Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

First-principles Raman spectra of MoS_2, WS_2 and their heterostructures

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1 The influence of laser frequency on the intensity ratio

Note that in the non-resonant first-order Raman scattering, the laser frequency ω_i only appears in the prefactor $(\omega_i \mp \omega_j)^4$ in Equation 1 in the main text, and ω_i is typically in the range 18797-30769 cm^{-1} (laser wavelength typically in the range 325-532 nm), much larger than frequencies ω_j of E^1_{2g} and A_{1g} modes (between 358 and 423 cm^{-1}). Thus, the prefactor $(\omega_i \mp \omega_j)^4 \approx \omega_i^4$ and hence the laser frequency affects both modes identically, exerting essentially no influence on the intensity ratio.

2 Supplementary figures

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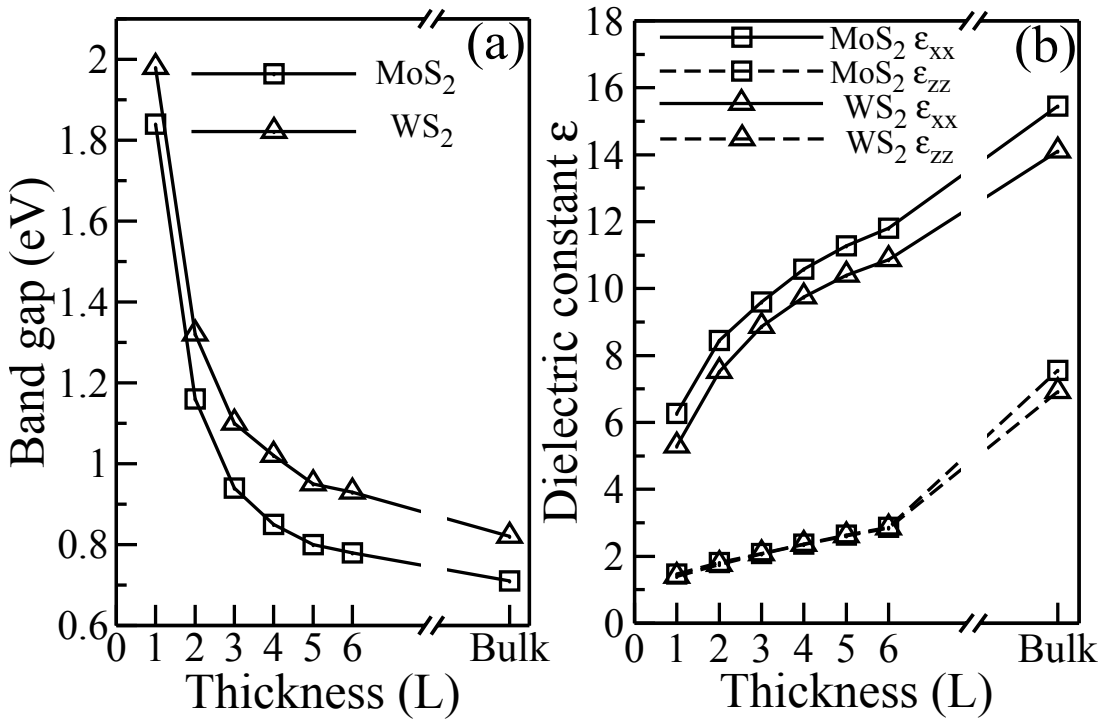

Figure S1 Thickness dependence of (a) electronic band gaps and (b) dielectric constants of MoS_2 and WS_2. For dielectric tensor ε, non-diagonal elements are zero and $\varepsilon_{xx} = \varepsilon_{yy}$.

