

**Step-Edge-Guided Nucleation and Growth of Aligned WSe<sub>2</sub> on Sapphire *via* a  
Layer-Over-Layer Growth Mode**

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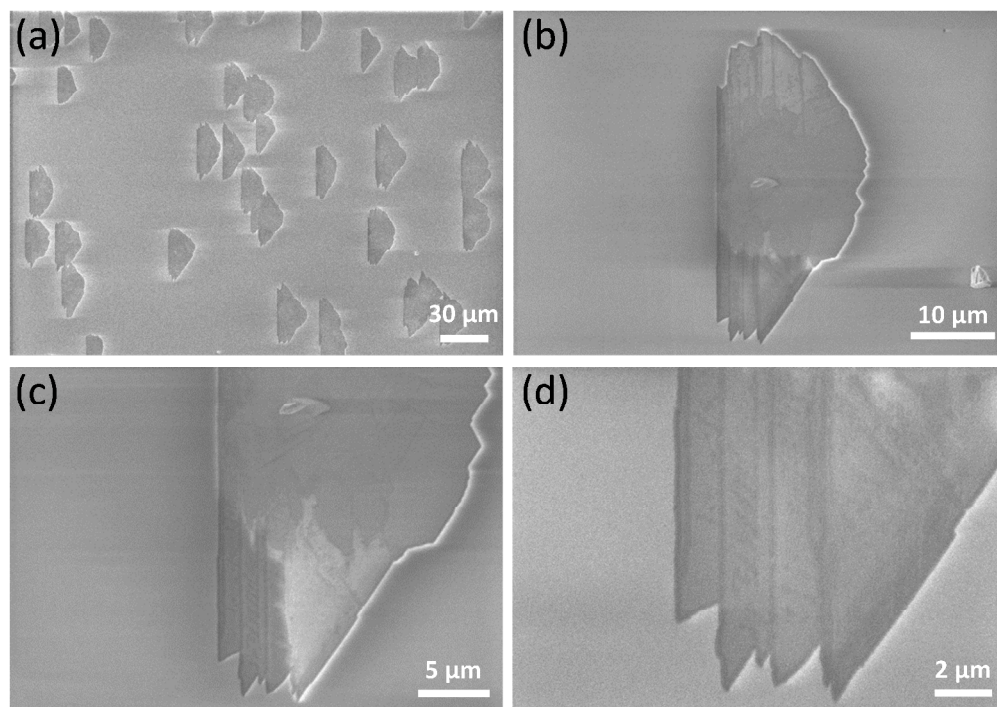


Figure S1. SEM images of aligned WSe<sub>2</sub> flakes on C-plane sapphire. (a) Aligned WSe<sub>2</sub> flakes with one aligned direction. (b)-(d) SEM examinations on the wing-like areas near the based edges of WSe<sub>2</sub> flakes which were grown at 990 °C.

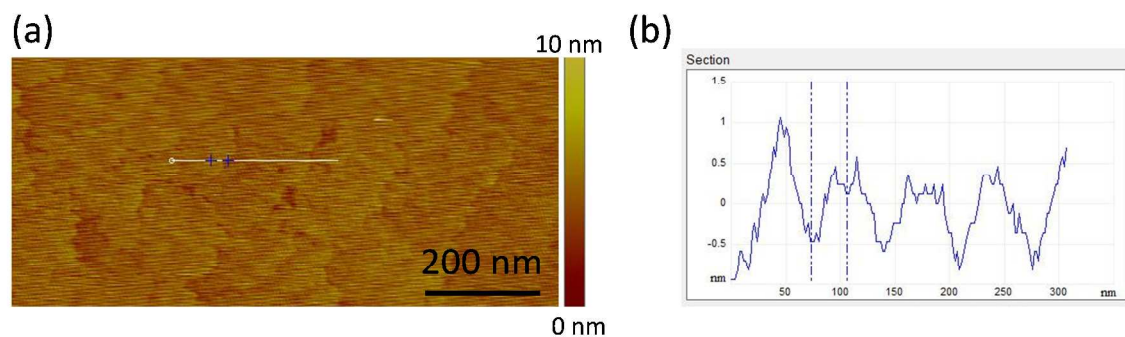


Figure S2. AFM characterization of pristine C-plane sapphire substrates. (a) An AFM height image shows isolated and irregular atomic steps on pristine sapphire surface before high temperature treatment. (b) Cross-section height profile of the atomic steps along the white line in (a).

