Electron Correlation and Charge Transfer Instability in Bilayered Two Dimensional Electron Gas

Sergio Conti(1,2) and Gaetano Senatore(3,2)

(1) Scuola Normale Superiore, Piazza dei Cavalieri, I-56126 Pisa, Italy
(2) Istituto Nazionale di Fisica della Materia
(3) Dipartimento di Fisica Teorica, Università di Trieste, Strada Costiera 11, I-34014 Trieste, Italy
(September 27, 1996)

We prove that the predicted charge transfer state in symmetric bilayers of two dimensional electron gases is always unstable at zero bias voltage, due to interlayer correlation and/or tunneling. This is most easily seen by resorting to a pseudospin formalism and considering coherent states obtained from the charge transfer state through rotations of the pseudospins. Evidently, the charge transfer state is stabilized by a sufficiently strong gate voltage, as found in recent experiments. We show that a simple model, in which the layers are strictly two dimensional, is able to account quantitatively for such experimental findings, when correlation is properly included.

Two dimensional electron gas (2DEG) systems confined in semiconductor space charge layers (e.g., Si inversion layers, GaAs heterojunctions, and quantum wells) have provided for the last twenty years an ideal laboratory for studying various electron–electron interaction effects under almost ideal 2D jellium conditions. The zero temperature phase diagram has attracted a lot of attention since Wigner pointed out that at low density electrons would crystallize to minimize the potential energy. A stable spin polarized phase at intermediate density was predicted by Hartree-Fock (HF) calculations, as a consequence of competition between kinetic and exchange energy. The most recent Diffusion Monte Carlo (DMC) simulations by Rapisarda and Senatore (RS) confirm this picture and show that the 2DEG is paramagnetic at low density 2DEG is negative, Ruden and Wu (RW) argued that exchange and correlation could overcome the kinetic and Hartree energy costs stabilizing a charge transfer state (CTS), in which one layer contains all the electrons and the other is empty. Their HF computation, restricted to the ideal zero-tunneling symmetric case, predicts stability of the CTS for

\[ r_s > \frac{3\sqrt{2} + 1}{16} \left( 1 + 2 \frac{d}{a_B} \right) \simeq 1.42 \left( 1 + 2 \frac{d}{a_B} \right), \]

which is well into the range of the experimentally attainable electron densities for reasonable layer separations \( d \) (e.g., with GaAs parameters, \( r_s = 4 \) or \( n_{2D} = 2 \cdot 10^{10} \text{cm}^{-2} \) for \( d < 90 \text{Å} \), according to Ref. [3]). In fact higher values of the coupling (\( r_s \approx 20 \)) have been achieved [10] working with holes, rather than electrons. Note that in the bilayer we relate the density/coupling parameter \( r_s \) to the total areal density.

Evidence for a CTS in GaAs/AlAs double quantum well structures in the presence of bias voltage was recently reported [11–13]. The experimental setup allowed to measure independently the charge contained in the two layers as a function of bias voltage, via Shubnikov-de Haas oscillations. For low voltages the transferred charge was found to depend linearly on \( V^c \), in quantitative agreement with the predictions of a simple equivalent circuit model, but for high enough voltages \( V^c \approx 1V \) the charge of one layer dropped abruptly from around 20% of the total charge to 0.

Here we shall demonstrate that, for two dimensional electron layers and at zero bias voltage, the CTS is unstable at any density and layer separation \( d \). A convenient model Hamiltonian for the coupled electron layers is obtained introducing a pseudospin variable \( \tau_i \), which labels the planes with the convention that \( \tau_i^{(z)} = 1/2 \) (−1/2) if electron \( i \) is in the upper (lower) plane. For fixed interlayer distance and background densities and in the absence of tunneling, the Hamiltonian takes the form

\[
H = \sum_i \frac{\tau_i^2}{2m} + \frac{1}{2} \sum_{i \neq j} \left( \frac{e^2}{r_{ij}} + 4\tau_i^{(z)} \tau_j^{(z)} \right) \frac{\tau_i^{(z)} \tau_j^{(z)}}{2} + \frac{e^2}{\sqrt{d^2 + r_{ij}}} \left( 1 - 4\tau_i^{(z)} \tau_j^{(z)} \right) + V^c \sum_i \tau_i^{(z)},
\]

