DOCTORAL THESIS

Organic Photovoltaic and Photomultiplication Detectors for Imaging Applications

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Abstract

In comparison with the commercial inorganic semiconductor-based photodetectors, the solution-processable organic photodetectors (OPDs) offer an attractive option for use in imaging applications, security monitoring and artificial intelligence. This research work aimed at developing high-performance solution-processable OPDs, including transparent near-infrared (NIR) OPDs and the novel photomultiplication (PM)/photovoltaic (PV) dual-mode OPDs for imaging applications. The recent advances in low-bandgap organic materials and solution fabrication technologies have provided an encouraging pathway for imaging applications using OPDs. The PhD work included: (1) analyzing the performance of the NIR OPDs, (2) investigating the photoresponses of the transparent NIR OPDs and the application in NIR visualization, (3) examining the unique filter-free spectral selective photoresponse behavior of the novel PM/PV dual-mode OPDs, and (4) demonstrating the use of the dual-mode OPDs for imaging applications.

The photoresponse of the solution-processed large-area transparent NIR OPDs with a high conductivity poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS) upper transparent electrode has been analyzed. The transparent large-area self-powered NIR OPDs have a high specific detectivity of >10^{12} Jones at 850 nm, and a high -3 dB cutoff frequency of 71 kHz and a linear dynamic range of 154 dB. The visualization of NIR light can be attempted through monolithic integration of transparent NIR OPDs with perovskite light-emitting diodes (PeLEDs). In an NIR-to-visible upconversion device, the transparent NIR OPD unit acts a charge-injection layer to adjust the emission behavior in the LED unit. The hole-electron current balance in the NIR-to-visible upconversion device is controlled by the photocurrent generated in the NIR PD that responds to the NIR

portion of the incoming light. The visible light emission in the LED unit is realized in area where the effective charge injection occurs, adjusted by the NIR PD unit in the presence of the NIR light, such that the objects reflecting or illuminating NIR light can be visualized.

In parallel to the optimization of transparent NIR photodetection component, the efficiency of the emission component is another crucial factor for determining the overall performance of the NIR-to-visible upconversion devices. Among different types of reported LEDs, the emerging PeLEDs are very attractive for use in NIR-to-visible upconversion devices because they have a high color purity with narrow emission spectrum, high brightness, and high photoluminescence quantum yield. The performance of the large-area cesium lead bromide ($CsPbBr_3$)-based PeLEDs has been optimized through a controlled growth of the perovskite emission layer modified using a hybrid additive approach. The results reveal that the use of the hybrid additive helps to suppress both the structural defects and nonradiative recombination-induced leakage current and assists in improving the electron-hole current balance in the PeLEDs.

Apart from the effort in developing NIR visualization devices, spectral crosstalk-free photodetection is another important technique with practical impacts for use in image sensors. The present commercial inorganic semiconductor-based broadband photodetectors require dedicated color filters to realize spectral selective photodetection. The use of the filters increases the complexity and cost of the image sensors. In this work, a novel bias-switchable PM/PV dual-mode OPD has been developed for image applications. The PM/PV dual-mode OPD has a heterostructure photoactive layer, comprising a tandem organic bulk-heterojunction (BHJ) architecture. The first BHJ absorbs the short-wavelength portion of the incoming light, whereas the second BHJ has an extended absorption to longer wavelengths. The spectral photo-selective photodetection is realized by adjusting the difference in wavelengths between the transmission cutoff wavelength of the first BHJ and the absorption edge of the second BHJ in the dual-mode OPDs. Both BHJs are connected in series to form a back-to-back structure. When a forward bias is applied across the ITO and Al contacts, the second BHJ is under its forward current. The first BHJ is under its reverse-biased condition for generating the photocurrent due to the absorption of the short-wavelength portion of the incoming light. Likewise, when a reverse bias is applied across the ITO and Al contacts, the first BHJ is under its forward-biased conduction to conduct the current. The second BHJ is under its reverse-biased condition to generate the photocurrent due to the absorption of the long-wavelength portion of the incoming light.

To suppress the spectral crosstalk in the PM/PV dual-mode OPDs, a long-wavelength absorbing functional layer has been adopted in the second BHJ. The use of the PM effect helps to promote the external quantum efficiency of the dual-mode OPDs over 100%, leading to a high photosensitivity for the long-wavelength portion of the incoming light. The dual-mode OPDs with a spectral selective photoresponse in two distinct bands over the visible light spectrum have been realized. For example, the dual-mode OPD, having a device configuration of ITO/ZnO/F8T2:$PC_{61}BM$/MoO_3:PEDOT:PSS/P3HT:$PC_{71}BM$/Al, has a high photoesponse over the wavelength of < 500 nm, operated under the forward bias. It also has a high photoresponse over the wavelength range from 625 to 650 nm when it is operated under the reverse bias. The high-performance spectral selective dual-mode OPDs have been used for blue and red color imaging analysis, demonstrating an excellent filter-free color imaging capability.

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