## Supporting Information for

# Substrate modified thermal stability of mono- and few-layer MoS<sub>2</sub>

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## 1. Experimental setup for thermal annealing

The exfoliated  $MoS_2$  flakes on different substrates (mica,  $SiO_2/Si$ .  $Al_2O_3$ ) were pushed into the thermostatic zone of a quartz tube, heated to 250, 270, 290 and 300 °C at a rate of 20 °C /min , and then held for 15 minutes before opening the lid to cool. See the methods for details.



Fig. S1 Schematic diagrams of the thermal annealing equipment setup.

## 2. Evidence of the presence of MoO<sub>3</sub> particles after thermal annealing

Comparing Fig. S2a and Fig. S2b, it was found that part of the bilayer region was thinned into a monolayer after thermal annealing at 290 °C. Meanwhile, particles appeared in the thinned area, as shown in Fig.S2c. These particles were most likely to be MoO<sub>3</sub>.



Fig. S2 MoO<sub>3</sub> particles on SiO<sub>2</sub> substrate after thermal annealing. Optical images of a bilayer MoS<sub>2</sub> before (a) and after (b) annealed at 290 °C under 10 kPa pressure. (c) AFM topography image after the annealing at 290 °C.

#### 3. Method of calculating the length of etching

Equation s1.1 shows the calculation of the average length of etched triangles, where L1, L2 and L3 represent the three side lengths of a triangle. Fig. S3 specifically describes the measurement of one of the triangles.



Fig. S3 The calculation of average triangle length.

#### 4. Etching of MoS<sub>2</sub> flakes on mica substrate

Fig. S4a and S4b are microscopic images of MoS<sub>2</sub> flakes on mica before and after thermal annealing. After annealed at 270°C, many etched triangular pitches appeared on the monolayer MoS<sub>2</sub>, while the bilayer and multilayers MoS<sub>2</sub> remained less changed. Fig. S4c shows an AFM image of Fig. S4b, emphasizing the different etching behaviors between monolayer MoS<sub>2</sub> and multilayer MoS<sub>2</sub>.



Fig. S4 Opical images of  $MoS_2$  on mica before (a) and after (b) annealed at 270 °C at 10 kPa pressure. (c) AFM topography image of  $MoS_2$  on mica after annealed at 270 °C at 10 kPa pressure.



#### 5. Thicknesses of monolayer MoS<sub>2</sub> on different substrates

Fig. S5 (a) AFM images and (b) height scanning curves of the monolayer  $MoS_2$  on  $Al_2O_3$ , SiO<sub>2</sub> and mica substrates.

#### 6. Determination of mica surface crystal orientation and composition

Referring to Fig. S6a, after comparison with the standard card, the mica we used was found to have a monoclinic crystalline structure, and the normal crystalline orientation is along the <001> direction. Fig. S6c shows the XPS spectra of mica, with a detailed analysis of each peak; the content of each element in mica can be semiquantitatively determined. Its chemical formula can be written as KAl<sub>2</sub> [AlSi<sub>3</sub>O<sub>10</sub>] [OH]<sub>2</sub>.



**Fig S6 (a)** Diffraction image and **(b)** SEM image of mica, demonstrating a monoclinic crystal of space group C2 with a (001) crystal plane, whose orientation is in the <001> direction perpendicular to the crystal plane. **(c)** XPS measurement showed that the chemical formula of the mica used is KAl<sub>2</sub> [AlSi<sub>3</sub>O<sub>10</sub>] [OH]<sub>2</sub>.

#### 7. Schematic of mica crystal surface



**Fig. S7** A schematic showing mica crystal (001) surface structure (referring to Cheng, et.al, Ref. 45 in the maintext).

### 8. Thermal etching of MoS<sub>2</sub> grown by chemical vapor deposition (CVD)



Fig. S8 AFM topography images of CVD monolayer  $MoS_2$ , which were originally grown on a SiO<sub>2</sub>/Si substrate and then transferred to mica (a) and SiO<sub>2</sub>/Si (b) substrates. After annealed at 240°C for 15min at atmospheric pressure, the monolayer  $MoS_2$  flakes were almost completely etched both on mica (c) and on SiO<sub>2</sub>/Si (d) substrates.