

Supporting Information

A simple medium-bandgap quinoidal A-D-A non-fullerene acceptor for ternary organic solar cells

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Experimental details

Theoretical calculations were carried out within the density functional theory (DFT) framework by using the Gaussian 16, applying density functional theory at the B3LYP level. The basis set of 6-31G* was used in the calculations (Supercomputation Service of UCLM).

The thin film steady state PL spectra of the materials are recorded on the Shimadzu fluorescence spectrophotometer. Time resolved PL spectra of the materials were measured on time resolved photoluminescence system (HORIBA make).

The TRPL spectra of the materials in thin film were recorded on the time-resolved photoluminescence with the time correlated single photon counting (TCSPC) (HORIBA make). The normalization of the absorption spectra and PL spectra of the materials is done by dividing whole spectrum by the maximum absorbance value.

The J-V characteristics under illumination of AM1.5 G (100 mW/cm^2) were measured using Keithley 2400 source meter and the devices were illuminated with AAA solar simulator. The EQE spectra were recorded utilizing the spectral response measurement system (Bentham make).

The indoor J-V measurements were performed under the illumination of device with standard white (SW) and cool white (CW) light emitting diode (1000 Lux) and intensity was confirmed by the luxmeter and converted into $\mu\text{W/cm}^2$ (which is 300.3 and $317.5 \mu\text{W/cm}^2$).

Device fabrication and characterization

The indium tin oxide (ITO) glass substrates were washed three times with deionized water, acetone and isopropanol alternatively in ultrasonic bath. After surface oxygen enrichment with UV-ozone cleaner, PEDOT:PSS was spin coated onto the cleaned ITO substrate at 3500 rpm for 40 s and subsequently, the films were annealed on a hot plate at 120 C for 10 min. A solutions of PBDB-T:Y6 (with different weight ratios, 14 mg/mL in total), PBDB-T:1QDT (with different weight ratios, 14 mg/mL, in total) and PBDB-T:1QDT:Y6 (with different weight ratio between 1QDT and Y6, keeping concentration of PBDB-T constant, 14 mg/mL in total) was stirred at 40 C for 2 hours). Before the spin coating process, 0.25 % 1-CN (v/v) was added to the solutions. The blend solution was spin coated at 3000 rpm for 40 s over the PEDOT:PSS to form an active layer. All the films were thermal annealed at 110 C for 10 min. Subsequently, a thin layer of PFB-Br (0.5 mg/ml in methanol with 0.5 % acetic acid (vol %)) was spin coated onto the active layer at a rate of 3500 rpm for 40 s. Finally, 100 nm Ag was deposited on the films under a pressure less than 10^{-5} Pa , with a shadow mask to maintain the active area of devices (0.04 cm^2).

The hole and electron mobilities of the devices were evaluated from the space charge limited current (SCLC) method with hole only structure of ITO/PEDOT:PSS/blend films/Au and the electron-only structure of ITO/Ag/blend film/Ag, respectively. The corresponding charge mobilities were calculated by fitting the Mott-Gurney square equation $J = (9\epsilon_r\epsilon_0\mu V^2/8L^3)$, where J is the current density, ϵ_r is the dielectric permittivity of the active layer, ϵ_0 is the vacuum permittivity, μ is the hole or electron mobility, and L is the thickness of the active layer.

The values of photocurrent density (J_{ph}) and effective voltage (V_{eff}) are estimated using following expressions:

$J_{ph} = J_L - J_D$, where J_L and J_D are the current density under illumination and in dark, respectively.

$V_{eff} = V_o - V_a$, where V_o is the voltage, when $J_{ph}=0$ and V_a is the applied voltage.

The transient photocurrent and photo-voltage of solar cells over time to analyse the transport mechanisms of photo-generated charge carriers and obtain the Charge Transfer Lifetime (from TPC) and Charge Recombination Lifetime (from TPV) for the better understanding of the charge carrier dynamics of devices. The experimental setup to perform a combined TPV/TPC measurement requires two light sources: one spectrally broad, continuously operated light source for the background illumination and one fast-switching, probing light source with intensities well below the background illumination, a function generator connected to the probing light source can provide the required pulses, while the solar cell should be connected to an oscilloscope at resistance 50 Ω (for TPC) and 1 M Ω (for TPV), which is triggered by the function generator.

