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Effect of Gate Voltage in Organic Phototransistors Based on Polythiophene/ Fullerene Bulk Heterojunction Nanolayers

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Here we report the effect of gate voltage on the performance of organic phototransistors made using bulk heterojunction films of regioregular poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM). To understand the illumination effect, a monochromatic light (520 nm) corresponded to the maximum absorption of present P3HT:PCBM blend film was employed. Results showed that the present device followed a p-type field-effect transistor in the dark and under illumination though the saturation trend of output curves became weakened under illumination. The responsivity exhibited a two-stage trend as a function of gate voltage. The maximum responsivity reached 0.13 A/W under illumination of green light at 81 $\mu W/cm^2$.

Keywords Gate voltage; organic phototransistors; PCBM; P3HT; responsivity; saturation

Introduction

Recently organic semiconductors have attracted great attention in a variety of applications such as displays, memory devices, energy conversion devices, photosensors, etc. These applications include organic field-effect transistors (OFETs) [1], organic solar cells (OSC) [2], organic light-emitting devices (OLEDs) [3,4], organic photodiodes (OPDs) [5], and/or organic phototransistors (OPTRs) [6–10]. Of these applications, OPTRs have hold less spotlight though they could replace expensive conventional photodetectors based on inorganic semiconductors. In principle, OPTRs are a kind of organic photodetectors in which light detection and signal amplification are combined in a single organic device (transistor). Hence OPTRs are considered as one of the feasible applications of OFETs owing to the broad absorption characteristics of organic semiconducting materials which can be ranged

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from ultraviolet to near infrared regions though a tailored synthesis is necessary to achieve the whole range absorption. This advantageous and integrated feature allows OPTRs to be applied for light-induced switches, light-triggered amplifier, integrated detection circuits, and highly-sensitive image sensor arrays.

Recently, series of researches on OPTRs have been carried out using organic small molecules and polymers, while their characteristics have been examined by varying the incident light intensity, the wavelength of incident light, the illumination direction, etc. Most of these studies were focused on the pristine organic semiconductor-based OPTRs [11]. However, no detailed study has been carried out for well-known polymer/fullerene bulk heterojunction blend systems though couple of brief reports has been out without information on the gate voltage effect [12]. In this work, we attempted to make polymer/fullerene phototransistors and examined the photoresponse characteristics as a function of gate voltage under illumination of low light intensity green light (520 nm, 81 $\mu W/cm^2$).

Experimental

Poly(4-vinylphenol) (PVP) and poly(melamine-*co*-formaldehyde) (MMF) were purchased from Aldrich and then used without further purification. PVP and MMF were mixed using propylene glycol-1-monomethyl ether-2-acetate (PGMEA) as a solvent. Regioregular poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} (PCBM) were purchased from Rieke and Nano-C, respectively. Blend solutions of P3HT:PCBM (1:0.7 by weight) were prepared using chlorobenzene as a solvent. A patterned 100 nm thick indium tin oxide (ITO)-coated glass was used as a substrate. The substrates were cleaned ultrasonically using acetone and isopropyl alcohol, followed by drying with a nitrogen flow. The PVP/MMF solution was spun on the ITO-glass substrates and then thermally cured using a hot plate at 100°C for 10 min and 200°C for 30 min to remove PGMEA in the cross-linked PVP/MMF films (thickness = ~800 nm). Next, P3HT:PCBM films (~50 nm thick) were spin-coated on the PVP/MMF films. These samples were loaded into a vacuum chamber system to deposit source and drain electrodes (top contact, 60 nm thick) by thermal evaporation of silver (Ag) in a vacuum of $\sim 10^{-6}$ torr through a shadow mask. The channel width and length of resulting top contact OPTRs was 3000 μm and 70 μm, respectively (Fig. 1).

The thickness of films and electrodes was measured using a thickness profiler (Alpha Step 200, Tencor). The basic transistor characteristics were measured using a semiconductor analyzer (Keithley SCS-4200) in the dark, while the photoresponse characteristics were examined under illumination using a integrated system equipped with a light source (TH-high power, 100 W-L, Spectral Product) and monochromator (CM-110, Spectral Product). The incident light intensity was 81 $\mu W/cm^2$ at 520 nm.

