Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

Liquid-phase Catalyst Pre-seeding for Controlled Growth of Layered MoS_2 Films over a Large Area via Chemical Vapor Deposition

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Experimental

Sample Synthesis

We employed a catalyst pre-seeding, liquid-phase Chemical Vapor Deposition (CVD) approach for the synthesis of MoS_2. Initially, we subjected the SiO_2/Si substrate to an ultrasonic cleaning process in acetone and isopropyl alcohol, effectively removing surface organic impurities. The clean substrate was then prepared for precursor application via spin coating, followed by an O_2 plasma treatment at 30W for 30 seconds, which converted the hydrophobic surface into a hydrophilic one.

The liquid-phase mixture, comprising transition metal precursors MoO_3 (Sigma-Aldrich,

99.99%) and sodium chloride (Sigma-Aldrich, 99%) in ammonia at varying concentrations, was then spun onto the SiO_2/Si substrate at 3,000 rpm for 60 seconds. This coated substrate was then dried on a hot plate at 80 ℃ for 5 minutes. This coating process was repeated until the desired number of seeds per unit area was achieved.

The prepared substrate and 500mg of S powder (Sigma-Aldrich, 99.99%) were positioned in Zone-1 and Zone-2 of a CVD furnace, respectively. The substrate in Zone-1 was heated to the growth temperature (700-800 ℃) over 14 minutes, while Zone-2 was maintained at 160 ℃. Upon reaching 750 ℃ in Zone-1, Zone-2 was held at 160 ℃ for an additional 8 minutes in a 25 sccm Ar and 4 sccm H_2 atmosphere. Finally, the furnace was naturally cooled to 25 ℃ under an Ar gas atmosphere at 25 sccm (Fig. S1).

Characterization of MoS_2

We employed a suite of characterization tools to examine the structural and compositional properties of the grown and transferred MoS_2. Optical Microscopy (OM) images were captured using an Olympus DX51 microscope. The surface morphology was examined through Scanning Electron Microscopy (SEM; JEOL JSM7401F). The microstructure of the samples was analyzed using Transmission Electron Microscopy (TEM; FEI, Tecnai G2 F20, 200 kV). We evaluated the thickness and surface topography via Atomic Force Microscopy (AFM, Veeco, Dimension 3100), while Raman spectra were obtained using micro-Raman microscopy (Renishaw, India Basic) with a 532 nm laser. Lastly, X-ray

Photoelectron Spectroscopy (XPS; Thermo Scientific, ESCALAB 250Xi) with a Mg K_α X-ray source was used to scrutinize the chemical bonding characteristics.

Fabrication of MoS_2 FETs

We fabricated back-gated Field-Effect Transistors (FETs) using a conventional photolithography process, following established literature. We used the transferred MoS_2 as the electrode, depositing layers of Cr and Au of 20 nm and 50 nm thickness, respectively. The field-effect charge mobility was then measured using a Keithley SCS-4200 equipped probe station, providing valuable insights into the electrical properties of the fabricated FETs.

