【Grant-in-Aid for Scientific Research (S)】

Broad Section D

Title of Project : Establishment of strong light field photoscience in solids and application to materials science

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Research Project Number : 21H05017　Researcher Number : 90212034
Term of Project : FY2021-2025　Budget Allocation : 146,400 Thousand Yen
Keyword : Extreme nonlinear optics, High harmonic generation, Strong field physics

【Purpose and Background of the Research】

The purpose of this study is to construct the theory of extreme nonlinear optical phenomena caused by the interaction between high-intensity light fields and solid materials.

From the research so far, it has become clear that **high-order harmonic generation (HHG)** occurs when a solid is irradiated with high-intensity light. In the case of the band insulator for which one electron approximation holds, the physical mechanism is the generation of electron-hole pairs by the tunnel process (Zener-Tunneling) by high-intensity light and the motion under an electric field. It has been also clear that the in-band (inter-band) drive mechanism plays a major role when the frequency of the generated harmonics is lower (higher) than the bandgap of the insulator. According to this scenario, materials with large bandgap are less likely to undergo tunneling processes, so HHG intensities are expected to decrease as the gap energy increases. However, for strongly correlated electron systems such as Mott insulators, we have found a tendency that is completely against such expectations.

Therefore, we decided to study systematically nonlinear optical phenomena under high-intensity light fields such as HHG in strongly correlated electron materials and materials showing charge order, in addition to band insulators for which one electron approximation holds.

【Research Methods】

In order to avoid the destruction of the solid surface by high-intensity light, high-intensity light in the mid-infrared region to terahertz region is utilized, and high-order harmonic generation in a wide wavelength region from the infrared region to the ultraviolet region is frequency-dependent and polarized. We will systematically investigate basic characteristics such as dependence and carrier doping dependence.

In addition, sub-cycle time-resolved spectroscopy and sideband spectroscopy, which can clarify the distribution and coherence of electronic states in one cycle of excitation light, are performed under the condition of high-order harmonic generation. In strongly correlated electron materials, charged particle pair production under high-intensity light fields may be completely different from that of ordinary semiconductors, so we will construct a theory that describes an extreme nonlinear response at the same time.

【Expected Research Achievements and Scientific Significance】

By studying strongly correlated electron materials and materials showing charge order in addition to band insulators, the material parameters that determine the extremely nonlinear optical phenomena that appear under high-intensity light fields will be clarified. This makes it possible to construct the theory of high-strength field photo-science for a wide range of solid materials. We also expect that the understanding will open a pathway to a novel measurement technology to evaluate strongly correlated electron materials.

This research has high academic originality and originality from the following viewpoints:

● Forerunner of HHG study in strongly correlated electron systems

We have clarified for the first time in the world the peculiar behavior of HHG in strongly correlated electron materials.

● Advanced research on HHG using ultra-thin films

In order to minimize the propagation effect (phase matching condition) and absorption loss in the nonlinear optical process, we will measure HHG in the reflection geometry or using ultra-thin films.

● HHG research from the perspective of material science

We will focus on materials with multiple stable phases and measure HHG with changing temperature around the phase transition point. This will clarify the relationship between the material order and the non-linear optical property.

【Publications Relevant to the Project】

・ N. Yoshikawa, T. Tamaya, and K. Tanaka, Science 356, 736-738 (2017).
・ N. Yoshikawa, K. Nagai, K. Uchida, Y. Takaguchi, S. Sasaki, Y. Miyata, K. Tanaka, Nature Communications 10, 3709 (2019).
・ H. Nishidome, K. Nagai, K. Uchida, Y. Ichinose, Y. Yomogida, K. Tanaka and K. Yanagi, Nano Letter 20 (8), 6215-6221 (2020).

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