学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Synthesis of Highly Luminescent Cesium Tin Halide Perovskite Nanocrystals
（高発光効率セシウムスズハライドペロブスカイトナノ粒子の合成）

Colloidal halide perovskite nanocrystals (NCs) have garnered substantial attention in recent years due to their excellent optoelectronic properties, which encompass tunable bandgaps, high photoluminescence efficiency, and substantial absorption coefficients. These attributes have propelled their widespread utilization across diverse optoelectronic applications, including lasers, light-emitting diodes, and solar cells. However, amidst the burgeoning interest in harnessing metal halide perovskite for optoelectronic devices, concerns over lead toxicity in lead-based halide perovskite materials necessitate the exploration of lead-free alternatives. Tin halide perovskites have emerged as promising substitutes owing to their reduced toxicity and comparable perovskite-like characteristics. Nonetheless, the synthesis of high-quality colloidal $CsSnX_3$ (X= Cl, Br, I) perovskite NCs presents a formidable challenge, impeding their widespread adoption in next-generation optoelectronic applications. This thesis delves into the synthesis mechanism and precursor chemistry governing the formation of tin halide perovskite NCs, culminating in the development of effective synthetic methodologies conducive to yielding high-quality products.

Chapter 1 provides an introduction to the research background of metal halide perovskite materials, including synthetic methods for metal halide perovskites, and summarizes the effective strategies reported to enhance the optical properties of metal halide perovskite materials. Additionally, it outlines the research progress of tin-based halide perovskite NCs. Finally, it elucidates the research topic's conceptual framework.

In chapter 2, the investigation focuses on elucidating the precursor chemistry involved in the hot-injection synthesis method for representative tin halide perovskite NCs, particularly $CsSnI_3$ NCs. Through comprehensive analysis, including nuclear magnetic resonance (NMR) spectroscopy, significant insights are gleaned regarding the role of polymeric alkanoate iodides, formed as intermediate products during the reaction between tin and iodide precursors, in modulating NCs properties such as photoluminescence quantum yield (PLQY), size, morphology, and uniformity.

In chapter 3, a straightforward method based on ion exchange is proposed, showcasing its effectiveness in generating high-quality $CsSnX_3$ NCs with excellent PLQY and adjustable size. $CsSnI_3$ NCs with a remarkable PLQY of 34.4% were obtained, surpassing the previously reported maximum value of 18.4%. The controlled reaction kinetics inherent in this approach enable the synthesis of NCs with excellent size uniformity, crucial for optoelectronic applications.

In chapter 4, further exploration into controlled ion exchange processes, facilitated by incorporating an additional tin source during the synthesis of $CsSnI_3$ NCs, unveils a pathway to enhance crystal quality. This endeavor resulted in a significant enhancement of the PLQY of $CsSnI_3$ NCs to 49.7%, accompanied by uniform size distribution. In-depth analysis, including in-situ photoluminescence (PL) and high-resolution transmission electron microscopy (HRTEM), elucidates the role of the additional tin source in accelerating the reaction rate that minimizes uncoordinated Sn and I atoms and thus boosts the PLQY of $CsSnI_3$ NCs. Additionally, the versatility of this methods is demonstrated through the synthesis of tin halide perovskite NCs with varied halide compositions.

In summary, this thesis advances the understanding of synthesis strategies for high-quality

tin halide perovskite NCs, offering valuable insights into precursor chemistry, reaction mechanisms, and synthetic methodologies. The developed approaches hold immense potential for facilitating the widespread utilization of tin halide perovskite NCs in diverse optoelectronic applications, thereby propelling the evolution of next-generation optoelectronic devices.