



Double-parallel-junction hybrid solar cells based on silicon nanocrystals



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ABSTRACT

A novel type silicon nanocrystal-based hybrid solar cell is demonstrated here, where two individual junctions are designed carefully and arranged in parallel with each other. It is found that complementary absorption can be realized by double parallel junctions, and more photons in a wide energy range can be absorbed. As a result, device efficiency has been enhanced more than twice compared to single junction reference device. In addition, its working principles are also studied extensively.

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1. Introduction

Concerning the global growing energy demand as well as increasing environmental problems, solar cells as one kind of low-cost and renewable energy devices have been attracting much interest and dramatic achievements have been realized in recent decades [1]. Up to date, numerous solar cells fabricated with different materials and in various structures have been proposed in order to further enhance their performance [2–7]. Among them, multijunction tandem solar cells present promising future, where several individual subcells with complementary absorption spectra are arranged in series [8,9]. Comparing to single junction device, they harvest a greater part of the solar spectrum [10,11]. The whole device has an identical photocurrent with each subcells, while the voltage is equal to the sum of all subcell voltages, yielding a dramatic efficiency promotion [12,13]. The main weakness of tandem device is that its fabrication process is relatively demanding because each subcell needs to be carefully designed in order to

match the light absorption spectrum along with photocurrent [14]. Similar to tandem solar cells, complementary absorption in a wide range also can be realized in polymer solar cells by blending three (or more) different donor and acceptor materials in the active region, which are commonly denoted as ternary (or polynary) polymer solar cells [15,16]. However, device performance have not been improved too much, conversely, device efficiency has deteriorated in many cases [17]. Moreover, several challenges need to be carefully addressed in order to thoroughly unravel the fundamental operating principles and to further design and improve such devices [18].

On the other hand, few works have been reported focusing on solar cells where several subcells are arranged in parallel. It is interesting to know whether and how to promote solar cell performance by arranging subcells in parallel. Hence, in this manuscript, a novel hybrid solar cell based on silicon nanocrystals (Si NCs) has been demonstrated, where a PTB7 and Si NCs bulk heterojunction is arranged in parallel with a PTB7 and PC₇₁BM bilayer heterojunction. It is found that similar to tandem solar cells, complementary absorption has been realized by these two parallel junctions and more photons can be converted to carriers. Therefore, comparing to single junction control device, solar cell efficiency has been improved dramatically in such double-parallel-junction (DPJ)

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hybrid solar cells, which mainly benefits from adding up photocurrent from both subjunctions. In addition, the working mechanisms of DPJ device are also extensively studied.

2. Experimental

2.1. Silicon nanocrystal fabrication

Si NCs have been employed in this work as have several merits which has been extensively discussed elsewhere [19–21]. Briefly speaking, they have promising potential in more efficient light-harvesting, exciton dissociation, carrier transportation, and so on; whilst keeping the superiorities which organic materials possess, such as flexible and solution-processable. Si NCs can be synthesized by several methods such as aqueous solution synthesis [22], laser ablation from bulk silicon [23], and so on [24]. In this work, free-standing Si NCs were fabricated by using very high frequency (VHF) non-thermal plasma as described in elsewhere [25–28]. A mixture of argon (Ar), hydrogen (H₂), and silicon tetrachloride (SiCl₄) gases with a certain flow ratio was employed as the reaction precursor. Nanocrystals synthesized in the plasma were collected downstream of the plasma on a mesh. As-produced Si NCs were then etched thoroughly with hydrofluoric acid (HF, 50%) vapour at room temperature, and finally, hydrogen terminated fresh Si NCs were transported in a glovebox (<1 ppm water and oxygen) for further use. Fresh Si NCs are single-crystallized and have an average size of 6 nm, although it is still difficult to observe quantum effect in such relatively large size particles, it would be possible in future works by reducing particle size. In addition, it works as n-type material as confirmed by fabricating thin film transistors (TFT) (see Fig. S1 in Supporting Information); some basic properties of fresh Si NCs are also illustrated in Fig. S1.

2.2. Solar cell fabrication

Device fabrication processes were conducted in glovebox. PTB7 was dissolved in chlorobenzene (CB) at a concentration of 10 mg/ml. Fresh Si NCs were also dispersed in CB at the same concentration. After homogenization, the Si NC solution was mixed with PTB7 solution in 1:1 volume ratio. Blended solution was stirred at 40 °C for 24 h before use.

