

Spin-Selective Exciton–Exciton Scattering as a Valley Depolarization Channel in Monolayer WSe₂

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Abstract. We develop a microscopic first-quantization theory of exciton–exciton interaction (EEI) in monolayer transition metal dichalcogenides that explicitly resolves the full spin–valley–band fine structure of the initial and final two-exciton states. Retaining the long- and short-range parts of the electron–hole exchange on equal footing with the direct and carrier-exchange Coulomb channels, we derive a compact classification of all symmetry-allowed scattering processes for each optical excitation condition (σ^\pm , linear, elliptical) and identify those that act as valley-pseudospin flips. A group-theoretical reduction under $D_{3h} \otimes T$ collapses the twelve kinematically open channels into five symmetry-independent classes and proves that the dominant single valley-flip amplitude carries a chirality-2 angular form factor enforced by C_3 invariance alone, tying it as a symmetry-sibling to the Maialle–Silva–Sham (MSS) long-range exchange field. Applied to monolayer WSe₂, the theory predicts that EEI-mediated intervalley transfer is dominated by the carrier-exchange channels; within these, the hole-exchange contribution outweighs electron-exchange, owing to the larger valence-band spin–orbit coupling of W-based TMDs, and the total EEI rate becomes comparable to the MSS precession rate for exciton densities $n_X \gtrsim 3 \times 10^{11} \text{ cm}^{-2}$. We compare this rate with excitation-induced dephasing, phonon-assisted intervalley scattering to the KA dark reservoir, exciton–impurity scattering, and the closely related Förster and Dexter energy-transfer mechanisms. The comparison resolves a long-standing ambiguity in the interpretation of density dependent valley-polarization measurements and identifies a regime in which EEI, rather than MSS or phonons, controls the optical valley coherence.

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