Evidence for unconventional superconductivity in twisted bilayer graphene

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The emergence of superconductivity and correlated insulators in magic-angle twisted bilayer graphene (MATBG) has raised the intriguing possibility that its pairing mechanism is distinct from that of conventional superconductors3–4, as described by the Bardeen–Cooper–Schrieffer (BCS) theory. However, recent studies have shown that superconductivity persists even when Coulomb interactions are partially screened5,6. This suggests that pairing in MATBG might be conventional in nature and a consequence of the large density of states of its flat bands. Here we combine tunnelling and Andreev reflection spectroscopy with a scanning tunnelling microscope to observe several key experimental signatures of unconventional superconductivity in MATBG. We show that the tunnelling spectra below the transition temperature $T_c$ are inconsistent with those of a conventional $s$-wave superconductor, but rather resemble those of a nodal $s$-wave superconductor with an anisotropic pairing mechanism. We observe a large discrepancy between the tunnelling gap $\Delta_t$, which far exceeds the mean-field BCS ratio (with $2\Delta_t/k_B T_c \sim 25$), and the gap $\Delta_{AR}$ extracted from Andreev reflection spectroscopy ($2\Delta_{AR}/k_B T_c \sim 6$). The tunnelling gap persists even when superconductivity is suppressed, indicating its emergence from a pseudogap phase. Moreover, the pseudogap and superconductivity are both absent when MATBG is aligned with hexagonal boron nitride. These findings and other observations reported here provide a preponderance of evidence for a non-BCS mechanism for superconductivity in MATBG.

Tunnelling measurements of the quasiparticle density of states (DOS), the energy gap and electron–phonon coupling in conventional superconductors have provided key experimental evidence for the BCS theory of superconductivity7. Similar measurements on correlated superconductors, most notably using scanning tunnelling microscopy (STM) and angle-resolved photoemission spectroscopy, have shown their properties to be qualitatively different from those of BCS superconductors8,9. For the high-$T_c$ cuprate superconductors, whereas tunnelling spectra in the overdoped regime can be captured by the DOS of a BCS-like model with a $d$-wave order parameter, the yet-to-be-understood pseudogap phenomenon at reduced doping causes the spectroscopic properties of the cuprates to strongly deviate from this picture8.

Superconductivity has been observed at remarkably low carrier densities at partial fillings of the flat bands of MATBG10–13. Although these qualities suggest an unconventional pairing mechanism, conclusive evidence for any mechanism beyond the BCS paradigm is absent. We use density-tuned scanning tunnelling and point-contact spectroscopy (DT-STS and DT-PCS) to show that the superconducting phase of MATBG, specifically when hole-doping its flat valence band, shares a remarkable number of features with unconventional superconductors. Our experiments show a V-shaped gap at low temperatures and an unusual pseudogap precursor phase at higher temperatures and magnetic fields from which phase-coherent superconductivity emerges. The low-energy region of the V-shaped gap supports an anisotropic pairing mechanism with nodes in the superconducting gap function, as anticipated by some theoretical studies14–17. The pseudogap state may signify pairing without phase coherence or a secondary phase forming above $T_c$ and $\Delta_{AR}$. Both the pseudogap and superconductivity are absent when MATBG is commensurately aligned with the hexagonal boron nitride (hBN) substrate, suggesting that the structural characteristics and/or the $C_2T$ symmetry of unaligned MATBG is required for stabilizing these ground states. Although we cannot rule out a phonon-based pairing mechanism18–20, our results provide key constraints for an accurate theory of superconductivity in MATBG.

We performed our experiments in a home-built dilution-refrigerator STM21 instrument on devices sketched in Fig. 1a. MATBG, biased at the sample voltage $V_s$, rests on hBN/SiO$_2$/Si, while a gate voltage $V_g$ applied to Si tunes the carrier density (see Methods). Figure 1b shows a topographic image22–24 of unaligned MATBG/hBN, while Fig. 1c shows DT-STS...
Fig. 1 | STS of the tunnelling gap of superconducting MATBG. a, The experimental set-up, showing an STM tip (from which the tunnelling current is measured). MATBG, biased at \( V_g \), sits atop hBN/SiO\(_2\)/Si, while \( V_s \) is applied to Si to tune the carrier density. b, STM topographic image of MATBG. c, Tunnelling \( dI/dV(V_g, V_s) \) taken at the centre of an AA site in device A (1.13°, 0.4% strain) shows the conductive and valence flat bands pinned to \( E_F \). The red dashed-line box highlights a set of gaps in the valence flat band. d, Higher resolution \( dI/dV(V_g, V_s) \) acquired at 250 mK at the centre of an AA site in device A (see Supplementary Information for AB/BA data). Figure 1c shows that the conductive (valence) flat band is pinned to the Fermi energy \( (E_F; \ V_g = 0 \text{ V}) \) when \( V_s \) tunes \( E_F \) above (below) the charge neutrality point \( \text{(CNP;} \ V_{\text{CNP}} = 3.7 \text{ V}) \), while the valence (conduction) flat band onsets at \( V_s \) \(-20 \text{ mV} \) \((V_g > 20 \text{ mV}) \) and displays significant energy broadening due to charge fluctuations\(^{25} \). At millikelvin temperatures, we observe features in DT-STS attributed to a cascade of transitions at partial band fillings\(^{20,21} \), but they appear weaker and broader in energy than those observed at higher temperatures \((T > 4 \text{ K}) \), which may be related to high-entropy isospin fluctuations\(^{24,25} \).

