Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information for

Valley depolarization in monolayer transition-metal dichalcogenides with zone-corner acoustic phonons

Tae-Young Jeong,[1,2†] Soungmin Bae,[3,4†] Seong-Yeon Lee,[1] Suyong Jung,[2]

Yong-Hoon Kim,[3,*] and Ki-Ju Yee[1,*]

[1]Department of Physics, Chungnam National University, Daejeon 34134, Republic of Korea

[2]Quantum Technology Institute, Korea Research Institute of Standards and Science, Daejeon 34113, Republic of Korea

[3]School of Electrical Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Korea

[4]Department of Physics, Yokohama National University, Hodogaya-ku, Yokohama 240-8501, Japan

† These authors equally contributed to this work.

*e-mail: y.h.kim@kaist.ac.kr (Y.-H.K.); kyee@cnu.ac.kr (K.J.Y.)

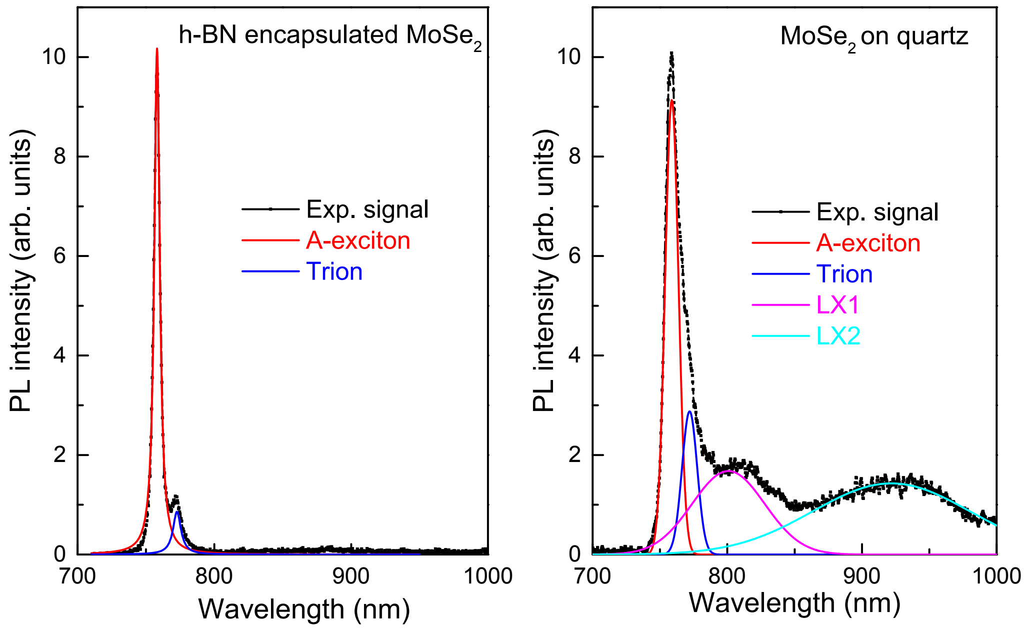

Figure S1. Photoluminescence of *h*-BN encapsulated $MoSe_2$ and bare $MoSe_2$ on quartz. Photoluminescence of an *h*-BN encapsulated monolayer $MoSe_2$ (left) and a monolayer $MoSe_2$ on quartz substrate (right) at 80 K obtained with an excitation wavelength of 532 nm. While *h*-BN encapsulated $MoSe_2$ exhibits an A-exciton at 758 nm and a trion peak at 772 nm, the $MoSe_2$ on quartz exhibits strong signals from localized excitons at 800 and 992 nm (cyan and magenta lines) as well, indicating a much greater number of defects in the bare $MoSe_2$ case.

