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Landau level degeneracy in twisted bilayer graphene: Role of symmetry breaking

Ya-Hui Zhang, Hoi Chun Po, and T. Senthil

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA

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The degeneracy of Landau levels flanking charge neutrality in twisted bilayer graphene is known to change from eightfold to fourfold when the twist angle is reduced to values near the magic angle of $\approx 1.05^\circ$. This degeneracy lifting has been reproduced in experiments by multiple groups and is known to occur even in devices which do not harbor the correlated insulators and superconductors. We propose $C_3$-symmetry breaking as an explanation of such robust degeneracy lifting and support our proposal by numerical results on the Landau-level spectrum in near-magic-angle twisted bilayer graphene. Motivated by recent experiments, we further consider the effect of $C_2$-symmetry breaking on the Landau levels.

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I. INTRODUCTION

The discovery [1,2] of correlated insulators and superconductivity in magic-angle twisted bilayer graphene (TBG) has sparked tremendous experimental and theoretical activity. The original results of Refs. [1,2] have been confirmed and extended in Refs. [3,4]. Ferromagnetism, accompanied by an anomalous (possibly quantized) Hall effect, has also been observed at $3/4$ filling of the conduction band for some devices [4,5]. In addition, gate-tunable correlated insulators [6] as well as signs of superconductivity [7] have been demonstrated in the moiré bands of ABC trilayer graphene aligned with a hexagonal boron nitride (h-BN) substrate. Very recently, twisted double bilayers of graphene have also been studied and are shown to host spin-polarized correlated insulators [8–10] and superconductivity [8,9].

In spite of the experimental progress, theoretically there is very little understanding of the many-body physics of these systems. In this paper, we address one aspect of the phenomenology of the normal metallic state of near-magic-angle twisted bilayer graphene. Specifically, we will focus on understanding the Landau-level degeneracy near charge neutrality. As we discuss below, the same pattern of a Landau fan, which conveys information on the nature of the charge carriers and the possible patterns of symmetry breaking, may shed light on the nature of the correlated phenomena closer to the magic angle.

Here, we show that the experimentally observed Landau fan can be reproduced within a free-fermion model of twisted bilayer graphene for various angles close to the magic angle. Our main result is that the sequence is stabilized by a weak breaking of $C_3$ symmetry. As is well known, $C_3$-symmetry breaking splits the Van Hove singularity (VHS) in the density of states [12,13]. For concreteness, let us focus on the conduction band. In the energy range between the two split Van Hove singularities, the equal-energy contours in the momentum space (within each valley) take a qualitatively different shape compared with those allowed in the symmetric case: there is a single electron-like orbit which encloses both of the Dirac points. In contrast, without $C_3$-symmetry breaking, the equal-energy contours either consist of two disjoint pockets each enclosing a Dirac point, or a single hole-like pocket.

1 $C_2$ refers to 180 degree spatial rotation and $T$ to time reversal.

2 For instance, see supplementary data in Ref. [3] on the 1.27° device (D2) at the ambient pressure of 0 GPa.
pocket enclosing the band maximum at \( \Gamma \). In the presence of a magnetic field, these new equal-energy contours give rise to fourfold degenerate Landau levels stemming out from charge neutrality. We find that, even for a relatively small degree of the \( C_3 \) breaking, the degeneracy in the vicinity of the magic angle is fourfold at the magnetic field strengths at which the experiments are done (\( \lesssim 1 \) T).

We trace the origin of this effect to the large susceptibility of near-magic-angle TBG to such a \( C_3 \) breaking perturbation, which leads to a significant modification of the electronic spectrum even when the “bare” \( C_3 \) breaking is weak. Such symmetry breaking may arise due to strain effects (known to be generally present in TBG) or to spontaneous symmetry breaking driven by interactions. While we leave open the physical origin of the \( C_3 \) breaking, we remark that the smallness of the symmetry breaking required suggests that the presence of strain in the sample is a plausible explanation.

In some devices of twisted bilayer graphene; for example, in Ref. [5], it is known that there is very likely also a breaking of \( C_2 \) symmetry. For the device in Ref. [5], this is due to the near alignment with a h-BN substrate. Such \( C_2 \) breaking gaps out the Dirac points and results in an insulator at charge neutrality. In the device studied in Ref. [4], it is observed that the system is insulating at neutrality. A possible explanation is that \( C_2 \) is broken in this system as well, although it is unclear if the symmetry breaking is again due to alignment with h-BN or is interaction driven. In light of these experimental results, we also study the expected Landau-level degeneracy in devices with \( C_2 \)-symmetry breaking, both with and without an additional \( C_3 \) breaking.

We remark that multiple theoretical attempts have already been made to address the neutrality Landau fan in small-angle TBG [14–18]. While all of the emergent symmetries of TBG are preserved in these earlier works, some of them did identify a specific choice of parameters for which the experimental sequence of \( \pm 4 \), \( \pm 8 \), \( \pm 12 \), \ldots can be observed. For instance, Ref. [18] found numerically that the experimental sequence can be reproduced in a narrow range of parameters near the magic angle. However, the sequence is also found to depend very sensitively on the choice of model parameters [18] and might not explain the experimental robustness (across groups and samples) of the Landau-level sequence.

