

## Supporting Information

### **Photoluminescence of Monolayer MoS<sub>2</sub> Modulated by Water/O<sub>2</sub>/Laser Irradiation**

*Chao Hou,<sup>a</sup> Jingwen Deng,<sup>a</sup> Jianxin Guan,<sup>a</sup> Qirong Yang,<sup>a</sup> Zhihao Yu,<sup>a</sup> Yilin Lu,<sup>b</sup>*

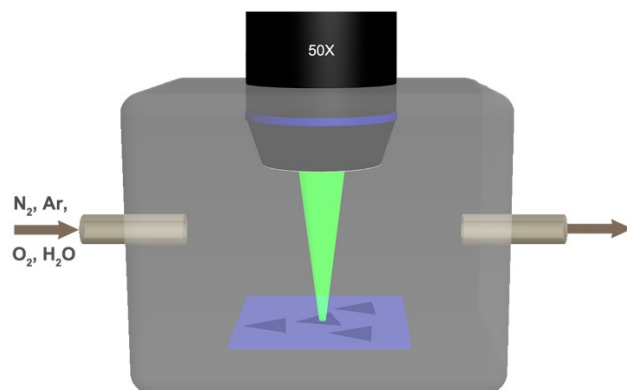
*Zihan Xu,<sup>c</sup> Zefan Yao <sup>a</sup> and Junrong Zheng <sup>\*a</sup>*

<sup>a</sup> College of Chemistry and Molecular Engineering, Beijing National Laboratory for Molecular Sciences, Peking University, Beijing 100871, China

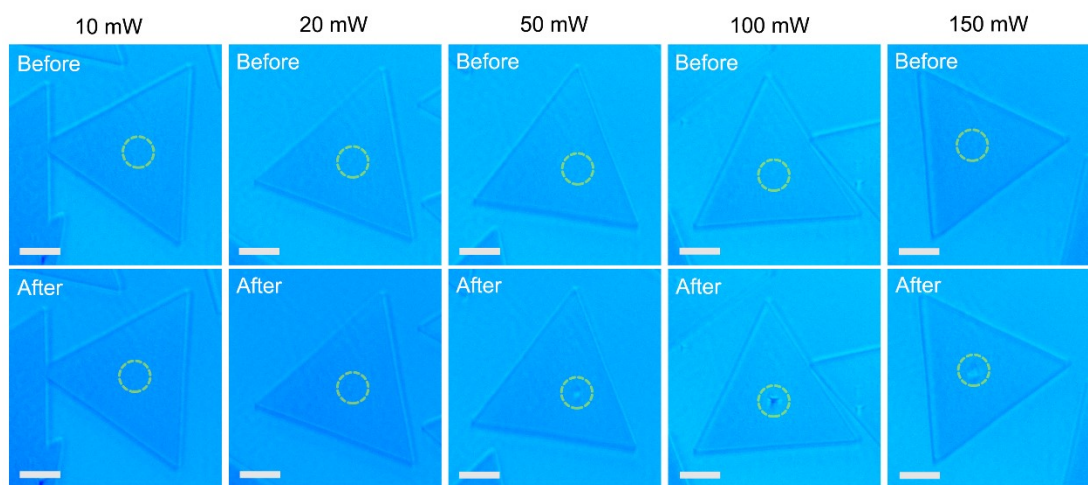
<sup>b</sup> Institutes of Physical Science and Information Technology, Anhui University, Hefei 230601, China

<sup>c</sup> Shenzhen Sixcarbon Technology, Shenzhen 518106, China

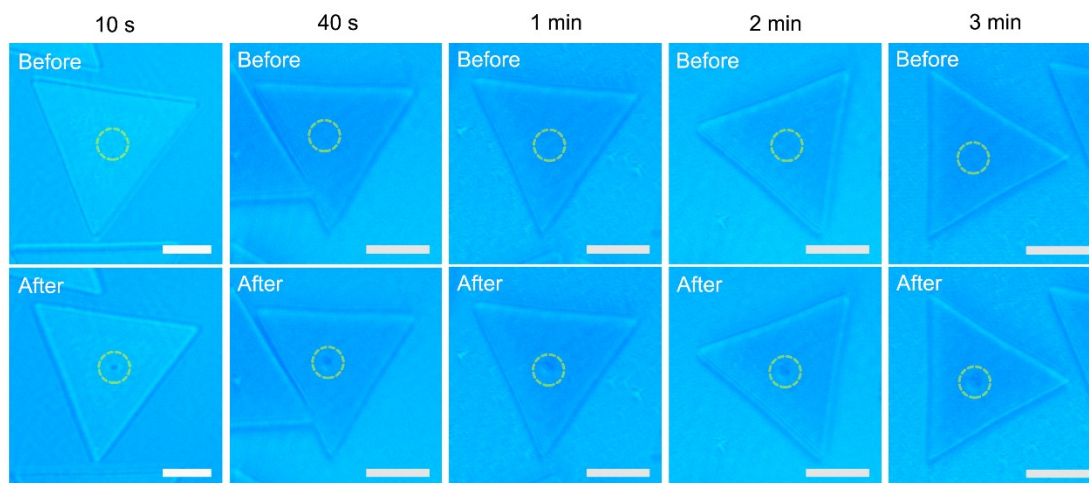
\*E-mail: junrong@pku.edu.cn, zhengjunrong@gmail.com.



