Supporting Information

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Observation of Electrically Tunable van Hove Singularities in Twisted Bilayer Graphene from NanoARPES

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Supplementary Note 1. Electronic structure maps of the device

Figure S1a - Figure S1d present real space maps composed from the integrated $(E,k)$-intensity indicated by correspondingly colored boxes in the ARPES spectra shown in Figure S1e - Figure S1h. Each map presents a distinct component of the device. Likewise, the ARPES spectra were averaged over the areas demarcated by boxes in Figure S1a - Figure S1d. Clean twisted bilayer graphene (twBLG) areas are characterized by two sharp Dirac cones (see panels a and e). The polycrystalline gold contacts are easily identified from the homogeneous intensity seen in panels b and f. The valence band maximum of the hBN is not observed in the relevant regions in panels c and g, possibly because the dispersion is not aligned with our detection window. We find that some spots within the twBLG flake display spectra of much poorer quality (see panel h). These are identified as having a lower intensity in the map as seen in panel a and marked by red dots in panel d. These spots likely contain trapped dirt or bubbles from the fabrication process and are avoided in the further detailed analysis.

![Supplementary Note 1. Electronic structure maps of the device](image)

Figure S1. $(E,k,x,y)$-dependent photoemission intensity. a) - d) $(x,y)$-dependent ARPES intensity maps corresponding to the spectral features marked with coloured boxes in (e)-(h). e) - h) ARPES intensity obtained by integrating the signal within the colored outlines in (a)-(d). Note that the square boxes in (c) indicate areas selected for the further analysis in Figure S2. The data were taken at a photon energy of 60 eV.
We explore the spatial homogeneity of the sample by inspecting detailed nanoARPES maps collected from the areas marked by square boxes in Figure S1c and presented in detail in Figure S2a - Figure S2c. Within the detailed maps, we select six (0.5 \( \mu \text{m} \times 0.5 \mu \text{m} \)) regions and investigate the spatial dependence of the \( k \)-separation (\( \Delta K \)) between the Dirac points of the two graphene layers and of the linewidth quantified via the full width at half maximum (FWHM) of momentum distribution curves (MDCs) (see sketches in Figure S2d). The spectra from the correspondingly colored areas are presented in Figure S2e - Figure S2j. In panel f we see an additional weak branch appearing near the top cone (see green arrow and dashed line), signaling that two slightly different rotational domains are captured within our beam spot size (see green square in panel a). A variation of linewidth is also observed with the most dramatic increase in width shown by purple arrows in Figure S2h, attributed to a lower quality of the sample in the area of the map marked by a filled purple box in Figure S2b.

The extent of these changes is quantified by extracting MDCs along the directions (labeled \( y \)) marked by open rectangles in panels a-c. The MDCs taken at the Fermi level provide \( \Delta K \) via MDC fits using a function containing two Lorentzian peaks and a linear background (see fitted peak positions in Figure S2k). The twist angle is calculated from the resulting values of \( \Delta K \) using the expression \( \theta = 2\arcsin \left( \frac{\Delta K}{2|\vec{K}|} \right) \), where \( |\vec{K}| = 1.7 \text{ Å}^{-1} \) (see sketch in Figure S2d). The spatial dependence of \( \theta \) shown in Figure S2l reveals a mean value of 12.2\(^\circ\), but microscopic domains are observed with a range of angles between 11.5\(^\circ\) - 13.0\(^\circ\). The FWHM values of corresponding MDC fits carried out at a binding energy of 0.5 eV are displayed in Figure S2m, summarizing the position dependent linewidth broadening.
Figure S2. Spatial dependence of electronic states and twist angle. a) - c) Detailed ($x, y$)-dependent ARPES maps of the regions indicated by color-coded boxes in Figure S1c. The intensity scale was obtained by integrating the signal across the entire spectrum. d) Schematic representation of the relationship between twist angle $\theta$, Dirac point separation $\Delta K$ between the two layers, and indication of MDC full width at half maximum (FWHM). e) - j) ARPES spectra from the correspondingly coloured ($0.5 \, \mu m \times 0.5 \, \mu m$) regions in (a)-(c). k) - m) Spatial dependence of (k) MDC peak positions obtained at $E_F$, (l) of the twist angle and (m) of the average FWHM for each linear branch of the two Dirac cones at a binding energy of 0.5 eV. The curves and markers have been color-coded according to the color of the open rectangles in (a)-(c), which indicate the line profiles where the MDCs were obtained.
Supplementary Note 2. MDC analysis of twBLG Dirac cones

Here we provide details on MDC fits of the twBLG Dirac cones for the binding energy range of 0.4 eV enclosed by dotted lines in Figure S3a. The fits were performed for each gate voltage using a function composed of four Lorentzian components (one per branch of each cone) and a linear background, leading to an excellent description of the data as shown in Figure S3b. The fits provide $k_F$ as sketched in Figure S3a, leading to the $V_g$-dependence shown in Figure S3c. The fitted peak positions also demonstrate the linear dispersion as seen in Figure S3d with the band velocities given in Figure S3e and FWHM values displayed in Figure S3f.

Figure S3. MDC analysis at different gate voltages. a) ARPES spectrum for $V_g = -9$ V. The horizontal dotted lines demarcate the binding energy range where the MDCs are extracted. The arrows denote the Fermi wavevector for top (red) and bottom (blue) layers, respectively. The dotted lines represent the fitted dispersion. b) MDCs (colored markers) with fits (black curves) consisting of four Lorentzian components on a linear background, shown for all measured gate voltages. c) $k_F$ as a function of gate voltage. d) Peak positions (markers) of the four Lorentzian functions fitted in (b) along with linear dispersion fits (black lines). e) Band velocity as a function of doping. f) FWHM of the Lorentzian peaks for each gate voltage.
Supplementary Note 3. EDC fits of van Hove singularities

EDCs used to determine the binding energy of the van Hove singularities as a function of gate voltage are extracted from the cuts along the given high symmetry lines in the mini BZ in Figure S4a - Figure S4c. The EDCs are shown for each gate voltage in Figure S4d - Figure S4f along with fits to a function composed of two Lorentzian peaks on a linear background. The peak positions provide the binding energy of the van Hove singularity and band maximum below the mini gap.

Figure S4. EDC analysis at different gate voltages. a) - c) ARPES spectra for $V_g = -9$ V along the given directions of the mini BZ. The dashed lines correspond to the single EDCs through the van Hove singularities. In (a), the image was taken near the edge of our acquisition region. d) - f) EDCs extracted along the vertical dashed lines shown in (a)-(c) (colored markers) along with fits to a function consisting of two Lorentzian components on a linear background, shown for all measured gate voltages. Linear fits to peak positions are indicated by blue and green lines and are intended to guide the eye only.
Supplementary Note 4. Estimate of nanoARPES spatial resolution

The spatial resolution can be estimated by assuming that the measured features are much sharper than our beam profile. To do so, we investigate several sharp boundaries in the ARPES maps, as presented in Figure S5a - Figure S5d. We fit a step function convoluted by a Gaussian to the line profile across the edge (see insets). Upon analysis of multiple such profiles, we find that the full width half maximum of our beam is $\Delta s = (690 \pm 80) \text{ nm}$.

Figure S5. Line profile analysis. a) - d) $(x,y)$-dependent ARPES maps around the edges of different device components. Insets: line profiles (open circles) taken along the direction indicated by the red arrow, together with the fit curves (blue lines) of a step function convoluted with a Gaussian beam profile.