Collective Modes in Classical Mass-Asymmetric Bilayers

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Using the quasi-localized charge approximation (QLCA), we analyze the effects of asymmetry on the long-wavelength longitudinal collective mode dispersion in a variety of strongly correlated electronic bilayer liquids, most notably, the mass-asymmetric electron-hole bilayer in its Coulomb liquid and dipole liquid phases. We point out the marked differences between the strong coupling (QLCA) and weak coupling (random-phase-approximation) descriptions of the way the asymmetry affects the collective mode structure.

In order to better understand the collective mode behavior of Coulomb bilayers actually realized in laboratory quantum well structures, there is a need to take into account the mass asymmetry. For example, in electron-hole bilayers (EHBs), one can hardly ignore the marked disparity between the electron and hole masses. However, how this asymmetry affects the mode structure depends on the value of the coupling parameter r_s. Here we study the strong coupling regime via a classical model. We calculate the longitudinal sound speeds and long-wavelength finite-frequency (energy) gaps for closely spaced mass-asymmetric EHB and electron bilayer (EBL) systems with the aid of the quasi-localized charge approximation (QLCA). The present study generalizes earlier collective mode studies for symmetric EBLs [1–3] and EHBs [4–6], the latter both in its Coulomb liquid and dipole liquid phases [7, 8].

It is instructive to first display the collective mode frequencies for bilayers in the Coulomb liquid phase with $n_1 \neq n_2$, $Z_1 \neq Z_2$, and $m_1 \neq m_2$. We introduce the parameters

$$p^2 = \frac{Z_2 n_2}{Z_1 n_1}, \qquad q^2 = \frac{Z_2 m_1}{Z_1 m_2} \tag{1}$$

and the nominal 2D reference frequency:

$$\omega_1^2 = \frac{2\pi n_1 Z_1^2 e^2}{a m_1}, \qquad \pi a^2 \sqrt{n_1 n_2} = 1. \tag{2}$$

The longitudinal collective modes frequencies are calculated from the dispersion relation

$$||\omega^2 \delta_{AB} - C_L^{AB}(k)|| = 0; \tag{3}$$

k is the in-plane wave number. Introducing the convenient dimensionless notation $\bar{k} = ka$ and $\bar{r} = r/a$, the long-wavelength QLCA dynamical matrix elements are given as

$$C_L^{11}(k \to 0) = \omega_1^2 \left[p^2 W + \bar{k} + U_{11}\bar{k}^2\right], \tag{4}$$

$$C_L^{12}(k \to 0) = \omega_1^2 pq \left[-W + \bar{k} - \bar{d}\,\bar{k}^2 + U_{12}\bar{k}^2\right], \tag{5}$$

$$C_L^{22}(k \to 0) = \omega_1^2 q^2 \left[W + p^2\bar{k} + p^2 U_{22}\bar{k}^2\right]; \tag{6}$$

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where

$$W=\frac{1}{2}\int_0^\infty \frac{\mathrm{d}\bar{r}\,\bar{r}}{\left(\bar{r}^2+\bar{d}^2\right)^{3/2}}g_{12}(r)\left[1-3\frac{\bar{d}^2}{\bar{r}^2+\bar{d}^2}\right] \tag{7}$$

is the $k=0$ interlayer correlation-generated finite-frequency contribution and

$$U_{11} = \frac{5}{16}\int_0^\infty \mathrm{d}\bar{r}h_{11}(r), \qquad U_{22}=\frac{5}{16}\int_0^\infty \mathrm{d}\bar{r}h_{22}(r), \tag{8}$$

$$U_{12} = \frac{5}{16}\int_0^\infty \frac{\mathrm{d}\bar{r}\,\bar{r}^3}{\left(\bar{r}^2+\bar{d}^2\right)^{3/2}}h_{12}(r)-\frac{9}{16}\bar{d}^2\int_0^\infty \frac{\mathrm{d}\bar{r}\,\bar{r}^3}{\left(\bar{r}^2+\bar{d}^2\right)^{5/2}}h_{12}(r) \tag{9}$$