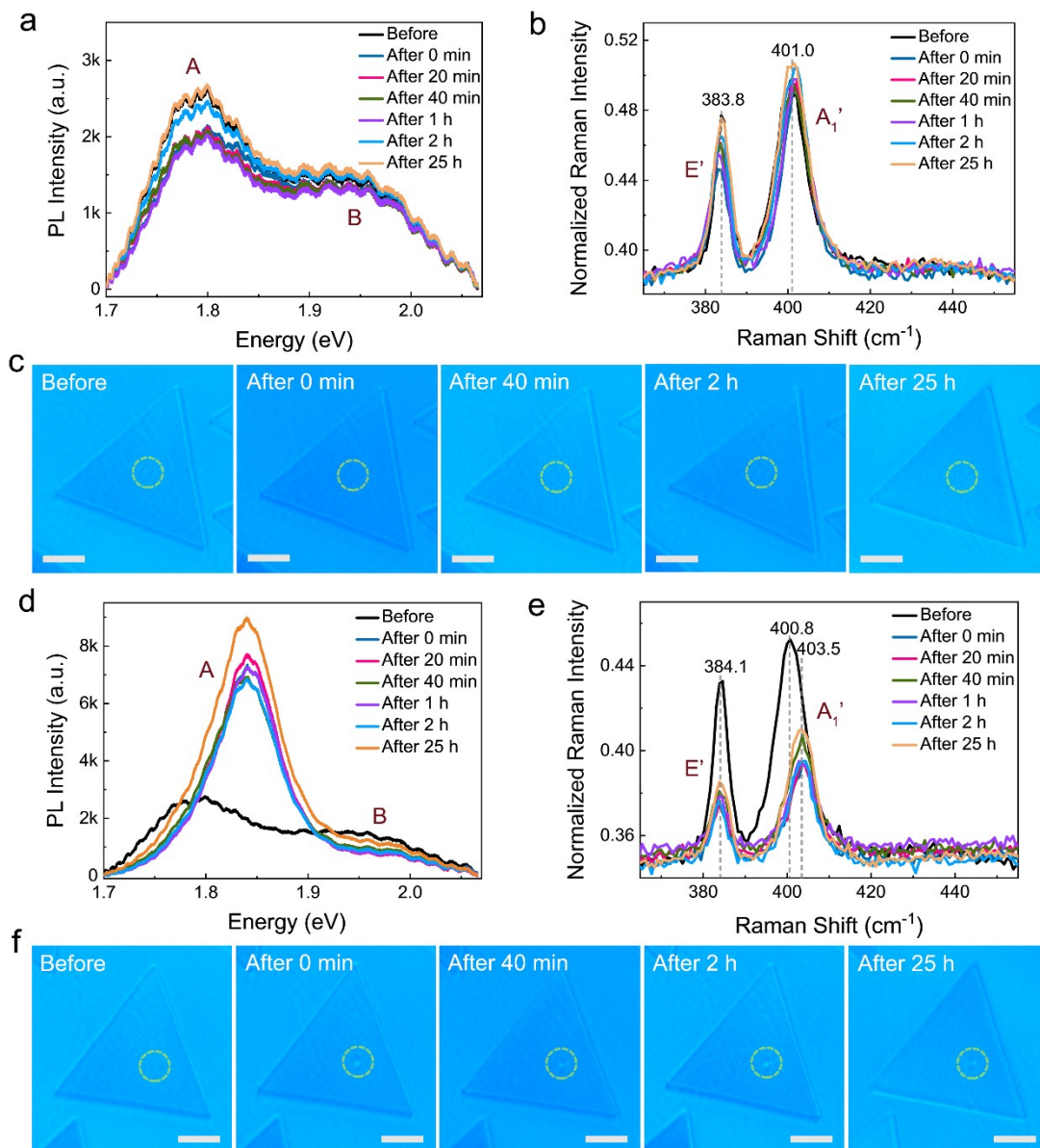
**Fig. S1** A schematic of atmosphere controllable cell, illustrating the *in-situ* PL/Raman measurement under different gas species.



