

Exciton valley depolarization in monolayer MoS₂: non-Markovian quantum dynamics, intervalley scattering, and the breakdown of the Dyakonov-Perel mechanism

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In monolayer transition metal dichalcogenides, exciton valley relaxation is typically attributed to the Dyakonov-Perel (DP) mechanism, where frequent intravalley scattering with phonons or defects suppresses the intervalley exchange-induced precession and the role of intervalley scattering is considered secondary. Employing a first-principles nonequilibrium exciton Green's function (NEGF) approach with the generalized Kadanoff-Baym ansatz (GKBA), which treats exchange-driven precession and exciton-phonon scattering on an equal footing across the full Brillouin zone, we demonstrate that even in the small momentum regime most favorable to DP physics, realistic exciton-phonon scattering is too weak to induce the motional narrowing that defines the DP regime.

Upon accounting for excitons across the entire Brillouin zone, large momentum intervalley scattering becomes the dominant pathway for valley relaxation, shortening the depolarization by a factor of 3-4 relative to intravalley-only models. We find that valley depolarization and decoherence time are approximately 50 fs at 300 K and lengthen to 130 fs at 10 K. By comparing our results with a Lindblad-type collision framework [1], we explicitly demonstrate the role of non-Markovian effects and reveal that the Markovian Lindblad framework is highly basis dependent. In the valley-pseudospin basis underlying prior analyses, the Markovian Lindblad approach artificially amplifies scattering-induced equilibration, biasing the dynamics toward a DP interpretation. Our study provides a comprehensive picture of valley relaxation, and establishes the exciton density matrix approach derived from NEGF+GKBA as a powerful tool for investigating ultrafast exciton dynamics [2].

[1] J. Xu, A. Habib, R. Sundararamn, Y. Ping, Phys. Rev. B 104, 184418 (2021)

[2] Y.H. Chan, Jonah B. haber, Mit H. Naik, Felipe H. da Jornada, Diana Y. Qiu, arXiv:2607.08479 (2026).