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Electronic Supplemental Information (ESI)
for

Molten-droplet-driven growth of MoS_2 flakes with controllable morphology transition for hydrogen evolution reaction

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1 Experiment parameters

Table. S1 The experiment parameters of the MDD growth of MoS_2 on Si/SiO_2 surfaces by chemical vapor deposition.

C_{NaOH} (mM)	M_{MoO_3} (mg)	Heating up time (min)	Growth time (min)	growth temperature (°C)	Gas flow (sccm)
200	80	20	10	800	200
200	80	20	10	820	200
200	80	20	10	840	200
200	80	20	10	860	200
200	80	20	10	880	200

2 Supplementary Figures

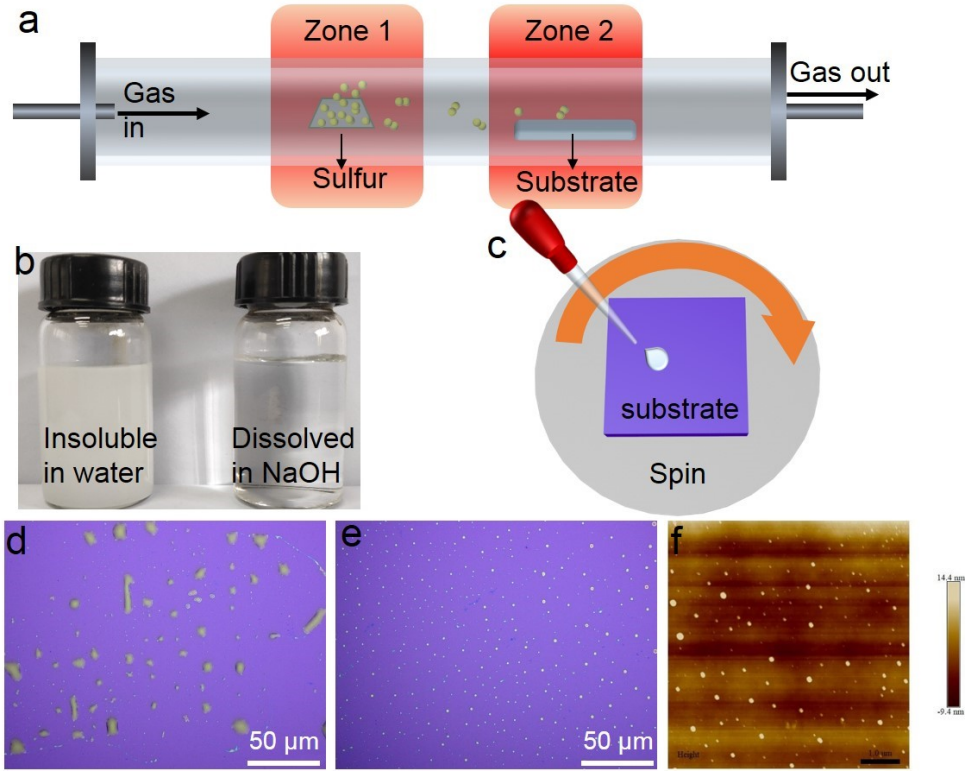


Fig. S1 (a) Schematic diagram of the dual-temperature CVD system for synthesizing MoS_2 domains. (b) Optical images of MoO_3 powder dissolved in NaOH solutions, showing the solubility of MoO_3 in NaOH. (c) Schematic diagram of liquid-precursor uniformly spin-coated on SiO_2/Si for the growth. (d) Optical image of liquid precursor spun-coated on SiO_2/Si substrate. (e) Optical and (f) AFM topography image of the liquid precursor spun-coated on the SiO_2/Si substrate after drying.