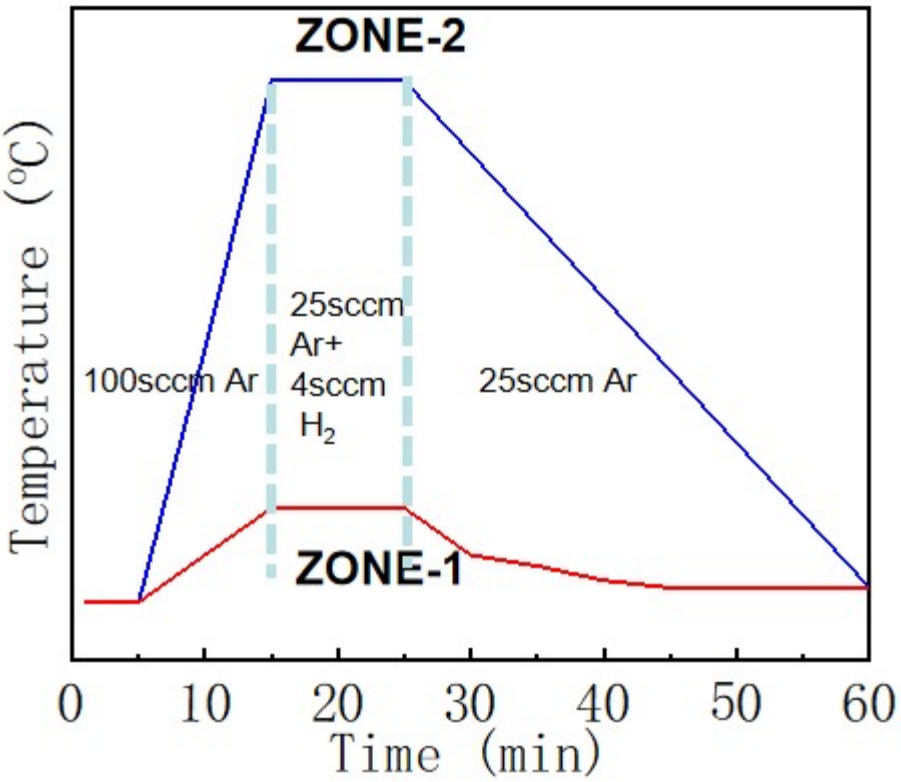
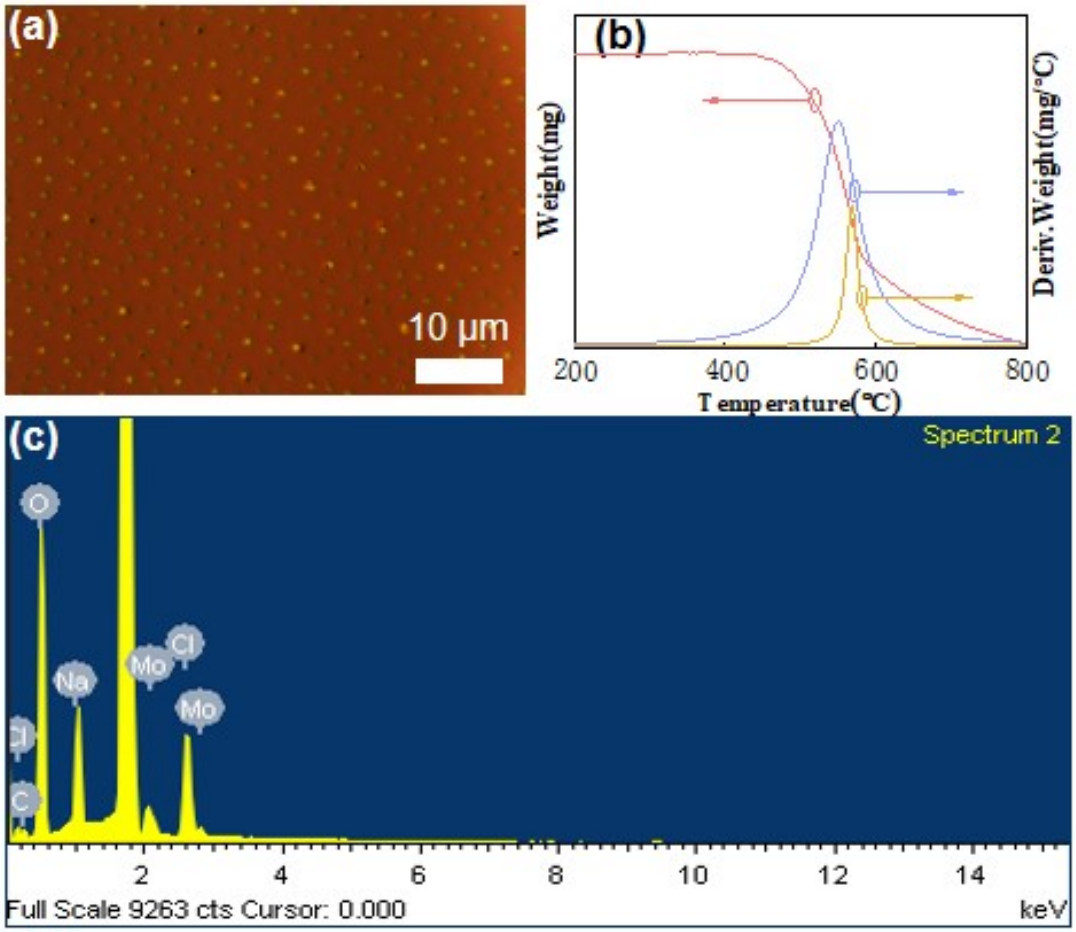


Figure. S1 Schematic representation of the gas flow dynamics during the CVD process. The chart correlates the growth control time with the corresponding temperature, capturing the crucial phases of the growth process, enabling the synthesis of high-quality MoS_2.



