Temporal dynamics of the coulomb screening in single layer MoS_2

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Abstract. We measure exciton dynamics in single-layer MoS_2 with unprecedented temporal resolution and we directly extract the characteristic time-scale for the Coulomb screening dynamics, which ranges between 15 and 35 fs.

1 Introduction

The optical response of single-layer (1L) Transition Metal Dichalcogenides (TMDs) in the visible range is strongly renormalized by excitonic effects caused by the quantum confinement in two dimensions [1]. The reduced screening of the Coulomb interaction between electrons and holes leads to a dramatic increase of the exciton binding energy, which is estimated to be hundreds of meV. For this reason, excitons in 1L-TMDs can be highly stable even at room temperature. A direct consequence of this effect is that the optical and the electronic gap, whose difference is set by the binding energy of the exciton, are well separated in energy. This peculiar property offers the interesting possibility to study the optical response of a semiconductor when carriers or excitons are selectively photoexcited in the system by tuning the pump photon energy respectively above or below the electronic gap.

In the last years, the non-equilibrium optical response of TMDs has been extensively investigated. However, most of the works focused the attention on the study of the relaxation channels that determine the decay dynamics of TMDs. On the other side, only few experimental works have been dedicated to the study of the exciton formation process. Moreover the typical temporal resolution of these experiments (i.e. ~ 100 fs) has not allowed to clearly resolve physical processes, like Coulomb screening dynamics, which is the fingerprint of transient bandgap renormalization [6] and exciton formation dynamics [2,3]. Such processes are supposed to occur on much a faster timescale [4].

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In the present work, we overcome this issue and we study the non-equilibrium optical response of 1L-MoS_2 by performing optical pump-probe experiments with an unprecedented temporal resolution: both pump and probe are few-cycle optical pulses, resulting in a temporal duration lower than 20 fs.
The sample under study is a 1L-MoS_2 sample grown by chemical vapour deposition on a SiO_2/Si substrate. The left panel of Fig. 1 shows a sketch of the band structure of 1L-MoS_2 in the K point of the hexagonal Brillouin zone. A and B optical transitions are strongly renormalized by excitonic effects and are centered at 1.9 eV and 2.05 eV, respectively. The energy difference between A and B excitons is attributed to the splitting of the valence band, induced by spin-orbit interaction.

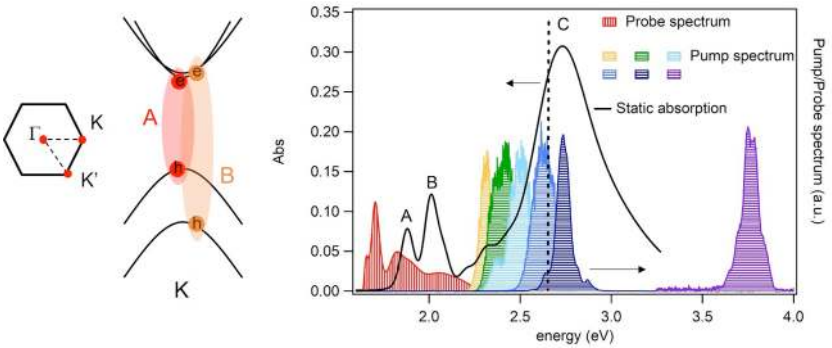

Fig. 1. (Left panel) Sketch of the band structure of 1L-MoS_2 in the K point of the hexagonal Brillouin zone. (Right panel) The black curve is the static absorption spectrum of 1L-MoS_2 measured at room temperature. On the same graph, the spectra of pump and probe pulses are displayed, together with the estimated energy of the electronic gap (dashed line).

As shown in the right panel of Fig. 1, the probe pulse has a broad spectrum covering both the A and B excitonic peaks, while the pump photon energy is progressively tuned from below to above the electronic gap (dashed line), whose energy has been previously estimated to be around 2.7 eV [5]. The temporal resolution for each pump-probe experiment has been measured by the cross frequency-resolved optical gating method and for each pump pulse it is estimated to be below 25 fs. In the case we excite the system below the electronic gap we mainly photo-inject excitons, while if we pump above the gap free carriers are generated.