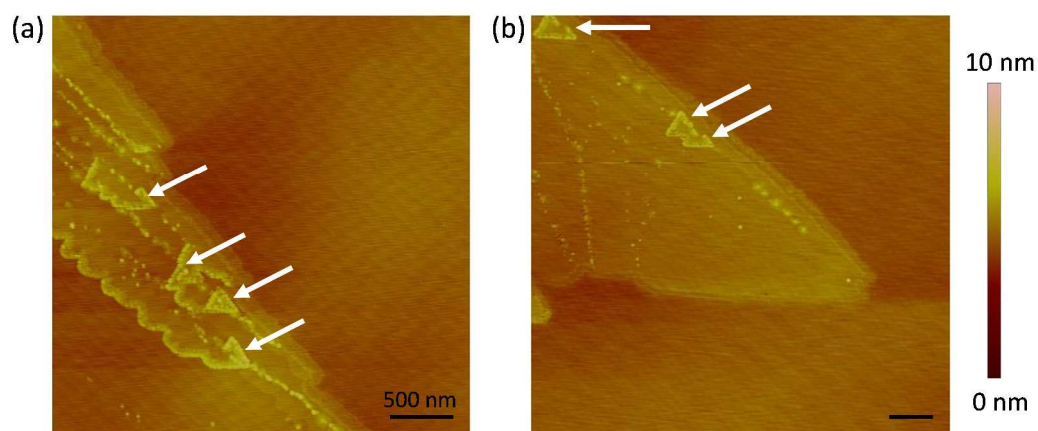


Figure S3. AFM height images show aligned nuclei along with periodic atomic steps on sapphire surface. The white arrows indicate some of the triangular nuclei of  $\text{WSe}_2$ .

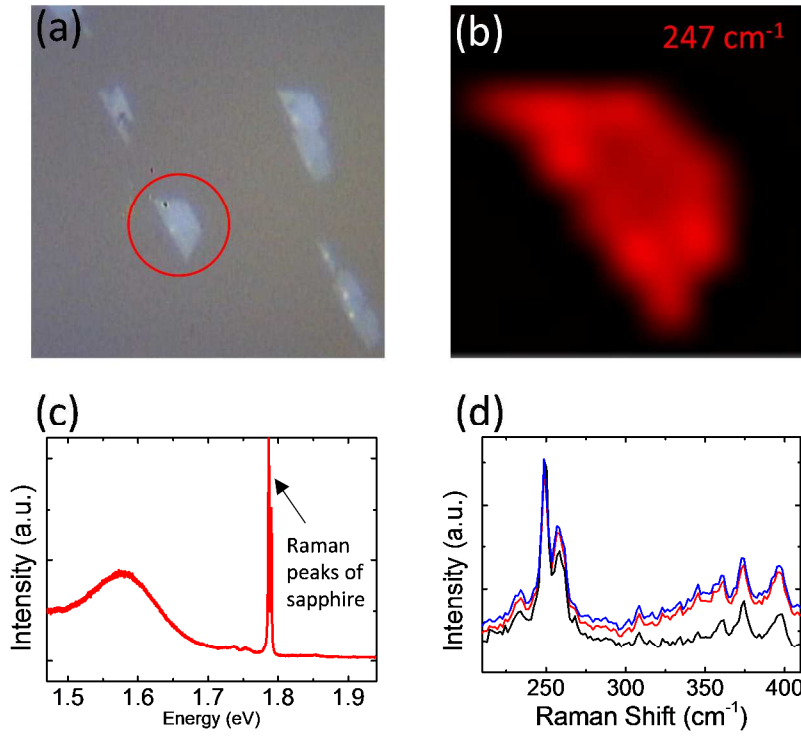


Figure S4. Raman and photoluminescence (PL) measurements on trapezoid WSe<sub>2</sub> flakes. (a) Optical microscopy images of typical flakes. (b) Raman intensity mapping at 247 cm<sup>-1</sup> of the flake highlighted in (a). (c) PL spectrum from the same flake in (a) shows broad and weak PL peaks at ~1.58 eV. (d) More Raman spectra from as-grown WSe<sub>2</sub> flakes, revealing they are few-layer flakes.

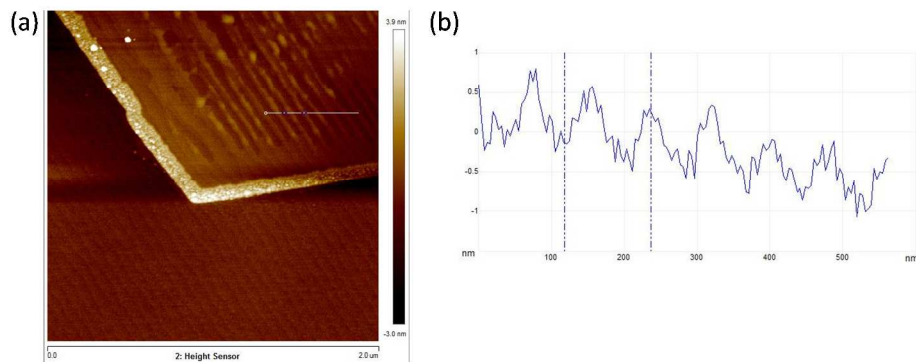


Figure S5. An additional AFM image on the flake showing in Figure 3 in the main text, showing the morphology of sapphire steps are well duplicated on the as-grown WSe<sub>2</sub> flake.

