

Ultrafast Nonlinear Optical Effects in Semiconductor Quantum Wells Resonantly Driven by Strong Few-Cycle Terahertz Pulses

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Abstract:

Intraexciton transitions in semiconductor quantum wells are modulated by strong and tunable few-cycle terahertz pulses. Time-resolved terahertz-pump and optical-probe measurements demonstrate that the $1s$ heavy-hole and light-hole exciton resonances undergo large-amplitude spectral modulations when the terahertz radiation is tuned near the $1s$ - $2p$ intraexciton transition. The strong nonlinear optical transients exhibit the characteristics of Rabi sidebands. The spectral features also reveal the dephasing properties of the optically dark $2p$ states. A microscopic theory shows that the $2p$ -dephasing rate is three times that of the $1s$ -state. The ultrafast nonlinear optical effects and their quantum nature suggest promising applications to ultrahigh-speed optical signal processing and quantum information processing in the THz region.

Keywords:

few-cycle terahertz pulse, nonlinear terahertz effect, quantum well, exciton, Rabi sideband

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1. Introduction

Interaction of strong terahertz (THz) pulses with semiconductors is an uncharted subject, yet it not only provides fresh insights into fundamental physical phenomena such as many-body interactions in correlated systems, but also grants potential applications to quantum information science and ultrahigh-speed optical signal processing. THz spectroscopy is a powerful tool to investigate the material properties inaccessible until recently [1]. In particular, the interaction of THz radiation with semiconductor is of great interest because many elementary excitations involving carrier dynamics and lattice vibrations have characteristic energies in this spectral region. The majority of THz studies of semiconductors have been concerned with linear optical properties measured by weak THz probe beams. A handful of recent studies, however, have explored the uncharted regime of nonlinear phenomena and non-equilibrium states induced by strong THz radiation [2-7]. These THz nonlinear effects are associated with intraband transitions in semiconductors.

Excitons in semiconductor quantum-wells (QWs) have the simple energy-level structure of a two-dimensional hydrogen-like system. The exciton binding energy in GaAs-based QW structures is in the range of ~ 10 meV, corresponding to the photon energy of THz radiation (4.14 meV at 1 THz). THz-induced nonlinear optical effects of excitons in QWs are of great interest because they provide new insights into the quantum coherence and wavepacket dynamics in semiconductors. For example, an experiment/theory study demonstrated that strong single-cycle THz pulses induce coherent extreme-nonlinear optical transients in semiconductor QWs, where the rotating wave approximation breaks down and the ponderomotive effects become prominent [6].

We present experimental observations and theoretical calculations of strong interactions between THz light and excitons, demonstrating the ultrafast dynamics of excitonic quantum coherence in GaAs/AlGaAs QWs resonantly driven by intense narrowband THz pulses. Figure 1 illustrates the THz-exciton interaction, where the exciton level structure is reduced to a cascaded three-level system including the two main exciton states, $1s$ and $2p$. The driving THz-field is tuned near the resonance of the $1s$ - $2p$ intraexciton transition. The strong THz field couples the two exciton states, producing nonstationary dressed states. An ultrafast optical probe is employed to observe the time evolution of the nonlinear optical effects of the $1s$ exciton level. Being simultaneously driven by the THz and the optical pulses, the QW system sustains two coherences involving interband and intraexciton transitions. Their dephasing times are in the picosecond range. The unique experimental arrangement of the ultrafast optical (90 fs) and THz (3 ps) pulses makes it possible to observe the THz-induced nonlinear optical transients of the coherent excitons.

2. Experimental setup

Figure 2 shows the schematic diagram of the experimental setup for time-resolved THz-pump and optical-probe measurements. The light source of the experiment was 1-mJ, 800-nm optical pulses from a 1-kHz Ti:sapphire regenerative amplifier (Coherent Inc., Legend). The pulse compressor of the amplifier introduced a linear chirp to stretch the optical pump pulses to ~ 4 ps. The optical beam was split into two by a beam splitter. The minor portion of the optical power (5%) was used as an optical probe to measure the transmission spectra of the $1s$ -exciton resonance. The probe pulses went through a pulse compressor which reduced the pulse duration to 90 fs. They transmitted through the QW sample and the transmission spectra were measured by a spectrometer. The major portion of the optical power (95%) was used for strong narrowband

THz pulse generation. The pump beam was split into two, and a relative time delay was introduced between them. They were orthogonally polarized, and eventually recombined to co-propagate. The two linearly-chirped and orthogonally-polarized optical beams produced strong few-cycle THz pulses via type-II difference-frequency generation in a 1-mm ZnTe crystal [8]. The THz pulses passed through a THz pass filter which blocked the optical pump pulses. The THz pulse energy was in the range of 1 nJ, and the pulse duration was ~ 3 ps. The maximum amplitude of electric field reached ~ 5 kV/cm. The central frequency of the spectrum is continuously tunable from 0.5 to 2.5 THz with the bandwidth of 0.2-0.5 THz.

