalized by rotating into the space spanned by the in-phase (+) and out-of-phase (−) directions: the resulting matrix elements are $\varepsilon_\pm(\mathbf{k}) = \varepsilon_{11}(\mathbf{k}) \pm \varepsilon_{12}(\mathbf{k})$, $\chi_\pm(\mathbf{k}) = \chi_{11}(\mathbf{k}) \pm \chi_{12}(\mathbf{k})$, etc.

The elements of the inverse dielectric matrix follow from equation (13):

$$\eta_{11}(k=0) = -\eta_{12}(k=0) = \frac{1}{2}\frac{L-N}{L-N+2\tilde{\Gamma}\bar{d}} \tag{14}$$

[12, 14] in agreement with the perfect screening sum rule (3). With increasing layer separation, N should rapidly vanish and L should approach a value L_{2D} appropriate for the isolated 2D layer. For the classical bilayer, $L - N = 1$ at $d = 0$ since, in this limit, N exactly cancels the correlational contribution to L.

In the case of the superlattice, the lowest-order correction to equation (11) expression for $\bar{\chi}(\bar{k}=0,q)$ is at most an $O(\bar{k}^2)$ term. With this stipulation, the small-k expansion of equation (2) results in the 'in-phase' ($q = 0$) and 'out-of-phase' ($q \neq 0$) dielectric response functions

$$\varepsilon(\bar{k}\to 0, q=0) = \frac{1}{L(q=0)}\left[\frac{4\tilde{\Gamma}}{\bar{k}^2\bar{d}} + L(q=0) + \frac{\tilde{\Gamma}\bar{d}}{3}\right] + O(\bar{k}^2) \tag{15}$$

$$\varepsilon(\bar{k}\to 0, q\neq 0) = 1 + \frac{2\tilde{\Gamma}\bar{d}}{L(q)(1-\cos qd)} + O(\bar{k}^2). \tag{16}$$

Equation (15) is in agreement with the perfect screening sum rule (4). We note the manifest similarity between equations (15) and (16) and their bilayer counterparts $\varepsilon_\pm(k\to 0)$ formed from equations (13).

We turn now to the derivation of the compressibility rules for the full density response functions. For the bilayer, one finds

$$\chi_{\pm}(k \to 0) = \chi_0 X_{\pm}(k \to 0) \tag{17}$$

$$X_{+}(k \to 0) = \frac{\bar{k}}{4\tilde{\Gamma}} - (L + N - 2\tilde{\Gamma}\bar{d})\left[\frac{\bar{k}}{4\tilde{\Gamma}}\right]^2 + O(\bar{k}^3) \tag{18}$$

$$X_{-}(k \to 0) = \frac{1}{L - N + 2\tilde{\Gamma}\bar{d}} + \frac{4\tilde{\Gamma}^2\bar{d}^2}{[L - N + 2\tilde{\Gamma}\bar{d}]^2}\frac{\bar{k}}{4\tilde{\Gamma}} + O(\bar{k}^2). \tag{19}$$

The corresponding in-phase ($q = 0$) and out-of-phase ($q \neq 0$) compressibility rules for the superlattice are

$$\chi(k \to 0, q) = \chi_0 X(k \to 0, q) \tag{20}$$

$$X(k \to 0, q = 0) = \frac{\bar{k}^2\bar{d}}{4\tilde{\Gamma}} - \left[L(0) + \frac{\tilde{\Gamma}\bar{d}}{3}\right]\left[\frac{\bar{k}^2\bar{d}}{4\tilde{\Gamma}}\right]^2 \tag{21}$$

$$X(k \to 0, q \neq 0) = \frac{1 - \cos qd}{L(q)(1 - \cos qd) + 2\tilde{\Gamma}\bar{d}} + O(\bar{k}^2). \tag{22}$$

As expected, the classical limit of the in-phase bilayer expression (18) closely resembles that of the isolated 2D electron plasma layer of density $2n$ when (18) is recast in terms of the dimensionless wavenumber $k_{2D} = k/\kappa_{2D} = \bar{k}/(2\Gamma)$; $\kappa_{2D} = (2\pi ne^2\beta)/\varepsilon_r$ is the 2D Debye wavenumber. By the same token, the classical limit of the in-phase superlattice expression (21) closely resembles that of the 3D one-component plasma (OCP) with density n/d when (21) is recast in terms of dimensionless wavenumber: $k_{3D}^2 = k^2/\kappa_{3D}^2 = (\bar{k}^2\bar{d})/(4\Gamma)$; $\kappa_{3D} = \sqrt{(4\pi ne^2\beta)/(\varepsilon_r d)}$ is the 3D Debye wavenumber.