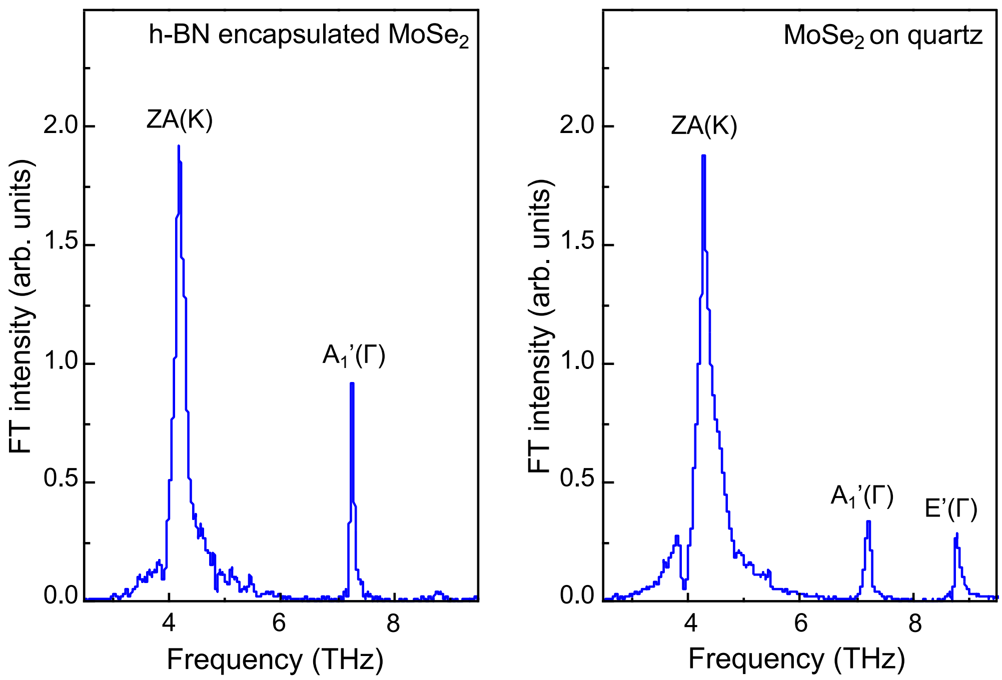

Figure S2. Coherent phonon spectrum of *h*-BN encapsulated $MoSe_2$ and bare $MoSe_2$ on quartz. FT spectrum of CP oscillation from *h*-BN encapsulated $MoSe_2$ (left) and that from bare $MoSe_2$ on quartz (right) measured using an excitation wavelength of 755 nm. Despite a distinct difference between the defect contributions in the photoluminescence signals (Fig. S1), the ZA(K) modes of the two samples are comparable here in strength. For the bare $MoSe_2$ sample, the signal at 8.77 THz, corresponding to the $E'_1(\Gamma)$ mode, likely appears owing to the defect-induced breaking of lattice symmetry.

