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Supplementary Materials



Figure S1 (a) AFM image of a pristine twist bilayer TaS2 nanosheet. (b) The thickness profile of the bilayer TaS2 nanosheets.



Figure S2 XPS spectra of pristine twist bilayer TaS2 nanosheets. (a) Ta $4f_{7/2}$ spectra; and (b) S $2p_{3/2}$ spectra.



Figure S3. The methodology for twist angle measurements from FFT patterns. The estimated uncertainty in the FFT angle measurement is merely 0.1°.

The uncertainty originates from the definition of FFT spot centers during the angle estimations. Figure S3a shows the variation of measurement (β - α) in a typical FFT image (from the Figure 2g). Figure S3b is the enlarged image from the red dashed box, which clearly indicates the measured deviation. This deviation can be quantitatively measured by intensity line scan. Figure S3c displays the intensity profile from the blue frame in Figure S3b, verifying the measured angle error is only 0.1° (that is 0.1°/25° = 0.4%).



Figure S4. (a, b) Low and (c) high magnification STEM images of twist bilayer TaS₂. The inset of (a) is a SAED pattern taken from a large area.

The twist bilayer samples generally lay flat over large regions, except for the sample edges which indicate obvious curly structure, as shown in Figure S4. From the large field-of-view STEM images and corresponding selected area (large than 1 μ m in diameter) electron diffraction (the inset of Fig. S4a), it can be confirmed that the twist angles keep consist over the entire bilayered nanosheets, rather than discrete regions with different twist angles. Moreover, the twist angles from the selected area electron diffraction are consistent with these determined by FFT.



Figure S5. Histogram of each twist angle observed frequency in STEM images. Total 50 bilayer nanosheets were examined. (#pls add the interlayer distance in the upper X-axis or plot separately)



Figure S6. (a) The schematic of bilayer TaS2 with curled edge. (b) High resolution STEM image indicates the curled up edge structure. (c, d) From intensity line scan measurements, the estimated uncertainty of the interlayer distance is only $\sim 2.6\%$.

The interlayer distances were measured from bilayers with curled-up edges, following the method reported in the reference (NPG Asia Mater 2016, 8, e266). The geometry of curled twist bilayer TaS₂ is showed in Figure S6a. Similar to the methodology used in Figure S3, the errors come from the definition of two Mo atomic plane centers during the interlayer distance estimations, as shown in Figure S6c and d. The error values can be quantitatively measured by intensity line scans. Figure S6c is the enlarged image from the red frame of Figure S6b, Figure S6d displays the intensity profile from the blue frame of Figure S6c, verifying the measured error is only about 0.17 Å (that is 0.17Å/6.5Å = 2.6%).