

## Supplementary Information

### **Remarkable quality improvement of as-grown monolayer MoS<sub>2</sub> by sulfur vapor pretreatment of SiO<sub>2</sub>/Si substrate**

Peng Yang<sup>1</sup>, Yabing Shan<sup>1</sup>, Jing Chen<sup>1</sup>, Garel Ekoya<sup>1</sup>, Jinkun Han<sup>1</sup>, Zhi-Jun Qiu<sup>1</sup>, Junjie Sun<sup>2</sup>, Fei Chen<sup>2</sup>, Haomin Wang<sup>3</sup>, Wenzhong Bao<sup>4</sup>, Laigui Hu<sup>1</sup>, Rong-Jun Zhang<sup>1</sup>, Ran Liu<sup>1</sup>, and Chunxiao Cong<sup>1, 5,\*</sup>

<sup>1</sup> State Key Laboratory of ASIC and System, School of Information Science and Technology, Fudan University, Shanghai 200433, China

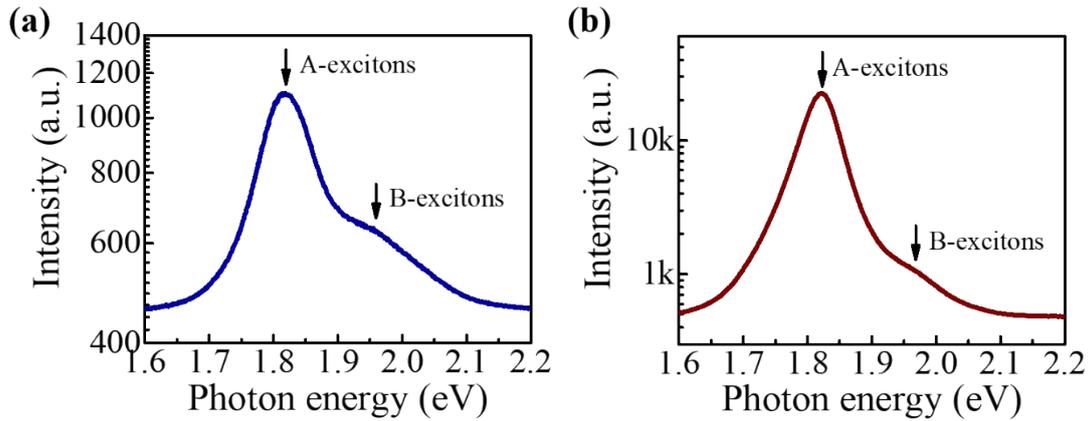
<sup>2</sup> State Key Laboratory of Laser Interaction with Matter, Innovation Laboratory of Electro-Optical Countermeasures Technology, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, 3888 Dongnanhu Road, Changchun Jilin 130033, China

<sup>3</sup> State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Changning Road 865, Shanghai 200050, China

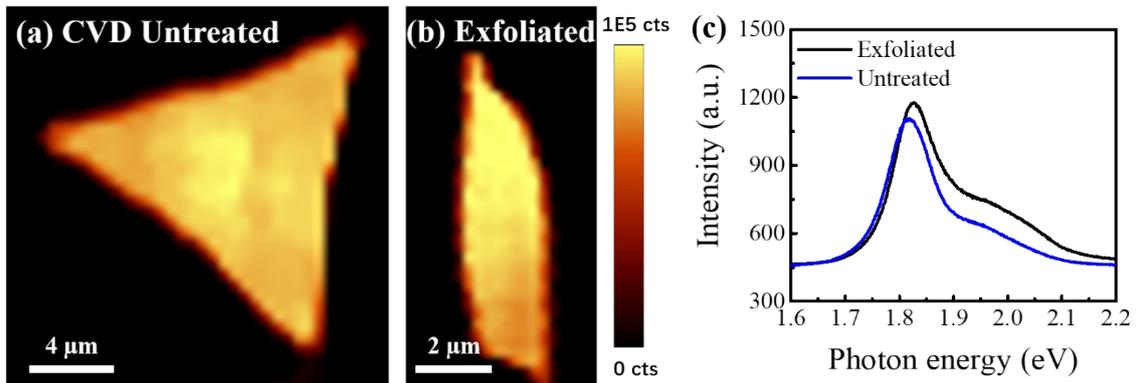
<sup>4</sup> State Key Laboratory of ASIC and System, School of Microelectronics, Fudan University, Shanghai 200433, China

<sup>5</sup> Academy for Engineering and Technology, Fudan University, Shanghai 200433, China

