



Simultaneous study of exciton dissociation and charge transport in a light harvesting assembly: Lead selenide nanocrystal/zinc oxide interface



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ABSTRACT

Simultaneous study of exciton dissociation between lead selenide (PbSe) and zinc oxide (ZnO) nanocrystals, and charge transport in the electron acceptor (ZnO) was carried out using the field effect transistor (FET) arrangement in which a bi-layer of PbSe and ZnO is formed onto the SiO₂ substrate. Using multiple potential probes between the source and drain electrodes, change in the electrical conductance in the ZnO, proportional to exciton dissociation at the electron donor/acceptor (PbSe/ZnO) interface, was measured excluding the Au/ZnO contact resistance. Surface ligand capping dependent exciton dissociation at the ethanedithiol-treated PbSe/ZnO interface was compared with the biphenyl-4,4'-dithiol-treated PbSe/ZnO interface using the FET geometry, demonstrating that a shorter ligand is more favorable to exciton dissociation. From the light intensity dependent conductivity change in the ZnO layer, the origin of carrier recombination is discussed.

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1. Introduction

Photoinduced charge transfer at the interface between nanocrystal (NC) sensitizers and metal oxide acceptors has been the main topic in assembling quantum dot (QD) solar cells (SCs) [1–3]. Colloidal quantum dot (CQD) Schottky SCs in which exciton dissociation occurs at the metal oxide/QD interface have been extensively researched to enhance the power conversion efficiency through tuning electrical and optical properties of CQD NCs [4,5]. Recombination of electrons and holes generated in the QD film and the back-transfer of holes to the metal electrode has limited the open circuit voltage and short circuit current [6–8]. CQD-sensitized SCs employ a monolayer of CQD film as a light absorber coupled to electron acceptor layers, preventing recombination of electrons and holes due to the energy barrier offered by the electrolyte interfacial with the electron acceptor [7]. Depleted hetero-junction (DH) CQD SCs possess the electron donor/acceptor interface creating charge depletion region at both sides [8,9]. Built-in electric field in the depletion region drives electrons and holes to the carrier collecting electrodes. DH CQD SCs provide flexibility in designing a high efficiency QD SC through controlling thickness of the component layers or inserting exciton blocking layers, resulting in an increased short circuit current [10,11].

In the devices, the short circuit current is determined by a series of charge transfer and transport processes at a number of interfaces [9, 12,13]. Electrons transferred to the electron acceptor from the electron donor travel to the cathode electrode through the electron acceptor layer and the acceptor/cathode electrode interface which can serve as

the potential trapping centers for electrons. Similarly, holes left in the electron donor layer transport to the transparent conducting anode electrode through the electron donor and the donor/anode electrode interface. The presence of the potential trapping and recombination centers in the path and the back transfer of excitons not only reduce the short-circuit current but also complicate understanding of exciton dissociation at the electron donor/acceptor interface, hindering simultaneous study of the charge transport in the bulk and photoinduced charge transfer at the interface. Moreover, in the conventional excitonic QD SCs, the thickness of light absorbing materials is thicker than the typical exciton diffusion lengths ranging tens of nanometer [9,10]. Recombination of excitons in the donor layer before reaching the interface is unavoidable, therefore, causing a photocurrent loss.

To understand exciton dissociation and the following charge transport minimizing the effect of carrier traps distributed in the bulk and interface within the complex solar cell structure, it is required to build a platform in which all of the photogenerated carriers are available for exciton dissociation suppressing carrier loss due to back transfer of excitons and (or) recombination in the electron donor layer. Here, we created a light harvesting system consisting of a PbSe/ZnO bi-layer in the bottom-contact field effect transistor (FET) structure and probed exciton dissociation and charge transport at the interface. In the arrangement, all of the electrons and holes excited in a drag-coated PbSe QD film the thickness of which is less than the exciton diffusion length can reach the interface. In the system, back transfer of electrons and holes is not allowed due to the geometry of the FET in which only electron donor/acceptor interface serves as the active layer between the source and drain electrodes. Using the light harvesting system, ligand capping dependent exciton dissociation was probed using two different capping molecules. The number of excitons dissociated at

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the interface is proportional to the conductance of the underlying ZnO layer, enabling a simultaneous study of charge transport and exciton dissociation.

2. Experimental procedure

2.1. Synthesis of PbSe NCs

Lead oxide (PbO), oleic acid (OA), trioctylphosphine (TOP), diphenylphosphine (DPP), and 1-octadecene (ODE) were purchased from Sigma-Aldrich Co. PbO (4 mmol) and OA (10 mmol) were mixed in ODE producing a precursor solution. The solution in a three-neck round bottom flask was degassed by heating at 160 °C for 1 h under nitrogen atmosphere. Se was mixed in TOP in argon filled glove box and DPP (4.5 mM) was dissolved in the solution (TOP-Se). 1 M TOP-Se solution was injected into the lead oleate solution to yield PbSe NCs. After a particular reaction time, the reaction was terminated by cooling the flask through dipping into an ice-water bath. For precipitation of PbSe QDs, ethanol was added followed by centrifugation and washing with hexane/methanol for purified

PbSe QDs. The size of PbSe QDs was controlled by temperature and reaction time in our experiments.

