Ultrashort laser pulse trains to enhance the induced Au nanoparticles formation

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Outline

Motivation for coherent control

Mechanism of coherent control

Pulse shaping

Results: control of Au nanoparticles formation

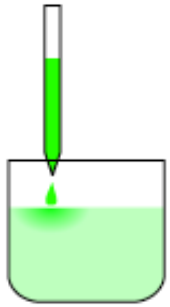
Conclusions

Conventional methods of chemical control

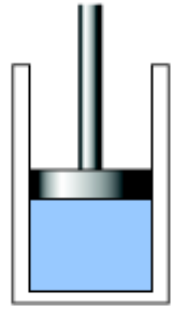
Macroscopically

temperature

concentration



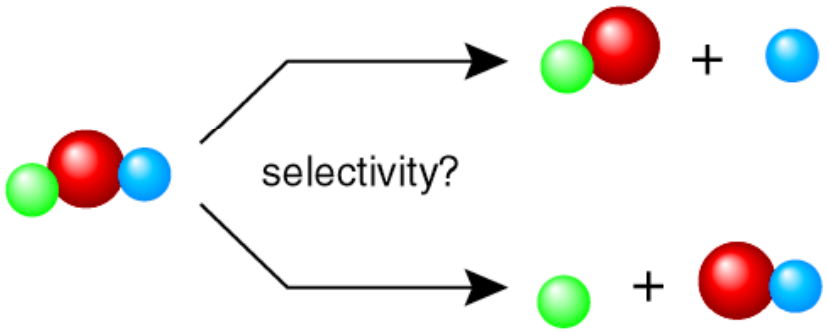
pressure



Microscopically

catalysis

Coherent Control

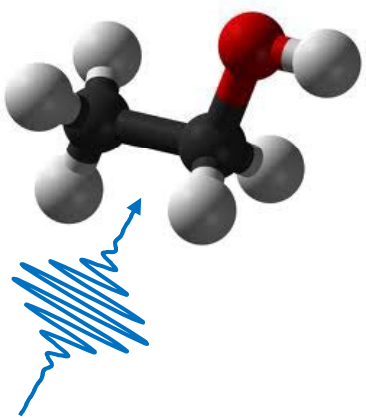


How to change a chemical reaction pathway and make different products?

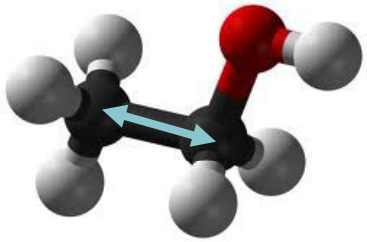
Use a light with a given frequency to excite the chemical bond we would like to break.

Intramolecular Vibrational Redistribution

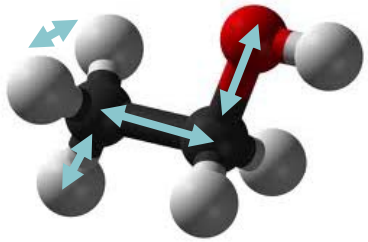
excite one bond



the bond vibrates



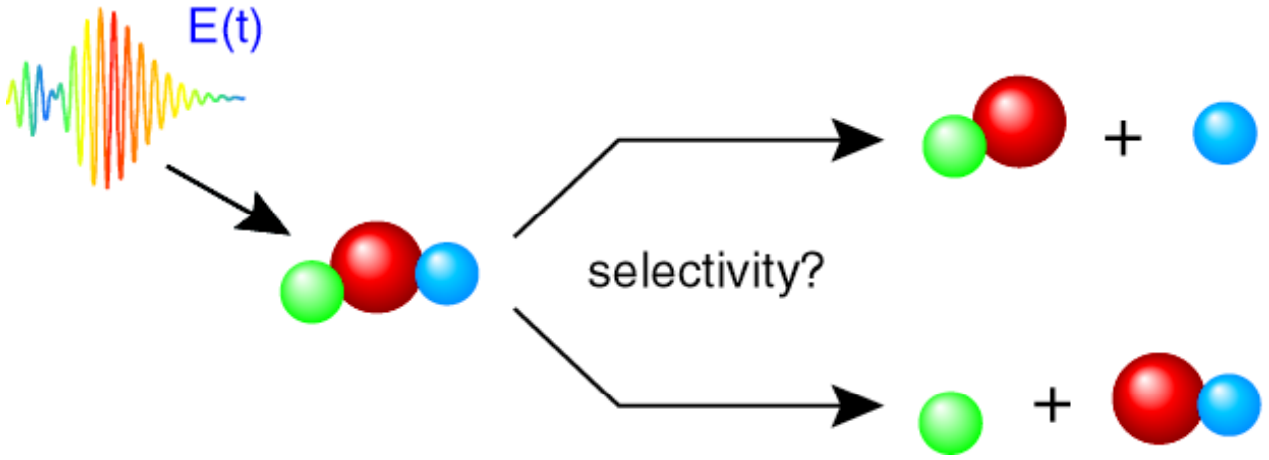
After a few fs the entire molecule is vibrating



IVR occurs on a few-fs, therefore long pulses excite the entire molecule, and the weakest bond breaks, no matter which bond was excited.

Coherent control: using shaped fs pulses

Can we used fs-pulses to cause a molecular vibration in such a way that a chosen bond is broken ?

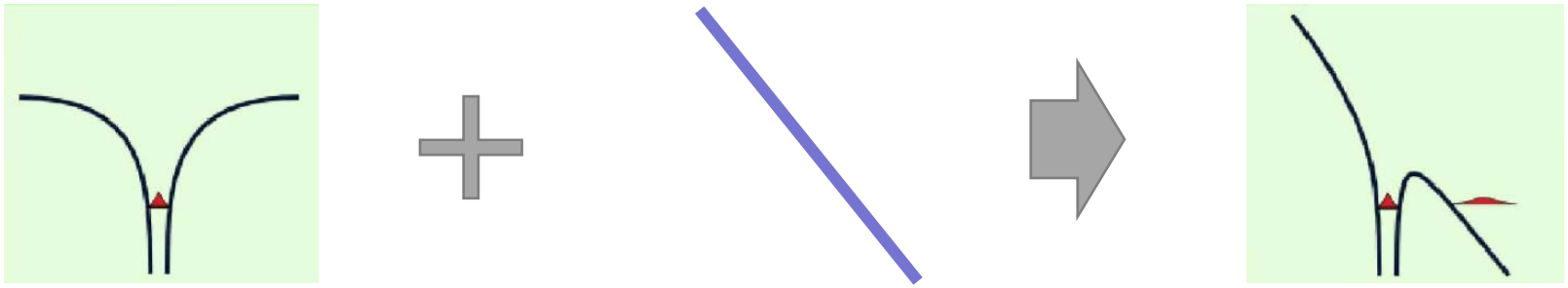


E(t) ?