Alternatively, Ref. [15] proposed a VHS-induced Landau-level splitting mechanism that is essentially the same as the one we discuss below. As emphasized above, we find that this picture is valid only if \( C_3 \)-symmetry breaking is invoked. This is consistent with the more microscopic calculation presented in Ref. [16], which did not observe the experimental sequence for the parameter range suggested in Ref. [15] when all the emergent symmetries are kept. Finally, we also note that Ref. [13] suggested that layer-dependent strain could lead to the observed Landau-level sequence near neutrality, although they did not perform an explicit calculation on this point. The mechanism they considered, however, is different from the one discussed in the present work. In particular, in our model the \( C_3 \) symmetry does not differentiate the two layers. Very recently Ref. [19] studied the effects of such strain on the pairing structure of the superconductor in TBG but that work did not address the Landau fans which are our interest here.

\section*{II. Hofstadter Butterfly Calculations}

Our starting point is the single-valley continuum model for twisted bilayer graphene [20,21]. Let \( q = \frac{4 \pi}{5a} \theta \) with \( a = 2.46 \) Å being the lattice constant of monolayer graphene, and define the wave vectors \( \mathbf{q}_1 = -q \hat{y} \), \( \mathbf{q}_2 = q (\sin \frac{2\pi}{5} \hat{x} - \cos \frac{2\pi}{5} \hat{y}) \), and \( \mathbf{q}_3 = g (\sin \frac{4\pi}{5} \hat{x} - \cos \frac{4\pi}{5} \hat{y}) \). The Hamiltonian is given by

\begin{equation}
H = \left( \begin{array}{cc}
\mathbf{H}_M & \sigma_3/2 \\
\mathbf{T}(r) & -\mathbf{H}_M
\end{array} \right),
\end{equation}

where \( \sigma_3 \equiv e^{i \phi \sigma_1/2} e^{-i \phi \sigma_2/2} \), \( T(r) = \sum_{j=1}^3 T_j e^{-i \mathbf{g}_j \cdot \mathbf{r}} \) with

\begin{equation}
T_j = \mathbf{T}_j \left( \begin{array}{cc}
\alpha & e^{-i \mathbf{q}_j \cdot \mathbf{r}} \\
0 & \alpha
\end{array} \right),
\end{equation}

and \( \mathbf{g}_j = \mathbf{q}_j - \mathbf{q}_1 \) are reciprocal-lattice vectors for the moiré potential. We use \( v = 10^6 \) m/s, \( \mu_B = 110 \) meV [21], and \( a = 0.8 \) to incorporate the effect of lattice relaxation [22].

A perpendicular magnetic field can be incorporated by substituting \( -i \mathbf{V} \rightarrow -i \mathbf{V} - e \mathbf{A}/\hbar \), with the vector potential \( \mathbf{A} = -B y \hat{x} \) in the Landau gauge. As in Ref. [14], the spectrum for nonzero \( B \) can be solved by first going into a Landau-level basis for two decoupled layers, and then reexpressing the interlayer potential \( T(r) \) in this basis (more details can be found in Appendix A). In practice, in the numerical calculations we keep only finite Landau levels (typically several hundreds) close to the charge neutrality. Note that, in the limit \( B \rightarrow 0 \), an infinite number of Landau levels should be kept, and so we only consider fields \( B > 1.2 \) T.

Equation (1) has all the effective symmetries of twisted bilayer graphene. In the following, we also consider the effect of symmetry breaking (at the single-particle level). In particular, we set \( (1 - \beta) T^1 = T^2 = T^3 \), such that \( \beta \neq 0 \) controls the degree of \( C_3 \)-symmetry breaking. We also introduce \( C_2 \)-symmetry breaking through \( H_M = M_0 \sigma_3 \otimes M_0 \sigma_3 \), which corresponds to a staggered chemical potential that would be present if the coupling to the hBN substrate were significant.

We focus on small twist angles \( \theta > 1.1^\circ \), slightly away from the true magic angle of \( \theta \approx 1.05^\circ \), for the following reasons: (1) In the experiment, the fourfold degeneracy is observed in a large range of angles, including in devices where the correlated insulators and superconductors are not observed. For instance, the sequence \( \pm 4 \), \( \pm 8 \), \( \pm 12 \), \ldots has been seen in the device at 1.27° in Ref. [3] under ambient pressure. This suggests that the key physics leading to the fourfold degeneracy should be in effect over a relative broad range of angles. (2) Due to the very small bandwidth exactly at the magic angle, the Landau levels are not well separated from each other because of the small cyclotron frequency, which complicates the analysis.

