Supporting Information
Surface Properties of Laser-treated Molybdenum Disulfide Nanosheets for Optoelectronic Applications

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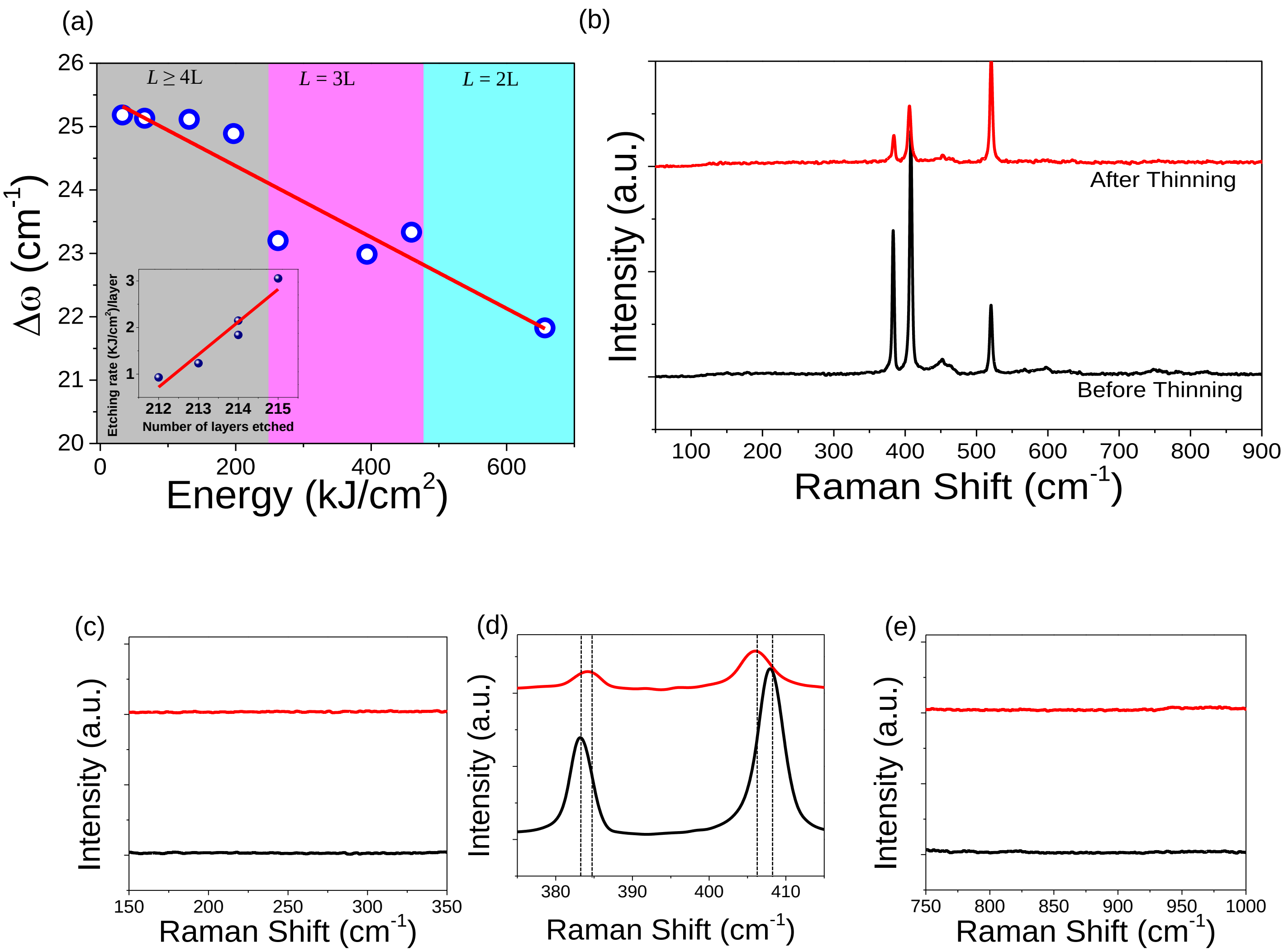

Figure S1. (a) Δω vs. the energy density of the incident 532nm laser. The fitted red line has a slope of 5.623×10^{-6} and a y intercept of 25.51. The inset shows the varying etching rate as a function of the etched number of layers. (b) Raman spectra before and after laser thinning. (c), (d), and (e) are zoomed in views of the spectra measured before and after laser thinning showing the absence of MoO_3 Raman peaks. The downshift of the A_{1g} peak after the treatment indicates successful laser thinning.

