Supporting Information

Photocurrent Spectroscopy of Dye-Sensitized Carbon Nanotubes

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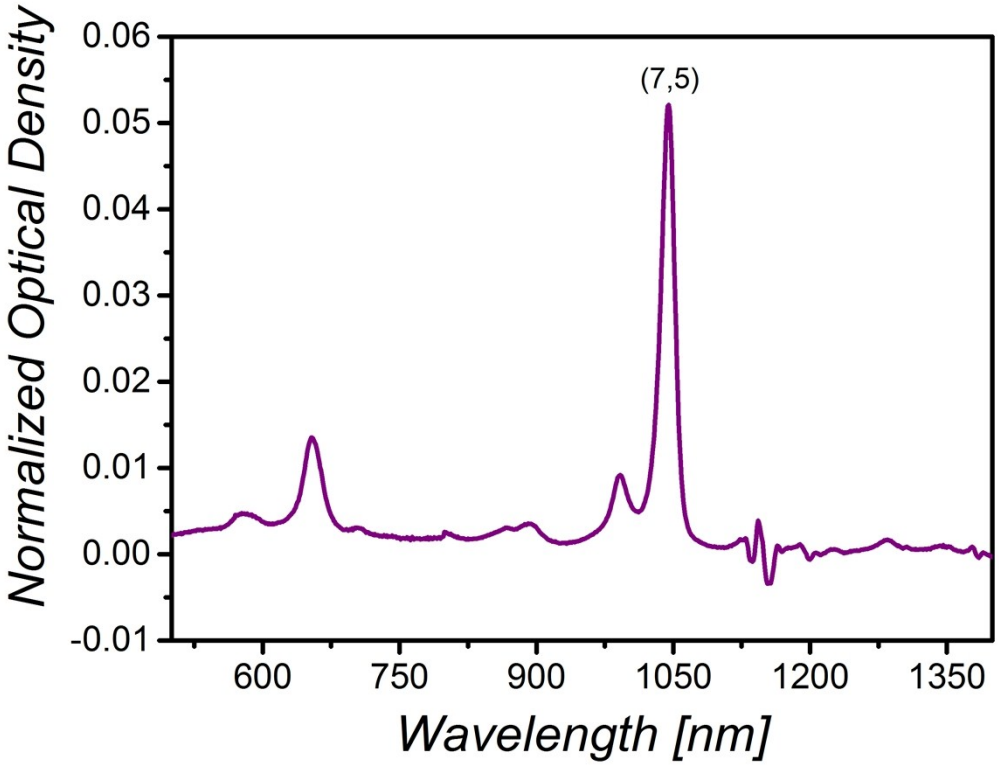


Fig. 1S Solution absorption measurement of PFO wrapped (7,5) carbon nanotubes.

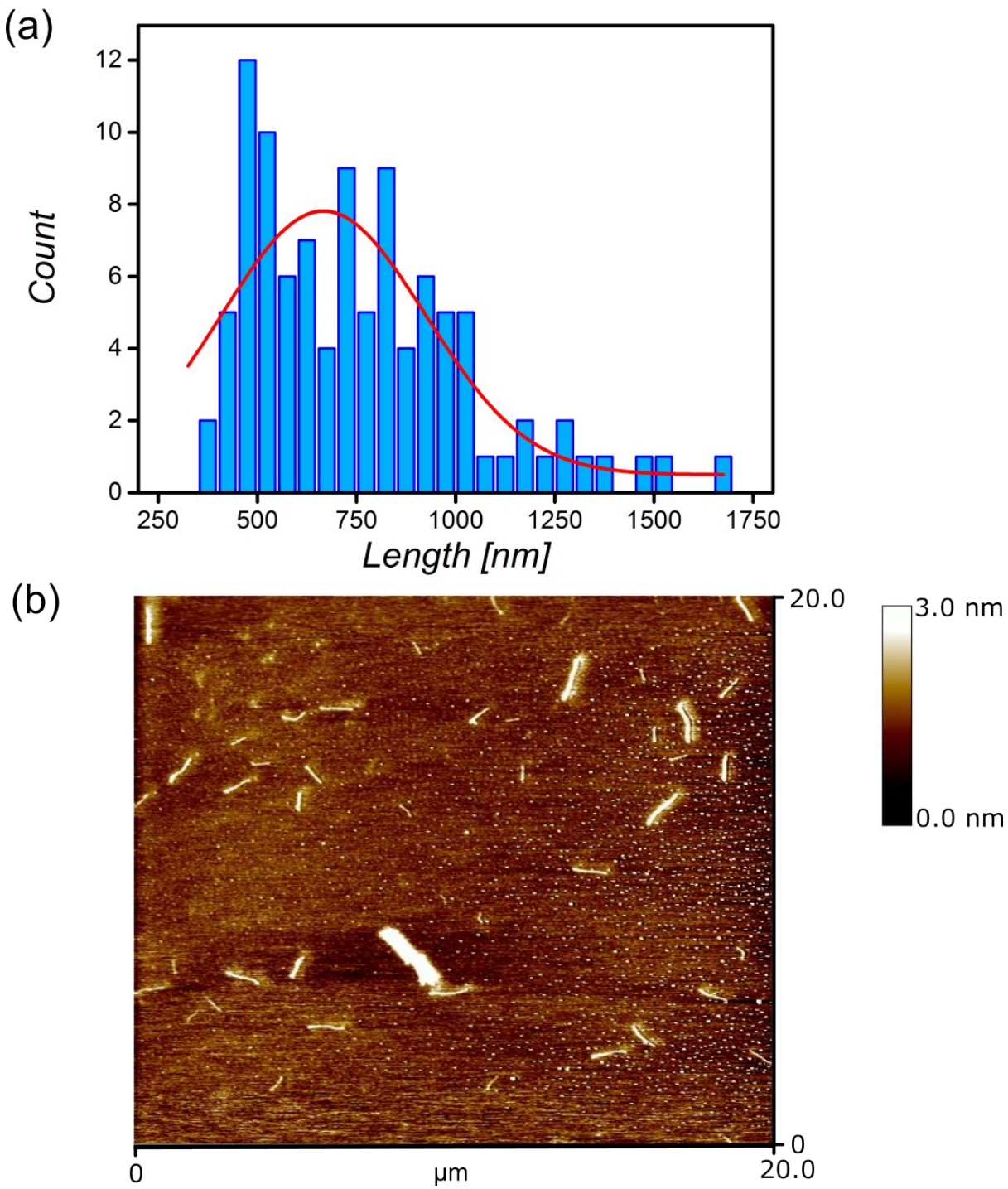


Fig. 2S (a) Histogram of carbon nanotube lengths determined from (b) AFM measurements.