Theoretical Calculations

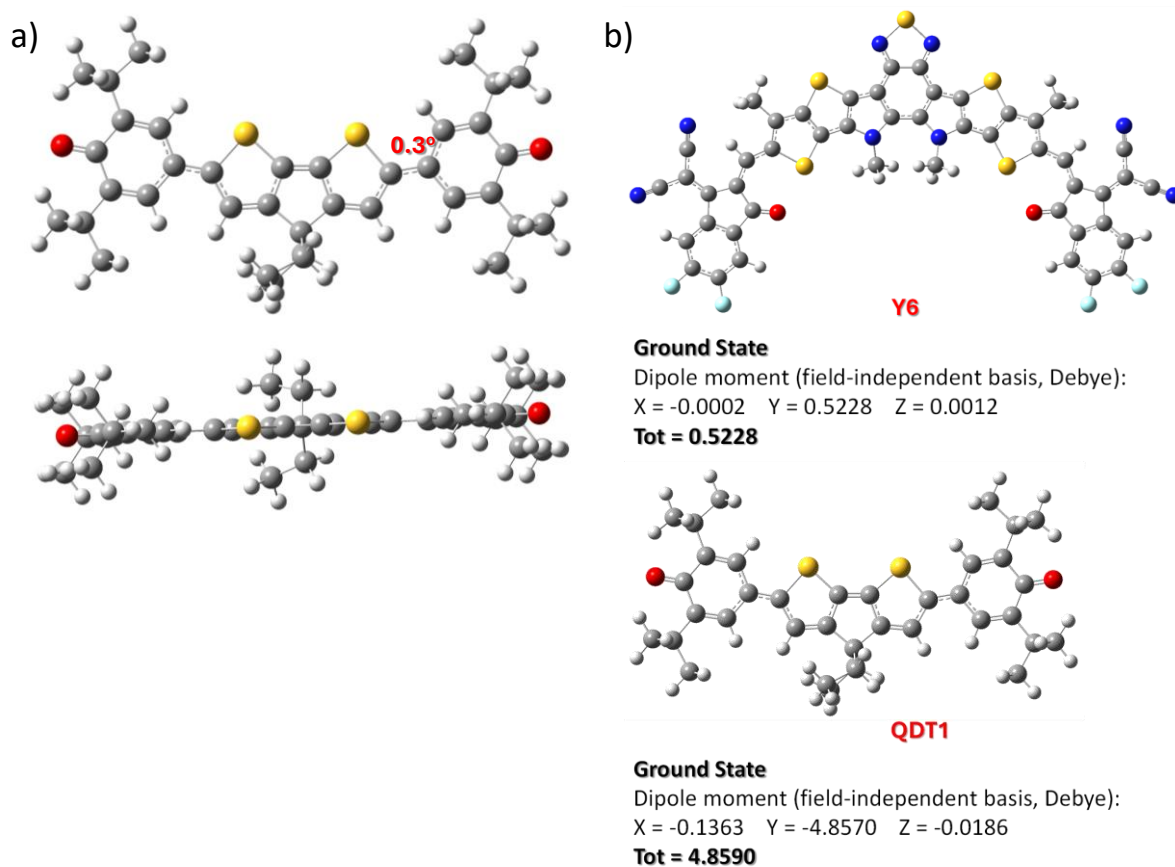


Figure S1. a) Optimized geometry for **QDT1** and b) dipole moments of **Y6** and **QDT1** (Gaussian 09W, DFT-B3LYP, 6-31G*).

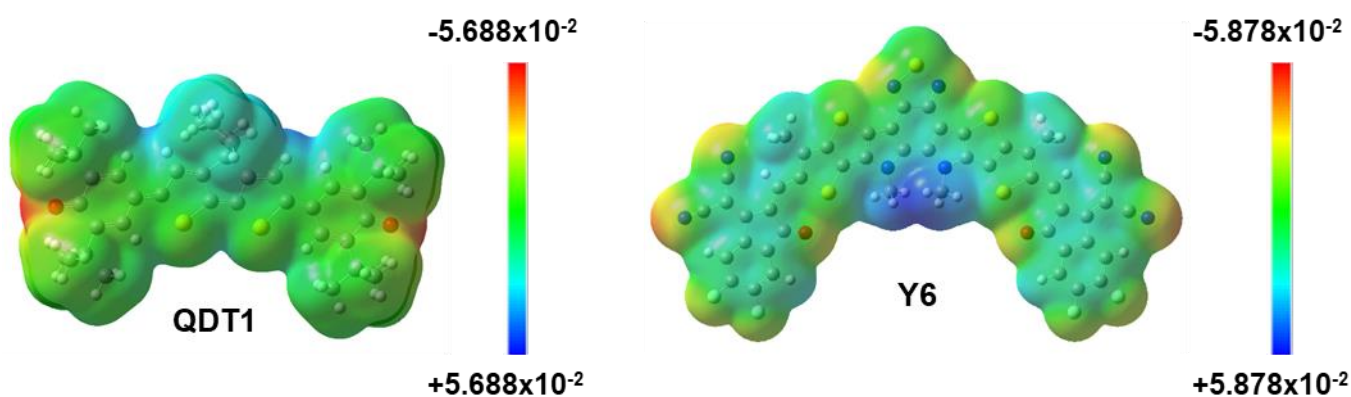


Figure S2. Molecular electrostatic potential maps of **Y6** and **QDT1**. Deepest red colour corresponds to the lowest electrostatic potential energy value in atomic units (hartree) and blue as the highest.