4. Structure functions and frequency-moment sum rules

We turn now to the analysis of the in-phase and out-of-phase dynamical structure functions. It is known that in the long-wavelength ($k \to 0$) domain of interest, the Feynman representation of the dynamical structure function in terms of δ-functions at the collective mode frequencies provides in most cases a reasonable physical picture. Here we extend this representation to include a $\omega = 0$ diffusive peak. Accordingly,

$$S_{\pm}(k \to 0, \omega) = \pi\,\{2p_{\pm}(\mathbf{k})\delta(\omega) + q_{\pm}(\mathbf{k})[\delta(\omega - \omega_{\pm}(\mathbf{k})) + \delta(\omega + \omega_{\pm}(\mathbf{k}))]\} \tag{23}$$

for the bilayer (see also [15]) and

$$S(k \to 0, q, \omega) = \pi\,\{2P(\mathbf{k}, q)\delta(\omega) + Q(\mathbf{k}, q)[\delta(\omega - \omega(\mathbf{k}, q)) + \delta(\omega + \omega(\mathbf{k}, q))]\} \tag{24}$$

for the superlattice; $q_{\pm}(\mathbf{k})$, $Q(\mathbf{k}, q)$ are the weight factors of the collective peaks and $\omega_{\pm}(\mathbf{k})$, $\omega(\mathbf{k}, q)$ are the collective mode frequencies; $p_{\pm}(q)$, $P(\mathbf{k}, q)$ are the weight factors for the central peak. While the inclusion of the central peak in the present context is novel, it has its antecedents in the literature. In his comment the reference [16] analysis of the dispersion relation for classical monatomic liquids and amorphous solids, Knipp [17] convincingly argues that the central 'Rayleigh' peak must be included to take account of thermal diffusion. Indeed, what makes Knipp's argument all the more compelling is the fact that the three-peak representation necessarily implements all three fundamental frequency-moment sum rules to *uniquely* specify the spectral weights $p_{\pm}(\mathbf{k})$, $q_{\pm}(\mathbf{k})$ of the central and collective peaks,

respectively, and the oscillation frequencies $\omega_{\pm}(\mathbf{k})$ of the collective modes. By contrast, in the more traditional Feynman-like representation for the OCP featuring only the two collective peaks, one of the three sum rules must be ignored as though it simply never existed.

In this work, the introduction of the central peak is further motivated by the observation that in multicomponent systems, hydrodynamic diffusion dominates the low-frequency behaviour and its representation in $S(\mathbf{k}, \omega)$ is indispensable in view of the fact that $S(k = 0) \neq 0$. Now, charged particle bilayers and superlattices are akin to multicomponent systems; thus, their central peaks are at least on a par with the resonances representing the out-of-phase longitudinal collective modes. Note that the representation of the central peak through a δ-function should not be problematic: only the integral over the peak matters in the forthcoming analysis. On the other hand, one should realize that the δ-function representation at the collective mode frequencies implies that the modes are undamped which, although a limitation, is still an acceptable approximation: see discussion below equation (46).

The fundamental sum rules for the bilayer and superlattice are customarily stated in terms of the respective frequency moments

$$\langle\omega^s\rangle_{\pm}(\mathbf{k}) = (1/\pi)\int_{-\infty}^{\infty} \mathrm{d}\omega\omega^s \,\mathrm{Im}\,\chi_{\pm}(\mathbf{k},\omega) \qquad \langle\omega^s\rangle(\mathbf{k},q) = (1/\pi)\int_{-\infty}^{\infty} \mathrm{d}\omega\,\omega^s\,\mathrm{Im}\,\chi\,(\mathbf{k},q,\omega).$$

