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Visible and Near-infrared imaging with non-fullerene based photodetectors

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ABSTRACT

Solution-processed organic photodetectors underpin an emerging technology with inherent implications in biological sensors and imaging displays. Conventional organic photodiodes, the core element of an organic photodetector, rely mainly on fullerene-based acceptors, which in combination with high and middle bandgap donors, limit the spectral photosensitivity to the visible range. Even in the case of low bandgap polymers the oscillator strength and thus the extinction coefficient are usually limited in the NIR due to the nature of molecular orbital hybridisation. Instead, we show that pairing prototypical poly(3-hexylthiophene) (P3HT) with rhodanine-benzothiadiazole-coupled indacenodithiophene (IDTBR), a non-fullerene electron acceptor absorbing beyond 750 nm, as the photoactive material of a simple photodiode results in a highly efficient organic photodetector with a record responsivity of 0.42 A W^{-1} and EQE of 69% in the NIR (755 nm). Non-fullerene based photodiodes were processed on amorphous silicon active matrix backplanes to realize large area flat panel photodetector imagers able to detect objects under visible and NIR light conditions with an exceptional combination of responsivity, dynamic response and image crosstalk.

Organic semiconductors are envisioned to positively impact human life through applications ranging from sensors and displays to lighting and photovoltaics.^[1–3] As opposed to most inorganic counterparts, organic devices can be printed from solution on plastic substrates, allowing for extremely thin, lightweight and conformable products that can be manufactured over large areas with freedom of shape.^[4,5] Notably, the current success of this technology is mainly driven by the development of organic light emitting diodes (OLED), which is now routinely employed in display technology.^[6] In the last decade, however, organic photovoltaics (OPV), leveraging the impressive improvement in device efficiency and stability, gradually moved from a lab curiosity to a niche market.^[7,8] Conversely, organic photodetectors (OPD), a technology based on organic photodiodes and thus closely related to OPV, have not experienced a similar trajectory of growth, mainly due to the lack of understanding of how to simultaneously control critical device parameters such as diode rectification, external quantum efficiency (EQE) and temporal responsivity.^[9] For instance, very few exemplary photodetectors successfully match the requirement to convert low levels of light into a detectable electrical signal, without the need of applying a significant external negative voltage. The latter limits the range of applicability but is a generally accepted measure to enhance charge collection and response time. Understanding how photoactive materials and device layouts can be engineered to alleviate the need for high voltages while maintaining a large on/off ratio is a widely desired goal and is expected to further drive this technology.

A particularly appealing characteristic of OPD devices, which derives from the ability of processing on plastics as well as the ease with which the organic semiconductor can be chemically functionalized, is the integration into complex systems of color vision, with recent examples emerging in both biomedical and soft robotic systems.^[10,11] Artificial retinas represent one exciting example.^[12,13] Furthermore, the chemical design of organic

chromophores with a relatively narrow and specific absorption spectrum allows organic photodetectors to be explored for selective narrowband photodetection, with special interest in the near infrared (NIR) region, where cost effective material choices for biomedical applications and process monitoring remain scarce.^[14] Typically, inorganic materials, such as InGaAs or epitaxial grown quantum dots, are either cost intensive, have reduced compatibility with large area devices or require filters to meet the same level of spectral sensitivity.^[9,15]

Similar to the working principle of OPV, controlling the structural disorder and excitonic nature of organic materials is the key driver for OPD performance.^[16] Maintaining efficient charge carrier transport over large distances (hundreds of nanometers) is a general prerequisite for elevated photosensitivity of printed and spray coated OPDs.^[17] In this context, the recent fast pace of OPV development upon the advent of highly efficient non-fullerene electron acceptor (NFA) materials with superior light harvesting, stability and charge recombination behavior over fullerene-based devices is expected to be of particular relevance.^[18–20] Recently, we observed unusually low recombination rates, enabling close to unchanged device performance for NFA based solar cells with up to 450 nm active layer thickness.^[21] This observation anticipates a similarly advantageous impact on the performance of OPDs, where thick active layers usually contribute to reduced dark currents.^[22,23] Thus, bulk heterojunction (BHJ) organic photodetectors consisting of a donor polymer and an NFA represent a chance to move the detection into the NIR spectral region without sacrificing EQE. Rhodanine-benzothiadiazole-coupled indacenodithiophene (O-IDTBR) is a small molecule NFA that could fulfil the aforementioned prerequisites, with its high extinction coefficient, long wavelength absorption, and good electron transport.^[24,25] Photovoltaic devices based on O-IDTBR and the well-known polymer P3HT deliver consistently power conversion efficiencies (PCE) of 6%.^[24] Significantly, the devices show nearly unchanged photostability over a time period of at least 2000 h.^[26] The fact that P3HT:O-IDTBR BHJ devices feature

