Lattice Theory of Pseudospin Ferromagnetism in Bilayer Graphene: Competing Orders and Interaction Induced Quantum Hall States

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In mean-field-theory bilayer graphene’s massive Dirac fermion model has a family of broken inversion symmetry ground states with charge gaps and flavor dependent spontaneous inter layer charge transfers. We use a lattice Hartree-Fock model to explore some of the physics which controls whether or not this type of broken symmetry state, which can be viewed as a pseudospin ferromagnet, occurs in nature. We find that inversion symmetry is still broken in the lattice model and estimate that transferred areal densities are \( \sim 10^{-5} \) electrons per carbon atom, that the associated energy gaps are \( \sim 10^{-2} \) eV, that the ordering condensation energies are \( \sim 10^{-7} \) eV per carbon atom, and that the energy differences between competing orders at the neutrality point are \( \sim 10^{-9} \) eV per carbon atom. We explore the quantum phase transitions induced by external magnetic fields and by externally controlled electric potential differences between the layers. We find, in particular, that in an external magnetic field coupling to spontaneous orbital moments favors broken time-reversal-symmetry states that have spontaneous quantized anomalous Hall effects. Our theory predicts a non monotonic behavior of the band gap at neutrality as a function of interlayer potential difference in qualitative agreement with recent experiments.

I. INTRODUCTION

Recent experimental progress\(^1\) in isolating and measuring the electronic properties of graphene single and multilayers has opened a new topic in two-dimensional electron system (2DES) physics\(^2\). Electronic wavefunctions in graphene systems are often described using a pseudospin language in which the spinors specify wavefunction components on different sublattices. Although the properties of graphene 2DES’s can often be successfully described using an effective non-interacting electron model, studies of electron-electron interactions effects have revealed some qualitative differences compared to ordinary 2DES’s that are related to these sub-lattice pseudospin degrees-of-freedom.

In the case of AB stacked bilayer graphene, there are four C sites and four \( \pi \)-orbitals per unit cell, but two of these are repelled from the neutral system Fermi level by interlayer hopping. This circumstance leads to a low-energy massive chiral fermion model\(^3\), without two-component spinors, and a crystal-momentum \( \vec{p} \)-dependent pseudo-magnetic field with a magnitude that varies as \( p^2 \) and an orientation angle twice as large as the momentum orientation angle \( \phi_p \). Neutral system states have one occupied pseudospin for each distinct set of momentum, spin, and valley labels. Recently\(^4\) Min et al. pointed out that when Coulombic electron-electron interactions are added to the massive chiral fermion model, the mean-field-theory ground state pseudospins of each spin-valley flavor break symmetry by rotating out of the \( x-y \) plane, developing \( \hat{z} \) components with a common spontaneously chosen sign and magnitudes which are larger at small \( p \). Because the two sublattices from which the two-band-model pseudospins are constructed are located in opposite layers, the broken symmetry transfers charge between layers. It is therefore characterized in momentum space by a vortex with vorticity \( v = 2 \) and a flavor-dependent core polarized along one of the polar directions, and in real space by flavor-dependent uniform layer polarization. Different members of the family of states are distinguished by the spin-valley flavor dependence of the sense of layer pseudospin orientation in the momentum-space vortex cores. The origin of the broken symmetry, which we refer to here as pseudospin ferromagnetism, is the \( p^2 \) pseudospin-splitting at small \( p \), which leads to infrared divergences in particle-hole polarization loops, combined with the frustrating effect of pseudospin chirality which leads to relatively stronger exchange interactions for \( \hat{z} \)-polarized pseudospins.

The broken symmetry states can be classified\(^9,10\) in a manner which highlights their valley and spin dependent momentum space Berry curvatures, which are in turn closely related\(^11\) to the Hall responses of the system. Indeed, recent experiments\(^11\) demonstrate that bilayer graphene exhibits a quantized quantum Hall effect in the absence of an external magnetic field. Over and above the basic science interest deriving from this presently unique property, it is possible that pseudospin order in bilayer graphene could play a role in graphene electronics\(^12\) by introducing hysteresis and dramatically enhancing the influence of gates on conductance.

In this article we report on a study of pseudospin ferromagnetism in a \( \pi \)-orbital tight-binding model for bilayer graphene, which is conveniently able to capture and establish the role of some bilayer graphene \( \pi \)-band features neglected in the massive chiral fermion model. We estimate that the density shift for each flavor is \( \sim 10^{-5} \) electrons per carbon atom, that the gaps are \( \sim 10^{-2} \) eV, that the total condensation energy is \( \sim 10^{-7} \) eV per carbon atom, and that the energy differences between competing ordered states is \( \sim 10^{-9} \) eV per carbon atom. The energy differences between competing ordered states, which are between one to two orders or magnitude smaller than the condensation energy, are sensitive to roughly estimated lattice scale details of the model we employ. The competing states are classified in the first place as either anomalous Hall states, in which opposite valleys are polarized toward opposite layers, or valley Hall states in which they are polarized toward the same layer. We find that valley Hall states have slightly lower energies. Because anom-
lous Hall states have spontaneous orbital magnetism they are favored by external magnetic fields. We estimate that, because the energy differences between states are very small, extremely weak fields are sufficient to induce phase transitions between valley Hall and anomalous Hall states. On the other hand potential differences between layers favor valley Hall states. When the spin degree of freedom is accounted for we find that the spontaneous energy gap in neutral bilayers first decreases and then increases with potential difference, in qualitative agreement with experiment.

In the following two sections we explain the model Hamiltonian we employ and some of the technical details of the calculations we have carried out. The main results are presented in the Section IV where we introduce the topological classification of solutions and use it to discuss the properties of broken symmetry solutions for vanishing, moderate, and strong externally controlled electric potential differences between the layers. (Below we refer to interlayer potential differences, which always play a key role in graphene bilayer physics, as potential biases.) We also discuss coupling to an external magnetic field which can drive transitions between competing states, some of which have broken time-reversal symmetry with associated anomalous Hall effects and orbital magnetism.

