Gate-Tunable Fractional Chern Insulators in Twisted Double Bilayer Graphene

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We predict twisted double bilayer graphene to be a versatile platform for the realization of fractional Chern insulators readily targeted by tuning the gate potential and the twist angle. Remarkably, these topologically ordered states of matter, including spin singlet Halperin states and spin polarized states in Chern number $\mathcal{C} = 1$ and $\mathcal{C} = 2$ bands, occur at high temperatures and without the need for an external magnetic field.

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Introduction.—Following the remarkable discovery of superconductivity and correlated states in magic angle twisted bilayer graphene [1–6], understanding the phase diagram resulting from electron-electron interactions in different Moiré heterostructures have attracted the interest of many. As a natural progression to the study of the acclaimed twisted bilayer graphene, recent investigations have geared toward twisted double bilayer graphene (TDBG) systems [7–12], formed when two bilayers of graphene—instead of monolayers—are rotated with respect to each other. Experiments on TDBG have established it as a promising platform for interaction-driven states. In particular, it has been reported [13–16] that spin polarized correlated insulator states arise in the narrow flat conduction band.

An important class of states that could arise due to the strong electron-electron interactions in an isolated flat band is fractional Chern insulators (FCIs) [17,18]. These are lattice analogs of the conventional fractional quantum Hall (FQH) effect in two-dimensional electron gases, with the advantage that they occur at zero magnetic field and with potentially significantly larger gaps. There has been a growing interest in investigating different kinds of FCIs in Moiré systems [19–24]. Most saliently, recent theoretical studies pointed to the existence of valley polarized FCIs at fractional filling of the topological $|\mathcal{C}| = 1$ valence or conduction band of twisted bilayer graphene aligned with boron nitride [19–21], as well as their competing gapless phases induced by strong particle-hole symmetry breaking terms which make the fine-tuning of experimental parameters necessary [19].

While very encouraging, twisted bilayer graphene aligned with boron nitride only provides flat bands with $|\mathcal{C}| = 1$, where FCIs can be directly mapped to conventional FQH states due to the topological equivalence between $|\mathcal{C}| = 1$ bands and Landau levels [25–27]. By contrast, a single band with a higher Chern number cannot be simply mapped to multiple decoupled Landau levels [28], leading to novel FCIs in $|\mathcal{C}| > 1$ flat bands that do not have conventional FQH states as continuum counterparts [28–34]. These $|\mathcal{C}| > 1$ FCIs have a “color-entangled” nature which is absent for conventional multicomponent FQH states [28], thus, providing a richer category of FQH physics. However, they were not reported yet in Moiré systems without an external magnetic field.

Motivated by these findings and open questions, here, we explore the premises for FCIs in TDBG. TDBG serves as a natural platform for probing FCIs for a number of reasons. First, its layer configuration breaks the C_2 symmetry by default, hence, removing the band touching at the Dirac points without needing to add a substrate layer. This results in separated bands around charge neutrality that could be individually studied. Second, TDBG bandwidth is highly controllable by tuning the twist angle θ and, more significantly, via the application of an electric field U , providing an extra degree of tunability in experimental setups that are absent in twisted bilayer graphene systems in the regime of twist angles $\theta = 0.6^\circ$ – 1.4° that we consider. This is to be contrasted with twisted bilayer graphene in the regime of very tiny twist angles $\theta \ll 1^\circ$ where electric fields have a qualitatively different impact on the properties [40–45]. In TDBG, the band topology is also controlled by varying U and θ . In particular, the first conduction band, which we focus on, has Chern number values ranging between $\mathcal{C} = 0$ and $\mathcal{C} = 3$ [7–9], allowing us access not only to Landau levels alike bands with $|\mathcal{C}| = 1$ but also to $|\mathcal{C}| > 1$ bands that are topologically distinct from continuum Landau levels. The appearance of an easily accessed $\mathcal{C} = 2$ flat band opens the possibility of realizing

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novel $\mathcal{C} = 2$ FCIs [28–34] in realistic materials without an external magnetic field.

