



## Research paper

# Study on interface engineering of layer-by-layer structure for applications in organic photodetector



Shuanghong Wu<sup>a</sup>, Junjie Yang<sup>a</sup>, Wenbin Ye<sup>a</sup>, Han Zhou<sup>a</sup>, Xiangru Wang<sup>a</sup>, Xiongbang Wei<sup>a</sup>, Silu Tao<sup>a</sup>, Zhenyu Chen<sup>b</sup>, Chundong Wang<sup>c</sup>, Qihui Wu<sup>d,\*</sup>, Zhi Chen<sup>a,e,\*\*</sup>

<sup>a</sup> School of Optoelectronic Information and State Key Laboratory of Electronic Thin Films & Integrated Devices, University of Electronic Science and Technology of China, Chengdu 610054, China

<sup>b</sup> Nano Top Electronic Technology Co. Ltd., Beijing 101407, China

<sup>c</sup> School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China

<sup>d</sup> Department of Materials Chemistry, School of Chemical Engineering and Materials Science, Quanzhou Normal University, Quanzhou 362000, China

<sup>e</sup> Department of Electrical & Computer Engineering, University of Kentucky, Lexington, KY 40506, USA

## ARTICLE INFO

## Keywords:

Organic

UPS

Photodetector

Interface

## ABSTRACT

The interfacial electronic structure of layer-by-layer 4-(dicyanomethylene)-2-t-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTb)/fullerene C<sub>60</sub> was investigated using ultra-violet photoemission spectroscopy (UPS). Photoemission data of period of DCJTb/C<sub>60</sub>/DCJTb films suggested the formation of surface dipole and interfacial band bending across the interfaces, which greatly facilitates the charge transfer from DCJTb to C<sub>60</sub> and from C<sub>60</sub> to DCJTb layer as well. When applied this layer-by-layer structure to a near-infrared photodetector, a maximum of photocurrent was achieved by the device with 3 periods of DCJTb/C<sub>60</sub> thin films. Finally, the detailed work mechanism of this detector was discussed.

## 1. Introduction

Organic photodetectors (PDs), which possess the advantageous properties of low cost, light weight, and flexibility, have been widely studied due to their tremendous potential in industrial and scientific applications [1–16]. To improve the performance of PDs, a variety of strategies have been pursued including the development of new materials with high absorption and high mobility [1–4], new methods to choose materials and control film quality for improving charge transport and reducing recombination [5–12], and novel device architectures with different functional layers [13–16]. Though significant progress has been made in developing the materials and high performance organic PDs, many fundamental aspects of the organic–organic semiconductor interfaces remain to be understood. A great deal of the physics occurs at the organic/organic interfaces that governs the behaviour of molecules for electronics. For example, the nature of organic interfaces determines the fate of excitons to be either stabilised for efficient organic light emitting diodes or destabilised for efficient PDs at the interfaces. Therefore, by selecting organic semiconductors with proper band-edge offsets between their highest occupied molecular orbitals (HOMOs) and lowest unoccupied molecular orbitals (LUMOs),

different device characteristics can be readily achieved. Consequently, interface engineering, a novel approach towards high-performance PDs, has attracted considerable attention. Moreover, the huge success in realizing organic electronic devices over the past 20 years has motivated many scientists to turn towards the field of organic/organic interfaces, and the number of active groups is still increasing today.

Recently, researchers implemented layer-by-layer structures in optoelectronic devices for preparation of organic PDs [13,17], dye-sensitized solar cells [18], and organic solar cells [19,20]. Peumans and co-workers reported an efficient, high-bandwidth organic PD exhibiting external quantum efficiencies of 75% across the visible spectrum using Cu–phthalocyanine (CuPc) and 3,4,9,10-perylene-tetracarboxylic bisbenzimidazole (PTCBI) as donor (D) and acceptor (A) materials, respectively [13]. It was demonstrated that photogenerated excitons could efficiently dissociate into free electrons and holes by rapid charge-transfer across the several closely spaced organic layer interfaces and escape of the photogenerated carriers from potential wells formed by the multilayers due to tunneling prior to recombination resulting in the high efficiency. This suggests that the interface in the device is very important and the layer-by-layer structure has a bright future for high efficient organic electronic devices. Vonhoeren et al.

\* Corresponding author.

\*\* Corresponding author at: School of Optoelectronic Information and State Key Laboratory of Electronic Thin Films & Integrated Devices, University of Electronic Science and Technology of China, Chengdu, 610054, China.

E-mail addresses: [qhwwu@qztc.edu.cn](mailto:qhwwu@qztc.edu.cn) (Q. Wu), [zhichen@engr.uky.edu](mailto:zhichen@engr.uky.edu) (Z. Chen).

<https://doi.org/10.1016/j.synthmet.2017.10.001>

Received 1 August 2017; Received in revised form 25 September 2017; Accepted 6 October 2017

Available online 20 November 2017

0379-6779/© 2017 Published by Elsevier B.V.

