Electronic Supplementary Information

Dark Current Reduction Strategies Using Edge-on Aligned Donor Polymers for High Detectivity and Responsivity Solution-processed Organic Photodetectors

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Synthesis of monomers

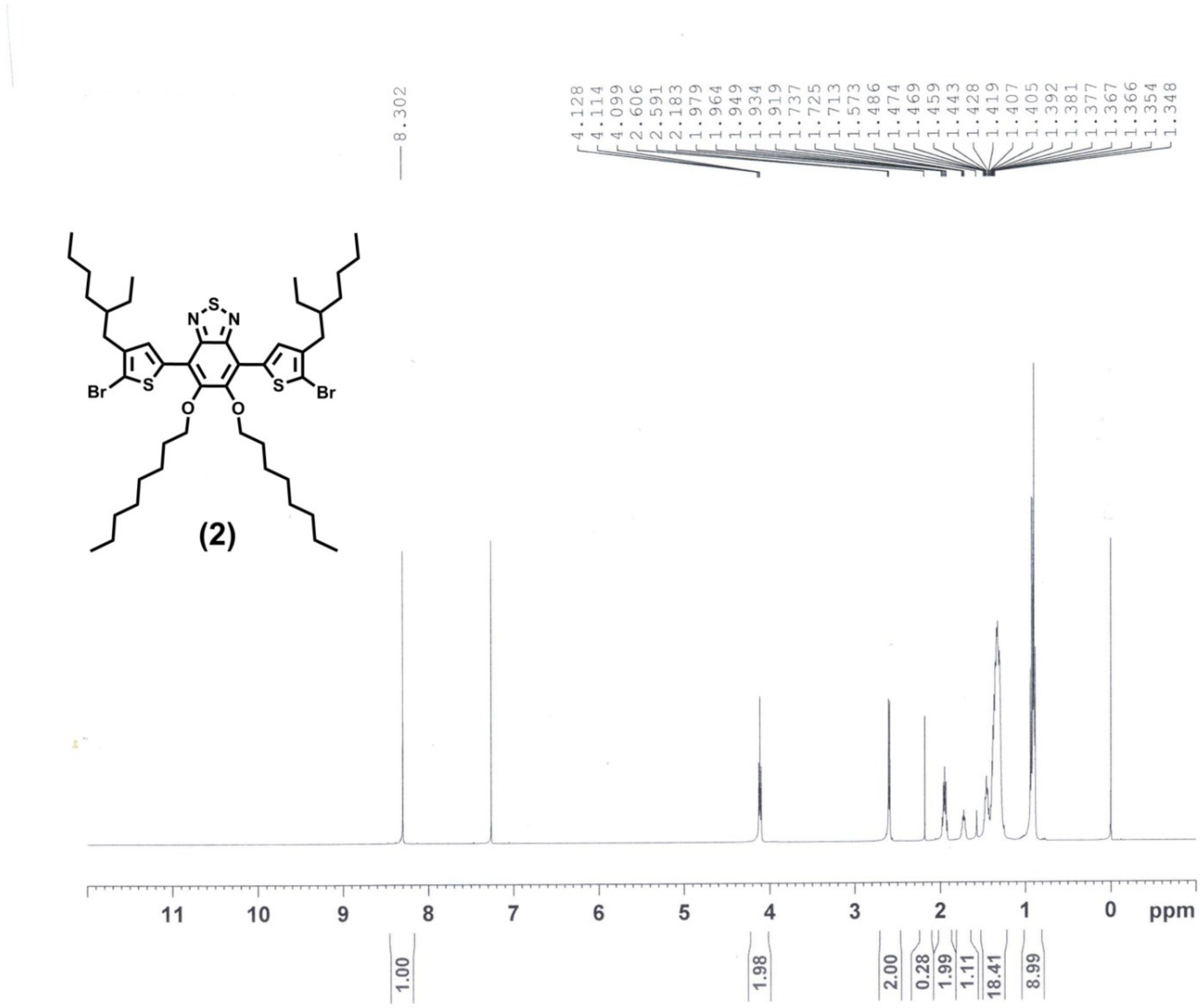


Figure S1. ^{1}H NMR spectra of 4,7-bis(5-bromo-4-(2-ethylhexyl)thiophen-2-yl)-5,6-bis(octyloxy)benzo[*c*][1,2,5]thiadiazole (2)