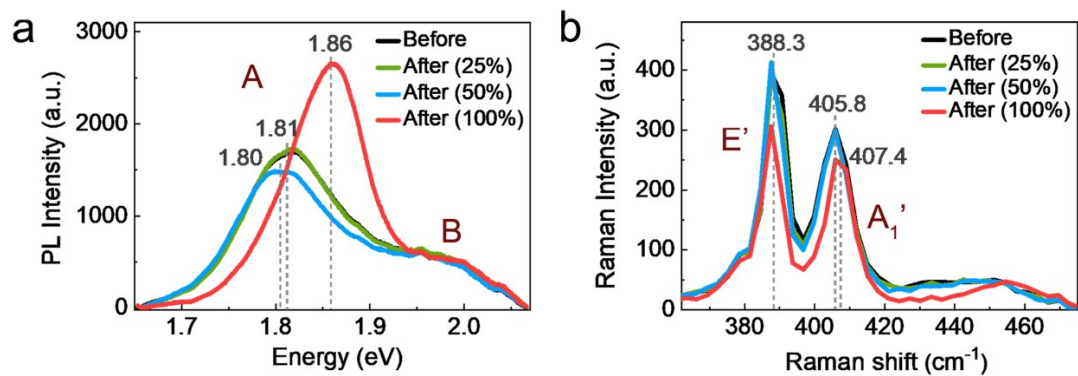
**Fig. S2** Optical images of the MoS<sub>2</sub> single domain before and after being irradiated by 532 nm laser in the air for two minutes with different irradiation laser powers. The yellow circles denote the spots been irradiated by laser. Sale bars are all 10  $\mu$ m.



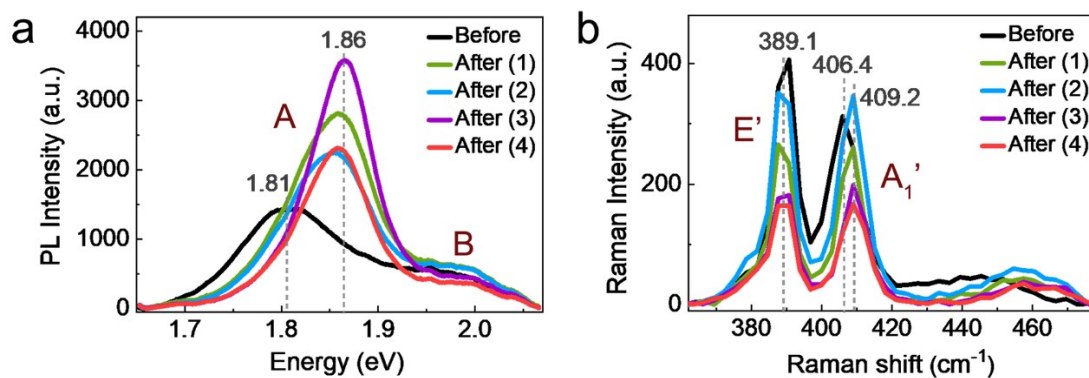
**Fig. S3** Optical images of the MoS<sub>2</sub> single domain before and after 50 mW of 532 nm laser irradiation in the air for different times. The yellow circles denote the spots been irradiated by laser. Sale bars are all 10  $\mu\text{m}$ .



