Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Ultrafast Nonlinear Photoresponse in Single-Wall Carbon Nanotubes: A Broadband Degenerate Investigation

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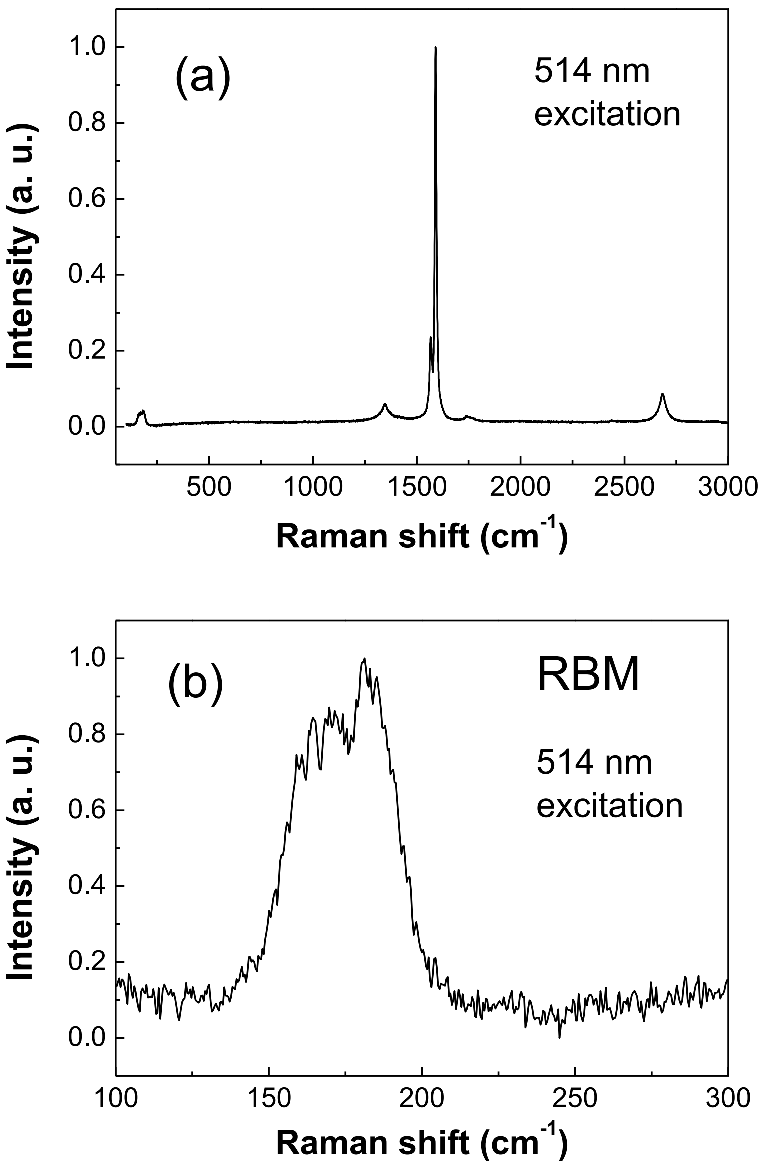

Figure S1. (a) Raman spectrum of SWNTs, (b) Raman spectrum of SWNTs showing Radial Breathing Mode (RBM), under 514 nm excitation. From Ref [28] (M. S. Strano et al., Nano Lett.) of the manuscript, it can be deduced that the RBM spectrum corresponds to tube diameter ranging from 1.3-1.6 nm.

