

Electronic Supplementary Information (ESI)

Anomalous excitonic resonance Raman effects in few-layer MoS₂

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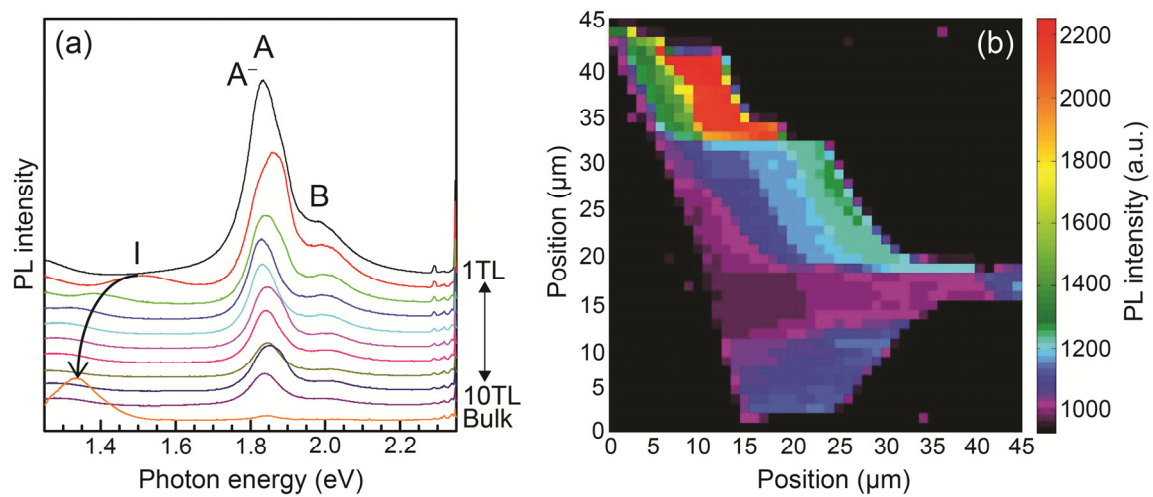


Figure S1. (a) Photoluminescence (PL) spectra of few-layer MoS₂. (b) PL intensity image of A excitation.

