

Supporting Information

MoS₂-OH Bilayer Mediated Growth of Inch-Sized Monolayer MoS₂ on Arbitrary Substrates

*Juntong Zhu^{1, #}, Hao Xu^{2, #}, Guifu Zou^{1, *}, Wan Zhang¹, Ruiqing Chai³, Jinho Choi¹,
Jiang Wu^{2, *}, Huiyun Liu², Guozhen Shen³, Hongyou Fan^{4, 5, *}*

¹College of Energy, Soochow Institute for Energy and Materials Innovations, and Key Laboratory of Advanced Carbon Materials and Wearable Energy Technologies of Jiangsu Province Soochow University, Suzhou 215006, China;

²Department of Electronic and Electrical Engineering, University College London, Torrington Place, London WC1E 7JE, UK;

³State Key Laboratory for Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, China;

⁴Center for Integrated Nanotechnology, Sandia National Laboratory, Albuquerque, NM 87185, United States

⁵Chemical and Biological Engineering, Center for Micro-Engineered Materials, University of New Mexico, Albuquerque, NM 87122, United States

* Corresponding author(s) to whom correspondence should be addressed:

Guifu Zou: zouguifu@suda.edu.cn; Jiang Wu: jiang.wu@ucl.ac.uk; Hongyou Fan: hfan@sandia.gov

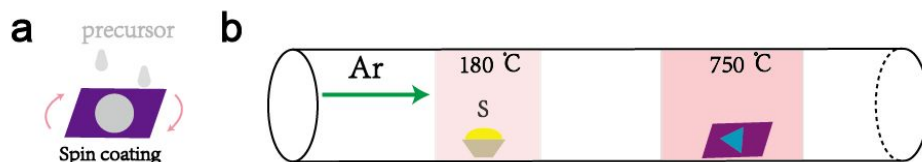


Figure S1. Schematic illustration of the MoS₂ growth process including (a) spin-coating precursor solution on the substrate and (b) Annealing process in the tube furnace.

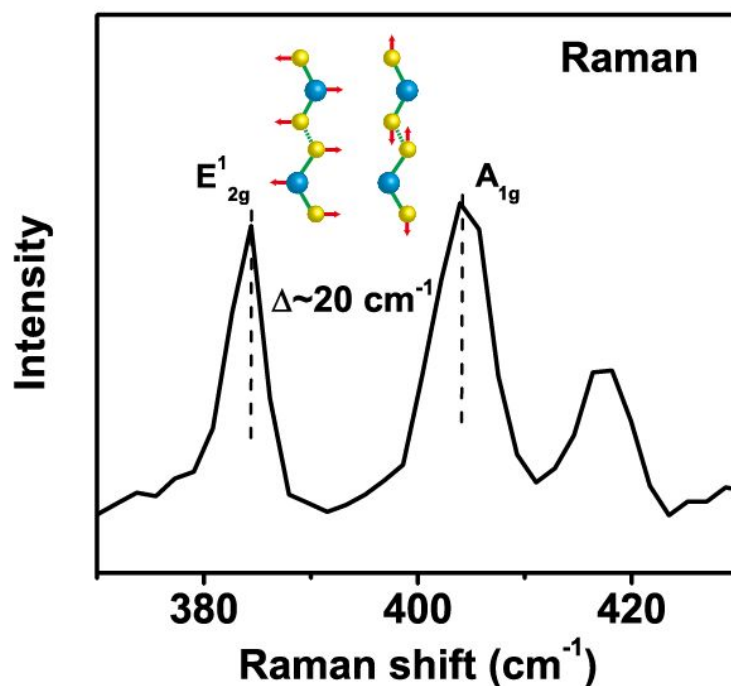


Figure S2. Raman spectra of the randomly selected position in the as-grown MoS₂ film. Raman modes of E_{2g}^1 and A_{1g} are located at 384.3 cm⁻¹ and 404.6 cm⁻¹, respectively, suggesting the monolayer nature. The peak at 418 cm⁻¹ is resolved to the sapphire substrate.

