Magnetic-field induced intervalley dynamics of excitons in monolayer transition metal dichalcogenides

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The use of external magnetic fields is a powerful tool to investigate the electronic properties of atomically thin semiconductors. Here, we describe our investigation of the intervalley scattering dynamics of intralayer excitons in $MoSe_2$ monolayers and twist-angle controlled $MoS_2/MoSe_2$ heterobilayers. Due to the ultrafast dynamics of the intervalley scattering mechanisms and the recombination processes, the magnetic-field induced valley polarization in monolayers transition metal dichalcogenides is generally explained is terms of a simple population imbalance[1]. However, in these materials the depolarization via electron-hole exchange interaction occurs at a rate comparable to that of the exciton relaxation toward the low-energy Zeeman-split exciton state and should be accounted for to achieve a more accurate description of the intervalley dynamics.

We consider this effect by comparing the dependence of the magnetic-field induced degree of circular polarization of CVD grown and mechanically exfoliated $MoSe_2$ monolayers and of moiré intralayer excitons in a $MoS_2/MoSe_2$ heterostructure (see figure 1(a)). We show that the different trends of the degree of circular polarization (DCP) versus magnetic field shown in figure 1(b) can be explained within a model that accounts for resonant intervalley scattering mechanisms and phonon-assisted exciton relaxation. The valley polarization of the charged excitonic complexes is instead explained by considering the magnetic field dependent of resident carriers in a system with a non-degenerate electron density.

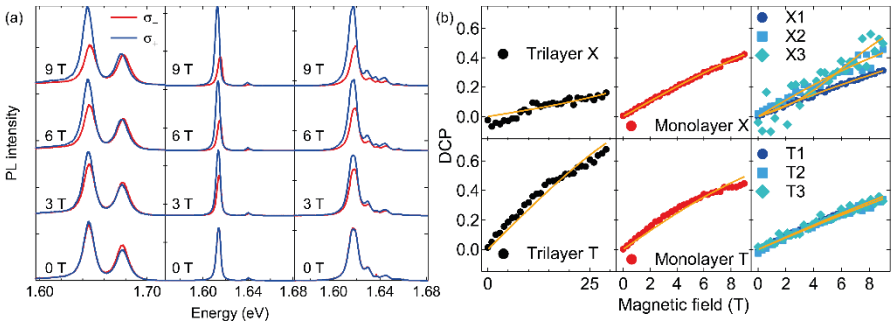

Figure 1. (a) MagnetoPL spectra of a CVD grown $MoSe_2$ monolayer (left panel), a mechanically exfoliated $MoSe_2$ monolayer (central panel) and intralayer moiré excitons in a $MoS_2/MoSe_2$ heterostructure (right panel). (b) Magnetic-field induced degree of circular polarization of the neutral and charged intralayer excitons of the three samples in panel (a). The yellow line are fits to models.

[1] Zhang, et al., Advanced Functional Materials 31, 2006064 (2021)