

Ultrafast carrier and spin dynamics in two-dimensional semiconductors

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In layered semiconductors, such as transition metal dichalcogenides (TMD), the electron-electron interaction is strongly enhanced by both quantum confinement and reduced screening [1]. Furthermore, in these materials the valley polarization can be optically controlled by means of circularly polarized light [2]. Here we discuss the ultrafast optical response of TMDs, focusing on two aspects: time-resolved measurements of exciton dynamics and spin/valley relaxation processes. We first study exciton dynamics in single-layer (1L) MoS₂ by broadband femtosecond transient absorption spectroscopy combined with time-resolved *ab-initio* simulations [3] based on the non-equilibrium Green's functions and density-functional theory. This comparison indicates that the non-equilibrium optical properties of TMDs are influenced by the renormalisation of both band gap and exciton binding energies caused by photo-excited charge carriers. We further use sub-20-fs pump pulses tuned above the electronic gap to study the exciton formation process in 1L-MoS₂, and determine a build-up time of ≈ 35 fs. This is consistent with a scenario where free carriers, initially excited above the quasi-particle gap, relax towards lower energy states and finally form the exciton state via the strong Coulomb interaction.

The exciton valley relaxation dynamics is investigated by time-resolved Faraday rotation [4]. We observe a double exponential decay, with an initial fast (~ 200 fs) decay due to scattering of spin-polarized excitons from K to K'. This is in good agreement with the time scale predicted by the Maialle-Silva-Sham electron/hole exchange interaction mechanism, which can be interpreted as a virtual annihilation of a bright exciton in one valley followed by the creation of an exciton in the opposite valley. Finally, we 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. Spin-polarized carriers are injected by a circularly polarized pump pulse resonant with the A exciton, while the co-circularly polarized probe pulse is tuned around the B excitonic peak. In this configuration, the scattering of the electrons from the upper to the lower conduction band level (where they cannot radiatively recombine) is detected by measuring the build-up dynamics of the bleaching signal around the B exciton caused by Pauli blocking. We also show that spin-conserving intervalley scattering dynamics occur on a faster time-scale than the intraband spin-flip process.

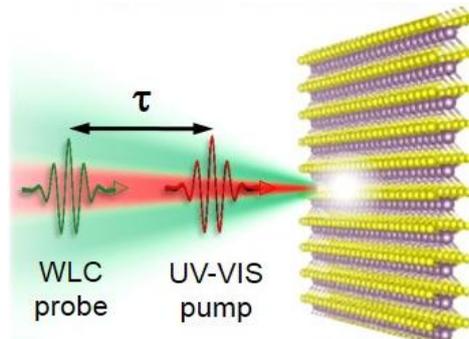


Figure 1. broadband ultrafast optical spectroscopy of single-layer TMDs.

References

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