

Sum rules for electron-hole bilayer and two-dimensional point dipole systems

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We formulate and analyze the third-frequency-moment sum rules for the two-dimensional (point) dipole system (2DDS) and the mass-symmetric electron-hole bilayer (EHB) in their strongly coupled liquid phases. The former, characterized by the repulsive interaction potential $\varphi_D(r) = \mu^2/r^3$ (μ is the electric dipole moment), reasonably well approximates the latter in the $d \rightarrow 0$ limit (d is the interlayer spacing), a conjecture that is further supported by the findings of the present work. We explore the extent to which the in-phase sum rule for the closely spaced EHB may or may not reconcile with its 2DDS sum-rule counterpart. This is the main emphasis of the present work.

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I. INTRODUCTION

Satisfaction of the third-frequency-moment sum rule serves as an important gauge of the accuracy of model dynamical theories of many-body systems, particularly in the strongly correlated liquid phase. To date, third-frequency-moment sum rules have been established and extensively analyzed for a variety of charge-neutral and Coulomb fluids, most notably (i) low-temperature interacting Bose liquids (e.g., liquid ^4He) [1–3], (ii) dense Lennard-Jones fluids [4], (iii) three- and two-dimensional one-component plasmas in the classical and quantum domains [5–12], (iv) binary ionic mixtures in a neutralizing uniform background of rigid degenerate electrons [13], and (v) layered electron liquids [14–16].

Third-frequency-moment sum rules have not been formulated, however, for two companion systems that have attracted a great deal of attention in recent years: (i) the closely spaced electron-hole bilayer (EHB) and (ii) the two-dimensional dipole system (2DDS) characterized by a repulsive $1/r^3$ interaction potential. The derivation and analysis of these sum rules are the main goals of the present work.

In the EHB, the charges in the two layers have opposite polarities ($\pm e$) and, for sufficiently small layer separation d , the positive and negative charges bind to each other in dipolelike excitonic formations. Quantum [17] and classical [18] Monte Carlo (MC) simulations and molecular dynamics (MD) [19] simulations have independently confirmed the existence of the excitonic phase in the EHB in both the zero-temperature quantum and high-temperature classical domains. The Ref. [18] classical simulations have revealed the existence of four phases in the strong-coupling regime: Coulomb liquid and solid phases and dipole liquid and solid phases. The Ref. [17] quantum MC simulations distinguish three: the excitonic phase, Coulomb plasma phase, and the Wigner crystal phase.

Based on the phase diagrams [17,18], the closely spaced EHB in its dipolelike excitonic phase can be, in a fairly good approximation, modeled as a two-dimensional (2D) monolayer

of interacting point dipoles, each of mass $m = m_e + m_h$. The model 2DDS is described as a collection of N -point dipoles occupying the large but bounded area A ; $n = N/A$ is the average density in the infinitely thin layer. The dipoles are free to move in the xy plane with dipolar moment oriented in the z direction. Accordingly, the repulsive interaction potential is given by $\varphi_D(r) = \mu^2/r^3$, with μ being the electric dipole strength. In recent years, the 2DDS model has been considered by a number of investigators [20–26]. While such an idealized system has yet to be realized in the laboratory, the concept of it does open up new lines of theoretical and computer simulation explorations pertaining to the dynamics of Bose systems. The third-frequency-moment sum rule could play a pivotal role in such explorations.

The plan of the paper is as follows. In Sec. II we derive the third-frequency-moment sum rule for the 2DDS, valid for arbitrary degeneracy. At long wavelengths, a dipole interaction integral that embodies the static correlation effects via the radial distribution function $g(r)$ emerges as the dominant contribution to the sum rule in strong-coupling regimes, the regimes of interest in the present work. In Sec. III we first formulate the in-phase and out-of-phase third-frequency-moment sum rules for the symmetric EHB when the layer spacing d lies in the domain of the Coulomb liquid phase. We then initiate a procedure for accessing the $d \ll a$ domain ($a = 1/\sqrt{\pi n}$) of the dipole (bound electron-hole pairs) liquid phase. In this latter domain, we compare the resulting in-phase sum rule with its 2DDS counterpart sum rule of Sec. II. As to the EHB out-of-phase sum rule, a preliminary study, presented at the end of this paper, is expected to provide information about the internal degrees of freedom of the dipoles.

There is an intimate connection between the sum-rule coefficients of a system and its mode dispersions in the strong-coupling domain. A technique based on this relationship has been elevated to an approximation scheme for strong coupling by Iwamoto *et al.* [10] and Tkachenko and co-workers [12]. By the same token, there is a relationship between the sum-rule coefficients and the results of the quasilocalized approximation scheme [22,25,27,28], even though this latter relies on its own physical foundations. These connections will be pointed out in more detail in Sec. IV.

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II. TWO-DIMENSIONAL DIPOLE SYSTEM

In this section we establish the third-frequency-moment sum rule for the 2DDS characterized by the repulsive interaction potential $\varphi_D(r) = \mu^2/r^3$. Following the standard procedure dictated by the Kubo sum-rule theorem [10,29], we calculate the $\ell = 1, 3$ frequency moments

$$\langle \omega^\ell \rangle(\mathbf{q}) = \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \omega^\ell \text{Im}\chi(\mathbf{q}, \omega) \quad (1)$$

in the high-frequency expansion

$$\text{Re}\chi(\mathbf{q}, \omega \rightarrow \infty) = -\frac{1}{\omega^2} \langle \omega \rangle(\mathbf{q}) - \frac{1}{\omega^4} \langle \omega^3 \rangle(\mathbf{q}) - \dots \quad (2)$$

resulting from the Kramers-Kronig formula linking the real and imaginary parts of the external density response function $\chi(\mathbf{q}, \omega)$:

$$\langle n(\mathbf{q}, \omega) \rangle = \chi(\mathbf{q}, \omega) U^{\text{ext}}(\mathbf{q}, \omega). \quad (3)$$

Here $\langle n(\mathbf{q}, \omega) \rangle$ is the Fourier transform of the average density response $\langle n(\mathbf{r}, t) \rangle$ to a weak external potential energy perturbation $U^{\text{ext}}(\mathbf{r}, t)$.

