Phases in Strongly Coupled Electronic Bilayer Liquids

V.I. Valtchinov (a,b), G. Kalman (a) and K.B. Blagoev (a)

(a) Department of Physics, Boston College, Chestnut Hill, MA 02167, USA
(b) Department of Radiology, Brigham and Women’s Hospital, Harvard Medical School, Boston, MA 02115, USA

The strongly correlated liquid state of a bilayer of charged particles has been studied via the HNC calculation of the two-body functions. We report the first time emergence of a series of structural phases, identified through the behavior of the two-body functions.

PACS numbers: 71.45.-d, 73.61.-r, 64.70.Ja, 64.90.+b

Electronic bilayer systems, consisting of two quasi-two-dimensional layers of electron (or hole) liquids, separated by a distance \( d \) comparable to the interparticle separation \( a \) (the Wigner-Seitz radius) within the layers, have revealed an unexpected richness of features and have been the object of many recent investigations. While most of the interest has originated from the quantum Hall effect, and thus the related studies have been directed at bilayers in strong magnetic fields, more recently it has been recognized that unmagnetized bilayers also constitute a physical system of remarkable complexity. Electronic bilayer systems in semiconductors can now be quite routinely fabricated by modern nano-technology in a fairly wide range of densities (i.e. \( r_s \) values) and interlayer separations. Interestingly, virtually similar systems have come into existence also under quite different circumstances in ionic traps, where the charged particles are classical ions and the layers develop as a result of the system seeking its minimum energy configuration.

At high enough \( r_s = a/a_B, a_B = \frac{4\pi\hbar^2}{m_e e^2} \) (the effective Bohr radius) or \( \Gamma = \frac{e^2}{\alpha k_B} \) (estimated to be between \( \Gamma = 98 \) and \( \Gamma = 222 \)) the bilayer is expected to crystallize into a Wigner lattice. While there is no direct experimental evidence on the formation of such an electronic solid in semiconductor bilayers without magnetic field (since the required \( r_s \) values do not seem to be attainable by present-day experimental technique), the formation of layered crystalline structures has been observed in molecular dynamics computer simulations and in ionic traps. At the same time, a series of rather thorough theoretical investigations have been carried out predicting the formation of classical Wigner lattices in electronic or ionic bilayers. These studies have revealed the existence of distinct structural phases, whose characteristics depend on the layer separation, i.e. on the \( d/a \) ratio: (see inset in Fig. 2) the staggered rectangular (ii); the staggered rhombic (iv), and the staggered triangular (v). Structure (ii) comprises the simple triangular (i) and the staggered square (iii), as special cases. A similar quantum mechanical study for the magnetized bilayer has led to very similar predictions.

The understanding of the experimentally more important (and theoretically perhaps more challenging) liquid phase, on the other hand, is quite incomplete. Its analysis hinges upon the availability of the intralayer and interlayer two-body functions, \( g_{11}(r) \) and \( g_{12}(r) \), respectively. While attempts have been made to determine the \( g_{ij} \)-s through various approximations, none of the results obtained can be considered reliable especially with regard to correctly reflecting the developing short range order: Zheng and McDonald and Neilson and collaborators have used the STLS (Singwi-Tosi-Land-Sjolander) approach, whose limitations are well-known; Kalman, Ren and Golden have used a quasi-iterative method, which breaks down for small layer separations.

The purpose of this Letter is to provide the first reliable calculation of the pair correlation functions \( g_{ij}(r) \) and related quantities such as the structure functions \( S_{ij}(k) \) and the dielectric matrix \( \epsilon_{ij}(k) \) for a bilayer system. The results show that different short range ordered structures develop already in the liquid phase, at relatively modest coupling values, emulating the different structural phases in the Wigner lattices. The gradual increase of the layer separation is accompanied by dramatic changes in \( g_{ij}(r) \), \( S_{ij}(k) \) and \( \epsilon_{ij}(k) \) that can be partially interpreted in terms of the structural changes referred to above, partially in terms of the new type of a substitutional ordered-disorder transition. The bilayer seems to be a unique liquid system exhibiting structural transformations of the phase pursuant to the change of a system parameter: such transformations are commonly known only in the solid phase.