Results and Discussion

As shown in the output curves (Fig. 2a), the present device exhibited a typical *p*-type trend with the grate voltage (V_G) though the saturation behavior seems to be slightly weak as a function of drain voltage (V_D). Here the interesting point is that the drain current (I_D) became already as high as -10 nA at $V_D = -60$ V without applying any bias on the gate electrode, which is attributed to the high dark current (background

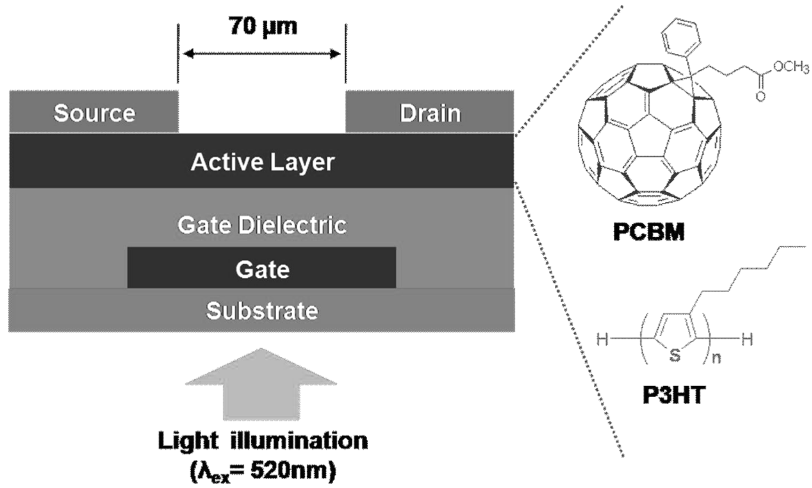
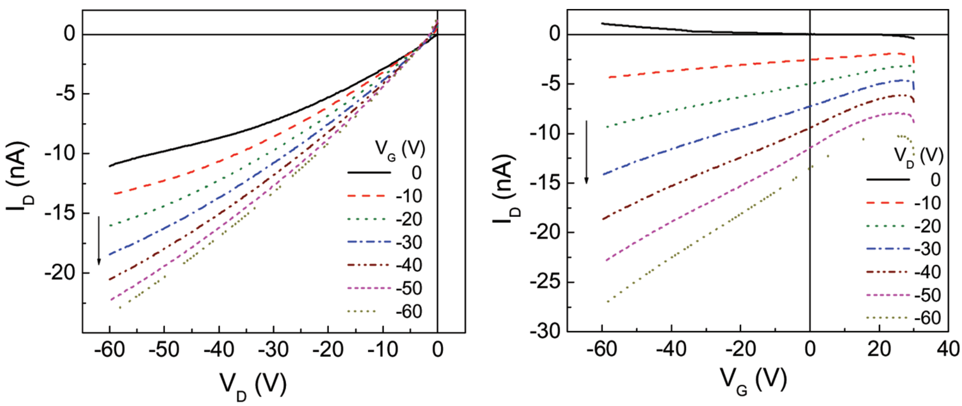


Figure 1. Schematic illustration of device structure and chemical structure of organic materials used in this work.

charges) generated by the large offset energy (0.7 eV) between donor (P3HT) and acceptor (PCBM) [2]. Nevertheless, the drain current was increased as the gate voltage increased at a fixed drain voltage, whilst the slope of $I_D - V_G$ became steeper as the drain voltage became higher (see transfer curves in Fig. 2b). This evidences that the present active layer (P3HT:PCBM) plays a *p*-type semiconductor role in spite of the included *n*-type molecules (PCBM). Here we note that almost no *n*-type trend (electron accumulation mode) was observed from the present device, which can be ascribed mainly to the work function of source and drain electrodes (Ag). The calculated hole mobility of the present devices was $\sim 5.3 \times 10^{-6}\,cm^2/Vs$ under dark.

The output characteristics of device under illumination are shown in Figure 3a. Compared to the dark output curves (Fig. 2a), the drain current under illumination was greatly increased over the entire drain voltages even though the shape of output curves became more deviated from a saturation. This deviation became more pronounced as the gate voltage increased higher. However, the intensity order in drain current with the gate voltage was still maintained under illumination. This result proves that the incident photons indeed involved in increasing the population of hole charge carriers, indicating that the present device did properly function as a phototransistor. As shown in Figure 3b, the apparent responsivity (R_A), which is the ratio of drain current to the incident light intensity, was increased in a sublinear way with the gate voltage at the fixed drain voltage ($V_D = -60\,V$). This nonlinearly increasing drain current is primarily attributed to the influence of dark drain current generated by field effect phenomenon.

