Spontaneous symmetry breaking and topology in twisted bilayer graphene: the nature of the correlated insulating states and the quantum anomalous Hall effect

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We theoretically study the nature of the correlated insulating states and the quantum anomalous Hall (QAH) effect in twisted bilayer graphene (TBG) at the magic angle from the perspective of spontaneous symmetry breaking. Using a generic unrestricted Hartree-Fock method applied to all the energy bands, and including all the valley, spin, and sublattice dependence of the exchange interactions, for the first time, we have successfully explained both the experimentally observed correlated insulating states at ±1/2 fillings and the QAH effect at 3/4 filling of the flat bands near the magic angle. Our results indicate that the correlated insulating states at ±1/2 filling can be states with both valley polarized order and intervalley coherent order (IVC). Both types of order parameters can be stabilized and are the ground states for certain interaction parameters. The valley polarized state breaks time-reversal (T) symmetry, but still exhibits vanishing anomalous Hall effect due to the combined $C_2T$ symmetry associated with each valley. The IVC ordered states break the valley charge conservation, which is massively degenerate due to the separate spin $SU(2)$ symmetry operation of each valley and the valley $U(1)$ symmetry operation. When a hexagonal boron nitride (hBN) substrate is aligned with the TBG, our calculations show that the valley polarized states are energetically favored over the intervalley coherent states at ±1/2 fillings, giving rise to QAH insulating states with Chern numbers ($C$) ±2 by virtue of the $C_2$ symmetry breaking induced by the substrate. Such valley polarized $C = ±2$ QAH states are orbital ferromagnetic and spin paramagnetic states, which would be stabilized by vertical magnetic fields, but would be strongly suppressed by in-plane magnetic fields. Within the same theoretical framework, our calculations indicate that the $C = ±1$ QAH states at ±3/4 filling of the magic-angle TBG would emerge only in the presence of hBN substrate. We further predict that the QAH states at the electron and hole fillings would exhibit hysteresis loops with opposite chiralities. All the QAH phenomena in TBG can be succinctly described by the pseudo-Landau-level representation of TBG.

Moiré graphene systems has been an exciting area of condensed matter physics since the discoveries of the correlated insulating states [1–6] and unconventional superconductivity [2, 7–9] in twisted bilayer graphene (TBG). At small twist angles, the low-energy states of TBG can be characterized by four bands contributed by the two nearly decoupled valleys $K$ and $K'$ [10, 11]. Around the first magic angle, the bandwidths of the four low-energy bands become vanishingly small, and these nearly flat bands are believed to be responsible for the intriguing correlation effects observed in TBG. Numerous theories have been proposed to understand the electronic structures [12–22], the correlated insulating phase [12, 23–33], and the mechanism of superconductivity [12, 24, 27, 29, 34–41] in TBG. However, despite the numerous theoretical proposals, up to date, the nature the correlated insulating states observed at ±1/2 filling of the flat bands of TBG around the magic angle is still an open question.

Besides the correlation effects, the flat bands in magic-angle TBG were found to exhibit nontrivial topological properties characterized by the odd winding of the Wilson loops [16, 17, 20, 42]. In particular, it has been proposed that the moiré potential in TBG would generate some opposite pseudo magnetic fields in the AA regions of TBG, which are coupled with two flavors of Dirac fermions [20]. The two flat bands (per spin per valley) are just the two zeroth pseudo Landau levels (LLs) resulting from the opposite pseudo magnetic fields, which carry opposite Chern numbers ±1 and possess opposite sublattice polarizations [20]. Thus the eight flat bands (including valley and spin) in magic-angle TBG are equivalent to eight zeroth pseudo LLs carrying opposite Chern numbers, sublattice polarizations, and spin quantum numbers. As the kinetic energy is quenched for the flat bands in magic-angle TBG, exchange Coulomb interactions would split the eight-fold degenerate pseudo LLs at any filling factor that could completely fill up an integer number of the pseudo LLs, i.e., at 0, ±1/4, ±1/2, and ±3/4 filling of the flat bands. As each pseudo LL carries nonzero Chern number ±1, it is natural that these interaction-driven insulating states may carry nonzero total Chern numbers (denoted by $C$), as suggested by the quantum anomalous Hall (QAH) effect observed at 3/4 filling of TBG aligned with hBN substrate [43, 44].

In this paper, we try to understand the nature of the correlated insulating states and the possible interaction-driven QAH states at different fillings of magic-angle TBG from the perspective of spontaneous symmetry breaking. We have developed a generic unrestricted Hartree-Fock (HF) method including all the valley, spin, and sublattice dependence of the exchange interactions, and have applied the HF methods to all energy bands of the continuum model of TBG [11] at the magic angle. Both the correlated insulating states observed at ±1/2 fillings in TBG and the QAH state observed at 3/4 filling of the hBN-aligned TBG can be successfully explained within such a generic HF framework. Our results
indicate that the correlated insulating states at $\pm1/2$ filling are states with two kinds of order parameters: the intervalley coherent (IVC) order, and the valley polarized (VP) order. The IVC order breaks the valley $U(1)$ charge-conservation symmetry, but (approximately) each valley still has its own spin $SU(2)$ degrees of freedom, leaving the ground state massively degenerate (at the mean-field level) in an eight-dimensional subspace due to the $SU(2) \times SU(2)$ symmetry operation. On the other hand, the VP state breaks time-reversal ($T$) symmetry, but it does not exhibit any anomalous Hall effect nor orbital magnetism due to the combined $C_{2z}T$ symmetry associated with each valley. When a hBN substrate is aligned with the TBG, however, our calculations show that the VP states are energetically favored over the IVC states at $\pm1/2$ fillings, giving rise to quantum anomalous Hall (QAH) insulating states with Chern numbers ($C$) $\pm2$ by virtue of the $C_{2z}$ symmetry breaking induced by the hBN substrate. Such valley polarized $C = \pm2$ QAH states at $\pm1/2$ fillings would exhibit unusual response to external magnetic fields: the QAH states would be stabilized by the out-of-plane magnetic fields due to the orbital Zeeman effect; but they can be strongly suppressed by in-plane magnetic fields as the spin Zeeman splitting would compete with the valley polarization, and tend to diminish the topological gaps. We find that an in-plane magnetic field $\lesssim 10\, \text{T}$ would be strong enough to completely exclude the $C = \pm2$ QAH states from our phase diagram.

Using the same HF method, we have also studied the QAH states at $3/4$ filling of the flat bands in hBN-aligned TBG at the magic angle. Our results indicate that the $C = \mp1$ QAH state is indeed a state with co-existing spin and orbital ferromagnetic orders at $\pm3/4$ filling, which is consistent with some of the previous theoretical studies and conjectures [22, 31, 33]. We further show that the $C = \pm1$ QAH state would show up only if a significant staggered sublattice potential is present, i.e., only when the TBG system is aligned with the hBN substrate to maxiamally break the $C_{2z}$ and/or sublattice symmetry. This is consistent with the experimental observations [43, 44]. We further predict that, for the same valley polarization, the orbital magnetizations are the same for $\pm p$ ($p = 1/2$ or $3/4$) fillings of the flat bands in hBN-aligned TBG, but the anomalous Hall conductivities are exactly opposite for the $\pm p$ fillings. This implies that the hysteresis loops would have opposite chiralities for the QAH states at the electron and hole fillings, which will be a unique experimental signature for the QAH effects in TBG. All the QAH phenomena in hBN-aligned TBG, including the $C = \pm2$ states at $\pm1/2$ filling and the $C = \mp1$ states at $\pm3/4$ fillings, can be succinctly described using the pseudo-LL representation of the flat bands for magic-angle TBG.

