Long range Coulomb interaction in the ground state of bilayer graphene

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We report on our studies of interacting electrons in bilayer graphene in a magnetic field. We demonstrate that the long range Coulomb interactions between electrons in this material are highly important. We show that in the unbiased bilayer (where both layers are at the same electrostatic potential), the interactions can cause mixing of Landau levels in moderate magnetic fields. For the biased bilayer (when the two layers are at different potentials), we demonstrate that the interactions are responsible for a change in the total spin of the ground state for half-filled Landau levels in the valence band.

Monolayer graphene is a two-dimensional hexagonal crystal of carbon atoms which exhibits a number of physical and electronic properties that have made it the subject of intense study since it was first isolated in 2004 [1]. The existence of a gapless, conical low energy quasiparticle spectrum $E = \hbar v_F k$ near the $K$ points of the Brillouin zone (with the slope of the dispersion relation defined by the Fermi velocity $v_F \approx c/300$) has lead to the possibility of realizing relativistic effects in a table-top solid state experiment [2]. Additionally, the unusual chiral nature of the charge carriers was confirmed by the observation of an anomalous integer quantum Hall effect [3] with Landau level (LL) spectrum $E_n = \sqrt{n}\hbar v_F/\lambda_B$, including a LL at zero energy and $\sqrt{B}$ dependence of the spectrum on the magnetic field ($\lambda_B$ is the magnetic length). From a more practical point of view, the unusually high electron mobility and conductance of graphene at room temperature [4] makes this a very promising material for the fabrication of electronic devices.

Bilayer graphene (BLG) [5] consists of a pair of monolayers bonded by relatively weak dimer bonds perpendicular to the plane of the monolayer sheets (see Fig. 1). In this material, both the conduction and valence bands have low energy structure consisting of two quadratic branches separated by the energy associated with the dimer bond, $\gamma_1$. The lower conduction band and upper valence band are degenerate at the $K$ points. The existence of chiral charge carriers with a Berry’s phase of $2\pi$ was confirmed in the observation of the integer quantum Hall effect [6] where the low energy LL spectrum is approximately linear in the field with $\epsilon_n \approx \pm \hbar \omega_c \sqrt{n(n-1)}$ for $n \geq 0$ where $\omega_c$ is the cyclotron frequency. This spectrum includes a doubly degenerate LL at zero energy [7].

It has been predicted theoretically [7, 8, 9] and observed experimentally (first by doping of the bilayer [10], and then by applying electrostatic gates [11]) that a gap can be induced in the low energy band structure of BLG by breaking the symmetry between the two layers. Switching off of the conduction current (and hence, proof of principal for switching devices made from so-called ‘biased bilayer graphene’) by sweeping the Fermi energy through the gapped region has been observed at low temperatures [12], and this development has lead to a surge in interest in this system.

The single particle theory of this material is well known [5, 7, 8, 9], it has been shown that the electron-electron interactions are significant in monolayer graphene [13, 14]. The Coulomb interaction has been studied in the unbiased (and hence, ungapped) bilayer [15], while the biasing potential was considered in the context of a ferromagnetic

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transition due to short-range interactions in the mean-field approximation at zero magnetic field \[16\]. Also, certain collective modes leading to an intra-Landau level cyclotron resonance have been predicted in the presence of a magnetic field within Hartree-Fock theory \[17\]. However, the effect of the long range Coulomb interaction on the ground state of the biased system in a strong magnetic field has not been systematically investigated, and we address this problem in the current Letter.

We find that the long range nature of the Coulomb interaction makes significant changes to the properties of the low energy charge carriers for BLG in a magnetic field. Specifically, the interactions are significantly stronger for electrons in the lowest LL, and this manifests itself in an observable way. It allows the possibility of mixing of the LLs which were well-separated in energy when the interactions were not considered. By calculating the explicit form of the ground state wave function, we show how this mixing fundamentally changes the nature of the ground state in the biased bilayer, and how this change manifests itself in the total spin of the ground state of the interacting system for half-filled LLs.

In BLG, each sheet contains two inequivalent triangular sublattices of carbon atoms which we label \(A_u\), \(B_u\), \(A_l\), and \(B_l\) for the \(A\) and \(B\) sublattices of the upper and lower layers respectively (see Fig. 1). In the Bernal stacking arrangement, the inter-layer bonds consist of dimers formed from atomic orbitals associated with the \(B_u\) and \(A_l\) sublattices. The energy associated with this bond is denoted \(\gamma_1\) and throughout this Letter we assume that \(\gamma_1 = 0.4\text{eV}\).

