

Supporting Information to: Reductive exfoliation of substoichiometric MoS₂ bilayers using hydrazine salts

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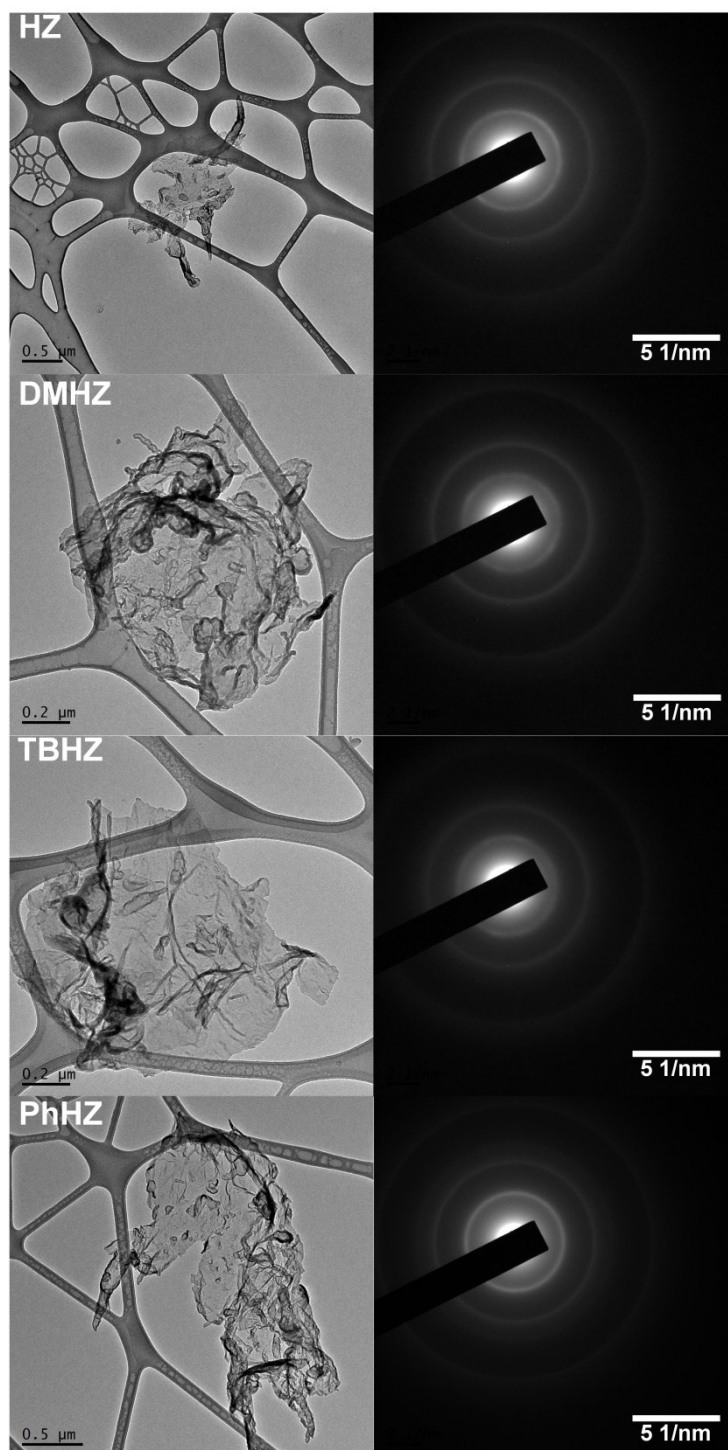


Figure SI-1: TEM images and selected area electron diffraction (SAED) patterns of 2D nanosheets exfoliated with different hydrazine derivatives as indicated in the TEM images. The diffraction patterns feature less detail as the one in the main manuscript since the Jeol 1010 was used for this images which provides less resolution. The image in the main manuscript was collected using the 2100F instrument. The lattice spacings were identical in all cases.

