

Polymer Chemistry

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Recent advances on polymer phototransistors

Pengcheng Gu,^{a,b,†} Yifan Yao,^{b,†} Linlin Feng,^{a,b} Shujie Niu,^{a,b} Huanli Dong^{a,b,*}Received 00th January 20xx,
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Phototransistor, a kind of optoelectronic devices that realizes the functionality of light detection and signal magnification in a single device, is an important component of optoelectronic integration. Polymer phototransistors, compared to traditional inorganic phototransistors, demonstrate attractive advantages of light-weight, low-cost, easy solution processing and flexibility, and are the the best candidates for *Plastic Optoelectronics*. Over the past years, significant advances have been achieved for polymer phototransistors with the development of new polymer semiconductor materials and the optimization of device fabrication techniques. In this review, we will give an overview of recent advances on polymer phototransistors, from a brief introduction of the device geometry, working mechanism, performance parameters to the development of device performance and the exploration of their flexible optoelectronic devices. Finally, a perspective and conclusion is also briefly addressed.

1 Introduction

Organic semiconductors have attracted considerable attention over the past decades due to their attractive advantages such as light weight, low cost and easy processing, which make them demonstrate potential applications in large-area flexible organic devices/circuits.¹⁻³ Among them, organic field-effect transistors (OFETs), a key component of organic electronics, have received remarkable progress⁴⁻⁶ with the field-effect mobility significantly increased from the initial value⁷ of $10^{-6}\sim 10^{-5}$ to the state-of-art value of $43\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for small-molecule semiconductors⁸ and $36.2\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for polymer semiconductors.⁹ Under the development of high performance OFETs, recently, bi/multi-functional OFETs, where two or more types of physical properties are combined together in a single device, have also received increasing interest due to their scientific and technological significance in organic electronic integration, for example organic phototransistors (OPTs).¹⁰⁻¹⁴ The concept of phototransistors was first proposed by William Shockley in 1951.¹⁵ Compared to photodiodes, phototransistors can exhibit much higher sensitivity and lower noise due to the unique working mechanism of field-effect transistors. Moreover, phototransistors combine light detection and signal amplification together in a single device, and can be easily integrated into various optoelectronic devices/circuits, such as light switchers, light memories and light detectors, etc., as shown in Figure 1.¹⁰

OPTs are generally divided into two types, such

small-molecule OPTs (SOPTs) and polymer OPTs (PPTs) depending on the types of active layers. Recently, the photoresponsivity of SOPTs has already reached up to 12000 AW^{-1} , which is comparable to that of inorganic phototransistors.¹³ Over the past several years, great achievements have also been obtained in PPTs under the considerable efforts of chemical scientists for the development of polymer semiconductors and device physicists for the improvement of device fabrication techniques.^{14,16} In comparison with small molecules, polymer semiconductors exhibit much better solution-processing ability, and compatibility with plastic substrates. Moreover, from molecule perspective, conjugated polymers have much larger conjugation length and broader optical absorption, which enable them more beneficial for optoelectronic devices.^{13,14} In this *MiniRev*, we attempt to present a systematic summary of these significant advances on PPTs. In section 2, we will first give a brief introduction with the purpose of offering an intuitive understanding of OPTs including typical structure geometry, working mechanism and basic performance parameters. In section 3, we will focus on the specific research progress of PPTs including the development of material systems and the improvement of device physics. Finally, the exploration of flexible devices and circuits based on PPTs and concise perspectives and conclusion are also briefly demonstrated.

^a Beijing Key Laboratory for Optical Materials and Photonic Devices, Department of Chemistry, Capital Normal University, Beijing 100048, China.

^b Institute of Chemistry, Chinese Academy of Science, Beijing 100190, China.

† The authors contributed equally.

*Corresponding author email: dhl522@iccas.ac.cn

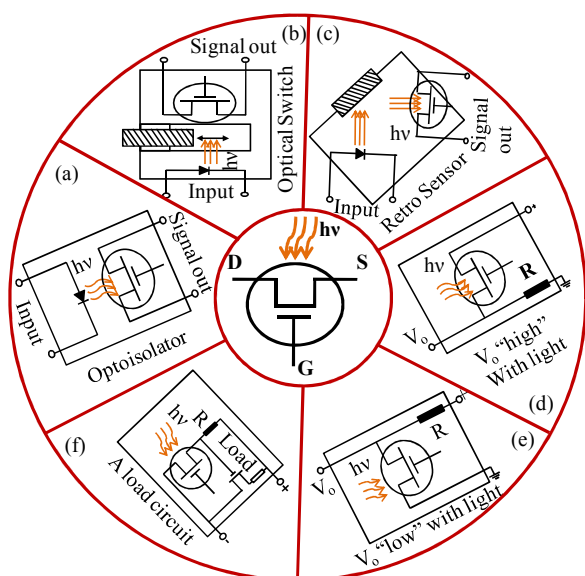


Figure 1. Demonstration of the potential applications of phototransistors. G: gate, D: drain electrode, S: source electrode. (a) optoisolators: the optoisolator is similar to a transformer in that the output is electronically isolated from the input; (b) highly sensitive optical switches: an object is detected when it enters the gap of the optical switch and blocks the light path between the emitter and detector; (c) retro sensors: the retro sensor detects the presence of an object by generating light and then looking for its reflectance off of the object to be sensed; (d-e) basic circuits; and (f) a circuit with greater voltage-switching capability. Copyright with permission from ref. [10] Copyright 2007, John Wiley & Sons.

2 A brief illustration of OPTs

In order to give a comprehensive understanding of OPTs, in this section, we will first briefly outline the fundamentals of device structures, working principles and basic performance parameters for OPTs.

2.1 Device structures of OPTs

The device structures of OPTs are mainly derived from that of OFETs, and also consist of four typical structures: bottom-gate/top-contact (BGTC), bottom-gate/bottom-contact (BGBC), top-gate/top-contact (TGTC) and top-gate/bottom-contact (TGBC), according to the relative positions of dielectric layer, active organic layer and source/drain electrodes. The only difference for OPTs from OFETs is the fourth terminal, light illumination incorporated in the device geometry. According to the position of light illumination, the structures can be further divided into top-light illumination (TL-) and bottom-light illumination (BL-), as shown in Figure 2. It should be stated that whether for TL- or BL- geometries, the gate dielectric layer used in these structures, such as TL-TGBC, TL-TGTC, BL-BGTC and BL-BGBC should be transparent enough in order to guarantee the light efficiently

arriving on the active organic semiconductor layer for high device performance.

