Supporting Information

The Role of Blend Ratio in Polymer:Fullerene Phototransistors

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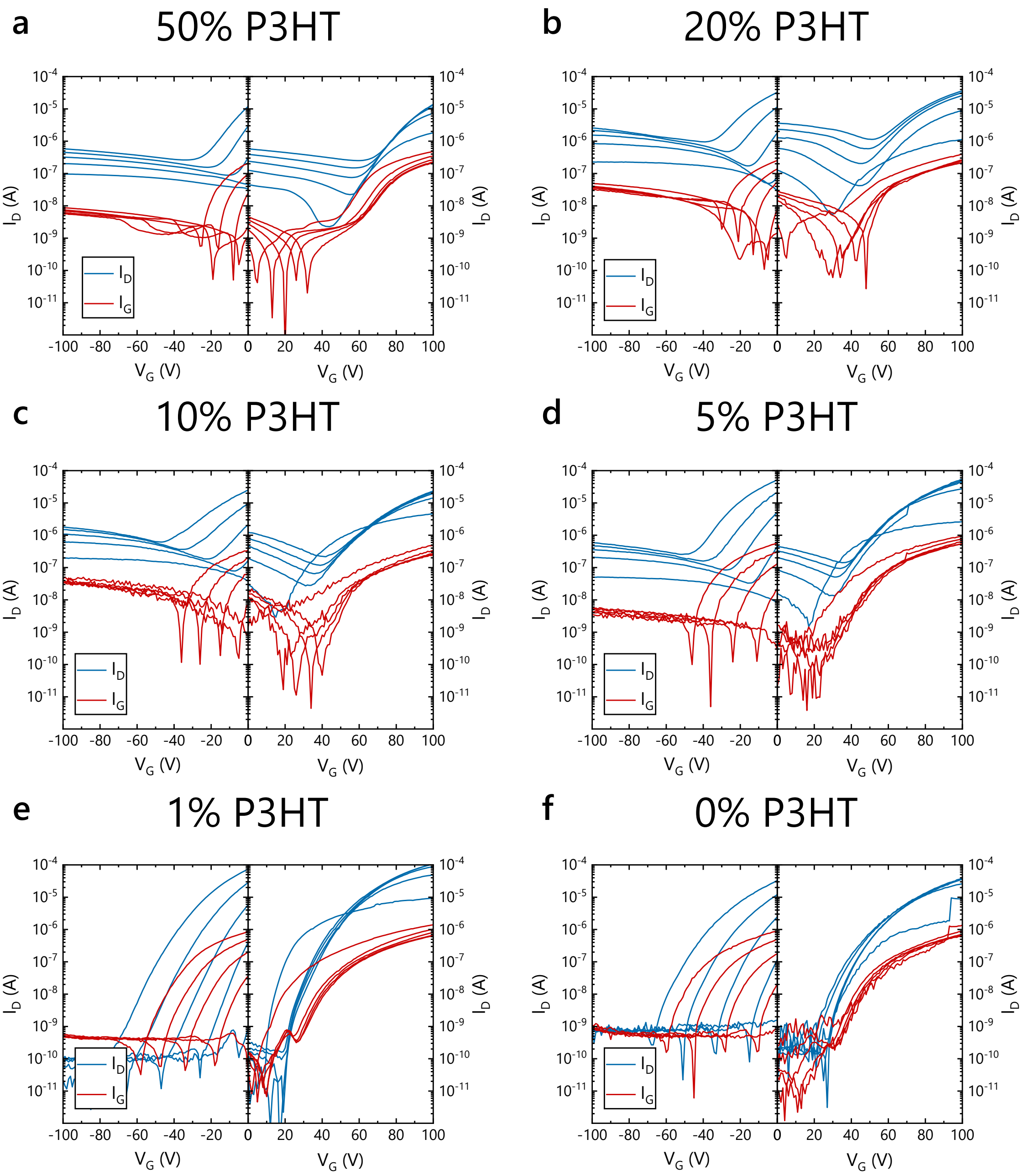
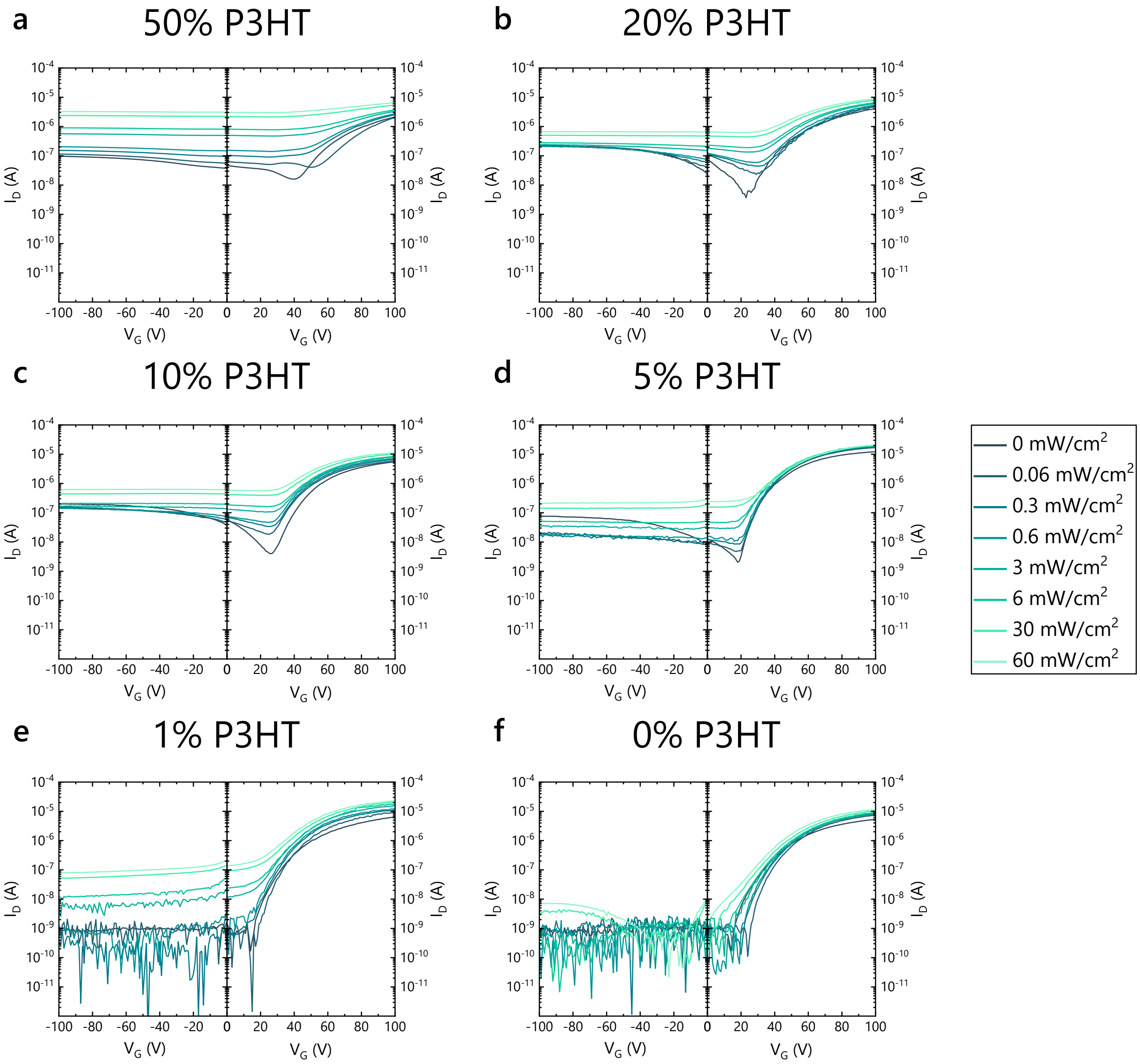
S1. Transfer Characteristics in the Dark



