Anomalous Hall effect at half filling in twisted bilayer graphene

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Magic-angle twisted bilayer graphene (tBLG) displays a variety of symmetry-broken phases, correlated Chern insulators, orbital magnetism and superconductivity. In particular, the anomalous Hall effect has been observed when the bands are filled with an odd number of electrons per moiré unit cell, indicating the emergence of a zero-field orbital magnetic state with spontaneously broken time-reversal symmetry. Here we present measurements of two tBLG devices with twist angles slightly away from the magic angle and report the observation of the anomalous Hall effect at half filling of both the electron and hole moiré bands. We suggest that two factors—the increased band dispersion away from the magic angle, and substrate potentials from the encapsulating boron nitride—probably play critical roles in stabilizing a valley-polarized substrate potentials. Around the magic angle of \( \theta = 1.1° \), strong Coulomb coupling enters the tBLG Hamiltonian and is the bandwidth) have so far predicted ground-state orders that are broadly consistent with experimental observations at each integer band filling factor, \( \nu \).

The ratio of \( U/W \) describing the interaction strength decreases as the twist angle is tuned away from the magic angle, and eventually the band dispersion can no longer be treated as a small perturbation to the Coulomb energy. This regime of intermediate coupling strength, in which \( U/W \sim 1 \), has been the subject of substantially less theoretical and experimental attention so far, but may hold important clues necessary for developing a complete understanding of the correlated regime.

Figure 1a,b shows Landau fan diagrams for our two devices (D1 with \( \theta = 0.96° \) and D2 with \( \theta = 1.20° \), respectively; Extended Data Fig. 1). Below each, we plot the device resistivity within the flat band (\( \sim 4 \leq \nu \leq 4 \)) acquired in the absence of a magnetic field and at our nominal base temperature of \( T \leq 100 \text{ mK} \). We observe many features typical of magic-angle tBLG at high field, including a cascade of correlated Chern insulator states and associated fans of quantum oscillations projecting to various integer \( \nu \) values. However, we do not observe robust zero-field insulating states for any integer \( \nu \), most probably indicating that the samples are in a moderately correlated regime.

Our primary result is the observation of the AHE within a small region of doping around half filling. Figure 1c,d shows a representative measurement of the Hall resistance, \( \rho_{xy} \), in devices D1 and D2 as the magnetic field, \( B \), is swept back and forth at \( \nu = +1.93 \) and \( -1.99 \), respectively. We observe hysteresis and Barkhausen jumps indicating the presence of magnetism in these states.

Figure 2a shows the temperature dependence of the AHE in device D1 at \( \nu = +1.93 \), acquired by taking the average difference of \( \rho_{xy} \) between the two field-sweeping directions, \( \Delta \rho_{xy} = \langle \rho_{xy}^+ \rangle - \langle \rho_{xy}^- \rangle \). Figure 2b plots \( \Delta \rho_{xy}/2 \) as a function of temperature. Although the Curie temperature is \( \sim 5.5 \text{ K} \), the amplitude of the AHE grows slowly as the device is cooled to base temperature, and saturates to a value of less than 1 kΩ. Taken at face value, these observations imply that the ground state at \( \nu = +2 \) is an ungapped symmetry-broken metal, given that the AHE amplitude is far from the quantized value of \( h/2e \) anticipated at half filling. However, twist-angle disorder arising due to unintentional strain in the device may instead play a
role in obscuring a small intrinsic gap at $\nu = +2$. In either case, our observations sharply contrast the behaviour of the trivial correlated insulating states that are conventionally observed in devices closer to the magic angle, which exhibit insulating longitudinal and Hall resistance without associated hysteresis$^{1-4}$.

