Predictive computational approaches to complex excited state phenomena in functional materials

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Optical and excited-state phenomena are key ingredients in functional materials characterization, dominating emerging applications such as optimization of solar energy conversion and storage, designing photon emitters, and generating Bose-Einstein condensates. Excited-state properties, including linear and non-linear absorption, as well as radiative and non-radiative exciton decay mechanisms and lifetimes, are strongly related to material structure. Recent advances allow a controlled design of structurally complex materials, along with close tracking of excited-state processes and their dependence in structural modifications. Understanding such structural effects on the excitonic phenomena from a theory perspective is challenging, as it demands predictive access to the involved excitations, strongly determined by the structural perturbation. In this talk I will describe a computational assessment of the relation between excitonic phenomena and material structural complexities, using many-body perturbation theory within the GW and Bethe-Salpeter equation (GW-BSE) approach. I will discuss the effect of atomic defects and heterostructures on the excitonic properties in layered transition metal dichalcogenides (TMDs), where the structural complexity leads to mixed excitonic transitions between states of different nature and localization, determining unique and tunable selection rules and absorption. I will further present a GW-BSE-based approach to study exciton transport with relation to material structure and symmetry, demonstrated on selected systems of varying dimensionalities.