crystals

MDPI

Supplementary Materials

Batch Production of Wafer-Scale Monolayer MoS_2

Zheng Wei [1], Xingdong Sun [1], Yongqing Cai [2,*], Yao Liang [1] and Zhihua Zhang [1]

[1] School of Materials Science and Engineering, Dalian Jiaotong University, Dalian 116028, China
[2] Shenzhen Institute for Quantum Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China
* Correspondence: caiyq@sustech.edu.cn

(1) Experimental setup

Batch production of monolayer MoS_2 wafers were grown by CVD and Figure S1 presents the photograph for the experimental setup. The CVD system consists of a three-temperature-zone tube furnace, a vacuum chamber and the gas circuit. Sulfur powder and molybdenum trioxide powder were employed as precursors and were loaded in a quartz boat and an inner quartz tube at low temperature zone I and zone II, respectively. 1-inch wafer-scale sapphire substrates were stuck in the grooves of the tailor-made sample holder erectly and were placed in high temperature zone III. Monolayer MoS_2 wafers were produced in batch in this CVD system under proper temperature and flow rates. Actually, batch production of oxygen doped MoS_2 wafers were also synthesized in this system and the growth parameters were adjusted slightly compared with those for intrinsic MoS_2 wafers.

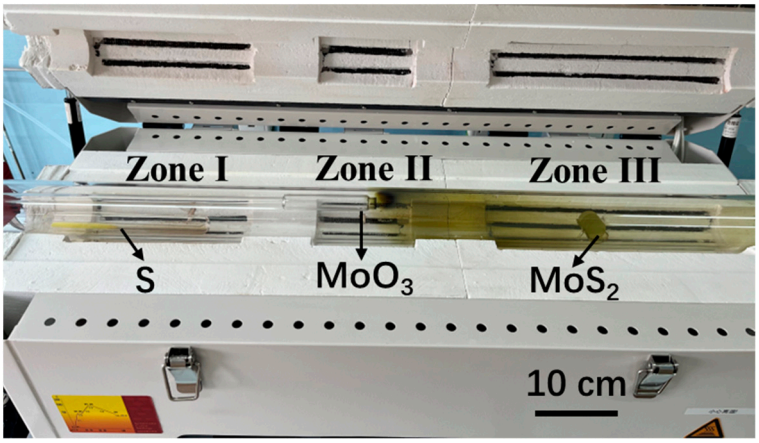

Figure S1. Experimental setup for batch production.

(2) Optical microscopy images

Optical microscopy images of the four monolayer MoS_2 wafers from one batch are shown in Figure S2 and a-d are images for wafer numbered 1-4, respectively. All of the optical microscopy images exhibit clean surfaces and similar contrasts, indicating the great cleanliness and homogeneity for the wafers in the same batch. Note that the scratches on the surface of samples are made by tweezers artificially for the comparison of MoS_2 monolayers and sapphire substrates.

