



Title	Studies of photoinduced electron transfer and exciton dynamics in halide perovskite films and single particles [an abstract of dissertation and a summary of dissertation review]
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Citation	北海道大学. 博士(環境科学) 甲第15137号
Issue Date	2022-09-26
Doc URL	http://hdl.handle.net/2115/87515
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Type	theses (doctoral - abstract and summary of review)
Additional Information	There are other files related to this item in HUSCAP. Check the above URL.
File Information	Bhagyashree_Mahesha_Sachith_abstract.pdf (論文内容の要旨)



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学 位 論 文 内 容 の 要 旨

博士 (環境科学)

氏 名 Bhagyashree Mahesha Sachith

学 位 論 文 題 名

Studies of photoinduced electron transfer and exciton dynamics in halide perovskite films and single particles

(ハロゲン化物ペロブスカイト膜と単一粒子における光誘起電子移動と励起子ダイナミクスの研究)

Lead halide perovskites become the most fascinating semiconductor materials for light-harvesting and light-emitting applications. These materials in the nanocrystalline forms obtained by reliable colloidal synthesis approaches show high photoluminescence quantum yield, high charge carrier mobilities, longer photoluminescence lifetimes, and high photostability. However, their exciton and charge carrier properties and interfacial electron transfer dynamics in self-assembled films and single particles need optimization for next-generation perovskite devices. This study mainly focuses on the electron donor-acceptor systems involving perovskite nanocrystal films and single-particles. I study the exciton, charge carrier, and electron-transfer dynamics of perovskite films and single nanocrystals by addressing the current issues related to interfacial charge transfer processes in perovskite solar cells. This thesis is summarized in five chapters.

In **chapter 1**, I discuss the general properties and significance of lead halide perovskites. I introduce their history followed by the structural composition and stability factors. I explain various synthesis methods for perovskite nanocrystals to control their shape and dimensionality. The preparation methods for self-assembled perovskite nanocrystal thin films and different characterization techniques are discussed in the second section of this chapter. In the third section, complete details about the band-gap properties and fundamental optical properties are presented. Also, I describe the charge carrier properties owing to the quantum confinement in closely packed films. In the final section, I explain the applications of halide perovskites to solar cells, photodetectors, and light-emitting devices (LEDs) followed by my research motivation and objectives.

In **chapter 2**, I provided complete details about the materials, synthesis methods, samples, and instrumentation techniques in this study. Perovskite nanocrystals are synthesized by various methods such as hot injection, ligand-assisted reprecipitation, and modified spray technique. Next, I give a detailed ex

planation of the working principles and instrumental setups of various spectroscopic (UV-vis absorption, steady-state and time-resolved fluorescence spectroscopy, and transient absorption spectroscopy) and microscopic (single-particle fluorescence microscopy, transmission electron microscopy, and scanning electron microscopy) techniques in this study.

In **chapter 3**, I summarize the extent of carrier diffusion, the degree of radiative loss, and the rate of diffusion-controlled interfacial electron transfer in heterojunction films of cesium or formamidinium lead bromide nanocrystals with C_{60} or TiO_2 . The electron transfer and charge separation were confirmed by measuring the photoluminescence lifetime and intensity, and transient absorption spectra.

In **chapter 4**, I demonstrate the electron transfer dynamics at the single-particle level by analyzing the photoluminescence blinking of single perovskite nanocrystals treated with tetracyanoquinodimethane or tetracyanobenzene molecules. The electron transfer rates are determined from time-resolved spectroscopy and the Gibbs free energy change is calculated using the differential pulse voltammetry. Further, the statistical analysis of > 450 single perovskite nanocrystals, and the ON-time and OFF-time probability distributions help confirm the electron transfer process.

In **chapter 5**, I combined all the aspects of exciton plasmon interactions by exploring the photophysical properties of perovskite nanocrystals on Au nanoparticle films and show a huge photoluminescence enhancement. The interactions significantly change the optical properties of perovskite nanocrystals. I discuss the dynamics of photoluminescence enhancement and quenching for perovskite nanocrystals with or without surface ligands in terms of chemical coupling of Au-Br and localized surface plasmon of Au. The significant increase in the OFF time of the plasmon coupled state and the decrease in the photoluminescence lifetime and transient kinetics are discussed precisely. The radiative recombination in the perovskite nanocrystals is dramatically altered by coupling with the Au localized surface plasmon resonance. The proximity of perovskite nanocrystals to Au nanoparticles increases the decay rate. This change in the excited-state relaxation results in remarkable increases in the photoluminescence intensity and decreases in the photoluminescence lifetime. The finite-difference time-domain simulations confirm the near-field intensity and hot spots at the boundary corner between perovskite nanocrystals and Au nanoparticles.