Supporting Information

Defect-Rich Monolayer MoS_2 as a Universally Enhanced Substrate for Surface-Enhanced Raman Scattering

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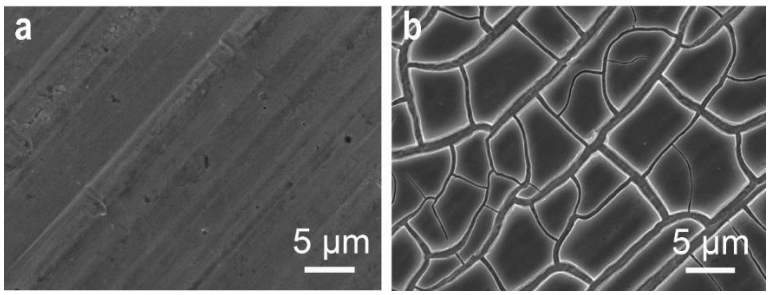

Figure S1. (**a**, **b**) SEM image of the untreated and electroplated surface of Mo foil.

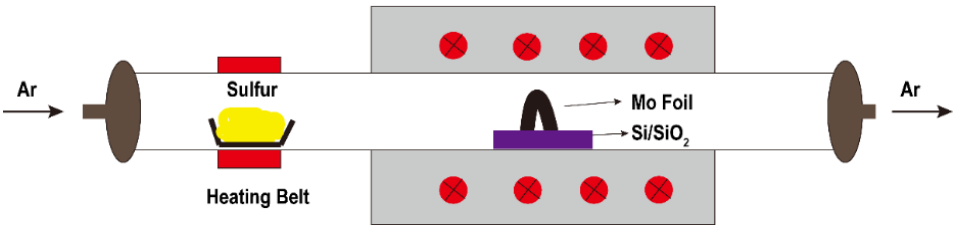

Figure S2. Schematic illustration of CVD system growing monolayer MoS_2.

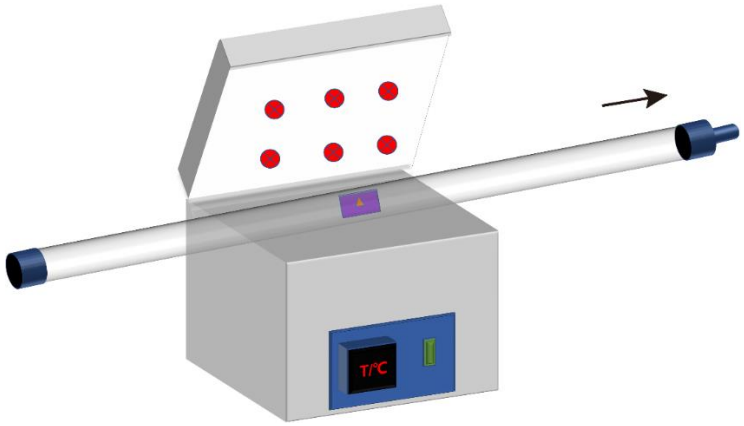

Figure S3. Schematic illustration of the etching process of monolayer MoS_2. Etching of monolayer MoS_2 is performed at temperatures below the growth temperature. The MoS_2 grown on the SiO_2/Si substrate is placed in the center of the glass tube, which is sealed at one end and connected to a vacuum oil pump at the other end.