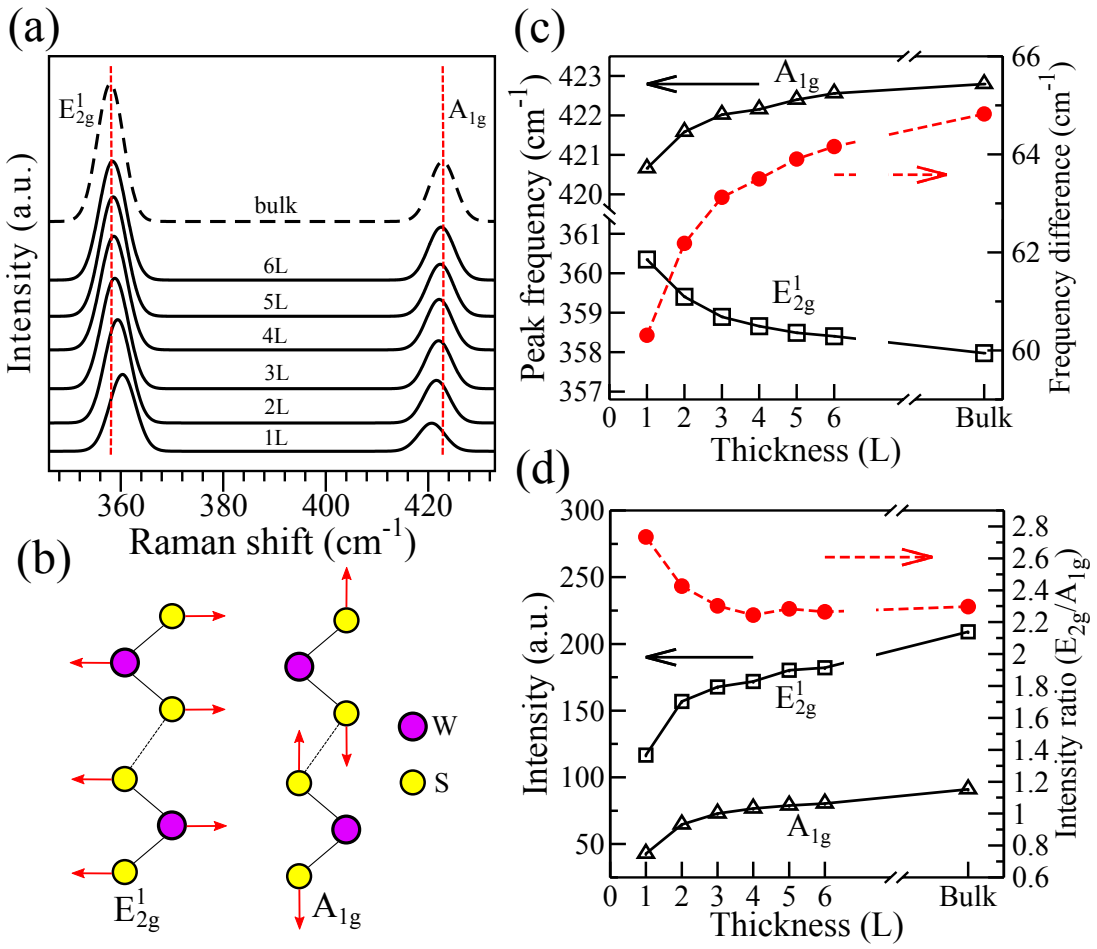

Figure S2 (Color online). (a) *Ab initio* Raman spectra of free-standing *n*-layer (*n*L) and bulk WS_2, with laser polarizations $\mathbf{g}_i$=$\mathbf{g}_s$=(1,0,0) and wavelength 488 nm. (b) Schematic of Raman-active modes E^1_{2g} and A_{1g} of bulk WS_2. (c) Frequencies of E^1_{2g} and A_{1g} (left vertical axis) and their difference (right vertical axis) as a function of layer number. (d) Peak intensities of E^1_{2g} and A_{1g} (left vertical axis) and their intensity ratio (right vertical axis) as a function of layer number.

