Unraveling Exciton Dynamics in 2D Van der Waals Heterostructures

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Two-dimensional (2D) van der Waals (vdW) heterostructures offer a fascinating platform to pursue fundamental science and novel device applications. Owing to quantum size confinement and reduced dielectric screening, electron-electron interaction and excitonic effect are prominent in 2D materials, which could dominate their electron and exciton dynamics. Recent experiments have revealed ultrafast charge (~100 fs) and energy (~1 ps) transfer dynamics in transition metal dichalcogenides (TMDs) heterostructures, which is surprising giving the strong electron-hole binding of the interlayer excitons. Most theoretical calculations to understand the experiments were performed based on local and semi-local exchange-correlation functionals, thus cannot capture the excitonic effect accurately. In this talk, I will introduce a first-principles method that combines non-adiabatic molecular dynamics (with the fewest switch surface hopping algorithm) and linear-response time-dependent density functional theory. Importantly, the method is formulated in the planewave bases and PAW pseudopotentials with range-separated hybrid functionals. As a result, the method can capture the excitonic effect accurately in extended 2D heterostructures. Using this method, we can shed light on the ultrafast charge transfer dynamics in TMD vdW heterostructures and elucidate the role of “hot” excitons in promoting ultrafast charge transfer despite the momentum mismatch in twisted heterostructures. We can also examine the properties of interlayer and intralayer excitons trapped by the moiré superlattices in the twisted heterostructures, and tune their properties and lifetimes by changing the twist angle, pressure and electric field.