



## Efficient organic ternary solar cells with the third component as energy acceptor

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### ABSTRACT

A smart ternary strategy was designed to simultaneously improve utilization of excitons on PCDTBT and enhance photon harvesting in long wavelength range by doping appropriate SQ into the dominating PCDTBT:PC<sub>71</sub>BM system. The phase separation can be finely optimized by adjusting SQ doping ratio, resulting in efficient charge carrier transport channels and the enhanced photon harvesting in the ternary active layers. The dopant SQ provides a potential route for further utilizing the excitons on PCDTBT, especially for the excitons unattainable to the PCDTBT/PC<sub>71</sub>BM interfaces. The excitons on PCDTBT can transfer their energy to SQ through Förster energy transfer, and then be dissociated into free charge carriers at SQ/PC<sub>71</sub>BM interface. The holes generated on SQ can be effectively transferred to PCDTBT and then transported along the channels formed by PCDTBT to anode. Consequently, the champion PCE of ternary solar cells arrives to 7.62% with 9 wt% SQ doping ratio in donors, which is much higher than the PCE of 6.54% for PCDTBT:PC<sub>71</sub>BM-based or 1.95% for SQ:PC<sub>71</sub>BM-based solar cells.

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### 1. Introduction

Solution-processed organic solar cells (OSCs) have gained rapid development during the past 10 years, which possess the unique properties of low cost, large-area fabrication, mechanical flexibility and synthetic variability [1–5]. To date, the state-of-the-art single junction OSCs have achieved the power conversion efficiency (PCE) of more than 10% based on the delicate material design and device engineering [6–9]. Though impressive, the further investigations on OSCs are still needed from materials and device physics to meet the commercial standard and generate renewable green energy. As we know, the PCE of solar cells is proportional to the product of short circuit density ( $J_{SC}$ ), open circuit voltage ( $V_{OC}$ ) and fill factor (FF). One of the fundamental challenges for single junction solar cells is the photon harvesting ability and range dependence on the optical band gap and energy levels of used materials, which will strongly influence the trade-off between  $J_{SC}$  and  $V_{OC}$ , named as Shockley-Queisser efficiency limit or thermalization losses [10]. Since the  $V_{OC}$  is mainly determined by the difference between the highest occupied molecular orbital (HOMO)

energy level of electron donor and the lowest unoccupied molecular orbital (LUMO) energy level of electron acceptor. It is important to achieve the more balanced trade-off between a narrow bandgap donor for high  $J_{SC}$  and a deep HOMO energy level for high  $V_{OC}$  while keeping enough driving force for exciton dissociation. Furthermore, the relatively narrow absorption window ( $\approx 100$  nm) of organic materials is another limitation for sufficient photon harvesting in binary single junction solar cells [11–13].

Tandem configuration solar cells can enhance the light absorption by stacking two or more sub-cells with complementary absorption range in the sub-cells, which can circumvent the Shockley-Queisser efficiency limit [14–16]. However, the extremely intricate fabrication technology results in enormous challenges to obtain highly efficient tandem solar cells with high rate of finished products, large scale and low cost [17–19]. Recently, organic ternary solar cells have attracted more and more attention due to their versatile characteristics, integrating the advantages of enhanced photon harvesting in tandem solar cells and simple processing conditions used in single junction solar cells [20–23]. For ternary solar cells, the active layer generally contains three components: the dominating donor:acceptor (D:A) system and the third component, where the third component can be electron donor [24,25], electron acceptor [26,27] or nonvolatile additive

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[28–30]. The main fundamental working mechanisms of ternary solar cells can be summarized as the followings: charge transfer [17,31,32], energy transfer [33,34], parallel linkage [35,36] and alloy structure [37–39], which are significantly different from between each other, as well as the mechanism of binary solar cells. There may be one or more mechanisms that govern the photo-voltaic process in one kind of ternary solar cells. More importantly, each mechanism has its own advantages and disadvantages [18,23]. It is vital important for ternary solar cells to synergistically employ the advantages of these mechanisms. In addition achieving an improved  $J_{SC}$  by enhancing photon harvesting, the  $V_{OC}$  and FF should be kept a decent value for improving the performance of ternary solar cells.

