Quantum Correlations, Spatiotemporal Control & Quantum Information Processing

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CHEMISTRY*CENTER FOR LASERS & PHOTONICS*DESIGN PROGRAM

Indian Institute of Technology Kanpur

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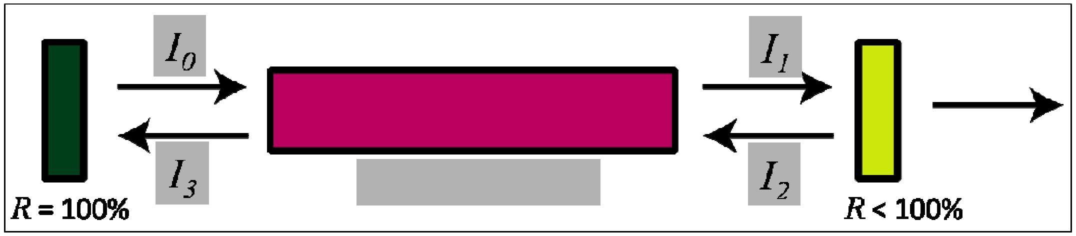
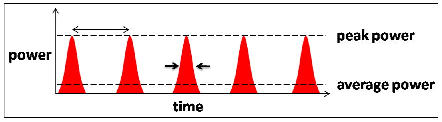
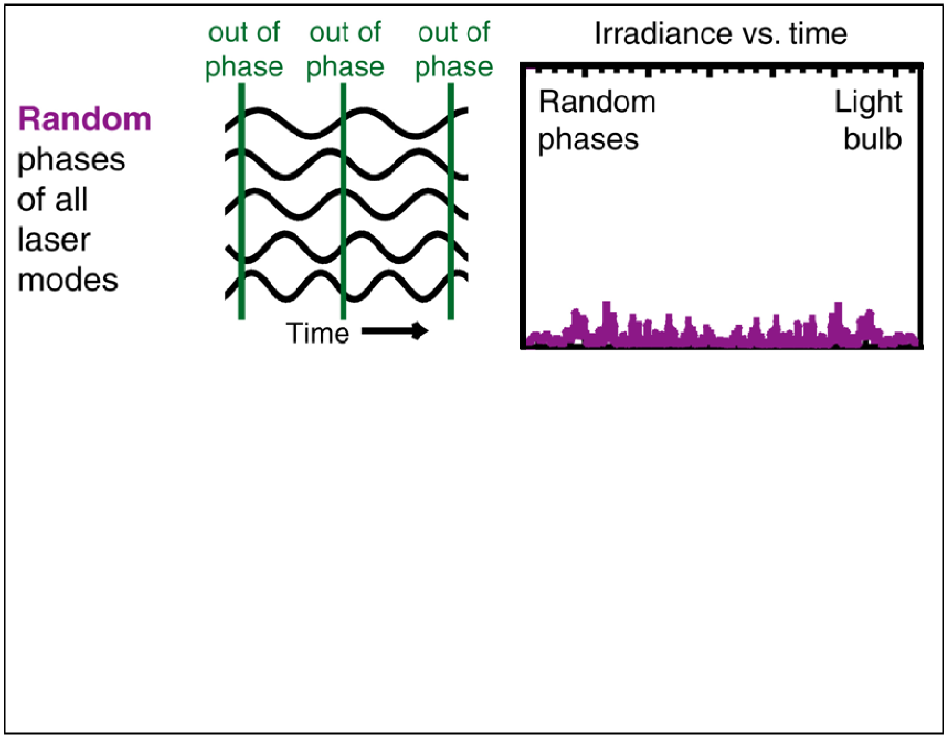
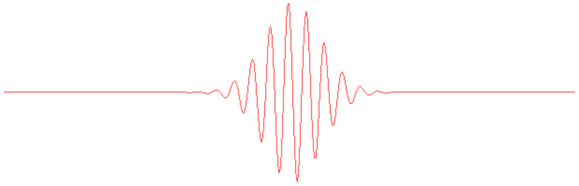
Laser sources and pulse characterization

What is an ultra-short light pulse?









$$\tilde{E}^+(t) = \frac{1}{2}\varepsilon(t)e^{i\phi_0}e^{i\phi(t)}e^{i\omega_l t}$$

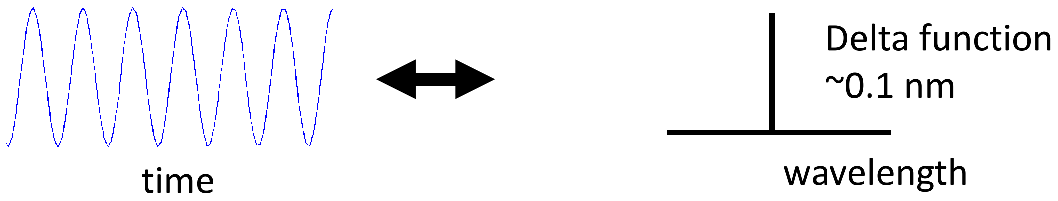
$$\tilde{E}^+(\Omega) = \left|\tilde{E}(\Omega)\right|e^{i\phi(\Omega)} = \tilde{E}(\Omega)$$

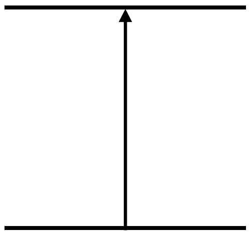
$$(\text{for } \Omega \geq 0)$$

$\tau\Delta\nu$ = constant ~ 0.441 (Gaussian envelope)

Laser Time-Bandwidth Relationship

- For a CW Laser

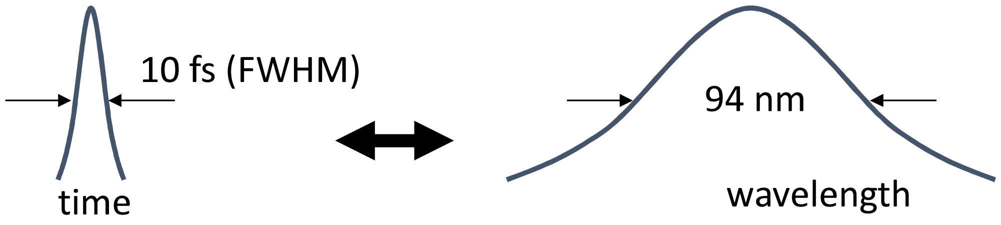


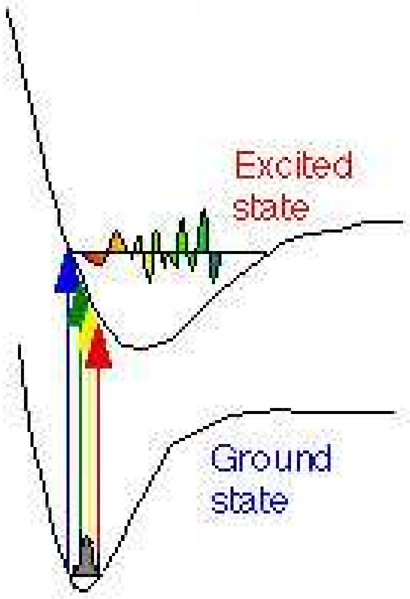


- An Ultrafast Laser Pulse
  - Coherent superposition of many monochromatic light waves within a range of frequencies that is inversely proportional to the duration of the pulse

Short temporal duration of the ultrafast pulses results in a very broad spectrum quite unlike the notion of monochromatic wavelength property of CW lasers.

e.g.
Commercially
available
Ti:Sapphire
Laser at 800nm

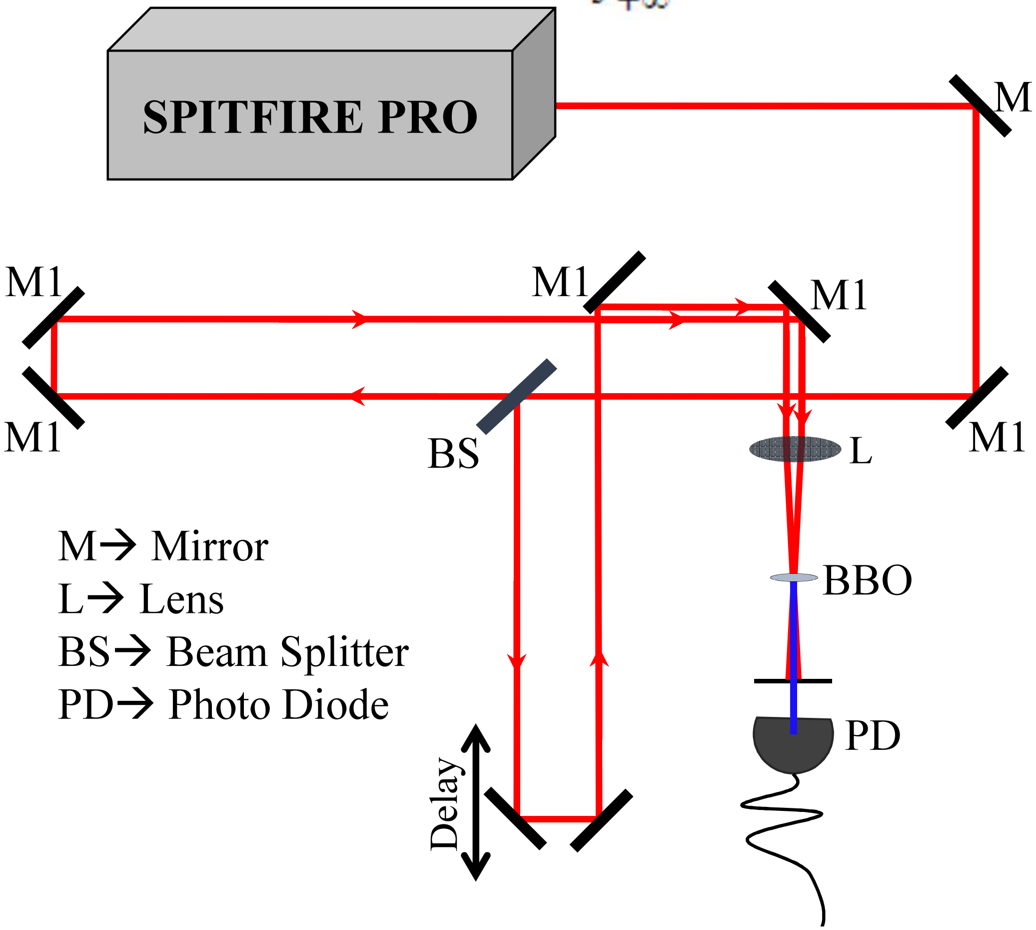




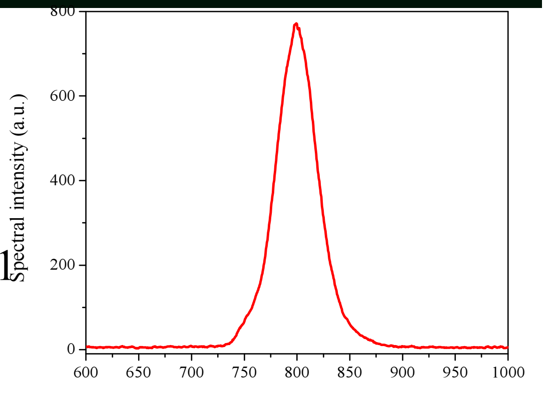
Pulse Characterization: Intensity Autocorrelation

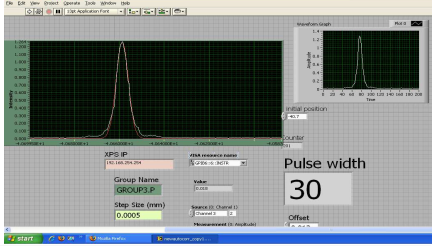
Non-collinear Intensity autocorrelation

$$A(\tau) = \int_{+\infty}^{-\infty} I(t)I(t-\tau)dt$$



M→ Mirror
L→ Lens
BS→ Beam Splitter
PD→ Photo Diode





Initial position -40.7

Counter 201

XPS IP 192.168.254.254

VISA resource name GPIB6::6::INSTR

Group Name GROUP3.P

Value 0.018

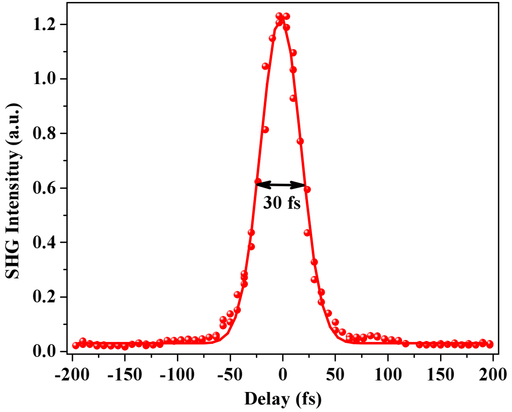
Step Size (mm) 0.0005

Source (0: Channel 1) Channel 3 2

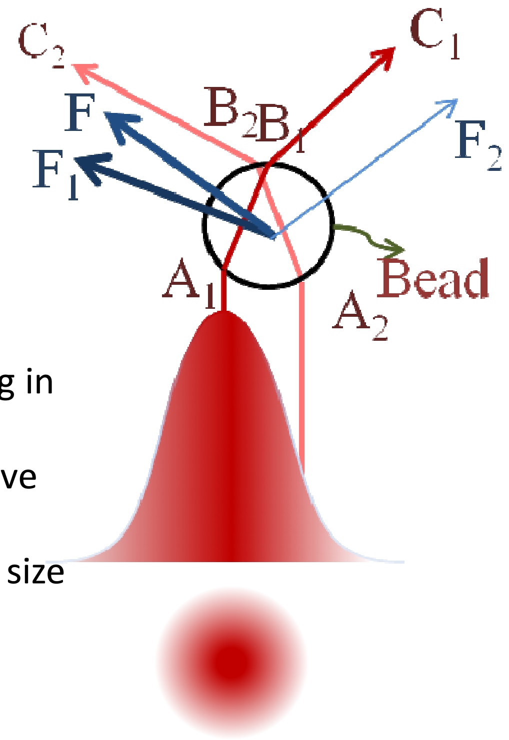
Measurement (0: Amplitude)

Pulse width 30

Offset



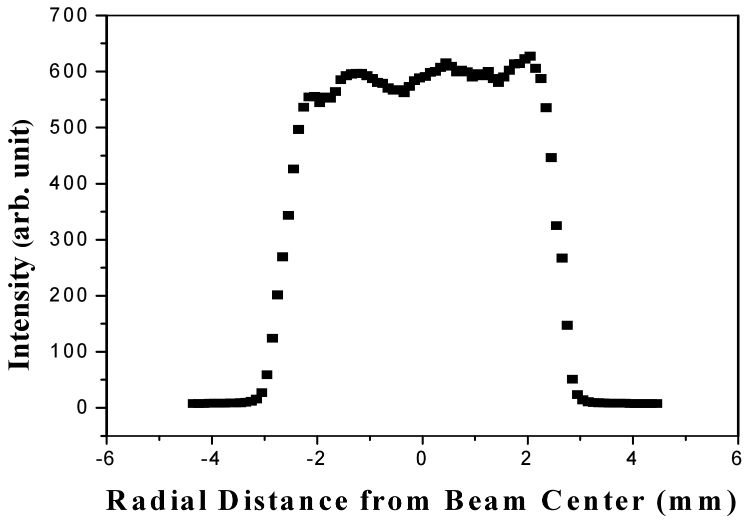
Spatial Control: Basics of optical trapping



