Supporting Information for

Probing Local Strain at MX_2−Metal Boundaries with Surface Plasmon-Enhanced Raman Scattering

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1. Optical images, Raman spectra, and PL spectra of exfoliated monolayer MoS_2 without and with 5 nm Ag deposition.

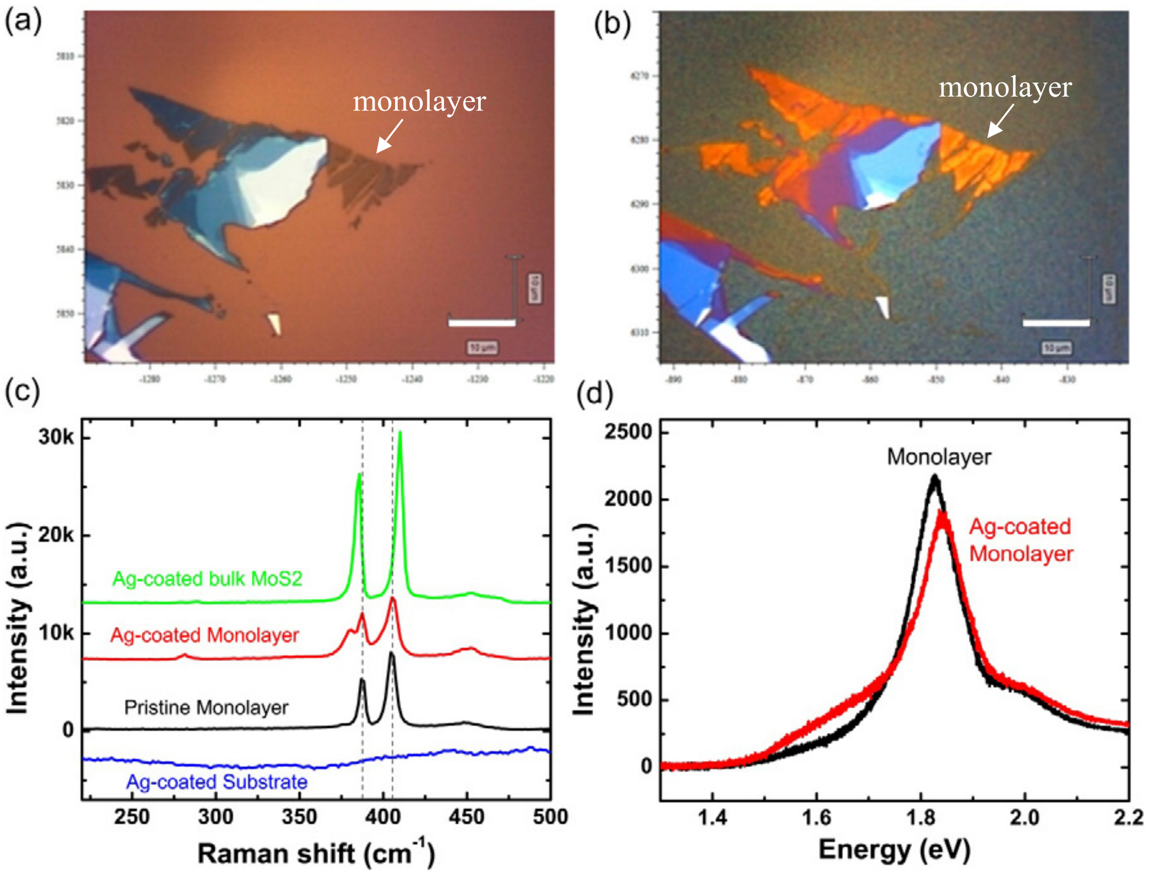

Figure S1. (a) Optical image of pristine exfoliated MoS_2 monolayer and few layers. (b) Corresponding optical image of MoS_2 layers after 5 nm Ag deposition. (c) Raman spectra of pristine MoS_2 monolayer, Ag-coated monolayer, Ag-coated bulk, and Ag-coated SiO_2/Si substrate. The nominal Ag thickness is 5 nm. The E^1_{2g} mode splits into two peaks for Ag-coated monolayer. The energy splitting of E^1_{2g} Raman mode can be estimated to be 7.5 cm^{-1} from a Lorentzian multi-peak fitting. The spectra are shifted vertically for clarity. (d) PL spectra of pristine monolayer and Ag-coated monolayer. A shoulder PL peak appears at an energy of ~0.15 eV below the A exciton PL.

