



Interfacial trap-induced confinement of mobile carriers during illumination at a pentacene/lead-sulfide functional interface



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ABSTRACT

Interfacial electronic properties at the pentacene/lead sulfide (PbS) interface in the dark under illumination were studied. With a small size of PbS colloidal quantum dot (CQD) (2.7 nm), photoinduced charge transfer at the pentacene/PbS CQD interface leads to a large threshold voltage shift in a pentacene/PbS CQD field effect transistor (FET). Through estimation of interfacial trap density by sub-threshold slope measurements in an FET structure, the mechanism by which threshold voltage shift occurs is discussed.

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1. Introduction

Charge transfer at the interface including organic semiconducting materials and nanocrystals is a crucial component in creating functional interfaces as well as in operating electronic devices [1–3]. Exciton dissociation, charge transfer, and charge trapping at various functional interfaces, therefore, have been extensively researched due to applications to various optoelectronic and memory devices [4–9]. Among various component layers comprising functional interfaces, organic semiconductors and colloidal quantum dots (CQDs) feature capability to tune charge transfer efficiency through triplet dissociation and size dependent optoelectronic properties, respectively [3,10,11]. Among organic semiconducting materials, pentacene is a very attractive semiconducting material used in a *p*-channel field effect transistor. Its electronic and structural properties have been extensively studied [12–14]. More interestingly, it has been reported that pentacene features singlet exciton fission in which two low energy triplet states are formed from a higher energy singlet exciton during illumination, improving photo-induced charge transfer efficiency [14–16]. Lead sulfide (PbS) CQDs with a Bohr radius of ~20 nm feature tunable optoelectronic properties due to quantum confinement effect [17,18]. The size dependent optoelectronic properties of PbS CQDs provide functional flexibility in designing

highly efficient optoelectronic devices operated by charge transfer and transport at the interface with various semiconducting materials [19–23].

Here, a functional interface consisting of PbS CQDs and pentacene is created and the effect of interfacial electronic properties on charge transfer during illumination is probed. As a result of photo-induced charge transfer, confinement of mobile carriers into a 2D carrier positioned near the interface is demonstrated through photo-induced threshold voltage measurements in a pentacene/PbS FET. Presence of the interfacial trap estimated through the sub-threshold measurements provides insight into understanding of the possible mechanism by which 2D carrier sheet is formed under illumination.

2. Experimental details

2.1. Synthesis of PbS CQDs and structural characterizations

Lead (II) Oxide (PbO, 0.45 g) was mixed with an oleic acid (20 ml) and 1-octadecene mixture in a three-neck flask. Hexamethyldisilathiane (TMS) solution, prepared in a glove box, was rapidly injected into the PbO solution at 110 °C. After 3 min, the solution was cooled down in an ice bath and transferred for centrifugation to collect a PbS CQD solution (40 mg/ml) in hexane. Surface morphology of PbS CQDs and pentacene films was investigated using tapping mode atomic force microscopy (AFM). To describe energetic level alignment at the PbS/pentacene interface, the energy band gap of PbS and pentacene films were extracted

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using optical absorption spectroscopy. Using high resolution transmission electron microscopy (HRTEM) and X-ray diffraction (XRD), the structural properties of PbS CQDs including crystallinity and quantum dot size were characterized.

2.2. Fabrication of a bottom-contact FET and electrical characterization

To structure a bottom-contact FET, 200 nm thick SiO₂ was used as a gate dielectric layer. Source and drain electrodes (Au (80 nm)/Ti (5 nm)) were pre-patterned onto the SiO₂ gate dielectric using the conventional photolithography process. PbS CQDs dispersed in hexane were spin-coated onto the gate dielectric, producing a thickness of ~10 nm. To form a pentacene/PbS interface, the FET device was transferred to a vacuum chamber equipped with a built-in electrical feed-through, allowing for *in-situ* electrical measurements. The *in-situ* measurement system prevents exposure of completed FET devices to the atmosphere which can degrade the interfacial properties. Electrical characterizations of FETs were carried out using a semiconductor parameter analyzer.

