Femtosecond pulses in nonlinear optics: microfabrication and pulse shaping

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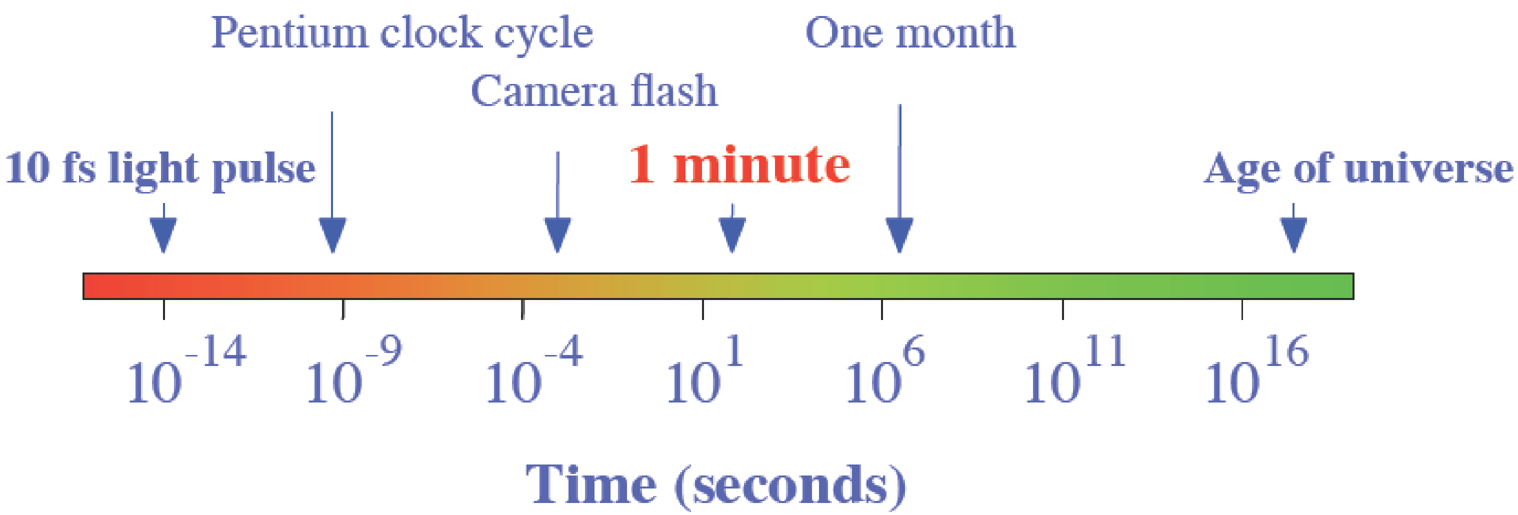
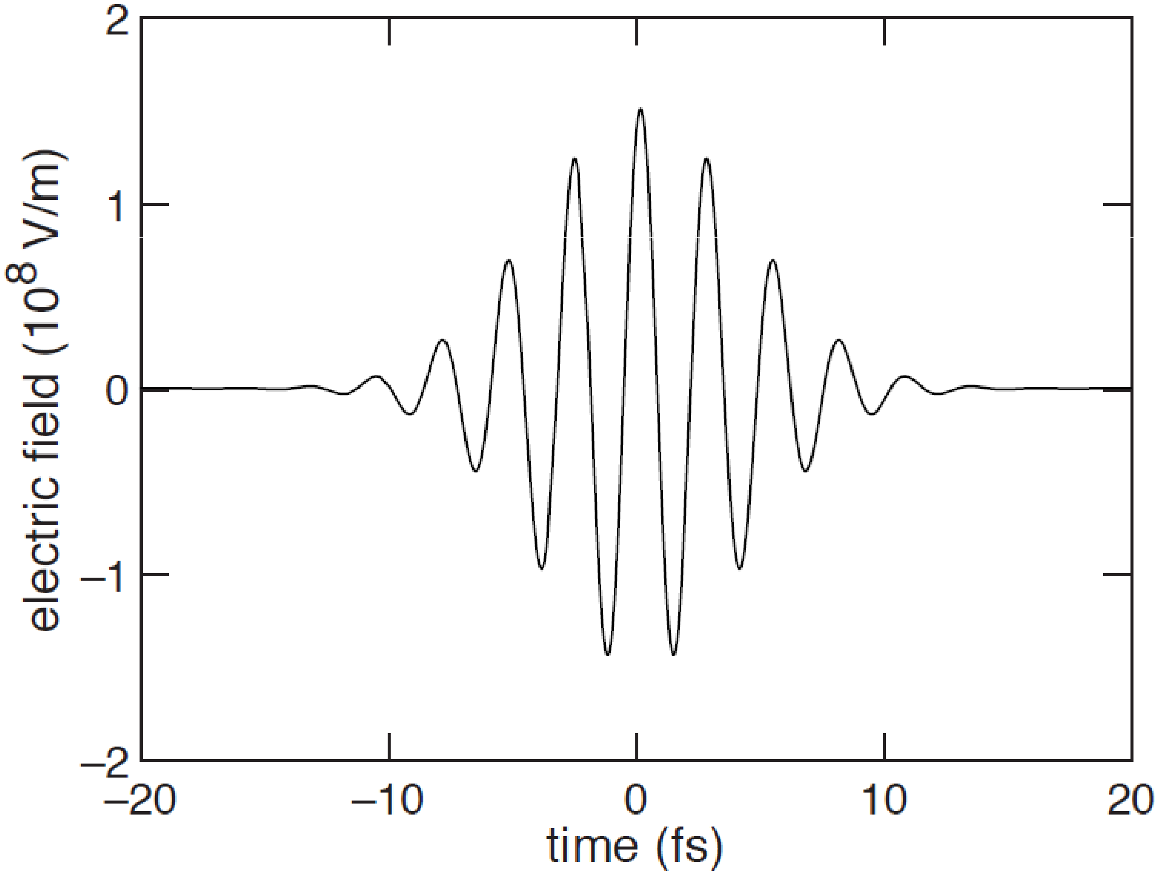
introduction

how short is a femtosecond pulse ?

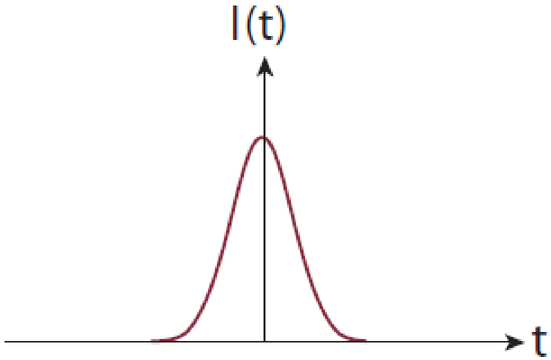
1 fs = 10^{-15} s

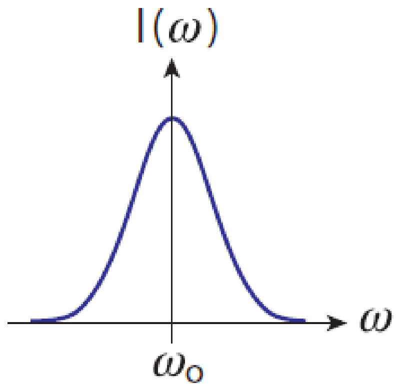


how short is a femtosecond pulse ?



introduction



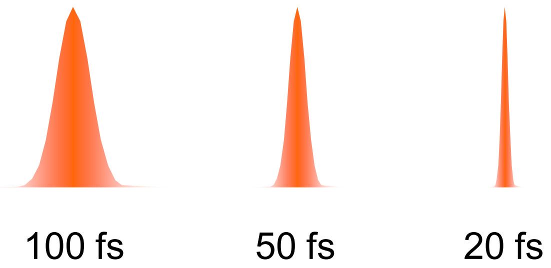


short duration → high intensities

broad spectral band → pulse shaping

fs-laser micromachining

Ti:Sapphire lasers



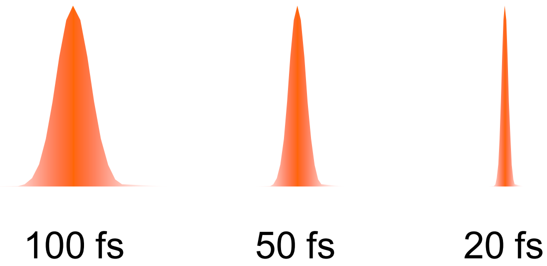
Very intense light

Laser intensities ~ 100 GW/cm^2
1×10^{11} W/cm^2

Laser pointer: 1 mW/cm^2 (1×10^{-3} W/ cm^2)

fs-laser micromachining

Ti:Sapphire lasers



Very intense light

Nonlinear Optical Phenomena

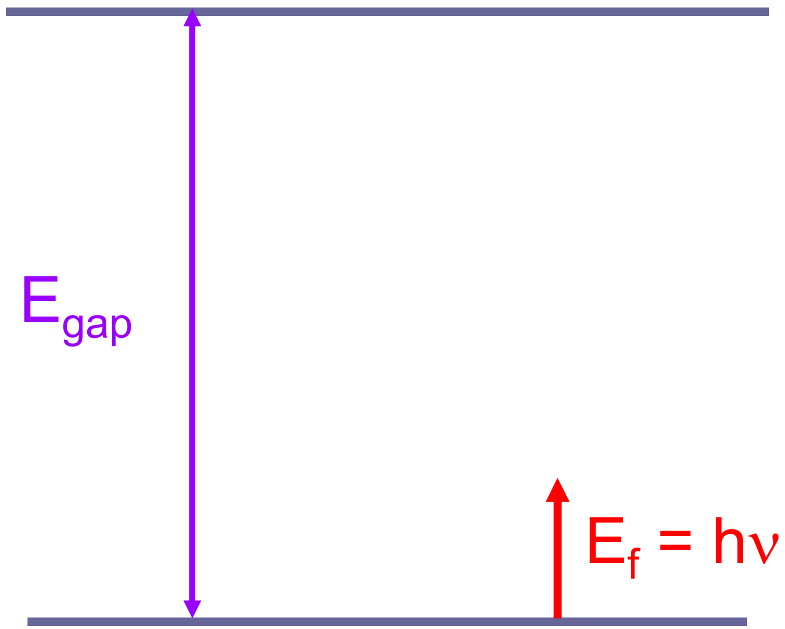
fs-laser microfabrication

photon energy < bandgap

nonlinear interaction

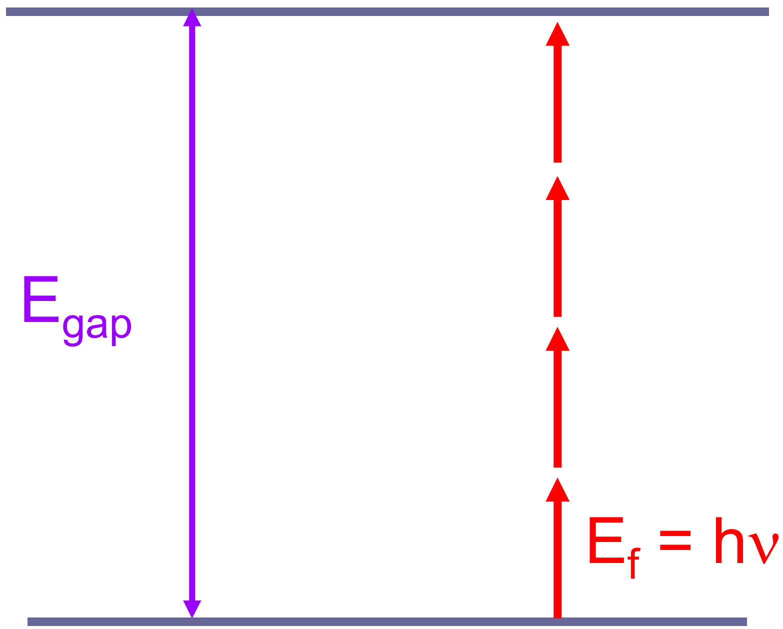
fs-laser microfabrication

nonlinear interaction



fs-laser microfabrication

nonlinear interaction



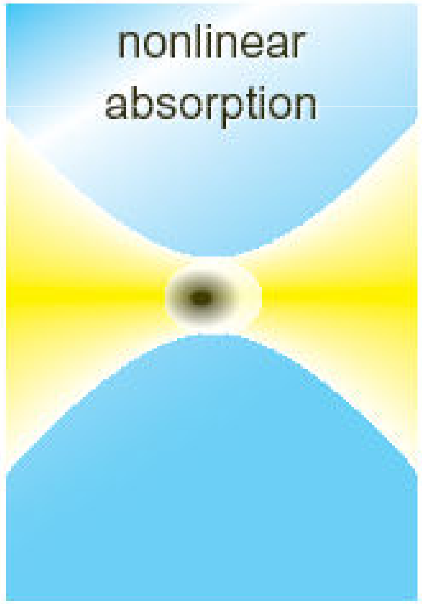
multiphoton absorption

Multi-photon absorption

Nonlinear interaction provides spatial confinement of the excitation

fs-microfabrication

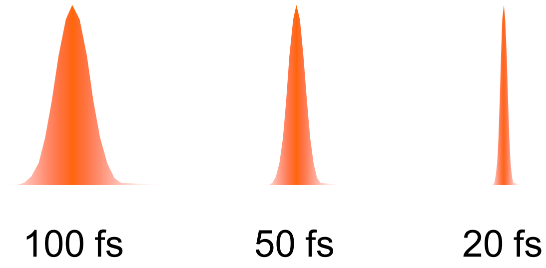
$$\alpha = \alpha_0$$



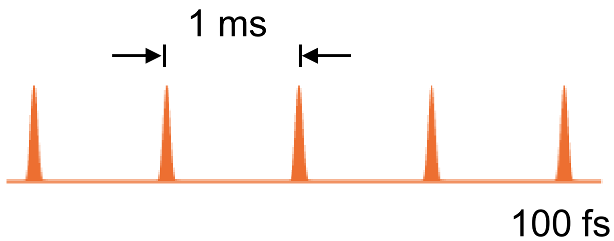
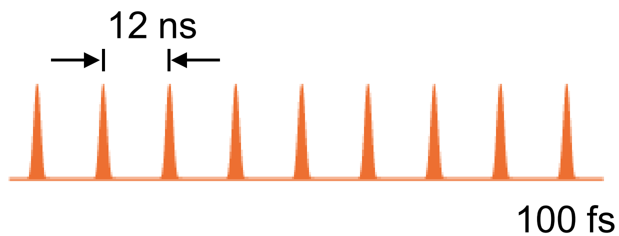
$$\alpha = \alpha_0 + \beta I$$

femtosecond pulses

Ti:Sapphire lasers

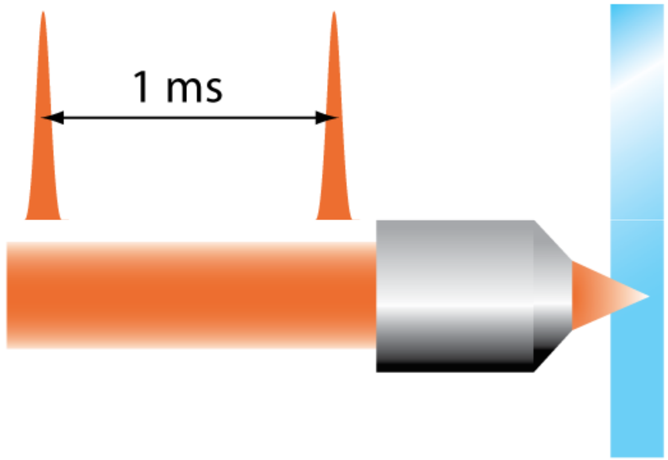


1 fs = 10^{-15} s

Repetition rate		Energy
1 KHz		mJ
86 MHz		nJ

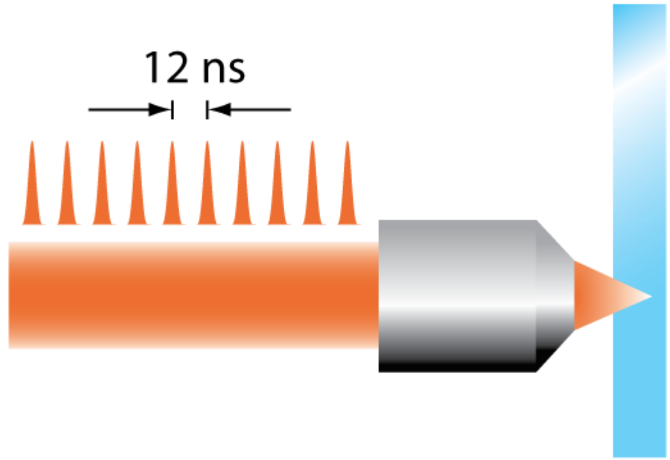
femtosecond pulses

amplified laser



repetitive

oscillator

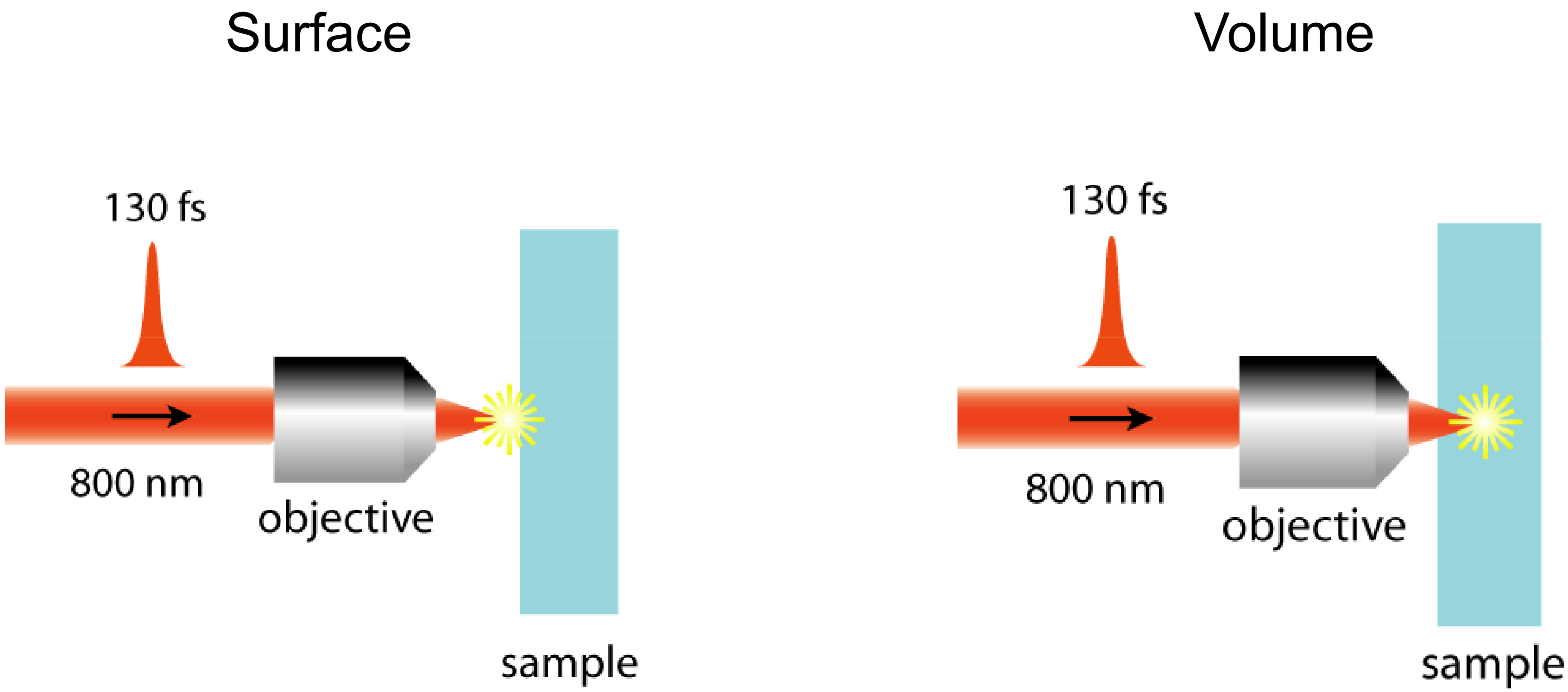


cumulative

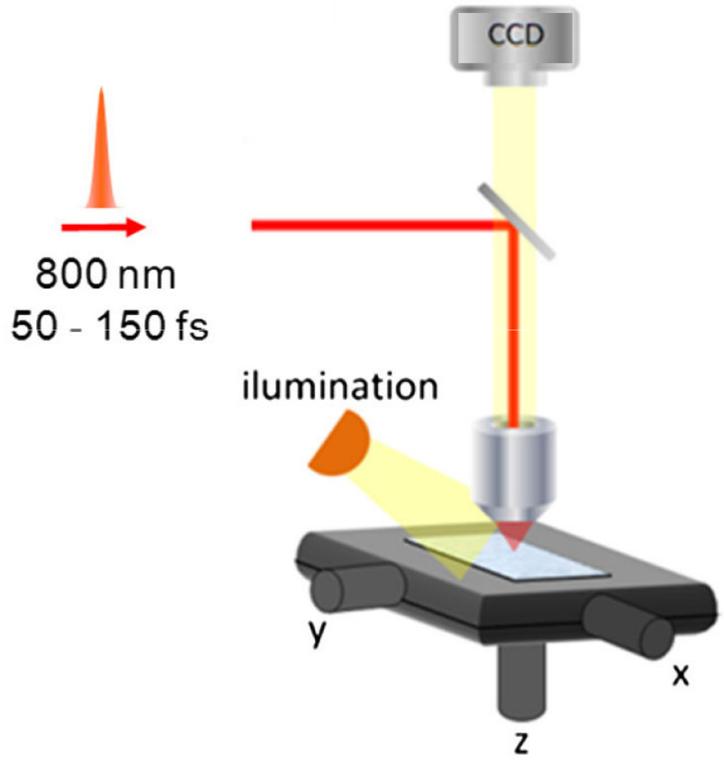
two main techniques

- fs-laser micromachining
- microfabrication via two-photon polymerization

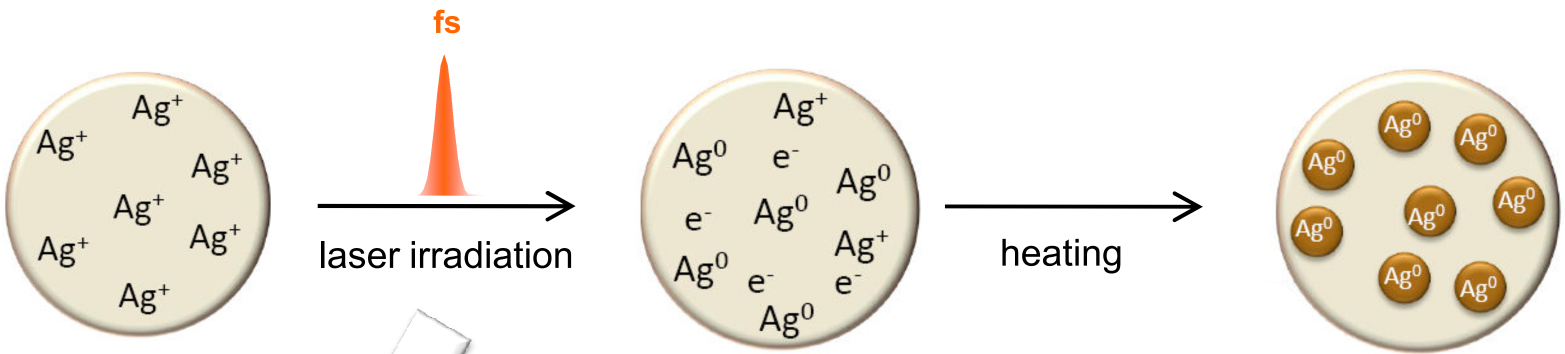
fs-laser micromachining



fs-laser microstructuring experimental setup



Generation of Ag nanoparticles



Ag nanoparticles are generated only in the irradiated area due to the **fs-laser induced photoreduction**