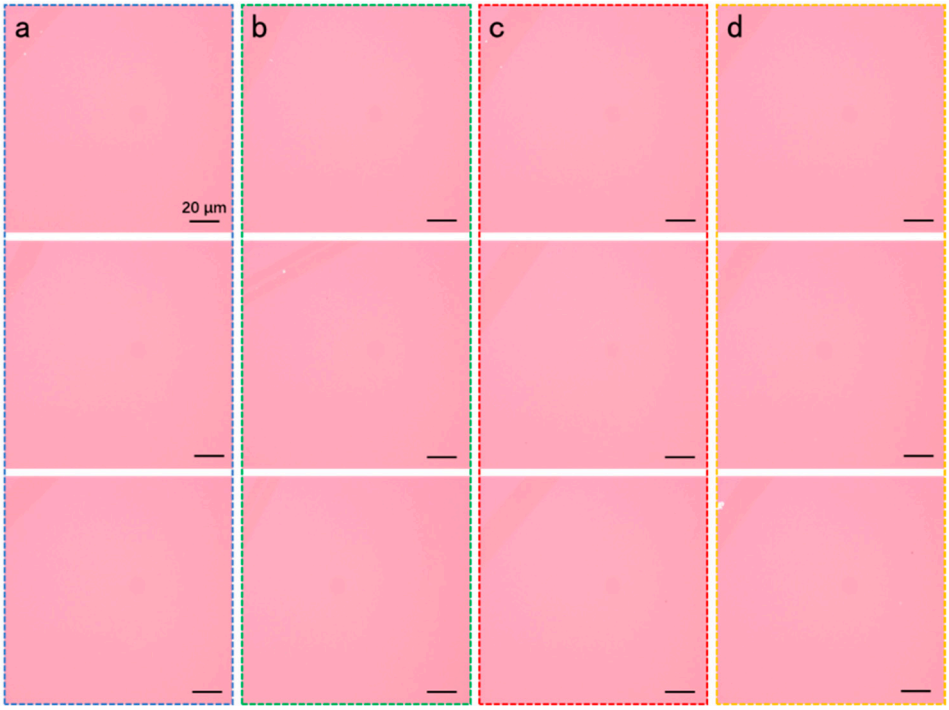

Figure S2. Optical microscopy images of MoS_2 monolayers in one batch. (**a**–**d**) Optical microscopy images for wafer 1 - wafer 4. Scale bars: 20 μm.

(3) AFM of sapphire

As depicted in Figure S3, the AFM image demonstrates that the sapphire substrate exhibits smooth morphology with steps on the surface. Through analysis of the AFM image, the roughness of the sapphire substrates is calculated to be 0.2 nm. These substrate steps play a pivotal role in facilitating the nucleation process during the monolayer MoS_2 growth.

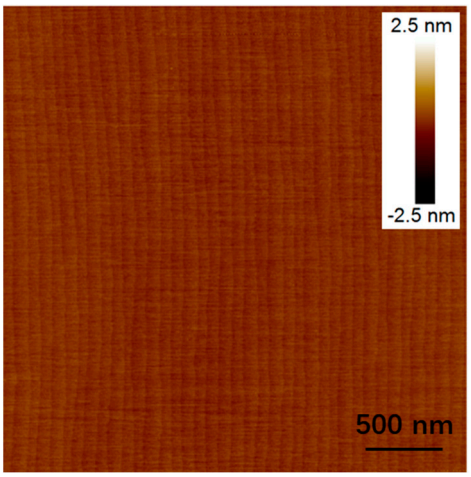

Figure S3. AFM image of sapphire substrate. Inset shows the color-scale of height.

(4) Thickness of MoS_2

The layer number of MoS_2 samples is characterized by AFM. Figure S4 demonstrates the AFM image of MoS_2 ribbon on sapphire substrate. The measured height at the ribbon's edge is approximately 0.6 nm, consistent with the thickness of a monolayer MoS_2 film.

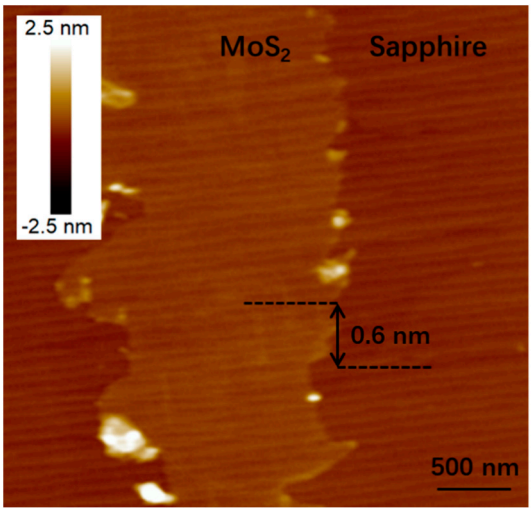

Figure S4. AFM image of the MoS_2 ribbon on sapphire substrate. Inset is the color-scale bar.

(5) Grain size of monolayer MoS_2

We have estimated the grain size of MoS_2 monolayer by AFM. In order to figure out the location and shape of grain boundaries, we fumigated the sample with steam. Figure S5 shows the AFM image of the MoS_2 sample with clear grain boundaries and the grain size of MoS_2 is measured around 1 μm.

