Collective Modes in Strongly Coupled Electronic Bilayer Liquids

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We present the first reliable calculation of the collective mode structure of a strongly coupled electronic bilayer. The calculation is based on a classical model through the 3rd frequency-moment-sum rule preserving quasi-localized-charge approximation, using the recently calculated hypernetted-chain pair correlation functions. The out-of-phase spectrum shows an energy gap at $k = 0$ and the absence of a previously conjectured dynamical instability. [S0031-9007(99)08877-8]

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Electronic bilayers exhibit a rich pattern of behavior, both on the static and on the dynamic level. While at high r_s values ($r_s > r_s^{\text{crystal}}$), the bilayer is expected to crystallize (according to [1,2], $r_s^{\text{crystal}} \geq 20$), and at very low r_s values the random-phase approximation (RPA) description is largely sufficient, the most interesting behavior occurs in the $1 < r_s < r_s^{\text{crystal}}$ domain, where the system is in the liquid state. This is the domain we focus on in this Letter.

The most remarkable feature on the static level is that the system exhibits a series of abrupt structural changes as the ratio of the interlayer distance d and the 2D Wigner-Seitz radius a is varied [3]. These structural changes parallel the structural phase transitions in the solid phase [2,4], but at finite temperature they are also combined with entropy increasing substitutional order-disorder transitions [3] where particles in layer 1(2) occupy positions appropriate for particles in layer 2(1): this is signaled by the two pair correlation functions (PCF) $h_{11}(r)$ and $h_{12}(r)$ becoming identical as $d \to 0$.

The dynamical behavior, the collective mode structure in particular, has been studied by Swierkowski *et al.* [5], by Gold [6], by Zhang and Tzoar [7], by Golden, Kalman, and collaborators [8–10], and by Moudgil *et al.* [11]. (Some of these studies pertain to a superlattice [infinite number of layers], rather than to a bilayer; in qualitative terms the results can, however, be easily interpreted for the bilayer.) Classical bilayer and multilayer structures that form in charged particle traps have also been studied theoretically by Dubin [12(a)] and have recently been observed in ion traps [12(b)]. There are two problematic issues that make the predictions based on the calculations less than reliable [13]. The first issue relates to the approximation technique used: most of the works cited [5–7,11] use methods which violate the 3rd frequency-moment-sum rule, whose satisfaction is well recognized [14] to be an important criterion for providing an acceptable description of the collective mode behavior. The second issue concerns the use of the intralayer and interlayer correlation functions as inputs in all the calculations cited. No reliable PCF data—either for classical or quantum bilayer systems—have been available until fairly recently: thus predictions of the collective mode structure (which turns out to be extremely sensitive to the behavior of the inputted PCF) have been compromised from the outset.

In this Letter we present the first consistent and reliable calculation of the collective mode spectrum of a strongly coupled electronic bilayer liquid. Our results show features which are qualitatively different from the weakly coupled RPA results; they also show that earlier claims concerning the possible emergence of a dynamical instability [5,6,9,11] cannot be supported by a more consistent treatment of the correlations. The calculation is based on a purely classical model: (i) two 2D electron liquids separated by distance d; (ii) scattering on impurities, etc., neglected; (iii) no interlayer tunneling; (iv) the system is described as a binary electron liquid with interaction potentials $\varphi_{11}(r) = \varphi_{22}(r) = e^2/r$, $\varphi_{12}(r) = e^2/\sqrt{(r^2 + d^2)}$, where exchange and other quantum effects are neglected. This approximation is reasonable in the strong coupling domain where the particles are well localized. The intralayer coupling is characterized by the parameter $\Gamma = e^2/aT$ where $a = 1/\sqrt{\pi n}$ and T is the kinetic energy per particle—the temperature in a classical system and $(1/2)\varepsilon_F$ in a zero temperature 2D electron gas. Hence the equivalence $\Gamma \to 2r_s$. The calculation of the dielectric matrix $\varepsilon_{ij}^{\mu\nu}(\mathbf{k}\omega)$ is carried out in the quasi-localized-charge approximation (QLCA), which has been applied successfully for the description of other strongly coupled Coulomb systems [15–17]; in the QLCA $\varepsilon(\mathbf{k}\omega)$ becomes a functional of the intralayer and interlayer PCFs $h_{11}(r)$ and $h_{12}(r)$ or of the corresponding structure functions $S_{11}(k)$ and $S_{12}(k)$,