We close the paper with a summary and some suggestions for further research.

II. FOUR-BAND $\pi$-ORBITAL TIGHT-BINDING MODEL OF BILAYER GRAPHENE

We describe bilayer graphene using a lattice model with one atomic $2p_z$ orbital per carbon site. We write the model’s Bloch basis states in the form

$$\psi_{k\kappa} (r) = \frac{1}{\sqrt{N}} \sum_{\tau} e^{i(r \cdot R_\tau - \tau \cdot \kappa)} \phi (r - R_\tau - \tau \kappa),$$

where $N$ is the total number of unit cells in the system, $\phi (r)$ is the band’s Wannier wavefunction, and $\kappa$ labels the carbon site with position $\tau \kappa$ relative to the triangular lattice vector $R_\tau$. (We comment later on the possible role of screening effects from the $p$ and $s$ orbitals which form the $\sigma$ and $\sigma^*$ bonds neglected in this model.) Following the convention used in Refs.[6,15], we use the notations $A$, $B$, $\bar{A}$, $\bar{B}$ for the four sublattice indexes $\kappa$, where $B$ and $\bar{A}$ are the opposite-layer near-neighbor-pair sites. With this convention, the four band tight-binding model Hamiltonian of a graphene bilayer is:

$$H_0 = \begin{pmatrix} 0 & \gamma_0 f & \gamma_0 f & \gamma_0 f \\
\gamma_0 f & 0 & \gamma_0 f & \gamma_0 f \\
\gamma_0 f & \gamma_0 f & 0 & \gamma_0 f \\
\gamma_0 f & \gamma_0 f & \gamma_0 f & 0 \end{pmatrix},$$

with $a = 2.46\AA$ arises from a sum over the three near-neighbor hops within a layer. We have neglected differences in on-site energies and next nearest neighbor hopping processes which give rise to electron-hole asymmetry and do not play an important role in pseudospin ferromagnetism. The tight-binding model parameters $\gamma_i$ should not be confused with the Slonczewski-Weiss, McClure[16] model parameters for bulk graphite, despite the obvious similarities in notation. In our calculations we adopt conventions similar to those of Ref.[17] for bilayer graphene, taking the values $\gamma_0 = -3.12 \text{eV}$, $\gamma_1 = -0.377$, $\gamma_2 = -0.29 \text{eV}$ and $\gamma_4 = -0.12 \text{eV}$ for the hopping parameters. Only the intralayer nearest neighbor ($\gamma_0$) process and interlayer tunneling ($\gamma_1$) process are retained in the minimal tight-binding model. The trigonal warping ($\gamma_2$) process which connects the $A$ and $B$ sites is responsible for the leading circular symmetry breaking near the valley points, while the ($\gamma_4$) process which connects $A$ and $\bar{A}$ sites influences the intralayer charge imbalance between sublattices $A$ and $B$.

III. LATTICE MODEL MEAN-FIELD THEORY

Because of the importance of non-local exchange in graphene systems, a Hartree-Fock mean-field theory approximation[15] is a natural first step in considering electron-electron interaction effects. When Coulomb interactions are added to the $\pi$-band tight-binding model the interaction terms in the mean-field Hamiltonian take the form:

$$V_{\text{HF}} = \sum_{k\lambda k'} U_{k\lambda k'}^{X} N_{\lambda} c_{k\lambda}^\dagger c_{k'\lambda} - W_{k\lambda k' \lambda}^{X} c_{k\lambda}^\dagger c_{k'\lambda} c_{k'\lambda'} c_{k'\lambda'},$$

where $\lambda$ is a composite label for sublattice $\kappa$ and spin $\sigma$. The first term on the right hand side of Eq. (4) is the Hartree term:

$$N_{\lambda} = N_{\kappa \sigma} = \sum_k \langle c_{k\lambda}^\dagger c_{k\lambda} \rangle = \sum_{k'} n_{k' \lambda}$$

$$U_{k\lambda k'}^{X} = \frac{\delta_{\sigma,\sigma'}}{A} \sum_{G} \exp [ \mu \langle G (\tau \kappa - \tau \kappa') \rangle] \langle |G| \rangle^2 V_{\kappa \lambda \kappa'}^{X} (|G|),$$

where $G$ is a reciprocal lattice vector. The second is the Fock (exchange) term:

$$W_{k\lambda k' \lambda}^{X} = \sum_k U_{k\lambda k'}^{X} (k' - k) \langle c_{k'\lambda}^\dagger c_{k\lambda} \rangle$$

$$U_{k\lambda k'}^{X} (q) = \frac{1}{A} \sum_{G} \exp [ \mu \langle G (\tau \kappa - \tau \kappa') \rangle] \times \langle |G - q| \rangle^2 V_{\kappa \lambda \kappa'}^{X} (|q - G|).$$

In Eq.(6) and Eq.(8), the two-dimensional Coulomb interaction $V_{\kappa \lambda \kappa'}^{X} (q) = 2\pi e^2 / (|q| \varepsilon_C)$ when $\kappa$ and $\kappa'$ refer to the same layer and $(2\pi e^2 / (|q| \varepsilon_C)) \exp [- |q| \varepsilon_C]$ when $\kappa$ and $\kappa'$ refer to the opposite layers. Here $\varepsilon_C$ is the relative dielectric constant, $\varepsilon_C = 3.35\AA$ is the interlayer separation, $A$ is the total area of the graphene sheet, and we use

$$\tilde{f} (q) = (1 - (r_0 q)^2) / ((1 + (r_0 q)^2)^4)$$

$$f (q) = 1 + 2e^{-i k a / 2\sqrt{3}} \cos \left( \frac{k a}{2} \right)$$
as a form factor which accounts for the spread of the π-orbital charge on each site. This simple form assumes an isotropic site-localized charge distribution. Eq. (9) was obtained by Fourier transforming the radial charge distribution of a hydrogenic 2p orbital. The use of \( r_0 = a_0 = a_0/\sqrt{30} \) would yield a root mean square radius corresponding to the covalent radius of the carbon atom \( a_0 = 0.77\text{Å} \). If we consider screening from the σ band electrons neglected in our model and the fact that the charge density distribution of a p_z orbital is far from spherical we expect that larger values of \( r_0 \), which effectively reduce onsite repulsion, would be more appropriate. For most of our calculations we have therefore used the value \( r_0 = 3a_0 \).

