Abstract

After graphene, transition metal dichalcogenides (TMDC) have appeared as the front-runners of the layered semiconducting family, offering a plethora of intriguing optoelectronic properties. Due to reduced dielectric screening and strong Coulomb potential, optoelectronic properties in these classes of semiconductors are governed by various quasiparticles like excitons, trions, bi-excitons besides free electrons and holes. This thesis explores quasiparticle dynamics in transition metal dichalcogenides and one of its successor- metal monochalcogenides (MMC) upon ultrafast light-matter interaction.

Excitonic bandgap in TMDCs can be tuned by altering the Coulomb field strength between its constituents by modifying the surrounding cloud of charge-carrying quasiparticles and hence, the effective dielectric permittivity. As an approximation, static approaches are commonly used to comprehend the experimental outcomes of bandgap tuning through the injection of free carriers, engineering of the dielectric environment, and photo-excitation. However, such an electrostatic framework overestimates the excitonic screening effect as the screening is particularly sensitive to some 'characteristic frequencies.' Despite some theoretical arguments on frequency-dependent exciton screening, there was no direct experimental evidence, to the best of my knowledge. This thesis provides first-ever experimental evidence and hence, a first stepping stone towards the perception of exciton-induced dynamic screening of excitonic Coulomb potential. As a prototype TMDC material, multilayered molybdenum disulfide (MoS_2) has been used. The resonance energy of an exciton state appears to be the characteristic frequency at which Coulomb screening from other excitons is efficient. On the other hand, the quasiparticle bandgap of semiconductors can be altered effectively by quasi-static screening from electron-hole plasma. Earlier, it has been demonstrated that plasma formation is retarded by a timescale related to the inverse of the plasma frequency. However, the experimental distinction between the carrier and electron-hole plasma and the transition of the former to the latter remained unexplored in a wider context. Electron-hole plasma formation is experimentally observed through exciton-plasma interaction. Under intense photo-excitation $\sim 10^{19}$ per cm^3, huge damping destroys the Coulomb-correlation and hinders the plasma formation until a majority of the free-carriers recombine and plasma oscillation period becomes sufficiently smaller than the damping time constant. Moreover, only 1-3% of the injected free carriers form Coulomb-correlated plasma. This study sheds light on exciton-plasma interactions and quasi-static Coulomb screening, which play pivotal roles in device engineering.

Under intense photo-excitation, light-matter interactions lead to various nontrivial quasiparticle dynamics that remained unexplored. As the excitation density is tuned near the Mott threshold, excitonic quasiparticles become unstable, and enhanced trion formation is observed. In a photo-induced non-thermal regime, bandedge A excitons display linewidth narrowing. In stark contrast to available studies that observe photo-induced exciton linewidth broadening, the reduced exciton-phonon coupling is demonstrated in the non-thermal regime following intense photo-excitation. Such anomalous behavior has never been reported earlier in any transition metal dichalcogenides.

Free carriers dominate the light-matter interaction of layered semiconductors with weak exciton binding energies. The author explores the photo-induced carrier dynamics in a metal monochalcogenide- Gallium Telluride (GaTe) and observe indirect carrier relaxation in the direct bandgap material. Such unique carrier dynamics make this material ideal for photodetection as the direct bands ensure high absorption oscillator strength, and slow, indirect carrier recombination facilitates efficient carrier extraction leading to sizable light-to-electricity conversion. This work explains the physical origin behind the excellent photo-detection properties of GaTe.

As the technology is inching towards atomically thin semiconductor-based compact devices, external control on the optoelectronic properties is of prime importance. Due to reduced dielec-

tric screening, optical properties of TMDCs are dominated by Coulomb-bound quasiparticles, like excitons, trions, and electron-hole plasma, which are effectively tuned by the photo-induced population through screening and other many-body interactions. In contrast, excitons are weakly bound in layered metal monochalcogenides, and free-carrier induced transition dictates the optical properties. Here, photo-induced population enables tunability in optical properties through state-filling due to the Pauli exclusion principle. This thesis explores photo-induced ultrafast quasiparticle dynamics that facilitate the tunability of optical properties in these layered semiconductors.

Keywords: Transient absorption spectroscopy, Transition metal dichalcogenides, transition metal monochalcogenides, carrier dynamics, Coulomb screening, Exciton, Trion, Electron-hole plasma.