Energy gap in the bilayer: Understanding the failure of the Singwi-Tosi-Land-Sjolander approximation

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We demonstrate that the conflicting predictions of the STLS (Singwi-Tosi-Land-Sjolander) and the quasilocalized charge approximations concerning the lack of, or the existence of, an energy gap, respectively, in the collective excitation spectrum of an electronic bilayer can be traced to a formal defect of the STLS scheme which renders it inappropriate for the analysis of the $k\rightarrow 0$ behavior of collective modes in binary systems. We conclude that in addition to the cogent physical reasons that mitigate in favor of its existence, the structural isomorphism between the bilayer and binary systems also mandates an energy gap in the former. [S0163-1829(98)04312-4]

Electronic bilayer systems consisting of two quasi-two-dimensional electron or hole gases separated by a fixed distance d in a double quantum well have been of intense interest in recent years. Another class of layered systems, electronic superlattices, consisting of a large number of equidistant layers, exhibit similar physical features. Such systems can now be routinely fabricated through modern semiconductor nanotechnology. Relatively high r_S $[=a/a_B;\ a=(1/n\pi)^{1/2}$ is the interparticle distance within a layer, $a_B=\hbar\varepsilon/e^2m^*$ is the Bohr radius] values have been[1] or are expected to be achieved. In this strong-coupling regime ($r_S\gg 1$), layered systems exhibit remarkable features, both on the static and dynamic levels.[2–26] One of the issues under investigation is the structure of collective excitations in strongly coupled bilayer systems where the conventional random-phase approximation (RPA) is inapplicable. Earlier approaches were based on focusing on intralayer correlations while ignoring interlayer correlations.[3,12,16] The unphysical consequences of this inconsistent approach are by now well realized.[4,16,17] Within the realm of a more consistent approximation there have been two major approaches to this problem. One approach consists of applying the well-established STLS (Singwi-Tosi-Land-Sjolander) (Refs. 27–29) approximation to the problem.[2–5,19,26] The other approach is based on the more recent QLC (quasilocalized charge) method.[11–14,16–18] The predictions of the two methods are quite different. In particular, the QLC method predicts the existence of a finite energy gap ($\omega>0$ for $k=0$) in the excitation spectrum of the out-of-phase modes: no such energy gap appears as the result of the STLS calculations. This paper addresses the question of the origin of this discrepancy.

It is well known that the electronic bilayer can be mapped onto a single two-component two-dimensional layer.[12] Thus we focus on the formal description of a multicomponent system of charged particles, possibly with a neutralizing background. Using a matrix formalism in species space,[30] the dielectric matrix can be expressed quite generally in terms of the screening function $\mathbf{u}(\mathbf{k}\omega)$:

$$\varepsilon(\mathbf{k}\omega)=1-\varphi(\mathbf{k})\chi_0(\mathbf{k}\omega)\{1-\mathbf{u}(\mathbf{k}\omega)\chi_0(\mathbf{k}\omega)\}^{-1}. \quad (1)$$

$\mathbf{u}(\mathbf{k}\omega)$ is related to the more customary ''mean-field'' $\mathbf{G}(\mathbf{k}\omega)$ by $\mathbf{u}_{AB}(\mathbf{k}\omega)=\varphi_{AB}(\mathbf{k}\omega)G_{AB}(\mathbf{k}\omega)$. $\chi_{0,AB}(\mathbf{k}\omega)$ is the partial density response matrix[30,31] for the noninteracting system (and thus diagonal in species space); the interaction potential $\varphi(\mathbf{k})$ is not necessarily pure Coulombic, since it may absorb other features of the interaction.

The screening function $\mathbf{u}(\mathbf{k})$ is static (i.e., independent of ω) both in the STLS and in the QLC approximations. The two approximation schemes also agree with each other in that $\mathbf{u}(\mathbf{k})$ is determined as a functional of the equilibrium pair correlation function $h(\mathbf{r})$ or its Fourier transform $h(\mathbf{k})$. Where they deviate from each other is both the underlying physical picture and the precise functional dependence of $\mathbf{u}(\mathbf{k})$ on $h(\mathbf{k})$.

