Density functional theory-based ab initio Molecular dynamics of a chiral perovskite's spectroscopic properties

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ABSTRACT

The market of photovoltaic panels is under a constant growth due to the high demand of new sources of renewable energy [1]. The development of new technologies viable for industrial use depends on factors such as the efficiency in energy conversion provided by the technology, the durability of the materials and the low cost of production.

The development of hybrid perovskites as semiconductors started in the last twenty years. Our study aims at testing using Density Functional Theory (DFT) the optoelectronic properties of a Hybrid Organic-Inorganic Perovskite (HOIP) previously characterized experimentally [2], (S/R)-3BrMBA$_2$PbI$_4$. This perovskite is formed by layers of chiral organic cations (3-BrMBA) in-between a lead-halide (iodine) perovskite lattice.

The study of the spectroscopic properties of the crystal structure was conducted using picosecond-scale *ab initio* Molecular Dynamics (AIMD) performed with Quantum Espresso to explore the conformational space. The absorption and circular dichroism spectra were then obtained by convolution of vertical transitions calculated in the framework of time-dependent-DFT (TD-DFT).

Due to the presence of a non-negligible charge transfer within the crystal structure, a pure functional such as PBE showed a largely shifted spectra. Thus, we run a large benchmark of functionals to evaluate their effect on the spectroscopic properties. Namely, six functionals, APFD, ωB97XD, HSEH1PBE CAMB3LYP, M11 and LC-ωHPBE, have been chosen.

As expected, the functionals including long-range corrections show promising results. Yet, the computed spectra obtained still lack the excitonic excitation peak observed experimentally at 500nm. We can also notice that the CD often shows the opposite expected sign for the studied chirality. Further, highlighting the complex interplay between conformational space and electronic structure exploration.

References

[1] IEA (2023), *World Energy Outlook 2023*, IEA, Paris

[2] Shangpu Liu *et al.,* Bright circularly polarized photoluminescence in chiral layered hybrid lead-halide perovskites. *Sci. Adv.,* **2023** *9*, 5083