Flat band carrier confinement in magic-angle twisted bilayer graphene

Nikhil Tilak\textsuperscript{1}, Xinyuan Lai\textsuperscript{1}, Shuang Wu\textsuperscript{1}, Zhenyuan Zhang\textsuperscript{1}, Mingyu Xu\textsuperscript{2,3}, Raquel de Almeida Ribeiro\textsuperscript{2,3}, Paul C. Canfield\textsuperscript{2,3} & Eva Y. Andrei\textsuperscript{1}**

Magic-angle twisted bilayer graphene has emerged as a powerful platform for studying strongly correlated electron physics, owing to its almost dispersionless low-energy bands and the ability to tune the band filling by electrostatic gating. Techniques to control the twist angle between graphene layers have led to rapid experimental progress but improving sample quality is essential for separating the delicate correlated electron physics from disorder effects. Owing to the 2D nature of the system and the relatively low carrier density, the samples are highly susceptible to small doping inhomogeneity which can drastically modify the local potential landscape. This potential disorder is distinct from the twist angle variation which has been studied elsewhere. Here, by using low temperature scanning tunneling spectroscopy and planar tunneling junction measurements, we demonstrate that flat bands in twisted bilayer graphene can amplify small doping inhomogeneity that surprisingly leads to carrier confinement, which in graphene could previously only be realized in the presence of a strong magnetic field.
When two sheets of graphene are superposed with a relative twist angle, they develop a moiré superstructure with an angle-dependent wavelength which reflects the local stacking variation of the two crystal lattices. Hybridization between the two sets of electronic bands produces a strongly modified, twist angle-dependent band structure. Close to a "magic" angle (~1°), this leads to very narrow, almost dispersionless (flat) low-energy bands. These flat bands are isolated from the dispersive bands by single-particle bandgaps. Since the kinetic energy of the electrons in the flat bands is quenched, e-e interactions become important and give rise to phenomena such as correlated insulating states, superconductivity, emergent ferromagnetism, etc. The electronic properties of twisted bilayer graphene (TBG) were initially explored in naturally occurring TBG synthesized by chemical vapor deposition, through the use of scanning tunneling microscopy/spectroscopy (STM/STS). More recently, the development of techniques to control the twist angle and the doping level has expanded the range of capabilities to include global measurements such as magnetotransport, electronic compressibility, angle-resolved photoemission spectroscopy, as well as gated local probes including STM/STS, nano-SQUID on-tip microscopy, local compressibility.

Obstacles to progress in this field include twist angle and doping inhomogeneity. Whereas efforts to address the former are being undertaken, the effects of local doping variation have thus far been ignored. Naively, this may be justified by the fact that spatial doping variations in samples using hexagonal boron nitride (hBN) substrates can be as low as 10\(^{10}\) cm\(^{-2}\), which is two orders of magnitude lower than the typical charge density in magic-angle TBG, \(\sim 10^{12}\) cm\(^{-2}\). However, as we demonstrate below, even such low levels of density inhomogeneity can radically change the response and electronic properties of the system, obscuring the moiré physics when the Fermi level is aligned to the edge of the flat band.

Using tunneling experiments with a traditional STM as well as a novel planar tunneling device, we find that near the edges of the flat bands, the local doping variations which are ubiquitous in TBG devices, produce patches of conducting regions separated by insulating regions. This leads to carrier confinement on a scale typically larger than the moiré wavelength which can conceal the magic-angle physics.

**Results**

We begin by discussing the results of tunneling measurements on a TBG device fabricated by a tear-and-stack technique (see "Methods" for details). The schematic experimental setup is shown in Fig. 1a. We navigate to the micron size sample using a capacitance-based technique. We identify a magic-angle region via STM topography and spectroscopy measurements. The honeycomb lattice of the carbon atoms in graphene is composed of two triangular sublattices labeled A and B. The flat bands are localized mostly on the AA stacking regions, where every atom in the top layer is positioned directly on top of an atom from the bottom layer. These appear as circular bright spots in the topography image when the flat bands are occupied (Fig. 1b). Surrounding the AA regions are six darker regions called AB/BA. In the AB (Bernal stacked) regions, top layer A sublattice atoms are positioned directly above bottom B sublattice atoms while the top layer B sublattice atoms have no partners in the bottom layer. The BA regions are defined similarly via sublattice symmetry. These stacking arrangements are illustrated in Fig. 1c.