The physics of coherent control

Nonperturbative nonlinear optics: strong field regime

The pulse electric field perturbs the molecule and can dissociates it



Molecule

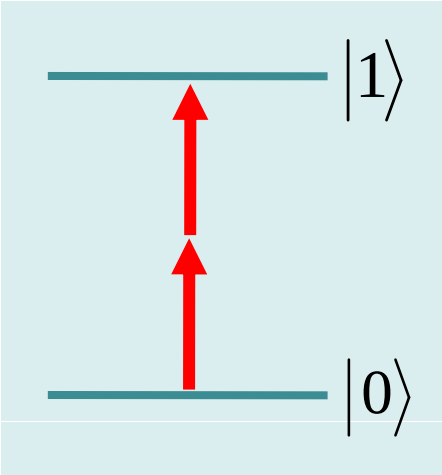
E-field of laser

Perturbed System

The required electric field of the pulse needs to be properly chosen

The physics of coherent control

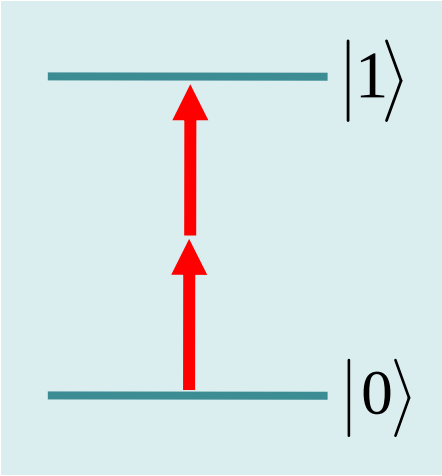
Perturbative nonlinear optics



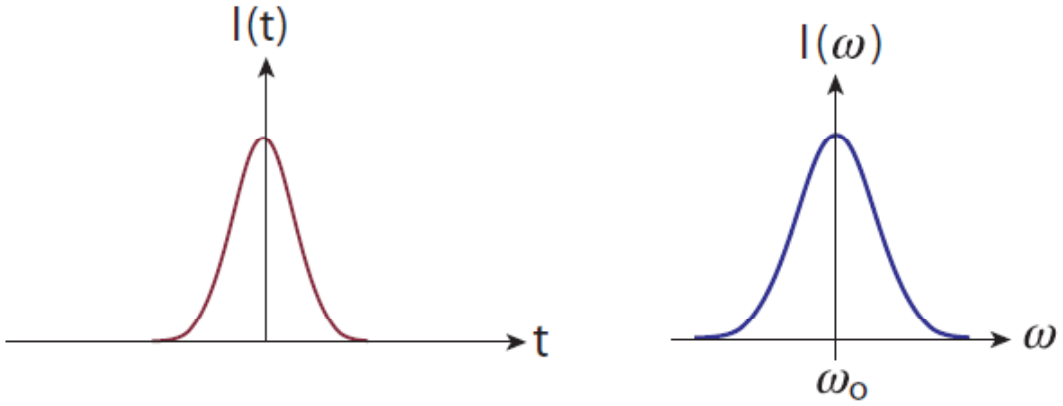
Using multi-photon absorption to excite a molecular system

induce photoreaction

The physics of coherent control



multi-photon absorption induced by ultrashort pulses

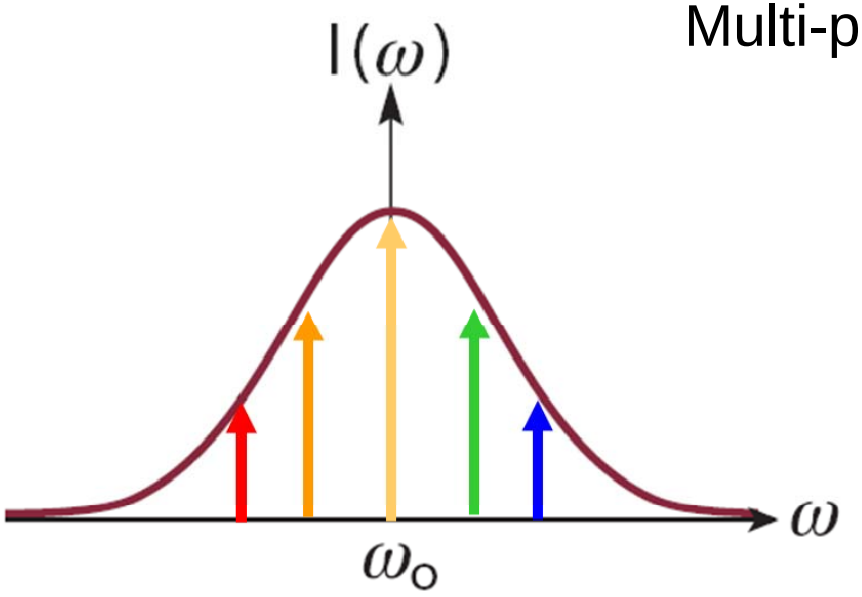


broad spectral band

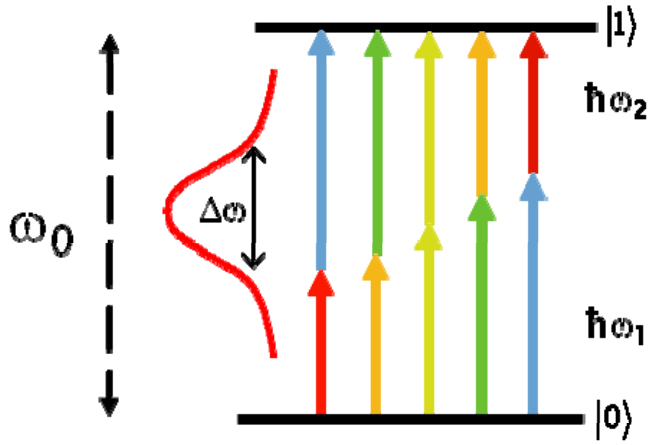
distinct photons of the pulse can promote two-photon absorption (**nondegenerate**)

multi-photon intrapulse interference

The physics of coherent control

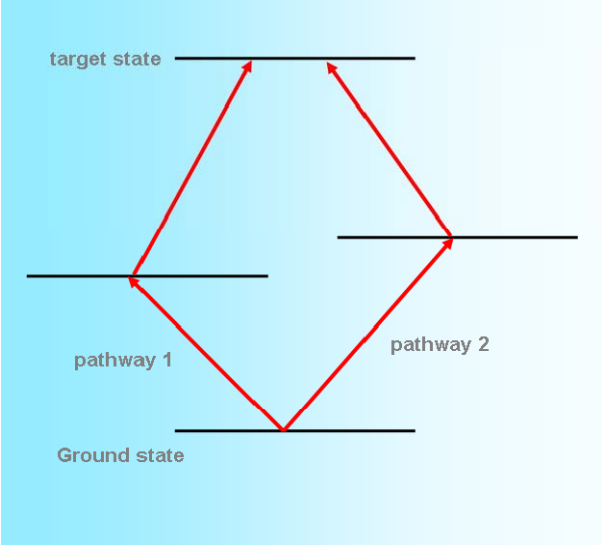


Multi-photon intrapulse interference



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



Theory for coherent control

The complete Hamiltonian for the system needs to be known

$$H_{system} = H_{molecule} + H_{radiation} + H_{interaction}$$

$H_{radiation}$ → **known** 

$H_{molecule}$ → small molecules: **approximate**

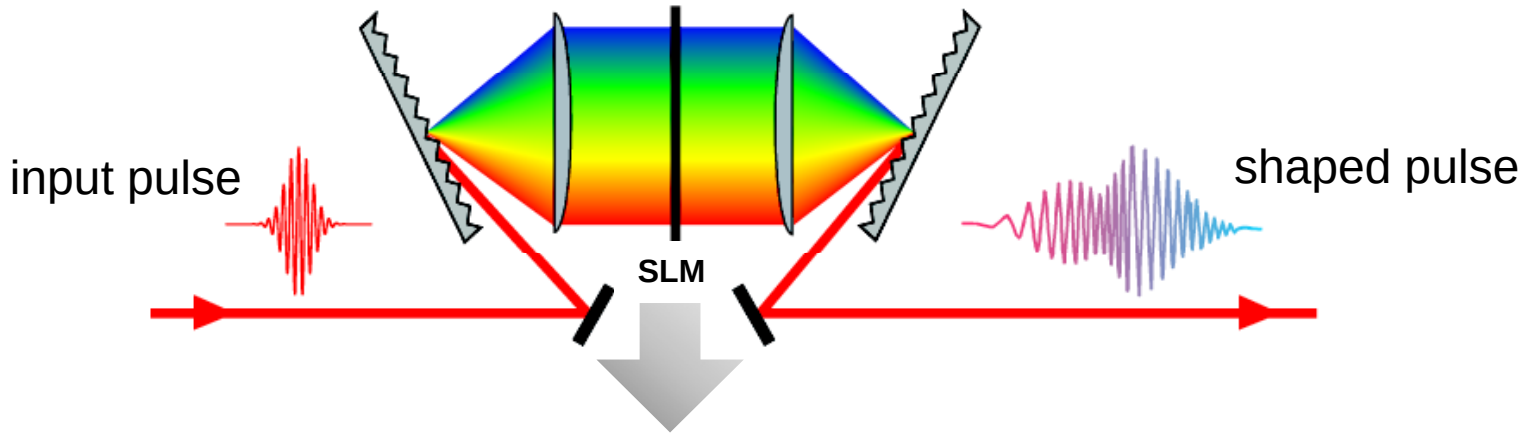
large molecules: **unknown**

$H_{interaction}$ → weak field: **known** $\langle \psi_f | \mu \cdot E | \psi_i \rangle$

