The crystal structure of a material creates a periodic potential that electrons move through giving rise to the electronic band structure of the material. When two-dimensional materials are stacked, the twist angle between the layers becomes an additional degree freedom for the resulting heterostructure. As this angle changes, the electronic band structure is modified leading to the possibility of flat bands with localized states and enhanced electronic correlations\(^1-6\). In transition metal dichalcogenides, flat bands have been theoretically predicted to occur over a range of twist angles below ~7 degrees\(^4\). Here we show the existence of a flat band in the electronic structure of a 3° twisted bilayer WSe\(_2\) sample using scanning tunneling spectroscopy. Direct spatial mapping of wavefunctions at the flat band energy have shown that the flat band is localized in the form of a hexagonal network in excellent agreement with first-principle density functional theory calculations\(^4\).
The formation of moiré superlattices (MSL) between two layers of van der Waals materials, with either a twist angle or lattice mismatch between them can dramatically alter the band structure and hence their electronic properties. Novel electronic states, such as replica Dirac cones and the Hofstadter butterfly pattern have been observed in graphene/hexagonal boron nitride (hBN) heterostructures. In a certain range of small twist angles, the folding of the band structure into a mini-Brillouin zone can form flat bands, giving rise to the localization of electronic states and the enhancement of electron-electron interactions. When the Coulomb potential surpasses the kinetic energy of the band electrons, strong correlated states such as unconventional superconductivity and Mott insulating behavior can arise from the flat bands. Recent transport experiments revealed these states for both magic-angle twisted bilayer graphene (MATBG) and aligned trilayer graphene/hBN heterostructures.

Apart from graphene-based MSL, twisted bilayer transition metal dichalcogenides (tTMDs) are also predicted to host flat bands at small twist angles for both homobilayers and heterobilayers. In contrast to MATBG, where the flat bands only forms within ±0.2° around a magic angle, in tTMDs flat bands are predicted to form over a wide range of angles and their band width monotonically decrease with the twist angle, which allows a more versatile platform for the design of flat band devices. For heterobilayer TMDs, recent scanning tunneling experiments have shown quantum-confined states related to the sharp peaks around band edges for aligned MoS2-WSe2 heterobilayers. For homobilayer TMDs, optical studies have shown band gap variations due to stacking effects in twisted bilayer MoS2. However, a direct measurement of moiré flat bands and localized states in the small angle regime (0° ≤ θ ≤ 7°) for tTMDs have not been performed yet. In this study we present local characterization of the 3° twisted bilayer WSe2 via scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS). We observe spectroscopic signatures of a flat band around the valence band edge of tWSe2 and directly image the localized nature of this flat band.

An optical image of the device and a schematic of our experimental setup are shown in Figs. 1a, b. All measurements were performed in ultra-high vacuum at a temperature of 4.6 K. The sample was fabricated by a dry transfer technique with controlled rotational alignment between the two layers of WSe2. The tWSe2 sits on a bilayer graphene (BLG) flake to provide a conducting substrate for collecting the tunneling current from the STM tip. A hBN flake partially covers the tWSe2 in order to clamp it down and prevent it from rotating from the designed twist angle. Fig. 1c shows the STM topography of the sample, both a graphene-hBN moiré (~11 nm) and the tWSe2 moiré (~6nm) are visible. The graphene-hBN moiré arises from the BLG and the bottom hBN having only a small twist angle between them. In order to extract the precise local twist angle and identify the different stacking configurations of the tWSe2, we filtered out the graphene-hBN moiré with a low-pass filter in Fig. 1d. The high symmetry points are labeled as AA, AB,
BA and Br, where AA means eclipsed stacking with W atom over W atom, Se atom over Se atom; AB (not to be confused with 2H or Bernal stacking in other literatures) means staggered stacking with W over Se; BA means staggered stacking with Se over W; Br means the bridge that connects neighboring AA sites. Applying an uniaxial heterostrain model19 and using the closest distance between the AA sites in 3 directions (L1, L2, L3) as input parameters, we find a twist angle $\theta = 3.00^\circ$ and a uniaxial strain $\varepsilon = 0.42\%$ (See Supplementary Information for details about the twist angle and strain determination). This is in excellent agreement with the designed twist angle of 3°. Fig. 1e illustrates the top view and the side view of the different high symmetry stackings that are identified in the tWSe2 MSL.

