Supporting Information

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

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## SUPPLEMENTARY FIGURES



**Figure S1.** Optical images of the as-grown WSe<sub>2</sub> flakes using (a) 25 mM  $C_{24}H_{40}NaO_5 + 125$  mM NaCl and (b) 25 mM  $C_{24}H_{40}NaO_5 + 500$  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  $C_{24}H_{40}NaO_5$  and 250 mM NaCl as the growth promoter.



**Figure S4.** Optical images of the as-grown  $WSe_2$  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  $C_{24}H_{40}NaO_5$  and (c,d) 25 mM  $C_{24}H_{40}NaO_5$  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  $C_{24}H_{40}NaO_5$  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  $WSe_2$  by fast Fourier transform (FFT) decomposition of the 2L regions: (a-c) 60° twist angle or AA' stacking; (d-f) 15° twist angle; (g-i) 0° 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  $WSe_2$  flakes as a function of the instantaneous layer sizes L1 (bottom layer) and L2 (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.