Twisted Trilayer Graphene: a Precisely Tunable Platform for Correlated Electrons

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We introduce twisted trilayer graphene (tTLG) with two independent twist angles as an ideal system for the precise tuning of the electronic interlayer coupling to maximize the effect of correlated behaviors. As established by experiment and theory in the related twisted bilayer graphene system, van Hove singularities (VHS) in the density of states (DOS) can be used as a proxy of the tendency for correlated behaviors. To explore the evolution of VHS in the twist-angle phase space of tTLG, we present a general low-energy electronic structure model for any pair of twist angles. We show that the basis of the model has infinite dimensions even at a finite energy cutoff and that no Brillouin zone exists even in the continuum limit. Using this model, we demonstrate that the tTLG system exhibits a wide range of magic angles at which VHS merge and the DOS has a sharp peak at the charge-neutrality point through two distinct mechanisms: the incommensurate perturbation of twisted bilayer graphene’s flat bands or the equal hybridization between two bilayer moiré superlattices.

Introduction. — Electronic properties in stacked graphene layers can be tuned by a small twist angle that modifies the interlayer interaction strength, an effect referred to as “twistronics” [1]. As the twist angle approaches a critical “magic angle” ($\sim 1.05^\circ$ in twisted bilayer graphene), the two van Hove singularities (VHS) in the density of states (DOS) of each monolayer merge, resulting in a sharp peak associated with flat bands, leading to the emergence of strongly correlated electronic phases [2]. The small twist angle gives rise to large-scale repeating patterns, known as moiré patterns. Unconventional correlated states have now been observed in many van der Waals (vdW) heterostructures with one twist angle, such as twisted bilayer graphene (tBLG) and twisted double bilayer graphene [3–15]. In these systems, electrons responsible for the correlation effects localize at the moiré scale [16–18].

The addition of a third layer introduces a new degree of freedom, the second twist angle, allowing for the further tuning of electron correlations. In twisted trilayer graphene (tTLG) with two consecutive twist angles, $\theta_{12}$ and $\theta_{23}$, the beating of two bilayer moiré patterns leads to higher-order patterns (moiré of moiré). The length scale of these is orders of magnitude larger than the bilayer moiré pattern (Fig. 1a) [19–21]. Unlike in tBLG where only the lowest-order moiré pattern dominates in the continuum limit, the dominant harmonic is twist-angle dependent in tTLG. For a given moiré of moiré harmonic labelled by $(m,n)$, the primitive reciprocal lattice vectors are given as the column vectors of $G_{mn}^H = mG_{12} - nG_{23}$, where the matrix $G_{ij}$ spans the bilayer moiré reciprocal space between layers $i$ (Li) and $j$ (Lj). The real space moiré of moiré supercell $A_{mn}^H$ is obtained by $A_{mn}^H = \frac{1}{T}(G_{mn}^H)^{-T}$, and the norm of the column vectors of $A_{mn}^H$ is the moiré length. Figure 1b shows the dominant moiré length as a function of twist angles, in which each lobe corresponds to the region where a different harmonic $(m,n)$ dominates. The moiré of moiré patterns can be discerned visually only near the $(N,1)$ or $(1,N)$ lobes for $N \in \mathbb{Z}$. Generally, multiple harmonics have competing length scales (see Supplementary Material Sec. I for detail). Therefore, tTLG cannot be approximated by two aligned tBLG and a general expression for the trilayer supercell does not exist, making it fundamentally different than multi-layered vdW heterostructures with a single twist angle [22–25]. The lack of a supercell approximation and the large length scale pose many computational challenges to the theoretical modeling of tTLG. While aspects of tTLG have been studied theoretically [19, 26, 27], a electronic structure model applicable to any pair of twist angles is lacking; this severely restricts our ability to investigate its electronic properties and the potential for correlated phases, which have been observed recently in tTLG at the moiré of moiré scale [28].

Here, we present tTLG as a platform to precisely tune twistronic correlations, using the VHS intensity as a proxy for strong correlations. We derive a general momentum-space model to study the electronic properties of the two-independent-twist-angle tTLG system using a low-energy $k \cdot p$ model, which provides computational efficiency and removes the constraint on the twist angle imposed by atomistic calculations with supercells. Using this model, we explore the tTLG phase space. We find that the two bilayer moiré superlattices hybridize when the two twist angles are equal, minimizing the energy separation between the two lowest VHS at a critical twist angle. At general twist angles, there exists a wide range of values at which the VHS merge and the DOS is sharply peaked at the charge-neutrality point (CNP). These magic angles can be understood as a tBLG magic angle modified by an incommensurate perturbative po-
tential from the third layer. Our analysis is well suited to guide experimental searches for correlation effects and enables the interpretation of otherwise unclear experimental findings [28].

FIG. 1: Illustration of moiré of moiré pattern in tTLG for \(\theta_{12} = 2.6^\circ\), \(\theta_{23} = 2.8^\circ\). Red and blue points represent the lattice points of the bilayer moiré supercells between L1–L2 and L2–L3 respectively: the moiré lattice vectors (red and blue arrows) are slightly rotated and have different lattice constants. Black arrows are the dominant moiré of moiré supercell lattice vectors. A blow-up of the small boxed area is shown below, with points representing the atomic lattice points of the bilayer moiré supercells between L1–L2 is shown below, with points representing the atomic

\(\begin{align*}
\mathcal{H}(k) = &\begin{pmatrix}
H^1(k) & T^{12} & 0 \\
T^{12\dagger} & H^2(k) & T^{23} \\
0 & T^{23\dagger} & H^3(k)
\end{pmatrix}.
\end{align*}\)

The diagonal blocks are the monolayer graphene tight-binding Hamiltonians in the rotated basis [29], representing the intralayer hopping. The off-diagonal blocks represent the interlayer hopping. The interlayer terms that connect two momentum degrees of freedom \(k^{(i)}\) and \(k^{(j)}\) in Li and Lj are given as,

\(\begin{align*}
T^{ij}_{\alpha\beta}(k^{(i)}, k^{(j)}) &= \frac{1}{|\Gamma|} \sum_{G^{(i)},G^{(j)}} e^{iG^{(i)} \cdot \tau^{(i)}_{\alpha}} \times \Gamma^{ij}_{\alpha\beta}(k + k^{(i)} + G^{(i)}) e^{-iG^{(j)} \cdot \tau^{(j)}_{\beta}} \delta_{k^{(i)} - G^{(i)}, k^{(j)} - G^{(j)}},
\end{align*}\)

where \(|\Gamma|\) is the monolayer unit cell area, \(\tau^{(i)}_{\alpha}\) (\(\tau^{(j)}_{\beta}\)) is the position of the sublattice \(\alpha\) (\(\beta\)), \(G^{(i)}\) is a reciprocal space lattice vector in \(L\ell\), and \(\Gamma^{ij}_{\alpha\beta}(p)\) is the momentum space hopping parameter between sublattice \(\alpha\) in Li and sublattice \(\beta\) in Lj. The \(\delta\)-function imposes the constraint on the values of \(k^{(i)}\) in the basis set and dictates the interlayer scattering selection rule. The above expressions are equivalent to a real space tight-binding model in the Bloch basis (See Supplementary Material Sec. IIA for the detailed derivation).

