

## Supporting Information

# Self-Limiting Opto-Electrochemical Thinning of Transition Metal Dichalcogenides

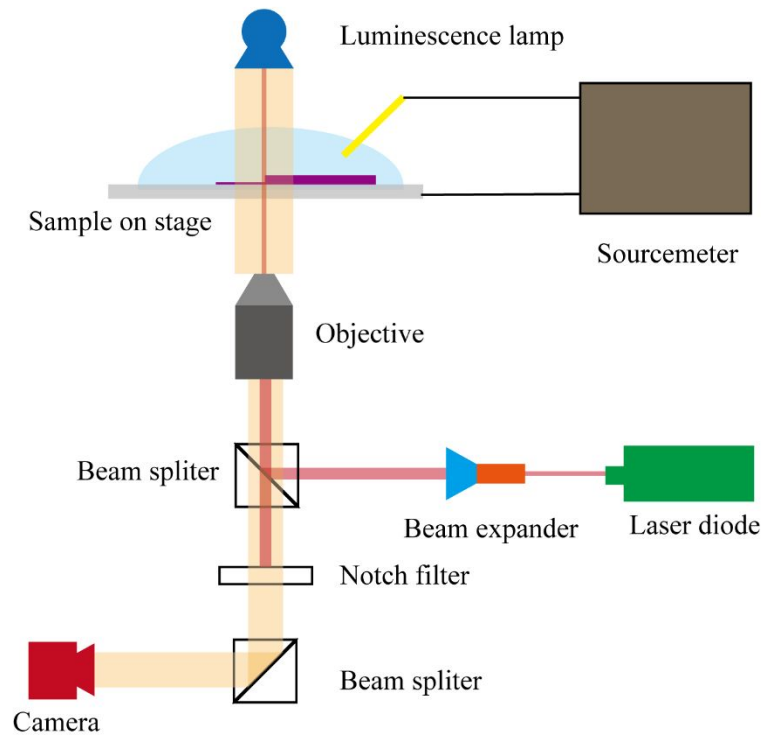
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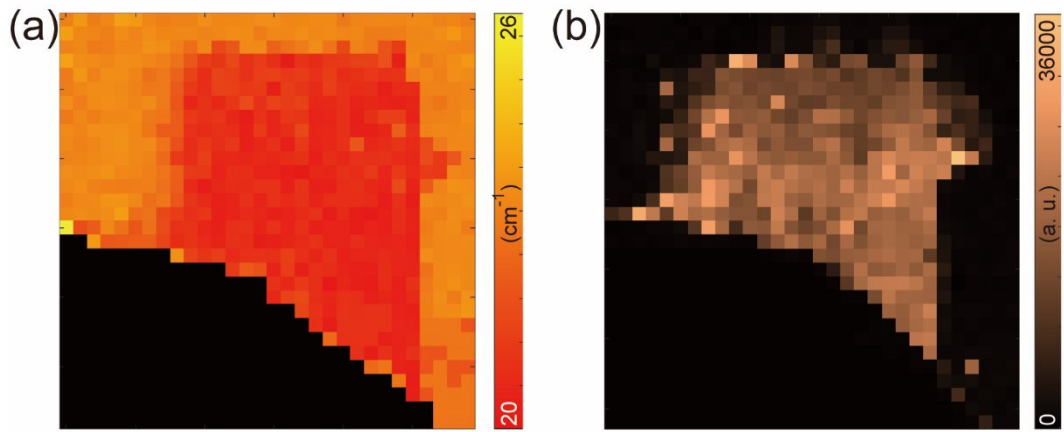
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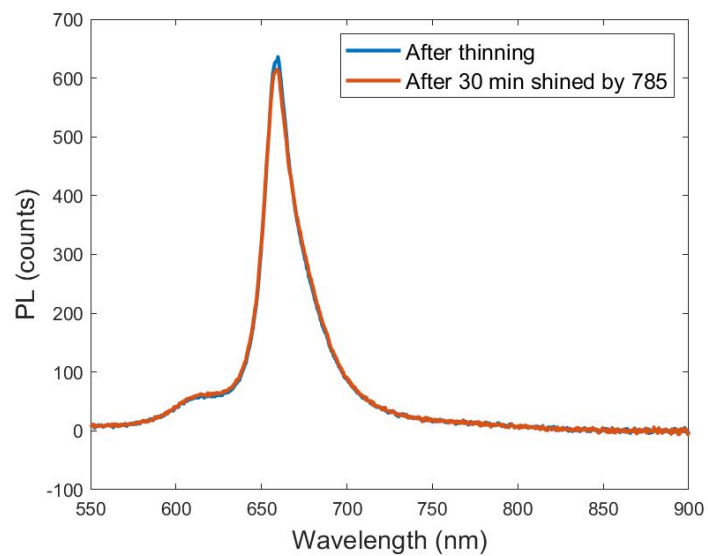
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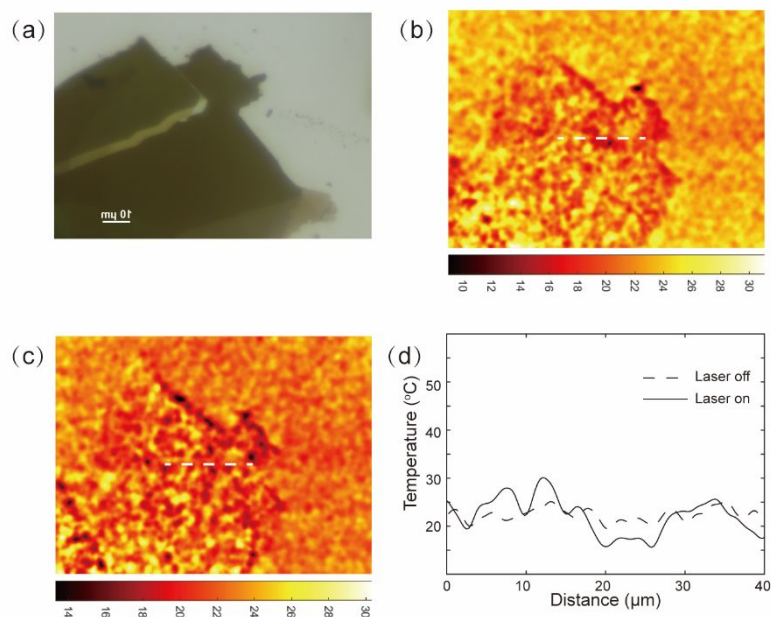
**Figure S1.** Schematic showing the experimental setup of sOET.