As explained in the introduction, pseudospin ferromagnetism in bilayer graphene can be neatly described using the two-band massive chiral fermion model. This approach has two shortcomings which the present calculation is intended to alleviate. First of all, the model has to rely on a crude ultraviolet-cutoff to account for the limited range of energy \( \sim \gamma_1 \) over which it is applicable. At moderate interaction strengths the amount of charge transferred between layers determined in the massive chiral fermion model calculation is strongly influenced by this cut-off. Secondly, the calculation described in Ref. (7) relies on the model’s circular symmetry for a number of simplifications. When direct hopping between the low-energy A and B sites, the \( \gamma_1 \) process, is included in the Hamiltonian the model’s Fermi lines are no-longer circular and the continuum model loses some of its attractive simplicity. These processes are known to be essential at very weak interaction strengths since they remove the infrared divergences responsible for the instabilities of the massive chiral fermion model. The present lattice model reduces to the continuum model at low energies, accounts naturally for the limited validity range of two-band models by retaining all four π-bands, and can deal with the loss of circular symmetry without any additional complication.

The main challenge which arises in practical implementation of the lattice model mean-field-theory lie in the numerical optimization of a problem in which the small portion of the Brillouin-zone close to one of the valley points plays a dominating role and must be sampled densely. It is essential that we have sufficiently dense k-point sampling near the Dirac points, and at the same time sample the full Brillouin zone. The increase of computational load with k-point sampling density is particularly rapid in the Hartree-Fock calculations because the Hartree-Fock matrix element at one k-point depends on the occupied wavefunctions at all other k-points. In Fig. 1 we illustrate the honeycomb lattice reciprocal space primitive cell and the k-point sampling scheme we have chosen. In an effort to achieve a satisfactory compromise between computational load and accuracy we, first of all, make use of the hexagonal symmetry inherent in the problem. This allows us to limit our calculations to 1/6th of the total Brillouin zone area when we distinguish K and K' valleys, and 1/12th of the total area when we do not. In addition, instead of using a uniform grid in the whole Brillouin zone we use a k-point mesh which is denser near the Dirac point. In our calculations we have used 18 \times 18, 32 \times 32 or 42 \times 42 coarse grids for the full primitive cell and multiplication factors of up to 128 for the finer k-point mesh near the valley centers. The fine mesh densities were either 2304 \times 2304 or 2048 \times 2048 for typical calculations at zero potential bias, and 672 \times 672 when smaller densities were enough to converge the calculations in the case of strongly biased bilayers. Dense k-point meshes were normally employed within \( \sim 0.5/a \) of a Dirac point, and wider regions were used for strong bias cases. (Here \( a = 2.46\text{Å} \) is the lattice constant of the triangular periodic lattice structure of graphene.) Nevertheless k-point sampling approximations remain the main source of numerical inaccuracies. The self-consistent-field calculations were iterated until convergence to 14 significant figures in the total energy per electron was achieved for a given choice of k-point sampling. In order to resolve the small energy differences between solutions of the self-consistent field equations corresponding to different states we needed to compare results obtained with the same k-point sampling scheme. The k-point sampling we used achieved convergence to within a few parts per thousand for charge density differences and band gaps.

IV. LATTICE MODEL PSEUDOSPIN FERROMAGNET

The inversion symmetry breaking instability in bilayer graphene occurs nearly independently at the two points and, within mean-field-theory, entirely independently for each electron spin. It is strongest in electrically neutral
Initial studies carried out at the Hartree-Fock level \cite{8, 15, 21, 23} identified a family of competing pseudospin ferromagnet states. Perturbative renormalization group calculations \cite{8, 21, 22} have confirmed that these instabilities can survive beyond mean-field-theory.

The 16 states can be classified into two basic types of overall layer polarization. By this classification are included the layer ferrimagnetic states in which three of the four flavors have overall polarization $\mathbf{A}$, and values $\mathbf{377eV}$, $\gamma = \gamma_0 = 0$, and dielectric screening parameter $\varepsilon_r = 4$. Right panel: Ongoing k-dependent exchange potentials of the spontaneously broken symmetry state on the four bilayer sublattices. The onsite potential is larger in magnitude on the low-energy sites that do not have an opposite layer neighbor, and lower on average in the layer with the larger density.