where \( V^c \) is the energy difference between the two planes, given by \( 2\pi \varepsilon^2 (\rho_1 - \rho_2) d \) in the absence of external fields, \( \rho_1 \) and \( \rho_2 \) being the background densities of the two planes. The prime on the second sum indicates that...
the term with $k = 0$ should be omitted when the sum is rewritten in reciprocal space. It is clear that with $N$ electrons there are two CTS’s, corresponding to two eigenstates of the total pseudospin component along $z$, $\mathcal{T}(z) = \sum \tau_i^{(z)} = \pm N/2$. In other words CTS is the same as (pseudospin) ferromagnetism, with the magnetization oriented along the $z$ axis. We shall show that without bias voltage there can be no such ferromagnetic phase transition. If any, in the presence of tunneling a transition takes place to a (coherent) correlated state, which has the same symmetry as the charge transfer state but does not discharge any charge. The charge transfer phase transitions can take place in the presence of significant sample asymmetry and/or gate voltage.

It is interesting to consider the ideal case $d = V^e = 0$, where both real spin $\sigma$ and pseudospin $\tau$ are conserved and the system is actually a 4-component monolayer 2DEG. At large density (small $r_s$) the ground state is completely paramagnetic, with the four components equally populated to minimize the Fermi energy; in the opposite low density regime the ground state is known to be a Wigner crystal and essentially insensitive to spin polarization. In the intermediate regime the 4-component 2DEG can be expected to mimic the above-mentioned behavior of the 2DEG (and 3DEG), with the appearance of a spin-polarized ground state. In HF, the 4-component 2DEG is unstable towards the 2-component phase at $r_s = 3\pi/16(\sqrt{2} - 1) \approx 1.42$ and then towards the 1-component phase at $r_s = 3\pi/8\sqrt{2}(\sqrt{2} - 1) \approx 2.01$, the first being obviously identical with the $d \rightarrow 0$ limit of the RW results and the latter with the usual spin polarization transition of the 2DEG in HF.

To assess the actual presence of these phase transitions we performed Slater-Jastrow Variational and Fixed Node Diffusion Monte Carlo simulations for the 4-component 2DEG, and compared them with the RS results. The method is completely analogous to the one employed by RS, with the only difference that our DMC code moves all particles at every step, therefore requiring smaller time steps which, in turn, makes time step extrapolations unnecessary. To check the code we duplicated one of the RS data points, finding excellent agreement within statistical noise. Our results are reported in Table II, and compared in Figure 1 with the data by RS on the 2 and 1-component 2DEG. At variance with the simple HF prediction, the figure clearly shows that the 4-component phase crystallizes at $r_s \approx 42$, being always stable at smaller couplings with respect to the other two fluid phases that we have considered. One could argue that backflow corrections might spoil the validity of our results, but we do not think this to be the case since backflow corrections are known to lower the paramagnetic energies more than the ferromagnetic ones.

As we have already mentioned, a total charge transfer state is described in our formalism as an eigenstate of each $\tau_i^{(z)}$ with eigenvalue $+1/2$. We remark that $\tau_i^{(z)}$ commutes with the Hamiltonian even at finite $d$ and $V^e$ (but zero tunneling) and we label with $|\Psi_{ch.tr.}\rangle$ the lowest eigenstate within the charge transfer subspace. In the symmetric $V^e = 0$ case it can be easily seen that the coherent state $|\Psi_{coh.}\rangle = \exp\left\{i\frac{\pi}{4} \sum_j \tau_j^{(y)}\right\} |\Psi_{ch.tr.}\rangle$ has an average energy lower than $|\Psi_{ch.tr.}\rangle$. In fact, (i) $1/x > 1/\sqrt{d^2 + x^2}$ for any $x$; and (ii) $\langle \Psi_{ch.tr.}|4\tau_i^{(z)}\tau_j^{(z)}|\Psi_{ch.tr.}\rangle = 1$, whereas $\langle \Psi_{coh.}|4\tau_i^{(z)}\tau_j^{(z)}|\Psi_{coh.}\rangle = 0$. The variational principle implies that $|\Psi_{ch.tr.}\rangle$ cannot be the ground state of the system, even if $|\Psi_{coh.}\rangle$ is not an eigenstate of $H$.

