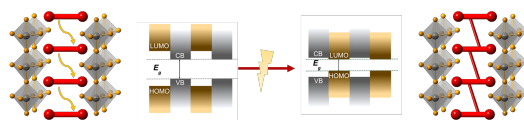


### Acetylene-Based Layered Hybrid Perovskites

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Hybrid organic-inorganic metal halide perovskites have become one of the dominant semiconductors in the field of photovoltaics. They are soft yet crystalline mixed ionic-electronic conductors that have unique optoelectronic and optoionic properties. However, they suffer from instability under device operating conditions.<sup>[1,2]</sup> This stimulated the development of low-dimensional and layered (2D) perovskites as a more stable alternative based on hydrophobic organic cations that act as spacers, templating halide perovskite slabs.<sup>[2]</sup> However, conventional organic spacers are mostly electronically insulating species that form quantum well electronic structures with charges confined within the inorganic layers, limiting their transport and lowering power conversion efficiency in solar cells.<sup>[2-4]</sup> This could be addressed by incorporating extended (opto)electronically active organic moieties within layered hybrid perovskite frameworks, which remains challenging. We demonstrate the capacity to extend the functionality of Ruddlesden-Popper and Dion-Jacobson 2D perovskite materials by incorporating acetylene-based organic spacers.<sup>[5]</sup> The formation of hybrid 2D perovskites is evidenced by a combination of techniques, including X-ray diffraction and solid-state NMR spectroscopy. Moreover, we assess their opto(electro)ionic characteristics by UV-vis absorption, photoluminescence emission, and impedance spectroscopy, showcasing changes in their functionality in response to light. This opens perspectives toward multifunctional hybrid materials in opto(electro)ionics.



*Schematic representation of layered hybrid perovskites with their quantum well electronic structure (left) and light-induced changes of their functionality (right)*

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