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学 位 論 文 審 査 の 要 旨

博士 (環境科学)

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学 位 論 文 題 名

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)

The candidate focused her doctoral research on optimizing the properties and charge carrier dynamics in lead halide perovskite (LHP) quantum dots and nanocrystals, one of the most promising classes of semiconductor materials for future laser, light-harvesting, sensor, and lighting applications, by halide vacancy filling, assembly preparation, chemical conjugation, and spectroscopic and microscopic studies. Several challenges, including low stability, carrier diffusion limitations, and inefficient carrier extraction are addressed in the thesis to improve the quality of LHPs and their hybrid systems. The candidate's research aimed to enhance the stability of nanocrystals via defect passivation, improve carrier diffusion through assembly formation, and optimize carrier extraction by integrating multiwall carbon nanotubes (MWCNTs) as electron acceptors. The thesis is structured into five chapters, covering synthesis methodologies, optical characterizations, and device performance enhancements.

Chapter 1 introduces perovskite structures, their photoluminescence (PL) behavior, and carrier diffusion mechanisms. Special attention is given to perovskite nanocrystal (PNC) films, emphasizing their optimal thickness for maximizing carrier transport. This chapter also discusses carrier extraction strategies and practical applications. Chapter 2 describes the synthesis methodologies for PNCs and perovskite quantum dots (PQDs) using the hot-injection method. It details the functionalization of MWCNTs, which were utilized to improve charge extraction efficiency. The experimental section also includes microscopic and spectroscopic tools, such as optical microscopy, scanning transmission electron microscopy (STEM), steady-state absorption spectroscopy, steady-state PL spectroscopy, time-resolved (TR) PL spectroscopy, transient absorption (TA) spectroscopy, Energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy (XPS). Chapter 3 focuses on defect passivation in CsPbBr₃ PNCs, primarily affecting nonradiative recombination and material instability. The candidate performed STEM-EDX analysis, confirming the presence of bromide vacancies leading to reduced PL quantum yield (QY) and PL lifetime. The PNCs were post-treated with various inorganic halide sources, among

which NaBr proved to be the most effective, increasing PLQY from 58% to 98% and PL lifetime from 11.6 ns to 13.8 ns. Chapter 4 concentrates on polymer-induced assembly formation of FAPbBr₃ PQDs, demonstrating solvent evaporation and polymer shrinkage-driven self-assembly. The study highlights redshifted PL spectra, extended PL lifetimes, and temperature-controlled carrier diffusion, illustrating the potential of such assemblies in stimuli-responsive optoelectronic devices. Chapter 5 focuses on fast charge carrier extraction from CsPbBr₃ PNCs using MWCNTs as electron acceptors. Functionalized MWCNTs were integrated into the PNC system via post-mixing and in situ hot-injection conjugation techniques. The conjugates exhibited hot-carrier transfer, biexciton-mediated nonradiative Auger recombination suppression, and enhanced charge transport efficiency, as revealed by TRPL and TA studies. The candidate's research significantly improves the quality of perovskite optoelectronic materials by addressing stability, carrier diffusion, and charge extraction challenges. The findings pave the way for enhanced efficiency for high-performance, durable perovskite-based devices.

審査員一同は、これらの成果を高く評価し、また研究者として誠実かつ熱心であり、大学院博士課程における研鑽や修得単位などもあわせ、申請者が博士（環境科学）の学位を受けるのに十分な資格を有するものと判定した。