Supporting Information for:

Unveiling the Complex Phonon Nature and Phonon Cascades in 1L to 5L WSe_2 Using Multiwavelength Excitation Raman Scattering

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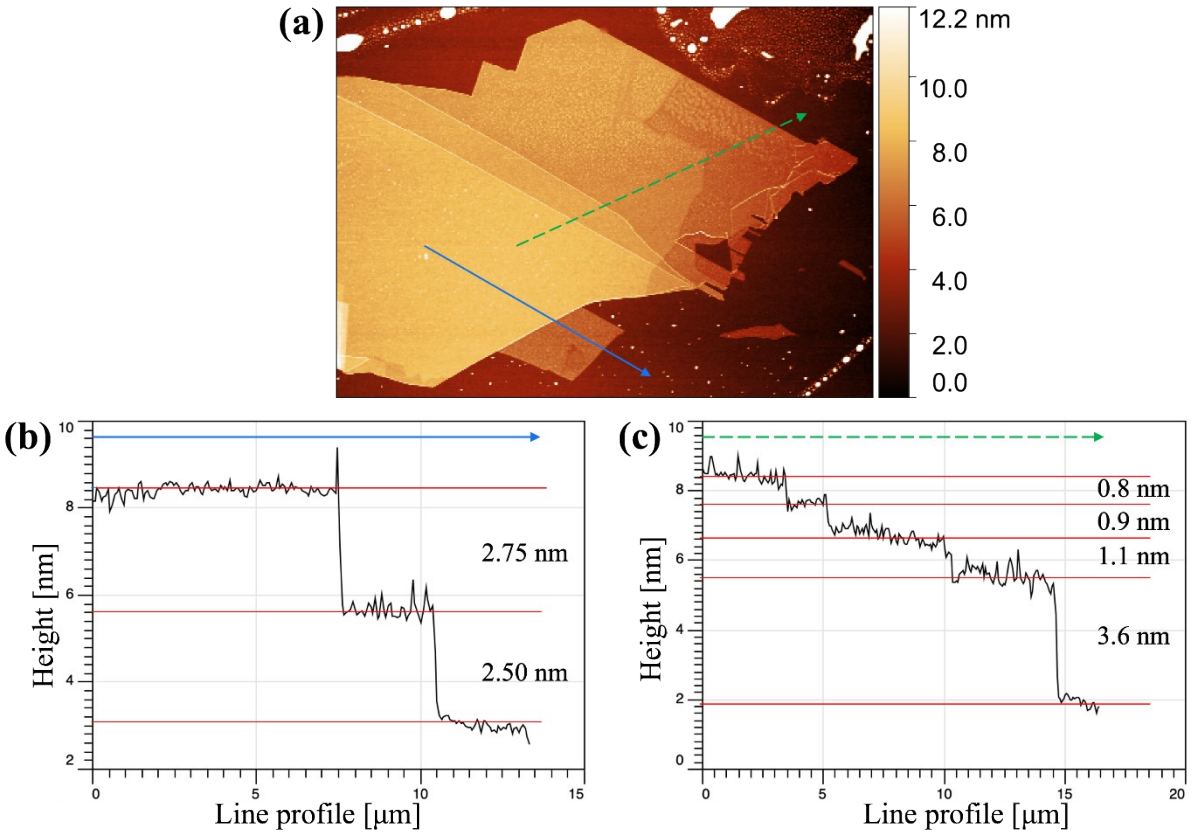

Figure S1. AFM characterization of WSe_2 flake. (a) AFM image of the WSe_2 flake measured in tapping-mode showing indicating to line profiles which were used for the identification of the number of layers. (b) and (c) AFM line profiles showing differences in the layer thicknesses and allowing identification of 1L to 5L layers.

