

## Electronic Supplementary Information

### High-quality round-shaped monolayer MoS<sub>2</sub> domain and its transformation

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### **Round-shaped MoS<sub>2</sub> domain synthesis**

The CVD process was carried out in a furnace with two-independent-temperature zones, and a dual-tube configuration was built using two quartz tubes with the diameter of 10.0 cm and 3.5 cm, respectively. 3-5 mg MoO<sub>3</sub> powder (Alfa Aesar, 99.5%) and SiO<sub>2</sub> (300 nm)/n<sup>+</sup>-Si substrate which was precleaned with acetone and ethanol were put in a quartz boat, whilst 400 mg sulfur powder (ZNX, 99.999%) was upstream in another quartz boat. The precursors and substrate were both inserted into the inner tube. The distance between two precursors was 14 cm and that between the substrate and MoO<sub>3</sub> was 6 cm except specified elsewhere. Before the growth, the tube was pumped to a vacuum and then introduced of 500 sccm pure Ar (99.999%), which repeated three times for better purging. After purging, the temperature of MoO<sub>3</sub> was gradually increased from room temperature to the set temperature with the same rate (i.e., 750 °C in 40 min) and growth time was in the range of 5 min - 45 min with a 50 sccm Ar flow. After the growth, the furnace was cooled down to the room temperature with a normal cooling rate of about 10 °C/min if not specified elsewhere. The temperature of sulfur precursor was correspondingly adjusted.

### **MoS<sub>2</sub> Characterization**

Raman spectroscopy and photoluminescence (PL) spectroscopy were achieved on a Raman microscope (Bruker Optik GmbH, SENTERRA) with a 532 nm laser excitation and a laser power of 2 mW. The Raman and PL mapping images were obtained with a 100× objective and a 100 nm scanning step.

The SEM images were obtained on a Hitachi S4800 field-emission SEM system with an accelerating voltage of 3.0 kV. The high-resolution transmission electron microscopy operating at 200 kV (HRTEM, FEI Tecnai G2 F20) was used to study the morphology and crystal structure. The surface morphology of samples were characterized by an atomic force microscopy (Bruker Dimension Edge).

### **Devices Fabrication and Characterization**

Fabrication of the MoS<sub>2</sub> FET devices is as follows. The ar-p 5320 photoresist was spin-coated at 4000 rpm on the as-prepared samples and then was patterned by photolithography (SUSS MA6) and lift-off techniques. The deposition of the Cr (ca. 5 nm) adhesive layer and the Au (ca. 60 nm) source/drain electrodes was performed by an electron-beam evaporator (Angestron). The channel length and width of the FETs are 4.0 μm and 6.0 μm, respectively. Electrical characterization of the devices was performed using a Keithley 4200-SCS semiconductor parameter analyser (Keithley) in a vacuum (30 Pa) box after the devices had been annealed at 300 °C for 1h in an argon atmosphere.

### **Additional results**

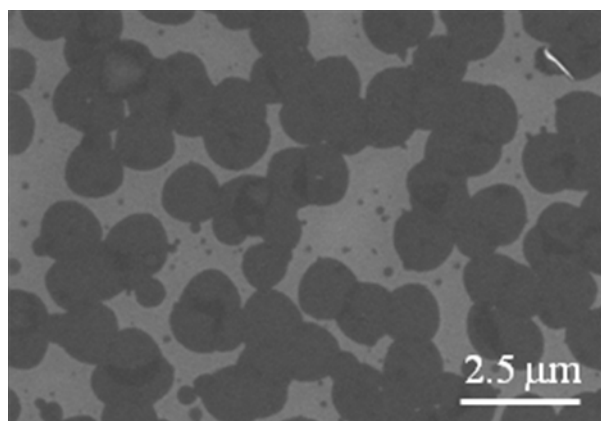


Figure S1. SEM image of round MoS<sub>2</sub> domains grown for 30 min at 750 °C, the same as that in Figure 2b.

