Flavour Hund’s coupling, Chern gaps and charge diffusivity in moiré graphene

Interaction-driven spontaneous symmetry breaking lies at the heart of many quantum phases of matter. In moiré systems, broken spin/valley ‘flavour’ symmetry in flat bands underlies the parent state from which correlated and topological ground states ultimately emerge\(^{1-10}\). However, the microscopic mechanism of such flavour symmetry breaking and its connection to the low-temperature phases are not yet understood. Here we investigate the broken-symmetry many-body ground state of magic-angle twisted bilayer graphene (MATBG) and its nontrivial topology using simultaneous thermodynamic and transport measurements. We directly observe flavour symmetry breaking as pinning of the chemical potential at all integer fillings of the moiré superlattice, demonstrating the importance of flavour Hund’s coupling in the many-body ground state. The topological nature of the underlying flat bands is manifested upon breaking time-reversal symmetry, where we measure energy gaps corresponding to Chern insulator states with Chern numbers 3, 2, 1 at filling factors 1, 2, 3, respectively, consistent with flavour symmetry breaking in the Hofstadter butterfly spectrum of MATBG. Moreover, concurrent measurements of resistivity and chemical potential provide the temperature-dependent charge diffusivity of MATBG in the strange-metal regime\(^{11}\)—a quantity previously explored only in ultracold atoms\(^{12}\). Our results bring us one step closer to a unified framework for understanding interactions in the topological bands of MATBG, with and without a magnetic field.

In condensed matter systems with flat electronic bands, the Coulomb interaction between electrons can easily surpass their kinetic energy and give rise to a variety of exotic quantum phases, including Mott insulators, quantum spin liquids and Wigner crystals\(^{13-15}\). In this strongly correlated regime, electrons may spontaneously order themselves to minimize the total Coulomb energy at the cost of increasing their kinetic energies, leading to the breaking of certain symmetries. Such broken-symmetry states can occur at a relatively high energy scale, and act as a parent state for phases that appear at lower energy scales, such as superconductivity. Furthermore, when there is a nontrivial topology in the system, the interplay between strong correlations and the underlying topology could lead to novel phases of matter. Understanding the physics behind this interplay could guide us in designing next-generation strongly correlated topological quantum materials.

MATBG serves as a unique platform to investigate interaction-driven phenomena in a highly tunable flat-band system. When two layers of monolayer graphene (MLG) are stacked with a small twist angle of \(\theta = 1.1^\circ\), the interlayer hybridization in the resulting moiré superlattice renormalizes the Fermi velocity and creates flat bands at low energies\(^{16,17}\). In this regime, a plethora of exotic correlated and topological phenomena have been experimentally demonstrated, including correlated insulator states, superconductivity and the quantum anomalous Hall effect\(^{1,2,4,7}\). Scanning tunnelling and single-electron transistor experiments have directly shown the importance of Coulomb-induced phase transitions that break the spin/valley symmetry\(^{18,19,21}\). Despite substantial experimental and theoretical progress, the microscopic picture that underlies the broken-symmetry states and their possible connections to the correlated phases and superconductivity requires further investigation.

**Flavour Hund’s coupling in MATBG**

Here we study the interplay between interaction-driven symmetry breaking and nontrivial topology in MATBG by directly measuring the combined thermodynamic and transport properties of its many-body ground state. We use the technique described in ref. \(^{22}\) to sense the chemical potential of MATBG. The MATBG is separated from an MLG layer by an ultrathin layer of hexagonal boron nitride (hBN; about 1 nm thick; Fig. 1a). We use the top gate voltage \(V_{tg}\) and back gate voltage \(V_{bg}\) to control the densities in MLG and MATBG, and measure the transport properties of the two layers simultaneously. Direct probing of the chemical potential \(\mu\) of one layer is achieved by sensing the screening of the electric field from the gates by the other layer\(^{23}\) (Fig. 1b, Supplementary Information). In particular, when one layer is at the charge-neutrality point (CNP), for example, \(n_{\text{MLG}} = 0\), where \(n_{\text{MLG}}\) is the carrier density of MLG, the chemical potential \(\mu_{\text{MATBG}}\) of the other

