Correlated insulators in twisted bilayer graphene

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Experiments on graphene bilayers, where the top layer is rotated with respect to the one below, have displayed insulating behavior when the moiré bands are partially filled. We calculate the charge distributions in these phases, and estimate the excitation gaps.

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I. INTRODUCTION

Graphene bilayers, where the top layer is rotated with respect to the bottom, show remarkable properties [1–10]. These arise from the presence of a long-wavelength moiré superlattice. For twists near certain “magic angles,” the low-energy bands become very flat, and interactions dominate [11]. Moreover, the bands have nontrivial topological indices. Experimentally, one observes insulating phases at certain rational fillings of the bands. Electrostatically doping away from these rational fillings leads to superconducting phases, whose transition temperatures are large compared to the bandwidth [1,2]. An important part of understanding the physics of these systems is to identify the structure of the correlated insulating states. Here, we conduct a variational study of the various possible charge-density, spin-density, and valley-density waves, which are the most natural candidates. We find that there are “stripe” ordered spin and valley ferromagnets at fillings of \( \frac{1}{8}, \frac{3}{8}, \frac{5}{8}, \frac{7}{8} \). At fillings of \( \{1/4, 1/2, 3/4\} \), the ground state is a spatially homogeneous spin and valley ferromagnet. The stripe order should give rise to birefringence.

There is significant prior work on this problem. Two separate groups, Seo, Kotov, and Uchoa [12], and Kang and Vafek [13], recently argued for a ferromagnetic state at 1/8 filling. Kang and Vafek’s results are similar to ours, in that they find charge ordering in addition to the spin ordering. Our approach is complementary in that we explore different models for the electron-electron interactions. Kang and Vafek use a short-range interaction, while we consider two models: a long-range Coulomb interaction, and a dipolar interaction which accounts for image charges in the back gate. All three models give similar results, pointing to the robustness of the phenomena.

Related analysis is found in the Supplemental Material of Choi et al. [3], which considers the half-filled case. Lu et al. [14] found signatures of an orbital ferromagnetic state at 3/8 filling. Studies of Wigner crystallization by Padhi, Setty, and Phillips [15,16] have relevance. Also notable is the slave-spin treatment of Pizarro, Calderon, and Bascones [17], the Hartree-Fock analysis of Xie and MacDonald [18], and the projection-based technique of Repellin, Dong, Zhang, and Senthil [19]. All of these reveal different aspects of the correlated insulators.

In addition to those already described [1,2], experimental studies of the correlated insulators include measurements of the compressibility [3,4], magnetic response [5], tunneling spectroscopy [6–8], and further transport properties [9,10].

Beyond finding the lowest-energy charge configurations, we investigate the energy cost of adding a particle, adding a hole, or adding a particle-hole pair. We find that the interaction energy from these defects is large compared to the width of the moiré minibands. Consequently, one expects large interaction-driven mixing in of higher bands, which would need to be included in quantitative models. Our present study, however, is a necessary prerequisite for those calculations.

II. MODEL

Graphene forms a honeycomb lattice, with a unit cell containing two sites, denoted \( A \) and \( B \). For small twist angles (near 1°), the bilayer system displays a large moiré unit cell. There are three notable regions, denoted by \( AA \), \( AB \), and \( BA \), each forming triangular lattices (see Fig. 1). In the \( AA \) regions, the two lattices align. In the \( AB \) regions, the \( A \) sites of the lower layer line up with the \( B \) sites of the upper layer. The \( BA \) region is the opposite. The \( AB \) and \( BA \) sites together form a honeycomb lattice.