Noting that the $s = 1$ f-sum rule for $\langle\omega\rangle_{\pm}(\mathbf{k}) = \langle\omega\rangle(\mathbf{k},q) = -n\bar{k}^2/(ma^2)$ is the same for both systems, the remaining compressibility ($s = -1$) and third-frequency-moment [18] ($s = 3$) sum rules that the in-phase and out-of-phase dynamical structure functions (23) and (24) are required to satisfy are

$$\langle\omega^{-1}\rangle_{\pm}(\bar{k}\to 0) = \chi_{\pm}(\bar{k}\to 0) \qquad \text{bilayer} \tag{25}$$

$$\langle\omega^{-1}\rangle(k\to 0,q) = \chi(k\to 0,q) \qquad \text{superlattice} \tag{26}$$

$$\langle\omega^3\rangle_{\pm}(\mathbf{k}) = -\frac{n\bar{k}^2}{ma^2}\left\{\omega_{\pm 0}^2(k) + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 + D_{\pm}(\mathbf{k}) + \left[\frac{\hbar\,\bar{k}^2}{2ma^2}\right]^2\right\} \qquad \text{bilayer} \tag{27}$$

$$\langle\omega^3\rangle(\mathbf{k},q) = -\frac{n\bar{k}^2}{ma^2}\left\{\omega_0^2(k,q) + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 + D(\mathbf{k},q) + \left[\frac{\hbar\bar{k}^2}{2ma^2}\right]^2\right\} \qquad \text{superlattice.} \tag{28}$$

Here, $\omega_{\pm 0}^2(k) = \frac{n\bar{k}^2}{ma^2}\phi_{\pm}(k) = \omega_{2\mathrm{D}}^2(k)\,[1 \pm \exp(-kd)]$ and $\omega_0^2(k,q) = \frac{n\bar{k}^2}{ma^2}\phi(k,q)$ are the respective bilayer and superlattice mean field frequencies; $\omega_{2\mathrm{D}} = \sqrt{(2\pi ne^2k)/(\varepsilon_r m)}$ is the plasma frequency of a single isolated layer; $\langle E_{\mathrm{kin}}\rangle$ is the average kinetic energy per particle for the interacting system. The D-functions take account of the exchange-correlation contributions to the third-frequency-moment sum rule coefficients through the $S_{ij}(k)$ static structure functions. For the bilayer, to lowest order in $\bar{k}$,

$$D_+(k\to 0) = -\omega_0^2\gamma_+\bar{k}^2 \tag{29}$$

$$D_-(k\to 0) = \omega_0^2 g^2 - \omega_0^2\gamma_-\bar{k}^2 \tag{30}$$

$$g = \sqrt{\frac{1}{2}\left|\int_0^{\infty}\mathrm{d}x\,x^2 S_{12}(x)\exp(-x\bar{d})\right|} \tag{31}$$

$$\gamma_{\pm} = -\frac{5}{32}\int_0^{\infty}\mathrm{d}x[S_{11}(x)-1] \mp \frac{5}{32}\int \mathrm{d}x\left[1 - \frac{11}{5}x\bar{d} + \frac{3}{5}x^2\bar{d}^2\right]S_{12}(x)\exp(-x\bar{d}). \tag{32}$$

$\omega_0 = \sqrt{2e^2/(\varepsilon_r m a^3)}$ is a nominal 2D plasma frequency, $x = k'a$, and $\bar{d} = d/a$. For the superlattice, one similarly finds

$$D(k \to 0, q = 0) = -\omega_0^2 \gamma(q = 0)\bar{k}^2 \tag{33}$$

$$D(k \to 0, q \neq 0) = \omega_0^2 [g(q)]^2 - \omega_0^2 \gamma(q)\bar{k}^2 \tag{34}$$

$$g(q) = \sqrt{\frac{1}{2}\left|\sum_{m=1}^{\infty}[1 - \cos(qmd)]\int_0^{\infty} \mathrm{d}x\, x^2 S_{m0}(x)\exp(-xmd)\right|} \tag{35}$$

$$\gamma(q) = -\frac{5}{32}\int_0^{\infty}\mathrm{d}x[S_{00}(x) - 1] - \frac{5}{32}\int_0^{\infty}\mathrm{d}x \sum_{m\neq 0}\left[1 - \frac{11}{5}x\,|m|\,\bar{d} + \frac{3}{5}x^2\,|m|^2\,\bar{d}^2\right] \times \cos(qmd) S_{m0}(x)\exp(-x|m|\bar{d}). \tag{36}$$

