

Polarizing Organic Photovoltaics

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Today's most prevalent information display technology is the liquid crystal display (LCD). Unfortunately, LCDs are energy inefficient, as most of the backlight energy (around 75%) is lost to the orthogonal polarizers. Here, we demonstrate a novel energy recycling concept called polarizing organic photovoltaics (ZOPVs), which can potentially boost the function of an LCD by working simultaneously as a polarizer, a photovoltaic device and an ambient light or sunlight photovoltaic panel. The ZOPV film was created by the uniaxial orientation of an organic conjugated polymer. A novel inverted quasi-bilayer structure was used to produce ZOPV devices. Significant anisotropic optical and photovoltaic effects were obtained, indicating the great potential of ZOPV as a promising green technology.

As an electromagnetic wave (EM), light can be divided into two linear oscillation components: the parallel ($//$) and perpendicular (\perp) polarizations.^[1] They can be separated by a linear polarizer to provide linear polarized light,^[2] and have vast applications. One of the most prevalent applications is for LCD technology.^[3] Figure 1a shows schematically the construction of LCD panels. The fundamental principle underpinning LCD operation is the modulation of light using a combination of two orthogonal polarizers with liquid crystal molecules between these two polarizers to form light valves. Currently, the most commonly used linear polarizer is the absorptive polarizer, which absorbs and wastes the unwanted polarization component while allowing the other component to be transmitted. From an energy point of view, these absorptive polarizers in LCDs are rather inefficient as far as the usage of the backlight photons is concerned. The power consumption of the backlight units takes up approximately 80–90% of the total power consumption in LCD modules.^[4] Unfortunately, most of the backlight energy is lost to these absorptive polarizers (75%). This loss is at a maximum when the pixel displays the color black (this is when the polarizers are completely crossed), since the backlight is still fully on.

In this work, we innovate on the biggest energy loss component, the polarizers, by turning the polarizer into an energy-generating photovoltaic (PV) unit, creating a polarizing organic photovoltaic device (we use the acronym ZOPV in this manuscript rather than POPV, which is often used to refer to polymer organic photovoltaics). A unique advantage of the organic conjugated materials is that the molecular chains can be easily oriented, leading to anisotropic response

to polarized incident light.^[5–8] This feature makes organic PV systems superior to inorganic PV or organic-hybrid PV systems^[9] for the purpose of polarizing PV. ZOPV devices integrated into an LCD panel (Figure 1b) have three potential benefits: i) polarization, whereby the EM wave component with an electric field perpendicular to the oriented molecular chain (*s*-mode polarized light^[7]) propagates through the film without absorption, serving its conventional role in LCDs; ii) as a PV device, the ZOPV film harvests the EM wave component parallel to the molecular chain orientation (*p*-mode polarized light, which is absorbed and wasted in a conventional LCD), converting it into electricity; and iii) ambient light or sunlight PV panel, when the ZOPV device is integrated into the LCD panel, its photovoltaic function remains even when the LCD panel is not in use, producing electricity through conversion of photons from ambient light or sunlight.^[10]

ZOPV films can be realized by the uniaxial orientation of polymer conjugated molecules.^[7,11,12] Here, the polymer used is poly(3-hexylthiophene) (P3HT), which is the most popular *p*-type polymer in organic photovoltaic (OPV) technology.^[13–19] By rubbing P3HT film with velvet cloth at 150 °C, polymer chains were oriented along the rubbing direction, resulting in a polarizing P3HT (z-P3HT) film with anisotropic optical properties. Figure 1c,d compare the anisotropic appearance of z-P3HT film. The pictures were taken by placing a traditional linear polarizer in front of a digital camera as an analyzer. In Figure 1c, the z-P3HT (behaving as a polarizer) and the analyzer had parallel molecular orientation; therefore, the polarized light could pass through both of them, resulting in a transparent appearance. In Figure 1d, the z-P3HT film and analyzer were placed with perpendicular molecular orientation. This resulted in a dark view due to the crossed-polarization by the two polarizers. Subsequent spin-drop-casting of phenyl-C₆₁-butyric acid methyl ester (PC₆₀BM) on the z-P3HT film created a structure, which we term a quasi-bilayer structure. As has been reported earlier,^[20–27] in spite of the sequential spin coating, PC₆₀BM molecules will diffuse into the oriented P3HT matrix forming an interpenetrating layer and giving the P3HT-PC₆₀BM quasi-bilayer ZOPV films. This is not a true bilayer structure but closer to the bulk-heterojunction (BHJ) structure required for efficient operation of an OPV device.^[26,27]