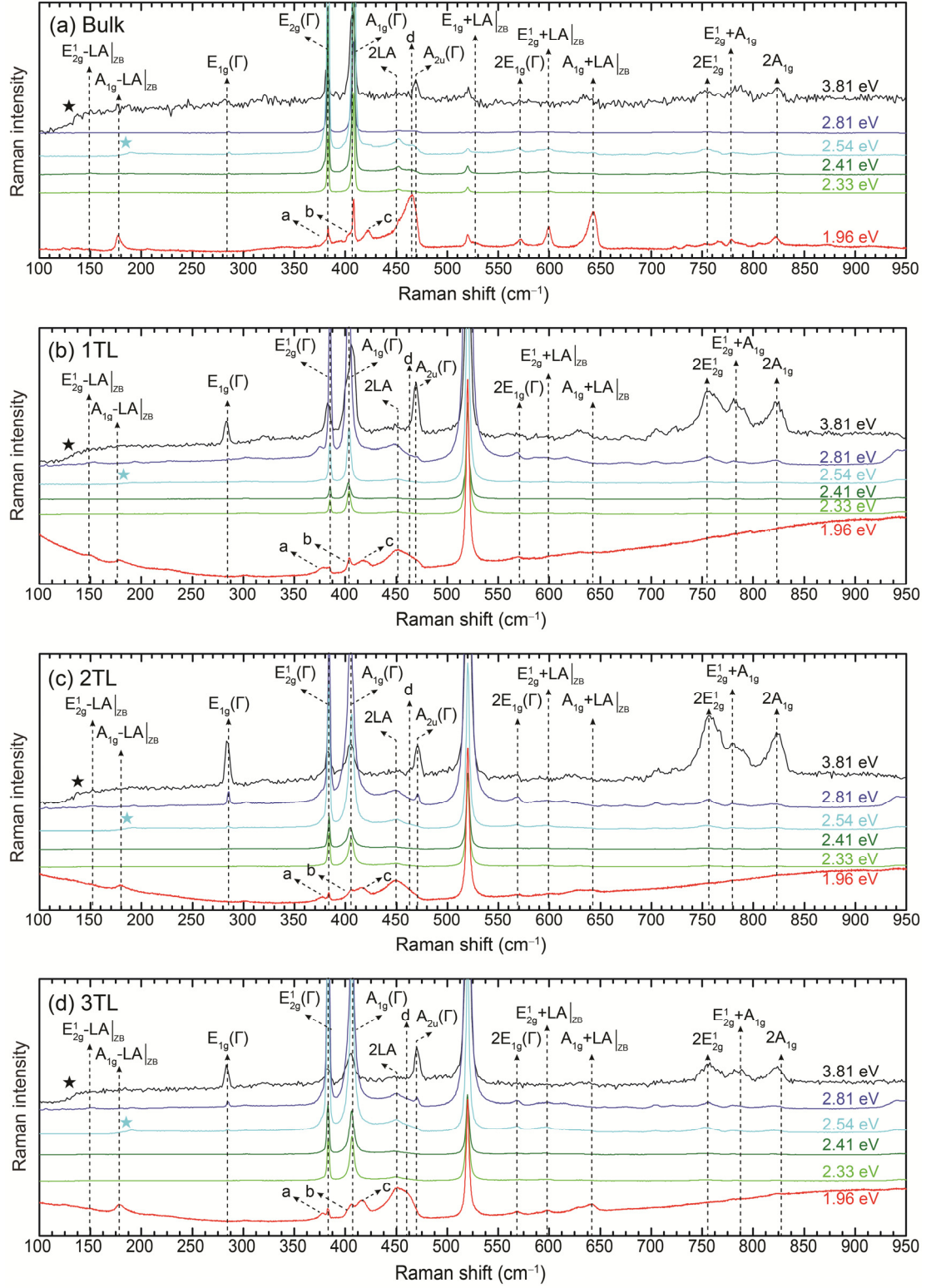


Figure S2. Raman spectra of (a) bulk, (b) 1TL, (c) 2TL, and (d) 3TL MoS₂ measured with 6 excitation energies: 3.81 eV, 2.81 eV, 2.54 eV, 2.41 eV, 2.33 eV, and 1.96 eV. Features indicated by (★) are experimental artefacts due to the cutoff of the edge filters.

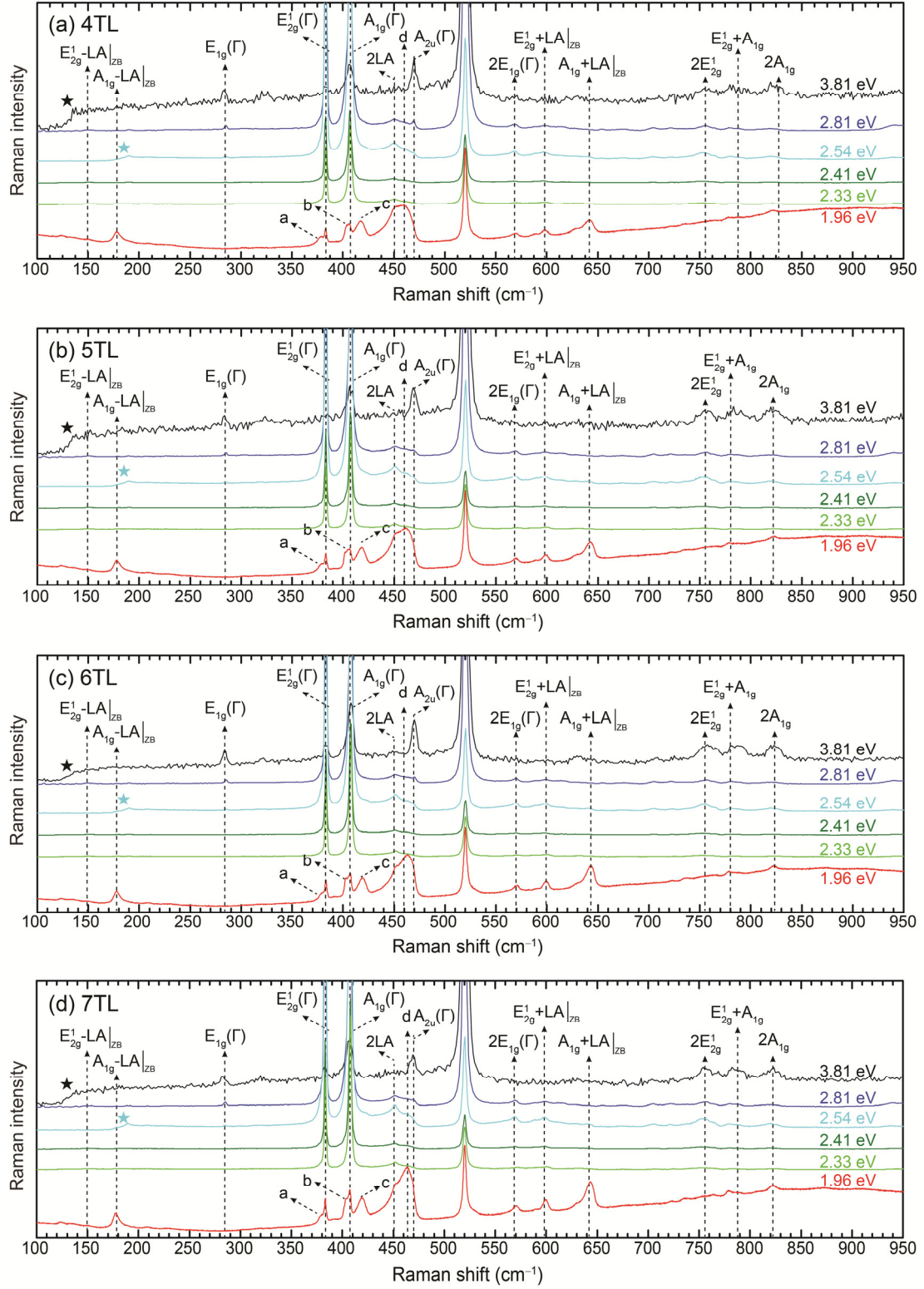


Figure S3. Raman spectra of (a) 4TL, (b) 5TL, (c) 6TL, and (d) 7TL MoS₂ measured with 6 excitation energies: 3.81 eV, 2.81 eV, 2.54 eV, 2.41 eV, 2.33 eV, and 1.96 eV. Features indicated by (★) are experimental artefacts due to the cutoff of the edge filters.

