

## Supporting information

# Flash Phase Engineering of MoS<sub>2</sub> Nanofilms for Enhanced Photoelectrochemical Performance

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## Experimental section

**Preparation of 1T-MoS<sub>2</sub>.** The 1T-MoS<sub>2</sub> was obtained by exfoliation of bulk MoS<sub>2</sub> *via* Li intercalation according to the literature [1]. In brief, 300 mg of MoS<sub>2</sub> powder was immersed in 5 mL hexane containing 1.6 M *n*-butyllithium for 48 h. Then, excess *n*-butyllithium was removed by washing with anhydrous ethanol for three times. The resulting solids were subsequently immersed in 45 mL water. To accelerate the delamination, the solution was shaken manually for 15 min and sonicated for an additional 15 min. The resulting solution was centrifugated at 6000 rpm for 30 min to remove the unexfoliated MoS<sub>2</sub> and multi-layers of MoS<sub>2</sub> (repeated 3 times). Another centrifugation step at 11000 rpm for 30 min was performed to remove nanoparticles resulting from side reactions and small-sized MoS<sub>2</sub>. Finally, the resulting solution was degassed with Ar for 30 min and stored in the refrigerator at 4 °C. The concentration was quantified as 0.5 mg mL<sup>-1</sup> after vacuum drying.

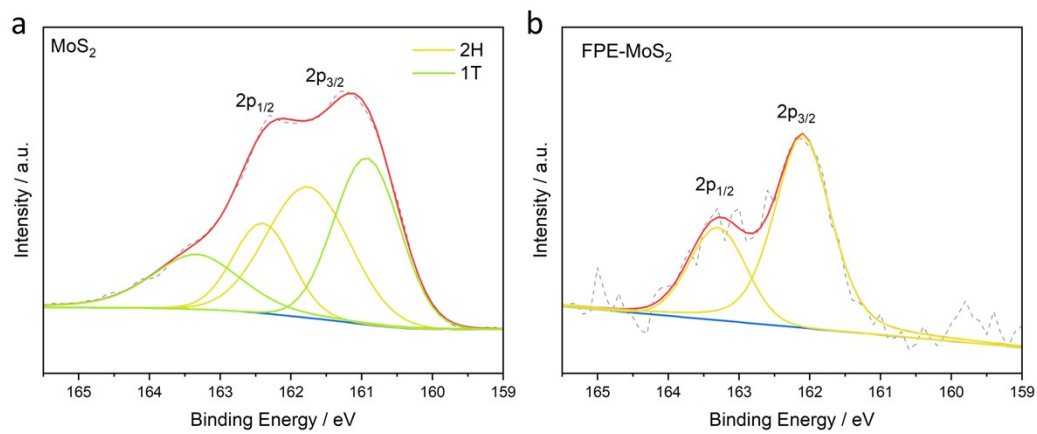
**Fabrication of 1T-MoS<sub>2</sub> substrate nanofilm.** 1T-MoS<sub>2</sub> substrate nanofilm was obtained *via* wet-chemistry exfoliation and vacuum filtration with a 25 nm pore sized membrane (Merck, MCE-Millipore). By adjusting the volume during the fabrication process, nanofilms of different thicknesses could be achieved.

**Laser direct writing setup.** The laser direct writing setup is a 200 mW TOPTICA iBeam smart 488-S laser with a wavelength of 488 nm (TOPTICA Photonics AG, Germany) [2]. The laser setup can be flexibly tuned in scanning speed and laser intensity, with a 1/e<sup>2</sup> focus spot of 15 μm.

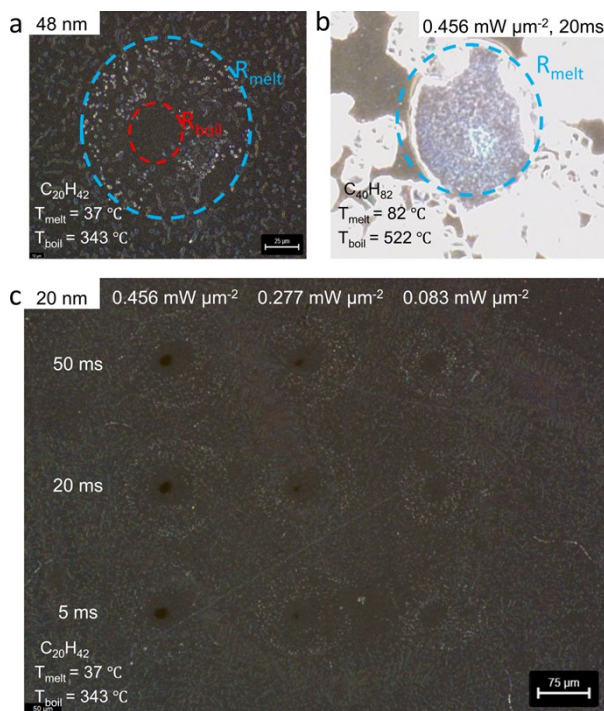
**Fabrication of 1T-2H MoS<sub>2</sub> heterophase.** 1T-2H MoS<sub>2</sub> heterophase was obtained by FPE. The laser set up was employed for precise control 1T and 2H phases by flexibly tuning its power density, scan speed and other parameters.

