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# Improvement of electron/hole injection balance based on nanowire/nanocube hybrid perovskite

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## ABSTRACT

The methyl ammonium lead iodide perovskite nanowire/nanocube can be fabricated with an improved two-step spin-coating process by adjusting the amount of dimethyl formamide. The hybrid structure enhanced the efficiency of electron/hole separation and charge transfer at the electron transport layer/perovskite/hole transport layer interfaces, benefiting from the improvement of electron/hole injection balance. The dynamic time parameters based on the half-baked cells, only containing electron transport layer or the hole transport layer, could assert rationality of the hypothesis by photoluminescence decays.

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## 1. Introduction

Recently, the hybrid organic/inorganic lead halide perovskite (e.g.  $\text{CH}_3\text{NH}_3\text{PbX}_3$ ,  $X=\text{I}, \text{Br}, \text{Cl}$ ) have been recognized as the most promising materials in fabricating solar cells due to its high absorption efficiency, long diffusion length, and ambipolar semiconductor [1]. The power conversion efficiency (PCE) of the device has been enhanced from 3.8% to 20.1% rapidly in the past 6 years [2,3]. Several facile synthesis of perovskite, including the one-step or two-step spin-coating techniques, were applied to prepare the active layer between the electron transport layer (ETL) and the hole transport layer (HTL) [4,5]. Under illumination, the photo-generated carriers were induced from the perovskite film and then selectively separated to the ETL and HTL. The PCE of perovskite devices might greatly depend on charge separation and transfer efficiency at the ETL/perovskite and/or perovskite/HTL interfaces. The ETL/perovskite interfacial engineering could be chosen to reduce interfacial barriers and optimize a charge transport pathway to achieve a high PCE [6]. For the same reason, the hole accumulation at the perovskite/HTL interface could also be solved by the interfacial engineering [7,8]. Therefore, the interfaces of ETL/perovskite/HTL could be optimized to maintain charge balance and gain high photocurrent.

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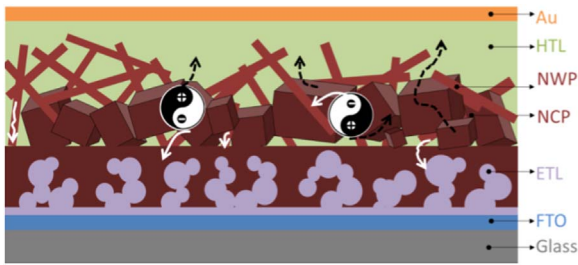
E-mail address: [yqwang@seu.edu.cn](mailto:yqwang@seu.edu.cn) (Y. Wang).

It is well-known that the size, orientation and phase could have a great effect on the photoelectrochemical properties. For instance, the perovskite nanowire and nanorod could improve the photo-to-electron or electro-to-optical conversion efficiency in the optoelectronic devices, such as a semiconductor laser and light-emitting diode [9,10].  $\text{CH}_3\text{NH}_3\text{PbI}_3$  nanowire based solar cells have achieved a PCE of 14.71% because of their effective electron/hole separation [11]. Compared with  $\text{CH}_3\text{NH}_3\text{PbI}_3$  nanocubes, the nanowires exhibit higher external quantum efficiency, longer carrier lifetime, and lower dark current [12]. To achieve an effective carrier transport in the ETL/perovskite/HTL interfaces, we report solar cell composed of nanowire/nanocube hybrid perovskite by controlling the ratio of N,N-Dimethylformamide (DMF) in the  $\text{CH}_3\text{NH}_3/\text{isopropanol}$  solution. The addition of nanowires into nanocubes could improve the injective balance of the electron/hole. (Fig. 1).

