Emergence of intrinsically isolated flat bands and their topology in fully relaxed twisted multi-layer graphene

Xianqing Lin,1,* Haotian Zhu,1 and Jun Ni2

1College of Science, Zhejiang University of Technology, Hangzhou 310023, People’s Republic of China
2State Key Laboratory of Low-Dimensional Quantum Physics and Frontier Science Center for Quantum Information, Department of Physics, Tsinghua University, Beijing 100084, People’s Republic of China

(Dated: April 20, 2021)

We study the electronic structure and band topology of fully relaxed twisted multi-layer graphene (TMLG). Isolated flat bands emerge in TMLG with the number of layers \( |M + N| \) up to 10 and with various stacking orders, and most of them are on the hole side. The touched bands of FLGs around the Fermi level are split by the moiré coupling through the electron-hole asymmetry in low-energy bands of FLGs and by the vertical hopping between next-nearest layers. The full structural relaxation leads to global gaps that completely isolate a flat band. For TMLG with given \( M \) and \( N \), the highest magnitude of Chern numbers \( |C| \) of the separable flat bands reaches \( M + N - 1 \) and can be hosted by certain isolated bands. The \( |C| = 9 \) occurs in the isolated flat valence band of several configurations with 10 layers. Such high \( |C| \) originates from the lifting of the band-state degeneracy in the weak regime of moiré coupling or from the topological phase transitions induced by the strong moiré coupling. Moreover, large orbital magnetic moments arise in isolated flat bands with high \( |C| \) and depend on the structural configurations of TMLG.

I. INTRODUCTION

The emergence of low-energy flat bands\(^1\)–\(^4\) and the observation of associated superconductivity and correlated-insulator phases\(^5\)–\(^12\) in magic-angle twisted bilayer graphene (TBG) have inspired great interest in exploring the peculiar electronic structure of graphene moiré systems\(^13\),\(^14\). Finite Berry phases around the Dirac cones of monolayer graphene are maintained in TBG\(^15\)–\(^18\), while the breaking of \( C_2T \) symmetry is required to obtain separable flat bands with nontrivial Chern numbers \( |C| \)\(^19\)–\(^21\), which can be achieved by carefully aligning TBG with hexagonal boron nitride to commensurate twist angles between them\(^22\)–\(^26\). In such heterostructures, quantized anomalous Hall conductivity (QAHC) \( \sigma_{xy} = C e^2/h \) is observed at an odd filling of a flat band together with spontaneous orbital ferromagnetism\(^27\),\(^28\), QAHC with \( |C| = 2 \) has been realized in chirally stacked trilayer graphene aligned with BN\(^29\). In contrast to TBG, intrinsic flat Chern bands in a valley arise in BA-AB stacked twisted double bilayer graphene (TDBG) and their \( |C| \) can reach \( \sqrt{3} \)\(^30\)–\(^35\). This suggests that higher-order flat Chern bands may occur in thicker twisted multi-layer graphene (TMLG) composed of two few-layer graphene (FLGs) with relative rotation. Moreover, complete isolation of a flat Chern band by global gaps from other bands is also essential to obtain QAHC that is contributed purely by edge states\(^25\). Therefore, it is important to explore TMLG systematically to identify isolated flat bands with high-order topology in graphene moiré systems.

The electronic structure of TMLG depends on the stacking orders of the FLGs and the twist angle \( \theta \)\(^32\),\(^36\)–\(^41\). The flat bands around the Fermi level \( E_F \) of TMLG were found to be entangled with each other or with other dispersive bands by band crossings\(^32\),\(^38\)–\(^41\). Most studies introduced external electric field to separate the flat bands so that their band topology becomes well defined, while the produced \( C \) is limited to small values\(^39\),\(^41\). For TMLG composed of chirally stacked FLGs, the flat valence and conduction bands in a valley can be separated from other bands and their total \( C \) were demonstrated to increase with the layer numbers of TMLG\(^32\). This suggests that high \( C \) may occur in one flat band if it can become isolated. We note that only rigid moiré superlattices of TMLG were considered in these previous studies\(^32\),\(^38\)–\(^41\), while full relaxation has been shown to be able to enhance the band separation in TBG and TDBG\(^35\),\(^42\)–\(^46\). In addition, the interlayer coupling was limited to that between adjacent layers\(^32\),\(^38\)–\(^41\). The coupling between next-nearest layers may also play an important role to isolate the flat bands.

Here, we have identified various stacking orders of fully relaxed TMLG with isolated flat bands. Certain isolated flat valence bands in configurations with \( M + N \) layers can host \( |C| \) as large as \( M + N - 1 \) and also large orbital magnetic moments for \( M + N \) up to 10. The mechanism behind the emergence of isolated flat bands and their significant band topology have been revealed.