Figure 2 shows the results of an optical study of the films. In Figure 2a, the absorption of regular (non-rubbed) P3HT film was studied to monitor changes in the film as a result of subsequent processing steps to fabricate a complete PV device. Dichloromethane (DCM) is a poor solvent for P3HT. When spin-coating pure DCM onto the P3HT film, no significant difference is observed (see Figure 2a, series 1 and 2), indicating the resistance of P3HT film against DCM solvent. When PC₆₀BM was coated onto P3HT from its DCM solution

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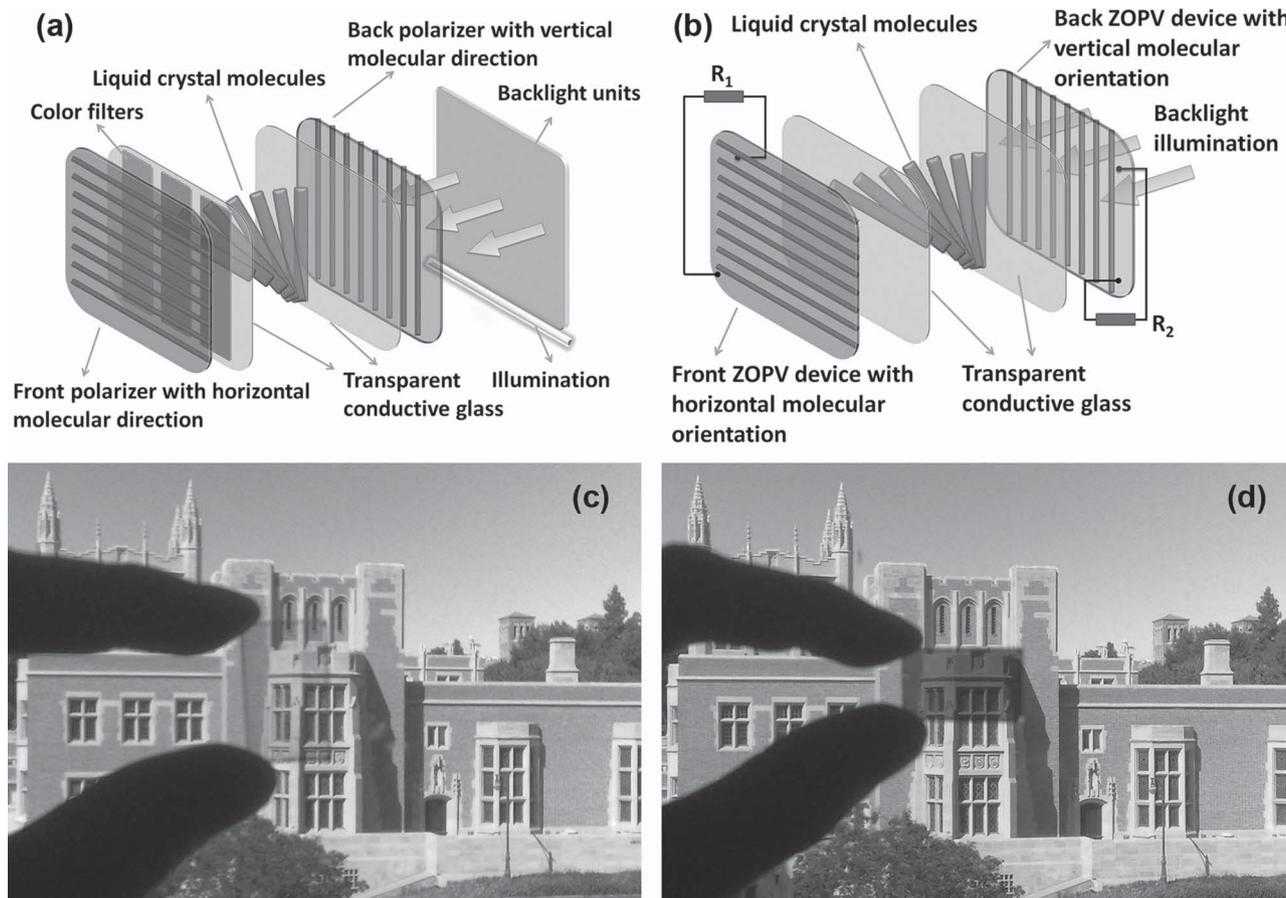