In this Letter, we provide compelling evidence for the existence of FCIs in TDBG through a detailed microscopic study of projected Coloumb interactions onto the first conduction band for both $\mathcal{C} = 1$ and $\mathcal{C} = 2$ regimes. We find a variety of ferromagnetic and spin singlet FCIs at different filling factors that are highly tunable i.e., could be accessed by moving around in the $U - \theta$ space.

Setup.—We consider electrons interacting via the screened Coulomb potential in the TDBG Moiré superlattice. We choose the Yukawa potential $V(\mathbf{q}) = [e^2/(4\pi\epsilon_r\epsilon_0 S)](2\pi/\sqrt{|\mathbf{q}|^2 + \kappa^2})$ to describe the screening of the Coulomb interaction, where e is the electron charge, ϵ_0 is the dielectric constant of vacuum, $\epsilon_r \approx 4$ is the relative dielectric constant of the material [9], S is the area of the Moiré superlattice, and κ measures the screening strength which we set as $\kappa = 1/a_M$ with a_M the lattice constant of TDBG [35]. We emphasize that our results are not sensitive to the details of the screening mechanism [35]. Unless otherwise stated, we assume both polarized spin and valley degrees of freedom for electrons. When the electrons are doped above the charge neutrality point, the first conduction band is partially filled. If the first conduction band is isolated from other bands below and above, it is fair to project the total Hamiltonian to this active band, leading to

$$H^{\text{proj}} = \sum_{\mathbf{k}} E(\mathbf{k}) c_{\mathbf{k}}^\dagger c_{\mathbf{k}} + \sum_{\{\mathbf{k}_i\}} V_{\mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3 \mathbf{k}_4} c_{\mathbf{k}_1}^\dagger c_{\mathbf{k}_2}^\dagger c_{\mathbf{k}_3} c_{\mathbf{k}_4}, \quad (1)$$

where $E(\mathbf{k})$ is the dispersion of the first conduction band, $c_{\mathbf{k}}^\dagger$ ($c_{\mathbf{k}}$) creates (annihilates) an electron with momentum $\mathbf{k}$ in the first conduction band (per spin and valley), and all $\mathbf{k}_i$'s are in the Moiré Brillouin zone (MBZ). The matrix element $V_{\mathbf{k}_1 \mathbf{k}_2 \mathbf{k}_3 \mathbf{k}_4}$ can be derived based on the effective model of TDBG as detailed in the Supplemental Material [35]. To obtain a realistic model of TDBG, we include trigonal warping and particle-hole asymmetry terms and choose $w_1 = 100$ meV and $w_0/w_1 = 0.7$ throughout this work, where w_0 and w_1 are the AA and AB tunneling strengths between two sheets of bilayer graphene, respectively [35]. Despite the fact that the band dispersion is often neglected in studies of FCIs in other settings in order to emphasize the interaction effects, here, we keep this term for considering a more realistic situation. In what follows, we impose periodic boundary conditions on finite samples and use extensive exact diagonalization to study the low-energy properties of the Hamiltonian (1) at various filling factors $\nu = N/(N_1 N_2)$, where N is the number of electrons in the first conduction band, and N_1 and N_2 are the number of unit cells in the two basic directions $\mathbf{a}_1$ and $\mathbf{a}_2$ of the TDBG Moiré superlattice. Each

eigenstate of the Hamiltonian (1) can be labeled by a two-dimensional momentum (K_1, K_2) .

Band isolation.—One of the most attractive advantages of TDBG is the high tunability of low-energy bands via the twist angle θ and the vertical voltage bias U . In particular, the first conduction band can be easily isolated and shows a rich phase diagram of band topology when θ and U are varied [8]. Before studying the interaction driven physics, first, we explore the $U - \theta$ space to find the regions where the first conduction band is well isolated such that the band topology is well defined and the projected Hamiltonian (1) may apply.

