Chemically brightened and highly valley polarized trions in monolayer MoS₂

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Monolayer (1L) transition metal dichalcogenides (TMDs) MX₂, where M and X are a transition metal and a chalcogen, respectively, are attractive for exploiting valleytronics or optovalleytronics in fast and energy-saving information processing applications. Their valley polarized excitons or charged excitons (trions) are candidates as information carriers [1-5] in optovalleytronics. Trions, which are three-body charged excitonic bound states of electrons and holes, are expected to exhibit a much longer valley lifetime compared to excitons. However, the relatively weak PL of trions prohibits the realistic application of trions in these materials.

Here, we use a p-type F₄TCNQ chemical dopant [6] to treat 1L MoS₂. Chemical dopant treatment is one of the effective methods to enhance PL emission efficiency [6,7]. The polarization resolved point PL mapping method [8] was used to obtain the information of the whole sample at 15 K. PL and valley polarization characteristics of 1L MoS₂ on a SiO₂/Si substrate are evaluated and compared before and after F₄TCNQ treatment. Valley polarization the measurements were carried out using a home-built optical setup under σ + circularly polarized excitation with a wavelength of 633 nm (1.96 eV), which is near the resonance of A-exciton absorption in 1L MoS2 at 15 K and is known to yield relatively high valley polarization [4].

Figure 1 shows the polarization resolved PL spectra at 15 K before (a) and after (b) the F_4TCNQ treatment [9]. The trion PL intensity was enhanced ~ 3 times after the treatment which was mainly attributed to the extension of the trion lifetime via the PL lifetime measurements [9]. Figure 2 shows a valley polarization map of trions (a) and a plot of trion valley polarizations at various positions on the sample versus corresponding PL intensities at the positions. The valley polarizations were kept near constant (~ 0.75) before and after the chemical treatment for the whole sample and



Figure 1. Polarization resolved PL spectra of 1L MoS₂ before (a) and after (b) the treatment. L and X⁻ are the PL signals from localized states and trions, respectively. σ + and σ - indicate the circularly polarized components of the PL spectra.



Figure 2. (a) Valley polarization map of trions in 1L MoS₂ at 15 K before and after the F_4TCNQ treatment. (b) Valley polarization as a function of trion PL intensity at various spots before and after the treatment.

were independent on the trion PL intensity. This insensitivity of the trion valley polarization on their PL intensity can be understood considering the very long valley lifetime of trions (> \sim 1 ns) and the phenomenological relation between the valley polarization, valley lifetime, and PL lifetime of trions [9].

These results showed that the brightening of trions and keeping its high valley polarization are kept compatible in chemically treated 1L MoS₂. The findings further highlight the usefulness of bright and highly valley polarized trion PL in chemically treated 1L MoS₂ in optovalleytronics.

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