Unlike twisted bilayers [30–34], the momentum space basis in tTLG is infinitely dimensional and lacks a Brillouin zone even in the continuum limit. In the bilayer case, the coupled momentum states satisfy the selection rule \(k^{(1)} - k^{(2)} = G^{(1)} - G^{(2)}\) [32]. Note that for a given \(G^{(1)} = m b_1^{(1)} + n b_2^{(1)}\) for \(m, n \in \mathbb{Z}\), we also have \(G^{(2)} = m b_1^{(2)} + n b_2^{(2)}\) for the same \(m, n\), where \(b^{(i)}_\ell\) is the \(\ell\)-th component of the primitive reciprocal lattice vector of the layer \(\ell\), since other hopping processes are much higher in energy. As the magnitude of \(G^{(i)}\) increases, the scattered momentum \(k'\) becomes farther away from the Dirac point. Therefore, to implement a finite cutoff, we can simply constrain the magnitude of the scattered momentum \(k' = G^{(i)}\) for \(\ell = 1, 2\). Physically, \(k'\) is a monolayer reciprocal lattice point that can scatter to a nearby momentum in the other layer. In contrast, in trilayers, the momentum states that form the basis of the Hamiltonian are connected in a more complicated way. A given \(k^{(1)}\) can couple to a momentum state \(k^{(2)}\) in L2 that satisfies \(k^{(2)} = k^{(1)} + G^{(2)} - G^{(1)}\), same as the bilayer case. Each \(k^{(2)}\) can then couple to a momentum state \(k^{(3)}\) in L3 through the second selection rule (Eq. (2)), resulting in the following final momentum:

\(\begin{align*}
k^{(3)} &= k^{(1)} + (G^{(2)} - G^{(1)}) + (G^{(3)} - G^{(2)}),
\end{align*}\)

where the reciprocal lattice vectors satisfy \(G^{(2)} - G^{(1)} = m b_1^{(12)} + n b_2^{(12)}\) and \(G^{(3)} - G^{(2)} = m' b_1^{(23)} + n' b_2^{(23)}\) for \(m, n, n', m' \in \mathbb{Z}\), with \(b^{(i)}_\ell\) being the bilayer moiré reciprocal space lattice vectors. Eq. (3) suggests that even though we do not allow the direct interlayer hopping between L1 and L3, they are coupled through L2. Unlike the simple 2D momentum crystal in bilayers, here the incomensuration between \(b^{(12)}_k\) and \(b^{(23)}_k\) creates for \(k^{(3)}\) a 4D structure that is projected onto 2D.

Equation (3) suggests that in \(L\ell\) of tTLG, the scattered momentum is given by \(k' = G^{(i)} + G^{(j)}\) for \(\ell \neq i, j\). To implement a cutoff, we should impose \(|k'| \leq k_c\) for some cutoff value \(k_c\). However, the incomensurability of twisted trilayers suggests that \(|k'|\) can be arbitrarily small and simply imposing \(|k'| \leq k_c\) still leads to an infinite basis. For example, in Fig. 2a, even though \(G^{(2)}\) lies outside of the cutoff radius, the resulting \(k'\) is still a relevant low-energy degree of freedom, due to the two-step scattering process. A similar construction can be made for all other \(G^{(2)}\) outside of the cutoff radius, which means within a finite energy cutoff, there are infinitely many coupled momentum states. In practice,
another set of cutoff conditions need to be implemented, namely $|G^{(\ell)}| \leq k_c$. With the constraint on $|G^{(\ell)}|$, the $k'$ in Fig. 2a, for example, is no longer allowed. In this way, we are ignoring the cases where $|G^{(\ell)}|$ is large but $|k'|$ is small, leading to the neglect of low-energy degrees of freedom and hence convergence is not guaranteed; this issue merits future work (see Supplementary Material Sec. IID for a convergence study). In this work, we choose $k_c = 4|b^{(\ell)}|$, with $\sim 5600$ momenta, such that the properties of interest (e.g., DOS maximum and the VHS location) do not change significantly as $k_c$ increases.

We take the low-energy limit by expanding around the Dirac point of each layer $K_{\text{L1}}$, letting $k^{(\ell)} = q^{(\ell)} + K_{\text{L1}}$. The intralayer Hamiltonian becomes the rotated Dirac equation, $H^\ell = v_F q \cdot (\alpha x^\ell - \alpha y^\ell)$, where $\alpha x^\ell = \sigma_x \cos \theta - \sigma_y \sin \theta$ and $\alpha y^\ell = \sigma_x \sin \theta + \sigma_y \cos \theta$ are rotated Pauli matrices with $\theta_\ell = \theta_{\text{T1L2}} = 0$, $\theta_\ell = 0$, $v_F = 0.8 \times 10^6$ cm/s is the Fermi velocity [35], and $q = k + k^{(\ell)} - K_{\text{L1}}$. For the interlayer hopping, we make the approximation that $\mathcal{I}^{ij}_{\alpha\beta}(k + k^{(\ell)} + G^{(\ell)}) \approx \mathcal{I}^{ij}_{\alpha\beta}(G^{(\ell)} + K_{\text{L1}})$ since $|k^{(\ell)}|, |q^{(\ell)}| \ll |K_{\text{L1}}|, |G^{(\ell)}|$, for a center site near the Dirac point. Due to the rapid decay of $\mathcal{I}^{ij}_{\alpha\beta}(p)$ as $p$ increases [2, 35, 36], we keep only the first shell in the summation in Eq. (2):

$$T^{ij}_{\alpha\beta}(q^{(\ell)} + q^{(j)}) = \sum_{n=1}^{3} T^{ij}_{\alpha\beta}(q^{(n)}),$$

where $q^{(i)}_1 = K_{\text{L1}} - K_{\text{L1}}$, $q^{(j)}_2 = R^{-1}(2\pi/3)q^{(j)}_1$, and $q^{(j)}_3 = R(2\pi/3)q^{(j)}_1$ using a counterclockwise rotation matrix $R(\theta)$. We include out-of-plane relaxation by letting $t^{i}_A = t^{j}_A = \omega_n = 0.07$ eV and $t^{i}_B = t^{j}_B = \omega_1 = 0.11$ eV [31, 37]. In the matrix form,

$$T^{i}_1 = \left[ \begin{array}{cc} \omega_0 & \omega_1 \\ \omega_1 & \omega_0 \end{array} \right], T^{j}_2 = \left[ \begin{array}{cc} \omega_0 & \omega_1 \phi \\ \omega_1 \phi & \omega_0 \end{array} \right], T^{i}_3 = \left[ \begin{array}{cc} \omega_0 & \omega_1 \phi \\ \omega_1 \phi & \omega_0 \end{array} \right],$$

where $\phi = \exp(i2\pi/3)$, $\bar{\phi} = \exp(-i2\pi/3)$.

In tBLG, with the low-energy expansion, momenta $q^{(1)}_i$ and $q^{(2)}$ form a hexagonal lattice in reciprocal space with the neighboring rhombus corners representing states from alternating layers (a moiré momentum lattice) [2, 32]. In tTLG, on top of each lattice point of the L1–L2 moiré momentum lattice, the additional scattering process creates a copy of the L2–L3 moiré momentum lattice (Fig. 2b), suggesting the absence of a Brillouin zone.

Density of states — We use Gaussian smearing to obtain the total DOS, summing over the two bilayer moiré Brillouin zones [38] (see Supplementary Material Sec. IIC for an explicit expression). We discretize each moiré Brillouin zone using a 22 x 22 grid. To properly normalize the DOS, we first calculate the DOS of only the intralayer Hamiltonian, which reduces to three independent copies of monolayer graphene [29]. We then obtain a normalization constant by fixing the prefactor to the expected low-energy monolayer DOS and using the same constant for the DOS of the full Hamiltonian. We adapt the Gaussian full-width-half-maximum, $\kappa$, based on the twist angle $\theta_{\ell,\ell+1}$: for $\theta_{\ell,\ell+1} \leq 2^\circ$, $\kappa = 0.35$ meV; for $\theta_{\ell,\ell+1} \in (2^\circ, 3.9^\circ)$, $\kappa = 1.2$ meV; for $\theta_{\ell,\ell+1} > 3.9^\circ$, $\kappa = 2.4$ meV.