Figure 1. Schematic constructions of LCD (a) and the potential LCD system integrated with ZOPV devices (b). Z-P3HT film with the rubbing direction parallel (c) or perpendicular (d) to the molecular orientation in the analyzer.

followed by annealing, PC₆₀BM molecules would diffuse into the P3HT films to form the interpenetrating quasi-bilayer P3HT-PC₆₀BM PV film.^[23–27] From Figure 2a series 3 and 4, it is clear that the diffusion of PC₆₀BM molecules does not affect the absorption of P3HT films. The absorption shoulder around 600 nm, which indicates the packing of P3HT chains,^[15] is not changed after thermal annealing. This suggests that the PC₆₀BM molecules only move in the disordered regions of P3HT film, but do not affect the ordered packing of P3HT chains.^[26–28] Figure 2b shows the polarized absorption of the z-P3HT film to *s*- and *p*- polarized incident light. Under *s*-polarized light, the optical density (OD) at 610 nm was only 0.0494. In contrast, when the absorption was taken under *p*-polarized light, a much stronger OD of 0.705 was observed at 610 nm. The ratio between absorption for *p*- and *s*-polarized light (defined as dichroic ratio, DR) can reach up to 14.3. This suggests highly efficient molecular alignment induced by the surface rubbing process. Figure 2c shows the polarized absorption of the quasi-bilayer P3HT-PC₆₀BM ZOPV film without annealing treatment. The DR peaks at 604 nm with a high value of around 12.4. This indicates that P3HT molecular chains still keep the high orientation after the coating of PC₆₀BM. More importantly, PC₆₀BM has weak absorption in the visible range and this does not affect the final anisotropic absorption of the ZOPV film a lot. After

annealing at 150 °C for 5 min, PC₆₀BM molecules diffused into the aligned P3HT matrix. From the polarized absorption spectra in Figure 2d, we note that the final ZOPV film still have the high DR value of approximately 11.3 at 604 nm, indicating that the aligned P3HT molecular chains maintain high orientation after thermal annealing. From these spectra, it can be concluded that PC₆₀BM molecules do not affect the P3HT molecular orientation and packing of P3HT when they are diffusing into the aligned P3HT matrix to form the quasi-bilayer ZOPV films.

Rubbing of P3HT films might create scratches on the surface. This will lead to the short circuits in the film, resulting in failure of the regular bilayer structure.^[5,6,24–26] Although by introducing a quasi-bilayer structure (wherein we spin coat PC₆₀BM on top of P3HT) we fill most of the cracks, still a thick buffer layer between the active layer and the electrode is required to prevent shorts in the device. An ideal buffer layer would be thick enough to prevent the evaporated metal electrode from penetrating the active layer and also be electrically conductive to facilitate OPV operation. One such candidate is the conductive polymer poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS). However, it is a *p*-type polymer, which necessitates that an inverted architecture is utilized for device fabrication.^[29] Moreover, PC₆₀BM spin-coated after P3HT would form a Schottky contact with PEDOT:PSS.