Free electron generation

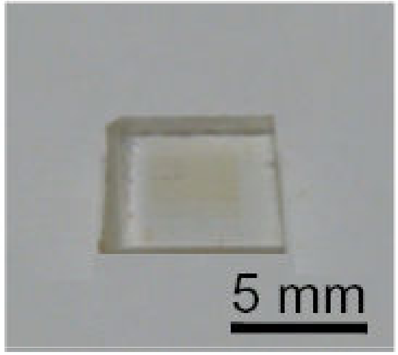
Photoreduction reaction

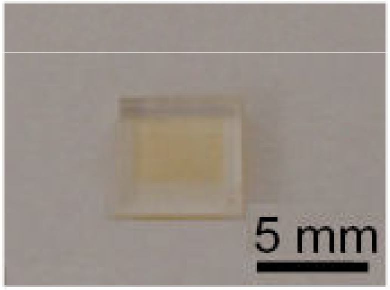
$Ag^{+} + e^{-} \rightarrow Ag^{0}$

Generation of Ag nanoparticles

Silver doped barium borate glass (Ag:BBO)

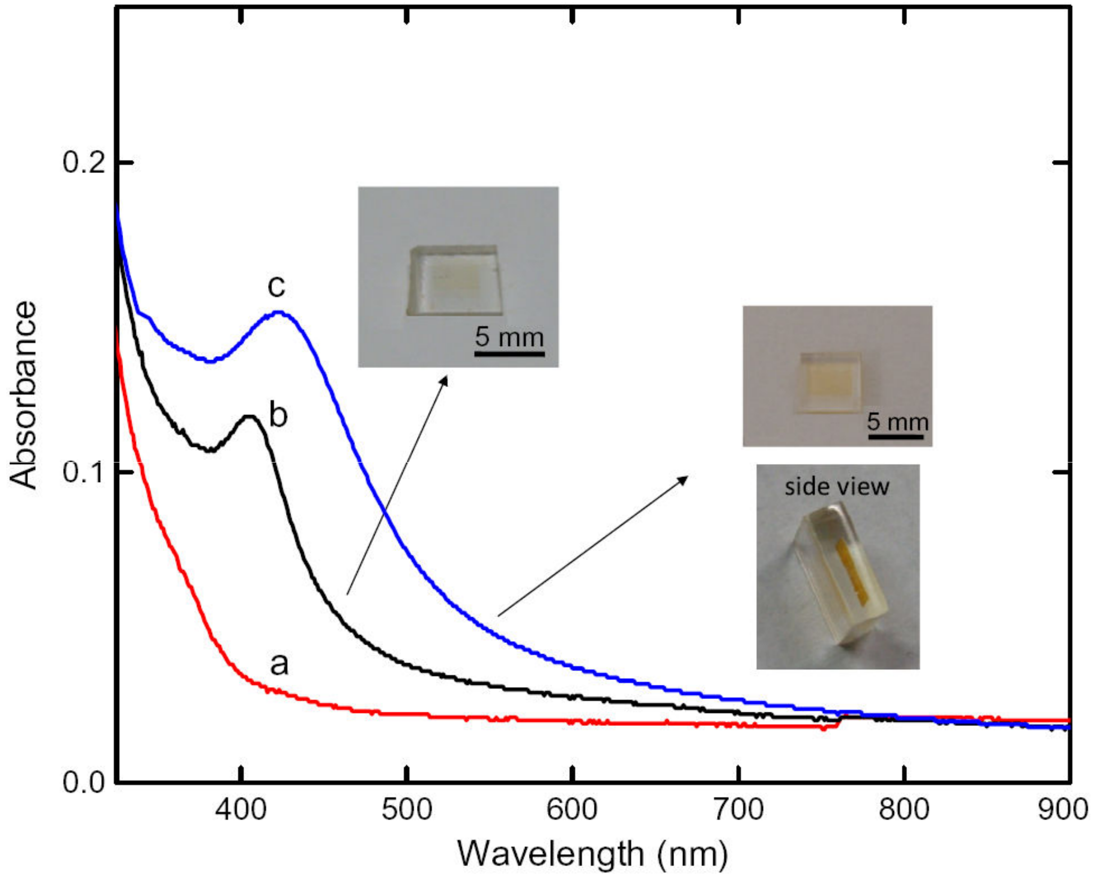
Sample after irradiation with the amplified fs-laser (1 kHz) and subsequent thermal treatment at 400 C for 1 h





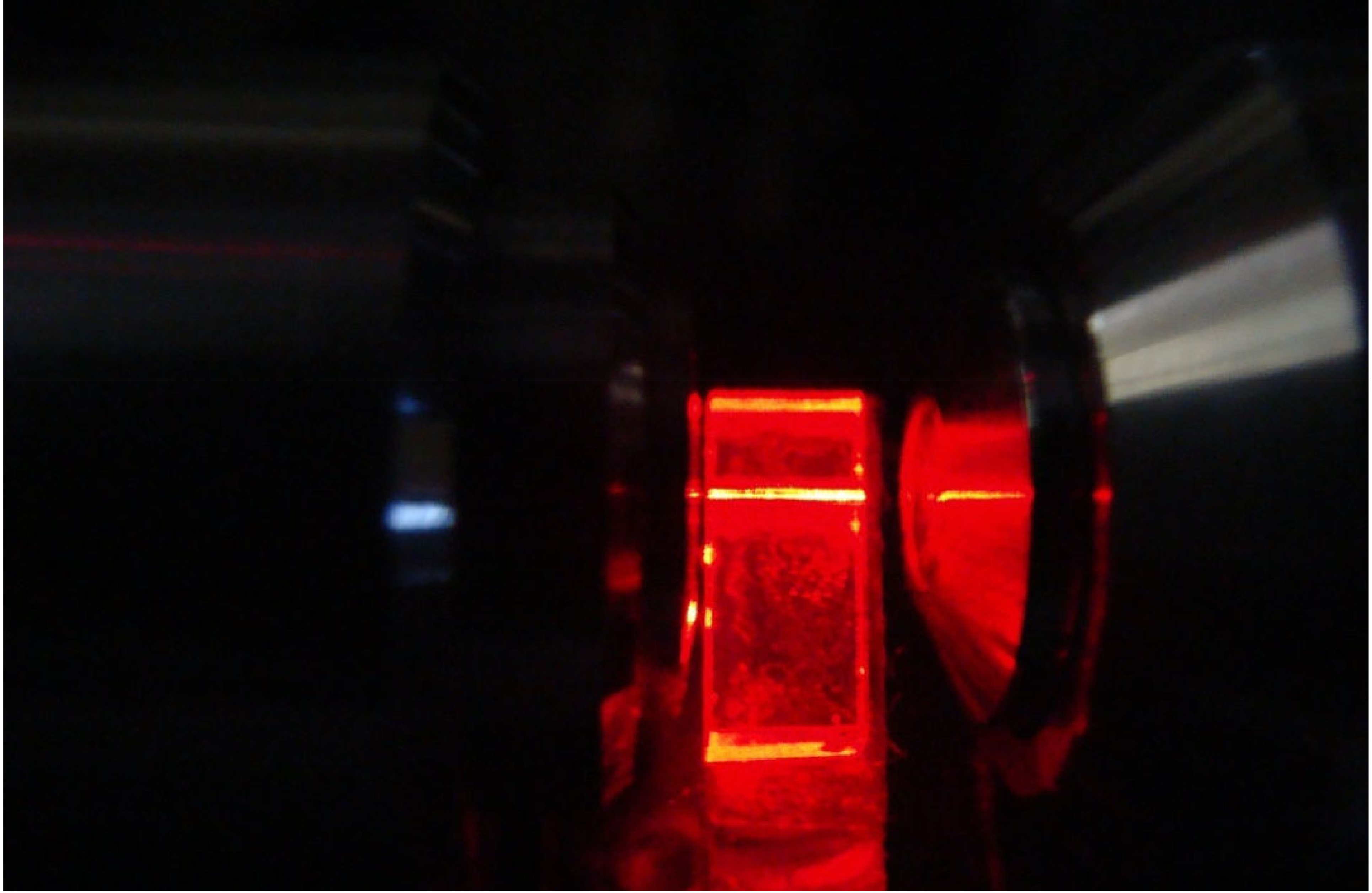
Sample after irradiation with the 5 MHz fs-laser

Generation of Ag nanoparticles



Absorption spectrum of the Ag:BBO sample as prepared (a), after irradiation with the 5 MHz fs-laser (b) and after irradiation with the amplified fs-laser (1 kHz) and subsequent thermal treatment.

fs-laser waveguides fabrication



Waveguides fabrication

Sample:

Ag:P7W3

Tungsten lead pyrophosphate glass - $(70Pb_2P_2O_7\text{-}30WO_3)\text{:}1AgCl$ (%mol)

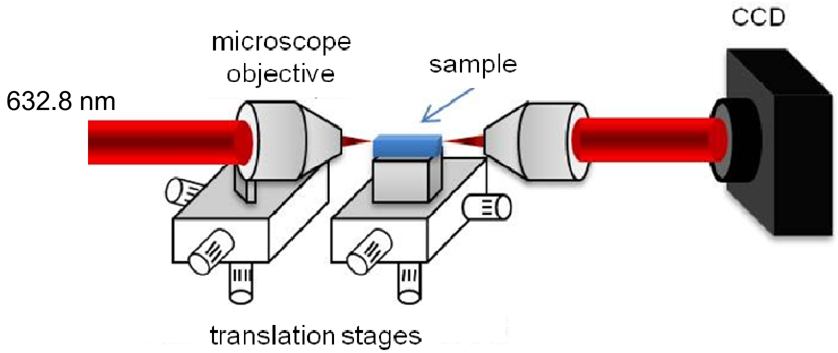
Waveguides fabricated using the 5-MHz laser system (50 fs) with 37 nJ/pulse and v = 10 μm/s

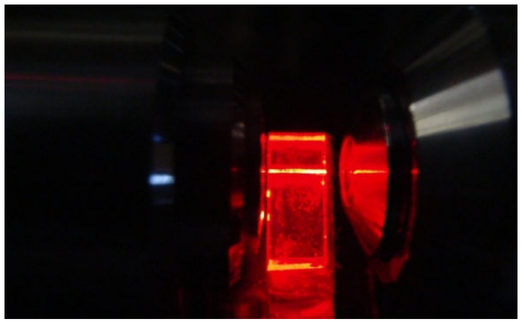
Top view

Cross-section view

Waveguides fabrication

Coupling light into the waveguides





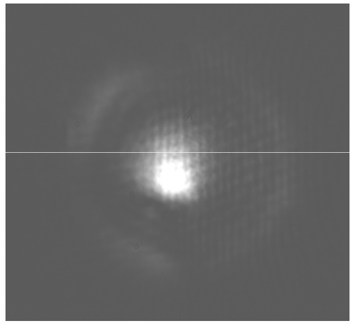
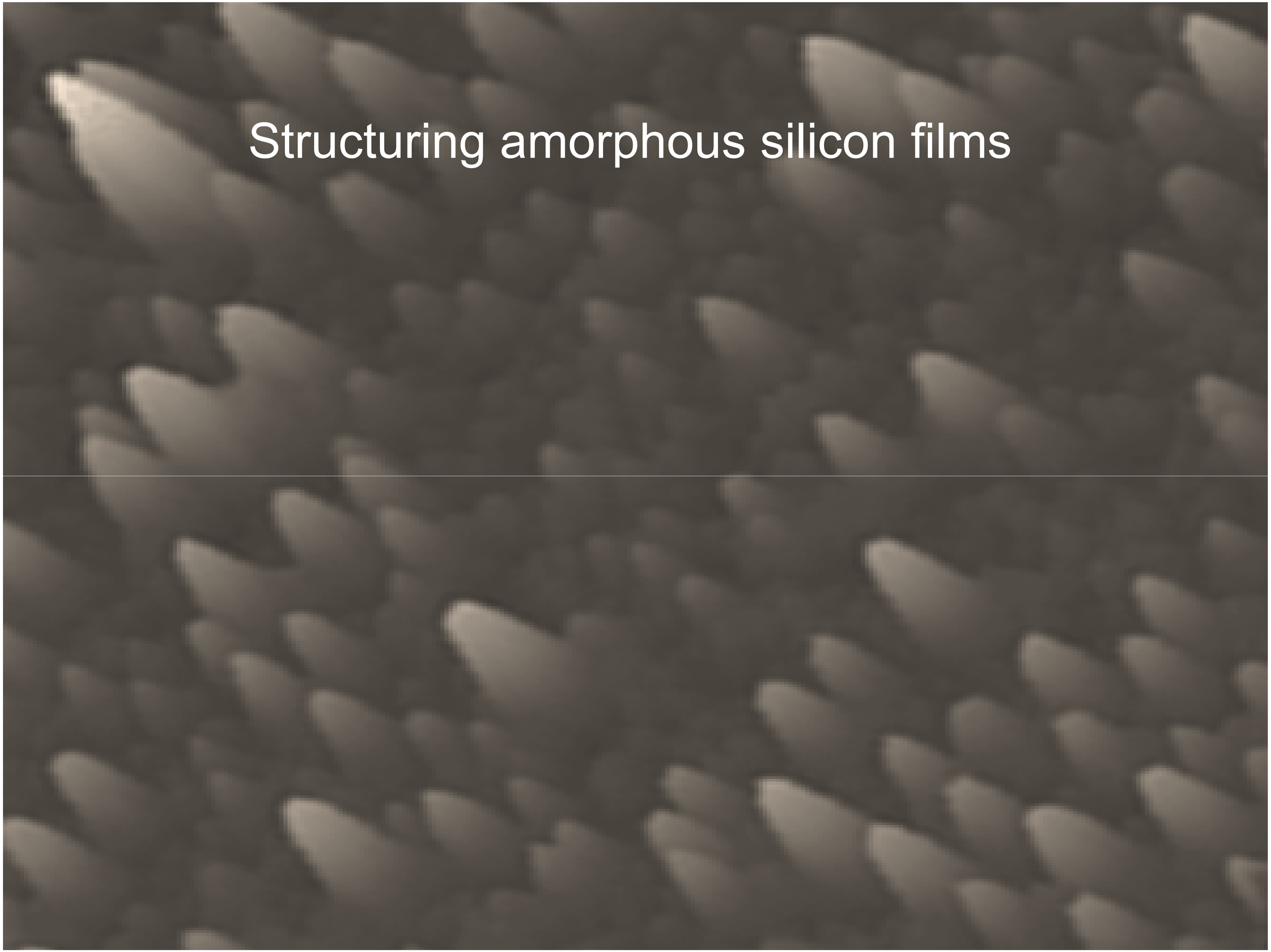


image of the waveguide output

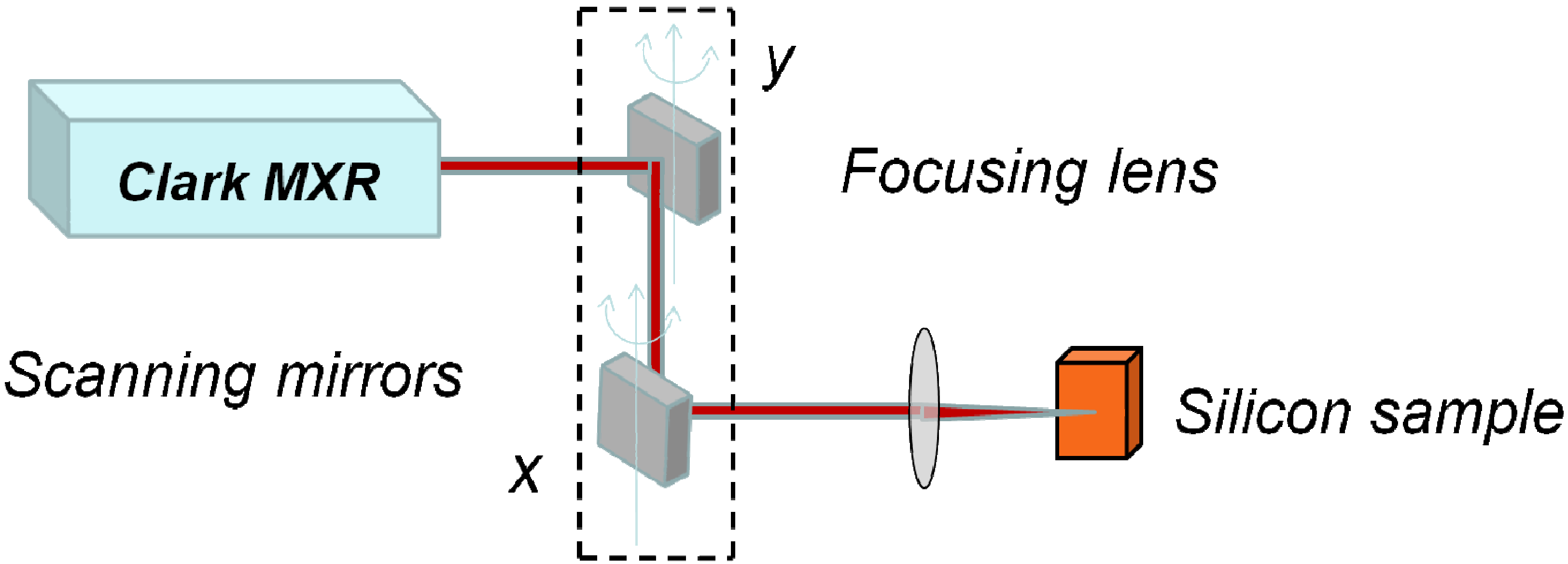
measured waveguide loss L = 1.3 dB/mm

Structuring amorphous silicon films



structuring amorphous Si surface

Experimental setup uses a pair of scanning mirrors

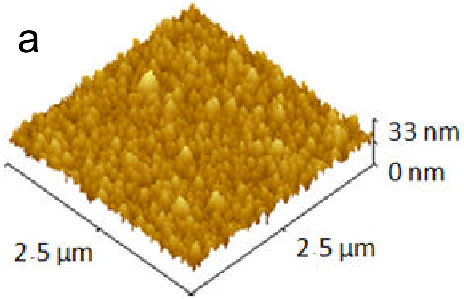


150 fs, 775 nm, 1 KHz, v = 5 mm/s, f= 20 cm

structuring amorphous Si surface

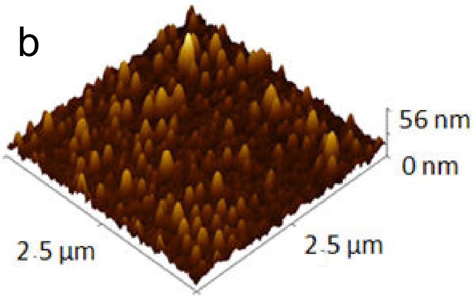
AFM micrographs of aSi microstructures at different laser intensities

a



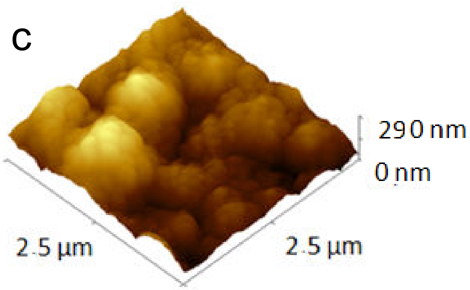
before irradiation

b



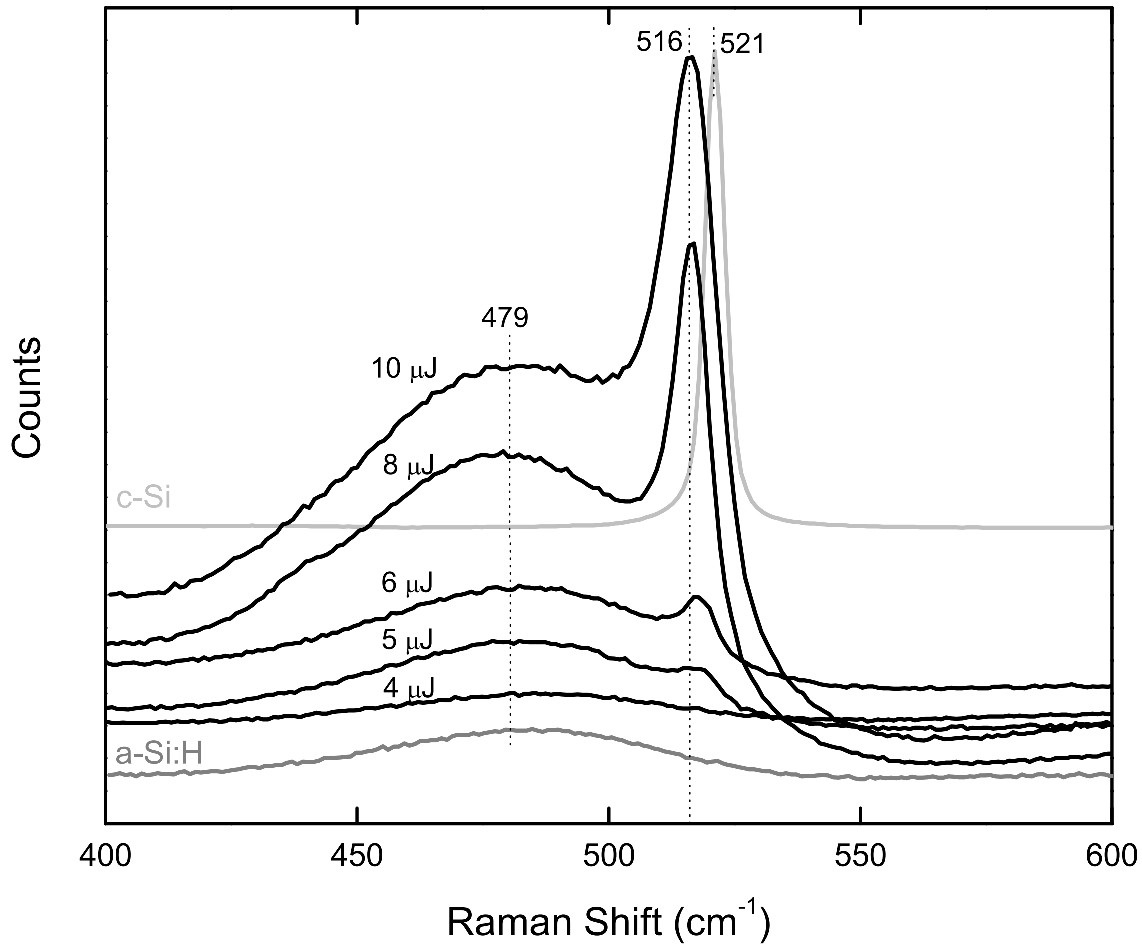
E = 5 µJ/pulse

c



E = 8 µJ/pulse

structuring amorphous Si surface



Micro-Raman analysis reveals the crystallization of the aSi upon fs-laser irradiation