Evolution of VHS. — We explore next the behavior of VHS as a function of twist angles in tTLG, by investigating the DOS enhancement and the narrowing of the separation between VHS (referred to as the VHS gap). We define a magic angle approximately as a geometry where both these features are achieved. Figure 3a shows the DOS of tTLG at equal twist angles. The bright regions represent VHS. As the twist angle decreases, the VHS gap first decreases and increases after reaching a minimum at $\sim 2.1^\circ$. This behavior is similar to the evolution of VHS in tBLG, in which changing the twist angle tunes the hybridization between two monolayer Dirac cones. Similarly in tTLG with $\theta_{12} = \theta_{23}$, varying the twist angle changes the hybridization strength between the two identical bilayer moiré superlattices. However, the two VHS can never merge at the CNP when $\theta_{12} = \theta_{23}$. The minimum VHS gap at $2.1^\circ$ is still around 20 meV and the DOS is orders of magnitude lower than at the tBLG magic-angle. For general twist angles, Fig. 3b shows the DOS as a function of $\theta_{12}$ with $\theta_{23} = 3^\circ$. Unlike in the equal twist angle case, the two VHS approach each other as the twist angle decreases and merge between $\theta_{12} = 1.6^\circ$ and $\theta_{12} = 1.3^\circ$, resulting in a sharp peak in the DOS.

To investigate the nature of DOS enhancements in tTLG, we performed calculations over an entire region

FIG. 2: Degrees of freedom in the momentum space basis for tTLG at $\theta_{12} = 2.2^\circ$, $\theta_{23} = 2^\circ$. Red, blue, and green are the reciprocal lattice vectors of L1, L2, and L3 respectively. The origin is chosen to be the Dirac point of L2. (a) Extended zone scheme, with the orange circle indicating the cutoff in $|k'|$. $k' = G^{(2)} + G^{(3)}$ falls within the cutoff radius 10Å $^{-1}$ despite both $|G^{(2)}|$ and $|G^{(3)}|$ being large. The momenta of L3 have their origin centered at $G^{(2)}$. (b) Reduced zone scheme, folded back to the monolayer Dirac points, $q^{(i)} = k^{(i)} - K_{\text{L1}}$. This basis of tTLG corresponds to the same twist angle as (a) but with an additional cutoff constraint $|G^{(i)}| \leq k_c = 6|b^{(i)}|$, leading to 26921 momenta.
of the \(\theta_{12}, \theta_{23}\) parameter space. Figure 4 shows the DOS maximum and the VHS gap, \(\Delta E\), as a function of both twist angles [39]. The magic-angle condition is met at a wide range of twist angles that follows a smooth curve but disappears near the diagonal. Although there is no significant DOS enhancement at \(\theta_{12} = \theta_{23}\), the DOS maximum near the diagonal is higher compared to the nearby regions where \(\theta_{12}\) and \(\theta_{23}\) differ slightly (light yellow region within the dotted lines in Fig. 4a).

We now examine the magic angle curve away from the diagonal. In the limit where \(\theta_{12} \gg \theta_{23}\) or \(\theta_{12} \ll \theta_{23}\), tTLG decomposes into a decoupled twisted bilayer moiré supercell and a graphene monolayer; the decoupled monolayer does not contribute significantly to the low-energy features. Therefore, we observe that the tTLG magic-angle curve asymptotically approaches the tBLG magic angle (dashed lines in Fig. 4a) for large \(\theta_{12}\) or \(\theta_{23}\). We checked numerically that when one twist angle is very large, \(\theta_{12} = 40^\circ\) for instance, the DOS maximum occurs exactly when the other twist angle, \(\theta_{23}\), is at the tBLG magic angle. This continuous magic angle curve and its asymptotic behavior suggest that these magic angles can be understood as the hybridization between two bilayer moiré superlattices.

The evolution of VHS along the diagonal likely has a different origin than the magic angles for \(\theta_{12} \neq \theta_{23}\). The perturbation theory argument predicts that \(v_F^*\) can reach 0 when \(\theta_{12} = \theta_{23}\), which we do not observe in Fig. 3a. The curve given by Eq. (7) intersects with the diagonal at \(\theta_{12} = \theta_{23} = 1.72^\circ\), which deviates from the twist angle with the minimal VHS gap in the numerical calculation \((2.1^\circ)\). These observations suggest that features near the diagonal are more aptly described by the hybridization between the two bilayer moiré superlattices with a shared middle layer, rather than between two independent unit cells as in the case of tBLG. Therefore, the perturbation argument does not apply.

**Moiré of moiré.** — In magic-angle tBLG, correlated states occur at the half-filling of the bilayer moiré supercell by filling the two isolated flat bands [3, 4, 16–18]. In magic-angle tTLG, even though the origin of some magic angles is perturbed tBLG, we argue that filling each flat band corresponds to filling the moiré of moiré supercell rather than the smaller bilayer moiré cell. This is because the incommensurability between the tBLG and the effective potential \(V\) changes the relevant superlattice area.

We compare our results to a simplified model, in
which tTLG is approximated as two aligned bilayer moiré cells [27]. While we observe similar qualitative behaviors, the simplified model fails to capture physics at the moiré scale and does not predict as drastic a DOS enhancement as our work. Most importantly, the simplified model requires a new basis for a different set of twist angles and it is difficult to generalize — limitations that our model overcomes. We have included a detailed comparison between the two models in Supplementary Material Section IV.

In summary, we explore the rich electronic behavior of tTLG in its twist angle phase space. We offer a general momentum space model to obtain electronic structure in tTLG at any pair of twist angles. We show that the twisted trilayer momentum space model does not have a Brillouin zone and has an infinitely-sized basis. Although we do not predict correlation strengths directly, we can use the presence of VHS as a proxy for electronic correlation. We show that the tTLG system exhibits a wide range of magic angles with merging VHS at the CNP. Away from the equal twist angle, the origin of the magic angles can be understood as tBLG in an incommensurate perturbative potential. Near the equal twist angle regime, the electronic properties are a result of the hybridization between two bilayer moiré superlattices that share the middle layer. Tuning the twist angle makes it possible to traverse between these two regimes. Our MATLAB code for the model is openly available [40].

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Supplementary Material for “Twisted Trilayer Graphene: a Precisely Tunable Platform for Correlated Electrons”

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The Supplementary Material includes four sections. In Section I, we discuss the geometry of the twisted trilayer graphene (tTLG) and calculate the higher-order moiré of moiré lengths. In Section II, we present a detailed derivation of the momentum space model and test its convergence. In Section III, we derive analytically the magic angles in tTLG. Finally, we compare and contrast our model with a simplified model proposed by Mora et al. [S1] in Section IV.

