Mesures pompe-sonde pour le diagnostic des plasmas laser créés lors de la nanostructuration des matériaux

-Laser technologies
-Carrier plasmas
-Pump-probe review
-3D structuring

R. STOIAN

LaHC, Saint Etienne, France

Laser processing

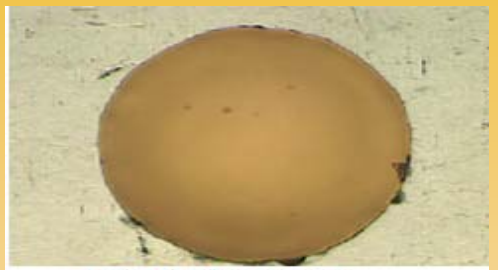
Micro and Nano fabrication

- Key technological issue in manufacturing

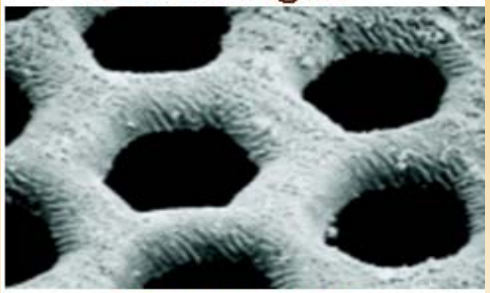
2D surface engineering

Fabrication

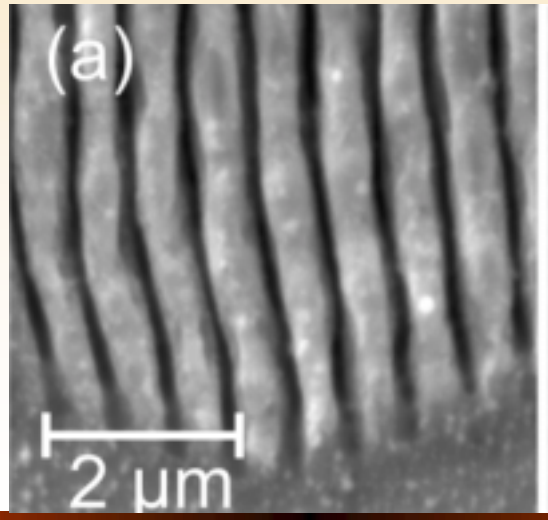
- Drilling
- Functional Microstructures
- Tribology
- Mechanics
- Electronics
- etc…..



IFSW Stuttgart

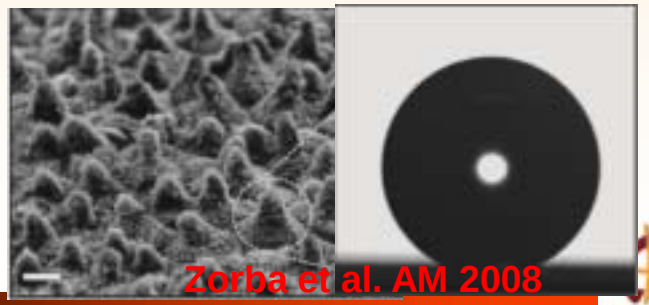


LZH Hannover



LaHC Saint Etienne

FORTH Crete



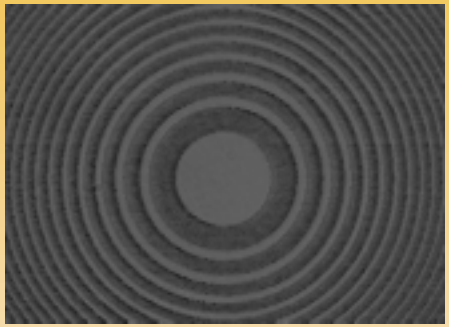
Zorba et al. AM 2008

3D optical functions

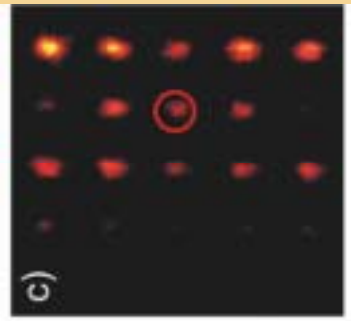
Fabrication

- Embedded optical elements
- Embedded lasers
- Photonic crystals
- Quantum information
- Optofluidics
- Astrophotonics
- etc…..

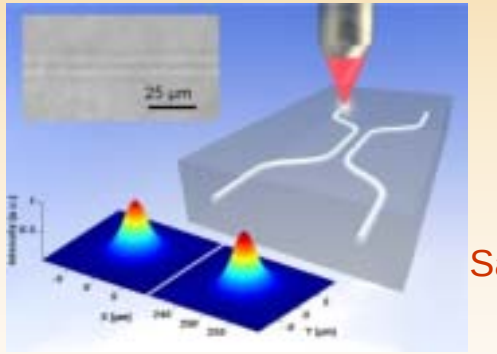
Watanabe OE 2002



Marshall OL 2008

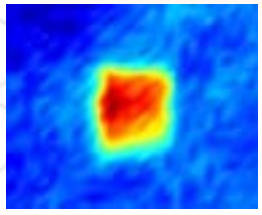


Szameit OE 2006



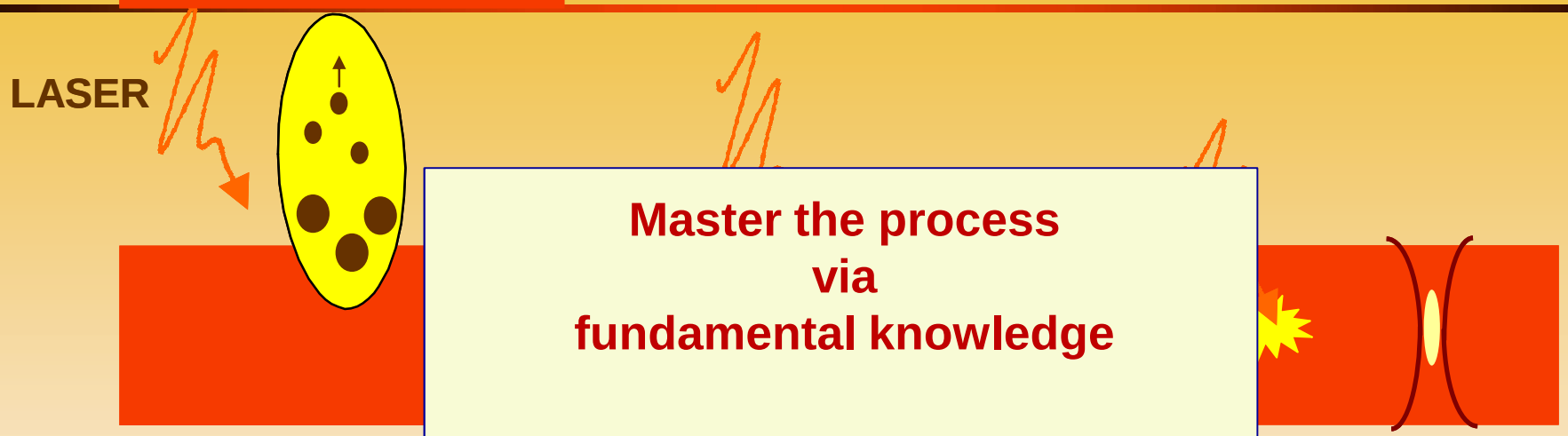
Sansoni PRL 2010

Cheng & Sugioka 2006



Thomson OE 2011

laser irradiation: enabling tool



Master the process
via
fundamental knowledge

-Laser ablation
-Material transfe
-Elemental spec

rocessing
ctive index change
onic functions

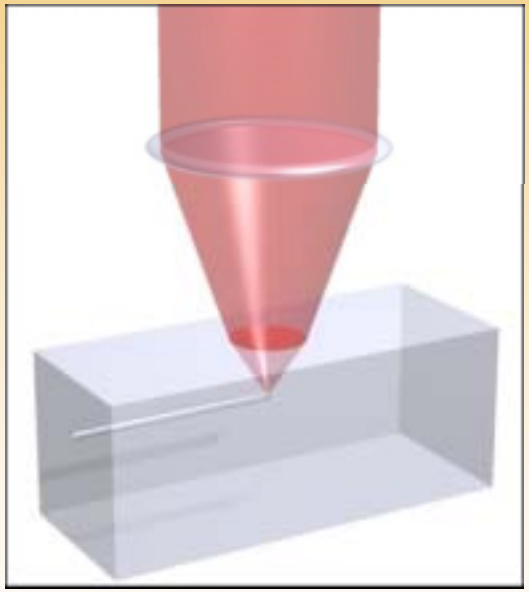
Probing laser processes
-a review of ultrafast probing

Material functions

Fabrication

- Involve lasers

But which lasers?



Long versus short pulses

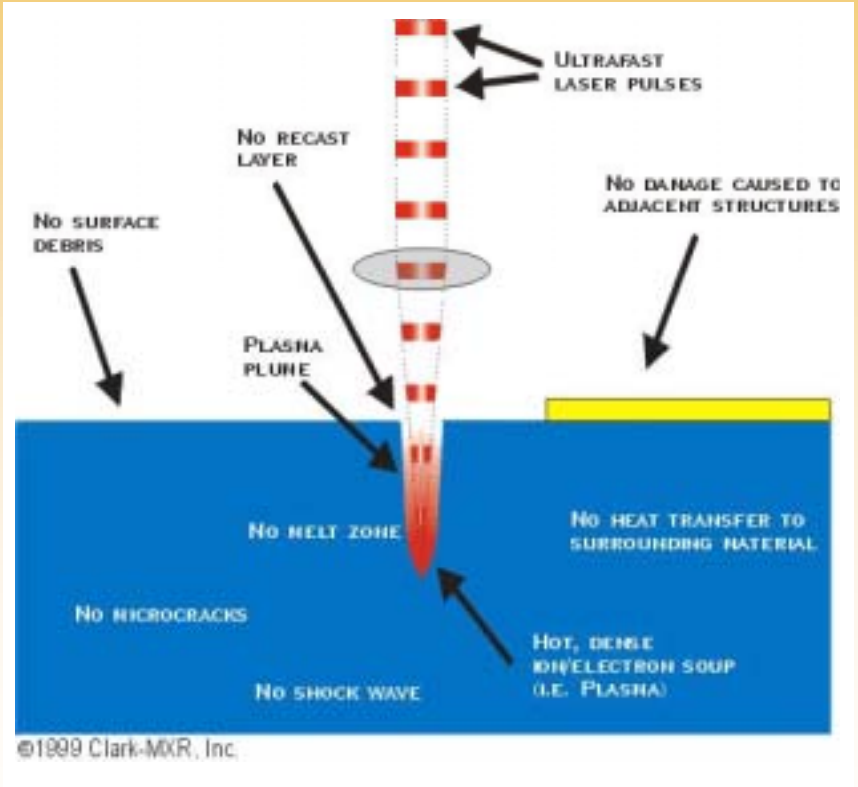
Long pulse

$$P = \frac{E}{\tau} = \frac{1mJ}{100\,fs} = 10GW$$

$$P = \frac{E}{\tau} = \frac{1J}{10\,fs} = PW$$

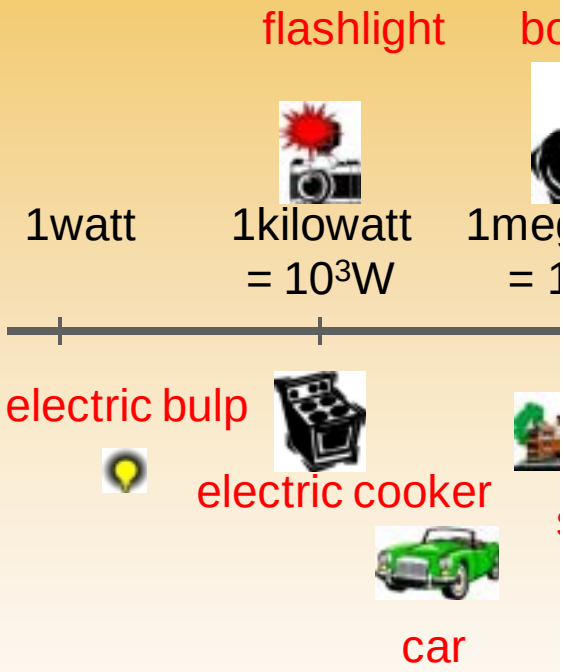
©1999 Clark-MXR, Inc.

Short pulse
femtosecond



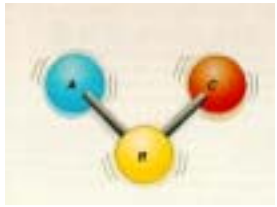
Clark-MXR web site

Long versus short pulses

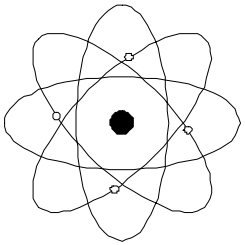


High intensities
(comparable with atomic fields)

Molecular Rotation /Vibration



Electron Motion

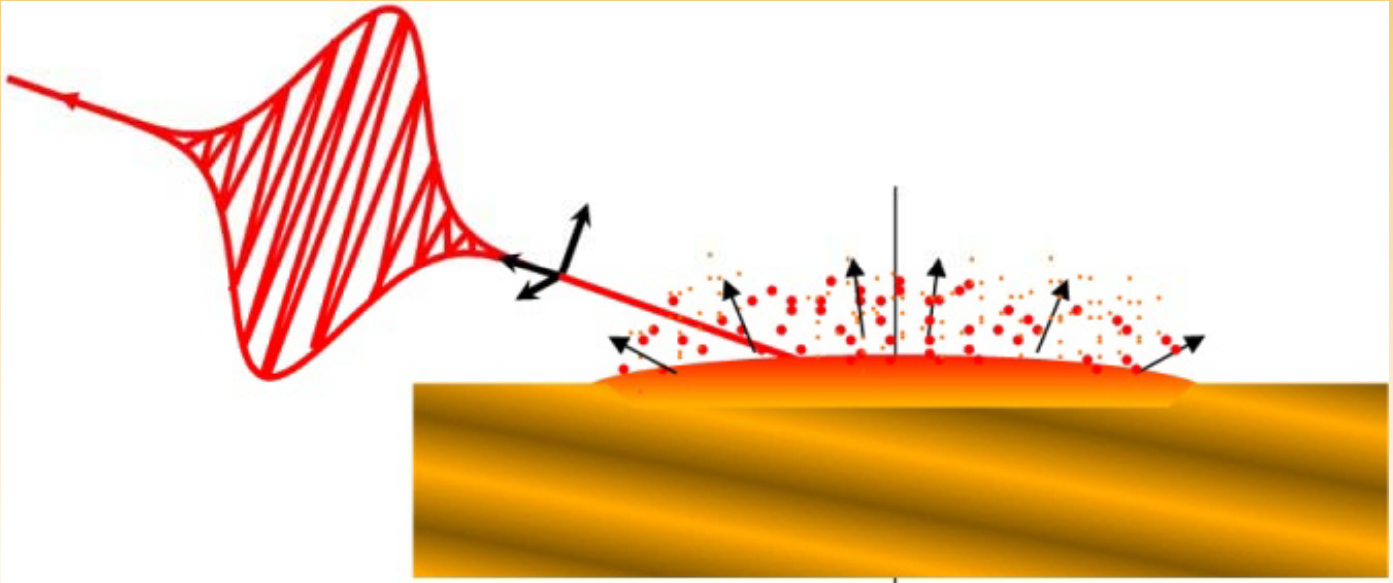


10^{-15} sec
femto

10^{-18} sec
atto

Laser action

Laser structuring = consequence of laser ablation



Energy coupling

⬇

Heating

⬇

Transformation

⬇

Ablation

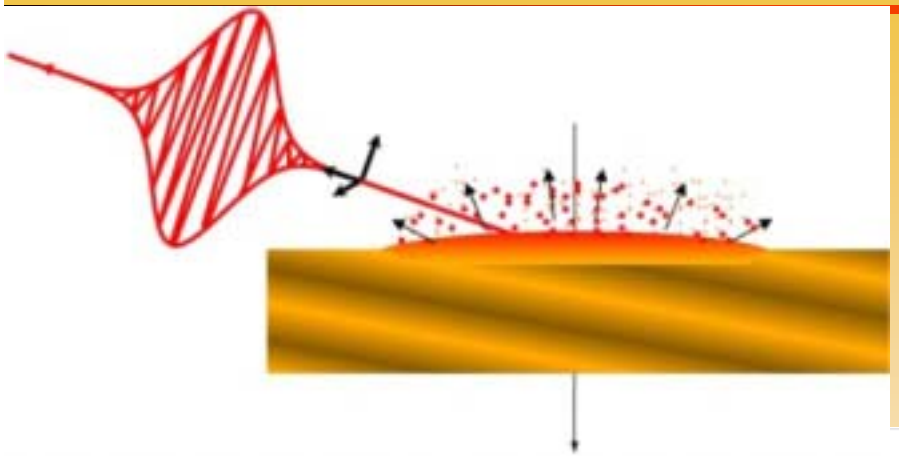
CAN THIS BE CONTROLLED ?

STRUCTURING

(achieve upgrade beyond the material limits?)

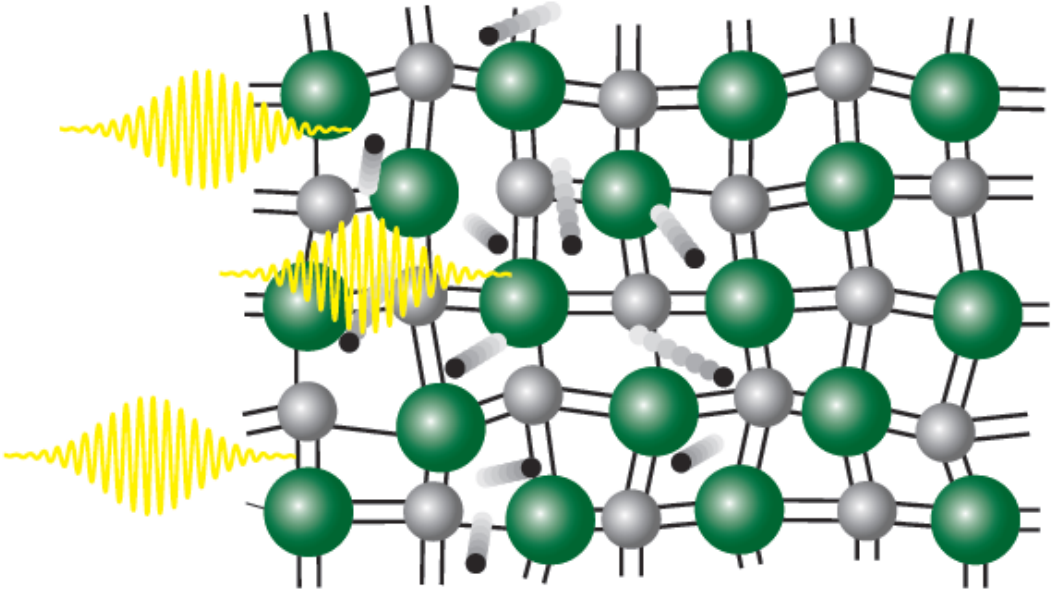
What is happening to the material: optically, structurally

Process dynamics



Laser ablation

- **Initial state**
- **Excitation**
- **Change**
- **Probe**



Dynamics

in 1878

Optical stroboscopy
10^{-1} to 10^{-3} seconds.

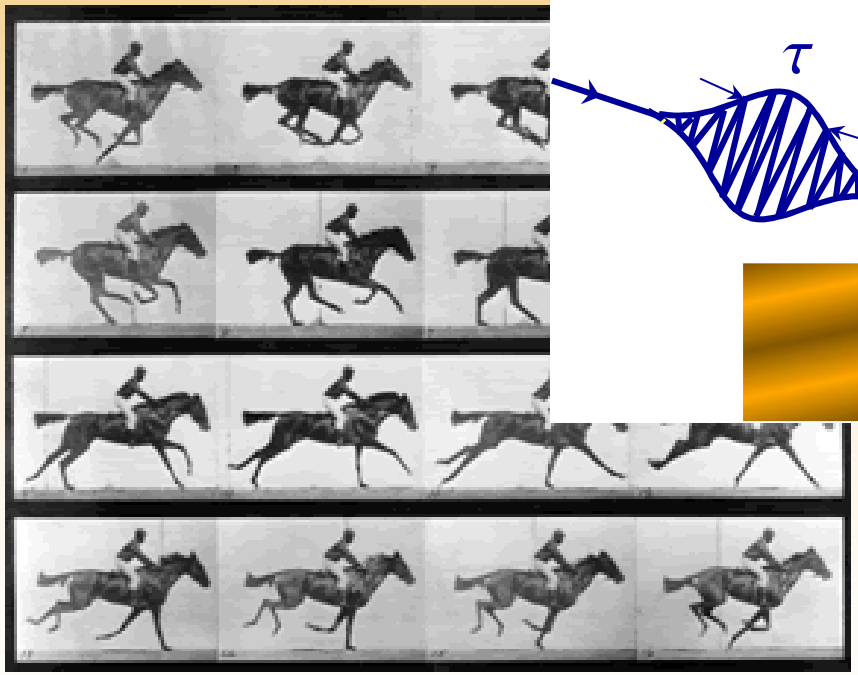
Ref. Eadweard Muybridge 1878

Dynamics

R. Trebino, GeorgiaTec

in 1878

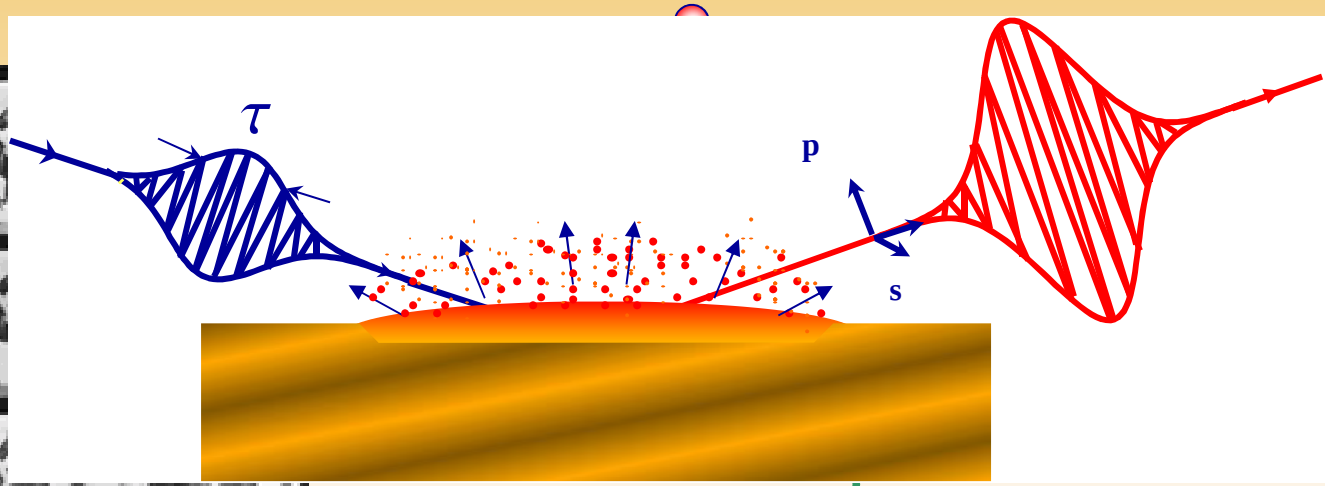
Optical stroboscopy
10^{-1} to 10^{-3} seconds.



