
Supplementary information

Observation of flat bands in twisted bilayer graphene

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Observation of flat bands in twisted bilayer graphene

Simone Lisi,¹ Xiaobo Lu,² Tobias A. de Jong,³ Petr Stepanov,² Jose R. Duran,² Florian Margot,¹ Irène Cucchi,¹ Edoardo Cappelli,¹ Andrew Hunter,¹ Anna Tamai,¹ Viktor Kandyba,⁴ Alessio Giampietri,⁴ Alexei Barinov,⁴ Johannes Jobst,³ Vincent Stalman,³ Maarten Leeuwenhoek,^{3,5} Kenji Watanabe,⁶ Takashi Taniguchi,⁶ Louk Rademaker,⁷ Sense Jan van der Molen,³ Milan Allan,³ Dmitri K. Efetov,² and Felix Baumberger^{*1,8}

¹Department of Quantum Matter Physics, University of Geneva,
24 Quai Ernest-Ansermet, 1211 Geneva 4, Switzerland

²ICFO – Institut de Ciències Fotoniques, The Barcelona Institute of Science and Technology, Castelldefels, Barcelona, Spain

³Huygens-Kamerlingh Onnes Laboratory, Leiden Institute of Physics,
Leiden University, Niels Bohrweg 2, 2333 CA Leiden, The Netherlands

⁴Elettra-Sincrotrone Trieste S.C.p.A., Basovizza, 34149 Trieste, Italy

⁵Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628CJ Delft, The Netherlands

⁶National Institute for Materials Science, 1-1 Namiki, Tsukuba, 305-0044, Japan

⁷Department of Theoretical Physics, University of Geneva,
24 Quai Ernest-Ansermet, 1211 Geneva 4, Switzerland

⁸Swiss Light Source, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

I. SUPPLEMENTARY INFORMATION

A. Fabrication

Supplementary Fig. 1 illustrates the different steps of the device fabrication described in the methods section of the main paper.

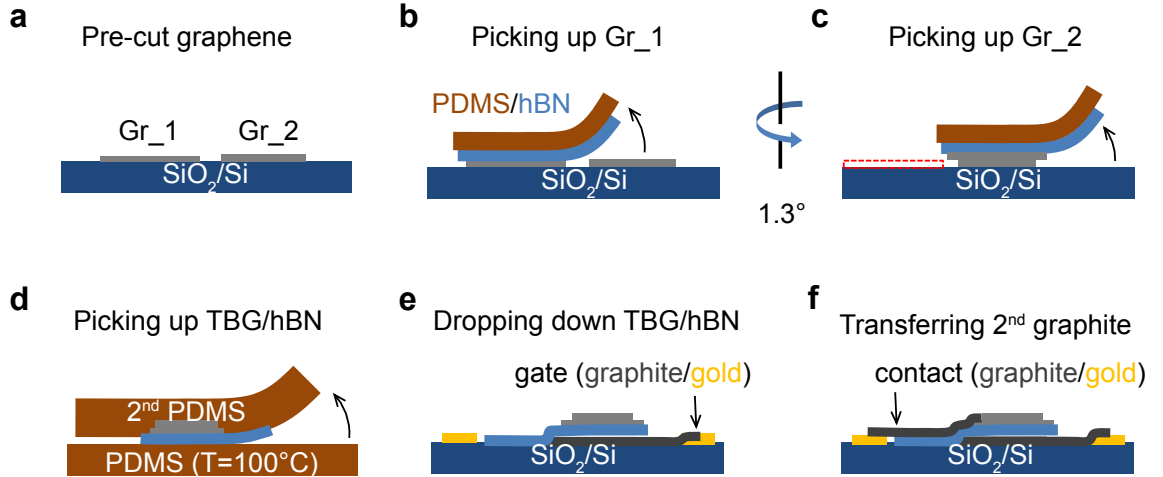


FIG. 1: Schematic of the device fabrication process. a) A graphene flake exfoliated on Si/SiO₂ is cut with an AFM tip in two mutually aligned flakes. b,c) The first graphene flake is picked up with pre-exfoliated hBN on a PDMS stamp, twisted manually by 1.3°, and then, used to pick up the second graphene flake. d) The heterostructure is flipped over and picked up with a second PDMS stamp. e) The heterostructure is transferred on top of a graphite flake connected with the gate electrode; e) a second graphite flake is stamped to connect the TBG to the ground electrode.