strong field: **unknown**

It might be possible to solve the problems for **VERY** simple system/molecules

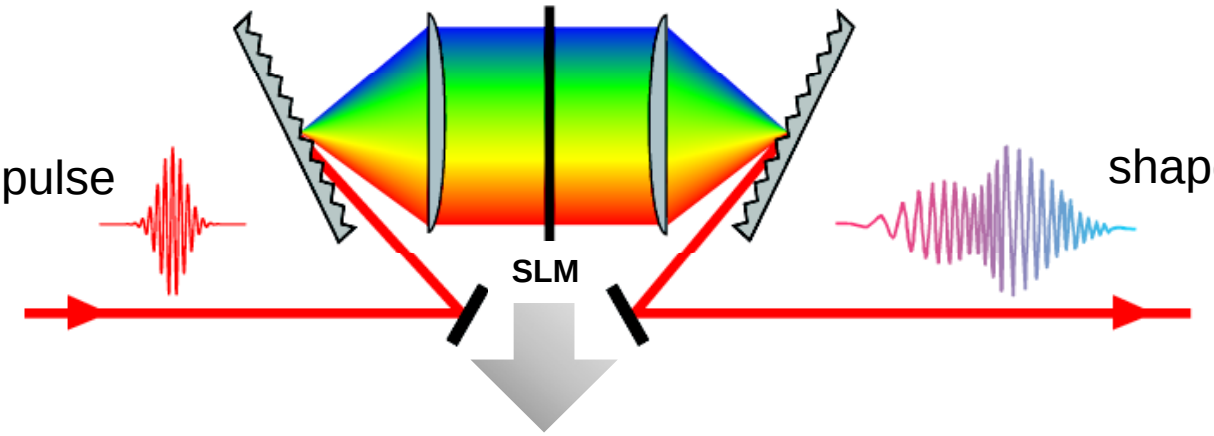
Pulse-shaping for coherent control



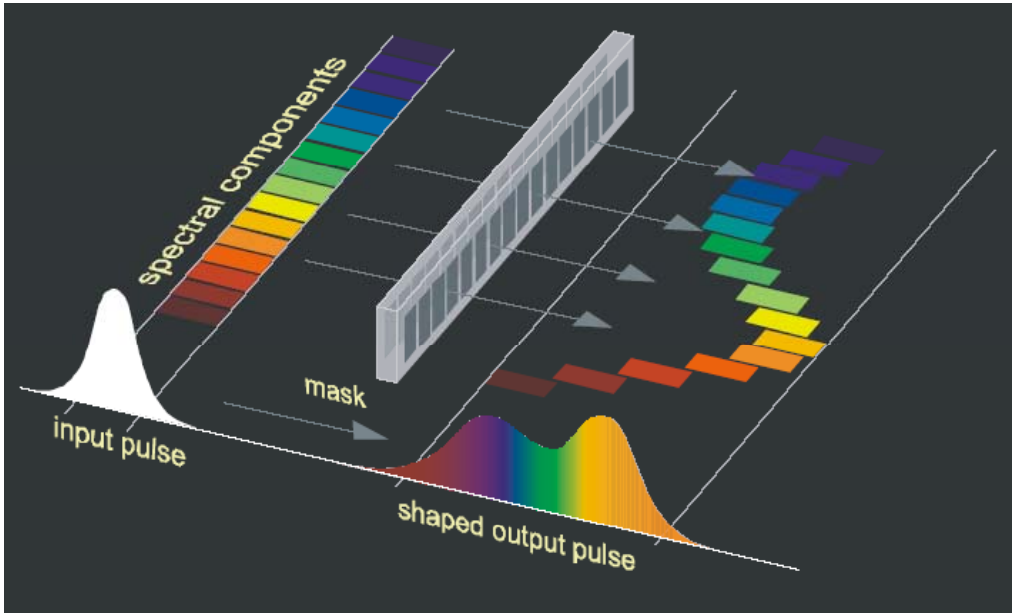
To generate pulses that are able to control optically-induced processes