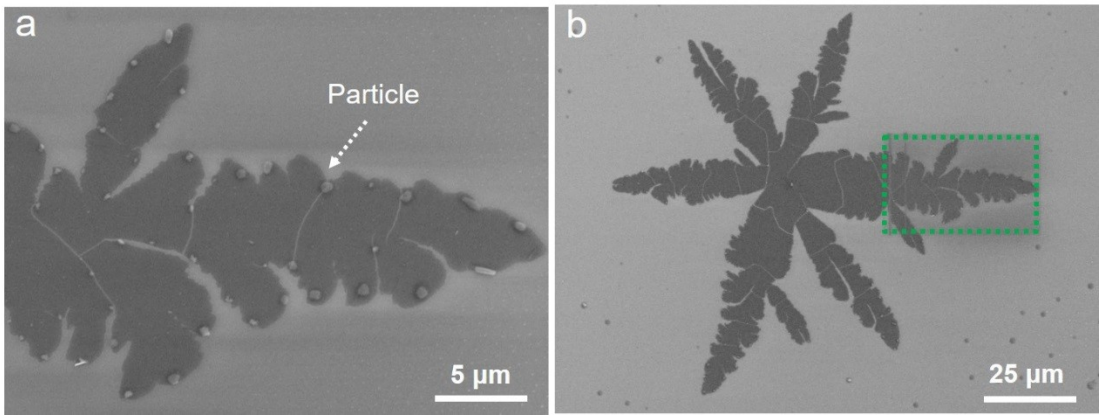


Fig. S2 (a) High-magnified SEM image of the spike taken from the green frame in (b) low-magnification SEM of a MDD-grown MoS_2 flake at growth temperature of 880°C, showing that the edges of the MoS_2 flakes are uniformly surrounded by solidified droplets.

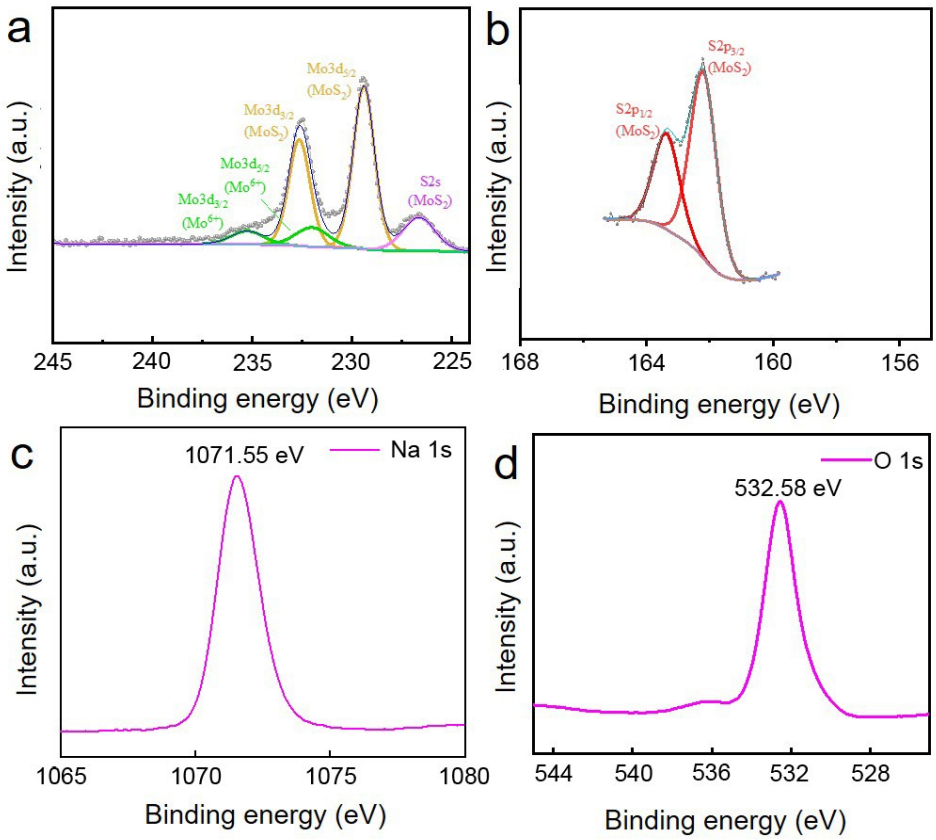
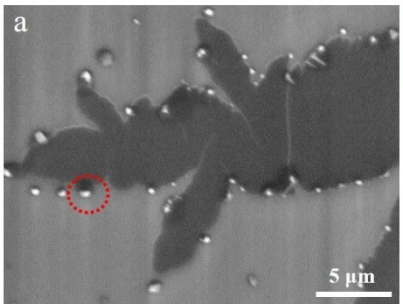


Fig. S3 XPS spectra of the MDD-grown MoS_2 flakes, showing (a) Mo 3d, (b) S 2p, (c) Na 1s, and (d) O 1s XPS peak.



Element	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)
Mo	5.03	0.83	21.52	2.01
S	10.08	0.94	13.84	0.99
O	81.52	3.56	60.32	0.85
Na	3.37	1.55	4.32	1.78

Fig. S4 (a) High-magnification SEM of a MDD-grown MoS_2 edge at growth temperature of 880°C, and (b) the corresponding EDS analysis for the particles denoted as the red circle in (a). Please note that here the irrelevant elements (Si and C) are not included for the EDS analysis.