In the low-temperature quantum domain, the 2DDS is characterized by the Hamiltonian

$$H = -\frac{\hbar^2}{2m} \sum_i \nabla_i^2 + \frac{1}{2} \sum_i \sum_{j \neq i} \frac{\mu^2}{|\mathbf{x}_i - \mathbf{x}_j|^3}. \quad (4)$$

The basic ingredient in the derivation of the sum rule coefficients (1) is the fluctuation-dissipation theorem

$$\chi(\mathbf{q}, \omega) = -\frac{i}{\hbar A} \int_0^\infty dt \exp(i\omega t) \langle [n_{\mathbf{q}}(t), n_{-\mathbf{q}}(0)] \rangle, \quad (5a)$$

$$\text{Im}\chi(\mathbf{q}, \omega) = -\frac{1}{2\hbar A} \int_{-\infty}^{\infty} dt \exp(i\omega t) \langle [n_{\mathbf{q}}(t), n_{-\mathbf{q}}(0)] \rangle. \quad (5b)$$

Equation (5b), when substituted into Eq. (1), results in the first two frequency moments

$$\langle \omega \rangle(\mathbf{q}) = \frac{1}{i\hbar A} \langle [\dot{n}_{\mathbf{q}}, n_{-\mathbf{q}}] \rangle, \quad (6)$$

$$\langle \omega^3 \rangle(\mathbf{q}) = \frac{1}{i\hbar A} \langle [\ddot{n}_{\mathbf{q}}, \dot{n}_{-\mathbf{q}}] \rangle. \quad (7)$$

The angular brackets denote averaging over the equilibrium ensemble; the binary products are equal-time ($t = 0$) products; $n_{\mathbf{q}} = \sum_i \exp(-i\mathbf{q} \cdot \mathbf{x}_i)$ is the Fourier transform of the local density operator in the Heisenberg representation. The textbook $\ell = 1$ f sum rule

$$\langle \omega \rangle(\mathbf{q}) = -\frac{nq^2}{m} \quad (8)$$

readily results from performing the routine commutator algebra for the Hamiltonian and local density operators.

Addressing next the evaluation of moment (7), the calculations of $\dot{n}_{\mathbf{q}}$ and $\ddot{n}_{\mathbf{q}}$ from their Heisenberg equations readily yield

$$\dot{n}_{\mathbf{q}} = \frac{i\hbar q^2}{2m} n_{\mathbf{q}} - \frac{i}{m} \sum_i [\exp(-i\mathbf{q} \cdot \mathbf{x}_i)] (\mathbf{q} \cdot \mathbf{p}_i), \quad (9)$$

$$\begin{aligned} \ddot{n}_{\mathbf{q}} = & -\frac{\hbar^2 q^4}{4m^2} n_{\mathbf{q}} + \frac{\hbar q^2}{m^2} \sum_i [\exp(-i\mathbf{q} \cdot \mathbf{x}_i)] (\mathbf{q} \cdot \mathbf{p}_i) \\ & - \frac{1}{m^2} \sum_i [\exp(-i\mathbf{q} \cdot \mathbf{x}_i)] (\mathbf{q} \cdot \mathbf{p}_i)^2 \\ & - \frac{3i\mu^2}{2m} \sum_i \sum_{j \neq i} \frac{\mathbf{q} \cdot (\mathbf{x}_i - \mathbf{x}_j)}{|\mathbf{x}_i - \mathbf{x}_j|^5} \\ & \times [\exp(-i\mathbf{q} \cdot \mathbf{x}_i) - \exp(-i\mathbf{q} \cdot \mathbf{x}_j)]. \end{aligned} \quad (10)$$

Here $\mathbf{p}_i = -i\hbar \nabla_i$ is the momentum operator. Assigning the labels 1, 2, 3, and 4, respectively, to the four right-hand-side members of (10), one can write the commutator in Eq. (7) in the form

$$[\ddot{n}_{\mathbf{q}}, \dot{n}_{-\mathbf{q}}] = \sum_{\ell=1}^4 [\ell, \dot{n}_{-\mathbf{q}}], \quad (11)$$

where

$$[1, \dot{n}_{-\mathbf{q}}] = -i \frac{N\hbar^3 q^6}{4m^3}, \quad (12)$$

$$[2, \dot{n}_{-\mathbf{q}}] = \frac{iN\hbar^3 q^6}{2m^3} + \frac{2i\hbar^2 q^4}{m^3} \sum_i (\mathbf{q} \cdot \mathbf{p}_i), \quad (13)$$

$$\begin{aligned} [3, \dot{n}_{-\mathbf{q}}] = & -\frac{iN\hbar^3 q^6}{2m^3} - \frac{2i\hbar^2 q^4}{m^3} \sum_i (\mathbf{q} \cdot \mathbf{p}_i) \\ & - \frac{3i\hbar q^2}{m^3} \sum_i (\mathbf{q} \cdot \mathbf{p}_i)^2, \end{aligned} \quad (14)$$

$$\begin{aligned} [4, \dot{n}_{-\mathbf{q}}] = & -3 \frac{i\hbar q^2 \mu^2}{m^2} \sum_i \sum_{j \neq i} \frac{1 - 5 \cos^2 \phi}{|\mathbf{x}_i - \mathbf{x}_j|^5} \\ & \times \{\exp[i\mathbf{q} \cdot (\mathbf{x}_i - \mathbf{x}_j)] - 1\} \\ & + 3 \frac{\hbar q^2 \mu^2}{m^2} \sum_i \sum_{j \neq i} \frac{\mathbf{q} \cdot (\mathbf{x}_i - \mathbf{x}_j)}{|\mathbf{x}_i - \mathbf{x}_j|^5}. \end{aligned} \quad (15)$$