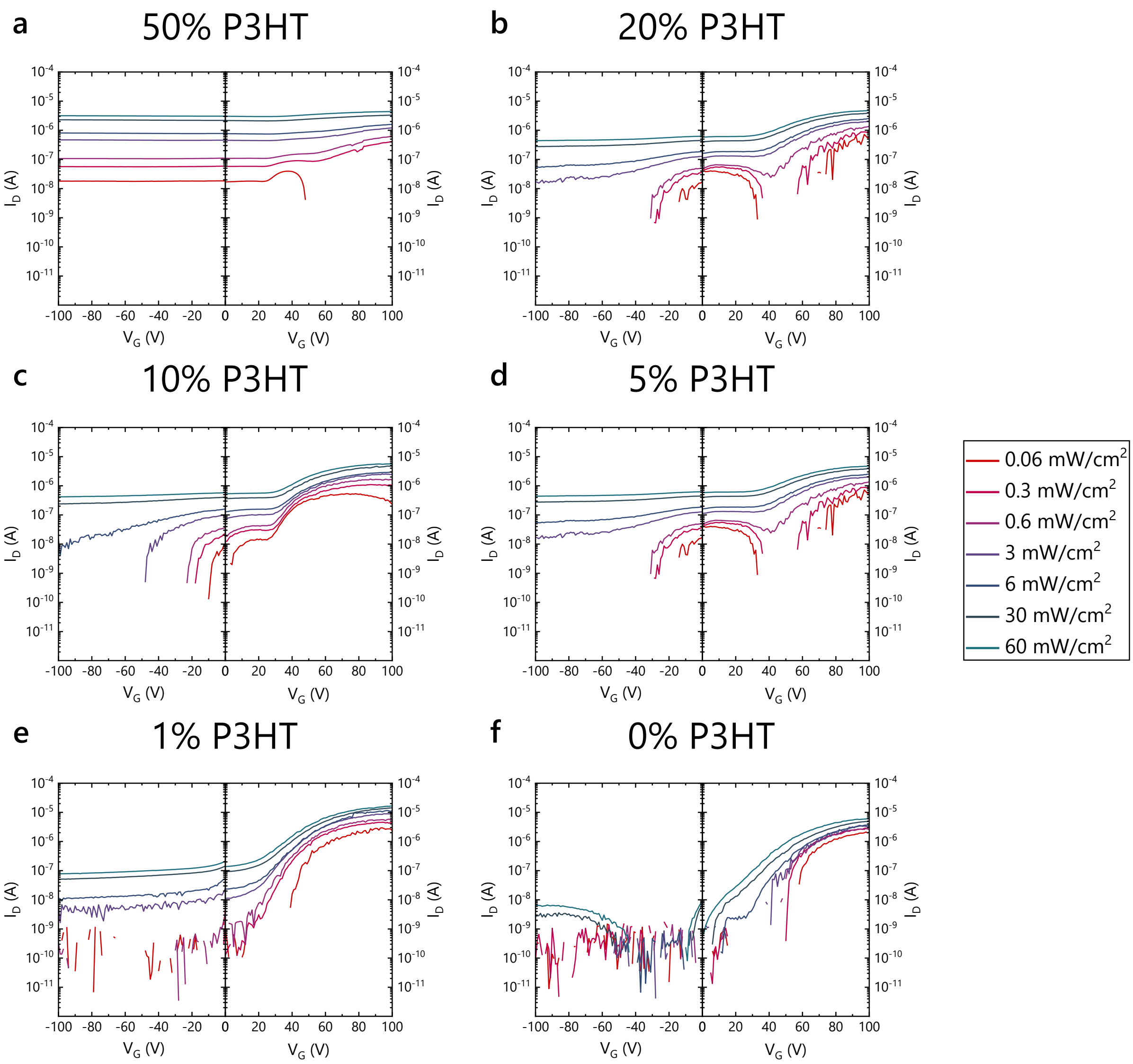
Supplementary Figure S1 p-Type (left) and n-Type (right) transfer characteristics (source-drain current, I_D, as a function of gate voltage, V_G) of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C$_{61}$-butyric acid methyl ester (PCBM) blend thin-film transistors (TFT) measured in the dark. The weight % of P3HT was **a** 50%, **b** 20%, **c** 10%, **d** 5%, **e** 1%, **f** 0%. Source-drain current (I_D) is shown in blue and gate leakage current (I_G) is shown in red. Each device has a channel length of 20 μm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. Each line is a measurement carried out at a different drain voltage (V_D) between V_D = -20V and -100V for p-type measurements and V_D = 20V and 100V for n-type measurements. Devices were measured under atmospheric pressure N_2 at room temperature. All TFTs measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text.

