Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

"Coherent Control of Interlayer Vibrations in Bi_2Se_3 van der Waals Thin-Films"

Tae Gwan Park[1], Hong Ryeol Na[2], Seung-Hyun Chun[2], Won Bae Cho[3], Sunghun Lee[2,*] and Fabian Rotermund[1,*]

[1]Department of Physics, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea

[2]Department of Physics and Astronomy, Sejong University, Seoul 05006, Korea

[3]Welfare & Medical ICT Research Department, Electronics and Telecommunications Research Institute (ETRI), Daejeon 34129, Korea

[*]Correspondence to: kshlee@sejong.ac.kr (S.L.) and rotermund@kaist.ac.kr (F.R.)

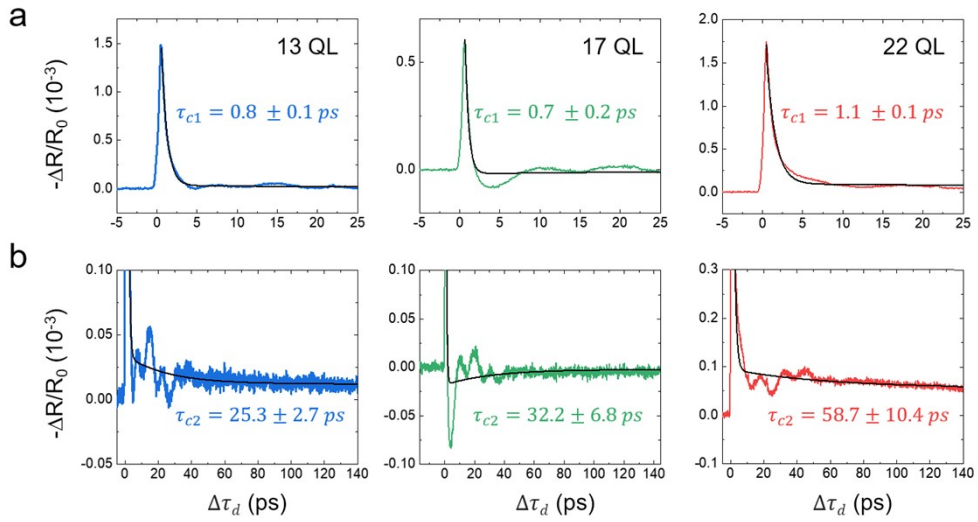

Fig. S1. Transient reflectivity (TR) signals in 13, 17 and 22 QL Bi_2Se_3. The TR traces are shown at **(a)** short and **(b)** long range of the time delay for a convenient display showing fast and slow decay components. The obtained decay constants are listed in each plot.

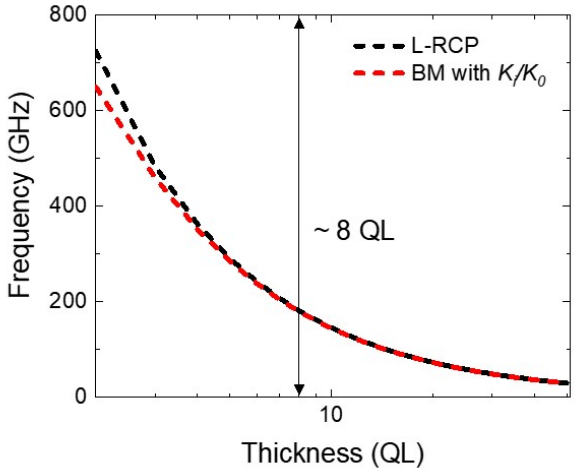

Fig. S2. Calculated frequencies of breathing mode (BM) and longitudinal resonant coherent phonon (L-RCP) mode without substrate coupling effect (free standing).

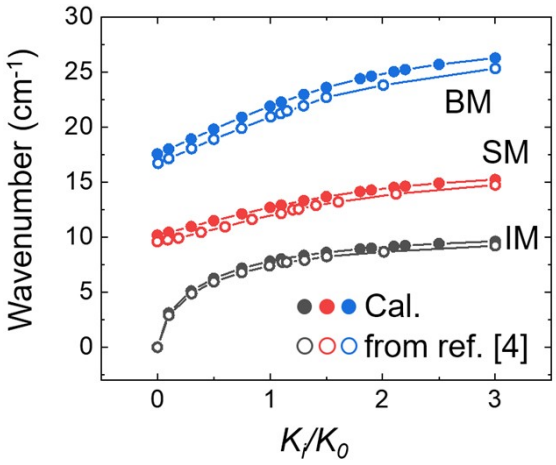

Fig. S3. Comparison of the calculated interlayer eigenmodes of 3 QL Bi_2Te_3 based on the modified linear chain model including substrate effect with DFT results taken from Ref. 1 in a unit of wavenumber (cm^{-1}). The parameters of mass density[1] ($\mu = 7.5 \times 10^{-6}$ kgm^{-2}) and out-of- and in-plane force constants[1] ($K_z = 8.21 \times 10^{19}$ Nm^{-3} and $K_x = 2.76 \times 10^{19}$ Nm^{-3}) of Bi_2Te_3 were used in the calculation.

