Electronic Supplementary Information

Bandgap Recovery of Monolayer MoS_2 using Defect Engineering and Chemical Doping

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MoS_2 characterization and analysis

Photoluminescence (PL) and Raman spectra were recorded with a Horiba XploRa Confocal Raman Microscope at 532 nm of excitation. The two prominent Raman peaks located around 402 cm^{-1} and 383 cm^{-1} assigned to the out-of-plane (A_{1g}) and in-plane (E_{2g}) phonon vibration modes respectively (Figure S1c). The exact position of these peaks can be found by fitting with the Lorentz function. The band separation df = 19-20 cm^{-1} corresponds to 1L MoS_2. With the increasing number of layers, the difference df will also increase, for instance, df = 21-22 cm^{-1} corresponds to bilayer of MoS_2. Raman mapping show spatial homogeneity of E_{2g} (Figure S1a) and A_{1g} (Figure S1b) over the MoS_2 flake area: the intensity and variation for both peaks were negligible, we have identified as grown flakes as pristine monolayers.

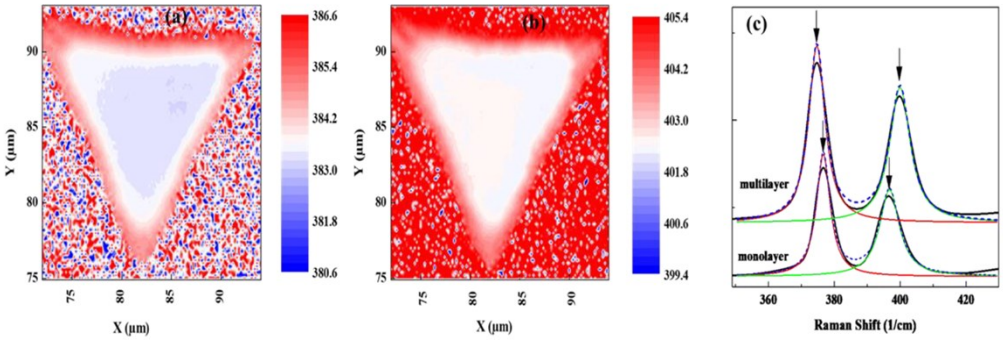

Figure S1 Raman intensity map: (a) E_{2g} at 383 cm^{-1}, and (b) A_{1g} at 402 cm^{-1}; (c) Raman spectrum from the center of MoS_2 flake.

MoS_2 samples were irradiated in a Zeiss Helium Ion Microscope operating at accelerating voltage of 30kV with doses 10E13 – 10E16 He^+ ions / cm^2. For defected samples there are two shoulders: (around 362 cm^{-1}) on the left of E_{2g} mode and one (around 415 cm^{-1}) to the right of A_{1g} mode, which are assigned as defect modes. Both peaks were significantly enhanced upon increase of irradiation dose, confirming the introduction of defects (Figure S2).

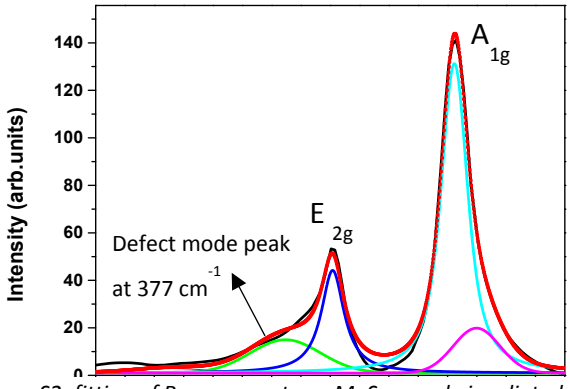

Figure S2: fitting of Raman spectrum MoS_2 sample irradiated with 10E14 dose.

The MoS_2 PL spectra contains three contributions: B-exciton at 1.96 eV, A-exciton at 1.82 eV, and trion (X^-) at 1.76 eV. We used Gauss function to extract position and intensity of trion peak as it shown on Figure S3.