We focused the THz pulses on a GaAs/AlGaAs QW sample, and measured its optical transmission spectra $T(\omega)$ with the weak 800-nm, 90-fs optical probe pulses as varying the relative time delay (t_d) between the THz and optical pulses. The sample of a semiconductor nanostructure contains ten high-quality, undoped 12-nm-wide GaAs QWs separated by 16-nm-wide $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ barriers. The sample was placed in a liquid helium cryostat keeping the temperature at 5K. The THz radiation was polarized in the QW plane so that it couples with intraexciton transitions, but not with intersubband transitions.

We characterized the THz pulses with a THz Michelson interferometer composed of two gold mirrors and a mylar beam splitter [Fig.3(a)]. THz interferometric autocorrelations were measured by a Si-bolometer. Figure 3(b) shows the time-resolved interferogram of the THz pulse, where the optical pulse duration and the relative time delay were 4.11 ps and 1.90 ps, respectively. The THz radiation is narrow band: the spectrum centered at 2.34THz has a spectral width of 0.2 THz with an associated quality factor of ~ 11 . Figure 3(c) shows the spectra for several time delays, demonstrating continuous frequency tuning. The overall tuning range was 0.3-2.5 THz. The spectral width of the THz pulses was ~ 0.2 THz. The data show a water

resonance at 2.23 THz, which reduces the measured pulse width when the central frequency is slightly off resonance. The experimental data indicate that the THz frequency linearly depends on the time delay, and the chirping parameter was found to be 0.613 ps^{-2} .

3. Results and Discussion

Figure 4(a) and (b) shows a time-resolved THz waveform measured by electro-optic sampling and its power spectrum when the THz radiation is tuned at 1.9 THz, and the peak THz field amplitude is $\sim 5 \text{ kV/cm}$. In Fig. 4(c-e), the dotted lines represent the unperturbed $1-T$ spectrum of the sample at 5 K, showing the heavy-hole (HH) and light-hole (LH) exciton resonances at 1.5401 and 1.5468 eV, respectively. In the presence of strong THz fields, the exciton resonance lines undergo remarkable changes in their spectral shape. Figure 4(c-e) displays the spectral modulations in a set of $1-T$ spectra (solid lines) taken at the optical probe time delays, $t_d = -1.7$, 0.9, and 1.7 ps.

The exciton lines broaden and shift to the lower energy side. Furthermore, a shoulder is seen to rise and fall near the HH exciton peak on the higher energy side. The modulated HH exciton line shape becomes strongly asymmetric, indicating line splitting obscured by broadening. This feature suggests that the THz radiation and the exciton levels are strongly coupled. The energy splitting ($\sim 2.5 \text{ meV}$) is consistent with an estimation based on the three-level model, $\hbar\Omega_R = d_{1s \rightarrow 2p} E_{THz} \sim 3 \text{ meV}$, where the exciton dipole moment $d_{1s \rightarrow 2p} \sim 5 e \cdot \text{nm}$ and the THz field amplitude $E_{THz} \sim 5 \text{ kV/cm}$.

Calculations based on a microscopic theory confirm that the main features of the experimental results are accounted for by the strong coupling of THz radiation with excitonic polarizations [7]. The theoretical analysis also shows that some peculiar features of the spectral modulations cannot be explained by the simple three-level model. First, the HH exciton line

shape becomes strongly asymmetric and the high-energy peak is much less pronounced compared with the low-energy peak. The primary reason for this is that the THz radiation can make transitions to the higher exciton levels, $3p$, $4p$, ..., and the conduction band. Another contributing factor is that the $2p$ polarization decays faster than the $1s$ polarization. Based on the calculations of a microscopic theory, the $1s$ and $2p$ polarization have different dephasing constants $\gamma_{1s}=0.5$ meV and $\gamma_{2p}=1.5$ meV, respectively [7]. Second, the exciton lines undergo appreciable spectral broadening which involves the continuum states in the conduction band. As shown in the unperturbed spectrum, the exciton resonances sit on the tail of the interband absorption spectrum. Consequently, THz-induced ionization contributes to the dephasing and the subsequent line broadening. Third, the whole HH exciton spectrum shifts to the high energy side. The blue shift is associated with the quivering motion of the charge carriers driven by the oscillating THz fields. The energy shift (~ 1 meV) is in the same order of magnitude with the ponderomotive energy, $e^2 E_{THz}^2 / 4m_e^* \omega_{THz}^2 \sim 1.4$ meV.