Beam bending in higher refractive index micron size beads

Para-axial Gaussian Mode:
$E=E_0\exp(-2r^2/w^2)$

Radiation pressure from photon flux

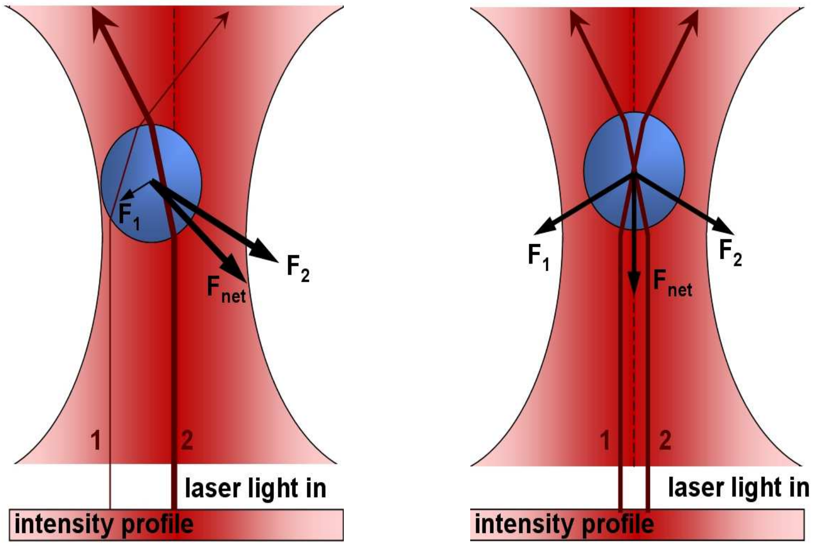


Flat-top Gaussian Mode

- ❖No trapping was observed as $\nabla E^2 \cong 0$

- ❖ For single beam optical trap, paraxial Gaussian beam is essential spatially
- ❖ Temporally, however, laser can be either cw or pulsed

OPTICAL TWEEZER



Para-axial Gaussian Mode:

$$E=E_0\exp(-2r^2/w^2)$$

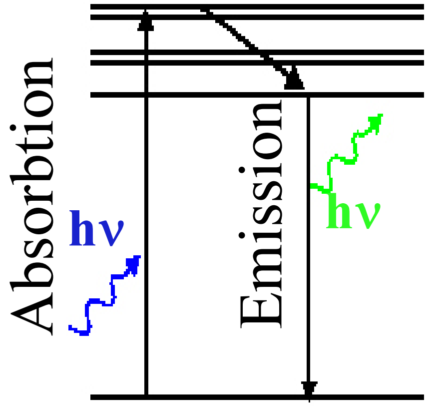
For Rayleigh Particles: $F\propto\nabla E^2$

using CW Laser $= E_0^2\exp(-4r^2/w^2)$

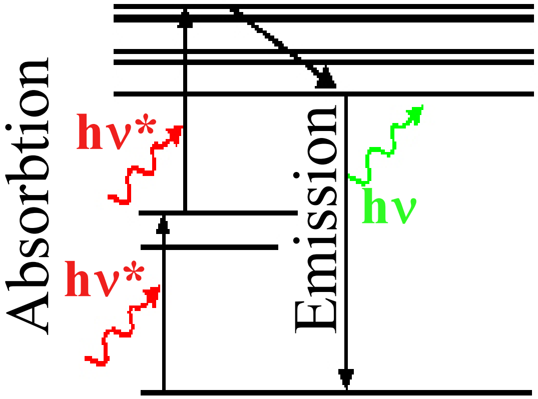
$\propto -r$ (for small r)

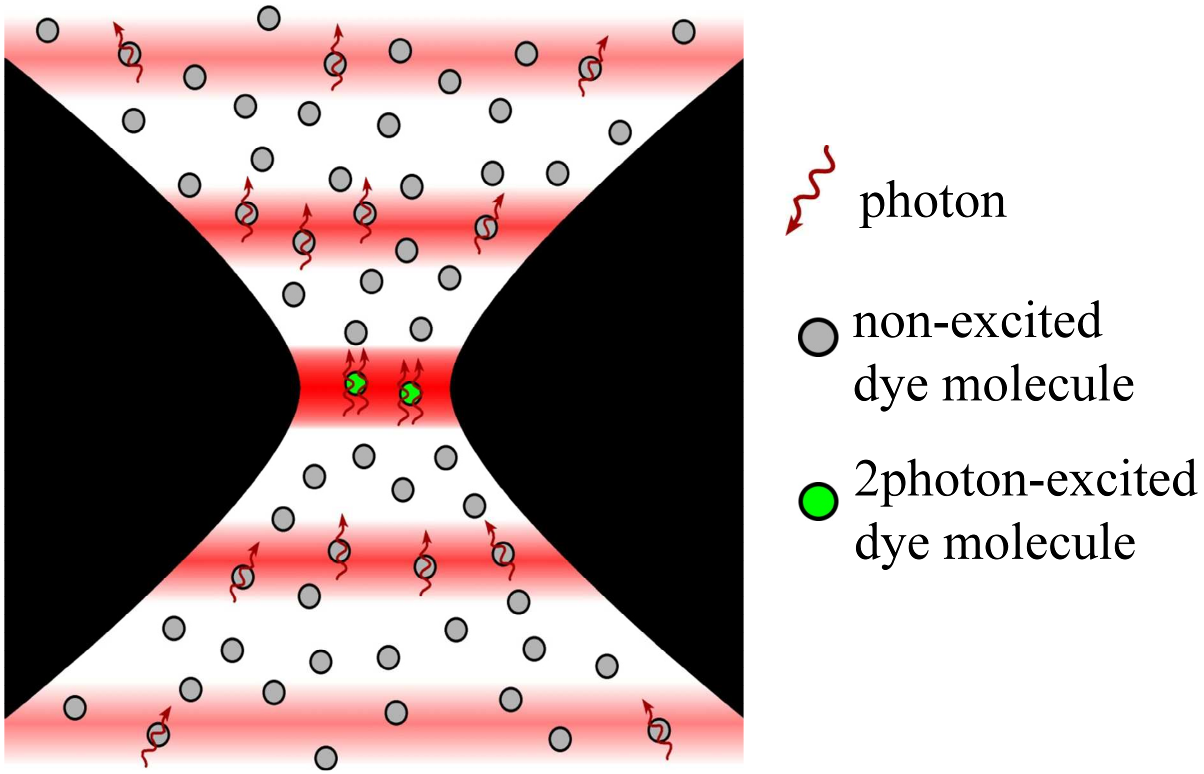
TWO PHOTON FLUORESCENCE

Single-photon Excitation

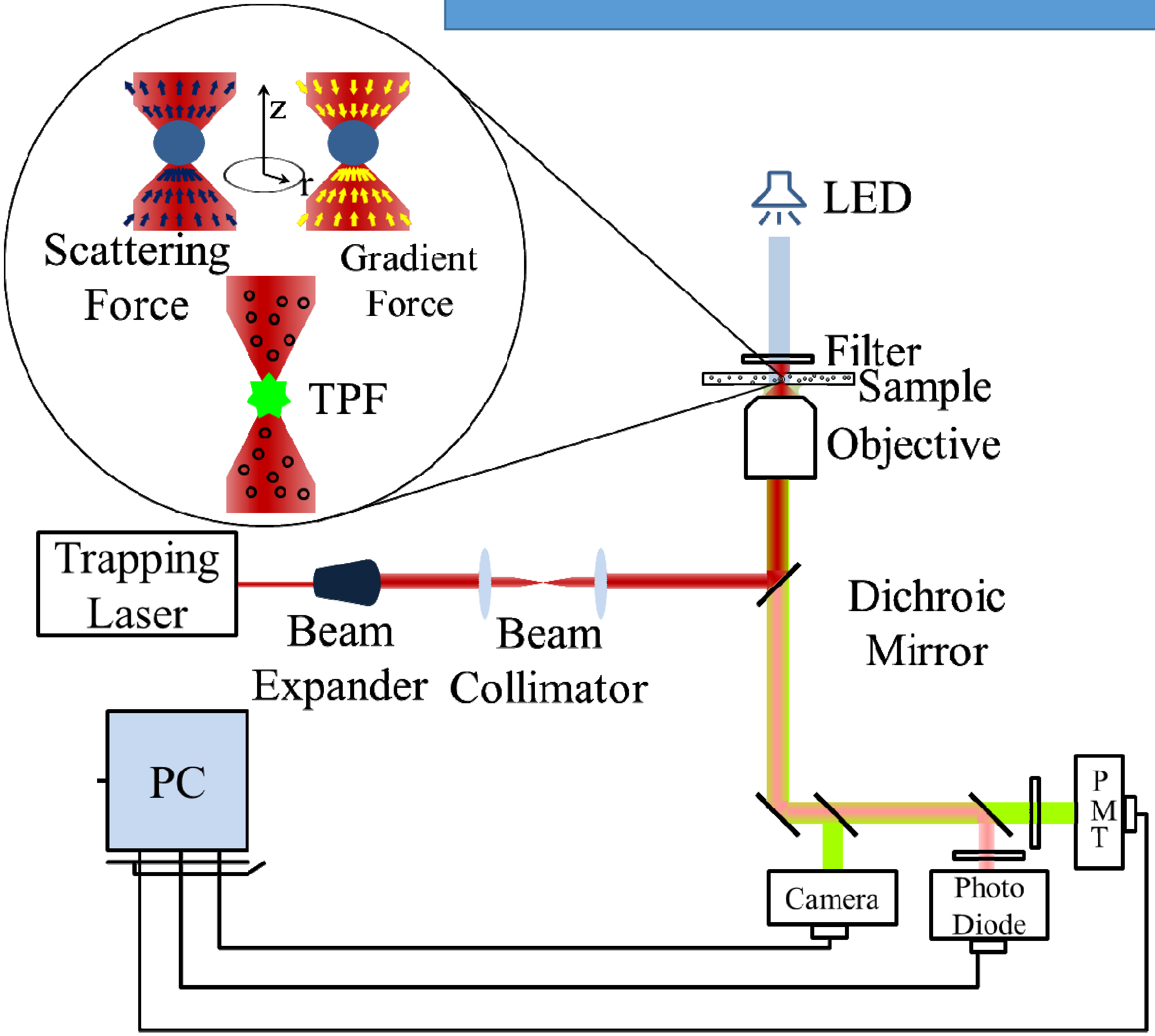


Two-photon Excitation





EXPERIMENTAL SETUP

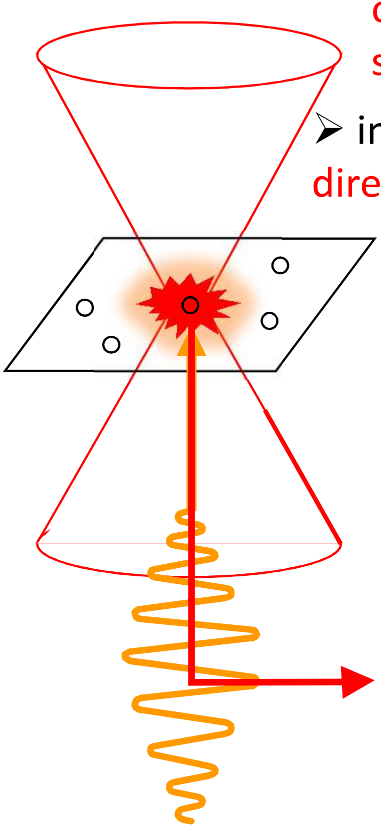


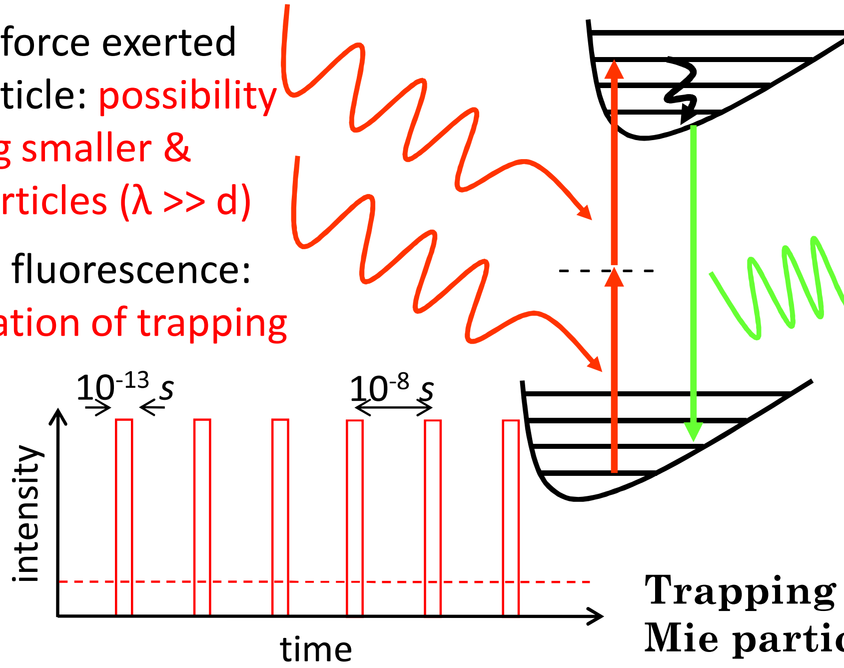
Advantages offered by a femtosecond pulsed laser

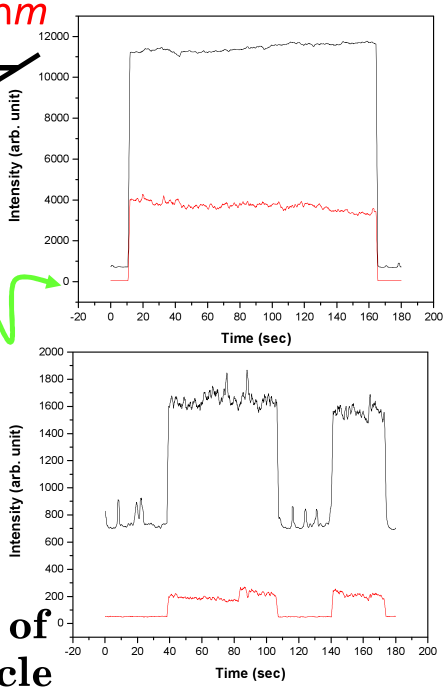
- ■ simultaneous detection of two-photon fluorescence and back-scattered light
- ■ bright-field video imaging ■ continuous-wave/mode-locked laser operation

➢ molecular fluorescence (two-photon fluorescence) at ~800 *nm*

- ➢ ~10^5 times force exerted on the particle: possibility of trapping smaller & smaller particles (λ >> d)
- ➢ intrinsic 3D fluorescence: direct observation of trapping

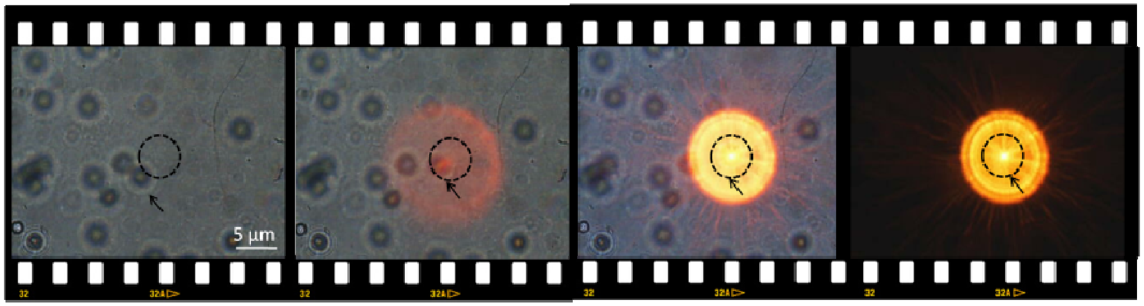






Trapping of Mie particle

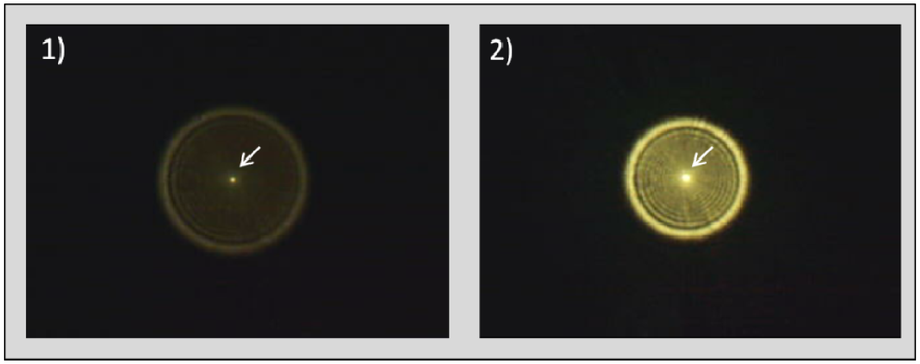
Trapped 4 and 1 µm particles stably with fs 800nm