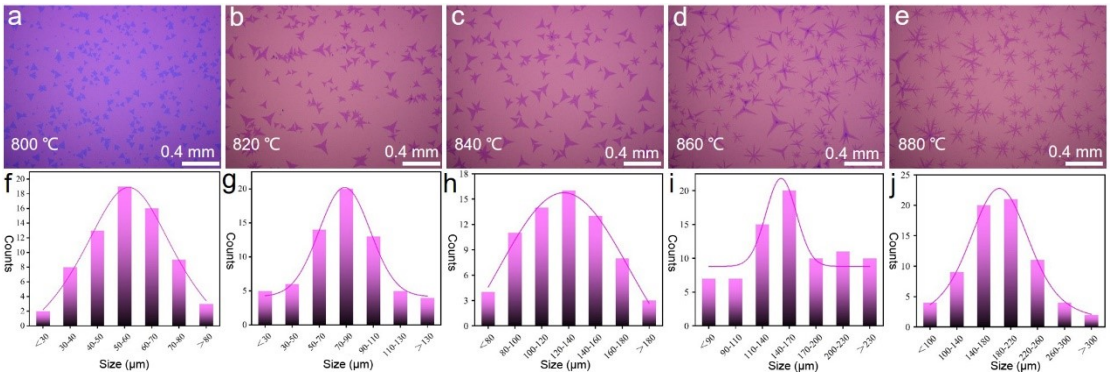


Fig. S5 (a-e) Optical images of MDD-grown MoS_2 flakes at growth temperatures of (a) 800, (b) 820, (c) 840, (d) 860, and (e) 880 °C, respectively. (f-j) The corresponding lateral size statistics of MoS_2 flakes in (a-e).

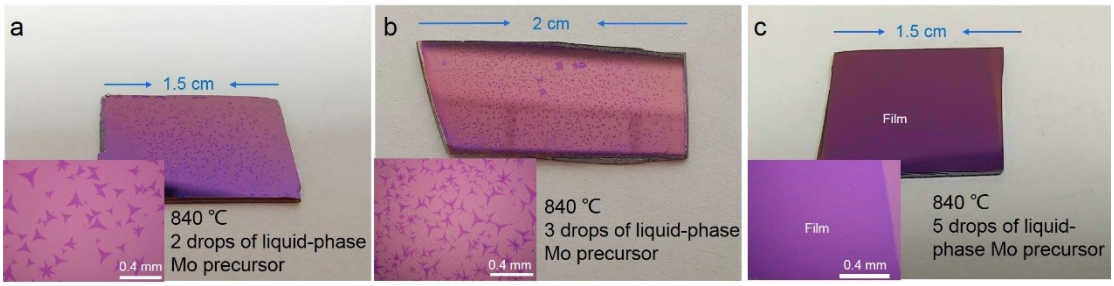


Fig. S6 (a-c) Optical image of MoS_2 flakes on SiO_2/Si substrate grown by MDD growth with different Mo precursor amount at fixed 840 °C and 10 min growth. The insets are randomly taken from the corresponding substrate.

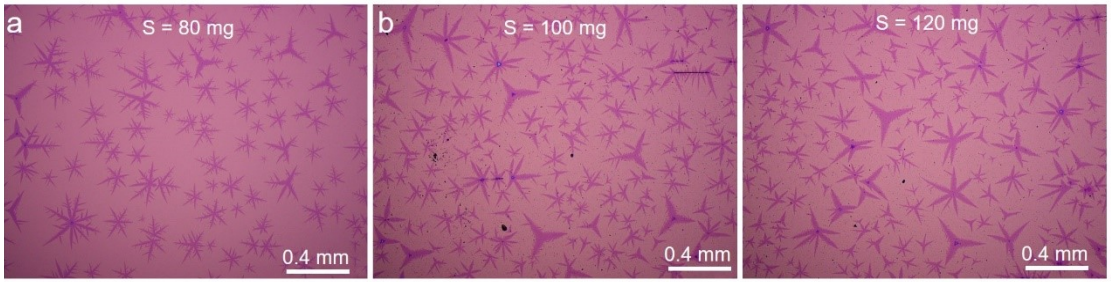


Fig. S7 (a-c) Optical images of MoS_2 flakes on SiO_2/Si substrate grown by MDD growth with different S precursor amount at fixed 880 °C and 10 min growth.