We measured the optical transmission at several THz frequencies from 1.4 to 2.2 THz. Figure 5 shows the $1-T(\omega_{opt})$ spectra at $\nu_{THz}=1.45, 1.68, 1.86,$ and 2.15 THz, which correspond to the detuning from the $1s-2p$ resonance, $\Delta = \nu_{THz} - \nu_{1s-2p} = -0.30, -0.07, 0.09,$ and 0.40 THz, respectively. The effects of the THz field are clearly different at different frequencies. The THz-induced changes at 1.45 THz ($\Delta=-0.30$ THz) are relatively small, while the slight red-shift of the HH exciton line and the emergence of the shoulder around 1.542 eV signify the formation of dressed states. The THz effects become significantly stronger near and above the $1s-2p$ resonance frequency. The differential transmission ($\Delta T = T(\text{with THz field}) - T(\text{w/o THz field})$) at $\Delta = \nu_{THz} - \nu_{1s-2p} = -0.07, 0.09,$ and 0.40 THz reaches remarkably large amplitude ranging from

-0.3 to 0.3. As the detuning increases, the rise of the broad shoulder in the high-energy side of the HH exciton line becomes more significant.

The temporal evolution of the THz-induced nonlinear optical effects exhibits the ultrafast dynamics of the excitonic quantum coherence. Figure 6(a) shows the driving THz waveform tuned at 1.86 THz. The contour plot of Fig. 6(b) displays the time-resolved differential transmission spectra $\Delta T(\omega_{\text{THz}})$ with a time step of 1/15 ps. The fanning-out spectral oscillations in the early time stage from -4 to -1 ps demonstrate transient coherent nonlinearity of the exciton system induced by the THz pulses. The coherent transient lasts ~2 ps corresponding to the dephasing time of the excitonic interband polarization. In the later time, the spectrum develops a strong asymmetry, signifying the formation of intraexcitonic coherence. The deep and broad spectral feature near 1.542 eV in the high-energy side of the HH exciton resonance indicates the evolution of the quantum coherence. Figure 6(c) shows characteristic changes in the ΔT spectra at several time delays, $t_d = -3.0, -2.0, -1.0, 0.0, 1.0, \text{ and } 2.0$ ps. The spectral shift of the dip near 1.542 eV is clearly seen; the dashed line to guide the eye.

4. Conclusion

In conclusion, coherent excitons in semiconductors, being resonantly driven by strong THz pulses tuned at the $1s$ - $2p$ intraexciton transition, exhibit pronounced ultrafast nonlinear optical effects. The coherent dynamics of the quantum system is obscured by strong dephasing effects. The remarkably large signal of the nonlinear optical transients betokens promising applications to high-speed optical communications and quantum information processing.

Acknowledgements

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Figure Captions

FIG 1. Energy-level diagram of an exciton resonantly driven by strong few-cycle THz pulses. The cascaded three-level model describes the THz and optical interaction with excitons and the formation of the dressed states resulting in Rabi splitting.

FIG 2. Experimental setup for THz-pump and optical-probe measurements with intense narrowband THz pulses produced by type-II difference frequency generation.

FIG 3. (a) Michelson interferogram of narrowband THz pulses tuned at 2.3 THz, which were generated by type-II difference frequency generation in a 1-mm ZnTe crystal with 4.11-ps optical pulses at 1.90 ps time delay. (b) Normalized power spectra of narrowband THz pulses for time delays from 1.50 ps to 2.03 ps. The inset shows the central frequencies of the spectra as a function of pulse delay. The solid line fits the data.

FIG 4. (a) Waveform of the narrowband THz radiation measured by electro-optic sampling and its power spectrum. (b) 1-Transmission spectra (solid lines) of heavy-hole and light-hole exciton resonances in a THz-driven GaAs/AlGaAs QW system at 5 K, where the delay time is (c-1) $t_1 = 1.7$ ps, (c-2) $t_2 = 0.9$ ps, and (c-3) $t_3 = 1.7$ ps. The dotted lines indicate the unperturbed spectrum.