The remainder of the paper is organized as follows. In Sec. I we introduce the moiré lattice structures of TBG, and the continuum model in describing the low-energy states of TBG. In Sec. II we introduce the HF formalism in treating the Coulomb interactions. In Sec. III we discuss the nature of the experimentally observed correlated insulating states at $\pm1/2$ filling of the flat bands of TBG at the magic angle. In Sec. IV we discuss the (possible) QAH effects at $\pm1/2$ and $\pm3/4$ fillings of TBG aligned with hBN substrate and the response to external magnetic fields. In Sec. V we summarize our results.

I. PRELIMINARIES

A. The moiré lattice structure

The commensurate moiré pattern is formed when the top-layer graphene is rotated with respect to the bottom layer by the angle $\theta(m)$, where $m$ is an integer obeying the condition $\cos \theta(m) = (3m^2 + 3m + 1)/((3m^2 + 3m + 1))$ [45]. There are periodic modulations of the AA, AB and BA regions in the moiré pattern. In the AA region, the A (B) sublattice of the top layer is roughly on top of the same sublattice of the bottom layer; while in the AB (BA) regions, the B (A) sublattice of the top layer is roughly on top of the A (B) sublattice of the bottom layer. The lattice vectors of the moiré superlattice can be chosen as $\mathbf{t}_1 = [\sqrt{3}L_s/2, L_s/2]$, and $\mathbf{t}_2 = [\sqrt{3}L_s/2, -L_s/2]$, where $L_s = t_1 = a/(2\sin \theta/2)$ is the superlattice constant of the moiré supercell, and $a = 2.46 \, \text{Å}$ is the lattice constant of monolayer graphene. The corresponding moiré reciprocal lattice vectors $\mathbf{g}_1 = [2\pi/(\sqrt{3}L_s), 2\pi/L_s]$, and $\mathbf{g}_2 = [2\pi/(\sqrt{3}L_s), -2\pi/L_s]$. After the twist, the $K$ ($K'$) points of the two monolayers $K_1$ ($K'_1$) and $K_2$ ($K'_2$) are mapped to the $K_s$ and $K'_s$ points of the moiré supercell Brillouin zone (BZ), as shown in Fig. 1(a). It worth to note that the interlayer distance in TBG varies

\[ \begin{align*}
\begin{array}{c}
\text{(a)}
\end{array}
\end{align*} \]

FIG. 1: (a) The lattice structure and Brillouin zone of the twisted bilayer graphene (TBG). The bandstructure of TBG at the magic angle $\theta = 1.05^{\circ}$ are shown in (b)-(c): (b) without the hBN substrate, and (c) aligned with the hBN substrate. See text for more details.
in real space [46]. As schematically shown in Fig. 1(a), in the AB(BA) region the interlayer distance \( d_{AB} \approx 3.35 \text{ Å} \), while the interlayer distance \( d_{AA} \approx 3.6 \text{ angstrom} \) [47] in the AA-stacked region. Such atomic corrugations can be modeled as [14]

\[
d_z(\mathbf{r}) = d_0 + 2d_1 \sum_{j=1}^{3} \cos (\mathbf{b}_j \cdot \delta(\mathbf{r})) ,
\]

where \( \mathbf{b}_1 = (2\pi/a, 2\pi/(\sqrt{3}a)) \), \( \mathbf{b}_2 = (-2\pi/a, 2\pi/(\sqrt{3}a)) \), and \( \mathbf{b}_3 = \mathbf{b}_1 + \mathbf{b}_2 \) are the three reciprocal lattice vectors of monolayer graphite. \( \delta(\mathbf{r}) \) is a 2D vector indicating the local in-plane shift between the carbon atoms in the two layers around position \( \mathbf{r} \) in the moiré supercell. In the AA region \( \delta \approx (0, 0) \) while in the AB region \( \delta \approx (a/\sqrt{3}, 0) \). We take \( d_0 = 3.433 \text{ Å} \) and \( d_1 = 0.0278 \text{ Å} \) in order to reproduce the interlayer distances in AA- and AB-stacked bilayer graphene.

**B. The continuum model for TBG and the symmetries**

The low-energy states of TBG can be well described by the continuum model proposed in Ref. 11. The low-energy electronic structures can be interpreted as follows. First, due to the formation of the moiré supercell, the Dirac cones from the bottom and top graphene monolayers are folded into the moiré supercell BZ, leading to a massive amount of linearly crossed bands which are entangled with each other. The characteristic bandwidth of the folded Dirac cone in the moiré BZ \( w \sim 2\pi \hbar v_F/L_s \sim 4\pi \hbar v_F \sin(\theta/2)/a \). Second, the Dirac states folded into the moiré BZ are further coupled with moiré potential (denoted by \( U(\mathbf{r}) \)) generated by the moiré pattern. At relatively large twist angles, the moiré potential is small compared with the moiré bandwidth, and can be considered as perturbations which may lead to avoided crossings at moiré BZ boundaries and give rise to some sets of bands that are isolated from the others. However, when the twist angle \( \theta \) is small enough such that the bandwidth is comparable or smaller than the moiré potential, the effects of the moiré potential becomes non-perturbative, and flat bands at a set of magic angles start to show up [11]. We will focus at the first magic angle \( \theta = 1.05^\circ \) throughout this paper.

It worth to note that there are two valleys \( K \) and \( K' \) for each graphene monolayer, and the states around either \( K \) or \( K' \) valleys can be separately folded into the moiré BZ, leading to valley degeneracy in additional to the spin degeneracy. Moreover, at small twist angles, the separation between the low-energy states around the \( K \) and \( K' \) valleys \( \sim |\mathbf{K} - \mathbf{K}'| = 4\pi/(3a) \) is much greater than the size of the moiré reciprocal lattice vector \( |\mathbf{g}_1| = 4\pi/(\sqrt{3}L_s) \) as shown in Fig. 1(a). As a result, the coupling between the low-energy states around the two valleys can be neglected at small twist angles, because the Fourier components of the corresponding moiré potential \( U(|\mathbf{K} - \mathbf{K}'|) \) is vanishingly small given that the potential \( U(\mathbf{r}) \) is smooth on the moiré length scale and that \( L_s \gg a \). Since the low-energy states of TBG from the two valleys are decoupled with each other, the charge is separately conserved for each valley, and the low-energy states of the system has an emergent valley symmetry (dubbed as \( U_v(1) \) symmetry in Ref. 12). Moreover, as spin-orbit coupling is negligible in graphene, there is separate spin \( SU(2) \) symmetry for each valley, such that the entire system has a \( U_v(1) \times SU(2) \times SU(2) \) symmetry.

The above idea can be formulated using the following continuum Hamiltonian [11]:

\[
H^\circ_\mu = \begin{pmatrix} -\hbar v_F(K - K^\circ_\mu) \cdot \sigma^\mu & U^\circ_\mu \\ U^\circ_\mu & -\hbar v_F(K - K_\mu) \cdot \sigma^\mu \end{pmatrix},
\]

where \( \sigma^\mu = [\mu \sigma_x, \sigma_y] \), \( \mu = \pm \) is the valley index, with \( K^- = K \) and \( K^+ = K' \). \( U_\mu(\mathbf{r}) \) denotes the moiré potential for the \( \mu \) valley:

\[
U_\mu(\mathbf{r}) = \begin{pmatrix} u_0g_\mu(\mathbf{r}) & u_0'g_\mu(\mathbf{r} + i\mathbf{r}_{AB}) \\ u_0'g_\mu(\mathbf{r} - i\mathbf{r}_{AB}) & u_0g_\mu(\mathbf{r}) \end{pmatrix} e^{-i\mathbf{K} \cdot \mathbf{r}},
\]

where \( \mathbf{r}_{AB} = (\sqrt{3}L_s, 3/3) \). \( u_0' \) and \( u_0 \) denote the intersublattice and intrasublattice interlayer coupling parameters, and \( u_0 < u_0' \) if the effects of atomic corrugations are taken into account [14]. The phase factor \( g(\mathbf{r}) \) is defined as \( g_\mu(\mathbf{r}) = \sum_{j=1}^{3} e^{-i\mathbf{q}_j \cdot \mathbf{r}} \), with \( \mathbf{q}_1 = (0, -4\pi/3L_s) \), \( \mathbf{q}_2 = (-2\pi/\sqrt{3}L_s, -2\pi/3L_s) \), and \( \mathbf{q}_3 = (2\pi/\sqrt{3}L_s, -2\pi/3L_s) \). \( \Delta \mathbf{K} = \mathbf{K}_2 - \mathbf{K}_1 = [0, 4\pi/(3L_s)] \). In addition to the \( U_v(1) \times SU(2) \times SU(2) \) symmetry, the continuum model of each valley has the symmetry generators \( C_{3z} \), \( C_{2z} \mathcal{T} \), and \( C_{2z} \mathcal{T} \), where \( \mathcal{T} \) is the time-reversal operation for spinless fermions (i.e., complex conjugation). The two valleys can be mapped to each other by \( \mathcal{T} \), \( C_{2z} \), or \( C_{3y} \) operations.