For the control device, commercially available patterned ITO glass was used as the transparent electrode. It was rinsed sequentially with chloroform, acetone, isopropanol and ethanol, finally treated by UV-Ozone cleaner (Filgen, UV253-OZ) for 15 min. Filtered PEDOT:PSS (Clevios PH1000, Heraeus) was spin-cast on ITO and annealed at 130 °C for 15 min. PEDOT:PSS layer thickness was controlled to about 40 nm and works as the hole transporting layer (HTL) in device. Afterwards, the Si NC/PTB7 blend solution was spin-cast on the HTL at 1100 rpm to form an active layer with a thickness of ~120 nm; finally, a 100 nm-thick aluminium (Al) electrode was evaporated on the top. The device has a well-defined active area of 4.6 mm².

For DPJ device, A PC₇₁BM thin layer with thickness about 40 nm was spin-cast between Si NC/PTB7 layer and aluminium electrode. Other conditions are identical to control device.

2.3. Characterization

Fourier transform infrared spectroscopy (FTIR) spectrum was taken by depositing Si NCs directly on one window of an air-free thallium bromide (KRS-5) cell and the corresponding spectrum was recorded in transmission mode with a resolution of 4 cm⁻¹ (JASCO FT/IR-6100). Transmission electron microscopy (TEM) images were obtained using a JEOL JEM-2010F transmission

electron microscope. Device current density-voltage (*J-V*) curves were measured under AM 1.5G illumination with a Keithley 2400 Digital Source Meter. Light source was calibrated by Asahi Spectra Co., Ltd. Incident light power is checked before every measurement by using a standard silicon photodiode. Devices were also encapsulated to confirm *J-V* and external quantum efficiency (EQE) spectra were also measured on a Hypermonolight system in open air (Bunkoukeiki, CEP-25BX).

3. Results and discussion

3.1. Control device

Fig. 1a shows the schematic diagram of control device. Mixing Si NCs in PTB7 introduces a new Si NC/PTB7 interface; this interface has a desirable energy offset as shown in the band alignment diagram in Fig. 1b, which promotes exciton dissociation generated in PTB7 under light illumination [29,30]. Moreover, a type-I staggered band alignment is established in the whole device, which facilitates carrier transportation and collection [31,32]. Electrons can flow smoothly from PTB7 to Si NCs and finally be collected through a Si NC/Al ohmic contact; meanwhile, holes will be collected, without any obvious potential barrier, to the opposite ITO electrode through PEDOT:PSS. Therefore, control Si NC/PTB7 hybrid device has the similar working principle with bulk heterojunction polymer solar cell [19,20,33]. In addition, it has been confirmed from an earlier study that, Si NCs can distribute almost closely and uniformly in the whole active layer using a 1:1 Si NC/PTB7 weight ratio [34]. This guarantees sufficient exciton separation by providing an extensive Si NC/PTB7 interface, efficient electron tunnelling transportation

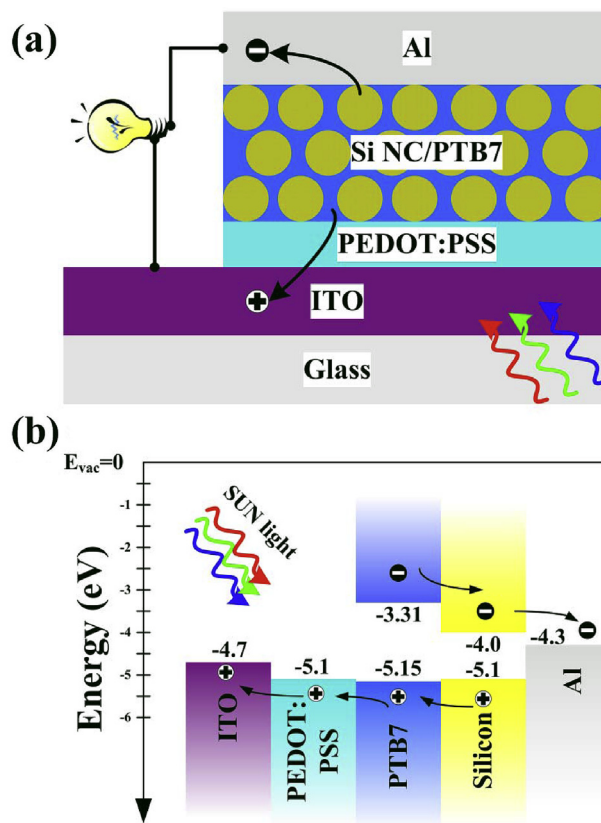


Fig. 1. (a) Schematic diagram of control device structure, and (b) its corresponding band alignment.

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