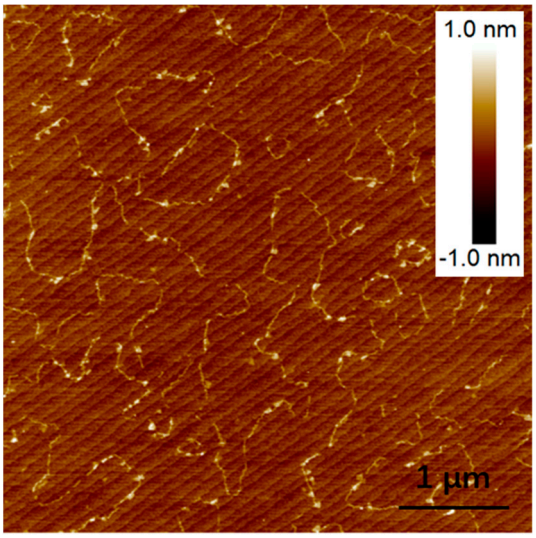

Figure S5. Estimate of the crystal grain size of MoS_2 by AFM image. Inset shows the color-scale of height.

(6) Raman spectra

To investigate the homogeneity of the batch-produced MoS_2 monolayers across the entire wafer scale, we conducted optical spectroscopy. The normalized raw data of the Raman spectra for MoS_2 monolayers along the X-direction and Y-direction are presented in Figure S6a and S6b, respectively. The spectra show typical E_{2g} and A_{1g} Raman peaks and all of the Raman peaks exhibit coincident peak position and uniform intensity in both directions, indicating the excellent homogeneity of the entire monolayer MoS_2 wafers along both orientations.

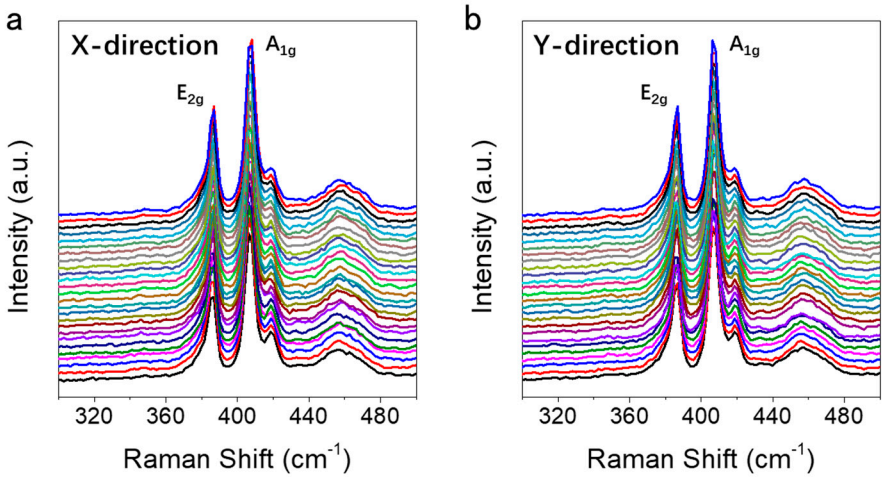

Figure S6. Raman spectra of MoS_2. (**a**,**b**) Raman spectra of wafer-scale monolayer MoS_2 along X- and Y-direction, respectively.

(7) PL spectra

Similar to the Raman spectra, we also performed PL spectra of the batch-produced MoS_2 monolayers across the entire wafer scale. Figure S7a and S7b are the normalized raw data of the PL spectra for MoS_2 monolayers along the X-direction and Y-direction. The strong PL peaks are clearly observed and show constant wavelength and intensity for the spectra in both directions, which indicate the great uniformity of monolayer MoS_2 wafers grown by batch production as well.

