Supporting Information

Growth of 2H Stacked WSe_2 Bilayers on Sapphire

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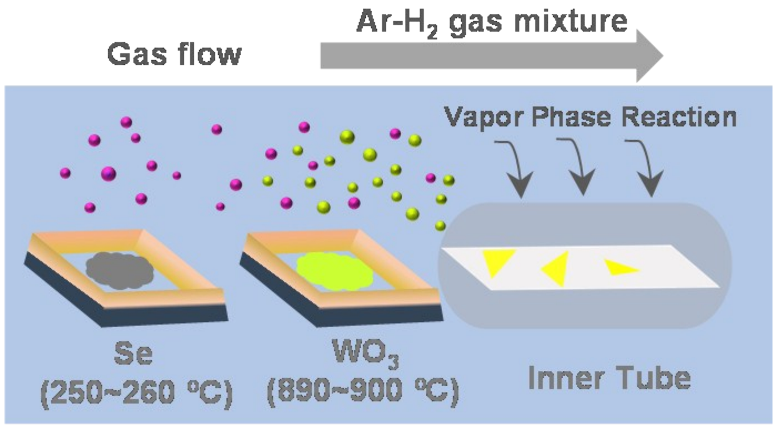

Figure S1. Schematic illustration of the CVD setup and the relative position between WO_3, Se and substrate. The distance between Se powder and WO_3 powder is ~25 cm.

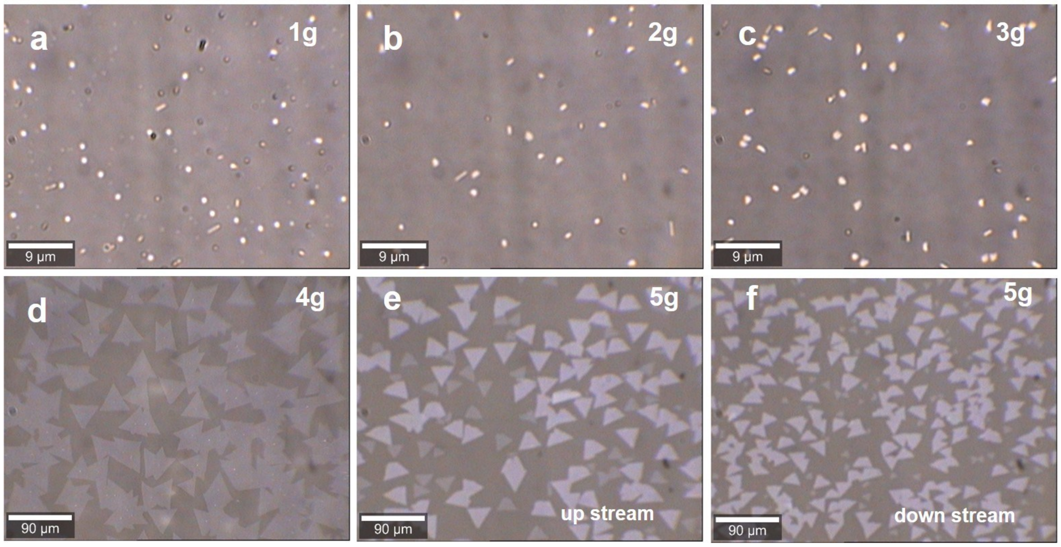

Figure S2. Optical images for the WSe_2 growth by CVD method. The mass amount of WO_3 is 0.3g, while the amount of Se powder is increased. The high-purity of H_2/Ar is as the carrier gas with a fixed flow rate of 5/65 sccm/sccm. The T_{Se} (temperature of Se) is maintained at 250 ^oC while T_{WO_3} (the temperature of WO_3) is kept at 895 ^oC. The growth pressure of the furnace is 10 torr for the whole CVD growth. The growth time is 15 mins.