FIG 5. $1-T(\omega_{\text{opt}})$ spectra of the exciton lines at different THz frequencies, (a) 1.45, (b) 1.68, (c) 1.86, and (d) 2.15 THz. The time delays between optical and THz pulses are 1.0, 1.2, 1.2, and 1.2 ps, respectively. The peak THz field amplitudes are ~ 5 kV/cm at 1.45, 1.68, and 1.86 THz and ~ 4 kV/cm at 2.15 THz. The insets show the power spectra of the THz radiation.

FIG 6. Temporal evolution of excitonic coherence driven by THz pulses. (a) Waveform of the driving THz pulses turned at 1.86 THz. (b) Differential transmission ΔT of the LH and HH excitons as a function of time and photon energy. The magnitude of ΔT is scaled in shade. (c) ΔT spectra at $t_d = -3.0, -2.0, -1.0, 0.0, 1.0,$ and 2.0 ps.

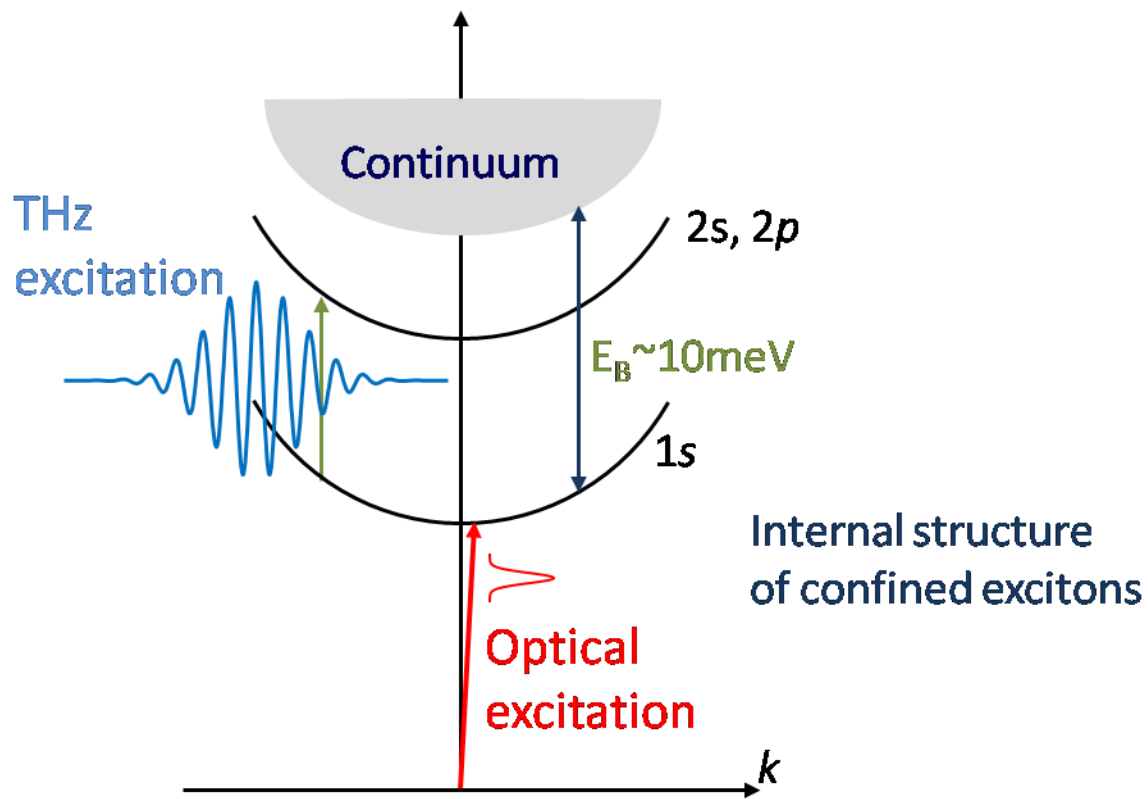


FIG 1.