In this work, a series of organic ternary solar cells were fabricated with poly[N-9'-heptadecanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)];[6,6]-phenyl-C71-butyric acid methyl ester (PCDTBT:PC<sub>71</sub>BM) as dominating system and 2,4-bis[4-(N,N-diisobutylamino)-2,6-dihydroxyphenyl] squaraine (SQ) as the third component. The optimized ternary active layers show a broad photon harvesting range compared with the PCDTBT:PC<sub>71</sub>BM blend films and relatively strong absorption intensity compared with the SQ:PC<sub>71</sub>BM blend films in the short wavelength range. In the ternary solar cells, the collection of holes generated on SQ molecules can be impressively improved along the hole transport channels formed by PCDTBT. The exciton on PCDTBT can't arrive at the PCDTBT/PC<sub>71</sub>BM interfaces, which can transfer its energy to SQ through Förster energy transfer, and then be dissociated into free charge carriers at SQ/PC<sub>71</sub>BM interface. Consequently, the champion PCE of ternary solar cells arrives to 7.62% with 9 wt% SQ doping ratio in donors, which is higher than the PCE value of 6.54% for PCDTBT:PC<sub>71</sub>BM-based or 1.95% for SQ:PC<sub>71</sub>BM-based solar cells, respectively. Based on the high PCE of PCDTBT:PC<sub>71</sub>BM based binary solar cells, it is convincing that ternary strategy should be an efficient method to improve the performance of OSCs. The most important is that an efficient method is firstly developed to investigate charge transfer between donors and transport process in the ternary active layer by studying the  $J-V$  curves of OSCs covered with a cut-off filter.

## 2. Experimental

The patterned indium tin oxide (ITO) glass coated substrates (sheet resistance  $15 \Omega/\square$ ) were consecutively cleaned in ultrasonic baths containing detergent, de-ionized water and ethanol, respectively. The cleaned ITO substrates were blow-dried by high pure nitrogen gas and then treated by oxygen plasma for 1 min to improve its work function and clearance. The transmission spectra of ITO substrate and PEDOT:PSS/ITO were measured and are shown in Fig. S1. Subsequently, poly-(3,4-ethylenedioxythiophene): poly-(styrenesulphonicacid) (PEDOT: PSS) (clevios P VP Al 4083, purchased from H.C. Starck co. Ltd.) thin films were fabricated on the cleaned ITO substrates by spin-coating method at 5000 round per minute (RPM) for 40 s, and then annealed at 150 °C for 10 min in ambient conditions. Then the ITO substrates coated PEDOT:PSS films were transferred to a high-purity nitrogen-filled glove box to fabricate active layers. The materials PCDTBT and PC<sub>71</sub>BM (purchased from 1-Material and used as received), SQ (purchased from Luminescence Technology Corp. and used as received) were dissolved in chlorobenzene:1,2-dichlorobenzene solvent mixture (1:3 vol) to prepare 20 mg/ml blend solutions, respectively. The doping weight ratios of SQ in donors (PCDTBT and SQ) are 0, 3 wt%, 6 wt%, 9 wt%, 12 wt%, 15 wt%, 30 wt%, 50 wt%, 100 wt% and the weight ratio of donors to acceptor is kept constant as 1:4 (PCDTBT<sub>1-x</sub>:SQ<sub>x</sub>:PC<sub>71</sub>BM<sub>4</sub>, x represents SQ doping ratios in donors). The blend solutions were spin-coated on PEDOT:PSS films at 800 RPM for 40 s in a high purity nitrogen-filled glove box to fabricate