Our method of approach is based on the classical HNC (Hyper-Netted Chain) approximation. This method has proven to be extremely reliable and accurate for Coulomb systems, both in three dimensions and two dimensions. We have adopted the HNC method to the bilayer situation, by mapping the (single component) bilayer onto a two-component 2D system, with an interaction potential matrix

\[
\varphi_{11} = \varphi_{22} = \frac{2\pi e^2}{r}; \varphi_{12} = \varphi_{21} = \frac{2\pi e^2}{\sqrt{r^2 + d^2}},
\]

(1)
and using the two-component equivalent of the HNC method. The two layers are assumed to have equal densities \( n \). The resulting system of equations for \( g_{ij}(r) = h_{ij}(r) + 1, c_{ij}(r) \), and their Fourier transforms \( h_{ij}(k) \) and \( c_{ij}(k) \)

\[
 h_{ij}(r) = e^{h_{ij}(r) - c_{ij}(r) - \phi_{ij}(r)} - 1; h_{12}(k) = \frac{c_{12}(k)}{[1 - nc_{11}(k)]^2 - [nc_{12}(k)]^2}; h_{11}(k) = \frac{c_{11}(k) + nc_{12}(k)h_{12}(k)}{1 - nc_{11}(k)}; \tag{2}
\]

has been solved following Lado’s original work on 2D systems. The underlying model is a classical one, where the average kinetic energy is represented by the inverse temperature parameter \( \beta \) and the interlayer coupling by the parameter \( \Gamma = \frac{\hbar^2}{4mk_b} \). This description is appropriate for bilayers arising in ionic traps if the confining potential is steep enough so that the degree of freedom perpendicular to the bilayer planes is suppressed; it is expected to be also quite adequate for the analysis of the spin-independent structural phases of the zero-temperature electron liquid, in the domain of high enough coupling \( (a > a_B) \) where the electrons are quasi-localized and the effect of intralayer exchange becomes small. Interlayer tunneling effects also disappear under the usually less stringent \( d > a_B \) condition. Under these circumstances, invoking the equivalence \( \Gamma \approx 2r_s \) is a reasonable prescription for the interpretation of the results derived from the present model of semiconductor bilayers.

Our result for the intralayer \( g_{11}(r) \) and interlayer \( g_{12}(r) \) two-body functions are summarized in Figs.1 which show their variation as functions of the coupling \( \Gamma \) and of the interlayer separation \( d/a \). In order to quantitatively analyze the information contained in the results we have used as principal indicators:

(i) \( g_{12}(0) \), the measure of the probability of finding a particle in layer 2 above another particle in layer 1: when the interlayer correlations become significant, \( g_{12}(r) \) approaches zero;

(ii) \( R_I, R_{II}, R_{III} \) and \( Q_I, Q_{II}, Q_{III}, \) the positions of the first three maxima of \( g_{11}(r) \) and \( g_{12}(r) \), respectively: these maxima identify the correlation-shells around a given particle, both in its own layer and in the layer adjacent to it;

(iii) the intralayer coordination numbers \( \rho_I, \rho_{II}, \rho_{III} \) and the interlayer coordination numbers \( \sigma_I, \sigma_{II}, \sigma_{III}, \) associated with each shell; the coordination numbers are defined by \( \rho_i = 2n\pi \int_{M'_i} r g_{11}(r)dr, \sigma_i = 2n\pi \int_{N'_i} r g_{12}(r)dr \)

where \( M'_i \) (\( N'_i \)) and \( M''_i \) (\( N''_i \)) are the minima preceding and following \( R_i(Q_i) \).

Short range order, signaled by the onset of oscillating behavior for \( g_{11}(r) \) appears around \( \Gamma_s \approx 3 \), more or less the same value as for the isolated 2D layer. The value of \( \Gamma_s \) doesn’t seem to be significantly affected by the layer separation \( d \). Once the short range order has developed, the most dramatic novel feature of the two-body functions is the series of quite abrupt shifts in the positions and changes in the amplitudes of the first few peaks both of \( g_{11}(r) \) and of \( g_{12}(r) \). This can be well observed in Figs. 1 and 2. These features unambiguously point at the formation and phase-transformation-like changes of short range structures in the bilayer electron liquid, which can be paralleled with the results of recent studies of the bilayer crystalline phase.

The physical features of the system are revealed through the two-body functions, the structure functions and the dielectric matrix of the system. In order to bring out the characteristic features in the details of the structure of two-body functions we have used the rather high \( \Gamma = 80 \) value for the latter. For the structure functions and for the dielectric matrix we have used the more realistic \( \Gamma = 20 \) and \( \Gamma = 10 \) values.

1. **Two-body functions** \( g_{11}(r), g_{12}(r) \).

