



# Emergence of correlations in alternating twist quadrilayer graphene

G. William Burg<sup>1</sup>, Eslam Khalaf<sup>2,3</sup>, Yimeng Wang<sup>1</sup>, Kenji Watanabe<sup>4</sup>, Takashi Taniguchi<sup>5</sup> and Emanuel Tutuc<sup>1</sup>✉

**Alternating twist multilayer graphene (ATMG) has recently emerged as a family of moiré systems that share several fundamental properties with twisted bilayer graphene, and are expected to host similarly strong electron–electron interactions near the magic angle. Here, we study alternating twist quadrilayer graphene (ATQG) samples with twist angles of 1.96° and 1.52°, which are slightly removed from the magic angle of 1.68°. At the larger angle, we find signatures of correlated insulators only when the ATQG is hole doped, and no signatures of superconductivity, and for the smaller angle we find evidence of superconductivity, while signs of the correlated insulators weaken. Our results provide insight into the twist angle dependence of correlated phases in ATMG and shed light on the nature of correlations in the intermediate coupling regime at the edge of the magic angle range where dispersion and interaction are of the same order.**

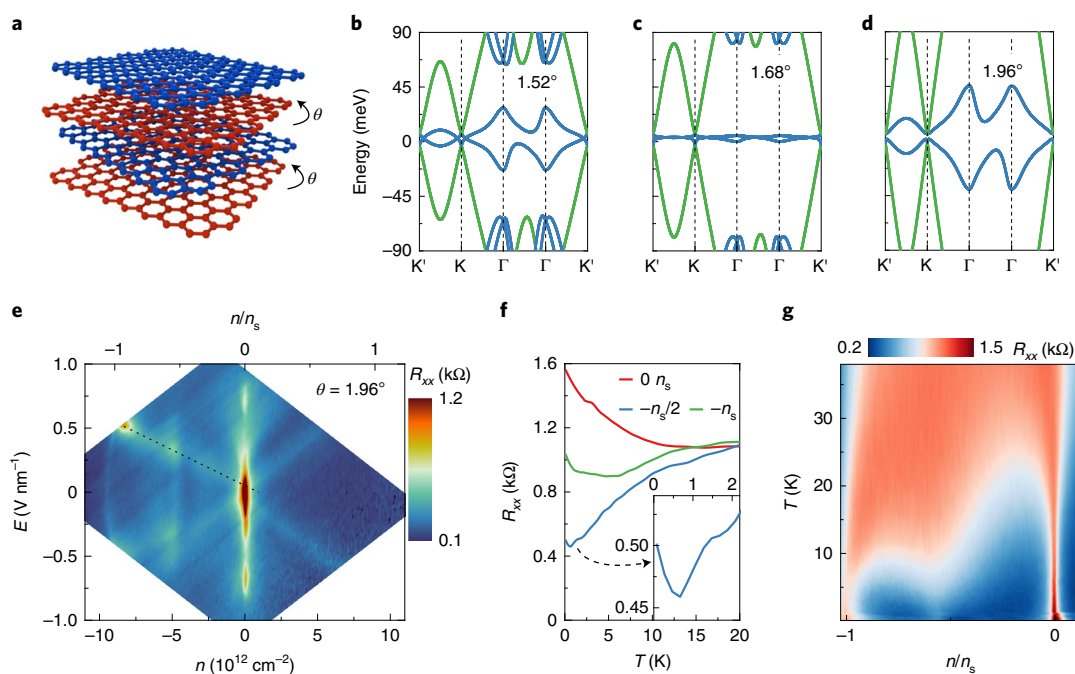
The emergence of two-dimensional van der Waals materials has enabled a new field of study in which moiré superlattices are engineered by stacking multiple van der Waals materials and applying a controlled twist between layers. For certain moiré heterostructures there are a series of ‘magic angles’ at which the lowest lying energy bands of the moiré band structure become extremely flat, enabling the Coulomb interaction strength to greatly exceed the kinetic energy of electrons in the band, favouring electron–electron correlations<sup>1</sup>. Starting with the discovery of correlated insulators and superconductivity within the flat bands of magic-angle twisted bilayer graphene (MATBG)<sup>2,3</sup>, substantial effort has been made to better understand the fundamental physics of moiré flat bands, and to expand the family of moiré heterostructures. Recent studies have revealed additional correlation-driven states in MATBG, such as ferromagnetism<sup>4,5</sup> and Chern insulators<sup>6–9</sup>, while others have focused on new types of twisted bilayers consisting of monolayer–bilayer graphene<sup>10–12</sup>, bilayer–bilayer graphene<sup>13–17</sup>, trilayer graphene on hexagonal boron nitride (h-BN)<sup>18,19</sup> and transition metal dichalcogenides<sup>20–22</sup>. These systems exhibit similar correlated phases, as in twisted bilayer graphene (TBG), but they differ in some fundamental aspects related to symmetry and band topology. In addition, they show no or rather weak signatures of superconductivity, in contrast to MATBG where robust superconductivity has been observed.

One family of moiré systems that share most of the TBG properties is alternating twist multilayer graphene (ATMG), a family of graphene moiré patterns consisting of  $m \geq 3$  graphene monolayers with a twist angle ( $\theta$ ) that alternates between  $+\theta$  and  $-\theta$  between each successive pair of layers<sup>23</sup>. The ATMG band structure can be exactly decomposed into  $m/2$  distinct TBG band structures for even  $m$ , each at an effective twist angle different than  $\theta$ , or  $(m-1)/2$  TBG band structures plus a Dirac cone for odd  $m$ . At the  $m$ -dependent magic angle, ATMG hosts flat bands, similar to those of MATBG, concurrently with dispersive bands, and for  $m \geq 5$  it can host multiple coexisting flat bands<sup>23,24</sup>. Realizing these systems requires overcoming the technical challenge of controlling several twist angles at once.

The simplest member of the ATMG family is alternating twist trilayer graphene (ATTG), consisting of three graphene layers with relative twist angles  $+\theta$  and  $-\theta$ . ATTG maps to a TBG with the interlayer coupling scaled by a factor of  $\sqrt{2}$  in addition to a dispersive Dirac band. Several recent studies have examined ATTG at the magic angle, revealing robust electric field-tunable superconductivity that persists across a wide range of densities<sup>25–29</sup>. However, the interplay between superconductivity and correlated insulators in ATMG, and their dependence on  $\theta$  remains unexplored. Here, we report the fabrication and characterization of alternating twist quadrilayer graphene (ATQG) close to, but slightly removed from, the magic angle of 1.68°. We focus on two samples with  $\theta = 1.96^\circ$  and  $1.52^\circ$ . We find that above the magic angle resistance maxima suggestive of correlated insulators appear at half-filling of the hole-doped side of the moiré flat bands, while superconductivity is absent. Below the magic angle we observe the opposite trend, wherein evidence of superconductivity is present, while the correlated insulators weaken. In both samples a small perpendicular magnetic field enhances the correlated insulating features, and quickly suppresses nascent superconductivity when present. In contrast, an in-plane magnetic field has little effect on the correlated insulators, and evidence of superconductivity persists in fields up to 7 T. A transverse electric field appears to enhance both the correlated insulators in the large-angle sample and the superconductivity in the small-angle sample.

Figure 1a shows a schematic of ATQG, consisting of four graphene monolayers with an alternating twist angle between each layer pair. Figure 1b–d shows the band structure of ATQG, taking into account relaxation<sup>23,24</sup>, at the two sample angles and at the magic angle of 1.68°. At the magic angle, the band structure can be separated into two TBG subsystems: one TBG at the first magic angle, giving rise to a pair of flat bands (blue lines in Fig. 1c), and another TBG away from the magic angle, which contributes two dispersive bands (green lines in Fig. 1c). At the two sample angles, the flat bands become more dispersive, but the overall character

<sup>1</sup>Microelectronics Research Center, Department of Electrical and Computer Engineering, The University of Texas at Austin, Austin, TX, USA. <sup>2</sup>Department of Physics, Harvard University, Cambridge, MA, USA. <sup>3</sup>Department of Physics, The University of Texas at Austin, Austin, TX, USA. <sup>4</sup>Research Center for Functional Materials, National Institute of Materials Science, Ibaraki, Japan. <sup>5</sup>International Center for Materials Nanoarchitectonics, National Institute of Materials Science, Ibaraki, Japan. ✉e-mail: [etutuc@mail.utexas.edu](mailto:etutuc@mail.utexas.edu)



**Fig. 1 | Transport and temperature dependence in ATQG.** **a**, Schematic of ATQG, with the second and fourth graphene monolayers (red) rotated by  $\theta$  relative to the first and third monolayers (blue). **b–d**, Band structure of ATQG at  $\theta=1.52^\circ$  (**b**),  $1.68^\circ$  (**c**) and  $1.96^\circ$  (**d**), displaying the flat bands of the TBG subsystem near the magic angle (blue) and the dispersive bands of the TBG subsystem away from the magic angle (green). **e**,  $R_{xx}$  versus  $n$  and  $E$  measured in the  $\theta=1.96^\circ$  sample at  $T=160$  mK. The top  $x$  axis shows  $n$  normalized to  $n_s$ . Correlated insulators appear when the valence band is half filled, in small ranges of  $E$  values. **f**,  $R_{xx}$  versus  $T$  measured at  $n=0$ ,  $-n_s/2$  and  $-n_s$  along the dotted line in panel **e**. The inset shows a close-up of the  $R_{xx}$  versus  $T$  at  $n=-n_s/2$  below 2 K. **g**,  $R_{xx}$  versus  $n/n_s$  and  $T$  along the dotted line in **e**.

of the band structure is similar. Figure 1e shows the longitudinal resistance ( $R_{xx}$ ) contour plot as a function of carrier density ( $n$ ) and transverse electrical field ( $E$ ) measured in the  $\theta=1.96^\circ$  sample at a temperature  $T=160$  mK.