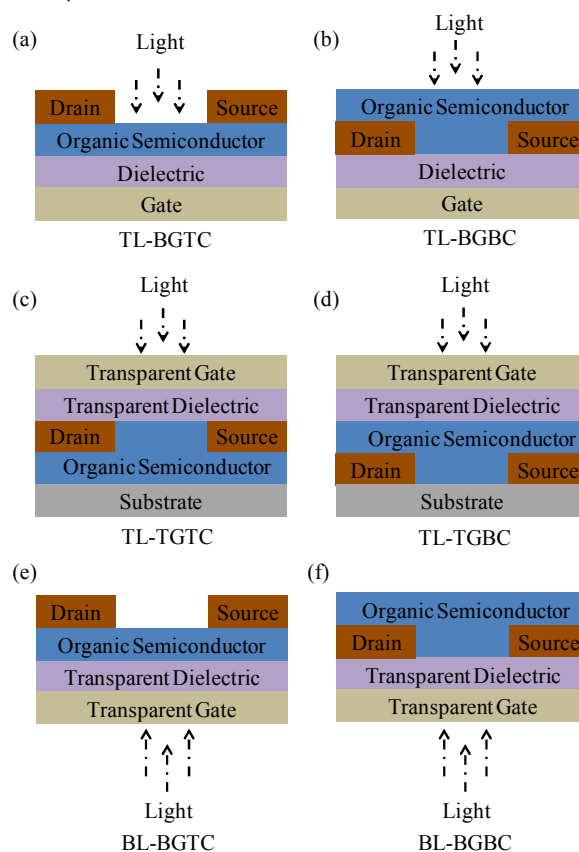


Figure 2. Typical device geometries used for OPTs, specifically including six types (a-f) according to the relative positions of active semiconductor layer, source/drain electrode, dielectric layer and light illumination.

2.2 Photoelectrical characteristics of OPTs

Similar with OFET measurement, transfer and output curves measured under dark and light illumination are two typical characteristics of OPTs, as shown in Figure 3a and 3b (an PPT constructed based on a donor-acceptor copolymer of HXS-1, detailed molecular structure is shown in Scheme 1).¹⁷ Under light illumination, an obvious increase of source-drain current compared to that in dark condition is observed in the corresponding transfer and output curves due to the contribution of photocurrent. Moreover, in OPTs, the incident light can be used as an independent variation to tune the transistor operation, like the function of gate bias in OFETs, working as a typical light and electric bi-functional device (as shown in Figure 3b).

2.3 Working principles of OPTs

There are generally two typical operation models in OPTs reflected by the relationship of photocurrent (I_{ph}) and incident optical power (P_{opt}): photovoltaic mode and photoconductive mode,¹⁸ which will be discussed in detail below.

Photovoltaic mode. When photovoltaic effect is dominant in OPTs, the shift of threshold voltage (V_{th}) is towards more positive (negative) values for p-channel (n-channel) devices when under light irradiation. The photo-generated excitons in the semiconductor dissociate into free charge carriers (electrons and holes) under extra illumination. These photo-generated electrons and holes accumulate under drain and source electrode, respectively. As a result, the large number of trapped photo-generated electrons effectively lowers the potential injection barrier between the source electrode and organic semiconductor surface. Moreover, contact resistance is significantly decreased under the effect of light irradiation. Thereby, the lowered injection barrier and contact resistance result in a positive shift of threshold voltage (V_{th}) and a significant increase in the drain current (I_d). The photocurrent caused by photovoltaic effect can be described in Figure 3c,¹⁹ which can be derived according to the following equation (1)²⁰:

$$I_{ph,pv} = G_M \Delta V_{th} = AkT \cdot [\ln(1 + \eta q \lambda P_{opt} / I_{pd} hc)] / q \quad (1)$$

Where η is the photo-generation quantum efficiency, P_{opt} is the incident optical power, I_{pd} is the dark current for minority charges, hc/λ is the photon energy, G_M is the trans-conductance, ΔV_{th} is the threshold voltage shift, and A is a proportionality parameter.

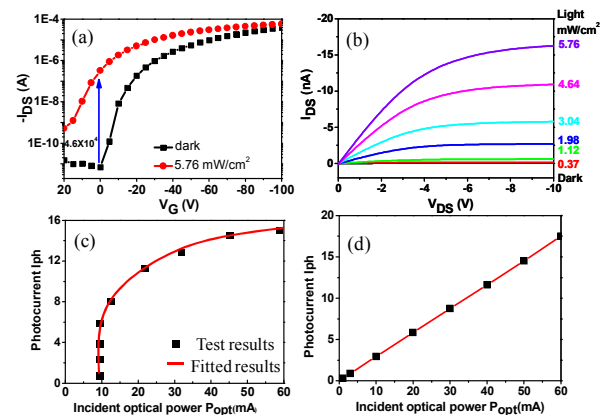


Figure 3. Photoelectrical characteristics of OPTs: (a) transfer and (b) output curves under dark and light illumination for a PPTs based on HXS-1 polymer semiconductor. [17] Copyright 2011, John Wiley & Sons. Two classical working modes: (c) photovoltaic effect and (d) photoconductive effect in phototransistors. [19] Copyright 2004, American Institute of Physics.

Photoconductive mode. When the transistor operates in the depletion region ($V_g > V_{th}$), photo-generated I_d appears a linear increase with optical power, also called photoconductive effect. It is similar to the two-terminal devices without the effect of gate voltage. The transverse electric field induced by the extra gate voltage will contribute to the enhancement of photo-generated carriers. Typical characteristic of photocurrent as a function of the incident optical power under

photoconductive effects is shown in Figure 3d.¹⁹ The current can be calculated according to equation (2):

$$I_{ph,pc} = (q \mu_p E) W D = B P_{opt} \quad (2)$$

Where μ_p is charge carrier mobility, p is charge carrier concentration; E is the electric field in the conducting channel, and W is the gate width, D is the depth of absorption region, and B is a proportionality factor.

2.4 Performance parameters of OPTs

Except for the basic characteristic parameters of OFETs, such as charge carrier mobility (μ), threshold voltage (V_{th}) and on/off ratio, photoresponsivity (R) and photosensitivity (that is photocurrent on/off ratio) (P) are another two basic parameters for evaluating the phototransistor performance. The R and P values can be calculated through the following equations (3) and (4) based on their corresponding transfer curves:

$$R = (I_{DS,i} - I_{DS,d}) / (S P_i) \quad (3)$$

$$P = (I_{DS,i} - I_{DS,d}) / I_{DS,d} \quad (4)$$

Where $I_{DS,d}$ and $I_{DS,i}$ are the drain current in dark and under illumination, respectively. S is the effective area of devices and P_i is the power of incident optical in unit area.

