Computational Understandings Of Hybrid Perovskites For Their Application In Photovoltaics

Solar cells based on the hybrid halide perovskite, CH₃NH₃PbI₃, have now reached a confirmed efficiency of 18%, demonstrating a pace for improvements with no precedents in the solar energy arena. Despite such explosive progress, the microscopic origin behind the success of such material is still debated and in particular it is not clear what role the organic component play in the light-harvesting process. We will show using electronic structure calculations that the organic molecules do contribute to the band structure close to the bandgap and they play a fundamental role in determining the crystal geometry. The high-temperature cubic phase of CH₃NH₃PbI₃ allows the molecules CH₃NH₃ to rotate, thus causing the octahedral PbI₆ cage to distort. Such distortion is the direct result of van der Waals interactions that once we include in our calculation reveal drastic consequences on the electronic structure. Charge transport properties of hybrid halide perovskites are also investigated with a combination of density functional theory including van der Waals interaction and the Boltzmann theory for diffusive transport in the relaxation time approximation. Our analysis suggests that the mobility is probably not a key factor in determining the high solar-harvesting efficiency of this class of materials.

Abstract
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