

Supporting Information

# Controlled Growth of Large-Area Bilayer Tungsten Diselenides with Lateral $P$ - $N$ Junctions

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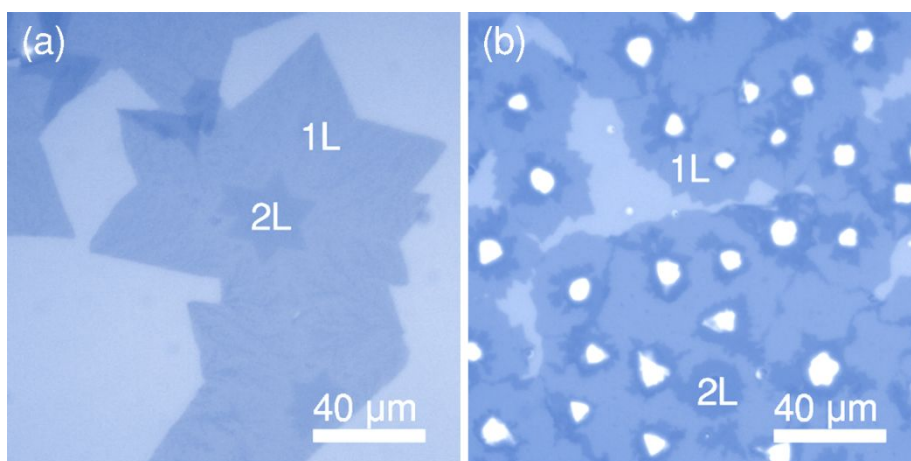
## AUTHOR INFORMATION

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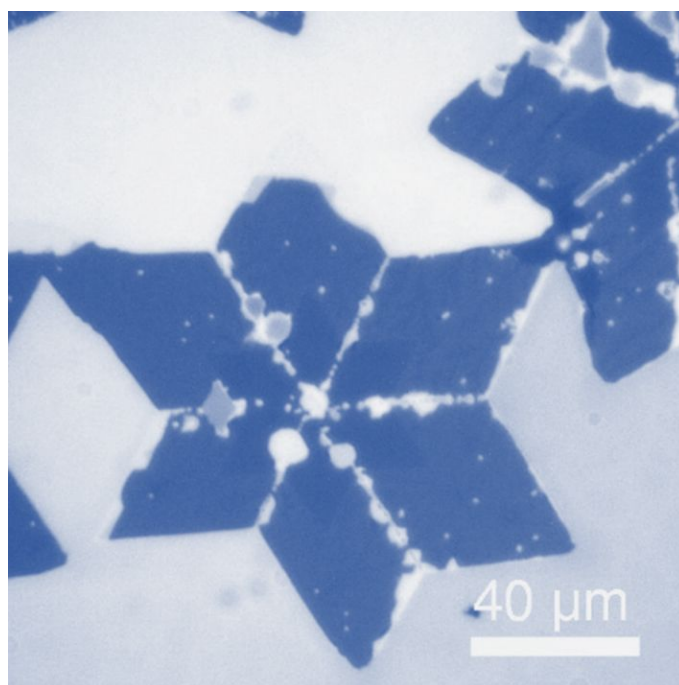
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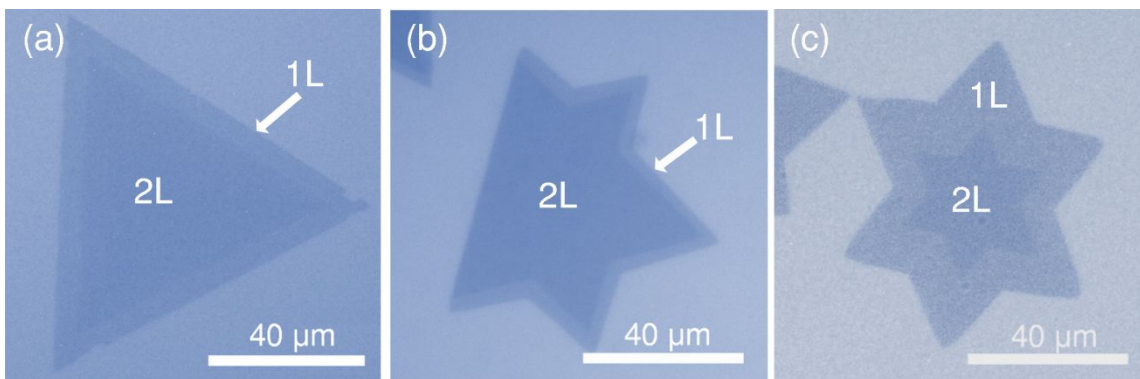
## SUPPLEMENTARY FIGURES