\section*{III. Landau Fan for the Symmetric Case}

First, we show the results for the fully symmetric model, which retains the \( C_3 \) and \( C_2 \) symmetries. We show that the

\footnote{Defined here as the angle at which the active band has a minimum bandwidth, for the parameters we used.}
Landau fan close to charge neutrality should have eightfold degeneracy, in agreement with the experiments at twist angle \( \theta = 1.8^\circ \) but in disagreement with the results near the magic angle \([1,3]\). The spectrum is shown in Fig. 1. From neutrality, only Landau levels \( n = 0, \pm 1, \pm 2 \) are well developed, and the higher levels are superseded by the Landau levels coming from the top and bottom of the active bands. In a semiclassical picture, such blending of Landau levels occurs near the zero-field Van Hove singularity separating the equal-energy contours enclosing the two mini Dirac points from those enclosing the band extrema located at \( \Gamma \).

Importantly, each Landau level emanating from charge neutrality is eightfold degenerate. This degeneracy can be explained by noting the doubling due to spin, valley and layer degrees of freedom. We remark that the layer index is not invariant case, there are several clear differences. First, more Landau levels from neutrality can be seen, suggesting that the Van Hove singularity is pushed further away from charge neutrality. Such a shift of a Van Hove singularity has also been observed in STM experiments at neutrality \([12]\). Second, the previously eightfold degenerate Landau level is split into two groups when magnetic field is larger than a critical field \( B_c(n) \) for each Landau level \( n \). \( B_c(n = 0) = 6 \) T and \( B_c(n = \pm 1) = 1.2 \) T. For \( |n| \geq 2, B_c \) is smaller than the lowest field we can reach in the calculation. Later we argue that \( B_c(n) \sim \frac{1}{|n|} \), which is confirmed for a smaller \( C_3 \) breaking parameter \( \beta = 0.01 \) in the Appendix.

Here we try to give a simple, qualitative explanation of the splitting of the eightfold degenerate Landau level based on a semiclassical picture derived from the band structure at zero magnetic field. With \( C_2 \) symmetry preserved, the fourfold degeneracy from spin and valley is robust. In our numerics, we have explicitly checked that the splitting happens within each fixed spin-valley sector, and so for the discussion here we can focus on a fixed sector. In the fully symmetric case (i.e., for \( \beta = 0 \)), there are two Dirac cones at the \( K \) and \( K' \) points of the mini Brillouin zone (MBZ). At doping slightly above charge neutrality, the Fermi surface consists of independent electron pockets, each enclosing one of the Dirac points \([Fig. 4(a)]\). As discussed in the previous section, the Landau levels arising from such orbits exhibit a twofold mini-valley degeneracy, which, when combined with valley and spin degeneracy, leads to eightfold degenerate Landau levels. When the doping is increased beyond the Van Hove singularity, the Fermi surface becomes a single hole pocket enclosing the \( \Gamma \) point, which leads to fourfold degenerate Landau levels coming from the superlattice gap at the top of the active bands.

Importantly, the breaking of the \( C_3 \) symmetry unpins the Dirac cones from the \( K \) and \( K' \) points and distorts the dispersion. Similar to before, at doping slightly above charge neutrality, the Fermi surface consists of two independent electron pockets, and one expects mini-valley degeneracy in the corresponding Landau levels. However, at higher doping the mentioned distortion opens up the possibility of a new form of Fermi surface, which is electron-like and encloses both of the Dirac points \([Fig. 4(b)]\). The corresponding Landau level

![FIG. 1. Spectrum at twist angle \( \theta = 1.15^\circ \) with both \( C_2 \) and \( C_3 \) symmetry. From neutrality, only \( n = 0, \pm 1, \pm 2 \) Landau levels can be resolved. Each of them is eightfold degenerate and the Landau fan sequence is \( \pm 4, \pm 12, \pm 20 \).](image1)

![FIG. 2. Spectrum at twist angle \( \theta = 1.15^\circ \) with \( C_3 \) breaking parameter \( \beta = 0.07 \). Compared with the \( C_3 \)-symmetric case, more Landau levels emerging from neutrality can be clearly resolved. Meanwhile, in the magnetic field \( B > 1.2 \) T, each Landau level is split into, each being fourfold degenerate. This leads to the Landau fan sequence \( \pm 4, \pm 8, \pm 12, \ldots \). \( E_0 \) is the energy of the first Van Hove singularity point discussed in Sec. IV.](image2)
would appear to emerge from charge neutrality but would no longer showcase mini-valley degeneracy. In other words, the eightfold degenerate Landau levels split. Equivalently, the emergence of such a new form of Fermi surface can be seen from the splitting of the Van Hove singularity as $β$ increases [Fig. 4(c)]. Such splitting from $C_3$-symmetry breaking is also discussed in Ref. [12]. Note that the preceding discussion assumes $β > 0$; while the Van Hove singularity still splits for negative values of $β$, the new equal-energy contours are open orbits and do not lead to fourfold degenerate Landau levels in the semiclassical picture. Consistently, we find that the Landau levels do not split for $β < 0$ (Appendix B). We think the open energy contour is an artifact of the simple model we used here. For a more realistic modeling of $C_3$ breaking, the Landau-level split may be quite stable.