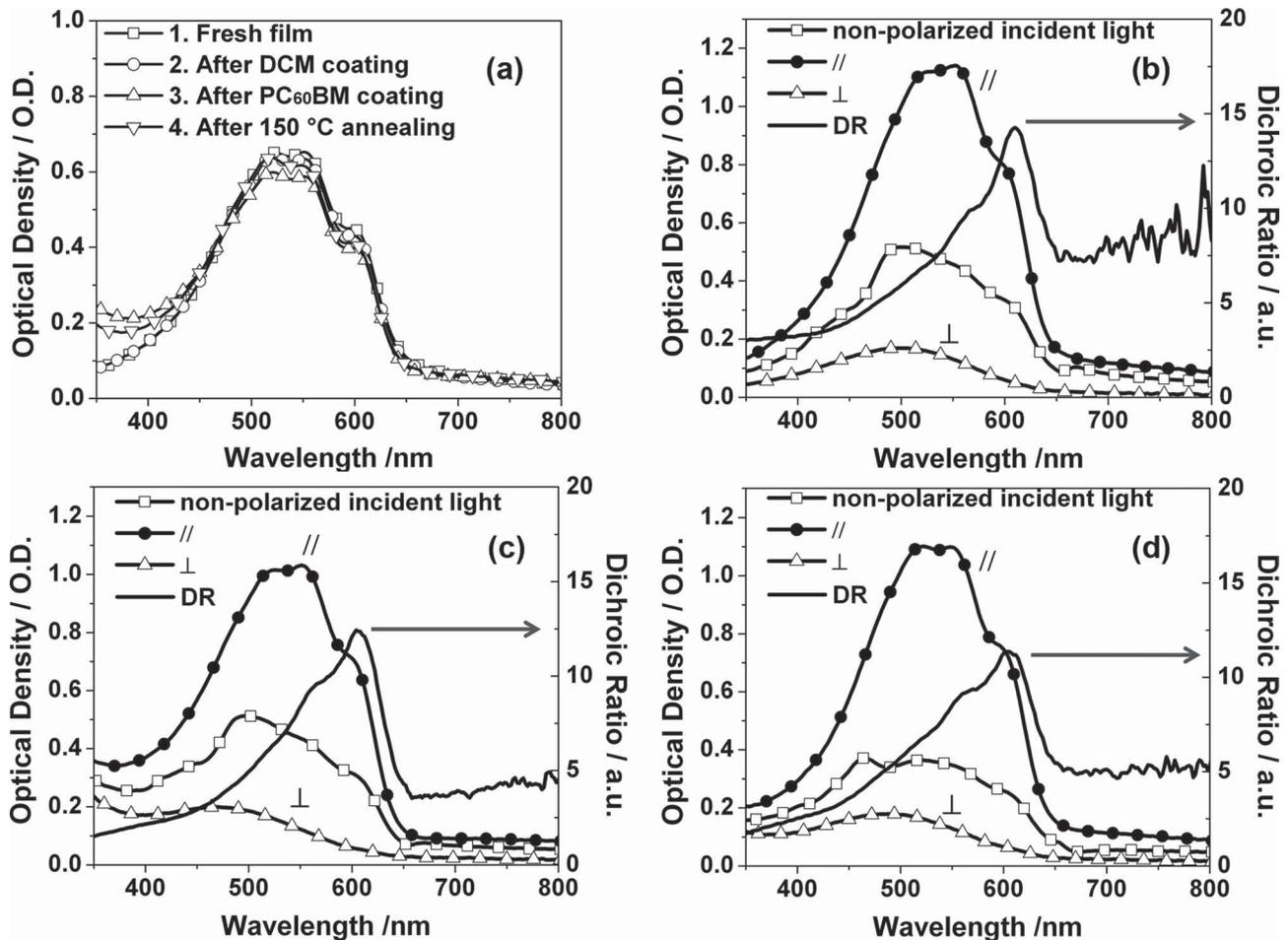


Figure 2. Optical study of the films. a) Absorption of P3HT films upon different treatments: 1) fresh film, 2) after spin-drop-coating DCM solvent, 3) quasi-bilayer P3HT-PC₆₀BM PV film, and 4) absorption of the film in (3) after 150 °C thermal annealing for 5 min. b) Polarized absorption of the z-P3HT films obtained with either s- (\perp) or p- (\parallel) polarized incident light. c) Polarized absorption of the quasi-bilayer P3HT-PC₆₀BM film without annealing. d) Polarized absorption of the quasi-bilayer P3HT-PC₆₀BM film in (c) after annealing at 150 °C for 5 min.