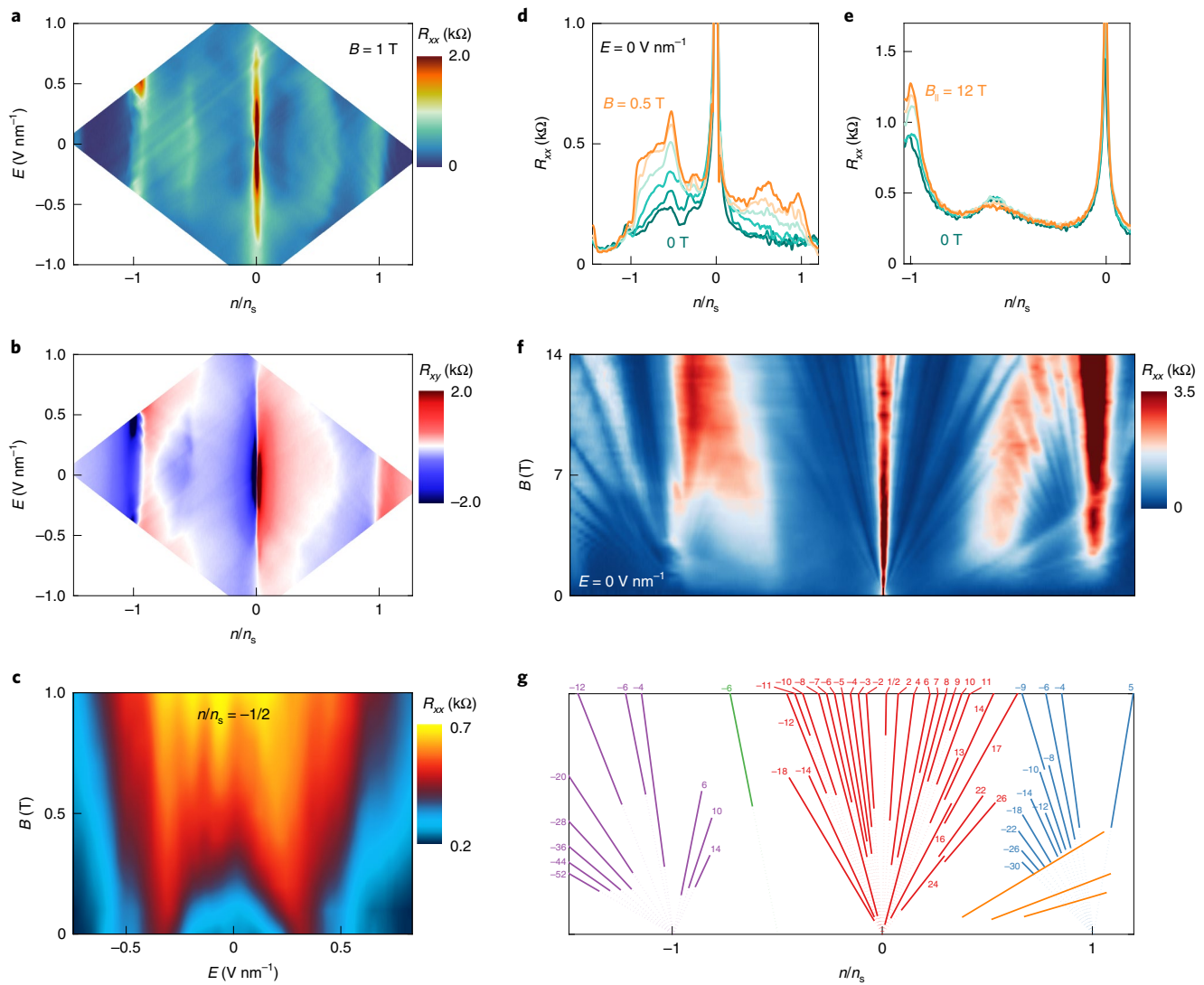
We observe  $R_{xx}$  maxima at charge neutrality and at  $n=-8.2 \times 10^{12} \text{ cm}^{-2}$  and  $-4.9 \times 10^{12} \text{ cm}^{-2}$ , which correspond to full-filling and half-filling of the flat valence band, respectively. While in the single-particle picture a gap at neutrality is not expected due to the semi-metallic dispersive bands, an interaction-generated gap in the flat band can be induced in the dispersive bands, which, unlike in ATTG, occurs because the two TBG subsystems are not distinguished by symmetry<sup>24</sup>. The full-filling density is denoted as  $n_s$  and is related to  $\theta$  by  $n_s \cong 8\theta^2/\sqrt{3}a^2$ , where  $\theta$  is in radians and  $a=2.46 \text{ \AA}$  is the graphene lattice constant. In Fig. 1e the upper  $x$  axis shows  $n$  normalized to  $n_s$ . For  $E$  near zero, the resistance at  $n=-n_s/2$  and  $-n_s$  is small due to residual conductance from the dispersive bands, but reaches a maximum as a gap opening is induced in the dispersive bands with increasing  $E$ <sup>24</sup>. At large  $E$ , no resistance maxima are observed at half-filling, consistent with calculations that show the two TBG subsystems hybridize and increase the bandwidth of the flat band. The appearance of correlated insulators exclusively in the valence band is in contrast with what is typically observed in TBG, in which correlated insulators are usually more robust when filling the conduction band; although there is some evidence that correlated insulators become stronger on the hole side at angles larger than the magic angle<sup>30,31</sup>.

Figure 1f shows the temperature dependence of the maximum  $R_{xx}$  at  $n=0$ ,  $-n_s/2$  and  $-n_s$ . For each dataset,  $R_{xx}$  shows an upward trend at low  $T$ , suggestive of insulating behaviour (see also Extended Data Fig. 1). Figure 1g shows  $R_{xx}$  versus  $n/n_s$  and  $T$  along the dotted line in Fig. 1e. At high  $T$ , only the band insulators are visible, while below 15 K the correlated insulator begins to appear and regions of low resistance develop on either side of it.

To further elucidate the nature of the correlated insulators, we examine the effect of an applied magnetic field. Figure 2a,b shows  $R_{xx}$  and the Hall resistance ( $R_{xy}$ ), respectively, as a function of  $n/n_s$  and  $E$  in a perpendicular field  $B=1$  T. At this field, the  $R_{xx}$  maxima of the correlated and band insulators extend to a wider range of  $E$  fields relative to  $B=0$  T and appear on the electron side as well. At  $n=-n_s/2$  and between  $E=\pm 0.5 \text{ V nm}^{-1}$ , the  $R_{xy}$  data show an abrupt change in value away from zero, a feature also observed in MATBG<sup>2,32</sup> and ATTG<sup>25</sup> and associated with a phase transition at the correlated insulator<sup>31,33</sup>. At larger  $E$ ,  $R_{xy}$  transitions through zero smoothly at  $n=-n_s/2$ , as expected when filling a single-particle band, suggesting a suppression of correlations and consistent with the disappearance of  $R_{xx}$  maxima. A similar behaviour is observed at  $n=+n_s/2$ , although in general signatures of the correlated insulator are weaker on the electron side.

An interesting feature of the  $B=1$  T data is the expansion of the half-filling resistance maxima to a larger range of  $E$  fields compared to the zero  $B$  field data (see also Extended Data Fig. 2). Figure 2c shows  $R_{xx}$  at  $n=-n_s/2$  as a function of  $E$  and  $B$ . For small  $B$ , the  $R_{xx}$  maxima are confined to small regions near  $|E|=0.3 \text{ V nm}^{-1}$ , but quickly expand and merge by  $B=0.5$  T. The applied  $B$  field induces gaps in the dispersive band through the formation of Landau levels. For small  $B$ , a combined  $E$  and  $B$  field is required to sufficiently open the band gap to reveal the correlated insulators, but above a critical  $B$  the correlated insulators are visible for all small  $E$ . These data support the scenario where correlated insulators are most likely to emerge if the flat bands are spectrally isolated from neighbouring bands<sup>13</sup>. Figure 2d shows  $R_{xx}$  versus  $n/n_s$  at  $E=0$ , for  $B$  fields ranging from 0 T to 0.5 T, demonstrating the marked increase in the correlated and full-filling insulator resistances for small  $B$  fields.

