



Nanocomposite perovskite based optical sensor with broadband absorption spectrum

Zahra Moradi^a, Hamidreza Fallah^{a,b,*}, Morteza Hajimahmoodzadeh^{a,b,*}

^a Department of Physics, Faculty of Science, University of Isfahan, Isfahan 81746-73441, Iran

^b Quantum Optics Research Group, University of Isfahan, Isfahan, Iran



ARTICLE INFO

Article history:

Received 3 February 2018

Received in revised form 30 June 2018

Accepted 16 July 2018

Keywords:

Optical sensor

Organic-inorganic hybrid perovskite

Nanocomposite

Photoresponsivity

Detectivity

ABSTRACT

Recently, organic-inorganic hybrid lead halide perovskite optical sensors have attracted lots of attention for their unique and remarkable optoelectronic properties like low-cost solution process deposition. Also, inorganic nanoparticles with narrow bandgap as photocarrier generator has been used in high efficiency low cost optical sensors to increase absorption spectrum. In this paper, we designed and fabricated a solution processed optical sensor by using nanocomposite of Methylammonium lead iodide and PbS nanoparticles as the active layer. It has high absorption in the broad range of spectrum that can work in small driving voltages (less than 1 V). It has low dark current and high photocurrent that are important parameters in optical sensors. This device achieves excellent photoresponsivity and high external quantum efficiency for broad range of wavelength from 370 nm to 940nm.

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1. Introduction

Optical sensors or photodetectors (PDs) have a lot of applications such as imaging, environmental monitoring and medical sensing [1]. The conventional optical sensors are made of SiC for detection in UV region, Si for detection in the visible region and HgCdTe for the infrared region. These kinds of the optical sensors usually have narrowband response [2,3].

Recently, organic-inorganic perovskite materials are used as absorbers in optical sensors and have attracted lots of interest for their unique potential to enable low cost, broadband spectrum detection [4–10]. Methylammonium lead halide perovskites have excellent semiconducting properties such as direct and tunable bandgap, wide absorption range, high carrier mobility and long charge diffusion length [3,11–13]. These materials can be prepared and processed from solution process methods that make them suitable to produce low cost optoelectronic devices such as photovoltaic (PV) solar cells [14], optical sensors [4,15] light emitting diodes (LEDs) [16–19] and field effect transistors (FET) [20]. In this study, $\text{CH}_3\text{NH}_3\text{PbI}_3$ is chosen as the active layer with low bandgap (~ 1.5 eV) among organohalide perovskite materials to have near Infrared absorption [21–23].

Today, inorganic nanoparticles with narrow bandgap have been used as photo charge generator in high efficiency low cost optical sensors. Colloidal nanoparticles with narrow bandgap such as lead sulfide (PbS) and lead selenide (PbSe) can be used in perovskite layer as nanocomposite to enhance absorption spectrum [18,24].

The goal of this work is design, fabrication and characterization of the optical sensor based on nanocomposite of Methylammonium lead iodide and PbS nanoparticles as the active layer with high response for low incident powers, broadband absorption spectrum and high detectivity for small driving voltages (less than 1V) by using low cost solution processes deposition.

The device structure is exhibited in Fig. 1. The transparent electrode, made of fluorine-doped tin oxide (FTO), followed by an electron transporting material (ETM) made of compact and mesoporous TiO_2 . TiO_2 is one of the best ETM for perovskite devices that collects and passes photogenerated electrons to the fluorine-doped tin oxide (FTO) collector electrode. Active layer consists of nanocomposite of Methylammonium lead iodide and PbS nanoparticles. Hole transport material (HTM) is deposited on top of nanocomposite layer.

HTM should be used in the device to have better charge transport. HTMs have energy levels adapted to the bandgap of perovskite material to make better hole transport from the perovskite layer to contact [25]. Nowadays, 2,2',7,7'-tetrakis[*N,N*-di(4-methoxyphenyl)amino]-9,9-spirobifluorene (Spiro-OMeTAD) is the best material used in perovskite devices as HTM [26–29]. The HTM should be coated on top of the perovskite layer. Finally, a

* Corresponding authors at: Department of Physics, Faculty of Science, University of Isfahan, Isfahan 81746-73441, Iran.