II. STRUCTURAL CONFIGURATIONS OF TMLG

We study TMLG with the top FLG (tFLG) rotated by \( \theta \) counterclockwise and the bottom FLG (bFLG) fixed, as seen in Fig. 1(a). The layer numbers of bFLG and tFLG are denoted by \( M \) and \( N \), respectively. We consider the strictly periodic moiré superlattices of TMLG with \( M + N \) up to 10 and \( \theta \) from 1.890° to 1.018°\(^\circ\). Starting from the twist interface, the layers in bFLG and tFLG are indexed by \( n = 1 \sim M \) and \( \tilde{n} = \tilde{1} \sim \tilde{N} \), respectively.
Besides the tunable $\theta$, there exist $2^{M+N-3}$ inequivalent stacking orders for TMLG with $M + N$ layers, which are represented by the stackings of tFLG and bFLG. For example, a configuration composed of tFLG with the BA stacking and bFLG with the ABCA stacking is denoted by BA-ABCA. The geometry of moiré superlattices in TMLG is detailed in the Supplemental Material (SM).

Within a FLG, a pair of sites from two adjacent layers with one site directly above the other are referred to as dimer sites, and there is a relatively strong interlayer hopping ($\gamma_1$) between them. In a chirally stacked FLG, the sites in inner layers are all dimer sites, and the non-dimer sites which constitute the low-energy band states are on the surface layers. For a general stacking of FLG, the layers can be decomposed into a sequence of chiral subsets, such as (AB)/(ABC). Such chiral decomposition of tFLG and bFLG in TMLG determines some characters of the electronic structure.

In TMLG, the local stacking between layer $\tilde{1}$ of tFLG and layer 1 of bFLG varies continuously across the moiré superlattice. Then the optimal local spacing between these layers determined by the local stacking also varies with the in-plane position in the superlattice, leading to the corrugation of the layers. More importantly, the spatially varying potential at the twist interface can drive the in-plane structural relaxation. Full relaxation has been performed for each configuration of TMLG employing the continuum elastic theory, as detailed in the SM.

### III. ELECTRONIC STRUCTURE OF TMLG

For each relaxed configuration of TMLG, we have built a tight-binding Hamiltonian ($\hat{H}$) taking into account the effects of out-of-plane corrugation and in-plane structural deformation. The Hamiltonian parameters and computational approaches of the electronic structure are given in the SM. It is noted that the vertical next-nearest-layer hopping ($\gamma_2$) within the top and bottom FLGs is included in $\hat{H}$. To diagonalize $\hat{H}$, the plane-wave-like basis functions are adopted and denoted by $|n\alpha, \mathbf{k}\rangle$, where $n$ is the layer index, $\alpha = A,B$, and $\mathbf{k}$ represents the momentum of the state in the reciprocal space of the pristine FLGs.

By calculating the two-dimensional (2D) energy bands in the entire Brillouin zone (BZ) for relaxed TMLG with different $\theta$ and stackings, we find that completely isolated flat bands emerge around $E_F$ for various configurations of TMLG, even for those with 10 layers. We consider flat bands with widths smaller than 10 meV. For an isolated band, global gaps are opened above and below this band. The bands around $E_F$ show rather strong electron-hole asymmetry and most isolated flat bands are on the hole side. The stacking orders that host isolated flat valence bands in a relatively large range of $\theta$ are listed in Table I and those with flat valence bands that are only isolated at some $\theta$ can be seen in Table SI of the SM. Figure 1(b-e) show the band structures of four typical configurations with different types of stacking orders and their DOS can be seen in Fig. S3. Since only very few configurations have isolated flat conduction bands (see Table SII), we focus on isolated flat valence bands in the following.

