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Phys. Rev. B **101**, 125411 — Published 12 March 2020

DOI: [10.1103/PhysRevB.101.125411](https://doi.org/10.1103/PhysRevB.101.125411)

Topological flat bands without magic angles in massive twisted bilayer graphenes

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Twisted bilayer graphene (TBG) hosts nearly flat bands with narrow bandwidths of a few meV at certain *magic* twist angles. Here we show that in twisted gapped Dirac material bilayers, or massive twisted bilayer graphenes (MTBG), isolated nearly flat bands below a threshold bandwidth W_c are expected for continuous small twist angles up to a critical θ_c depending on the flatness of the original bands and the interlayer coupling strength. Narrow bandwidths of $W \lesssim 30$ meV are expected for $\theta \lesssim 3^\circ$ for twisted Dirac materials with intrinsic gaps of ~ 2 eV that finds realization in monolayers of gapped transition metal dichalcogenides (TMDC), silicon carbide (SiC) among others, and even narrower bandwidths in hexagonal boron nitride (BN) whose gaps are ~ 5 eV, while twisted graphene systems with smaller gaps of a few tens of meV, e.g. due to alignment with hexagonal boron nitride, show vestiges of the magic angles behavior in the bandwidth evolution. The phase diagram of finite valley Chern numbers of the isolated moire bands expands with increasing difference between the sublattice selective interlayer tunneling parameters. The valley contrasting circular dichroism for interband optical transitions is constructive near 0° and destructive near 60° alignments and can be tuned through electric field and gate driven polarization of the mini-valleys. Combining massive Dirac materials with various intrinsic gaps, Fermi velocities, interlayer tunneling strengths suggests optimistic prospects of increasing θ_c and achieving correlated states with large U/W effective interaction versus bandwidth ratios.

I. INTRODUCTION

The field of atomically thin two dimensional (2D) materials has seen an enormous progress over the last fifteen years since the successful experiments identifying the massless Dirac cones in graphene¹⁻³. In recent years the research directions have shifted towards tailoring the electronic structure of 2D material heterojunctions by selective material control and stacking arrangement⁴⁻⁷. New families of hexagonal 2D materials beyond graphene including group IV layered materials⁸⁻¹³, semiconducting and metallic transition metal dichalcogenides (TMDC)¹⁴⁻¹⁷ expand the list of the so called Dirac materials whose relevant band structure near the Fermi level is located at the K and K' Dirac points at the Brillouin zone corners. Modifications in the stacking arrangements are expected to introduce significant changes in the electronic structures of 2D materials assemblies. One prototypical example is the twisted bilayer graphene¹⁸⁻⁴⁵ where the relative rotation of one of the layers gives rise to new features in the electronic structure such as van Hove singularities and secondary Dirac cones^{46,47} that can be understood based on the associated moire bands^{25,31,34,48-50}. Observations of superconductivity⁵¹⁻⁵³ and correlated phases⁵⁴⁻⁵⁶ in twisted bilayer graphene have sparked a renewed interest in searching the properties of twisted bilayer graphene and flat bands in other types of twisted 2D materials. New proposals for flat bands in hybrid heterostructures include transition metal dichalcogenides⁵⁷⁻⁶⁰, hexagonal boron nitride bilayers^{61,62}, van der Waals patterned dielectric superlattices⁶³, multilayer graphene systems such as twisted bilayer graphene⁶⁴⁻⁶⁶, and rhombohedral trilayer graphene on hexagonal boron nitride where gate tunable correlated and superconducting phases have been observed experimentally^{67,68}. When the Dirac material layers are gapped by applying vertical electric fields^{64,69-74} or when the gaps are already open like in BN/BN bilayers⁶¹, the bandwidths remained reduced continuously for small twist angles rather

than only at a discrete set of magic angles like in twisted bilayer graphene³¹. In this work we investigate the behavior of the low energy bands bandwidth for different massive twisted bilayer graphene (MTBG) Hamiltonians and find that nearly flat bands below a threshold bandwidth W_c can be achieved for a continuous range of twist angles up to a critical angle θ_c that increases with the band gap size and with the reduction of the Fermi velocity. Our study indicates that low energy nearly flat bands can be expected in the vicinity of band edges for small enough twist angles in a variety of gapped materials including semiconducting TMDC, silicon carbide (SiC), and hexagonal boron nitride (BN) and other 2D materials twisted bilayers when they can be appropriately modeled by gapped Dirac Hamiltonians. This flexibility in the twist angle choice should enormously facilitate the preparation of twistronics devices to tailor the flat bands. Furthermore, we calculated the associated topological valley Chern numbers phase diagram that can lead to spontaneous quantum Hall phases in the absence of magnetic fields⁷⁵⁻⁷⁹ when the degeneracy of these bands are lifted under the effects of spin or valley polarising time reversal symmetry breaking perturbations^{65,69,80}, and whose signatures have been measured in recent experiments^{56,80,81}. The interband transition and circular dichroism in twisted gapped Dirac materials is studied as a function of twist angle, electric fields and stacking alignment between the layers to illustrate the tunability of the optical properties in the system.

The manuscript is organized as follows. We present in Sec. II the theoretical details of the Hamiltonian model used in Sec. III to analyze the bandwidth dependence as a function of Hamiltonian parameters. In Sec. IV we discuss the analytical solutions of the eigenvalues at the symmetry points of the moire Brillouin zone. Then in Sec. V we discuss the valley Chern number phase diagram, and in Sec. VI the circular dichroism of the interband transition oscillator strengths before we close the manuscript in Sec. VII with the summary and discussions.

FIG. 1. (Color online) Interband coherence leading to band isolation and flattening between gapped Dirac cones in the moire Brillouin zone (mBZ). (a) Real-space moire pattern and moire Brillouin zone showing the followed k -path at the mBZ of the K macrovalley. (b) Hyperbolic band dispersion $E(k) = (\Delta^2 + (v_F p)^2)^{1/2}$ that is parabolic at small p and linear at large momenta due to an intralayer band gap 2Δ . The rotation between the layers separates the bands in momentum space by $K_\theta = K\theta$ proportional to the twist angle. Flatter bands are expected for smaller Fermi velocities v_F and large Δ . A system with finite interlayer coupling $\omega_1 = \omega_3 = 0.01$ eV and $\omega_2 = 0.05$ eV illustrates the opening of a secondary gap δ_s near $\tilde{\Gamma}$, and avoided crossings δ_M near $\tilde{M}$, and suppressions in the bandwidth W . (c) Schematic representation of multiple rotated Dirac cones, gapped Dirac cones, and the generation of isolated flatbands in the presence of interlayer tunneling in the top row, and side view of actual band structure surface plots in the bottom row.

II. TWISTED GAPPED DIRAC HAMILTONIAN MODEL

The twisted gapped Dirac materials systems we consider are described based on the conventional assumption of the moire bands theory that the interlayer tunneling strength varies slowly with respect to the 2D position on an atomic scale^{25,31,34,82}. Our continuum Hamiltonian shares the same periodicity of the moire pattern and we employ the Bloch's theorem to formulate the effective low energy electronic structure in the moire Brillouin zone it defines, see Fig. 1(a). The moire bands theory used in graphene on graphene (G/G) and subsequent extension for more general material combinations including graphene on hexagonal boron nitride (G/BN) have been discussed in Ref.³⁴, where a recipe is proposed to inform the model Hamiltonian parameters from first principles calculations from the crystal Hamiltonian that goes beyond the two center approximation for the interatomic interactions. The associated continuum models are generalized to include additional diagonal and off-diagonal moire pattern terms to the Bistritzer-MacDonald model resulting in:

$$H_{\text{IMBG}}(\mathbf{k}) = \begin{pmatrix} h_{\theta/2}^t(\mathbf{k}) & T(\mathbf{r}) \\ T^\dagger(\mathbf{r}) & h_{-\theta/2}^b(\mathbf{k}) \end{pmatrix} \quad (1)$$

where the bottom and top layers are twisted symmetrically in opposite senses to preserve the orientation of the moire pattern

and therefore of the moire Brillouin zone (mBZ)

$$h_\theta^l(\mathbf{k}) = (v_F^l \hat{R}_{-\theta} \mathbf{p} + \mathbf{A}^l(\mathbf{r})) \cdot \boldsymbol{\sigma}_{xy} + V^l(\mathbf{r}) \mathbb{1} + \Delta^l(\mathbf{r}) \sigma_z \quad (2)$$

where $\mathbf{p} = \mathbf{k} - \mathbf{K}$ is centered at the $\mathbf{K}$ point of each rotating layer, the index l distinguishes which layer. The triangular moire patterns for the scalar and vector potentials are given by

$$V(\mathbf{r}) = V_0 + 2V_1 \text{Re}[e^{i\phi_V} f(\mathbf{r})], \quad (3)$$

$$\Delta(\mathbf{r}) = \Delta_0 + 2\Delta_1 \text{Re}[e^{i\phi_\Delta} f(\mathbf{r})], \quad (4)$$

$$\mathbf{A}(\mathbf{r}) = A_0 + 2A_1 \hat{z} \times \nabla \text{Re}[e^{i\phi_A} f(\mathbf{r})], \quad (5)$$

where $f(\mathbf{r}) = \sum_{m=1}^6 e^{i\mathbf{G}_m \cdot \mathbf{r}} (1 + (-1)^m)/2$ that respects the triangular symmetry is defined in terms of $\mathbf{G}_m = \hat{R}_{2\pi(m-1)/3} \mathbf{G}_1$ with $m = 1, \dots, 6$ the six first shell moire reciprocal lattice vectors generated rotating successively $\mathbf{G}_1 \simeq (0, 4\pi\theta/\sqrt{3}a)$ by an angle of $2\pi/3$ ^{34,83} is equivalent to a sum of three cosines with alternating first shell G-vectors⁸⁴

$$M(\mathbf{r}) = 2C \text{Re}[e^{i\phi} f(\mathbf{r})] = 2C \sum_{m=2,4,6} \cos(\mathbf{G}_m \mathbf{r} + \phi), \quad (6)$$

where $M(\mathbf{r})$ is a placeholder for the potentials in Eqs. (3,4) and the constants C and ϕ are defined for different materials in Table I. The operator $\hat{R}_\theta$ accounts for the rotation of the electrons in momentum space through small phase differences

between the top and bottom layers and is responsible for the chiral circular dichroism observed in TBG for twist angles in opposite senses^{85,86}. For the analysis presented in this work we neglect these small additional phases because their effect is generally small and we can obtain electron-hole symmetric bands some simple interlayer tunneling values.

We use the conventional form of the interlayer tunneling in the small angle approximation distinguishing the different sublattice resolved tunneling terms^{31,34}

$$T_{s,s'}(\mathbf{r}) = \sum_j e^{-i\mathbf{Q}_j \cdot \mathbf{r}} T_{s,s'}^j, \quad (7)$$

where the three $\mathbf{Q}_j$ vectors $\mathbf{Q}_0 = K\theta(0, -1)$ and $\mathbf{Q}_{\pm} = K\theta(\pm\sqrt{3}/2, 1/2)$ are proportional to twist angle θ and $K = 4\pi/3a$ is the Brillouin zone corner length of the Dirac material of lattice constant a where $a = 2.46 \text{ \AA}$ for graphene and $a = 3.51 \text{ \AA}$ for MoS_2 . The interlayer coupling matrices between the two rotated adjacent layers are generically given by

$$T^0 = \begin{pmatrix} \omega_1 & \omega_2 \\ \omega_2^* & \omega_3 \end{pmatrix}, \quad T^{\pm} = \begin{pmatrix} \omega_1 & \omega_2 e^{\mp i2\pi/3} \\ \omega_2^* e^{\pm i2\pi/3} & \omega_3 \end{pmatrix} \quad (8)$$

where we distinguish three different ω_1 , ω_2 and ω_3 interlayer sublattice tunneling obtained averaging over all possible stacking configurations and assume for simplicity that they are real values. The convention taken here for the T^j matrices assume an initial AA stacking configuration $\tau = (0, 0)$ ³⁴ and differs by a phase factor with respect to the initial AB stacking $\tau = (0, a/\sqrt{3})$ ³¹. We use a similar naming convention for the AA, AB and BA local stacking arrangements of the unit cell atoms also for the 60° or equivalently 180° degrees alignment of the layers where the two sublattice atoms of the top layer are switched regardless of choice for the rotation center.

The moire band Hamiltonians were modeled from existing parameters in the literature for G/G^{64,69}, TMDC/TMDC^{84,87}, and informed from first-principles calculation for SiC/SiC and BN/BN for this work. The parameter fitting procedure follows closely those of Ref.³⁴ while the details for SiC/SiC are presented in appendix.

III. BANDWIDTH PHASE DIAGRAM IN TWISTED GAPPED DIRAC MATERIALS

Here we show that the low energy moire bands in twisted gapped Dirac materials have narrow bandwidths for a continuous range of twist angles in contrast to the discrete set of magic angles in twisted bilayer graphene. Specifically we show that the bandwidth of the low energy moire bands remain extremely narrow below a threshold value W_c up to critical twist angle of θ_c which scales almost linearly with the intrinsic band gap 2Δ and interlayer tunneling $\omega_i = \omega$, and is inversely proportional to the Fermi velocity v_F . Hints of this behavior were observed in ABC-trilayer graphene on BN under a perpendicular electric field⁶⁹, in the evolution of bandwidth in twisted BN/BN systems^{61,62}, and more recently in twisted double bilayer graphene subject to electric fields⁷⁰⁻⁷⁴,

TABLE I. Lattice parameters, inter-site tunneling energies and band gaps of twisted gapped Dirac materials bilayer systems. The Hamiltonian parameters for G/G systems are from Refs. [69] for rigid and Ref. [64] for out of plane relaxed geometries. Most TMDC parameters are taken from Ref. [87] except for WSe_2 parameters from Ref. [84], and the BN and SiC parameters have been calculated from the LDA-DFT band structures for this work. The intralayer moire patterns have been considered when modeling bilayers of BN, SiC, and WSe_2 , and all energies are given in eV. The coefficients $C_{0\mu}, C_{\mu}$ are given in eV and angle ϕ_{μ} in degrees.

