Supporting Information

Topography preserved microwave plasma etching for top-down layer engineering in MoS_2 and other van der Waals materials

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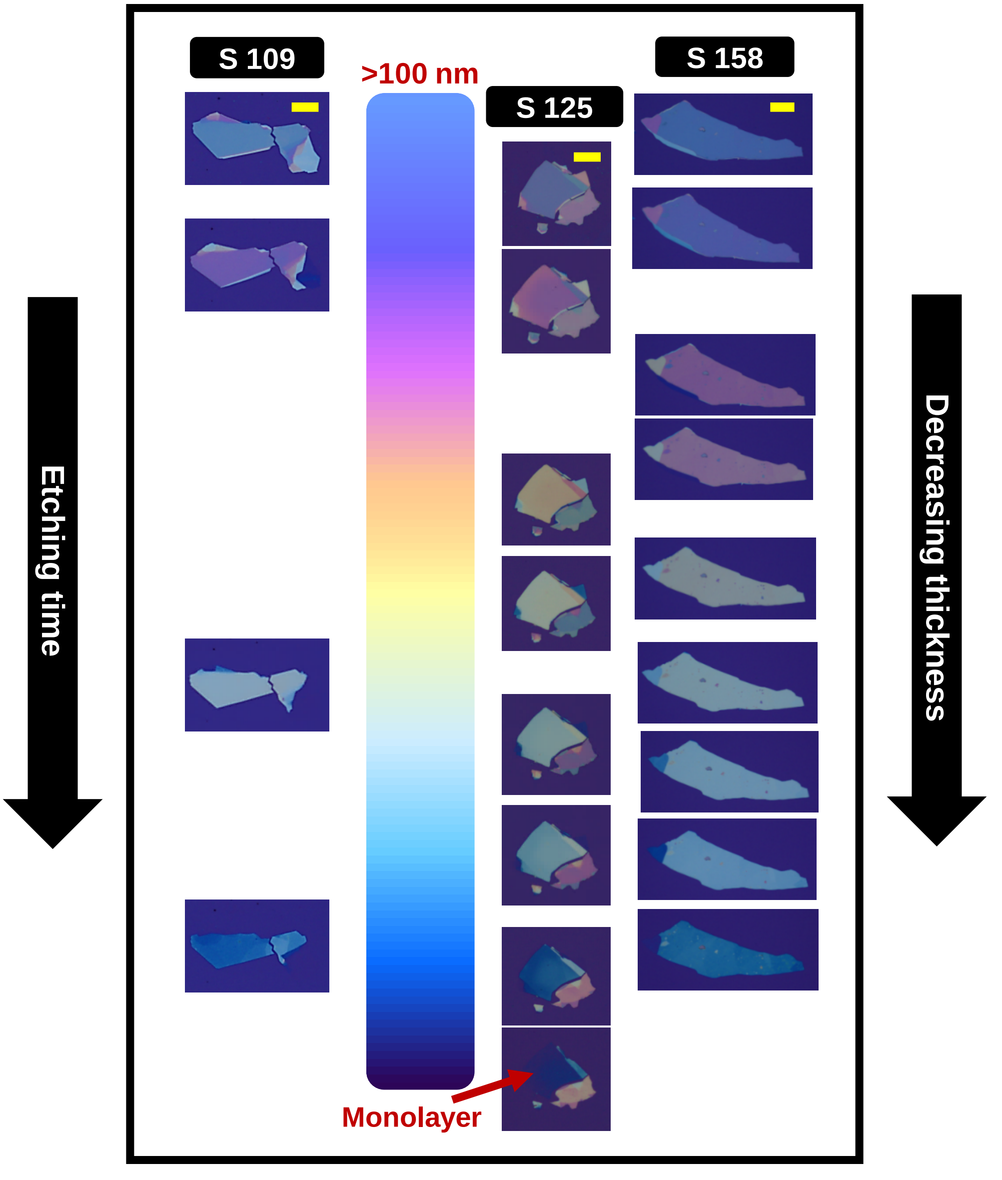
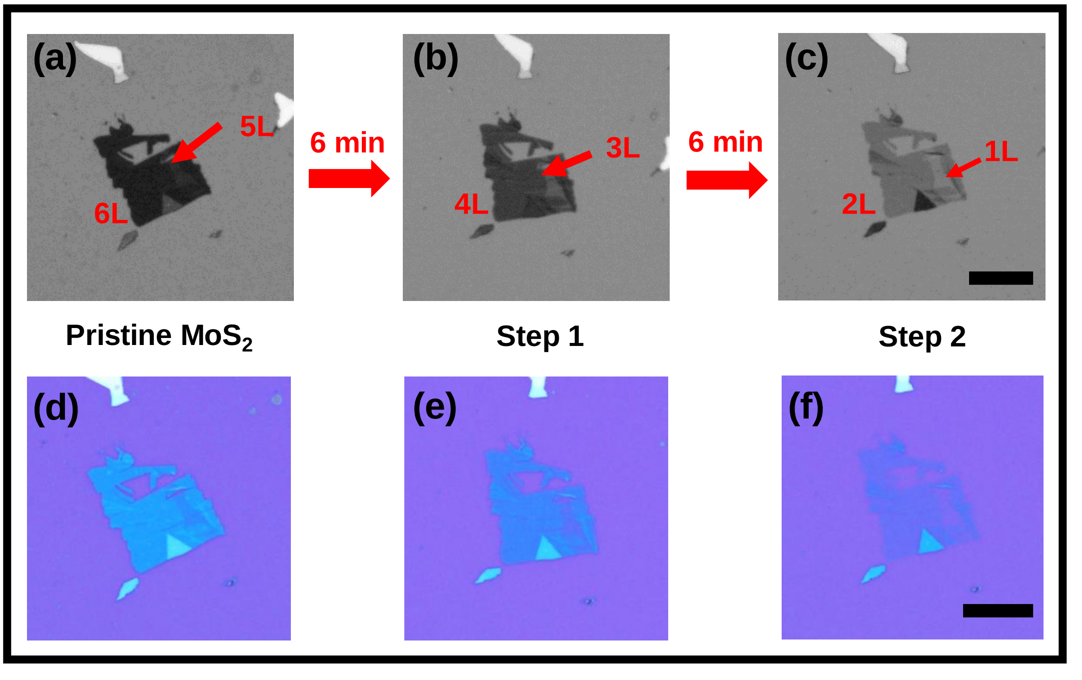
S1: Thickness dependent color profile of MoS_2 flakes