As described in Ref. [20], the electronic structure of the low-energy bands is built from Wannier states which are centered at the \( AB \) and \( BA \) sites. Each Wannier state has a three-lobed spatial structure, with each lobe centered on an \( AA \) region. Thus the Wannier state centers are in the \( AB \)-\( BA \) regions, but the charge density is peaked in the \( AA \) regions.
We use the interaction model derived by Koshino et al. [20]. They found that the Coulomb interaction energy from lobes in the $AA$ regions $r_i$ and $r_j$ is well approximated by

$$V_{ij} = \begin{cases} \frac{e^2}{9\varepsilon_0L_M} \frac{1}{|r_i - r_j|}, & r_i \neq r_j, \\ \frac{e^2}{9\times0.28L_M}, & r_i = r_j, \end{cases}$$  

(1)

where $-e/3$ is the charge in each lobe, and $\varepsilon$ is the dielectric constant, which we take to equal to $10\varepsilon_0$. Here, $L_M$ is the distance between neighboring $AB$ and $BA$ regions. We use $L_M = 13.4$ nm, corresponding to a twist angle of $\theta = 1.05^\circ$. We also include an exchange interaction, which lowers the energy by $J$ when two overlapping occupied orbitals have the same spin and valley quantum numbers. We use the values for the exchange energies from Ref. [20]. These are dominated by the nearest-neighbor and next-nearest-neighbor terms: $J = 0.376, 0.0645\varepsilon^2/(\varepsilon L_M)$. All other exchange energies are smaller than $0.0014\varepsilon^2/(\varepsilon L_M)$. Terms beyond the fifth-nearest neighbor are ignored.

The Coulomb interaction energy from a two-dimensional (2D) array of charges is linearly divergent with system size. The relative energies of the configurations with the same average density, however, are well defined. Some care must be taken in calculating these energies, since even after subtracting off the leading divergence, the remaining sums are conditionally convergent. The process is regularized by using 2D Ewald sums [24,25]:

$$\sum_{w \in \Omega} \frac{1}{|r - w|} = f_i(r) + f_i(w) + C,$$  

(2)

$$f_i = \frac{2\pi}{\Omega} \sum_{k \in \Omega \setminus 0} \frac{e^{ik\cdot r}}{2k} \text{erfc} \left(\frac{k\eta}{\sqrt{2}}\right),$$  

(3)

$$f_s = \sum_{w \in \Omega} \frac{1}{|r - w|} \text{erfc} \left(\frac{|r - w|}{2\sqrt{2}\eta}\right) - \frac{4\sqrt{\pi}}{\Omega} \eta.$$  

(4)

Here, $\eta$ is an arbitrary parameter which separates the interactions into long-range and short-range parts. The sum is over a Bravais lattice $\Omega$, which in our case is generated by the supercell vectors $v_1, v_2$. The reciprocal lattice is $\bar{\Omega}$, and $\bar{\Omega} \setminus \{0\}$ represents the reciprocal lattice with the origin removed. Here, $C$ is an irrelevant infinite constant, which physically represents the Coulomb energy from a uniform charge distribution. The sums in Eqs. (3) and (4) are absolutely convergent. We chose $\eta$ to give a reasonably fast convergence rate, typically taking it to be $1/3$ of the lattice constant.

In the experimental setup of Ref. [2], a metallic back gate sits roughly 30 nm behind the sample. We supplement our long-range Coulomb calculations by also calculating energies where we include a set of image charges in this layer. The Appendix gives the resulting expressions for the energies. To leading order, these images uniformly shift the energies of all configurations by the same amount (corresponding to the classical capacitance). Configuration-dependent corrections are exponentially small in the ratio of the supercell periodicity to the distance from the back gate, and we find that for the experimental geometry the results for ground state energies are unchanged when we add the images.
Thus we find that at fillings \( \nu = \frac{1}{2}, \frac{1}{3}, \frac{3}{4}, \frac{1}{4} \), all electrons have the same spin and valley quantum numbers. At \( \nu = \frac{3}{4} \), there are not enough states for all electrons to carry the same spin and valley quantum numbers. Instead, two-thirds of the electrons carry one set, while the other one-third carry another set. At \( \nu = \frac{1}{2} \), things are similar, but an equal number of electrons carry each of the two sets of quantum numbers. Larger \( \nu \)'s are simply particle-hole mirrors of these configurations. At the level of our model, the exact quantum numbers do not matter—what matters is just that they follow this pattern.