(d)

Element	Weight%	Atomic%
C K	7.59	11.30
O K	62.02	70.21
Na K	13.98	10.87
Cl K	14.31	7.22
Mo L	2.09	0.40
Totals	100.00	

Figure. S2 Detailed characterization of seed precursors. (a) An Optical Microscopy (OM) image of the seeds, devoid of the sulfur source, grown at a temperature of 750 °C, illustrating the morphology without sulfur integration; (b) Thermogravimetric Analysis (TGA) data of the seeds, with a Mo^{+}: Na^{+} molar ratio of 1:7, demonstrating the stability and decomposition characteristics; (c) Energy Dispersive X-ray Spectroscopy (EDS) spectrum of the seeds, highlighting the elemental distribution and abundance; (d) Elemental composition data for the seed solution, indicating the stoichiometry of the precursor mix.

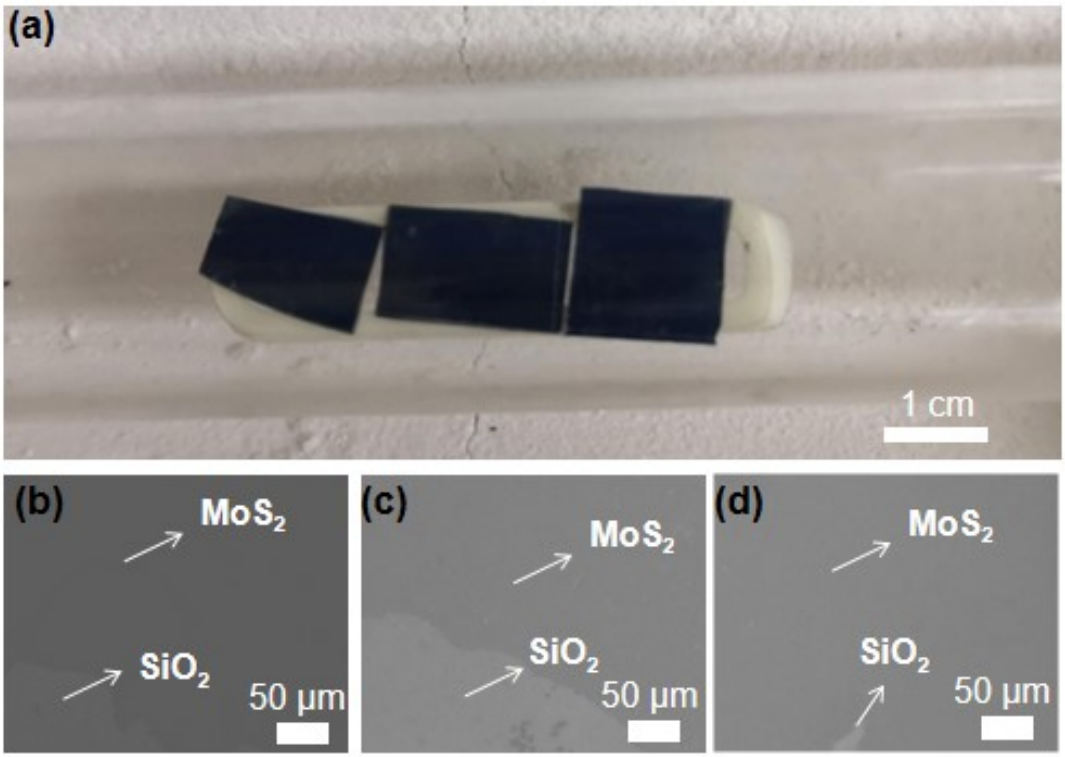


Figure. S3: Post-growth characterization of MoS_2 samples. (a) A photograph of the clean chamber following the CVD growth, exhibiting an absence of residual contaminants; (b), (c), and (d) SEM images of MoS_2 samples after the growth process, revealing the surface morphology and structure at a microscale level.