I. CALCULATION OF MOIRÉ OF MOIRÉ LENGTHS

The atomic and reciprocal space geometry of tTLG with two independent twist angles are shown in Fig. S1a. The monolayer lattice vectors are defined as the column vectors of the following matrix:

\[ A_0 = a_G \begin{bmatrix} 1 & 1/2 \\ 0 & \sqrt{3}/2 \end{bmatrix} = [a_1 \ a_2] , \]

where \( a_G = 2.4768 \) Å is the graphene lattice constant (as obtained from DFT). The \( \ell \)-th layer will be referred to as \( L_\ell \). We assume that \( L_2 \) is unrotated, with \( L_1 \) rotated clockwise by \( \theta_{12} \) and \( L_3 \) rotated counterclockwise by \( \theta_{23} \). Defining the counterclockwise rotation matrix

\[ R(\theta) = \begin{bmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{bmatrix} , \]

the lattice vectors of the three layers can be written as \( A_1 = R(-\theta_{12})A_0, A_2 = A_0, \) and \( A_3 = R(\theta_{23})A_0 \) respectively, with the column vectors denoted as \( a_i^{(\ell)} \) for \( \ell = 1,2,3 \) and \( i = 1,2 \). The monolayer reciprocal lattice vectors are given by the columns of \( G_\ell = 2\pi A_\ell^{-T} \). For example, the reciprocal lattice vectors of \( L_2 \) are \( b_1^{(2)} = 2\pi/a_G(1,-\sqrt{3}/3) \) and \( b_2^{(2)} = 2\pi/a_G(0,2/\sqrt{3}) \). The K point of \( L_2 \) is given as \( K_L^{(2)} = (2b_2^{(2)} + b_1^{(1)})/3 = (4\pi/(3a_G),0) \). The reciprocal lattice vectors of layers \( L_1 \) and \( L_3 \) can be obtained by acting \( R(-\theta_{12}) \) and \( R(\theta_{23}) \) on \( b_i^{(1)} \) for \( i = 1,2 \). We also denote the monolayer unit cell of layer \( \ell \) to be \( \Gamma^{(\ell)} \) and the reciprocal space to be \( \Gamma^{(\ell)*} \).

FIG. S1: Lattice geometries of the tTLG system. (a) The twisted trilayer graphene system in real space (left) and momentum space with their original monolayer reciprocal lattice vectors (right). (b) Bilayer moiré Brillouin zone for \( \theta_{12} = \theta_{23} \) with high symmetry points.
The twisted trilayer system exhibits higher order moiré of moiré patterns due to the interference between the two bilayer moiré patterns. To the lowest order, the bilayer moiré length between layers $i$ and $j$ is given by $a^{ij}_M = a_G / \sin \theta_{ij}$. We denote the bilayer moiré superlattice between layers $ij$ to be $\Gamma^{ij}$, spanned by the column vectors of matrix $A_{ij} = (a^{(ij)}_1 a^{(ij)}_2)$. The bilayer moiré Brillouin zone between layers $i$ and $j$ are given by the column vectors of $G_{ij} = G_j - G_i = 2\pi(A^{T}_j - A^{T}_i)$. The lattice vector of the moiré superlattice is the column vectors of $A_{ij} = 2\pi G^{-T}_{ij}$. After some algebra, we obtain the lattice vectors for the two bilayer supercells, $a^{12}_i, a^{23}_i$ for $i = 1, 2$:

$$a^{12}_i = \frac{a_G}{2(1 - \cos \theta_{12})} \left[ (1 - \cos \theta_{12}) \hat{x} - \sin \theta_{12} \hat{y} \right],$$

$$a^{23}_i = \frac{a_G}{2(1 - \cos \theta_{23})} \left[ (1 - \cos \theta_{23}) \hat{x} + \sin \theta_{23} \hat{y} \right] ,$$

$$a^{12}_1 = \frac{a_G}{2(1 - \cos \theta_{12})} \left[ \frac{1 - \cos \theta_{12}}{2} + \frac{\sqrt{3}}{2} \sin \theta_{12} \right] \hat{x} + \left( -\frac{1}{2} \sin \theta_{12} + \frac{\sqrt{3}}{2} \left(1 - \cos \theta_{12}\right) \right) \hat{y} ,$$

$$a^{23}_2 = \frac{a_G}{2(1 - \cos \theta_{23})} \left[ \frac{1 - \cos \theta_{23}}{2} - \frac{\sqrt{3}}{2} \sin \theta_{23} \right] \hat{x} + \left( \frac{1}{2} \sin \theta_{23} + \frac{\sqrt{3}}{2} \left(1 - \cos \theta_{23}\right) \right) \hat{y} .$$

Note that there is a small twist angle between the two bilayer moiré superlattices. Moreover, for general twist angles $\theta_{12} \neq \theta_{23}$, the two bilayer moiré cells have a lattice mismatch. The twist angle $\phi$ and the lattice mismatch $\delta$ between the two bilayer moiré patterns give rise to higher-order moiré of moiré lengths. The primitive reciprocal lattice vectors of a given harmonic $(m, n)$ are given as the column vectors of $G_{mn}^{H} = mG_{12} - nG_{23}$. Inverting $G_{mn}^{H}$, we obtain the moiré of moiré supercell in real space $A_{mn}^{H} = \frac{1}{2\pi}(G_{mn}^{H})^{-T}$. The norm of the column vectors are the moiré of moiré lengths, denoted as $\lambda_{mn}^{H}$. For $m = n = 1$, the moiré of moiré length $\lambda_{11}^{H}$ is explicitly given as

$$\lambda_{11}^{H} = \frac{(1 + \delta) a_{12}^M}{\sqrt{2(1 + \delta)(1 - \cos \phi) + \delta^2}}, \quad (S4)$$

where $\phi = \cos^{-1} \left( \frac{a^{(12)}_1}{a^{(23)}_1} \right)$ is the twist angle between the bilayer moiré supercells and $\delta = \frac{\sin \theta_{23}}{\sin \theta_{12}} - 1$ is the lattice mismatch between the two bilayer moiré supercells such that $a_{12}^M = (1 + \delta) a_{12}^H$. Eq. (S4) agrees with the first-order approximation for the moiré length for a twisted bilayer with a lattice mismatch $[S2]$, with the lattice constant being the bilayer moiré length between L1 and L2.

A dominant moiré of moiré length does not necessarily exist nor evolve smoothly under the continuous change of twist angle. To see this, we will consider different harmonics of the higher order moiré patterns. To find the dominant harmonic for an arbitrary pair of twist angles, we calculate $A_{mn}^{H}$ for $|m|, |n| \leq 15$ numerically and find the $(m, n)$ such that the norm of $G_{mn}^{H}$ is the smallest, or, equivalently, that the moiré of moiré length $\lambda_{mn}^{H}$ is largest. We are neglecting the cases where higher order harmonics dominate, such as the cases where $\theta_{12}$ and $\theta_{23}$ are different by more than a factor of 15. In those cases, the two bilayers moiré supercells have very different sizes and become essentially decoupled, which is not the focus of our study. Figure S2a shows the moiré of moiré harmonics for varying $\theta_{12}$ at a fixed $\theta_{23} = 2.8^\circ$, indicating the non-smooth dependence of the dominant moiré of moiré length on the twist angle. In tTLG, there exists a supercell approximation when there is a clear dominant harmonic, that is when $\theta_{12} \approx N\theta_{23}$ or $\theta_{23} \approx N\theta_{12}$ for $N \in \mathbb{Z}$. For example, at $\theta_{12} = 2.6^\circ, \theta_{23} = 2.8^\circ$, the dominant harmonic is $(1, 1)$ (see Fig. S2b) and at $\theta_{12} = 1.35^\circ, \theta_{23} = 2.8^\circ$, the dominant harmonic is $(2, 1)$ (see Fig. S2d). However, there are cases where there is no clear dominant moiré of moiré. For example, in Fig. S2c, it is difficult to visually discern a large repeating pattern and the estimated moiré of moiré lattice vectors fail to capture the relevant length scale. This is because near $\theta_{12} = 1.8^\circ$, many harmonics, such as $(3, 2), (5, 4)$, and $(5, 3)$, all have comparable lengths (see Fig. S2a the corresponding point).