The local twist angle (\(\theta\)) is determined by measuring the average moiré wavelength \((L_M)\) in the three crystallographic directions using the relation

\[ L_M = a / (2 \sin(\theta/2)), \]

where \(a = 0.246\) nm is the lattice constant of monolayer graphene. The data presented below were collected in a region with a twist angle of 1.12°, over at least 30 moiré unit cells. The heterostrain in the region was estimated to be 0.2% following reference.

Each flat band in TBG is fourfold degenerate owing to the valley and spin degrees of freedom. In total, it carries a carrier density of eight electrons per moiré cell to completely fill both the flat bands. For a twist angle \(\theta\) this corresponds to a carrier density

\[ n = \frac{1}{e} \left( V_g - V_{g0} \right) \left( \frac{d_1}{\epsilon_1} + \frac{d_2}{\epsilon_2} \right)^{-1}. \]

Here \(V_{g0}\) is the gate voltage needed to tune the system to charge neutrality, \(\epsilon_0\) is the permittivity of vacuum, \(e\) is the electronic charge, \(d_1\) and \(d_2\) are the thickness and dielectric constant where \(i = 1, 2\) correspond to hBN and SiO\(_2\), respectively. Given the geometry of our device and the measured twist angle, it takes 75–85 V of backgate to completely fill the empty flat bands.

Figure 1d shows a typical STS curve, which provides a measure of the local density of state (LDOS) within the AA regions when the flat bands are completely filled. The two sharp peaks in the LDOS correspond to two Van Hove singularities (VHS) in the electronic spectrum where the density of states diverges. The rapid decrease in the LDOS near the band edges suggests that close to the empty or full band the electronic properties are particularly susceptible to local doping variations, caused by twist angle inhomogeneity, impurities or defects. We measure the full width at half maximum of the electron and hole side flat bands to be \(\sim 16\) meV and the two VHS are separated by \(\sim 18\) meV. The dips in the \(dI/dV_b\) between the flat bands and the remote bands correspond to the single-particle superlattice gaps.

Next, we measured the gate voltage dependence of the STS at an AA site (Fig. 2a). In addition to the flat band and the dispersive bands, a series of sharp peaks were observed in the \(dI/dV_b\) spectra when the Fermi level was tuned close to the full filling of the electron–side flat band (Fig. 2c). These peaks, which are almost equidistant \((\Delta V_b \approx 60\) meV\) in bias voltage, move toward higher bias values as the flat bands are filled. This is opposite to the expected gating behavior for normal features in the DOS which should evolve toward smaller bias values as the bands are filled.

Furthermore, when the peaks intersect the flat band at the Fermi level \((V_b = 0\) mV\), a series of diamond-like structures appear in the gate dependence map which resembles Coulomb diamonds seen in quantum dots. These can be seen more clearly in the zoomed-in view of the gate dependence map as shown in Fig. 2b. After eliminating trivial explanations of the origin of these peaks (such as dirt stuck to the tip), we concluded that the effect is intrinsic to the sample and not a tip artifact (see Supplementary Note 8). Recalling that, owing to the chiral nature of the quasiparticles, backscattering in graphene is suppressed, the observation of Coulomb diamonds may be surprising. In fact, Coulomb diamonds in graphene are not observed without applying a strong out-of-plane magnetic field. The magnetic field splits the bands into flat Landau levels separated from each other by an energy gap which depends on the magnitude of the applied magnetic field. Split gates or tip-induced band bending can be used to locally bend the Landau levels which produce...
quantum dots in such samples, which are observed as Coulomb diamonds\(^{31}\) in conductance vs gate voltage maps.