* Email address: felix.baumberger@unige.ch

B. ARPES data

In supplementary Fig. 2 we show the raw un-symmetrized ARPES constant energy contours together with same calculations shown in the main text.

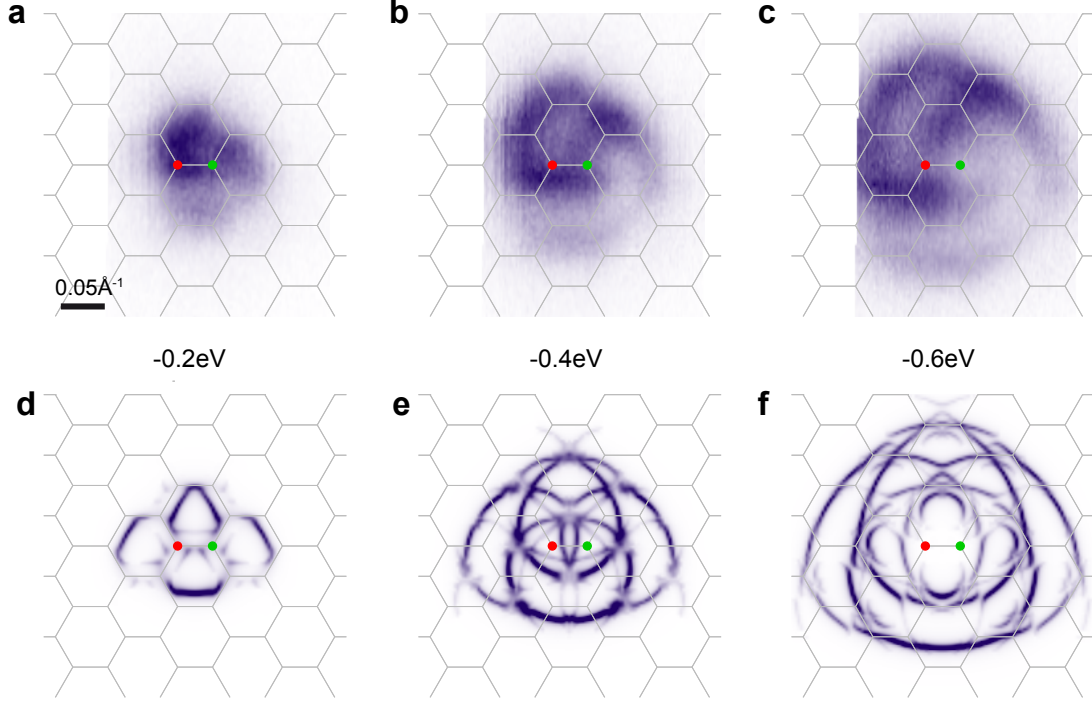


FIG. 2: a-c) Raw ARPES constant energy contours. d-f) Calculations of the spectral weight within the continuum model described in section D. A Lorentzian broadening of 10 meV has been applied to the calculation.

C. Scanning tunneling microscopy

Supplementary Fig. 3 shows two topographic images of the TBG surface taken $1 \mu\text{m}$ apart. The Fourier transforms of the images used to determine the twist angles, as described in the methods section, are shown as insets.

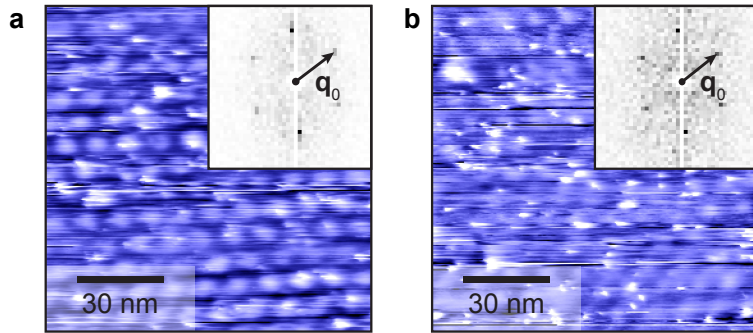


FIG. 3: a, b) STM topographies of the TBG surface are shown together with their respective Fourier transform (insets). The arrows in the insets indicate the reciprocal moiré wave vector used to determine the twist angle. Both images were acquired with bias voltage $V_b = 400 \text{ mV}$ and tunneling current $I = 20 \text{ pA}$.