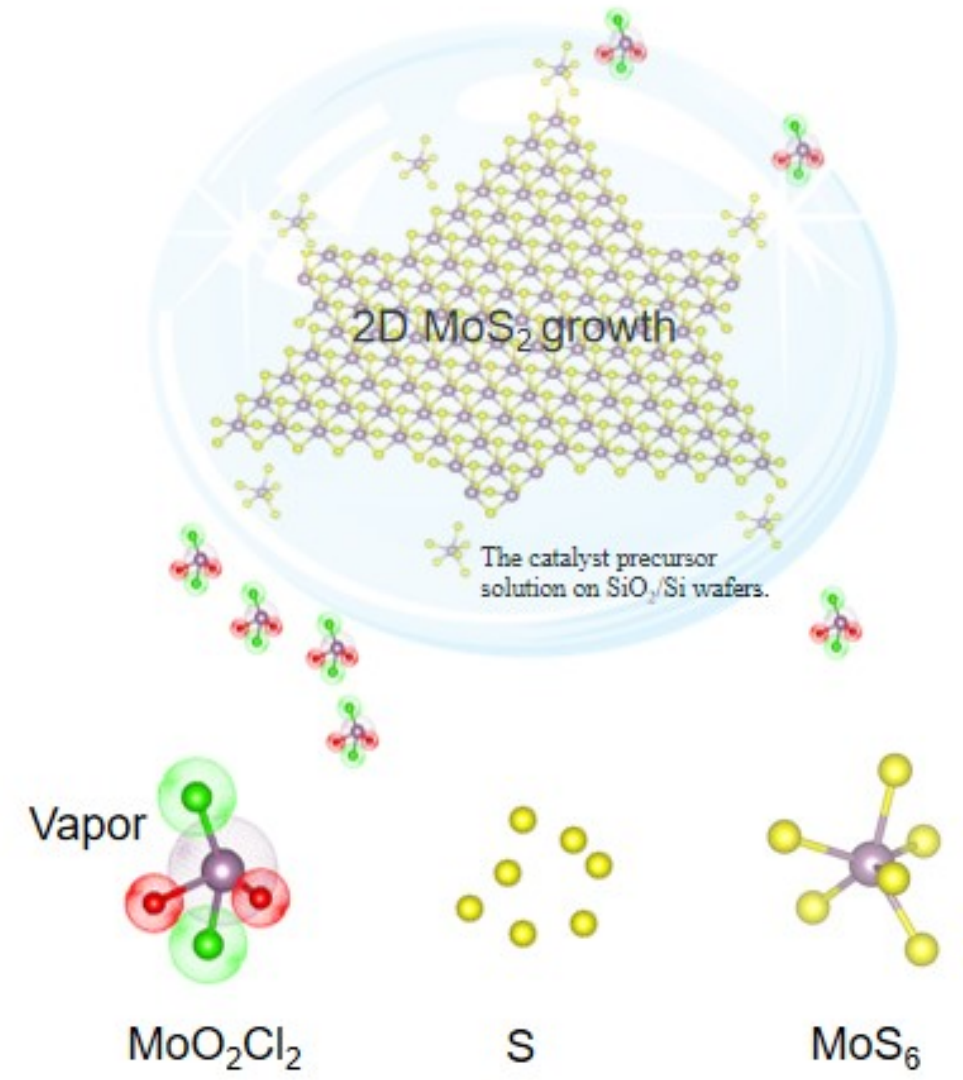


Figure. S4: Schematic illustration of the rapid and large-scale growth mechanism of MoS_2. This figure elucidates the process that allows for rapid growth over a large area, enabling the production of extensive MoS_2 films, a critical feature for commercial applications.