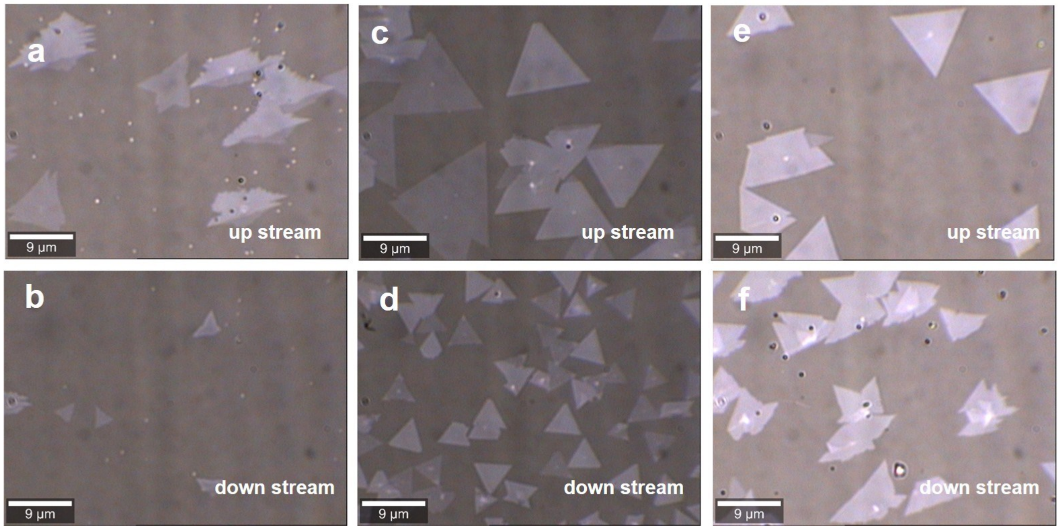

Figure S3. Optical images for the WSe_2 growth by CVD method. The mass amount of WO_3 is 0.3 g. The high-purity of H_2/Ar is as the carrier gas with a fixed flow rate of 5/65 sccm/sccm. **a-b,** The amount of Se powder is 5.5 g, T_{Se} = 250 ^{o}C and T_{WO_3} = 890 ^{o}C; **c-d,** The amount of Se powder is 5.0 g, T_{Se} = 260 ^{o}C and T_{WO_3} = 895 ^{o}C; **e-f,** The amount of Se powder is 5.5 g, T_{Se} = 250 ^{o}C and T_{WO_3} = 900 ^{o}C.

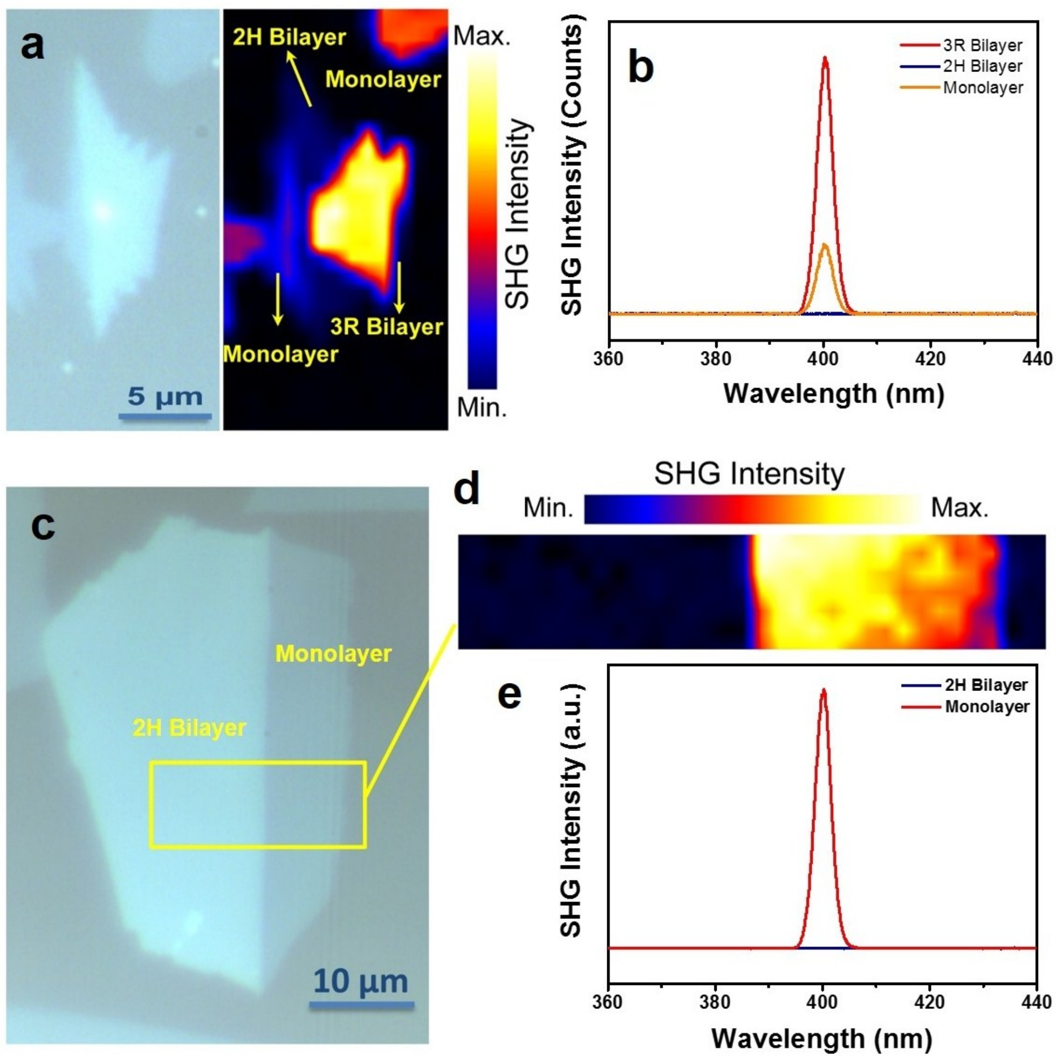

Figure S4. a, Left: optical micrograph of cloud bilayer WSe_2 crystal with monolayer WSe_2 as reference; Right: The corresponding SHG mapping intensity obtained by pixel-to-pixel spatial scanning on the crystals in Fig. S4a; **b,** The SH signal spectra of different layer number; **c,** Optical micrograph of irregular bilayer WSe_2 crystal with monolayer WSe_2 as reference; **d,** The corresponding SHG mapping intensity obtained by pixel-to-pixel spatial scanning on the crystals in Fig. S4c; **e,** The SH signal spectra of different layer number.

