Carrier concentration-dependent excitons in van der Waals heterostructures from ab initio many-body perturbation theory

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Interfacing transition metal dichalcogenides (TMDs) into van der Waals heterostructure bilayers with type-II level alignment has led to recent reports of interlayer excitons with large binding energies, long lifetimes, and signatures of exciton condensation at elevated temperatures. Atomically flat two-dimensional molecular crystals (MC) on TMD monolayers is an emerging interfacial quantum materials platform with tunable level alignment, exciton binding energies, and photoluminescence, given the heightened sensitivity of the organic layers to their environment. In addition to non-local adsorbate/substrate screening, free charge carrier screening is particularly relevant in MC-TMD bilayer heterostructures, and controllably altering these distinct modes of screening can lead to new phenomena. Using a dielectric embedding ab initio GW plus Bethe-Salpeter equation (GW-BSE) approach [1], we compute energy level alignment as well as neutral excitations at MC-TMD interfaces (MC = PDI or PTCDA ; TMD = MoS_2 or WS_2), exploring new emergent optical transitions, such as those associated with interlayer excitons characterized by electrons and holes separated between the MC adsorbate and the TMD, respectively [2-3]. Using an extended plasmon-pole model reducing computational cost with no loss of accuracy [4-5] and explicit addition of electrons, we also explore the role of free charge carriers in screening electron-hole interactions, quasiparticle energy level alignment, and the nature of interlayer excitons.

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