We also consider the situation that TBG is placed on top of an hBN substrate, which is aligned with the bottom graphene layer. This is actually the device used in Refs. 43 and 44, in which (quantum) anomalous Hall effect has been observed at \( 3/4 \) filling of the conduction flat band around the magic angle. The aligned hBN substrate imposes two effects on the electronic structures of TBG. First, the alignment of the hBN substrate with the bottom graphene layer would impose a staggered sublattice potential on the bottom layer grapheme and break the \( C_{2z} \) symmetry, which opens a gap at the Dirac points of the flat bands of TBG. Actually the two flat bands per spin for the \( K \) valley acquires nonzero Chern numbers \( \pm 1 \) (\( \mp 1 \) for the \( K' \) valley) once a gap is opened up at the Dirac points. Second, the hBN substrate would generate a new moiré pattern, which approximately has the same period as the one generated by the twist of the two graphene layers [48, 49], which only
generate some additional sub-bands below and above the flat bands of TBG \cite{43,44}. Therefore, it is a good approximation to neglect the moiré potential generated by the hBN substrate. This is actually the approximation widely adopted in previous theoretical studies \cite{22,33}. With such an approximation, the effective Hamiltonian for the hBN-aligned TBG system is simplified as

$$H_\mu = H_\mu^0 + H_{\text{mass}}$$

where $H_{\text{mass}}$ is the “Dirac mass” term at the bottom layer graphene generated by the hBN substrate, which is expressed as

$$H_{\text{mass}} = \begin{pmatrix} \Delta \sigma_z & 0 \\ 0 & 0 \end{pmatrix}.$$  

(5)

$\Delta = 15$ meV is the staggered sublattice potential exerted on the bottom graphene layer. The $H_{\text{mass}}$ term would break the $C_{2z}F$ symmetry associated with each valley, which will show is crucial in generating the anomalous Hall effect and orbital ferromagnetism.

II. THE MOIRÉ HARTREE-FOCK FORMALISM

The Coulomb interactions between the electrons in TBG can be expressed in momentum space as

$$H_C = \frac{1}{2N_s} \sum_{\alpha k \sigma} \sum_{\sigma' k' q} V(q) c_{k+q,\alpha \sigma}^\dagger c_{k',-q,\alpha' \sigma'}^\dagger c_{k,\alpha' \sigma'} c_{k,\alpha \sigma}$$

(6)

where $k$ is the wavevector of the Dirac fermion in graphene, $\alpha$ is the combined layer and sublattice index, and $\sigma$ is the spin index. $c_{k,\alpha \sigma}^\dagger$ and $c_{k,\alpha \sigma}$ represent the creation and annihilation operators of the Dirac fermions. $N_s$ is the total number of moiré supercells in the entire system. $V(q)$ is Fourier transform of the Coulomb interaction

$$V(q) = \frac{1}{\Omega_M} \int d\mathbf{r} e^2 \frac{e^{-\kappa |\mathbf{r}|}}{4\pi \epsilon_0 |\mathbf{r}|} e^{-i \mathbf{q} \cdot \mathbf{r}},$$

where $\Omega_M$ is the area of the moiré supercell, $\kappa$ is introduced as the inverse screening length, and $\epsilon$ is the background dielectric constant. $\kappa$ and $\epsilon$ will be treated as two free parameters in the following calculations.

If we are interested in the low-energy states around the Dirac points $K$ and $K'$ in graphene, the Dirac fermions can be assigned with the valley index $\mu = \pm 1$. One can re-define the wavevectors $k$ and $k'$ as those expanded around the Dirac point $K'^\mu$, then the Coulomb interactions in Eq. (6) can be divided into the intra-valley term $H_{C,\text{intra}}$ and the inter-valley term $H_{C,\text{inter}}$

$$H_{C,\text{intra}} = \frac{1}{2N_s} \sum_{\alpha k \mu \mu' \alpha' \sigma' \sigma} \sum_{kk'q} V(q) c_{k+q,\mu \sigma}^\dagger c_{k',-q,-\mu' \alpha' \sigma'} c_{k',\mu' \alpha' \sigma'} c_{k,\mu \sigma}$$

(8)

and

$$H_{C,\text{inter}} = \frac{1}{2N_s} \sum_{\alpha k \mu \mu' \alpha' \sigma' \sigma} \sum_{kk'q} V(|K - K'|) c_{k+q,\mu \sigma}^\dagger c_{k',-q,-\mu' \alpha' \sigma'} c_{k',\mu' \alpha' \sigma'} c_{k,\mu \sigma}$$

(9)

Eq. (8) represents the Coulomb scatterings of two electrons which are created and annihilated in the same valley, while Eq. (8) represents some kind of “pair-hopping” process in momentum space in which two electrons are created in $\mu$ and $-\mu$ and get annihilated in $-\mu$ and $\mu$ valleys. The characteristic interaction strength of $H_{C,\text{intra}}$ is $V(|K - K'|) \approx 100$ meV, while the characteristic interaction strength of the intervalley term is $V(|K - K'|) \approx 1.7$ meV at the magic angle. We thus expect the intravalley interaction would dominate over the inter-valley term. The intravalley term $H_{C,\text{intra}}$ preserves the valley charge conservation and the separate spin rotational symmetry of each valley, i.e., the $U_s(1) \times SU(2) \times SU(2)$ symmetry; but the $H_{C,\text{inter}}$ term only preserves the valley charge conservation and a global $SU(2)$ symmetry, i.e., the $U_s(1) \times SU(2)$ symmetry.

Now we make Hartree-Fock approximations to Eq. (8) and Eq. (9). The Hartree term of $H_{C,\text{intra}}$ reads

$$H_{H,\text{intra}} = \frac{1}{N_s} \sum_{\mu \sigma} \sum_{Q} \sum_{\mu' \sigma' \alpha} A(Q) c_{k+Q,\mu,\sigma}^\dagger c_{k,\mu,\sigma}$$

(10)

where the Hartree order parameter $A(Q)$ associated with the moiré reciprocal lattice vector $Q$ is:

$$A(Q) = \sum_{k' \mu' \sigma' \alpha'} V(Q) c_{k'-Q,\mu' \sigma' \alpha'} c_{k',\mu' \sigma' \alpha'}.$$  

(11)

The Hartree potential $A(0)$ represents a spatially homogeneous constant electrostatic potential, which is expected to be canceled by some positive charge background in the system, thus is dropped in our calculations. Then the leading-order Hartree terms are the six first-neighbor $A(Q)$ terms with $|Q| = 4\pi/(\sqrt{3}L_s)$.