We stress that both $|\Psi_{ch.tr.}\rangle$ and $|\Psi_{coh.}\rangle$ have the same symmetry properties under exchange of the particle’s spatial coordinates, as this is not affected by pseudospin rotation. In particular the two states are both fully pseudospin polarized, though in different directions. The average charge distribution in $|\Psi_{coh.}\rangle$ corresponds to half electrons in one layer and half in the other, and therefore it does not lose in Hartree energy with respect to the normal state, still having the exchange energy gain deriving from the full antisymmetry of the spatial part of the wavefunction. The difference with the above-mentioned HF predictions is due to the fact that no state with the same symmetry as $|\Psi_{coh.}\rangle$ was considered by RW.

No such clearcut statement can be made in the asymmetric $V^e \neq 0$ case. In general, it can be seen that the term $V^e \sum \tau_i^{(z)}$ favours finite values of $\mathcal{T}(z)$, i.e. (partial) pseudospin polarization. A quantitative estimate would rely on the detailed pseudospin dependence of the correlation energy for the bilayer system, which — to the best of our knowledge — has not yet been determined. Com-
mon wisdom saying that the phase transition in jellium models occurs directly from unpolarized to fully polarized implies the presence of a maximum in the internal energy at intermediate polarizations. In turn, this implies that if the value of $V^c$ is sufficient to raise the polarization up to this point, which may still be far below saturation, it will spontaneously saturate. This behavior would look like a 2-component to 1-component phase transition driven by an increasing $V^c$, and can possibly explain the experimental results reported in Refs. [1, 2].

It must be noticed that the state $|\Psi_{coh.}\rangle$ is a broken symmetry state, since all states of the form $|\Psi_{coh.}(\phi)\rangle = \exp\left\{i\frac{\pi}{2} \sum_j (\tau_j^{(y)} \cos \phi + \tau_j^{(x)} \sin \phi)\right\} |\Psi_{coh.\;tr.}\rangle$ are degenerate. The order parameter is the total pseudospin $T = \sum \tau$, corresponding to interlayer phase coherence, and behaves as an easy-plane ferromagnet, the z component being frozen by the interplay of correlation and exchange. The inset reports the pseudospin polarization $\xi$ as a function of bias potential $V_g$.

We shall further neglect finite thickness and tunneling effects. In the following RS, on the ground that the approximation is easily obtained from the energies of Table I and of Ref. [2] for the high $B$ situation can be directly extended to this zero field case. The role of a tunneling Hamiltonian, which can be conveniently written as

$$H_t = i \sum \tau_i^{(x)},$$

with the tunneling matrix element $t > 0$, is to break the $\phi$ symmetry and stabilize the coherent state $|\Psi_{coh.}\rangle = |\Psi_{coh.}(\phi = 0)\rangle$, in which $\langle T^{(x)}\rangle = 0$ and $\langle T^{(z)}\rangle = -N/2$. In the large $t$ case all electrons trivially lie in the symmetric state, which has exactly the same form as $|\Psi_{coh.}\rangle$. For small enough layer separation $d$ and tunneling amplitude $t$ a perturbative estimate of the critical amplitude $t^*$ at which the fully coherent state $|\Psi_{coh.}\rangle$ becomes stable is easily obtained from the energies of Table I and of RS, on the ground that the $d$ dependence is negligible, as it vanishes in first order, contrary to the tunneling term. Thus one gets $t^*/2 = E_1(r_s) - E_2(r_s)$, which at $r_s = 2$ corresponds to $t^* = 0.8 \text{ meV} = 9K$ with GaAs parameters.

