FEL based Four Wave Mixing

C. Masciovecchio
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- **Introduction to FERMI Free Electron Laser**
- **FERMI End Stations**
- **EIS program (TIMEX & TIMER)**
- **MULTICOLOR Spectroscopy**

Why Free Electron Lasers ?

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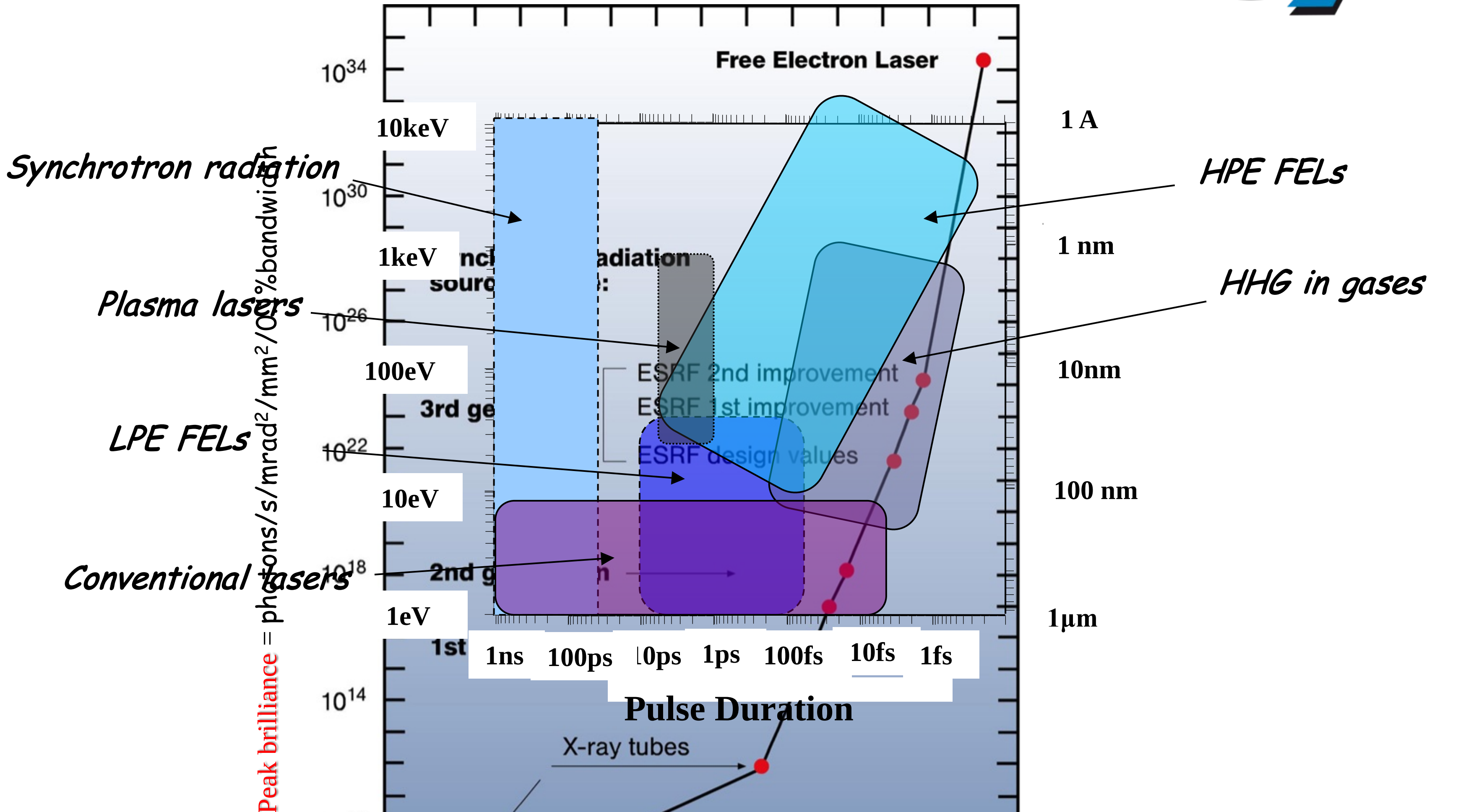
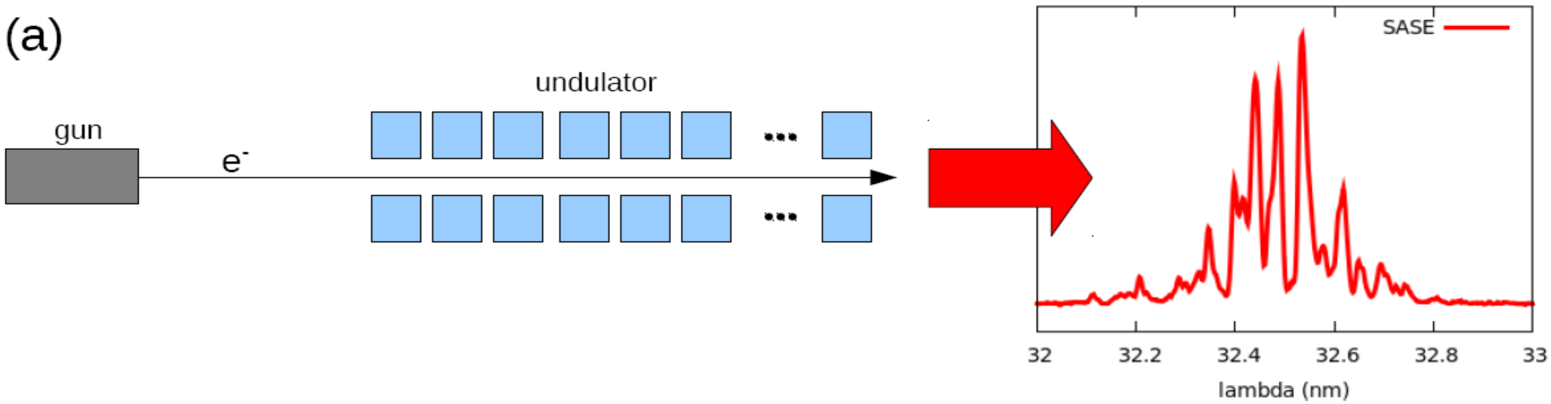


