Broken-symmetry states at half-integer band fillings in twisted bilayer graphene

Saisab Bhowmik, Bhaskar Ghawri, Nicolas Leconte, Samudrala Appalakondaiah, Mrityunjay Pandey, Phanibhusan S. Mahapatra, Dongkyu Lee, K. Watanabe, T. Taniguchi, Jeil Jung, Arindam Ghosh and U. Chandni

The dominance of Coulomb interactions over the kinetic energy of electrons in flat moiré bands of magic-angle twisted bilayer graphene (TBG) gives rise to a variety of correlated phases, including correlated insulators, superconductivity, orbital ferromagnetism, Chern insulators and nematicity. Most of these phases occur when the carrier density is at or near an integer number of carriers per moiré unit cell. However, the demonstration of ordered states at fractional moiré band fillings at zero applied magnetic field is more challenging. Here we report the observation of states near half-integer band fillings 0.5 and ±3.5 at zero magnetic field in TBG proximitized by tungsten diselenide. Furthermore, at a band filling near −0.5, a symmetry-broken Chern insulator emerges at high magnetic field that is compatible with the band structure calculations within a translational symmetry-broken supercell with twice the area of the original TBG moiré cell. Our results are consistent with a spin or charge density wave ground state in TBG in the zero-magnetic-field limit.

In two-dimensional (2D) electron systems, a quantizing magnetic field \( B \) is essential for the experimental realization of both integer and fractional quantum Hall states. The Haldane model was the first proposal for a zero-\( B \) analogue of the integer quantum Hall effect, and interacting lattice models with nearly flat Chern bands have since proposed fractionally quantized states in the absence of \( B \) (refs. 13–14). The latter require extremely flat bands featuring non-trivial topological properties, making twisted bilayer graphene (TBG) an ideal material platform to investigate such zero-field states. Recent experiments have revealed a plethora of broken-symmetry phases in TBG, including correlated insulators \( \text{C} \), Chern insulators \( \text{C} \), and related moiré materials \( \text{C} \), where the external tuning of several parameters leads to the desired symmetry breaking. For example, \( \text{C} \) inversion symmetry can be broken by aligning the TBG layers with hexagonal boron nitride (hBN), and the application of a \( B \) field breaks time-reversal symmetry, \( T \). Incompressible states stabilized by electron-electron interactions within the flat bands were found to appear at large \( B \) fields, in the form of Hall plateaux featuring Chern numbers \( \text{C} \) at various band filling indices \( \nu \) (refs. 7–14). Recently, transport measurements in TBG uncovered a high \( B \)-field-induced state at \( \nu = 3.5 \), suggesting the possibility of a new van Hove singularity and fractional Chern insulator. However, states originating from fractional fillings of the unit cell at zero or weak \( B \) fields have remained rather elusive in TBG devices. States featuring fractional \( C \) and/or fractional \( \nu \), which unify the possibility of many exotic states such as spin or charge density wave phases (fractional \( s \)), symmetry-broken Chern insulators (integer \( C \), fractional \( \nu \)) and fractional Chern insulators (fractional \( C \), \( \nu \)) are thus of great interest to our growing understanding of this fascinating material platform. Although all these states emerge from strong electron-electron interactions within the flat bands, subtle changes in experimental configurations can modify the anisotropy energy that decides the preferred ordered phase when the degenerate phases are in close competition.

