Nonlinear Photocarrier Dynamics in Multilayer WSe_2 Driven by Strong THz Pulses

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Abstract: We demonstrate that strong THz pulses enhance THz absorption in optically excited WSe_2. The increase in THz conductivity indicates that strong THz fields drive photocarriers into higher mobility states.

Transition metal dichalcogenides (TMDs) are 2D semiconductors which exhibit strong optical responses such as exciton resonances and spin-dependent photocarrier dynamics owing to their unique band structure [1]. High-field THz spectroscopy, a powerful tool for manipulating carrier dynamics in photoexcited materials [2], has been utilized to modulate optical responses in TMDs such as excitonic resonances in monolayer MoS_2 [3]. In this study we investigate high-field photocarrier dynamics in CVD grown large-grain multilayer WSe2, employing time-resolved high-field THz spectroscopy.

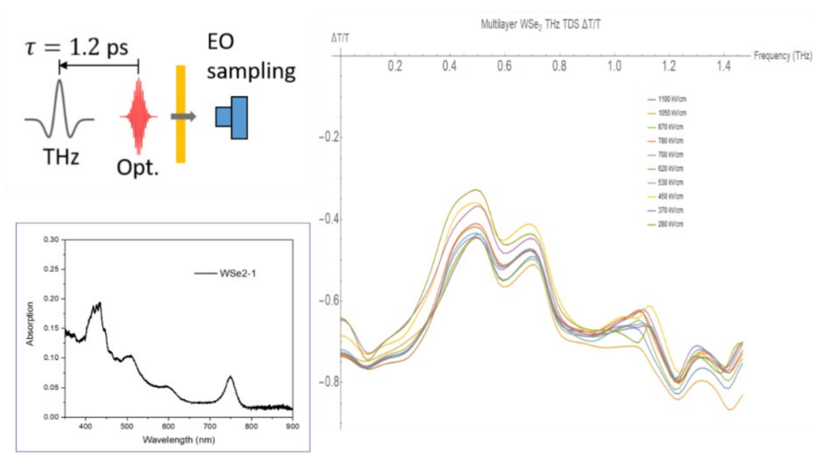

FIG. 1. (a) schematic (b) absorption spectrum of monolayer WSe2. (c) Time-resolved THz transmission of the optically excited WSe2 sample.

Figure 1a provides a rudimentary schematic of the optical pump/THz probe experiment. The WSe_2 sample was optically excited by 400-nm and 800-nm 100-fs pulses, whose photon energies are above and below the bandgap of monolayer WSe_2, respectively. We generate intense single-cycle THz pulses by tilted-pulse-front optical rectification in a Mg:LiNbO3 prism [4]. The field amplitude of the broadband THz pulses reached 1.1 MV/cm at an optical pulse energy of 1 mJ. We measured THz pulse energy using a pyroelectric detector and obtained THz waveforms employing electro-optic sampling in a 1-mm ZnTe crystal. Figure 1c shows that an optical excitation, instantaneously injecting photocarriers, induces strong THz absorption in WSe2. The photocarriers undergo slow relaxation in hundreds of picosecond time scale.

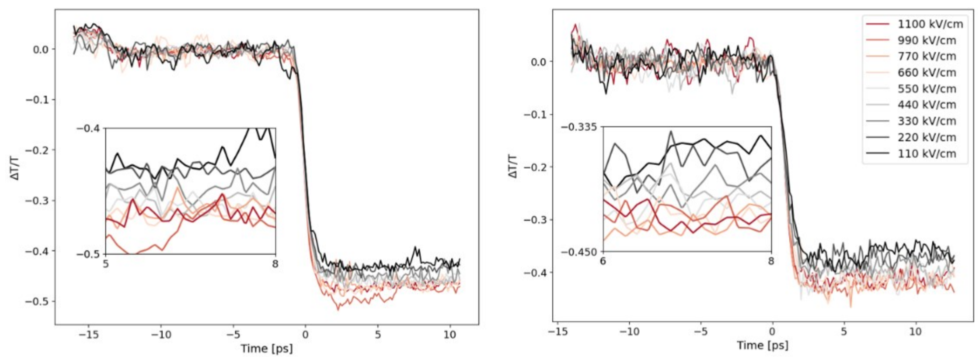

FIG. 2. Time-resolved differential THz transmission of WSe2 at the peak THz field amplitude of 110, 220, 330, 440, 550, 660, 770, 990 and 1100 kV/cm for (a) 400-nm and (b) 800-nm optical pumping.

Figure 2 shows the time-resolved differential THz transmission of the WSe2 sample at various peak THz field strengths from 110 to 1100 kV/cm for (a) 400-nm and (b) 800-nm optical pumping. The THz transmission reduction induced by optical excitation exhibits nonlinear responses in both pumping schemes. The differential THz transmission decreases from 43% at 110 kV/cm to 48% at 1100 kV/cm for the 400-nm pumping, and from 37% to 42% for the 800-nm pumping.

In order for more detailed examination about the nonlinear THz responses, we performed high-field THz time-domain spectroscopy. Figure 3a shows the waveforms transmitted through the sample, and the corresponding transmission spectra are shown in Fig.3b. The decrease in transmission as the THz-field strength increases is notable, along with the nonlinear decrease in the magnitude of this induced change as the strength increases.

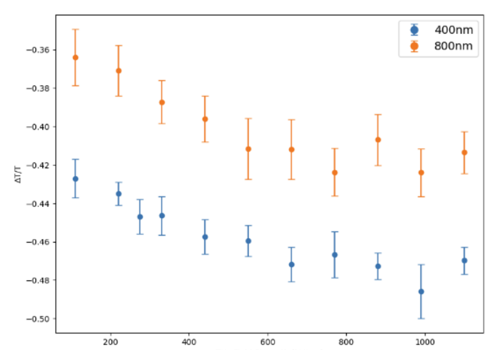

FIG. 3. (a) Waveforms (b) Spectra (c) Differential transmission

Figure 3c showcases the averaged differential transmission for various THz field strengths. In Figure 3a, we see that above 600 kV/cm the 400nm pumped WSe2 begins to stop experiencing field strength dependent effects. In Figure 3b, a similar behavior is observed, but at a lower strength of roughly 500 kV/cm

Microscopic mechanisms for the field induced nonlinear THz absorption in optically excited WSe2 are still a work in progress. We speculate that the strong THz fields drive photocarriers into sidebands of high mobility and enhance the THz conductivity of the material. An alternative mechanism is that the strong THz fields are freeing electrons trapped in defect states, also resulting in rise of the conductivity.

References

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