Fig. 2a shows the constant height STS measurements of the local density of states (LDOS) taken on the 4 different high symmetry sites shown in Fig. 1d. For each of these measurements the tip height was stabilized at a bias voltage of -2.4 V and tunnel current of 100 pA. Then the feedback circuit was switched off, a small ac voltage (10 mV) was applied to the bias voltage and the differential conductance $dI/dV$ was measured as a function of bias voltage using lock-in detection. The constant height $dI/dV$ shows a band gap of 2.2eV for the AA site and 2.1eV for all the other sites, an energy difference which is mainly contributed by a valence band edge shift of ~80 meV. While the constant height $dI/dV$ can accurately measure the LDOS around the $\Gamma$-point in the center of the Brillouin zone, it fails to detect other states with a large parallel momentum, such as the states that near the K-point29 (See Supplementary Information for details about Brillouin zone location assignments). This can be overcome by employing a constant current spectroscopy method, where the tip height is adjusted by a feedback loop when the effective tunnel barrier changes due to higher parallel momentum. Fig. 2b shows the constant current STS measurements of the critical points in the valence band for the 4 high symmetry sites. For all of these measurements the tunnel current was fixed at 10 pA, and the differential conductance was measured. While the peak at -1.9V aligned well for all the locations, the features around the band edge shows significant location dependence. The most striking feature are the sharp peaks present at the AB, BA and bridge sites, indicating evidence of flat bands in this type of system as predicted by theory4,6.

To better identify the locations of the critical points at different high symmetry sites, we fit each constant current $dI/dV$ curve with Lorentzian functions (see Fig. 2c for an example and Supplementary Information for all fits) and compare their peak positions in Fig 2d. For each $dI/dV$ curve there are a set of 3 closely spaced peaks between -1.0 V and -1.2 V and another isolated peak near -1.9 V. We first notice that the $\Gamma$ point for the AA site is shifted down by ~0.2 V compared with other high symmetry sites, which is consistent with the constant height $dI/dV$ measurements. Other bands only show small shifts between different stackings. There is also a small splitting between the first two Q points in the conduction band and a bigger splitting between the first two K points in the valence band. These splittings are due to the lack of
inversion symmetry of the sample, since the in-plane dipole momentum of the top and bottom layers of WSe$_2$ are almost parallel to each other. While such a splitting is not present in the inversion symmetric 2H-stacked bilayer WSe$_2$ where the in-plane dipole momentum of the two layers are anti-parallel$^{30}$. The observed spin-orbit-induced splitting (~130meV) in 3° tWSe$_2$ is comparable to the calculated spin-orbit splitting in isolated AA stacked bilayer WSe$_2$ (100 ~ 140mV)$^4$.

To further study the location dependence of the critical points, we perform spatially resolved tunneling spectroscopy at constant current along a line (indicated in Fig. 3a) crossing all the high symmetry points. There are a set of states located between -1.0 V and -1.4 V that evolve with position as seen in Fig. 3b. The continuous shift of the $\Gamma_v$ point resembles the smooth transition between the high symmetry points in the MSL. The sharp peak around -1.1V due to the presence of the flat band shows only a slight spatial variation within the AB-Br-BA region, comparable to the small variation of the $K_v$ and the $\Gamma_2$ points. In contrast, this sharp peak is almost completely missing on the AA sites, indicating that the flat band is localized on the AB-Br-BA region and away from the AA sites.

We verify the localization of the flat band by scanning over the surface at a fixed bias voltage and acquiring the LDOS at constant current. Fig. 4a is the LDOS map at the flat band energy, in excellent agreement with the predicted wave function of the flat band for a relaxed ($\epsilon \geq 0.3\%$) MSL$^4$, it features a conductive hexagon enclosing the insulating AA region. On the other hand, LDOS maps at energies away from the flat band (Fig. 4b, c) show the AA regions as triangular bright spots indicating that at these energies the electrons are at the center of the unit cells. These energies correspond to the top of the first and second valence band at the $\Gamma$ point. The Supplementary Information contains additional LDOS images for all of the band edges identified in the experiment which show similar behavior to the $\Gamma$ point.