Figure 3a shows a zoomed-in field-symmetrized Landau fan diagram of $\rho_{xy}$ for device D1. At high field, we observe a fan of two-fold degenerate quantum oscillations that project to $\nu = +2$ at $B = 0$ and disperse towards larger filling factor, away from the charge neutrality point (CNP, $\nu = 0$). These indicate the formation of a new Fermi surface with reduced size compared with the isospin-unpolarized phase nearer the CNP, as has been observed regularly in magic-angle tBLG owing to a doping-dependent symmetry-breaking cascade$^{22,23}$. Figure 3b shows the corresponding field-antisymmetrized measurement of the Hall resistance, $\rho_{xy}$. At slight underdoping of $\nu = +2$, we see a region in which the Hall effect reverses sign, corresponding to a weakly developed quantum oscillation with an apparent Chern number of $C = -4$ projecting towards smaller filling factor. This state is eventually interrupted at high field by the formation of a $C = +3$ correlated Chern insulator projecting to $\nu = 1$ at $B = 0$. Figure 3c summarizes the most robust gapped states we observe, with trivial insulators denoted in red, correlated Chern insulators in blue, and additional quantum oscillations in grey and green. The $\rho_{xy}$ sign reversal and the associated $C = -4$ state depicted by the green line in Fig. 3c contrast the typical behaviour of magic-angle tBLG devices, in which the cascade of symmetry-breaking transitions arise only very near each integer filling and result in fans of quantum oscillations that disperse exclusively towards larger band filling$^{22-24}$. The precise details of these isospin Stoner transitions depend sensitively on the value of $U/W$ (refs. 21), potentially accounting for the difference in the behaviour of the quantum oscillations we observe in this device.
Although the $\rho_{xy}$ sign reversal persists to $B=0$, careful measurements of the AHE suggest that its origin is different below and above the coercive field, $B_c$. For $B>B_c$, the sign change arises owing to the C=−4 state discussed earlier. For $B<B_c$, the sign change instead arises as a consequence of a reversal of the orbital magnetic state upon doping. We perform two distinct measurements to clearly identify the latter effect. First, in Fig. 3d we plot $\Delta \rho_{xy}/2$ acquired by sweeping $B$ back and forth at fixed $\nu$, and observe an abrupt sign change precisely at $\nu=+2$. Figure 3e shows $\rho_{xy}$ traces at selected values of $\nu$, in which we see that the sense of the AHE flips upon doping across half filling. Second, in Fig. 3f we plot $\rho_{xy}$ acquired by sweeping $\nu$ from small to large values at fixed $B$, $\rho_{xy}$ traces at $B=\pm 50 \text{ mT}$ acquired for both sweeping directions of $\nu$, as indicated by the black dashed lines. The blue and red arrows indicate the field sweeping direction. The data are acquired at $T=20 \text{ mK}$ in all measurements.

Previous measurements of the magnetism underlying the AHE in tBLG indicate that it is driven primarily or exclusively by orbital magnetic moments. Owing to the extraordinarily weak spin-orbit coupling (SOC) in graphene, the AHE we observe here is almost certainly also driven by orbital magnetism. Consequently, a correlated ground state at half filling with a spontaneous valley polarization is the most natural mechanism consistent with all of our observations described above. Such a state spontaneously breaks time-reversal symmetry, resulting in an AHE due to the large Berry curvature concentrated at the band extrema. Breaking the combined $C_2 T$ symmetry ($C_2$ is a two-fold rotation and $T$ is time reversal) is the most straightforward mechanism for achieving valley imbalance at half filling. In the absence of $C_2$ symmetry, the tBLG bands acquire a staggered sublattice potential mass that opens a gap at the Dirac point, separating the eight flat bands into two distinct groups of four. Each moiré subband carries a valley Chern number of either +1 or −1, with subbands in opposite valleys carrying opposite signs of the valley Chern number owing to their time-reversed relationship. Both of our devices exhibit a base temperature resistivity above 25 kΩ, nearly an order of magnitude larger than typical tBLG devices, indicating weak $C_2$ symmetry breaking from close (but not exact) rotational alignment with the encapsulating BN (see Methods and Extended Data Figs. 3 and 4 for further discussion).