The experimental setup of Ref. [1] includes a metallic gate at a large distance $D$ from the two 2DEG’s and some charged dopants, whose location and amount are unspecified and irrelevant for the present purposes. The 2DEG’s are realized with $150(180) \text{ Å}$ thick GaAs quantum wells with a midpoint separation $d$ of $220(194) \text{ Å}$ for sample A(B), and the tunneling energy $t = 0.005K (5K)$. From the work of Zheng and MacDonald [3] and from numerical simulations [8] it is clear that interlayer correlations are relevant only if $d$ becomes smaller than the average interparticle distance $r_{s,A/B}$. As in the present situation $r_{s,A/B} \sim 150 \text{ Å}$ and $d \sim 200 \text{ Å}$ correlation effects can be safely neglected. In the following we shall further neglect finite thickness and tunneling effects.

We propose a simple model which correctly includes intralayer correlations and interlayer mean field interactions and is closely related to the one used by Eisenstein et al [1] to discuss a similar experimental setup. Let us consider the gate potential $V_g^{(0)}$ corresponding to equal density $n^{(0)}$ in the two ‘active’ layers, i.e. to zero electric field between the layers. [The value of $V_g^{(0)}$ depends on details of the sample design, as location and amount of dopants, geometry, etc]. With reference to this state and under the assumption that no net charge flows in the system with varying $V_g$, the electrochemical equilibrium condition between the two 2DEG’s is written as

$$\mu(n_1) - \mu(n^{(0)}) = \mu(n_2) - \mu(n^{(0)}) + 4\pi e^2 d(n_2 - n_0)$$

and between the upper layer and the gate as

$$-e(V_g - V_g^{(0)}) = \mu(n_1) - \mu(n^{(0)}) + 4\pi e^2 D(n_1 + n_2 - 2n^{(0)})$$

We now proceed to solve numerically these two coupled equations for the unknowns $n_1$ and $n_2$ at various values of $V_g$. While the sample parameters $d$, $n^{(0)}$ and $V_g^{(0)}$ are given in Ref. [2], $D$ is not, and was determined by fitting the measured variation of the average density $(n_1 + n_2)/2$ with $V_g$. The chemical potential $\mu(n) = d(nE(n))/dn$ for the idealized single layer 2DEG is derived from the precise equation of state obtained by RS with DMC. The results shown in Figs. 2 are in very good agreement with the experimental data from Ref. [1], with significant disagreement only in the tunneling-dominated region around $n_1 = n_2$. In fact quantitative agreement (see Fig. 3) can be obtained even in this region computing the
FIG. 3. Layer densities for the bilayer B specified in the text, as a function of the bias voltage. Triangles and diamonds give the experimental results of Ref. [12]. The full curve results from the strictly two dimensional model discussed in the text, while the dashed curve is obtained by accounting for tunneling as explained in the text.

subband densities from the layer densities with no tunneling \( \tilde{n}_1, \tilde{n}_2 \) by exact diagonalization of a noninteracting electron hamiltonian with finite tunneling,

\[
H = \sum_k \left[ \left( \frac{\hbar^2 k^2}{2m} - \mu^0(\tilde{n}_1) \right) a^\dagger_{k,1} a_{k,1} + \left( \frac{\hbar^2 k^2}{2m} - \mu^0(\tilde{n}_2) \right) a^\dagger_{k,2} a_{k,2} + t \left( a^\dagger_{k,1} a_{k,2} + \text{c.c.} \right) \right]. \tag{6}
\]

We remark that neglecting intralayer interactions would lead to the usual approximation \( \mu^0(n) = C_q n \), where the so-called quantum capacitance \( C_q \) is given by \( \pi \hbar^2 / m \). By making this assumption for both layers the above equations are linear and can be solved analytically, obtaining results in fair agreement with experiments but completely missing the nontrivial charge transfer effect signaled by the peak in the upper curve of Fig. 2.

In conclusion, we presented a pseudospin formalism to describe bilayer 2DEG systems and used it to rigorously prove the instability of the charge transfer state and to argue for the stabilization of coherent states at small finite tunneling. We also reported results from DMC simulations, which reveal the stability of the pseudospin unpolarized state at \( d = V_\varepsilon = 0 \). Finally we have shown that the presently available experimental data, being in a regime where interlayer correlations are negligible, can be quantitatively accounted for by a simple strictly two dimensional model. Clearly the more challenging and interesting situations, in which interlayer correlations are discernible and determine the details of the charge transfer, is still awaiting for both experimental and theoretical investigations.