**Distinguishing nodal superconductivity**

DT-STS shows several gapped phases (Fig. 1c) starting with band insulators \( V_s = \pm 4 \text{ V} \), where the electron filling per moiré unit cell relative to the CNP. Here we focus on partial fillings of the valence flat band near \( -3 < V < -2 \text{ mV} \), where transport studies\(^{1–3} \) report superconductivity in MATBG (Fig. 1c, red box). Figure 1d, f shows tunnelling spectra \( dI/dV(V_g, V_s) \) from two devices (device A as in Fig. 1c and device B), which display a gap at \( V = -2 \text{ mV} \) that opens and closes at \( E_F \) with decreasing \( V_g \), followed by the opening of a new gap that persists in the range \(-3 < V < -2 \text{ mV} \). The density dependence of these gaps is highlighted by \( dI/dV(V_g = 0 \text{ V}) \) as a function of \( V_s \) shown in Fig. 1d, f. We observe a clear transition between the two gapped phases, consistent with the phase diagram of MATBG from transport studies\(^{1–3,5} \) in which a correlated insulator at \( V = -2 \text{ mV} \) transitions into a superconductor that persists for \(-3 < V < -2 \text{ mV} \). In Fig. 1e, g, we plot tunnelling gaps for the \( V = -2 \text{ mV} \) correlated insulator (red curves) and the \(-3 < V < -2 \text{ mV} \) superconductor (blue curves) measured in each device. The \(-3 < V < -2 \text{ mV} \) tunnelling gap is significantly larger than \( k_B T \) observed in transport experiments\(^{1–3} \) and is an order of magnitude larger than an in-plane tunnelling gap in a MATBG p–n junction\(^{26} \) (presumably, the lateral p–n junction probes only the edge of the superconducting dome adjacent to the correlated insulator, instead of optimal doping, due to the junction’s doping gradient). Before examining the shapes of the tunnelling spectra further, we discuss our method for distinguishing between gapped insulating and superconducting phases by complementing DT-STS with PCS measurements.

As both correlated insulators and superconductors show suppressions in \( dI/dV(V_g = 0 \text{ V}) \), we require complementary information that distinguishes these two phases. We performed PCS by reducing the tip height above the sample until the tip makes point contact with the sample surface (sketched in Fig. 2a) and then measuring the two-terminal tip–sample conductance \( G(V_g, V_s) \) (see Methods; see Supplementary Information for discussion of possible tip-induced pressure and strain during PCS). This measurement is particularly sensitive to the local region beneath the tip (see Supplementary Information).

The PCS zero-bias conductance \( G(V_g = 0 \text{ V}, V_s) \) (see Methods; see Supplementary Information for discussion of possible tip-induced pressure and strain during PCS). This measurement is particularly sensitive to the local region beneath the tip (see Supplementary Information). The PCS zero-bias conductance \( G(V_g = 0 \text{ V}, V_s) \) (see Methods; see Supplementary Information for discussion of possible tip-induced pressure and strain during PCS). This measurement is particularly sensitive to the local region beneath the tip (see Supplementary Information).
superconductivity in this doping range. More direct evidence for superconductivity is revealed by the voltage-bias dependence of PCS $G(V_g)$ in Fig. 2c, d. These spectra are indicative of Andreev reflection\cite{22,23}, where incoming electrons from the metallic tip are reflected as holes while Cooper pairs propagate into the superconducting sample (Fig. 2a). This results in enhanced conductance at low biases and ‘excess current’ when the sample is superconducting. Signatures of Andreev reflection in PCS $G(V_g)$ (black boxes in Fig. 2e, f) are limited to fillings $-3 < v < -2$, magnetic fields $B < B_c$, 50 mT and temperatures $T < T_c$ $1.2$ K, all of which are consistent with transport measurements\cite{13}. A side-by-side comparison of STS and PCS (Fig. 2g) at the same sample location shows how PCS can clearly distinguish tunnelling gaps associated with superconductivity from those associated with insulators. Despite the presence of many correlation-driven gaps at $E_F$ in STS, only the filling range $-3 < v < -2$ shows both a V-shaped gap in STS and a zero-bias conductance peak in PCS.

