Dirac spectroscopy of strongly correlated phases in twisted trilayer graphene

Received: 21 April 2022
Accepted: 2 November 2022
Published online: 22 December 2022

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Magic-angle twisted trilayer graphene (MATTG) hosts flat electronic bands, and exhibits correlated quantum phases with electrical tunability. In this work, we demonstrate a spectroscopy technique that allows for dissociation of intertwined bands and quantification of the energy gaps and Chern numbers $C$ of the correlated states in MATTG by driving band crossings between Dirac cone Landau levels and energy gaps in the flat bands. We uncover hard correlated gaps with $C = 0$ at integer moiré unit cell fillings of $\nu = 2$ and $3$ and reveal charge density wave states originating from van Hove singularities at fractional fillings $\nu = 5/3$ and $11/3$. In addition, we demonstrate displacement-field-driven first-order phase transitions at charge neutrality and $\nu = 2$, which are consistent with a theoretical strong-coupling analysis, implying $C_2T$ symmetry breaking. Overall, these properties establish a diverse electrically tunable phase diagram of MATTG and provide an avenue for investigating other related systems hosting both steep and flat bands.

Fundamental signatures of quantum phases often emerge as gap openings, which signify phase transitions in band topology, electronic correlations and elementary excitations. In flat band moiré systems, which have recently emerged as a robust and tunable platform to investigate strongly correlated electrons, exotic gapped quantum states of correlated insulators, superconductivity and topological Chern insulators have been uncovered. In particular, the recently realized mirror-symmetric twisted trilayer graphene system with a magic angle $\theta \approx 1.5^\circ$ (MATTG) has evolved as an interesting new platform (Fig. 1a). Its energy spectrum, however, contains a coexisting moiré flat band and a steep Dirac cone (Fig. 1b), which makes the detection of correlation-induced insulating states very difficult because multiband transport through the Dirac cone obscures these. Explicitly detecting and quantifying the non-trivial gapped states in the flat bands of MATTG requires a new technique that can selectively untie the electrons in the intertwined bands.

In this work, we overcome these challenges by introducing a Dirac spectroscopy approach, which makes it possible to quantify the Chern numbers and gap sizes of the interaction-driven gaps in the flat bands. This is accomplished by studying in detail the band crossings of these gaps with the Landau levels of the Dirac cone (D-LLs) under a small perpendicular magnetic field $B$ (Fig. 1c). As shown in Fig. 1a, the device in this study consists of a double-gated, ultra-clean MATTG device with a twist angle of $\theta \approx 1.5^\circ$ (Methods), where the two gates make it possible to induce a displacement field and also change the electron filling per moiré unit cell $\nu$.

Figure 1d,e shows the Landau fans of longitudinal magnetoresistance $R_{xx}$ and Hall resistance $R_{xy}$ versus $B$ and $\nu$ at zero $D$, and Fig. 1f shows the corresponding $R_{xy}$ versus $\nu$ line traces for selected fixed $B$. To resolve the crossing features between the correlated flat band gaps and the D-LLs, we focus only on a narrow $(\nu, B)$ phase space from $0.5 < \nu < 3.5$ and $0 < B < 3$ T. The key crossing features in this range, however, are seen for all integer $\nu$ throughout the entire moiré valence and conduction bands (Extended Data Fig. 1). Here, we observe a multitude of gapped features at integer fillings $\nu$ and Landau levels originating from these, which follow linear trajectories in the $(\nu, B)$ phase space.
and 

