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Strongly Interacting Phases in Twisted Bilayer Graphene at the Magic Angle

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Twisted bilayer graphene near the magic angle is known to have a cascade of insulating phases at integer filling factors of the low-energy bands. In this Letter we address the nature of these phases through an unrestricted, large-scale Hartree-Fock calculation on the lattice that self-consistently accounts for all electronic bands. Using numerically unbiased methods, we show that Coulomb interactions produce ferromagnetic insulating states at integer fillings $\nu \in [-3, 3]$ with maximal spin polarization $M_{\text{FM}} = 4 - |\nu|$. We find that the $\nu = 0$ state is a pure ferromagnet, whereas all other insulating states are spin-valley polarized. At odd filling factors $|\nu| = 1, 3$ those states have a quantum anomalous Hall effect with Chern number $\mathcal{C} = 1$. Except for the $\nu = 0, -2$ states, all other integer fillings have insulating phases with additional sublattice symmetry breaking and antiferromagnetism in the remote bands. We map the metal-insulator transitions of these phases as a function of the effective dielectric constant. Our results establish the importance of large-scale lattice calculations to faithfully determine the ground states of TBG at integer fillings.

Introduction.—When two graphene sheets are twisted by a small angle, dubbed the “magic angle,” their low energy bands reconstruct into flat bands [1, 2]. A cascade of insulating phases has been recently observed in twisted bilayer graphene (TBG) near the magic angle at integer filling fractions of the flat bands [3–7], some of which are in proximity to low-temperature superconducting phases [8–12]. Magic angle TBGs are considered strongly correlated electron systems, with evidence of a strange metal phase at finite temperature [13, 14] and of low-energy electronic collective modes that are strongly coupled to the charge carriers [15]. Because of the quantum geometry of the flat bands and the degeneracy between spin, valley, and sublattice degrees of freedom in graphene [16], interactions in combination with magnetic field, strain and substrate effects can drive various strongly interacting phases. Recent experiments observed evidence of quantum anomalous Hall (AQH) states [17–20], charge order [21], ferromagnetism [18], and Kekule intervalley coherent states [22–24] in the presence of strain.

The nature of intrinsic correlated insulating phases at zero field, in the absence of strain or substrate effects, is not clear. For instance, the insulating state at half-filling ($\nu = 0$) is unknown. Quantum Monte Carlo results in effective models predicted the possibility of valley quantum Hall [25, 26], intervalley coherent (IVC) states and a valence bond solid [25]. Hartree-Fock (HF) calculations found an antiferromagnet [27], a valley Chern insulator [28] and an IVC state [29]. Very little numerical results exist about the $|\nu| = 1$ states. A HF calculation found a Chern insulator with IVC states [23]. At $\nu = -2$, HF [27] and exact diagonalization [30] results encountered a pure spin polarized ferromagnet, whereas other HF results proposed a pure valley polarized state [31] and also an IVC state [23]. A growing consensus between HF and exact diagonalization at $\nu = 3$ indicates that

this state may be a spin-valley polarized Chern insulator [30, 32, 33]. The determination of these phases could shed light in the nature of the superconducting states in TBG and remains an outstanding open question.

At the magic angle $\theta \approx 1.1^\circ$, the Moiré unit cell with size $L \sim a/[2 \sin(\theta/2)] \approx 12$ nm, where a is the graphene lattice constant, has $\sim 10^4$ lattice sites, making *ab initio* methods prohibitively expensive. Previous numerical studies based on Hartree-Fock [28, 29, 31, 32, 34–36], mean-field [37–39], exact diagonalization [30, 33, 40], quantum Monte Carlo [25, 26], and density matrix renormalization group [41, 42] relied on effective low-energy models in the continuum approximation, or else in effective lattice models. The latter were derived either with the extraction of Wannier orbitals from the low-energy flat bands [43–48], phenomenologically [49], or else with the introduction of an energy cutoff in the remote bands [27, 38, 39].