In this Letter we report the observation of novel states at half-integer moiré band fillings in TBG devices proximitized by a layer of tungsten diselenide (WSe\(_2\); Fig. 1a); these persist at \( B \approx 0 \), suggesting the possibility of a spin or charge density wave order. We focus on magnetotransport and thermoelectric transport measurements on two devices D1 and D2, with twist angles \( \theta \approx 1.14^\circ \) and \( 1.16^\circ \), respectively. In Fig. 1b we show the four-probe longitudinal resistance \( R_{xx} \) as a function of band filling fractions \( \nu \) for \( \nu = 1, 2 \) and 3 and a resistive peak at \( \nu = 4 \). We also observe linear-in-\( T \) behaviour of \( R_{xx}(T) \) at \( \nu = 1, 2 \) and 3 that persists up to \( T = 30 \) K (Supplementary Fig. 8). Device D2 exhibits similar correlated states at different \( \nu \) across various contact configurations (Supplementary Fig. 7). Figure 1c presents the Landau fan diagram for device D1, where the linearly dispersing features emanating from the charge neutrality point \( \nu = 0 \) and other band fillings can be characterized by fitting the Diophantine equation, \( n/\nu = \phi/\phi_0 + s \), where \( n/\nu = \phi/\phi_0 + s \), where \( n/\nu = \phi/\phi_0 + s \), where \( n \) is the density corresponding to one carrier per moiré unit cell, \( \phi \) is the magnetic flux per moiré unit cell, \( \phi_0 = h/e \) is the flux quantum, \( h \) is Planck's constant, \( e \) is the charge of an electron and \( s \) is the band filling index or the number of carriers per unit cell at \( B = 0 \). The slope gives the Chern number via the Streda formula, \( C = (\hbar/e)/(\pi B) \). The Landau levels diverging from \( \nu = 0 \) have filling factors of 4, 8, 12 and 16 (black lines), indicative of the four-fold spin-valley degeneracy. For \( B > 5 \) T, we observe an additional state nucleating from the charge neutrality point \( \nu = 1 \) at \( B = 0 \), with a slope of 2 (green line). Figure 1d shows Hall conductivity \( \sigma_{xy} \) as a function of \( \nu \) at a perpendicular \( B = 1 \) T. We find that, although zero crossings of \( \sigma_{xy} \) are observed, as expected, at \( \nu = 0 \) and 4, a ‘reset’ behaviour is observed at \( \nu = 2, 3 \), namely \( \sigma_{xy} \) decreases to zero and rises again through \( \nu = 2 \) and 3 without any sign change. Such features have been attributed to enhanced Coulomb interactions-driven gap opening when the Fermi energy \( E_F \) crosses...
the van Hove singularity at $\nu = 2$ and 3 (ref. 1). Surprisingly, some novel observations are the additional zero crossings in $\sigma_x(n)$ at $\nu = 1$ and half-integers $\nu = 0.5$ and 3.5. Intriguingly, a minor peak in $R_{xx}(n)$ around $\nu = 3.5$ was seen even at $B = 0$ in several later thermal cycles, as shown in the inset to Fig. 1d. It is often illustrative to plot the Hall density $n_H$ calculated from Hall resistance $R_{xy}$, as $n_H = \frac{e}{2}\frac{\partial R_{xy}}{\partial B}$, which provides insights into the van Hove singularities and Fermi surface topology. In Fig. 1c, $n_H$ diverges with opposite signs on the low- and high-density sides of $\nu = 0.5, 1$ and 3.5. Furthermore, no sign-reversal in $n_H$ is observed at $\nu = 2, 3$. Remarkably, we find robust states at $\nu = 0.5$ and 3.5, even at $B$ fields as low as $\sim 0.3$ T in device D1 (Extended Data Fig. 1).

To further investigate the presence and robustness of states at fractional fillings, we report in Fig. 2 detailed magnetotransport (Extended Data Fig. 1). Although Hall measurements demonstrate the existence of states at half-integer fillings for low $B$, the technique precludes direct observation of these states at $B = 0$. We have performed thermoelectric transport measurements at $B = 0$ for device D1 (details are provided in the Methods). In Fig. 3a, the dependence on $\nu$ of thermovoltage $V_{2\omega}$ shows a series of zero crossings at $\nu = 0, 1, 2, 3$. Most importantly, we observe that the sign-reversal of $V_{2\omega}$ also occurs at $\nu = 3.5$ at $B = 0$. In fermionic systems at very low temperatures ($T \ll T_F$, where $T_F$ is the Fermi temperature), where scattering of charge carriers is diffusive, the semiclassical Mott relation establishes a connection between thermoelectric power ($S = V_{2\omega}/\Delta T$) and electrical resistance ($R_{xx}$)\(^{21,22}\) as

$$S_{\text{Mott}} = \frac{\pi^2 k_B^2 T}{3|e|} \frac{d n_H}{d n} \frac{d E}{E},$$

