International Research Center for Elements Science – Nanophotonics –

http://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) development of nanoparticle assemblies with new optical functionalities, and 3) ultrafast optical spectroscopy of excited states of semiconductor nanostructures.

KEYWORDS

Femtosecond Laser Spectroscopy
Semiconductor Nanoparticles
Perovskites
Single Photon Spectroscopy
Solar Cells

Selected Publications

Tahara, H.; Sakamoto, M.; Teranishi, T.; Kanemitsu, Y., Harmonic Quantum Coherence of Multiple Excitons in PbS/CdS Core-shell Nanocrystals, *Phys. Rev. Lett.*, **119**, 247401/1-6 (2017).

Yamada, T.; Yamada, Y.; Nishimura, H.; Nakaike, Y.; Wakamiya, A.; Murata, Y.; Kanemitsu, Y., Fast Free-Carrier Diffusion in $CH_3NH_3PbBr_3$ Single Crystals Revealed by Time-Resolved One- and Two-Photon Excitation Photoluminescence Spectroscopy, *Adv. Electron. Mater.*, **2,** [1500290-1]-[1500290-5] (2016).

Ihara, T.; Kanemitsu, Y., Absorption Cross-section Spectrum of Single CdSe/ZnS Nanocrystals Revealed Through Photoluminescence Excitation Spectroscopy, *Phys. Rev. B*, **92,** [155311-1]-[155311-5] (2015).

Yamada, Y.; Yamada, T.; Le, P. Q.; Maruyama, N.; Nishimura, H.; Wakamiya, A.; Murata, Y.; Kanemitsu, Y., Dynamic Optical Properties of $CH_3NH_3PbI_3$ Single Crystals as Revealed by One- and Two-photon Excited Photoluminescence Measurements, *J. Am. Chem. Soc.*, **137,** 10456-10459 (2015).

Yamada, Y.; Nakamura, T.; Endo, M.; Wakamiya, A.; Kanemitsu, Y., Photocarrier Recombination Dynamics in Perovskite $CH_3NH_3PbI_3$ for Solar Cell Applications, *J. Am. Chem. Soc.*, **136,** 11610-11613 (2014).

Observation of Ultrafast Multiexciton Coherence in PbS Nanocrystals Probed by Using a Phase-locked Interference Detection Technique

Generation and recombination dynamics of multiexcitons in nanocrystals have been investigated to understand fundamental physics and use them for device applications. However, their initial generation processes have not yet been clarified. To directly observe ultrafast generation processes of multiexcitons, we developed a new pump-probe system using a phase-locked interference detection technique. Using this system, we successfully observed dipole oscillations of multiexcitons. Furthermore, we found that high-frequency dipoles, i.e., harmonic dipole oscillations, are generated by multiexcitons, even though excitation photon energy is set to the low-frequency exciton resonance. These results provide important insight into multiexciton generation mechanisms.

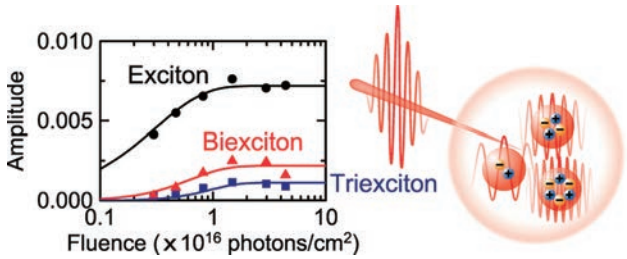

Figure 1. Amplitudes of multiexciton dipole oscillations.

Charged Exciton and Biexciton Dynamics in $CsPbBr_3$ Perovskite Nanocrystals

Lead halide perovskite nanocrystals are attracting much attention owing to their highly luminescent properties. However, nonradiative recombination processes such as Auger recombination of charged excitons (trions) and biexcitons have not yet been understood sufficiently. In this study, we clarified that relaxation dynamics of photoexcited carriers in $CsPbBr_3$ nanocrystals is dominated by three components of exciton, charged exciton, and biexciton, using femtosecond transient-absorption (TA) and single-dot second-order photon correlation spectroscopy. The relaxation lifetimes of charged excitons and biexcitons are determined to be ~40 ps and ~200 ps, respectively. Furthermore, we confirmed the existence of charged excitons even under weak photoexcitation based on the second-order photon correlation measurements.

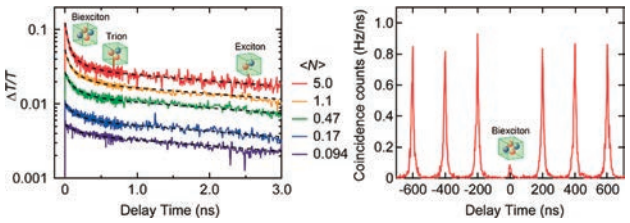

Figure 2. Transient absorption dynamics and second-order photon correlation of $CsPbBr_3$ nanocrystals.

Charge Extraction Processes in $CH_3NH_3PbI_3$ Perovskite Solar Cells

Lead halide perovskites are promising candidates for cost-effective electronic devices including solar cells and light-emitting diodes. In this work, we investigated the photocarrier extraction dynamics in high-efficiency $CH_3NH_3PbI_3$ perovskite solar cells by means of time-resolved photoluminescence (PL) and photocurrent (PC) measurements. We found a peculiar slowdown in the PL lifetime of the perovskite layer for high laser excitation powers, followed by a decrease in the external quantum efficiency of PC. This result indicates that the carrier-extraction process experienced a bottleneck under strong light illumination, limiting the device performance. We conclude that the carrier-extraction rate is sensitive to the photocarrier density, and consequently, the carrier-extraction bottleneck strongly enhances photocarrier recombination losses in the perovskite layer.

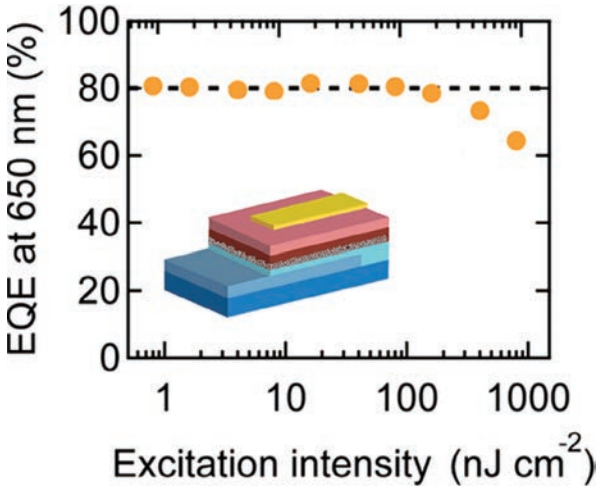

Figure 3. Excitation intensity dependence of external quantum efficiency of perovskite solar cells.