Charge order and broken rotational symmetry in magic-angle twisted bilayer graphene

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Bilayer graphene can be modified by rotating (twisting) one layer with respect to the other. The interlayer twist gives rise to a moiré superlattice that affects the electronic motion and alters the band structure1–4. Near a ‘magic angle’ of twist1–4, where the emergence of a flat band causes the charge carriers to slow down5, correlated electronic phases including Mott-like insulators and superconductors were recently discovered5–8 by using electronic transport. These measurements revealed an intriguing similarity between magic-angle twisted bilayer graphene and high-temperature superconductors, which spurred intensive research into the underlying physical mechanism5–8,14. Essential clues to this puzzle, such as the symmetry and spatial distribution of the spectral function, can be accessed through scanning tunnelling spectroscopy. Here we use scanning tunnelling microscopy and spectroscopy to visualize the local density of states and charge distribution in magic-angle twisted bilayer graphene. Doping the sample to partially fill the flat band, we observe a pseudogap phase accompanied by a global stripe charge order that breaks the rotational symmetry of the moiré superlattice. Both the pseudogap and the stripe charge order disappear when the band is either empty or full. The close resemblance to similar observations in high-temperature superconductors5–8 provides new evidence of a deeper link underlying the phenomenology of these systems.

In the limit of strong correlations, the Coulomb interaction slows down the electrons in a material or even localizes them in a Mott insulating phase, characterized by a spectral gap that opens at integer fillings. Doping a Mott insulator produces fascinating correlated quantum phases that result in phenomena such as the pseudogap and high-temperature superconductivity, with broken symmetries to accommodate the new order parameters. In the case of magic-angle twisted bilayer graphene (TBG), doping the Mott-like insulating states leads to correlation-driven superconducting phases at low temperatures6,10. Using scanning tunnelling microscopy (STM)/scanning tunnelling spectroscopy (STS) to study the electronic properties of magic-angle TBG at temperatures above the superconducting transition, we observe a correlation-driven pseudogap state characterized by partial gapping of the low-energy density of states (DOS) when the flat band is partially filled. This pseudogap state is accompanied by a local charge polarization with a quadrupole symmetry and a global charge-ordered stripe phase that breaks the rotational symmetry of the moiré superstructure.

The STM topography of a TBG sample near the magic angle measured in a gated device geometry (Fig. 1a) is shown in Fig. 1b. The bright regions, which correspond to AA stacking where each top-layer atom is positioned directly above a bottom-layer atom, form a triangular moiré superlattice. The twist angle between the two layers, \( \theta \), can be estimated from the moiré period, \( L \), through the geometrical relation \( L \approx \frac{a}{2} \sin(\theta/2) \), where \( a = 0.246 \) nm is the graphene lattice constant1,2. Surrounding each bright region are six darker areas consisting of alternating AB/BA Bernal stacking. In the AB (BA) stacking each top-layer atom in the A (B) sublattice sits directly above a B (A) atom in the bottom layer, while top-layer B (A) atoms have no partner in the bottom layer. This is directly seen in the triangular structure of the atomic-resolution topography (Extended Data Fig. 1), where the triangles in the AB and BA regions show only one of the two sublattices. Earlier work on TBG revealed two van Hove singularities (VHS) in the local DOS that flanked the Dirac point2,4,22,23 with an energy separation of \( \Delta E \approx \hbar v_F K \Delta_\perp = -2 t_x \). Here \( \hbar \) is the reduced Planck constant, \( v_F \) is the Fermi velocity, \( K \Delta_\perp \Delta_\perp = 2 K \sin(\theta/2) \) is the magnitude of the wave vector at the Brillouin zone corner of the original graphene lattice, and \( t_x \) is the interlayer hopping that can vary from sample to sample depending on the fabrication technique or external pressure2. As the twist angle is reduced, the two VHS approach each other until, at the ‘magic angle’, \( \theta_m \approx 1.1^\circ \), they merge, producing an almost flat band with vanishingly small Fermi velocity3,4. Owing to the reduced kinetic energy, this narrow band is susceptible to interaction effects, so that when the Fermi energy \( E_F \) is brought within the band, interactions become dominant and the system can lower its energy through the formation of correlated electron phases.