D. Theoretical calculations

The theoretical ARPES intensity is calculated using the continuum model of twisted bilayer graphene[2, 3]. Below, we briefly outline this model. An untwisted graphene layer will have lattice vectors $\mathbf{a}_1 = a(1, 0)$ and $\mathbf{a}_2 = a(1/2, \sqrt{3}/2)$ with $a = 0.246$ nm. The reciprocal lattice vectors are $\mathbf{G}_1 = \frac{2\pi}{a}(1, -1/\sqrt{3})$ and $\mathbf{G}_2 = \frac{2\pi}{a}(0, 2/\sqrt{3})$. The rotated layers have reciprocal lattice vectors $\mathbf{G}_{\ell,i} = R(\pm\theta/2) \cdot \mathbf{G}_i$, and the reciprocal moiré vectors can be found as follows

$$\mathbf{G}_i^M = R(-\theta/2) \cdot \mathbf{G}_i - R(+\theta/2) \cdot \mathbf{G}_i \quad (1)$$

The moiré unit cell lattice vectors can be found from the relation $\mathbf{L}_i^M \cdot \mathbf{G}_j^M = 2\pi\delta_{ij}$. The Moiré lattice constant is $|\mathbf{L}_i^M| = \frac{a}{2\sin\theta/2}$. The K and K' points are given by $\mathbf{K}_\ell^{(\xi)} = -\xi(2\mathbf{G}_{\ell,1} + \mathbf{G}_{\ell,2})/3$, where $\xi = \pm 1$ denotes the valley index. The continuum model Hamiltonian per valley ξ can be written as a 4×4 matrix in the sublattice space (A_1, B_1, A_2, B_2) ,

$$H^{(\xi)} = \begin{pmatrix} H_1 & U^\dagger \\ U & H_2 \end{pmatrix}. \quad (2)$$

For the intralayer Hamiltonian we used the standard monolayer graphene nearest-neighbor hopping model, with $t = 2.90$ eV hopping. The Hamiltonian matrix is

$$H_\ell = -t \begin{pmatrix} 0 & \gamma_\ell(\mathbf{k}) \\ \gamma_\ell^*(\mathbf{k}) & 0 \end{pmatrix} \quad (3)$$

where $\gamma_\ell(\mathbf{k}) = \sum_{i=1}^3 e^{-i\mathbf{k}\delta_\ell^{(i)}}$ where $\delta_\ell^{(i)}$ are the vectors connecting the A to the B lattice sites in layer ℓ .

The matrix U is the effective interlayer coupling between the twisted layers

$$U = \begin{pmatrix} u & u' \\ u' & u \end{pmatrix} + \begin{pmatrix} u & u'\omega^{-\xi} \\ u'\omega^\xi & u \end{pmatrix} e^{i\xi\mathbf{G}_1^M \cdot \mathbf{r}} + \begin{pmatrix} u & u'\omega^\xi \\ u'\omega^{-\xi} & u \end{pmatrix} e^{i\xi(\mathbf{G}_1^M + \mathbf{G}_2^M) \cdot \mathbf{r}}. \quad (4)$$

The interlayer coupling parameters are set to $u' = 110$ meV and $u = 0.8u'$. The parameters u, u' are unequal to model the lattice relaxation that favors AB stacking over AA stacking.

Because the interlayer coupling U allows for scattering between neighboring moiré Brillouin zones, we include several such small Brillouin zones around the K or K' points of the original graphene layers up to a cut-off distance of $\sim 5|\mathbf{G}^M|$. The Bloch eigenstates are given by a sum over all these mini-Brillouin zones, $\psi_{n\mathbf{k}}(\mathbf{x}) = \sum_{\mathbf{G}} c_{n\mathbf{k}}^{\mathbf{G}} e^{i(\mathbf{k}+\mathbf{G})\mathbf{x}}$. To obtain the ARPES intensity of a given Bloch eigenstate, following Refs. [4, 5], we sum $|c_{n\mathbf{k}}^{\mathbf{G}=0}|^2$ over all the four sublattices. To visualize the intensity, we broaden the spectral points with a Lorentzian of width $\gamma = 10$ meV. All calculations shown in the main text are performed for a twist angle $\theta = 1.34^\circ$ in valley $\xi = 1$.

