Hybrid functionals for ABX_3 halide perovskites: Influence of Spin-Orbit Coupling

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ABSTRACT

Hybrid organic/inorganic halide perovskites are promising candidates for their use as solar light absorber materials in next-generation photovoltaic cells, which exhibit competitive power conversion efficiencies. The refinement of their chemical composition is key for obtaining fine-tuned optoelectronic properties and long-term stability. High-throughput screening approaches are used for the sampling of their chemical composition, which require an optimized and accurate prediction of their properties.

On the one hand, hybrid functionals can be optimized to yield a description of structural and electronic properties of materials in good agreement with experiment, at the same level as most sophisticated methods based on the GW approximation [1, 2]; on the other hand, for some perovskites, the Spin-Orbit Coupling (SOC) is needed to obtain a good description of data, such as their band gap [3]. In this work, we investigate the relevance of the explicit inclusion of SOC in hybrid functionals on ABX_3 perovskites (A = K, Rb, Cs ; B = Ge, Sn, Pb and X = Cl, Br, and I). The effect of the Fock mixing with or without SOC has been explored: whatever its value, the SOC does not induce changes on structural properties; however, its inclusion affects electronic properties, with the lifting of degeneracy in the band structure and the reduction of the band gap for heavier B elements. For Fock mixing reproducing the experimental band gap, the SOC has a negligeable effect on properties such as effective masses or band widths.

References

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