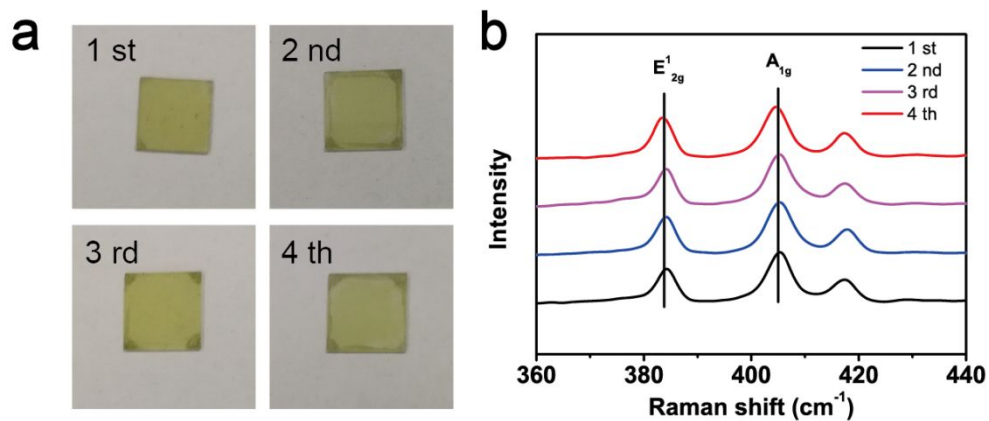


Figure S3. Photographs and Raman spectra of as-grown MoS₂ film on the sapphire substrate reused for four times. (a) Photographs show uniform films, (b) Raman spectra of four MoS₂ samples, E_{2g}^1 and A_{1g} are located at ~ 384 cm⁻¹ and ~ 404 cm⁻¹, respectively, corresponding to the monolayer.

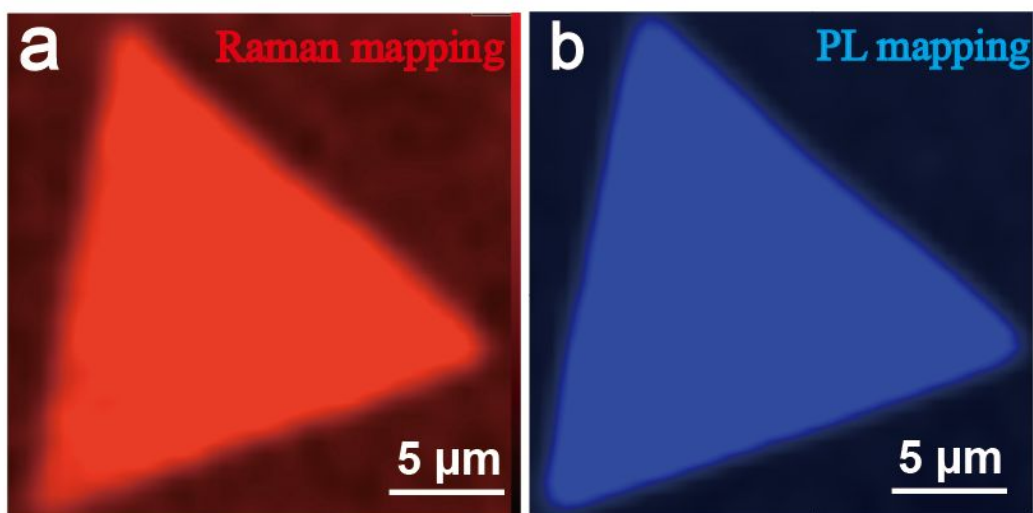


Figure S4. The monolayer MoS₂ keeps good uniformity in thickness. (a) Raman intensity map of A_{1g} peak, (b) PL map of the A-exciton.