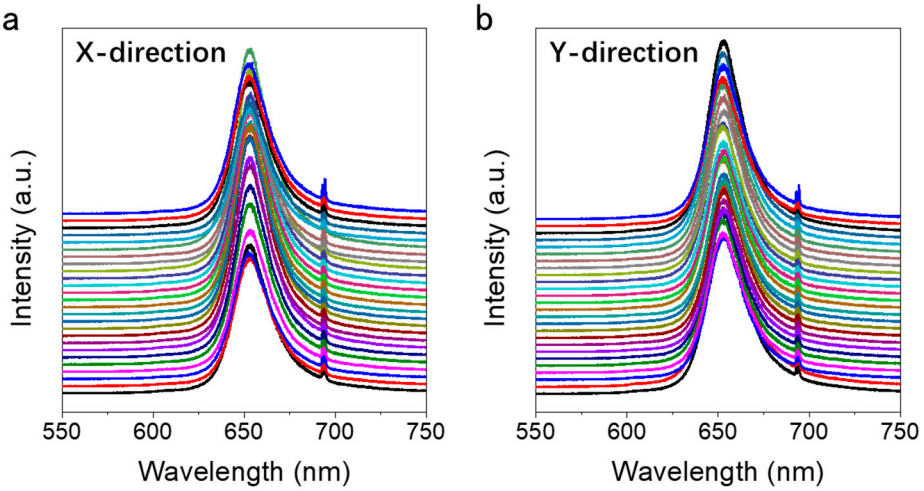

Figure S7. PL spectra of MoS_2. (**a**,**b**) PL spectra of wafer-scale monolayer MoS_2 along X- and Y-direction, respectively.

(8) Transfer characteristic curves

Back-gate FETs were fabricated on SiO_2/Si substrates to characterize the electrical performance of MoS_2 monolayers grown through batch production. Figure S8 presents the transfer characteristic curves of 20 transistors and the curves exhibit typical n-type semiconductor behavior. Field-effect mobility and current on/off ratio of these MoS_2 transistors are calculated and the average value of mobility and on/off ratio are 10.5 $cm^2V^{-1}s^{-1}$ and 1.3×10^6, respectively. The results of electrical performances confirm the high quality of monolayer MoS_2 produced by batch production process and the distributions of transfer curves, field-effect mobility and current on/off ratio indicate the outstanding homogeneity of MoS_2 samples.

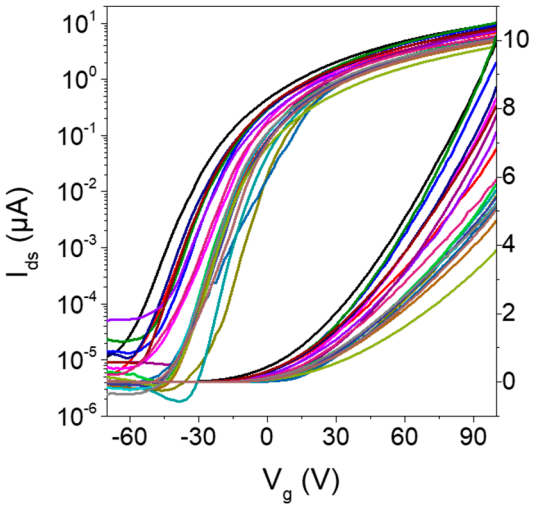

Figure S8. Transfer characteristic curves of MoS_2 FETs.

(9) Comparison of FET performance

In order to compare the performance of MoS_2 FET with previous reported results, especially the MoS_2 samples grown by chemical vapor deposition, we have meticulously compiled a comprehensive summary in Table S1. In this work, the average mobility of the MoS_2 FETs is calculated to be 10.5 $cm^2V^{-1}s^{-1}$ and the on/off ratio averages at 1.3×10^6, which is at a high level among the previous results. The results of electrical performance of FETs confirms the high quality of the monolayer MoS_2 films synthesized by batch production.

Table S1. Comparison of performance for MoS_2 FET.

Ref.	Field-Effect Mobility ($cm^2V^{-1}s^{-1}$)	On/Off Ratio
Liu, L et al. [1]	3.2	1×10^6
Liu, K et al. [2]	6	1×10^6
Zhang, J et al. [3]	7	5×10^6
Yang, P et al. [4]	11.2	7.7×10^5
This work	10.5	1.3×10^6

(10) Comparison of MoS_2 and $MoS_{2-x}O_x$

For purpose of comparing the contrast of intrinsic MoS_2 and oxygen doped MoS_2, photograph of original sapphire, MoS_2 and $MoS_{2-x}O_x$ wafers is shown in Figure S9. The MoS_2 wafer show the color of olive green, while the color of $MoS_{2-x}O_x$ wafer is yellowish brown. Although the samples present different contrasts, they show clean and homogeneous surfaces across the whole wafer, indicating the high quality of the batch-production samples.

