Abstract

Exploring the Optical Properties of Two-Dimensional Transition Metal Dichalcogenides Moiré Superlattice

By

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Atomically thin two-dimensional (2D) monolayer transition metal dichalcogenides (TMDs) have garnered significant attention due to their fascinating optical and electrical properties. Unique optical and electronic properties from TMD monolayer can be observed such as high optical absorption, large exciton binding energy, and direct band gap, making them promising candidates for ultrathin potential optoelectronic devices. Moreover, monolayer TMDs exhibit valley-selective optical properties and spin-valley locking, which offer opportunities for manipulating electronic and excitonic new physical degrees of freedom.

Furthermore, the emergence of moiré superlattice occurs when two transition metal dichalcogenides monolayers are stacked together with a slight twist angle and small lattice mismatches, resulting in periodic quantum confinement known as moiré potentials. These moiré potentials serve as a potential trap for the electron-hole pair, "exciton" and giving rise to novel excitonic state as quantum ensembles. The spatial modulation of the electronic potential landscape in moiré superlattice offer strong light-matter interactions and enhanced exciton binding energies. This manifests in sharp photoluminescence peaks, indicative of stable and long-lived excitonic states. Additionally, the moiré excitons exhibit tunable valley and spin degree of freedom at the three-fold rotational symmetry C_3. These characteristics offer promising avenues for valleytronic and spintronic applications. The unique optical and electronic characteristic of moiré exciton in the TMD heterostructure have motivated me to explore their fundamental nature further.

In this thesis, I investigated the fundamental optical properties arising from moiré excitonic systems such as moiré exciton and moiré trion in $MoSe_2/WSe_2$ heterostructure by

photoluminescence (PL) spectroscopy. Initially, my focus was on studying the dynamical behavior of the moiré exciton in *R*-type $MoSe_2/WSe_2$ heterostructure. From the temperature dependent PL experiments, I discovered the presence of dark state lying above the bright state within the moiré potential. Moreover, I elucidated that the dynamics of moiré exciton is predominantly influenced by phonon-assisted transition between bright and dark states with increasing temperature. Additionally, I conducted excitation power dependence of PL spectroscopy, which revealed the emergence of an additional peak at higher energy side, corresponding to the emission from triplet moiré exciton with long lifetime.

Then, I introduced a novel approach to fabricate $MoSe_2/WSe_2$ heterostructure using focused ion beam technique. This approach leads to realize the observation of smaller number of moiré exciton peaks by minimizing the inhomogeneity of the moiré potentials. With taking advantage of this approach, I successfully observed moiré charged excitons (trions) through electrostatic doping and conducted experiments to investigate their dynamics, through temperature dependent and circularly polarization resolved PL spectroscopy. Notably, I observed intriguing negative valley polarization from moiré trions with a temperature-independent long valley relaxation lifetime.

Lastly, I proposed new way to manipulate the dimensionality of moiré excitons by introducing very thin *h*-BN spacer between the TMD monolayers. The *h*-BN spacer strongly suppresses the moiré potentials landscape and leads to dimensionality transition from 0D moiré excitons to 2D free exciton. The change of PL peak position, and strong temperature dependence of radiative exciton decay rate strongly indicate the modulation of moiré potential landscape and change in dimensionality of exciton. Furthermore, I observed long valley polarization in 2D free interlayer exciton reaching to 100 ns under low temperature condition, which is attributed to weak electron-hole exchange interactions.

The studies of the moiré excitons and trions with their dynamics including valley polarization, provide me comprehensive understanding of fundamental optical characteristics in the TMD heterostructure. These investigations have uncovered valuable insights that could serve as a foundational platform for advancing research in quantum potential applications. By elucidating the behavior and optical properties of moiré excitons and trions, I pave the way for exploring new avenues in quantum technology and device engineering.