ultra-low charge recombination in the presence of a thick active layer of $\sim 0.5\mu\text{m}$ makes this blend ideally suited for large area printing and coating applications.^[27] Within the framework of the present work, we further noted that under low light levels ($1\text{--}10\text{ mW cm}^{-2}$) close to “shunt-free” solar cell devices would maintain their high fill factor (FF) values, consistent with low charge recombination, and anticipating suitable performance for low light detection (**Figure S1**, in the supporting information). Motivated by the combined benefits of P3HT:O-IDTBR, we transferred the optoelectronic properties of P3HT:O-IDTBR to realize efficient organic photodetectors. P3HT:O-IDTBR based OPDs feature simultaneously high external quantum efficiency ($\sim 70\%$), low dark current ($J_d=30\text{ nA cm}^{-2}$) and, to the best of our knowledge, the highest responsivity of 0.42 A W^{-1} at 755 nm for solution processed organic photodetector reported so far. By integrating solution processed active layers on top of an amorphous silicon active matrix backplane (a-Si AM), we realized large area flat panel imagers able to detect objects under visible and NIR light conditions with an unprecedented combination of responsivity, dynamic response and image crosstalk.

Results and Discussion

Figure 1a shows the chemical structure of P3HT and O-IDTBR and the absorption profile of the blend. The absorption of O-IDTBR extends between the near UV and NIR, thus, complementing the characteristic absorption of P3HT. To study the performance of P3HT:O-IDTBR as the photoactive layer of an organic flat panel photodetector (imager), we processed the blend on an a-Si thin film transistors array as active matrix backplane with an Indium Tin Oxide (ITO) bottom electrode per pixel on glass substrate.^[15] For this purpose, a ZnO solution (electron transporting layer) was first coated on the a-Si/ITO wafer. The BHJ blend was then either spray coated or doctor bladed on ZnO, with the results from both coating methods shown below. Notably, these steps were all done in air. The device was finished by processing

layers of thermally evaporated MoOx (hole selective layer) and Aluminum (Al). **Figure 1b** and **1c** show a photograph of the flat-panel imager and the device configuration, respectively. **Figure 1d** depicts qualitatively the resulting sensitivity of the photodetector. The images of shaking hands and an individual hand are shadow casts of transilluminated photographic slides using a green LED source (peaked at 532 nm). **Figure S2** collects different images obtained from the imager. The vertical and horizontal lines derive from dead pixels on the silicon wafer. The quality perception of an image (signal to noise ratio, brightness, sharpness, etc.) is crucially dependent on the optoelectronic characteristics of the photodiode that forms the photodetector, particularly the dark/photocurrent (on/off ratio and diode rectification), the quantum efficiency for converting photons into electrons (spectral sensitivity) and frequency response. Thus, we first analyze one critical device parameter, the dark current density (J_d), of inverted P3HT:O-IDTBR devices processed via doctor blading and spray coating.

Figure 1 a) Chemical structure of the materials used and their absorption profiles. b) Photograph (active area 2.5x2.5 cm) and c) schematic device stack of the organic imager. d) Visible shadow cast under green illumination (532 nm) of a slide showing a handshake and a hand. The stripes in the images are the result of interconnection failures of flexible printed circuits (FPCs) on the contacting pads of the glass and are not due to defects of the active area.

Figure 2a shows the J-V characteristics from -5V to +5V of representative photodiodes. Interestingly, we measured low J_d for doctor bladed and spray coated devices of 30.4 nA cm^{-2} and 21 nA cm^{-2} , respectively. Moreover, we calculate a rectification ratio, defined as the ratio of J_d at $\pm 5\text{V}$ of $\sim 10^6$, supporting that these devices form excellent diodes. Importantly, illuminating the device with monochromatic green light (532 nm and a power density of $780 \mu\text{W cm}^{-2}$) the current density increases by almost four orders of magnitude, leveraging the good charge extraction capability of the P3HT:O-IDTBR based devices, in agreement with our previous observations in analogous photovoltaic cells. This result is impressive for several reasons. It shows that this semiconducting polymer blend can be processed using different scalable coating techniques with little implications on the optoelectronic properties. Furthermore, the blend can be processed with high thickness (700 nm), while still preserving low **series** resistance and high on/off ratios. It is thus conceivable that a blend of this nature could be easily adapted for efficient large area manufacturing. Table 1 depicts relevant device characteristics of this NFA-based photodiode.

