Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for

Photoluminescence of Monolayer MoS_2 on $LaAlO_3$ and $SrTiO_3$ substrates

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1. Calculation of the enhancement factor for Raman and Photoluminescence (PL) intensity normalization.

The enhancement factor is defined as $\Gamma^{-1} = \frac{I_{MoS_2}^{freesanding}}{I_{MoS_2}^{on-substrate}}$. The calculation method is based on the model including optical interference and absorption effects due to the substrate geometry, which is described in Ref.[1] By multiplying Γ^{-1} for each measured Raman and PL spectrum, the optical interference effect attributed to the different samples is eliminated and intrinsic difference between each system can be compared. Fig.S1(a) shows the calculated enhancement factor Γ^{-1} of monolayer to four-layer MoS_2 on SiO_2, $LaAlO_3$, Gel-film and $SrTiO_3$ substrate. Fig.S1(b) and (c) show the comparison of the raw and normalized Raman and PL spectra of monolayer MoS_2 on different substrates.

2. Thickness-dependence of Raman spectra for MoS_2 on different substrates

Fig.S2 shows the normalized Raman spectra of 1-4L MoS_2 on different types of substrates.

From the Raman spectra, for each kind of substrate, both E^1_{2g} and A_{1g} modes of MoS_2 are clearly observed. The dependence of these two mode frequencies on layer thickness exhibits similar trend for four selected substrates, i.e. E^1_{2g} mode moves to lower frequency end while A_{1g} mode shifts to higher frequency end when the number of layers increases.

3. Thickness-dependence of PL spectra for MoS_2 on different substrates

Fig.S3 shows the normalized PL spectra of 1-4L MoS_2 on different substrates. It is known that, for monolayer MoS_2, PL spectrum is composed with two excitonic peaks A and B, associated with direct optical transitions from the lowest conduction bands to the highest spin-split valence bands.[2] From Fig.S3, both excitonic peaks can be clearly observed in all samples. Meanwhile, all the samples show very similar thickness-dependence, i.e. when the number of layers decreases, the intensity of peak A dramatically increases and reaches to the maximum for monolayer thin film, due to the indirect-to-direct band transition. For the bilayer samples, we also observed an emission peak around 1.55 eV (Peak I) which is ascribed to indirect band transition. Besides the change of peak intensity, the position and shape of peak also changes with thin film thickness. Peak A moves to higher energy end when thin film thickness decreases.

4. The fitting of PL spectra of monolayer MoS_2 on different substrates.

We fit the experimental PL data of monolayer MoS_2 on SiO_2, $LaAlO_3$, Gel-film and $SrTiO_3$ substrate. The fits are composed by three Lorenz functions corresponding neutral excitons (A^0), trions (A^-) and the direct band transition (peak B) . The fitting results are shown in Fig.S4.

5. The electron density calculation

We estimate the electron density in monolayer MoS_2 from the analysis of PL intensity of neutral excitons and trions emissions. According to mass action model which is based on the dynamic equilibrium between neutral excitons (A^0), free electrons and trions (A^-), the following relationship is obtained:[3-5]

$$\frac{N_{A^0} n_{el}}{N_{A^-}} = (\frac{4 m_{A^0} m_e}{\pi \hbar^2 m_{A^-}}) k_B T \exp(-\frac{E_b}{k_B T})$$

Where k_B is the Boltzmann constant, E_b is the trion binding energy (~20 meV),[6] T is the temperature. m_e (0.35 m_0) , m_{A^0} (0.8 m_0) and m_{A^-} (1.15 m_0) are the effective mass of neutral excitons, trions and electrons, respectively, where m_0 is the mass of free electrons.[7] N_{A^0} and N_{A^-} are the population of neutral exciton and trion, repectively. n_{el} is the electron density.