II. MOMENTUM SPACE MODEL

In this section, we offer a detailed derivation of our momentum space model and density of states formalism, and study the convergence as a function of the momentum space cutoff radius.
A. Detailed derivation of momentum space model

To model the electronic structure of the tTLG system, we start from a tight-binding approximation for each individual layer; we take into account the interlayer hopping in a transverse tight-binding approximation between nearest-neighbors. We start by writing the Hamiltonian for the trilayer as a sum of the following terms

\[ H = \sum_{\ell=1}^{3} H^{\ell} + \sum_{l=1,2} (V^{\ell,\ell+1} + V^{\ell+1,\ell}), \]

where \( H^{\ell} \) is the Hamiltonian for the \( \ell \)-th layer and \( V^{ij} \) describes the interlayer hopping. For simplicity, we only consider the interlayer couplings between adjacent layers. DFT calculations predict that the interlayer coupling between L1 and L3 is roughly 10 times smaller than the coupling between adjacent layers (e.g., between L1 and L2) [S3]. In a second quantized notation, \( H^{\ell} \) can be written as

\[ H^{\ell} = -t \sum_{R^{(\ell)}} c_{\ell,A}^{\dagger}(R^{(\ell)}) c_{\ell,B}(R^{(\ell)}) + c_{\ell,B}(R^{(\ell)} - a_{1}^{(\ell)}) + c_{\ell,B}(R^{(\ell)} - a_{2}^{(\ell)}) + h.c., \]

where \( c_{\ell,\alpha}^{\dagger} \) and \( c_{\ell,\alpha} \) are the creation and annihilation fermionic operators of the orbital \( \alpha \) in layer \( l \), \( a_{1,2}^{(\ell)} \) are the lattice vectors of layer \( l \), and \( t \) is the hopping parameter between nearest-neighbors. As for the interlayer coupling, we define the following overlap matrix element in the tight-binding basis

\[ t_{ij}^{\alpha\beta}(R^{(i)}, R^{(j)}) = \langle i, R^{(i)}, \alpha | H | j, R^{(j)}, \beta \rangle, \]
where $\alpha$ and $\beta$ denotes the sublattice degree of freedom. The interlayer Hamiltonian in the second-quantized notation is

$$V^{ij} = \sum_{R^{(i)},\alpha,R^{(j)},\beta} c^\dagger_{i,\alpha}(R^{(i)}) t^{ij}_{\alpha\beta}(R^{(i)}, R^{(j)}) c_{j,\beta}(R^{(j)}). \quad (S8)$$

We obtain the Hamiltonian in the momentum basis at a center site momentum $\mathbf{k}$. Defining $Q^{(l)} = \mathbf{k}^{(l)} + \mathbf{k}$ for $\mathbf{k}^{(l)} \in \Gamma^{(l)*}$. We perform the Fourier transform as follows

$$c^\dagger_{\ell,\alpha}(R^{(l)}) = \frac{1}{\sqrt{\mid \Gamma^{(l)*} \mid}} \int_{\Gamma^{(l)*}} d\mathbf{k}^{(l)} e^{iQ^{(l)}(\mathbf{R}^{(l)} + \tau^{(l)}_{\alpha})} c^\dagger_{\ell,\alpha},$$

$$c_{\ell,\alpha}(R^{(l)}) = \frac{1}{\sqrt{\mid \Gamma^{(l)*} \mid}} \int_{\Gamma^{(l)*}} d\mathbf{k}^{(l)} e^{-iQ^{(l)}(\mathbf{R}^{(l)} + \tau^{(l)}_{\alpha})} c_{\ell,\alpha}, \quad (S9)$$

where the integral is over the Brillouin zone of the $\ell$-th layer, $\tau^{(l)}_{\alpha} = 0$, $\tau^{(l)}_{\beta} = 1/3(a^{(l)}_1 + a^{(l)}_2)$. The inverse of the transform in Eq. (S9) is

$$c^\dagger_{\ell,\alpha}(R^{(l)}) = \frac{1}{\sqrt{\mid \Gamma^{(l)*} \mid}} \sum_{R^{(l)}} e^{-iQ^{(l)}(\mathbf{R}^{(l)} + \tau^{(l)}_{\alpha})} c^\dagger_{\ell,\alpha}(R^{(l)}),$$

$$c_{\ell,\alpha}(R^{(l)}) = \frac{1}{\sqrt{\mid \Gamma^{(l)*} \mid}} \sum_{R^{(l)}} e^{iQ^{(l)}(\mathbf{R}^{(l)} + \tau^{(l)}_{\alpha})} c_{\ell,\alpha}(R^{(l)}), \quad (S10)$$

where $\mid \Gamma^{(l)*} \mid$ is the area of the Brillouin zone in the $\ell$-th layer. The intralayer Hamiltonian in the Bloch basis can now be written as follows

$$H^{\ell} = -\frac{t}{\mid \Gamma^{(l)*} \mid} \sum_{R^{(l)}} \int_{\Gamma^{(l)*}} d\mathbf{k}^{(l)} \int_{\Gamma^{(l)*}} d\mathbf{k}^{(l)} e^{i(k^{(l)} - k^{(l)}) \cdot \mathbf{R}^{(l)}} \sum_{s^{(l)}} e^{ik \cdot s^{(l)}} c^\dagger_{\ell,k^{(l)},\alpha} c_{\ell,k^{(l)},\beta},$$

$$= -\frac{t}{\mid \Gamma^{(l)*} \mid} \sum_{R^{(l)}} \int_{\Gamma^{(l)*}} d\mathbf{k}^{(l)} \sum_{s^{(l)}} e^{iQ^{(l)} \cdot s^{(l)}} c^\dagger_{\ell,k^{(l)},\alpha} c_{\ell,k^{(l)},\beta}, \quad (S11)$$

where we use the Poisson summation formula, $\sum_{R^{(l)}} e^{ik^{(l)} \cdot R^{(l)}} = \mid \Gamma^{(l)*} \mid \sum_{G^{(l)}} \delta_{k^{(l)} \cdot G^{(l)}}$. We also define $s^{(l)}$ to describe the nearest neighbor separations between $A$ and $B$ sublattices, which are given as $s^{(l)}_1 = 1/3(a^{(l)}_1 + a^{(l)}_2)$, $s^{(l)}_2 = 1/3(-2a^{(l)}_1 + a^{(l)}_2)$, $s^{(l)}_3 = 1/3(a^{(l)}_1 - 2a^{(l)}_2)$. The intralayer Hamiltonian in the basis of $c_{\ell,k^{(l)},\alpha}$ can then be written as

$$H^{\ell}(Q^{(l)}) = -t \begin{bmatrix} 0 & f^{\ell}_{Q^{(l)}} \\ f^{\ell}_{Q^{(l)}}^\dagger & 0 \end{bmatrix}, \quad (S12)$$

where $f^{\ell}_{Q^{(l)}} = \sum_{s^{(l)}} e^{iQ^{(l)} \cdot s^{(l)}}$. The Hamiltonian is equivalent to the monolayer graphene tight-binding model at a given momentum $Q^{(l)}$. For the intralayer Hamiltonian, there is no constraint on $Q^{(l)}$.