Monolayer parameters and interlayer tunneling						
Bilayer	a (Å)	$ t_o $	ω			2Δ
			ω_1	ω_2	ω_3	
G/G (rigid)	2.46	2.6	0.098			-
G/G (relaxed)	2.46	3.1	0.098	0.12	0.098	-
MoS_2/WS_2	3.195	1.1/1.37	0.01			1.66/1.79
$\text{MoS}_2/\text{MoS}_2$	3.193	1.1	0.01			1.66
WS_2/WS_2	3.197	1.37	0.01			1.79
BN/BN (0°)	2.48	2.5	0.178	0.147	0.078	4.58
BN/BN (60°)	2.48	2.5	0.208	0.148	0.078	4.59
SiC/SiC	3.06	1.7	0.165	0.413	0.063	2.383
$\text{WSe}_2/\text{WSe}_2$	3.32	1.261	0.0011	0.0	0.0097	1.2

Intralayer moire patterns						
i-j	A-A	B-B	A'-A'	B'-B'	A-B	A'-B'
BN/BN (0°)						
C_{0ii}	2.333	-2.247	2.333	-2.247	-	-
C_{ij}	0.0180	0.0098	0.0180	0.0098	0.0044	0.0044
ϕ_{ij}	-67.9	-77.9	67.9	77.9	-145	145
BN/BN (60°)						
C_{0ii}	2.368	-2.222	-2.222	2.368	-	-
C_{ij}	0.0027	0.0026	0.0026	0.0027	0.0059	0.0059
ϕ_{ij}	-85.8	72.91	72.707	-85.8	120.12	120.12
SiC/SiC						
C_{0ii}	1.319	-1.064	1.319	-1.064	-	-
C_{ij}	0.0426	-0.0085	0.0426	-0.0085	0.0	0.0
ϕ_{ij}	42.71	-26.56	-42.71	26.56	0.0	0.0
$\text{WSe}_2/\text{WSe}_2$						
C_{0ii}	1.2	0.0	1.2	0.0	-	-
C_{ij}	0.0068	0.0089	0.0068	0.0089	0.0	0.0
ϕ_{ij}	-89.7	-91.0	89.7	91.0	0.0	0.0

and twisted transition metal dichalcogenides⁵⁷⁻⁶⁰. To understand the behavior of the moire bands in our MTBG systems it is useful to review the behavior of the discrete set of magic angles in the Bistritzer-MacDonald model of twisted bilayer graphene where the band structure scaling parameter $\alpha = \omega/(\theta v_F K)$ relating the interlayer coupling strength ω with the twist angle θ and the Fermi velocity v_F was used to identify the angles where the Fermi velocity vanishes at the Dirac point³¹. This scaling parameter α summarizes the relationship between the system parameters indicating that flat bands can be achieved more easily for greater interlayer coupling ω , and for smaller Fermi velocities v_F and twist angles θ . Fig. 1(b) illustrates how these parameters defining α can affect the band structures. For instance, finite interlayer tunneling terms $\omega_i \neq 0$ introduce coherence between the moire bands opening a secondary δ_s gap at Γ which together with the primary δ_p gap at $\bar{K}$ allows the formation of isolated Chern

FIG. 2. (Color online) Bandwidth of the low energy valence flat bands as a function of twist angle in twisted gapped Dirac material bilayers as a function of the primary band gap magnitude for (a) massive twisted bilayer graphene, and (b) using parameters of other twisted gapped Dirac material bilayers listed in Table I. The material parameters for TMDC/TMDC corresponds to MoS_2/WS_2 heterobilayers. Traces of the magic angle bandwidth dips of gapless twisted bilayer graphene gradually disappear as the intralayer gap magnitudes become larger and they are almost inexistent already for gaps of the order of ~ 0.2 eV. The bands are electron-hole symmetric for equal interlayer tunneling $\omega_i = \omega$ and in the absence of intralayer moire patterns and if we neglect the twist angle dependent phases. We observe that the bandwidths remain flat for a greater range of twist angles when the intrinsic band gaps are large. The non-monotonic behavior of the SiC/SiC valence band W and lack of electron-hole symmetry (not shown) can be traced to the unequal ω_i values for the interlayer tunneling parameters.

bands, see Fig. 1(c). The bandwidth W can be defined as the energy difference of the band energy at $\tilde{\Gamma}$ and the band edge at $\tilde{K}$ or $\tilde{K}'$, and we can define δ_M as the avoided gap at the $\tilde{M}$ point located between $\tilde{K}$ and $\tilde{K}'$.

The linear relationship between ω and θ suggested by the structure of α for the first magic angle was confirmed by explicit bandwidth calculations that numerically satisfy the $\theta_m = C_m \omega / v_F$ relationship for the m^{th} magic angle both in twisted bilayer graphene⁶⁹ and for the first magic angle corresponding to $m = 1$ in the minimal model of twisted bi-bilayer graphene⁶⁴ confirming that the magic twist angles should increase together with interlayer coupling strength as expected from this scaling relation^{41,69}. These magic angles were defined as the θ values that give rise to bandwidth local minima rather than the angles where the effective Fermi velocity vanishes at the K -points of the mBZ³¹ since this definition would become ambiguous in twisted bi-bilayers or gapped Dirac materials whose band edges already have zero Fermi velocity.

The bandwidths generally increase with twist angles and decrease with the band gap as shown in Fig. 2, where we have represented the evolution of the bandwidth W with twist angle of gapped graphene with a finite Δ mass term. Similar evolutions of W are shown also for other gapped Dirac materials that we modeled through other system parameters listed in Table I that aims to capture the behavior of the band edges near the K -points, or the (macro)valleys of single layers, of

FIG. 3. (Color online) Critical twist angle values θ_c to achieve (a) $W = 0.02$ eV and (b) $W = 0.03$ eV as a function of the band gap 2Δ obtained from the numerical results, the analytical bandwidths obtained from the first harmonic approximation discussed in Sec. IV, and line fits of the numerical data in Eq. (9). Here $v_F^0 = 0.84 \times 10^6$ m/s is the LDA Fermi velocity in a single layer graphene.

a variety of materials comprising twisted bilayers of gapped G, TMDCs including MoS_2 and WS_2 , SiC, BN. While increase in bandwidth W is expected for large twist angles, a non monotonic behavior can be present depending on the details of the Hamiltonian like in our example of SiC/SiC low energy valence band. In all the materials considered showing monotonic increase of W with twist angle we observe that the critical twist angle θ_c required to achieve a given bandwidth W_c increases for increasing Δ and ω_i and decreasing v_F . Hence, we can normally expect that the bandwidth will remain smaller than W_c for all the angles below the critical θ_c . A plausible alternative definition of θ_c not used here would be the ratio $R_c = U/W_c$ between the Coulomb repulsion and bandwidth. The relationship between θ and the system parameters is captured for the twisted gapped Dirac bilayer model with a single tunneling parameter $\omega_i = \omega$ through

$$\theta = \frac{C_D |\Delta| + C_\omega |\omega| + C_W W}{|t_0|} \quad (9)$$

where we use the dimensionless constants of $C_D = 1.6$, $C_\omega = 26$, $C_W = 45$, and t_0 is the nearest neighbor hopping amplitude in a honeycomb lattice that can be related with the Fermi velocity of the Dirac Hamiltonian through $v_F = |t_0| \sqrt{3}a/2\hbar$. The Eq. (9) shows the interdependence of W with the system parameters θ , Δ , ω and can be used to determine the critical angle θ_c associated to a specific critical bandwidth W_c that we aim for. The Fig. 3 compares numerically calculated θ_c with approximate analytical forms discussed in Sec. IV and confirms the interdependence of the parameters in Eq. (9) relating the twist angle θ , interlayer coupling strength ω_i , the Fermi velocity v_F of each layer, and the staggered potential Δ between the sublattices within each layer that gives rise to the intrinsic gaps.

Our calculations show that the MTBG is advantageous for the generation of flat bands in comparison to TBG for two

FIG. 4. (Color online) (a) Evolution of the electronic structure of $\tilde{G}/\tilde{G}$ at $\theta = 1^\circ$ twist angle as a function of band gap $2\Delta = 0, 0.1, 0.5, 1, 3$ eV where we can observe an enhancement of the secondary isolation gap δ_s together with the increase in the primary gap δ_p . The band structures have been obtained using equal interlayer coupling parameters $\omega_i = \omega = 0.098$ eV. (b) Colormap of the bandwidth W phase diagram in the parameter space of ω and θ where we observe an expansion of the narrow bandwidth region (dark color) when Δ is increased. (c) Colormap of the isolation gap δ_s in ω and θ phase space where we expect to find isolated flat bands that are more susceptible to Coulomb interactions. The δ_s secondary gap is calculated as the energy difference between (minimum of E_{C_2}) - (maximum of E_{C_1}) and becomes negative when both bands overlap in energy. (d) Evolution of the energy spacing between the first three low lying electron-hole symmetric conduction or valence bands evaluated at $\bar{K}$. While the lowest energy band remains approximately constant for different twist angles we find a roughly linear growth in the energy level position with twist angle for the higher energy bands. Corresponding band gap is indicated in parenthesis of the legends. (e) Bandwidth versus twist angle in a log-log scale shows a bandwidth evolution of the form $W(\theta) \simeq b\theta^m$ where the $b(\theta)$, $m(\theta)$ parameters indicates the local evolution of W as a function of θ that can be fitted with lines in a log-log scale for sufficiently large twist angles, whereas the plateaus and non-monotonic behavior for small twist angles are limitations in numerical accuracy. The inset illustrates the almost linear relationship between 2Δ and critical θ_c corresponding to fixed values of bandwidth W_c . (f) Evolution of the slope $m(\theta)$ for the tangent line fits in panel (e) that decrease progressively for larger twist angles following the hyperbolic dispersion of a single gapped Dirac band. (g) Illustration of the rapid change in the bandwidth for nearly flat bands on the order of nano-eV for small variations in the twist angle of $\sim 0.1^\circ$.

important reasons. First there is no need to aim for a specific magic twist angle to achieve narrow bandwidths when the gaps are sufficiently large, and second the gap opening allows

to achieve flat bands for larger twist angles than in TBG where the structural stability of the moire pattern is greater⁸⁸. Flat bands for larger twist angles implies in turn that stronger cor-

related phases are in principle achievable because the moire Coulomb interactions $U \sim \theta e^2 / 4\pi\epsilon_r\epsilon_0 a$ are roughly proportional to twist angle, where e is the electron charge and a is the lattice constant of the individual triangular lattice.

This MTBG model is a well defined model that can illustrate the behavior of various gapped Dirac material combinations both for the small and large gap limits. In the small gap limit a massive graphene layer has experimental realization in graphene aligned with hexagonal boron nitride (G/BN)^{88–95} that opens a band gap of ~ 15 meV or larger depending on sample preparation methods. Likewise band gaps of the order of few tens of meV are expected in silicene, germanene layers due to intrinsic spin-orbit coupling effects⁹⁶. The massive twisted bilayer graphenes can also serve as approximate models for other gapped Dirac materials such as SiC, hBN and TMDC⁸⁷ type triangular lattice 2D materials.

The evolution of MTBG flat bands in massive twisted bilayer graphenes are clearly summarized in Fig. 4 where we show the resulting band structure, bandwidth and isolation gaps expected as a function of gap size 2Δ , twist angle θ , and interlayer coupling ω for a wide range of parameters that can describe several twisted gapped Dirac materials. In Fig. 4(a) we show the band structure from electron-hole symmetrized Bistritzer-MacDonald model of twisted bilayer graphene that uses a single interlayer tunneling parameter $\omega_i = \omega$ and removes the rotation phases in each graphene layer. The results are obtained for a fixed twist angle $\theta = 1^\circ$ and variable gap sizes between $2\Delta = 0$ till $2\Delta = 3$ eV where it is clearly shown that finite values of intralayer gaps creates a band gap at the primary Dirac point $\delta_p > 0$ and also generates a secondary isolation gap $\delta_s > 0$ whose magnitude increases progressively as 2Δ becomes larger. It is important that both primary and secondary gaps remain finite in order to isolate the low energy bands and allow a stronger effective Coulomb interaction. When intralayer band gaps are large enough the primary δ_p and secondary δ_s gaps open simultaneously in a larger system parameter space of ω and θ . In the limit of small intralayer gaps of a few tens of meV like in aligned G/BN structures this reduced bandwidth region concentrate around the magic angle lines in the parameter space and thus we can still identify vestiges of the zero gap limit. A progressive increase in the gap size alters the phase diagram of the bandwidth in ω vs θ space merging the discrete traces of magic angle lines into a larger area in the phase diagram expanding the parameter space where we can find finite secondary gaps δ_s . This opening of the secondary isolation gap δ_s even when all three interlayer tunneling parameters $\omega_i = \omega$ are the same. This broadening of the parameter space leads to the disappearance of the discrete magic angles and gives rise to a continuous range of twist angles with reduced bandwidth when the band gaps are larger than a few hundreds of meV. We have typically explored twist angles from 0.05° to 2.5° for $\tilde{G}/\tilde{G}$, where the tildes indicates the presence of gaps, and up to 5° for larger gap materials, and interlayer coupling ω values that span between 0 to 0.25 eV, over two times larger than $\omega \sim 0.1$ eV in bilayer graphene. For models with unequal ω_i tunneling values we define $\omega = \omega_2$ and keep the same fixed ratios of ω_2/ω_1 and ω_2/ω_3 when we scale the strength

of interlayer coupling for the different materials listed in Table I using $\omega/\omega_i = 1$ for rigid $\tilde{G}/\tilde{G}$, $\omega_2/\omega_1 = 1.22$ for relaxed $\tilde{G}/\tilde{G}$, $\omega_2/\omega_1 = 0.83$, $\omega_2/\omega_3 = 1.89$ for BN/BN(0°), $\omega_2/\omega_1 = 0.71$, $\omega_2/\omega_3 = 1.89$ for BN/BN(60°), and $\omega_2/\omega_1 = 2.5$, $\omega_2/\omega_3 = 6.5$ for SiC/SiC.