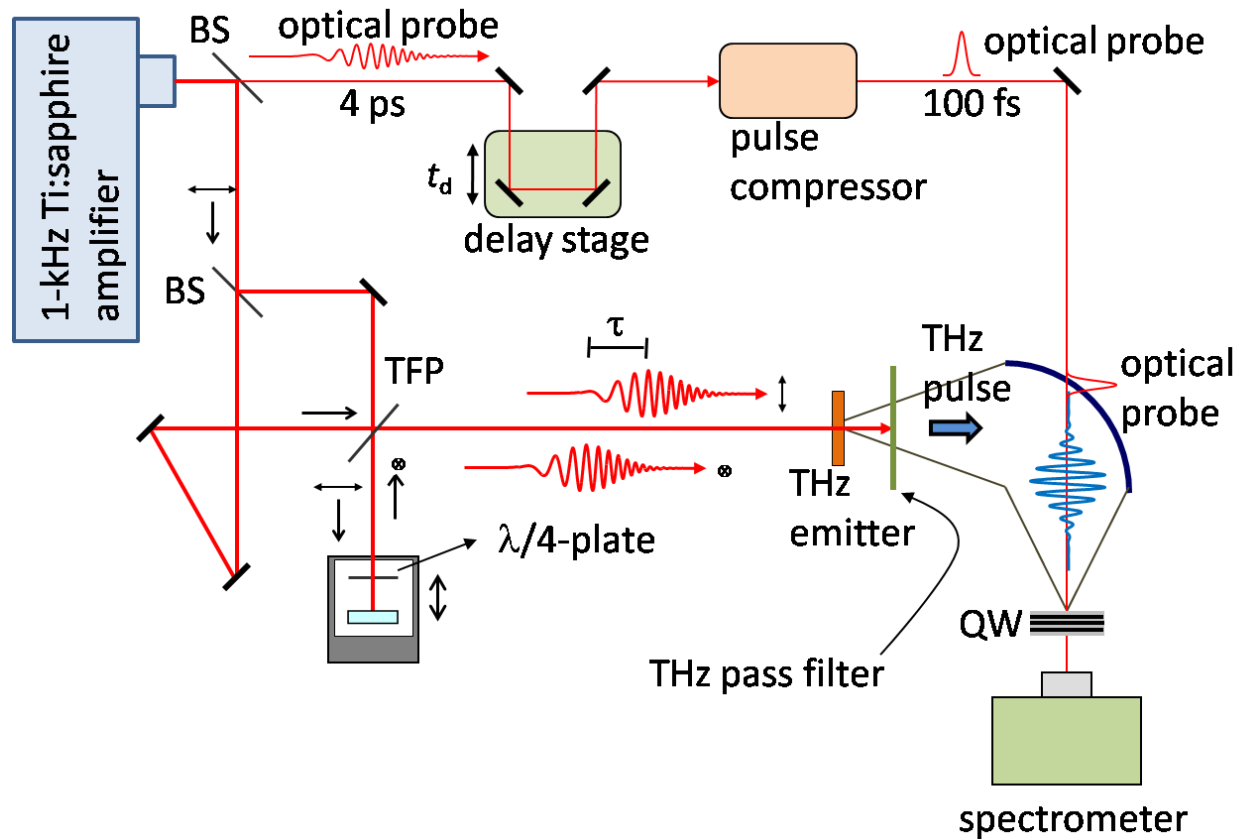


FIG 2.

