### **Electronic Supplementary Information**

# Fast and efficient electrochemical thinning of ultra-large supported and free-standing MoS<sub>2</sub> layers on gold surfaces

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#### A) Height profiles

Figure S1a-b shows AFM images of MoS<sub>2</sub> flake before and after electrochemical thinning process. The cyan line in both images indicates the region where height profiles were acquired. Figure S1c shows that the initial height of the flake is approximately 150 nm. After electrochemical thinning process, the edges are higher than the basal plane, as indicated in Figure S1d. In order to obtain a more reliable height profile in the center region of the flake we transfer the electrochemically thinned MoS<sub>2</sub> flake onto a Si substrate, which has a smoother surface. The transfer process was done using the same sucrose method used to transfer the bulk flakes. Figure S1e and S1f shows the images of two thinned flakes on Si substrate. As can be observed, folds and wrinkles are observed in the image due to the transfer process. On Si it was possible to set a reliable baseline from edge to edge, as indicated in Figure S1g-h. As can be observed, the height profile reached approximately a monolayer (0.85 nm) in the center region of thinned MoS<sub>2</sub>, as can be observed in Figures S1g-h.



**Figure S1.** AFM image before (a) and after (b) electrochemical thinning process. c-d) Height profiles before and after electrochemical thinning process on Au substrate, respectively. AFM images of thinned flakes transferred to Si substrates (e,f) and their respective height profiles (g,h).

## B) Raman spectra



Figure S2. Raman spectra at the thinned area.

## C) Single-flake electrochemistry



**Figure S3.** Cyclic voltammograms (i) and optical microscope images (ii-v) obtained for flakes of 60 (a) and 480 nm (b). The CVs were obtained in 0.5M  $H_2SO_4$  at 50 mV s<sup>-1</sup>. The area of the flakes are (a) 9984  $\mu$ m<sup>2</sup> and (b) 21260  $\mu$ m<sup>2</sup>. The scale bars in (ii)-(v) are 100  $\mu$ m.

## D) Ultra-large MoS<sub>2</sub> flakes



**Figure S4.** Optical microscope image of an ultra-large area thinned  $MoS_2$  flake. The length of black and red dashed lines are: (a) 554.5 and 511.0  $\mu$ m and (b) 813.5 and 410.8  $\mu$ m, respectively.

#### E) Electrochemical thinning on FTO electrode

We investigated the electrochemical thinning process of  $MoS_2$  on FTO electrodes. By using the same transfer method, the transfer yield was lower when compared to Au. Next, we conducted the same exfoliation process and monitored large flakes. As can be observed in Figure S5 below large regions at the center and bottom right in the image, highlighted by the red line, were lixiviated from the surface.



**Figure S5.** a) Large MoS<sub>2</sub> flake on FTO substrate. b) Electrochemically thinned MoS<sub>2</sub> flake. c) Raman spectra obtained at two different positions of the substrate. The black dots represent the position where the spectra was taken.

F) Electrochemical thinning in the absence of  $O_2$ 



**Figure S6.** Optical microscope image of  $MoS_2$  before (a) and after (b) electrochemical thinning in the absence of dissolved oxygen. We purged the electrolyte with  $N_2$  for 1h before the electrochemical experiment. Scale bars are 50  $\mu$ m.

#### G) Raman mapping

Raman maps were obtained after electrochemical thinning of  $MoS_2$  flakes of different thicknesses. Figure S7 shows the stereomicroscope images of the thinned flakes (i-iii) and their respective raman maps (iv-vi) at the indicated region (dashed rectangle), respectively. The arrows in the maps indicate the width of the bulk left on the edges. The measured width was the following: (iv)  $2.0 - 5.0 \mu m$ , (v)  $4.5 - 6.0 \mu m$  and (vi)  $9.0 - 12 \mu m$ .



**Figure S7.** (I - iii) Optical microscope images of the electrochemically thinned flakes and their respective raman maps (iv – vi). The thicknesses of the bulk flakes before the thinning process was 60 nm (i), 143 nm (ii) and 480 nm (iii).  $MoS_2$  flakes (i) and (iii) were the same used in Figure S3 while flake (ii) was shown in Figure 2.

# H) AFM images of $MoS_2$ suspended on the microhole



Figure S8. a) AFM image of electrochemically thinned  $MoS_2$  on the microhole. b-c) Height profiles.