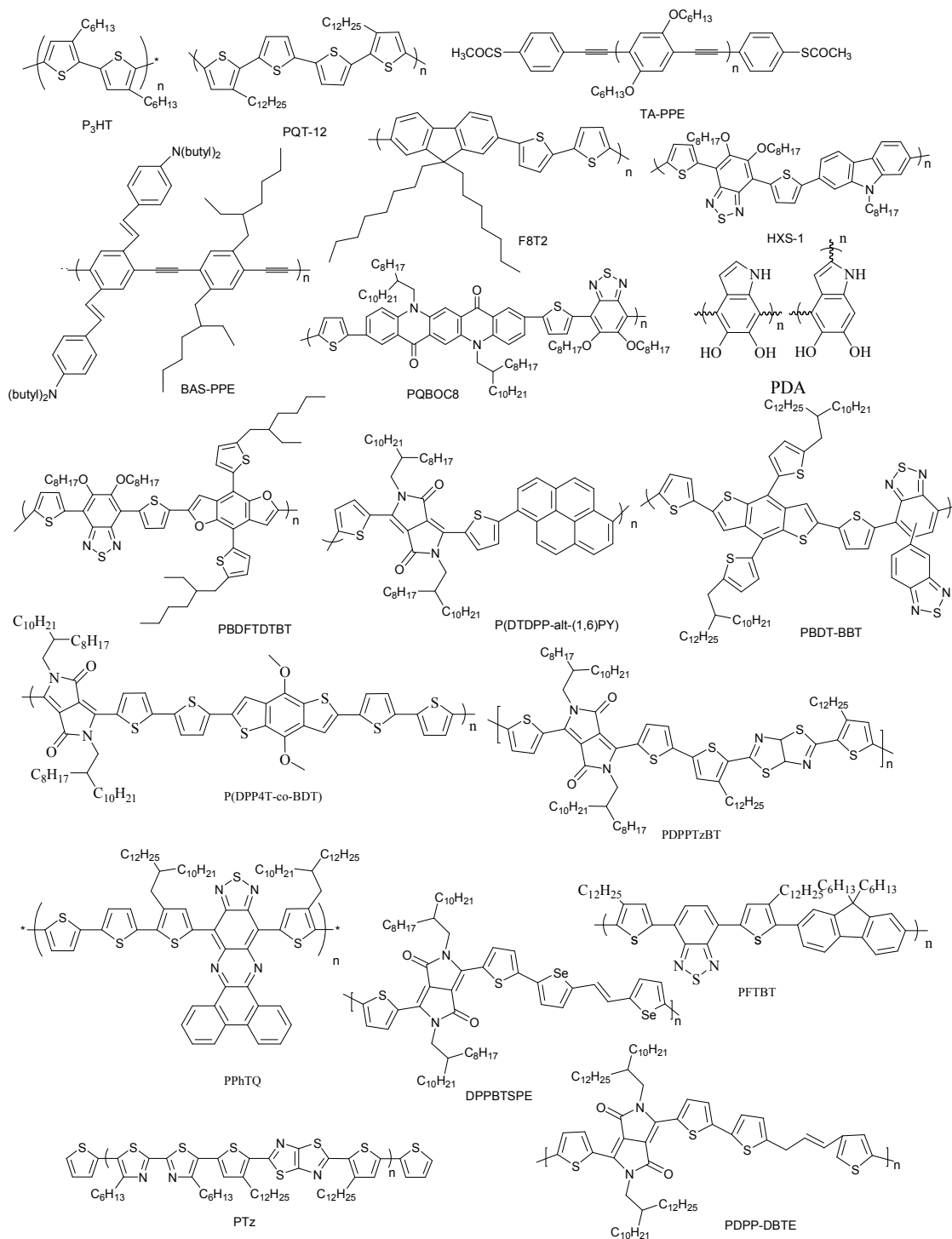
3 Recent advances on PPTs

The first PPT in literatures was demonstrated by Narayan's group in 2001, which was constructed based on BL-BGTC geometry with poly(3-octylthiophene-2,5-diyl) (P3OT) as the active layer and polyvinylalcohol (PVA) as the transparent gate dielectric layer.²¹ This P3OT-based PPT demonstrated a R value of 1 A W^{-1} , which was higher than that of its corresponding planar two-terminal photo detectors, however, it was still obviously lower than that of inorganic phototransistors and SOPTs,^{10,18} probably due to the slow relaxation process in polythiophene layer.²² So, in following, how to further improve the device performance of PPTs has been the research focus in this field. With the development of polymer semiconductors⁴ and device fabrication techniques of OFETs,⁵ significant improvement has also been achieved for PPTs. A timeline of literature publication numbers and device performance of PPTs as a function of years are simply depicted in Figure 4. Obviously, over the past 14 years, the PPT device performance, with P and R as parameter references has undergone remarkable increase with tens and even more than two orders of magnitude, and the highest P and R value of PPTs is approaching $10^{8[49]}$ and $\sim 1 \times 10^{6[49]}$ A W^{-1} , respectively.

The basic requirements for polymer semiconductors used in PPTs are high carrier mobility and wide absorption, in order to facilitate the whole process of effective light absorption, exciton dissociation and charge carrier transport like that in polymer solar cells.²³ Scheme 1 shows the chemical structures of typical polymer semiconductors recently used in PPTs. It can be seen that the polymer material systems are becoming more and more rich, including not only the classical semiconducting polymers,

such as P3HT, P3OT, TA-PPE, etc., but also the recently investigated field-effect donor-acceptor (D-A) copolymers. Besides polymer semiconductor themselves, device fabrication techniques and device structures are also crucial for the

resulting device performance. In following, we will give more detailed discussions for the development strategies of high performance PPTs according to the types of active layers used.



Scheme 1. Chemical structures of polymer semiconductors recently used in PPTs.

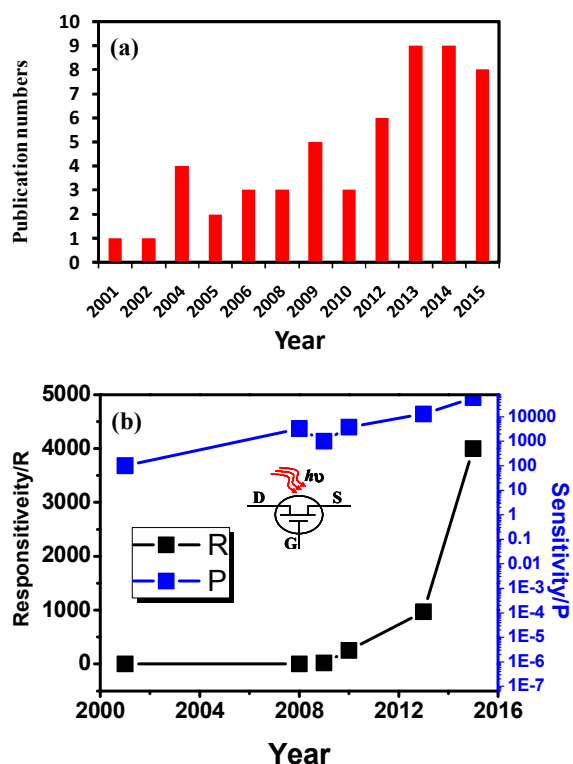


Figure 4. The numbers of publications and R and P values of PPTs as a function of years