**Photoelectrochemical (PEC) measurements and GSH detection.** The chronoamperometry curves (*i*-*t*) were measured with an electrochemical potentiostat (Gamry, USA) with a three-electrode system in 0.5 M Na<sub>2</sub>SO<sub>4</sub> solution with Pt as counter electrode and Ag/AgCl as reference electrode, under a fiber-coupled white LED lamp (455 nm, 300 mW cm<sup>-2</sup>). The working electrode was achieved by FPE on 1T-MoS<sub>2</sub> substrate nanofilm under different laser parameters. The response of MoS<sub>2</sub> and FPE-MoS<sub>2</sub> electrodes to GSH was evaluated by successively adding 100 nM GSH solution every 50 s. Measurements were performed at 0.5 V vs. Ag/AgCl electrode under white light irradiation.

**Instrument.** Ultraviolet-Visible (UV-Vis) absorption spectra were measured on a SHIMADZU UV- 3600i Plus UV-vis-NIR spectrophotometer. Atomic Force Microscopy (AFM) measurement was conducted on a Park NX10 AFM with tapping mode. Raman spectroscopy was conducted on a WITec alpha 300 R-Raman Imaging Microscope with the excitation at 532 nm, the diameter of focus spot is about 8 μm, which is centered at the printed lines. X-ray photoelectron spectroscopy (XPS) was measured on an ESCALAB 250Xi spectrometer.