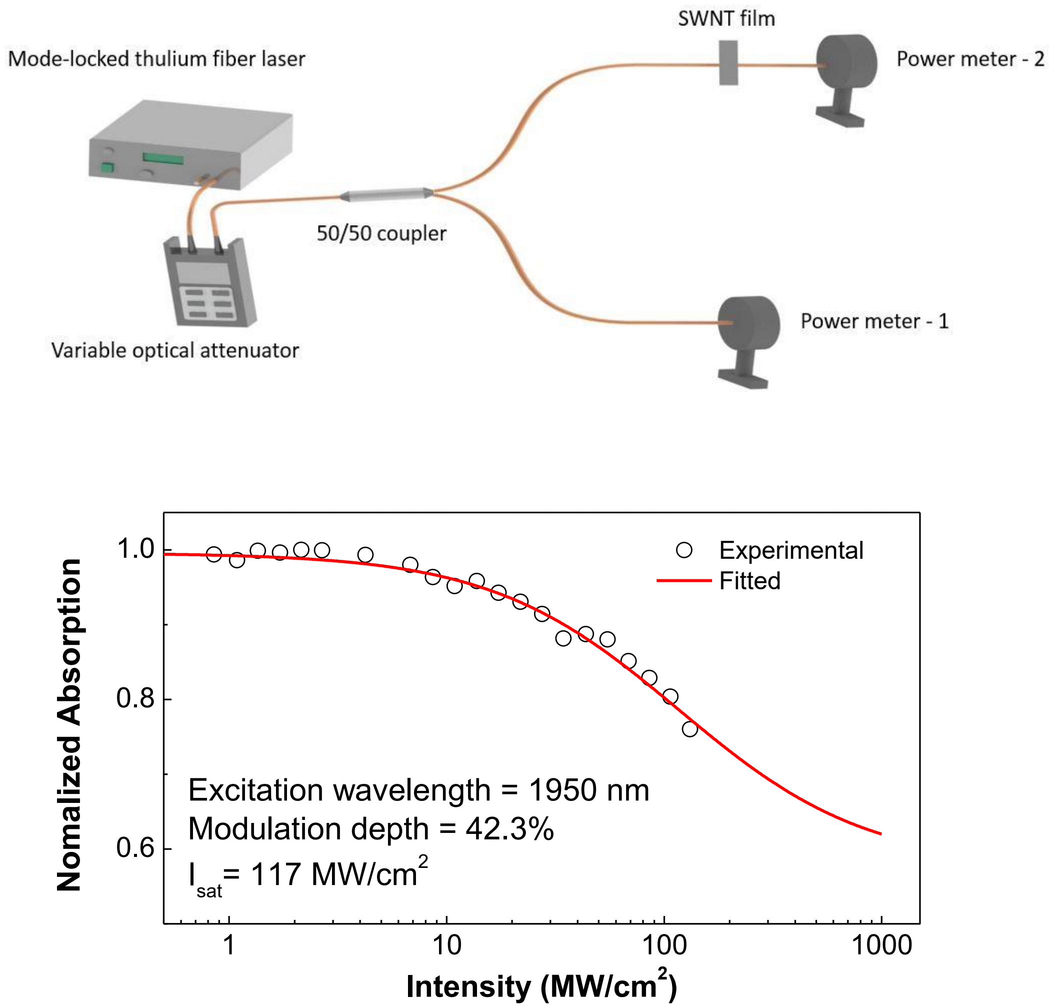

Figure S2. Nonlinear absorption characterization using a mode-locked thulium fiber laser, operating at 1950 nm (NPI Lasers, Inc.). The pulses incident onto the SWNT sample is ~ 6 ps. From the characterization, a modulation depth of 42.3%, and a saturation intensity of ~ 117 MW/cm^2 can be obtained.

