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## Hybrid bulk-heterojunction solar cells based on all inorganic nanoparticles

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## ABSTRACT

Hybrid bulk-heterojunction (HBH) nanostructure enables high excitons splitting efficiency by virtue of increasing contact area in the active layer. In this work, we have demonstrated a HBH solar cell based on all inorganic p-type and n-type nanoparticles. In order to facilitate charge transportation and collection after efficient excitons splitting, nanotetrapod shaped CdTe is used as p-type semiconductor and CdSe quantum dot as n-type material. The operation mechanism of HBH solar cells is demonstrated to be a donor–acceptor (D–A) model based on splitting of excitons. Besides the morphology and absorption characterization, photoluminescence quenching behavior is researched to show the excitons splitting process. Solar cells with HBH structure show promising photovoltaic performance, which benefits from the percolated donor–acceptor networks in the hybrid film. Our preliminary research shed light on the new development of solution based nanoparticles thin film solar cells.

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## 1. Introduction

Solar cells based on quantum dots (QDs) have caused more and more attention in view of their compatibility with materials' solution synthesis, devices' low cost fabrication and large area flexibility. Compared with their counterparts of organic solar cells which also possess these potentials, QDs solar cells offer easy tuning of light response in a broad range by using the quantum size-effect of colloidal QDs. Earlier research of all inorganic nanocrystals solar cells based on CdTe/CdSe bilayer structure generated an efficiency as high as 2.9% [1]. Afterwards, solar cells adopting PbS colloidal QDs underwent a rapid development, with their efficiency rapidly increasing from 2% in the Schottky structure [2] to 6% in reversed bilayer heterojunction structure [3], and recently to 6.6% with a p–n type PbS QDs heterojunction [4].

As regard to the presently researched photovoltaic device with a Schottky contact or bilayer heterojunction structure, it is suggested that the photocurrent is generated from charge separation under the driving force of built-in electric field in the depletion region which locates at the semiconductor–metal contact [5,6] or p–n interface [7,8]. Photogenerated excitons must diffuse a long way to the p–n depletion region before their splitting, which takes a high risk of recombination considering a relatively large QDs

thickness as well as a small depletion width. The depletion region was broadened by forming bulk-heterojunction contact where PbS QDs were penetrated into a TiO<sub>2</sub> porous layer [9]. Besides, quantum funnels structure was found to be helpful in driving minority electrons under a graded energy band structure [10]. However, these nanostructures were either complicated to be achieved or difficult to form enough heterojunction contact areas. Other nanostructures are needed for an efficient formation of depletion region and splitting of excitons.

It is well known that organic solar cell possesses very high excitons dissociation and charges collection efficiencies due to the hybrid bulk-heterojunction (HBH) structure [11,12]. P-type organic molecules are mixed with n type molecules in a nano-sized phase separation. With the help of the two interpenetrated phases, charge carriers could be conveniently transported and collected after excitons splitting. In this work, the HBH concept is introduced to fabricate all inorganic QDs solar cells. Another critical problem during applying this structure in inorganic solar cell is that continuous charges transportation should also be required at the same time. Thus, it might not be a good idea if both spherical shaped p and n type nanocrystals are blended together, which is difficult to form network pathways for electrons and holes. Nanocrystals with a hyperbranched shape can overcome the trade-off between efficient excitons separation, charges transportation and collection. Herein, CdTe nanotetrapods (NTs) are used to mix with CdSe QDs to form the HBH nanostructure with a type-II energy band alignment [13–15]. It is for the sake of a densely

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compacted hybrid film that the spherical shaped CdSe is used, not the same tetrapod shape. Hybrid film morphology and excitons splitting property were characterized to demonstrate the base of photovoltaic effect. By tuning different device parameters, such as mass ratio, film thickness and annealing temperature, we successfully demonstrate all inorganic HBH nanostructured solar cells composed of nanotetrapods and quantum dots, which opens up new opportunity in developing efficient and solution based nanoparticles (NPs) thin film solar cells.

## 2. Experimental section

### 2.1. Synthesis of CdTe NTs and CdSe QDs

CdTe NTs and CdSe QDs were synthesized according to the procedure in literature [16] with some modifications. Typically, a Cd-precursor solution (1 mmol of CdO dissolved in 3 mL of oleic acid together with 3 g tri-n-octylphosphine oxide (TOPO)) was heated to 280 °C. A Te-source solution (0.5 mmol of Te dissolved in 3 mL tri-n-octylphosphine (TOP)) was quickly injected into the Cd precursor solution and the reaction was performed for 3–4 min at 260 °C to synthesize CdTe tetrapod nanocrystals. As to CdSe QDs, similar recipe and procedure were used by changing Te into Se powder with a volume of 1.0 mmol. The synthesized CdTe NTs and CdSe QDs underwent the same purification process firstly by centrifuging the reaction solution after adding a little volume of ethanol and then cleaning the precipitates with chlorobenzene/ethanol solvent/antisolvent for at least three times. The obtained products were dissolved in chlorobenzene with a certain concentration.