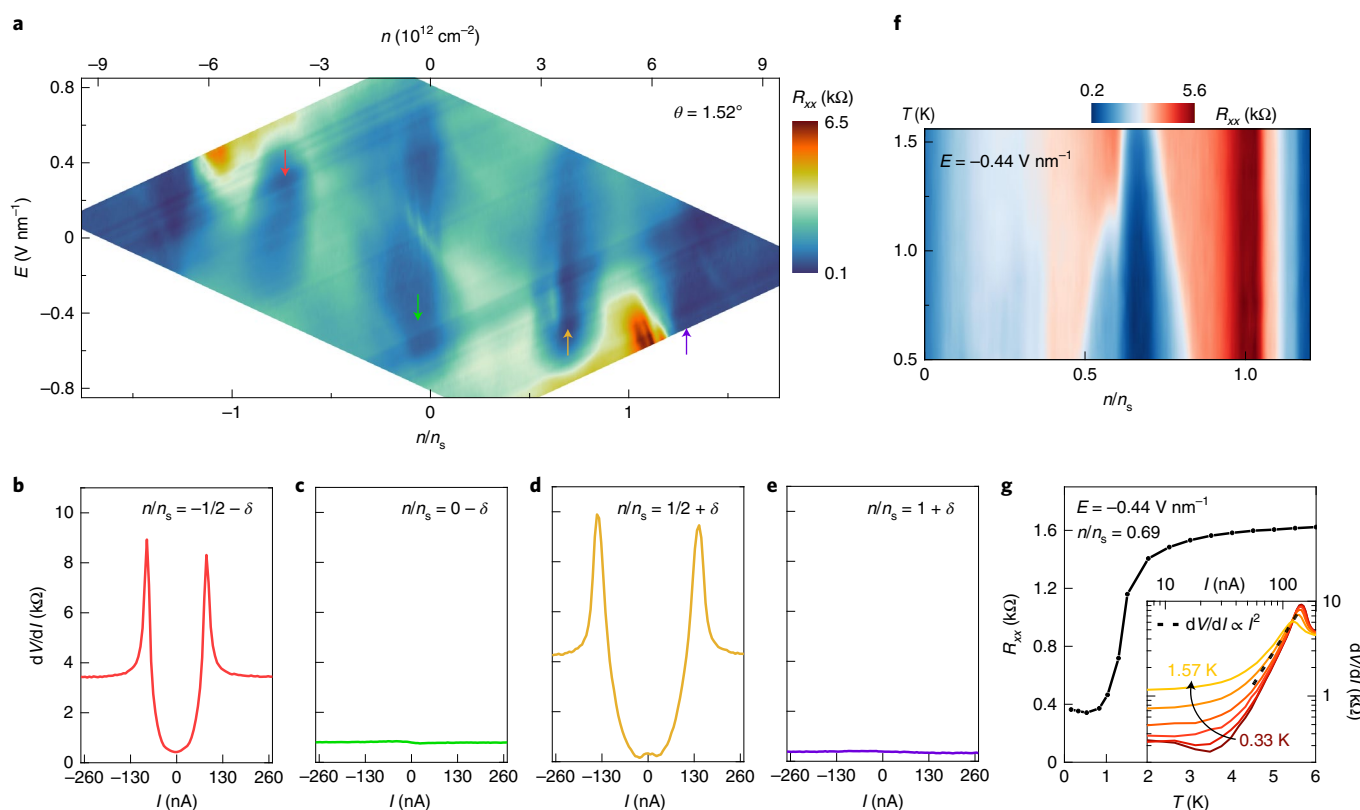
We also consider the effect of an in-plane magnetic field ( $B_{||}$ ). In the limit of zero layer thickness, an in-plane magnetic field only couples to the spin degree of freedom via the Zeeman effect, but because of the finite layer thickness of the electron system



**Fig. 2** |  $\theta = 1.96^\circ$  ATQG in a magnetic field. **a, b**,  $R_{xx}$  (**a**) and  $R_{xy}$  (**b**) versus  $n/n_s$  and  $E$  at  $B = 1$  T and  $T = 160$  mK. **c**,  $R_{xx}$  versus  $E$  and  $B$  along a vertical slice of **a** at  $n/n_s = -1/2$ . Resistance maxima that are initially localized to small regions of  $E$  expand and merge with increasing  $B$ . **d**,  $R_{xx}$  versus  $n/n_s$  at  $E = 0$  V nm $^{-1}$  as a function of  $B$  between 0 and 0.5 T, in increments of 0.1 T, showing correlated and full-filling insulators respond strongly to a small  $B$  field. **e**,  $R_{xx}$  versus  $n/n_s$  along the dotted line in Fig. 1e as a function of  $B_{||}$  between 0 and 12 T, in increments of 4 T. The correlated insulator shows little dependence on  $B_{||}$ . **f**,  $R_{xx}$  versus  $n/n_s$  and  $B$  at  $E = 0$  V nm $^{-1}$ , which shows oscillations associated with the formation of Landau levels originating from correlated and band insulators. **g**, Fits and labels of individual Landau levels in **f**.

considered here,  $B_{||}$  can also couple via an orbital effect<sup>24,34</sup>. The orbital coupling can be understood through a layer-dependent vector potential  $d(l\hat{z} \times \mathbf{B}_{||})$ , where  $l = 1, 2, 3, 4$  is the layer index,  $d = 3.4$  Å is the interlayer distance and  $\hat{z}$  is the unit vector perpendicular to the sample plane. The resulting orbital coupling is obtained by modifying the kinetic component of the Hamiltonian in layer  $l$  as  $h_l = v_F \sigma \cdot \nabla \rightarrow v_F \sigma \cdot \nabla - g_{\text{orb}} \mu_B \sigma \cdot (l\hat{z} \times \mathbf{B}_{||})$ , where  $g_{\text{orb}} = ev_F d / \mu_B \approx 6$  is the orbital  $g$ -factor, which is of the same order as the Zeeman splitting;  $e$  is the electron charge,  $v_F$  is the Fermi velocity of monolayer graphene and  $\mu_B$  is the Bohr magneton. For ATQG, a recent theoretical study has shown that the effective orbital coupling in the TBG subsystem hosting the flat band is smaller by a factor of 20 compared to TBG<sup>24</sup>, leading to a reduced sensitivity of correlated states to  $B_{||}$ . Figure 2e shows  $R_{xx}$  versus  $n/n_s$  at  $B_{||}$  from 0 T to 12 T. The correlated insulator at  $n = -n_s/2$  undergoes a slight broadening and reduction of the resistance peak up to  $B_{||} = 12$  T, which is consistent with the previous discussion and indicative of a spin-unpolarized state, as a Zeeman gap should increase with  $B_{||}$ .

Figure 2f shows  $R_{xx}$  as a function of  $n/n_s$  and  $B$  at a constant  $E = 0$  V nm $^{-1}$ , and exhibits quantum oscillations arising from the formation of Landau levels in a perpendicular magnetic field (Methods). We observe Landau fans emerging from  $n/n_s = -1, -1/2, 0, +1$ , with the filling factor ( $\nu$ ) of individual Landau levels labelled and the sub-bands indicated by different colours in Fig. 2g. The fan at  $n/n_s = 0$  shows single integer Landau levels, signalling a full lifting of the fourfold spin and valley degeneracy, and is indicative of high sample quality. Furthermore, the robust fans at  $n = \pm n_s$  suggest good angle uniformity. At  $n = -n_s/2$ , there is a single Landau level corresponding to  $\nu = -6$ , which is expected to occur for a band with Chern number  $-2$ , and a contribution of  $-4$  from the zeroth Landau level of the dispersive band. The emergence of a Chern  $-2$  band at  $n = -n_s/2$  at finite  $B$  field is also present in MATBG<sup>3,35</sup>. We observe an additional fan at positive  $n$  and smaller  $B$  fields with much lower slopes than the other fans, which we attribute to the partial filling of the dispersive band, as also seen in ATTG<sup>25,26</sup>.



**Fig. 3 | Evidence of superconductivity in  $\theta = 1.52^\circ$  ATQG.** **a**,  $R_{xx}$  versus  $n/n_s$  and  $E$  at  $T = 160$  mK. The top  $x$  axis shows the total density,  $n$ . Multiple areas of low resistance appear near charge neutrality, half-filling and full-filling of the flat bands. **b–e**,  $dV/dI$  versus  $I$  at  $n/n_s = 1/2 - \delta$  (**b**),  $0 - \delta$  (**c**),  $1/2 + \delta$  (**d**) and  $1 + \delta$  (**e**) at the  $n$  and  $E$  values labelled by the corresponding colour arrows in **a**. Regions near  $\pm n_s/2$  show critical current characteristic while other low-resistance regions do not. **f**,  $R_{xx}$  versus  $n$  and  $T$  along  $E = -0.44$  V nm $^{-1}$ , showing a dome of low resistance at  $n_s/2 + \delta$ . **g**,  $R_{xx}$  as a function of  $T$  at  $n/n_s = 0.69$  and  $E = -0.44$  V nm $^{-1}$ , illustrating the abrupt resistance decrease at 2 K. The inset shows  $dV/dI$  versus  $I$  on a logarithmic scale at different temperatures and the black dashed line shows a line of  $dV/dI \propto I^2$ , which is used to determine  $T_{\text{BTK}}$ .

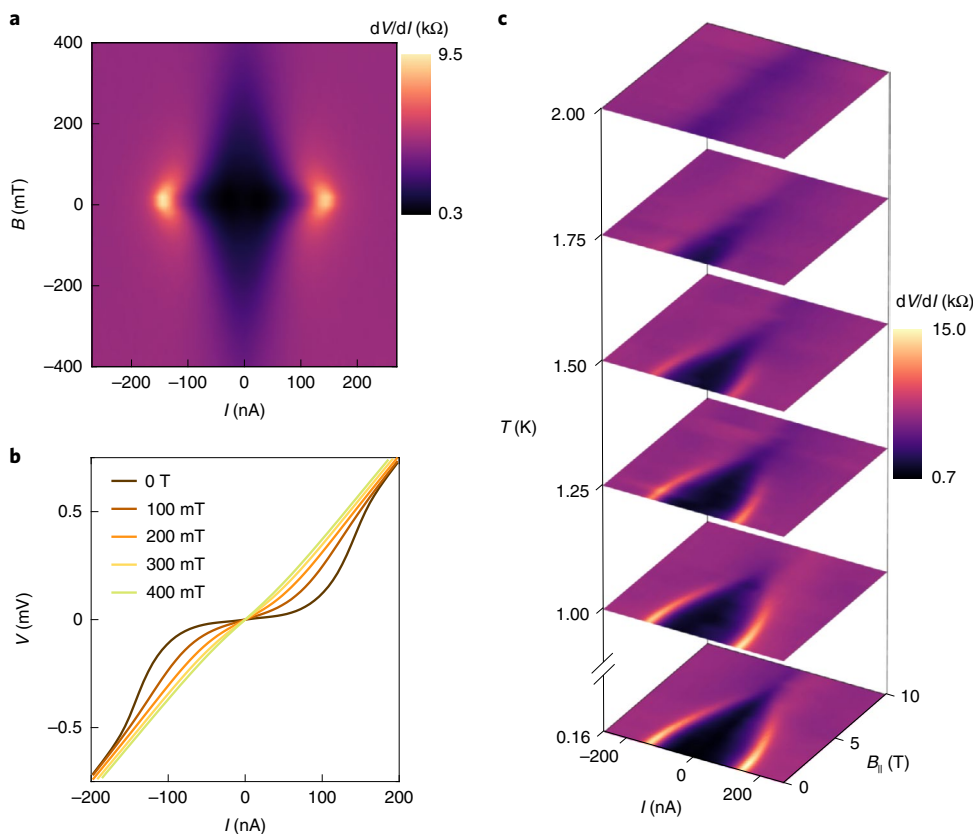
We now consider the ATQG sample with  $\theta = 1.52^\circ$ , which is closer to but smaller than the  $1.68^\circ$  magic angle. Figure 3a shows  $R_{xx}$  versus  $n/n_s$  and  $E$  at  $T = 160$  mK. The data show broad,  $E$ -dependent resistance maxima near  $\pm n_s/2$  and  $\pm n_s$ , as well as several regions of low resistance at  $(\pm n_s/2 + \delta)$ ,  $(\pm n_s + \delta)$  and  $0 - \delta$ , where  $\delta$  represents a small density value. It is instructive to examine these regions more closely, in particular near half-filling, where superconductivity most commonly occurs in TBG. Figure 3b–e shows the four-point differential resistance ( $dV/dI$ ) versus current ( $I$ ) at four representative points labelled by the coloured arrows in Fig. 3a. In the regions near charge neutrality (Fig. 3c) and full-filling (Fig. 3e) the differential resistance remains insensitive to the current. Conversely, near half-filling (Fig. 3b,d) the  $dV/dI$  data show a pronounced critical current behaviour, characterized by sharp peaks in  $dV/dI$  at the transition from a low to a high resistance state. The regions that exhibit the strongest critical current behaviour occur at finite  $E$  fields (Extended Data Fig. 3), consistent with similar observations in ATTG, and in agreement with calculations suggesting an increase in the critical temperature with  $E$  for twist angles close to but smaller than the magic angle<sup>24</sup>.