An additional, critical parameter establishing the suitability of a photoactive semiconductor for imaging applications is the photon to electron conversion efficiency. We therefore performed external quantum efficiency (EQE) measurements. (**Figure S3**). **Figure 2b** shows the maximum EQE value (at 755 nm) of P3HT:O-IDTBR as a function of negative voltage for both blade and spray coated devices. We found very high EQE values of $\sim 70\%$ in the NIR and photoactive response beyond 800 nm, as detailed in **Table 1**. To put our results

into perspective, we highlight that P3HT:PCBM, a prototypical blend for organic photodiodes, achieves ~75% EQE at 550 nm in the best case, but drops sharply after 630 nm.^[28] As such, the observation of high EQE combined with the very low dark current discussed earlier render the P3HT:O-IDTBR blend an excellent candidate for low-cost photodetector applications across the visible and NIR.

Figure 2 a) Current-voltage measurements of doctor-bladed and spray coated P3HT:O-IDTBR devices in the dark and under green light conditions. b) Responsivity (solid symbols) and External Quantum Efficiency (open symbols) data at 755 nm as a function of voltage for the same devices as in a). c) Responsivity of doctor-bladed P3HT:O-IDTBR devices as a function of bias. d) Measured cut-off frequency at -5 V reverse bias under green light illumination. The dashed horizontal lines represent a common metric to show the frequency at which the signal amplitude (photocurrent) drops to $1/\sqrt{2}$ (-3 dB) and $1/2$ (-6 dB) of the initial dc value.

Finally, from the EQE data we calculate the responsivity (R) of our devices using the following equation (1):

$$R(\lambda) = EQE \frac{\lambda q}{hc} \quad (1)$$

where λ is the wavelength, q the elementary charge, h the Plank's constant and c the speed of light. A value for R of 0.42 A W^{-1} at -5V and 755 nm for doctor bladed device (**Figure 2c**) and 0.38 A W^{-1} for spray coated diodes (**Figure S4**) was achieved, which, to the best of our knowledge, is the highest responsivity value reported so far for an organic photodetector. To better evaluate the significance of our results, we have included in **Table 1** and **Figures S5** and **S6** typical device parameters for champion photodetectors using different technologies. Importantly, within this comparison P3HT:O-IDTBR emerges as the most efficient organic photodetector for wavelengths beyond 700 nm .

Table 1. Diode parameters of P3HT:O-IDTBR photodetectors were measured at -5V reverse bias. EQE and R were calculated for 755 nm light excitation.

	J_D [A cm ⁻²]	EQE [%]	R [A W ⁻¹]	f _{-3dB} [Hz]	f _{-6dB} [Hz]	Ref.
P3HT:O-IDTBR Spray-coated	2.1×10^{-8}	63.4 @755 nm	0.38 @755 nm	12.4×10^3	45.2×10^3	This work
P3HT:O-IDTBR Blade-coated	3.4×10^{-8}	68.5 @755 nm	0.42 @755 nm	52.6×10^3	141.9×10^3	This work
P3HT:PCBM	7×10^{-8}	76 @550 nm	0.39 @550 nm			[28]

P3HT:indigo	1.8×10^{-8}	80 @650 nm	0.4 @650 nm	-	-	[25]
PTT:PC ₆₀ BM	1.0×10^{-6}	46 @800 nm	0.26 @800 nm	4×10^6	-	[29]
BP3T/SnPc/SnPc:C60/BCP (vacuum)	1.1×10^{-5}	-	0.36 @750 nm	-	-	[30]
Hamamatsu S1227 (Silicon)	-	-	0.36 @700 nm	-	-	SI
UDT Sensors UV-100 (Silicon)	-	-	~0.5 @750 nm	-	-	SI