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layer is given by $\mu_{\text{MATBG}} = -\left(\frac{eQ_{\text{btg}}}{C_{\text{t}}}ight)V_{\text{btg}}$, where $Q_{\text{btg}}$ and $C_{\text{t}}$ are the geometric capacitances per unit area of the top and middle hBN dielectrics, respectively, and $e$ is the electron charge. The MLG layer used in our experiments has very low disorder of $<3 \times 10^3$ cm$^{-2}$ (Fig. 1c). The MATBG layer has a twist angle of $\theta = 1.07^\circ \pm 0.03^\circ$, and exhibits correlated states at integer filling factors $\nu_{\text{MATBG}} = 1, \pm 2, 3$, as well as superconducting domes (blue) at $-2 - \delta$ and $-2 + \delta$, respectively. Combined plot of the resistance of MLG and MATBG, represented by the purple and orange colour scales, respectively, and overlaid on the same axes. As a proof of principle, we use the CNP of MATBG (orange diagonal feature) to probe the chemical potential of MLG, at $B = 0$ (e) and $B = 1$ T (f). The horizontal purple stripes are the resistive features in MLG. From the CNP of MATBG, we extract the chemical potential of MLG versus density, which is shown in the insets of e–f. The yellow line in the inset of e is a fit to $\mu_{\text{MLG}} = -\hbar v_F \sqrt{2e\hbar B \delta_1 / M_{\text{MLG}}}$. The red ticks in the inset of f denote the expected Landau level energies $\pm v_F \sqrt{2e\hbar B}$, where $v_F = 1.12 \times 10^6$ m s$^{-1}$ and $\delta_1$ is an integer.

Information), from which we determine the MLG Fermi velocity to be $v_F = 1.12 \times 10^6$ m s$^{-1}$ by fitting to $\mu_{\text{MLG}} = -\hbar v_F \sqrt{2e\hbar B \delta_1 / M_{\text{MLG}}}$ ($\hbar$, reduced Planck constant). In a magnetic field of $B = 1$ T, the spectrum of MLG is quantized into discrete Landau levels at energies of $\pm v_F \sqrt{2e\hbar B \delta_1 / M_{\text{MLG}}}$, as expected. Our technique can thus determine the chemical potential of either layer with a sensitivity of $\pm 1$ meV.

The chemical potential of MATBG is shown in Fig. 2a. Hereafter we will simply use $n$ (v) and $\mu$ to denote $n_{\text{MATBG}}$, $\mu_{\text{MATBG}}$ and $\mu_{\text{MLG}}$. The $V_{\text{bg}}$ axis is directly proportional to $\mu$ when tracking the CNP of MLG (shown as the green curve). The longitudinal resistance $R_{\text{xx}}$ of MLG (purple) and MATBG (orange) is overlaid for qualitative comparison, and the grey dashed lines indicate the integer filling factors of $\nu = 0, \pm 1, \pm 2, \pm 3$. Around the MATBG CNP ($\nu = 0$), $\mu$ rises quickly with $\nu$, consistent with a minimal density of states (DOS) at the Dirac point. However, once we shift electrons into the flat band, its slope decreases quickly and $\mu$ reaches a local maximum around $\nu = 0$. Surprisingly, it then decreases, exhibiting a negative inverse compressibility $\chi' = d\mu/dn$.
flavour symmetry breaking occurs before each integer filling factor (except \( T = 2 \text{ K} \) near interaction-driven chemical potential stabilization at \( \nu = 1 \). The chemical potential can be explained qualitatively when both terms are non-zero.

Illustration of interaction-driven chemical potential stabilization at \( \nu = 1 \). The chemical potential curve at \( T = 2 \text{ K} \) near \( \nu = 1 \) is shown. A phase transition associated with flavour symmetry breaking occurs before each integer filling factor (except \( \nu = 4 \)). The exchange energy stabilizes the filled flavour when the filling factor is close to 1. See Methods for further details. The temperature dependence of the chemical potential of MATBG from \( T = 2 \text{ K} \) to \( T = 70 \text{ K} \), probed with the MLG CNP.

A clear pinning behaviour at integer filling factors persists up to \( T = 20 \text{ K} \).