Through examination of variations of the electronic...
properties with different configurations of TMLG, we category the stacking orders into four types, i.e. cases with chiral stacked FLGs (type I), those with Bernal-stacked FLGs that have even numbers of layers (type II), other cases without a single layer in the stacking decomposition of FLGs (type III) and with a single layer in decompositions (type IV), as listed in Tables I and SI. The isolated flat valence band is separated from other bands by global gaps at the charge neutrality point ($\Delta_0$) and just below it ($\Delta_h$). $\Delta_0$ is smaller than 4 meV for all cases, while $\Delta_h$ can have a rather large value, especially for type-I cases, as shown in Fig. 2(a). Most type-IV configurations have extremely narrow $\Delta_h$ as some characters of the low-energy linear dispersions contributed by the decomposed single layer in the pristine FLGs are maintained. The $\Delta_0$ of most type-II cases are quite large, while those of many type-III cases are rather small. The widths ($W_v$) of isolated valence bands begin to become smaller than 10 meV at $\theta \approx 1.5^\circ$, as shown in Fig. 2(b). For type-I cases, $W_v$ can be narrower than 5 meV only when $\theta$ is around or below $1.1^\circ$, while the $W_v$ of type-III and type-IV cases can reach such narrow $W_v$ at about $1.3^\circ$. The electronic behavior of different stacking types also depends on the thickness of TMLG. Only when $N \geq 3$, isolated flat bands can emerge for type-I stacking, as seen in Table II. For $N \geq 7$, only type-III and type-IV cases can have isolated flat bands. No isolated flat bands exist for systems with $N = 1$. In addition, similar to pristine FLGs$^{47,48}$, the 2D energy dispersions of the isolated flat bands exhibit trigonal warping, as seen in Fig. S2.

To reveal the mechanism behind the emergence of isolated flat bands in TMLG, we have compared the band structures and state composition of some configurations with different parts of the Hamiltonian, as shown in Fig. 3 for the type-I (BA)-(ABCA) stacking and the type-II (CA)-(AB)(AB) stacking at $\theta = 1.35^\circ$. Starting from the rigid superlattice without moiré coupling between FLGs and without the $\gamma_2$ hopping within FLGs, the moiré coupling, the $\gamma_2$ hopping, the corrugation effect, and the in-plane relaxation effect are included in the Hamiltonian successively.

Without moiré coupling, the low-energy bands of bFLG (tFLG) touch at the corner $\bar{K}$ ($\bar{K}'$) [see Fig. S1(b)] of the supercell BZ, and the bands of bFLG cross with those of tFLG, as seen in Figs. 4(a), 4(f) and S4. The band crossings can be eliminated by the moiré hopping between layers 1 and 1 in a way similar to TBG, as these degenerate states at crossings have contributions from these interface layers. In contrast to TBG always with touched valence and conduction bands at the BZ corners, state splitting occurs around the moiré coupling for TMLG. In the pristine bFLG, the states composed of non-dimer sites have zero energy at $\bar{K}$. These states are just the non-dimer basis functions with the momentum of $K_+$, which is at the corner of the bFLG BZ [see Fig. S1(a)]. When bFLG is coupled to tFLG, the energy of the non-dimer states $\bar{K}'$ in the first layer of bFLG rises above zero and the other non-dimer states still have zero energy. We will show that the positive energy of $|1B, K_+\rangle$ is due to the electron-hole asymmetry in low-energy bands of tFLG.

For the chiral tFLG whose zero-energy states are at $\bar{K}'$, three degenerate conduction states ($|\psi_{c,n}\rangle$ with $n = 1 - 3$) and three degenerate valence states ($|\psi_{v,n}\rangle$) have low energies ($\varepsilon_{c,n}$ and $\varepsilon_{v,n}$) at $\bar{K}$, as seen in Fig. S4. By second-order perturbation approximation, the energy ($\varepsilon_{1B}$) of $|1B, K_+\rangle$ can be expressed as

$$
\varepsilon_{1B} = \sum_{n=1}^{3} \left| \langle 1B, K_+ | \hat{H} | \psi_{c,n}\rangle \right|^2 - \varepsilon_{c,n} + \sum_{n=1}^{3} \left| \langle 1B, K_+ | \hat{H} | \psi_{v,n}\rangle \right|^2 - \varepsilon_{v,n}
$$

(1)

Considering only the interlayer hopping $\gamma_1$ between
layers. With the electron-hole symmetry, half of the squared norm of $|\langle \psi_{c,n} | H | \psi_{c,n} \rangle|$ decreases the same value ($\Delta_B, \Delta_{0}$) in layer 1 and $\tilde{1}$ produces the same value ($u$) of the Hamiltonian elements between $|1B, K_{+}\rangle$ and $|1\alpha, K_{+}\rangle$. For the BA stacked tFLG, $|\langle1B, K_{+}\rangle H | \psi_{c,n} \rangle$ becomes $u/\sqrt{2}$ as layer $\tilde{1}$ contributes half of the squared norm of $|\psi_{c,n}\rangle$. For other tFLG, $|\langle1B, K_{+}\rangle H | \psi_{c,n} \rangle$ decreases slowly with its number of layers. With the electron-hole symmetry, $\varepsilon_{v,n} = -\varepsilon_{c,n}$, then $\varepsilon_{1B}$ is still zero.