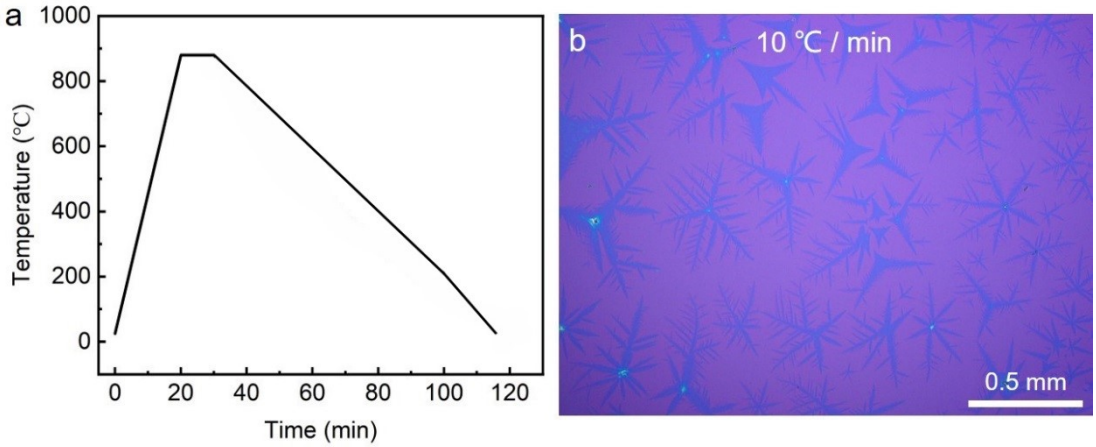


Fig. S8 (a) The temperature curve with a slow cooling rate of 10 °C/min in the MDD growth process. (b) Optical images of MoS_2 crystals at a growth temperature of 880 °C and a cooling rate of 10 °C /min with otherwise identical conditions.

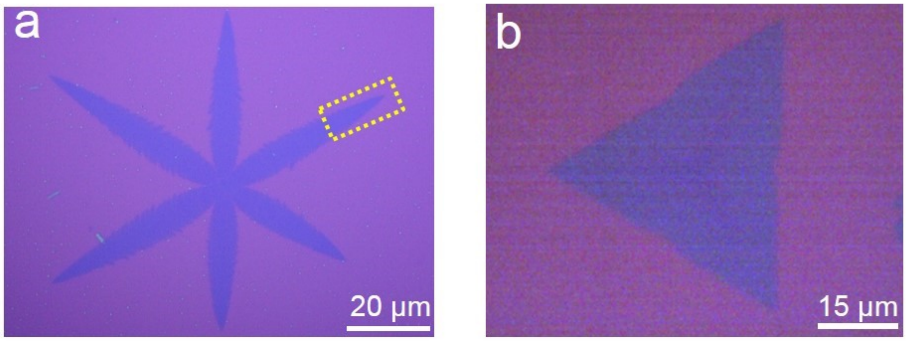


Fig. S9 (a) Optical image of dendritic MDD-grown MoS_2 flake for the optical properties characterization. The inset yellow frame is the area for Raman and PL mapping measurements shown in Fig. 3c and e. (b) Optical image of triangular MDD-grown MoS_2 flake for the optical properties characterization.

