

An All-Organic Self-Powered Photodetector with Ultraflexible Dual-Polarity Output for Biosignal Detection

ntingting Yan, Ziqing Li, Fa Cao, Jiaxin Chen, Limin Wu, and Xiaosheng Fang*

ndowing photodetectors with mechanically flexibility and actual funcntionality are current research issues in developing optoelectronic devices. nt book week to be a constant of the sector of nt to the realization of ultraflexible electronics. Thus, an ultraflexible all-organic nbotodetector (all-OPD) is designed by innovatively introducing symmetrical not contract to the substitute the widely applied indium-
toped tin oxide (ITO)/Ag electrodes. Specifically, this all-OPD exhibits a high nt bottocurrent remains about 80% of the original performance after being bent n 20 000 circles, and can output steady biosignals for photo-plethysmography nt (PPG) application. More importantly, this all-OPD outputs dual-polarity not occurrent as it is flipped or folded. Benefitting from the ordered phase ndistribution and designed Schottky barrier heights, the photogenerated holes n will be transferred and collected by nearer electrode, while electrons will be nt speed in the thick bulk heterojunction (BHJ) as a result of the long channel. nt bis work offers a new avenue toward developing a multifunctional and ultrand transported to the straightforward all-solution method, and it is expected to be more compatible in complex application scenarios.

1. Introduction

Abundant organic semiconductors have been developed recently, thus, offer us more options in designing organic electronic devices. Internet options in designing organic electronic devices in the energy levels in designing organic electronic electronic

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An organic photodetector (OPD), with organic semiconductors as photosensitive layer, has proved to rival that of a low-noise silicon photodetector, almost in all metrics^[10] and has promising applications in health monitoring and image sensing.[11-13] In the meantime, flexible organic semiconductors are advantageous substitutes for inorganic semiconductors in wearable electronics because conformal thin-film devices can be manufactured with them, therefore, can better adhere and fit human skin.^[14,15] Although organic semiconductors can make up for the poor flexibility of inorganic semiconductors to some extent, the metal-based or metal oxide electrodes in OPD remain a block to the realization of ultraflexible devices.^[13] Simultaneously, metal-based or metal-oxide-based electrodes require vacuum thermal evaporation or electron beam deposition techniques that will complex the preparation process and increase production costs.^[15] Introducing intrinsic flexible organic electrodes may

 help solve the above mentioned problems. The polymer conductor help solve the above mentioned problems. The polymer conductor help solve the above mentioned problems. The polymer conductor of polymer conductor problems is polymer by the problem of the most promising electrode in skin-like wearable electronics.

Dual-polarity photocurrent response endows optoelectronic devices with multifunctional characteristics that have proved to be applied in switchable light imaging, optical communication, and spectral bands' distinction.^[16-19] While traditional p-n junction photodiodes should obey the physical principles of the unidirectional current migration, hence dual-polarity photocurrent response in the previous researches was realized by taking advantage of the concomitance among photovoltaic, photo-electrochemical, and photo-thermoelectric effects,^[16,17,20,21] which has a high demand in selecting materials and fabricating devices. While in organic semiconductor systems, donors and acceptors blended to form interpenetrating bulk-heterojunction (BHJ) networks,^[22] by reasonably designing the work functions of electrodes, migrations of electrons/holes can be effectively regulated thus can easily switch the polarity of electronic signals.^[23,24] Distinctive characteristics of BHJ in OPD inspired us a straightforward and low-cost way in developing dual-polarity photocurrent response optoelectronic devices.

In this work, we introduced transparent organic electrodes PH1000/PH1000 to fabricate an all-organic photoeltector (all-OPD) with a vertically aligned structure PH1000/ poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate), Al 4083







Figure 1. Device structure of the all-OPD and properties of PH1000. a) Device structure of the all-OPD, top PH1000 was connected to ground.
 b) Chemical structures of each layer. c) Surface morphology of PH1000 film. d) Transmittance of PH1000 and energy levels of each layer. e) Surface.