The interlayer hopping $\gamma_4$ between a non-dimer site and a dimer site introduces electron-hole asymmetry in the bands of FLGs. The supercell momentums of the basis functions expanding $|\psi_{c,n}\rangle$ and $|\psi_{c,n}\rangle$ all have the same length $k_0 \approx 4\pi\theta/3a$. With only $\gamma_1$ and $\gamma_4$ as well as the intralayer nearest-neighbor hopping ($-t_0$) for chiral tFLG with $N$ layers, $\varepsilon_{v,n}$ and $\varepsilon_{c,n}$ can be expressed analytically as\cite{47,48}

$$\varepsilon_{v,n} = -\left(\sqrt{3t_0 a k_0/2}\right)^N/\gamma_1^{N-1} + 3t_0 \gamma_4 a^2 k_0^2/(2\gamma_1),$$
$$\varepsilon_{c,n} = \left(\sqrt{3t_0 a k_0/2}\right)^N/\gamma_1^{N-1} + 3t_0 \gamma_4 a^2 k_0^2/(2\gamma_1),$$

which lead to $-\varepsilon_{v,n} < \varepsilon_{c,n}$. Numerical calculations show that $\varepsilon_{v,n}$ is also closer to zero than $\varepsilon_{c,n}$ for non-chiral FLGs. In addition, the norms of all the Hamiltonian elements in Eq. (1) are still approximately equal when including $\gamma_4$. Therefore, $\varepsilon_{1B}$ has a positive value. For the BA stacked tFLG, $\varepsilon_{1B}$ is given analytically by

$$\varepsilon_{1B} = \frac{u^2}{\theta^2} \frac{9\gamma_1}{8\pi^2 t_0^2} \left(\frac{1}{1-2\gamma_1/t_0} - \frac{1}{1+2\gamma_4/t_0}\right).$$

As $\gamma_4$ is much smaller than $t_0$, we have $\varepsilon_{1B} \approx \frac{9\gamma_1 \gamma_4 u^2}{(2\pi^2 t_0^3 \theta^2)}$.

At $K'$, similar analysis shows that the positive energy of the non-dimer state $|1B, K_{+}\rangle$ in layer 1 of tFLG can also be attributed to the electron-hole asymmetry in the bands of bFLG.

When only moiré coupling is included in the Hamiltonian of the (CA)-(AB)(AB) configuration with non-chiral bFLG, three states contributed by the non-dimer sites in the bottom three layers of bFLG still have zero energy at $K'$, as seen in Fig. 3(g). The turning on of the $\gamma_2$ hopping between the 2A and 4A sites can split these degenerate states, as shown in Fig. 3(h). Then only the $|3B, K_{+}\rangle$ state is located at almost zero energy.

For the rigid superlattice with all interlayer hopping, the flat bands around $E_F$ remain overlapped with other dispersive bands. The corrugation effect reduces such band overlapping by making the flat valence band narrower, as shown in Figs. 3(d) and 3(i). The in-plane relaxation has a more significant impact on the band dispersions. The flat valence band can become completely gapped from other bands with full relaxation, while $\Delta_0$ may be decreased by the in-plane relaxation, especially for configurations with a non-chiral FLG, as shown in Figs. 3(e) and 3(j).

To further demonstrate the importance of full relaxation for the emergence of isolated flat bands, the variations of $W_v$, $\Delta_h$ and $\Delta_0$ with $\theta$ considering both out-of-plane and in-plane relaxation are compared with those considering only corrugation in Fig. 4. For (BA)-(ABC) with only corrugation effect, $W_v$ is overestimated at $\theta$ around $1.1^\circ$, $\Delta_h$ is underestimated at all $\theta$, and $\Delta_0$ is also underestimated at small $\theta$. For (CA)-(AB)$(AB) only with full relaxation can $\Delta_h$ and $\Delta_0$ become positive at small $\theta$. The trends of these electronic properties with $\theta$ are also rather different for the two stackings. In particular, the maximum value of $\Delta_h$ is located at about $1.5^\circ$ for (BA)-(ABC), while it is at about $1.2^\circ$ for (CA)-(ABC). In addition, the first local minimum of $W_v$ is also at a larger $\theta$ for (BA)-(ABC).

