Electron-hole asymmetry and band gaps of commensurate double moiré patterns in twisted bilayer graphene on hexagonal boron nitride

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Spontaneous orbital magnetism observed in twisted bilayer graphene (tBG) on nearly aligned hexagonal boron nitride (BN) substrate builds on top of the electronic structure resulting from combined G/G and G/BN double moiré interfaces. Here we show that tBG/BN commensurate double moiré patterns can be classified into two types, each favoring the narrowing of either the conduction or the valence bands on average, and obtain the evolution of the bands as a function of the interlayer sliding vectors and electric fields. Finite valley Chern numbers ± 1 are found in a wide range of parameter space when the moiré bands are isolated through gaps, whereas the local density of states associated with the flatbands are weakly affected by the BN substrate invariably concentrating around the AA-stacked regions of tBG. We illustrate the impact of the BN substrate for a particularly pronounced electron-hole asymmetric band structure by calculating the optical conductivities of twisted bilayer graphene near the magic angle as a function of carrier density. The band structures corresponding to other N -multiple commensurate moiré period ratios indicate it is possible to achieve a narrow width of $W \lesssim 30$ -meV isolated folded band bundles for tBG angles $\theta \lesssim 1^\circ$.

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I. INTRODUCTION

The electronic properties of magic angle twisted bilayer graphene (tBG) and other graphene moiré systems have become an intense focus of research in recent years after observations of strong correlations and superconductivity in transport experiments [1–6]. One of the striking observations revealing the inherent topological character of the bands in graphene moiré systems [7,8] is the spontaneous anomalous Hall effects at $3/4$ filling in tBG that appear even in the absence of an external magnetic field [9–11], which can be viewed as a solid-state realization of the Haldane-like model in a honeycomb lattice [12]. Theories attempting to explain the spontaneous Hall effects relied on band structures that included a small mass term in the Dirac Hamiltonian contacting the boron nitride (BN) substrate [13,14] that give rise to precursor bands of the spontaneous quantum Hall

phases. The fact that this effect in tBG has been observed so far only in tBG samples that are nearly aligned with BN [15–17] motivates further research on the effects of the BN substrate in modifying the electronic structure of twisted bilayer graphene. Recent literature on flatband twisted two-dimensional materials have shown remarkable Coulomb interaction effects manifested in the insulating gaps, superconducting phases, and orbital magnetism to mention a few examples. Our single-particle results aim to provide clear reference electronic structures that are precursors of the ordered phases that we can expect when Coulomb interactions are included.

In this paper we show that commensurate double moiré patterns of tBG on BN can have significant electron-hole asymmetric bands in addition to gaps between the moiré bands which provide the conditions for the formation of valley Chern bands prone to strong correlations. Here we focus our discussions on commensurate double moiré patterns of tBG/BN, whose continuum Hamiltonian can be conveniently formulated in terms in a unique moiré Brillouin zone. The electronic structure of double moiré patterns was addressed in the past for arbitrary relative twist angles in systems, such as BN encapsulated graphene [18–20] or twisted trilayer graphene [21] giving rise to supermoiré patterns. Earlier studies of BN/G/BN double moiré systems [18] have shown that the

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largest double moiré interference effects are seen when the patterns are commensurate and in these cases the electronic structures undergo significant modifications when interlayer sliding is introduced.

Electronic structure calculations for tBG/BN relying on commensurate double moiré patterns carried out recently have shown the opening of gaps between moiré bands that approximately maintains electron-hole symmetry [22,23]. In contrast, in our paper we show that the moiré bands can have significant electron-hole asymmetry depending on the relative sliding between the moiré patterns and the twist angles. For type-1 double moiré systems we can obtain, on average, a narrow conduction band and a wider valence band, whereas we predict that type-2 double moiré systems can form ordered phases for both conduction and valence bands slightly favoring the valence bands. Our results provide a possible justification that the spontaneously broken-symmetry phases have been observed in experiments only for the conduction bands for the type 1 of double moiré arrangements [6,9], whereas correlated phases are likely for the valence bands for type 2. In both cases we find that isolation of the flatbands required to endow finite valley Chern numbers is facilitated by the opening of primary and secondary gaps. We find that valley Chern numbers are finite with values of ± 1 over a large parameter space in keeping with experimentally observed quantized Hall conductivities, whereas the local density of states associated with the flatbands invariably concentrate around the AA-stacked regions of tBG regardless of the BN arrangement. We illustrate for a particularly pronounced electron-hole asymmetric band structure the impact of the BN moiré pattern by showing the differences in the optical conductivities between tBG and tBG/BN as a function of carrier density. Band structures corresponding to other rational numbers N -multiple commensurate moiré periods indicate that it is possible to find narrow width $W \lesssim 30$ -meV isolated folded band bundles for tBG angles $\theta \lesssim 1^\circ$.

The paper is structured as follows. In Sec. II we introduce the model Hamiltonian for the doubly commensurate tBG on BN. In Sec. III we present the exact geometric conditions under which the double moiré patterns become commensurate with the same period and orientations. We then move on to analyze the electronic structures in Sec. IV where we present the band structures for different relative sliding vectors paying particular attention to the bandwidth, gaps, and valley Chern numbers, and study the effects of interlayer potential differences due to a perpendicular electric field. In Sec. V we illustrate the effects of the BN substrate in the optical conductivity near the magic angle twisted bilayer graphene for a clearly electron-hole asymmetric electronic structure with a narrow conduction band. In Sec. VI we discuss the electronic structures for commensurate double moiré patterns with different periods. Finally we close the paper in Sec. VII with the conclusions.

II. MODEL HAMILTONIAN

The model Hamiltonian for doubly commensurate tBG/BN can be conveniently formulated through the moiré bands theory with a single Brillouin zone [24,25] without needing to recourse to a supermoiré framework

for incommensurate double moiré patterns [18–20,26]. The Hamiltonian matrix elements arising from the two moiré interfaces from tBG and tGBN lead to two types of arrangements illustrated in Figs. 1(a)–1(f) that can be built by properly matching the orientation of the moiré pattern angles with the moiré reciprocal lattice vectors, see Figs. 1(e)–1(j). The continuum model Hamiltonian is concisely given as $H = H_{\text{tBG}}(\theta) + H_{\text{BN}}^M(\mathbf{r})$, where $H_{\text{tBG}}(\theta)$ is the Hamiltonian of tBG for the K valley,

$$H_{\text{tBG}}(\theta) = \begin{pmatrix} h_b(-\theta/2) & T(\mathbf{r}) \\ T^\dagger(\mathbf{r}) & h_t(\theta/2) \end{pmatrix} + V, \quad (1)$$

where $V = \text{diag}(\eta, \eta, -\eta, -\eta)/2$ represents the interlayer potential difference resulting from an applied electric field in the perpendicular direction and $H_{\text{BN}}^M(\mathbf{r})$ describes the moiré potential corresponding to BN placed underneath tBG, where $h_{b,t}(\pm\theta/2)$ represents a 2×2 Dirac Hamiltonian $v_F \mathbf{p} \cdot \boldsymbol{\sigma}$ for the bottom (b) and the top (t) layers of tBG rotated by $\pm\theta/2$ ($+$: counterclockwise, $-$: clockwise), yielding

$$h_{b,t}(\pm\theta/2) = D(\mp\theta/2) v_F \mathbf{p} \cdot \boldsymbol{\sigma} D(\pm\theta/2), \quad (2)$$

where $D(\pm\theta) = e^{\pm i\theta\sigma_z/2}$ and the Fermi velocity is $v_F = t_0\sqrt{3}a_G/2\hbar$, choosing $t_0 = 3.1$ eV [27]. Here, $\boldsymbol{\sigma} = (\sigma_x, \sigma_y)$, and σ_z is the z component of the Pauli matrix. The interlayer coupling $T(\mathbf{r})$ is given by

$$T(\mathbf{r}) = \sum_{j=0,\pm} e^{-iq_j \cdot \mathbf{r}} T_{s,s'}^j, \quad (3)$$

where $\mathbf{q}_0, \mathbf{q}_\pm$ are given as $\mathbf{q}_0 = \theta k_D(0, -1)$, $\mathbf{q}_\pm = \theta k_D(\pm\sqrt{3}/2, 1/2)$ if the twist angle θ is small enough. Here, $k_D = 4\pi/3a_G$ is the magnitude of the graphene BZ corner wave vector where the lattice constant of graphene is given as $a_G = 2.461$ Å. The interlayer tunneling matrices $T_{s,s'}^j$ were formulated for the AA initial stacking in Ref. [25] as

$$T^j = e^{-i\mathbf{G}_j \cdot \boldsymbol{\tau}} \begin{pmatrix} \omega_{A,A'} & \omega_{A,B'} e^{-ij\varphi} \\ \omega_{B,A'} e^{ij\varphi} & \omega_{B,B'} \end{pmatrix}, \quad (4)$$