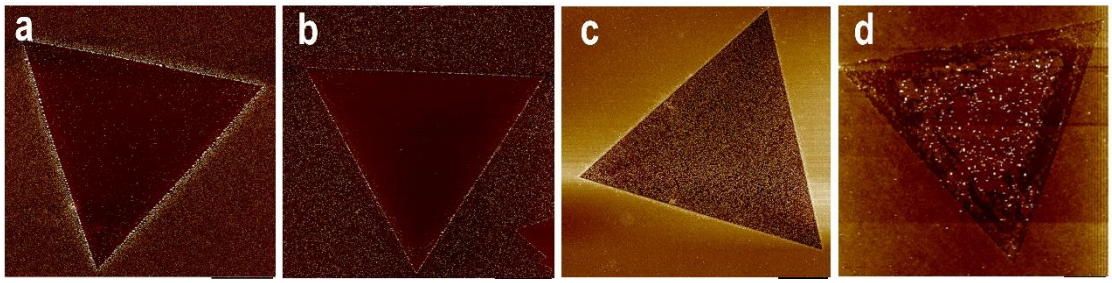

Figure S4. AFM image of (**a**) monolayer MoS_2, (**b**) mild etching monolayer MoS_2, (**c**) moderate etching monolayer of MoS_2, (**d**) severe etching monolayer of MoS_2.

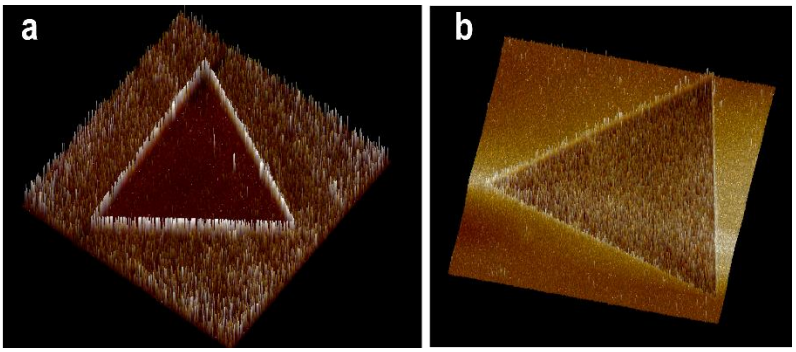

Figure S5. 3D AFM image of (**a**) monolayer MoS_2 and (**b**) moderate etching monolayer MoS_2.

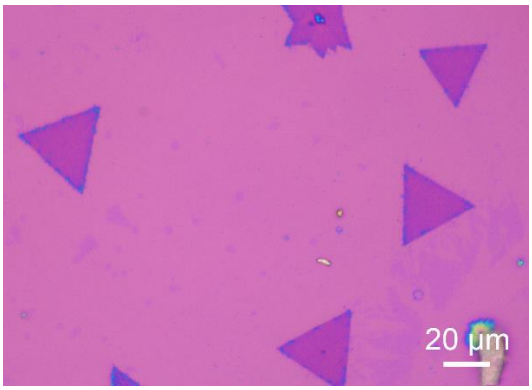

Figure S6. Optical photograph of dye molecules added dropwise on unetched monolayer MoS_2.

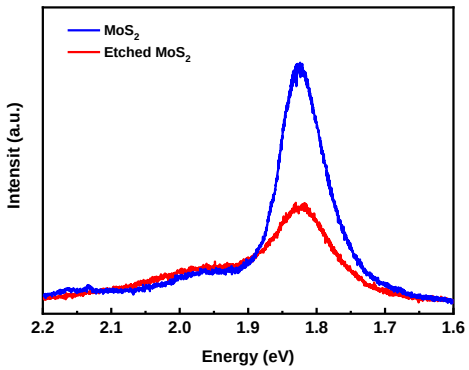

Figure S7. PL spectra of monolayer MoS_2 and etched monolayer MoS_2.

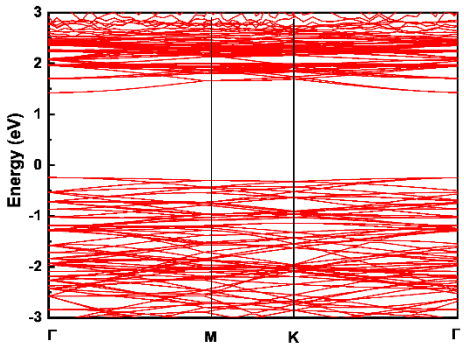

Figure S8. The calculated band structures of MoS_2 take Fermi level as reference.

