Hybrid Systems for Next-Generation Photodetectors: From Layered Inorganic/Organic Heterojunctions to Perovskites

Abstract

In an era of rapid technological advancement, innovations in optoelectronics have enabled a diverse range of applications in sensing, communications, and energy conversion. At the heart of many such optoelectronic technologies lies a photodetector that serves as a bridge between an optical signal and an electronic gadget. Their ability to sense light and turn it into an electrical signal makes them indispensable in many critical applications, including medical imaging, environmental monitoring, image sensors, and telecommunications, among others. In recent years, organic-inorganic hybrid materials have gained tremendous interest due to their remarkable and unique optoelectronic properties. These hybrid materials leverage the added benefits of both material systems. This thesis focuses on the fabrication and characterization of high-performance and stable photodetectors based on organic-inorganic hybrid materials.

This work explores two categories of hybrid materials: layered inorganic-organic heterojunctions (HJs) and organic-inorganic hybrid perovskites, with an emphasis on developing photodetectors that show concurrent enhancement of multiple performance metrics. The first part of the thesis investigates devices based on layered inorganic-organic HJs. Mechanically exfoliated layered transition metal dichalcogenides (TMDs) served as the inorganic component, paired with poly(3-hexylthiophene) (P3HT) as the organic counterpart. To start with, a lateral heterostructure was created using tungsten diselenide (WSe_2) and P3HT to boost responsivity and speed. This WSe_2/P3HT hybrid device demonstrates enhanced photoresponse and broad detection capabilities (400−1100 nm) attributed to better exciton separation and carrier collection from the formation of a type-II HJ, along with long-range energy transfer from P3HT to WSe_2. Furthermore, the device maintains stable performance in ambient conditions, achieving a peak responsivity of 17.6 A W^{-1} for low incident light intensities ($<10^{-5}$ W cm^{-2}) at a wavelength of 640 nm, with a rapid response speed of 9.5/5.1 μs. Following this, the device structure was altered to a three-terminal field-effect transistor configuration, enabling gate-controlled photocurrent mechanisms. An n-p HJ phototransistor was developed using n-type molybdenum diselenide ($MoSe_2$) and p-type P3HT. This architecture facilitates high photo-gain and reduced dark current through its intrinsic amplification. The hybrid phototransistor achieves a responsivity of 40.4 A W^{-1} and a detectivity of 2.03×10^{10} Jones at 660 nm while also ensuring a quick response speed of 363/370 μs. These findings underscore the potential of hybrid systems and interfacial engineering in attaining high responsivity and speed, paving the way for their integration into advanced optoelectronic devices.

In the second part of the thesis, the photodetection capabilities of organic-inorganic hybrid perovskites are investigated. Recent advancements in device design and material synthesis have brought perovskite photodetectors closer to achieving their potential in imaging, sensing, and optical communication, due to their excellent performance. However, critical performance metrics such as linear dynamic range (LDR) and low-light detection capability, both essential for high-sensitivity imaging and sensing applications, remain underexplored. First, a photodetector was fabricated using $FAPbI_3$ (formamidinium lead iodide) perovskite film exhibiting outstanding LDR and excellent low-light detection capability. To optimize its performance, the effect of hole transport layers (HTLs) on device operation is systematically investigated. This study reveals the significance of HTL selection on key metrics such as charge extraction efficiency, speed, and linearity. The $FAPbI_3$ devices with optimized HTLs exhibit a large LDR that exceeds 200 dB and can detect light as faint as a few pW cm^{-2}. The best $FAPbI_3$ devices also exhibit a fast response speed of nearly 1 μs. Subsequently, lower-dimensional structures, specifically $FAPbI_3$ perovskite quantum dots (PQDs), were also explored for their suitability for photodetector applications. PQDs offer enhanced spectral tunability and stronger light absorption due to the quantum confinement effect. Additionally, the presence of surface ligands in PQDs facilitates defect passivation, improving their stability and reducing non-radiative recombination. The $FAPbI_3$ PQD device exhibits high EQE reaching 80% in the visible range. It also demonstrates a high linear dynamic range of 181 dB and a rapid response time of 505 ns. Overall, these results demonstrate the suitability of hybrid perovskites for developing practical and high-performance photodetectors.