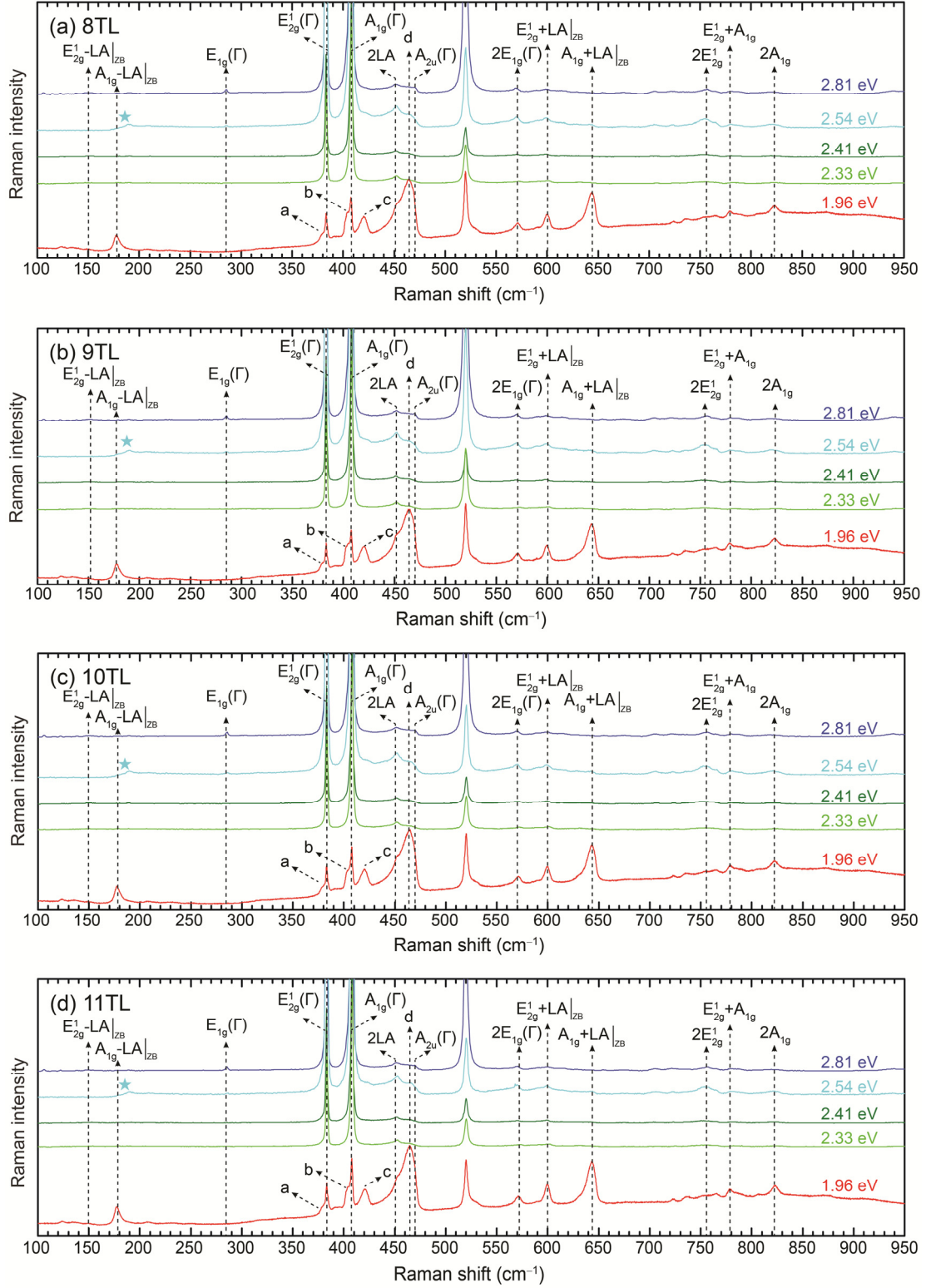


Figure S4. Raman spectra of (a) 8TL, (b) 9TL, (c) 10TL, and (d) 11TL MoS₂ measured with 5 excitation energies: 2.81 eV, 2.54 eV, 2.41 eV, 2.33 eV, and 1.96 eV. Features indicated by (★) are experimental artefacts due to the cutoff of the edge filters.