Similarly, we write the interlayer Hamiltonian in the $c^\dagger_{\ell,k^{(l)},\alpha}$ basis

$$V^{ij} = \int_{\Gamma^{(i)*}} \int_{\Gamma^{(j)*}} d\mathbf{k}^{(i)} d\mathbf{k}^{(j)} \sum_{\alpha\beta} c^\dagger_{\ell,k^{(i)},\alpha} T^{\alpha\beta}_{ij}(k^{(i)}, k^{(j)}) c_{\ell,k^{(j)},\beta}, \quad (S13)$$

where we use Eq. (S9) and

$$T^{\alpha\beta}_{ij}(k^{(i)}, k^{(j)}) = \frac{1}{\sqrt{\mid \Gamma^{(i)*} \mid \mid \Gamma^{(j)*} \mid}} \sum_{R^{(i)},R^{(j)}} e^{iQ^{(i)} \cdot (R^{(i)} + \tau^{(i)}_{\alpha})} t^{ij}_{\alpha\beta}(R^{(i)}, R^{(j)}) e^{-iQ^{(j)} \cdot (R^{(j)} + \tau^{(j)}_{\beta})}. \quad (S14)$$

We now apply the two center approximation

$$t^{ij}_{\alpha\beta}(R^{(i)}, R^{(j)}) = t^{ij}_{\alpha\beta}(R^{(i)} + \tau^{(i)}_{\alpha} - R^{(j)} - \tau^{(j)}_{\beta}). \quad (S15)$$
and write the interlayer coupling in terms of a two-dimensional Fourier Transform

\[ t_{ij}^{\alpha\beta}(R(i), R(j)) = t_{ij}^{\alpha\beta}(R(i) + \tau_{ij}^{(i)} - \tau_{ij}^{(j)}) \]

\[ = \int \frac{dp}{(2\pi)^2} e^{i p \cdot (R(i) + \tau_{ij}^{(i)} - R(j) - \tau_{ij}^{(j)})} \tilde{t}_{ij}^{\alpha\beta}(p). \]  

(S16)

Plugging Eq. (S16) into Eq. (S14), the interlayer coupling matrix element in momentum space is

\[ T_{\alpha\beta}^{ij}(k(i), k(j)) = \frac{1}{\sqrt{|\Gamma(i)| |\Gamma(j)|}} \int \frac{dp}{(2\pi)^2} \sum_{R(i), R(j)} e^{i (Q(i) + p - R(i)) \cdot \tau_{ij}^{(i)}} t_{ij}^{\alpha\beta}(p) e^{-i (Q(j) + p - R(j) - \tau_{ij}^{(j)})} \]

\[ = \sqrt{|\Gamma(i)| |\Gamma(j)|} \sum_{G^{(i)}, G^{(j)}} \int \frac{dp}{(2\pi)^2} e^{i G^{(i)} \cdot \tau_{ij}^{(i)}} \tilde{t}_{ij}^{\alpha\beta}(p) e^{-i G^{(j)} \cdot \tau_{ij}^{(j)}} \delta_{k + k(i) - p, G^{(i)}} \delta_{k + k(j) - p, G^{(j)}} \]

\[ = \frac{1}{|\Gamma|} \sum_{G^{(i)}, G^{(j)}} e^{i G^{(i)} \cdot \tau_{ij}^{(i)}} t_{ij}^{\alpha\beta}(k(i) + k - G^{(i)}) e^{-i G^{(j)} \cdot \tau_{ij}^{(j)}} \delta_{k(i) - G^{(i)}, k(j) - G^{(j)}}. \]  

(S17)

In the last step, we use the Poisson summation rule and \(|\Gamma(i)| = 4\pi^2 |\Gamma|^{-1}\), where \(|\Gamma|\) is the monolayer unit cell area. We have obtained the scattering selection rule \(k(i) - G^{(i)} = k(j) - G^{(j)}\) for \(i = j \pm 1\), which imposes the constraint on the values of allowed \(k(i)\).

Combining the intralayer and interlayer terms, the Hamiltonian in the \(c^3_{L,k^{(i)},\alpha}\) basis can be represented as a \(3 \times 3\) block given in Eq. (1) of the main text.

B. Low-energy limit

We can greatly simplify the model by taking the low-energy limit. Each \(H^l\) can be expanded around its Dirac point, \(k^{(l)} = K_{L^l} + q^{(l)}\), as a rotated Dirac Hamiltonian \(H_D^l(q)\) for \(q = k + k^{(l)} - K_{L^l}\):

\[ H^1(k) \approx H_D^1(q) = v_F \begin{bmatrix} 0 & e^{i q_+ q_+} \\ e^{-i q_+ q_-} & 0 \end{bmatrix}, \]

\[ H^2(k) \approx H_D^2(q) = v_F \begin{bmatrix} 0 & q_+ \\ q_- & 0 \end{bmatrix}, \]

\[ H^3(k) \approx H_D^3(q) = v_F \begin{bmatrix} 0 & e^{-i q_+ q_+} \\ e^{i q_+ q_-} & 0 \end{bmatrix}, \]  

(S18)

where \(q_{\pm} = (q_x \pm iq_y)\). For the interlayer coupling, we substitute \(k^{(l)} = q^{(l)} + K_{L^l}\) into Eq. (S17),

\[ T_{\alpha\beta}^{ij}(q^{(i)}, q^{(j)}) = \frac{1}{|\Gamma|} \sum_{G^{(i)}, G^{(j)}} e^{i G^{(i)} \cdot \tau_{ij}^{(i)}} \tilde{t}_{ij}^{\alpha\beta}(k + K_{Li} + q^{(i)} + G^{(i)}) e^{-i G^{(j)} \cdot \tau_{ij}^{(j)}} \delta_{q^{(i)}, K_{Li} + G^{(i)}, q^{(j)} + K_{Lj} - G^{(j)}}. \]  

(S19)

For momenta near the Dirac point, since \(|q^{(i)}|, |k| \ll |K_{Li}|, |G^{(i)}|\) we can approximate \(\tilde{t}_{ij}^{\alpha\beta}(k + K_{Li} + q^{(i)} + G^{(i)}) \approx \tilde{t}_{ij}^{\alpha\beta}(K_{Li} + G^{(i)})\). This approximation can lead to the suppression of particle-hole asymmetry in the tight-binding model [S5, S6]. Due to the rapid decay of the hopping parameter \(\tilde{t}(p)\) as \(p\) increases [S7], we keep only the first shell in the summation in Eq. (S19):

\[ T_{\alpha\beta}^{ij}(q^{(i)}, q^{(j)}) = \sum_{n=1}^{3} T_{n,\alpha\beta}^{ij} \delta_{q^{(i)}, -q^{(j)}, -q_1^{(i)}}. \]  

(S20)

where \(q_1^{(i)} = K_{Li} - K_{Lj}, q_2^{(i)} = R^{-1}(2\pi/3)q_1^{(i)}, \) and \(q_3^{(i)} = R(2\pi/3)q_1^{(i)}\) (see Fig. S1b). We include out-of-plane relaxation by letting \(t_{ij}^{AA} = t_{ij}^{BB} = \omega_0 = 0.07 \text{eV}\) and \(t_{ij}^{AB} = t_{ij}^{BA} = \omega_1 = 0.11 \text{eV}\), which matches with the interlayer coupling in
FIG. S3: Top: DOS obtained using different sizes of momentum space basis, $n_k$. Bottom: errors in the DOS corresponding to the vertical lines on the top with the same color. The error is defined as the difference between the DOS value at the given energy and the DOS at the largest cutoff shown on the top. The dashed lines are guides to the eyes showing power law scaling of the error as a function of $n_k$.

Nam and Koshino [S8] and Carr et al. [S5]. In matrix form,

$$T_{ij}^1 = \begin{bmatrix} \omega_0 & \omega_1 \\ \omega_1 & \omega_0 \end{bmatrix}, T_{ij}^2 = \begin{bmatrix} \omega_0 & \omega_1 \bar{\phi} \\ \omega_1 \phi & \omega_0 \end{bmatrix}, T_{ij}^3 = \begin{bmatrix} \omega_0 & \omega_1 \phi \\ \omega_1 \bar{\phi} & \omega_0 \end{bmatrix},$$  \hspace{1cm} (S21)

where $\phi = \exp(i \frac{2\pi}{3})$, $\bar{\phi} = \exp(-i \frac{2\pi}{3})$.

