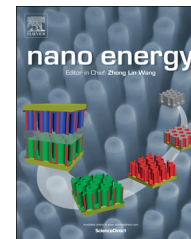


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## RAPID COMMUNICATION

# From binary to multicomponent photoactive layer: A promising complementary strategy to efficient hybrid solar cells

Changwen Liu<sup>a,b</sup>, Zeliang Qiu<sup>a,b</sup>, Feng Li<sup>c</sup>, Weili Meng<sup>a,b</sup>,  
 Wenjin Yue<sup>a,b</sup>, Fapei Zhang<sup>c</sup>, Qiquan Qiao<sup>d</sup>, Mingtai Wang<sup>a,b,\*</sup>

<sup>a</sup>Institute of Plasma Physics, Chinese Academy of Sciences, Hefei 230031, PR China

<sup>b</sup>Key Lab of Novel Thin Film Solar Cells, Chinese Academy of Sciences, Hefei 230031, PR China

<sup>c</sup>High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, PR China

<sup>d</sup>Center for Advanced Photovoltaics, Department of Electrical Engineering and Computer Sciences, South Dakota State University, Brookings, SD 57007, USA

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## Abstract

A strategy is demonstrated for fabrication of highly efficient hybrid solar cells based on the polymer/nanoarrays with complementary multicomponents in photoactive layer, including a scenario to controllably synthesize ternary ZnO/CdS/Sb<sub>2</sub>S<sub>3</sub>-core/shell/shell nanoarrays (ZCS-NAs) for a high open-circuit voltage ( $V_{oc}$ ) and short-circuit current and an approach to dope amorphous polymer with lithium bis(trifluoromethanesulfonyl) amide at nanoscale for a remarkably improved fill factor. With the integrated benefits from the complementary multicomponents having optimized nanoarray structure and doping concentration, an efficiency up to 5.01% under AM 1.5 illumination (100 mW/cm<sup>2</sup>) is achieved in the polymer/ZCS-NA devices with poly(2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene) as the polymer. To the best of our knowledge, this is the highest efficiency in the polymer/nanoarray devices. It is found that the  $V_{oc}$  in the multicomponent solar cells is determined by the band level difference between ZnO core and polymer, and sufficient photo-excitation of the polymer is necessary for efficient photocurrent generation. The component effects on device performance are elucidated and a model concerning the effective polymer phase and illumination attenuation between nanorods is proposed for understanding the charge generation from polymer absorption in the multicomponent solar cells.

\*Corresponding author at: Institute of Plasma Physics, Chinese Academy of Sciences, Hefei 230031, PR China.  
 Tel./fax: +86 551 65593171.

E-mail address: [mtwang@ipp.ac.cn](mailto:mtwang@ipp.ac.cn) (M. Wang).

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## Introduction

Hybrid polymer-based solar cells (HPSCs) using a photoactive layer formed by a conjugated polymer as electron donor (D) and inorganic nanostructure as electron acceptor (A) have shown promises in pursuit of cost-effective photovoltaic devices [1-6], due to their particular advantages of integrating the properties of organic polymer (good flexibility, light-weight, and easy-film formation) and inorganic nanostructures (high electron mobility, high electron affinity, and good stability). In principle, the polymer absorbs photons to generate excitons (bound electron-hole pairs); the photogenerated excitons diffuse to the D/A interface for dissociation into free charge carriers (electrons and holes) that are transported within D and A components to the respective electrodes for photocurrent generation. The photoactive layer in HPSCs is often structured into planar and bulk architectures. The D and A components are sequentially deposited on top of each other in the planar architecture, providing bilayer heterojunction devices with a single and two-dimensional D/A interface. However, nanostructured A component is dispersed in polymer matrix in the bulk architecture, resulting in bulk-heterojunction (BHJ) solar cells with three-dimensionally distributed D/A interfaces. With the use of BHJ architecture, a high D/A interface area for exciton dissociation is created, and the problem due to the limited exciton diffusion length ( $L_D$ ) in common conjugated polymers, e.g.,  $L_D < 10$  nm in poly(2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylene vinylene) (MEH-PPV) [7-9] and poly(3-hexylthiophene) (P3HT) [10,11], is addressed by nanoscale phase separation that results in a bicontinuous interpenetrating network of D and A phases in photoactive layer. The power conversion efficiency ( $\eta$ ) of around 5% has been achieved in the polymer/CdTe devices with combined bilayer and BHJ architectures [12] and the P3HT/TiO<sub>2</sub> bulk devices fabricated by infiltrating P3HT into TiO<sub>2</sub>/Sb<sub>2</sub>S<sub>3</sub> coaxial nanowire arrays [13]. Seok and co-workers [14-16] have achieved the efficiencies of 5.13%, 6.18% and 6.3% in the BHJ devices based on Sb<sub>2</sub>S<sub>3</sub>-sensitized mesoporous TiO<sub>2</sub> films in combination with P3HT, poly(2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta [2,1-b;3,4-b']dithiophene)-alt-4,7(2,1,3-benzothiadiazole)) (PCPDTBT), and PCPDTB T-PCBM ([6,6]-phenyl-C61-butyrac acid methyl ester) blend, respectively. However, the disordered electron transport formed by nanoparticles may cause a serious charge recombination; in particular, the nanomorphology formed by the simple blend of polymer and nanoparticles will encounter a low thermal stability under illumination.