3.1 PPTs based on single component film of polymer semiconductors

PPTs based on single component film of polymer semiconductors, are referring to those devices where the active layer is composed of only one kind of polymer semiconductor, which is also the simplest device structure. The first reported PPT was constructed based on such a device structure, where spin-coated 100 nm P3OT film used as the active layer.²¹ This device demonstrated a P value of ~ 100 under 532 nm light illumination at 1 mW cm^{-2} , which was further increased to $\sim 10^3$ for devices based on a thicker P3OT film (150 nm). Subsequently, PPTs based on P3HT or other P3AT single component films were further constructed by the same group. Unexpectedly, polythiophene-based PPTs usually exhibit a slow relaxation of photo-induced excess charge carriers, resulting in a slow decay rate upon switching off the photoexcitation.²² One explanation is because that the photo-induced charge carrier generation depends on the V_{GS} , light intensity and temperature, while the transport characteristics are controlled by the carrier mobility and factors influencing the local built-in electric field parameters.²⁴ Such a concept of slow relaxation of excess charges in OFETs was further used for construction of memory devices by Dutta *et al.*, where the memory states was controlled by V_{GS} applied on the OFETs, realizing the write, store and erase processes (Figure 5a).²⁵ Subsequently, Wasapinyokul *et al.*, fabricated PPTs based on a single component of PQT-12 film and systematically investigated the effect of incident light intensity

on device optoelectronic properties.²⁶ Under light illumination with a fixed wavelength but various intensities from dark to $1100 \mu\text{W cm}^{-2}$, the authors found that for PQT-12-based PPTs, the source-drain current demonstrated relatively weak dependence on the light illumination intensity, where the current first increased with the increase of light intensity but then decreased after a threshold value at a high light intensity, due to the oversaturation behavior of electron-hole recombination under light illumination with high power.

Poly(*para*-phenylene ethynylene)s (PPEs) are another type of polymer semiconductors early investigated in PPTs, which possess ideal rigid-rod conjugated structure and conductivity, and are usually used as molecular wires in nano/molecular devices.²⁷⁻²⁹ In 2004, Xu *et al.*, constructed PPTs based on single component film of a cross-conjugated polymer, 2,5-bis(dibutylaminostryl)-1,4-phenylene-b-alkyne-b-1,4-bis(2-ethylhexyl)benzeneterpolymer (BAS-PPE) with a BL-BGBC geometry.²⁷ Typical photoelectrical characteristics were observed for BAS-PPE based PPTs, giving a high P value of 6×10^3 and a R value of 5 mA W^{-1} . In following, Nakashima *et al.*, synthesized another novel rigid-rod PPE derivative with thioacetylene-functionalized groups (TA-PPE).³¹ Hu *et al.*, first fabricated polymer nanoscale photoswitches and phototransistors based on the self-assembled TA-PPE molecules and planar nanogap electrodes. With light illumination switching on and off, these devices exhibited typical on and off states, with a fast switching rate of 400 Hz and high P ratio of $\sim 10^3$ under the effect of tunnelling current.³² Dong *et al.*, further fabricated PPTs based on TA-PPE films with a TL-BGTC device structure, as expected which also demonstrated high device performance with a P ratio of 3.3×10^3 , and R value of 36 mA W^{-1} at $V_{GS}=0 \text{ V}$.^{33,34} Moreover, the optoelectronic properties for polymer thin films is significantly affected by the molecular orientation in solid state, and the higher orders of polymer chains are, the higher the device performance is.³⁴ Additionally, different from P3AT-based devices,^{22,24} PPE-based phototransistors and photoswitches both exhibit relatively fast on and off switching rate under the effect of light illumination on and off.^{34,35} Two effects, photoconductive and photovoltaic effects, were both characterized in the same transistors, depending on the gate bias. With gate bias, the polymer phototransistors were dominated by photovoltaic effect, while without gate bias, they were dominated by photoconductive effect.

Recently, D-A copolymers have also been applied into PPTs due to their attractive advantages of high charge carrier mobility and broad absorption region.^{23,36} Poly(9,9-dioctylfluorene-co-bithiophene) (F8T2) is the first copolymer used in PPTs. Hamilton *et al.*, systematically studied the photoelectrical properties of F8T2-PPTs based on a BL-BGBC structure,³⁷ which demonstrated a significant increase of current in the off-state, while a small change in the on-state under light illumination, attributed to the different effect of photoconductive and photovoltaic. A R value of 0.7 mA W^{-1} in

the strong accumulation region and a P value of $\sim 10^3$ in the off-state were obtained for F8T2-PPTs. Further, the authors examined the effect of light wavelength on the device performance of F8T2-based PPTs. It was found that high photo-energy photons produced by short wavelength were easier to be absorbed by the polymers, thus giving high density of charges in the active layer and high device performance of PPTs.³⁸ Later, a higher R value of 18.5 A W^{-1} was further obtained for F8T2-PPTs by Wang *et al.*³⁹ Inspired by previous works, more recently, a variety of narrow-band gap D-A copolymers have been used as the active layer in PPTs. For example, the large planar benzo[1,2-b:4,5-b']dithiophene (BDT) unit is an attractive building block for design of novel D-A conjugated polymers. In 2009, Qin *et al.*, designed and synthesized a novel BDT-based copolymer (HXS-1), which demonstrated very high photovoltaic performance with a power conversion efficiency (PCE) of over 5%, due to its close molecular packing in the solid state and a wide range of visible absorption.⁴⁰ Subsequently, Dong *et al.*, fabricated PPTs based on the thin films of HXS-1 polymer and obtained a high charge carrier mobility of $0.06 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, on/off ratio of over 10^7 as well as high photoelectric property with a P value of $\sim 4.6 \times 10^4$, which is also the highest P value in its reported time.¹⁷ Huang *et al.*, fabricated PPTs based on another BDT-based copolymer,

poly{4,8-bis(2'-ethylhexylthiophene)benzo[1,2-b:3,4-b']difuran-5,6-bis(4',7'-di-2-thienyl-5',6'-dioctyloxy-2',1',3'-benzothiazole) (PBDFTDTBT), and achieved a P value of 1.2×10^5 and R value of 360 mA W^{-1} under white light illumination.⁴¹ Li *et al.*, constructed PPTs based on the self-assembled ultrathin crystalline copolymer films of PQBOC8 copolymer,⁴² which showed a R value of 970 A W^{-1} , and a P value of 1.36×10^4 under a very low white light irradiation (43 mW cm^{-2}). Another BDT-based copolymer, PBDT-BBT of 4,8-bis(2-thienyl)benzo[1,2-b:4,5-B']dithiophene and 5,5'-bibenzo[c][1,2,5]thiadiazole with a wide absorption of 300-800 nm has recently also been used in PPTs, which demonstrated a R value of 3200 mA W^{-1} and a P value of 4×10^5 .⁴³ Diketopyrrolopyrrole (DPP)-based semiconductors are another type of promising building block for copolymer materials, due to their easily tuned structures, solubility and high crystallinity in solid state.⁴⁴ In 2013, Yanget *et al.*, designed and synthesized two novel DPP-based copolymers, P(DTDP-alt-(1,6)PY) and P(DTDP-alt-(2,7)PY) (with the different anchoring positions in the pyrene).⁴⁵ These two polymers exhibit not only good charge transport performance with carrier mobility of $0.23 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ but also high photoelectrical property with a high P value of 1.2×10^5 and R value of 360 mA W^{-1} in

