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Near Infrared Photoresponsive Organic Field-effect Transistors by Utilizing Pentacene/Lead Phthalocyanine Heterojunction

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Abstract: Photoresponsive organic field-effect transistors (phOFETs) were fabricated by utilizing organic heterojunction based on pentacene and lead phthalocyanine (PbPc). Under a near-infrared light illumination (wavelength 808 nm and a power intensity of 124 mW/cm²), the heterojunction photoresponsive organic field-effect transistors (HJ-phOFETs) exhibited a maximum photosensitivity of 4.4×10^4 , and a maximum photoresponsivity of 118 mA/W, which were 766 times and 785 times higher than that of PbPc single-layer phOFET, respectively. It was observed that the maximum photosensitivity and the maximum photoresponsivity stabilized around 5.4×10^4 and 326 mA/W after 120 h, respectively. The high performance of HJ-phOFET is attributed to the utilization of PbPc as photosensitive layer which has high absorbance in near infrared region (NIR) and pentacene as channel layer with high hole mobility. These results indicate that the HJ-phOFET based on pentacene and PbPc is proved to be a NIR photodetector with excellent photosensitivity and stability.

Key words: heterojunction; pentacene; PbPc; near infrared; photoresponsive organic field effect transistorCLC number: TN386Document code: ADOI: 10.3788/fgxb20143503.0342

基于并五苯/酞菁铅异质结的近红外光敏有机场效应管

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摘要:采用并五苯(Pentacene)和酞菁铅(PbPc)两种有机材料作为有源层,制备了异质结有机光敏场效应 管。在波长为808 nm、强度为124 mW/cm²的近红外光照条件下,异质结 phOFET 获得最大的光暗电流比达 4.4×10⁴,栅压为-50 V时的最大光响应度为118 mA/W,比单层酞菁铅 phOFET 分别高出766 倍和785 倍。 在经过120 h后,器件的最大光暗电流比和最大光响应度分别稳定于5.4×10⁴和326 mA/W 附近。由于在异 质结 phOFET 中采用了对近红外光具有高吸收效率的酞菁铅作为光敏层,而高空穴迁移率的并五苯材料作为 靠近栅介质的沟道层,光生载流子的产生与传输能力得到了有效的提高。实验结果表明,基于并五苯/酞菁 铅的有机异质结应用于光敏有机场效应管的结构设计中,可以使 phOFET 成为一种同时具有良好光敏性及稳 定性的近红外光探测器件。

关键 词:异质结;并五苯;酞菁铅;近红外;光敏有机场效应管

1 Introduction

In recent years, photoresponsive organic fieldeffect transistors (phOFETs) have attracted increasing research interests due to their low cost, mechanical flexibility. Comparing with organic photodiodes (OPDs), phOFETs possess great application prospect because of their higher sensitivity and lower noise levels^[1-3]. The heterojunction structure has been widely used for organic electronic devices to improve their performance, naturally, which is also applied in phOFETs to enhance the device photosensitivity and the transport speciality of carrier^[3-5]. Nevertheless, most organic heterojunctions adopted in the organic electronic devices are anisotype heterojunction, similar to the p-n heterojunctions in inorganic semiconductor devices, which contain two semiconductors with dissimilar types of conductivity. Moreover, the isotype organic heterojunctions consisting of two semiconductors with same types of conductivity have been rarely studied and applied^[6-7]. Although superior performance can be achieved in many organic devices with the anisotype heterojunction structure, most n-type organic semiconductor materials are very sensitive to moisture and oxygen as exposed to air, which may lead to a rapid decline or invalidation of devices. The p-p organic heterojunctions applied in organic eletronic devices can not only achieve high performances but also are able to possess excellent stability, which are beneficial to the researches of phOFETs^[8-10].

