Ultrafast broadband study of photocarrier dynamics in MoS_2 single layer

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We present a time-resolved study of charge carrier dynamics in single-layer MoS_2 (1L-MoS_2) by ultrafast transient absorption spectroscopy. Using tunable pump pulses and broadband probing, we monitor the relaxation dynamics of the photo-excited states with unprecedented spectral coverage (the entire visible range). The sample is a 10 x 30 μm^2 1L-MoS_2 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 C (at 1.9, 2.1 and 2.9 eV) and a red-shifted photoinduced absorption, Fig. 1. These features do not depend on the excitation energy, which is tuned to be resonant and non-resonant with the excitonic transitions. Pauli blocking cannot explain, alone, the simultaneous bleaching of the three excitonic transitions and the corresponding photoinduced absorption. Instead, we believe that a transient band gap renormalization caused by the presence of photo-excited carriers should be also considered. A static strong renormalization of both electronic band gap and exciton binding energy was previously reported in $MoSe_2$ due to the interaction with the substrate [2]. Here we compare our data with simulations combining non-equilibrium Green's functions with *ab-initio* methods [3,4]. The comparison of experimental data with simulations allows us to shed light on the delicate interplay among Pauli blocking, band gap renormalization and electron-phonon relaxation, which are the key phenomena governing the carrier dynamics after photo-excitation.

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Figure.1 Transient Absorption of 1L-MoS_2. 100 fs-pulse excitation in the visible range determines simultaneous bleaching of the A, B, C excitons, with the absorption surviving up to hundreds ps. a) Transient absorption map with λ_{pump}= 400nm; b) Transient absorption spectrum at fixed delays; c) Relaxation dynamics of C-exciton bleaching with λ_{pump}= 400, 600 and 650 nm.