where $\mathbf{G}_0 = (0, 0)$, $\mathbf{G}_\pm = k_D(-3/2, \pm\sqrt{3}/2)$, and $\boldsymbol{\tau} = (\tau_x, \tau_y)$ describe a relative sliding of the top G layer with respect to the bottom layer of graphene, $\omega_{A,A'} = \omega_{B,B'} = \omega'$, and $\omega_{A,B'} = \omega_{B,A'} = \omega$, we get

$$T^0 = \begin{pmatrix} \omega' & \omega \\ \omega & \omega' \end{pmatrix}, \quad T^\pm = \begin{pmatrix} \omega' & \omega e^{\mp i\varphi} \\ \omega e^{\pm i\varphi} & \omega' \end{pmatrix} \quad (5)$$

for local AA stacking corresponding to $\boldsymbol{\tau} = (0, 0)$, where $\varphi = 2\pi/3$. The local AB stacking corresponds to $\boldsymbol{\tau} = a_G(0, 1/\sqrt{3})$, and the local BA stacking is given by $\boldsymbol{\tau} = a_G(0, 2/\sqrt{3})$ or, equivalently, $\boldsymbol{\tau} = a_G(0, -1/\sqrt{3})$. By taking different $\omega' = 0.0939$ and $\omega = 0.12$ eV's for the intra- and intersublattice couplings between layers related through $\omega' = A\omega^2 + B\omega + C$ where $A = -0.5506$, $B = 1.036$, and $C = -0.02245$ following the exact exchange and random phase approximations total energy minima parametrization [27,28].

For tBG/BN, the moiré potential Hamiltonian $H_{\text{BN}}^M(\mathbf{r})$ has the form

$$H_{\text{BN}}^M(\mathbf{r}) = \sum_{m=1}^6 (W_m e^{i\tilde{\mathbf{G}}_m \cdot \mathbf{r}}) \oplus \mathbf{0}_{2 \times 2}, \quad (6)$$

$$W_m = \begin{pmatrix} C_{AA}e^{i[(-1)^m\phi_{AA}]} & C_{AB}e^{i[(-1)^m\phi_{AB}-\varphi_m]} \\ -C_{AB}e^{i[(-1)^m\phi_{AB}+\varphi_m]} & C_{BB}e^{i[(-1)^m\phi_{BB}]} \end{pmatrix}. \quad (7)$$

$\tilde{G}_m$ for tBG. Here we use the moiré potential parameter set given by $C_{AA} = -14.88$ meV, $\phi_{AA} = 50.19^\circ$, $C_{BB} = 12.09$ meV, $\phi_{BB} = -46.64^\circ$, $C_{AB} = 11.34$ meV, $\phi_{AB} = 19.60^\circ$ following Refs. [25,29].

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$\mathbf{G}_i$ ($i = 1, \dots, 6$). On the other hand, for tGBN we have $\mathbf{G}'_i = \epsilon \mathbf{G}_i - \theta \hat{z} \times \mathbf{G}_i$. We assume that for the commensurate angle sets (see the next section for further details), the mBZ of tBG and tGBN have the same size and share their two corners in momentum space as shown in Figs. 1(e) and 1(j) (see Appendix A for further details).

We label each commensurate stacking in real space with three characters, A, B, and C to label from bottom to top layers the relative position of the left sublattice of the two atoms unit cell of G or BN. For instance, when the local AA-stacking region in tBG is AA stacked on top of hBN (AA-tBG/BN), a type-1 system gives rise to three AAA, ABC, and ACB local stackings at the symmetry points, whereas AAA, ACA, and ABA local stackings appear in type-2 systems. We plot each of the commensurate stacking configurations of tBG/BN with six small colored circles under the moiré patterns for type 1 in Figs. 1(b)–1(d) and for type 2 in 1(g)–1(i).

III. COMMENSURATE TWIST ANGLE SETS

Among the many possible commensurate double moiré patterns [18–20] here we want to focus on the subset that can be obtained by requiring two constraints: (1) moiré length: the moiré lengths of tBG and tGBN should be the same, and (2) moiré angle: the difference of the moiré angle should be 0 (mod 60°). The moiré length of tBG is

$$L_{\text{tBG}}^M = \frac{a_G}{2 \sin(\theta/2)}, \quad (8)$$

and the moiré length of tGBN is

$$L_{\text{tGBN}}^M = \frac{a_G}{\sqrt{\alpha^2 - 2\alpha \cos \theta' + 1}}, \quad (9)$$

where $\alpha = a_G/a_{\text{BN}} = 1 + \epsilon$ and $\epsilon = (a_G - a_{\text{BN}})/a_{\text{BN}}$ [25] are parameters that represent the lattice mismatch between two layers. We use for the lattice constant of graphene $a_G = 2.461 \text{ \AA}$ and boron nitride $a_{\text{BN}} = 2.504 \text{ \AA}$, which results in $\epsilon = -0.017172$. Here, we use the notation for angles measured with respect to the bottom graphene (G) layer for convenience since both moiré interfaces are sharing it; we define $\theta = \theta_{\text{tBG}}$ as the twist angle of tBG which is measured with respect to the bottom G layer, and $\theta' = -\theta_{\text{tGBN}}$ is the twist angle of BN measured with respect to the contacting bottom G layer at the tGBN interface and, hence, the minus sign. The condition for coincident moiré lengths $L_{\text{tBG}}^M = L_{\text{tGBN}}^M$ requires the following relation between θ and θ' :

$$\theta = \cos^{-1} \left[\frac{(1 + \alpha)(1 - \alpha) + 2\alpha \cos \theta'}{2} \right]. \quad (10)$$

Accordingly, the rotation angle $\phi = \phi_{\text{tBG}}$ of the moiré pattern in tBG with respect to the bottom G layer is [25]

$$\phi = \tan^{-1} \left(\frac{\sin \theta}{\cos \theta - 1} \right). \quad (11)$$

This expression reduces to

$$\phi = -\text{sgn}(\theta) \frac{\pi}{2} + \frac{\theta}{2}, \quad (12)$$

indicating that the moiré pattern orientation rotates with respect to the bottom G by one half of the total twist angle between the graphene layers. On the other hand, the tGBN

moiré pattern rotation angle $\phi' = \phi_{\text{tGBN}}$ measured with respect to the bottom G layer is given by

$$\phi' = \tan^{-1} \left(\frac{\alpha \sin \theta'}{\alpha \cos \theta' - 1} \right). \quad (13)$$

The condition for the two equal period moiré patterns to be commensurate is

$$\Delta\phi = \phi' - \phi = \frac{n\pi}{3}, \quad (14)$$

where n is an integer number [18]. There is no analytical solution for $n = 0$, but we have for $n = 1$ the type-1 solution at $\Delta\phi = 60^\circ$ or, equivalently, -120° because of the π periodicity of the tangent function and have for $n = 2$ the type-2 solution at $\Delta\phi = -60^\circ$ or, equivalently, 120° . The rotation of the moiré patterns and associated moiré Brillouin zones are shown graphically in Figs. 1(a) and 1(f) in real space, Figs. 1(e) and 1(j), and in Appendix A in momentum space respectively. The main differences between the two solutions are that in type-1 systems the top G layer and the bottom hBN layer are rotated in the same sense, e.g., ($\theta > 0$, $\theta' > 0$) with respect to the bottom G layer, whereas in type 2 they are rotated in the opposite directions with respect to the bottom G layer, e.g., ($\theta > 0$, $\theta' < 0$). Imposing equal moiré length $L_{\text{tBG}}^M = L_{\text{tGBN}}^M$ conditions from Eqs. (8) and (9) leads to

$$4 \sin^2(\theta/2) = [\alpha - \cos(\theta')]^2 + \sin^2(\theta'), \quad (15)$$

that is equivalent to Eq. (14) with $n = 1$ when we use the $\theta = 2\theta'$ relation [22,30] reducing to

$$\alpha \pm \sqrt{3} \sin(\theta') = \cos(\theta'), \quad (16)$$

that yields same sign angles for type-1 solutions ($\theta, \theta' = (1.1329^\circ, 0.5664^\circ)$). The equations for unequal sign angles for type-2 solutions can be formulated by combining both Eqs. (14) and (15) for $n = 2$ and their numerical solutions are ($\theta, \theta' = (1.1463^\circ, -0.5932^\circ)$). Geometric discussions of the double moiré commensuration conditions in real and momentum spaces are discussed in Appendix A. The real-space moiré patterns of the commensurate double moiré patterns for the three different stacking configurations between the top and the bottom G layers are shown for both type-1 in Figs. 1(b)–1(d) and type-2 systems in Figs. 1(g)–1(i) where we can clearly see their differences both in the real-space patterns as well as in their momentum space-band structures.

