



# High performance photomultiplication perovskite photodetectors with PC<sub>60</sub>BM and NPB as the interlayers



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## ABSTRACT

Organic-inorganic hybrid perovskites have attracted more attention as successful light harvesting materials for solution-processed semiconductors and exhibit remarkable optoelectronic properties. Here photomultiplication-type photodetectors based on perovskite CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> are demonstrated. By introducing suitable interlayers at the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/electrode interfaces, the performance of the photodetector is significantly improved. The optimized device with a *N,N*-di(naphthalene-1-yl)-*N,N*-diphenylbenzidine anode interface layer and [6,6]-phenyl-C<sub>60</sub>-butyric acidmethyl ester cathode interface layer shows a broadband response with a high photocurrent gain of about 177 and a high detectivity of  $4.6 \times 10^{13}$  Jones, which are higher than the reference device. Besides, the response speed of the device is also increased. The improvement is attributed to the improved charge carrier collection efficiency and suppressed dark current of the device.

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## 1. Introduction

Photodetectors are the tools which can capture incident light and convert it into electronic signals, the light absorption material is thus critical for the devices. Recently, rapid progresses have been made in organic-inorganic perovskite solar cells [1–7], reaching a certified power conversion efficiency of 22.1% [8]. The organometal halide perovskite materials with layered structure present the properties of inorganic semiconductor materials and organic materials at the same time, and the cooperation of organic and inorganic components could greatly enhance the properties for each other. They do not only have the inorganic material properties of ultrafast charge generation, high mobility, and long charge carrier lifetime, but also enjoy the organic properties of easy fabrication and no deep-level defects. Semiconductor photodetectors with organic–inorganic halide perovskite materials which have high light absorption coefficients, large static dielectric constants (40–70) [9,10], high free carrier mobility [11], a partially tunable energy gap around 1.6 eV

[12,13], and long diffusion lengths [3,14–18] have many important applications in both industry and defense as well as scientific research [19–23]. Generally, there are typically two types of perovskite photodetectors, e.g., photodiode- and photomultiplication (PM)-types. For a photodiode-type photodetector, the external quantum efficiency (EQE) is generally lower than unity due to their limited light absorption efficiency, electron-hole pair (or exciton) dissociation efficiency, and charge-carrier collection efficiency [24–26]. On the other hand, the PM-type photodetector exhibits an EQE much higher than unity due to the mechanism that one incident photon can trigger more than one hole (or electron) flowing across the photodetector, indicating that a photocurrent gain of this device [27,28]. Due to their high responsivity and detectivity, PM-type perovskite photodetectors have drawn more and more attention recently [20,29]. Moehl. et al. observed a strong photocurrent amplification in a perovskite solar cell [30], and a direct contact of fluorine-doped tin oxide and perovskite is essential for the photocurrent amplification [31]. Dong et al. demonstrated a high photocurrent gain of about 500 in a perovskite photodetector by introducing a large concentration of Pb<sup>2+</sup> cations in the top surface of the perovskite film [20]. Meanwhile, Liu et al. reported a perovskite

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photodetector with a photocurrent gain of about 45 through incorporating PbS quantum dots as the electron trap states [32]. Although high performance perovskite photodetectors have been demonstrated, their performance should be further improved for practical applications.

One of the factors that limit the performance of a photodetector is its dark current. The dark current can be controlled by the interface layer between the perovskite film and electrode. Thus appropriate choice of electrode interlayers is critical to improve the performance of perovskite photodetectors [21,26,33]. Besides, the charge carrier collection efficiency can also be modulated by the interface layers. Such a strategy has been demonstrated in photodiode-type perovskite photodetector [34]. However, it has never been exploited in PM-type counterparts. In this work, PM-type perovskite photodetectors based on  $\text{CH}_3\text{NH}_3\text{PbI}_3$  (MAPbI<sub>3</sub>) are fabricated, which shows a broad spectral response ranging from 300 to 800 nm. By adding *N,N'*-di(naphthalene-1-yl)-*N,N'*-diphenylbenzidine (NPB) and [6,6]-phenyl-*C*<sub>60</sub>-butyric acidmethyl ester (PC<sub>60</sub>BM) as the anode interface layer and cathode interface layer, respectively, the photodetector exhibits a low dark current and thus increased photocurrent gain of about 177 and detectivity of  $4.6 \times 10^{13}$  Jones at  $-1$  V.

## 2. Experimental detail

Three photodetectors with the structures shown in Fig. 1a were fabricated:

- PD1: ITO/MAPbI<sub>3</sub>/TAPC/MoO<sub>3</sub>/Ag  
 PD2: ITO/PC<sub>60</sub>BM/MAPbI<sub>3</sub>/TAPC/MoO<sub>3</sub>/Ag  
 PD3: ITO/PC<sub>60</sub>BM/MAPbI<sub>3</sub>/NPB/MoO<sub>3</sub>/Ag

