Abstract of the thesis

Graphene, a two-dimensional allotrope of sp^2-bonded honeycomb carbon (C) lattice, has been studied extensively since its discovery in 2004. Despite many intriguing properties, graphene has limited applications in optoelectronic and photovoltaic devices, since pristine graphene lacks any electronic bandgap and absorbs only ~2.3% of incident radiation over the broad electromagnetic spectrum which is not quite good enough for the realization of commercial photodetectors and solar cells. To overcome this limitation, various efforts were made e.g. formation of nanostructures from graphene, e.g. graphene nanoribbons, graphene oxide, graphene quantum dots, etc. Graphene Quantum Dots (GQDs) is the ultra-small nano-fragments of graphene with lateral size less than 10 nm. GQDs are well known for their unique size-dependent optical properties along with excellent biocompatibility. However, pristine GQDs consist of several restrictions such as low quantum yield, low charge carrier density, etc., which limit their utilization applications. Functionalization of GQDs can tune their electronic, chemical, optical properties, etc. thus opening new gateways to a multitude of applications. In the current thesis, different synthesis methods for both pristine GQDs and GQDs functionalized by several heteroatoms such as Oxygen (O), Nitrogen (N), Sulphur (S), and Selenium (Se) have been reported and consequent device applications have been explored.

Changes in optical and compositional properties of GQDs through the thermal annealing method have been investigated. When GQDs, synthesized by carbonization of citric acid, are annealed at 100° C, 200° C, and 300° C, their PL emission peak is red-shifted from 450 nm to 580 nm. Also, low-temperature PL measurement reveals a crossover from positive thermal quenching to negative thermal quenching phenomena. The results indicate that the synthesized luminescent GQDs, with negative thermal quenching characteristics on optimized annealing, are attractive for selective and tunable optical emitters in the visible region, making them useful as potential biomarkers and optical thermometry applications (chapter 2). The influence of size and localized defect states on photogenerated carrier recombination dynamics, which affects the performance of graphene quantum dots (GQDs)-based Si-compatible near-UV heterojunction photodetectors, is reported. GQDs of varying sizes from ∼3.0 to ∼8.0 nm have been prepared by a top-down method of oxidative cutting of graphene oxide followed by hydrothermal reduction and gradient centrifugation at different speeds. 6.0 nm GQDs show the highest carrier lifetime revealed by the time-resolved PL decay study. Photodetector devices fabricated by using ∼6.0 nm diameter GQDs displayed the highest peak responsivity of 3.5 A/W (chapter 3). We have used size-tunable nitrogen functionalized graphene quantum dots with blue (BGQDs) and green (GGQDs) emissions to excite $CsPbI_3$ perovskite nanocrystals in a remote double-layer phosphor stack to fabricate warm white LEDs. A correlated color temperature of ~5182 K is achieved, which is suitable for daylight-white) tonality along with a high-quality color rendering index (~90%) and exceptionally high luminous efficiency of radiation. The best device exhibited an LER of 251 lumens/watt (chapter 4). We have synthesized nitrogen-doped graphene quantum dots (N-GQDs) with tunable emission and electrical properties by changing the reaction solvent. While switching from protic H_2O solvent to aprotic DMF solvent, the emission wavelength of N-GQDs shifts from green to red. As-synthesized N-GQDs have been devised as an absorbing layer in quantum dots sensitized solar cells (QDSSCs). Our best device outperforms similar quantum dot sensitized solar cells in terms of power conversion efficiency (>5%) (chapter 5). We have developed a facile method to synthesize UV-NIR dual absorbing hybrid nanostructures for two-band self-powered photodetector devices. S-doped GQDs and Se-doped GQDs were synthesized by solvothermal methods, and nanocomposites of them with lead chalcogenides were synthesized. As synthesized S-GQD/PbS and Se-GQD/PbSe nanocomposites showed excellent absorption and PL emission behaviors in the NIR region. MSM type photodetector devices were made by spin coat our devices operate at a broad wavelength range of 400-1800 nm and show peak responsivity of 1.8 A/W at NIR wavelengths under zero-bias conditions, making them best performing self-powered NIR photodetectors made from colloidal quantum dots. Our devices also perform well at near UV-blue wavelengths with a peak responsivity of 2.5 A/W, making them quite attractive for two-colors/dual-band self-powered photodetectors (chapter 6).

Keywords: Graphene quantum dots, functionalized GQDs, UV-visible photodetectors, NIR photodetectors, solar cells, light emitting devices, phosphor LEDs