structuring amorphous Si surface

The crystalline volume fraction

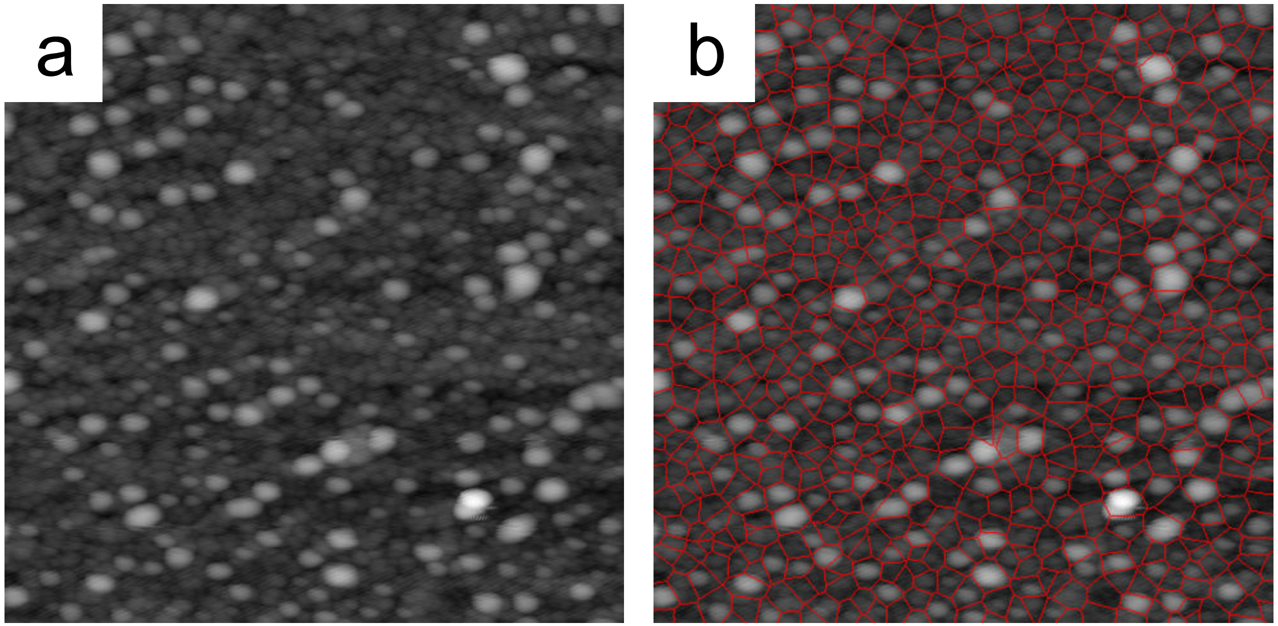
$$X_c = I_{cp} / \left(I_{cp} - \sigma I_{ap} \right)$$

in which I_{cp} and I_{ap} are the intensities of the crystalline peak and at the center of the amorphous band

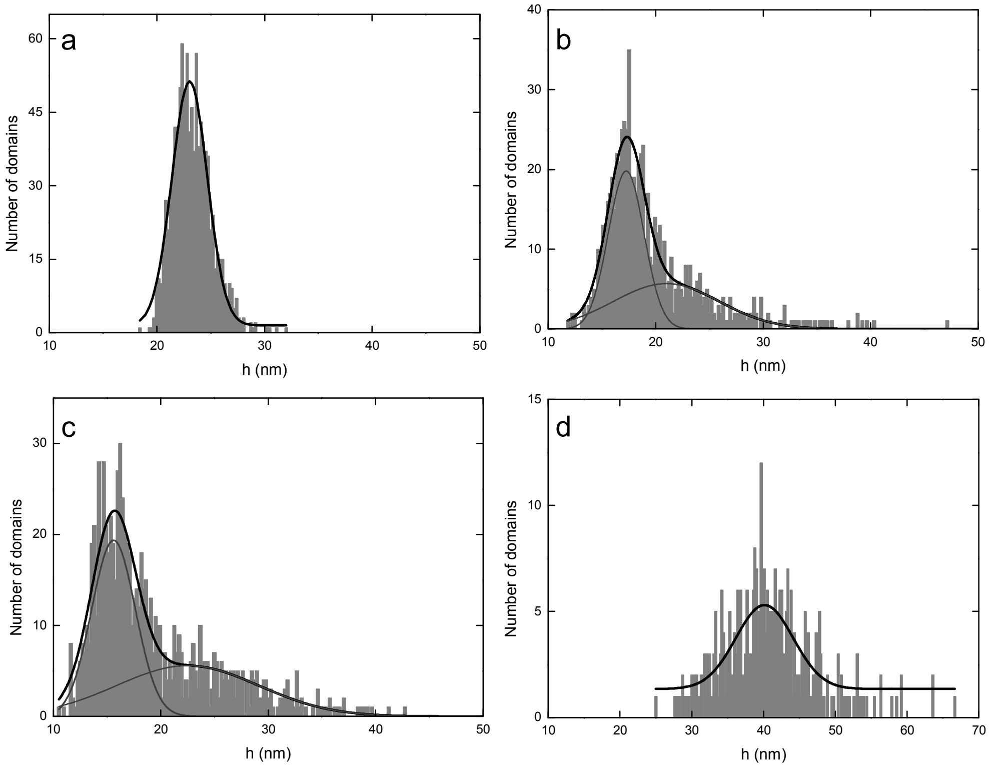
Pulse energy (μJ)	X_c (%)	Raman peak (cm^{-1})	Nanocrystal diameter (nm)
4	0	-	-
5	43	519	9
6	48	518	6
8	62	517	5
10	73	516	4

structuring amorphous Si surface

AFM image of a a-Si:H sample irradiated with 5 μJ (a) and its corresponding segmentation obtained using the Voroni´s diagram method (b).

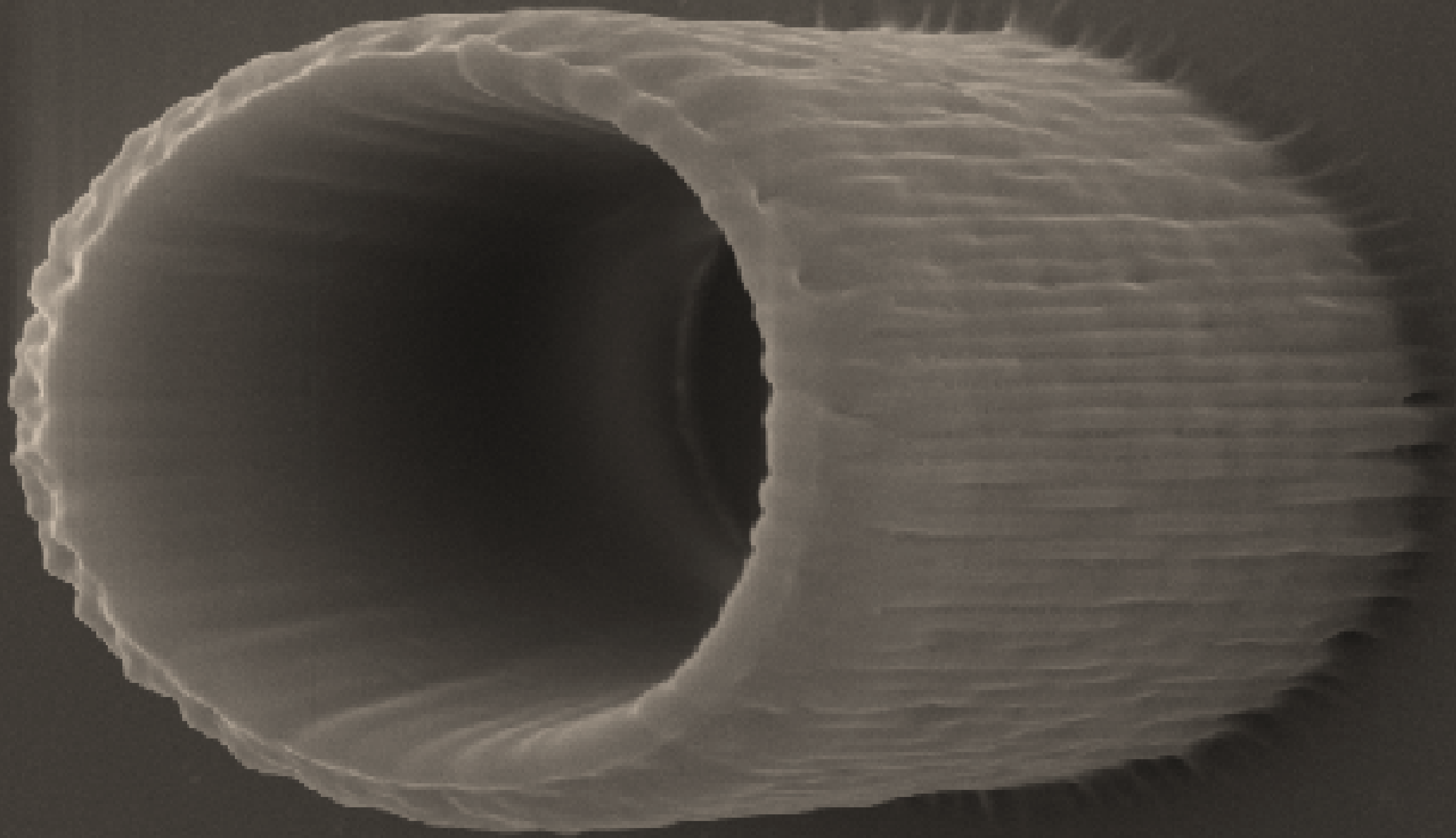


structuring amorphous Si surface



Height histograms of the domains on the sample surface
(a) before laser irradiation
(b) E = 4 μJ
(c) E = 5 μJ
(d) E = 6 μJ

fs-laser microfabrication



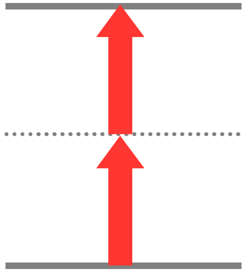
fabrication of microstructure using fs-laser
and nonlinear optical processes

Two-photon polymerization

Monomer + Photoinitiator $\rightarrow$ Polymer

light

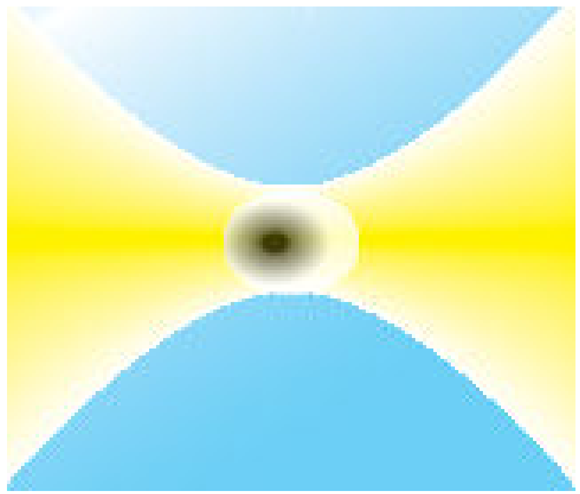
Photoinitiator is excited by **two-photon absorption**



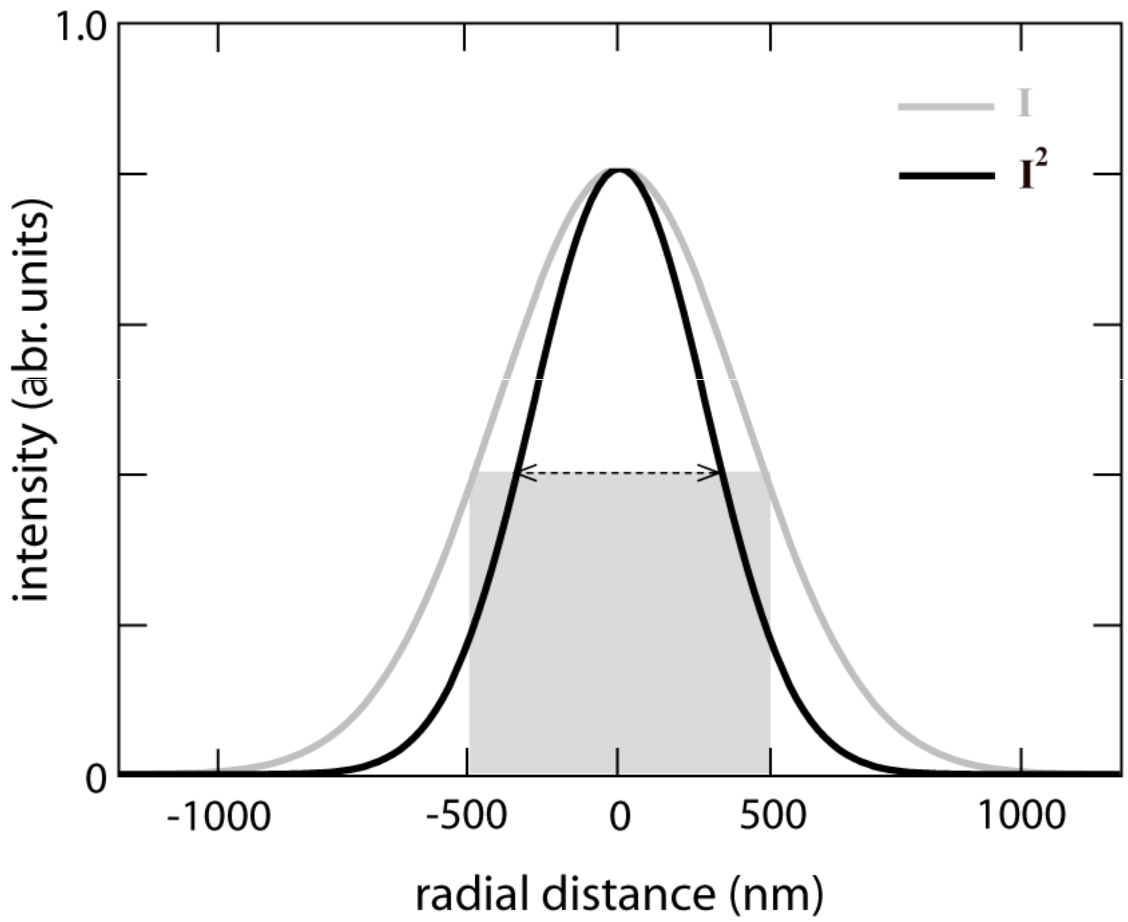
$$R_{2PA} \propto I^2$$

The polymerization is confined to the focal volume.