The combination of equations (25)–(36) and the fluctuation-dissipation theorems

$$\mathrm{Im}\,\chi_{\pm}(\mathbf{k}, \omega) = -\frac{n}{\hbar}\tanh\left(\frac{\beta\hbar\omega}{2}\right) S_{\pm}(\mathbf{k}, \omega) \qquad \text{bilayer} \tag{37}$$

$$\mathrm{Im}\,\chi(\mathbf{k}, q, \omega) = -\frac{n}{\hbar}\tanh\left(\frac{\beta\hbar\omega}{2}\right) S(\mathbf{k}, q, \omega) \qquad \text{superlattice} \tag{38}$$

yields the weight factors

$$p_{\pm}(k \to 0) = \frac{1 - \exp(-\beta E_0)}{\beta E_0} X_{\pm}(\mathbf{k}) - \frac{\bar{k}^2}{\beta m a^2 \omega_{\pm}(\mathbf{k})} \tag{39}$$

$$q_{\pm}(k \to 0) = \frac{\hbar\bar{k}^2}{2ma^2\omega_{\pm}(\mathbf{k})\tanh[\beta\hbar\omega_{\pm}(\mathbf{k})/2]} \qquad \text{bilayer} \tag{40}$$

$$P(k \to 0, q) = \frac{1 - \exp(-\beta E_0)}{\beta E_0} X(\mathbf{k}, q) - \frac{\bar{k}^2}{\beta m a^2 \omega^2(\mathbf{k}, q)} \tag{41}$$

$$Q(\bar{k} \to 0, q) = \frac{\hbar\bar{k}^2}{2ma^2\omega(\mathbf{k}, q)\tanh[\beta\hbar\omega(\mathbf{k}, q)/2]} \qquad \text{superlattice} \tag{42}$$

and long-wavelength in-phase and out-of-phase plasmon oscillation frequencies

$$\omega_+^2(k \to 0) = 2\omega_0^2\bar{k}\left[1 - \frac{\bar{k}\bar{d}}{2}\right] + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 - \omega_0^2\gamma_+\bar{k}^2 \tag{43}$$

$$\omega_-^2(k \to 0) = \omega_-^2(k = 0) + \omega_0^2\bar{k}^2\bar{d}\left[1 - \frac{\bar{k}\bar{d}}{2}\right] + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 - \omega_0^2\gamma_-\bar{k}^2 \qquad \text{bilayer} \tag{44}$$

$$\omega^2(k \to 0, q = 0) = \omega_{3D}^2\left[1 + \frac{\bar{k}^2\bar{d}^2}{12}\right] + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 - \omega_0^2\gamma(q = 0)\bar{k}^2 \tag{45}$$

$$\omega^2(k \to 0, q \neq 0) = \omega^2(k = 0, q \neq 0) + \frac{\omega_0^2\bar{d}}{1 - \cos\,qd}\bar{k}^2 + \frac{3\langle E_{\mathrm{kin}}\rangle}{ma^2}\bar{k}^2 - \omega_0^2\gamma(q)\bar{k}^2 \qquad \text{superlattice.} \tag{46}$$

Note the 2D OCP-like character of the in-phase plasmon frequency (43) and the 3D OCP-like character of its superlattice counterpart equation (45) ($\omega_{3\mathrm{D}} = \sqrt{4\pi ne^2/(\varepsilon_r md)}$