As a complementary point of view, one can consider the Landau levels associated with the mini Dirac cones, which have energies $E_n = v_{\text{eff}} \sqrt{2e\hbar n B}$ for $n \geq 0$. In this picture, the lifting of the mini-valley degeneracy originates from the hybridization between the two sets of mini-Dirac Landau levels. Qualitatively, such hybridization is expected to become significant when $E_n \gtrsim E_C$ [155], where $E_n$ denotes the energy of the first split Van Hove singularity arising from $C_3$ breaking. As such, we expect the Landau-level splitting to be observable for $B \gtrsim B_C(n)$ where $B_C(n) \propto 1/n$. This trend is in qualitative agreement with the numerics shown in Fig. 2 and Appendix C. However, we caution that such a simple picture does not explain, for instance, the splitting of the zeroth Landau level shown in Fig. 2.

V. LANDAU FAN WITH $C_3$ BREAKING

Recent experiments have seen signatures of $C_2$ breaking [4,5]. A typical $C_2$ breaking term is the staggered potential

$$H_M = M_f \psi^\dagger \sigma_z \psi_f + M_b \psi^\dagger \sigma_z \psi_b,$$

where $M_f, M_b$ are the staggered potential strengths for top and bottom layer, respectively. $\sigma$ denotes Pauli matrices in the $A, B$ sublattice subspace.

If the sample is well aligned with only the top h-BN substrate, one expects $M_f \neq 0, M_b \approx 0$ [23,24]. In this case, the zeroth Landau level is always split into four groups, each of which has only twofold spin degeneracy (see Appendix D). Therefore, the first two Landau fan sequences are always $±2, ±4$, which has indeed been observed in both Refs. [5] and [4]. In this case, the activation gap for the $v = 2$ quantum Hall sequence is proportional to the magnitude of $|M_f| - |M_b|$ and therefore is almost a constant when changing magnetic field. This feature may be a useful test for this scenario in the experiment. Note that the valley splitting for the zeroth Landau level is from the valley-sublattice locking and should not be thought as a simple valley Zeeman coupling in the semiclassical picture. To get a strong $v = 2$ sequence in the single-particle level, we find it necessary to invoke a strong mirror-reflection breaking through a finite $|M_f| - |M_b|$. Therefore the sample in Ref. [4] should also have a strong mirror-reflection symmetry breaking to explain the observed $v = ±2$ sequence. The origin of the mirror-reflection symmetry breaking is not clear because no obvious signature of hBN alignment is reported in Ref. [4].

Next, we turn to the higher Landau levels. For $|n| \geq 1$ the valley splitting is quite small, as shown in Fig. 5. Without $C_3$ breaking, the $|n| \geq 1$ Landau levels from mini $K$ and $K'$ overlap with each other and therefore we still expect eightfold
landau level degeneracy in twisted bilayer graphene

VI. DISCUSSION

Since the first experiment on near-magic-angle twisted bilayer graphene, the Landau fan near neutrality has been a mystery. The observed fourfold Landau fan is in striking contrast with the naive theoretical expectation of an eightfold degeneracy which is also seen at larger twist angles. The fourfold degeneracy is seen in devices that approach but need not be too close to the magic angle. In this paper we show how to understand these observations through explicit calculations of the Landau fan within the continuum model of the band structure. While there exist previous such calculations with mixed success in explaining the experiments, we find that a key ingredient is to include C3-symmetry breaking into the continuum Hamiltonian. Upon approaching the magic angle the decreasing bandwidth (and the concomitant increase in density of states) leads to an amplified response of the electronic structure to a weak C3-breaking anisotropy. This makes it easier for the equal-energy contours near the mini-Dirac points to touch and reconstruct. Beyond a low-energy scale the equal-energy contours then enclose both mini-Dirac points, thereby reducing the Landau fan degeneracy from eight to four. We also consider the effect of C2 breaking.

With this understanding let us now revisit the experimental results on various devices. The samples in Ref. [1–3] behave as a semimetal at neutrality, suggesting a good C2T symmetry. To explain the fourfold degeneracy of a Landau fan, we require the existence of C3 breaking, either from strain or from interaction-driven symmetry breaking. Such C3 breaking has been seen in scanning tunneling microscope (STM) experiments [12, 26] on other near magic angle twisted bilayer graphene samples.

The sample in Ref. [5] is aligned with the h-BN substrate on one side. This leads to an explicit breaking of both C2 and the effective mirror (a 180° rotation in three dimensions about an axis that is parallel to and bisects the two layers) symmetries. The C2 breaking leads to a gap at neutrality, consistent with the high resistivity observed at neutrality compared with the devices in Ref. [1–3]. Extensive Landau fan data are not available in Ref. [5], although the reported Landau levels ±2, ±4 are reproduced by our analysis that takes into account both C2 and mirror breaking.