S2. Transfer Characteristics under Illumination



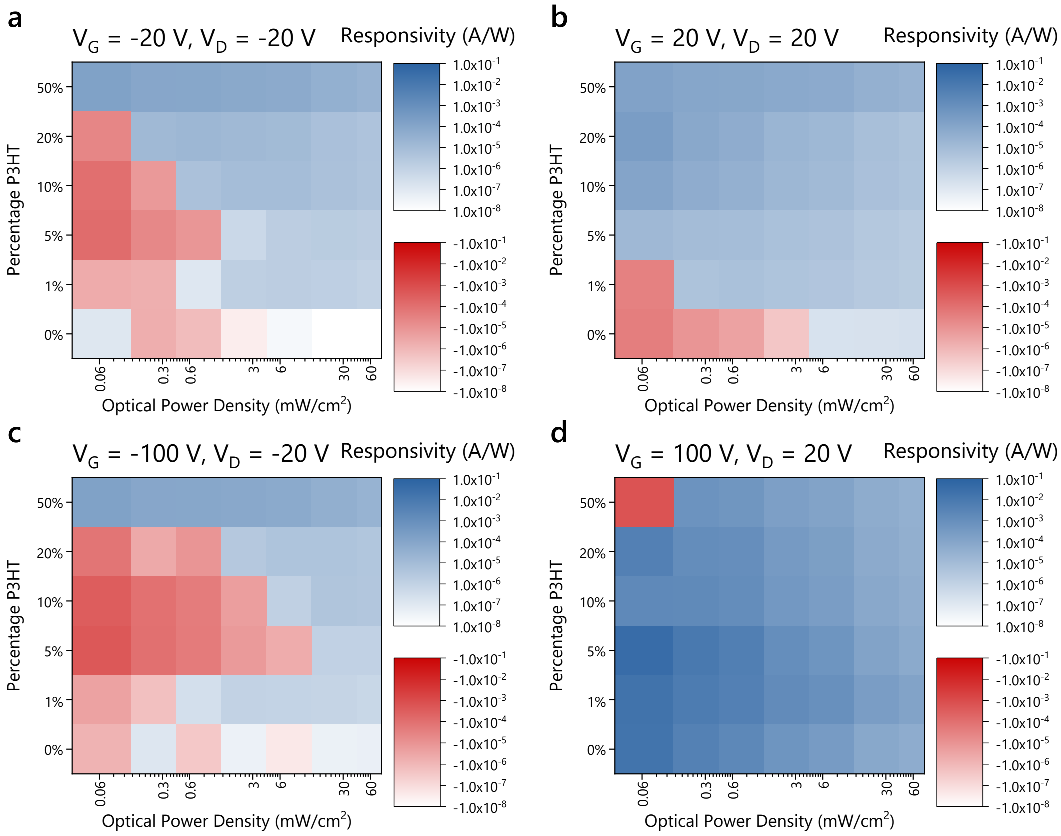
Supplementary Figure S2 p-Type (left) and n-Type (right) transfer characteristics (source-drain current, I_D, as a function of gate voltage, V_G) for thin film transistors (TFTs) formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM), measured under various intensities of green (λ_{max} = 525 nm) light. The weight % of P3HT was **a** 50%, **b** 20%, **c** 10%, **d** 5%, **e** 1%, **f** 0%. All TFTs measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text. All devices had a channel length of 20 µm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. Drain voltages of V_D = -20 V were applied for negative V_G (left panels) and V_D = +20V, were applied for positive V_G (right panels). All devices were measured under atmospheric pressure N_2 at room temperature. Legend shows incident optical power density.

S3. Photocurrent



Supplementary Figure S3 p-Type (left) and n-Type (right) photocurrent ($I_{\mathrm{ph}} = I_D^{(\mathrm{illuminated})} - I_D^{(\mathrm{dark})}$) for thin film transistors (TFTs) formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM), measured under various intensities of green (λ_{max} = 525 nm) light. The weight % of P3HT was **a** 50%, **b** 20%, **c** 10%, **d** 5%, **e** 1%, **f** 0%. Negative values of I_{ph} are not displayed. All TFTs measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text. All devices had a channel length of 20 µm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. Drain voltages of V_D = -20 V were applied for negative V_G (left panels) and V_D = +20V, were applied for positive V_G (right panels). All devices were measured under atmospheric pressure N_2 at room temperature. Legend shows incident optical power density.

