Terahertz Nonlinear Spectroscopy in Solids

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Recent development of ultrashort pulse technologies allows us to drive large amplitude motion of electron and ion coherently. The intense terahertz (THz) pulse resonant with the vibration frequency is promising to drive vibrations more directly and in coherent manner. In the case of semiconductors, one may coherently control the electronic system in the sub-level structures of quantum structures with intense THz waves. We recently performed the generation of intense terahertz pulse using tilted wave-front technique with a Ti:Sapphire regenerative amplifier. The maximum electric field is as large as 250 kV/cm, which is strong enough to induce nonlinear optical effects in solids [1]. In this talk, we would like to focus THz nonlinear optical phenomena in semiconductor and molecular vibrations in solids. First, we will show strong spectral modulations in the heavy-hole and light-hole exciton absorption band in ZnSe/ZnMgSSe multiple-quantum-well (MQW) structure induced by intense THz electric field. The peak-shift of exciton absorption clearly indicates deviation from the Stark effect, which implies that the interaction between exciton and THz wave enters the non-perturbative regime [2]. Similar effects have been confirmed in GaAs/AlGaAs MQWs. In this case, we clearly observed crossover from red shift to blue shift for the THz electric field in the heavy hole exciton absorption. These results indicate that AC Stark effect and dynamical Franz-Keldysh effect should be competed in this field strength. We will also report on coherent drive of anharmonic oscillator system. We experimentally demonstrated 20-steps ladder climbing on the anharmonic intermolecular potential in the amino acids micro crystals with intense monocycle terahertz pulse [3]. Absorption spectra show suppression of the absorption peak and enhancement of the low frequency absorption for the incident electric field amplitude. These results are reproduced by simulations based on coherent transition processes between quantum levels in the anharmonic potential. The appearance of such nonlinearity allows us to control macroscopic motion via phase-controlled terahertz pulse.

References

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