Unlike in graphene monolayer, where continuum models are justified by the separation of the valleys through kinematic constraints [16], in TBG the valleys are not good quantum numbers even in the absence of backscattering [50, 51]. Besides, at length scales shorter than the size of the Moiré unit cell L , Coulomb interactions can be one order of magnitude larger than the energy gap separating the low-energy bands from the remote ones. A more conclusive determination of the ground state needs to account for virtual processes that connect the two sets of bands [52] and may require treating the remote bands on an equal footing with the flat ones through an unbiased calculation on the lattice.

In this Letter we perform an unrestricted, self-consistent, large-scale HF calculation on the lattice with Coulomb interactions screened by metallic gates. The calculation fully accounts for all remote bands in the emergence of many-body states. We find that the in-

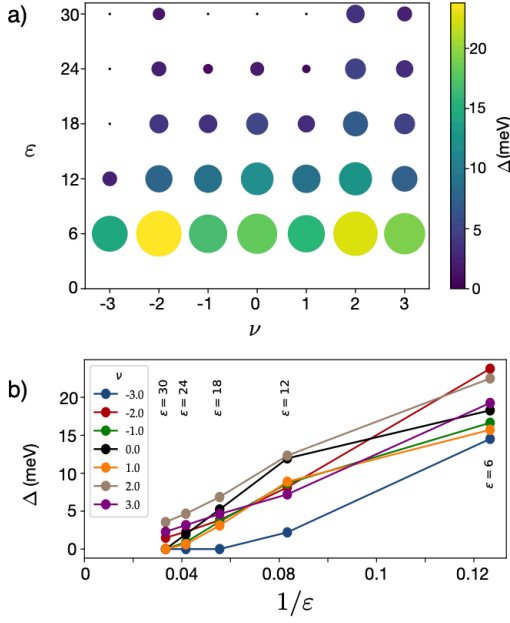


Figure 1: a) Phase diagram of strongly interacting phases in magic angle TBG at integer fillings $\nu \in [-3, 3]$ of the flat bands for different values of the effective dielectric constant ε . Size of the circles and color indicate the amplitude of the many-body gap Δ . Small dots are metallic states (zero gap), indicating the presence of a metal-insulator transition for a critical value of ε . b) Scaling of the many-body gap Δ vs $1/\varepsilon$ for different integer fillings.

insulating ground states of integer fillings $\nu \in [-3, 3]$ are ferromagnetic, with a magnetic moment $M_{\text{FM}} = 4 - |\nu|$ due to the spin polarization of the flat bands. In particular, the $\nu = 0$ state is a pure spin ferromagnet. Valley polarization (VP) is found at all integer ν away from $\nu = 0$. Except for $\nu = 0, -2$ we identified antiferromagnetism (AF) in the remote bands, which appears with sublattice polarization (SLP). Our results also indicate that odd filling factors $\nu = \pm 1, \pm 3$ have quantum anomalous Hall (QAH) states with Chern number $\mathcal{C}=1$.

Those insulating states have a metal-insulator transition (MIT) at critical values of the effective dielectric constant ε , as shown in Fig. 1. The amplitudes of the many-body gaps for $\nu = \pm 3$ are strongly particle-hole *asymmetric* in the moderate- and small-gap regimes, in agreement with experiments [5, 7, 53] and in contrast with continuum models, which are particle-hole symmetric [54]. We conjecture that most experimental samples are in the vicinity of the $\nu = 0$ state MIT. In this regime, we find that the $\nu = \pm 2, 3$ states have prominent gaps of a few meV, whereas the other filling factors $\nu = -3, \pm 1$ are either metallic or have very small many-body gaps, also in agreement with experiment. We suggest that these MITs can be experimentally observed.