A. Total layer density and Chern number classifications of competing states

Because each bilayer flavor can polarize toward either of the two layers, there are a total number of $2^4 = 16$ possible configurations of the broken symmetry state. The layer polarization for each spin and valley determines the sign of the mass term in its continuum model and has implications for the topological properties in the system as we will now discuss. The 16 states can be classified by overall layer polarization as being ferromagnetic, ferrimagnetic, or layer antiferromagnetic. By this classification there are two layer ferromagnetic states in which all flavors choose the same polarization, eight layer ferrimagnetic states in which three of the four flavors choose the same polarization, and six layer antiferromagnetic states with no overall polarization. The layer antiferromagnetic states are electrostatically favored in the absence of a potential bias.
TABLE I: Summary of Hall transport properties for valley $\tau_c = K, K'$ and spin $\sigma = \uparrow, \downarrow$, dependent layer polarization $\lambda_c = T, B$. These states are classified as layer ferromagnetic (F), layer ferrimagnetic (Fi) or layer antiferromagnetic (AF) following reference \[7,10\]. F, Fi, and AF states have respectively four, three, and two valley-spin components polarized toward the same layer ($\lambda_c = T$). We have listed only eight out of a total of sixteen configurations omitting the equivalent configurations obtained by layer reversal for every flavor. Each valley contributes a finite Hall conductivity with magnitude $\frac{\sigma_{xy}}{n}$ and a sign that reverses with both valley interchange and layer polarization. The Hall conductivities are assigned to particular valleys on the basis of approximate Chern indices obtained by integrating Berry curvatures over the portion of the Brillouin zone near $K$ or $K'$. The edge-state structure is expected to depend on each of the partial Hall conductivities. The total Hall conductivity is separated into contributions from separate spins and separate valleys using $\sigma_{xy}^\sigma = \sigma_{xy}^{K\uparrow} + \sigma_{xy}^{K\downarrow} = \sigma_{xy}^{T\uparrow} + \sigma_{xy}^{T\downarrow}$. The F configuration have zero total Hall conductivity, whereas all Fi configurations have a finite total anomalous Hall conductivity of two units. The A configurations are expected to be electrostatically favored in the absence of an external layer bias potential and include three types of solutions with different Hall properties, one with a total anomalous total Hall conductivity, one with zero Hall conductivity but finite spin Hall conductivity, and another with zero Hall and spin Hall conductivities because of opposite Hall conductivity contributions from $K$ and $K'$ valleys for both spins.

|          | $\lambda_c$ | $\tau_c$ | $\sigma_\tau$ | $\sigma_{xy}$ | $\sigma_{xy}^{K\uparrow}$ | $\sigma_{xy}^{K\downarrow}$ | $\sigma_{xy}^{T\uparrow}$ | $\sigma_{xy}^{T\downarrow}$ |
|----------|-------------|-----------|---------------|---------------|----------------------------|----------------------------|---------------------------|---------------------------|
| **F**    | (T $\uparrow$) | (T $K'$ $\uparrow$) | (T $K$ $\downarrow$) | (T $K'$ $\downarrow$) | 1 | -1 | 1 | -1 | 0 | 0 | 2 | -2 |
| **Fi**   | (T $\uparrow$) | (T $K'$ $\uparrow$) | (T $K$ $\downarrow$) | (B $K'$ $\downarrow$) | 1 | -1 | 1 | -1 | 0 | 2 | 0 | 2 |
|          | (T $\uparrow$) | (T $K'$ $\uparrow$) | (B $K'$ $\downarrow$) | (T $K$ $\downarrow$) | 1 | 1 | -1 | -1 | -2 | 0 | -2 | -2 |
|          | (T $K'$ $\uparrow$) | (B $K'$ $\downarrow$) | (T $K$ $\downarrow$) | (T $K'$ $\downarrow$) | -1 | -1 | 1 | -1 | 2 | 2 | 0 | 0 |
| **AF**   | (T $\uparrow$) | (B $K'$ $\uparrow$) | (T $K$ $\downarrow$) | (B $K'$ $\downarrow$) | 1 | 1 | 1 | 1 | 2 | 2 | 2 | 4 |
|          | (T $\uparrow$) | (B $K'$ $\uparrow$) | (T $K'$ $\downarrow$) | (B $K$ $\downarrow$) | 1 | 1 | -1 | -1 | 2 | -2 | 0 | 0 |
|          | (T $\uparrow$) | (T $K'$ $\uparrow$) | (B $K$ $\downarrow$) | (B $K'$ $\downarrow$) | 1 | -1 | -1 | 1 | 0 | 0 | 0 | 0 |

vortices\[9,10,27\] in the broken symmetry states. Because the vorticity $\nu$ is opposite for opposite valleys, the integrated Berry curvature gives rise to a Hall conductivity $11,12,28,30$ with magnitude $e^2/h$ for each flavor, and a sign that changes with valley as well as with layer polarization. The Berry curvature reflects the handness of Bloch electrons and captures intra-cell circulating currents which generate a finite orbital magnetic moment proportional to the angular momentum due to self-rotating Bloch wave packets.\[29\] The Berry curvature of the system can be evaluated using\[11\]

$$\Omega_{n}(k) = \sum_{n' \neq n} \left[ \frac{(u_{n'} | \frac{\partial}{\partial k_x} | u_{n'}) (u_{n'} | \frac{\partial}{\partial k_y} | u_{n'})}{(E_{n'} - E_n)^2} - c.c. \right].$$

where $|u_{n'}\rangle$ represents the Bloch eigenstates of the system and $E_n$ are the associated eigenvalues for each $k$. The finite orbital moment generated by these wave packets has a similar expression and can be evaluated through\[11\]

$$m_n(k) = -\frac{\hbar}{2m} \sum_{n' \neq n} \left[ |u_{n'}| \frac{\partial}{\partial k_x} |u_{n'}\rangle \langle u_{n'}| \frac{\partial}{\partial k_y} |u_{n'}\rangle - c.c. \right] = \hbar \sum_{n' \neq n} \left[ |u_{n'}| \frac{\partial}{\partial k_x} |u_{n'}\rangle \langle u_{n'}| \frac{\partial}{\partial k_y} |u_{n'}\rangle - c.c. \right].$$