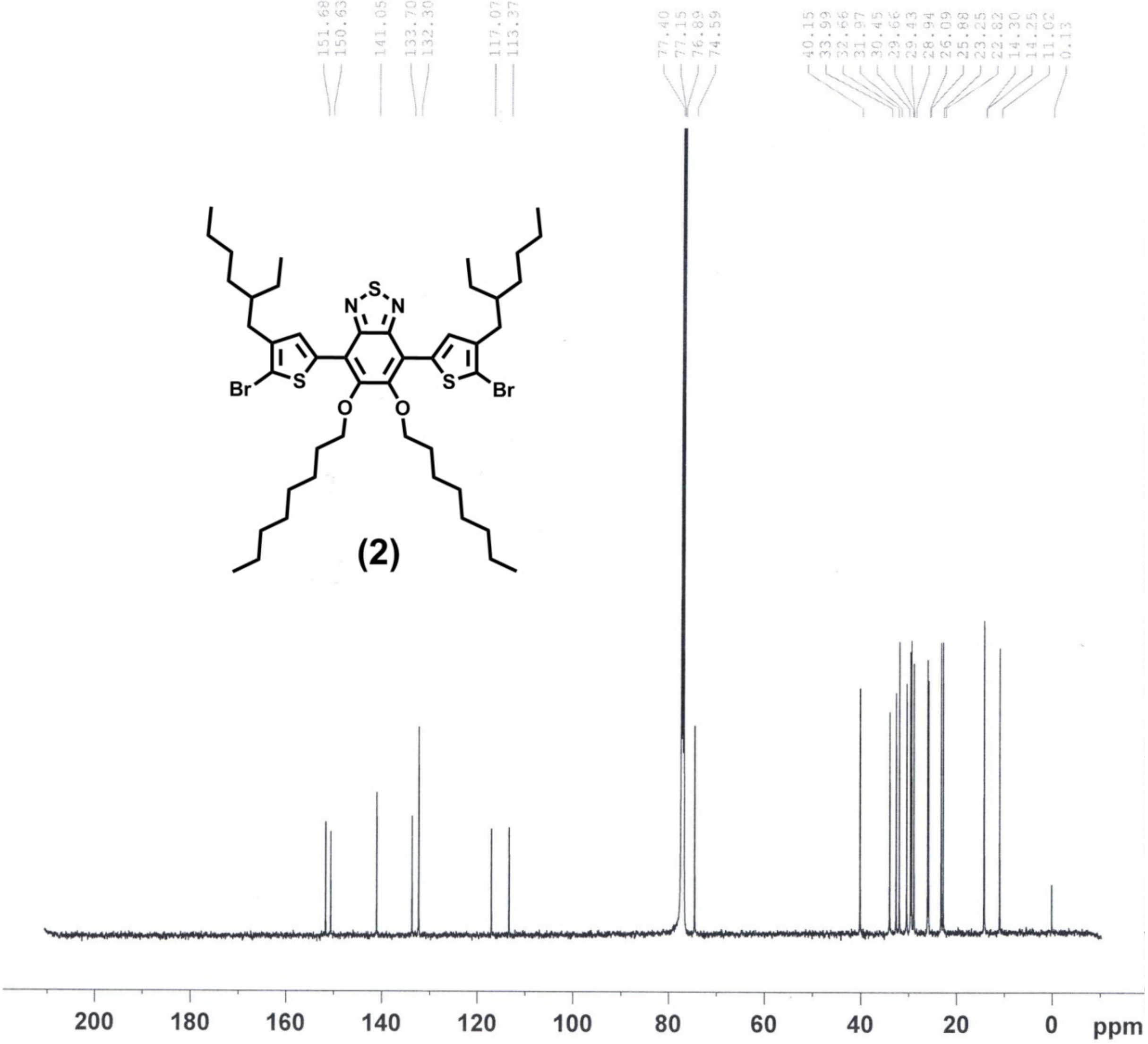


Figure S2. ^{13}C NMR spectra of 4,7-bis(5-bromo-4-(2-ethylhexyl)thiophen-2-yl)-5,6-bis(octyloxy)benzo[*c*][1,2,5]thiadiazole (2)