Replacing disordered A-phase pathways formed by nanoparticles with vertically aligned nanorod/nanowire arrays offers the way to prepare the HPSCs with an ideal BHJ architecture for long-term application [1-6,17,18]. The nanoarray devices have a high area and stabilized spatial distribution of D/A interfaces for exciton dissociation, and the straightforward and interdigitated nanochannels for electron transport. Zinc oxide nanorod/nanowire arrays (ZnO-NAs) are widely used in such aligned BHJ devices [17-22], due to their facile synthesis and environment-friendly characteristics [23-25]. Incorporating a

conjugated polymer into ZnO-NAs provides the polymer/ZnO-NA solar cells with binary components (i.e., polymer and ZnO) in photoactive layer. However, the  $\eta$  in the polymer/ZnO-NA devices is not high enough yet ( $\eta=0.2-0.5\%$ ). The efficiency of a solar cell is determined by the relationship  $\eta=J_{sc} \times V_{oc} \times FF/P_{in}$ , where  $J_{sc}$  is the short-circuit current density,  $V_{oc}$  is the open-circuit voltage, FF is the fill factor, and  $P_{in}$  is the incident light power density. The polymer/ZnO-NA devices with MEH-PPV and P3HT are suffering from a small  $V_{oc}$  ( $\sim 0.1$  to  $0.5$  V) and a narrow polymer absorption spectrum (400–600 nm) for a low  $J_{sc}$  [19-22].

The structural characteristics at the D/A interface governing charge generation and recombination are crucially important. Interfacial modification has been attempted to improve the performance in polymer/ZnO-NA devices. Modification of ZnO-NA with amphiphilic organic molecules can enhance the  $J_{sc}$  to a certain extent as a result of the improved compatibility between polymer and ZnO, but has a limited effect on increasing  $V_{oc}$  [26-29]. Moreover, modification of ZnO-NA with inorganic semiconductors can enhance  $V_{oc}$  up to above 0.8 V, but has an unsatisfactory improvement in  $J_{sc}$  [30-34]. Notably, these organic [28,29] or inorganic [32,33] modifications often do not improve remarkably the device FF ( $\sim 30\%$  to  $40\%$ ); in particular, the improved  $J_{sc}$  from those modifications is still limited to the scale of several mA/cm<sup>2</sup>. Reasonably, the binary nanoarrays (i.e., modifier and ZnO) and the resulting ternary photoactive layer (binary nanoarray and polymer) obtained from the modifications with an organic or inorganic modifier mainly improve either  $J_{sc}$  or  $V_{oc}$ . Therefore, it still remains challenging to get a significantly high  $V_{oc}$  and  $J_{sc}$ , as well as FF, in an individual device for a breakthrough at the efficiency of the polymer/ZnO-NA solar cells.

We previously coated the ZnO nanorods in ZnO-NA with a polycrystalline film of CdS quantum dots (QDs), and the resulting heterostructured ZnO/CdS-core/shell nanorod array (ZC-NA) generated the HPSCs with a  $V_{oc}$  up to 0.82 V, for which a not high efficiency of  $\eta=0.87\%$  is mainly due to a still rather low  $J_{sc}$  and FF [33]. Here we demonstrate, for the first time, a strategy to fabricate the efficient HPSCs based on polymer/ZnO-NA systems by using multicomponents with complementary properties in photoactive layer. Our approach includes the controllable synthesis of ternary core/shell/shell nanoarrays for a high  $V_{oc}$  and  $J_{sc}$ , and the nanoscale doping of conjugated polymer for a remarkably improved FF. The chemical processes for ternary nanoarray synthesis, the solar cell architecture and the charge transfer processes involved are depicted in Figure 1. As illustrated in Figure 1a, the preformed ZnO nanorods in ZnO-NA are first coated with a polycrystalline film of CdS-QDs as an inner shell via successive ion layer adsorption and reaction (SILAR) technique, producing a binary ZnO/CdS-core/shell nanorod array (i.e., ZC-NA); afterwards, a Sb<sub>2</sub>S<sub>3</sub> outer shell is formed by depositing Sb<sub>2</sub>S<sub>3</sub> nanocrystals on the CdS shell through a two-step process that involves the chemical bath deposition (CBD) and thermally induced crystallization of amorphous Sb<sub>2</sub>S<sub>3</sub>, providing finally a ternary ZnO/CdS/Sb<sub>2</sub>S<sub>3</sub>-

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