The Fock term of Eq. (8) is expressed as

$$H_{F,\text{intra}} = -\frac{1}{N_s} \sum_{\mu \sigma \mu' \sigma' \alpha' \alpha} B_{\mu' \sigma' \alpha', \mu \sigma \alpha} (k + Q) c_{k+Q,\mu \sigma}^\dagger c_{k,\mu' \sigma' \alpha'}$$

(12)

where the Fock order parameter

$$B_{\mu' \sigma' \alpha', \mu \sigma \alpha} (k + Q) = \sum_{k'} V(|k + Q - k'|) c_{k'-Q,\mu' \sigma' \alpha'} c_{k',\mu \sigma \alpha}.$$  

(13)

In this paper, we make the approximations that the exchange interactions are local in real space, such that the wavevector dependence of the Fock order parameters can be neglected, i.e.,

$$H_{F,\text{intra}} \approx -\frac{1}{N_s} \sum_{\mu \sigma \alpha \mu' \sigma' \alpha'} B_{\mu' \sigma' \alpha', \mu \sigma \alpha} (k, \mu \sigma \alpha).$$  

(14)
in which the Fock order parameter $B_{\mu'\sigma'\alpha'\mu\sigma\alpha}$ is no longer dependent on the wavevector $k$. Moreover, we further assume that the Fock order parameters are independent of the layer index, since the two layers have already been equally mixed by the moiré potential to give rise to the non-interacting flat bands at the magic angle [20]. Therefore, in the end, the intravalley Fock terms are dependent on the valley, spin, and sublattice indices, which have 64 independent order parameters.

Next we consider the Fock term of $H_{\text{inter}}$, which can be expressed as

$$H_{\text{F,inter}} = -\frac{1}{N_s} \sum_{k} \sum_{\mu \sigma' \alpha \alpha'} C_{\mu \sigma \alpha, \mu \sigma' \alpha'} c_{k, \mu \sigma \alpha} c_{k', \mu \sigma' \alpha'},$$

where

$$C_{\mu \sigma \alpha, \mu \sigma' \alpha'} = \sum_{k} V(|k - k'|) c_{k, \mu \sigma \alpha}^\dagger c_{k', \mu \sigma' \alpha'},$$

Note that, Eq. (16) is vanishing for intervalley coherent (IVC) order, and the valley polarized (VP) order. The IVC order always breaks the valley charge conservation symmetry, which has the form $\tau_z \sigma_i$ and/or $\tau_y \sigma_j$ ($i, j = 0, x, y, z$), where $\tau$, $\sigma$ and $\alpha$ are the Pauli matrices representing the valley, spin, and sublattice degrees of freedom of the system. However, as discussed in Sec. II, if we neglect the Hartree term of the inter-valley Coulomb interactions (given by Eq. (9)), for the IVC ordered states each valley has its own spin rotational ($SU(2)$) symmetry, such that each IVC state still has a $SU(2) \times SU(2)$ degeneracy. As a result, all the IVC orders $\tau_z \sigma_j$ and/or $\tau_y \sigma_j$ ($j = x, y, z$) can be connected to $\tau_z \sigma_0$ and/or $\tau_y \sigma_0$ by two separate unitary transformations in the spin sectors of the two valleys, i.e., one can always find two $2 \times 2$ unitary matrices $U_1$ and $U_2$ such that

$$\begin{pmatrix} 0 & \sigma_0 \\ \sigma_0 & 0 \end{pmatrix} = \begin{pmatrix} U_1^\dagger & 0 \\ 0 & U_2^\dagger \end{pmatrix} \times \begin{pmatrix} 0 & \sigma_j \\ \sigma_j & 0 \end{pmatrix} \times \begin{pmatrix} U_1 & 0 \\ 0 & U_2 \end{pmatrix},$$

(18)

i.e., $s_0 = U_1^\dagger s_j U_2 = U_1^\dagger s_j U_1$ with $j = x, y, z$. The $SU(2) \times SU(2)$ symmetry makes the resulted IVC state massively degenerate such that the states with any linear combinations of the form $\sum_{i=0, y} \sum_{j=0, x, y, z} \lambda_{ij} \tau_i \sigma_j$ are degenerate with each other, which forms an eight-dimensional degenerate subspace. The tendency of breaking spin $SU(2)$ symmetry is expected to be overwhelmed by the fluctuations within the ground-state manifold, leading to a paramagnetic, and almost featureless “hidden-ordered” insulating state. If the IVC ordered state is the actual ground state of the system, the transition from the IVC ordered phase to the disordered phase may be characterized by a Berezinskii-Kosterlitz-Thouless (BKT) transition with the melting of the IVC vortex and anti-vortex pairs. Including the Hartree term of Eq. (9) would split the $\tau_{x(y)} \sigma_0$-like states from the spin-ordered $\tau_{x(y)} \sigma_j$ ($j = x, y, z$)-like states, but the split is expected to be small as the intervalley interaction is very weak. On the other hand, the VP order can be generically written as $\tau_{x(y)} \sigma_j$ and/or $\tau_{y} \sigma_j$, which preserves the valley charge conservation but breaks $T$ symmetry, and may also break spin rotational symmetry and/or sublattice symmetry.

We have applied the HF method discussed in Sec. II to all the energy bands at $\pm 1/2$ filling of the flat bands of the magic-angle TBG [1], without considering the effects from the hBN substrate. We make the following two approximations on the correlated insulating states at $\pm 1/2$ fillings based on experimental observations: (i) the correlated insulating states preserve the moiré lattice translational symmetry, which is strongly supported by the experimental results from scanning tunneling microscopy (STM) [3–6]; and (ii) we do not consider the situations of breaking rotational symmetries. This is because the TBG systems have heterostrians which break the $C_{3z}$ symmetry at the single-particle level. Thus it is not completely clear the nematicity observed in Refs. 3, 4 are driven by the heterostrain or purely driven by Coulomb interactions.

With these two approximations, all the order parameters can be divided into two categories: the intervalley coherent (IVC) order, and the valley polarized (VP) order. The IVC order always breaks the valley $U(1)$ charge-conservation symmetry, which has the form $\tau_z \sigma_i$ and/or $\tau_y \sigma_j$ ($i, j = 0, x, y, z$), where $\tau$, $\sigma$ and $\alpha$ are the Pauli matrices representing the valley, spin, and sublattice degrees of freedom of the system. However, as discussed in Sec. II, if we neglect the Hartree term of the inter-valley Coulomb interactions (given by Eq. (9)), for the IVC ordered states each valley has its own spin rotational ($SU(2)$) symmetry, such that each IVC state still has a $SU(2) \times SU(2)$ degeneracy. As a result, all the IVC orders $\tau_z \sigma_j$ and/or $\tau_y \sigma_j$ ($j = x, y, z$) can be connected to $\tau_z \sigma_0$ and/or $\tau_y \sigma_0$ by two separate unitary transformations in the spin sectors of the two valleys, i.e., one can always find two $2 \times 2$ unitary matrices $U_1$ and $U_2$ such that

$$\begin{pmatrix} 0 & \sigma_0 \\ \sigma_0 & 0 \end{pmatrix} = \begin{pmatrix} U_1^\dagger & 0 \\ 0 & U_2^\dagger \end{pmatrix} \times \begin{pmatrix} 0 & \sigma_j \\ \sigma_j & 0 \end{pmatrix} \times \begin{pmatrix} U_1 & 0 \\ 0 & U_2 \end{pmatrix},$$

(18)

i.e., $s_0 = U_1^\dagger s_j U_2 = U_1^\dagger s_j U_1$ with $j = x, y, z$. The $SU(2) \times SU(2)$ symmetry makes the resulted IVC state massively degenerate such that the states with any linear combinations of the form $\sum_{i=0, y} \sum_{j=0, x, y, z} \lambda_{ij} \tau_i \sigma_j$ are degenerate with each other, which forms an eight-dimensional degenerate subspace. The tendency of breaking spin $SU(2)$ symmetry is expected to be overwhelmed by the fluctuations within the ground-state manifold, leading to a paramagnetic, and almost featureless “hidden-ordered” insulating state. If the IVC ordered state is the actual ground state of the system, the transition from the IVC ordered phase to the disordered phase may be characterized by a Berezinskii-Kosterlitz-Thouless (BKT) transition with the melting of the IVC vortex and anti-vortex pairs. Including the Hartree term of Eq. (9) would split the $\tau_{x(y)} \sigma_0$-like states from the spin-ordered $\tau_{x(y)} \sigma_j$ ($j = x, y, z$)-like states, but the split is expected to be small as the intervalley interaction is very weak. On the other hand, the VP order can be generically written as $\tau_{x(y)} \sigma_j$ and/or $\tau_{y} \sigma_j$, which preserves the valley charge conservation but breaks $T$ symmetry, and may also break spin rotational symmetry and/or sublattice symmetry.