is the bulk plasma frequency); the exchange-correlation γ coefficients act to modify the slopes of the dispersion curves in the usual manner. Most importantly, we observe that the out-of-phase plasmon frequencies (44), (46) develop a finite-frequency 'energy gap' at $k = 0$ characteristic of optic mode behaviour, that is, $\omega_-(k = 0) = \omega_0 g \neq 0$ for the bilayer and $\omega(k = 0, q \neq 0) = \omega_0 g(q) \neq 0$ for the superlattice. This is in marked contrast to the customary acoustic dispersion predicted by the RPA (formally recovered by setting $D_- = 0$ in (27) and $D(q \neq 0) = 0$ in (28)). The energy gap was predicted some time ago first for strongly coupled classical superlattices [8] and later for classical bilayers [9] using the quasilocalized charge approximation (QLCA) or a third-frequency-moment sum rule equivalent of the QLCA. Recent molecular dynamics simulations have now confirmed its existence in classical bilayers [10]. The results of the present analysis go much further in that they predict that the finite-frequency energy gap persists for *arbitrary* coupling strengths and *over the entire classical to quantum domain all the way down to zero temperature*.

The representations (23) and (24) for the dynamical structure functions, of course, preclude the possibility of describing the damping of the collective modes. Some qualitative assessment, however, can be made and here we confine our attention to the out-of-phase plasmon modes only. The two major damping mechanisms to be considered are Landau damping (single-particle excitations) and collisional damping (multiple pair excitations). According to the QLCA analysis of [9(a)], as long as the layer separation is not too large ($d < 1.5a$), for the small-k values of this work the out-of-phase plasmon is well outside the pair continuum and is thus immune to Landau damping. In the strong coupling regime, collisional damping is also operative; however, it becomes prominent only at higher k values well beyond the long-wavelength domain of the present theory.

Finally, the knowledge of the in-phase and out-of-phase dynamical structure functions makes it possible to calculate the corresponding static structure functions via $S_\pm(k \to 0) = p_\pm(k \to 0) + q_\pm(k \to 0)$ and $S(k \to 0, q) = P(k \to 0, q) + Q(k \to 0, q)$. Although it is not manifest from equations (39) and (41), the amplitude of the central peak $p_\pm(k \to 0)$, $P(k \to 0,\ q)$ is always positive, as it should be (see table 1), further attesting to the soundness of representations (23) and (24) at long wavelengths.

The results of the analysis of the in-phase and out-of-phase dynamical structure functions in the classical and quantum domains are summarized in table 1. The following summarizes the salient features of the table.

- The fact that the central peak is related to thermal diffusion is clearly demonstrated by its strong temperature dependence: while in the classical domain (high temperatures) the central peak saturates at a value dictated by the compressibility sum rule, in the quantum domain (low temperatures), it is proportional to T both for the in-phase and out-of-phase structure functions and vanishes at $T = 0$.
- The strength of the out-of-phase central peak always dominates the strength of the in-phase central peak. This is especially pronounced in the case of the superlattice.
- At low temperatures, the in-phase and out-of-phase static structure functions are dominated by their collective peaks. In contrast, at high temperatures this dominance prevails only for $S_+(k \to 0)$ and $S(k \to 0, q = 0)$, whereas for $S_-(k \to 0)$ and $S(k \to 0, q \neq 0)$, the main contribution shifts to the central peak.
- In the classical limit, the table 1 formulae for the bilayer and superlattice static structure functions are *exact* [14, 19] in that they can be established directly from the compressibility rule and the static fluctuation-dissipation theorem without the need to invoke the Feynman-like representations (22) and (23).

Table 1. The table shows the leading terms (in powers of k) for the in-phase and out-of-phase static structure functions and the strengths of the central and collective peaks of the corresponding dynamical structure functions; $\bar{k} = ka$, $k_{2D} = k/\kappa_{2D}$ and $k_{3D} = k/\kappa_{3D}$, here $\kappa_{2D} = 2\pi ne^2\beta/\varepsilon_r$ and $\kappa_{3D} = \sqrt{4\pi ne^2\beta/(\varepsilon_r d)}$ are 2D and 3D Debye wavenumbers.