C. Density of States

The DOS at a given energy $\epsilon$, $D(\epsilon)$, for an incommensurate tight-binding model is defined as [S9]

$$D(\epsilon) = \sum_r \frac{1}{N} \sum_{n=1}^N \delta(\epsilon - \epsilon_n) |\psi_n(r)|^2,$$  \hspace{1cm} (S22)

where the $r$ sum is over all real space lattice positions, $n$ is the band index, and $\psi_n(r)$ is the corresponding eigenfunction. To obtain the density of states numerically, we use a Gaussian function $\phi_{\epsilon, \kappa}(x) = \frac{2\sqrt{\ln 2}}{\sqrt{\pi\kappa}} \exp\left[-4 \ln 2 \frac{(x-\epsilon)^2}{\kappa^2}\right]$ to approximate the $\delta$-function, and $\kappa$ is the full-width-half-maximum of the Gaussian, which determines the energy resolution of the DOS [S10]. We can transform the DOS equation to momentum space:

$$D(\epsilon) = \frac{N}{2} \sum_{\alpha=A,B} \sum_{\ell=1,2} \int_{\Gamma(\ell,\ell+1)} \sum_n \phi_{\epsilon, \kappa}(\epsilon_n, k) |\psi_n, k|^2 \, dk,$$  \hspace{1cm} (S23)

where $N$ is a normalization constant, $\epsilon_n, k$ is an energy within the energy window $[\epsilon - \Delta \epsilon/2, \epsilon + \Delta \epsilon/2]$, $\Delta \epsilon$ is the energy interval, $\psi_{n,k}$ and $\epsilon_{n,k}$ is an eigen-pair of the Hamiltonian $H(k)$ in Eq. (1) of the main text associated with the center site $k$ and band $n$. The integral is evaluated over the bilayer moiré Brillouin zone between layers $\ell$ and $\ell+1$, $\Gamma(\ell,\ell+1)^*$, and we discretize $\Gamma(\ell,\ell+1)^*$ using a $22 \times 22$ grid to evaluate the integral. We adapt $\kappa$ based on the area of the integration domain $\Gamma(\ell,\ell+1)^*$ as $\theta_{\ell,\ell+1}$ changes.

In order to make a direct comparison between the DOS at different twist angles, we need to properly normalize the DOS. For a given cutoff radius, we first calculate the DOS of the intralayer Hamiltonian only, which reduces to
three independent copies of monolayer graphene. Near the charge-neutrality point, the DOS per eV per nm$^2$ is given by [S4]

$$D(\epsilon) = \frac{6}{\pi} \frac{|\epsilon|}{v_F^2},$$  \hspace{1cm} (S24)

where the prefactor includes a factor 3 from the number of layers as well as a factor of 4 from spin and valley degeneracies. We then obtain a normalization constant by fixing the prefactor to the expected slope given in Eq. (S24) and use the same constant for the DOS of the full Hamiltonian.

D. Convergence

The incommensurability of the tTLG system leads to an infinite number of coupled momenta within any finite cutoff radius. Due to the additional constraints we impose on the magnitude of $G^{(\ell)}$, we neglect degrees of freedom that can contribute to the low energy states. As a result, there is no guaranteed convergence. Figure S3a-d shows the DOS and the corresponding errors for different numbers of momentum degrees of freedom for tTLG with two different sets of twist angles. In both cases, as the cutoff increases, the error does not decay significantly. Note that in the case of $\theta_{12} = 2^\circ$, $\theta_{23} = 2.4^\circ$, the drop in error is most likely a numerical artifact and further increasing the cutoff will not likely to reduce the error. However, the physically relevant features, such as the magnitude of the DOS maximum and the positions of the VHS, are relatively stable as the cutoff increases. In contrast, Fig. S3e, f shows the fast convergence of the DOS in tBLG as a function of cutoff radius. This is because in tBLG, increasing the cutoff radius does not change the number of relevant low-energy degrees of freedom. In this work, we choose a cutoff at the 4th honeycomb shell (i.e., $k_c = 4 |\ell^{(\ell)}|$ corresponding to $\sim 5,600$ momenta). This choice was made by considering both computational efficiency and the accuracy of physical properties of interest.

III. EFFECTIVE HAMILTONIAN AND RENORMALIZED FERMI VELOCITY

We examine the limit in which the momentum-space is truncated at the first honeycomb shell. The truncation gives rise to the following $14 \times 14$ Hamiltonian:

$$\mathcal{H}(q) = \begin{pmatrix}
H_D^1(q + q_1^{12}) & H_D^2(q + q_1^{12}) & (T_1^{12})^\dagger \\
T_1^{12} & T_2^{12} & H_D^3(q + q_2^{23}) \\
H_D^3(q) & T_1^{23} & T_2^{23} & T_3^{23} \\
(H_D^3(q + q_2^{23}))^\dagger & (T_1^{23})^\dagger & (T_2^{23})^\dagger & (T_3^{23})^\dagger
\end{pmatrix}. \hspace{1cm} (S25)
$$

This Hamiltonian acts on seven two-component spinors $\Psi = (\psi_{11}, \psi_{12}, \psi_{13}, \psi_{20}, \psi_{31}, \psi_{32}, \psi_{33})$, where $\ell$ and $j$ in $\psi_{\ell j}$ denote the layer and the momentum basis index respectively. Using this Hamiltonian, we can derive an expression for the renormalized Fermi velocity $v_F^*$. We first define the dimensionless quantities $\alpha_{12} = \omega/v_F k_{\theta_{12}}$ and $\alpha_{23} = \omega/v_F k_{\theta_{23}}$, where $k_{\theta_{ij}} = \frac{8\pi \sin(\theta_{ij}/2)}{3a_0}$. For simplicity, we assume $\omega_0 = \omega_1 = \omega$ and neglect the angular dependence in $H_D^\ell$ by letting $H_D^\ell$ be an unrotated Dirac Hamiltonian: $H_D^\ell(q) = v_F \sigma \cdot q$, where $\sigma = (\sigma_x, -\sigma_y)$ is the Pauli matrix. The zero-energy state of the Hamiltonian satisfies $\mathcal{H}\Psi = \sum_{j=1}^7 c_j \Psi_j = 0$, where $c_j$ is the column vectors of $\mathcal{H}$, and $\Psi_j$ is the $j$-th component of the spinor $\Psi$. Therefore, we obtain the following relation between components of $\Psi$

$$\Psi_j = -(H_D^\ell)^{-1}T^4\psi_{20}, \hspace{1cm} (S26)$$
where \( j \neq 4 \) (\( \Psi_j \) is not a state on \( L_2 \) or \( \psi_{20} \)). Using this, the effective Hamiltonian to the leading order in \( q \) is

\[
\langle \Psi | H_D^2(q) | \Psi \rangle = \frac{v_F^*}{1 + 6(\alpha_{12}^2 + \alpha_{23}^2)} \psi_{20}^\dagger \left\{ \sigma \cdot q + \sum_{n=1}^{3} T_n^{12}(H_D(q + q_n^{12}))^{-1}(\sigma \cdot q)(H_D(q + q_n^{12}))^{-1}T_n^{12} \right\} \psi_{20} 
+ T_n^{23}(H_D^3(q + q_n^{23}))^{-1}(\sigma \cdot q)(H_D^3(q + q_n^{23}))^{-1}T_n^{23} \right\} \psi_{20},
\]

where the renormalized Fermi velocity \( v^* \) is

\[
\frac{v_F^*}{v_F} = \frac{1 - 3(\alpha_{12}^2 + \alpha_{23}^2)}{1 + 6(\alpha_{12}^2 + \alpha_{23}^2)}. \tag{S28}
\]

Fig. S4a shows the \( v_F^* \) to \( v_F \) ratio as a function of \( \theta_{12} \) at a few values of \( \theta_{23} \). As \( \theta_{23} \) increases, the \( v_F^* / v_F \) ratio approaches the tBLG curve. Fig. S4b shows \( v_F^* / v_F \) for equal twist angles, which shows that perturbation theory predicts that \( v_F^* \) can still go to zero at 1.72°. However, in our numerical calculation using the full Hamiltonian, we do not observe a complete flattening of bands at this twist angle.