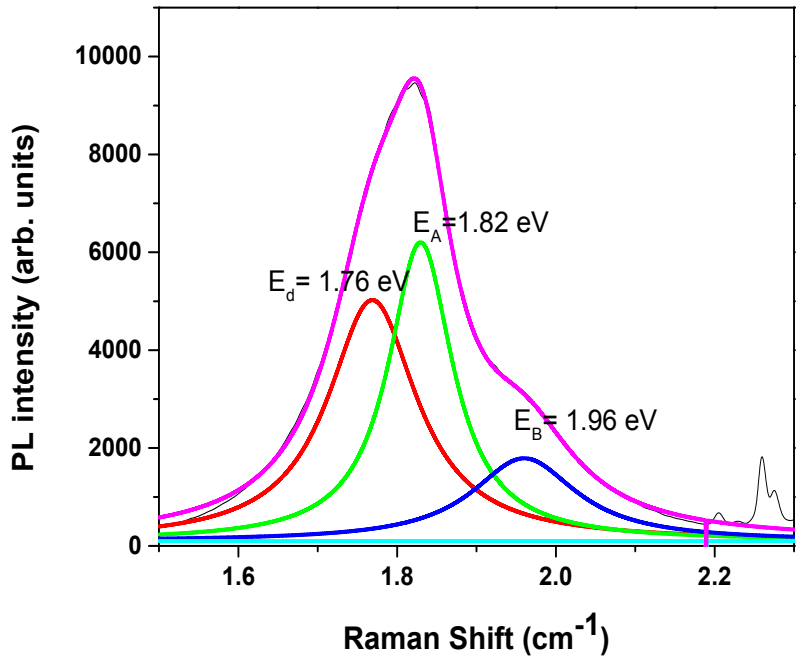

Figure S3: PL of MoS_2

Atomic force microscopy was used to study topography of treated MoS_2 sample (Figure S4). According to our previous research [S1] the inter-defect distance for 10E14 dose is 10 nm.

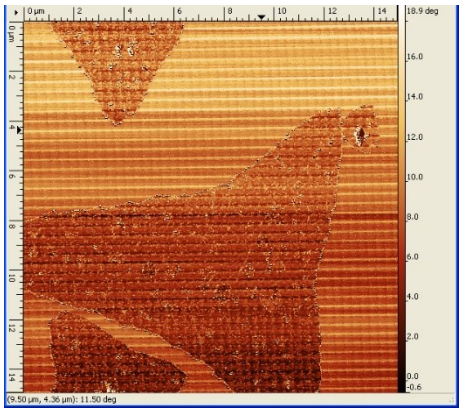

Figure S4: AFM topography and phase of irradiated MoS_2 sample

Attachment of F4TCNQ molecules was verified with TEM (Figure S5)

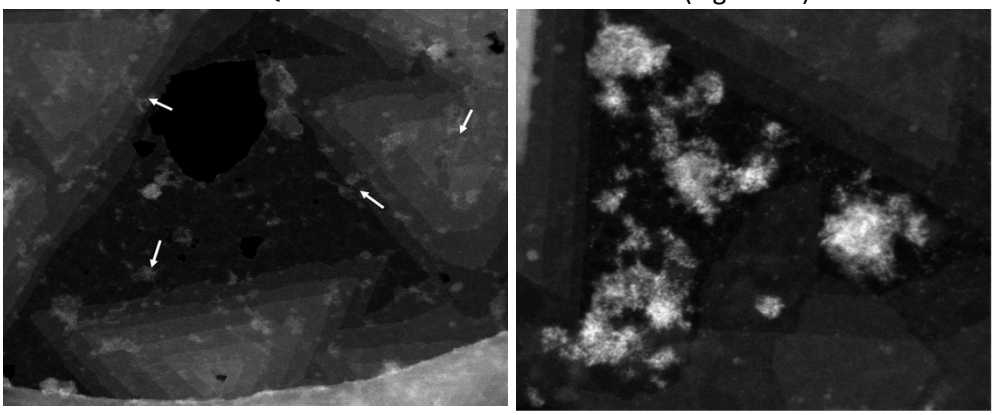

Figure S5: Transmission Electron Microscopy of F4TCNQ molecule on 2D MoS2

References:

[S1] F. Aryeetey, T. Ignatova and S. Aravamudhan, Quantification of defects engineered in single layer MoS_2, *RSC Advances*, 2020, **10**, 22996–23001.