An additional effect we observe from the gap increase is the progressive flattening of the higher energy bands, where we observe bandwidths smaller than a few meVs giving rise to a practically discrete set of energy spectra as found in atomic or quantum dot levels when individual layer primary gaps assume values of a few eVs for twist angles around $\sim 1^\circ$. The twist angle dependence of these quasi-flat higher energy bands follows an approximately linear evolution with θ and is summarized in Fig. 4(d), where we show differences in the energy levels of $E_{21} \sim 40$ meV between the lowest two energy levels, and smaller than $E_{32} \sim 10$ meV for the energy spacing between the second and third levels when the twist angles are changed by $\sim 1^\circ$. The bandwidth evolution for the higher energy bands in MTBG for different gap sizes are shown in the appendix. Remarkable flattening of the bands down to numerical values of $W \sim 10^{-12}$ eV are represented in Fig. 4(e)-(g). For small twist angles they are widened rapidly with increasing twist angle as $W(\theta) \simeq b\theta^m$ with powers as large as $m \sim 25$ that gradually reduces with increasing twist angles approaching the $m \sim 1$ limiting behavior of hyperbolic bands in the small gap limit. This rapid compression in W for sufficiently small θ implies that the density of states around the average band energy $E_{av} \sim (E_{max} + E_{min})/2$ of a given band

$$D(E_{av}) = \int_{\text{mBZ}} \frac{d\mathbf{k}}{(2\pi)^2} \delta(E_{av} - E(\mathbf{k})) \propto \theta^2/W \propto \theta^{2-m} \quad (10)$$

will roughly be proportional to the moire Brillouin zone area that scales with θ^2 and is inversely proportional to the bandwidth W in the absence of van Hove singularities. According to this estimate we expect nearly flat bands prone to Coulomb interaction driven ordered phases whose bandwidths evolve with twist angle as $D(E_{av}) \propto \theta^{2-m}$ even if we do not have divergences associated to saddle points in the Fermi surface. The rapid enhancement in the joint density of states between the flat bands, when not limited by the broadening due to disorder, will more than compensate the decrease of the oscillator strengths in the flat bands to enhance the optical absorption in the system.

The twist angle dependence of the electronic structure and DOS for a variety of materials and the associated density of states (DOS) and local density of states (LDOS) are shown in Fig. 5. We notice that when the primary gap δ_p near charge neutrality and the secondary isolation gap δ_s are simultaneously present the low energy flat bands are isolated from the neighboring higher energy bands and they can acquire a well defined integer valley Chern number^{65,97,98}. As we commented earlier on, the electronic structure of large gap MTBG like in BN/BN bilayers whose individual layer gaps are above several eV shows that higher energy bands are also flattened. These high energy flatband states should be accessible through carrier gating techniques for marginally twisted BN bilayers when the electron densities for each moire band and the interband energy spacing between the contiguous flat

FIG. 5. (Color online) (a) Electronic band structures and the corresponding density of states showing the evolution of flat bands as a function of twist angle in gapped Dirac material bilayer systems. Here, the $\tilde{G}/\tilde{G}$ consist of bilayers whose single layer gap is $2\Delta = 0.1$ eV. We notice the flattening of higher energy bands as the gaps become larger. (b) The local density of states in the bilayer systems at $\theta = 1^\circ$, and (c) In the top panel we show the stacking resolved real space representation of the local density of states at the valence band van Hove singularity (VHS), and in the bottom panel we represent the local stacking configuration of the moiré pattern, where the blue lines delimit the LDOS plot area of the top panel, and the stacking types in gapped bilayers with 0° and 60° alignments.

bands are reduced.

A different perspective for the interdependence of bandwidth and system parameters in $\tilde{G}/\tilde{G}$ is seen in the parameter space of ω and θ in Fig. 6, and in the parameter space of Δ and θ in Fig. 17 in the appendix, where we can observe how the magic angle traces are erased for $2\Delta \gtrsim 0.2$ eV, consistent with the observations in Fig. 2. Apart from the obliteration of the magic angles, the increase in the single layer bandgap gives rise to a nonzero primary gap δ_p , generally smaller than

2Δ , that lifts the band degeneracy at charge neutrality, and opens a secondary isolation gap δ_s , as already noted earlier for the case of equal interlayer tunneling parameters $\omega_i = \omega$. We also observe that the increase of interlayer coupling allows to achieve flat bands of tBG at greater twist angles as noted in earlier works^{41,98}, suggesting that we can look for materials with stronger interlayer coupling as a pathway to achieve stronger correlated phases due to reduced moiré pattern period, as evidenced in our SiC/SiC proposal.

FIG. 6. (Color online) Phase diagram colormaps of the bandwidth W , secondary gap δ_s , primary gap δ_p , the U_{eff}/W ratio in $\tilde{G}/\tilde{G}$, MoS₂/WS₂ TMDC, BN/BN and SiC/SiC bilayers for variable parameters ω and θ . The intralayer gaps considered in $\tilde{G}/\tilde{G}$ make the bandwidths less sensitive to twist angle especially as the gaps increase. They are taken as $2\Delta = 0.1$ eV in the first row and shows how a gap introduces broadening in the magic angle lines that were sharply defined in the limit of zero intralayer gaps. Likewise, increase of interlayer tunneling strength generally favors the flattening of the bands. Here, $\omega = \omega_2$ is the y-axis, while the remaining ω_1 and ω_3 parameters are proportional to ω_2 with fixed ratios following Table I, as explained in the main text. The ω_2 values obtained from DFT for each system are indicated with a green horizontal line in each phase diagram. The threshold value of the $U_{\text{eff}}/W = 1$ ratio where Coulomb interactions are expected to be significant is indicated with a blue line. In systems with large band gaps we find suppressed bandwidths in a large parameter space of twist angle versus interlayer tunneling.

Since the primary gap δ_p near the charge neutrality and the secondary gap δ_s near the Γ point of the mBZ are important factors that can influence the onset of interaction-driven phase ordering, we have studied these isolation gaps in the parameter space of $(\theta, 2\Delta)$ and (θ, ω) for all the massive systems listed in Table I. Greater band isolation should reduce screening due to electrons in neighboring bands and therefore strengthen the effective Coulomb interactions. We have represented in Fig. 6

the U_{eff}/W ratio obtained using the screened Coulomb potential⁶⁴

$$U_{\text{eff}} = \frac{e^2}{4\pi\epsilon_r\epsilon_0 l_M} \exp(-l_M/\lambda_D) \quad (11)$$

where the moire length is $l_M = a/\theta$, and the Debye length $\lambda_D = 2\epsilon_0/e^2 D(\delta_p, \delta_s)$ uses the 2D density of states $D(\delta_p, \delta_s) = 4(|\delta_p|u(-\delta_p) + |\delta_s|u(-\delta_s))/(W^2 A_M)$ that assumes a value proportional to the band overlap ratio $\delta_{p/s}/W$ when $\delta_{p/s} < 0$, where $u(x)$ is the heaviside step function. We use the relative dielectric constant $\epsilon_r = 4$, and there are four valley-spin degenerate electrons per moire unit cell area $A_M = \sqrt{3}l_M^2/2$ for each filled moire band. The phase diagram for U_{eff}/W of gapped $\tilde{G}/\tilde{G}$ systems in the parameter space of θ and ω shows that for small gaps we can still identify traces of the magic angle lines present in gapless tBG⁶⁹ together with the suppression of the bandwidth and isolation through δ_p and δ_s gaps, whereas in large gapped systems we can find a larger continuous parameter space of narrow band widths with large U_{eff}/W ratio.

IV. ANALYTICAL EXPRESSIONS OF W , δ_s , δ_p AND δ_M .

Analytical expressions of the physical quantities relevant for describing the flat bands including the bandwidth W , the primary gap δ_p , the isolation gap δ_s and avoided gap δ_M can be obtained from the eigenvalues at the different symmetry points in the moire Brillouin zone by solving the truncated moire bands Hamiltonian in the first shell approximation^{31,34}. The electronic structure of twisted Dirac bilayers results from the coherence between the constituent layers, ranging from the perturbative weakly coupled regime for large twist angles to progressively stronger coupling at small twist angles where multiple momenta scattering is required in order to capture the electronic structure of the coupled bilayer. The truncation for the Hamiltonian that we use is therefore just appropriate to describe the systems with large enough twist angles where the narrow bandwidth starts to become more dispersive. For the analysis in this section we consider the minimal model of the gapped Dirac cones connected by the interlayer tunnelings matrices given in Eq. (8) and preserve the convenient electron-hole symmetry eliminating the rotation phases $e^{\pm i\theta/2}$ phases that accompany the twists in each layer and present our analysis for the conduction flat bands which can be equally applied for the valence bands. We assume for simplicity $\omega_1 = \omega_3$ in the tunneling terms and allow for a different ω_2 which enhances the isolation gap δ_s . We denote by $E_i(\mathbf{k})$ the i^{th} conduction band. By using $E_1(\mathbf{k})$ and $E_2(\mathbf{k})$ the band energies of the first and second conduction bands we can define

$$W = E_1(\tilde{\Gamma}) - E_1(\tilde{K}) \quad (12)$$

$$\delta_s \simeq E_2(\tilde{\Gamma}) - E_1(\tilde{\Gamma}) \quad (13)$$

$$\delta_p = 2E_1(\tilde{K}) \quad (14)$$

$$\delta_M = E_2(\tilde{M}) - E_1(\tilde{M}) \quad (15)$$

following the properties of the moire bands shown in Fig. 1(b). The $E_1(\tilde{K})$ conduction band minimum of the flat band is always at the $\tilde{K}$ point, whereas the maximum $E_1(\tilde{\Gamma})$ always

FIG. 7. (Color online) Schematic of high-symmetry points in the mBZ. The numbers 1, ..., 6 and 4' are the gapped Dirac cone indices, and the red and yellow circles mark the gapped Dirac points from two different layers. $E(\tilde{K})$ is evaluated from a 8-band model consists of four gapped Dirac cones in the dashed brown circle. $E(\tilde{\Gamma})$ is calculated from a 12-band model which includes six gapped Dirac cones inside the dotted blue square. $E(\tilde{M})$ can be obtained from two gapped Dirac cones in the green solid oval. The interlayer tunnelings among the contiguous cones are T^0 , $T^\pm$ as given in in Eq. (8), and we discuss the case $\omega_1 = \omega_3 \neq \omega_2$.

happens at the Γ point. Consequently, the bandwidth of the conduction band (W) and the primary gap (δ_p) are obtained from $E_1(\tilde{K})$ and $E_1(\tilde{\Gamma})$. The minimum of the second conduction band is $E_2(\tilde{\Gamma})$ for small θ , so the isolation gap is correctly given by Eq. (13) for sufficiently small twist angles when $E_2(\tilde{\Gamma})$ is a minimum. For large twist angles, the second conduction band minimum moves away from the $\tilde{\Gamma}$ point starting to deviate from Eq. (13) but this estimate can still be useful for discriminating the isolation of the flat bands. A comparison of our analytical expressions against numerically obtained δ_s is offered in the appendix. The avoided gap δ_M or the eigenenergies at $E_i(\tilde{M})$ give additional information about the band structure at the zone boundary of the moiré Brillouin zone.

In Fig. 7 we show the schematic of high-symmetry points in the mBZ which are used to obtain the analytical expressions, where the $j = 1, \dots, 6$ and $j = 4'$ odd and even indices are used to label the gapped Dirac cones of the bottom and top layers respectively. The eigenvalues $E_i(\tilde{K})$ are evaluated from an 8-bands model consisting of four gapped Dirac cones in the dashed brown circle, while the $E_i(\tilde{\Gamma})$ are calculated from a 12-bands model which includes six gapped Dirac cones inside the dotted blue square. The $E_i(\tilde{M})$ energies for the avoided gaps can be obtained from a 4-bands model of two intersecting gapped Dirac cones within the green oval in Fig. 7. The interlayer tunneling among the contiguous cones are T^0 , $T^\pm$ as given in Eq. (8), and here we limit the discussions to the case of $\omega_1 = \omega_3 \neq \omega_2$. The displacement between the contiguous Dirac points is $K_\theta = K\theta$, where $K = 4\pi/3a$ is the distance between Γ to K in the single layer BZ. The analytical resolution of the eigen energies and eigenvectors for moiré systems should also be useful to understand the topological properties of the band structures from point group symmetry considerations⁹⁹.

A. Calculation of $E(\tilde{K})$ from an 8×8 model

We consider an 8-bands model including only the four gapped Dirac cones inside the dashed circle given in Fig. 7 to derive the analytical form of $E(\tilde{K})$. Our approach is similar to the scheme used in Ref.³¹ to calculate the renormalized Dirac-point band velocity in TBG. The 8-bands model Hamiltonian connecting one gapped Dirac cone with three surrounding gapped cones reads

$$H_{8 \times 8}(\tilde{K}) = \begin{pmatrix} h^1(\tilde{K}) & T^0 & T^+ & T^- \\ T^0 & h^2(\tilde{K}) & 0 & 0 \\ T^+ & 0 & h^6(\tilde{K}) & 0 \\ T^- & 0 & 0 & h^{4'}(\tilde{K}) \end{pmatrix}, \quad (16)$$

whose diagonal blocks are gapped Dirac Hamiltonians

$$h^j(\mathbf{k}) = \begin{pmatrix} \Delta_j & \hbar v_F |\mathbf{k} - \mathbf{q}_j| e^{-i\theta_{\mathbf{k}-\mathbf{q}_j}} \\ \hbar v_F |\mathbf{k} - \mathbf{q}_j| e^{i\theta_{\mathbf{k}-\mathbf{q}_j}} & -\Delta_j \end{pmatrix}, \quad (17)$$

where $\mathbf{k}$ is the mBZ momentum, and $\theta_{\mathbf{k}}$ is the momentum orientation with respect to the k_x axis. The j is the gapped Dirac cone index, so $\mathbf{q}_j = \{q_{x,j}, q_{y,j}\}$ determines the center of the j -th Dirac cone. The eigenstate consists of four two-component spinors $\Psi^T = \{\psi_1^T, \psi_2^T, \psi_3^T, \psi_4^T\}$. We find that at the K point, the spinors for the first conduction band always follows the relations $\psi_2 = \mathcal{U} \psi_4$, $\psi_3 = \mathcal{U}^* \psi_4$, where $\mathcal{U}$ is a diagonal matrix given by

$$\mathcal{U} = \begin{pmatrix} 1 & 0 \\ 0 & e^{i2\pi/3} \end{pmatrix}. \quad (18)$$

The discussions for the valence bands are closely similar and they are shown in the appendix. Consequently, the first conduction band energy at the $\tilde{K}$ point is given by

$$E(\tilde{K}) = \frac{\Delta}{3} + \frac{2}{3} \sqrt{4\Delta^2 + 3\rho_\theta^2 + 9\omega_1^2 + 9\omega_2^2} \cos\left(\frac{2\pi - \varphi}{3}\right), \quad (19)$$

where

$$\rho_\theta \equiv \hbar v_F K \theta$$

$$\varphi = \arccos \frac{8\Delta^3 + 9\Delta\rho_\theta^2 - 54\Delta\omega_1^2 + 27\Delta\omega_2^2}{(4\Delta^2 + 3\rho_\theta^2 + 9\omega_1^2 + 9\omega_2^2)^{3/2}}.$$

For $\omega_1 = \omega_3$, the twisted gapped Dirac bilayer model given in Eq. (1) preserves electron-hole symmetry in the absence of the aforementioned rotation phases, and the lowest band edges reside at the $\tilde{K}$ point. Hence the resulting analytical expression of the primary gap is

$$\delta_p = 2E(\tilde{K}), \quad (20)$$

where $E(\tilde{K})$ is given by Eq. (19).

