

Supporting Information for “Surface-Enhanced Raman Scattering of Hydrogen Plasma-Treated Few-Layer MoTe₂”

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1. Experimental Methods

Sample Preparation. Few layer MoTe₂ films were prepared by mechanical exfoliation method from the bulk MoTe₂ crystal, and transferred onto 300 nm SiO₂/Si substrates. Then the samples were treated with hydrogen plasma at 100 w under the pressure of 5 Pa.

Characterization. The Dimension ICON AFM equipment was used to identify the thickness of the original and hydrogen plasma treated MoTe₂ (H₂-MoTe₂) samples, and the Thermo Fisher XPS equipment was used to detect the atomic concentrations and valence state of the original and H₂-MoTe₂ samples. All Raman spectra were collected by the LABRAM HR-800 Raman equipment with the laser line of 532nm.

SERS measurement. The $10^{-7} \sim 10^{-13}$ M R6G aqueous solution were diluted from 10^{-3} M aqueous solution in the certain proportion. The prepared samples were soaked in a specific solution for 40 minutes (The samples were soaked in the solution for enough time to ensure that the concentration of the R6G/RhB molecules on MoTe₂ sample is the same as in the solution), then taken out and rinse with ethanol, and finally blown with nitrogen. Raman spectrum were collected with a $50 \times$ L objective lens; the laser spot diameter was about 1 μm ; the laser power was 0.5 mW and the acquisition time was 40 s. Raman spectra were measured at 15 different locations on each sample, and the mean value of signal intensity was taken as the standard deviation for the final analysis standard.

2. Optical microscopy images of pristine and H₂-MoTe₂ film

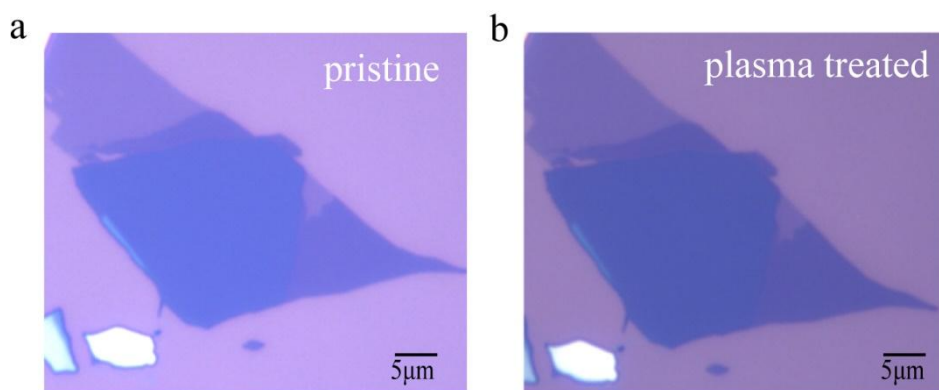


Fig. S1. Optical microscopy images of pristine and hydrogen plasma treated MoTe₂ films on 300-nm SiO₂/Si substrates.

3. Raman spectra of R6G molecules coated on pristine MoTe₂ and H₂-MoTe₂ substrates

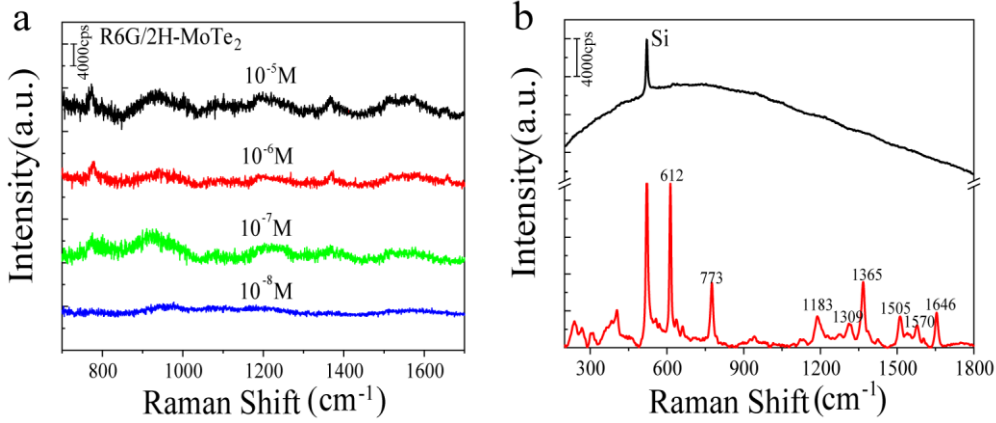


Fig. S2. (a) SERS spectra of R6G coated on pristine MoTe₂ flakes (3L) with various concentrations from 10⁻⁵ to 10⁻⁸ M. (b) Raman spectra of the R6G molecules deposited on bare SiO₂ (black line) and on 2D H₂-MoTe₂ (red line). The signal integration times were both 40 s.

4. The calculated enhancement factors

The Raman enhancement factors (EF) of the substrates were estimated according to the following equation (1)^[1, 2]

$$E_f = \frac{I_{SERS}/N_{SERS}}{I_{bulk}/N_{bulk}} \quad (1)$$

where I_{SERS} and I_{bulk} represent the Raman intensity of probe molecules on hydrogen plasma treated MoTe₂ substrate and the Raman intensity measured from the bulk probe molecules without substrate, respectively. N_{SERS} and N_{Bulk} denote the corresponding numbers of probe molecules on H₂ plasma treated MoTe₂ substrate and that without substrate, respectively. N_{SERS} is written as $N_{SERS} = \frac{CVN_A A_{Raman}}{A_{sub}}$ and N_{bulk} is written as $N_{bulk} = \frac{\rho h A_{Raman} N_A}{M}$. C is the corresponding concentration of probe molecules (10⁻⁷ M) and V is the volume of the droplet (20 μ L), h is the effective layer

depth of $21 \mu\text{m}^{[3]}$. N_A is the Avogadro constant and A_{Raman} is the laser spot area. A_{sub} is the effective area of the substrate, which is approximately $9\pi \text{ mm}^2$. M is the molar mass of dye (479.02 g/mol for R6G) and density ρ of bulk R6G is 1.15 g cm^{-3} .