We have focused on TMLG without potential differences between layers. It is noted that equal potential differences (\Delta) between adjacent layers produced by applied
FIG. 3. (Color online) The band structures and state composition for TMLG with the (BA)-(ABCA) stacking (a-e) and the (CA)-(AB)(AB) stacking (f-j) at $\theta = 1.35^\circ$. Starting from the rigid superlattice without moiré coupling between the FLGs and without the $\gamma_2$ hopping in the FLGs (a, e), the moiré coupling (b, g), the $\gamma_2$ hopping terms (c, h), the corrugation effect (d, i), and the in-plane relaxation effect (e, j) are included in the Hamiltonian successively. The band energies are directly calculated by diagonalizing the Hamiltonian without energy shifting, so the Fermi levels do not necessarily lie at the zero energy here.

out-of-plane electric field may separate crossed bands locally for some configurations, while only a rather small $\Delta$ can enhance the gaps around the isolated flat bands slightly and a larger $\Delta$ tends to close these global gaps, as shown for example in Fig. S5 for the (BA)-(ABCA) and (BA)-(ABCA) configurations.

IV. VALLEY CHERN NUMBERS AND ORBITAL MAGNETIC MOMENTS

At an odd filling of an isolated flat band, spontaneous valley polarization may occur due to the electron-electron interaction. If such a valley polarized band hosts a non-zero Chern number, the TMLG can support QAHC. The Chern numbers $C$ of flat bands in the $\xi = +$ valley have been obtained explicitly by integral of the Berry curvature ($\Omega_z$) in the supercell BZ as detailed in the SM, and the $C$ of a band in the $\xi = -$ valley is just the opposite of that for $\xi = +$. Chern numbers are also calculated for separable flat bands which are separated from nearby bands by local gaps larger than 0.5 meV to characterize the dependence of $C$ on stacking orders of TMLG.

We also focus on the Chern numbers ($C_v$) of flat valence bands in the $\xi = +$ valley. Among all configurations with $M + N$ layers, the largest $|C_v|$ is $M + N - 1$. The largest $|C_v|$ can occur in certain configurations with isolated flat bands, such as those shown in Figs. 1(b) and 1(d) and listed in Table SI. We note that the isolated flat valence band in a configuration with 10 layers [see Fig. 1(d)] has $C_v = -9$, which is the largest magnitude of $C_v$ for all considered TMLG with $N + M \leq 10$.

Systematic calculations of all configurations with different stackings and $\theta$ show that most flat valence bands with high $C_v$ slightly overlap with other bands and such band overlapping is related to the layers numbers ($M$ and $N$) of FLGs in TMLG, as illustrated in Fig. 5. The appearance of highest $C_v$ generally becomes less likely with increasing $N+M$. For $N + M = 6$, more cases with $|C_v| = 5$ appear for $M = 4$ than for $M = 3$ as type-II stacking is only possible for $M = 4$. In contrast, there are much fewer cases with the largest $|C_v|$ for $M = 6$ than for $M = 5$ when $M + N = 10$, which is related to the occurrence of type-III configurations only for $M = 5$. We notice that it is mainly the overlapping of the va-
The highest valence band states around \( \bar{K} \) in the pristine FLGs, and the sign of the Berry phases is fixed by the fact that the valence state at \( \bar{K} \) is mainly contributed by the chiral bFLG (tFLG), whose \( \Omega_\nu \) can also be already present at large \( \theta \), such as the (BA)-(ABCA)(CB) stacking with \( C_v = -7 \) [see Fig. S6(a)] for \( \theta \) from 1.89° to 1.47° at which the flat valence band becomes isolated.

In the weak coupling regime for (CA)-(AB)(AB), the \( \Omega_\nu \) peaks around \( K' \) originating from tFLG contribute +1 to \( C_v \), while there are both positive and negative peaks around \( K \) as bFLG is non-chiral, then \( C_v \) has a relatively small value of 2. With decreasing of \( \theta \), dipole-like pairs of positive and negative peaks form and band inversion between the middle bands occur at about 1.30°.
leading to the largest $C_v$ of 5. The next topological phase transition occurs at about 1.16°. We note that full relaxation is required to drive the topological phase transition with $C_v$ rising to 5 in the strong coupling regime, as shown in Fig. 4(d). The $C_v = -9$ for the (CBA)(BA)-(AB)(AB) stacking shown in Fig. 1(e) also occurs at small $\theta$, and the variation of $\Omega_z$ maps with $\theta$ can be seen in Fig. S6(b).

The non-trivial topology in isolated flat bands suggests that spontaneous orbital magnetic moments ($m$) may be observed at odd fillings of the flat bands. We evaluate the magnitude of $m$ contributed by the isolated flat valence band by setting the chemical potential at the middle of the $\Delta_0$ gap. The magnitude of $m$ can reach 10$\mu_B$ per supercell for $\theta$ around 1.1°, as shown in Fig. 7(a). For configurations with positive $m$, its value is related to the layer numbers of TMLG with $N+M = 8$ cases generally having the largest $m$ at a $\theta$. $m$ is roughly proportional to $C_v$, while the spanning of its value is still rather large for a given $C_v$, especially for small $|C_v|$. We note that for certain cases with a small $|m|$ but a large $|C_v|$, the sign of the total orbital magnetic moment may be inverted by tuning the chemical potential across the gap.