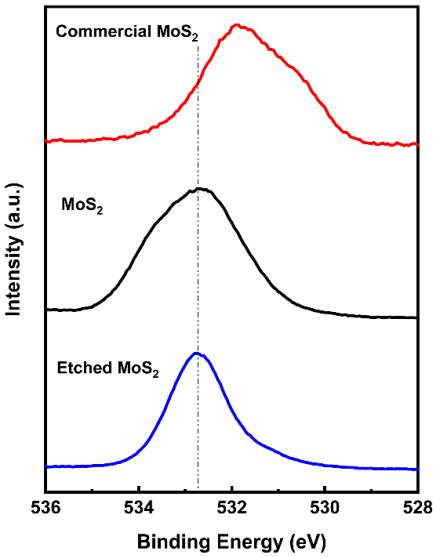

Figure S9. O 1s XPS spectrum of commercial MoS_2 powder, CVD grown monolayer MoS_2, and etched monolayer MoS_2.

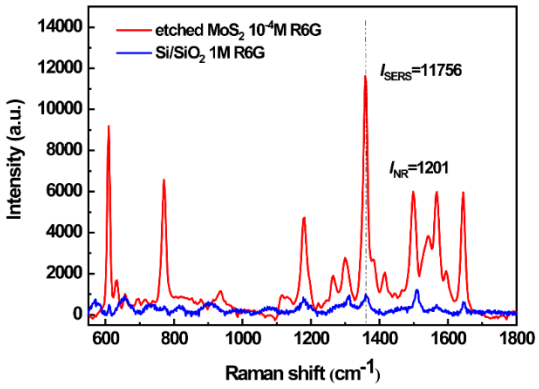

Figure S10. The enhancement factor (EF) of etched MoS_2 overlay 10^{-4}M R6G. Under the same test conditions as "Materials and Methods", we tested the Raman spectra of 10^{-4} M R6G on etched MoS_2 substrates and the Raman spectra of 1 M R6G on SiO_2/Si substrates. The final enhancement factor was obtained 10^5 by selecting the data where 1360 cm^{-1} was located using the calculation formula.

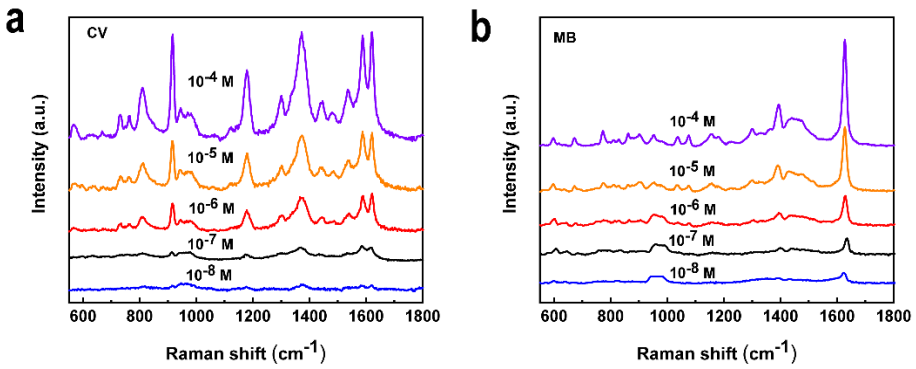

Figure S11. SERS measurements of (**a**) crystal violet (CV), and (**b**) methylene blue (MB) molecules probes on etched monolayer MoS_2 substrates.

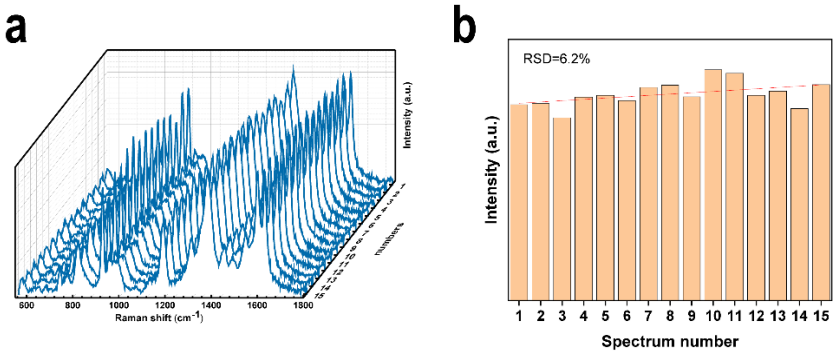

Figure S12. (**a**) SERS spectra of CV (10^{-5} M) coated on etched MoS_2 taken from 15 randomly chosen spots. (**b**) Histogram distribution of the Raman intensities at 1620 cm^{-1}.