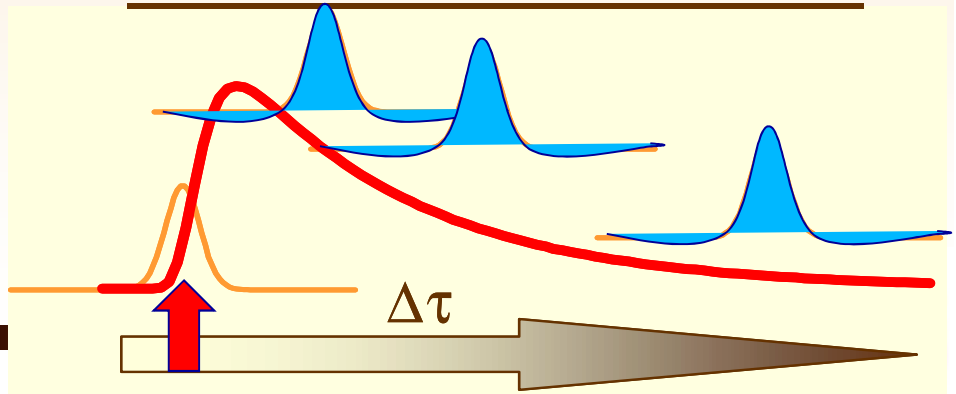
Ref. Eadweard Muybridge 1878

In > 2000

Ultrafast spectroscopy by pulsed laser pump probe 10^{-10} to 10^{-15} s



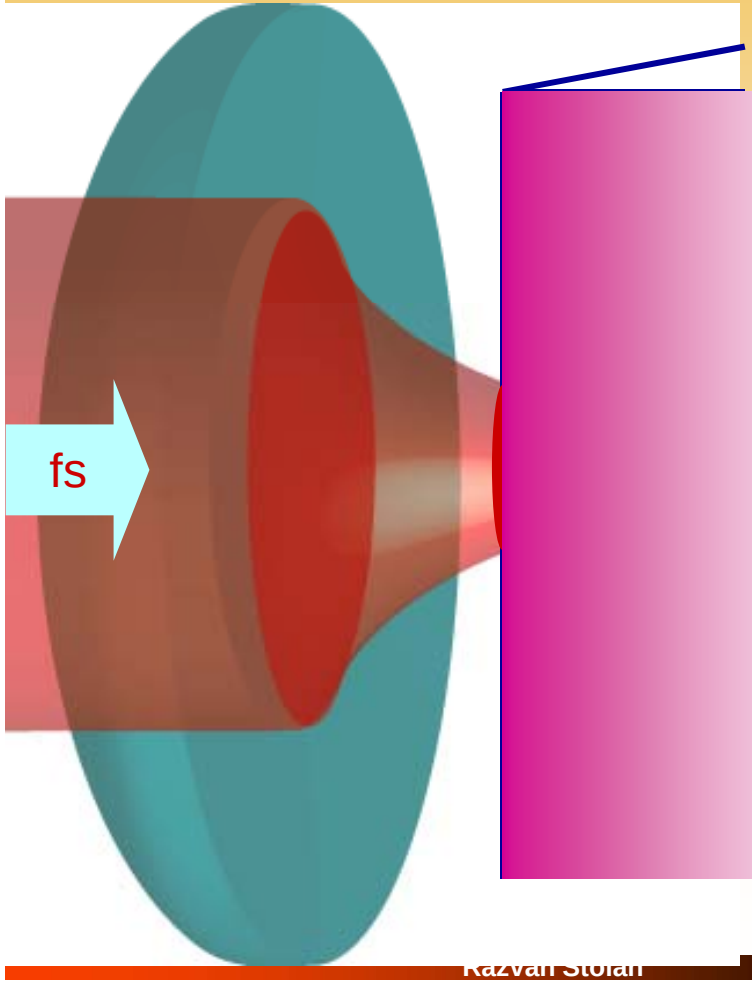
Molecular motion in real time



Laser-matter interaction

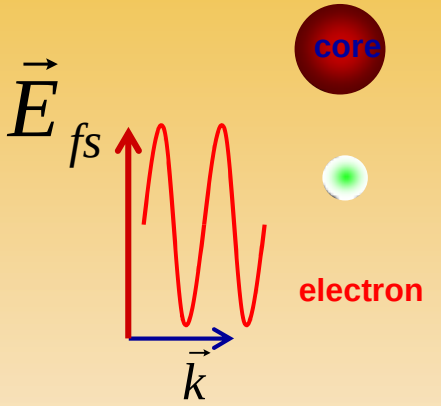
- **How is the laser energy deposited in materials?**
  - **ELECTRONS (CARRIER PLASMAS)**
  - **(confined in condensed matter)**

3D nonlinear excitation: ultrafast laser pulses

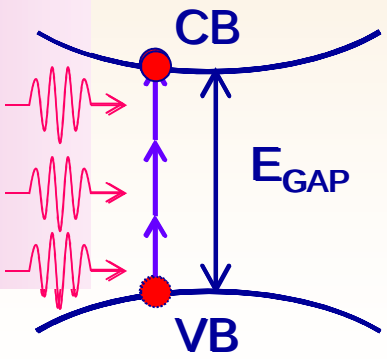


esponse
linear response

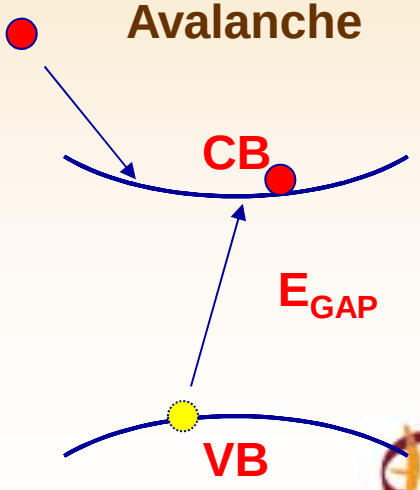
•Inv. Bremsstrahlung



DIONIZATION
ltiphotonic



COLLISIONAL
Avalanche



Electron; the forced (or free) oscillator

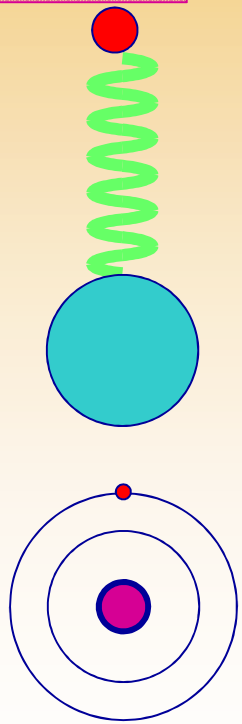
- Electron on a spring with position $x_e(t)$, and driven by a light wave, $E_0 \exp(-i\omega t)$:

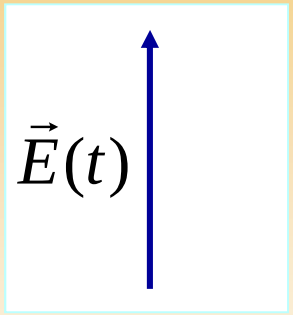
$$m_e d^2 x_e / dt^2 + m_e \omega_0^2 x_e = eE_0 \exp(-i\omega t)$$

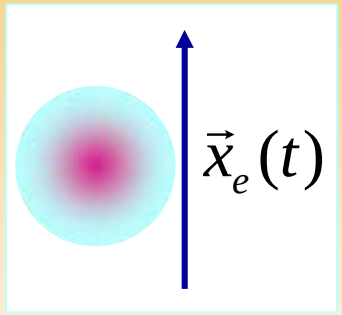
The solution is:

$$x_e(t) = \left[\frac{(e/m_e)}{(\omega_0^2 - \omega^2)} \right] E_0 \exp(-i\omega t)$$

Infinite amplitude at $\omega = \omega_0$



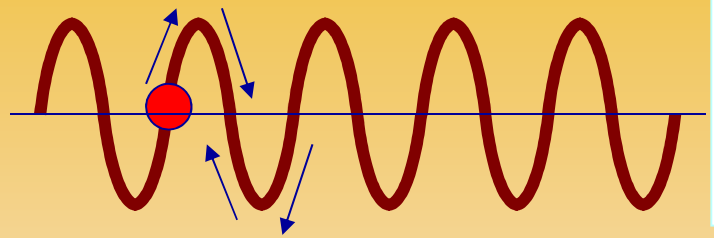


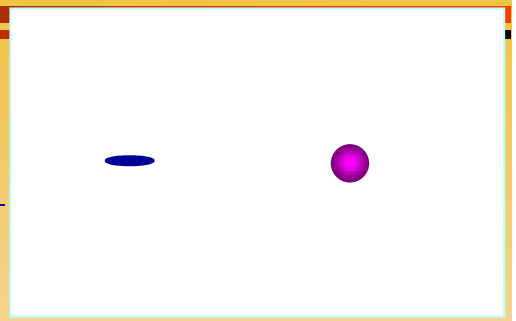


Free electrons

Force Oscillator

$$x_e(t) = \left[\frac{-(e/m_e)}{(\omega^2 + i\omega\gamma)} \right] E(t)$$

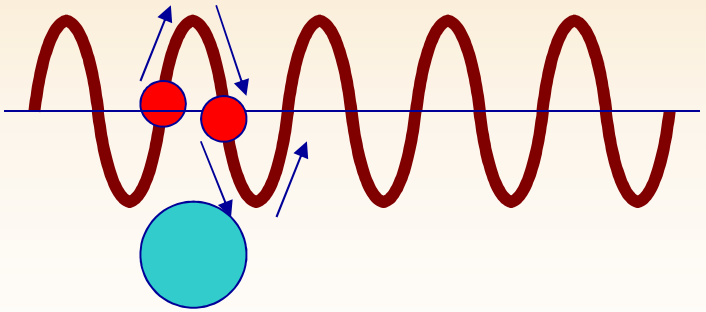




Energy stays constant $E_{kin}=E_{pot}$
no absorption on free electrons

(no energy and momentum conservation)

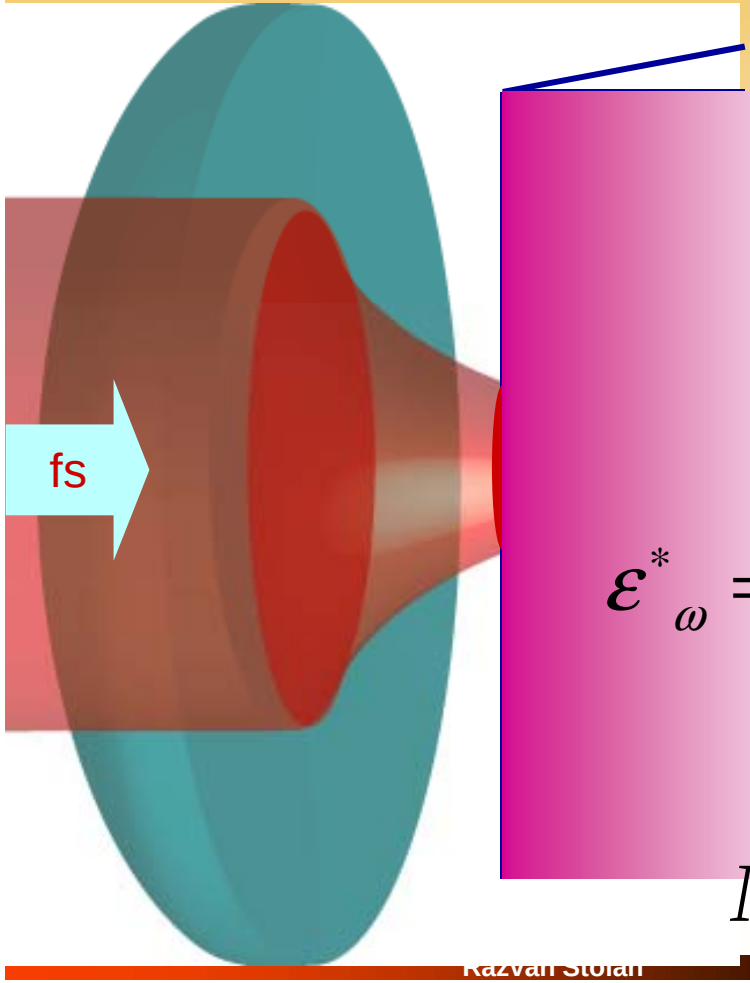
$$x_e(t) = \left[\frac{-(e/m_e)}{(\omega^2 + i\omega\gamma)} \right] E(t)$$

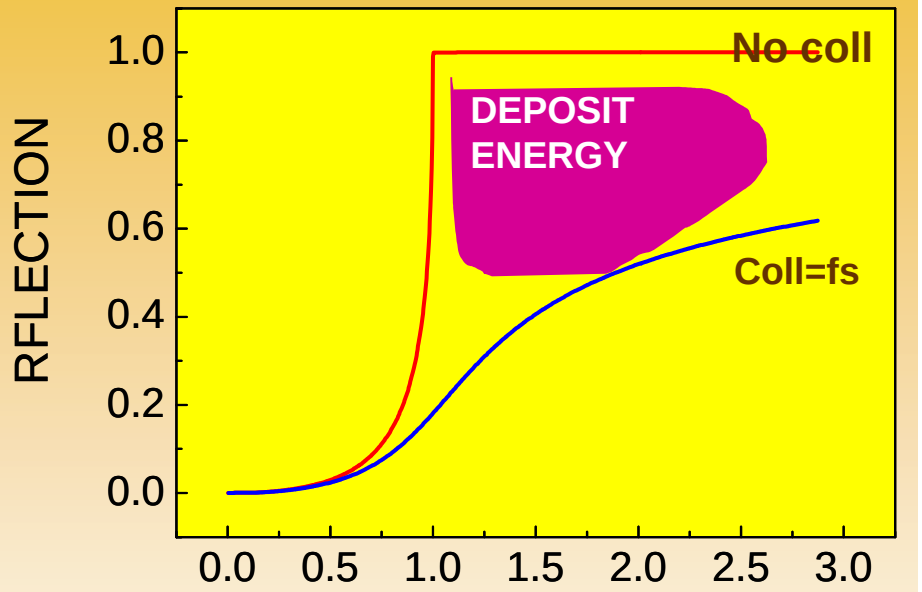


Energy varies because of collisions
ABSORPTION

3D nonlinear excitation: ultrafast laser pulses

Drude model



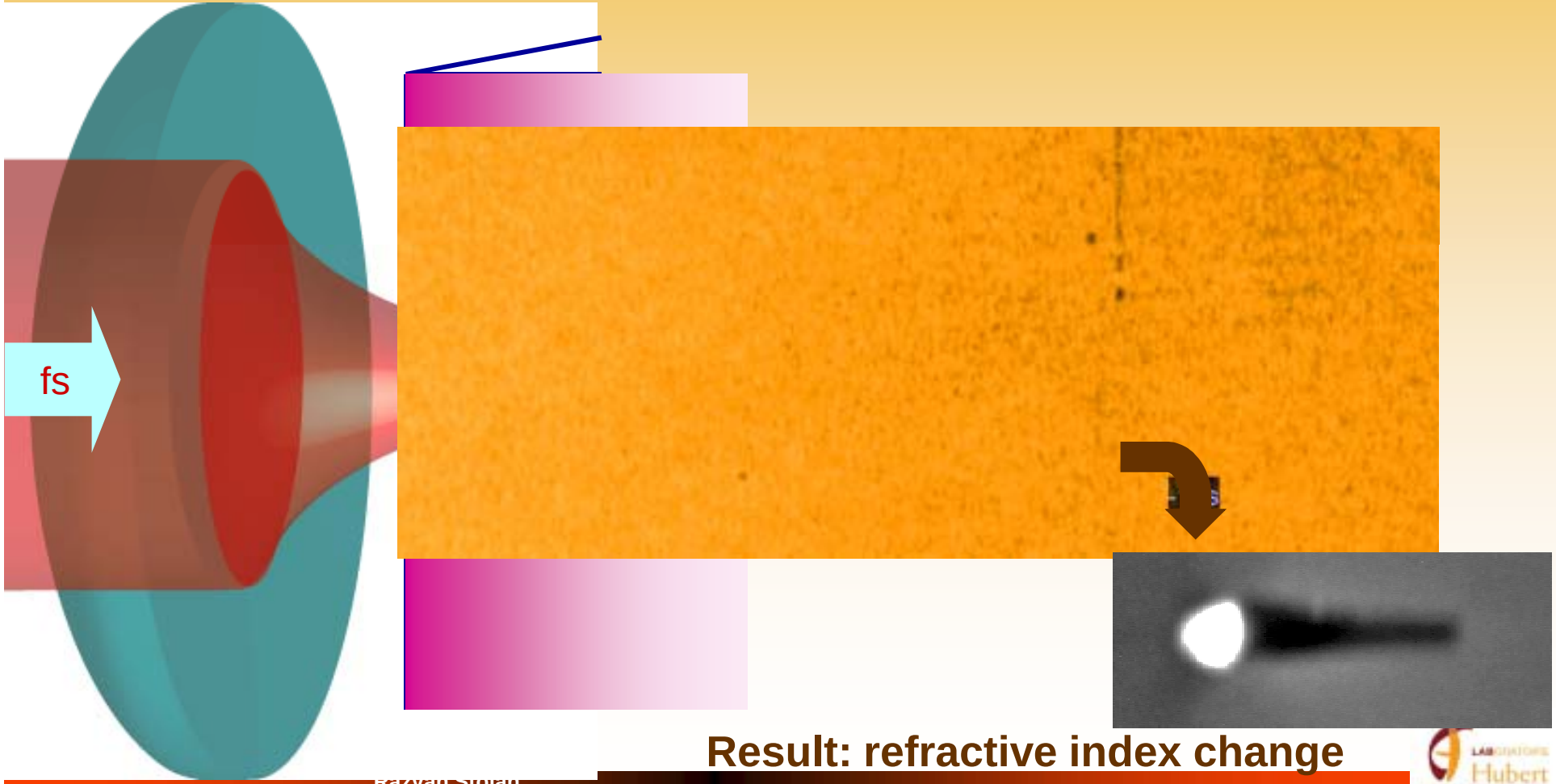


$$\varepsilon^*_\omega = \varepsilon_0 - \frac{N_e}{N_{cr}} \frac{1}{1 + i\dfrac{1}{\omega\tau}} = \varepsilon_r + i\varepsilon_i$$

N_e τ m_e

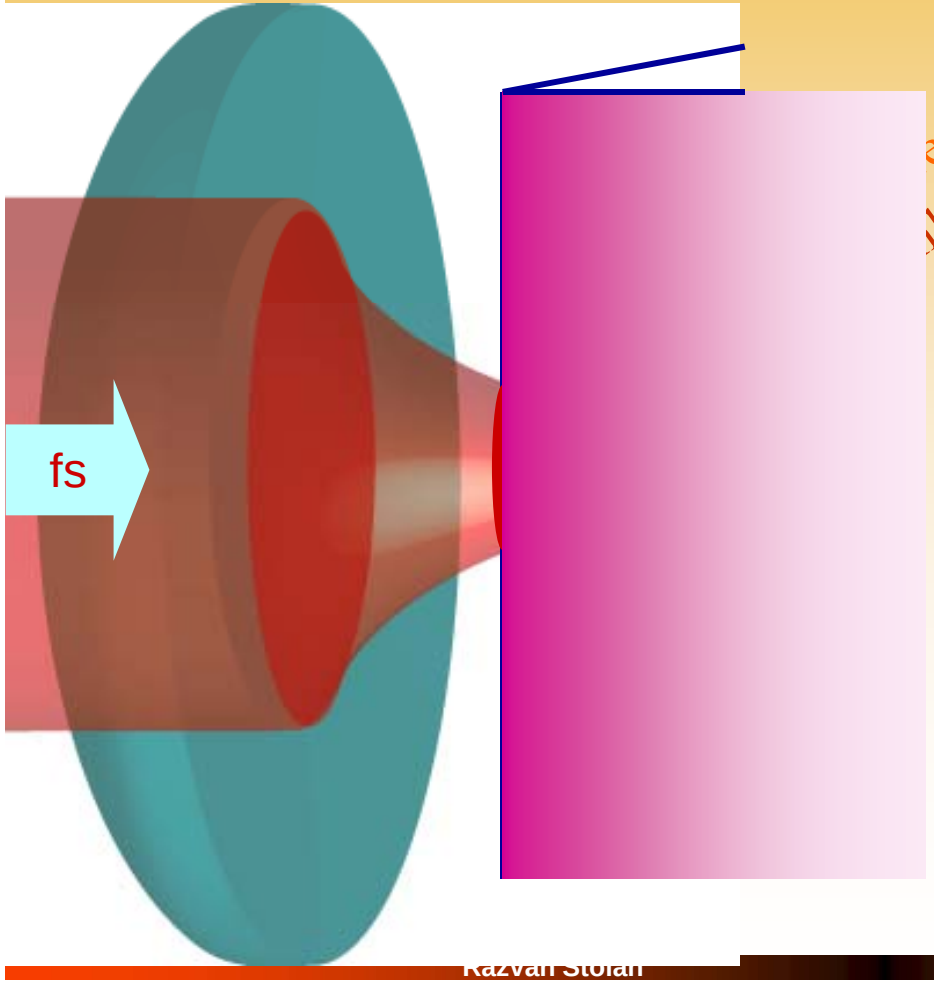
$$N_{cr} = \varepsilon_0 m^*_e \omega^2 / e^2 \qquad \mathrm{Re}\left[\varepsilon^*_\omega\right] = 0$$

3D nonlinear excitation: ultrafast laser pulses



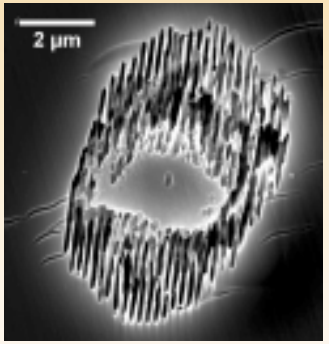
Result: refractive index change

Nonlinear excitation: ultrafast laser pulses



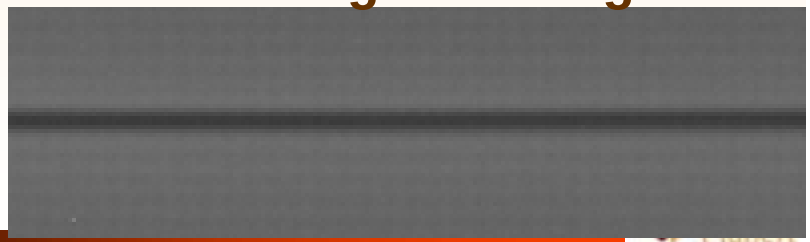
esponse
linear response

Microexplosions:
New matter states



Self – organization

Index changes: waveguides



Excitation of carrier plasmas

Q1: Ionization/Excitation

Optical dynamics:

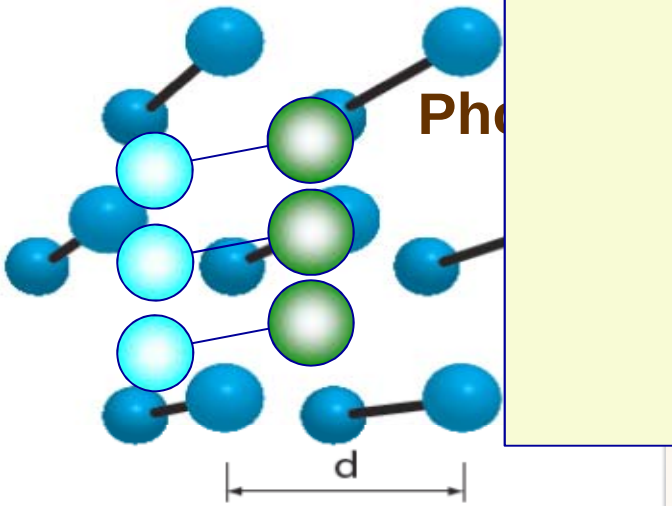
Changes in optical properties

-Reflectivity

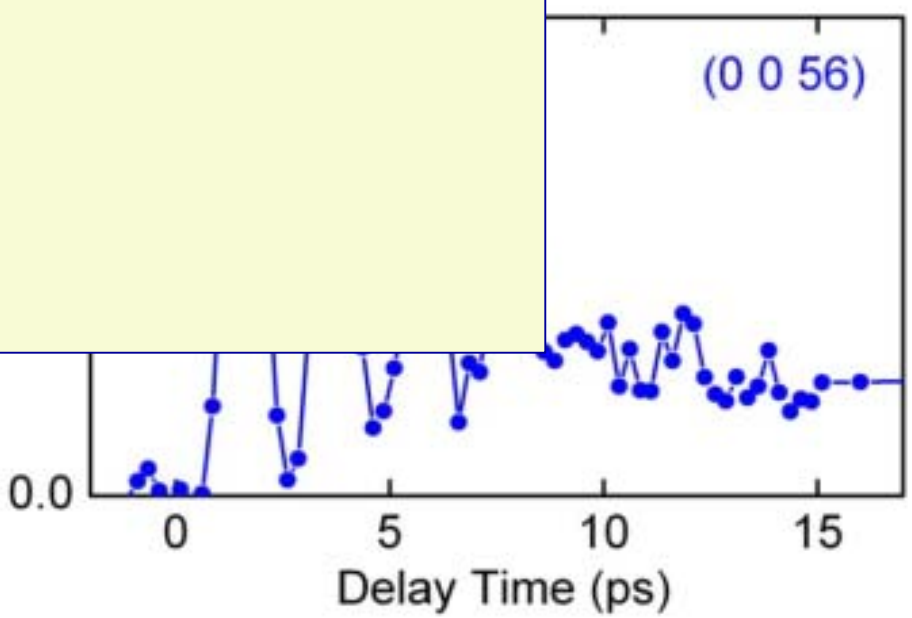
-Absobtivity

Example1: Coherent vibrations

ration to heat



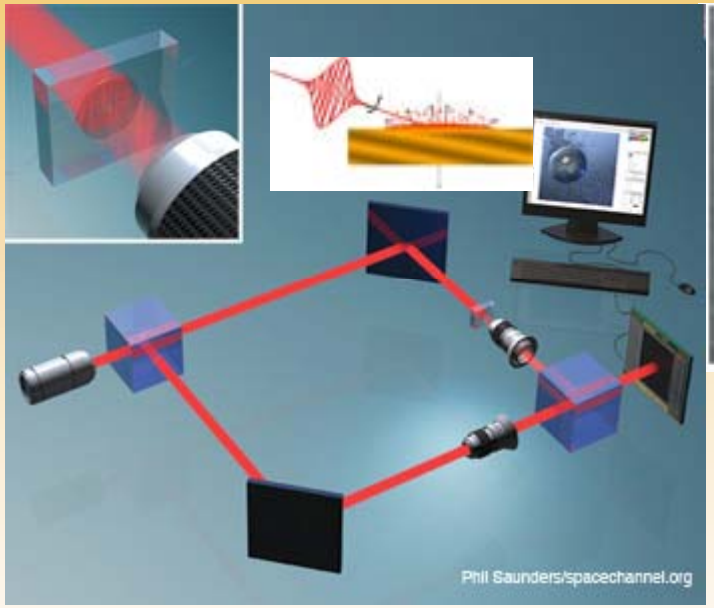
Energy coupling



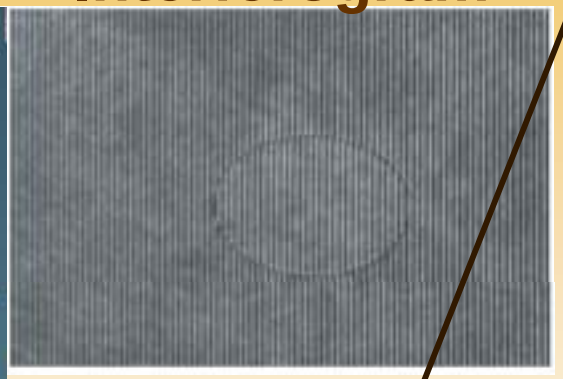
Digital holography

$$n^* \simeq n_0 - \frac{N_e}{2N_{cr}} \frac{1}{1+(1/\omega\tau)^2}$$

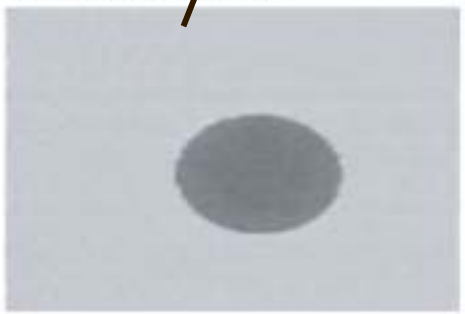
$$k^* \simeq \frac{N_e}{2N_{cr}} \frac{1/\omega\tau}{1+(1/\omega\tau)^2}$$



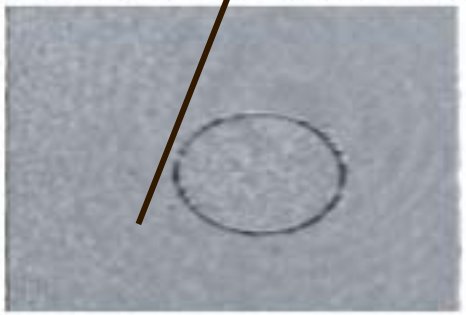
Interferogram



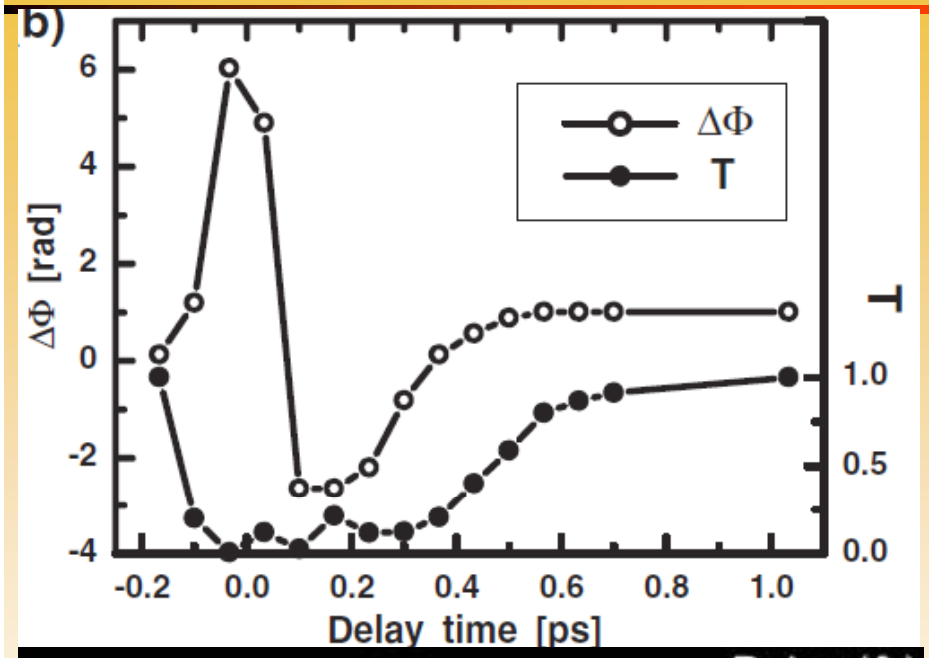
Phase: φ

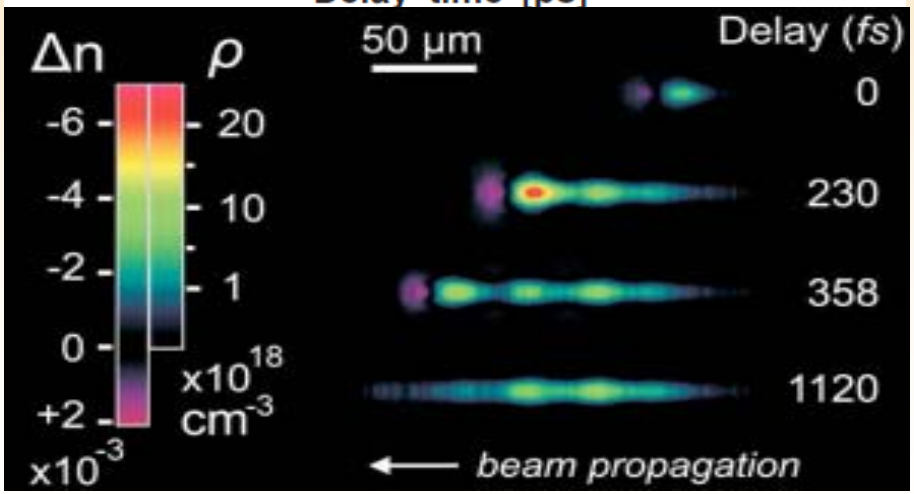


Amplitude T

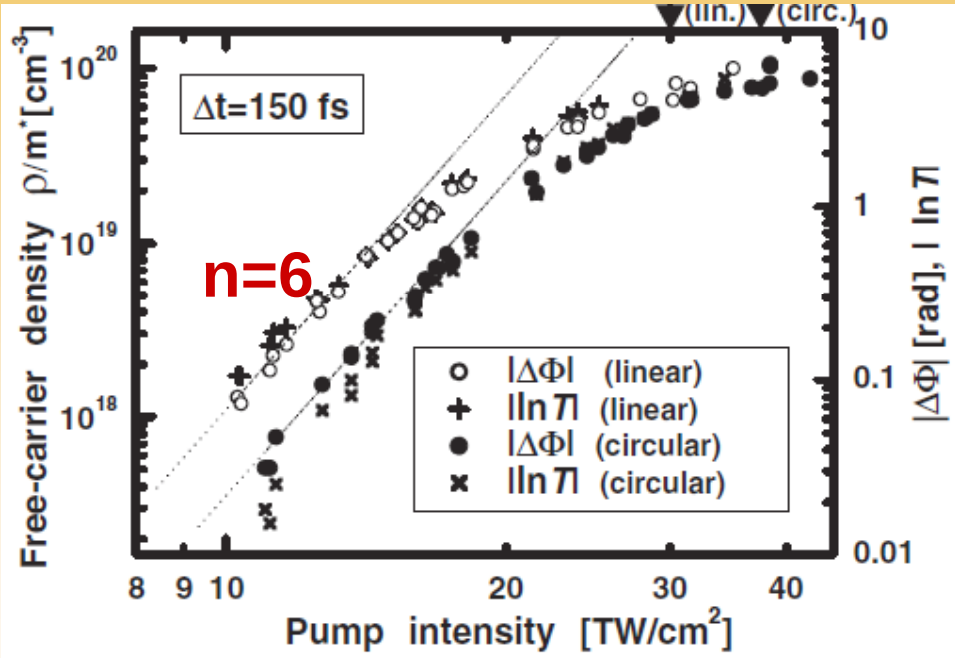


Ionization of carrier plasmas in dielectrics





sapphire



$$\frac{dN_e}{dt} \sim I^n \qquad \frac{N_e}{m_e}$$

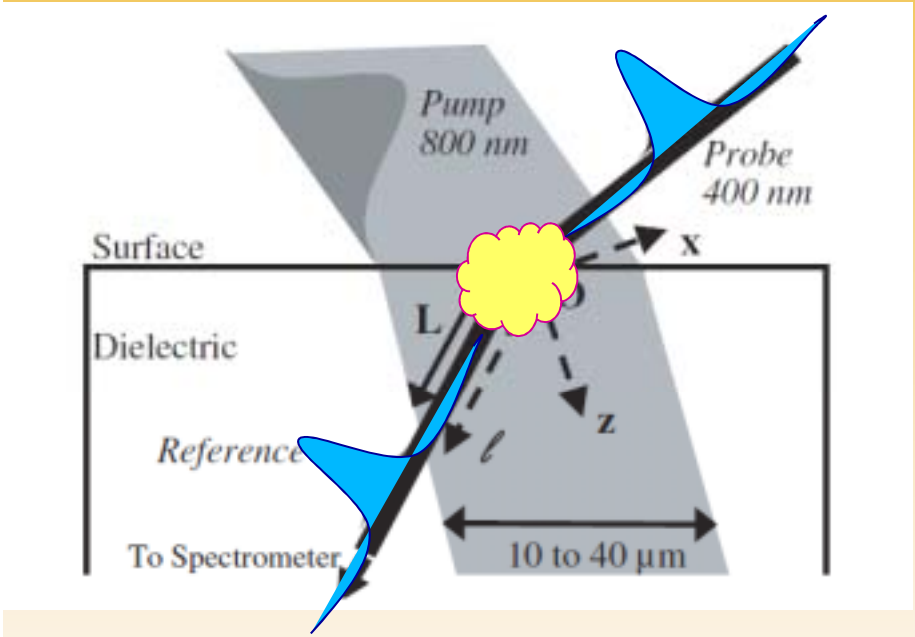
Temnov PRL 2006, Papazoglou APL 2009

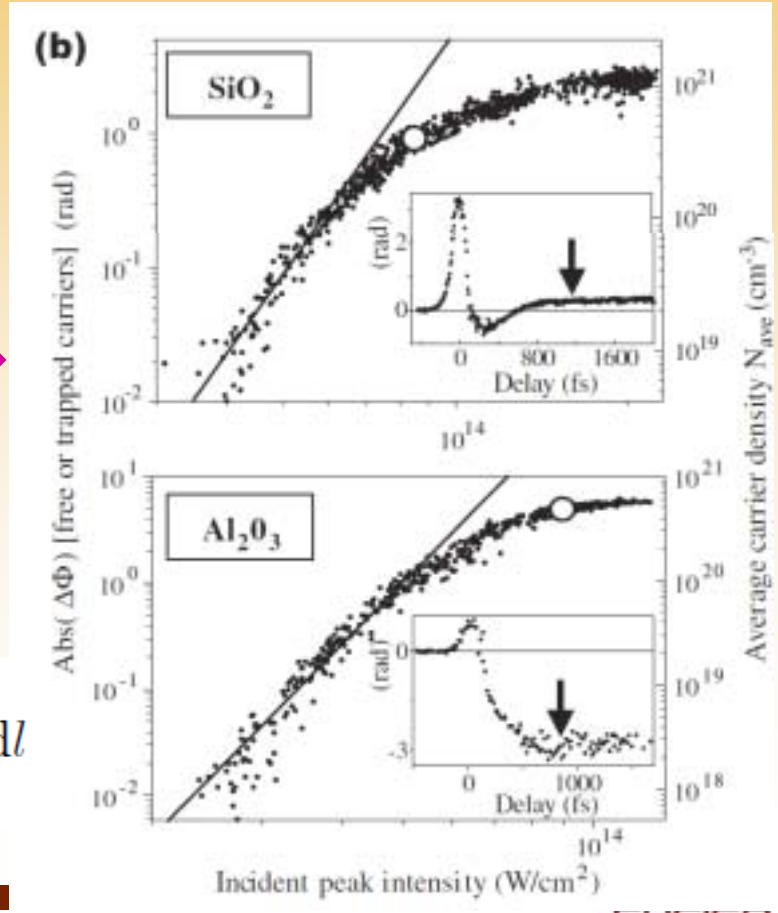
Excitation of carrier plasmas

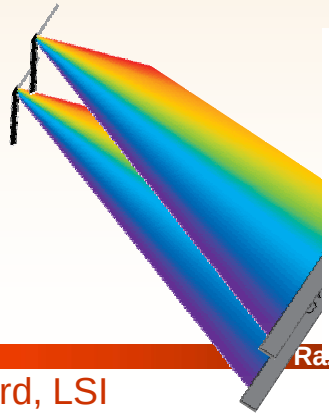
Q2: Relaxation

Spectral interferometry

$$n^* \simeq n_0 - \frac{N_e}{2N_{cr}} \frac{1}{1+\left(1/\omega\tau\right)^2}$$

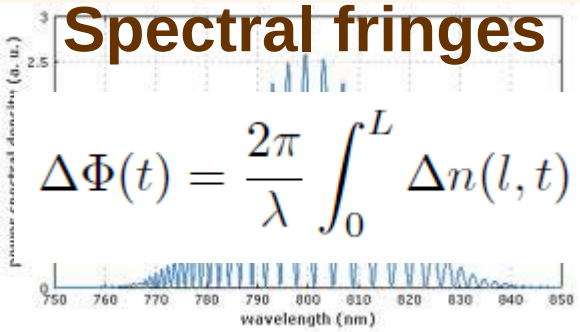






Spectral fringes

$$\Delta\Phi(t) = \frac{2\pi}{\lambda}\int_0^L \Delta n(l,t)\cdot \mathrm{d}l$$

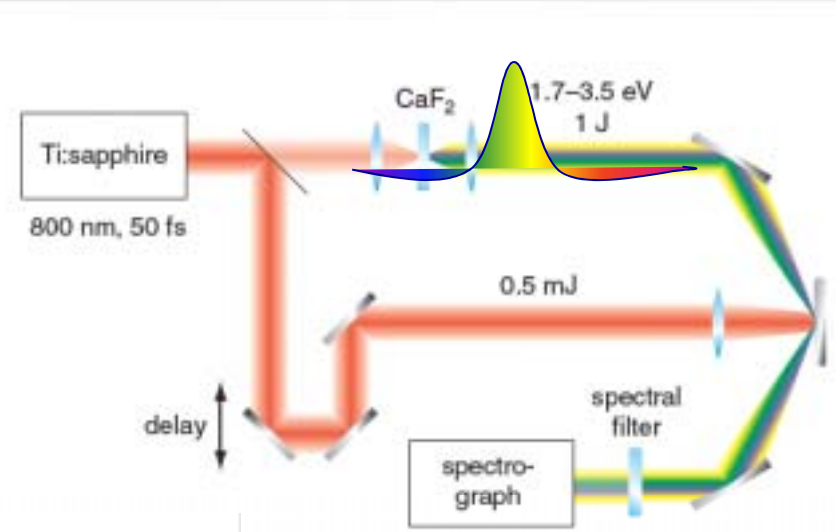


Optical dynamics: time-resolved elipsometry

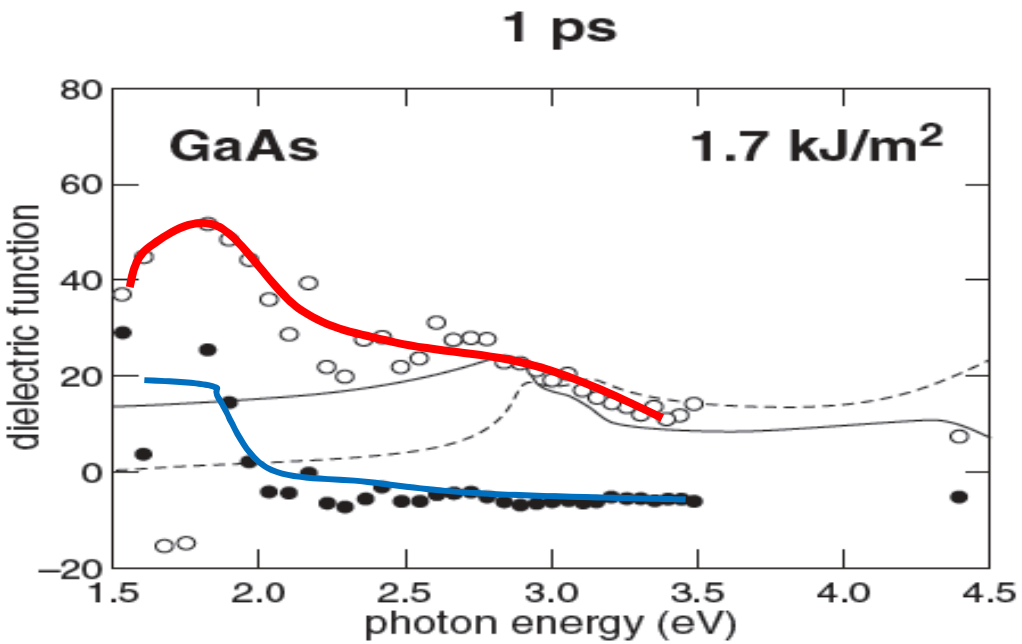
Changes in optical properties

-dielectric function

Re(ε) via Fresnel eq.
Im(ε)

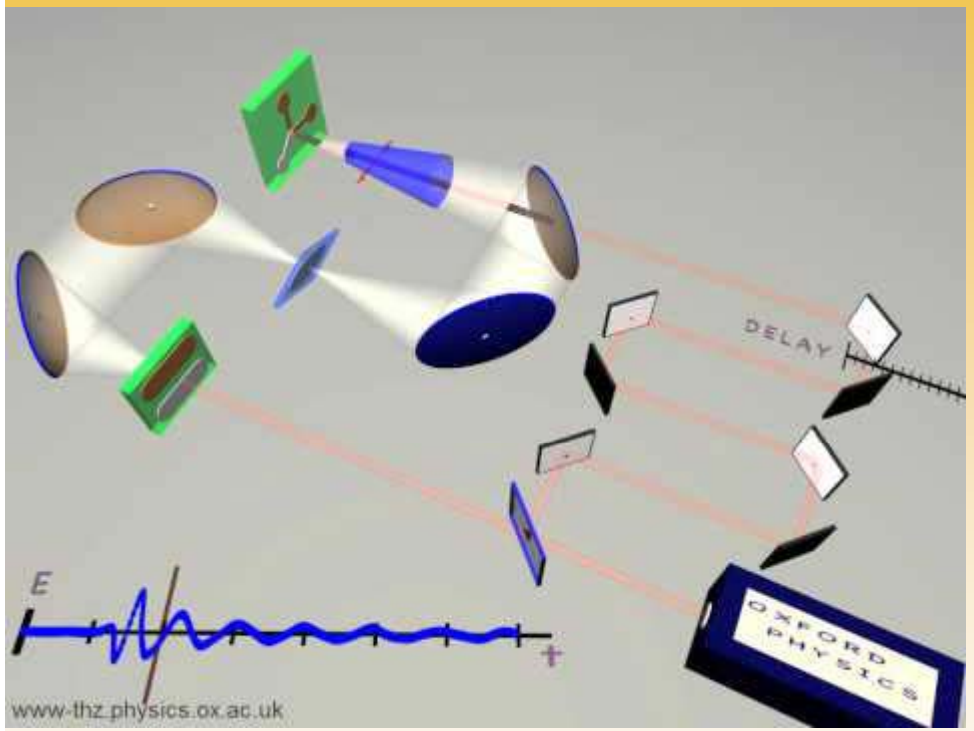


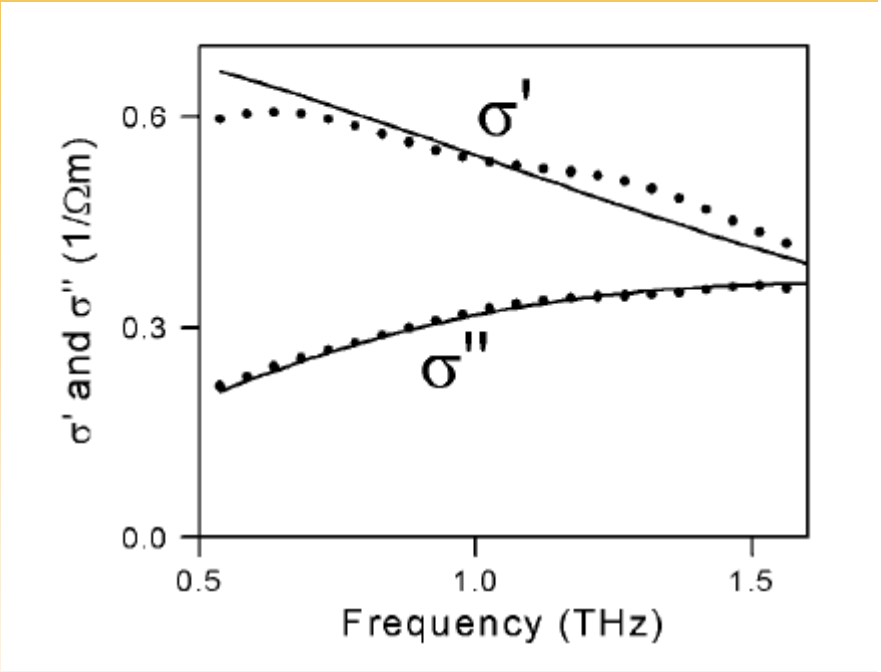
Phase transitions



SOLID TO LIQUID

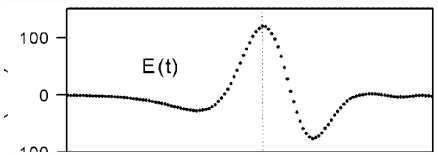
Time-domain techniques

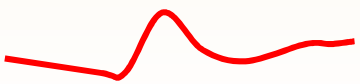




THz waves

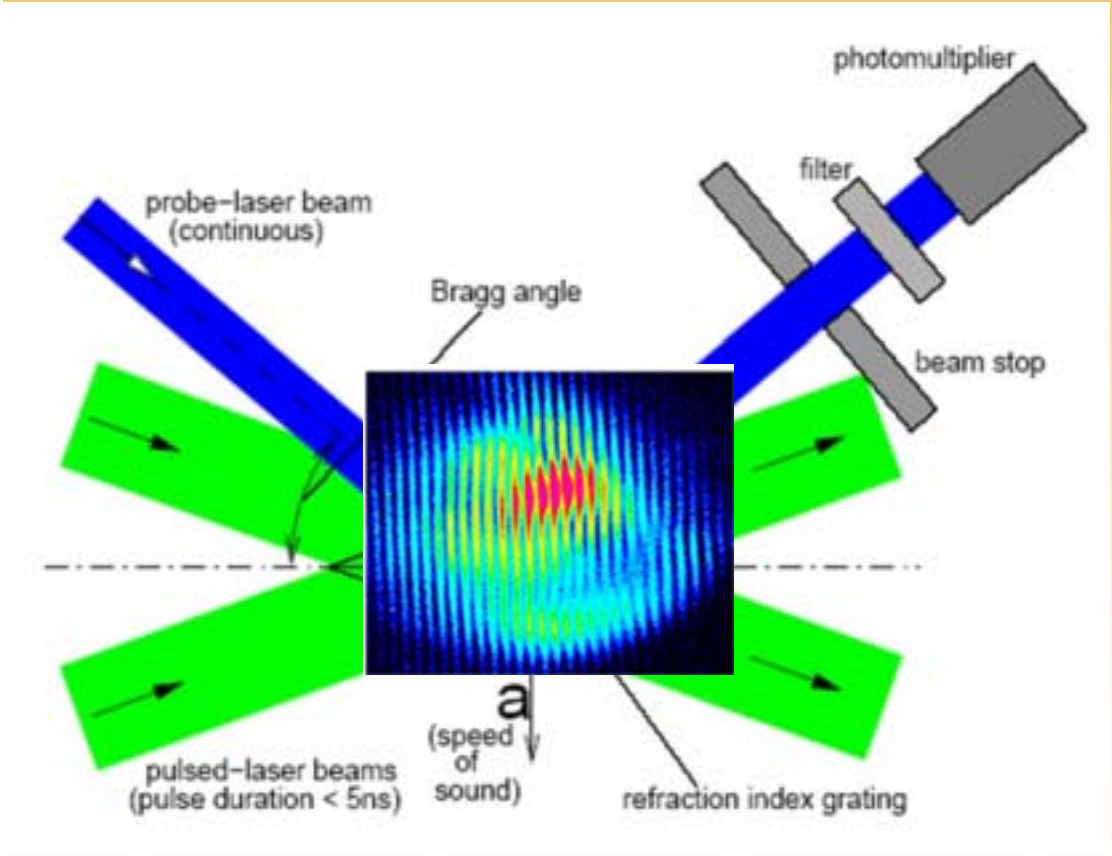
THz time-domain spectroscopy





M. Bonn, T. Heinz, MIT

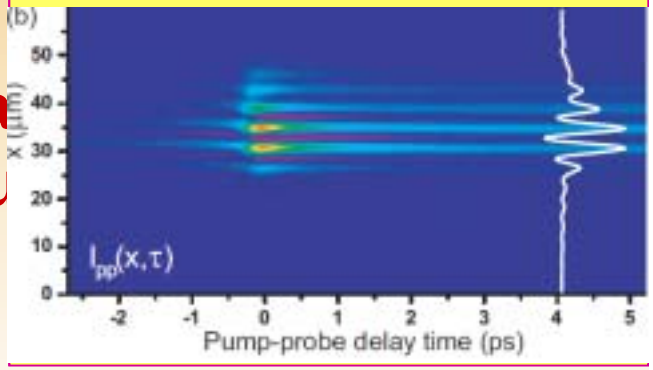
Transient gratings: ultrafast laser pulses



