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# Improved performance of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based photodetector with a MoO<sub>3</sub> interface layer

Ye Wang <sup>a, b</sup>, Qiaogang Song <sup>a, b</sup>, Tong Lin <sup>a, b</sup>, Yue Fu <sup>a, b</sup>, Xue Sun <sup>a, b</sup>, Bei Chu <sup>a</sup>, Fangming Jin <sup>a</sup>, Haifeng Zhao <sup>a</sup>, Wenlian Li <sup>a</sup>, Zisheng Su <sup>a, \*</sup>, Yantao Li <sup>a, \*\*</sup>

<sup>a</sup> State Key Laboratory of Luminescence and Applications, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun, 130033, PR China

<sup>b</sup> University of Chinese Academy of Sciences, Beijing, 100039, PR China

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# ABSTRACT

Planar CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite based photodetectors are fabricated by a facile and low-cost one-step method. The devices show broad spectral photoresponse from the ultraviolet to whole visible region and the performance can be significantly improved by the introduction of a bipolar transporting MoO<sub>3</sub> interface layer between CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film and Au electrode. The photocurrent of the device with an optimized MoO<sub>3</sub> layer is about twice that of the reference device without MoO<sub>3</sub> layer, which results in a high ON/OFF current ratio of  $5.9 \times 10^3$  at 5 V. Besides, slightly increased photoresponse speed is also found in the optimized device with rise time and decay time of 21.6 and 9.9 ms, respectively. The improvement can be attributed to the improved hole and electron collection efficiency and the quickly filled in or emptied trap states at the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/Au interface due to the introduction of the bipolar transporting MoO<sub>3</sub> layer.

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# 1. Introduction

Hybrid organolead halide perovskites,  $CH_3NH_3PbX_3$  (X = Cl, Br, I) have attracted significant interests as light absorbing materials in electronic and photonic applications recently [1-6], because of their excellent intrinsic optoelectronic properties, such as a direct bandgap, large absorption coefficient, high carrier mobility [5,7,8], long exciton diffusion length [9,10], and so forth. Furthermore, the series of CH<sub>3</sub>NH<sub>3</sub>PbX<sub>3</sub> perovskites can be fabricated by a facile and low-cost all-solution self-assembly method [5,8]. A variety of photodetectors based on hybrid organolead halide perovskites have been reported since 2014 [2]. Several research groups have successfully fabricated CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite based photodetectors with attractive features such as broad band, flexibility, and ultrasensitivity. For example, Hu et al. demonstrated the first organolead halide perovskite based broadband photodetector in 2014 [1]. Zhang and coworkers reported a novel ultrasensitive photodetector  $(I_{\text{light}}/I_{\text{dark}} > 10^4 \text{ at a bias of 30 V under an incident light of}$  ~ $6.59 \text{ mW/cm}^2$ ) based on unique CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite films with island-structured morphology in 2015 [11]. Other researchers have also explored the photodetector applications based on CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite [2,12,13].

The property at the perovskite/electrode interface plays an important role in determining the performance of a photodetector. In a planar type photodetector, the anode and cathode are synchronously deposited with the same metal material. Zhao et al. have systematically investigated the effects of the electrode on the performance of the perovskite photodetectors [14]. However, there are no reports about modification of the perovskite/electrode contact by introducing interface layer. In order to match with the fabrication process of photodetector, this interface layer should be a bipolar transporting material to facilitate not only the hole but also the electron collection. MoO<sub>3</sub> is non-poisonous and stable in ambient condition, and has been considered as a kind of promising material as interface layer of photoelectric devices, such as organic solar cells, organic light emitting diodes, and organic field-effect transistors [15–17]. Besides, MoO<sub>3</sub> is a kind of bipolar transporting material, and this feature makes it more attractive as interface layer for perovskite photodetectors. In this work, we have improved the performance of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite photodetectors by introducing MoO<sub>3</sub> between CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film and Au





<sup>\*</sup> Corresponding author.

<sup>\*\*</sup> Corresponding author.

E-mail addresses: suzs@ciomp.ac.cn (Z. Su), liyt@ciomp.ac.cn (Y. Li).

electrode. It is observed that the photoconductive performance is dependent on the thickness of MoO<sub>3</sub> layer. The device with an optimized MoO<sub>3</sub> layer thickness of 7.5 nm achieves an about two times ON/OFF ratio (~ $5.9 \times 10^3$  at a bias of 5 V) of the reference device without any interlayer. Besides, the device exhibits a fast photoresponse speed with rise and decay time of about 21.6 and 9.9 ms, respectively.

