Supporting Information for

Ultrasensitive Detection of Crystal Violet Using a Molybdenum Sulfide-Silver Nanostructure-based Sensing Platform: Roles of the Adsorbing Semiconductor on SERS Signal Enhancement

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Calculation of limit of detection (LOD)

The standard curve of linear detecting range was given as:

$$Y = A + B \times Log(X) \tag{1}$$

where A and B are intercept and slope of regression equation obtained through the plot of the logarithmic SERS intensity (Y) – logarithmic concentration (X).

The LOD is calculated using the following equation [1]:

$$LOD = 10^{[(Y_{blank} + 3SD)/Y_{blank} - A]/B} \tag{2}$$

where Y_{blank} and SD are the SERS signal and the standard deviation of blank sample, respectively.

SD is calculated via the well-known formula:

$$SD = \sqrt{\frac{1}{n-1} \times \sum_{i}^{n} (x_i - x_{average})^2} \tag{3}$$

where x_i if the "i" sample of the series of measurements, $x_{average}$ is the average value of SERS signal obtained from the blank sample repeated n times.

Calculation of relative standard deviation (RSD)

The RSD value of repeatability and reproducibility is calculated via the well-known formula:

$$\text{RSD} = \frac{SD \times 100}{x_{average}} \tag{4}$$

where SD is the standard deviation that calculates using equation 4 and $x_{average}$ is the average value of SERS signal obtained from each measurement.

Calculation of enhancement factor (EF)

The EF value is calculated according to the well-established equation, which was employed in several published studies [2, 3]:

$$EF = \frac{I_{SERS}}{I_{Raman}} \times \frac{N_{bulk}}{N_{surface}} \quad (5)$$

where I_{SERS} and I_{Raman} are Raman signal intensity of the analyte with and without SERS from the substrate, respectively; and N_{bulk} is the number of analyte molecules that are probed on the Raman spectrum, while $N_{surface}$ is the number of analyte molecules probed using SERS.

N_{bulk} can be calculated following:

$$N_{bulk} = \frac{A_{laser} \times h \times \rho}{M} \times N_A$$

(6)

where A_{laser}, h, ρ and m are the laser spot area, the focal length, the density of the solid analyte and its molecular weight, respectively; and N_A is the Avogadro number.

$N_{surface}$ can be expressed as:

$$N_{surface} = \frac{C \times V}{A_{substrate}} \times N_A \times A_{laser} \quad (7)$$

where C, V, $A_{substrate}$ are the concentration, the volume drop-casted of the analyte, and the area of the substrate, respectively; N_A is the Avogadro number; and A_{laser} is the laser spot area.

Thus, EF can be calculated as:

$$EF = \frac{I_{SERS}}{I_{Raman}} \times \frac{N_{bulk}}{N_{surface}} = \frac{I_{SERS}}{I_{Raman}} \times \frac{h \times \rho \times A_{substrate}}{M \times C \times V}$$

(8)

In our case, I_{SERS} and I_{Raman} is Raman signal intensity with and without SERS substrate of Crystal Violet (736 cm^{-1}), h = 2 μm = 2×10^{-4} cm, ρ = 1.19 g/cm^3, M = 408 g/mol, $A_{substrate}$ = 4π mm^2, C = 10^{-7}mol/L, V = 5 μL = 5×10^{-6} L.

