Ultrafast charge transfer in heterostructures of two-dimensional materials

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Heterostructures (HS) of two-dimensional materials offer unlimited possibilities to design new materials for applications to optoelectronics and photonics. In such HS the electronic structure of the individual layers is well retained because of the weak interlayer van der Waals coupling. Nevertheless, new physical properties and functionalities arise beyond those of their constituent blocks, depending on the type and the stacking sequence of layers. In this presentation we use high time resolution ultrafast transient absorption (TA) and two-dimensional (2D) spectroscopy to resolve the interlayer charge scattering processes in HS.

We first study a WSe_2/$MoSe_2$ HS, which displays type II band alignment with a staggered gap, where the valence band maximum and the conduction band minimum are in the same layer. By two-colour pump-probe spectroscopy, we selectively photogenerate intralayer excitons in $MoSe_2$ and observe hole injection in WSe_2 on the sub-picosecond timescale, leading to the formation of interlayer excitons. The temperature dependence of the build-up and decay of interlayer excitons provide insights into the layer coupling mechanisms. We also employ 2D spectroscopy, which guarantees simultaneously high temporal and spectral resolution, to time resolve the sub-50-fs interlayer exciton formation in a MoS_2/WS_2 HS.

Finally, we investigate a graphene/WS_2 HS where, for excitation well below the bandgap of WS_2, we observe the characteristic signal of the A and B excitons of WS_2, indicating ultrafast charge transfer from graphene to the semiconductor. The nonlinear excitation fluence dependence of the TA signal reveals that the underlying mechanism is hot electron/hole transfer, whereby a tail the hot Fermi-Dirac carrier distribution in graphene tunnels through the Schottky barrier. Hot electron transfer is promising for the development of broadband and efficient low-dimensional photodetectors.