

## Abstract

Perovskite solar cells manifested an outstanding evolution in efficiency beyond 25%. Their high defect tolerance and compositional flexibility allow solution processing and tunability of optoelectronic properties, stability, efficiency, and even chemical safety through the facile change of both cations and anions. There are a plethora of device fabrication protocols and thus deviations are evident in the reported properties and performance metrics which pose questions regarding the mechanisms behind the liquid-to-solid transition and especially the solution dynamics in the perovskite precursor inks. To address this, spectroscopic methods such as solution nuclear magnetic resonance, ultraviolet-visible absorption, Fourier-transform infrared and photoluminescence spectroscopy were used in this work to investigate the nature of perovskite precursor inks. The found dynamic solution equilibria are influenced by the choice of solvents, additives, and overall composition with structures in dimethyl sulfoxide (DMSO) and dimethyl formamide (DMF) resembling solid-state ones. The main goal of this thesis was to minimize the toxicity and safety risks of perovskite precursor inks and solar cells. Therefore, two holistic approaches were developed to eliminate the necessity of toxic solvents in  $\text{MAPbBr}_3$  and  $\text{MAPbI}_3$  inks by using the protic ionic liquid methylammonium propionate in combination with water and alcohol. The solution chemistry and liquid-to-solid transition of these greener inks were investigated through spectroscopic techniques such as in-situ PL spectroscopy manifesting different solution species and liquid-to-solid transition mechanisms compared to the DMF- and DMSO-based precursor inks. Secondly, lead was replaced by less toxic bismuth to design new perovskite-inspired materials, which are thoroughly studied towards optoelectronic properties, solar cell performance and stability to learn new strategies of optimizing and designing greener absorber materials.