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

KEYWORDS

Femtosecond Laser Spectroscopy
Single Photon Spectroscopy
Semiconductor Nanoparticles
Solar Cells
Perovskites

Selected Publications

Yamashita, D.; Handa, T.; Ihara, T.; Tahara, H.; Shimazaki, A.; Wakamiya, A.; Kanemitsu, Y., Charge Injection at the Heterointerface in Perovskite $CH_3NH_3PbI_3$ Solar Cells Studied by Simultaneous Microscopic Photoluminescence and Photocurrent Imaging Spectroscopy, *J. Phys. Chem. Lett.*, **7,** 3186-3191 (2016).

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).

Biexciton Cascade Emission and Absorption Cross Section of Single Semiconductor Nanocrystals

The sequential two-photon emission process known as biexciton cascade emission is a characteristic phenomenon that occurs in photoexcited semiconductor nanocrystals (NCs). This process occurs when a biexciton state is created in the NCs; thus, the occurrence of the process is related to the photoabsorption properties of the NCs. We presented a simple equation that connects the photoabsorption of single NCs and the biexciton cascade emission. The equation is found to be independent of the quantum yields of photoluminescence (PL). With this equation and using an analysis of second-order photon correlation, the absolute absorption cross section of the single NCs could be evaluated, obtaining values on the order of 10^{-14} cm^2.

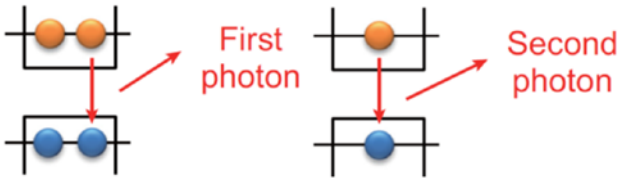

Figure 1. Schematic of the biexciton cascade emission process.

Microscopic Photoluminescence and Photocurrent Imaging of $CH_3NH_3PbI_3$ Perovskite Solar Cells

Metal halide perovskite semiconductors are attracting much attention owing to their high photovoltaic properties in solution-processed solar cells. Detailed understanding of carrier recombination and transport processes is needed to further improve the power conversion efficiency. We analyzed these processes in perovskite solar cells by microscopic imaging of time-resolved photoluminescence (TR-PL) and photocurrent (PC). Simultaneous measurements of TR-PL and PC show a positive correlation between PL intensity and PL lifetime, and a negative correlation between PL and PC intensities. We found that these correlations are caused by the competition between the photocarrier injection from the perovskite layer to the charge transport layer and the photocarrier recombination in the perovskite layer.

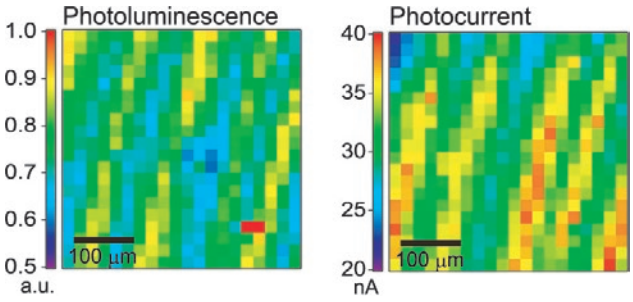

Figure 2. Microscopic image of photoluminescence and photocurrent intensities in perovskite solar cell.

Free Carriers Versus Excitons in $CH_3NH_3PbI_3$ Perovskite Thin Films at Low Temperatures

Organometal halide perovskites are an excellent class of materials for developing cost-effective electronic devices including solar cells, light-emitting diodes, and lasers. Therefore, insightful knowledge of photocarrier recombination dynamics in perovskites, which determines fundamentally performance of perovskite-based devices, is important. We investigated the dynamic responses of photocarriers in $CH_3NH_3PbI_3$ ($MAPbI_3$) thin films at low temperatures using various time-resolved laser spectroscopy. The spectroscopic data consistently indicate that the high-temperature tetragonal phase remains in $MAPbI_3$ films. We observed a fast charge transfer from the major orthorhombic phase to the minor tetragonal phase, which likely prevents the formation of excitons in the orthorhombic phase. Thus, the photocarrier recombination dynamics in two phases could be described by a free-carrier model, rather than an exciton model, even at low temperatures.

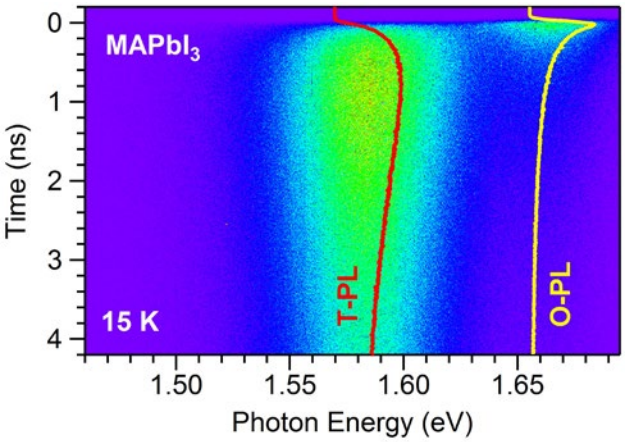

Figure 3. Two-dimensional contour image of a time-resolved photoluminescence spectrum of $MAPbI_3$ thin films at 15 K.