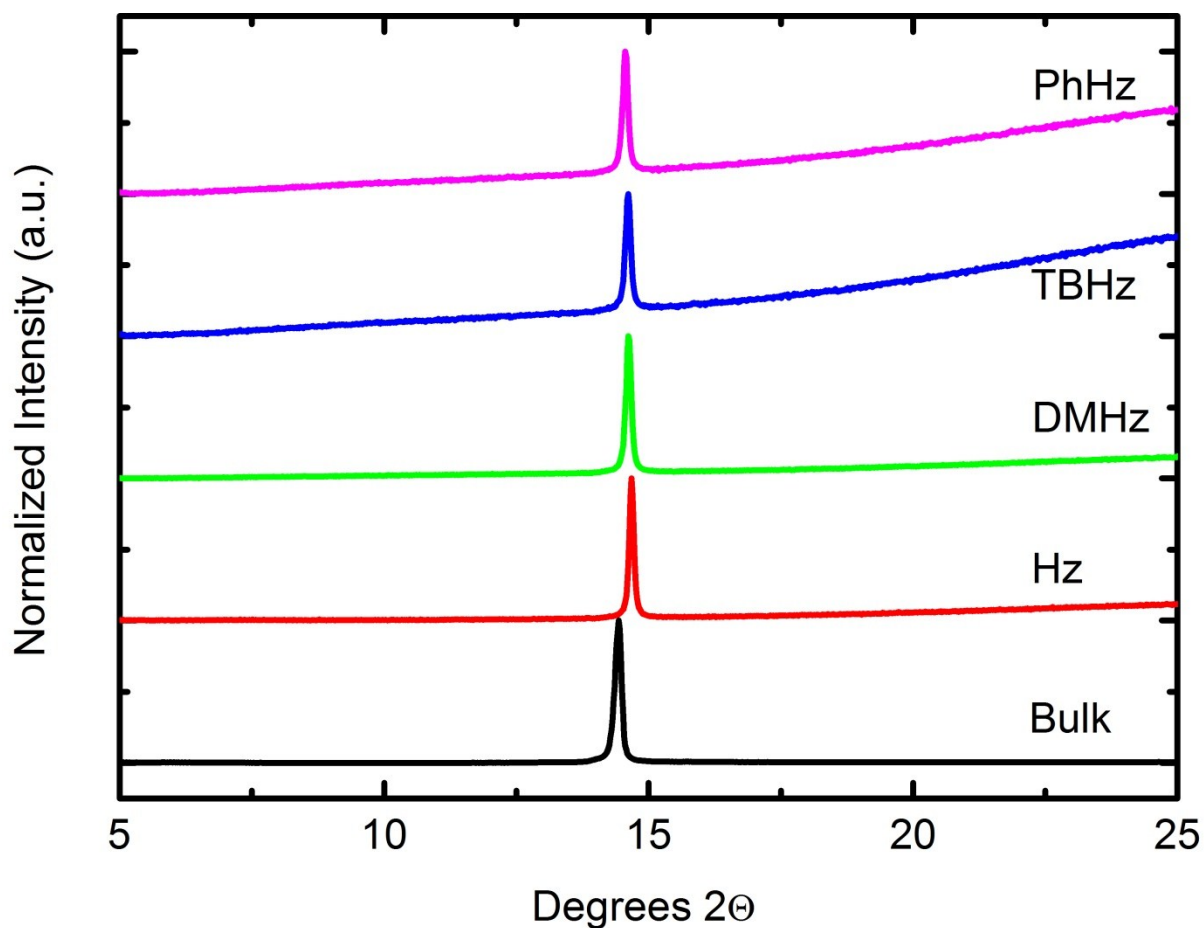


Figure SI-2: XRD patterns of residual bulk particles which failed to fully exfoliate during the 1 hr reaction time. The black line labelled as bulk refers to the pristine unreacted starting material. The samples were washed as described in the main paper and drop casted onto glass slides. Samples not washed featured identical peak positions for the main peak (data not shown). The broad peak visible at higher angles in TBHz and PhHz is due to the glass substrate. The small variation in the main peak position might be attributed to variations in sample height with reference to the detector and are within the error of the measurement.

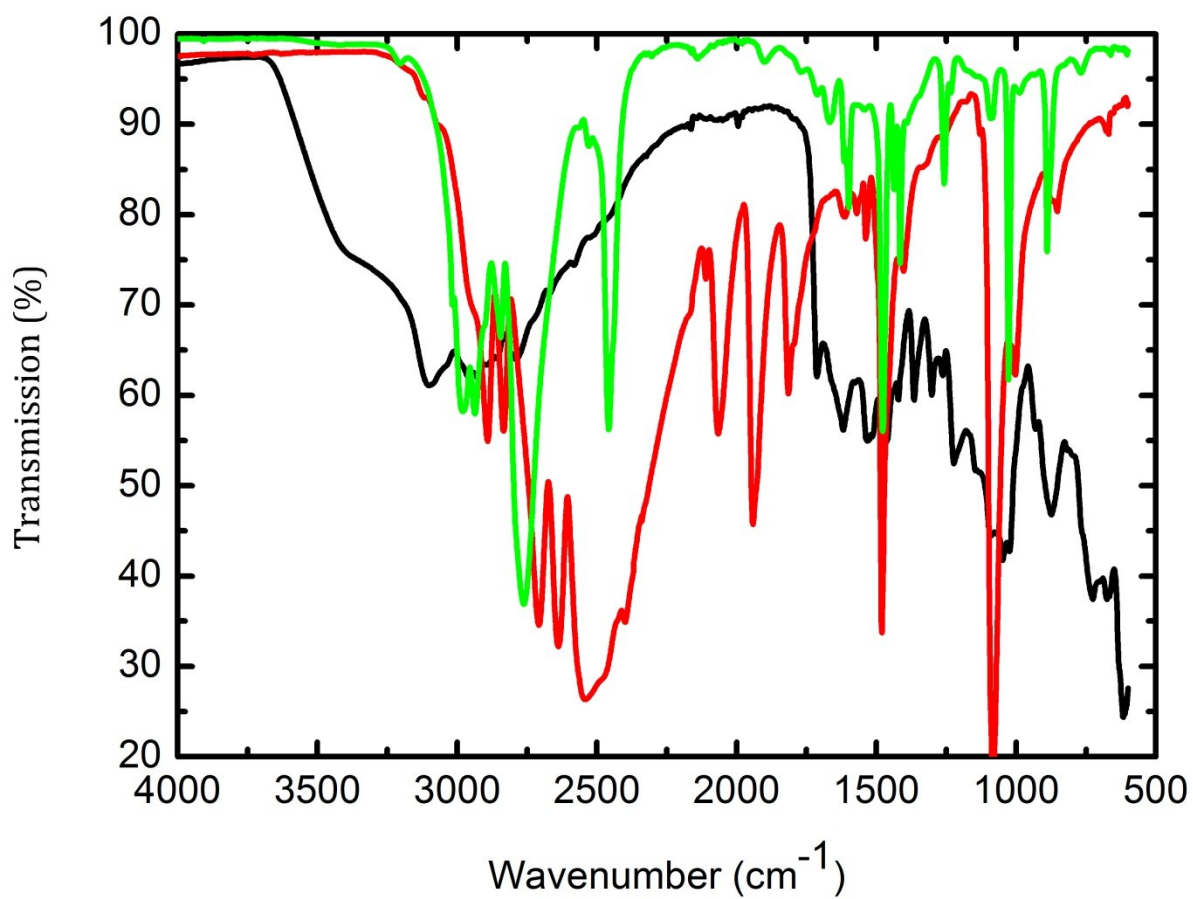


Figure SI-3: Fourier-transform IR (FTIR) spectroscopy data for hydrazine hydrochloride (red) and the side products from the Hz reaction, white crystals (green) and the isolated brown oil (black).