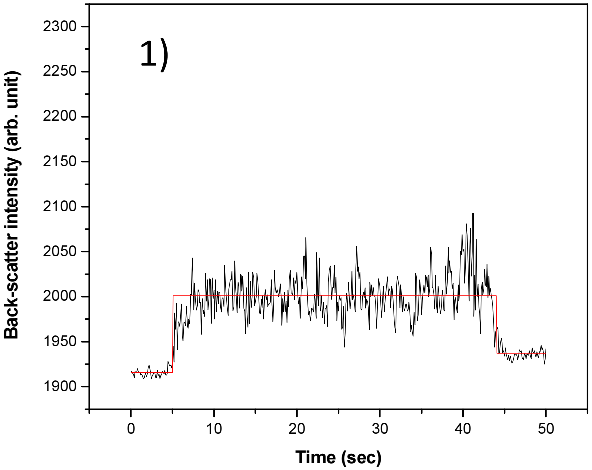
Direct Trapping of Rayleigh or sub-diffraction sized particles (λ >> d)

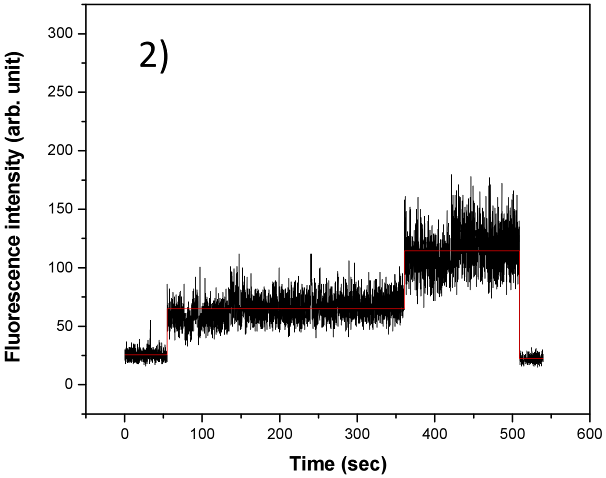
Trapping of 100 nm Latex Bead

Observed through Two-photon Fluorescence Signal only — calibrated from back-scattering data of 4 & 1 micron



Particle aggregation can also be tagged through this extremely sensitive technique (see case 2)



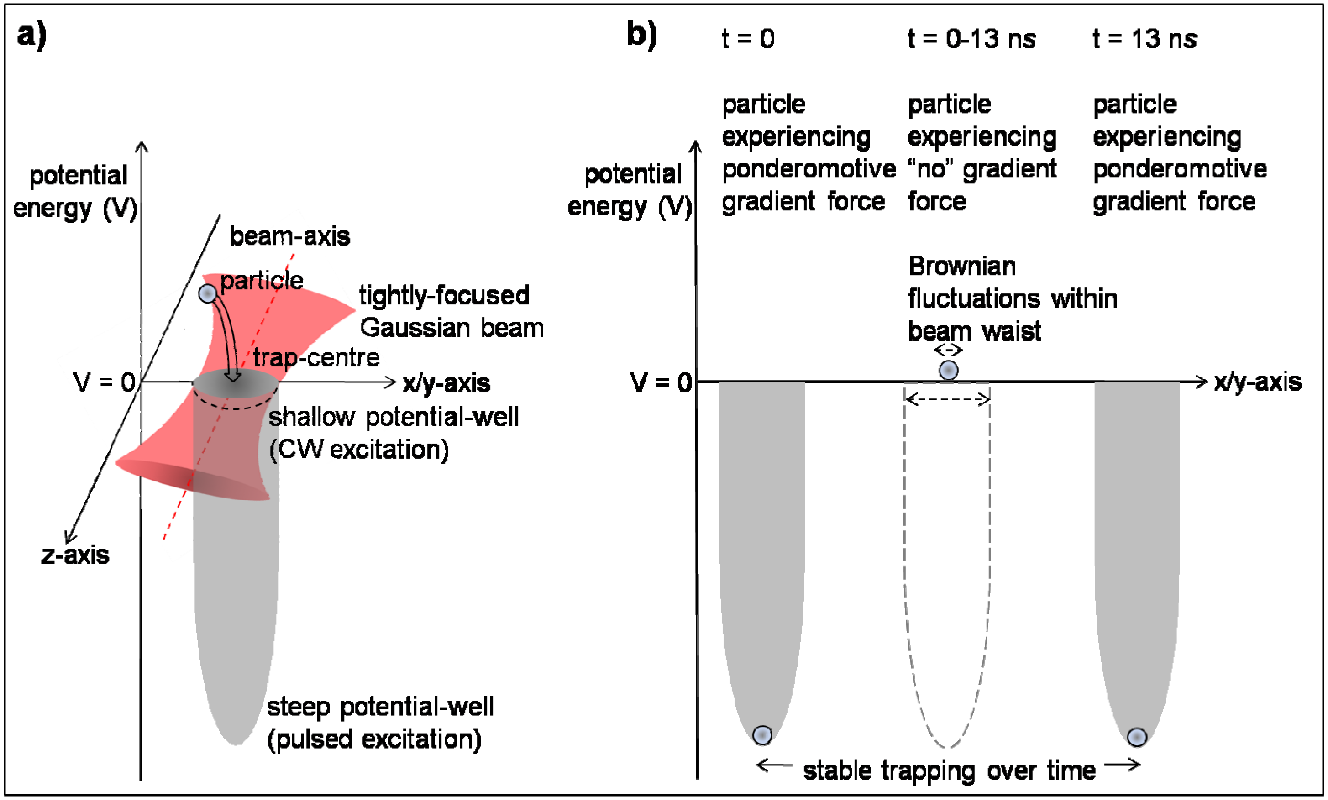


Simultaneous coupling pulse shaping to this efficient and sensitive ultrafast optical tweezers is possible to let us reach our goal towards *spatiotemporal* control and spectroscopy

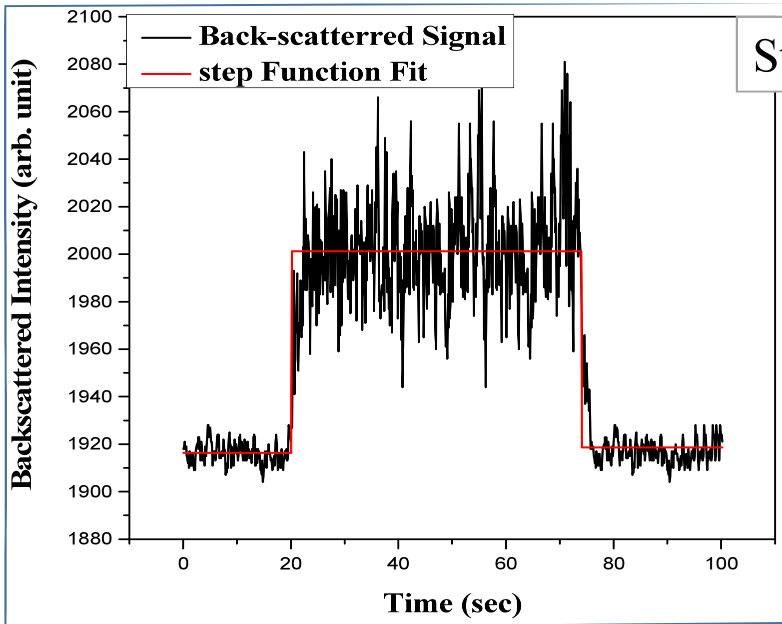
Spatial Trapping: Optical trapping—towards trapping of single macromolecules

Trapping of Rayleigh (λ»d) particles

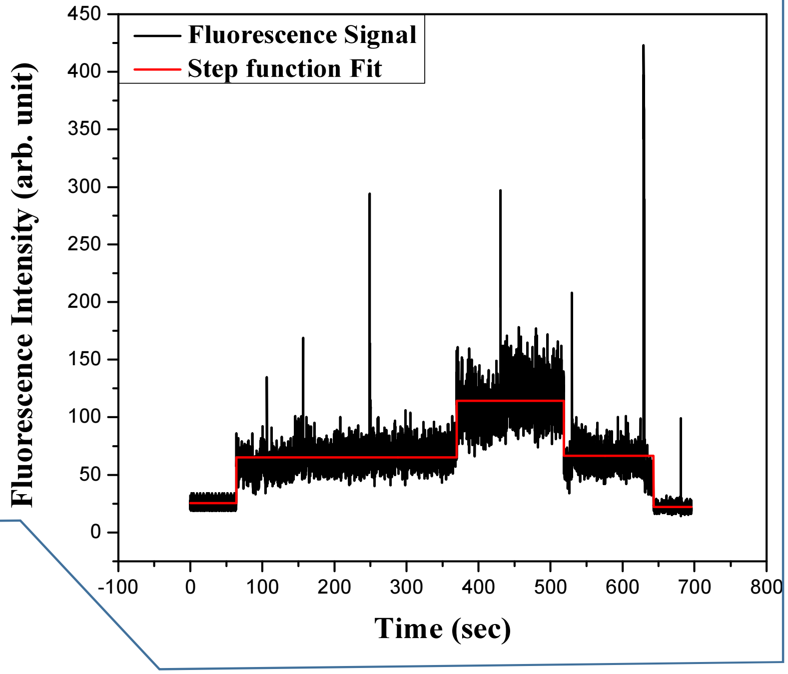
- force depends on polarizability: e.g., latex nano-particles are hard to trap
- high peak power of an ultra-short pulse but '**Repetition Rate is Critical**'
- requires high repetition rate of the pulses

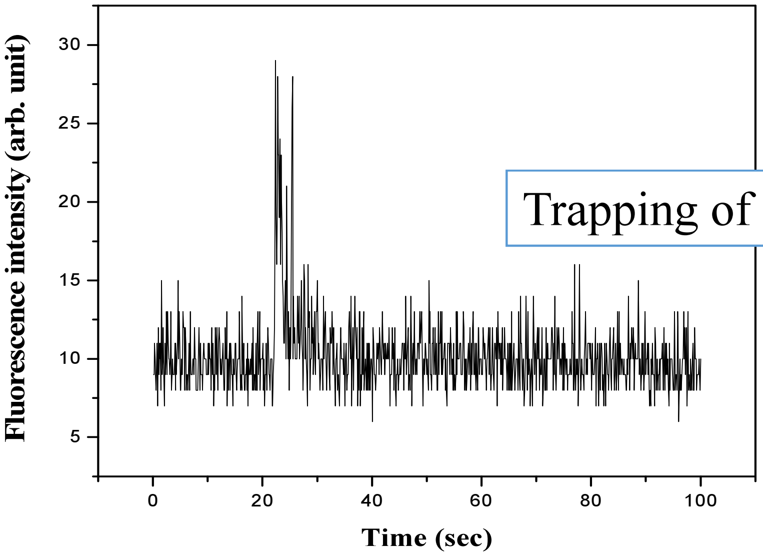


➢A. K. De, D. Roy, A. Dutta, D. Goswami, *Appl. Opt.*, **48**, G33 (2009)