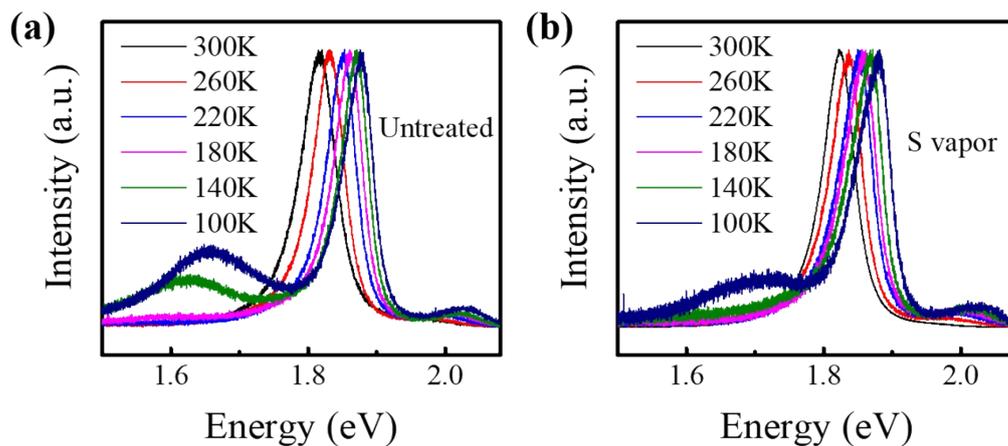
\*Address correspondence to [cxcong@fudan.edu.cn](mailto:cxcong@fudan.edu.cn)



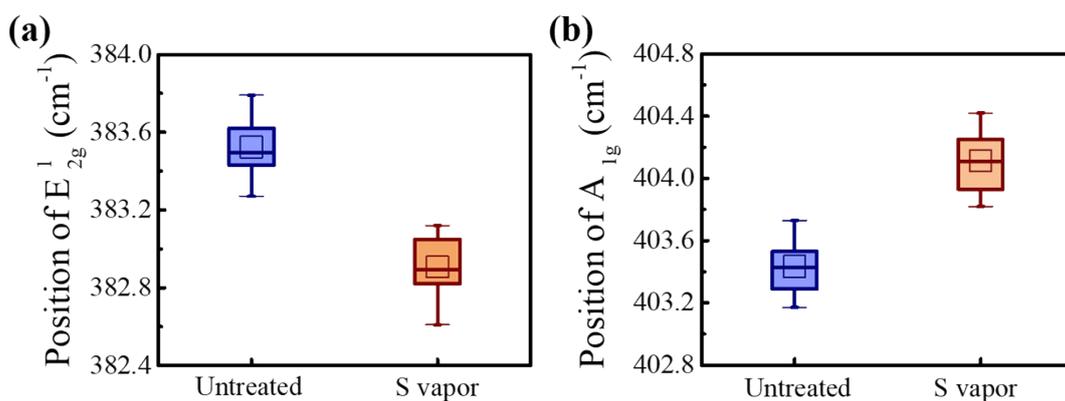
**Figure S1** PL spectra of MoS<sub>2</sub> grown on untreated SiO<sub>2</sub>/Si substrate (a) and sulfur vapor pretreated SiO<sub>2</sub>/Si substrate (b) with intensity on a log-scale.



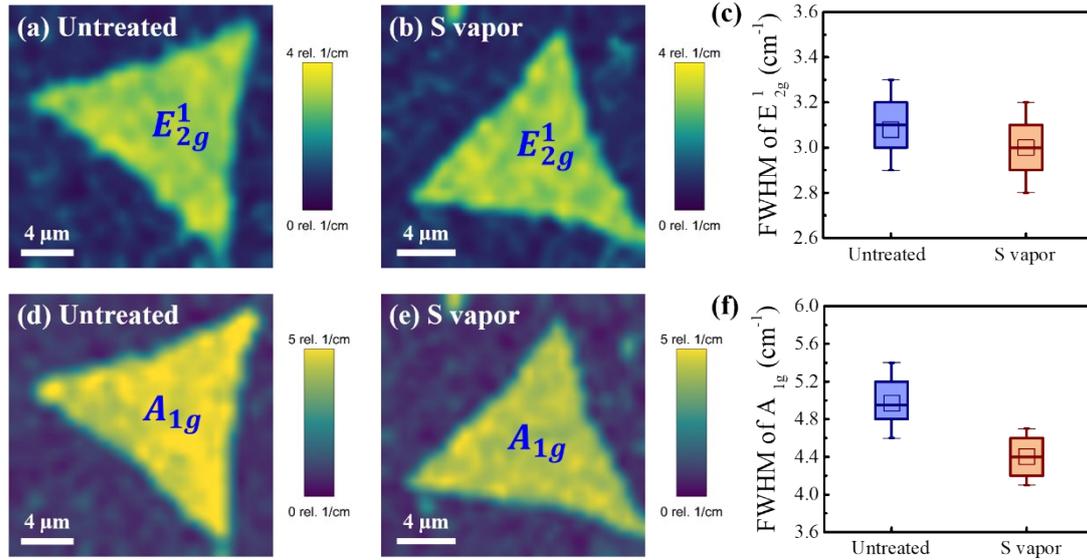
**Figure S2.** (a) PL peak intensity mapping of MoS<sub>2</sub> domain corresponding to Figure 1(b), it should be noted that the intensity scale bar in Figure 1(b) is twenty fold than that of Figure S1(a). (b) PL peak intensity mapping of mechanical exfoliated monolayer MoS<sub>2</sub>. (c) Typical PL spectra extracted from (a) blue line, and (b) black line, respectively.



