

Chemical vapor deposition merges MoS₂ grains into high-quality and centimeter-scale films on Si/SiO₂

Mukesh Singh^a, Rapti Ghosh^{b,c,d}, Yu-Siang Chen^c, Zhi-Long Yen^a, Mario Hofmann^a, Yang-Fang Chen^a and Ya-Ping Hsieh^{c,d†}

^aDepartment of Physics, National Taiwan University, Taipei 106, Taiwan

^bDepartment of Physics, National Central University, Chung Li 320, Taiwan

^cInstitute of Atomic and Molecular Sciences, Academia Sinica, Taipei 115, Taiwan

^dMolecular Science and Technology, Taiwan International Graduate Program, Academia Sinica, Taipei 115, Taiwan

†Correspondence to: yphsieh@gate.sinica.edu.tw (Ya-Ping Hsieh)

Supplementary data S1: Uniformity of the in-situ O₂-processed continuous MoS₂ films

Uniformity of the MoS₂ films was measured using AFM and it showed ~1 – 2 nm thickness. Figure 1 represents the AFM images of different oxygen-assisted MoS₂ samples and its surface height profile, which confirms about the reproducibility as well as large area uniformity. To confirm the large area uniformity of the film thickness, we have measured the thickness at several locations (~10 places) using AFM and all measurements show the same result of ~1 – 2 nm. (Here, we have

shown the AFM image and corresponding surface height profile of a few places.) These results show that films are quite uniform in large area.

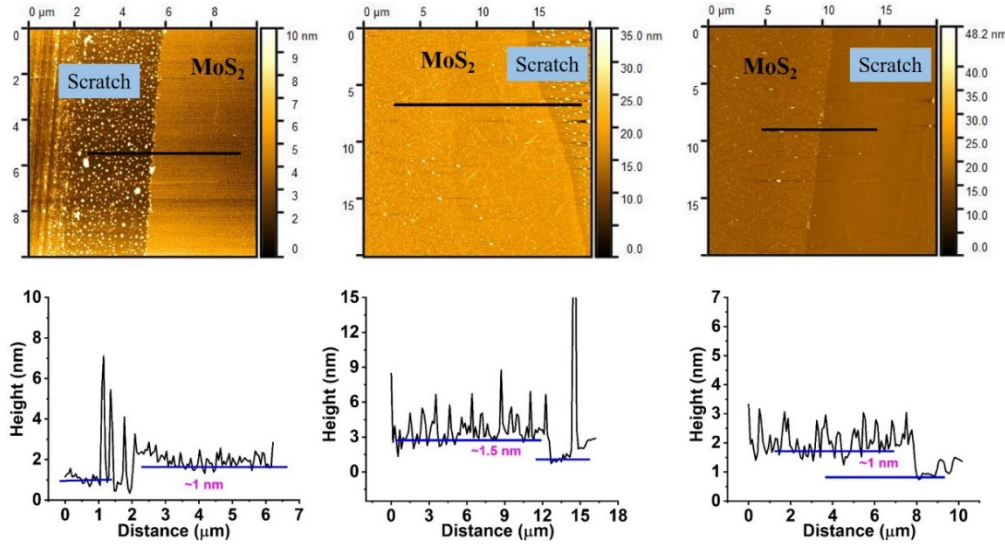


Figure 1: AFM images of different oxygen-assisted MoS₂ films and its surface height profile, which confirms about the reproducibility as well as large area uniformity.

Supplementary data S2: Transistor device fabrication on Si/SiO₂

Field effect transistor devices were fabricated on Si/SiO₂ (90 nm) using photolithography followed by thermal evaporation of Ti (3 nm) / Au (80 nm) for source and drain electrodes on top of MoS₂. Thermally grown 90 nm SiO₂ acts as gate dielectric and Si acts as gate electrode. The channel length of the device was 6 and 25 μm for the films processed without oxygen and in-situ oxygen samples respectively. After completion of lift-off process, devices were annealed in Ar environment at 200 $^{\circ}\text{C}$ for 2h, for removing the organic residue. All the measurements were performed in ambient condition.