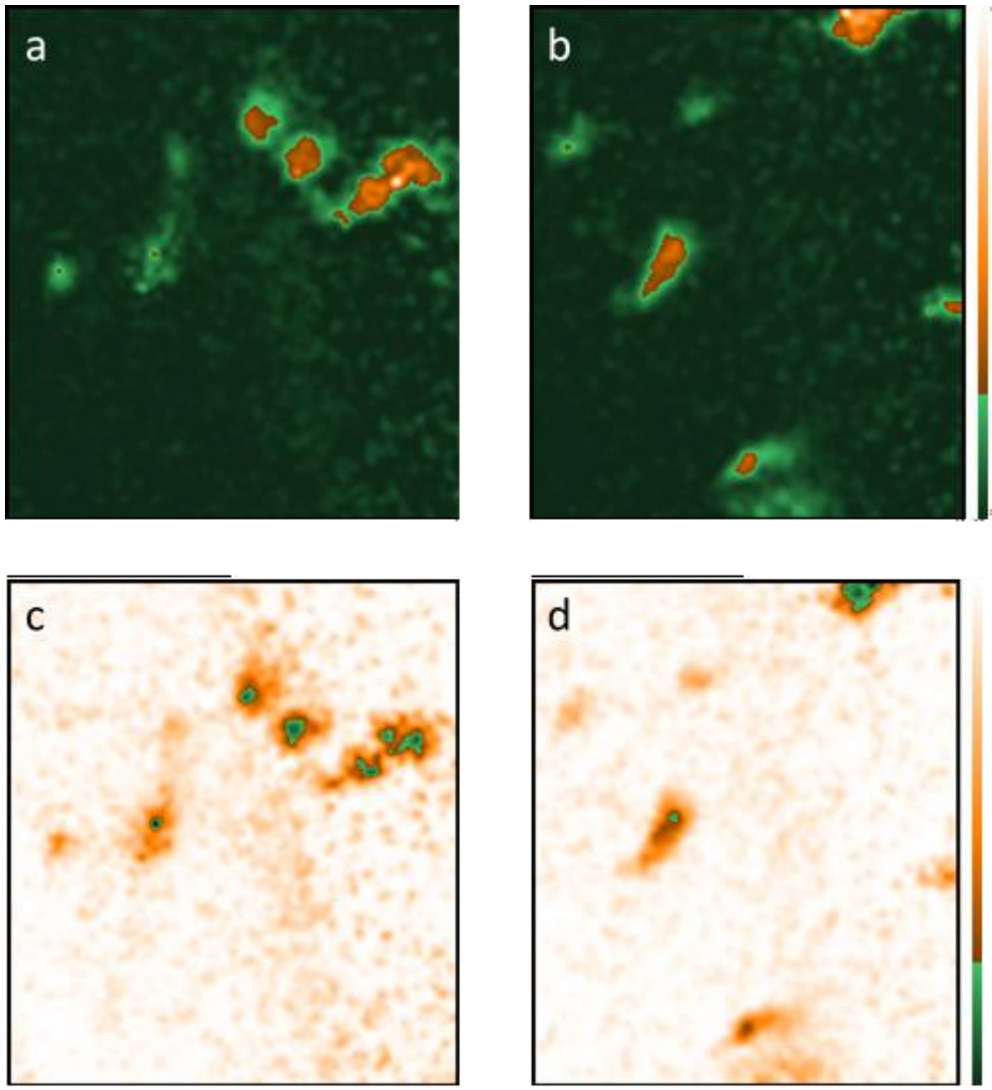

Figure S2. Processed MoS_2 $3d^5$ image of the laser-treated and untreated MoS_2 nanosheets under (a) ambient and (b) vacuum conditions. Processed Si 2p images under (c) ambient and (d) vacuum conditions. Image size was 161 µm × 178 µm for ambient conditions and 153 µm × 178 µm for vacuum conditions. Images were processed consistent with the Methods in the main text.

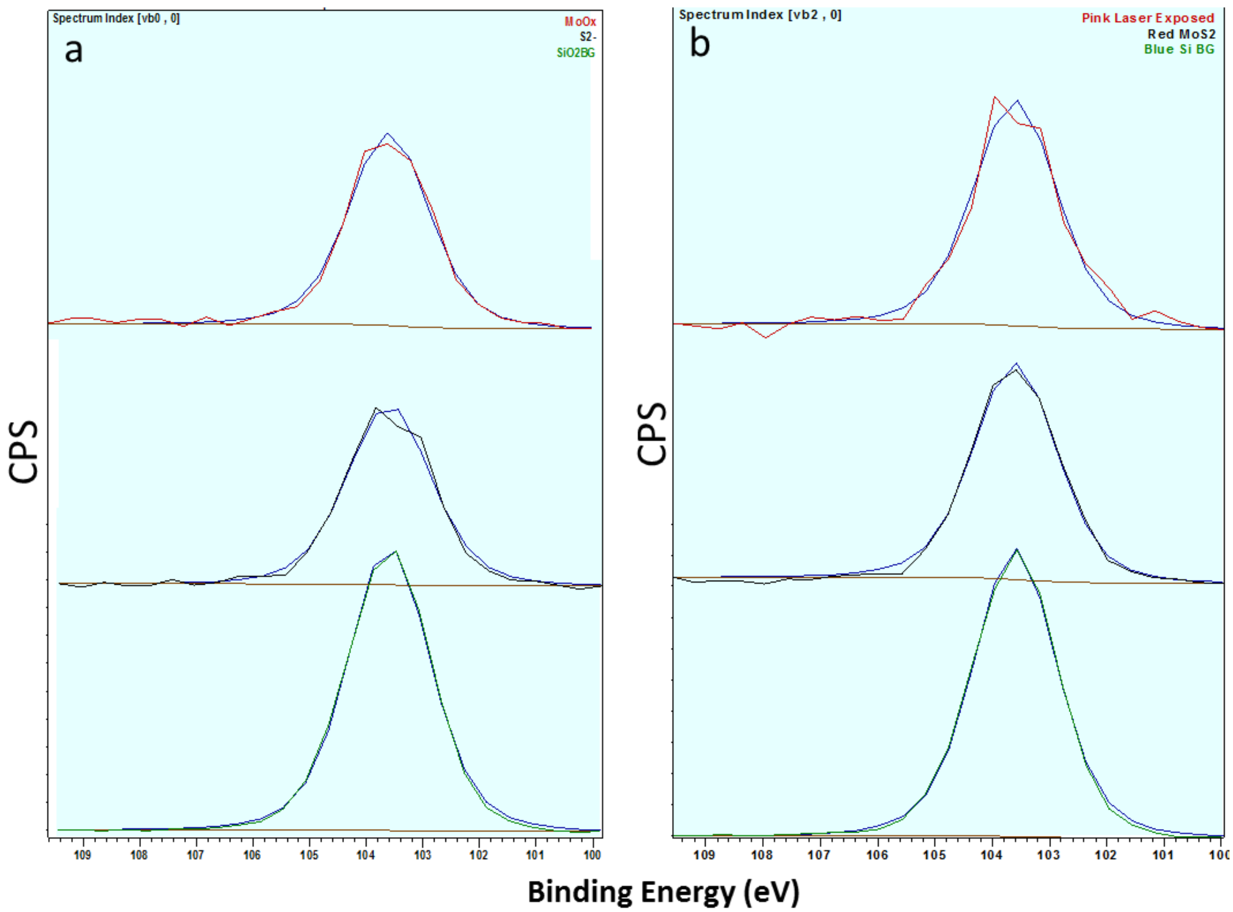

Figure S3: Si 2p extracted spectra for (top to bottom) laser-treated MoS_2, native MoS_2, and SiO_2 background for the (a) ambient and (b) vacuum conditions.

