Imaging moiré flat bands in three-dimensional reconstructed WSe$_2$/WS$_2$ superlattices

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Moiré superlattices in transition metal dichalcogenide (TMD) heterostructures can host novel correlated quantum phenomena due to the interplay of narrow moiré flat bands and strong, long-range Coulomb interactions$^{1-9}$. However, microscopic knowledge of the atomically reconstructed moiré superlattice and resulting flat bands is still lacking, which is critical for fundamental understanding and control of the correlated moiré phenomena. Here we quantitatively study the moiré flat bands in three-dimensional (3D) reconstructed WSe$_2$/WS$_2$ moiré superlattices by comparing scanning tunnelling spectroscopy (STS) of high-quality exfoliated TMD heterostructure devices with ab initio simulations of TMD moiré superlattices. A strong 3D buckling reconstruction accompanied by large in-plane strain redistribution is identified in our WSe$_2$/WS$_2$ moiré heterostructures. STS imaging demonstrates that this results in a remarkably narrow and highly localized K-point moiré flat band at the valence band edge of the heterostructure. A series of moiré flat bands are observed at different energies that exhibit varying degrees of localization. Our observations contradict previous simplified theoretical models but agree quantitatively with ab initio simulations that fully capture the 3D structural reconstruction. Our results reveal that the strain redistribution and 3D buckling in TMD heterostructures dominate the effective moiré potential and the corresponding moiré flat bands at the Brillouin zone K points.

Moiré superlattices in two-dimensional (2D) heterostructures provide an attractive platform to explore novel correlated physics, since nearly flat electronic bands can be engineered to enhance the effects of electron–electron correlations. This was first seen in graphene-based moiré superlattices, where correlated insulator states, superconductivity and ferromagnetic Chern insulators have been observed in both twisted bilayer$^{10-12}$, double bilayer$^{13,14}$ and ABC trilayer$^{15-17}$ moiré systems. The TMD-based moiré superlattice can have even flatter minibands, thus enhancing the role of the long-range Coulomb interactions. They have recently emerged as a new model system to explore novel strongly correlated quantum phenomena, such as the correlated insulators and generalized Wigner crystal$^{18-20}$. More exotic emerging states, such as charge transfer insulators and pair density waves, have been predicted to emerge from theoretical models of hole-doped WSe$_2$/WS$_2$ moiré heterostructures$^{21-23}$. These correlated phenomena, however, depend sensitively on the precise structural and electronic properties of the underlying moiré superlattice, due to the delicate interplay amongst the atomic geometry, moiré band structure and Coulomb interactions. Fundamental understanding and quantum control of TMD-based moiré phenomena thus require both quantitative knowledge of three-dimensional (3D) superlattice reconstructions at the atomic level and flat band electronic structure at the meV energy level, something that has hitherto been missing.

Scanning tunnelling microscopy (STM) provides a powerful tool to characterize the atomic and electronic structure of moiré superlattices. Previous STM studies have successfully observed localized moiré flat bands and correlated electronic gaps in twisted bilayer graphene$^{24-26}$, and demonstrated moiré site-dependent electronic structure in TMD moiré superlattices$^{27-29}$. Narrow moiré flat bands at the valence band edge of TMD heterostructures, however, have not yet been reported. Part of the challenge is the difficulty in fabricating high-quality exfoliated TMD moiré heterostructures on insulating substrates that are suitable for STM characterization. As a result, previous STM studies often focused on chemical vapour deposition (CVD) grown TMD heterostructures on conducting graphite. CVD growth, however, yields lower sample quality and very limited control of stacking order and twist angle of the TMD heterostructure compared to exfoliation and stacking techniques. A graphite substrate can also pin the Fermi level of the TMD material and induce undesirable electronic screening and modification of TMD band structure$^{27-29}$.