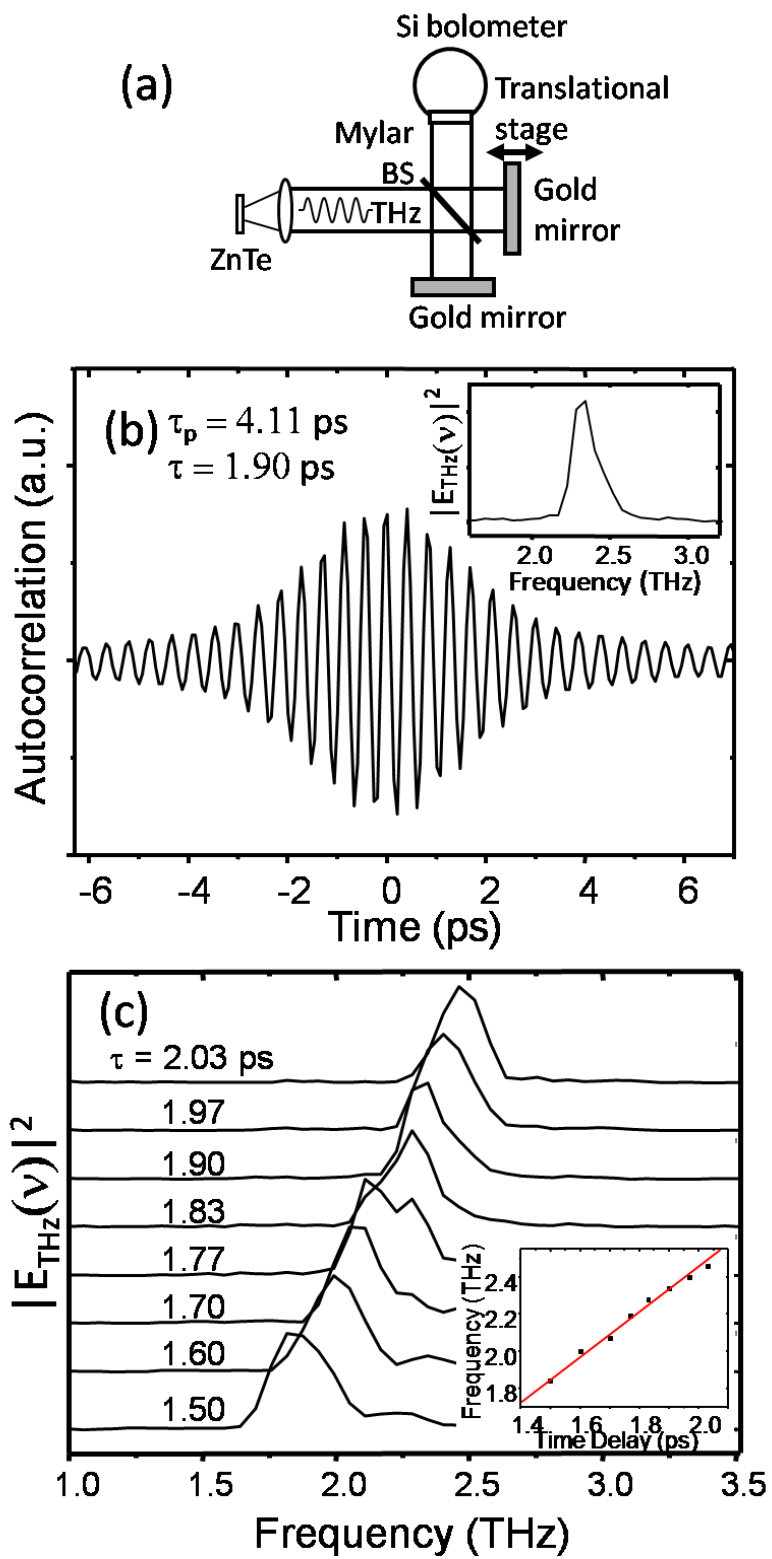


FIG 3.