High spatial resolution

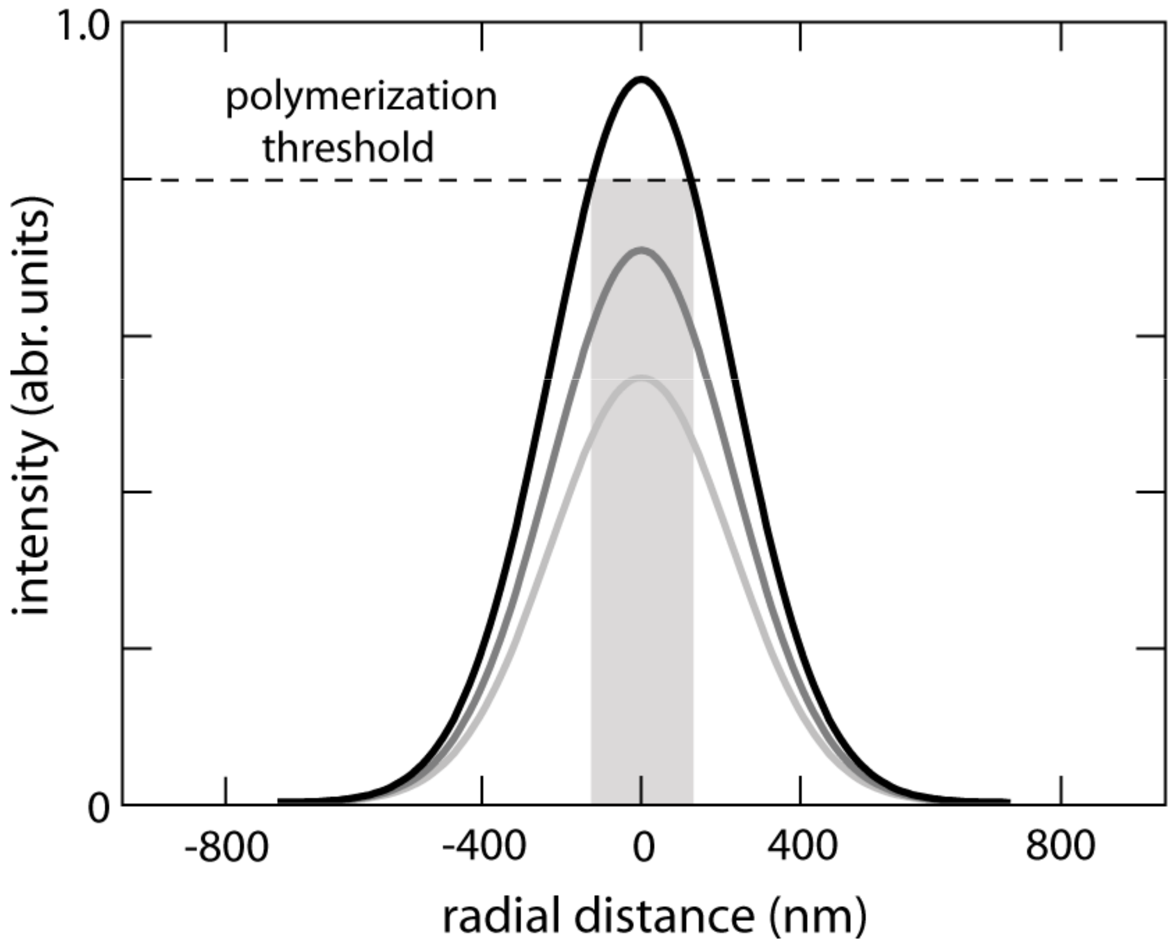


Two-photon polymerization



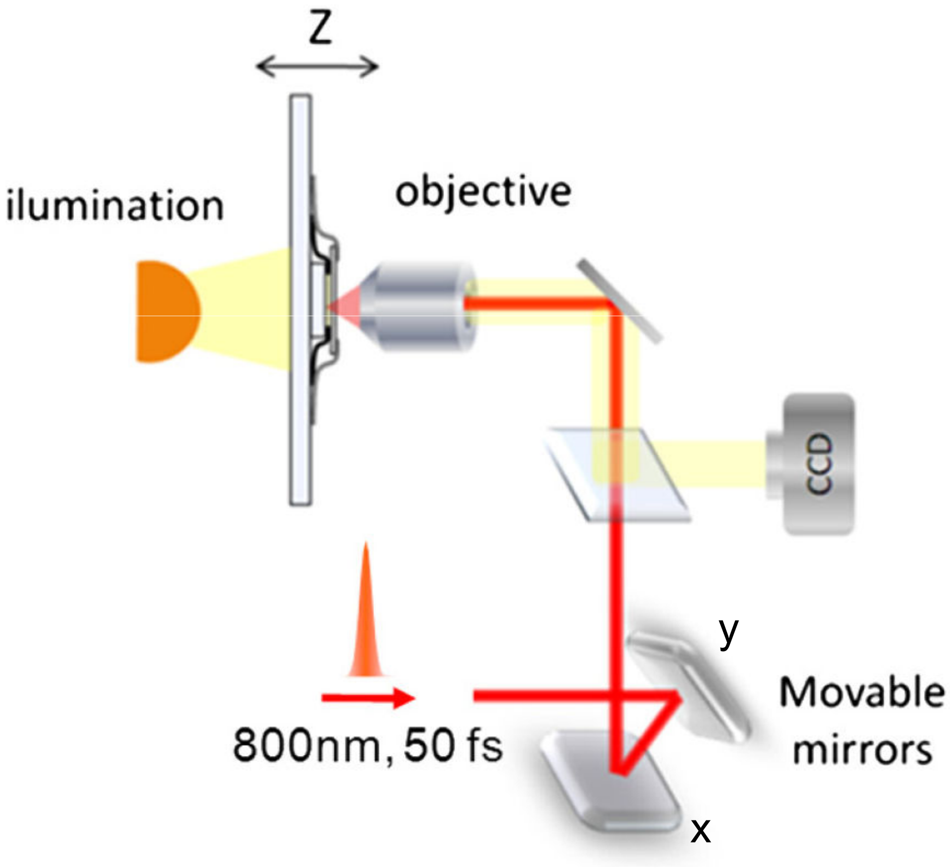
bellow the diffraction limit

Two-photon polymerization



even higher spatial resolution

Two-photon polymerization setup



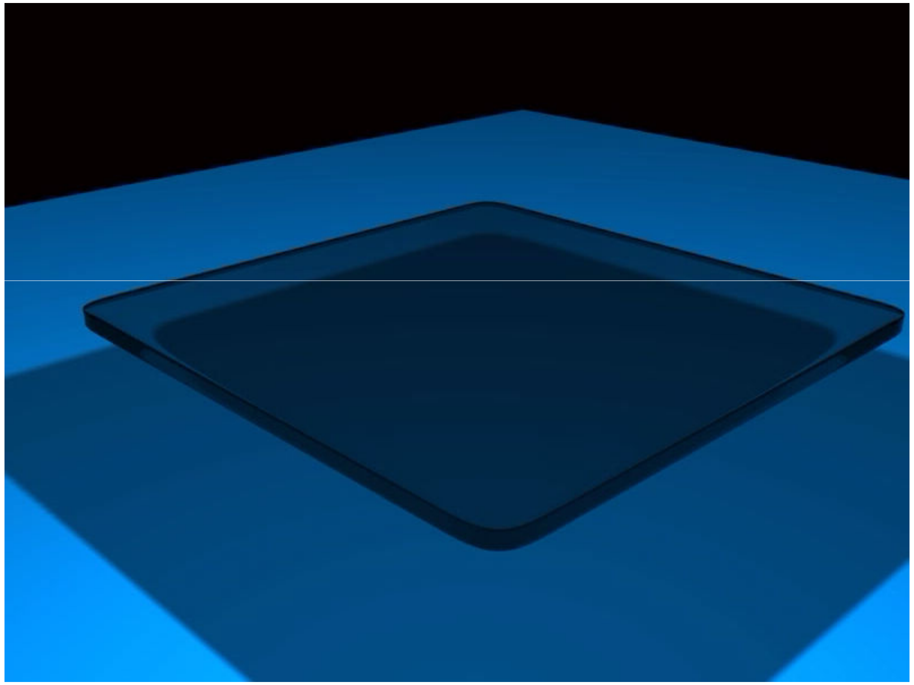
Ti:sapphire laser oscillator

- 50 fs
- 800 nm
- 80 MHz
- 20 mW

Objective

40 x
0.65 NA

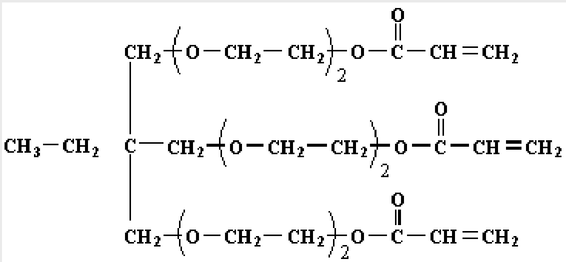
Two-photon polymerization



Resin preparation

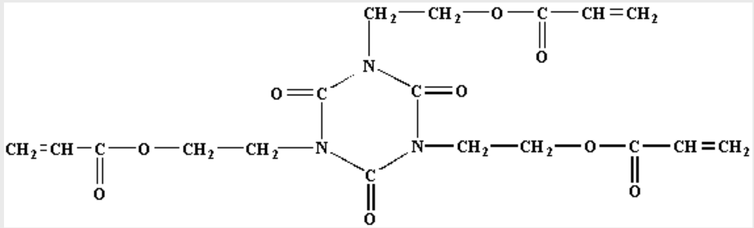
Monomers

Monomer A



reduces the shrinkage upon polymerization

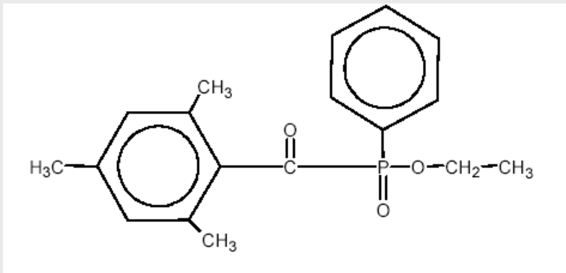
Monomer B



gives hardness to the polymeric structure

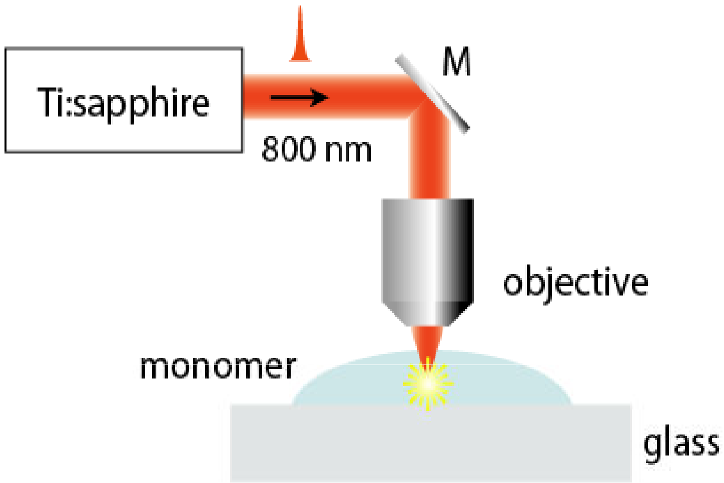
Photoinitiator

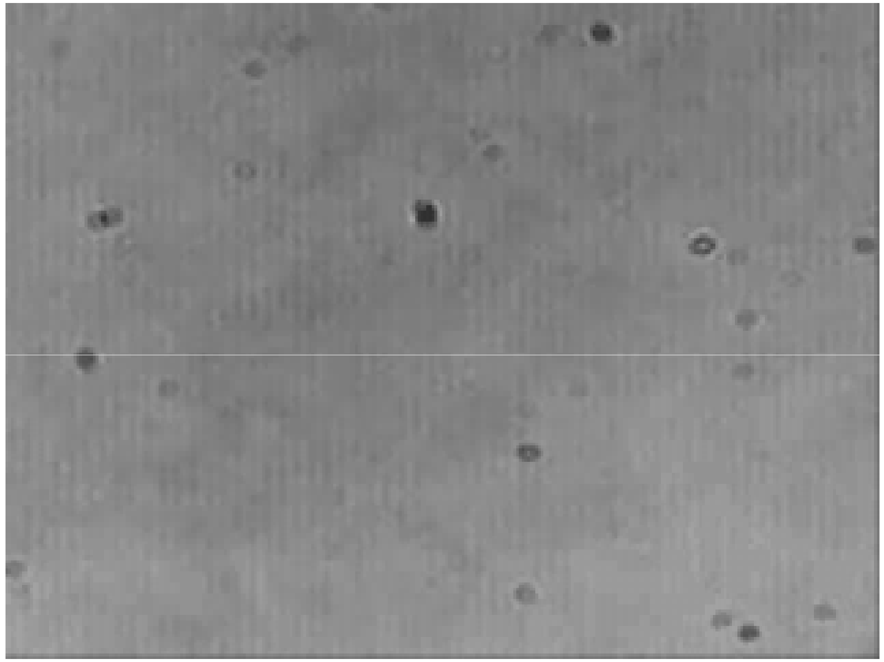
Lucirin TPO-L



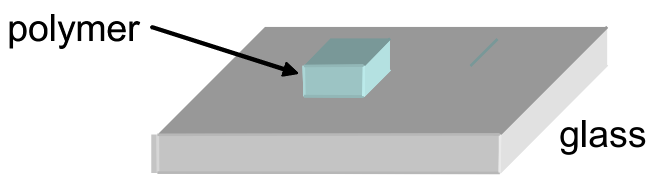
Appl. Phys. A, 90, 633–636 (2008)

Two-photon polymerization

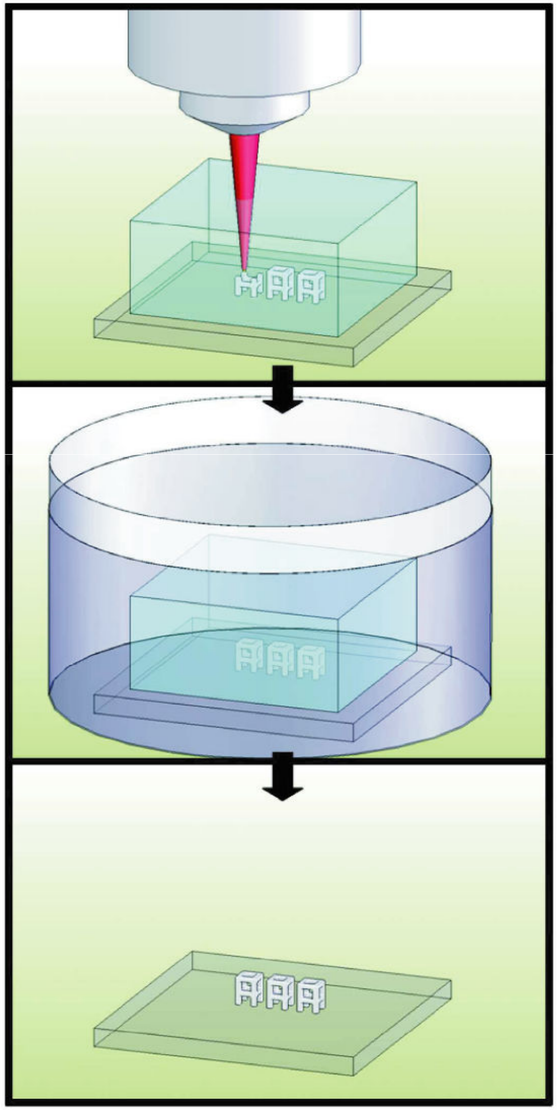




30 μm x 30 μm x 12 μm cube



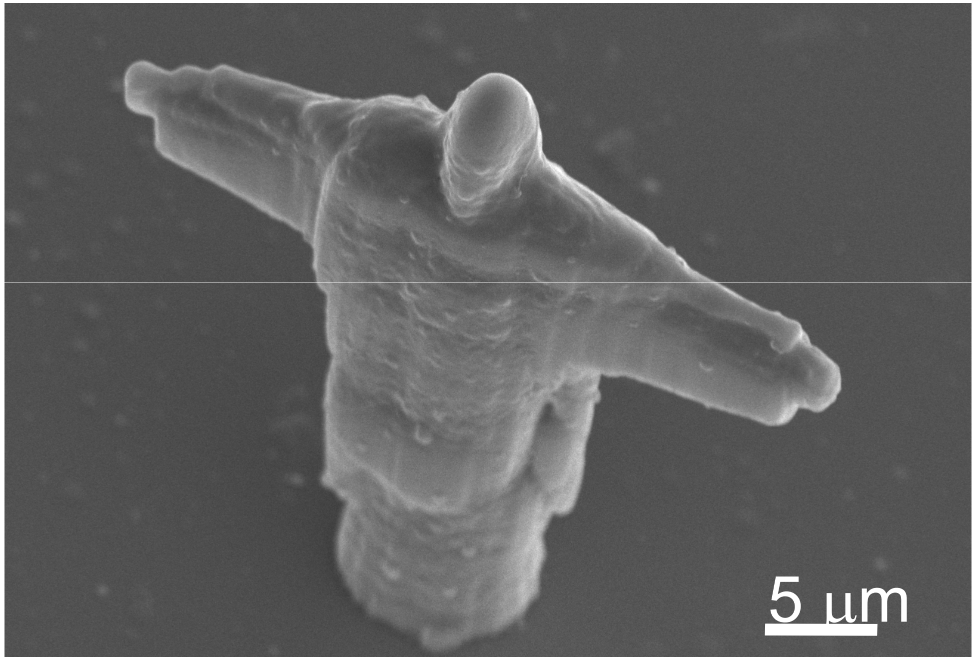
Two-photon polymerization



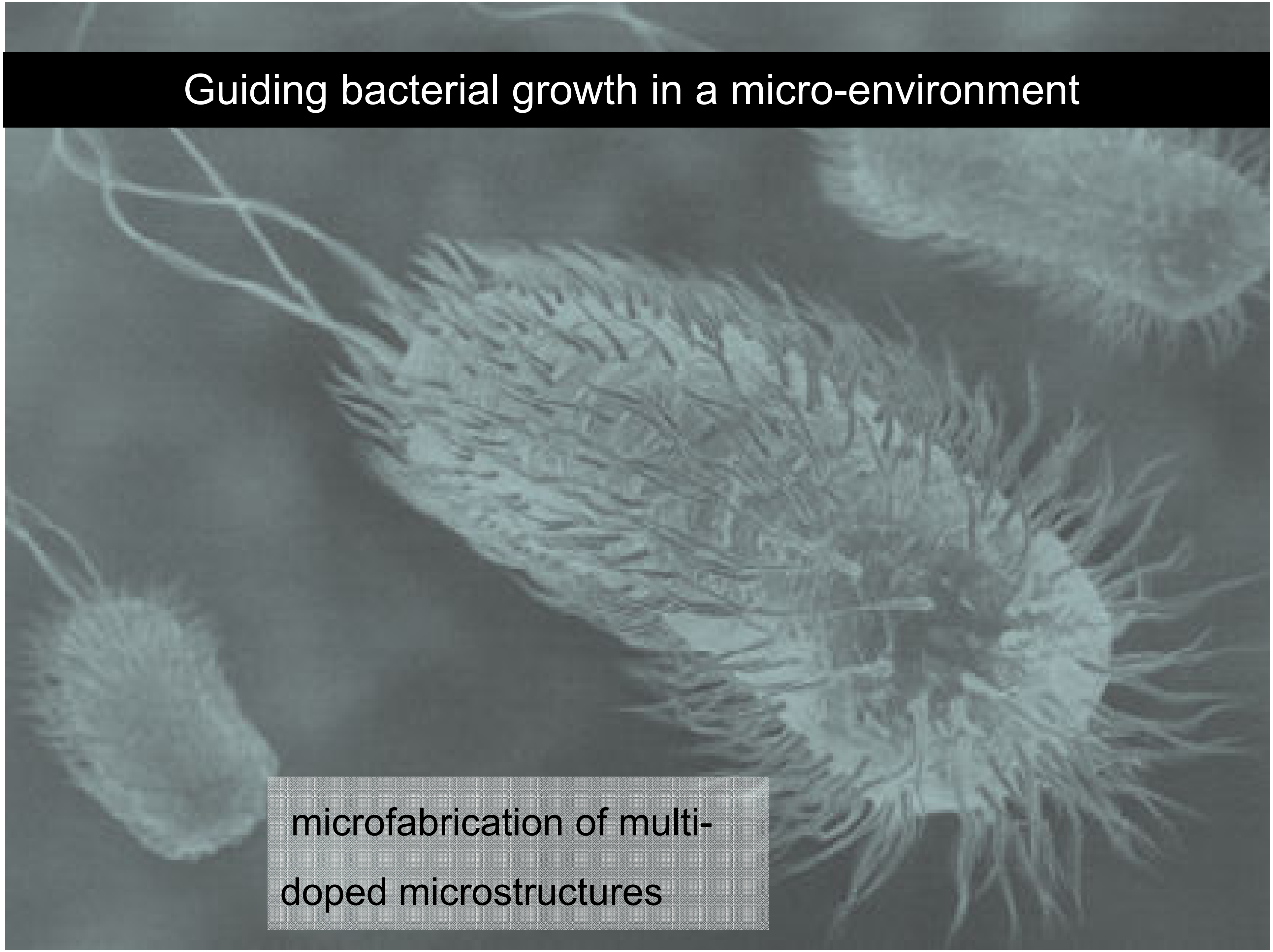
After the fabrication, the sample is immersed in ethanol to wash away any unsolidified resin and then dried

two-photon polymerization

Microstructure fabricated by two-photon polymerization



Guiding bacterial growth in a micro-environment

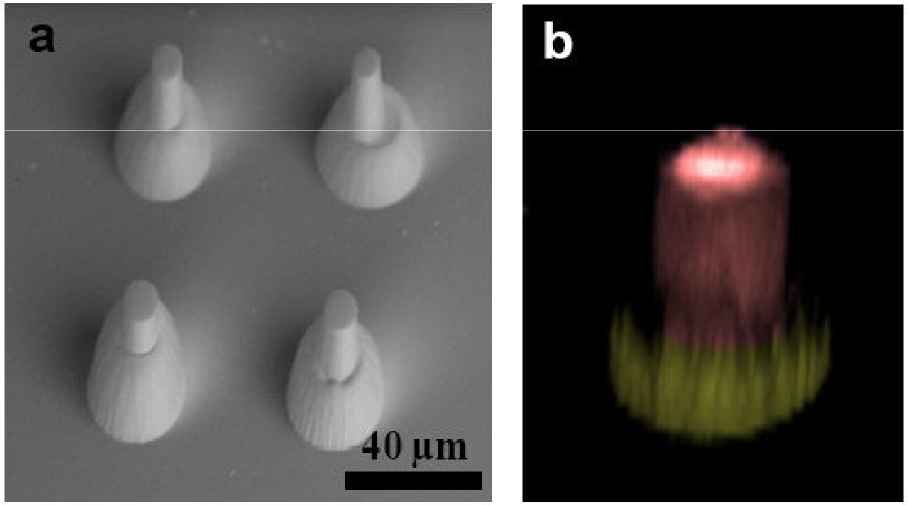


microfabrication of multi-doped microstructures

Guiding bacterial growth in a micro-environment

to study bacterial growth it was needed to develop
double doped microstructures

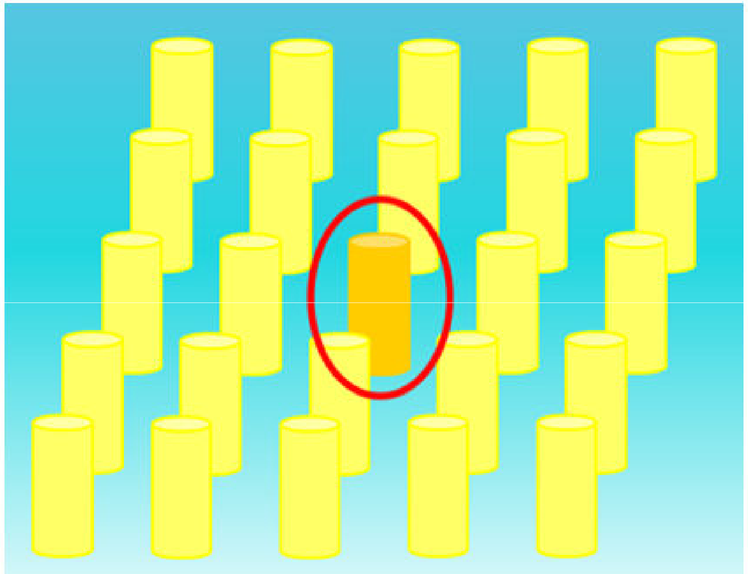
microstructure containing Fluorescein and Rhodamine

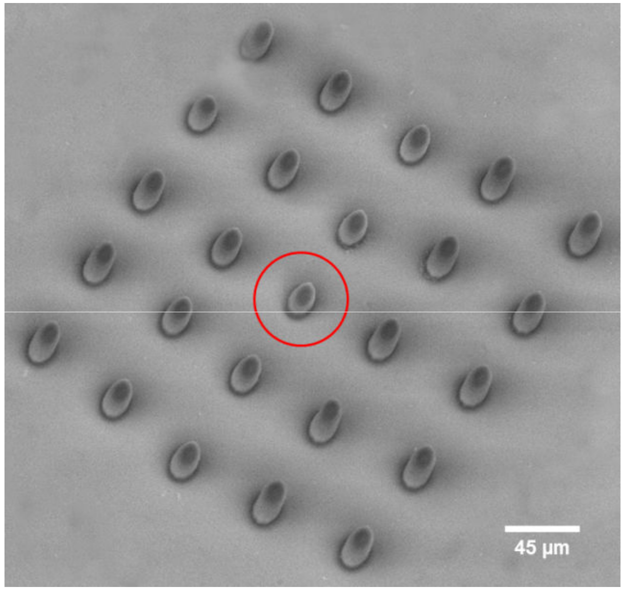


(a) SEM of a double-doped microstructure (top view).
(b) Confocal fluorescent microscopy image of the same microstructure.

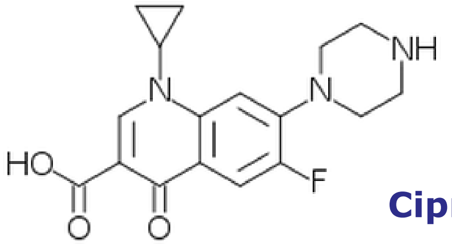
Guiding bacterial growth in a micro-environment

Study the development of E. coli in micro-environments:





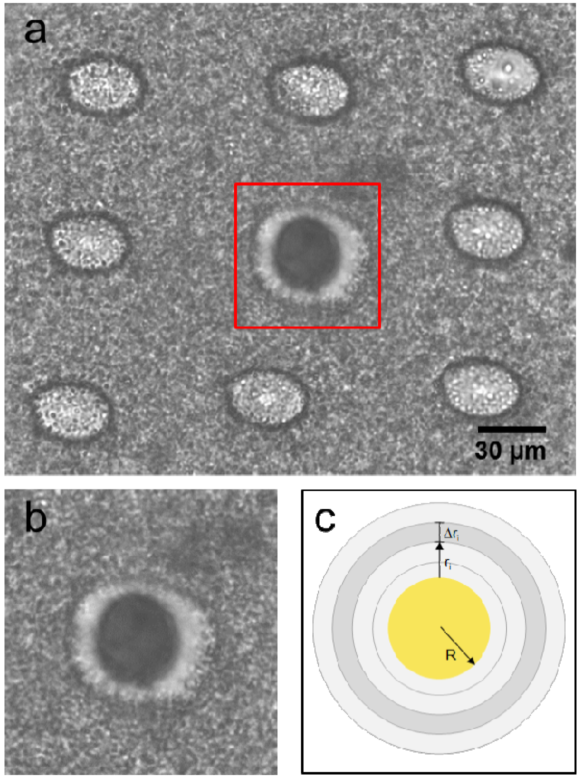
micro-environment in which the central structure contains antibiotic.



