1D Exciton Diffusion on Semiconducting Nanotubes Using Photoabsorption Spectroscopy

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Power law population relaxation ($\propto 1/\sqrt{t}$)

History dependence of PA Lineshape
$\Rightarrow$ exciton confinement energy

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OFFICE OF SCIENCE U.S. DEPARTMENT of ENERGY

Experimental

HipCO Tubes dissolved in 2% SDBS solution and purified

Pump: 0.1-1.0 mJ/cm^2 @ 725 nm

0.1-0.5 mJ/cm^2 @ 670 nm

Probe: S_{11} absorption from 920-1430 nm;

0.2 ps < t < 120 ps

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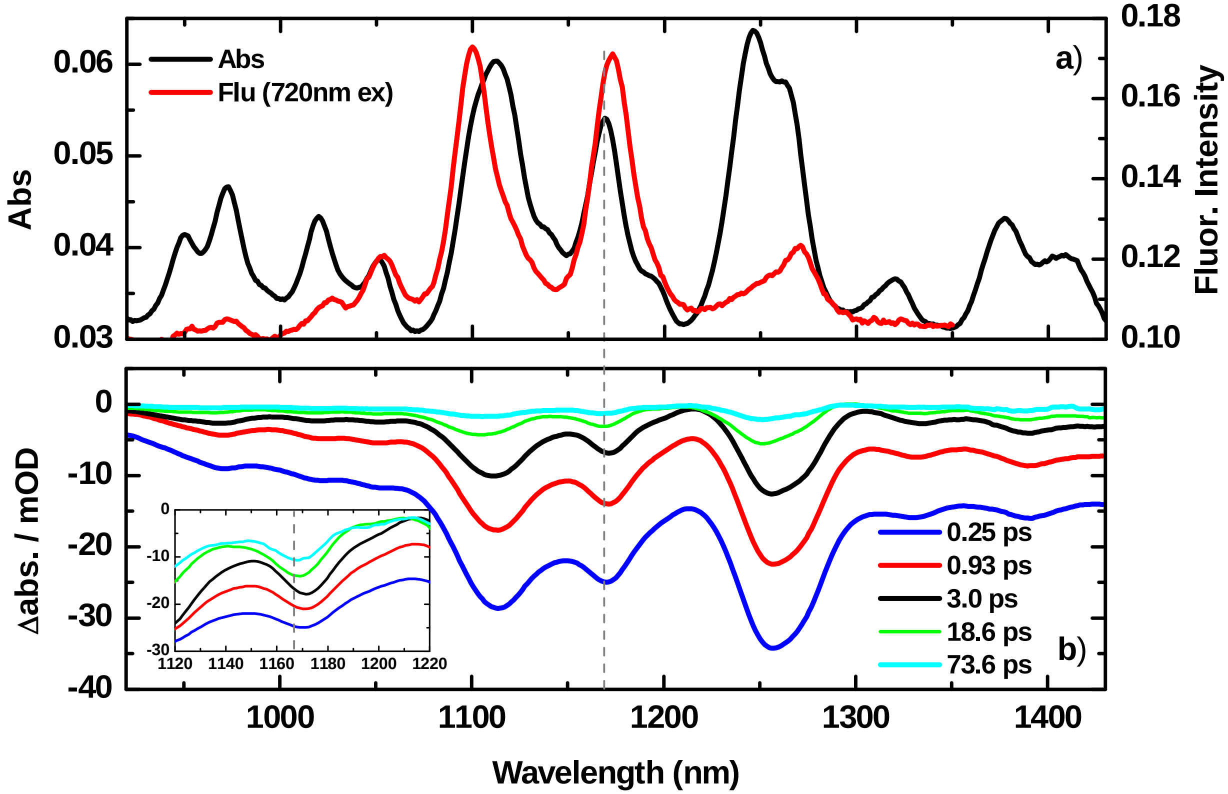
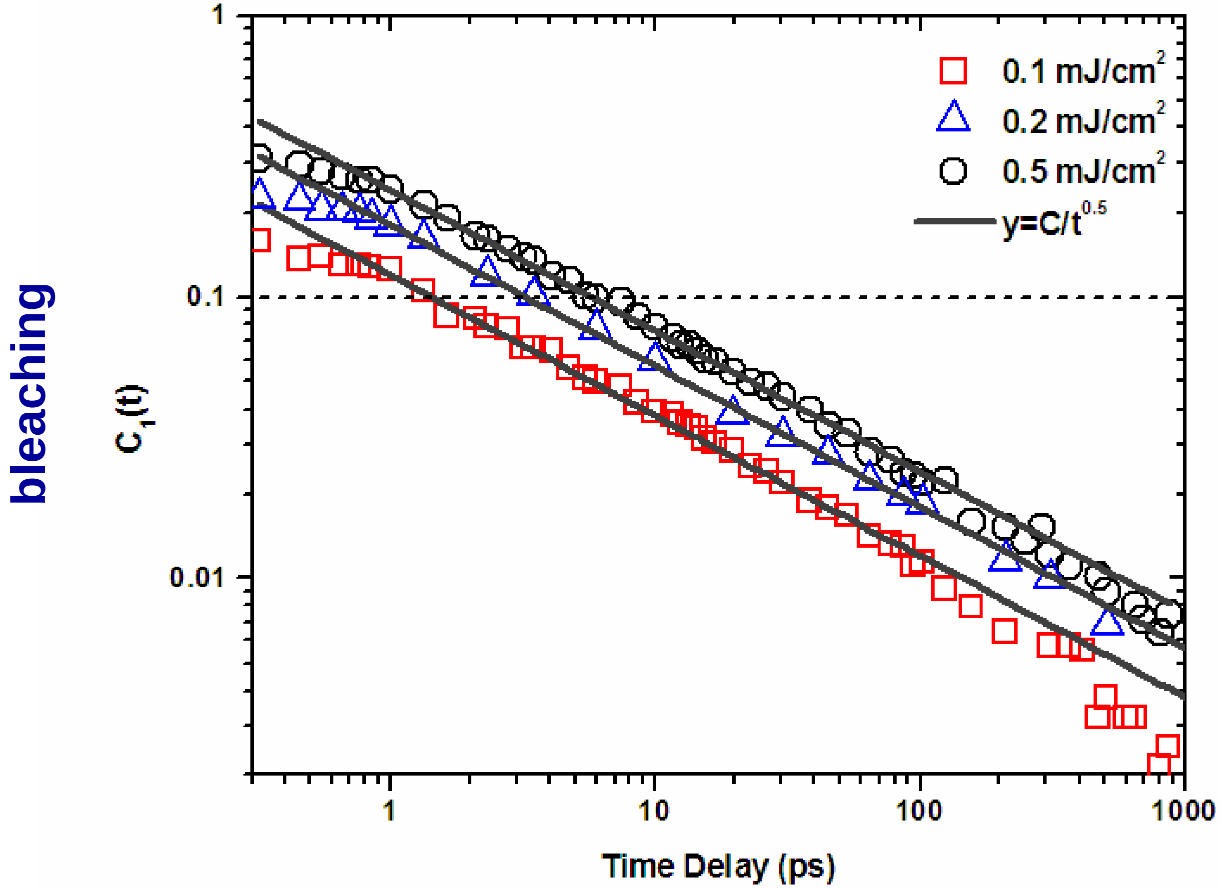
Measure $\Delta\alpha(\lambda,t)$ after primary excitation