We allow for a static electric potential \(U\) to be applied between the upper and lower layers, so that the upper layer has potential \(U/2\), and the lower layer \(-U/2\). In a strong magnetic field we can write the tight-binding Hamiltonian using the four component basis \(\{A_u, B_l, A_l, B_u\}\) with \(\xi = +1\) in the \(K\) valley, and the basis \(\{B_l, A_u, B_u, A_l\}\) with \(\xi = -1\) in the \(K'\) valley as \[7\]

\[ H_0 = \begin{pmatrix}
\xi U/2 & 0 & 0 & \xi v_F \pi^\dagger \\
0 & -\xi U/2 & \xi v_F \pi & 0 \\
0 & \xi v_F \pi^\dagger & -\xi U/2 & \gamma_1 \\
\xi v_F \pi & 0 & \gamma_1 & \xi U/2 \\
\end{pmatrix} \]  

(1)

where \(\pi\) and \(\pi^\dagger\) are the operators corresponding to electron hops between neighbouring atoms (in opposite sublattices in the same layer). The eigenvalues \(\varepsilon_n^\xi\) of Hamiltonian \(H_0\) are found from the quartic polynomial equation \[9\]

\[ \left[2n - (\xi \delta + \varepsilon_n^\xi)^2\right] \left[2(n + 1) - (\xi \delta - \varepsilon_n^\xi)^2\right] = t^2 \left[(\varepsilon_n^\xi)^2 - \delta^2\right] \]  

(2)
where \( n \in \mathbb{Z}^+ \). The energies \( t \propto \gamma_1, \delta \propto \frac{\epsilon_c}{2} \) and \( \varepsilon_\xi \) are measured in units of \( \frac{\hbar v_F}{e} \). Additionally, we denote the band of a particular LL by placing a ‘+’ (‘-’) after the level index for the conduction (valence) band. The wave functions associated with the LLs of this spectrum for \( n \geq 1 \) are given by

\[
\psi_{n\pm} = e^{iky} \left( a_{n\pm}\varphi_{n+1}, b_{n\pm}\varphi_{n-1}, c_{n\pm}\varphi_n, d_{n\pm}\varphi_n \right)^T
\]

where the functions \( \varphi_n \) for \( n \geq 0 \) are the magnetic oscillator functions in the Landau gauge and the coefficients are defined so that the overall wave function is normalized to unity. There are also levels with \( n = 0 \pm \), which have wave functions \( \psi_{0+}^{K'} = e^{iky} (\varphi_0, 0, 0, 0) \) and \( \psi_{0-}^{K'} = e^{iky} (\varphi_0, 0, 0, 0) \) with \( \varepsilon = \pm \delta \); and \( \psi_{0+}^{K} \) and \( \psi_{0-}^{K} \) are defined by Eqns. \[2\] and \[3\] with the appropriate substitutions for \( n \) and \( \xi \). When \( U = 0 \), these four states are degenerate yielding the eight-fold degeneracy (including the factor of 2 for spin) seen in the integer quantum Hall effect in BLG \[6\].

Note that for a sufficiently strong inter-layer potential, the single particle energies described here can cross at certain magnetic field strengths. We include the fermionic properties of the electrons by constructing Slater determinants for the non-interacting many body state wave functions.

To include the effects of the long-range Coulomb interaction we consider the Hamiltonian

\[
\mathcal{H}_{\text{Coul}} = \frac{1}{2} \sum_{i \neq j} \frac{e^2}{|\vec{r}_i - \vec{r}_j|}
\]

where the vectors \( \vec{r}_{i,j} \) label the positions of the electrons, and \( \epsilon \) is the dielectric constant of graphene. For graphene mounted on an SiO\(_2\) substrate, \( \epsilon_{\text{SiO}_2} = 2.5 \) \[18\]. Other methods of isolating graphene could be considered by changing the value of this constant. The effect of the interaction term is to mix the non-interacting states into some linear combination which minimizes the total energy of the system. Our analysis is conducted by employing the exact diagonalization scheme \[19\] in which we calculate the linear combination of non-interacting basis states which gives the ground state of the Hamiltonian \( \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{Coul}} \). This method entails dividing the infinite sheet into rectangular cells of dimension \( L_x \times L_y \) \[20\] and applying periodic boundary conditions to the wavefunctions at the edges of each cell. The matrix elements of the interaction over the single particle states described above are evaluated exactly by calculating the integrals over the spatial extent of the cell numerically. The interaction between the cells is taken into account by adding the Madelung energy of a charged lattice \[21\].