Due to the electron-hole symmetry, the first valence band energy at $\tilde{K}$ point $E^v(\tilde{K}) = -E(\tilde{K})$ is available from Eq. (19). For the valence band states at $\tilde{K}$ we can define the two-component spinors for the 8×8 model in a similar way to what we have done for the conduction band. The eigenstate consists of four two-component spinors $\Psi^T =$

FIG. 8. (Color online). Plots in the top row show the eigenvector phases of the (a) first and (b) second $E_i(\tilde{\Gamma})$ conduction bands, where the ϕ_i labels in the plot denote the eigenenergies $E_{\phi_i}(\tilde{\Gamma})$ for $\phi_1 = \{0, 2\pi/3\}$, $\phi_2 = \{2\pi/3, -2\pi/3\}$, and $\phi_3 = \{-2\pi/3, 0\}$. Plots in the second row show the differences between different $E_{\phi_i}(\tilde{\Gamma})$ in the space of Δ and θ . The parameters used in these calculations are $\omega_1 = \omega_2 = \omega_3 = 0.098$ eV and $t_0 = -2.6$ eV.

$\{\psi_1^T, \psi_2^T, \psi_3^T, \psi_4^T\}$, and the first valence band always follows the relations $\psi_2 = \mathcal{U}_v \psi_4$, $\psi_3 = \mathcal{U}_v^* \psi_4$, where $\mathcal{U}_v$ is a diagonal matrix given by

$$\mathcal{U}_v = \begin{pmatrix} e^{i2\pi/3} & 0 \\ 0 & 1 \end{pmatrix}. \quad (21)$$

B. Analytical expression of $E(\tilde{\Gamma})$

The eigenenergy at the $\tilde{\Gamma}$ point can be calculated from the 12×12 Hamiltonian consisting of six gapped Dirac cones inside the blue dotted square in Fig. 7. The interlayer tunneling between contiguous gapped Dirac cones are given by the T_0 , $T^\pm$ matrices, and the Hamiltonian reads

$$H_{12 \times 12}(\tilde{\Gamma}) = \begin{pmatrix} h^1(\tilde{\Gamma}) & T^0 & 0 & 0 & 0 & T^+ \\ T^0 & h^2(\tilde{\Gamma}) & T^- & 0 & 0 & 0 \\ 0 & T^- & h^3(\tilde{\Gamma}) & T^+ & 0 & 0 \\ 0 & 0 & T^+ & h^4(\tilde{\Gamma}) & T^0 & 0 \\ 0 & 0 & 0 & T^0 & h^5(\tilde{\Gamma}) & T^- \\ T^+ & 0 & 0 & 0 & T^- & h^6(\tilde{\Gamma}) \end{pmatrix}. \quad (22)$$

The eigenstate consists of six two-component spinors $\Psi^T = \{\psi_1^T, \psi_2^T, \psi_3^T, \psi_4^T, \psi_5^T, \psi_6^T\}$, and we find that the spinors always follow the relations $\psi_3 = \mathcal{V} \psi_1$, $\psi_5 = \mathcal{V}^* \psi_1$, $\psi_4 = \mathcal{V} \psi_2$, $\psi_6 = \mathcal{V}^* \psi_2$, and $\mathcal{V}$ is a diagonal matrix

$$\mathcal{V} = \begin{pmatrix} e^{i\eta_1} & 0 \\ 0 & e^{i\eta_2} \end{pmatrix} \quad (23)$$

where $\{\eta_1, \eta_2\}$ only have three different combinations, which are $\phi_1 : \{\eta_1 = 0, \eta_2 = 2\pi/3\}$, $\phi_2 : \{\eta_1 = 2\pi/3, \eta_2 = -2\pi/3\}$, and $\phi_3 : \{\eta_1 = -2\pi/3, \eta_2 = 0\}$. The eigenvalues for the valence bands at the $\tilde{\Gamma}$ point have electron-hole symmetry and are related to the conduction band energies through $E_{\phi_1}^v(\tilde{\Gamma}) = -E_{\phi_3}(\tilde{\Gamma})$, $E_{\phi_2}^v(\tilde{\Gamma}) = -E_{\phi_2}(\tilde{\Gamma})$ and $E_{\phi_3}^v(\tilde{\Gamma}) = -E_{\phi_1}(\tilde{\Gamma})$. Likewise the valence band eigenvectors can be defined with the same spinor structure for the conduction bands but using the ϕ_i phases that follow from this electron-hole symmetry of the eigenenergies. For each ϕ_i ($i=1,2,3$), the eigenenergy problem is then changed to solving a quartic equation

$$x^4 + \alpha x^2 + \beta x + \gamma = 0. \quad (24)$$

For the three ϕ_i sets, the coefficients in Eq. (24) become different. By solving Eq. (24) for each ϕ_i one can get 4 roots, among which two are positive and two are negative. We need the smaller positive root in our calculation, so for three ϕ_i s we obtain three different energy eigenvalues and denote them as $E_{\phi_i}(\tilde{\Gamma})$. The details of the calculation of $E_{\phi_i}(\tilde{\Gamma})$ are given in appendix, we use the following approximations

$$\begin{aligned} E_{\phi_1/\phi_3}(\tilde{\Gamma}) &\approx \sqrt{\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2} - \sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2} \\ &\quad \mp \frac{3\Delta\omega_1^2}{2\sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}, \\ E_{\phi_2}(\tilde{\Gamma}) &= \sqrt{\Delta^2 + \rho_\theta^2 + \omega_1^2 + 4\omega_2^2} - 2\sqrt{\Delta^2\omega_1^2 + 4\rho_\theta^2\omega_2^2 + 4\omega_1^2\omega_2^2}. \end{aligned} \quad (25)$$

We represent in Fig. 8 the ϕ_i parameters that determine the eigenstates of the first and second conduction flat bands. In

this figure the first conduction band energy is given by either

FIG. 9. (Color online) Effective mass m^*/m_e with respect to the rest mass of the electron as a function of 2Δ and θ for the massive twisted bilayer graphene. In the left panel, the effective mass is obtained from the numerical derivation of Hamiltonian in Eq. (1) at the band edge. In the (right) panel, we show line cuts that illustrate the divergence of the effective mass near a critical twist angle that depends on Δ , that for large enough twist angles when the band edges locate at $\tilde{K}$ and $\tilde{K}'$ approaches $m^* = \Delta/v_F^2$.

$E_{\phi_1}(\tilde{\Gamma})$ or $E_{\phi_2}(\tilde{\Gamma})$, while the second conduction band energy is given by either $E_{\phi_1}(\tilde{\Gamma})$, $E_{\phi_2}(\tilde{\Gamma})$ or $E_{\phi_3}(\tilde{\Gamma})$ depending on the Hamiltonian parameters Δ and θ . Consequently, the expressions of the bandwidth and the isolation gap are

$$\begin{aligned} W &= \min(E_{\phi_1}(\tilde{\Gamma}), E_{\phi_2}(\tilde{\Gamma})) - E(\tilde{K}), \\ \delta_s &= |\min(E_{\phi_2}(\tilde{\Gamma}), E_{\phi_3}(\tilde{\Gamma})) - E_{\phi_1}(\tilde{\Gamma})|. \end{aligned} \quad (26)$$

C. Analytical models for $E(\tilde{M})$

The moire bands in reciprocal space are repeated periodically by the moire reciprocal lattice vectors G_m . The coherence between the two overlapping bands of the rotated layers can be approximated in the simplest perturbative limit by a four by four Hamiltonian neglecting all but the smallest momenta coupling equivalent to two interacting gapped Dirac cones where the bands would otherwise intersect between $\tilde{K}$ and $\tilde{K}'$ as illustrated in Fig. 9.

$$H_{4 \times 4}(\tilde{M}) = \begin{pmatrix} h^1(\tilde{M}) & T^0 \\ T^0 & h^2(\tilde{M}) \end{pmatrix}. \quad (27)$$

The energy eigenvalue at the $\tilde{M}$ -point can be obtained by diagonalizing Eq. (27). For the simplest case when $\omega_1 = \omega_2 = \omega_3$, the two conduction band energies at the $\tilde{M}$ -point are given by

$$E(\tilde{M}) = \sqrt{\Delta^2 + (\rho_\theta/2)^2 + \omega_1^2} \pm |\omega_1|, \quad (28)$$

where $\rho_\theta = \hbar v_F K \theta$ from which we can conclude that the energy difference for the avoided gap is

$$\delta_M = 2|\omega_1|. \quad (29)$$

If $\omega_1 = \omega_3 \neq \omega_2$, the $\tilde{M}$ -point conduction band energy eigenvalues are

$$\begin{aligned} E(\tilde{M}) &= \\ &\sqrt{\Delta^2 + (\rho_\theta/2)^2 + \omega_1^2 + \omega_2^2 \pm 2\sqrt{\Delta^2 \omega_1^2 + (\rho_\theta/2)^2 \omega_2^2 + \omega_1^2 \omega_2^2}}. \end{aligned} \quad (30)$$

In the limit of large Δ , one can approximate $E(\tilde{M})$ to obtain

$$\delta_M \approx 2 \frac{\sqrt{\Delta^2 \omega_1^2 + (\rho_\theta/2)^2 \omega_2^2 + \omega_1^2 \omega_2^2}}{\sqrt{\Delta^2 + (\rho_\theta/2)^2 + \omega_1^2 + \omega_2^2}}, \quad (31)$$

which further simplifies to

$$\delta_M \approx |\omega_1| + |\omega_2| \quad (32)$$

if we assume $\omega_2 = \omega + \delta\omega$, $\omega_1 = \omega - \delta\omega$ and $\delta\omega \ll \omega_1, \omega_2$.

The above Hamiltonian and associated solutions can be further simplified to a 2×2 model in the limit of large band gaps thanks to the almost perfect sublattice polarization of the conduction and valence bands. In this limit, we can build a two by two Hamiltonian that couples two conduction or two valence mutually coupled through a single tunneling parameter ω , and in the presence of an interlayer potential difference V_g we have

$$\hat{H}_{2 \times 2}(k) = \begin{pmatrix} \varepsilon_{c(v)}(k + \frac{K_\theta}{2}) + \frac{V_g}{2} & \omega \\ \omega^* & \varepsilon_{c(v)}(k - \frac{K_\theta}{2}) - \frac{V_g}{2} \end{pmatrix} \quad (33)$$

that couples through ω the two conduction or valence band edges $\varepsilon(k) = \varepsilon_c(k) = -\varepsilon_v(k) = (\Delta^2 + (\hbar v_F(k))^2)^{1/2}$ arising from the top and bottom layers whose bands represented along the line that connects the two shifted moire zone $\tilde{K}$ -points are shown in Fig. 9 for $V_g = 0$. By denoting $\varepsilon^\pm(k) = \varepsilon(k \pm K_\theta/2)$ for the band edges we obtain the following eigenvalues

$$E_{c(v)}(k) = \left(\frac{\varepsilon_{c(v)}^-(k) + \varepsilon_{c(v)}^+(k)}{2} \right) \pm \sqrt{|\omega|^2 + \left(\frac{\varepsilon_{c(v)}^-(k) - \varepsilon_{c(v)}^+(k) - V_g}{2} \right)^2} \quad (34)$$

Keeping in mind that at the $\tilde{M}$ point $\varepsilon^+(0) = \varepsilon^-(0) = \varepsilon(\pm K_\theta/2)$ the avoided gap reduces to

$$\delta_M = 2|\omega| \quad (35)$$

in agreement with Eq. (29) and Eq. (32). The effective mass m^* of the electron given by

$$m^* = \hbar^2 / \frac{\partial^2 E(k)}{\partial k^2} \quad (36)$$

has an analytical approximation near $(\tilde{K}, \tilde{K}')$ given by,

$$m^* = \frac{\varepsilon(k)^3}{\Delta^2 v_F^2} \quad (37)$$

that reduces to the K point value $m^* = \Delta/v_F^2$ of a single gapped Dirac cone in the limit of large twist angles¹⁰⁰. The numerical m^* from the low energy bands are represented in Fig. 9 as a function of gap and twist angle.

V. TOPOLOGICAL FLAT BANDS AND VALLEY CHERN NUMBERS

Isolated bands in the moire Brillouin zone can develop integer valley Chern numbers different from zero depending on the details of the Hamiltonian^{69,97,101}. A finite charge Hall conductivity will develop when the valley degeneracy is lifted by valley polarization in an otherwise time reversal invariant system^{65,98}. This type of selective valley and spin filling mechanisms have been proposed to underlie the observed ferromagnetism and anomalous Hall effects in twisted bilayer graphene aligned with a boron nitride substrate^{56,81}, and the ordered phases in twisted double bilayer graphene⁷⁰⁻⁷³. On a closely related topic for our work, opposite valley Chern numbers of $C = \pm 1$ have been proposed in twisted bilayer graphene with a small gap of ~ 15 meV in one of the layers aligned with the hexagonal boron nitride substrate^{65,102} while Coulomb interactions could trigger $C = \pm 2$ low energy bands even in the absence of substrate effects¹⁰³. Analogous valley Chern bands due to intralayer moire patterns have also been proposed in twisted semiconducting TMDC⁸⁴, mosaics of local valley Hall topological order¹⁰⁴ and moire Berry curvatures¹⁰⁵. Recent observations of quantized anomalous Hall conductance at zero magnetic field in TBG⁸¹ or non quantized orbital moments⁵⁶, and at small magnetic fields of ~ 0.4 T in ABC trilayer graphene on hexagonal boron nitride in the presence of electric field induced gaps of ~ 20 meV⁸⁰ suggest optimistic prospects of finding exotic zero magnetic field quantum Hall states when the device qualities are sufficiently improved.