A common metric of comparison for the detection limit of photodetectors in general is the specific detectivity (D^*). Assuming that the device performance is shot noise limited, the detectivity D^* can be calculated using equation 2:

$$D^* = \frac{R}{\sqrt{2qI_D}} \quad (2)$$

We calculated D^* to be 4.57×10^{12} Jones and 3.98×10^{12} Jones for spray and doctor bladed devices, respectively, using the aforementioned responsivities and dark currents. We note that while the calculation of D^* using equation 2 is a very common approach it may lead to an overestimation of the detectivity.^[31] The calculation of the D^* neglects the contribution of the flicker noise (1/f-noise) and the thermal noise to the total noise current of the photodiode.^[32]

Finally, study the evolution of the J-Vs characteristics of P3HT:O-IDTBR under different light illumination. As depicted in **Figure S7** the current density calculated at -4V follows a linear behavior with the light intensity for more than 4 orders of magnitude, in line with the reduced charge recombination processes observed for O-IDTBR-based devices.^[21]

For practical applications of flat-panel imagers, an important parameter to be determined is the photodiode frequency response. Ideally, the frequency response should be much beyond 30 Hz for appliances with video rate performance. **Figure 2d** shows the dynamic (damping) behavior of P3HT:O-IDTBR diodes in a frequency range between 1 Hz and 1 MHz. From the frequency response, one can extract the commonly reported cut-off frequencies at -3dB and -6dB, which corresponds to the decay of the photocurrent signal to $1/\sqrt{2}$ and $1/2$ of the dc amplitude (non-modulated photocurrent), respectively. For spray coated devices, frequencies of 12.4 kHz (3 dB) and 45 kHz (-6 dB) were measured, whereas in the case of doctor bladed diodes we measure a faster response of 52.6 kHz (-3 dB) and 141.9 kHz (-6 dB). These values are among the highest frequency responses reported for solution processed organic photodiodes (**Table 1**), suggesting that P3HT:O-IDTBR is a promising candidate for imaging applications.^[33,34] While higher frequency responses have been previously reported, those devices relied on evaporated semiconductors.^[35,36] To validate the figure of merit of our NFA based photodiodes, we tested the photosensitivity of the non-fullerene blend in an organic flat panel photodetector. **Figure 3a** shows a reconstructed image of three gears under 532 nm illumination. When illuminating with light corresponding to the tail of the photo-responsivity of O-IDTBR using a 780 nm diode instead, we were still able to clearly detect the object, i.e., P3HT:O-IDTBR photodetectors have the ability of detecting images across the visible and NIR spectrum. The quality (contrast) of an image is often limited by the crosstalk between the pixels of an imager. The crosstalk is best characterized by illuminating the active area over a sharp metal edge covering a region of interest (inset of **Figure 3c**). We observed that the signal falls between the illuminated and dark area within one pixel with a pixel pitch of 98 μm . To quantify the resolution, we calculated the contrast at a fixed spatial frequency, i.e. the modulation transfer function (MTF) as line pairs per millimeter (lp mm^{-1}) in a region of interest (ROI) devoid of major defects.^[37,38] As depicted in the inset of **Figure 3c**, the measured points are linearly fitted and extrapolated to a maximum resolution of 5 lp mm^{-1} ,

which is in line with the pixel pitch. This means that the sensitivity of the photoactive P3HT:O-IDTBR blend is so high as to not obscure the native resolution given by the active matrix thin film transistor panel.

Figure 3 Visible (a) and infrared (b) shadow cast at 532 nm and 780 nm, respectively, of a slide showing gears. The stripes in the images are the results of poorly connected flexible

printed circuits (FPCs) on the contacting pads of the glass and are not due to defects of the active area. c) Image crosstalk shown as the response of a pixel row at a sharp metallic edge under green light illumination. Inset: spatial frequency response of the infrared imager.

In conclusion, we report organic photodetectors based on P3HT and a non-fullerene molecular acceptor: rhodanine-benzothiadiazole-coupled indacenodithiophene (O-IDTBR). Our OPDs feature simultaneously high external quantum efficiency ($\sim 70\%$), low dark current ($J_d = 30 \text{ nA cm}^{-2}$) and, to the best of our knowledge, the highest responsivity in the NIR of 0.42 A W^{-1} for an organic photodetector reported so far. Compared to the workhorse polymer blend P3HT:PCBM, the use of O-IDTBR allows extending the absorption profile beyond 800 nm, a key driver for enhanced OPD operation. The combination of O-IDTBR with P3HT is particularly interesting because it allows NIR detection using a structurally simple and cost effective high bandgap polymer thus circumventing the need for a chemically more complex and thus costly low bandgap polymer.

