



Thermal-assisted band to band tunneling at the electron donor/acceptor energy harvesting assembly



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ARTICLE INFO

Article history:

Received 3 October 2016

Received in revised form 25 January 2017

Accepted 30 January 2017

Available online 31 January 2017

Keywords:

Tunneling

P3HT

Contact resistance

Zinc oxide

Recombination

ABSTRACT

Understanding interfacial electronic properties at the electron donor/acceptor energy harvesting assembly in photovoltaic devices is important in that it can provide insights into revealing charge carrier recombination mechanism at the interface. From temperature dependent contact resistance measurements at the aluminum-doped zinc oxide (AZO)/poly-hexylthiophene (P3HT)/aluminum-doped zinc oxide (AZO) structure in which AZO (electron acceptor) acts as highly conductive source and drain electrodes, charge transfer mechanism at the electron donor/acceptor interface is elucidated. In the mechanism, charge transfer process is initiated by band to band tunneling (BTBT) of hole carriers from AZO to P3HT through localized electronic states populated near the highest occupied molecular orbital (HOMO) level of the P3HT close to the AZO favored by energetic alignment between the conduction band edge of AZO and the HOMO of P3HT. Following BTBT, charge transfer process is completed by thermal activated hopping in the P3HT (electron donor) close to the AZO. It is found that thermally activated transport in the P3HT close to the AZO dominates the magnitude of the contact resistance, providing insights into the origin of carrier recombination mechanism at the donor/acceptor interface in photovoltaic devices.

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1. Introduction

Understanding charge transfer mechanism at the electron donor/acceptor energy harvesting assembly associated with energetic alignment and the electronic localized states is of paramount importance because it can provide a clue to reveal the origin of carrier recombination, unavoidable at the interfaces during operation of many optoelectronic devices. Particularly, in photovoltaic devices operated by exciton diffusion and separation at the donor/acceptor interface followed by charge transport, identifying and probing the charge transfer mechanism at the interface are crucial [1–3].

In charge transfer process at the electron donor/acceptor interface, energy alignment and the carrier density of states (DOS) dominate the charge transfer mechanism because energy band offset at the interface works either as a driving force or an activation energy to overcome for charge transfer [4,5]. Moreover, the presence of localized electronic states at the interface, called interfacial traps, can significantly alter the path of carriers during charge transfer process [6,7]. Indeed, the magnitude of the maximum achievable open circuit voltage, set by the difference between the lowest unoccupied molecular orbital (LUMO) of the electron acceptor and the highest occupied molecular orbital (HOMO) of the electron donor, is significantly modified by the charge

transfer mechanism associated with interfacial traps which act as main recombination centers [8–10]. Further, the presence of recombination centers (interfacial traps) may influence charge transfer/transport mechanism by changing energetic alignment between the donor/acceptor interface altering the magnitude of the photocurrent [11–13].

In most cases, electron donor/acceptor interfaces are buried in the vertically stacked photovoltaic devices, challenging analysis of electrical properties resulting from the charge transfer/transport mechanism at the interface. A series of functional interfaces designed for device operation are arranged within the photovoltaic devices complicating a separate study of the donor/acceptor interface. As demonstrated in many spectroscopic studies, further, investigating the effect of localized states residing at the buried interface on the electrical properties in a real device arrangement requires a number of assumptions relating to charge transfer/transport although energetic alignments between the component layers have been addressed by spectroscopic tools such as ultraviolet photoelectron spectroscopy [14,15]. In other words, with only spectroscopic analysis, information of electrical charge transfer/transport associated with localized states and energy band bending is not available.

Here, interfacial charge transfer mechanism at the electron donor/acceptor assembly consisting of a representative conjugated polymer and a metal oxide was investigated using temperature dependent contact and channel resistance measurements with transmission line method (TLM) [16,17]. In the arrangement, the electron acceptor serves

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as the source/drain electrodes, a reservoir of charge carriers, allowing for measurement of the electrical contact resistance at the electron donor/acceptor interface. This enables direct probing of charge transfer/transport at the interface leading to simultaneous measurements of carrier transport and the contact resistance whereby the charge transfer/transport mechanism associated with carrier recombination at the interface can be revealed. As an electron donor/acceptor assembly, poly-hexylthiophene (P3HT) and aluminum-doped zinc oxide (AZO) were chosen because they have been demonstrated as a representative donor/acceptor assembly in hybrid photovoltaic devices [18, 19]. Particularly, highly conductive metal oxide layers including AZO have attracted much interest as an electron extracting layer because its high conductivity compensates for increase in thickness reducing structural defects with a very thin metal oxide film [20–22].

