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Ultrasensitive all-solution-processed field-effect transistor based perovskite photodetectors with sol-gel SiO_2 as the dielectric layer

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ABSTRACT

In this work, we present a high-performance field-effect transistor (FET) based perovskite photodetector by using a sol-gel SiO_2 layer as the dielectric layer. The FET-based perovskite photodetector Al(Gate)/SiO_2/$CH_3NH_3PbI_3$/Au(Drain, Source) shows a high responsivity of 10.72 A/W and a high specific detectivity of 6.2×10^{13} Jones under 0.37 μW/cm2 532 nm laser, and the underlain mechanism is discussed. Therefore, this kind of all solution-processed method provides a promising way to fabricate high-performance perovskite-based optoelectronic devices on flexible substrates.

1. Introduction

Recently, hybrid organic-inorganic lead halide perovskites ($CH_3NH_3PbX_3$, where X = Cl, Br, and I) have been employed as ideal light absorption materials in optoelectronic devices such as light-emitting diodes (LEDs) [1], solar cells [2,3] and photodetectors [4–6] for their excellent optoelectronic properties. To achieve high-performance photodetectors, the active materials need high absorption extinction coefficient and large charge mobility (μ) to get sufficient light absorption and high photocurrent. Therefore, $CH_3NH_3PbI_3$, being with advantages of direct band gap, large absorption coefficient, high carrier mobility and long diffusion length, gets an excellent candidate as the active material applied in photodetectors [7,8]. Lee et al. [5] reported a field-effect transistor (FET) based perovskite photodetector with a high photoresponsivity of 180 A/W and a photodetectivity of about 10^9 Jones at an incident light power of 1 μW. In their cases, the active perovskite layer was spin-coated on a SiO_2/Si substrate as the outmost layer. Therefore, the active perovskite layer will be degraded after its exposing to the air directly. As we know, the moisture and oxygen in the air will accelerate the decomposition of perovskite [9–11]. Guo et al. [8] and Li et al. [12] spin-coated a fluorous polymer and a polymethylmethacrylate (PMMA) as the protect-layer to keep perovskite layer away from the moisture and oxygen, respectively. However, the Si wafers used as the gate electrode in these devices are expensive as compared with other metal electrodes, and the SiO_2 layer was usually grown on silicon substrate by a thermally oxidizing process which needs a high reaction temperature of 900–1200 °C. Furthermore, the SiO_2/Si substrate in those cases, using as the dielectric layer and the gate electrode respectively, is not flexible and will hinder their development in flexible applications.

Therefore, in this paper, we present an all-solution-processed high-performance FET-based photodetector, in which a solution-processed sol-gel SiO_2 layer was used as the dielectric layer. In this way, the expensive Si wafer was replaced by a low-cost Al

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electrode as the gate electrode, and the active perovskite layer was isolated from the moisture and oxygen by the additional sol-gel SiO_2 layer. As a result, the FET-based perovskite photodetector exhibited a photoresponsivity of 10.72 A/W and a specific detectivity of 6.2×10^{13} Jones at 0.37 μW/cm2 532 nm laser. Therefore, such a kind of all-solution-processed method is very simple and low-cost at room temperature and it provides a promising way to fabricate FET-based perovskite photodetectors on flexible substrates.

2. Experimental

2.1. Materials

Lead (II) Iodide (PbI_2, >99.99%), methylamine (33 wt% in absolute ethanol) and hydroiodic acid (57 wt% in water) were purchased from Alfa Aesar. Tetramethyl orthosilicate (TMOS, 99%), methyltrimethoxysilane (MTMS, 97%), formic acid (>98%) and *N*,*N*-dimethylformamide (DMF, >99.99%) were purchased from Acros Organics. 1-propanol (>99%), isopropanol (>99.7%) and diethyl ether (>99%) were purchased from the Beijing Chemical Works. All chemicals were used as received.

2.2. Synthesis of $CH_3{-}Si(OCH_3)_3$

$CH_3{-}Si(OCH_3)_3$ was synthesized by following a previously reported procedure [13]. Firstly, 15.86 g of tetramethyl orthosilicate (TMOS) and 14.2 g of methyltrimethoxysilane (MTMS) were mixed in a polypropylene jar, and then it was followed by injecting 6 g of 1-propanol. Then 13.2 g of 2 M formic acid in water was added in. The mixture was reacted at 36 °C for 120 min in a water bath. After that, 20.6 g of water and 184.8 g of 1-propanol were added to dilute the hydrolysis mixture. Finally, the water and alcohol in the mixture were removed at 50 °C for 30 min by rotary evaporators (~0.1 Mpa).

2.3. Synthesis of CH_3NH_3I

CH_3NH_3I was synthesized according to a previously reported procedure [14]. Firstly, methylamine (33 wt% in absolute ethanol) and hydroiodic acid (57 wt% in water) were mixed at 1:1 M ratio in a round-bottom flask at 0 °C for 2 h stirring. After that, the resulting solution was evaporated at 50 °C for 1 h by a rotary evaporator to crystallize CH_3NH_3I. The product was washed with diethyl ether by stirring the solution for 30 min, which was repeated three times. Finally, the product was dried overnight at 60 °C in a vacuum oven.