The fundamental assumption of the STLS approximation is that even when the system is outside equilibrium due to an external perturbation, particle correlations are well approximated by their equilibrium value. The resulting expression for $\mathbf{u}(\mathbf{k})$ is[27–29]

$$u_{AB}(\mathbf{k})=\frac{1}{V}\sum_{\mathbf{q}}\frac{\mathbf{k}\cdot\mathbf{q}}{k^2}\varphi_{AB}(\mathbf{q})h_{AB}(\mathbf{k}-\mathbf{q}) \quad \text{(STLS)} \quad (2)$$

We are interested in the $k\rightarrow 0$ behavior of the collective excitations of the system: thus we analyze $\mathbf{u}(k\rightarrow 0)$. Consider first a three-dimensional binary system (say, a binary ionic mixture[32,33]) with pure Coulomb interaction

$$\varphi_{AB}(\mathbf{k})=\frac{4\pi e^2}{k^2}Z_AZ_B, \quad (3)$$

where eZ_A and eZ_B are the species charges. We focus on the $k\rightarrow 0$ limit of $\mathbf{u}(\mathbf{k})$. One finds for the diagonal and off-diagonal terms

$$u_{AA}(k\rightarrow 0)=\frac{2}{3\pi}e^2Z_A^2\int_0^\infty dq h_{AA}(q) \quad \text{(STLS)}, \quad (4)$$

$$u_{AB}(k\to 0)=\frac{2}{3\pi}\,e^2 Z_A Z_B\int_0^\infty dq h_{AB}(q) \quad \text{(STLS)}$$

$$(A\neq B). \quad (5)$$

The important feature of the above relations is that both the diagonal and the off-diagonal elements of $\mathbf{u}(\mathbf{k})$ approach a constant (negative) k-independent value in the $k\to 0$ limit.

Consider now the system of our principal interest, an electronic bilayer, consisting of two two-dimensienal (2D) electron layers in a neutralizing background, separated from each other by distance d. The system can be mapped onto a single 2D layer with two ''components.''[12] The corresponding potentials are

$$\varphi_{11}(k)=\varphi_{22}(k)=\frac{2\pi e^2}{k},$$
$$\varphi_{12}(k)=\frac{2\pi e^2}{k}\,e^{-kd}, \quad (6)$$

and, in the $k\to 0$ limit, the diagonal and off-diagonal elements of $\mathbf{u}(\mathbf{k})$ assume the form

$$u_{AA}(k\to 0)=\tfrac{1}{2}e^2\int_0^\infty dq h_{AA}(q), \quad (7)$$

$$u_{AB}(k\to 0)=\tfrac{1}{2}e^2\int_0^\infty dq e^{-qd}h_{AB}(q) \quad \text{(STLS)} \quad (A\neq B). \quad (8)$$

Next we turn to exhibiting the calculation of $\mathbf{u}(\mathbf{k})$ in the QLC approximation. The approximation is a dynamical one, that focuses on the oscillatory motion of the quasilocalized particles trapped in local potential fluctuations. The resulting general expression for $\mathbf{u}(\mathbf{k})$ is given again in terms of the pair-correlation function[34]

$$u_{AB}(\mathbf{k})=\frac{1}{V}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^4}\left\{\varphi_{AB}(\mathbf{q})h_{AB}(\mathbf{k}-\mathbf{q})-\delta_{AB}\sum_C \varphi_{AC}(\mathbf{q})h_{AC}(q)\right\} \quad \text{(QLC)}. \quad (9)$$

We now consider the two cases as before, in the $k\to 0$ limit. For binary mixtures one finds

$$u_{AA}(k\to 0)=-\frac{4\pi e^2 Z_A Z_B}{V}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^4 q^2}h_{AB}(\mathbf{q})$$
$$=+\frac{4\pi}{3}\frac{e^2 Z_A Z_B}{k^2} \quad \text{(QLC)}, \quad (10)$$

$$u_{AB}(k\to 0)=+\frac{4\pi e^2 Z_A Z_B}{V}\sum_{\mathbf{q}}\frac{(\mathbf{k}\cdot\mathbf{q})^2}{k^4 q^2}h_{AB}(\mathbf{q})$$
$$=-\frac{4\pi}{3}\frac{e^2 Z_A Z_B}{k^2} \quad \text{(QLC)} \quad (A\neq B). \quad (11)$$