We calculate the indirect gap $\Delta \equiv \min(\Delta_e, \Delta_v)$ of the first conduction band, where Δ_e and Δ_v are the indirect gaps to the next excited band and the valence band, respectively. We observe three main regions with significant Δ in the $U - \theta$ space [Fig. 1(a)], in which the isolated first conduction band has Chern number $\mathcal{C} = 0, 1$, and 2, respectively (similar results were reported in Ref. [8] for different model parameters). Here, we define $\mathcal{C} = \int_{\text{MBZ}} d\mathbf{k} \Omega_{\mathcal{C}}(\mathbf{k})/(2\pi)$ with $\Omega_{\mathcal{C}}(\mathbf{k}) = i(\langle \partial\mu(\mathbf{k})/\partial k_x | \partial\mu(\mathbf{k})/\partial k_y \rangle - \langle \partial\mu(\mathbf{k})/\partial k_y | \partial\mu(\mathbf{k})/\partial k_x \rangle)$ the Berry curvature of the first conduction band and $|\mu(\mathbf{k})\rangle$ the corresponding band Bloch eigenfunction. In addition, we have checked that the interaction with the valence band does not change this Chern number in the regime of parameters that we are considering [35]. In the following,

FIG. 1. (a) The indirect gap of the first conduction band as a function of the vertical voltage bias U and the twist angle θ . The Chern number $\mathcal{C}$ of the first conduction band is given in the three main regions where the band is isolated. (b)–(d) Non-interacting band structures near charge neutrality for selective values of θ and U along $\Gamma = (0, 0)$, $K = K_M(\sqrt{3}/2, 1/2)$, $M = K_M(\sqrt{3}/4, 3/4)$, and $K' = K_M(0, 1)$ with $K_M = 4\pi/3a_M$. The first conduction band is shown in red with its Chern number $\mathcal{C}$. Plots are generated with $(t_0, t_1, t_3, t_4, \delta, w_1, w_0) = (2610, 361, 283, 138, 15, 100, 0.7w_1)$ meV [35].

FIG. 2. Evidence of $\nu = 1/3$ FCIs in the $\mathcal{C} = 1$ region with $\theta = 0.75^\circ$, $U = 20$ meV. (a) The low-lying energy spectrum for $N = 8, N_1 \times N_2 = 4 \times 6$, $N = 10, N_1 \times N_2 = 5 \times 6$, and $N = 12, N_1 \times N_2 = 6 \times 6$. (b) The spectral flow for $N = 10, N_1 \times N_2 = 5 \times 6$, where Φ_y is the magnetic flux insertion in the $\mathbf{a}_2$ direction. (c) The finite-size scaling of the energy gap (green) and the ground-state splitting (red) for $N = 4, 5, 6, 8, 10, 12$ without flux insertions. We define the energy gap and the ground-state splitting as $E_4 - E_1$ and $E_3 - E_1$, respectively, where E_i is the i th energy level in ascending order. (d) The particle entanglement spectrum for $N = 10, N_1 \times N_2 = 5 \times 6, N_A = 5$, with 23256 levels below the entanglement gap (the dashed line).

we will focus on the $\mathcal{C} = 1$ and $\mathcal{C} = 2$ regions in Fig. 1(a) and probe the existence of robust FCIs in the first conduction band in both regions.

FCIs in the $\mathcal{C} = 1$ region.—Now, we address the possibility of FCIs stabilized by the screened Coulomb interaction in the $\mathcal{C} = 1$ region. First, we examine $\nu = 1/3$, where one may, in general, expect robust FCIs as the lattice analogs of the celebrated Laughlin state [46]. Indeed, we observe clear three-fold ground-state degeneracies in the energy spectrum of the Hamiltonian (1) for $U = 20$ –30 meV and $\theta = 0.6^\circ - 0.8^\circ$ [Fig. 2(a)] where the three approximately degenerate ground states have momenta (K_1, K_2) which are consistent with the prediction of Haldane statistics for the $\nu = 1/3$ FCIs in a $\mathcal{C} = 1$ band [47–49]. The three-fold ground-state degeneracy persists during the magnetic flux insertion through the handles of the toroidal system, thus, further confirming the robustness of the finite-size fingerprint of a topological degeneracy [Fig. 2(b)]. The energy gap separating the three ground states from excited states becomes much larger than the ground-state splitting as the system size grows and is very likely to survive in the thermodynamic limit as suggested by a finite-size scaling [Fig. 2(c)]. Remarkably, the gap corresponds to a temperature of about 10 K, which is at least an order of magnitude higher than required by the conventional FQH states in two-dimensional electron gases. Importantly, this energy scale is still smaller than the band gap, Δ , confirming the validity of the band projection.