To compensate for distortions in the pulses

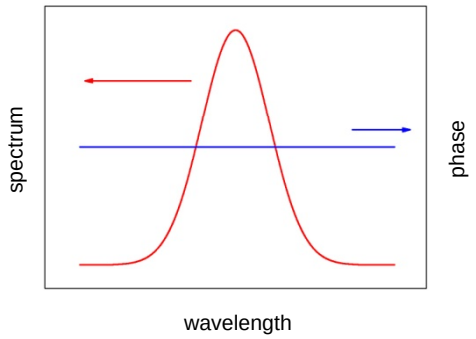
Pulse-shaping for coherent control



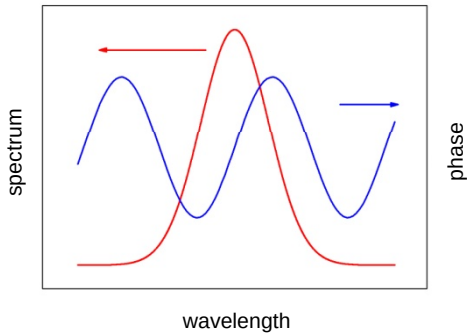
liquid crystal display

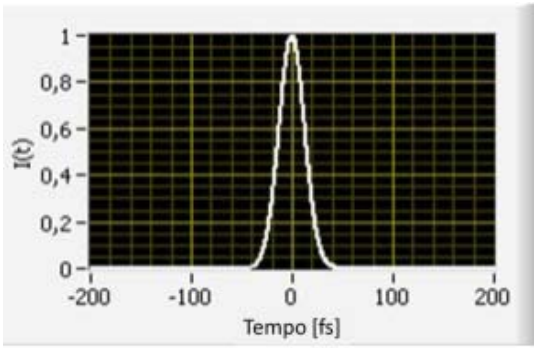


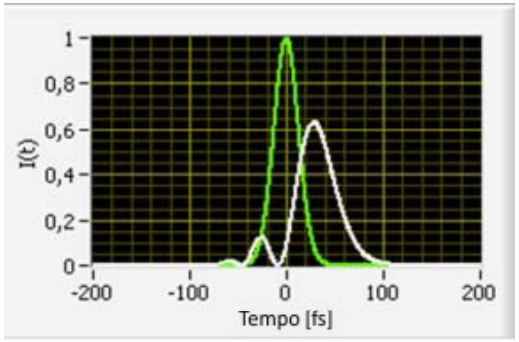
Pulse-shaping for coherent control



pulse shaper





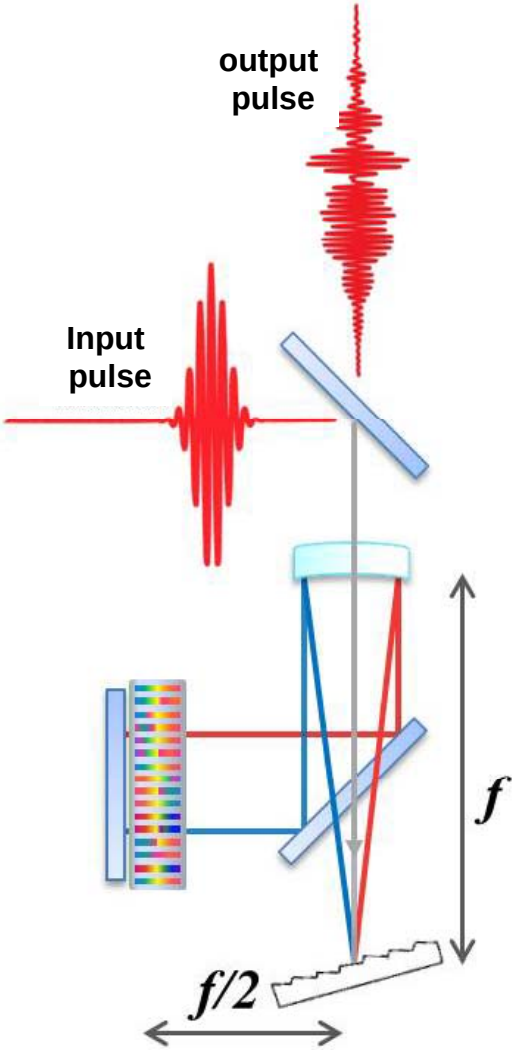


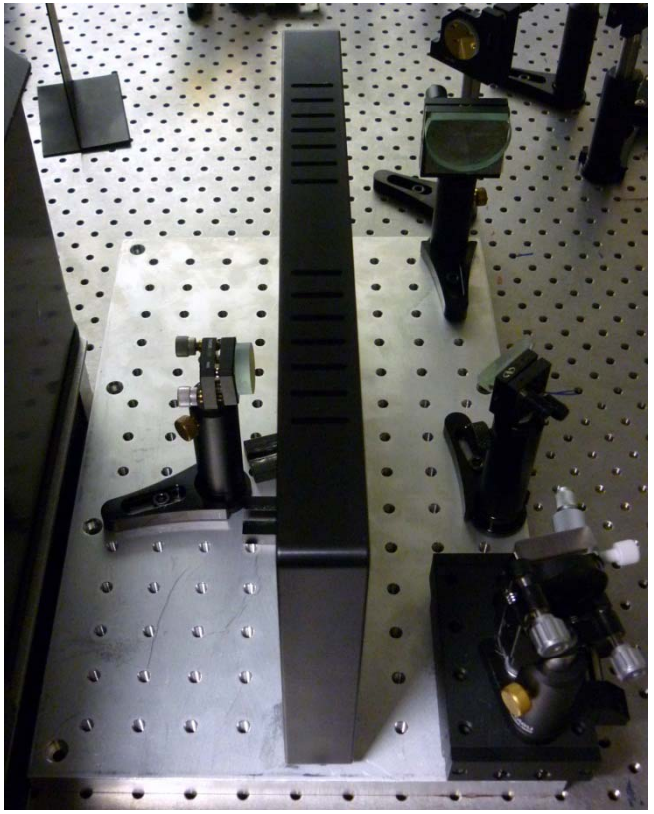
Experiment

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

τ = 30 fs
λ = 800 nm
f =1 kHz

fs-laser

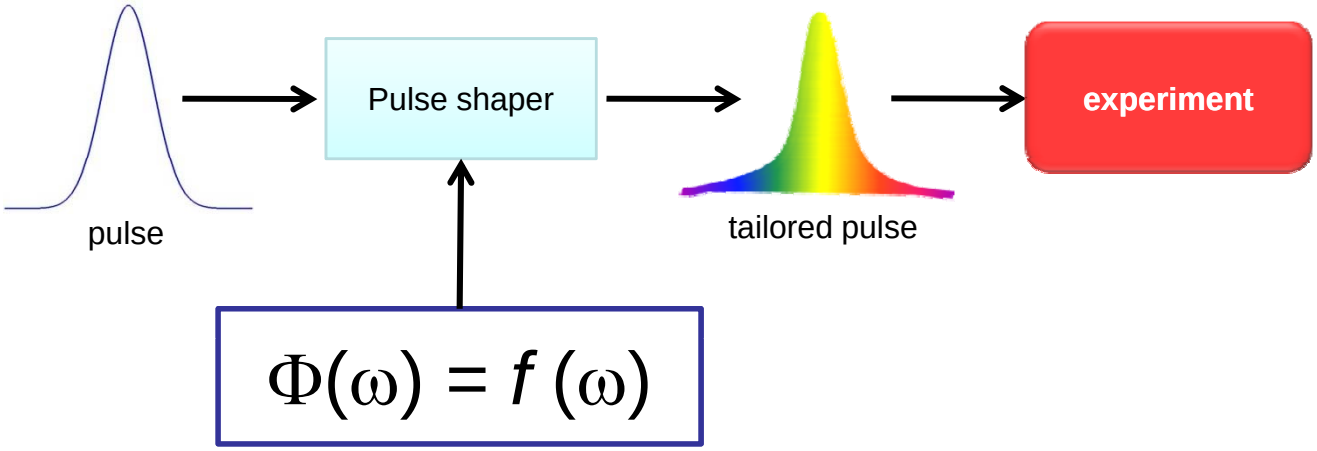
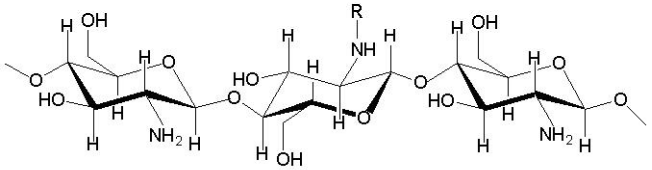
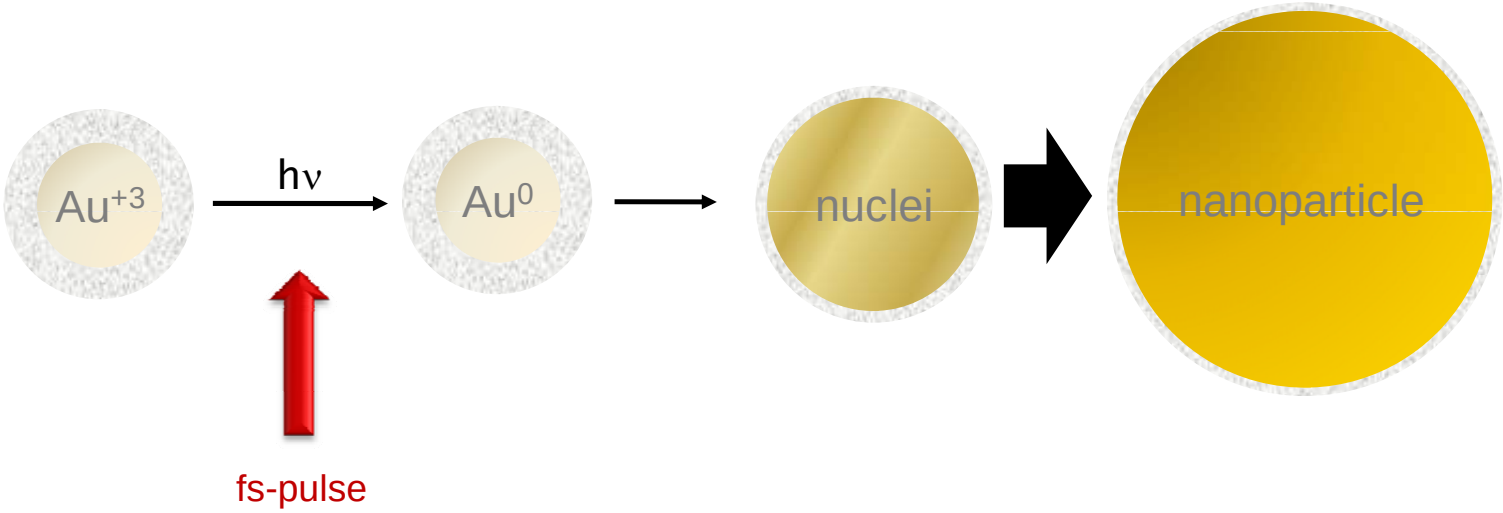