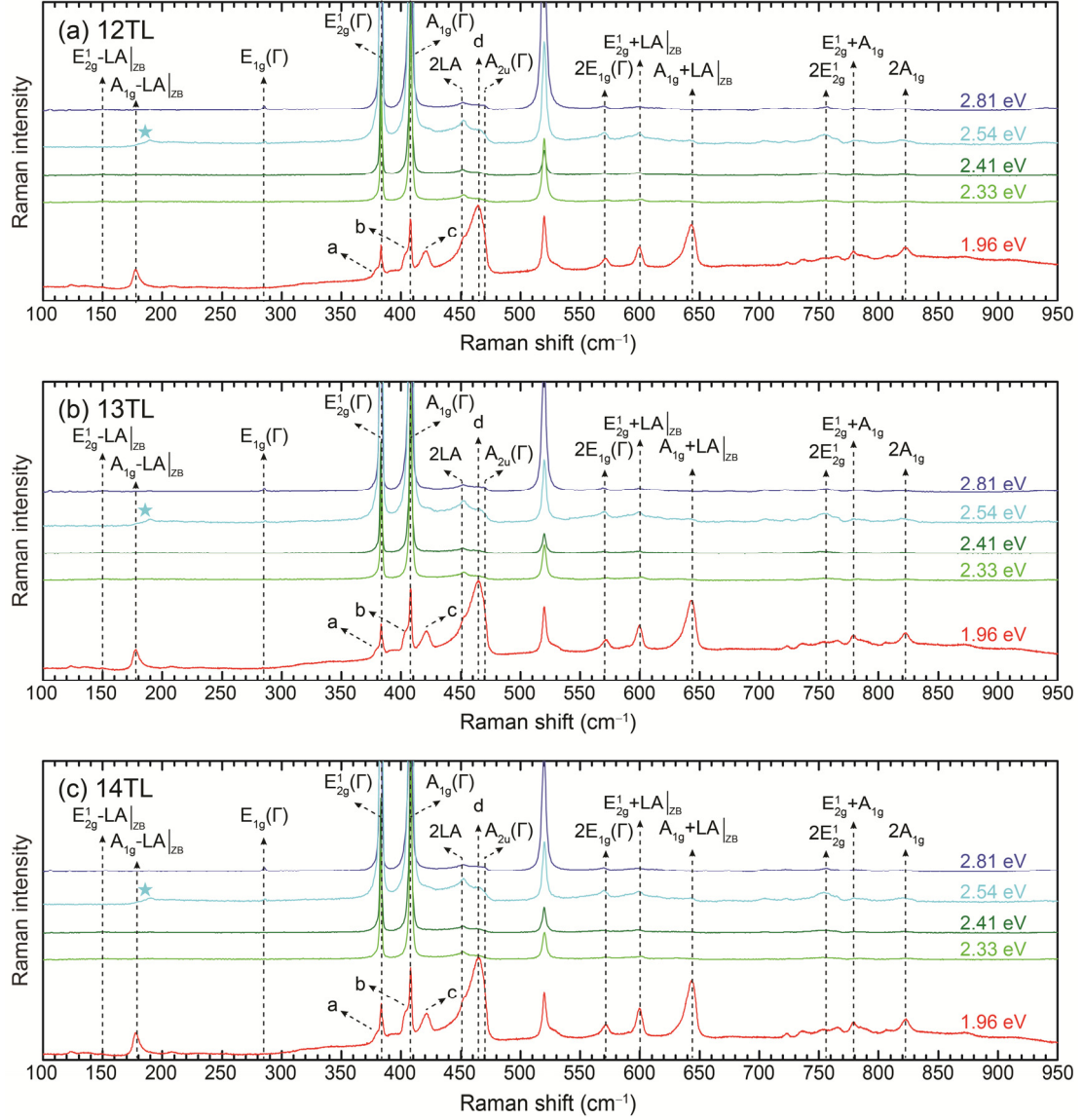


Figure S5. Raman spectra of (a) 12TL, (b) 13TL, and (c) 14TL MoS₂ measured with 5 excitation energies: 2.81 eV, 2.54 eV, 2.41 eV, 2.33 eV, and 1.96 eV. Features indicated by (★) are experimental artefacts due to the cutoff of the edge filters.

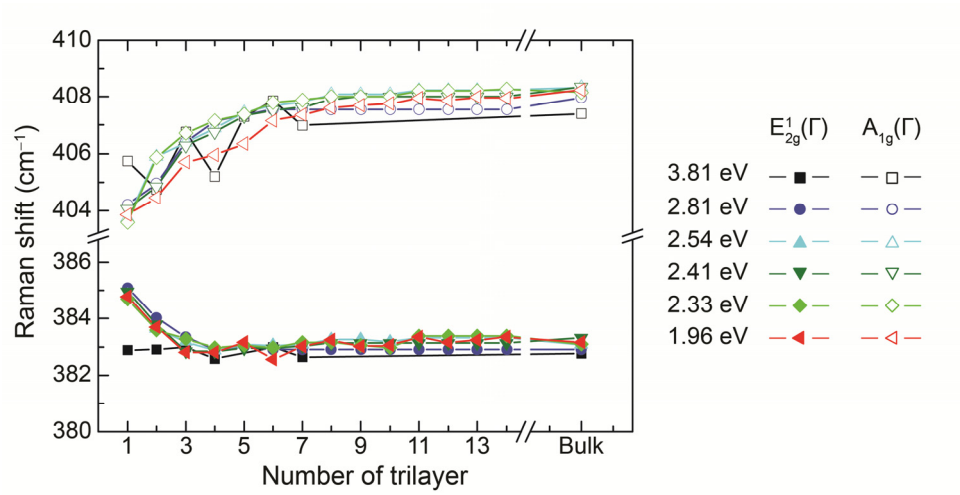


Figure S6. Peak positions of E_{2g}^1 and A_{1g} modes up to 14TL and bulk for 6 excitation energies. The data for the 3.81 eV excitation show larger scatter due to a lower spectral resolution at short wavelengths.

