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種別(和文)	要約
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Thesis title:

Microscopic Fluorescence Study of Ion Migration in Lead Halide Perovskite Heterostructures

Thesis outline:

In this thesis, we systematically study the migration behavior of halide anions and A-site cations in lead halide perovskites heterostructures using fluorescence microscopy.

Chapter 1 provides a comprehensive background on lead halide perovskites, highlighting the fundamental issues related to ion migration, and the innovative approaches involving perovskite heterostructures to mitigate these challenges. It also introduces the importance of microscopic fluorescence studies in unveiling the intricate mechanisms of ion migration at the micro-scale.

Chapter 2 details the chemicals, experimental instrumentation, and methodologies employed in this research, laying a solid foundation for the subsequent experimental analyses and characterizations.

In **Chapter 3**, we investigate the effect of the local halide environment on halide anion migration in $CsPbX_3$@MOF(X) composites. The metal-organic framework MOF(X) serves as a lead source for the in-situ formation of $CsPbX_3$ and provides a controlled halide environment for ion modulation. By analyzing the crystallization kinetics of $CsPbX_3$@MOF(X) during the initial stages and its spectral dynamics after complete reaction, we reveal a large compositional heterogeneity of the embedded mixed halogen perovskites and their non-monotonic photoluminescence spectral evolutions under illumination. This reflects the dynamic modulation of ion migration induced by changes

in the local environment.

In **Chapter 4**, we explore the migration of cations in vertical $CsPbBr_3/PEA_2PbBr_4$ nano-heterostructures prepared through post-growth self-assembly. Our investigations reveal a layer-by-layer cation migration and the resulting sequential restructuring at the 3D/2D epitaxial interface under continuous illumination. This cation migration is reversible, similar to the behavior observed for halide anions, reflecting the local environment-induced dynamic nature of ion migration. The structural dynamics at the 3D/2D interface ultimately leads to optimized distribution quasi-2D phases and efficient cascade-like energy transfer from the 2D to the 3D phase.

In **Chapter 5**, we investigate the cation exchange behavior of MA (methylamine) and FA (formamidine) within pure 2D PEA_2PbBr_4 nanoplates. Our findings reveal that both MA and FA exhibit higher in-plane migration compared to out-of-plane migration. After rapid in-plane exchange, ion migration is suppressed due to increased lattice mismatch at the 3D/2D lateral interface. This spatially heterogeneous migration and interface ion management lead to the formation of 3D/2D lateral heterostructures of $MAPbBr_3/PEA_2PbBr_4$ and $FAPbBr_3/PEA_2PbBr_4$. Among these, $FAPbBr_3/PEA_2PbBr_4$ shows high spectral purity and superior spectral stability due to the higher spatial resistance of the relatively larger FA cations compared to MA cations.

Finally, **Chapter 6** concludes the work done in this thesis. We highlight the critical importance of interface design and ion selection in structure optimizing, enhancing energy flow, and improving spectral purity and stability. These findings advance our understanding of ion migration mechanisms and will provide valuable feedback for the further development of more stable and efficient perovskite-based devices.