Ciprofloxacin

Guiding bacterial growth in a micro-environment

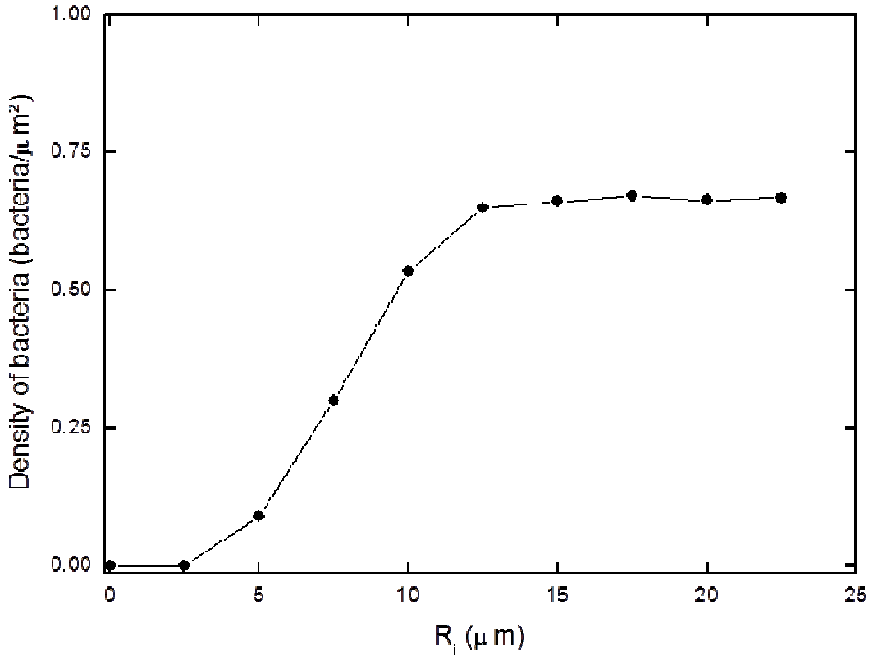
Study the development of E. coli in micro-environments:



after 3 hours, we observed that a small region around the doped structure does not show bacterial growth.

such inhibition zone was analyzed by determining the bacterial density in concentric rings

Guiding bacterial growth in a micro-environment



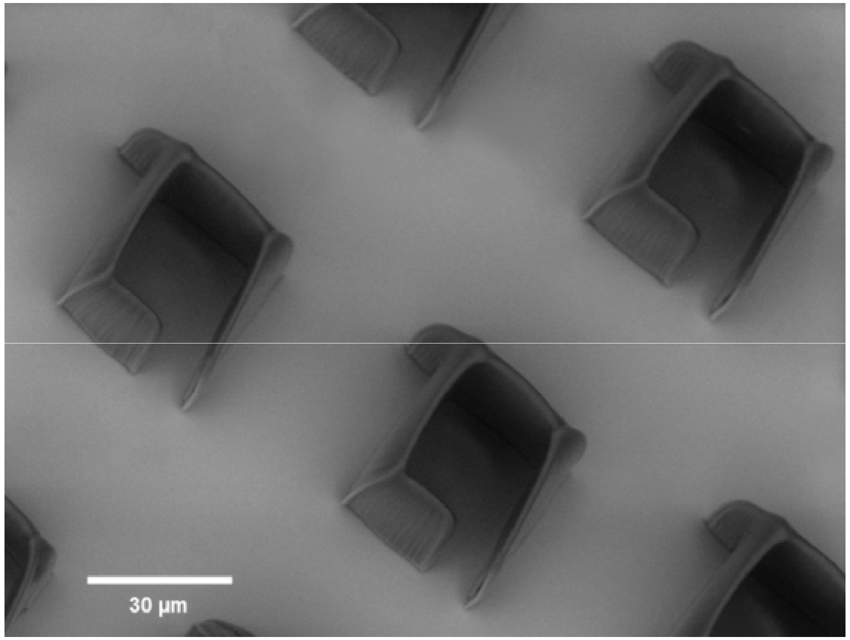
the density of bacteria grows monotonically with r_i

saturating when r_i reaches approximately 12 µm in about 0.7 bacteria/μm^2

the inhibition zone has a maximum range of approximately 10 µm, being more effective as one gets closer to the microstructure impregnated with ciprofloxacin

Guiding bacterial growth in a micro-environment

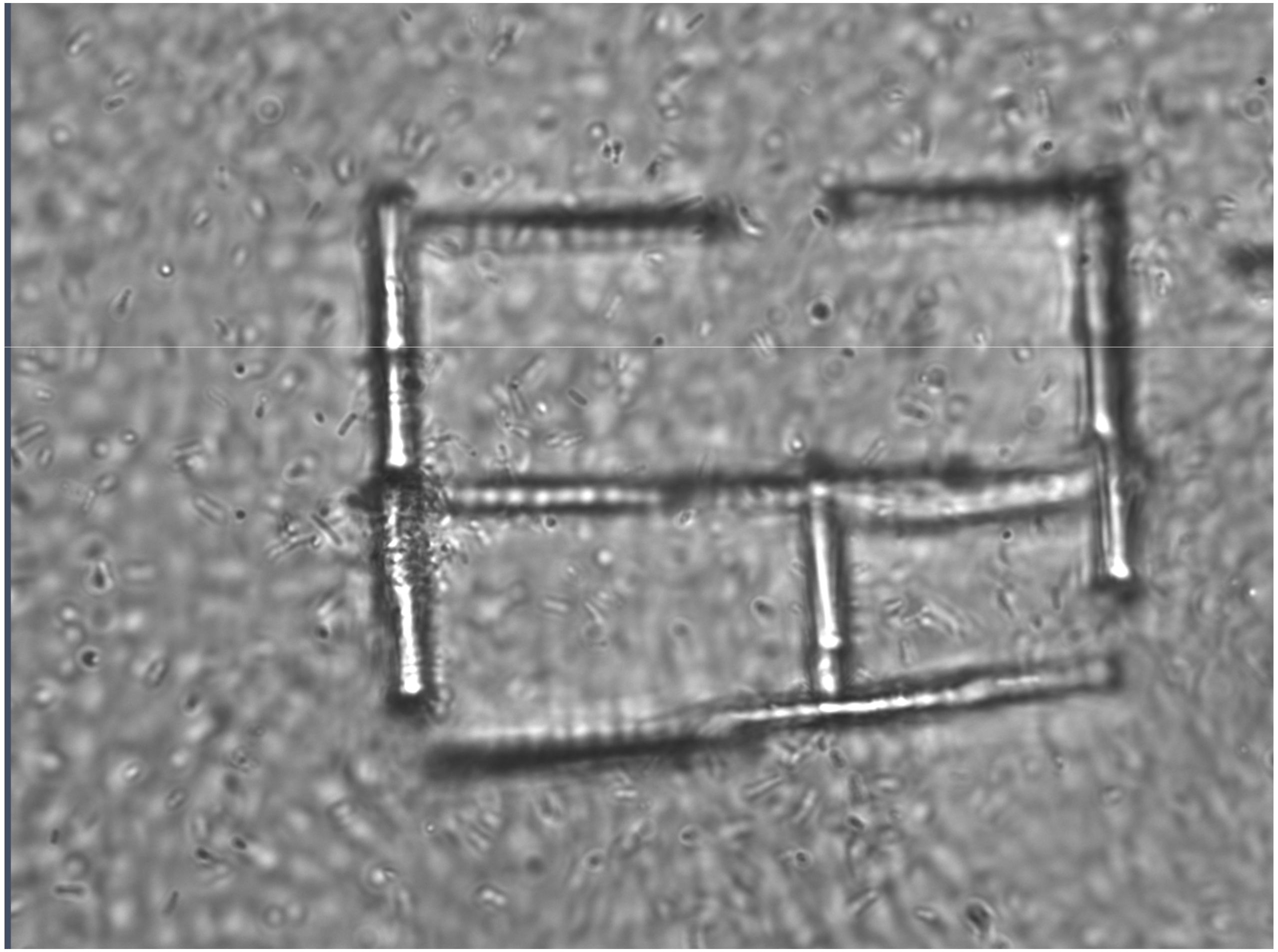
Bacteria microtraps



using micro-environments to study the dynamics of bacterial migration

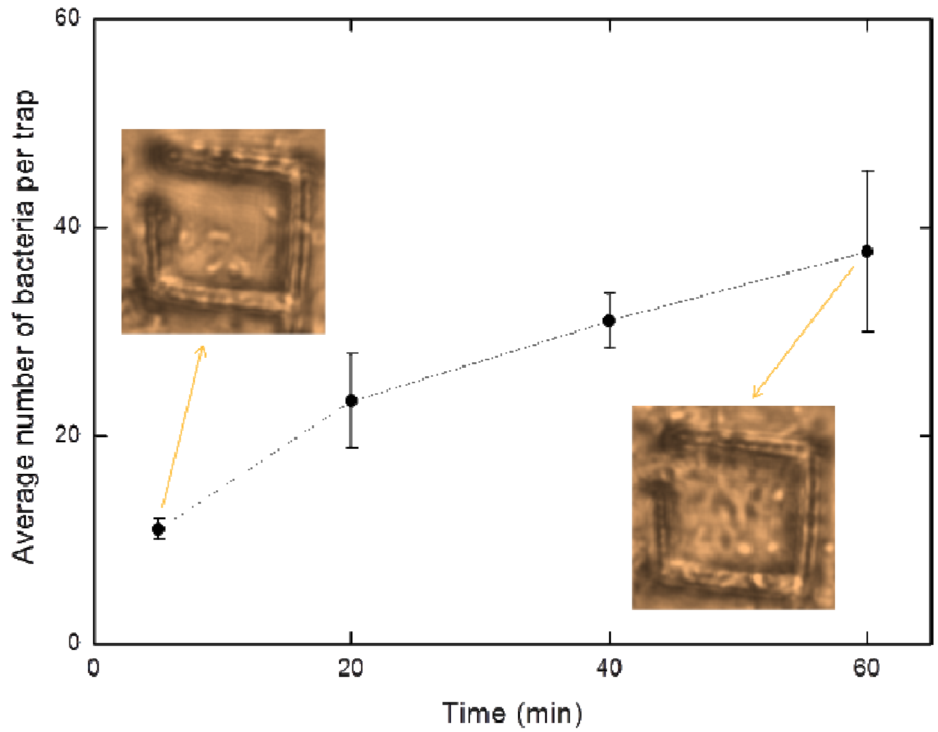
Guiding bacterial growth in a micro-environment

Bacteria microtraps



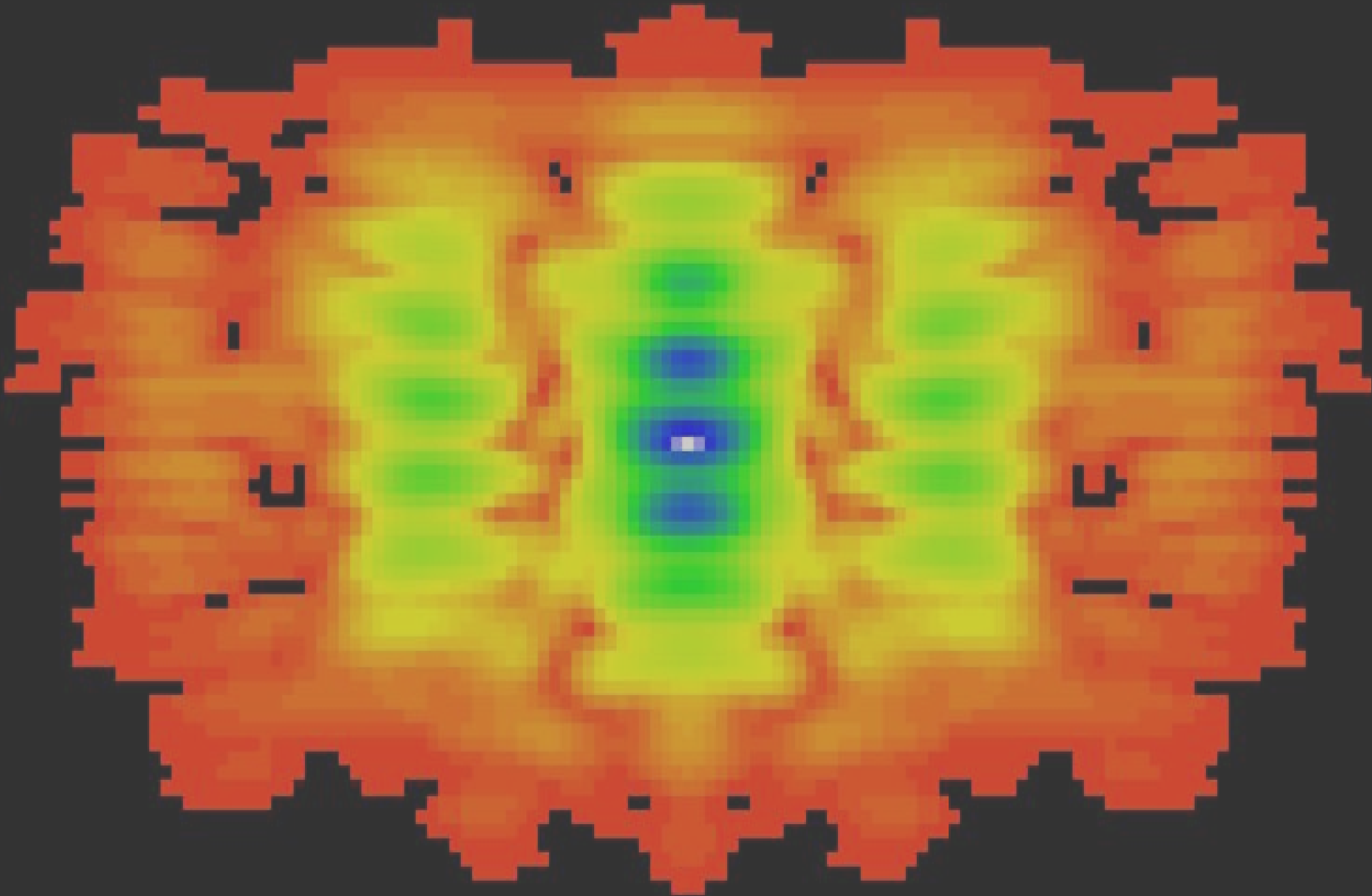
Guiding bacterial growth in a micro-environment

Bacteria microtraps



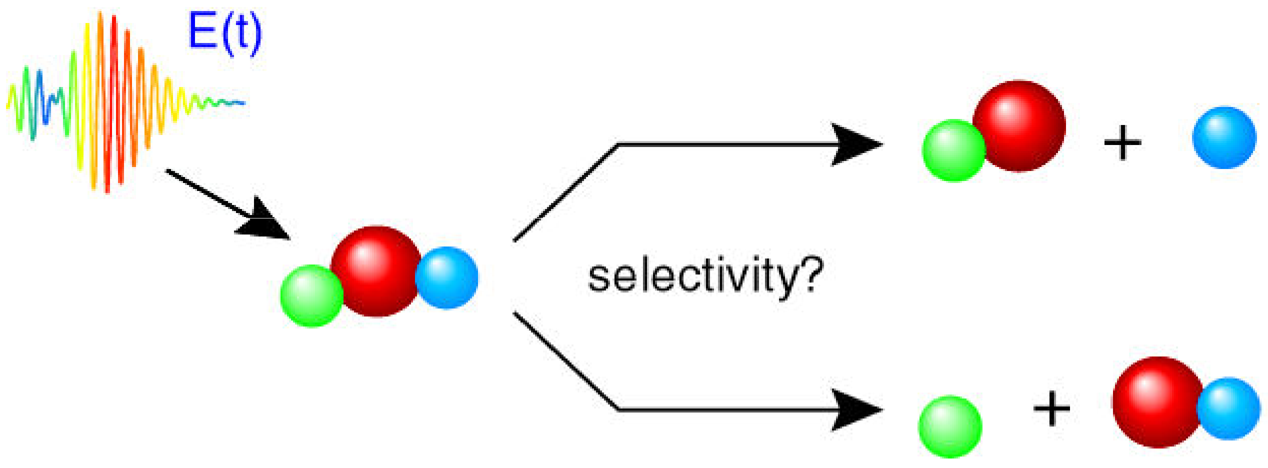
using micro-environments to study the dynamics of bacterial migration

Shaping fs-pulses



shaped fs pulses

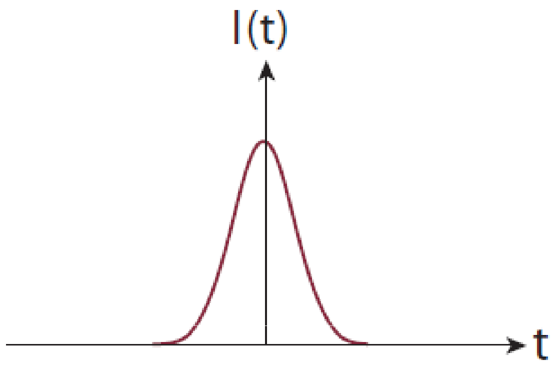
Can we used shaped fs-pulses to drive a given optical phenomenon to a specific results ?

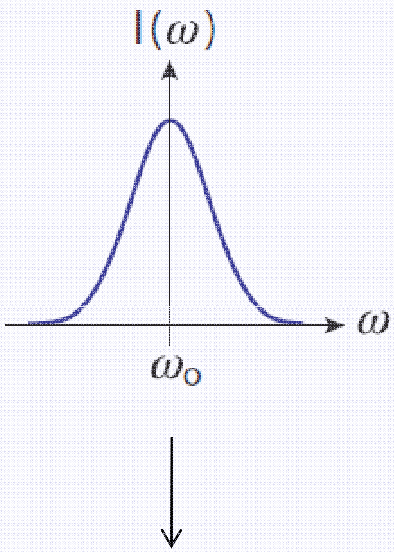


E(t) ?

shaped fs-pulses

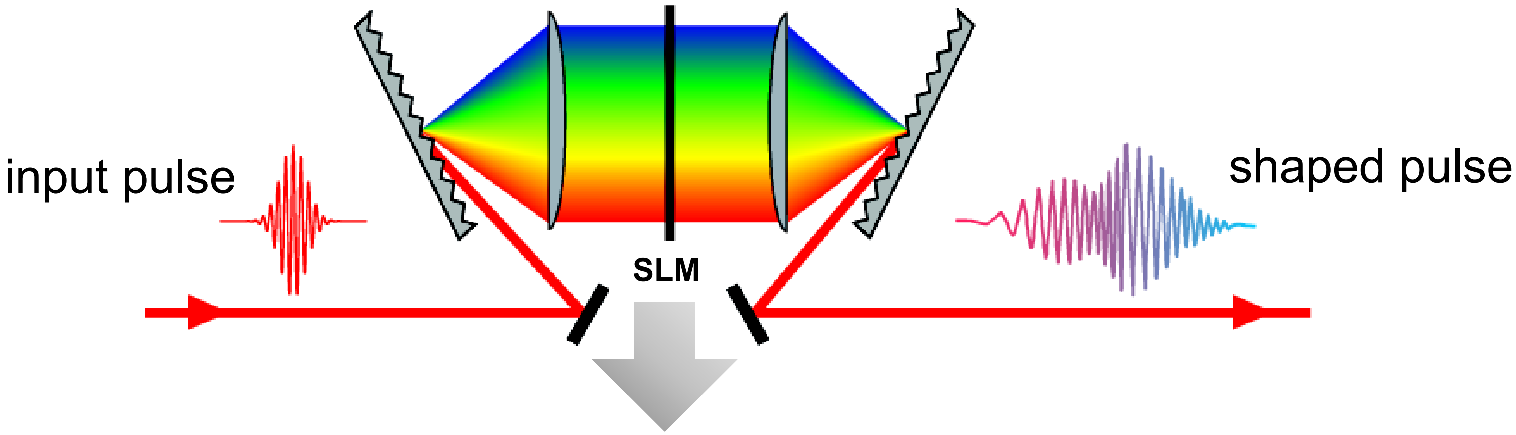
fs-pulses present a broad spectral band





We take advantage of the broad spectral band to shape the pulse in the frequency domain

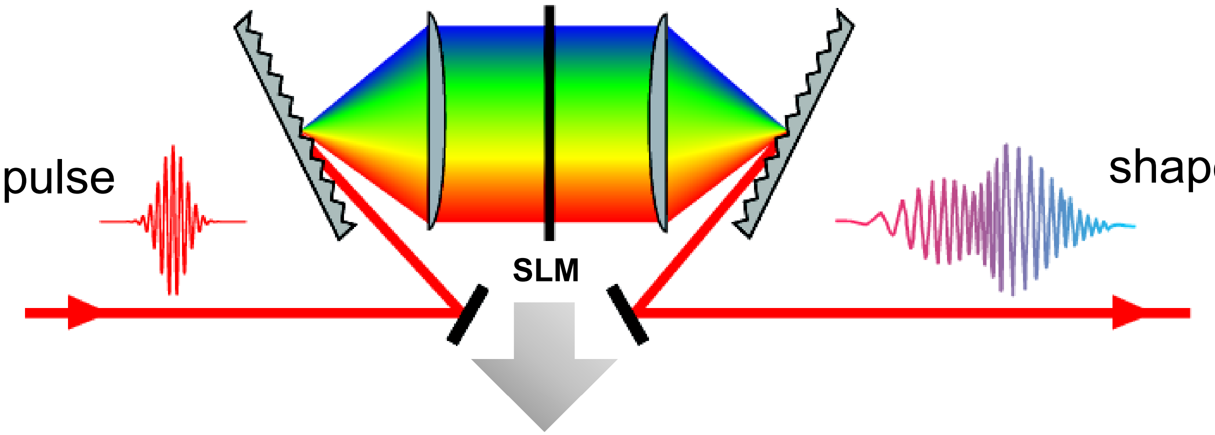
Pulse-shaping



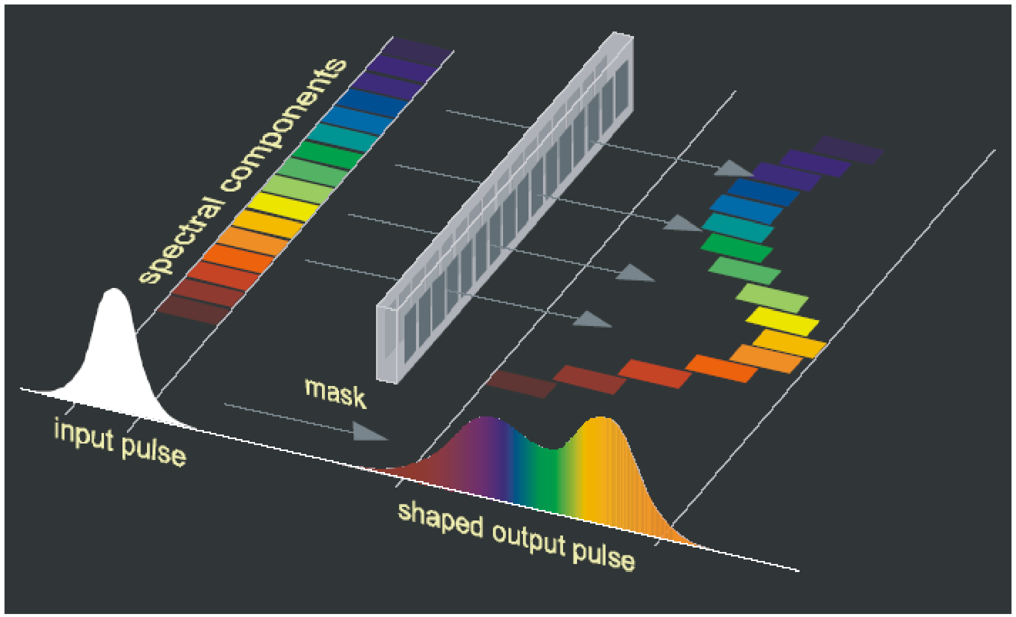
To generate pulses that are able to control optically-induced processes