In the recent study of Ref. [4], a number of additional features are reported. In contrast with Refs. [1–3] but somewhat similar to Ref. [5], charge neutrality is insulating with a sizable gap ≈0.8 meV. Furthermore, a Landau fan sequence ±2, ±4, ±8, . . . is reported near neutrality. The insulating behavior at neutrality could be due to C2T breaking or from explicit alignment with at least one of the h-BN substrates, or spontaneously due to interactions.3 In Ref. [4] the observed Landau fan sequence near neutrality is ±2, ±4, ±8, . . . . In our calculations, if we further retain the effective mirror symmetry, then we are unable to reproduce the observation of the ±2 level. Thus we conclude that the mirror symmetry must be strongly broken. An additional C3 breaking is needed to further reduce the degeneracy of higher Landau levels to be fourfold. Thus we think it likely that C2, C3, and effective mirror are broken in the device in Ref. [4].

The C3 and C2 breaking that we have crucially invoked in understanding the Landau fan clearly sets the stage on which the other correlated phenomena happen. An immediate open question is whether it is an explicit or spontaneous symmetry breaking driven by interactions. We hope to study this in the future.

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4 Alternately, it could be due to an interaction-driven intervalley coherence order [27].
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APPENDIX A: CALCULATION OF HOFSTADTER BUTTERFLY

In this section we describe the procedure of our calculation of Hofstadter butterfly spectrum. The method we used is essentially the same as in Ref. [14].

We first describe a general scheme for any graphene moiré superlattice systems. Then calculation for the twisted bilayer graphene is straightforward.

For moiré systems with large lattice constant, the two valleys always form a separated spectrum and here we only treat valley + as an example. At zero magnetic field, a moiré system is described by a continuum model:

\[ H = H_0 + H_M. \]  

(A1)

\( H_0 \) is just the effective model for the valley:

\[ H_0 = \sum_k h_{ab}(k)f^\dagger_a(k)f_b(k), \]  

where \( \alpha, \beta \) is the combination of layer and sublattice index.

\( H_M \) is the moiré hopping or superlattice potential term. It involves the scattering of momentum from \( k \) to \( k + G_M \), where \( G_M \) is the reciprocal vector of the small mini Brillouin zone (MBZ) folded by \( H_M \). A general \( H_M \) term looks like

\[ H_M = \sum_k \sum_Q f^\dagger_{\alpha\beta}(Q)f_{\alpha\beta}(k + Q). \]  

(A3)

When the moiré superlattice is formed by twisting two layers, the momenta of the Dirac cones (or more general band-crossing points) from the two layers are different. Here we follow the notation of Ref. [21] and define the momentum \( \vec{k} \) in \( \psi_{\alpha\beta}(k) \) as the momentum relative to the Dirac point in the corresponding layer fixed by \( \alpha \). In this convention \( \vec{Q}_j \) does not belong to the reciprocal vector of the MBZ.

The key idea to calculate the spectrum under magnetic field is the following: We first ignore \( H_M \) and add a magnetic field to \( H_0 \). We can easily get the new eigenstate in the Landau-level basis. Then we express \( H_M \) in the Landau-level basis, which generically mix different Landau levels. Numerically we can still solve the resulting Hamiltonian and get the spectrum.

First let us solve \( H_0 \) plus magnetic field. The Hamiltonian is labeled by dynamical momenta \( \pi_x = p_x - eA_x \) and \( \pi_y = p_y - eA_y \) with the commutation relation:

\[ [\pi_x, \pi_y] = i\frac{l_B^2}{\hbar}. \]  

(A4)

where \( l_B^2 = \frac{\hbar}{eB} \).

We can define guiding center coordinates \( R_x = x + l_B^2\pi_y \) and \( R_y = y - l_B^2\pi_x \) with the commutation relation

\[ [R_x, R_y] = -il_B^2. \]  

(A5)

We also have \([\pi_x, R_y] = 0\). \( \pi_x \) generates the Landau-level index \( n \) and \( R_y \) generates the orbital within each Landau level.

Define the ladder operators

\[ a = \frac{i\hbar}{\sqrt{2}}(\pi_x + i\pi_y), \]

\[ a^\dagger = \frac{i\hbar}{\sqrt{2}}(\pi_x - i\pi_y). \]  

(A6)

One can easily check that \([a, a^\dagger] = 1\). Therefore, \( a \) can be viewed as an annihilation operator. The Landau-level \( n \) eigenstate satisfies

\[ a|n\rangle = \sqrt{n}|n\rangle. \]  

(A7)

We use the Landau gauge: \( A = (-By, 0) \); then \( R_x = x + p_yl_B^2 \) and \( R_y = -p_xl_B^2 \). We can use \( R_y = -p_xl_B^2 \) to label the basis of each Landau level. Each basis is then labeled by Landau-level index \( n \) and \( k_x \).

We label the function \( \psi_{n,k_x}(y) \) as the wave function in real space for the basis \([n, k_x]\). The corresponding annihilation operator is labeled as \( \psi_{n,k_x,\alpha} \), where \( \alpha = A_1, B_1, A_2, B_2 \) is the sublattice index.

