Non equilibrium optical properties of monolayer MoS_2 probed by ultrafast spectroscopy

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Abstract

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 circular polarized light [2]. We investigate single-layer MoS_2 (1L-MoS_2) with ultrafast transient absorption spectroscopy (Fig.1a) and time resolved ab-initio simulations [3] based on the non-equilibrium Green's functions and density-functional theory [4]. 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. The exciton valley relaxation dynamics [5] is investigated by time-resolved Faraday rotation. A circularly polarized pump pulse creates a spin and valley polarized population in the conduction/valence band, which causes the rotation of the linear polarization of a delayed probe pulse. The probe pulse energy is tuned below the absorption gap to be sensitive only to the helicity-dependent light scattering of the photoexcited electrons and holes. Since probe photons couple to the charge carriers orbital momentum, which in 1L-MoS_2 is locked to the valley index, the rotation angle θ_F probes the intervalley relaxation processes. We observe a double exponential decay (Fig.1b), with an initial fast (~200fs) 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.

References

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Figures

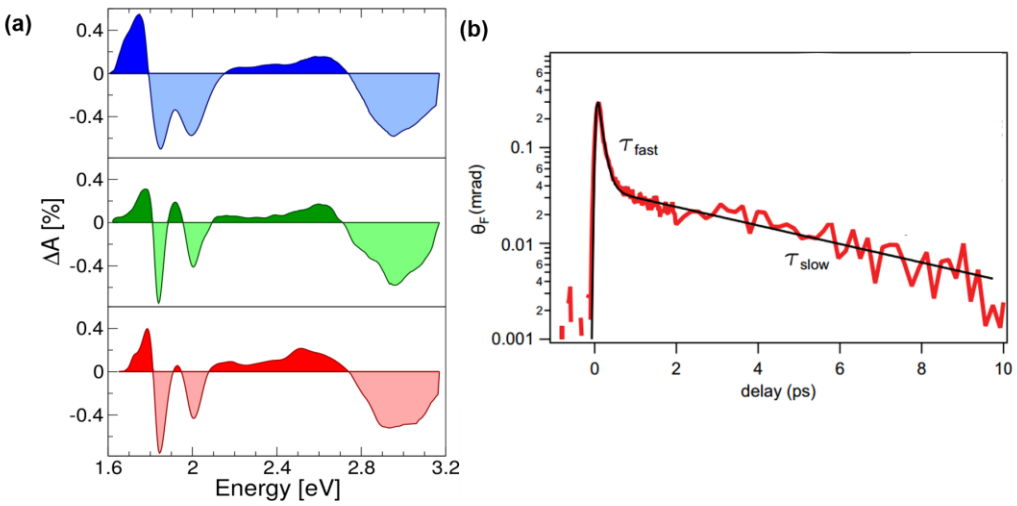

Figure 1. Non equilibrium optical properties of 1L-MoS_2. (a) Transient absorption spectra (ΔA) at different excitation energies (ω_{pump}); (b) Time-resolved Faraday rotation (θ_F)