

# International Research Center for Elements Science – Nanophotonics –

<https://www.scl.kyoto-u.ac.jp/~opt-nano/index-e.html>



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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    Single Photon Spectroscopy  
Semiconductor Nanoparticles        Perovskites  
High Harmonic Generation

## Recent Selected Publications

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.; Ootobe, T.; Kanemitsu, Y.; Hirori H., Modifying Angular and Polarization Selection Rules of High-order Harmonics by Controlling Electron Trajectories in k-Space, *Nature Commun.*, **11**, [3069-1]-[3069-7] (2020).  
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<sub>3</sub>NH<sub>3</sub>PbCl<sub>3</sub> Single Crystals: Photon Recycling of Excitonic Luminescence, *Phys. Rev. Lett.*, **120**, [057404-1]-[057404-6] (2018).

## THz Pulse Excitation Generates Hot Carriers by Hot Phonon Bottleneck Effect

Halide-perovskite semiconductors are a promising material for future optoelectronic devices such as solar cells, and their excellent photocarrier properties are thought to originate from the characteristic electron-phonon interactions and phonon-mode characters. In this study, we explored the potential of THz pulses, which can resonantly excite the low-energy optical phonons, to modulate the photocarrier dynamics in  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite films. After THz pulse excitations, their impact on the photoinduced carrier dynamics was investigated by time-resolved photoluminescence (PL) spectroscopy. We observed the PL intensity was strongly modulated upon THz excitation in the picosecond time scale. Time-resolved PL spectra showed that the carrier temperature transiently increased, and furthermore, the PL intensity with high photon energies was enhanced by THz pulse excitation. These features indicate that the lifetime of hot carriers is enhanced by the phonon excitation induced by THz pulses, through the mechanism of hot-phonon bottleneck effect. These first observations suggest that the phonon systems, which are usually regarded as a passive heat bath, can be utilized as a new degree of freedom to efficiently harness photocarrier energy.

## Ultrastrong Coupling between Phonons in Halide Perovskites and THz Vacuum Photons

When a matter is strongly coupled to the light field, a hybridized state emerges and behaves differently from the original electronic state. As the coupling strength increases, the hybrid system reaches the regime of ultrastrong coupling (USC). Especially, the USC between phonons and the vacuum field in a cavity is attracting much attention, because it opens up a novel way to material manipulation. However, this phononic USC remains difficult to achieve in solids because of the small dipole moments of phonon excitations. In our study, we found a way to increase coupling strength by enhancing the vacuum field, and thus realized the phononic USC. The system is made of the cavity of split ring resonator (SRR) and the phonon modes of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite semiconductor films at THz frequencies (Figure 1). Our analysis reveals that as the gap of SRR is narrowed, the enhancement of the vacuum field overcomes the decrease of the dipole number in a gap, finally resulting in the USC in a nano-size gap.

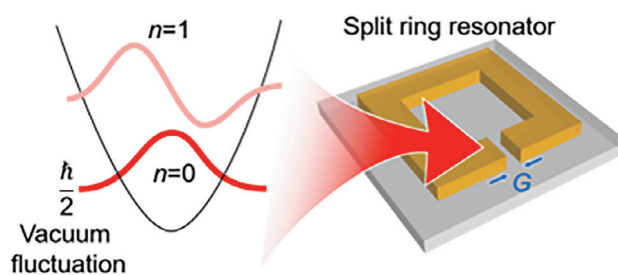


Figure 1. Image of vacuum field in a split ring resonator.

## Size-dependent Exciton-phonon Coupling in Single Lead-halide Perovskite Nanocrystals

Lead-halide perovskite nanocrystals (NCs) are promising optical materials for use in several light-emitting devices. Recently, it has been reported that single perovskite NCs are suitable for single-photon sources because of their stable light emission and long exciton coherence time. In particular, exciton coherence time is strongly affected by the exciton-phonon coupling. To reveal the exciton-phonon coupling in perovskite NCs, we studied the NC size dependence of the PL spectra of  $\text{FAPbX}_3$  (FA =  $\text{HC}(\text{NH}_2)_2$ , X = Br, I) NCs by using single-dot spectroscopy at liquid-helium temperature. We found four peaks below the exciton PL peak. By measuring size dependence of the PL peak energy and PL intensity, we concluded that they consist of two LO-phonon replicas of the exciton and emissions of charged exciton (trion) and biexciton (Figure 2). The binding energies of biexcitons and trions are size-dependent because of enhanced Coulomb interaction in smaller NCs. The LO-phonon energies are size-independent, but the strength of exciton-phonon coupling (the Huang-Rhys factors) becomes larger in smaller NCs. This phenomenon is probably due to the size-dependent dielectric constants and spatial separation of the electron and hole wavefunctions.

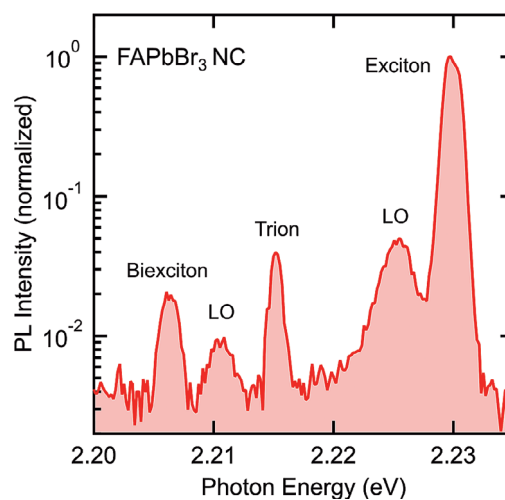


Figure 2. PL spectrum of a single perovskite nanocrystal.