and perpendicular magnetic field. Black solid lines mark the band edges A and 

crossings between the D-LLs with their dispersion relation 

as is schematically demonstrated in Fig. 1c. This shows the band 

features are associated with the flat bands, the latter are attributed to 

magnetic fields that emerge from charge neutrality, which appear 

$\nu$ as non-monotonic features in the ($\nu$, $B$) phase space. While the first D-LL and the second D-LL cross with band edges A and B, respectively. Illustrative schematics (right) of both gapped flat band and D-LL (phase I), where the total Chern number is $C = C^1 + (4N + 2)$ and $N$ is an integer. Circled numbers (1) and (2) link the positions in the schematics with the positions in the measurements in (d), (e) and (f). d.e, Landau fan diagram shown by longitudinal resistance $R_{xx}$ (d) and transverse Hall resistance $R_{xy}$ (e) at zero displacement field. e, linecuts of $R_{xy}$ at different magnetic fields B, as taken from (e) and marked there by colored bars. Grey and blue shading denote correlated states for a flat band charge with a correlated insulator (CI) and a charge density wave (CDW) at integer fillings $\nu = 1, 2, 3$ and fractional filling $\nu = 5/3$, respectively.

We also observe an extra set of quantum oscillations at much lower magnetic fields that emerge from charge neutrality, which appear as non-monotonic features in the ($\nu$, $B$) phase space. While the first features are associated with the flat bands, the latter are attributed to the Dirac cone.

Overall, the features around integer fillings $\nu = i$ in Fig. 1d,e can be well understood within a complex crossing picture between the LLs formed in the Dirac band and the correlated gaps in the flat band, as is schematically demonstrated in Fig. 1c. This shows the band crossings between the D-LLs with their dispersion relation $\mu^{D\text{-LL}}_n = \nu_{B} \sqrt{2 e \hbar N \text{sign}(N) B}$, and the interaction-induced gaps of the flat band (see also Extended Data Fig. 2). Here, $\nu_{B}$ is the elementary charge, $\hbar$ the reduced Planck constant, $N = 0, \pm 1, \pm 2, \ldots$ is the Landau level index, $\mu^{D\text{-LL}}_n$ and $\mu^{LL \text{-LL}}_{n+1}$ are the chemical potential of the $n$th D-LL and the $n$th D-LL (at the Dirac point), respectively, and $\nu_{B}$ is the Fermi velocity of the Dirac cone. When both the flat band and the D-LL are gapped and the chemical potential of the flat band is equal to $\mu^{f} = \mu^{D\text{-LL}}_n$, then the total Chern number of the combined gap is $C = C^f + C^D$, where $C^f$ is the Chern number of the flat band gap and $C^D = 4N + 2 = 2, 6, 10, \ldots$ is the Chern number of the D-LL gaps. As we will show, these considerations will allow us not only to directly extract the Chern numbers of the correlation-induced gaps $C^f$, but also to extract their gap size.

**Zero Chern numbers at integer fillings**

The main clue to understanding these characteristics comes from Fig. 1f, which shows a well-quantized Hall resistance $R_{xy}$ at integer fillings $\nu = h/(Ce^2)$ and $R_{xx}$ 0 inside the correlated gaps at integer fillings $\nu$, which follow linear trajectories $dn/d\nu = C_e/h$ in the $\nu$--$B$ phase diagram as expected from the Streda formula. The quantized plateaus follow a robust sequence where $\nu = 0, \pm 1, \pm 2, \ldots$ is the Landau level index, $\mu^{D\text{-LL}}_n$ and $\mu^{LL \text{-LL}}_{n+1}$ are the chemical potential of the $n$th D-LL and the $n$th D-LL (at the Dirac point), respectively, and $\nu_{B}$ is the Fermi velocity of the Dirac cone. When both the flat band and the D-LL are
as is schematically explained in Fig. 1c. Since the sequence of Chern numbers inside the correlated gap is precisely what is expected from Dirac electrons, the transport must be carried through the edge states formed between the D-LL, while the electrons in the flat bands must be fully localized. Hence we can conclude that the interaction-driven states at integer fillings \( \nu = 3 \) are correlated insulators with Chern numbers \( C = 0 \). While we cannot resolve the correlated states for other fillings in such great detail, the slopes in the Landau fan suggest that all these also have Chern numbers \( C = 0 \) (Extended Data Fig. 1 and Supplementary Information, section A).