Studies of the active layers.

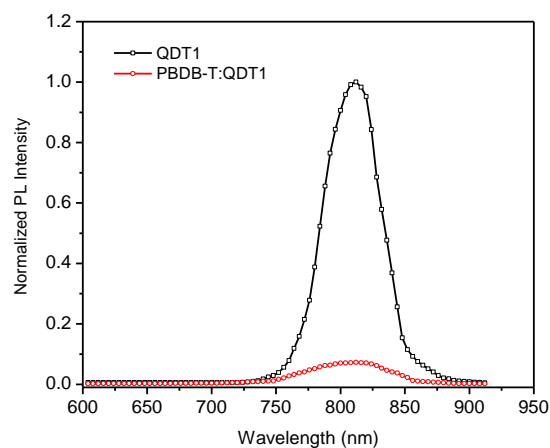


Figure S3. PL spectra of **QDT1** and **PBDB-T:QDT1** (excitation wavelength 690 nm).

Table S1. Photovoltaic parameters of the OSCs based on ternary **PBDB-T:QDT1:Y6** with different weight ratios of **QDT1** and **Y6**, keeping the concentration of **PBDB-T** constant.

PBDB-T:QDT1:Y6 weight ratio	J_{sc} (mA/cm²)	V_{oc} (V)	FF	PCE (%)
1.0:0:1.2	20.78	0.812	0.662	11.17
1.0:0.1:1.1	21.02	0.827	0.678	11.79
1.0:0.2:1.0	22.84	0.839	0.697	13.31
1.0:0.4:0.8	21.42	0.844	0.682	12.33
1.0:0.7:0.5	17.62	0.849	0.645	9.65
1.0:1.2:0	14.36	0.854	0.613	7.52

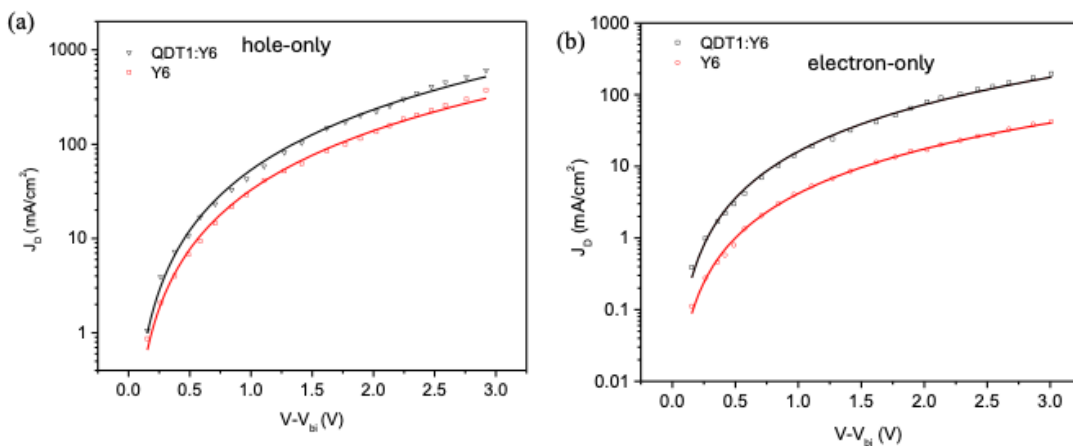


Figure S4. Dark J-V characteristics for: (a) hole only and (b) electron only devices, employing PBDB-T:Y6 and PBDB-T:QDT1:Y6 active layers.