A weak external magnetic field will tend to favor a state in which the orbital magnetizations of all valleys are aligned, and the anomalous Hall conductivity is correspondingly maximized. This observation suggests the possibility of valley optoelectronics that exploits the circular dichroism of interband transitions\[30\] in the broken symmetry states. The Kerr and Faraday effect measurements with linearly polarized light can be a useful tool to detect signatures of broken time reversal symmetry.\[44\] In Fig. 3 we present the $k$-dependent magnetization evaluated for one self-consistent gapped state in the presence of an interlayer bias. Similar results have been obtained previously using the massive Dirac-fermion continuum model. An estimate of the zero field magnetization per valley-spin degree of freedom $M_{c,\sigma} = \sum_k |m_n(k)| \frac{d h}{2\pi} \approx 2\pi \sum_k |m_n(k)| k d h \frac{d h}{2\pi} \frac{d k}{d h}$ can be obtained integrating the orbital moment around each one of the valleys for a given spin component. For the broken symmetry state with $\mathcal{E}_c = 4$ each component integrates to $M_{c,\sigma} = 10^{-3} \mu_B$ per carbon atom, compared to $M_{c,\sigma} = 1.4 \cdot 10^{-3} \mu_B$ for the non-interacting system with bias $\mathcal{E} = 0.1 V/nm$ and $M_{c,\sigma} = 2.2 \cdot 10^{-3} \mu_B$ per carbon atom for $\mathcal{E} = 0.4 V/nm$. The individual flavor orbital magnetizations and anomalous Hall contributions cancel in states that do not have broken time-reversal symmetry.

In the non-interacting electron biased bilayer graphene state, opposite contributions from the two valleys lead to vanishing total Hall conductivity, consistent with the absence of broken time-reversal symmetry. The nature of the layer polarized broken symmetry we describe here is analogous to the biased bilayer in the sense that the charge transfer can be attributed to the $(k$-point dependent) exchange potential difference between low-energy sites on opposite layers, as illustrated in Fig. 2. Because of the non-locality of the exchange interaction, however, we are able to find self-consistent solutions in which the effective interlayer bias potential has opposite signs in the two valleys, and the total Hall conductivity is finite. In Table 1 we present a list of the 16 different configurations for which we have found self-consistent solutions, which are characterized by the sense of layer ($\lambda_c = T, B$) po-
Hall conductivity, in which electrons are polarized towards the anomalous Hall (AH) state, with four quantized units of energy penalty. As summarized in Table I, there are three dis-similarities with those for non-interacting electrons with an external interlayer bias.

We compare results for the self-consistent broken symmetry states with those for non-interacting electrons with an external interlayer bias.

B. Broken inversion symmetry states in an unbiased bilayer

We start by examining the layer antiferromagnetic charge balanced configurations which are lowest in energy in the absence of an external bias because of the absence of a Hartree energy penalty. As summarized in Table I, there are three distinct types of layer antiferromagnets. One configuration is the anomalous Hall (AH) state, with four quantized units of Hall conductivity, in which electrons are polarized towards the same layer for both spins, and towards opposite layers for opposite valleys. Second is the spin Hall (SH) state which has opposite layer polarization on opposite valleys, and in each valley opposite layer polarization for opposite spin. The mean-field Hamiltonian for this state is similar to that of a non-interacting system with intrinsic spin orbit coupling. Finally there is a solution with the same layer polarization in the two valleys, but opposite layer polarizations for opposite spins. This state has a finite valley Hall (VH) conductivity for each spin, zero valley Hall conductivity when summed over spins, and zero total Hall conductivity. Optical measurements based on linearly polarized light using the Kerr and Faraday effect can be used to identify the anomalous Hall state which has broken time reversal symmetry state.

In the continuum model formulation the three distinct solutions are degenerate. In the lattice model Hartree-Fock theory the energies of the AH and SH states are still exactly degenerate because the exchange energy is spin diagonal and the energy within each spin is independent of the Hall effect sign. We refer to these two states collectively as the anomalous Hall states. The valley Hall (VH) solution does have a different energy however, because the relative sense of layer polarization in the two valleys influences inter-valley exchange potentials. Table II presents our results for the condensation energy of the broken symmetry state and for the differences in energy between the VH and AH/SH solutions separated into different contributions. We first note that the condensation energies are reasonably independent of the model parameters $\epsilon_F$ and $r_0$, which are not precisely known and dependent on the sample’s dielectric environment. The scale of the condensation energy should be compared with the value of the Coulomb interaction at the momentum scale $k^*$ ($e^2/k^*$) over which the two-band model applies to bilayer graphene. We find $k^*$ by setting the low-energy model parabolic band energy equal to the isolated layer energy, which gives $k^* \sim \gamma_1 / \hbar v$. The Coulomb energy at this momentum scale is $(\epsilon_g^2/\alpha_g r_0) \times \gamma_1 = \alpha_g x \gamma_1 \sim 100$ meV. This energy scale gives an estimate of the size of the gap which would be expected if all states with $k < k^*$ were fully layer polarized. We find a gap which is approximately four times smaller and density shifts between layers that are smaller than $\pi k^*/(2\pi)^2 \sim (\gamma_1/2\pi \alpha_g)^2$ by a similar fraction. These numerical values indicate that the broken symmetry arises mainly from those bilayer band states that are reasonably well described by the low-energy two-band models used in the original mean-field calculations, as expected. We note that the condensation energy is smaller by several orders of magnitude than the total interaction energy, which involves electrons across the full $\pi$-band.