We have applied the HF method discussed in Sec. II to all the energy bands at $\pm 1/2$ filling of the flat bands,
i.e., filling 2 out of the 8 flat bands for TBG at the magic angle. We treat the background dielectric constant \( \epsilon \) and the inverse screening length \( \kappa \) (in units of \( \text{Å}^{-1} \)) as two free parameters, with \( 1 \leq \epsilon \leq 7 \), and \( 0.001 \leq \kappa \leq 0.014 \). We have compared the energies of the system with the calculated HF ground states in the parameter space spanned by \((\kappa, \epsilon)\) as shown in Fig. 2. In Fig. 2(a) we show the indirect gaps of the HF ground states at -1/2 filling. Clearly there is a large region in the \((\kappa, \epsilon)\) space in which the ground states are insulating. Moreover, such gapped states exhibit vanishing anomalous Hall effect as shown by the calculated AHC in Fig. 2(b).

We have carefully checked the HF order parameters of the insulating states at -1/2 filling, and find that both the VP order and the IVC order can be numerically stabilized, and that in some region of the parameter space they can even co-exist with each other. For example, for \( \kappa = 0.001 \text{ Å}^{-1} \) and \( \epsilon = 3 \), the ground state is insulating with an indirect gap \( \sim 0.0085 \text{ eV} \), and the dominant order parameters are the IVC orders \( \Delta^{ijx}_x \tau_x s_j \sigma_x \) and \( \Delta^{ijx}_y \tau_y s_j \sigma_x \) \((j = x, y, z)\) with \( \Delta^{ijx}_x, \Delta^{ijx}_y \sim 70 \text{ meV} \). The corresponding band structures are shown in Fig. 3(a).

We see that every band is twofold degenerate due to the \( SU(2) \times SU(2) \) symmetry discussed above. To be specific, the full HF Hamiltonian with the generic IVC order parameters \( \tau_x s_j \sigma_x \) and/or \( \tau_y s_j \sigma_x \) \((j = x, y, z)\) can always be transformed to the one with the spin-independent order parameter \( \tau_x s_y \sigma_x \) and/or \( \tau_y s_x \sigma_x \) by two independent unitary transformations \( U_1 \) and \( U_2 \) (see Eq. (18)). Every band of the HF Hamiltonian with \( \tau_x(y) = 0 \)-type of order parameters is twofold spin degenerate, and such twofold degeneracy cannot be lift by unitary transformations shown in Eq. (18). Therefore, the band structures of the HF Hamiltonians with the generic IVC order parameters would remain twofold degenerate.

For \( \kappa = 0.005 \) and \( \epsilon = 2 \), the dominant order parameters are the VP-type orders with \( \tau_0 s_n \sigma_z \) and \( \tau_z s_n \sigma_z \), where \( s_n \) is the Pauli matrix representing the spin operator pointing along the direction \( \hat{n} \). The choice \( \hat{n} \) is arbitrary due to the \( SU(2) \) degeneracy. Such ordered states are valley polarized states with antiferromagnetic-like spin ordering on the \( A \) and \( B \) sublattices, which break \( T \) symmetry and spin rotational symmetry, but preserves a spinor \( C_{2z} \) symmetry, whose symmetry operator is represented by \( \tau_x \sigma_0 \sigma_z K \) (\( K \) for complex conjugation). The orbital part of the \( C_{2z} \) symmetry enforces that both the AHC and orbital magnetization have to vanish, which is consistent with the calculated AHC of the ordered states as shown in Fig. 2(b). Moreover, the full spinor \( C_{2z} \) symmetry also guarantees that the energy bands at every \( k \) point must be twofold degenerate as shown in Fig. 3(b). This is because the spinor \( C_{2z} \) operation would leave the \( k \) point invariant but would flip the physical spin, thus the spinor \( C_{2z} \) symmetry enforces that the states with opposite spins must be degenerate at the same \( k \) point.

Sometimes the VP order and the IVC order may co-exist in the phase diagram of Fig. 2. For example, when \( \epsilon = 2 \), and \( \kappa = 0.007 \text{ Å}^{-1} \), the ground state is insulating with a gap \( \sim 7.5 \text{ meV} \). The order parameters include both the IVC and VP types of orders: \( \Delta_{IVC} \sum_{x,y,z} (\tau_x s_y \sigma_x + \tau_y s_x \sigma_x) + \Delta_{VP} \tau_z s_0 \sigma_z \), where both \( \Delta_{IVC} \) and \( \Delta_{VP} \) are around 10 meV. Such a state with co-existing IVC and VC orders breaks \( T \) and spin rotational symmetry, and it does not have any special symmetry protecting the band degeneracy. Thus each band at a generic \( k \) point is non-degenerate as shown in Fig. 3(c). However, the state with co-existing IVC and VP orders still has an orbital \( C_{2z} \) symmetry represented by \( \tau_x s_0 \sigma_z K \). The orbital \( C_{2z} \) symmetry enforces that the AHC and orbital magnetization must vanish, such that the system is a \( C = 0 \) trivial insulator.

We have also checked that the correlated gaps in all the VP-ordered, IVC-ordered, and the co-existing VP and IVC ordered states (shown in Fig. 3(a)-(c)) tend to decrease with the increase of external magnetic fields due to spin Zeeman effects. This is in perfect agreement with the experimental observations [1].

For large dielectric constant and short screening length, the Coulomb interactions are not strong enough...
FIG. 3: Band structures for the Hartree-Fock ground states of TBG at -1/2 filling at the magic angle: (a) $\epsilon = 3, \kappa = 0.001 \text{Å}^{-1}$, (b) $\epsilon = 2, \kappa = 0.005 \text{Å}^{-1}$, (c) $\epsilon = 3, \kappa = 0.007 \text{Å}^{-1}$, and (d) $\epsilon = 6, \kappa = 0.01 \text{Å}^{-1}$, where $\epsilon$ is the dielectric constant, and $\kappa$ is the inverse screening length.

to open a gap in the system. In such cases, the system tend to stay in a valley-splitting metallic phase with the order parameter $\tau_2 \sigma_0 \sigma_j$ ($j = 0, x, y, z$), as marked in the upper right corner in Fig. 2(a). For example, when $\epsilon = 6$ and $\kappa = 0.01 \text{Å}^{-1}$, the dominant order parameter is $\Delta^{\text{Dirac}} \tau_2 \sigma_2 \sigma_j$ with $\Delta^{\text{Dirac}} \sim -5 \text{meV}$. Such a VP ordered state breaks $T$ symmetry due to the valley polarization, but preserves the orbital $C_{2z}T$ symmetry, which kills the anomalous Hall effect as shown by the calculated AHC in Fig. 2(b). The band structures of the valley-splitting metallic phase are shown in Fig. 3(d). We see that every band is spin degenerate, and the system remains metallic. The phase diagram at $+1/2$ filling (filling 6 out of the 8 flat bands) is similar to that at the $-1/2$ filling. Namely, when the interaction is weak with large dielectric constant and short screening length, the system tends to stay in a valley-splitting metallic phase. When the Coulomb interaction becomes stronger, a global gap opens up in the energy bands, and the system becomes a correlated insulator with both VP and IVC types of order parameters. Similar to the case of $-1/2$ filling, both the valley-splitting metallic states and the correlated insulating states preserve the orbital $C_{2z}T$ symmetry, which kills the anomalous Hall effect. We refer the readers to Supplementary Material for more details about the results at $+1/2$ filling.