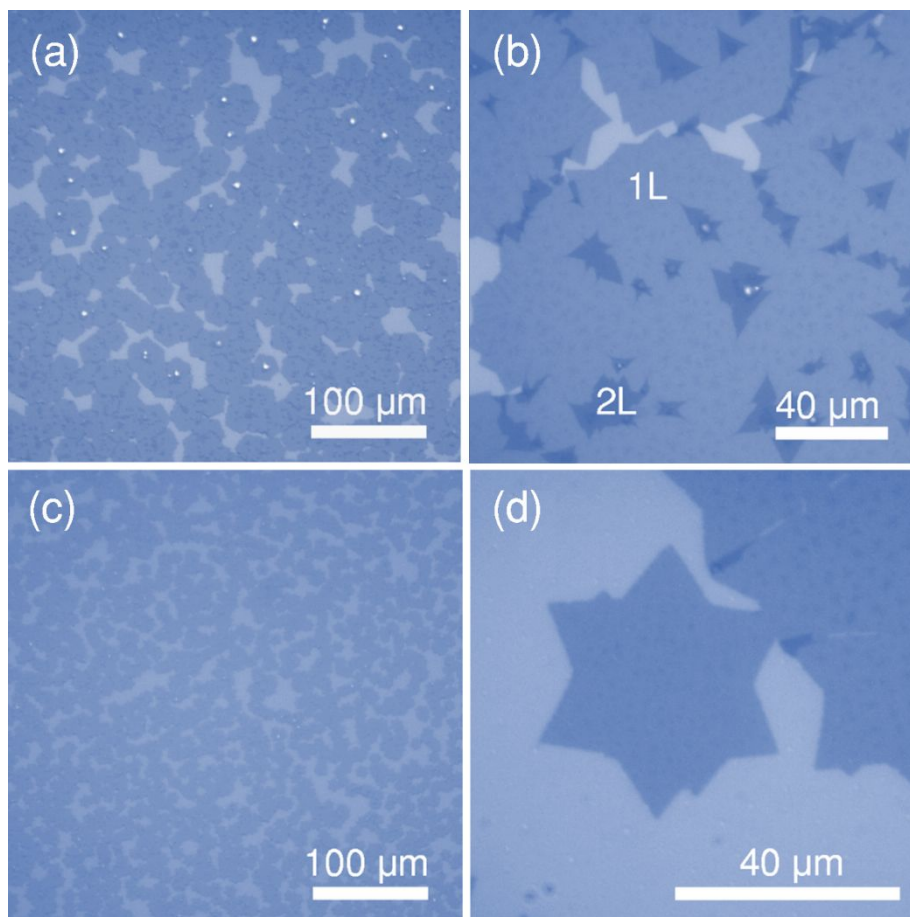
**Figure S1.** Optical images of the as-grown WSe<sub>2</sub> flakes using (a) 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> + 125 mM NaCl and (b) 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> + 500 mM NaCl as the growth promoter. The bright spots in (b) are multilayer WSe<sub>2</sub>.