The second line follows from $(1/V)\Sigma_{\mathbf{q}}\mathbf{h}_{AB}(\mathbf{q})=h_{AB}(r=0)=-1$ for a repulsive Coulomb interaction. Similarly, for a bilayer system, one has

$$u_{AA}(k\to 0)=-\frac{e^2}{2}\frac{1}{k^2}\int_0^\infty dq\; q e^{-qd}h_{AB}(q)$$
$$=-u_{AB}(k\to 0) \quad \text{(QLC)} \quad (A\neq B). \quad (12)$$

The precise value of the integral in Eq. (11) depends on $h(\mathbf{k})$ but it is certainly finite. The obvious and profound difference between the STLS equations (4), (5), (7), and (8) and the QLC equations (10), (11), and (12), respectively, is, then, that in the $k\to 0$ limit the latter expressions become infinite while the former remain bounded. In order to see the implications of this difference we now analyze the dispersion relation for the longitudinal collective modes. The dispersion relation is obtained from the

$$\|\varepsilon(\mathbf{k},\omega)\|=0 \quad (13)$$

condition. For $\chi_0(\mathbf{k},\omega)$ we adopt the high-frequency

$$\chi_{AB}(\mathbf{k},\omega)=\delta_{AB}\frac{n}{m}\frac{k^2}{\omega^2} \quad (14)$$

expression, which can also be interpreted as the expression resulting from the neglect of the random motion of the particles.

Consider first the binary mixture: the combination of Eqs. (1), (3), (4), (5), (13), and (14) yields ($\omega_p^2=4\pi e^2[(Z_1^2 n_1/m_1)+(Z_2^2 n_2/m_2)]$)

$$\omega^2=\omega_p^2+O(k^2) \quad \text{(STLS)} \quad (15)$$

for the STLS approximation, whereas the combination of Eqs. (1), (10), (11), (13), and (14) yields

$$\omega^2=\omega_p^2+\delta\omega^2(Z_2/Z_1, m_2/m_1, n_2/n_1)+O(k^2) \quad \text{(QLC)} \quad (16)$$

for the QLC approximation. The significant difference between Eqs. (15) and (16) is manifested by the finite frequency shift $\delta\omega^2$ in $k\to 0$ limit in the QLC description (the precise value of $\delta\omega^2$, which is positive, is not relevant here; the interested reader can find the corresponding expression and the discussion in Ref. 34); in contrast, no such shift appears in the STLS approximation.

It is now not surprising that for the case of the bilayer a similar result ensues. For the STLS approximation, Eqs. (1), (6), (7), (8), (13), and (14) combine into (n_s is the surface density)

$$\omega^2=\frac{2\pi e^2 n_s d}{m}k^2+O(k^2) \quad \text{(STLS)}. \quad (17)$$

This is the well-known RPA out-of-phase plasma acoustic mode, softened by the negative $O(k^2)$ correlational correction. This result was reported in Refs. 2, 3, 4, 5, and 26. In contrast, the QLC calculation via Eqs. (1), (11), (12), and (13) leads to

$$\omega^2 = \omega^2(0) + \frac{2\pi e^2 n_s d}{m} k^2 + O(k^2), \quad \text{(QLC)} \qquad (18)$$

where the leading term is the k-independent $\omega^2(0)$

$$\omega^2(0) = -\frac{e^2 n}{m} \int_0^\infty dq\ q^2 e^{-qd} h_{12}(q). \qquad (19)$$

In other words, there is an energy gap $\omega(0)$ at $k=0$, whose value is determined by Eq. (19). This is the result obtained in Refs. 12, 13, 14, 16, and 17 for the case of an infinite superlattice, and in Refs. 12 and 18 for the case of a bilayer.