Photo-reduction

$HAuCl_4$ + chitosan

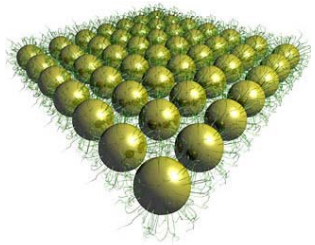




Control of Au nanoparticles formation

$Ch + nh\nu \rightarrow Ch^{+} + e^{-}$

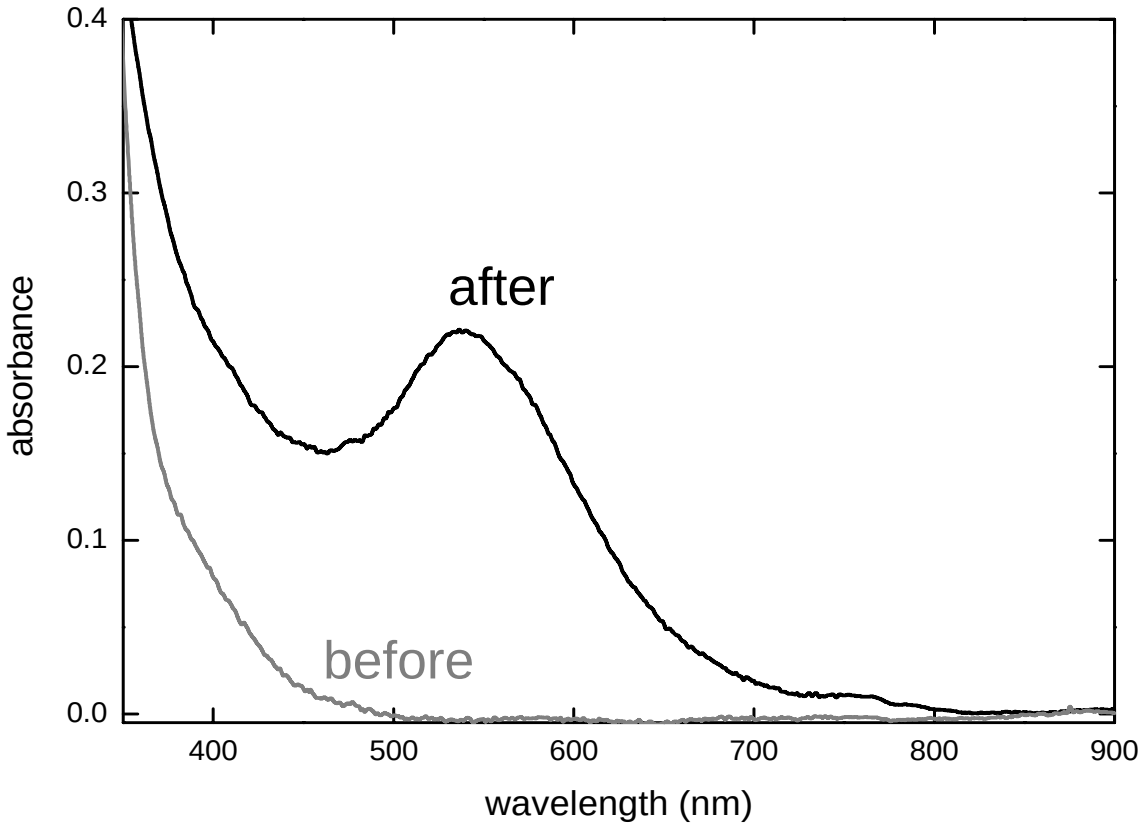
$Au^{+} + e^{-} \rightarrow Au^{0}$



excition laser used was a KMLabs – Dragon (multipass amplifier)