We use STM topography together with STS to identify the magic angle and the flat-band condition in situ. To study the electronic structure of TBG near the magic angle, we measure the \( dI/dV \) spectrum \((I \) is the current, \( V \) the bias), which is proportional to the local DOS. The flat band in the DOS produces a peak in the \( dI/dV \) spectrum centred at the Dirac point (Fig. 1c). The width of this peak is narrowest when two VHS have merged and the band is full or empty (Fig. 1c), providing a practical criterion for identifying the magic angle. Using the peak width as a guide, we find the narrowest peak in sample regions with no heterostrain24, as identified by a perfectly triangular moiré pattern (Fig. 1b) (Methods). For the samples discussed here the magic angle is \( \theta_m \approx 1.07^\circ \) (\( L = \approx 13.2 \) nm), from which we extract \( t_x \approx 104 \) meV.

In order to separate the intrinsic band structure from correlation-induced band-reconstruction effects, we first show (Fig. 1c) the \( dI/dV \) spectra obtained in the AA region in the highly n-doped regime, for a gate voltage \( V_g = 55 \) V where the band is fully occupied, and in the highly p-doped regime \( V_g = -55 \) V where the band is empty. In both cases we observe a single spectral peak in the AA region, indicating the presence of the flat band. This peak is absent in the AB/BA regions (Fig. 1c inset), consistent with previous reports25,26. The single-peak structure is observed only very near the magic angle, but as the angle deviates from its magic value, the peak splits into two VHS that flank the Dirac point (Extended Data Fig. 2), consistent with the single-particle band structure theory for TBG3,4. It is worth noting that, while two similar VHS observed for non-magic twist angles are seen in all STS experiments, at the magic angle the DOS structure exhibits large sample-to-sample variations26–28. This reflects the fact that near the magic angle the band structure is very sensitive to lattice relaxation, interlayer coupling, strain and interaction strength29,30, which are influenced by sample preparation techniques and device geometry.

As we move the Fermi level into the flat band by adjusting the gate voltage, the single peak in the AA region broadens and splits, revealing a pseudogap feature (Fig. 1d), which suggests a band reconstruction associated with the emergence of a correlation-induced phase. In
As the Fermi level enters the flat band, we observe a spectral weight redistribution away from the Fermi energy and into the sidebands. This gives rise to a pseudogap feature flanked by two peaks, the relative height of which depends on doping, as shown in Fig. 2a. The two nearest dips near the same fillings—ν = 0, ±1/4, ±1/2, ±3/4, ±1—as those observed in the conductance measurements, correspond to 0, 1, 2, 3, 4 electrons (+) or holes (−) per moiré cell, respectively.

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**Fig. 2** | Doping dependence of $dI/dV$ spectra. a, The dependence on gate voltage (lower x axis) or filling fraction (upper x axis) of the $dI/dV$ intensity at the Fermi level ($V_g = E_F$) shows clear dips (vertical shaded bars are guides to the eye) at filling fractions $\nu = 0, \pm 1/4, \pm 1/2, \pm 3/4, \pm 1$. Here $+$ ($-$) correspond to the electron (hole) doped sectors, respectively. b, Colour map of gate-voltage-dependent (lower x axis) or carrier-density-dependent (upper x axis) $dI/dV$ spectra in the AA region, highlighting the doping-induced spectral shift between the LB and UB (arrowed). The vertical dashed line marks the charge neutrality point (taken at $V_g = −300$ mV, $I = 20$ pA). c, Gate-voltage dependence of $dI/dV$ spectra taken at the centre of an AA domain close to charge neutrality. The curves (offset for clarity) correspond to doping levels ranging from $−0.86 \times 10^{12}$ cm$^2$ (at $V_g = −12$ V) to $0.86 \times 10^{12}$ cm$^2$ (at $V_g = +12$ V). The spectral weight shift between the LB and the UB with doping is clearly seen. d, DMFT simulation of local DOS projected to the AA-centred local functions at different filling fractions, as discussed in the text. The data discussed in the main text were taken on sample 1. The data taken on another sample (sample 2) are shown in Extended Data Fig. 9.

