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# 1 Enhanced Photomultiplication in Filter-Free Organic Photodetectors for

## 2 Red and NIR Light Sensing Using Minimal Nonfullerene Blends

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### 20 Calculations

21 The external quantum efficiency and responsivity values of OPDs were derived according to

22 the number of collected charges and the number of incident photons by Equations (1-3):<sup>[1]</sup>

$$EQE = \frac{J_{\rm ph}/e}{P_{\rm in}/h\nu},\tag{1}$$

$$EQE = \frac{\chi \tau \mu V}{L^2}$$
(2)

25 
$$R = \frac{J_{\rm ph}}{P_{\rm in}} = \frac{EQE \cdot e}{hv}$$
(3)

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27  $J_{\rm ph}$  is the photogenerated current density (the measured current density under light ( $J_{\rm L}$ ) minus 28 the dark current density ( $J_{\rm d}$ )), *h* is the Planck constant, and *v* is the frequency of incident light, 29 *hv* is photon energy, and  $P_{\rm in}$  is the illumination power density.  $\chi$  is the ratio of captured carriers 30 to the total exciton dissociation rate,  $\tau$  is the lifetime of the captured carriers, *V* is the value of 1 the applied bias voltage,  $\mu$  is the mobility of the uncaptured carriers, and *L* is the device 2 thickness.

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4  $D^*$  of the device calculated from  $i_n$  is according to formula (3-4):<sup>[2]</sup>

$$D^* = \frac{R\sqrt{AB}}{i_n},\tag{4}$$

6 A is the active area of the PM-OPDs, B is the bandwidth.

Dynamic range (DR) is referred as the predictable photoresponse generated from the highest and lowest detectable illumination power densities, when the slope ( $\beta$ ) of the photocurrent density versus light intensity curve is less than 1, exhibiting the sublinear response. It often exists in the photodetectors with gain or photomultiplication. The photodiodes often show a linear dynamic range (LDR) with  $\beta$ =1. Thus the DR and LDR of the devices are calculated by the following equations: <sup>[3-5]</sup>

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$$DR = 20\log\frac{P_{\text{high}}}{P_{\text{low}}} \neq 20\log\frac{I_{\text{high}}}{I_{\text{low}}}$$
(5)

$$LDR = 20\log\frac{P_{\text{high}}}{P_{\text{low}}} = 20\log\frac{I_{\text{high}}}{I_{\text{low}}}$$
(6)

where  $P_{\text{high}}$  and  $P_{\text{low}}$  are the highest and lowest detectable illumination power densities of the photocurrent density versus light intensity curve.  $I_{\text{high}}$  and  $I_{\text{low}}$  are the highest and lowest photocurrent density of the photocurrent density versus light intensity curve.

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#### **Supplementary Figures**





20 Figure S1 (a) J-V characteristics measured for the PM-OPDs with a 2.8 µm thick P3HT active 21 layer in dark and under illumination of the 660 nm LED having a power density of 1.27

- $1\,$  mW/cm². (b) EQE spectra measured for the P3HT based PM-OPDs with different cathode
- 2 contacts, operated under the bias of -60 V.



- 4 Figure S2 Schematic diagrams illustrating the formation of traps at the (a) P3HT/Al, (b)
- 5 P3HT/Cu (Ag) interfaces.



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7 Figure S3 Schematic diagram illustrating the tunneling hole injection at the P3HT/A1 interface

8 in a P3HT-based PM-OPD, operated under a reverse bias, under illumination of a 660 nm LED.



2 Figure S4 Absorption spectra of the acceptors used in preparation of different PM-OPDs in this

3 work.



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5 Figure S5 Semi-log EQE spectra of the P3HT:Y6 -based PM-OPD operated at different biases.





2 Figure S6 Optical field distribution calculated for PM-OPDs with 2.8 µm thick active layers of

3 (a) P3HT:PCBM (100:1) and (b) P3HT:IT-4F (100:1), and photogenerated electron profiles

4 calculated for PM-OPDs with a 2.8 μm thick active layers of (c) P3HT:PCBM (100:1) and (d)

5 P3HT:IT-4F (100:1).



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7 Figure S7 (a) J-V characteristic, and (b) EQE spectra operated under the bias of -60 V for 8 P3HT:Y6 BHJ PM-OPD with different active layer thicknesses.



2 Figure S8 (a) *J-V* characteristic (b) semi-log EQE spectra operated under the bias of -60 V for
3 P3HT:Y6 BHJ PM-OPDs with different D:A ratio.



5 Figure S9 Response times for the P3HT:Y6 BHJ PM-OPD, operated at the bias of -60 V, under

6 illumination of LEDs with emission peak wavelengths of (a) 660 nm and (b) 850 nm.



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- 1 Figure S10 Transient responses measured for the P3HT:Y6 BHJ PM-OPD, operated at different
- 2 bias, under illumination of LEDs with emission peak wavelengths of 660 nm and 850 nm.
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Frequency (FI2)
Figure S11 Noise spectral density measured for P3HT:Y6 (100:1)-based PM-OPD operated





- 7 8
- 9 Figure S12 Frequency response of the P3HT:Y6 (100:1)-based PM-OPD operated under bias
  10 of -60 V.



2 Figure S13  $R(\lambda)$ – $P_{in}$  characteristics measured for the P3HT:Y6 (100:1)-based PM-OPD under

3 illumination of (a) 660 nm and (b) 850 nm LEDs.



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- 5 Figure S14 J-V characteristics in dark and under illumination (850 nm, 1.27 mW/cm<sup>2</sup>) of the
- 6 freshly prepared P3HT:Y6 BHJ PM-OPD device and its corresponding performances after the
- 7 device aged in air over one week.
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