Supplementary data S3: Responsivity data comparison for the two terminal co-planar geometry (in ambient condition)

Responsivity (A/W)	Wavelength and laser power	Area of devices (L/W)* (in μm)	Materials	References
1 at $V_{\text{ds}} = 1.5 \text{ V}$	532 nm, 200 μW	A few micron / tens to 100 micron	CVD MoS_2	1
1×10^{-3} at $V_{\text{ds}} = 1 \text{ V}$	405 nm, 100 μW	2 / 20	CVD MoS_2	2
59 at $V_{\text{ds}} = 1.2 \text{ V}$	532 nm, $1.69 \times 10^{-3} \text{ W/cm}^2$	5 / ~ 30	Exfoliated MoS_2	3
**780 at $V_{\text{ds}} = 1 \text{ V}$	532 nm, $1.3 \times 10^{-4} \text{ W/cm}^2$	---	CVD MoS_2	4
1.1×10^6 at $V_{\text{ds}} = 0.15 \text{ V}$	460 nm, 0.33 pW	---	Exfoliated MoS_2	5
1.1×10^{-3} at $V_{\text{ds}} = 1.5 \text{ V}$	514.5 nm, 1 μW	0.8 / 5	CVD MoS_2	6
0.42×10^{-3} at $V_{\text{ds}} = 1 \text{ V}$	550 nm, 80 W/cm^2	2.1 / 2.6	Exfoliated MoS_2	7
7 at $V_{\text{ds}} = 1 \text{ V}$	488 nm, 1 μW	---	CVD MoS_2	8
420 at $V_{\text{ds}} = 15 \text{ V}$	532 nm, 10^{-5} W/cm^2	125 / 480	CVD MoS_2	This work

* L is channel length; W is channel width. ** $V_{\text{g}} - V_{\text{th}} = 100 \text{ V}$

CVD (Chemical Vapor Deposition)

Supplementary data S4: I-T curve of oxygen assisted MoS_2 thin films

For the stability of photosensor, I-T measurement were performed for 10 cycles with a laser power of $9.5 \times 10^{-3} \text{ W/cm}^2$. A uniform sustainable of photocurrent for longer duration indicate that defect

density in oxygen-assisted MoS₂ is decreased, as shown in figure 2. Due to decrease in trap states, a fast recovery time of the order of millisecond is observed. The rise and fall time are 350 ms and 310 ms respectively.

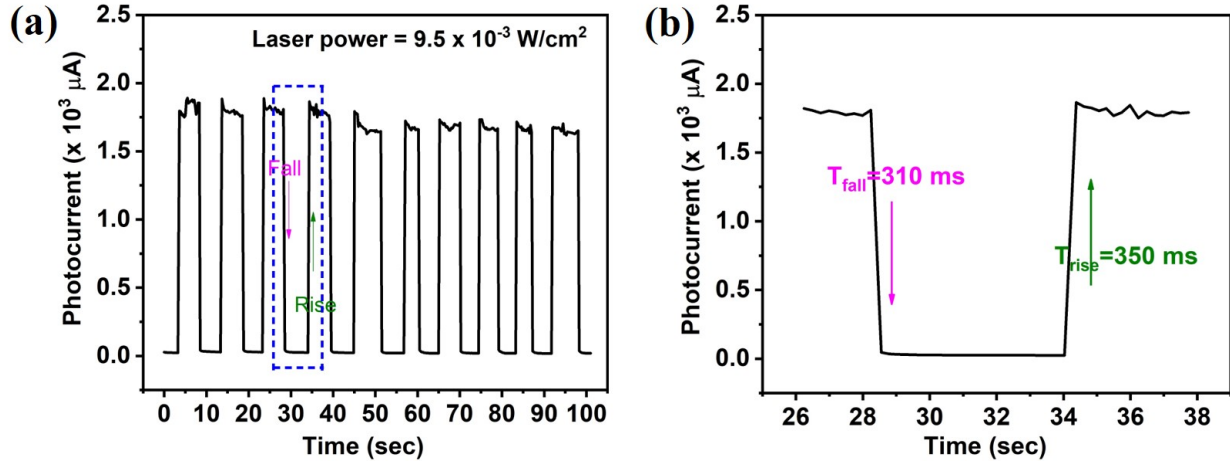


Figure 2: (a) I-T curve of oxygen assisted MoS₂ thin films for 10 cycles at a fixed biasing of V_{ds} 15V and (b) is corresponding fall and rise time 310 ms and 350 ms respectively.

