# Studies on the Efficiency and Durability Control of Quasi-2D Perovskite Light-Emitting Diodes and Related Mechanisms

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論 文 名　: Studies on the Efficiency and Durability Control of Quasi-2D Perovskite Light-Emitting Diodes and Related Mechanisms
（擬二次元ペロブスカイト LED の効率と耐久性及びメカニズム解析に関する研究）

区　　分　: 甲

# 論 文 内 容 の 要 旨

Quasi-2D PeLEDs have earned much attention from researchers because of their rapidly improving EQEs. However, how the structural properties (multi-*n* configuration, domain passivation effect, and etc.) of quasi-2D perovskite films affect the emission properties remains controversial. Besides, quasi-2D PeLEDs reported by far still had short working lifetimes and severe EQE roll-off, for which the mechanisms need investigation.

The work presented in the thesis reports on the essential factors impacting the emission efficiencies and working lifetimes of quasi-2D PeLEDs and the mechanisms of how these factors work.

In **Chapter 1**, the definition of metal halide perovskites and their structural and optoelectronic properties were introduced. The background of constructing quasi-2D perovskite in PeLEDs was illustrated. The development and remaining issues of quasi-2D PeLEDs were discussed.

In **Chapter 2**, how the stoichiometric engineering impacts the defect passivation and domain distribution of quasi-2D perovskites was systematically investigated to understand the influence to the emission efficiencies. By tuning the ratio of perovskite precursors, not only the *n* of the emission domains was changed, but also the non-emission low-*n* domains were controlled. These low-*n* domains were further proved unfavorable for obtaining efficient emission of quasi-2D perovskites.

As a result, in perovskite films prepared from stoichiometric quasi-2D precursor compositions, large organic ammonium cations function well as passivators. In comparison, compositions of simply adding a large organic halide salt into the 3D perovskite precursor ensure not only the defect passivation but also the effective formation of quasi-2D perovskite domains, with avoiding unfavorable low-*n* domains appearing, ending up with remarkably high exciton binding energies. Quasi-2D perovskite films fabricated with a well-designed precursor composition achieved a high PLQY of 95.3% and an EQE of 14.7% in LEDs

In **Chapter 3**, the mechanisms of the degradation and EQE roll-off on the basis of ion migration were discussed. From the trends of EQE and voltage evolutions, two device decay stages with different mechanisms were found. The recovery behaviors of the quasi-2D PeLEDs indicate that some ion migration dominates the EQE decay. The migrated ions were then discovered to be ammonium cations attached on the domain boundaries, and their migration was also responsible for the severe EQE roll-off.

As a result, it was concluded that the migration of ligand cations through domain boundaries of quasi-2D perovskite films induces the gradual losing of defect passivation at the boundaries, which results in the reversible PeLED degradation and severe EQE roll-off. When the device operation time is long, the mobile cations enter and interact with the electron transport layer, leading to the stage of irreversible PeLED degradation. The device degradation mechanisms discovered here are constructive for developing quasi-2D PeLEDs with smaller efficiency roll-off and better operational durability.

In **Chapter 4**, based on the discoveries introduced above, the perspective regarding the strategies on achieving shorter emission wavelengths, higher efficiency, and better durability for quasi-2D PeLEDs were discussed.