Classical bilayer	Classical superlattice
In-phase	
$p_+(k\to 0) = [3-(L+N)-2\Gamma\gamma_+]\frac{k_{2D}^2}{4}$	$P(k\to 0, q=0) = [3-L(0)-2\Gamma\gamma(0)]k_{3D}^4$
$q_+(k\to 0) = \frac{k_{2D}}{2} - [3-2\Gamma\gamma_+ - \kappa_{2D}d]\frac{k_{2D}^2}{4}$	$Q(k\to 0, q=0) = k_{3D}^2 - \left[3-2\Gamma\gamma(0)+\frac{\kappa_{3D}^2 d^2}{12}\right]k_{3D}^4$
$S_+(k\to 0) = \frac{k_{2D}}{2} - [L+N-\kappa_{2D}d]\frac{k_{2D}^2}{4}$	$S(k\to 0, q=0) = k_{3D}^2 - \left[L(0)+\frac{\kappa_{3D}^2 d^2}{12}\right]k_{3D}^4$
Out-of-phase optical	
$p_-(k\to 0) = \frac{1}{L-N+2\Gamma\bar{d}} + \frac{\Gamma\bar{d}^2\bar{k}}{[L-N+2\Gamma\bar{d}]^2}$	$P(k\to 0, q\neq 0) = \frac{1-\cos qd}{L(q)[1-\cos qd]+2\Gamma\bar{d}} + O(\bar{k}^2)$
$q_-(k\to 0) = \frac{\bar{k}^2}{2\Gamma\bar{g}^2} + O(\bar{k}^4)$	$Q(k\to 0, q\neq 0) = \frac{\bar{k}^2}{2\Gamma[g(q)]^2} + O(\bar{k}^4)$
$S_-(k\to 0) = \frac{1}{L-N+2\Gamma\bar{d}} + \frac{\Gamma\bar{d}^2\bar{k}}{[L-N+2\Gamma\bar{d}]^2}$	$S(k\to 0, q\neq 0) = \frac{1-\cos qd}{L(q)[1-\cos qd]+2\Gamma\bar{d}} + O(\bar{k}^2)$
Zero-temperature bilayer	Zero-temperature superlattice
In-phase	
$p_+(k\to 0) = 0$	$P(k\to 0, q=0) = 0$
$q_+(k\to 0) = \frac{\bar{k}^{3/2}}{4\sqrt{r_s}} + O\left(\bar{k}^{5/2}\right)$	$Q(k\to 0, q=0) = \frac{\bar{k}^2\bar{d}}{4\sqrt{r_s}} + O(\bar{k}^4)$
$S_+(k\to 0) = \frac{\bar{k}^{3/2}}{4\sqrt{r_s}} + O(\bar{k}^{5/2})$	$S(k\to 0, q=0) = \frac{\bar{k}^2\bar{d}}{4\sqrt{r_s}} + O(\bar{k}^4)$
Out-of-phase optical	
$p_-(k\to 0) = 0$	$P(k\to 0, q\neq 0) = 0$
$q_-(k\to 0) = \frac{\bar{k}^2}{4g\sqrt{r_s}} + O(\bar{k}^4)$	$Q(k\to 0, q\neq 0) = \frac{\bar{k}^2}{4g(q)\sqrt{r_s}} + O(\bar{k}^4)$
$S_-(k\to 0) = \frac{\bar{k}^2}{4g\sqrt{r_s}} + O(\bar{k}^4)$	$S(k\to 0, q\neq 0) = \frac{\bar{k}^2}{4g(q)\sqrt{r_s}} + O(\bar{k}^4)$

5. Summary

In this paper, we analyse the long-wavelength behaviour of the dynamical structure functions for bilayers and superlattices. The analysis is based on a Feynman-like representation featuring a zero-frequency peak in addition to the two collective peaks. Inclusion of the central peak, on the one hand, recognizes the fact that layered systems are akin to multicomponent plasmas. On the other hand, it guarantees satisfaction of all three principal frequency-moment sum rules. Of particular significance are the compressibility sum rules for bilayers and superlattices and their similarities and differences with each other and with the OCP in two and three dimensions.

The collective mode frequencies, including the interlayer correlation-induced energy gap in the out-of-phase spectrum, that result from the dynamical structure functions persist over the entire temperature domain down to zero temperature. The recent molecular dynamics simulations of Donko *et al* [10] confirm the existence of the energy gap in the classical regime. In the quantum domain as far as experimental verification is concerned, the existing observations on semiconductor bilayers at small-r_s and high-$\bar{k}$ values can be reconciled with the small energy gap that would exist in this parameter range [20]. The ultimate verification of the existence of the energy gap at zero temperature awaits Raman scattering experiments on high-r_s multiple quantum well structures.