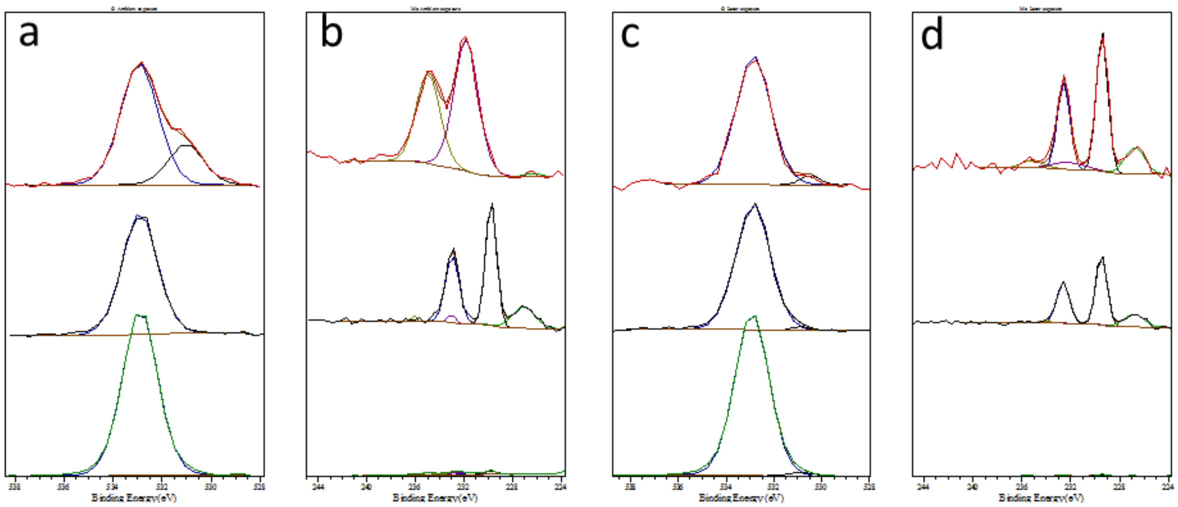

Figure S4: (a-d) Fitted spectra extracted from ROIs in the composite map for native and laser-treated MoS_2 nanosheets.

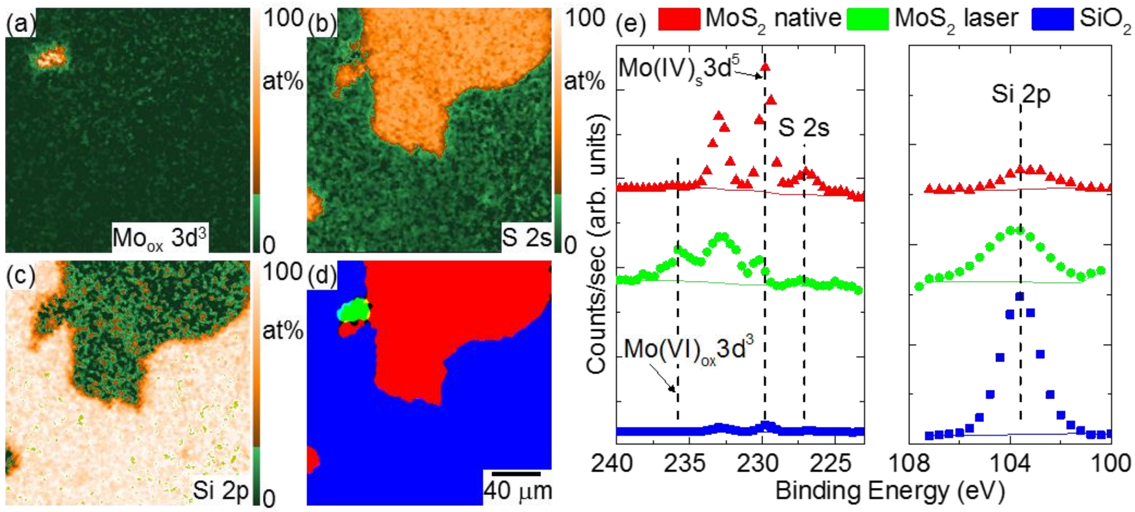

Figure S5: Replicate of ambient exposure conditions for *i*-XPS. (a) Mo_{ox}, (b) $S_{sulfide}$, (c) Si 2p and (d) composite mask images. (e) Extracted spectra from the raw image stack using the masks for the ROIs.

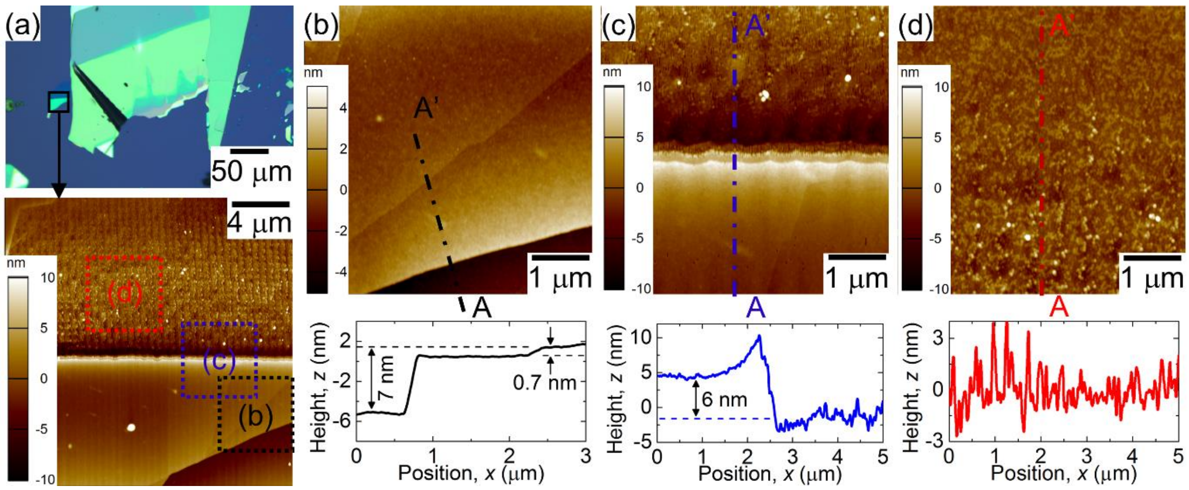

Figure S6. (a) Optical and AFM images of laser-thinned MoS_2. The highlighted part of the optical image shows the location of the nanosheet. AFM images of (b) native MoS_2 before thinning, (c) the edge between native MoS_2 and thinned MoS_2, and (d) MoS_2 after thinning.