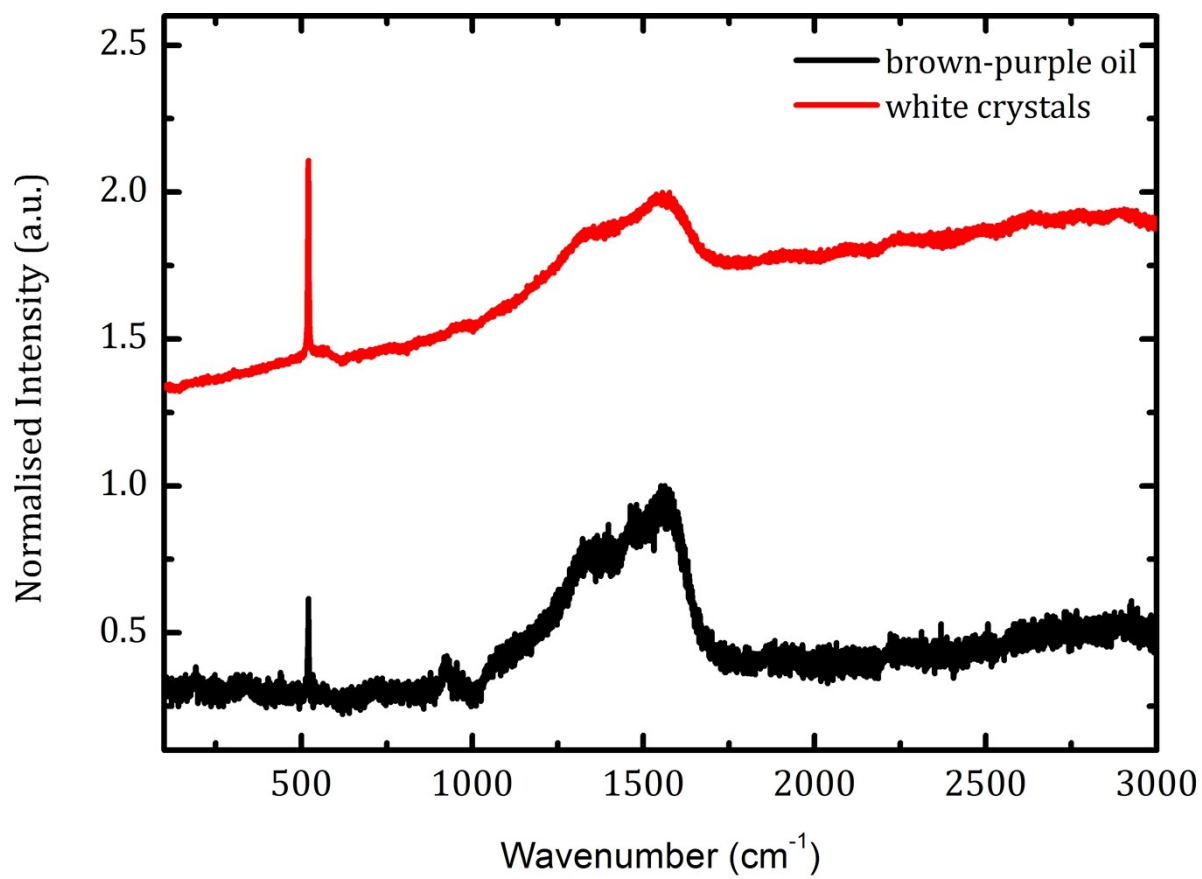


Figure SI-4: Raman spectra for the side products from the Hz reaction, white crystals (red) and the isolated purple-brown oil (black).

