Bandgap renormalization governs the ultrafast response of MoS_2

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Two-dimensional crystals of transition metal dichalcogenides (TMDs) are attracting growing interest due to their unique electronic and optical properties, resulting in the demonstration of functional devices. On the more fundamental side, they represent ideal systems for exploring the physics of 2D semiconductors, where the electron-electron interaction is strongly enhanced by both quantum confinement and the reduced screening. Here we present a time-resolved study of charge carrier dynamics in single-layer MoS2 (1L-MoS2) that combines ultrafast transient absorption spectroscopy and ab-initio simulations.

Using tunable pump pulses and broadband probing, the relaxation dynamics of the photo-excited states is monitored with unprecedented spectral coverage (the entire visible range). The sample is a 10x30 μm^2 1L-MoS2 prepared by micromechanical exfoliation and transferred onto a transparent fused silica substrate [1]. The transient absorption spectrum has three prominent features, each consisting of a bleaching at the energies of the excitonic transitions A, B and C (at 1.9, 2.1 and 2.9 eV), and a red-shifted photoinduced absorption, see Figure 1. These features do not depend on the excitation energy, which is tuned to be resonant and non-resonant with the excitonic transitions.

Comparison with ab-intio simulations based on non-equilibrium Green's functions [2,3] allows us to shed light on the delicate interplay among the key phenomena governing the carrier dynamics after photo-excitation. In particular, a transient band gap renormalization caused by the presence of photo-excited carriers is found to give rise to the simultaneous bleaching of the three excitonic transitions and the corresponding photoinduced absorption bands, which cannot be explained by Pauli blocking alone [4].

References

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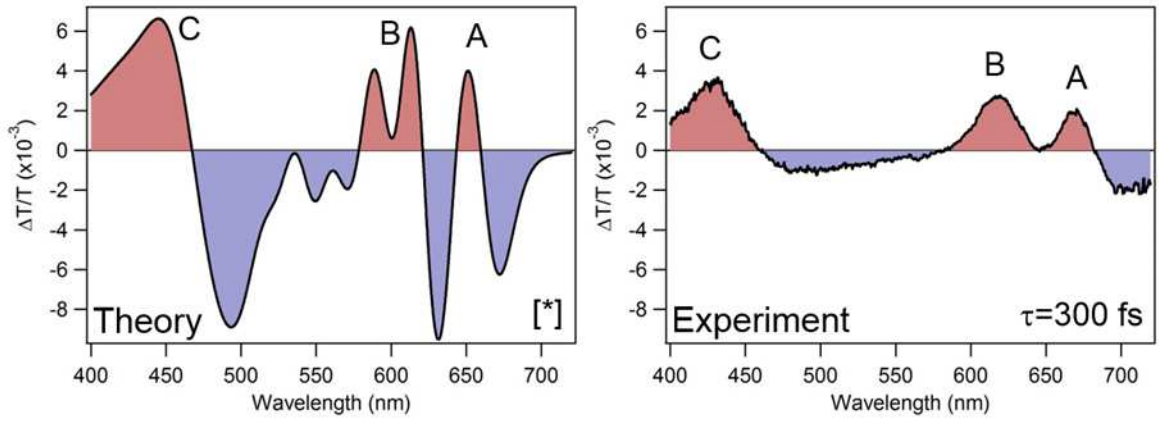

Figure 1: Simulated (left panel) and experimental (right panel) transient transmittivity of 1L-MoS2, following fs-pulse excitation with λ_{pump} = 400 nm at fixed time delay t = 300 fs, demonstrating the simultaneous bleaching of the exciton transitions A, B and C (red), and the photoinduced absorptions (blue).