where $\frac{d n_H}{d n}$ represents the density of states. We compare $V_{2\omega}$ with the derivative of $R_{xx}$ with respect to $n$, shown in the bottom panel of Fig. 3a. We observe exact mapping of zero crossings at $\nu = 1, 2$ and 3,
Fig. 2 | Symmetry-broken states at $\nu \approx -0.5$ and $\pm 3.5$ in device D2, with $\theta \approx 1.16^\circ$. a. Plot of $n_{xy}$ versus $\nu$ at $B = 2$ T and $T = 5$ K. b. $R_{xy}$ as a function of $\nu$ for different $B$ up to 9 T. The colour bar shown here is in a logarithmic scale. Slopes corresponding to the various $R_{xy}$ minima emanating from $\nu = 0, 1$ and $\pm 2$ are as indicated. An additional state develops at $B \geq 6.5$ T, the position of which at $B = 9$ T is marked by the red arrow on the top axis. c. Plot of $R_{xy}$ versus $(\nu, B)$ at $T = 5$ K, showing multiple states at different $\nu$. The red and blue arrows on the x-axis mark $\nu = +3.5$ and $\nu = -3.5$, respectively, where $R_{xy}$ changes sign. In the bottom panel, these states are labelled by $(C, \nu) = (\pm 4, 0), (\pm 2, 0)$ (solid blue), $(1, 3), (+2, +2), (-2, -2)$ (3, 1) (solid green) and $(−4, −0.5)$ (solid red). d. $n_{xy}$ exhibits sign changes at $\nu = -3.5$ (d) and $+3.5$ (e) for high magnetic fields (5–7 T). f. Plot of $R_{xy}$ at $\nu \approx -0.5$ for different $B$, along the red line in the lower panel in c, approaching the quantized limit at $R_{xy} = h/4e^2$ as the field is increased.

with peaks in $R_{xy}$. Although $\frac{dn_{xy}}{d\nu}$ shows a weak peak at $\nu = 3.5$, a clear sign change is observed in $V_{xy}$. Intriguingly, the state at $\nu = 0.5$ does not appear at $B = 0$ in either $R_{xy}$ or $V_{xy}$. This, we believe, is because the sign changes in $V_{xy}$ occur in accordance with the resistive peaks in $R_{xy}$, whereas the sign changes in $\sigma_{xy}$ and $n_{xy}$ are intrinsically connected to the Fermi surface topology via $R_{xy}$. The temperature dependence of $V_{xy}$ in Fig. 3b shows the sign reversals as seen by a series of vertical red and blue bands throughout the measured $T$ range. Although the transitions at $\nu = 0, 1, 2$ are robust and persist up to the measured temperature of $T = 40$ K, the feature at $\nu = 3.5$ vanishes for $T \geq 18$ K.

The emergence of distinct features in magnetotransport and thermoelectricity at half-integer band fillings for two TBG-WSe$_2$ devices, despite the inevitable variability typically seen in TBG samples, suggests a robust mechanism behind our observations. We speculate that strong correlations between electrons may generate a spin or charge density order that breaks translation symmetry in the moiré lattice, leading to Brillouin zone folding with electron wavefunctions extending over a unit cell with doubled area. In the non-interacting, single-particle band structure of TBG, a van Hove singularity is expected to appear at $|\nu| = 2$ (refs. 11,22). Broken translational symmetry and the appearance of a folded Brillouin zone may favour van Hove singularities at $\nu = 1$ and other partial fillings instead, as observed in our devices. Although higher-order translational symmetry breaking (tripling, quadrupling) is a possibility, we cannot rule out the presence of an inherent, accidental van Hove singularity at $\nu = 0.5$ either. As described in the theoretical model below, additional spin splitting of bands can lead to a complex density of states hierarchy, which can explain the features at half-integer fillings. Furthermore, the emergence of a Chern insulator at $\nu \approx -0.5$ in device D2 strengthens the possibility of a doubled moiré unit cell. We also remark that the observation of a state with a slope of $C = 2$, as obtained from the Streda formula from a band filling of $s = 1$ for device D1, is reminiscent of an odd-parity ($p = C + s$) Chern insulator, attributed to reconstruction of Chern bands driven by the modulation of charge density waves over the real-space moiré unit cell’6.

Figure 4a shows the band structure of TBG subject to a 1D density wave potential that doubles the moiré unit cell area (Fig. 4b) and hence the number of bands, similar to ref. 11. The four-fold degenerate conduction and valence bands will turn into two eight-fold degenerate bands upon zone-folding when the supercell area is doubled. In Fig. 4c, we schematically illustrate the degeneracy lifting of these bands, resulting in a total of 16 split bands in the presence of spin or charge density potential patterns. The states at half-integer fillings observed in experiments are therefore compatible with an odd-integer filling of the zone-folded split bands. Our proposal of 1D patterned phases whose periods are commensurate with the moiré patterns can give rise to fractionally filled flat bands, analogous to the Bloch band filling index $s = \pm 1/3$, 1/2 states seen in Landau levels$^{14}$ that can be justified in the presence of Wigner crystal phases$^{26}$. Our tight-binding Hamiltonian in the carbon π-orbitals basis incorporates this 1D pattern through a sinuousoidal function and further incorporates the sublattice staggering as well as the intrinsic and Rashba spin–orbit coupling potentials that are often introduced when describing proximity spin–orbit coupling of graphene on WSe$_2$ (see Methods for details). These terms introduce an energy anisotropy that favours a certain spin and valley isospin polarization
The system parameters for the band structure are ad hoc choices that illustrate the degeneracy lifting of the folded flat bands, which the Coulomb interactions can further amplify. We note that proximity-induced spin–orbit coupling in TBG-WSe₂ heterostructures is experimentally well established. Supplementary Fig. 2 describes the bands associated with the alternative \( \phi = \pi/2 \).
cosine-like 1D potential that illustrates the overall robustness to shifts in its origin.