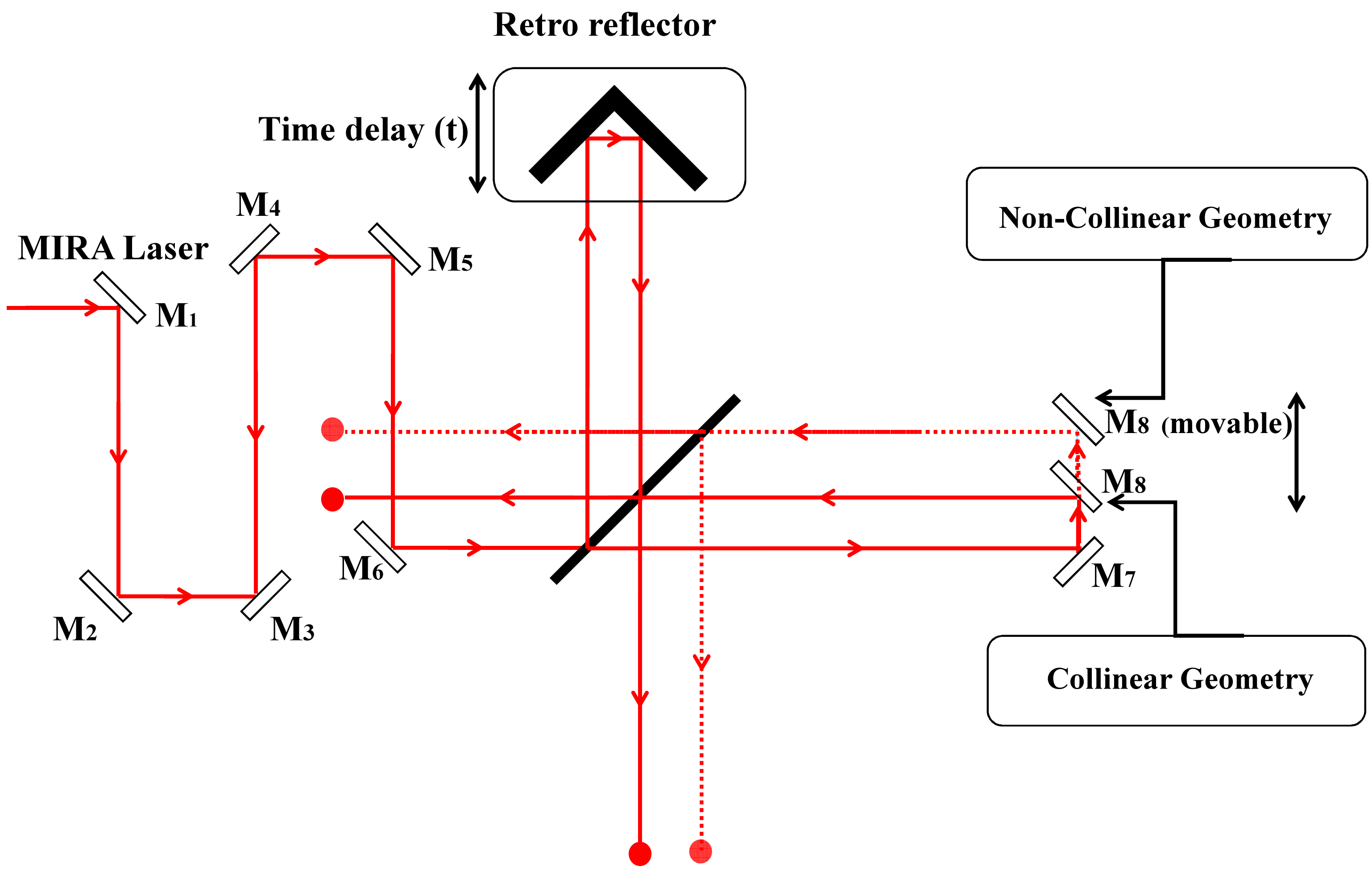
Stable trapping of 100 nm latex beads



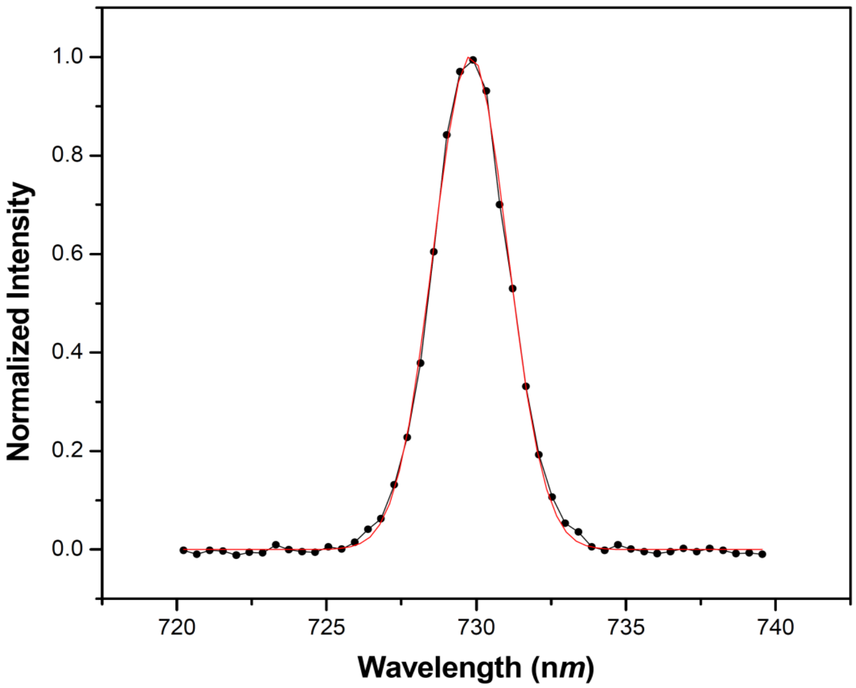


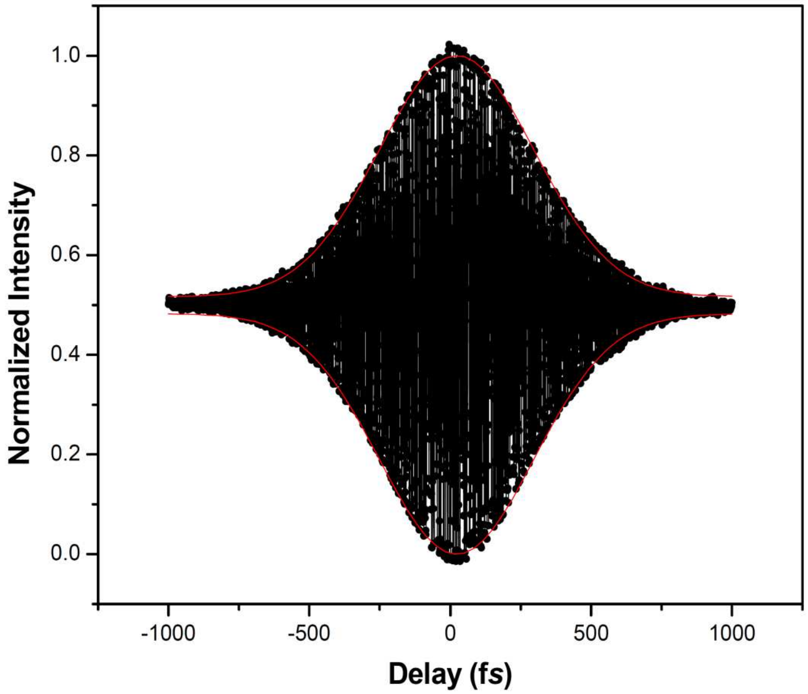
Trapping of 16 nm Q-dot

Capabilities of Single Beam Femtosecond Optical trap


Retro reflector
Time delay (t)
M4
MIRA Laser
M5
M1
Non-Collinear Geometry
M8 (movable)
M8
M6
M7
M2
M3
Collinear Geometry

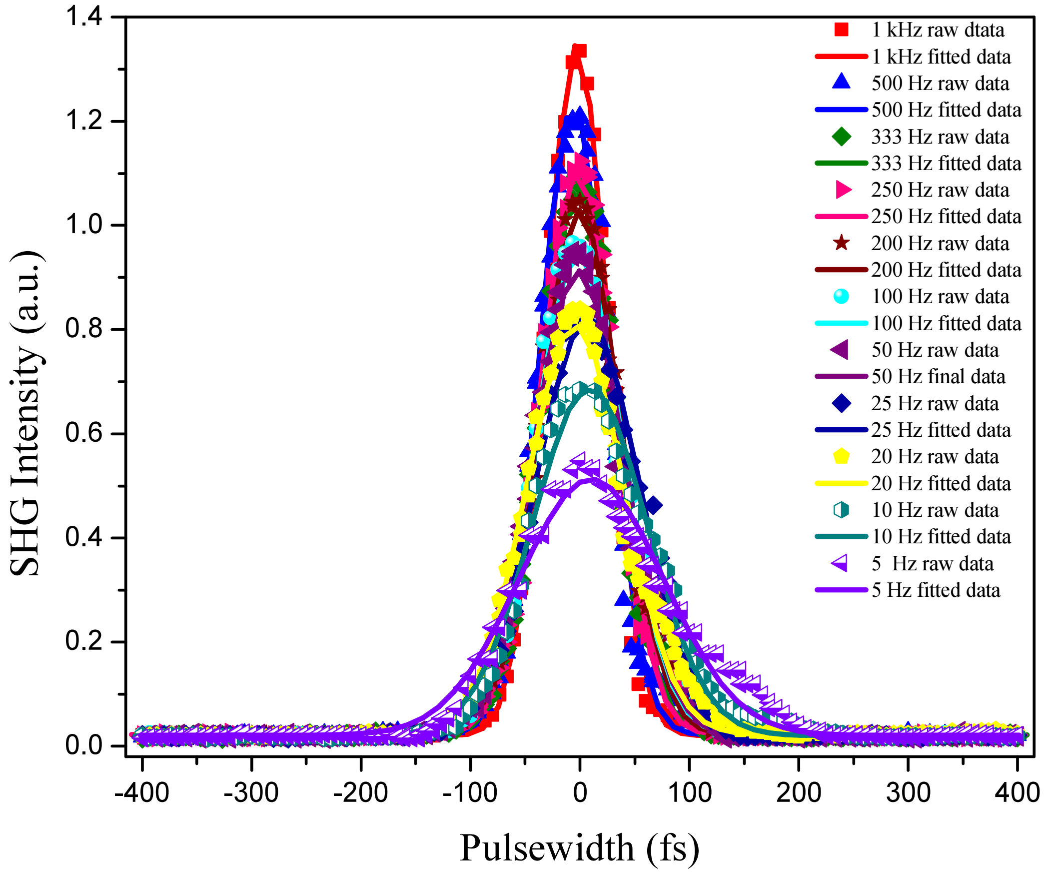
Laser Pulse Profile





- Laser central wavelength ~730 nm, Pulse width: ~180 fs

Pulse Characterization Under Different Repetition rate



Laser repetition rate (Hz)	Pulse width (fs)
1000	47
500	52
333	58
250	59
200	62
100	67
50	69
25	80
20	81
10	88
5	111

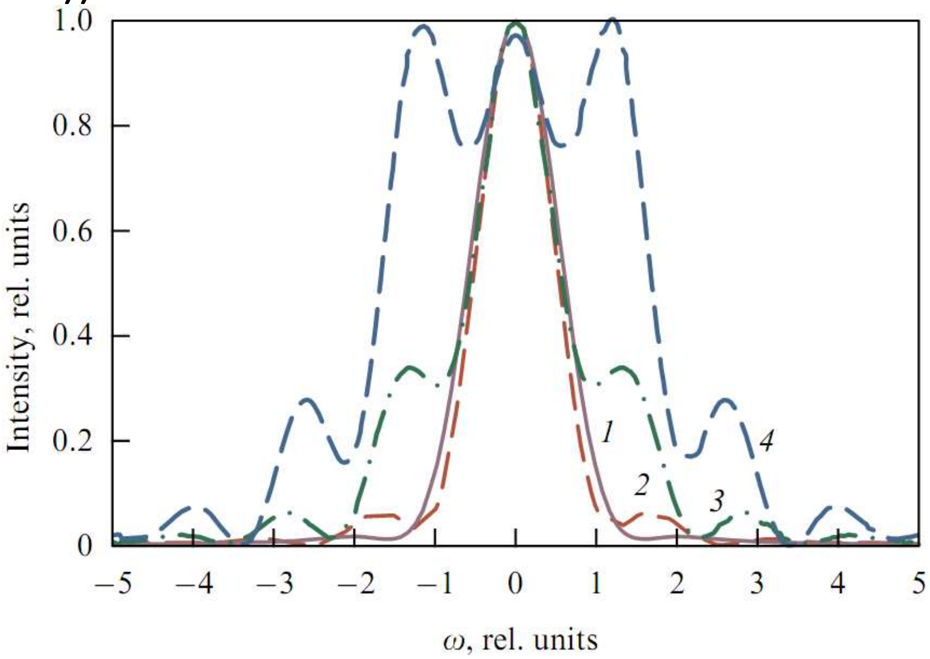
Supercontinuum

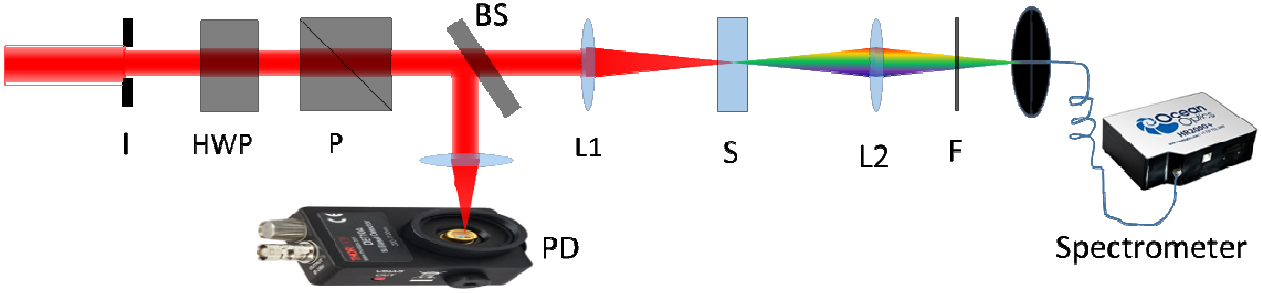
- Spectacular nonlinear phenomenon (true for every transparent dielectrics).
- Wide spectral range, highly coherent, low divergent beam.
- Broadband time resolved spectroscopy, optical parametric amplification, optical pulse compression.
- Generated through pulse self actions (self-, cross-phase modulation, four-wave mixing, filamentation due to multi-photon excitation (MPE)).

Self-phase modulation

$$\varphi_{NL}(\tau) = \int_0^l n_2 I(z,\tau) \frac{\omega_0}{c} dz$$

- Intensity dependent Kerr-nonlinear effect.
- Symmetric broadening, modulation of spectra.

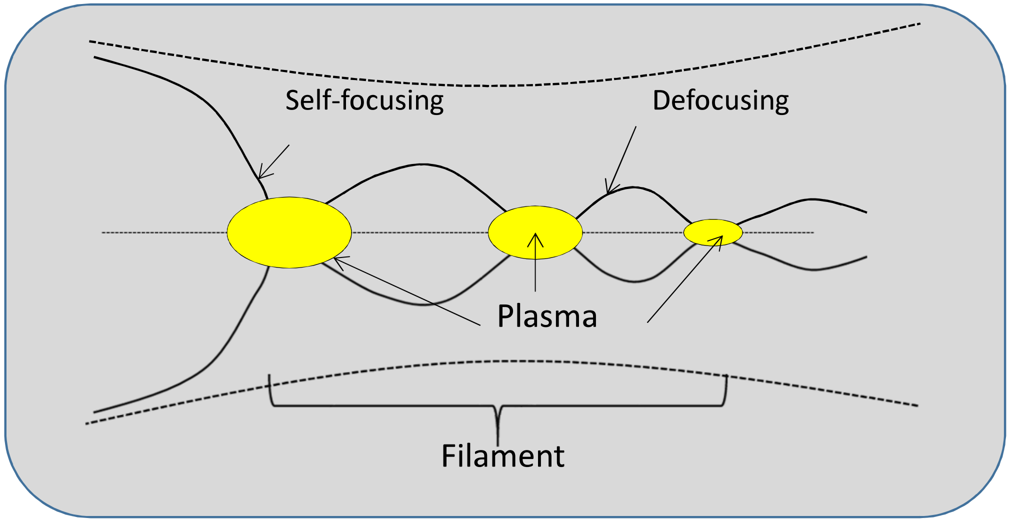


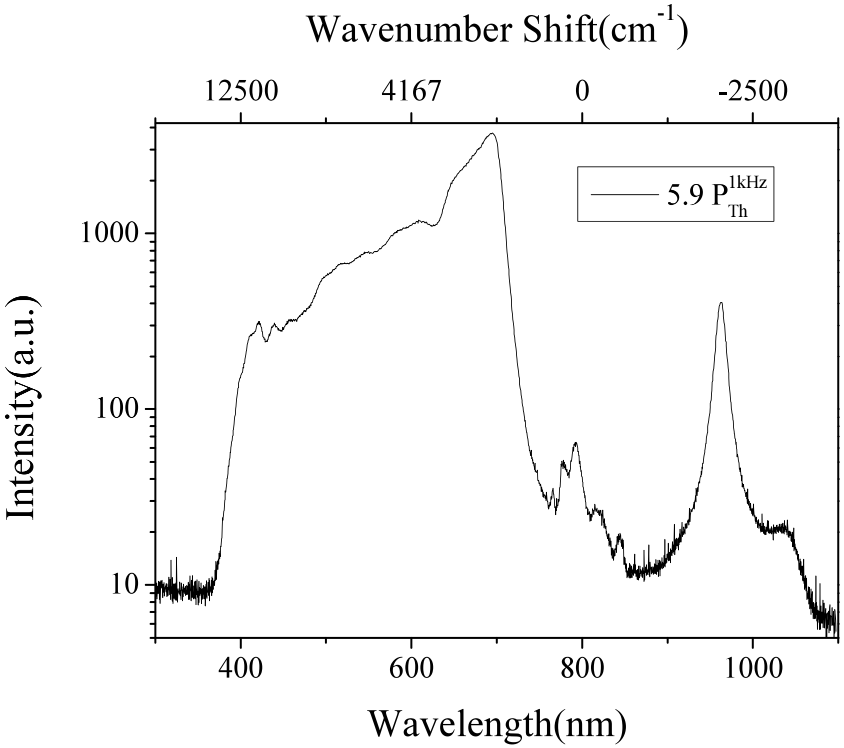


I=iris; HWP=half-waveplate; P=polariser; L1, L2=lens; S=sample cuvette; F=filter

Filamentation

- Self effect of LASER pulse.
- Mainly due to Self-focusing and defocusing owing to free electron generation via MPE.