Here $\cos \phi = \mathbf{q} \cdot (\mathbf{x}_i - \mathbf{x}_j) / q |\mathbf{x}_i - \mathbf{x}_j|$. Consolidating Eqs. (11)–(15) and ensemble averaging, one then obtains

$$\begin{aligned} \langle [\ddot{n}_{\mathbf{q}}, \dot{n}_{-\mathbf{q}}] \rangle = & -\frac{iN\hbar^3 q^6}{4m^3} - 3 \frac{i\hbar q^2}{m^3} \sum_i \langle (\mathbf{q} \cdot \mathbf{p}_i)^2 \rangle \\ & - \frac{i\hbar}{m^2} q_\mu q_\nu \sum_i \sum_{j \neq i} \left\langle \{\exp[i\mathbf{q} \cdot (\mathbf{x}_i - \mathbf{x}_j)] - 1\} \right. \\ & \times \left. \frac{\partial^2}{\partial x_{i\mu} \partial x_{j\nu}} \frac{\mu^2}{|\mathbf{x}_i - \mathbf{x}_j|^3} \right\rangle. \end{aligned} \quad (16)$$

The angular brackets denote an ensemble average. The 2DDS third-frequency-moment sum-rule coefficient follows from Eqs. (7) and (16):

$$\langle \omega^3 \rangle(\mathbf{q}) = -\frac{nq^2}{m} \left[\frac{\hbar^2 q^4}{4m^2} + 3 \frac{q^2}{m} \langle E_{\text{kin}} \rangle + C(\mathbf{q}) \right], \quad (17)$$

$$C(\mathbf{q}) = \frac{1}{2} \omega_D^2 \int_0^\infty d\bar{r} \bar{r} g(r) K(\mathbf{r}, \mathbf{q}), \quad (18a)$$

$$\begin{aligned} K(\bar{r}, \mathbf{q}) = & -\frac{a^2}{\pi} \int_0^{2\pi} d\phi [\exp(i\mathbf{q} \cdot \mathbf{r}) - 1] \frac{(\mathbf{q} \cdot \nabla)^2}{q^2} \frac{1}{\bar{r}^3} \\ = & \frac{3}{\bar{r}^5} [3 - 3J_0(\bar{q}\bar{r}) + 5J_2(\bar{q}\bar{r})], \end{aligned} \quad (18b)$$

where $\bar{r} = r/a$, $\bar{q} = qa$, $J_0(qr)$ and $J_2(qr)$ are ordinary Bessel functions, $\omega_D = \sqrt{2\pi n\mu^2/ma^3}$ is a nominal 2D dipole frequency, $a = 1/\sqrt{\pi n}$ is the 2D Wigner-Seitz radius, and $g(r)$ is the statistics- and coupling-dependent pair distribution function. (To avoid confusion about the mass notation used for the EHB in the next section, this is a good place to remind the reader that in this section, $m = m_e + m_h$.) Equations (17) and (18) are valid for arbitrary degeneracy. The second right-hand-side member of sum rule (17), proportional to the average kinetic energy per particle for the interacting 2D system $\langle E_{\text{kin}} \rangle = \langle p^2/2m \rangle$, is a well-known feature of third-frequency-moment sum rules (note that $\langle E_{\text{kin}} \rangle = 1/\beta$ when the particles obey classical statistics). At long wavelengths ($\bar{q} < 1$) and in high-coupling regimes, however, the dominant contribution by far is the third right-hand-side member of (17) portraying the dipole-dipole interactions (18a) and (18b).

To see that the dipole $g(r \rightarrow 0)$ tends to zero sufficiently fast to guarantee convergence of the $C(\mathbf{q})$ integral (18) in the zero-temperature quantum domain, we observe that when two point dipoles are in close proximity to each other, the ground-state pair wave function $\psi(r)$ and consequently the pair distribution function $g(r) \propto |\psi(r)|^2$ are determined by the solution of the two-particle Schrödinger equation in the $r \rightarrow 0$ limit. Paralleling Kimball's electron-gas calculation [30], one readily finds that the dipole $g(r \rightarrow 0) \propto K_0^2(2\sqrt{r_0/r}) \approx (\pi/4)\sqrt{r/r_0} \exp(-4\sqrt{r_0/r})$, where K_0 is the modified Bessel function of the second kind and $r_0 = m\mu^2/\hbar^2$ is the dipole equivalent of the Bohr radius [20(a)]. This small- r behavior has also been pointed out in Ref. [20(a)]. In the high-temperature classical domain, convergence is guaranteed as long as $g(r \rightarrow 0) \propto \exp[-\beta\mu^2/r^3]$, as expected. The structure of $C(\mathbf{q})$ for arbitrary coupling, and consequently of the third-frequency-moment sum rule itself, implies that the 2DDS does not have a random-phase-approximation (RPA) limit, i.e., $g(r) = 1$ is ruled out. This crucial point has already been emphasized in Refs. [22,25]. The long-wavelength limit of sum rule (17) can be expressed in terms of the average dipole-dipole interaction energy per particle

$$\langle E_{\text{int}} \rangle = \frac{n}{2} \int d^2r \varphi_D(r) g(r) \quad (19)$$

as

$$\langle \omega^3 \rangle(q \rightarrow 0) = -\frac{nq^2}{m} \left[3 \frac{q^2}{m} \langle E_{\text{kin}} \rangle + \frac{33}{8} \frac{q^2}{m} \langle E_{\text{int}} \rangle \right]. \quad (20)$$