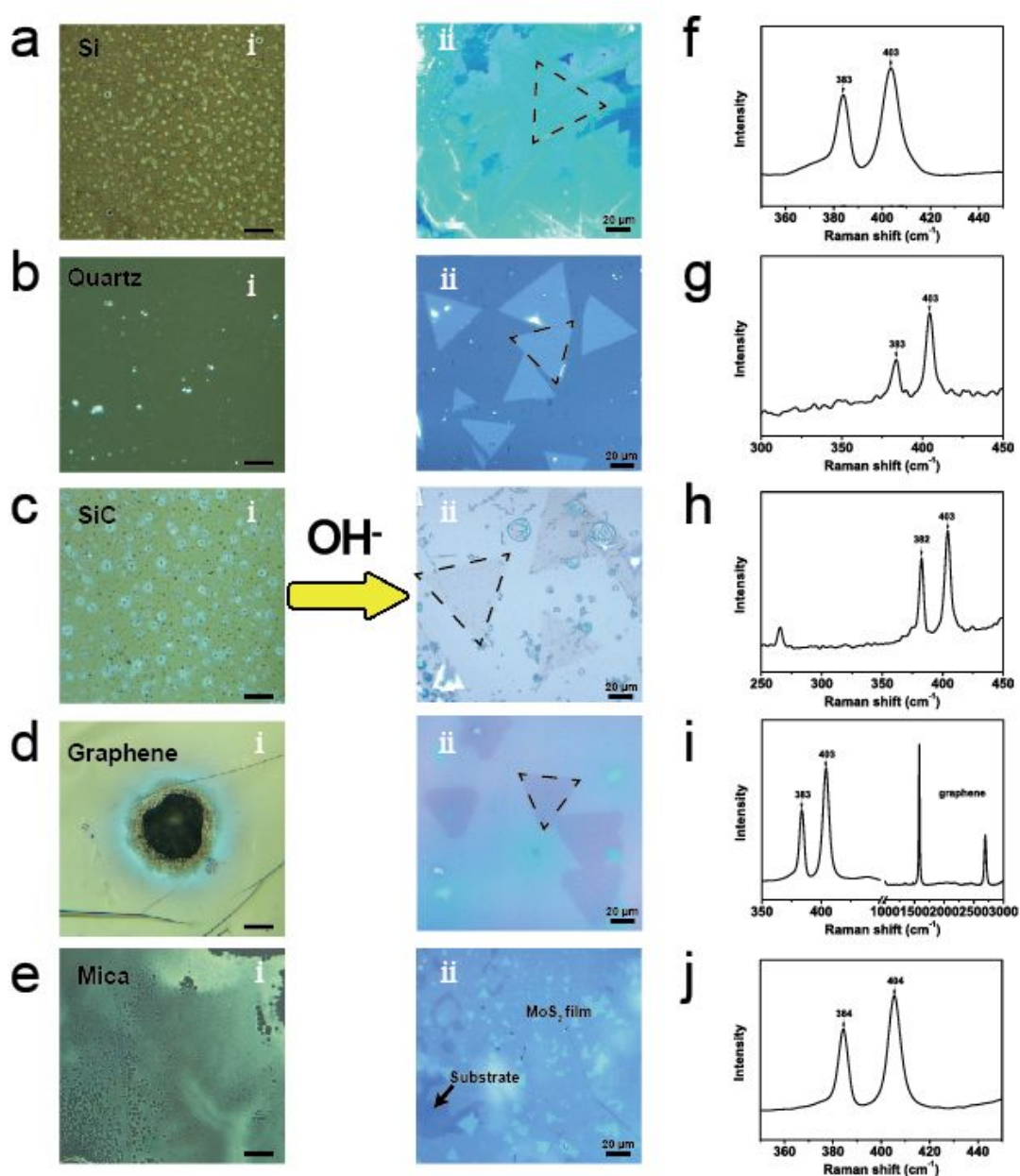


Figure S5. Substrate-independent growth of monolayer MoS₂. Optical microscopy (OM) images of MoS₂ grown on diverse substrates without (i) and with -OH (ii), respectively. (a) Si, (b) quartz, (c) SiC, (d) graphene, (e) mica. (f - j) Corresponding Raman spectra of as-grown MoS₂ with -OH on diverse substrates from ii, respectively.