At $\nu=−2$, two of the eight moiré subbands are (un)occupied. In this case, there are a handful of nearly degenerate ground states predicted by Hartree–Fock calculations in the absence of $C_2$ symmetry, including (1) a valley-polarized, spin-unpolarized quantum
a more rigorous theoretical treatment of tBLG, very probably taking into account the role of the increased band dispersion away from the magic angle (that is, towards the intermediately coupled regime) and symmetry-breaking substrate potential terms from the encapsulating BN.

Finally, we discuss some of the implications of our findings in the context of other recent observations in tBLG. The AHE has been reported at \( \nu = +2 \) in a magic-angle tBLG device assembled on a monolayer of WSe\(_2\) (ref. \( ^{21} \)). However, in this case the proximity-induced SOC in the tBLG couples the spin and valley degrees of freedom, and a generalized Hund’s term explicitly favours the VP-QAH state at \( \nu = 2 \). A separate report of a gate-tunable Josephson junction tBLG device (without WSe\(_2\)) revealed evidence for some form of magnetic ordering at \( \nu = -2 \) (ref. \( ^{39} \)). However, the AHE was not reported, and the usual trivial correlated insulator was observed instead. The origin and nature of the magnetic order underlying the AHE in our devices is therefore likely to be different from these earlier reports in tBLG. Orbital magnetism has been observed at \( \nu = -2 \) in a magic-angle tBLG device with strong dielectric screening of the Coulomb interactions, and may be of similar nature to that found here, but required the assistance of a perpendicular magnetic field to emerge over a competing zero-field ground state\(^{36} \). Finally, we note that we do not observe any signatures of superconductivity in our devices (Fig. 1a,b), despite it having been seen previously in devices with similar twist angles\(^{20–24} \). Although it is impossible to rule out moiré disorder as the cause of its absence, a more likely possibility is that the valley-imbalanced ground-state order we observe at half filling is incompatible with pairing (see Methods for further discussion). Our results highlight the need for further measurements of devices slightly away from the magic angle to help disentangle the potential interplay of various forms of symmetry breaking and superconductivity.

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Methods

Device fabrication. The tBLG devices were fabricated using the cut-and-stack method\(^\text{35}\), in which exfoliated graphene flakes are isolated using an atomic force microscopy tip, and then stacked on top of one another at the desired twist angle. Samples were assembled using standard dry-transfer techniques with a polycarbonate/polydimethyl siloxane stamp\(^\text{35}\). Both tBLG devices are encapsulated in flakes of BN with a graphite bottom gate, and then transferred onto a Si/SiO\(_2\) wafer. Device D2 additionally has a graphite top gate. The temperature was kept below 18°C during device fabrication to preserve the intended twist angle. Standard electron-beam lithography, CHF\(_3\)/O\(_2\) plasma etching, and metal deposition techniques (Cr/Au) were used to define the complete stack into a Hall bar geometry\(^\text{36}\). Device D2 was measured while mounted onto a piezo-based apparatus capable of applying uniaxial strain to the device. All results, here, were acquired without any bias applied to the piezos, which corresponds closely to zero external strain (the results of the strain measurements will be discussed elsewhere).

Transport measurements. Transport measurements below 1.5 K were performed in Bluefors dilution refrigerators with heavy low-temperature electronic filtering. Measurements above 1.5 K were performed in a Cryomagnetics VTI system or in a PPMS DynaCool system. All measurements were conducted in a four-terminal geometry with an a.c. current excitation of 1–10 nA using standard lock-in techniques at a frequency of 17.8 Hz. A voltage was applied to the Si gate to dope the region of the graphene contacts overlapping the graphite back gate to a high charge carrier density and to reduce the contact resistance. Extended Data Fig. 1 shows optical micrographs of the two devices. Unless explicitly noted otherwise, all data from device D1 (D2) were acquired using contacts C–E (A–B) for \(\rho_{xx}\) and E–(A–F) for \(\rho_{xy}\).