Our spectroscopic measurements have proved the existence of a moiré flat band in 3° tWSe$_2$ originating from the highest valence band at the $\Gamma$ point. In contrast to the MATBG, where the filled flat bands are localized on the AA sites$^{10,18,19,21}$, the filled flat band in 3° tWSe$_2$ is localized on the hexagonal network separating the AA sites. Our results match well with the theoretical predictions$^4$, and open up the possibility of probing flat bands in a vast family of small angle tTMDs. Future, gate-tunable experiments could reveal the phenomenology of correlated states in these systems, such as topological insulating states$^6$, when the flat bands are tuned to be partial filled.
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Competing Interests statement: The authors declare no competing interests.

Main Figure Legends:

Figure 1. Stacking configurations of the tWSe2. a, Optical image of the measured device. Blue and Gray dashed lines highlight the hBN and BLG flakes, red dashed lines mark the tWSe2 region. b, Schematic of the STM setup on the tWSe2 device. c, Atomic-resolution STM topography on the 3.00° tWSe2 sample, with a set bias voltage \( V_B = -2.5 \) V, set current \( I = 100 \) pA. d, A zoomed-in view of panel c with the graphene-BN moiré filtered out. e, Illustration of the different stacking configurations: AA, AB, BA. The blue and cyan colors denote the W atoms, while pink and yellow denote the Se atoms in the two layers.

Figure 2. dI/dV spectra on the 4 high symmetry points. a, Constant height mode dI/dV vs. bias voltage data, acquired at \( I =100 \) pA. b, Constant current mode dI/dV vs. bias voltage data, acquired at \( I =10 \) pA. c, Constant current dI/dV vs. bias voltage data measured at the AA point (black), along with fitting functions (red). d, Bias voltage values at fitted peak positions at the four high symmetry points (left panel). and a zoom-in plot around valence band edge (right panel).

Figure 3. Line cut dI/dV spectra. a, Illustration of the line along where the dI/dV spectra was taken. b, Constant current dI/dV spectra line cut, acquired at \( I = 10 \) pA. The dashed vertical lines label the location of high symmetry sites.

Figure 4. Spatially resolved LDOS at different energy. a, LDOS maps at the flat band energy, acquired at \( I = 10 \) pA. b and c, LDOS maps at the \( \Gamma \) energies that are away from the flat band, acquired at \( I = 10 \) pA.
Methods

Sample Fabrication

Our tWSe$_2$ sample was fabricated by sequential pickup steps using a hemispherical handle substrate with rotational control. Starting with a large single grain monolayer WSe$_2$ trimmed into two separate sections by plasma etching, we sequentially picked up the two sections by a BLG-hBN heterostructure attached to the hemispherical handle, with the BLG in direct contact to tWSe$_2$. Between the first, and second WSe$_2$ section pick-up, the substrate was rotated by 3° to create the tWSe$_2$. Another hBN flake partially covering the tWSe$_2$ was subsequently picked up in order to clamp the tWSe$_2$ down and prevent rotation from the designed twist angle. The stacking structure was then placed on a SiO$_2$/Si substrate with tWSe$_2$ and hBN clamp on top. Metal electrodes were defined and deposited with Ni/Au to complete the device.

STM Measurements

STM/STS measurements were performed in the ultrahigh-vacuum LT-STM (Omicron) operating at 4.6K. dI/dV spectroscopies were acquired by adding a 10mV a.c. voltage at 617Hz to the bias voltage and measuring the current with lock-in detection. dI/dV spectroscopy was performed in two different modes, a constant height mode where tip height was stabilized at a particular bias voltage and the feedback circuit was turned off while ramping the bias voltage. A second mode was a constant current mode, where the current feedback was left on while the bias voltage changed allowing the tip to change its height. Electrochemically etched tungsten tips were used for imaging and spectroscopy. All the tips were first checked on an Au surface to ensure that they had the proper work function based on the decay of the tunnel current with distance from the sample. In addition, dI/dV spectroscopy was performed on the Au surface to ensure that the tip had a constant density of states.

