IFSC-USP Fotônica

UNIVERSIDADE DE SÃO PAULO
Instituto de Física de São Carlos

Coherent control of light matter interaction

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USP

ultrashort laser pulses

Mode-locking

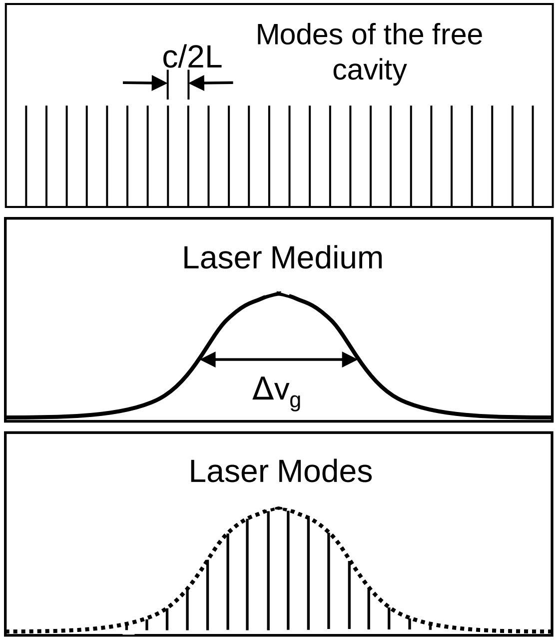
$$E_n(t) = E_n \cdot \exp\left[i\left(2\pi\nu_n t + \phi_n\right)\right] \qquad n = 1,2,3,\ldots$$

$$\nu_n = n\frac{c}{2L} \qquad \nu_c \equiv \frac{c}{2L} \Rightarrow f_{\text{Laser System}}$$

$$N \approx \Delta\nu_g / \nu_c$$

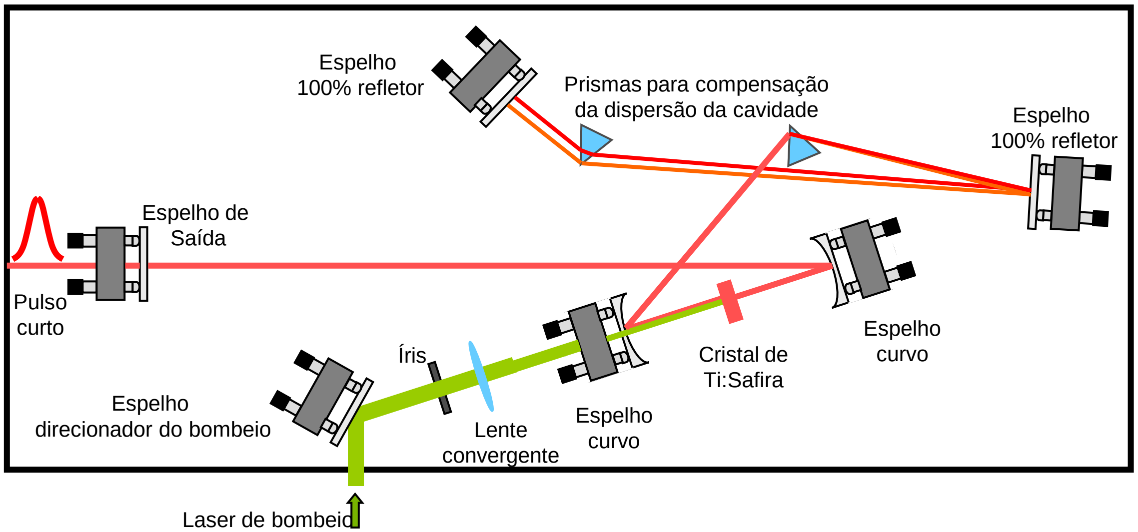
$$\text{Femtosecond lasers}: \; N \approx 10^5 - 10^6$$

$$\tau_{pulse} \propto 1/\Delta\nu_g$$

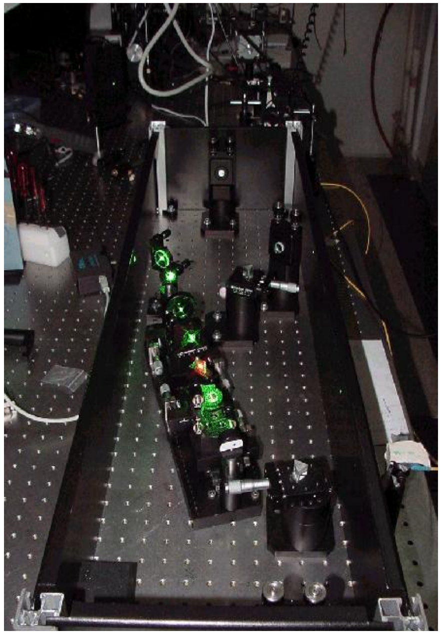


ultrashort laser pulses

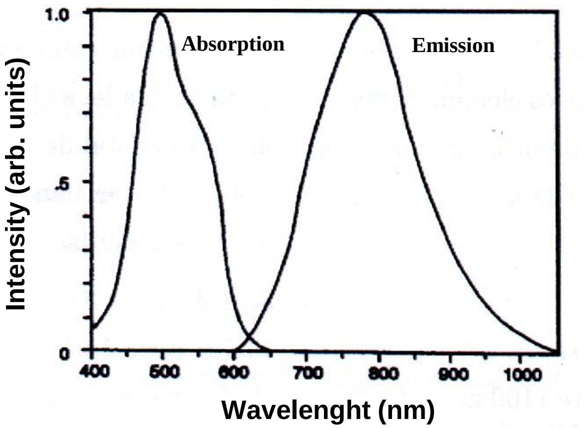
Laser Cavity



Ti:Sapphire laser oscillator



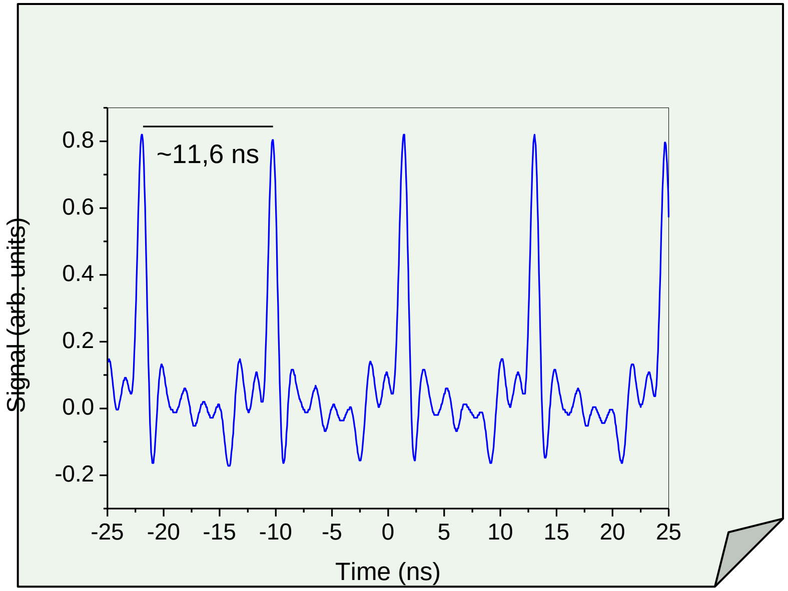
Ti:Sapphire Crystal



$$\tau_{pulse} \propto 1/\Delta\nu_g$$

ultrashort pulses

Femtosecond pulses supplied for the Ti:Sapphire oscilador laser

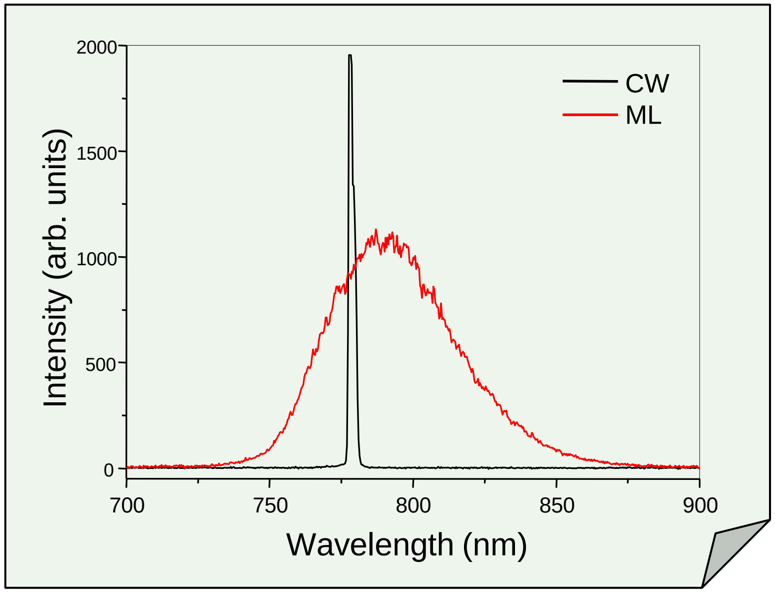


$\Delta t_{Peak} \sim 11{,}6\ \mathrm{ns}$ $\quad \overline{P}_{\mathrm{ML}} \sim 450\ \mathrm{mW}$

$\mathrm{f} \sim 86\ \mathrm{MHz}$ $\quad \overline{\mathrm{E}}_{\mathrm{pulse}} \sim 5\ \mathrm{nJ}$

ultrashort pulses

Emission spectra of the Ti:Sapphire laser oscillator in CW and ML modes



$$\Delta\lambda_p^{FWHM} \sim 55\,\mathrm{nm}$$

$$\lambda_0 \sim 785\,\mathrm{nm}$$

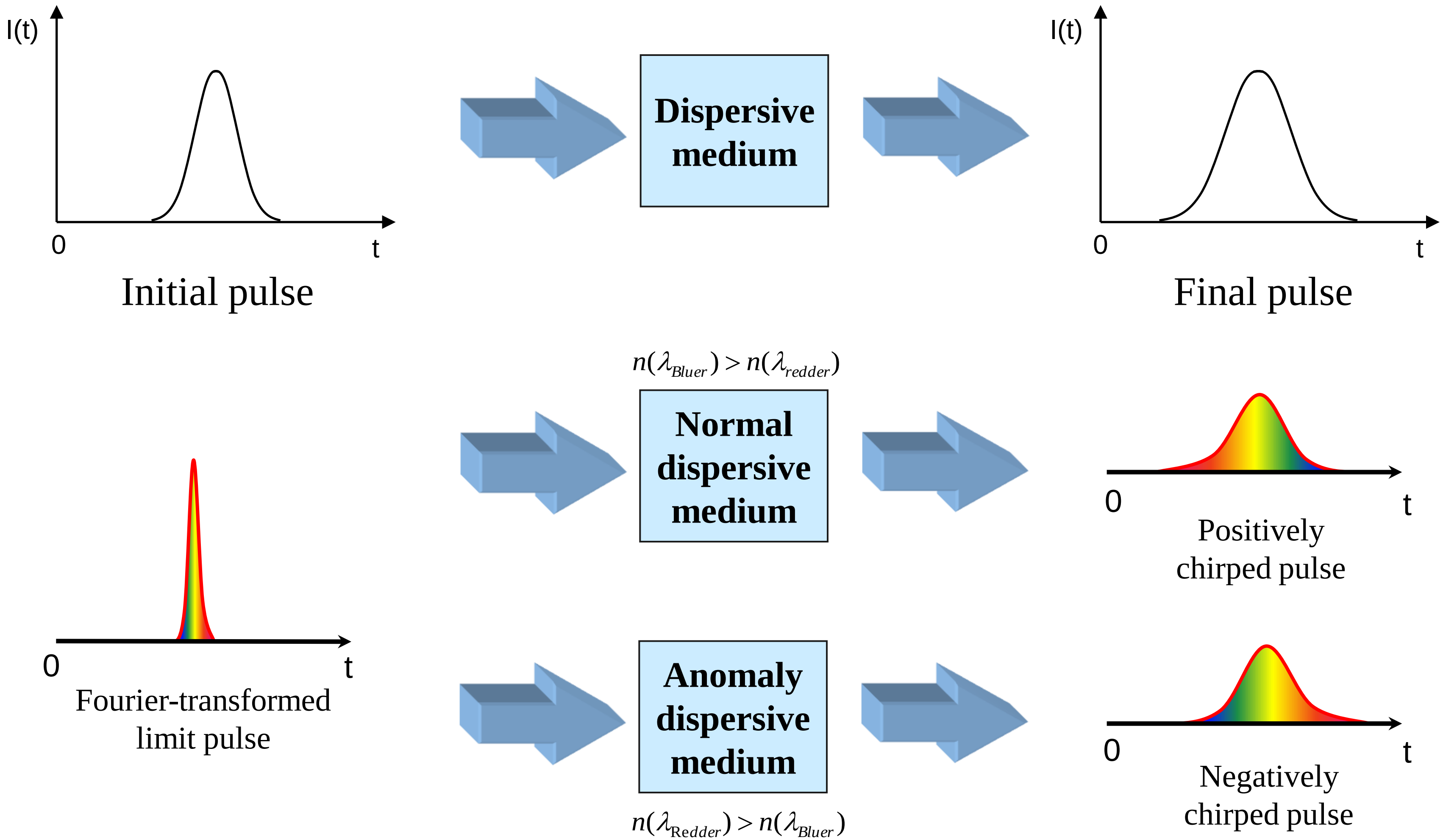
$$\overline{P}_{CW} \sim 250-300\,\mathrm{mW}$$

$$\overline{P}_{\mathrm{ML}} \sim 450\,\mathrm{mW}$$

$$\tau_{TL} = 0{,}441 \cdot \frac{\lambda_0^2}{c\Delta\lambda_p} \quad \Rightarrow \quad \tau_{TL} \sim 16{,}5\,\mathrm{fs}$$