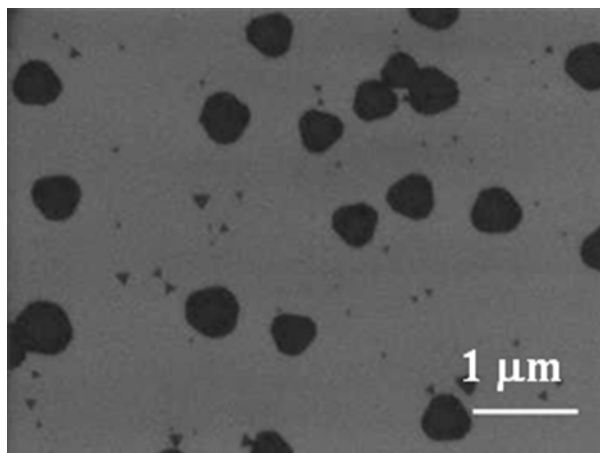


Figure S2. SEM image of truncated triangle (or nearly round) MoS<sub>2</sub> domains grown for 5 min at 750 °C, the same as that in Figure 5b.

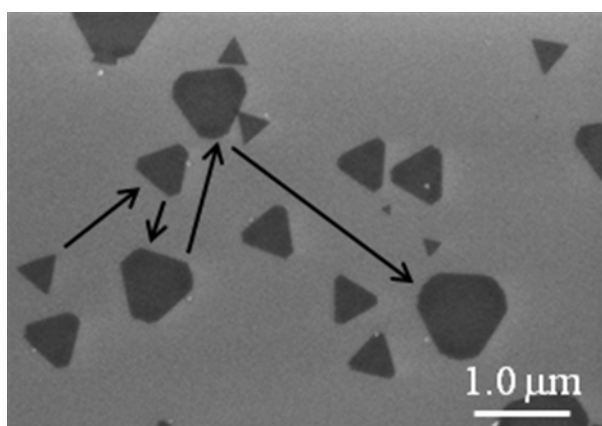


Figure S3. SEM image of different-shaped MoS<sub>2</sub> domains grown for 20 min at 750 °C with only large quartz tube, the same as that in Figure 5d.

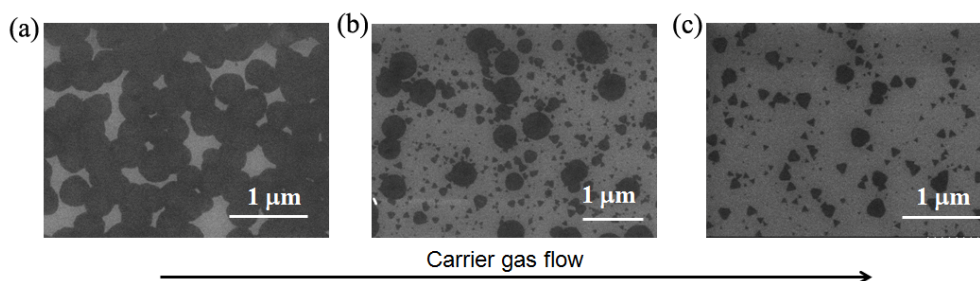


Figure S4. Shape evolution as a function of the distance between the the substrate and the MoO<sub>3</sub> precursor and. (a) 4 cm of the substrate away from the MoO<sub>3</sub>, (b) 8 cm of the substrate away from the MoO<sub>3</sub>, (c) 10 cm of the substrate away from the MoO<sub>3</sub>. With the increase of the distance, the concentration of reactants decreased, and the growth regime gradually shifted to thermodynamically favored process. The domain shape evolved from round to triangle, and the size and density decreased simultaneously.

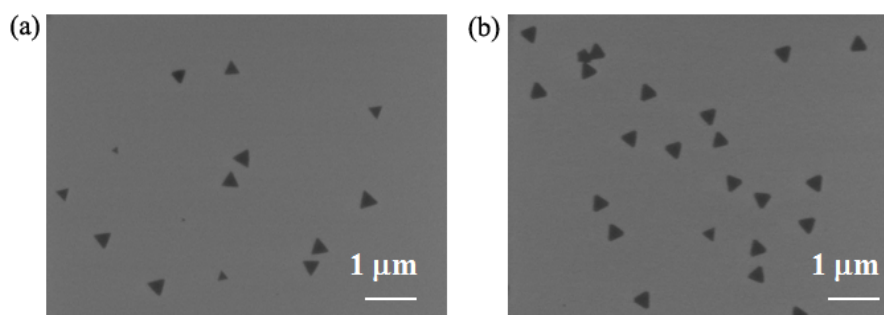


Figure S5. SEM image of triangle MoS<sub>2</sub> domains grown for 40 min at 710 °C.