Data Availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.
Figure 1

(a) Image showing the layers of materials: hBN, tWSe$_2$, BLG, and SiO$_2$. The layers are labeled with Ni/Au markers.

(b) Schematic diagram showing the alignment of materials and the application of bias ($V_{\text{Bias}}$).

(c) High-resolution image showing the atomic structure at a scale of 5nm.

(d) Detailed atomic-level view with labels L1, L2, L3, and atomic configurations AA, AB, BA.

(e) Schematics of AA, AB, and BA atomic configurations.
Figure 2

(a) Graph showing various bias voltages (V) with labels for $\Gamma_2$, $\Gamma_v$, $Q_c$, and $K_C$. The graph plots $(dI/dV)$ in arbitrary units (a.u.).

(b) Graph similar to (a) with additional labels for AA, AB, Br, and BA.

(c) Graph showing $(dI/dV)$ for bias voltages with labels for $\Gamma_2$, $\Gamma_v$, and $K_v$.

(d) Graphs showing $(dI/dV)$ peak position with markers for $K_C$, $K_v$, $\Gamma_2$, $Q_2$, $\Gamma_v$, $Q_c$, and $K_2$.
Figure 4
Supplementary Information

Flat bands in small angle twisted bilayer WSe₂

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1. Determination of local twist angle and strain

Fig. S1 | Filtering of the topography. a, Atomic-resolution STM topography on the 3° tWSe₂ sample, with a set bias voltage VB = -2.5 V and set current I = 100 pA. b, Fourier transform of panel a. c, Filtered Fourier transform, black area at the center indicates the components being removed. d, Topography image after the short-pass filter was applied, white dashed lines indicate the distance between nearest-neighbor AA sites in 3 different directions.

In order to properly determine the twist angle and strain of the tWSe₂ sample, we need to remove the effect of the moiré pattern due to the bilayer graphene and hexagonal boron nitride layers underneath. These two layers are aligned such that they create an ~6 nm moiré pattern. We apply a spatial short-pass filter by removing all the features with wavelength longer than 9.5 nm from the Fourier transform of the topography.
image. Fig. S1a is the original STM topography taken with a setpoint voltage of $V = -2.5$ V and current of $I = 100$ pA. We have performed an FFT on this image to obtain the wavevectors present in the image and the results are displayed in Fig. S2b. The resulting FFT is filtered to remove the long wavelength components which occur at the center of the image as shown by the black circle in Fig. S1c. The inverse Fourier transform then leads to the image shown in Fig. S2d, which contains only the moiré pattern from the tWSe$_2$.

To determine the local twist angle and strain of the tWSe$_2$, we first fit the 3 nearest AA sites with Gaussian functions to obtain the coordinates of their centers and thus the shortest distance between them ($L_1$, $L_2$, $L_3$). Then we follow a uniaxial heterostrain model$^1$ to express $L_1$, $L_2$ and $L_3$ by:

$$L_1 = \frac{4\pi}{\sqrt{3}k}\left[\left(\cos \theta_T - \frac{\cos^2 \theta_S}{1+\varepsilon} - \frac{\sin^2 \theta_S}{1-\delta\varepsilon}\right)^2 + \left(\sin \theta_T + \frac{\sin \theta_S \cos \theta_S}{1+\varepsilon} - \frac{\sin \theta_S \cos \theta_S}{1-\delta\varepsilon}\right)^2\right]^{1/2}$$

$$L_2 = \frac{8\pi}{\sqrt{3}k}\left[\left(\cos \theta_T - \sqrt{3}\sin \theta_T - \frac{\cos^2 \theta_S - \sqrt{3} \sin \theta_S \cos \theta_S}{1+\varepsilon} - \frac{\sin^2 \theta_S + \sqrt{3} \sin \theta_S \cos \theta_S}{1-\delta\varepsilon}\right)^2 + \left(\sin \theta_T + \sqrt{3} \cos \theta_T - \frac{\sqrt{3} \sin^2 \theta_S - \sin \theta_S \cos \theta_S}{1+\varepsilon} - \frac{\sqrt{3} \cos^2 \theta_S + \sqrt{3} \sin \theta_S \cos \theta_S}{1-\delta\varepsilon}\right)^2\right]^{1/2}$$