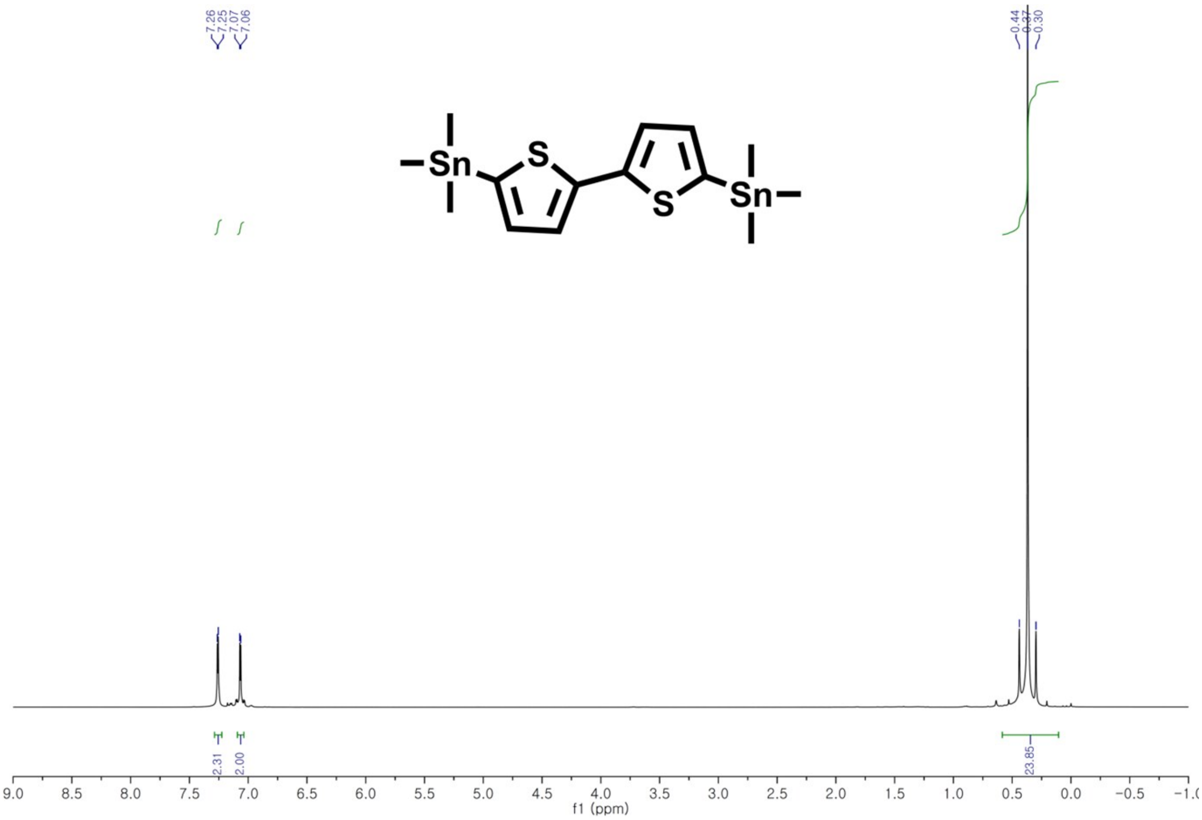


Figure S3. ^{1}H NMR spectra of 5,5'-bis(trimethylstannyl)-2,2'-bithiophene

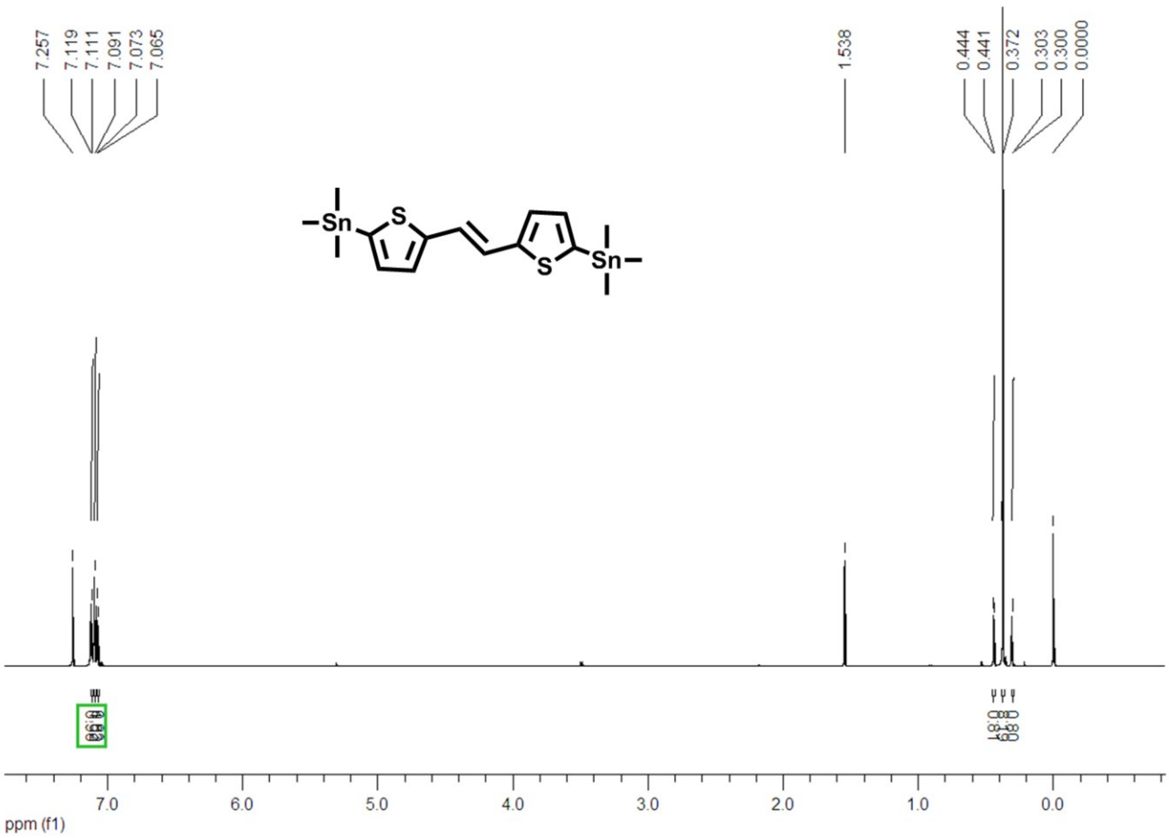


Figure S4. ^{1}H NMR spectra of (*E*)-1,2-bis(5-(trimethylstannyl)thiophen-2-yl)ethene

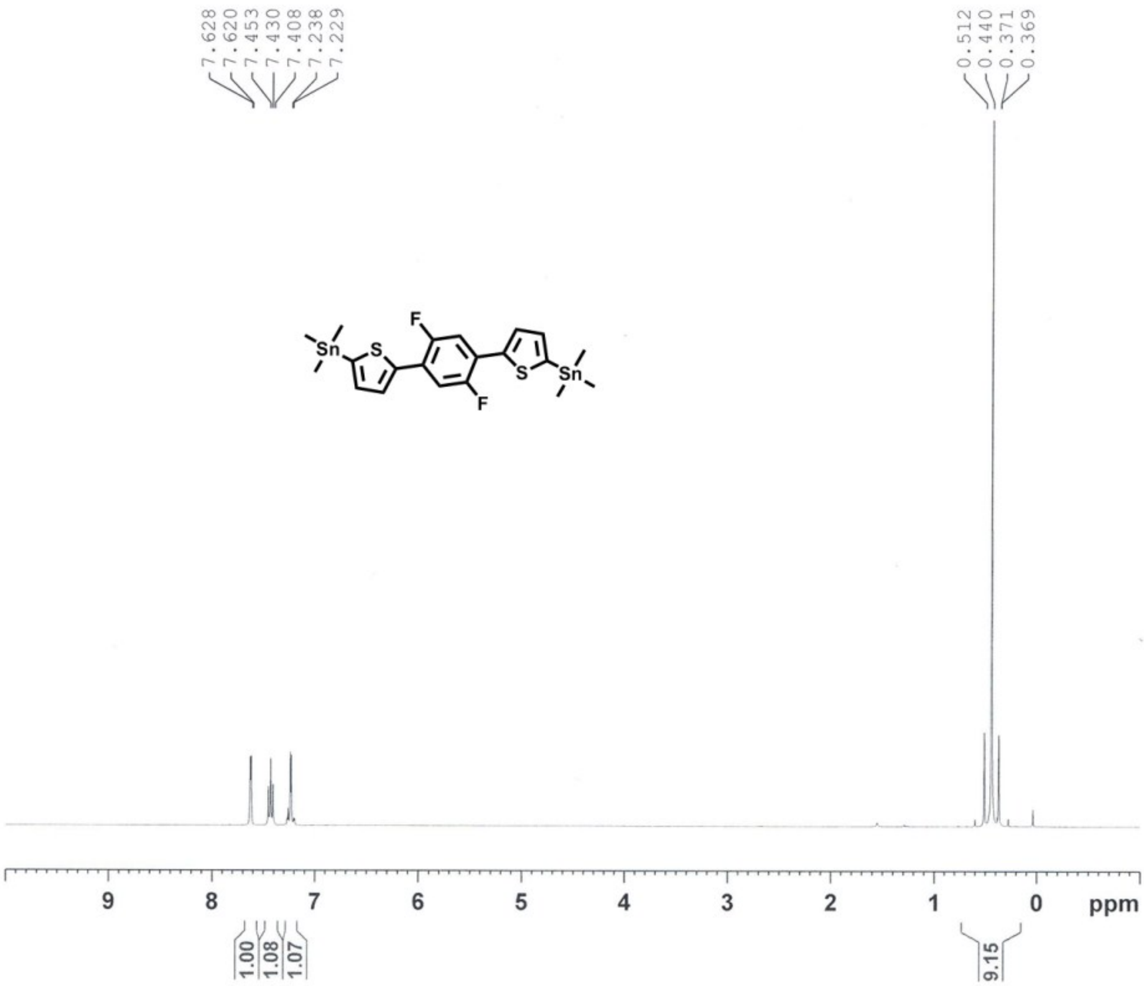


Figure S5. ^{1}H NMR spectra of ((2,5-difluoro-1,4-phenylene)bis(thiophene-5,2-diyl))bis(trimethylstannane)