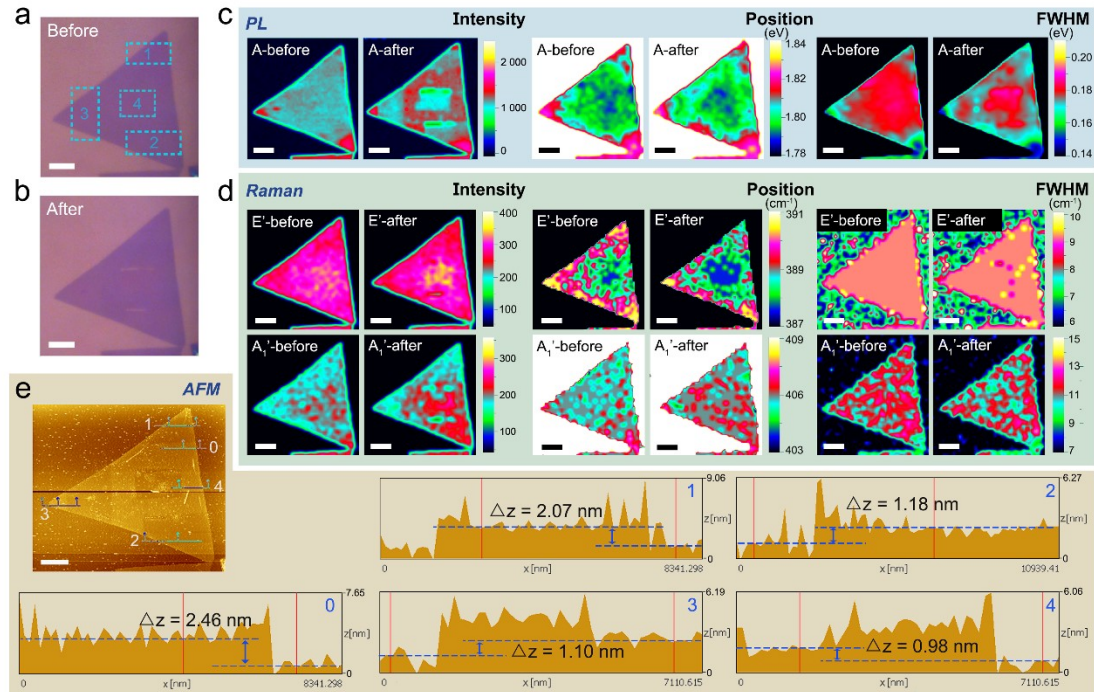
**Fig. S4** (a) PL and (b) Raman spectra, and (c) the corresponding optical images of MoS<sub>2</sub> monolayer single domain before laser treatment and from 0 minute to 25 hours after laser irradiation (532 nm laser, 20 mW, 2 minutes) in the air. (d-f) The results for the same measurements while only changing the laser irradiation power to 50 mW. Spectra are taken with excitation power of 5 mW for 10 s. The Raman spectra are normalized to peak 520 cm<sup>-1</sup> of Si/SiO<sub>2</sub> substrate. The yellow circles denote the spots been irradiated by laser. Sale bars are all 10 μm.



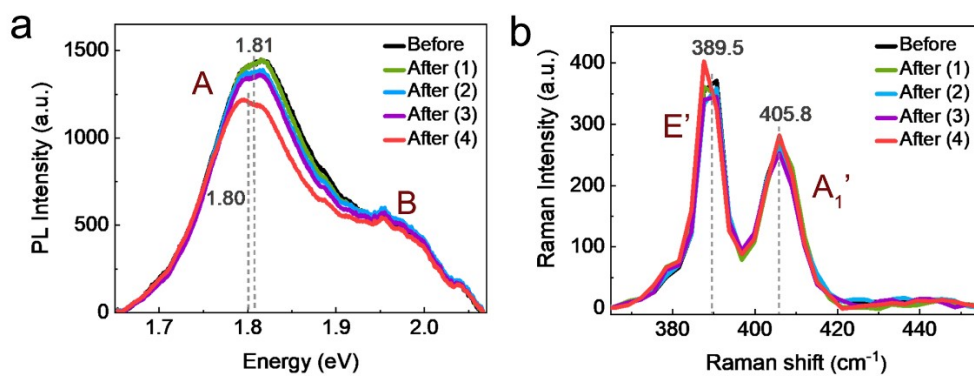
**Fig. S5** (a) PL and (b) Raman average single spectrum from the area before and after laser treatment with different powers corresponding to Fig. 2 in the main text.



