

Supporting information:

A Metrological Investigation of (6,5) Carbon Nanotube Absorption Cross-Section

*Laura Oudjedi^{1,2}, A. Nicholas G. Parra-Vasquez^{1,2}, Antoine G. Godin^{1,2}, Laurent Cognet^{1,2}, and
Brahim Lounis^{1,2,*}*

¹ Univ Bordeaux, LP2N, F-33405 Talence, France

² CNRS & Institut d'Optique, LP2N, F-33405 Talence, France

Laser beam profile determination:

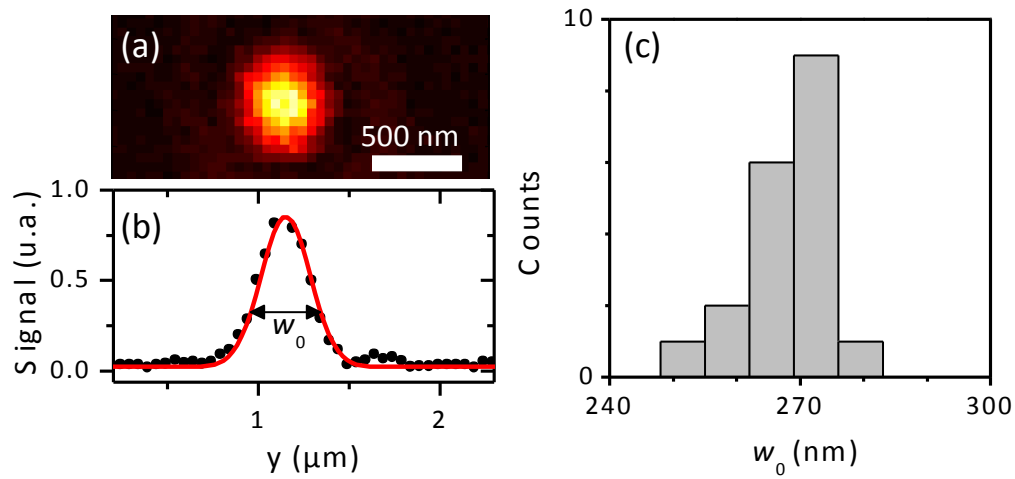


Figure 1S: (a) The beam profile is determined by acquiring fluorescence confocal images of individual 20 nm luminescent spheres. (b) By fitting the profile with a Gaussian function we measure the beam waist (w_0). (c) From 45 fluorescent spheres we obtain $w_0 = 269 \pm 7$ nm.

Beam displacement determination:

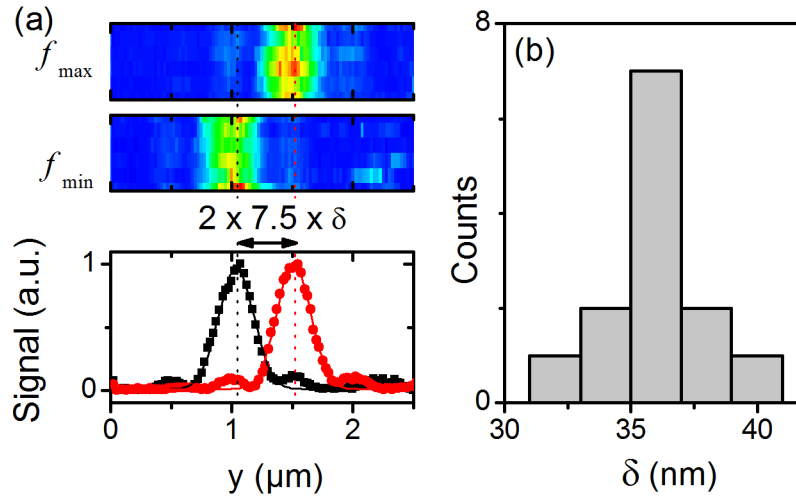


Figure 2S: Calibration of the beam displacement. (a) Two images of the same semiconducting nanocrystal are acquired for two different RF frequency driving the acousto-optic modulator (AOM). The frequency difference is chosen such as the beam displacement in (a) is 15 times δ , the spatial modulation amplitude used in the absorption measurements. Both images profiles fit to Gaussian functions. (b) The measured values of δ from 13 acquisition are gathered in this histogram and $\delta = 36.4 \pm 1.9$ nm.

Selection of isolated (6,5) nanotubes:

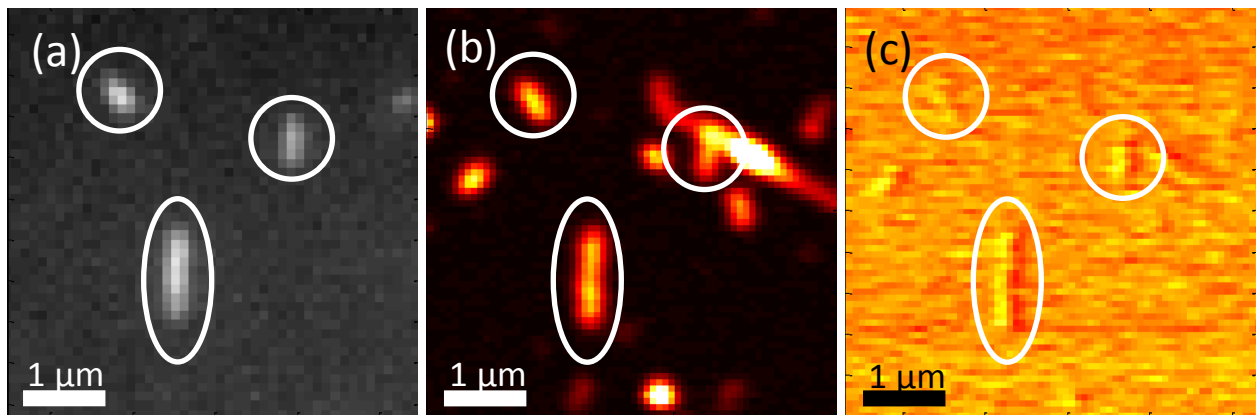


Figure 3S: Luminescence (a, 300 ms integration time), photothermal (b, 5 ms/pixel) and modulated absorption (c, 25 ms/pixel) images of the same sample region. (6,5) carbon nanotubes are encircled. Only well-isolated long vertical nanotubes are studied.

Photoluminescence excitation spectra S_{22} transition of individual (6,5) nanotubes:

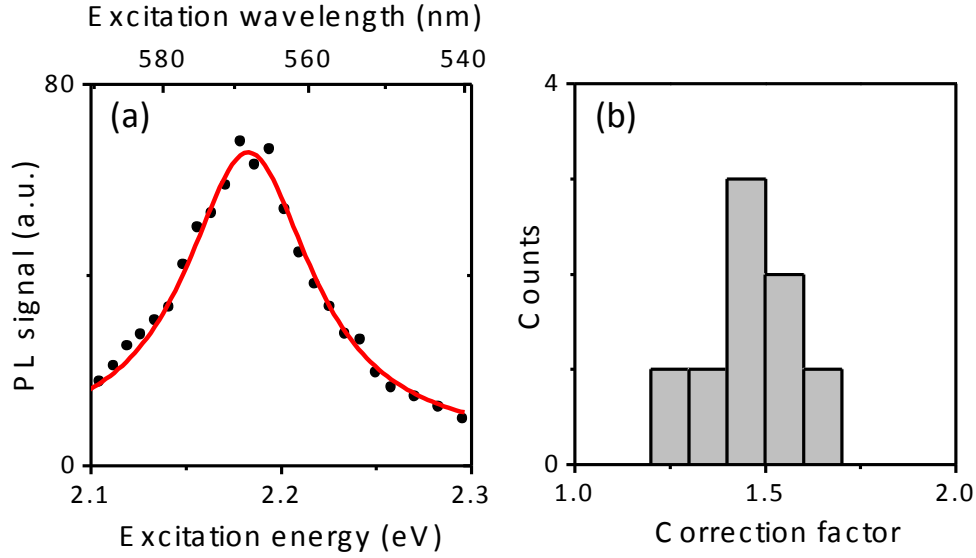


Figure 4S: (a) Photoluminescence excitation spectrum of an individual (6,5) carbon nanotube with its Lorentzian fit with a peak S_{22} transition found at 569 ± 2 nm. (b) From such spectra, a correction factor of 1.47 ± 0.12 is deduced for the determination of σ_c at the S_{22} peak transition using the measured values at 561nm (see main text).

Steady-state exciton concentration calculation with laser scanning microscopy modality

The exciton concentration $c(x)$ along a carbon nanotube of length L was theoretically derived from the diffusion equation with a source of excitons generated by the absorption of a Gaussian

beam with a profile along the tube axis $I(x) = I_0 e^{-\frac{2(x-x_0)^2}{w_0^2}}$. In the low excitation regime, when the local exciton generation is proportional to the excitation intensity the diffusion equation writes

$$\frac{\partial c(t,x)}{\partial t} = D \frac{\partial^2 c(t,x)}{\partial x^2} - \frac{c(t,x)}{\tau} + \sigma_c n_c \frac{I_0}{h\nu} e^{-\frac{2(x-x_0)^2}{w_0^2}}$$
 with D the exciton diffusion constant, τ the exciton lifetime, n_c the number of carbon atoms per tube length, σ_c the absorption cross section per carbon atom and $h\nu$ the photons energy. In the stationary regime it becomes:

$\frac{\partial^2 c(x)}{\partial x^2} - \frac{c(x)}{l_D^2} + \Lambda e^{-\frac{2(x-x_0)^2}{w_0^2}} = 0$, with boundary conditions $c(x \leq 0) = c(x \geq L) = 0$, $l_D = \sqrt{D\tau}$ the exciton diffusion length and $\Lambda = \frac{\sigma_c n_c I_0}{D h \nu}$.

The data presented in figure 4, are simulated for $L=5 \mu\text{m}$ and for different exciton diffusion lengths. Each curve is obtained by (i) solving the stationary diffusion equation for different positions x_0 of the beam along the tube, (ii) calculating the emission distribution in the detection plane as the convolution $c(x)$ with the emission point spread function of the imaging optics and detection optics (microscope objective NA=1.45, emission wavelength 985 nm) and (iii) integrating the resulting profile over the area of the detector (equivalent radius of $1.1 \mu\text{m}$ in the sample plane).

Figure 5S present two examples, of the exciton concentration generated in the nanotube for two beam positions (at the end and at center of the tube) and the corresponding PL photon distribution in the detection plane using $l_D = 200 \text{ nm}$

The absorption, which is proportional to the integral of the Gaussian that overlaps the nanotube, is also presented for comparison with the PL profile.