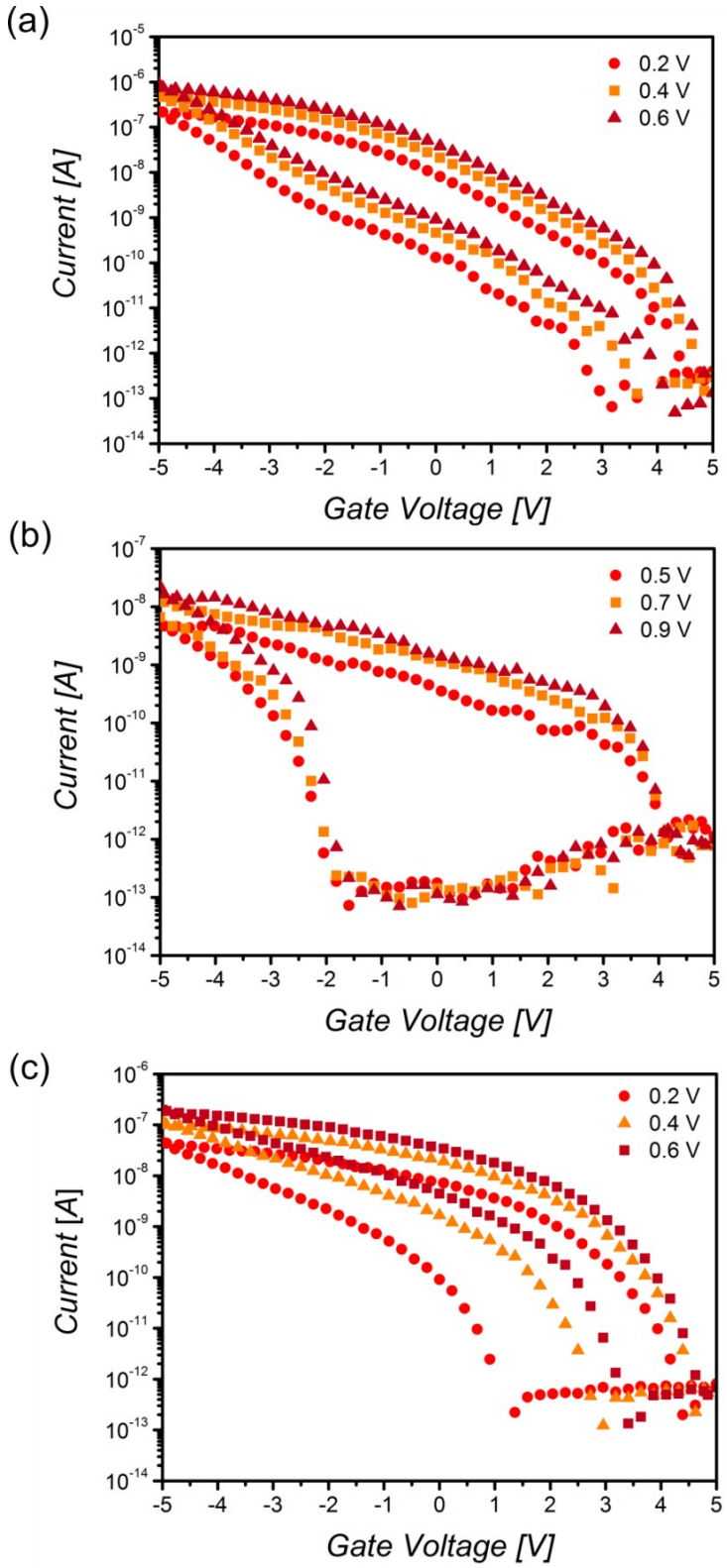


Fig. 3S Transconductance measurements of the devices prior to functionalization with (a) A532, (b) A565 and (c) A610.

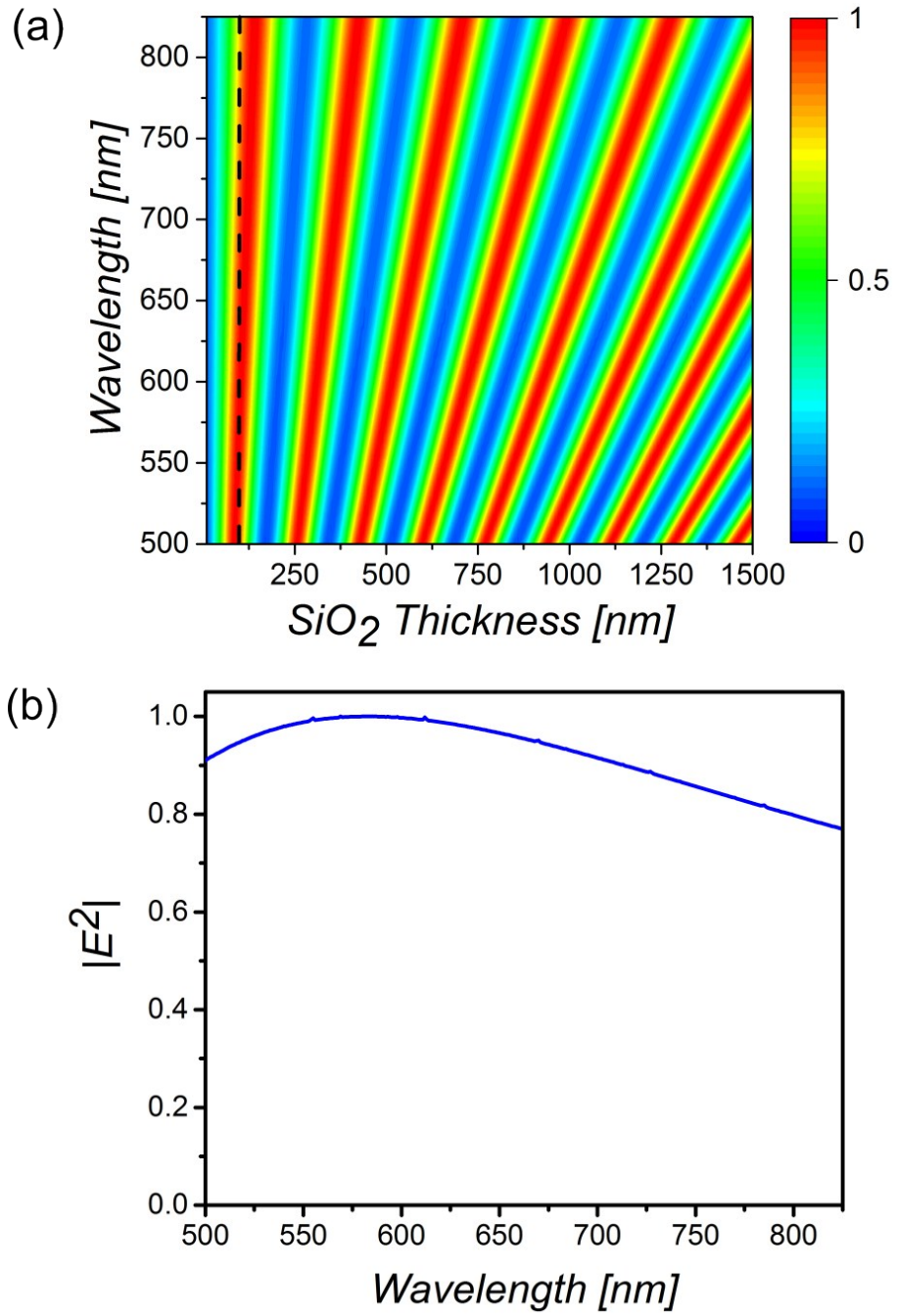


Fig. 4S (a) Normalized electric field intensity of light incident on a SiO_2/Si wafer with variable oxide thickness and (b) a cross section for 100 nm SiO_2 used in this work