The continuum model can be written in terms of the operator \( f_{\alpha\beta,k}, \) which is related to \( \psi \) through

\[ f_{\alpha\beta,k} = \sum_n \tilde{\psi}_{n,k}(k_x)|\psi_{n,k_x,\alpha}\rangle, \]  

(A8)

where \( \tilde{\psi}_{n,k}(k_x) \) is the Fourier transformation of \( \psi_{n,k_x}(y) \) with respect to \( y \).

We have the following translation property:

\[ \tilde{\psi}_{n,k_x}(y) = \tilde{\psi}_{n,0}(y + k_xl_B^2), \]

\[ \tilde{\psi}_{n,k_x}(k_y) = \tilde{\psi}_{n,0}(k_y)e^{ik_xk_yl_B^2}. \]  

(A9)

\( H_0 \) can be expressed in the following form:

\[ H_0 = \sum_{k_x, n} \sum_{\alpha\beta} \psi^\dagger_{\alpha m,\beta n}(k_x)\psi_{\alpha m,\beta n,k_x}. \]  

(A10)

Here \( k_x \) is defined in the whole range of \( R = [\infty, \infty] \). In the graphene problem, \( h_{nm} \) is usually very simple. For example, in monolayer graphene, it is just an off-diagonal term from \( B, n \) to \( A, \ n + 1 \).

\( H_M \) generically scatters momentum. In terms of \( \psi_{\alpha m,\beta n}, H_M \) in Eq. (A3) can be rewritten as

\[ H_M = \sum_{k_x} \sum_Q \sum_{nm} T_{\alpha\beta}(Q)F_{nm,k_x}(Q)|\psi_{\alpha m,\beta n,k_x} + Q\rangle, \]  

(A11)

where

\[ F_{nm,k_x}(Q) = \sum_{k_y} \tilde{\psi}_{n,k}(k_y)\tilde{\psi}_{m,k_y,Q}(k_y + Q_y) \]

\[ = F_{nm,0}(Q)e^{ik_yQ_y}l_B^{-2}, \]  

(A12)

where we have used Eq. (A9). We have

\[ F_{nm,0}(Q) = \sum_{k_y} \tilde{\psi}^\dagger_{n,0}(k_y)\tilde{\psi}_{m,0}(k_y + Q_y)e^{ik_yQ_y}l_B^{-2}. \]  

(A13)

where \( (z_x, z_y) = \frac{1}{\sqrt{2}}(Q_x, Q_y) \). \( L_m^n(z) \) is the associated Laguerre polynomials.
If we view \( k_x = k_x^0 + x \Delta \) with \( x \in \mathbb{Z} \) as a site in a one-dimensional (1D) chain, Eq. (A11) is basically a hopping term in this 1D chain. The lattice constant is \( \Delta = Q_x \) for the smallest nonzero \( Q_x \). Here \( \alpha, \beta \) just is the orbital label of each site labeled by \( k_x \). At each site \( k_x \), the hopping term \( t(k_x) = F_{\text{meck}}(L) \) is a function of the site \( k_x \). Generically, we have a quasiperiodic 1D model because \( t(x) \sim e^{iQ_x a \Delta x} \sim e^{iQ_x a \Delta x} \) is oscillating according to Eq. (A12).

With the commensurate condition \( \Delta Q_x a = 2\pi \frac{\pi}{p} \), we have a unit cell with \( p \) sites. Then we can solve the model with good quantum numbers \( k_x \) as the crystal momentum of this 1D chain. For TBG, the corresponding model is basically a hopping term in this 1D chain. The lattice constant is \( \Delta = Q_x \) for the smallest nonzero \( Q_x \). Here \( \alpha, \beta \) are just the orbital label of each site labeled by \( k_x \). At each site \( k_x \), the hopping term \( t(k_x) = F_{\text{meck}}(L) \) is a function of the site \( k_x \).

Now we apply the above general scheme to the TBG. We keep \( M \) Landau levels. Typical \( M \) is several hundreds. For the flux corresponding to \( \Phi = \frac{2\pi}{p} \), we can construct a \( 4pM \) matrix for \( H_0 + H_M \) corresponding to each fixed \( (k_1, k_2) \). We use \( f_{\alpha,n,j}(k_1, k_2) \) to label the state with momentum \( k_2 \) in the 1D chain fixed by \( k_1 \). \( j = 1, 2, \ldots, p \) is the sublattice index for each unit cell.

The Hamiltonian corresponding to \( H_0 \) is

\[
H_0 = \frac{-\sqrt{2} \hbar}{l_B} \sum_{k_n=0,1,\ldots,M} \sum_{k_{n+1}} \left[ e^{i\frac{\pi}{2}\sqrt{n+1}} f_{\alpha,n+1,j}^* (k) f_{\beta,n,j} (k) \right] + \text{H.c.} + e^{-i\frac{\pi}{2}\sqrt{n+1}} f_{\alpha,n+1,j} (k) f_{\beta,n,j}^* (k) + \text{H.c.}
\]

\[
+ \sum_k \sum_n M_{\alpha} [f_{\alpha,n,j}^* (k) f_{\alpha,n,j} (k) - f_{\beta,n,j}^* (k) f_{\beta,n,j} (k)]
\]

\[
+ \sum_k \sum_n M_{\beta} [f_{\beta,n,j}^* (k) f_{\beta,n,j} (k) - f_{\beta,n,j}^* (k) f_{\beta,n,j} (k)].
\]

Note that here \( k = (k_1, k_2) \) is just a label of the eigenstate of the 1D chain. It is not the true two-dimensional (2D) momentum.