IV. BAND STRUCTURES

In this section we present the band structures of tBG/BN corresponding to the two types of commensurate stacking arrangements introduced in earlier sections as a function of moiré pattern sliding and perpendicular external fields. We will be paying attention to the bandwidth, the gaps, and the electron-hole asymmetry. Knowledge of the band structure allows to estimate regimes of strong correlations that satisfy $U_{\text{eff}}/W \gtrsim 1$ where the bandwidths W are sufficiently narrow versus the effective Coulomb interaction,

$$U_{\text{eff}} = \frac{e^2}{4\pi\epsilon_r\epsilon_0 l_M} \exp(-l_M/\lambda_D), \quad (17)$$

where we used the dielectric constant of graphene as $\epsilon_r = 4$, the moiré length $l_M = a_G/[2 \sin(\theta/2)] \simeq a_G/\theta$, and the

screening length $\lambda_D = 2\epsilon_0/e^2 D(\delta_p, \delta_s)$ that depends on the two-dimensional density of states

$D(\delta_p, \delta_s) = 4[|\delta_p|u(-\delta_p) + |\delta_s|u(-\delta_s)]/(W^2 A_M)$ defined in terms of the Heaviside step function $u(x)$ that includes screening due to overlap between neighboring bands [27]. The valley Chern number of the n th energy band $C_n = \int_{\text{mBZ}} d^2\mathbf{k} \Omega_n(\mathbf{k})/(2\pi)$ is obtained integrating the Berry curvature [31] given by $\Omega_n(\mathbf{k}) = -2 \sum_{n' \neq n} \text{Im}[\langle n | \frac{\partial H}{\partial \mathbf{k}_x} | n' \rangle \langle n' | \frac{\partial H}{\partial \mathbf{k}_y} | n \rangle / (E_{n'} - E_n)^2]$. Additional information on the electronic structure can be found in Appendix C where we present the normalized local density of states (LDOS) whose maxima are expected at the AA-stacking sites of tBG.

A. Type-1 and type 2 electronic structures

We can distinguish two arrangements of double moiré patterns that we classify as type-1 and type-2 depending on the relative twist of the layers in the tBG and tGBN substrate that, in turn, impacts the relative rotation of the associated moiré patterns $\Delta\phi = \phi' - \phi$. For double moiré patterns of type 1 we require a relative rotation of $\Delta\phi = -120^\circ$ or, equivalently, $\Delta\phi = 60^\circ$ between the mBZs of tBG and tGBN as shown in Fig. 1(e). On the other hand, for type 2 we require $\Delta\phi = -60^\circ$ (see Appendix A) or, equivalently, $\Delta\phi = 120^\circ$ as shown in Fig. 1(j). The impact in the band structure of tBG/BN is represented along the high-symmetry line of the mBZ of tBG ($\tilde{K}-\tilde{K}'-\tilde{\Gamma}-\tilde{\Gamma}-\tilde{K}$), we show the band structures for Figs. 1(b)–1(d) for type-1 and Figs. 1(g)–1(i) for type-2 for different displacements of the top G layer for an electronic structure model with an unequal interlayer tunneling value that incorporates an effective vertical relaxation [27,28]. In our analysis the twist angles of tBG compatible with the double commensuration defined by Eqs. (13) and (14) essentially depend on the value of the lattice constant mismatch $\alpha = a_G/a_{\text{BN}}$. The double commensuration angles $\theta \simeq 1.13^\circ$ and $\theta \simeq 1.14^\circ$ result from our choice of graphene and BN lattice constants. We can expect the bandwidths will remain narrow provided that these angles remain close to the experimental [32] $\theta_{\text{MA}}^{\text{exp}} \simeq 1.08^\circ$ and theoretical [24,27,28] $\theta_{\text{MA}}^{\text{th}} \simeq 1.05^\circ$ values where flatbands emerge. We estimated the changes in the electronic structure for twist angles θ of tBG away from the magic angle by using an approximation scheme that preserves commensuration with the hBN moiré pattern potential which shows a similar qualitative behavior in the vicinity of the magic twist angle for variations $\delta\theta$ on the order of $\sim 10\%$ or smaller, see Appendix C.

The electronic structures depend strongly on the relative sliding between the moiré patterns of tBG and tGBN. We use a vector $\boldsymbol{\tau} = (\tau_x, \tau_y)$ to indicate the sliding of the top G layer prior to rotation and with this notation the local AA, AB, and BA stackings at the origin correspond to $(\tau_x, \tau_y) = (0, 0)$, $a_G(0, 1/\sqrt{3})$, and $a_G(0, 2/\sqrt{3})$. The local stacking maps between the two graphene sheets and the BN substrate are traced by assuming that the BN substrate is locked at the AA-stacking configuration with respect to the bottom graphene layer. For instance, the AA-tBG/BN case where tBG is AA stacked prior to twisting corresponds to Figs. 1(b) and 1(g). For type 1 we can find a small gap of ~ 2.5 meV at $\tilde{K}'$ whereas for type-2 the lowest-energy bands are quasiflat

with a large gap of ~ 35 meV at $\tilde{K}'$ and ~ 20 meV at $\tilde{\Gamma}$. For AB-tBG/BN corresponding to Figs. 1(c) and 1(h), type 1 bands consist of mutually overlapping bands with large secondary gaps δ_s for both conduction and valence bands that separate the lowest-energy bands with the next band that is farther in energy from charge neutrality, whereas for type 2 we find isolated low-energy conduction and valence bands with a δ_p gap of size ~ 15 meV at $\tilde{\Gamma}$. For BA-tBG/BN corresponding to Fig. 1(d), we see a large electron-hole asymmetric electronic structure separated with a small primary gap $\delta_p \simeq 3.5$ meV consisting of a nearly flat low-energy conduction band with $W \sim 5$ meV and a wider valence-band $W \sim 30$ meV that recalls the experimental observations of spontaneous Hall effects for electrons but not for holes [6,9]. Conversely for type 2 we have wider conduction and narrower valence bands that are separated by a finite gap of $\delta_p \simeq 5.6$ meV for the entire mBZ.

For both types of solutions, the secondary gap δ_s for the conduction and valence bands remain open for the three AA, AB, and BA symmetric-stacking geometries, and we will show shortly that this is the case for the entire range of sliding geometries. This is in contrast to the behavior of the primary gap δ_p that clearly closes when the top G layer is placed at $(\tau_x, \tau_y) = a_G(0, 1/\sqrt{3})$ in a type-1 system.

The Berry curvatures for discrete three commensurate slidings of the top G layer for the conduction and valence bands and the normalized local density of states $\tilde{D}(r, E) = D(r, E)/\max[|D(r, E)|]$ at the van Hove singularity can be found in Appendix C where we illustrate the impact of the BN substrate in altering those quantities.

Having identified the distinct behavior of the bands for type-1 and type-2 moiré pattern arrangements at three different symmetric-stacking configurations we discuss in the following the dependence of the band structure to other continuous-stacking sliding geometries and interlayer potential differences.

B. Sliding and electric-field dependent electronic structure

Here we extend our analysis of the lowest-energy bands to other continuous stacking geometries as a function of $\boldsymbol{\tau} = (\tau_x, \tau_y)$ and explore the changes in the electronic structure due to an interlayer potential difference η introduced by a perpendicular electric field. For all possible local stacking geometries we have obtained the maxima, the minima, the average, and the standard deviations of the bandwidths, which we succinctly summarize in Table I. From these data we can observe a more pronounced electron-hole asymmetry for type-1 systems with narrower conduction bands on average and smaller bandwidth fluctuations that posit the preference of the conduction bands to form ordered phases with respect to the valence bands. On the other hand, we expect a higher likelihood of observing ordered phases for both conduction and valence bands for type-2 systems because the electron-hole asymmetry is generally less pronounced, whereas narrower valence bands with smaller fluctuations are found for the average results.

The phase diagram maps for the bandwidth W , the U_{eff}/W ratios, the δ_p , δ_s gaps, and the valley Chern number maps for continuous-stacking geometries are shown in Fig. 2. The

FIG. 2. The bandwidth W , the secondary δ_s , the primary δ_p gaps, the ratio of the effective Coulomb interaction to W , and the valley Chern number of conduction and valence bands as a function of the sliding vector of graphene on graphene $\tau = (\tau_x, \tau_y)$ for type-1 (upper row) and type-2 (lower row) systems. The commensurate stacking with a translation by $\tau_y = a_G/\sqrt{3}$ and $\tau_y = 2a_G/\sqrt{3}$ along the line $\tau_x = 0$ represents the local-AB and local-BA stackings in tBG, respectively.

stacking configurations with the narrowest bandwidth and largest gaps have greater chances of enhanced Coulomb interactions that can lead to ordered phases. For roughly half of the stacking configuration space we can observe favorable $U_{\text{eff}}/W \gtrsim 1$ conditions, and they largely coincide with the regions that have finite valued valley Chern numbers. The regions with suppressed U_{eff}/W are mainly due to the closure of the primary gap δ_p that overlaps the two low-energy flatbands and enhances the Coulomb screening.

