# TERAHERTZ NONLINEAR OPTICS AND SPECTROSCOPY

by

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# ABSTRACT

This dissertation focuses on the study of THz nonlinear optics and spectroscopy. By using recently developed intense, few-cycle THz pulse sources, nonlinear optical responses of selected materials at THz frequencies are investigated. The characteristics of these nonlinear phenomena are different from the corresponding effects in the microwave and visible frequency ranges.

This dissertation starts with the study of carrier dynamics under the influence of an intense THz field, which are important to both fundamental research and the application of ultrafast electronics. By using a pump-probe-technique, a nonlinear transmission enhancement of intense THz waves in a photoexcited GaAs sample is observed. This nonlinear response is attributed to the intervalley scattering of electrons in the presence of a high THz electric field. The Drude model is used to fit the frequency-dependent photoconductivity in the GaAs sample. The deviation from a Drude response of the observed photoconductivity spectrum at high THz fields, compared with the fitted Drude behavior of the photoconductivity spectrum at low THz fields, verifies the nonlinear response due to the THz-induced intervalley scattering.

In addition, THz-induced cross-phase-modulation (XPM) is observed in a ZnTe crystal, in which the phase of an optical pulse is modulated due to the presence of transient birefringence induced by an intense THz pulse through the Pockels effect. The significant phase shift on the sub-picosecond time scale causes strong changes in the spatiotemporal profile of the optical pulse, yielding spectral shifting, broadening, and lensing effects. One distinctive feature of this THz-induced cross-phase-modulation is that it is a second-order nonlinear process, while its counterpart in the visible/infrared range is normally a third-order nonlinear process.

THz-induced third-order nonlinear processes, including THz-field-induced second-harmonic generation (TFISH) and THz Kerr effect, are also investigated in this dissertation. These processes can be described by a THz-related four-wave-mixing (FWM) mechanism. For TFISH, an optical wave at a frequency of 2ω is generated from the mixing of two optical fundamental waves and a THz wave at frequencies of ω, ω and $\Omega_{THz}$ respectively. In the THz Kerr effect, a transient refractive index change proportional to the intensity of THz waves is produced. Furthermore, when using a β-

BBO crystal as the medium for TFISH, the optical second-harmonic generation in the β-BBO crystal through a second-order nonlinear process serves as an efficient local oscillator and mixes with the TFISH, yielding an measurable interference term which is linearly proportional to the THz field. This heterodyne technique provides an alternative method for the coherent characterization of intense THz waves.

With these studies of THz nonlinear optics and spectroscopy, this dissertation seeks to further our understanding of the fundamental laws of wave-matter interactions in the THz range which could lead to future applications in optical coherence control, THz devices, and THz communications.