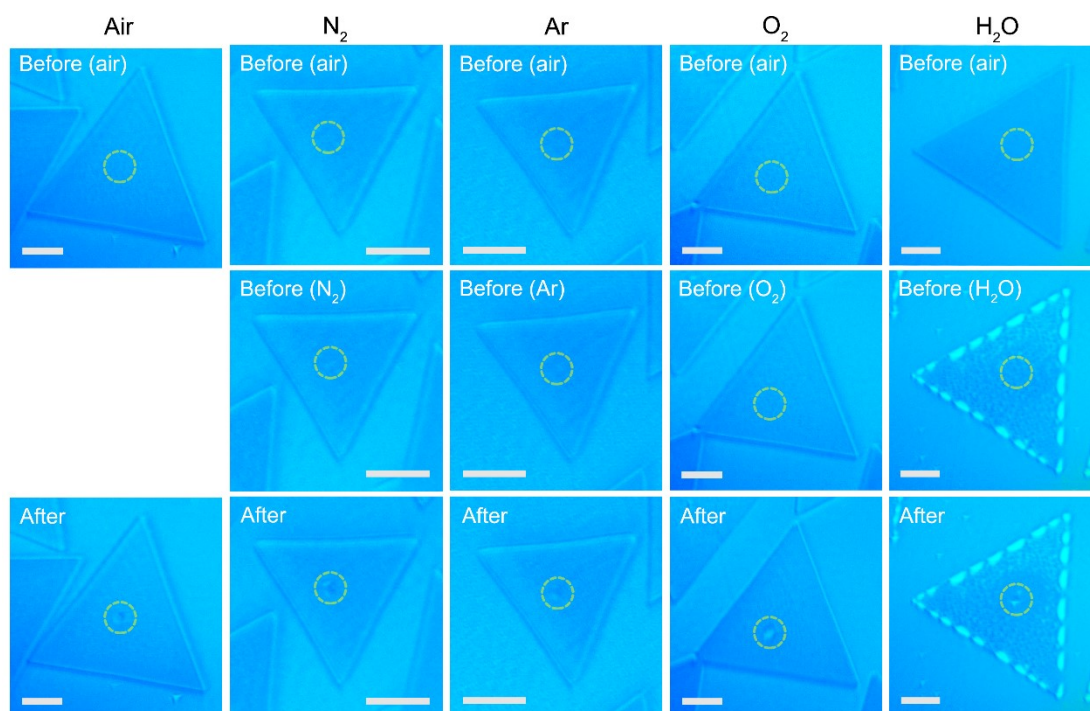
**Fig. S6** (a) PL and (b) Raman average single spectrum from the area before and after 100% power of laser treatment for different times corresponding to Fig. 3 in the main text.



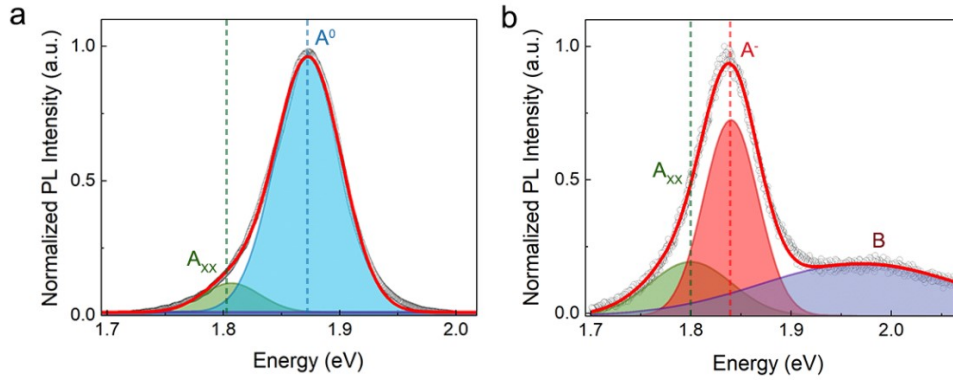
**Fig. S7** Optical images of the monolayer MoS<sub>2</sub> triangle domain (a) before and (b) after 532 nm laser irradiation with 50% power for once, twice, three times and four times, respectively. (c) PL and (d) Raman mappings of intensity, peak position, and FWHM for A emission peak, and E'/A<sub>1</sub>' mode, respectively. (e) The corresponding AFM results after laser treatment. Numbers 0 denotes the area before treatment. Number 1~4 represent areas after laser treatment with 50% power for once, twice, three times and four times, respectively.  $\Delta z$  reveals the thickness of the corresponding area. Sale bars are all 5  $\mu\text{m}$ .



**Fig. S8** (a) PL and (b) Raman average single spectrum from the area before and after 50% power of laser treatment for different times corresponding to Fig. S7.



**Fig. S9** Optical images of the MoS<sub>2</sub> single domain before laser irradiation in ambient air, before and after 532 nm laser irradiation (50 mW for 2 minutes) in different atmospheres (from left to right): ambient air (with humidity 10~20%), N<sub>2</sub>, Ar, O<sub>2</sub>, H<sub>2</sub>O (with humidity 90~98% in ambient air). The yellow circles reveal the spots been irradiated by laser. Sale bars are all 10  $\mu\text{m}$ .



**Fig. S10** (a) PL spectrum of monolayer MoS<sub>2</sub> after adsorbing excessive water (there are water droplets on the surface). It is fitted with Gaussian peaks ( $A_{xx}$ ,  $A^0$ ), revealing neutral exciton ( $A^0$ ) dominates the spectrum. (b) PL spectrum of pristine monolayer MoS<sub>2</sub> excited by low power density of laser excitation (80 W/cm<sup>2</sup>). The fitting result shows the trion ( $A^-$ ) peak dominates the spectrum when the excitation power is low enough. The low power density is realized using a 10× objective lens. Because of different samples, the peaks' positions in (a, b) are slightly different with that in Fig. 6 in the main text.