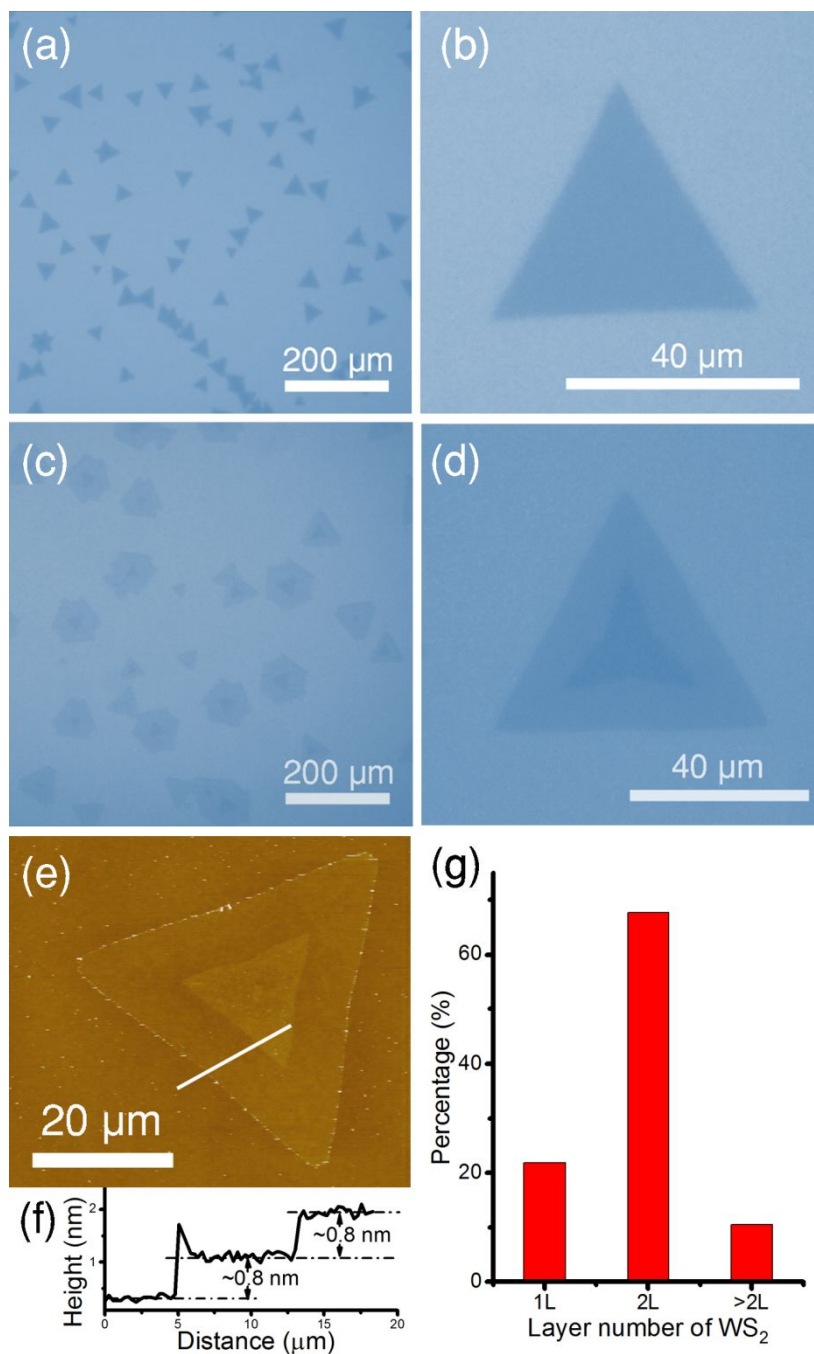
**Figure S2.** Optical image of a partially oxidized bilayer WSe<sub>2</sub> flake, in which the grain boundaries are clearly observed.