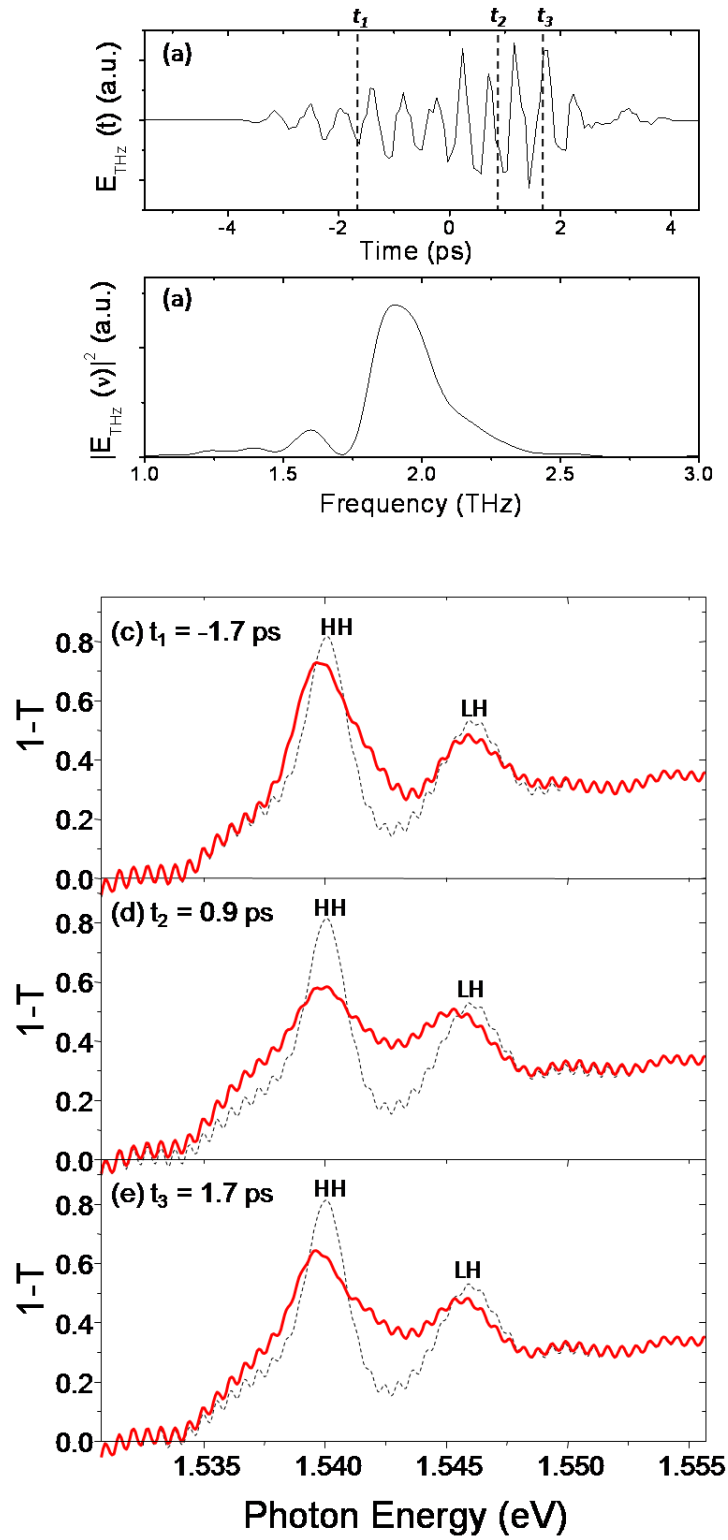


FIG 4.