The observed \( C = 0 \) at \( \nu = 2, 3 \) can be traced down to the zero field limit \( B < 0.3 \) T, and can survive up to a large perpendicular magnetic field \( B > 8 \) T without transitioning to a Chern insulator state with a non-zero Chern number \( |C| = 4 - |\nu| \), as is observed in magic-angle twisted bilayer graphene due to time-reversal symmetry breaking of its valley-projected Chern bands (Extended Data Fig. 1)\(^{27-30}\). For the \( C = 0 \) ground state, one candidate mechanism is translation symmetry breaking, which doubles the number of bands and leads to a natural description with eight \( C = 0 \) bands and eight \( C = \pm 1 \) bands; there are then natural states at \( \nu = 3 \) with zero total \( C \) that are obtained by filling combinations of these 16 bands\(^{30}\). Alternatively, \( C = 0 \) states at both non-zero fillings \( \nu = 2, 3 \) are also consistent with the recently proposed time-reversal symmetric 'incommensurate Kekulé spiral' order which appears in the presence of strain\(^{28}\). Further studies are needed to uncover the competitions of these Chern insulator states in MATTG which are determined by symmetry breaking, a process that is sensitive to specific parameters such as strain and magnetic field.

### Charge density waves at fractional fillings

Beyond the integer filling states, we also find similar features at fractional fillings \( \nu = 5/3 \) and \( 11/3 \). As can be seen in Figs. 1d,e and 2c, at these fractional fillings, we observe similar features as for the integer fillings, which signifies the formation of interaction-driven \( C = 0 \) gaps in the flat band. Our Hartree–Fock simulations of the hole-doped side of \( \nu = 2 \) (Fig. 2a and Supplementary Information, section B), show the existence of van Hove singularities (VHSS) with six hole pockets which merge into a single ring-shaped Fermi surface. In particular, \( \nu = 5/3 \) is extremely close to these VHSS in energy and its Fermi surface is nearly nested with respect to the wavevectors that connect moiré K and K' points. These wavevectors are the translation-symmetry-breaking wavevectors that correspond to a \( \sqrt{3} \times \sqrt{3} \) tripling of the moiré unit cell in a \( C_6 \) symmetric way. Breaking \( C_6 \) symmetry is believed to destroy the CDW state by ruining the above nesting. This accounts for the rare observation of CDW in other devices as \( C_6 \) symmetry is often broken by strain effects. We also study the temperature dependence of these CDW states as shown in Fig. 2b. The melting of the correlated states at increased temperature, manifested by the vanishing of Hall resistance, shows a faster melting of the \( \nu = 5/3 \) state than for the \( \nu = 2 \) state, in accordance with the expected smaller inter-site Coulomb interaction responsible for fractional filling states as compared to stronger on-site Coulomb interaction for integer fillings.