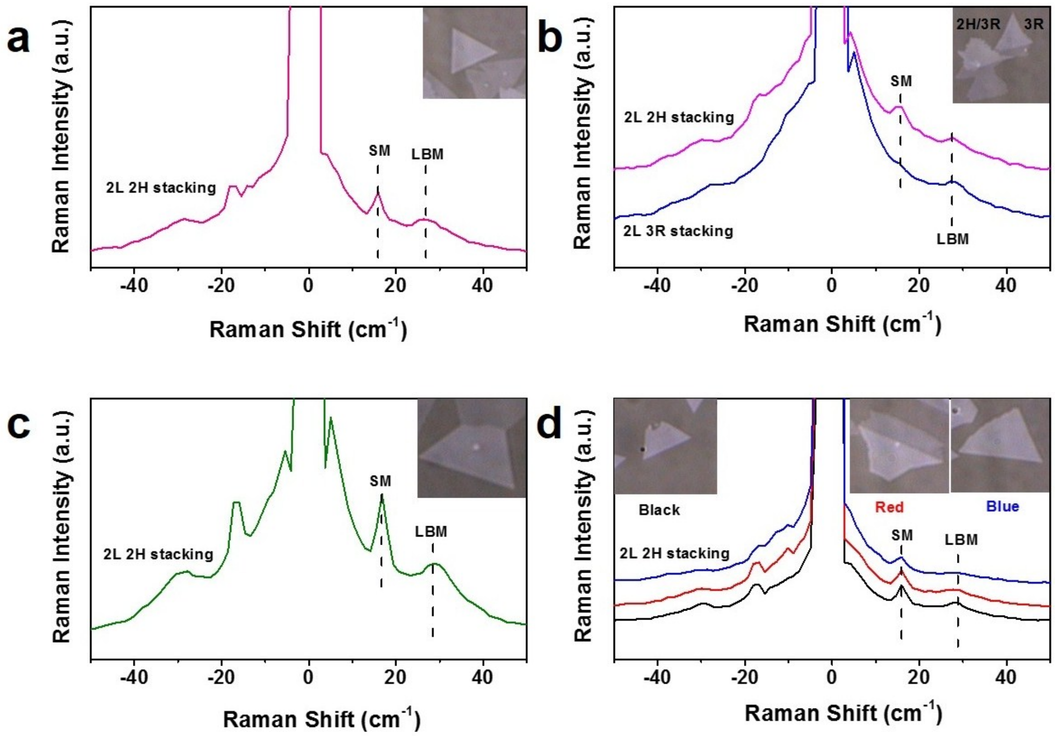

Figure S5. Low-frequency Raman spectra of bilayer WSe_2 crystals for 2H and 3R stacking configurations with different morphologies.

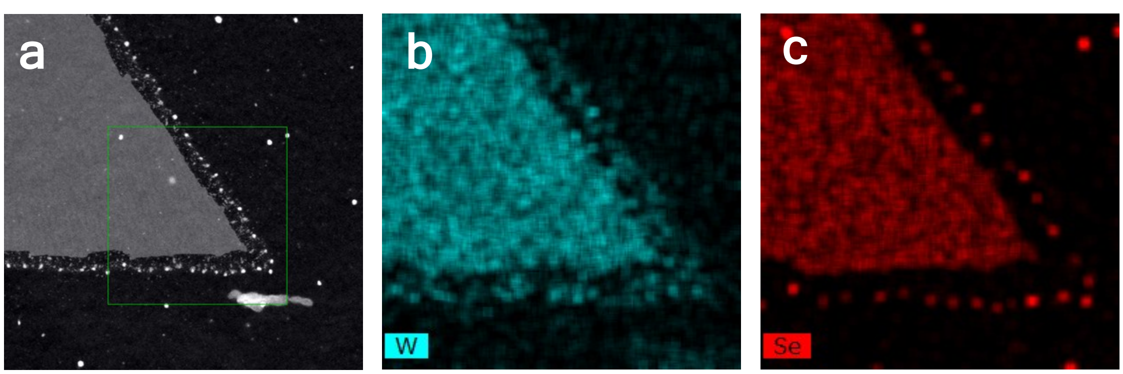

Figure S6 a, Low-magnification HAADF-STEM image of top-view bilayer WSe_2 sample; **b-c,** elemental mapping of the region (green frame) in Fig.a.