are intra- and interlayer correlation energy contributions to the dispersion; $\bar{d}=d/a$ is the dimensionless layer spacing; $h_{ij}(r)$ is the equilibrium pair correlation function and $g_{ij}(r)=1+h_{ij}(r)$ is the pair distribution function. Eqs. (3) to (9) provide the collective mode frequencies for $n_1\neq n_2$, $Z_1\neq Z_2$, and $m_1\neq m_2$:

$$\begin{aligned}\omega_+^2(k\to 0) &= \frac{q^2\left(1+p^2\right)^2}{p^2+q^2}\omega_1^2\bar{k}-\frac{p^2q^2\left(1-q^2\right)^2\left(1+p^2\right)^2}{\left(p^2+q^2\right)^3 W}\omega_1^2\bar{k}^2\\ &\quad -\frac{2p^2q^2\bar{d}}{p^2+q^2}\omega_1^2\bar{k}^2+\frac{q^2}{p^2+q^2}\left[U_{11}+2p^2U_{12}+p^4U_{22}\right]\omega_1^2\bar{k}^2\end{aligned} \tag{10}$$

$$\begin{aligned}\omega_-^2(k\to 0) &= \left(p^2+q^2\right)\omega_1^2W+\frac{p^2}{p^2+q^2}\left(1-q^2\right)^2\omega_1^2\bar{k}\\ &\quad +\frac{p^2q^2\left(1-q^2\right)^2\left(1+p^2\right)^2}{\left(p^2+q^2\right)^3 W}\omega_1^2\bar{k}^2+\frac{2p^2q^2\bar{d}}{p^2+q^2}\omega_1^2\bar{k}^2\\ &\quad +\frac{p^2}{p^2+q^2}\left[U_{11}-2q^2U_{12}+q^4U_{22}\right]\omega_1^2\bar{k}^2.\end{aligned} \tag{11}$$

Note the emergence of the $k=0$ finite frequency gap in Eq. (11), previously reported for symmetric Coulomb bilayers [2–4]. Focusing on oscillation frequency (10), we observe that to lowest order in k,

$$\omega_+^2(k\to 0)=\tilde{\omega}^2\bar{k}, \tag{12}$$

where

$$\tilde{\omega}^2=\frac{q^2\left(1+p^2\right)^2}{p^2+q^2}\omega_1^2=2\pi n\frac{\langle Z\rangle^2e^2}{a\langle m\rangle} \tag{13}$$

is a characteristic frequency related to the average atom in the virtual crystal approximation [9–11]; $\langle Z\rangle$ and $\langle m\rangle$ are defined to be the average charge and mass:

$$\langle Z\rangle=\frac{Z_1n_1+Z_2n_2}{n_1+n_2}, \qquad \langle m\rangle=\frac{m_1n_1+m_2n_2}{n_1+n_2}. \tag{14}$$

This behavior has also been noted for binary Yukawa systems [12]. Clearly the heavier mass is dominant in (12). The marked contrast between the formally correlation-independent QLCA frequency (12) and its weak coupling (RPA random-phase-approximation) counterpart [13]

$$\omega_{\text{RPA}}^2(k\to 0)=\left(\omega_1^2+\omega_2^2\right)\bar{k}=\left(1+p^2q^2\right)\omega_1^2\bar{k}, \tag{15}$$

(where the lighter mass is dominant) is a consequence of the localization of the particles inherent in the QLCA. For the special case $q^2=1$, the QLCA frequency (12) is identical to the RPA frequency (15). We turn now to the special case of mode dispersion in the EHB in both the Coulomb liquid and dipole liquid phases.