It is instructive to compare the dipole sum rules (17) and (20) with their 2DOCP counterparts [10(b)]:

$$\langle \omega^3 \rangle(\mathbf{q}) = -\frac{nq^2}{m_e} \left[\frac{\hbar^2 q^4}{4m_e^2} + 3 \frac{q^2}{m_e} \langle E_{\text{kin}} \rangle + C_{\text{Coul}}(\mathbf{q}) \right], \quad (21)$$

$$C_{\text{Coul}}(\mathbf{q}) = \frac{1}{2} \omega_0^2 \int_0^\infty d\bar{r} \bar{r} g(\bar{r}) K_{\text{Coul}}(\bar{r}, \mathbf{q}), \quad (22a)$$

$$\begin{aligned} K_{\text{Coul}}(\bar{r}, \mathbf{q}) &= -\frac{a^2}{\pi} \int_0^{2\pi} d\phi [\exp(i\mathbf{q} \cdot \mathbf{r}) - 1] \frac{(\mathbf{q} \cdot \nabla)^2}{q^2} \frac{1}{\bar{r}} \\ &= \frac{1}{\bar{r}^3} [1 - J_0(\bar{q}\bar{r}) + 3J_2(\bar{q}\bar{r})], \end{aligned} \quad (22b)$$

where $\omega_0 = \sqrt{2\pi ne^2/m_e a}$ is a nominal 2D plasma frequency. The appealing structural likeness of (17) and (21) notwithstanding, we nevertheless note the marked difference between the potential-dependent kernel functions (18b) and (22b). In attempting to generate a small- q expansion from (22a) and (22b), one encounters a divergent integral. This mandates writing (22a) in the form where the contribution to the integral that exhibits the large- r divergence for $q \rightarrow 0$ can be separated:

$$C_{\text{Coul}}(\mathbf{q}) = \frac{1}{2} \omega_0^2 \int_0^\infty d\bar{r} \bar{r} [1 + h(\bar{r})] K_{\text{Coul}}(\bar{r}, \mathbf{q}). \quad (23)$$

Here $h(\bar{r})$ is the equilibrium pair correlation function. It is then the 1 contribution to the integral that can be identified as the offending term. However, this integral can be exactly evaluated for arbitrary \bar{q} :

$$\int_0^\infty \frac{d\bar{r}}{\bar{r}^2} [1 - J_0(\bar{q}\bar{r}) + 3J_2(\bar{q}\bar{r})] = 2\bar{q}. \quad (24)$$

The correct procedure is then to substitute Eq. (23) with (24) into sum rule (21). This results in

$$\langle \omega^3 \rangle(\mathbf{q}) = -\frac{nq^2}{m_e} \left[\frac{\hbar^2 q^4}{4m_e^2} + 3 \frac{q^2}{m_e} \langle E_{\text{kin}} \rangle + \omega_0^2 \bar{q} + D(\mathbf{q}) \right], \quad (25a)$$

$$D(\mathbf{q}) = \frac{1}{2} \omega_0^2 \int_0^\infty d\bar{r} \bar{r} h(\bar{r}) K_{\text{Coul}}(\bar{r}, \mathbf{q}). \quad (25b)$$

This procedure is tantamount to separating out the $\omega_0\sqrt{\bar{q}}$ RPA plasmon frequency and $D(\mathbf{q})$ non-RPA exchange-correlation contributions. At long wavelengths,

$$\langle \omega^3 \rangle(q \rightarrow 0) = -\frac{nq^2}{m_e} \left[3 \frac{q^2}{m_e} \langle E_{\text{kin}} \rangle + \omega_0^2 \bar{q} + \frac{5q^2}{8m_e} \langle E_{\text{Coul}} \rangle \right], \quad (26a)$$

where

$$\langle E_{\text{Coul}} \rangle = \frac{n}{2} \int d^2r \varphi_{\text{Coul}}(r) h(r) \quad (26b)$$

is the average Coulomb potential energy, with $\varphi_{\text{Coul}}(r) = e^2/r$. Comparing now the behavior of the 2DDS governed by the $1/r^3$ short-range interaction and the 2D Coulomb layer governed by the $1/r$ long-range interaction, we see why the separation of the leading RPA plasmon frequency from the exchange-correlation contribution is necessary. Such a separation does not exist for the dipole short-range interactions.

III. ELECTRON-HOLE BILAYER LIQUID

In this section we establish the third-frequency-moment sum rules for the closely spaced EHB liquid, with the goal of showing how the in-phase sum rule relates to its 2DDS counterpart (17). Here it suffices to confine our study to the mathematically more tractable *mass-symmetric* EHB ($m_e = m_h = \bar{m}$) [17,18,27,28]. This idealized model consists of two large but bounded infinitely thin layers, each of area A , spaced a distance d apart; layer 2 is populated by nA electrons, each endowed with charge $-|e|$; layer 1 is populated by nA holes, each endowed with charge $+|e|$. The interaction potentials are

given as

$$\begin{aligned}\varphi_{11}(r) &= \varphi_{22}(r) = \frac{e^2}{r}, \quad \varphi_{12}(r) = -\frac{e^2}{\sqrt{r^2 + d^2}}; \\ \varphi_{11}(q) &= \varphi_{22}(q) = \frac{2\pi e^2}{q}, \quad \varphi_{12}(q) = -\frac{2\pi e^2}{q} \exp(-qd).\end{aligned}\quad (27)$$

As we have emphasized in the Introduction, the 2DDS model is expected to be a good description for the closely spaced EHB liquid in the $d \rightarrow 0$ limit. For finite- d values, the situation is more complicated. Although both classical [18] and quantum [17] simulations seem to establish a critical coupling-dependent d^* value below which the system is in the dipole state, the physical meaning of d^* is unclear. The exact phase diagram (in the $d - \Gamma$ or $d - r_s$ parameter space) of the EHB is not known; it is not even established whether the transition from the Coulomb liquid state to the dipole liquid state is a phase transition or a gradual crossover. Should the latter be the case, one would have to deal with a three-component mixture of concentrations determined by equilibrium requirements. In the following, we will work by assuming that we can choose a domain $d \ll d^*$ such that the system is entirely (i.e., overwhelmingly) in the dipole state, while for the domain $d \gg d^*$ it is entirely in the (spin-unpolarized) Coulomb state.