The phase diagram maps for the bandwidth W , the gaps, and the resulting U_{eff}/W ratios in Fig. 3 shows that a perpendicular electric field directed in the appropriate sense can enhance the narrowing of the bands and has an overall effect of enhancing the secondary gap δ_s , whereas the opening and closure of the primary gap δ_p follows a more complex behavior as a function of stacking and applied electric fields.

C. Band gap analysis at the $\tilde{K}'$ point

As discussed earlier, distinct electronic low-energy bands are expected depending on the moiré pattern-stacking types

TABLE I. The bandwidth W maxima, the minima, and the average taken from every possible local sliding configuration and the corresponding standard deviation for type-1 and type-2 moiré pattern stacking arrangements. We find narrower bandwidths with smaller fluctuations for the type-1 conduction and type-2 valence bands.

W (meV)	Type 1		Type 2	
	Conduction	Valence	Conduction	Valence
Max	15.2	29.2	26.5	21.7
Min	6.0	6.0	8.2	8.9
Average	11.6	13.1	15.0	14.8
Standard deviation	1.9	4.6	4.8	2.6

that we classify as type 1 and type 2. For example, in type 1 of AA-tBG/BN there is a small gap ~ 2.5 meV at $\tilde{K}'$ in contrast to the large gap ~ 35 meV in type 2. A band gap near charge neutrality in tBG/BN double moiré systems can be expected to appear due to moiré pattern interference effects. We illustrate this behavior by obtaining the analytical expressions for the size of the gaps at $\tilde{K}'$ for different moiré pattern types and relative stacking arrangements as a function of the model parameters that define the moiré patterns. For this purpose we consider a truncated 8×8 Hamiltonian as a simple model to describe the tBG system that captures the scattering of the electrons from the three Dirac cones of the bottom layer to one Dirac cone on the top layer [24]. At $\tilde{K}'$ the eigenstates can be represented by $\Psi = (\psi_1, \psi_2, \psi_3, \psi_4)^T$ consisting of four two-component sublattice pseudospin spinors [33]. Here the ψ_2, ψ_3 spinors can be rewritten in terms of ψ_4 by multiplying with unitary matrices $\psi_2 = U\psi_4$ and $\psi_3 = U^\dagger\psi_4$ and, thus, allowing to further reduce our Hamiltonian to a 3×3 matrix to obtain a simpler characteristic equation. Then, the expression for the band gap at $\tilde{K}'$ is as follows:

$$E_C(\tilde{K}') - E_V(\tilde{K}') \approx \frac{6(A_3 - B_3)}{3\omega^2 + 3\omega^2 + Q^2}, \quad (18)$$

where $Q = \hbar v_F \theta k_D$. This expression agrees up to two significant digits with the gap size obtained by diagonalizing numerically the eight-bands model. The above equation holds for both type-1 and type-2 solutions and for the three symmetric AA, AB, and BA stackings, but the analytical coefficients A_3 and B_3 are different for each case. Below we present the expression for the A_3 and B_3 coefficients for type-1 BA-tBG/BN for the electronic structure showing the largest electron-hole asymmetry that incorporates the effect of the interlayer potential difference η that can be induced by a perpendicular electric field and the moiré pattern potentials generated by the BN substrate that affects the magnitude of the gap. For sufficiently small magnitudes of η the A_3 and B_3 coefficients

FIG. 3. The bandwidth W , the secondary δ_s , the primary δ_p gaps, and the ratio of the effective Coulomb interaction to W for type 1 (upper row) and type 2 (lower row) as a function of continuous sliding of the top G layer in the y -direction (τ_y) prior to rotation obtained for various values of the interlayer potential difference η . We see that both stacking configuration and electric fields can substantially alter the bandwidth and band isolation of the low-energy moiré bands.

in Eq. (18) can be expressed in terms of the moiré pattern coefficients stemming from the hBN substrate as follows:

$$A_3 = \omega^2 C_{AA} \cos(\phi_{AA} + \varphi) + \omega^2 C_{BB} \cos(\phi_{BB} - \varphi) + \omega' \omega C_{AB} \cos(\phi_{AB} - \varphi) - (\eta/6)[-Q^2 + 4C_{AA} \cos(\phi_{AA} + \varphi)C_{BB} \cos(\phi_{BB} - \varphi) - 4C_{AB}^2 \cos^2(\phi_{AB} - \varphi) - 2\sqrt{3}QC_{AB} \cos(\phi_{AB} - \varphi)], \quad (19)$$

and

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA} - \varphi) + \omega^2 C_{BB} \cos(\phi_{BB}) + \omega' \omega C_{AB} \cos(\phi_{AB} + \varphi) - (\eta/6)[-Q^2 + 4C_{AA} \cos(\phi_{AA} - \varphi)C_{BB} \cos(\phi_{BB}) - 4C_{AB}^2 \cos^2(\phi_{AB} + \varphi) - 2\sqrt{3}QC_{AB} \cos(\phi_{AB} + \varphi)]. \quad (20)$$

In Appendix B we show a more detailed derivation of the analytic expressions for other types of solutions and stackings.

V. OPTICAL CONDUCTIVITY

Electronic structure reconstruction in the energy ranges of a few tens of meV in the vicinity of the Dirac cone of graphene can be conveniently explored by means of infrared and terahertz spectroscopies. Even though an experimental study of the optical conductivity of tBG/BN is difficult due to the large sample sizes that are normally required to obtain sharp optical features, typical of a few tens of microns for infrared and terahertz wavelengths, photocurrent-based measurements [34] would allow to extract reliable optical absorption spectra even in small area devices. Here we show the numerical results for the optical absorption in type-1 BA-tBG/BN as well as tBG without a BN substrate in an effort to estimate the differences in the optical signals that can be expected in these two systems. The real part of the longitudinal optical conductivity normalized by the universal optical conductivity of graphene $\sigma_0 = \pi e^2/2h$ is given

by [35–39]

$$\text{Re}[\sigma_{xx}(\omega)]/\sigma_0 = \frac{16}{\omega} \int \frac{d^2 \mathbf{k}}{(2\pi)^2} \sum_{i,j} [f(\epsilon_{k,i}) - f(\epsilon_{k,j})] |\langle \mathbf{k}, i | J_x | \mathbf{k}, j \rangle|^2 \times \delta[\omega + (\epsilon_{k,j} - \epsilon_{k,i})/\hbar], \quad (21)$$

where $J_\alpha = -\partial H/\partial k_\alpha$ is the current operator and $f(\epsilon)$ is the Fermi-Dirac distribution function. $\epsilon_{k,i}$ is the i th eigenstate energy at $\mathbf{k} = (k_x, k_y)$. In Fig. 4(a) we show the energy bands for type-1 BA-tBG/BN (black solid line) and tBG (red dotted line)

FIG. 4. (a) Energy bands under the effective vertical relaxation for BA-tBG/BN (black solid line) in type 1 and for tBG (red dashed line) for comparison. The horizontal magenta, blue, and green lines represent the positions of the Fermi energy that empties or fills a nearly flatband. Longitudinal optical conductivity in (b) tBG and in (c) tBG/BN in the case of the set of twist angles $(\theta, \theta') = (1.13 291^\circ, 0.5664^\circ)$ for different three values of chemical potential $\mu = -0.021$ eV (magenta circle), 0.012 eV (blue triangle), and 0.035 eV (green square) corresponding to the horizontal lines in panel (a).

TABLE II. The calculated (θ, θ') angle sets for doubly commensurate tBG/BN type-1 and type-2 systems where the moiré patterns are aligned mod 60° angles when the moiré patterns can be related through $L_{\text{tGBN}}^M = NL_{\text{tBG}}^M$.

