Exploring Spin and Valley Physics in 2D Materials Using Optical Vortex Beam

Abstract:

Monolayer transition metal dichalcogenides (TMDs) with broken triangular sublattice symmetry originate intriguing valley physics, where electron spin is coupled with the valley degree of freedom due to high spin-orbit splitting of valence and conduction bands. Spin-valley coupling in monolayer TMDs gives rise to temperature-dependent photoluminescence (PL), valley polarization, and coherence effect, limited by intervalley scattering caused by exciton-phonon, exciton-impurity, and electron-hole exchange interaction (EHEI). We demonstrate a unique approach to create excitons at higher momentum states controlled by the topological charge (l) of the optical vortex beam (OV) at near-resonant excitation. We identify the effect of intravalley spin-flip scattering in spin-valley coupled two-dimensional systems by transferring the momentum of light into exciton center of mass using OV beams. By varying the dispersion of light using l of the OV beam, we demonstrate a unique approach to control the intravalley spin-flip scattering rate of excitons. From our photoluminescence measurements, we demonstrate that the intravalley scattering rate in W-based TMDs can be tuned externally by OV beams. Variation of PL intensity with l shows a crossover temperature (> 150 K), indicating competition among time scales involving radiative recombination, spin-flip scattering, and thermal relaxations. Simultaneously we tune intervalley EHEI by controlling excitons center of mass momentum (COM) utilizing the photon distribution of higher-order OV beam. By virtue of this, we have observed excitons COM-dependent valley depolarization and decoherence, which gives us the ability to probe the valley relaxation timescale in a steady-state measurement. But for non-resonant excitation, the phonon cascade process creates a mixture of coherent and incoherent excitons controlled by excitation-detuning energy. At far-resonance excitation (520 nm laser) in WSe_2 for low temperature, PL intensity variation with l mimics the variation expected at a higher temperature regime, indicating thermalization of excitons. Our approach of controlling bright and dark excitons generation rates via relative strength between longitudinal and transverse electric field components is corroborated by observing enhancement of defect peaks and a decrease in trion intensity with l number of OV beam. Our proposed technique utilizing structured light beams can open up a new paradigm to explore the physics of excitons in two-dimensional systems.

Keywords: Optical vortex beam, Angular momentum of light, Transition metal dichalcogenides, Valleytronics, Monolayers, Excitons, e-h exchange interaction, Valley scattering.