In this work we determine the moiré flat band electronic structure of 3D reconstructed WSe$_2$/WS$_2$ moiré superlattices by combining scanning tunnelling spectroscopy (STS) of high-quality exfoliated TMD heterostructure devices with ab initio simulations of both the atomic geometry and electronic band structure of TMD moiré superlattices. Our STM imaging and theoretical simulations reveal a striking 3D buckling reconstruction of the WS$_2$/WSe$_2$ heterostructures that is accompanied by strong strain redistribution...
within the moiré superlattice. We observe multiple moiré flat bands at the valence band edge that originate from the K point, as well as a separate set of deep-lying moiré flat bands that originate from the Γ point (our convention is to refer to the K and Γ points of the unfolded WSe₂ Brillouin zone (BZ) instead of the moiré BZ). The topmost valence flat band from the K point is prominently narrow with a width of only 10 meV, and is expected to be responsible for the recently observed novel correlated insulator behaviour and generalized Wigner crystal states\(^{18,19,24,25}\). The strong localization of this band at the B\(^{WS}\) stacking site revealed by STS spatial mapping contradicts previous simplified density functional theory (DFT) calculations, which predict localization at the AA site\(^{14,18}\). The STS results, however, are fully consistent with our DFT results obtained using a calculated large 3D reconstructed moiré superlattice. Our results show that the unexpected 3D moiré reconstruction and strain redistribution play a dominant role in determining the lowest energy moiré flat bands in WS₂/WSe₂ heterostructures.

The schematic of our WSe₂/WS₂ heterostructure device is shown in Fig. 1a. We used an array of graphene nanoribbons (GNRs) as contact electrodes, and the silicon substrate as a back gate to control the carrier density of the heterostructure. Details of the device fabrication are presented in the Supplementary Information. Figure 1b shows an ambient atomic force microscopy image of the top surface of the device: an array of GNRs (each separated by 100 to ~200 nm) partially covers the WSe₂/WS₂ heterostructure. Figure 1c shows an enlarged large-scale ultrahigh vacuum STM image of the heterostructure. The moiré superlattice can be clearly resolved in both the exposed TMD and GNR-covered areas, demonstrating the high quality of the heterostructure device.

Figure 2a shows a zoomed-in STM image of the moiré superlattice in the exposed TMD area. It shows a moiré period of 8.16 nm, which is consistent with the period expected for an aligned WSe₂/WS₂ heterostructure with a near-zero twist angle. Exposed WSe₂/WS₂ and graphene-covered areas are labelled. The WS₂, with smaller lattice constant, has the opposite strain distribution within the WSe₂ layer of the heterostructure. The moiré superlattice shows a strong moiré superlattice reconstruction that includes both a large in-plane strain distribution and a prominent out-of-plane buckling. Figure 2e shows the calculated in-plane strain distribution within the WSe₂ layer of the heterostructure. The moiré superlattice tends to increase the area of the interlayer-locked AB stacking regions due to its lower energy than AA stacking. As a result, the WSe₂ layer becomes locally compressed at the AB stacking regions due to its larger lattice constant compared with WS₂. The residual tensile strain localizes to the AA stacking region (Fig. 2e). The WSe₂ layer, with smaller lattice constant, has the opposite strain distribution (Supplementary Fig. 7). To partially release this strain, the heterobilayer reconstructs in three dimensions by an in-phase buckling in the out-of-plane direction (Fig. 2h.i). The simulated height distribution of the top WSe₂ layer (Fig. 2f) perfectly
reproduces our STM image (Fig. 2a,b). Figure 2g shows a side view of the 3D reconstructed heterostructure from both experiment and theory. The buckling above the AA ‘valley’ causes the AB sites to rise. It is noteworthy that the presence of an hexagonal BN (hBN) substrate only slightly reduces the buckling effect (see Supplementary Information for more details). The simulated line profile agrees well with our experimental data (Fig. 2g).

The moiré superlattice reconstruction has a profound impact on the electronic properties of the moiré flat bands. To observe this effect, we used STS to probe the local electronic structure of the WSe₂/WS₂ heterostructure. Figure 3a displays the STS dI/dV spectra acquired at different moiré sites for −3 V < Vₜₐₐ < 2 V. Differences are seen in the spectra obtained at different moiré sites, which are consistent with previous studies performed on bilayer heterostructures grown on graphite²¹,²².