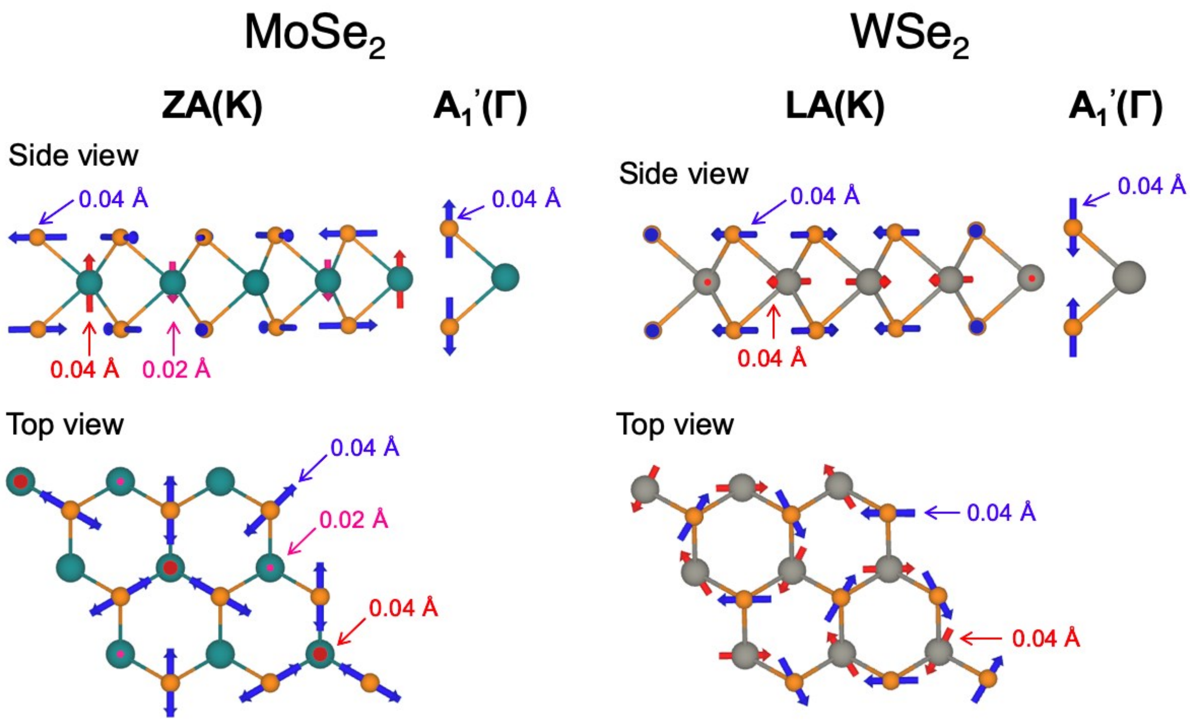

Figure S3. Atomic displacements of specific phonon modes in monolayer $MoSe_2$ and WSe_2. Phonon direction and atomic displacements of monolayer $MoSe_2$ and WSe_2 are displayed with the injection of A_1'(Γ), ZA(K) (in $MoSe_{2,}$ left), and LA(K) (in $WSe_{2,}$ right) phonons with the one phonon per formation unit.

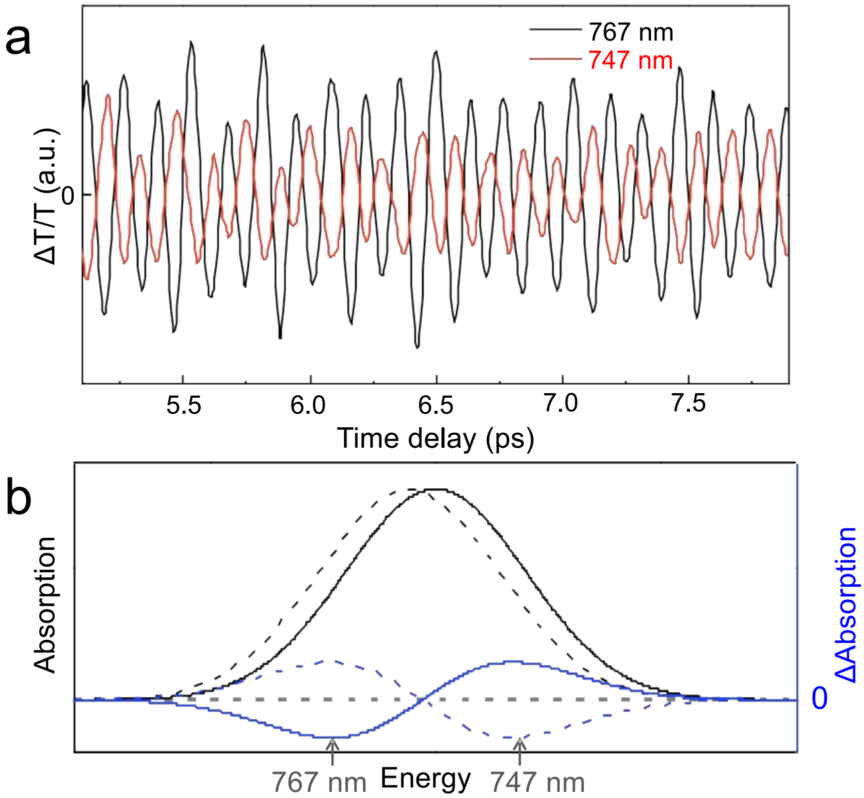

Figure S4. Energy-dependent phase change of $A_1^{'}$(Γ) phonon oscillation. (a) ΔT/T oscillations at excitation wavelengths of 767 nm and 755 nm, which are, respectively, below and above the A-exciton resonance. The apparent π-phase shift between the two wavelengths supports that energy gap modulation follows from $A_1^{'}$(Γ) phonon oscillation. (b) Schematic of the resonance shift due to atomic motion (black lines) and the absorption differences (blue lines) revealing opposite signal signs depending on whether the energy is below or above the resonance peak.