Similarly:

-plasma waves (conserve k)

Diffra
modu



Temnov OE 2009

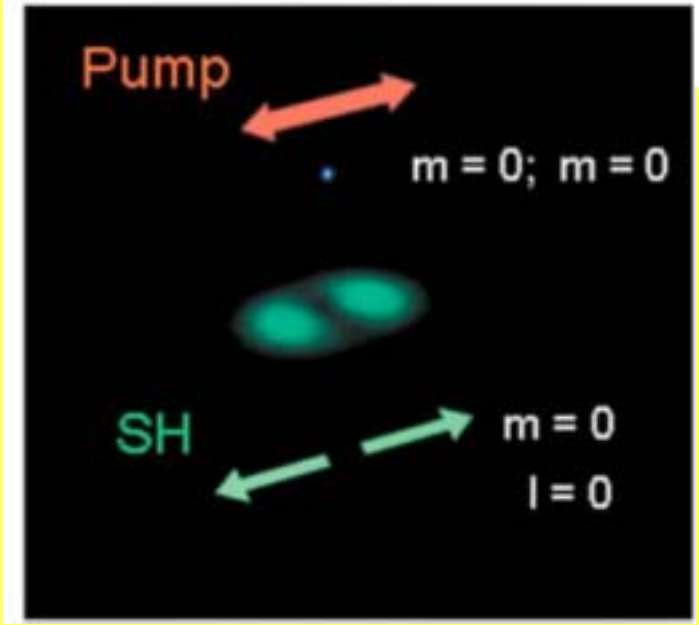
DLR, Liu APL 2011, Suntsov APL 2009

Light conversion

-Generation of frequencies and phase matching

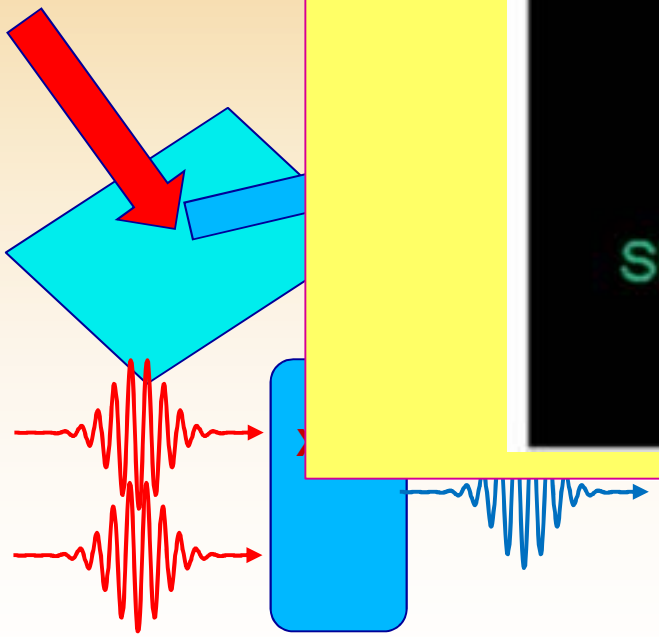
χ^2 sensitive to crystalline anisotropy

In glasses? Electronic gradients



$$\vec{P}(2\omega) = \chi_{fe}(2\omega)\left[2\nabla E^2/2 + 2\vec{E}(\vec{E}\cdot\nabla \ln n_e)/\varepsilon_p\right]$$

Beresna at al APL 2009



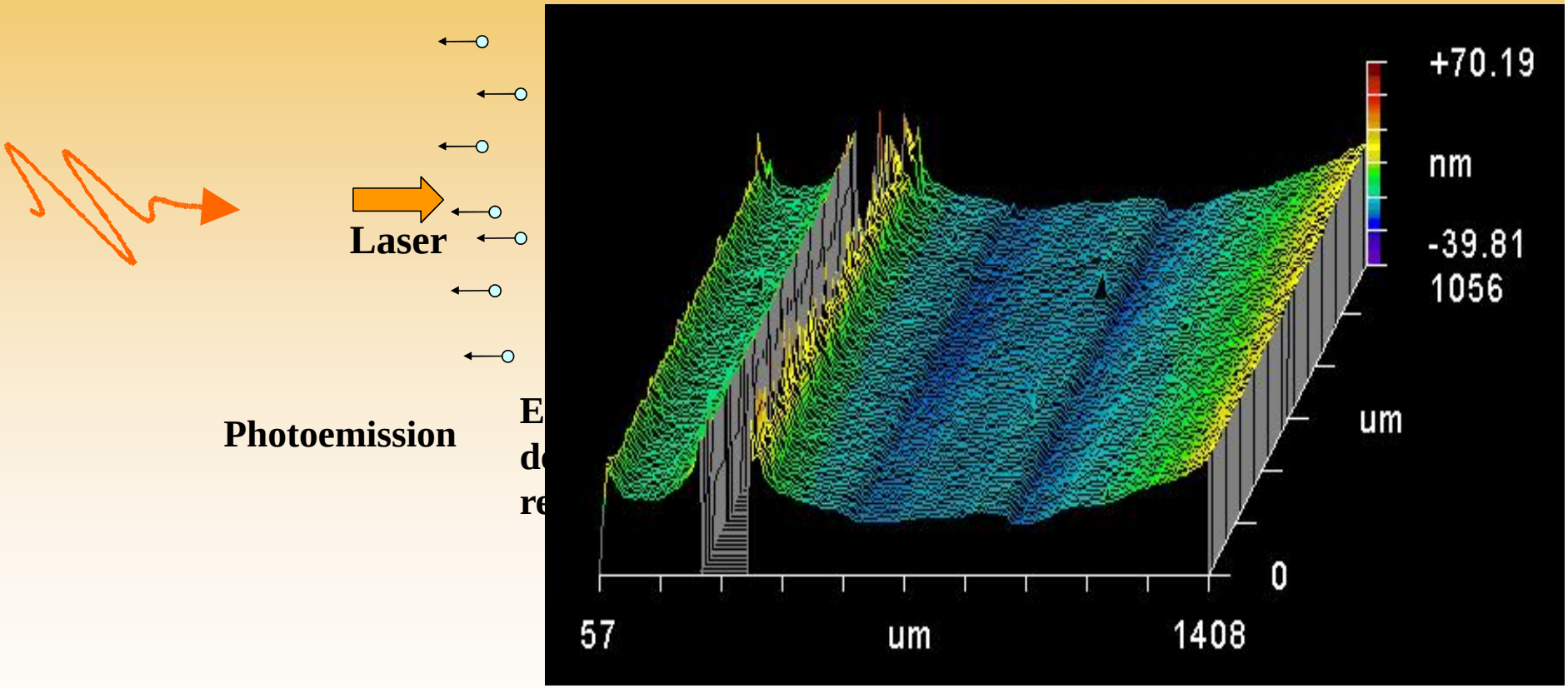
SOLID TO LIQUID

Tom et al PRL 1998

Excitation of carrier plasmas

Q3: Material ejection

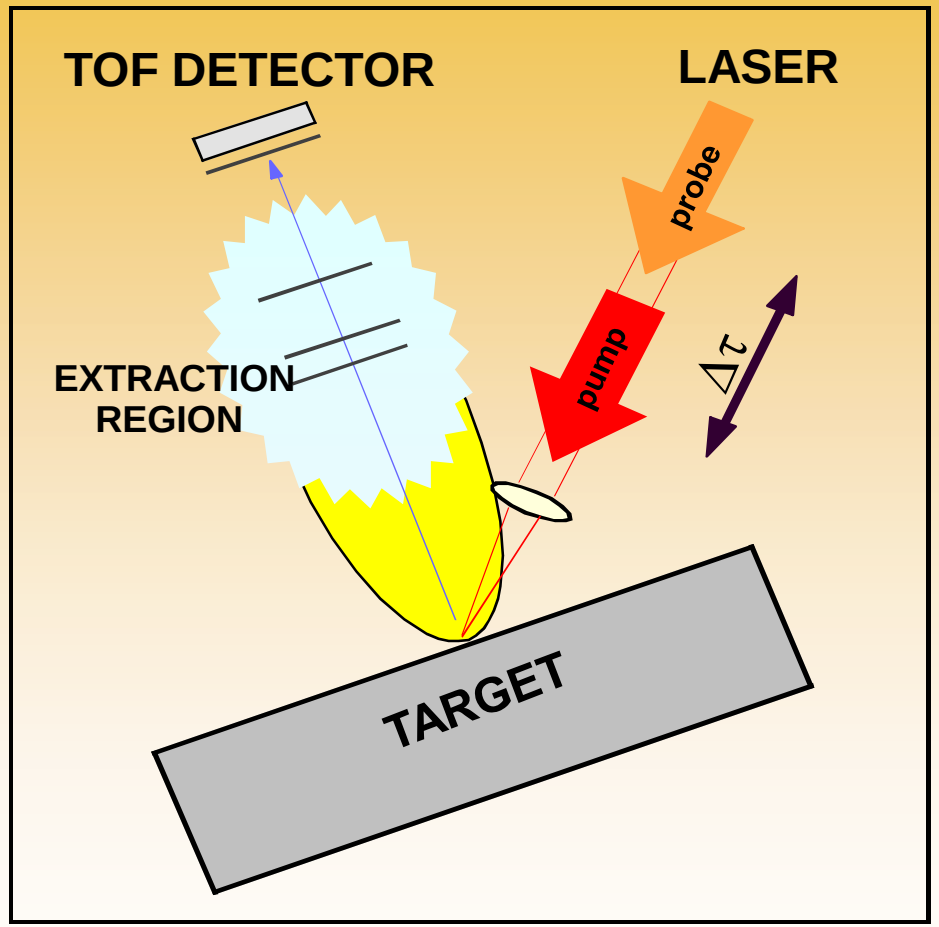
Material removal: ultrafast laser pulses



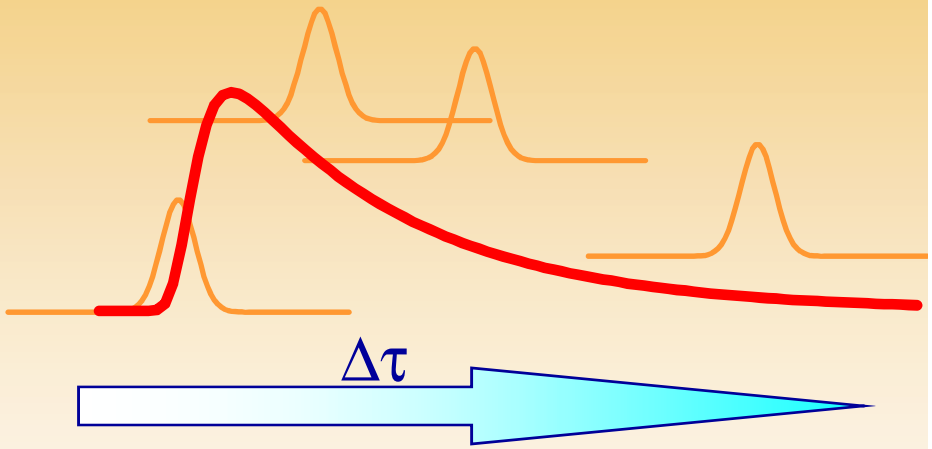
Coulomb explosion

R. Stoian PRL 2002

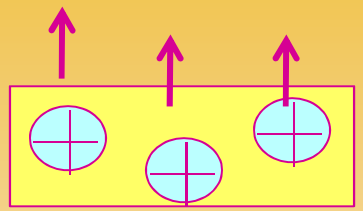
Material removal: ultrafast laser pulses



decay of excitation



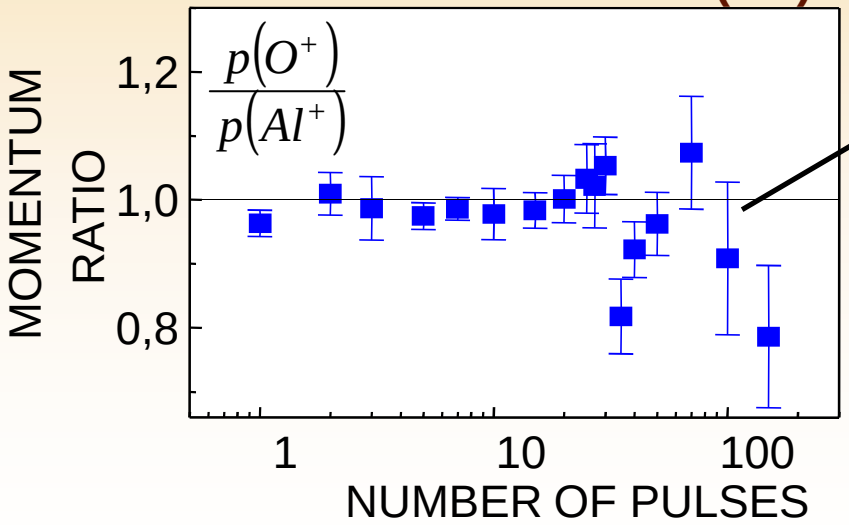
Material removal: ultrafast laser pulses

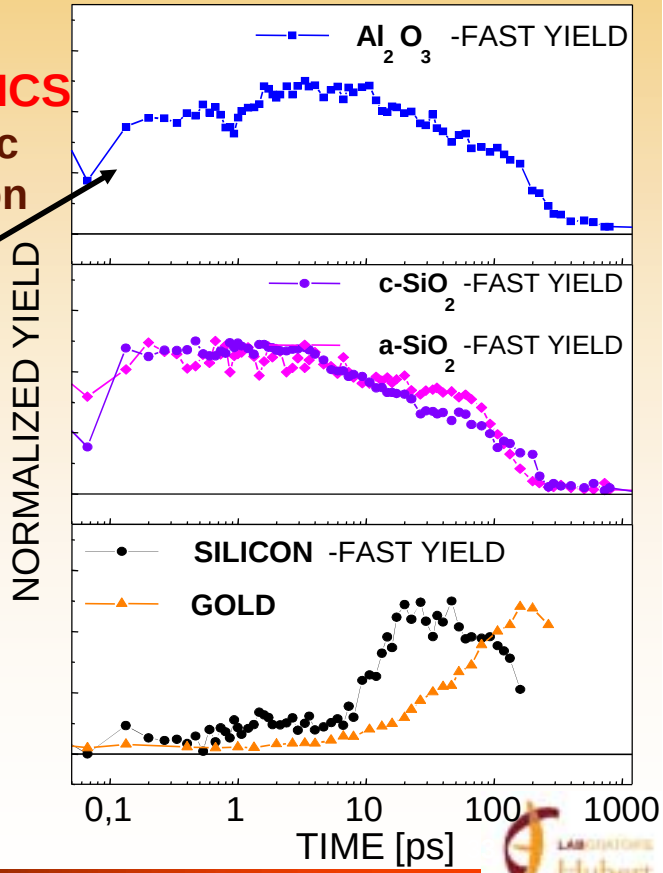


DIELECTRICS
electrostatic ion emission

METALS
thermal

COULOMB EXPLOSION (CE)





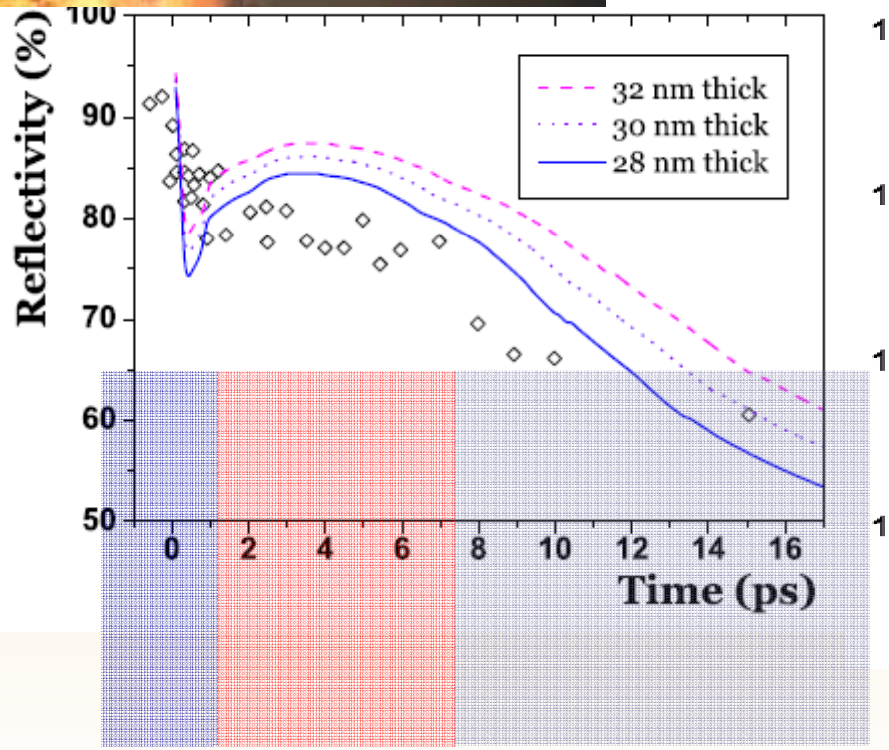
R. Stoian PRL 2002

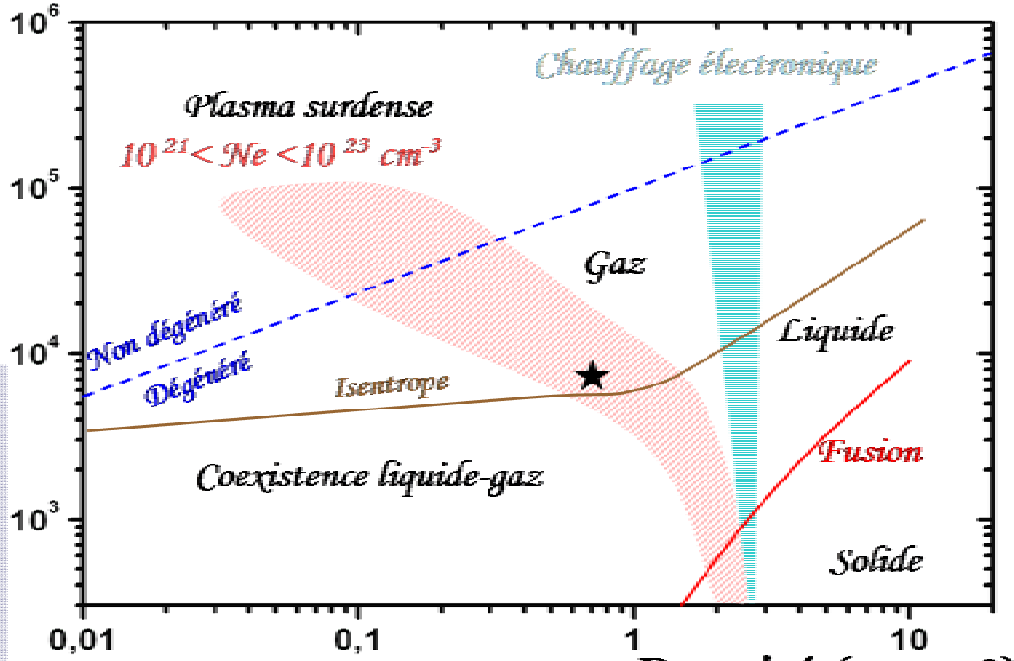
Excitation of carrier plasmas

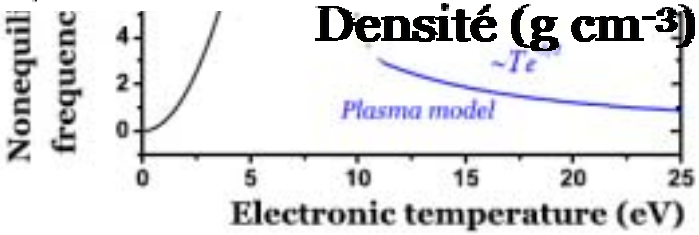
Q4: Internal energy;
Scattering, plasma radiation

Energy of plasmas: ultrafast laser pulses

Simulation codes

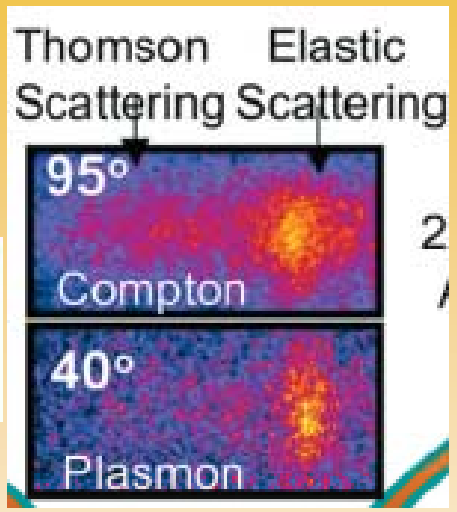


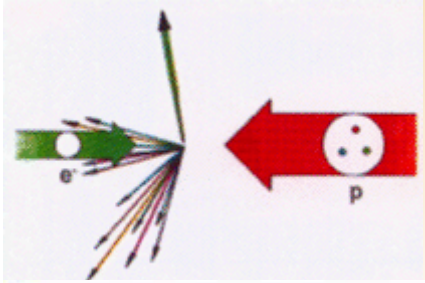




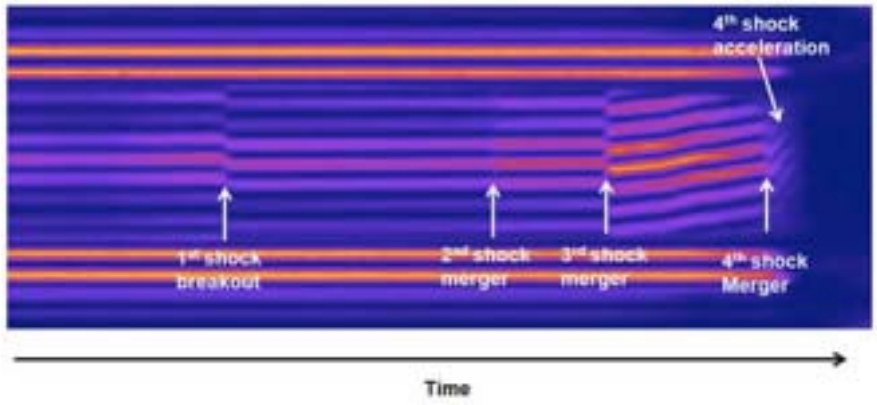
Electron heating **Lattice heating** **Hydrodynamic expansion**

