論文の内容の要旨

Strong electric field processes in solids induced by an intense mid-infrared light source

（高強度中赤外光源によって誘起される
固体中の強電場過程の研究）

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Recent progress of ultrafast laser technology has opened a new research area in strong-field physics where atoms and molecules in strong laser fields exhibit extremely nonlinear responses such as tunnel ionization and high harmonic generation. Using such extreme nonlinearity, attosecond science has been well developed in the past decade, and our understanding of ultrafast electron dynamics in atoms and molecules have been expanded tremendously. Meanwhile, strong field physics in condensed matters is still unexplored because it is difficult to nondestructively apply intense optical fields to solids. In order to apply strong electric fields to solids, intense and long-wavelength sources are needed whose photon energy is much smaller than their bandgap energies. We propose and demonstrate a novel scheme to produce intense mid-infrared pulses with stable carrier-envelope phases. We successfully apply electric fields of several tens MV/cm to crystalline solids without inducing optical breakdown, and observe extremely nonlinear responses such as high harmonic generation and field-sensitive change in transient absorption.

The thesis contains three achievements. First is the generation and complete characterization of carrier-envelope phase-stable intense mid-infrared fields with a peak electric field strength exceeding 50 MV/cm. A newly developed technique of optical parametric amplification, namely dual-wavelength optical parametric amplification (OPA), is demonstrated to produce intense mid-infrared pulses. Owing to the intrinsic simplicity of the technique, a long-term carrier-envelope phase stability is assured. A novel scheme for the generation of 6.5-fs visible pulses is also demonstrated and used

for the complete characterization of the mid-infrared fields. This scheme provides ultrashort visible pulses with smooth and stable spectra which are crucial to be used as a probe in sub-cycle spectroscopy. Complete waveform characterization of the mid-infrared field is realized via electro-optic sampling with a LGS crystal which has an extremely broad transparency range from ultraviolet to mid-infrared.

Second achievement is the generation of high harmonics in a crystalline solid with ellipsometric analysis to trace the electron dynamics in the band structure. High harmonics are produced in a semiconductor GaSe crystal, which cover the spectral range from mid-infrared to visible, extending beyond the bandgap energy of ~2 eV. We employ ellipsometric detection of emitted harmonics, and observe unusual appearance of the polarization components in the direction perpendicular to the driving mid-infrared field. Detailed analysis shows that the transition from the perturbative to non-perturbative regime occurs around ~10 MV/cm. The polarization rotation of high harmonics reflects the electronic structure of solids, especially the band gradients near the Brillouin-zone boundary. This method, high harmonic ellipsometry, can extract rich information on the electron dynamics and band structures, which is analogous to the high harmonic spectroscopy of atoms and molecules in attosecond sciences.

Third, we demonstrate the sub-cycle resolved transient absorption spectroscopy of a semiconductor GaSe crystal around its bandgap. Under the presence of weak electric-fields (<10 MV/cm), Franz-Keldysh oscillations above the bandgap are clearly observed. At high electric fields (>10 MV/cm), we discover novel absorption features where the absorption spectrum is modulated on the sub-cycle time scale, which suggest strong-field-driven coherent electron dynamics around the bandgap.

This series of experimental works is just the beginning of the strong field physics in condensed matters. Further development of mid-infrared sources and the theory of strongly-driven electronic states will deepen our understanding of extremely fast electron processes in solids and may become a foundation of new optical technologies in future.