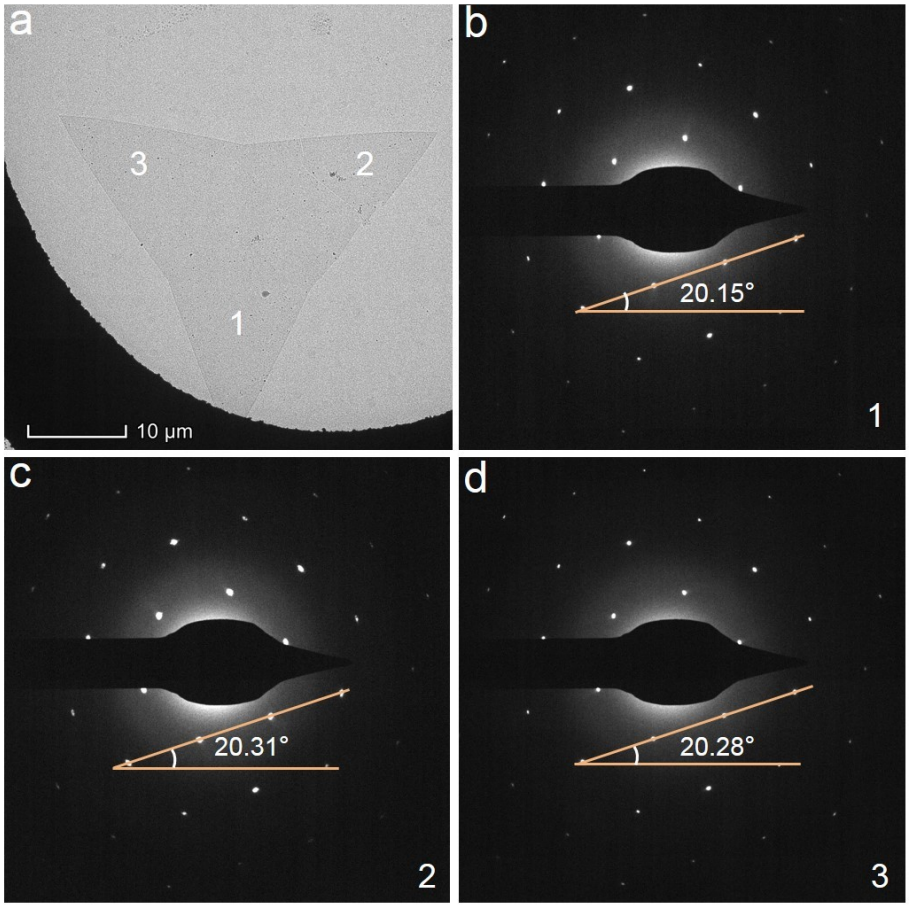


Fig. S10 (a) TEM images of the triangle-shaped MoS_2 flake transferred on copper grid. (b-d) Selected area electron diffraction (SAED) patterns for selected regions at different locations marked as 1, 2, and 3 in (a).