Energy of plasmas: ultrafast laser pulses





Proton scattering



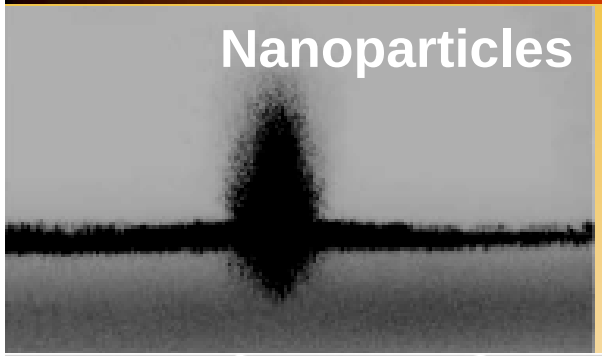
Front evolution

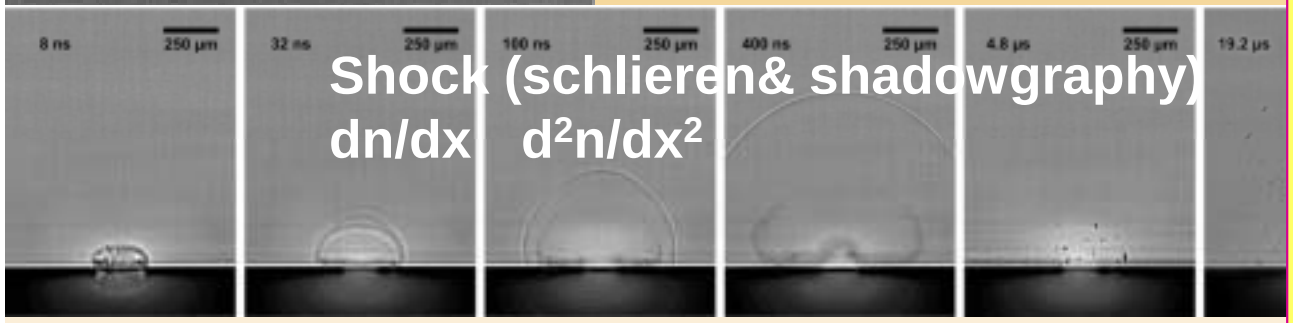
Properties:

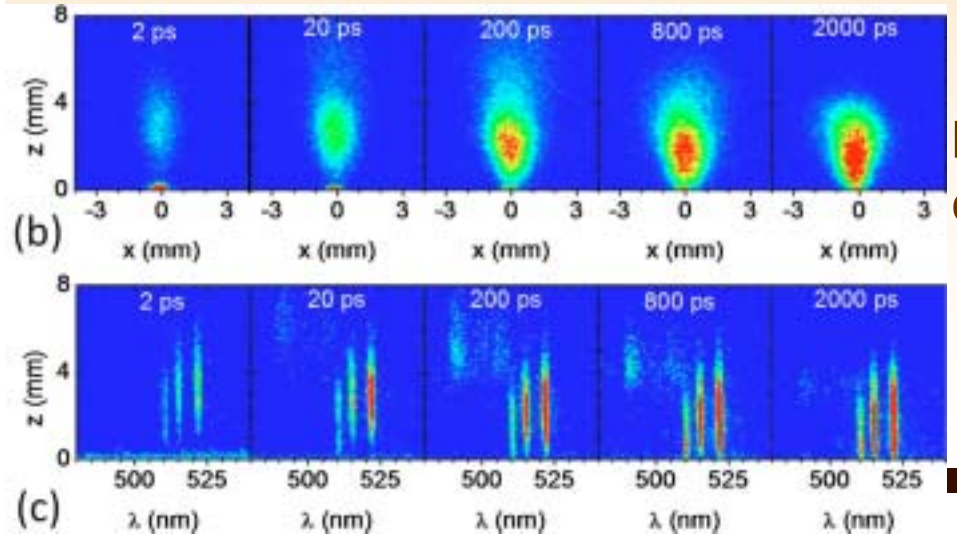
-density
-structure
-temperature

Various labs: SLAC, DESSY, Los Alamos, CEA, Berkley

Ablation pulses: ultrafast laser pulses







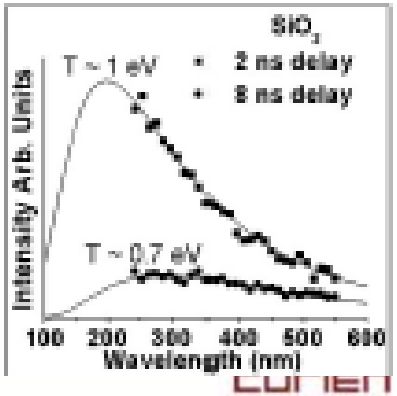
Properties:

-acoustic
-optical
-luminescence
-electrical
-structural
-waves
-ions (core level)

Luminescence and excitation temperature

$$I_\lambda \sim e^{-E/kT}$$

Amoruso APL 2009
Hermann JAP
Lippert JAP 2010
Car OL 2005

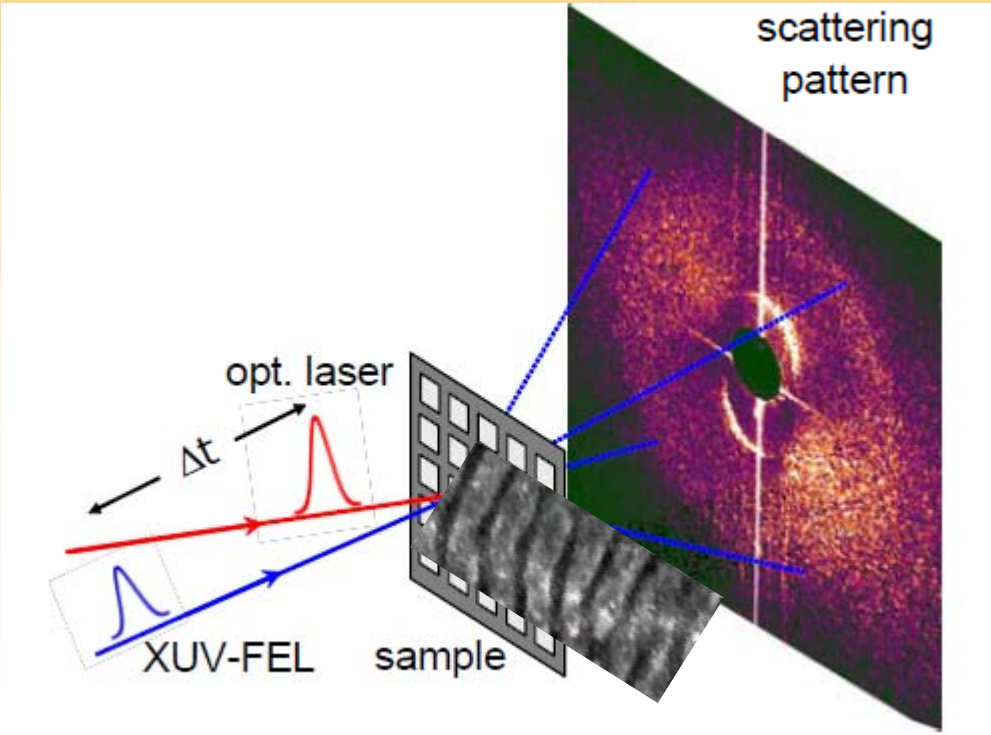


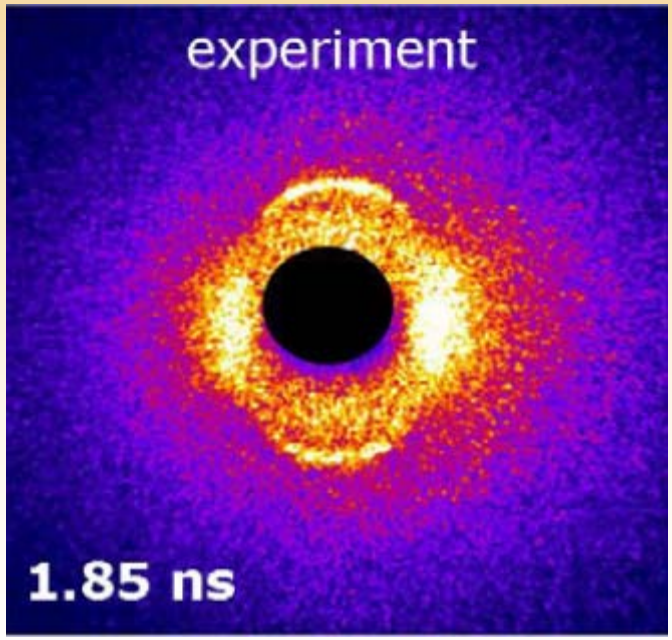
Topology transformations

Challenge: dynamics of the surface movement?

Optical methods:

-visualizing surface modulation





Time-resolved diffraction?

The interest?

What is happening with the material

-changes in the dielectric function: OPTICAL

-changes in the structure: THERMODYNAMIC

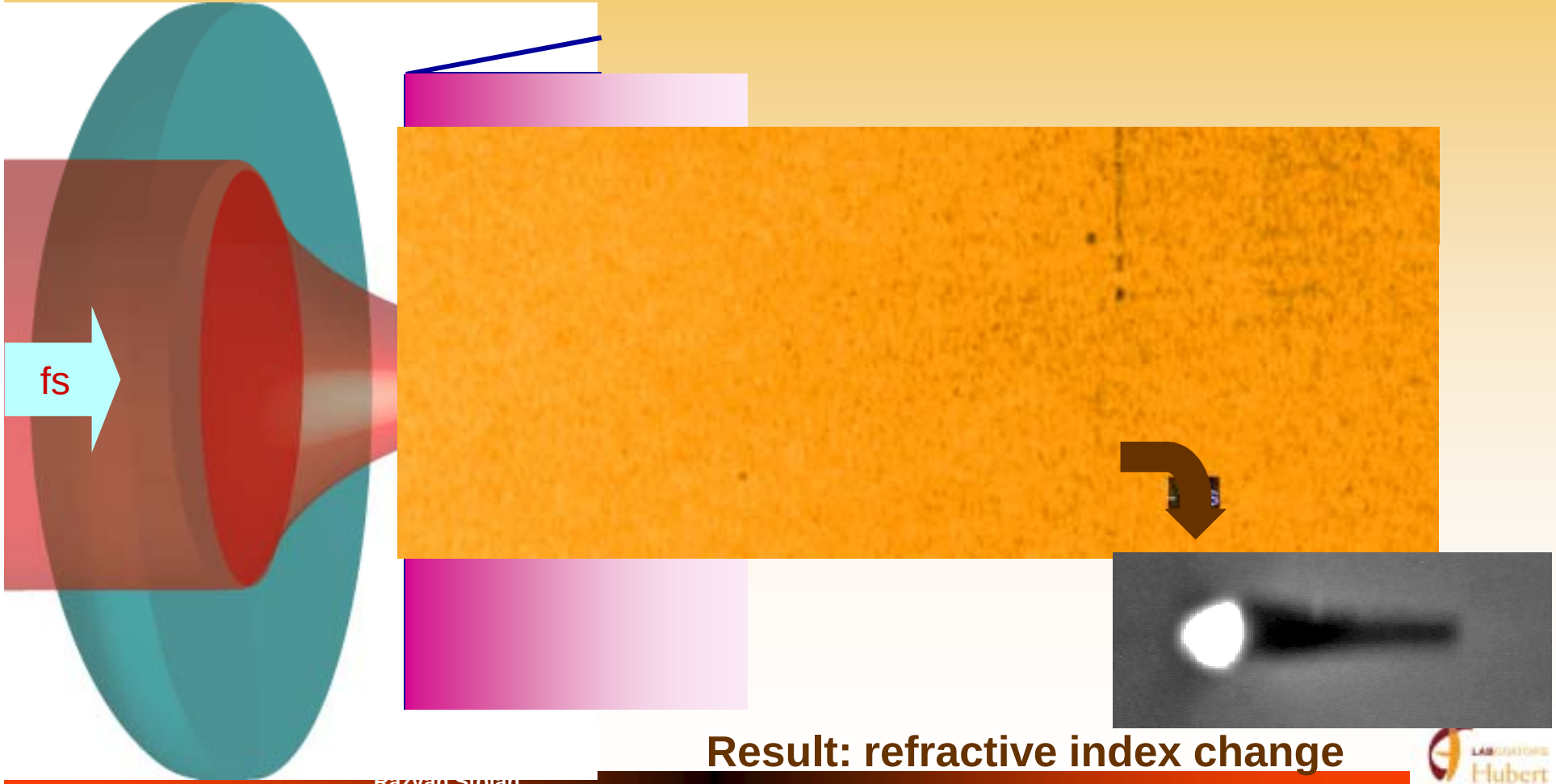
-changes in the shape: FUNCTION

Monitoring and control of carrier plasmas

Q5: Carrier dynamics

Ex: Application to 3D structuring

3D nonlinear excitation: ultrafast laser pulses

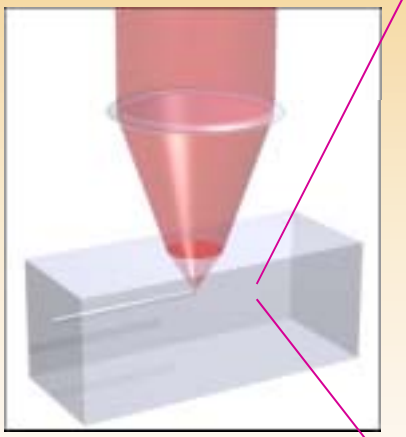


Result: refractive index change

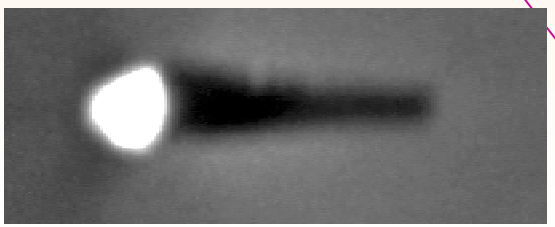
3D material modifications

refractive index Δn

- Building block of embedded optical functions



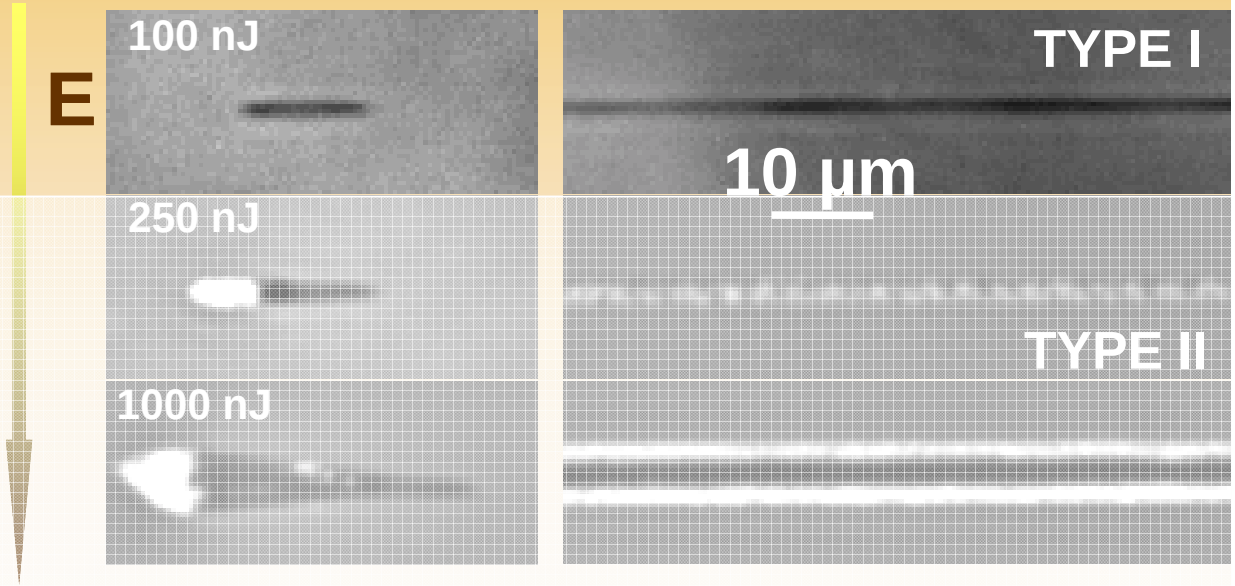
Light guiding
Davis et al OL 1996



Energy density regimes: a-SiO_2

Phase contrast

NA: 0.45 - 150 fs



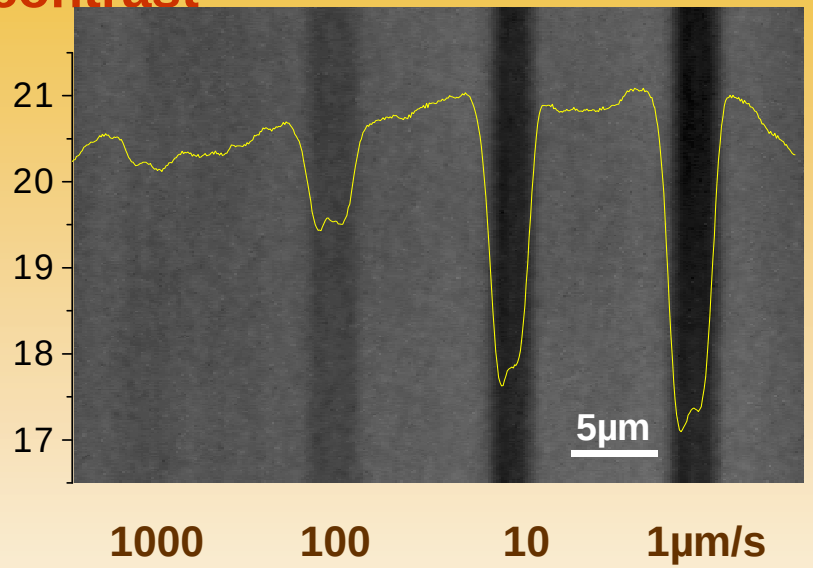
Black Δn>0 enables guiding - core

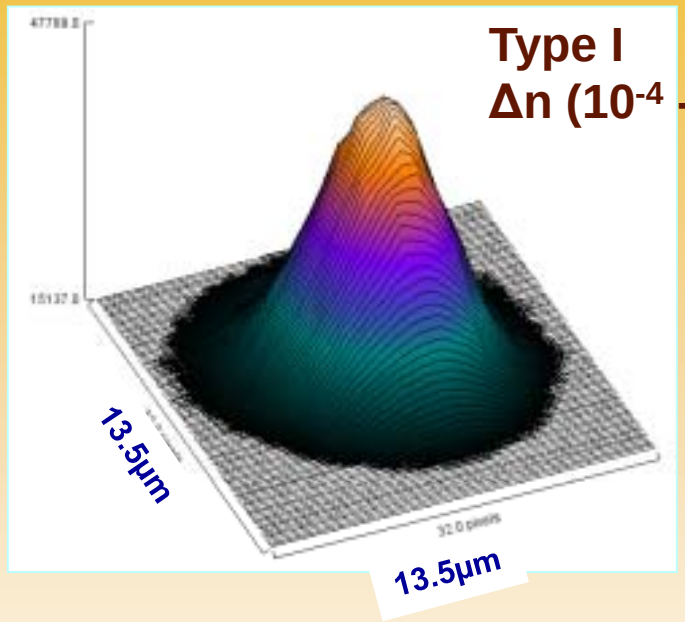
White Δn<0 defines guiding -cladding

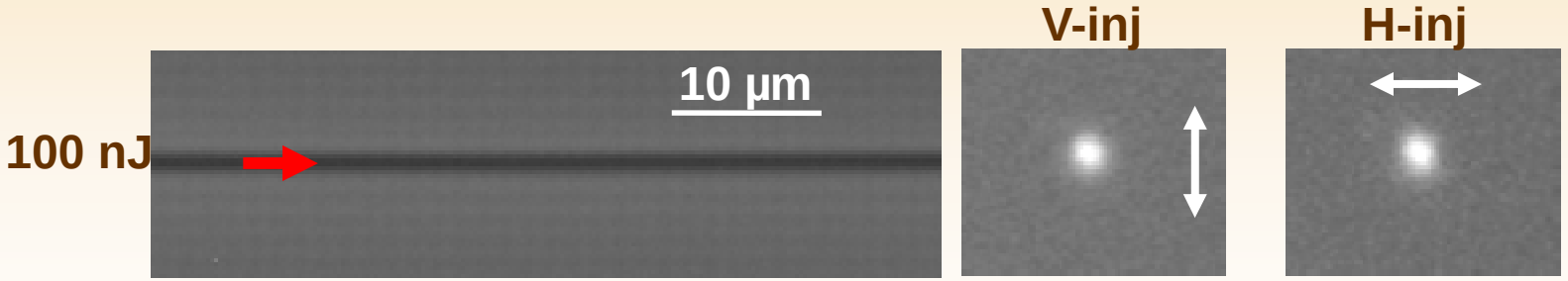
Type I regime: a-SiO_2

Optical functions

Phase contrast

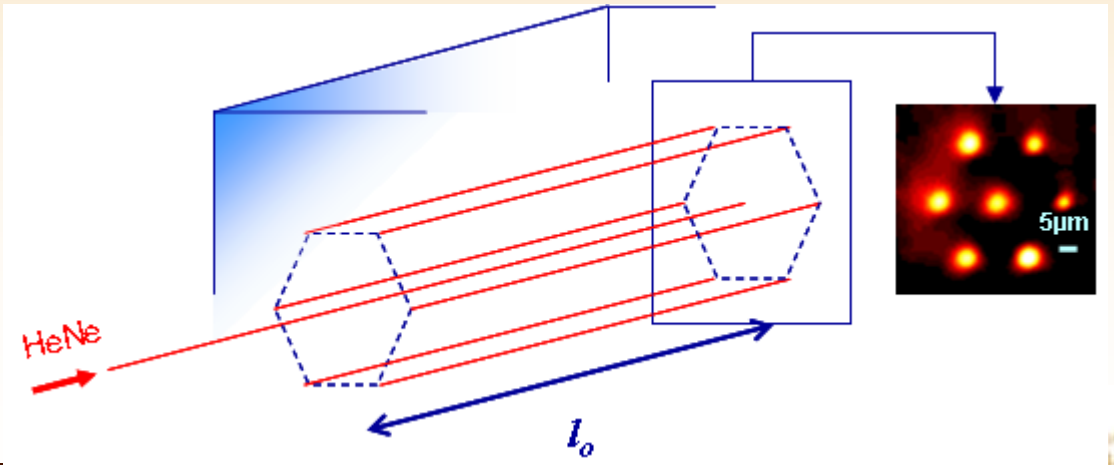






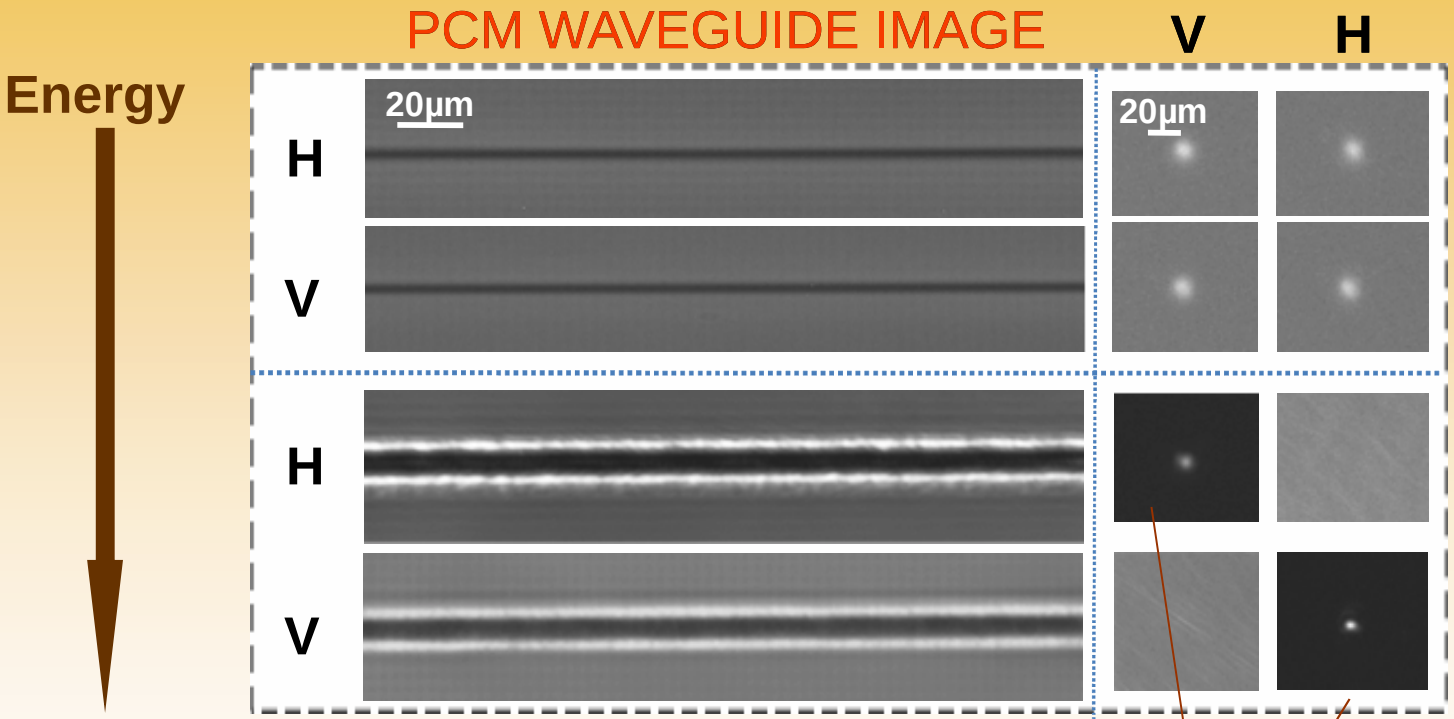
-NO STRONG POLARIZATION SENSITIVITY
-ISOTROPIC OPTICAL GUIDING
-LOW LOSSES <0.5dB/cm