The entanglement spectroscopy of the ground-state manifold with the particle-cut entanglement spectrum (PES) [47,50,51] further corroborate that the ground states are topologically nontrivial. By dividing the whole system into N_A and $N - N_A$ electrons and labeling each PES level by the total momentum (K_1^A, K_2^A) of those N_A electrons, we find a clear entanglement gap $\Delta\xi \approx 0.3$ separating the low-lying PES levels from higher ones [Fig. 2(d)]. The number of levels below this gap exactly matches the pertinent counting of quasihole excitations in the $\nu = 1/3$ Abelian FCIs [47–49], which rules out competing possibilities such as charge density waves.

We have also considered other filling factors. At $\nu = 2/3$, we find the particle-hole conjugate of the $\nu = 1/3$ FCIs reported above [35]. Moreover, we observe vestiges of possible five-fold ground-state degeneracies at $\nu = 2/5$ and $\nu = 3/5$ [35], which may suggest the $\nu = 2/5$ Jain state and its particle-hole conjugate. However, these states are fragile against band dispersion and compete with spinful states (as discussed below).

FCIs in the $\mathcal{C} = 2$ region.—Previous studies have reported novel FCIs residing in flat bands with higher Chern numbers [28–34]. Unlike FCIs in $|\mathcal{C}| = 1$ flat bands, these high- $\mathcal{C}$ FCIs do not have usual continuum FQH states as counterparts. In particular, application of composite fermion theory to Bloch bands predicts a series of high- $\mathcal{C}$ FCIs at $\nu = r/(rk|\mathcal{C}| + 1)$ [34], where $k > 0$ is the number of flux attached to each particle in the composite fermion theory (k should be even for fermions) and r is the number of fully filled composite-fermion bands. Note that r can be negative, which corresponds to negative flux attachment. The energy gap usually decays with the decreasing of ν —specifically with increasing denominators—which makes the states more fragile and harder to observe both in finite-size calculations and experiments. Consequently, we set $k = 2$ and $r = -1$ in the following to consider the largest filling $\nu = 1/3$ in the $\nu = r/(2rk + 1)$ branch for the $\mathcal{C} = 2$ region of TDBG.

Remarkably, we find clear three-fold ground-state degeneracies for relatively large systems ($N \geq 8$ electrons) with $U = 50$ –70 meV and $\theta = 1.2^\circ - 1.4^\circ$ [Fig. 3(a)]. While the energies vary more during the flux insertion than at $\nu = 1/3$ in the $\mathcal{C} = 1$ region, the three-fold topological degeneracy still persists in the spectral flow [Fig. 3(b)]. The finite-size scaling for the three available system sizes within our computational limit suggests that an energy gap of ~ 5 K probably survives in the thermodynamic limit [Fig. 3(c)]. Moreover, we also obtain similar results [35] in the exactly flat $\mathcal{C} = 2$ band in the chiral limit of TDBG [10] where the FCIs at $\nu = 1/3$ should be topologically equivalent to what we observe here in the realistic TDBG model. In that case, we further observe a clear entanglement gap in the PES, which unambiguously

FIG. 3. Evidence of $\nu = 1/3$ FCIs in the $\mathcal{C} = 2$ region with $\theta = 1.35^\circ$, $U = 60$ meV. (a) The low-lying energy spectrum for $N = 8, N_1 \times N_2 = 4 \times 6$, $N = 10, N_1 \times N_2 = 5 \times 6$, and $N = 12, N_1 \times N_2 = 6 \times 6$. (b) The spectral flow for $N = 10, N_1 \times N_2 = 5 \times 6$, where Φ_y is the magnetic flux insertion in the $\mathbf{a}_2$ direction. (c) The finite-size scaling of the energy gap (green) and the ground-state splitting (red) for $N = 8, 10, 12$ without flux insertions. We define the energy gap and the ground-state splitting as $E_4 - E_1$ and $E_3 - E_1$, respectively, where E_i is the i th energy level in ascending order.

rules out the charge density wave and suggests a nontrivial topological order distinct from the $\nu = 1/3$ Laughlin state [35]. All these results confirm the topological nontrivial property of the $\nu = 1/3$ states in the $\mathcal{C} = 2$ region of TDBG.