Figure 3f shows the temperature dependence of  $R_{xx}$  versus  $n/n_s$  at a constant  $E = -0.44$  V nm $^{-1}$ . At densities near  $n_s/2 + \delta$ , a clear dome of low resistance forms, while little temperature dependence is observed for all other  $n/n_s$ , including the low-resistance regions near charge neutrality and full-filling. Figure 3g shows the temperature dependence of  $R_{xx}$  at the optimal doping of Fig. 3f (orange arrow in Fig. 3a) over a larger temperature range. Around 2 K, the resistance drops sharply, but plateaus at a finite resistance of  $\sim 350 \Omega$  for temperatures

below 1 K. Using 50% of the normal-state resistance as the criterion for the critical temperature ( $T_c$ ) gives  $T_c \approx 1.34$  K. The inset of Fig. 3g shows  $dV/dI$  versus  $I$  on logarithmic scales. The temperature at which the  $dV/dI$  characteristic near the critical current is proportional to  $I^2$  (dashed line) gives the Berezinskii–Kosterlitz–Thouless transition temperature ( $T_{\text{BTK}}$ ) and, to best fit, the data yield  $T_{\text{BTK}} \approx 1.29$  K.

Overall, the  $dV/dI$  versus  $I$  and temperature-dependence data are strongly suggestive of nascent superconductivity (see also Extended Data Figs. 4 and 5). However, given the finite resistance at the lowest temperatures, it is best to exercise diligence. Joule heating has been proposed as a mechanism to produce non-linear current–voltage characteristics and strong temperature dependence, particularly in twisted double bilayer graphene<sup>17</sup>. In these cases, the data consistently show gradual transitions at a finite current leading to broad peaks or no peaks in the  $dV/dI$  versus  $I$ <sup>16,17,19,21,36</sup>, in contrast to the sharp  $dV/dI$  peaks observed here. A second normal-state mechanism that can produce abrupt transitions in the  $dV/dI$  is non-equilibrium transport<sup>37</sup>, where velocity saturation leads to an apparent critical current behaviour. However, these characteristics arise at very large current densities of the order of  $50 \mu\text{A} \mu\text{m}^{-1}$ , several orders of magnitude larger than the currents used here ( $\sim 100$  nA  $\mu\text{m}^{-1}$ ). Furthermore, a critical current is observed at all densities away from neutrality in the non-equilibrium regime, and not exclusively near fractional filling of the bands, as we observe (Fig. 3b–e).

A key test of a superconducting state is the response to a small perpendicular  $B$  field, which has been used in MATBG to identify states with finite resistance at the lowest temperatures<sup>5</sup>. The Joule



**Fig. 4 | Magnetic field response of nascent superconductivity.** **a**,  $dV/dI$  versus  $I$  and  $B$  at  $n/n_c = 0.69$  and  $E = -0.44 \text{ V nm}^{-1}$ . The critical current is quickly suppressed by a small  $B$  field. **b**,  $V$  versus  $I$  as a function of  $B$  between 0 and 400 mT, in 100 mT steps. **c**,  $dV/dI$  versus  $I$  and  $B_{||}$  at multiple temperatures. At the lowest temperature, the critical field is  $B_{||} \approx 7 \text{ T}$ . The critical current and field both shrink with increasing  $T$ , and are entirely absent at 2 K.

heating effect has not been considered in a magnetic field<sup>17</sup> and the critical current that arises from non-equilibrium transport was shown to be insensitive to a magnetic field up to 2 T (ref. <sup>37</sup>).

Figure 4a shows  $dV/dI$  as a function of  $I$  and  $B$  at  $T = 160 \text{ mK}$  at the same  $E$  and  $n$  values as in Fig. 3d,g. The critical current is strongly dependent on the applied  $B$  field, steadily decreasing with increasing field, and is fully extinct at  $\sim 350 \text{ mT}$ , consistent with a nascent superconducting state. Figure 4b shows the corresponding  $I$  versus  $V$  data at 100 mT intervals, demonstrating the marked reduction of the switching behaviour at the critical current as the characteristic trends towards linear with  $B$  field. The data for Fig. 4a,b give an approximate critical  $B$  field ( $B_c$ ) of 250 mT, which can be used to estimate the Ginzburg–Landau coherence length ( $\xi$ ) with  $B_c = [\Phi_0/(2\pi\xi)](1 - T/T_c)$ , where  $\Phi_0 = h/(2e)$  is the superconducting flux quantum and  $h$  is Planck’s constant. Using the given relation, we find  $\xi \approx 34 \text{ nm}$ . We do not observe Fraunhofer oscillations in the  $B$  field data, which are often seen in MATBG<sup>3,30,32</sup>, and occur due to phase-coherent transport through small normal regions, which arise from charge inhomogeneity, interspersed within the superconducting bulk. It is possible that in this sample only isolated pockets of the channel are superconducting and the normal regions separating them are too large to maintain phase coherence, which may also explain the non-zero minimum resistance.

In contrast to a perpendicular magnetic field, in-plane magnetic fields are expected to have little effect on superconducting states in all  $m$ -layer ATMG. Odd-layer ATMG is explicitly protected from pair breaking in an in-plane field due to its mirror and time-reversal symmetry<sup>35</sup>, and has been demonstrated experimentally in ATTG in which superconductivity survives substantially beyond the Pauli limit<sup>38</sup>. Even-layer ATMG lacks mirror symmetry and should respond

to  $B_{||}$  via orbital coupling in the same way as MATBG. The orbital effect in TBG is obtained by projecting the modified Hamiltonian onto the flat bands, which yields a momentum-dependent correction to the dispersion that has opposite sign between the two valleys, leading to pair breaking for intervalley superconductivity at a field that depends on both doping and field direction, but is roughly the same as the Pauli field<sup>24,39,40</sup>. However, this effect in ATQG is expected to be scaled down by a factor of 20 (ref. <sup>24</sup>). To better illustrate the reduced effect of an in-plane  $B$  field, we first calculate the theoretical Pauli limit ( $B_p$ ), assuming a weak coupling superconductor and a  $g$ -factor of 2, using  $B_p = 1.86 [T/K] T_c$ , which for our sample yields  $B_p \approx 2.5 \text{ T}$ . Figure 4c shows  $R_{xx}$  versus  $I$  and  $B_{||}$  at multiple temperatures and, indeed, the evidence of superconductivity persists up to a critical field of roughly 7 T at the lowest temperature, well beyond the expected Pauli limit and almost an order of magnitude larger than in MATBG<sup>40</sup>. With increasing temperature, the critical field and critical current both decrease, and at  $T = 2 \text{ K}$  no signs of superconductivity are observed, consistent with the Fig. 3g data. Furthermore, we note that the data for Fig. 4a,c were taken using different pairs of contacts and that, in general, the observed transport characteristics are consistent across multiple sets of contacts (Extended Data Fig. 6).

The data presented here demonstrate the existence of correlations in ATQG, even when removed from the exact magic angle. While the physics of ATQG is closely analogous to TBG, there are distinct similarities and differences as a function of  $\theta$ . A comparison between our ATQG samples and TBG can be made by dividing the ATQG twist angle by the golden ratio ( $\approx 1.62$ ) to find an equivalent TBG twist angle. The  $\theta = 1.96^\circ$  and  $\theta = 1.52^\circ$  samples map to TBGs with  $\theta = 1.21^\circ$  and  $\theta = 0.94^\circ$ , respectively. The appearance of stronger insulating states on the hole side compared to the electron

side in the large-angle sample is similar to earlier results in TBG<sup>31</sup>, where correlated insulators probed through electronic compressibility measurements become stronger on the hole side for angles approaching 1.20°. In addition, the observation of superconductivity without correlated insulators for angles as low as 0.79° has been reported in TBG<sup>41</sup>. Our observations depart from the TBG picture when considering the asymmetry in the twist angle dependence of the superconductivity and correlated insulators, where the former is more prominent at smaller angles and the latter is more prominent at larger angles, while in TBG superconductivity is more robust on moving away from the magic angle in both directions<sup>30,41,42</sup>. While more data are needed to confirm this as a distinct feature of ATQG, it suggests that the presence of the dispersive bands may play a role in the stability of different correlated phases.