## 2. Experimental section

### 2.1. Device assembly

All the experimental procedures were carried out in an improved glove box with a humidity of about 20%. FTO glasses ( Pilkington TEC-8,  $8\ \Omega\ \text{sq}^{-1}$ ) were cleaned by distilled water, ethanol and isopropanol for 3 times each.  $\text{TiO}_2$  blocking layer was deposited by spray pyrolysis [4] and the 20 nm-sized  $\text{TiO}_2$  particles were spin-coated and calcined at 500 °C for 30 min.  $\text{PbI}_2$  (934 mg)



**Fig. 1.** Schematic diagram of charge separation and transfer in the nanowire/nanocube hybrid perovskite solar cell.

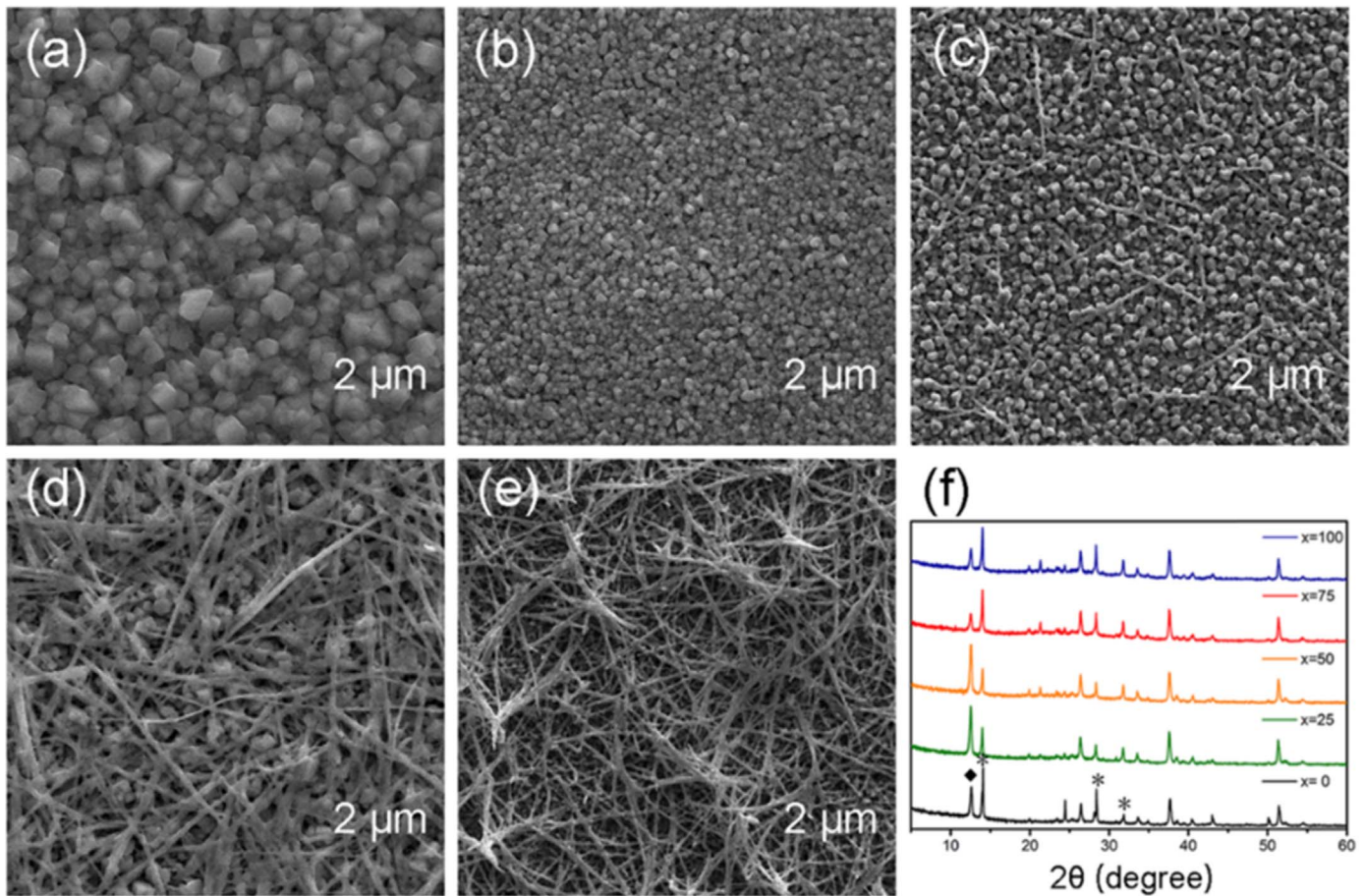
was dissolved in 2 mL of DMF (Sinopharm) and heated at 70 °C, then the  $\text{PbI}_2$  was spin-coated onto the  $\text{TiO}_2$  film at a speed of 2000 rpm for 5 s and 6000 rpm for 5 s. After the  $\text{PbI}_2$  substrate stood for 90 s,  $\text{CH}_3\text{NH}_3\text{I}$  solution (300  $\mu\text{L}$ , 7 mg/mL) including X- $\mu\text{L}$  of DMF ( $X=0, 25, 50, 75, 100$ ) was loaded for 1 min which was spun at 4000 rpm for 30 s and then dried at 100 °C for 5 min. Next, 50  $\mu\text{L}$  of spiro-MeOTAD solution was spin-coated on the  $\text{CH}_3\text{NH}_3\text{PbI}_3$  film at 2000 rpm for 30 s. A spiro-MeOTAD solution was prepared by dissolving 122.5 mg of spiro-MeOTAD in 1 mL of chlorobenzene, to which 28.8  $\mu\text{L}$  of 4-tert-butylpyridine, 17.5  $\mu\text{L}$  of lithium bis(trifluoromethanesulfonyl) imide (Li-TFSI) solution (520 mg in 1 mL of acetonitrile (Sinopharm)) was added. Finally, Au (80 nm) was thermal evaporated as counter electrode.

