Ultrafast characterization of saturable absorption in monolayer $MoSe_2$

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Single and few-layer atomically thin materials have received significant attention since the isolation of graphene. Among Transition Metal Dichalcogenides (TMDs), monolayer molybdenum diselenide (ML-$MoSe_2$) is emerging as a promising direct bandgap semiconductor. Similarly to other low-dimensional materials, such as graphene[1] and carbon nanotubes [2], TMDs[3] exhibit strong saturable absorption (SA) properties, like intensity dependent absorption, together with picosecond (ps) recovery times. They are already widely employed to passively Q-switch or mode-lock lasers in a wide range from 800 nm to 3 μm, but all of them are still limited in the infrared (IR) wavelengths. Only TMD-based Q-Switching has recently reached visible wavelengths[4]. Here we use reflection-based nonlinear absorption microscopy and ultrafast degenerate pump-probe spectroscopy to investigate the SA properties of ML-$MoSe_2$, which has a bandgap in the VIS-near IR range. The extracted key SA parameters are the *spectrally dependent absorption* (720-810nm), the *saturation intensity* (~2.5MW/cm^2), the *modulation depth* (~80%), the *non-saturable losses* (~20%), and the time scale for absorption recovery after a saturating pulse, i.e. the *recovery time* (~300ps). Reflection-based nonlinear absorption microscopy shows a uniform intensity dependent change of the absorption across the region of the lowest energy exciton (~790nm) together with a constant modulation depth (see Fig. 1). Pump-probe spectroscopy reveals that ML-$MoSe_2$ fully recovers from absorption saturation within few hundred ps (see Fig. 2), indicating that this material can be efficiently applied as a slow saturable absorber (with recovery time well above the pulse duration).

References

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[3] Mohanraj J. et al., Optical Materials, 60 (2016) 601-617
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Figures

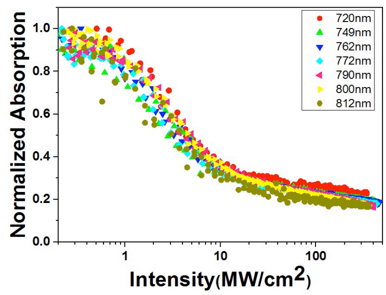

Figure 1: Pump intensity dependent normalized absorption of ML-$MoSe_2$. Overlapping curves at different excitation wavelengths reveal the same characteristic modulation depth.

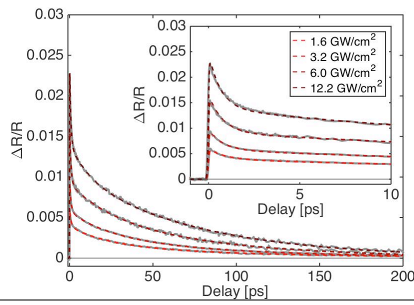

Figure 2: Degenerate pump-probe dynamics at different pump intensities (solid grey lines), together with best fit tri-exponential decay functions (dashed lines). In the inset dynamics in the first 10 ps delay. Pump and probe pulses at 760 nm.