A preliminary account of the above study was presented at the INFM-FORUM workshop on 2DEG, held in Pisa, FORUM, June 96, and at the Conference The Electron Quantum Liquid in Systems of Reduced Dimensions held in Trieste, ICTP, July 96. We acknowledge stimulating discussions with B. I. Halperin and with A. H. MacDonald.

Note added—While completing the present manuscript we became aware of a recent preprint on the same topic by Das Sarma et al [16], which is somewhat complementary to this work.

[1] F. Rapisarda and G. Senatore, Aust. J. Phys. 49, 161 (1996).
[2] K. Moon, H. Mori, K. Yang, S. M. Girvin, A. H. MacDonald, L. Zheng, D. Yoshioka, and S. C. Zhang, Phys. Rev. B 51, 5138 (1995).
[3] P. P. Ruden and Z. Wu, Appl. Phys. Lett. 59, 2165 (1991).
[4] L. Świerkowski, D. Neilson, and J. Szymański, Phys. Rev. Lett. 67, 240 (1991).
[5] D. Neilson, L. Świerkowski, J. Szymański, and L. Liu, Phys. Rev. Lett. 71, 4035 (1993).
[6] M. Alatalo, P. Pietiläinen, T. Chakraborty, and M. A. Salmi, Phys. Rev. B 49, 8277 (1994).
[7] S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. 73, 1971 (1994).
[8] F. Rapisarda, Phase Diagram of coupled electron layers, PhD Thesis, University of Trieste (1996); F. Rapisarda and G. Senatoro, in preparation.
[9] A. H. MacDonald, Phys. Rev. B 37, 4792 (1988).
[10] S. Shapira, U. Sivan, P.M. Solomon, E. Buchstab, M. Tischler, and G. Ben Yoseph, preprint.
[11] Y. Katayama, D. C. Tsui, H. C. Manoharan, and M. Shayegan, Surf. Sci. 305, 405 (1994).
[12] X. Ying, S. R. Parihar, H. C. Manoharan, and M. Shayegan, Phys. Rev. B 52, R11611 (1995).
[13] Y. Katayama, D. C. Tsui, H. C. Manoharan, S. Parihar, and M. Shayegan, Phys. Rev. B 52, 14817 (1995).
[14] L. Zheng and A.H. MacDonald, Phys. Rev B 49, 552 (1994).
[15] J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, Phys. Rev. B 50, 1760 (1994).
[16] S. Das Sarma, M. W. Ortolano, and L. Zheng, preprint cond-mat/9609182.
TABLE I. Fixed-node DMC total energies of the 4-component 2DEG for 52 particles and in the bulk limit, in Ryberg per particle. Variational results used for the size-extrapolation are also given. These data are accurately reproduced by the same fitting formula used by RS, with parameters $a_0 = -0.88115$, $a_1 = 4.439$, $a_2 = 0.14063$, $a_3 = 1.9034$.

|       | $r_s = 2$  | $r_s = 10$ | $r_s = 20$ | $r_s = 30$ | $r_s = 50$ |
|-------|------------|------------|------------|------------|------------|
| $N = 36$ | -0.5882(4) | -0.17102(2) | -0.092275(6) | -0.063561(5) | -0.039386(3) |
| $N = 52$ | -0.5728(1) | -0.17030(2) | -0.092085(4) | -0.063474(3) | -0.039354(1) |
| $N = 84$ | -0.58062(7) | -0.17055(2) | -0.092123(5) | -0.063481(4) | -0.039355(2) |
| $N = 100$ | -0.5758(1) | -0.17036(2) | -0.092062(4) | -0.063457(4) | -0.039344(2) |
| $N = 180$ | -0.57676(8) | -0.17031(2) | -0.092050(5) | -0.063453(4) | -0.039340(2) |
| $VMC_\infty$ | -0.5750(1) | -0.17019(3) | -0.092002(4) | -0.063426(3) | -0.039330(1) |
| $DMC_{52}$ | -0.5838(4) | -0.17232(4) | -0.092891(9) | -0.063975(8) | -0.039639(3) |
| $DMC_\infty$ | -0.5860(5) | -0.17221(6) | -0.09281(1) | -0.063927(9) | -0.039615(4) |