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Interlayer exciton thermalization probed by moiré-split intralayer states in WSe_2/WS_2 heterobilayers

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Abstract

Moiré interlayer excitons (IXs) in transition metal dichalcogenides heterostructures have recently attracted significant attention due to their unique optical properties and potential for novel optoelectronic applications [1]. Strong interlayer coupling enables the observation of IXs primarily via photoluminescence. While several studies have attempted to probe IXs absorption [2] and dynamics via transient spectroscopy [3], investigations into their thermalization processes have remained limited due to weak absorption signals and the insufficient temporal resolution of time-resolved photoluminescence techniques.

Here, we investigate the thermalization dynamics of IXs by probing the moiré-induced splitting of intralayer exciton states with distinct spatial distribution in WSe_2/WS_2 heterobilayers [Figs. 1(a) and 1(b)] [4]. Using ultrafast transient reflectance spectroscopy, we monitor IX thermalization by deconvoluting spectral parameters such as peak energy, linewidth, and spectral weight. We find that IX thermalization proceeds on a ~10 ps timescale [Fig. 1(c)], dominated by phonon-exciton scattering under low excitation densities below the moiré density. At higher excitation densities, carrier-carrier interactions provide an additional scattering channels, accelerating the thermalization process [Fig. 1(d)]. Our findings reveal insights into the interplay between phonons, carriers, and moiré potentials in IX thermalization dynamics, and introduces a new approach for pump–probe spectroscopy based on spatial exciton distributions, extending beyond the conventional momentum-resolved band framework.