## 2. Experimental section

#### 2.1. Materials

All chemicals were purchased commercially and used as received.  $CH_3NH_3PbI_3$  precursor solution was synthesized by mixing  $CH_3NH_3I$  and lead iodide (PbI<sub>2</sub>) at 1:1 (0.162: 0.462 g) equimolar ratio in *N*,*N*-dimethylformamide (DMF, 0.7 ml) and dimethyl sulfoxide (DMSO, 0.3 ml) at 70 °C, stirring for 6 h inside a nitrogenfilled glove box.

#### 2.2. Device fabrication

Glass substrates were cleaned by ultra-sonication in acetone, ultrapure water, and isopropanol for 30 min, respectively, and then dried naturally. After that they were treated in an ultraviolet-ozone chamber for 10 min before transferred to the glove box for deposition of  $CH_3NH_3PbI_3$  perovskite layer. The devices were fabricated by spin-coating of the as-synthesized  $CH_3NH_3PbI_3$  precursor solution onto the pre-cleaned glass substrate at a rate of first 500 rpm for 5 s and then 2000 rpm for 60 s. After 40 s delay, 800  $\mu$ l diethyl ether was quickly added during the spin-coating procedure. The substrates were then heated at 70 °C and 105 °C for 2 min and 10 min, respectively. During the annealing process, the color of the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film changed from light yellow to black, indicating that the ABO<sub>3</sub> structure was gradually formed at high temperature during the evaporation of the DMF and DMSO solvents. MoO<sub>3</sub> layer and Au electrodes were thermally deposited onto CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film in a vacuum chamber at a pressure of ~3 × 10<sup>-4</sup> Pa through a shadow mask, defining the channel length and width of 1000 and 60 µm, respectively.

#### 2.3. Device characterization

X-ray diffraction (XRD) pattern was measured with a diffractometer (Rigaku D/Max- 2500) using Cu K $\alpha$  radiation ( $\lambda = 1.54056$  Å). The current-voltage (I-V) characteristics were measured using a programmable source meter (Keithley 2400) in the dark and under illumination of 100 mW/cm<sup>2</sup> with AM 1.5G simulated solar spectrum from a solar simulator (Newport 94023A). Scanning electron microscopy (SEM) images were measured on a Hitachi S4800. Absorption spectrum of the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film on a glass substrate was recorded with a Shimadzu UV-3101PC spectrophotometer. The external quantum efficiency (EQE) data was measured with a Stanford SR803 lock-in amplifier under monochromatic illumination at a chopping frequency of 130 Hz by a Stanford SR540 chopper. All measurements were performed under ambient condition and at room temperature.

#### 3. Results and discussion

The surface morphology and cross-sectional image of the  $CH_3NH_3PbI_3$  film on a glass substrate are shown in Fig. 1a and b. The



Fig. 1. (a) Surface morphology, (b) Cross-sectional image, (c) XRD pattern, and (d) absorption spectra of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film on a glass substrate.



Fig. 2. (a) Schematic device structures and (b) energy level diagram of the photodetectors. (c) SEM image of the fabricated whole device.

grain size of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> ranges from about 100 to 500 nm, and the large grain size can reduce the grain boundary and hence increase charge carrier collection efficiency. The cross section image shows that the thickness of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film is about 690 nm. Fig. 1c depicts the XRD pattern of the prepared CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite film. The XRD of as-deposited film has peaks at 14.14°, 20.05°, 28.45°, 31.92°, 40.71°, and 43.25°, which can be indexed to the reflections from (110), (112), (220), (310), (224), and (314) crystal planes, respectively, indicating a tetragonal perovskite structure with a = 8.855 Å and c = 12.659 Å, which is consistent with literature reported [18]. The CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film shows a broad absorption band from the ultraviolet to whole visible region with an edge of about 780 nm (Fig. 1d), corresponding to a bandgap of 1.59 eV.

Based on the as-synthesized CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film, planar photodetectors with two kinds of structures are fabricated, as shown in Fig. 2a. Here, CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> film serves as the photosensitive material to absorb light. Au electrodes can collect photogenerated charge carriers, and MoO<sub>3</sub> layer is able to improve the contact at the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/Au electrode interface. From the SEM image of the whole device shown in Fig. 2c, it can be seen that the width of the gap between the two Au electrodes is about 60  $\mu$ m.

