

Summary of thesis:
High harmonic generation from perovskite manganese oxide
 $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$

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In the thesis, we report a new characteristic of high harmonic generation (HHG) from strongly correlated materials. HHG is an extreme nonlinear optical phenomenon. In the early stage, it has been studied for atomic gasses. With the development of high-power and narrow pulse-width lasers, further investigation on HHG has progressed both in gas and solid materials. In 2011, HHG from a condensed material in the non-perturbative regime was reported in ZnO, where light-matter interaction cannot be regarded as a perturbation. Since then, solid HHG has drew considerable attention, and it has opened the door to the investigation of physical properties of materials that conventional techniques cannot explore. For example, HHG can be used as a tool for band reconstruction and high-spatial-resolution spectroscopy.

Under these circumstances, HHG in strongly correlated materials has attracted much attention, where we cannot neglect the strong interaction between an itinerant electron driven by the intense laser field and other electrons. In these materials, there is an expectation that HHG can extract information on the physical properties of material qualitatively different from well-studied band insulators or semiconductors. So far, a number of theoretical approaches have been proposed to deal with HHG from strongly correlated materials. However, few experimental investigations on HHG in these materials have been reported.

The goal is to find the key factors necessary for understanding HHG from strongly correlated materials. For this purpose, we choose hole-doped manganite $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ (PCMO) as the target system. PCMO system is an example of strongly correlated materials with a coupling between spin, charge, and orbital (lattice) degrees of freedom, and it exhibits a variety of electronic and magnetic phases, depending on hole-doping level x and the temperature. The easily controllable electronic phase enables a comprehensive study of HHG in strongly correlated materials.

Here, we investigated HHG properties relevant to respective magnetic and electronic phases from two compounds; $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$ and $\text{Pr}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$. The compositions of these two materials are not so different. However, they undergo different phase transitions with decreasing temperatures. Hence, the temperature dependence of HHG properties differs in these two compounds. $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$ has charge/orbital-ordered and spin-

ordered ground state. As the temperature decreases, the harmonic intensities gradually increase below the transition temperature where $\text{Mn}^{3+}/\text{Mn}^{4+}$ charge ordering and Mn e_g -orbital ordering can be observed, with further decreasing temperature, it exhibits antiferromagnetic spin ordering. Whereas $\text{Pr}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$ has orbital-ordered and inhomogeneous ferromagnetically spin-ordered ground state.

The high harmonic intensity of PCMO ($x=0.4$) monotonically increases below charge/orbital ordering temperature as the temperature decreases. For PCMO ($x=0.2$), the temperature variation of high harmonic intensity shows a clear peak at the magnetic transition temperature where the spin momentum of Mn aligns ferromagnetically.

Comparing the results of these two compounds, we concluded that to understand the physical properties of HHG from PCMO system, two factors have to be included. One of which has been previously proposed for Mott insulating system. It considers the correlation between the itinerant electrons driven by the light field and the thermal fluctuation of ordered states. Another one is peculiar to the materials exhibiting intrinsic inhomogeneity in their ground state, where metallic-like regions exist in the macroscopically insulating medium. This study demonstrates that HHG from strongly correlated materials is extremely sensitive to the local electronic correlation. Further investigation using HHG spectroscopy may enable us to explore the ambiguous nature of complicated systems.

In Chapter 1, we review some basic properties of HHG in solids and recent studies on HHG from strongly correlated materials. In Chapter 2, we show what has attracted researchers to investigate PCMO, highlighting the importance of investigating HHG from this system. Information on the preparation and characterization of the crystal samples is also included here. In Chapter 3, we describe the experimental method for HHG measurement. In Chapters 4 and 5, we report the HHG measurements performed on PCMO ($x=0.4$) and PCMO($x=0.2$), respectively. In Chapter 6, we discuss the overall experimental results and provide the future aspects of this study.