3. Results and discussion

The first excitonic peak of PbS CQDs at a particular size is determined by the band gap of the PbS CQDs. In Fig. 1(a) the first excitonic peak was observed at 680 nm and the band gap of the PbS CQDs was estimated from the Tauc plot in the inset of Fig. 1(a). The size of PbS CQDs based on the band gap was estimated to be 2.7 nm [24]. From Scherrer equation based on the XRD data of PbS CQDs in Fig. 1(b), the average crystallite size of PbS CQDs was calculated to be 2.6 nm consistent with the size calculated from the energy band gap. The lowest unoccupied molecular orbital (LUMO) and the highest occupied molecular orbital (HOMO) levels of PbS CQDs

were calculated from the energy band gap and the size. The magnitude of the LUMO and HOMO levels are in good agreement with those predicted from the position of the first absorption peak of the PbS CQDs [24,25]. The HRTEM image of PbS CQDs in Fig. 1(c) as well as the XRD data in Fig. 1(b) indicates that PbS CQDs are highly crystalline. Energy levels at the pentacene/PbS interface derived from the above spectroscopic methods are exhibited in Fig. 1(d). The LUMO level of PbS is deeper than that of pentacene and the HOMO level of PbS is close to that of pentacene. The LUMO (3.2 eV) and HOMO (5.0 eV) levels of pentacene were calculated by the Tauc plot from optical absorption measurement and the ionization energy measurement from photoelectron spectroscopy in air (Data not shown) [24].

In the bottom-contact FET structure with the width of Z and the channel length of L , the drain current, I_D , is determined by the FET mobility, μ and the carrier concentration, $C_{ox}(V_G - V_T)$, as shown in below Eq. (1):

$$I_D = \frac{Z}{L} \mu C_{ox} (V_G - V_T) V_D \quad (1)$$

where C_{ox} is the capacitance of the gate dielectric, V_T is the threshold voltage, and V_D is the drain voltage. When the gate voltage, V_G , is far larger than the drain voltage, the carrier concentration in the channel region becomes uniform, achieving the linear transport regime in transistor operation. In the linear transport regime, the threshold voltage is defined as the required gate voltage to induce mobile carriers in the channel between the source and drain electrodes [26,27]. Above the threshold voltage, the drain current linearly increases with the carrier concentration increasing at a given FET mobility, following the relation in Eq. (1).

Pentacene/PbS interface was incorporated into a bottom-contact FET structure shown in the inset of Fig. 2(a). The

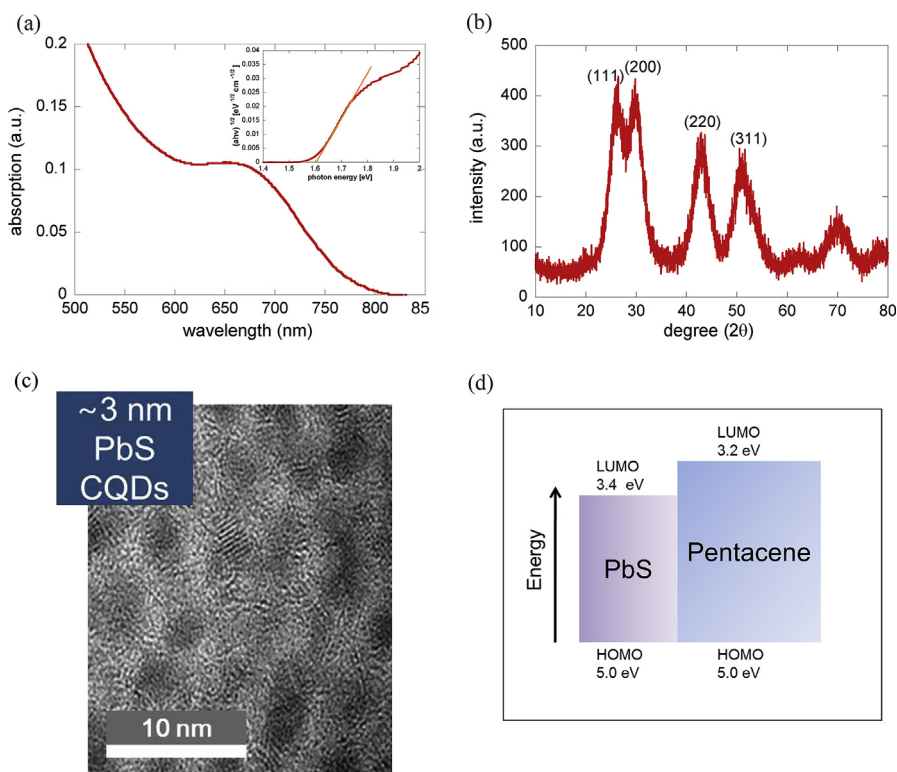


Fig. 1. (a) Optical absorption spectrum of PbS CQDs (~3 nm). The inset shows the Tauc plot for calculation of the energy band gap (1.6 eV). (b) XRD data for PbS CQDs (c) HRTEM image of PbS CQDs. The average size estimated from the image is close to 3 nm. (d) Energy band diagrams of PbS CQD and pentacene.

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