## Abstract

Most commercial photodiodes that work in a broad spectral range covering visible to near-infrared (NIR) wavelengths are based on Si. However, these suffer from the limitations such as high cost and complex manufacturing processes. This doctoral research focuses on the realization of broadband photodetectors with enhanced responsivity throughout the visible to NIR spectral region compared to the commercially available Si photodetectors by coating a single thin organic film on top of crystalline Si surfaces. Organic semiconductors have emerged as a potential active material for the fabrication of fundamental electronic devices such as photovoltaics, photodetectors, field-effect transistors, etc., owing to their excellent mechanical flexibility, spectral tunability, and easy processability. Apart from these, organic semiconductors, being low dielectric materials, are popularly known to exhibit spin-distinct states of 0 and 1, singlets and triplets, respectively, upon photoexcitation. The larger diffusion lengths of triplet excitons, due to their longer lifetime than their singlet counterpart, make them quite advantageous in photo-sensing devices. In literature, many reports focus on exploiting the organic triplets in photo-sensing devices, but they all have complex and expensive manufacturing processes. In this dissertation, we have fabricated organic-inorganic heterojunction-based photodetector devices with the most straightforward design, responsive in a broad spectral range of 400 to 1100 nm. Tin (IV) naphthalocyanine dichloride (SnNcCl<sub>2</sub>) is vacuum-deposited as the organic semiconductor material on crystalline p-Si to form a heterojunction. The growth of SnNcCl<sub>2</sub> is studied extensively with atomic force microscopy (AFM) and x-ray reflectivity (XRR) analysis on the planar p-Si surface to achieve efficient junction properties of the fabricated photodiodes. The ultrafast transient absorption spectroscopy (UTAS) measurements unveiled SnNcCl<sub>2</sub> as a potential triplet donor in the subpicoseconds regime with more than 60% yield. One of the most important outcomes of this thesis is that we have shown that the higher-energy triplet (T<sub>n</sub>) excitons of SnNcCl<sub>2</sub> could dissociate at the junction contributing to the photoresponsivity of the fabricated devices, which distinguishes our work from the reported results focus on dissociating the first-order triplets forming heterojunctions with low-bandgap materials. In the later part of the thesis work, a coevaporated film of SnNcCl<sub>2</sub> and PTCDI-C<sub>8</sub> (N,N'-Dioctyl-3,4,9,10-perylenedicarboximide) is thermally grown on the pyramidal-textured p-Si surface to fabricate photodetector devices. The reduction of reflection loss from the Si surface and the transfer of SnNcCl<sub>2</sub> triplets (T<sub>1</sub>) to PTCDI-C<sub>8</sub> contribute to the broad spectral responsivity of the devices. A typical final device shows photoresponse greater than 0.1 A/W at -2 V with a maximum responsivity of 0.45 A/W at 1000 nm throughout the spectral region from 400 nm to 1100 nm with detectivity of the order of 10<sup>11</sup> Jones, which shows its efficacy compared to other reports on SnNcCl<sub>2</sub>-based and other similar devices till date.

Keywords: Organic-inorganic heterojunction, growth study, atomic force microscopy (AFM), x-ray reflectivity (XRR), broadband photodetectors, photoresponsivity, ultrafast transient absorption spectroscopy (UTAS), triplet excitons, triplet donor, SnNcCl<sub>2</sub>, PTCDI-C<sub>8</sub>