To compensate for distortions in the pulses

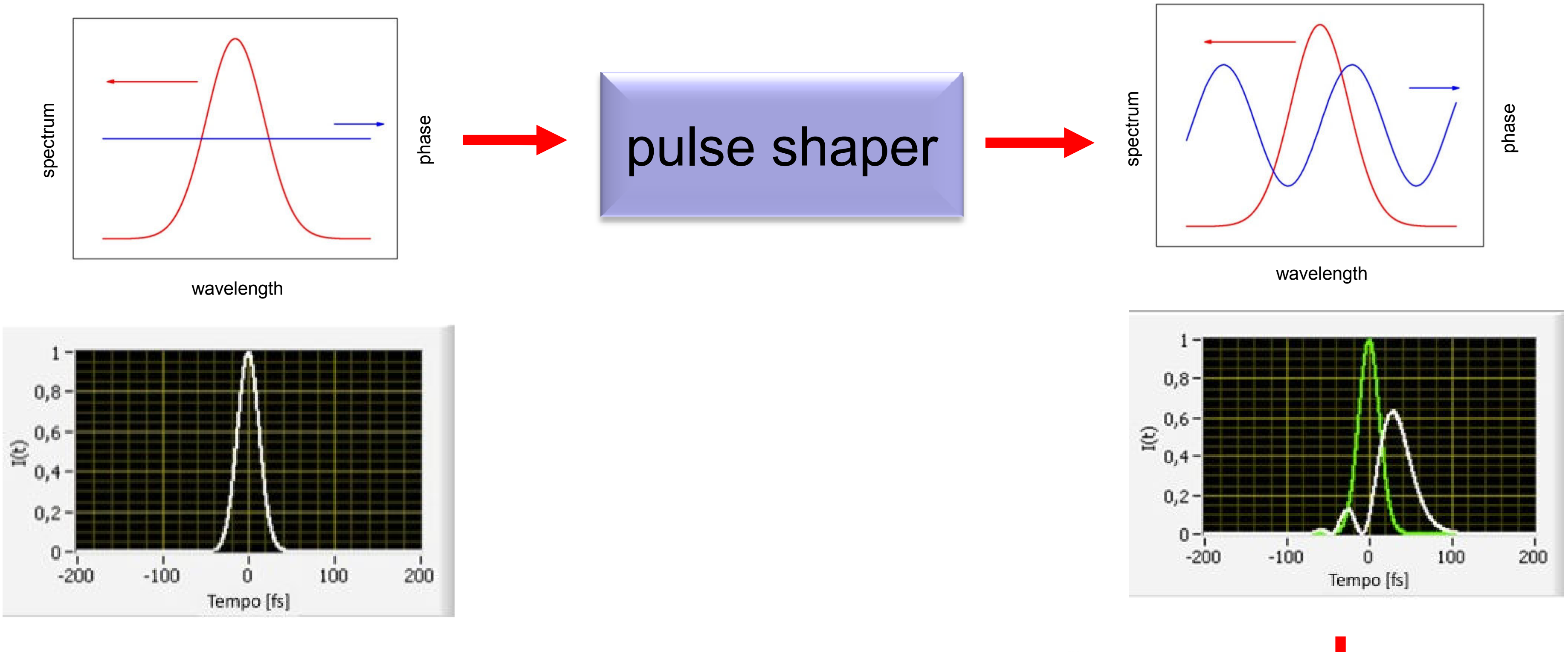
Pulse-shaping for coherent control



liquid crystal display



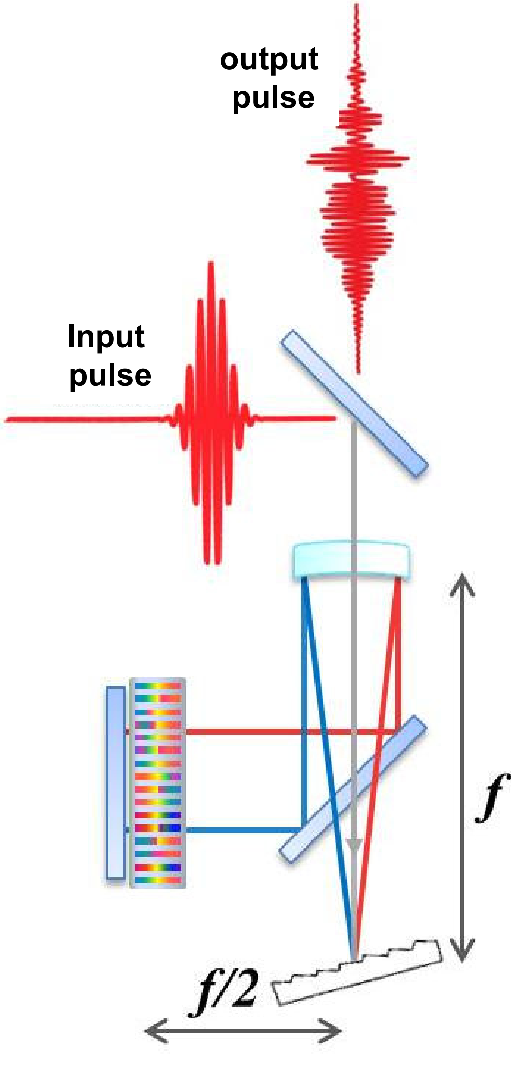
Pulse-shaping for coherent control

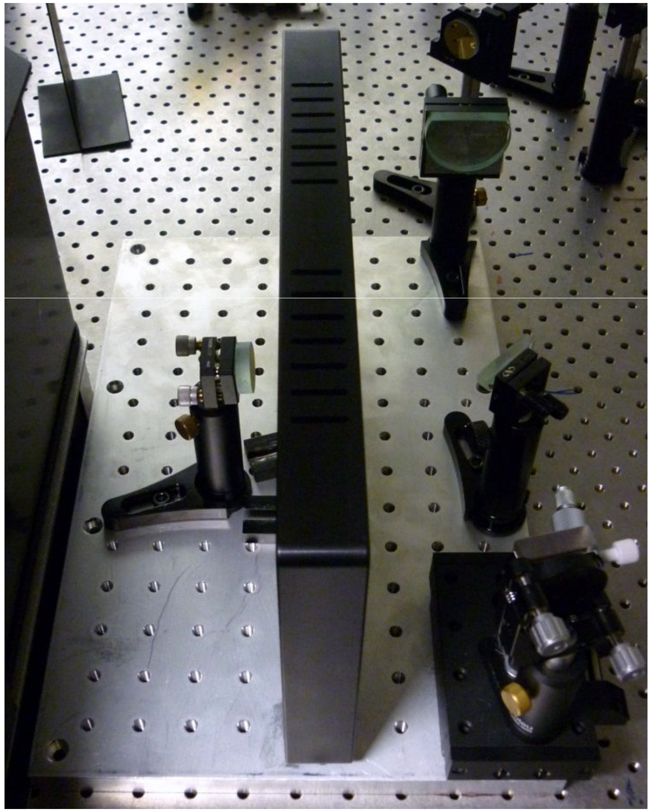


By changing the pulse shape we can alter the results of an experiment

Pulse-shaper

Reflection system





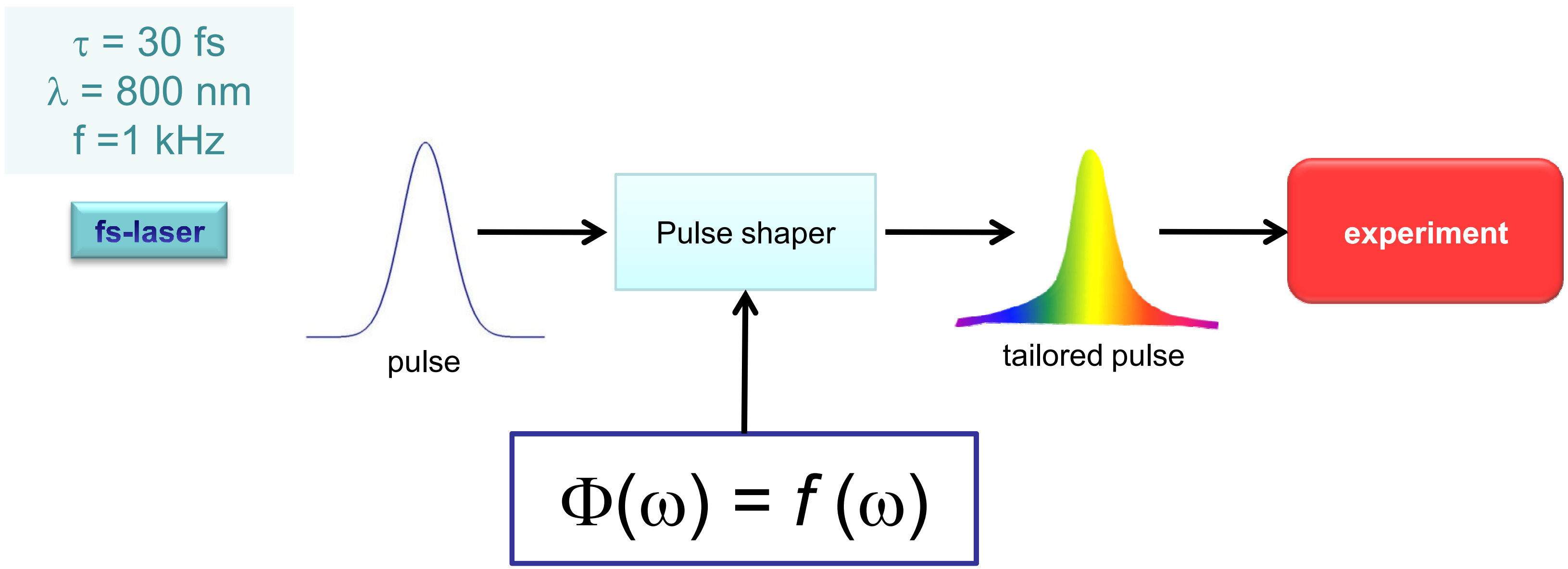
Shaping the pulse

How to define which pulse shape to use ?

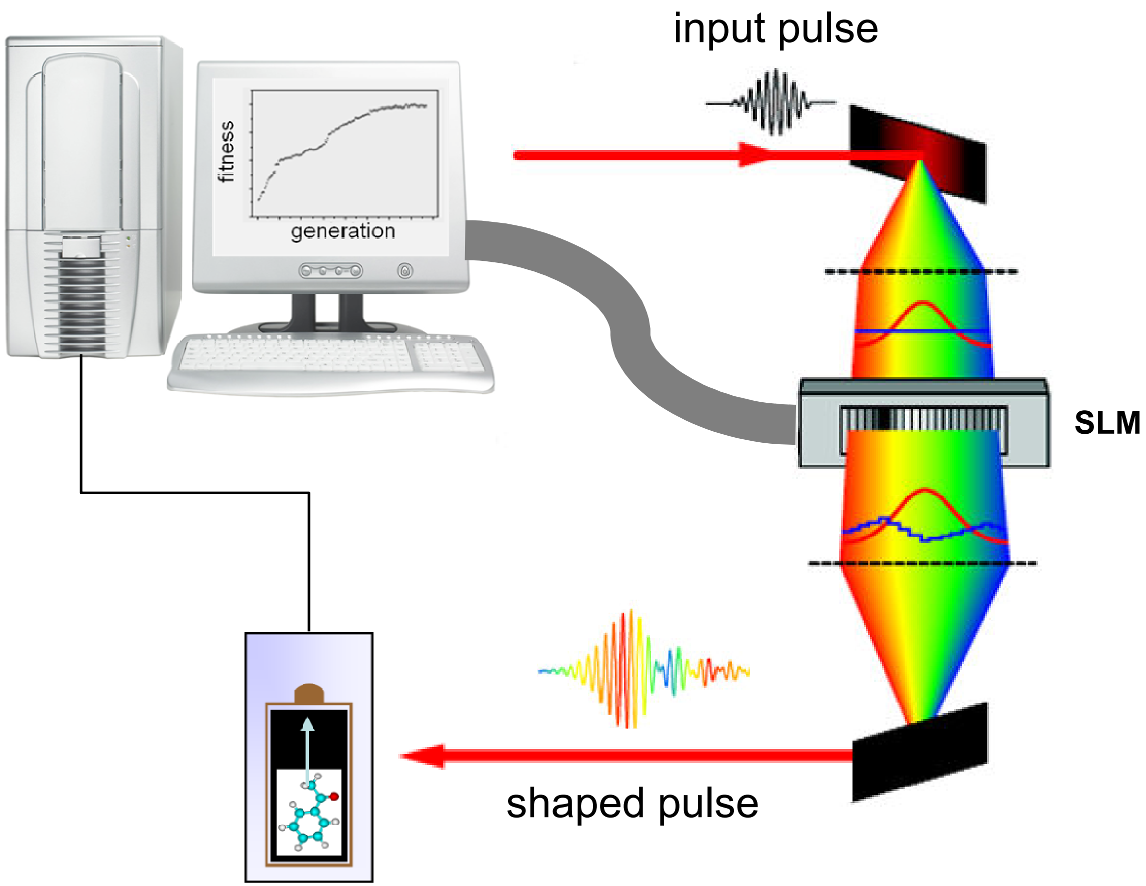
Learning algorithms

Defined phase masks

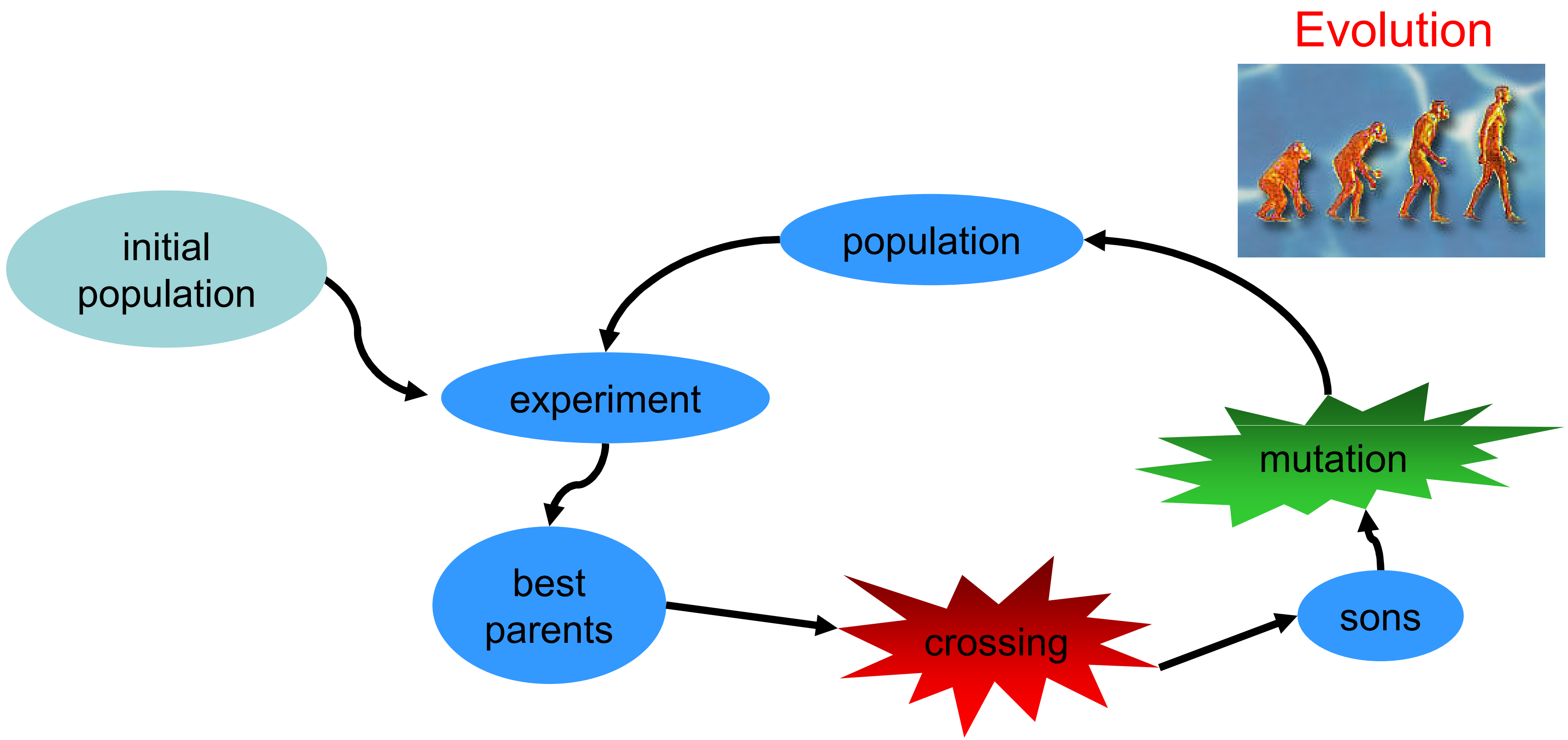
Coherent control: defined phase masks



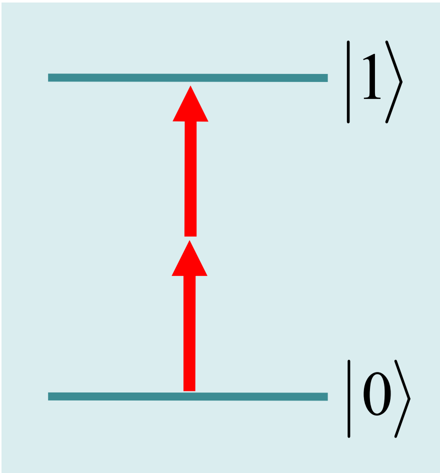
Coherent control: learning algorithm



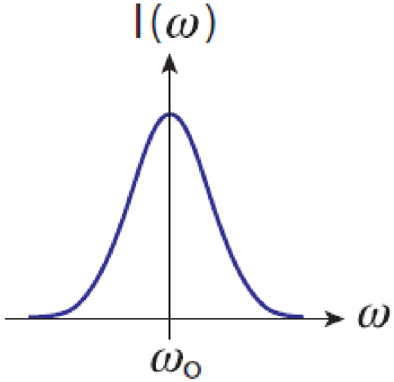
Genetic Algorithm



The physics of coherent control



multi-photon absorption induced by ultrashort pulses

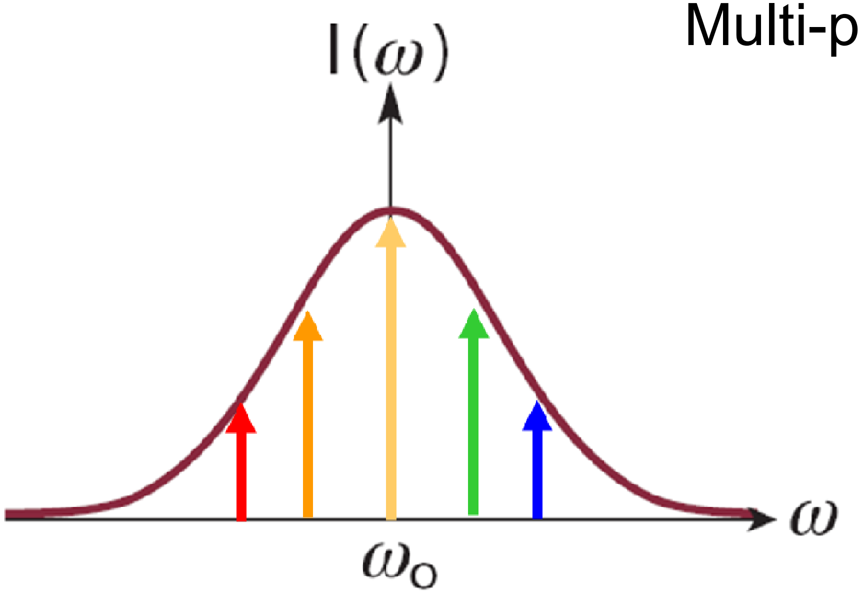


broad spectral band

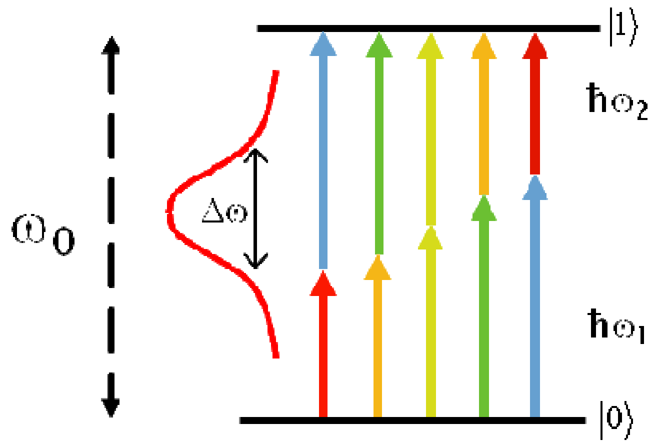
distinct photons of the pulse can promote two-photon absorption (**nondegenerate**)

multi-photon intrapulse interference

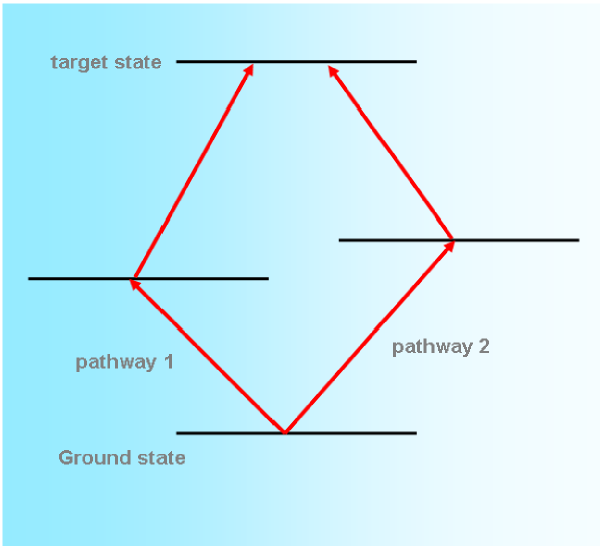
The physics of coherent control



Multi-photon intrapulse interference



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

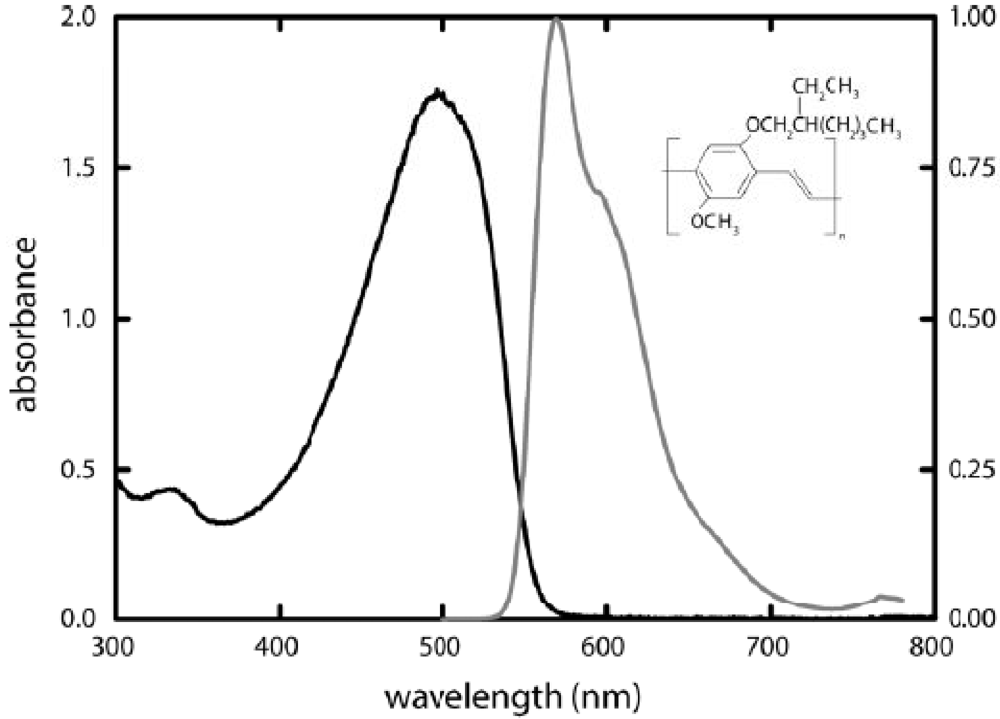


It is needed to “shape” the phase of the pulse

Results

1) control of MEH-PPV photodegradation
2) control of emission in Y-shaped molecules
3) control of Au nanoparticles formation in chitosan