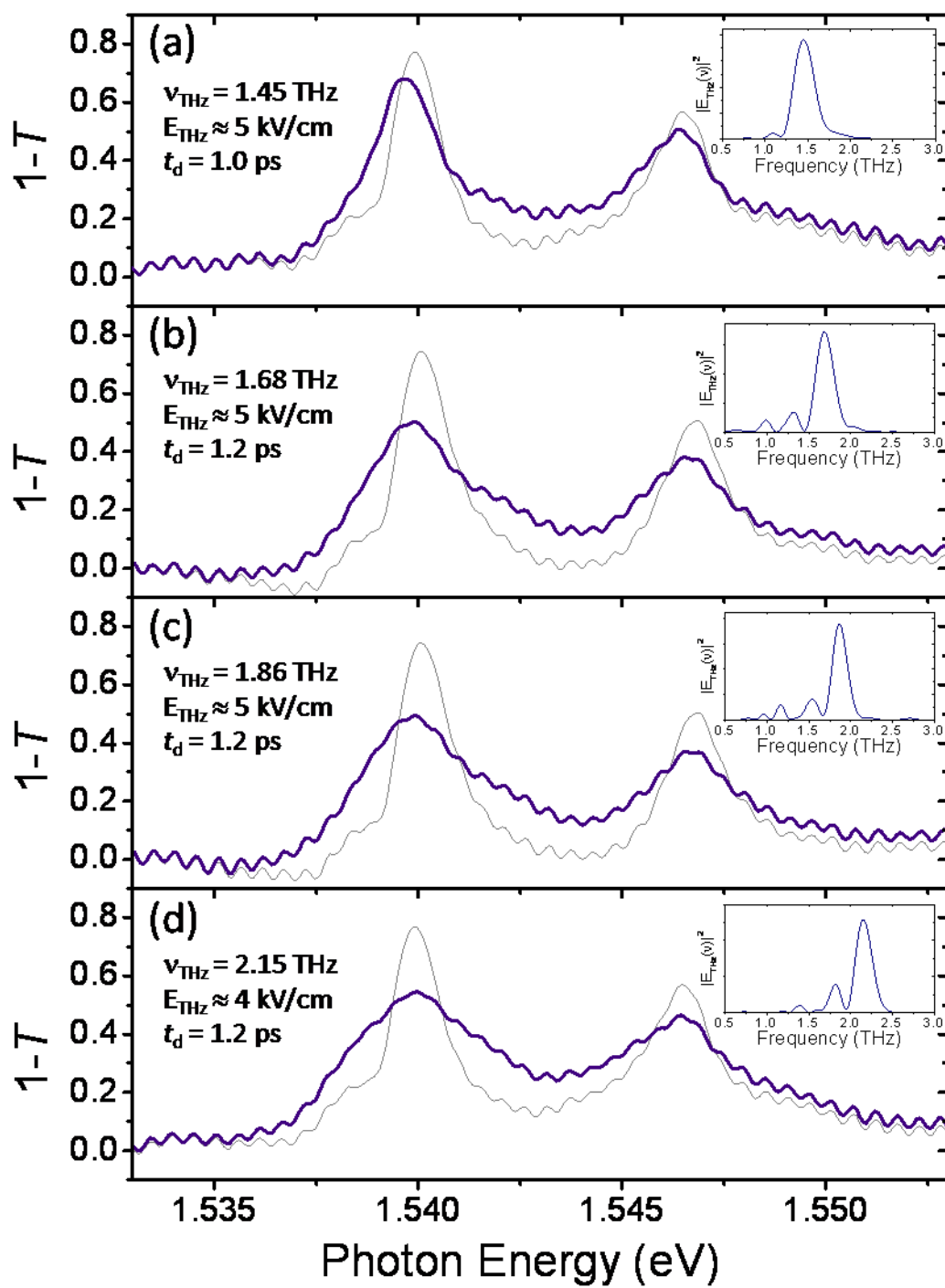


FIG 5.

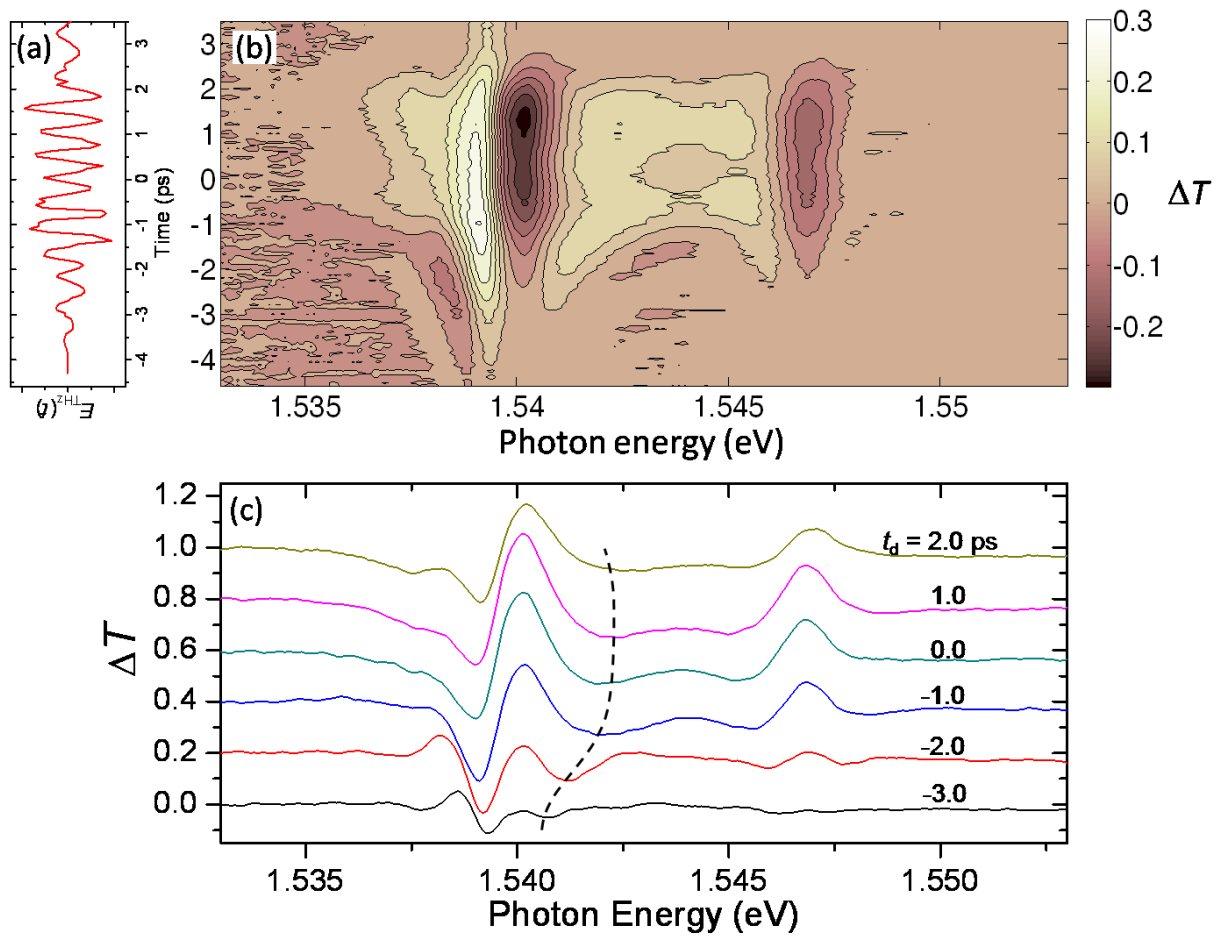


FIG 6.