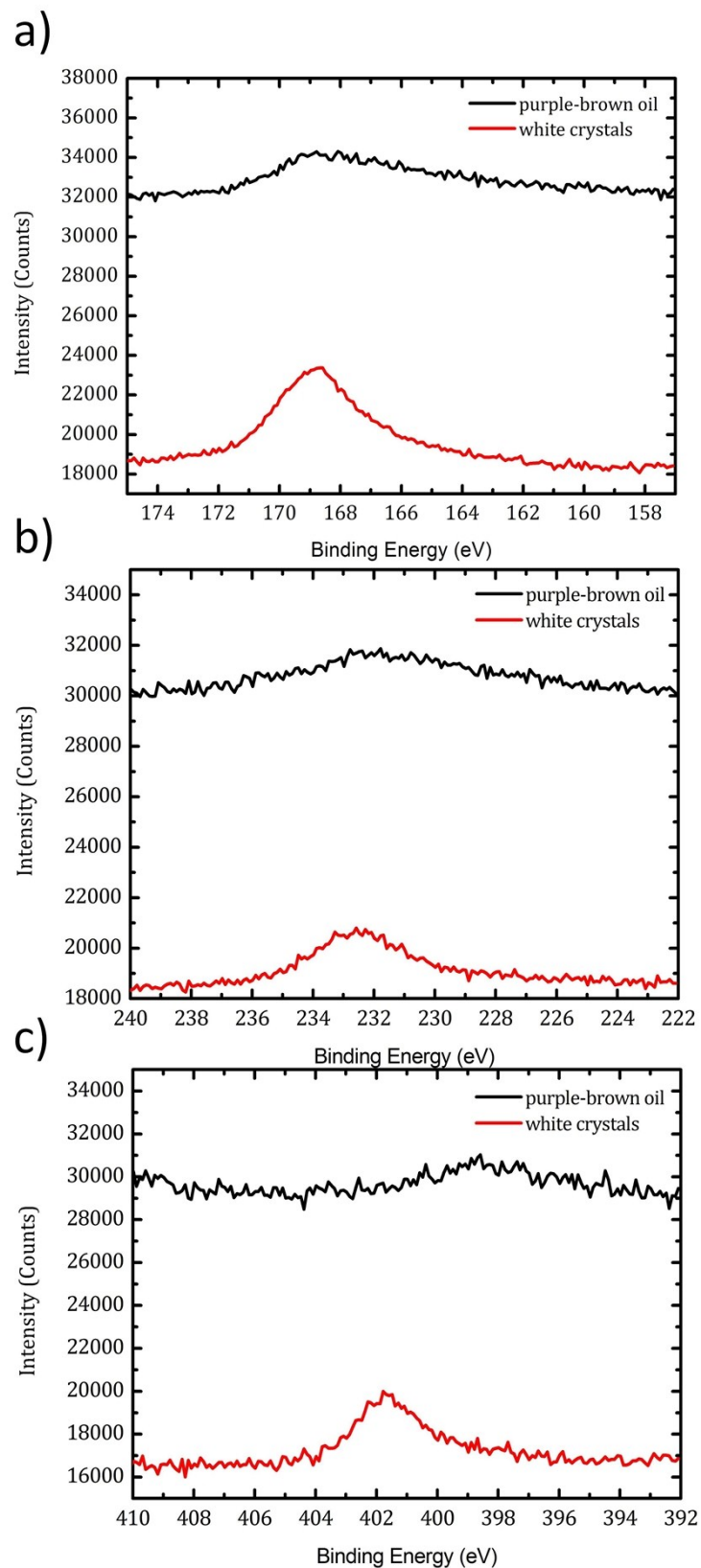


Figure SI-5: X-ray photoelectron spectroscopy (XPS) for the side products from the Hz reaction, white crystals (red) and the isolated purple-brown oil (black) for a) the sulphur region, b) the molybdenum region and c) the nitrogen region.