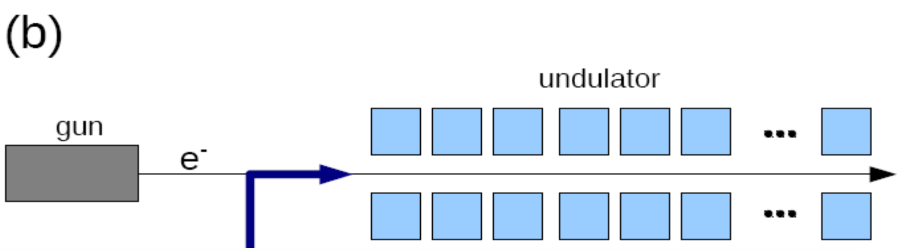
Imaging with high Spatial Resolution (~ λ): fixed target imaging, particle injection imaging,..

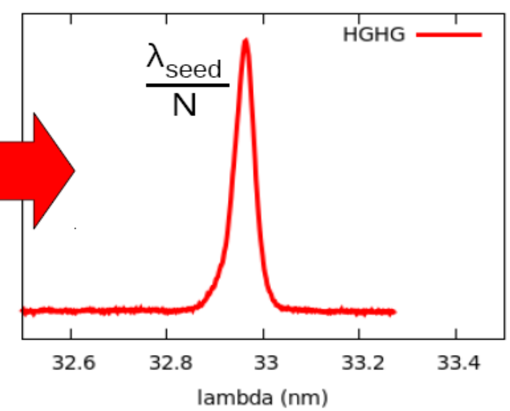
Dynamics: four wave mixing (nanoscale), warm dense matter, extreme condition, ...

Resonant Experiments: XANES (tunability), XMCD (polarization), chemical mapping, ……

