Insight into electronic and structural properties of $CH_3NH_3PbX_3$ (X=Cl, Br, I) hybrid perovskites from tight binding modeling and NMR spectroscopy

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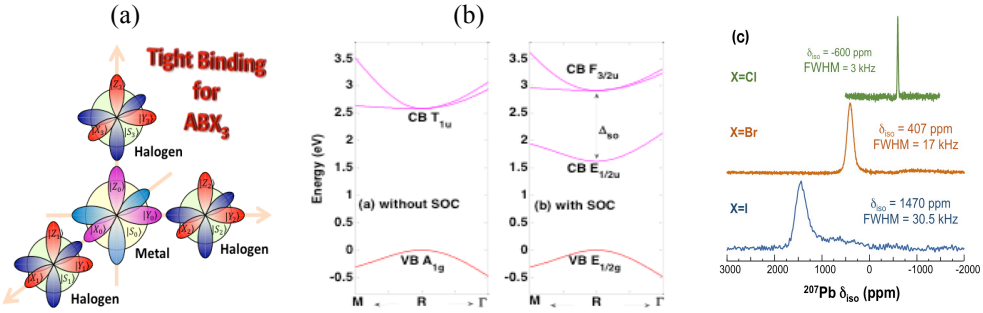
A significant breakthrough was achieved in the field of photovoltaics thanks to the use of hybrid perovskites in the context of solar cells. Currently, this extends to other technological applications such as lasers or detectors. The leading or prototype material has the general formula $CH_3NH_3PbX_3$, where X is one or a mixture of halogen elements. The accelerating pace of scientific discovery and technological innovation calls for deeper insight into the intrinsic physical properties of these materials with major issues related to technological barriers.

To this end, we herein combine theoretical and experimental atomic-scale tools and investigate thoroughly the prototype hybrid perovskite for X=Cl, Br, I. First, solid-state physics concepts and semi-empirical simulations are implemented to investigate their optoelectronic properties, taking the pseudo-cubic reference perovskite structure. For instance, results from a newly developed tight-binding (TB) Hamiltonian, including spin-orbit coupling (SOC), are compared to those afforded by state-of-the-art density functional theory (DFT) calculations. Such a semi-empirical tool provides a low cost alternative to process more complex hybrid perovskite structures [1].



(a) Illustration of sp^3 TB model (b) TB computed diagram with and without SOC (c) ^{207}Pb static NMR spectrum obtained at 14T on $CH_3NH_3PbX_3$.

Next, we show that static ^{207}Pb and low temperature ^{2}H NMR are relevant tools to study the local order and the dynamics in such hybrid perovskites [2]. It is demonstrated that the nature of the halogen element has a dramatic impact on the isotropic shift of ^{207}Pb. ^{2}H NMR lineshapes recorded between room temperature (RT) down to 25 are consistent with freezing of the dynamical disorder existing in a RT plastic phase, and an orientational glassy behavior at low temperature [2].

References

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