**Fig. 3** | Spatial charge modulation in the correlated phase. a, STM topography in a 10 nm $\times$ 10 nm area centred on an AA region ($V_b = −200$ mV, $I = 20$ pA). The red circle labels the AA region; see d for the coloured arrow. b, $dI/dV$ map over the same area as a for the LB (left panel) and UB (right panel) at $V_g = 0$ V (10 nm $\times$ 10 nm, $V_b = −200$ mV, $I = 50$ pA). c, Map of the net charge obtained by the method described in the text. Red corresponds to electron doping and blue to hole doping. The four dashed lobes mark sectors of alternating electron (e) and hole (h) doping; see d for the coloured arrow. d, Spatial dependence of $dI/dV$ curves along the coloured arrow in a and c shows the spectral weight shift between the LB and the UB with position. e, Gate-voltage dependence of filling fraction (symbols) within the flat band extracted from Fig. 2c. The dotted line shows the gate dependence of the filling fraction, $\nu$, obtained from the gate voltage as described in the text. f, Position dependence of filling fraction from d along the path indicated by the coloured arrow in c. The filling fraction was obtained from the normalized area under the LB peak, as discussed in the text (see Methods).
a single peak similar to the experimental result, even though the intrinsic non-interacting DOS is split into two narrow peaks, because the Coulomb interaction broadens the peaks and smears out the double-peak structure. At fractional fillings (ν = 0, ± 1/4, ± 1/2), DMFT predicts a Mott gap in the local spectra, in which the occupied (empty) bands correspond to the lower (upper) Hubbard bands. The lower Hubbard band loses spectral weight with doping, while the upper band gains the weight, in qualitative agreement with the STM measurement.

To reveal the electronic structure, and the underlying symmetry of the emergent correlated state in the partially doped flat band, we study the spatial dependence of the topography (Fig. 3a) and spectroscopy (Fig. 3b–d, f) at the charge neutrality point, V_g = 0 V. In topography, the AA (bright) region shows up as a perfect disk, consistent with the symmetry of the moiré pattern which, in the limit of small twist angles and a continuum approximation, is C_6v. In contrast to the topography, the dI/dV maps taken in the same region at the energy corresponding to the LB (Fig. 3b left panel) and the UB (Fig. 3b right panel) form ellipses with major-axis orientations that are roughly orthogonal to each other and clearly break the C_6v symmetry. To elucidate the origin of the broken symmetry revealed by the dI/dV maps, we plot in Fig. 3d the position dependence of the spectra taken along the arrow shown in Fig. 3a. We note that, even though the back-gate voltage is fixed at V_g = 0 V, we nevertheless observe a clear spectral weight transfer from the LB, which is dominant in the bottom part of the AA region (red end of the arrow), to the UB, which dominates in the top part of the AA region (green end of the arrow). This is remarkably similar to the spectral weight transfer observed in the centre of the AA region as a function of doping (Fig. 2c). Because the back-gate voltage is set to a fixed value (V_g = 0 V), it is then natural to interpret the position dependence of the spectral weight transfer in terms of a spatial charge redistribution within the AA region. Using the same analysis as in Fig. 3e to calculate the local charge from the peak areas A_{LB} and A_{UB}, we obtain the position dependence of the local charge shown in Fig. 3e. Following this analysis, the high-intensity elliptical region for the UB map corresponds to hole doping, whereas the high-intensity elliptical region for the LB map corresponds to electron doping. In order to extract the spatial dependence of the local doping in the flat band, we subtract the intensity of the UB peak from the LB peak at every point on the map in Fig. 3b. Following this subtraction procedure, which directly visualizes the spatial charge modulation (Fig. 3c), we observe that within the AA region (red circle) the charge modulation exhibits a quadrupole geometry, consisting of four quadrants with alternating positive (blue) and negative (red) charge, ranging from about 1 hole to about 1.5 electrons per moiré cell. Similar charge polarization patterns are observed for ν = ±1/4 fillings (Extended Data Fig. 4). Importantly, as the Fermi level is outside the flat band (Extended Data Fig. 5), or for twist angles far from the magic value (Extended Data Fig. 6), the C_6v symmetry of the spectral maps is restored. This confirms that the broken symmetry is associated with a correlation-induced ordered phase, and rules out interpretations in terms of artefacts such as tip anisotropy, or strain effects (see Methods).