We show that similar to the Landau levels in graphene, the flat bands in magic-angle TBG amplify local density variations, but without the need of applying a magnetic field. To this end, we determined the gate voltage at which the flat bands are almost completely full, so that the Fermi level lies at the edge of the flat bands. At this gate voltage, 68 V, we collected spatial \(dI/dV_b\) maps, shown in Fig. 3a for bias voltage \(V_b = 0\) mV. These maps reveal prominent contrasting patches of bright and dark regions, corresponding to charge puddles created by the local doping inhomogeneity. A comparison of spectra (Fig. 3b) gathered in the bright and dark regions at positions marked by the green and red crosses respectively, shows the flat band is completely full in the dark regions while it is partially empty in the bright regions, directly confirming the spatial doping variation in this sample.

To further characterize this doping variation, we plot in Fig. 3c a line-cut across the conducting region in Fig. 3a along a path denoted by the yellow arrow. The energy of the charge neutrality point \(E_{\text{CNP}}\) is marked by the solid black line as a guide to the eye. In the absence of any doping variation, one would expect the \(E_{\text{CNP}}\) to be constant regardless of the position for a given filling. The black line traces the shape of the potential well created by the doping disorder at this gate voltage. Since there are no electronic states available near \(E_F\) in the regions immediately surrounding the bright islands, the carriers in the bright regions should be confined within them. They can only occupy discrete energy levels whose separation depends on the shape and size of the potential well. Carriers from the tip can tunnel into the bright regions only into these discrete energy levels. These confined carriers can tunnel from the bright conducting islands to the nearby islands through the short insulating barrier separating them. We illustrate this situation with a tunneling diagram in Fig. 3d. Hereafter we refer to these conducting regions as quantum dots.

Valuable information about quantum dots can be extracted by analyzing their charging characteristics. Upon analyzing the Coulomb diamonds in Fig. 2b, the capacitances of the quantum dot to the tip \(C_d\), backgate \(C_g\) and the surrounding conducting regions \(C_r\) were found to be in the ratio \(C_d : C_r : C_g = 1 : 37 : 36\) (see Supplementary Note 3). This implies that although \(V_b\) is typically small compared to \(V_g\), the tip is very efficient at gating the dot because of its larger capacitive coupling. This explains the positive slope of the oblique charging lines in the gate dependence map (Fig. 2a). The size of the quantum dot estimated by using a disc model \((61 \pm 4.8\) nm) is consistent with the size of the conducting island shown in Fig. 3a.

We also performed high resolution \(dI/dV_b\) spatial mapping in the conducting region indicated by the dotted magenta square in Fig. 3a in order to directly image the confined carriers (Fig. 3e).
The three bright wavefronts in Fig. 3e correspond to three lowest energy states in the conducting region. See Supplementary Note 5 for more detailed analysis of the evolution of these wavefronts.

Next, we comment on the possible reasons behind the observed doping variation. The TBG rests on a thin hBN flake (23.5 nm). The hBN is exfoliated on top of a commercial highly p-doped Silicon chip with a thermally grown oxide layer (WaferPro, LLC). Silicon dioxide grown on Silicon wafers is known to suffer from doping variation. The TBG rests on a thin hBN, which is a wide gap dielectric, while the top hBN is ultra-thin (~20 nm). This device consists of an hBN encapsulated TBG with a local metallic backgate. The thick bottom hBN acts as a gate dielectric, while the top hBN is ultra-thin (~4 layers) and acts as a tunneling barrier. After locating a clean TBG region with atomic force microscopy (AFM), a metallic tunneling electrode is deposited on top of the thin hBN layer through a 600 nm diameter circular hole etched in a thick hBN (see "Methods"). A bias voltage is applied between the tunneling electrode and the TBG and the resulting tunneling current is amplified and measured. Similar to the STM measurement, we obtain the differential conductance ($dI/dV_b$) using a standard lock-in technique and tune the Fermi level of the sample by applying a gate voltage.