Our results also demonstrate the robustness of the superconducting phase to an in-plane magnetic field, exhibiting large Pauli limit violation similar to ATTG. This strongly suggests that superconductivity in both systems is not spin singlet. Additionally, the insensitivity of the insulating features to an in-plane field indicates a non-spin-polarized parent state. Both observations point towards a spin-valley-locked state as the most likely parent state, which yields a superconducting order that consists of a mixture of spin singlet and triplet<sup>43,44</sup>. Our findings show that ATQG, and ATMG more generally, is a unique playground for probing interactions beyond TBG. It highlights a decoupling of the superconducting phase and correlated insulators with twist angle, as well as a robustness to an in-plane magnetic field that is absent in TBG.

*Note added in proof:* During the preparation of this manuscript we became aware of two related studies, which show robust superconductivity in ATQG at the magic angle<sup>45,46</sup>.

## 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/s41563-022-01286-2>.

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## Methods

**Sample fabrication.** To prepare ATQG samples, h-BN and graphene were separately exfoliated onto Si/SiO<sub>2</sub> substrates. Large-area graphene flakes, typically with a length of at least 80 μm, were identified using optical microscopy and the single atomic layer thickness was confirmed with Raman spectroscopy. The h-BN flakes were similarly identified and their thickness and surface roughness determined with atomic force microscopy (AFM). The four constituent graphene layers were predefined by cutting the graphene flake using an AFM tip in contact mode. A hemispherical polymer stamp consisting of polydimethylsiloxane and a thin layer of spin-coated polypropylene carbonate on a glass slide was used to first selectively pick up an h-BN flake with a thickness of approximately 30 nm. Then, individual graphene layers were picked up sequentially by the top h-BN, while alternating the twist angle between each successive layer. The target angles during this process were usually ~0.3° larger than the final desired angle, to allow for relaxation during further processing. All pick-ups were performed at a temperature of 60 °C. The stack was then released onto a graphite/h-BN bottom gate at 170 °C, and a graphite top gate was subsequently transferred onto the heterostructure using a similar procedure to that described. Finally, electron beam lithography (EBL) and plasma etching were used to define the Hall bar geometry, and metal edge contacts were evaporated onto the sample using EBL and physical vapour deposition. The optical micrographs of the samples presented in the main text are shown in Extended Data Fig. 6.

**Measurement set-up.** Four-point transport measurements were performed using a low-frequency lock-in amplifier, with an excitation current ranging from 1 to 10 nA. The charge density ( $n$ ) and transverse electric field ( $E$ ) were independently controlled using top ( $V_{TG}$ ) and bottom ( $V_{BG}$ ) gate voltages. The density is given by  $n = (V_{TG}C_{TG} + V_{BG}C_{BG})/e$ , where  $C_{TG}$  and  $C_{BG}$  are the top and bottom gate capacitances, respectively, and  $e$  is the electron charge. The electric field is given by  $E = (V_{TG}C_{TG} - V_{BG}C_{BG})/2\epsilon_0$ , where  $\epsilon_0$  is the vacuum permittivity.

Measurements were performed in two different refrigerators. A dilution refrigerator was used for measurements performed at temperatures between 160 mK and 1.5 K, and a pumped <sup>4</sup>He refrigerator was used for measurements at temperatures of 1.5 K and higher.

**Assignment of full-filling density.** The value of  $n_s$  was experimentally determined by fitting Landau fans at large perpendicular magnetic fields (Fig. 2g and Extended Data Fig. 7). At high magnetic fields, the Landau level gaps in the dispersive bands are sufficiently large that the gate-induced charge density predominantly fills the flat bands and  $n_s$  can be accurately determined. At zero magnetic field, there is a partitioning of carriers between the flat and dispersive band, and consequently the half- and full-filling resistance maxima in Figs. 1e and 3a do not exactly align with the designated  $-n_s/2$  and  $-n_s$  values on the respective  $x$  axes.

**Quantum oscillations in a perpendicular magnetic field.** In a perpendicular magnetic field ( $B$ ), electrons take on discrete cyclotron orbits known as Landau levels, with quantized energies that are proportional to  $B$  and have a level index, or

filling factor,  $\nu$ . Each sub-band ( $s$ ) of the band structure splits into Landau levels, which manifests in transport as minima in the longitudinal resistance every time the charge density ( $n$ ) is modulated to fill a Landau level. Landau levels are indexed by  $s$  and  $\nu$ , and are related by  $n = \nu(eB/h) + sn_s$ , where  $e$  is the electron charge and  $h$  is Planck's constant. Experimentally, these appear as 'fans' of low resistance that extend outward from band or correlated insulators (Fig. 2 and Extended Data Figs. 7 and 8).

## Data availability

Source data are provided with this paper. All other supporting data are available from the corresponding author upon reasonable request.

## Code availability

The supporting code for this paper is available from the corresponding author upon reasonable request.

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## Author contributions

G.W.B. and E.T. conceived and designed the experiment. G.W.B. fabricated and measured the samples, and Y.W. assisted with measurements. G.W.B., E.K. and E.T. analysed the data. E.K. provided band structure calculations. K.W. and T.T. provided the boron nitride crystals. All authors contributed to discussions and writing of the manuscript.

## Competing interests

The authors declare no competing interests.

## Additional information

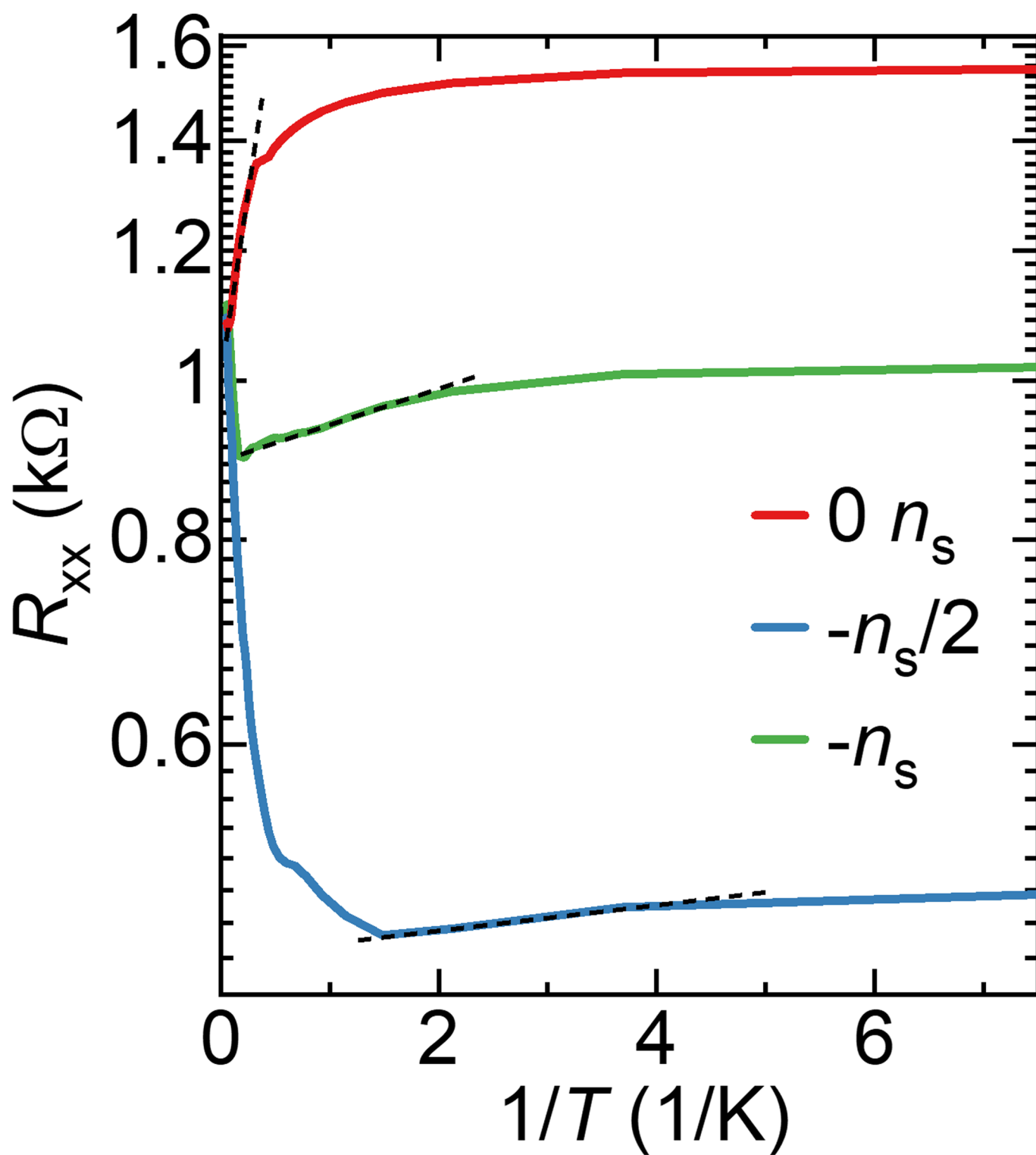
**Extended data** is available for this paper at <https://doi.org/10.1038/s41563-022-01286-2>.

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**Correspondence and requests for materials** should be addressed to Emanuel Tutuc.