(PEDOT:PSS)/poly(3-hexylthiphene) (P3HT):phenyl-C71-butyricacid-methyl ester (PC71BM)/polyethylenimine ethoxylated (PEIE)/ PH1000 that obtained a high self-powered responsivity (R) of more than 100 mA W⁻¹ among 500–600 nm. Interestingly, by virtue of this device configuration, the polarity of the outputting photocurrent was changed as we flipped over or folded our all-OPD. The polarity of net current depended on the illumination position, where more photogenerated charge carriers would flow to and be collected by the nearer electrode. Two possible applications that may result from this effect were position monitoring and motion detections, which can be realized by detecting the electric signals as rotating or folding the all-OPD. Meanwhile, excellent flexibility of this device offers more application possibilities; biosignals were detected by sticking this soft all-OPD on finger pulp combined with a red light-emitting diode (LED). Given the simplicity of the device structure and fabricating process, this all-OPD will have great application potential in realizing multifunctional, portable, and wearable electronic devices.

2. Results and Discussion

Here, 500 nm bulk heterojunction was formed with the blends of organic semiconductor donor P3HT and acceptor PC71BM (Figure S1, Supporting Information) which acted as the photosensitive layer. S1, Supporting Information) which acted as the photosensitive layer. S1, Supporting Information) which acted as the photosensitive layer. S1, Support is sensitive layer with a subset of sensitive layer state with the sensitive layer and layer are listed in Figure 1b. PEDOT:PSS and PEIE were and electrodes.^[26–28]

In order to obtain an all-OPD with intrinsic flexibility, organic electrodes PH1000/PH1000 were introduced to substitute conventional rigid indium-doped tin oxide (ITO)/Ag electrodes in P3HT:PC71BM-based system (see Experimental Section for details). Smooth PH1000 films with a roughness of 2.61 nm were formed by a solution method on an ultrathin poly(ethylene terephthalate) (PET) substrate as a bottom electrode and on PEIE layer as a top electrode (Figure 1c). Additionally, PH1000 film was transparent enough, where nearly over 90% incident light around 250-800 nm can be transmitted from either top PH1000 electrode or bottom PH1000 electrode to photosensitive layers (Figure 1d); thus, the light loss arose from the reflection of the electrode would be minimized. The work function of PH1000 was determined to be -5.0 eV by Kelvin probe in air (Figure 1e; Figure S2, Supporting Information). Energy band diagrams of different layers were listed







Figure 2. Optoelectronic properties of rigid OPD based on ITO/Ag electrodes and the flexible all-OPD based on PH1000/PH1000 electrodes.
a,b) Device structure of ultraflexible of properties of rigid OPD based on ITO/Ag electrodes and the flexible all-OPD based on PH1000/PH1000 electrodes.
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The all-OPD was successfully prepared with the device configu-PET/PH1000/PEDOT:PSS/P3HT:PC71BM/PEIE/PH1000 ration (Figure 2a). The evolution of sheet resistances of PET/PH1000 was tested as they were bending 0°, 90°, and 180°, as shown in Figure S3 (Supporting Information), and it was found that PET/ PH1000 almost depicted the same resistance of its original value upon bending strain, which indicated that PET/PH1000 has superiority in prospect of practical application in flexible device. Optoelectronic properties of the ultraflexible all-OPD under different wavelengths were further investigated at 0 V. Additionally, optoelectronic properties of rigid OPD with conventional rigid electrodes ITO/Ag were fabricated and characterized for comparison (Figure 2b). R is a key parameter to evaluate optoelectronic conversion capability of a photodetector, which is defined by equation R = $I_{\rm ph}/PS$, where $I_{\rm ph}$ represented the photocurrent of photodetector, P is the power density, and *S* is the effective area of the photodetector. Detectivity (D) indicates the capacity of the photodetector to detect weak signals and can be calculated by *R*, $D = R/(2qJ_d)^{1/2}$, where *q* is the electronic charge and *I*_d is the dark-current density.^[29]