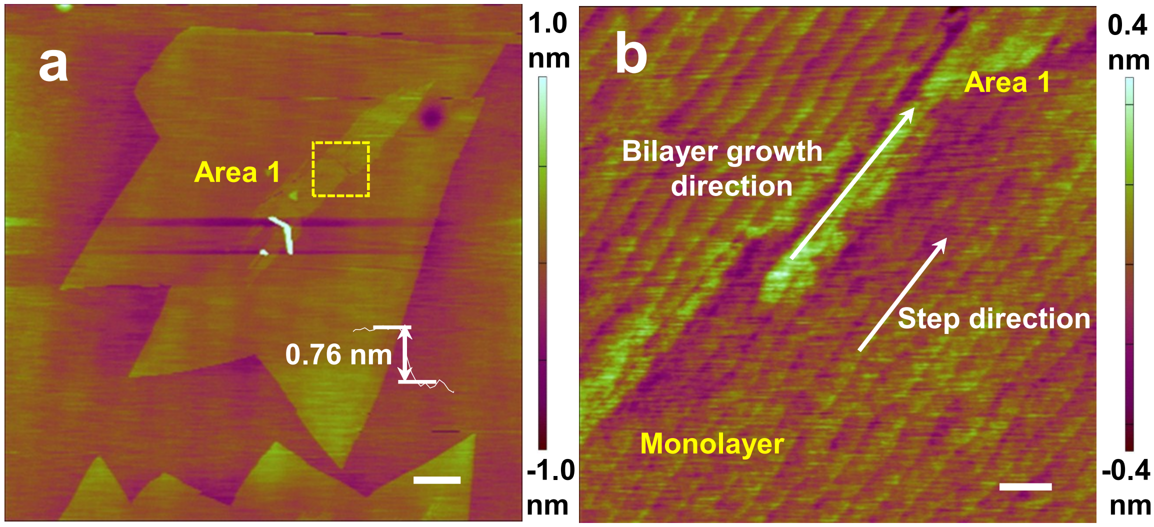

Figure S7. a, AFM topographic image of the monolayer WSe_2 grain boundary crystal with bilayer nuclei in the center area; **b,** The zoom-in AFM topographic image of area 1, indicative of initial WSe_2 bilayer nuclei aligned growth on the atomic steps. Scale bars: a, 2 μm; b, 100 nm.