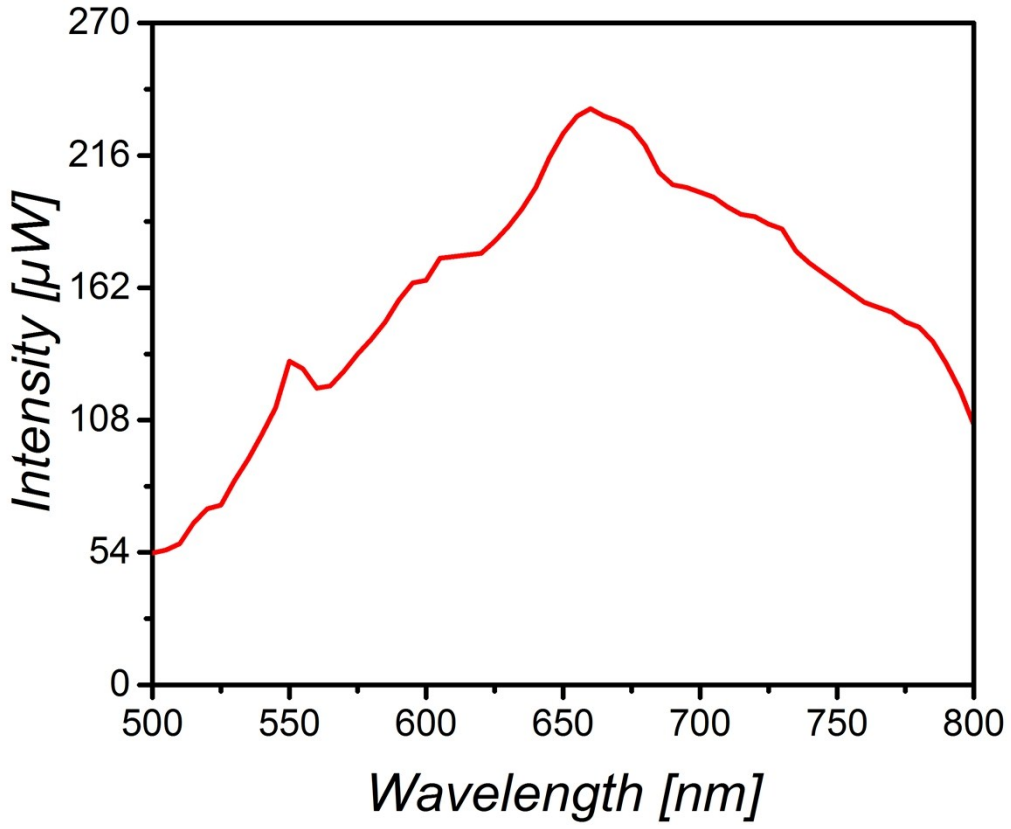


Fig. 5S Power output of the light source measured with a calibrated photodiode sensor (PD300R, Ophir).

Calculations for the generated number of excitons per pulse in a single CNT

S_{22} Wavelength (λ) = 650 nm,
Average power (P_{AVG}) = 2.25×10^{-4} W,
Repetition Rate = 80 MHz,
Pulse Width = 6 ps,
Absorption cross-section per carbon of (7,5) at S_{22} (σ_{22}) = 0.62×10^{-21} m^2/Carbon,
Diameter of (7,5) = 0.82nm, Length of a CNT = 800 nm.

Peak power

$$P_{peak} = \frac{P_{AVG}}{Repetition\ Rate \times pulse\ width} = \frac{2.25 \times 10^{-4}\ W}{(80 \times 10^{6}\ Hz) \times (6 \times 10^{-12} s)} = 0.47\ W.$$

Number of photon per second

$$N_p = \frac{P_{PEAK}}{hc/\lambda} = \frac{0.47\ W}{(6.63 \times 10^{-34}\ Js) \times (3 \times 10^{8}\ ms^{-1}) / (650 \times 10^{-9}\ m)}$$
$$= 1.54 \times 10^{18}\ s^{-1}.$$

Number of carbon atoms per unit length

$$N_c = \frac{4\sqrt{n^2 + nm + m^2}}{3a_{cc}} = \frac{4\sqrt{7^2 + 35 + 5^2}}{3\ \times 0.142} = 98\ nm^{-1}.$$

Total number of carbon atoms in 800 nm $N_c^{'} = 98\ nm^{-1} \times 800\ nm = \ 78400$

Total absorption cross-section $\sigma_{22}^{'} = 0.62 \times 10^{-21} m^2/atom \times 78400\ atom$

$$\sigma_{22}^{'} = 4.86 \times 10^{-17}\ m^2.$$

Area of the laser spot size (A_{SPOT}) =

$$\frac{\pi(1.22\lambda)^2}{4} = \frac{\pi\{1.22 \times (650 \times 10^{-9}\ m)\}^2}{4} = 4.94 \times 10^{-13} m^2.$$

Generated number of excitons per pulse (N_G) $= \left(\frac{N_p \times \sigma_{22}^{'}}{A_{SPOT}}\right) \times pulse\ width$

$$= \left(\frac{(1.54 \times 10^{18}\ s^{-1}) \times (4.86 \times 10^{-17}\ m^2)}{(4.94 \times 10^{-13} m^2)}\right) \times 6 \times 10^{-12}\ s = 909\ excitons.$$