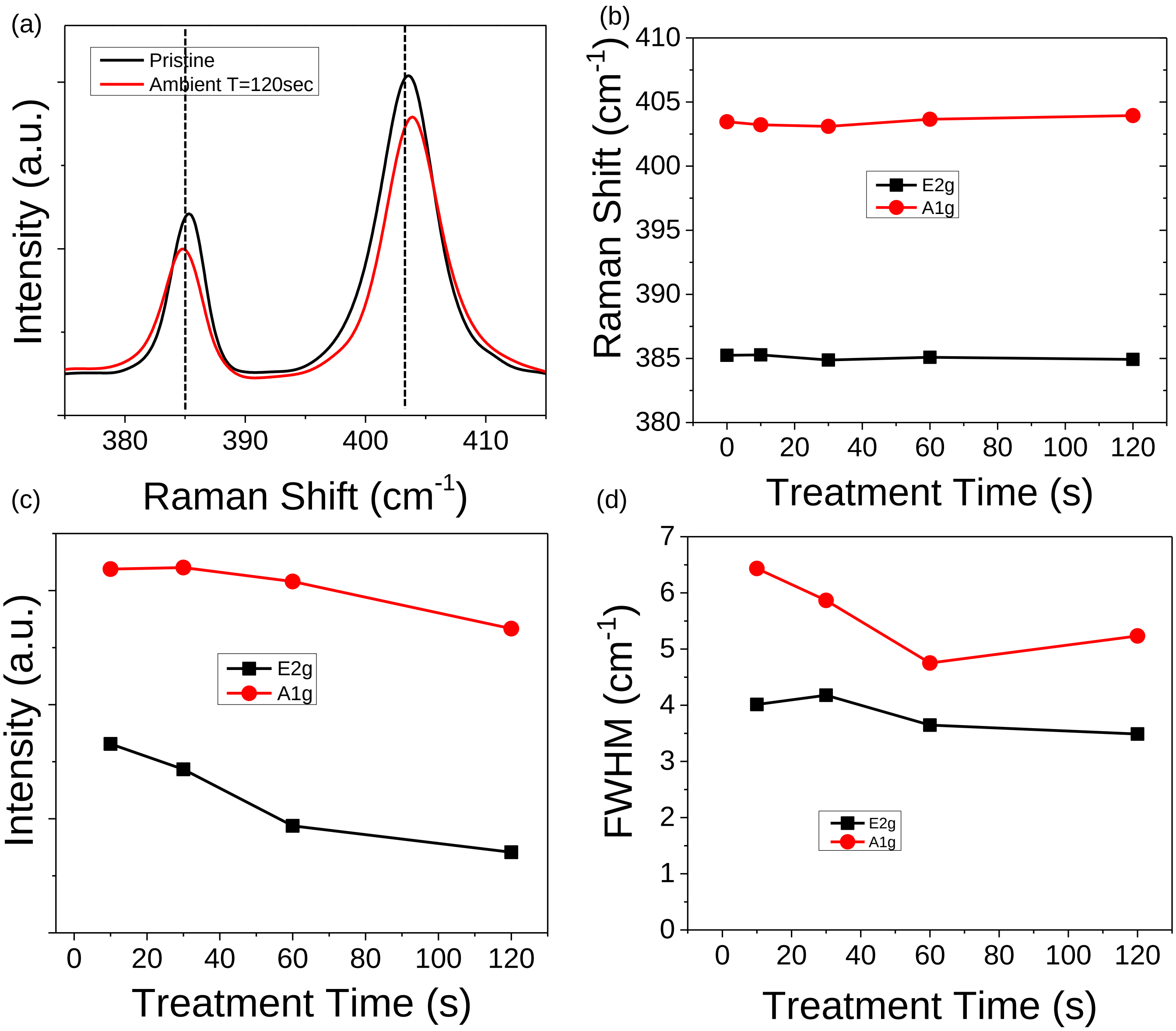

Figure S7. (a) Raman spectra before and after laser treatment in ambient conditions. (b) Shift (c) intensity, and (d) FWHM of the A_{1g} and E_{2g} vibrational peaks at different treatment times.

