Electronic Supplementary Material (ESI) for Nanoscale.

Modulating the periods and electronic properties of striped moiré superstructures for monolayer WSe_2 on Au(100) by varied interface coupling

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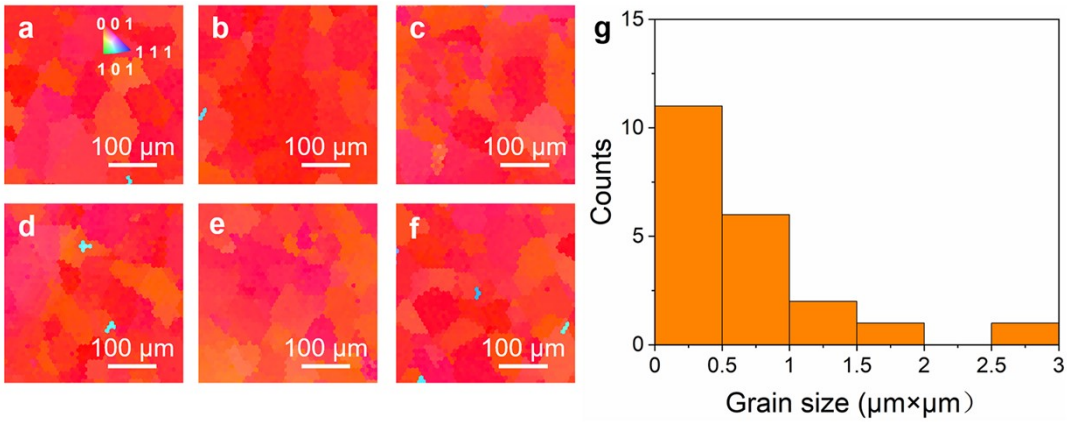

Figure S1. Typical grain size statistics of the Au(100) facet. (a-f) EBSD maps from six local regions, respectively. (g) Statistical histogram of the grain sizes of the Au(100) facet.

According to the statistical results, grain sizes of the Au(100) facet are mainly distributed within 0-1 um^2. Treatment of Au foils: Polycrystalline gold foils were first ultrasonically cleaned, and then loaded into the CDW chamber for a long standard annealing treatment time (~950 °C for 5 h in atmosphere).

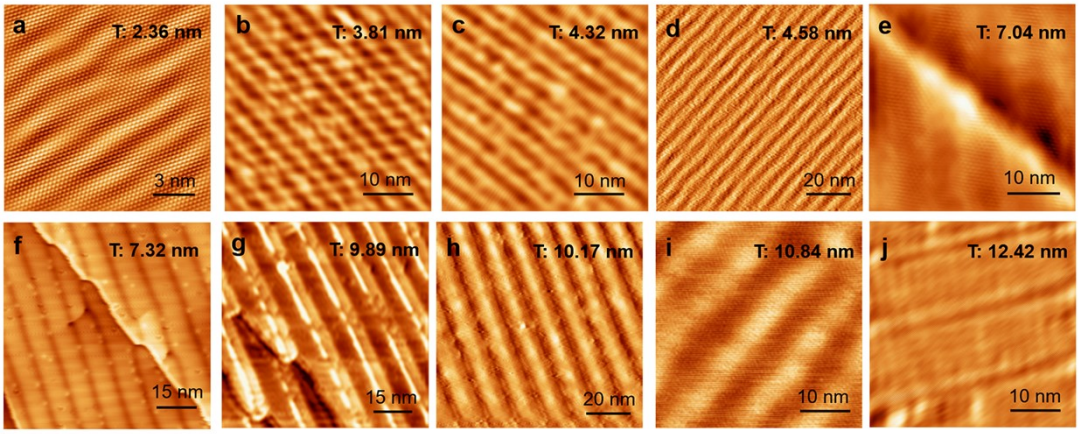

Figure S2. STM characterizations of striped moiré superstructure patterns for monolayer WSe_2 on Au(100) after annealing process at ~340 °C. "T" represents the periodicities or the inter-stripe distances for striped moiré superstructures.

After ~340 °C annealing process, the typical morphology of the striped moiré superstructure patterns for monolayer WSe_2 on Au(100) was obtained, as shown in Figure S2. The periodicities of striped patterns fall in a range of 0-15 nm, indicating nearly random orientations of monolayer WSe_2 domains on the Au(100) facet.