V. SUMMARY AND CONCLUSIONS

For TMLG with $M+N$ layers, full relaxation has been performed for the $2^{M+N-3}$ inequivalent stacking orders at varying $\theta$. Isolated flat bands emerge in relaxed TMLG with $M+N$ up to 10 and with various stackings, and most of them are on the hole side. The stacking orders that host isolated flat bands can be categorized into four types based on the electronic behavior of flat bands and the stacking decomposition. For type-I configurations with both chiral FLGs, the touched bands of FLGs around $E_F$ are split by the moiré coupling through the electron-hole asymmetry in low-energy bands of FLGs while the hopping $\gamma_2$ between the next-nearest layers is further required for the state splitting in other types with non-chiral FLGs. The corrugation effect reduces the band overlapping around $E_F$ and the full structural relaxation leads to global gaps that completely isolate a flat band. For TMLG with given $M$ and $N$, the highest Chern number $|C|$ of the separable flat bands reaches $M+N-1$ and can be hosted by certain isolated bands. The $|C| = 9$ occurs in the isolated flat valence band of several configurations with 10 layers. Such high $|C|$ originates from the lifting of the band-state degeneracy in the weak regime of moiré coupling or from the topological phase transitions induced by the strong moiré coupling. Moreover, large orbital magnetic moments $m$ arise in isolated flat bands with high $|C|$ and depend on the structural configurations.
Orbital magnetic moment ($\mu_B$)

FIG. 7. (Color online) (a) The orbital magnetic moments ($m$) contributed by the isolated flat valence band for configurations with different layer numbers ($M+N$). (b) $m$ versus $C_v$ for all configurations with isolated flat valence bands.

ACKNOWLEDGMENTS

We gratefully acknowledge valuable discussions with D. Tománek, H. Xiong, and S. Yin. This research was supported by the National Natural Science Foundation of China (Grants No. 11974312 and No. 11774195). The calculations were performed on TianHe-1(A) at National Supercomputer Center in Tianjin.

* E-mail: xqlin@zjut.edu.cn

1 R. Bistritzer and A. H. MacDonald, “Moiré bands in twisted double-layer graphene,” Proc. Natl. Acad. Sci. U.S.A. 108, 12233 (2011).

2 J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto, “Continuum model of the twisted graphene bilayer,” Phys. Rev. B 86, 155449 (2012).

3 S. Fang and E. Kaxiras, “Electronic structure theory of weakly interacting bilayers,” Phys. Rev. B 93, 235153 (2016).

4 G. Tarnopolsky, A. Jura Kruchkov, and A. Vishwanath, “Origin of Magic Angles in Twisted Bilayer Graphene,” Phys. Rev. Lett. 122, 106405 (2019).

5 Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. Y. Luo, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, R. C. Asghoori, and P. Jarillo-Herrero, “Correlated insulator behaviour at half-filling in magic-angle graphene superlattices,” Nature 556, 80 (2018).

6 Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, “Unconventional superconductivity in magic-angle graphene superlattices,” Nature 556, 43 (2018).

7 X. Lu, P. Stepanov, W. Yang, M. Xie, M. A. Aamir, I. Das, C. Urgell, K. Watanabe, T. Taniguchi, G. Zhang, A. Bachtold, A. H. MacDonald, and D. K. Efetov, “Superconductors, orbital magnets, and correlated states in magic angle bilayer graphene,” Nature 574, 653 (2019).

8 Y. Xie, B. Lian, B. Jäck, X. Liu, C.-L. Chiu, K. Watanabe, T. Taniguchi, B. A. Bernevig, and A. Yazdani, “Spectroscopic signatures of many-body correlations in magic-angle twisted bilayer graphene,” Nature 572, 101 (2019).

9 A. Kerelsky, L. J. McGilly, D. M. Kennes, L. Xian, M. Yankowitz, S. Chen, K. Watanabe, T. Taniguchi, J. Hone, C. Dean, A. Rubio, and A. N. Pasupathy, “Maximized electron interactions at the magic angle in twisted bilayer graphene,” Nature 572, 95 (2019).

10 Y. Jiang, X. Lai, K. Watanabe, T. Taniguchi, K. Haule, J. Mao, and E. Y. Andrei, “Charge order and broken rotational symmetry in magic-angle twisted bilayer graphene,” Nature 573, 91 (2019).