To establish the relationship between PL intensity and the population of neutral exciton and trion, we consider a three level model that includes a trion, an exciton and the ground state.[8] Based on this model, the PL intensity weight can be related to the population of neutral exciton and trion as:

$$\frac{I_{A^-}}{I_{total}} = \frac{I_{A^-}}{I_{A^-} + I_{A^0}} = \frac{\frac{\gamma_{A^-}}{\gamma_{A^0}} \frac{N_{A^-}}{N_{A^0}}}{1 + \frac{\gamma_{A^-}}{\gamma_{A^0}} \frac{N_{A^-}}{N_{A^0}}}$$

Where I_{A^0} and I_{A^-} are integrated PL intensity of neutral exciton and trion, respectively. γ_{A^0} and γ_{A^-} are the relative decay rate of the neutral exciton and trion, respectively. The value of $\frac{\gamma_{A^-}}{\gamma_{A^0}}$ is ~ 0.15 according to Mouri et al.'s study.[8]

Combining the three level model and mass action model, the n_{el} can be expressed as:

$$n_{el} = \frac{\frac{I_{A^-}}{I_{total}}}{\frac{\gamma_{A^-}}{\gamma_{A^0}} (1 - \frac{I_{A^-}}{I_{total}})} \left[(\frac{4 m_{A^0} m_e}{\pi \hbar^2 m_{A^-}}) k_B T \exp(-\frac{E_b}{k_B T}) \right]$$

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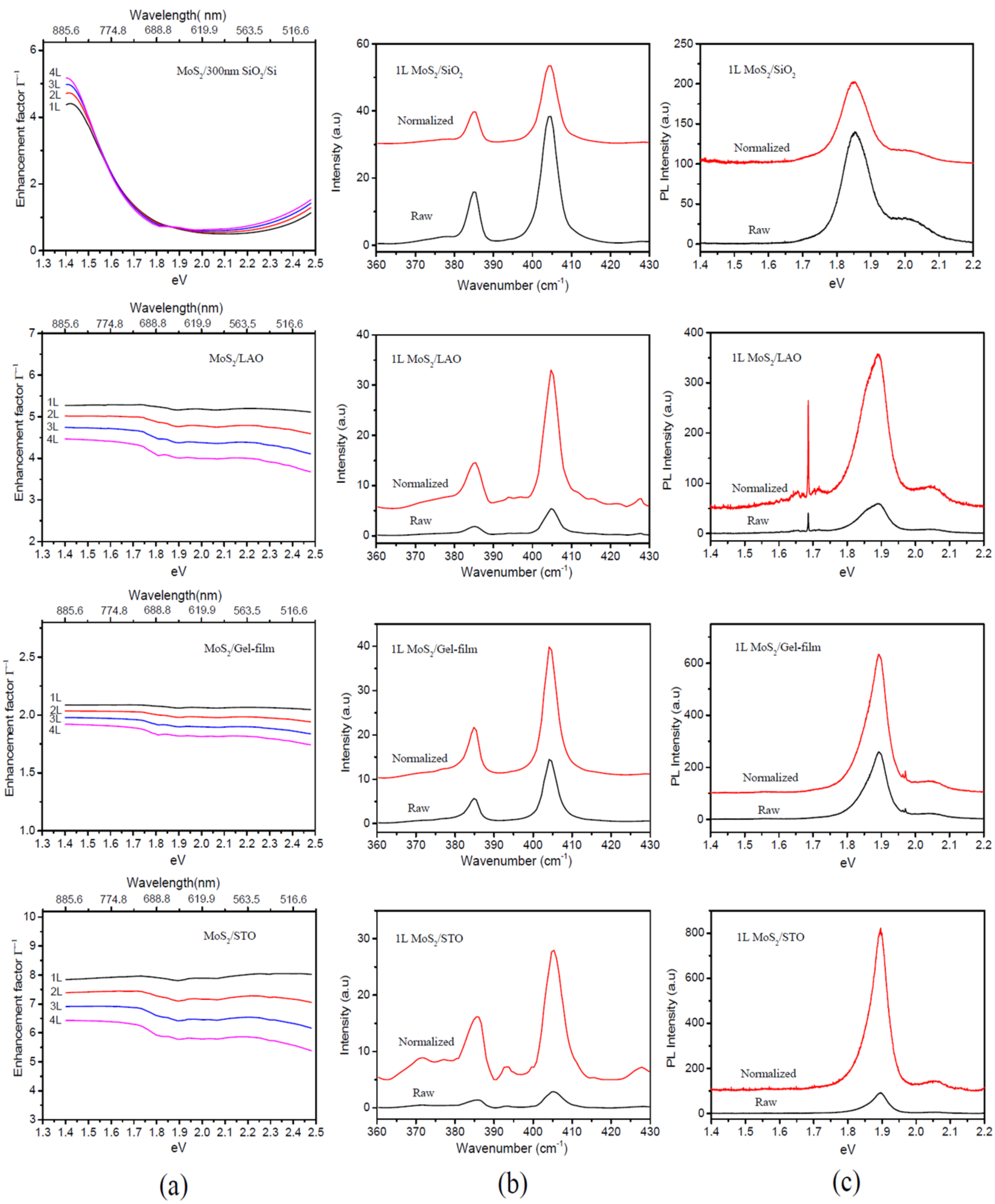