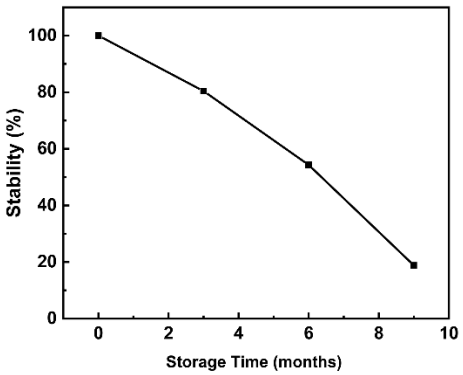

Figure S13. The stability ratio of 10^{-5} M R6G coated on etched MoS_2.

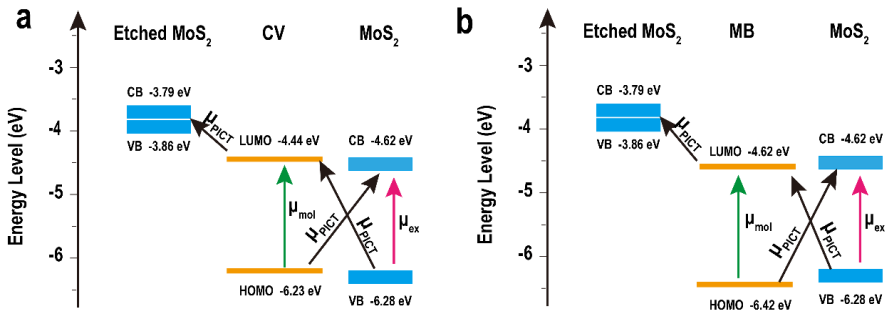

Figure S14. Energy level diagram and charge transfer transitions in the diagram comparing the charge-transfer pathways in (**a**) CV/etched MoS_2 and (**b**) MB/etched MoS_2.

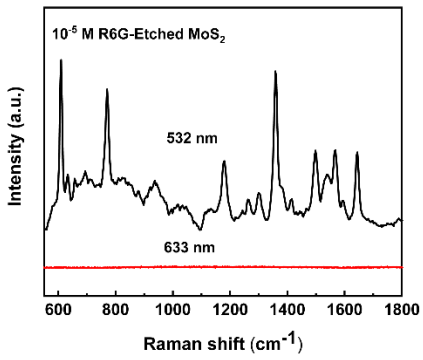

Figure S15. The Raman signals of 10^{-5} M R6G coated on etched MoS_2 were irradiated at 532 nm and 633 nm, respectively.

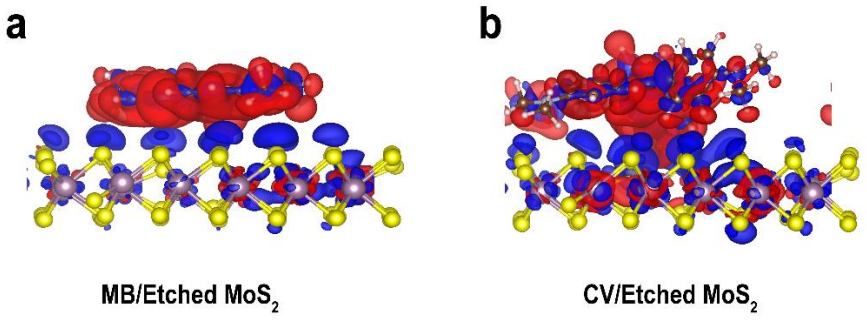

Figure S16. Side views of the electron density difference isosurface for MB (**a**), and CV (**b**) molecule absorbed on etched monolayer MoS_2.