Photocurrent and dark current of the two devices were acquired by the current- and dark current of the two devices were acquired by the current-time (*I-t*) response test under eilumination of different wavelengths on–off at 0 V. It is found in Figure 2c,d that both the all-OPD and the rigid OPD could achieve a high *R* of about 20–150 mA W⁻¹. Especially, the allOPD achieved its highest R between 500 and 600 nm, which would be beneficial to be applied in photo-plethysmography (PPG) technology for the detection of biosignals. In addition, it was found that the all-OPD depicted a lower R than that in rigid-OPD, and there may be two reasons. It may correspond to the limited driving force from the symmetrical electrodes PH1000/ PH1000 for promoting carrier's transportation. Also, the quality of organic layers on ultraflexible substrate would decline to some extent; thus, the charge recombination may happen before they separated. Compared to the nearly transparent top PH1000/PEIE^[26] (Figure 1d), there was an intense absorption of Ag over the wavelength of 400 nm,^[30] which caused a large energy loss by the electrode Ag reflection or absorption thus leading to the reduction of photocurrent (Figure S4, Supporting Information) and suppression of R in wavelength between 400 and 600 nm for ITO/Ag-based rigid devices. D of the two devices exhibited the same variation trends as R, while there was also a slight loss of *D* in the all-OPD as a result of its higher dark current. Further, the response speed of P3HT:PC71BMbased OPD at 0 V was conducted, and it was found that steady and regular current signals under the transient light response without applied bias could be outputted, as shown in Figure S5 (Supporting Information). The response time was estimated to be 0.5 ms to rise and 3.2 ms to fall, which could satisfy the requirements of detecting of biosignals in this work.

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light intensity illuminations of 350 nm at 0 V (Figure 2e,f) light intensity illuminations of 350 nm at 0 V (Figure 2e,f) light intensity illuminations of 350 nm at 0 V (Figure 2e,f) light intensity illuminations of 350 nm at 0 V (Figure 2e,f) light is disconsistent intensity intensities of 350 nm at 0 V (Figure 2e,f) light of 350 nm at 0 V (Figure 2e,f) light of 350 nm at 0 V (Figure 2e,f) light of 0 li

 dark current were basically in the same order of magnitude, and the dark current cannot be ignored under weak light.

An ultraflexible self-powered all-OPD with high R and D An ultraflexible self-powered all-OPD with high R and D An ultraflexible self-powered all-OPD with high R and D around 500–600 nm has been teded to be another allound that be another another another another another be another another another another another another another all-OPD with distinctive opticelectronic properties that the photocurrent polarity and value were dependent on the device rotating angles. As shown in Figure 3a, as the all-OPD was



ing figure 3. Dual-polarity photocurrent response and working principles of the all-OPD during flipping and folding. a) Schematic of device flipping. Figure 3. Dual-polarity photocurrent response and working principles of the all-OPD during flipping and folding. a) Schematic of device flipping. Interview of the subscription of the subscription of the subscription of the subscription of the subscription. Schematic of the subscription of the subscription of the subscription of the subscription. Schematic of the subscription of the subscription of the subscription of the subscription. Schematic of the subscription of the subscription of the subscription of the subscription. Schematic of the subscription of the subsc

Notably, considering the ultraflexibility of this all-OPD, we further explored the photocurrent output responses as it was folded to 0°, 90°, and 180°. As demonstrated in Figure 3c, the photocurrent of the all-OPD decreased to a minimum, as it was folded to 90° because almost half of the device was parallel to incident light under this circumstance. Then, as this all-OPD continued to be folded to 180°, the polarity of photocurrent was switched, and the photocurrent value of the fully folded device increased to more than 6 µA. The inside reason was that when the all-OPD was fully folded (180°), the incident light was switched to be irradiated from the bottom PH1000, hence, presented a similar switchable photocurrent polarity effect. Multifunctional properties of ultraflexibility and dual-polarity response were realized based on our fabricated all-OPD. The photocurrent polarities were strongly dependent on the folding angles of the all-OPD, which may have application prospect in the motion detection field.

Unlike conventional dual-polarity p-n junction inorganic photodetector, the switchable photocurrent in the all-OPD was intensely connected with the distinctive BHJ structure of polymer donor P3HT and fullerene acceptor PC71BM. It is found that in the polymer:fullerene system, blend properties may vary spatially perpendicular to substrate via selfassembly.^[23,32] Due to the vertical phase segregation of the polymer and fullerene, PC71BM tended to concentrated at the bottom PH1000/PEDOT:PSS interface and P3HT tended to accumulated at the PEIE/top PH1000 interface.[33] However, since the mass ratio of the PC71BM and P3HT was 1.5:1 in this work, PC71BM remained dominated in the BHJ perpendicular to the substrate, as schematically shown in Figure 3d, and may form Schottky barriers after contacting electrode PH1000. While Schottky barrier height (SBH) in bottom PH1000/BHJ interface was larger than that in BHJ/top PH1000 because PC71BM was more enriched close to the substrate. Further, schematic energy level alignment diagrams of the generated electric field at the bottom PH1000/BHJ and BHJ/top PH1000 interfaces were made to help better understand carrier's migrations, as shown in Figure S6 (Supporting Information). When incident light was illuminated from top PH1000 side (Figure 3e), photogenerated excitons were stimulated at the shallow layer of the BHJ surface. Electrons and holes separated under the driving forces from donor-acceptor phase interfaces and Schottky junction, where electrons flowed to bottom PH1000 electrode and would be trapped in the thick BHJ film (≈500 nm) with the long channel. Simultaneously, sufficient holes could be transported from semiconductors to top PH1000 electrode with transported from semiconductors to top PH1000 electrode with transported from semiconductors to top PH1000 electrode with transpin or therminoring in the toron top PH1000 electrode with transpin or top top top electrode with transport of the transport of transport of the transport of tra