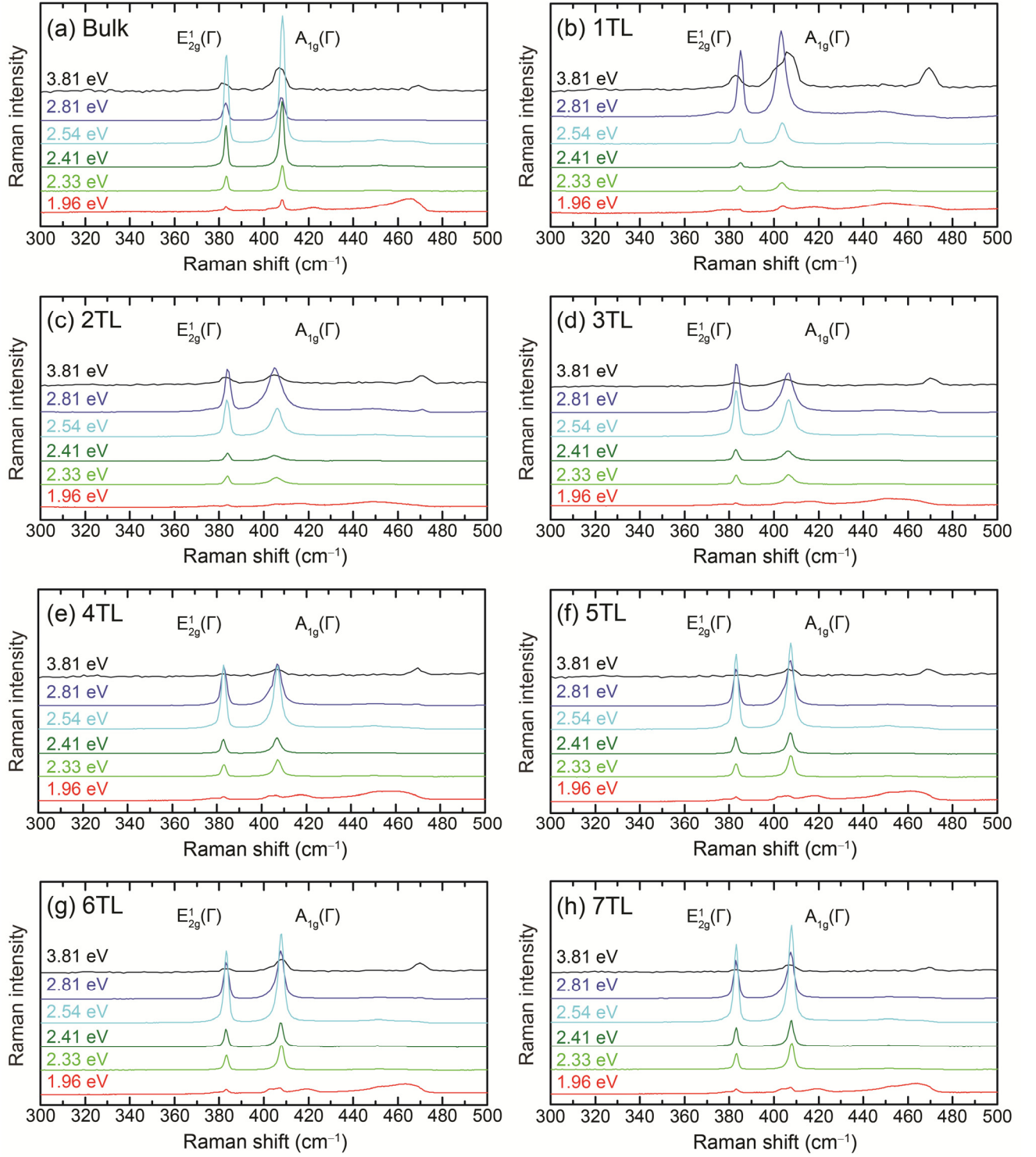


Figure S7. Dependence of E_{2g}^1 and A_{1g} modes on excitation energies for (a) bulk and (b) 1TL – (h) 7TL MoS₂.