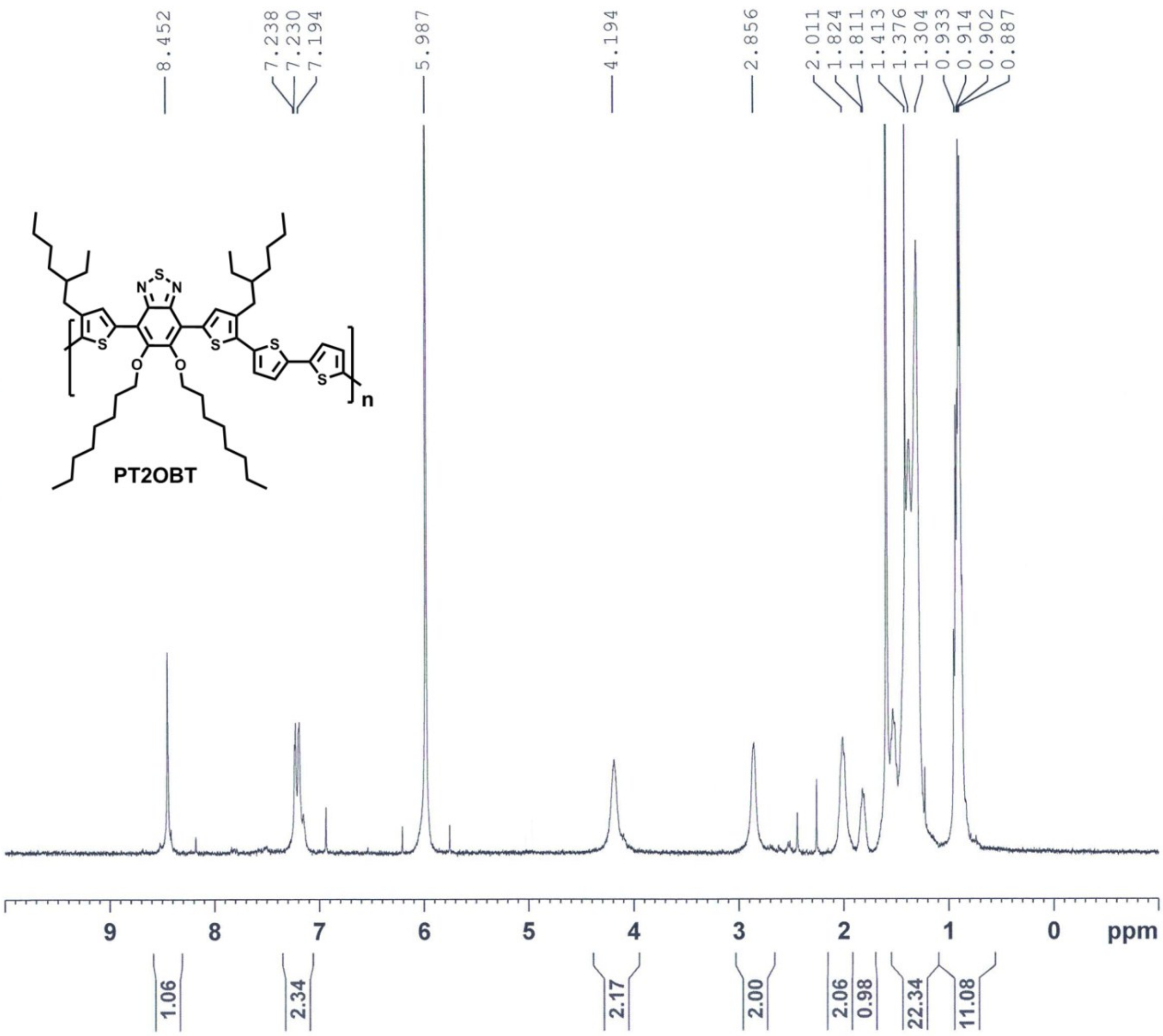


Figure S6. [1]H NMR spectrum of PT2OBT

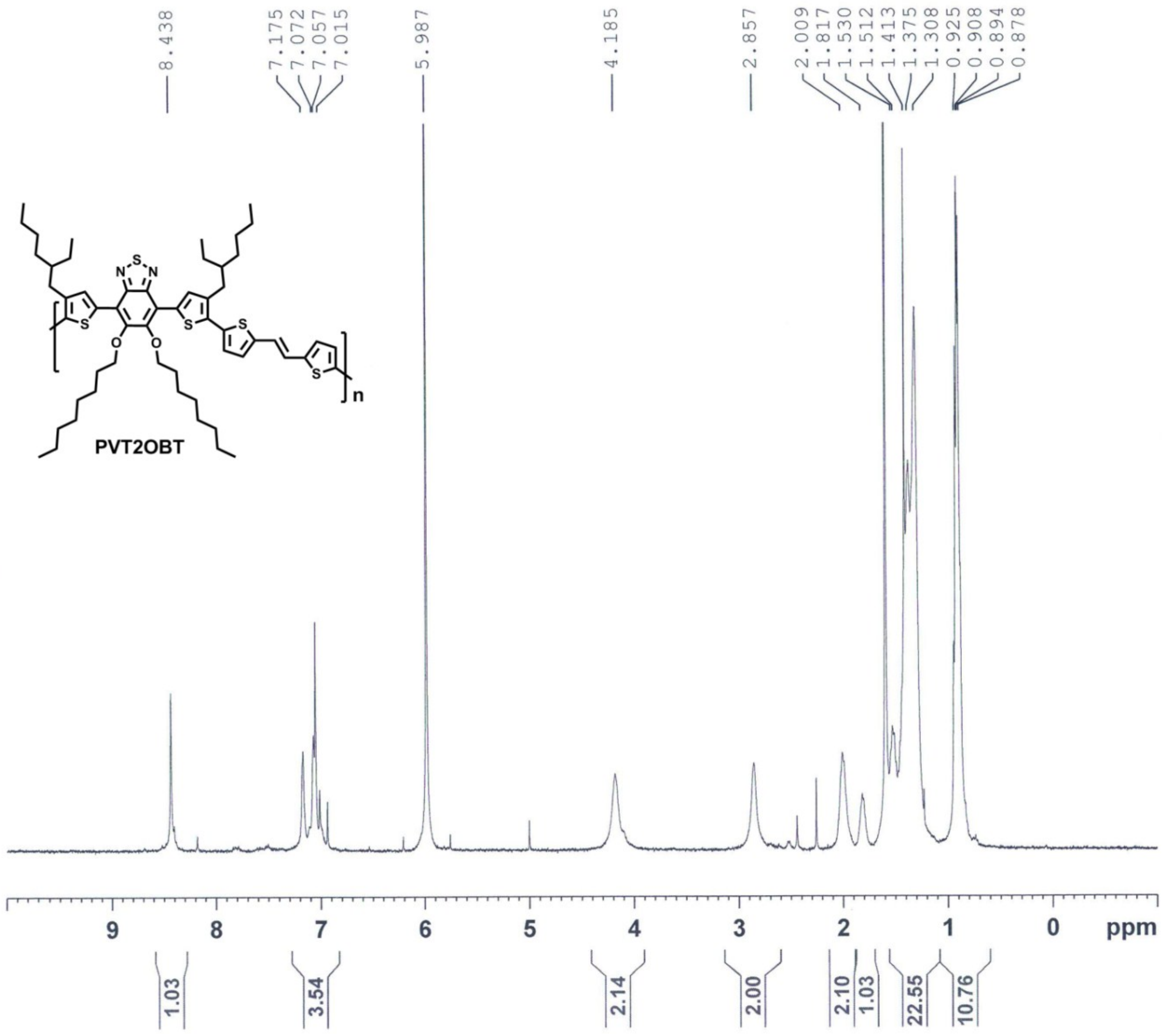


Figure S7. ^{1}H NMR spectrum of PVT2OBT

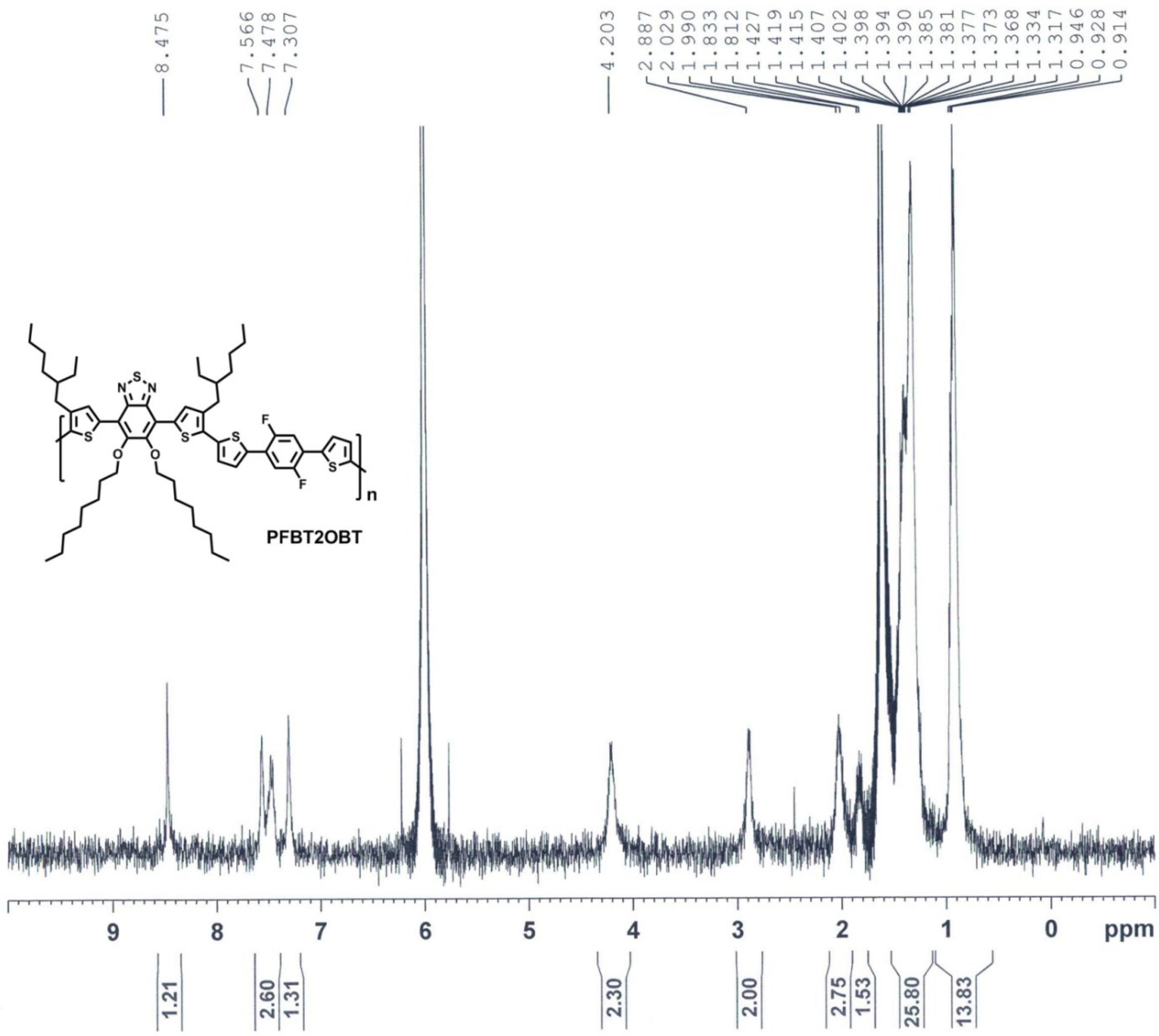
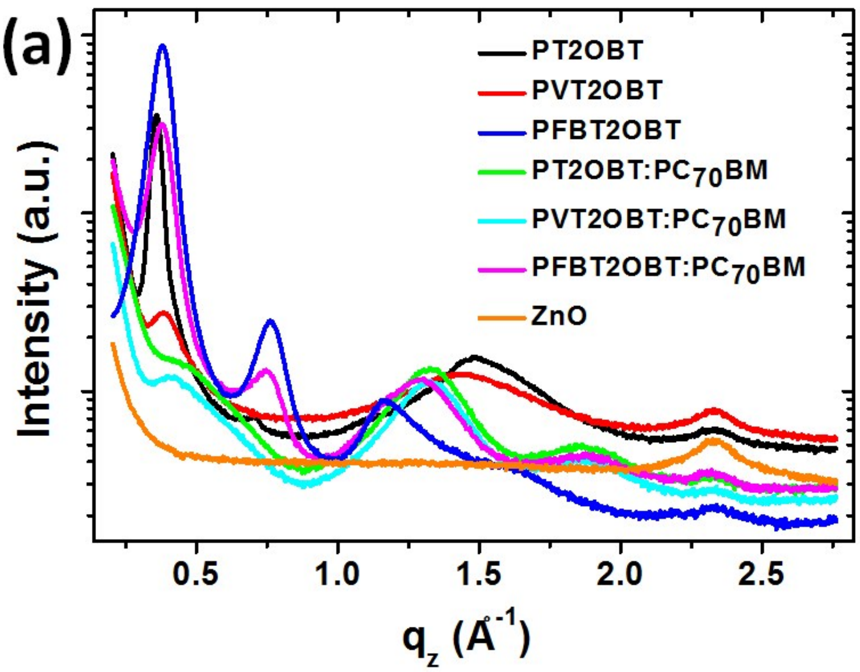


Figure S8. [1]H NMR spectrum of PFBT2OBT



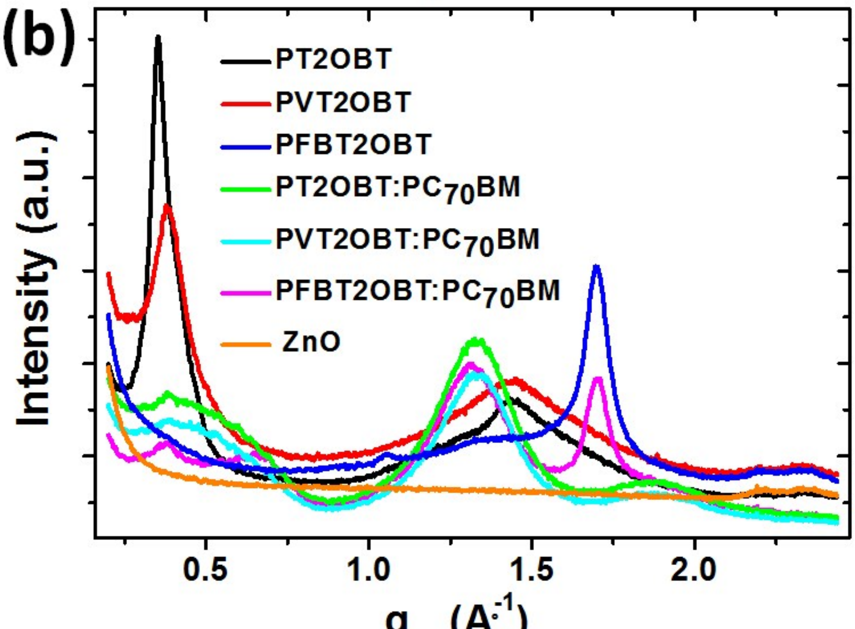


Figure S9. (a) Out-of-plane and (b) in-plane line cuts of neat polymers (PT2OBT, PVT2OBT and PFBT2OBT) and blend films (PT2OBT:PC_{70}BM, PVT2OBT:PC_{70}BM and PFBT2OBT:PC_{70}BM)

Steady-state space-charge-limited current (SCLC) mobility

The charge carrier mobilities were calculated using the SCLC model, where the SCLC is described by $J = 9\varepsilon_0\varepsilon_r\mu V^2/8L^3$, where J is the current density, L is the film thickness of the active layer, μ is the hole mobility, ε_r is the relative dielectric constant of the transport medium, ε_0 is the permittivity of free space (8.85×10^{-12} F m^{-1}), V is the internal voltage in the device, and $V = V_{appl} - V_r - V_{bi}$, where V_{appl} is the applied voltage to the device, V_r is the voltage drop due to contact resistance and series resistance between the electrodes, and V_{bi} is the built-in voltage due to the relative work function difference of the two electrodes.