**Figure S2.** Raman scattering and PL mapping of the thinned  $\text{MoS}_2$  in Figure 1. (a) Spatial mapping of the Raman frequency distance between  $E_{12g}$  mode and  $A_{1g}$  mode of  $\text{MoS}_2$ . (b) Spatial mapping of the integrated PL intensity from 600 nm to 750 nm. Both the reduced Raman frequency distance of  $\sim 20.5 \text{ cm}^{-1}$  and enhanced PL intensity indicate the monolayer feature in the laser scanned area.

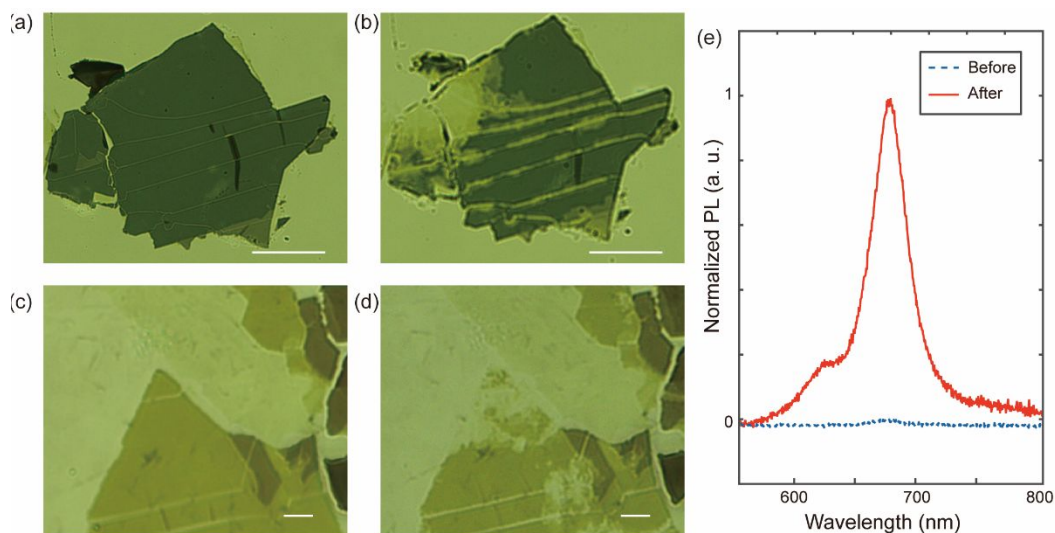


**Figure S3.** PL spectra of the thinned MoS<sub>2</sub> before and after laser excitation of 30 min. No obvious difference in the PL intensity was observed.