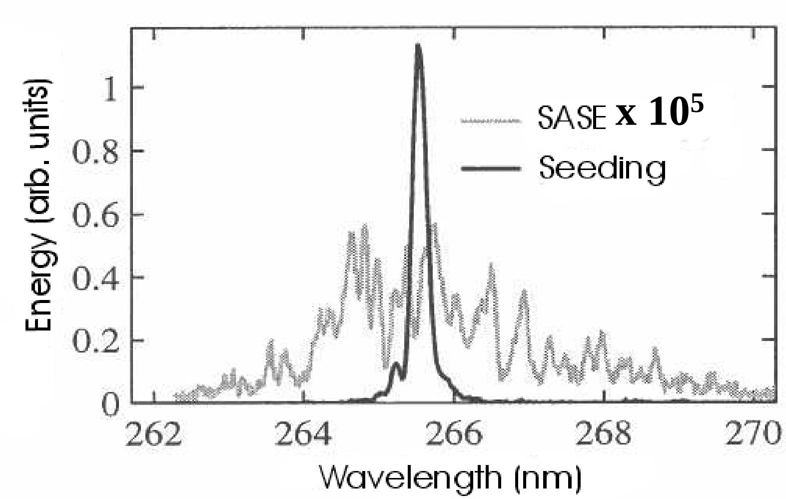
SASE vs Seeded







F. Bencivenga et al., Adv. In Phys. (2015)



L. H. Yu et al., PRL (2003)

The FERMI Free Electron Laser

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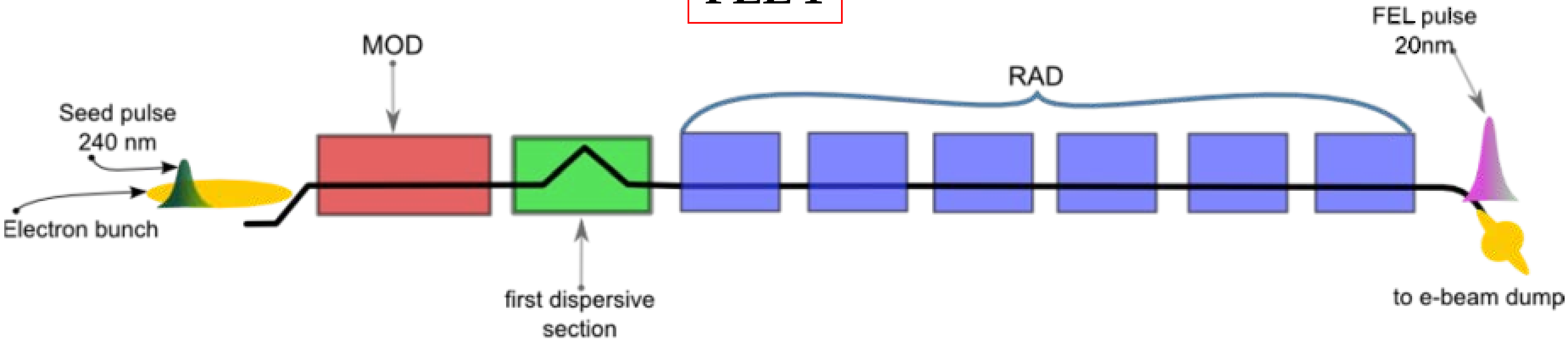
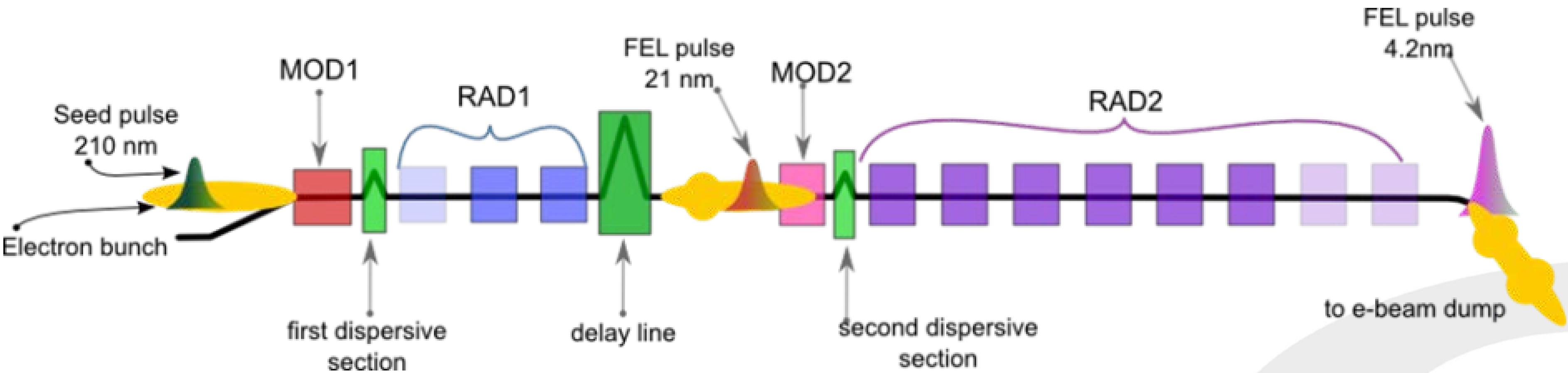
FEL 1



FEL 2



nature photonics
ARTICLES
PUBLISHED ONLINE: 23 SEPTEMBER 2012 | DOI: 10.1038/NPHOTON.2012.233

