The Impact of Dark Excitons on Polariton Spatiotemporal Dynamics in 2D Semiconductors

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The rich exciton landscape of transition metal dichalcogenides (TMDs) provides intervalley scattering pathways via momentum-indirect excitons [1]. These dark states have been extensively studied in bare TMD monolayers, where they facilitate rapid energy relaxation and have recently been demonstrated to play a key role in polariton absorption [2] and photoluminescence (PL) [3]. However, the relaxation dynamics of TMDs in the strong coupling regime is not as well understood. In particular, understanding the role of the polariton bottleneck effect is important, as it impedes phonon-driven scattering into low-momentum states, hindering the development of room-temperature polaritonic devices and the investigation of polariton condensates. Using a microscopic Wannier-Hopfield approach [4], supported by detuning-dependent cryogenic PL measurements, we have investigated the momentum- and time-resolved relaxation of exciton polaritons in an MoSe2 monolayer integrated within a microcavity [5]. For a suitable cavity detuning, momentum-dark excitons provide an efficient reservoir to rapidly populate the polariton ground state via phonon-assisted scattering, thereby circumventing the bottleneck effect. We also predict striking evidence of these intervalley pathways through the presence of angle-resolved phonon sidebands in low-temperature PL spectra. Furthermore, we report recent work exploring exciton polariton transport at elevated temperatures, in particular, the crossover from the ballistic to diffusive regime due to efficient scattering with phonons. By solving the polaritonic Boltzmann equations, taking into account the full spatiotemporal dynamics of the exciton reservoir, we investigate the expansion of the entire exciton cloud and how the rapid in-plane propagation of polaritons impacts the thermalization of the system.

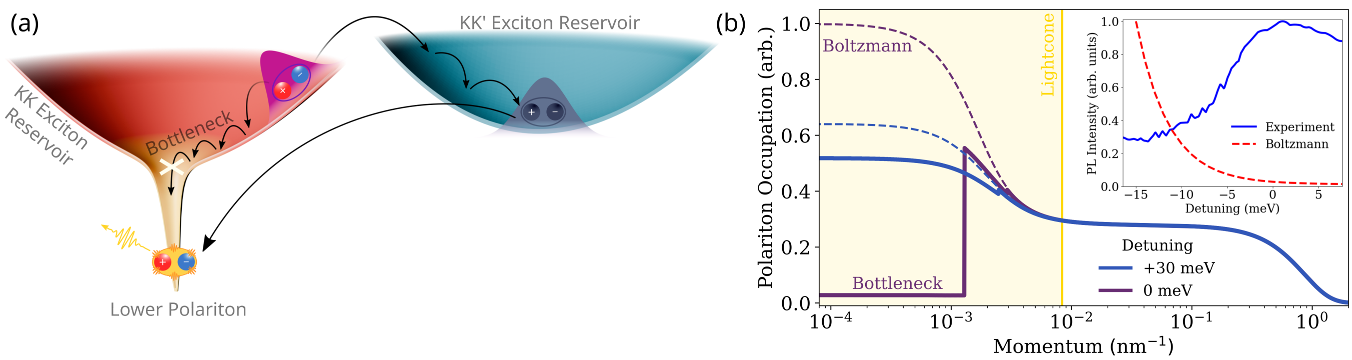

Figure 1: (a) Schematic of possible relaxation processes for the lower branch of a molybdenum-based exciton polariton. (b) Detuning dependence of the lower polariton occupation at room temperature, revealing a bottleneck at low momenta for detunings where intervalley scattering from the KK' valley is not possible. The inset shows experimental PL intensity at cryogenic temperatures along the lower polariton dispersion (blue curve). A vastly different detuning dependence is found compared to the PL emission from a thermalized Boltzmann population (dashed red curve), revealing a bottleneck.

References

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