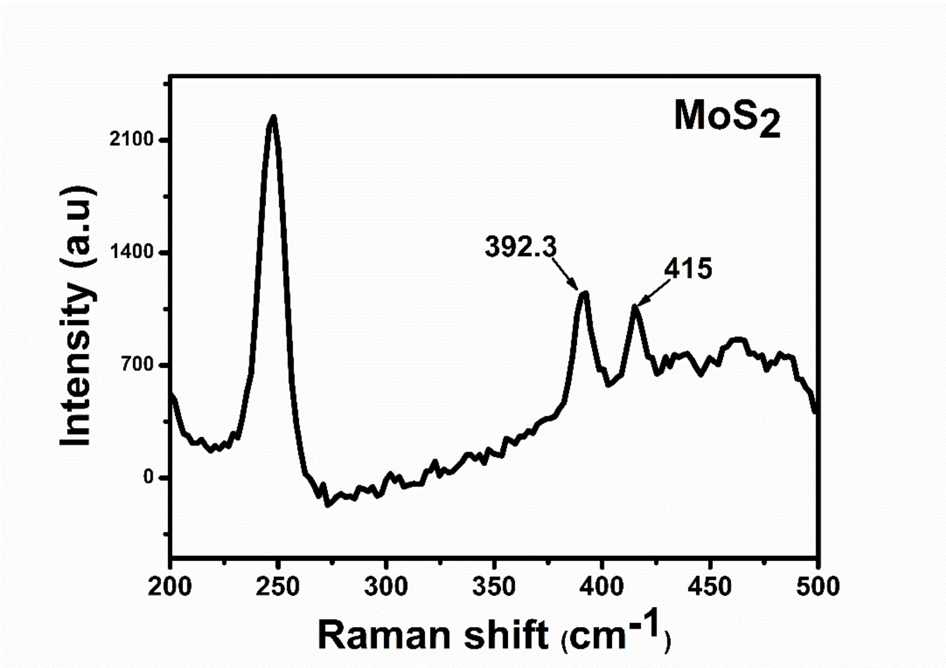


Figure S1: Raman spectrum of MoS_2 nanosheets.

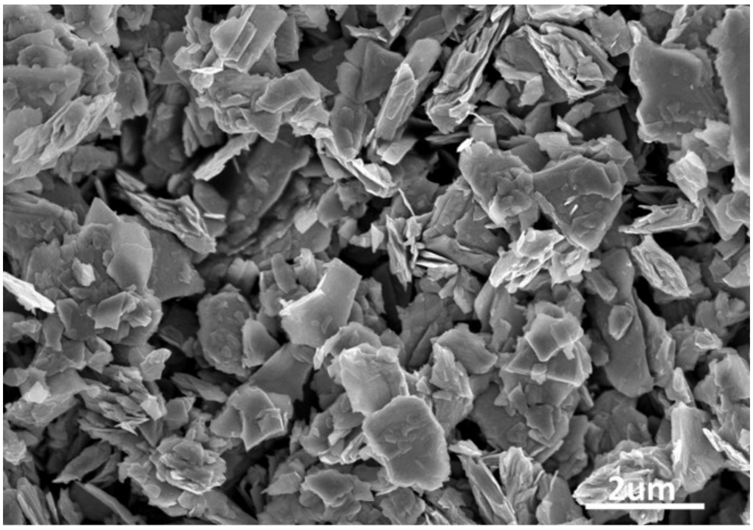


Figure S2: SEM image of MoS_2

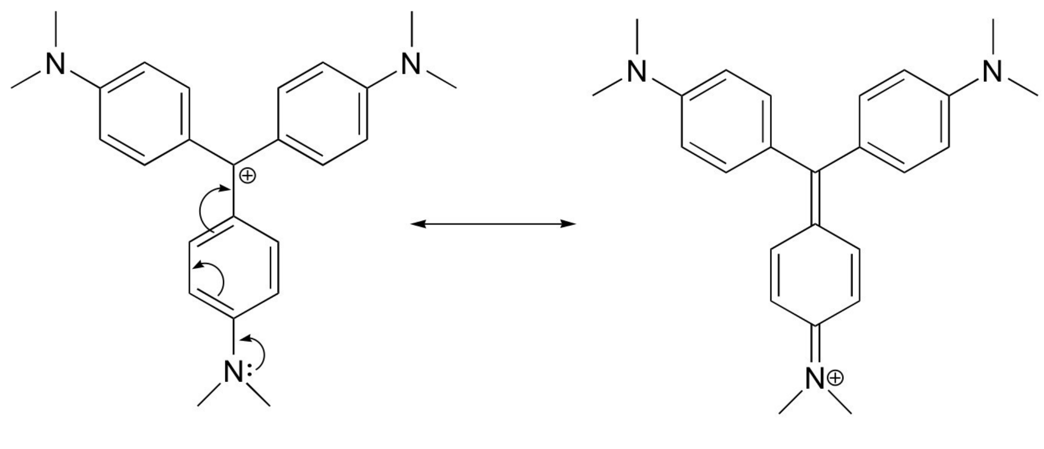


Figure S3: Molecular structure of CV and its delocalization of electrons.

