Electronic Supplementary Information

Colloidal 2D $Mo_{1-x}W_xS_2$ Nanosheets: An atomic- to ensemble-level spectroscopic study

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Table S1) Stoichiometry and amount of precursor to obtain $Mo_{1-x}W_xS_2$ NS alloys of targeted composition.

x in $Mo_{1-x}W_xS_2$	$MoCl_5$ (µmol, mg, eq.)	WCl_6 (µmol, mg, eq.)	S (mmol, mg, eq.)
0	18.5, 5, 1	0	62.25, 8, 3.4
0.25	13.88, 3.8, 0.75	4.63, 1.8, 0.25	62.25, 8, 3.4
0.5	9.25, 2.5, 0.5	9.25, 3.6, 0.5	62.25, 8, 3.4
0.75	4.63, 1.3, 0.25	13.88, 5.5, 0.75	62.25, 8, 3.4
1	0	18.5, 7.3, 1	62.25, 8, 3.4

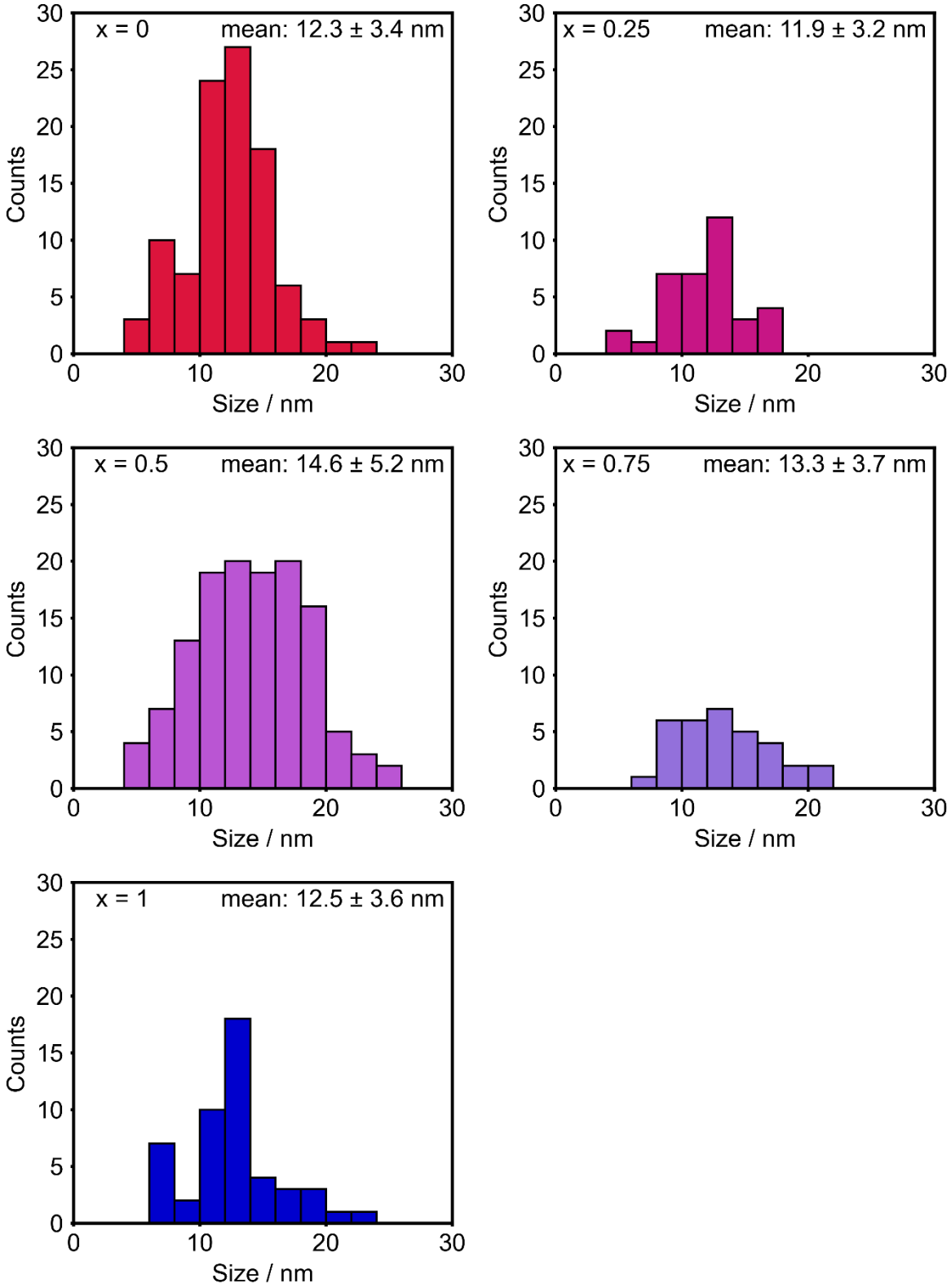

Fig. S1: Size distribution for $Mo_{1-x}W_xS_2$ NSs as observed in TEM images. Sizes are distributed between 5 and 25 nm and centered around 12 to 14 nm.