The derivation of the sum rules is to be carried out in two stages. In the first stage, we infer from an earlier work [15] the in-phase and out-of-phase third-frequency-moment sum rules for the EHB in its Coulomb liquid phase. This phase now is characterized by an interlayer spacing $d \gg d^*$. In the second stage, we concentrate on the $d \ll d^*$ domain characterizing the excitonic liquid phase. We adapt the first-stage EHB sum rules to this domain by carrying out what amounts to an expansion in powers of d/a after we have modeled the interlayer pair distribution function $g_{12}(r, d)$ to take account of its evolution as $d \rightarrow 0$ [17,18,31].

Proceeding with the first-stage formulation, one customarily defines the (external) density response matrix $\chi_{AB}(\mathbf{q}, \omega)$ through the constitutive relation [32]

$$\begin{aligned}\langle n_A(\mathbf{q}, \omega) \rangle &= \sum_B \chi_{AB}(\mathbf{q}, \omega) \hat{U}^B(\mathbf{q}, \omega) \\ &= \sum_B \chi_{AB}(\mathbf{q}, \omega) e_B \hat{\Phi}^B(\mathbf{q}, \omega)\end{aligned}\quad (28)$$

linking the Fourier transform of the average density response $\langle n_A(\mathbf{r}, t) \rangle$ of layer A field particles to an external potential energy perturbation $\hat{U}^B(\mathbf{r}, t)$; its companion external scalar potential $\hat{\Phi}^B(\mathbf{r}, t)$, which acts on the layer B field particles [each endowed with charge label e_B ; $\hat{U}^B(\mathbf{r}, t) = e_B \hat{\Phi}^B(\mathbf{r}, t)$], can originate from an external charge source Q placed in either layer. Thus, if the charge density $Q(t)$ is placed in layer 1 at $\mathbf{r} = \mathbf{0}$, then

$$\begin{aligned}\hat{\Phi}^1(\mathbf{q}, \omega) &= \frac{2\pi Q(\omega)}{q}, \quad \hat{U}^1(\mathbf{q}, \omega) = +|e|\hat{\Phi}^1(\mathbf{q}, \omega); \\ \hat{\Phi}^2(\mathbf{q}, \omega) &= \frac{2\pi Q(\omega)}{q} \exp(-qd), \\ \hat{U}^2(\mathbf{q}, \omega) &= -|e|\hat{\Phi}^2(\mathbf{q}, \omega).\end{aligned}\quad (29)$$

The first- and third-frequency-moment sum-rule coefficients, defined by

$$\begin{aligned}\langle \omega \rangle_{AB}(\mathbf{q}) &= \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \omega \text{Im} \chi_{AB}(\mathbf{q}, \omega) \\ &= \frac{1}{i\hbar A} \langle [\dot{n}_{A,\mathbf{q}}, n_{B,-\mathbf{q}}] \rangle,\end{aligned}\quad (30a)$$

$$\begin{aligned}\langle \omega^3 \rangle_{AB}(\mathbf{q}) &= \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \omega^3 \text{Im} \chi_{AB}(\mathbf{q}, \omega) \\ &= \frac{1}{i\hbar A} \langle [\ddot{n}_{A,\mathbf{q}}, \dot{n}_{B,-\mathbf{q}}] \rangle,\end{aligned}\quad (30b)$$

can be readily formulated by adapting electron multilayer equations (8) and (9) of Ref. [15] to the EHB characterized by interaction potentials (27). One obtains the sum-rule coefficients

$$\langle \omega \rangle_{AB}(\mathbf{q}) = -\frac{nq^2}{\bar{m}} \delta_{AB}, \quad (31)$$

$$\begin{aligned}\langle \omega^3 \rangle_{AB}(\mathbf{q}) &= -\frac{nq^2}{\bar{m}} \left\{ \left[\frac{\hbar q^2}{2\bar{m}} \right]^2 \delta_{AB} + 3 \frac{q^2}{\bar{m}} \langle E_{\text{kin}} \rangle \delta_{AB} \right. \\ &\quad \left. + \frac{nq^2}{\bar{m}} \varphi_{AB}(q) + D_{AB}(\mathbf{q}) \right\},\end{aligned}\quad (32a)$$

where

$$\begin{aligned}D_{AB}(\mathbf{q}) &= -\frac{n}{\bar{m}} \int d^2\mathbf{r} h_{AB}(r) [\exp(-i\mathbf{q} \cdot \mathbf{r}) - \delta_{AB}] \\ &\quad \times \frac{(\mathbf{q} \cdot \nabla)^2}{q^2} \varphi_{AB}(r) + \frac{n}{\bar{m}} \delta_{AB} \\ &\quad \times \sum_{C \neq A} \int d^2\mathbf{r} h_{AC}(r) \frac{(\mathbf{q} \cdot \nabla)^2}{q^2} \varphi_{AC}(r)\end{aligned}\quad (32b)$$

is the longitudinal exchange-correlation matrix. Its elements can be expressed in terms of the computationally tractable configuration space integrals:

$$\begin{aligned}D_{11}(\mathbf{q}) &= \frac{\omega_0^2}{2} \left\{ \int_0^\infty d\bar{r} \frac{1}{\bar{r}^2} h_{11}(r) [1 - J_0(qr) + 3J_2(qr)] \right. \\ &\quad \left. - \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} h_{12}(\rho) \left[1 - 3\frac{d^2}{\rho^2} \right] \right\},\end{aligned}\quad (33)$$

$$\begin{aligned}D_{12}(\mathbf{q}) &= \frac{\omega_0^2}{2} \left\{ \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} h_{12}(\rho) \left[1 - 3\frac{d^2}{\rho^2} \right] \right. \\ &\quad \left. - \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} h_{12}(\rho) [1 - J_0(qr) + 3J_2(qr)] \right\} \\ &\quad + \frac{3\bar{d}^2 \omega_0^2}{2} \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^5} h_{12}(\rho) [1 - J_0(qr) + J_2(qr)],\end{aligned}\quad (34)$$

