Computational Insights into Organic/Inorganic Halide Perovskite Solar Devices

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Hybrid organic/inorganic halide perovskite solar cells (PSCs) have become a promising class of materials for solar cells. Researchers have recently developed numerous methods to reduce defects, suppress non-radiative recombination, and enhance device efficiency and stability. Among these, additive and interface engineering methods have attracted extensive attention and are particularly popular for high-efficiency and stable PSCs. However, fundamental knowledge of the underlying atomic-level processes leading to improved performances and lifespan is often lacking. We employ DFT calculations, classical and ab-initio molecular dynamics (MD) simulations, with the help of state-of-the-art, polarizable classical force-fields and machine-learned interatomic potentials, trained on ab-initio MD trajectories, to unveil the underlying mechanisms connected to the facilitated interfacial charge transfer and improved stability of various PSCs devices. In particular, we apply our methodology to assess the stabilizing nature, as well as the optoelectronic properties of recently proposed hydrophobic organic cations in the context of layered two-dimensional perovskite materials and inverted PSCs.

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