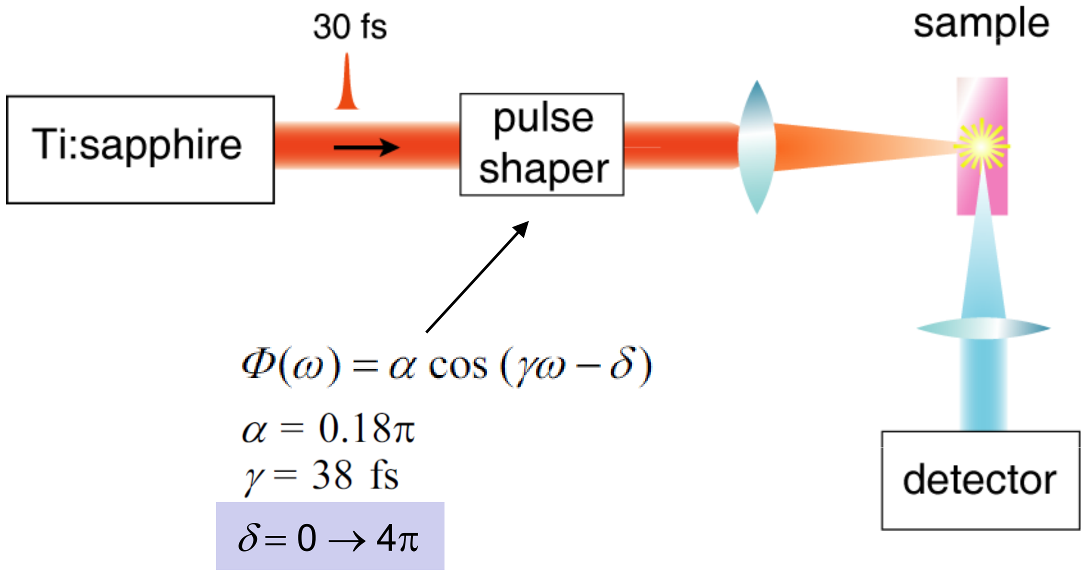
Control of MEH-PPV photodegradation



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

Control of MEH-PPV photodegradation

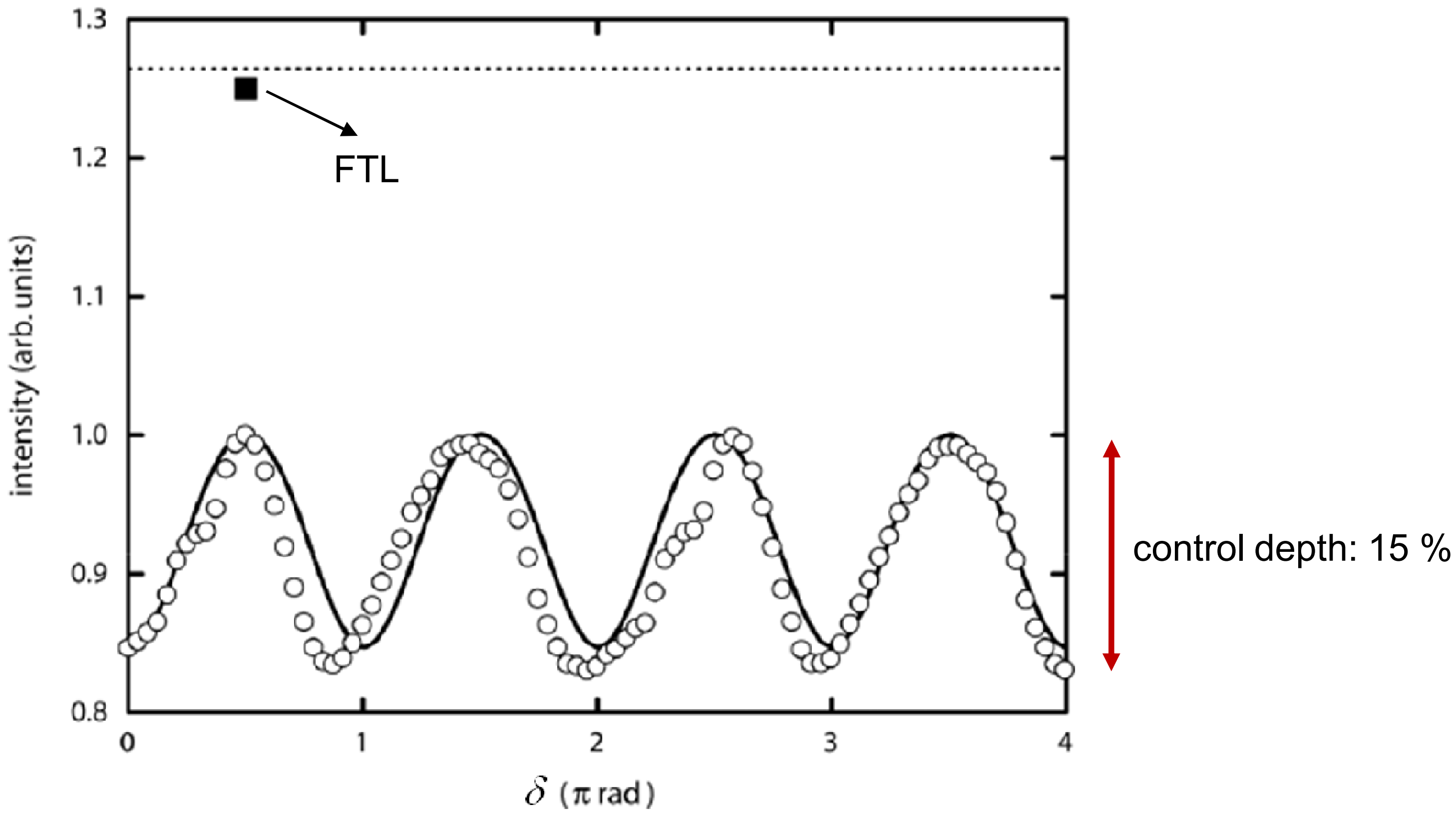


Measured

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

2 - photodegradation for distinct phase masks

Control of MEH-PPV photodegradation



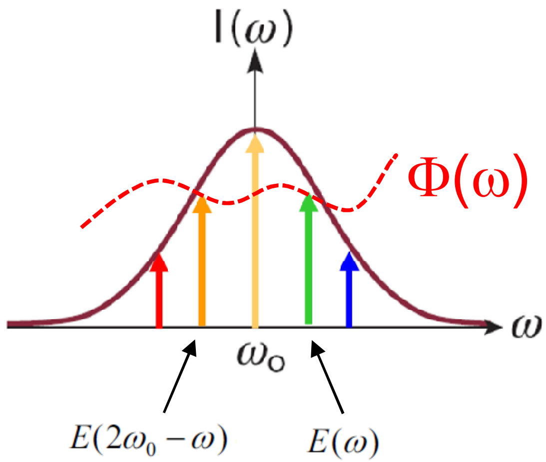
The emission with FTL pulses is ~ 25% higher than the one with the phase mask

Control of MEH-PPV photodegradation

Two-photon absorption transition probability for an atomic system

$$S^{(2)} \propto \left| \int_0^\infty E(\omega)\, E(2\omega_0 - \omega)\, \mathrm{d}\omega \right|^2 \mathrm{d}\omega_0$$

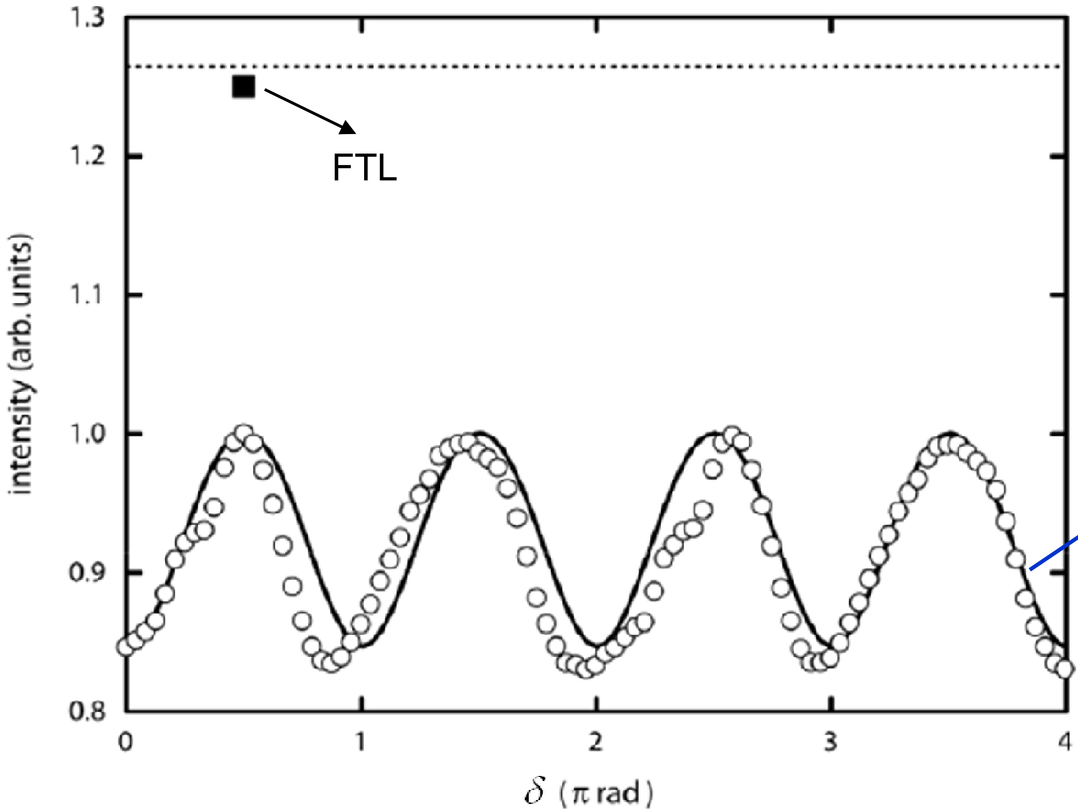
$$E(\omega) = A(\omega) \exp\left[\mathrm{i}\Phi(\omega)\right]$$



For a molecular system it is needed to include the integral on the 2PA spectrum $g(2\omega_0)$

$$S^{(2)} \propto \int g(2\omega_0) \left| \int_0^\infty E(\omega)\, E(2\omega_0 - \omega)\, \mathrm{d}\omega \right|^2 \mathrm{d}\omega_0$$

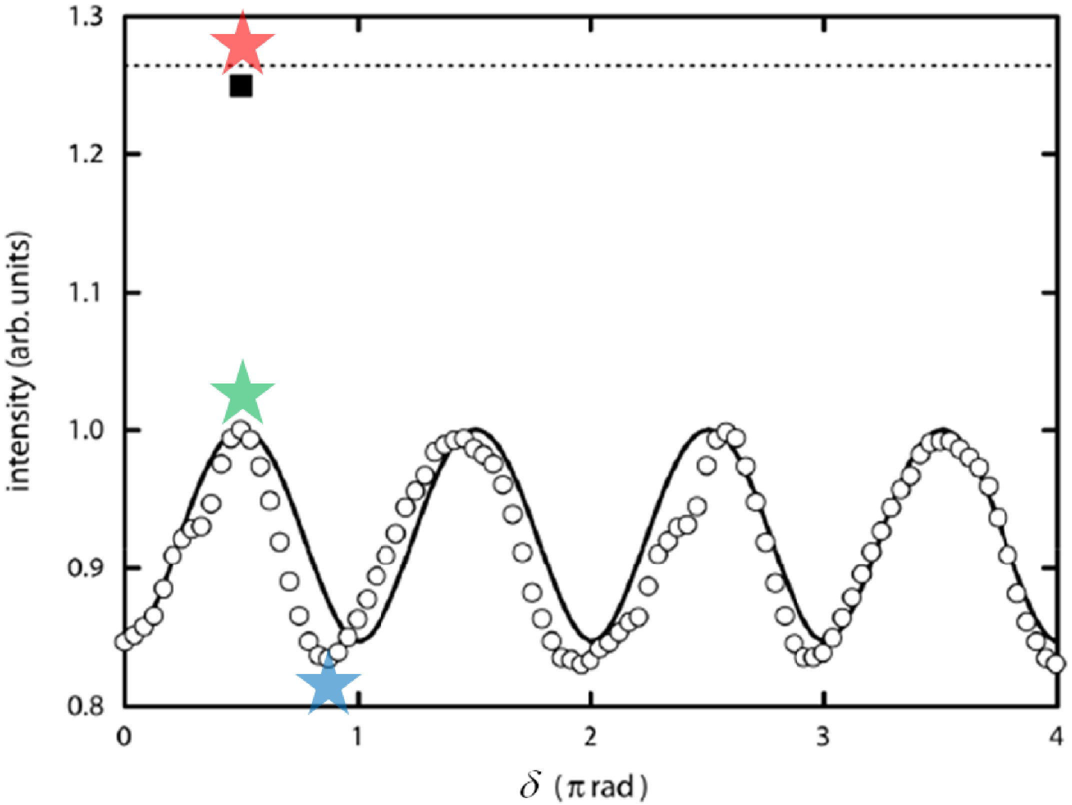
Control of MEH-PPV photodegradation



$$S^{(2)} \propto \int g(2\omega_0) \left| \int_0^\infty E(\omega)\, E(2\omega_0 - \omega)\, \mathrm{d}\omega \right|^2 \mathrm{d}\omega_0$$

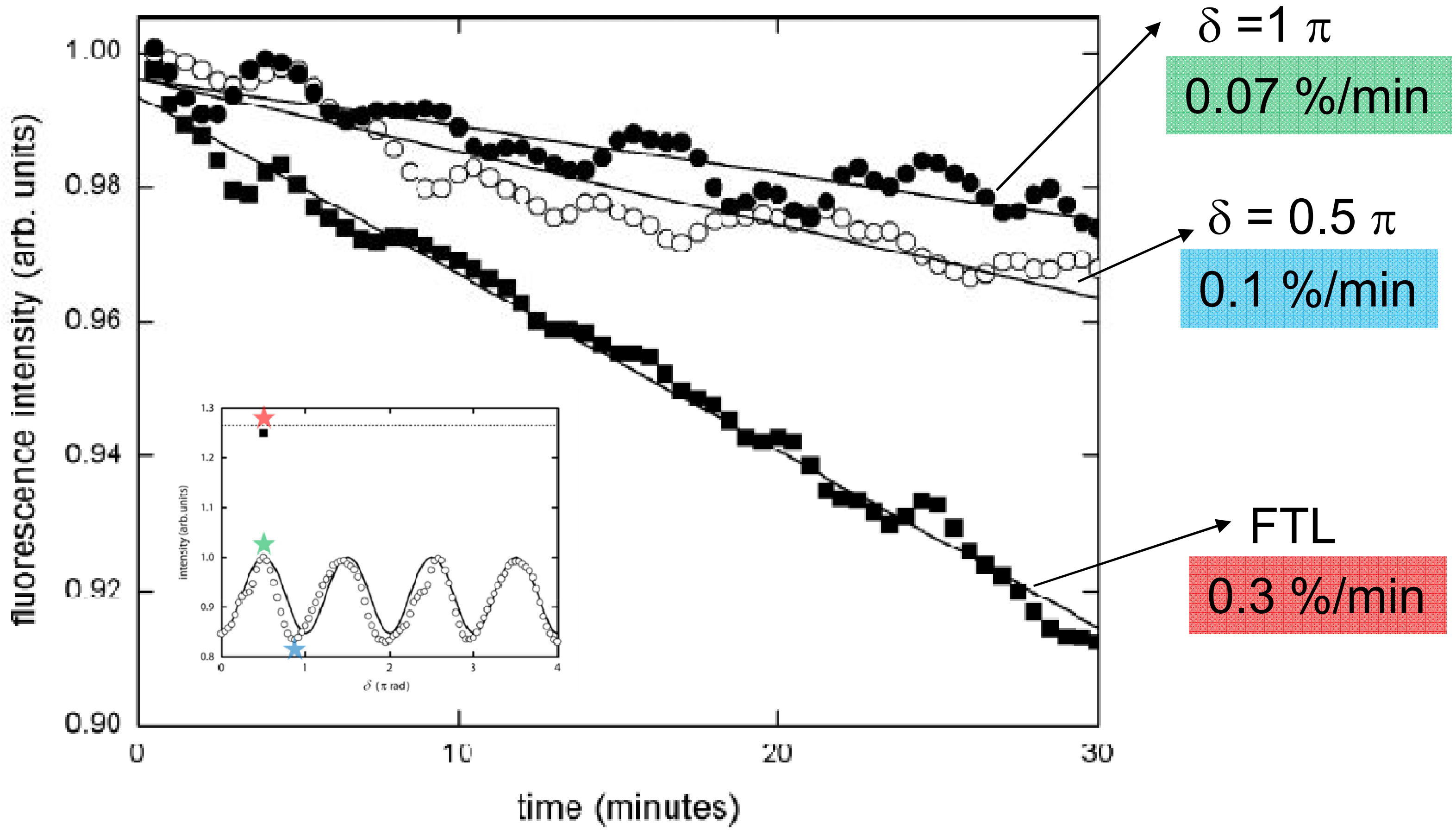
modeling of the coherent control

Control of MEH-PPV photodegradation



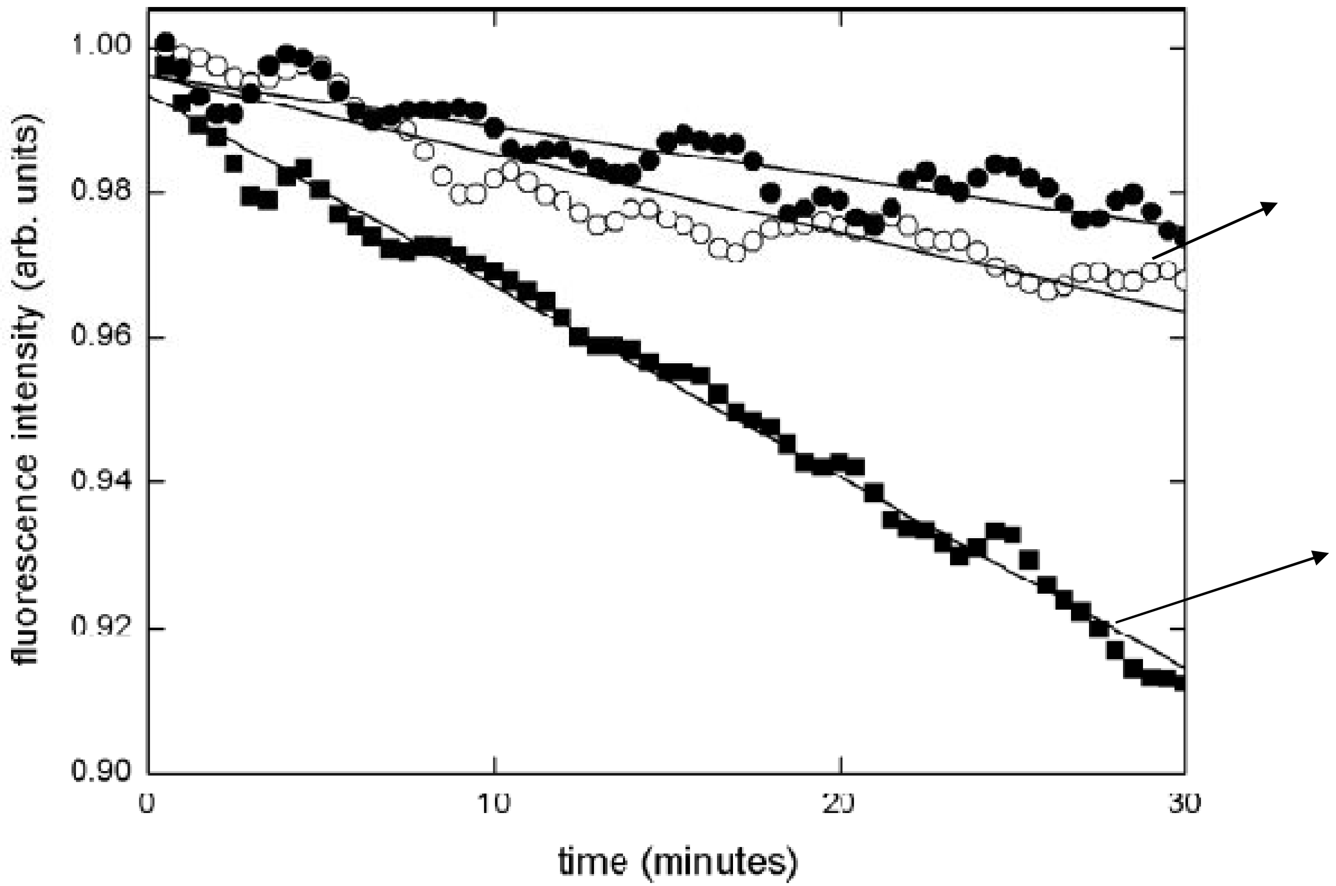
measuring the photobleaching rate with three diferent phase-masks

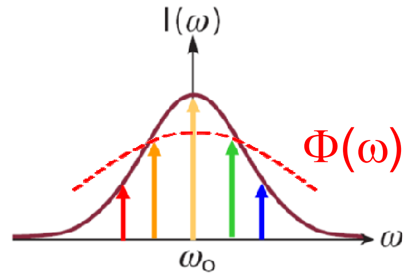
Control of MEH-PPV photodegradation



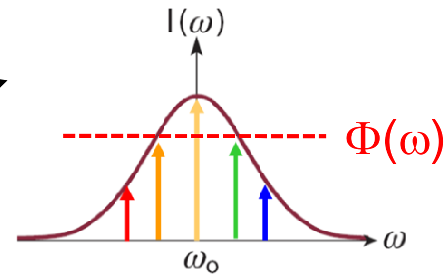
photobleaching rate <u>decreases 3 times</u> for the phase-masked pulses