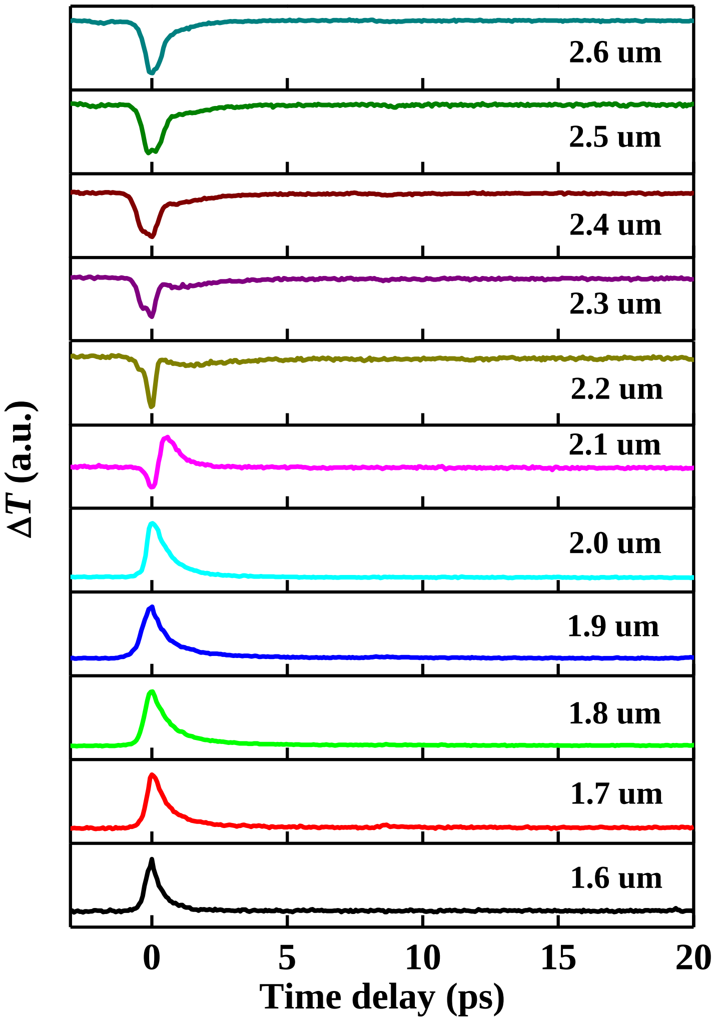

Figure S3. Non-degenerate pump-probe results on SWNTs thin film sample (without polymer matrix). The pump wavelength is 800 nm, with a fluence of 1.05 mJ/cm^2. A transition from photobleaching (PB) to photoinduced absorption (PA) is found to occur at a wavelength of ~ 2100 nm, in agreement with the degenerate results shown in Fig.5 of the manuscript. Such observation confirms that the onset of the PA band at ~ 2100 nm is intrinsic to the SWNTs used in this work.