Supplementary data S5: Uniform wafer scale photosensitivity of MoS₂

Wafer scale continuous MoS₂ films were grown on Si/SiO₂ using 3-inch CVD system. Figure 3 represents the large scale growth of MoS₂ in centimetre dimensions and its photoluminescence histogram around the whole wafer. Photoluminescence of the sample were measured randomly in one direction from one end to another end with a step of 2 mm. The histogram of photoluminescence intensity shows almost uniform single layer MoS₂ films over the entire substrate. However, a small decrease in photoluminescence intensity is observed at the other end of the samples. This may be due to the bilayer MoS₂, which could be possible due to the

concentration gradient of MoO₃ vapor in large area i.e. higher towards MoO₃ source and lower far away from the source.

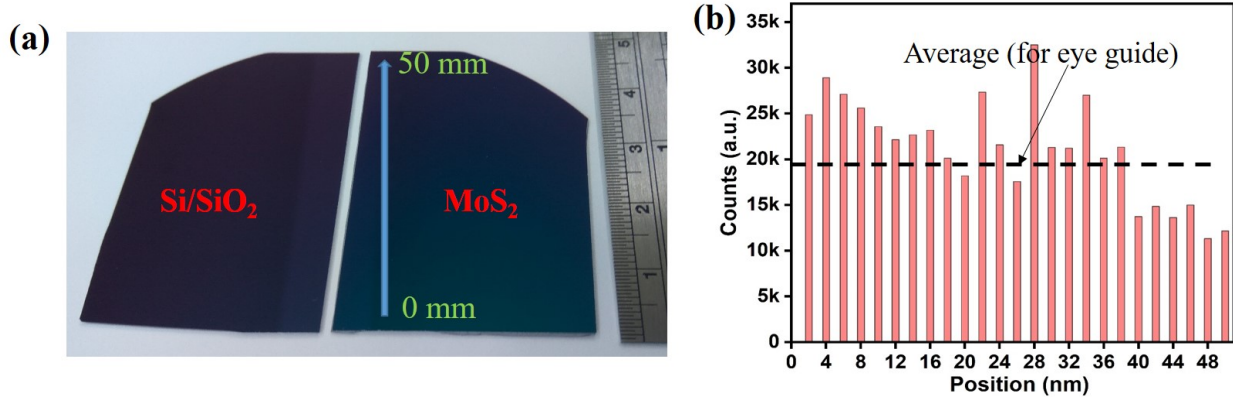


Figure 3: (a) Wafer scale continuous MoS₂ growth on Si/SiO₂ substrate (right side). A blank Si/SiO₂ substrate (left side) is shown here for the reference of contrast image. (b) Histogram of photoluminescence intensity measured on the wafer scale samples from one end to another end in one direction with 2 mm step. (Black dashed line is represented for eye guide only).

References

- 1 S. Khadka, T. E. Wickramasinghe, M. Lindquist, R. Thorat, S. H. Aleithan, M. E. Kordesch and E. Stinaff, *Applied Physics Letters*, 2017, **110**, 261109.
- 2 A. E. Yore, K. K. H. Smithe, S. Jha, K. Ray, E. Pop and A. K. M. Newaz, *Applied Physics Letters*, 2017, **111**, 043110.
- 3 W. Tang, C. Liu, L. Wang, X. Chen, M. Luo, W. Guo, S. W. Wang and W. Lu, *Applied Physics Letters*, 2017, **111**, 153502.
- 4 W. Zhang, J. K. Huang, C. H. Chen, Y. H. Chang, Y. J. Cheng and L. J. Li, *Advanced Materials*, 2013, **25**, 3456–3461.
- 5 R. Nur, T. Tsuchiya, K. Toprasertpong, K. Terabe, S. Takagi and M. Takenaka, *Communications Materials*, 2020, **1**, 103.

- 6 N. Perea-López, Z. Lin, N. R. Pradhan, A. Iñiguez-Rábago, A. L. Elías, A. McCreary, J. Lou, P. M. Ajayan, H. Terrones, L. Balicas and M. Terrones, *2D Materials*, 2014, **1**, 011004.
- 7 Z. Yin, H. Li, H. Li, L. Jiang, Y. Shi, Y. Sun, G. Lu, Q. Zhang, X. Chen and H. Zhang, *ACS Nano*, 2012, **6**, 74–80.
- 8 J. Jadwiszczak, G. Li, C. P. Cullen, J. J. Wang, P. Maguire, G. S. Duesberg, J. G. Lunney and H. Zhang, *Applied Physics Letters*, 2019, **114**, 091103.