$$\varepsilon(\mathbf{k}\omega) = \mathbf{I} - \omega_0^2(ka)[\omega^2\mathbf{I} - \mathbf{D}(\mathbf{k})]^{-1}. \qquad (1)$$

[$\omega_0 = (\frac{2\pi e^2 n}{ma})^{1/2}$, the nominal plasma frequency of a single 2D layer] with [15]

$$D_{ij}^{\mu\nu}(\mathbf{k}) = \frac{1}{mA}\sum_{\mathbf{q}} q^\mu q^\nu \Big[\varphi_{ij}(q)S_{ij}(|\mathbf{k} - \mathbf{q}|) - \delta_{ij}\sum_l \varphi_{il}(q)S_{il}(q) \Big]. \qquad (2)$$

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I is the identity matrix. In Cartesian space $\varepsilon(\mathbf{k}\omega)$ and similarly $\mathbf{D}(\mathbf{k})$ are reducible to longitudinal $\varepsilon^L(\mathbf{D}^L)$ and transverse $\varepsilon^T(\mathbf{D}^T)$ matrices. The dispersion relation for the longitudinal modes is then obtained from

$$||\varepsilon^L(\mathbf{k}\omega)|| = 0 \tag{3}$$

which leads to

$$\omega^2 = \omega_0 ka(1 \pm e^{-kd}) + D_{11}^L(\mathbf{k}) \pm D_{12}^L(\mathbf{k}). \tag{4}$$

With the neglect of retardation effects, the dispersion relation for the transverse modes is derived from

$$||\varepsilon^T(\mathbf{k}\omega)^{-1}|| = 0 \tag{5}$$

which yields

$$\omega^2 = D_{11}^T(\mathbf{k}) \pm D_{12}^T(\mathbf{k}). \tag{6}$$

The **D** functions are to be expressed in terms of the structure functions, according to Eq. (2). The latter have recently been calculated [3] through the hypernetted-chain integral equation for a wide range of Γ and d values. Inputting them into the corresponding **D** functions and using the latter in Eqs. (4) and (6) one can generate a full description of the collective mode spectrum. The results are portrayed in Figs. 1–3 and the qualitative features of the collective mode dispersion are summarized below. (The figures are given for $\Gamma = 40$, corresponding to $r_s = 20$: this r_s value, while high enough for correlations to be dominant is within the domain of experimental realizability.)

(1) The spectrum of collective modes comprises four modes: two (longitudinal and transverse) in-phase modes [corresponding to the + sign in Eqs. (4) and (6)] and two (longitudinal and transverse) out-of-phase modes [corresponding to the − sign in Eqs. (4) and (6)].

(2) The in-phase modes (where the two layers oscillate in unison) are not qualitatively different from the similar modes of an isolated 2D layer [16]. In particular, for $k \to 0$ the longitudinal (plasmon) mode has the typical, quasiacoustic $\omega \sim \sqrt{k}$ dispersion, while the transverse (shear) mode is acoustic, $\omega \sim k$; both modes are softened by intralayer and interlayer correlations.

(3) The out-of-phase modes (where the oscillations of the two layers exhibit a 180° phase difference) are characterized by an $\omega(k = 0) > 0$ energy gap. The physical reason for the existence of an energy gap for layered systems has already been discussed elsewhere [8]. The out-of-phase longitudinal mode in the RPA has been identified as the *acoustic plasmon* [18,19] since for $k \to 0$, $\omega \to 0$ as k. The present calculation clearly shows the marked difference brought about by the strong correlations. Since at $k = 0$ the isotropy of the system is unbroken, the plasmon and the shear modes share a common gap value.

