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

Broad Section B

Title of Project : Explorations of ultrafast quantum phase transitions in strongly correlated electron systems by high-intensity terahertz/mid-infrared pulses

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【Purpose and Background of the Research】

Recently, attempts for rapid controls of electronic structure and physical property of a solid by irradiating it with light (photoinduced phase transition) have been actively made. To achieve the application of this phenomenon to optical switching devices, the key is how to cause changes in physical properties by light in an ultrafast time scale and with high efficiency. From this viewpoint, the principal investigator has been focusing on strongly correlated systems. As typical examples of their photoinduced phase transitions, photoinduced Mott-insulator to metal transition and ionic to neutral transition are known. However, in these phenomena, the temperature rise of the system is often an essential problem in terms of both elucidation of non-equilibrium dynamics and realization of ultrafast switching. A promising method for solving this is the control of physical properties by infrared light.

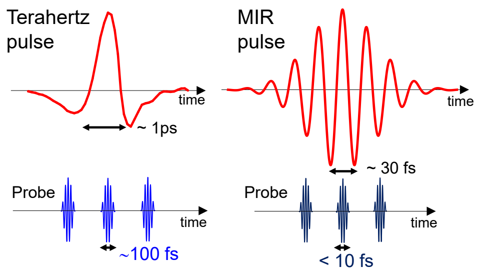
In this study, we will excite strongly correlated systems (Mott insulators, electronic dielectrics, multiferroics, etc.), with terahertz pulses or mid-infrared (MIR) pulses with large amplitudes in the transparent region and explore insulator-metal transition by quantum tunneling process, paraelectric-ferroelectric transition by intermolecular electron transfers, and polarization and magnetization control by a new mechanism.

【Research Methods】

In this study, by irradiating strongly correlated systems with an electromagnetic field pulse with amplitudes several times larger than before, we aim to achieve ultrafast electronic transitions with new mechanisms. There are two items: (1) searching for a new electromagnetic field-induced transition by increasing the intensity of a terahertz pulse, and (2) searching for a new electric field-induced transition by increasing the intensity of an MIR pulse and using a broadband ultrashort probe pulse.

The advantage of a terahertz pulse is that a single-cycle electromagnetic field can be obtained (the left figure). Using this pulse, a strong electric field (magnetic field in the vertical direction) can be applied in a specific direction. In this study, a terahertz pulse with a large electric-field amplitude is generated, and the electronic-state change along the electric field is detected by an ultrashort pulse from the visible to MIR region. By using a pulse with an amplitude much larger than 1 MV/cm, it is possible to induce a phase transition triggered by carrier generation and electron transfer by quantum tunneling processes in various materials. This pulse is also a magnetic field pulse with a large amplitude. However, in simple ferromagnets in which spin conservation law holds, dynamics of magnetization should be slower than picoseconds, so that it is difficult to control magnetization by a magnetic-field component of this pulse alone. In this study, therefore, we aim at control of magnetization that effectively uses an electric field or both an electric field and a magnetic field, focusing on the systems (double exchange or multiferroic systems), in which spin and charge are coupled.

In the MIR region, we can generate an oscillating electromagnetic-field pulse (the right figure) with an amplitude exceeding 10 MV/cm. In this study, we will establish a method to detect the response to this pulse using a probe pulse from the visible to near-IR region with a time width less than 10 fs Using this, we will achieve an electronic phase control based on a new mechanism, such as a phase transition via a floquet state induced under a periodic strong electric field.



【Expected Research Achievements and Scientific Significance】

If we succeed in inducing collective motions of electrons and spins by irradiating a strong electromagnetic field pulse in a strongly correlated systems, we will be able to realize a new quantum phase transition with low energy loss and at ultrafast speed. In most of photo-induced transitions caused by a visible pulse, a transition occurs from ordered phase to less-ordered one. Using the approach of this study, it may be possible to generate novel electronically ordered phases such as charge-ordered phase and electronic-type dielectric phase, which cannot be stabilized in the steady state. Therefore, our study is expected to open a new way for material phase control.

【Publications Relevant to the Project】

- H. Yamakawa *et al*., "Mott transition by an impulsive dielectric breakdown", *Nature Materials* **16**, 1100 (2017).
- T. Miyamoto *et al*., "Probing ultrafast spin-relaxation and precession dynamics in a cuprate Mott insulator with sevenfemtosecond optical pulses", *Nature Communications* **9**, 3948 (2018).
- T. Terashige *et al*., "Doublon-holon pairing mechanism via exchange interaction in two-dimensional cuprate Mott insulators", *Science Advances* **5**, eaav2187 (2019).
- H. Yamakawa et al., "Terahertz-field-induced polar charge order in electronic-type dielectrics", *Nature Communications* **12**, 953 (2021).

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