Understanding Delocalization and Dissociation of Excitons in Heterogeneous Semiconductors from First Principles Computational Modeling

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Excitons are composite quasiparticles which consist of correlated photoexcited electron-hole pairs. Understanding exciton formation, delocalization and dissociatiation in complex heterogeneous semiconductors is of key importance for their deployment to optoelectronic applications, including photovoltaics, lighting and lasing. Development of new computational modeling techniques are capable to describe interactions between excitons and other quasiparticles constitutes a frontier first principles computational modeling of materials. The state of the art approach to compute optical excitation energies in semiconductors and insulators is the GW+Bethe-Salpeter Equation (GW+BSE) approach [1,2]. This framework is extremely powerful and provides the foundation of new methods aimed at describing complex excited state phenomena.

In this talk, I will present our recent first principles study of optical excitations in several complex organic-inorganic layered lead-halide perovskites, including Ruddelsden-Popper, Dion Jacobson layered perovskites and perovskite-intergrowth hetero-structures [3-6] using state-of-the-art GW+BSE. I will show how the exciton binding energy and spatial delocalization of excitons can be readily tuned by controlling structural and chemical properties of the organic and inorganic layers of these systems,. Time permitting, I will also briefly present a new first principles approach we have developed, based on GW+BSE, which allows us to understand the impact ionic vibrations have on the dielectric screening of excitons from first principles [7,8] and compute temperature dependent exciton binding energies and dissociation rates [8].

References

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