We first focus our analysis of the STS spectra on the moiré flat band closest to the valence band edge, where the effects of strongly correlated states have been observed previously¹⁴,¹⁵. A challenge in the STM study of TMD materials is how to distinguish electronic states arising from K or Γ points in the single-layer BZ²¹,²². Here we utilize two distinct features of the K-point states to identify them. We first use the fact that, due to the much larger in-plane momentum of K-point states, their wavefunction decays faster outside the TMD layer (see details in the Supplementary Information). Figure 3b shows the height-dependent dI/dV spectra measured at one of the AB sites (ultimately confirmed as a BSTM/W site) in the moiré pattern. Two prominent peaks are observed near Vₜₐₐ = −1.7 V and Vₜₐₐ = −1.5 V. The peak near −1.5 V exhibits a much stronger height dependence than the peak near −1.7 V, suggesting that the peaks at −1.5 V and −1.7 V correspond to electronic states at the K and Γ points, respectively. We next use the two facts that the TMD K-point electron wavefunctions have large in-plane momentum and are mainly contributed by the W d orbital with angular momentum ℓ = ±2, which will induce atomic-scale alternating constructive and destructive interference patterns as illustrated in Fig. 3d (see details in Supplementary Information). The high-resolution dI/dV mapping at −1.5 V shows pronounced dI/dV signal oscillation over atomic-scale distances that match the WSe₂ lattice (Fig. 3e), while the dI/dV mapping at −1.7 V varies more smoothly (Fig. 3f). This behaviour confirms that the −1.5 V peak originates from K-point states at the valence band edge, whereas the −1.7 V peak originates from Γ-point states.

We performed larger scale dI/dV mapping to directly visualize the localization of the flat bands in real space. Figure 3g,i shows dI/dV maps obtained at the two peak energies −1.52 V (labelled K1) and −1.73 V (labelled Γ1). The LDOS for both of these flat band states are found to be strongly localized at the BSTM/W site. Atomic-scale site dependence in the dI/dV signal for K-point states is again reflected in the K1 dI/dV mapping. Figure 3h,j shows the dI/dV mapping at slightly lower energies. The LDOS distribution is seen to change dramatically and now shows LDOS minima, where previously there were maxima at the BSTM/W site.

To better determine the energy-dependent LDOS of the moiré flat bands, Fig. 3k shows a density plot of dI/dV spectra for the bias range −1.72 V < Vₜₐₐ < −1.42 V along the BSTM/WBerry/AA direction, indicated by the yellow path marked in Fig. 3g. Figure 3l shows the same plot, but over a different bias range: −1.86 V < Vₜₐₐ < −1.63 V. Figure 3k shows a prominent moiré flat band at the valence band minimum that is strongly localized at the BSTM/W site. This K-point moiré flat band is isolated from deeper moiré flat bands by a gap of ~50 meV. Figure 3c displays a high-resolution dI/dV spectrum at the BSTM/W site, which exhibits a peak having a full width at half maximum (FWHM) of 12 mV ± 1 mV. After a deconvolution eliminating the impact of the modulation voltage, the FWHM of the dI/dV peak is 10 mV ± 1 mV (see Supplementary Information for details), which sets an upper limit on the bandwidth of the moiré flat band of WSe₂/WS₂. The occupied moiré miniband in twisted bilayer graphene, by comparison, has an experimental bandwidth of 10 to ~40 meV (refs. 17–20). The narrowness of the WSe₂/WS₂ moiré flat band in combination with the strong long-range Coulomb interactions in 2D...
semiconductors makes this TMD heterostructure an excellent platform to explore highly correlated quantum phenomena.

In addition to the valence band edge moiré flat band, Fig. 3k shows that deeper moiré flat bands having different wavefunction spatial characteristics are present, but these bands are not well isolated. One such flat band can be tentatively identified near $-1.59$ V in Fig. 3h. The $dI/dV$ mapping of this flat band (labelled K2) shows a ring-shaped electron wavefunction around the BSe/W site in this case reminiscent of the first excited states of a harmonic oscillator (additional $dI/dV$ mapping and position-dependent $dI/dV$ spectra are included in the Supplementary Information).