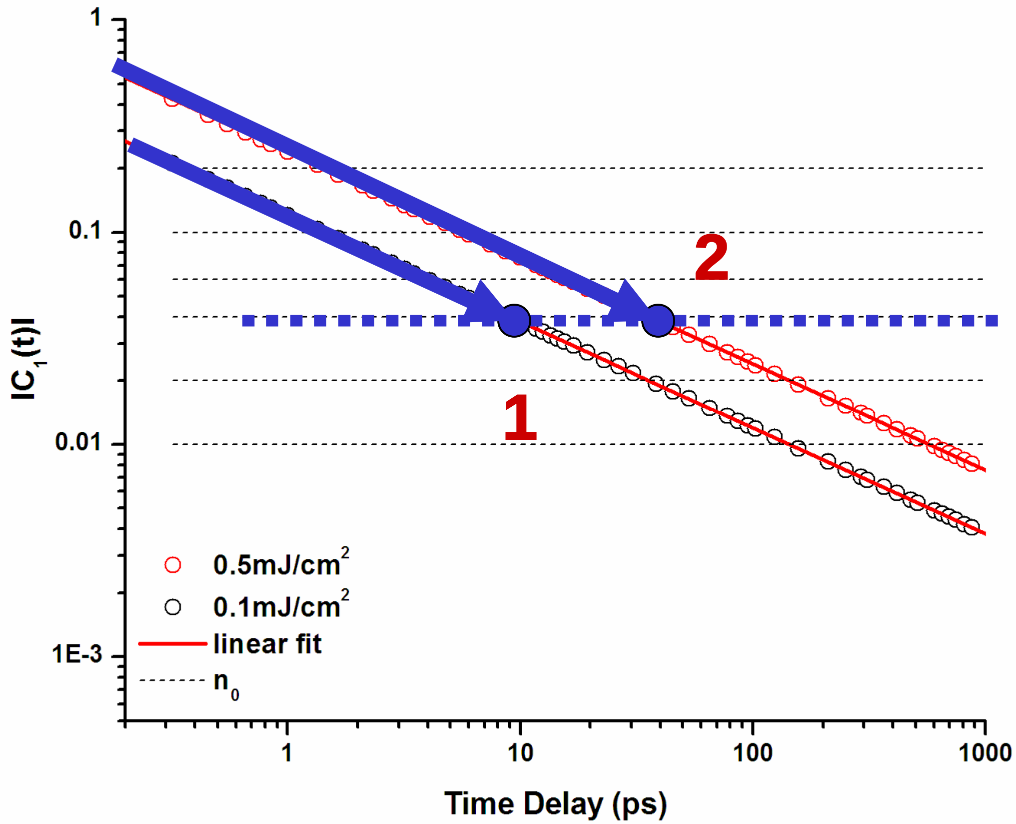


Power Law Relaxation of Excited Population



R.M Russo et al. Phys. Rev. B 74, R041405 (2006)

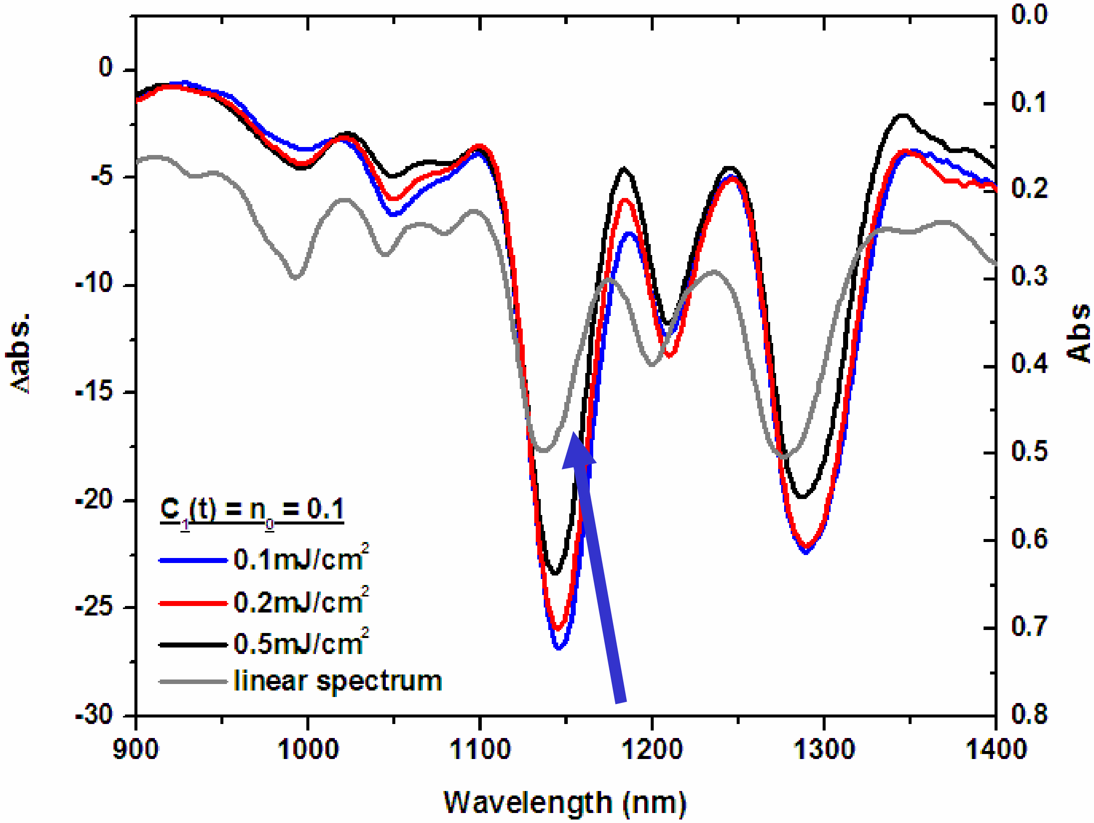
For two systems evolved to same density



$$\frac{\dot{n}_1}{\dot{n}_2} > 1$$

system evolved from the higher density is the "slower"