In addition to the local charge ordering observed in the scans of Fig. 3, large-area scans (Fig. 4) exhibit globally ordered charge stripes aligned with a crystallographic axis of the moiré superlattice, which break the initial C_6v symmetry of the moiré lattice and reduce it to C_2v. As seen in Fig. 4c, the global stripe order emerges from the alignment of the quadrupole charge lobes that are centred on the AA regions. The individual quadrupole lobes are not perfectly aligned with the stripe direction, but are slightly tilted (about 16°) with respect to it (Extended Data Fig. 7). Although we observed the charge symmetry breaking in all magic-angle samples studied here (Extended Data Figs. 8,9), we expect that there are many competing quantum states near the magic angle, and therefore slightly different sample preparations could possibly result in different correlated ground states.

Earlier reports have shown that the phenomenology of magic-angle TBG bears a tantalizing resemblance to that of high-temperature superconductors:5,6 the emergence of superconductivity in a doped Mott-insulator, the dome-like doping dependence of the superconducting critical temperature, the linear temperature dependence of the resistivity, and the relatively large ratio of critical temperature to the Fermi energy. Both the pseudogap phase and its stripe charge order revealed here closely resemble similar observations in the pnictides18,19 and copper oxides17,20, in which the high-temperature superconductivity is believed to be intimately connected to the pseudogap phase and stripe order is common15. The findings reported here add a new piece to the puzzle connecting TBG and high-temperature superconductors, and identify an important constraint on the nature of the correlated electronic states in these systems.

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in sample 1. The d
Data Fig. 5 the topography and d
in the partially filled flat band or of the band structure itself, we show in Extended
metry and charge modulation occurs in the partially filled flat band. To address the
Absence of broken symmetry in the full band.

Absence of broken symmetry at non-magic twist angles. In Extended Data Fig. 6a, we show an area where the twist angle (θ = 1.5°) is away from the magic angle. The moiré period here is ~9.4 nm, and the DOS shows two VHS peaks separated by 80 mV (Extended Data Fig. 6a inset). The dI/dV map at the energy of one of the VHS is given in Extended Data Fig. 6b, where the circular shape of the subtraction map in these two regions (panels b and c) shows that no stripe orientations are slightly different, indicating the presence of charge-order domains. Importantly, the observation of domains with different stripe orientations helps rule out interpretations in terms of artefacts such as tip anisotropy.

Charge order in a different sample. The charge-ordered phase that forms in the partially filled flat band in magic-angle TBG was observed repeatedly, both in different regions of the same sample 1 (Extended Data Fig. 8) and in another magic-angle sample (sample 2)(Extended Data Fig. 9). Both the local charge modulation (Extended Data Fig. 9c) and the global stripe charge order (Extended Data Fig. 9f) are observed in this sample.

Excluding possible artefacts. Here we use the following results to exclude possible artefact-induced charge-order effects. (1) With the same tip in the same area, we observe no broken symmetry in the full band, as shown in Extended Data Fig. 5. This excludes the possibility that the observed charge order is a result of local strain or an artefact arising from an anisotropic tip. (2) We observe no broken symmetry at non-magic twist angles (Extended Data Fig. 6), again ruling out artefacts such as strain or an anisotropic tip. (3) The energy shift in the LB and UB as a function of position can also help to rule out a tip effect. In Extended Data Fig. 10, we show an extension of the data in Fig. 3d taken out to a distance of 6.8 nm along the coloured arrow. On this length scale, an energy shift in both LB and UB is clearly seen. (4) In Extended Data Fig. 9, the shape of the high-intensity regions of the DOS maps measured in the LB and the UB are more complex than those in Fig. 3b. A distorted shape could be attributed to tip anisotropy, this explanation is inconsistent with the fact that the shape changes with bias voltage and with position in the sample, and with the fact that the energies of the UB and LB change with position along the sample.

