# SUPPORTING INFORMATION

## Unexpected hole transfer leads to high efficiency single-walled carbon nanotube hybrid photovoltaic

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**Device fabrication**

SWNTs with diameters in the range of 1-1.5 nm are synthesized by chemical vapor deposition on ST cut quartz (Hoffman Materials) and degenerate doped Si with 300 nm of $SiO_2$ substrates. E-beam evaporated Fe (3 Å) was used as catalysts. Growth of SWNTs were carried out on 1 inch quartz tube furnace using $CH_4$ (1900 sccm) and $H_2$ (300 sccm) at 900 $^oC$ under ambient pressure for 60 minutes. E-beam evaporated 50 nm of Al(Pd) was used as contact electrodes to SWNTs. Furthermore, a 200 nm $SiO_2$ layer was e-beam evaporated over the electrode area to prevent leakage. To form SWNT/P3HT photovoltaic devices, 65 nm thick film of P3HT (American Dye Source, 10 mg/ml in chlorobenzene) was spin-coated (1500 rpm, 60 sec) over the SWNTs.  Following this, a 200 nm ITO contact was RF sputter coated (Enerjet, Lesker) using shadow mask  at RF power 700 W, DC bias -42.5 V, Ar pressure 7 mT and Ar flow rate of 40 sccm. Finally, 300 nm of $SiO_2$ capping layer was e-beam evaporated through a shadow mask on ITO to protect samples against environmental degradation. Impressively, the device characteristics have not changes for more than half a year while all measurements were carried out under ambient conditions.

**Electrical and optical characterizations**

I-V characteristics of all SWNT/P3HT devices were measured using a DAQ (National Instruments) in series with a current pre-amplifier (Ithaco, DL instruments 1211). A source meter (Kiethley 2400) was also used to provide voltage beyond 10 V in the electrostatic gating measurements. Single junction SWNT/P3HT devices were excited using a 532 nm CW laser (Coherent Verdi 10) focused using a 30X objective. Excitation at 800 nm was obtained by the use of a MIRA-900 Ti:sapphire (Coherent) pulse laser system. AM1.5G intensity was obtained using a calibrated solar simulator (Newport 96000 150 W). Spectral resolved photocurrent measurement setup consist of a monochromator (SP 2300 Princeton Instruments) coupled with a 250 W tungsten halogen lamp (Hamatsu). Optical power measurements were carried out using a 1931C power meter (Newport) coupled to a UV enhanced 918UV Si photodetector (Newport).

**Effective external quantum efficiency calculation of the SWNT/P3HT device**

A simple model (Fig. S3) is used to find the ratio between the generated electrons to the incident number of photons within the SWNT/P3HT interface. This ratio, termed as the effective EQE of the system, provides useful insight to the fundamental efficiency of carrier generation at the SWNT/P3HT junctions. As shown in Fig. S3, illumination of the photovoltaic device is taken at normal incidence to the heterojunction coming from the ITO electrode. Zero reflection of light is considered at each interface. Absorbed photons within the top P3HT layer (bulk-P3HT) will recombine before coming to the P3HT/SWNT interface and therefore will not generate photocurrent. Resultant, useful transmitted light power density per wavelength ($P_n(x)$), incident at a cross section within an exciton diffusion length of P3HT ($\Lambda_E$) from the heterojunction is therefore, calculated using Eqn. (1),

$$P_n(D-\Lambda_E) = T(n)P_n(0)e^{-\alpha_n(D-\Lambda_E)} \qquad (1)$$

Where, $P_n(0)$, $T(n)$, $D$, $\Lambda_E$, $\alpha_n$ are taken as incident optical power density per wavelength $n$, ITO transmission at wavelength $n$ (Fig. S4 black line), P3HT thickness (65 nm), P3HT exciton diffusion length (8.5 nm) and P3HT absorption coefficient (Fig. S4 red line), respectively.

In Eqn. (1) an exponential decay of the transmitted light by extinction within the P3HT layer is assumed together with transmission loss at ITO contact. To obtain the optical power from the power density term above, the effective device area is calculated. Effective device area per SWNT is calculated as $2\times\Lambda_E\times L$ assuming a rectangular area of twice the exciton diffusion length by the SWNT channel length ($L$). Therefore, the total effective area is found by scaling the above value with the number of SWNTs within the active layer of the device ($N$) given by ($2\times\Lambda_E\times L\times N$). Hence the optical power per wavelength ($P_n'$) incident within the heterojunction vicinity is calculated as,

$$P'_n = 2\times\wedge_E\times L\times N\times P_n(D-\wedge_E) \qquad (2)$$

Therefore, the effective EQE ($EQE_n$) is calculated using the $P_n'$ and short circuit current density per wavelength ($I_n$) using Eqn. 3.

$$EQE_n = \frac{I_n E_n}{qP'_n} \qquad (3)$$

Where, $q$ and $E_n$ is charge of an electron and the energy per wavelength $n$, respectively. Substituting Eqn. (1) and Eqn. (2) in Eqn. (3) the effective EQE of the SWNT/P3HT device is found as:

$$EQE_n = \frac{I_n E_n}{2q \times \wedge_E \times L \times N \times T(n) P_n(0) e^{-\alpha_n (D - \wedge_E)}} \qquad (4)$$