Change of nonlinear refractive index in the medium

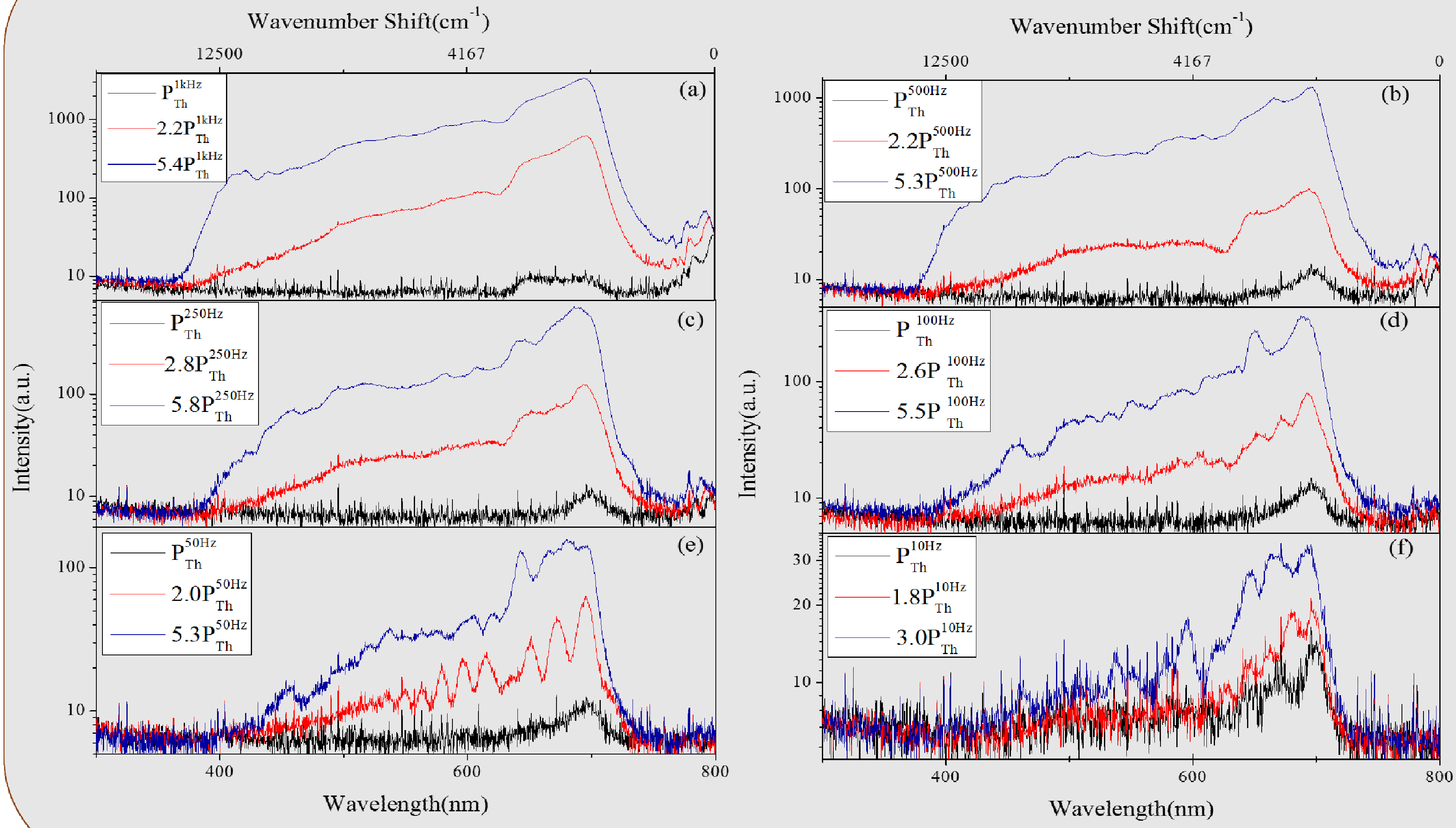
$$\Delta n \approx n_2 I(\boldsymbol{r}, t) - \frac{2\pi e^2 N_e(\boldsymbol{r}, t)}{m_e \omega_0^2}$$

Spectral broadening due to self-focusing and filamentation

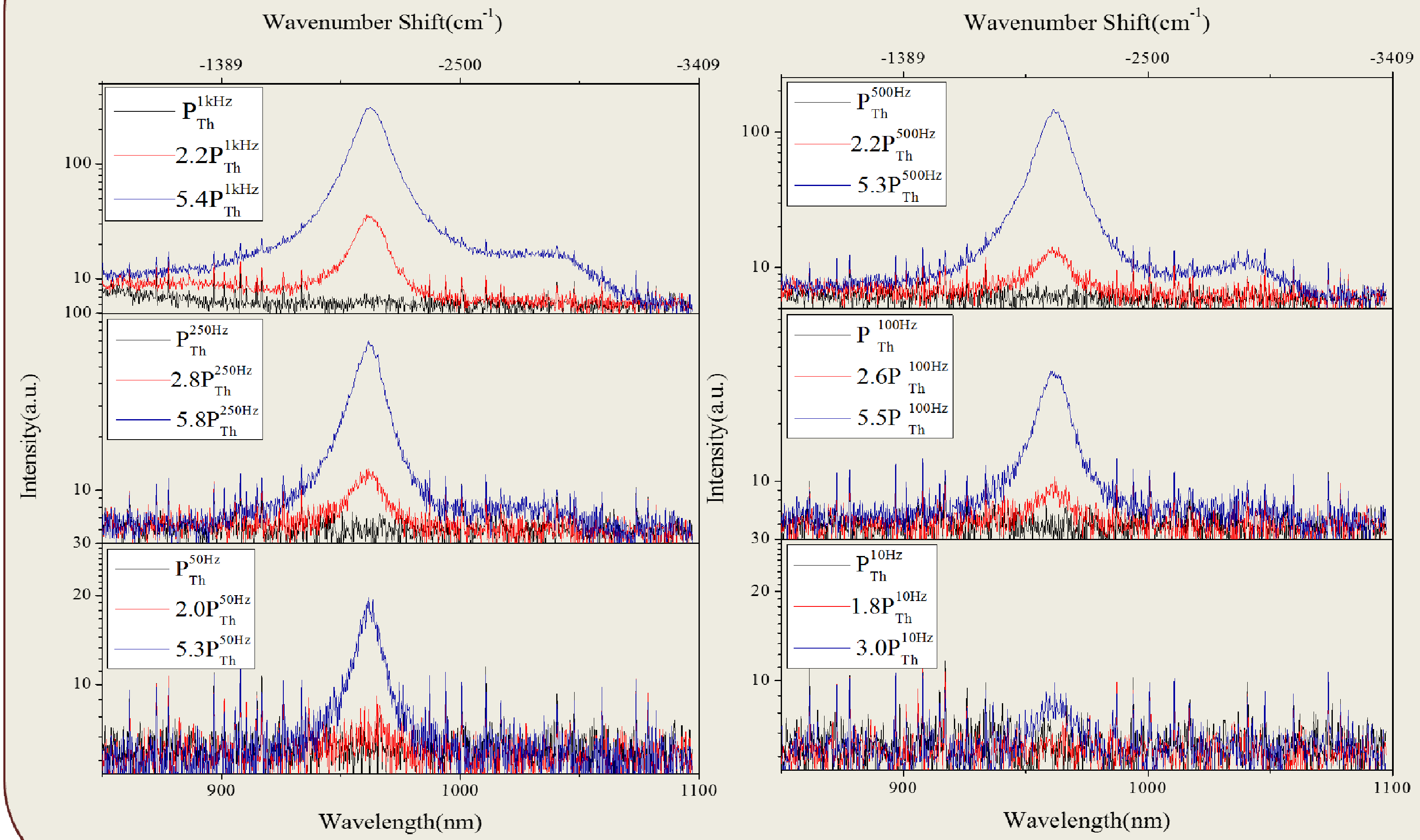
$$\Delta\omega = -\frac{\partial \Delta\varphi}{\partial t} = -\frac{\omega_0 n_2 z}{c}\frac{\partial I(\boldsymbol{r}, t)}{\partial t} + \frac{2\pi e^2 z}{m_0 \omega_0 c}\frac{\partial N_e(\boldsymbol{r}, t)}{\partial t}$$

Inhomogeneity in n_2 influences the group velocity of the propagating pulse. This nonlinearity causes the group velocity of edges of the pulse larger than the group velocity of peak thus creates a spike in intensity envelop.

Structured supercontinuum spectrum due to interference between filaments

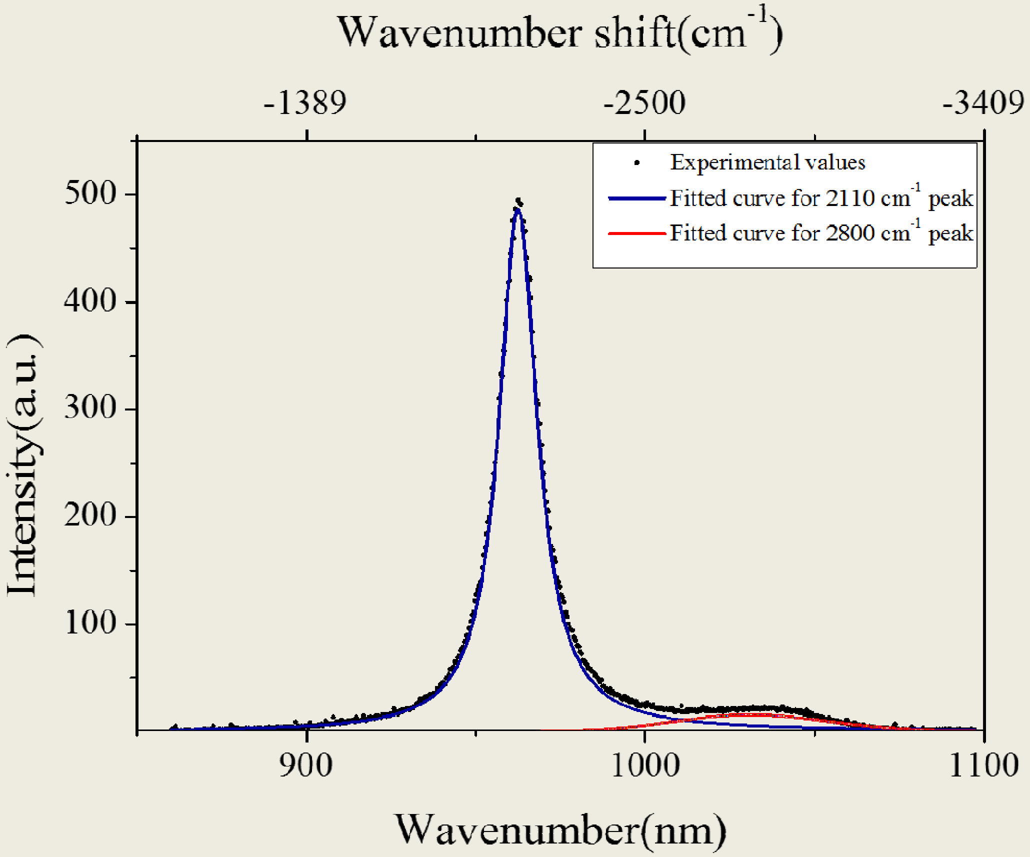


Combination band of water



Stokes region of the supercontinuum of water

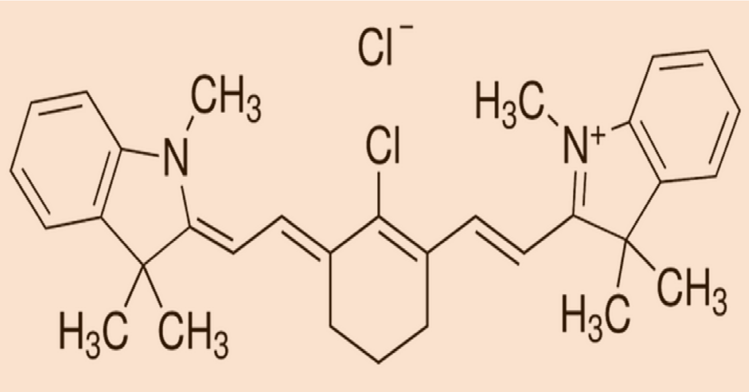
Explaining the Novel Underlying Interference Structures in the Supercontinuum Spectrum of Water



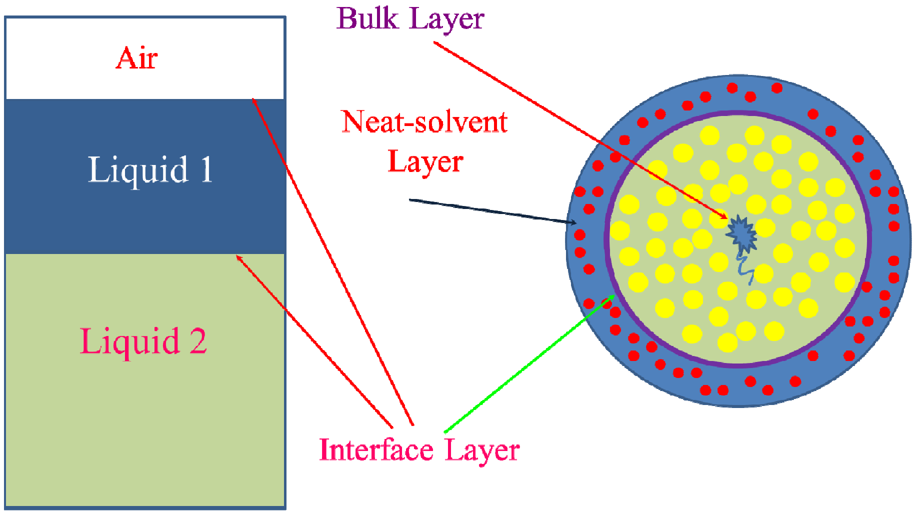
- $2110cm^{-1}$ peak due to combination of scissoring and liberation mode.
- $2800cm^{-1}$ peak due to $H_{2n}O_n^+$ and appears after intensity clamping.
- At threshold significant amount of H-bond left.
- At high intensity more water molecules cresses the chaotic region and forms significant amount of $H_{2n}O_n^+$ species.

