Dexter induced inverted valley polarization in monolayer WSe_2

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A highly efficient, but not particularly explored, intervalley scattering mechanism in transition metal dichalogenides resembles a Dexter coupling in the reciprocal space. This mechanism couples exciton states with the same spin in different valleys [1].

Here, we demonstrate that the Dexter-type intervalley scattering affects not only the neutral exciton [1], but also other excitonic complexes of a monolayer WSe_2 . While the photoluminescence (PL) spectrum is co-polarized with the excitation laser when this is tuned far-off resonance with either A or B exciton, its degree of circular polarization (DCP) becomes negative when the laser light is tuned resonant with B exciton, as shown in Fig. 1(a). This vanishing or negative polarization of the excitation and biexciton complexes is confirmed by the polarization-resolved PL excitation (PLE) of Fig. 1(b,c,d). Pump-probe measurements shown in Fig. 1(e,f) demonstrate that immediately after an excitation pulse resonant with A exciton is absorbed, the DCP is negative, thus supporting the Dexter coupling as main mechanism driving the exciton dynamics in this condition.



Figure 1: (a) PL spectra of encapsulated monolayer WSe₂ with detection of the light co-polarised $(\sigma^+\sigma^+)$ and cross-polarised $(\sigma^+\sigma^-)$ to excitation. Excitation energy between A_{2s} and B_{1s} in top panel and resonant with B_{1s} exciton in the bottom panel. (b,c,d) Excitation energy dependent PL intensity of the co- and cross-polarised emission and calculated degree of circular polarisation (DCP) for A_{1s}, XX and XX⁻, respectively. (e) Photoinduced absorption of the B_{1s} exciton and (f) degree of circular polarization for an excitation resonant to A_{1s} exciton.

[1] G. Berghäuser, et al., Nature Communications 9, 971 (2018).