Nonlinear Optical Transmission in WSe_2 Induced by Intense THz Fields

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Abstract: In WSe_2 intense THz fields are found to enhance transmission at 400 nm, while reducing it at 800 nm. The differential transmission is proportional to the field amplitude. The nonlinear responses are fast, yet non-adiabatic.

Transition metal dichalcogenides (TMDs) are semiconducting 2D materials exhibiting strong optical responses including exciton resonances and spin-dependent photocarrier dynamics due to their unique band structure [1]. High-field terahertz (THz) spectroscopy provides a powerful tool to manipulate photocarriers in semiconductors, which has great potential for applications in high-speed optoelectronics [2,3]. It has been demonstrated that a strong THz field modulates the excitonic resonances of monolayer MoS_2 [4]. In this study, we investigate ultrafast dynamics of photocarriers and nonlinear optical effects in CVD grown large-grain WSe_2 monolayer and multilayers induced by strong THz fields, employing time-resolved optical-pump/THz-probe and THz-control/optical-probe spectroscopy.

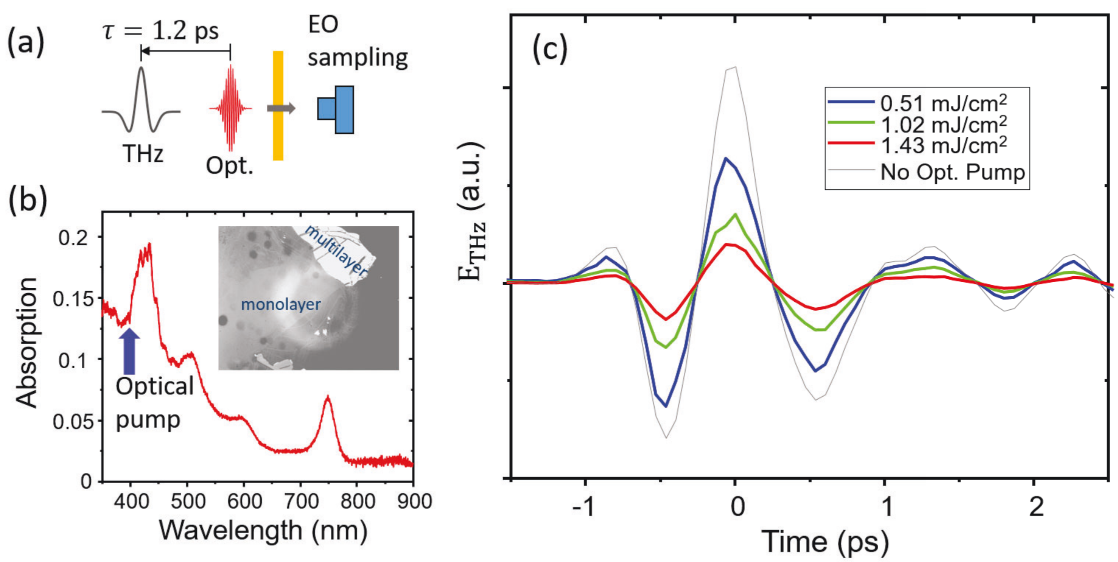

FIG. 1. (a) Schematics of the optical pump/THz probe setup (b) Optical absorption spectrum of the WSe_2 sample. A microscopic image of the sample is shown in inset. (c) THz waveforms transmitted through the sample for the optical pump fluence 0.51, 1.02 and 1.43 mJ/cm^2. The gray line presents the waveform of no optical excitation.

We performed time-domain THz spectroscopy of an optically excited WSe_2 sample. Intense single-cycle THz pulses were generated by tilted-pulse-front optical rectification in a Mg:$LiNbO_3$ prism [5]. The field amplitude of the broadband THz pulses (central frequency, 0.7 THz; bandwidth, 0.7 THz) reached 1.2 MV/cm at an optical pulse energy of 1 mJ. We measured the THz waveforms via electro-optic sampling in an 1-mm ZnTe crystal. Figure 1a shows a schematic diagram of the optical pump/THz probe experimental setup. We optically excited the sample with 400-nm, 100-fs pulses, where the photocarriers are injected just above the C-exciton resonance at 420 nm. The optical absorption spectrum of the WSe_2 sample is shown in Fig. 1b. The optical excitation above the bandgap induced large absorption of THz radiation in the sample, while the unexcited sample, an insulator, is transparent in the THz range. Figure 1c shows that the THz waveforms transmitted through the sample after 1.2 ps of the optical excitation are significantly reduced, demonstrating strong interaction between the THz pulses and photocarriers.