IV. THE QAH EFFECTS IN TBG ALIGNED WITHBORON-NITRIDE SUBSTRATE

We continue to study the magic-angle TBG system aligned with hBN substrate at different integer fillings of the flat bands. As discussed in Sec. I B, the alignment of the hBN substrate would impose two effects on the TBG system. First, when hBN is aligned with TBG, the boron (nitrogen) atom is below the $A(B)$ sublattice of the bottom-layer graphene, which strongly breaks the $C_{2z}$ symmetry (namely, $A/B$ sublattice symmetry) of the system. As a consequence, each valley of TBG no longer has the $C_{2z}T$ symmetry, and the AHE and orbital magnetizations are expected to show up once the system becomes valley polarized. Second, the slight mismatch between the lattice constants of hBN and graphene would generate another moiré potential, but it is an order of magnitude weaker than that generated by the mutual twist of the two graphene layers. Therefore, it is an excellent approximation to neglect the moiré potential generated by the hBN substrate. Then the low-energy effective Hamiltonian of hBN-aligned TBG is just given by Eq. (4). In what follows we will apply the moiré HF methods introduced in Sec. II to hBN-aligned TBG at different fillings of the flat bands at the magic angle. QAH insulating states with different Chern numbers are obtained at different fillings. We will also discuss the response of the QAH states to external magnetic fields, and the interpretation of these QAH states from the perspective of the pseudo-Landau-level representation [20].

A. QAH effects at $\pm 1/2$ filling

We first consider the spontaneous symmetry breaking and the possible correlated insulating states at $\pm 1/2$ fillings in hBN-aligned TBG. We find that if the "Dirac-mass term" with $\Delta = 0.015 \text{eV}$ (Eq. (5)) is included, the HF ground states are generally VP states. Since the $C_{2z}$ symmetry is broken by the hBN substrate, the VP states can exhibit non-vanishing AHE and orbital magnetizations. In Fig. 4 (a) and (c) we show the indirect gaps (in units of eV) and the AHC (in units of $e^2/h$) of the HF ground states at $-1/2$ filling of the hBN-aligned TBG. As clearly shown in the figure, the ground state is predicted to be a $C = 2$ QAH insulating state at intermediate interaction strengths. When the interaction is weak, the system becomes a valley-splitting "Chern metal" characterized by large but non-quantized AHC, as indicated in the upper-right corner of Fig. 4(c). The valley polarized QAH insulating states and the valley-splitting metallic states here are similar to the valley-polarized insulating and metallic states discussed in Sec. III. The essential difference is that the alignment of the hBN substrate with TBG would break the orbital $C_{2z}T$ symmetry of each valley, such that non-zero Berry curvatures and Chern numbers become symmetry allowed.

In Fig. 4(b)-(d) we show the indirect gaps and the AHC of the HF ground states at $+1/2$ filling of the hBN-aligned TBG. In the intermediate interaction regime, the calculated HF ground state of the system is a valley-polarized insulating state with quantized AHC $-2e^2/h$, which is opposite to that at the $-1/2$ filling. On the other hand, with the same valley polarization, the orbital magnetizations in the $C = 2$ and $C = -2$ QAH insulating states at $-1/2$ and $+1/2$ fillings have the same sign, which implies that the system would exhibit hysteresis.
loops with exactly opposite chiralities at $-1/2$ and $1/2$ fillings as schematically shown in the insets of Fig. 4(a) and (b). The opposite hysteresis loops at the electron and hole fillings would be a unique experimental signature for the QAH effects in the moiré graphene systems. In the weak interaction regime, i.e., in upper-right corner, the system becomes a valley-split Chern metal with opposite AHC to that at $-1/2$ filling.

The band structures of the $C = \pm 2$ QAH insulators and the valley-splitter Chern metals at $\mp 1/2$ fillings are shown in Fig. 5. In Fig. 5(a) and (c) we plot the band structures of the QAH insulating states with $\epsilon = 3$, $\kappa = 0.005 \, \text{Å}^{-1}$, at $+1/2$ and $-1/2$ fillings with $C = +2$ and $-2$ respectively. The dominant order parameters are $\Delta_{00z} \tau_0 \sigma_0 \sigma_z$ with $\Delta_{00z} \sim 20 \text{meV}$, $\Delta_{00x} \tau_0 \tau_z \sigma_0 \sigma_z$ with $\Delta_{00x} \sim 3.5 \text{meV}$, $\Delta_{00x} \tau_0 \tau_z \sigma_0 \sigma_z$ with $\Delta_{00x} \sim -8.3 \text{meV}$, and $\Delta_{00z} \tau_0 \tau_z \sigma_0 \sigma_z$ with $\Delta_{00z} \sim 2.65 \text{meV}$. The resulted ground state breaks both $T$ and $C_{2z}$ symmetries, but preserves spin rotational symmetry, thus every band is spin degenerate. The spin rotational symmetry can be broken in the ground states at some other $(\kappa, \epsilon)$ parameters, but it does not change the quantized anomalous Hall conductivity which is protected by the topological gap.

In Fig. 5(b) and (d) we show the HF band structures with $\kappa = 0.005$ and $\epsilon = 5$ at $+1/2$ and $-1/2$ fillings. The system stays in the valley-splitting metallic phase, in which the dominant order parameters are $\Delta_{00z} \tau_0 \sigma_0 \sigma_z$ with $\Delta_{00z} \sim 6 \text{meV}$, $\Delta_{00x} \tau_0 \tau_z \sigma_0 \sigma_z$ with $\Delta_{00x} \sim -4 \text{meV}$, and $\Delta_{00z} \tau_0 \tau_z \sigma_0 \sigma_z$ with $\Delta_{00z} \sim 1.8 \text{meV}$. The Fock potentials from the VP ordering are not strong enough to open up a global gap, but the valley-splitting states still exhibit giant anomalous Hall effect by virtue of the $C_{2z}$ symmetry breaking induced by the hBN substrate.

### B. Response to external magnetic fields

The predicted QAH states at $\pm 1/2$ fillings of hBN-aligned TBG exhibit unconventional responses to external magnetic fields. First, when the hBN substrate is aligned with the TBG system, the valley-polarized $C = \pm 2$ QAH states at $\mp 1/2$ fillings are always associated with giant orbital magnetizations $\sim 5-12 \mu_B$ per moiré supercell. In other words, the valley-polarized states are orbital ferromagnetic states when the $C_{2z}$ symmetry is broken by the hBN substrate. Therefore, a weak vertical external magnetic field will assist to stabilize the valley-polarized state by virtue of the giant $g$ factors associated with the orbital Zeeman effect. We thus expect that applying weak vertical magnetic fields will greatly enlarge the area of the $C = \pm 2$ QAH phases in the phase diagrams of Fig. 4(c)-(d).