For TBG, \( H_M \) is

\[
H_M = \sum_k \sum_{nm} \left[ F_{0,nn}^0 (Q) e^{i\frac{\pi}{2} Q l_B^2} e^{i\Delta Q l_B^2 j} + \sum_{j=1,2} e^{iQ_j} F_{nm}^0 (Q) e^{i\frac{\pi}{2} Q l_B^2} e^{i\Delta Q l_B^2 j} \right] \times f_{\alpha,n,j}^* (k) T_{\alpha,n,j}^j (k) f_{\beta,n,j} (k).
\]

where \( T_{\alpha,n,j}^j \) is the interlayer moiré hopping matrix. We have \( T_{\alpha}^j = T_{\beta}^j = T_j \). Here \( t, b \) means top and bottom layer, respectively. The momentum transfer is \( Q_1 = (0, \frac{2\pi}{3a_B}) \), \( Q_2 = (-\frac{2\pi}{3a_B}, \frac{2\pi}{3a_B}) \), and \( Q_3 = (\frac{2\pi}{3a_B}, -\frac{2\pi}{3a_B}) \).

\[
T_j = t^M \left( \frac{\alpha}{e^{-i\pi(j-1)}}, \frac{e^{-i\pi(j-1)}}{\alpha} \right).
\]
FIG. 8. Wannier plot at \( \theta = 1.24^\circ \) for various \( C_3 \)-breaking strength. \( C_2 \) symmetry is preserved. With \( \beta = 0.07 \), we can still see the sequence \( n = \pm 8/\Phi_1 \) at magnetic field \( B > 2.5 \text{T} \).

where \( \varphi = \frac{2\pi}{3} \). \( \alpha \leq 1 \) is a parameter to incorporate lattice relaxation. We use \( \alpha = 0.8 \) in our calculation.

**APPENDIX B: LANDAU LEVELS WHEN \( \beta < 0 \)**

In the main text we show that a \( C_3 \) breaking with \( \beta > 0 \) can reduce the degeneracy to fourfold. Here we discuss the case with \( \beta < 0 \). As is shown in Fig. 6, each Landau level from the charge neutrality is still eightfold degenerate for \( \beta < 0 \). This implies that a very specific form of \( C_3 \) is required to explain the experimental result of a fourfold degenerate Landau fan.

**APPENDIX C: LANDAU FAN AT LARGER TWIST ANGLE**

For \( \theta = 1.24^\circ \), the spectrum and the Wannier plots are shown in Figs. 7 and 8. Clearly, even at this larger angle, \( C_3 \) breaking can lead to a fourfold degenerate Landau fan at finite magnetic field, even though the bandwidth is as large as 40 meV here.

**APPENDIX D: Landau Fan with \( C_2 \) Breaking**

Recent experiments have seen signatures of \( C_2 \) breaking [4,5]. A typical \( C_2 \)-breaking term is just the staggered potential

\[
\mathcal{H}_{C_2} = M_t \psi_t^\dagger \sigma_z \psi_t + M_b \psi_b^\dagger \sigma_z \psi_b,
\]

where \( M_t, M_b \) are the staggered potential strength for the top and bottom layer, respectively. \( \sigma \) is a Pauli matrix in the \( A, B \) sublattice subspace.

For \( C_2 \) breaking coming from alignment with the top (bottom) h-BN layer, we have \( M_t = M, M_b = 0 (M_t = 0, M_b = M) \).

For \( C_2 \) breaking from spontaneous symmetry breaking, a natural choice is \( M_t = M_b = M \) [25]. Alignment with both top and bottom h-BN layers also gives this term.

In the following we discuss the Landau fan close to neutrality from the above two different \( C_2 \)-breaking Ansätze.

1. Zeroth Landau levels

We start from the \( n = 0 \) Landau level with an emphasis on the observed \( \nu = \pm 2 \) sequence [4,5]. When \( C_2 \) is preserved, there are two Dirac cones at \( K \) and \( K' \) of the MBZ for each spin-valley sector.

Let us suppress the spin index. For the Dirac cone at \( K \) (or \( K' \)) of valley \( a = \pm \), we can write a \( 2 \times 2 \) effective model:

\[
\mathcal{H}_{\text{eff}} = \sum_k \psi^\dagger(k)
\begin{pmatrix}
M_{K,K'} & \nu_{\text{eff}}(k_x \mp i k_y) \\
\nu_{\text{eff}}(k_x \pm i k_y) & -M_{K,K'}
\end{pmatrix}
\psi(k),
\]

where \( \psi(k) \) is a two-component spinor. Note that the pseudospin basis of \( \psi(k) \) is a mixture of the sublattice and layer.