1.1 Interlayer correlations, as inferred from \( g_{11}(0) \) seem to completely disappear for about \( d/a = 3 \), even for very high \( \Gamma \) values (see inset Fig. 2). Thus for layer separation higher than this value the two layers behave as virtually independent 2D systems.

1.2 The positions, heights, and widths, and the resulting coordination numbers of the correlation-shells shift and change drastically as the interlayer separation is increased (see Fig. 2). These changes can be brought into correspondence with the expected formation of the different lattice structures in the solid phase as discussed above. Since each lattice structure carries a precise set of nearest neighbor positions \( \{R_a\}, \{Q_a\} \) and coordination number \( \{\rho_a\}, \{\sigma_a\} \) (with \( a = 1, 2, 3... \) representing the first, second, third, etc. nearest neighbors in the lattice), these can be identified with the previously defined \( \{R_a\}, \{Q_a\}, \{\rho_a\}, \) and \( \{\sigma_a\} \) (values with \( a = I, II, III, \) representing the first, second and third shell positions). (The identification is not one-to-one; a given shell, in general, with the possible exception of the first shell, contains more than one set of nearest neighbors). Thus the formation of a series of "liquid phases" emulating the underlying lattice structural phases can be observed.

1.3 At \( d = 0 \), when the two layers are collapsed into a single 2D system with double density \( 2n \), their two two-body functions \( g_{11}(r) \) and \( g_{12}(r) \) are identical ,with \( R_{II}/R_{I} = Q_{II}/Q_{I} \approx 1.9 \) ratio, slightly higher than the characteristic value of a triangular lattice. The \( R_I = Q_I = 1.25 \) value itself is somewhat below the lattice constant \( R_I = 1.34 \), similarly to what has been obtained in the Monte Carlo and HNC calculations for an isolated 2D electron liquid. The double density triangular structure can also be regarded as a staggered rectangular lattice with a side
The identity of the two-body functions $g_{11}$ and $g_{12}$ and the accompanying equality of the coordination numbers, $\rho_1 = \sigma_1$, however, clearly indicates that the two species are in a complete substitutional disorder, occupying the lattice sites at random.

With $d$ increasing away from the zero value, the substitutional disorder is rapidly replaced by a substitutional order appropriate for the staggered rectangular structure: by $d/a = 0.5$, $\rho_1$ and $\sigma_1$ assume their expected $\rho_1 = 2$ and $\sigma_1 = 4$ values. At the same time, the rectangular unit cell deforms toward a square-like shape: this is well shown by the reduction of $R_{11}/R_1$ to the $\sim 1.5$ value. The substitutional disorder that prevails in the domain $0 < d/a < 0.5$ would seem to require that vestiges of the peaks of $g_{11}(r)$ show up in $g_{12}(r)$ and vice versa. This, however, is apparently not the case: $R_1$ and $Q_{11}, R_{11}$ and $Q_{111}$, etc. are quite distinct. Nevertheless, the separations between $R_1$ and $Q_1$ remain small enough and the peaks wide enough to accommodate the substitutionally ill-positioned particles. In fact, the $Q_{111}$ shell appears to be thriving on the ill-positioned population: by the establishment of the substitutional order the $II$-shell at $Q_{11} = 3.65$ vanishes and is replaced by the more distant position at $Q_{111} = 4.99$.

The transformation to an underlying staggered square lattice structure becomes complete at $d/a = 0.7$. The indicators of this transformation are the jump of $R_{11}$ from 2.30 to 3.89 and $R_{11}$ from 3.87 to 5.63, and the accompanying jump of the coordination numbers $\rho_1$ from $\sim 2$ (same as $\rho_1$ for the rectangular lattice) to $\sim 7$ (close to $\rho_1 + \rho_2 = 8$ ($\rho_1 = 4, \rho_2 = 4$) for the square lattice). All these changes reflect the higher symmetry of the square lattice.

In the domain $d/a > 0.7$ the staggered square lattice gradually transforms into a staggered rhombic lattice with $90^\circ > \varphi > 60^\circ$: this allows the energetically favorable increase of the lattice constant, at the cost of the decrease of the energetically less important second and third neighbors. This trend is very clearly observable in the behavior of $R_1$, $R_{11}$, and $R_{111}$ also detectable in $Q_1$, and $Q_{111}$. There is no significant change in the corresponding shells.

Around $d/a = 1.6$, $\varphi$ reaches the $60^\circ$ value and in each layer the underlying structure becomes a standard $2D$ triangular (hexagonal) lattice, with lattice constant appropriate for density $n$. This can be inferred from $R_1$ reaching its maximum and $R_{11}$, $R_{111}$ and $Q_1$, $Q_{11}$, $Q_{111}$ reaching their minimum values, the former very close to $\sqrt{2}$ times the $R_1(d = 0)$, $R_{11}(d = 0)$ and $R_{111}(d = 0)$ values respectively.

