

Lead-Free Perovskites for Photocatalytic H₂ Production: (In-)stability and Reaction Mechanism from First-Principles Simulations

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Metal-halide perovskites (MHPs) are promising materials for a variety of energy conversion applications due to their outstanding optoelectronic properties. In principle, MHPs bring along favorable properties for heterogeneous photocatalysis due to their tunable electronic band gap and electronic energy levels, but the poor stability hampered the development of MHP photocatalysts. In the last few years, to our surprise, water-stable and photocatalytically active tin-halide perovskites (THPs) and double perovskites [1, 2] have been demonstrated, paving the way towards the design of lead-free photocatalysts for hydrogen production. In this talk, I will present recent results from first-principles simulations that unveil the atomistic origins of the (in-)stability of THPs in water and compare these to their lead-based counterparts. In detail, ab initio molecular dynamics simulations on THP/water interfaces demonstrate the origin of degradation of MASnI₃ in water and shed light on the surprising water-stability of DMASnBr₃ [3]. Furthermore, high-level density functional theory (DFT) calculations on the key steps of the hydrogen production mechanism at the perovskite/water interface are presented [4]. The peculiar importance of surface defects and localized charge carriers at the perovskite surface are discussed for both THPs and Cs₂AgBiX₆ (X=Br, Cl) double perovskites.

References:

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