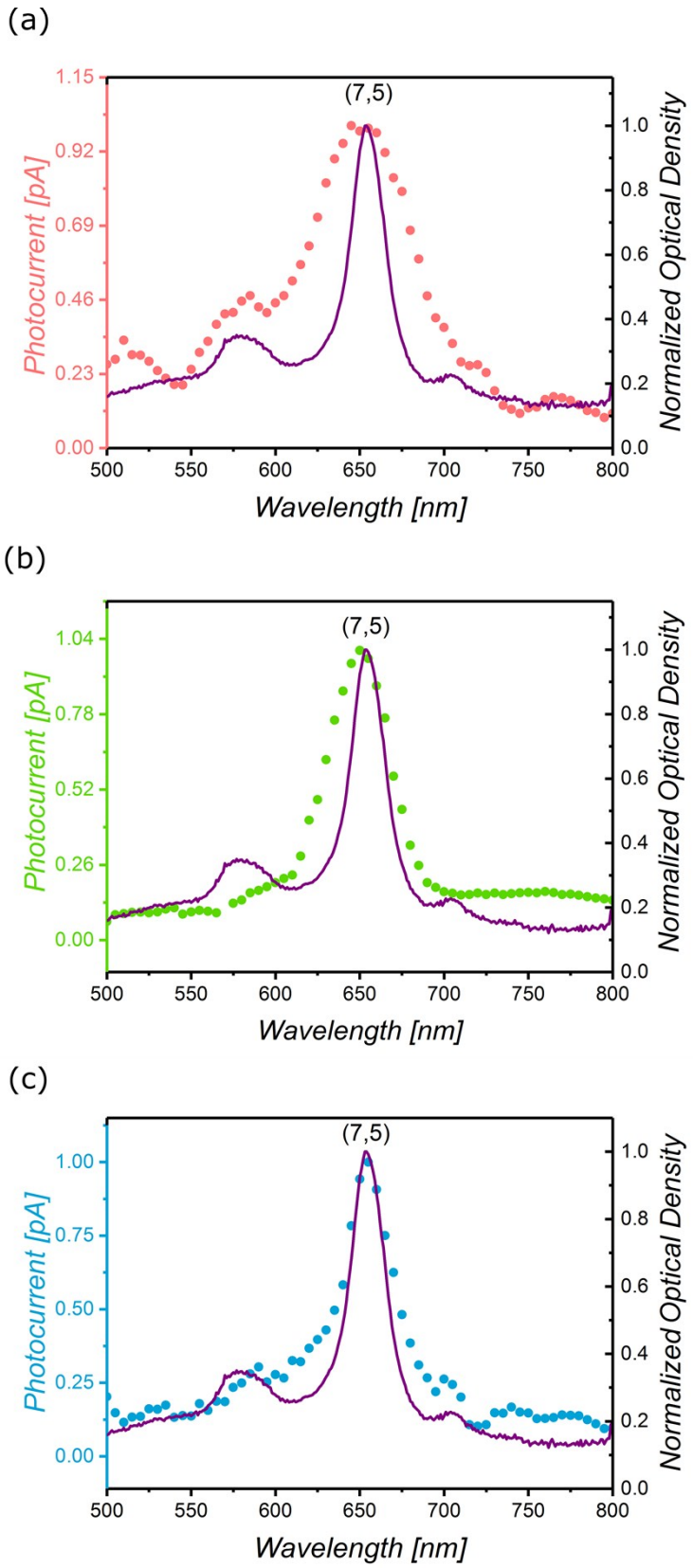
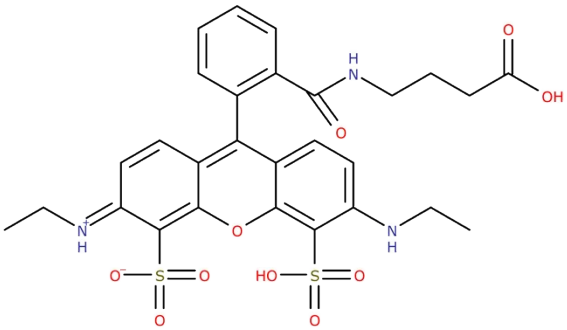


Fig. 6S Photocurrent spectra of the 3 devices shown in Fig. 2 prior to modification with (a) A532, (b) A565 and (c) A610.

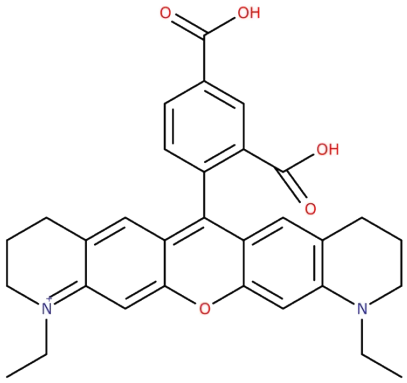
Properties	**ATTO 532**	**ATTO 565**	**ATTO 610**
$Absorption_{max}$ (nm)	532	564	616
$Emission_{max}$ (nm)	552	592	632
Quantum Yield (%)	90	90	70
Absorption Cross Section (cm^2)	4.39×10^{-16}	4.58×10^{-16}	5.73×10^{-16}
Fluorescence Lifetime (ns)	3.8	4	3.2

Table 1S Properties of the ATTO dyes in ethanol solution used in this work.

(a) A532



(b) A565



(c) A610

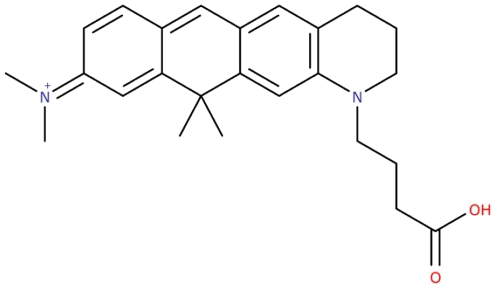


Fig. 7S Chemical structure of the (a) A532, (b) A565, (c) A610 dyes

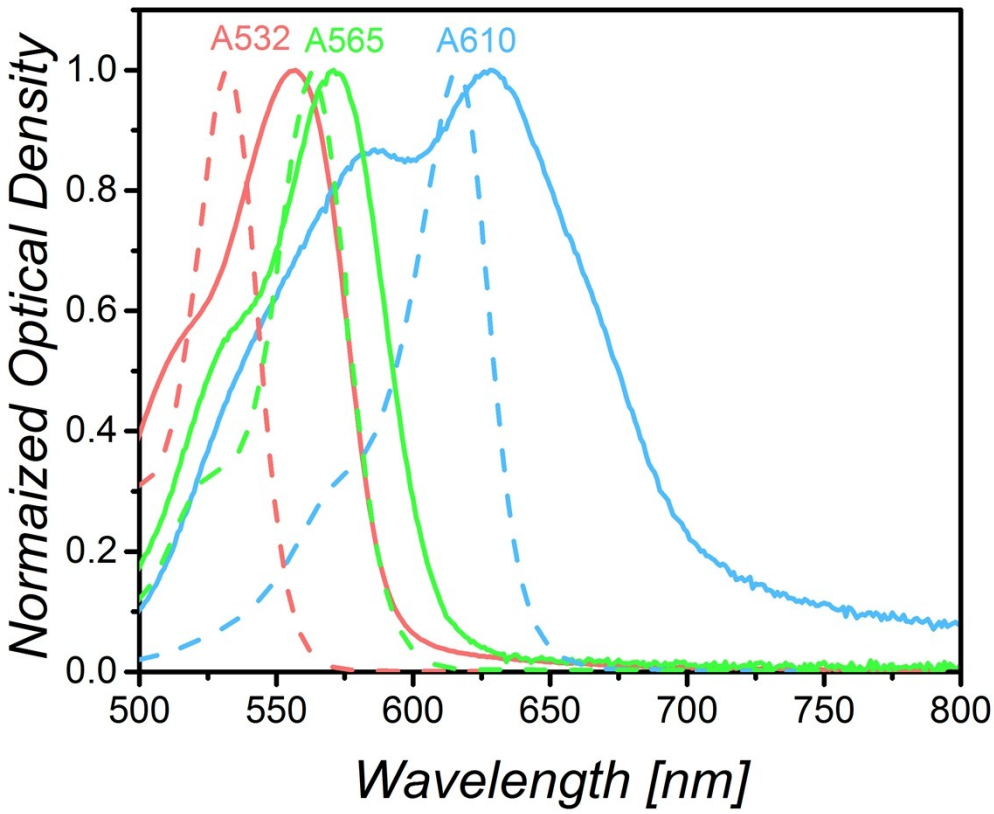


Fig. 8S Absorption measurements of the different ATTO dyes used in this work both (a) in ethanol solution (dotted lines) and (b) as a film on a glass surface (solid lines).