**Two distinct energy scales and the pseudogap**

Both STS and PCS provide complementary evidence for an anisotropic pairing mechanism of superconductivity in MATBG. Moreover, these measurements establish two distinct energy scales. Low-energy STS spectra (Fig. 3a) are clearly incompatible with an isotropic $s$-wave pairing symmetry, and the best fits to such a model require introducing unphysically large quasiparticle broadening (equivalent to an electron temperature above 2 K; for comparison, see Supplementary Information for STS on superconducting Al). Often STS spectra on MATBG have a finite conductance at zero energy, but V-shaped spectra with zero conductance at zero bias have also been observed (Fig. 1g). These STS spectra resemble the quasiparticle DOS of a nodal superconductor, as for higher-angular-momentum (for example, $p$- or $d$-wave) pairing with an anisotropic gap function (Fig. 3b shows this fit for device A, $V_s = -25.8$ V—see the Supplementary Information for fits at other $V_s$).
Although the nodal fit describes this spectrum well, one should be cautious about this interpretation given the similar appearance of this gap to that of the pseudogap above $T_c$ and $B_c$ described below. Nevertheless, we extract an energy scale of $\Delta_{a} = 0.9\,\text{meV}$ from this fit, which roughly corresponds to half the separation of the shoulders in the spectrum. Similarly, the Andreev reflection spectra in PCS resemble predictions from the Blonder–Tinkham–Klapwijk (BTK) model\(^{29}\) using a nodal superconducting gap function (Fig. 3c and Supplementary Information). However, a BTK-model fit yields an energy scale $\Delta_{b} = 0.3\,\text{meV}$ (device A; $V_g = -22.8\,\text{V}$), $3 - 5$ times smaller than $\Delta_a$. For $T = 1.2\,\text{K}$ (measured through PCS), the observed ratio $2\Delta_b/k_B T_c = 25$ (device A; $V_g = -22.8\,\text{V}$) is significantly higher than the expected ratio for a tunnelling gap of a BCS superconductor ($2\Delta_b/k_B T_c = 3.53$). The Andreev energy-scale ratio $2\Delta_a/k_B T_c = 6$ also appears to be higher than the BCS ratio. As noted above, Andreev reflection disappears when phase-coherent superconductivity is absent, with both $\Delta_a$ and the Andreev excess current vanishing above $T_c$ and $B_c$ (Figs. 2d and 3d). In contrast, the STS gap $\Delta_a$ persists when phase-coherent superconductivity vanishes above $T_c$ (see Fig. 3e and Supplementary Information) and well above $B_c$ (Fig. 4).

A similar dichotomy between the energy scales describing tunnelling and Andreev reflection has been documented for the undoped cuprate superconductors\(^{37}\), where Andreev reflection also tracks the onset of phase coherence at $T_{c}$ while the tunnelling gap persists above $T_{c}$, as we observe in MATBG (see high-temperature data in the Supplementary Information; see also ref. \(^{23}\)). Compared to studies\(^{38,39}\) that examine the relationship between the pseudogap and superconductivity in the cuprates, in MATBG, we have the advantage that application of a relatively weak magnetic field in the hole-doped regime. ($V_g$ is the Landau-level filling factor.) Near $-3 < v < -2$, we observe an unconventional superconducting phase at low magnetic fields, which transitions into a pervasive pseudogap regime at high magnetic fields. QH, quantum Hall.
sharp pseudogap in the absence of phase-coherent superconductivity occurs when the van Hove singularity associated with the valence band overlaps with $E_g$. In this situation, the gain in the exchange energy may favour the formation of an isospin (spin/valley)-polarized/coherent ground state (or some other ordered state), which may be responsible for the pseudogap with sharp side peaks shown in Fig. 4a. However, given the remarkable resemblance between the shapes of the STS gaps in the pseudogap and superconducting phases, it is also possible that such a gap is driven by the formation of incoherent pairs for $B > B_c$ and $T > T_c$ (ref. 38). Regardless of the origin, the correlations responsible for the pseudogap are clearly compatible with the onset of phase-coherent superconductivity.