E-mail addresses: hfallah@sci.ui.ac.ir (H. Fallah), m.hajimahmoodzadeh@sci.ui.ac.ir (M. Hajimahmoodzadeh).

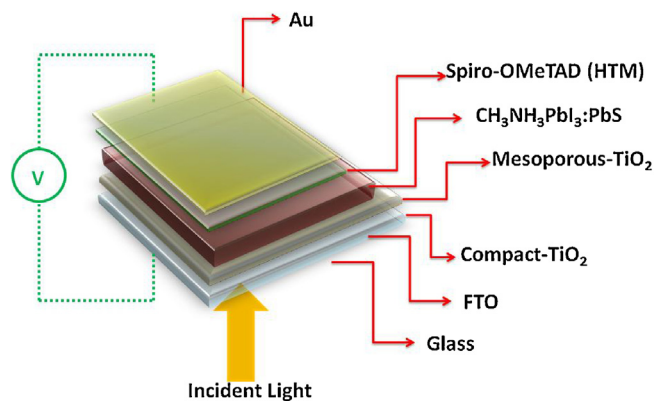


Fig. 1. Schematic of nanocomposite perovskite optical sensor with vertical configuration: FTO/c-TiO₂/meso-TiO₂/CH₃NH₃PbI₃:PbS/Spiro-OMeTAD(HTM)/Au.

metallic electrode such as gold (Au) or silver (Ag), is deposited on top of HTM by evaporation.

2. Experimental

Fluorine doped tin oxide (FTO) coated glass substrates (14 × 14 mm, ~15Ω/sq resistance) were etched with zinc powder and HCl (2 M). The substrates were cleaned with soap (Extran® MA 02 neutral) and sonicated for 15 min in a solution of water and soap. Then, the sheets were rinsed with MilliQ water and sonicated for 15 min in ethanol (95%). Finally, the substrates were rinsed with acetone and dried with compressed air. Before deposition of compact TiO₂, an Ultraviolet/Ozone treatment was performed for 15 min. Then, TiO₂ blocking layer was deposited onto the substrates by spin coating at 450 °C, using 5 mL of a titanium diisopropoxide bis(acetylacetonate) (75% in isopropanol, Sigma-Aldrich) solution diluted in ethanol (0.5:4.5, v/v), using oxygen as carrier gas. After the coating process the films were kept at 450 °C for 30 min and left to cool to room temperature. A mesoporous TiO₂ layer was deposited by spin-coating 100 μL of a mixture of TiO₂ (30 NR-D Dyesol) in ethanol (150 mg TiO₂:1 mL ethanol) at 2000 rpm during 10 s with 2000 rpm/s. After drying at 100 °C during 10 min, the TiO₂ layers were heated at 500 °C for 30 min and cooled to room temperature. The method reported in references [24,30] was used for preparing nanocomposite of CH₃NH₃PbI₃:PbS. 10 mg/ml⁻¹ of colloidal PbS nanoparticles were added into 3 mL of dimethylformamide (DMF) and dimethylsulfoxide (DMSO) 9:1 solution containing 350 mg of PbI₂ and 150 mg of CH₃NH₃I and stirred for 10 min. Then nanocomposite layer of

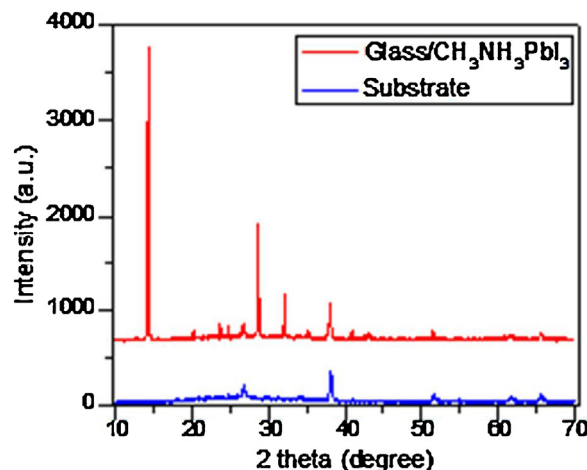


Fig. 3. XRD pattern of CH₃NH₃PbI₃ thin film on glass substrate.