For two systems that evolve to same density



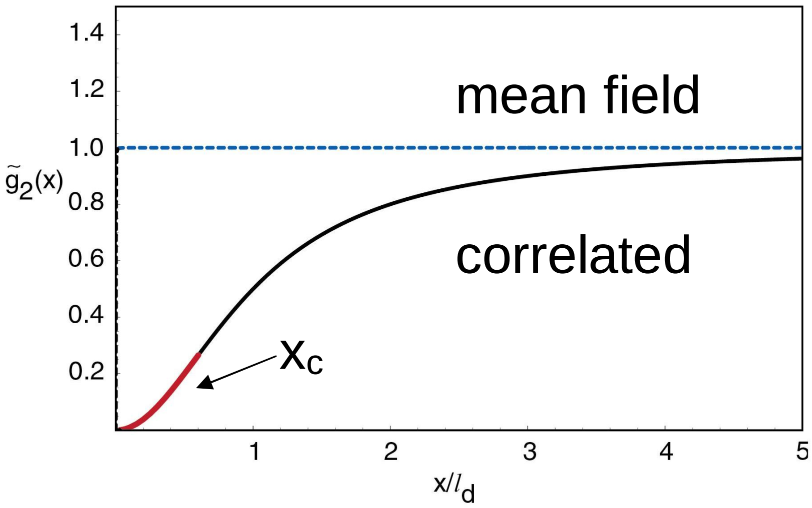
.. and its bleaching spectrum is shifted

"*history dependence*"

Diffusion Limited Pair Annihilation

In mean field theory: $$\frac{dn}{dt} = -Cn^2$$

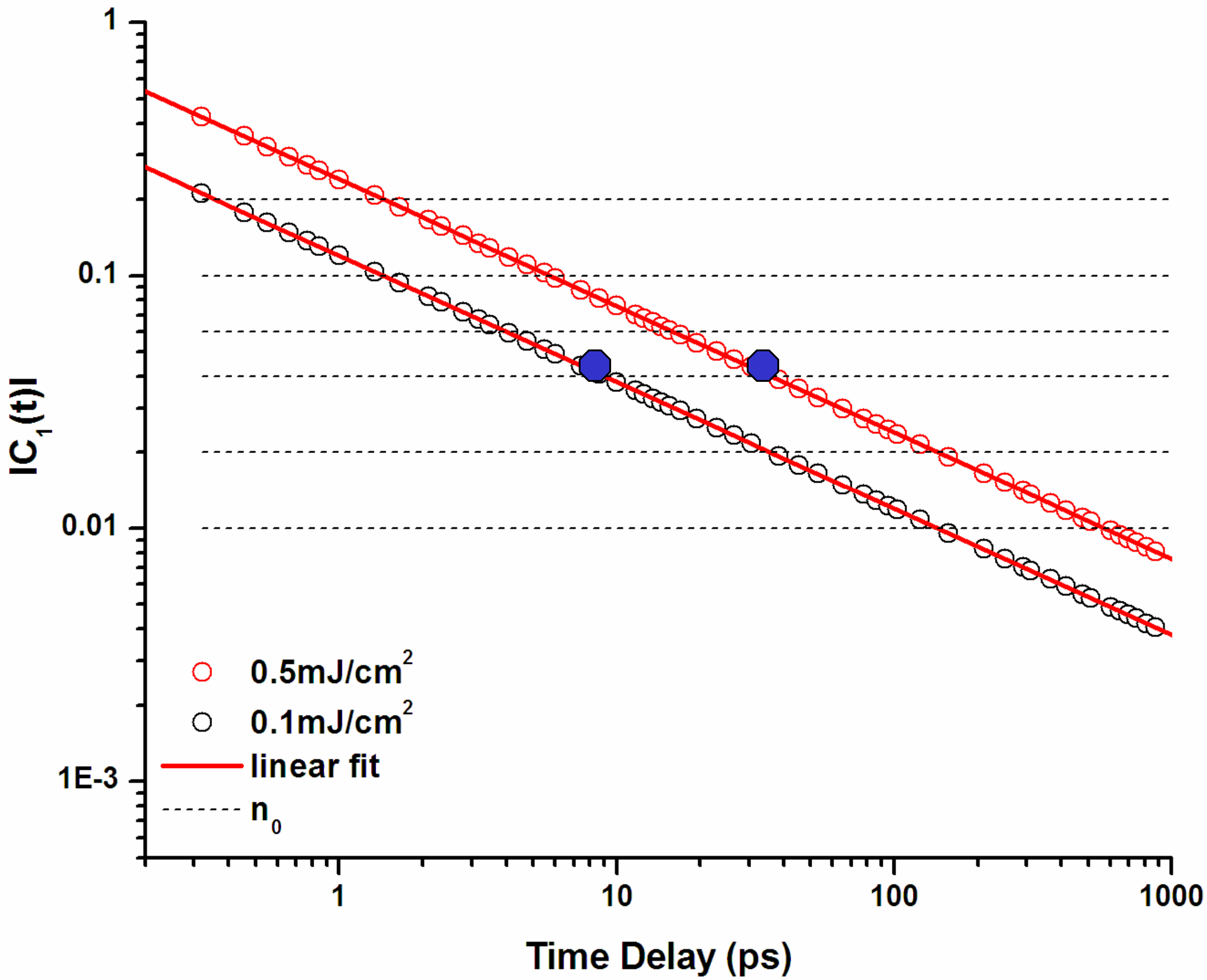
but in a correlated distribution: $$\frac{dn}{dt} = -C \times \tilde{g}_2(x_c, t) \times n^2$$