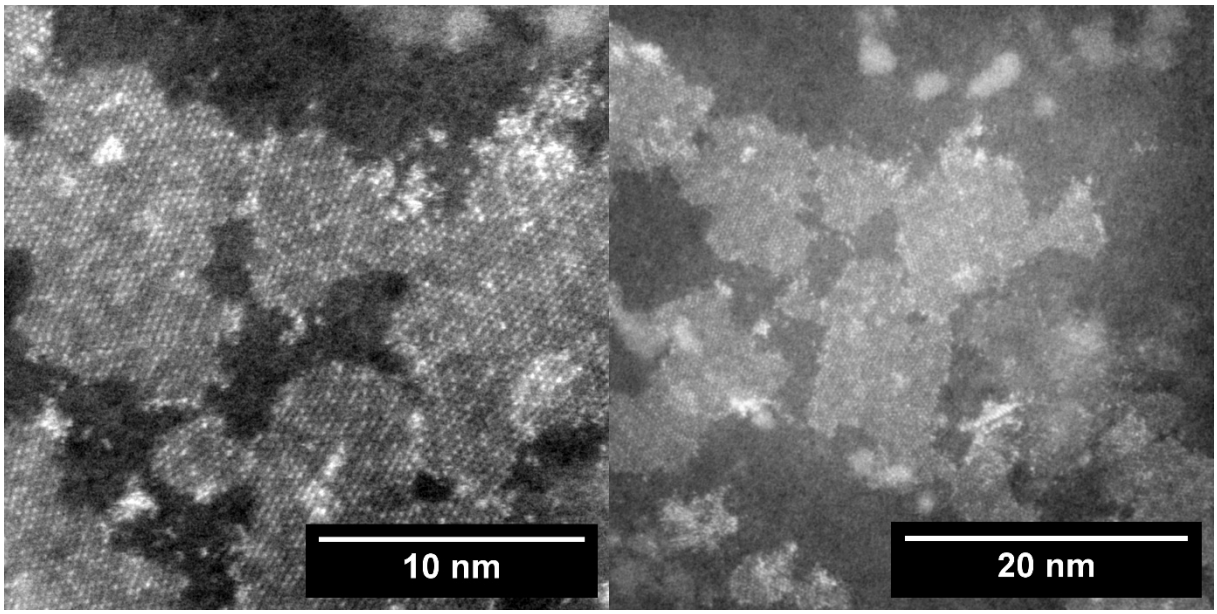

Fig. S2: Examples of flat-lying monolayers of colloidal $Mo_{1-x}W_xS_2$ NSs covering the graphene surface of the TEM grid.

Raman spectroscopy peak fitting

Fig. S2 shows the steps taken to process and subsequently fit the raw Raman data for the example of the $Mo_{0.25}W_{0.75}S_2$ alloy (Fig. 3c (*right*) of the main manuscript). First, the area of interest of the Raman spectra are normalized and smoothed. Then the background, manually fitted as a third-order polynomial, is subtracted. Symmetric, non-skewed, Gaussian functions were used to fit the individual Raman modes E', E^1_{2g}, and A_{1g}.

