

Supplementary Information

Quantification of Defect Engineering in Single Layer MoS₂

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Wet Etch Transfer of Monolayer Molybdenum Disulfide (MoS₂) onto TEM grid

PMMA (950K A4) was spin coated on MoS₂ monolayer on the Si/SiO₂ in a two-step cycle. The first step was run for 15 secs at 400 rpm followed by second step that was run for 45 secs at 1500-2000 RPM. PMMA coated film was then heated at 180°C for 15 min on a hot plate to completely remove residual solvent. Then, PMMA/MoS₂/SiO₂/Si stack was immersed in 1M of KOH. After 5-10 hours, PMMA/MoS₂ stack detached from the Si/SiO₂ and started to float in KOH [1]. PMMA/MoS₂ stack was then picked up on a Quanta Foil TEM grid and then heated on a hot plate at 100°C for 15 mins to remove any remaining solvent. PMMA was lastly, removed using acetone.

1. A. Gurarslan, Y. Yu, L. Su, Y. Yu, F. Suarez, Shanshan Yao, Y. Zhu, M. Ozturk, Y. Zhang, and L. Cao, Surface-Energy-Assisted Perfect Transfer of Centimeter-Scale Monolayer and Few-Layer MoS₂ Films onto Arbitrary Substrates. ACS Nano 2014 8 (11), 11522-11528 DOI: 10.1021/nn5057673

Calculation of Average of Inter-Defect Distance in 1L MoS₂.

Helium Ion Microscope was used to controllably introduce defects in 1L MoS₂ thereby creating atomic vacancies (1Mo + yS). Helium ions provide gentle etching extracting multiple sulfur atoms with only one molybdenum atom. The predominant defects present were single molybdenum-based vacancies (1Mo + yS) while the number of missing sulfur atoms differed. The exposure dose was in a range of 1E14 to 1E16. L_D value was calculated manually from Scanning Transmission Electron Microscopy (STEM) maps as it shown on Figure 1S for the dose 1E14. Results of calculation of mean L_D value with corresponding standard deviation was populated in a Table 1S.

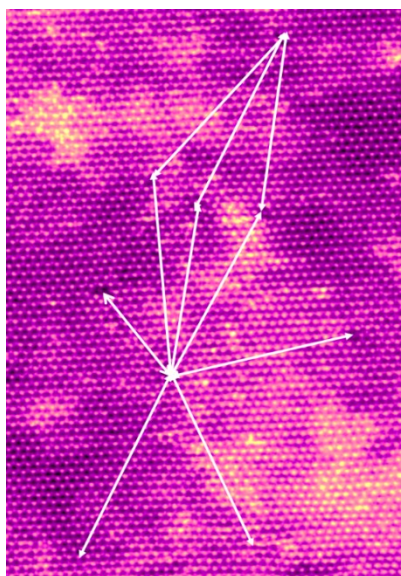


Figure 1S: STEM image of suspended molybdenum disulfide after helium ion irradiation. White arrows show distance between absent Mo sites

Table 1S. Calculation of mean LD value from STEM for respective helium ion dose

Helium Ion dose (Ions/cm ²)	1E14	3E14	3.75E14	4E14	5E14	1E15	5E15	1E16
L _D , nm (from STEM)	10.3±1.1	9.5±1.9	8.6±1.6	7.1±1.9	7.5±2.0	5.4±1.0	4.1±1.0	3.7±0.4

Raman Studies of Irradiated MoS₂ on TEM grid

The in-plane E_{2g}/out-of-plane A_{1g} vibrational modes after irradiation are red/blue shifted, and both are widened. There is a shoulder (at around 362 cm⁻¹) on the left of E_{2g} peak and one (around 415 cm⁻¹) to the right of A_{1g} peak, which are assigned as defect modes.

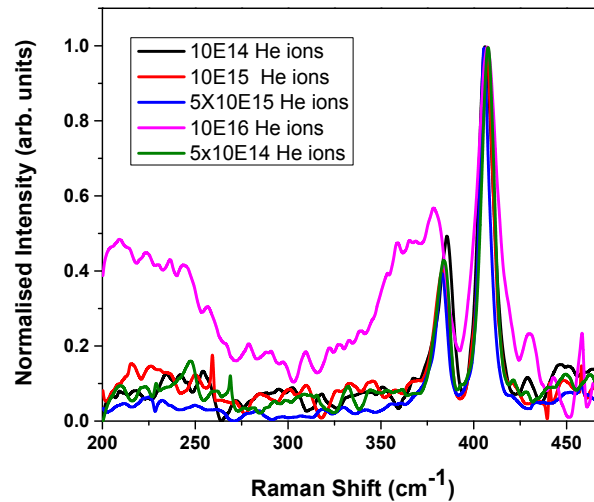


Figure 2S: Raman studies of Irradiated suspended 1L MoS₂; the first order Raman modes (E_{2g} / A_{1g}) are red/blue shifted and the second order mode LA (M) arises upon dosage increase.

AFM Characterization

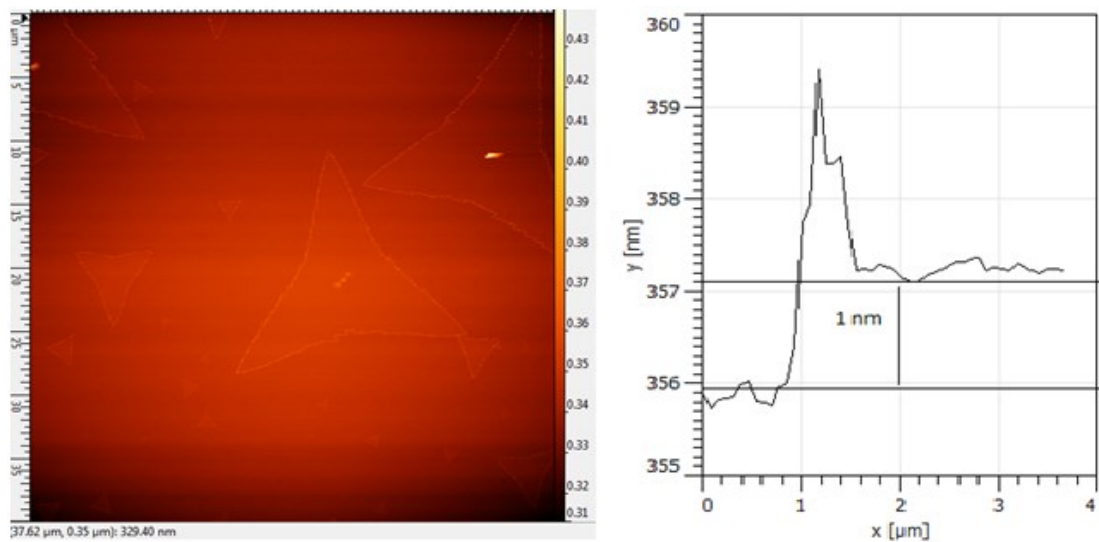


Figure 3S: AFM topography of monolayer of molybdenum disulfide