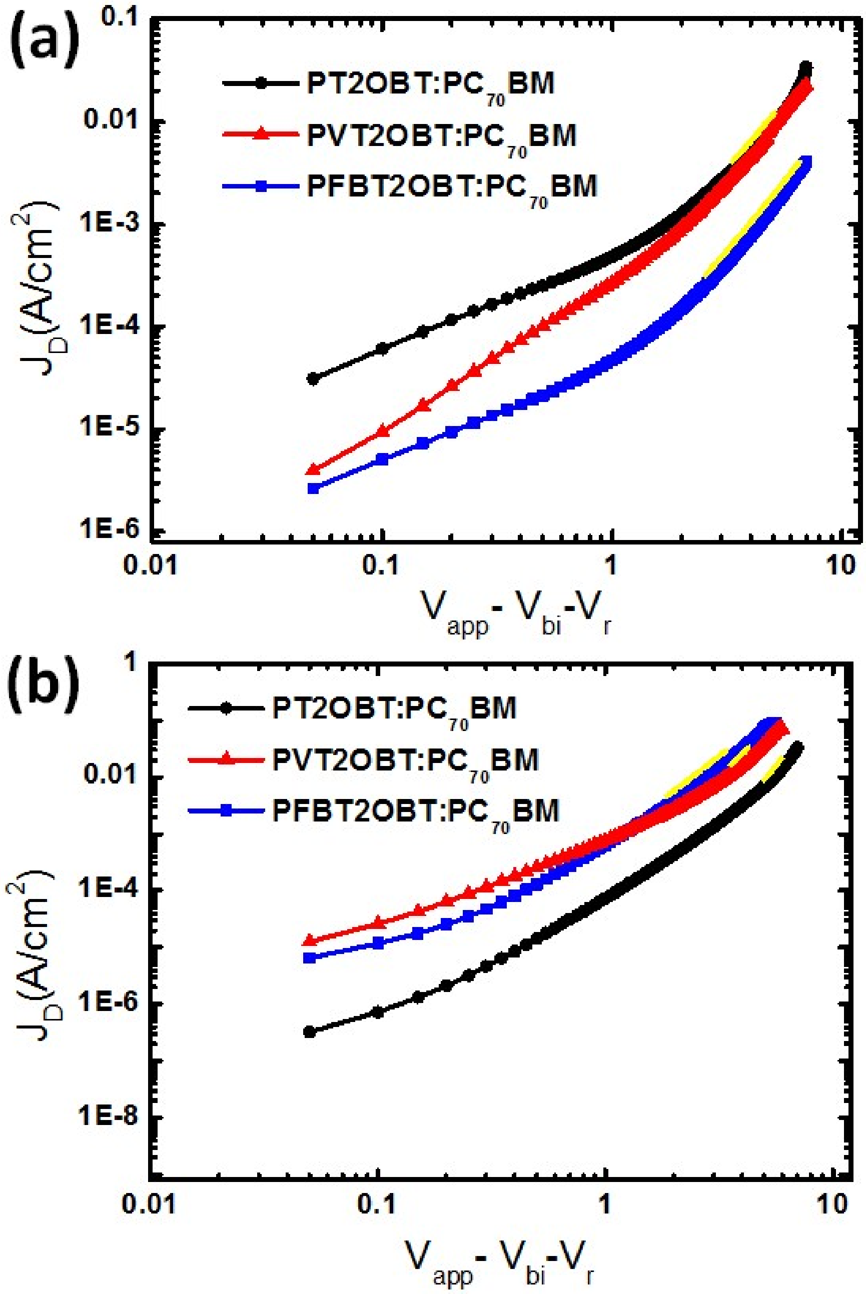


Figure S10. *J–V* characteristics of (a) the hole-only devices with the structure of ITO/PEDOT:PSS/Polymer:$PC_{70}BM$ (1:1.5 w/w in ODCB)/MoO_3/Ag and (b) the electron-only devices with that of ITO/ZnO/Polymer:$PC_{70}BM$ (1:1.5 w/w in ODCB)/LiF/Al. The charge mobilities were calculated by fitting the *J–V* curves in the SCLC regime.