### 2.2. Fabrication of HBH solar cells using CdTe:CdSe blended hybrid

The fabrication process of HBH solar cells is as follows. A 40 nm thick layer of poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) is spin-coated onto the ITO substrate that is precleaned by soap water followed by deionized water, acetone and isopropanol. The ITO/PEDOT:PSS substrate is annealed at 140 °C for 1 h and then it is transferred into a glove box together with the CdTe:CdSe mixed solution. The mixture is prepared by mixing CdTe NTs and CdSe QDs chlorobenzene solution and thereafter stirring for at least 24 h. HBH films with different thickness are achieved by spin-coating the mixture at different speed for 60 s in N<sub>2</sub> atmosphere. For a hybrid film thicker than 500 nm, two or three spin-coating process was carried out on the substrate, and between each spin-coating of hybrid solution, the substrate was washed with methanol and then dried under vacuum. At last, the substrates with HBH films were annealed at different temperature for 10 min. Afterward, onto the hybrid film, a ZnO buffer layer of about 20 nm is obtained as is usually done [17]. The solar cells fabrication is finished by thermally depositing a 100 nm aluminum cathode on top. The HBH solar cell has a final structure as: ITO/PEDOT:PSS/CdTe NTs:CdSe QDs/ZnO/Al, as shown in Fig. 1 (b).

### 2.3. Characterization

The shape of CdTe NTs and CdSe QDs was determined by a transmission electron microscopy (TEM) on a Hitachi H-800 at an acceleration voltage of 80 kV. HBH thin film surface, cross-section morphology and energy dispersive X-ray spectroscopy (EDS) were measured by field emission scanning electron microscopy (FESEM, JEOL 7006 F) as well as TEM. The EDS characterization was carried out at a scanning voltage of 15 kV. As about the TEM test, a thin film of CdTe:CdSe hybrid was fabricated firstly by spin-coating the mixed

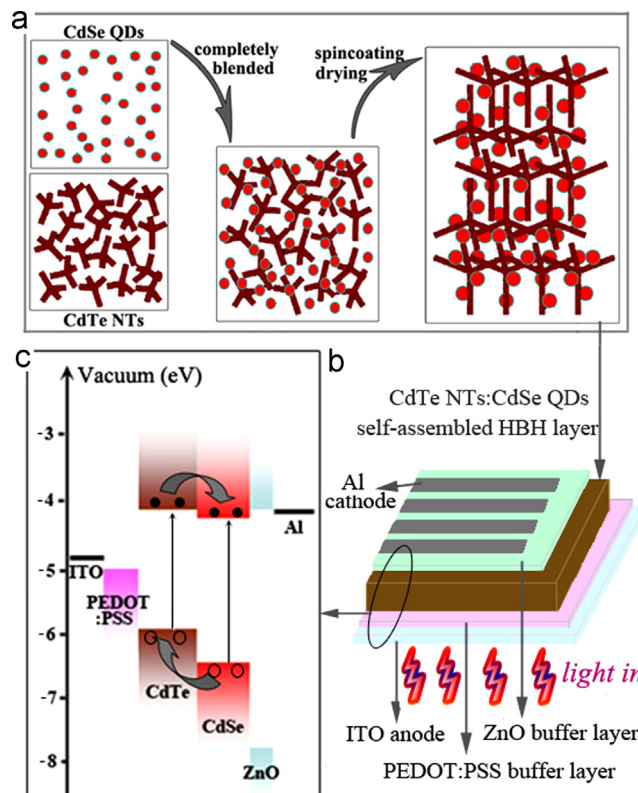


Fig. 1. Schematic images showing (a) the formation process of NTs:QDs HBH structure, (b) hybrid solar cell structure and (c) its corresponding energy level alignment.

solution on PEDOT:PSS layer, annealing at 150 °C and then immersing the substrate in water to dissolve the PEDOT:PSS layer; this process left a separated hybrid film that can be attached on copper net for TEM testing. Also, traditional TEM samples were made by dropping and drying the solution on copper net. X-ray diffraction pattern was recorded on a DX-2700 X-ray diffractometer with Cu K<sub>α</sub> radiation. Light absorption (Abs) and photoluminescence (PL) measurements were carried out on Varian U-3000 model ultraviolet-visible spectrophotometer and Varian Cary Eclipse fluorescence spectrophotometer respectively. Photocurrent characterization was carried out on Growntech IPCE test station with Keithley 2000 multimeter. The current–voltage (*J–V*) measurements on CdTe:CdSe HBH solar cells were performed on Keithley 2400 source in forward bias mode under AM 1.5 (100 mW cm<sup>-2</sup>) illumination.

## 3. Results and discussions

### 3.1. NTs:QDs HBH device structure and its operating mechanism

As a novel hybrid structure, the formation process of NTs:QDs HBH is supposed to be like this (as depicted in Fig. 1(a)): loosely but completely blended CdSe QDs and CdTe NTs mixture is formed firstly in the hybrid solution, and after spin-coating and drying, self-assembly of the nanoparticles happens so that CdSe QDs infiltrate in the gaps among the branched CdTe NTs under the driving force of space steric hindrance. This HBH active layer is sandwiched between PEDOT:PSS and ZnO buffer layer as shown in the skeleton image in Fig. 1(b). Theoretically, this device structure enables a suitable energy level distribution for the generation and collection of photocurrent (Fig. 1(c)). Herein, considering the nature of hybrid bulk-heterojunction structure, PEDOT:PSS buffer layer is used to provide an electrons barrier to the ITO anode and

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