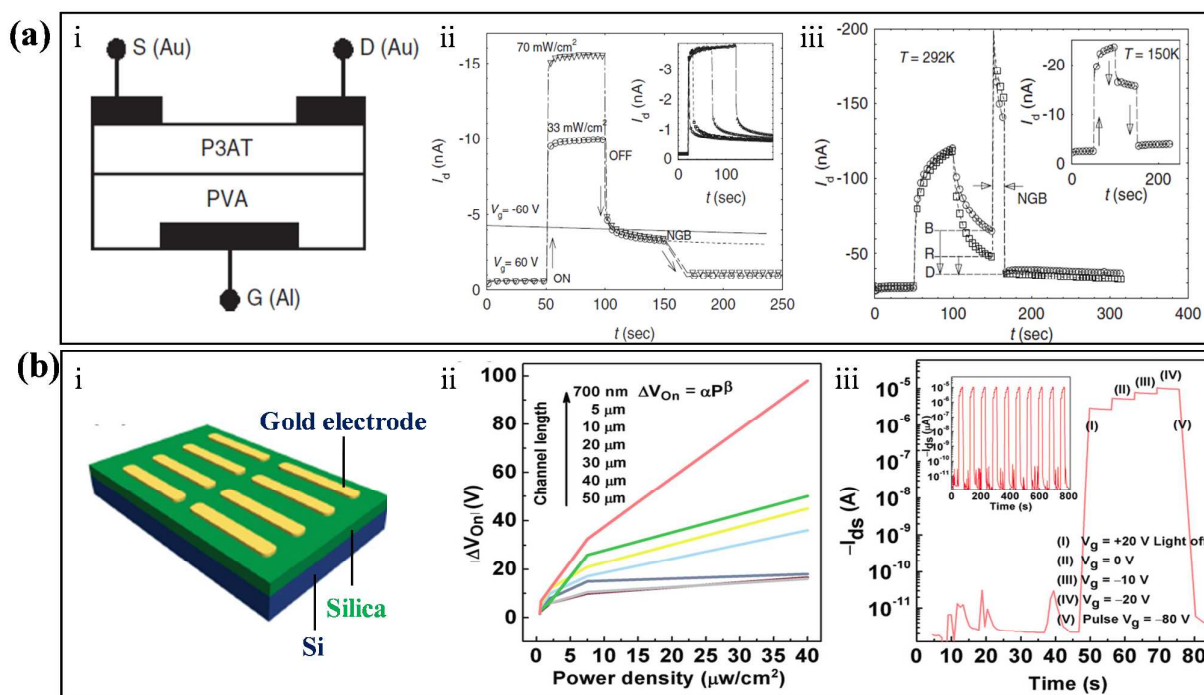


Figure 5. Slow relaxation effect in PPTs for exploration of memory applications. (a-i) Schematic of the PPTs based on P3AT active layer with a TL-BGTC geometry, (a-ii) Memory effect in P3AT-based PPTs measured at 20 K, which can be modulated by pulse gate voltage, (a-iii) Memory effect under different wavelength of light illumination. (b-i) Short channel gold electrode arrays for PPTs with PDPTzBT thin film as active layer, (b-ii) Positive shift of onset gate voltage versus power of illumination with decreasing channel-lengths, (b-iii) Detailed process of a light-writing and erasing cycle. To test its reproducibility, ten cycles are conducted repeatedly with light intensity of $7.5 \mu\text{W cm}^{-2}$ and V_{ds} of -20 V (inset). [25] and [49] Copyright 2004 and Copyright 2014, John Wiley & Sons.

PPTs. Recently, Zhan *et al.* designed and synthesized a novel DPP-based copolymer, P(DPP4T-co-BDT) by the polymerization of

3,6-bis(5'-bromo-[2,2'-bithiophen]-5-yl)-2,5-bis(2-octyldodecyl)pyrrolo-[3,4-c]-pyrrole-1,4(2H,5H)-dione and 2,6-bis(trimethyltin)-4,8-dimethoxybenzo[1,2-b:3,4-b']dithiophene.⁴⁶ The copolymer P(DPP4T-co-BDT)-based thin film PPTs showed excellent photo-response property with R up to $4 \times 10^3 \text{ A W}^{-1}$ (the highest value to date) and P up to 6.8×10^5 under a low intensity of light illumination, which was much higher than that of inorganic single crystal silicon phototransistor. Nam and co-workers demonstrated OPTs based on a novel dense and conformal surface PDA thin film, which was produced using self-polymerization of a homogenous PDA solution under oxygen bubbling instead of a conventional air stream.⁴⁷ Device measurement results showed that the PDA thin film device performance could be tailored by adjusting the incident light intensity and gate voltage (with the highest R value of A W^{-1} at $V_G = 12 \text{ V}$ and P value of 6.9×10^4 at $V_G = -2 \text{ V}$) and a prompt reproducible photo-switching property was obtained for this novel PDA-based OPTs with a fast response rate (rise and decay time: $\sim 0.5 \text{ s}$ and 1.2 s , respectively). Li *et al.*, fabricated PPTs based on a novel ambipolar D-Acopolymer (PPhTQ), and achieved a maximum photo-responsivity of 400 A W^{-1} and a balanced hole ($0.09 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) and electron ($0.06 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) charge transport mobility.⁴⁸ Except for the development of polymeric materials, the advances of device fabrication techniques, such as adopting short-channel geometry in devices, also could enable high performance of PPTs. More recently, Wang *et al.*, successfully constructed an ultrasensitive and high performance PPT based on a short-channel device geometry with a BGTC structure and diketopyrrolopyrrole-thiazolothiazole copolymer (PDPPTzBT) as the active layer.⁴⁹ A series of short conducting channel arrays were constructed through a simple and effective 'coffee ring lithography' inkjet printing technique via a $50 \mu\text{m}$ orifice nozzle, giving the channel lengths from several micrometers down to 700 nm (Figure 5b-i). A high carrier mobility of $1.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, and photo-responsivity with an extremely high R of 10^6 A W^{-1} were obtained for PDPPTzBT-based PPTs with 700 nm channel length. Moreover, it is found that the onset gate voltage (ΔV_{on})

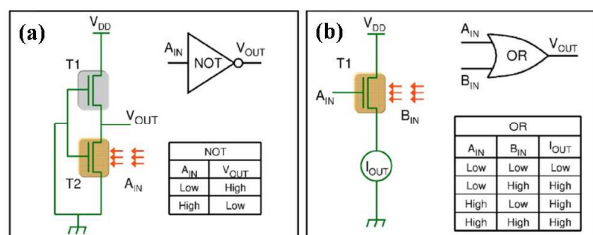


Figure 6. Logic circuits based on ambipolar OPTs and unipolar OFETs. (a) Circuitry of the electro-optical NOT gate together with its symbolic representation and the truth table. (b) Circuitry of the electro-optical OR gate together with its symbolic representation and the truth table. The circuit combines two

inputs, one electrical (A IN) and one optical (B IN). Copyright with redraw permission from ref. [51] Copyright 2007, American Institute of Physics.