Hamiltonian.—We begin with the real space lattice Hamiltonian of TBG, $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_C$, where $\mathcal{H}_0 =$

$\sum_{\alpha\beta} \sum_{ij} h_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) d_{\alpha,i,\sigma}^\dagger d_{\beta,j,\sigma}$ is the hopping term, with $d_{\alpha,i,\sigma}$ the annihilation operator of an electron with spin $\sigma = \uparrow, \downarrow$ on sublattice α of unit cell i , and $h_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j)$ is the corresponding tight-binding matrix element between sublattices α and β located in unit cells centered at $\mathbf{R}_i$ and $\mathbf{R}_j$ respectively. The second term describes a screened Coulomb interaction between any two lattice sites,

$$\mathcal{H}_C = \frac{1}{2} \sum_{\alpha\beta} \sum_{ij} \hat{n}_{\alpha,i} V_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) \hat{n}_{\beta,j}, \quad (1)$$

where $\hat{n}_{\alpha,i} = \sum_{\sigma} d_{\alpha,i,\sigma}^\dagger d_{\alpha,i,\sigma}$ is the local density operator and $V_{\alpha\beta}(\mathbf{R}) = e^2/(\varepsilon\xi) \sum_{m=-\infty}^{\infty} (-1)^m [(\boldsymbol{\tau}_\alpha - \boldsymbol{\tau}_\beta + \mathbf{R})/\xi]^2 + m^2]^{-\frac{1}{2}}$ is the screened form of the interaction in the presence of symmetric gates located at the top and bottom of the TBG heterostructure [55, 56], with e the electron charge and $\boldsymbol{\tau}_\alpha$ the position of a site in sublattice α measured from the center of its unit cell. $\xi \approx 10$ nm is the distance between the plates of the metallic gates in most experiments [10, 12], and ε is the effective dielectric constant, which we treat as a free adjustable parameter. The value of the dielectric constant of graphene encapsulated in hexagonal boron nitride (hBN) is $\varepsilon \approx 6$, although polarization effects due to remote bands in TBG could effectively make it several times larger [27, 30]. We regularize the on-site Coulomb interaction by choosing the onsite Hubbard term $V_{\alpha\alpha}(\mathbf{R} = 0) = 12.4/\varepsilon$ eV of single layer graphene [57] and then smoothly interpolating it with Eq. (1) through $V_{\alpha\beta}(\mathbf{R}) \approx 1.438/[\varepsilon(0.116 + |\boldsymbol{\tau}_\alpha - \boldsymbol{\tau}_\beta + \mathbf{R}|)]$ eV [58]. Although this choice is not unique, the results do not depend on the details of the regularization.

Strain effects can favor ground states that break the translational symmetry of the Moiré pattern [22–24]. To the best of our knowledge, no broken translation symmetry has been observed to date in TBG without strain [59, 60]. We assume the absence of strain effects and rewrite the Hamiltonian in momentum representation,

$$\mathcal{H} = \sum_{\alpha\beta} \sum_{\mathbf{k}, \sigma} h_{\alpha\beta}(\mathbf{k}) d_{\alpha,\mathbf{k},\sigma}^\dagger d_{\beta,\mathbf{k},\sigma} + \mathcal{H}_C, \quad (2)$$

where

$$\mathcal{H}_C = \frac{1}{2} \sum_{\alpha\beta} \sum_{\mathbf{q}} \hat{n}_\alpha(\mathbf{q}) V_{\alpha\beta}(\mathbf{q}) \hat{n}_\beta(-\mathbf{q}), \quad (3)$$

with $V_{\alpha\beta}(\mathbf{q})$ being the Fourier transform of the Coulomb interaction in Eq. (1).

Near the magic angle $\theta \approx 1.085^\circ$ the unit cell of the commensurate structure has $N_b = 11164$ lattice sites, and $\alpha = 1, \dots, N_b$. The tight-binding hopping parameters $t(\mathbf{r})$ are calculated from the standard Slater-Koster functions parametrized by *ab initio* calculations [61, 62]. We define $h_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) \equiv t(\mathbf{r}_\alpha - \mathbf{r}_\beta) - \mu \delta_{\alpha\beta} \delta_{ij}$, with $\mathbf{r}_\alpha = \boldsymbol{\tau}_\alpha + \mathbf{R}_i$ the position of the lattice sites and μ the chemical potential. We account for lattice corrugation