2.4. Device fabrication

Fig. 1 shows the schematic structure of the FET-based perovskite photodetector. Firstly, deionized water was used to clean the glass substrates and followed by ultrasonic treatment in isopropanol, acetone and ethanol for 15 min, sequentially. Then Au interdigitated metal electrodes (with 100 μm channel length) were evaporated on the substrates through a shadow mask [15]. For the fabrication of $CH_3NH_3PbI_3$ film, PbI_2 were dissolved in *N*,*N*-dimethylformamide with a concentration of 462 mg/ml and stirred for 12 h at 50 °C. Then the solution was spin-coated on Au electrode at 4000 rpm for 15 s and dried at 70 °C for 10 min CH_3NH_3I were dissolved in isopropanol with a concentration of 10 mg/ml. The solution were dropped onto the PbI_2 film and stayed for 30 s. Then it was spin-coated at 6000 rpm for 5 s (~300 nm) and dried at 70 °C for 30 min. After cooling down, $CH_3{-}Si(OCH_3)_3$, which was dissolved in methylbenzene with a volume ratio of 2:1, was spin-coated on the $CH_3NH_3PbI_3$ film and then the substrate was dried at 50 °C for 5 min, and then it became a SiO_2 dielectric layer. Finally,

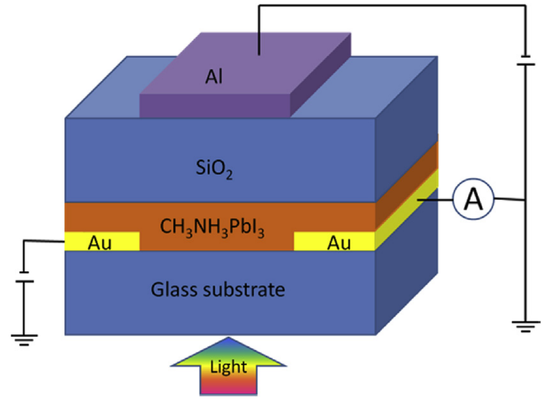

Fig. 1. Schematic diagram of the FET-based perovskite photodetector Al(Gate)/SiO_2/$CH_3NH_3PbI_3$/Au(Drain, Source) and the circuit for its characterization. The device is illuminated from the bottom.

the Al gate electrode was fabricated by thermal evaporation through a shadow mask. The device fabrication was conducted under nitrogen environment in the glove box.

3. Results and discussions

Fig. 2(a) shows the absorption spectra of the $CH_3NH_3PbI_3$ film, showing a good light-harvesting capability in visible region since there is a strong absorption for the incident wavelength from 380 nm to 780 nm. X-ray diffraction (XRD) were used to investigate the microstructure of the perovskite film, and the inset shows the XRD pattern (red line) of the as-prepared $CH_3NH_3PbI_3$ film and

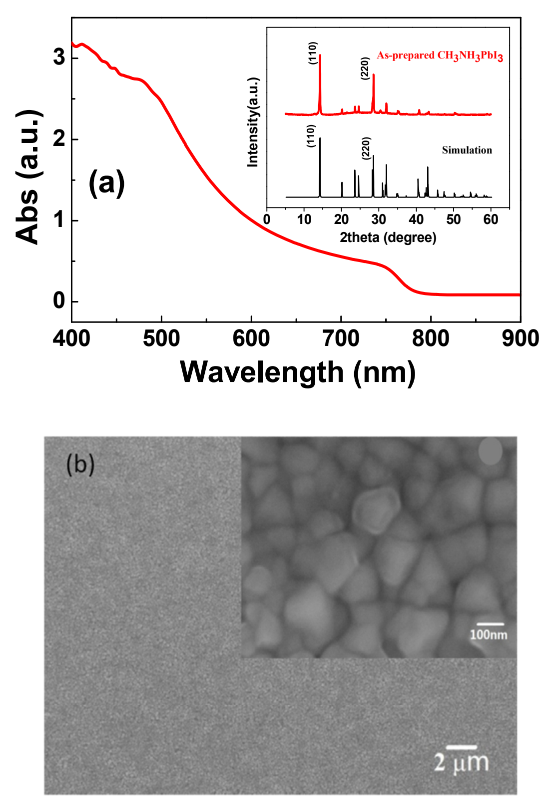

Fig. 2. (**a**) Absorption spectrum of $CH_3NH_3PbI_3$ film, and the inset shows its XRD spectra and the theoretically simulated XRD spectra of tetragonal $CH_3NH_3PbI_3$ single crystal; (**b**) SEM image of $CH_3NH_3PbI_3$ film surface, and the inset shows its HR-SEM.