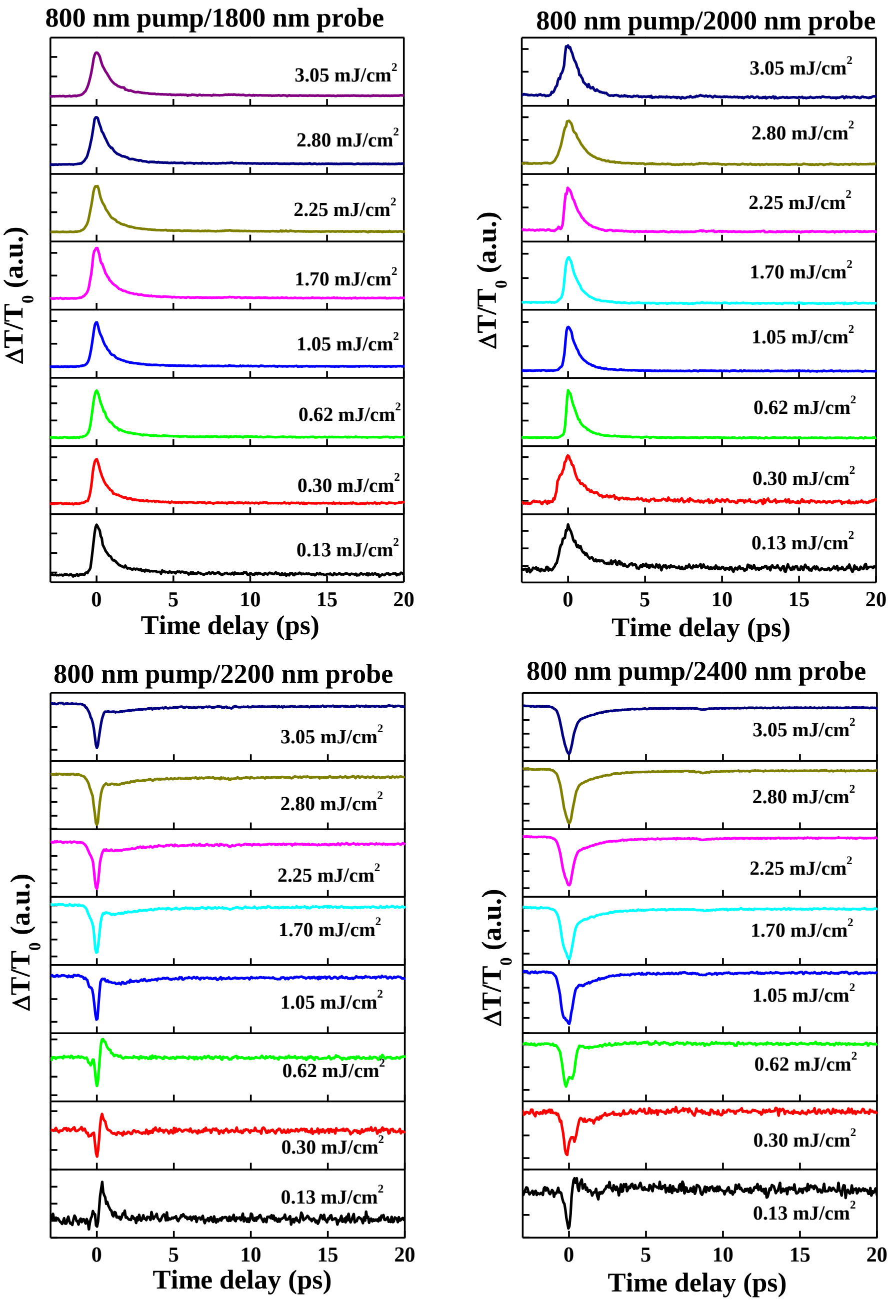

Figure S4. Non-degenerate pump-probe results on SWNTs thin film sample (without polymer matrix). The pump wavelength is 800 nm, the pump fluence is tuned from 0.13-1.05 mJ/cm^2. For wavelength below 2100 nm, i. e. 1800 nm and 2000 nm, PB signal is always observed, whereas for wavelength above 2100 nm, i.e. 2200 nm and 2400 nm, PA is primarily observed.

Supplementary Note:

The absorption property of individual nanotubes makes up the macroscopic behavior of the nanotube array. Our previous work has put forward a phenomenological model for suspended individual nanotubes, in which the experimental absorption spectra are produced within 20% accuracy (Text Ref [9]). The absorption spectrum of individual nanotubes is simulated by the following formulas

$$A_{nm}(E) = \sum_p \frac{\Sigma_p}{\pi} \cdot \frac{w_p}{(E - E_p)^2 + w_p^2} + \frac{\Sigma_p}{a \cdot \pi} \cdot conv\left(\frac{b \cdot w_p}{E^2 + (b \cdot w_p)^2}, \frac{\vartheta[E - (E_p + \Delta)]}{\sqrt{E - (E_{ii} + \Delta)}}\right)$$

where n,m is the chiral number of the nanotube and p denotes the order of the transition. We consider that the diameter of nanotube sample obey certain distribution function $f = f(d)$, where d is the diameter. We could then obtain the optical absorption of the sample containing an ensemble of nanotubes by

$$A(E) = \Sigma_{nm} f(d) \cdot A_{nm}(E)$$

In this paper, $f(d) = 1$, $d \in (1.3,\ \ 1.6)$ nm is chosen to represent the evenly distributed tube chiralities. We note that when the width of the transition peak w_p is set to three to five times of the values used for individual nanotube (accounting for dielectric screening by other tubes in the bundles and other effects), best approximation of the experimental tube absorption can be achieved.

Text Ref [9] K. H. Liu et al. “Systematic determination of absolute absorption cross-section of individual carbon nanotubes.” Proceedings of the National Academy of Sciences **111**, 7564 (2014).