### Extracting the gap size of the correlated gaps

For brevity, and without loss of generality, we focus on the \( \nu = 2 \) state for the rest of this paper, since all integer filling states show similar behaviour. We estimate the size of the interaction-induced gap of the \( \nu = 2 \) state in Fig. 3a. Such spectroscopy can be achieved by measuring the chemical potential jump across these gaps, which can be achieved by measuring the crossing points of its band edges with two consecutive D-LLs in \( \nu = 2 \), as is demonstrated in Figs. 1c and 3b. By tracking the Oth D-LL (\( N = 0 \)) in the \( \nu = 2 \) phase diagram, the chemical potential of the flat band \( \mu' \) relative to the Dirac point can be extracted as \( \mu' - \mu'_{\text{Dirac}} \) (ref. 20). This way the chemical potential jump of \( \mu' \) at the correlated gap of the flat band can directly manifest itself by the change of magnetic field at the crossing of the D-LLs in the \( \nu = 2 \) phase space. We obtain the exact band edge points \( A_N \) and \( B_N \) for each LL index \( N \) (see also Fig. 1c) by tracking the crossing state that corresponds to partial filling of D-LLs at the Fermi level, which is marked by an \( R_{xy} \) peak. When this state occurs between the D-LLs and the correlated gap (phase I in Extended Data Fig. 2), it features a transition between two adjacent \( R_{xy} \) plateaus with \( C = 4N \pm 2 \) by changing the magnetic field \( B \). When this phase reaches the crossing between the D-LL and the flat band edges, the \( R_{xy} \) plateau transition disappears and the \( R_{xy} \) peak merges into the two flanking peaks that manifest the edges of the flat band. The thus extracted magnetic fields at the band edge points \( A_N \) and \( B_N \) determine the corresponding chemical potentials in these points \( \mu'_{AN} \) and \( \mu'_{BN} \) relative to zeroth D-LL, and allow us to estimate the gap size of the correlated states via \( \Delta' = \mu'_{BN} - \mu'_{AN} \), where a value of \( \nu < 10^7 \text{ m}^2 \text{s}^{-1} \) is used according to theoretical and recent experimental results\(^{29-31}\). We find a value of \( \Delta'(\nu = 2) = 4.8 \) meV at \( D = 0 \) for the correlated gap of the \( \nu = 2 \) state, which has a similar value as the half-filled gaps previously extracted for magic angle twisted bilayer graphene.
and perpendicular magnetic field, where DOS is the density of states. c. Extracted chemical potential of the band edges relative to the 0th D-LL and correlated gap Δν (ν = 2) as a function of the displacement field for the flat band. The extraction is carried out with D-LL index N = 1. Error bars represent the uncertainty in the values of the chemical potential and the gap size calculated from the magnetic field of crossing points Aν and Bν, which show an uncertainty of ∼0.01 T in a.

**Fig. 3** | Extraction of the interaction-driven gap at ν = 2. a. Quantum oscillations at low magnetic field shown by longitudinal resistance Rxy and transverse Hall resistance Rxy, at a variety of displacement fields. Blue dashed lines mark Rxy peaks and Rxy transitions of C = 4ν + 2 in a magnetic field B, which corresponds to a phase II where half-filled D-LLs cross the correlated gap of the flat band. b. Illustrative schematics of gap measurement by tuning carrier density and perpendicular magnetic field, where DOS is the density of states. c. Extracted chemical potential of the band edges relative to the 0th D-LL and correlated gap Δν (ν = 2) as a function of the displacement field for the flat band. The extraction is carried out with D-LL index N = 1. Error bars represent the uncertainty in the values of the chemical potential and the gap size calculated from the magnetic field of crossing points Aν and Bν, which show an uncertainty of ∼0.01 T in a.

**D-driven gap enhancement and Dirac cone shifting**

The application of a displacement field D in MATTG renormalizes the single particle band structure, where the Dirac cone and the flat bands hybridize, as has been shown in Fig. 1b. To study the effect of D on the ν = 2 state (see Supplementary Information, section A for the discussion on other integer filling states) we repeat this spectroscopy for different |D| < 0.2 V nm⁻¹, where we assume that νν is not significantly altered (Supplementary Information, section C), and find that Δ⁺ (ν = 2) shows a sizeable enhancement with respect to increased D, as can be seen in Fig. 3c (see also Extended Data Fig. 3). Strong-coupling analytics and Hartree–Fock simulations can explain this trend, where for small D and not-too-large twist angles, the dispersion of the twisted bilayer graphene (TBG) subsystem bands is increased. This increased dispersion can combine with the interaction-induced gap such that the total gap is increased (Supplementary Information, section B). In previous reports on MATTG, also in our device, the nearby superconductor at ν = 2 + δ also shows a superconducting transition temperature Tc enhancement with moderate D (Supplementary Information, section D). This observed increase in both Δ⁺ (ν = 2) and Tc hints at a relation between correlated insulator and the nearby superconductor, and is compatible with a scenario where the pairing scale is controlled by the TBG subsystem dispersion³⁴.