The second block of columns present differences in energy between VH states and AH/SH states. For the parameters we have considered the total energies for the VH states are lower than the AH/SH thanks to the exchange energy advantage of the former states. The differences in energy between these states become substantially smaller than the condensation energy when the carbon radius model parameter $r_0$ is increased. Larger values of $r_0$ improve the accuracy of the continuum model and differences in energy between the different solutions rapidly decrease. We have previously argued that values of $r_0 \sim 3 \bar{a}_0$ in the form factor are appropriate. If so, the differences in energy between different solutions are $\sim 10^{-9}$ eV per carbon atom, about 100 times smaller than the ordering condensation energy. We have also explored an alternative formulation of the Hartree-Fock equations which evaluates the non-local exchange potentials in real space. This formulation allows the model’s onsite repulsion $U$ to be adjusted separately from the longer ranged tails and thus allow a more direct assessment of the impact of lattice scale details of the effective Coulomb interaction, and leads to similar conclusions.
TABLE II: Energy differences between distinct mean-field solutions in the absence of a potential bias. The first group of columns represent energy differences between the broken symmetry pseudospin ferromagnet states in the anomalous Hall (AH) configuration and the unbroken symmetry state that has no gap. The latter has been obtained constraining the self-consistent HF calculation to preserve inversion symmetry. The third column represents the exchange energy difference $\Delta E_X$ of the broken symmetry self-consistent state with respect to the reference exchange energy of the non-interacting state. The remaining columns represent energy differences between between anomalous Hall and valley Hall (VH) solutions. The calculated energy differences depend sensitively on the choice of the model parameter $r_0$ used in the form factor. The total exchange energy differences $\Delta E^{tot} = 4(\Delta E^{KK}_X + \Delta E^{KK}_X)$ can be expressed as a sum of intravalley $(KK)$ and intervalley $(KK')$ contributions, where the four-fold factor is due to the twofold degeneracy in both spin and valley. We have verified that the energy differences depend very weakly on the $\gamma_1$ and $\gamma_2$ parameters which are excluded in the minimal model. The two anomalous Hall states have the same energy in mean-field theory as explained in the text. The values compiled in this Table have been evaluated using $\varepsilon_r = 4$ and the indicated carbon atom radii $r_0$. All energies are expressed in eV per carbon atom unit.

| $r_0$ | $\Delta E^{tot}$ | $\Delta E_X$ | $\Delta E^{TB}$ | $\Delta E^{tot}$ | $\Delta E_X$ | $\Delta E^{KK}_X$ | $\Delta E^{KK}_X$ |
|-------|------------------|---------------|------------------|------------------|---------------|------------------|------------------|
| $a_0$ | $-4.84 \cdot 10^{-8}$ | $-1.13 \cdot 10^{-7}$ | $-5.10 \cdot 10^{-4}$ | $-1.03 \cdot 10^{-7}$ | $-9.72 \cdot 10^{-7}$ | $-1.75 \cdot 10^{-7}$ | $-6.75 \cdot 10^{-8}$ |
| $2a_0$ | $-5.17 \cdot 10^{-8}$ | $-1.24 \cdot 10^{-7}$ | $-6.15 \cdot 10^{-4}$ | $-4.66 \cdot 10^{-9}$ | $-1.92 \cdot 10^{-9}$ | $-4.09 \cdot 10^{-9}$ | $-6.97 \cdot 10^{-10}$ |
| $3a_0$ | $-6.17 \cdot 10^{-8}$ | $-1.58 \cdot 10^{-7}$ | $-5.09 \cdot 10^{-4}$ | $-1.14 \cdot 10^{-9}$ | $-2.76 \cdot 10^{-9}$ | $-5.71 \cdot 10^{-10}$ | $-1.18 \cdot 10^{-10}$ |

TABLE III: Transferred charge per valley-spin flavor in the charge balanced anomalous Hall and valley Hall solutions. In our mean field Hartree-Fock calculations the VH solutions have slightly larger band gaps and interlayer charge transfers than their anomalous Hall counterparts. In this table, charge densities are in units of $10^{11}$ cm$^{-2}$ and band gaps in meV units. We have used the tight-binding model parameters $\gamma_0 = -3.12$ eV, $\gamma_1 = 0.377$ eV, $\gamma_2 = 0.20$ eV, and $\gamma_3 = -0.32$ eV. The $\gamma_1$ parameter has only a marginal influence on the broken symmetry state. The value chosen for $\gamma_1$ term tends to accumulate extra electrons at the low energy sites in the bilayer but does not influence the strength of the broken symmetry. The results reported here were obtained using dielectric constant $\varepsilon_r = 4$ and carbon atom radius $r_0 = 3a_0$. In the rightmost set of columns we show the results obtained when a smaller carbon atom radius $r_0 = 2a_0$ is used in the form factor calculation in Eq. (9).

Anomalous Hall / Spin Hall (AH/SH)

| $\varepsilon_r$ | $\Delta n_t$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta \gamma_{gap}$ |
|----------------|-------------|----------------|----------------|----------------|-------------|----------------|----------------|----------------|-------------|----------------|----------------|----------------|-------------|----------------|
| 4              | 0.52        | 0.38           | 0.28           | 0.31           | 0.52        | 5.67           | 4.59           | 4.07           | 31           | 0.72           | 0.25           | 0.31           | 0.47         | 31           |
| 5              | 0.42        | 0.60           | 0.18           | 0.22           | 0.41        | 5.38           | 4.97           | 4.61           | 21           | 0.54           | 0.16           | 0.22           | 0.38         | 22           |
| 6              | 0.34        | 0.46           | 0.12           | 0.16           | 0.34        | 5.20           | 4.86           | 4.63           | 29           | 0.42           | 0.10           | 0.16           | 0.32         | 16           |

Valley Hall (VH)

| $\varepsilon_r$ | $\Delta n_t$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta n_t^a$ | $\Delta n_t^b$ | $\Delta n_t^c$ | $\Delta n_t^l$ | $\Delta \gamma_{gap}$ |
|----------------|-------------|----------------|----------------|----------------|-------------|----------------|----------------|----------------|-------------|----------------|----------------|----------------|-------------|----------------|
| 4              | 0.52        | 0.38           | 0.32           | 0.33           | 0.52        | 5.70           | 5.18           | 5.55           | 4.03         | 0.89           | 0.40           | 0.32           | 0.49         | 32           |
| 5              | 0.42        | 0.61           | 0.19           | 0.22           | 0.42        | 5.40           | 4.98           | 4.60           | 4.18         | 0.64           | 0.24           | 0.22           | 0.40         | 22           |
| 6              | 0.34        | 0.47           | 0.13           | 0.16           | 0.34        | 5.21           | 4.87           | 4.62           | 4.28         | 0.33           | 0.15           | 0.17           | 0.33         | 17           |

Large values of $U$ imply strong short-range correlations which are not accurately described by the continuum model. From the results shown in table I, however, we see that only moderate short-range screening from degrees-of-freedom outside the $\pi$-band model are required to make deviations from the continuum model small. Given the abundance of separate evidence that interaction effects in graphene systems are accurately described by a continuum model, we assume that the required short-range screening is in fact present.