Dependence of the chemical potential of MATBG on the in-plane magnetic field, \( B \perp \), at \( T = 4 \text{ K} \) and \( B = 0.7 \text{ T} \), probed with the N = 1 MLG Landau level. The pinning of the chemical potential around odd filling factors \( \nu = \pm 1 \) gets intensified as \( B \perp \) is applied, whereas those at filling factors \( \pm 2 \) do not display substantial change. Inset, magnetization \( M \) in units of \( \mu_0 \) per moiré unit cell, showing that all states at \( \nu = \pm 1, \pm 2, \pm 3 \) are magnetized in an in-plane field. Error bars (blue) correspond to the 95% confidence interval. Zoom-in of the chemical potential (top) and transport resistance (bottom) aligned for comparison, shown for \( \nu = +1, +2 \). The plots for other fillings are shown in Extended Data Fig. 2. The spacing between ticks on the horizontal axis is 0.2.

Fig. 2 | Chemical potential of MATBG as a function of temperature and in-plane magnetic field. (a) Sensing the chemical potential of MATBG using the CNP of MLG. Measurement taken at \( B = 0 \text{ T} \) and \( T = 4 \text{ K} \). The green line shows the extracted chemical potential of MATBG. Gray dashed lines mark the integer filling factors of MATBG, which agree with the MATBG correlated resistive features. The chemical potential is pinned at each filling factor, showing the stabilization of the state. The inset shows the same features probed by tracking the \( N = 1 \) MLG Landau level at \( B = 0.7 \text{ T} \). (b) Mean-field estimate of the chemical potential with various Coulomb repulsion energies \( U \) and exchange energies \( J \) in units of the single-particle bandwidth, \( W = 1 \). The experimental data are best explained qualitatively when both terms are non-zero. Illustration of interaction-driven chemical potential stabilization at \( \nu = 1 \). The chemical potential at \( T = 2 \text{ K} \) near \( \nu = 1 \) is shown. A phase transition associated with flavour symmetry breaking occurs before each integer filling factor (except \( \nu = 4 \)). The exchange energy stabilizes the filled flavour when the filling factor is close to 1. See Methods for further details. (d) Temperature dependence of the chemical potential of MATBG from \( T = 2 \text{ K} \) to \( T = 70 \text{ K} \), probed with the MLG CNP.

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(Ref. 23), and gets pinned at a local minimum around \( \nu = 1 \). Subsequently, \( \mu \) rises again until it reaches the next maximum. This intriguing pinning behaviour is repeated at each integer filling factor, including \( \nu = 4 \) (Fig. 2a inset). On the hole-doped side (\( \nu < 0 \)), the pinning behaviour is reversed and weaker (that is, creates weak maxima in \( \mu \)). The total bandwidth estimated from \( \mu \) is around 40 meV. We also investigated \( \mu \) versus temperature from 2 K to 70 K (Fig. 2d). The observed pinning behaviour persists prominently up to 20 K. We point out that the pinning of \( \mu \) should not be interpreted as a measure of the gaps of the insulator states because its energy scale (visible up to 70 K) is much greater than the typical energy scale of the insulator states (typically below 10 K). Instead, the insulator state, and possibly also the superconducting state, might be thought of as low-energy states that emerge from the ‘parent’ states of the broken flavour symmetry. We also note that the pinning on the hole-doped side occurs at slightly more negative values of \( \nu \) (Supplementary Information).

The pinning of \( \mu \) at all integer \( \nu \) is reminiscent of the stabilization of half-filled or fully filled electronic shells in atoms, which is known as Hund’s rule for maximum spin multiplicity and stems from the Coulomb exchange interaction between the electrons. In MATBG, the pinning behaviour of the chemical potential is naturally explained when both the on-site inter-flavour Coulomb repulsion energy \( U \) and the inter-site intra-flavour exchange energy \( J \) are considered. We focus on \( \nu > 0 \) in the following description. Figure 2b shows \( \mu \) calculated with a mean-field model for different values of \( U \) and \( J \) (Supplementary Information), which qualitatively reproduces the experimentally measured \( \mu \) only when both \( U \) and \( J \) are non-zero and of similar magnitude (purple solid curve), beyond the currently established understanding9,10,21. A possible mechanism for such stabilization of \( \mu \) at \( \nu = 1 \) is illustrated in Fig. 2c and elaborated in Methods. We note that the mean-field treatment of the Coulomb interactions correctly captures the many-body compressibility to leading order21, but might not give the same ground state as the exact solution. Other mechanisms, such as the formation of a Wigner crystal24, might also be relevant to the observation of negative compressibility.