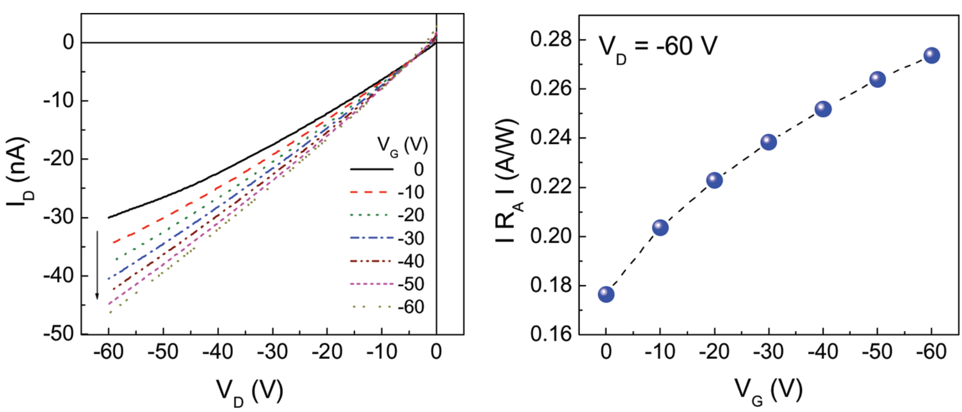
To understand the net effect of incident photons, we calculated the corrected responsivity (R_C) by subtracting the dark current (Fig. 2a) from the overall drain current under illumination (Fig. 3a). As shown in Figure 4a, the R_C value was quite linearly increased at lower drain voltages but it was likely to enter a saturation mode at higher drain voltages. In addition, the R_C value was increased with the gate voltage and its increment became bigger at higher gate voltages.



(a) (b)

Figure 2. Output (a) and transfer (b) characteristics of devices under dark.

Interestingly, the absolute value of R_C showed a two-stage behavior: (1) quick increase up to $V_G = -20\,V$ and (2) saturating tendency above $V_G = -20\,V$ (see Fig. 4b). This might tell us about the correlation between charge amplification and charge recombination according to the intensity of gate voltage: At lower gate voltages the improved charge transport by initial charge amplification played a positive role in quick improvement in R_C, whereas the excess charge generation by further increasing the gate voltage might be involved in the increased charge recombination with photogenerated counter charges. As a result, we achieved the maximum $R_C = \sim 0.13\,A/W$ at higher gate and drain voltages ($V_D = -60\,V$ and $V_G = -60\,V$). This responsivity value is comparable to those (0.005~1 A/W) reported by other research groups who used different types of organic materials including conjugated polymers [6,10,13,14].



(a) (b)

Figure 3. (a) Output characteristics of devices under illumination (520 nm, 81 μW/cm^2). (b) Apparent responsivity (R_A) as a function of gate voltage at $V_D = -60\,V$.

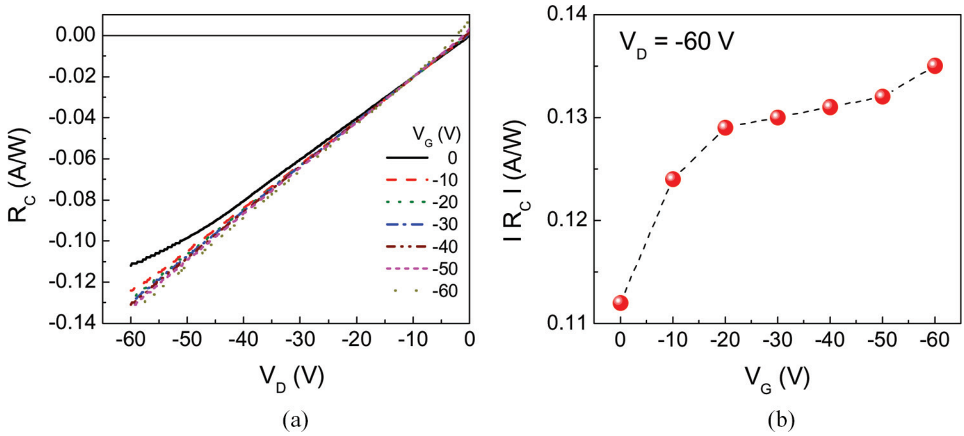


Figure 4. (a) Corrected responsivity (R_C) of devices as a function of drain voltage under illumination (520 nm, 81 μW/cm^2). (b) Corrected responsivity (R_C) as a function of gate voltage at $V_D = -60$ V.

Conclusions

Organic phototransistors have been fabricated using bulk heterojunction films of P3HT and PCBM. The devices showed a typical *p*-type transistor behavior under dark and this nature was maintained under illumination. However, the saturation tendency was weakened under illumination. The apparent responsivity was measured to increase sublinearly with the gate voltage, whereas the corrected responsivity exhibited a two-stage behavior (quick increase-saturation). Consequently, the maximum (corrected) responsivity of present phototransistor reached 0.13 A/W at $V_D = -60$ V and $V_G = -60$ V.

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