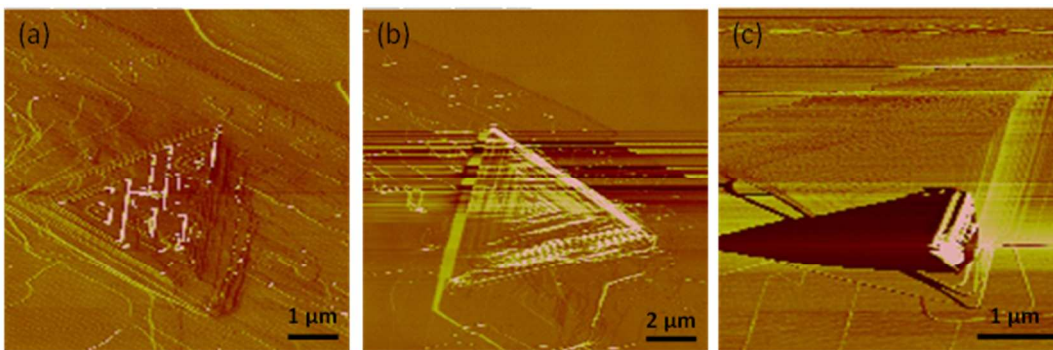


Figure S6. Additional AFM images on the screw-dislocation cores. Such screw dislocation hillocks exist at almost all of the aligned WSe<sub>2</sub> flakes.

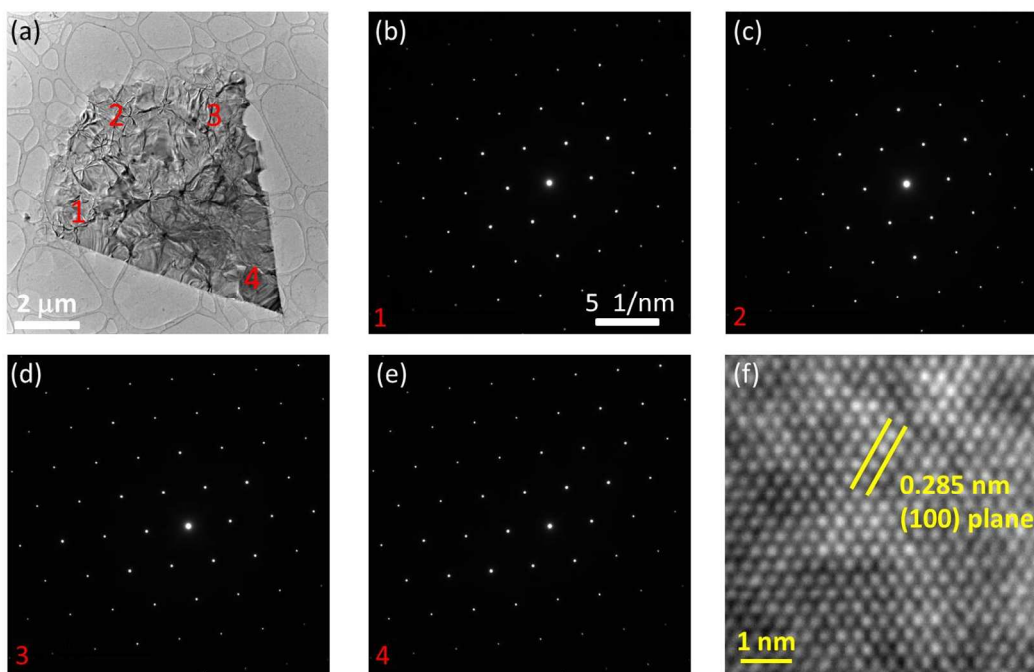


Figure S7. TEM characterization of WSe<sub>2</sub> flakes. (a) TEM image of a WSe<sub>2</sub> flake after being transferred onto TEM grid using PMMA-mediated transfer method. The flake becomes crumpled after transferring. (b)-(e) Diffraction patterns taken at locations 1-4 as indicated in image (a). These results show that the flake is a single crystal. (f) HRTEM image shows high crystalline WSe<sub>2</sub> flakes. The spacing is measured to be 0.285 nm for WSe<sub>2</sub> (100) lattice planes.