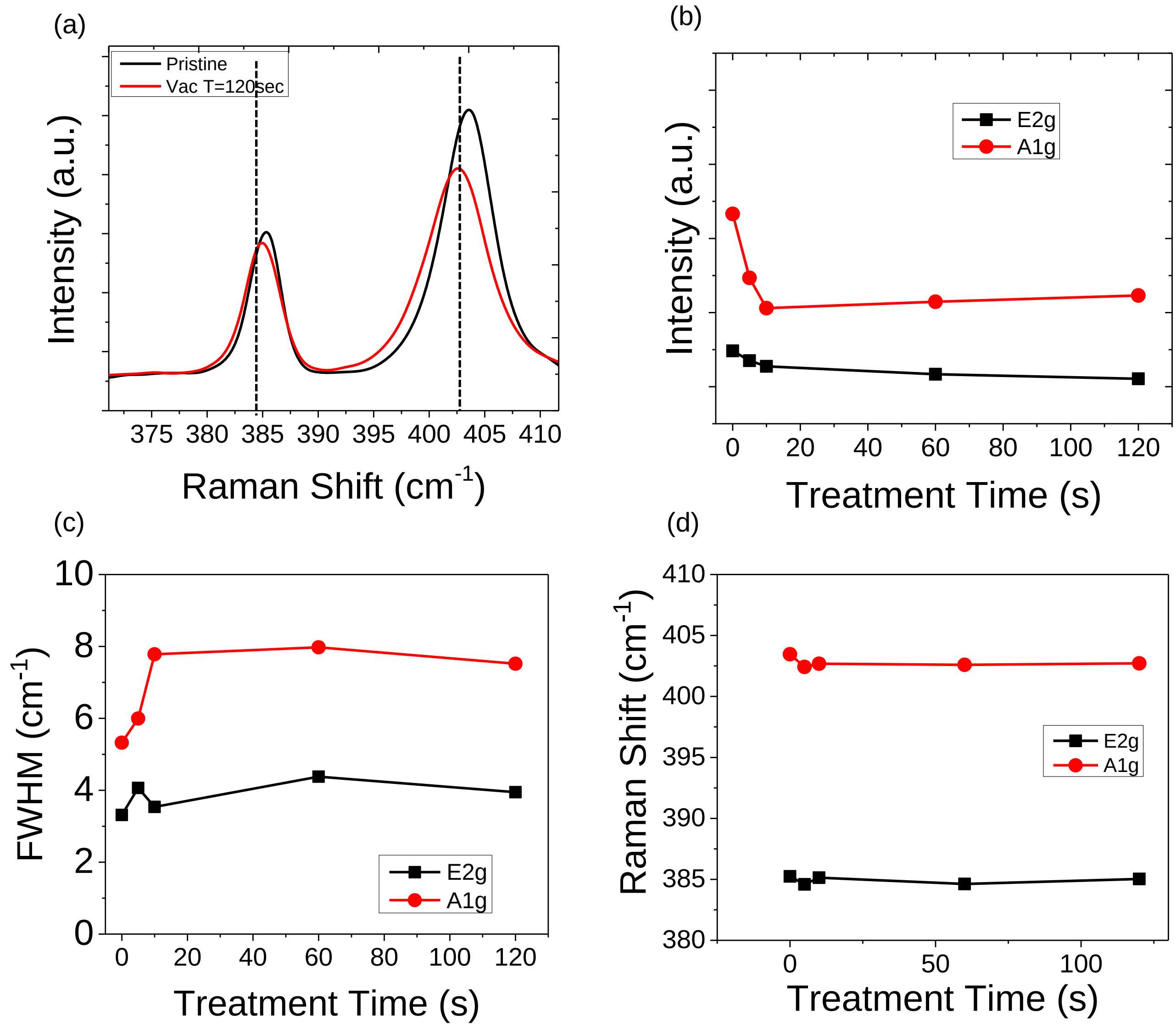

Figure S8. (a) Raman spectra before and after laser treatment in vacuum. (b) Intensity (c) FWHM, and (d) shift of the A_{1g} and E_{2g} vibrational peaks at different treatment times.

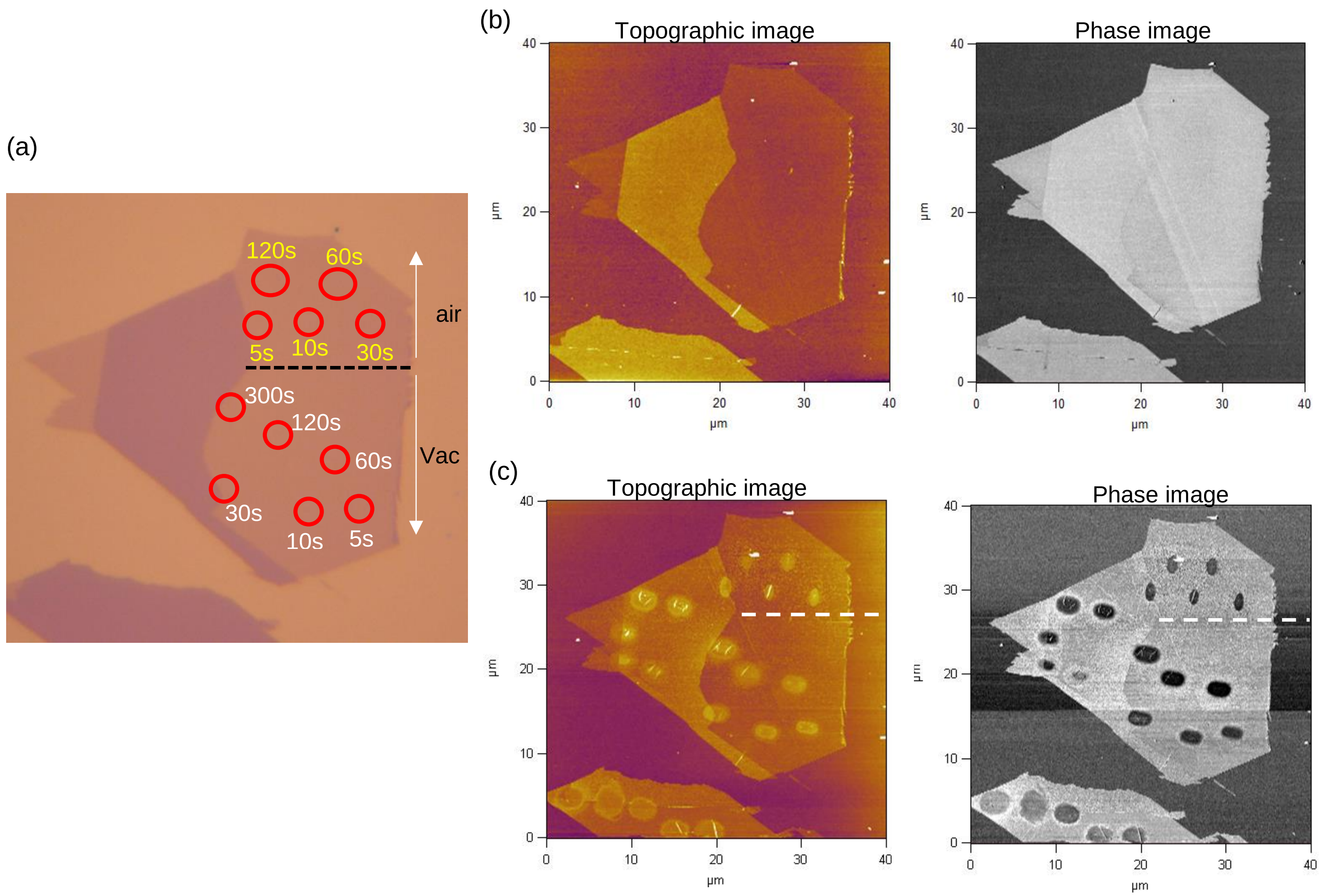

Figure S9. (a) Optical image of laser-treated monolayer in vacuum and ambient. AFM measurements of a laser-treated monolayer (b) before treatment (c) after treatment. The dashed line in (c) separates the regions between treatments in air and vacuum, as illustrated in (a).