2. Raman and PL spectra of monolayer and bilayer MoS$_2$ with 1 nm Au deposition.

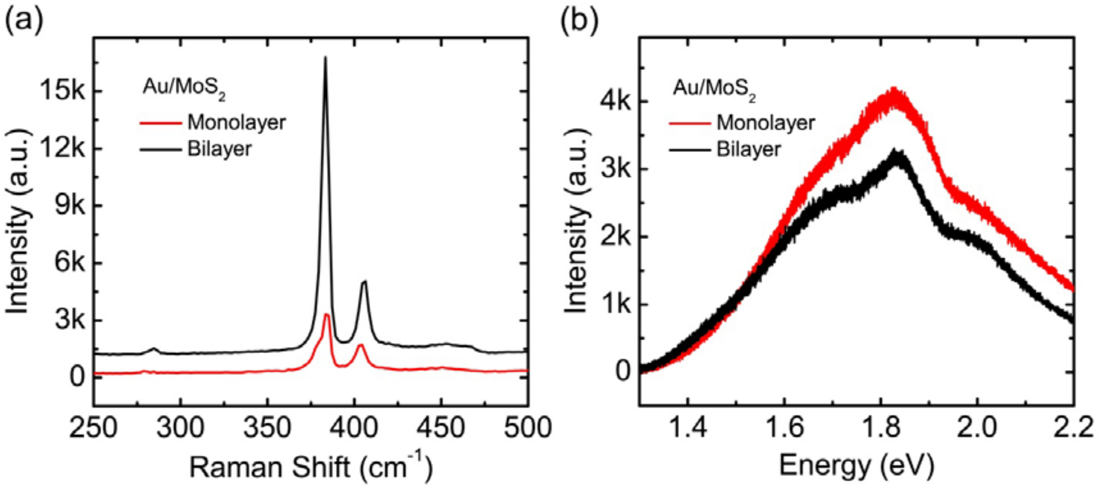

Figure S2. (a) Raman spectra of Au-coated monolayer and bilayer MoS$_2$ excited by a 488 nm laser. The nominal Au thickness is 1 nm. It shows that the E^1_{2g} Raman mode also splits for Au-coated monolayer, and the energy splitting is estimated to be 4.4 cm^{-1} from a Lorentzian multi-peak fitting. (b) PL spectra of Au-coated monolayer and bilayer MoS$_2$.

3. Reflection spectrum of Ag NPs and Raman spectra of Ag/MoS$_2$ at different excitation wavelengths.

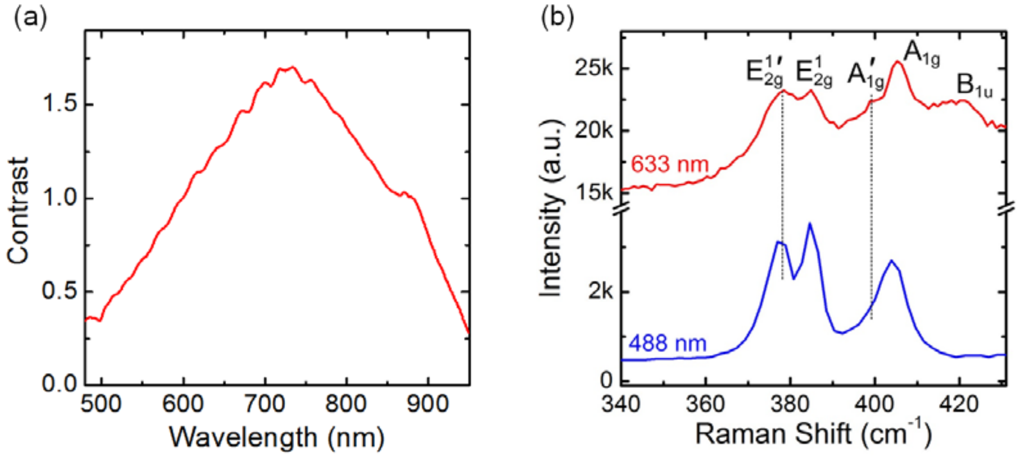

Figure S3. (a) Reflection spectrum of Ag nanoparticles on monolayer MoS_2. (b) Raman spectra of Ag-coated monolayer MoS_2 at different excitation wavelengths. The thickness of Ag is 1 nm nominally.