S4. Responsivity



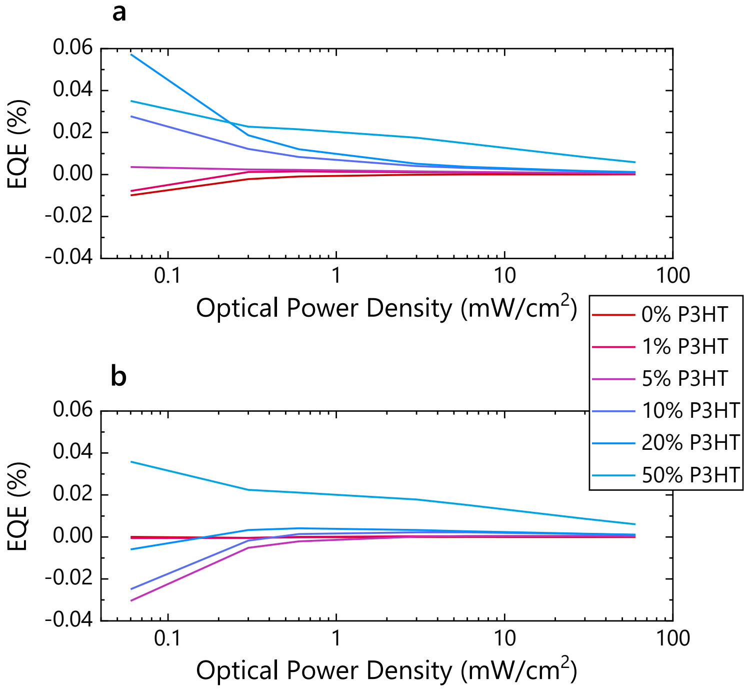
Supplementary Figure S4 Responsivity of phototransistors formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM) with various weight % of P3HT, measured under various intensities of green (λ_{max} = 525 nm) light. Responsivities were extracted with applied drain (V_D) and gate voltages (V_G) of **a** V_D = -20 V and V_G = -20 V, **b** V_D = 20 V and V_G = 20 V, **c** V_D = -20 V and V_G = -100 V, **d** V_D = 20 V and V_G = 100 V, respectively. Blue represents positive responsivities and red represent negative responsivities. All phototransistors measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text and had a channel length of 20 µm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. All devices were measured under atmospheric pressure N_2 at room temperature.

S5. External Quantum Efficiency

External Quantum Efficiency (EQE) can be evaluated from responsivity via Equation S1:[1]

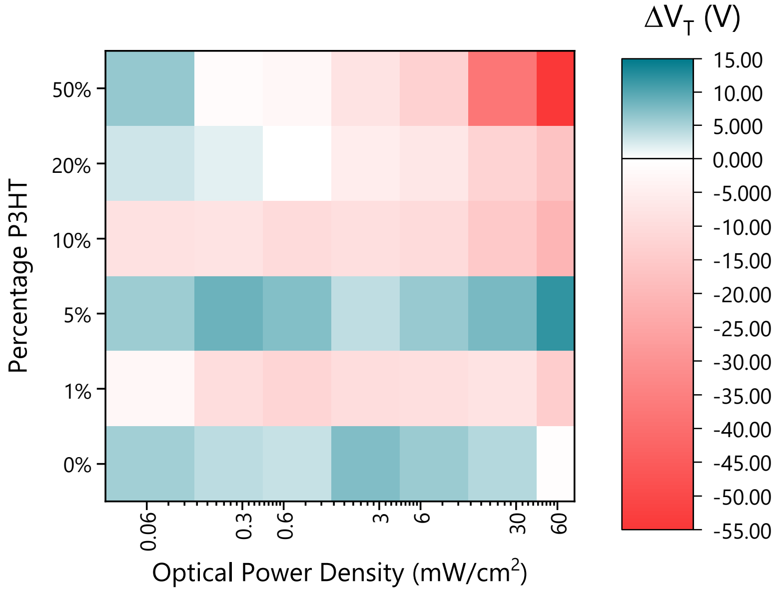
$$EQE = R\frac{hc}{e\lambda} = \frac{I_{\mathrm{ph}}}{PLW}\frac{hc}{e\lambda} \tag{S1}$$

Here R is the responsivity, h is Planck's Constant, c is the speed of light *en vacuo*, e is the fundamental unit of change, λ is the wavelength of incident photons (we here use the peak wavelength λ = 525 nm), I_{ph} is the photo-induced change in drain current under illumination ($I_{\mathrm{ph}} = I_D^{(\mathrm{illuminated})} - I_D^{(\mathrm{dark})}$), P is the incident optical power density, L is the transistor channel length, and W is the transistor channel width. In Figure S5 the EQE is evaluated as a function of blend ratio and incident optical power density for applied voltages of **a** V_D = -20V, V_G = -20V and **b** V_D = 20V, V_G = 20V.