		$L_{\text{tGBN}}^M = NL_{\text{tBG}}^M$						
N		1/4	1/3	1/2	1	2	3	4
Type 1	θ°	0.284447	0.379079	0.568072	1.132910	2.253321	3.362061	4.459900
	θ'°	0.576353	0.575245	0.573035	0.566455	0.553515	0.540851	0.528445
Type 2	θ°	0.285284	0.380567	0.571420	1.146307	2.306986	3.483098	4.675807
	θ'°	-0.583049	-0.584172	-0.586426	-0.593244	-0.607136	-0.621395	-0.636048

line) without a BN substrate where we achieve double commensuration of equal period moiré patterns by using the twist angles of $(\theta, \theta') = (1.13\ 291^\circ, 0.5664^\circ)$ as we introduced earlier. The horizontal lines are the three different chemical potentials $\mu = -0.021$ (magenta circle), 0.012 (blue triangle), and 0.035 eV (green square) considered, and we plot the real part of the linear optical absorption in the longitudinal direction for tBG in Fig. 4(b) and type-1 BA-tBG/BN in Fig. 4(c). The presence of the BN substrate leads to markedly different asymmetric optical absorptions for $\mu = -0.021$ eV (magenta circle) and 0.035 eV (green square), and one can observe small gaps less than 10 meV for $\mu = -0.021$ eV (magenta circle), 0.012 eV (blue triangle) in type-1 BA-tBG/BN as shown in Fig. 4(c), unlike without BN.

VI. COMMENSURATE DOUBLE MOIRÉ ANGLE SETS

So far we have considered doubly commensurate systems with equal moiré pattern lengths, and here we extend our band structure analysis to commensurate double moiré systems with unequal individual moiré lengths. An example of a doubly commensurate supermoiré built from unequal length moiré patterns was realized in twisted double bilayer graphene on hexagonal boron nitride arranged in a way that it gives rise to a Kekule-type supermoiré lattice whose moiré bands are flattened and isolated [40]. We expect that a similar bandwidth narrowing and band isolation behavior due to zone folding and formation of avoided gaps could take place in tBG/BN systems. Here we explore the band structures of supermoiré systems formed by combining unequal individual moiré lengths that are multiples of each other and have mod 60° alignment. The Hamiltonian matrix elements of the commensurate double moiré pattern can be built from the matrix elements of each moiré pattern labeled by the respective moiré reciprocal lattice vectors whose magnitude and relative orientations will change depending on the twist angle sets considered.

Our analysis focuses on the $L_{\text{tBG}}^M > L_{\text{tGBN}}^M$ cases when the moiré lengths of tBG are greater than those of tBG for $N = 1/2, 1/3$, and $1/4$ ratios considering that the moiré length $L_{\text{tBG}}^M \simeq a_G/\theta$ for tBG and $L_{\text{tGBN}}^M \simeq a_G/(\theta'^2 + \epsilon^2)^{1/2}$ for tGBN as a function of twist angles θ, θ' are given in Eqs. (8) and (9). These commensurate supermoiré patterns can be achieved in the limit of small tBG twist angles $\theta \lesssim 1^\circ$ and gives rise to zone folding as the moiré Brillouin zone reduces in size. The set of twist angles that satisfy the commensuration conditions are listed in Table II for type-1 and type-2 systems when $L_{\text{tGBN}}^M = NL_{\text{tBG}}^M$ for some integer and rational values of N .

The resulting band structures are presented in Fig. 5 for the three different starting stackings, AA, AB, and BA where we can observe bundles of narrow bands that we highlight with blue shades that are separated from each other forming bands of moiré bands. Likewise we have also highlighted with red shades the isolated nearly flat moiré bands that are separated from each other through avoided gaps. In particular, we find that the band bundles for $N = 1/4, 1/3$ commensurate ratios can have bandwidths of the order of ~ 30 meV.

These folded nearly flatbands are weakly dispersive in the reduced moiré Brillouin zones in the limit of small tBG twist angles. We expect that the moiré pattern features that appear at low energies due to the BN substrate can introduce gaps that separate the folded band bundles that enhance their isolation and reduce their screening. The notable changes in the band structures that we can observe for different relative sliding configurations for the doubly commensurate geometries indicate that small deviations of the twist angles can, in principle, lead to significant changes to the electronic structures, whereas we might also expect formation of gaps between the folded moiré band bundles due to the BN substrate. As we approach the regime of marginally twisted graphene bilayers we expect that the commensuration strain effects will become more important and should be considered in a more complete theory.

VII. CONCLUSION

We have investigated the electronic structure of commensurate double moiré patterns of tBG on BN in an effort to understand the effects introduced by a BN substrate when it is brought to near alignment with the graphene layers and identify the conditions that are favorable for the appearance of gapped moiré bands with finite valley Chern numbers that ultimately lead to the anomalous Hall effects observed in experiments.

The band structure of tBG has been calculated based on a continuum model that effectively accounts for vertical interlayer relaxations by using unequal interlayer tunneling parameters. The effects of a second moiré pattern produced by the BN substrate have been modeled assuming a moiré pattern that is perfectly commensurate with tBG which conveniently allows us to study the physics of the system based on a single moiré Brillouin-zone continuum Hamiltonian model. For these doubly commensurate geometries we have highlighted the importance of aligning properly the moiré reciprocal lattice upon rotation to properly construct the continuum moiré Hamiltonian and have illustrated the role of the BN moiré patterns and interlayer potential differences in opening the

FIG. 5. Band structures for the twist angle sets of N -multiple moiré lengths for $N = 1/4, 1/3, 1/2$ for twist angles listed in Table II for single moiré tBG and double moiré tBG/BN type 1 and type 2 systems. The AA, AB, and BA labels indicate the starting stacking of the top G layer prior to rotation. Isolated folded band bundles with narrow width $\lesssim 30$ meV are indicated by the blue-shaded regions, whereas the isolated single flatbands of width $\lesssim 20$ meV are indicated by red-shaded regions.

primary band gaps by obtaining the analytical form of the gaps near $\bar{K}'$ for a truncated eight-bands Hamiltonian model.

When the periods of each moiré pattern are equal, we have identified two types of commensurate double moiré patterns depending on the relative twist angles of the tBG layers with respect to the BN substrate that we classified as type 1 and type 2 and whose band structures have revealed markedly different features in their electron-hole asymmetry, the formation of band gaps, and the dependence on local interlayer sliding arrangement. In type-1 double moiré systems we find narrower overall conduction bands than valence bands with the most prominent asymmetry appearing in the vicinity of the BA-tBG/BN-stacking configuration where the conduction band has the smallest bandwidth ~ 5 meV suggesting that conduction bands are more prone for broken symmetry phases than the valence bands, whereas the electron-hole symmetry is partially restored in type-2 solutions that slightly favor, on average, the narrowing of the valence bands suggesting that ordered phases will likely appear for both conduction and valence bands. The impact of the BN substrate on the electronic structure should be observable not only through the transport measurement, but also through the optical absorption probes as we illustrated for the specific BA-tBG/BN example versus tBG without a substrate.

We have extended our electronic structure study to other commensurate double moiré systems with unequal moiré pattern lengths $L_{\text{tBG}}^M > L_{\text{tGBN}}^M$ focusing on the smaller bilayer graphene twist angles and verified that they often lead to separated folded moiré bands bundles with reduced widths

~ 30 meV. Hence, our results suggest that tBG twist angles in the range of 0.3° – 0.5° clearly below $\sim 1^\circ$ can also be candidate systems for exhibiting correlated phases.

The analysis presented in this paper for commensurate double moiré patterns for a variety of different sliding geometries shows how the BN substrate impacts the electronic structure of tBG, specifically leading to band-gap openings that isolate the moiré bands. Additionally, these band structures provide a useful starting point for understanding the electronic structure of incommensurate double moiré tBG/BN systems that can be pictured as a coherent combination of the commensurate states with different stacking. A more detailed study of the incommensurate double moiré patterns as they depart from perfect commensuration as well as the impact of lattice relaxations in the electronic structure is desirable to understand in greater detail the precursor states leading to the experimentally observed anomalous Hall orbital magnetization in tBG/BN.

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FIG. 6. Geometric solution of the two sets of commensurate angles for tBG/BN in momentum space for (a) type 1 ($\theta > 0$, $\theta' > 0$) and (b) type-2 ($\theta > 0$, $\theta' < 0$). The two dashed (purple) lines on the lower row for both type 1 and type 2 have the equal length. (c) The leftmost column represents the real-space representation of mass term $H_z^M(\mathbf{r})$ and the real part of the in-plane gauge field $\text{Re}[H_{AB}^M(\mathbf{r})]$ in tGBN for type 1 (upper two rows) and type 2 (lower two rows). The red arrow indicates the lattice vector of tGBN. The rest of the three columns represent the mass term and the real part of the in-plane gauge term in tBG/BN for type 1 (upper two rows) and type 2 (lower two rows) for three different $\Delta\phi$ values 0° , -60° , and -120° . The black arrows correspond to the lattice vector of tBG, and the red arrows represent that of tBG/BN. Note that the moiré patterns generated from tBG/BN coincide with those from tGBN (on the leftmost column) when $\Delta\phi$ (the angle difference between red and black arrows) is -120° for type 1 and $\Delta\phi = -60^\circ$ for type 2.