In this paper, a near-infrared heterojunction photoresponsive organic field effect transistors (HJ-phOFETs) with pentacene and lead phthalocyanine (PbPc) as active layer were fabricated, in which silver(Ag) was used as the source and drain electrodes. PbPc, as a good NIR photoresponsive material with a strong absorption in the near infrared (NIR) region (780 ~ 900 nm), has been widely applied in organic solar cells (OSCs) and NIR photodetectors^[11-12]. Pentacene is a typical hole transport organic material with high mobility, and the hole mobility of single crystal pentacene OFET reaching up to 35 cm² · V⁻¹ · s⁻¹ was reported^[13].

Pentacene is very suited for using as an active layer in red photodetectors due to its high absorption in red light^[14]. Therefore, it can be considered that PbPc plays the part of the NIR photosensitive layer and pentacene acts as the channel transport layer in the structure of HJ-phOFETs. Under NIR illumination, photo-induced excitons are generated in PbPc layer and diffuse to other zone due to concentration gradient. The diffused excitons nearby the source electrode dissociate into free electrons and holes under the strong electric field of electrode/semiconductor interface, and then the free holes transport in the pentacene channel layer. The HJ-phOFETs, in which generation of photo-induced excitons and transportation of carriers occur in different active layers, show high photoresponsivity and excellent stability.

2 Experiments

Pentacene and PbPc were purchased from J&K Chemical Ltd., and Sigma-Aldrich Ltd., respectively, and used as received. Bottom-gate top-contact geometry was adopted to fabricate the phOFETs, as shown in the inset of Fig. 1. A heavily n-doped Si substrate with a resistivity of 0.03 $\Omega \cdot$ cm acted as the gate electrode, on which there was a 950 nm thermally grown SiO₂ layer as the gate dielectric. The substrates were ultrasonically cleaned by acetone, ethanol and de-ionized water, and was dried with N₂ gas blowing and baked in an oven at 60 °C for 30 min. Then the surface of cleaned SiO₂ was treated with octadecyl silane (OTS) by vapour deposition



Fig. 1 UV-Vis absorption spectra of PbPc (dash) and pentacene (solid) thin film. Inset: Schematic structure of the HJ-phOFET.

in an oven at 120 °C for 2 h. For the device fabrication, a 50-nm-thick pentacene film was deposited on the top of SiO₂ substrate by vacuum thermal evaporation. The vacuum was kept at 2×10^{-3} Pa and the substrate temperature at 65 °C during the deposition. Then a 20-nm-thick PbPc film was vacuum deposited on the top of the pentacene film. Ag as source/drain electrodes was vacuum deposited through a shadow mask which defined a channel length(*L*) and width(*W*) of 25 µm × 2 mm. Meanwhile, single-layer OFETs with pentacene and PbPc as the active layers respectively were fabricated for comparison.

For optical absorption measurement, single-layer thin films of pentacene (50 nm) and PbPc (20 nm) were vacuum deposited on cleaned quartz substrates, respectively. TU-1901 spectrometer was used for the measurements of absorption spectra. For the measurement of photosensitive effects, a NIR laser diode with a wavelength of 808 nm and a power intensity of 990 mW/cm² was used. Different neutral filters were used to obtain variable optical power intensity for illumination. The measurement was implemented in a vacuum environment (<10 Pa), and all devices were preserved under a vacuum of about 10 Pa.

3 Results and Discussion

As shown in Fig. 1, the absorption spectrum of the PbPc film exhibits strong absorption in the NIR region, while the pentacene film exhibits strong absorption in the visible light region and no significant absorption in the NIR region. Hence, the absorbance of organic heterojunction based on pentacene/ PbPc at 808 nm is comparable to that of a neat PbPc film.