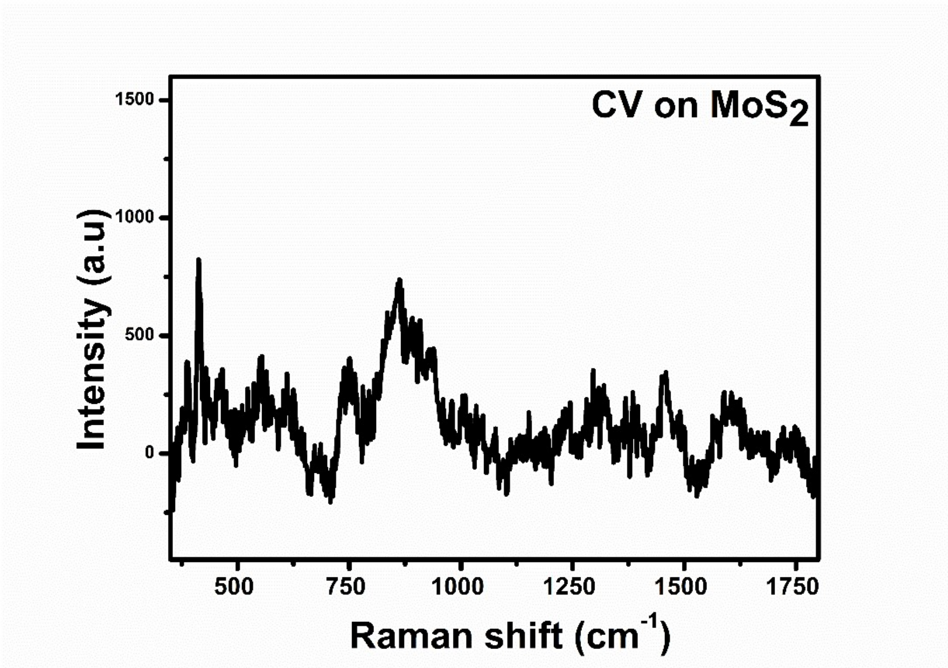


Figure S4: SERS spectrum for CV (10^{-5} M) on MoS_2.

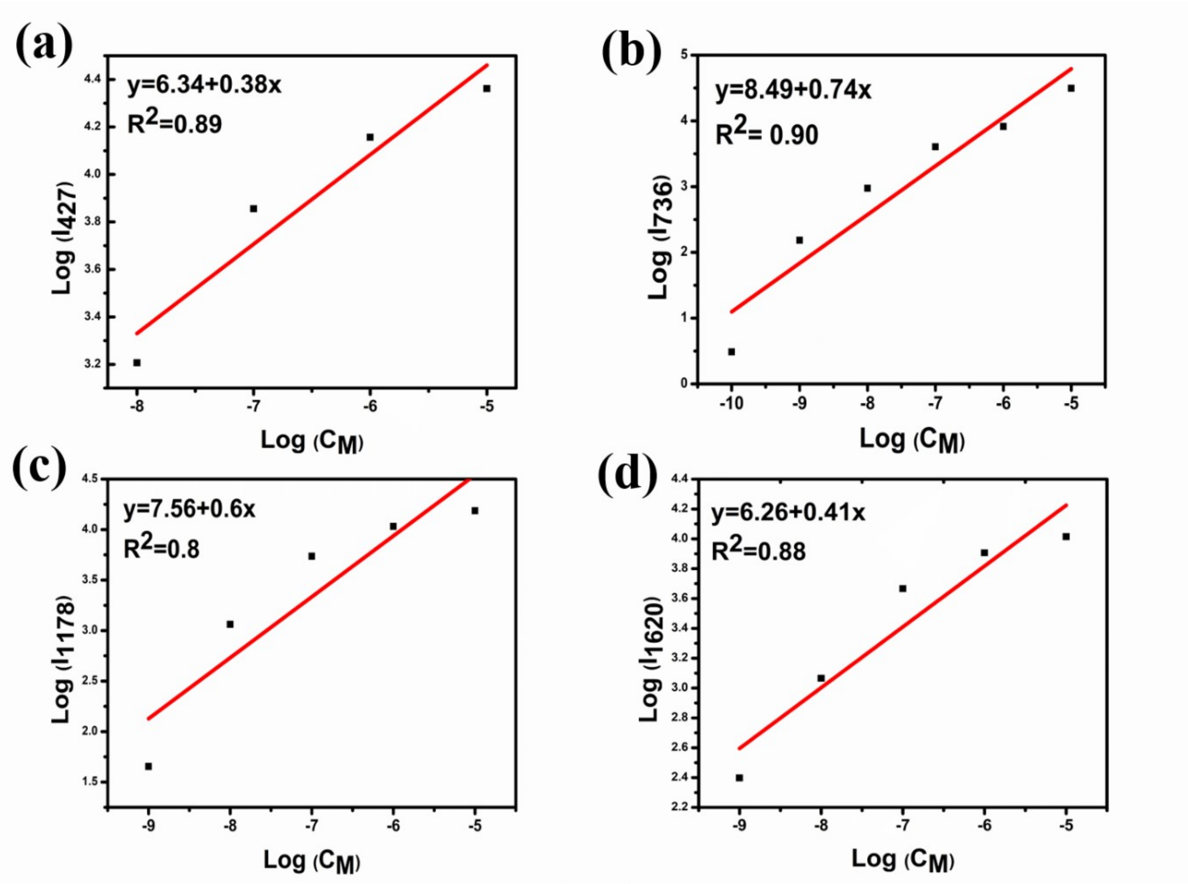
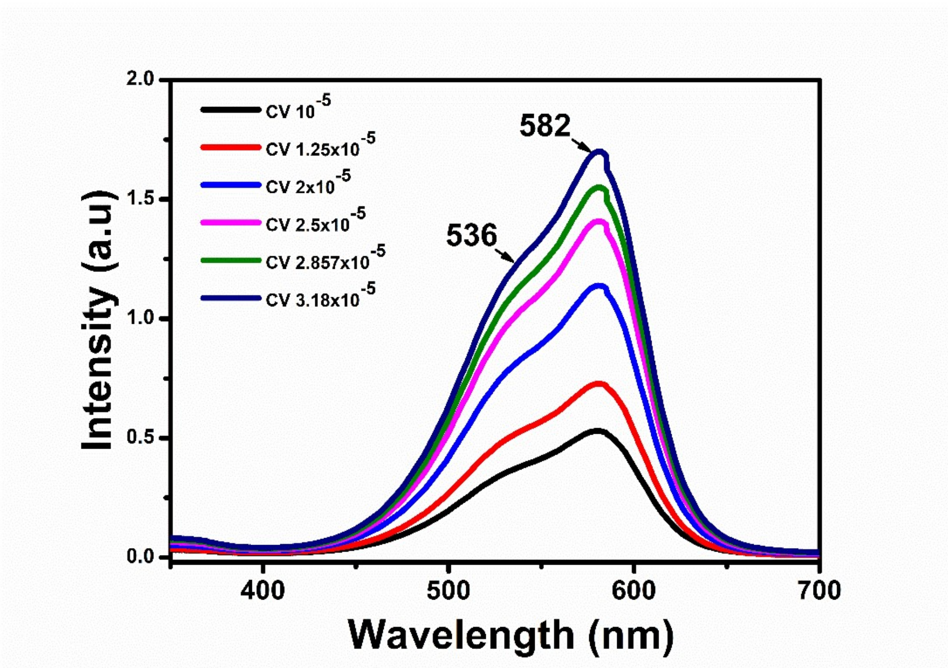