2 Results and Discussion

The transient reflectivity ($\Delta R/R$) maps measured at different pump photon energies exhibit qualitatively the same features, i.e. a positive variation of the signal in correspondence of A and B absorption peaks (see the map in Fig. 2a). These transient features are the result of two effects: phase space filling, due to the Pauli blocking of the probe absorption, and a red-shift of the optical band gap, due to the transient variation of the Coulomb screening induced by the pump excitation, namely the bandgap renormalization [6].

Here we focus on the build-up dynamics of the $\Delta R/R$ signal of both A and B transitions. The build-up time τ_{rise} is estimated by fitting the dynamics with an exponential function convoluted with a gaussian peak, accounting for the instrumental response function of the experiment. As shown in panel b (A exciton) and c (B exciton) of Fig. 2, we find that when

the pump photon energy is lower than the energy of the electronic gap (2.7eV) the transient signal exhibits an almost instantaneous rise time τ_{rise}, which gradually slows down when the energy of the pump approaches the electronic gap. Surprisingly, for higher pump photon energies the build up time does not increase anymore.

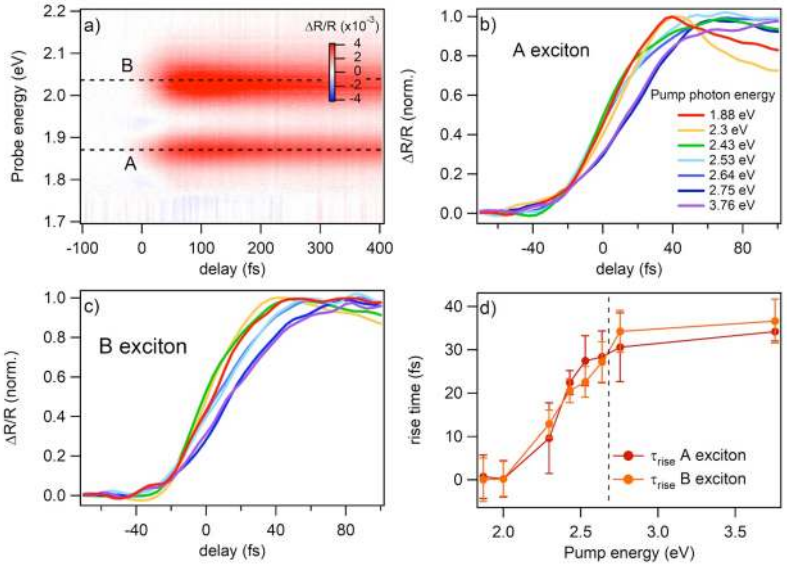

Fig. 2. (a) Transient reflectivity map of 1L-MoS_2 measured at room temperature. (b-c) Temporal traces cut of the ΔR/R map around the A/B excitonic peak measured at different pump photon energy. All the traces are normalized to unity. (d) Rise time of the traces vs photon energy.

Panel d of Fig. 2 reports the extracted rise time τ_{rise} of both A and B excitonic transitions as a function of the pumping photon energy. We can clearly distinguish between two different regimes. One is below the electronic gap, in which the rise time of A/B monotonically increases from less than 15 fs to 35 fs, as the pump photon energy is tuned from 1.8eV to 2.7eV. The second one is above the electronic gap, from 2.7eV to 3.7eV, in which the 35 fs build up does not increase anymore.

We argue that the observation of the finite rise time of the differential reflectivity signal 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 extracted characteristic time constant for the Coulomb screening ranges between 15 fs and 35 fs.

References

1. D. Qiu *et al.*, Phys. Rev. Lett. **111**, 216805 (2013).
2. P. Steinleitner *et al.*, Nano Letters, **17** (3), 1455 (2017).
3. F. Ceballos *et al.*, Nanoscale, **22** (8), 11681 (2016).
4. C. Robert *et al.*, Phys. Rev. B **93**, 205423 (2016).
5. K. Yao *et al.*, Phys. Rev. Lett., **119** (8), 087401 (2017).
6. E.A.A. Pogna *et al.*, ACS Nano, **10,** 1182 (2016).