Power-driven sign reversal of interlayer exciton valley polarization in a WS_2/WSe_2 moiré heterobilayer under sub-gap excitation

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Properties of heterostructures made of monolayers of transition metal dichalcogenides (TMDCs) make them a rich platform for exploring correlated many-body phenomena in semiconductor physics. Moiré superlattices in TMDC heterobilayers host interlayer excitons whose valley degree of freedom is governed by the quasi-angular momentum of the trapping site, yet conventional above-gap excitation accesses these states only indirectly through an intralayer cascade that obscures the intrinsic selection rules. We use circularly polarized sub-gap excitation, 50–230 meV below the WSe_2 A-exciton resonance, to create interlayer excitons directly in an R-type WS_2/WSe_2 moiré heterobilayer at 7 K. Power-driven sign reversal of the degree of circular polarization of the IX_1 ground state is observed, from −9% to +12%, absent under above-gap conditions. A spectral threshold at 1.577 eV separates two regimes with opposite valley injection (Fig.1): magneto-photoluminescence confirms the same emitting state in both regimes, but reversed valley population imbalances, which provide direct evidence for coexisting moiré miniband states with co-circular and cross-circular selection rules. A rate-equation model quantitatively reproduces the sign reversal through nearly symmetric injection, exchange depolarization, chiral-phonon intervalley scattering, and biexciton-mediated valley purification. A Lower-energy interlayer exciton feature exhibits the same polarization sign as the ground state but with a photoluminescence intensity that scales as the square of the ground state intensity and an amplified valley asymmetry at high density, consistent with a collision-induced phonon sideband; G_0W_0+BSE calculations identify momentum-dark states at similar energies as a possible subdominant contribution. Sub-gap excitation spectroscopy thus opens a direct spectroscopic window into the excitonic miniband structure and valley-contrasting selection rules of moiré superlattices.

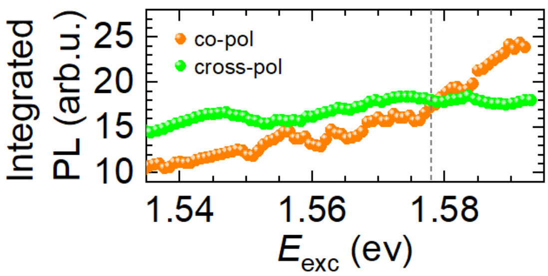

Fig. 1 Integrated IX_1 PL intensity in co- and cross-polarized configurations as a function of E_{exc}.