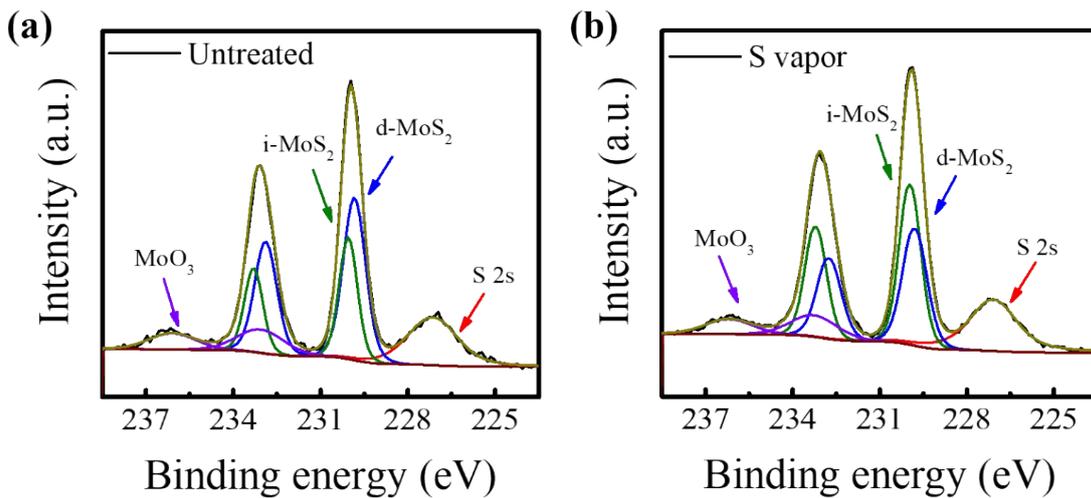
**Figure S3.** Temperature-dependent PL spectra of MoS<sub>2</sub> flake grown on untreated (a) and sulfur vapor pretreated (b) SiO<sub>2</sub>/Si substrate. All the spectra are normalized by the intensity of the neutral exciton.



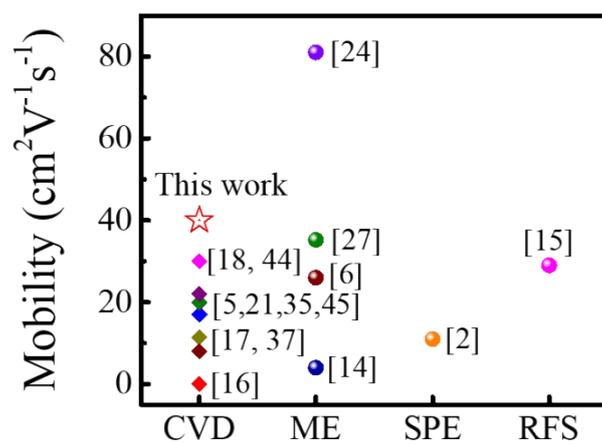
**Figure S4** Statistic peak position of E<sub>1</sub> 2<sub>g</sub> (a) mode and A<sub>1g</sub> (b) mode of MoS<sub>2</sub> grown on untreated and sulfur vapor pretreated SiO<sub>2</sub>/Si substrate



**Figure S5.** (a, b) Raman mapping of FWHM of  $E_{12g}$  mode of MoS<sub>2</sub> domains corresponding to Figure 2(b) and 2(d), respectively. (c) The statistical results of peak width of  $E_{12g}$  mode extracted from the mapping area of (a, b). (d, e) Raman mapping of FWHM of  $A_{1g}$  mode of MoS<sub>2</sub> domains corresponding to Figure 2(c) and 2(e), respectively. (f) The statistical results of peak width of  $A_{1g}$  mode extracted from the mapping area of (d, e).



**Figure S6.** XPS spectra of Mo 3d and S 2s core levels of CVD grown MoS<sub>2</sub> on untreated SiO<sub>2</sub>/Si substrate (a) and sulfur vapor pretreated SiO<sub>2</sub>/Si substrate (b).



**Figure S7.** Statistics of mobility of MoS<sub>2</sub> in the corresponding references. Note: CVD (chemical vapor deposition), ME (mechanical exfoliation), SPE (solution-processable exfoliation), RFS (radio frequency sputtering).