We estimate the twist angle from the measured energy separation of the VHS peaks $\Delta E_{\text{VHS}}$:

$$\Delta E_{\text{VHS}} \approx \frac{\hbar v_F}{a} \Delta \theta$$

where $\hbar$ is Plank’s constant, $v_F$ is the Fermi velocity of electrons in graphene, $a$ is the lattice constant of graphene, and $\Delta \theta$ is the interlayer tunneling parameter. A typical $dI/dV_b$ spectrum of TBG at $V_b = 0$ V is shown in Fig. 4b. Two peaks corresponding to the VHS which flank the charge neutrality point are consistent with STS results on TBG close to the magic angle. The measured $\Delta E_{\text{VHS}}$ of $48 \pm 2$ meV yields a twist angle of $(1.25 \pm 0.05)$

Assuming that the work function of TBG is close to the work function for Bernal stacked bilayer Graphene (4.7 eV), and that the work function of the tunneling electrode (Cr/Au) is close to that of Chromium (4.5 eV), we can estimate a work function difference of the order of 100 meV between the TBG and the tunneling electrode. This work function difference results in the TBG becoming electron doped and creates a potential well, approximately the size of the tunneling electrode, where charge carriers can be confined. Similar results were seen previously in STM experiments on Bernal bilayer graphene, where a potential well was created by charging the impurities in hBN. This situation closely resembles the potential wells observed in the STM device, albeit with different length scales and origins of the confinement potentials. The schematic diagram (Fig. 4d) illustrates this scenario. Indeed, gate dependence maps measured at 4.2 K (Fig. 4c) show Coulomb diamond-like features. On further cooling the sample to 0.3 K (Fig. 4e) and performing high resolution
**Fig. 3** Evidence of local doping variation in the sample. 

**a** Shows a \( \frac{dI}{dV_b}(x, y, V_g = 0 \text{ mV}, V_g = 68 \text{ V}) \) spatial map of a 246 \( \times \) 246 nm\(^2\) region (scale bar 50 nm). Several bright regions, indicating a high density of states, which are surrounded by dark regions, indicating a low density of states are visible. The thin yellow lines are a guide for the eyes. 

**b** Shows individual \( \frac{dI}{dV_b} \) spectra measured at the positions marked by the red and green crosses in **a**. A lateral shift of the two spectra with respect to the \( E_F \) indicates a difference in the local chemical potential. The approximate location of the charge neutrality point taken as the dip between the two VHS is also labeled. 

**c** is a line-cut across the conducting island in **a** in a direction indicated by the yellow arrow. The variation of the local charge neutrality point energy (\( E_{\text{CNP}} \)) as a function of position can be seen which indicates local doping variation. The black line is a 5th order polynomial fit. Error bars represent uncertainty in the determination of CNP due to finite energy resolution. The bright conducting regions surrounded by the darker insulating regions act like quantum dots. 

**d** A tunneling diagram of the system. There are two tunneling barriers: the large vacuum barrier between the tip and the dot and the shorter barrier between the dot and the bulk of the sample. The quantum dot is defined by a potential well which has the shape indicated by the local charge neutrality point. Confined states within this potential well are indicated by horizontal lines. There are low-energy electron tunneling events from the tip into the confined states of the quantum dot and then into the bulk of the sample (blue arrow). There also exist direct tunneling from the tip to the higher energy bands of the bulk of the sample (red arrow). 

**e** Shows a high resolution \( \frac{dI}{dV_b}(x, y, V_g = -224 \text{ mV}, V_g = 57 \text{ V}) \) spatial map of the conducting island indicated by the magenta square in **a**. The three bright wavefronts are three single electron charging states confined in the bright island. The shape of the bright wavefronts approximately matches the shape of the bright island.

**Fig. 4** Carrier confinement in a TBG planar tunneling device. 

**a** Schematic of the planar tunneling device. An ultra-thin hBN acts as a tunneling barrier between the metallic tip and the TBG. 

**b** Tunneling spectrum measured at 0 V backgate voltage showing the flat bands. The sample is highly n-doped because of the work function difference between the tunneling electrode and the TBG. 

**c** Gate dependence map of the STS. Flat bands can be seen as two bright parallel lines at negative bias. The other features are a result of confinement. 

**d** Schematic of the position dependence of the charge neutrality point. \( \mu_0 \) is the shift in the charge neutrality point under the tip compared to the bulk region. 