### Quenching pairing and pseudogap with hBN

Further insight into superconductivity and the pseudogap phase in MATBG is provided by studying MATBG aligned with hBN. Anodically, transport experiments do not report superconductivity in MATBG samples that are presumed to be well aligned with hBN (refs. 31,32). In examining the role of hBN alignment, STM studies are particularly advantageous, as they can directly visualize and distinguish the graphene–graphene (G–G) and graphene–hBN (G–hBN) moiré features. Figure 5b shows a set of representative topographic images of device C, taken at different $V_s$ and $V_g$, to disentangle the different structural roles of the two moiré patterns (see Supplementary Information). Surprisingly, these images show perfect alignment between the AA sites of the G–G moiré and the carbon–boron regions of the G–hBN moiré. This suggests a propensity for MATBG aligned to hBN to undergo a moiré-scale incommensurate–commensurate transition when the two moiré length scales are similar. In the schematic in Fig. 5a, we label these substrate-modified AA sites as AAb sites to reflect this alignment configuration. Likewise, the AB/BA sites of MATBG are made inequivalent by the hBN, forming AAb (BAa) regions where atoms in the top (bottom) graphene sheet are in register with atoms in the top hBN layer. This incommensurate–commensurate transition contrasts with the formation of a super-superlattice due to a long-wavelength interference between the two moiré patterns.

**DT-STS and DT-PCS on non-superconducting MATBG aligned to hBN.**

*Fig. 5.* a, STM topographic image of MATBG that is perfectly commensurate with the underlying hBN substrate. Atomicistic schematics show the stacking configurations of carbon, boron and nitrogen for different regions of the moiré pattern. b, STM topographic images of MATBG aligned to hBN for different values of $V_s$ and $V_g$, highlighting the graphene (G–G) moiré pattern and the graphene–hBN (G–hBN) moiré pattern. c, Side-by-side comparison of tunnelling $dI/dV(V, V_g)$ into an AAb site and point-contact $G(V, V_g)$ for device C (1.08° G–G twist angle, 0.1% G–G interlayer strain, 0.5 ± 0.1° G–hBN twist angle), which uses a graphite gate instead of a silicon gate. No signatures of a superconducting gap or a pseudogap or of Andreev reflection can be seen in either measurement. d, $dI/dV(V_g)$ spectra from c, offset by 15 mV (left) and 20 mV (right) for clarity. e, Tunnelling $dI/dV(V_s)$ and PCS $G(V_s)$ line cuts from c for $V_g = 0$ V. See Supplementary Information for tunnelling and spectroscopy parameters.
Discussion
Cumulatively, our findings provide substantial evidence that pairing in MATBG is unconventional and distinct from that of a BCS mechanism. STS does not show an isotropic gap with a size consistent with that expected from a $T_c \sim 1.2$ K x-wave BCS superconductor, but shows a V-shaped DOS consistent with that of a nodal superconductor, where the details of the spectra vary with twist angle and strain (see Supplementary Information). The PCS measurements corroborate this picture and additionally show an unusual linear suppression of the Andreev excess current approaching $T_r$ (Fig. 3d). This behaviour is similar to that reported in other unconventional superconductors$^{36,41}$ and has been suggested to be related to pair-breaking effects due to inelastic scattering from bosonic modes. There are many candidates for bosonic modes in MATBG, ranging from phonons to more exotic collective isospin fluctuations$^{42}$; however, a key ingredient for this scenario is the presence of a sign-changing order parameter, which makes scattering from such modes pair breaking$^{43}$. As an aside, if pairing is spin-triplet in nature, the ratio of the enhanced conductance near zero bias to the background conductance in the Andreev spectra is incompatible with an equal-spin-pairing order parameter (see Supplementary Information). Moreover, like the underdoped cuprate superconductors$^{35}$, MATBG shows contrasting behaviour between the energy scales describing tunnelling and Andreev reflection. Without further experiments, it is difficult to distinguish between different explanations for this dichotomy (that is, a precursor broken-symmetry phase or preformed pairing without coherence$^{30}$). Overall, the experiments presented here provide clear constraints for constructing a model of the pairing mechanism in this novel electronic material that lies beyond the BCS paradigm.

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in Fig. 2d were acquired by heating the 3He–4He mixture to a pressure of the conductance around the Andreev peaks\(^48\). See the Supplementary Information for non-Ohmic contact, possibly between the graphene and the Ti/Au surface. This does not damage the graphene. Differential conductance \(G\) measurements were performed by moving the STM tip a few nanometres (relative to the tip height during tunnelling) into the MATBG surface. This does not damage the graphene. Differential conductance \(G(V_g, V_s)\) was then measured through lock-in detection of the a.c. current in response to a a.c. modulation \(V_{ac}\) added to \(V_s\). Initial tunnelling parameters for STS were chosen to avoid phonon-induced inelastic tunnelling\(^{44}\).