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APPENDIX A: GEOMETRICAL DERIVATION OF COMMENSURATE TWIST ANGLES AND MOIRÉ PATTERN ROTATIONS

There are two classes of solutions for moiré length $L_{\text{tBG}}^M = L_{\text{tGBN}}^M$ commensurate tBG/BN as shown in Fig. 1. Here we present a geometrical proof for both types of solution in momentum space and a more detailed analysis about how type-1 and type-2 moiré potentials in real space lead to relative moiré pattern rotation angles of $\Delta\phi = -120^\circ$ and $\Delta\phi = -60^\circ$, respectively.

We assume that the bottom and top G layers are rotated by $-\theta/2$ and $\theta/2$, respectively. In Fig. 6 we plot the BZ of the bottom G layer with a thick blue solid line and the BZ of the top G layer with a thick green solid line, generating the mBZ of tBG which is plotted by a black thin solid line. The thick brown solid line represents the BZ of hBN. For tBG/BN to be commensurate, the mBZ of tBG and of tGBN should have the same size and share their two corners. The geometrical understanding for type 1 is straightforward since the simple relation $\theta = 2\theta'$ holds as shown in Fig. 6(a). For type 2, on the other hand, if we assume the length from the center to the corner of the BZ of the hBN layer is $l\alpha$, then the length from the center to the corner of the BZ of the G layer is l , and the two lengths indicated by the dashed purple line in Fig. 6 must

be the same to share the two corners of the mBZ of tBG and tGBN. Then we get an equation which is equivalent to the $L_{\text{tBG}}^M = L_{\text{tGBN}}^M$ condition in Eq. (10). The shaded areas in red and blue in the upper row in Figs. 6(a) and 6(b) correspond to the mBZ depicted in Fig. 1(e) and 1(j), respectively.

We now move on to discuss the rotation of the moiré patterns. The moiré potential in Eq. (6) can be rewritten for the bottom G layer with a 2×2 matrix as follows:

$$H^M(\mathbf{r}) = \sum_{m=1}^6 W_m e^{i\tilde{\mathbf{G}}'_m \cdot \mathbf{r}} = V_{AA}(\mathbf{r}) \left(\frac{\sigma_0 + \sigma_z}{2} \right) + V_{BB}(\mathbf{r}) \left(\frac{\sigma_0 - \sigma_z}{2} \right) + \mathbf{A}(\mathbf{r}) \cdot \boldsymbol{\sigma}, \quad (\text{A1})$$

where $\tilde{\mathbf{G}}'_m$ are the moiré reciprocal vectors of tGBN,

$$V_{AA}(\mathbf{r}) = 2C_{AA} \text{Re}[e^{i\phi_{AA}} f(\mathbf{r})], \quad (\text{A2})$$

$$V_{BB}(\mathbf{r}) = 2C_{BB} \text{Re}[e^{i\phi_{BB}} f(\mathbf{r})], \quad (\text{A3})$$

and

$$\mathbf{A}(\mathbf{r}) = 2C_{AB}(\hat{\mathbf{z}} \times \nabla) \text{Re}[e^{i\phi_{AB}} f(\mathbf{r})]/|\tilde{\mathbf{G}}|. \quad (\text{A4})$$

Here, $f(\mathbf{r}) = \sum_m [1 + (-1)^m] e^{i\tilde{\mathbf{G}}'_m \cdot \mathbf{r}}/2$ and $|\tilde{\mathbf{G}}|$ is the magnitude of the moiré reciprocal lattice vector $\tilde{\mathbf{G}}_m$ of tBG. In the pseudospin basis $H_0^M(\mathbf{r}) = [V_{AA}(\mathbf{r}) + V_{BB}(\mathbf{r})]/2$ represents the periodic moiré potential, and

FIG. 7. (a) The bandwidth W , (b) the secondary gaps δ_s , (c) the primary gaps δ_p , (d) U_{eff}/W , and (e) the valley Chern number of the lowest conduction band (red filled circle) and valence band (blue empty circle) for type 1 (left column) and type 2 (right column) as a function of sliding in y -direction τ_y . Here, AAA stacking corresponds to AA-tBG/BN [$\tau = (0, 0)$], AAB stacking is AB-tBG/BN [$\tau = a_G(0, 1/\sqrt{3})$], and AAC stacking corresponds to BA-tBG/BN [$\tau = a_G(0, 2/\sqrt{3})$].

$H_z^M(\mathbf{r}) = [V_{AA}(\mathbf{r}) - V_{BB}(\mathbf{r})]/2$ is the local mass term that can give a gap at the neutrality point. The $\mathbf{A}(\mathbf{r})$ is a non-Abelian SU(2) gauge potential in the in-plane direction due to the unequal hopping probability between different sublattices of a honeycomb lattice [41,42]. We can rewrite the $\mathbf{A}(\mathbf{r}) \cdot \boldsymbol{\sigma}$ term as $H_{AB}^M(\mathbf{r})\sigma_+ + H_{BA}^M(\mathbf{r})\sigma_-$ where $\sigma_{\pm}$ is $(\sigma_x \pm i\sigma_y)/2$, and then from the real-space maps of the rotated moiré potentials $H_z^M(\mathbf{r})$ and $H_{AB}^M(\mathbf{r})$ we can verify the correct alignment required for the moiré reciprocal lattice vectors of both interfaces to assign the moiré pattern coefficients in the Hamiltonian matrix.

The leftmost column of Fig. 6(c) shows the mass term $H_z^M(\mathbf{r})$ in real space and the real part of gauge field $\text{Re}[H_{AB}^M(\mathbf{r})]$ of tGBN, and the red arrows represent the one of moiré lattice vectors resulted from the moiré patterns. On the other hand, the right three columns give $H_z^M(\mathbf{r})$ and $\text{Re}[H_{AB}^M(\mathbf{r})]$ of tBG/BN for different $\Delta\phi$'s. The black arrows are the moiré lattice vector directions of tBG, and the red arrows are those of tBG/BN. Note in Fig. 6(c) that the two components of the moiré potential, $H_z^M(\mathbf{r})$ and $H_{AB}^M(\mathbf{r})$ in real space for tGBN are in agreement with those of tBG/BN when $\Delta\phi = -120^\circ (-60^\circ)$ for the case of type 1 (type 2) as also illustrated in more detail in Figs. 1(e) and 1(j).

APPENDIX B: BAND GAP ANALYSIS AT $\tilde{K}'$

Here derive the analytic expressions for the energy gap at a specific high-symmetry point $\tilde{K}'$ that often is a useful estimate of the actual band gap at charge neutrality. We first consider truncating the range of $\tilde{\mathbf{G}}$ vectors for an eight-bands model of tBG [24] possessing three Dirac Hamiltonians for the bottom layer and one Dirac Hamiltonian for the top layer. We use the fact that among the four two-component pseudospin eigenstates in the basis $\Psi = (\psi_1, \psi_2, \psi_3, \psi_4)^T$ [33], the two spinors ψ_2 and ψ_3 can be written at $\tilde{K}'$ in terms of ψ_4 by a unitary transformation as follows, for example, for AA-tBG/BN,

$$\psi_2 = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\varphi} \end{pmatrix} \psi_4, \quad \psi_3 = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\varphi} \end{pmatrix}^\dagger \psi_4 \quad (\text{B1})$$

for the conduction (valence) band in type 1 (type 2), and

$$\psi_2 = \begin{pmatrix} e^{-i\varphi} & 0 \\ 0 & 1 \end{pmatrix} \psi_4, \quad \psi_3 = \begin{pmatrix} e^{-i\varphi} & 0 \\ 0 & 1 \end{pmatrix}^\dagger \psi_4 \quad (\text{B2})$$

for the valence (conduction) band in type 1 (type 2), we reduced our 8×8 Hamiltonian into a 3×3 matrix to find a secular equation. In type 1 for the conduction band,

$$\begin{pmatrix} -E_C & 3e^{-i\varphi}\omega & 3\omega' \\ e^{i\varphi}\omega & 2C_{AA}\cos(\phi_{AA} + \varphi) - E_C & 2C_{AB}\cos(\phi_{AB})e^{-i\varphi} + Qe^{i\pi/6} \\ \omega' & 2C_{AB}\cos(\phi_{AB})e^{i\varphi} + Qe^{-i\pi/6} & 2C_{BB}\cos(\phi_{BB}) - E_C \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \quad (\text{B3})$$

and for the valence band,

$$\begin{pmatrix} -E_V & 3\omega' & 3e^{i\varphi}\omega \\ \omega' & 2C_{AA}\cos(\phi_{AA}) - E_V & 2C_{AB}\cos(\phi_{AB} - \varphi/2)e^{i\varphi/2} + Qe^{i\pi/6} \\ e^{-i\varphi}\omega & 2C_{AB}\cos(\phi_{AB} - \varphi/2)e^{-i\varphi/2} + Qe^{-i\pi/6} & 2C_{BB}\cos(\phi_{BB} - \varphi) - E_V \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \quad (\text{B4})$$