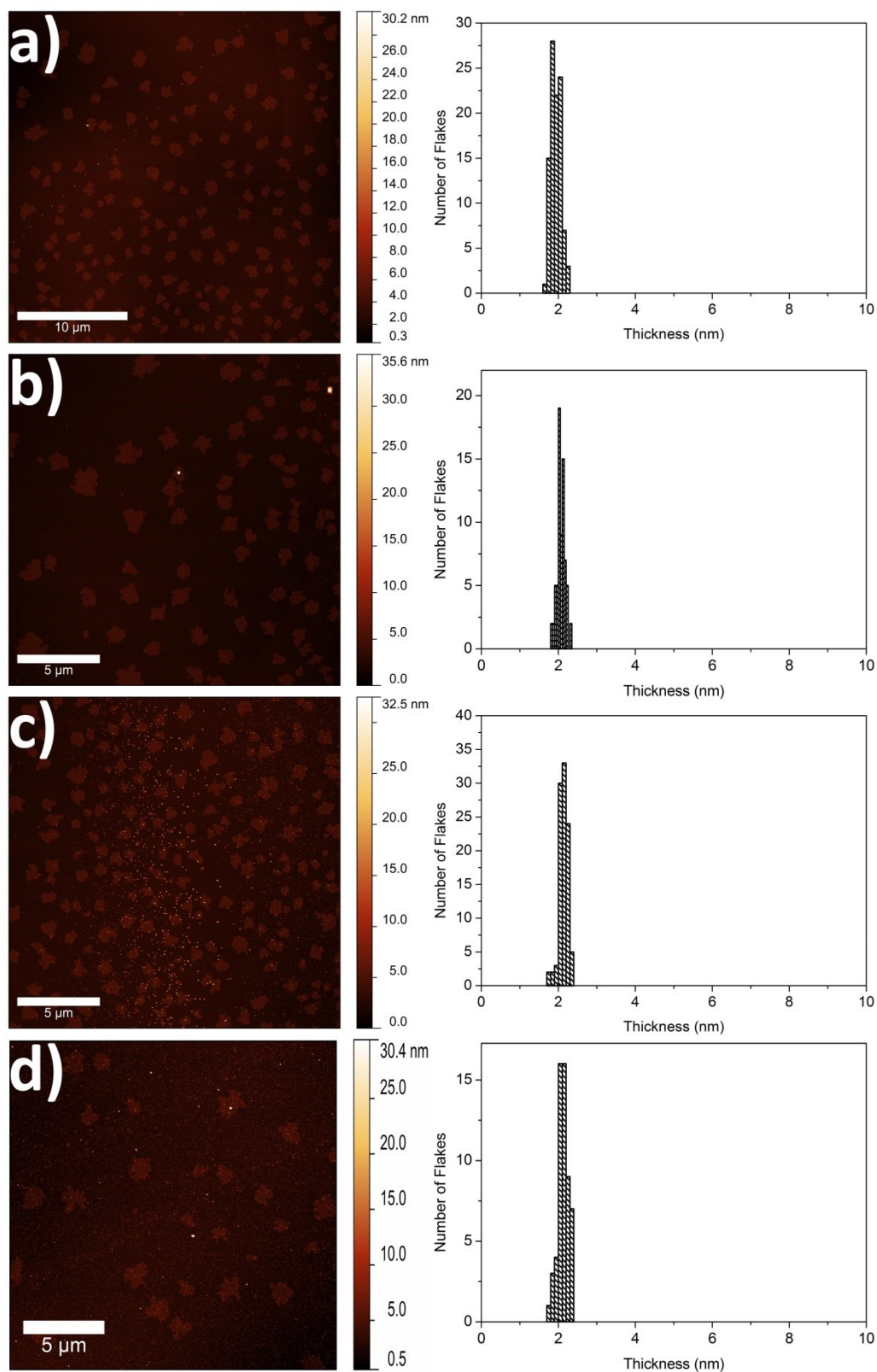


Figure SI-6: AFM Images and thickness histograms. a) HZ, b) DMHZ, c) TBHZ and d) PhHZ. Between 50 and 100 flakes were measured for each histogram. The average thicknesses are HZ: 1.9 ± 0.2 nm; DMHZ: 2.0 ± 0.10 nm; TBHZ: 2.09 ± 0.11 nm; PhHZ: 2.1 ± 0.15 nm

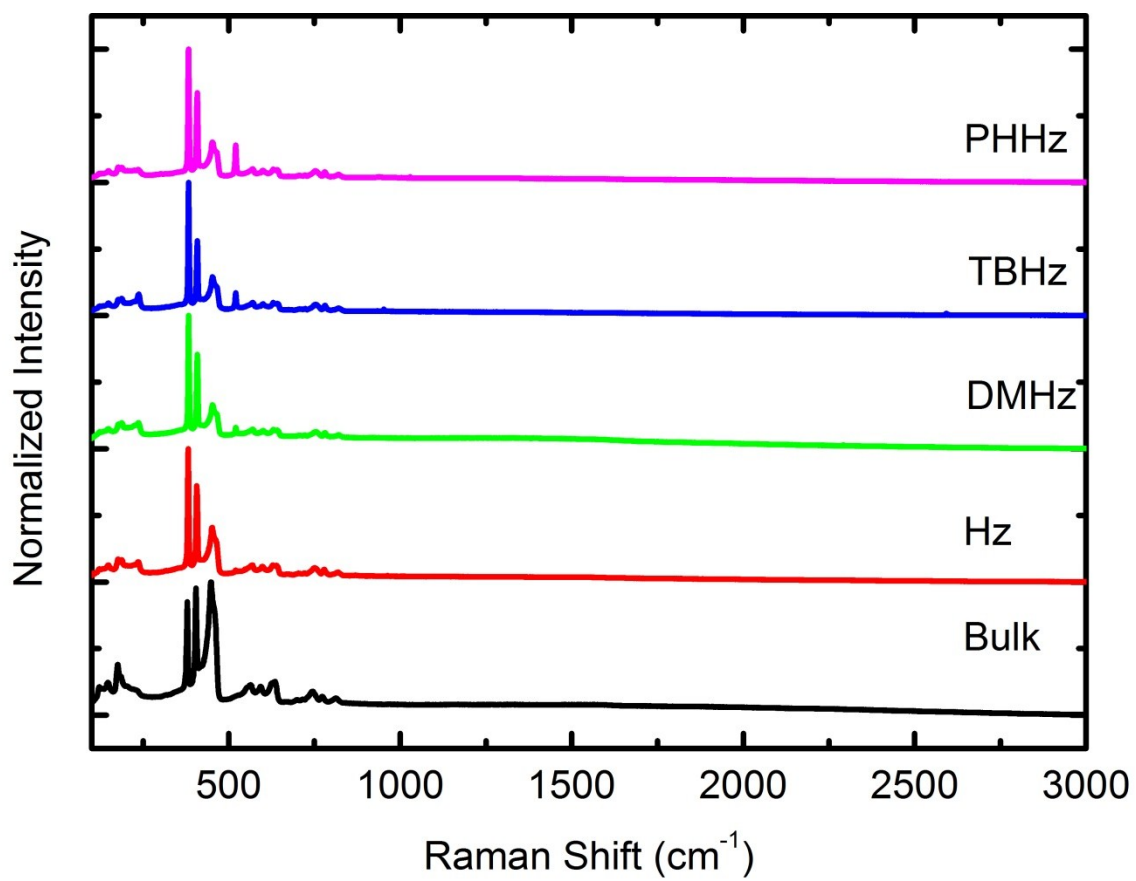


Figure SI-7: Extended Raman spectra.