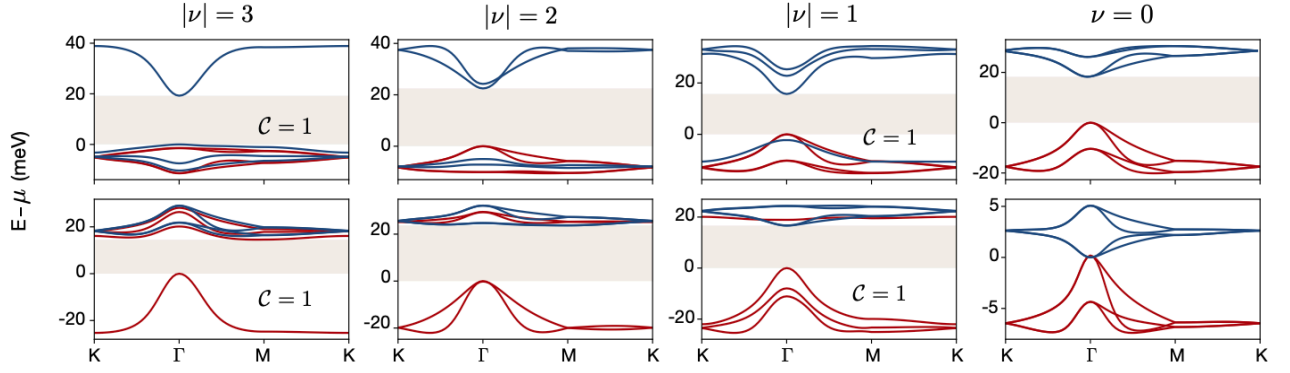


Figure 2: Band structure of the low energy spin polarized bands for integer filling factors. Top row from left to right: $\nu = 3, 2, 1, 0$ for $\varepsilon = 6$. Bottom row (left to right): $\nu = -3, -2, -1$ for $\varepsilon = 6$ and $\nu = 0$ for $\varepsilon = 30$ (metallic state). The gray shadow indicates the size of the many-body gap. Red (blue) lines represent spin σ ($-\sigma$). For $\nu < 0$ ($\nu > 0$) all red (blue) bands are topological, while blue (red) bands are trivial. Odd and even filling factors have a net Chern number $\mathcal{C} = 1$ and zero, respectively. At $\varepsilon = 6$ all integer filling factors have insulating states. The insulating state at $\nu = 0$ is ferromagnetic, with zero valley polarization. All non-zero filling states are spin and valley polarized. The $\nu = 0, \pm 1$ states have MITs in the range $24 < \varepsilon < 30$ (see text).

effects by allowing the layers to relax in the z direction [44, 62–64].

Hartree-Fock calculation.—We proceed to identify the many-body ground states of the problem through a large-scale, unrestricted, self-consistent HF lattice calculation. A HF decomposition of the Hamiltonian reduces the problem to one of diagonalizing the effective Hamiltonian

$$\mathcal{H}_{\text{HF}} = \sum_{\mathbf{k}, \sigma} \sum_{\alpha \beta} \bar{h}_{\alpha \beta}(\mathbf{k}, \sigma) d_{\alpha, \mathbf{k}, \sigma}^{\dagger} d_{\beta, \mathbf{k}, \sigma}. \quad (4)$$

Here $\bar{h}_{\alpha \beta}(\mathbf{k}, \sigma) = h_{\alpha \beta}(\mathbf{k}) + h_{\alpha \beta}^{\text{H}}(\mathbf{k}, \sigma) + h_{\alpha \beta}^{\text{F}}(\mathbf{k}, \sigma)$ is the renormalized matrix elements due to both Hartree,

$$h_{\alpha \beta}^{\text{H}}(\mathbf{k}, \sigma) = \delta_{\alpha \beta} \sum_{\gamma, \mathbf{k}', \sigma'} V_{\beta \gamma}(0) \rho_{\gamma \gamma}(\mathbf{k}', \sigma'), \quad (5)$$

and Fock contributions,

$$h_{\alpha \beta}^{\text{F}}(\mathbf{k}, \sigma) = - \sum_{\mathbf{k}'} V_{\alpha \beta}(\mathbf{k} - \mathbf{k}') \rho_{\alpha \beta}(\mathbf{k}', \sigma). \quad (6)$$