On the other hands, the valley-polarized $C = \pm 2$ QAH states are also spin paramagnetic states [51], which implies that the spin Zeeman effect would compete with the topological gap generated by the valley polarization. It follows that if one applies an in-plane magnetic field which only couples to spin magnetization, the $C = \pm 2$ QAH states at $\mp 1/2$ fillings will be strongly suppressed. In Fig. 6 (a) and (c) we show the indirect gaps and the AHC of the HF ground states at $-1/2$ filling of hBN-aligned TBG in the presence of an in-plane magnetic field $B_{//} = 5 \text{T}$. Comparing Fig. 6(c) with Fig. 4(c), it is clearly shown that the region of the QAH phase has been significantly reduced due to the presence of the weak in-plane magnetic field. In Fig. 6(b) and (d) we show the
indirect gaps and AHC of the HF ground states at $-1/2$ filling with the in-plane magnetic field $B_{\parallel} = 10$ T. We see that the QAH phase has been completely ruled out from the phase diagram. Therefore, the $C = \pm 2$ QAH states at $\mp 1/2$ fillings exhibit contrasting responses to external magnetic fields, i.e., out-of-plane magnetic field will help to stabilize the $C = \pm 2$ QAH states, while an in-plane magnetic field tends to suppress the QAH states. Such extremely anisotropic responses to external magnetic fields result from the facts that the QAH ground states at $\pm 1/2$ fillings are orbital ferromagnetic but spin paramagnetic states, which are unique properties of the QAH effects of hBN-aligned TBG at $\pm 1/2$ fillings.

C. QAH effects at $\pm 3/4$ filling

Quantum anomalous Hall effects with the Chern number $\pm 1$ has been observed at $3/4$ filling of the flat bands in hBN-aligned TBG at the magic angle [43, 44]. It has been emphasized in Refs. 43 that the alignment of the hBN substrate with the graphene layer is essential in achieving the anomalous Hall effect. When hBN is aligned with graphene, the $C_{2z}$ symmetry is maximally broken generating a staggered sublattice potential $\Delta \sim 15$ meV on the bottom layer graphene, which split the two flat bands (per spin valley per valley) of TBG in such a way that each band becomes topologically nontrivial and carries $\pm 1$ Chern numbers. Including the spin and valley degrees of freedom, there are in total eight topological flat bands with the Chern numbers $\pm 1$. If one fills one out of the eight topological flat bands, or fill seven of the eight flat bands, it is natural that the occupied bands would split from the unoccupied bands due to the exchange interactions, which opens a gap with nonzero Chern number $\pm 1$. This is believed to be the physical origin of the QAH effects observed in hBN-aligned TBG at $3/4$ filling of the flat bands.

Here we apply the moiré HF method discussed in Sec. II to the magic-angle TBG at $\pm 3/4$ fillings of the flat bands. We calculate the ground states of the system both with and without the alignment of the hBN substrate, i.e., with the staggered sublattice potential $\Delta = 0$ and $\Delta = 15$ meV (see Eq. (5)). When $\Delta = 0$, the calculated indirect gaps and AHC of the HF ground states at $-3/4$ filling are shown in Fig. 7(a) and (c) respectively. The indirect gaps are negative in most part of the phase diagram suggesting that the system remains metallic even for relatively strong interactions. These metallic states turn out to be valley-split metals with the dominant order parameters being $\tau_z s_0 \sigma_x$ and $\tau_z s_0 \sigma_0$. Moreover, these metallic states exhibit vanishing AHE as indicated in Fig. 7(c) because the valley-splitting metallic state still has $C_{2z}^{\tau_0}$ symmetry which guarantees that the Berry curvature vanishes at every $\mathbf{k}$ point, thus prohibits anomalous Hall effect. The indirect gaps and AHC of the calculated HF ground states at $+3/4$ filling for $\Delta = 0$ are shown in Fig. 7(b) and (d). Again, the system stays in the valley-splitting metallic phase in most part of the phase diagram, and the AHC vanishes due to the $C_{2z}^{\tau_0}$ symmetry.

The situations are drastically different when an hBN
substrate is aligned with the TBG system. When the staggered sublattice potential $\Delta = 15 \text{meV}$, the calculated HF ground states in the parameter space $(\kappa, \epsilon)$ at $-3/4$ and $3/4$ fillings are presented in Fig. 8. In particular, in Fig. 8(a) and (c) we show the indirect gaps and the AHC of the HF ground states at $-3/4$ filling. In addition to the valley-splitting metallic phase, for some interaction parameters $(\kappa, \epsilon)$ the ground state can be a QAHI insulator with the Chern number $+1$ as marked by “QAHI” in Fig. 8(c). In Fig. 8(b) and (d) we show the indirect gaps and AHC of the HF ground states at $+3/4$ filling. For the same valley polarization, we see that the quantized AHC at $+3/4$ filling is opposite to that at $-3/4$ filling. Moreover, the orbital magnetizations at $3/4$ fillings have the same sign, thus the QAH effects at $\pm3/4$ fillings would have hysteresis loops with opposite chiralities as is the case of $\pm1/2$ fillings. When the interaction is weak, i.e., in the upper-right corners of Fig. 8(c) and (d), the system becomes a valley-split metallic phase with non-vanishing AHC due to the broken $C_{2z} \mathcal{T}$ symmetry.

In Fig. 9(a) and (c) we show the band structures of the QAH states with $\epsilon = 2.5, \kappa = 0.005 \text{ Å}^{-1}$ at $-3/4$ and $3/4$ fillings respectively. The order parameters with $\epsilon = 2.5, \kappa = 0.005 \text{ Å}^{-1}$ at $-3/4$ filling (denoted by $\Delta_{-3/4}$) can be expressed as:

$$\Delta_{-3/4} \approx \Delta_{0z} \tau_0 s_0 \sigma_z + \Delta_{z0z} \tau_z s_0 \sigma_z + \Delta_{z0z} \tau_z s_0 \sigma_z + \Delta_{0nz} \tau_0 s_n \sigma_z + \Delta_{znz} \tau_z s_n \sigma_z,$$

where $\Delta_{0z} \approx 28 \text{meV}, \Delta_{z0z} \approx -12.7 \text{meV}, \Delta_{0nz} \approx -5 \text{meV},$ and $\Delta_{znz} \approx 4 \text{meV}$. $\sigma_n$ is the Pauli matrix representing the spin operator pointing along $\hat{n}$ direction, where the choice of the $\hat{n}$ should be arbitrary due to the spin $SU(2)$ symmetry. Such a state exhibits both orbital and spin ferromagnetic orderings.

In Fig. 9(b) we show the band structures with $\epsilon = 5, \kappa = 0.005 \text{ Å}^{-1}$ at $-3/4$ filling. The system now stays in a valley-split metallic phase with finite orbital magnetization but vanishing spin magnetization. Thus every band is spin degenerate. The order parameters of the QAH states and the valley-split metallic states at $+3/4$ filling are similar to those at the $-3/4$ filling. The corresponding band structures are shown in Fig. 9(c) in the QAH insulating phase, and in Fig. 9(d) in the valley-split metallic phase.

### D. Interpretation from the pseudo Landau level representation

In Ref. 20, the authors of this paper have introduced the pseudo-Landau-level representation of TBG. Here we will show that all the intriguing QAH phenomena in TBG at different fillings can be succinctly described using the pseudo-LL representation of TBG. The essential conclusion of Ref. 20 is that, the effects of the moiré potential in TBG is to generate some opposite pseudo magnetic fields in the AA regions of the moiré pattern. For each valley and each spin, the opposite pseudo magnetic fields are coupled with two flavors of Dirac fermions from the two graphene layers, which generate two sets of pseudo LLs with opposite Chern numbers. The higher LLs are mutually coupled to each other by some other intrasublattice coupling terms in the continuum Hamiltonian. However, the two zeroth pseudo LLs of Dirac fermions with op-
The eigenstates for electrons moving in the $AA$ regions of the moiré pattern. On the other hands, it is well known that in $k$ space the Bloch functions near the $K_s$ and $K'_s$ are localized in the $AA$ regions, thus the two low-energy bands per spin per valley would be flat around $K_s$ and $K'_s$ points at the first magic angle since they are just the zeroth pseudo LLs. As the twist continues to decrease, $\Omega_{AA}$ could enclose multiple pseudo-magnetic flux quanta, i.e. $\Omega_{AA} = n \pi l_B^2$, the corresponding twist angle $\theta_n$ is just the $n$th magic angle.

