Ultrafast carrier and spin dynamics of two-dimensional semiconductors

Transition Metal Dichalcogenides (TMDs) are emerging materials for electronic, spintronic and quantum information processing applications. When thinned to a single layer (1L), they become direct bandgap two-dimensional semiconductors, with optical response dominated by tightly bound (up to 0.5 eV binding energy) excitons, due to the strong quantum confinement and the reduced Coulomb screening.

In addition, the large spin-orbit coupling lifts the spin degeneracy of the valence and the conduction bands; due to the lack of inversion symmetry, spin and valley indexes are locked and valley-polarized carriers can be excited by circularly polarized light. Finally, layers of different TMDs can be easily integrated vertically by mechanical stacking, forming van der Waals heterostructures with rich physics.

In this talk I will present results on the non-equilibrium optical response of 1L TMDs investigated by a variety of ultrafast optical spectroscopy techniques. We first study exciton dynamics of 1L-MoS₂, by broadband femtosecond transient absorption, showing that the non-equilibrium optical response is dominated by the renormalisation of both band gap and exciton binding energies caused by photo-excited charge carriers.

We then use two-colour helicity-resolved pump-probe spectroscopy in order to disentangle the intervalley and intravalley spin-flip processes of electrons in the conduction band of 1L-WS₂, time resolving the formation of the dark excitons which are responsible for photoluminescence quenching in this material.

Finally, we study heterostructures of WSe₂ and MoSe₂ and time resolve the sub-picosecond build-up time of the interlayer exciton formed upon injection of a hole from MoSe₂ to WSe₂, as well as its temperature-dependent lifetime.