Twist angle determination. The twist angle, \(\theta\), was first determined from the values of the charge doping, \(n\), at which the insulating states at full band filling (\(n = \pm 4\)) appeared, following \(n = 8\sqrt{\pi^2}\nu^2/\pi^2\), where \(\nu = 0.246\) nm is the lattice constant of graphene. It was then confirmed and refined by fitting the quantum oscillations observed at high field (see Fig. 3a–c for an example). The filling factor is defined as \(\nu = \sqrt{3}\lambda/\pi\), where \(\lambda\) is the period of the moiré. We extracted a twist-angle inhomogeneity of less than 0.05° in our devices (Extended Data Fig. 1).

Absence of the AHE away from half filling. We do not see evidence of the AHE at any filling factor outside of a small range of doping surrounding \(\pm 2\). One possibility is that the symmetry-broken states at \(\nu = 1\) and 3 are inherently non-magnetic in our devices. Another is that symmetry breaking at these integer fillings requires assistance from a magnetic field, thus precluding the AHE at \(B = 0\). Although we see weak zero-field resistance bumps at certain odd values of \(\nu\), none shows clear resonant features normally associated with symmetry-broken states in tBLG. A third possibility is that an intrinsic, but weak, AHE at odd fillings requires assistance from a magnetic field, thus precluding the AHE at \(B = 0\). Although this phenomenon could be completely obscured by the twist-angle inhomogeneity. We are unable to experimentally distinguish these three cases.

Candidate ground states at half filling in C\(_2\)-symmetric tBLG. Hartree–Fock calculations performed on tBLG with \(C_2\) symmetry in the strong coupling limit predict either a Kramers intervalley coherent (K-IVC) or an incommensurate Kekulé spiral (IKS) order\(^\text{37}\) (refs.\(^\text{14,18}\)), depending on the precise magnitude of heterostrain in the structure\(^\text{37}\). The IKS state is time-reversal symmetric\(^\text{19}\), and, although the K-IVC features local magnetic moments on the moiré scale, it is also time-reversal symmetric on spatial averaging\(^\text{20}\). None of these states naturally supports an AHE, and they are therefore inconsistent with our observations. It is therefore highly likely that the magnetic ground state we observe at half filling requires \(C_2\)-symmetry breaking (although there could be more exotic alternatives we have not considered here).

Possible alignment with the encapsulating BN. In principle, \(C_2\) symmetry can break spontaneously; however, it is typically observed when the tBLG is within a few degrees of rotational alignment with one of the encapsulating BN crystals. A large AHE approaching quantization to \(\pm 4\) was observed at \(\theta = \pm 3\) in previous tBLG devices with BN misalignment of less than 1°, most simply understood as arising from a complete spin and valley polarization\(^\text{21}\). Our observations contrast with these results, however, as we instead observe a metallic AHE only at \(\theta = \pm 2\) or \(\pm 4\).

Although we do not have a direct measure of the twist angle of either encapsulating BN crystal with the tBLG, we do not observe any signatures of additional superlattice minibands indicative of near-0° alignment. Nevertheless, we do observe significant large C\(_2\)-NPS resistivity in device D1 (Extended Data Fig. 4a), and extended Data Fig. 4a, as well as a weak thermal activation behaviour in device D1 (we did not measure this carefully in device D2). Additionally, careful inspection of the crystalline edges of the graphene and BN flakes provides additional evidence of their close angular alignment (Extended Data Fig. 3). Previous measurements of BN-encapsulated monolayer graphene have revealed the formation of multi-electronvolt-scale gaps at the graphene C\(_2\)NPs, even in devices in which the rotational misalignment of the BN approaches 5° (ref.\(^\text{22}\)) — well beyond the ability to reach full filling of the resulting moiré miniband with gating. We therefore speculate that both of our devices are probably somewhat (but not precisely) aligned to at least one of the encapsulating BN crystals, providing a natural mechanism for \(C_2\) symmetry-breaking. The orbital magnetism may depend sensitively on the precise alignment and near commensurabilities of the twisted graphene and graphene/BN moiré superlattices\(^\text{23–25}\), and the local fluctuations of the atomic registry may provide an additional source of magnetic disorder in the form of a rapid spatial fluctuation in the sign of the local mass gap\(^\text{26}\).