Fig. 3 depicts the I-V characteristics of the photodetectors in the dark and under one sun (100 mW/cm<sup>2</sup>) AM 1.5G simulated solar illumination. Both the devices with and without MoO<sub>3</sub> layer have dark current on the order of  $10^{-10}$  A. This indicates that the

introduction of MoO<sub>3</sub> layer has little effect on the dark current of the devices. All the I-V curves of the devices present nearly linear dependence on applied voltage under illumination, indicating the formation of Ohmic contact between CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> or CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/ MoO<sub>3</sub> and Au electrodes, which demonstrates the photoconductive nature of the as-fabricated photodetectors. That is, the device can be considered as an insulator under dark circumstance, and the photocurrent arises dramatically upon illumination. Here, the photocurrent can be attributed to the separation of electron-hole pairs generated by the incident photons with energy equal or larger than the band gap of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>. It is worth noting that the photocurrent of the devices with different thickness of MoO<sub>3</sub> layer exhibits significant difference. The photocurrent of the reference device without MoO\_3 layer is about 5.48  $\times$   $10^{-7}$  A at 5 V, corresponding to an ON/OFF ratio of about  $2.7 \times 10^3$  as compared with its dark current ( $2.0 \times 10^{-10}$  A). The photocurrent of the optimized device with 7.5 nm MoO<sub>3</sub> layer increases to  $1.24 \times 10^{-6}$  A with the similar dark current of  $2.1 \times 10^{-10}$  A, corresponding to an ON/OFF ratio of about  $5.9 \times 10^3$ , which is about twice that of the reference device. The ON/OFF ratio of our optimized device is among one of the highest values reported for planar CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite based photodetectors [1,11,12,19]. It is also found that although the MoO<sub>3</sub> layer has a little effect on the dark current of the devices, the photocurrents are significantly improved under illumination. As mentioned above, the device can be considered as an insulator under dark circumstance. Here, the dark current is determined by



**Fig. 3.** I-V curves of the photodetectors with different thickness of  $MoO_3$  layer under dark and solar simulator irradiation with the white light intensity of 100 mW/cm<sup>2</sup>.



Fig. 4. Responsivity and detectivity of the  $\rm CH_3NH_3PbI_3$  photodetector with a 7.5 nm  $\rm MoO_3$  layer.



Fig. 5. Photoresponse behaviours of the CH<sub>3</sub>NH<sub>3</sub>Pbl<sub>3</sub> (black line) and CH<sub>3</sub>NH<sub>3</sub>Pbl<sub>3</sub>/MoO<sub>3</sub> (red line) photodetectors of (a) six cycles and (b) one cycle. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the bulk property of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> layer. By contrast, the device exhibits a typical photoconductive mode upon illumination, and the interface of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/Au plays an important role in determining the photocurrent. The introduction of MoO<sub>3</sub> layer significantly improved the charge carrier collection efficiency and hence increases the photocurrent.

The energy level diagram of the device is shown in Fig. 2b with the data quoted from literature [20,21]. It can be seen that the valence and conduction band of MoO<sub>3</sub> are 9.6 and 6.3 eV, respectively, while the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> are 5.4 and 3.9 eV, respectively. Although the valence band of MoO<sub>3</sub> is much lower than the HOMO level of CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>, the holes can be extracted via the gap states in MoO<sub>3</sub> layer, because the vacuum level of MoO<sub>3</sub> can shift towards higher energy due to the interfacial dipoles at MoO<sub>3</sub>/Au interface [20]. On the other hand, the electrons can be extracted through the conduction band of MoO<sub>3</sub>. Therefore, the introduction of MoO<sub>3</sub> layer facilitates both the hole and electron collection and thus increases the photocurrent of the devices under illumination. When the thickness of MoO<sub>3</sub> layer further increased to 10 nm, as shown in Fig. 3, the photocurrent decreased by about 20%. With the thickness of MoO<sub>3</sub> layer increasing, the series resistance of the device increase, which will decrease the charge carrier collection efficiency of the device.