τ = 30 fs
λ = 800 nm
f =1 kHz
E = 2 mJ

Control of Au nanoparticles formation



FTL pulse

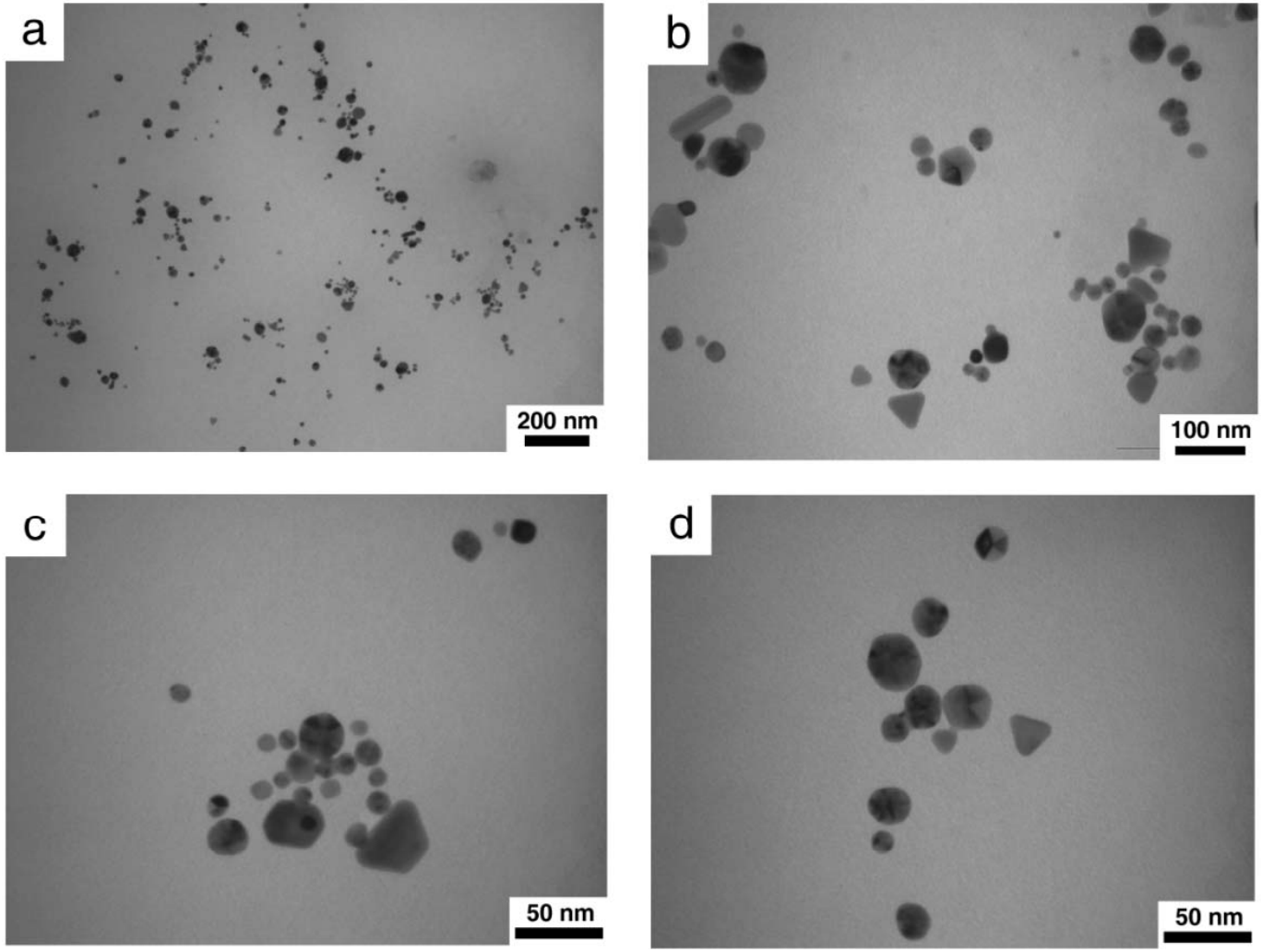
E = 185 μ*J*
I = 5 x 10^{11} W/cm^2

λ= 800 nm

sample absorption spectrum before and after irradiation

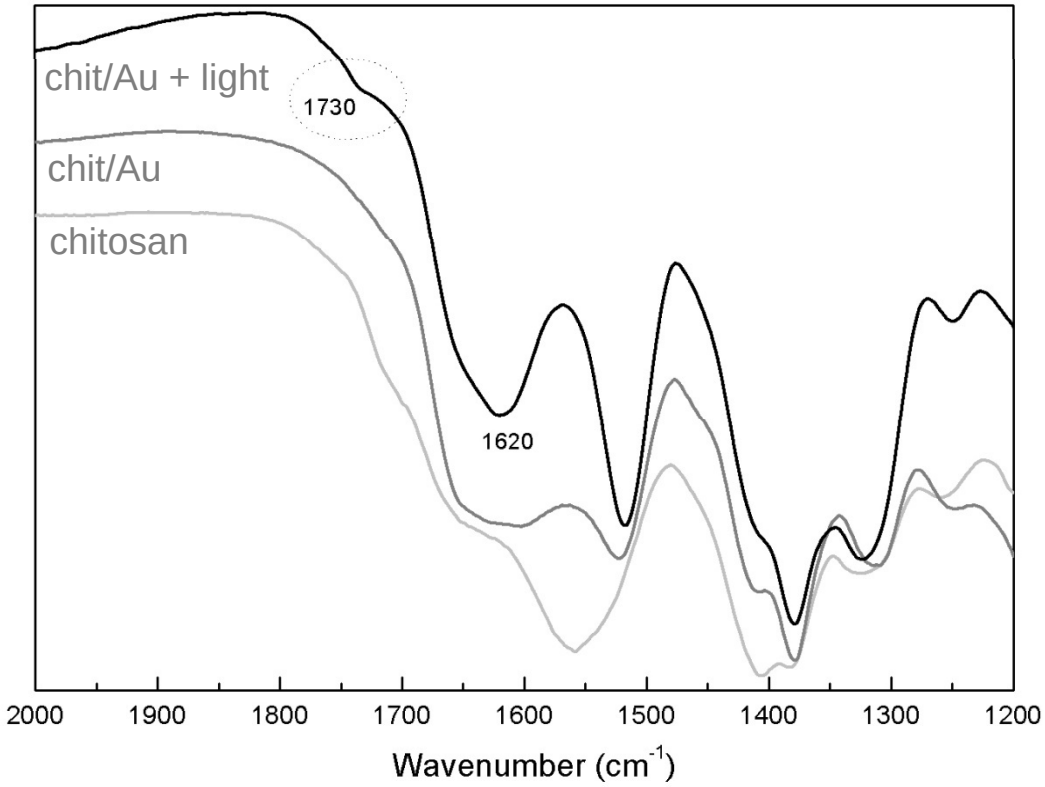
Results

Gold nanoparticles – TEM images



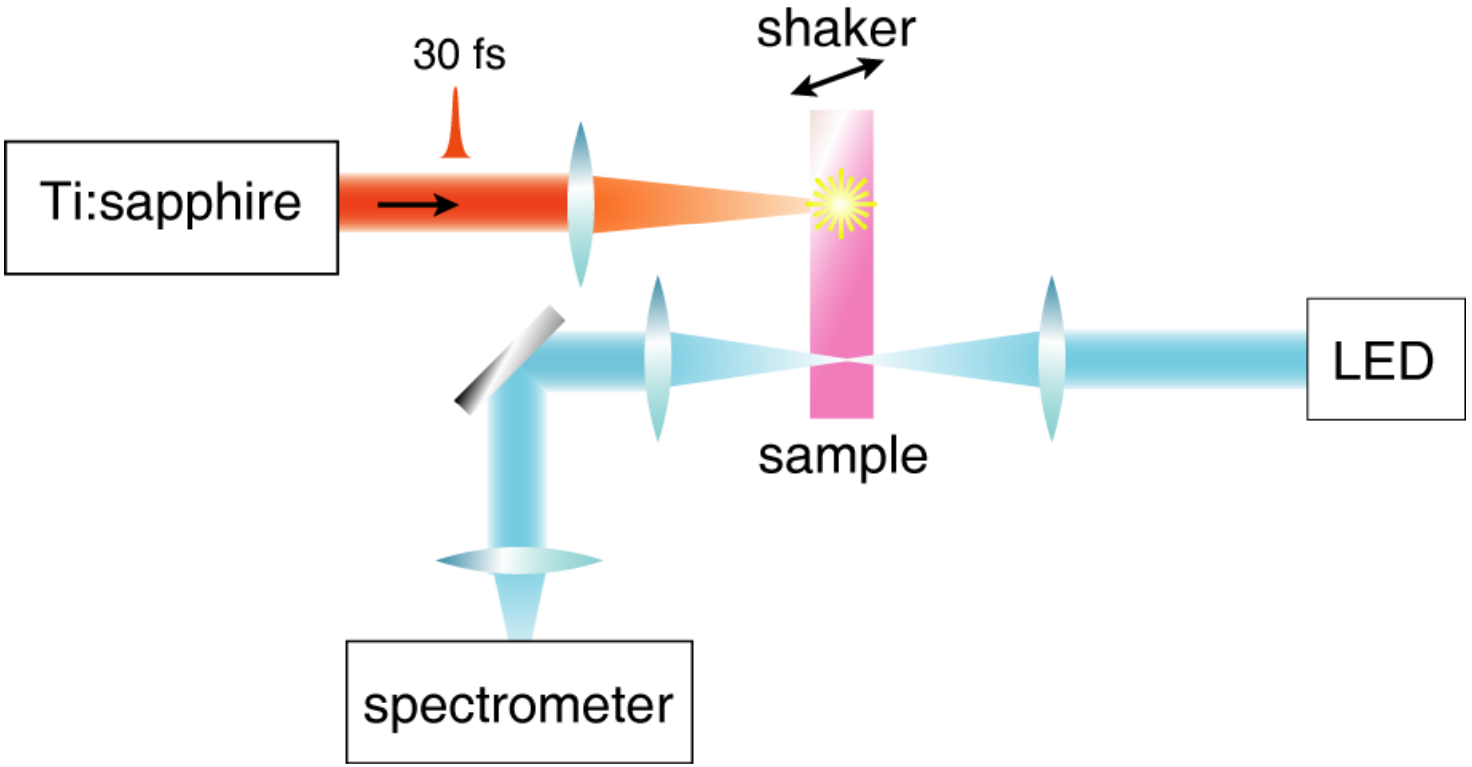
Control of Au nanoparticles formation

FTIR spectra of the samples



indicates that the reduction of the gold ions for the formation of the gold nanoparticles is related to the oxidation of hydroxyl groups in chitosan to carbonyl groups