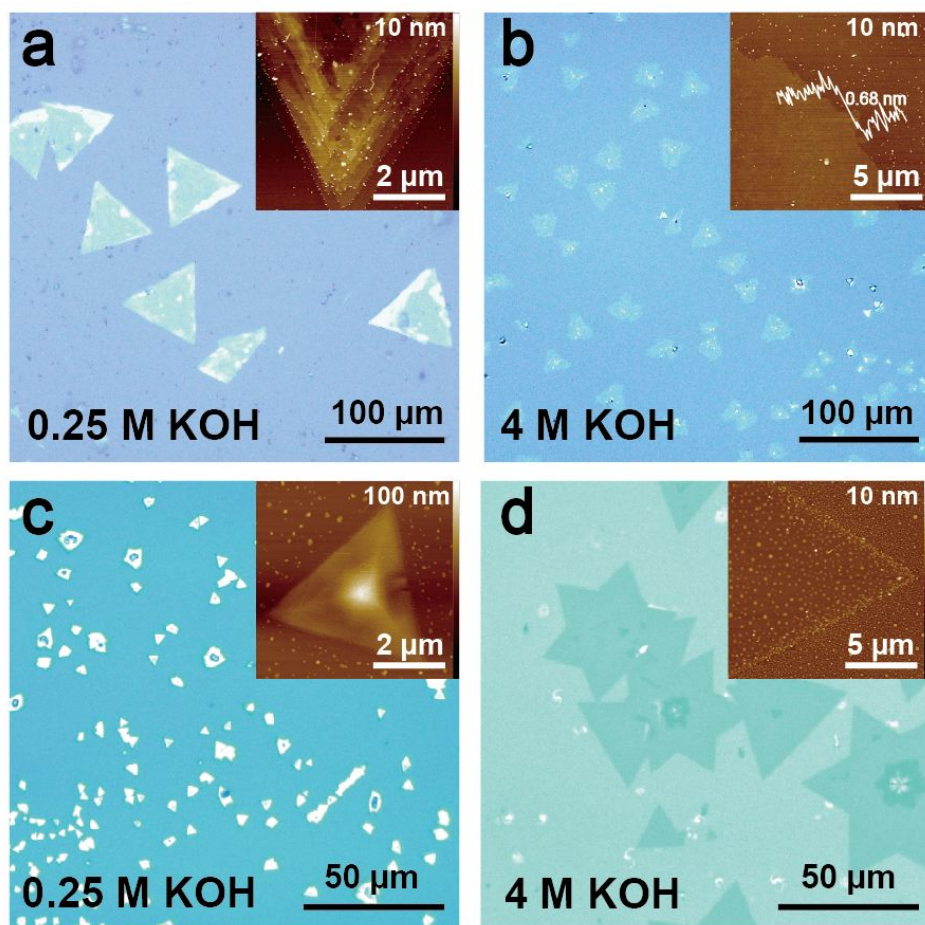


Figure S6. Insufficient and excess amount of -OH in the growth of monolayer MoS₂ (a, b) MoS₂ grown on sapphire with 0.25 and 4 M KOH, respectively. (c, d) MoS₂ grown on SiO₂ substrate with 0.25 and 4 M KOH, respectively.