The triangular lattices emerging as the end product of the rhombic structure are staggered in such a way that the vertices of layer 2 lie over the midpoints of the sides of the triangles in layer 1 (and vice versa). This is not the case: $Q_{11}$ becomes dominant, approaching $\sim 25$. This behavior is rapidly replaced by a substitutional disorder which requires that $n \int d^2r \{h_{11}(r) + h_{12}(r)\} = -1$. For high $d$ values $L_{11}$ becomes dominant, approaching the value appropriate for the isolated 2D layer, which, for $\Gamma \geq 1$ is $L_{11} = 1 \sim 0.821 \Gamma$. The inset in Fig.2 shows the $L_{11} - L_{12}$ values, extracted from $S_{11}(k)$ and $S_{12}(k)$, corroborating the expected behavior.

The inverse of the static dielectric response matrix $\eta(k) = \varepsilon^{-1}(k)$ can be expressed in terms of the structure functions as $\eta_{kij} = \delta_{ij} - \beta n \varphi_{\eta}(k) S_{ij}(k)$. It determines the total potential $\Phi_1$ in the two layers when either of them is perturbed by an external potential $\Phi$: $\Phi_1 = \eta_{kij} \Phi_j$.

At small layer separation the effective dielectric function $\varepsilon_{eff}^{kij}$ has the features of the dielectric function of an isolated 2D layer (see Fig. 4) with the characteristic anti-screening behavior for $k < k_0$ ( $\sim 25$). This behavior becomes qualitatively different precisely at the layer separation where $L_{11} - L_{12}$ changes from positive to negative (see inset in Fig. 4): $\varepsilon_{11}$ and $\varepsilon_{12}$ develop alternatively anti-screening and screening domains.

We can compare our results concerning the critical values of $d/a$ at the phase transition boundaries with those of Goldoni and Peeters (GP) into our $d/a$ values Table I shows the comparison. The $d/a$ ([ii] $\rightarrow$ [iii]) value has been taken along the liquid-solid phase boundary at the lowest $\Gamma$ value ($\Gamma = 118$) in Fig. 8 of their paper. GP claim a wide range of stability $(0.79 < d/a < 1.10)$ for the staggered square lattices. We don’t see this happening: the gradual transformation into a rhombic structure seems to follow immediately the formation of the square lattice. The expected first order character of the (iv) - (v) transition is masked in the present description. Neither is there a clear indication of the transition taking place at $\varphi = 69.6^\circ$ predicted by GP rather than at $\varphi = 60^\circ$ the indicator would
be a slight decrease in the $\rho_i$ values accompanying the detectable slight increase in the $Q_i$ values). The possibility for substitutional disorder is not part of the GP lattice model which allows for structural changes only, although, as our analysis reveals, it would be, for a finite temperature, a determining factor of the phase structure of the system in the $d/a < 0.5$ domain.

In summary, we have calculated, the two-body functions, structure functions and the static dielectric matrix of a strongly correlated electronic bilayer liquid through the classical HNC approach. We have found a series of dramatic changes in the liquid structure as the layer separation varies from $d/a = 0$ to $d/a = 3$. These changes can be brought into correspondence with the structural transformations and with substitutional order-disorder transformations in the underlying lattice structure. This behavior seems to be unparalleled in other strongly correlated liquid systems. We have also shown how the structure functions and the static dielectric matrix reflect these structural changes.

This work has been partially supported by NSF Grant PHY-9115714. Part of the work was done while GK was visiting at the University of California at San Diego and is grateful to Tom O’Neil for his hospitality. We would like to thank Dan Dubin for the discussions and for making his unpublished data available and to Ken Golden of the University of Vermont for a series of useful conversations.