To present a comprehensive understanding of the internal mechanism of the dual-polarity outputted photocurrent of our all-OPD. We further adjusted the work function of the top electrode by doping silver nanowires (AgNWs) to construct OPDs with asymmetric counter electrode. As AgNWs were introduced to replace half of the top PH1000 electrode (PH1000:AgNWs), device outputted a high photocurrent of over 3 µA (Figure 4a). The slight decrease of current compared with that in the all-OPD may be due to the higher roughness of the top PH1000:AgNW electrodes with a lower charge collection efficiency (Figure S7, Supporting Information). Similarly, when AgNWs completely replaced PH1000 as the top electrode, it was found that ideal AgNWs' conductive networks were difficult to be built on the organic surface thus outputting the lowest current (Figure 4a). Meanwhile, the minimal exponent (0.66) in PH1000/AgNWbased device implied more bimolecular recombination, which would result in the loss of photocurrent (Figure S8, Supporting Information). Consequently, the huge photocurrent difference directly influenced the R of PH1000/PH1000:AgNW-based and PH1000/AgNW-based devices (Figure 4b).

To investigate the relationship of photocurrent polarity with the work function of electrodes, additional I-t experiments were conducted by flipping the OPDs with PH1000/ PH1000:AgNWs and PH1000/AgNWs as electrodes. When AgNWs (work function = 4.2 eV)^[34] were introduced, surface potential of top electrode dropped from 5.1 eV (PH1000) to 4.8 eV(PH1000:AgNWs) (Figure S9, Supporting Information), and there was still a photocurrent polarity reverse as the device was flipped to over 90° (Figure 4c), where Schottky barrier in BHJ/top PH1000:AgNWs' interface (Φ_3) was nearly the same as Φ_1 therefore had the same mechanism as in PH1000/ PH1000-based system (Figure 4d). However, as the top electrode was completely substituted by AgNWs, dual-polarity photocurrent response disappeared as it was flipped from 0° to 180° (Figure 4e). In this PH1000/AgNW-based OPD, Schottky barrier was much lower in BHJ/top AgNWs' interface (Φ_4) than that in bottom PH1000/BHJ interface (Φ_2), as incident light electrons would easily get over the Schottky barrier (Φ_4) and collected by external circuit than holes; thus, the polarity of net photocurrent became reversed as well (Figure 4f).

The mechanical bending test was of great significance in evaluating the ultraflexible all-OPD, as demonstrated in Figure 5a. A stable photocurrent could be outputted and remained to be about 5 µA after the all-OPD was bent for 20 000 circles. Small







Figure 4. Optoelectronic properties and mechanisms of OPDs by varying work functions of top electrodes. a) I-t curves of OPDs with PH1000, Figure 4. Optoelectronics and mechanisms of OPDs by varying work functions of top electrodes. a) I-t curves of OPDs with PH1000, PH1000, and New Set on the electrodes of the term of term of

nt current spikes could arise as the incident light was switching on without an external potential, which is corresponding to the photogenerated carriers stimulated at the interface rapidly photocurrent.^[35,36] Variation of photocurrents with bending times is displayed in detail in Figure 5b; photocurrent dropped about 20% after bending 5000 times due to the morphology damage under the constant and cyclic bending stress. The photocurrents of the all-OPD would maintain at 80% of its original value as it was further bent to 20 000 circles. The physical pictures of ultraflexible all-OPD in twisted, crimped, and folded states are also shown in Figure 5b. Therefore, our designed all-OPD had great bending resistance, which was promising to work efficiently in continuous bending operation. In addition, as shown in Figure S10 (Supporting Information), the performance of our fabricated all-OPD after it was put in the air for 10 days was monitored, where the dark current and photo-

current were almost the same with the original results, which could eliminate the possibility of the device performance degradation arising from the environment.