DMFT simulations of doping-dependent LDOS for a flat band. To construct the tight-binding model, we followed refs 4,27 and approximate the Fourier transform of the interlayer tunnelling by $t(q) = \text{exp}(-\gamma|q|T)$ with parameters $\gamma = 1.066$ eV, $\alpha = 0.15$, $\gamma = 1.25$, $d = 3.34$ Å. The hopping within the graphene layers is $t = 2.73$ eV. For the DMFT calculation, the set of four localized correlated orbitals is derived as in ref. 29. While these orbitals are very localized and contain almost all the weight of the narrow band, their projection to the tight-binding bands breaks the particle–hole symmetry, so that the occupied (unoccupied) part of the higher-energy band at the $\Gamma$ point has large (small) projection to the localized set of orbitals. Consequently, the DMFT correlated spectra appear asymmetric at the charge-neutral point ($\nu = 0$), in disagreement with experiment. In the orbital set of ref. 29, we use equal phase for the projection to the top and the bottom layer of the bilayer graphene. We found that the phase difference between the localized orbitals on the top and the bottom layer controls this particle–hole asymmetry. Moreover, we found that the purely imaginary phase between the two layers forms an optimized projector for the DMFT method, which not only restores the particle–hole symmetry, but also increases the amount of overlap between the narrow band and the localized set of functions (from 95% to 99.9%). Hence, this modified set of localized orbitals was used here. Finally, the Coulomb interaction among the correlated orbitals is accounted for by the DMFT method, where we assumed the screening of the interaction by dielectric constant $\varepsilon = 5$, which leads to a Coulomb interaction strength of $U = 150$ meV between the local orbitals, as in ref. 29.

Data availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.

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Author contributions Y.J. and J.M. performed STM experiments. Y.J., J.M. and E.Y.A. performed data analysis and wrote the paper with input from all authors. K.H. provided calculations. X.L. fabricated the devices. K.W. and T.T. provided hexagonal boron nitride. E.Y.A. supervised the project.

Competing interests The authors declare no competing interests.

Additional information

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Extended Data Fig. 1 | Atomic-resolution STM of AB and BA regions in TBG near the magic angle. a, Large-area STM topography, 8.9 nm × 8.9 nm, taken at $V_b = -200$ mV, $I = 30$ pA. b, c, Magnified views of the green and blue boxed areas, respectively, in a. A schematic drawing of the graphene lattice is superposed on the STM topography to highlight the sublattice polarization due to the AB (BA) stacking.
Extended Data Fig. 2 | $dI/dV$ spectra and evolution of VHS with doping away from the magic angle in TBG. a, Back-gate ($V_g$) dependence of $dI/dV$ spectra for TBG at $\theta \approx 1.2^\circ$. Assignments of peaks to VHS 1 and VHS 2 are indicated. b, Zoomed-in image of the boxed area in a. c, Evolution of the $dI/dV$ spectra with back-gate ($V_g$) for a TBG at $\theta \approx 1.7^\circ$. 
Extended Data Fig. 3 | Estimate of the filling fraction from the area under the LDOS peak. a, $dI/dV$ curve for $\nu \approx -0.3$ ($V_g = -10$ V in Fig. 2, sample 1). The dashed line represents the background subtraction.