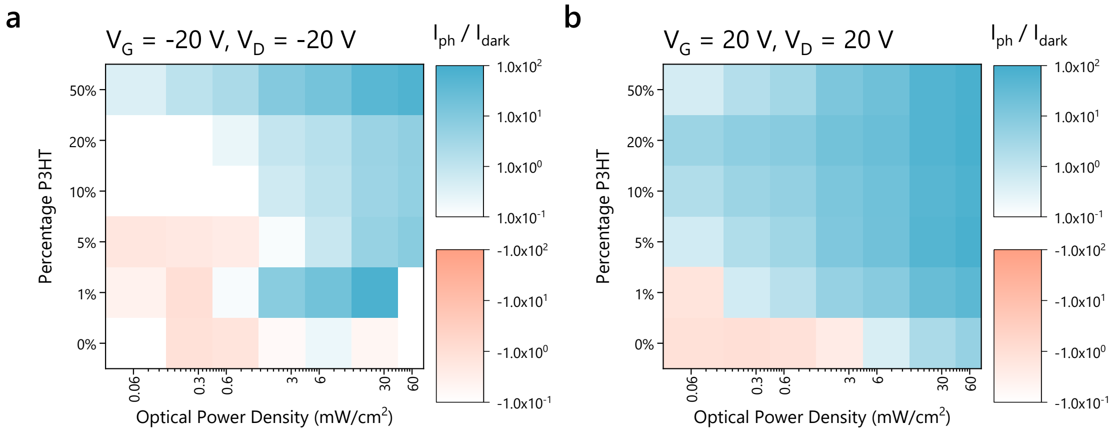
Supplementary Figure S5 External Quantum Efficiency (EQE) of phototransistors formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM) with various weight % of P3HT, measured under various intensities of green (λ_{max} = 525 nm) light, evaluated using Equation S1. EQEs were extracted with applied drain and gate voltages of **a** V_D = -20 V and V_G = -20 V, and **b** V_D = 20 V and V_G = 20 V, respectively. All phototransistors measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text and had a channel length of 20 μm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. All devices were measured under atmospheric pressure N_2 at room temperature.

S6. Change in Threshold Voltage under Illumination



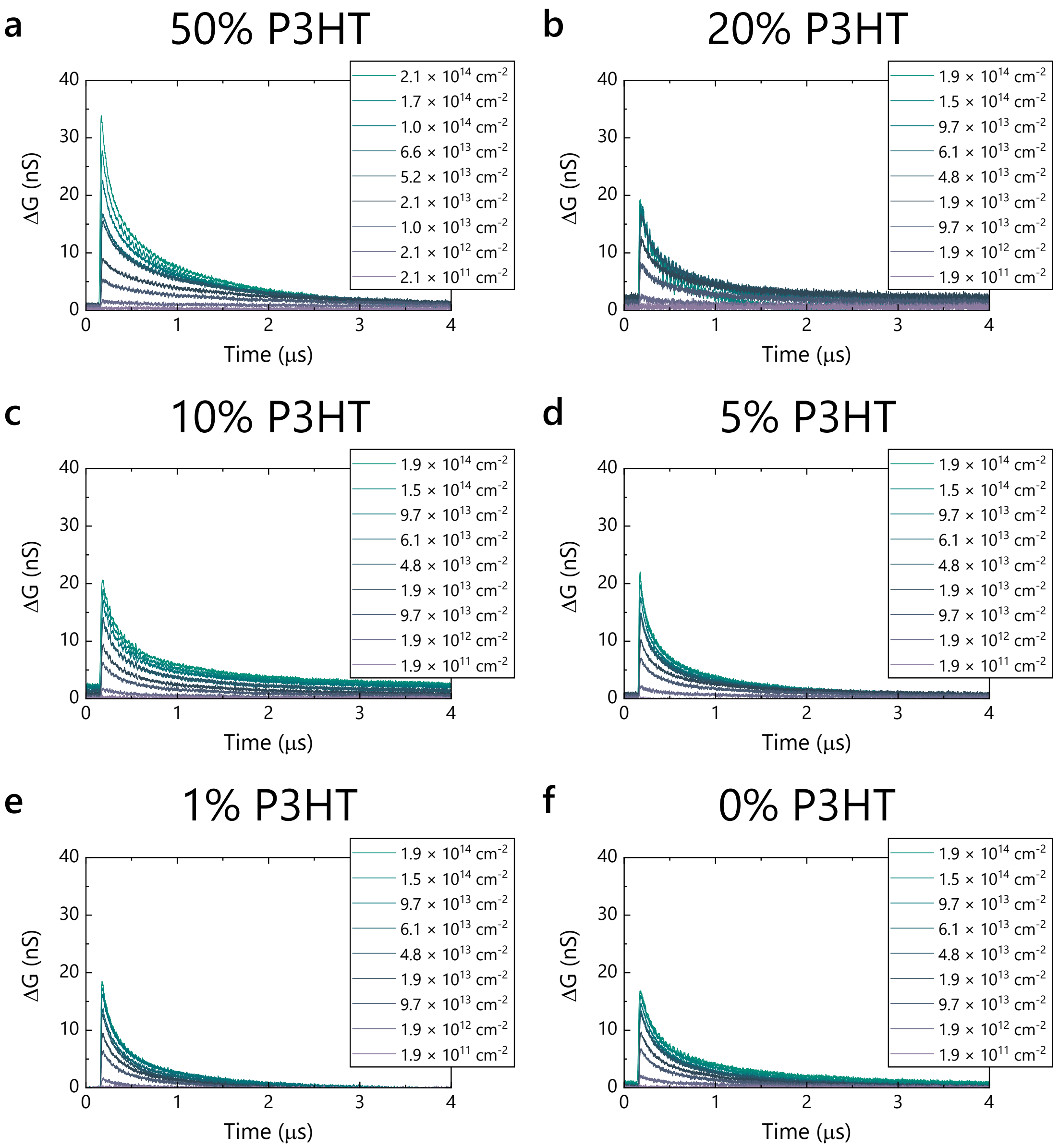
Supplementary Figure S6 Change in n-channel threshold voltage (ΔV_T) of phototransistors formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM) with various weight % of P3HT, measured under various intensities of green (λ_{max} = 525 nm) light. ΔV_T was extracted with an applied drain voltage of V_D = 20 V. Green represents positive changes in V_T and red represent negative changes in V_T. All phototransistors measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text and had a channel length of 20 μm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. All devices were measured under atmospheric pressure N_2 at room temperature.