Supplementary Figs. 3 and 4 show a superimposition of contours of all the states computed in our model (red lines) and the calculated spectral weights that are directly compared with the ARPES data in the main text.

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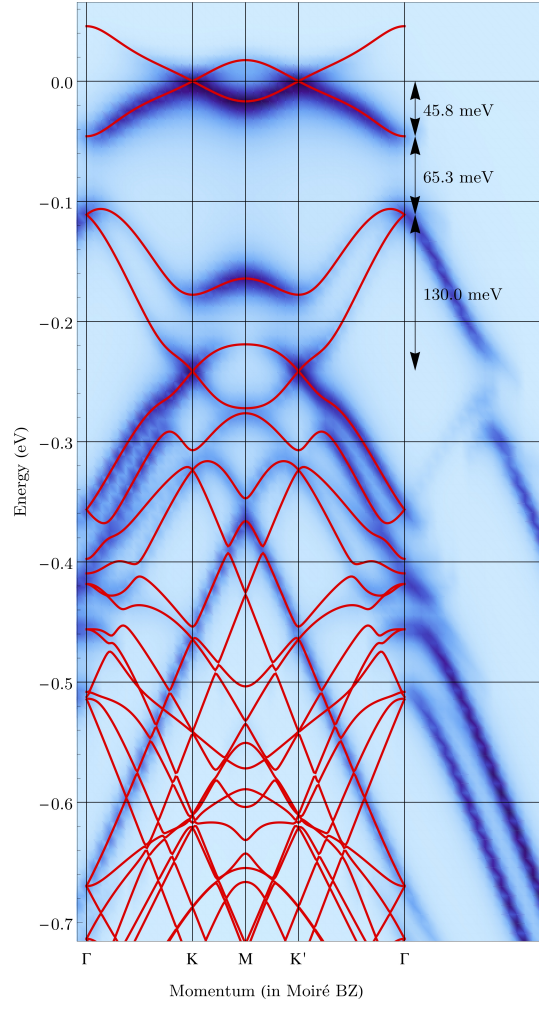


FIG. 4: TBG band structure in the continuum model along high-symmetry lines of the moiré mini-Brillouin zone superimposed on spectral weights as they are presented in Fig. 4 of the main text.

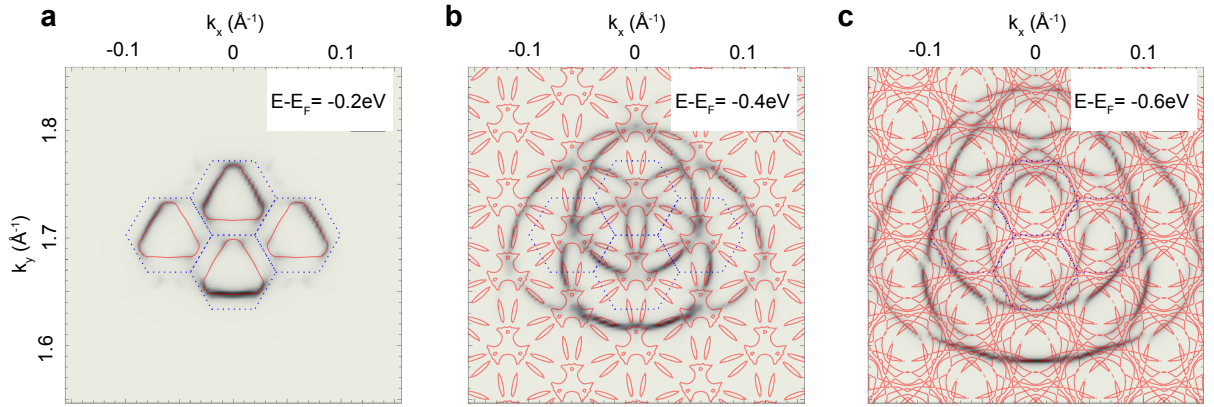


FIG. 5: Constant energy contours of eigenvalues superimposed on the spectral weights presented in Fig. 3 of the main text.