Mass Transfer and Stability Properties of Functinalized 2D Perovskite Interfaces

Two-dimensional (2D) halide perovskites are an attractive class of hybrid perovskites that have additional optoelectronic tunability due to their accommodation of relatively large organic ligands. Nevertheless, contemporary ligand design depends on either expensive trial-and-error testing of whether a ligand can be integrated within the lattice or conservative heuristics that unduly limit the scope of ligand chemistries.

This work begins by investigating simulation-guided ligand design. Employing molecular dynamics (MD) simulations and machine learning (ML) models, systematic ligand exploration unveils the principles governing the stability and structural relationships of these perovskites. Promising ligand candidates undergo a refinement process informed by feasibility considerations, followed by experimental synthesis and characterization to underscore the effectiveness of simulation-informed design strategies.

Delving into the realm of anionic diffusion, a pivotal factor in 2D perovskite stability, the study examines this property using both experimental methods and simulation techniques. This parallel examination underscores the alignment between simulation predictions and real-world observations, offering nuanced insights derived from molecular simulations. Importantly, simulations serve as potent tools for hypothesis validation when ligands originate from experimental synthesis, affirming conjectures stemming from empirical insights.

The exploration extends to comprehending the molecular insights of experimental observations, thereby shedding light on factors that enhance device efficiency. We explore mechanisms for mitigating phase disproportionation, optimizing ion diffusion, modulating molecular interactions between perovskite and polymeric hole-transporting materials, and uncovering the single-molecule behavior that leads to high photoluminescence quantum yields. Notably, all simulation outcomes align with experimental findings, further validating the utility of MD analysis in the context of 2D perovskite systems.

Furthermore, this work addresses a crucial aspect of MD simulations, namely the refinement of force field models. Previously developed topology automated fixed-charge force-field interactions (TAFFI) is augmented through the incorporation of polarizability using classical Drude oscillators, resulting in a novel framework termed TAFFI-Drude. This approach enhances electrostatic properties while retaining transferability and consistency from the existing TAFFI model.

The thesis concludes with a comprehensive discussion of the findings across the aforementioned areas, highlighting the impact of simulation-driven design and insights in advancing 2D perovskite research. The implications of these discoveries for optoelectronic applications and the broader field of materials science are explored, emphasizing the potential for innovation and improvement within the realm of 2D halide perovskites.