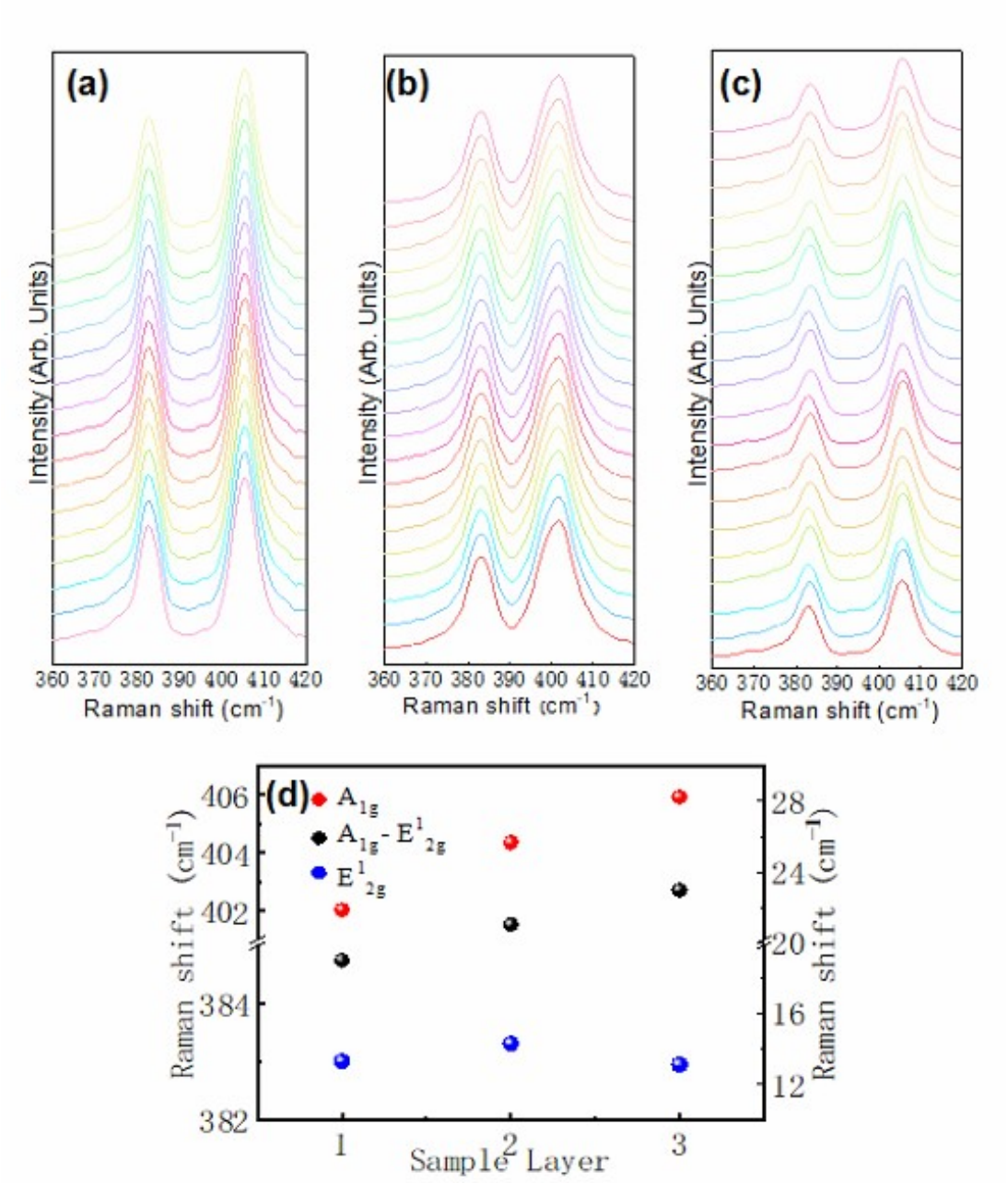


Figure S5: Raman spectroscopic investigation of MoS_2 films at different stages. (a) Raman spectra of MoS_2 film monolayers at 20 distinct points in the film-covered area, showing the vibrational properties of single-layer MoS_2; (b) Raman spectra of MoS_2 film bilayers at various locations, providing insight into the interaction between layers; (c) Raman spectra of MoS_2 film trilayers, illustrating the change in vibrational properties with increasing layer number; (d) Statistical analysis of Raman peak positions for monolayer, bilayer, and trilayer MoS_2, showcasing the shifts due to interlayer interactions.

Table S1 Comparison between literature and experimentally determined electronic properties for the MoS_2 monolayer.

Mobility	On/Off Ratio	Reference
0.1	10^2	[41]
9.6	10^5	[42]
5.9	10^5	[43]
22	-	[44]
32.1	10^8	**Present study**