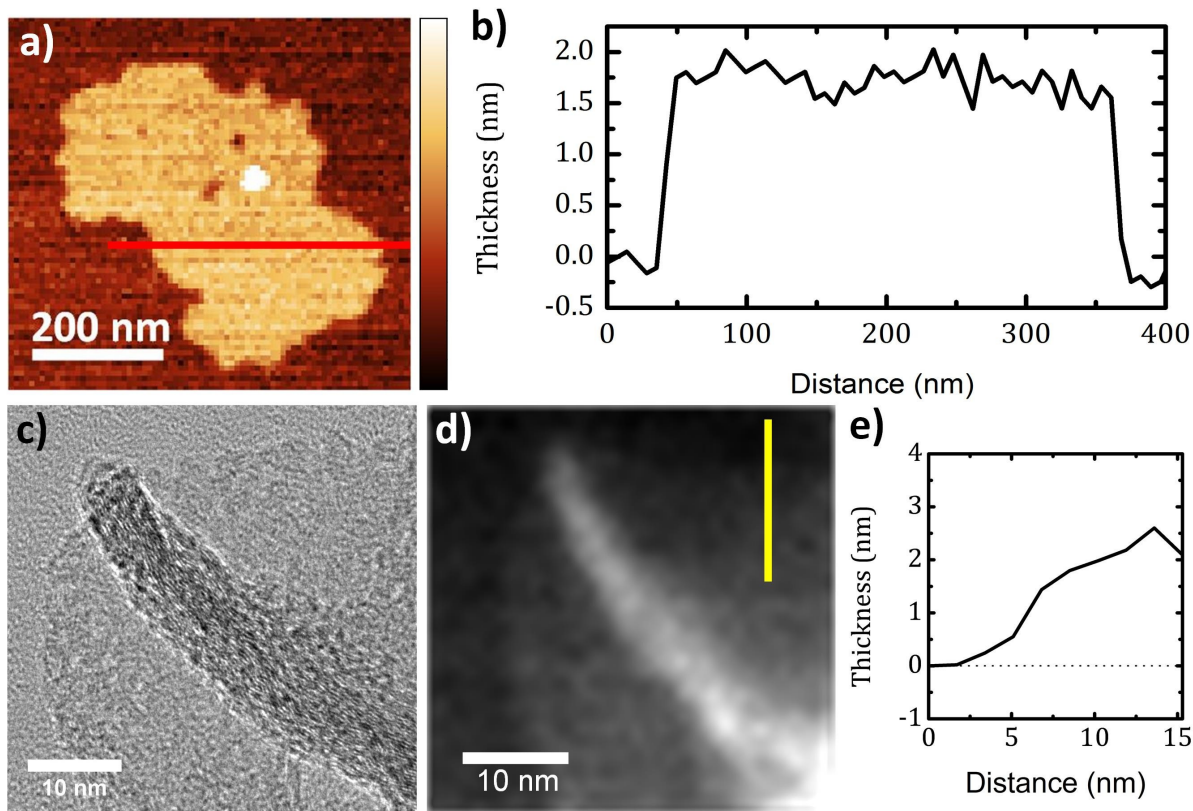


Figure SI-8: a) High resolution AFM image of a small $\text{MoS}_{1.2}$ sheet with the thickness scale ranging from 0 nm (black) to 5 nm (white) and the red line indicating the position of the thickness profile shown in b). c) High resolution transmission electron microscopy (HRTEM) image of the region that was analysed using electron energy loss spectroscopy (EELS), d) EELS map with the yellow line indicating the region investigated for the thickness profile shown in e)

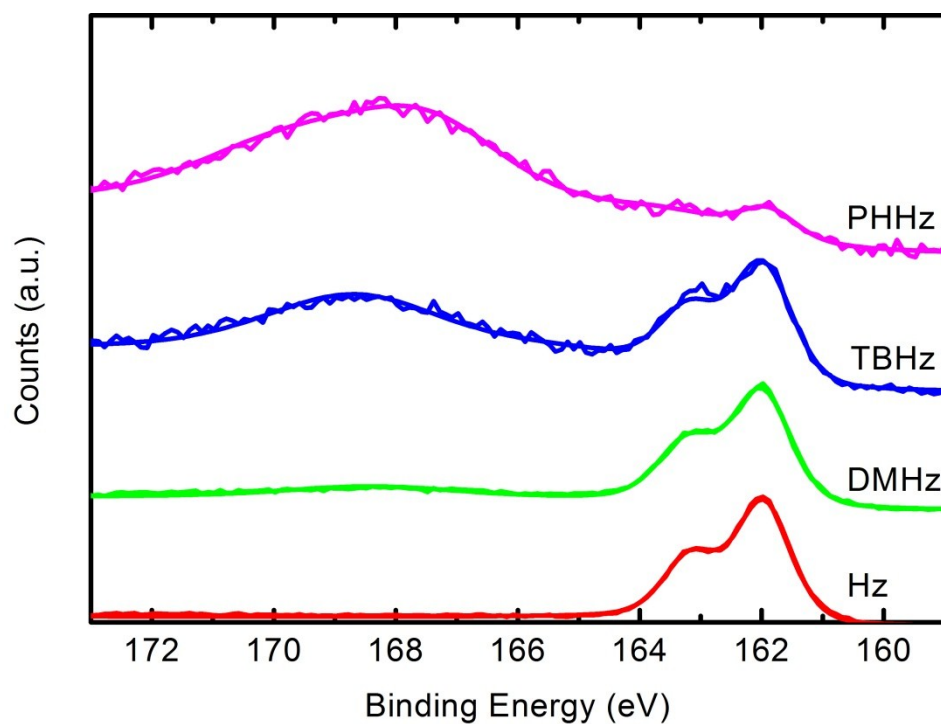


Figure SI-9: Extended S p2 XPS spectra. The broad peak at ~168 eV is associated with metal sulphates.