To probe the magnetic properties of the correlated states, we measured \( \mu \) as a function of in-plane magnetic field up to 11 T (Fig. 2e, f for \( \nu = +1 \) and \( \nu = +2 \); Extended Data Fig. 2 for \( \nu = -1, -2, +3 \)). At \( B = \pm 1 \), the pinning of \( \mu \) is clearly strengthened by \( B \perp \) (the magnetic field parallel to the plane), as is the peak in \( \rho_{xx} \) (Methods, Extended Data Fig. 3), suggesting that the \( \nu = \pm 1 \) states develop a spin polarization in response to the magnetic field. To confirm this, we directly obtained the magnetization by integrating Maxwell’s relation26 (Fig. 2e inset). We indeed find that the magnetization reaches a value of the order of 1 Bohr magneton \( (\mu_B) \) at \( \nu = ±1 \), consistent with a spin-polarized state at finite field, which would indicate either a very soft paramagnetic state or a ferromagnetic state at zero field. The \( \nu = ±2 \) states, on the other hand, have been speculated to be spin-unpolarizing insulating states9,25,26. However,
we find that, whereas the transport peak is indeed suppressed by $B_i$ (see Fig. 2f, Extended Data Fig. 3), $\mu$ measured at $v = \pm 2$ does not show any evident dependence on the in-plane magnetic field (Fig. 2f). Furthermore, the in-plane magnetization $M_i$ does not return to zero when $v$ is tuned from $\pm 1$ to $\pm 2$ (Fig. 2e inset). Although the lack of dependence of $\mu$ on the in-plane magnetic field can be partially captured by our theoretical model (Supplementary Information), the persistence of magnetization near $v = \pm 2$ is at odds with the finite-field spin-unpolarized ground state inferred from transport. These observations suggest that in an in-plane field, $v = \pm 2$ gaps might select a ground state with nontrivial spin and/or valley texture, instead of simply occupying two flavours with opposite spins.

Our experiments also constrain the possible mechanism of superconductivity in MATBG. The superconducting dome lies in the region where $\chi^1$ is high (Extended Data Fig. 4b), with the maximum $T_c$ corresponding to a maximum in $\chi^1$. Because a Bardeen–Cooper–Schrieffer (BCS)-type superconductivity in the weak-coupling limit would be enhanced when the DOS is high (and thus $\chi^1$ is low), our observation of an opposite trend indicates that it is hard to reconcile the superconductivity in MATBG with a weakly coupled BCS theory. Future theories attempting to model the superconductivity in MATBG will probably need to consider the importance of Coulomb interactions, including both repulsion and Hund’s coupling, and the consequent phase transitions.

**Correlated Chern insulators**

We now turn to the topological properties of MATBG. By measuring $\mu$ in a perpendicular magnetic field, we can observe the energy gaps that result from the interplay between the Hofstadter spectrum and the Coulomb interactions\(^{-39-39}\). The helical nature of the Dirac electrons in graphene endows each flat band of MATBG with a Chern number of $C = \pm 1$, which is usually explicitly manifested when the composite $C_2T$ symmetry is broken, either by alignment to the hBN substrate (breaks $C_2$) or by applying a magnetic field (breaks $T$). Figure 3c shows the Hofstadter butterfly spectrum of TBG, in which the topologically nontrivial gaps ($C = \pm 1$) and the trivial gaps ($C = 0$) are shown. The former gaps are smoothly connected to the Landau level gaps at $v_{LL} = v/\phi_0 = \pm 4$ at low fields, where $\phi$ is the magnetic flux per unit cell and $\phi_0 = h/e$ is the flux quantum.
quantum. Without interactions, the only possible total Chern number in this picture is \( C = 0, \pm 4 \), given that all factors are in the same gap. The Coulomb interactions cause their Chern numbers to be different and give rise to new hierarchies of Chern gaps.