11 A. Uri, S. Grover, Y. Cao, J.A. Crosse, K. Bagani, D. Rodan-Leegrain, Y. Myasoedov, K. Watanabe, T. Taniguchi, P. Moon, M. Koshino, P. Jarillo-Herrero, and E. Zeldov, “Mapping the twist-angle disorder and Landau levels in magic-angle graphene,” Nature 581, 47
K. P. Nuckolls, M. Oh, D. Wong, B. Lian, K. Watanabe, T. Taniguchi, B. A. Bernevig, and A. Yazdani, “Strongly correlated Chern insulators in magic-angle twisted bilayer graphene,” Nature 588, 610 (2020).

E. Y. Andrei and A. H. MacDonald, “Graphene bilayers with a twist,” Nat. Mater. 19, 1265 (2020).

E. Y. Andrei, D. K. Efetov, P. Jarillo-Herrero, A. H. MacDonald, K. F. Mak, T. Senthil, E. Tutuc, A. Yazdani, and A. F. Young, “The marvels of moiré materials,” Nat. Rev. Mater. 6, 201 (2021).

Z. Song, Z. Wang, W. Shi, G. Li, C. Fang, and B. A. Bernevig, “All magic angles in twisted bilayer graphene are topological,” Phys. Rev. Lett. 123, 036401 (2019).

H. C. Po, L. Zou, T. Senthil, and A. Vishwanath, “Faithful tight-binding models and fragile topology of magic-angle bilayer graphene,” Phys. Rev. B 99, 155455 (2019).

J. Ahn, S. Park, and B.-J. Yang, “Failure of nielsen-ninomiya theorem and fragile topology in two-dimensional systems with space-time inversion symmetry: Application to twisted bilayer graphene at magic angle,” Phys. Rev. X 9, 021013 (2019).

J. Liu, J. Liu, and X. Dai, “Pseudo landau level representation of twisted bilayer graphene: Band topology and implications on the correlated insulating phase,” Phys. Rev. B 99, 155415 (2019).

N. Bultinck, S. Chatterjee, and M. P. Zuzelte, “Mechanism for Anomalous Hall Ferromagnetism in Twisted Bilayer Graphene,” Phys. Rev. Lett. 124, 166601 (2020).

Y.-H. Zhang, D. Mao, and T. Senthil, “Twisted bilayer graphene aligned with hexagonal boron nitride: Anomalous Hall effect and a lattice model,” Phys. Rev. Research 1, 033126 (2019).

J. Liu and X. Dai, “Anomalous Hall effect, magneto-optical properties, and nonlinear optical properties of twisted graphene systems,” npj Comput. Mater. 6, 57 (2020).

X. Lin and J. Ni, “Symmetry breaking in the double moiré superlattices of relaxed twisted bilayer graphene on hexagonal boron nitride,” Phys. Rev. B 102, 035441 (2020).

T. Cea, P. A. Pantaleón, and F. Guinea, “Band structure of twisted bilayer graphene on hexagonal boron nitride,” Phys. Rev. B 102, 155136 (2020).

X. Lin, K. Su, and J. Ni, “Misalignment instability in magic-angle twisted bilayer graphene on hexagonal boron nitride,” 2D Mater. 8, 025025 (2021).

J. Shi, J. Zhu, and A. H. MacDonald, “Moiré commensurability and the quantum anomalous Hall effect in twisted bilayer graphene on hexagonal boron nitride,” Phys. Rev. B 103, 075122 (2021).

D. Mao and T. Senthil, “Quasiperiodicity, band topology, and moiré graphene,” Phys. Rev. B 103, 115110 (2021).

M. Serlin, C. L. Tschirhart, H. Polshyn, Y. Zhang, J. Zhu, K. Watanabe, T. Taniguchi, L. Balents, and A. F. Young, “Intrinsic quantized anomalous Hall effect in a moiré heterostructure,” Science 367, 900 (2020).

A. L. Sharpe, E. J. Fox, A. W. Barnard, J. Finney, K. Watanabe, T. Taniguchi, M. A. Kastner, and D. Goldhaber-Gordon, “Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene,” Science 365, 605 (2019).

G. Chen, A. L. Sharpe, E. J. Fox, Y.-H. Zhang, S. Wang, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, Z. Shi, T. Senthil, D. Goldhaber-Gordon, Y. Zhang, and F. Wang, “Tunable correlated Chern insulator and ferromagnetism in a moiré superlattice,” Nature 579, 56 (2020).

J. Y. Lee, E. Khalaf, S. Liu, X. Liu, Z. Hao, P. Kim, and A. Vishwanath, “Theory of correlated insulating behaviour and spin-triplet superconductivity in twisted double bilayer graphene,” Nat. Commun. 10, 5333 (2019).