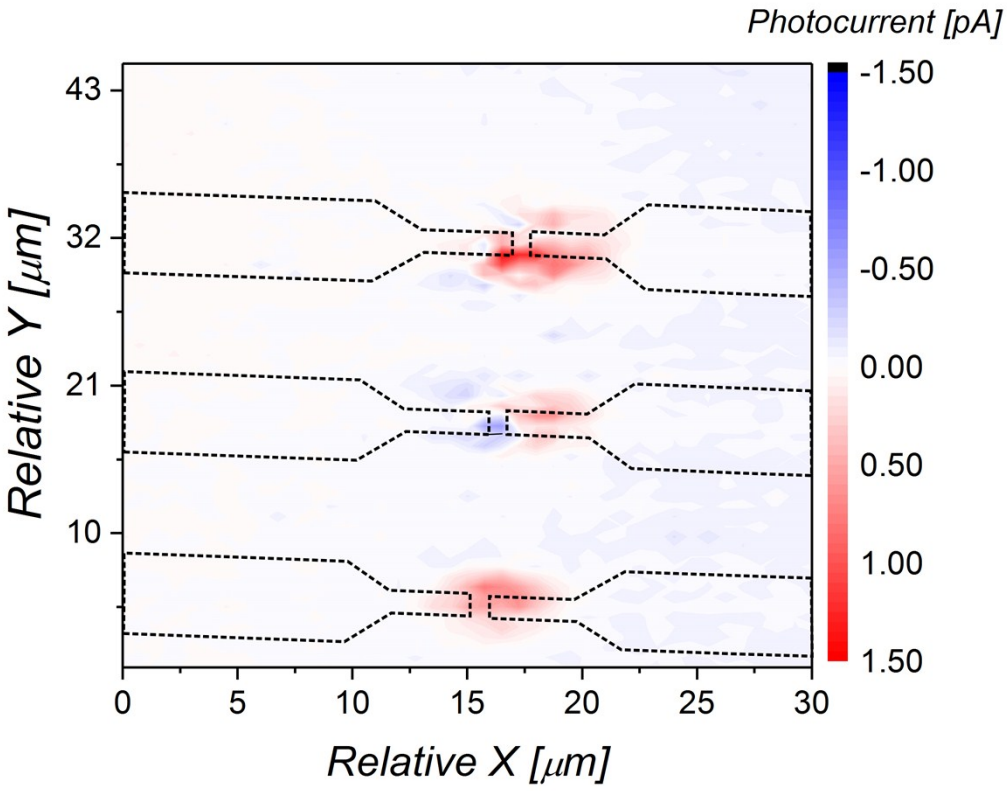


Fig. 9S Photocurrent map of the CNT devices prior to A565 modification as shown in Fig. 1d.

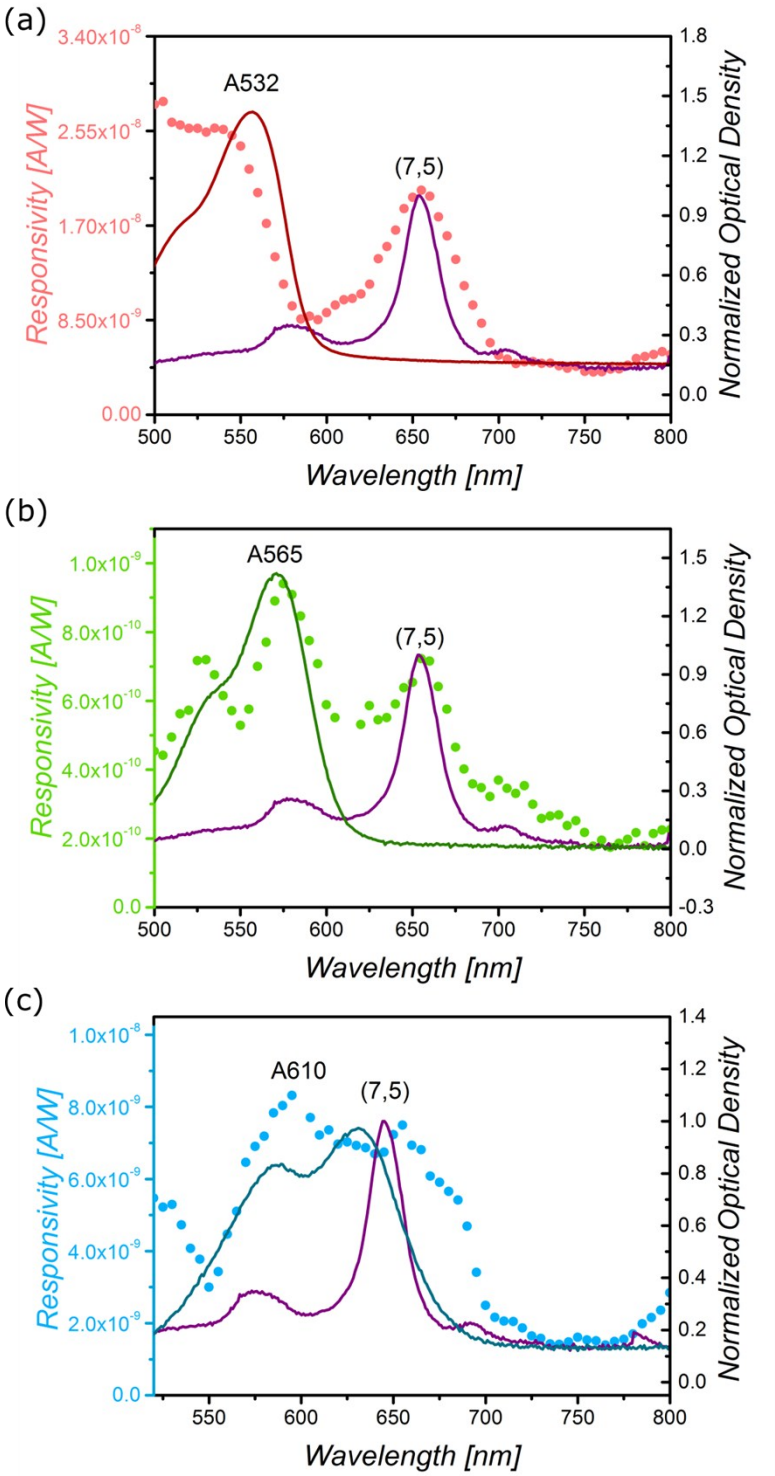


Fig. 10S Responsivity spectra of devices functionalized with (a) A532, (c) A565 and (e) A610 dyes. The photocurrent spectra (dotted) are correlated with the absorption spectra of the dyes (same color) and the (7,5) dispersion (Purple).

