International Research Center for Elements Science – Nanophotonics –

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Scope of Research

Our research interest is to understand optical and quantum properties of nanometer-structured materials and to establish opto-nanoscience for creation of innovative functional materials. Space- and time-resolved laser spectroscopy is used to study optical properties of semiconductor quantum nanostructures and strongly correlated electron systems in low-dimensional materials. The main subjects are as follows: 1) investigation of optical properties of single nanostructures through the development of a high-resolution optical microscope, 2) ultrafast optical spectroscopy of excited states of semiconductor nanostructures, and 3) photophysics of solar cell materials.

KEYWORDS

Femtosecond Laser Spectroscopy
Semiconductor Nanoparticles
High Harmonic Generation
Single Photon Spectroscopy
Perovskites

Selected Publications

Sanari, Y.; Otobe, T.; Kanemitsu, Y.; Hirori, H., Modifying Agular and Polarization Selection Rules of High-order Harmonics by Controlling Electron Trajectories in k-Space, *Nature Commun.*, **11,** [3069-1]-[3069-7] (2020).

Handa, T.; Tahara, H.; Aharen, T.; Kanemitsu, Y., Large Negative Thermo-optic Coefficients of a Lead Halide Perovskite, *Sci. Adv.*, **5,** [eaax0786-1]-[eaax0786-8] (2019).

Sanari, Y.; Tachizaki, T.; Saito, Y.; Makino, K.; Fons, P.; Kolobov, A. V.; Tominaga, J.; Tanaka, K.; Kanemitsu, Y.; Hase, M.; Hirori, H., Zener Tunneling Breakdown in Phase-Change Materials Revealed by Intense Terahertz Pulses, *Phys. Rev. Lett.*, **121,** [165702-1]-[165702-6] (2018).

Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Quantum Coherence of Multiple Excitons Governs Absorption Cross-Sections of PbS/CdS Core/Shell Nanocrystals, *Nat. Commun.*, **9,** [3179-1]-[3179-8] (2018).

Yamada, T.; Aharen, T.; Kanemitsu, Y., Near-Band-Edge Optical Responses of $CH_3NH_3PbCl_3$ Single Crystals: Photon Recycling of Excitonic Luminescence, *Phys. Rev. Lett.*, **120,** [057404-1]-[057404-6] (2018).

Origin of Negative Thermo-optic Coefficients of Lead Haide Perovskites

Lead halide perovskites are promising materials for flexible and efficient optoelectronic device applications. Recent studies have shown that the temperature dependence of their optical properties is quite different from that of conventional semiconductors. Understanding the origin of the unique temperature dependence is required from both fundamental physics and device application viewpoints. In this project, we clarified the temperature dependence of the refractive indices in lead halide perovskite $CH_3NH_3PbCl_3$. We found that $CH_3NH_3PbCl_3$ possesses negative thermo-optic coefficients for the entire visible wavelength region and its magnitude increases near the absorption edge. Analysis based on the Lorentz oscillator model revealed that the large thermal expansion coefficient leads to the large decrease in the valence electron density upon temperature rise, which consequently results in the unique negative thermo-optic coefficients.

Polarization Control of High-order Harmonics Induced by Two-color Orthogonally Polarized Laser Fields

Irradiation with a high-intensity laser pulse to a material causes light emission with a broad spectrum consisting of many harmonics of the excitation laser frequency. This phenomenon is called high-order harmonic generation (HHG). HHG has been actively studied toward new optical technologies, but the mechanisms of HHG are still under debate. In our experiments, in order to understand HHG mechanisms, a GaSe crystal was simultaneously excited by two orthogonally polarized optical pulses with wavelengths of λ_1 = 2400 nm and λ_2 = 1300 nm (Figure 1). We observed a broadband emission and multiple peaks originating from higher-order harmonic generation. Furthermore, it was clarified that the polarization of higher-order harmonics is determined by the motion of electrons that are strongly driven two-dimensionally by the electric field of orthogonal laser light. These results will lead not only to the technology for controlling the properties of high-order harmonics, but also to the ultrafast optoelectronics controlled by the electric field of laser light.

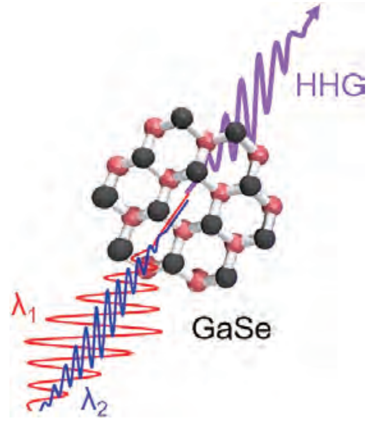

Figure 1. Experimental setup. Two orthogonally polarized pulses are focused on the sample.

Improvement of Optical Gain in Halide Perovskite Nanocrystals by Double Pump Method

Lead halide perovskite nanocrystals are expected for light-emitting device materials because they have superior optical properties. They exhibit high photoluminescence quantum yields and emission wavelength tunability by controlling the particle size and halide composition. However, a reabsorption process caused by exciton-exciton interactions suppresses the optical gain in photoexcited nanocrystal ensembles. This is one of the serious problems for realizing nanocrystal-based lasers. We evaluated the reabsorption process and the optical gain threshold of $CsPbI_3$ nanocrystals by double-pump transient absorption spectroscopy. Our study revealed that the efficient generation of asymmetric-hot biexciton states by the double-pump method is beneficial to suppress the reabsorption process and reduce the optical gain threshold (Figure 2).

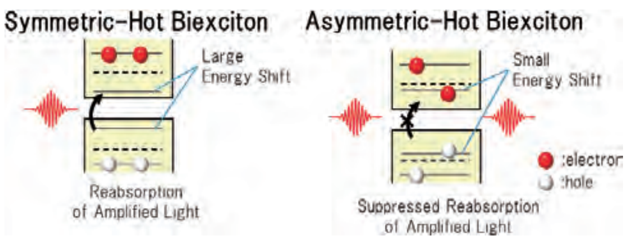

Figure 2. Schematics of the reabsorption processes in a symmetric-hot biexciton and an asymmetric-hot biexciton.