Spinful FCIs.—So far, we have assumed both valley and spin polarization. Now, motivated by experiments, we keep valley polarization [14] but bring the spin degree of freedom back. In this case, as both the band dispersion and the interaction are independent on the spin flavor if one neglects Hund’s coupling terms that are generically much weaker than the Coulomb interaction, the spinful many-body Hamiltonian [35] is $SU(2)$ symmetric within each valley, which allows us to label each energy level by the

total spin S and its z component $S_z = (N_\uparrow - N_\downarrow)/2$, with $N_\uparrow$ and $N_\downarrow$ the number of spin-up and spin-down electrons, respectively. At $\nu = 1/3$ in both the $\mathcal{C} = 1$ and $\mathcal{C} = 2$ regions, we find three-fold ground-state degeneracies in all S_z sectors, and the ground energies with different S_z are identical (Fig. 4). This confirms the assertion that the $\nu = 1/3$ FCIs observed in both $\mathcal{C} = 1$ and $\mathcal{C} = 2$ regions are, indeed, ferromagnetic with total spin $S = N/2$.

On the other hand, we find that such ferromagnetism can disappear at other filling factors, leading to spin-singlet ground states. For example, at $\nu = 2/5$ in the $\mathcal{C} = 1$ region with U around 20 meV and $\theta = 0.65^\circ - 0.75^\circ$, the ground states are in the $S_z = 0$ sector, thus, having total $S = 0$. Remarkably, five-fold ground-state degeneracies appear in this case (Fig. 5) strongly suggesting the Halperin (332) state as the ground state. As it was found in other graphene based Moiré systems that ferromagnetic states are favored with the increasing of w_0/w_1 [20], there could be a phase transition from the Halperin (332) state to the $\nu = 2/5$ Jain state at w_0/w_1 larger than the value 0.7 chosen in this work.

Discussion.—In this work, we have established twisted double bilayer graphene as a flexible platform for a plethora of fractional Chern insulators which are stabilized by tuning a gate voltage and the twist angle. Remarkably, these exotic states occur at high temperatures, and some of them occur in parameter regimes that are directly experimentally accessible. In particular, the novel $\mathcal{C} = 2$ FCIs at filling $\nu = 1/3$, which we predict around a range of twist angles $\theta \approx 1.2^\circ - 1.4^\circ$, are perfectly within the reach of current experiments in which TDBG samples with $\theta \approx 0.84^\circ - 2^\circ$ have been manufactured [13–16]. While it remains challenging in experiments to realize TDBG samples with θ as low as $0.6^\circ - 0.8^\circ$ where we numerically observe the $\mathcal{C} = 1$ FCIs, future advances in manufacturing van der Waals heterostructures might make this possible. Prospective experimentally realizable FCIs, indeed, provide an intriguing path toward the possibility of many technological applications. In the latter context, the investigation of possible non-Abelian states provides a particularly intriguing outlook.

FIG. 4. The low-lying energy spectra at $\nu = 1/3$ for $N = 8$ spinful electrons on the $N_1 \times N_2 = 4 \times 6$ lattice. The Chern number of the first conduction band is $\mathcal{C} = 1$ in (a) and $\mathcal{C} = 2$ in (b), with $\theta = 0.75^\circ$, $U = 20$ meV and $\theta = 1.35^\circ$, $U = 60$ meV, respectively.

FIG. 5. The low-lying energy spectra at $\nu = 2/5$ for (a) $N = 6$ spinful electrons on the $N_1 \times N_2 = 3 \times 5$ lattice and (b) $N = 8$ spinful electrons on the $N_1 \times N_2 = 4 \times 5$ lattice. Here, we choose $\theta = 0.65^\circ$, $U = 20$ meV.

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