Robust trion emission in two-dimensional $Mo(S_xSe_{1-x})_2$ alloys

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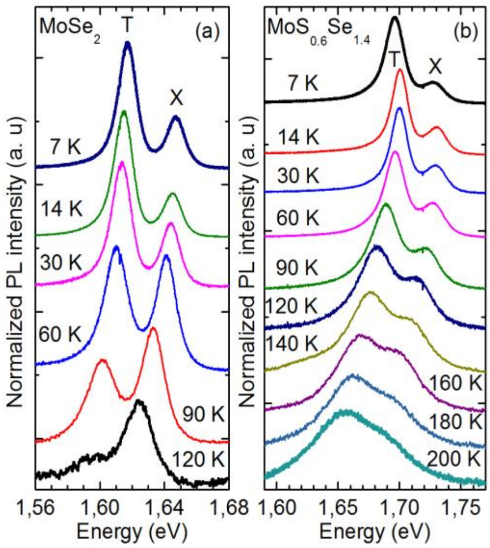

Fig. 1 The temperature evolution of the PL spectra of (a) $MoSe_2$ and (b) $MoS_{0.6}Se_{1.4}$.

Transition metal dichalcogenides (TMDCs) have attracted significant attention due to the discovery of the indirect-to-direct bandgap transition and the coupling of the spin and valley degrees of freedom when reducing thickness to one monolayer. Due to the strong confinement to single layer, the TMDC excitons have very high binding energies of a few hundreds of meV. In optical spectra of $MoSe_2$ and WSe_2 well-resolved exciton and charged exciton (trion) transitions were detected. The strong spin-orbit coupling in these materials leads to the spin splitting of A-exciton between the dark and bright states, approximately equal to the splitting of the conduction band Δ^{cb}_{so}. As predicted in theoretical calculations, Δ^{cb}_{so} for WSe_2 is negative, whereas for $MoSe_2$ it is positive. This mainly results in the contrasting lowest energy exciton sub-bands: dark in WSe_2 and bright in $MoSe_2$. Despite the same value of trion binding energy in both diselenides, there is a significant difference in the temperature evolution of the trion emission intensity. In $MoSe_2$, trion PL intensity strongly decreases with the increase of temperature and is negligible in the spectra at temperatures above $T > 100$ K, whereas in WSe_2 trion emission is prominent even up to $T \approx 200$ K. In both materials, the neutral exciton PL intensity increases with the increasing temperature compared to trion and finally exceeds it at the highest temperatures.

Here, we employ temperature-dependent micro-photoluminescence (μ-PL) spectroscopy to probe variation in the relative intensity ratio between the neutral and charged excitons in $Mo(S_xSe_{1-x})_2$ for the small sulfur content x ($0.1<x<0.3$). We have found that in contrast to $MoSe_2$ and WSe_2, for all samples with sulfur admixture, the trion emission intensity exceeds exciton intensity in the entire temperature range from $T = 7$ K to 200 K. We also present the complementary μ-Raman scattering measurements performed at $T = 300$ K in backscattering geometry. As the composition x slightly increases, we observe the characteristic splitting of the out-of plane vibration A'_1 into two phonon branches. They evolve in different manner as a function of the composition x, and they are likely related to the different distribution of the chalcogenide atoms within the $Mo(S_xSe_{1-x})_2$ layers (Se-Se and Se-S pairs). Interestingly, the trion binding energy, determined from PL spectra, takes the value accurately from the middle of the frequency range definite by corresponding phonon branches. This leads to distinctive resonant condition that makes these 2D semiconductors an excellent platform to study exiton-phonon interaction. We interpret the robust thermal trion emission in $Mo(S_xSe_{1-x})_2$ alloys to strong increase of trion-exciton coupling mediated by optical phonon.