Figure.S1. (a).The calculated enhancement factor of monolayer to 4-layer MoS_2 on SiO_2, LAO,Gel-film and STO substrates. (b). The raw and normalized Raman spectra of monolayer MoS_2 on different substrates . (c). The raw and normalized PL spectra of monolayer MoS_2 on different substrates.

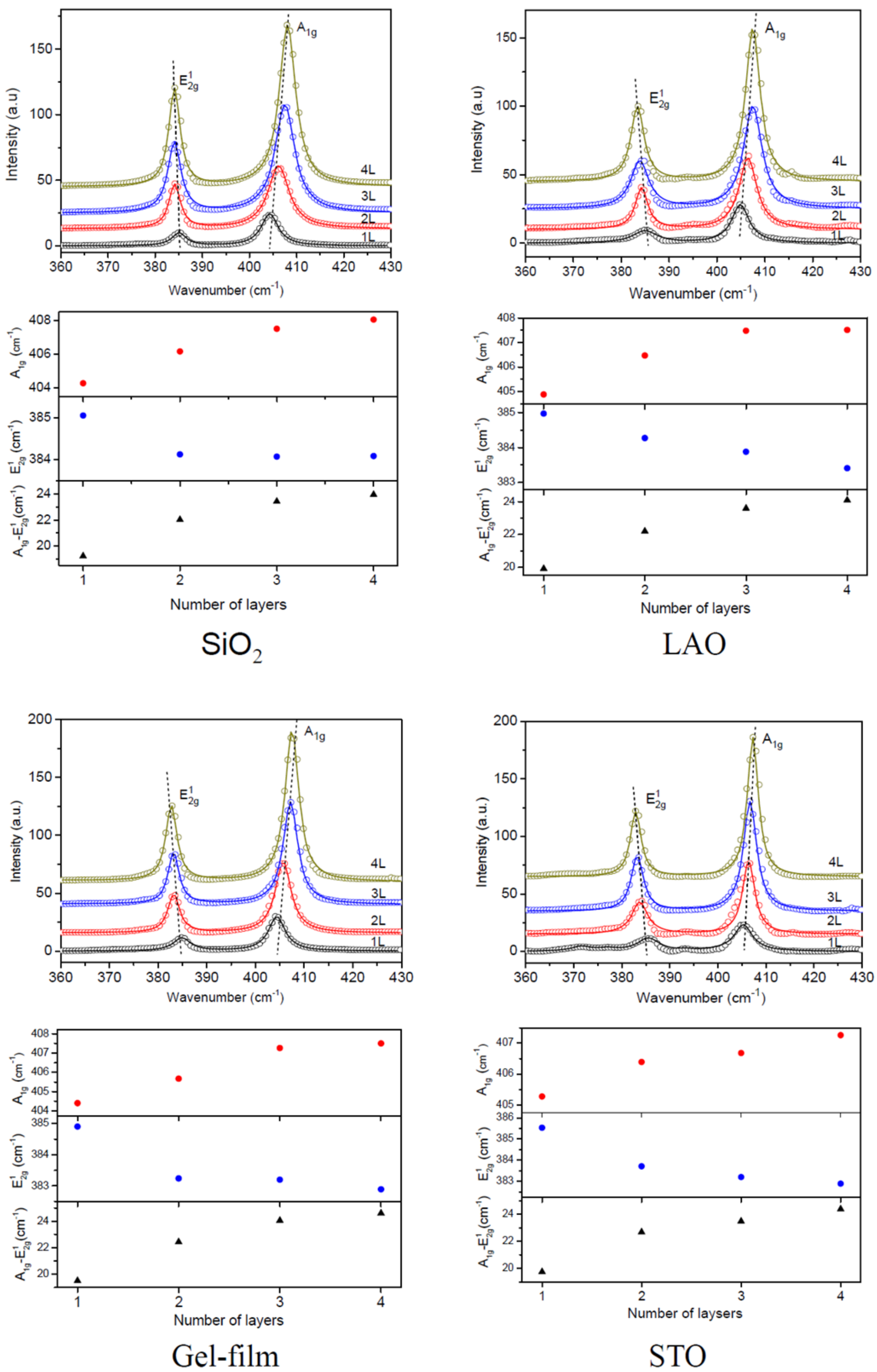

Figure S1. Normalized Raman spectra, frequencies of the A_{1g} and $E^1{}_{2g}$ modes and the difference between the two modes of monolayer to 4-layer MoS_2 on SiO_2, LAO, STO and Gel-film substrates.