To fabricate a working PV device, we introduce another novel structure where we introduce optimum annealing of PC₆₀BM so that PC₆₀BM penetrates to the bottom of the P3HT layer to contact the ITO electrode forming optimum electron extraction morphology through the bulk active layer.^[26,27,30] This will also expose some of the P3HT on the top layer to form ohmic contact (for holes) with the PEDOT:PSS anode. **Figure 3a** shows the inverted quasi-bilayer device structure based on the non-rubbed P3HT film. In this device, the P3HT-PC₆₀BM quasi-bilayer PV film was prepared on the cathode modification layer (TiO₂:CsCO₃).^[31] The anode modification layer, PEDOT:PSS, was coated onto the P3HT-PC₆₀BM quasi-bilayer film. **Figure 3b** gives the current density versus voltage (J - V) characterization of the inverted quasi-bilayer devices based on the non-rubbed P3HT film (control device). The measurement was performed under regular AM 1.5G 1 sun irradiation. A power conversion efficiency (η_e) of 2.62% was obtained with a short-circuit current density (J_{sc}) of 6.45 mA cm⁻², an open-circuit voltage (V_{oc}) of 0.61 V and fill factor (FF) of 66.6%. The high device efficiency is evidence that the PC₆₀BM diffuses through the P3HT matrix to the bottom TiO₂:Cs₂CO₃

to form a continuous transport route for efficient electron collection.^[30]

Based on the inverted quasi-bilayer device architecture, we fabricated ZOPV devices using the aligned P3HT film. Both reflective electrodes (Al, 100 nm, **Figure 4a**) and semitransparent electrodes (Au, 10 nm, **Figure 4b**) have been used for the devices. **Table 1** summarizes the device performance. The devices were tested under both unpolarized and polarized AM1.5G illumination. When tested under unpolarized AM1.5G 1 sun condition, the ZOPV devices with reflective Al electrode reached a power conversion efficiency of $\eta_e = 1.38\%$, with $J_{sc} = 3.69$ mA cm⁻², $V_{oc} = 0.60$ V, and $FF = 62.2\%$. Compared with control device using regular P3HT film (**Table 1**), the low J_{sc} in the ZOPV device is because only the p-portion of the incident light can be efficiently absorbed and transferred into electricity. The ZOPV device with semitransparent Au electrode gave a $\eta_e = 0.69\%$, with $J_{sc} = 2.74$ mA cm⁻², $V_{oc} = 0.60$ V, and $FF = 42.2\%$. These results confirm that the PEDOT:PSS layer provides an effective sealing layer and prevents the short circuit. The low FF for the semitransparent ZOPV device is primarily because of the high series resistance of the 10 nm Au electrodes on the PEDOT:PSS film.

