Ultrafast valley-polarized charge transfer in a $WSe_2/MoSe_2$ heterostructure

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Abstract

Van der Waals heterostructures (HS), obtained by stacking two different monolayers on each other, have been intensively investigated because of their possible applications in electronics and optoelectronics [1]. Type II HS, where the valence band maximum and the conduction band minimum are in separate layers, allow efficient charge separation, which is particularly beneficial for photovoltaics based on atomically thin materials.
We use pump-probe spectroscopy to study the charge transfer dynamics in a type II $WSe_2/MoSe_2$ HS [2]. The pump beam is resonant with the smaller optical gap of the two monolayers (that of $MoSe_2$) and injects excitons in the K/K' valleys while the probe beam is resonant with the optical gap of WSe_2 to reveal any excited state population transfer from $MoSe_2$ to WSe_2. Excitation of an isolated $MoSe_2$ monolayer leads to a reduced absorption resonance (bleaching due to Pauli blocking) within the instrumental time resolution (100 fs). In the HS, on the other hand, we observe a clear delayed formation of the WSe_2 bleaching signal. We attribute this finite rise time to a fast interlayer hole transfer occurring in less than 1 ps, which can be clearly time-resolved in our experiment. The hole transfer yields an interlayer exciton (i.e. a bound state of an electron and a hole residing in two separate layers). Moreover, time resolved circular dichroism shows that the valley polarization is transferred from the excited layer to the adjacent one. The relaxation dynamics of the interlayer exciton is considerably slower (i.e. ~1 ns) compared to the excitons in isolated monolayers.

References

[1] A. K. Geim and I. V. Grigorieva, Nature 499 (2013), 419.
[2] Z. Wang et al. in preparation.

Figures

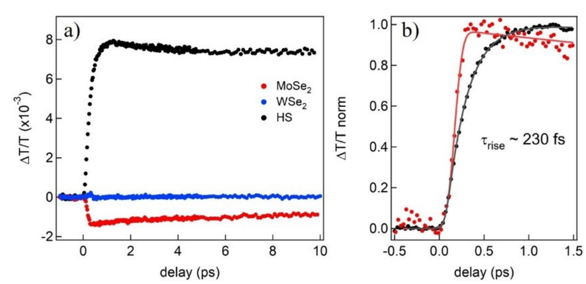

Figure 1: (a) Relative change ΔT/T of transmittance measured with pump and probe on resonance respectively with $MoSe_2$ and WSe_2 optical gap on $MoSe_2$ alone (red), WSe_2 alone (blue) and the $WSe_2/MoSe_2$ heterostructure (black). All the measurements are performed at T=9K. b) Normalized □ΔT/T traces at short delays in order to highlight the slower build up time of the HS signal compared to the single layer alone.