Exciton optics and dynamics in organic and organic/TMD heterostructures

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Organic semiconductors are exciting candidates for photovoltaic and biosensor technologies. They are cheap, flexible, relatively easy to fabricate and posses excellent light-matter coupling, crucial for a host of application. Currently however there are limitations preventing these materials reaching the level of efficiency of competing devices. These materials possess low mobilities and cannot be easily mechanically modified by strain. These limitations can be overcome by interfacing organic crystals with transition metal dichalcogenides (TMDs). In this joint theory-experiment work, we study the exciton landscape in organic molecular crystals as well as organic/TMD heterostructures, with a particular emphasis on their optical response. We demonstrate both theoretically and experimentally that the low-temperature photoluminescence (PL) is dominated by the formation of interlayer excitons, with the electron and hole located on the TMD and molecule layer, respectively [1]. We find that additional sidebands emerge as a result of the phonon-mediated indirect recombination of excitons. Furthermore, we study the exciton landscape within an organic crystal layer focusing on tetracene and pentacene crystals, We find a unique polarisation- and temperature-dependence of absorption and PL spectra, stemming from the Davydov-split excitons [2]. We describe the exciton dynamics in these crystals, where the flatness of the exciton bands gives rise to phonon-bottlenecks in the exciton relaxation, where momentum-indirect excitons offer crucial relaxation channels. We then turn our attention to energy transfer processes in organic/TMD heterostructures, which is governed by the Förster interaction, and explore the resulting signatures in the differential absorption and PL spectra. Our joint theoretical-experimental work unveils the behaviour of excitons in organic TMD heterostructures, and sheds light on the optical and dynamical response of these materials, which is imperative for understanding and designing future device architectures.

References

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Figures

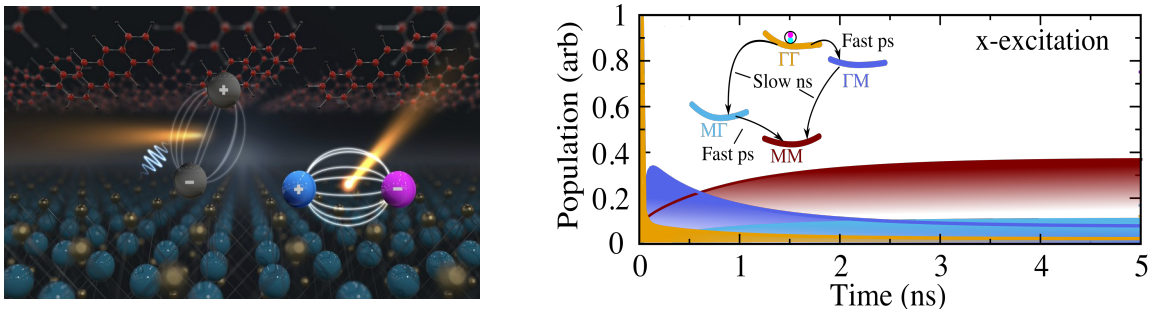

Figure 1: (Left) Artist's illustration of inter- & intralayer exciton landscape in an organic/TMD heterostructure. (Right) Exciton relaxation dynamics in an organic semiconductor.