As is further plotted in Fig. 3c, we also find that at the band edge points Aν and Bν, the chemical potentials relative to the Dirac point, µν Aν = µν Aν LL and µν Bν = µν Bν LL, are decreased by increased D. We interpret this behaviour to be due to an upward shift in the energy of the Dirac point with D, as can be seen in the single particle band-structure calculations for a finite D in Fig. 1b. While single particle simulations predict that the Dirac cone immediately hybridizes with the flat bands for arbitrarily small D, due to the breaking of the mirror symmetry, our band crossing results rather suggest a hybridization that is gradual and proportional to D. This can be seen in the quadratic shifting of the hybridized Dirac points with D, which is in contrast to a linear shifting predicted by the single particle picture. This failure of the single particle picture is explained with our strong-coupling and Hartree–Fock predictions (SI section B).

**Displacement-field-driven phase transitions for large D**

Due to the strong hybridization of the Dirac cone with the flat band at elevated D, the Dirac spectroscopy cannot be further applied. To further illuminate the effect of large displacement field D on the many-body states in MATTG we can however use transport measurements. Figure 4a,b shows the phase diagrams of Rxy versus v and D at B = 1 T and longitudinal resistivity ρ versus v and D at B = 0 T, respectively, which show a multitude of correlated states at various fillings and two robust superconducting regions in the interval of |ν|v = 2 + δ (0 < δ < 1), in good agreement with previous work²²,²³. Most importantly, both plots show a multitude of displacement-field-driven transitions of Rxy and ρ versus D for the different integer filling states. Focusing again on the ν = 2 state and taking the line cut of ρ versus D, as is plotted in Fig. 4c, we can identify several trends. Starting from D = 0 we observe a low-resistivity metallic state, which gradually increases with D and develops a localized non-gapped peak around a critical displacement field value of |Dν | = 0.4 V nm⁻¹. Above another critical displacement field of |Dν | = 0.6 V nm⁻¹ the state becomes gapped, showing clear temperature-activated behaviour (inset of Fig. 4c) with an extracted gap of Δν = 0.26 meV, and above |Dν | > 1.1 V nm⁻¹ the state transitions again into a low-resistivity metallic state. We further mark these critical displacement fields in Fig. 4a,b, which clearly show a consistency of these transitions both in the ρ and the Rxy data.

To understand this behaviour, we propose an evolution of states as schematically depicted in Fig. 4d. Here the transition |Dν | marks the transition of a non-hybridized Dirac cone to a strongly hybridized one, where the resistance peak arises due to the Fermi surface coinciding with the Dirac point as the Dirac cone is gradually shifted upwards with D (see more detailed discussion in Supplementary Information, section K). The transitions to a gapped state at |Dν | and again to a non-gapped metallic state at |Dν |, we interpret in the framework of first-order transitions at the Dirac cone point between states that are driven by a tunable competition between energetically competitive symmetry-breaking orders, in particular the Kramer intervalley coherence (KIVC), valley
Hall (VH), valley polarization (VP) and semimetal (SM) states. Figure 4e plots the energies of these orders tuned by displacement field from numeric Hartree–Fock simulations, where the KIVC state is gapless and dominates at moderate displacement field and VH, which breaks $C_{3v}T$, is fully gapped for $D \geq 0$ and is lower in energy for larger displacement fields, before ultimately a symmetric semimetal becomes the ground state at the largest fields (Supplementary Information, section B). The displacement field regime of the gapped $\nu = 2$ state is consistent with the theoretical prediction, where notably the superconductivity is weakened in the regime of the displacement field where VH is developed (Fig. 4b), consistent with the absence of superconductivity in samples with explicit $C_{3v}T$ breaking with, say, an aligned hexagonal boron nitride (hBN) substrate. This is also compatible with theoretical proposals based on skyrmion pairing or pseudospin fluctuations. Such a phase transition of a Dirac cone from non-gapped KIVC to gapped VH is also emergent at charge neutrality (Fig. 4b and see more discussions on $D$-driven effects at other integer fillings in Supplementary Information, section K).