Figure S5: Plot of log of SERS intensity against CV concentration at (a) 427 cm^{-1}; (b) 736



cm^{-1}; (c) 1178 cm^{-1}; and (d) 1620 cm^{-1}.

Figure S6: UV-vis absorption spectrum of CV at different concentrations.

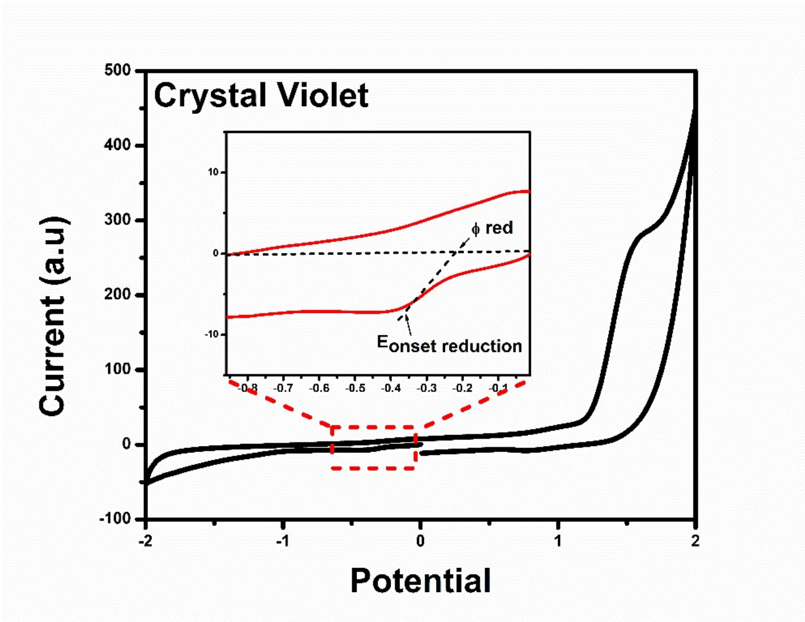


Figure S7: Cyclic voltammogram of Crystal Violet and magnified curve to determine the onset reduction potential (ϕ_{red}).