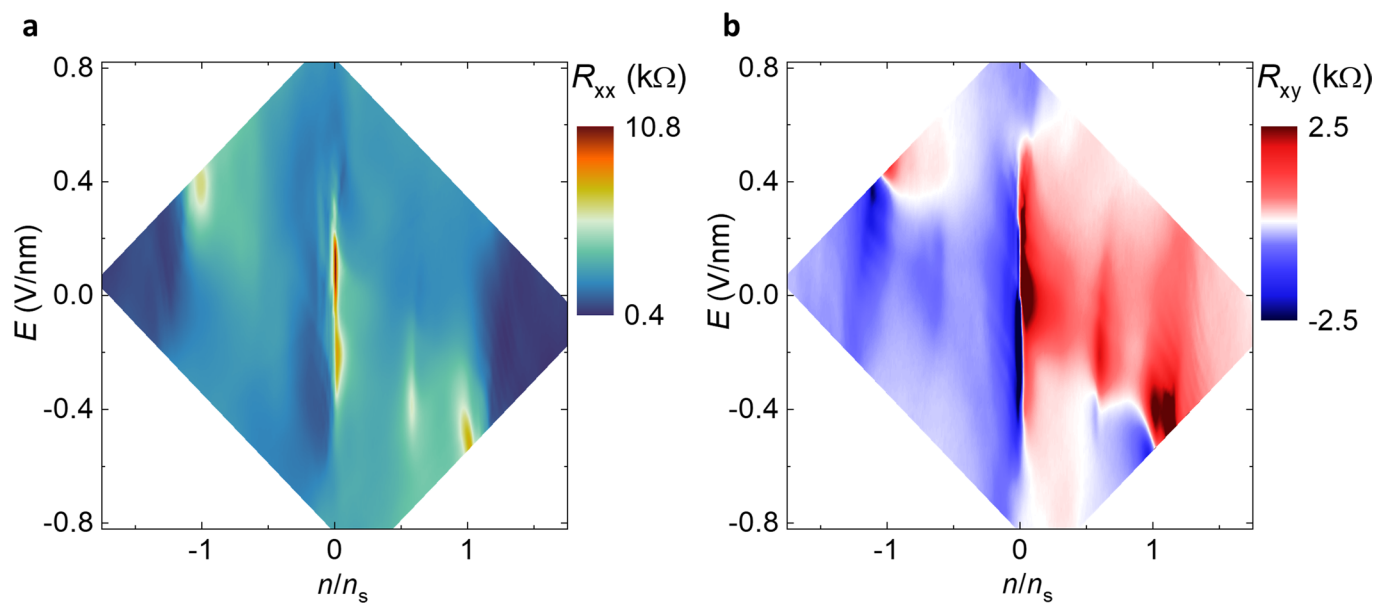
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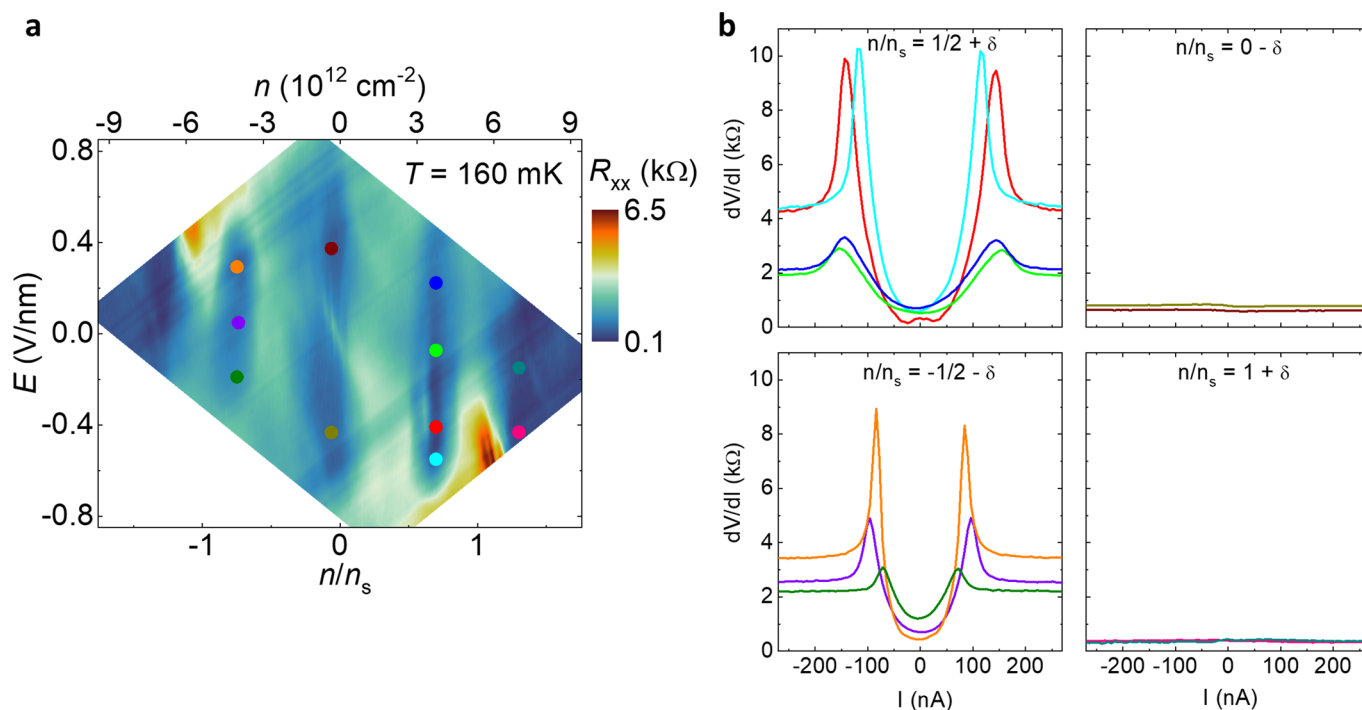


**Extended Data Fig. 1 | Arrhenius plot of correlated and band insulators in  $\theta = 1.96^\circ$  ATQG.** Log-scale  $R_{xx}$  vs.  $1/T$  at  $n/n_s = 0, -1/2$ , and  $-1$ , using the same data as in Fig. 1f of the main text. The linear portions of each trace are marked by a dashed line.

