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| Title | Exploring exciton recombination, migration, and transfer in halide perovskite systems [an abstract of dissertation and a summary of dissertation review] |
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| Degree Grantor | 北海道大学 |
| Degree Name | 博士(環境科学) |
| Dissertation Number | 甲第16235号 |
| Issue Date | 2025-03-25 |
| Doc URL | https://hdl.handle.net/2115/95354 |
| Rights(URL) | https://creativecommons.org/licenses/by/4.0/ |
| Type | doctoral thesis |
| File Information | KHATUN_Farida_abstract.pdf, 論文内容の要旨 |

学 位 論 文 内 容 の 要 旨/Dissertation Abstract

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学 位 論 文 題 名/Title of Dissertation

Exploring exciton recombination, migration, and transfer in halide perovskite systems
（ハロゲン化物ペロブスカイト系における励起子の再結合、移動、および転移の探求）

Lead halide perovskites have emerged as interesting semiconductors for a broad spectrum of applications, such as solar cells, lasers, light-emitting diodes, and photodetectors. This is attributed to their outstanding characteristics, including facile synthesis, strong light absorption capabilities, tunable bandgaps, and exceptional charge carrier mobility. However, despite their immense potential, these materials are constrained by challenges such as carrier recombination and nonradiative energy losses, which significantly impede their overall efficiency. The basic aim of this thesis is to enhance the efficiency of charge-carrier transfer to the transport layers by mitigating both radiative and nonradiative recombination losses. I have done this by synthesizing defect-passivated perovskite nanocrystals (PNCs), increasing carrier diffusion in PNC assembly, and quickly extracting the carriers using multiwall carbon nanotube (MWCNT) as an acceptor molecule. The thesis is structured into five chapters. Chapter 1 provides an introduction to perovskites, focusing on their structures, photoluminescence (PL) behavior, and carrier diffusion dynamics across different perovskite sample types. Special emphasis is placed on the use of PNC films of optimal thickness to maximize carrier diffusion. This chapter also explores various carrier extraction strategies and potential applications of perovskites. Chapter 2 details the synthesis methodologies for PNCs and perovskite quantum dots (PQDs) using the hot-injection method. The functionalization process of MWCNT is also detailed here. It also includes microscopic and spectroscopic tools, viz., steady-state absorption spectroscopy, steady-state fluorescence spectroscopy, time-resolved single-photon counting, transient absorption spectroscopy, Raman spectroscopy, X-ray photoelectron spectroscopy, Fourier-transform infrared spectroscopy, scanning transmission electron microscopy, and single-particle microscopy. Chapter 3 focuses on addressing nonradiative recombination, which is commonly associated with defects in PNCs. $CsPbBr_3$ PNCs synthesized via hot injection come with defects that reduce PL quantum yield (PLQY) and PL lifetime. Among various halide salts tested for defect passivation, NaBr was identified as the most effective, increasing the PLQY from 58% to 98%. Single-particle blinking studies were conducted for rationalizing the defects and optimizing the PL properties. Defect passivation significantly reduced nonradiative recombination, improving the photophysical properties of the PNCs. Anyway, to increase the carrier diffusion, PNC assembly formation is necessary. In Chapter 4, I demonstrated the polymer-induced assembly formation which increases the carrier lifetime and carrier diffusion. Additionally, temperature-controlled carrier diffusion was also demonstrated, highlighting the potential of these assemblies for

external stimuli-responsive optoelectronic devices. Although increased carrier diffusion helps the carrier to reach the charge transport layer, delayed carrier transport from perovskite to charge transport layers increases the likelihood of radiative recombination. In Chapter 5, I focus on the quick extraction of carriers from PNCs using functionalized MWCNT. The PNCs bind to MWCNTs, forming PNC-MWCNT conjugates. These conjugates exhibit hot-carrier transfer from PNCs to MWCNTs, reduced nonradiative Auger recombination, and minimized radiative recombination at the band edge. Overall, this thesis not only helps to clarify the charge-carrier migration in lead halide perovskites but also demonstrates their harvesting by interfacial coupling with MWCNT, presenting strategies for improving charge transport and extraction beneficial for developing highly efficient and durable optoelectronic devices.