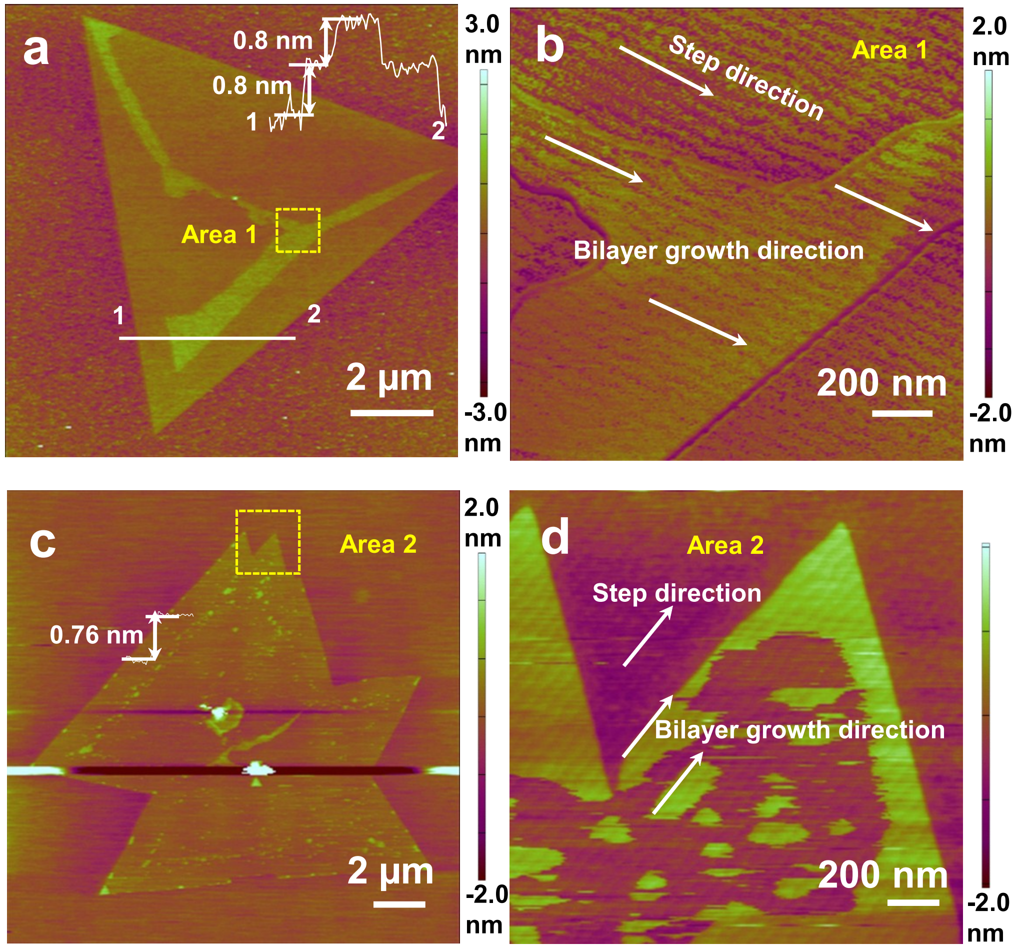

Figure S8. a-b, AFM topographic images of monolayer WSe_2 crystal with bilayer nuclei in different areas. The inset height profile is~0.8 nm, indicating a thickness of WSe_2 monolayer. The zoom-in AFM image in Fig.R1b shows WSe_2 bilayer nuclei initial growth on the atomic steps of sapphire; **c-d,** AFM topographic images of monolayer WSe_2 crystal with bilayer nuclei in different areas. The inset height profile is ~0.76 nm, indicating a thickness of WSe_2 monolayer The zoom-in AFM image in Fig. R1d shows WSe_2 bilayer nuclei initial growth on the atomic steps of sapphire.

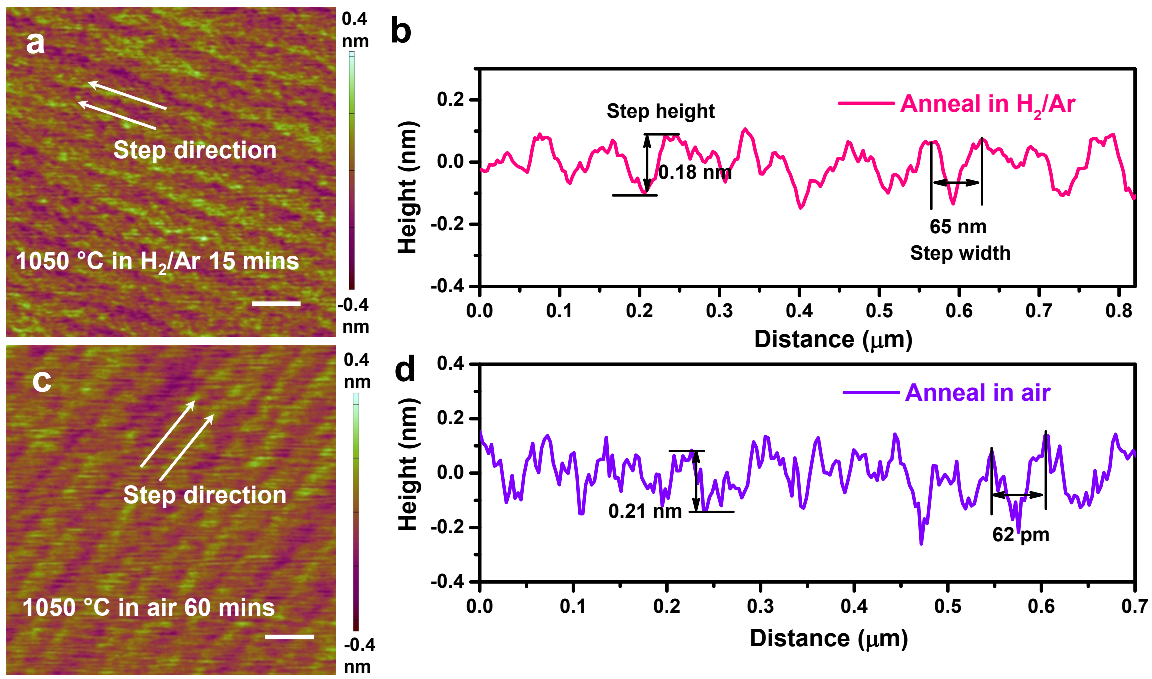

Figure S9. a, AFM topographic image of sapphire surface after high-temperature treatment (1050 °C in H_2/Ar for 15 mins); **b,** the corresponding cross-section height profile of the atomic steps along the vertical step direction in Fig. S7a; **c,** AFM topographic image of sapphire surface after high-temperature treatment (1050 °C in air for 60 mins); **d,** the corresponding cross-section height profile of the atomic steps along the vertical step direction in Fig. S7c. Scale bars: a, 100 nm; c, 100 nm.