the active layers. The thickness of the active layers is  $\sim 80$  nm, which was measured by Ambios Technology XP-2 stylus Profiler. After that, the conjugated polyelectrolyte poly[9,9-bis(3'-(N,N-dimethylamino)-propyl-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) solutions (0.2 mg/ml in methanol with 0.25 vol% acetic acid) were spin-coated on the top of active layers at 3000 RPM for 30 s. The cathode of Al (100 nm) film was deposited by thermal evaporation with a shadow mask under  $10^{-4}$  Pa and the thickness was monitored by a quartz crystal microbalance. The active area is about  $3.8 \text{ mm}^2$ , which is defined by the vertical overlap of ITO anode and Al cathode. To give the more accurate active area of cells, the size of ITO and Al electrode was measured by Ambios Technology XP-2 stylus Profiler and is about 2 mm and 1.9 mm, as shown in Fig. S2. The device configuration of fabricated PSCs is ITO/PEDOT:PSS (40 nm)/Active layer (80 nm)/PFN (10 nm)/Al (100 nm). The chemical structure and absorption spectra of used materials, as well as the schematic diagram of cells are shown in Fig. 1.

The ultraviolet-visible (UV-vis) absorption spectra of neat and blend films were obtained using a Shimadzu UV-3101 PC spectrometer. The current-voltage ( $I-V$ ) curves of all OSCs were measured in ambient conditions using a Keithley 4200 source meter. AM 1.5G irradiation at  $100 \text{ mW/cm}^2$  provided by an ABET Sun 2000 solar simulator, which was calibrated by standard silicon solar cells (purchased from Zolix INSTRUMENTS CO. LTD). All the characterization of ternary solar cells were carried out in air conditions. The external quantum efficiency (EQE) spectra of OSCs were measured by a Zolix Solar Cell Scan 100. Photoluminescence (PL) spectra of neat and blend films were measured by a HORIBA Fluorolog<sup>®</sup>-3 spectrofluorometer system. Time-resolved transient photoluminescence (TRTPL) spectra were obtained using a FluoroCube-01-NL and FluoroCub-NL from Jobin Yvon, the excitation light was provided by a NanoLED-460 with pulse width  $< 1.3 \text{ ns}$  and power  $7 \text{ pJ/pulse}$ . The electrochemical cyclic voltammetry (CV) was conducted on an electrochemical workstation (CHI660D Chenhua Shanghai) with Pt plate as working electrode. The water contact angle images of neat and blend films were recorded by using a KRÜSS DSA 100 instrument under ambient conditions. The morphology of the active layers was investigated by atomic force microscopy (AFM) using a multimode Nanoscope IIIa operated in tapping mode. Transmission electron microscopy (TEM) images of the active layers were obtained by using a JEOL JEM-1400 transmission electron microscope operated at 80 kV.

## 3. Results and discussion

All the binary and ternary solar cells were fabricated and measured under the same conditions based on the structure of ITO/PEDOT:PSS/active layer/PFN/Al (see Experimental Section). The absorption spectra of used materials PCDTBT, SQ and PC<sub>71</sub>BM exhibit a complementary absorption in the range from 300 nm to 800 nm, which should be beneficial for photon harvesting in ternary solar cells. The absorption spectra of PCDTBT: SQ blend films with different SQ doping ratios are shown in Fig. 2a. It is apparent that the absorption intensity of the blend films is gradually enhanced in the range from 600 nm to 800 nm along with the increase of SQ doping ratios in donors. Meanwhile, the absorption intensity of blend films is slightly reduced in the range from 300 nm to 600 nm due to the decreased PCDTBT content in the blend films. The similar phenomenon was also observed in the absorption spectra of PCDTBT: SQ:PC<sub>71</sub>BM ternary blend films along with the increase of SQ doping ratios in donors, as shown in Fig. S3. Fig. 2b shows current density-voltage ( $J-V$ ) curves of all cells under AM 1.5G light illumination with an intensity of  $100 \text{ mW/cm}^2$ . The key photovoltaic parameters were summarized and are listed in Table 1 according to the  $J-V$  curves. It is obvious that the PCE values of ternary solar cells are continuously increased along with SQ doping ratios up to 9 wt% and then

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