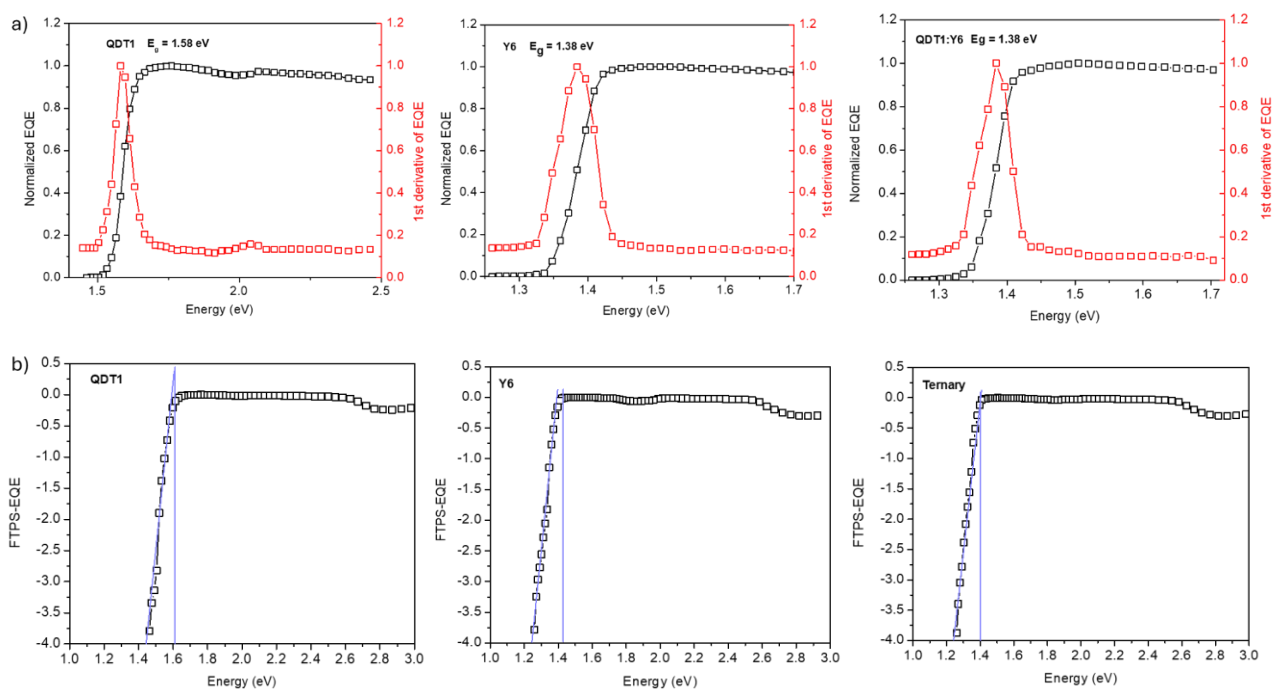


Figure S5. (a) Normalized EQE and 1st derivatives of EQE, and (b) FTPS-EQE spectra of binary and ternary devices.

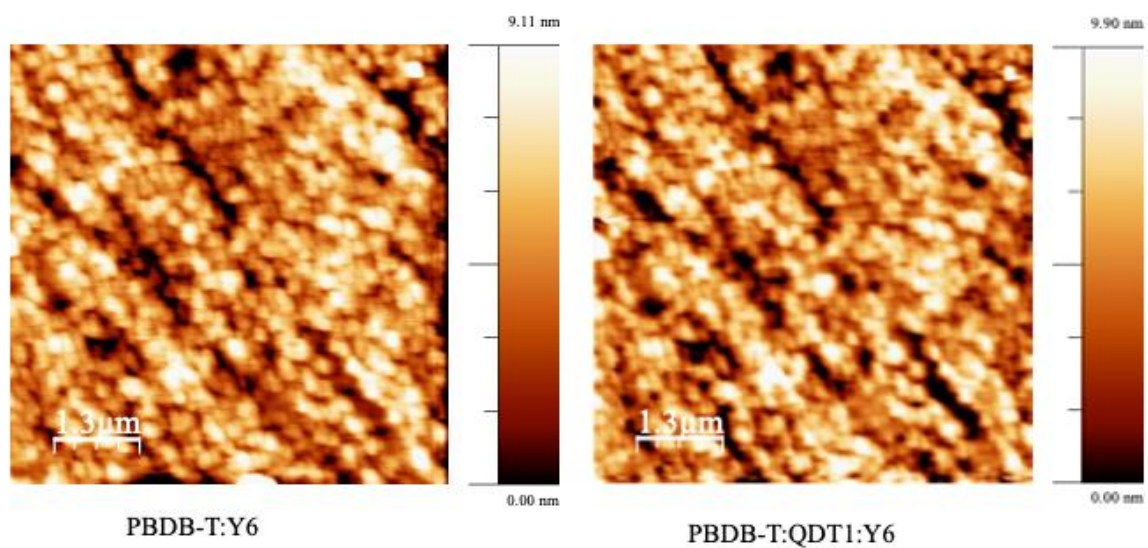


Figure S6. AFM images of binary PBDB-T:Y6 and ternary PBDB-T:QDT1:Y6 films.