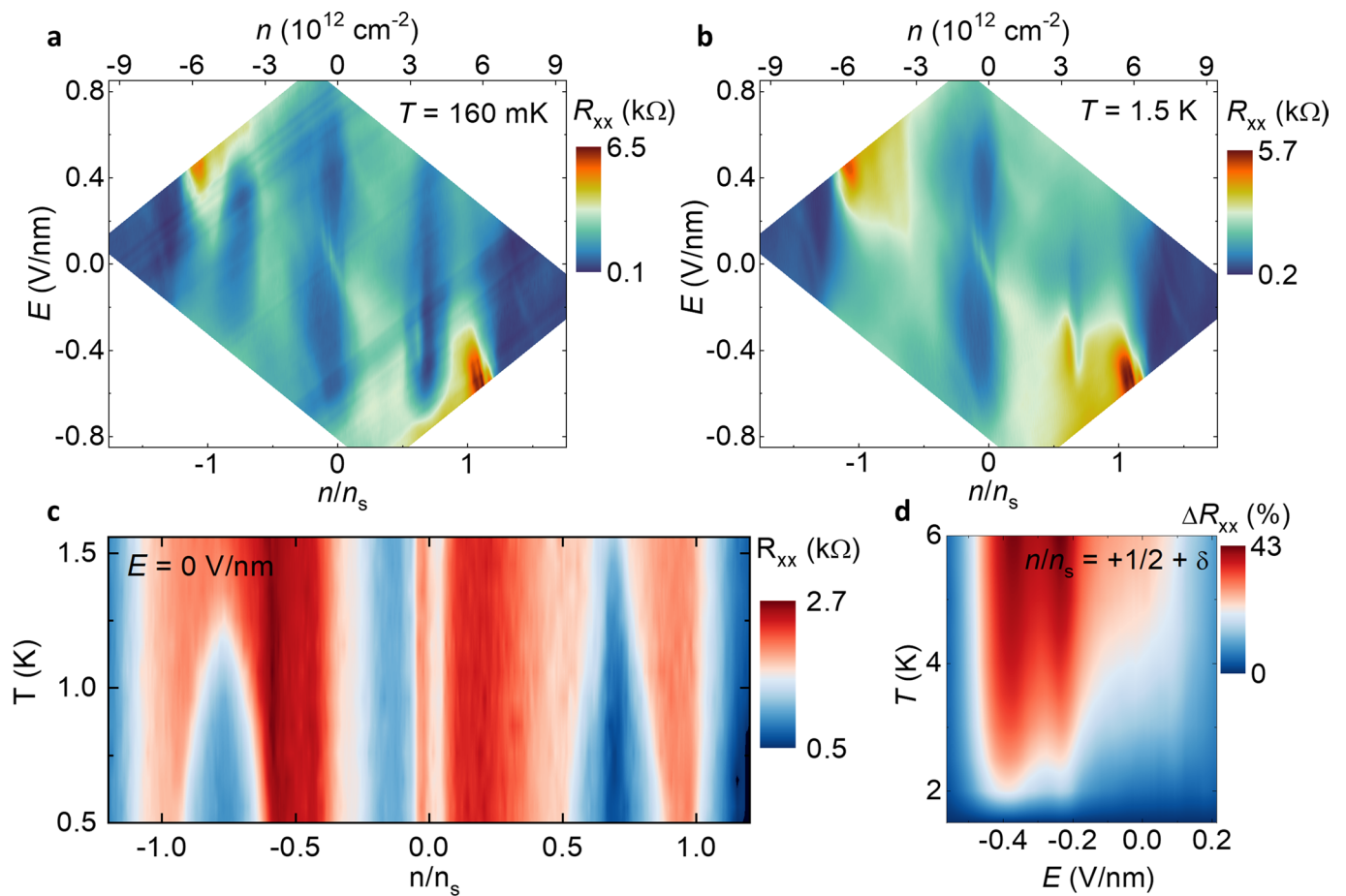




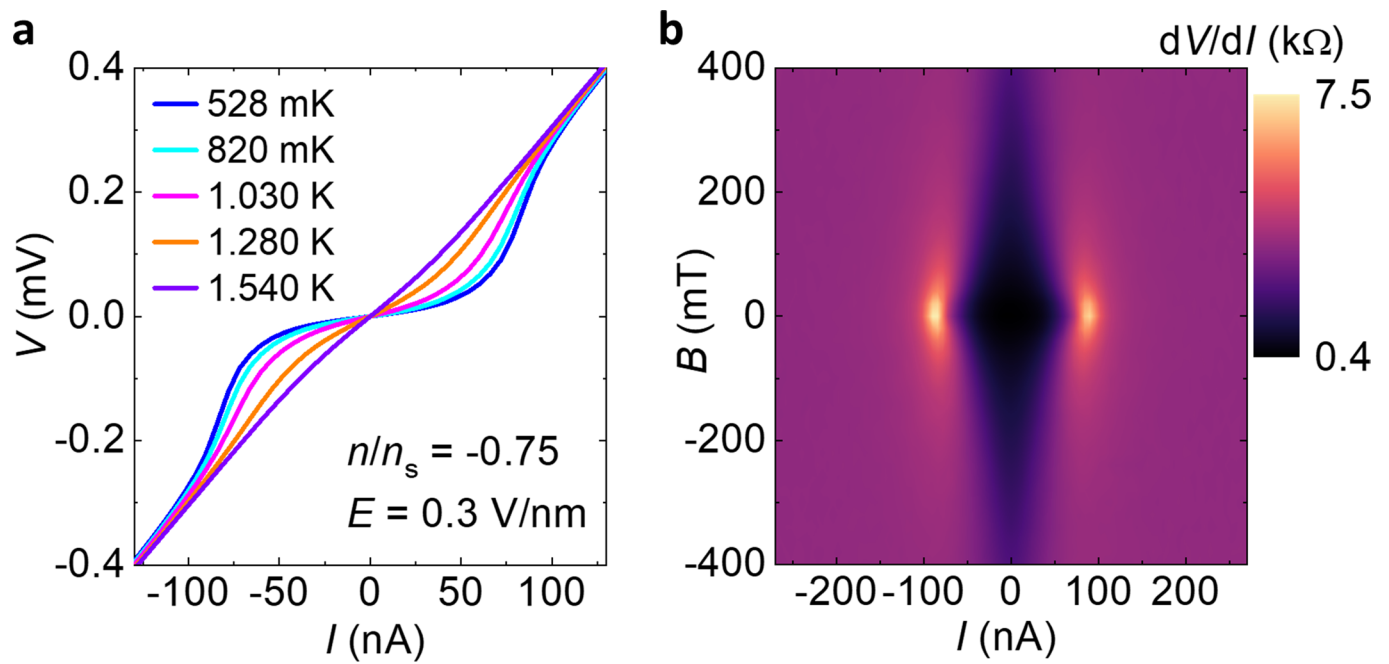
**Extended Data Fig. 2** |  $\theta = 1.52^\circ$  ATQG at  $B = 1$  T. **a-b**,  $R_{xx}$  (**a**) and  $R_{xy}$  (**b**) vs.  $n/n_s$  and  $E$  at  $B = 1$  T. Similar to the Fig. 2 data for  $\theta = 1.96^\circ$  ATQG,  $R_{xx}$  maxima become more apparent at half and full-filling and  $R_{xy}$  data show trend reversals indicative of a phase transition at the correlated insulators.



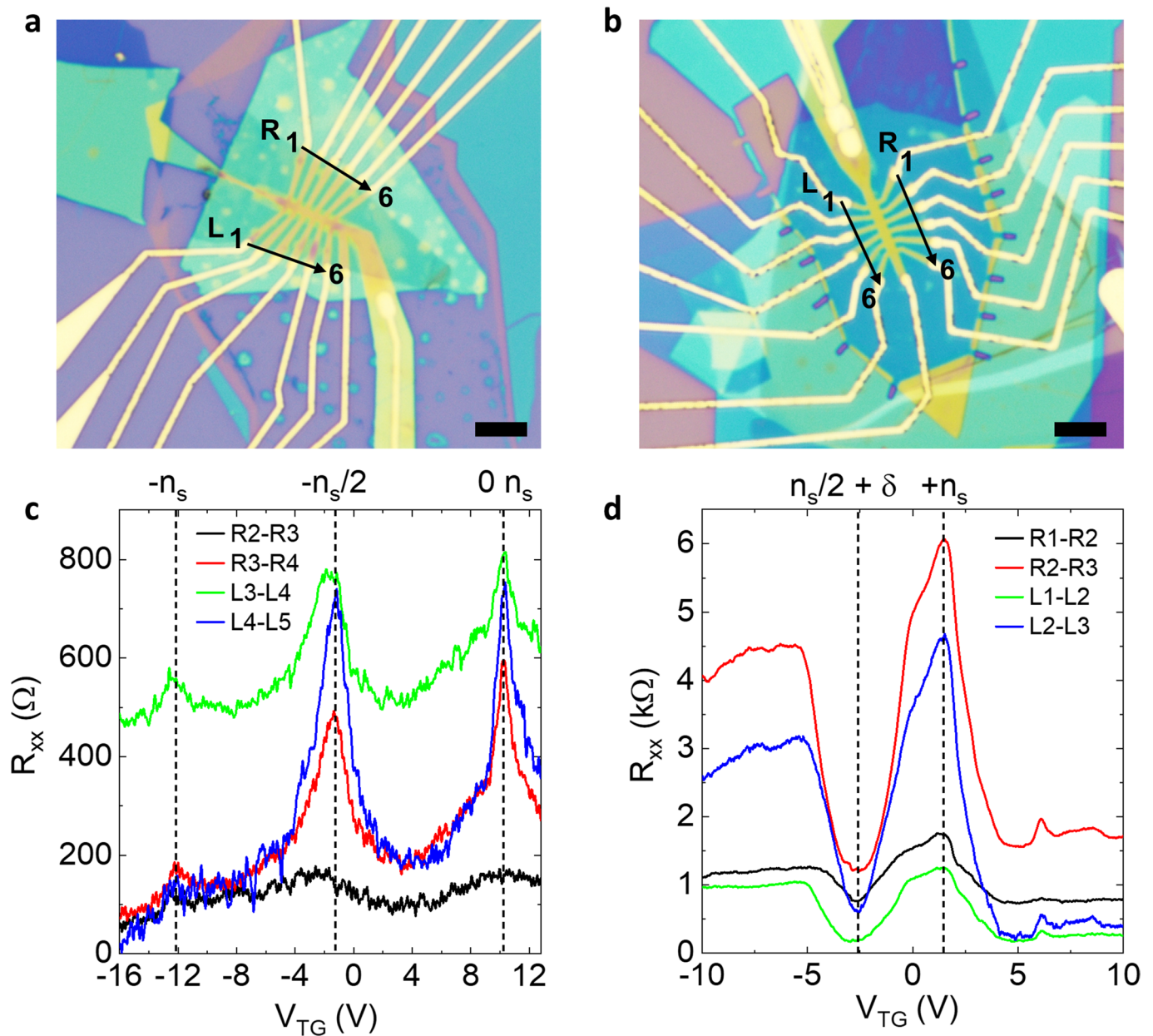
**Extended Data Fig. 3 |  $dV/dI$  vs  $I$  at different  $n/n_s$  and  $E$ .** **a**,  $R_{xx}$  vs.  $n/n_s$  and  $E$  in the  $\theta = 1.52^\circ$  sample, the same data shown in Fig. 3a of the main text. The dots indicate points where the  $dV/dI$  vs.  $I$  measurements were taken. **b**,  $dV/dI$  vs.  $I$  at  $n/n_s = 1/2 + \delta$  (upper left),  $0 - \delta$  (upper right),  $-1/2 - \delta$  (lower left), and  $1 + \delta$  (lower right). The line colors correspond to the dot colors in **a**. Two primary observations can be made from the data. First, regions near half-filling show a critical current in the  $dV/dI$  vs.  $I$  at all  $E$ , while data acquired at other fillings show a  $dV/dI$  that is insensitive to  $I$ . Second, the peaks in  $dV/dI$  are generally most pronounced at larger  $E$ , in agreement with the theoretical picture that the critical temperature increases at finite  $E$  for samples with a twist angle below the magic angle<sup>24</sup>.