e.g:

$$g_2(x,t) = \frac{x^2}{x^2 + Dt}$$

Relate the Relaxation Rates

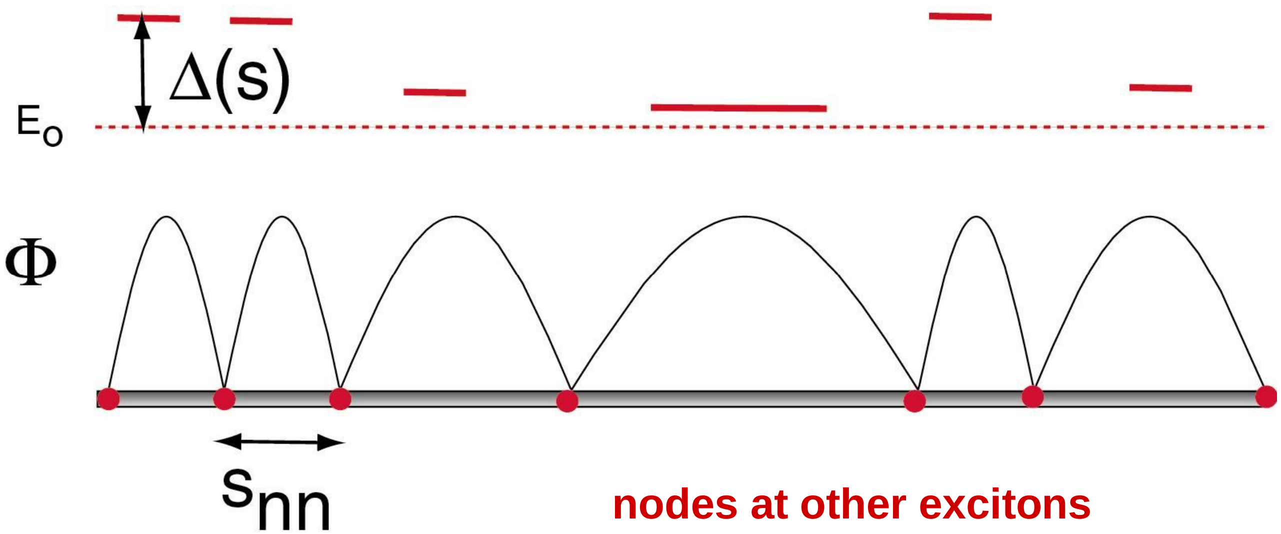


$$\frac{\dot{n}_1}{\dot{n}_2} = \frac{\tilde{g}_2(x_c, t_1)}{\tilde{g}_2(x_c, t_2)}$$