4. Analysis of strain distribution in monolayer and bilayer MoS_2 coated with Ag NP.

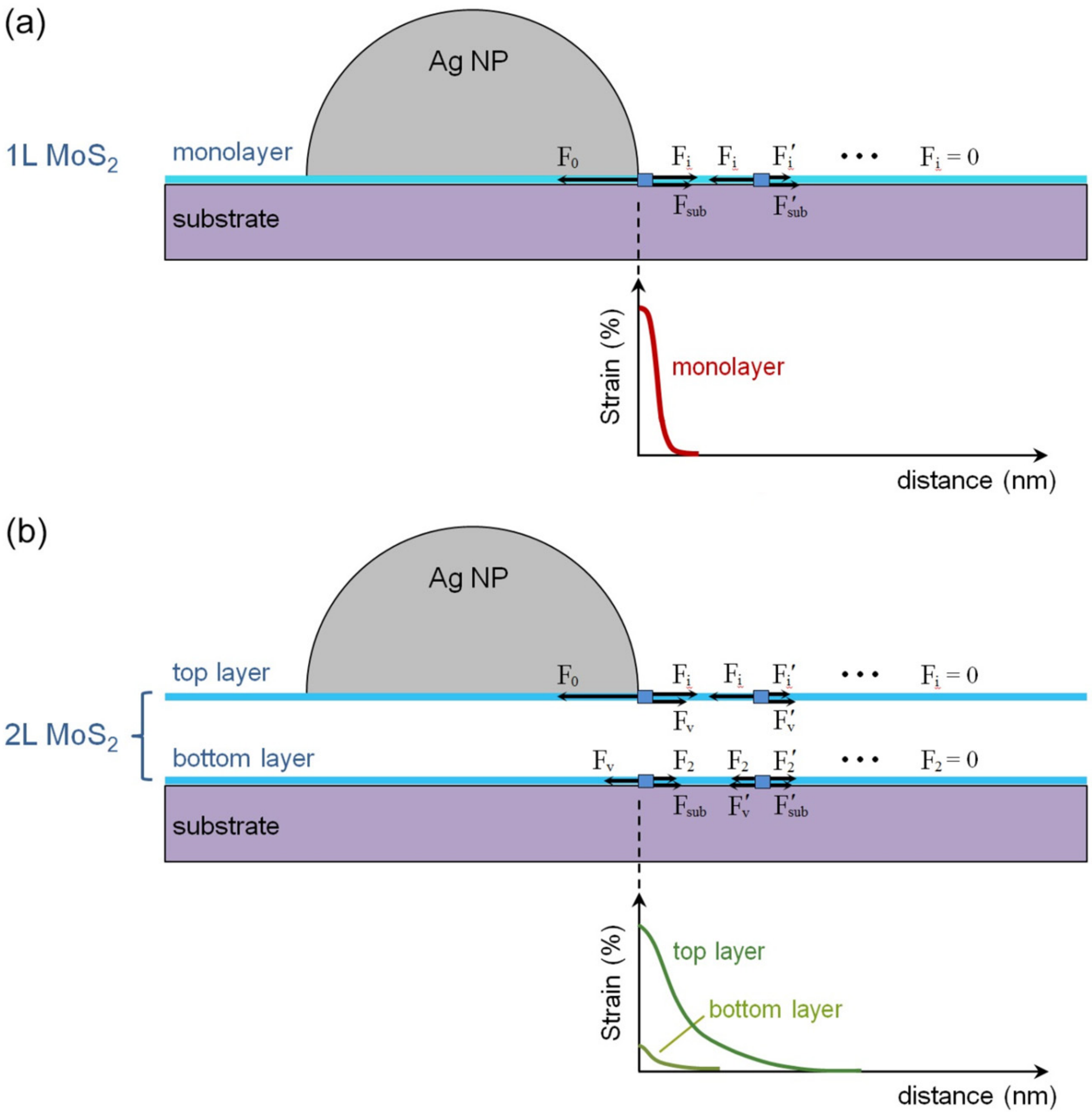

Figure S4. Force analysis and diagram of strain relaxation in monolayer (a) and bilayer (b) MoS_2 deposited with Ag NP.

In Figure S4, we provide a qualitative model for the strain distribution in monolayer and bilayer MoS_2. Local force analysis for the element nearest to the Ag NP and the proximate elements are shown in Fig. S4a for monolayer. $\mathbf{F}_0$ is the force coming from the intrinsic stress. $\mathbf{F}_\mathrm{i}$ ($\mathbf{F}'_\mathrm{i}$) is the internal force in monolayer, which monotonously decreases until the strain is fully relaxed. $\mathbf{F}_\mathrm{sub}$ ($\mathbf{F}'_\mathrm{sub}$) is the external force from substrate, representing the load transfer to substrate. Similarly, the local force analysis for the elements in both top layer and bottom layer is shown in Fig. S4b for bilayer. $\mathbf{F}_\mathrm{v}$ ($\mathbf{F}'_\mathrm{v}$) is the interlayer friction coming from interlayer van de Waals force, representing the load transfer from the top to the bottom layer. $\mathbf{F}_2$ ($\mathbf{F}'_2$) is the internal force in bottom layer, which also monotonously decreases until the strain is fully relaxed.

For the monolayer MoS_2 or the top layer of bilayer MoS_2, the transfer equation of intrinsic stress can be described as

$$dF/A = (F_i - F'_i)/(\Delta wh) = F'_{sub}(or\ F'_v)/(\Delta wh) = \mu f_{V-W}/h \cdot dx \qquad \text{(Eq. S1)}$$

where A is the cross-section area, h is the thickness of monolayer, Δw is the width of the selected element, μ is the friction coefficient between the top layer and the underneath interface, f_{V-W} is the van der Walls force in an unit area of the top layer, and *dx* is the slide distance at that position (the last step is only true up to the sliding point).