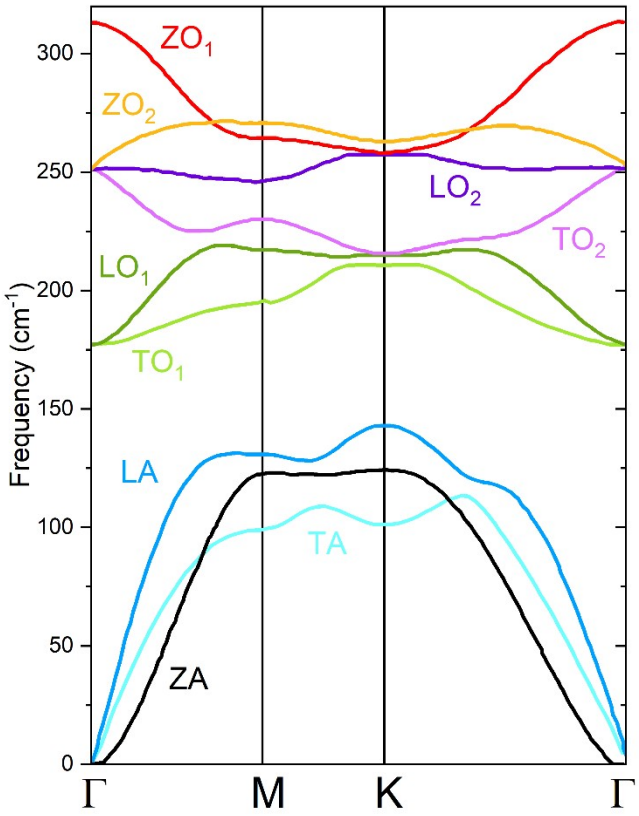

Figure S2. Density Functional Theory (DFT) calculated phonon dispersion of monolayer WSe2 adapted from Ref. [1], with labeled phonon branches. Monolayer WSe_2 has three atoms per unit cell, resulting in total of nine phonons. The three acoustic phonons correspond to the out of plane (ZA) vibrations, in-plane transverse (TA) vibrations, and in-plane longitudinal (LA) vibrations. These phonons are located at 0 cm^{-1} at the Γ point and correspond to the lowest frequency phonon branches across the Brillouin zone. The E" and E' phonon modes are located at 176 and 250 cm^{-1} at the Γ point, and as doubly degenerated modes, are split into two optical branches each, over the Brillouin zone. The lower frequency phonon branches correspond to transverse (TO_1 and TO_2) vibrations, while the higher frequency phonon branches correspond to longitudinal (LO_1 and LO_2) vibrations of E" and E' modes, respectively. The A_1' and A_2" modes are located at 250 and 310 cm^{-1} at the Γ point and correspond to two out of plane (ZO_1 and ZO_2) vibrations, respectively. They are represented with the highest frequency phonon branches.

[1] M. D. Luca, X. Cartoixà, J. Martín-Sánchez, M. López-Suárez, R. Trotta, R. Rurali and I. Zardo, New insights in the lattice dynamics of monolayers, bilayers, and trilayers of WSe2 and unambiguous determination of few-layer-flakes' thickness, *2D Mater.*, 2020, **7**, 025004.