The optical images of MoS_2 flakes on 300 nm SiO_2/Si substrate taken using Olympus BX51M trinocular microscope (with U-25LBD color temperature conversion filter, preset illumination and 10 ms exposure time) after various stages of etching are shown. Bulk MoS_2 flakes which are around 100 nm thick appear bright sky-blue in color. As the flakes are uniformly etched down they show a color change due to an increase in transparency on approaching the few-layer-limit. S-109, S-125 and S-158 are representative samples. S-125 has been thinned down to the monolayer after several controlled etching steps. The scale bars in yellow correspond to 20 µm.

S2: Thickness analysis using optical contrast method for the fine etch process

We utilize the thickness identification by optical imaging technique put forward by Wang *et al,* 2012 *Nanotechnology* **23** 495713. to determine the thickness of MoS_2 flakes upto 6 layers. To demonstrate the fine etching process and the layer-by-layer etching nature, we start with a sample having two distinct regions (plateaux) marked as 6L and 5L respectively in the R-channel image (a) below. The first step of fine-etching is carried out for 6 minutes, during which two layers are removed from either regions to yield 4L and 3L regions respectively as shown in (b). The second step, also for 6 minutes, leads to subsequent etching of 4L to 3L regions down to 2L to 1L respectively, preserving the initial topography as shown in (c). The scale bar is 20 µm. (a), (b) and (c) are the red channel images of (d), (e) and (f) respectively which are used to extract the thickness information using the optical contrast method.



S3: Roughness analysis of pristine and plasma exposed MoS_2 samples

Scan area 1 µm X 1µm and 2 µm X 2µm		
Sample code	RMS roughness (nm)	Average roughness (nm)
S-153	0.42	0.32
S-136	0.47	0.35
S-51 (pristine)	0.38	0.30
S-42 (pristine)	0.23	0.18