Control of MEH-PPV photodegradation





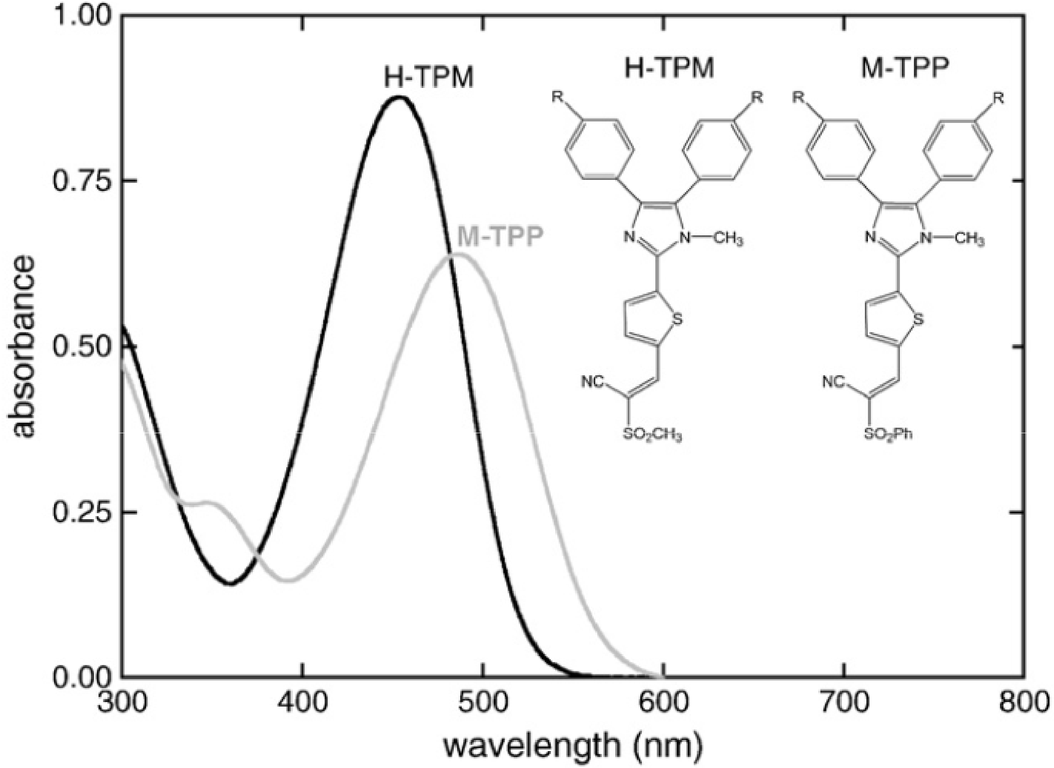
pairs in phase



all in phase

cosine-like mask: smaller amount of molecules is excited $\rightarrow$ less photobleaching

Control of emission in Y-sahped molecules

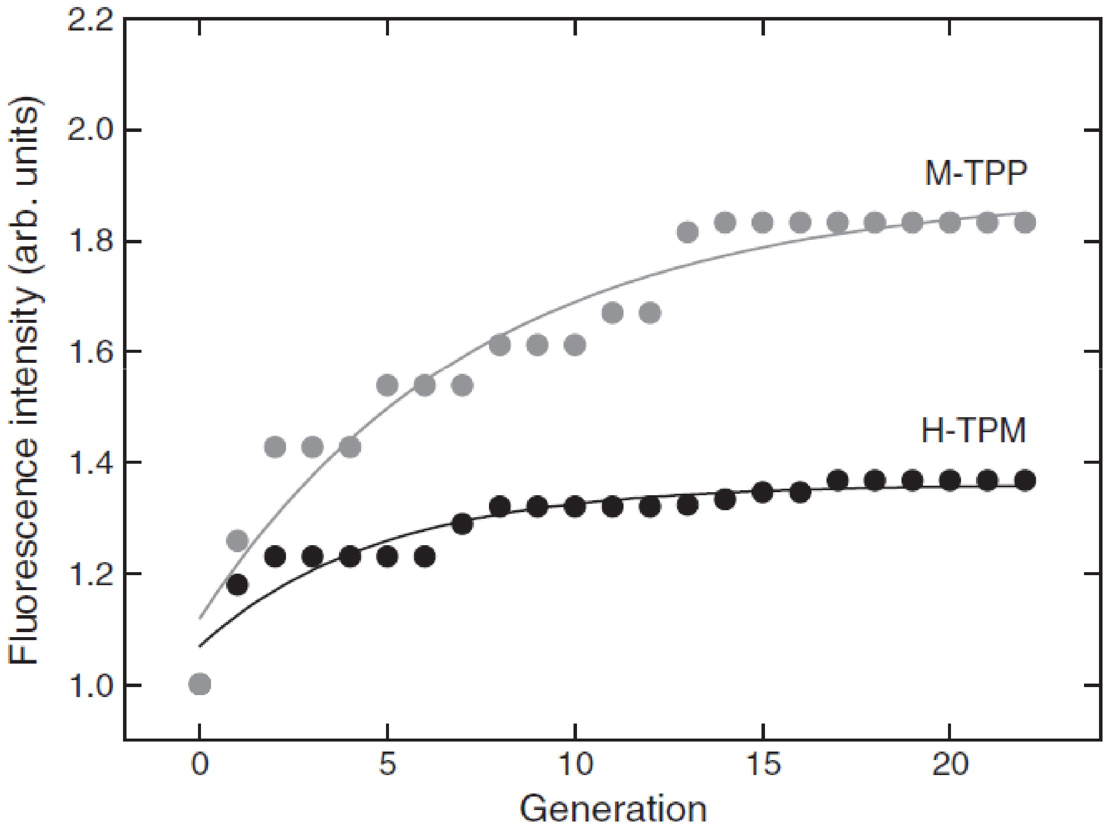


Optimize the two-photon excited emission of Y-shaped molecules using GA

Understanding the coherent control on the 2PA in molecular systems can lead to the development of new strategies to enhance ONL

Control of emission in Y-sahped molecules

Controlling the emission by Genetic Algorithm



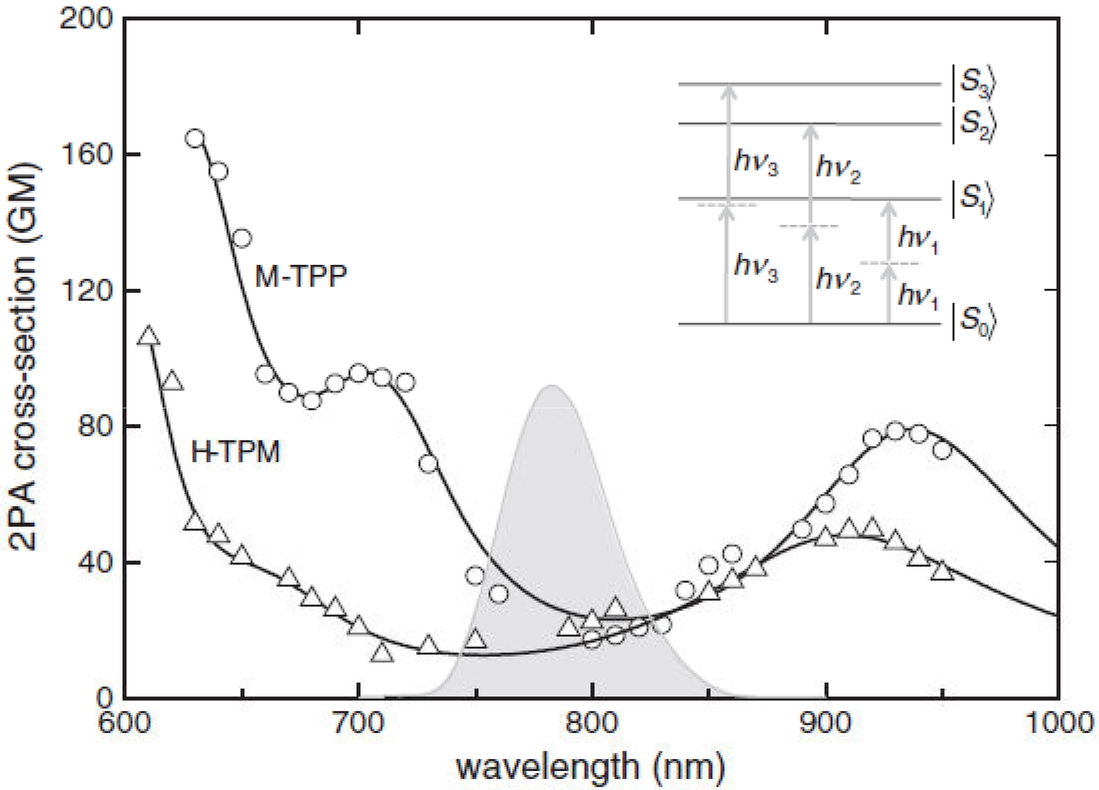
$\bar{R} = 1.9 \pm 0.2$

$\bar{R} = 1.4 \pm 0.1$

the final pulse is FTL for both molecules

specific molecular features affect the control over the 2PA process

Control of emission in Y-sahped molecules



Intra pulse multi-photon interference

$$S^{(2)} \propto \int_{-\infty}^{\infty} g^{(2)}(2\omega_0) \left| \int_{-\infty}^{\infty} E(2\omega_0 - \Omega) E(\Omega) d\Omega \right|^2 d\omega_0$$

distinct 2PA spectra

The detuning between the 2PA and the pulse spectrum is smaller for M-TPP

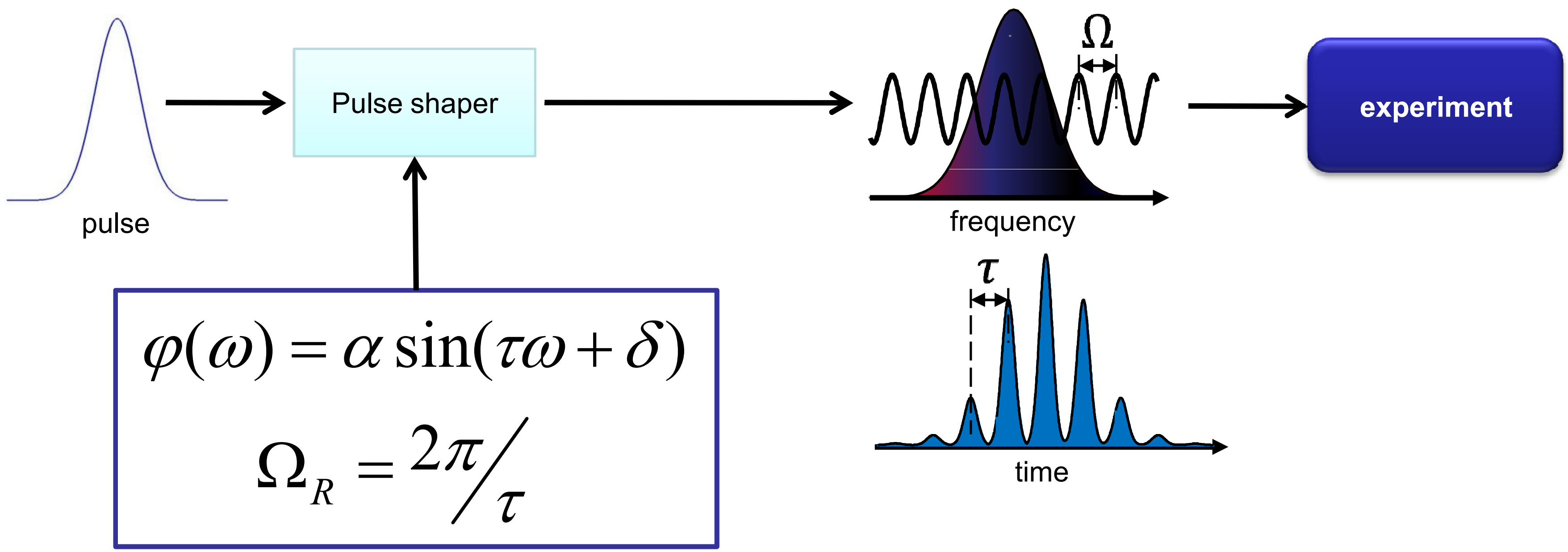
modeling the growth rate

M-TPP $\bar{R}_{teo} = 1.8$

H-TPM $\bar{R}_{teo} = 1.5$

Pulse shaping

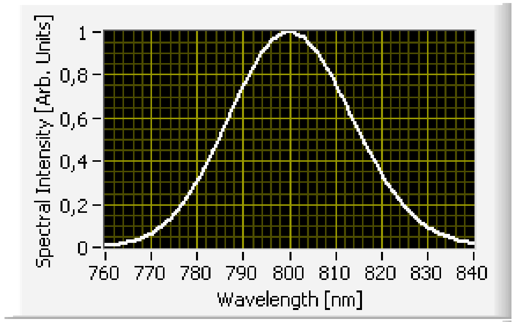
creating a pulse train



$$\varphi(\omega) = \alpha \sin(\tau\omega + \delta)$$

$$\Omega_R = 2\pi / \tau$$

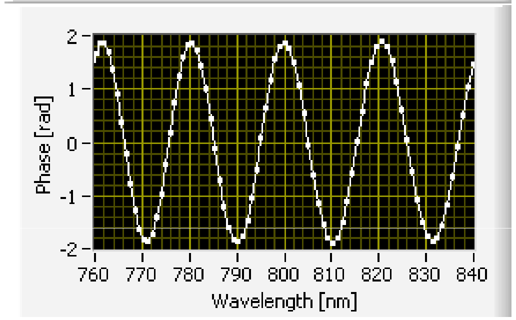
Control of Au nanoparticles formation

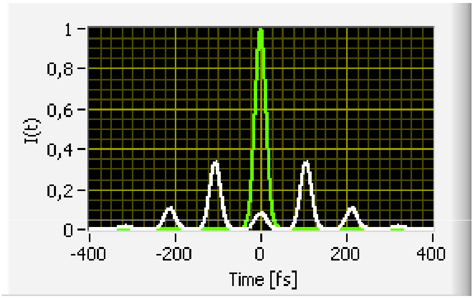


varying the period of the sinusoidal phase mask

$$\emptyset(\omega) = \alpha \, sin(\gamma\omega + \delta)$$

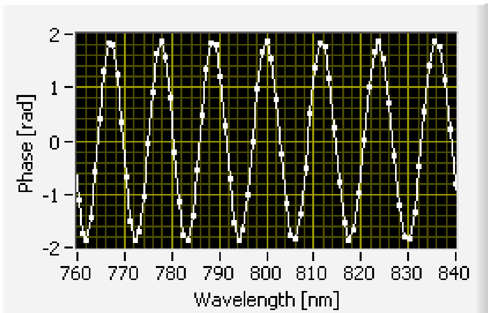
generate pulse trains with distinct separation time

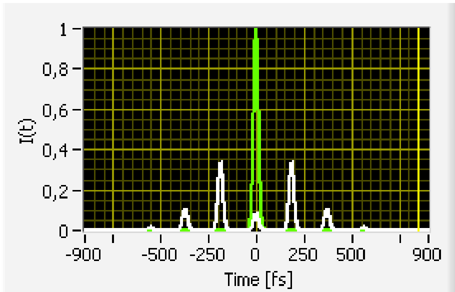




$N_{periods} = 4$

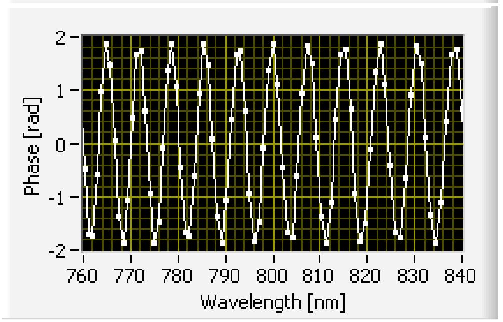
$t_{sep} = 106$ fs

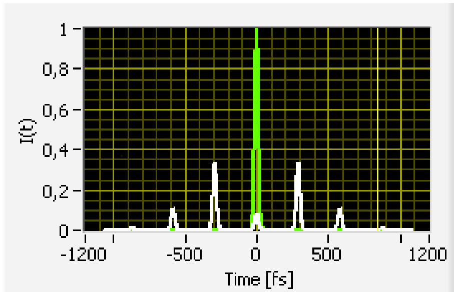




$N_{periods} = 7$

$t_{sep} = 186$ fs

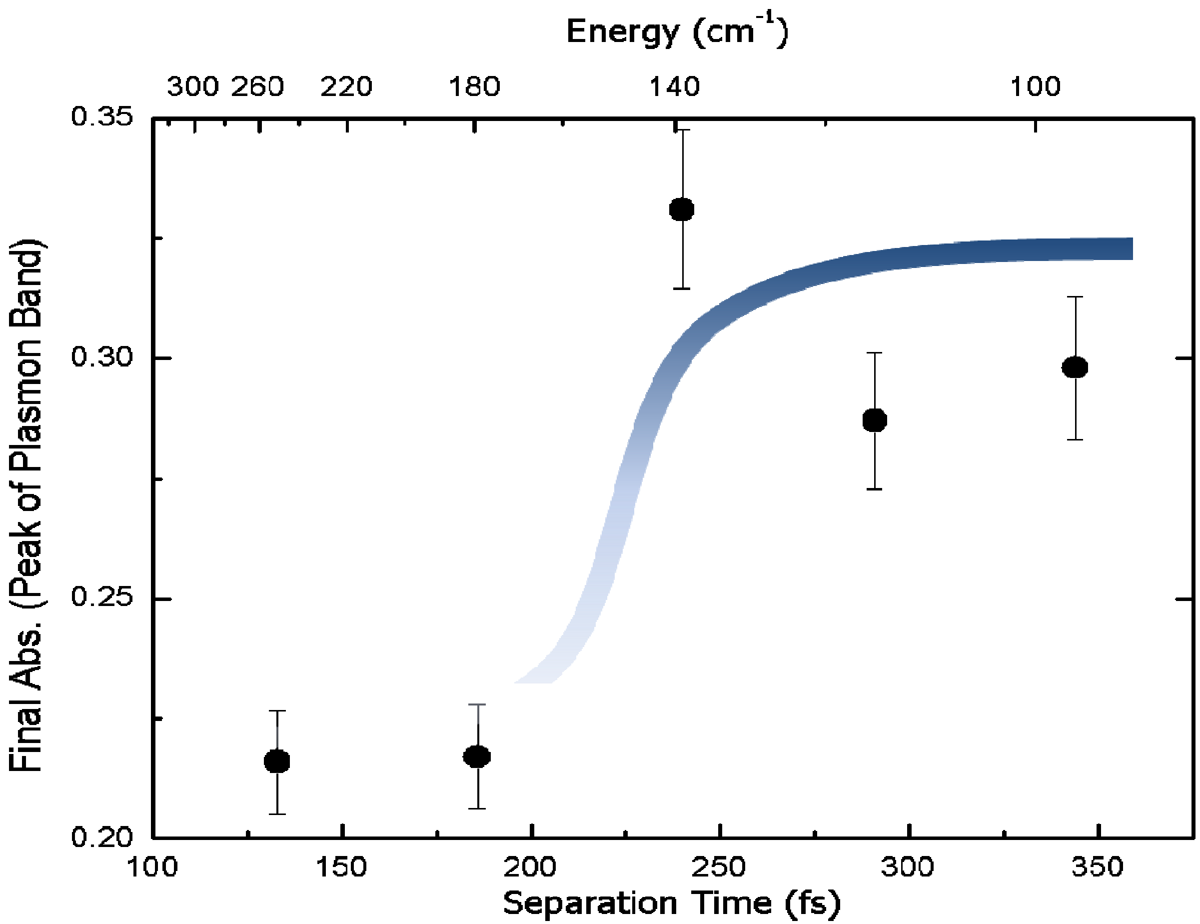




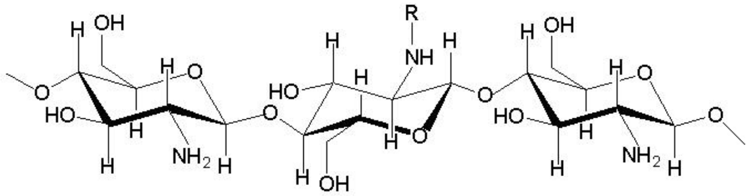
$N_{periods} = 11$

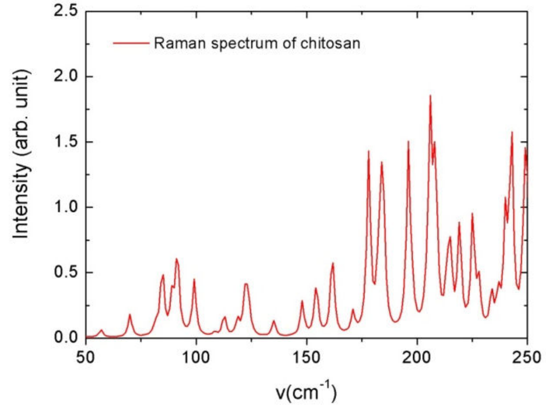
$t_{sep} = 291$ fs

Control of Au nanoparticles formation

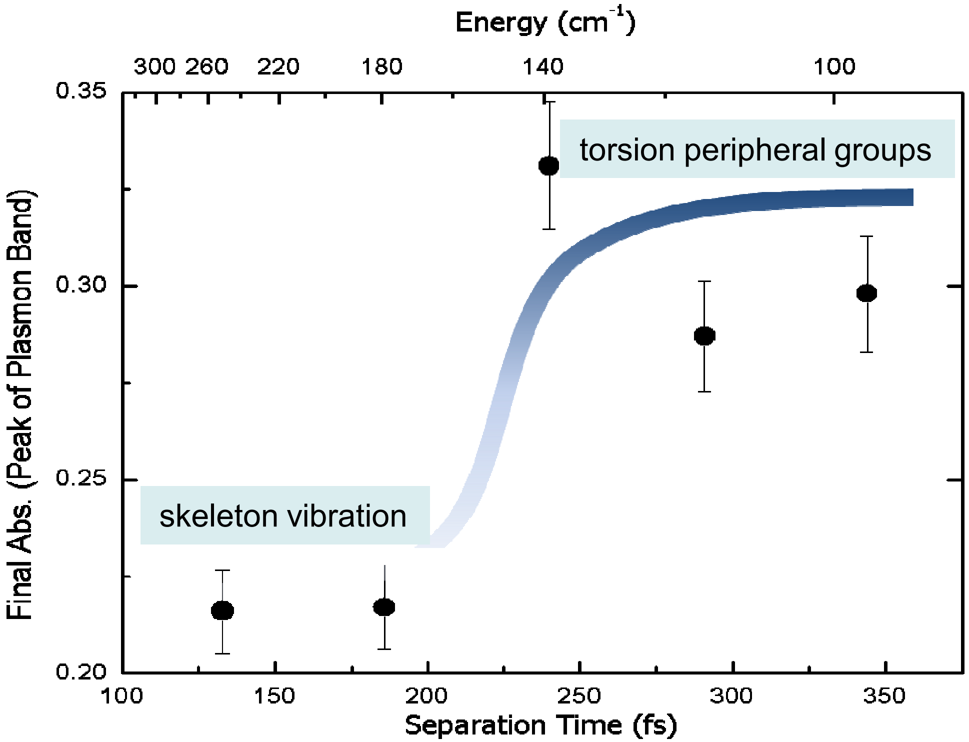


Control of Au nanoparticles formation





lower frequencies are related to peripheral groups (OH and NH_2), which are propably related to the gold photoreduction



Acknowledgments

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Thank you !

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