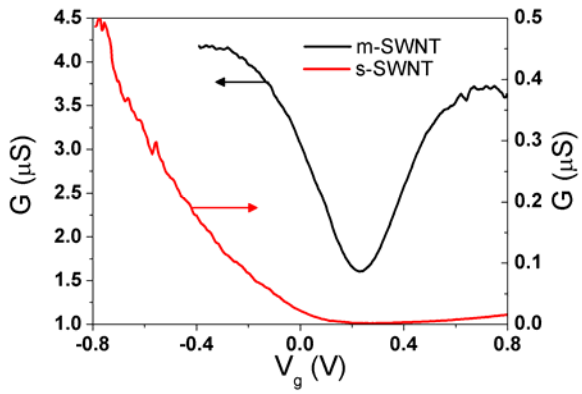


**Fig. S1**. $G$-$V_g$ measurement of individual SWNT on quartz substrate using a solution gate method. The red and black line indicates the $G$-$V_g$ characteristics of a semiconducting SWNT and metallic SWNT respectively. Here, deionized water droplet deposited on exposed SWNT area was used the gate dielectric. A metal tip contacted with the water droplet was used as the gate electrode. $V_g$ was limited to -800 mV to 800 mV to prevent dielectric break down.

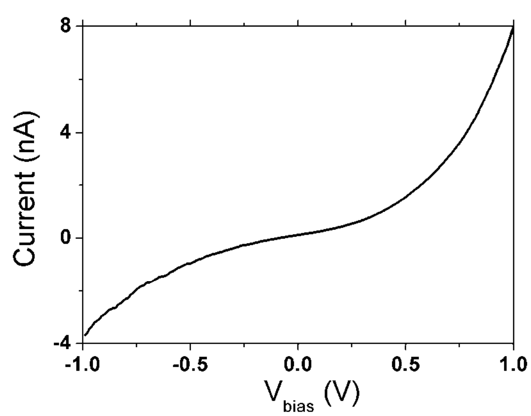


**Fig. S2.** Dark current-voltage measurements of 6-SWNT ensemble Al/SWNT/P3HT/ITO device with bias applied on the Al electrode, and current collected through ITO contact.

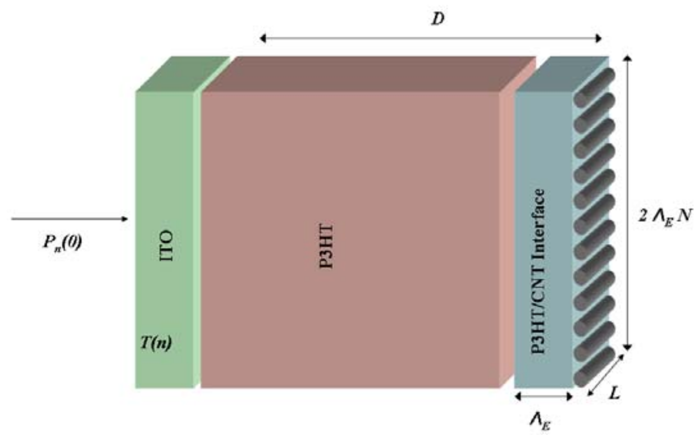


**Fig. S3.** Schematic illustration of the model used for the effective external quantum efficiency calculation of the SWNT/P3HT device. Top ITO contact is given in green. For clarity, the P3HT is shown to consist of bulk-P3HT layer (red) and a thin SWNT/P3HT interface layer (blue) which is one diffusion length ($\Lambda_E$) wide. The terms $\boldsymbol{P_n(0)}$, $\boldsymbol{T(n)}$, $\boldsymbol{D}$, $\boldsymbol{\Lambda_E}$, $\boldsymbol{L}$, $\boldsymbol{N}$ are taken as incident optical power density per wavelength *n,* ITO transmission factor at wavelength *n* (Fig. S4), P3HT thickness (65 nm), P3HT exciton diffusion length (8.5 nm), SWNT channel length (3 µm) and the number of SWNTs within the active area.

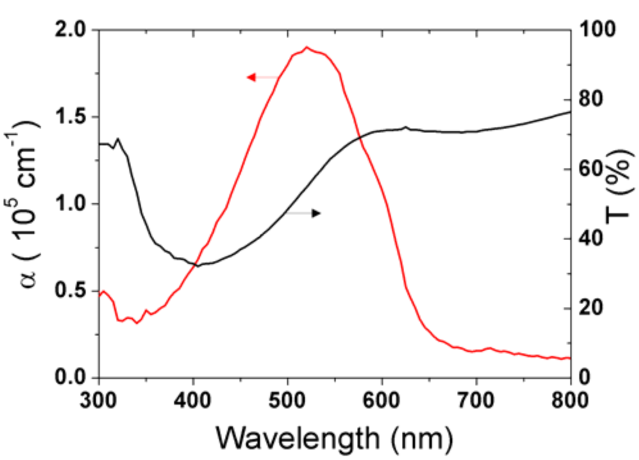


**Fig. S4** The absorption coefficient of a 65nm thin film of P3HT (red curve). Optical transmission of an ITO film (200 nm) deposited by RF sputtering is shown in black.