In the following we show in Fig. 10 the valley Chern number phase diagram expected in MTBG for the first conduction and valence bands expanding beyond the parameter region studied in earlier related work^{65,98,102} showing the interdependence of the mass terms with other system parameters. Further analysis of the Chern number phase diagrams for different parameter sets Δ , v_F , θ , ω_i ($i=1,2,3$) and V_g are shown in 18 in the appendix. We calculate numerically the valley

FIG. 10. (Color online) Valley Chern number phase diagram of massive twisted bilayer graphene as a function of the mass terms Δ_t and Δ_b for the top and bottom layers for the first conduction and valence bands obtained for interlayer coupling parameters $\omega_1 = \omega_2 = 0.85\omega$ with $\omega = 0.098$ eV and twist angles $\theta = 1^\circ$ and $\theta = 3^\circ$. The panels on the left are zoomed in for $|2\Delta| \leq 0.1$ eV to magnify the small gap regime. While the Chern numbers are $C_v = 0, \pm 1$ for most of the phase space we notice the presence of small regions where they assume $C_v = \pm 2$ values. For large Δ values, the increase of twist angle helps to expand the parameter space region that turns the trivial low energy bands into Chern bands. We further show in the expanded Fig. 18 in the appendix that the phase space of non-trivial Chern bands increases with the differences in magnitude of the tunneling ω_i between different sublattices, and that the high energy bands undergo rapid variations in the Chern numbers indicating the multiple band crossings and reduced phase spaces where the gaps are well developed.

Chern numbers with valley index $v = \pm 1$ through the standard formula¹⁰⁶

$$C_v = \int_{\text{mBZ}} \mathbf{dk} \Omega_n(\mathbf{k}) / 2\pi \quad (38)$$

and using the Berry curvature of the n^{th} band

$$\Omega_n(\mathbf{k}) = -2 \sum_{n \neq n'} \text{Im} \left[\frac{\langle u_n | \frac{\partial H}{\partial k_x} | u_{n'} \rangle \langle u_{n'} | \frac{\partial H}{\partial k_y} | u_n \rangle}{(E_n - E_{n'})^2} \right] \quad (39)$$

where E_n and u_n are the eigenenergies and eigenvectors. The maxima of Berry curvatures often concentrate around the different symmetry points of the mBZ at $\tilde{K}$, $\tilde{K}'$ or $\tilde{\Gamma}$, and near the moire Brillouin zone boundaries where the avoided gaps are formed. Details of Berry curvature plots can be found in Fig. 19 in the appendix. Some general conclusions we can anticipate from our calculations of the Chern number phase diagram of twisted gapped Dirac bilayers are that (i) the conduction and valence bands remain topologically trivial if $\omega_1 = \omega_2 = \omega_3$ and their relative magnitudes sensitively contribute in determining the phase space of topological bands, (ii) as the band gap of the system becomes larger we need to increase the twist angle θ to turn the lowest band into a Chern band, (iii) the intralayer moire patterns contribute in the determination of the Chern number phase diagram, and (iv) small values of interlayer bias comparable in magnitude

FIG. 11. (Color online) Chern number results for the first four conduction and valence bands calculated with respect to θ for a variety of twisted massive Dirac cone systems modeled through the parameters in Table. I. VB and CB indicate the results for the valence bands and conduction bands. The $\tilde{G}$ represents an intrinsically gapped graphene with $2\Delta = 0.01$ eV of comparable magnitude of the gap in G/BN, while G represents a gapless graphene. We find relatively wide twist angle regions where the Chern numbers are well quantized especially for the lower energy bands. When the bands are not well isolated the valley Chern numbers are not quantized to an integer value and can show rapid variations when the system parameter changes.

with the interlayer tunneling can lead to higher energy band crossings and changes in Chern number, as was seen in models of TMDC based on effective intralayer moire potentials⁵⁷, see Fig. 20 in the appendix.

In Fig. 10 we represent the Chern number phase diagrams for the first four conduction bands of two twisted gapped Dirac materials as a function of gap magnitudes. The phase diagrams for the valence bands are similar to that of conduction bands with opposite Chern numbers except for a small electron-hole symmetry breaking stemming mainly from unequal ω_i values, see Fig. 18 in the appendix for a more complete model parameter space sweep. By comparing the phase diagrams corresponding to different twist angles of $\theta = 1^\circ$ and $\theta = 3^\circ$ in Fig. 10's (a) and (b) panels respectively we find that shortening of the moire period by increasing θ can turn the low energy bands into Chern bands in large gap systems. This behavior can be explained by the fact that an increase in twist angles enlarges the mBZ area and prevents the Berry curvature weights at the low energy bands to be pushed to higher energy bands. We have shown results for the first valence and conduction bands for systems with different interlayer tunneling terms ω_1 , ω_3 and variable gap values Δ_b (Δ_t) of the bottom (top) layer for interlayer tunneling parameters of $\omega_1 = \omega_3 = 0.85\omega_2$ with $\omega = \omega_2 = 0.098$ eV. Nonzero valley Chern numbers result in two belt regions when either of the diagonal tunneling terms ω_1, ω_3 becomes different from the

off-diagonal ω_2 . This region becomes wider as the difference between ω_1 and ω_2 increases. We have verified that a finite Chern number can be obtained when any two ω_i terms assume unequal values. Hence, the low energy Chern bands are possible only when there are at least two different ω_i values coupling different sublattices in Eq. (8), and a sufficiently small finite mass term in at least one of the layers. The nonzero valley Chern band parameter region is generally larger in Δ_t, Δ_b space when the differences in the tunneling parameters are also large, as we can verify through the calculations in the appendix where we compare $\omega_1 = \omega_3 = 0.85\omega_2$ together with other various tunneling parameter sets. The case illustrated in Fig. 10 uses model parameters that are relatively close to the experimental TBG where we have one graphene layer aligned with BN. For equal gaps $\Delta_b = \Delta_t$ we find that beyond a gap of ~ 50 meV the low energy band becomes trivial, whereas when $\Delta_t = 0$ we can still find nonzero valley Chern numbers up to a larger $\Delta_b \sim 140$ meV. While relatively small mass terms are required to see low energy Chern bands we can still find finite Chern bands for higher energy bands when the mass terms are as large as a few eVs by taking large enough twist angles. Valley Chern bands can appear also for in the limit of small twist angles $\theta \lesssim 1^\circ$ in systems modeled through intralayer moire patterns like WSe₂/WSe₂ discussed in the appendix. We have represented in Fig. 11 the twist angle θ dependence of the Chern numbers for several massive twisted bilayer graphene

systems that we modeled from the parameters in Table I. For small band gaps we find nonzero valley Chern numbers for the lowest energy bands in a wide range of twist angles $\theta \gtrsim 1^\circ$ in the limit of small band gaps. When the intralayer band gaps are larger we notice a tendency for higher energy bands to acquire finite Chern numbers for sufficiently large twist angles $\theta \gtrsim 3^\circ$ for interlayer tunneling dominated moire pattern systems like SiC/SiC and BN/BN bilayers.

VI. VALLEY CONTRASTING OPTICAL TRANSITIONS

Numerous optical experiments for semiconducting transition metal dichalcogenides have verified optical dichroism in broken inversion symmetry single layer materials^{107–109} associated with the chirality of the layers in twisted multilayer systems^{85,86,110}. In the simplest picture, circular dichroism is expected in gapped Dirac materials due to valley contrasting orbital moments associated with the Berry curvatures at the band edges for circularly polarized dipole optical interband transitions involving angular momentum changes of $\Delta l = \pm 1$ ¹⁰⁸, and leading to selection rules of the form $m = w \pm 1$ for the promotion of angular momentum m excitons in w -chiral gapped Dirac systems^{107,109}. Here we investigate how in a gapped twisted bilayer graphene system the circular dichroism for the interband optical transitions are modified going from the decoupled layers limit consisting of two independent gapped Dirac Hamiltonian layers to two coupled gapped Dirac Hamiltonians leading to strongly hybridizing flatbands. In our models this crossover in behavior can be achieved either by changing the rotation angle between the bilayers or by modifying the interlayer coupling strength.

In a single gapped Dirac Hamiltonian model the interband optical oscillator strength for each k -point for circularly polarized light is given by¹¹¹

$$f_{nn'}^{\pm}(\mathbf{k}) = \frac{2|P_{nn'}^{\pm}(\mathbf{k})|^2}{m_e \hbar \omega_{nn'}(\mathbf{k})} = \frac{2|P_{nn'}^x(\mathbf{k}) \pm i P_{nn'}^y(\mathbf{k})|^2}{m_e \hbar \omega_{nn'}(\mathbf{k})} \quad (40)$$

where $P_{nn'}^{x/y} = m_e \langle u_n | \hat{v}_{x/y} | u_{n'} \rangle$ and $\hbar \omega_{nn'} = E_n(\mathbf{k}) - E_{n'}(\mathbf{k})$ is the energy difference between the u_n and $u_{n'}$ states, and we define the degree of circular polarization for each k -point to be¹⁰⁸

$$\eta_{nn'}(\mathbf{k}) = \frac{|P_{nn'}^+(\mathbf{k})|^2 - |P_{nn'}^-(\mathbf{k})|^2}{|P_{nn'}^+(\mathbf{k})|^2 + |P_{nn'}^-(\mathbf{k})|^2}. \quad (41)$$

For sake of simplicity the present analysis focuses on the strongest features of interband transitions in the dipole approximation and neglects the cross σ_{xy} terms responsible for the optical activity due to twist angle dependent phase difference between top and bottom layers⁸⁶ and the higher order terms proportional to the magnetic fields¹¹⁰. Studies about the selection rules for the promotion of the excitons^{107,109} and discussions related with the moire excitons^{112–114} for twisted gapped Dirac bilayer systems will be discussed elsewhere.

In Fig. 12 we show a schematic illustration of the band edges near each macrovalley K and K' for twisted bilayers

FIG. 12. (Color online). Schematic illustration of twisted gapped Dirac bilayers for zero and finite interlayer potential difference near 0° and 60° rotation alignments giving rise to same and opposite mass signs respectively at the minivalleys $\tilde{K}$ and $\tilde{K}'$. We can expect macrovalley K and K' contrasting circular dichroism near 0° alignment and suppressed circular dichroism near 60° . For the latter it is possible to introduce a finite circular dichroism by applying interlayer bias and Fermi level change that leads to layer and minivalley polarization (within a macrovalley).

near 0° and 60° alignments where we expect different optical responses because the band edges near $\tilde{K}$ and $\tilde{K}'$ minivalleys within a macrovalley could have the same or opposite mass signs. Circular dichroism is present near 0° alignment due to the alignment of the mass signs in each macrovalley, while near 60° alignment the valley contrasting circular dichroism is canceled although not completely if we take into account the phases acquired due to the rotation of the layers^{85,110} that should also be accounted for in our twisted gapped Dirac systems. We can see that this cancellation can also become nonzero when the minivalleys are polarized by the external electric field that polarizes layer and a finite carrier doping that shifts the Fermi level.

In Fig. 13 we illustrate how the interband optical transition oscillator strengths are progressively modified in twisted systems going from two weakly coupled gapped Dirac cones for large twist angles of $\sim 3^\circ$ to strongly coupled bands leading to flat band systems near $\sim 1^\circ$. The separate representation of $|P_{nn'}^+(\mathbf{k})|^2$ and $|P_{nn'}^-(\mathbf{k})|^2$ allows to distinguish the sensitivity to opposite circular polarization of the states at different k -points in the mBZ. In the limit of large twist angles the distribution of the oscillator strengths in the mBZ are in qualitative agreement with the expected interband transitions obtained juxtaposing two gapped Dirac cones next to each other separated by K_θ in momentum space and coupled through the interlayer tunneling parameters ω_i . For large enough twist angles and small primary gaps the interband transition oscillator strengths clearly peak around the $\tilde{K}$ and $\tilde{K}'$ points in keeping with the optical properties of a single gapped Dirac Hamiltonian¹⁰⁰. As the twist angle is reduced and the interlayer

FIG. 13. (Color online) Interband transition oscillator strengths $f_{nm'}^{\pm}(\mathbf{k})$ and dichroism $\eta_{nm'}(\mathbf{k})$ for circularly polarized light in twisted gapped Dirac material bilayers for various twist angles showing clear peaks around the gapped Dirac cone band edges at $\tilde{K}$ and $\tilde{K}'$. These peaks become broader and lower as the twist angle is progressively reduced and the quasiparticle velocities decrease. The comparison between the twisted bilayers near 0° and 60° alignment shows oscillator strength peaks with the same and opposite signs near each minivalley. We observe same sign and opposite sign circular dichroism $\eta(\mathbf{k})$ near the minivalleys indicative of the switch from macrovalley to minivalley contrasting physics depending on alignment. The transitions between selected bands are distinguished with blue (v1 and c1), black (c1 and c2), red (v2 and c2) and green (v1 and c2) solid lines. We used the interlayer coupling $\omega_t = 0.098$ eV and $|t_0| = 2.6$ eV, with a gap of $2\Delta = 0.3$ eV.

coupling strength is enhanced we observe a progressive flattening of the bands and reduction of quasiparticle velocities that in turn results in a reduction of the oscillator strength in Eq. (40). Its evolution at the mini Dirac points represented in Fig. 14 clearly shows a superlinear suppression of the oscillator strengths as a function of the band gap, and its evolution is faster for smaller than larger twist angles. However, the almost divergent increase in the density of states $D(E) \propto \theta^{2-m}$ in Eq. (10) due to squeezing bandwidth should increase the absorption rate as the bands become flatter until the flatness is limited by the broadening width due to disorder or temperature. This band flattening leads to smoother spread out and broad distribution of the Berry curvatures near the band edges in the mBZ. However, even in the limit of very narrow bandwidths near $\sim 1^\circ$ the oscillator strength for circularly polarized light remains predominantly centered around the minivalleys $\tilde{K}$ and $\tilde{K}'$ in the mBZ resulting in a polarization function $\eta(\mathbf{k})$ close to unity near the minivalleys. Due

to the almost complete flattening of the bands we notice that an external electric field that shifts the position of the band edges near the minivalley points can introduce distortions to the band structure that are significant enough to modify the oscillator strengths.