In Table III we report on the layer distribution of charge when inversion symmetry breaking occurs within a particular valley. The results presented here are single valley-spin results based on the AH and VH solutions. We have performed these calculations for band models which include and exclude tight-binding hopping parameters other than in plane nearest neighbor hopping $\gamma_0$ and interlayer tunneling $\gamma_1$. The trigonal warping $\gamma_1$ term connects the sites $A$ and $B$ and introduces a triangular distortion in the band structure near the Dirac point, hence breaking the approximately circular symmetry of the bands. It also makes the dispersion become linear instead of quadratic at the lowest energy scales and works against symmetry breaking. We notice that the reduction of the band gap due to trigonal warping varies between two per cent and ten per cent depending on the short-range interaction strength, in-
indicating that trigonal warping plays a relatively minor role. The $\gamma_4$ term increases the accumulation of charge density at lattice sites $A$ and $B$ relative to that at the high energy sites $B$ and $B$. The gap size and total transferred charge ($\Delta n_l$) remain virtually unchanged, although the distribution of charge between sublattices on the same layer ($\Delta n_i^L$) is altered in some states.

The temperature dependence of the band gap and the transferred charge per valley-spin component in mean-field-theory is plotted in Fig. 4. As the temperature is increased both the charge density and band gaps are reduced, but their ratio is approximately fixed. The decay trend is similar for the different dielectric constants we have considered. The critical temperatures obtained in this mean-field calculation provide an estimate of the maximum temperature at which local order survives in the absence of disorder. The band gap decreases more quickly than the charge-density transfer order parameter as the interaction is made weaker; the charge density per valley-spin varies approximately like $\varepsilon_r^{-1}$ whereas the gap varies as $\varepsilon_r^{-1.55}$.

C. Quantum phase transitions in the low magnetic field and low bias regime

To first order in magnetic field, the magnetic contribution to energy is simply proportional to spontaneous magnetization discussed previously, which is entirely orbital in character and depends on the flavor-dependent layer polarizations. States that are related by layer polarization reversal are no longer equal in energy in the presence of a magnetic field. If we take $\Delta E_{tot} \sim 10^{-9} eV$ per carbon atom for the mean field energy differences between AH and VH states from Table II, and use the relation $\Delta E_{tot} \sim M_{\sigma_\uparrow,\sigma_\downarrow} \cdot B$, we obtain from the orbital magnetization values we have calculated that a magnetic field $B_c \sim 0.004T$ is sufficient to favor the AH state with orbital magnetization parallel to the magnetic field over VH states. Considering the small energy differences between the different competing states as shown in Table II we can expect that the coupling between magnetic field and the orbital moment in the system can play a decisive role in selecting the minimum energy ground-state. Because the electron densities at which gaps occur are magnetic field dependent in states with finite Hall conductivity, small fluctuations in the density like those associated with electron-hole puddle domains will also have an important influence on the nature of the ground-state at low magnetic fields.

We now consider the case of zero magnetic field and finite electric field. In presence of an external bias the inversion symmetry of a graphene bilayer is explicitly broken, favoring charge accumulation in one of the layers. When the external bias becomes large enough the charge balanced broken symmetry antiferromagnetic (AF) configuration gives way to ferrimagnetic (Fi) and eventually ferromagnetic (F) configurations. These solutions can become energetically favored for certain ranges of the external bias potential thanks to their intrinsic broken inversion symmetry configuration with spontaneous charge transfer. In Fig. 5 we present the total energies of the different types of solutions in the low bias regime, obtained by starting the self-consistent calculations from different seeds. For low potential bias the charge balanced AF structure remains lowest in energy. Eventually the external field becomes large enough to flip the layer polarization of one valley-spin component towards the layer favored by the electric field, giving rise to the Fi type solutions. The electrostatic charge imbalance of the Fi states is approximately half of that associated with the F state in which all four components are polarized towards the same layer and have the same magnitude. In the Fi configuration all three charge density components polarized towards the same layer do not have exactly the same value when the electron spins are different, but they are similar in magnitude. In the F configuration all four density components are polarized towards the same layer and have the same magnitude.
FIG. 5: Bias dependence of the total energies of the system for different valley-spin polarization configurations for a system with $\varepsilon_r = 4$. The system undergoes transitions from the AF to Fi and then to F states as a function of external field, each one displaying clearly different quantum Hall conductivity properties. The origin of energy has been arbitrarily shifted for presentation convenience.

The behavior of the band gaps in the low bias regime illustrates the character of each solution. The AF band gap continually decreases when the electric field is increased. In this case the electric field always works against the charge density distribution of two components directly contributing in the reduction of the band gap. For the F configuration we have a completely opposite trend; the gap always increases since the electric field favors charge accumulation in the layer in which the density is already higher. In the Fi configuration we find an intermediate situation. We find an initial increase of the gap in presence of an electric field thanks to the reduction of the Hartree energy penalty associated to the spontaneous charge imbalance. Beyond a certain point the band gap starts to decrease, reflecting the fact that the electric field is working against the layer polarization of one component. Eventually when the effect of the external field becomes dominant the system undergoes a phase transition to the F configuration. Every time there is a crossover of the minimum energy levels we will see an abrupt change in the band gap and charge density. The Hall conductivity of the system will also see discontinuous changes following the classification of table I.