Mass-Asymmetric EHB Coulomb liquid

($n_1 = n_2 = n$, $Z_1 = +1$, $Z_2 = -1$ and $m_1 \neq m_2$)

$$\omega_+^2(k \to 0) = \frac{4\pi ne^2 \bar{d}}{a\,(m_1+m_2)}\bar{k}^2 + \frac{4\pi ne^2}{a\,(m_1+m_2)}\left[U_{11} - U_{12}\right]\bar{k}^2 \tag{16}$$

$$\begin{aligned}\omega_-^2(k \to 0) &= -\frac{2\pi ne^2}{a}\left[\frac{1}{m_1}+\frac{1}{m_2}\right]W + \frac{2\pi ne^2}{a}\left[\frac{1}{m_1}+\frac{1}{m_2}\right]\bar{k} - \frac{4\pi ne^2\bar{d}}{a\,(m_1+m_2)}\bar{k}^2 \\ &\quad + \frac{2\pi ne^2}{am_1m_2\,(m_1+m_2)}\left[\left(m_1^2+m_2^2\right)U_{11} + 2m_1m_2U_{12}\right]\bar{k}^2\end{aligned} \tag{17}$$

For the EHB, note that $W < 0$. In deriving Eq. (16) from (10), we observe that at $p^2 = -1$, the virtual atom frequency (12) disappears leaving an acoustic dispersion comprised of the RPA term (proportional to $\bar{d}$) and a correlational term.

The effect of mass asymmetry in EHBs was first considered some time ago [13, 14]: Using the self-consistent field approximation (SCFA), Eguiluz *et al* [13] noted that the acoustic plasmon is dominated by the heavier hole mass, in accordance with the first right-hand-side member of Eq. (16). Subsequently, Liu *et al* [15], within the framework of a Singwi-Tosi-Land-Sjolander mean-field theory description, conjectured the emergence of a soft mode associated with a charge-density-wave, thought to possibly preempt the onset of excitonic bound states discussed below.

The acoustic frequency (16) indicates the possible occurrence of an instability ($\omega^2 < 0$) for $d < d_c = a(U_{12} - U_{11})$, pertaining to the transition from the Coulomb liquid to dipole liquid phase. To demonstrate that this reasonably well reconciles with the Monte Carlo (MC)-generated phase diagram of Ref. [8], one can estimate the QLCA d_c first by observing that at high layer separations $d/a \geq 2$, there is no appreciable interlayer correlation [8]. Thus, $d_c/a \approx -U_{11} = a|E_{11}|/e^2 = O(1)$, where E_{11} is the in-layer correlation energy per particle. One would expect that such an instability is the precursor to transition from the EHB Coulomb liquid phase to the dipole liquid phase. The above order-of-magnitude estimate of d_c is consonant with the Ref. [8] MC prediction $d_c/a \sim 1 - 1.5$. In the absence of particle correlations ($h_{11}(r) = h_{12}(r) = 0$), we observe that the $k = 0$ finite frequency energy gap $\propto W$ vanishes and Eqs. (16) and (17) morph into the actual RPA $\omega \propto k\sqrt{d}$ acoustic and Eq. (15) $\omega \propto \sqrt{k}$ frequencies for the EHB [4, 13].

Mass-Asymmetric EHB Dipole Liquid

The calculation of the long-wavelength collective mode frequencies for the dipole liquid phase is more intricate. Here, one proceeds by evaluating the finite-k dynamical matrix element

$$C_L^{12}(k) = \omega_1^2 pq\left[\bar{k}\,\exp(-kd) + \tilde{U}_{12}(k) - W\right] \tag{18}$$

$$\begin{aligned}\tilde{U}_{12}(k) &= \frac{1}{2}\int_0^\infty \frac{\mathrm{d}\bar{r}\,\bar{r}}{\left(\bar{r}^2+\bar{d}^2\right)^{3/2}}h_{12}(r)\left[1 - J_0(kr) + 3J_2(kr)\right] \\ &\quad -\frac{3}{2}\bar{d}^2\int_0^\infty \frac{\mathrm{d}\bar{r}\,\bar{r}}{\left(\bar{r}^2+\bar{d}^2\right)^{5/2}}h_{12}(r)\left[1 - J_0(kr) + J_2(kr)\right],\end{aligned} \tag{19}$$

first in the $\bar{d} \to 0$ limit; this entails invoking the approximation $G_{12}(r) = g_{11}(r) + G(r)$, where $G(r)$ represents a steep Gaussian contribution: $g_{12}(0) = G(0) \propto \Gamma/\bar{d}^3$ [8]. The subsequent small-k expansion then follows.