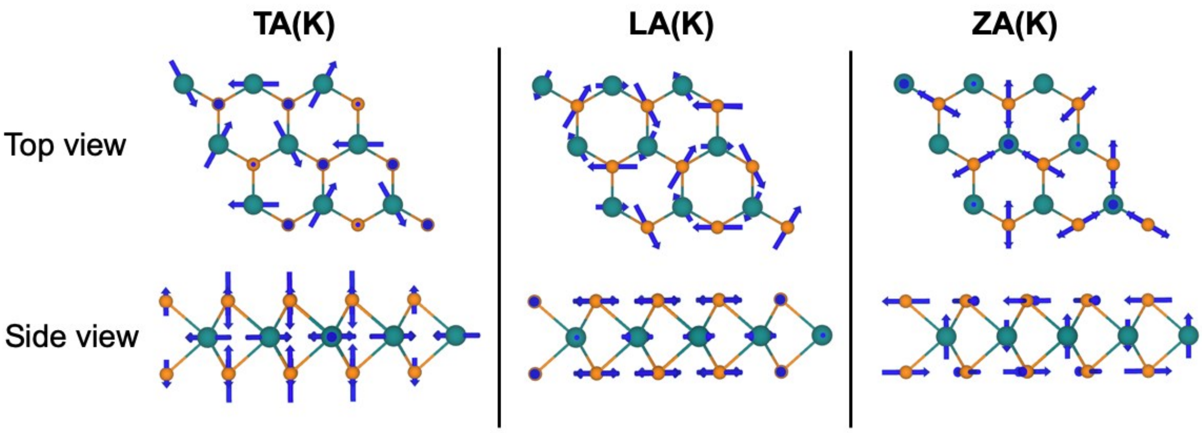

Figure S5. Symmetry of K-point acoustic phonons. Illustrations of phonon vibrations in monolayer $MoSe_2$ corresponding to the TA(K) (left), LA(K) (middle), and ZA(K) (right) modes. The green and orange circles represent Mo and Se atoms, respectively. While the TA(K) and LA(K) modes are invariant under the inversion operation with respect to the center plane, as indicated with a dotted line in the side view, the ZA(K) mode lacks such inversion symmetry.

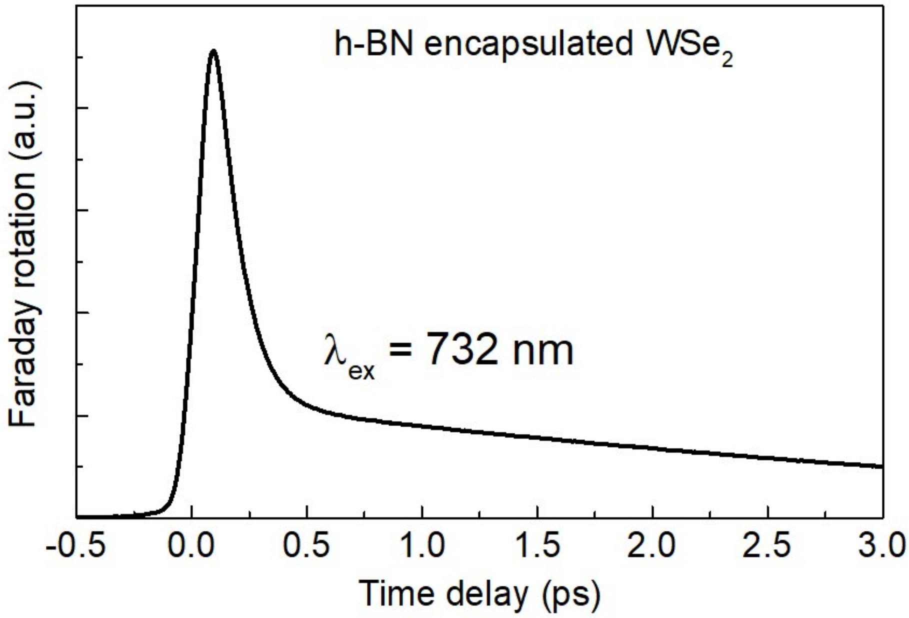

Figure S6. Valley depolarization dynamics in monolayer WSe_2. Time-resolved Faraday rotation measured at the A-exciton resonance, λ_{ex} = 732 nm, of *h*-BN encapsulated monolayer WSe_2. Bi-exponential signal fitting reveals a fast decay constant of 110 fs, which is comparable to the half-period of the LA(K) oscillation in WSe_2.

