Understanding Excitons in Metal Halide Perovskites using First Principles Computational Modeling and High Performance Computing

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Organic-inorganic metal-halide perovskites are one of the most exciting families of optoelectronic materials, attracting a wide interest in the research community cross disciplines, including in fundamental Condensed Matter Physics, Materials Science and Chemistry, as well as Applied Physics and Optoelectronics. Implementation of this family of materials in optoelectronic devices has shown rapid progress, including in solar cells with record break efficiencies, LEDs, photo-detectors, etc. However, despite this growing success, this materials family is largely unexplored with many fundamental properties not well understood, leaving an untapped opportunity to drive innovation through targeted materials design. In this project we aim to fill this knowledge gap using state-of-the-art first principles computational materials modeling and high-performance computing.

3D Halide perovskite have a formula unit of ABX3 (with A = Cs, MA, FA, etc, B = Pb, Sn and X = Cl, Br, I); several new derived materials have recently been designed and synthesized including 3D halide double perovskites Cs 2 BB'X 6 (with B = Bi, Sb, In and B'=Cu, Ag, Au) [1-4], as well as an even wider variety of quasi-2D (Q2D) layered perovskites, including Ruddlesden-Popper and Dion-Jacobson type perovskites and perovskite-intergrowth hetero-structures [5,6].

In this presentation I will give a brief outline of some of our recent results in understanding the optoelectronic properties of this wide variety of halide perovskites with different chemistries and dimensionalities using first principles many-body perturbation theory methods within the GW approximation [7] and the Bethe-Salpeter equation [8]. I will present recent GW+BSE calculations and highlight the excitonic properties in prototypical CsPbX3 and mixed-halide 3D perovskites, Cs2BB'X6 halide 3D double perovskites [9], quasi-2D halide perovskites and hetero-structures [6], and discuss the impact of structure and chemical composition in these systems. Furthermore, I will highlight the role of the A-site organic cation in the screening of excitons, alongside the impact of dimensionality optoelectronic properties of quasi-2D Ruddlesden Popper halide perovskites, and show how hetero-structures impact optical properties of quasi-2D layered perovskites. I will summarize by outlining the intuition drawn from these closely related materials and give my outlook for future work.

[1] Filip, Volonakis & Giustino, Handbook of Materials Modeling: Applications: Current and Emerging Materials, Springer International Publishing, 295-324 (2020).
[2] Volonakis, Filip, Haghighirad, Sakai, Wenger, Snaith & Giustino, J. Phys. Chem. Lett. 7 (7), 1254-1259 (2016).
[3] Filip, Hillman, Haghighirad, Snaith & Giustino, J. Phys. Chem. Lett. 7 (13), 2579-2585 (2016).
[4] Volonakis, Haghighirad, Milot, Sio, Filip, Wenger, Johnston, Herz, Snaith & Giustino, J. Phys. Chem. Lett. 8 (4), 772-778 (2017)
[5] Smith, Crace Jaffee & Karunadasa, Annu. Rev. Mater. Res., 48, 111-136 (2018).
[6] Aubrey, Valdes, Filip, Connor, Lindquist, Neaton & Karunadasa, Nature, 597, 355-359 (2021)
[7] Hybertsen & Louie, Phys. Rev. B 34, 5390 (1986).
[8] Rohlfing & Louie, Phys. Rev. Lett. 81, 2312 (1998).
[9] Biega, Filip, Leppert & Neaton, J. Phys. Chem. Lett.12 (8), 2057-2063 (2021).