Fig. 2 shows the output and transfer characteristic curves of single-layer PbPc phOFET in the dark and under illumination. Typical p-type operating characteristic is observed. In the dark, the drain current $I_{\rm d}$ is merely -0.6 nA at the gate-source voltage $V_{\rm gs} = -50$ V and the drain-source voltage $V_{\rm ds} = -50$ V. For illumination with a power intensity of 124 mW/cm², $I_{\rm d}$ increases to -2.4 nA at $V_{\rm gs} =$ -50 V and $V_{\rm ds} = -50$ V, which is 4 times the size of that in the dark. This result indicates that there is an evident photoresponse of PbPc phOFET under NIR illumination. According to theory of FETs, the saturation drain current is given by:

$$I_{d,sat} = \frac{W}{2L} \mu_{sat} C_{i} (V_{gs} - V_{th})^{2}, V_{ds} \ge V_{gs} - V_{th},$$
(1)
$$\int_{\frac{1}{2}}^{0.5} \int_{-0.5}^{(a)} \int$$

Fig. 2 (a) Output characteristics of PbPc single-layer phOFET in the dark (solid) and under illumination (open). (b) Tranfer characteristics of PbPc single-layer phOFET.

from where threshold voltage $V_{\rm th}$ and the saturated carrier mobility $\mu_{\rm sat}$ of phOFETs can be extracted^[15]. The threshold voltage of PbPc phOFET in the dark($V_{\rm th,dark}$) is -48 V and the saturated holes mobility in the dark($\mu_{\rm sat,dark}$) is 5.5×10^{-5} cm² · V⁻¹ · s⁻¹. Under illumination, $V_{\rm th}$ shifts forward to -33 V and $\mu_{\rm sat}$ increases to 5.8×10^{-5} cm² · V⁻¹ · s⁻¹. The variation of $V_{\rm th}$ may result from the photogenerated holes accumulated at the interface between electrode and semiconductor and at the interface between gate dielectric and semiconductor under illumination^[16].

An important parameter of phOFETs is photosensitivity (P), which is defined as the ratio of the photocurrent to the dark current^[15]:

$$P = \frac{I_{\rm ph}}{I_{\rm dark}} = \frac{I_{\rm ill} - I_{\rm dark}}{I_{\rm dark}}, \qquad (2)$$

where $I_{\rm ph}$ is the photocurrent, $I_{\rm dark}$ is the drain current in the dark and $I_{\rm ill}$ is the drain current under illumination. Another important parameter is photoresponsivity $(R)^{[15]}$, defined as following:

$$R = \frac{I_{\rm ph}}{P_{\rm opt}} = \frac{I_{\rm ill} - I_{\rm dark}}{P_{\rm inc}A}, \qquad (3)$$

where P_{opt} is the effective optical power on the channel of the device, P_{inc} is the incident optical power intensity, A is the effective illuminated area, namely, the channel area of device $(L \times W)$. Enhancement of photosensitivity and photoresponsivity of phOFETs is beneficial to lower the noise levels of devices and improve the precision of photodetectors. The maximum photosensitivity of PbPc single-layer phOFET is calculated from Fig. 2 (b) and Eq. (2) to be 57. And a maximum photoresponsivity of PbPc single-layer phOFET of 0.15 mA/W was obtained at $V_{gs} = -50$ V and $V_{ds} = -50$ V (see Table 1). Ag was utilized as the source and drain electrodes, whose work function (4.26 eV) was seriously mis-matched with the highest occupied molecular orbital (HOMO)



Fig. 3 (a) Output characteristics of pentacene/PbPc HJphOFET in the dark (solid) and under illumination (open). (b) Tranfer characteristics of pentacene/ PbPc HJ-phOFET.

of PbPc $(5.2 \text{ eV})^{[11,17]}$. Therefore, a high barrier is formed at Ag/PbPc interface and which further hindered the holes-injection from source electrodes to PbPc layer. Furthermore, the hole mobility of Pb-Pc thin film is very low, which impedes transferring of the massive photo-induced carriers, as a result, I_{dark} and R of PbPc single-layer phOFET are both poor.