Equation S1 indicates that the propagation of intrinsic stress with distance strongly depends on $\mu f_{V-W}/h$. According to the two dimensional Young's modulus measurement,[2] the modulus of bilayer MoS_2 is much lower than twice the value of monolayer, probably due to the interlayer sliding, which suggests that the interaction between top and bottom layers of bilayer is weaker than the interaction from substrate. In monolayer MoS_2, therefore, μf_{V-W} are larger than the value in the top layer of bilayer MoS_2 because of the strong clamping by the substrate. As a result, the strain relaxes faster with distance in monolayer MoS_2 than in the top layer of bilayer MoS_2.

In the bottom layer of bilayer MoS_2, stress transfer can be similarly described as

$$dF/A = (\mu^{sub} f_{V-M}^{sub} - \mu^{top} f_{V-M}^{top})/h \cdot dx \qquad \text{(Eq. S2)}$$

However, because the initial local stress in the bottom layer is much lower than the top layer due to the weak van der Waals interaction, the strain in the bottom layer should be much smaller than that in the top layer. Curves of strain distribution are also illustrated in Fig. S4.

References:

1. van der Zande, A. M.; Huang, P. Y.; Chenet, D. A.; Berkelbach, T. C.; You, Y.; Lee, G.-H.; Heinz, T. F.; Reichman, D. R.; Muller, D. A.; Hone, J. C., Grains and grain boundaries in highly crystalline monolayer molybdenum disulphide. *Nature Materials* 2013, 12, 554-561.
2. Bertolazzi, S.; Brivio, J.; Kis, A., Stretching and Breaking of Ultrathin MoS_2. *ACS Nano* 2011, 5, 9703-9709.