b, $dI/dV$ spectrum after background subtraction. Coloured areas are used to estimate the filling fraction as described in the text. $A_{LB}$ and $A_{UB}$ are the areas under the lower and upper bands, respectively.
Extended Data Fig. 4 | Charge polarization within moiré cells of TBG at ±1/4 filling. a, b, dI/dV curves and maps at +1/4 filling (a) and −1/4 filling (b), taken at $V_b = -200$ mV, $I = 50$ pA. The left two panels show the dI/dV curves at +1/4 filling (upper panel) and −1/4 filling (lower panel), the centre panels show the dI/dV maps at the LB energy in dI/dV curves (−30 mV for +1/4 filling and −17 mV for −1/4 filling), and the right panels show the dI/dV maps at the UB energy (11 mV for +1/4 filling and 33 mV for −1/4 filling).
Extended Data Fig. 5 | Absence of broken symmetry in the full flat band in sample 1. STM topography (a) and dI/dV map (b) of the same area discussed in the main text, measured at the energy corresponding to the centre of the flat band (dashed line in inset) in the highly n-doped regime ($V_g = +55$ V) corresponding to the fully filled flat band (taken at $V_b = 200$ mV, $I = 15$ pA). Inset, dI/dV spectra in AA/AB ($V_g = +55$ V).
Extended Data Fig. 6 | Absence of broken symmetry at non-magic twist angles. a, STM topography of TBG away from the magic angle (θ = 1.5°), centred on the AA region (shown by the dotted circle; data taken at \( V_b = -150 \text{ mV}, I = 20 \text{ pA} \)). Inset, \( \frac{dI}{dV} \) spectra in the AA/BA regime. b, \( \frac{dI}{dV} \) map of the same area as shown in a, at the energy of the left VHS (−29 mV), which is labelled by the dashed line in the inset in a (\( V_b = -150 \text{ mV}, I = 50 \text{ pA} \)).
Extended Data Fig. 7 | Relative orientation of the negative charge lobes and the charge stripe direction. a, Charge order extracted from a large area in sample 1, showing stripe charge orientation along a crystallographic axis of the moiré lattice (same as Fig. 4c; see Fig. 4c legend for details). b, The relative orientation angle between the charge quadrupole lobes (green lines) and the charge stripes (red lines), $16^\circ \pm 2^\circ$, is roughly constant within this region.
Extended Data Fig. 8 | Different charge-order orientations. a, $dl/dV$ maps at the energy of the LB (left panel) and the UB (right panel) for the same sample (sample 1) as that discussed in the main text, but in a different region in which the charge stripe is along a different direction. b, Charge modulation map obtained by subtracting the two intensity maps shown in a. The black arrow shows the direction of the electron lobe, and the green arrow marks the direction of the global charge stripe, which coincides with a crystallographic axis of the moiré pattern (see inset). Inset, large STM image of the moiré pattern. The scale bar is 10 nm. c, For comparison we show Fig. 3c, illustrating that the orientation (labelled by the black arrow) has changed compared to that in b. Inset, large STM image of the moiré pattern in c. The scale bar is 10 nm.
Extended Data Fig. 9 | Charge modulation in magic-angle TBG (sample 2). a, STM topography in a 10 nm × 10 nm area centred on the AA region (red circle; taken at $V_b = -100$ mV, $I = 40$ pA). b, $dl/dV$ map over the same area as a for the LB (left panel) and the UB (right panel) at $V_g = 0$ V ($V_b = -100$ mV, $I = 40$ pA). c, Map of net charge obtained by the method described in the main text. Red corresponds to electron doping and blue to hole doping. The four dashed lobes mark the sectors with alternating electron (e) and hole (h) doping. d, Spatial dependence of $dl/dV$ curves along the coloured arrows in a and c shows the shift of spectral weight between the LB and the UB with position. e, Position dependence of filling fraction from d along the path indicated by the arrow in c. The filling fraction was obtained from the relative area under the LB peak, as discussed in the text. f, Large-scale $dl/dV$ map (40 nm × 40 nm) of net charge, obtained by the method described in the text and in Fig. 3c.
Extended Data Fig. 10 | Energy shift in the LB and UB peak positions.

In the left panel (which is the same as Fig. 3d), the distance spanned, about 5 nm, is too small to distinguish the energy shift in the two bands (dashed lines). In the right panel, the data is extended out to \( r = 6.8 \) nm, where an energy shift in both LB and UB can be seen. See Methods for details.