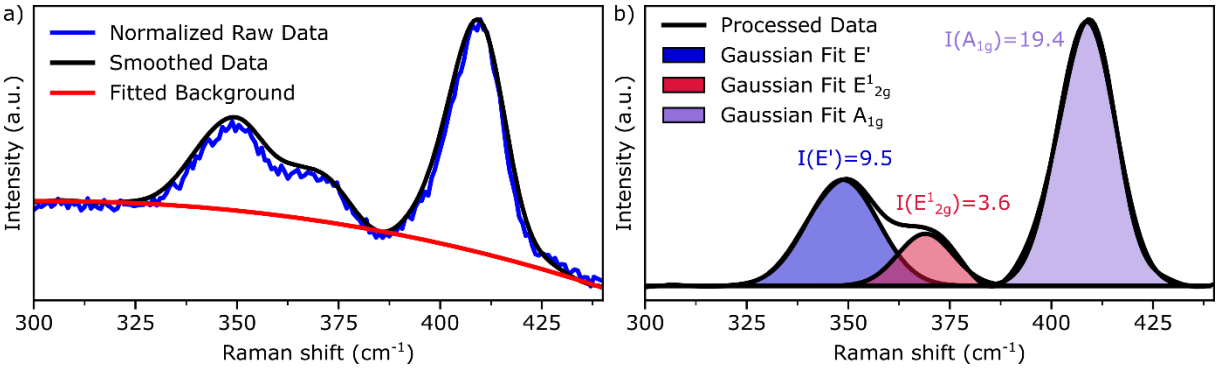

Fig. S3: a) Smoothing and background fitting of the raw Raman data, as illustrated for the example of the spectrum of the $Mo_{0.25}W_{0.75}S_2$ alloy. b) The final, processed data is fitted using three symmetric, non-skewed, Gaussian functions. The intensities of the individual peaks are indicated.

The intensity ratio $I(E^1_{2g})$ / I(E'), which can be used as an indication for the composition of the alloy, is influenced by the specific smoothing, background correction and fitting parameters, limiting the numerical robustness of this method. In this specific case of parameters, the exact intensity ratio $I(E^1_{2g})$ / I(E') of the $Mo_{0.25}W_{0.75}S_2$ alloy is calculated to be 0.38. To account for the lack of robustness of this method, the approximate value for the alloy composition (~ 1/3) is displayed in the main manuscript.

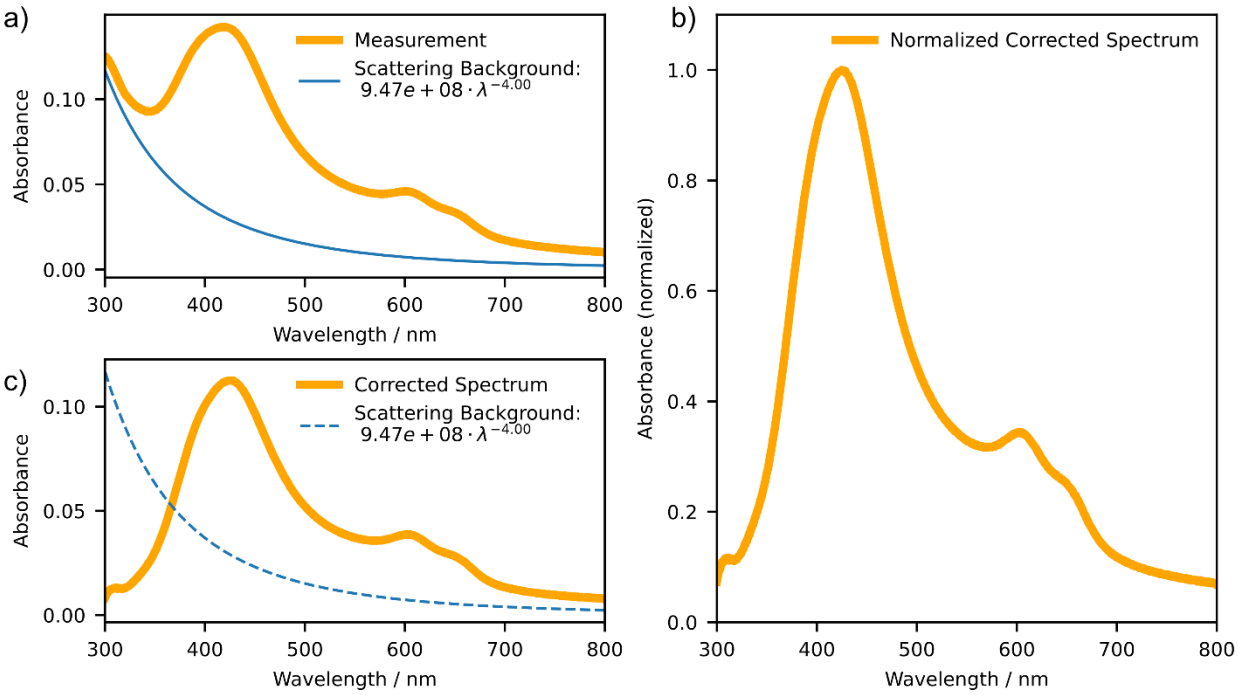

Fig. S4: a) Absorbance of MoS_2 NSs plotted against the wavelength (λ) with scattering background (Rayleigh scattering) proportional to λ^{-4}. b) Corrected absorbance spectrum. c) Corrected spectrum normalized.