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Top illuminated organic photodetectors with dielectric/metal/dielectric transparent anode

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ABSTRACT

The top illuminated organic photodetectors (OPDs) with a Dielectric/Metal/Dielectric (DMD) transparent anode are fabricated. The transparent electrode is composed of molybdenum trioxide (MoO_3)/silver (Ag)/MoO_3 layers and zinc oxide (ZnO)/aluminum (Al) is used for bottom cathode. The optimized DMD electrode has an optical transmittance of 85.7% at the wavelength of 546 nm and sheet resistance of ~6 Ω/sq. The fabricated OPDs exhibit a high detectivity and wide range linearity.

1. Introduction

Over two decades, organic photovoltaics and photodetectors have been extensively studied due to their easy deposition methods, large-area processing capabilities, and fine tunability of optical absorption coefficients [1,2]. Especially, organic photodiodes (OPD) have been actively studied as potential low-cost, high-performance alternatives to amorphous silicon (a-Si) photodiode for flat panel imagers [3–5]. Among various kinds of the imagers, indirect type imager based on CsI(Tl) scintillator looks very promising for OPD application due to its reduced lateral light scattering, high light output, fast response time, and suitable peak emission spectra [6,7]. Among various organic materials for such application, Poly(3-hexylthiophene-2,5-diyl):Phenyl-C_{61}-butyric acid methyl ester (P3HT:PCBM) bulk heterojunction (BHJ) blend system appears to be very desirable candidate [4,5,7–9]. Ng et al. used 4 µm thick poly[(2-methoxy-5-ethylhexyloxy)-1,4-phenylenevinylene] (MEHPPV): PCBM BHJ blend layer as photoactive layer in combination with a flexible a-Si thin-film transistor (TFT) backplane to realize an imager array with 35% external quantum efficiency [3]. They used solution processed ITO nanoparticles as top transparent electrode which had a very high sheet resistance of 1 MΩ/sq, resulting in significant current loss during signal readout. Alternatively, Tedde et al. demonstrated a concept of a-Si:H TFTs active pixel sensor (APS) imager integrated with the OPD. The APS provides on-pixel amplification up to 10 compared to passive pixel sensor (PPS) [4]. They adapted conventional structure of OPDs with top Ca/Al transparent cathode. When low-workfunction metals are used as transparent electrode, post-process encapsulation is required. The long-term stability of such devices cannot be guaranteed, also the Ca/Al transparency is rather low. To enhance the device stability in air, the inverted organic solar cell or photodiode architectures are investigated. In such devices a high workfunction metal such as Au, Ag, or PEDOT:PSS are used as top transparent electrodes [10–16]. Although a large number of works have been reported for inverted type organic solar cells [10–14] or inverted type organic photodetector with the bottom illumination [15], only a few inverted type organic photodetectors with top

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illumination geometry have been reported so far. For example, Baierl et al. demonstrated the inverted type P3HT:PCBM OPD with PEDOT:PSS as a top electrode [17,18]. In such device configuration the PEDOT:PSS suffers from a high acidity, hygroscopic behavior, resulting in inferior stability [19]. Alternatively, ultra-thin metallic films can also be used as a transparent electrode in OPV and OPD application [4,5,20,21]. In such configuration, the reflection from the metal surface is too high and a large portion of incident light can be wasted. To suppress such high reflections, an optically transparent dielectric material can be employed as an anti-reflection layer, leading to the Dielectric/Metal/Dielectric (DMD) multilayer electrode configuration. Although a number of papers on the application of the DMD transparent electrode to organic light emitting diode and organic solar cell have been published [22–26], to our best knowledge, no research on the top anode, top-illumination OPD with the DMD configuration has been reported. In this paper, we demonstrate a top-anode, top-illumination OPD with engineered MoO_3/Ag/MoO_3 DMD semi-transparent electrode that could be used for large-area imager application. The optical simulation and experimental measurement are conducted to investigate the effect of the DMD semi-transparent electrode on the OPD properties. Also, OPD electrical characteristics including current–voltage properties, quantum efficiency, noise equivalent power, and detectivity are evaluated.