(4) From Eqs. (4) and (6) the gap value can be expressed as

$$\omega^2(0, d) = -\frac{\omega_0^2}{2} \int_0^\infty d(qa)\,(qa)^2 e^{-qd} S_{12}(q). \tag{7}$$

With increasing d and consequently decreasing interlayer correlations, $\omega(0)$ shows a decreasing tendency and it virtually vanishes for $d > 1.5$ when the separated layers become practically uncorrelated [3]. This downward trend is, however, preceded by a slight upturn for $0 < d < 0.16$ (for $\Gamma \geq 30$). Although the details of this behavior are not well understood, it is most likely due to the *substitutional disorder* that prevails in this region [3]: the eigenfrequencies of the localized modes in the substitutionally disordered phase are expected to be higher than in the substitutionally ordered phase [20]. Within the domain investigated, the Γ dependence of $\omega(0)$ is quite mild, but with the QLCA being a strong coupling approximation, no inference concerning the behavior of $\omega(0)$ in the moderately coupled ($\Gamma < 10$) domain can be drawn from this observation. In fact, it is expected that for low enough Γ values, $\omega(0)$ tends to zero to match at $\Gamma = 0$ the predicted RPA behavior [18].

(5) For finite k values all four dispersion curves develop an oscillatory behavior, generated by the similar behavior of the inputted structure functions. This behavior has also been identified for the isolated 2D layer [16,17(b)]. The structure of the out-of-phase plasmon

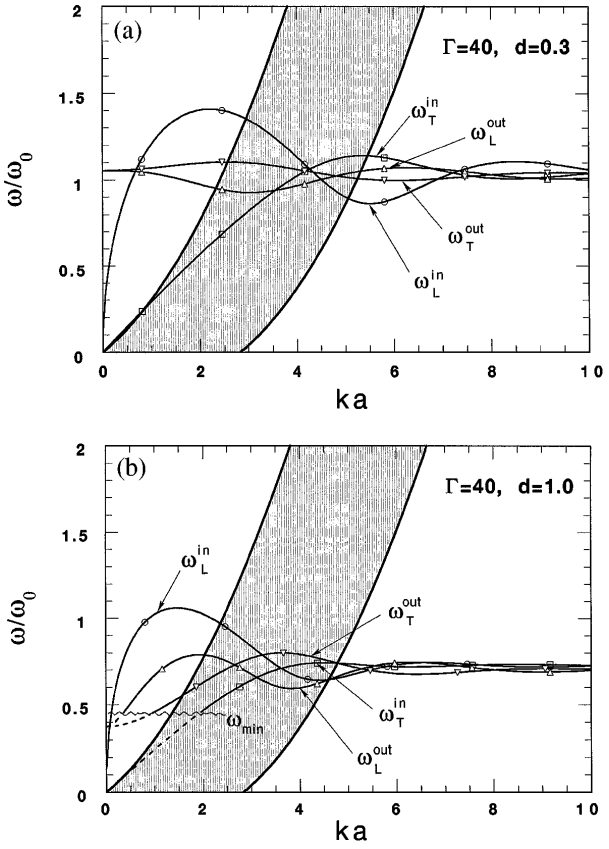

FIG. 1. The four principal modes for $\Gamma = 40$ ($r_s = 20$): (a) $d/a = 0.3$; (b) $d/a = 1.0$. The shaded region is the pair excitation continuum. For $d/a = 1.0$ ($\omega_{\min} \sim 0.45\omega_0$) the diffusional domain in which the shear mode is nonpropagating and the out-of-phase plasmon exhibits an RPA-like behavior is indicated.