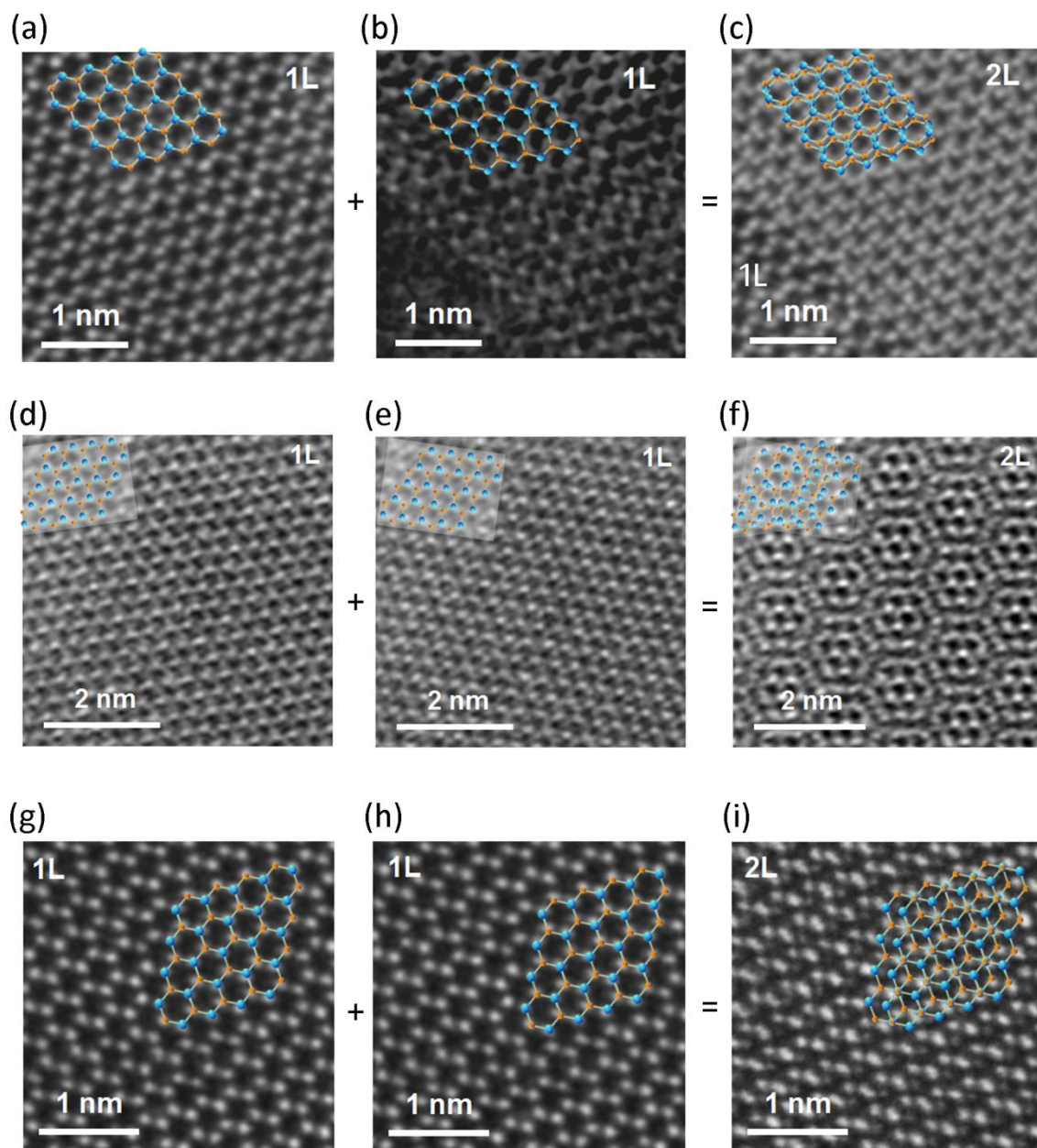
**Figure S3.** Optical images of bilayer WSe<sub>2</sub> flakes with varied morphologies using a mixture of 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> and 250 mM NaCl as the growth promoter.



**Figure S4.** Optical images of the as-grown WSe<sub>2</sub> flakes using (a,b) 75 mM NaCl and (c,d) 25 mM NaCl as the growth promoter.

