

Symmetry-analysis and atomistic simulations of layered perovskites: drawing the roadmap for current and future investigations

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On the wave of their success in light emission and energy conversion applications, layered metal halide perovskite frames have been subject to intense investigations. As a result, we are currently testifying the accumulation of a huge body of evidences related to the optical and electronic properties of these materials, in reference for instance to the role of the dielectric contrast between the inorganic and organic component,[1] or to the shift in paradigm from the traditional quantum-well level alignment to a “heterojunction-like” type II interface, following the incorporation of the π -conjugated organic spacers.[2] On the other hand, orienting in such rapidly evolving field is getting harder and harder, hence calling for the identification of a few concepts of general-interest acting as a road-map for more specific discussions.

A sketch of this roadmap will be presented here, taking advantage of very general symmetry-based, group theory analysis, so to pave the ground for broader discussions, like the identification of the expected evolution of the electronic and excitonic structure of halide perovskite materials, when going from 3D, to 2D.[3] These results will be then referred to recent spectroscopic experiments[4-6] and to cutting edge first-principle calculations,[7] so demonstrating the usefulness of modern first-principle simulations tools to support the interpretation of experimental investigations. Excitons dominate the optical response of two-dimensional layered perovskite materials, where quantum and dielectric confinement enhance the electron-hole interactions;[1] still, they are shown to play a fundamental role also for double perovskites already in 3D,[8-9]. Time permitting, the discussion will also include electron/exciton phonon interactions in a very simplistic way, so moving from zero Kelvin, frozen atom perspective to more realistic effects, as related to cooling of the photogenerated carriers.[10]

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