Fourier Transform limited pulse

Dispersion of ultrashort pulse

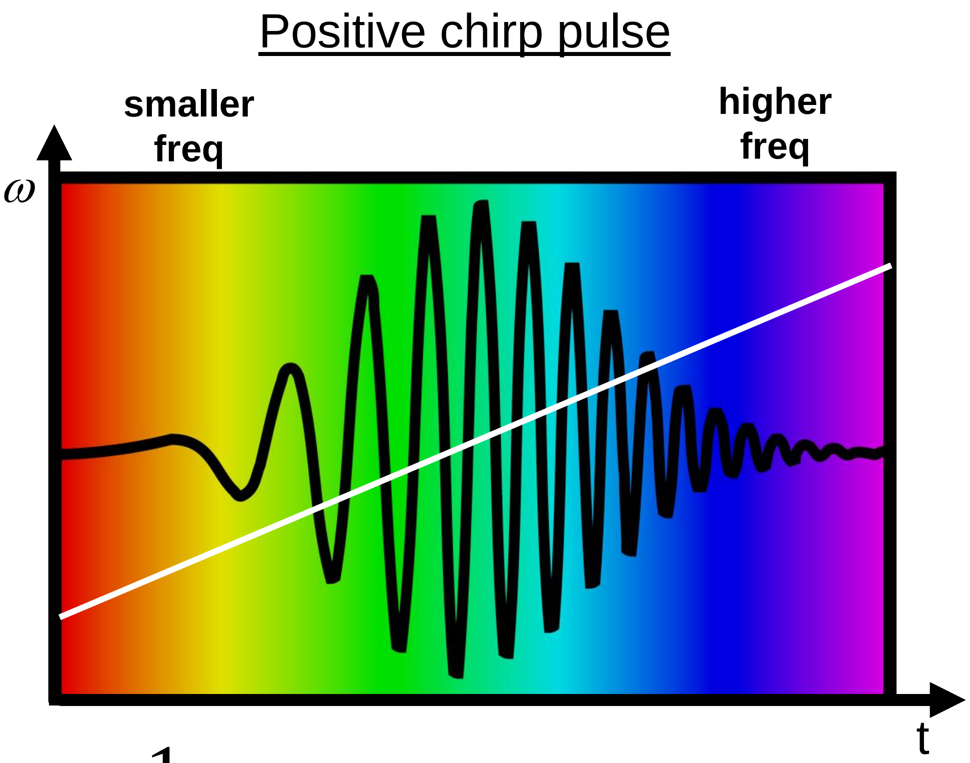


Dispersion of ultrashort pulse - Chirp

- *chirp* $E(t) = \mathrm{Re}\left\{\sqrt{I(t)}\,\exp\{i[\omega_0 t - \phi(t)]\}\right\}$

$$\omega_{inst}(t) \equiv \omega_0 - \frac{\mathrm{d}\phi}{\mathrm{d}t}$$

$$\phi(t) = \phi_0 + \phi_1 t + \frac{1}{2}\phi_2 t^2 + \ldots$$



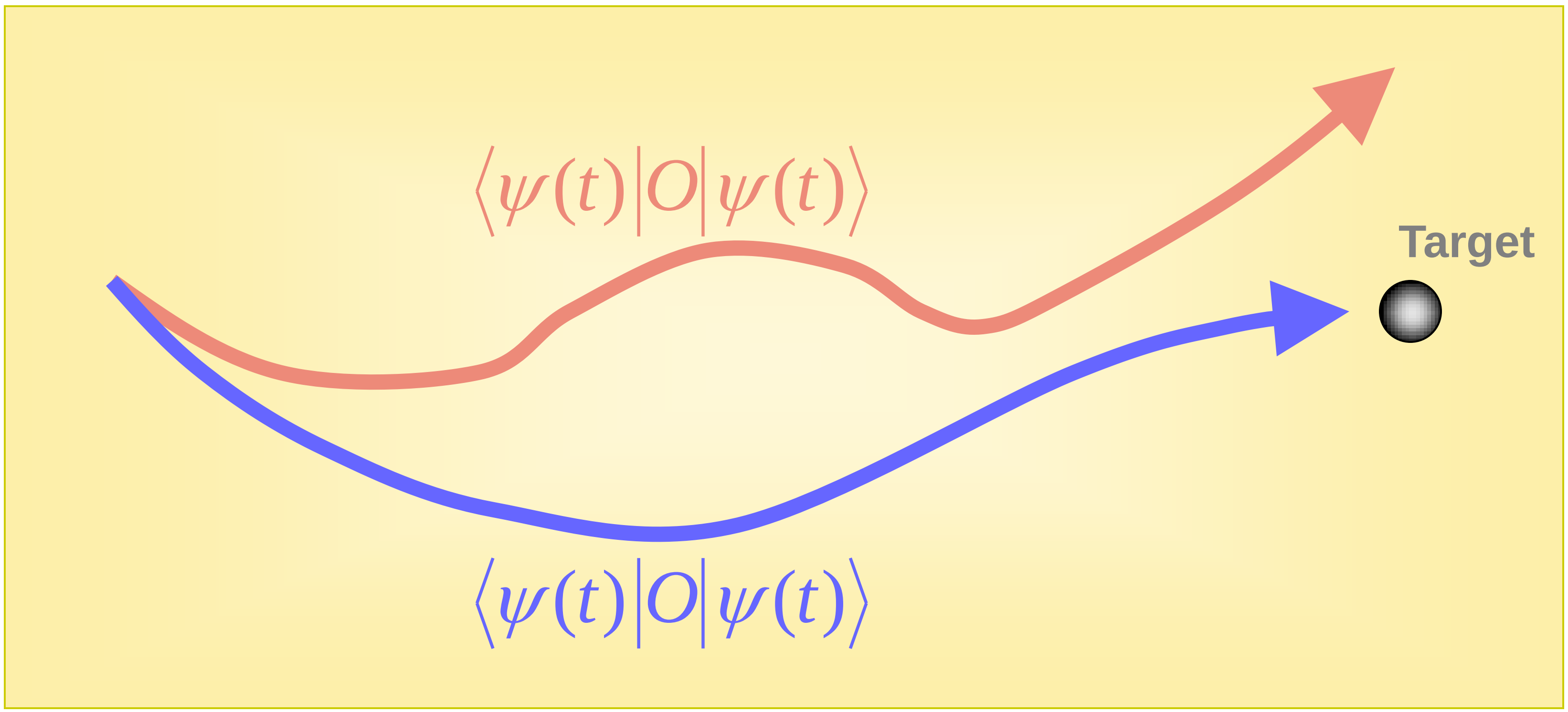
$$\varphi(\omega) = \varphi_0 + \varphi_1(\omega - \omega_0) + \frac{1}{2}\varphi_2(\omega - \omega_0)^2 + \ldots$$

ultrashort pulses

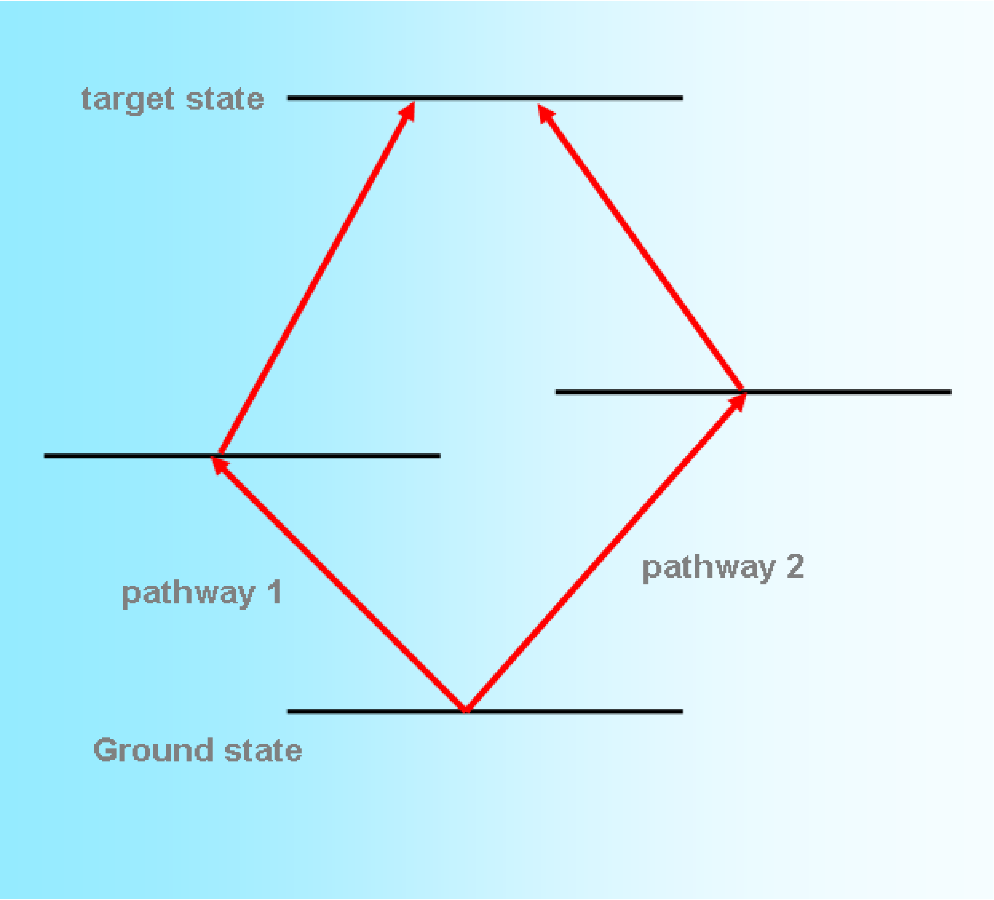
- Short temporal duration
  - High light intensity
  - Allows nonlinear effects
- wide spectral band
  - Control nonlinear process and photo-reactions

Coherent control

General idea: use the broad spectral band of ultrashort pulses to direct a given optical process



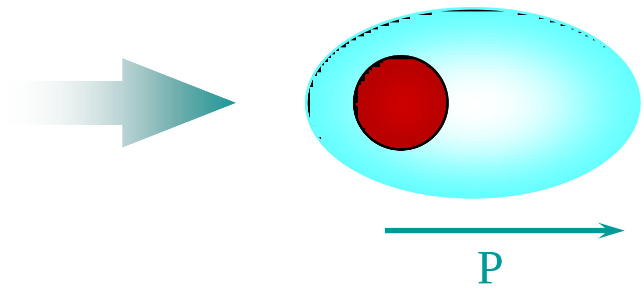
Coherent control



Distinct combinations of photons of the same pulse can lead the system to a given final state through different pathways

How does that actually work ?

Nonlinear optics



- *High intensities*

$E_{rad.} \sim E_{inter.}$

Charges:
Anharmonic Oscillator

$$\vec{P} = \chi^{(1)} . \vec{E} + \chi^{(2)} : \vec{E}\vec{E} + \chi^{(3)} \vdots \vec{E}\vec{E}\vec{E} + \ldots$$

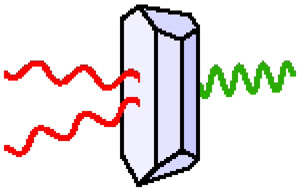
nonlinear effects are observaded with high intensities

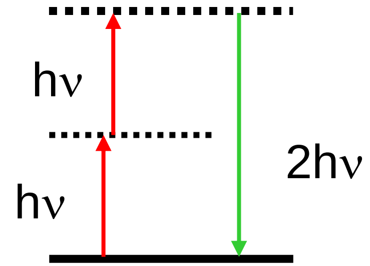
Nonlinear optics

1961:

- Second Harmonic Generation (SHG), Franken *et al.*

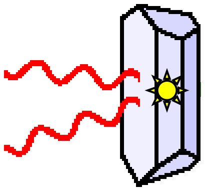
Ruby laser beam (694.2nm) → Quartz Crystal (SiO_2) → Laser light (347.1nm)

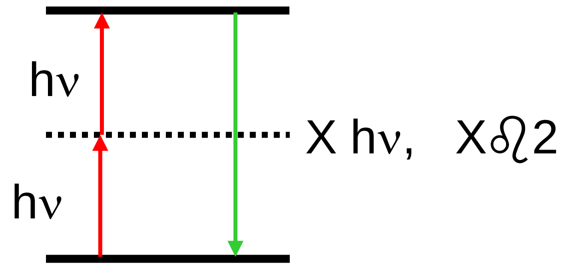




- Two-photon Absorption (2PA), Kaiser and Garret

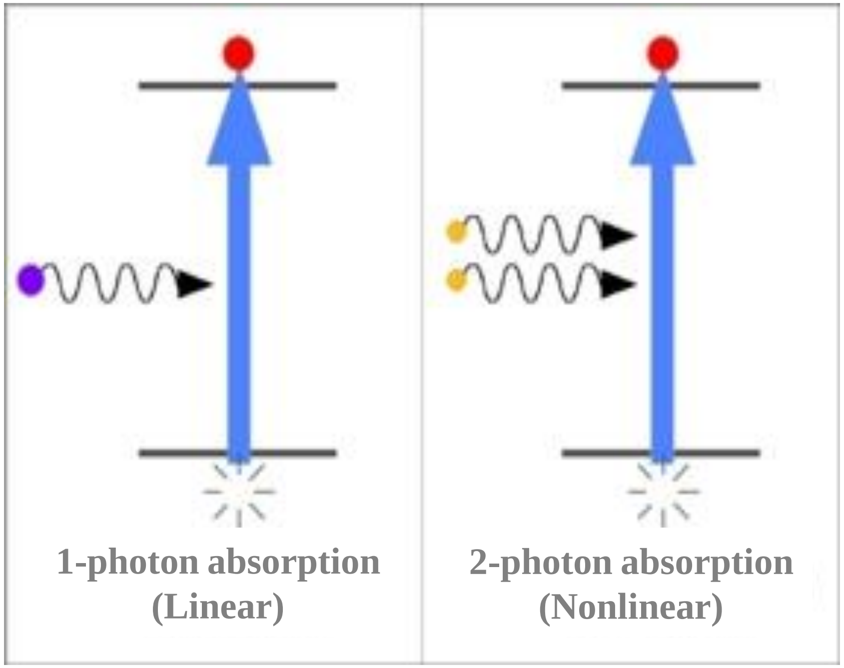
Ruby laser beam (694.2nm) → Eu_2:CaF_2+ Crystal → Fluorescent light (425 nm)





Two-photon absorption (2PA) process

Phenomenon does not described for the Classical Physics and does not observed until the development of the Laser.