Use of a Dye Molecule

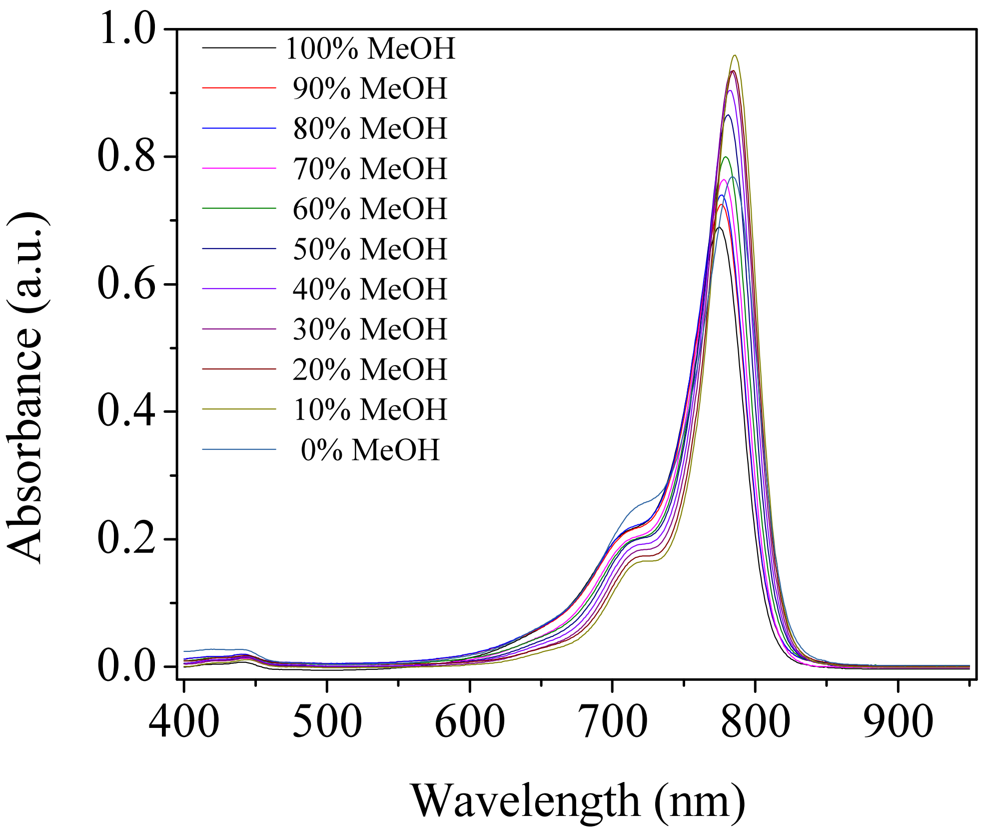
Structure of IR775Molecule

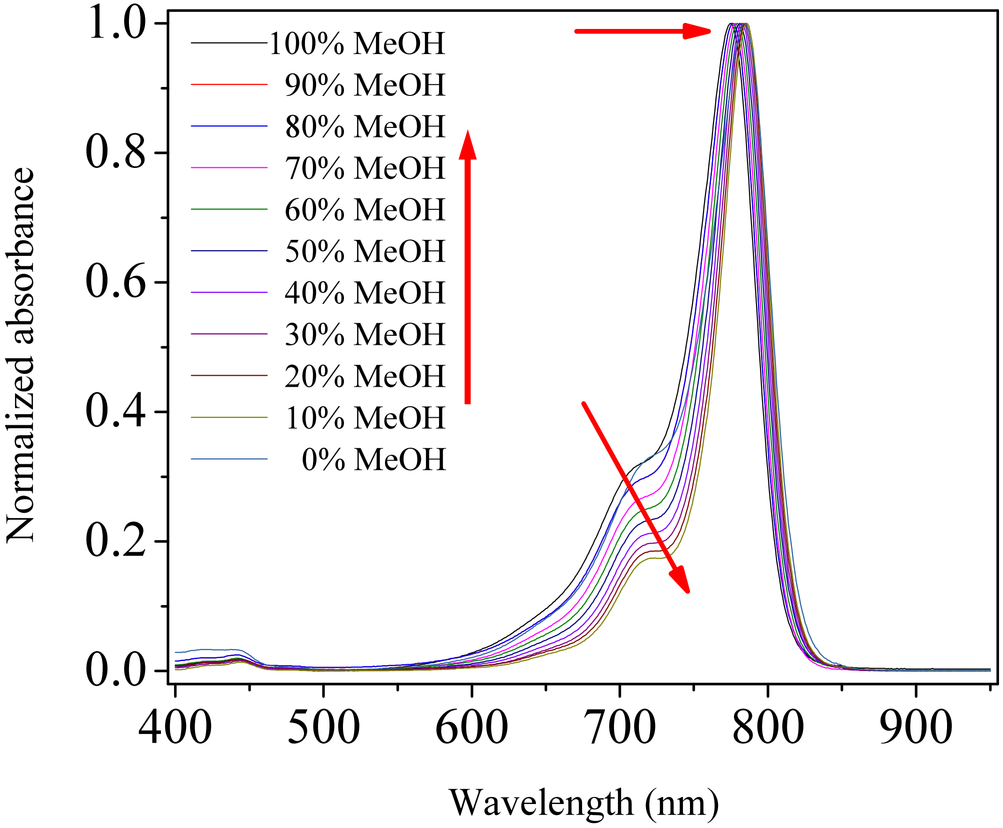


IR775

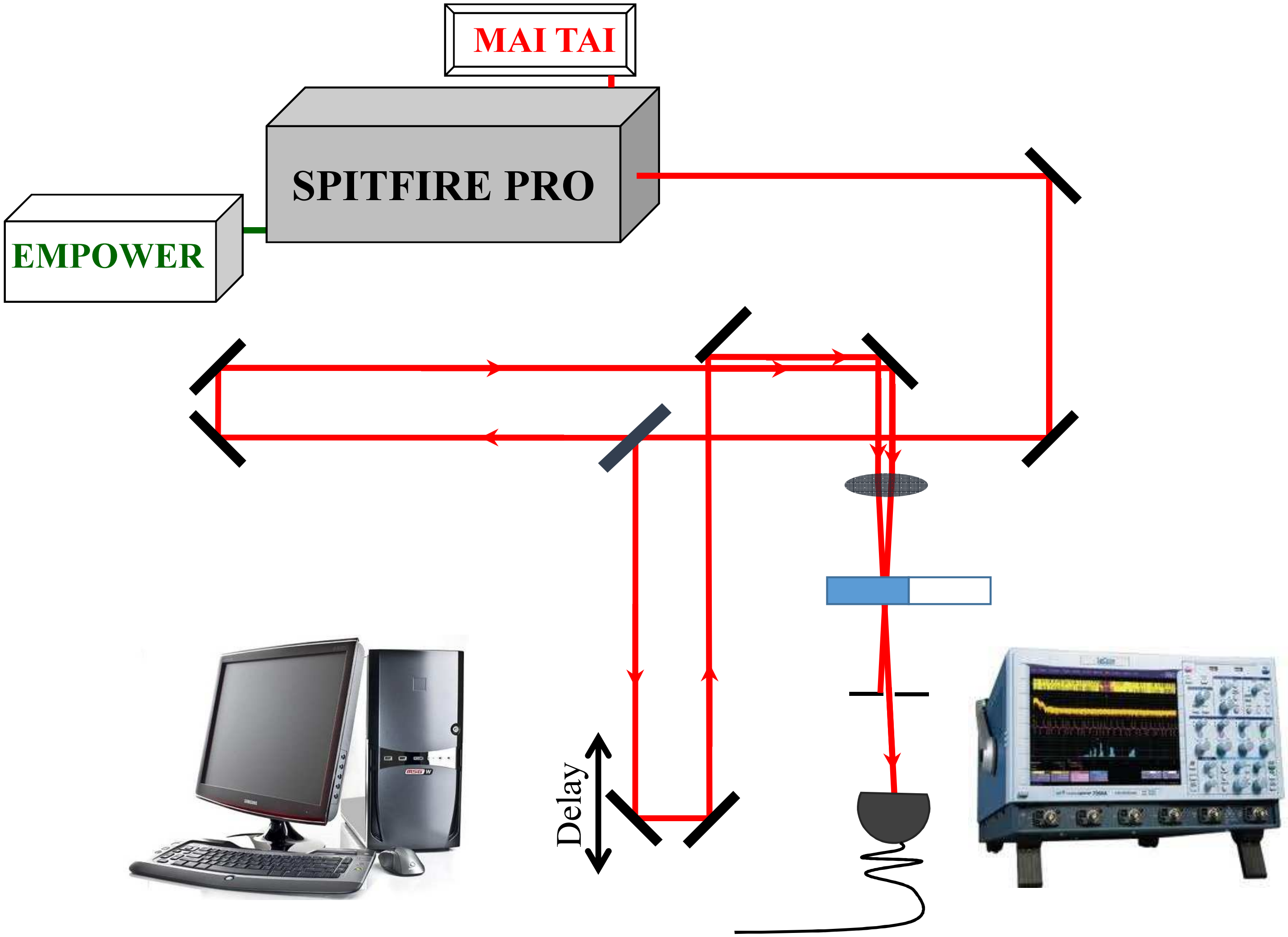


Fluorescence spectra of IR775 in the binary mixture of MeOH-$CHCl_3$

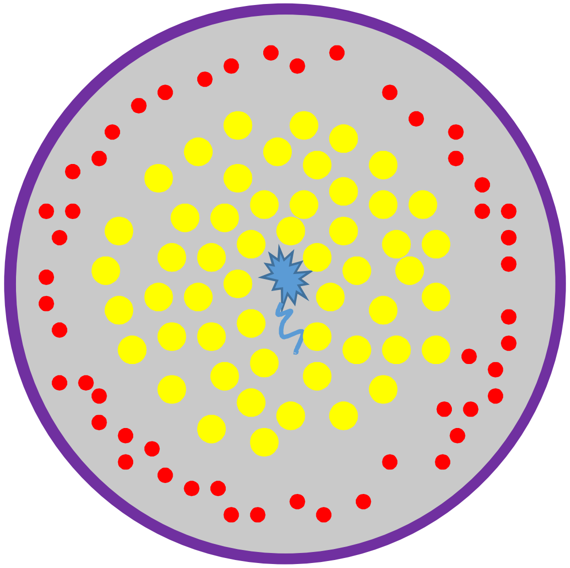


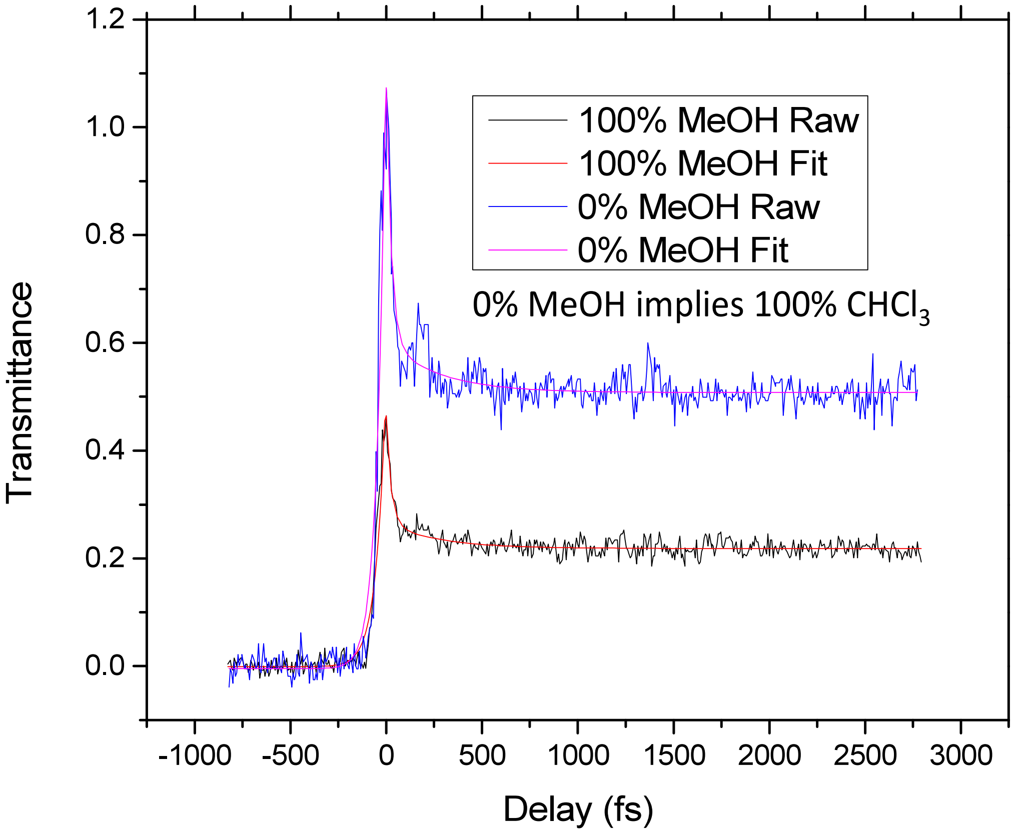


Schematic Experimental Setup



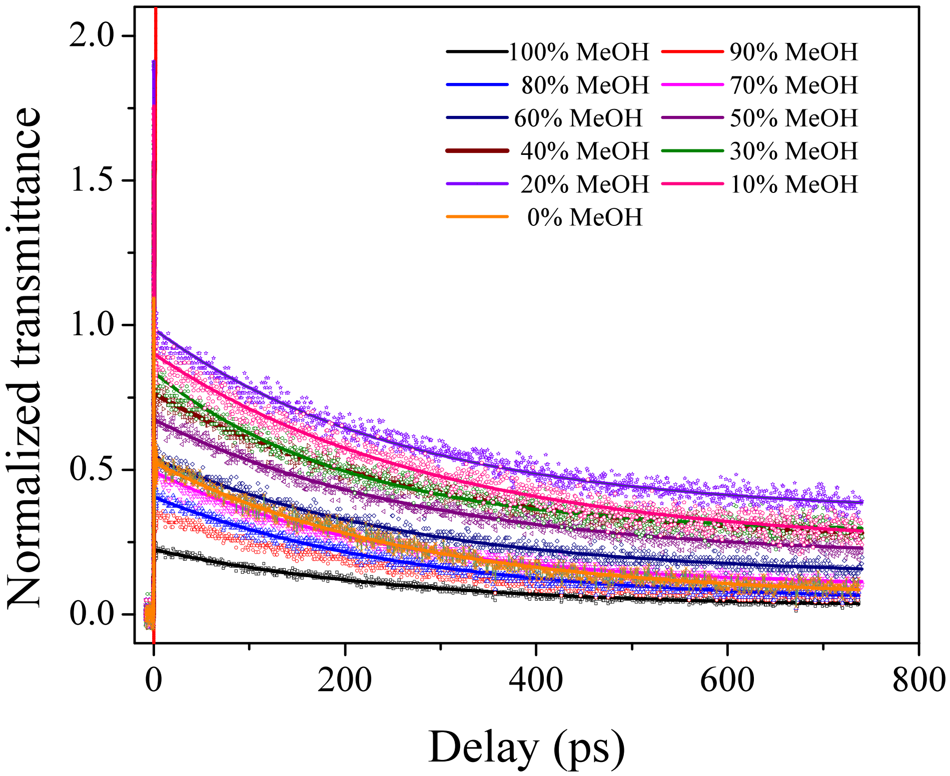
Experimental Data

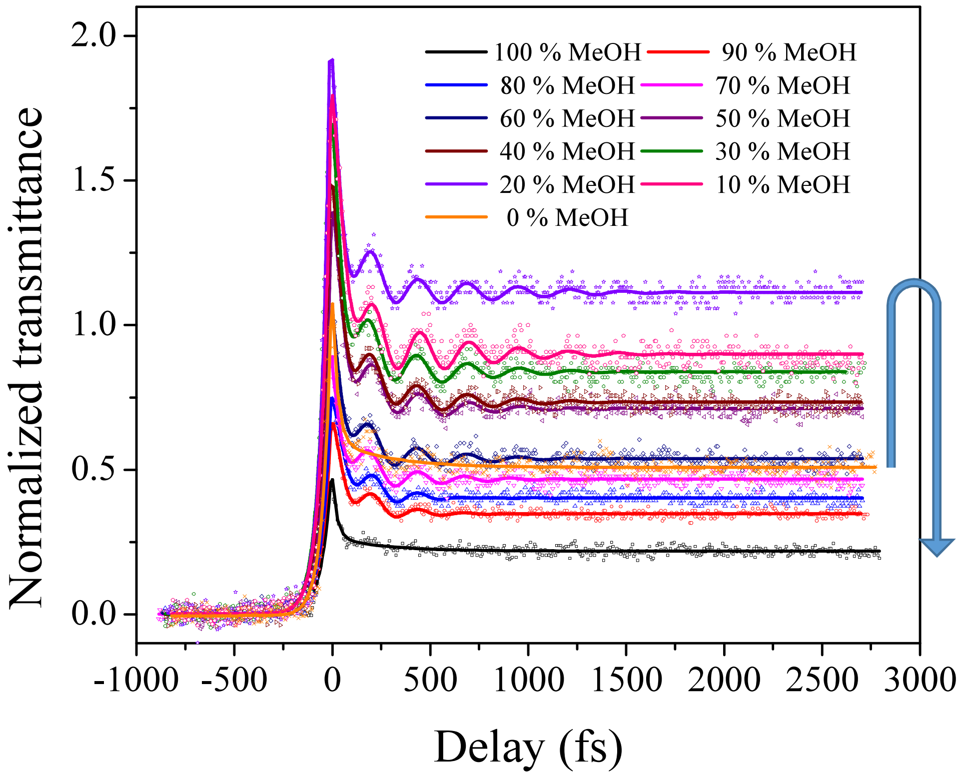




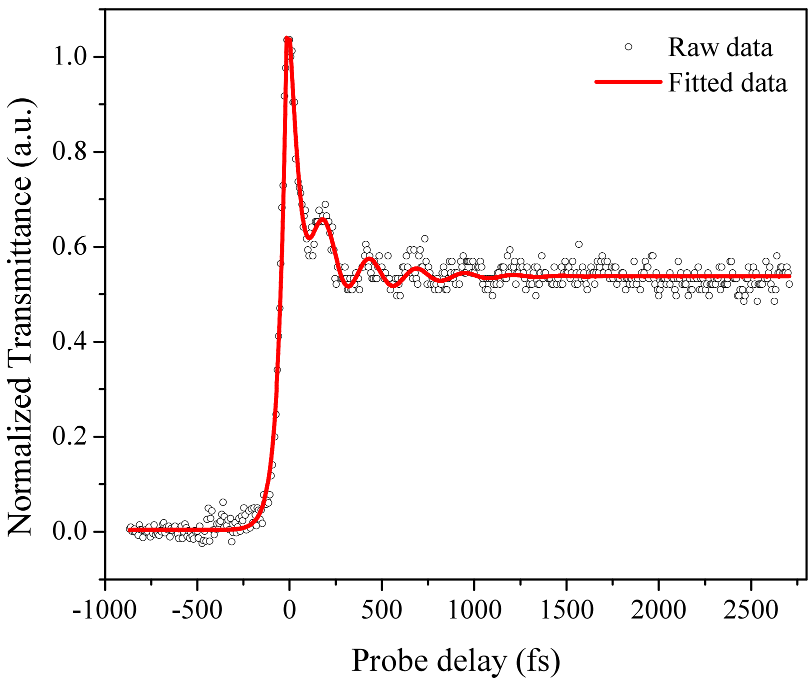
0% MeOH implies 100% $CHCl_3$

Decay of the transient absorption of IR775 in the binary mixture of MeOH-$CHCl_3$