Our work shows that the generation of flat bands by means of moiré superlattices is enriched by the possibility of superposing periodic potentials that are commensurate with the original moiré pattern\textsuperscript{15}. The favoured ground state among several quasi-degenerate phases will depend on the anisotropy energy for different spin/valley isospin polarization that can change with details in dielectric environment, sample preparation and device assembly methods. The details of spin and valley isospin ordering will impact system properties like the spin or orbital magnetism stemming from the topological nature of the bands, the superconducting pairing symmetry, as well as the instabilities in the electron–electron and electron–hole channels. We have only considered the $s = 1/2$ Bloch band filling index scenario, and future research attention should be given to $s = \pm 1/3$ for the Kekulé patterns with three times larger superlattice area, as seen in experiments with graphene/hBN\textsuperscript{24,25}. Our work suggests that using transition metal dichalcogenide (TMD) dielectrics is a promising pathway to further understand and explore the nature of fractional-filling-correlated states in the limit of vanishing magnetic fields.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01557-4.

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Methods

Device fabrication. The devices in this work were fabricated using a modified ‘ear and stack’ method (Supplementary Fig. 6). The different layers of the heterostructures were sequentially picked up using polypropylene carbonate (PPC) films coated on polydimethylsiloxane (PDMS) stamps. The Hall bar devices were fabricated using electron-beam lithography, reactive ion etching using CHF$_3$/O$_2$ and thermal evaporation of ohmic edge contacts using Cr/Au (5 nm/60 nm). WS$_2$ layers of thickness 2–3 nm were exfoliated from bulk crystals procured from 2D semiconductors.

Transport measurements. Electrical and thermoelectric transport measurements were carried out in a pumped helium cryostat with a 9-T magnetic field. Magnetotransport measurements were performed with a bias current of 10–100 nA using an SR830 low-frequency lock-in amplifier at 17.81 Hz. For the thermoelectric measurements, a temperature gradient across the entire TBG-WS$_2$ region was created by passing a sinusoidal current (I$_{AC}$) through single-layer graphene contacts located outside the TBG-WS$_2$ region (Fig. 3a inset). A range of sinusoidal currents (300–400 nA) were used at an excitation frequency of $\omega = 17.81$ Hz, and the resulting second-harmonic thermovoltage ($V_{2\omega}$) was recorded using the same lock-in amplifier. The linear variation of $V_{2\omega}$, with $I_{AC}$ ensured a linear response regime for the range of heating currents used. To estimate the twist angle, we used the relation $n_t = 80^2/3\pi^2$, where $a = 0.246$ nm is the lattice constant of graphene and $n_t$ $(a = 4)$ is the charge carrier density corresponding to a fully filled superlattice unit cell.

Real-space lattice calculations. The tight-binding Hamiltonian incorporating the 1D density wave potential can be written as

$$H = \sum_{l,m} \sum_{\sigma} c^\dagger_{l,m,\sigma} c_{l,m,\sigma} + \sum_{l} \left( C_0 + C_1 \right) \sin \left( \frac{2\pi}{n_t} \right) c^\dagger_{l,\uparrow} c_{l,\downarrow}$$

$$+ \sum_{l,m} \Delta c^\dagger_{l,m,\sigma} c_{l,m,\sigma} + \sum_{(l,m)} \nu_{l,m} t e^{i2\pi l \Delta} c^\dagger_{l,m,\sigma} c_{l,m,\sigma}$$

$$+ \frac{\lambda}{2} \sum_{l,m,\sigma,\sigma'} c^\dagger_{l,m,\sigma} c_{l,m,\sigma'} \left[ \hat{z}_{\sigma} \times \hat{d}_{l,m,\sigma} \right]_{\sigma,\sigma'}.$$  