VII. SUMMARY AND DISCUSSION

Recent research on twisted van der Waals materials is rapidly expanding beyond twisted bilayer graphene (TBG) to include twisted layered materials with intrinsic gaps. In this work we have identified and explained the practical advantages of twisted gapped Dirac materials over TBG for the formation of narrow bandwidth flat bands. We studied the conditions for the generation of narrow bandwidth flat bands as a function of twist angle based on the extended Bistritzer-MacDonald model of twisted bilayer graphene with a finite

FIG. 14. (Color online) Oscillator strength corresponding to f_{lcv}^+ transitions between the highest valence and lowest conduction bands evaluated at the mini valley point $\tilde{K}$ where maxima values are expected in gapped Dirac materials. The suppression of the oscillator strength for smaller twist angles and intralayer band gap 2Δ illustrates the reduction of the quasiparticle velocity associated with the flattening of the bands. In the left panel we show that the suppression of the oscillator strength is faster than the growth rate of 2Δ for a fixed value of θ . In the right panel we show more clearly the steeper increase of the oscillator strength with twist angle in the small angle regime.

mass term in each one of the layers, allowing for intralayer moire patterns, and using up to three different interlayer tunneling parameters ω_i with $i = 1, 2, 3$ for a more precise description of the interlayer coupling. We have identified the evolution of the low energy bandwidth (W) and found its dependence as a function of the band gap (2Δ), twist angles (θ), the Fermi velocity (v_F) and interlayer coupling (ω_i). The fitting equation Eq. (9) is expected to be valid in the parameter range where several realistic 2D material combinations lie including gapped graphene due to alignment with hexagonal boron nitride (BN), transition metal dichalcogenides (TMDC), silicon carbide (SiC), and our analysis should be valid when the band edges near the K point can be described with gapped Dirac Hamiltonians. Systems whose single layer band edges are at Q or Γ points in the original Brillouin zone can in principle be analyzed in a similar way by rotating the bands and using the interlayer coupling terms at appropriate points in momentum space.

One of the main conclusions we draw is that a finite gap in the constituent layers of the twisted gapped Dirac materials makes the generation of flat bands simpler than in TBG because already for band gaps of ~ 250 meV the band flattening in twisted bilayers happens for a continuous range of small twist angles without requiring specific magic angles. Moreover, the larger the gaps the greater the suppression of the band width allowing to achieve narrower bands for similar twist angles, a fact that should facilitate achieving stronger effective Coulomb interactions $U \propto \theta$ that scales with twist angle. In moderately gapped TMDC materials or large band gap hBN materials we find that the bandwidths can remain below ~ 10 meV even for twist angles as large as $\sim 3^\circ$. Stronger interlayer coupling parameters ω_i also allows to achieve narrow bandwidths for larger twist angles. In the example case of twisted SiC bilayers perturbed by relatively strong intralayer

moire patterns and interlayer coupling the interplay of three unequal ω_i gave rise to valence bands with bandwidths on the order of ~ 20 meV even for twist angles as large as $\sim 7^\circ$, which implies a seven fold enhancement of the Coulomb interaction strength with respect to magic angle TBG based on the scale of the moire pattern periods. Our conclusions based on numerical calculations are complemented by the analytical solutions of the band eigenvalues at the symmetry points of the mBZ that provides estimates for the bandwidth as a function of the different system parameters such as twist angle, interlayer coupling and band gap.

The topological nature of the nearly flat bands in twisted gapped Dirac materials have been studied by calculating the phase diagrams for the valley Chern numbers associated to the first four conduction and valence bands. In particular, it was shown that the interlayer tunneling terms ω_i have to be different from each other for a valley Chern band to emerge in the limit of small intralayer band gaps like in systems of graphene on hexagonal boron nitride. The increase of twist angle θ in general helps to expand the phase space for the low energy valence and conduction bands to acquire a finite valley Chern number although this can effectively weaken the influence of the interlayer moire patterns and the magnitude of the isolation secondary gaps. For larger gap systems like in semiconducting TMDC or in the limit of large gap systems like hexagonal boron nitride, the lowest energy levels of a twisted bilayer remain topologically trivial but the higher bands can remain topological for large enough twist angles. These higher energy bands should be accessible either by gating techniques in devices in the small twist angle limit when the electron density per band is small, or through optical measurements. The valley contrasting circular dichroism in twisted gapped Dirac materials inherits the properties of single gapped Dirac layers whose band edges locate at the minivalley $\tilde{K}$ and $\tilde{K}'$ points of the moire Brillouin zone around which maxima in the optical transition oscillator strength for circularly polarized light and Berry curvature are often found. This behavior was found to persist even in the limit of multiple nearly flat bands where the traces of the original gapped Dirac cone band edges could not be clearly identified. From the local density of state plots these nearly flat bands can be viewed as discrete atomic-like levels of a lattice of nearly decoupled quantum dots than Landau levels whose electron densities are evenly distributed. Qualitatively distinct optical response to circularly polarized light is expected between twisted bilayer systems near 0° and 60° (or equivalently 180°) alignment where the phase winding and Berry curvature values of the bands in each minivalley could be aligned to point in the same or opposite directions respectively.

In summary, our analysis suggests optimistic prospects of finding isolated flat bands in systems other than twisted bilayer graphene in a variety of twisted gapped Dirac materials in particular when the original bandwidth of the building block materials are narrow to begin with, or when a strong interlayer interaction allows to access nearly flat bands for a larger range of twist angles.

ACKNOWLEDGMENTS

This work was supported by the Samsung Science and Technology Foundation under project no. SSTF-BA1802-06 for J. S. and J. J., and from the Korean NRF grant number NRF-2016R1A2B4010105 for S. J. Financial support for J. S. has also been granted by the National Natural Science Foundation of China (Grant No. 11604166), Zhejiang Provincial Natural Science Foundation of China (Grant No. LY19A040003) and K. C. Wong Magna Fund in Ningbo University. This work was partly performed at the Aspen Center for Physics, which is supported by National Science Foundation grant PHY-1607611.

Appendix A: Hamiltonian parameters fitting procedure from DFT calculations

Here we outline the methodology followed to obtain the Dirac Hamiltonian model parameters for materials involving SiC that were calculated from LDA-DFT calculations with the Perdew-Zunger parametrization¹¹⁵. For SiC bilayers we started from the full tight-binding (FTB) model Hamiltonian H_{mono} for monolayer SiC using the hopping parameters extracted from the maximally localized Wannier functions by means of a two by two Hamiltonian¹¹⁶ by means of a two by two Hamiltonian

$$H_{\text{mono}} = \begin{bmatrix} H_{\text{Si-Si}} & H_{\text{Si-C}} \\ H_{\text{C-Si}} & H_{\text{C-C}} \end{bmatrix}. \quad (\text{A1})$$

This monolayer Hamiltonian is then used to construct the bilayer SiC Hamiltonian through

$$H_{\text{bilayer}} = \begin{bmatrix} H_{\text{mono}} & T^0 \\ T^{0*} & H_{\text{mono}} \end{bmatrix} \quad (\text{A2})$$

where, is the FTB model Hamiltonian of monolayer SiC, and T^0 is the interlayer tunneling matrix as given in Eq. (8). We have determined the tunneling matrix elements ω_i that define T_0 by assuming that for large gap systems ω_1 , ω_3 determines the splitting in conduction, valence bands respectively, whereas ω_2 controls the interaction between conduction and valence bands. When fitting the tunneling matrix elements to reproduce the DFT-bands near the K -point for different standard stacking structures (AA, AB and BA) we followed the criteria that (a) the low energy bands should be parabolic at K -point so as to match with the gapped Dirac cone shaped DFT bands, (b) for all three standard SiC bilayer staking, at least four low energy bands (2-conduction,2-valence) should reproduce the DFT bands near K -point, and (c) the optimum ω_i parameters set should be unique and reproducible. The equilibrium interlayer distances have been relaxed within self-consistent local density approximation. With these criteria in mind the optimum tunneling parameters for AA-stacked bilayer SiC, are $\omega_1 = 0.485$ eV, $\omega_2 = \omega_2^* = 0$, and $\omega_3 = 0.19$ eV, while for AB-stacking, $\omega_1 = \omega_2 = \omega_3 = 0$ and $\omega_2^* = 1.24$ eV, and for BA-stacking $\omega_1 = \omega_2^* = \omega_3 = 0$ and $\omega_2 = 1.24$ eV. The tunneling matrix elements obtained from above procedure are then averaged for the three standard stacking structures and

they are $\omega_1 = 0.165$ eV, $\omega_2 = \omega_2^* = 0.413$ eV and $\omega_3 = 0.063$ eV as listed in Table I.

Appendix B: Analytical wave functions and comparison with numerical calculations

FIG. 15. (Color online) Comparison of the analytical and the numerical results for $\omega_1 = \omega_2 = \omega_3 = 0.098$ eV and $t_0 = -2.6$ eV. The left column shows the results for $2\Delta = 0.3$ eV corresponds to a small gap case, and the right column is for a large value $2\Delta = 4.5$ eV which is close to the gap size in BN/BN. We find a satisfactory agreement between both results especially in the regime of large twist angles where the analytical results are expected to work better. The difference of δ_s between the numerical and analytical data defined as $\delta_s = E_2(\Gamma) - E_1(\Gamma)$ for large twist angles is because the second conduction (valence) band minimum (maximum) starts to happen away from the Γ point rather than the due to lack of accuracy in the analytical expressions.

In this appendix we provide further details on the analytical solutions of the wave functions evaluated at the symmetry points. As we have discussed in the main text, $\{\eta_1, \eta_2\}$ only have three different combinations, which are $\phi_1 : \{\eta_1 = 0, \eta_2 = 2\pi/3\}$, $\phi_2 : \{\eta_1 = 2\pi/3, \eta_2 = -2\pi/3\}$, and $\phi_3 : \{\eta_1 = -2\pi/3, \eta_2 = 0\}$. Once ϕ_i , so the values of $\{\eta_1, \eta_2\}$ are fixed, the eigenenergy problem is then changed to solving a quartic equation

$$x^4 + \alpha x^2 + \beta x + \gamma = 0, \quad (\text{B1})$$

which has a general formula for roots, but the roots are in quite complicated forms. Alternatively, we discuss the calculation of approximate solutions below. For three different ϕ_i s, the coefficient in Eq. (B1) have different values. For ϕ_1 , the

coefficients are

$$\begin{aligned}\alpha &= -2(\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2), \\ \beta &= -6\Delta\omega_1^2, \\ \gamma &= (\Delta^2 + \rho_\theta^2)^2 - \Delta^2(5\omega_1^2 - 2\omega_2^2) - \rho_\theta^2(4\omega_1^2 + 2\omega_2^2) \\ &\quad + (2\omega_1^2 + \omega_2^2)^2.\end{aligned}\quad (\text{B2})$$

If $\beta = 0$, Eq. B1 becomes a quadratic equation and the 4 roots are given by

$$E_{\phi_1}^0(\tilde{\Gamma}) = \pm \sqrt{\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2 \pm \sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}. \quad (\text{B3})$$

Due to the ignorance of the $\beta = -6\Delta\omega_1^2$ term, $E_{\phi_1}^0(\tilde{\Gamma})$ is a good approximation only for small Δ and ω_1 . The energy correction δE_{ϕ_1} can be obtained by substituting $x = E_{\phi_1}^0(\tilde{\Gamma}) - \delta E_{\phi_1}$ to Eq. B1 with the coefficients given by Eq. B2, and expanding to the first order of δE_{ϕ_1} . After a straight forward

calculation, one can obtain

$$\delta E_{\phi_1} \approx \frac{\beta}{4(E_{\phi_1}^0)^2 + 2\alpha}, \quad (\text{B4})$$

and hence

$$E_{\phi_1}(\tilde{\Gamma}) \approx E_{\phi_1}^0(\tilde{\Gamma}) - \delta E_{\phi_1}. \quad (\text{B5})$$

The effect of β term in Eq. B2 is to add an energy correction to the roots given in Eq. B3. Among the 4 roots in Eq. B3, the one corresponds to the lower conduction band is

$$E_{\phi_1}^0(\tilde{\Gamma}) = \sqrt{\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2 - \sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}, \quad (\text{B6})$$

and the energy correction due to the β is

$$\delta E_{\phi_1} = \frac{3\Delta\omega_1^2}{2\sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}. \quad (\text{B7})$$

Thus the lower conduction band energy for ϕ_1 case is given by

$$\begin{aligned}E_{\phi_1}(\tilde{\Gamma}) &\approx E_{\phi_1}^0(\tilde{\Gamma}) - \delta E_{\phi_1} \\ &= \sqrt{\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2 - \sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}} \\ &\quad - \frac{3\Delta\omega_1^2}{2\sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}.\end{aligned}\quad (\text{B8})$$

For ϕ_3 case, α and γ values for are the same as those in the ϕ_1 case as given in Eq. B2, the only difference is that $\beta = 6\Delta\omega_1^2$, which is with opposite sign in comparison with Eq. B2. Hence $E_{\phi_3}(\tilde{\Gamma})$ has a similar form as $E_{\phi_1}(\tilde{\Gamma})$ and is given by

$$E_{\phi_3}(\tilde{\Gamma}) \approx E_{\phi_1}^0(\tilde{\Gamma}) + \delta E_{\phi_1}. \quad (\text{B9})$$

The coefficients for ϕ_2 case are

$$\begin{aligned}\alpha &= -2(\Delta^2 + \rho_\theta^2 + \omega_1^2 + 4\omega_2^2), \\ \beta &= 0, \\ \gamma &= (\Delta^2 + \rho_\theta^2)^2 - \Delta^2(2\omega_1^2 - 8\omega_2^2) - \rho_\theta^2(-2\omega_1^2 + 8\omega_2^2) \\ &\quad + (\omega_1^2 - 4\omega_2^2)^2.\end{aligned}\quad (\text{B10})$$

The energy corresponds to ϕ_2 at $\tilde{\Gamma}$ point has a simpler form

$$E_{\phi_2}(\tilde{\Gamma}) = \sqrt{\Delta^2 + \rho_\theta^2 + \omega_1^2 + 4\omega_2^2} - 2\sqrt{\Delta^2\omega_1^2 + 4\rho_\theta^2\omega_2^2 + 4\omega_1^2\omega_2^2} \quad (\text{B11})$$

When $\omega_1 = \omega_3$, the gapped Dirac system preserves the electron-hole symmetry. We briefly discuss the energy eigenvalues the related ϕ_i phases for the three valence bands close to the Fermi level. As we have introduced in Section. IV. B, for each ϕ one may obtain 4 roots, which are 4 eigenenergies

of the 12 by 12 Hamiltonian. The three lowest conduction band energies are given by $E_{\phi_i}(\tilde{\Gamma})$ ($i=1,2,3$), where $E_{\phi_i}(\tilde{\Gamma})$ is one of the 4 roots when the vector phases are given by ϕ_i . Among the other three roots for each ϕ_i , there exists one root which corresponds to one of the three valence bands close to the Fermi level. The valence bands energies are

$$E_{\phi_1/\phi_3}^v(\tilde{\Gamma}) \approx -\sqrt{\Delta^2 + \rho_\theta^2 + 2.5\omega_1^2 + \omega_2^2} - \sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2} \mp \frac{3\Delta\omega_1^2}{2\sqrt{10\Delta^2\omega_1^2 + 9\rho_\theta^2\omega_1^2 + 9/4\omega_1^4 + 4\rho_\theta^2\omega_2^2 + \omega_1^2\omega_2^2}}, \quad (\text{B12})$$

$$E_{\phi_2}^v(\tilde{\Gamma}) = -\sqrt{\Delta^2 + \rho_\theta^2 + \omega_1^2 + 4\omega_2^2} - 2\sqrt{\Delta^2\omega_1^2 + 4\rho_\theta^2\omega_2^2 + 4\omega_1^2\omega_2^2}.$$

One can find that the valence band energies at the $\tilde{\Gamma}$ point are related to the conduction band energies as $E_{\phi_1}^v(\tilde{\Gamma}) = -E_{\phi_3}(\tilde{\Gamma})$, $E_{\phi_2}^v(\tilde{\Gamma}) = -E_{\phi_2}(\tilde{\Gamma})$ and $E_{\phi_3}^v(\tilde{\Gamma}) = -E_{\phi_1}(\tilde{\Gamma})$.

