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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 exciton-plasmon coupling in luminescent semiconductor quantum dot-gold nanoparticle systems
（発光半導体量子ドット-金ナノ粒子系における励起子-プラズモン結合の研究）

The candidate's doctoral research focused on enhancing the photoluminescence (PL) properties of two promising classes of semiconductor quantum dots (QDs), namely methylammonium lead iodide ($MAPbI_3$) perovskite QDs and cadmium selenide–zinc sulfide (CdSe–ZnS) QDs, by coupling their excitonic states with the localized surface plasmon resonance (LSPR) of gold nanorods (Au NRs). These QDs, widely studied for applications in lasers, light harvesting, sensing, and solid-state lighting, typically suffer from PL quenching due to nonradiative exciton recombination caused by carrier trapping in charged states. This results in stochastic blinking and low average emission intensity, limiting their practical utility. The core objective of the candidate's doctoral research was to improve the emission intensity of individual QDs by inducing exciton–plasmon coupling through direct contact with Au NRs or integration within a plasmonic cavity. The thesis is organized into five chapters, each systematically advancing from fundamental principles to experimental realizations, analysis, and exciton-plasmon coupling mechanisms.
Chapter 1 provides a comprehensive introduction to the fundamental concepts of excitons, plasmons, and their coupling mechanisms. It presents background knowledge on the excitonic behavior of QDs, the optical characteristics of gold nanostructures, plasmonic cavities, and key examples of exciton–plasmon coupling in both organic dye–metal and semiconductor QD–metal systems. Chapter 2 details the experimental methodology and materials used. This includes the synthesis of $MAPbI_3$ QDs with and without surface ligands, achieved through ligand-assisted reprecipitation and precursor spraying methods, respectively. It also describes the preparation of Au NRs and the fabrication of QD–plasmonic hybrid systems. Characterization tools included optical microscopy, scanning transmission electron microscopy, steady-state and time-resolved PL spectroscopy, transient absorption spectroscopy, and single-particle PL measurements. Chapter 3 explores the optical behavior of perovskite QDs deposited on glass and Au NRs, investigated at ensemble and single-particle levels. The integration of QDs with Au NRs

led to a significant enhancement in PL intensity, attributed to plasmon-assisted biexciton generation and accelerated radiative recombination. These results demonstrate the effectiveness of LSPR coupling in amplifying the PL of halide perovskite and metal chalcogenide QDs. Chapter 4 examines the exciton–plasmon coupling in CdSe–ZnS QD–Au NR systems. When the plasmon resonance of Au NRs was matched with the QD exciton energy, a notable suppression of PL blinking and an increase in PL intensity were observed. These effects were linked to the quenching of nonradiative processes and improved electron–hole recombination efficiency due to near-field plasmonic interactions. Chapter 5 investigates the formation of a low-energy hybrid exciton–plasmon state within a plasmonic cavity composed of a gold mirror, gold nanorods, and embedded QDs within polymer layers between the mirror and rods. The hybrid state showed further PL intensity enhancement and extended exciton lifetimes, resulting from energy transfer processes between the cavity and QDs. In conclusion, the candidate's research provides significant advancements in the understanding and application of exciton–plasmon coupling to enhance QD photoluminescence. By demonstrating substantial emission enhancement and blinking suppression in both halide perovskite and CdSe–ZnS QDs, this work contributes valuable insights toward the development of next-generation optoelectronic devices with improved performance and reliability.

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充分な資格を有するものと判定した。