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学 位 論 文 審 査 の 要 旨

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学 位 論 文 題 名

Investigation of photoinduced electron transfer from CdSe-CdS and FAPbBr₃ nanocrystal assemblies to molecular acceptors

(CdSe-CdSおよびFAPbBr₃ナノ結晶集合体から分子受容体への光誘起電子移動の研究)

The candidate conducted her doctoral research on photoinduced electron transfer processes from solutions, assemblies, and films of lead halide perovskite (LHP) nanocrystals (NCs) and cadmium chalcogenide quantum dots (QDs) to molecular acceptors. These luminescent NCs and QDs exhibit exceptional optical and electronic properties, making them highly promising for future optoelectronic applications such as solar cells, light-emitting devices, lasers, optical displays, and photodetectors. The research in this thesis aims to elucidate the fundamental mechanisms governing charge carrier transport and improve electron transfer efficiency through structural optimization and controlled assembly formation of NCs and QDs. The study addressed challenges related to charge transport limitations in isolated nanocrystals, demonstrating that NC and QD assemblies can enhance carrier diffusion and electron transfer efficiency.

The thesis is structured into five chapters. Chapter 1 introduces the fundamental properties of perovskite nanomaterials and chalcogenide QDs, focusing on the quantum confinement effect and its impact on absorption and emission properties. It also gives an overview of photoinduced electron transfer. Various synthesis methods for perovskites and chalcogenide NCs and QDs are discussed. The chapter highlights the significance of film preparation and assembly formation in enhancing charge transport and concludes with the research objectives and practical applications of these materials in optoelectronics. Chapter 2 details the preparation methodologies for formamidinium lead bromide (FAPbBr₃) LHP NCs, cadmium selenide zinc sulfide (CdSe-CdS) QDs, and mesoscopic assemblies and thin films of these NCs and QDs. Also, the candidate included instrumental techniques employed in characterizing the above samples and their optical and electron transfer properties, including the theoretical principles and operational methodologies of spectroscopic tools such as steady-state fluorescence spectroscopy, UV-visible absorption spectroscopy, differential light-scattering (DLS) technique, scanning transmission electron microscopy (STEM), and time-resolved photoluminescence (TRPL) spectroscopy. Chapter

3 explores the electron transfer dynamics from perovskite nanocrystals and their assemblies to specific molecular acceptors. Electron acceptors such as 7,8,8-tetracyanoquinodimethane (TCNQ), fullerene (C60), and functionalized fullerene (C60-RCOOH) were selected based on their high electron affinity. While isolated nanocrystals exhibited limited charge transfer due to ligand-induced restrictions, assemblies and thin films significantly enhanced electron transfer efficiency. Investigations conducted in solution and film states confirmed that assembly structures facilitated charge carrier diffusion, improving overall performance. Chapter 4 extends the findings of Chapter 3 by examining the electron transfer properties of CdSe-CdS QD assemblies. Core-shell QDs were chosen for their exceptional optical properties and suitability for device applications. Controlled assembly formation was achieved using 1,6-hexane dithiol as a cross-linker, enabling precise analysis of photoinduced electron transfer processes. The study revealed that structural organization is crucial in optimizing charge transport efficiency. Chapter 5 introduces a bio-inspired approach to generating water-soluble mesoscopic CdSe-CdS assemblies. This novel technique utilized functionalized CdSe-CdS QDs modified with carboxyl (-COOH) groups and further aggregated using an amide condensation reaction of lysine. Trypsin-mediated cleavage of the amide bonds facilitated controlled disassembly, leading to well-dispersed mesoscopic QD assemblies. Characterization via STEM and DLS confirmed the successful formation of these assemblies, highlighting their potential for biocompatible optoelectronic applications. The candidate's research significantly advances the understanding of electron transfer processes in perovskite NC and chalcogenide QD films and assemblies. Demonstrating the impact of controlled structural organization on charge carrier dynamics offers valuable insights into optimizing these materials for next-generation photovoltaic and optoelectronic devices. The research contributes to the fundamental design principles of nanocrystal assemblies, paving the way for enhanced device efficiency and broader applications in advanced energy materials.

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。