We used two experimental protocols for avoiding unwanted local gating from the tip\(^{45}\). First, we used an STM tip that had been freshly prepared (field emission, pulsing, poking) and calibrated on a cleaned single-crystal metal, paying particular attention to protecting the tip from polymer residue contamination that often lies on the surface of two-dimensional material devices. Second, we used an STM tip and metal crystal made of materials (for example, tungsten and copper) that are work-function-matched with graphene. Careful preparation of the tip and sample are essential because when polymer residue on the device’s surface attaches to the tip, spectroscopic features of the tunnel junction are compromised, and topographic images often show ‘drag patterns’ caused by the motion of a particle in the tunnel junction or by flexing of the tip apex\(^{46}-^{47}\). As these drag patterns may be misinterpreted as tip-induced strain effects, we provide evidence of our clean and stable tip–sample junctions in Supplementary Fig. 16, which shows two topographic images without a drag pattern that are essentially identical despite a three-orders-of-magnitude change in the junction resistance.

### PCS measurements

PCS measurements were performed by moving the STM tip a few nanometres (relative to the tip height during tunnelling) into the MATBG surface. This does not damage the graphene. Differential conductance \(G(V_g, V_s)\) was then measured through lock-in detection of the a.c. current in response to a a.c. modulation \(V_{ac}\) added to \(V_s\), while \(dG/dV(V_g, V_s)\) is simply the numerical derivative of the measured \(G(V_g, V_s)\). We note that the conductance \(G(V_g)\) appears to be slightly suppressed around zero bias in the metallic state of MATBG at millikelvin temperatures, but this suppression vanishes at \(T = 1.3\, {\text{K}}\). As this suppression is present at all \(V_g\) and at magnetic fields above \(B_s\), we conjecture that this is due to non-Ohmic effects, possibly between the graphene and the Ti/Au electrodes. When MATBG is superconducting, the finiteness of the critical current and the proximity effect may also contribute to the suppression of the conductance around the Andreev peaks\(^{46}\). See the Supplementary Information for more details on the PCS measurements. The data in Fig. 2b, c, e, f were acquired together, and the data in Fig. 2d, g, h were acquired together. Between these two sets of data, the tip was withdrawn from the surface, and then point contact was re-established in the same location. The temperature-dependent data in Fig. 2d were acquired by heating the \(^{3}\text{He}–^{4}\text{He}\) mixture to \(T = 1.3\, {\text{K}}\) and then measuring PCS as the dilution refrigerator was cooled. The temperatures in Fig. 2d were measured via a RuO\(_2\) thermometer in the STM head. The tip probably drifts relative to the sample during this measurement.

As Yankowitz et al.\(^2\) have shown that superconductivity in twisted bilayer graphene can be tuned with pressure, we examined the role of tip-induced pressure/strain during a PCS measurement. Supplementary Fig. 6 shows tip-height-dependent PCS, showing that the energy scale for Andreev reflection \(\Delta_{\text{AR}}\) is unchanged as the tip is pressed further into MATBG. This, along with the fact that the density range, \(\Delta\), and \(B_s\) of superconductivity in PCS match those of transport experiments, verifies the one-to-one correspondence of STS and PCS at the same location. See Supplementary Section D for further discussion.

### Sample preparation

Devices were fabricated using a ‘tear-and-stack’ method\(^{49}\) in which a single graphene sheet is torn in half by van der Waals interaction with hBN. The two halves are rotated relative to each other and stacked to form MATBG. As device B is device A from ref. \(^{23}\), a full description of the fabrication procedure can be found therein. To summarize, graphene and hBN are picked up with polyvinyl alcohol. Then, to flip the heterostructure upside down, the heterostructure is pressed against an intermediate structure consisting of polymethyl methacrylate/transparent tape/Sylgard 184, and the polyvinyl alcohol is dissolved via water injection. The heterostructure is then transferred to a SiO\(_2\)/Si chip with pre-patterned Ti/Au electrodes. Residual polymer is dissolved in dichloromethane, water, acetone and isopropanol alcohol. This chip is annealed in ultrahigh vacuum at 170 °C overnight and 400 °C for 2 h. Device A is prepared in a similar manner, except the polymethyl methacrylate is replaced with Elvacite 2550, and N-methyl-2-pyrrolidone is added as a solvent. For device C, the intermediate structure consists only of Sylgard 184 on a glass slide, and a graphite gate is added to the heterostructure.

### Data availability

The data that support the findings of this study are available at https://doi.org/10.25281zenodo.5722484.

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