These topological gaps are directly observed in Fig. 3a. Near charge neutrality, we observe the gaps as steps in \( \mu \) at the Landau level filling factors \( \nu_L = 0, \pm 2, \pm 4 \), the positions of which evolve according to the Streda formula \( \nu_L = \frac{1}{\hbar v_F T} \), where \( \rho \) is obtained by fitting the linear-in-\( T \) range and extrapolating to \( T = 0 \).

A more quantitative analysis is performed on the chemical potential measured at \( B_z = 6 \) T (Fig. 3e). From the steps in \( \mu \), we extract the values of the Landau level gaps at \( \nu_L = -2, 0, 2, 4 \) to be 5.9, 3.3, 5.9, 2.3, 4.9 meV, respectively. The small values of the gaps at \( \nu_L = 0, \pm 4 \) translate to a Fermi velocity of approximately \( v_F = 6 \times 10^4 \) m s\(^{-1} \), consistent with previous experiments\(^{27}\). The Chern gaps at \( \nu = 1 + 3\phi/\phi_0, 2 + 2\phi/\phi_0 \) and \( 3 + \phi/\phi_0 \) are extracted to be 2.2, 5.0 and 1.9 meV, respectively. The larger gap at \( 2 + 2\phi/\phi_0 \) is consistent with the fact that this state is more readily resolved in electronic transport experiments\(^{12,14,28,35}\). Its difference with the gaps at \( \nu = 1 + 3\phi/\phi_0 \) and \( 3 + \phi/\phi_0 \) might be attributed to different magnetic ground states. These gaps have a weak dependence on \( B_z \) (Extended Data Fig. 5), consistent with the Hofstadter spectrum (Fig. 3c).

**Charge diffusivity of a ‘strange metal’**

In correlated metals with multiple bands near the Fermi energy, the atomic Hund’s coupling is known to play an important role in their many-body physics, including the strange-metal regime\(^{31}\). In MATBG, recent experiments have reported evidence for strange-metal behaviour\(^{15}\), manifested as resistivity linear with temperature \( T \) from very low \( T \). As shown in Fig. 4a, c, the resistivity in our MATBG sample is largely linear with \( T \) over a range of densities around the correlated states, and has a slope that is weakly dependent on \( n \) (refs. \(^{11,32}\)). It has been hypothesized that the strange-metal behaviour can be
universally described by a ‘Planckian’ scattering rate bound of \( D = k_B T / h \) (\( k_B \), Boltzmann constant) in the framework of incoherent non-quasiparticle transport\(^{36,37}\). However, the construction of a microscopic picture for this bound is still in progress\(^{36,37}\).

A universal framework to investigate the strange-metal regime is the Nernst–Einstein relation, which connects the resistivity \( \rho \), compressibility \( \chi \) and charge diffusivity \( D \) of a generic conductor by \( \rho / \chi T = e^2 D / \hbar \). A linear-in-\( T \) resistivity could thus originate from: (i) \( \chi \propto T \), which could arise from thermodynamic contributions\(^{38,39}\) when \( k_B T \geq W \) (\( W \) is the single-particle bandwidth); (ii) \( D \propto T \), which represents a linear scattering rate; or (iii) a combined \( T \) dependence of both. Differentiating between these possibilities could help to constrain theoretical models for strange-metal behaviour\(^{40,41}\). However, to the best of our knowledge, there have been no reported measurements of the compressibility or charge diffusivity of strange metals, and only recent experiments have begun to explore this physics in ultracold atoms\(^2\).