**e** High resolution \( \frac{dI}{dV_b} \) gate dependence maps measured at 0.3 K showing coulomb diamonds, a result of electron confinement.
spectroscopic measurements, the resulting gate dependence shows well-developed Coulomb diamonds reflecting the carrier confinement in the device. The charging energy ($E_C$) extracted from the diamond-like features in Fig. 4e, ~2 meV, is much smaller than that observed in the STM device, consistent with a much larger dot size. The diameter of the QD in this case, estimated by using a 2D disc capacitance model ($\sim 640$ nm), agrees well with the size of the tip electrode. This supports the scenario of electron confinement due to the strong amplification of potential inhomogeneities in the flat band of magic-angle TBG.

**Planar tunneling device.** The planar tunneling device consists of two parts, a top hBN layer containing etched holes plus an ultra-thin hBN tunneling layer (t-hBN), and a TBG/hBN stack with a local metal gate.

**Preparation of top hBN with etched holes.** Patterning of exfoliated hBN on SiO$_2$ was carried out using e-beam lithography. Then the patterned holes were etched out through CHF$_3$/O$_3$ plasma followed by a lift-off process.

**Assembly of the hBN/t-hBN/TBG/gold stack.** The hBN/t-hBN/TBG/gold stack was assembled with the twist transfer method in a glovebox (Argon atmosphere), using a stamp consisting of polypropylene carbonate film and PDMS. The hBN film with etched holes (30–50 nm thick) is firstly picked up. Then the t-hBN (~4 layers) is picked up by the hBN on the stack. The hBN/t-hBN stack is then deposited onto the TBG/hBN/gold stack that is prepared separately in step (2). During the assembly of the stack, the temperature is kept below 160 °C. AFM and electrostatic force microscopy are subsequently used to identify the region of the TBG prior to depositing the electrical contacts (Cr/Au) for tunneling measurements. The tunneling measurements are carried out in He-3 system with a base temperature of 0.3 K. The $dI/dV$ measurements are used standard lock-in technique by adding AC excitation of 0.2–1 mV at the bias of 7.1 Hz.

**Growth of crystalline hBN.** Single-crystalline hBN was grown at high temperature and high pressure using a Rockland Research, cubic-multi-anvil press system. Elemental Mg and 11B in a Mg$_2$B$_3$O$_5$ molar ratio, were placed in a 7 mm long, 5.7 mm inner diameter BN crucible with any empty space filled by extra BN powder. The crucible was then taken to 3.3 GPa at room temperature and heated to 1380 °C over 2 h. After dwelling at 1380 °C for 1 h, the system was cooled to 580 °C over 6 h and then quenched to room temperature. Once at room temperature the pressure was brought back to ambient conditions. Post growth, the crucible was removed from the pressure media and sealed in an amorphous silica amouple. Excess Mg was distilled from the hBN by heating one end of the ampoule to 750 °C, while the other end hung out of the end of a horizontal tube furnace for 200 min. Thin, clear flakes of hBN with planar dimensions of up to 1 × 2 mm$^2$ were separated from the growth crucible.

**Data availability**

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

Received: 5 October 2020; Accepted: 14 June 2021; Published online: 07 July 2021

**References**

1. Li, G. et al. Observation of Van Hove singularities in twisted graphene layers. *Nat. Phys.* 6, 109–113 (2010).

2. Andrei, E. Y., Li, G. & Du, X. Electronic properties of graphene: a perspective from scanning tunneling microscopy and magnetotransport. *Rep. Prog. Phys.* 75, 056501 (2012).

3. Bistritzer, R. & MacDonald, A. H. Moire bands in twisted double-layer graphene. *Proc. Natl Acad. Sci. USA* 108, 12233–12237 (2011).

4. Lopes dos Santos, J. M. B., Peres, N. M. R. & Castro Neto, A. H. Graphene bilayer with a twist: electronic structure. *Phys. Rev. Lett.* 99, 256802 (2007).

5. Cao, Y. et al. Superlattice-induced insulating states and valley-protected orbits in twisted bilayer graphene. *Phys. Rev. Lett.* 117 https://doi.org/10.1103/PhysRevLett.117.116804 (2016).