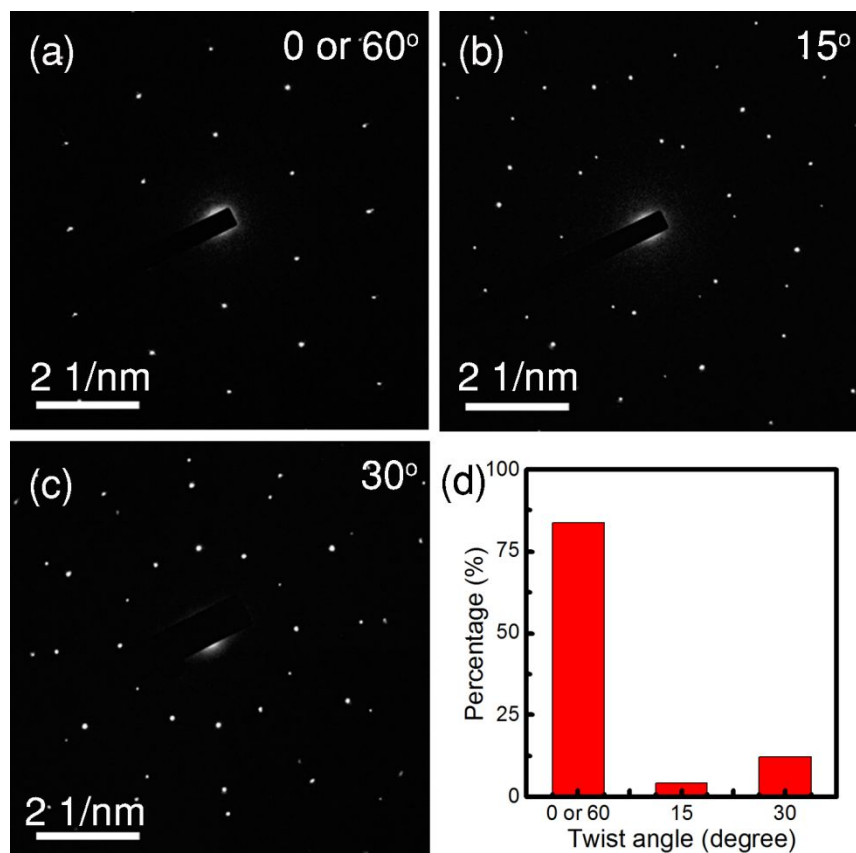


**Figure S5.** Optical images of WS<sub>2</sub> flakes grown by CVD using (a,b) 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> and (c,d) 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> and 250 mM NaCl as the growth promoter. (e) Atomic force microscope image of a bilayer WS<sub>2</sub> flake and (f) corresponding height profile; (g) Statistical analysis on the layer number of WS<sub>2</sub> flakes grown using 25 mM C<sub>24</sub>H<sub>40</sub>NaO<sub>5</sub> and 250 mM NaCl as the growth promoter, showing the preferential growth of bilayer material.

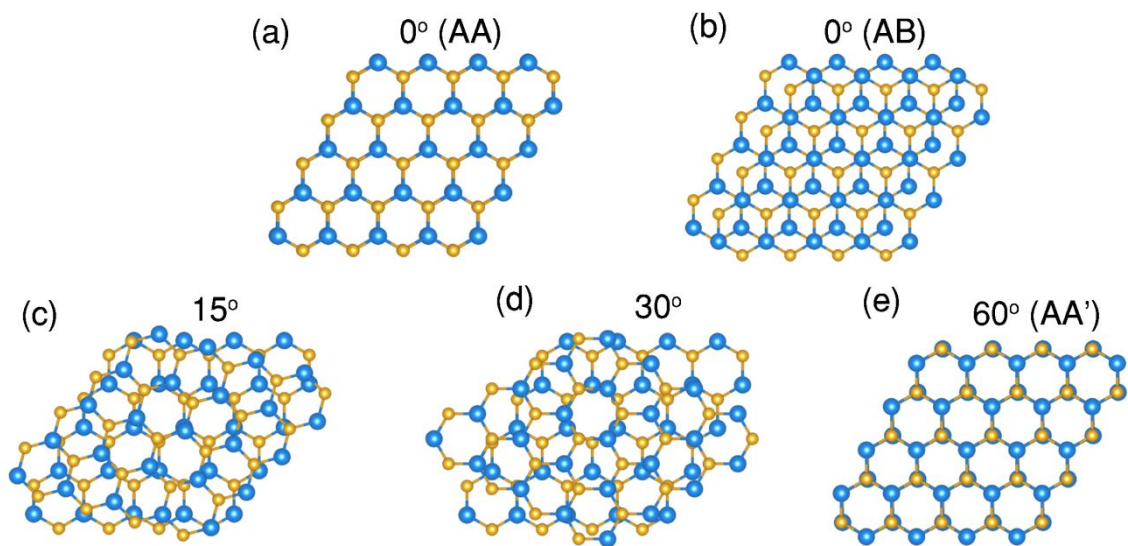


**Figure S6.** Analysis of the stacking orientation and atomic structure of bilayer  $\text{WSe}_2$  by fast Fourier transform (FFT) decomposition of the 2L regions: (a-c)  $60^\circ$  twist angle or AA' stacking; (d-f)  $15^\circ$  twist angle; (g-i)  $0^\circ$  twist angle or AB stacking. In each set of the 3 images, the 2L regions is decomposed into two monolayers by FFT to identify the corresponding atomic models. The related inset images present the corresponding atomic models.