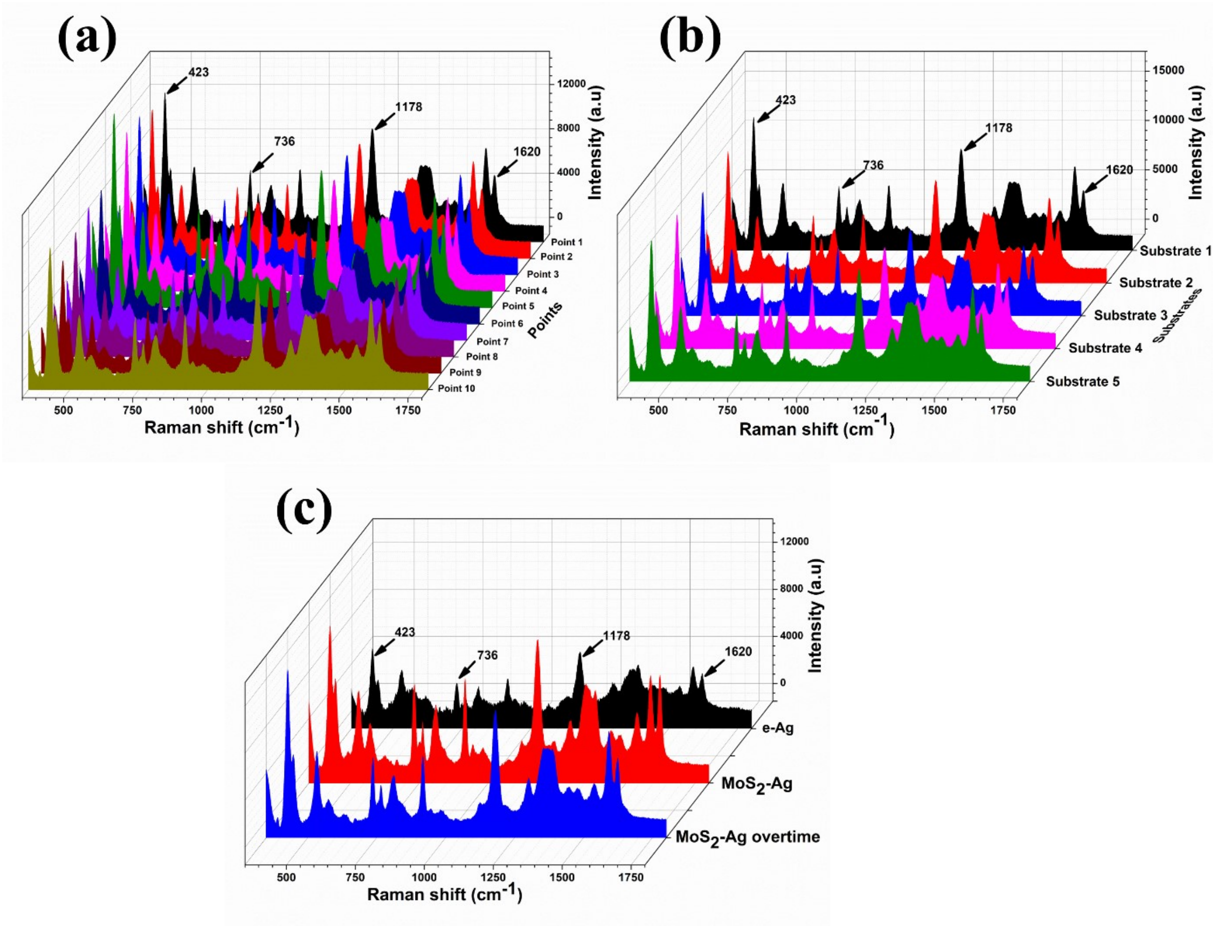


Figure S8: (a) Repeatability and (b) Reproducibility of SERS sensors for CV on MoS_2/Ag overtime; (c) Raman spectrum of CV 10^{-6}M comparing e-Ag, MoS_2-Ag and MoS_2-Ag overtime.

References

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(2) Le Ru, E.C.; Blackie, E.; Meyer, M.; Etchegoin, P.G. Surface Enhanced Raman Scattering Enhancement Factors: A Comprehensive Study. *J. Phys. Chem. C*, 2007, **111**, 33, 13794-13803. https://doi.org/10.1021/jp0687908.

(3) Fu, W. L.; Zhen, S. J.; Huang, C. Z; One-pot green synthesis of graphene oxide/gold nanocomposites as SERS substrates for malachite green detection. *Analyst*, 2013, **138**, 3075-3081. https://doi.org/10.1039/C3AN00018D.