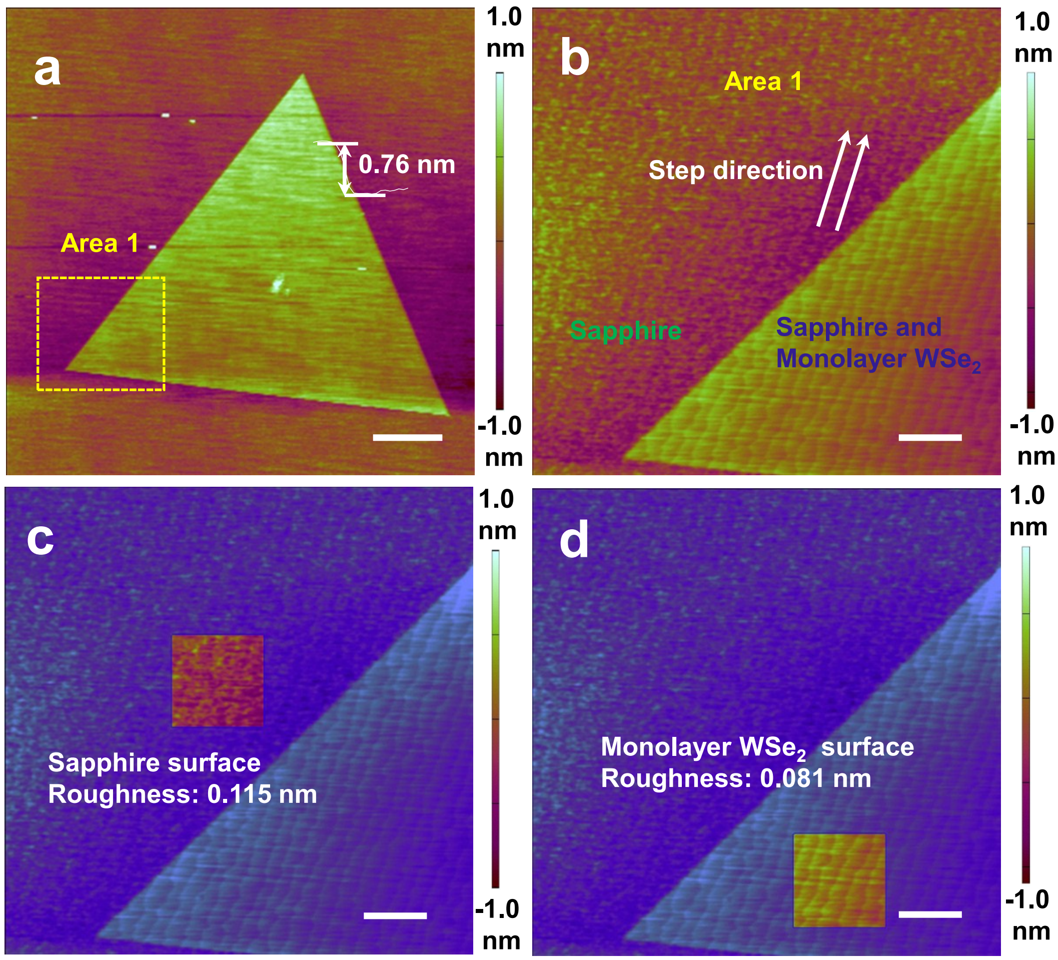

Figure S10. a, AFM topographic image of one WSe_2 crystal. The inset height profile was ~0.8 nm, indicating a monolayer thickness; **b,** The zoom-in AFM image in Fig. S8a. And the image showed irregular atomic steps on bare sapphire surface without any pre-treatment. In contrast, the apparently periodic atomic steps were shown after covering monolayer WSe_2; **c,** The selected area for the roughness calculation of bare sapphire surface (300 nm x 300 nm); **d,** The selected area for the average roughness calculation of sapphire surface with monolayer WSe_2 covering (300 nm x 300 nm). Scale bars: a, 2 µm; b, 200 nm; c, 2 µm; d, 200 nm.

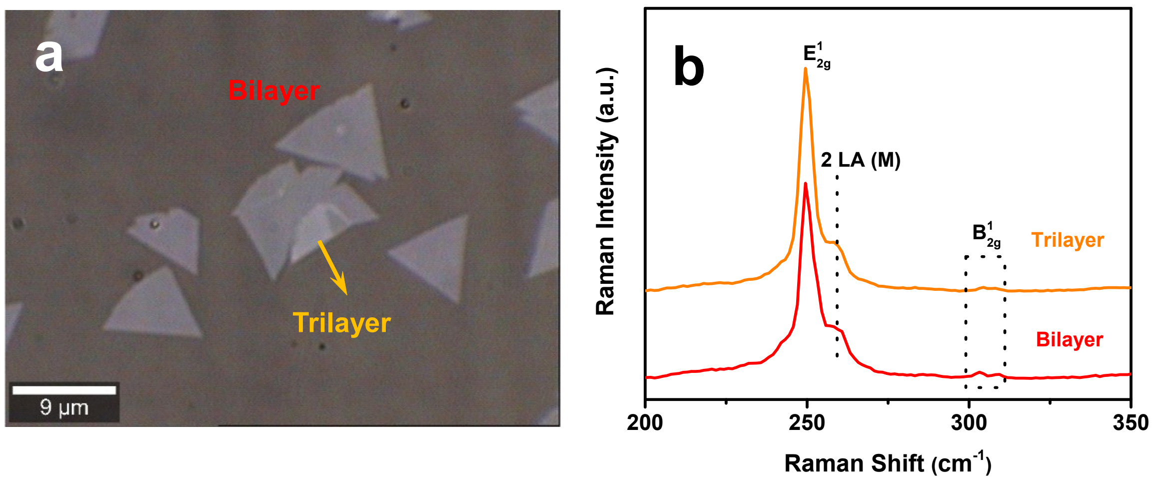

Figure S11. a, Optical micrograph of bilayer/trilayer WSe_2 crystals as-grown on c-plane sapphire substrate; **b,** The Raman spectra measurements for bilayer (red) and trilayer (orange) WSe_2 crystals.