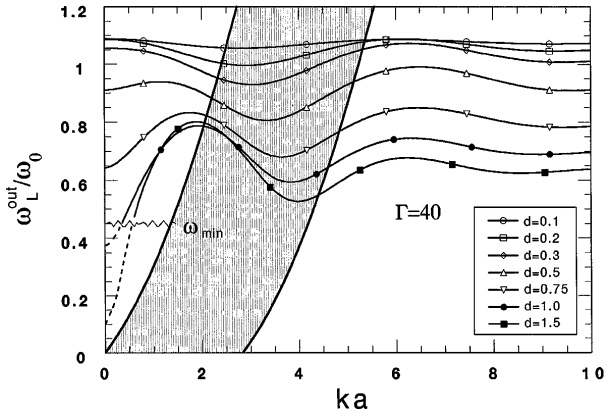

FIG. 2. The gapped out-of-phase ("acoustic") plasmon for different layer separations. The shaded region is the pair excitation continuum. In the $\omega < \omega_{\min}$ domain the $d/a = 1.0$, 1.5, and 2.0 gap values are spurious and the dispersion should resemble the RPA acoustic-type behavior.

mode is of special interest here: the first sharp rotonlike minimum has attracted attention in earlier studies [5,9,11] which were based on the neglect or on a highly approximate treatment of the interlayer correlations. It was suggested that the minimum of ω^2 may dip below $\omega^2 = 0$ [5(a),9,11] or may, at least, reach the close vicinity of $\omega^2 = 0$ [5(b),5(c)]. The former behavior would indicate a dynamical instability (heralding the onset of charge-density-wave-type ground state), [5(a),11]; the latter has been interpreted as the onset of a new high-k, low frequency mode [5(b),5(c)]. Our results show that the roton minimum never drops below the value already reached by the dispersion curve of the 2D layer. The consistent treatment of the interlayer and intralayer correlations thus precludes the existence of the effects conjectured in [5,9,11], virtually independently of any other approximation used.

(6) At high k values, for a given d all the dispersion curves approach the same asymptotic frequency value, the frequency of a localized mode, a particle oscillating in the screening environment of the two layers,

$$\omega^2(\infty, d) = \frac{-1}{mA}\sum_{\mathbf{q}}\{\varphi_{11}(q)[S_{11}(q) - 1] + \varphi_{12}(q)S_{12}(q)\}q^2 = -\frac{\omega_0^2}{4}\int_0^\infty d\,(qa)\,(qa)^2 \times [S_{11}(q) - 1 + S_{12}(q)e^{-qd}]. \tag{8}$$

This result is, probably, only of academic interest, since it is unlikely that high-k modes would survive the damping mechanisms operating in the system.

The QLCA is not geared to describe damping processes and therefore our calculation fails to provide information on the damping of the collective modes. However, some qualitative statements can be made. (We concentrate on the out-of-phase modes only.) There are three major damping mechanisms to be considered. These are (i) single pair excitations (Landau damping), (ii) multiple pair excitations, and (iii) diffusive-migrational damping [17]. The effect of the single pair excitations can be easily assessed from Figs. 1 and 2. Both of them show the pair excitation domain: it is clear that as long as the layer separation is not too large ($d/a < 1.5$) for small k values both the out-of-phase plasmon and shear modes are well outside the continuum and are thus immune to Landau damping. In a highly correlated plasma multiparticle excitations are, however, also operative, with increasing importance at higher k values. The high-k portion of the dispersion curve emerging from the continuum would probably be heavily damped by this process. The diffusive-migrational shifting of the quasisites [17] can originate from quasithermal diffusion or from tunneling between neighboring minima of the fluctuating potential. For Γ sufficiently high ($\Gamma \geq 40$, i.e., $r_s \geq 20$, probably close to crystallization [1]), the latter effect should be significantly diminished. The former, however, is sizable when its characteristic time becomes comparable with the period of the mode under consideration. Based on our earlier estimate [10] of the lowest surviving oscillation frequency, $\omega_{\min}$ and on the molecular dynamics results of Ref. [21] (see also [17]) concerning the lowest propagating wave-number value for the shear mode, one can conclude that for $\Gamma = 40$ ($r_s = 20$) the $d/a < 0.8$ domain and for $\Gamma = 20$ ($r_s = 10$) the $d/a < 0.6$ domain can be safely assumed not to be seriously affected by this damping mechanism for either of the modes (cf. Fig. 1a). For higher d values the gap frequency $\omega(0)$ is below $\omega_{\min}$ and thus the shear mode would be propagating for $\omega > \omega_{\min}$ only; in the domain $\omega < \omega_{\min}$ the longitudinal mode would be stripped of its correlational features and would revert to an RPA-type acoustic plasmon with $\omega(k \to 0) \to 0$ (cf. Fig. 1b).