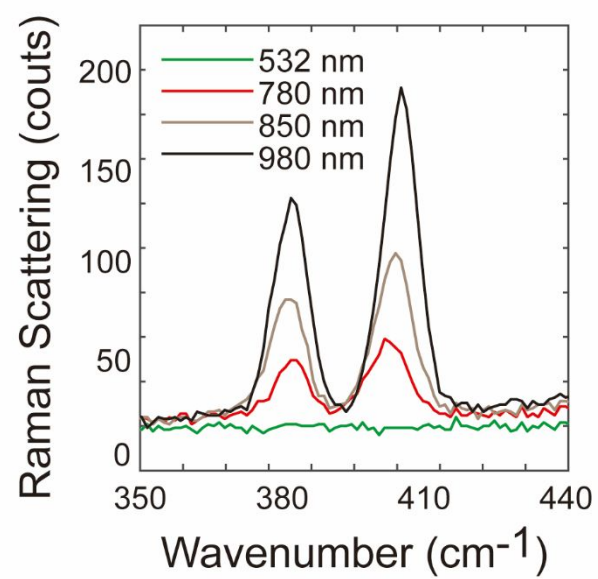


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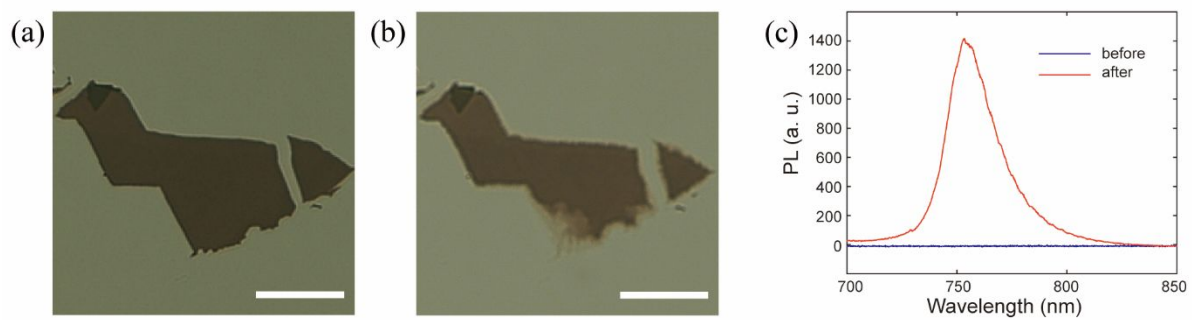
**Figure S4.** Temperature increase induced by a 0.102 mW  $\mu\text{m}^2$  785 nm lasers irradiation on a MoS<sub>2</sub> flake in DI water. (a) Optical image of the MoS<sub>2</sub> flake; (b) Background temperature distribution when the laser was off, the ambient temperature was  $\sim 22$  °C; (c) Temperature distribution when a 785 nm laser was directed on the MoS<sub>2</sub> flake. (d) Cross-sections of the temperature distribution with the laser off/on made at the white dashed line in (b) and (c). Scale bar: 10  $\mu\text{m}$ .



**Figure S5.** Thinning of MoS<sub>2</sub> on thin gold film and graphene with no bias. a-b) Optical images of a MoS<sub>2</sub> flake on 5 nm gold film before (a) and after (b) illuminated by a  $0.256 \text{ mW } \mu\text{m}^{-2}$  785 nm laser for 30 s in DI water. c-d) Optical images of a MoS<sub>2</sub> flake on monolayer graphene (glass substrate) before (c) and after (d) illuminated by a  $0.257 \text{ mW } \mu\text{m}^{-2}$  785 nm laser for 30 s DI water. Even without applying bias, the thickness of the multilayer MoS<sub>2</sub> was reduced. (e) PL spectra before (blue dashed) and after thinning. Scale bars are 25  $\mu\text{m}$ .



**Figure S6.** Raman spectra of MoS<sub>2</sub> flakes after scanning by different lasers.



**Figure S7.** sOET of WSe<sub>2</sub>. (a) Optical image of the WSe<sub>2</sub> flake before thinning. (b) Optical image of the WSe<sub>2</sub> flake after thinning. (c) PL spectra before and after thinning. Scale bars are 10 μm.