First-principles modelling of excitons in metal-halide perovskites and beyond

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Metal-halide perovskites are a structurally, chemically, and electronically diverse class of semiconductors with applications ranging from photovoltaics to radiation detectors and sensors. Understanding neutral electron-hole excitations (excitons) is key for predicting and improving the efficiency of energy-conversion processes in these materials. First-principles calculations have played an important role in this context, allowing for detailed insight into the formation of excitons in many different types of perovskites [1]. Such calculations have demonstrated that excitons in some perovskites significantly deviate from canonical models due to the chemical and structural heterogeneity of these materials [2-5]. In this presentation, I will provide an overview of our current understanding of excitons in metal-halide perovskites. I will focus on results based on Green's function-based many-body perturbation theory in the GW+Bethe-Salpeter Equation approach, the prevalent method for calculating excitons in extended solids [6, 7]. This approach readily considers the anisotropic electronic structures and dielectric screening present in many perovskites and other heterogeneous semiconductors, and important effects such as spin-orbit coupling. However, the complex and diverse electronic structure of these materials, and its intricate coupling to pronounced and anharmonic structural dynamics [8 - 10], pose challenges which are currently not fully addressed within the GW+Bethe-Salpeter Equation approach, and which I will also briefly discuss in this contribution.

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