Appendix C: Bandwidth evolution in higher energy bands, Chern number phase diagrams and Berry curvatures

As noted in the main text the bandwidth of the moire energy bands are reduced when the twist angles become sufficiently small or when the intralayer gaps are increased. The narrowing of the bandwidth in the three low lying conduction or valence bands for increasing 2Δ is illustrated in Fig. 16 and is also reflected in the progressive reduction of the y-axis scale. For the Hamiltonian approximation where we have used with equal ω_i and neglected the twisting phases we have electron-hole symmetry that results in equal bandwidths for the valence and conduction bands. The bandwidth compression happens most effectively for the lowest energy valence and conduction bands as the band gap 2Δ is increased.

If we focus on the behavior of the lowest energy bands we can find in Fig. 17 the bandwidth evolution as a function of system parameters Δ and θ that offers a complementary perspective of the Fig. 6 in the main text of the different quantities W , δ_s , δ_p and the ratio between Coulomb interaction versus bandwidth U/W .

The Chern number phase diagram in Fig. 18 shows that the valence and conduction bands are closely electron-hole symmetric in our simplified model where the twist angle dependent phases are not included, and deviations in the electron-

FIG. 16. (Color online) Evolution of the bandwidth corresponding to the first three low energy bands for different band gaps 2Δ . We observe a clear decrease in the bandwidths of the low lying energy bands when the band gaps become larger. The bandwidth narrowing is more effective for the lowest energy conduction or valence band represented with a green line extending for larger θ values as the band gap 2Δ increases.

hole symmetry that distinguish the results of the valence and conduction bands stem from the use of unequal interlayer tunneling values ω_i . Sensitive changes of Chern number as a function of gap magnitude found for large twist angles and high energy bands indicate the complex avoided crossing structure at higher energies. We have listed in each column the Chern bands phase diagram for same v_F parameter as in graphene and use different interlayer tunneling terms ω_1 , ω_3 and variable gap values Δ_b (Δ_t) of the bottom (top)

FIG. 17. (Color online) Phase diagram colormaps of the bandwidth W , secondary gap δ_s , primary gap δ_p , the U_{eff}/W ratio in $\tilde{G}/\tilde{G}$, MoS₂/WS₂ TMDC, BN/BN and SiC/SiC bilayers similar to the results in Fig. 6 but represented for fixed ω and variable 2Δ , θ . This plot illustrates how the range of angles for which we find narrow bandwidths disappears for gradually increasing band gap 2Δ . The 2Δ of each system which are obtained from DFT(except for $\tilde{G}/\tilde{G}$) are indicated with a green horizontal line in each phase diagram. The threshold value of the $U_{\text{eff}}/W = 1$ ratio where Coulomb interactions are expected to be significant is indicated with a blue line. In systems with large band gaps we find suppressed bandwidths in a large parameter space of twist angle versus interlayer tunneling.

layer. In the first and second columns we show the results for the interlayer tunneling parameters of $\omega_1 = \omega_3 = 0.85\omega_2$ and $\omega_1 = \omega_3 = 0.5\omega_2$. As noted earlier, if we choose the values of ω_i ($i=1,2,3$) to be identical, both conduction and valence

bands are trivial in the Δ_b , Δ_t phase space, but if the diagonal tunneling terms $\omega_1 = \omega_3$ become distinct from the off-diagonal ω_2 the integer valley Chern number emerges in two belt regions that become wider as the difference between ω_1 and ω_2 increases. Hence, the low energy Chern bands are possible only when there are at least two different ω_i values coupling different sublattices in Eq. (8), and a sufficiently small finite mass term in at least one of the layers. The nontrivial Chern band parameter region is generally larger in Δ_t , Δ_b space when the differences in the tunneling parameters are also large, as we can verify comparing $\omega_1 = \omega_3 = 0.85\omega_2$ and $\omega_1 = \omega_3 = 0.5\omega_2$ columns, and we have an intermediate situation for $\omega_1 = 0.85\omega_2$, $\omega_3 = 0.5\omega_2$ where we additionally include a difference between the diagonal hopping parameters ω_1 , ω_2 . The former case is closely related with the experimental situation of TBG where one layer is aligned with BN. When $\Delta_b = \Delta_t$ the maximum allowed gap is ~ 50 meV before the level becomes trivial, whereas if $\Delta_t = 0$ the maximum Δ_b allowed to preserve a topological band is about ~ 140 meV. A qualitatively different phase diagram is found when $\omega_1 = \omega_3 = 1.3\omega_2$ when the diagonal tunneling elements become larger than the off-diagonal interlayer terms, while we still require small enough mass terms to preserve a Chern band. However, even when the mass terms are as large as a few eVs it is possible to find Chern bands for large enough twist angles and higher energy bands. The valley Chern bands appear even for small twist angles $\theta \lesssim 1^\circ$ in WSe₂/WSe₂ system modeled mainly through intralayer moire patterns.

We plot in Fig. 19 the Berry curvatures obtained from Eq. (39) in the main text to illustrate that they are peaked most strongly at the symmetry points in the Brillouin zone. This information can be useful to identify the momentum space regions contributing to specific interband optical transitions and exciton promotion for circularly polarized light. In the cases of $\tilde{G}/\tilde{G}$ and $\tilde{G}/G$, which are closest to the actual experiments of twisted bilayer graphene nearly aligned with hexagonal boron nitride, we find that the Berry curvature concentration shifts from $\tilde{\Gamma}$ to $\tilde{K}$ and $\tilde{K}'$ points as the twist angles become larger and we progressively approach the picture of two weakly coupled massive Dirac cones.

In Fig. 20 we supplement the valley Chern number phase diagram as a function of twist angle presented in the main text Fig. 11 by introducing interlayer potential differences of $V_g = 0.05$ eV and 0.1 eV. We find that these experimentally achievable interlayer potential values are able to alter the Chern number phase diagram especially for higher energy bands and for systems whose moire patterns are defined mainly by intralayer potentials than interlayer tunneling.

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¹ K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, science **306**, 666 (2004).

² A. K. Geim and I. V. Grigorieva, Nature **499**, 419 (2013).

³ Y. Zhang, Y.-W. Tan, H. L. Stormer, and P. Kim, Nature **438**, 201 (2005).

⁴ A. F. Young and P. Kim, Nature Physics **5**, 222 (2009).

⁵ Y. Gong, J. Lin, X. Wang, G. Shi, S. Lei, Z. Lin, X. Zou, G. Ye, R. Vajtai, B. I. Yakobson, *et al.*, Nature materials **13**, 1135 (2014).

- ⁶ C. Huang, S. Wu, A. M. Sanchez, J. J. Peters, R. Beanland, J. S. Ross, P. Rivera, W. Yao, D. H. Cobden, and X. Xu, *Nature materials* **13**, 1096 (2014).
- ⁷ X. Duan, C. Wang, J. C. Shaw, R. Cheng, Y. Chen, H. Li, X. Wu, Y. Tang, Q. Zhang, A. Pan, *et al.*, *Nature nanotechnology* **9**, 1024 (2014).
- ⁸ N. D. Drummond, V. Zólyomi, and V. I. Fal'ko, *Phys. Rev. B* **85**, 075423 (2012).
- ⁹ P. Vogt, P. De Padova, C. Quaresima, J. Avila, E. Frantzeskakis, M. C. Asensio, A. Resta, B. Ealet, and G. Le Lay, *Phys. Rev. Lett.* **108**, 155501 (2012).
- ¹⁰ C.-C. Liu, W. Feng, and Y. Yao, *Phys. Rev. Lett.* **107**, 076802 (2011).
- ¹¹ Z. Ni, Q. Liu, K. Tang, J. Zheng, J. Zhou, R. Qin, Z. Gao, D. Yu, and J. Lu, *Nano letters* **12**, 113 (2011).
- ¹² M. Dávila, L. Xian, S. Cahangirov, A. Rubio, and G. Le Lay, *New Journal of Physics* **16**, 095002 (2014).
- ¹³ L. Matthes, O. Pulci, and F. Bechstedt, *Journal of Physics: Condensed Matter* **25**, 395305 (2013).
- ¹⁴ J. Wilson and A. Yoffe, *Advances in Physics* **18**, 193 (1969).
- ¹⁵ L. F. Mattheiss, *Phys. Rev. B* **8**, 3719 (1973).
- ¹⁶ Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, *Nature nanotechnology* **7**, 699 (2012).
- ¹⁷ X. Xu, W. Yao, D. Xiao, and T. F. Heinz, *Nature Physics* **10**, 343 (2014).
- ¹⁸ J. Hass, F. Varchon, J. E. Millán-Otoya, M. Sprinkle, N. Sharma, W. A. de Heer, C. Berger, P. N. First, L. Magaud, and E. H. Conrad, *Phys. Rev. Lett.* **100**, 125504 (2008).
- ¹⁹ D. L. Miller, K. D. Kubista, G. M. Rutter, M. Ruan, W. A. de Heer, P. N. First, and J. A. Stroscio, *Science* **324**, 924 (2009).
- ²⁰ D. L. Miller, K. D. Kubista, G. M. Rutter, M. Ruan, W. A. de Heer, P. N. First, and J. A. Stroscio, *Phys. Rev. B* **81**, 125427 (2010).
- ²¹ M. L. Sadowski, G. Martinez, M. Potemski, C. Berger, and W. A. de Heer, *Phys. Rev. Lett.* **97**, 266405 (2006).
- ²² W. A. De Heer, C. Berger, X. Wu, M. Sprinkle, Y. Hu, M. Ruan, J. A. Stroscio, P. N. First, R. Haddon, B. Piot, *et al.*, *Journal of Physics D: Applied Physics* **43**, 374007 (2010).
- ²³ I. Brihuega, P. Mallet, H. González-Herrero, G. Trambly de Laisardière, M. M. Ugeda, L. Magaud, J. M. Gómez-Rodríguez, F. Ynduráin, and J.-Y. Veuillen, *Phys. Rev. Lett.* **109**, 196802 (2012).
- ²⁴ T. Ohta, J. T. Robinson, P. J. Feibelman, A. Bostwick, E. Rotenberg, and T. E. Beechem, *Phys. Rev. Lett.* **109**, 186807 (2012).
- ²⁵ J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto, *Phys. Rev. B* **86**, 155449 (2012).
- ²⁶ J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto, *Phys. Rev. Lett.* **99**, 256802 (2007).
- ²⁷ S. Shallcross, S. Sharma, E. Kandelaki, and O. A. Pankratov, *Phys. Rev. B* **81**, 165105 (2010).
- ²⁸ S. Shallcross, S. Sharma, and O. A. Pankratov, *Phys. Rev. Lett.* **101**, 056803 (2008).
- ²⁹ W. Landgraf, S. Shallcross, K. Türschmann, D. Weckbecker, and O. Pankratov, *Phys. Rev. B* **87**, 075433 (2013).
- ³⁰ S. Shallcross, S. Sharma, and O. Pankratov, *Phys. Rev. B* **87**, 245403 (2013).
- ³¹ R. Bistritzer and A. H. MacDonald, *Proceedings of the National Academy of Sciences* **108**, 12233 (2011).
- ³² P. Moon and M. Koshino, *Phys. Rev. B* **87**, 205404 (2013).
- ³³ P. Moon and M. Koshino, *Phys. Rev. B* **85**, 195458 (2012).
- ³⁴ J. Jung, A. Raoux, Z. Qiao, and A. H. MacDonald, *Phys. Rev. B* **89**, 205414 (2014).
- ³⁵ P. San-Jose, J. González, and F. Guinea, *Phys. Rev. Lett.* **108**, 216802 (2012).
- ³⁶ P. San-Jose and E. Prada, *Phys. Rev. B* **88**, 121408 (2013).
- ³⁷ T. Stauber, P. San-Jose, and L. Brey, *New Journal of Physics* **15**, 113050 (2013).
- ³⁸ R. Bistritzer and A. H. MacDonald, *Phys. Rev. B* **81**, 245412 (2010).
- ³⁹ Z. Wang, F. Liu, and M. Chou, *Nano letters* **12**, 3833 (2012).
- ⁴⁰ H. Schmidt, J. C. Rode, D. Smirnov, and R. J. Haug, *Nature communications* **5** (2014).
- ⁴¹ S. Carr, S. Fang, P. Jarillo-Herrero, and E. Kaxiras, *Phys. Rev. B* **98**, 085144 (2018).
- ⁴² M. Koshino, N. F. Q. Yuan, T. Koretsune, M. Ochi, K. Kuroki, and L. Fu, *Phys. Rev. X* **8**, 031087 (2018).
- ⁴³ J. Kang and O. Vafek, *Phys. Rev. Lett.* **122**, 246401 (2019).
- ⁴⁴ G. Tarnopolsky, A. J. Kruchkov, and A. Vishwanath, *Phys. Rev. Lett.* **122**, 106405 (2019).
- ⁴⁵ H. C. Po, L. Zou, T. Senthil, and A. Vishwanath, *Phys. Rev. B* **99**, 195455 (2019).
- ⁴⁶ A. Luican, G. Li, A. Reina, J. Kong, R. R. Nair, K. S. Novoselov, A. K. Geim, and E. Y. Andrei, *Phys. Rev. Lett.* **106**, 126802 (2011).
- ⁴⁷ G. Li, A. Luican, J. L. Dos Santos, A. C. Neto, A. Reina, J. Kong, and E. Andrei, *Nature Physics* **6**, 109 (2010).
- ⁴⁸ D. Wong, Y. Wang, J. Jung, S. Pezzini, A. M. DaSilva, H.-Z. Tsai, H. S. Jung, R. Khajeh, Y. Kim, J. Lee, S. Kahn, S. Tolabimazraehno, H. Rasool, K. Watanabe, T. Taniguchi, A. Zettl, S. Adam, A. H. MacDonald, and M. F. Crommie, *Phys. Rev. B* **92**, 155409 (2015).
- ⁴⁹ A. Kerelsky, L. McGilly, D. M. Kennes, L. Xian, M. Yankowitz, S. Chen, K. Watanabe, T. Taniguchi, J. Hone, C. Dean, A. Rubio, and A. J. Pasupathy, *arXiv:1812.08776*.
- ⁵⁰ Y. Choi, J. Kemmer, Y. Peng, A. Thomson, H. Arora, R. Polski, Y. Zhang, H. Ren, J. Alicea, G. Refael, *et al.*, *Nature Physics* , 1 (2019).
- ⁵¹ Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, *Nature* **556**, 43 (2018).
- ⁵² M. Yankowitz, S. Chen, H. Polshyn, Y. Zhang, K. Watanabe, T. Taniguchi, D. Graf, A. F. Young, and C. R. Dean, *Science* **363**, 1059 (2019).
- ⁵³ Y. Cao, D. Chowdhury, D. Rodan-Legrain, O. Rubies-Bigordà, K. Watanabe, T. Taniguchi, T. Senthil, and P. Jarillo-Herrero, *arXiv preprint arXiv:1901.03710* (2019).
- ⁵⁴ Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. Y. Luo, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, *et al.*, *Nature* **556**, 80 (2018).
- ⁵⁵ K. Kim, A. DaSilva, S. Huang, B. Fallahazad, S. Larentis, T. Taniguchi, K. Watanabe, B. J. LeRoy, A. H. MacDonald, and E. Tutuc, *Proceedings of the National Academy of Sciences* **114**, 3364 (2017).
- ⁵⁶ A. L. Sharpe, E. J. Fox, A. W. Barnard, J. Finney, K. Watanabe, T. Taniguchi, M. A. Kastner, and D. Goldhaber-Gordon, *Science* **365**, 605 (2019).
- ⁵⁷ F. Wu, T. Lovorn, E. Tutuc, and A. H. MacDonald, *Phys. Rev. Lett.* **121**, 026402 (2018).
- ⁵⁸ M. H. Naik and M. Jain, *Phys. Rev. Lett.* **121**, 266401 (2018).
- ⁵⁹ M. H. Naik, S. Kundu, I. Maity, and M. Jain, *arXiv preprint arXiv:1908.10399* (2019).
- ⁶⁰ Y. Pan, S. Fölsch, Y. Nie, D. Waters, Y.-C. Lin, B. Jariwala, K. Zhang, K. Cho, J. A. Robinson, and R. M. Feenstra, *Nano letters* **18**, 1849 (2018).
- ⁶¹ L. Xian, D. M. Kennes, N. Tancogne-Dejean, M. Altarelli, and A. Rubio, *Nano Lett.* **19**, 4934 (2019).
- ⁶² X.-J. Zhao, Y. Yang, D.-B. Zhang, and S.-H. Wei, *arXiv preprint arXiv:1906.05992* (2019).
- ⁶³ L.-k. Shi, J. Ma, and J. C. Song, *arXiv preprint*

