

Supplemental Information for:

Optical Detection of Single Nano-Objects by Transient Absorption Microscopy

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Expression for the Transient Absorption Signal of a Single Nanowire:

The signal in the transient absorption microscopy experiments depends on the non-linear cross-section S_{NL} of the nano-object, and the intensity of the pump and probe beams at the position (x,y) of the nano-object. Specifically, the change in the detected probe intensity is:

$$\Delta P_{probe} = -S_{NL} \times I_{pump}(x,y)I_{probe}(x,y) \quad (S1)$$

which is Equation (2) of the main text. S_{NL} implicitly depends on the frequencies of the pump and probe beams and the time delay between them, and includes contributions from transient absorption, stimulated emission and transient bleach. Written in this way, S_{NL} has units of $\text{m}^4 \text{W}^{-1}$. Treating the laser beams as Gaussian beams, the intensities at the focus of the microscope can be written as

$$I(x,y) = \left(2P/\rho w^2\right) \text{Exp}\left\{-2(x^2 + y^2)/w^2\right\}, \quad (S2)$$

where w is the beam waist, and P is the power carried by the beam. For a nanoparticle with size much less than w at the center of the beams, the intensities experienced by the particle are $I(0,0) = \left(2P/\rho w^2\right)$, which leads to Equation (3) of the main text, and subsequently Equation (4), which gives the signal-to-noise in the experiments.

For a nanowire, the intensities have to be integrated along one dimension. Consider a long nanowire, with a length much greater than w , that is orientated along the x-axis (arbitrary), and is at $y = 0$. In this case

$$\begin{aligned}
 DP_{probe} &= -S_{NL} \times \int_{-\infty}^{\infty} dx I_{pump}(x, 0) I_{probe}(x, 0) \\
 &= -S_{NL} \times \left(\frac{2P_{pump}}{\rho w_{pump}^2} \right) \left(\frac{2P_{probe}}{\rho w_{probe}^2} \right) \int_{-\infty}^{\infty} dx \text{Exp} \left[-2x^2 \left(\frac{1}{w_{pump}^2} + \frac{1}{w_{probe}^2} \right) \right]. \quad (S3) \\
 &= -S_{NL} \times \left(\frac{2P_{pump}}{\rho w_{pump}^2} \right) \left(\frac{2P_{probe}}{\rho w_{probe}^2} \right) \times \sqrt{\frac{\rho}{2}} \sqrt{\frac{w_{pump}^2 w_{probe}^2}{w_{pump}^2 + w_{probe}^2}}
 \end{aligned}$$

This expression can be written in a simpler form by noting that beam waist is related to the area of an equivalent “top hat” beam by $A = \rho w^2/2$.¹ This yields

$$\begin{aligned}
 DP_{probe} &= -S_{NL} \times \left(\frac{P_{pump}}{A_{pump}} \right) \left(\frac{P_{probe}}{A_{probe}} \right) \sqrt{\frac{A_{pump} A_{probe}}{A_{pump} + A_{probe}}} \\
 &= -S_{NL} \times \left(\frac{P_{pump} P_{probe}}{\sqrt{A_{pump} A_{probe}} \times \sqrt{A_{pump} + A_{probe}}} \right), \quad (S4)
 \end{aligned}$$

which leads to Equation (5) of the main text. Note that the units of S_{NL} are now $\text{m}^3 \text{W}^{-1}$, that is, S_{NL} in Equation (S4) and Equation (5) of the main text is implicitly the nonlinear susceptibility per unit length of the nanowire (compare to Equation (S1)).

1 See, for example, B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics*, New York, Wiley, 1991.