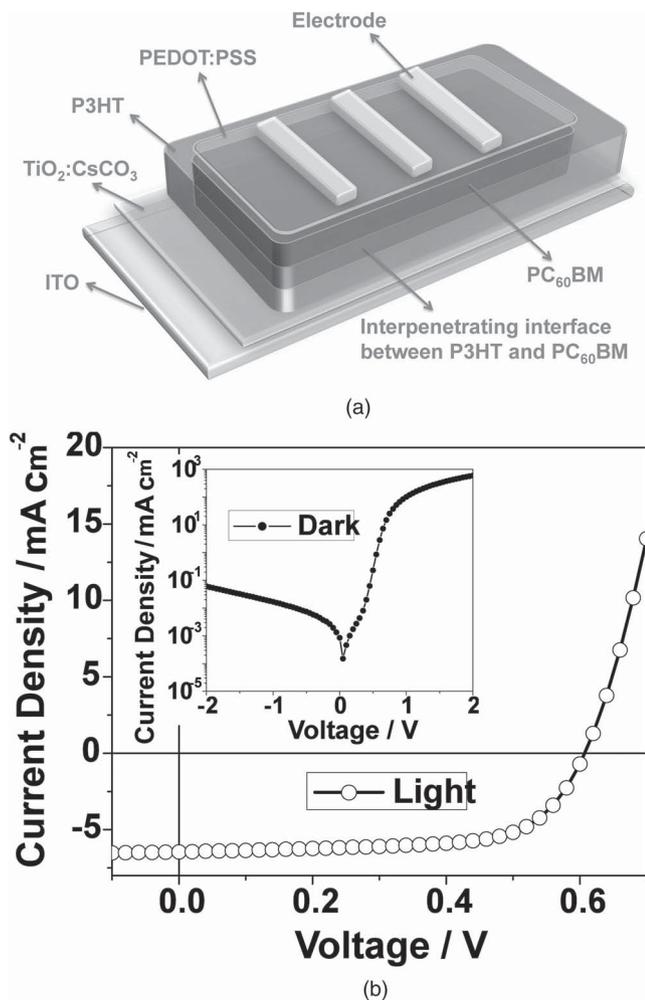


Figure 3. Characterization of the inverted quasi-bilayer OPV device: a) device architecture and b) J - V characterization (inset: dark current).

For the polarizing PV performance characterization of ZOPV devices, a polarizer was placed between devices and light source to provide polarized incident light. The polarized light intensity was approximately 33 mW cm^{-2} . Figure 4c show the dichroic photovoltaic effect of ZOPV device with reflective electrode. The reflective ZOPV device generated a J_{sc} of 1.88 mA cm^{-2} under p -mode polarized light, with $\eta_e = 2.00\%$, $V_{oc} = 0.57 \text{ V}$, and $FF = 62.2\%$. In contrast, when tested under s -mode polarized light, the J_{sc} was only 0.68 mA cm^{-2} , with $\eta_e = 0.64\%$, $V_{oc} = 0.528 \text{ V}$, and $FF = 58.53\%$. The ratio of $J_{sc}(p)$ to $J_{sc}(s)$ is 2.76. Polarized incident photon-to-current conversion efficiency (IPCE) was also utilized to identify the polarized J_{sc} , which gave $J_{sc}(p) = 1.92 \text{ mA cm}^{-2}$, $J_{sc}(s) = 0.71 \text{ mA cm}^{-2}$ and a J_{sc} ratio of 2.70. Figure 4d shows the polarized J - V characterization of the semi-transparent ZOPV device. A higher J_{sc} ratio of up to 3.54 was achieved with $J_{sc}(p) = 1.45 \text{ mA cm}^{-2}$, and $J_{sc}(s) = 0.41 \text{ mA cm}^{-2}$. Compared to the ratio in the reflective electrode, this is a little higher. This is because the incident light in the reflective ZOPV is reflected by the electrode, causing the incident light to propagate through the active layer twice and be absorbed twice. For the transparent ZOPV, the incident light is not reflected and

only absorbed once. Further investigation is under way to provide a much higher J_{sc} ratio. Large scale quasi-bilayer device can be fabricated using spray-coating deposition.^[32] Moreover, higher ZOPV performance can be expected if a better transparent electrode is used and this is also important for its future application in LCD panels.

To conclude, we have developed polarizing organic photovoltaic technology, a novel concept for energy harvesting and recycling technology. We have successfully achieved highly oriented P3HT films with high DR values of up to 14.3 through a surface rubbing method. To realize the ZOPV device, we also introduce novel inverted quasi-bilayer device architecture. The structure gives the flexibility to process P3HT and PC₆₀BM separately. This provides the feasibility to fabricate polymer donor films with additional functions, such as the anisotropic optical property shown here. Good device performance has proved that the PC₆₀BM molecules will penetrate through the highly aligned P3HT matrix and reach the bottom electrode while keeping the orientation of P3HT chains. Dichroic photovoltaic effect characterization has given a high J_{sc} ratio of 3.54. We also calculated the ideal energy recycling efficiency for the ZOPV devices (see the Supporting Information); at a value greater than 10%, this shows great promise for revolutionizing the architecture of the traditional LCD display. This method of both recycling otherwise wasted energy and harvesting the ambient light energy represents significant progress towards the development of another promising green technology.