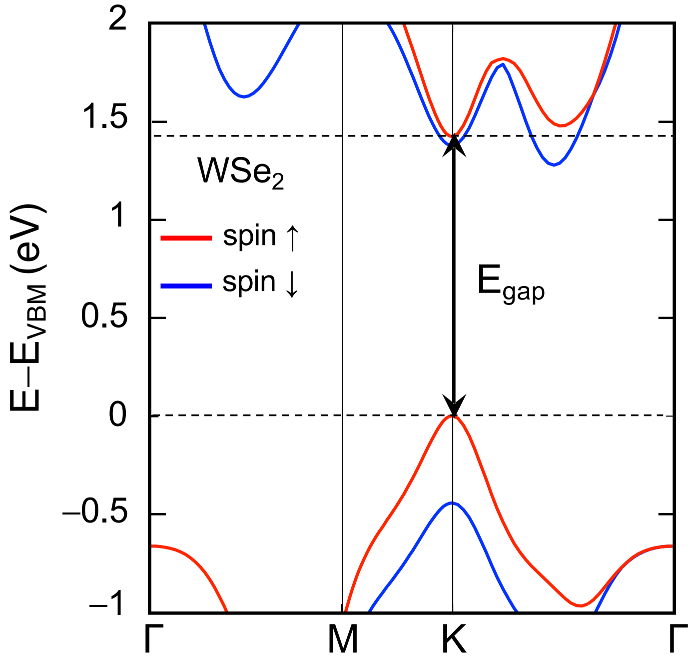

Figure S7. Electronic band structure of monolayer WSe_2. DFT calculation has been performed within the PBEsol exchange-correlation functional. The energy of bright exciton has higher energy comparing to the dark exciton, which is contrasted to the monolayer $MoSe_2$ case.

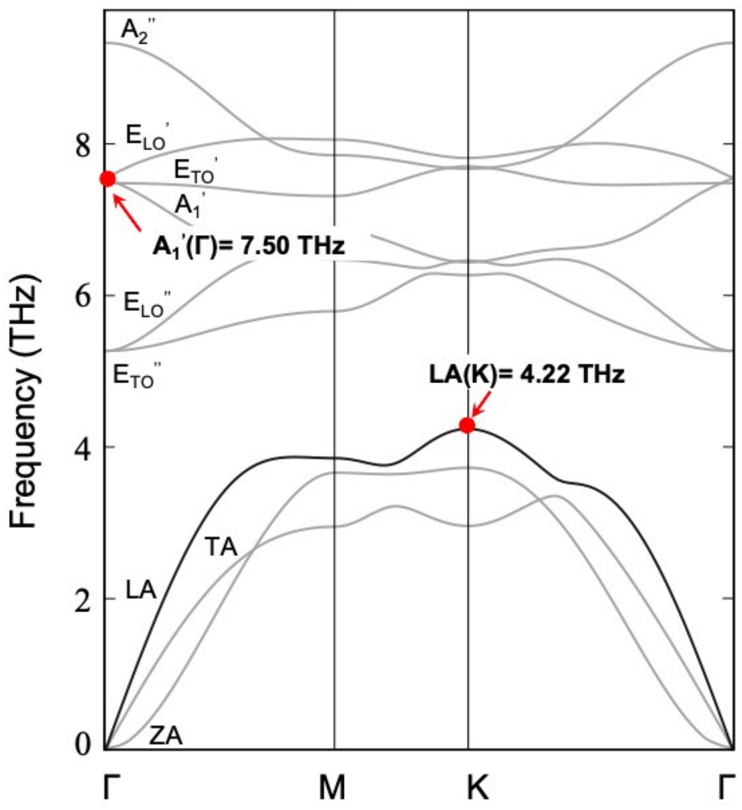

Figure S8. Phonon dispersion curve of monolayer WSe_2. Phonon dispersion curve of monolayer WSe_2 calculated using density functional theory as implemented in VASP package (DFT; PBEsol). The red dots indicate the phonon modes of $A'_1(\Gamma)$ at 7.50 THz, LA(K) at 4.22 THz, and ZA(K) at 3.69 THz.