The number of single particle states included in the Hilbert space from which the non-interacting many body basis states are constructed is determined as follows. There are four relevant quantum numbers: The LL index \( n \), the valley \( \xi \), the spin and the momentum \( k = \pi m / L_x \). The values of the momentum are fixed when the boundary conditions are applied to the cell, and are labelled by \( 0 \leq m \leq M - 1 \) with \( M = L_x L_y / (2 \pi L_\xi^2) \). Which LLs we select is governed by the details of the system we wish to model, and \( M \) is set by computational restraints.

In order to reduce the size of the many body system (and so to improve the speed of calculation), we observe that the Hamiltonian conserves the total momentum \( \mu = \sum_{i=1}^N m_i \mod M \). Therefore, we can perform the diagonalization separately for each value of \( \mu \) which reduces the basis size to approximately the \( 1/M \)th part. Finally, since there is no spin-dependent term in the Hamiltonian, the total projection of spin on the \( z \) axis \( S_z \) is also a good quantum number. We can fix \( S_z \) to its minimum value (which is 0 for even \( N \) and \( \frac{1}{2} \) for odd \( N \)) whilst still being able to recover all eigenstates of the total spin operator \( S^2 \) \[14\].

We consider two different cases: Firstly we demonstrate the strength of the interaction by calculating the shift in the energy of each LL due to interactions for \( U = 0 \) (an unbiased bilayer). Then we examine the system where the
\[ \nu - 3 - 2 - 1 - 0 \]

\[ \text{Energy shift} - 0.6443 - 0.6443 - 0.6443 - 0.6443 \]

\[ \nu - 1 - 2 - 3 - 4 \]

\[ \text{Energy shift} - 0.6316 - 0.6222 - 0.6148 - 0.6085 \]

**TABLE I**: Energy shift per electron due to the Coulomb interaction for integer filling factors in the \(0\pm\) LL for \(U = 0\). Energy units are \(e^2/(\epsilon\lambda_B)\), the number of momentum states \(M = 3\), and the magnetic field \(B = 3\)T.

\[
\begin{array}{|c|c|c|c|c|}
\hline
\text{Landau level } n & 1+ & 2+ & 3+ & 4+ \\
\hline
\nu_n = 1 & -0.4766 & -0.5001 & -0.5242 & -0.5160 \\
\nu_n = 2 & -0.4705 & -0.4880 & -0.5176 & -0.5110 \\
\nu_n = 3 & -0.4645 & -0.4759 & -0.5111 & -0.5061 \\
\nu_n = 4 & -0.4584 & -0.4638 & -0.5046 & -0.5012 \\
\hline
\end{array}
\]

**TABLE II**: Energy shift per electron due to the Coulomb interaction in LL \(n \geq 1\) at filling factor \(\nu_n\). Energy units are \(e^2/(\epsilon\lambda_B)\), the number of momentum states \(M = 3\), and the magnetic field \(B = 3\)T.

filling factor is negative, the inter-layer potential is sizeable and the magnetic field strong. In this case, we observe changes in the total spin of the ground state as a function of \(U\) and the magnetic field strength \(B\).

We model the unbiased bilayer near half-filling by taking a single particle Hilbert space consisting of electrons in the \(0+\) and \(0-\) LLs with all possible spin and valley states at \(U = 0\). Each integer value of the filling factor \(\nu\) is simulated by taking the number of electrons \(N = (\nu + 4)M\), and we have \(M = 3\). Table I shows the results of diagonalizing the resulting many body Hamiltonian and evaluating the change in energy from the non-interacting ground state for integer filling factors. We notice that the energy shift per electron reduces slightly as the LL fills.