Here ITO is the cathode, PC<sub>60</sub>BM is the cathode interface layer, MAPbI<sub>3</sub> is the active layer, the NPB or di-[4-(*N,N*-ditolyl)-amino]-phenyl]cyclohexan (TAPC) is the anode interface layer, MoO<sub>3</sub> is used for anode modification, and Ag is the anode. The MAPbI<sub>3</sub> layers were prepared by a two steps spin-coating method similar to the literature [20]. PbI<sub>2</sub> was dissolved in anhydrous *N,N*-dimethylformamide to give a concentration of 600 mg/ml and the CH<sub>3</sub>NH<sub>3</sub>I (MAI) was dissolved in 2-propanol to obtain a concentration of 43 mg/ml. ITO coated glasses were ultrasonically cleaned with acetone, ethanol, and deionized water for 15 min, respectively, and then treated with ultraviolet-ozone for 15 min before

fabrication of the photodetectors. PC<sub>60</sub>BM with a concentration of 10 mg/ml dissolved in chlorobenzene solution was coated on ITO glass at 2000 rpm for 30 s, which formed a layer of about 80 nm. Then the PbI<sub>2</sub> solution was spun on ITO substrate at 3000 rpm for 60 s and dried at 70 °C for 5 min. The MAI solution was spun on top of dried PbI<sub>2</sub> film at 3000 rpm for 60 s and then annealed at 100 °C for 1 h, which formed MAPbI<sub>3</sub> layers with different thicknesses depended on the substrates. The device fabrication processes above were done in a nitrogen filled glovebox (<0.1 ppm H<sub>2</sub>O and O<sub>2</sub>). Subsequently, TAPC or NPB film (6 nm), 12 nm MoO<sub>3</sub>, and 100 nm Ag were thermally evaporated sequentially on the MAPbI<sub>3</sub> layers under a pressure of  $5.0 \times 10^{-4}$  Pa. The photoactive area of the devices is 6 mm<sup>2</sup> as defined by the cross-bar configuration of the 2 mm wide ITO electrode and the 3 mm wide Ag electrode. X-ray diffraction (XRD) patterns were collected on a Bruker D8 Focus diffractometer with a Cu K $\alpha$  radiation ( $\lambda = 1.54056$  Å) operated at 40 kV and 40 mA. Scanning electron microscopy (SEM) images were measured on a Hitachi S4800. Absorption spectra of the films on ITO-coated glass substrates were recorded with a Shimadzu UV-3101PC spectrophotometer. EQE spectra were performed with a Stanford SR803 lock-in amplifier under monochromatic illumination at a chopping frequency of 130 Hz by a Stanford SR540 chopper. Current-voltage (J-V) curves of the devices in the dark and under illumination were measured with a Keithley 2400 sourcemeter.

## 3. Results and discussion

The morphologies of the MAPbI<sub>3</sub> films on ITO and ITO/PC<sub>60</sub>BM are shown in Fig. 2a and b. The two MAPbI<sub>3</sub> films have almost the same dense, homogeneous, and smooth morphology, indicating that the PC<sub>60</sub>BM layer has little effect on the morphology of MAPbI<sub>3</sub> films, and the average grain size is about 400 nm. On contrast, the two films exhibit different thicknesses, as shown in the cross-sectional SEM images in Fig. 2c and d. The thickness of MAPbI<sub>3</sub> film on ITO is about 550 nm, while the total thickness of PC<sub>60</sub>BM and MAPbI<sub>3</sub> film is 520 nm. This indicates MAPbI<sub>3</sub> film on PC<sub>60</sub>BM has a lower thickness, which is attributed to the different roughness and surface properties of ITO and PC<sub>60</sub>BM.

The XRD patterns of the two films are depicted in Fig. 3. Similarly, the two films also exhibit the same XRD pattern with no obvious impurity, indicating that the PC<sub>60</sub>BM film has no impact on

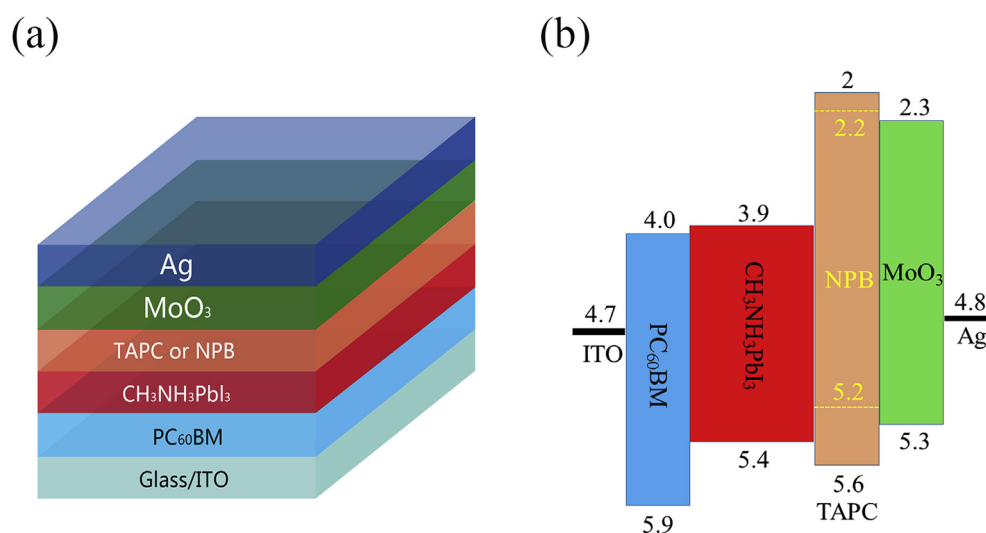


Fig. 1. (a) Device structure and (b) energy level diagram of the photodetectors.

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