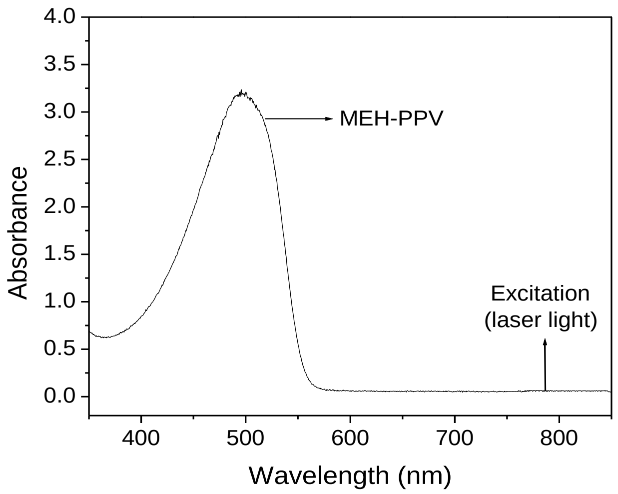
Maria Göppert-Mayer was born June 28th 1906 in **Kattowitz**. In 1963 she received the Nobel Prize in Physics.

Theoretical model: Maria Göppert-Mayer, 1931

Two photons from an intense laser light beam are simultaneously absorbed in the same "quantum act", leading the molecule to some excited state with energy equivalent to the absorbed two photons.

Two-photon absorption

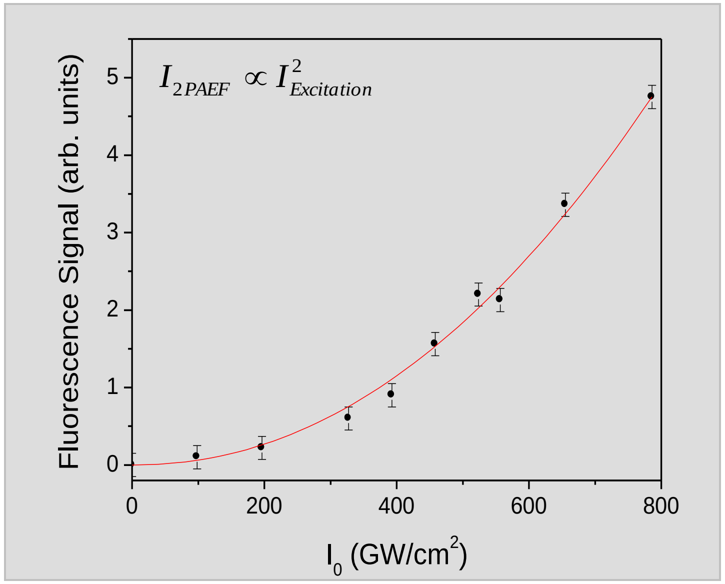
Materials are excited by non-ressonant light (outside of the absorption band)



Fluorescence (2PAEF)

$$\frac{dN_2}{dt} \approx N_1 \delta_{2PA} I^2$$

$$F \propto I^2$$

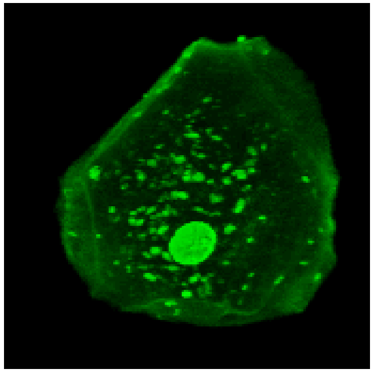


Spacial localization of the excitation

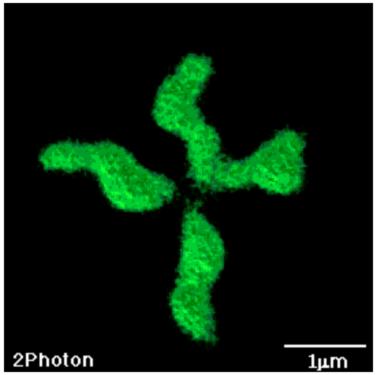
Amos, W.B. & White, J.G. (2003)

Applications of 2PA

- **Microscopy by 2PAEF**



Cell

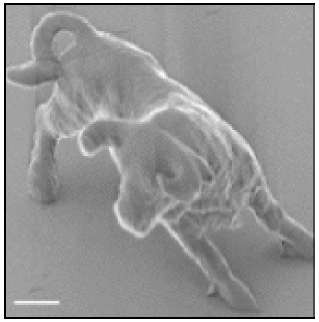


Human chromosome

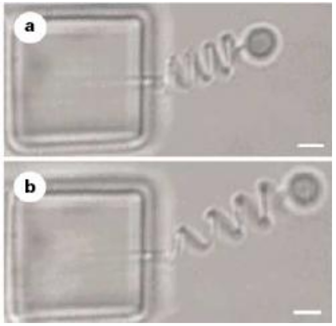
3D Imagems obtained via 2PAEF

- **3D Microfabrication by 2PA (Polimerization)**

Nature 412, 697-698 (2001)



Bull



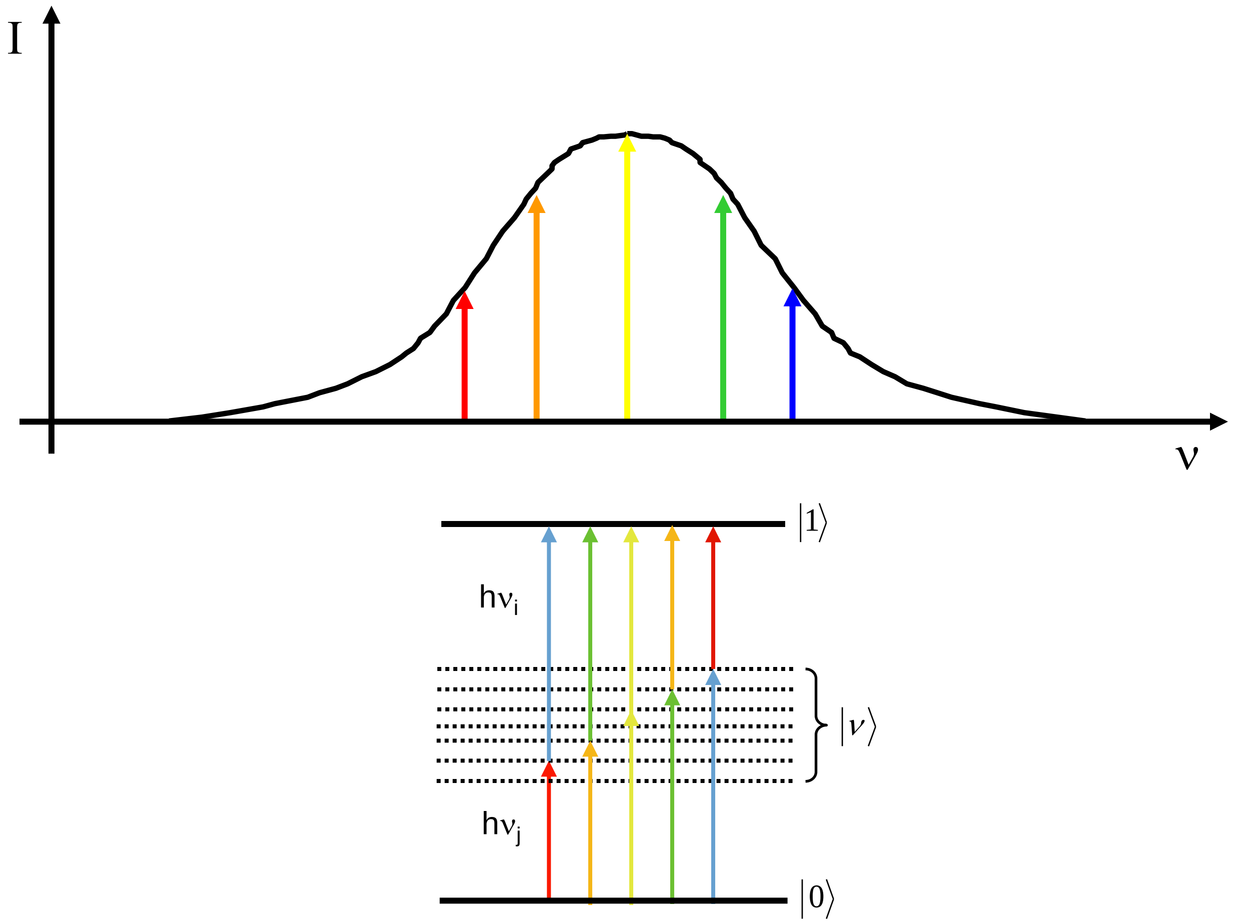
Mass-spring system

Applications of 2PA

Given all the applications of 2PA, it seems to be interesting to be able to directly control it

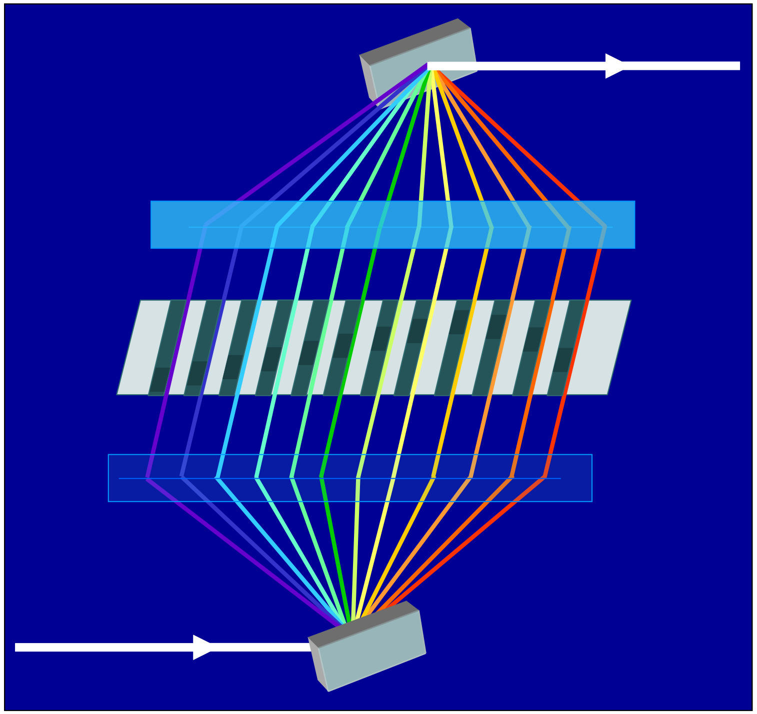
Just playing with the fs-pulse "shape"