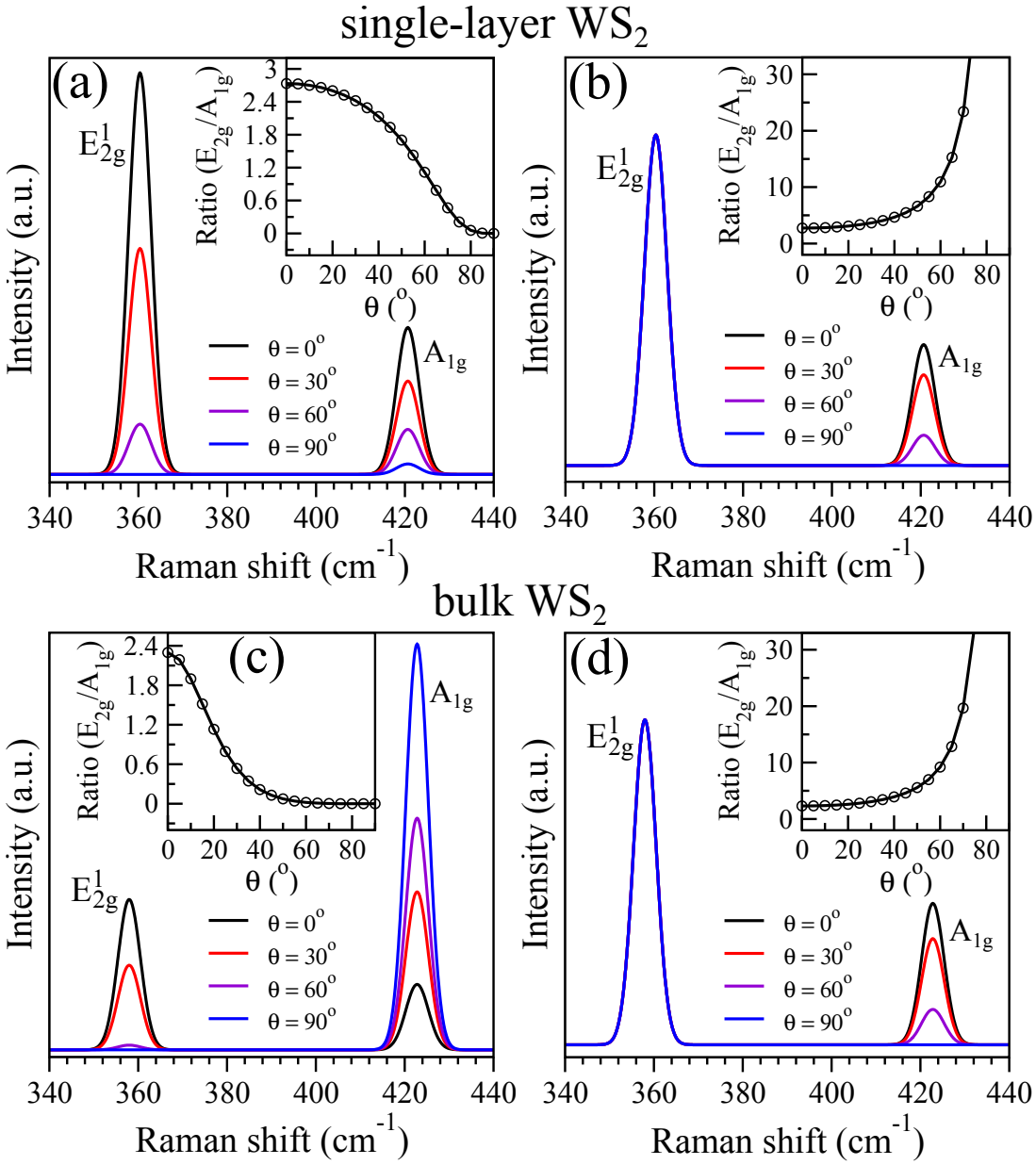

Figure S3 (Color online). Raman spectra of (a) single-layer and (c) bulk WS_2 with laser polarizations $\mathbf{g}_i=\mathbf{g}_s=(\cos\theta,0,\sin\theta)$. Raman spectra of (b) single-layer and (d) bulk WS_2 with laser polarizations $\mathbf{g}_i=(1,0,0)$ and $\mathbf{g}_s=(\cos\theta,\sin\theta,0)$. In (a) and (c), some of Raman spectra are scaled for comparison purpose. The inset figures show the intensity ratio between E^1_{2g} and A_{1g} as a function of θ.