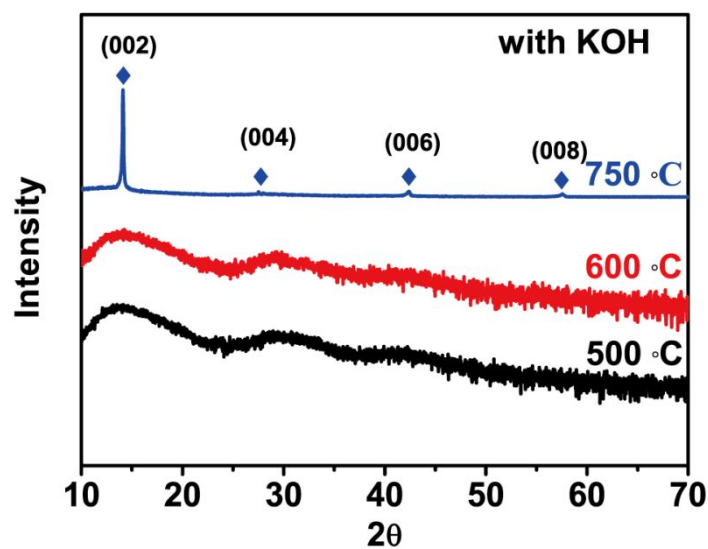


Figure S7. XRD patterns of the as-grown MoS₂ samples at different annealing temperature. MoS₂ samples have poor crystal quality at 500 and 600 °C but high-quality crystallinity at 750 °C.

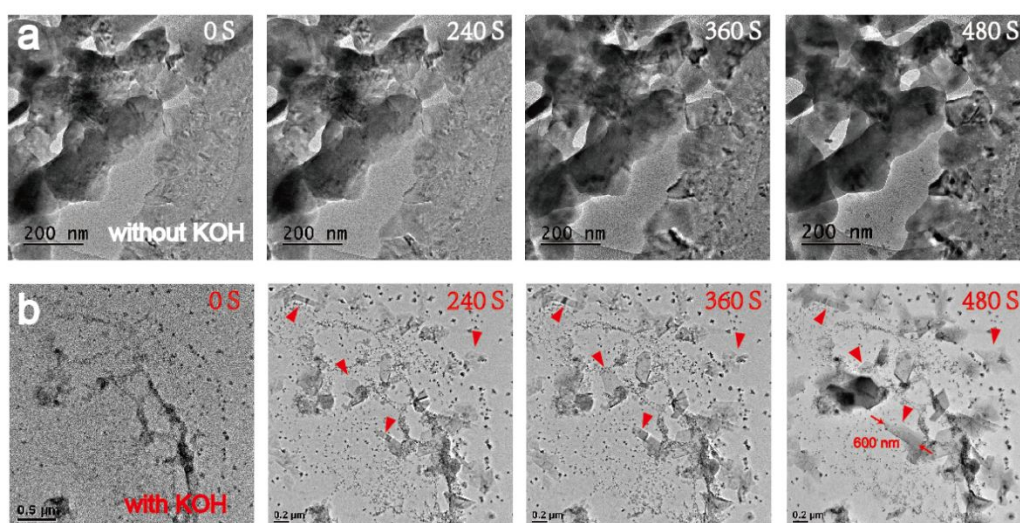


Figure S8. Long-time-resolved TEM images of the growth of MoS₂ at 750 °C. As a comparison, (a) MoS₂ grown in precursor solution without KOH, (b) MoS₂ grown in 1 M KOH precursor solution. The length of flake increased from 0 to 600 nm.