Each contribution can be cast in terms of the $N_b \times N_b$ zero-temperature density matrix for a given momentum and spin, $\rho_{\alpha \beta}(\mathbf{k}, \sigma) = \sum_n^{\text{occupied}} \phi_{\alpha, \mathbf{k}}^{(n)}(\sigma) \phi_{\beta, \mathbf{k}}^{(n)*}(\sigma)$, which is defined as a sum over all occupied bands, with $n = 1, \dots, N_b$. The N_b -component spinors $\phi_{\alpha, \mathbf{k}}^{(n)}(\sigma)$ describe the exact eigenvectors of Eq. (4), which need to be calculated self-consistently while enforcing a fixed total number of particles per unit cell.

The ground state at the HF level is found by self-consistently calculating the real space density matrix, $\hat{\rho}_{\alpha \beta}(\mathbf{R}, \sigma)$, from a 9×9 mesh in momentum space including the Γ point. Each self-consistent loop renormalizes the chemical potential of the previous step to ensure conservation of the total number of particles. All densities are measured away from the neutrality point.

The initial density matrix is extracted from the relaxed non-interacting tight binding Hamiltonian and self-consistently iterated until convergence [65]. In order to probe for many-body instabilities, we seed the initial density matrix with random noise, allowing for all symmetries (with the exception of translational symmetry) to be broken. After convergence, we then perform numerical measurements with the resulting density matrix to identify the ground states at different filling factors. This approach provides an unbiased method to probe for many-body instabilities at the HF level.

Ferromagnetic ground states.—In Fig. 2 we show the reconstructed low energy bands of magic angle TBG for integer filling factors $\nu \in [-3, 3]$ for $\varepsilon = 6$. At this value of ε we find that all integer fillings correspond to insulating states. Interactions increase the bandwidth of the low-energy flat bands by a factor of ~ 3 , while splitting their spin degeneracy. The lines in red (blue) are spin σ ($-\sigma$) bands, which are fully spin polarized, with large many-body gaps Δ (see Fig. 1a). The spin polarization M_{FM} changes in integer increments between integer filling factors and is maximal at half filling ($\nu = 0$), where the ground state has four occupied mini-bands with the same spin σ . We find that the system has maximum total spin polarization $M_{\text{FM}} = 4 - |\nu|$ for $\nu \in [-3, 3]$.

This behavior is observed in all insulating states at higher values of ε . For $\varepsilon = 12, 18, 24$ and 30 the many-body gaps progressively decrease while the low-energy bands become flatter, leading to a cascade of filling-factor-dependent MITs [66]. For instance, at $\varepsilon = 18$ the $\nu = -3$ state is metallic whereas the $\nu = 0, \pm 1$ states have a MITs in the range between $24 < \varepsilon < 30$ (see Fig. 1). The $\nu = \pm 2, 3$ states are more robust, persisting at much larger values of ε . The scaling of Δ with $1/\varepsilon$ for all integer fillings is shown in Fig. 1b.

The ferromagnetic order parameter can be defined di-

rectly from the real-space density matrix as $M_{\text{FM}} = \sum_{\alpha} s_{\alpha}$, with $s_{\alpha} = \hat{\rho}_{\alpha\alpha}(0, \sigma) - \hat{\rho}_{\alpha\alpha}(0, -\sigma)$ the local spin polarization summed over all lattice sites in the Moiré unit cell. The measurements of M_{FM} are summarized in Fig. 3a. Only the flat bands contribute to the magnetization and to the ensuing ferromagnetic order [67]. With the exception of half and quarter filling ($\nu = 0, -2$), we find SLP in all other insulating states. The order parameter for SLP breaks the symmetry between sublattices A and B in each monolayer and is defined as $m_{\text{SLP}} = \sum_{\sigma, \alpha \in A} \hat{\rho}_{\alpha\alpha}(0, \sigma) - \sum_{\sigma, \alpha \in B} \hat{\rho}_{\alpha\alpha}(0, \sigma)$. In Fig. 3b we show the contribution to SLP from both the low-energy and remote bands.