2. Experimental

2.1. Device fabrication

Low-cost soda lime glass substrates (Asahi glass) were cleaned by acetone, isopropyl alcohol, and deionized water by sonication for 5 min for each process, and treated by oxygen plasma for 5 min subsequently. 100 nm of aluminum layer was thermally evaporated using shadow mask for patterning bottom cathode. ZnO sol–gel solution was prepared by dissolving 0.5 M of zinc acetate dihydrate ($Zn(CH_3COO)_2{\cdot}2H_2O$, Sigma Aldrich) as a precursor in 2-methoxyethanol (2ME, Sigma Aldrich) solvent. 0.5 M of mono-ethanolamine was added as a stabilizer and the mixture was vigorously stirred at a temperature of 60 °C for 4 h. The solution was then cooled down and aged for more than 24 h. The synthesized ZnO solution was spin-coated on top of the patterned cathode layer with spin-coating speed of 2000 rpm for 30 s and annealed at 150 °C for 20 min, yielding 40 nm thickness of ZnO layer. The substrates were transferred into a glove box with nitrogen atmosphere for deposition of photoactive layer. 25 mg of P3HT (Rieke Metals) and 25 mg of PCBM (American Dye Source, Purity: >99.5%) were mixed into 1 mL of dichlorobenzene (DCB) and stirred by magnetic bar overnight for the BHJ solution. The solution was filtered by 0.45 μm syringe filter and then spin-coated onto the ZnO layer with different spin-coating speed to realize OPDs with active layer thickness of 200 nm, 320 nm and 450 nm. To prevent complete drying of P3HT:PCBM film, the spin-coating timing was adjusted for each process. The active layers were then solvent-assisted annealed [27] for 30 min to 1 h to ensure fully dried films in N_2 atmosphere. The substrates were transferred into thermal evaporator for deposition of 5 nm MoO_3, 10 nm silver and 35 nm MoO_3 subsequently. A shadow mask was used for top anode fabrication. All the devices were encapsulated with a thin slide glass sealed by UV curable epoxy resin. The device size was 0.04 cm^2 with square shape.

2.2. Device measurement

Optical transmittance and absorbance spectra of the semitransparent DMD, DM multilayer, and P3HT:PCBM BHJ film were measured using Agilent CARY-5E UV–vis spectrometer. Optical reflectance of the DMD, DM electrode were measured by Filmmetrics F20 thin-film measurement system. Sheet resistances of the electrodes are measured by 4-point probe method. A solar simulator (Oriel) equipped with Xenon lamp and band pass filter with peak wavelength 546 nm (FWHM = 2 nm), was used for the current density–voltage (*J*–*V*) characteristic measurement under illumination. The irradiance of the illuminated light was measured by Newport power meter. The *J*–*V* characteristics of all the organic photodiode under illumination and dark conditions were measured by HP2416A semiconductor measurement system with a probe station in a dark Faraday cage. External Quantum Efficiency (EQE) was measured with a setting of lock-in amplifier (Stanford Research Systems SRS 830), monochromator with a 100 W halogen tungsten lamp, light chopper, and UV-enhanced silicon photodetector (Newport UV808) for calibration.

2.3. Methods

Optical modeling method and detailed description of the OPD figures of merits can be found in the Supplementary Materials.

3. Results and discussion

3.1. DMD top electrode simulation, characterization, and optimization

The Dielectric/Metal/Dielectric (DMD) electrode in comparison to other transparent top electrode has desirable properties such as (i) the DMD electrode has anti-reflective MoO_3 dielectric layer on top, which maximizes the transmission of the light of interest, and (ii) the passivation of the top electrode with the additional MoO_3 layer allows direct deposition (e.g. thermal evaporation [28]) of CsI(Tl) scintillator materials, which is beneficial in achieving a higher scintillator gain [7]. Fig. 1 shows the schematic of the fabricated top illuminated OPD. To find optimum layer thicknesses ensuring that a maximum amount of incident light can pass through the top DMD multilayer transparent electrode, an optical simulation is carried out based on refractive indices of all the OPD layers measured by a spectroscopic ellipsometer. The measured refractive indices of MoO_3 and Ag layers over wavelength from 400 nm to 800 nm are shown in Fig. S1 (Supplementary