Absence of superconductivity: tBLG devices with similar twist angles to those we study here have previously been found to exhibit superconducting states surround half filling at low temperature\(^\text{27–30}\). However, our devices do not exhibit any obvious signatures of superconductivity. Most notably, the device resistivity surrounding half filling at base temperature is typically on the order of kilo-ohms (see the bottom panels of Fig. 1a,b). We cannot rule out trivial explanations, including temperature effects much lower than our base temperature which could suppress the occurrence of superconductivity due to severe local structural inhomogeneities in the sample. However, the proximity to the putative valley-polarized states at half filling motivates a more substantive explanation for its absence. Cooper pairs typically form with zero centre of mass momentum, and, as a consequence, electrons in tBLG are most likely to form intervalley pairs. Partial or full filling of the valley degeneracy therefore disfavours superconductivity, as the centre-of-mass momentum of the Cooper pairs becomes nonzero for weak valley polarization, and saturates to values of approximately the momentum of the K/K' corners of the graphene Brillouin zone when the valley is fully polarized (intravalley Cooper pairing limited). Although this approximation is consistent with the anticipated antagonistic relationship between the AHE at half filling and superconductivity at nearby doping. Although superconductivity and the AHE have been previously reported at disjoint band fillings in a single tBLG device\(^\text{26}\), so far they have never been seen proximate to one another.

Coexistence of competing ground states in device D2. We observe sharp dips in the resistivity of device D2 at filling factors slightly below \(\nu = -4, 2\) and \(\pm 3\) (Fig. 1b). These features project vertically upon applying a magnetic field. The resistance additionally becomes negative as \(B\) is increased near \(\nu = \pm 4\) and \(-2\), as shown in white in Fig. 1b. Four-terminal measurements of an insulating state often result in negative-resistance artefacts, especially in the presence of inhomogeneity, as the potential difference between the voltage probes can invert in the percolative transport regime. These artefacts therefore reveal the coexistence of the trivial insulators typically observed at these band fillings in magic-angle tBLG, presumably originating from a portion of the device with slightly smaller twist angle (\(\theta \approx 1.15°\)). Extended Data Fig. 5 shows additional transport measurements acquired with other contacts on the sample. The coexisting trivial correlated insulator at \(\nu = \pm 2\) is most clear in the contacts at the top of the device (corresponding to the region with AHE), and becomes progressively weaker in contacts towards the bottom of the device where the measured twist angle becomes slightly larger. The coexistence of the trivial correlated insulator and AHE states at \(\nu = \pm 2\) implies that they are probably nearly degenerate in this sample, with small changes in the twist angle responsible for the switching between the two. We note that we do not observe any similar features in device D1 (see Extended Data Figs. 6 and 7 for measurements acquired with additional contacts). The sense of the AHE does not flip as device D2 is doped across half filling (Extended Data Fig. 8), unlike for the case of device D1. We do not know the exact origin of this discrepancy, but speculate that the competing ground states and attendant magnetic disorder may play a role.

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.

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Author contributions
C.-C.T. and X.M. fabricated the devices. C.-C.T., X.M. and Z.L. performed the measurements. K.W. and T.T. provided the bulk BN crystals. C.-C.T., X.M., Z.L., J.-H.C. and M.Y. analysed the data and wrote the paper.

Competing interests
The authors declare no competing interests.