Experimental Section

Materials: Poly(3-hexylthiophene) (P3HT) was purchased from Rieke Metals, Inc. [6,6]-phenyl C₆₁-butyric acid methyl ester (PC₆₀BM) was purchased from Nano-C. Poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS, CLEVIOS P VP Al 4083) was purchased from H. C. Starck. All materials were used as received.

Polarizing P3HT (z -P3HT) Film Fabrication: The solution of P3HT in chlorobenzene (2 wt%) was spin-drop-cast onto substrate at a speed of 2500 rpm for 5 s. The film was then annealed at 150 °C for 10 min. The P3HT film was rubbed gently with a velvet cloth 80 times at 150 °C. See the Supporting Information for a video of the rubbing process for P3HT in detail; Figure S2 and Table ST1 (Supporting Information) show the increase in DR as a function of the rubbing time. We believe that the rubbing process induces orientation of the P3HT molecules. This orientation probably arises from two effects: i) the shear of the surface during rubbing and ii) the compression of the surface by the loading force. The resultant process propagates the orientation obtained at the surface to the bulk.^[33,34] Furthermore, the detailed physical mechanism behind orientation of the bulk film is under investigation.

Inverted Quasi-Bilayer Device Fabrication: The devices were fabricated on indium tin oxide (ITO)-coated glass substrates with a sheet resistance of $15 \Omega \text{ square}^{-1}$. TiO₂:CsCO₃ solution, prepared by blending 0.4 wt% and 0.4 wt% solutions of TiO₂ and Cs₂CO₃ in a 1:1 volume ratio, was spin-cast at 2500 rpm for 30 s, and the thermal annealing was performed at 150 °C for 15 min. The P3HT films (regular or polarizing films) were fabricated over the TiO₂:CsCO₃ layer. A 0.5 wt% solution of PC₆₀BM in DCM was spin-drop-coated onto the P3HT film while the substrate was spinning at 4500 rpm. The films were then annealed at 150 °C for 1 to 5 min to form a quasi-bilayer structure. Then, PEDOT:PSS with surfactants was spin-coated at 4000 rpm for 60 s and annealed at 80 °C for 2 min. The device fabrication was completed by thermal evaporation of Al (100 nm) as the reflective electrode or Au (10 nm) as the semitransparent electrode under vacuum at a base pressure of 2×10^{-6} Torr.