SLP is accompanied by a sublattice modulation of the ferromagnetism in the low-energy bands, and by a pattern of *staggered* magnetization in the *remote* bands, indicating AF order [67]. In Fig. 3c we display the AF order parameter $M_{\text{AF}} = \sum_{\alpha \in A} s_{\alpha} - \sum_{\alpha \in B} s_{\alpha}$ for different integer fillings. Both M_{AF} and m_{SLP} vanish at $\nu = 0, -2$ and closely follow each other at other integer filling factors, as shown in Fig. 3b, c. We see weak SLP and AF order at $\nu = 2$.

Topological states.—We next calculate the Chern number of the flat bands through the eigenstates extracted from the self-consistent density matrix, $\mathcal{C}_n(\sigma) = -(i/2\pi) \int_{\text{BZ}} d^2\mathbf{k} \cdot [\nabla_{\mathbf{k}} \times \langle \phi_{\mathbf{k}}^{(n)}(\sigma) | \nabla_{\mathbf{k}} | \phi_{\mathbf{k}}^{(n)}(\sigma) \rangle]$, with the integral set over the moiré Brillouin zone. The total Chern number of the occupied topological flat bands is $\mathcal{C} = 1$ at $\nu = \pm 1, \pm 3$ and zero at $\nu = 0, \pm 2$ for all values of ε with insulating states. At $\nu < 0$ ($\nu > 0$), all flat bands with majority (minority) spin shown in red (blue) in Fig. 2 are topological, whereas the ones with minority (majority) spin shown in blue (red) are trivial. All bands are trivial at $\nu = 0$.

We verify this result by doing numerical measurements of the QAH order parameter through the real-space density matrix, $m_{\text{QAH}} = \frac{1}{3\sqrt{3}} \text{Im} \sum_{\langle\langle \alpha, \beta \rangle\rangle} \eta_{\alpha\beta} \hat{\rho}_{\alpha\beta}(0, \sigma)$, which probes for chiral loop currents among next-nearest-neighbor sites $\langle\langle \alpha, \beta \rangle\rangle$, where $\eta_{\alpha\beta} = \pm 1$ for clockwise or counterclockwise hopping in the honeycomb plaquette of each monolayer [68]. As shown in Fig. 3d, $m_{\text{QAH}} = 0$ at even filling factors and is finite at odd fillings, in agreement with the calculated Chern numbers depicted in Fig. 3e. We note that the $\nu = -3$ state for $\varepsilon \geq 18$ and $\nu = \pm 1$ for $\varepsilon = 30$ are spin polarized metals [66]. Although we observe bulk loop currents in those states ($m_{\text{QAH}} \neq 0$), they do not generate chiral topological edge modes and the Chern number is not well defined because of the finite Fermi surface around the Γ point.

Valley polarization.—In TBG the valleys are emergent quantum numbers that set the overall degeneracy of the flat bands in the non-interacting limit [69, 70]. To measure VP effects we use the order parameter $m_{\text{VP}} = \frac{1}{3\sqrt{3}} \text{Im} \sum_{\langle\langle \alpha, \beta \rangle\rangle} \delta_{\alpha} \eta_{\alpha\beta} \hat{\rho}_{\alpha\beta}(0, \sigma)$, with $\delta_{\alpha} = \pm 1$ for $\alpha \in A$ or B , which sums over the difference between the loop