Additional information
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Extended Data Fig. 1 | Optical micrographs of the devices. a, Device D1. b, Device D2. The twist angle measured between neighboring pairs of contacts is also shown. The scale bars are 5 μm.
Extended Data Fig. 2 | Electric-field-reversal of the magnetic state in Device D1. a,b, $\rho_{xy}$ acquired by sweeping $\nu$ from small to large values (a), and from large to small values (b), at fixed $B$ and $T = 20$ mK. c,d, The same measurements performed at $T = 1.5$ K. At both temperatures, we observe hysteresis at low magnetic fields depending on the direction the doping is swept.
Extended Data Fig. 3 | Possible alignment of tBLG and BN. Optical micrographs of the completed heterostructures of a, Device D1 and b, Device D2 prior to device fabrication. The top and bottom BN flakes are outlined in green and blue dashed lines, respectively. Angular alignment between the tBLG and BN is estimated by comparing the orientation of the straight crystallographic edges of each. A representative graphene edge is denoted by the white dashed line, and the angular offset with a selected BN edge is denoted next to the corresponding black dashed lines. A few selected BN corners with modulo-30° angles are denoted by the solid black lines, establishing that these are likely zigzag or armchair edges. In both devices, the top BN and tBLG appear to have a 29–30° angular offset. This indicates a high likelihood of few-degree or smaller alignment of the graphene and top BN crystals, although there is inherent uncertainty in identifying perfect crystalline edges of graphene. The scale bars are 5 μm.
Extended Data Fig. 4 | Thermal activation of the CNP in Device D1. a, Resistivity of Device D1 at the CNP ($\nu = 0$) measured as a function of temperature, exhibiting insulating behavior below approximately 25 K. The red curve is measured in a VTI down to 1.5 K, and the blue curve is measured in a dilution fridge down to 20 mk. b, The same data shown on an Arrhenius plot. The CNP exhibits a small region of (approximately) activated behavior. We extract the band gap, $\Delta = 2.2$ meV, from the slope of the linear fit (red dashed line) using $\rho \propto e^{\Delta/kT}$, where $k$ is the Boltzmann constant.
Extended Data Fig. 5 | Transport measurements from additional contact pairs in Device D2. a–c, $\rho_{xx}$ and d–f, $\rho_{xy}$ Landau fan diagrams measured between different pairs of contacts corresponding to the contact labeling scheme in Extended Data Fig. 1b. The strength of the coexisting trivial insulating state at $\nu = -2$ varies substantially depending on the contact pair. g–i, Measurements of the AHE near $\nu = -2$ acquired using the same contacts as the associated $\rho_{xy}$ fans shown in (d–f). The data are acquired at the same gate voltage, which corresponds to a slightly different value of $\nu$ due to the twist angle disorder in the sample. The AHE is only observed in contact pairs A–F, despite the overall similarities of the Landau fans. For contacts B–G in particular, there is a large offset from $\rho_{xx} = 0$ due to mixing with $\rho_{xy}$. All measurements are acquired at $T = 100$ mK.
Extended Data Fig. 6 | Landau fan diagrams from additional contact pairs in Device D1. a–c, Landau fan diagrams of \( \rho_{xx} \) and d–f, \( \rho_{xy} \) measured between different pairs of contacts corresponding to the contact labeling scheme in Extended Data Fig. 1a. All measurements are acquired at \( T = 20 \) mK.
Extended Data Fig. 7 | AHE near $\nu = +2$ measured in additional contact pairs in Device D1. a, b, $\rho_{xx}$ and c, f, $\rho_{xy}$ acquired with the contacts denoted above each plot, following the labeling scheme shown in Extended Data Fig. 1a. The data are all acquired at the same gate voltage, which corresponds to a slightly different value of $\nu$ due to the twist angle disorder in the sample. All measurements are acquired at $T = 20$ mK.
Extended Data Fig. 8 | AHE versus doping in Device D2. \( \rho_{xy} \) measured as \( B \) is swept back and forth around at selected values of \( \nu \) around \( \nu = -2 \) in Device D2. The measurements are acquired at \( T = 100 \) mK.