In order to demonstrate the applicability in wearable electronics, transmission mode PPG was assembled by a red LED and our fabricated ultraflexible all-OPD, as shown in Figure 5c,d. The red LED was stuck on fingernail, which would emit red light that easily passed through the finger. flexible all-OPD was stuck on the finger pulp to detect the changes of optical signals caused on the finger pulp to detect the changes of optical all-OPD was stuck on the finger pulp to detect the changes of optical signals caused by changes of bood vessel volume in the microvascular be and the subscription of humans.^[37] Finally, Figure 5 edepicts that 18 periodic biological signals were stably output within 15 s, indicating that this wearable self-powered all-OPD would be an outstanding candidate for all-day health monitoring.

3. Conclusion

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Figure 5. Optoelectronic properties of the all-OPD after a constant bending test and output signals of PPG. a) *I–t* curves of the all-OPD after bending 0 time (black line), 2000 times (blue line), and 20 000 times (red line), the bending radius was about 3 mm. b) Variation of normalized photocurrent after bending different times and physical pictures of all-OPD. *I–t* tests conducted under simulated sunlight (175 mW cm⁻²) on–off switch at 0 V. c) Schematic and d) physical pictures of PPG based on the all-OPD. e) Outputting photocurrent of the PPG within 15 s. The power of the red LED was 0.2 W.

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4. Experimental Section

Materials: All solvents were purchased from Sinopharm China. P3HT and PC71BM were purchased prom Sinopharm China. P3HT and PC71BM were purchased proceed from Sinopharm China. P3HT and PC71BM were purchased from UC and Sinopharm China. Solvent and PC11BM were purchased from Heraeus, Inc., Germany. 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) and PEIE were purchased from Youxuan, Liaoning Co., Ltd.

Device Fabrication: 32 mg of P3HT and 48 mg of PC71BM were dissolved in 1 mL of chloroform and stirred overnight at 55 $^\circ$ C to

form a blended solution. PEIE was dissolved in dimethoxy ethanol to obtain 1 wt% PEIE solution. The volume of PH1000 and PEDOT:PSS aqueous solution was diluted with ethanol to three times the original volume before used. To fabricate a flexible device, PH1000 was sprayed on the ultrathin (30 μ m) PET, which was on a 120 °C hot plate to form a 300 nm film and annealed for 20 min. Thin film of PEDOT:PSS (15 nm) was further produced in the same way as on smooth PH1000 surface. P3HT:PC71BM solution was then spin-coated and baked at 100 °C to form an active layer of about 500 nm. PEIE was sprayed on the active layer to form a thin film of about 10 nm. At last, PH1000 was sprayed on PEIE on a 120 °C hot plate and annealed at 120 °C for 20 min to form a 300 nm film. For the convenience of the optoelectronic measurements, the organic electrodes were extracted by stretchable silver paste. To fabricate a rigid device, glasses with ITO were ultrasonically cleaned with soapy water, deionized water, acetone, and isopropanol for 20 min and plasma treatment for 5 min before use. PEDOT:PSS was spin-coated on the substrate at 3000 rpm and annealed at 120 °C for 20 min. P3HT:PC71BM solution was then spin-coated and baked at 100 °C to obtain the active layer. 10 nm BCP film and 20 nm Ag film were finally prepared via vacuum thermal deposition. The PH1000, PEDOT:PSS, and PEIE layers were fabricated in the air. The photosensitive layer P3HT:PC71BM was fabricated in the glove box with nitrogen. All characterizations of the devices were conducted in the air condition. The dimensions of all devices were $0.2 \text{ cm} \times 0.2 \text{ cm}.$

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Device Characterization: The transmittance of electrodes was calculated by a UV-vis spectrophotometer (Hitache of electrodes was calculated by a UV-vis spectrophotometer of electrodes was calculated by a UV-vis spectrophotometer (Hitache of electrodes was calculated by a UV-vis spectrophotometer of electrodes was calculated by a UV-vis spectrophotometer (Hitache of electrodes was calculated by a cuber spectrophotometer of the transmittance of electrodes was calculated by a cuber verse spectron of the spectrophotometer of the spec

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

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Keywords

<a>ull-organic photodetectors, all-solution processing method, biosignal detection, dual-polarity output

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