$$D_{22}(\mathbf{q}) = D_{11}(\mathbf{q}), \quad (35)$$

where $\rho = \sqrt{r^2 + d^2}$, $\bar{\rho} = \rho/a$, $\bar{r} = r/a$, and $\omega_0^2 = 2\pi n e^2 / \bar{m} a$. We note the recovery of the exchange-correlation integral (25b) in the $d \rightarrow \infty$ isolated 2D layer limit. Equations (32a) and (32b) can be rewritten as in-phase (+) and

out-of phase (−) sum rules:

$$\langle \omega^3 \rangle_{\pm}(\mathbf{q}) = -\frac{nq^2}{\bar{m}} \left\{ \left[\frac{\hbar q^2}{2\bar{m}} \right]^2 + 3\frac{q^2}{\bar{m}} \langle E_{\text{kin}} \rangle + \omega_0^2 \bar{q} [1 \mp \exp(-qd)] + D_{11}(\mathbf{q}) \pm D_{12}(\mathbf{q}) \right\}. \quad (36)$$

As expected, the Coulomb liquid phase sum rules (32) and (36) exhibit the separate RPA plasmon frequency and exchange-correlation matrix $D_{AB}(\mathbf{q})$ contributions, with the latter expressed in terms of the equilibrium pair correlation function $h_{AB}(r)$. As we have pointed out in the previous section, this is in sharp contrast to the 2DDS sum rule (17), which of course can have no RPA contribution and features not $h(r)$ in the dipole-dipole interaction integral, but rather the pair distribution function $g(r) = 1 + h(r)$. We proceed now to reformulate the sum rules in the $d \rightarrow 0$ limit following a rather intricate procedure detailed in the stage-two derivation below.

Taking our cue from the discussion in the paragraph below Eq. (36), one can mask the RPA contribution to (32a) by introducing the C_{AB} dynamical matrix elements:

$$C_{11}(\mathbf{q}) = \omega_0^2 \bar{q} + D_{11}(\mathbf{q}), \quad (37)$$

$$C_{12}(\mathbf{q}) = -\omega_0^2 \bar{q} e^{-qd} + D_{12}(\mathbf{q}). \quad (38)$$

The third-frequency-moment sum rules (36) now assume the more compact form

$$\langle \omega^3 \rangle_{\pm}(\mathbf{q}) = -\frac{nq^2}{\bar{m}} \left\{ \left[\frac{\hbar q^2}{2\bar{m}} \right]^2 + 3\frac{q^2}{\bar{m}} \langle E_{\text{kin}} \rangle + C_{11}(\mathbf{q}) \pm C_{12}(\mathbf{q}) \right\}. \quad (39)$$

Then, from Eqs. (33), (34), (37), and (38) and the integral formulas

$$\int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} \left[1 - 3\frac{d^2}{\bar{\rho}^2} \right] = 0, \quad (40)$$

$$\frac{1}{2} \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} [1 - J_0(qr) + 3J_2(qr)] - \frac{3}{2} \bar{d}^2 \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^5} [1 - J_0(qr) + J_2(qr)] = \bar{q} e^{-qd}, \quad (41)$$

one readily obtains the $C_{AB}(\mathbf{q})$ matrix elements by trading $h(r)$ for $g(r)$ in (33) and (34). The stage is now properly set for the derivation of the $C_{AB}(\mathbf{q})$ matrix elements and subsequent (\pm) third-frequency-moment sum-rule coefficients in the $d \ll d^*$ domain of bound electron-hole pairs

First, we observe that in this $d \ll d^*$ domain, classical simulations [18] indicate the existence of permanently bound electron-hole pairs through the correlated motion of the two terminal particles. Based on this observation, one expects that it is the combined mass $m = 2\bar{m}$ that should be featured in the (+) f sum rule and in the prefactor and kinetic energy contributions to the (+) third-frequency-moment sum rule (39). Replacement of \bar{m} with m in (39) goes hand-in-hand with the $g_{12}(\rho)$ conjecture (42) below. We will see that this replacement is consonant with the natural emergence of m , not \bar{m} , in the $C_+(\mathbf{q})$ dipole-dipole interaction contribution to

the closely spaced EHB sum-rule counterpart of Eq. (17). It should be noted, however, that this replacement of \bar{m} with m is a reasonable correction to the sum-rule derivation with a preset goal in mind, but by no means is on the same exact footing as the establishment of the parent equation (39). As mentioned before, a rigorous derivation of the result would require recognizing that, in the $d \ll d^*$ domain, we are no longer dealing with a two-layer architecture but rather with a ternary mixture of bound electron-hole pairs, along with free electrons and free holes; the derivation would then be reformulated to accommodate this latter architecture.

Next we need to take account of the rather dramatic evolution of $g_{12}(\rho)$ as $d \rightarrow 0$. Zero-temperature quantum and classical MC simulations of the symmetric EHB [17,18,31] reveal that in the excitonic liquid phase, $g_{12}(\rho)$ for both the spin-unpolarized and classical Coulomb liquids develops a steep Gaussian-like central peak $\Delta(\bar{\mathbf{r}}, d)$ for low- \bar{r} values around $\bar{r} = 0$. For quantum systems this is understandable because it follows from the assumption that $g_{12}(r \rightarrow 0) \simeq |\psi_{12}(r \rightarrow 0)\psi_{12}^*(r \rightarrow 0)|$, where $\psi_{12}(r)$ is the ground-state wave function for the 12 pair. For the classical system, the reason for the deviation from the expected (on thermodynamic grounds) $\exp[-\beta\varphi_{12}(\rho)]$ is discussed in Ref. [18]. Moreover, classical MC and MD simulations [18,27] indicate that, sufficiently well below the Coulomb liquid–dipole liquid phase boundary, $g_{12}(\bar{\rho}) \approx g_{11}(\bar{r}) + \Delta(\bar{\mathbf{r}}, d)$, with the Gaussian-like steepening markedly increasing with decreasing d . This suggests that, in the $d \rightarrow 0$ limit, it is reasonable to conjecture that