To interpret the observed moiré minibands, we performed large-scale DFT calculations on the force field-reconstructed moiré superlattice. Figure 4a shows the calculated valence band structure for the WSe$_2$/WS$_2$ moiré superlattice in the mini-BZ (left) and the corresponding plot of the density of states (DOS) (right). The valence band edge is set at $E=0$. The bands closest to the valence band edge lies at $-1.73$ V and is localized to the B$^{Se/W}$ site. At the slightly deeper energy of $-1.78$ V a new wavefunction distribution is seen and the LDOS now shows a minimum at the B$^{Se/W}$ site (Fig. 3i). The ring-shaped electron wavefunction around the B$^{Se/W}$ site in this case is once again reminiscent of the first excited states of a harmonic oscillator (additional $dI/dV$ mapping and position-dependent $dI/dV$ spectra are included in the Supplementary Information).

Fig. 3 | STS measurement of moiré-induced flat bands. a, Moiré site-dependent $dI/dV$ spectra with low current set point ($V_{bias} = -3$ V, $I=70$ pA). Peaks in the $-2$ V $< V_{bias} < -1.7$ V range show strong moiré site-dependent peak positions. b, Tip-sample distance ($d$)-dependent STS at the B$^{Se/W}$ site ($V_{bias} = -2.15$ V, $I=50$, 100, 200, 400, 800, 1,600 pA). A second peak near $V_{bias} = -1.5$ V emerges with decreased $d$, indicating that it has a larger decay constant and originates from K-point states. c, High-resolution $dI/dV$ spectrum measured at the B$^{Se/W}$ site. A sharp peak with FWHM of 12 mV ± 1 mV can be observed near $V_{bias} = -1.5$ V (uncertainty in the FWHM of this peak comes from the standard deviation of widths extracted from spectra obtained at different B$^{Se/W}$ sites). d, Illustration of the atomic-scale wavefunction interference pattern. K-point states have a 2π phase winding over the adjacent three W atoms, while Γ-point states have identical phases over all Se atom sites. e, f, High-resolution $dI/dV$ mappings measured at the same B$^{Se/W}$ region with biases corresponding to the K-point ($-1.5$ V) (e) and the Γ-point ($-1.71$ V) (f) peaks. Black hollow circles in e show the atomic-scale constructive interference points as illustrated in d. g, h, Large-scale $dI/dV$ mappings of K-point states for $V_{bias} = -1.52$ V (g) and $V_{bias} = -1.59$ V (h). i, j, Large-scale $dI/dV$ mappings and Γ-point states for $V_{bias} = -1.73$ V (i) and $V_{bias} = -1.78$ V (i). Panels g–j show the same region of the sample surface. Solid dots in g label the positions of B$^{Se/W}$ (red), B$^{W/S}$ (green) and AA (blue) sites. k, l, $dI/dV$ density plot of K-point (k) and Γ-point (l) states along the two-segment yellow path shown in g. The corresponding path is also shown in k, l. Horizontal arrows label the positions of the B$^{W/S}$ (green), B$^{Se/W}$ (red) and AA (blue) sites. White vertical arrows label the energies used in g, j. $V_{bias} = 50$ V for all measurements shown. The tip-sample distance is determined by the set point $V_{bias} = -2.15$ V, $I=800$ pA for all spectroscopy shown. The lock-in modulation is 20 mV for all panels except c, where the lock-in modulation is 5 mV.
In summary, we find that 3D moiré reconstruction dominates the low-energy moiré electronic structure, resulting in a narrow moiré flat band with 10 meV bandwidth at the valence band maximum in WS₂/WS₂ heterostructures. Such quantitative understanding of the atomic and electronic structure within a moiré superlattice is crucial for simulating the Hubbard model and for the future control of novel correlated phenomena in TMD-based moiré heterostructures.

**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/s41563-021-00923-6.

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Data availability
The data supporting the findings of this study are included in the main text and in the Supplementary Information files, and are also available at https://github.com/HongyuanLiCMP/Moire_STM_source_data.

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Author contributions
M.F.C., F.W. and S.L. conceived the project, and S.G.L. supervised the theoretical calculations. H.L. and S.L. performed the STM measurements, and M.H.N. carried out the DFT and GW calculations. H.L., J.X., X.L., J.W., W.Z., S.Z. and S.K. fabricated the heterostructure device. E.R. and D.W. performed the second harmonic generation measurements. K.Y., M.B. and S.T. grew WSe₂ and WS₂ crystals. K.W. and T.T. grew the hBN single crystal. All authors discussed the results and wrote the manuscript.

Competing interests
The authors declare no competing interests.

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