6. Cao, Y. et al. Correlated insulator behaviour at half-filling in magic-angle graphene superlattices. *Nature* https://doi.org/10.1038/nature26154 (2018).

7. Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. *Nature* https://doi.org/10.1038/nature26160 (2018).
8. Sharpe, A. L. et al. Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene. *Science* **365**, 605–608 (2019).
9. Serlin, M. et al. Intrinsic quantized anomalous Hall effect in a moiré heterostructure. *Science* **367**, 900–903 (2020).
10. Kim, K. et al. van der Waals heterostructures with high accuracy rotational alignment. *Nano Lett.* **16**, 1989–1995 (2016).
11. Yankowitz, M. et al. Tuning superconductivity in twisted bilayer graphene. *Science* **363**, 1059–1063 (2019).
12. Stepanov, P. et al. Uniting the insulating and superconducting orders in magic-angle graphene. *Nature* **583**, 375–378 (2020).
13. Lu, X. et al. Superconductors, orbital magnets and correlated states in magic-angle bilayer graphene. *Nature* **574**, 653–657 (2019).
14. Wu, S., Zhang, Z., Watanabe, K., Taniguchi, T. & Andrei, E. Y. Chern insulators, van Hove singularities and topological flat bands in magic-angle twisted bilayer graphene. *Nat. Mater.* **20**, 488–494 (2021).
15. Tomarkovitz, M. et al. Electronic compressibility of magic-angle graphene superlattices. *Phys. Rev. Lett.* **123**, 046601 (2019).
16. Utama, M. I. B. et al. Visualization of the flat electronic band in twisted bilayer graphene near the magic angle twist. *Nat. Phys.* https://doi.org/10.1038/s41567-020-0997-x (2020).
17. Jiang, Y. et al. Charge order and broken rotational symmetry in magic-angle twisted bilayer graphene. *Nature* **573**, 91–95 (2019).
18. Kerelsky, A. et al. Maximized electron interactions at the magic angle in twisted bilayer graphene. *Nature* **572**, 95–100 (2019).
19. Cho, Y. et al. Electronic correlations in twisted bilayer graphene near the magic angle. *Nat. Phys.* **15**, 1174–1180 (2019).
20. Dong, D. et al. Cascade of electronic transitions in magic-angle twisted bilayer graphene. *Nature* **582**, 198–202 (2020).
21. Uri, A. et al. Mapping the twist-angle disorder and Landau levels in magic-angle graphene. *Nat. Phys.* **581**, 47–52 (2020).
22. Zeng, D. et al. Cascade of phase transitions and Dirac revivals in magic-angle graphene. *Nature* **582**, 203–208 (2020).
23. Dean, C. R. et al. Boron nitride substrates for high-quality graphene electronics. *Nat. Nanotechnol.* **5**, 722–726 (2010).
24. Li, G., Luican, A. & Andrei, E. Y. Self-navigation of a scanning tunneling microscope tip toward a micron-sized graphene sample. *Rev. Sci. Instrum.* **82**, 073701 (2011).
25. Kouwenhoven, L. P., Schön, G. & Sohn, L. L. in *Mesoscopic Electron Transport* (eds Sohn, L. L., Kouwenhoven, L. P. & Schön, G.) 1–44 (Springer, 1997).
26. Li, G. & Andrei, E. Y. Observation of Landau levels of Dirac fermions in graphite. *Nat. Phys.* **3**, 623–627 (2007).
27. Li, G., Luican, A. & Andrei, E. Y. Scanning tunneling spectroscopy of graphene on graphite. *Phys. Rev. Lett.* **102**, 176804 (2009).
28. Allen, M. T., Martin, J. & Yacoby, A. Gate-defined quantum confinement in suspended bilayer graphene. *Nat. Commun.* **3**, 934 (2012).
29. Li, S.-Y. et al. Nanoscale detection of valley-dependent spin splitting around a magic-angle graphene quantum dot with orbital and valley splittings. *Nano Lett.* **16**, 5798–5805 (2016).
30. Jung, S. et al. Evolution of microscopic localization in graphene in a magnetic field from scattering resonances to quantum dots. *Nat. Phys.* **7**, 245–251 (2011).
31. Ando, T. Screening effect and impurity scattering in monolayer graphene. *J. Phys. Soc. Jpn.* **75**, 074716 (2006).
32. Nomura, K. & MacDonald, A. H. Quantum transport of massless dirac fermions. *Phys. Rev. Lett.* **98**, 076602 (2007).
33. Chen, J.-H., Jang, C., Xiao, S., Ishigami, M. & Fuhrer, M. S. Intrinsic and extrinsic performance limits of graphene devices on SiO2. *Nat. Nanotechnol.* **3**, 206–209 (2008).
34. Martin, J. et al. Observation of electron–hole puddles in graphene using a scanning single-electron transistor. *Nat. Phys.* **4**, 144–148 (2008).
35. Singh, B. R. Oxidation of silicon in the presence of chlorine and chlorine compounds. *J. Electrochem. Soc.* **125**, 453–461 (1978).
36. Luican, A., Li, G. & Andrei, E. Y. Quantized Landau level spectrum and its density dependence in graphene. *Phys. Rev. B* **83**, 041405 (2011).
37. Luican, A. et al. Single-layer behavior and its breakdown in twisted graphene layers. *Phys. Rev. Lett.* **106** https://doi.org/10.1103/PhysRevLett.106.126802 (2011).
38. Andrei, E. Y., Katzir, A. & Suss, J. T. Point defects in hexagonal boron nitride. III. EPR in electron-irradiated BN. *Phys. Rev. B* **13**, 2831–2834 (1976).
39. Wong, D. et al. Characterization and manipulation of individual defects in insulating hexagonal boron nitride using scanning tunnelling microscopy. *Nat. Nanotechnol.* **10**, 949–953 (2015).
40. Yu, Y.-J. et al. Tuning the graphene work function by electric field effect. *Nano Lett.* **9**, 3340–3344 (2009).
41. Hölzl, J. & Schulte, F. K. in *Solid Surface Physics* (eds Hölzl, J., Schulte, F. K. & Wagner, H.) 1–150 (Springer, 1979).
42. Velascos, J. et al. Visualization and control of single-electron charging in bilayer graphene quantum dots. *Nano Lett.* **18**, 5104–5110 (2018).
43. Altvater, M. A. et al. Electrostatic imaging of encapsulated graphene. *2D Mater.* **6**, 045034, https://doi.org/10.1088/2053-1583/ab254e (2019).