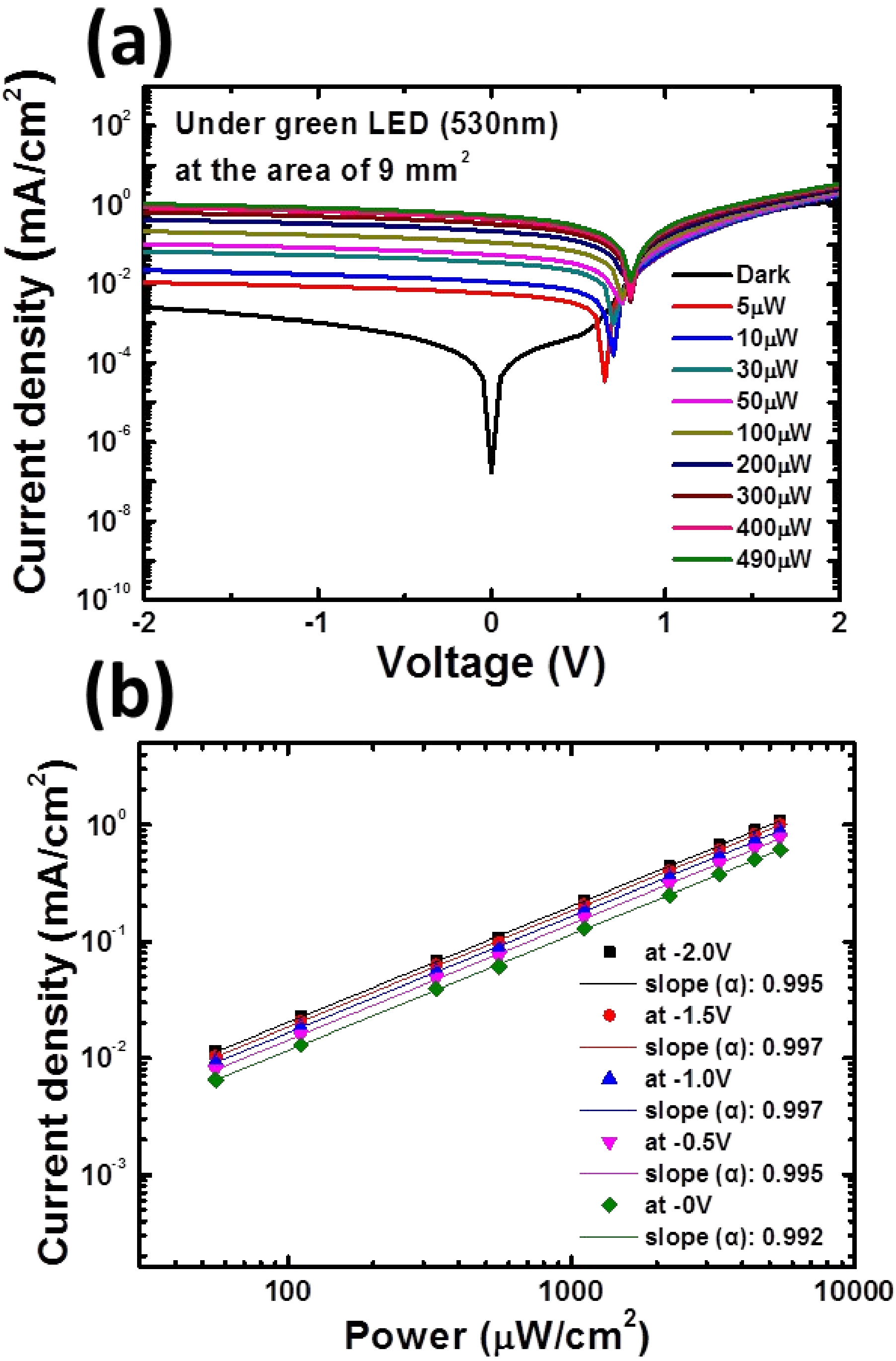


Figure S11. (a) *J-V* characteristices of PT2OBT:$PC_{70}BM$ devices at the active layer thickness of ~180 nm. (b) Current density vs light intensity at different reverse bias (0, -0.5, -1, -1.5 and -2 V).

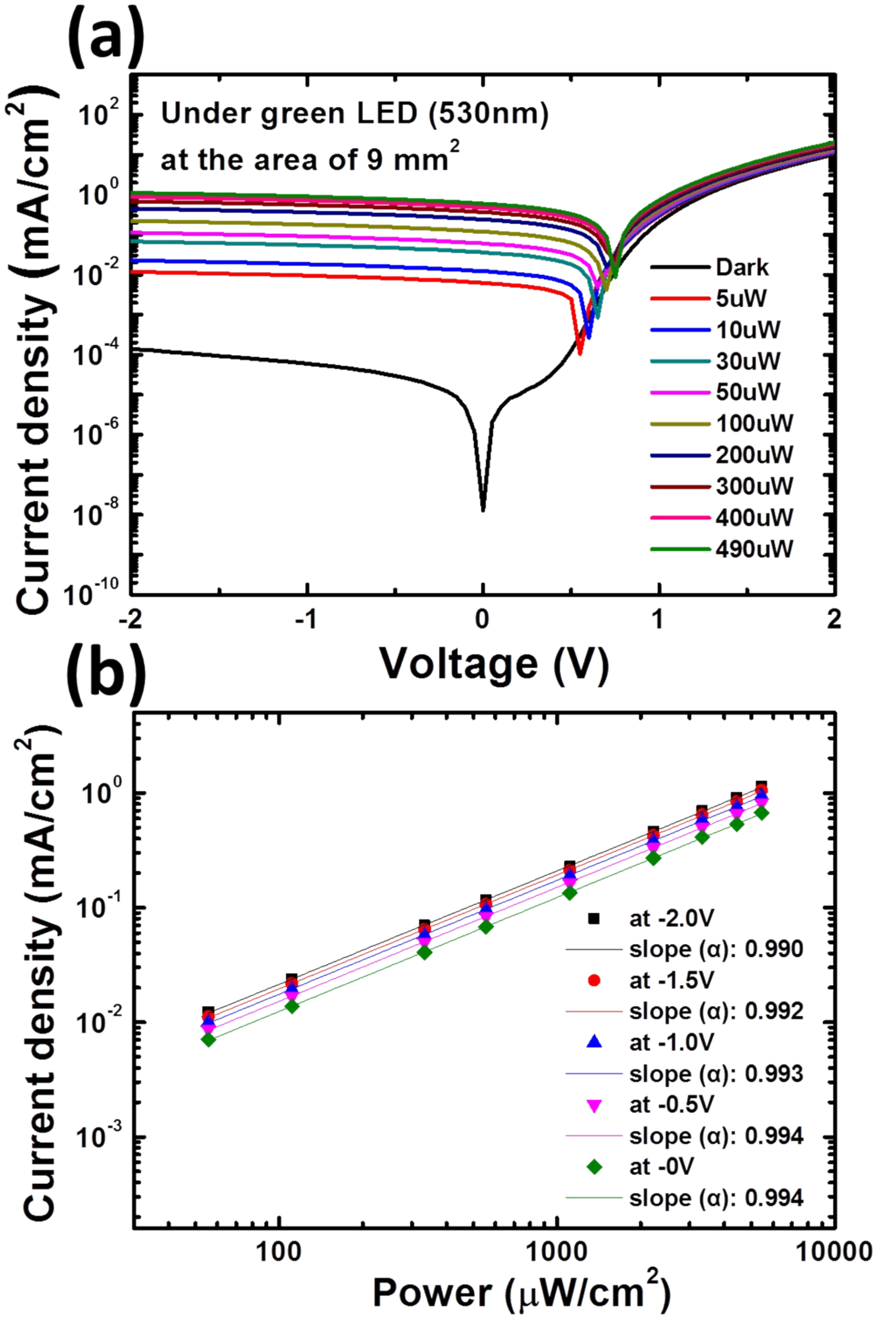


Figure S12. (a) *J-V* characteristices of PVT2OBT:$PC_{70}BM$ devices at the active layer thickness of ~180 nm. (b) Current density vs light intensity at different reverse bias (0, -0.5, -1, -1.5 and -2 V).