- arXiv:1904.07877 (2019).
- ⁶⁴ N. R. Chebrolu, B. L. Chittari, and J. Jung, Phys. Rev. B **99**, 235417 (2019).
 - ⁶⁵ Y.-H. Zhang, D. Mao, Y. Cao, P. Jarillo-Herrero, and T. Senthil, Phys. Rev. B **99**, 075127 (2019).
 - ⁶⁶ J. Liu, Z. Ma, J. Gao, and X. Dai, Phys. Rev. X **9**, 031021 (2019).
 - ⁶⁷ G. Chen, L. Jiang, S. Wu, B. Lyu, H. Li, B. L. Chittari, K. Watanabe, T. Taniguchi, Z. Shi, J. Jung, *et al.*, Nature Physics **15**, 237 (2019).
 - ⁶⁸ G. Chen, A. L. Sharpe, P. Gallagher, I. T. Rosen, E. Fox, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, J. Jung, Z. Shi, D. Goldhaber-Gordon, Y. Zhang, and W. Feng, Nature (2019).
 - ⁶⁹ B. L. Chittari, N. Leconte, S. Javvaji, and J. Jung, Electronic Structure **1**, 015001 (2019).
 - ⁷⁰ X. Liu, Z. Hao, E. Khalaf, J. Y. Lee, K. Watanabe, T. Taniguchi, A. Vishwanath, and P. Kim, arXiv preprint arXiv:1903.08130 (2019).
 - ⁷¹ J. Y. Lee, E. Khalaf, S. Liu, X. Liu, Z. Hao, P. Kim, and A. Vishwanath, arXiv preprint arXiv:1903.08685 (2019).
 - ⁷² C. Shen, N. Li, S. Wang, Y. Zhao, J. Tang, J. Liu, J. Tian, Y. Chu, K. Watanabe, T. Taniguchi, *et al.*, arXiv preprint arXiv:1903.06952 (2019).
 - ⁷³ Y. W. Choi and H. J. Choi, arXiv preprint arXiv:1903.00852 (2019).
 - ⁷⁴ M. Koshino, Phys. Rev. B **99**, 235406 (2019).
 - ⁷⁵ F. D. M. Haldane, Phys. Rev. Lett. **61**, 2015 (1988).
 - ⁷⁶ C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 146802 (2005).
 - ⁷⁷ R. Nandkishore and L. Levitov, Phys. Rev. B **82**, 115124 (2010).
 - ⁷⁸ J. Jung, F. Zhang, and A. H. MacDonald, Phys. Rev. B **83**, 115408 (2011).
 - ⁷⁹ F. Zhang, J. Jung, G. A. Fiete, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. **106**, 156801 (2011).
 - ⁸⁰ G. Chen, A. L. Sharpe, E. J. Fox, Y.-H. Zhang, S. Wang, L. Jiang, B. Lyu, H. Li, K. Watanabe, T. Taniguchi, *et al.*, arXiv preprint arXiv:1905.06535 (2019).
 - ⁸¹ M. Serlin, C. Tschirhart, H. Polshyn, Y. Zhang, J. Zhu, K. Watanabe, T. Taniguchi, L. Balents, and A. Young, arXiv preprint arXiv:1907.00261 (2019).
 - ⁸² J. R. Wallbank, A. A. Patel, M. Mucha-Kruczyński, A. K. Geim, and V. I. Fal'ko, Phys. Rev. B **87**, 245408 (2013).
 - ⁸³ J. Jung, E. Laksono, A. M. DaSilva, A. H. MacDonald, M. Mucha-Kruczyński, and S. Adam, Phys. Rev. B **96**, 085442 (2017).
 - ⁸⁴ F. Wu, T. Lovorn, E. Tutuc, I. Martin, and A. H. MacDonald, Phys. Rev. Lett. **122**, 086402 (2019).
 - ⁸⁵ C.-J. Kim, A. Sánchez-Castillo, Z. Ziegler, Y. Ogawa, C. Noguez, and J. Park, Nature nanotechnology **11**, 520 (2016).
 - ⁸⁶ E. S. Morell, L. Chico, and L. Brey, 2D Materials **4**, 035015 (2017).
 - ⁸⁷ D. Xiao, G.-B. Liu, W. Feng, X. Xu, and W. Yao, Phys. Rev. Lett. **108**, 196802 (2012).
 - ⁸⁸ J. Jung, A. M. DaSilva, A. H. MacDonald, and S. Adam, Nature communications **6**, 6308 (2015).
 - ⁸⁹ B. Hunt, J. Sanchez-Yamagishi, A. Young, M. Yankowitz, B. J. LeRoy, K. Watanabe, T. Taniguchi, P. Moon, M. Koshino, P. Jarillo-Herrero, *et al.*, Science **340**, 1427 (2013).
 - ⁹⁰ E. Wang, X. Lu, S. Ding, W. Yao, M. Yan, G. Wan, K. Deng, S. Wang, G. Chen, L. Ma, *et al.*, Nature Physics **12**, 1111 (2016).
 - ⁹¹ C. Woods, L. Britnell, A. Eckmann, R. Ma, J. Lu, H. Guo, X. Lin, G. Yu, Y. Cao, R. Gorbachev, *et al.*, Nature physics **10**, 451 (2014).
 - ⁹² H. Kim, N. Leconte, B. L. Chittari, K. Watanabe, T. Taniguchi, A. H. MacDonald, J. Jung, and S. Jung, Nano letters **18**, 7732 (2018).
 - ⁹³ M. Kindermann, B. Uchoa, and D. L. Miller, Phys. Rev. B **86**, 115415 (2012).
 - ⁹⁴ P. San-Jose, A. Gutiérrez-Rubio, M. Sturla, and F. Guinea, Phys. Rev. B **90**, 075428 (2014).
 - ⁹⁵ M. Yankowitz, K. Watanabe, T. Taniguchi, P. San-Jose, and B. J. LeRoy, Nature communications **7**, 13168 (2016).
 - ⁹⁶ C.-C. Liu, W. Feng, and Y. Yao, Phys. Rev. Lett. **107**, 076802 (2011).
 - ⁹⁷ J. C. W. Song, P. Samutpraphoot, and L. S. Levitov, Proceedings of the National Academy of Sciences **112**, 10879 (2015).
 - ⁹⁸ B. L. Chittari, G. Chen, Y. Zhang, F. Wang, and J. Jung, Phys. Rev. Lett. **122**, 016401 (2019).
 - ⁹⁹ C. Fang, M. J. Gilbert, and B. A. Bernevig, Phys. Rev. B **86**, 115112 (2012).
 - ¹⁰⁰ D. Xiao, W. Yao, and Q. Niu, Phys. Rev. Lett. **99**, 236809 (2007).
 - ¹⁰¹ Y.-H. Zhang, D. Mao, and T. Senthil, arXiv preprint arXiv:1901.08209 (2019).
 - ¹⁰² N. Bultinck, S. Chatterjee, and M. P. Zaletel, arXiv preprint arXiv:1901.08110 (2019).
 - ¹⁰³ A. H. MacDonald, Physics Online Journal **12** (2019).
 - ¹⁰⁴ Q. Tong, H. Yu, Q. Zhu, Y. Wang, X. Xu, and W. Yao, Nature Physics **13**, 356 (2017).
 - ¹⁰⁵ H. Yu, M. Chen, and W. Yao, arXiv preprint arXiv:1906.05499 (2019).
 - ¹⁰⁶ D. Xiao, M.-C. Chang, and Q. Niu, Rev. Mod. Phys. **82**, 1959 (2010).
 - ¹⁰⁷ X. Zhang, W.-Y. Shan, and D. Xiao, Phys. Rev. Lett. **120**, 077401 (2018).
 - ¹⁰⁸ W. Yao, D. Xiao, and Q. Niu, Phys. Rev. B **77**, 235406 (2008).
 - ¹⁰⁹ T. Cao, M. Wu, and S. G. Louie, Phys. Rev. Lett. **120**, 087402 (2018).
 - ¹¹⁰ T. Stauber, T. Low, and G. Gómez-Santos, Phys. Rev. Lett. **120**, 046801 (2018).
 - ¹¹¹ P. Y. Yu and M. Cardona, *Fundamentals of semiconductors: physics and materials properties* (Springer, 2010).
 - ¹¹² K. Tran, G. Moody, F. Wu, X. Lu, J. Choi, K. Kim, A. Rai, D. A. Sanchez, J. Quan, A. Singh, *et al.*, Nature **567**, 71 (2019).
 - ¹¹³ K. L. Seyler, P. Rivera, H. Yu, N. P. Wilson, E. L. Ray, D. G. Mandrus, J. Yan, W. Yao, and X. Xu, Nature **567**, 66 (2019).
 - ¹¹⁴ C. Jin, E. C. Regan, A. Yan, M. I. B. Utama, D. Wang, S. Zhao, Y. Qin, S. Yang, Z. Zheng, S. Shi, *et al.*, Nature **567**, 76 (2019).
 - ¹¹⁵ J. P. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981).
 - ¹¹⁶ N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Rev. Mod. Phys. **84**, 1419 (2012).

FIG. 18. (Color online) Valley Chern number phase diagram of massive twisted bilayer graphene as a function of the mass terms Δ_t and Δ_b for the top and bottom layers evaluated for the first four valence bands for a variety of massive twisted bilayer graphene systems using $\omega = 0.098$ eV that supplements the phase diagram presented in the main text for the first conduction and valence bands in Fig. 10.

FIG. 19. (Color online) Berry curvatures for the first four conduction bands of different twisted gapped Dirac Hamiltonian bilayers modeled through the parameters in Table I. The peaks of the Berry curvature are often located at the symmetry points of the moire Brillouin zone where the avoided gaps are produced and their values depend sensitively on the Hamiltonian details. For increasing twist angles the two twisted gapped Dirac Hamiltonians tend to effectively decouple for low energies, and therefore the Berry curvature tend to concentrate more sharply near the symmetry points of the mBZ where the gaps decrease.

FIG. 20. (Color online) Valley Chern numbers as a function of interlayer potential difference $V_g = 0.05\text{ eV}$ and 0.1 eV that supplements the calculation results in Fig. 11 for $V_g = 0$. It is shown that moderate values of interlayer potential differences V_g achievable in experiments introduces non-negligible changes in the valley Chern number phase diagram as a function of twist angle, especially for higher energy bands and when significant intralayer moire patterns are present.