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Role of interlayer coupling, phonons, and intervalley scattering in marginally twisted WSe_2 homobilayer

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Abstract

Excitons serve as a unique optical probe in moiré materials due to their sensitivity to both valley-dependent electronic structure and many-body excitonic interactions.[1] In angle-tuned twisted bilayers, lattice reconstruction becomes significant at marginal twist angles.[2] Here, we investigate the steady-state and ultrafast transient exciton behaviors in a twisted WSe_2 homobilayer (t-WSe_2) using reflection contrast (RC), photoluminescence (PL) and ultrafast pump-probe spectroscopy. We report the emergence of two distinct intralayer excitons exhibiting different doping-dependent valley coherence and population dynamics, arising from asymmetric interlayer coupling. This asymmetric interlayer coupling leads to a more rapid decay of X_1 compared to the decay of X_2. We employed a modified transfer matrix method (TMM) [3] based on the Lindblad master equation to analyze the population decay times and the pure dephasing rates. Notably, the dephasing rate of X_1 **is observed to increase in hole-doping, consistent with polarization-resolved PL. Through the modified TMM analysis, we attribute the fast-decaying component (**τ_1=0.5-1 ps**)** observed in pump-probe spectroscopy to radiative recombination within the excited optical light cone, combined with repopulation effects. The slower decay component (τ_2=100-300 ps) is linked to the exciton-phonon scattering and the population relaxation mediated by interlayer breathing phonons. **Our findings provide detailed insights into the exciton population dynamics in twisted homobilayers, emphasizing the roles of asymmetric coupling, intervalley interactions, and exciton-phonon coupling in transients of exciton complexes.**