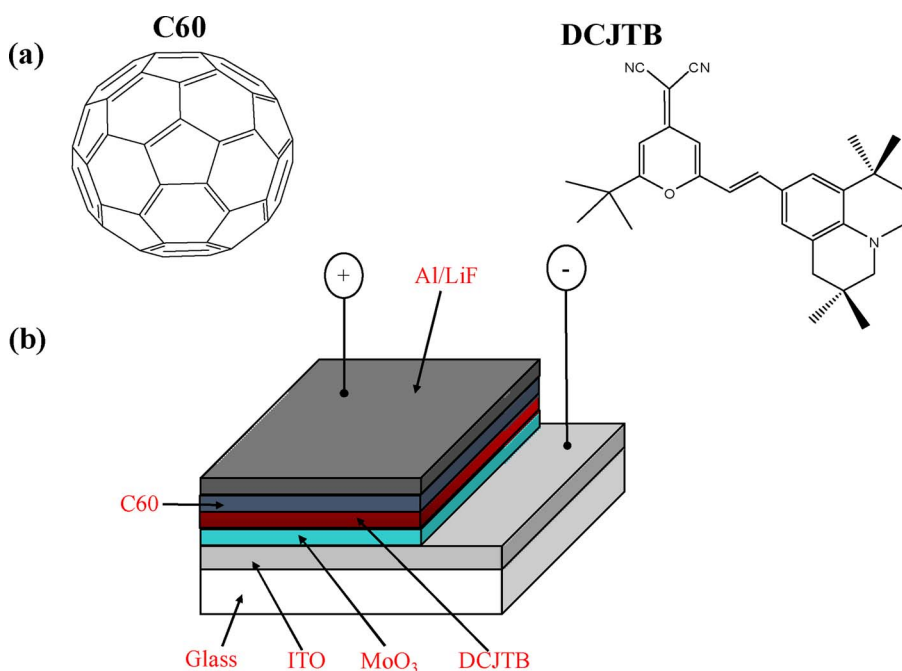


Fig. 1. (a) Molecular structures of C<sub>60</sub> and DCJTb, (b) schematic construction of double-layer device.

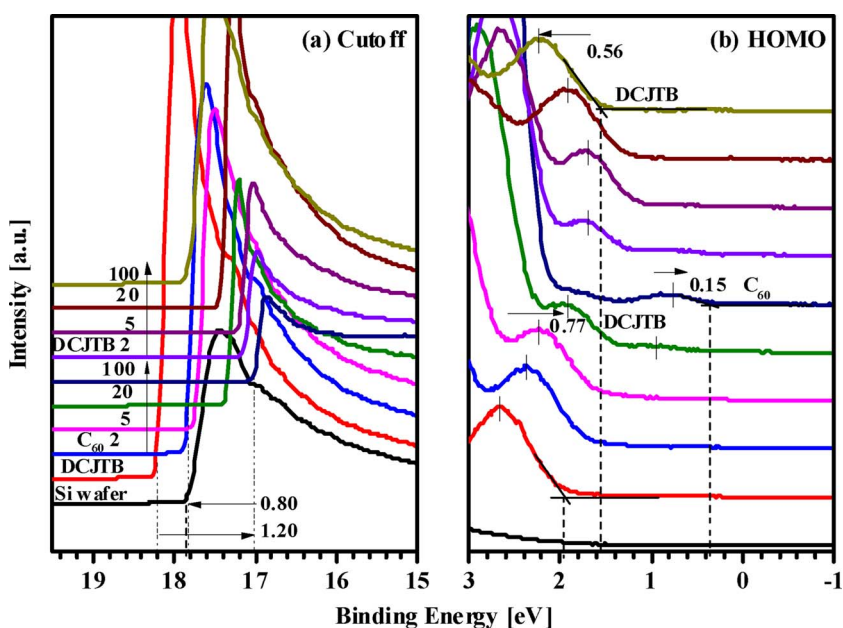


Fig. 2. Evolution of He I UPS spectra of upon deposition of C<sub>60</sub> on DCJTb on Si wafer and also upon the continuous deposition of DCJTb on C<sub>60</sub> near the (a) cutoff and (b) HOMO regions. The marked values of thickness in the unit of nm.

reported organic PDs with an active layer assembled by layer-by-layer deposition, which is sandwiched by an aluminum (Al) and an indium tin oxide (ITO) electrodes. This study showed that the layer-by-layer method is well suited for fabricating organic PDs with controlled chromophore content and that the number of active layers has a critical impact on the device performance [17]. The work reported by Hsu et al. was particularly interesting because in which they employed layer-by-layer approach to assemble graphene/TCNQ stacked films as conducting anodes for organic solar cells, which is quite attractive for next-generation flexible devices demanding high conductivity and transparency [19].

As an efficient red dye, 4-(dicyanomethylene)-2-t-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTb) has been widely used in the fabrication of organic electroluminescent devices, playing an important role in the improvements of device efficiency, operational stability, and color fidelity [21–24]. It was also used in solar cells for high

efficiency [25–28], however, it is seldom used in organic photo-detectors. In this paper, we study the interfacial electronic structure of DCJTb and C<sub>60</sub>, and report a near-infrared (NIR) PD based on them as the active materials with layer-by-layer structure. We demonstrate that the interfacial band bending and the formation of surface dipole at the junction of DCJTb/C<sub>60</sub> and C<sub>60</sub>/DCJTb detected using ultra-violet photoemission spectroscopy (UPS) greatly facilitates the charge transfer from DCJTb to C<sub>60</sub> and from C<sub>60</sub> to DCJTb films. The relationship between the number of active layers and PD performance were studied and a maximum of photocurrent was achieved by the device with 3 periods of DCJTb/C<sub>60</sub> thin films.

## 2. Experimental

All chemicals were purchased commercially and used without further purification. Organics and metal layers were sequentially deposited

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