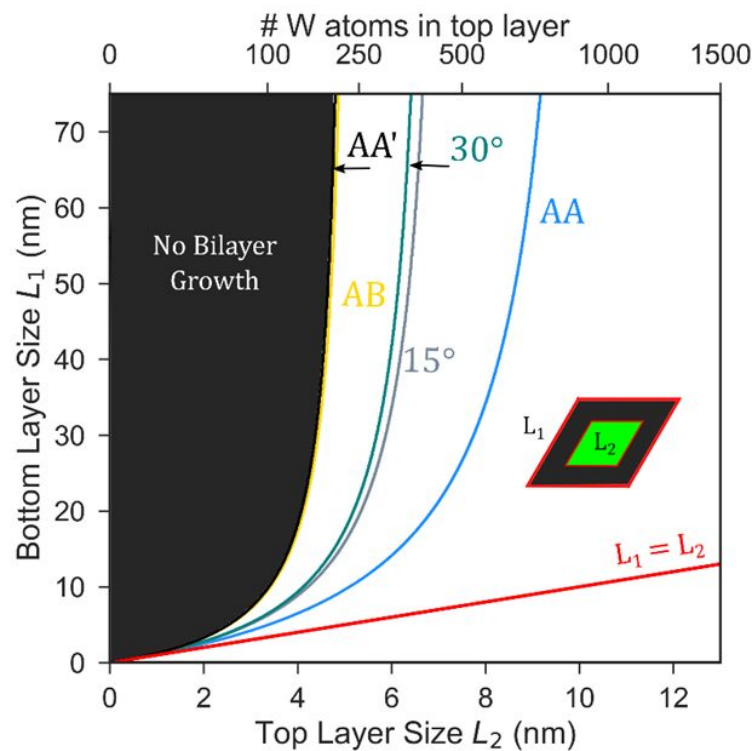




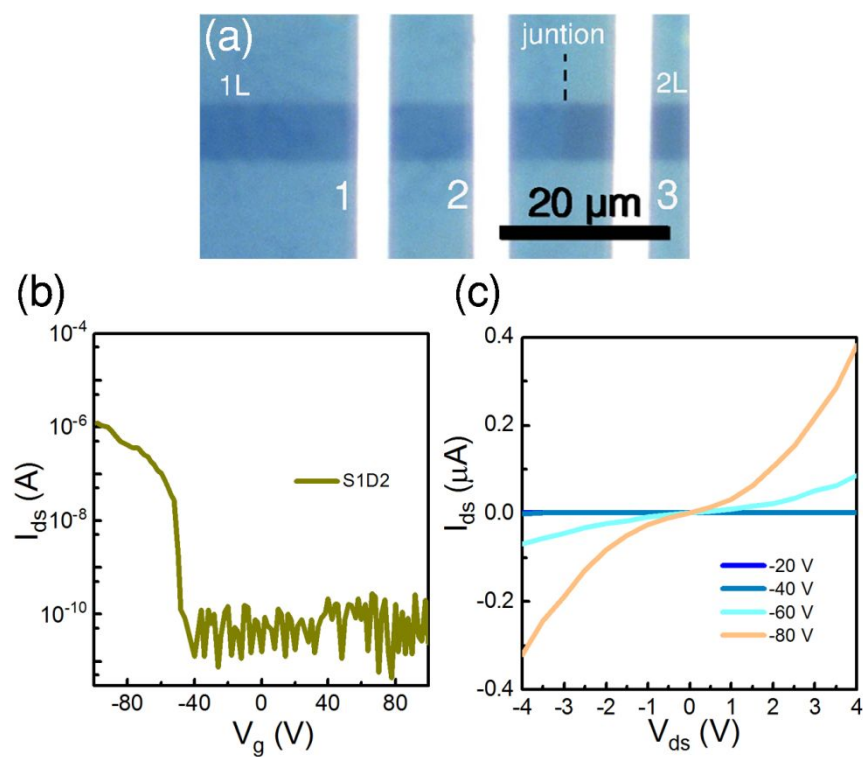
**Figure S7.** Selected area electron diffraction (SAED) patterns of bilayer WSe<sub>2</sub> flakes with (a) 0 or 60° twist, (b) 15° and (c) 30° twist angles; (d) Histogram of twist angle frequencies based on ~100 bilayer WSe<sub>2</sub> flakes.



**Figure S8.** Summary of the atomic models of bilayer WSe<sub>2</sub> with varied stacking orientations.



**Figure S9.** Thermodynamic growth diagram for bilayer WSe<sub>2</sub> flakes as a function of the instantaneous layer sizes  $L_1$  (bottom layer) and  $L_2$  (top layer). The lines mark the minimum required top layer size for thermodynamically favorable bilayer growth of triangular flakes over the range of bottom layer sizes.



**Figure S10.** (a) Optical micrograph of a device structure based on a bilayer flake containing a large monolayer region. The electrode pair S1D2 contacts the monolayer region. (b)  $I_{ds}$ - $V_g$  curves for the S1D2 FET device. (c)  $I_{ds}$ - $V_{ds}$  curves for the WSe<sub>2</sub>-based FET at different values of the backgate voltage.