

Supplementary Materials: Light-induced interlayer spacing dynamics via orbital phonon coupling

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Supplementary Section S1: Stacking configurations

Here, we compare the AA and AB bilayer and the AAA and ABA trilayer stackings for 1T'-transition metal dichalcogenide (TMD)s, as shown in Fig. S1. A and B denote laterally distinct layer stacking. In the schematic ABA configuration, the outer layers share a same configuration and the central layer is shifted (see Fig. S1a). Table S1 lists the relative stacking energies. A positive value of the defined energy difference indicates that AB or ABA is lower in energy than AA or AAA, respectively. This result indicates that ABA is more stable than AAA stacking. Based on this evaluation, we consider ABA stacking for the trilayer system in the main text.

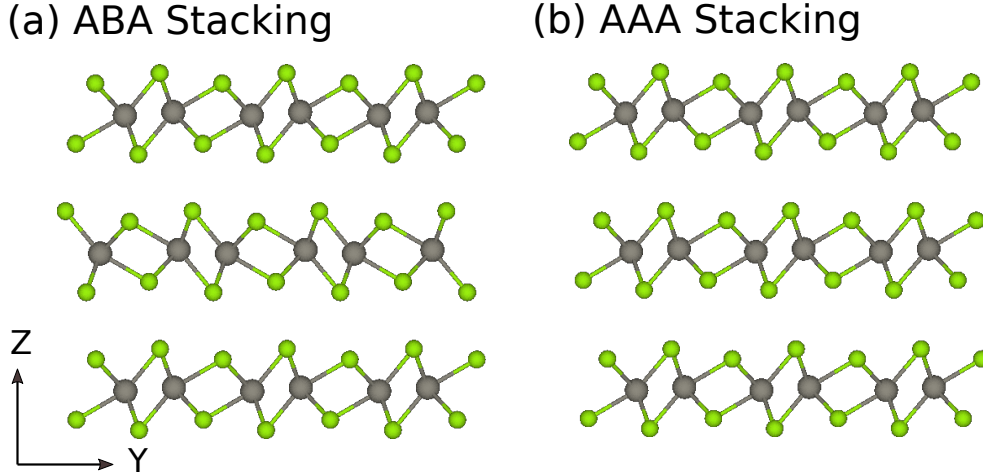


Fig. S1. Stacking configuration considered for bilayer and trilayer 1T'-TMDs. (a) AB bilayer and ABA trilayer configurations. (b) AA bilayer and AAA trilayer configurations.

Table S1. Relative stacking energies.

System	Energy difference	1T'-WSe ₂	1T'-WS ₂
Bilayer	$E_{AA} - E_{AB}$	7 meV	44 meV
Trilayer	$E_{AAA} - E_{ABA}$	13 meV	78 meV

Supplementary Section S2: Layer-dependent electronic structure

For 1T'-WS₂, the calculated monolayer gap is 151 meV, and the monolayer is classified as a quantum spin Hall insulator with $\mathbb{Z}_2 = 1$. Figure S2 presents the band structure and edge spectrum for the monolayer and the band structure for the bilayer and trilayer systems. The bilayer provide small band gap with $\mathbb{Z}_2 = 0$ and trilayer exhibit semimetallic band structures. Compared to 1T'-WSe₂ cases, overall electronic structure are consistent except the trilayer.

