

DFT Simulations and Device Modelling of MXene/Perovskite Solar Cells

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Perovskite solar cells (PSC) have gained a key role in the field of renewable energy due to their high efficiencies and low production costs [1]. However, the effort in tuning the perovskite crystal morphology and interface properties, in order to improve the device performances, is still huge [2]. Among the solutions proposed so far, MXenes have emerged for the possibility of being incorporated within the perovskite layer resulting in an important improvement of the PSC efficiency [3].

In this study, we investigate the interaction between MXenes and perovskite, performing first-principles calculations in the framework of density functional theory. Here, we consider the interface between a $\text{CH}_3\text{NH}_3\text{PbI}_3$ thick layer, in its high temperature cubic phase, and a $\text{Ti}_3\text{C}_2\text{T}_x$ MXene layer, with the T_x functional group being F, O or/and OH. In particular, we analyze the electrostatic potential profile in the direction orthogonal to the interface and the electronic band structure of the system. We find that the functionalization of the MXene surface plays a crucial role in determining the electronic properties of the MXene/perovskite system. Specifically, the perovskite work function is strongly affected by the presence of the MXene layer and a shift of the Fermi level with respect to the perovskite band edge is observed. Interestingly, these effects depend on the MXene surface functional group: on the one hand, the OH group causes a reduction of the work function and an upward shift of the Fermi level, on the other hand, the O terminal group has the opposite effect. Parameters extracted from DFT have been used to model the PSC behavior using a Drift-Diffusion model. Comparison between simulations and experiments permits to outline the important role of the 2D MXenes in improving the performance of PSC through work function tuning.

[1] <https://www.nrel.gov/pv/assets/pdfs/pv-efficiency-chart.20190103>

[2] A. Agresti et al., ACS Energy Lett., **2**, 279 (2017).

[3] Z. Guo et al., Small, **14.47**, 1802738 (2018).

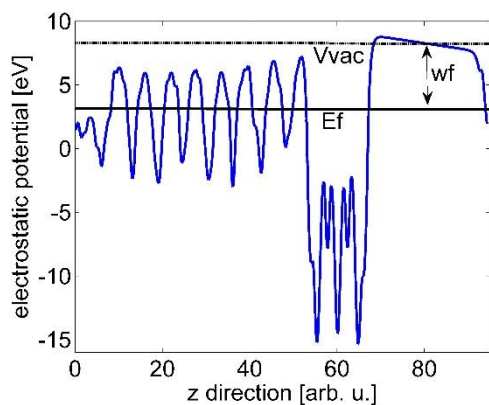


Fig.1: Electrostatic potential profile of $CH_3NH_3PbI_3/Ti_3C_2O_2$ heterojunction in the direction orthogonal to the interface.

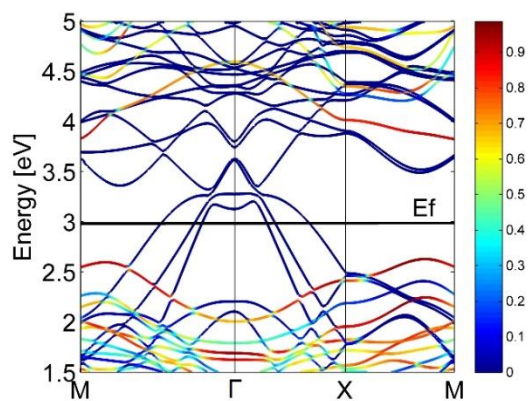


Fig.2: Projected band structure of $CH_3NH_3PbI_3/Ti_3C_2O_2$ heterojunction. The contribution from $CH_3NH_3PbI_3$ electronic orbitals is represented in red.

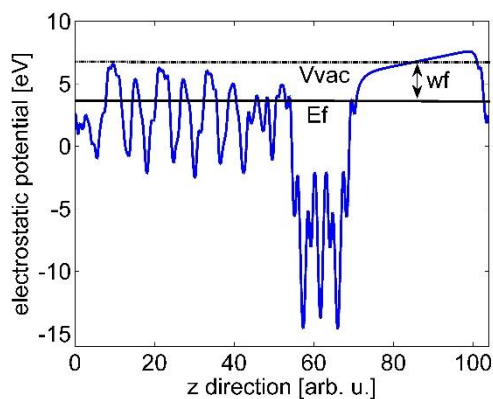


Fig.3: Electrostatic potential profile of $CH_3NH_3PbI_3/Ti_3C_2(OH)_2$ heterojunction in the direction orthogonal to the interface.

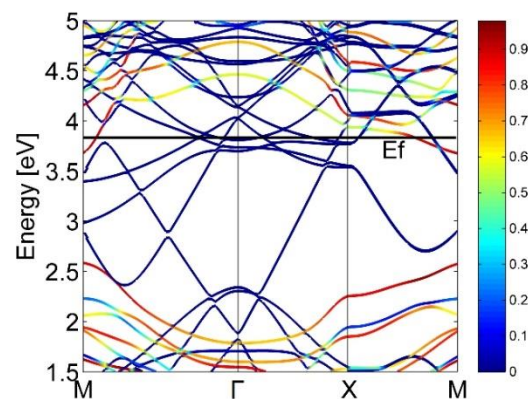


Fig.4: Projected band structure of $CH_3NH_3PbI_3/Ti_3C_2(OH)_2$ heterojunction. The contribution from $CH_3NH_3PbI_3$ electronic orbitals is represented in red.