FIG. 9. Spectra at twist angle 1.15\(^{\circ}\) for two different \( C_2 \)-breaking Ansätze. \( C_3 \) symmetry is preserved. The two colors indicate states in the two microscopic valleys. The zeroth Landau levels are split to four or two groups and do not disperse with magnetic field up to 8 T. For the second plot with a small \( |M_t| - |M_b| \), the splitting between \( K \) and \( K' \) states within each valley is very small. In this case, the \( \nu = \pm 2 \) quantum Hall sequence should be absent without invoking interaction effects.
When $M_l = M_b$, mirror reflection guarantees $M_K = M_{K'}$. When $|M_l| \gg |M_b|$, we also expect $|M_K| \gg |M_{K'}|$. Let us make $M_l = M_b = 0$ first. Now we add a magnetic field and get four zeroth Landau levels formed by $(\pm, K/K')$ for each spin sector. In total, the zeroth Landau level has eightfold degeneracy. Here we try to give an explanation with $C_3$ breaking parameter $\beta = 0.03$.

For higher Landau levels, Ref. [4] observes fourfold degeneracy. Here we try to give an explanation with $C_2$ breaking Ansätze. With $C_2$ breaking, the only exact degeneracy at finite magnetic field is the twofold spin degeneracy (we ignore the small spin Zeeman coupling). However, for $n \geq 1$, the valley splitting is quite weak, as shown in Fig. 9. For $M_l = 5 \text{ meV}$ and $M_b = 0$, the mirror reflection is strongly broken. However, the $n \geq 1$ Landau levels from $K$ and $K'$ for each valley still overlap with each other. Therefore the $n \geq 1$ Landau levels have eightfold degeneracy as long as $C_3$ is not broken. With $C_3$ breaking, the Landau levels from $K$ and $K'$ will couple with each other and reduce the degeneracy to fourfold, as discussed for $C_2$-symmetric case. We show the Wannier plots for several cases in Fig. 10. One can see that the ansatz with $M_l = M_b$ cannot give the $v = \pm 2$ quantum Hall sequence without invoking interaction effects.

2. Higher Landau Levels

For higher Landau levels, Ref. [4] observes fourfold degeneracy. Here we try to give an explanation with $C_2$ breaking Ansätze. With $C_2$ breaking, the only exact degeneracy at finite magnetic field is the twofold spin degeneracy (we ignore the small spin Zeeman coupling). However, for $n \geq 1$, the valley splitting is quite weak, as shown in Fig. 9. For $M_l = 5 \text{ meV}$ and $M_b = 0$, the mirror reflection is strongly broken. However, the $n \geq 1$ Landau levels from $K$ and $K'$ for each valley still overlap with each other. Therefore the $n \geq 1$ Landau levels have eightfold degeneracy as long as $C_3$ is not broken. With $C_3$ breaking, the Landau levels from $K$ and $K'$ will couple with each other and reduce the degeneracy to fourfold, as discussed for $C_2$-symmetric case. We show the Wannier plots for several cases in Fig. 10. One can see that the ansatz with $M_l = M_b$ cannot give the $v = \pm 2$ quantum Hall sequence without invoking interaction effects.

When $M_l = M_b$, mirror reflection guarantees $M_K = M_{K'}$. When $|M_l| \gg |M_b|$, we also expect $|M_K| \gg |M_{K'}|$. Let us make $M_l = M_b = 0$ first. Now we add a magnetic field and get four zeroth Landau levels formed by $(\pm, K/K')$ for each spin sector. In total, the zeroth Landau level has eightfold degeneracy. Here we try to give an explanation with $C_3$ breaking parameter $\beta = 0.03$.

For higher Landau levels, Ref. [4] observes fourfold degeneracy. Here we try to give an explanation with $C_2$ breaking Ansätze. With $C_2$ breaking, the only exact degeneracy at finite magnetic field is the twofold spin degeneracy (we ignore the small spin Zeeman coupling). However, for $n \geq 1$, the valley splitting is quite weak, as shown in Fig. 9. For $M_l = 5 \text{ meV}$ and $M_b = 0$, the mirror reflection is strongly broken. However, the $n \geq 1$ Landau levels from $K$ and $K'$ for each valley still overlap with each other. Therefore the $n \geq 1$ Landau levels have eightfold degeneracy as long as $C_3$ is not broken. With $C_3$ breaking, the Landau levels from $K$ and $K'$ will couple with each other and reduce the degeneracy to fourfold, as discussed for $C_2$-symmetric case. We show the Wannier plots for several cases in Fig. 10. One can see that the ansatz with $M_l = M_b$ cannot give the $v = \pm 2$ quantum Hall sequence regardless of whether $C_3$ is broken. The only Ansatz which can reproduce the sequence $\pm 2$, $\pm 4$, $\pm 8$, $\pm 12$, ... in Ref. [4] is the one shown in Fig. 5 in the main text.

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