Materials Theory of Halide Perovskites: Insights from Atomistic and Molecular Modeling

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Metal halide perovskites, known for their unique chemical and physical properties, have attracted considerable attention over the past decade for their potential in optoelectronic applications such as solar cells, LEDs, and photodetectors. Our research approach leverages a sophisticated blend of computational methods, including electronic structure calculations (DFT and tight binding) and reactive molecular dynamics simulations with ReaxFF and machine learning potentials. This comprehensive, multiscale approach enables us to thoroughly investigate the electronic and dynamical properties of halide perovskites, leading to significant insights and advancements.

One of our research focuses is on the analysis of defects in halide perovskites. Understanding and mitigating these defects is crucial for enhancing the efficiency and longevity of optoelectronic devices. Our approach includes the detailed determination of electronic energy levels and thorough examination of the dynamic properties of these materials. By identifying defects responsible for recombination losses and chemical degradation, we develop targeted strategies for their mitigation. These strategies involve engineering perovskite compositions and interfaces, using passivation agents, and optimizing the quality of perovskite films through precise control of synthetic chemistry and processing parameters.

Another key area of our research is exploring chirality in perovskites. Introducing chiral organic ligands into perovskite structures alters their mirror symmetry, leading to unique properties like chiral-induced spin selectivity (CISS) and chiroptical activity, both exhibiting temperature-dependent behaviors. We use first-principles methods and models for calculating circular dichroism (CD) and analyzing electron/spin transport. We study the impact of the dynamic microstructure of chiral perovskites under various temperature conditions, aiming to identify structural features that influence the optoelectronic responses. These insights provide valuable information for the design of new chiral perovskites with potential applications in novel optoelectronics, such as spin LEDs and chiral photodetectors.

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