Manipulating a two-photon process

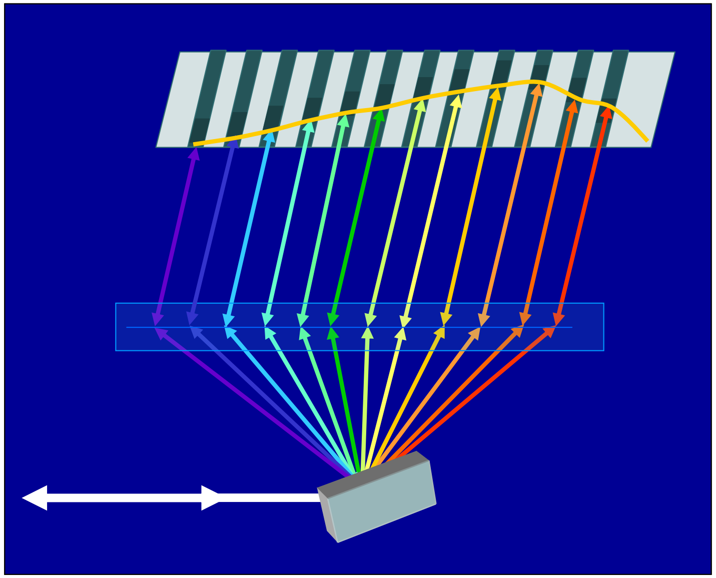


I´ll have to “shape” the phase profile of the pulse

Shaping the pulses



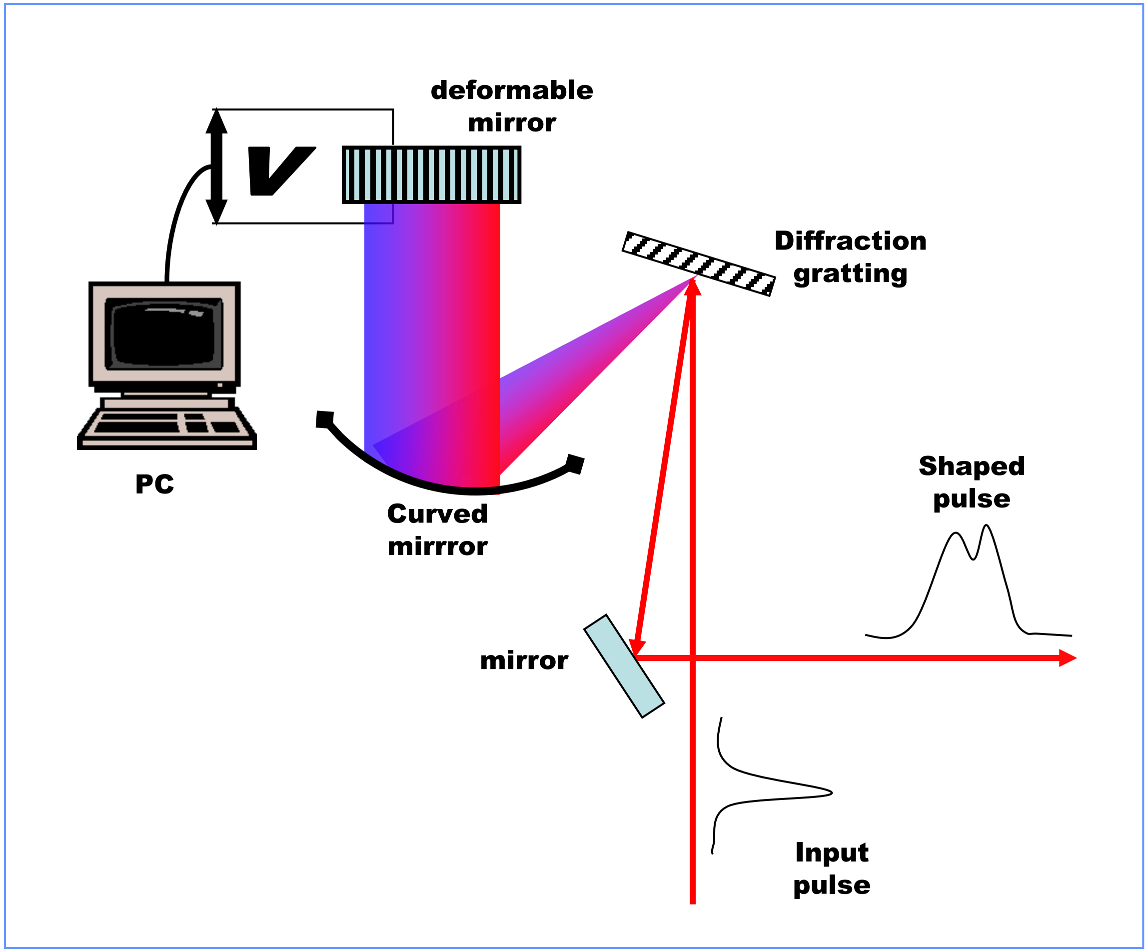
Transmission setup



Reflection setup

Shapping the pulses

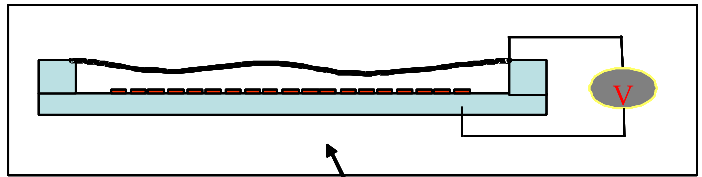
Deformable Mirror

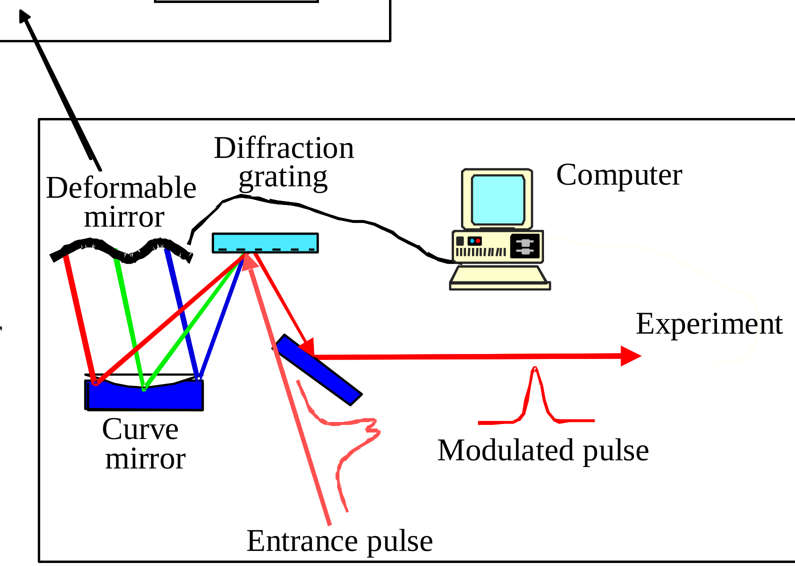


The deformable mirror acts in each spectral component of the pulse

Ultrashort pulse shaping technique

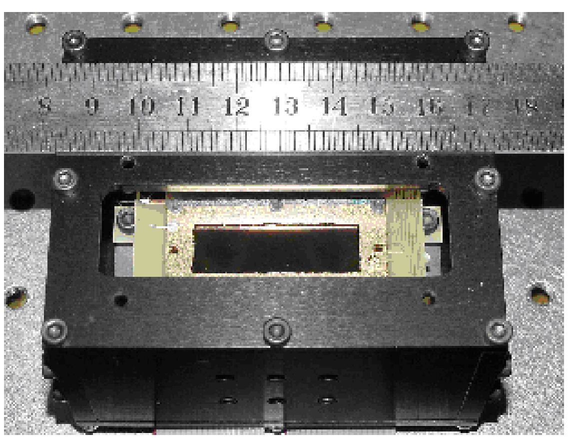
The MMDM is placed at the Fourier plane of a zero dispersion stretcher consisting of a 600 grove/mm ruled grating and a 25 cm focal-length mirror.





Experimental setup for ultrashort pulse shaping technique using deformable mirror

Micromachined deformable mirror (MMDM)



Shaping the ultrashort pulse in the phase domain.

MMDM is a 600 nm gold-coated silicon nitride membrane (8 mm x 30 mm) suspended over an array of 19 actuator electrodes on a printed circuit board. Potential applied to the actuator creates an electrostatic attraction between the membrane and the electrode, deforming the mirror surface.

Shapping the pulses

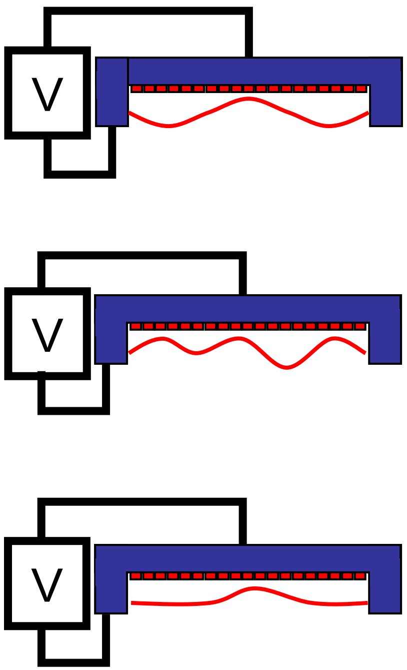
How to define which shape to use ?