$$L_3 = \frac{8\pi}{\sqrt{3}k}\left[\left(-\cos \theta_T - \sqrt{3}\sin \theta_T + \frac{\cos^2 \theta_S + \sqrt{3} \sin \theta_S \cos \theta_S}{1+\varepsilon} + \frac{\sin^2 \theta_S - \sin \theta_S \cos \theta_S}{1-\delta\varepsilon}\right)^2 + \left(-\sin \theta_T + \sqrt{3} \cos \theta_T - \frac{\sqrt{3} \sin^2 \theta_S + \sin \theta_S \cos \theta_S}{1+\varepsilon} - \frac{\sqrt{3} \cos^2 \theta_S - \sqrt{3} \sin \theta_S \cos \theta_S}{1-\delta\varepsilon}\right)^2\right]^{1/2}$$

Where $k$ is reciprocal wavevector of the unstrained WSe$_2$ lattice, $\theta_T$ is the twist angle between the two layers of WSe$_2$, $\theta_S$ is the angle between the direction of applied strain and the direction of one of the WSe$_2$ crystal lattice, $\varepsilon$ is the strain percentage, $\delta$ is the Poisson ratio of WSe$_2$ (estimated to be 0.19$^2$). We numerically solve for the three unknown optimization parameters $\theta_T$, $\theta_S$, and $\varepsilon$ that best fit the experimentally measured moiré wavelengths L1, L2 and L3. From Fig. S1c we obtained $L_1 = 6.51$ nm, $L_2 = 6.46$ nm, $L_3 = 6.05$ nm giving values of $\theta_T = 3.00^\circ$, $\theta_S = 17.74^\circ$, $\varepsilon = 0.42\%$. 






















2. Decay constant measurements and the assignment of band edges

Fig. S2 | Decay constant as a function of bias voltage at the high symmetry points for a, the valence band and b, the conduction band.

In the main text we have discussed that for \((\text{d}I/\text{d}V)_z\) it is difficult to detect states with a large parallel momentum, because the effective tunneling decay constant \(\kappa\) can be expressed by\(^3\):

\[
\kappa = \frac{2m\phi}{\hbar^2} + k_{\parallel}^2
\]

where \(m\) is the effective mass of the electrons, \(\phi\) is the effective work function for the STM junction, \(\hbar\) is the reduced Planck constant, and \(k_{\parallel}\) is the parallel momentum. Measuring the decay constants for different states allows them to be assigned to different band edges in momentum space. We measure the decay constant by directly measuring \(I(Z)\) where \(Z\) is the tip-sample separation. For each bias voltage the tip was first stabilized with a current setpoint of 10 pA, then the tip was gradually retracted by 5 Å while recording the current. The tunnel current exponentially depends on tip-sample separation \(I \propto e^{-2\kappa Z}\), allowing for the decay constant to be extracted. By fitting each \(I(Z)\) curve with an exponential function, we obtain the decay constant \(\kappa\) as a function of bias voltage, as shown in Fig. S2.

For the valence band (Fig. S2a), there is a small dip around -1.9V for all 4 high symmetry sites, indicating this state is at the \(\Gamma\) point in momentum space. At the valence band edge, on AB, Br and BA sites there is
a big dip along with two small peaks on both sides of the minimum, indicating the momentum space locations of the three states near the valence band edge are ordered as K-Γ-K. On the AA site, the decay constant starts to drop at -1.4V as the bias voltage is getting closer to the band edge, while for other sites the decay constant starts to drop at -1.2V, indicating that the state at Γ_v is shifted down in energy on AA site. Thus, we assign the highest three states in the valence band on AA site as K-K-Γ.

For the conduction band (Fig. S2b), there is a dip around 1.0V on all 4 high symmetry sites. We assign this state to the Q point because it has lower momentum than the K point, while the Γ point for the conduction band is located at much higher energy^4,5. The state that is around 1.3 V has no clear features in our decay constant measurements, this is because the K point and M^* point (defined as the midpoint between Γ point and the M point) in the conduction band are almost degenerate in energy^5, their contribution to the decay constant partially cancel each other. Thus, we assign the state around 1.3V as K_C + M^*_C and label it as just K_C for simplicity. The energy differences between our assigned band edges within the conduction band or valence band agrees well with the theory calculation^5.