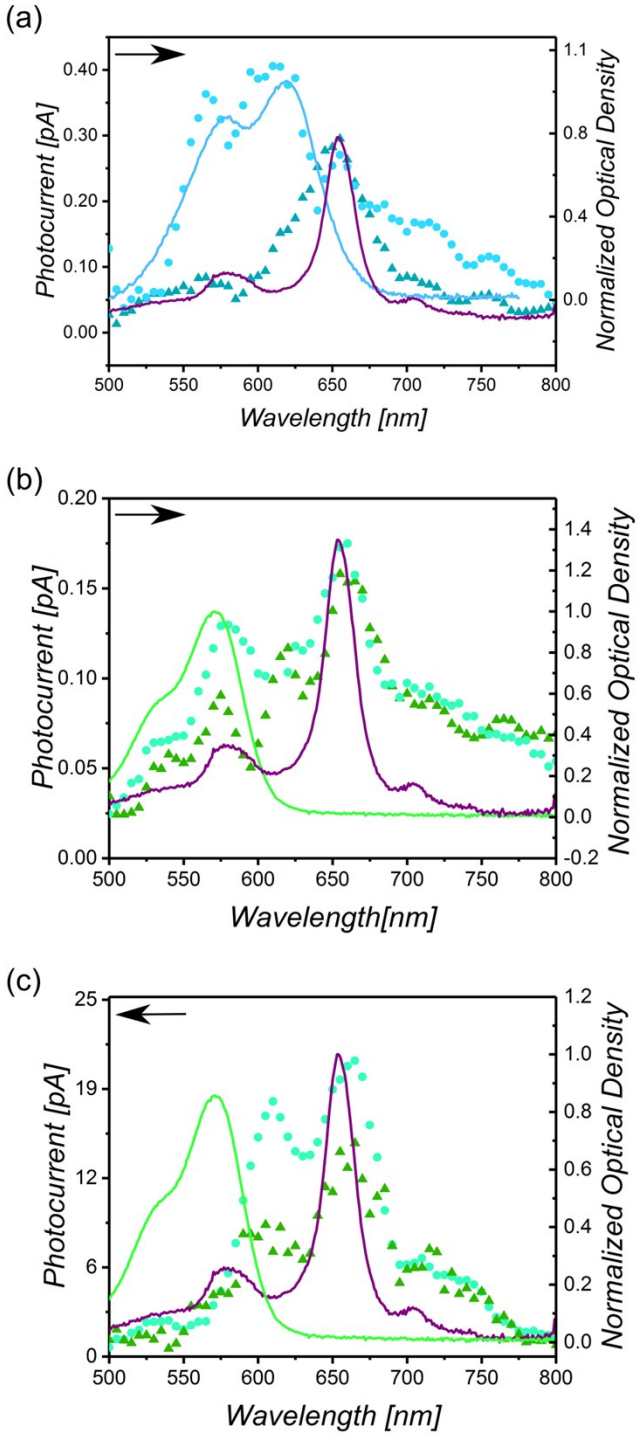


Fig. 11S Repeated photocurrent measurements of a device modified with (a) 40 µL of A610 and ((b) and (c)) 20 µL of A565. The scan direction has been indicated with arrows in the top left. For all cases, the subsequent spectrum (triangles) shows lower or almost no contribution from the dye in comparison to the initial measurement (circles).

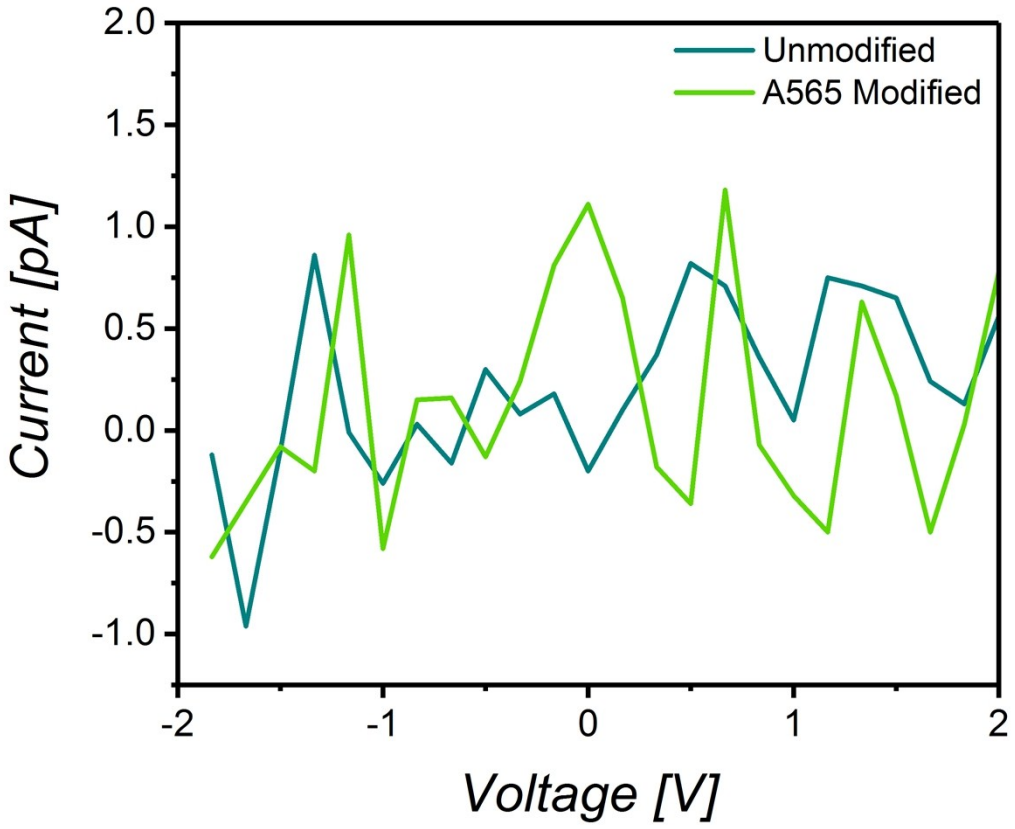


Fig. 12S I-V characteristics at 0 V gate bias and 0.5 mV source – drain voltage of a device before and after modification with 50 µL of A565.