In summary, we put forward a Dirac spectroscopy in MATTG that is employed to reveal and quantify many-body electron states, in particular of correlated states with zero Chern numbers at integer or fractional fillings. Such spectroscopy also casts light on the evolution of exotic phase transitions and complex correlated band structures in displacement fields. Our work also helps to bridge between correlated insulators and nearby unconventional superconductors, and provides an avenue to study correlated states in other twisted multilayer graphene with both steep and flat bands.

**Online content**

Any methods, additional references, Nature Portfolio 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/s41563-022-01428-6.

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Methods

Device fabrication

The whole stacking is fabricated by the van der Waals assembly technique. Graphite, hBN and monolayer graphene flakes are mechanically exfoliated on SiO$_2$ (285 nm)/Si$^+$ substrates and the optimal thickness chosen by optical contrast. A graphite layer a few nanometres thick, which acts as the top gate, is first picked up at 100 °C by a propylene carbonate film which is placed on a polydimethyl siloxane stamp. All the other layers are subsequently assembled by repeating this procedure to make the final graphite/hBN/MATTG/hBN/graphite sandwich as shown in Fig. 1a. To assemble the MATTG, we precut the graphene flake into three separate pieces to reduce the strain which is otherwise introduced during the picking-up process. The second and third graphene layers are rotated with a target angle around 1.6° and −1.6°, respectively, to achieve mirror symmetry. The whole stacking is finally released on a SiO$_2$ (285 nm)/Si$^+$ chip at 180 °C and further patterned into a Hall bar geometry by following a typical ebeam lithography and CHF$_3$/O$_2$ reactive ion etching process. Dual gates and Hall bars are edge-contacted with Cr (5 nm)/Au (50 nm) metal.

Measurements

The electronic transport measurements are carried out in a dilution fridge and at a base temperature of 35 mK unless specified. A standard lock-in technique with excitation frequency $f = 17.111$ Hz and a.c. current $<10$ nA is employed by using a Stanford Research SR860. The a.c. current is applied through a 10 MΩ resistor. The local top and bottom gate are connected to source meters (Keithley 2400) to tune the carrier density and displacement field. We further apply a global gate voltage $V_{g}$ and a.c. signal $V_{ac}$ to tune the carrier density and displacement field, which results in the correlated gap being slightly smaller. This is a consequence of the Dirac cone shifting in the displacement field at $\phi = 30°$ (with $\phi = 0°$, the gap of the Dirac cone is $\Delta = (4\nu - 2)e/h$). By increasing $\phi$ from $0°$ to $30°$, the Dirac cone is shifted by an amount $\phi\Delta = (4\nu - 2)e/h$, where $\phi\Delta$ is the shift in the peak position relative to $\nu = 0$. At each filling $\nu$, the Dirac cone moves up by $\phi\Delta$ with different slopes $d\nu/d\phi = (4\nu - 2)/\phi$.

Estimation of chemical potential, charge density and twist angle

Based on the filling status of D-LLs and flat bands at Fermi level $E_F$, the phases of a TTT in a perpendicular magnetic field are classified into four types (Supplementary Fig. 1): phase I, $E_F$ is located inside both an incompressible D-LL gap and a correlated flat band gap; phase II, $E_F$ is located inside a compressible Nth D-LL and an incompressible correlated flat band gap; phase III, $E_F$ is located inside both a compressible Nth D-LL and a flat band; phase IV, $E_F$ is located inside an incompressible D-LL gap and a compressible flat band. Phase I hosts a quantized $R_N$ plateau. Phases II and III host an $R_N$ peak as a result of partially filling a correlated D-LL. By changing the magnetic field, phase I with $R_N = h/(4N - 2)e$ transitions to that with $R_N = h/(4N + 2)e$. This transition corresponds to phase II.