2.2. Formation of PbS/ZnO interface in the FET structure

Source and drain electrodes (Cr 3 nm/Au 80 nm) were photolithographically defined on a SiO₂ gate dielectric. Highly doped silicon substrate worked as a gate electrode. Multiple voltage probes spaced by 120 μm were patterned between the metal electrodes. ZnO films were grown on a 200 nm thick SiO₂ substrate using radio frequency magnetron sputtering in argon atmosphere at a deposition rate of 3 nm/min, producing a final thickness of 60 nm. The surface morphology of the ZnO layer was observed by atomic force microscopy (AFM, Asylum MFP-3D, tapping mode) and scanning electron microscopy (SEM, LEO 1550 FESEM, operated at 8.0 kV).

3. Results and discussion

PbSe/ZnO interface was incorporated into a bottom-contact FET device. Fig. 1(a) shows a schematic diagram of a PbSe (~10 nm)/ZnO (~60 nm) FET structure in which multiple voltage probes are patterned

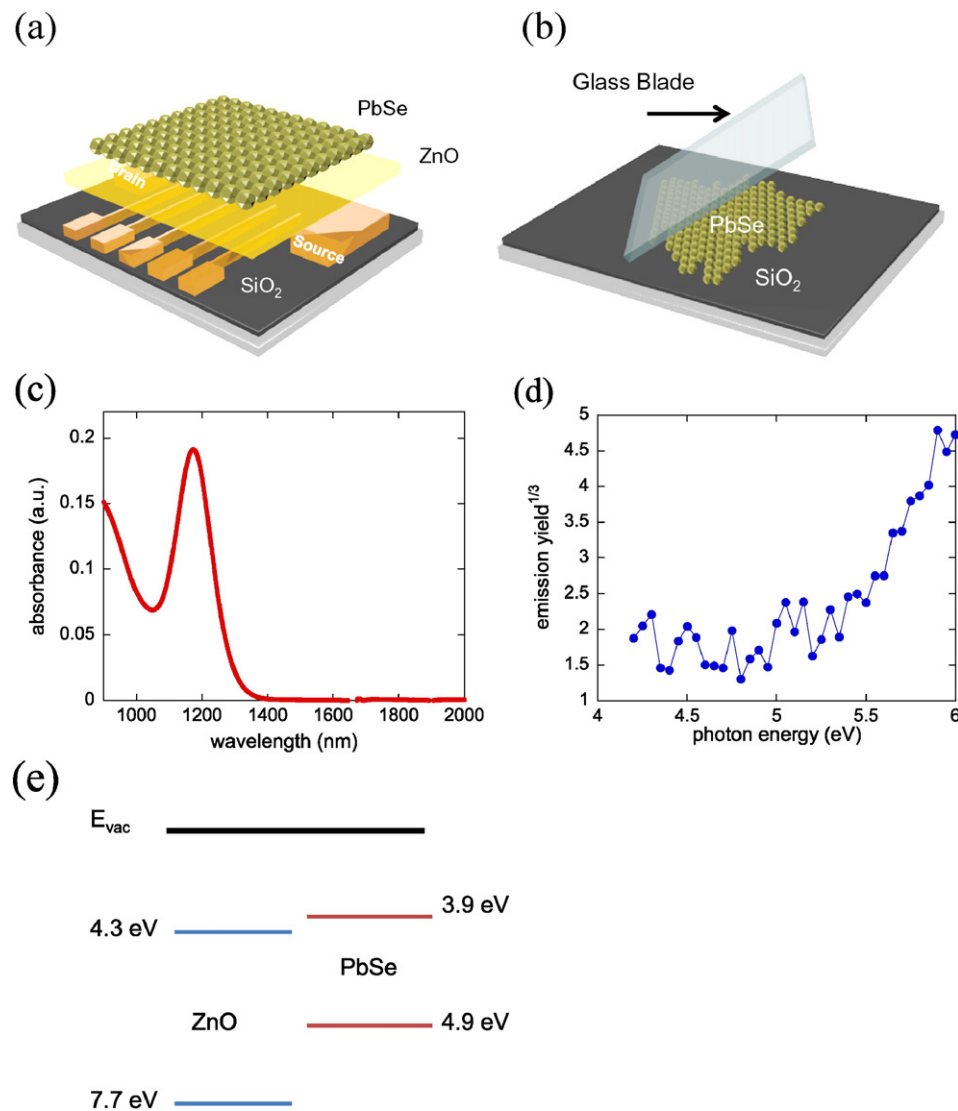


Fig. 1. Schematic diagrams of (a) an FET structure with a bi-layer of PbSe/ZnO and (b) film formation of a PbSe layer (~10 nm) on top of ZnO using drag-coating. The channel length was 160 μm and the multiple voltage probes were spaced by 25 μm. The valence and conduction band edges of ZnO were estimated from literatures mentioned in the text. (c) Optical absorption spectroscopic data for PbSe NCs. (d) Plot of emission yield as a function of photon energy for a EDT-PbSe film on an ITO coated glass. (e) The energy band diagram drawn from the optical absorption and PESA measurements.

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