Our combined resistivity and compressibility measurements allow us to extract the charge diffusivity of MATBG (Fig. 4b). Although \( \chi \) becomes negative before each integer filling factor at \( T \leq 20 \text{ K} \), as discussed above, at higher \( T \) it converges to a roughly constant value of the order of \( 10^3 \text{ eV nm}^2 \), regardless of \( v \). Figure 4d shows selected traces of \( \chi \) versus \( T \), which exhibit only a weak dependence on \( T \), albeit \( \rho \) exhibits a prominent linear-in-\( T \) behaviour, suggesting that the linear \( \rho \propto T \) behaviour in MATBG is mainly due to a \( T \)-dependent charge diffusivity. Figure 4e–f shows the \( T \) dependence of the extracted effective diffusivity \( D^\alpha = \chi / [\rho (\rho - p_0)] \) and its inverse, where \( p_0 \) is the residual resistivity extrapolated to zero temperature. These quantities indeed appear to roughly follow a \( \propto T^{-1} \) and a \( \propto T \) trend, respectively. Our observations therefore indicate that the strange-metal regime in MATBG is consistent with a scattering rate linear in \( T \). These arguments do not apply to regions with negative electronic compressibility, because the interpretation of diffusivity in this case needs to be modified\(^{42}\) (Supplementary Information). Interestingly, we find the extracted effective diffusivity \( D^\alpha(T) \) at all these fillings to be within about a factor of 2 of a diffusivity bound of \( D_{\text{bound}} = \hbar c^2 / (k_B T) \) proposed for incoherent metals\(^{38}\). Although this bound is known to be violated in the low-temperature region in a large-\( U \)-system\(^5\), this is not at odds with our observations if MATBG is in the intermediate \( U \) regime (\( U/W = 1 \), deduced from our modelling and other experiments\(^{40,41}\)).

**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/s41586-021-03366-w.

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Method

Sample fabrication
The multilayer heterostructure consists of one sheet of MLG and TBG with a small twist angle of $\theta = 1.1^\circ$, separated by a thin (-1 nm) hBN layer. This sandwich is encapsulated by two hBN flakes. All flakes are first exfoliated on SiO$_2$/Si substrates and subsequently analysed with optical microscopy and atomic force microscopy to determine their thicknesses and quality. The multilayer heterostructure is fabricated by a modified polymer-based dry pick-up technique, whereby a layer of poly(bisphenol A carbonate)/polydimethylsiloxane on a glass slide fixed on the micro-positioning stage is used to sequentially pick up the flakes. The order of the pick-up is hBN–MLG–hBN (1 nm)–MLG–MLG, and the last two MLG sheets are laser-cut from one MLG flake (see Supplementary Information) and twisted by an angle of $-1.1^\circ$. All hBN layers are picked up at 90 °C, whereas the MLG layers are picked up at room temperature. The hBN–MLG–hBN (1 nm)–MLG–MLG heterostructure is then released on the pre-stacked hBN–Pd/Au back gate at 175 °C. The Hall bar geometry for the transport measurements is defined with electron beam lithography and reactive ion etching for each of the MLG and MLG–MLG layers. The top gate and electrical edge contacts are patterned with electron beam lithography and thermal evaporation of Cr/Au.

Measurement setup
Electronic transport measurements were performed in a dilution refrigerator with a superconducting magnet, with a base electronic temperature of 70 mK. The current through the sample, amplified by 10$^7$A, and the four-probe voltage, amplified by 1,000, were measured with SR-830 lock-in amplifiers synchronized at the same frequency between 1–20 Hz. A current excitation of 1 nA or a voltage excitation of 50–100 μV was used for resistance measurements. We measured both MLG and MATBG layers simultaneously for accurate comparison. See Supplementary Information for details about the extraction of chemical potential from the data.

Flavour symmetry breaking and negative compressibility at $\nu = 1$
In Fig. 2c we schematically illustrate the spontaneous flavour symmetry breaking at $\nu = 1$ due to the Coulomb interactions, and how this generates negative compressibility when a finite $\chi$ is present. Near charge neutrality, as the density is increased, all four flavours are filled at the same rate (Fig. 2c (i)). At $\nu = 1$, the Coulomb repulsion between different flavours starts to surpass the kinetic energy penalty of filling up only one flavour. When $\nu$ reaches a certain value (still below 1), a flavour-symmetry-breaking phase transition occurs and all electrons are transferred into a single flavour to minimize the Coulomb repulsion 43 (Fig. 2c (ii)). From this phase-transition point to $\nu = 1$, that is, while a single flavour is being filled, the $\chi$ term does not have any contribution to the free energy, whereas the $\nu$ term decreases the total free energy as $-\mu \chi$ (see Supplementary Information). This term decreases the chemical potential and results in a negative inverse compressibility $\chi = D^3 - 2J_D$ (see Supplementary Information). When $\nu > D$, maximal stabilization by the exchange term $\nu \chi$ is reached. Further increase in $\nu$ (Fig. 2c (iv)) populates the other three empty flavours and increases the chemical potential before the next phase transition occurs.

