International Research Center for Elements Science – Nanophotonics –

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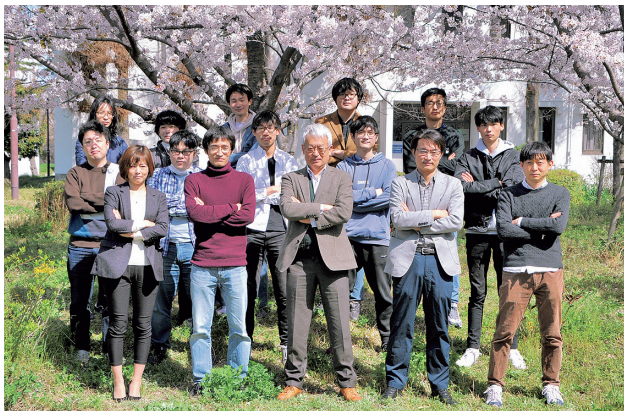
Scope of Research

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, 3) photophysics of solar cell materials, 4) engineering material properties with lights.



KEYWORDS

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

Recent Selected Publications

Nakagawa, K.; Hirori, H.; Sato, S. A.; Tahara, H.; Sekiguchi, F.; Yumoto, G.; Saruyama, M.; Sato, R.; Teranishi, T.; Kanemitsu, Y., Size-Controlled Quantum Dots Reveal the Impact of Intraband Transitions on High-Order Harmonic Generation in Solids, *Nature Phys.*, **18,** 874-878 (2022).

Handa, T.; Hashimoto, R.; Yumoto, G.; Nakamura, T.; Wakamiya, A.; Kanemitsu, Y., Metal-Free Ferroelectric Halide Perovskite Exhibits Visible Photoluminescence Correlated with Local Ferroelectricity, *Sci. Adv.*, **8,** [eabo1621-1]-[eabo1621-8] (2022).

Yumoto, G.; Hirori, H.; Sekiguchi, F.; Sato, R.; Saruyama, M.; Teranishi, T.; Kanemitsu, Y., Strong Spin-Orbit Coupling Inducing Autler-Townes Effect in Lead Halide Perovskite Nanocrystals, *Nature Commun.*, **12,** [3026-1]-[3026-7] (2021).

Sekiguchi, F.; Hirori, H.; Yumoto, G.; Shimazaki, A.; Nakamura, T.; Wakamiya, A.; Kanemitsu Y., Enhancing the Hot-Phonon Bottleneck Effect in a Metal Halide Perovskite by Terahertz Phonon Excitation, *Phys. Rev. Lett.*, **126,** [077401-1]-[077401-6] (2021).

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).

Ultrafast Spatiotemporal Dynamics of Exciton Spins in a Two-Dimensional Layered Halide Perovskite at Room Temperature

Along with their excellent optoelectronic properties, two-dimensional (2D) layered halide perovskites (LHPs) have unique spintronic properties. 2D LHPs have stable excitons with spins of ±1 and exhibit substantial exciton-exciton interactions and relatively long exciton spin relaxation times at room temperature. These properties provide potential for realizing room-temperature exciton spin transport. To investigate the spatiotemporal evolution of exciton spins, we developed polarization-resolved pump-probe microscopy with millidegree, submicrometer, and subpicosecond resolutions and performed time-resolved Faraday rotation imaging of room-temperature spin-polarized excitons in 2D LHP single crystals of $(C_4H_9NH_3)_2(CH_3NH_3)_3Pb_4I_{13}$. Under strong photoexcitation conditions, we observed that as the pump-probe delay time increases, a ring-like spatial pattern emerges in the spin-polarized exciton population and simultaneously ultrafast exciton spin transport occurs. Furthermore, we found that the anomalous spatiotemporal dynamics of the exciton spins originates from exciton-exciton exchange interactions. Our findings reveal the potential of 2D LHPs for room-temperature spin-optoelectronic applications.