We find that for \( \nu = \frac{1}{8} \), the occupied modes (all of which have the same spin and valley quantum numbers) form a striped configuration, as shown in Fig. 2. For \( \nu = \frac{1}{4} \), the occupations are uniform, with every Wannier state occupied by a single electron, all of which share the same spin and valley quantum numbers. For \( \nu = \frac{3}{8} \), the electrons with the dominant quantum numbers form a uniform pattern, while the ones with the other quantum number form the pattern from \( \nu = \frac{1}{8} \). For \( \nu = \frac{1}{2} \), the occupations are again uniform, with each Wannier site being doubly occupied.

Our results are confirmed by the experimental observations of Zondiner et al. [4], where they find that at an integer filling, the spin and valley degrees of freedom are not filled equally. Starting with \( \nu = \frac{1}{8} \), only one flavor fills it up. Then at the next integer filling, another additional flavor fills it up, and so on.

Despite the symmetry breaking at \( \nu = \frac{1}{8} \), the charge density is nearly uniform: Each AA site has a charge of \(-e\). The spatial symmetry breaking, however, should be apparent in optical measurements through a birefringence. One may also find that transport measurements are anisotropic.

The energy per particle in each of these four cases is \( E = -1.72, -2.39, 35.5, 49.4e^2/(e\Delta) \). In physical units, this corresponds to \( E = -18.4, -25.6, 381, 530 \) meV per particle. The relatively small difference between the energy per particle for \( \nu = \frac{1}{8} \) and \( \nu = \frac{1}{4} \) occurs because in these spin/valley polarized states, the direct Coulomb interaction is largely canceled by the exchange interactions. At higher fillings, we are adding electrons with different spin/valley quantum numbers, for which there is no such cancellation. These energies should all be understood as relative to a classical uniform charge distribution.

One way to quantify the stability of the striped state at \( \nu = \frac{1}{8} \), is to look at the next-lowest-energy state that we found (Fig. 3). This excited configuration has energy \(-17.7 \) meV per particle, which is only modestly higher than the ground state. The charge density for this pattern is again uniform, even though the pattern of occupied Wannier states is nonuniform. Note, one can create \( \nu = \frac{1}{8} \) configurations whose energies lie between these by alternating these two patterns.

Given the small energy difference, lattice defects, impurities, or other irregularities could play a role in the pattern with those from Fig. 2.
excitation energy would be twice what we found at $\nu = 1/8$, since the charge is twice as large. In fact, adding an electron at $\nu = 1/4$ costs much more than twice the $\nu = 1/8$ value. Given that the charge density is very same in both cases, this difference can be attributed to the exchange interaction. The energy to remove an electron is $E_e = 51.2 \text{ meV}$, which is somewhat less than twice the energy of the equivalent excitation at $\nu = 1/8$. The difference can be attributed to the detailed structure of the charge distribution in the Wannier states. The particle-hole excitation energy is $E_{eh} = 21.1 \text{ meV}$. A spin or valley flip excitation costs an energy of $E_s = 17.5 \text{ meV}$.

At $\nu = 3/8$, the ground state contains electrons with two different quantum numbers. Let us denote these by $\alpha$ and $\beta$. The $\alpha$ electrons are spread uniformly, while the $\beta$ electrons form the zigzag pattern in Fig. 2. The lowest-energy electronic excitation corresponds to adding another electron in the $\beta$ state, with an energy of $E_e = -28.1 \text{ meV}$. Clearly, this is just the sum of the values of $E_e$ for the $\nu = 1/8$ and $\nu = 1/4$ configurations. Note that if we add an electron with a different spin/valley, the value would have been $-20.7 \text{ meV}$, which is energetically costlier. The lowest-energy hole excitation comes from removing an electron from the $\beta$ state, which yields $E_h = 33.2 \text{ meV}$. The particle-hole excitation energy is $E_{eh} = 10.2 \text{ meV}$. A spin or valley flip excitation costs an energy of $E_s = 10.1 \text{ meV}$.