Finally, we show that our assumption in the analytic calculation of an unrotated Dirac Hamiltonian for the intralayer Hamiltonian and \( \omega_0 = \omega_1 \) does not significantly change the magic angle estimates. Fig. S5 compares the \( v_F^* \) obtained analytically and numerically and show that the two curves and the magic angle do not differ significantly. In the numerical calculation, we diagonalize the 14 \( \times \) 14 Hamiltonian with rotated Dirac equation for the intralayer terms and \( \omega_0 = 0.07 \text{ eV}, \omega_1 = 0.11 \text{ eV} \) for the interlayer terms. At \( \theta_{23} = 2.5^\circ \), the magic angle obtained analytically and numerically differ by 1.1%.

**IV. COMPARISON WITH A SIMPLIFIED MODEL**

In this section we compare our results with the model proposed by Mora et al. [S1] and use it to gain further insights into our findings. In this alternate model, a different momentum space basis is used by aligning the two bilayer moiré Brillouin zones (Fig. S6). This approximation ignores the incommensurability of the system, making a two-dimensional momentum space crystal with the periodicity of the bilayer moiré Brillouin zone. As a result, the problem’s complexity reduces to that of a bilayer. Formally, the Hamiltonian can still be written as the 3 \( \times \) 3 block as in Eq. (1) in the main text, but the size of the basis is reduced to be on the same order as tBLG. We implemented two cases: (1) \( \theta_{12} = \theta_{23} \) and (2) \( 2\theta_{12} = \theta_{23} \). Fig. S7 shows the momentum space basis for these two cases. In case (2), the larger bilayer Brillouin zone (L1, L2) is folded onto the smaller Brillouin zone (L2, L3) in momentum space. This
FIG. S5: Comparison between the renormalized Fermi velocity $v_F^*$ of the Hamiltonian in Eq. (S25) calculated analytically (solid line) and numerically (dashed line).

FIG. S6: Comparison of the bilayer moiré Brillouin zone geometry between our model and Mora et al. [S1] model with $\theta_{12} = \theta_{23}$. Left: two bilayer moiré Brillouin zones are misaligned by a small twist angle; right: the two bilayer moiré Brillouin zones are approximated to be aligned.

model essentially describes a system consisted of $2 \times 2$ L1, L2 moiré supercell and a L2, L3 moiré supercell. Fig. S9 shows a comparison between the DOS obtained from the two models. We keep the values of $\omega_0$ and $\omega_1$ the same as our model and use the same approach to normalize the DOS for a direct comparison. We cutoff at the 4th shell and use a grid size $22 \times 22$ for the density of states. The Gaussian FWHM we use is 5 meV for $\theta < 2^\circ$ and 8 meV for $\theta \geq 2^\circ$, where $\theta$ is the twist angle that determines the size of the Brillouin zone.

For $\theta_{12} = \theta_{23}$, Fig. S8a shows the DOS obtained with the simplified model, which agrees qualitatively with the DOS from our model (Fig. 3a of the main text). However, here the DOS has the sharpest peak between $1.7^\circ$ and $1.8^\circ$, and at $2.1^\circ$ the VHS have a larger width compared to our model. Fig. S8b-d also show that the location of peaks away from the CNP are also very different from our model.

For $2\theta_{12} = \theta_{23}$, the two models predict similar trend for the VHS evolution, and the simplified model makes the right prediction for the magic angle. This is expected from perturbation theory, since the magic angle condition does not rely on the existence of a moiré of moiré cell (as was shown in Section III). However, the magnitude of the DOS differs significantly between the two models. This is because there are two flat bands near the CNP in the simplified model, whereas in our full model, there is a large number of nearly overlapping flat bands due to incommensurability (Fig. S10). Fig. S10 compares the band structure from our model and the simplified model. The two band structures are qualitatively similar but our model shows a large number of bands due to the lack of a periodic Brillouin zone. Furthermore, the aligned-bilayer approximation will exclude correlated phases that depend on band-hybridization or symmetries from the moiré of moiré length scale. Note that we do not plot the relative layer weights (color) of the band structure in the simplified model because of the way that the Brillouin zone is wrapped — the L1 degrees of
FIG. S7: The momentum degrees of freedom in the low-energy limit of the simplified model for (a) \(\theta_{12} = \theta_{23}\) and (b) \(2\theta_{12} = \theta_{23}\).

FIG. S8: DOS obtained with the Mora et al. [S1] model DOS on a logarithmic color scale at equal twist angles (same color scale as Fig. 3a in the main text for a direct comparison) (b)-(d) DOS states along the black dashed line in (a) for (b) \(\theta = 1.8^\circ\), (c) \(\theta = 2.1^\circ\), (d) \(\theta = 2.5^\circ\), where the black solid lines are obtained using the Mora et al. [S1] model, and the blue dashed lines are obtained using the full model.

freedom are wrapped on top of the L3 degrees of freedom. Therefore, the wavefunction weights from the two models are not directly comparable for this particular high symmetry line cut.

We can use these results to further support our argument of bilayer moiré hybridization at equal twist angles. In this simplified model, sharpest VHS occur between 1.7° and 1.8°, which is in better agreement with the magic angle prediction from perturbation theory. In our model, the sharpest peak and the narrowest width occurs at a larger angle (2.1°). If this phenomenon is caused by moiré hybridization, the simplified model would not have it since it does not have the moiré of moiré scale. Indeed, the DOS from the two models differ most significantly at 2.1° (see Fig. S8c).

As we argue in the main text, adding electrons from the CNP at a low carrier concentration on the order of the tTLG moiré of moiré cells fills one flat band near the CNP in Fig. S10a at a time. Injecting electrons at a carrier concentration comparable to the bilayer moiré cell density would fill all these flat bands near the CNP. The simplified
FIG. S9: DOS as a function of $\theta_{12}$ with $2\theta_{12} = \theta_{23}$ using (a) the full model and (b) the Mora et al. [S1] model, both on a logarithmic color scale.

FIG. S10: Comparison of band structures and DOS at $\theta_{12} = 1.4^\circ, \theta_{23} = 2.8^\circ$ between our model (top) and the Mora et al. [S1] model (bottom) along the green dashed line in Fig. S1b. In (a), colors represent the weight of the wavefunctions at the center site. Red, blue, and green represents weights purely from L1, L2, L3 respectively, and colors in between represent hybridization between different layers. A colormap is provided on the top left corner. Note that even though the high symmetry points in (a) and (c) have the same label, the high symmetry lines are not exactly the same due to the different Brillouin zone geometries in the two models. The densities of the states of the from the two models are shown on the same scale.
model can again be used to understand this argument. The model also predicts some band flattening at certain twist angles, but there are only two flat bands near the CNP (Fig. S10c). Filling electrons to these two bands is equivalent to filling the bilayer moiré cell, since their momentum space basis has the periodicity of bilayer moiré Brillouin zone and there is no moiré of moiré length in this model. These two flat bands near the CNP can be qualitatively considered as the limit where all the flat bands from our model overlap exactly on top of each other. Therefore, in terms of filling the supercell, filling the two flat bands from the simplified model is equivalent to filling all flat bands in the full model.

In addition to its inability to make predictions about electronic behaviors at the moiré of moiré scale, another major limitation of the model is its difficulty to generalize to arbitrary twist angles. For each set of twist angles on a different \((m,n)\) harmonic, it requires the derivation of a new basis by folding the bilayer moiré Brillouin zone, while our model's basis is insensitive to the choice of angles and overcomes this limitation.

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