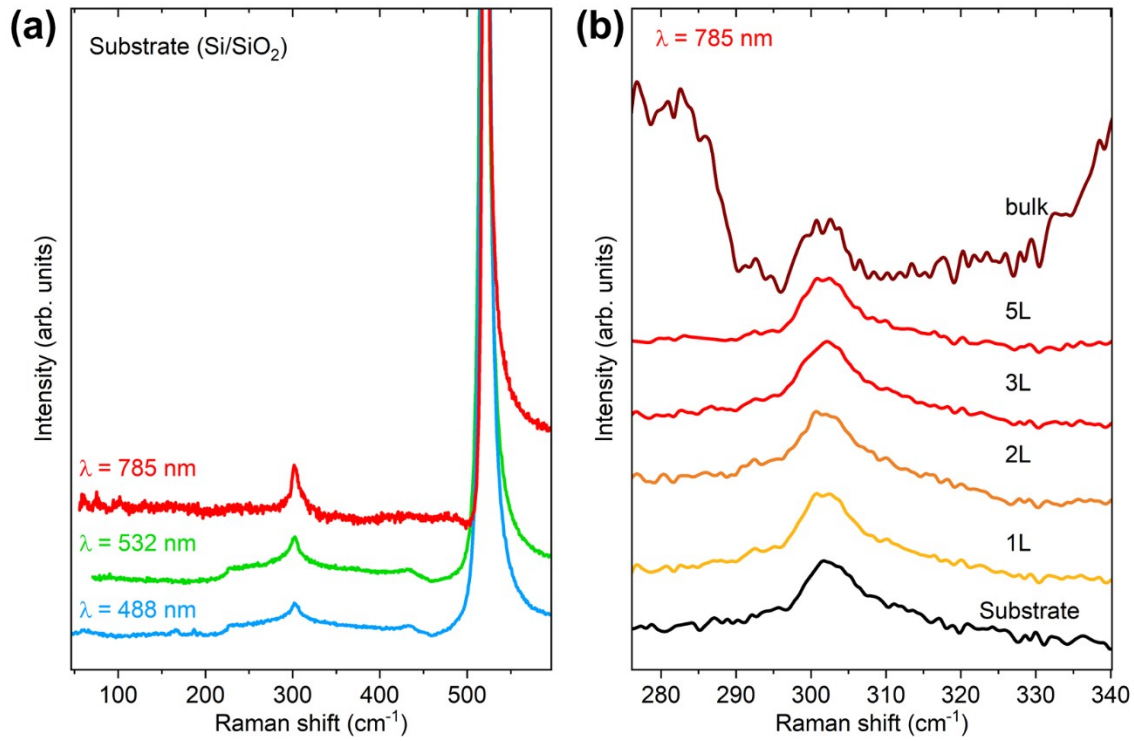

Figure S3. (a) Raman spectra of bare Si/SiO_2 substrate measured with 488, 532 and 785 nm laser excitations allowing easier discrimination of Raman peaks attributed to the substrate. (b) Comparison of 302 cm^{-1} Raman peak of 1L to 5L and bulk WSe_2, to bare Si/SiO_2 substrate. All Raman spectra were measured with 785 nm laser excitation and normalized to Si (520 cm^{-1}) peak.

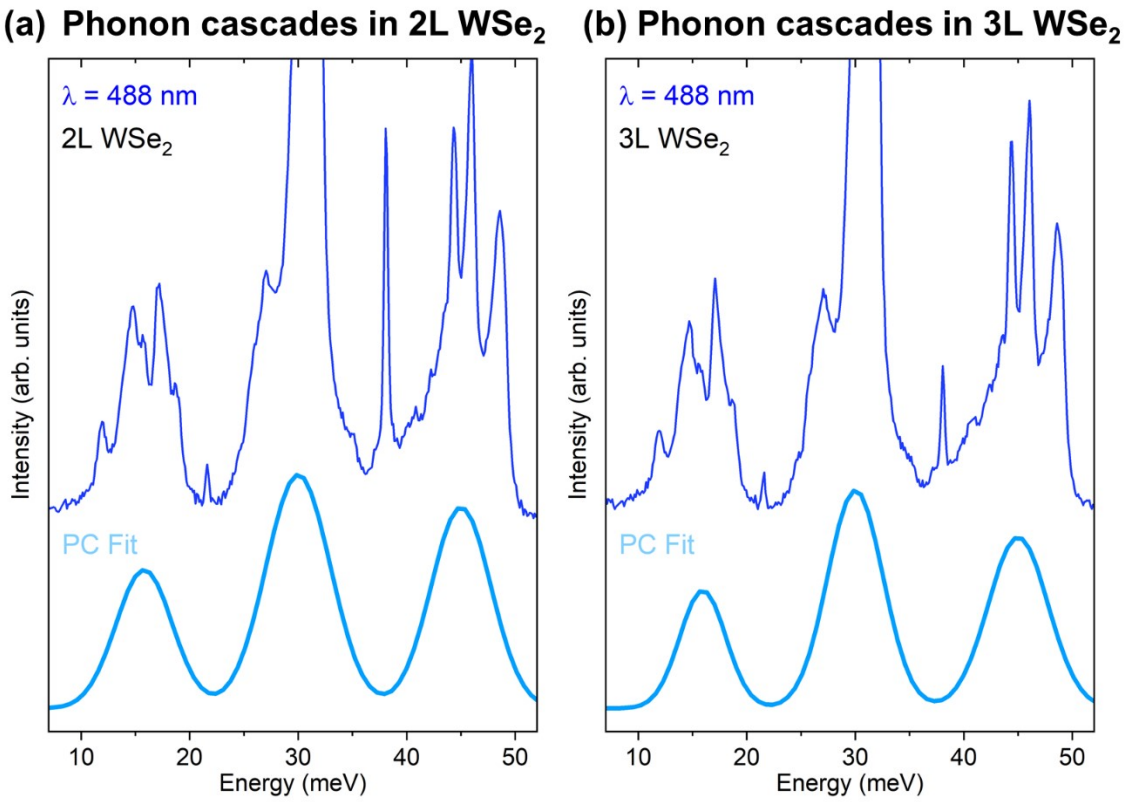

Figure S4. Raman spectra of **(a)** 2L and **(b)** 3L WSe_2 measured with 488 laser excitation, with phonon cascade fit (labeled as PC Fit) revealing 3 periodic Gaussian peaks, labeled as N_{PC} = 1, 2 and 3, at about 16, 31 and 46 meV, respectively.