Moreover, we realized large area flat panel imagers with P3HT:O-IDTBR as the photoactive blend able to detect objects under visible and NIR light conditions with a frequency response that satisfies many applications with video rate requirements. Similar to recent trends in organic photovoltaics it is plausible that the development of small molecule acceptors will further drive the spectral sensitivity and overall efficiency of OPDs. The presented results certainly position OPD technology as major contender for full color detection and image sensing, particularly in scenarios where conformability, weight and shape play a significant role.

Experimental Section

Materials: P3HT and O-IDTBR were synthesized as reported elsewhere.^[24]

Fabrication of Photodetector devices and Imagers: Pre-structured indium tin oxide (ITO) substrates were cleaned with acetone and isopropyl alcohol in an ultrasonic bath for 10 minutes each. After drying, the substrates were spin-coated with 30 nm of zinc oxide (ZnO)^[24] and ~450 nm thick active layer based on P3HT:O-IDTBR (1:1 ratio by weight) (35 g L⁻¹ and 9 g L⁻¹ for, respectively, doctor blading and spray-coating) in Chlorobenzene (CB). To complete the fabrication of the devices 100 nm of MoOx and 100 nm of Al were thermally evaporated through a customized mask (with a 10.4 mm² active area opening) under a vacuum of ~1x10⁻⁶ mbar. Imagers were fabricated under the same conditions on a-Si AM TFT backplane with ITO bottom contacts, with a 256x256 pixel array and a pixel pitch of 98 µm.

J-V measurements: The J-V characteristics were measured using a Keithley 6487 Picoammeter in an electrically and optically shielded box. For illumination, green light-emitting diodes (LED) with a peak emission wavelength of 532 nm at a power density of 780 µW cm⁻² were used. The EQE was measured using a set up illuminated by a xenon arc lamp with the monochromator chopped at 170 Hz, a preamplifier (Femto DHPA-100) and a lock-in amplifier (EGG&G5210). A calibrated c-Si photodiode was used as reference.

Cut-off: Dynamic measurements were made in an electrically and optically shielded box with a transimpedance amplifier (FEMTO DHPA-100) connected to a digital oscilloscope (Tektronix MDO3034). Pulsed green light was applied through the transparent ITO electrode using a high power LED and a signal waveform generator (TGA 1242) to create a sinusoidal wave with frequencies ranging from 1 Hz to 1 MHz. The calculated RC frequency (f_{RC}) of the devices were ~ 6 MHz.^[34]

Imager and characterization

The imager is measured through a custom-made readout and driving electronics, based on the highly integrated readout IC (ROIC) ISC9717 from FLIR, which enables to simultaneously integrate, amplify, low pass filter and analog-to-digital convert (14 bit) the input signal allowing low noise imaging even at low light intensities. The common top electrode of the pixels was biased at -5V and the ROIC was operated in an electrode collection mode. The light source was triggered with the readout electronics with an illumination time of 10 ms per image.

Image post processing using a low frequency filter and analysis are carried out using the free software ImageJ (<https://imagej.nih.gov/ij/plugins/se-mtf/index.html>).

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Supporting Information ((delete if not applicable))

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Supplementary Information

Visible and Near-infrared imaging with non-fullerene based photodetectors

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Figure S1. J-V characteristics of P3HT:O-IDTBR at 1 sun (a), from 1 sun to 0.01 sun (b) and from 0.01 to 0.001 (c) sun condition. d) FF values extracted from a,b,c, as a function of light intensity.

Figure S2. Visible shadow cast under green illumination (532) nm of a slide showing a butterfly (a) and the Gioconda portrait (b).

Figure S3. External quantum efficiency (EQE) of doctor bladed (a) and spray coated (b) P3HT:O-IDTBR photodetector as a function of reverse bias.

Figure S4. Responsivity of spray coated P3HT:O-IDTBR photodetector as a function of reverse bias.

Figure S5. Responsivity of commercial available Hamamatsu S1227 photodetector. Source: Hamamatsu website:

<https://www.hamamatsu.com/jp/en/product/category/3100/4001/4103/S1227-1010BQ/index.html#1328447245601>

Figure S6. Responsivity of commercial available UDT Sensors UV-100 photodetector.

Source: UDT website:

<http://www.perfectelectronicparts.com/productdetail.php?q=FILE-UV100&mfname=UDT+SENSORS+INC>

[Figure S7. Current density calculated at -4V as a function of light intensity of P3HT:O-IDTBR photodetector.](#)