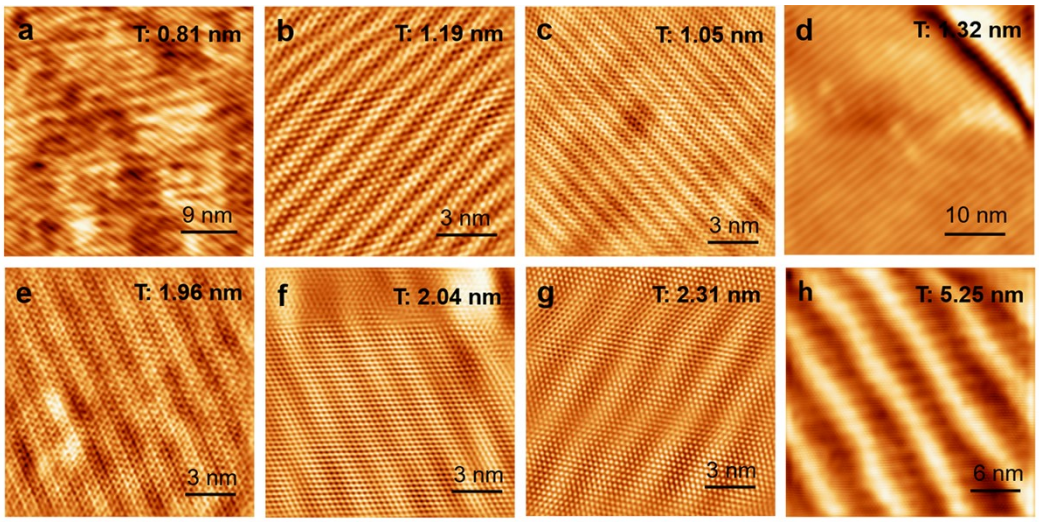

Figure S3. STM characterizations of striped moiré superstructures for monolayer WSe_2 on Au(100) after sample annealing process at ~420 °C for 4h.

After high-temperature annealing process at ~420 °C for 4h, the morphology of the striped moiré superstructure patterns for monolayer WSe_2 on Au(100) was achieved, as shown in Figure S3. The periodicities of the striped moiré superstructures fall in a narrow range.

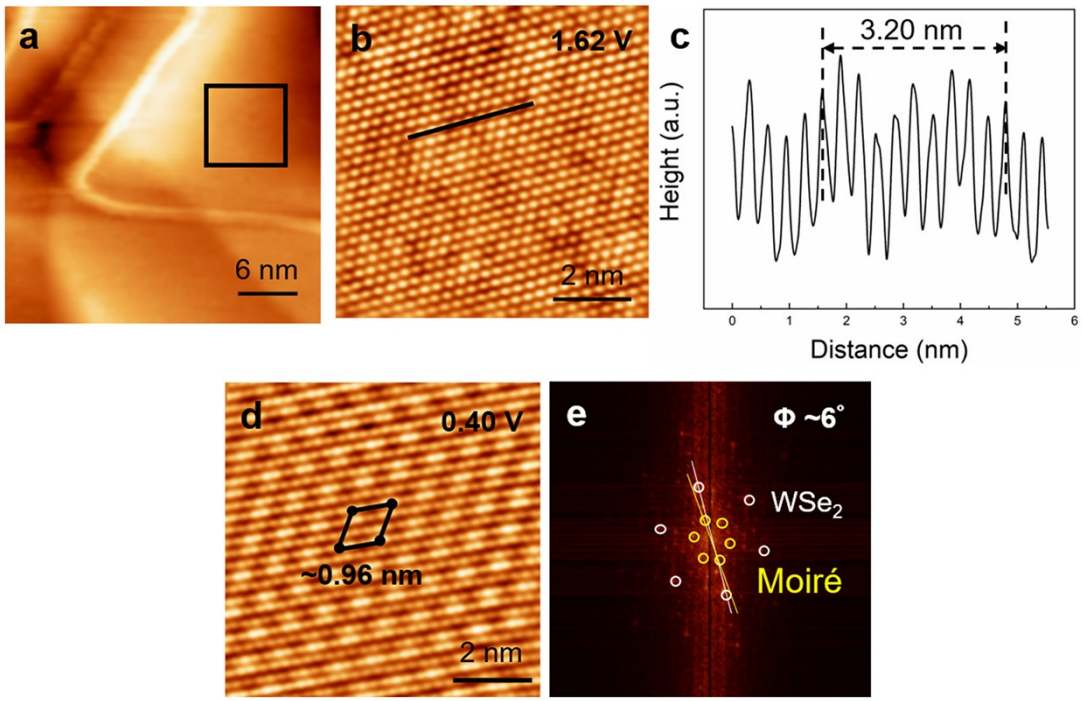

Figure S4. STM characterizations of monolayer WSe_2/Gr/Au vertical stack. (a) Large-scale STM image (1.63 V, 0.20 nA; 77 K) of monolayer WSe_2/Gr/Au. (b) Further magnification of the black rectangular region in (a). (c) Height profile along the black line in (b) showing a lattice constant of ~0.32 nm indicating the formation of WSe_2. (d) STM topography of monolayer WSe_2/Gr/Au (as also shown in Figure 5c) in the main text. (e) The corresponding 2D fast Fourier transform (2D-FFT) pattern from (d). The orientation angle between the atomic row of monolayer WSe_2 and the stripe direction of the moiré superstructure is ~6°.

