Dark excitonic ground state in $Mo_{0.68}W_{0.34}Se_2$ monolayers

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Monolayers (MLs) of semiconducting transition metal dichalcogenides (S-TMDs) such as $MoSe_2$ and WSe_2 exhibit direct bandgaps and strong excitonic effects, which govern their emission and absorption properties. Depending on the spin configuration of the ground excitonic state, MLs are classified as bright (e.g. $MoSe_2$) or darkish (e.g WSe_2) [1]. In $Mo_xW_{1-x}Se_2$ alloys the emission energy can be continuously tuned by adjusting the relative concentration of Mo and W atoms [2], enabling precise control over their optical properties for photonic applications. We investigate low-temperature photoluminescence (PL) of $Mo_{0.68}W_{0.34}Se_2$ ML encapsulated in hexagonal boron nitride (hBN) under in-plane magnetic ($B_{\parallel}$) fields up to 30 T. Figure 1(a) shows the field evolution of the PL spectra for $Mo_{0.68}W_{0.34}Se_2$. At zero field, the spectrum closely resembles that of bright $MoSe_2$ [1,3] and exhibits two well-resolved lines, labeled X^B and T, attributed to the neutral and charged bright excitons, respectively. Upon applying $B_{\parallel}$ above ~15 T, an additional emission line, X^D, appears. Following previous studies [4], we attribute this line to the dark exciton (X^D) located 16 meV below X^B, whose intensity increases quadratically with $B_{\parallel}$ [Fig. 1(b)], consistent with theory and experiment. This behavior is supported by band-unfolding calculations for $Mo_{0.75}W_{0.25}Se_2$ [Fig. 1(c)], which reveal opposite spin ordering of the valence-band maximum and conduction-band minimum, indicating a dark excitonic ground state in $Mo_{0.68}W_{0.34}Se_2$ ML. We additionally observe a pronounced reduction of the X^B intensity [Fig. 1(b)], evidencing angle-dependent interplay between bright and dark states. These findings reveal the darkish character of $Mo_{0.68}W_{0.34}Se_2$ ML and indicate a more complex magnetic-field-induced redistribution of emission intensity between X^D and X^B, a behavior not previously reported for S-TMD ML.

References

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Figures

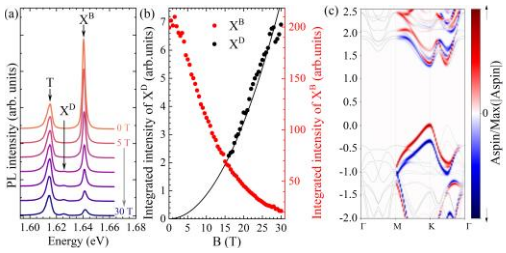

Figure 1: (a) Low-temperature PL spectra of $Mo_{0.68}W_{0.34}Se_2$ ML measured in $B_{\parallel}$ up to 30 T. (b) Quadratic increase of the X^D intensity as a function of $B_{\parallel}$. (c) Spin-resolved band-unfolding calculation at the DFT level for a 2×2×1 supercell of $Mo_{0.75}W_{0.25}Se_2$.