is strongly dependent on the channel length, that is, the reduction of channel lengths is accompanied by an increasing trend of ΔV_{on} , due to modulation of the gate voltage (Figure 5b-i). A gigantic shift of onset voltage (V_{on}) was observed in the sub-micron channel transistors, which was mounting to 90 V under the illumination of $40 \mu\text{W cm}^{-2}$. Additionally, PDPPTzBT-based PPTs demonstrated relatively long recovery time and exhibited an unique characteristic of persistent photoconductivity with a stable intermediate current state after switching off the light illumination. Such characteristic combining its gate controllable property enables these devices to function as reproducible light-triggered memory devices (Figure 5b-ii), where by applying appropriate light illumination and pulse gate voltage, the writing process, different memory states and erase process could be realized in a single device.

3.2 PPTs based on blend films of polymer semiconductors

The combination of bulk hetero-junction effect, where donor and acceptor materials are combined together in the active layer, are widely used in organic photovoltaic devices, due to the ultrafast long-range charge separation at the donor-acceptor interface.⁵⁰ Such a concept has also been used in PPTs. Anthopoulos *et al.*,⁵¹ reported light-sensing PPTs based on donor and acceptor blend films of '6,6'-phenyl-C61-butyric acid methyl ester (PC₆₁BM) and poly-2-methoxy-5-3,7-dimethyloctyloxy-p-phenylene vinylene (OC₁C₁₀PPV), which demonstrated not only ambipolar charge transport characteristic but also much faster response time than unipolar OPTs devices. Interestingly, by carefully tuning the ambipolar transport property, their photo-responsivity can be further controlled and improved. The fast response and controlled photo-responsivity property of blend film based PPTs demonstrated their potential applications in electro-optical switchers and logic circuits (Figure 6). Yasin *et al.*, further fabricated organic bulk-hetero-junction based on blend films of P3HT and PCBM and obtained a R value of 3 A W^{-1} at $V_{\text{DS}} = -8 \text{ V}$.⁵² In addition, except for polymer/small molecule blends, polymer/inorganic nanoparticles are also used as the blends in PPTs. In 2008, Mok *et al.* successfully introduced photo-sensitive composite films of P3HT and TiO₂ nanoparticles in PPTs.⁵³ These devices based on P3HT:TiO₂ active blend films demonstrated a rapid change in channel current under illumination due to the electrons trapped by TiO₂ nanoparticles, showing fast response time and high stability. Based on this result, they further deeply investigated the photo-responsive characteristics of P3HT:TiO₂-based devices, and found that their photo-response property was extremely sensitive to weak light due to an integration effect.⁵⁴ Moreover, the photo induced mechanism was investigated clearly that photons first absorbed by P3HT and then excitons effectively separated into free charge at the

P3HT:TiO₂ surface under the illumination of UV light. While excitons generated in the P3HT part and electron were trapped in the TiO₂ nanoparticles under visible light. So, excitations could be generated in the P3HT and TiO₂ nanoparticles, respectively. Therefore, both the visible and UV light illumination could result in electrons accumulated in TiO₂ nanoparticles, thus caused the high photosensitivity of the devices. The concept of using polymer/inorganic nanoparticles in PPT suggests potential applications in wavelength sensors by selecting different semiconducting polymers and inorganic nanoparticles.

3.3 PPTs based on Micro/nano-crystals of polymer semiconductors

As well known, the performance of devices is highly sensitive to solid state crystallinity, crystal grain size and molecular packing of organic semiconductors. However, the inevitable large amounts of defects and grain boundaries in thin films play a negative impact in improvement of device performance.⁵⁵ So, obtaining highly ordered microstructures is another effective approach to further realize the enhancement of device performance, and also beneficial for the integration of micro/nano- optoelectronic devices and circuits. In 2004, Pal *et al.*, reported PPTs based on regular P3HT fibers, which were prepared through high-boiling point solvent of *p*-xylene.⁵⁶ The high crystallinity of P3HT nanowires contributed to the high photoelectrical property of its PPTs with a R value of 250 A W⁻¹ and P value of 3.8 × 10³. Dierckx *et al.*, also fabricated PPTs based on the P3HT fibers and obtained a much higher P value of 6.8 × 10³ due to the further improvement of its crystallinity by optimizing the self-assembly process.⁵⁷ Liu *et al.*, prepared large-area nanowire arrays of poly{(9,9-dihexylfluorene-2,7-diyl)-alt-[4,7-bis(3-dodecylthiophen-5-yl)-2,1,3-benzothiadiazole-2,2'-diyl]}(PFTBT) through traditional anodic aluminium oxide (AAO) template methods by elaborately tuning and controlling the self-assembly of molecules into the AAO pores. PPTs based on PFTBT nanowires exhibited a R value of as high as 1700 mA W⁻¹ and switch P ratio of 2000.⁵⁸ Obviously, incorporating polymer microstructures in PPTs is a good way to improve the device performance because of the reduced defects compared to thin films, however, the crystallinity of these previously reported polymer nanostructures is still not high enough, or even some of them are still in amorphous state.⁵⁹ So, in this case, how to prepare high crystalline polymer micro/nanostructures is crucial not only for construction of high polymer optoelectronic devices but also establishing an accurate platform for repeated probing charge transport and photoelectrical property of polymer semiconductors.

Since 2000s, small-molecule organic single crystals have been widely investigated and remarkable progresses have been achieved in this field, including the growth of high quality organic single crystals, the development of feasible fabrication techniques and the construction of high performance devices, which indeed greatly contributed to the development of organic electronics.⁶⁰⁻⁶² However, how about are crystalline or single

crystals of polymer micro/nanostructures? Actually, it has been a challenging task in polymer science to grow large-size and high-quality polymer crystals, especially for the electron-rich

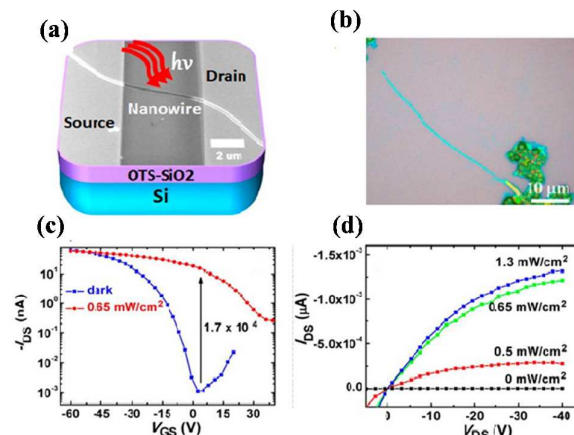


Figure 7. (a) Schematic of PPTs based on conjugated polymer nanowire. (b) Optical microscopy of PTz nanowire on Si/SiO₂ substrate. (c) Transfer characteristics of the PTz-nanowire based PPTs under dark and light irradiation. (d) Output characteristics of the PTz-nanowire based PPTs tuned by different light intensity. Copyright with redraw permission from ref. [67] Copyright 2013, American Chemical Society.