The output and transfer characteristic curves of HJ-phOFET in the dark and under illumination are displayed in Fig. 3. The dark current of HJ-phOFET is -750 nA at $V_{\rm gs} = -50$ V and $V_{\rm ds} = -50$ V, which is 1 250 times larger than that of PbPc singlelayer phOFET. The great improvement of dark current (I_{dark}) at HJ-phOFET operating is attributed to the high mobility of pentacene channel layer. For illumination with a power intensity of 124 mW/cm^2 , the drain current of HJ-phOFET increases to -1 358 nA, as a result the photocurrent ($I_{\rm ph} = 608$ nA) is 337 times larger than that (1.8 nA) of PbPc single-layer phOFET. The $V_{\rm th}$ and $\mu_{\rm sat}$ in the dark of HJ-phOFET are calculated from Fig. 3 and Eq. (1) to be -36 V and 2.5 × 10⁻² cm² · V⁻¹ · s⁻¹, respectively. Under illumination, the V_{th} shifts forward to -31 V and the μ_{sat} changes to 2.7×10^{-2} cm² · $V^{-1} \cdot s^{-1}$. From the data of Fig. 3 and Eq. (2) ~ (3), a maximum photosensitivity of 4.4×10^4 and a maximum photoresponsivity of 118 mA/W are obtained for HJ-phOFET at $V_{\rm gs}$ = -50 V and $V_{\rm ds}$ = -50 V. High hole-mobility pentacene has a HOMO of 5.0 eV^[17], which matches well with that of Pb-Pc. As a result, the free holes dissociated from photo generated excitons can easily transmit to the drain electrode through pentacene conducting channel, which significantly improves the photosensitive characteristics of devices.

The results were obtained at $V_{\rm gs} = -50$ V and under illumination with a power intensity of 124 mW/cm². Stability is an important parameter for photoelectronic devices. Fig. 4 shows the maximum photosensitivity, the maximum photoresponsivity ($V_{\rm gs} = -50$ V and $V_{\rm ds} = -50$ V) and the saturation hole mobility of HJ-phOFET in the dark as function of the storage time in a low vacuum around 10 Pa.

Table 1 Device performance details								
Devices	$I_{ m ph}/$	$R_{ m max}/$	$P_{_{ m max}}$	$V_{ m th,dark}/$	$V_{ m th,ill}/$	$\mu_{ m p,dark}$ /	$\mu_{ m p,ill}/$	
	nA	$(mA \cdot W^{-1})$		V	V	$(\mathrm{cm}^2\cdot\mathrm{V}^{-1}\cdot\mathrm{s}^{-1})$	$(\mathrm{cm}^2\cdot\mathrm{V}^{-1}\cdot\mathrm{s}^{-1})$	
PbPc	1.8	0.15	57	- 48	- 33	5.5×10^{-5}	5.8 × 10 ⁻⁵	
HJ-phOFET	608	118	4.4×10^{4}	- 36	- 31	2.5×10^{-2}	2.7×10^{-2}	

Table 1Device performance details

It can be seen from Fig. 4, the maximum photoresponsivity and saturation hole mobility both increase over time and then stabilize around constant values. After 120 h, the maximum photoresponsivity of HJphOFET increases from 118 mA/W to 326 mA/W, and the maximum photosensitivity increases from 4.4×10^4 to 5.4×10^4 . Furthermore, the saturation hole mobility of HJ-phOFET in the dark shifts from 2.5 × 10⁻² cm² · V⁻¹ · s⁻¹ to 3.5 × 10⁻² cm² · $V^{-1} \cdot s^{-1}$. The saturation hole mobility increase of HJ-phOFET in 120 h later leads to enhancement of the maximum photoresponsivity and the maximum photosensitivity. Because pentacene is a p-type organic semiconductor with good air-stability, and it is reported that phthalocyanines organic materials are also stable materials, thus the HJ-phOFET fabricated based on these can exhibit a good stable performance^[18-21].



Fig. 4 The maximum photoresponsivity at $V_{\rm gs} = -50$ V, $V_{\rm ds} = -50$ V and the maximum photoresponsivity of HJ-phOFET as function of storage time. Inset: The saturated hole mobility of HJ-phOFET in the dark as function of storage time.