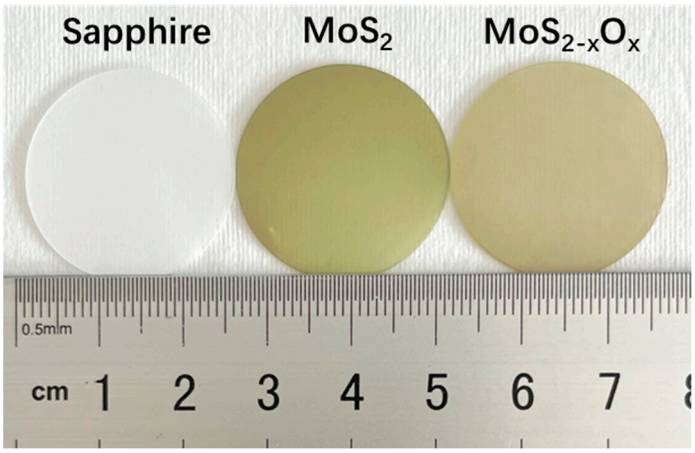

Figure S9. Comparison of monolayer MoS_2 wafer and monolayer $MoS_{2-x}O_x$ wafer.

(11) Grain size of monolayer oxygen-doped MoS_2

The grain size of oxygen-doped MoS_2 was estimated by optical microscopy image through measuring single-crystal samples. Figure S10 shows the optical microscopy image of oxygen doped MoS_2 grain and it exhibits the grain size of around 35 μm.

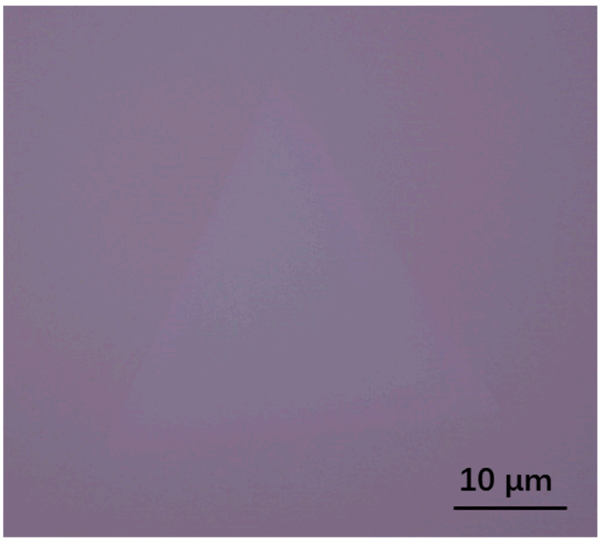

Figure S10. Estimate of the crystal grain size of MoS_2 by optical microscopy image.

References

1. Liu, L.; Qiu, H.; Wang, J.; Xu, G.; Jiao, L. Atomic MoS2 monolayers synthesized from a metal-organic complex by chemical vapor deposition. *Nanoscale* **2016**, *8*, 4486-4490.
2. Liu, K. K.; Zhang, W.; Lee, Y. H.; Lin, Y. C.; Chang, M. T.; Su, C. Y.; Chang, C. S.; Li, H.; Shi, Y.; Zhang, H. *et al.* Growth of large-area and highly crystalline MoS2 thin layers on insulating substrates. *Nano Lett* **2012**, *12*, 1538-1544.
3. Zhang, J.; Yu, H.; Chen, W.; Tian, X. Z.; Liu, D. H.; Cheng, M.; Xie, G. B.; Yang, W.; Yang, R.; Bai, X. D. *et al.* Scalable growth of high-quality polycrystalline MoS2 monolayers on SiO_2 with tunable grain sizes. *ACS Nano* **2014**, *8*, 6024-6030.
4. Yang, P.; Zhang, S.; Pan, S.; Tang, B.; Liang, Y.; Zhao, X.; Zhang, Z.; Shi, J.; Huan, Y.; Shi, Y. *et al.* Epitaxial growth of centimeter-scale single-crystal MoS2 monolayer on Au(111). *ACS Nano* **2020**, *14*, 5036-5045.