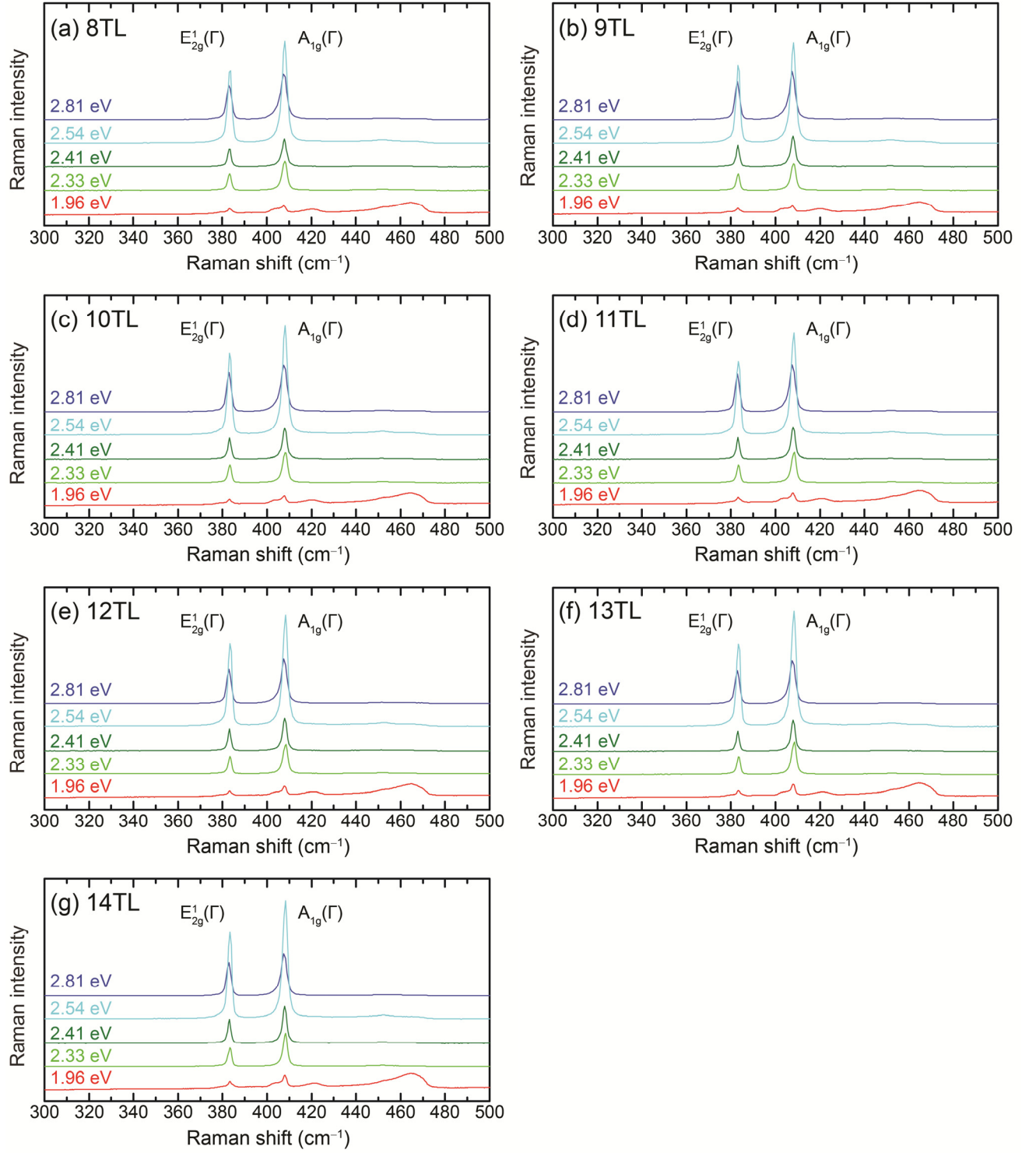


Figure S8. Dependence of E_{2g}^1 and A_{1g} modes on excitation energies for (a) 8TL – (g) 14TL MoS₂.