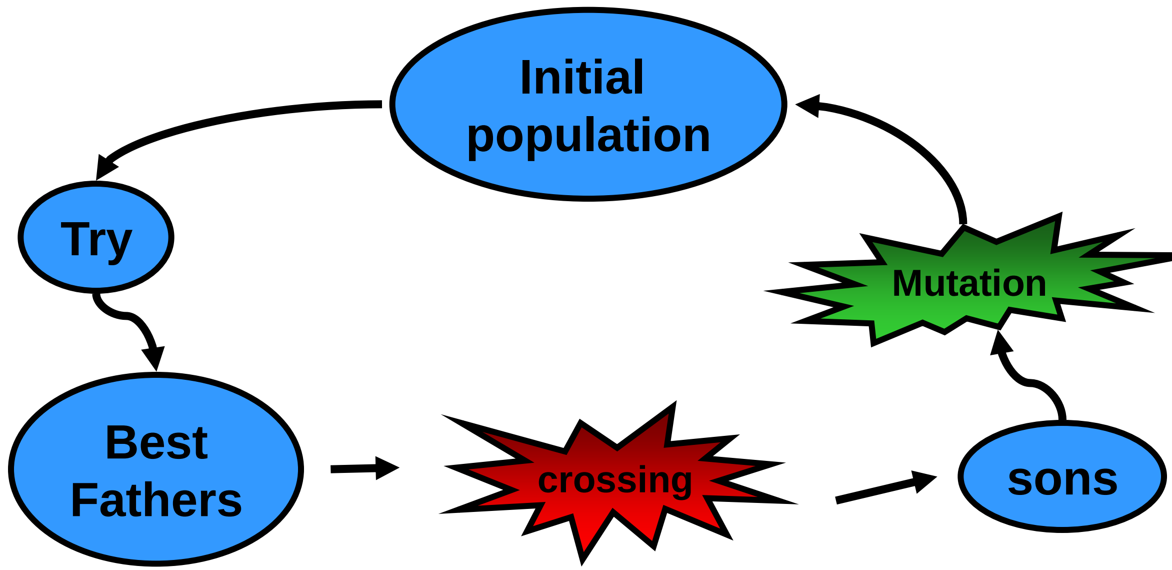
Genetic Algorithm

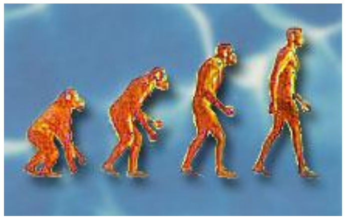
Phase Mask

Genetic Algorithm

Shaping the pulse + computer algorithm



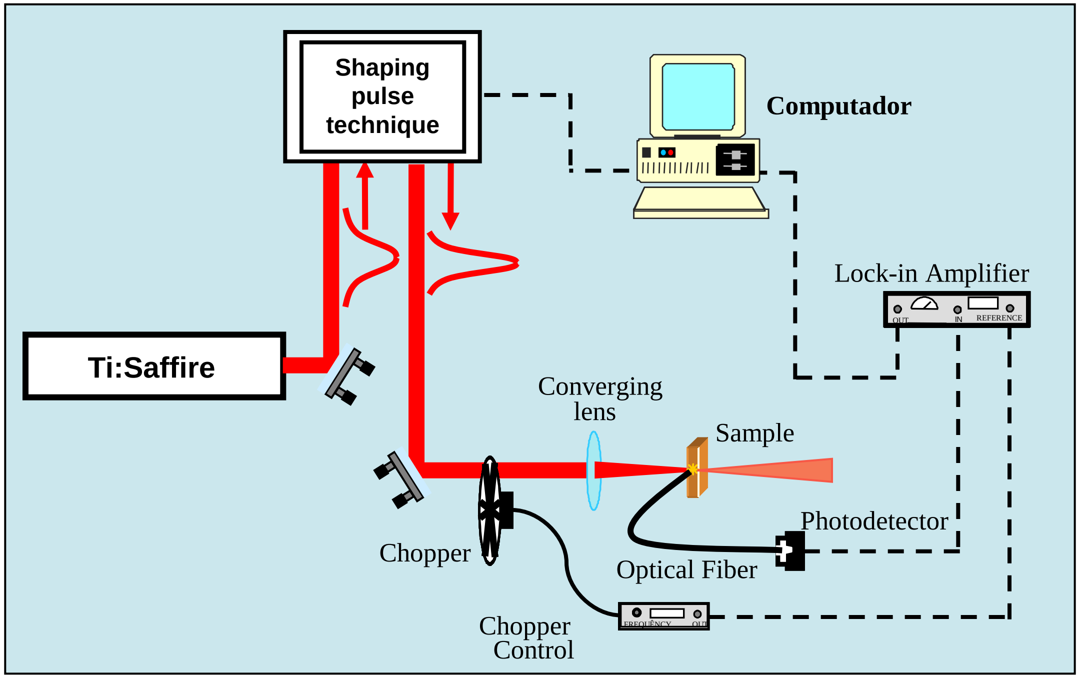


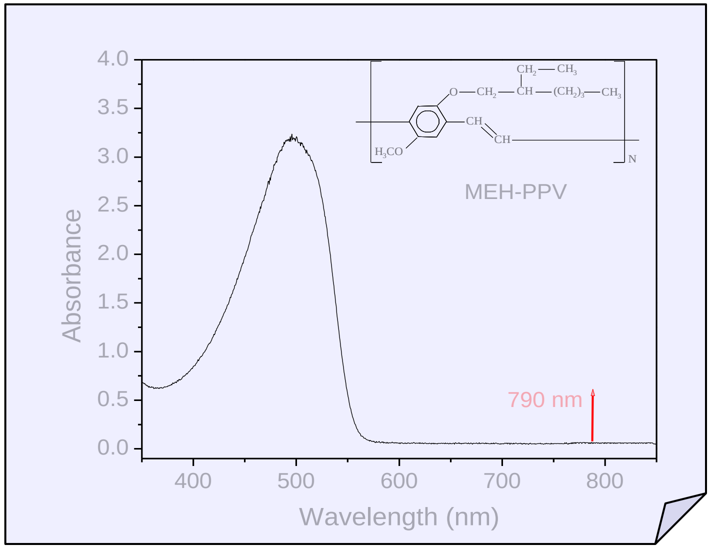


Evolution

In this case, the process is optimized but the mechanism is not well understood

Optimization of two-photon induced fluorescence

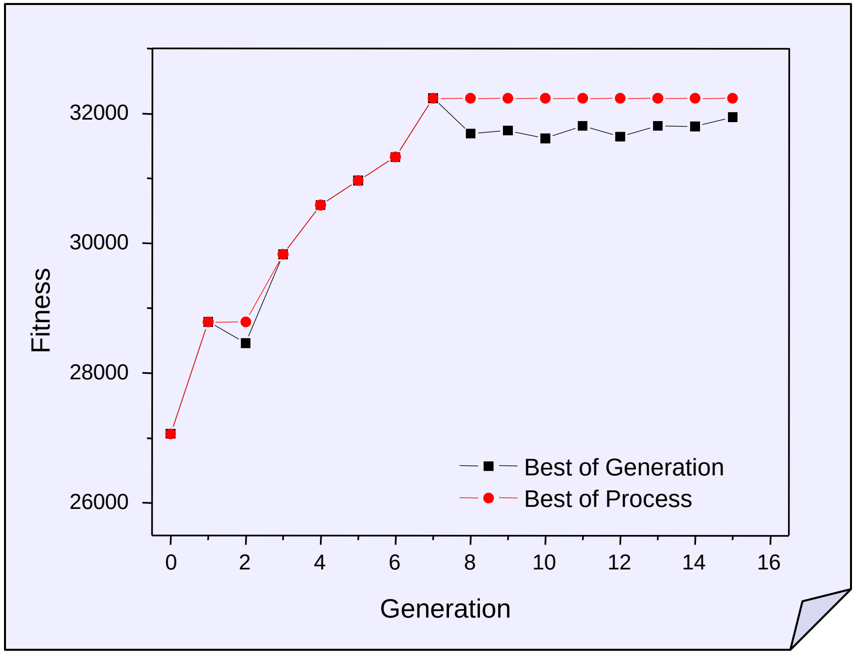


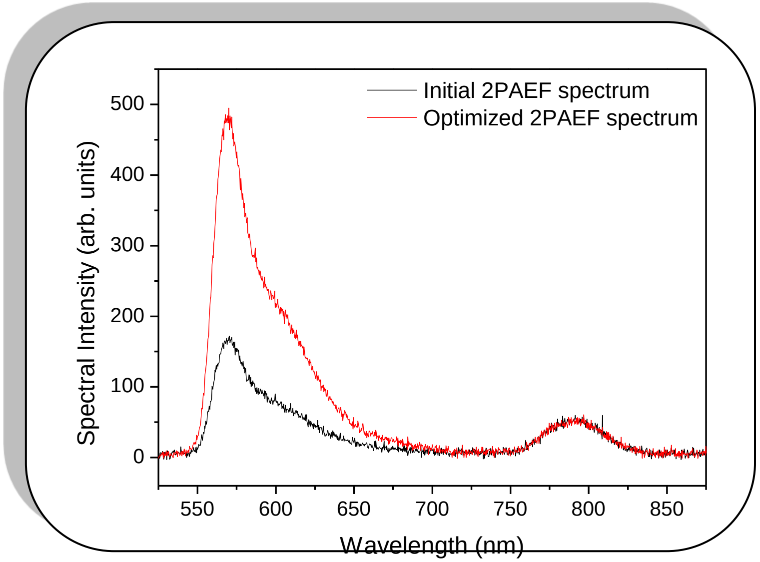


UV-Vis spectra of MEH-PPV in chloroform

Optimization of two-photon induced fluorescence

Optimization process of the 2PAEF spectrum

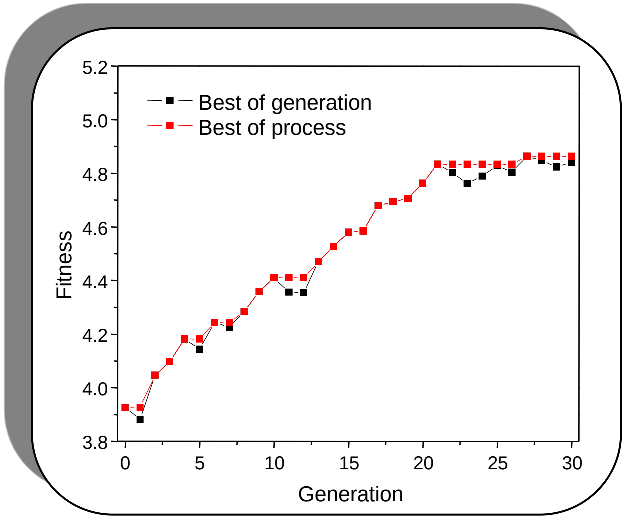




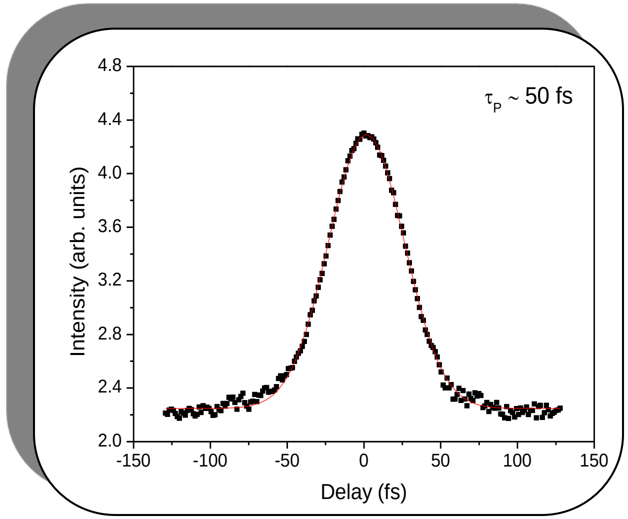
Optimization of two-photon induced fluorescence

What is happening with the pulse during its evolution

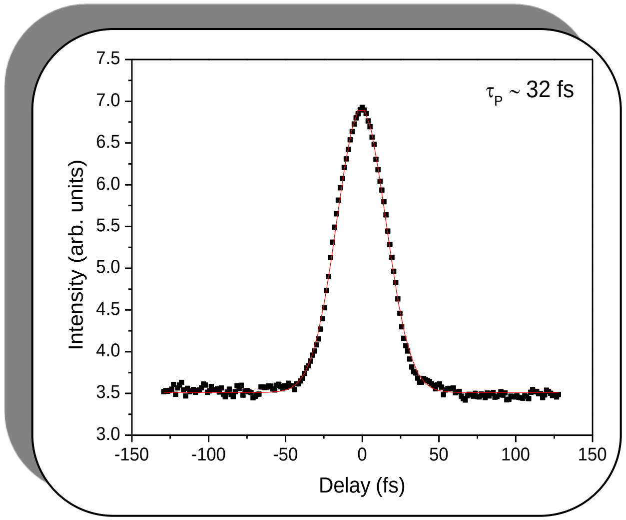
Optimization of the two-photon induced fluorescence



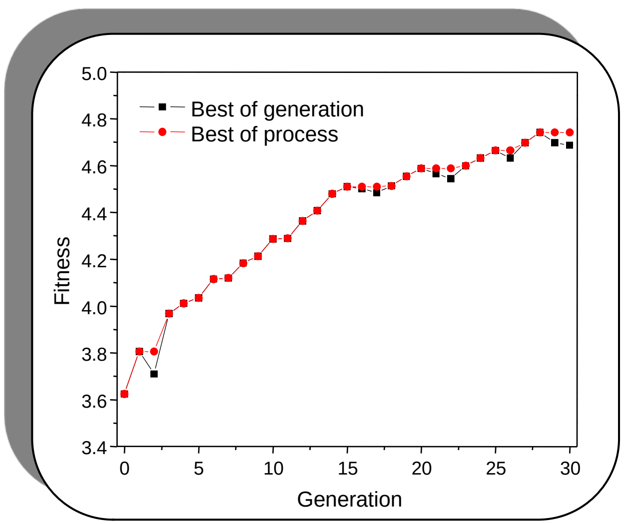
Evolution of the optimization process of the ultrashort pulse



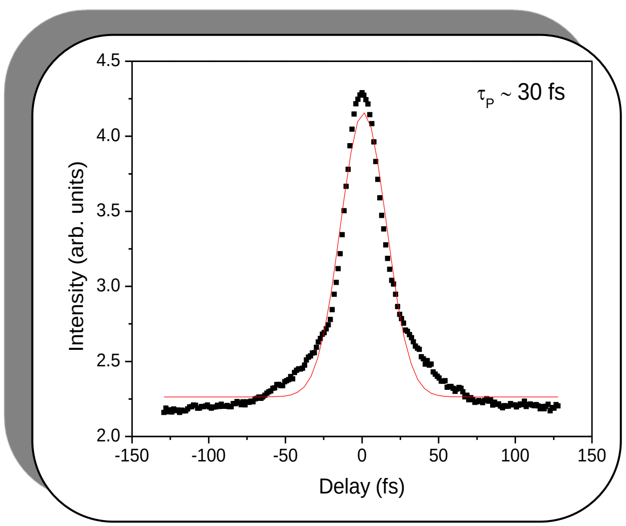
Autocorrelation obtained for non optimized pulse



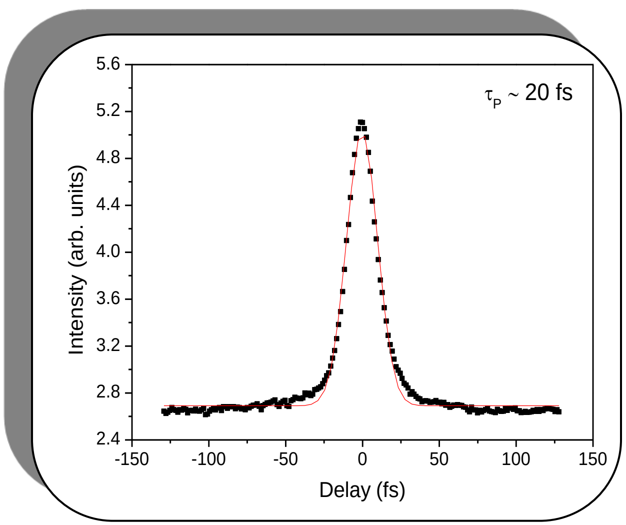
Autocorrelation obtained for optimized pulse



Evolution of the optimization process of the ultrashort pulse

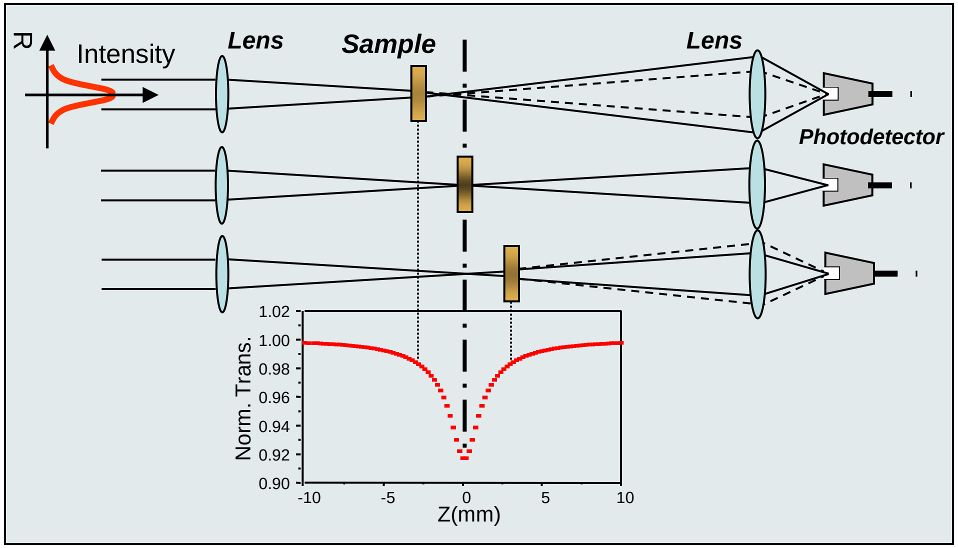


Autocorrelation obtained for non optimized pulse



Autocorrelation obtained for optimized pulse

Optimization of two-photon induced thermal lens

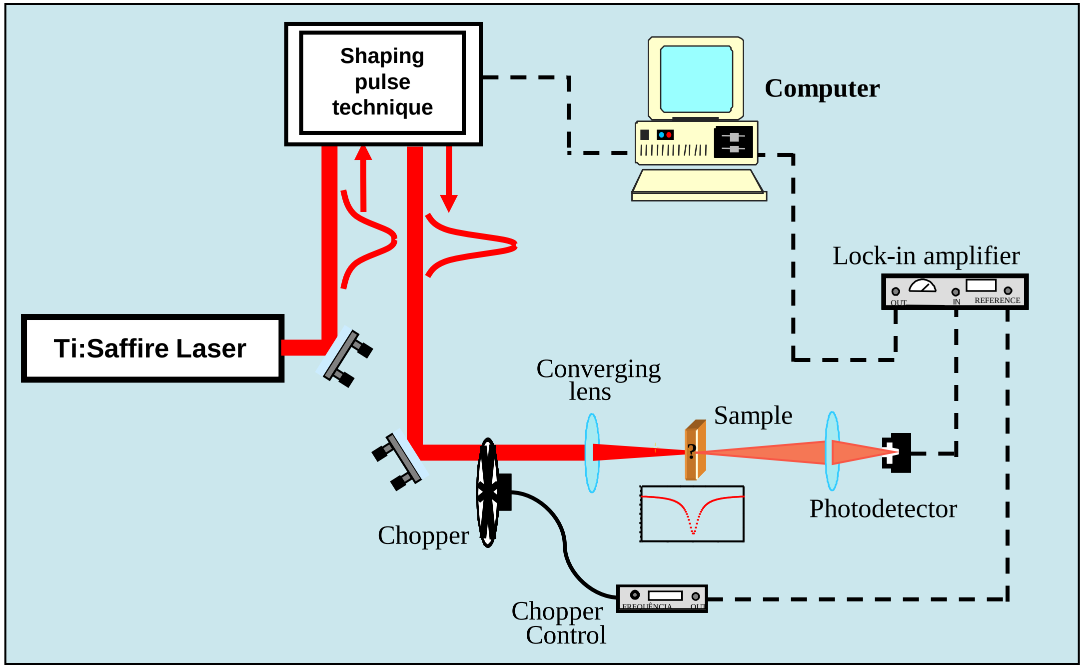


Z-Scan technique

(Nonlinear absorption)