After some algebra, one obtains

$$\omega_+^2(k \to 0, d \to 0) = \frac{33}{16}\bar{k}^2\omega_D^2 \int_0^\infty \frac{\mathrm{d}\bar{r}}{\bar{r}^2} g_{11}(r), \tag{20}$$

$$\begin{aligned}\omega_-^2(k \to 0, d \to 0) = & -\frac{2\pi n e^2}{a}\left[\frac{1}{m_1}+\frac{1}{m_2}\right] W + \frac{2\pi n e^2}{a}\left[\frac{1}{m_1}+\frac{1}{m_2}\right]\bar{k} \\ & + \frac{2\pi n e^2}{a}\left[\frac{1}{m_1}+\frac{1}{m_2}\right] U_{11}\bar{k}^2 - \frac{33}{16}\bar{k}^2\omega_D^2 \int_0^\infty \frac{\mathrm{d}\bar{r}}{\bar{r}^2} g_{11}(r);\end{aligned} \tag{21}$$

$$\omega_D^2 = \frac{2\pi n\mu^2}{a^3\,(m_1+m_2)}$$

is the characteristic dipole frequency [16]; $\mu = ed$. Evaluation of the gap term in (21) proportional to W provides the Kepler frequency, the classical equivalent of the bound-state energy

$$\omega_-^2(k = 0, d \to 0) \approx \frac{e^2}{\pi d^3}\left[\frac{1}{m_1}+\frac{1}{m_2}\right]. \tag{22}$$

We note that the small-k behavior of the plus (+) mode is acoustic with slope proportional to the interlayer spacing d [16, 17]. This is in marked contrast to the $\sqrt{d}$ dependence featured by the RPA contribution to the Eq. (16) acoustic mode [4], indicating that this mode portrays the density oscillation of the dipole. The (–) collective mode spectrum features the $k = 0$ finite frequency energy gap formula [see Ref. 4 for its symmetric EHB counterpart] which, in the dipole liquid phase, is dominated by the prominent Gaussian-like peak in the interlayer pair correlation function at $r = 0$ [4, 8]. Thus, the emergence of the Kepler frequency (22). Here, in contrast to the acoustic speed of the plus mode, it is the lighter particles (the electrons) that play the dominant role. Our calculated sound speed from Eq. (20) for the closely spaced EHB is in perfect agreement with the sound speed formula reported for the 2D dipole liquid with repulsive interaction potential [16, 17]

$$\varphi(r) = \mu^2/r^3 \tag{23}$$

In summary, we have used the quasi-localized charge approximation (QLCA) to describe the longitudinal collective mode dispersion in strongly coupled Coulombic asymmetric bilayer liquids. Two mode frequencies are identified to which we assign $(\pm)$ labels. The (–) mode exhibits the ubiquitous finite-frequency $k = 0$ energy gap first reported by us quite some time ago for the symmetric bilayer liquid; in the asymmetric configuration, the magnitude of the gap is dominated by the lighter species. For the most general configuration where the charges, masses, and densities of the two layers differ, the lowest order (in k) correlation-independent contribution to the (+) mode, while it resembles the weak coupling RPA oscillation frequency in its $\omega \propto \sqrt{k}$ dependence, in fact, differs markedly from the RPA: in the strong coupling regime, the plasma oscillation is dominated by the heavier species, in sharp contrast to the weak coupling regime where it is the lighter species that is dominant. This behavior has also been noted for binary Yukawa systems [12]. For the special case of the mass-asymmetric EHB in its Coulomb liquid phase, the (+) mode is acoustic ($\omega \propto k$) consisting of an RPA contribution softened by combined intralayer and interlayer correlation energy contributions. Our preliminary QLCA analysis indicates that for a given coupling strength, there is a critical layer spacing $d_c = O(a)$, below which $\omega_+^2(k \to 0) < 0$, suggesting a phase transition from the Coulomb liquid to dipole liquid, in accordance with the Monte Carlo-generated phase diagram of Ref. [8]. In the dipole liquid regime, the (–) mode is identified as the Kepler frequency, here dominated by the lighter species; the (+) mode is acoustic with phase velocity governed wholly by the average dipole potential energy in accordance with what has been reported by us for the 2D point dipole system [16].

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