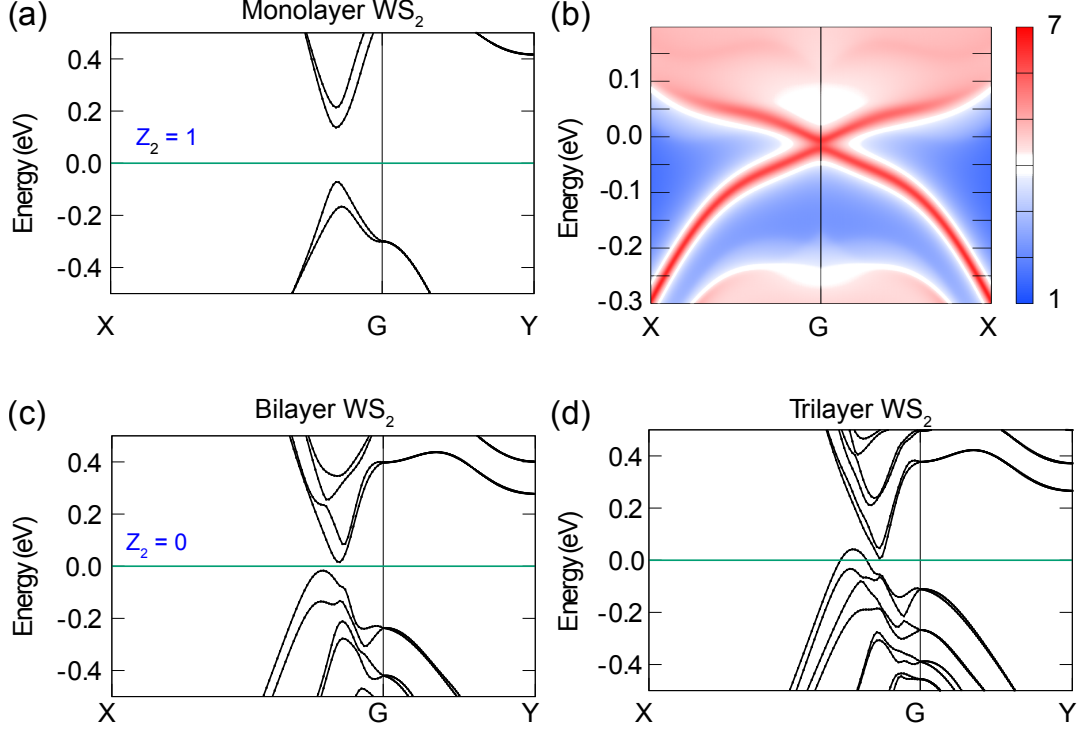


Fig. S2. Layer-dependent electronic structure of 1T'-WS₂. (a) Monolayer band structure, with a calculated gap of 151 meV and $\mathbb{Z}_2 = 1$. (b) Edge state angle resolved photoemission spectroscopy (ARPES) calculation for monolayer. (c,d) Band structures of the (c) bilayer and (d) trilayer, respectively.

Supplementary Section S3: Light-induced interlayer breathing motion with various laser frequencies

Here, we investigate light-induced interlayer breathing dynamics ($\Delta d(t) = (d(t) - d(0))/2$) at different laser photon energies. Trilayer 1T'-WS₂ is excited at photon energies $\hbar\omega = 0.1, 0.6$, and 1.5 eV, with laser intensities of 1.82×10^{11} , 1.43×10^{12} , and 4.09×10^{11} W/cm², respectively (See Fig. S3(a)). Excitation at 0.1 eV induces interlayer contraction, whereas excitation at 1.5 eV produces a negligible change in interlayer spacing. Notably, excitation at 0.6 eV induces interlayer expansion in trilayer 1T'-WS₂, in contrast to the contraction observed in trilayer 1T'-WSe₂ at the same photon energy.

We also investigate trilayer 1T'-WSe₂ under excitation at $\hbar\omega = 3.0$ eV with a laser intensity of 2.23×10^{12} W/cm². This excitation induces interlayer contraction, whereas excitation at the same photon energy produces interlayer expansion in trilayer 1T'-WS₂. The dynamics of 1.2 eV laser with 3.56×10^{12} W/cm² intensity is also investigated to distinguish it from two-photon process by 0.6 eV laser. As shown in Fig. S3(b), 1.2 eV laser induces the expansion of interlayer spacing, while 0.6 eV provides contraction. These results highlight the sensitivity of light-induced interlayer dynamics to both the material and the excitation conditions, including photon energy.

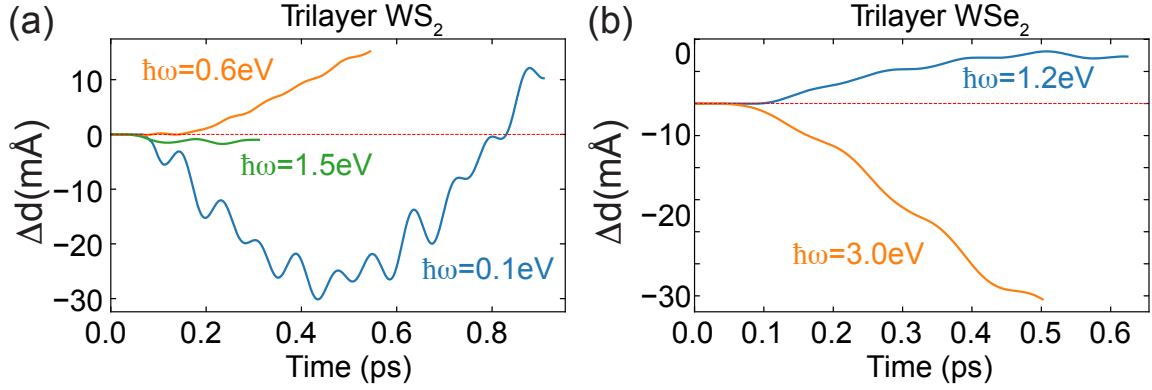


Fig. S3. TDDFT simulations of interlayer breathing motion with various frequencies of laser pulse (a) Light-induced interlayer-spacing change in trilayer 1T'-WS₂ with 0.1, 0.6, and 1.5 eV laser. (b) Light-induced interlayer-spacing change in trilayer 1T'-WSe₂ with 1.2 eV and 3.0 eV laser.