We note that systems with negative compressibility tend to phase separately in order to minimize the total free energy. The observation of a negative compressibility indicates that our system might be in a strong Coulomb frustration regime 43, which acts to suppress macroscopic phase separation that may otherwise occur in an unconstrained system.

Maxwell’s relations
Using Maxwell’s relations between thermodynamic variables, we can obtain information about various thermodynamic quantities by taking different derivatives of the chemical potential. The free energy of the system per unit area in the presence of magnetization can be written as $g = \mu - T s - M B_i$, where $\mu, s$ are the internal energy, in-plane magnetization and entropy per area, respectively. $g$ and $s$ satisfy

$$du = Tds + B_i dB_i + \mu dv,$$

$$dg = -sdT - M dB_i + \mu dv.$$

By taking the second derivative of $g$ with respect to $(v, B_i)$ in different orders, we can obtain the following Maxwell’s relationship

$$\frac{\partial M_i}{\partial v} = -\frac{\partial \mu}{\partial B_i} \bigg| _{T, v}.$$

Therefore, we can integrate from the $B_i$ derivative of $\mu$ to obtain the change in $M_i$ as a function of density $v$,

$$M_i(v=0) - \int_{v}^{v'} \frac{\partial \mu}{\partial B_i} \bigg| _{T, v} dv'.$$

The extracted $\partial M_i/\partial v$ and $M_i$ versus $v$ are shown in Extended Data Fig. 6. We extract the uncertainty (95% confidence interval) of $\partial M_i/\partial v$ from fitting $\mu$ with $B_i$, and propagate through the integration to obtain the uncertainty in $M_i$.

Thermal activation gap analysis
To determine the activation gap, a temperature-dependent background was first removed from the raw resistance $R_{xx}$ of MATBG to avoid being affected by the linear $R_{xx} - T$ behaviour in MATBG 43. The background-removed quantity is denoted as $R_{xx}'$ and shown in Extended Data Fig. 3a, b. Thermal activation gap analysis was performed on the basis of the Arrhenius formula $R_{xx}' \propto \exp\left(-\Delta/(2k_B T)\right)$, where $\Delta$ is the gap size. By fitting the gaps as a function of the in-plane magnetic field $B_i$ to $\Delta = g_B B_i$, we find effective transport $g_B$ factors of -1.31 for the $v = +2$ state and -0.57 for the $v = +1$ state, as shown in Extended Data Fig. 3c.

Data availability
The data that support the current study are available from the corresponding authors upon reasonable and well motivated request.

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Author contributions
J.M.P. and Y.C. fabricated the samples and performed transport measurements and numerical simulations. K.W. and T.T. provided hBN samples. J.M.P., Y.C. and P.J.-H. performed data analysis, discussed the results and wrote the manuscript with input from all co-authors.
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Extended Data Fig. 1 | Superconductivity and Landau fan diagram of MATBG. a, Superconducting curves for the $\nu = -2 - \delta$ and $+2 + \delta$ domes of MATBG. The maximum $T_c \approx 2.7$ K is determined from the 50% normal resistance of the $\nu = -2 - \delta$ curve. b, Landau fan diagram of MATBG at 1 K. The CNP shows the main sequence $\nu_{\text{LL}} = \pm 4, \pm 8, \ldots$ and the broken-symmetry states $\nu_{\text{LL}} = -1, \pm 2, \pm 3$. There are fans from $\nu = \pm 2$, where the sequence $\nu_{\text{LL}} = \pm 2, \pm 4, \pm 6$ and $\nu_{\text{LL}} = -2$ are seen, respectively. We also find transport evidence of a correlated Chern gap with Chern number $C = 3$ from $\nu = +1$. 
Extended Data Fig. 2 | In-plane magnetic-field dependence of $\mu$ and $R_{xx}$.

