Theoretical insight on quantum and dielectric confinement in metal-halide perovskites

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Both all inorganic and hybrid halide perovskites recently demonstrated undeniably remarkable characteristics for a wide range of optoelectronic applications. The perovskite fever began with 3D halide perovskites of chemical formula AMX_3 with A a small organic or an inorganic cation, M a metal (Pb^{2+}, Sn^{2+}, Ge^{2+}), and X a halogen (I^-, Br^-, Cl^-), which opened a route toward low-cost manufacture of solar cells, offering currently certified conversion efficiencies over 25%.[1] Since the initial breakthrough mid-2012,[2] halide perovskites attracted worldwide efforts from the scientific community[3] leading to an extensive exploration of their structural versatility and an ever-growing diversity of structures.[3] Prior to the perovskite fever, especially in the 90's, most experimental efforts on halide perovskites focused on chemistry and optical characterizations of monolayered halide perovskites of chemical formula $A'MX_4$, with A' a larger organic cation.[4]

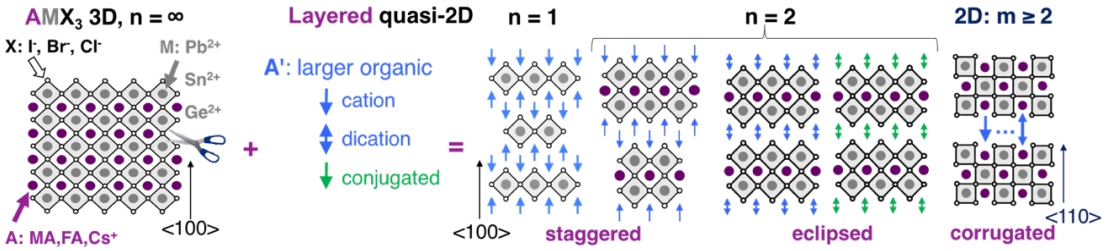

Figure : Layered metal-halide structures conceptually derived from the mother 3D perovskite network

Currently, many different perovskite as well as non-perovskite metal-halide networks are synthesized and their optoelectronic properties deserve to be unraveled (Figure). Among others, new compositions such as $A'_2A_{n-1}M_nX_{3n+1}$ afford layered structures with a controlled number (n) of octahedra in the perovskite layer and thus offer an ideal platform for fundamental understanding.[3,5] Here, through a couple of recent examples,[5,6] we will discuss their optoelectronic properties based on theoretical modeling. Impact of composition and structural pattern on properties will be inspected, with particular emphasis on the effect of quantum and dielectric confinements on charge carriers and excitons.

[1] https://www.nrel.gov/pv/cell-efficiency.html. [2] M. M. Lee & al. *JACS* **134**, 17396 (2012); H.-S Kim & al. *Sci. Rep.* **2**, 591(2012). [2] Special issue, *Chem. Rev.* **119**, 3033 (2019). [3] C. Katan & al. *Chem. Rev.* **119**, 3140 (2019). [4] G. C. Papavassiliou *Prog. Solid State Chem.* **25**, 125 (1997); D. B. Mitzi *Prog. Inorg. Chem.* John Wiley & Sons (1999). [5] L. Mao & al. Chem, in press (2019). [6] M. Smith & al. Chem. Sci. **8**, 1960 (2017); J.-C. Blancon & al. *Science* **355**, 1288 (2017); B. Traore & al. *ACS Nano* **12**, 3321 (2018); J.-C. Blancon & al. *Nat. Com.* **9**, 2254 (2018); M. Kepenekian & al. *Nano Lett.* **18**, 5603 (2018); J. Leveillee & al. *PRMater.* **2**, 105406 (2018) and submitted. X. Li & al. *Chem. Mater.* **31**, 3582 and *JACS* **141**, 12880 (2019).