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Modeling organohalide perovskites for photovoltaic applications: From materials to interfaces¹

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The field of hybrid/organic photovoltaics has been revolutionized in 2012 by the first reports of solid-state solar cells based on organohalide perovskites, now topping at 20% efficiency. First-principles modeling has been widely applied to the dye-sensitized solar cells field, and more recently to perovskite-based solar cells. The computational design and screening of new materials has played a major role in advancing the DSCs field. Suitable modeling strategies may also offer a view of the crucial heterointerfaces ruling the device operational mechanism. I will illustrate how simulation tools can be employed in the emerging field of perovskite solar cells. The performance of the proposed simulation toolbox along with the fundamental modeling strategies are presented using selected examples of relevant materials and interfaces. The main issue with hybrid perovskite modeling is to be able to accurately describe their structural, electronic and optical features. These materials show a degree of short range disorder, due to the presence of mobile organic cations embedded within the inorganic matrix, requiring to average their properties over a molecular dynamics trajectory. Due to the presence of heavy atoms (e.g. Sn and Pb) their electronic structure must take into account spin-orbit coupling (SOC) in an effective way, possibly including GW corrections. The proposed SOC-GW method constitutes the basis for tuning the materials electronic and optical properties, rationalizing experimental trends. Modeling charge generation in perovskite-sensitized TiO₂ interfaces is then approached based on a SOC-DFT scheme, describing alignment of energy levels in a qualitatively correct fashion. The role of interfacial chemistry on the device performance is finally discussed.

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