At $\nu = 1/2$, any added electron must have different quantum numbers than the rest of the electrons. The required energy is $E_e = -27.6 \text{ meV}$. As expected, this comes out to be twice the value of $E_e$ for $\nu = 1/4$. The energy required to remove an electron is $E_h = 65.0 \text{ meV}$. The particle-hole excitation energy is $E_{eh} = 21.1 \text{ meV}$. A spin or valley flip excitation costs an energy of $E_s = 17.5 \text{ meV}$. We note that the value of $E_s$ is the same for $\nu = 1/8$ and $\nu = 3/8$ (equal to 10.1 $\text{ meV}$), and for $\nu = 1/4$ and $\nu = 1/2$ (equal to 17.5 $\text{ meV}$).

Much of the structure in the excitation energies is related to the exchange interaction: The direct Coulomb energy (in units of $e^4/\epsilon a_0^2$) for adding a particle to the ground states corresponding to $1/8, 1/4, 3/8, 1/2$ fillings are 0.642, 1.284, 1.926, 2.568, which are in the ratio $1:2:3:4$. This scaling arises because the charge distribution is uniform in all cases.

V. SUMMARY AND OUTLOOK

To summarize, we have calculated the charge distributions of the correlated insulators at all integer fillings of magic angle twisted bilayer graphene. We have also estimated all possible excitation gaps over the insulating ground states. In all these computations, we have neglected the kinetic terms, as the effective tight-binding models show that the strength of the hopping terms is much less than the Coulomb interactions, and will only have a perturbative effect. One essential caveat is that because the Coulomb scale is so large, it likely leads to mixing between the flat bands and the spectator bands. Nonetheless we are able to explain much of the observed physics, such as the progression of fillings seen by Zondiner et al. [4].

It would be rewarding to apply similar techniques to characterize the correlated insulating phases of transition metal dichalcogenide (TMD) homobilayers and heterobilayers [26–30], as they have much simpler moiré band structures.
Moreover, in certain TMDs such as WSe$_2$, the flat bands and the resulting correlated states are found to exist over a continuum of angles [31] rather than a narrow range around some “magic angles.”

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APPENDIX: INTERACTIONS WITH IMAGE CHARGES

We model screening by a back gate sitting a distance $d/2$ from the sample by placing equal and opposite image charges at a distance $d$. We treat the charge distribution of each Wannier state as three delta functions, each of charge $q = e/3$. The Coulomb interaction between a single charge in one plane, and a Bravais lattice of charges in another is

$$V = \lambda \sum_{w \in \mathcal{L}} \frac{1}{\sqrt{|r - w|^2 + d^2}},$$  \hspace{1cm} (A1)

where $\lambda = q^2/(4\pi \epsilon)$, $w$ is a 2D lattice vector, and $r$ is the 2D location of the charge in the device. By Fourier transforming the potential, one can write

$$V = \lambda C + \frac{2\pi \lambda d}{\Omega} + \frac{2\pi \lambda}{\Omega} \sum_{q \in \mathbb{Z} \setminus 0} e^{-qd} |q| e^{iqr},$$ \hspace{1cm} (A2)

where the first term contains the same infinite constant as Eq. (2). The second term is the potential from a uniformly charged plane, with $\Omega$ being the area of the unit cell. The third term falls off exponentially with the separation. There, $\mathcal{L}$ is the 2D reciprocal lattice. Due to this exponential cutoff, the image charges play a very minor role in the energetics of different periodic charge configurations.

We follow the procedure in Lee and Cai [25] to write $V = V_s + V_l$ with

$$V_s = \sum_{w \in \mathcal{L}} \frac{\text{erfc} \left( \frac{\sqrt{|r - w|^2 + d^2}}{2\eta} \right)}{\sqrt{|r - w|^2 + d^2}} + \frac{2\pi d \text{erfc} \left( \frac{d}{2\eta} \right) - 4\sqrt{\pi} \eta e^{-\frac{d^2}{4\eta}}}{\Omega},$$

$$V_l = \frac{2\sqrt{\pi}}{\Omega} \sum_{k \in \mathcal{L} \setminus 0} \left[ e^{-k\delta} - e^{-k\delta} \text{erfc} \left( \frac{\delta}{2\eta} - k \eta \right) + e^{k\delta} \text{erfc} \left( \frac{\delta}{2\eta} + k \eta \right) \right].$$ \hspace{1cm} (A3)

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