for $x_c \ll \ell_d$

$$\approx \frac{x_c^2 / Dt_1}{x_c^2 / Dt_2} = \frac{t_2}{t_1}$$

Frequency Shifts from Exciton Confinement Energy



Lineshape from distribution of nn spacings

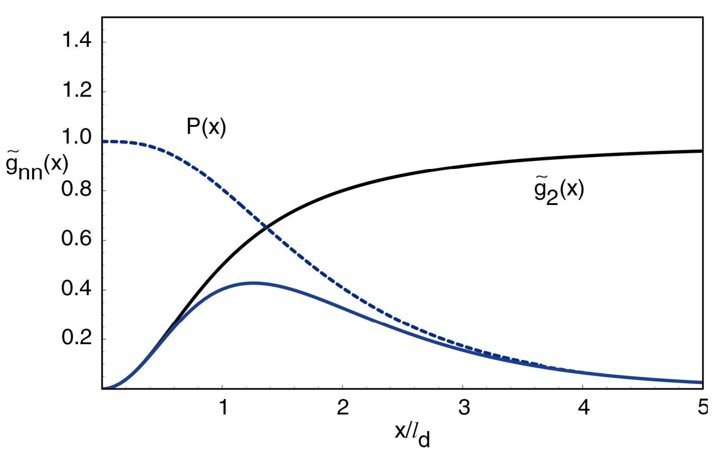
Time evolution of distribution of nn spacings

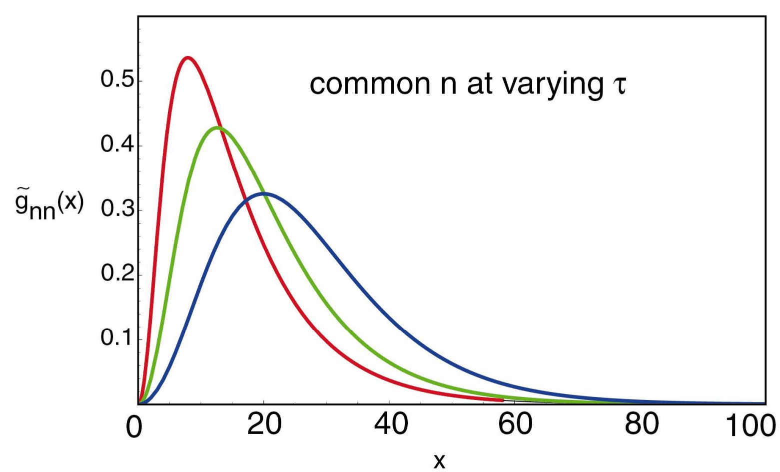
$$g_{nn}(x) = n \times \tilde{g}_2(x/\ell) \times P(x,t);\ \ell = \sqrt{Dt}$$

Distribution of pair separations

Probability of first neighbor at range x

$$P(x,t) = \exp\left(-n(t)\,x + n(t)\,\ell \times \arctan(x/\ell)\right)$$





Lineshape Calculation

Lineshape from ground state absorption

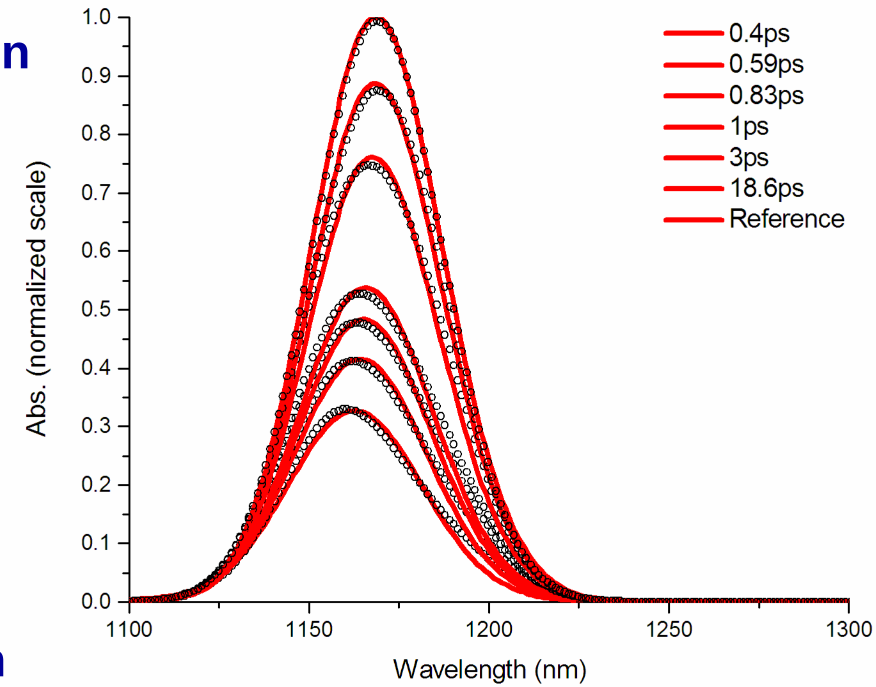
$$A(\omega,\omega_o) = L(\omega - \omega_o)$$