The potential of a flat band relative to the Dirac cone vertex $\mu' - \mu_{D-LL}^0$ with respect to charge filling $v$ is estimated through phase III. When the Nth D-LL crosses the flat band, $\mu' - \mu_{D-LL}^0$ equals $\mu_{N-LL} - \mu_{N-LL}^0$, where $\mu_{N-LL} - \mu_{N-LL}^0$ is the chemical potential difference between the Nth D-LL and the 0th D-LL, which can be extracted experimentally via $\mu_{N-LL} - \mu_{N-LL}^0 = \nu_I \Delta \mu_{N-LL}$. Here, $\nu_I$ is the Fermi velocity of the Dirac cone, $\Delta \mu_{N-LL}$ is the sign of the LL index $N = 0, \pm 1, \pm 2, \pm 3, \ldots$ and $B$ is a perpendicular magnetic field.

At lower displacement field $D$ where the flat band and the Dirac cone are decoupled, the charge density of a flat band $n_D$ is obtained as $n_D = n_t - n_n$, where $n_t$ is the total charge density which is acquired through the Hall effect or gate capacitance and $n_n$ is the charge density in the Dirac cone. Under a perpendicular magnetic field, when the Nth D-LL is half filled in phase III, $n_D$ is obtained as $n_D = 4e v^2/\Phi_0$, where $\Phi_0$ is the flux quantum and the factor 4 denotes the four-fold spin and valley degeneracy of the D-LL. This helps in calculating the charge density of a Dirac cone at zero magnetic field for various flat band fillings, for example, $n_D = 0.5 \times 10^{10}$ cm$^{-2}$ at integer filling $v = 2$ and $n_D = 1.2 \times 10^{10}$ cm$^{-2}$ at $v = 3$ at zero displacement field. The exact filling of the flat band is then defined by the relation $v = 4n_D/n_n$ ($n_n$ corresponds to full filling of the flat band). Alternatively, at each filling $v = 2, 3, 4, 5, 6, \ldots$, the trajectories of the Hall plateau or the longitudinal resistance $R_N$, minimum (or maximum) with different slopes $d\nu/d\phi = (4\nu - 2)e/h$ converge at zero magnetic field in an $n - B$ space. The $n'$ of the converging point is to the right of $n'$ for these integer or fractional fillings of the flat band. At higher displacement fields where the flat band and Dirac cone are maximally hybridized, $n'$ is directly obtained as $n' = n''$.

To extract the twist angle, we use the relation $n'' = \theta^2/\sqrt{3}a^2$, where $a = 0.246$ nm is the lattice constant of graphene.

Extraction of correlated gap

The precise measurement of the correlated flat band gap relies on precise determination of the crossing points $A_N$ and $B_N$ in the $n - B$ phase diagram. In Supplementary Fig. 1, the phase boundary between phase I and IV, or between phase II and III, corresponds to the state in which $E_F$ touches the flat band edge, the former of which produces resistive $R_N$ peaks in proximity to integer fillings due to D-LL edge state scattering with unfruno flat band charges in the tail of the split flat band (Fig. 3a), and the latter corresponds to $A_N$ and $B_N$. Meanwhile, phase II and the phase boundary between phases I and IV merge on $A_N$ and $B_N$. At critical points $A_N$ and $B_N$, the transition of the $R_N$ plateaus with $C = 4N \pm 2$ in the magnetic field, which is characteristic of phase II, disappears. In addition, the trajectory of the resistive $R_N$ peaks of phase II has a slope of $d\nu/d\phi = 4e/(4\nu - 2)e/h$ from phase I. This gives rise to a slope transition of resistive $R_N$ trajectories at $A_N$ and $B_N$ in the $n - B$ phase diagram. Notably, as flat band electrons exhibit negative compressibility in proximity to integer fillings, $B_N$ appears at the minimum of the magnetic field in the $n - B$ phase diagram. The overall features help to precisely identify $A_N$ and $B_N$. The correlated gap is extracted through the as-acquired magnetic field at $A_N$ and $B_N$. The error of our measurement is determined by the D-LL broadening, which is associated with device disorders and the magnetic field.

