Comparison of the excitonic upconversion photoluminescence in $MoSe_2$ monolayers with a different doping level

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Transition metal dichalcogenide monolayers represent unique platforms for studying both electronic and phononic interactions as well as intra- and intervalley exciton complexes [1]. The interaction between excitonic complexes in TMDC monolayers may be alternatively probed in upconversion (UPC) PL photoluminescence (PL) experiments. The excess energy required for the UPC process may be taken from phonons or resident electrons in the monolayer. Hence, the UPC PL provides information on both the energy spectra of the TMDCs as well as the scattering mechanism related to exciton–exciton, exciton–electron, and exciton–phonon interactions.

Here, we compare the UPC PL in an optically bright $MoSe_2$ monolayer systems revealing different doping level. In hBN-encapsulated $MoSe_2$ monolayers with a relatively weak electron concentration the UPC PL excitation reveals two pronounced resonances below the neutral 1s A-exciton (X). The resonance detected at an energy of about 25 meV below X coincides with the PL peak and binding energy of the singlet trion T_S. The second resonance at −18 meV with respect to the exciton transition is attributed to the neutral biexciton (XX^0). The mechanism of the exciton PL upconverted by the neutral biexciton is attributed to the interaction of the photocreated electrons at the K+ and K– valleys with zone-corner flexural acoustic ZA(K) phonons. The UPC of the X PL via the spin-singlet negative trion is assigned to a spin- and valley-conserving scattering process of the photocreated electron with the optical A_1 phonon mode whose energy at the K valleys matches the energy difference between the singlet trion and the neutral exciton [1].

Interestingly, in uncapped $MoSe_2$/hBN structures with the higher electron density the exciton UPC PL is hardly detected, whereas in hBN/$MoSe_2$/graphene/hBN heterostructures, where the doping by resident charge carriers is completely neutralized and results in a vanishing trion emission, the UPC energy gain is about 17 meV and resonates with the energy of the zone-corner flexural acoustic ZA(K) phonon mode.

Our results extend the current discussion about interactions of electrons with both optical and acoustic phonons at the K valleys and their role in the upconversion of exciton emission in $MoSe_2$ monolayers. We also provide further insights into resonant exciton–trion and exciton–biexciton couplings for optically exciting a 2D material within its nominal transparency range.

[1] J. Jadczak, J. Debus, J. Olejnik et al., *J. Phys. Chem. Lett.* **14**, (39) 8702-8708 (2023).