

Abstract

Light matter interactions in atomically thin van der Waals materials have attracted significant attention in recent days [1–6]. Although the thickness does not exceed few nanometers, such atomically thin materials alone or in combination with other nanostructures show exciting and unexpected photodetection properties [7–16].

Fabrication of atomically sharp junctions can be achieved with 2D van der Waals heterostructures, which significantly enhances the scope to design new types of physical systems, where novel phenomena can be studied [15, 17, 18]. Heterostructures combine properties of dissimilar materials resulting in improved device performances and hence, can be applied to multiple fields [19–21].

This thesis encompasses a photoresponse study of various atomically thin heterostructures made of graphene, bilayer-graphene (BLG) and MoS₂. A graphene-on-MoS₂ heterostructure, made of monolayer graphene and few atomic layers of MoS₂, combine superior electronic transport properties of graphene with the optical properties of MoS₂. Such hybrids exhibit enormous photoresponsivity, with values as high as $\sim 10^{10}$ A W⁻¹ at ~ 130 K and $\sim 10^8$ A W⁻¹ at room temperature, which make these the most photoresponsive materials available till date. Presence of tunable persistent photoresponse allows these to function as optoelectronic memory devices; where the persistent state shows near perfect charge retention within the experimental time scale of operation (~ 12 hrs).

Noise-free large gain ($10^9 - 10^{10}$) mechanism is one of the salient features of graphene-MoS₂ hybrids. Devices made from BLG-on-MoS₂ hybrids further help in improving the photoresponsive gain in these devices, and a large photoresponsivity ($\sim 10^9$ A W⁻¹) is maintained even when operating these devices at low channel bias ($V_{DS} < 50$ mV), or at a low range of channel current ($I_{DS} < 10$ nA). In an optimized operating condition, where circuit noise is lower than the signal from a single photoelectron, BLG-on-MoS₂ devices function as a number-resolved single-photon detector. High specific detectivity and low noise equivalent power of these devices, allow investigation of photon noise present in an optical source.

Along with the optoelectronic property study, various optical and electrical characterizations are adapted that explain the interface properties of graphene-MoS₂ heterostructures. For example, Raman spectroscopy and photoluminescence study at the interface

suggest strong interlayer coupling and efficient dissociation of excitons respectively, which play a key role in attaining large photoresponse. Interfacial barrier characteristics are also investigated in a vertical graphene-MoS₂ geometry, which shows that the barrier height can be tuned by applying an electrostatic field.

Various experimental techniques and instruments, such as heterostructure fabrication technique and setup, optical cryostat etc., were developed in house to accomplish experimental investigation, which are discussed in details.

Results of photoresponse study in van der Waals materials have opened up the possibility of designing a new class of photosensitive devices which can be utilized in various optoelectronic applications such as in biomedical sensing, astronomical sensing, optical communications, optical quantum information processing and in applications where low intensity photodetection and number resolved single photon detection attracts tremendous interest.