Feedback Signal

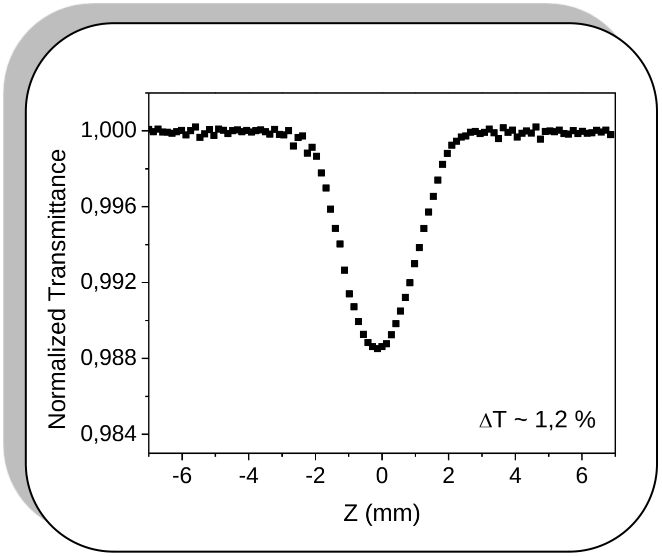
Nonlinear transmittance



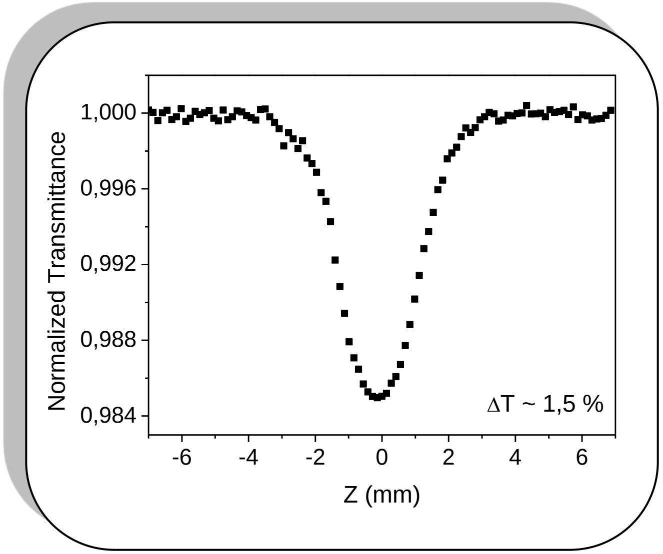
Samples

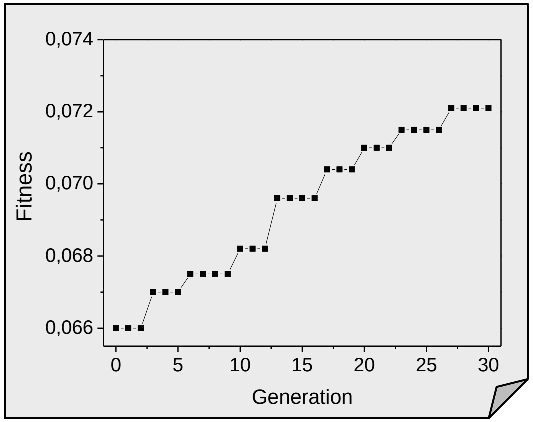
Fluorescents and non-fluorescents

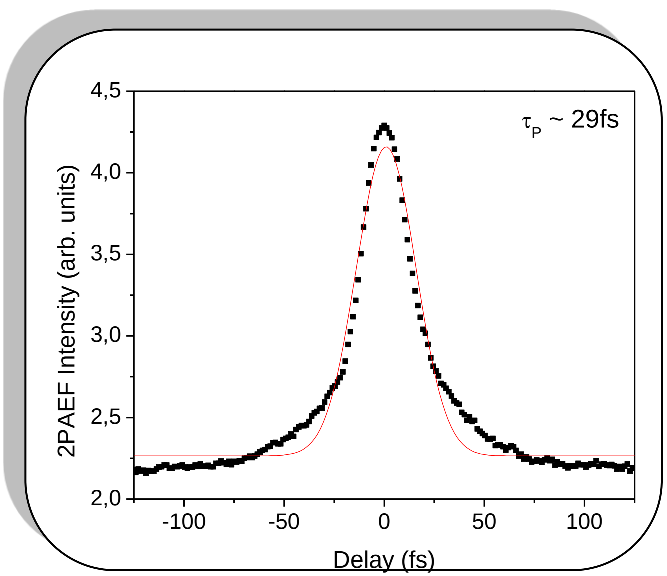
Optimization of two-photon induced thermal lens

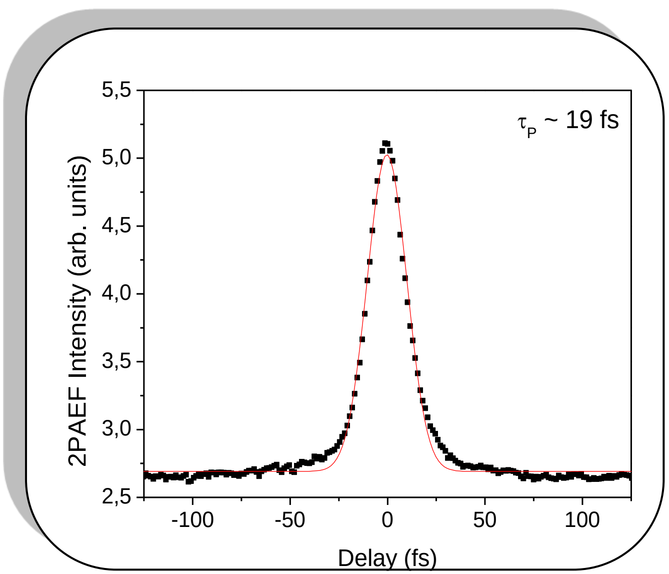


Sample: MEH-PPV

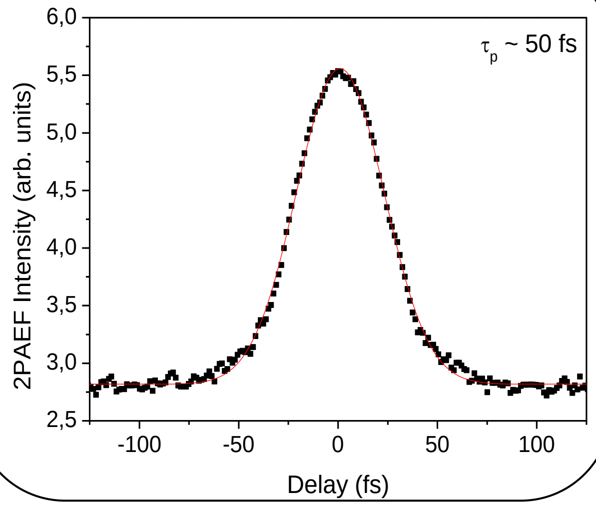




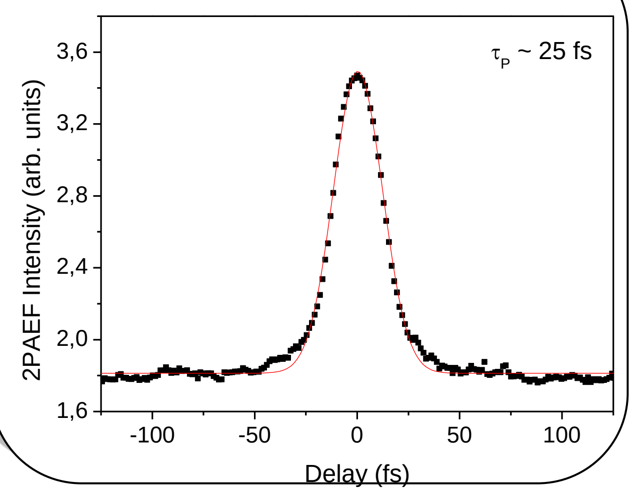


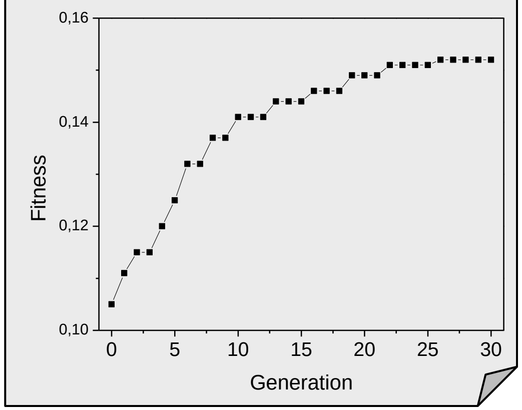


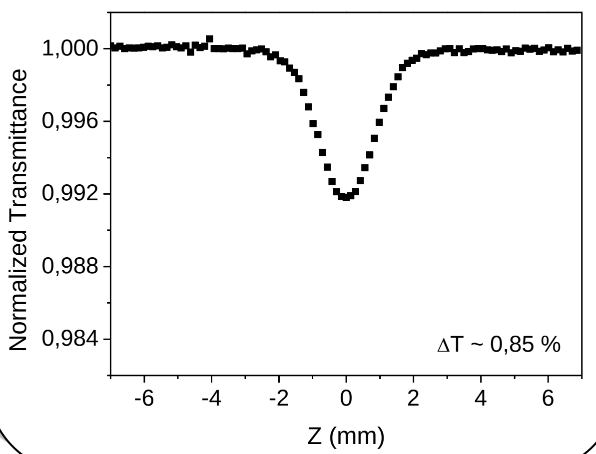
Optimization of two-photon induced thermal lens

