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Non-linear and non-equilibrium exciton diffusion in 2D perovskites

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Abstract

Two-dimensional hybrid perovskites exhibit strong excitonic effects due to quantum confinement and reduced dielectric screening. We study exciton transport in $(MBA)_2PbI_4$ using transient photoluminescence microscopy, comparing enantiomer-pure and racemic mixture samples. At room temperature, we observe initial efficient exciton propagation over > 100 nm, followed by localization of carriers at later times, with diffusion coefficients up to 1.5 cm^2/s. Enhanced exciton mobility in enantiomer-pure samples is linked to reduced disorder, as confirmed by photothermal deflection spectroscopy. We find that early-time diffusion strongly depends on excitation density, due to gradual filling of shallow trap states within the disordered energy landscape. At low temperatures, samples of the achiral 2D perovskite $(PEA)_2PbI_4$ show unusually high transient diffusion coefficients of up to several tens of cm^2/s, even under resonant excitation. In a joint theory-experiment study we show how the presence of dark excitonic states leads to the observed non-equilibrium dynamics and extremely rapid exciton propagation. These findings combined highlight the intriguing and rich physics of 2D perovskites with mobile excitonic quasiparticles serving as primary energy carriers.