Note that we measure the gap under different magnetic fields. By varying the D-LL index, the impacts of the change in magnetic field on the correlated gap can be revealed. At $v = 2$ and $D = 0$, when the D-LL index changes from $N = 1$ to $N = 2$, the magnetic field correspondingly changes from around 0.45 T to 0.25 T (Fig. 3a). This induces a reduction of the gap of $\sim 0.5$ meV (with $N = 2$, $\Delta = (v = 2)$ is calculated to be $\sim 4.3$ meV), which is much smaller than the extracted correlated gap. Note that due to the Dirac cone shifting in the displacement field at an elevated displacement field, phase II appears at a lower magnetic field, which results in the correlated gap being slightly smaller. This leads to a slight underestimate of the D-induced enhancement of the correlated gap at $v = 2$.

Data availability

Source data for the main figures and the Extended Data figures are provided along with this paper. Other supporting data are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Acknowledgements

We thank B. Andrei Bernevig, L. Xian and Q. Wu for fruitful discussions and I. Das, A. Jaoui, C.-W. Cho and B.A. Piot for the help with cryogenic measurements. P.J.L. acknowledges fruitful discussions with M. Christos and support by the Department of Defense (DoD) through the National Defense Science and Engineering Graduate Fellowship (NDSEG) Program. E.K. was supported by the German National Academy of Sciences Leopoldina through grant number LPDS 2018-02. A.V. was supported by a Simons Investigator award and by the Simons Collaboration on Light子...
Ultra-Quantum Matter, which is a grant from the Simons Foundation (grant number 651440). D.K.E. acknowledges support from the Ministry of Economy and Competitiveness of Spain through the ‘Severo Ochoa’ programme for Centres of Excellence in R&D (SES-0522), Fundació Privada Cellex, Fundació Privada Mir-Puig, the Generalitat de Catalunya through the CERCA programme, funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement number 852927). K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan (grant number JPMXP0112101001) and JSPS KAKENHI (grant numbers 19H05790, 20H00354 and 21H05233).

Author contributions
C.S. and D.K.E. conceived of the project. C.S. fabricated devices, performed transport measurements and analysed the experimental data. P.J.L., E.K. and A.V. performed the numeric simulations. K.W. and T.T. provided the hBN crystals. C.S., P.J.L., E.K., A.V. and D.K.E. discussed the data. C.S., P.J.L., E.K., A.V. and D.K.E. wrote the paper.

Competing interests
The authors declare no competing interests.

Additional information
Extended data is available for this paper at https://doi.org/10.1038/s41563-022-01428-6.

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41563-022-01428-6.

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Peer review information Nature Materials thanks Oleg Yazyev and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Landau fan diagram and Landau level crossing at $D=0\text{V}/\text{nm}$. a, Landau fan diagram shown by longitudinal resistance $R_{xx}$ and transverse Hall resistance $R_{xy}$. b, Schematic of Landau level structure as observed in panel (a) and panel (b).
Extended Data Fig. 2 | Phase diagram in perpendicular magnetic field and chemical potential measurement at $D = 0V/nm$. a, four types of phases in perpendicular magnetic field when Dirac cone coexists with moiré flat band. The dash lines denote phase boundaries. Phases II and III correspond to partially filling $N$th D-LL, which shows finite band broadening due to disorder effects. The phase boundary between phase II and III, namely $A_N$ and $B_N$, is marked with solid line. In illustration schematics for each phase, the emergent correlated gap is shown by flat band splitting. b, chemical potential measurement at $D = 0V/nm$. The dark cyan dots show energy difference between flat band and Dirac cone vertex, which is obtained via phase III with D-LL index $N = 1$. The yellow dash line denotes a tentative plotting of Dirac cone shifting as a function of charge filling, where the zero energy corresponds to energy of flat band charge neutrality.
Extended Data Fig. 3 | Zoom-in Landau fan diagram around ν=2 at moderate displacement field. The top, middle and bottom panels are mapping plots of longitudinal resistance $R_{xx}$ of two neighbour regions R1 and R2 and Hall resistance $R_{xy}$, respectively. From the left to the right panel, displacement field $D$ is $D = 0, 0.05, 0.1, 0.15$ and $0.2 \text{V/nm}$ in sequence.