conjugated polymers. In 2006, Kim *et al.*, first reported the single crystal microwires of P3HT polymer, which were prepared through a self-seeding method from a dilute chloroform solution.⁶³ The authors found that in the P3HT microcrystals, P3HT were arranged with their π - π stacking direction along the long axis of microwires. In 2009, Dong *et al.*, reported single crystal micro/nanowires of TA-PPE through a modified solvent assisted self-assembly method,⁶⁴ where a different polymer molecular packing from that of P3HT microwires was observed, i.e., the conjugated polymer backbones of TA-PPE were aligned along the long axis of nanowires.⁶³ The obtainment of high-quality crystals of P3HT and TA-PPE opens up a new area for the investigation of other conjugated polymer crystals as well as corresponding optoelectronic devices. To date, various conjugated polymers have been proved to form crystal microstructures, leading to the significant improvement of charge transport compared to their corresponding thin films,⁶⁵ and the highest mobility value for conjugated polymer crystals obtained recently was up to 24.0 cm² V⁻¹ s⁻¹.⁶⁶ In 2013, Liu *et al.*, first reported PPTs based on crystal microwires of bithiazole-thiazolothiazolecopolymer (PTz),⁶⁷ which were prepared through the solvent assisted self-assembly method similar with that of TA-PPE nanowires.⁶⁴ PPTs based on PTz nanowires exhibited high device performance with hole mobility of 0.46 cm² V⁻¹ s⁻¹ and R of 2531 A W⁻¹ and P of 1.7 × 10⁴ due to its close π -stacking and unique molecular packing with the conjugated backbones aligned to the long axis of nanowires, (Figure 7). More recently, Umet *et al.*,⁶⁶ fabricated PPTs based on single crystals of another D-A conjugated polymer, 1,2-bis(5-(thiophen-2-yl)selenophen-2-yl)ethene (DPPBTSPE),

which also showed high device performance with R of 1920 A W⁻¹ and P value of 10⁴. Much higher device performance is expected for conjugated polymer crystals and more deep

investigations are still needed in this field. Table 1 summarizes the device performance of PPTs discussed in the manuscript.

Table 1. Detailed device performance of PPTs

Polymer semiconductor	Structure(dielectric)	Mobility (cm ² V ⁻¹ s ⁻¹)	R A W ⁻¹ (light wavelength, intensity)	P	Ref
P3OT	BL-BGTC (PVA)	10 ⁻³ -10 ⁻⁴	1 (532nm,1mWcm ⁻²)	10 ² -10 ³	[21]
P3HT	TL-BGTC (SiO ₂)	0.01-0.07	245(Whit light, 51mW cm ⁻²)	3.8×10 ³	[56]
	TL-BGTC (SiO ₂)	NA	250(White light ,0.91mW cm ⁻²)	6.8×10 ³	[57]
PQT-12	BL-TGBC (PMMA/PVP)	1.1×10 ⁻³	6.6(525nm, 3μW cm ⁻²)	~10 ²	[26]
BAS-PPE	TL -BGBC (Polyimide)	NA	0.005 (White light, 4 μW cm ⁻²)	6×10 ³	[30]
TA-PPE	TL-BGTC (SiO ₂)	NA	0.036(Whitelight,5.76 mW cm ⁻²)	3.3×10 ³	[33]
F8T2	TL-BGTC (BCB/SiNx)	3×10 ⁻³	1 (White light,2.9 mW cm ⁻²)	10 ³	[37,38]
	TL-BGBC (SiO ₂)	1.2×10 ⁻⁴	18.5 (465 nm, 5 μW cm ⁻²)	~ 10 ²	[39]
HXS-1	TL-BGTC (SiO ₂)	0.06	NA(White light,5.76 mW cm ⁻²)	4.6×10 ⁴	[17]
PBDFDTBT	TL-BGTC (SiO ₂)	0.05	0.36(White light,1mW cm ⁻²)	1.2×10 ⁵	[41]
PQBOCB	BGTC (SiO ₂)	0.13	970(White light,0.28mW cm ⁻²)	1.36×10 ⁴	[42]
P(BDT-BBT)	TL-BGTC (SiO ₂)	6×10 ⁻³	3.2(White light, 5.51mW cm ⁻²)	4×10 ⁵	[43]
P(DTDP- <i>alt</i> (1,6)PY	TL-BGTC (SiO ₂)	0.23	0.36(White light, 1 mW cm ⁻²)	1.2×10 ⁵	[45]
P(DPP4T-co-BDT)	TL-BGTC (SiO ₂)	0.047	4000(White light, 9.7μW cm ⁻²)	6.8×10 ⁵	[46]
PDA	TL-BGTC (SiO ₂)	0.96	9(White light,19.3mW cm ⁻²)	6.9×10 ⁴	[47]
PDPPTzBT	BGTC(SiO ₂ PMMA)	1.8	10 ⁶ (White light)	10 ⁸	[49]
PPhTQ	TL-BGTC (SiO ₂ ,HMDS)	0.09(hole)	400(White light)	NA	[48]
		0.06(electron)			
P3HT/PCBM	TL-TGBC	1.6×10 ⁻⁴	0.047(Whitelight, 100mW cm ⁻²)	3	[52]
MDMOPPV:PC ₆₁ BM	BGBC (SiO ₂ ,HMDS)	10 ⁻⁴ -10 ⁻³	NA (469 nm)	10 ⁴	[51]
P3HT/TiO ₂	BGBC (SiO ₂)	NA	NA(370-590 nm, 0.2mW cm ⁻²)	~10 ³	[53,54]
PFTBT	TL-BGTC (SiO ₂)	NA	1.7(White light,5.76mW cm ⁻²)	2×10 ³	[58]
PT ₂	TL-BGTC (SiO ₂)	0.46	2531(White light,0.65mW cm ⁻²)	1.7×10 ⁴	[67]
DPPBTSPE	TL-BGTC (SiO ₂)	24	1920(632 nm, 24 mW cm ⁻²)	~10 ³	[66]
PTCDI-C13H27	BGBC (C-PVP)	NA	0.41 (532nm, 22.5mW cm ⁻²)	NA	[68]
PDPP-DBTE	TGBC (PEDOT:PSS)	0.26	NA(White light, 9mW cm ⁻²)	5	[69]

^aPVA: polyvinyl alcohol; PMMA: polymethyl methacrylate; PVP: polyvinyl pyrrolidone; BCB: benzocyclobutene; HMDS: hexamethyldisilazane , ^bNA refers to no related data reported in the literature.