For the SERS measurement, the intensity is obtained by taking average from measurements of 15 spots, and the number of analyte molecules is estimated by equation (1) on the assumption that the analyte molecules were distributed uniformly on the substrates. The peak of 1647 cm^{-1} was selected to calculate the enhancement factor. The Raman 1647 cm^{-1} intensity of R6G/ $\text{H}_2\text{-MoTe}_2$ (10^{-7} M) is 3940 counts with 40 s acquisition time, and that of bulk R6G is 3700 counts with 1 s acquisition time. From the observed spectra we obtain for the ratio of intensities $I_{\text{SERS}} / I_{\text{Bulk}} = (3940/40) / (3700/1) = 2.7 \times 10^{-2}$. The EF at 10^{-7} M dye concentration is estimated to be around 1.83×10^6 .

5. Raman spectra of RhB molecules on $\text{H}_2\text{-MoTe}_2$, MoTe_2 and SiO_2 substrates

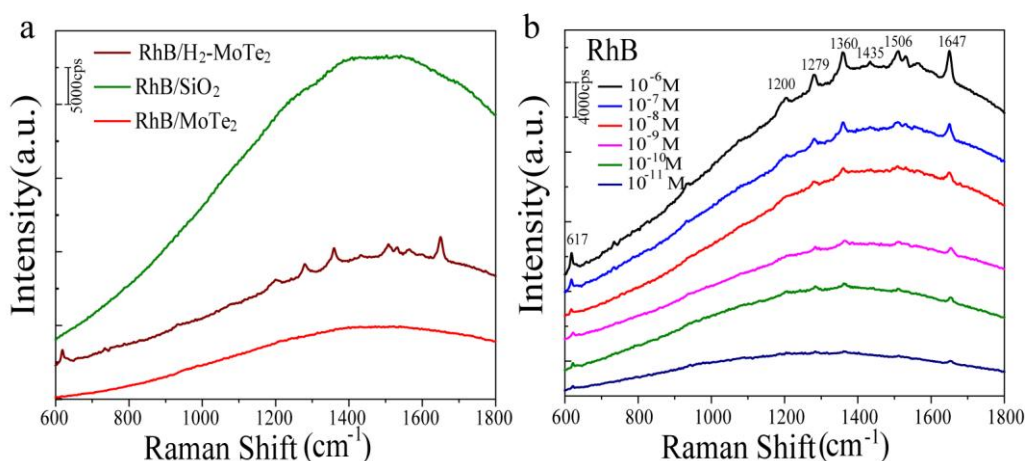


Fig. S3. (a) Raman spectra of RhB coated on $\text{H}_2\text{-MoTe}_2$, MoTe_2 and SiO_2 substrates with a fixed concentration of 10^{-7} M . (b) SERS spectra of RhB coated on $\text{H}_2\text{-MoTe}_2$ flakes (3L) with various concentrations from 10^{-6} to 10^{-11} M .

6. Raman spectra of R6G molecules on $\text{Ar}(\text{O}_2)\text{-MoTe}_2$ substrates

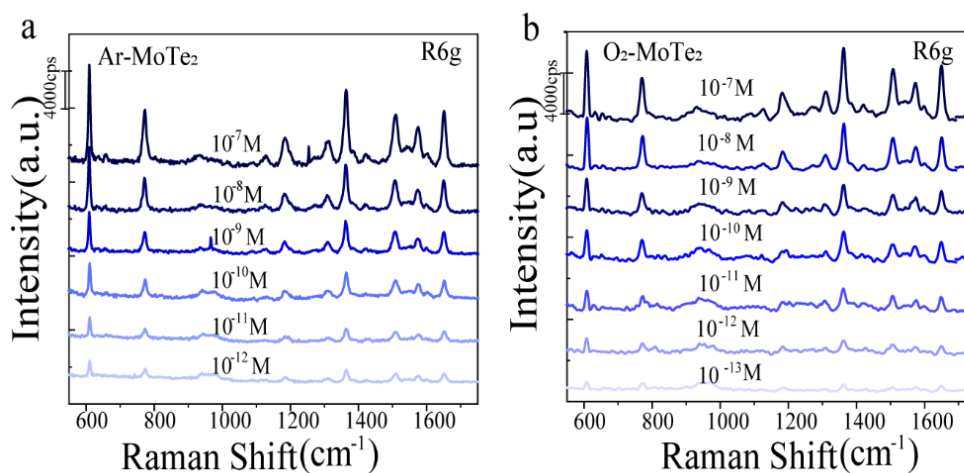


Fig. S4. (a) Raman spectra of R6g coated on Ar-MoTe₂ substrates with various concentrations from 10⁻⁷ to 10⁻¹² M. (b) Raman spectra of R6g coated on O₂-MoTe₂ substrates with various concentrations from 10⁻⁷ to 10⁻¹³ M.

Supplementary references

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- [2] Mehmet, Yilmaz, Esra, Babur, Ozdemir, Rebecca L, Giesecking, Yavuz, Dede and Ugur 2017 *Nature materials* **141** 4097
- [3] Cai W B, Ren B, Li X Q, She C X, Liu F M, Cai X W and Tian Z Q 1998 *Surface Science* **406** 9