$$g_{12}(\bar{\rho}) = \tilde{g}_{11}(\bar{r}) + \frac{1}{n} \delta(\mathbf{r}) = \tilde{g}_{11}(\bar{r}) + \pi \delta(\bar{\mathbf{r}}), \quad (42)$$

where $\tilde{g}_{11}(\bar{r})$ is practically the same as $g_{11}(\bar{r})$; indeed, since the Gaussian-like central peak of $g_{12}(\rho)$ contains but one particle out of N particles [18], we can take $\tilde{g}_{11}(\bar{r}) = g_{11}(\bar{r})$, accurate to order $1/N$. Thus the $C_{AB}(\mathbf{q})$ matrix elements become

$$C_{11}(\mathbf{q}) = \frac{\omega_0^2}{2} \left\{ \int_0^\infty d\bar{r} \frac{1}{\bar{r}^2} g_{11}(r) [1 - J_0(qr) + 3J_2(qr)] - \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} g_{11}(r) \left[1 - 3\frac{d^2}{\bar{\rho}^2} \right] + \frac{1}{\bar{d}^3} \right\}, \quad (43)$$

$$C_{12}(\mathbf{q}) = \frac{\omega_0^2}{2} \left\{ -\frac{1}{\bar{d}^3} + \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} g_{11}(r) \left[1 - 3\frac{d^2}{\bar{\rho}^2} \right] - \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} g_{11}(r) [1 - J_0(qr) + 3J_2(qr)] \right\} + \frac{3\bar{d}^2 \omega_0^2}{2} \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^5} g_{11}(r) [1 - J_0(qr) + J_2(qr)]. \quad (44)$$

Then applying the denominator expansion

$$\frac{1}{\bar{\rho}^3} \approx \frac{1}{\bar{r}^3} \left[1 - \frac{3}{2} \frac{\bar{d}^2}{\bar{r}^2} \right]$$

to Eqs. (43) and (44) provides

$$C_{11}(\mathbf{q}) = -\frac{\omega_0^2}{2} \int_0^\infty \frac{d\bar{r}}{\bar{r}^2} g_{11}(r) [J_0(qr) - 3J_2(qr)] + \frac{9}{4} \omega_0^2 \bar{d}^2 \int_0^\infty \frac{d\bar{r}}{\bar{r}^4} g_{11}(r) + \frac{e^2}{\bar{m} \bar{d}^3}, \quad (45)$$

$$C_{12}(\mathbf{q}) = \frac{\omega_0^2}{2} \int_0^\infty \frac{d\bar{r}}{\bar{r}^2} g_{11}(r) [J_0(qr) - 3J_2(qr)] - \frac{3}{4} \omega_0^2 \bar{d}^2 \times \int_0^\infty \frac{d\bar{r}}{\bar{r}^4} g_{11}(r) [3J_0(qr) - 5J_2(qr)] - \frac{e^2}{\bar{m}d^3}. \quad (46)$$

Convergence of the second right-hand-side integrals of (45) and (46) is guaranteed in virtue of the discussion above Eq. (19). The (+) third-frequency-moment sum rule for the EHB in the $d \ll d^*$ domain now readily follow from Eqs. (39), (45), and (46), with the stipulation that the combined dipole mass m replaces \bar{m} in the prefactor and kinetic energy contributions per the discussion in the paragraph following Eq. (27)

$$\langle \omega^3 \rangle_+(\mathbf{q}) = -\frac{nq^2}{m} \left\{ \left[\frac{\hbar q^2}{2m} \right]^2 + 3 \frac{q^2}{m} \langle E_{\text{kin}} \rangle + C_+(\mathbf{q}) \right\}, \quad (47)$$

$$C_+(\mathbf{q}) = \frac{1}{2} \omega_D^2 \int_0^\infty d\bar{r} \bar{r} g_{11}(r) K(\bar{r}, \mathbf{q}), \quad (48)$$

where $K(\bar{r}, \mathbf{q})$ is defined by Eq. (18b). Here we note that $\omega_D^2 = 2\pi n \mu^2 / (2\bar{m}) a^3$ is identical to the 2DDS characteristic frequency defined below Eq. (17) for $m_e = m_h$. At long wavelengths, (47a) simplifies to

$$\langle \omega^3 \rangle_+(q \rightarrow 0) = -\frac{nq^2}{m} \left[3 \frac{q^2}{m} \langle E_{\text{kin}} \rangle + \frac{33}{8} \frac{q^2}{m} \langle E_{\text{int}} \rangle \right], \quad (49)$$

where

$$\langle E_{\text{int}} \rangle = (n/2) \int d^2 r \varphi_D(r) g_{11}(r).$$

Clearly, the $C(\mathbf{q})$ and $C_+(\mathbf{q})$ dipole-dipole interaction terms in sum rules (17) and (47) reconcile, as do the sum rules (17) and (47) subject to the mass replacement proviso.

In this section we have followed an approach that, in effect, shows how the (+) sum rule (39) for the Coulomb liquid evolves with decreasing d into the (+) sum rule (47) in the $d \ll d^*$ bound-state domain. This replication of the 2DDS sum rule (17) notwithstanding, the approach has its limitations when it comes to formulating the (−) sum-rule companion to (47). For one thing, there is the question of which mass to assign to the prefactor and kinetic energy contributions to the third-frequency-moment sum rule [and to the (−) f sum rule as well]. While we do not yet have clear guidelines for selecting the appropriate mass, the assumption that we are dealing with a bound state suggests the reduced mass $\tilde{m} = \bar{m}/2$ to be the appropriate choice; this is further corroborated by Eq. (53) below and by the natural emergence of \tilde{m} in Eq. (52) for $C_-(q = 0)$.