**Acknowledgements**

N.T. and X.L. acknowledge support from the U.S. DOE-BES grant DOE-FG02-99ER45742. S.W. and E.Y.A. acknowledge support from the Gordon and Betty Moore Foundation’s EPiQS initiative grant GBMP4543. M.X. and P.C.C. were supported by the U.S. DOE-BES Division of MDE and the research was performed at the Ames Laboratory which is operated for the U.S. DOE by ISU under Contract No. DE-AC02-07CH11358. R.A.R. was supported by the Gordon and Betty Moore Foundation’s EPiQS initiative grant GBMP4411.

**Author contributions**

X.L. and N.T. fabricated and characterized the STM device. S.W. and Z.Z. fabricated and characterized the planar tunneling device. M.X., R.A.R., and P.C.C. grew the hBN crystal used to make the devices. N.T. and E.Y.A. wrote the paper with inputs from all authors. E.Y.A. supervised the research.

**Competing interests**

The authors declare no competing interests.

**Additional information**

*Supplementary information* The online version contains supplementary material available at https://doi.org/10.1038/s41467-021-24480-3.

**Correspondence** and requests for materials should be addressed to E.Y.A.

**Peer review information** *Nature Communications* thanks Yuan Cao, and the other, anonymous, reviewer(s) for their contribution to the peer review of this work. Peer reviewer reports are available.

**Reprints and permission information** is available at http://www.nature.com/reprints

**Publisher’s note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This article is licensed under a *Creative Commons Attribution 4.0 International License*, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2021