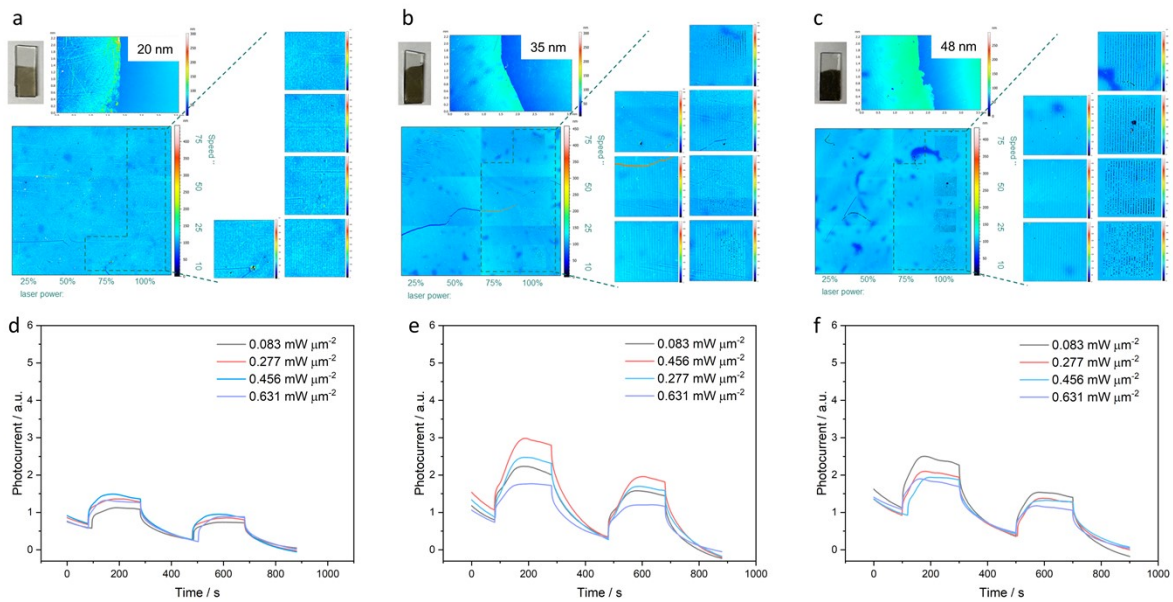


**Figure S1.** XPS S 2p spectra of MoS<sub>2</sub> and FPE-MoS<sub>2</sub>.

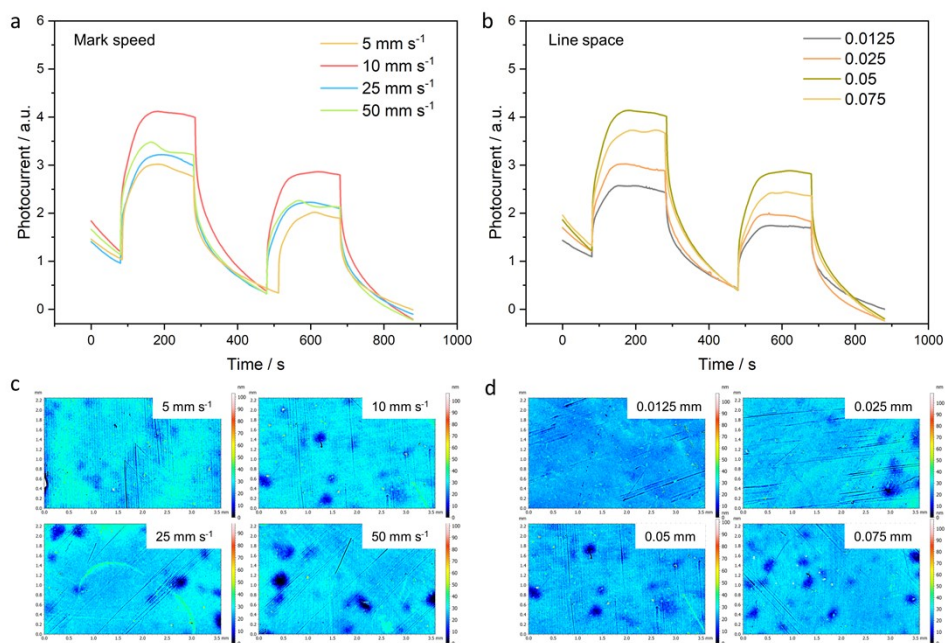


**Figure S2. Investigation of laser-induced MoS<sub>2</sub> surface temperature.** Light microscopy image of a 0.456 mW μm<sup>-2</sup> laser irradiation (20ms) on a 48 nm thick MoS<sub>2</sub> absorber with (a) a C<sub>20</sub>H<sub>42</sub> alkane thin film and (b) a C<sub>40</sub>H<sub>82</sub> alkane thin film. (c) Different laser irradiation parameters on 20 nm thick MoS<sub>2</sub> absorber, coated with a C<sub>20</sub>H<sub>42</sub> alkane thin film.

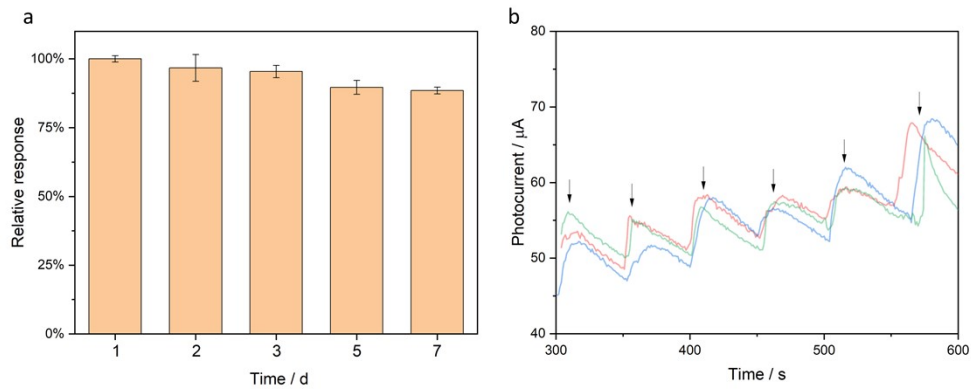
Here, the shorter chain alkane forms a melting ring (R<sub>melt</sub>) and a rupture hole (R<sub>boil</sub>) on 48 nm thick MoS<sub>2</sub> absorber after the laser irradiation, while the longer chain alkane only shows a melting ring (R<sub>melt</sub>). According to the boiling temperature of C<sub>20</sub>H<sub>42</sub> (373 °C) and C<sub>40</sub>H<sub>82</sub> (522 °C), the temperature on a 48 nm thick MoS<sub>2</sub> absorber could be estimated between 373 and 522 °C. Overlapping of the corresponding melting and boiling rings both appear on the 20 nm thick MoS<sub>2</sub> absorber, so that the temperature on all MoS<sub>2</sub> absorbers could be confirmed to reach >343 °C in the spot center.



**Figure S3.** (a, b, c) Optical images and vertical scanning interferometry (VSI) measurements of MoS<sub>2</sub> films of different thicknesses and line patterns fabricated with different laser parameters. (d, e, f) Corresponding photocurrent responses at 0.5 V vs. Ag/AgCl under white light irradiation.



**Figure S4.** Photocurrent responses on 35 nm thin MoS<sub>2</sub> film, processed with 0.277 mW  $\mu\text{m}^{-1}$  laser irradiation at (a) different laser mark speeds and (b) different laser pattern line spacing, at 0.5 V vs. Ag/AgCl under white light irradiation. (c, d) Corresponding vertical scanning interferometry (VSI) images.



**Figure S5.** (a) Stability measurements of the resultant FPE-MoS<sub>2</sub> electrode over 7 days. (b) Reproducibility measurement of three different electrodes.

## References

- [1] Eda, G.; Yamaguchi, H.; Voiry, D.; Fujita, T.; Chen, M. W.; Chhowalla, M. *Nano Lett.*, **2011**, 11, 5111-5116.
- [2] Zhang, J.; Zou, Y.; Eickelmann, S.; Njel, C.; Heil, T.; Ronneberger, S.; Strauss, V.; Seeberger, P. Savateev, A.; Loeffler, F. *Nature Communication*, **2021**, 12, 3224.