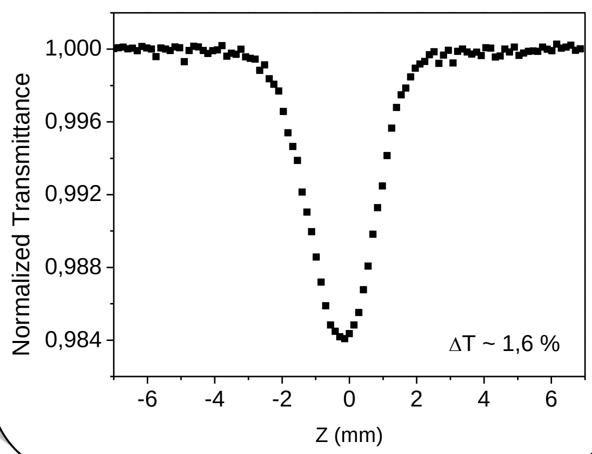


Sample: MEH-PPV





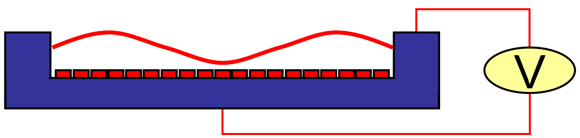


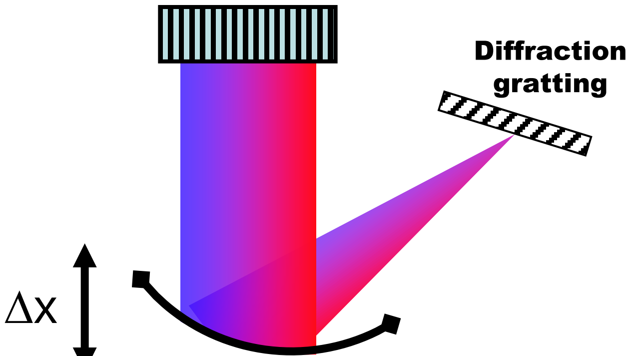


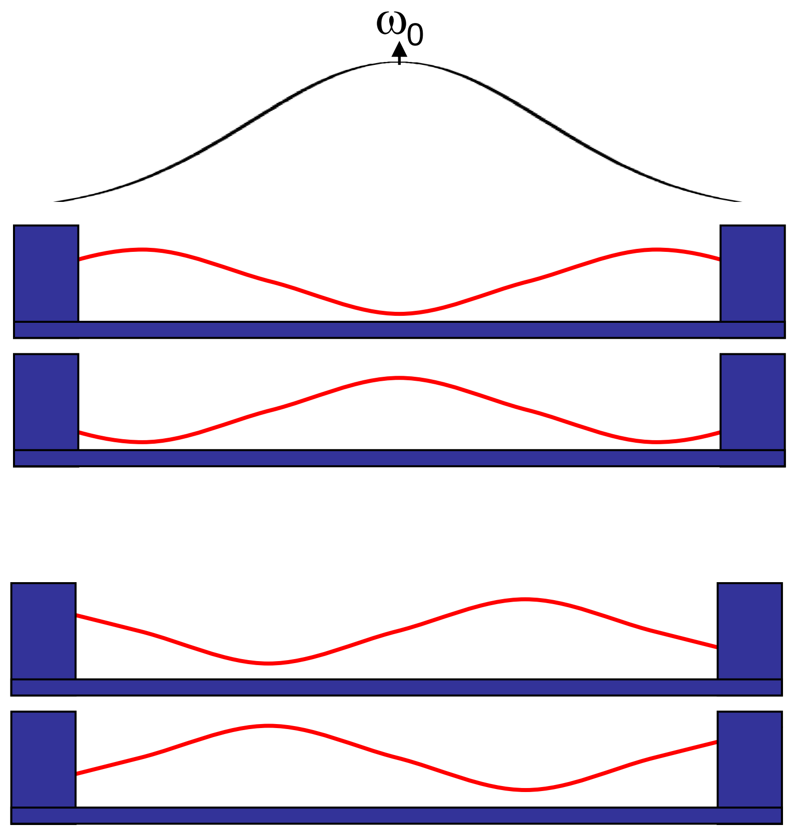
Phase Mask

In this case, we impose a known phase mask function to the ultra-short pulse

$$\Theta(\Omega) = \alpha \cos(\gamma\Omega + \delta)$$







Phase Mask

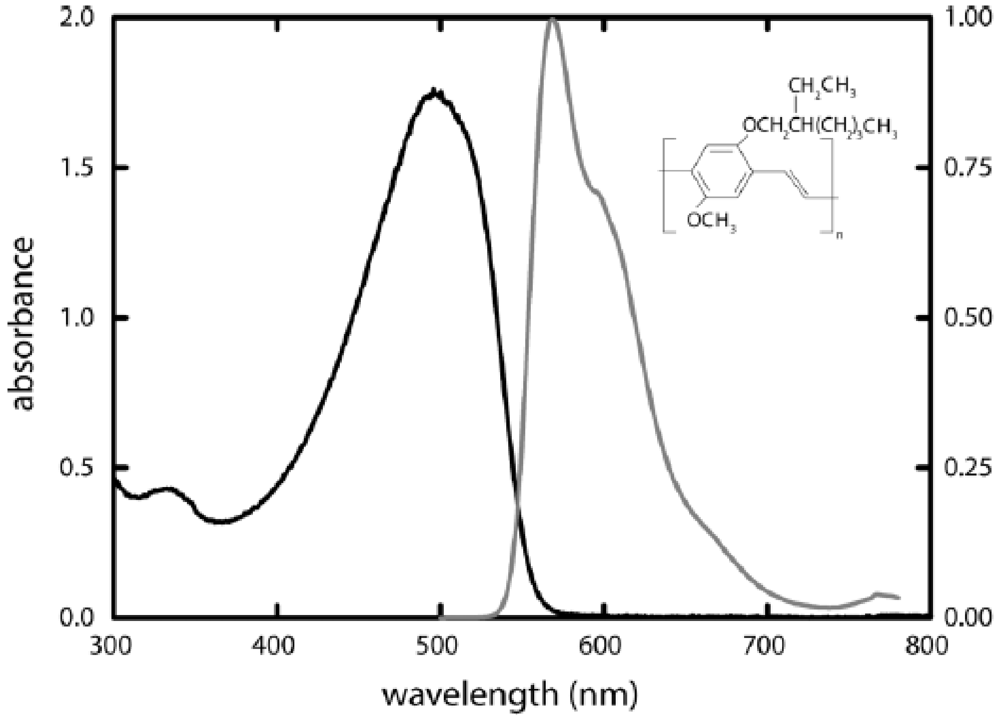
- Two-photon absorption transition is given by

$$S_2(\omega_0) \propto \left| \int_{-\infty}^{+\infty} A(\omega_0/2+\Omega)A(\omega_0/2-\Omega) \cdot \exp\left\{i[\varphi(\omega_0/2+\Omega)+\varphi(\omega_0/2-\Omega)]\right\} d\Omega \right|^2$$

Anit-symmetric phase mask: S_2 is independent of the phase

Symmetric phase mask: S_2 is minimized

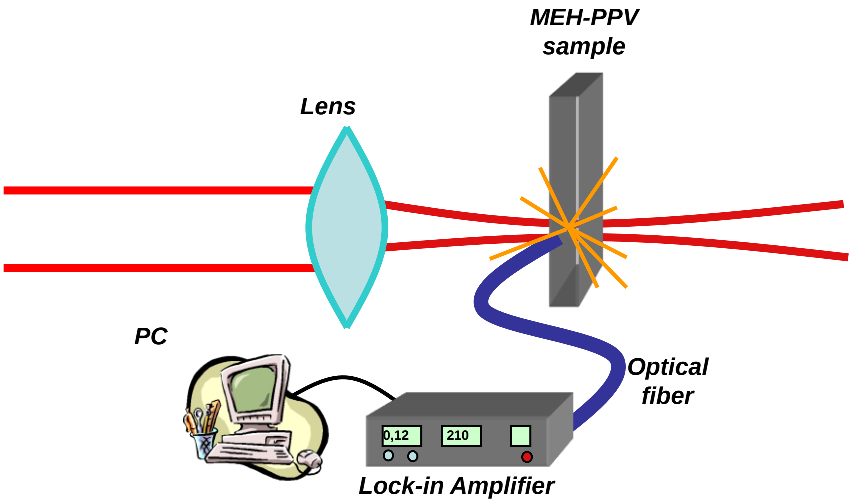
Controlling the MEH-PPV photo-degradation



MEH-PPV: conductive and luminescent polymer with interesting properties for applications

However, MEH-PPV photo-bleaches due to a photoxidation reaction, causing a decrease in its emission

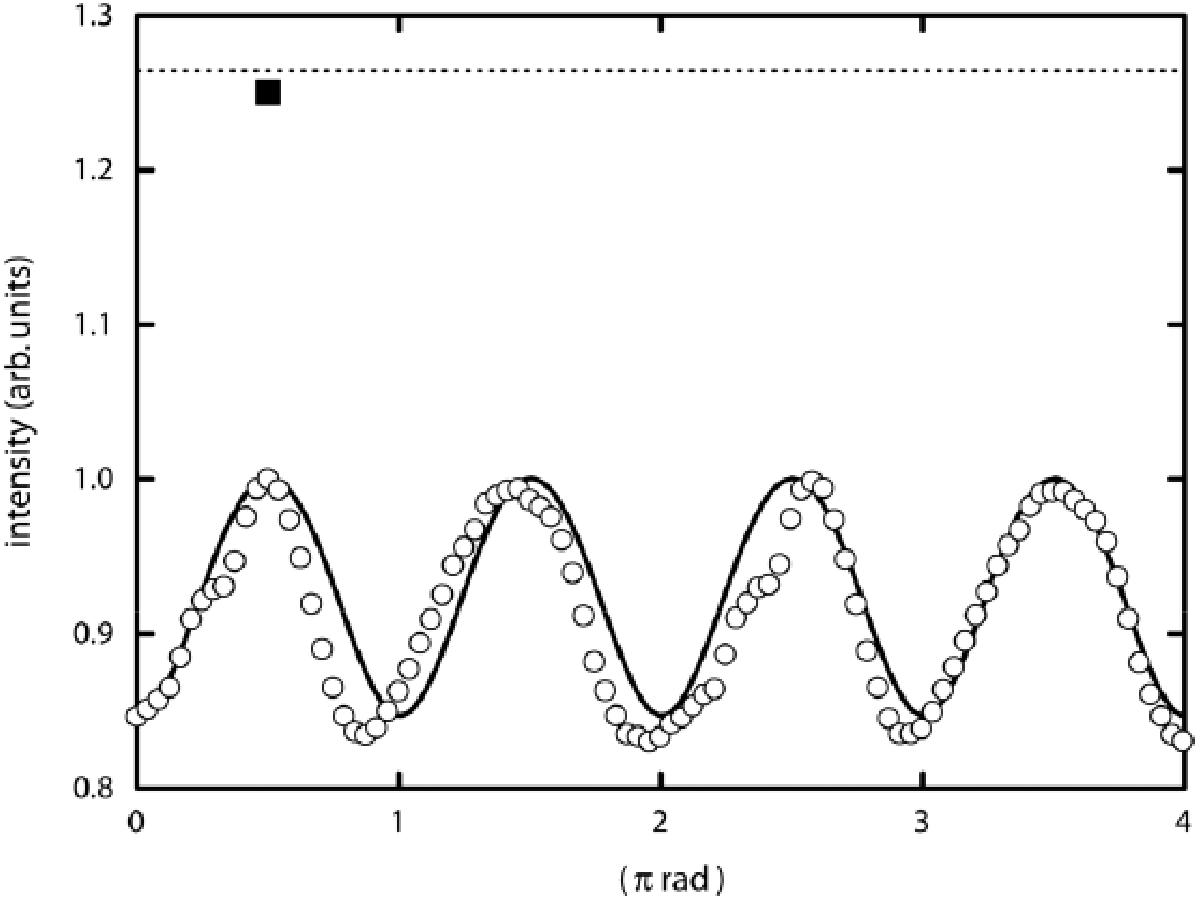
Controlling the MEH-PPV photo-degradation



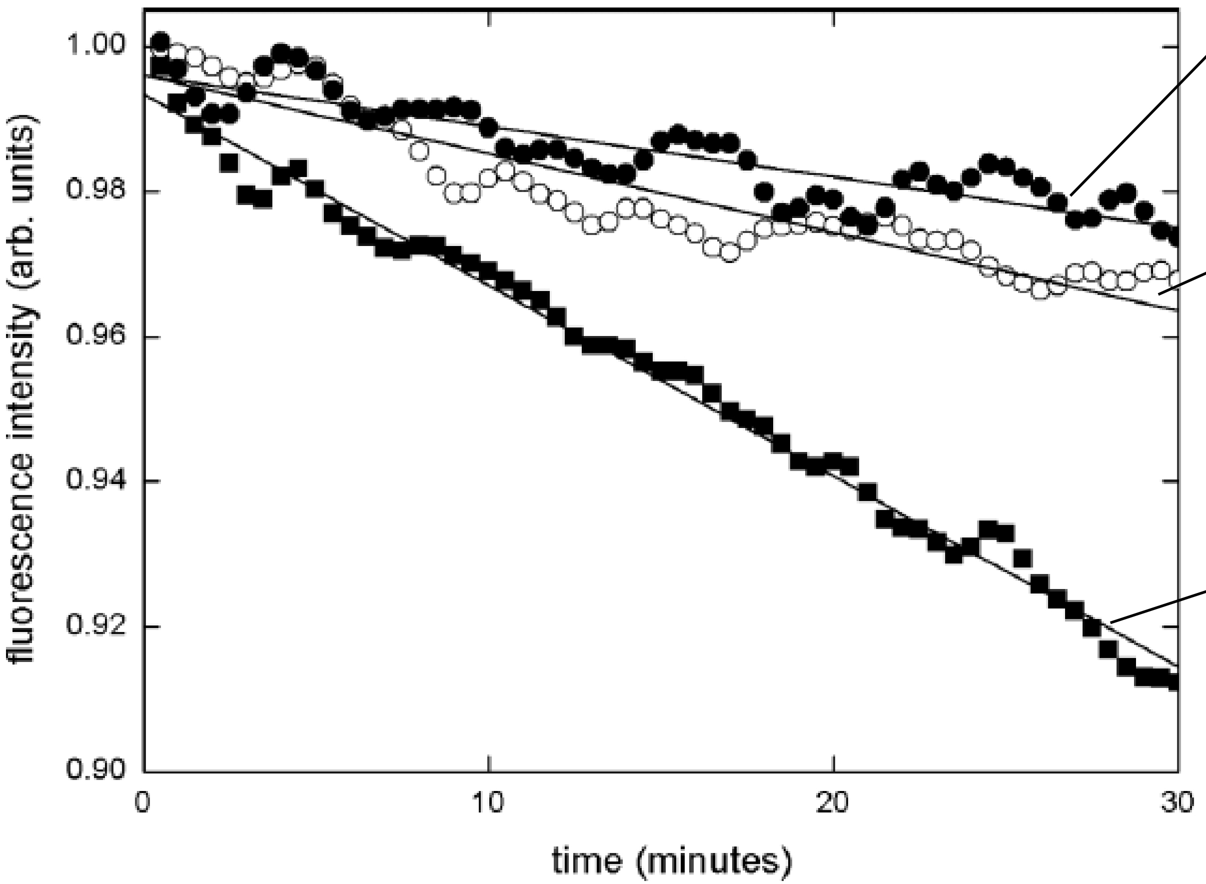
Observe the two-photon excited emission as a function of the phase-mask