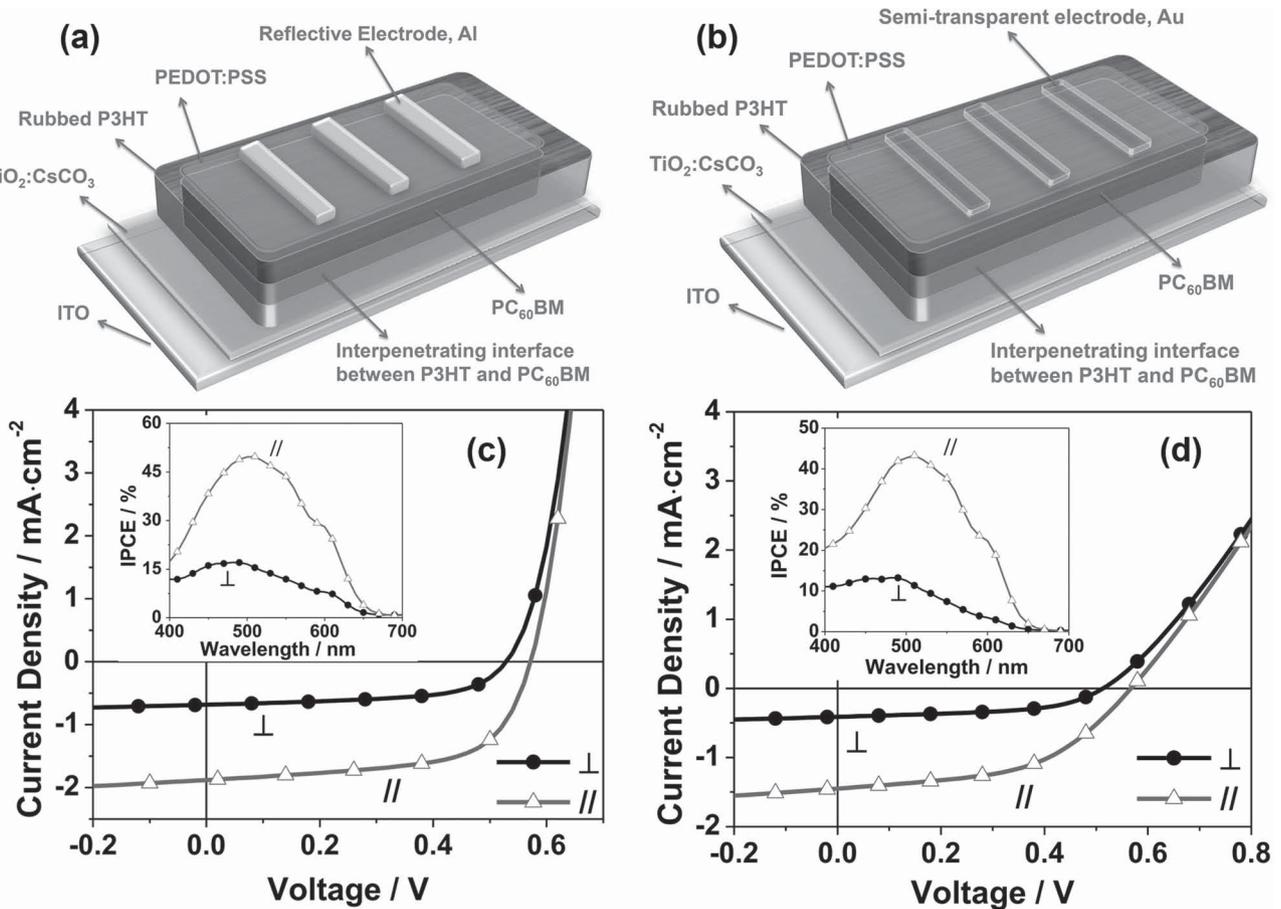


Figure 4. Inverted quasi-bilayer ZOPV device architecture using reflective Al electrode (a) or semitransparent Au electrode (b). Dichroic J - V characterization of the inverted quasi-bilayer ZOPV devices with reflective (c) and semitransparent (d) electrodes; inset: polarizing IPCE spectra.

Electrical, Optical, and Microscopic Characterization: Absorption spectra were taken using a Hitachi ultraviolet-visible spectrophotometer (U-4100). For the polarized absorption characterization, a prism polarizer accessory was placed between the light source and the

samples to provide the polarized incident light. J - V characteristics of photovoltaic cells were taken using a Keithley 2400 source unit under a simulated AM1.5G spectrum with an Oriel 91191 solar simulator. For the polarizing PV measurement, a polarizer was placed between the light

Table 1. Device performance summary.

Anode	Device type	Test condition	J_{sc} [mA cm ⁻²]	V_{oc} [V]	FF [%]	η_e [%]
Reflective electrode, PEDOT:PSS/Al (100 nm)	Control	AM1.5G, 1 sun, unpolarized light	6.45	0.61	66.6	2.62
	ZOPV	AM1.5G, 1 sun, unpolarized light	3.69	0.60	62.2	1.38
	ZOPV	s-mode polarized light, 0.33 sun, ⊥	0.68	0.53	58.5	0.63
	ZOPV	p-mode polarized light, 0.33 sun, //	1.88	0.57	62.2	2.00
Semitransparent electrode, PEDOT:PSS/Au (10 nm)	ZOPV	AM1.5G, 1 sun, unpolarized light	2.74	0.60	42.2	0.69
	ZOPV	s-mode polarized light, 0.33 sun, ⊥	0.41	0.51	53.1	0.33
	ZOPV	p-mode polarized light, 0.33 sun, //	1.45	0.57	50.3	1.25

source and device to provide the polarized incident light. The intensity of the polarized light was approximately 33 mW cm^{-2} , as calculated using a Si photodiode.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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