[1] D.H.E. Dubin, Phys. Rev. Lett. 66, 2076 (1991); *ibid.* 71, 2753 (1993); D.H.E. Dubin, Phys. Fluids B 5, 295 (1993).
[2] H. Totsuji and J.-L. Barrat, Phys. Rev. Lett. 60, 2484 (1988); R.W. Hasse and J.P. Schiffer, Ann. Phys. (NY) 203, 419 (1990); A. Rahman and J. Schiffer, Phys. Rev. Lett. 57, 1133 (1986); J.P. Schiffer, Phys. Rev. Lett. 71, 818 (1993).
[3] S.L. Gilbert, J.J. Bollinger, and D.J. Wineland, Phys. Rev. Lett. 60, 511 (1988); G. Birkl, S. Kassner, and H. Walther, Nature 357, 310 (1992); J.N. Tan, J.J. Bollinger, B. Jelenovcic, W.M. Itano, and D.J. Wineland, in *Physics of Strongly Coupled Plasmas*, ed. by W. D. Kraeft and M. Schlages, (World Scientific, 1996), p. 387.
[4] G. Goldoni and F.M. Peeters, Phys. Rev. B53, 4591 (1995).
[5] V.I. Falko, Phys. Rev. B49, 7774 (1994).
[6] K. Esfarjani and Y. Kawazoe, J. Phys. Condens. Matter 7
[7] S. Narasimhan and T.L. Ho, Phys. Rev. B62, 12291 (1995).
[8] L. Zheng and A.H. MacDonald, Phys. Rev. B49, 5522 (1994).
[9] L. Swierkowski, D. Neilson, and J. Szymanski, Australian J. Phys. 46, 423 (1993); D. Neilson, L. Swierkowski, J. Szymanski, and L. Liu, Phys. Rev. Lett. 71, 4035 (1993); *ibid.* 72, 2669 (1994).
[10] G. Kalman, Y. Ren, and K.I. Golden, in *Proceedings of the VI Workshop on the Physics of Nonideal Plasmas*, ed. by T. Bornath and W.D. Kraeft [Contrib. Plasma Phys. 33, 449 (1993)].
[11] G.S. Stringfellow, H. DeWitt, and W. Slattery, Phys. Rev. A41, 1105 (1989).
[12] F.J. Rogers, D.A. Young, H.E. DeWitt, and M. Ross, Phys. Rev. A28, 2990 (1983).
[13] H. Totsuji, Phys. Rev. A17, 399 (1978).
[14] R.C. Gann, S. Chukravarty, and G.V. Chester, Phys. Rev. B20, 326 (1979).
[15] F. Lado, Phys. Rev. B17, 2827 (1978).
[16] V.I. Valtchinov, G. Kalman, and K.B. Blagoev, in *Physics of Strongly Coupled Plasmas*, (Ref. 3), P139.
[17] G. Kalman and K.I. Golden, *unpublished*.
[18] K.I. Golden, G. Kalman, and Ph. Wyns, Phys. Rev. A61, 6940 (1990).

CAPTIONS FOR FIGURES.

Fig. 1. Variation of the structure of the two-body functions with increasing layer separations $d/a$ at $\Gamma = 80$: (a) $g_{11}(r)$; (b) $g_{12}(r)$.

Fig. 2. Characteristic variation with layer separation of the first, second and third maxima of $g_{11}(r)$ and $g_{12}(r)$: (a) positions $R_I$, $R_I(1)$ associated coordination numbers $\rho_{11}$, $\rho_{11}$, $\rho_{11}$ and $\sigma_1$, $\sigma_1$, $\sigma_{11}$. The insets show the five principle lattice structures and the value $g_{12}(r)$ at $r = 0$.

Fig. 3. Variation of the structure functions with increasing layer separations $d/a$ for $\Gamma = 20$ for $S_{11}(k)$ and $S_{12}(k)$. Note that the coordinates of $S_{11}$ and $S_{12}$ are shifted with respect to each other.

Fig. 4. Diagonal and off-diagonal elements of the static dielectric matrix for different layer separations (a) $d/a = 0.05$ ($L_{11} > L_{12}$) (b) $d/a = 0.1$ ($L_{11} < L_{12}$) at $\Gamma = 10$. The insets show $deti_{ij}$ and $L_{11} - L_{12}$, the difference between diagonal and transverse compressibilities.

CAPTION FOR TABLE
Phase boundaries ((i): simple triangular; (ii/a): substitutionally disordered staggered quadrangular; (ii/b): substitutionally ordered staggered quadrangular; (iii): staggered square; (iv): staggered rhombic; (v): staggered triangular)

| Phases          | $d/a$  |
|-----------------|--------|
| This work       | GP     |
| $(ii/a) \rightarrow (ii/b)$ | $\sim 0.5$ |
| $(ii) \rightarrow (iii)$     | $0.7 \sim 0.8$ | $0.79$ |
| $(iv) \rightarrow (v)$       | $1.5 \sim 2.0$ | $1.87$ |
$\Gamma = 10$
$d/a = 0.10$
$L_{11} - L_{12} < 0$