Theoretical insights to tune the surface dipoles and work functions of halide perovskites

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Abstract:

Lead halide perovskites have attracted considerable interest owing to their rich physics and rapidly burgeoning applications in the field of optoelectronic devices[1, 2]. Therefore, an accurate theoretical description of the link between surface dipoles and the work functions, especially to understand the interface effects in this class of materials is of scientific and practical interest[3]. Herein, we briefly introduce our methodology[4] that accentuates the relation between the surface dipoles and the work functions using concepts from classical physics combined with the state-of-the-art first-principles calculations. We extend the application of our methodology to inspect the fine-tuning of the work function of the prototypical model system i.e $CsPbBr_3$ perovskite surface via surface passivation mechanisms through dipolar ligand molecules. We show the influence of ligands at the surface, that cause an increase or decrease in the work function shifts, thereby considered as positive and negative dipoles, respectively. Furthermore, we study the ligand layer coverage (from 0% to 100% coverage) for tailoring the surface dipoles and accordingly the work functions, and discern that the latter can be tuned by several thousands of meV. At 100% coverage, we capture the steric hindrance for bulky ligands that agree with the previous experimental reports. Finally, we compare our results to those predicted from conventional classical approaches based on the "three capacitor models" that link the dipole interlayer-induced vacuum level shift and the molecular dipole moment. Our findings identify recipes to fine-tune materials work functions and provide valuable insights into the interfacial engineering of this family of optoelectronic devices.

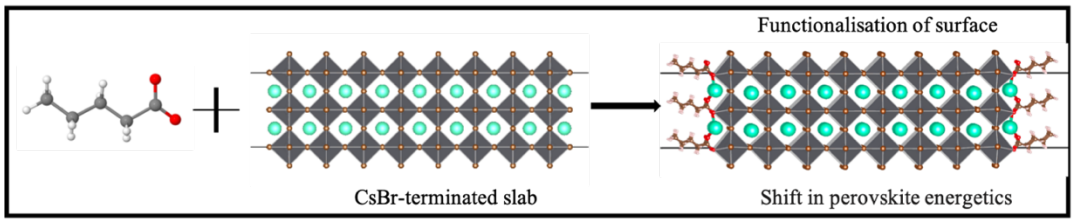

Figure1: Surface functionalization of perovskites via molecules

References:

1) Katan *et al.,* Chem. Rev. 119, 3140 (2019).
2) Traore *et al.,* ACS Nano 12, 3321 (2018).
3) Canil *et al.,* Energy Environ. Sci. 14, 1429 (2021).
4) Traore *et al.,* manuscript submitted.

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