Dynamics of Excitonic Valley Transfer in Transition Metal Dichalcogenide Multilayer Heterostructures

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The valley degree of freedom of excitons in atomically thin transition metal dichalcogenides (TMDs) is an intriguing candidate for implementing quantum information carriers in nanoscale optoelectronic devices. It has been shown, both theoretically and experimentally, that the dynamics of exciton valley pseudospin is governed by the electron-hole exchange Coulomb interaction, which is analogous to an in-plane pseudo-magnetic field acting on the pseudospin. However, in practice, multilayer stacking of atomically thin 2D materials is inevitable in designing functional nanoscale devices. In 2D van der Waals multilayer structures, excitons in different monolayer structures interact with each other via the excitonic transition dipole-dipole interaction, which opens channels for rapid interlayer exciton transfer. Interestingly, the interlayer excitonic dipole-dipole interaction is strong and comparable to the intralayer exchange Coulomb interaction. The interplay of these two excitonic valley transfer mechanisms provides new possibilities for potential applications.

In this work, we investigate the dynamics of valley-bound excitons in TMD monolayers embedded in 2D van der Waals multilayer structures using the quantum master equation approach. We found that the strength of the interlayer exchange Coulomb interaction does not sensitively depend on the separation between the monolayer structures, but is screened by the sandwiching dielectric materials. Our results show that, under the influences of the excitonic active monolayers, the flipping rate of valley polarization is enhanced due to the interlayer exciton transfer. That is, the depolarization of valley pseudospin can be tuned by inserting different dielectric materials between the two TMD monolayers. This makes the valley coherence control possible in designing nanoscale optoelectronic devices.