Ultrafast Optical Pump-Probe Studies of Photoexcited Carrier Dynamics in Mono-Layer MoS2

HAINING WANG, CHANGJIAN ZHANG, FARHAN RANA, Cornell University — Mono-layer MoS2 is a 2D semiconductor with a direct bandgap. We present, for the first time, ultrafast optical pump-probe measurements results on the relaxation dynamics of photoexcited carriers in mono-layer MoS2. Pulses at energies 2.74 eV and 1.37 eV with width 100 fs are used in our experiments. The pump photon energy is larger than the bandgap, and the probe is below the optical absorption edge. Our results show that the differential transmission of the probe is negative with three distinct features: i) an initial probe absorption due to two-photon absorption involving also the pump pulse ii) a fast relaxation transient lasting to about 1 ps in which the differential transmission recovers by almost 90%. iii) a very slow recovery of the transient that lasts about 200 ps. We explain the observed transients in terms of the relaxation of the carriers to the conduction band bottom, followed by the formation of excitons and trions, and the response of the excitons and trions. The extremely large exciton and trion binding energies make these states much more preferable than the free carrier states in the bands. The contributions of interband and intraband processes to the observed transients will be explained and the extracted relaxation and recombination rates will be discussed.