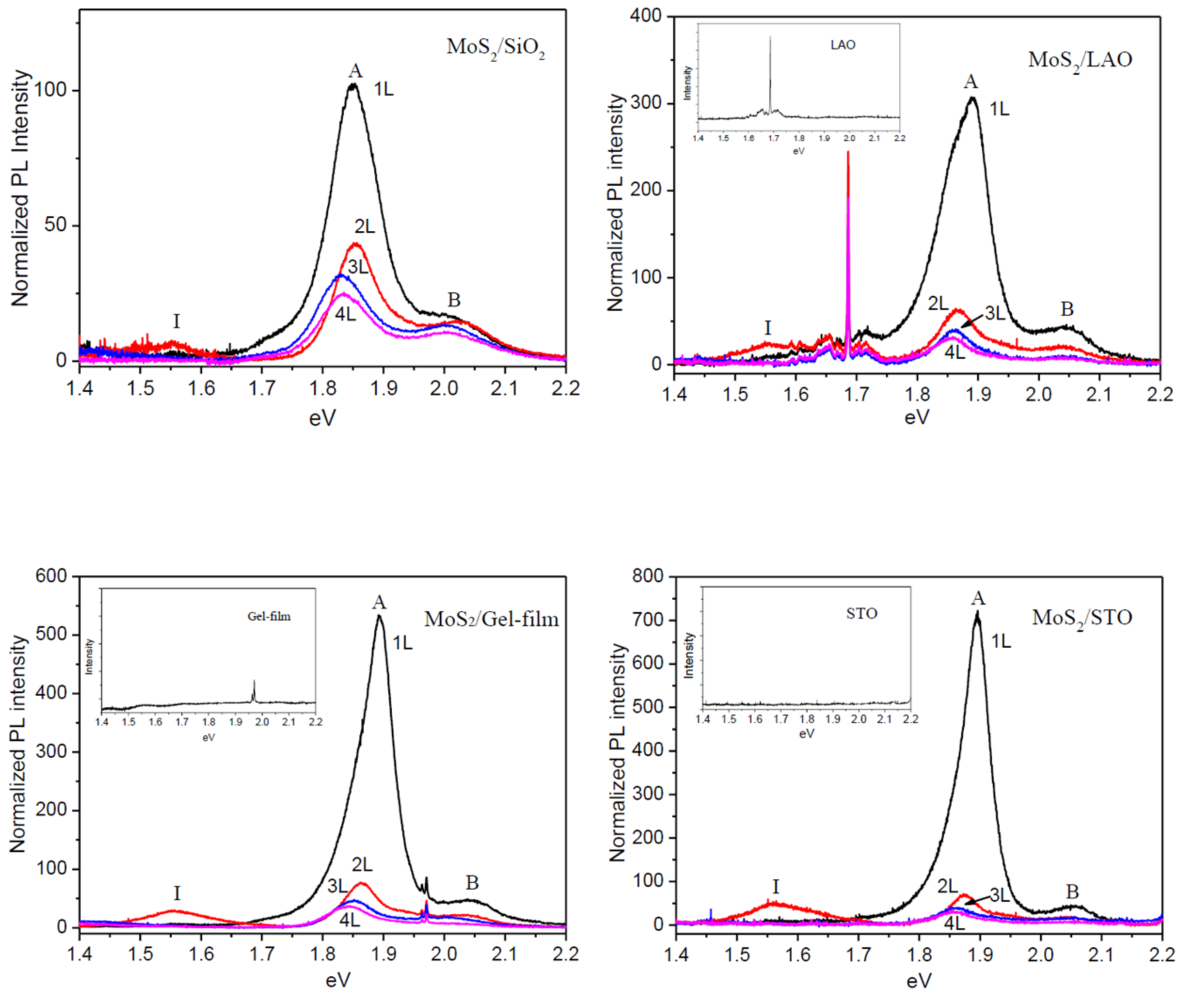

Figure S3. Thickness-dependent of the normalized PL spectra of MoS_2 on SiO_2, LAO, Gel-film and STO substrates. The inserts are PL spectra of substrates.