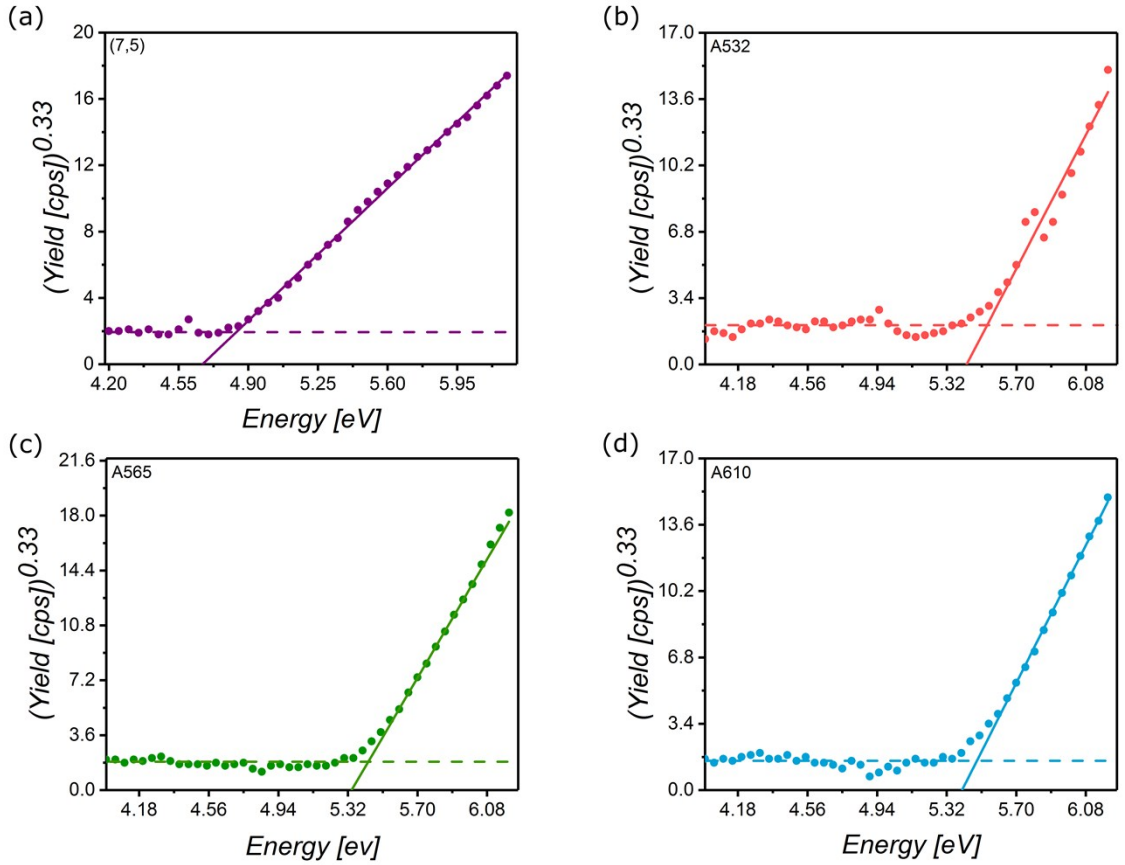


Fig. 13S PESA measurements of (a) (7,5) and the (b) A532 (c) A565 (d) A610 dyes cast onto a glass substrate.

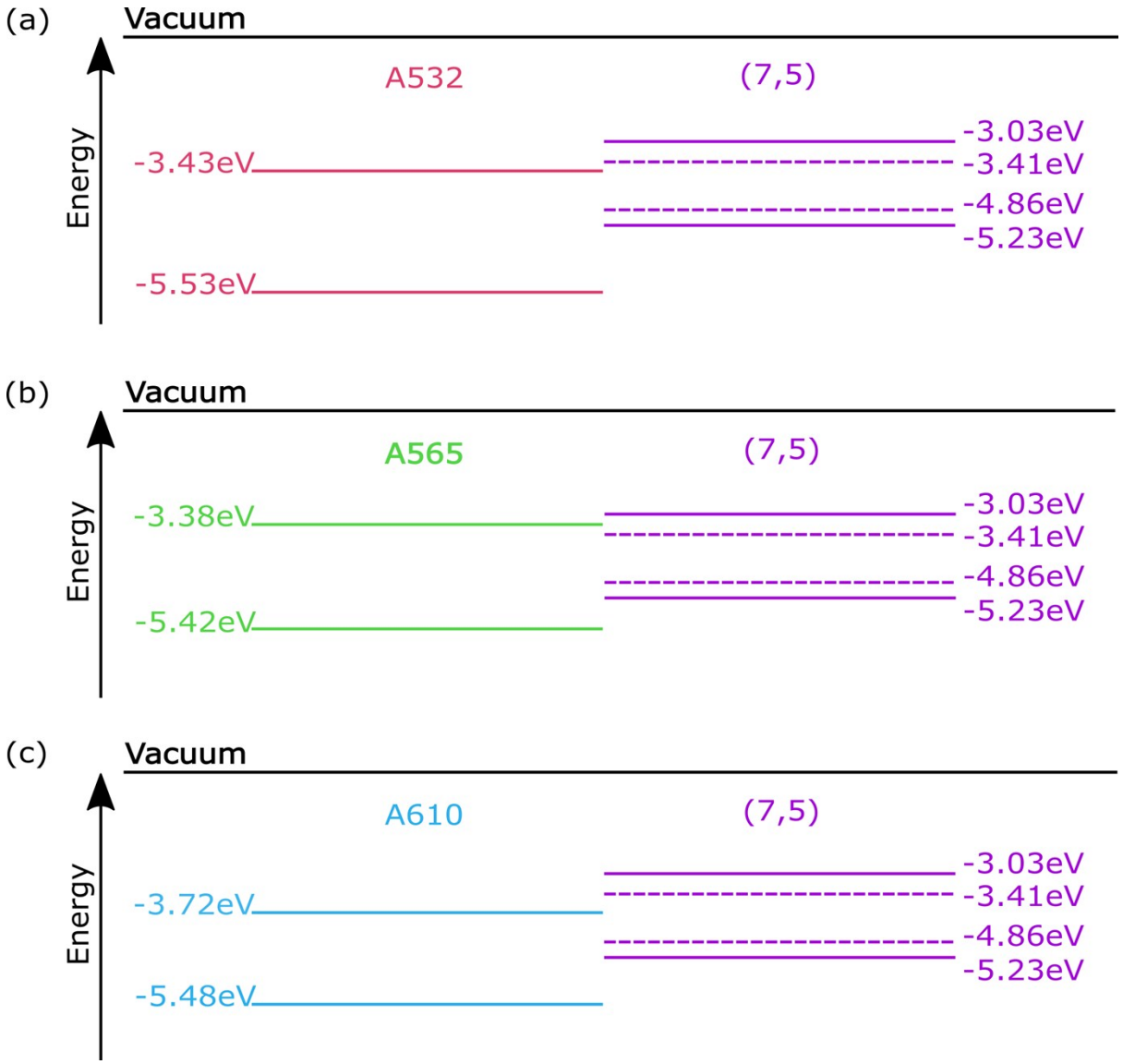


Fig. 14S Energy level diagram based on the HOMO level determined from PESA measurements shown in Fig. S12. The dashed lines represent the S_{11} of the (7,5). All three dyes form a Type II heterojunction with the (7,5).

