Summary of thesis: Impact of intraband transitions in high-order harmonic generation from solids

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High-order harmonic generation (HHG) is a nonlinear optical phenomenon where the strong excitation light pulse generates radiation that contains photons with integer multiples of frequencies of the excitation light, and HHG has already been applied to attosecond-pulse and coherent X-ray generation. HHG from atomic gases were discovered in the 1980s, and their generation mechanism was explained by the 3-step model: It describes the initial tunneling ionization of an electron, the acceleration, and then the recollision process of the electron. Since 2000, HHG from solids has been observed, and intensively studied to realize the highly efficient generation processes and for clarifying the generation mechanism in the non-perturbative regime. Some unique aspects have been observed in HHG from solids that are not observed in gases, such as linear dependence of cutoff energy on excitation intensity, multiple plateau regions, and enhancement of harmonic intensities under elliptically polarized light excitation. It has been considered that solid-state-specific intraband transitions of electrons, in addition to interband transitions, are crucial in the mechanism of HHG from solids. However, the most of studies have been performed on bulk crystals, and the role of intraband and interband transitions in HHG from solids has been unclear.

In this thesis, we clarify the role of intraband transitions by studying semiconductor nanocrystals (NCs) and graphene as samples. We can control the electronic states by the quantum confinement effect in NCs and thus intraband transitions, we can experimentally clarify the relationship between HHG and electronic states. In the study of graphene, we studied HHG using mid-infrared (MIR) pulse excitation in combination with terahertz (THz) pulse, which allows the generation of hot electrons and thus the manipulation of interband transitions. These studies reveal that the coupling of intraband and interband transitions is important for efficient HHG from solids.

Firstly, we discuss the mechanism of HHG with CdSe and CdS NCs. We prepared the NCs whose diameter was precisely controlled in nanoscale region by wet chemical synthesis. When the diameter becomes close to the wavelength of electron wavefunction, the quantum confinement effect allows the change in the electronic states with from a bulk-like continuous band to an atomic-like discrete state. The discrete states with the sub-bandgap energy are expected to suppress the electron motion and thus the intraband transitions for MIR pulse excitation. We observed HHG from a thin film of CdSe NCs under MIR pulse excitation and measured the diameter dependence of HHG efficiency. Though the HHG efficiency does not change significantly up to the size of 3 nm, it sharply reduces below it. We further compared the HHG between CdS and CdSe, whose electrons have different effective masses. Compared them with the same NC size, the HHG reduction in CdS NCs is smaller than that in CdSe and this result indicates that the suppression of

intraband transitions causes the HHG reduction. In other words, since the sub-bandgap is inversely proportional to the effective mass, the sub-bandgap of CdS NC with larger effective mass is smaller than that of CdSe NC for the same diameter. Then the smaller sub-bandgap of CdS NC does not suppress the intraband transitions as much as that of CdSe and thus HHG. The HHG calculations using the 1D dimer chain model also reproduced the observed behaviors. These results indicate that intraband transition is an important origin for HHG in solids.

Secondly, we show the results of HHG from graphene. The pioneering studies in graphene propose that the origin of the HHG enhancement under elliptically polarized light excitation is the gapless band structure, and recently the importance of coupling between intraband and interband transitions is discussed. Here, we study the role of intraband and interband transitions in HHG from graphene by using THz pulse excitation. Without THz excitation, 5th- and 7th-order harmonics were observed. On the other hand, with THz excitation, even-order harmonics appear and odd-order harmonic intensity decreases. Also, though the even-order harmonic intensity follows the time waveform of the THz electric field, the odd-order harmonic intensities remain reduced even after the THz pulse passed with the recovery time of a few picoseconds. The generation of even-order harmonics is attributed to the inversion symmetry breaking caused by the THz electric field, and the decrease in odd-order harmonic intensities is attributed to the generation of hot electrons. Then, we measured the MIR ellipticity dependence of 7th harmonic intensity under THz excitation and we found that the intensity decreases more significantly under elliptically polarized light excitation than that under linearly polarized case. The thermal distribution of hot electron induced by THz excitation follows a Fermi-Dirac distribution, and then the suppression of interband transitions by Pauli blocking is expected to reduce HHG under MIR excitation. Thus, the larger suppression of HHG under elliptically polarized light excitation than linearly polarized light excitation indicate that the lower-order interband transitions is more important for the elliptically HHG enhancement. The calculation results for changing the electron temperature also reproduces the experimental results. Our results show that the suppression of interband transitions by hot electron generation enables the control of HHG.

In this thesis, we study the intraband and interband transitions as the origin of HHG from solids. Our observation of HHG from semiconductor NCs shows the importance of intraband transitions specific to solids. In the HHG from graphene, we show that the interband transitions can be manipulated by hot electrons caused by THz pulse excitation, thus allowing the ultrafast control of the HHG. From our studies, it can be concluded that in solids, the manipulation of electronic states by nanosizing of material or external fields enables the control of intraband and interband transitions and thus HHG.