Most OFETs operate at unipolar transportation mode, that is, unipolar carriers (electrons or holes) primarily contribute to channel current as OFETs work. The high injection barrier which is resulting from the energy-level mismatch, interfacial effect and series-resistance effect^[22-23] enormously decreases the field-effect mobilities and the drain currents of heterojunction OFETs. By comparison of pentacene single-layer OFET and HJ-phOFET, it is found that the drain current and holes mobility of pentacene single-layer OFET (not here) are both much less than those of HJ-phOFET in the dark. The utilization of Ag as the source and drain electrodes causes an energy difference existing between the work function of Ag and the HOMO of pentacene, and a positive dipole layer is formed at the Ag/pentacene interface, which will result in a hole injection barrier of 0.8 eV at the Ag/pentacene interface, and obstruct the holes injecting into pentacene layer inevitably^[17], consequently, the dark current and hole mobility of pentacene single-layer phOFET are both much lower. When PbPc thin film is inserted between Ag and pentacene, PbPc can be regarded as the buffer layer for Ag and pentacene contact and enhance the injection efficiency of holes greatly^[24]. The electron affinity of PbPc (3.9 eV) is 1.0 eV higher than that of pentacene (2.9 eV), and the energy gap (E_g) of pentacene (2.1 eV) is much larger than that of PbPc (1.3 eV), these may cause that the holes in PbPc layer move to pentacene and a hole depletion layer is formed in $\mathrm{PbPc}^{[7\text{-}8,17]}.$ The hole depletion region in the PbPc layer mainly consisting of negative fixed-charge may lower the injection barrier of holes, in consequence the dark current and hole mobility of HJ-phOFET increase greatly in contrast with the single-layer phOFET. When the HJ-phOFET is illuminated by a NIR light, the operation mechanism of HJ-phOFET can described as: (I) Generation of photo-induced excitons in PbPc layer. (II) Diffusion of photo-induced excitons to the source electrode in PbPc layer. (III) Dissociation of excitons into free holes and electrons at the electrode/ semiconductor interface. (IV) Extraction of electrons by the source and transportation of holes in pentacene layer^[25]. The factors impacting on photosensitive properties of phOFETs are most probably the match of material energy levels, carrier mobility and interfacial effect of organic layers. For PbPc single-layer phOFET and HJ-phOFET, a huge number of free holes generated under illumination may enhance the carrier density in channel, and result in a decline of threshold voltage. However, there is almost no absorption of pentacene in the NIR region around 808 nm, the characteristics of pentacene OFETs are essentially unchanged under illumination. The well match of energy levels of PbPc and pentacene and the high hole mobility of pentacene facilitate the transportation of photo-induced carriers, as a consequence the photosensitive performance of HJ-phOFET is enhanced enormously. However, since pentacene with a high hole mobility is used as the channel layer, the dark current of HJphOFET correspondingly increases and exceeds the value of photo-generated current, which will instead hinders the dissociation of photo-induced excitons and thereby degrades the performance of photosensitive detectors. For this case, it is predicted that the adoption of a p-type organic materials with high photosensitivity and a p-type organic materials with high

mobility will be able to realize the excellent and stabilized phOFETs by appropriate modulation of energy levels.

4 Conclusion

HJ-phOFET based on pentacene and PbPc was fabricated, and its photosensitive characteristics were analyzed compared with single-layer phOFET. The results indicate that better photosensitive performance is achieved in HJ-phOFET than that in single laver phOFET. Under 808 nm and 124 mW/cm² NIR illumination, a maximum photosensitivity of $5.0 \times$ 10^4 and a maximum photoresponsivity of 118 mA/W $(V_{gs} = -50 \text{ V})$ are obtained in HJ-phOFET; while the maximum photosensitivity and photoresponsivity of single-layer phOFET are 57 and 0.15 mA/W, respectively. It is demonstrated that the organic heterojunctions with high hole-mobility pentacene as the channel transportation layer and high absorbance Pb-Pc as NIR photosensitive layer are beneficial to the dissociation of photo-induced excitons and the transportation of holes, which will greatly enhance the photo-induced current and improve the HJ-phOFET performance.

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