FIG. 8. Electronic structure of tBG/BN as a function of tBG twist angle θ for commensurate double moiré patterns. The dashed vertical line indicates the twist angle θ corresponding to the doubly commensurate moiré pattern conditions determined by the fixed lattice constants $a_G = 2.461 \text{ \AA}$ of graphene and $a_{BN} = 2.504 \text{ \AA}$ of hexagonal boron nitride. For other twist angles we assume that the twist angle θ' as well as the lattice constant of hBN in the tGBN moiré interface changes such that the moiré vectors coincide with those of tBG in both length and angle. From top to bottom rows we show the bandwidths, the secondary gaps, the primary gaps, and the ratio of effective Coulomb interaction to bandwidth (U_{eff}/W) of the lowest conduction (red filled circle) and valence band (blue empty circle) for three different stacking configurations, AA-tBG/BN [$\tau = (0, 0)$], AB-tBG/BN [$\tau = a_G(0, 1/\sqrt{3})$], and BA-tBG/BN [$\tau = a_G(0, 2/\sqrt{3})$] prior to rotation for type-1 systems in panels (a)–(c) and panels (d)–(f) for type-2 systems.

where the eigenstates of the conduction and valence bands for the eight-band model are given as $\psi_{C,1} = (c \ 0 \ a \ b e^{i\varphi} \ a \ b e^{-i\varphi} \ a \ b)^T$ and $\psi_{V,1} = (0 \ c \ a e^{-i\varphi} \ b \ a e^{i\varphi} \ b \ a \ b)^T$. Here, $Q = \hbar v_F \theta k_D$.

We get characteristic equations from the above 3×3 matrices. For example,

$$E_C^3 - 2A_1 E_C^2 - A_2 E_C + 6A_3 = 0, \quad (\text{B5})$$

and

$$E_V^3 - 2B_1 E_V^2 - B_2 E_V + 6B_3 = 0, \quad (\text{B6})$$

where

$$A_1(B_1) = C_{AA} \cos \Theta_{I,t}^{A(B)} + C_{BB} \cos \Theta_{II,t}^{A(B)}, \quad (\text{B7})$$

and

$$\begin{aligned} A_2(B_2) = & 3(\omega'^2 + \omega^2) + Q^2 + 4C_{AB}^2 \cos^2 \Xi_{I,t}^{A(B)} \\ & - 4C_{AA}C_{BB} \cos \Xi_{II,t}^{A(B)} \cos \Theta_{II,t}^{A(B)} \\ & + 2\sqrt{3}QC_{AB} \cos \Xi_{I,t}^{A(B)} \end{aligned} \quad (\text{B8})$$

for $t = 1$ (type 1) and $t = 2$ (type 2). For type 1, Θ and Ξ on the order of AA-, AB-, BA-tBG/BN are given as follows:

$$\Theta_{I,1}^A = \begin{cases} \phi_{AA} \\ \phi_{AA} \\ \phi_{AA} \end{cases}, \quad \Theta_{I,1}^B = \begin{cases} \phi_{AA} - \varphi \\ \phi_{AA} \\ \phi_{AA} - \varphi \end{cases}, \quad (\text{B9})$$

$$\Theta_{II,1}^A = \begin{cases} \phi_{BB} + \varphi \\ \phi_{BB} - \varphi \\ \phi_{BB} - \varphi \end{cases}, \quad \Theta_{II,1}^B = \begin{cases} \phi_{BB} \\ \phi_{BB} + \varphi \\ \phi_{BB} \end{cases}, \quad (\text{B10})$$

$$\Xi_{I,1}^A = \begin{cases} \phi_{AB} \\ \phi_{AB} - \varphi \\ \phi_{AB} - \varphi \end{cases}, \quad \Xi_{I,1}^B = \begin{cases} \phi_{AB} + \varphi \\ \phi_{AB} \\ \phi_{AB} + \varphi \end{cases} \quad (\text{B11})$$

$$\Xi_{II,1}^A = \begin{cases} \phi_{AA} \\ \phi_{AA} + \varphi \\ \phi_{AA} + \varphi \end{cases}, \quad \Xi_{II,1}^B = \begin{cases} \phi_{AA} - \varphi \\ \phi_{AA} \\ \phi_{AA} - \varphi \end{cases}. \quad (\text{B12})$$

FIG. 9. (Left) The normalized local density of states $\tilde{D}(r, E) = D(r, E)/\max|D(r, E)|$ at the van Hove singularity of conduction/valence bands for type 1 (left two columns) and type 2 (right two columns) of tBG/BN. The electrons are highly populated at the local-AA stacking in tBG for both valence and conduction bands of type 1 and type 2 for all three slidings as seen in (a) $\tau = a_G(0, 0)$, (b) $\tau = a_G(0, 1/\sqrt{3})$, and (c) $\tau = a_G(0, 2/\sqrt{3})$. (Right) Berry curvatures of type 1 (left two columns) and type 2 (right two columns) of valence band and conduction band for case of sliding the top G layer by (d) $\tau_y = 0$, (e) $\tau_y = a_G/\sqrt{3}$, and (f) $\tau_y = 2a_G/\sqrt{3}$.

Also, A_3 and B_3 are given as follows, for type-1 AA-tBG/BN:

$$A_3 = \omega^2 C_{BB} \cos(\phi_{BB} + \varphi) + \omega^2 C_{AA} \cos(\phi_{AA}) + \omega' \omega C_{AB} \cos(\phi_{AB}), \quad (\text{B13})$$

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA} - \varphi) + \omega^2 C_{BB} \cos(\phi_{BB}) + \omega' \omega C_{AB} \cos(\phi_{AB} + \varphi) \quad (\text{B14})$$

for type-1 AB-tBG/BN,

$$A_3 = \omega^2 C_{AA} \cos(\phi_{AA} + \varphi) + \omega^2 C_{BB} \cos(\phi_{BB} - \varphi) + \omega' \omega C_{AB} \cos(\phi_{AB} - \varphi), \quad (\text{B15})$$

and

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA}) + \omega^2 C_{BB} \cos(\phi_{BB} + \varphi) + \omega' \omega C_{AB} \cos(\phi_{AB}), \quad (\text{B16})$$

and for type-1 BA-tBG/BN,

$$A_3 = \omega'^2 C_{AA} \cos(\phi_{AA} + \varphi) + \omega^2 C_{BB} \cos(\phi_{BB} - \varphi) + \omega' \omega C_{AB} \cos(\phi_{AB} - \varphi), \quad (\text{B17})$$

and

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA} - \varphi) + \omega'^2 C_{BB} \cos(\phi_{BB}) + \omega' \omega C_{AB} \cos(\phi_{AB} + \varphi). \quad (\text{B18})$$

Since A_3 and B_3 are negligibly small, we can get three roots for each, $E_C = 0$, $E_C = A_1 \pm \sqrt{A_1^2 + A_2}$, and $E_V = 0$, $E_V = B_1 \pm \sqrt{B_1^2 + B_2}$, presuming $A_3 = B_3 = 0$. By retrieving A_3 and B_3 and plugging the lowest energies $E_C = 0$, $E_V = 0$, whose eigenvectors remain the same, into the original characteristic equations, we get a much more simplified expressions for the energy gap as written in Eq. (18). In the same manner, for other stackings of tBG the characteristic equations have the same form, but the coefficients are different since the unitary matrix U connecting the spinor bases is different, resulting in different characteristic equations.