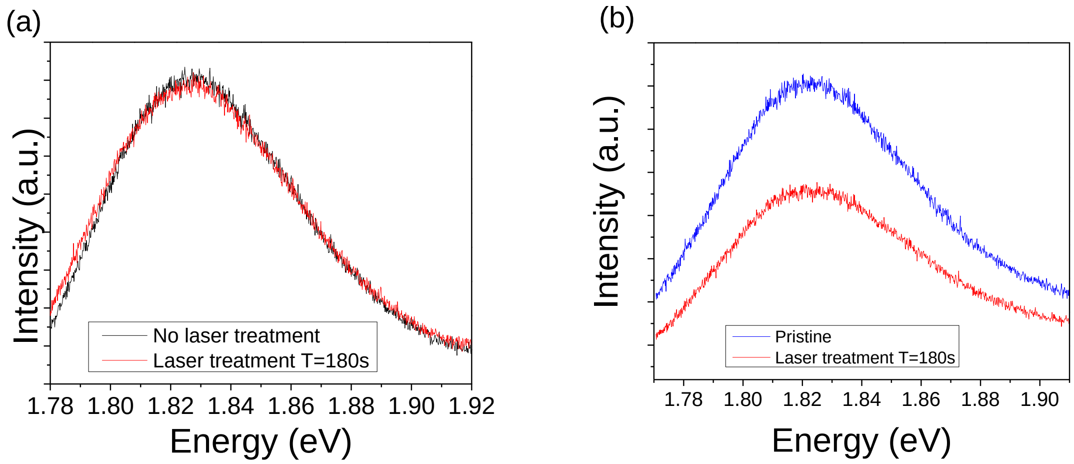

Figure S10. PL measurements before and after laser treatment using 0.93 mW for (a) 1L and (b) 5L MoS_2.

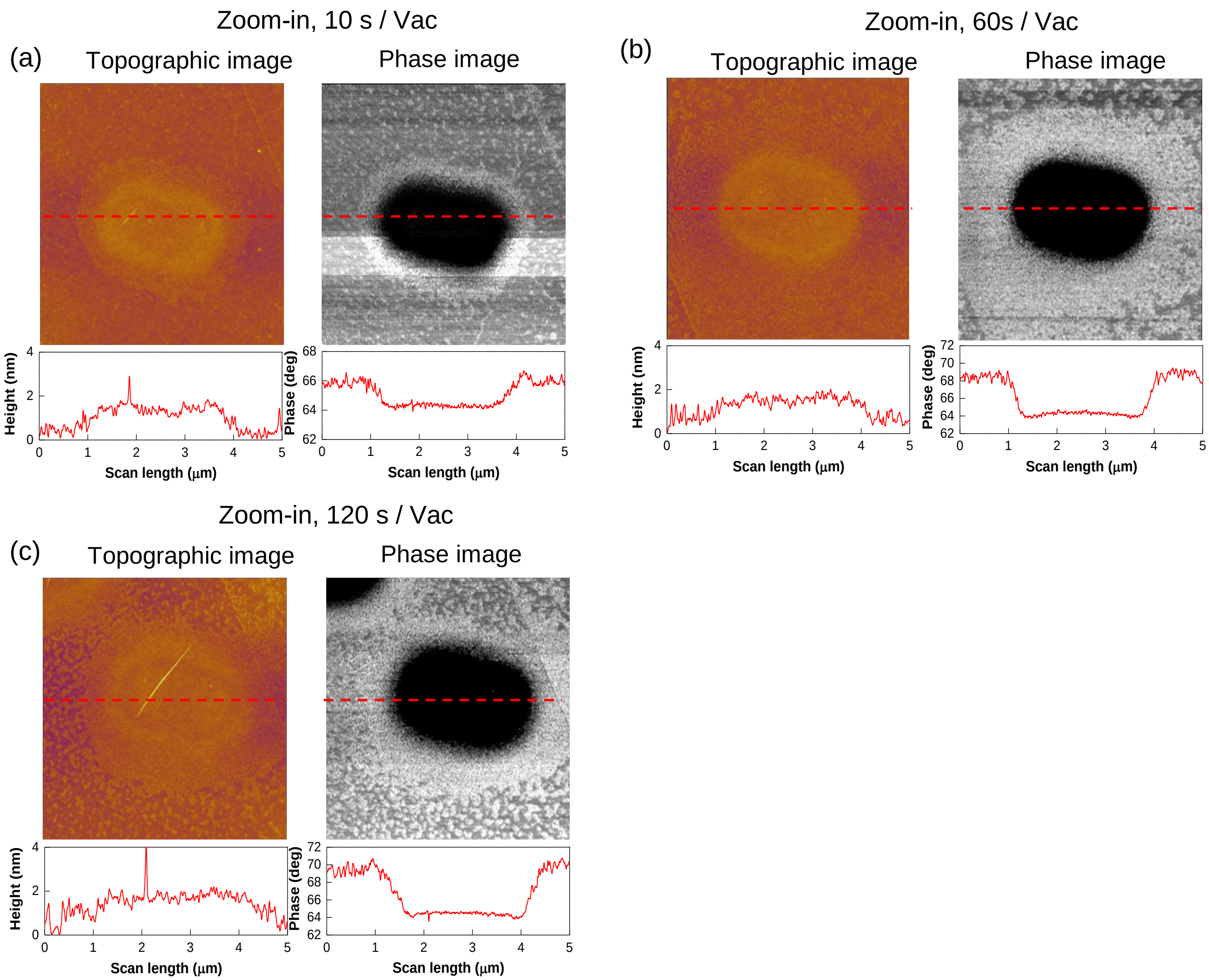

Figure S11. Selected laser-treatment spots in vacuum for (a) 10 s treatment (b) 60 s treatment, and (c) 120 s treatment.