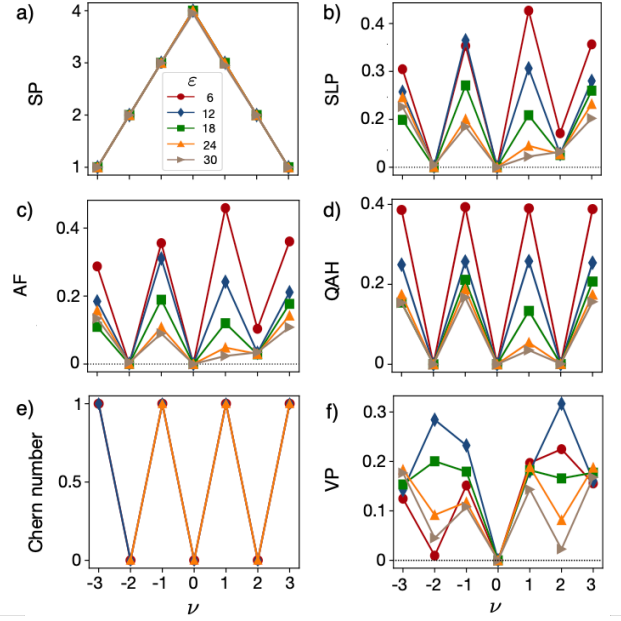


Figure 3: Magic angle TBG instabilities at $\varepsilon = 6$ (red dots), 12 (blue diamonds), 18 (green squares), 24 (orange triangles) and 30 (gray triangles) and for integer $\nu \in [-3, 3]$: a) Spin polarization, b) sublattice polarization, c) antiferromagnetism, d) quantum anomalous Hall, e) total Chern number and f) valley polarization. The $\nu = 0$ state has zero valley and sublattice polarization (see text).

currents in sublattices A and B in each monolayer [51]. In Fig. 3f we show the total VP calculated for integer fillings. The behavior of m_{VP} is not monotonic with the strength of interactions. For all values of ε we find that VP and SLP are Pauli blocked at half filling, $m_{\text{VP}} = m_{\text{SLP}} = 0$, due to the $\nu = 0$ state being maximally spin polarized ($M_{\text{FM}} = 4$), exhausting the occupation of the flat bands with the same spin.

Discussion.—Exact diagonalization results [30] and exact calculations in the chiral limit of the continuum model [71] indicate that unrestricted HF may faithfully capture the ground state of magic angle TBG at integer fillings, where correlation effects are minimal, and interactions are mostly dominated by exchange. Transport experiments indicate the prevalence of insulating states at $\nu = \pm 2, 3$ [5, 7, 53], in agreement with our results for $\varepsilon \gtrsim 24$, which show a strong particle-hole asymmetry between $\nu = \pm 3$ states.

We note that our results at $\nu = 0$ significantly depart from prior quantum Monte Carlo and HF results in low energy effective models [25, 25–28], and indicate the presence of a pure spin ferromagnet with maximal polarization. The $\nu = \pm 2$ states are spin-valley polarized with zero net Chern number, in contrast with prior exact diagonalization and HF calculations in low energy models, which predicted either spin [27, 30] or valley polarized states [31]. The $\nu = \pm 1, \pm 3$ states are found

to be spin-valley polarized Chern insulators. In particular, our results at $\nu = 3$ qualitatively agree with previous studies [30, 32, 33]. We find that SLP is accompanied with the emergence of AF order in the remote bands at $\nu = \pm 3, \pm 1, 2$, and with sublattice modulation of the ferromagnetically ordered state in the flat bands. Recent electron spin resonance transport measurements have found evidence of magnetism and AF order in magic angle TBG at $\nu = \pm 2, 3$ [7].

Our numerical calculations also shed light into the MIT transition of those insulating states. The $\nu = 0$ insulator has been observed in some transport experiments [3] but not in others [5, 7, 53]. According to the scaling of the many-body gaps shown in Fig. 1b, the typical transport gap of a few meV observed at $\nu = -2$ in most HBN encapsulated samples is consistent with an effective dielectric constant $\epsilon \sim 25 \pm 5$, close to the $\nu = 0$ state MIT. We predict that solid insulating behaviors at $\nu = 0, \pm 1$ could be observed in suspended samples. Our results set the framework for the use of large-scale HF lattice calculations in moire heterostructures.

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