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, and 4) engineering material properties with lights.

KEYWORDS

Femtosecond Laser Spectroscopy
Quantum Dots
High Harmonic Generation
Single Photon Spectroscopy
Perovskites

Recent Selected Publications

Zhang, Z.; Sekiguchi, F.; Moriyama, T.; Furuya, S. C.; Sato, M.; Satoh, T.; Mukai, Y.; Tanaka, K.; Yamamoto, T.; Kageyama, H.; Kanemitsu, Y.; Hirori, H., Generation of Third-Harmonic Spin Oscillation from Strong Spin Precession Induced by Terahertz Magnetic Near Fields, *Nature Commun.*, **14,** [1795-1]-[1795-7] (2023).

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).

Topics

The Second- and Third-Harmonic Generation from Spin Precession in a Canted Antiferromagnet

Recently, antiferromagnetic spintronics is considered as one of the important next-generation information technologies, owing to the low-loss and non-volatility of spin properties. Therefore, the spin dynamics in antiferromagnets with the resonance at terahertz (THz) frequencies, has attracted much interest. However, because of the lack of strong THz magnetic fields, the THz spin dynamics has not yet been well understood. In this study, we developed a novel micro-resonator to efficiently enhance the magnetic component of THz wave, which allows the generation of the world's strongest THz magnetic field up to 2 Tesla in an antiferromagnet $HoFeO_3$.[1)] By using this strong THz magnetic field, we succeeded in inducing the unprecedentedly large magnetization change and thus generating the second and third harmonics of fundamental spin frequency (Fig. 1), for the first time. Furthermore, our research provides a deep understanding of relationship between the harmonics and the spin structure. The efficient excitation method of spin dynamics and the understanding of nonlinear spin dynamics can help us establish novel nonlinear spintronics at terahertz frequencies.

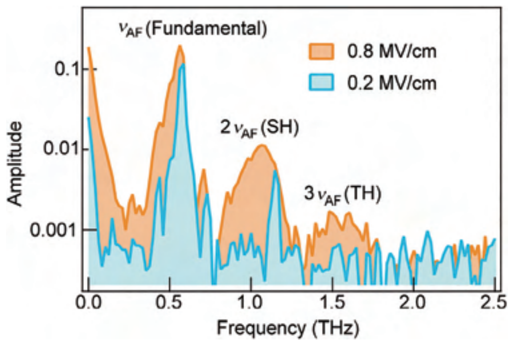

Figure 1. Spectrum of spin dynamics. Different colors indicate different excitation powers.

Size-Dependent Structural Phase Transition in Single Lead Halide Perovskite Nanocrystals

Lead halide perovskite nanocrystals (NCs) have attracted attention as a new light-emitting material because of their high photoluminescence (PL) quantum yields and bandgap tunability around the whole visible region. Halide perovskites have three crystal structures (orthorhombic, tetragonal, and cubic), and the PL peak energy depends strongly on the crystal structure. However, it is unclear how the NC size affects the phase transition temperature. In this study, we investigated the temperature dependence of the PL spectra in single perovskite $APbBr_3$ (A = FA ($HC(NH_2)_2$) and Cs) NCs from 5.5 to 200 K using single-dot PL spectroscopy.[2)] For $FAPbBr_3$ NCs, a redshift in PL peak energy due to the orthorhombic to tetragonal phase transition is observed between 100 and 150 K, while $CsPbBr_3$ NCs show no phase transition within the observed temperature range. We found that the phase transition temperature decreases in smaller $FAPbBr_3$ NCs (Fig. 2).

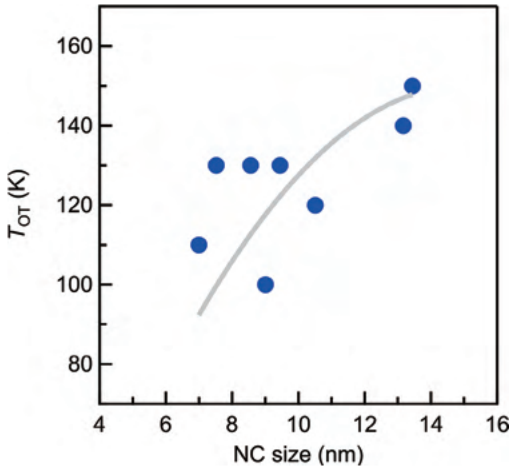

Figure 2. Size dependent orthorhombic-to-tetragonal phase transition temperature (T_{OT}) in $FAPbBr_3$ NCs.

Anomalous Band-Edge Optical Responses in a Two-Dimensional Ferroelectric Halide Perovskite

In two-dimensional (2D) halide perovskites, electrons and holes confined in atomically thin 2D perovskite layers form excitons with extremely large binding energies. The peculiar optical properties emerge from such stable excitons. In addition, because the lattice distortion of the perovskite structure can be controlled by the organic molecular cations contained in 2D halide perovskites, the unique structural properties appear: Large lattice distortion introduced by large-sized organic cations results in the emergence of ferroelectricity in 2D halide perovskites. Therefore, clarifying the correlation between the crystal structure and optical properties is important for understanding the physical properties of ferroelectric halide perovskites and developing new devices. In this study, we measured the temperature dependences of the absorption spectra, second harmonic generation (SHG) intensity, and lattice constants of 2D ferroelectric perovskite $(BA)_2(EA)_2Pb_3I_{10}$ single crystals (BA is n-butylammonium and EA is ethylammonium).[3)] We found the anomalous temperature behavior of the Urbach tail of the absorption spectrum exhibiting a significant change at around 200 K. We revealed that this change is correlated with the temperature dependences of the SHG intensity and the in-plane lattice distortion. We concluded that the randomness of the ferroelectric polarizations modifies the linewidth of the band-edge exciton transition and leads to the anomalous temperature dependence of the steepness parameter of the Urbach tail.

References

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2) K. Cho, T. Yamada, M. Saruyama, R. Sato, T. Teranishi, and Y. Kanemitsu, *J. Chem. Phys.* **158**, 201104 (2023).

3) C. Higashimura, G. Yumoto, T. Yamada, T. Nakamura, F. Harata, H. Hirori, A. Wakamiya, and Y. Kanemitsu, *J. Phys. Chem. Lett.* **14**, 8360 (2023).