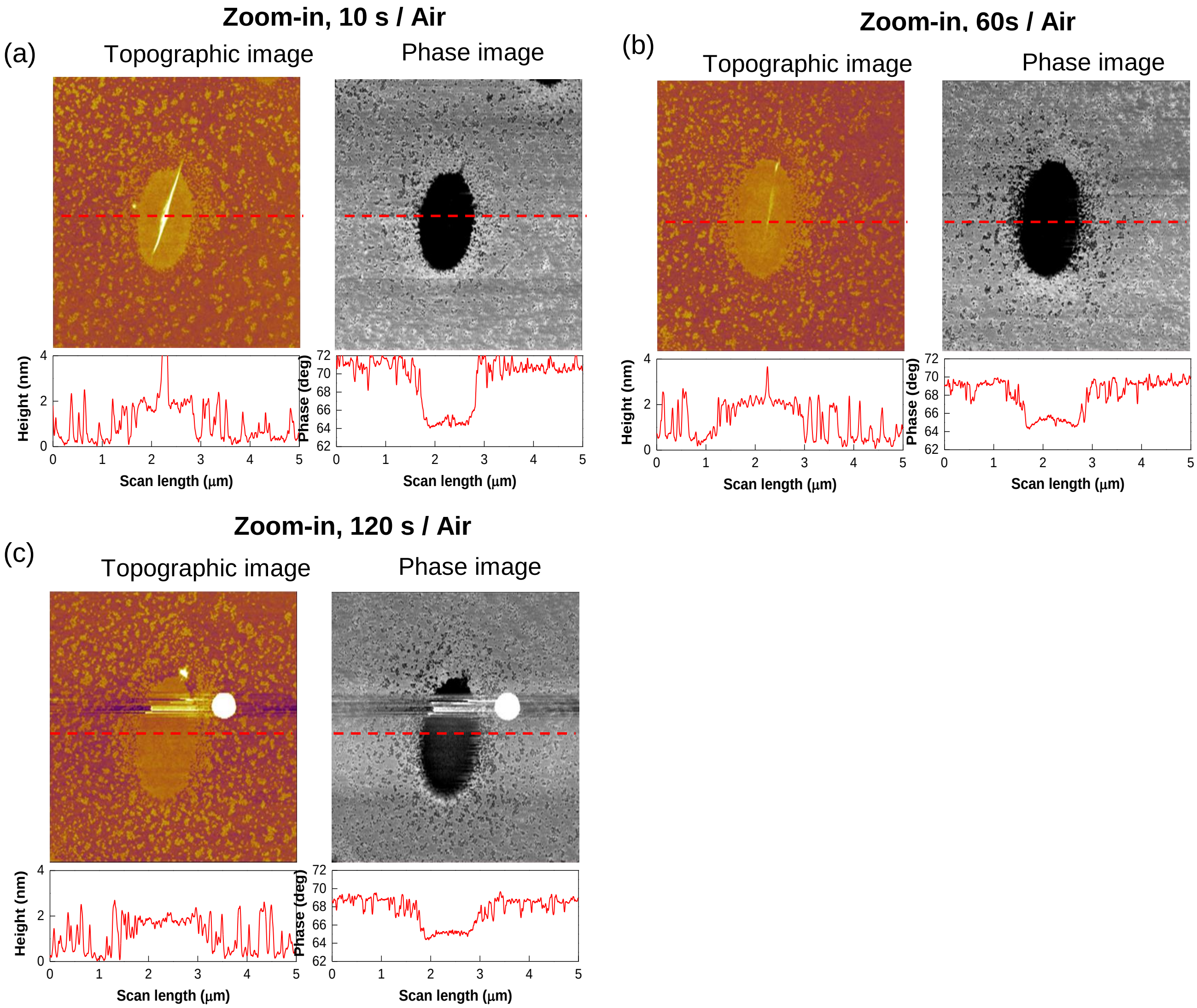

Figure S12. Selected laser-treatment spots in ambient for (a) 10 s treatment (b) 60 s treatment, and (c) 120 s treatment.

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(*This is the importer that W. A. Osborn made for importing the MWCNT images in the Gorham et al. Carbon 2016 paper*)
vmsImgImportFast[fp_] := Module[{str, retVal},
   str = OpenRead[fp];
   Skip[str, Record, 78]; (*sets the number of metadata rows to not import from the .vms file.*)
   retVal = Partition[ReadList[str, Number, 256^2], 256];
   (*takes the rest of the rows and turns them into 256 rows with 256 columns *)
   Close[str];
   retVal
  ];
(*-----------------------------------------------------------------------------------------------*)

(*This section was prepared by JM Gorham and is used for Batch processing all image files in a folder
 to be cropped by various, predetermined amounts to a new, uniform size*)
SetDirectory["C:\\Users\\..."]; (*sets the directory the files are extracted from and saved to*)
MoDataset = Map[vmsImgImportFast[#] &, FileNames["*.vms"]];
(*Imports the all images from set directory using the script Will wrote for the MWCNT imaging paper*)
StageDrift = {39, 35, 31, 36, 32, 30, 27, 26, 25, 23, 24, 22, 22, 21, 20, 18, 16, 20, 18, 19, 18, 18, 17, 16, 15,
  16, 14 , 15, 13, 13, 12, 14, 15, 13, 12, 13, 14, 13, 11, 13, 13, 12, 10, 13, 13, 8, 12, 11 , 10, 11, 11, 11, 10, 9
};
(*Manually load a list of drift for images until the drift stops.  Each element is the  drift calculated
 as the absolute value of the difference between where the drift starts and stops, IN PIXELS,
as it appears in Casa.  Drifting occurs from right to left over time, as it appears in Casa.*)
StageCorrect = 256 - (StageDrift);
(*Adjust to reflect columns to retain*)
ResizeAllStageCorrect = PadRight[PadRight[StageCorrect, 79, 245], 107, 256];
(*Pads the list to the size of the image set if the image set is bigger than the list of drifting
 images.  Added values are 256 because no pixels need be cropped*)
PadCropDataset =
 ImageData[ImageCrop[(*ImageData is absolutely essential or else the image will not go back into a Casa readable file*)
     ImageCrop[
      Image[MoDataset[[#]], "Byte"], (*Defines the image used*)
      {ResizeAllStageCorrect[[#]], 256}, Left], (*Corrects for the drift by deleting left columns of pixels *)
     {Min[ResizeAllStageCorrect], 256}, Right],
    "Byte"(*Crops all images from the right to obtain a constant size based on the maximum drift observed*)
   ] & /@ Range[1, Length[ResizeAllStageCorrect]];
(*Defines number of processes to be the number of the images defined in the stage drift file*)
(*-----------------------------------------------------------------------------------------------*)

