Many-body effects on the electronic and optical properties of quasi-two-dimensional materials

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Recently, transition metal dichalcogenides (TMDs), such as MoS_2, $MoSe_2$, WS_2, and WSe_2, have been isolated in mono- and few-layer forms. These atomically-thin materials exhibit many remarkable properties, such as valley-selective optical selections rules and excitons with huge binding energies, and are the subject of intense research interest. We have applied the GW and GW plus Bethe Salpeter equation (GW-BSE) approaches to the electronic and optical properties of these quasi-two-dimensional (2D) materials. We found that the TMDs have multiple series of excitons with binding energies in excess of 0.6 eV, and excitation spectra (excited exciton levels) that cannot be explained by the usual 2D hydrogenic model [1,2]. Both the quasiparticle band gaps and the binding energies are very sensitive to the environment in which the 2D materials are placed. We have also studied excitonic effects and energy loss spectra of electron-hole excitations at finite momentum transfer **q** in the TMDs. We found that the binding energies of the finite center-of-mass momentum excitons (e.g., those corresponding to transitions across the indirect gap with **q**= ½ (Γ➔K) in the Brillouin zone) are as large as those of the direct gap (**q**=0) excitons, and have similarly non-hydrogenic excitation spectra and unusual selection rules. The large binding energies, non-hydrogenic spectra, and the sensitivity of the bandgap and exciton binding energies to substrate screening are all found to be a consequence of a strong spatial variations in the screening of the many-electron interactions due to the quasi-2D nature of the material [3]. These three factors -- i) the necessity to include environmental screening, ii) the strong spatial variations in screening, and iii) the large extent of exciton wavefunctions in real space -- make the study of the optical properties of quasi-2D systems very computationally challenging. For example, it requires in general a sampling of more than 100,000 k-points in the Brillouin zone to obtain converged results. We have developed new interpolation methods and k-point sampling techniques that reduce the computational cost by orders of magnitude, making these studies computationally feasible [4].

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