## 2.2. Characterization and measurement

Scanning electron microscopy (SEM) images were obtained by FEI inspect F50. UV–Vis absorption was measured with UV–vis spectro-photometer CARY-60 with wavelength ranging from 400 nm to 800 nm. The crystal structures of the perovskite films were characterized by X-ray diffraction (XRD Smartlab3). Transient fluorescence decays was measured with an HORIBA FM-4P-TCSPC spectrometer. Current-voltage (IV) and dark current curves were measured by Keithley 2400 under Newport Oriol 91,192 simulated illumination ( $\text{AM1.5}, 100 \text{ mW cm}^{-2}$ ) and in the dark, respectively.

## 3. Results and discussions

The morphology of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  can be easily tuned by adding polar aprotic solvent DMF into the  $\text{CH}_3\text{NH}_3\text{I}$  solution during spin-coating process. It was found a random  $\text{CH}_3\text{NH}_3\text{PbI}_3$  riprap was formed without DMF (Fig. 2(a)). However, the huge cuboids were transformed to tiny cuboids ( $\sim 100 \text{ nm}$ , Fig. 2(b)) if 25  $\mu\text{L}$  of DMF was added into  $\text{CH}_3\text{NH}_3\text{I}$  solution. It is significant that a small quantity of nanowires start to appear as DMF was increased to 50  $\mu\text{L}$  (Fig. 2(c)), and much more nanocubes were transformed to nanowires by adding 75  $\mu\text{L}$  of DMF (Fig. 2(d)). The formation of one-dimensional nanowire perovskite can be mainly contributed to the polar aprotic solvent DMF [11]. Moreover, all nanocubes could completely transform to nanowires when 100  $\mu\text{L}$  of DMF was added (Fig. 2(e)). According to the XRD patterns (Fig. 2(f)), the diffraction peaks at 14°, 28.4° and 31.8° can be assigned to (110),



**Fig. 2.** SEM images of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  prepared from the solution of  $\text{CH}_3\text{NH}_3\text{I}$ /isopropanol (35 mg/5 mL) with different DMF volume ( $\mu\text{L}$ ): (a) 0, (b) 25, (c) 50, (d) 75, (e) 100. (f) XRD patterns of the corresponding samples from (a) to (e). Scale bars: 2  $\mu\text{m}$ . \* $\text{CH}_3\text{NH}_3\text{PbI}_3$  (JCPDS No. 46-1045), ♦ indicates  $\text{PbI}_2$  (JCPDS No. 80-1000), and other peaks referring to  $\text{SnO}_2$  (JCPDS No. 41-1445) substrate.

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