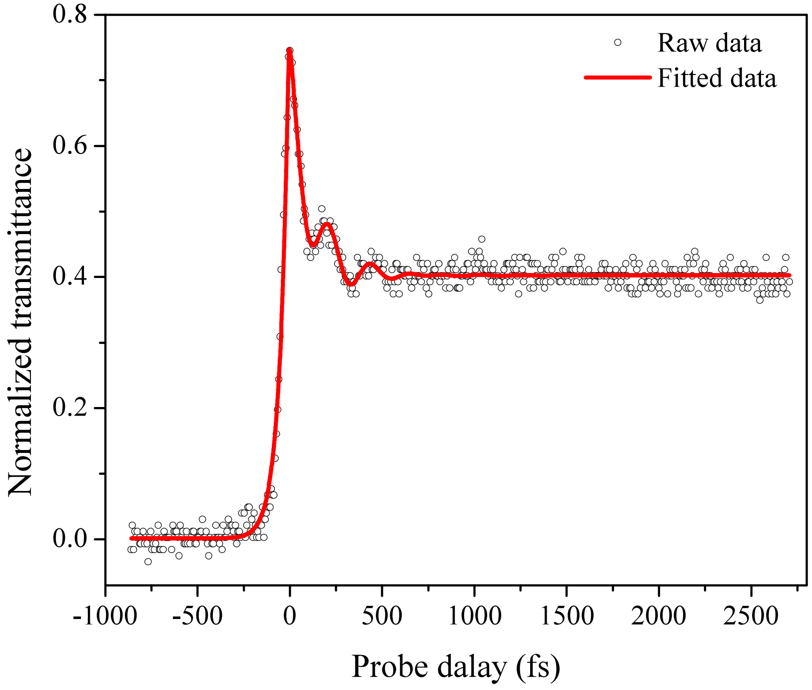




Time-resolved pump-probe dynamics of IR775 dye in binary mixture of methanol & chloroform



60% MeOH



80% MeOH

Microheterogeneity

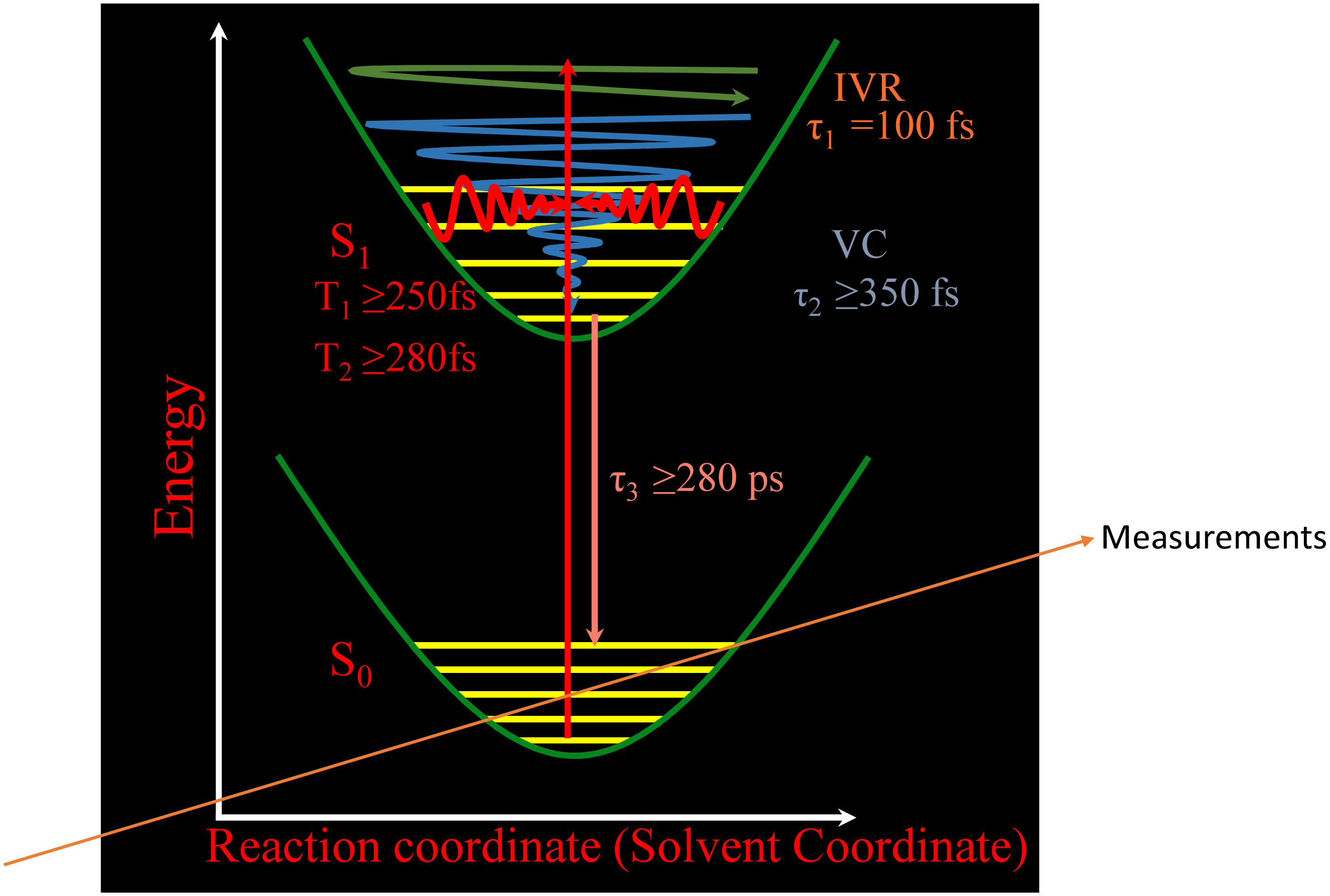
Contd…

$$f(t) = a_1 e^{-t/\tau_1} + a_3 e^{-t/\tau_3} + a_2 e^{-t/\tau_2}\left(A_1 \cos\left[\frac{2\pi t}{T_1} + \phi_1\right] + A_2 \cos\left[\frac{2\pi t}{T_2} + \phi_2\right]\right) + c$$

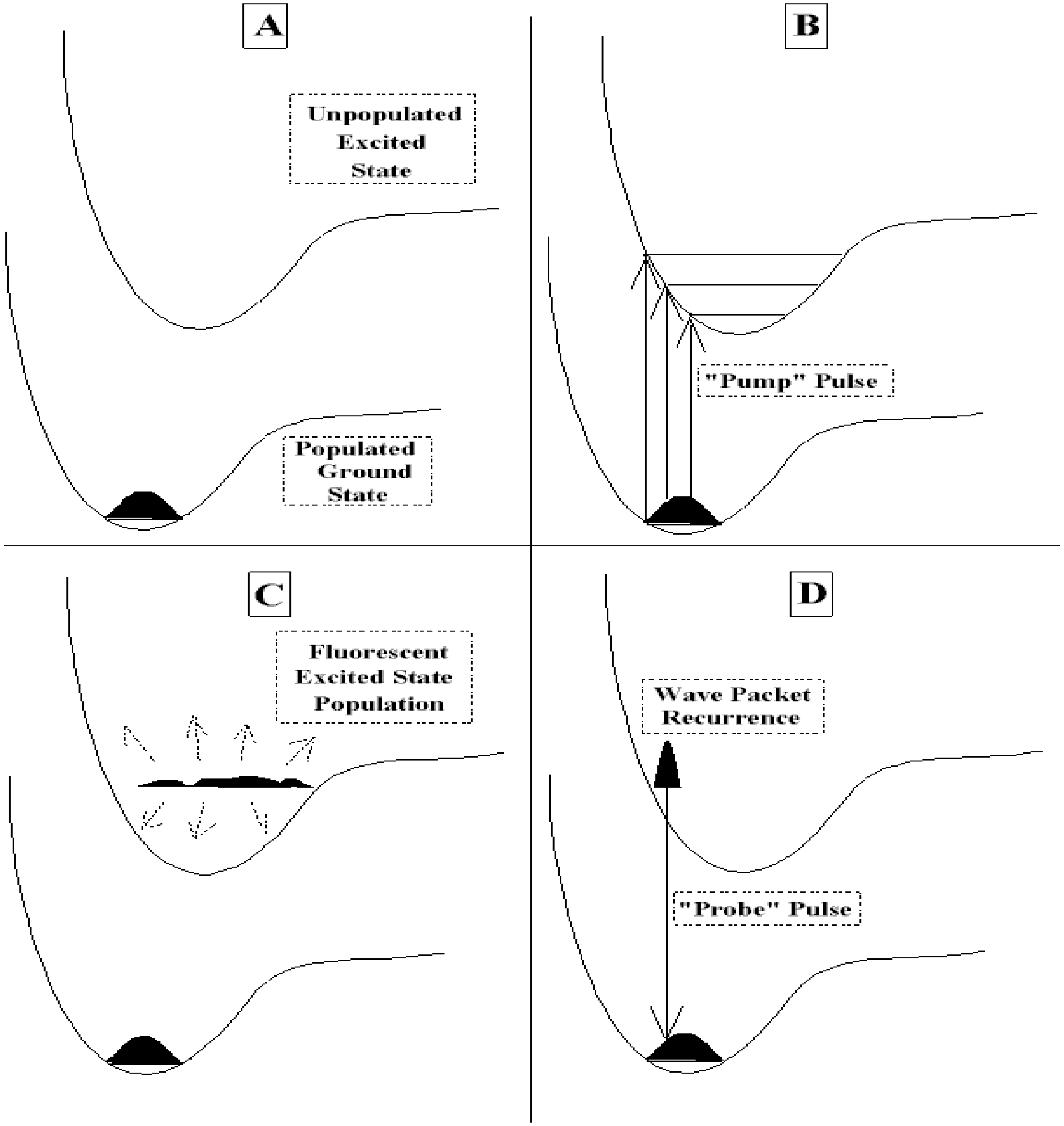
% methanol in IR775 in the binary mixture of methanol and chloroform	τ_1 (fs)	τ_2 (fs)	T_1 (fs)	T_2 (fs)	τ_3 (ps)
80	**101**	**219**	**270**	**205**	**271**
70	**97**	**215**	**270**	**281**	**245**
60	**101**	**219**	**261**	**270**	**272**
50	**124**	**205**	**262**	**270**	**302**
40	**123**	**220**	**264**	**271**	**274**
30	**100**	**230**	**265**	**277**	**265**
20	**100**	**300**	**260**	**282**	**283**
10	**100**	**290**	**260**	**280**	**280**

Excited state decay time constants of the binary mixture of MeOH-$CHCl_3$ in IR775

Contd…

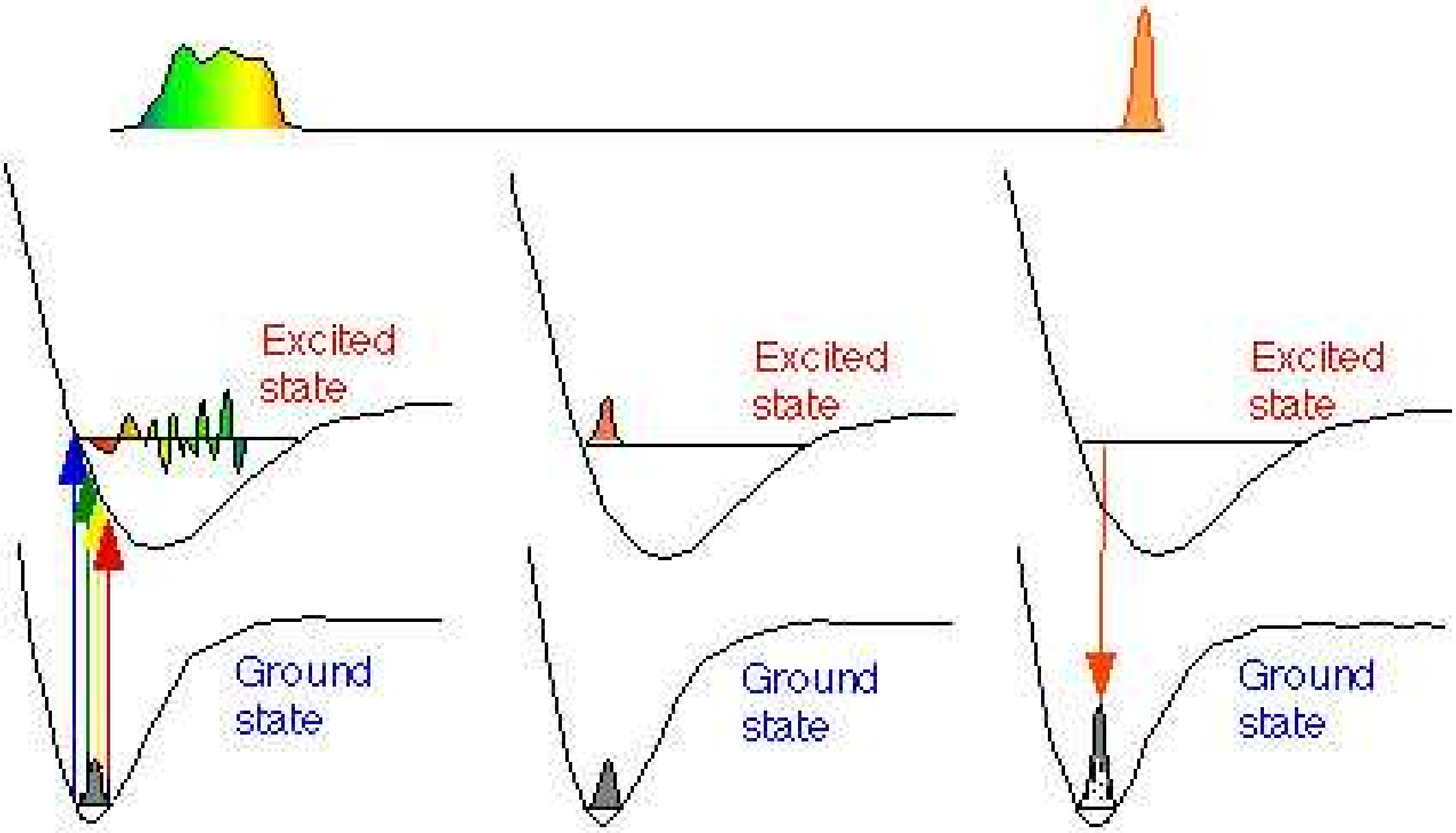


Schematic representation of the excited state dynamics


A
Unpopulated Excited State
Populated Ground State
B
"Pump" Pulse
C
Fluorescent Excited State Population
D
Wave Packet Recurrence
"Probe" Pulse