3. (dI/dV)_I and (dZ/dV)_I fitting details

In the main text Figure 2d, we have presented the fitted peak positions for the measured (dI/dV)_I spectroscopies at different high symmetry sites (AA, AB, Br and BA), the details of the fittings are shown in Fig. S3a and Fig. S4a for the valence band and the conduction band respectively. For each of the valence band curves, we have fit the spectra with four Lorentzian functions corresponding to the two upper most band edges at the K and Γ points. For the conduction band, we have used three Lorentzian functions for the lowest two band edges at the Q point and the lowest band at the K point. The middle panel shows the measured data (solid black curves) and the fitted curves (dashed red curves), the dashed green line shows a constant background from the fit. The top panel shows the fit residual, defined as the difference between the measured data and fitted curve. The Lorentzian fit curves for each individual peak are shown in the bottom panel.

An alternate way of identifying the band edges is a (dZ/dV)_I measurement, where the height signal as the input of the lock-in and dZ/dV is measured with the current feedback engaged. As the bias voltage is changed through a band edge, the density of states of the sample abruptly changes. With the feedback engaged, the tip must move to maintain the same tunnel current and therefore this is a peak in (dZ/dV)_I at the band edge. For all the (dZ/dV)_I measurements, the tunneling current was
fixed at 10pA and a 10mV ac voltage was applied to the bias voltage. Following the same method as we did with the \(\frac{dI}{dV}\) data, we fitted the \(\frac{dZ}{dV}\) data with Lorentzian functions. The details of the fittings are shown in Fig. S3b and Fig. S4b. Note that here we use a linear background to compensate for the Z movement due to the changing bias voltage. We plot the fitted peak locations versus high symmetry sites in Fig. S5, the position of the band edges and the energy separations between them are in excellent agreement with Fig. 2d in the main text obtained from \(\frac{dI}{dV}\) measurements.

**Fig. S3** | Fitting of the spectroscopies for the valence band. **a**, \(\frac{dI}{dV}\) fittings for 4 different high symmetry sites, top panel shows the fitting residuals; middle panel shows the measured curve (solid black line), fitted curve (dashed red line), and the background of the fitting (dashed green line); bottom panel shows the individual fit curves for each of the peaks. **b**, same as **a** except using the \(\frac{dZ}{dV}\) measurements.
Fig. S4 | Fitting of the spectroscopies for the conduction band. a, \((dI/dV)_{t}\) fittings for 4 different high symmetry sites, top panel shows the fitting residuals; middle panel shows the measured curve (solid black line), the fitted curve (dashed red line), and the background of the fitting (dashed green line); bottom panel shows the individual fit curves for each of the peaks. b, same as a except using the \((dZ/dV)_{t}\) measurements.

Fig. S5 | Bias voltage values of band edges from \((dZ/dV)_{t}\) data at the four high symmetry points (left panel), and a zoom-in plot around the valence band edge (right panel). The data is in agreement with the \((dI/dV)_{t}\) data presented as Fig. 2d in the main text.

4. Local density of state maps at other energies

In the main text, we have presented the local density of state maps for the energies of the flat band state, and the band edges at \(\Gamma_V\) and \(\Gamma_2\), here we show the local density of state maps for all the other assigned
band edges in Fig. 2D and Fig. S6. The peak in the local density of states occurs on different sites for different energies, due to band edge variations between the different stacking orders as shown in Fig. 3b of the main text. None of these images show the hexagonal network appearance seen for the flat band energy, indicating that the wavefunction of the flat band is distinctively different from the ordinary wavefunctions that arise from the moiré pattern.

**Fig. S2** | Local density of states maps at different energies. **a**, lowest energy band at Q in the conduction band. **b**, second band at Q in the conduction band. **c**, lowest band at K in the conduction band. **d**, highest band at K in the valence band. **e**, second highest band at K in the valence band.

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