It now remains to determine which one of the two results quoted can be regarded as providing a credible description of the collective mode behavior of the bilayer. We contend that there are at least two reasons that show that the QLC approach is correct, while the STLS is not. The first is that the QLC expression for $\varepsilon(\mathbf{k}\omega)$ satisfies the third frequency moment sum rule,[34–37] while the similar STLS expression does not. This can be easily shown by comparing the coefficients of the ω^{-4} terms in the high-frequency expansion of $\varepsilon(\mathbf{k}\omega)$ as constructed with the STLS and QLC expressions given for $\mathbf{u}(\mathbf{k})$ by Eqs. (2) and (8), respectively, with the expression given in the literature (see, e.g., Refs. 32 and 33). This difference between the STLS and QLC schemes is significant since satisfaction of the third frequency moment sum rule has been recognized[38] as an important criterion in the construction of an acceptable dynamic approximation.

The second reason to accept the QLC prediction as correct is that the correlational upward frequency shift of the plasmon mode in a binary ionic mixture is a known feature of such systems, and has been verified by computer simulations.[32,33] It should be clear from the line of argument followed in this paper that from the formal point of view the energy gap in the bilayer and the frequency shift in the binary mixture are closely related to each other and the latter in fact may be regarded as an ''upward shift'' of the RPA $\omega(k=0)=0$ frequency.

The inability of the STLS (or of any static mean field) scheme to satisfy both low- and high-frequency (third frequency moment) sum rules was recognized a long time ago.[39] The consequences of this inconsistency are, however, not the same for all physical systems. While it seems clear that the STLS scheme is seriously deficient as far as the description of the dynamics of two-component systems is concerned, at the same time it is well known to be qualitatively quite reliable for a single component system, such as the electron gas. When applied to the problem of the plasmon dispersion in the latter, both the STLS and QLC reproduce—albeit with different numerical coefficients—the negative plasmon dispersion which has been identified both experimentally[40] and through molecular dynamics simulations.[41] Thus, whereas the third frequency moment sum rule is violated by the STLS scheme both for single-component and multicomponent systems, the consequences for the former are mild, but for the latter quite dramatic.

A further comment concerns the fact that the QLC is an approximation scheme which is valid in the strong-coupling regime, i.e., for high enough $r_s(=a/a_B)$ or $\Gamma(=e^2/ak_BT)$ values, where local order, making quasilocalization possible, has already set in (this can be estimated to be around $r_s \sim 6$, or $\Gamma \sim 10$). In fact, a perturbation calculation for $\Gamma<1$, performed for the binary mixture,[35,36,42] indicates $\delta\omega \sim \Gamma^{2/3}$, quite in contrast to the Γ-independent $\delta\omega$ discussed here. Thus one can conjecture that in the same fashion a similar scenario would prevail in the case of the bilayer, ensuring a smooth transition from the RPA to the strong coupling regime; at the same time this expected scenario implies that a small but finite energy gap would exist even for Γ values well below the QLC regime.

Finally, we wish to emphasize that in addition to the formal reasoning put forward in this paper to show the correctness of the QLC formalism leading to the energy gap, there exist cogent physical arguments[12–14,18] that explain its existence. These are not repeated here, since the purpose of this paper has been only to clarify the formal reasons that lead to the result discussed. One may also relate the appearance of this gap to the existence of other gapped excitations in many body systems which are certainly not uncommon. Collective modes due to long range forces, such as the three-dimensional plasmon, and the optical frequencies of ionic and other crystals constitute one class of examples; others, such as the superconducting or excitonic gaps are generated by the formation of bound pairs in the ground state; a gap in the collective mode spectrum can also be due to a finite energy jump in the single-particle energy spectrum, a common occurrence in magnetized systems (cf. Ref. 43). The physical origin of the gap in the bilayer (or in the superlattice) seems to be different from any of the above mechanisms and should be sought in the spatial nonuniformity of the system.

In conclusion, we have shown that the STLS approximation scheme is structurally incapable of generating finite frequency shifts at $k=0$, even in cases where the existence of such a shift is well established; thus any prediction based on the STLS scheme concerning the $k=0$ behavior of collective modes in a bilayer should be regarded as unreliable. We have also shown that the prediction of the QLC approximation of a finite energy gap at $k=0$ in the collective-mode spectrum is an unavoidable formal consequence of the structure that has already led to the description of well-understood and experimentally verified effects in other situations.

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