D. Exchange screening effects in presence of a strong bias

In the presence of an external bias the inversion symmetry of graphene bilayer is explicitly broken, favoring the charge accumulation in one of the layers and opening a band gap, a fact that has been verified in several experiments and also predicted theoretically within tight-binding, tight-binding plus Hartree screening or ab-initio calculations. Here we explore the role that electron exchange can play when the system is subject to a strong external bias. In Fig. 7 we present the charge accumulation in one of the layers as a function of bias. In the strong bias limit we are in the F configuration in which all four components are polarized towards the same layer. Results obtained in the Hartree approximation follows a similar trend in the strong bias limit resulting in comparable amounts of total transferred charge in the range of bias we considered, with the Hartree only screening allowing more sloshed charge. A larger value of dielectric screening $\varepsilon_r$ that weakens the interaction strength also weakens the electrostatic screening and therefore more charge imbalance per layer is expected.

As we show in Fig. 7 the presence of the exchange term in
the Hamiltonian introduces a clear enhancement of the band gap that persists up to high bias potentials. This gap enhancement can be related to the exchange contribution that tends to introduce a strong asymmetry in charge distribution between $A$ and $B$ sublattices of a given layer that persists up to rather high values of external electric field as shown in Fig. 7. We may define this intralayer charge imbalance ratio as $(|\Delta n_A| - |\Delta n_B|) / |\Delta n_l|$. The enhancement of charge imbalance between the sublattices is one of the effects of intralayer exchange that contributes to changing the magnitude of the band gap. This quantity is more sensitive to the strength of the Coulomb interaction than to changes in the temperature.

V. SUMMARY AND CONCLUSIONS

We have presented a numerical description of the broken charge symmetry Hartree-Fock ground state in a graphene bilayer system for different dielectric screening of the media and temperature. The predicted gap sizes and temperature ranges in which this effect is expected to survive should be experimentally accessible. The size of the interlayer charge transfers suggests that the broken symmetry states will be suppressed by smooth disorder strong enough to produce charge puddles with density variations larger than around $10^{-5}$ per carbon atom. The total amount of spontaneous charge transfer from one layer to another in the broken ground state is ~ $10^{11} \text{cm}^{-2}$, smaller but comparable in magnitude to the densities that can be induced by gating the device or depositing impurities, and can therefore be relevant for interpreting gating experiments in bilayer graphene. The mechanism for broken symmetry described here is most effective for a neutral bilayer because the energy gain due to charge transfer is greatest for electrons nearest to the Dirac point. The broken symmetries are expected to be present only when the carrier density is smaller than about $10^{13} \text{cm}^{-2}$, hence a reduction of this effect is expected as the Fermi surface moves away from the charge neutrality point when the system is doped.

The Coulomb correlation neglected in the Hartree-Fock formalism usually plays a more important role in extended systems than in localized atomic systems where they are typically one order of magnitude smaller than exchange effects. Treatments of Coulomb correlation based on a renormalization group framework or RPA suggest that the broken symmetry addressed in this paper will survive when Coulomb correlation is considered. In our case we have used larger values of $\varepsilon_r$ than those based on estimates from dielectric functions alone in order to effectively reduce the strength of exchange to approximately account for screening effects. Application of a self-consistent dynamic screening approximation to this problem would be interesting but would require the high density of sampling $k$-points near the Dirac point to be implemented in a computationally efficient way.

An interesting feature of these broken symmetry states is the spontaneous quantum Hall effects which appear when opposite valleys have opposite layer polarizations. When opposite valleys have the same layer polarization, the system has only a valley Hall effect, which is not manifested in standard electrical measurements. There is no energy difference between these states in a continuum model which does not account for lattice-scale physics. We find that the energy difference between these states in a continuum model which does not account for lattice-scale physics. We find that the energy difference between these states decreases rapidly depending on the strength of the on-site effective interaction in our $\pi$-band only model, with valley Hall states being favored over anomalous Hall states. For a very weak on site interaction, anomalous Hall states could be favored over valley Hall states.


FIG. 7: External bias dependence of layer resolved charge density, band gaps and sublattice charge asymmetry. We have considered the $F$ configuration where all four valley-spin components are polarized towards the same layer. Upper panel: Charge accumulation dependence as a function of bias in presence of Hartree and Fock terms and charge accumulation dependence as a function of bias in presence of Hartree screening only. The evolution of $\Delta n_l$ as a function of bias has a slightly larger slope in the Hartree only approximation towards the same layer.

Middle panel: Band gaps obtained for Hartree-Fock and Hartree approximations using different values of $\varepsilon_r$ as a function of external electric field $E$. We notice a substantial enhancement of the band gap when the exchange term is included. Lower panel: Measure of deviation from neutrality in the charge differences for each one of the sublattices in a given layer normalized by the total sloshed charge. The asymmetric distribution of the charge between low energy sublattice $A$ and high energy sublattice $B$ of a given graphene layer is enhanced in presence of electron exchange. This asymmetry becomes smaller as the external bias becomes stronger.
we find that valley Hall states have lower energy than anomalous Hall states, but that the energy competition can be altered by even quite weak external magnetic fields. This scenario appears to be consistent with experiments from the Yacoby group in which a $v = 4$ quantized Hall effect persists down to very weak magnetic fields. The small energy differences between competing states could mean that domains with all characters are present, separated by domain walls, because of entropic and disorder considerations. In the presence of a magnetic field, coupling of spontaneous orbital magnetism to the external field favors anomalous Hall states and should also coarsen any domain structure. Similarly an externally applied potential bias favors valley Hall states and should also coarsen domain structures. In the absence of disorder we do find that the charge gap is first decreased by potential bias, and finally increased once both spins have valley Hall effects with the same layer polarization. This finding is qualitatively consistent with experiment.

We have also presented mean field theory estimates of the critical temperatures associated with spontaneous layer polarization states in graphene bilayers. According to these estimates, order will not survive to room temperatures and therefore is unlikely to be useful for applications. On the other hand, the exchange interactions which produce broken symmetry states at low temperatures, will still play a role in enhancing gaps produced by external potential biases at room temperature.

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