4 Flexible organic phototransistors (FOPTs)

The attractive advantages of light weight, low cost, easy solution processing of SOPTs and PPTs have recently advanced the exploration of their flexible and stretchable devices and circuits. For instance, Chou *et al.*,⁶⁸ presented flexible in-cell devices by incorporating a phototransistor and a photo switch together based on a transparent polyethersulfone (PES)

substrate with a cross-linked poly(4-vinylphenol) as polymer dielectric layer and N,N'-ditridecyl-3,4,9,10-perylenetetra-carboxylic diimide (PTCDI-C₁₃H₂₇) as the active layer. This flexible optoelectronic circuit demonstrated a low operation voltage (3.0 V) and high opto-electrical property with a R value of 0.41 A W⁻¹, indicating its potential application in in-cell touch screens (Figure 8a-c). Kim *et al.*, demonstrated another successful flexible polymer

phototransistors based on transparent and colourless polyimide plastic substrate through an all-solution process, such as roll-to-plate reverse offset printing (ROP), inkjet printing and bar coating (Figure 8d-e).⁶⁹ High crystallinity in the active layer poly[2,5-bis(2-octyldodecyl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione-(E)-1,2-di(2,2'-bithiophen-5-yl)ethene] (PDPP-DBTE) was obtained by optimizing the processing conditions, such as

high-temperature thermal treatment at 180 °C, which contributed to the resulting excellent electrical switching property with a fast response time of 11.2 s and high photosensitivity with photo-to-dark ratio property of 1.54×10^5 in off-state ($V_{GS} = 11$ V). These results provide us hope and confidence for the realization of large-area flexible optoelectronic applications of OPTs.

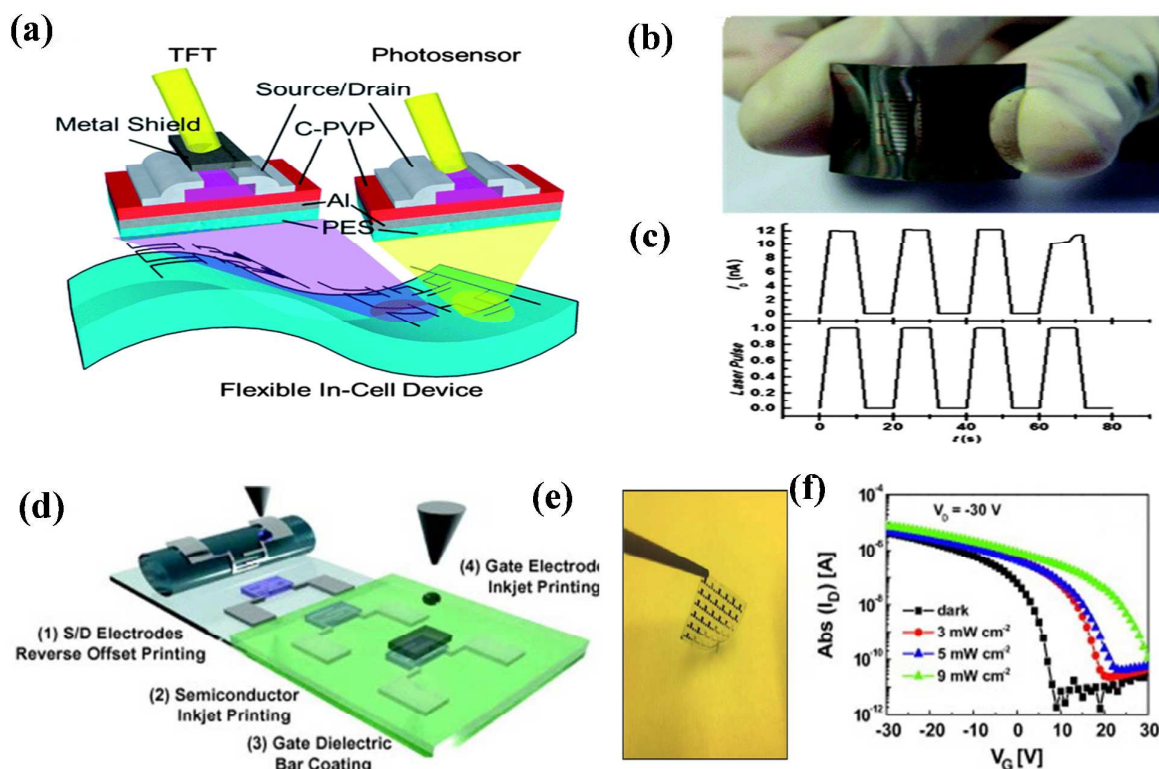


Figure 8. (a) Schematic process flow for fabrication of OPTs based on a combination of printing methods; (b) Transfer characteristics of this flexible device in the dark and under illumination at different light intensity and at $V_D = -30$ V. (c) Schematic of a flexible in-cell device incorporating a photo-OTFT and a switch-OTFT. (d) Photograph of the flexible testing devices. (e) Drain-source current as a function of time (upper panel) then the photo-OTFT is subjected to light pulses (lower panel). [68] Copyright 2014, Royal Society of Chemistry and ref. [69] Copyright 2014, Elsevier B.V.

5 Perspective and conclusion

In this review, we have systematically summarized the development of polymer phototransistors over the past years under the development of polymer semiconductors and device physics. Despite tremendous progress have been demonstrated for polymer phototransistors, there are still large space to further improve the device performance, such as by further (1) enriching the polymer material systems in PPTs, especially incorporating the recently reported high-mobility, narrow-band D-A copolymers; (2) improving the crystallinity of polymer chains

in solid state with appropriate molecular packing; (3) optimizing device structures from the aspects of interface contact and channel sizes; (4) with theoretical calculations to guide the experimental studies. Opportunities and challenges coexist and we believe under the joint efforts of material scientists, physical scientists and theoretical scientists, significant progresses could be further achieved for polymer phototransistors to advance their practical applications, especially in integrated flexible optoelectronic devices/circuits.

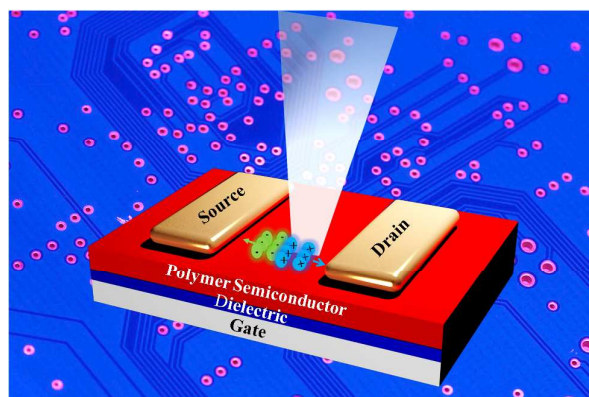
Acknowledgements

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The table of contents entry:**Authors:** P. C. Gu, Y. F. Yao, L. L. Feng, S. J. Niu, H. L. Dong**Title:** Recent advances on polymer phototransistors

In this *MiniRev*, we will give a highlight on the recent advances on polymer phototransistors.