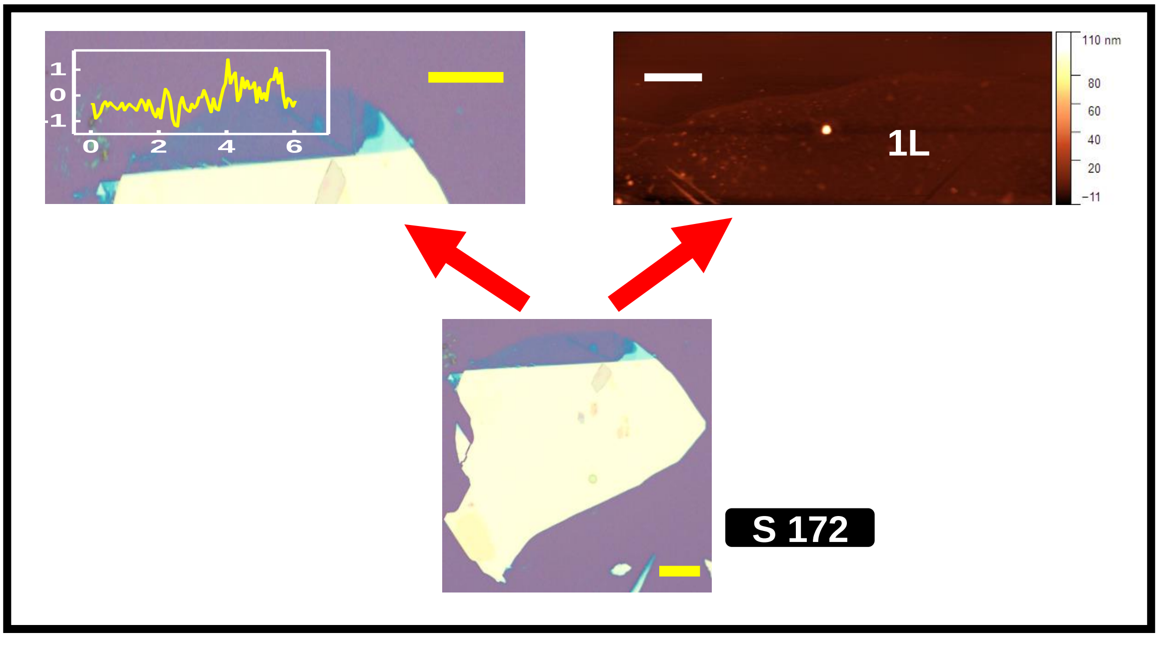
Roughness values

A Bruker Dimensions Edge atomic force microscope was used to obtain the height profile data for the above samples. The values of RMS roughness (R_q) and average roughness (R_a) are obtained using **Gwyddion**, a commonly used data processing software for AFM. The values were verified using **Nanoscope Analysis** software.

S4: AFM height profiles of monolayer and 4-layer samples

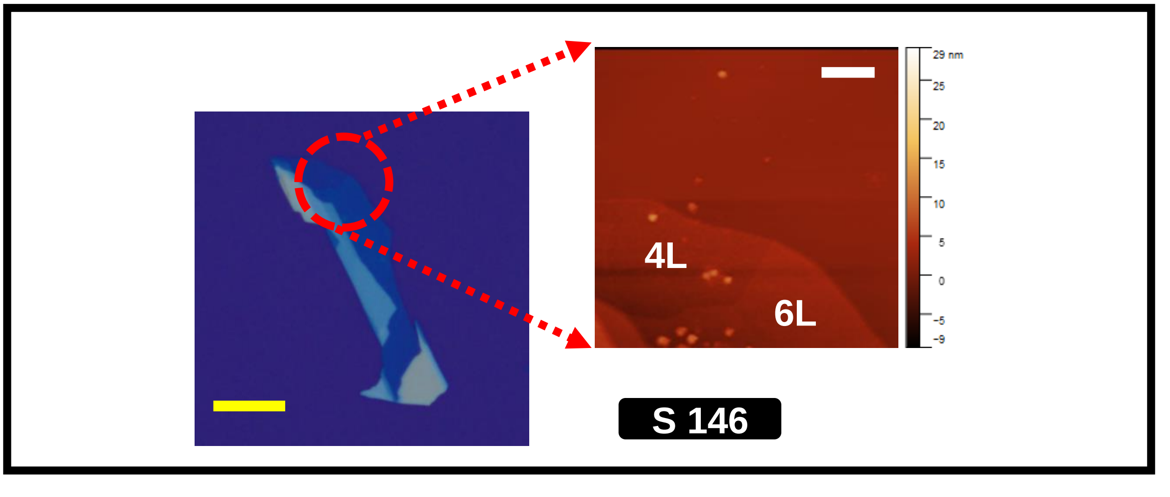
Monolayer MoS_2

AFM height profile and optical image of a large area plasma thinned monolayer MoS_2 (sample S-172). Scale bar is 20 µm.



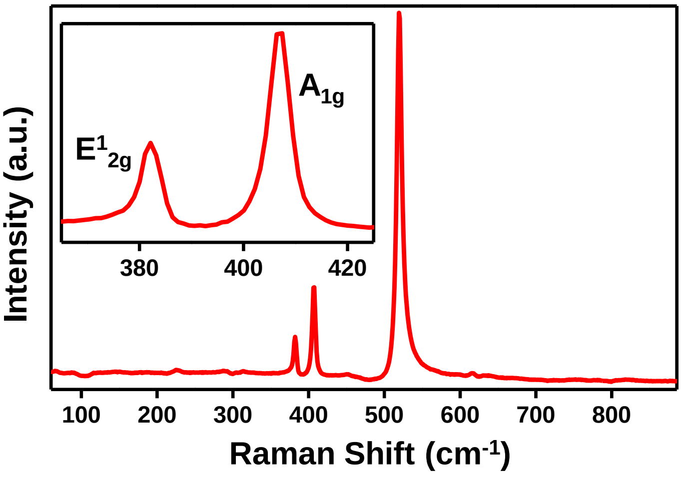
Four-layer MoS_2

AFM height profile and optical image of S-146 with regions corresponding to 6 layers and four layers as shown by AFM. The R-channel image of the sample is shown in the second column of Fig. 1 (b). Scale bar is 20 µm.

