ABSTRACT

Tunable light-matter couplings in dissimilar constituents (metal & semiconductors) have resulted in significant progress for the new class of mixed-dimensional hybrid materials. In this regard, the complexes fabricated using transition metal dichalcogenides (TMDs) and metal nanostructures play a leading role in the development of two-dimensional (2D) quantum hybrids along with their applications in Si-compatible photonics. In this dissertation, the many-body properties of the excitonic complexes in TMDs (MoS_2 & WS_2) and tunable exciton-plasmon coupling in metal-TMDs quantum hybrids have been studied using both steady-state and femtosecond pump-probe spectroscopy.

Pristine p-WS_2 and PVP encapsulated Ag-nanoparticle incorporated WS_2 (n-AGPW) layers have been synthesized using different solution-processable chemical exfoliation schemes. The intercalant impurity-free AGPW plays dual role by inducing induced chemical doping and plasmon enhanced light harvesting. Both of the 2D/3D heterojunction photodetectors (p-WS_2/n-Si & n-AGPW/p-Si) exhibits excellent rectification characteristics with peak responsivity of 1.1 A/W & 8.0 A/W at −10 V reverse bias, respectively, demonstrate the superiority of AGPW based plasmonic photodiode as compared to the non-plasmonic one. The ultrafast origin (Wannier-Mott nature) and carrier lifetimes of different excitonic complexes (excitons & biexcitons) at room temperature in pristine layered WS_2 dispersion have been investigated using a non-collinear femtosecond pump-probe spectroscopy technique. The binding energies of AA (~ 69 meV) and BB (~ 66 meV) biexcitons are extracted experimentally. Our detailed helicity resolved pump-probe investigations have been conceded to explore the ultrafast generation and evolution of individual bright exciton-plasmon polaritons (bright plexcitons) in self-assembled size-tunable Au nanostructure/layered WS_2 hybrids. A remarkably robust Rabi-splitting energy (~ 250 meV) and comparatively higher stable plexciton formation time (~ 7.0 ps) are realized for both the plexcitons, validating the strong-coupling conditions of polariton formation. The real-time investigation of double Fano lineshapes is also demonstrated in Au nanodisk-MoS_2 hybrid. The time-domain double Fano build-up starts at ~ 1.0 ps timescale, which sustains up to 5.0 ns. In summary, these diverse ultrafast light-matter coupling phenomena presented in this dissertation for layered TMDs and their plasmonic hybrids reveal their potential in Si-compatible 2D/3D heterojunction photodetectors and quantum photonics.