International Research Center for Elements Science – Photonic Elements Science –

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. Optical properties of semiconductor quantum nanostructures and strongly-correlated electron systems in lowdimensional materials are studied by means of space- and time-resolved laser spectroscopy. The main subjects are as follows: (1) Investigation of optical properties of single nanostructures through the development of 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 Carbon Nanotubes Semiconductor Nanoparticles Transition Metal Oxides Semiconductor Nanostructures



Selected Publications

Matsunaga, R.; Matsuda, K.; Kanemitsu, Y., Observation of Charged Excitons in Hole-doped Carbon Nanotubes Using Photoluminescence and Absorption Spectroscopy, *Phys. Rev. Lett.*, **106**, [037404-1]-[037404-4] (2011).

Yamada, Y.; Yasuda, H.; Tayagaki, T.; Kanemitsu, Y., Temperature Dependence of Photoluminescence Spectra of Undoped and Electrondoped SrTiO₃: Crossover from Auger Recombination to Single-carrier Trapping, *Phys. Rev. Lett.*, **102**, [247401-1]-[247401-4] (2009).

Matsunaga, R.; Matsuda, K.; Kanemitsu, Y., Evidence for Dark Excitons in a Single Carbon Nanotube Due to the Aharonov-Bohm Effect, *Phys. Rev. Lett.*, **101**, [147404-1]-[147404-4] (2008).

Hosoki, K.; Tayagaki, T.; Yamamoto, S.; Matsuda, K.; Kanemitsu, Y., Direct and Stepwise Energy Transfer from Excitons to Plasmons in Close-packed Metal and Semiconductor Nanoparticle Monolayer Films, *Phys. Rev. Lett.*, **100**, [207404-1]-[207404-4] (2008).

Quantized Auger Recombination of Biexcitons in Si_{1-x}Ge_x Nanocrystals

Fabrication and characterization of semiconductor nanocrystals (NCs) have been extensively studied due to interest both in the fundamental physics and potential applications in optoelectronic devices. We studied dynamics of quantized Auger recombination in $\text{Si}_{1-x}\text{Ge}_x$ NCs embedded in SiO_2 films by femtosecond intraband pump-probe spectroscopy. The temporal change of the electron-hole pair number under strong photoexcitation was well explained by the quantized Auger recombination model that considered the size distribution of NCs. On the basis of the dependence of the Auger decay rate on temperature and Ge composition, we confirmed the occurrence of breakdown of the *k*-conservation rule in quantized Auger recombination in Si and Si_{1-x}Ge_x NCs.



Figure 1. Time-resolved transmission spectra in Si nanocrystals.

Dynamics of Exciton-hole Recombination in Hole-doped Carbon Nanotubes

Carbon nanotubes are one of the excellent materials for studying the optical properties of excitons, because of their unique band structures and large exciton binding energies. We studied the exciton decay dynamics in holedoped single-walled carbon nanotubes (SWCNTs) by using femtosecond pump-probe transient absorption (TA) spectroscopy. By the doping of SWCNTs with holes, a fast decay component with the lifetime of a few picoseconds appears in TA signals, which corresponds to exciton decay through the Auger recombination between an exciton and a hole and the trion formation. We revealed that this exciton decay rate is quantized by the number of holes in a single SWCNT. The number of holes of a holedoped SWCNT is successfully evaluated on the basis of TA decay dynamics.



Figure 2. Schematic illustration of charged excitons in carbon nanotubes.

Carrier Extraction from Ge/Si Quantum Dot Solar Cells

Quantum dots (QDs) have attracted attention because of their interesting physical properties and potential applications in optoelectronic devices such as light emitters and solar cells. In QDs, physical processes of generation, relaxation, and recombination of carriers are determined by their nanostructures and differ from those in bulk crystals. We report studies of the carrier extraction mechanism in Si solar cells with Ge QDs, which enable the optical absorption of photons with energies below the band gap of the host. Photocurrent measurements revealed that the photocurrent in the QD solar cells increased superlinearly with increasing excitation intensity under strong photoexcitation, which differed greatly from the behavior of Si solar cells without Ge QDs. This nonlinear photocurrent generation indicates that the carrier extraction efficiency from QDs is enhanced under strong photoexcitation by nonlinear carrier extraction processes, such as two-step photon absorption and hot carrier generation via Auger recombination.



Figure 3. Schematic illustration of Ge/Si QDs (left). TEM image of Ge QD array (right).