Observe the photodegradation for distinct phase masks

Controlling the MEH-PPV photo-degradation



Controlling the MEH-PPV photo-degradation



$\delta = 1\ \pi$

0.07 %/min

$\delta = 0.5\ \pi$

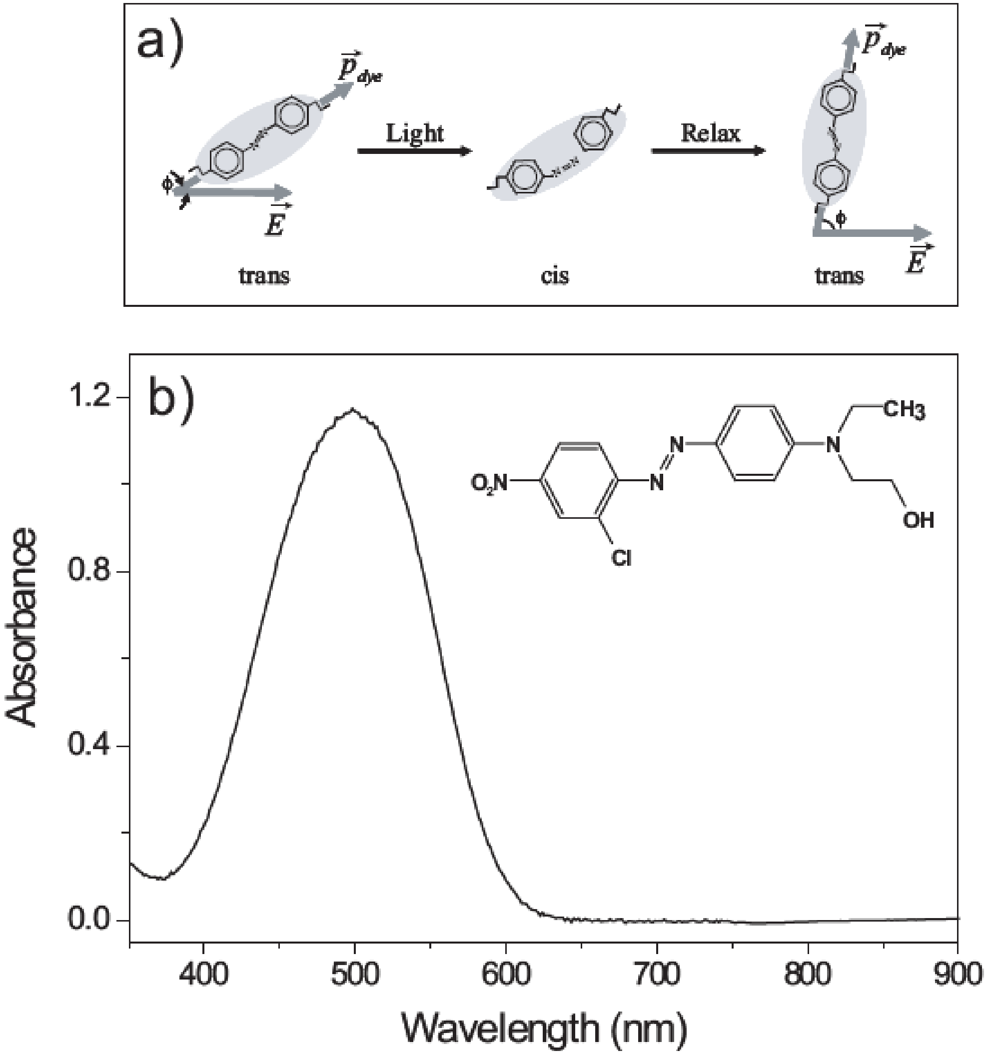
0.1 %/min

FTL

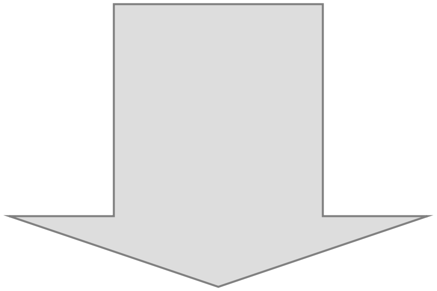
0.3 %/min

Photobleaching rate is smaller for the phase-masked pulses

Coherent control of molecular orientation

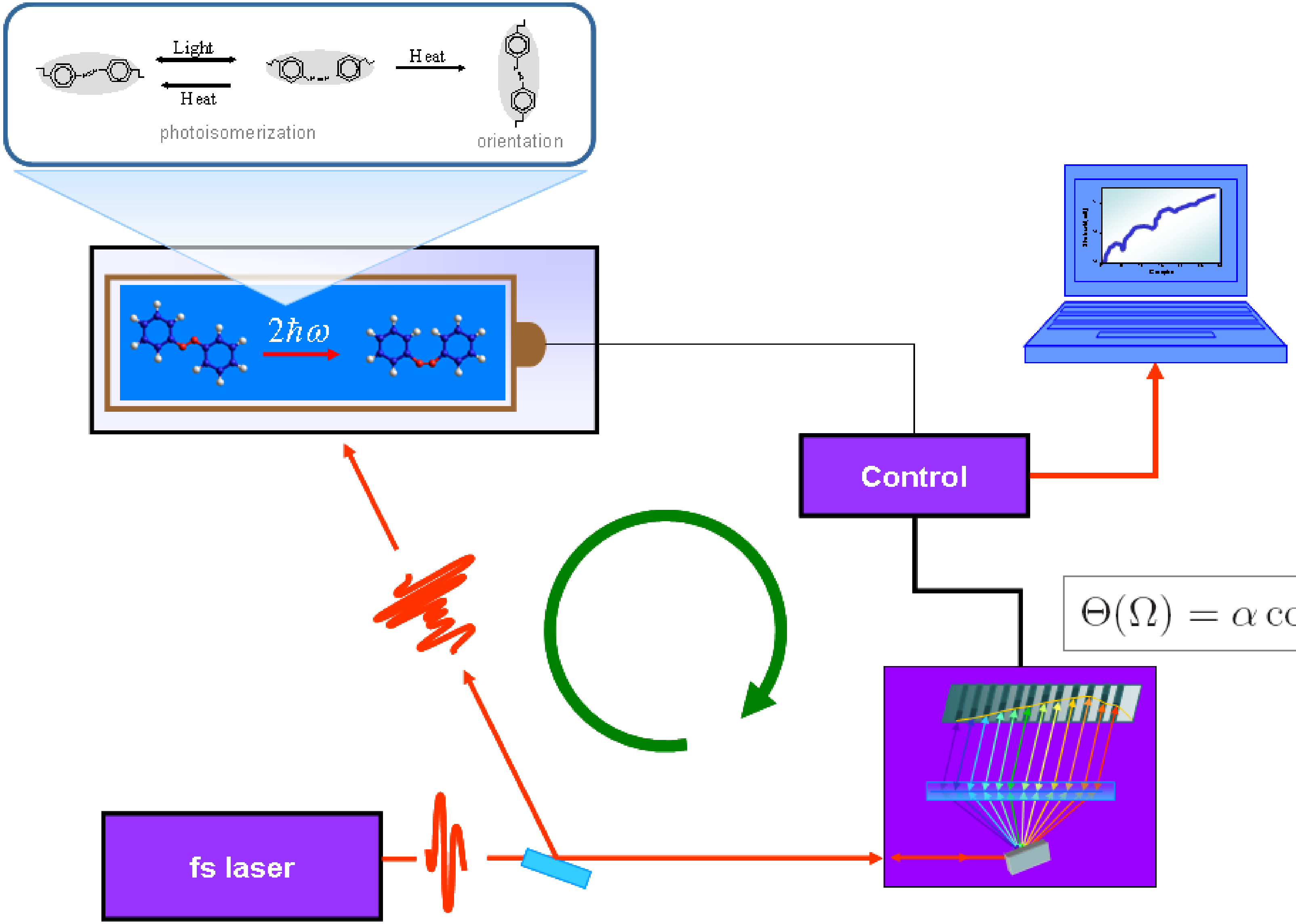


Use fs-laser (broad band) to induce two-photon absorption and, consequently, molecular orientation (optical storage)



Coherently control the molecular orientation

Coherent control of molecular orientation



$$\Theta(\Omega) = \alpha \cos(\gamma\Omega + \delta)$$

Coherent control of molecular orientation

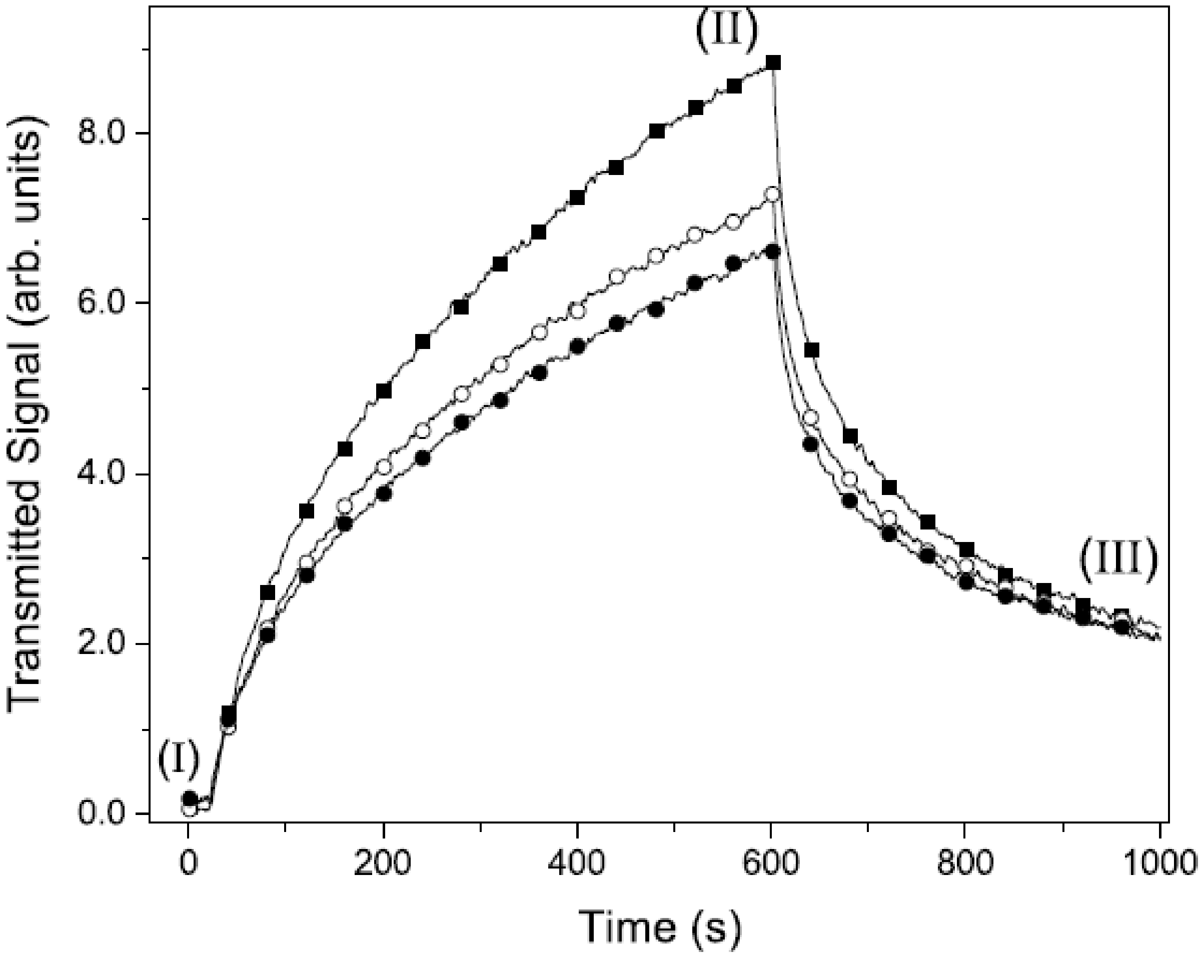
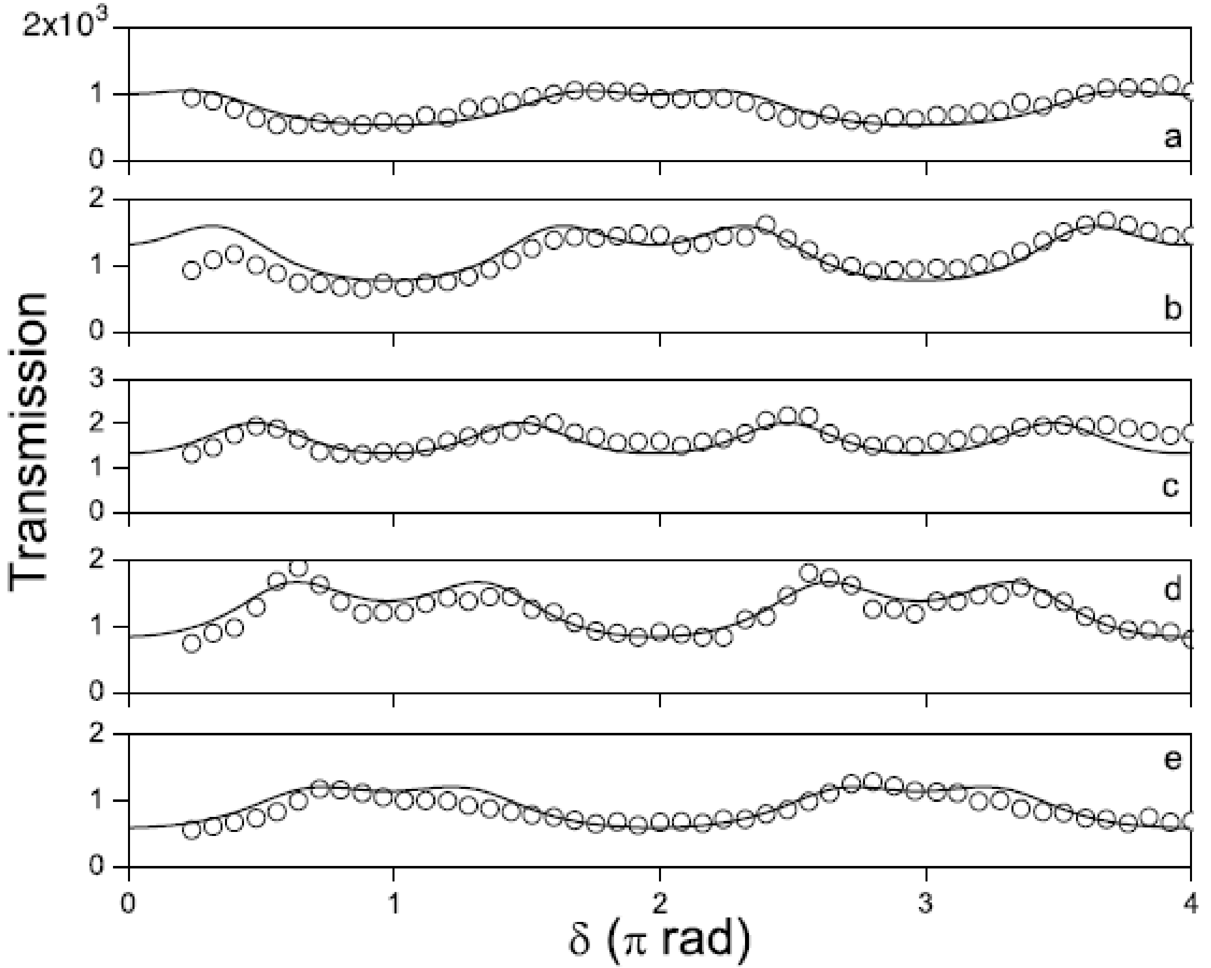


FIG. 2. Optical storage curves for the PMMA/DR13 film for three distinct δ values: $5\pi/10$(■), $7\pi/10$(○), and π(●).

Coherent control of molecular orientation

$$\phi(\Omega) = \alpha \cos(\gamma\Omega + \delta) + 1/2\beta\Omega^2$$



Conclusions

- Pulse shaping methods + coherent control of the nonlinear interaction seems to be an interesting method to further control nonlinear optical processes.

Thanks for your attention!!!