3D photonic structure

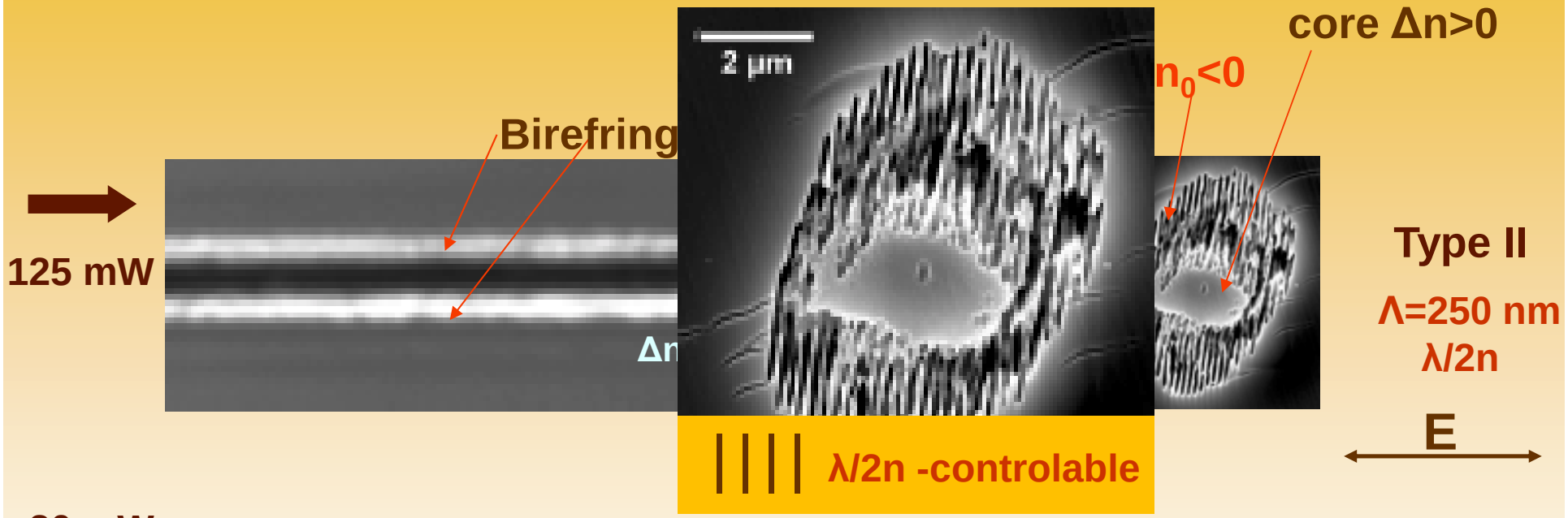


Mauclair et al. OE 2009

Role of polarization: a-SiO_2



Anisotropic regimes: a-SiO_2



Shimotsuma PRL 2003
Bhardwaj PRL 2006

Guiding when E is parallel to the planes

Cladding: form birefringence

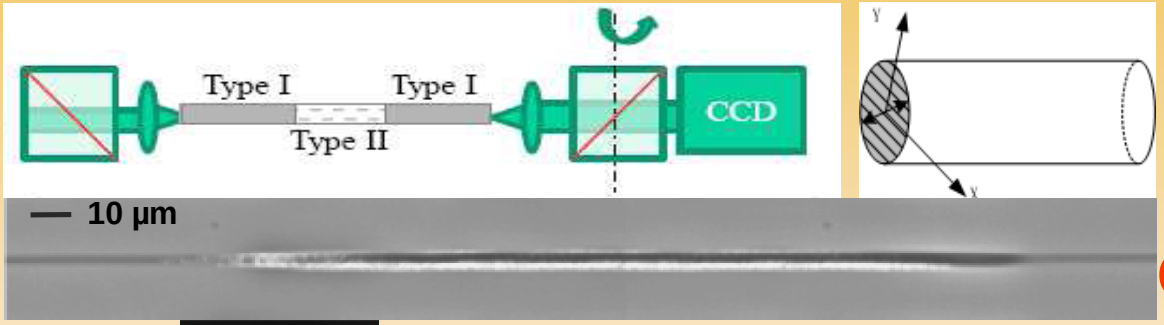
a-SiO_2

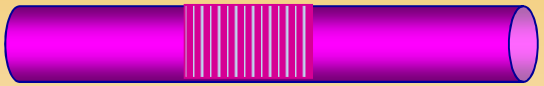
Polarization function

Cheng et al. OE 2009, 2010

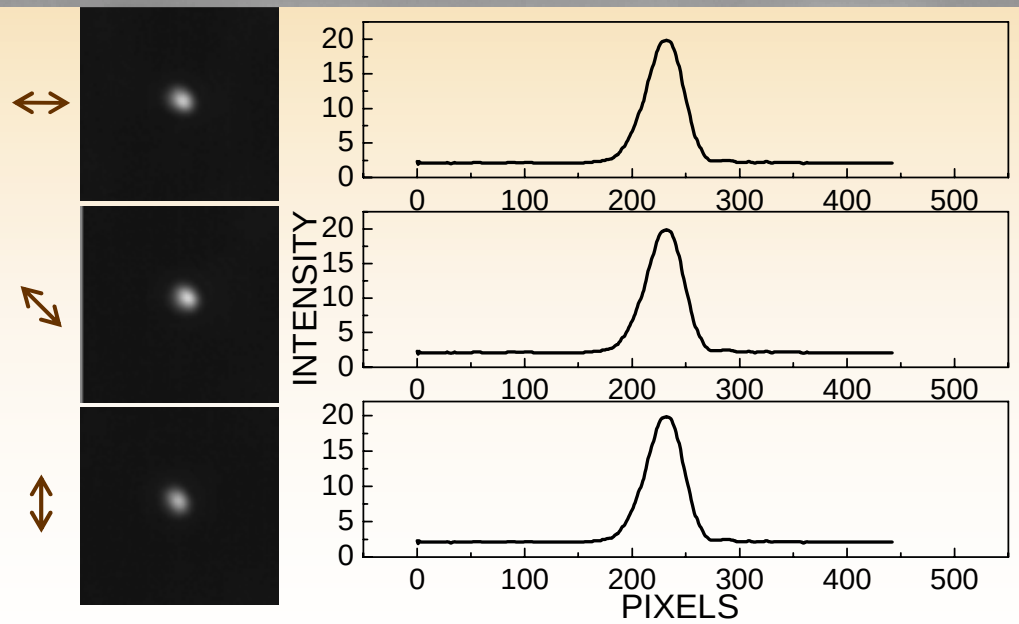
Optical functions: a-SiO_2

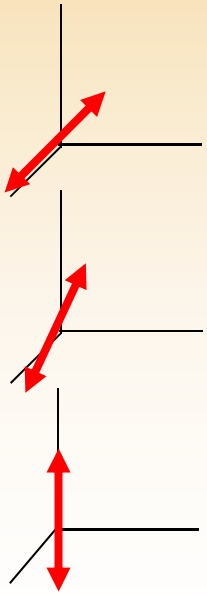
- Polarization maintaining waveguides
- Birefringent phase retardation properties





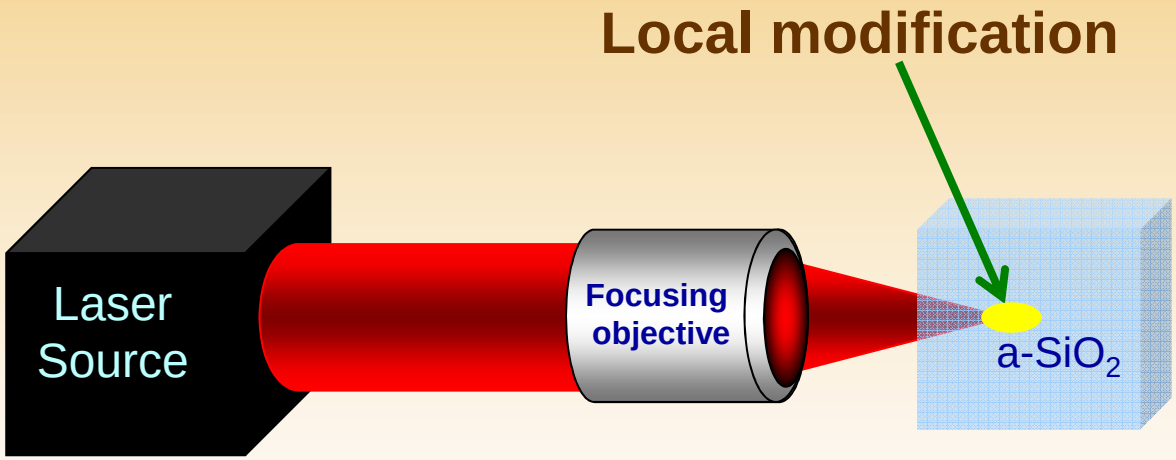
Quarter Wave Plate





Mishchik et al. OE 2010

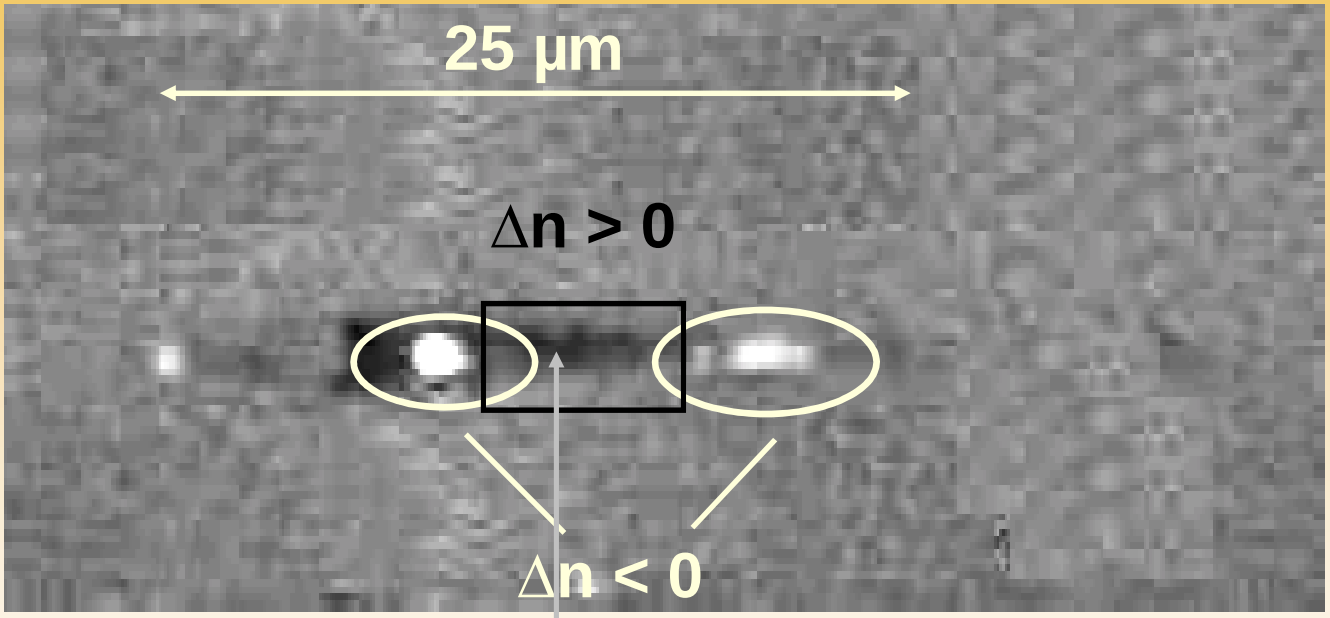
Refractive index changes



Can this be improved (controlled)?

Refractive index changes a-SiO_2

SINGLE PULSE EFFECT (N=1): a-SiO_2



PHASE-CONTRAST MICROSCOPY

FOCUS (NA= 0.4), 1µJ

Q1: How is the energy distributed?

Q2: How does the material react?

Refractive index changes

Q: How is the energy deposited?

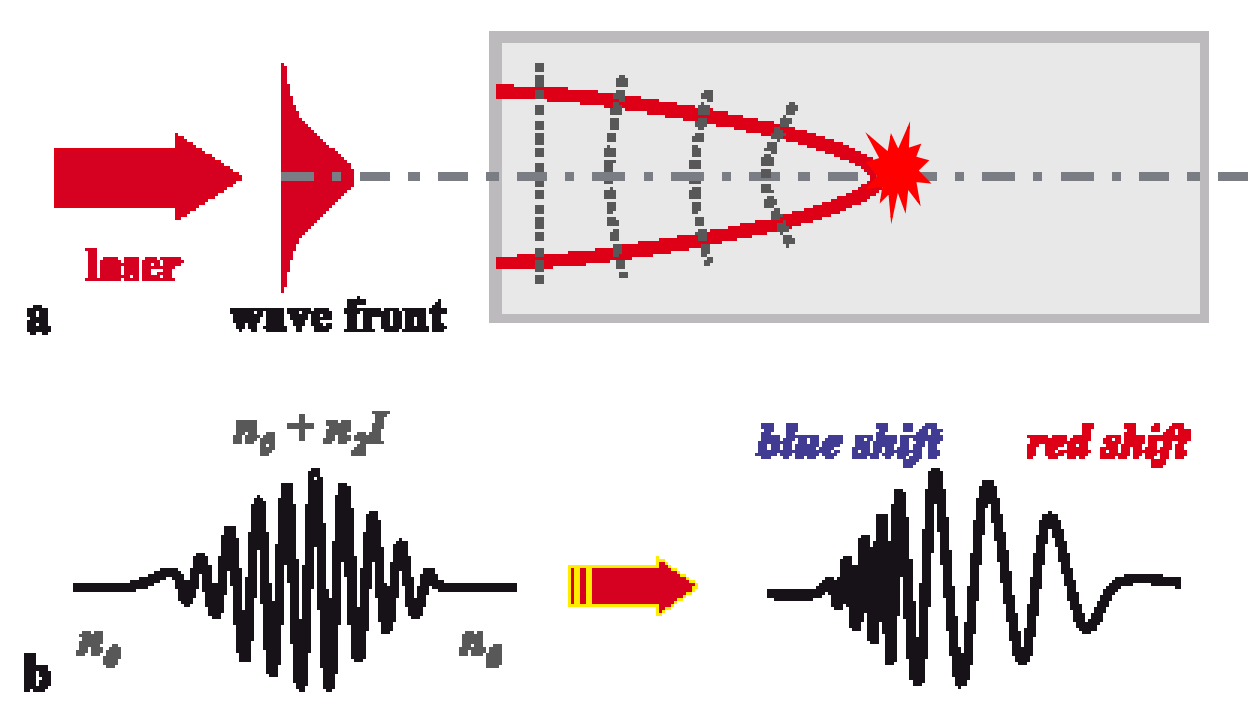
Nonlinear pulse propagation

Collaboration:
IT, Novosibirsk

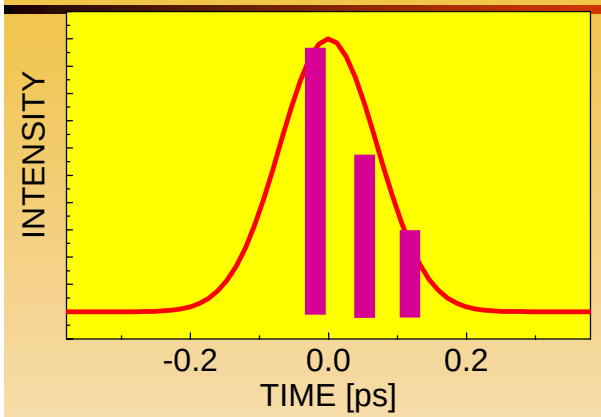
NLSE-Schrödinger

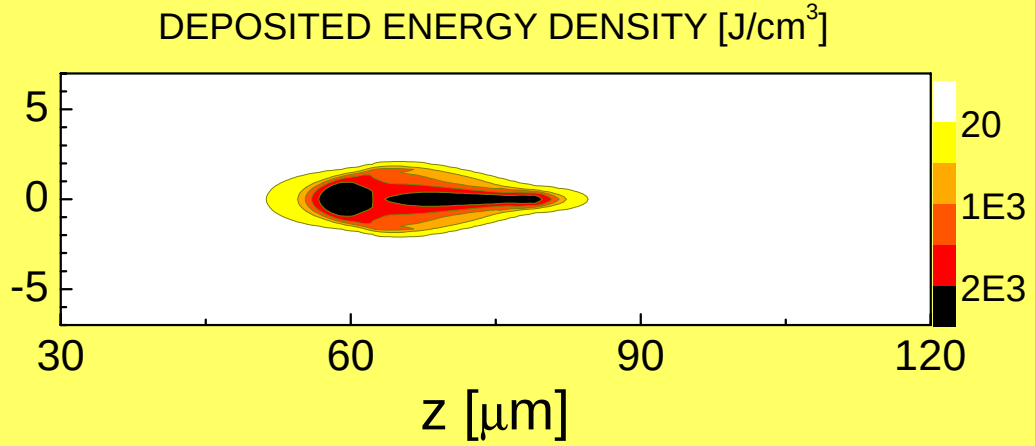
E-field propagation

-self-focusing and self-phase modulation
-plasma generation
-filamentation

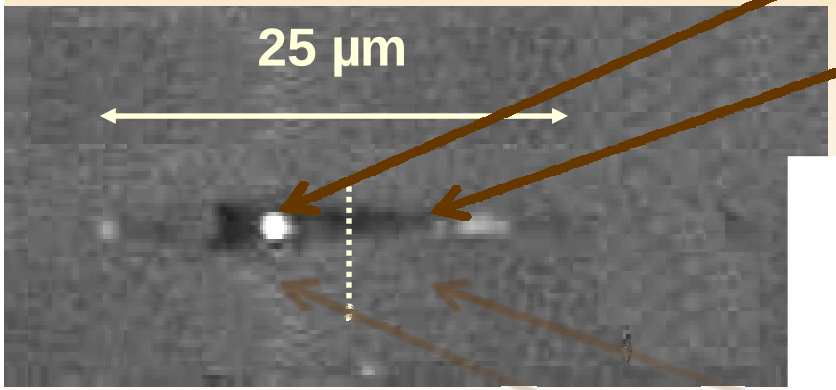


Pulse evolution: sequential energy deposition



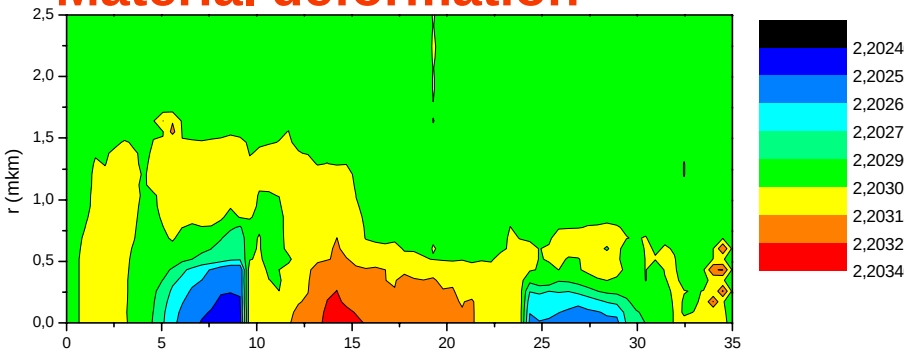


Thermo-elasto plastic model (dynamic elasticity)



Accumulation of energy (expansion & rarefaction)

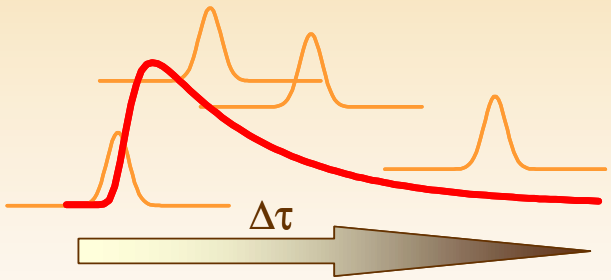
Material deformation



Densification

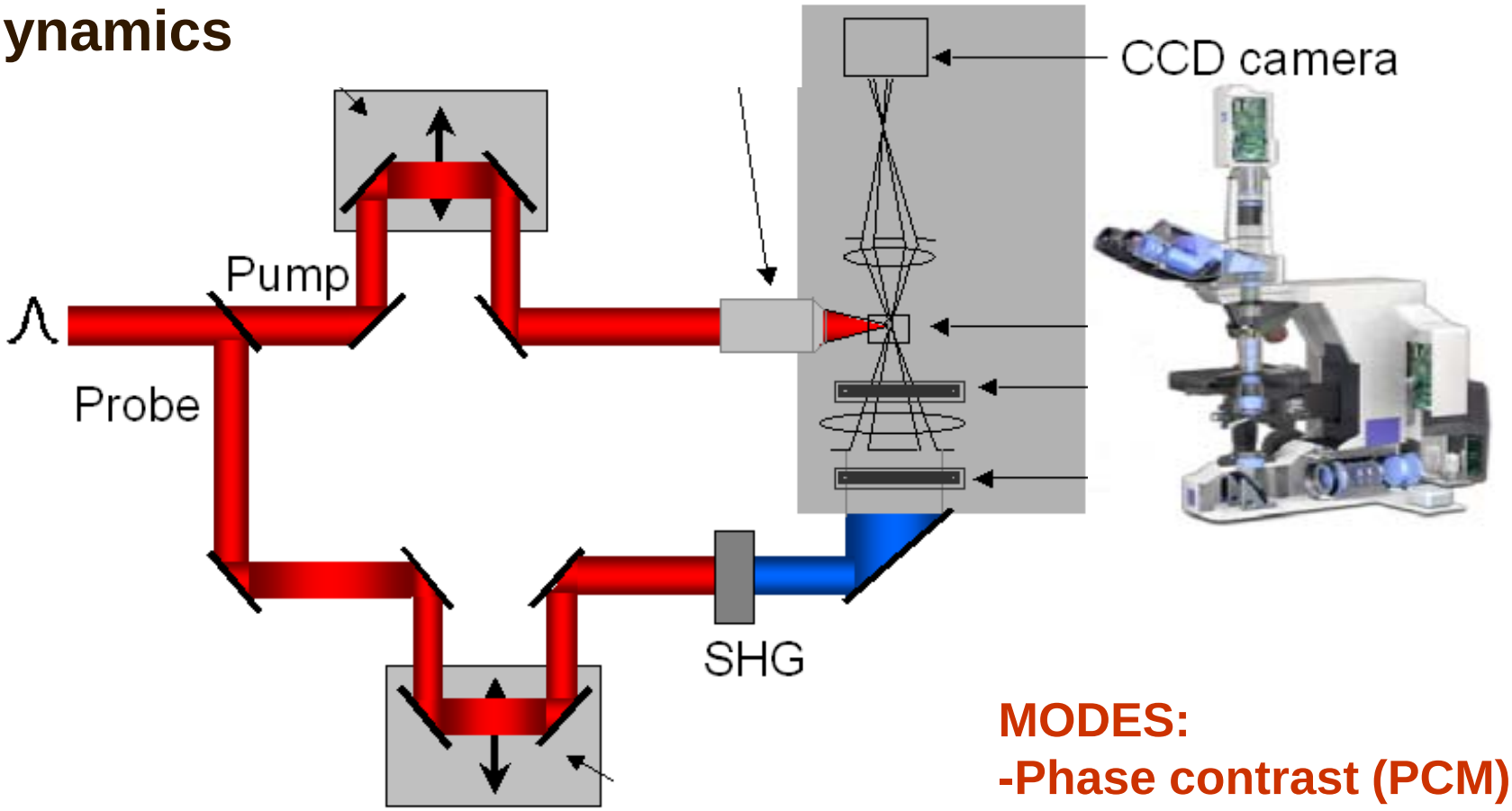
Refractive index changes a-SiO_2

Q2: how does the material react?