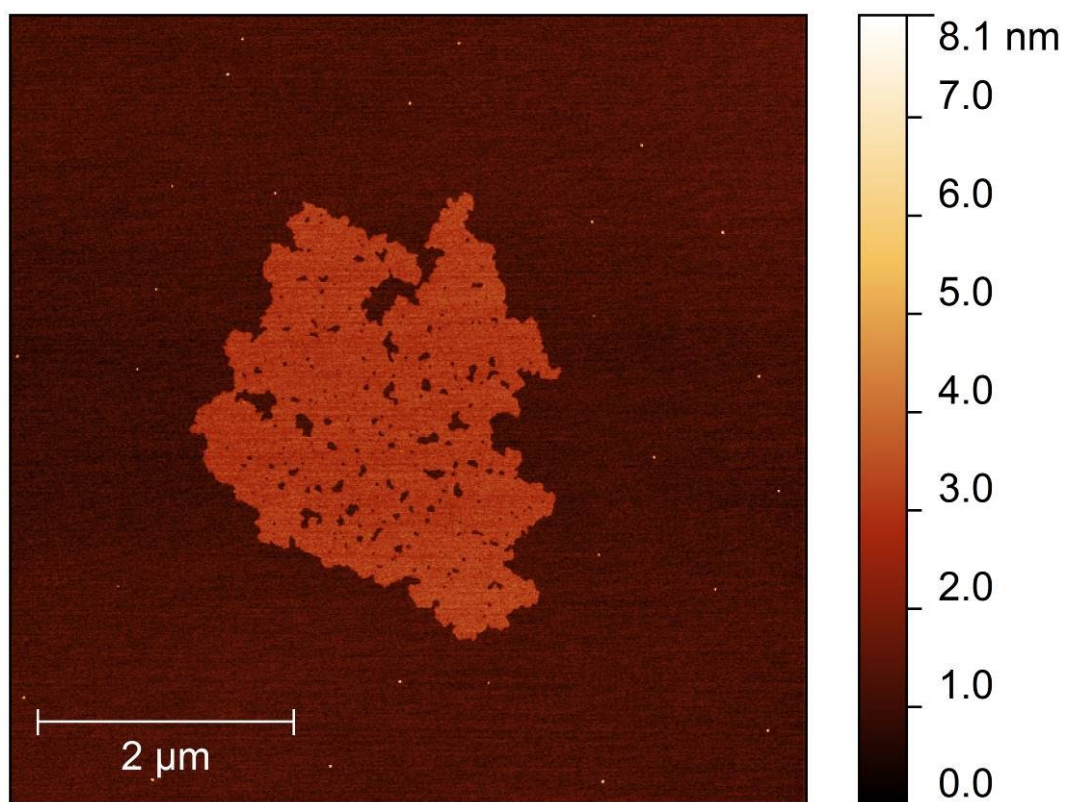


Figure SI-10: AFM image of single flake after 7 days of storage in DMF.

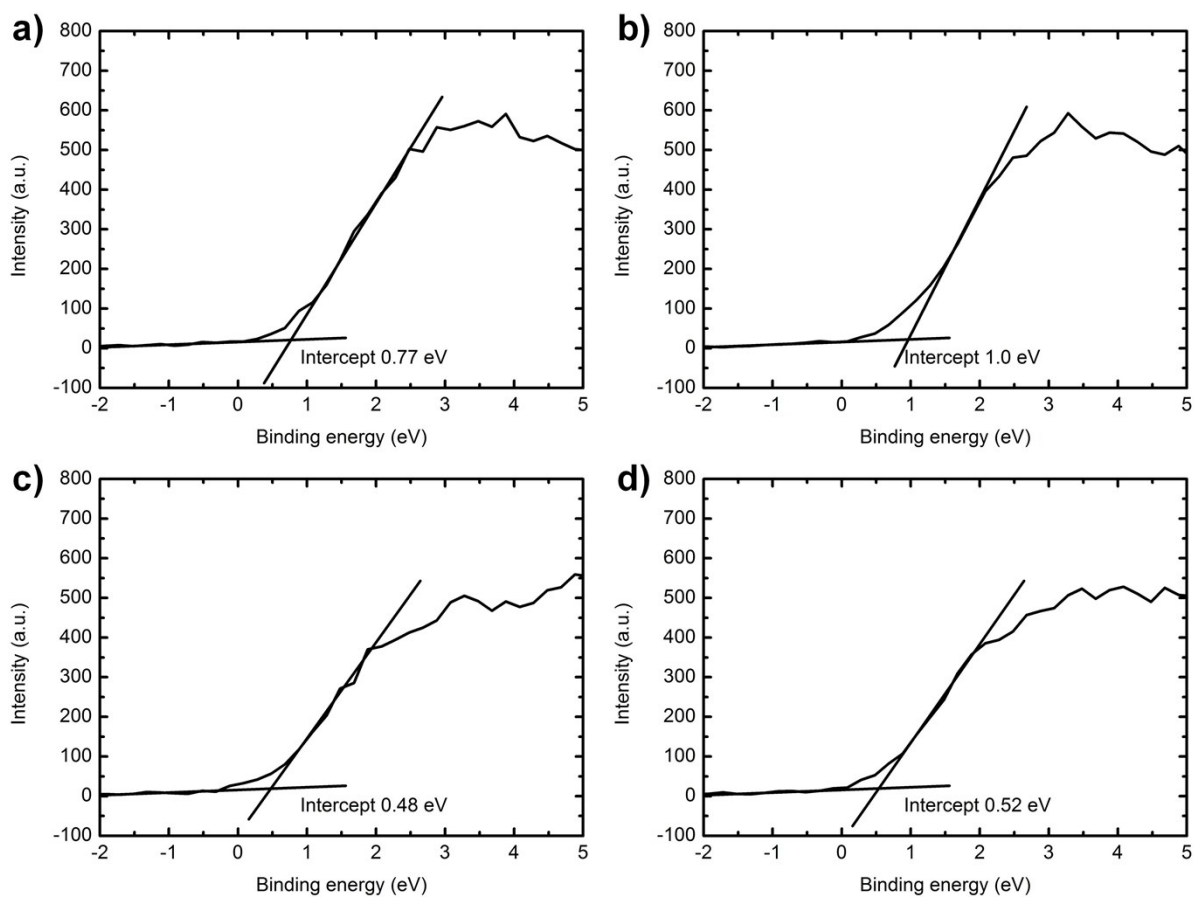


Figure SI-11: XPS valence band spectra for MoS_x samples exfoliated with a) HZ, b) DMHZ, c) TBHZ and d) PhHZ.

