

Supporting Information:

Strongly Coupled Electron-Phonon Dynamics in Few-Layer TiSe₂ Exfoliates

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This SI includes:
Figures S1-S3.

I. Additional UED Results

The time-resolved intensity behavior of the Bragg diffraction spots is shown in Figure S1, panel (a), at laser excitation fluences of 1.4, 2.3, 3.4, 4.6, and 5.7 mJ/cm². These temporal profiles are obtained by averaging over the six first-order diffraction spots shown in Figure 2 (A) of the manuscript. A biexponential function of the form $I = I_0 + A_1 e^{-\tau_1/t} + A_2 e^{-\tau_2/t}$ is used to accurately fit the transient decay. Figure S1 (b) shows the residual change in intensity obtained by subtracting the biexponential fit from the experimental time-resolved change in intensity of the Bragg diffraction spots. These residual plots show the high accuracy of the biexponential fits of the experimental data shown in Figure S1 (a). Figure S2 shows the time-dependent c-axis lattice constant of TiSe₂ sample obtained at non-normal incidence angles, that exhibits coherent breathing modes overlapped with sample expansion following photoexcitation. The residual spectra showing the acoustic oscillations is presented in Figure 4 of the manuscript.

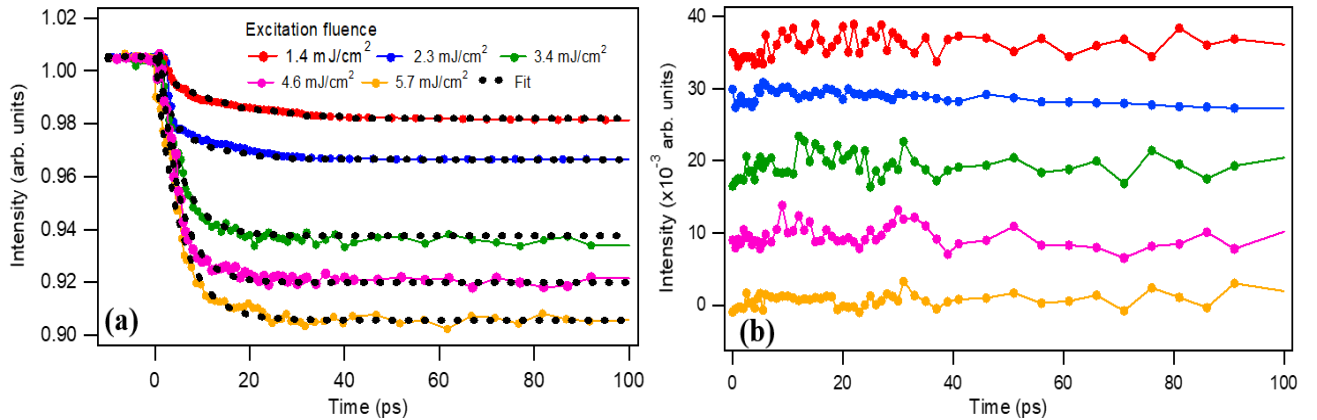


Figure S1. (a) Time-resolved intensity change of the Bragg diffraction spots and (b) residual change in intensity obtained by subtracting the biexponential fit from the change in intensity at laser excitation fluences of 1.4 (red dots), 2.3 (blue dots), 3.4 (green dots), 4.6 (pink dots), and 5.7 (yellow dots) mJ/cm². The dynamics of the change in intensity are well characterized by a biexponential fit.

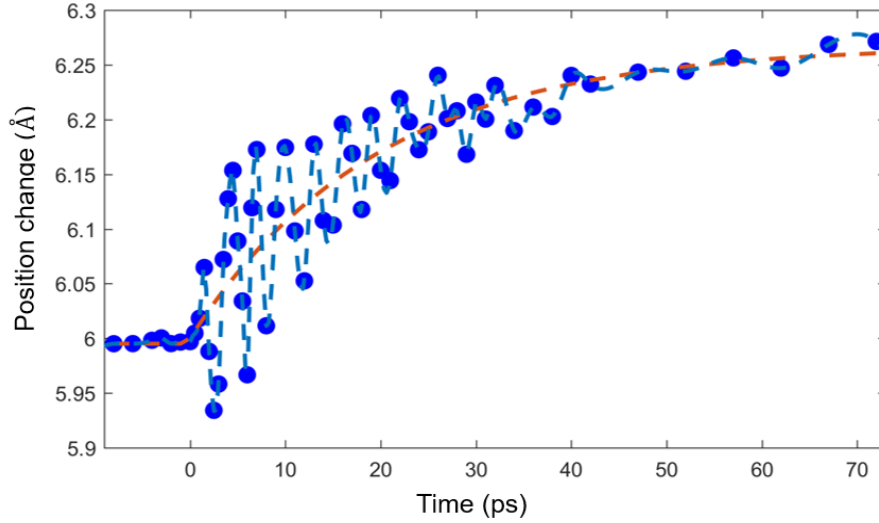


Figure S2. Time-dependent c-axis lattice constant of TiSe₂ sample exhibiting coherent breathing modes overlapped with simple expansion following photoexcitation.

II. Raman Spectroscopy

Figure S3 shows the Raman spectrum of single-crystal TiSe₂ sample after excitation at 532 nm. Both E_g and A_{1g} phonon modes are observed at around 135 and 200 cm⁻¹, respectively.

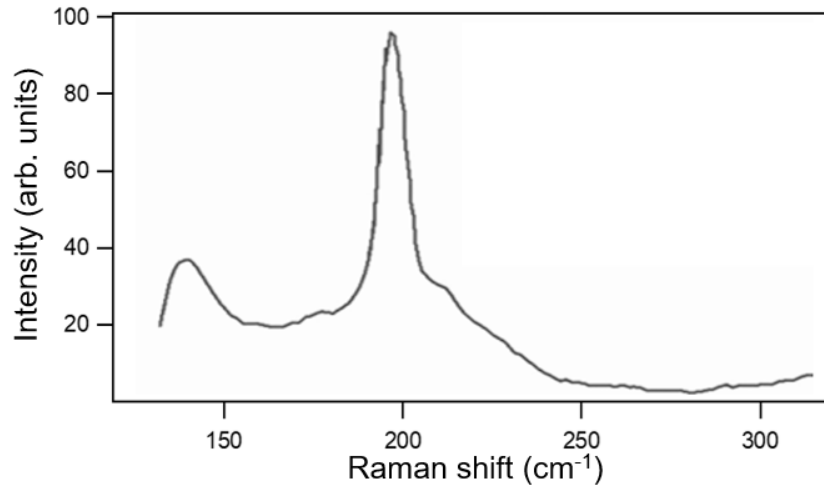


Figure S3. Raman spectrum of the single-crystal TiSe₂ sample showing the E_g and A_{1g} phonon modes.