In Table II, we show the energy shift due to the Coulomb interaction for electrons in higher LLs (i.e. for levels with \(n \geq 1\)) in the conduction band. We have taken a single particle Hilbert space consisting of all spin and valley states within one LL. The filling factor \(\nu_n\) within LL \(n\) can range between 0 (corresponding to an empty level) and 4 (a filled level), so that \(\nu_n = 4\) and \(\nu_{n+1} = 0\) describe the same overall filling factor. The number of electrons is set by \(N = \nu_n M\), and in order to allow direct comparison with the lowest LL we restrict ourselves to \(M = 3\). Table II shows that the energy associated with the interaction of electrons is very similar in each of the higher LLs, and that the interaction energy per particle is slightly reduced as the LL is filled. We have verified that the results are identical in the valence band.

Together, Tables I and II show that the effect of the long range electron-electron interactions is considerable, and that for this value of the magnetic field \((B = 3\)T\) the shift in the higher LLs is only about two-thirds that of the lowest LL. In Fig. 2 we show the energy shift and absolute energy of filled LLs as a function of the magnetic field. The strength of the interaction scales with \(e^2/(\epsilon\lambda_B) \propto \sqrt{B}\) with a roughly constant coefficient, while the LL spacing goes with \(\omega_c \propto B\), so for lower values of the field, the \(n = 0\pm\) level crosses the \(2-\) and \(1-\) levels as shown in Fig. 2(b). The data shown here were calculated with \(\epsilon = 2.5\), modelling graphene [18] deposited on an SiO\(_2\) substrate. For suspended graphene (where \(\epsilon \approx 1\)), it is conceivable that the effect of the interaction would be even stronger. Additionally, the effect of the inter-layer potential is to bring together the valence band LLs with low index, so for
FIG. 2: (a) The energy shift per electron of filled LLs. (b) The absolute energy per electron of filled LLs showing the crossing between the $n = 0 \pm$ degenerate level and the higher LLs in the valence band. In both plots $U = 0$, and $M = 5$.

FIG. 3: The total spin of the ground state of the (a) $\nu = -2$ and (b) $\nu = -6$ systems. $M = 2$ and $N = (\nu + 8)M$. The lines show the crossing points of the single particle states. The graining is due to the finite interval between data points.

finite bias it is plausible that the interactions will cause significant mixing between these levels.

Now we turn our attention to the system with an inter-layer potential applied, so that $U \neq 0$. With a finite gap between the 0+ and 0− levels and non-zero filling factor, it is possible to consider the negatively-doped system by taking only those single particle states which are in the valence band. Therefore we select the single particle states from which to form the Slater determinants describing the non-interacting basis states by taking all spin and valley states of the 0− and 1− LLs. We have $M = 2$ and the number of electrons is related to the filling factor by $N = (\nu + 8)M$. Diagonalizing this system for half-filled LLs (so for $\nu = -6$ and $\nu = -2$), and calculating the expectation value of the total spin operator $S^2$ over the resulting ground state as a function of the magnetic field and the inter-layer bias gives the data shown in the plots in Fig. 3. We have also superimposed lines which represent the energy at which the single particle energy levels cross, as labelled in the legend. The plots show that there is an abrupt change in the total spin of the ground state, and a range of parameters where there is a non-zero polarization of the spin. This transition is not directly related to the crossings of the single particle states, since the position and slope of the transitions do not match the corresponding lines which we have superimposed on the plots. This effect is therefore purely due to the Coulomb interaction, and in particular to the exchange contribution which acts to minimize the energy of spin-polarized many body states.
Figure 4 shows the occupation of the single particle levels in the interacting many body ground state. For simplicity, we display only the LL index of the states. The actual ground state is a coherent combination of several non-interacting basis states, where the combination of LL indices is consistent but different arrangements of momentum and valley states each come with identical prefactors in the linear combination. This figure shows that in the lower-right region of the parameter space considered, the electrons occupy as many of the 1− states as possible. As the parameters change to the upper-left region, the 0− levels become successively more populated. This demonstrates the fundamental effect of the inter-layer potential and the electron-electron interactions on the nature of the ground state.

In conclusion, we have shown that the long range Coulomb interaction between electrons plays an important rôle near the Dirac point in BLG. In the unbiased case, the interactions will cause electrons in the doubly-degenerate lowest LL to be lower in energy than those in the 1− level for small-to-moderate magnetic fields. If an inter-layer bias is applied to split the valence and conduction bands, the electron-electron interactions precipitate a transition in the total spin of the ground state of half-filled LLs for certain ranges of parameters. This effect will have fundamental implications for the design of devices made from this material.

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