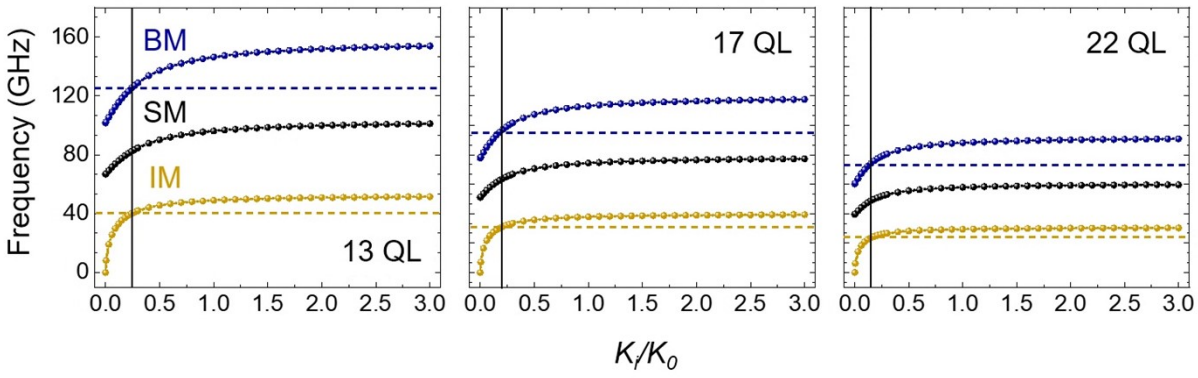

Fig. S4. Calculated frequencies of interface mode (IM), shear mode (SM), and breathing mode (BM) according to K_i/K_0. The horizontal dashed lines indicate the observed frequencies for fast (royal color) and slow modes (orange), respectively. The vertical lines denote the intersectional line for K_i/K_0 that simultaneously satisfies the BM and IM frequencies with the experimental values of fast and slow modes.