(*This section is used for batch exporting  image files to a folder that have already been processed.  Currently
 set for the above mass cropping script*)
SetDirectory["C:\\Users\\..."]; (*sets the directory the files are extracted from and saved to*)
FirstValues = Map[ReadList[OpenRead[#], String, 39] &, FileNames["*.vms"]];
(*extracts the first 39 values from each image in the selected directory including all energy values*)
Spatialvalues = ReadList[OpenRead["Spatial values.txt"], String, 39];
(*separate file including second 39 values from each image in the selected directory including all
 spatial values.  NOTE:  there are 6 values that are spatial that need to be corrected for each dataset*)
Export["CroppedVacuumImage" <> ToString[#] <> ".txt",
   Join[FirstValues[[#]], Spatialvalues, Flatten[PadCropDataset[[#]]]]] & /@ Range[1, Length[ResizeAllStageCorrect]]
(*Exports all Cropped XPS images as .txt files with their front 78 lines of metadata*)
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Figure S13. Mathematica Script of the script employed in *i*-XPS to correct for image drift.

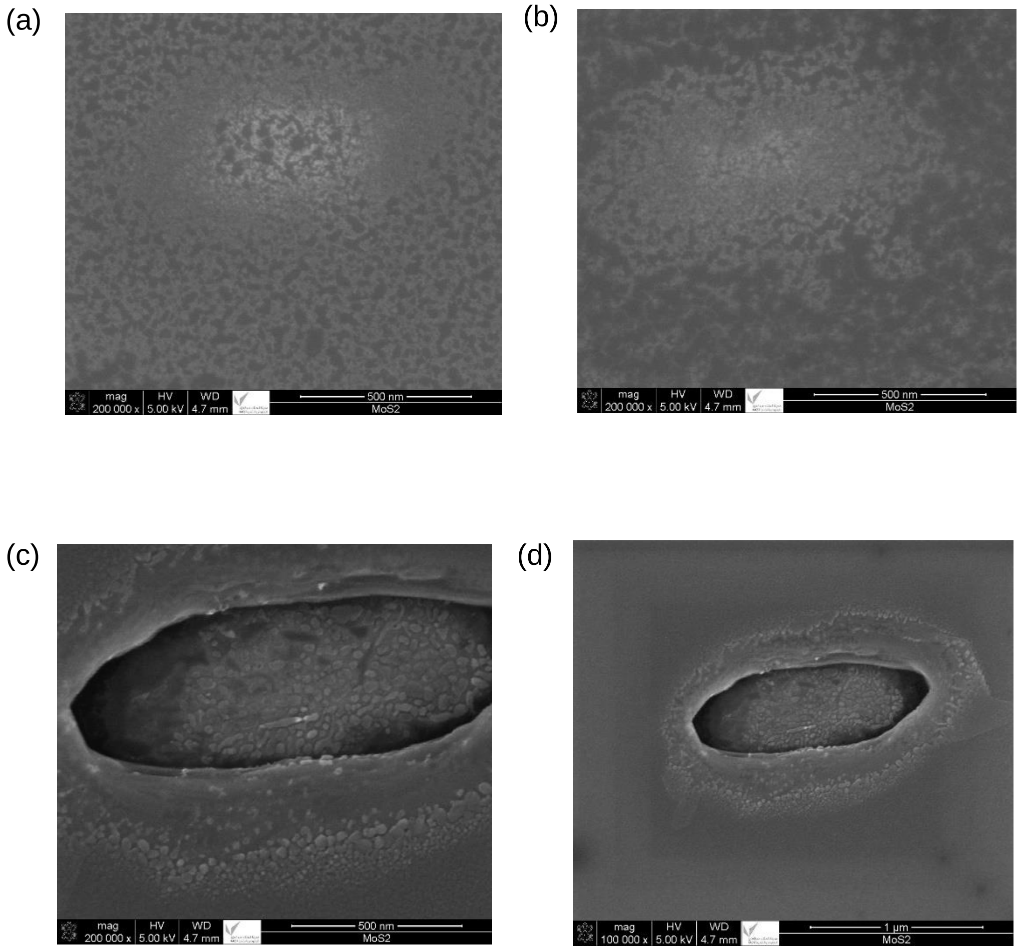

Figure S14. SEM images of laser-treated MoS_2 multilayer nanosheet. (a) and (b) SEM images showing particles formed on the surface of the treated spots. Laser treatments were carried out with 0.93 mW of 532-nm laser for 60 s. (c) and (d) SEM images showing laser thinning and the formation of anomalous particles around the edges. These particles vary in size and shape. The elliptical shape of the spot is due to stage drift in the setup. Laser treatments were carried out using 8.3 mW of 532-nm laser for 5 s. For all images, treatments were taken using 100× objective lens.

Table S1. Atomic percentages attributed to each chemical or elemental species for different masks or ROIs (underlined in the case of multiple elements).

	Mask/ROI ID	$\underline{Mo}_{ox}$	$\underline{Mo}_{sulfide}$	$\underline{O}_{Mo}$	$\underline{O}_{Si}$	$S_{sulfide}$	Si
Ambient	MoS_2 laser	8	0	16	51	1	24
	MoS_2 native	0	4	0	58	8	30
	SiO_2	0	0	0	63	0	37
Vacuum	MoS_2 laser	1	7	2	50	12	28
	MoS_2 native	0	4	1	56	7	33
	SiO_2	0	0	1	63	0	36

The results presented in Table S1 are presented without any error and are intended to be informational and estimates only. This is a result of one measurement, which precludes any type A error associated with multiple measurements in attempts to assess the heterogeneity across one specimen or between multiple specimens. With respect to type B uncertainty, there is insufficient information and calibration checks for all factors to accurately assign an uncertainty. Some of the factors not accounted for include knowledge of the variability in the intensity as a function of position on the detector, which is important due to shifts in the image region as a function of stage drift as well as energy dependent shifts. Attenuation in photoelectron intensity from the substrate was also not accounted for which may have resulted in a suboptimal rsf assignment and would be another source of error. Therefore, since the only error we have access to is an idea of the uncertainty associated with the fit, which would grossly underestimate the true error, we chose to only use these values as rough estimates with no assignment of error.