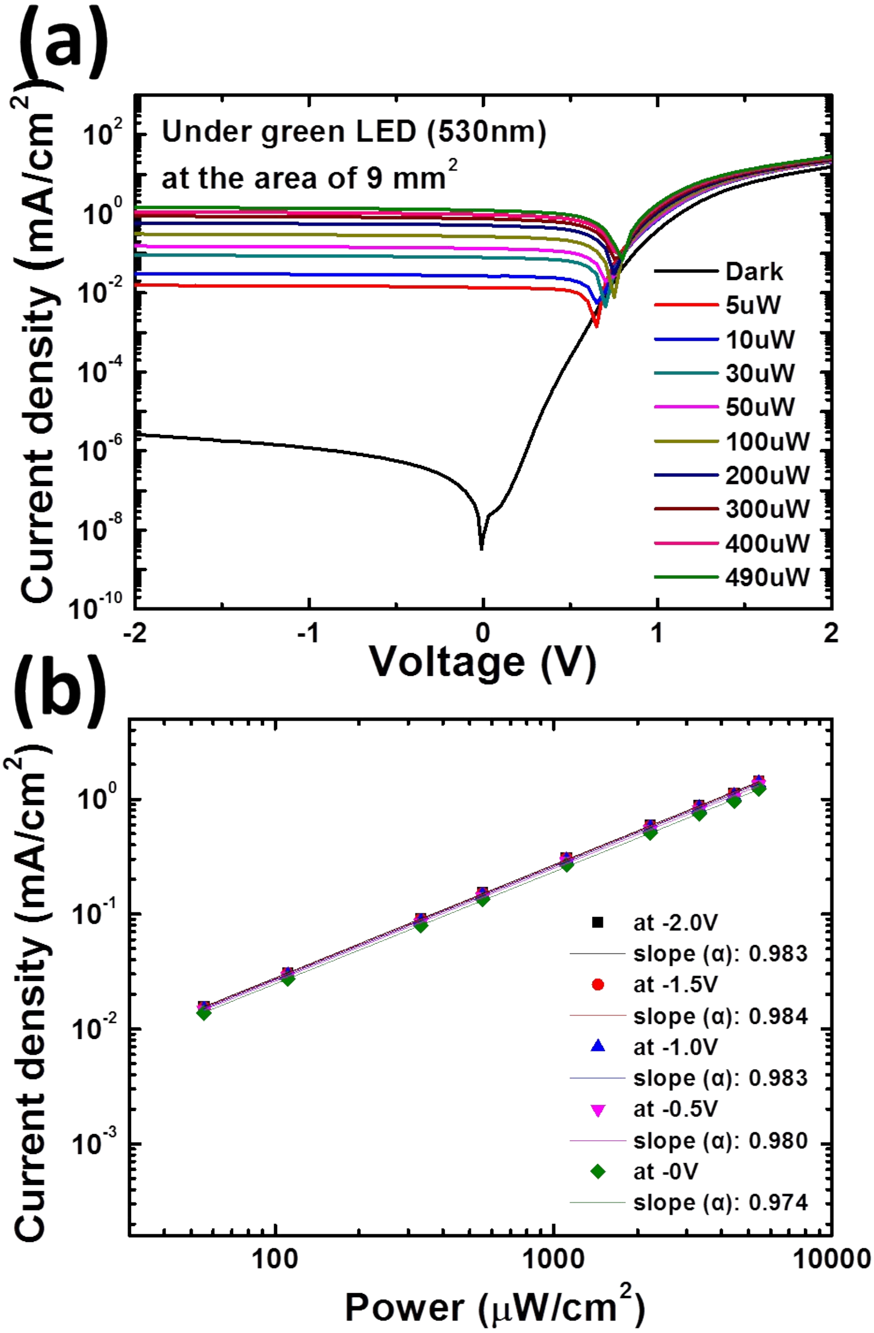


Figure S13. (a) *J-V* characteristices of PFBT2OBT:$PC_{70}BM$ devices at the active layer thickness of ~180 nm. (b) Current density vs light intensity at different reverse bias (0, -0.5, -1, -1.5 and -2 V).

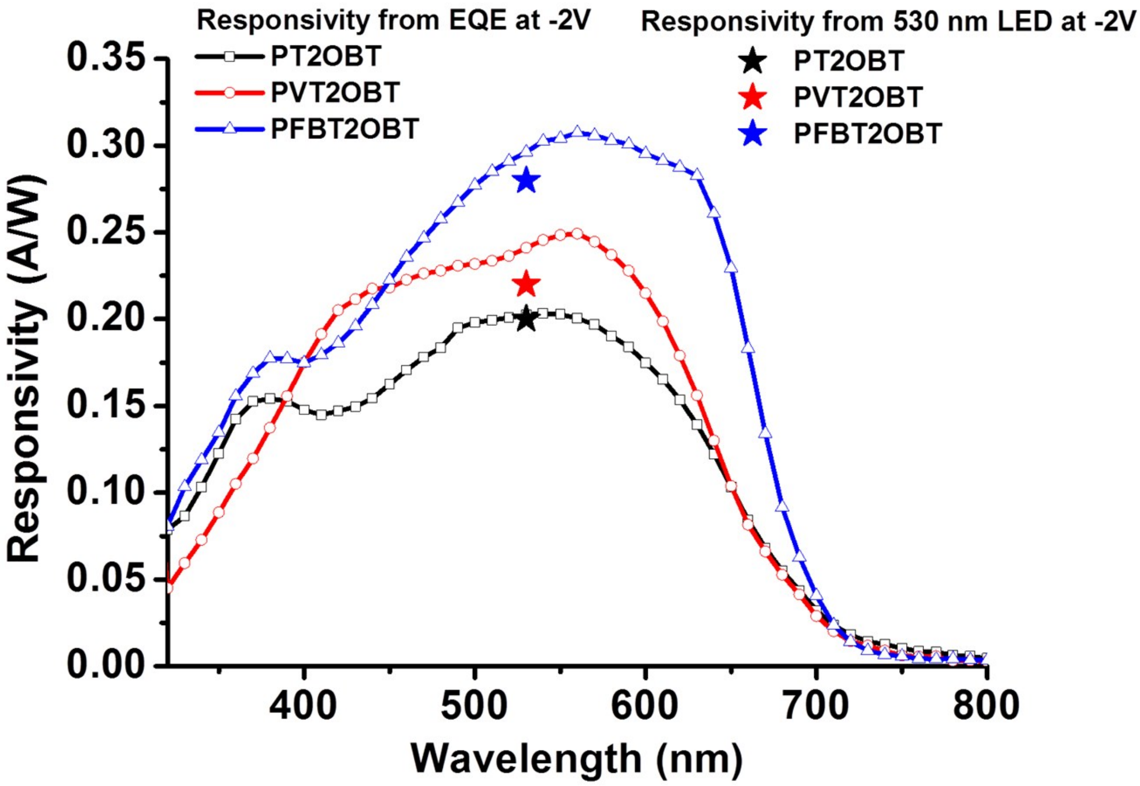


Figure S14. The caculated responsivity of PT2OBT, PVT2OBT and PFBT2OBT from EQE measurement at -2V (line + symbol) and the responsivity from direct measurement under 530 nm LED at -2V (star symbol).