perovskite and PbS nanoparticles was deposited by spin-coating 50 μL of this solution at 4000 rpm during 50 s with 4000 rpm/s. During the spin-coating procedure, diethyl ether was added to the substrate. Then, the film was heated at 100 °C for 3 min. The Spiro-OMeTAD was deposited on top of the perovskite substrates by spin coating at 4000 rpm, with 2000 rpm/s, for 30 s, using 50 μL of Spiro-OMeTAD solution. The Spiro-OMeTAD solution was prepared by dissolving 72.3 mg of (2,2',7,7'-tetrakis(N,N'-di-p-methoxyphenylamine)-9,9'-Spirobifluorene), 28.8 μL of 4-tert-butylpyridine and 17.5 μL of a stock solution of 520 mg/mL of lithium bis(trifluoromethylsulfonyl)imide in acetonitrile, in 1 mL of chlorobenzene. The deposition of top electrode made of gold was performed by thermal evaporation under ultrahigh vacuum conditions, using a commercial MBraun vacuum chamber. Before beginning the evaporation the chamber was evacuated until pressure of 1 × 10⁻⁶ mbar. Cross section Scanning electron microscope (SEM) image of nanocomposite device is shown in Fig. 2 (a) and SEM image of PbS nanoparticles in perovskite layer is shown in Fig. 2 (b).

XRD pattern of CH₃NH₃PbI₃ thin film is shown in Fig. 3. Reported XRD pattern of CH₃NH₃PbI₃ film only exhibited three low angle (<35°) diffraction peaks at 2θ of 13.9°, 28.3° and 31.7°, corresponding to the (110), (220) and (310) planes of the tetragonal perovskite structure.

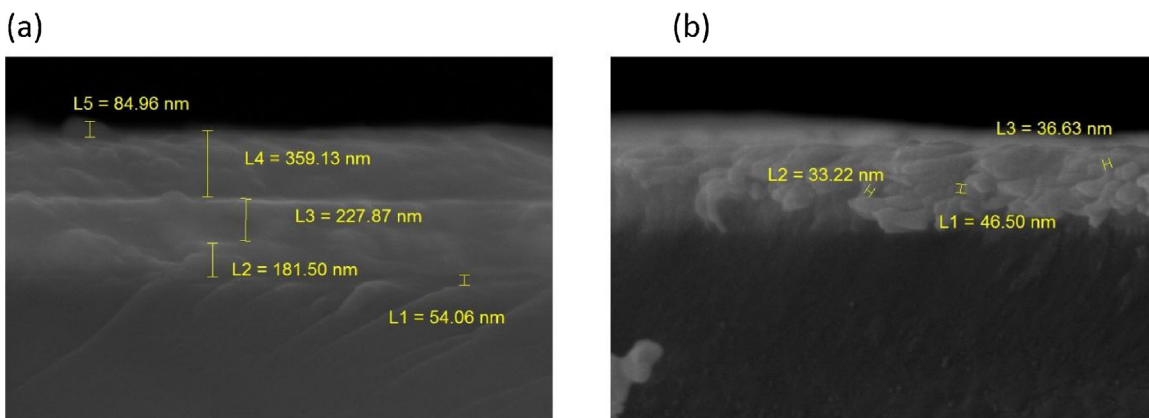


Fig. 2. SEM of (a) nanocomposite device with layers from bottom to up: compact-TiO₂ (L1)/ meso-TiO₂ (L2)/CH₃NH₃PbI₃:PbS (L3)/Spiro-OMeTAD (L4)/ Au (L5), and (b) Zoomed image of CH₃NH₃PbI₃ layer only showing PbS nanoparticles.

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