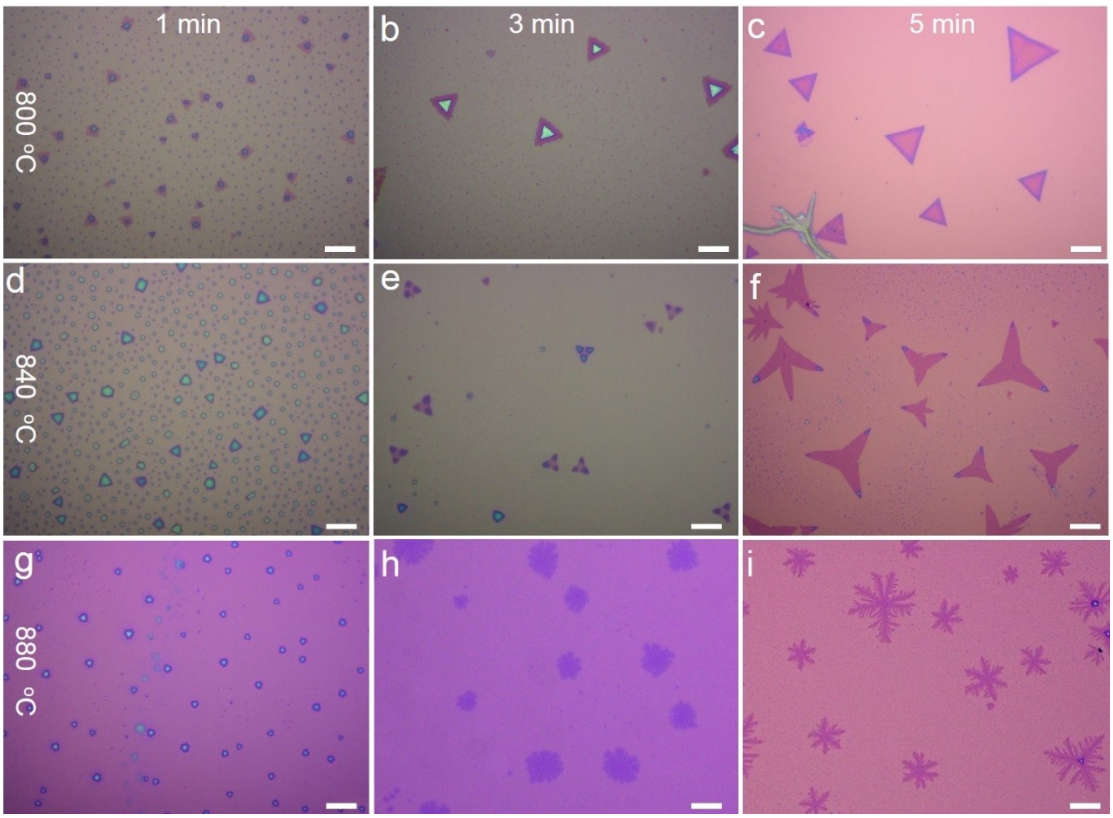


Fig. S11 (a-c) Low-magnification optical images of MDD-grown MoS_2 flake on substrate for 1, 3, and 5 min growth at 800 °C. (d-f) Low-magnification optical images of MDD-grown MoS_2 flake on substrate for 1, 3, and 5 min growth at 840 °C. (g-i) Low-magnification optical images of MDD-grown MoS_2 flake on substrate for 1, 3, and 5 min growth at 880 °C. All the scale bar is 20 μm.

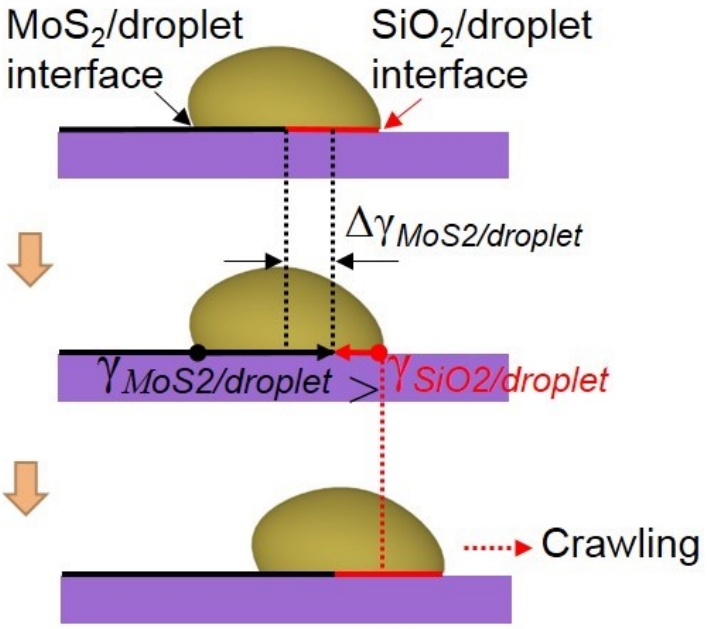


Fig. S12 Schematic of driven molten droplet with crawling movement during the growth of MoS_2. $\gamma_{MoS2/liquid}$ and $\gamma_{SiO2/liquid}$ are the interface energies of solid MoS_2/ and SiO_2/liquid droplet, respectively. $\Delta\gamma_{MoS2/liquid}$ is the interface energy of solid MoS_2/liquid droplet of accretion growth of MoS_2.

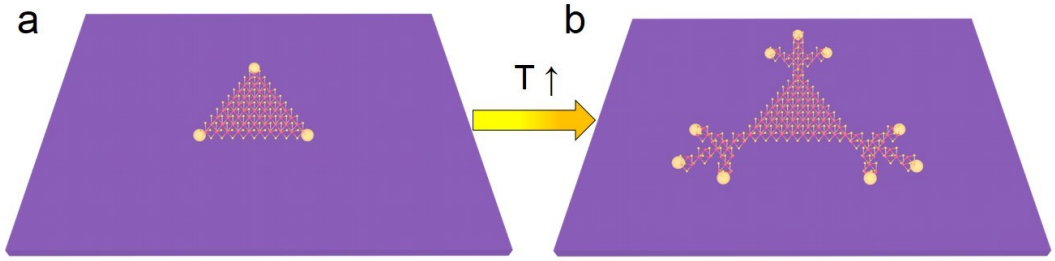


Fig. S13 (a, b) Schematic diagram of a MoS_2 flake grown at an initial growth temperature of 840°C (a) for 4 minutes and then rapidly ramped up to 880°C (b) for 5 minutes.

Supplementary Note

Calculation of the fractal dimension of MoS_2 flakes. In order to quantify the complexity of the MoS_2 morphology under different experimental conditions, we used box-counting to evaluate it. In this method, the image is filled with boxes of a certain number of pixels, and the number of pixels in the boxes is calculated by the size of the Fig.. The number of boxes for each filled Fig. is analyzed after enlarging and shrinking the Fig. by a certain scale. Therefore, we use the following method to calculate:

$$Log(N) = -DLog(s) + Log(C)$$

where N is the number of boxes, s is the scale of the graph, and D is the fractal dimension. The higher the fractal dimension, the more complex the graph structure.