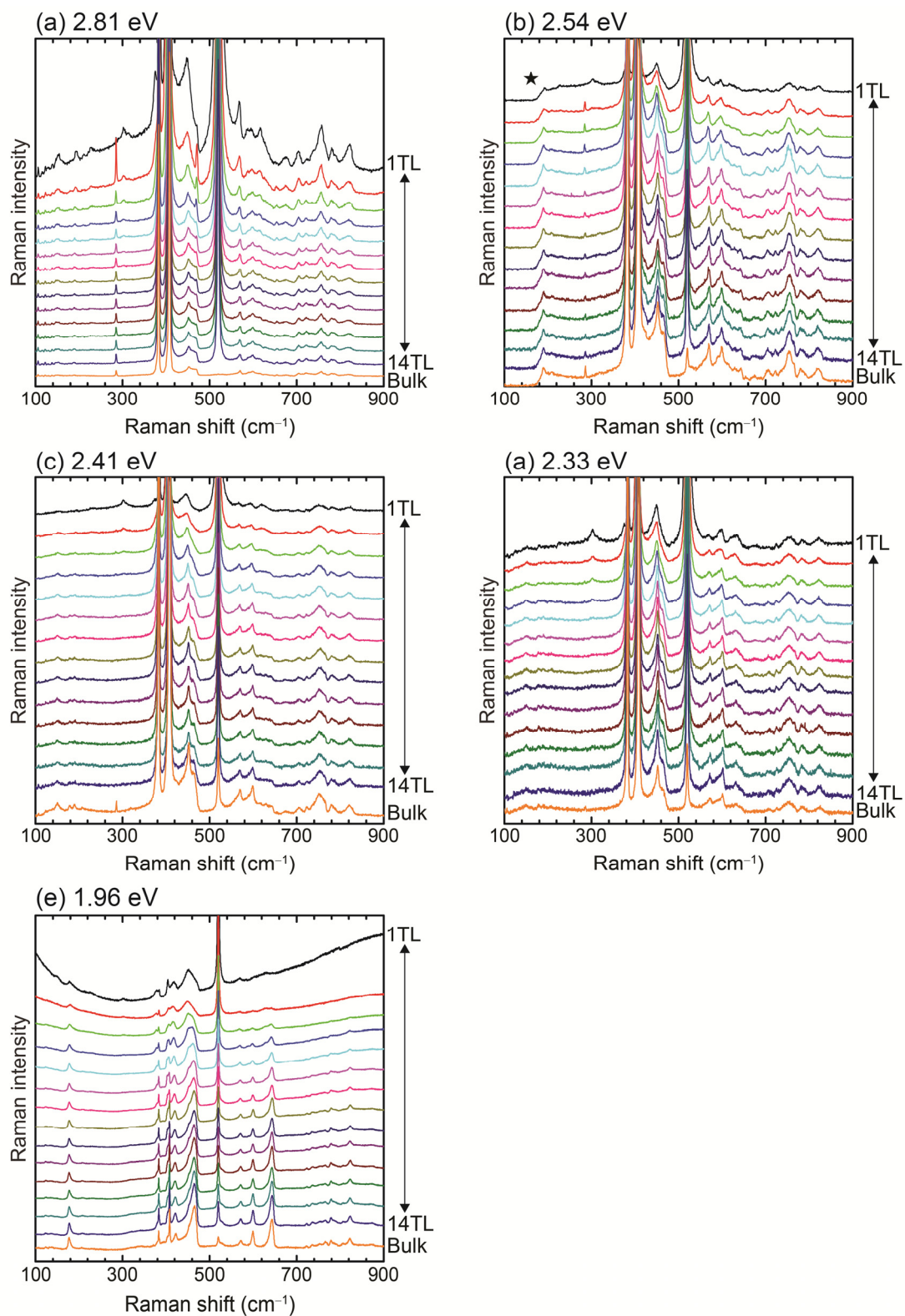


Figure S9. Thickness dependence of the Raman spectrum of MoS₂ measured with 5 excitation (a) 2.81 eV, (b) 2.54 eV, (c) 2.41 eV, (d) 2.33 eV, and (e) 1.96 eV. Features indicated by (★) are experimental artefacts due to the cutoff of the edge filters.