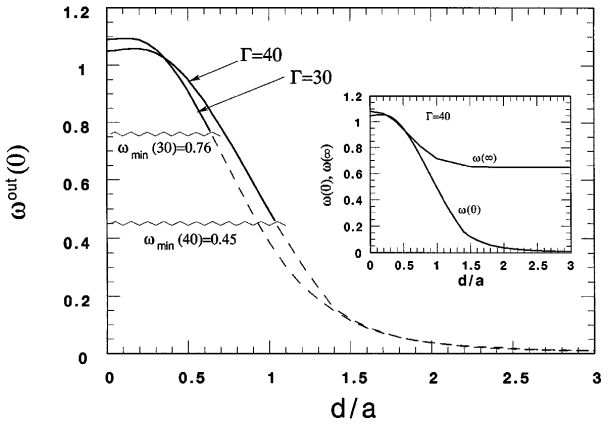

FIG. 3. Gap $[\omega(0)]$ value for $\Gamma = 40$ and $\Gamma = 30$ as functions of the layer separation. Below the estimated $\omega_{\min}(\Gamma)$ value the gap is spurious. The inset compares $\omega(0)$ and $\omega(\infty)$ for $\Gamma = 40$.

The collective mode structure of the strongly coupled bilayer liquid presented in this Letter bears a close relationship to the phonon spectrum of the bilayer solid, recently

calculated by Goldoni and Peeters [4]. The four modes in the solid phase can be identified as the transverse acoustic and the longitudinal quasiacoustic ($\sim \sqrt{k}$) phonons and the transverse and longitudinal optical phonons. The "gaps" exhibited by the latter are, in general, different because of the anisotropy of the lattice. In contrast, in the liquid state, there is only one single isotropic gap, as determined in this Letter. This liquid gap value is typically slightly above the arithmetic average of the optical frequencies of the transverse and longitudinal phonons in the solid phase.

Concerning the possible observation of the features predicted in this Letter, one can suggest three main areas that should lend themselves to direct experimental verification: (1) the existence and the nonmonotonic d-dependence of the $k = 0$ energy gap; (2) the existence of a transverse shear excitation with a high frequency and expected low damping (this is in sharp contrast to the usual scenario for the shear mode in the liquid phase, which vanishes for $k \to 0$ [17,21,22]); (3) the nonexistence of the predicted [5,6,9,11] instability or low frequency mode in the vicinity of the first roton minimum. We note that the reported Raman scattering experiments [23] are inconclusive because of the low r_s and relatively high k values involved. Recent advances in fabricating high r_s samples [24] and small layer separation should render the suggested experiments feasible.

In summary, we have obtained a comprehensive picture of the collective mode structure of an electronic bilayer in the strongly coupled liquid phase. This structure is qualitatively different both from that of the weakly coupled bilayer electron gas (describable in the RPA) and from the phonon spectrum of the bilayer solid and exhibits a number of remarkable features which should be experimentally verifiable. The calculation avoids the pitfalls of earlier approaches which stem from the inconsistencies in the approximations used for the interlayer and for the intralayer PCFs, and from the violation of the 3rd frequency-moment-sum rule.

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