DEVELOPMENT OF THE PEROVSKITE-POLYMER INKS AND UNDERSTANDING THEIR STRUCTURE-PROPERTY RELATIONSHIP FOR OPTOELECTRONIC APPLICATIONS

ABSTRACT

The functionality of perovskite-based opto-electronic devices, including solar cells, LEDs, transistors, and sensors, is significantly influenced by the fabrication of the perovskite active layer, its integration into suitable device architectures, and the optimization of device performance through various component adjustments. Understanding the structure-property correlation is essential for the development of this technology.

This thesis investigates the effects of macroscale material fabrication modifications on microscale charge carrier dynamics. By employing the '*polymer-in-perovskite*' approach, micro grains larger than 100μm were achieved for the PMMA-incorporated triple cation perovskite precursor, resulting in a compact film morphology without anti-solvent treatment. Detailed investigations of crystallization dynamics led to an enhancement in radiative recombination lifetime from 0.88 μs (for pristine FAMACs film) to 1.45 μs (for FAMACs film with PMMA incorporation). Additionally, microstructural findings of the photoactive layer were appraised against operating conditions in a device through transient photocurrent measurements, aiming to gain a deeper understanding of the '*polymer-in-perovskite*' approach for precursor development.

Furthermore, a methodology utilizing ligands and surfactants to engineer colloidal suspensions of perovskite was explored. This foundational approach, using methylammonium lead triiodide (MAPI) and polymethyl methacrylate (PMMA), offers an accessible alternative to post-

treatment of the photoactive layer. By integrating the properties of polymers and perovskites with selective additives, a novel '*perovskite-in-polymer matrix*' solution was developed for one-step coating of colloidal perovskite, enabling the production of distinct and tailored morphologies. This solvent system, featuring various additives, serves as a template to achieve similar morphologies, provided the additives contain related functional groups. These inks can be adjusted to produce different surface morphologies, governed by the control of colloidal composition and molecular arrangement dynamics, which are explored in detail in this dissertation. The study also investigates phase segregation observed in quasi-2D-3D perovskite polycrystalline films formed using these template inks.

Overall, this thesis delves into colloidal chemistry, molecular arrangement, film morphology, and the properties of particle structure, size, and shape, as well as their optoelectronic characteristics. Comprehensive analyses of these aspects are provided through a range of measurements, including X-ray diffraction (XRD), scanning electron microscopy (SEM), steady-state photoluminescence (PL), UV-vis absorbance, confocal laser scanning microscopy (CLSM), and small-angle X-ray scattering (SAXS), etc.