Y.-H. Zhang, D. Mao, Y. Cao, P. Jarillo-Herrero, and T. Senthil, “Nearly flat Chern bands in moiré superlattices,” Phys. Rev. B 99, 075127 (2019).

J. Liu, Z. Ma, J. Gao, and X. Dai, “Quantum Valley Hall Effect, Orbital Magnetism, and Anomalous Hall Effect in Twisted Multilayer Graphene Systems,” Phys. Rev. X 9, 031021 (2019).

M. Koshino, “Band structure and topological properties of twisted double bilayer graphene,” Phys. Rev. B 99, 235406 (2019).

N. R. Chebrolu, B. L. Chittari, and J. Jung, “Flat bands in twisted double bilayer graphene,” Phys. Rev. B 99, 235417 (2019).

X. Lin, H. Zhu, and J. Ni, “Pressure-induced gap modulation and topological transitions in twisted bilayer and twisted double bilayer graphene,” Phys. Rev. B 101, 155405 (2020).

A. Vela, M. V. O. Moutinho, F. J. Culchac, P. Venezuela, and R. B. Capaz, “Electronic structure and optical properties of twisted multilayer graphene,” Phys. Rev. B 98, 155135 (2018).

T. Cea, N. R. Walet, and F. Guinea, “Twists and the electronic structure of graphene materials,” Nano Lett. 19, 8683–8689 (2019).

G. A. Tritsaris, S. Carr, Z. Zhu, Y. Xie, S. B. Torrisi, J. Tang, M. Matthäakis, D. T. Larson, and E. Kaxiras, “Electronic structure calculations of twisted multi-layer graphene superlattices,” 2D Mater. 7, 035028 (2020).

S. Zhang, B. Xie, Q. Wu, J. Liu, and O. V. Yazyev, “Chiral decomposition of twisted graphene multilayers with arbitrary stacking,” arXiv:2012.11964.

J. Cao, M. Wang, C.-C. Liu, and Y. Yao, “Ab initio four-band Wannier tight-binding model for generic twisted graphene systems,” arXiv:2012.02575.

Z. Ma, S. Li, M.-M. Xiao, Y.-W. Zheng, M. Lu, H. Lu, J.-H. Gao, and X. C. Xie, “Moiré flat bands of twisted few-layer graphite,” arXiv:2001.07995.

N. N. T. Nam and M. Koshino, “Lattice relaxation and energy band modulation in twisted bilayer graphene,” Phys. Rev. B 96, 075311 (2017).

H. Yoo, R. Engelke, S. Carr, S. Fang, K. Zhang, P. Cazeaux, S. H. Sung, R. Hovden, A. W. Tseng, T. Taniguchi, K. Watanabe, G.-C. Yi, M. Kim, M. Luskin, E. B. Tadmor, E. Kaxiras, and P. Kim, “Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene,” Nat. Mater. 18, 448 (2019).

P. Lucignano, D. Alfè, V. Cataudella, D. Ninno, and G. Cantele, “Crucial role of atomic corrugation on the flat bands and energy gaps of twisted bilayer graphene at the magic angle θ ∼ 1.08°,” Phys. Rev. B 99, 195419 (2019).

F. Guinea and N. R. Walet, “Continuum models for twisted bilayer graphene: Effect of lattice deformation and hopping parameters,” Phys. Rev. B 99, 205134 (2019).

Y. W. Choi and H. J. Choi, “Intrinsic band gap and electrically tunable flat bands in twisted double bilayer graphene,” Phys. Rev. B 100, 201402 (2019).

M. Koshino and E. McCann, “Trigonal warping and berry’s phase Nπ in abc-stacked multilayer graphene,” Phys. Rev. B 80, 165409 (2009).
F. Zhang, B. Sahu, H. Min, and A. H. MacDonald, “Band structure of $abc$-stacked graphene trilayers,” Phys. Rev. B 82, 035409 (2010).

H. Min and A. H. MacDonald, “Chiral decomposition in the electronic structure of graphene multilayers,” Phys. Rev. B 77, 155416 (2008).

H. Min and A. H. MacDonald, “Electronic structure of multilayer graphene,” Progress of Theoretical Physics Supplement 176, 227 (2008).

J. Zhu, J.-J. Su, and A. H. MacDonald, “Voltage-Controlled Magnetic Reversal in Orbital Chern Insulators,” Phys. Rev. Lett. 125, 227702 (2020).

C. Repellin, Z. Dong, Y.-H. Zhang, and T. Senthil, “Ferromagnetism in narrow bands of moiré superlattices,” Phys. Rev. Lett. 124, 187601 (2020).