Pump-probe apparatus: time-resolved microscopy

Excitation and relaxation dynamics



MODES:
-Phase contrast (PCM)
-Optical transmission (OTM)

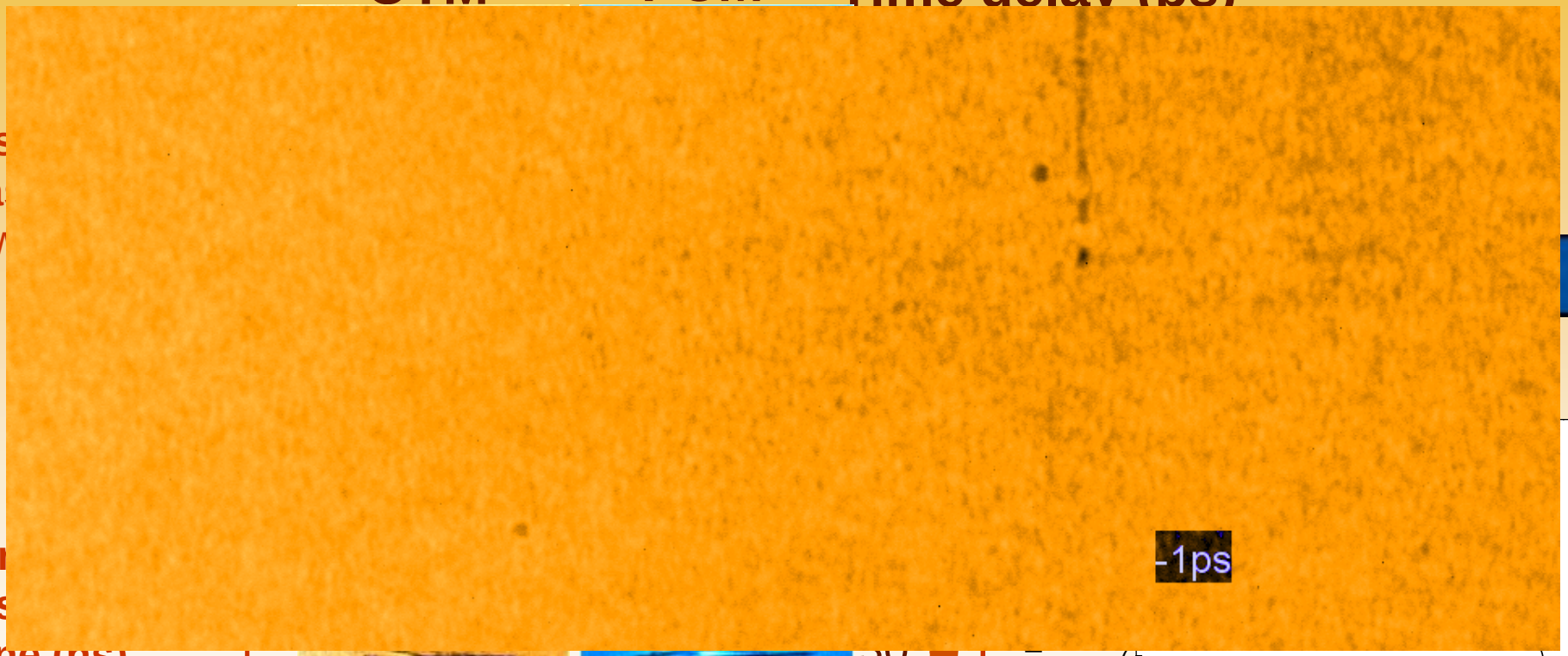
Refractive index changes a-SiO_2: time-sequence

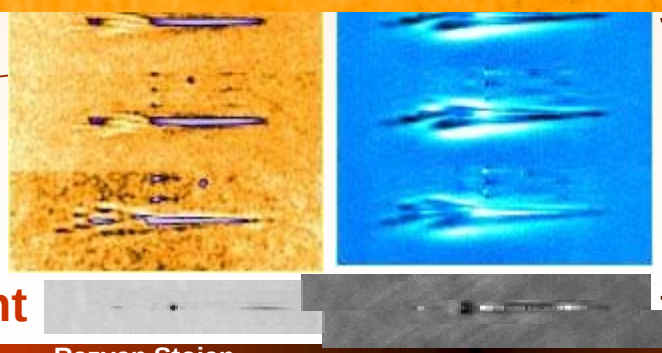
OTM PCM Time delay (ps)

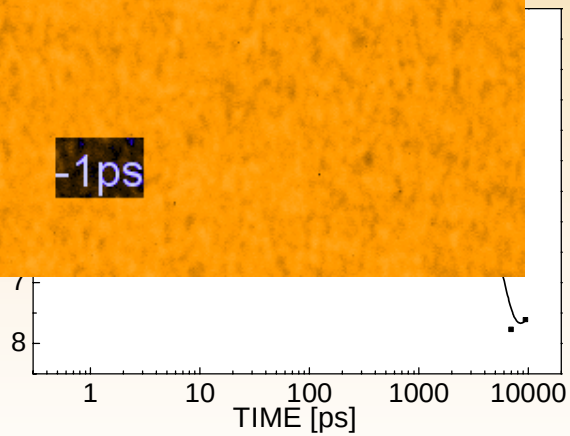
Fas
pla
(low

Per
abs
zone (ns)
-electrons?
-liquid phase?
-defects?

But not permanent



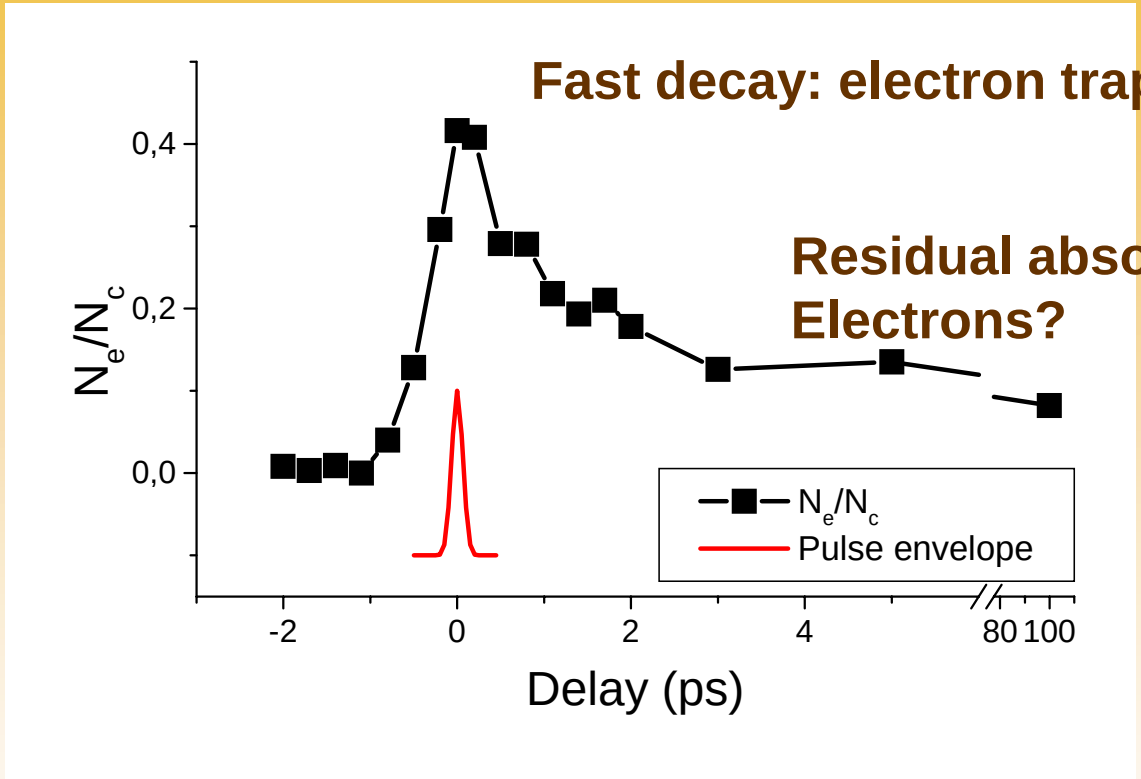




Matrix response

Thermo-mechanics

Refractive index changes a-SiO_2: electrons

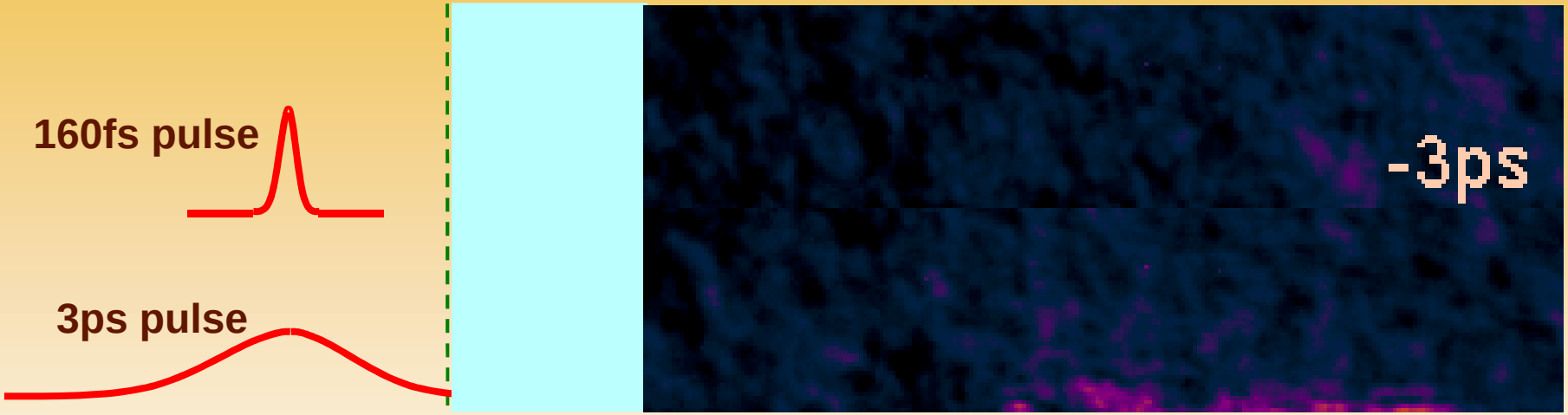


-Residual absorption can be linked to defects

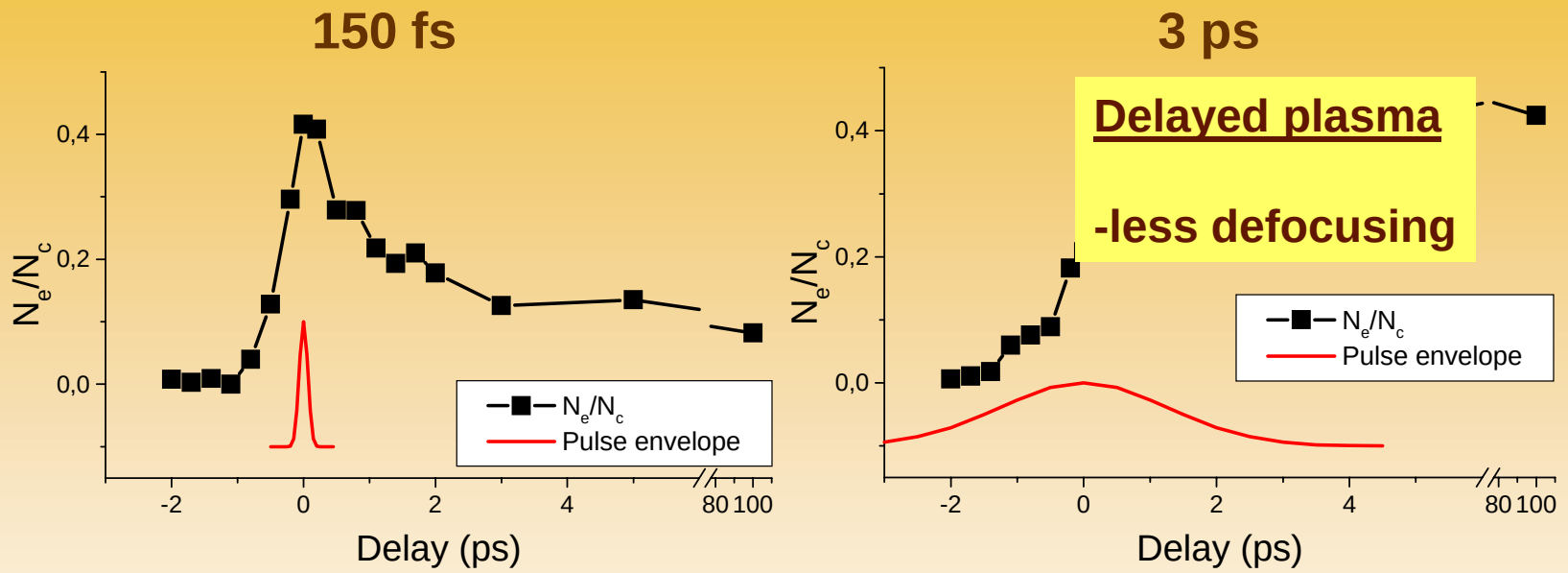
-But also to free electrons in liquid phase where trapping is not efficient (indication of a « phase transition»)

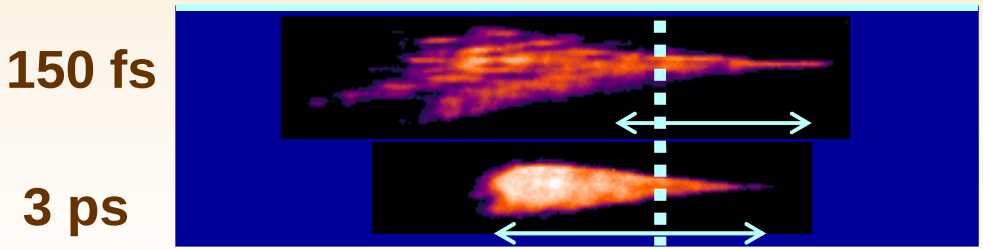
Refractive index changes a-SiO_2: time-sequence

Comparison short & long pulses 6.2µJ, NA=0.45



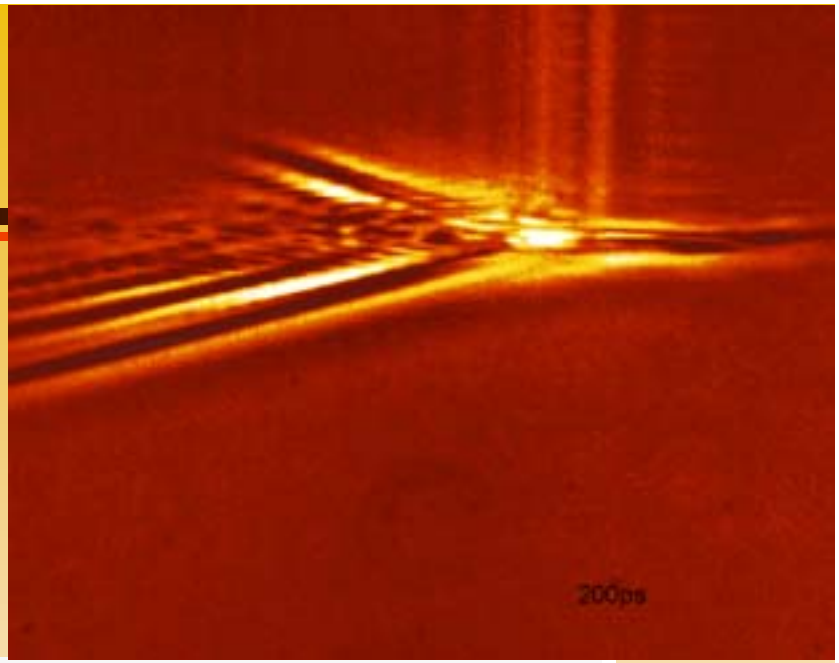
Refractive index changes a-SiO_2: electrons





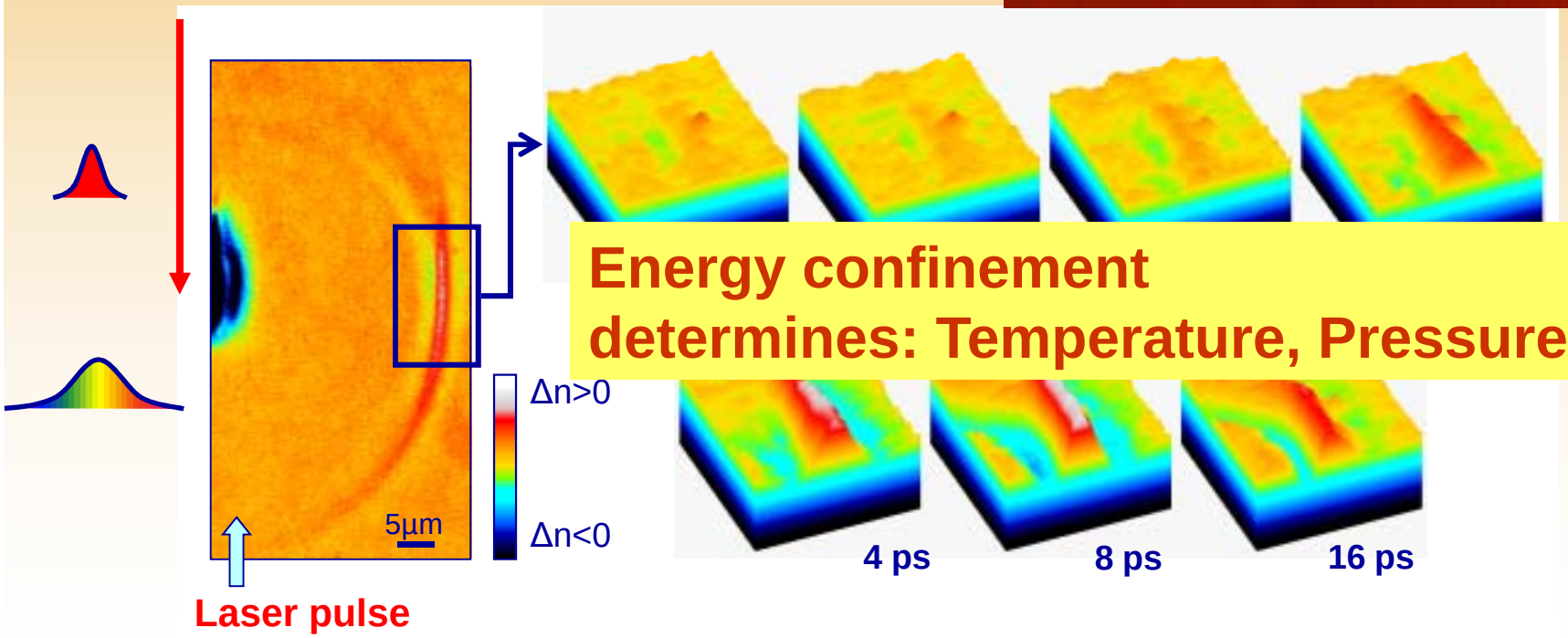
Better absorption and confinement

Energy confinement
role of pulse duration



Comparison fs/ps, single pulse irradiation

- **Pressure wave**
- ps pulse – Stronger amplitude of the PW

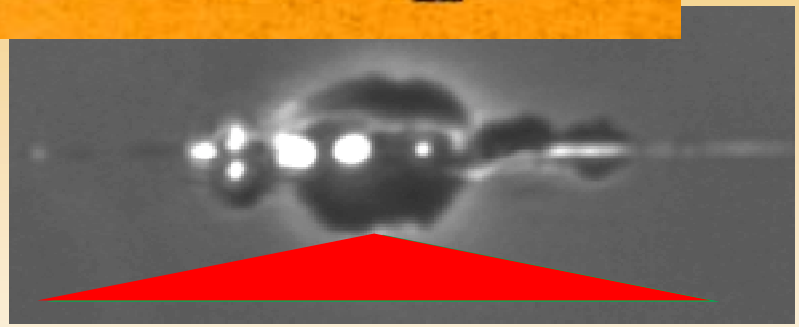


Energy confinement determines: Temperature, Pressure

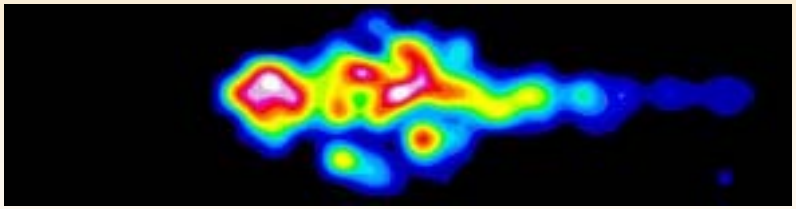
Electronic transitions a-SiO_2: spectra

N=1

a) Electron plasma at peak

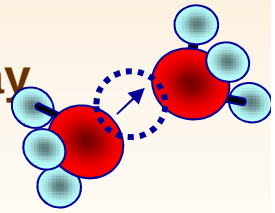


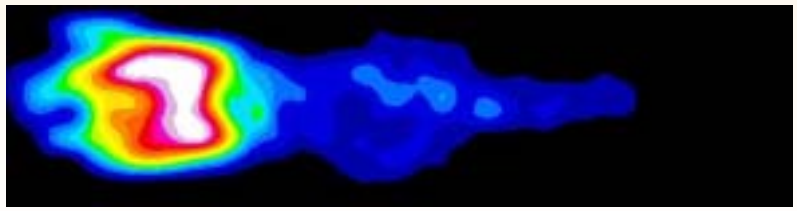
b) Permanent damage



ODC

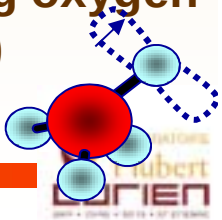
c) Oxygen defficiency centers
Slow el. decay

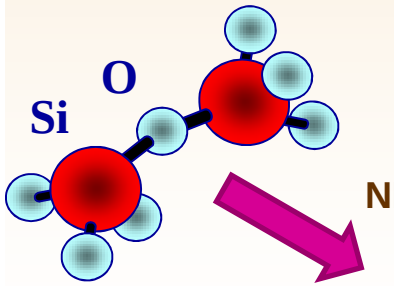




NBOHC

d) Non-bridging oxygen (Fast el. decay)



