MULTI-SCALE MODELING OF HYBRID PEROVSKITE PHOTOVOLTAICS

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Hybrid organic-inorganic perovskites (HPs) are a novel materials class in photovoltaic (PV) research. Their light-to-energy conversion efficiency has progressed extremely rapidly (Fig. 1), but further advancements require atomistic insight into materials processes that is difficult to obtain due to the complex structure of HPs. We address this structural complexity by developing a multi-scale model that combines quantum mechanical calculations with long-range electrostatic interactions.

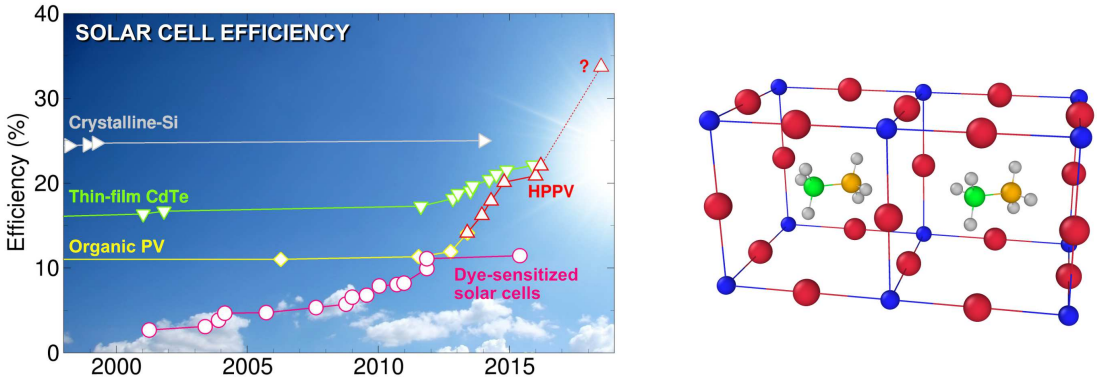

Figure 1: Left: Conversion efficiency evolution with time of different solar cell types. Right: An example of an MA^+ pair configuration in neighboring $MAPbI_3$ unit cells.

Our multi-scale model parameterizes the interaction between neighboring methylammonium cations (MA^+) in methylammonium lead triiodide ($CH_3NH_3PbI_3$, or $MAPbI_3$). It is based on our previous quantum mechanical study [1], in which we observed strong coupling between the MA^+ cations and the inorganic PbI_3^- lattice via hydrogen bonding and identified two preferred MA^+ orientations in the unit cell. The resulting 196 MA^+ pair combinations can be reduced to only 25 due to symmetry. We then perform density-functional theory (DFT) calculations for $3\times3\times3$ and $4\times4\times4$ supercell models to analyse the distribution of these 25 pairs and to determine their relative energy contributions.

We apply our pair model to larger supercells to develop a statistical description of the $MAPbI_3$ structure on the nanometer scale. We observe a large percentage of parallelly-aligned MA^+ configurations (Fig. 1) that arrange in linear domains. The length distribution of these linear domains suggests that they may be favorable to charge carrier transport through the material, which would benefit the efficiency of HPs.

[1] J. Li and P. Rinke, Phys. Rev. B **94** 045201 (2016).