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Appendix

In this section, we discuss the interpretation of the out-of-phase finite-frequency energy gap in the $d = 0$ limit. It is instructive to parallel the situation with the behaviour of the phonon spectrum calculated in [3] for the classical bilayer Wigner crystal. The spectrum exhibits two $k = 0$ energy gaps—one for the longitudinal mode and one for the transverse mode—that persist when the two layers are merged into one. In the case of the strongly coupled bilayer liquid where the isotropy of the system dictates that the two gap frequencies assume the same value, our calculation based on the gap coefficient (31) indicates that the finite-frequency gap persists as well in the $d = 0$ limit. In this limit, the pair correlation functions $h_{11}(r)$ and $h_{12}(r)$ become identical, whence

$$\omega_-(k = 0, d = 0) = \omega_0 \sqrt{\frac{1}{2}\left|\int_0^\infty \mathrm{d}x\, x^2\,[S_{11}(x) - 1]\right|}. \tag{A1}$$

Equation (A1) is identical to the $\omega(k \to \infty)$ Einstein frequency for the isolated 2D layer having density n [9(a), 9(d), 21]. However, the question then arises: how can (A1) be reconciled with the fact that in the strong coupling regime, the 2D electron liquid should feature only the two *in-phase* longitudinal plasmon (P) and transverse shear (S) mode frequencies, with both having the same $\omega(k = 0) = 0$ value?

To address this question it is useful to examine how this scenario plays out in the case of the classical zero-temperature bilayer crystal [3]. One first has to realize that after the two layers merge, a $k = 0$ out-of-phase phonon excitation in the bilayer *does not* become a $k = 0$ phonon in the resulting isolated 2D crystal. This is because in the staggered rectangular crystal structure prevailing at small layer separations [3], particles in the two layers occupy sites that become lattice points along alternating rows of the 2D hexagonal crystal that results when the two layers coalesce into one. Thus when the $k = 0$ P and S phonons are excited, the two layers develop two uniform, but different distributions of particle displacements that emerge as a single periodic (non-uniform) distribution in the combined 2D layer. With reference to the bilayer phonon dispersion curves displayed in [3] and to the 2D phonon dispersion curves of [22], one can then identify the $k = 0$ P and S optic frequencies at $d = 0$ with the same frequencies in the isolated 2D layer at $k = 2\pi/(\sqrt{3}a_0) = 1.905/a$. This is precisely the X-point midway between two vertices on the boundary of the hexagonal Brillouin zone (see [22], figure 1). Thus the 'persistence' of the $k = 0$ bilayer energy gap in the solid phase in the $d \to 0$ limit (as actually displayed in [3], figure 3(a)), in fact, can be well understood as a conversion into a $k \neq 0$ 2D excitation.

In the liquid phase of the bilayer, the situation is somewhat different. The source of the difference lies in the substitutional disorder [12], an inevitable consequence of the finite temperature. As a result, when the two layers merge, $h_{11}(r) = h_{12}(r)$, and the positions of the particles originating from layer 2 are no longer distinguishable from those in layer 1. Thus, in contrast to what happens in the classical zero-temperature bilayer crystal, the relative out-of-phase motion of the particles belonging to the two layers does not translate itself into a propagating 2D mode. What happens instead is rather similar to the $k = 0$ behaviour of an optic mode of a two-component system with two out-of-phase particles oscillating in a unit cell. Such an oscillation, not being part of the normal mode structure of the 2D OCP, must be regarded as being generated by the specific initial conditions that correspond to the merger of the two layers. In any case, one would expect that in the liquid phase, 'inter-species' diffusion would damp out these oscillations quickly. This is of course not apparent within the formalism of this work where damping is not properly accounted for. Thus the frequency of these oscillations as predicted by (A1) is of academic interest only. Nevertheless, the gap

coefficient (31) and its offspring equation (A1) can be understood by realizing that at $k = 0$, each layer oscillates rigidly and the relative equilibrium positions of the particles within each layer remain unaffected. Thus, forces acting on any singled out particle in one layer originate from the frozen equilibrium positions of the particles in the other layer; this is the condition for the excitation of the Einstein frequency, as given by (A1).

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