2. Experimental section

The contact resistance between AZO and P3HT in the AZO/P3HT/AZO structure was measured using TLM in which the electrical resistance of the P3HT channel between AZO electrodes are controlled by varying the channel length as shown in Fig. Fig. 1(a). Highly conductive AZO electrodes were fabricated by doping Al during sputtering followed by thermal annealing in a nitrogen atmosphere at 500 °C. In the sputtering process, AZO films were formed in argon atmosphere at 400 °C growth temperature using 2-inch ZnO single ceramic target doped with 3 wt% Al₂O₃. To fabricate the AZO/P3HT/AZO test structure, positive photoresist was spin-coated onto the AZO film followed by ultraviolet (UV) treatment, patterning the channel region. The photoresist on the

channel area is exposed to UV and etched away in the develop process. The defined channel region is dipped in a dilute HCl solution to remove the AZO film on which photoresist is not coated. After fabrication of AZO source and drain electrodes, P3HT solution in chloroform was spincoated on the structure at 1500 rpm for 30 s, producing a 60 nm thick P3HT film.

The P3HT channel resistance, R_{ch} and the AZO/P3HT contact resistance, R_c , comprise the total resistance, R_{tot} , as given in Eq. (1);

$$R_{tot} = R_{ch} + R_c = R_s \frac{L}{w} + R_c \quad (1)$$

Here, R_s is the sheet resistance, L the channel length, and w is the width. The total resistance is plotted as a function of channel length, L . The contact resistance is determined from the total resistance at which the channel length is zero from a linear fitting of the plot of the resistance as a function of channel length.

3. Results and discussion

In our previous study [23] it is found that tunneling current at the AZO/P3HT interface embedded in the AZO/P3HT/AZO structure is favored by energetic alignment between the CBE of AZO and the HOMO of P3HT, and the presence of interfacial electronic localized states in the P3HT close to the AZO. The presence of tunneling current at an inorganic/organic interface including a wide-bandgap semiconductor was also confirmed in our previous study of interfacial charge transfer between GaN and P3HT [24]. In the study, an *n*-GaN/P3HT/*n*-GaN

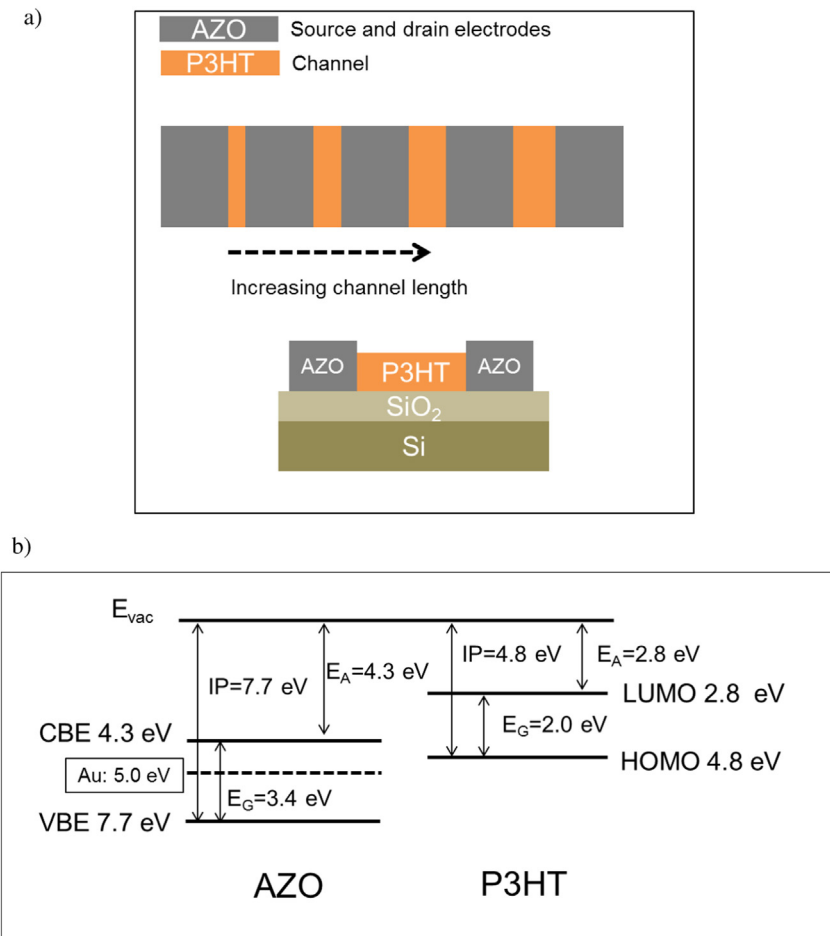


Fig. 1. (a) TLM structure for the contact resistance of AZO/P3HT. The P3HT (60 nm) channel length between AZO (80 nm) electrodes ranged between 4 and 50 μm and the width is 2 mm. (b) Energy band diagrams for AZO and P3HT.

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