S7. Ratio of Photocurrent to Dark Current

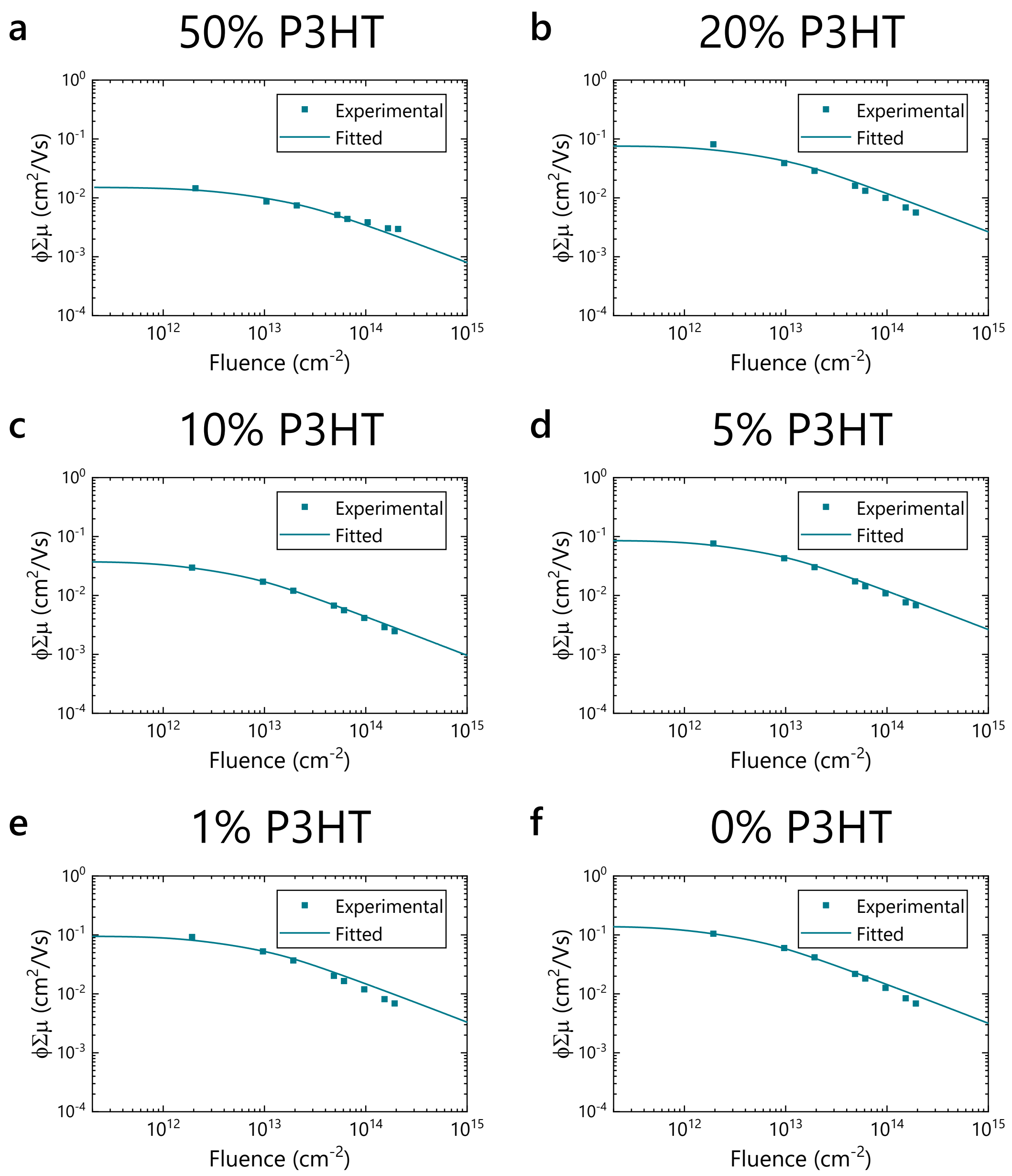


Supplementary Figure S7 Ratio of photocurrent ($I_{\mathrm{ph}} = I_D^{(\mathrm{illuminated})} - I_D^{(\mathrm{dark})}$) to dark current ($I_D^{(\mathrm{dark})}$) for phototransistors formed of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM) with various weight % of P3HT, measured under various intensities of green ($\lambda_{\max}$ = 525 nm) light. Ratios were extracted with applied drain (V_D) and gate voltages (V_G) of **a** V_D = -20 V and V_G = -20 V, **b** V_D = 20 V and V_G = 20 V, respectively. All phototransistors measured were in the bottom-gate, bottom-contact (BGBC) architecture, as shown in Figure 1a of the main text and had a channel length of 20 µm, a channel width of 1 cm, and a dielectric capacitance of 15 nF/cm^2. All devices were measured under atmospheric pressure N_2 at room temperature.

S8. Time-Resolved Microwave Conductivity (TRMC) Data



Supplementary Figure S8 Photoconductance (ΔG) of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM) as a function of time before, during, and after excitation from a green (532 nm) laser pulse. The weight % of P3HT was **a** 50%, **b** 20%, **c** 10%, **d** 5%, **e** 1%, **f** 0%. The fluence of the laser is provided in the legends. All measurements were carried out using time-resolved microwave conductivity (TRMC) in air at room-temperature. The laser pulse had a full width half-maximum (FWHM) of roughly 5 ns in all cases.