Including the valley and spin degrees of freedom, there are in total eight flat bands at the first magic angle. These eight flat bands are equivalent to zeroth pseudo LLs with opposite Chern numbers $\pm 1$, opposite sublattices, and opposite spin indices, as schematically shown in Fig. 10(a). Without Coulomb interactions nor the hBN substrate, all the eight pseudo LLs are degenerate. The alignment of the hBN substrate would impose a staggered sublattice potential to the system, which opens a gap $\sim 4$ meV between the states of the $A$ and $B$ sublattices. Without loss of generality, we suppose the $A$ sublattice states are lower in energy. Moreover, our calculations show that the staggered sublattice potential can be dynamically enhanced by Coulomb interactions, which increases the gap between the states with $A$ and $B$ sublattice polarizations. At $-3/4$ filling, only one of the four pseudo LLs with the $A$ sublattice polarization is occupied. The exchange Coulomb interactions would open a gap between the occupied and unoccupied pseudo LLs, leading a polarized pseudo-LL state as shown in Fig. 10(b). Such a state is a quantum anomalous Hall state with both orbital and spin ferromagnetic orderings. At $3/4$ filling, one needs to fill three out of the four pseudo LLs with $B$ sublattice polarizations, and the unoccupied pseudo LL would split from the occupied ones due to exchange interactions, as schematically shown in Fig. 10(c). As a result, the ground state is an orbital and spin ordered QAH state with opposite Chern number to that of $-3/4$ filling.

At $-1/2$ filling, one needs to fill two out of the four pseudo LLs with $A$ sublattice polarizations. The $C = 2$ QAH state is just a valley polarized and spin paramagnetic state with two $C = 1$ pseudo LLs being occupied with the total Chern number $2$, as shown in Fig. 10(d). Similarly, at $1/2$ filling, two out of the four pseudo LLs with $B$ sublattice polarizations, and the unoccupied pseudo LL would split from the occupied ones due to exchange interactions, as schematically shown in Fig. 10(c). As a result, the ground state is an orbital and spin ordered QAH state with opposite Chern number to that of $-3/4$ filling.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure10.png}
\caption{Interpreting the quantum anomalous Hall effects in TBG from the pseudo-Landau-level representations. (a) The eight degenerate zeroth pseudo Landau levels with opposite Chern numbers $\pm 1$, opposite sublattice polarizations $A$ and $B$, and opposite spins. (b) The $C = 1$ QAH state at $-3/4$ filling. (c) The $C = -1$ QAH state at $3/4$ filling. (d) The $C = 2$ QAH state at $-1/2$ filling. (e) The $C = -2$ QAH state at $1/2$ filling.}
\end{figure}

The pseudo magnetic field generated by the moiré potential also introduces a new length scale, the pseudo magnetic length $l_B = \sqrt{L_s \hbar v_F/(4 \pi u'_0)}$, where $L_s$ is the size of the moiré supercell, $u'_0$ is the strength of intersublattice component of the interlayer coupling, and $v_F$ is the Fermi velocity of graphene [20]. The corresponding loop area enclosing one pseudo magnetic flux quantum $S_0 = \pi l_B^2 = L_s \hbar v_F/(4u'_0)$. As $L_s \sim 1/\theta$, we note that $S_0 \sim 1/\theta^2$, while the area of the $AA$ region of the moiré primitive cell $\Omega_{AA} \sim 1/\theta^2$. When the twist angle $\theta$ is relatively large, $S_0$ is greater than $\Omega_{AA}$; but as $\theta$ decreases, $\Omega_{AA}$ increases faster than $S_0$, such that at some critical angle $\theta_c$, the two areas $\Omega_{AA}$ and $S_0$ would be equal to each other, i.e., the $AA$ region of each moiré primitive cell encloses exactly one pseudo magnetic flux quantum. $\theta_c$ is exactly the first magic angle. At the first magic angle, $S_0$ becomes commensurate with $\Omega_{AA}$ such that the electrons in the zeroth pseudo-LL states of each moiré primitive cell can perform the cyclotron motions without being coupled to each other, then the zeroth LLs are
V. SUMMARY

To summarize, we have developed a generic Hartree-Fock method including the valley, spin, and sublattice dependence of the exchange interactions. We have applied such a generic Hartree-Fock method to all energy bands of twisted bilayer graphene, and have constructed full phase diagrams at ±1/2 and ±3/4 fillings both with and without the hBN substrate. For the first time, we have successfully explained both the experimentally observed correlated insulating states at ±1/2 fillings and the quantum anomalous Hall effect at 3/4 filling of the flat bands near the magic angle. Our results indicate that the experimentally observed correlated insulating states at ±1/2 filling can be states with valley polarized order, or intervalley coherent order, or mixture of the two types of order parameters. Both the intervalley coherent order and the valley polarized order can be numerically stabilized, and are predicted as the ground states of the system at ±1/2 fillings for certain interaction parameters. The valley-polarized ordered state breaks time-reversal (T) symmetry, but the anomalous Hall effect and orbital magnetization is prohibited due to the presence of $C_{2z}T$ symmetry. On the other hand, the intervalley coherent order breaks valley charge conservation and may optionally break spin rotational symmetry. The intervalley coherent states form an eight dimensional degenerate subspace, which is generated by the separate spin SU(2) symmetry operation of each valley as well as the valley charge $U(1)$ symmetry operation. As a result, the intervalley coherent ordering may be overwhelmed by the fluctuations within the massively degenerate ground-state manifold, and the the system may become a “Mott-like” insulator.

When the hBN substrate is aligned with the TBG, our calculations show that the valley polarized states are energetically favored over the valley coherent states at ±1/2 filling, leading to quantum anomalous Hall (QAH) insulating states with Chern numbers ($C$) ±2 by virtue of the $C_{2z}$ symmetry breaking induced by the hBN substrate. We further predict that the valley polarized $C = \mp 2$ QAH states can be stabilized by vertical magnetic fields due to the significant orbital Zeeman effect, but would be suppressed by weak in-plane magnetic fields due to the competition between the spin Zeeman splitting and the valley polarization. Our calculations show that an in-plane magnetic field $\lesssim 10^T$ would be strong enough to completely rule out the $C = \mp 2$ QAH state in the entire phase diagram. We have also calculated the Hartree-Fock ground states at ±3/4 fillings of TBG at the magic angle. When the hBN substrate is absent, our results indicate that the system tends to stay in a valley-split metallic phase with vanishing anomalous Hall effect due to the $C_{2z}T$ symmetry. When the hBN substrate is aligned with TBG, a new $C = \mp 1$ QAH state with co-existing spin and orbital ferromagnetic ordering emerges for certain interaction parameters. Our theory is in perfect agreement with the experimental observations at 3/4 filling of the magic-angle TBG [43, 44]. We also predict that, for the same valley polarization, the orbital magnetizations have the same sign for ±p ($p = 1/2$ or 3/4) fillings of the flat bands in hBN-aligned TBG, but the anomalous Hall conductivities are exactly opposite for the ±p fillings. This implies that the hysteresis loops would have opposite chiralities for the QAH states at the electron and hole fillings.

Our work has clarified the nature of the experimentally observed correlated insulating states at ±1/2 filling of TBG, and has explained the quantum anomalous Hall effect at 3/4 filling of hBN-aligned TBG within the same theoretical framework. We have further predicted the emergence of $C = \pm 2$ QAH states at ±1/2 fillings of the flat bands in hBN-aligned TBG, and have shown that these QAH states exhibited opposite hysteresis loops at electron and hole fillings, and can be strongly suppressed by weak in-plane magnetic fields. Our work is a significant step forward in understanding the correlation effects and topological properties of twisted graphene systems, and will provide useful guidelines for future experimental and theoretical works.

Note added: During the preparation of our manuscript, we note the related work posted recently by Bultinck et al. [50], which performs Hartree-Fock calculations for magic-angle TBG including the effects from the remote bands, and have discussed the instability at the charge neutrality point.

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