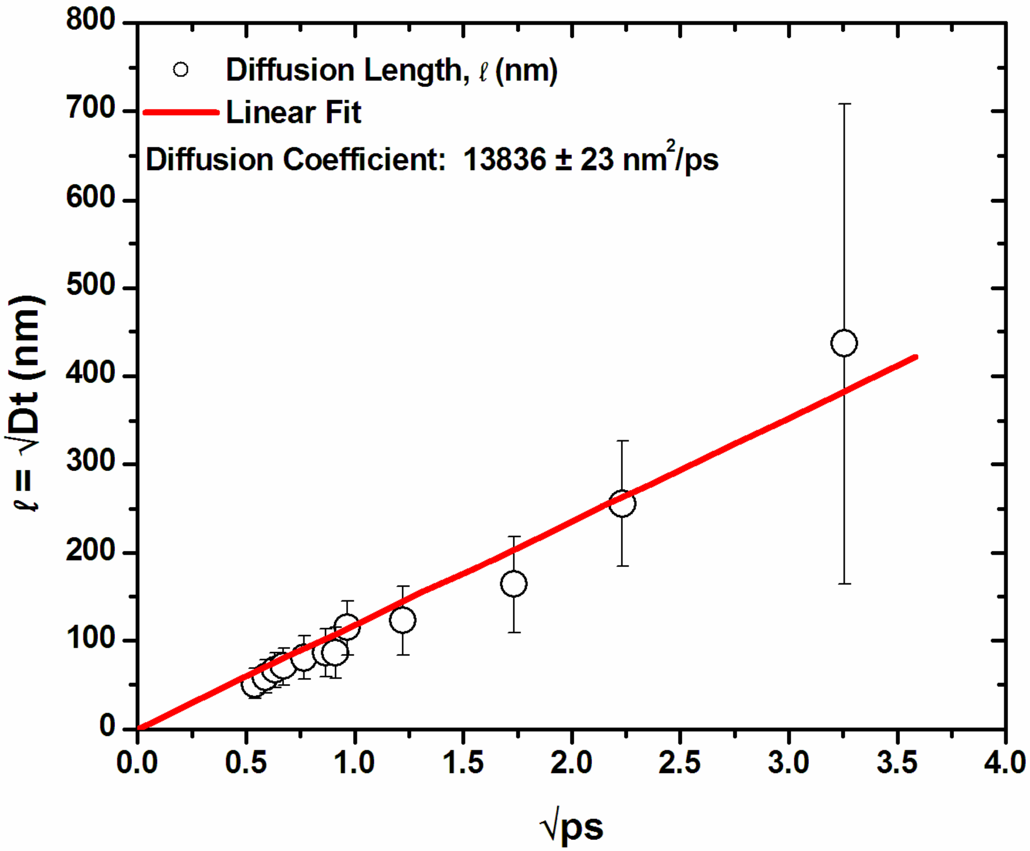
..shifted by the confinement energy

$$\omega_o \rightarrow \omega_o \left(1 + 2\left(\frac{\pi\hbar v_F}{sE_o} \right)^2 \right)$$

.. and averaged over the nn distribution

$$\langle A(\omega) \rangle = \int_0^\infty L(\omega - \omega_o(s))\, g_{nn}(s,t) \times \frac{s}{\xi} ds$$





Best fit: D=138 +/- 23 cm^2/sec

Compare: Korovyanko et al. D~120 cm^2/sec (PRL 92, 017403 (2004))
(from measurement of polarization memory)

Further Comments

Pairwise recombination ⇔ triplet-triplet annihilation?

Exciton density (alone) does not describe
short range correlations: population relaxation, PA…

Crossover from dense to dilute regime after first passage time
$L^2/D \sim 100$ ps

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