

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

Jamie Fitzgerald¹, Roberto Rosati¹, Hangyong Shan², Christian Schneider², Ermin Malic¹

¹Philipps-Universität, Marburg, Germany. ²Carl von Ossietzky Universität Oldenburg, Oldenburg, Germany

Abstract

The rich exciton landscape of transition metal dichalcogenides (TMDs) provides intervalley scattering pathways via momentum-indirect excitons. These dark states facilitate rapid energy relaxation in bare TMD monolayers, and have recently been demonstrated to play a key role in polariton absorption [1] and PL [2]. 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 [3], 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 [4]. 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.