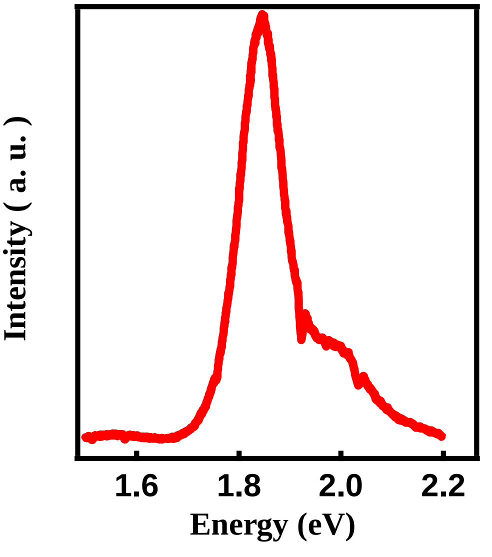


S5: Extended Raman Spectra of MoS_2

Wide range Micro-Raman Spectra of monolayer MoS_2 sample S- 172 using 532 nm excitation laser (Horiba Xplora Plus) is shown below. The characteristic E^1_{2g} and A_{1g} peaks are sharp and the peak at ~520 cm^{-1} corresponds to Si. The spectra shows no emergence of defect-related peaks. The characteristic Raman peaks of MoO_3 are also not observed. Similar spectra are observed for all of our plasma treated samples.

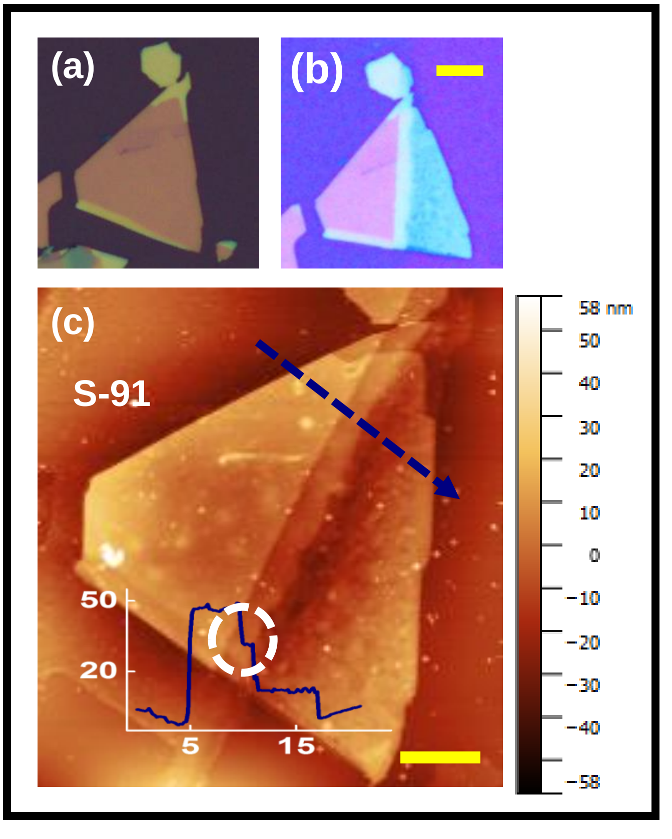


S6: Photoluminescence spectra of pristine monolayer sample



S7: Half etched sample (S-91)

We selectively etch-down a portion of a pristine sample exploiting the metal masking technique described in Fig. 1 (e) to study the change in surface roughness due to plasma exposure. Figure (a) shows the optical image of pristine sample S-91, and Figure (b) depicts the optical image of the sample after the metal-masking followed by etching. Figure (c) shows the AFM height profile of the sample; the left-half of the sample was masked while the right-half has been etched down to a thickness of 12 nm starting from a thickness of ~48 nm. The bottom inset shows the line-profile taken along the blue dashed arrow. The step in the height profile and the line-cut highlighted by the dotted circle in Fig. (c) is caused by the removal of a few layers of the material from the undercut region, an artefact of the photolithography. Scale bar is 20 µm.



S8: Transconductance of pristine MoS_2 sample

Transcondiuctance data of a pristine exfoliated MoS_2 sample. Inset shows optical image of the device. Scale bar is 20 µm.