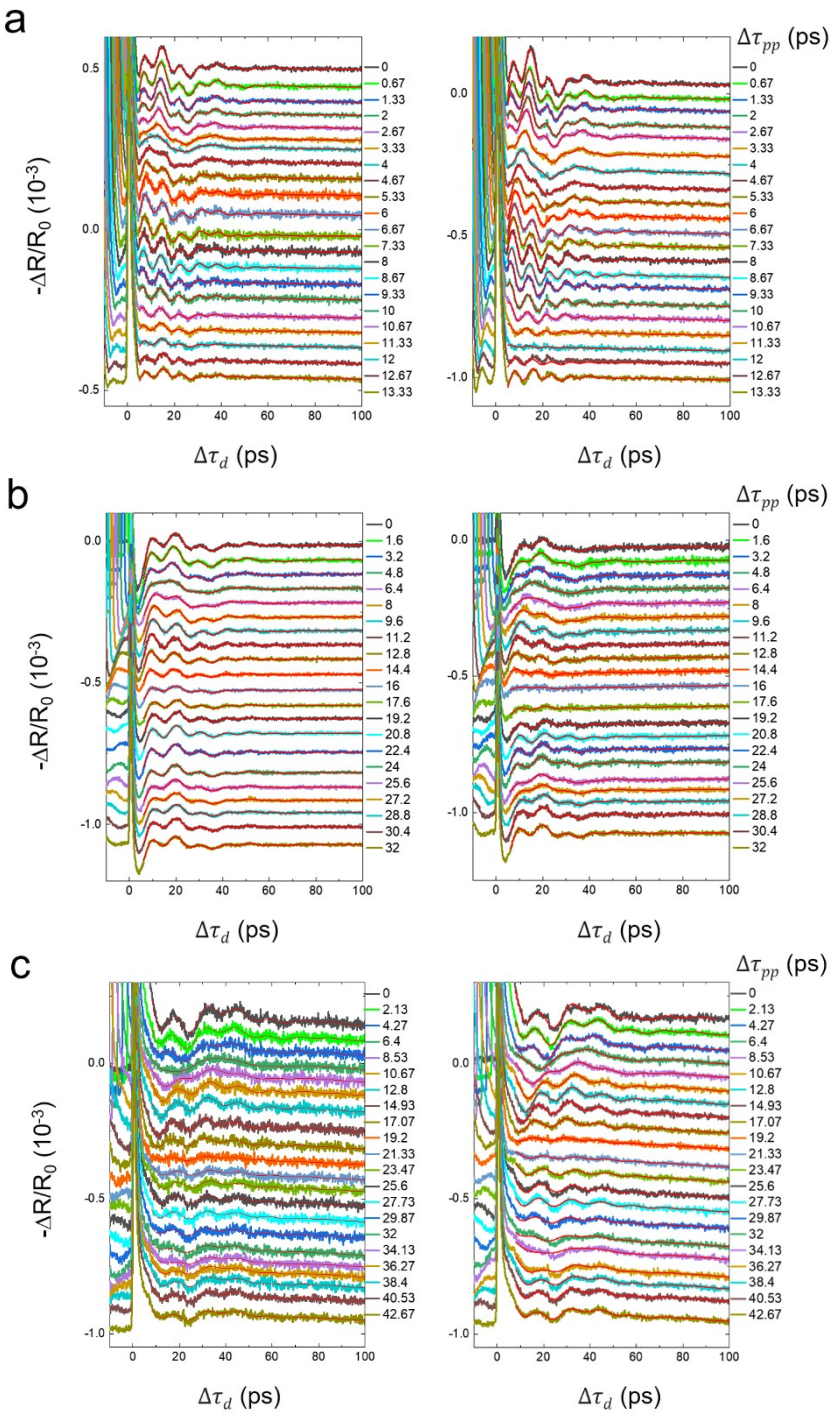

Fig. S5. Coherently controlled interlayer vibration traces in **(a)** 13, **(b)** 17, and **(c)** 22 QL Bi_2Se_3 at various delays between two pump pulses ($\Delta\tau_{pp}$). The red curves are fitting results based on Equation (1) in the main text.

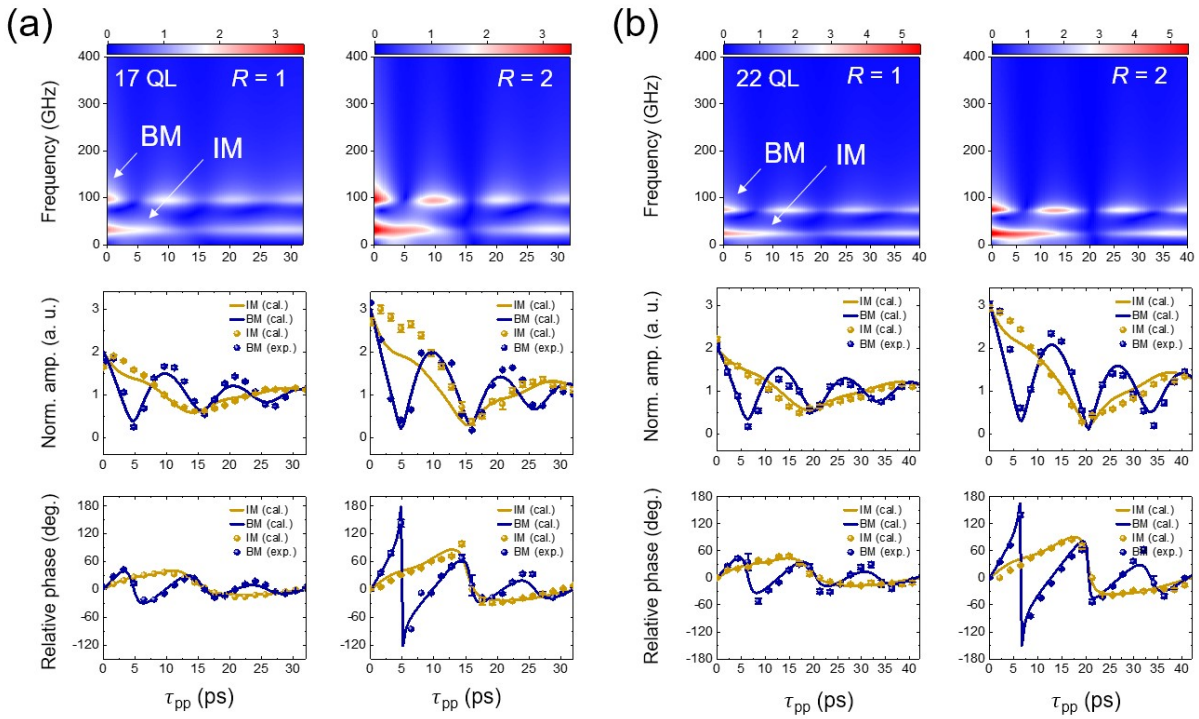

Fig. S6. Comparison of the simulated spectral responses of controlled interlayer vibrations with experimental results. **(a, b)** Simulated results based on coherent control of interlayer vibrations of 17 and 22 QL Bi_2Se_3 in the spectral domain. **(c, d)** Normalized amplitudes and **(e, f)** relative phases of controlled interlayer vibrations according to $\Delta\tau_{pp}$. The solid lines with beige and blue color are the simulated results taken from (a, b). The dots with the same color are experimental results obtained from Figure S2.

References

1. Zhao, Y.; Luo, X.; Zhang, J.; Wu, J.; Bai, X.; Wang, M.; Jia, J.; Peng, H.; Liu, Z.; Quek, S. Y., Interlayer Vibrational Modes in Few-Quintuple-Layer Bi_2Te_3 and Bi_2Se_3 Two-Dimensional Crystals: Raman Spectroscopy and First-Principles Studies. *Phys. Rev. B* **2014,** *90* (24), 245428.