We proceed now with the calculation of $C_-(\mathbf{q}) = C_{11}(\mathbf{q}) - C_{12}(\mathbf{q})$ from Eqs. (45) and (46). One readily obtains

$$C_-(\mathbf{q}) = \omega_K^2 - \omega_0^2 \int_0^\infty \frac{d\bar{r}}{\bar{r}^2} g_{11}(r) [J_0(\bar{q}\bar{r}) - 3J_2(\bar{q}\bar{r})] + \frac{3}{2} \omega_D^2 \int_0^\infty \frac{d\bar{r}}{\bar{r}^4} g_{11}(r) [3 + 3J_0(\bar{q}\bar{r}) - 5J_2(\bar{q}\bar{r})], \quad (50)$$

where $\omega_K = \sqrt{e^2/\tilde{m}d^3}$ is the Kepler frequency, apparently representing the intrinsic excitation of the bound electron-hole

pair. We gain further insights into the expected $\omega(\mathbf{q})$ dispersion that combines effects originating from the intrinsic excitations of the dipole and from the genuine out-of-phase propagating mode by evaluating the sum-rule ratio $\langle \omega^3 \rangle_-(\mathbf{q})/\langle \omega \rangle_-(\mathbf{q})$:

$$\frac{\langle \omega^3 \rangle_-(\mathbf{q})}{\langle \omega \rangle_-(\mathbf{q})} = \left[\frac{\hbar q^2}{2\tilde{m}} \right]^2 + 3 \frac{q^2}{\tilde{m}} \langle E_{\text{kin}} \rangle + C_-(\mathbf{q}); \quad (51)$$

the reduced mass replacements in the kinetic energy contributions go hand-in-hand with the natural emergence of the reduced mass in $C_-(\mathbf{q})$. From Eqs. (50) and (51)

$$\left. \frac{\langle \omega^3 \rangle_-(\mathbf{q})}{\langle \omega \rangle_-(\mathbf{q})} \right|_{q=0} = C_-(0) = \omega_K^2 - \frac{1}{\tilde{m}d^2} \langle E_{\text{int}} \rangle + 9\omega_D^2 \int_0^\infty \frac{d\bar{r}}{\bar{r}^4} g_{11}(r). \quad (52)$$

Equation (52) also follows directly from the application of the $g_{12}(\rho)$ conjecture (42) to

$$C_-(0) = \frac{n}{\tilde{m}} \int d^2 r g_{12}(r) \frac{(\mathbf{q} \cdot \nabla)^2}{q^2} \varphi_{12}(r) = -\omega_0^2 \int_0^\infty \frac{d\bar{r}}{\bar{r}^3} g_{12}(\rho) \left[1 - 3 \frac{d^2}{\rho^2} \right] > 0. \quad (53)$$

Clearly, the statistics-independent Kepler frequency plays the dominant role in (52). It also plays the dominant role in the description of the collective in-layer shear mode oscillations of two indirect excitons in a harmonic trap [33].

IV. CONCLUSION

We have derived and analyzed the third-frequency-moment sum rules for the mass-symmetric electron-hole bilayer and for the two-dimensional (point) dipole system characterized by the repulsive interaction potential $\varphi_D(r) = \mu^2/r^3$, where μ is the electric dipole moment. Our principal results are given by Eqs. (17), (20), (36), (47), (49), (51), and (52).

In the strong-coupling regimes of interest in the present study, we have supposed that Bose-Einstein condensation can be ignored since strong particle interactions destroy coherence, thereby suppressing the condensate fraction. This is borne out by quantum MC [17,20] and path-integral Monte Carlo simulations [34].

Our analysis shows the extent to which the 2DDS sum rule (17) may or may not reconcile with its companion EHB (+) sum rule (47). Our overall observation reinforces the justification of using the 2DDS as an approximation for the closely spaced EHB. The (−) EHB sum rule ratios (51) and (52), which relate to the excitations of the internal degrees of freedom of the bound electron-hole pairs, merit further exploration.

In order to find the connection between the sum-rule relationships and the mode structure, one may generate the $\langle \omega^3 \rangle_+(\mathbf{q})/\langle \omega \rangle_+(\mathbf{q})$ ratios, as suggested by Refs. [10,12], from the 2DDS sum rule (17) or its companion EHB in-phase (+) sum rule (47a). Calculated in the high-coupling regimes and at long wavelengths, they are dominated by the $C(\mathbf{q})$ term. This quantity can then be identified as the centerpiece of the quasilocized-charge-approximation (QLCA) description

of the collective excitations for the respective systems. As to the ratio $\langle\omega^3\rangle_{-}(\mathbf{q})/\langle\omega\rangle_{-}(\mathbf{q})$ of the out-of-phase sum-rule coefficients, we observe that the finite-frequency gap

$$\left.\frac{\langle\omega^3\rangle_{-}(\mathbf{q})}{\langle\omega\rangle_{-}(\mathbf{q})}\right|_{q=0} = -\omega_0^2 \int_0^\infty \frac{d\bar{r}\bar{r}}{\bar{\rho}^3} h_{12}(\rho) \left[1 - 3\frac{d^2}{\rho^2}\right] > 0, \quad (54)$$

the hallmark of the out-of-phase collective mode dispersion in strongly coupled EHB Coulomb liquids [27,28] is the survivor in the $q \rightarrow 0$ limit. In the $d \rightarrow 0$ limit, the resulting equation (52) shows the dominance of the Kepler frequency that can be identified with the internal excitation of the bound electron-hole pair. This is also consistent with the prediction

of our recent QLCA studies of the classical bilayer [27,28]: The Kepler frequency is also shown there to be the dominant feature of the long-wavelength energy gap in the out-of-phase collective excitation spectrum. However, the additional terms appearing in Eq. (52) have not been explored in the analysis of the EHB mode dispersion; their presence here indicates how at finite- d values, a correlation-induced energy gap would interfere with the Kepler frequency.

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