For AA-tBG/BN of type 2, the 3×3 matrix for a characteristic equation is given as

$$\begin{pmatrix} -E_C & 3e^{-i\varphi} \omega & 3\omega' \\ e^{i\varphi} \omega & 2C_{AA} \cos(\phi_{AA} + \varphi) - E_C & 2C_{AB} \cos(\phi_{AB}) e^{-i\varphi} + Q e^{i\pi/6} \\ \omega' & 2C_{AB} \cos(\phi_{AB}) e^{i\varphi} + Q e^{-i\pi/6} & 2C_{BB} \cos(\phi_{BB}) - E_C \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \quad (\text{B19})$$

$$\begin{pmatrix} -E_V & 3\omega' & 3e^{i\varphi} \omega \\ \omega' & 2C_{AA} \cos(\phi_{AA}) - E_V & 2C_{AB} \cos(\phi_{AB} - \varphi/2) e^{i\varphi/2} + Q e^{i\pi/6} \\ e^{-i\varphi} \omega & 2C_{AB} \cos(\phi_{AB} - \varphi/2) e^{-i\varphi/2} + Q e^{-i\pi/6} & 2C_{BB} \cos(\phi_{BB} - \varphi) - E_V \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \quad (\text{B20})$$

where the eigenstates of the conduction and valence bands for the eight-band model are given as $\psi_{C,2} = (0 \ c \ ae^{-i\varphi} \ b \ ae^{i\varphi} \ b \ a \ b)^T$ and $\psi_{V,2} = (c \ 0 \ a \ be^{i\varphi} \ a \ be^{-i\varphi} \ a \ b)^T$. This 3×3 matrix gives us two equations, Eqs. (B5) and (B6) as in type 1. The constants A_1 , B_1 , A_2 , and B_2 also follow the same equations Eqs. (B7) and (B8) with different Θ 's and Ξ 's as follows:

$$\Theta_{I,2}^A = \begin{cases} \phi_{AA} + \varphi \\ \phi_{AA} - \varphi \\ \phi_{AA} + \varphi \end{cases}, \quad \Theta_{I,2}^B = \begin{cases} \phi_{AA} \\ \phi_{AA} \\ \phi_{AA} - \varphi \end{cases}, \quad (B21)$$

$$\Theta_{II,2}^A = \begin{cases} \phi_{BB} \\ \phi_{BB} + \varphi \\ \phi_{BB} \end{cases}, \quad \Theta_{II,2}^B = \begin{cases} \phi_{BB} - \varphi \\ \phi_{BB} - \varphi \\ \phi_{BB} + \varphi \end{cases}, \quad (B22)$$

$$\Xi_{I,2}^A = \begin{cases} \phi_{AB} \\ \phi_{AB} + \varphi/2 \\ \phi_{AB} \end{cases}, \quad \Xi_{I,2}^B = \begin{cases} \phi_{AB} - \varphi/2 \\ \phi_{AB} - \varphi/2 \\ \phi_{AB} + \varphi/2 \end{cases}, \quad (B23)$$

$$\Xi_{II,2}^A = \begin{cases} \phi_{AA} + \varphi \\ \phi_{AA} - \varphi \\ \phi_{AA} + \varphi \end{cases}, \quad \Xi_{II,2}^B = \begin{cases} \phi_{AA} \\ \phi_{AA} \\ \phi_{AA} - \varphi \end{cases}, \quad (B24)$$

The A_3 and B_3 parameters for type-2 systems are given by

$$A_3 = \omega^2 C_{AA} \cos(\phi_{AA} + \varphi) + \omega^2 C_{BB} \cos(\phi_{BB}) + \omega' \omega C_{AB} \cos(\phi_{AB}), \quad (B25)$$

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA}) + \omega^2 C_{BB} \cos(\phi_{BB} - \varphi) - \omega' \omega C_{AB} \cos(\phi_{AB} - \varphi/2) \quad (B26)$$

for AA-tBG/BN,

$$A_3 = \omega^2 C_{AA} \cos(\phi_{AA} - \varphi) + \omega^2 C_{BB} \cos(\phi_{BB} + \varphi) - \omega' \omega C_{AB} \cos(\phi_{AB} + \varphi/2), \quad (B27)$$

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA}) + \omega^2 C_{BB} \cos(\phi_{BB} - \varphi) - \omega' \omega C_{AB} \cos(\phi_{AB} - \varphi/2) \quad (B28)$$

for AB-tBG/BN and

$$A_3 = \omega^2 C_{AA} \cos(\phi_{AA} + \varphi) + \omega^2 C_{BB} \cos(\phi_{BB}) + \omega' \omega C_{AB} \cos(\phi_{AB}), \quad (B29)$$

$$B_3 = \omega^2 C_{AA} \cos(\phi_{AA} - \varphi) + \omega^2 C_{BB} \cos(\phi_{BB} + \varphi) - \omega' \omega C_{AB} \cos(\phi_{AB} + \varphi/2) \quad (B30)$$

for BA-tBN/BN.

The analytical values of the gaps in the presence of an interlayer potential difference η will be shown only for the case of BA-tBG/BN in type 1. The associated 3×3 matrix for the characteristic equation is

$$\begin{pmatrix} \eta/2 - E_C & 3e^{i\varphi} \omega & 3e^{-i\varphi} \omega' \\ e^{-i\varphi} \omega & 2C_{AA} \cos(\phi_{AA} + \varphi) - \eta/2 - E_C & 2C_{AB} \cos(\phi_{AB} - \varphi) + Qe^{i\pi/6} \\ e^{i\varphi} \omega' & 2C_{AB} \cos(\phi_{AB} - \varphi) + Qe^{-i\pi/6} & 2C_{BB} \cos(\phi_{BB} - \varphi) - \eta/2 - E_C \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \quad (B31)$$

for the conduction band, and

$$\begin{pmatrix} \eta/2 - E_V & 3e^{-i\varphi} \omega' & 3\omega \\ e^{i\varphi} \omega' & 2C_{AA} \cos(\phi_{AA} - \varphi) - \eta/2 - E_V & 2C_{AB} \cos(\phi_{AB} + \varphi) + Qe^{i\pi/6} \\ \omega & 2C_{AB} \cos(\phi_{AB} + \varphi) + Qe^{-i\pi/6} & 2C_{BB} \cos(\phi_{BB}) - \eta/2 - E_V \end{pmatrix} \begin{pmatrix} c \\ a \\ b \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \quad (B32)$$

for the valence band where the eigenstates of the conduction and valence bands in the eight-band model are given as $\psi_{C,1} = (0 \ c \ ae^{i\varphi} \ be^{-i\varphi} \ ae^{-i\varphi} \ be^{i\varphi} \ a \ b)^T$ and $\psi_{V,1} = (c \ 0 \ ae^{-i\varphi} \ b \ ae^{i\varphi} \ b \ a \ b)^T$. This expression is the same as Eq. (18) for the gap at $\tilde{K}'$ but using with different coefficients.

APPENDIX C: SUPPLEMENTAL ELECTRONIC STRUCTURE, LOCAL DENSITY OF STATES, AND BERRY CURVATURES

Here we supplement information on electronic structure presented in the main text for the bandwidth W , the secondary band gaps δ_s , the primary band gaps δ_p , the ratio of the screened Coulomb interaction strength to the bandwidth U/W , and the valley Chern numbers by considering the effects of twist angles under the assumptions of commensurate double moiré patterns and provide information of the local density of states at the van Hove singularities and the Berry curvatures of the nearly flatbands.

We begin by presenting in Fig. 7 through one-dimensional plots various electronic structure information for variable τ_y

and fixed $\tau_x = 0$ that had been discussed in the main text. The topmost row (a) panel shows the fluctuations in the bandwidth for the low-energy conduction and valence bands as a function of τ_y , where we can find a relatively narrower conduction band versus valence band for type-1 systems and more electron-hole symmetric bands for type-2 systems. Subsequent rows represent (b) the secondary band gaps and (c) the primary band gaps. Information from (a)–(c) panels are used to determine (d) the U_{eff}/W ratio where we can clearly observe its relative enhancement for conduction bands in type-1 but maintaining comparable magnitudes for electrons and holes for type 2 systems. The valley Chern numbers in (e) showing well-quantized values or oscillating with noninteger values as a function of τ_y are in keeping with the fact that the bands are isolated when we have positive primary and secondary gaps.

In Fig. 8 we provide additional information about how the electronic structure will be modified when we allow the twist angle θ of tBG to change from the specific values determined by the double moiré commensuration condition. For this purpose we make the approximation that double commensuration is maintained for every value of θ

which implies that the moiré pattern period and angle also change continuously together with the pattern of tBG. The dashed vertical line indicates the commensuration angle θ for tBG consistent with the lattice constants of graphene $a_G = 2.461 \text{ \AA}$ and hexagonal boron nitride $a_{BN} = 2.504 \text{ \AA}$. We may expect that the double commensuration condition can still hold approximately in the vicinity of this vertical dashed line thanks to lattice relaxation effects that rotate and globally relax the lattices. The information in Fig. 8 allows to distinguish the expected electronic structures features in the lowest-energy conduction and valence bands for different initial interlayer sliding stacking geometries AA-tBG/BN [$\tau = (0, 0)$], AB-tBG/BN [$\tau = a_G(0, 1/\sqrt{3})$], and BA-tBG/BN [$\tau = a_G(0, 2/\sqrt{3})$] prior to rotation. Whereas

the relative sliding of the layers is shown to considerably modify the low-energy electronic structure as we illustrate in Fig. 9 through normalized LDOS, we also find that the electron localization largely concentrates at the AA-stacking sites of tBG regardless of the arrangement of the BN substrate. Quantitative differences in the normalized LDOS are still expected to be observable near AB or BA local stacking regions when relative sliding between layers are introduced. Likewise, the Berry curvature maps undergo changes in their hotspot distributions depending on which stacking configuration is chosen. These differences could impact the Hall transport and optical measurements especially when valley contrasting circularly polarized light is used.

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