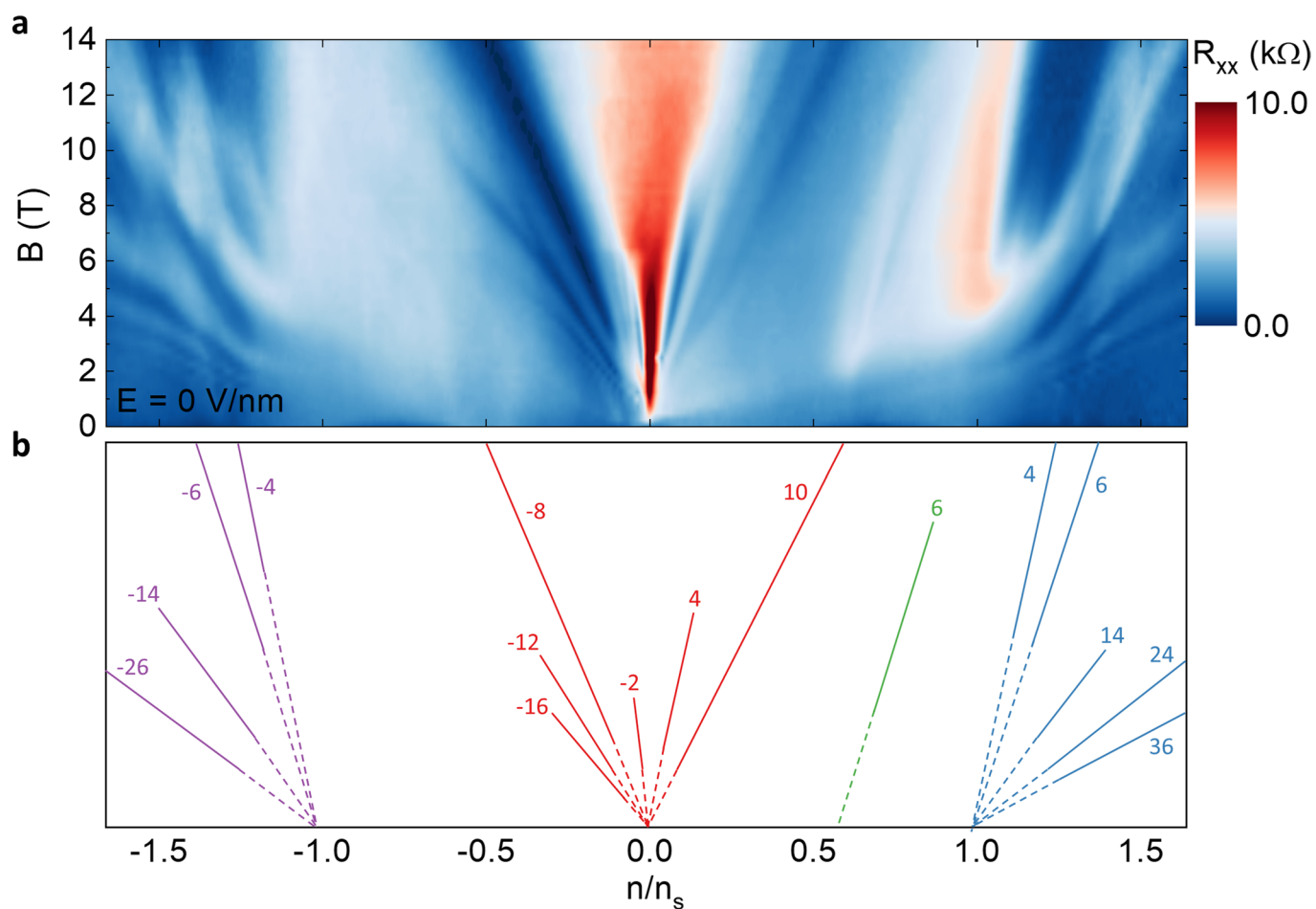
**Extended Data Fig. 4 | Additional temperature dependence in the  $\theta = 1.52^\circ$  ATQG.** **a-b**,  $R_{xx}$  vs.  $n/n_s$  and  $E$  at  $T = 160$  mK (**a**) and  $T = 1.5$  K (**b**). At the lower temperature,  $E$ -dependent regions of low resistance appear at  $n = \pm(n_s/2 + \delta)$  while all other regions remain insensitive to temperature between the two contour plots. **c**,  $R_{xx}$  vs.  $n/n_s$  and  $T$  at  $E = 0$  V/nm. Low resistance domes form on the higher density side of half-filling on both the electron and hole side. Consistent with **a**, **b**, the data suggest a higher critical temperature on the electron side compared to the hole side. **d**,  $\Delta R_{xx}$  vs.  $E$  and  $T$  at  $n/n_s = 1/2 + \delta$  for  $T$  between 1.5 K and 6 K, where  $\Delta R_{xx}$  is the percentage change in  $R_{xx}$  from the  $T = 1.5$  K value.  $R_{xx}$  changes more rapidly at larger  $E$ , suggesting an onset of correlations at higher temperatures with increasing  $E$ .



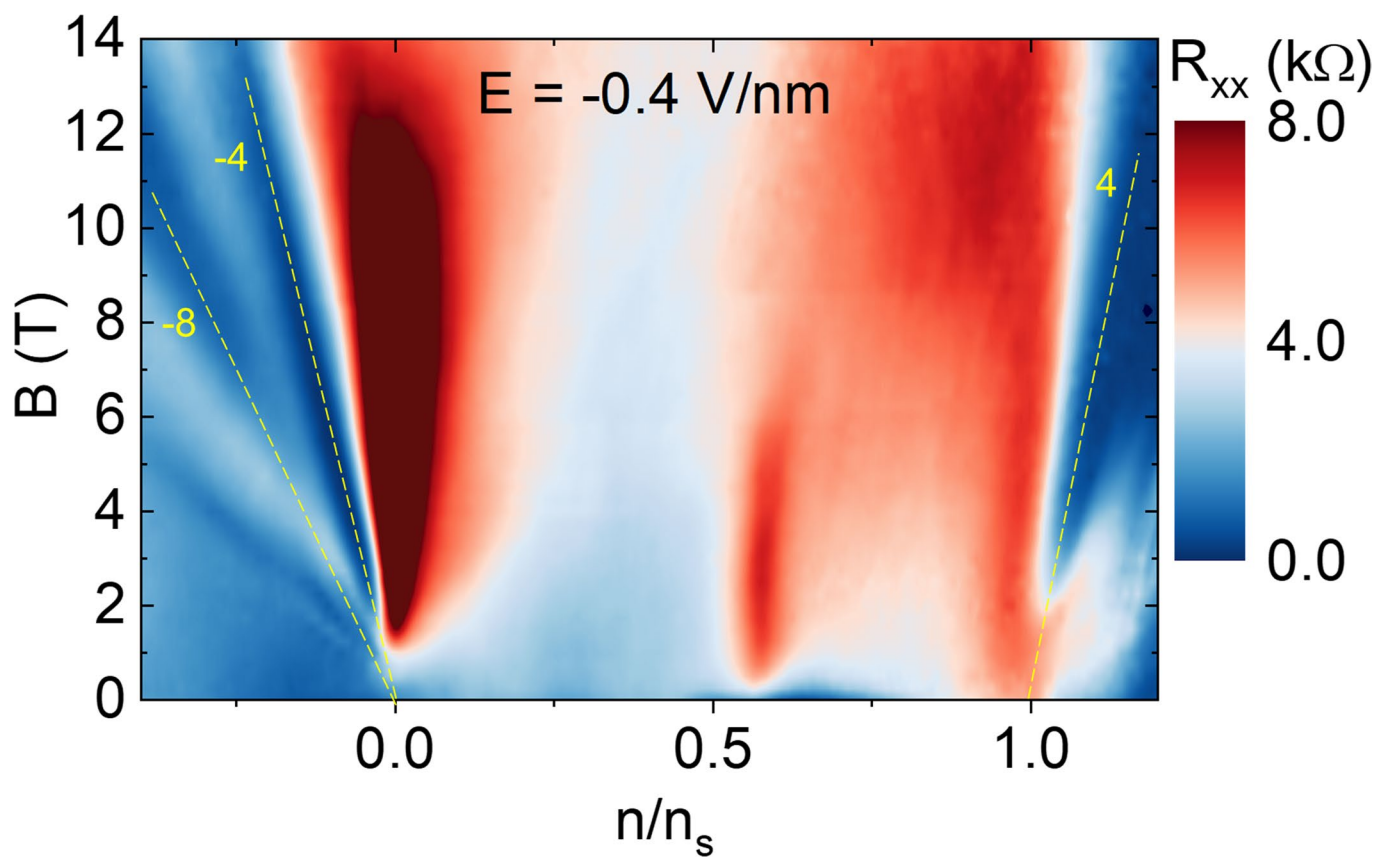
**Extended Data Fig. 5 | Evidence of superconductivity on hole side.** **a**,  $V$  vs.  $I$  at  $n/n_s = -0.75$  and  $E = 0.3$  V/nm for temperatures between 528 mK and 1.540 K in the  $\theta = 1.52^\circ$  sample. At the highest temperature, the critical current behavior is extinguished and the characteristic becomes linear. **b**,  $dV/dI$  vs  $I$  and  $B$  at the same  $n/n_s$  and  $E$  as in **a**. A small  $B$ -field suppresses the nascent superconductivity, similar to the Fig. 4a data of the main text.



**Extended Data Fig. 6 | Twist angle uniformity in the ATQG samples.** **a-b**, Optical micrographs of the  $\theta = 1.96^\circ$  (**a**) and  $\theta = 1.52^\circ$  (**b**) samples. The voltage probe contacts are labeled in each panel, and the scale bars are  $5 \mu\text{m}$ . **c-d**, representative traces of  $R_{xx}$  vs.  $V_{TG}$  in the  $\theta = 1.96^\circ$  (**c**) and  $\theta = 1.52^\circ$  (**d**) samples using four different voltage probe contact pairs. For the  $\theta = 1.96^\circ$  sample, the  $R_{xx}$  maxima corresponding to the band insulators at  $0 n_s$  and  $-n_s$  closely align, confirming a uniform twist angle to within  $\pm 0.01^\circ$  throughout the channel. Similarly, in the  $\theta = 1.52^\circ$  sample, the positions of the resistance maxima at  $+n_s$  and resistance dips at  $n_s/2 + \delta$  that precede the superconducting state closely match across all contact pairs.



**Extended Data Fig. 7 | Landau fans in the  $\theta = 1.52^\circ$  ATQG.** **a**,  $R_{xx}$  vs.  $n/n_s$  and  $B$  at  $E = 0 \text{ V/nm}$ , showing quantum oscillations as described in the Methods section. **b**, Landau fans observed in panel **a**, along with the Landau level filling factor. The filling factor of 6 at  $n = n_s/2$  is indicative of a band with Chern number 2, as discussed in the main text for the  $\theta = 1.96^\circ$  sample.



**Extended Data Fig. 8 | Landau fans near nascent superconductivity.**  $R_{xx}$  vs.  $n/n_s$  and  $B$  at  $E = -0.4 \text{ V/nm}$  in the  $\theta = 1.52^\circ$  sample. The prominent Landau levels are labeled. Near half-filling, the low resistance domes diminish at small  $B$  values, and a resistance maximum develops at the correlated insulator.