A large region of monolayer WSe_2/Gr/Au heterostructure is shown in Figure S4. Further magnification at the atomic scale image and its height profile shows a lattice constant of ~0.32 nm, indicating the formation of monolayer WSe_2. The bias-dependent transition from the atomic lattice of monolayer WSe_2 to the moiré pattern indicates the existence of monolayer WSe_2/Gr/Au heterostructure according to the structural simulation. The corresponding FFT image reveals the orientation angle between atomic row of monolayer WSe_2 and the stripe direction of the moiré superstructure (~6°).

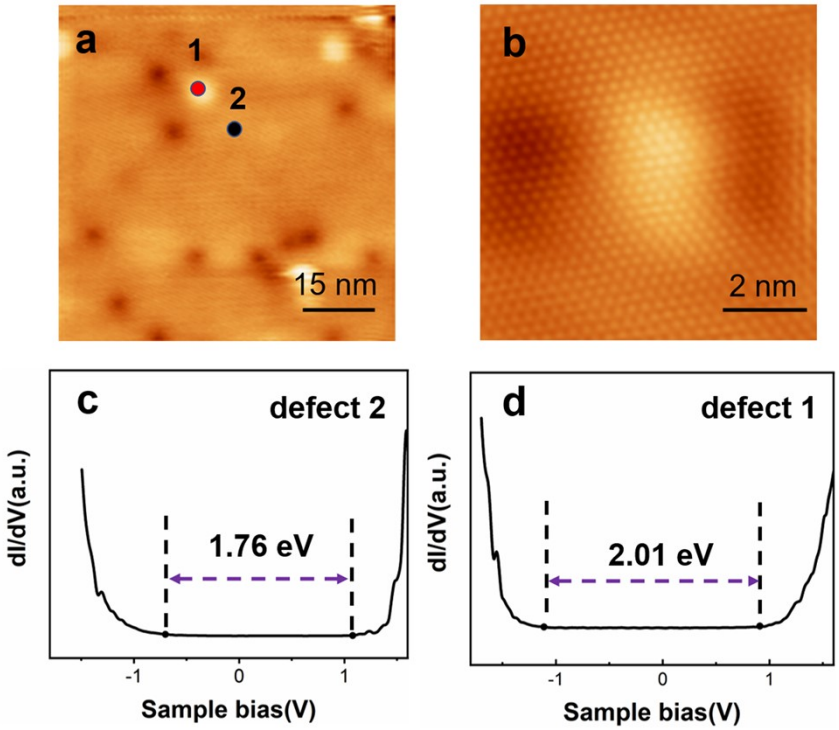

Figure S5. STM morphology of defect and defect-free areas for monolayer WSe_2/Au, and the corresponding STS spectra. (a) STM image (0.31 V, 0.20 nA; 77K) showing defect (bright protrusion marked by a red dot) and defect-free areas (marked by a black dot). The defects sites are characterized with dark spots or bright protrusions, corresponding to sulfur (S) vacancies or trapped atoms or molecules in the interfaces of monolayer WSe_2/Au. (b) Atomic resolution image (0.30 V, 0.20 nA; 77K) over the area in (a) marked with a red dot. (c-d) dI/dV spectra probed using STS at the defect-free areas (black dot) and the defect sites (red dot) from (a), respectively. (1.73 V, 0.80 nA, 10 mV, 932 Hz, 77K).

There are some light protrusions in the STM image, as shown in Figure S5 a. From successive zoom-in STM images in Figure S5 b, continuous crystal lattice can be observed to be decorated with dark or bright contrasts spots, corresponding to the sulfur (S) vacancies or trapped atoms or molecules in the interfaces of monolayer WSe_2/Au, respectively.

The STS spectra were collected with the STM tip positioned at the defect-free (black dot) and defect (red dot) regions, respectively. The VBM and CBM of the defect-free region are located at ~-0.70 eV and 1.06 eV, respectively, yielding a quasiparticle bandgap (Eg) of ~1.76 eV (Figure S5 c). At the defect site featured with bright protrusion (Figure S5 d), the VBM and CBM are located at ~-1.11 and 0.90 V, respectively, corresponding to band gap of ~2.01 eV, very close to the intrinsic band gap of monolayer WSe_2 on HOPG (~2.12 eV).[1] The interface trapped molecules or atoms are expected to weaken the interface coupling interaction between WSe_2 and Au, leading to more intrinsic electronic band gap than that of WSe_2/Au.

References

1 C. Zhang, Y. Chen, A. Johnson, M.-Y. Li, L.-J. Li, P. C. Mende, R. M. Feenstra and C.-K. Shih, *Nano Lett.*, 2015, **15**, 6494–6500.