Observation of High-Order Harmonic Generation from Semiconductor Quantum Dots

High-order harmonic generation (HHG) is attracting much attention as a new spectroscopic tool for understanding ultrafast carrier dynamics under strong electric fields and has a great potential for a coherent light source in the extreme ultraviolet region. Recently, the intensive studies on HHG from solids have been conducted both theoretically and experimentally and it is known that the mechanism of HHG from solids is completely different from that from gases. The HHG mechanism in solids is theoretically discussed in terms of two processes, the intraband and interband transitions, but there have been no experimental studies on the control of these two processes. To clarify the role of two transitions in the HHG efficiency, we investigated HHG from CdSe and CdS quantum dots. We can control the intraband transition process by utilizing the quantum confinement effects of dots. We observed that the high-order harmonic (HH) intensity increases with the dot size (Fig. 1). Our findings reveal that the intraband transitions play an important role in HHG from solids.

Exciton-Phonon and Trion-Phonon Couplings in Single $CsPbBr_3$ Perovskite Nanocrystals

Lead halide perovskite nanocrystals (NCs) are attracting significant attention as light-emitting device materials because of their superior optical properties such as high photoluminescence (PL) quantum yields and bandgap tunability. In these NCs, exciton-phonon couplings strongly affect their optical properties. However, the mechanism of exciton-phonon coupling in perovskite NCs remains unclear. Very narrow PL lines at low temperatures in the NCs enable LO-phonon-mode-resolved analysis of exciton-phonon couplings. We studied the PL spectra of single $CsPbBr_3$ NCs by using single-dot spectroscopy at 5.5 K to evaluate the strengths of exciton-phonon couplings (the Huang-Rhys factors). We observed that PL of trions becomes more dominant than that of excitons under strong photoexcitation conditions and LO-phonon replicas of trions clearly appear in addition to those of excitons (Fig. 2). Both the Huang-Rhys factors of excitons and trions become larger for smaller NCs. We also found that the size dependence of the Huang-Rhys factors for trions are weaker than that for excitons. These results are caused by the difference in the charge distributions of excitons and trions in the NCs.

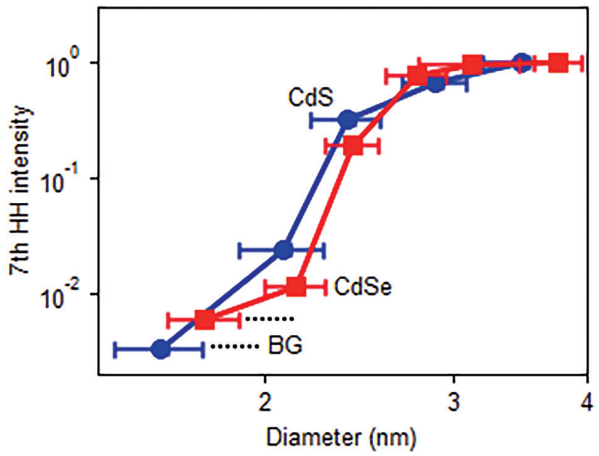


Figure 1. Diameter dependence of 7th HH intensity.

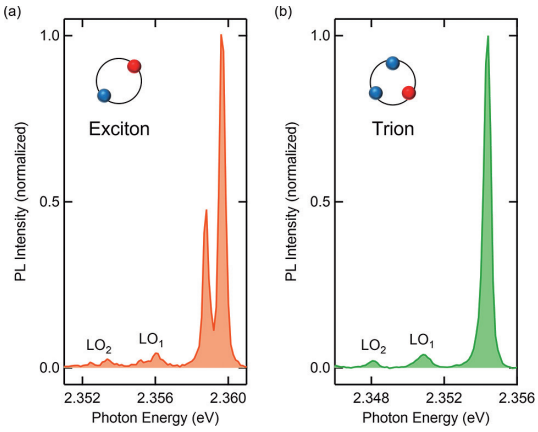


Figure 2. PL peaks of (a) exciton and (b) trion with their LO-phonon replicas.