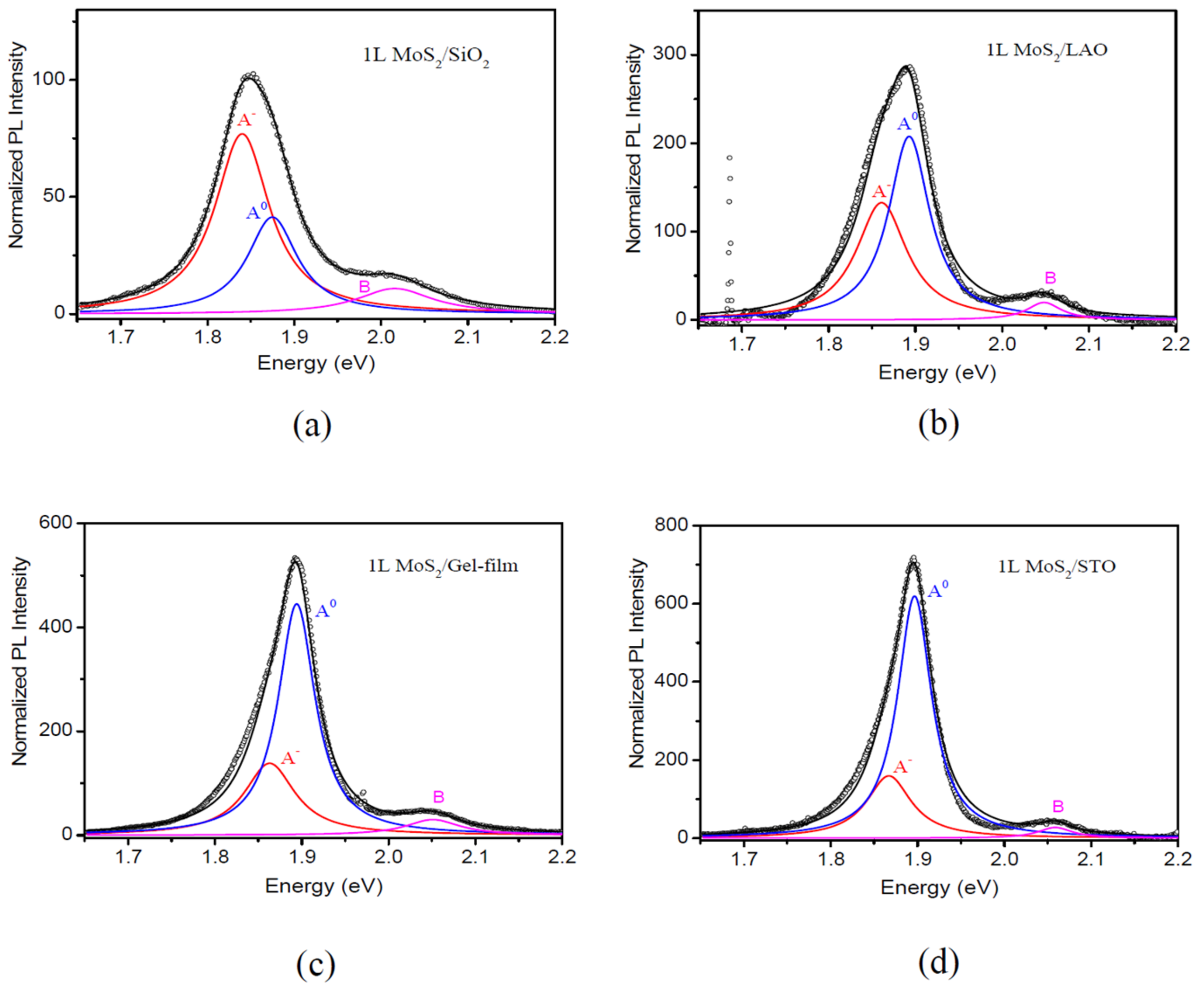

Figure.S4. The fitting of PL spectra of monolayer MoS_2 on SiO_2 (a), LAO (b),Gel-film (c),STO (d) substrates. The open circle symbols are experimental data. The thin solid lines are the Lorentz functions that compose the fit.