Supplementary Figure S9 TRMC figure of merit and proxy for carrier mobility, $\phi\Sigma\mu$, as a function of incident laser fluence for of blends of poly(3-hexylthiophene-2,5-diyl) (P3HT) and phenyl-C_{61}-butyric acid methyl ester (PCBM). The weight % of P3HT was **a** 50%, **b** 20%, **c** 10%, **d** 5%, **e** 1%, **f** 0%. $\phi\Sigma\mu = \phi(\mu_e + \mu_h)$ where μ_e and μ_h are the average electron and hole mobilites measured over the sample area, respectively. ϕ is the carrier-generation efficiency, i.e. the number of electron-hole pairs generated per absorbed photon ($\phi \in [0,1]$). The points are experimental values and the lines are fits to a numerical model accounting for bimolecular and Auger recombination during the laser pulse.[2] All measurements were carried out using time-resolved microwave conductivity (TRMC) in air at room-temperature. The laser pulse had a full width half-maximum (FWHM) of roughly 5 ns in all cases.

Supporting References

(1) Baeg, K.-J.; Binda, M.; Natali, D.; Caironi, M.; Noh, Y.-Y. Organic Light Detectors: Photodiodes and Phototransistors. *Adv. Mater.* **2013**, *25* (31), 4267–4295. https://doi.org/10.1002/adma.201204979.

(2) Labram, J. G.; Chabinyc, M. L. Recombination at High Carrier Density in Methylammonium Lead Iodide Studied Using Time-Resolved Microwave Conductivity. *J. Appl. Phys.* **2017**, *122* (6), 065501. https://doi.org/10.1063/1.4990802.