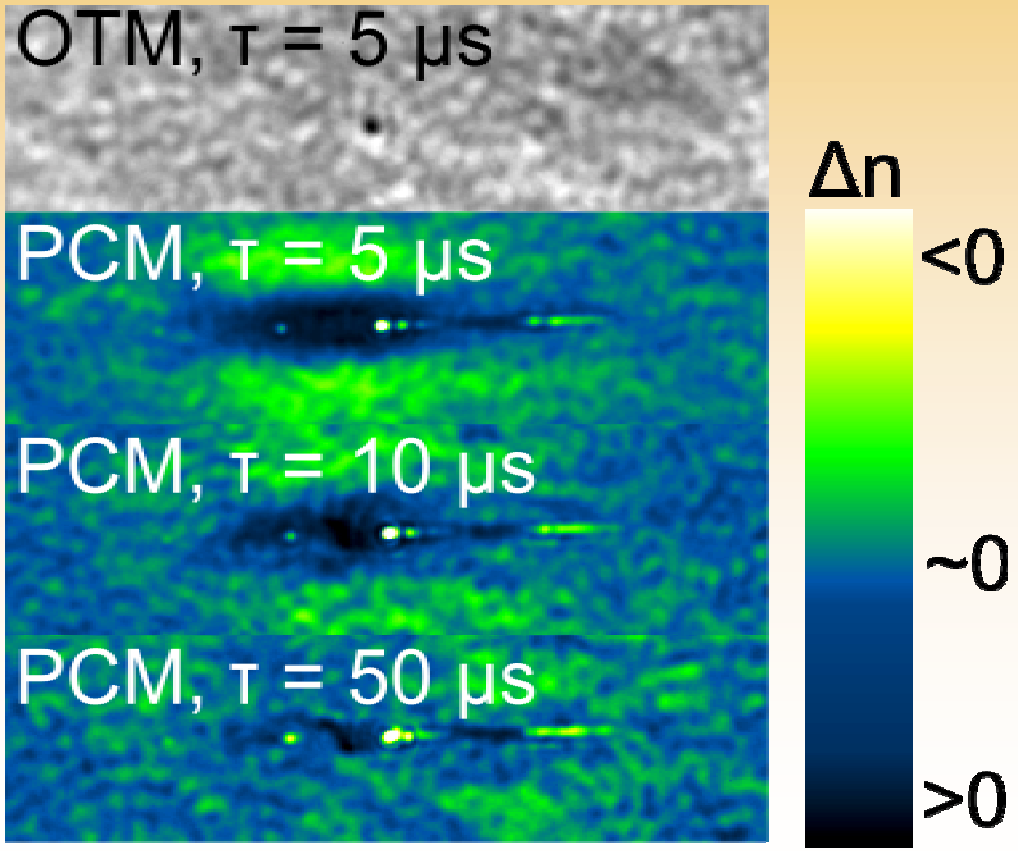


Refractive index changes a-SiO_2: stress

Slow relaxation

Slow Δn Dynamics

-stress relaxation



Mermillod-Blondin et al. RSI 2011

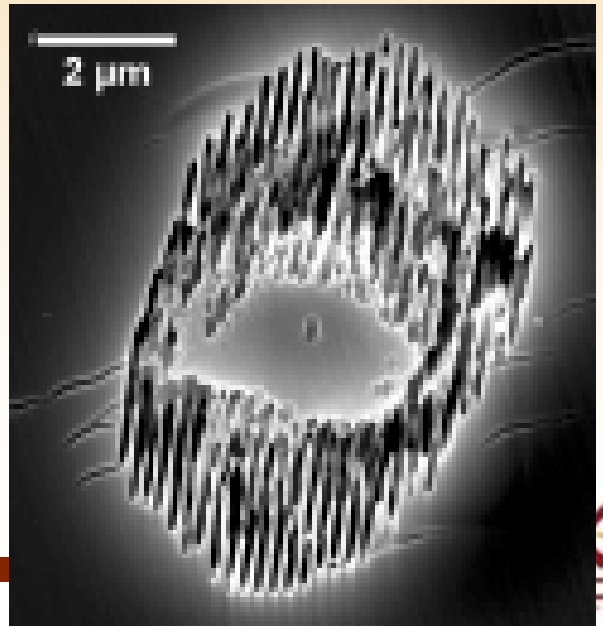
Refractive index changes

Q5: the energy density is important

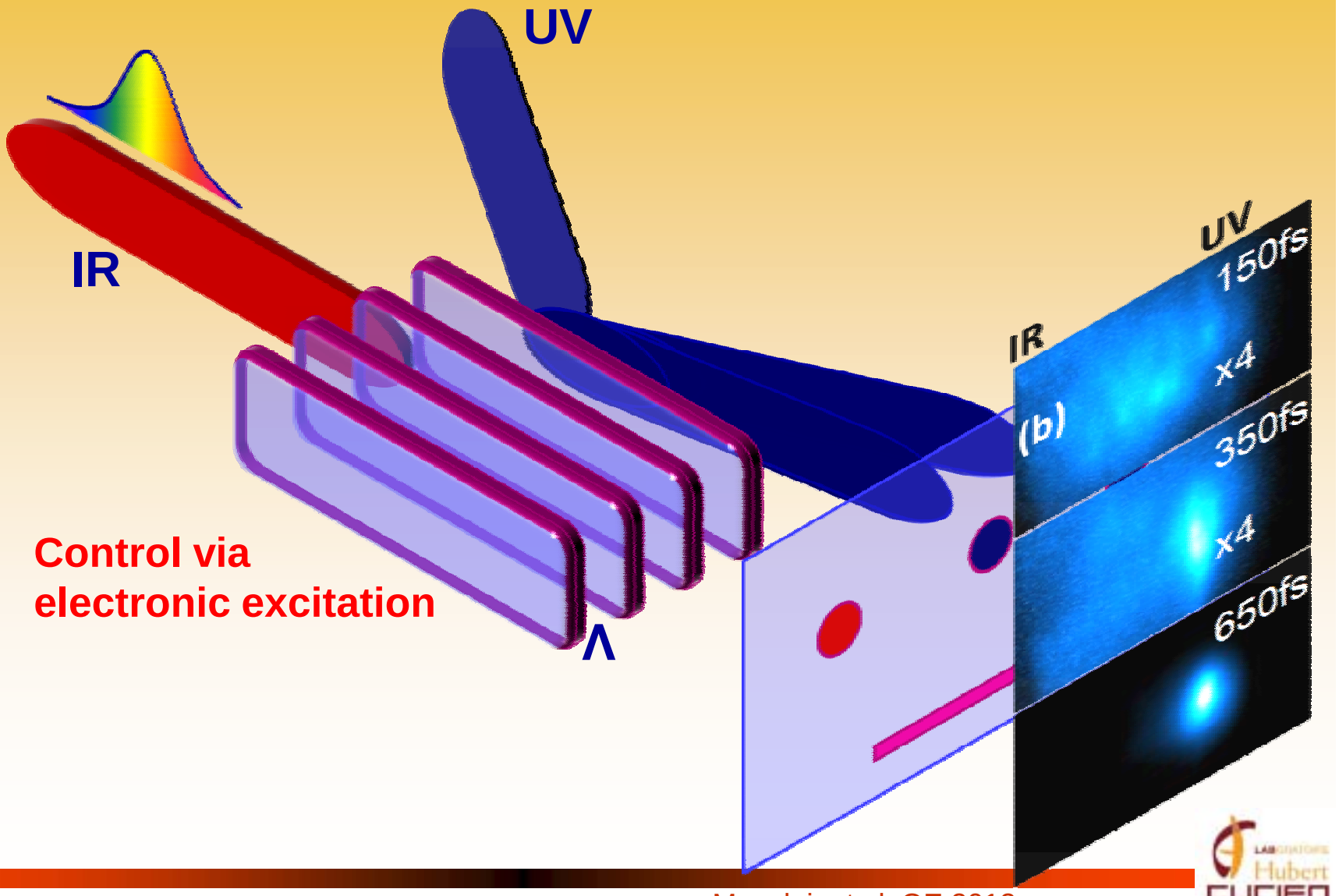
Energy density is important!!!
-regulates the physical excitation
-determines relaxation paths

Modulated refractive index changes

Q: How controllable is the nanoscale pattern?

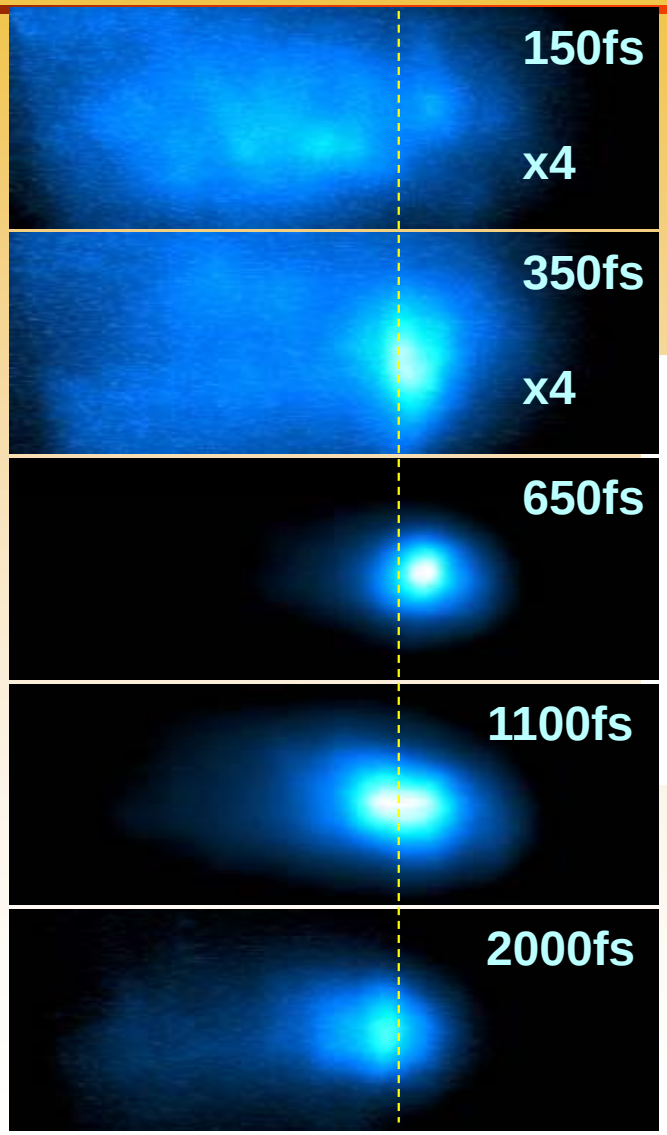


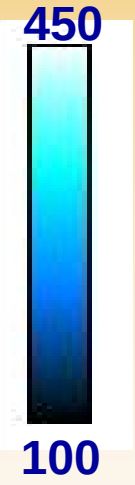
Nano-control via diffraction feedback: a-SiO_2

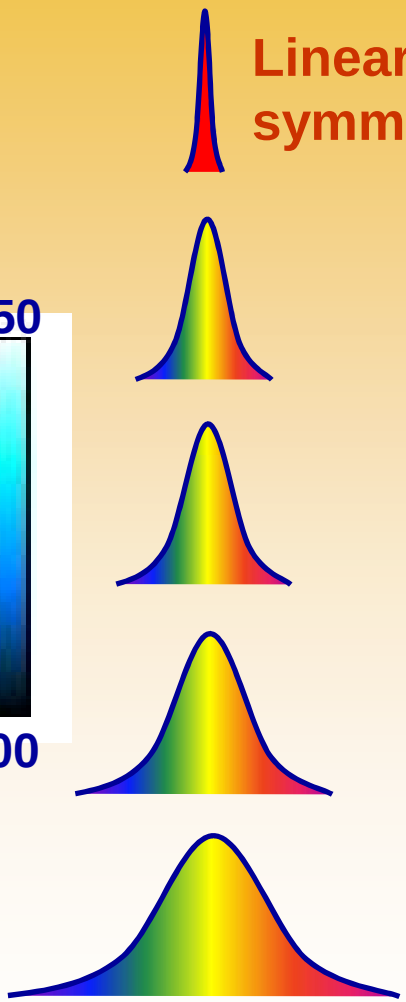


Mauclair et al. OE 2012

Nano-control via diffraction feedback: a-SiO_2







Linear chirp,
symmetric stretching

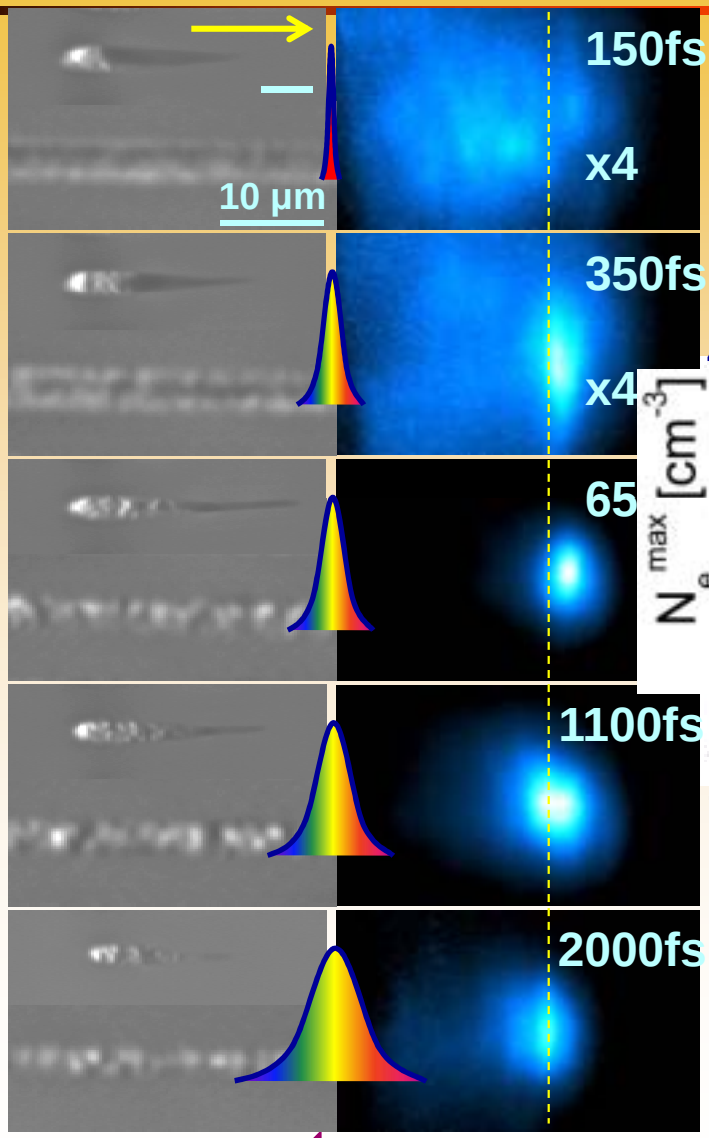
Controllable Periodicity

Controllable arrangement

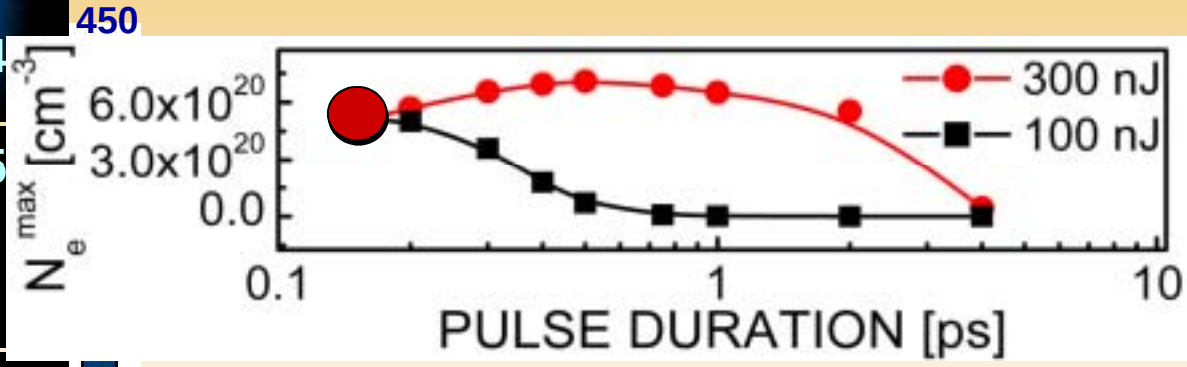
400 300 250 210 Λ

Mauclair et al. OE 2012

Nano-control via diffraction feedback: a-SiO_2



Electron density



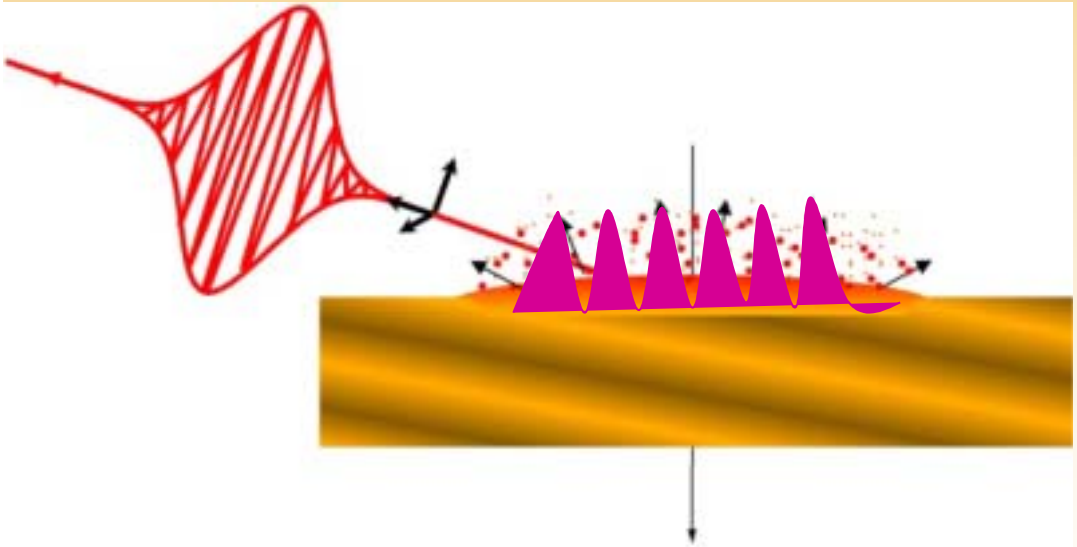
Non monotoneous N_e -max 0.6ps

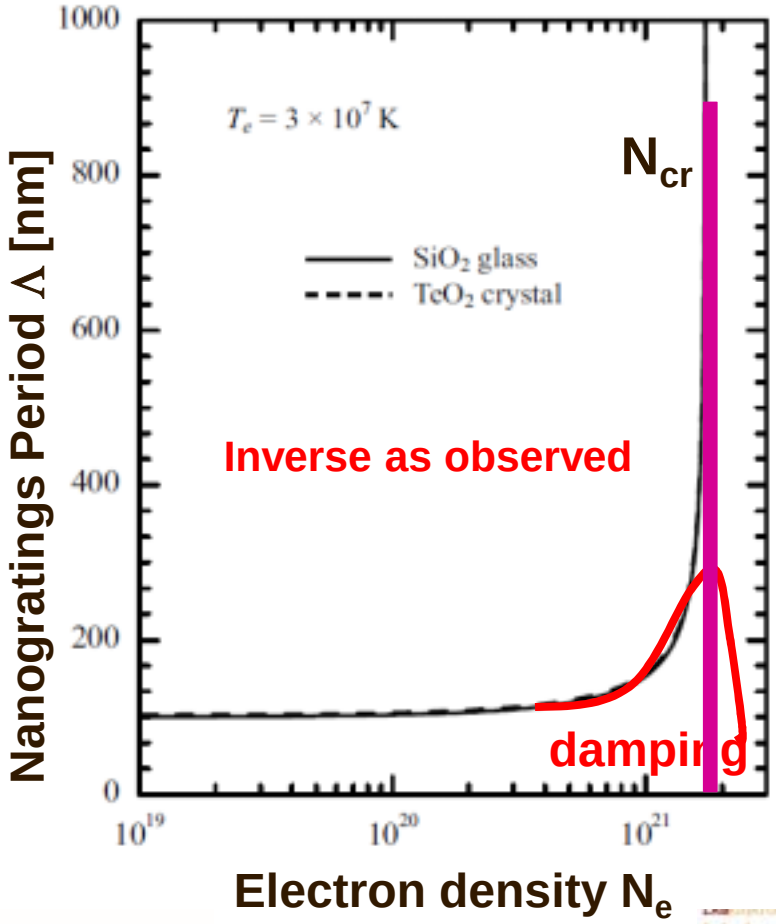
Mauclair et al. OE 2012

Nanostructuring hyphotesis: a-SiO_2

Plasmonic hypothesis?

$$\Delta = \lambda \sqrt{\frac{\varepsilon_r + 1}{\varepsilon_r}} \qquad \varepsilon_r = \varepsilon_{r0} - \frac{N_e}{N_{cr}}$$

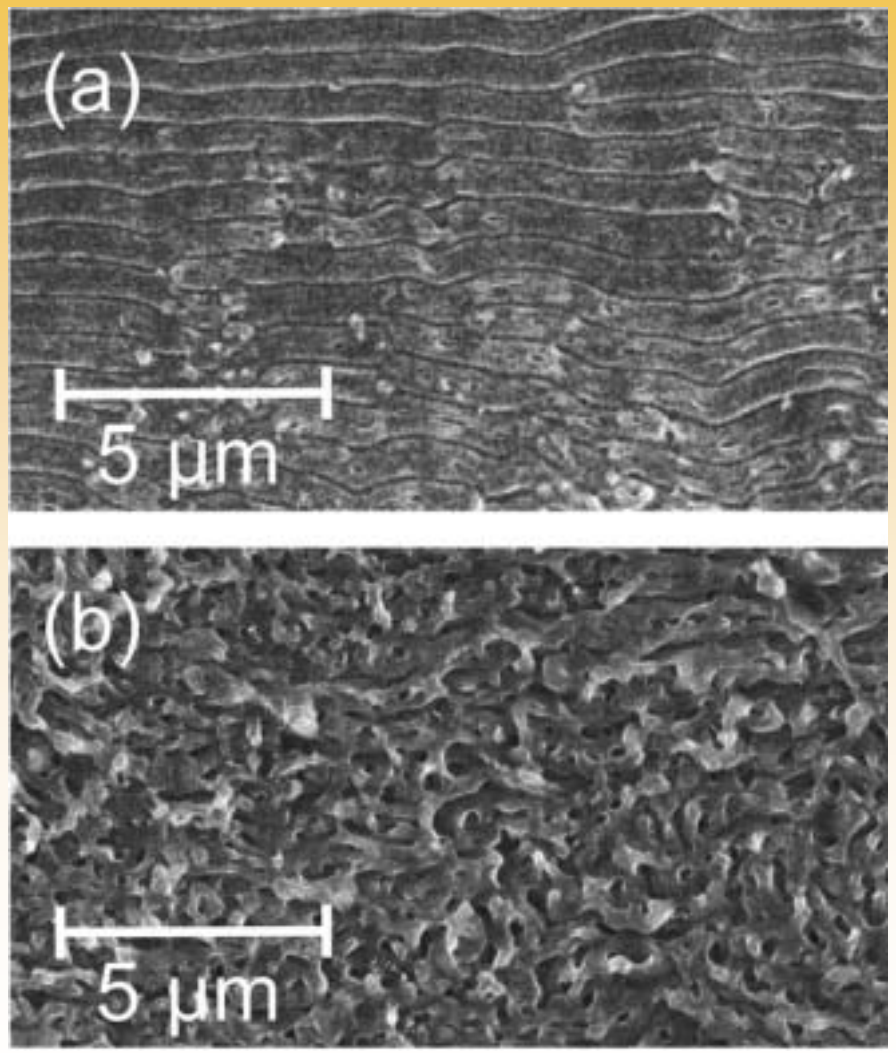


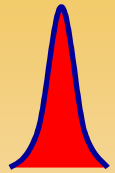


Shimotsuma et al. MPL 2005

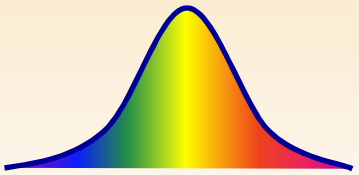
Nanoscale control

Transition Order-Disorder on polymers





150 fs

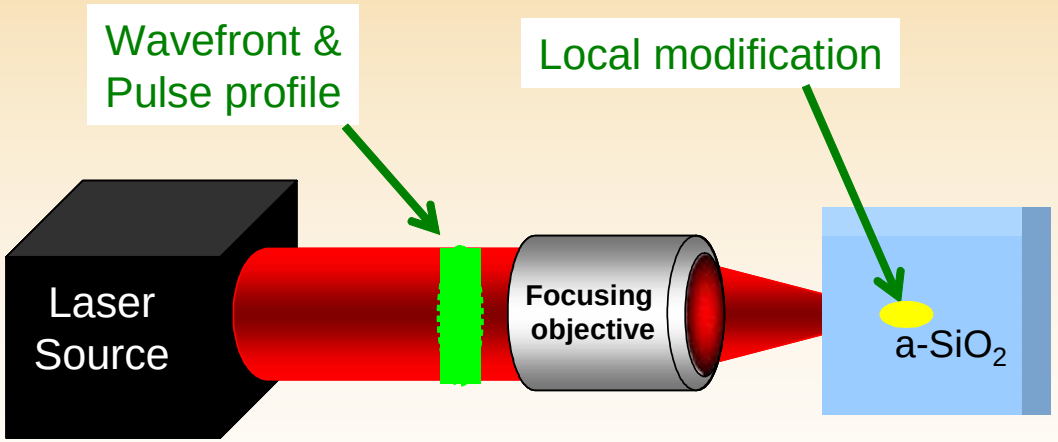


2 ps

Possible applications

What do we want to achieve ?

- **Flexibility in designing laser-interaction**



summary:

Its all about the electrons

Process control

thanks to:

LHC, St. Etienne

C. Mauclair

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MBI, Berlin

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M. Zamfirescu

XIOPM, XiAN

G. Cheng

IT, Novosibirsk

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N. M. Bulgakova

Y. Meschcheryakov

Max-Born-Institut