JCP **112, 5081 (2000)**

Using Shaped Pulses to Measure Molecular Potentials



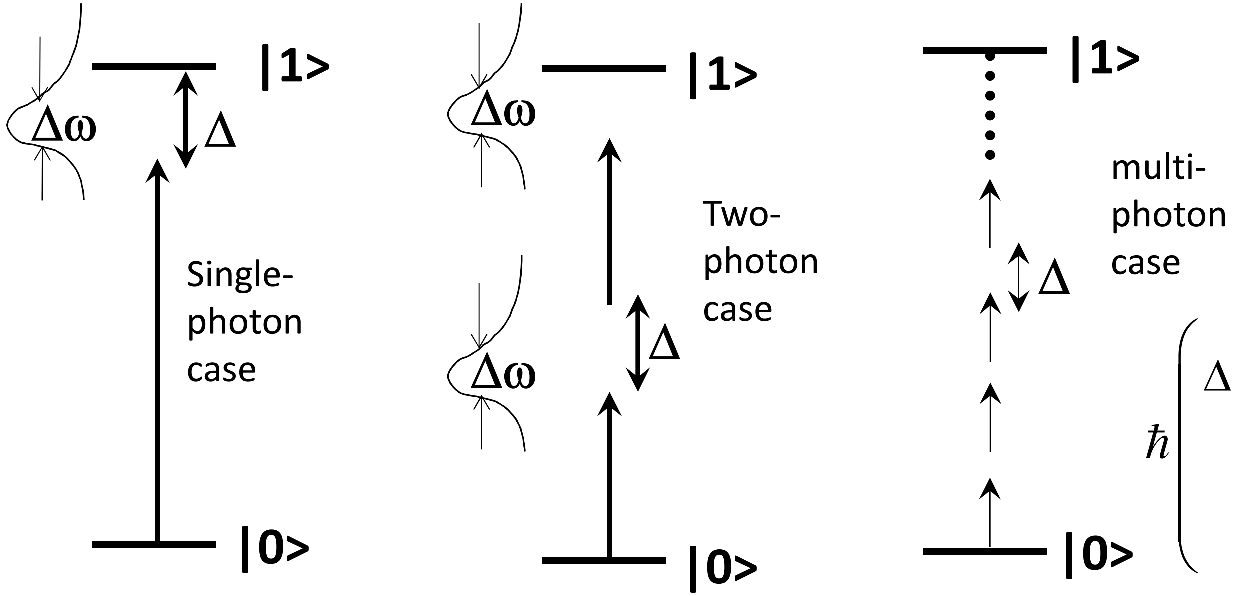
Chirped excitation pulse
(blue before red)

t=1 ps

Sharply refocused
wavepacket
(if chirp is correct)

t=1 ps

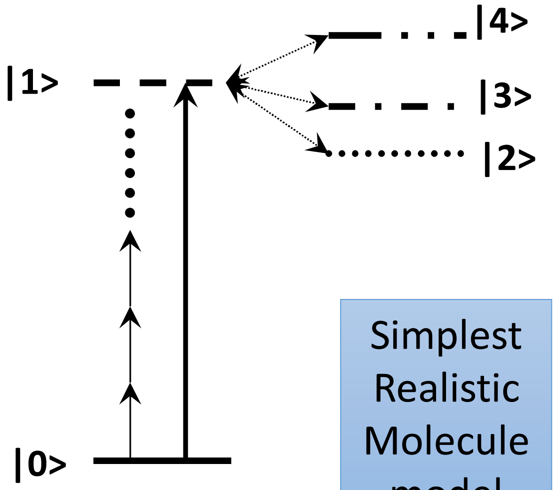
Ultrafast dump
pulse



Ideal 2-Level model

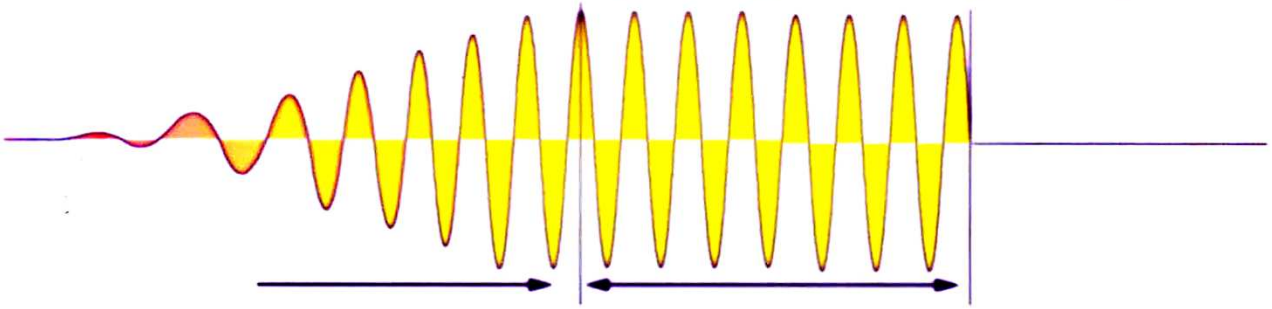
$$\hbar\begin{pmatrix} \Delta + N\dot{\phi}(t)) & \frac{\Omega_1}{2} \\ \frac{\Omega_1^*}{2} & 0 \end{pmatrix}$$

$$\begin{array}{cc} & \begin{array}{cccccc} |0\rangle & |1\rangle & |2\rangle & |3\rangle & |4\rangle & \cdots \end{array} \\ \hbar & \begin{pmatrix} 0 & \Omega_1(t) & 0 & 0 & 0 & \cdots \\ \Omega_1(t) & \delta_1(t) & V_{12} & V_{12} & V_{12} & \cdots \\ 0 & V_{12} & \delta_2(t) & V_{12} & V_{12} & \cdots \\ 0 & V_{12} & V_{12} & \delta_3(t) & V_{12} & \cdots \\ 0 & V_{12} & V_{12} & V_{12} & \delta_4(t) & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \end{pmatrix} \end{array}$$



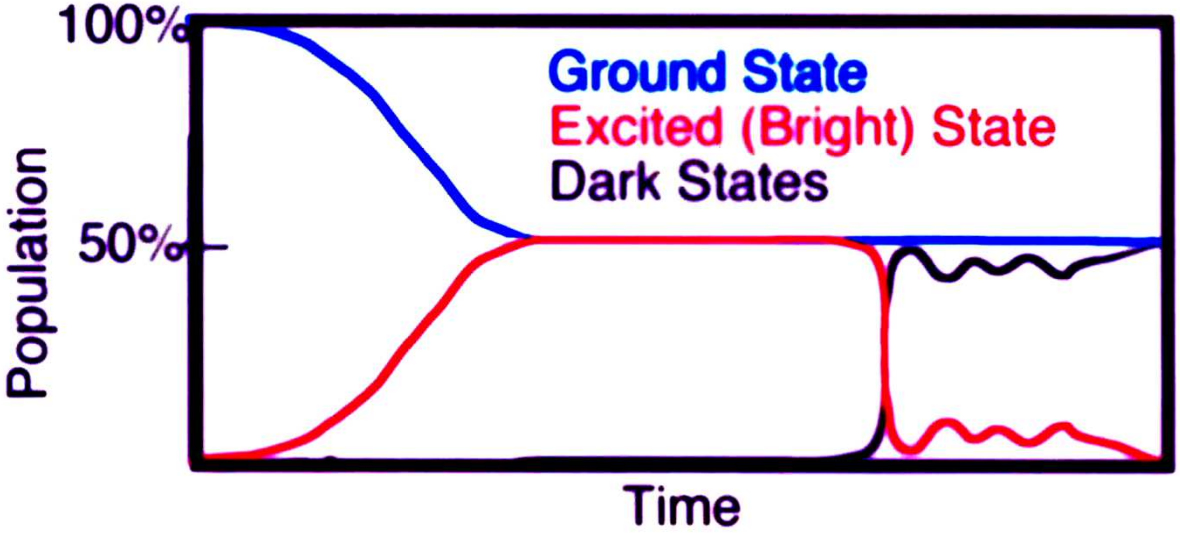
Simplest Realistic Molecule model

Adiabatic half passage in coupled systems:



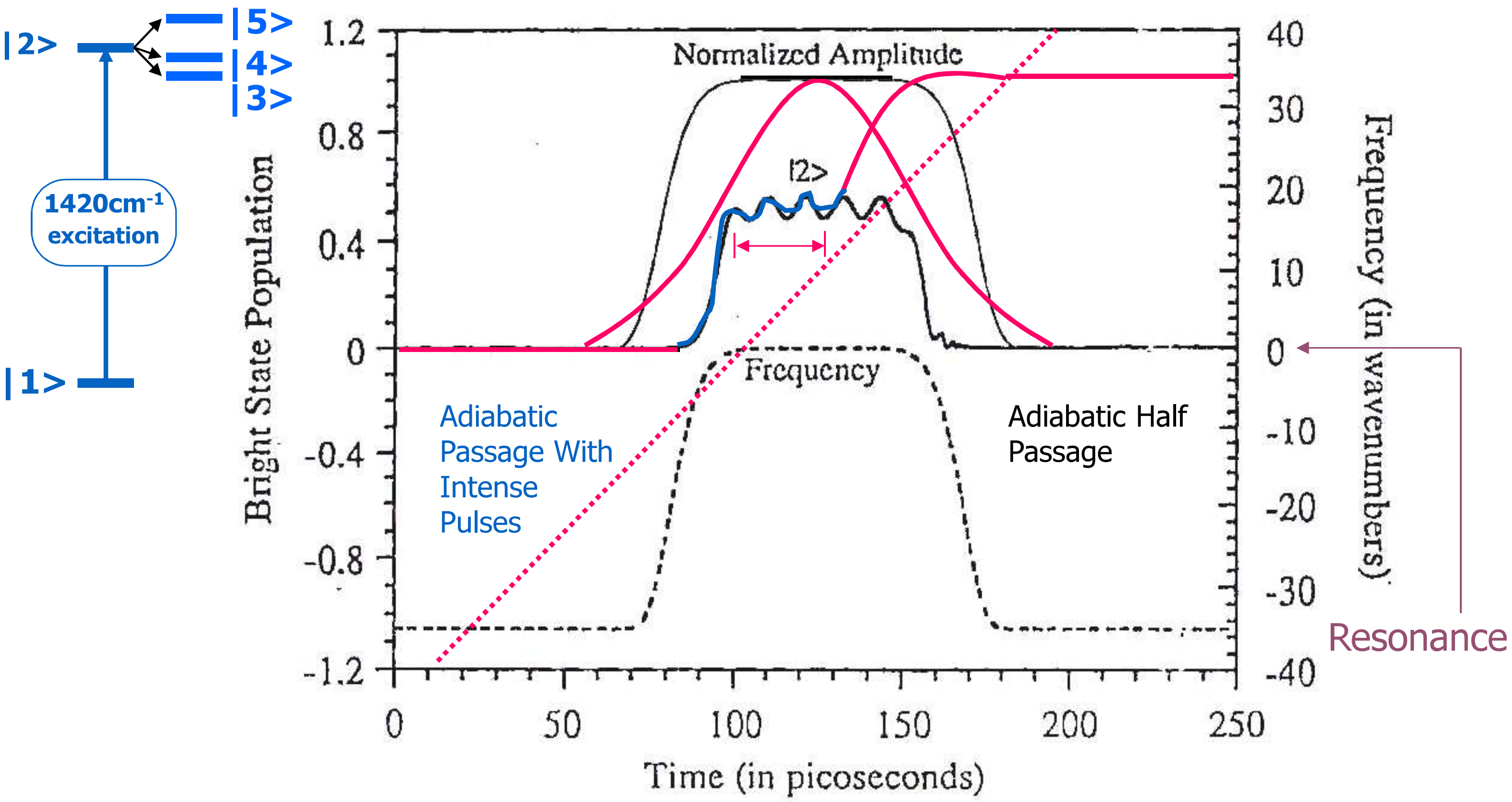
t<0: sweep to resonance

0<t<T: constant amplitude, $\mu \cdot E/\hbar \gg$ couplings to dark states

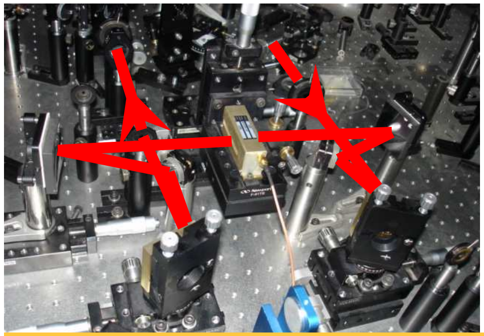


Model Calculations with Shaped Pulses

Anthracene



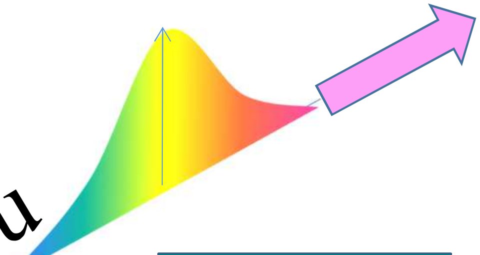
Phys. Rev. Lett. 88, 177901 (2002); J. Chem. Phys. 127, 124305 (2007)

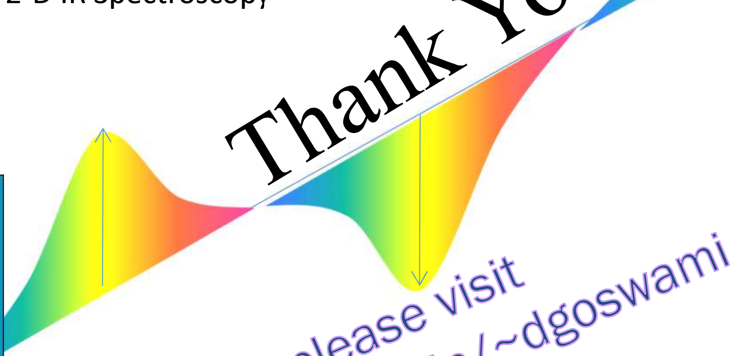


Femtosecond Pulse Shaper

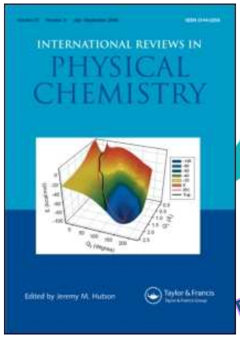
- Coherent Control
- Bioimaging
  - **Multiphoton Imaging**
  - **Optical Tweezers**
- 2-D IR Spectroscopy

Thank You





Measurement
of Nonlinearities



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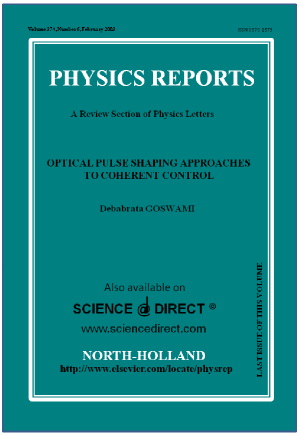

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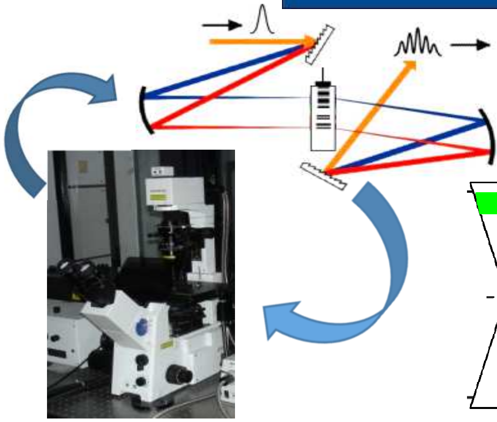

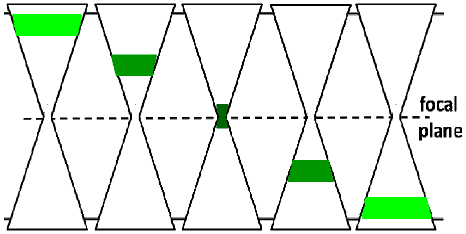

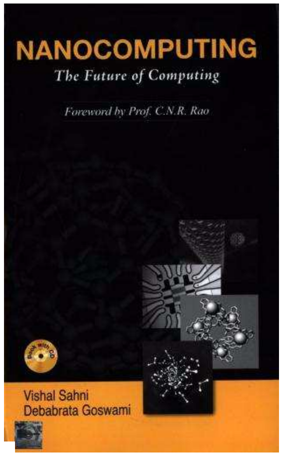

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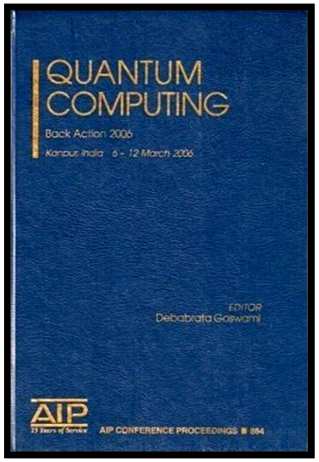
Vishal Sahni
Debabrata Goswami



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