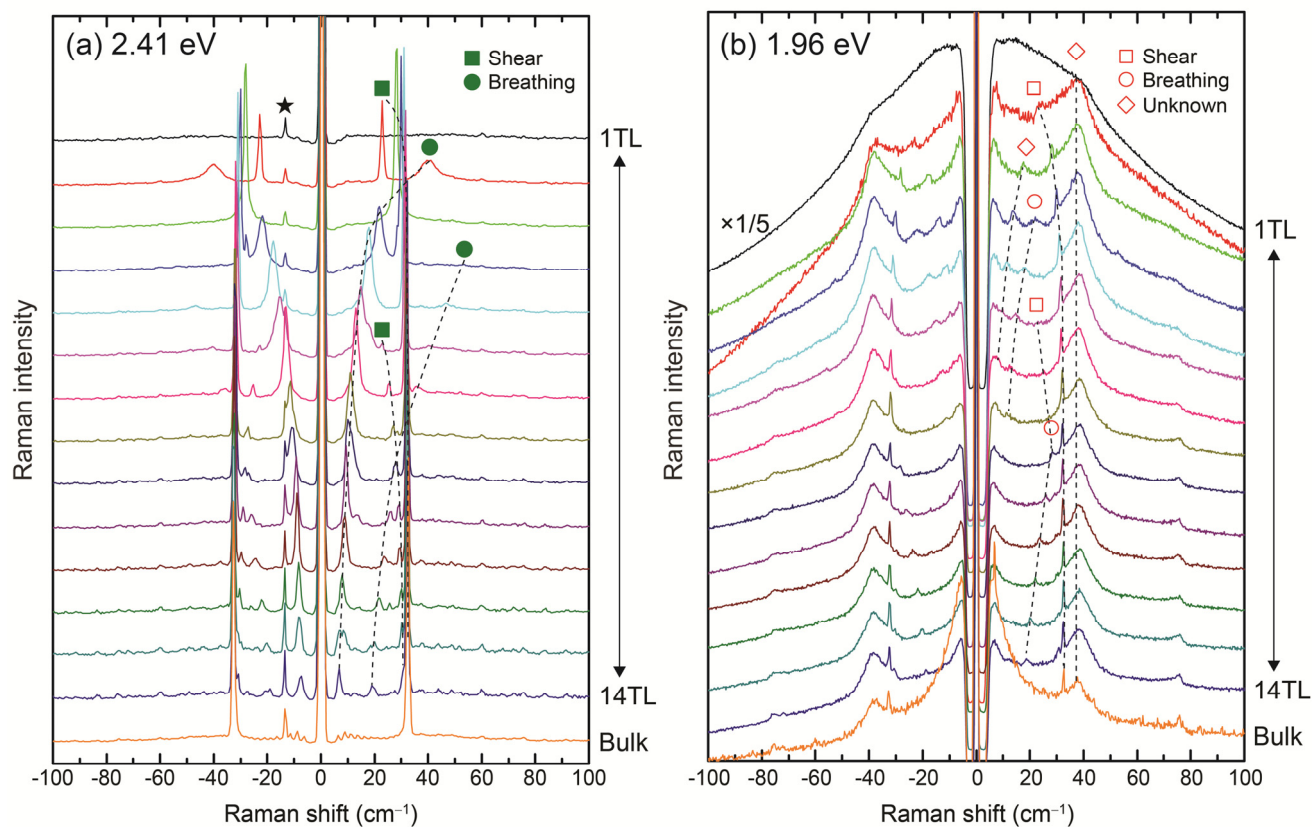


Figure S10. Low-frequency Raman spectra of MoS₂ measured with excitation energies of (a) 2.41 eV and (b) 1.96 eV. A plasma line of the 2.41-eV laser is indicated by (★).

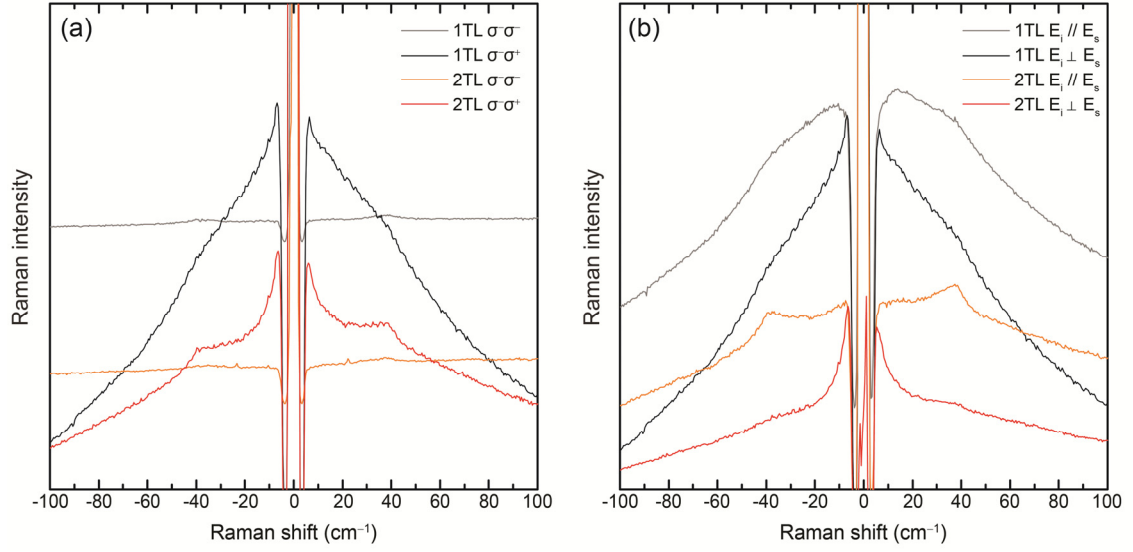


Figure S11. Polarization dependence of the low-frequency Raman spectra of 1TL and 2TL MoS₂ measured with excitation energy of 1.96 eV: (a) circular and (b) linear polarization dependence. In backscattering geometry, $(\sigma^-\sigma^+)$ or $(\sigma^+\sigma^-)$ correspond to spin-conserving scattering. (a) shows that the ‘central peak’ is due to a spin-conserving scattering mechanism.

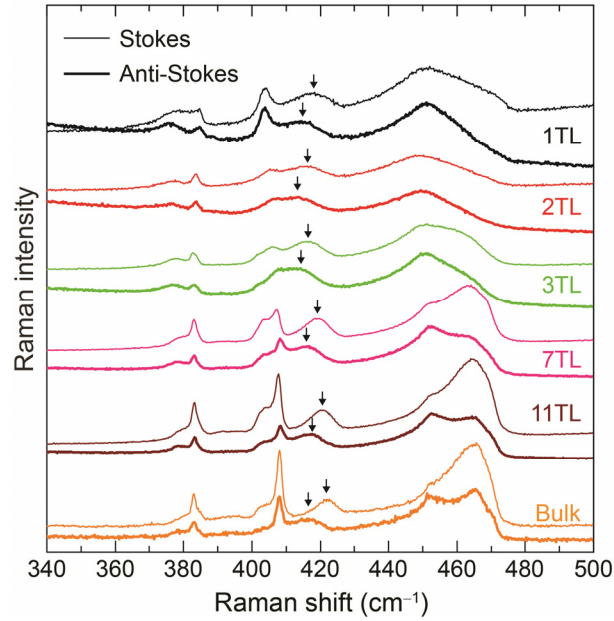


Figure S12. Comparison of Stokes and anti-Stokes Raman spectra of few-layer MoS₂ for an excitation energy of 1.96 eV. The peak *c* indicated by arrows exhibits a clear displacement between Stokes and anti-Stokes scattering.