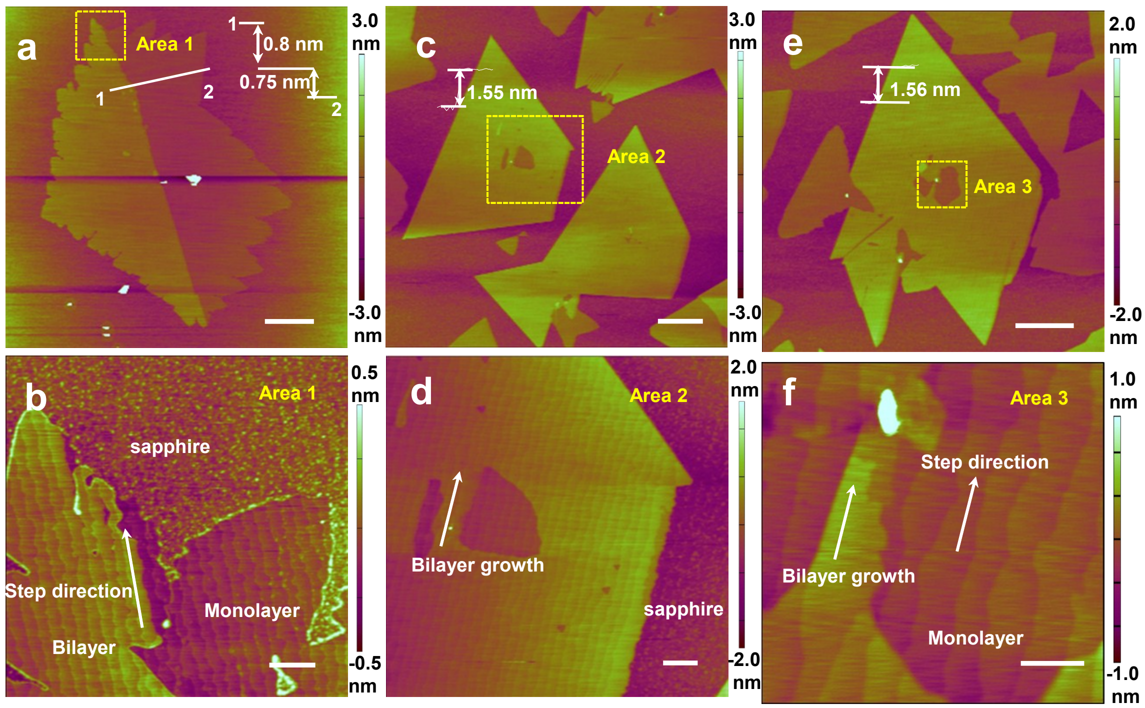

Figure S12. AFM topographic images of three representative bilayer WSe_2 crystals as-grown on sapphire surface. a, The bilayer WSe_2 crystal with irregular morphology. The inset height profiles were both ~0.8 nm, indicating a bilayer thickness in the left part and a monolayer thickness in the right part of the WSe_2 crystal; **b,** The zoom-in AFM image showed the bilayer WSe_2 nuclei growth orientation following the atomic steps; **c,** the bilayer WSe_2 crystal with truncated triangle morphology. The inset height profile demonstrated a bilayer thickness of WSe_2 crystal; **d,** The zoom-in AFM image showed bilayer WSe_2 nuclei growth orientation following the atomic steps; **e,** The bilayer WSe_2 crystal with grain boundary. The inset height profile demonstrated a bilayer thickness of WSe_2; **f,** The zoom-in AFM image showed bilayer WSe_2 nuclei growth orientation following the atomic steps. Scale bars: a, 2 μm; b, 200 nm; c, 1 μm; d, 200 nm; e, 1 μm; f, 100 nm.

Table S1. Roughness measurements on different surfaces

WSe_2 crystals	Sapphire Roughness	Monolayer Roughness	Bilayer Roughness	Trilayer Roughness	Sapphire Roughness (1050 ºC)[1]	Sapphire Roughness (1050 ºC)[2]
Sapphire (Fig. S7a)	-	-	-	-	0.056 nm	-
Sapphire (Fig. S7c)	-	-	-	-	-	0.060 nm
Crystal 1 (Fig. 2b)	-	0.070 nm	-	-	-	-
Crystal 2 (Fig. 2d)	0.109 nm	0.073 nm	0.068 nm	-	-	-
Crystal 3 (Fig. 3b)	0.129 nm	0.094 nm	0.097 nm	0.076 nm	-	-
Crystal 4 (Fig. S8b)	0.148 nm	0.056 nm	-	-	-	-
Crystal 5 (Fig. S10b)	0.110 nm	0.077 nm	0.082 nm	-	-	-
Crystal 6 (Fig. S10d)	0.125 nm	0.081 nm	0.073 nm	-	-	-
Crystal 7 (Fig. S10f)	-	0.075 nm	0.086 nm	-	-	-

[1]The as-supplied sapphire was annealed in the H_2/Ar for 15 mins;

[2]The as-supplied sapphire was annealed in the air for 60 mins.

