Tuning Optical Excitations in Quasi-Low Dimensional Hybrid Organic-Inorganic Metal-Halide Perovskites

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Hybrid organic-inorganic halide perovskites have advanced at an unparalleled pace, with perovskite solar cells exceeding the performance with established technologies after a decade of development. While 3D halide perovskites claim most of this success, quasi-2D (Q2D) hybrid organic inorganic metal-halide perovskites are increasingly popular due to interesting excited state properties enhanced by low dimensionality, as well as their chemical and structural heterogeneity [1]. New and evermore complex Q2D perovskites are continuously reported in literature [2], but in-depth understanding of their optoelectronic properties is not keeping up this rapid materials design pace.

In this talk I will present some of our recent results focused on understanding the role of structure and chemical composition in the excited state properties of Q2D layered perovskites using state-of-the-art first principles computational modeling methods based on many-body perturbation theory [3]. We approach this problem by first deconstructing the complex Q2D perovskite, and separately understanding how the structure and chemistry of the organic and inorganic components contribute to their optoelectronic properties, by computing their quasiparticle band structure and optical absorption spectra within the GW approximation and the Bethe-Salpeter equation framework (GW+BSE)[4,5]. In this presentation, I will show our studies of two types of layered perovskites distinguished by the alignment of adjacent inorganic layers. I will discuss how key structural features can facilitate tuning of the energy and spatial delocalization of excitons in these complex materials using model structures, and show how this may be realized in real Q2D perovskites [6].

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