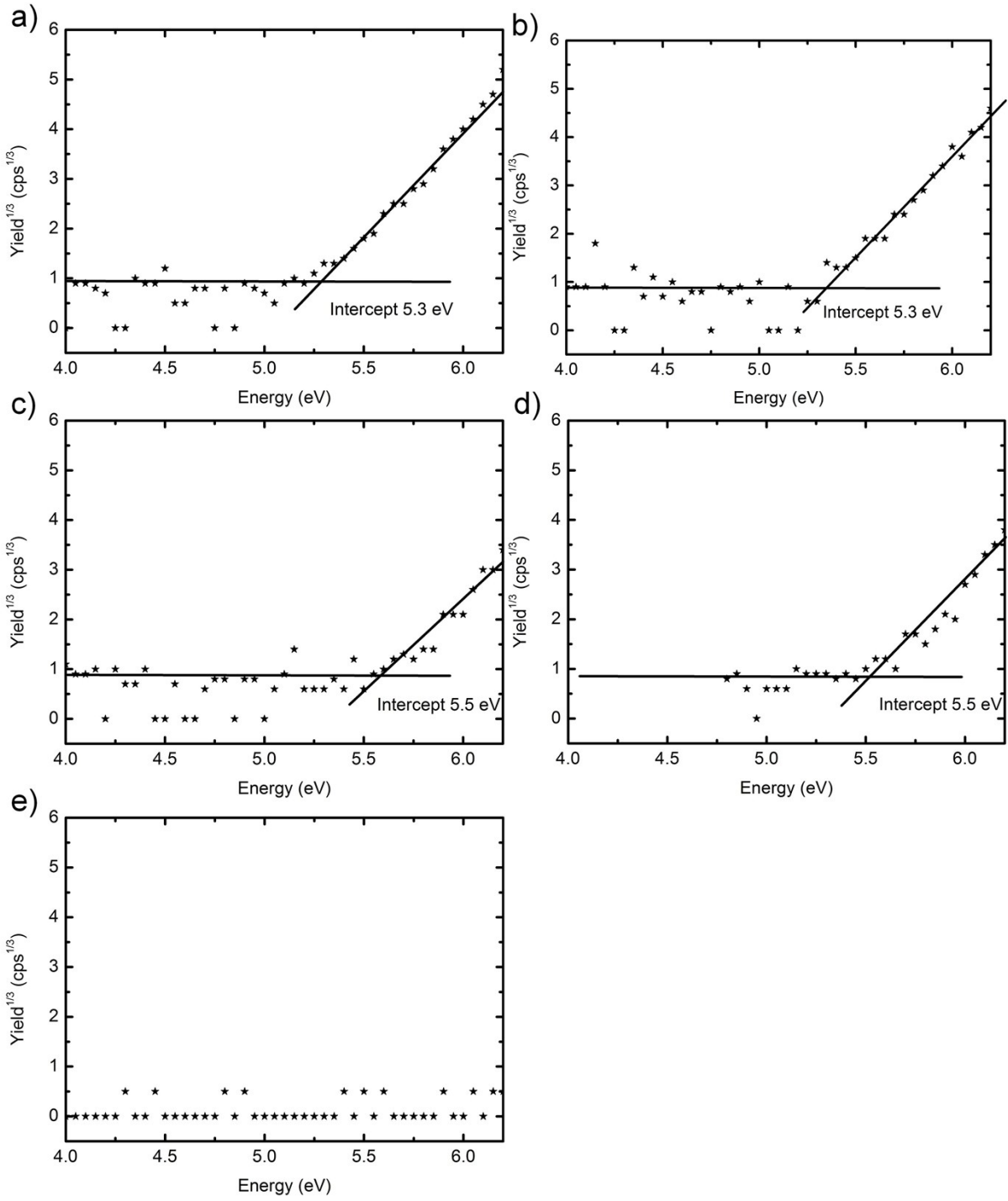


Figure SI-12: Photoelectron spectroscopy in air (PESA) measured for samples exfoliated with HZ (a), DMHZ (b), TBHZ (c), PhHZ (d) and for a blank glass slide (e).

Table SI-1: Results of a biexponential fit to the data shown in Figure 3. The equation used for the fit is $y = y_0 + A_1 \times [1 - e^{(-x/\tau_1)}] + A_2 \times [1 - e^{(-x/\tau_2)}]$

Parameter	Y_0	A_1	τ_1 (ns)	A_2	τ_2 (ns)	R^2
HZ	24588	-22257	0.62	-2238	3.35	0.9986
DMHZ	30343	-28790	0.43	-1492	2.38	0.9978
TBHZ	18449	-15918	0.93	-2555	4.60	0.9974
PhHZ	38757	-38001	0.52	-756	2.68	0.9983