Minicolloque n° MMPS28 × **Oral** ☐ **Poster**

Modeling the photophysical properties of Perovskite Solar Cells interfaces using Density Functional Theory: the case of MAPI/CBA/TiO_2

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With various advantages over other types of solar cells, such as easy fabrication and flexibility, hybrid organic-inorganic perovskites solar cells (PSCs) are considered as potential candidates for next generation photovoltaic devices [1]. They feature an absorber layer (usually a metal-halide perovskite) sandwiched between electron (typically TiO_2) and hole (i.e. Spiro-OMeTAD) transport layers. Although advances in synthesis techniques [2;3], compositional engineering [4;5] and device structure optimization [6] has led to the production of high-quality polycrystalline perovskite films rapidly boosting in a few years their performances, they however present high density levels of defects at the interfaces between the absorber layer and the charge transport substrates which act as non-radiative recombination sites for holes and electrons thereby affecting the electron transport properties of PSCs. Interface defects thus constitute one of the main causes for losses in PCE and interface engineering has become of crucial importance to address this issue. Interfacial tuning can be attained by employing a self-assembled monolayer (SAM) of bifunctional organic molecules linking the perovskite to the substrate and stabilizing their interface [7]. However, finding the best SAM candidates for interface modification is not an easy task due to lack of experimental technique able to characterize the full interface.

In this context, the ab-initio structural and electronic modeling of PSC interfaces can give interesting insights for their understanding and in silico optimization. Recently, a self-consistent electrostatic embedding approach (SC-Ewald) coupled to time-dependent density functional theory (TD-DFT) calculations has been developed by some of us [8;9] in order to model the ground and excited state properties of periodic materials such as interfaces, allowing to compute their photophysical properties at low computational cost.

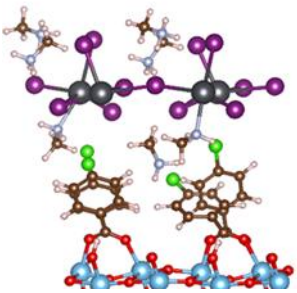

Figure 1: MAPI/CBA/TiO_2 interface model.

Here this computational strategy is applied to simulate the UV-Vis absorption spectrum of a model MAPI/CBA/TiO_2 interface, where the CBA (p-chlorobenzoic acid) molecules form the SAM connecting the MAPI and TiO_2 surfaces [7]. The results obtained allow to infer the role played by the SAM and the analysis of the excited states mostly involved in the absorption shows an electron density delocalization between the MAPI and the TiO_2 thereby suggesting efficient electron injection at the interface.

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