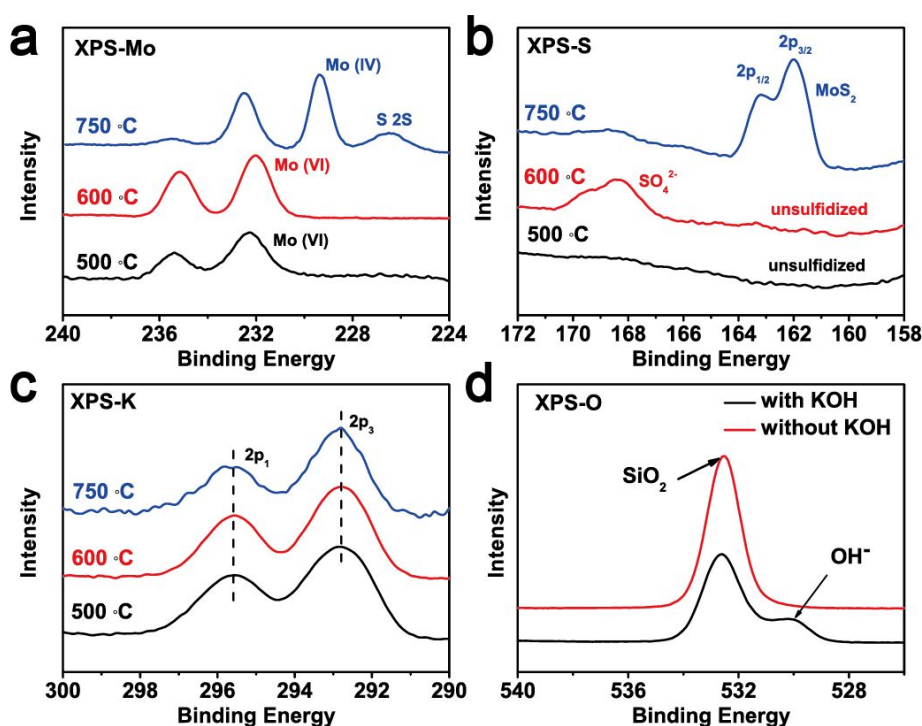


Figure S9. XPS spectra of as-grown MoS₂ samples at different temperature in annealing process. (a) Mo 3d, (b) S 2p, (c) K 2p, (d) O peak of the achieved MoS₂.

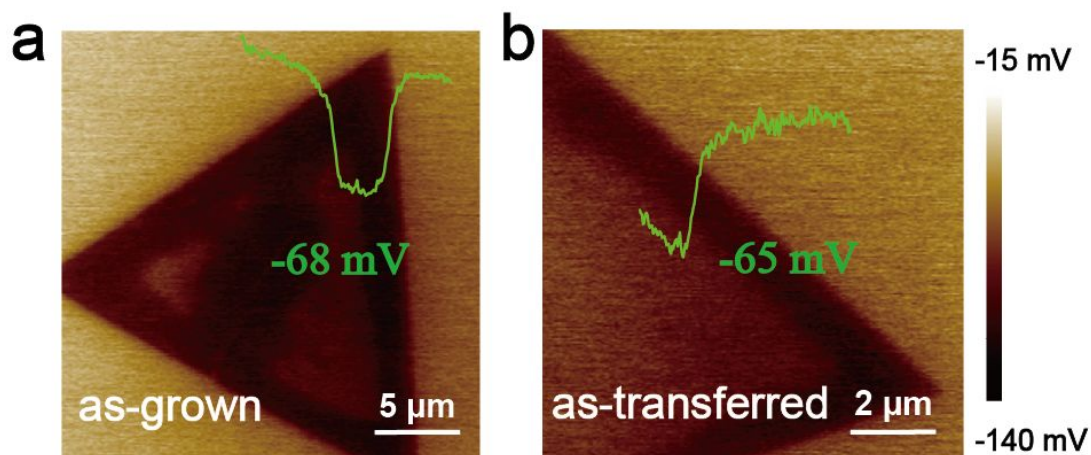


Figure S10. KPFM images of direct growth and as-transferred MoS₂ monolayers, respectively.

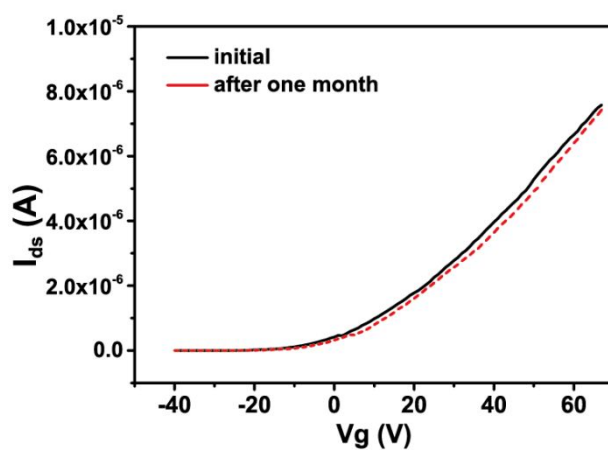


Figure S11. Transfer characteristics of the device before (black line) and after one month (red line) in air.

Supplementary Movie 1. The evolution of SAED patterns during the crystallization process.

Supplementary Movie 2. dynamical behavior of the increased flake at 750 °C.