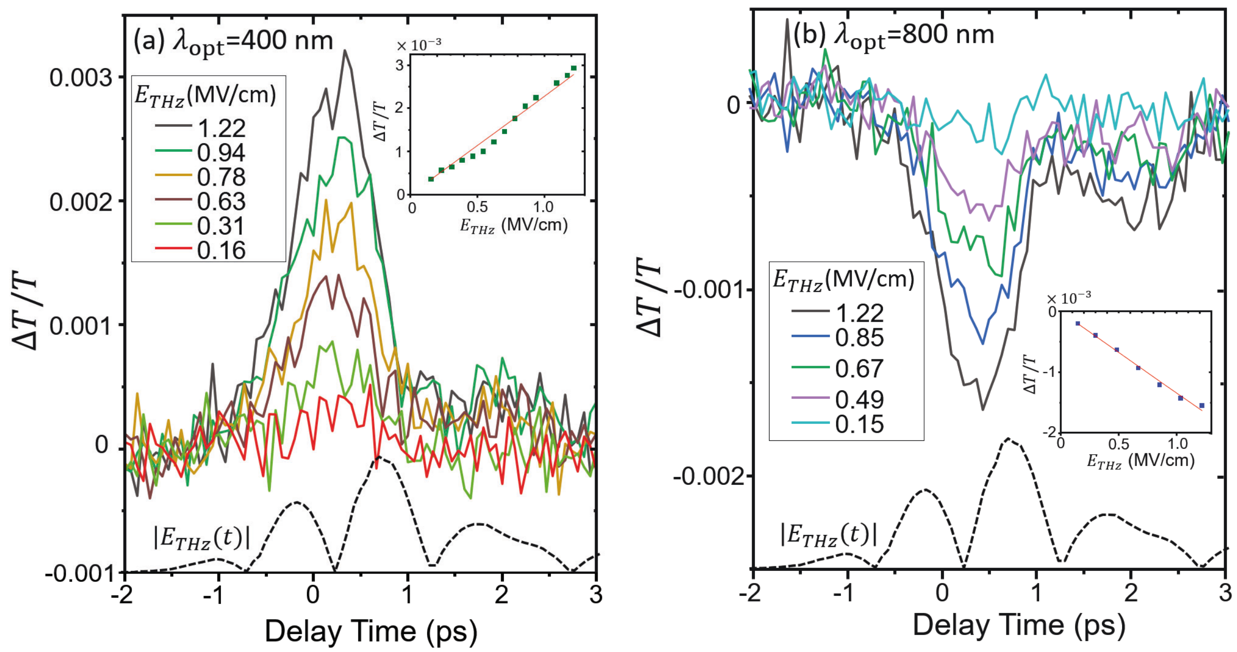

FIG. 2. Time-resolved differential transmission of optical probe pulses at (a) 400 nm and (b) 800 nm induced by intense THz fields. The peak THz field strength is varied from 0.15 MV/cm to 1.22 MV/cm. The insets show the maximum differential transmission vs. the peak THz field intensity. The dotted lines at the bottom presents the THz field magnitude as a function of the delay time.

We study the effect of intense THz fields on the optical properties in multilayer WSe_2, measuring optical transmission below (800 nm) and above (400 nm) the bandgap through the large size (~2 mm) multilayer flake shown in the inset of Fig. 1b near the top of the microscopic image of the sample. The transmission at 400 nm enhances as the THz field intensity increases, while the transmission at 800 nm reduces. We speculate that the field-induced nonlinear optical effects involve many-body interactions between photocarriers. The increase in absorption at 800 nm might be caused by the THz field induced broadening of the A exciton resonance at 750 nm [4]. The transmission enhancement at 400 nm is extraordinary, explained neither by broadening nor by blue-shift of the C exciton resonance at 420 nm. The differential transmission is proportional to the THz field strength, as shown in the Fig. 2 insets. It is noteworthy that the nonlinear optical effects are non-adiabatic: the optical responses have the time scale similar to the THz pulses, yet they are not completely transient.

In summary, we demonstrate that strong THz fields produce nonlinear optical effects in WSe_2. The different optical responses below and above the bandgap indicates that the hot and cold photocarriers undergo distinctive high-field dynamics.

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