Phonon-mediated upconversion photoluminescence processes in hBN-encapsulated monolayer TMDs

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The monolayer transition metal dichalcogenides (TMDs) emerge as an outstanding platform to study electron- electron and electron- phonon interaction [1, 2]. They host variety of intra- and inter- valley neutral and charged excitonic complexes, which are either optically active or inactive due to the conservation of a spin or a momentum. The higher number of excitonic features positioned energetically below the bright direct exciton have been observed in the low temperature emission spectra of W-based structures mainly due the dark exciton band lying at lower energy than the bright band. They have been identified as bright singlet and triplet trions, neutral and charged bright biexcitons, spin-forbidden dark exciton, dark trions and momentum-indirect dark excitons activated by scattering with defects or phonons.

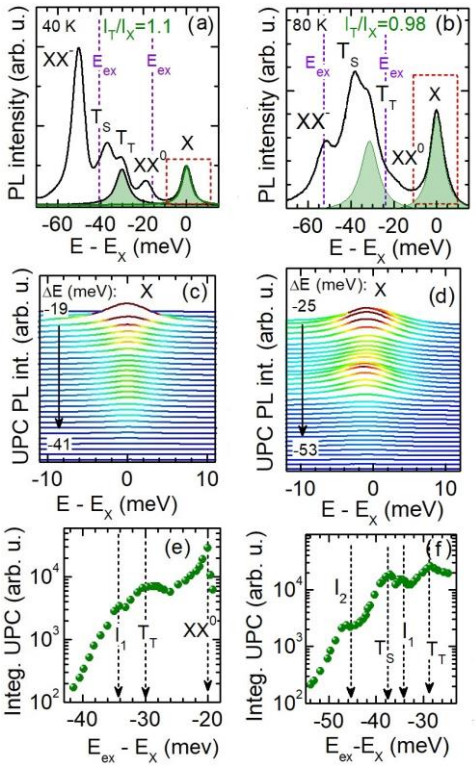

Fig. 1 (a-b) The PL spectrum of WSe_2 at 40 and 80 K, respectively. (c-d) The evolution of the UPC PL spectra as function of energy gain ($\Delta E=E_{ex}-E_X$). (e-f) Integrated UPC PL intensity of the exciton X as a function of ΔE.

Here, we demonstrate an observation of momentum- dark excitons in temperature- dependent upconversion photoluminescence (UPC PL) in hBN-encapsulated WSe_2 monolayers. The two momentum- dark excitons I_1 and I_2 show up in the excitonic UPC PL spectra as intensity enhancements 34 and 45 meV below the bright exciton (Fig.1a-f), correspondingly, which is in line with recent theoretical calculations [3]. We show that the observation of momentum- dark excitons in the upconversion emission process requires special conditions related to the electron doping as well as the temperature. Moreover, in the UPC PL spectra we observe prominent resonances at energies corresponding to the biexciton and both the spin- singlet and-triplet trions (Fig. 1e-f). The revealing of the fine trion structure in the UPC PL sheds a new light on previous studies of monolayer WSe_2, where only the triplet trion- exciton coupling was observed. For comparison we perform similar experiments in hBN- encapsulated monolayer $MoSe_2$, where the dark exciton band lying at higher energy than the bright band. By increasing the temperature from 7 to 120 K we probe phonon- mediated exciton- trion interaction as the normal PL intensity of the singlet trion gradually decreases.

[1] J. Jadczak et al., *Nature Communications* **10**, 107 (2019).
[2] J. Jadczak et al., *Materials* **14**, 399 (2021).
[3] S. Brem et al., *Nano Lett.* **20**, 4 (2020).