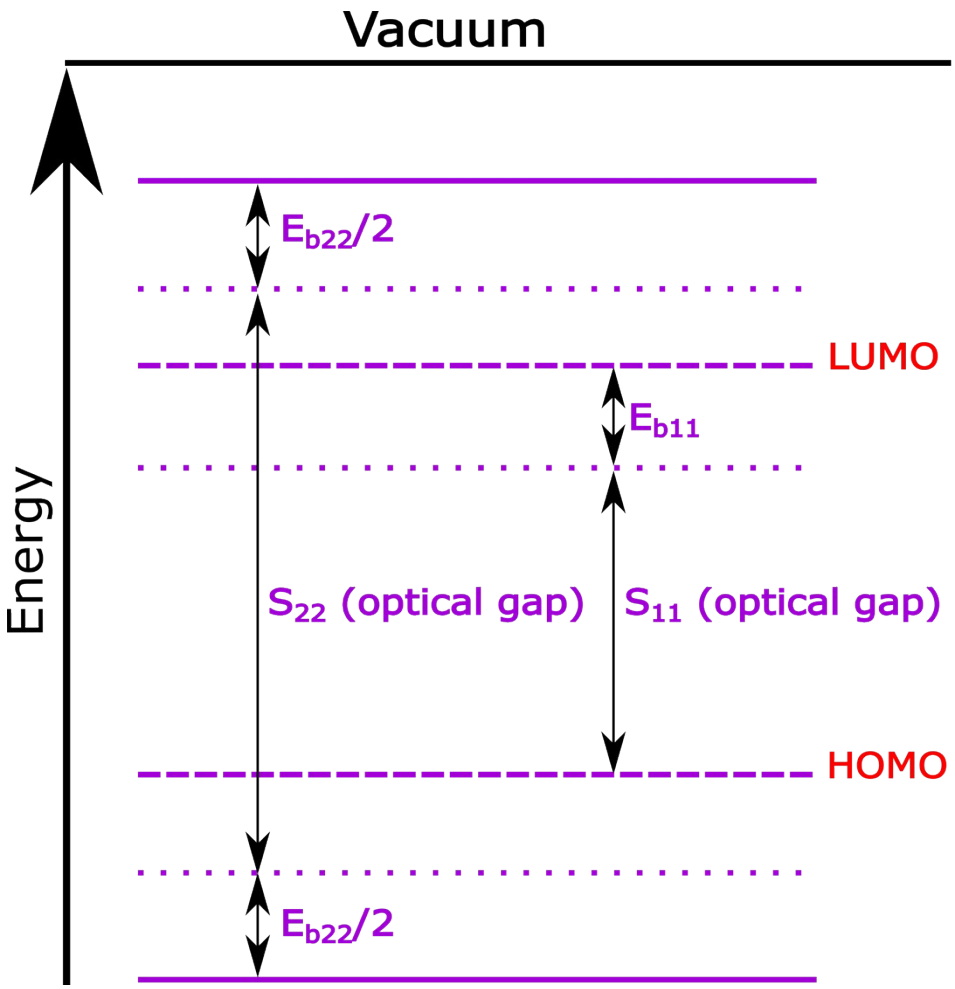


Fig. 15S Energy level diagram showing the HOMO, LUMO and S_{22} energy bands. The estimated free carrier levels after adding the binding energies (E_{b11} and E_{b22}) of the S_{11} (dashed) and S_{22} (continuous) are depicted in Fig. 13S.

Dye Coverage Estimation

Radius of (7,5) = 0.41nm, Length of a CNT = 800 nm, dye interlayer spacing = 0.35 nm,

Surface Area of a single A565 dye = 1.03 nm^2, CNT population =P_{cnt}

Surface coverage of the n^{th} layer of A565 dye $= \frac{P_{cnt} \times 2 \times \pi \times 800 \times (0.41 + 0.35(n-1))}{1.03}$

Total dye coverage $= \sum_{i=1}^{n} \frac{P_{cnt} \times 2 \times \pi \times 800 \times (0.41 + 0.35(n-1))}{1.03}$ molecules

For a single nanotube (P_{cnt} = 1), total dye coverage = 6.38×10^5 molecules, when n = 38.
Therefore, layer thickness = (37 × 0.35) + 0.41 = 13.36 nm

For 10 nanotubes (P_{cnt} = 10), total dye coverage = 6.8×10^5 molecules, when n = 12.
Therefore, layer thickness = (11 × 0.35) + 0.41 = 4.26 nm

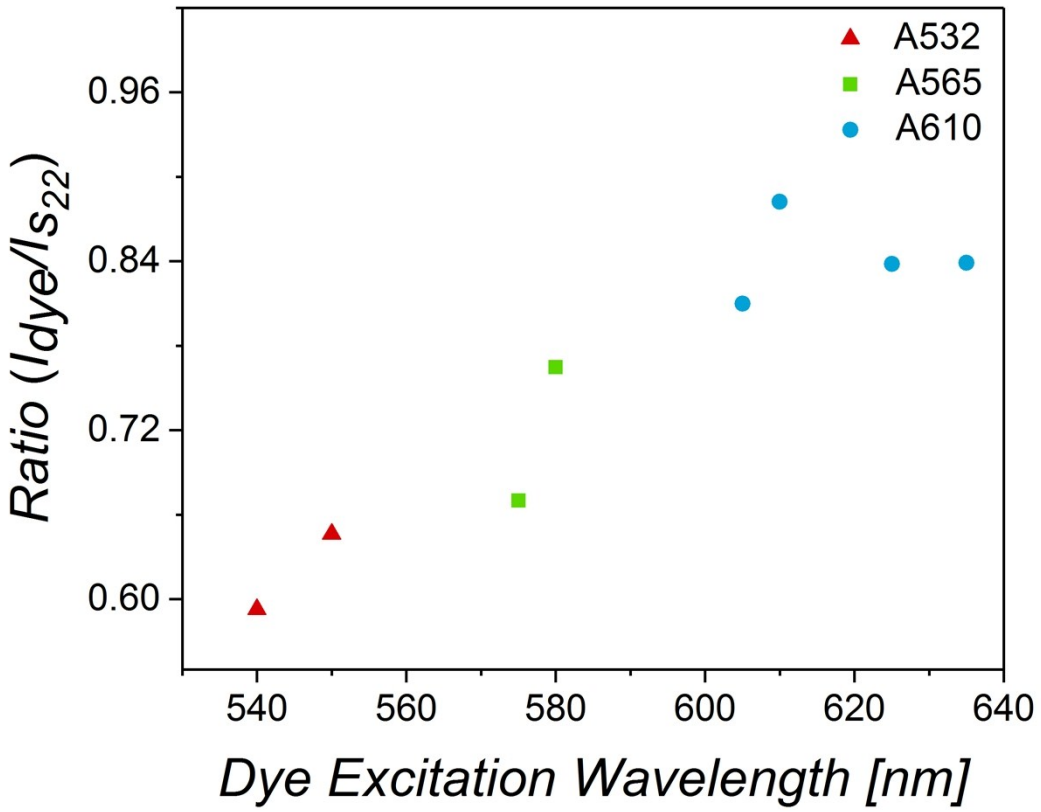


Fig. 16S Photocurrent intensity at the excitation wavelength of the dyes compare to S_{22} of (7,5).