Here, the first term describes the carbon interatomic hopping terms, which are tuned to generate the magic-angle flat bands at 1.08° (ref. 31). Supplementary Section IA describes the expression for the distance-dependent $t_{\alpha\tau}$ terms. The $l$, $m$ indices are lattice labels, $m_r$, where the $Y$ subscript refers to the top-layer sites that are contacting the TMD. The second 1D sine term describes a density wave potential repeating with a period of $2\Delta$ (twice the TBG moiré period), which defines a supercell that doubles the moiré area and allows folded bands to be obtained that split at half-integers of the unperturbed systems filling factors. The $\phi$ phase term is a control knob that shifts the potential origin position. The remaining three terms capture the WS$_2$ interface effect in the contacting graphene layer through a constant mass term with unequal sublattice resolved potentials, as well as through Kane–Mele and Rashba spin–orbit coupling models, where $\sigma_{m,\tau} = \pm 1$, following the conventions in ref. 14, where $\hat{z}$ represents the spin Pauli matrices and $\hat{d}_{l,m,\sigma}$ is the displacement vector between lattice sites $l$ and $m$, $\sigma_{m,\tau} = \pm 1$ indicates the presence of mass with opposite signs for the $A$, $B$ sublattice resolved potentials. Our Hamiltonian in equation (2), based on the Kane–Mele model 32, has a simpler form compared to the graphene/TMD models with unequal sublattice resolved spin–orbit coupling that leads to valley Zeeman terms 26, but is able to completely lift the degeneracy of the moiré bands.

The atomic positions were relaxed using LAMMPS molecular dynamics simulations 33 based on EXX-RPA-informed 8 force fields, and the electronic structure was obtained through real-space tight-binding calculations as outlined in Supplementary Section I. Both the band structure and density of states were calculated by a shift–invert diagonalization method 18 with a broadening of 0.8 meV, and the band structures in the absence of spin–orbit coupling terms were calculated using exact diagonalization. We used a $4\times3\times1$ Monkhorst–Pack $k$-point grid for the density of states calculation in a rectangular supercell containing 22,328 atoms. The graphene itself was modelled using the FG2 model 34. More details about the Hamiltonian implementation, analysis of the different terms in the Hamiltonian and phase-shifted patterns are provided in Supplementary Section I.

Data availability

Data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Code availability

The codes that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

S.B. fabricated the devices, performed the measurements and analysed the data. B.G., M.P. and P.S.M. assisted with measurements and analysis. K.W. and T.T. grew the hBN crystals. A.G. and U.C. advised on experiments. N.L., S.A., D.L. and J.J. performed the theoretical calculations. S.B., N.L., J.J. and U.C. wrote the manuscript, with input from other authors.

Competing interests

The authors declare no competing interests.

Additional information

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Correspondence and requests for materials should be addressed to Saibab Bhowmik, Jeil Jung or U. Chandni.

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Extended Data Fig. 1 | Low-field Hall data in device D1, $\theta \approx 1.14^\circ$. a. $n_h - \nu$ plot for $B = 0.3 - 1.2$ T. b. $\sigma_{xy}$ as a function of $\nu$ for the same range of $B$. The zero crossings at $\nu = 0.5, 1$ and 3.5 and reset at $\nu = 2$ and 3 are robust with $B$. 
Extended Data Fig. 2 | Low-field Hall data in device D2, $\theta \approx 1.16^\circ$. a. $n_\text{e}$ vs. $\nu$ plot for $B = 0.2 - 2$ T. The sign change at $\nu = 3.5$ appears at the lowest $B$-field of 0.2 T. In addition, the reset at charge carriers is observed at $\nu = \pm 2$. b. $\sigma_{xy}$ as a function of $\nu$ for the same range of $B$. 
Extended Data Fig. 3 | Hall conductivity at $B = 9$ T in device D2. \( \sigma_{xy} \) shows plateaus as \( \sigma_{xy} = C e^2/h \) associated with the minima in \( \sigma_{xx} \). Different color bars (blue for the CNP, green for \( \nu = 1, \pm 2, 3 \) and red for \( \nu = -0.5 \)) have been used to show the sequence of symmetry-broken nearly quantized states nucleating from several partial fillings of flat bands.
Extended Data Fig. 4 | Magneto-thermoelectricity measurements in device D1. Thermoelectric voltage $V_{\text{te}}$ measured at $B = 0, 1, 1.5,$ and $2 \, \text{T}$ and $T = 3 \, \text{K}$ for a heating current of 300 nA. The feature at $\nu = 3.5$ is clearly visible at all $B$. A tiny shoulder develops in the vicinity of $\nu = 0.5$, that is particularly prominent at $B = 2 \, \text{T}$.