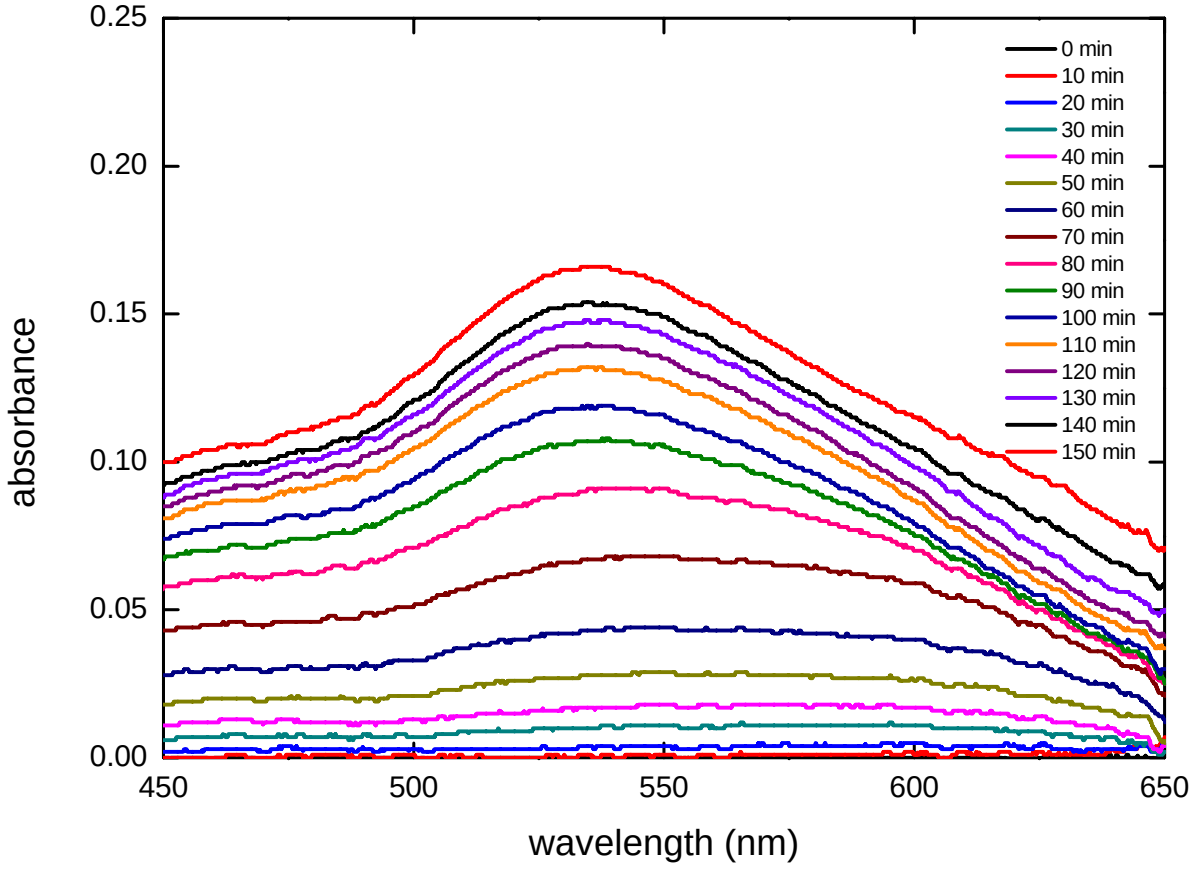
Control of Au nanoparticles formation



to determine the the dynamics of nanoparticles formation

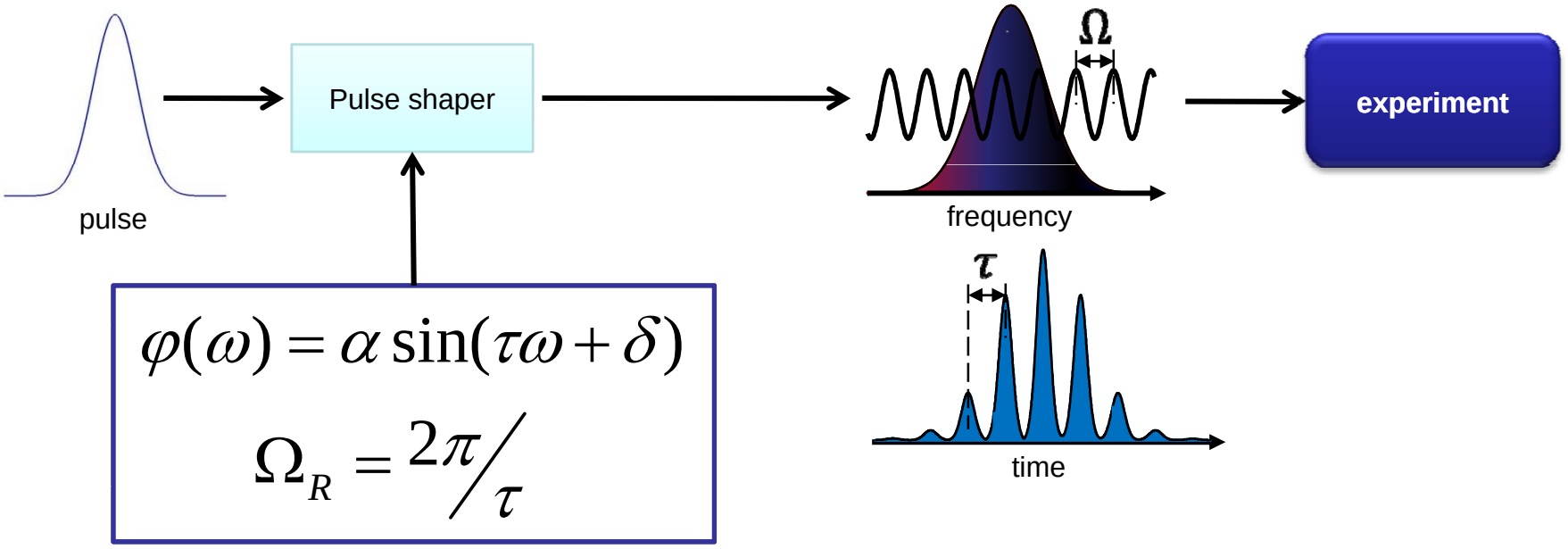
Control of Au nanoparticles formation

$I = 12 \times 10^{11}$ W/cm^2

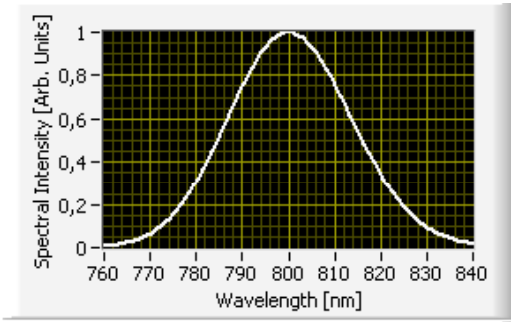


Coherent control

creating a pulse train



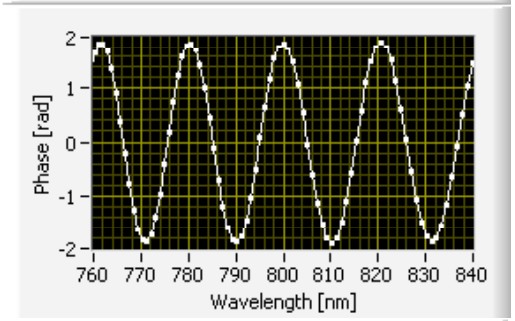
Control of Au nanoparticles formation

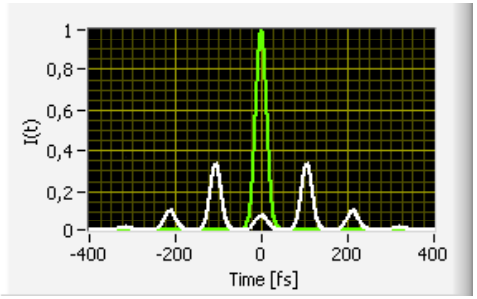


varying the period of the sinusoidal phase mask

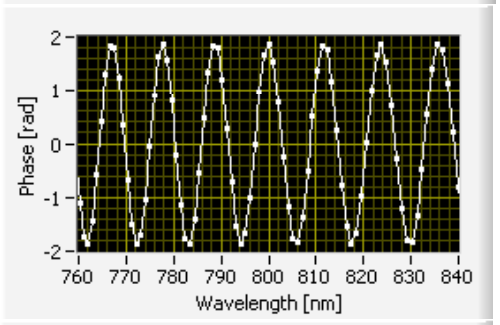
$$\emptyset(\omega) = \alpha\, sin(\gamma\omega + \delta)$$

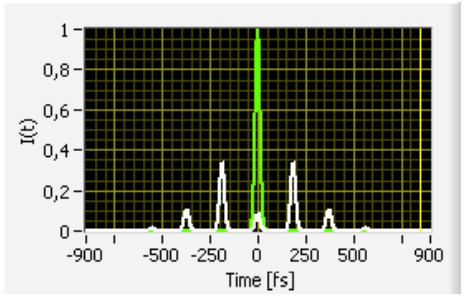
generate pulse trains with distinct separation time