a–c. The in-plane magnetic-field dependence of $\mu$ and $R_{xx}$ is shown at $\nu = -2$ (a), $\nu = -1$ (b) and $\nu = +3$ (c). a, b. The hole-doped side features are qualitatively similar to those on the electron-doped side, but weaker. We note that the ‘dip’ in $\mu$ on the electron-doped side is analogous to the ‘peak’ on the hole-doped side. For $\nu = -1$, the peak in $R_{xx}$ and the ‘peak’ feature in $\mu$ enhance under $B_{\parallel}$. For $\nu = -2$, the peak in $R_{xx}$ weakens upon applying $B_{\parallel}$, whereas the feature in $\mu$ does not exhibit a noticeable change. c. For $\nu = +3$, the trend in $\mu$ is similar to that for $\nu = +1$, that is, the ‘dip’ feature is enhanced under $B_{\parallel}$, although the dependence is generally weaker. There is no noticeable peak in $R_{xx}$ at $\nu = +3$. 

For $\nu = -1$, the peak in $R_{xx}$ and the ‘peak’ feature in $\mu$ enhance under $B_{\parallel}$. For $\nu = -2$, the peak in $R_{xx}$ weakens upon applying $B_{\parallel}$, whereas the feature in $\mu$ does not exhibit a noticeable change. For $\nu = +3$, the trend in $\mu$ is similar to that for $\nu = +1$, that is, the ‘dip’ feature is enhanced under $B_{\parallel}$, although the dependence is generally weaker. There is no noticeable peak in $R_{xx}$ at $\nu = +3$. 
Extended Data Fig. 3 | Thermal activation gap analysis and $g$ factors of the correlated states. 

**a, b.** Fitting of temperature-dependent resistance using the Arrhenius formula $R^*_\nu = \exp(-\Delta/(2k_B T))$ at $\nu = +1$ (a) and $+2$ (b) for in-plane magnetic fields of $B_\parallel = 0$–$11$ T. $R^*_\nu$ is the background-removed resistance of MATBG. 

**c.** $B_\parallel$ dependence of the thermal activation gap $\Delta$. The extracted $g$ factors are $-0.57$, $-1.31$ for the $\nu = +1$, $+2$ states, respectively.
Extended Data Fig. 4 | Overlay of inverse compressibility and superconducting dome, and full-range chemical potential data. a, Temperature dependence of inverse compressibility $\frac{d\mu}{dn}$ for $T = 2$–70 K at $B = 0$ T. Negative compressibility near $\nu = -1, +2$ persists up to $T \approx 20$ K. b, Comparison between $\frac{d\mu}{dn}$ and superconducting $T_c$ dome (red points; 20% normal-state resistance) near $\nu = -2 \pm 6$. The $T_c$ dome occurs near maximum $\frac{d\mu}{dn}$, which is unexpected within a weak-coupling BCS-type mechanism for the superconductivity. c, Same data as in Fig. 2d, but showing the chemical potential beyond $\nu = \pm 4$. 
Extended Data Fig. 5 | Perpendicular magnetic-field dependence of Landau level and Chern gaps.

a–e, Gap extraction from the chemical potential curves at $B_\perp = 0$–6 T. f–g, Magnetic-field dependence of Landau level gaps (f) and correlated Chern gaps (g). Whereas the $\nu_{LL} = \pm 4$, 0 Landau level gaps have an increasing trend with $B_\perp$, the $\nu_{LL} = \pm 2$ gaps show a relatively weak dependence. Similarly, the three correlated Chern gaps also exhibit weak dependence on $B_\perp$. The reason why the Chern gap at $\nu = 2 + 2\phi/\phi_0$ is larger than the other two might be the difference in their magnetic ground states, with contributions from both orbital and spin degrees of freedom.
Extended Data Fig. 6 | In-plane magnetization of MATBG. a, $\frac{dM_{\parallel}}{dv}$ versus $v$. Peaks are visible near $\nu_L = \pm 1$. $T = 4$ K. b, Magnetization $M_{\parallel}$ from integration of the curve in a. $M_{\parallel}$ persists near all filling factors $\nu = \pm 1, \pm 2, \pm 3$. The error bands correspond to a confidence level of 95%.