The responsivity (*R*) of a photodetector can be defined as: [22].

$$R = \Delta I / PS \tag{1}$$

where  $\Delta I (\Delta I = I_{photo} - I_{dark})$  is the difference between the photocurrent and dark current, *P* is the incident light intensity, and *S* is the effective illumination area. The detectivity (*D*<sup>\*</sup>) can be expressed by the relationship if the shot noise from the dark current is the major contribution: [23].

$$D^* = R / \sqrt{2qJd} \tag{2}$$

where *q* is absolute electron charge and  $J_d$  ( $J_d = I_{dark}/S$ ) is dark current density. The *R* and  $D^*$  of the photodetector with 7.5 nm MoO<sub>3</sub> layer at 1 V are illustrated in Fig. 4. Here, the *R* is calculated from the EQE spectrum. The device shows a broad photoresponse from 350 to 800 nm and the *R* begins to decrease from about 750 nm, which is consistent with the absorption spectrum of the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> layer shown in Fig. 1d. Correspondingly, the  $D^*$  of the device in the whole visible region is higher than  $10^{12}$  Jones with the maximum of  $1.71 \times 10^{13}$  Jones at 755 nm. This  $D^*$  is among one of the highest values reported for perovskite photodetectors [2,24–26].

Photoresponse speed is another key factor for evaluating photodetector performance. To quantitatively extract the photoresponse constant, the well accepted quantitative criterion is introduced. That is, the rise and decay time are defined as the time taken for the initial current to increase to 90% of the peak value, or vice versa, respectively. We studied the transient photoresponse behavior of the devices with the illumination switched on and off at the bias voltage of 1 V, as shown in Fig. 5a. An enlarged typical ON/ OFF circle is shown in Fig. 5b. According to the criterion we mentioned above, the rise and decay time of the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based reference device are about 22.3 and 11.4 ms, respectively, which is comparable with the best performance of planar CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> photodetectors reported elsewhere. While the rise and decay time of the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>/MoO<sub>3</sub> (7.5 nm) photodetector are about 21.6 and 9.9 ms, respectively. The density of the electronic trap states at the

#### Table 1

Comparison of the photoresponse speed between the reported and our photodetectors.

Active material	Rise time (s)	Decay time (s)	Ref.
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> thin film	<0.1 <sup>a</sup>	<0.1 <sup>c</sup>	[1]
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /TiO <sub>2</sub>	~0.02 <sup>a</sup>	~0.02 <sup>e</sup>	[12]
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> island	<0.05 <sup>b</sup>	<0.05 <sup>d</sup>	[11]
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /rGO	0.0409 <sup>a</sup>	0.0288 <sup>c</sup>	[30]
CH <sub>2</sub> NH <sub>2</sub> PbI <sub>2</sub> /NCG	<0.025 <sup>a</sup>	<0.025 <sup>c</sup>	[31]
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> thin film	~0.0223 <sup>a</sup>	~0.00114 <sup>c</sup>	This work
CH <sub>3</sub> NH <sub>3</sub> PbI <sub>3</sub> /MoO <sub>3</sub>	~0.0216 <sup>a</sup>	~0.0099 <sup>c</sup>	This work

<sup>a</sup> Rise up to 90%.

<sup>b</sup> Increase from 10% to 90%.

<sup>c</sup> Fall down to 10%.

<sup>d</sup> Decrease from 90% to 10%.

<sup>e</sup> Reach 1/*e* of the original photocurrent.

semiconductor/metal interface is one of the factors that determining the response speed of the photodetector [27]. Thus, the slightly increased photoresponse speed can be attributed to the quickly filled in or emptied trap states when the light is turned on or turned off due to the existence of MoO<sub>3</sub> interface layer [28,29]. A comparison of photoresponse speed between reported and our CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based photodetectors are listed in Table 1. It can be found that the response speed of our photodetector is the fastest among these devices [1,11,12,30,31].

#### 4. Conclusion

In conclusion, CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite based photodetectors are fabricated by a facile and low-cost one-step method. The prepared CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> photodetector with an optimized bipolar transporting MoO<sub>3</sub> interface layer (7.5 nm) exhibits enhanced performance on the ON/OFF ratio, responsivity, and photoresponse speed. The ON/ OFF ratio of the optimized device is about two times that of the reference device. At the same time, the rise and decay time change from 22.3 and 11.4 ms to 21.6 and 9.9 ms, respectively, which is the fastest photoresponse speed among CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based photodetectors with planar structure. These results suggest that the introduction of MoO<sub>3</sub> layer to CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based photodetector is promising for enhancing the performance of the device. Furthermore, the mechanism understanding also provides opportunities for introducing interface layer to the CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> based photodetector for further study.

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