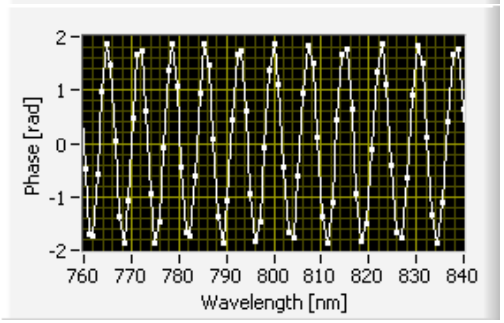


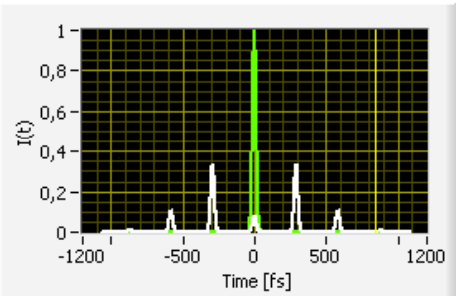
$N_{periods} = 4$
$t_{sep} = 106$ fs





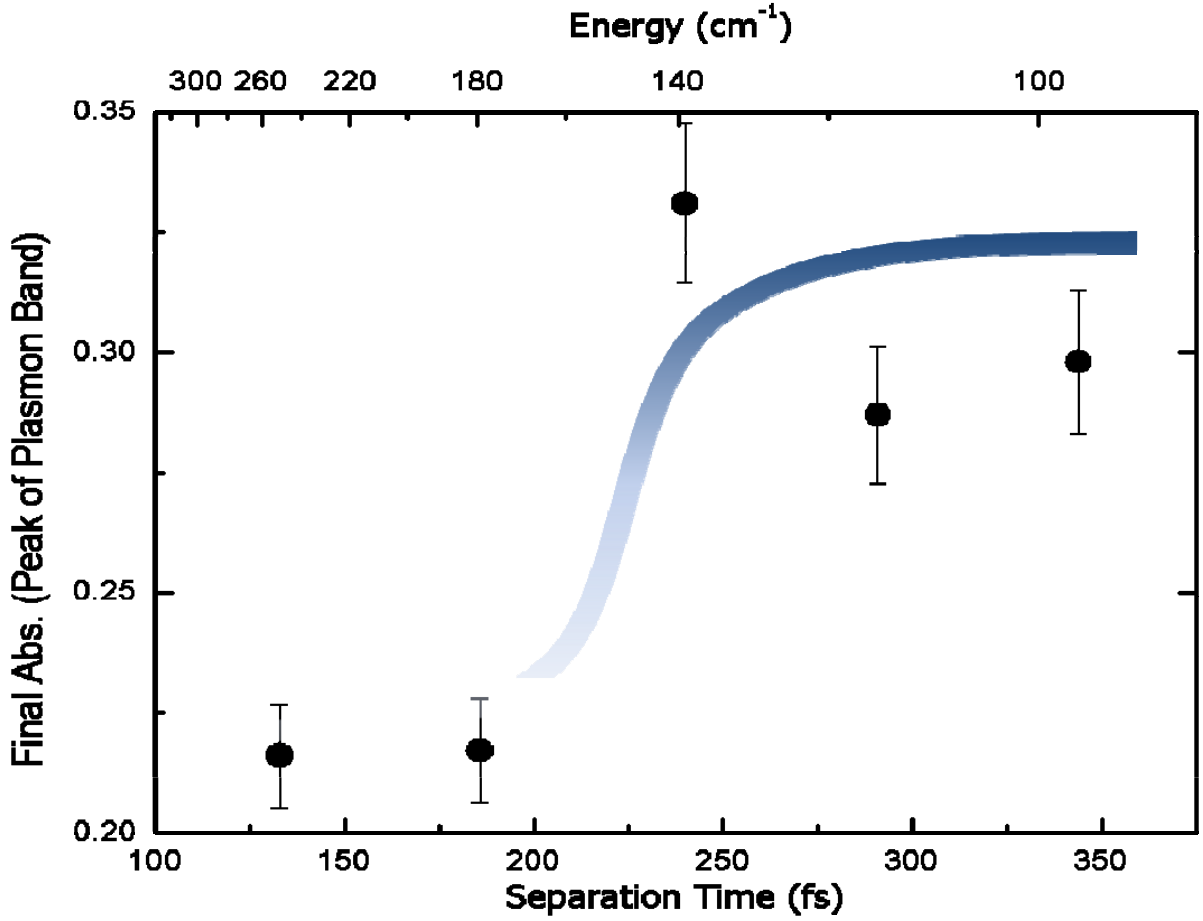
$N_{periods} = 7$
$t_{sep} = 186$ fs



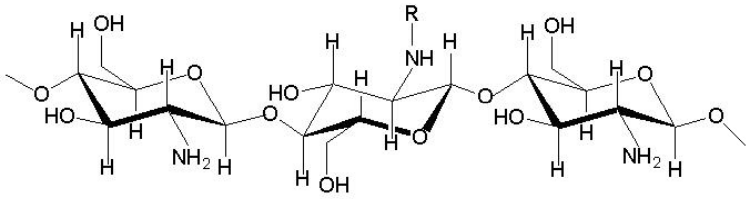


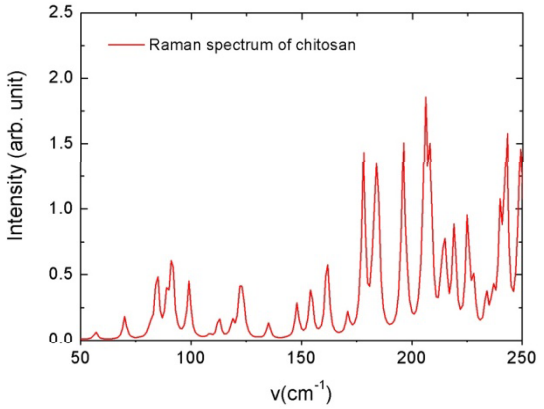
$N_{periods} = 11$
$t_{sep} = 291$ fs

Control of Au nanoparticles formation

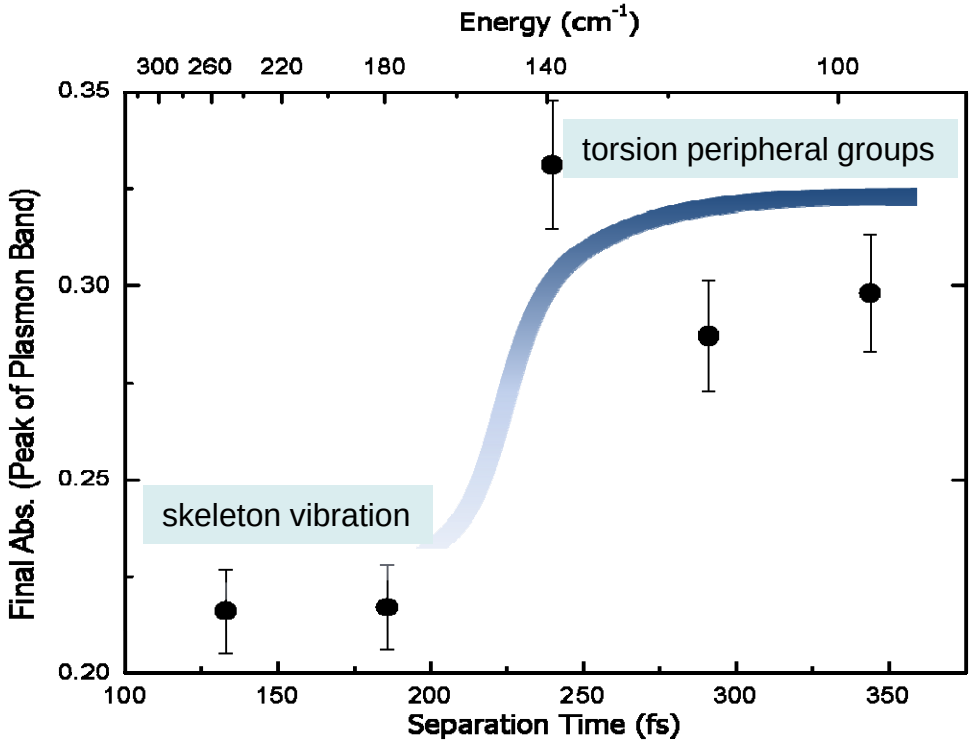


Control of Au nanoparticles formation





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



Conclusions

Coherent control methods seems to be an interesting option for the synthesis of gold nanoparticles production

Acknowledgments

www.fotonica.ifsc.usp.br

Acknowledgments

Thank you !

for a copy of this presentation

http://www.photonics.ifsc.usp.br

presentation