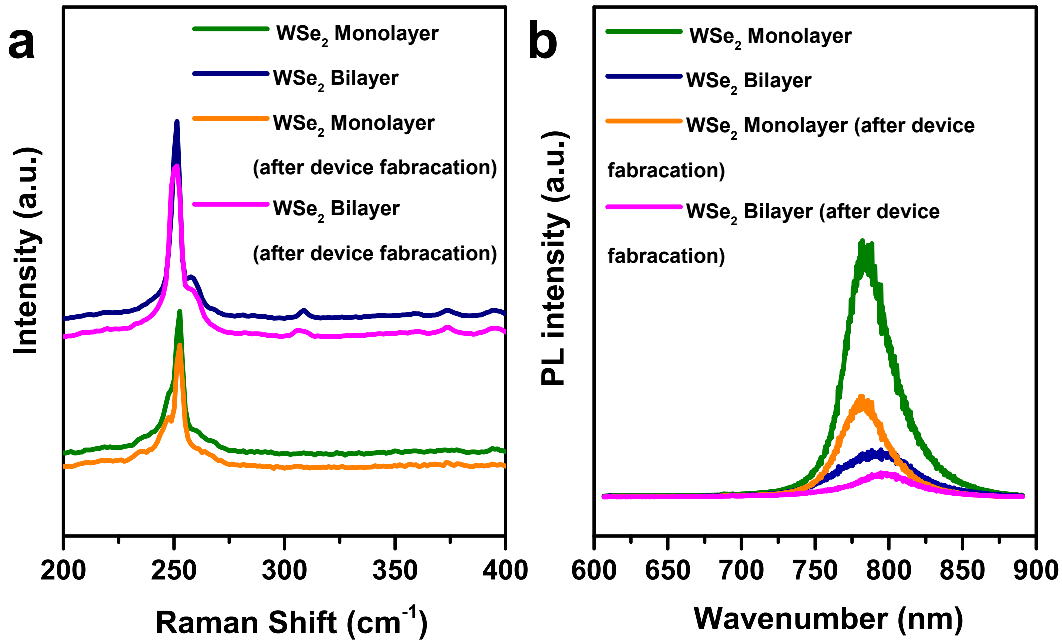

Figure S13. a. The Raman spectra measurements for WSe_2 monolayer and bilayer crystals before and after device fabrication on c-plane sapphire substrate; **b,** The PL spectra measurements for WSe_2 monolayer and bilayer crystals before and after device fabrication on c-plane sapphire substrate.

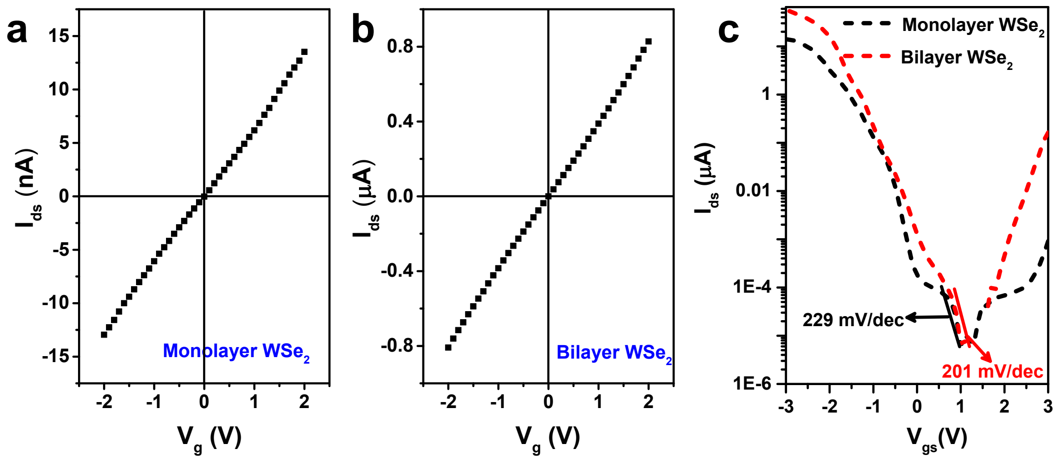

Figure S14. a, I_{ds} as a function of V_g for monolayer device with two terminal. I_{ds} as a function of V_g for bilayer device with two terminal; **b,** I_{ds} as a function of V_g for monolayer device. I_{ds} as a function of V_g for bilayer device. $C_g = 5.0\ \mu F/cm^2$; **c,** I_{ds} as a function of V_{gs} for monolayer/bilayer device. The subthreshold slopes of monolayer and bilayer crystals were measured to be 229 and 201 mV/dec, respectively.