Ab-initio calculations and simple models of electronic excitations in 2D materials and heterostructures

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Abstract

Many-body calculations based on the GW approximation to the electron self-energy and the Bethe-Salpeter equation (BSE) for the density response function are powerful methods for predicting band structures and optical excitations in real materials from first principles. We illustrate the high accuracy obtained by these methods through examples from our computational 2D materials database [1]. In 2D materials electron-electron interaction effects are particularly important because of the weak dielectric screening resulting in large self-energy corrections to band energies and strong excitonic effects. We show that the exciton binding energy can be approximated by a simple 2D Hydrogenic model taking an effective 2D dielectric constant and the exciton mass as input. Remarkably, the exciton binding energy depends only weakly on the exciton mass, and consequently the binding energy is found to scale directly with the size of the band gap – an effect unique to 2D excitons [2]. The simple Hydrogenic model also allows us to obtain the field-induced dissociation rates of 2D excitons by the technique of complex scaling to compute resonance life times. We show that the dissociation rates can be tuned significantly by embedding the active 2D material in a van der Waals heterostructures [3]. In general, periodic ab-initio calculations for van der Waals heterostructures are complicated by the incommensurable nature of the interfaces. We show that by neglecting the effect of hybridization and thus assuming a purely electrostatic coupling between the layers, it is possible to compute the dielectric properties of large incommensurable van der Waals heterostructures from the dielectric function of the individual layers [4]. The latter can be calculated once and for all and stored in a database. We illustrate how the Quantum Electrostatic Heterostructure (QEH) model can be used as a tool for modeling excitons, plasmons, and optical properties of van der Waals heterostructures [5].

References

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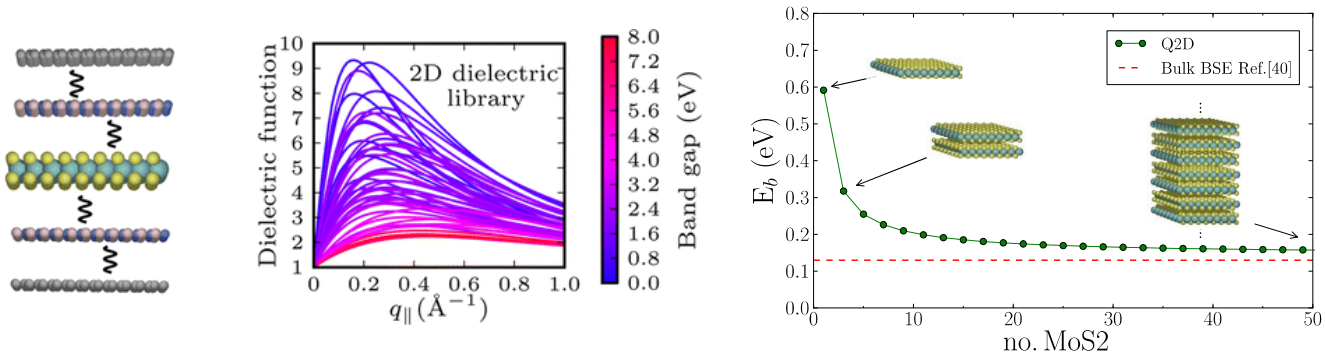

Figure: Left: The QEH model calculates the dielectric function of a vdWH from the dielectric functions of the individual 2D crystals assuming a pure Coulombic interaction between the layers. Middle: Static dielectric function of 50 semiconducting TMDs. Right: Exciton binding energy in MoS_2 slabs of varying thickness. The exciton is calculated using a (quasi-)2D Hydrogen model with screened electron-hole interaction obtained from the QEH model. The results are seen to converge towards the bulk result showing the importance of dielectric screening relative to quantum confinement (not taken into account).