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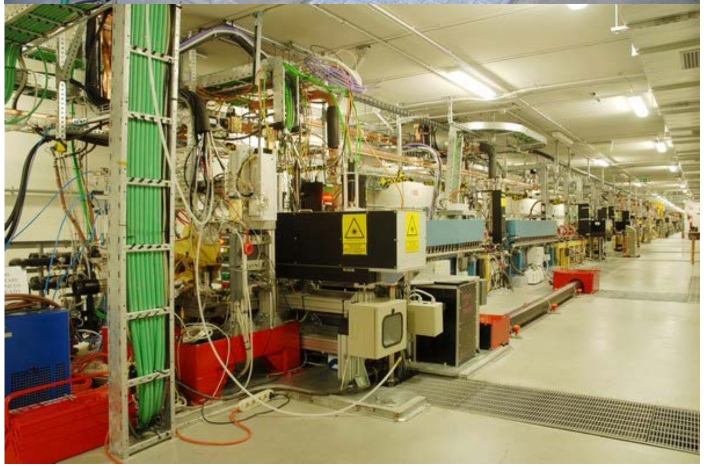
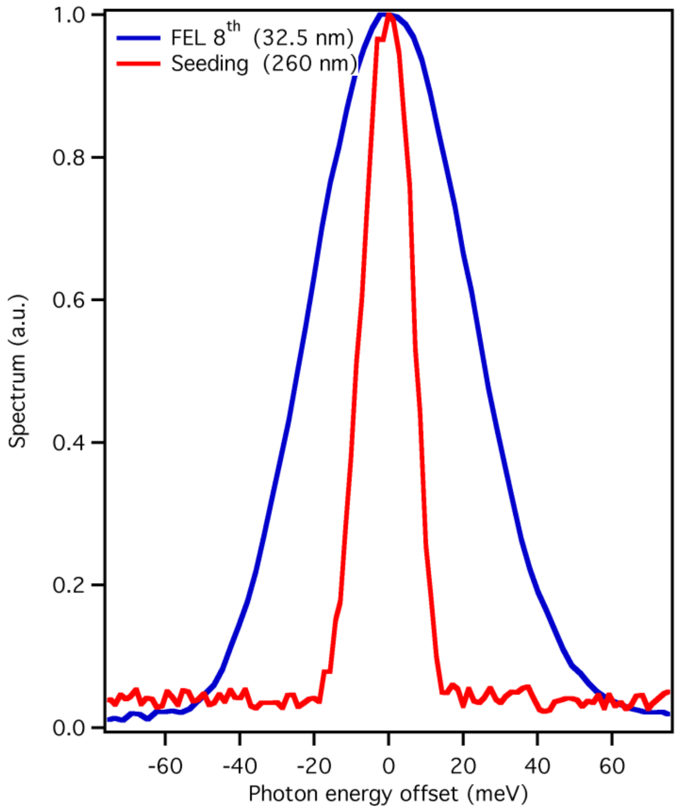
Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet

E. Allaria et al., Nat. Phot. (2012)





$\Delta t < 100$ fs

Flux ~ 10^{13} ph/pulse

E ~ 10 - 500 eV

Total Control on

Pulse Energy

Time Shape

Polarization

nature photonics

ARTICLES

PUBLISHED ONLINE: 20 OCTOBER 2013 | DOI: 10.1038/NPHOTON.2013.277

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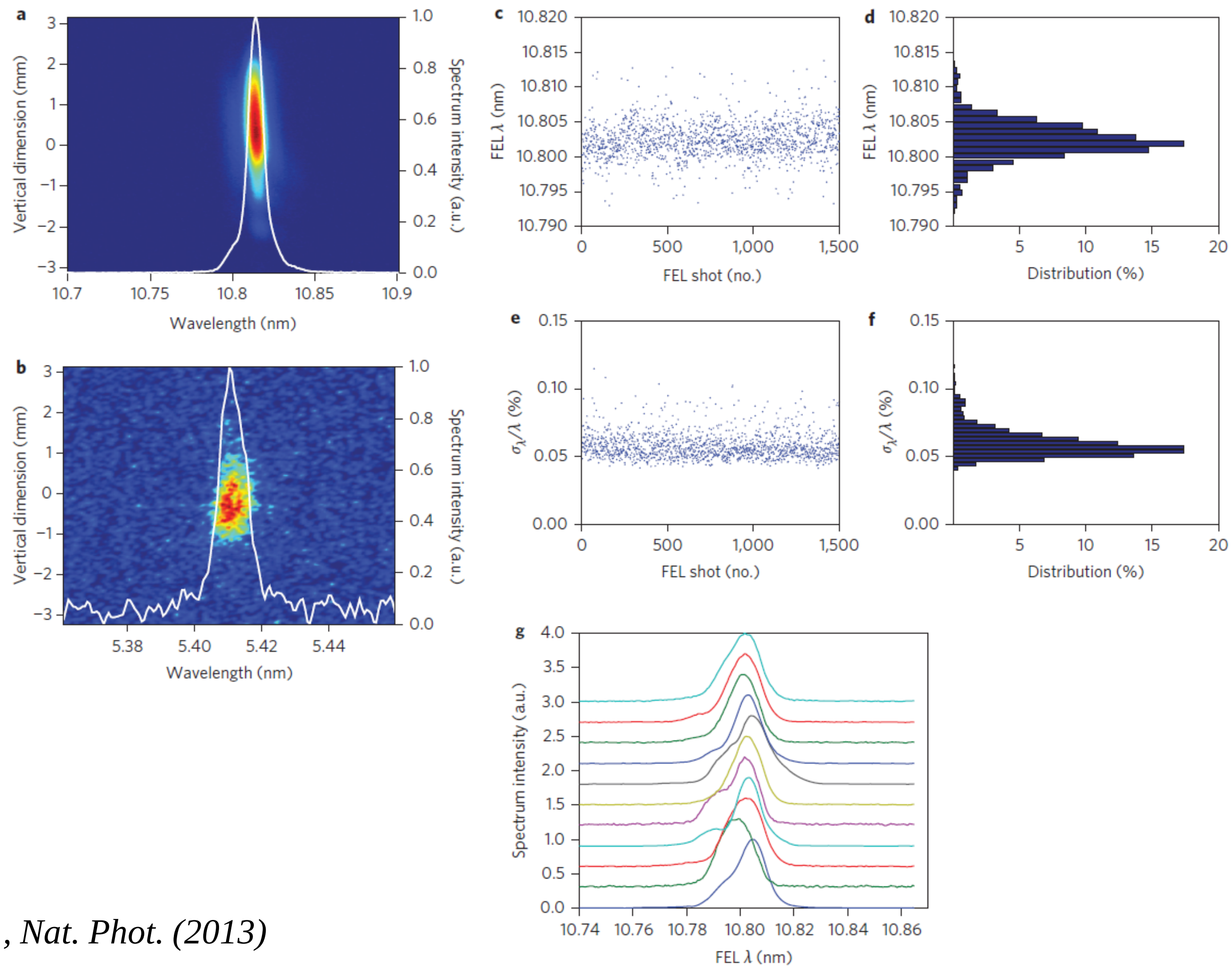
Two-stage seeded soft-X-ray free-electron laser



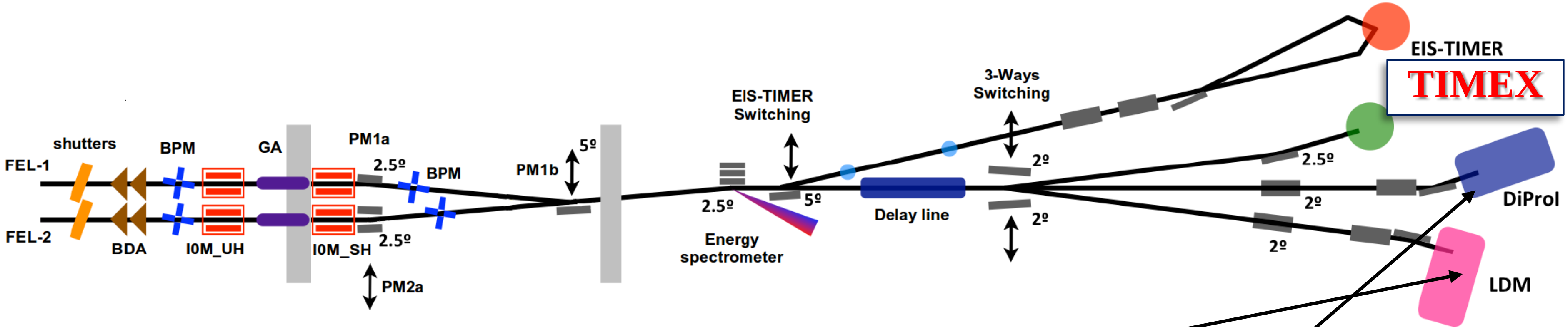
E. Allaria et al., Nat. Phot. (2013)

The Experimental Hall

EIS (Elastic & Inelastic Scattering)

C. Masciovecchio et al., J. Synch. Rad. (2015)

TIMER



TIMEX

LDM (Low Density Matter)

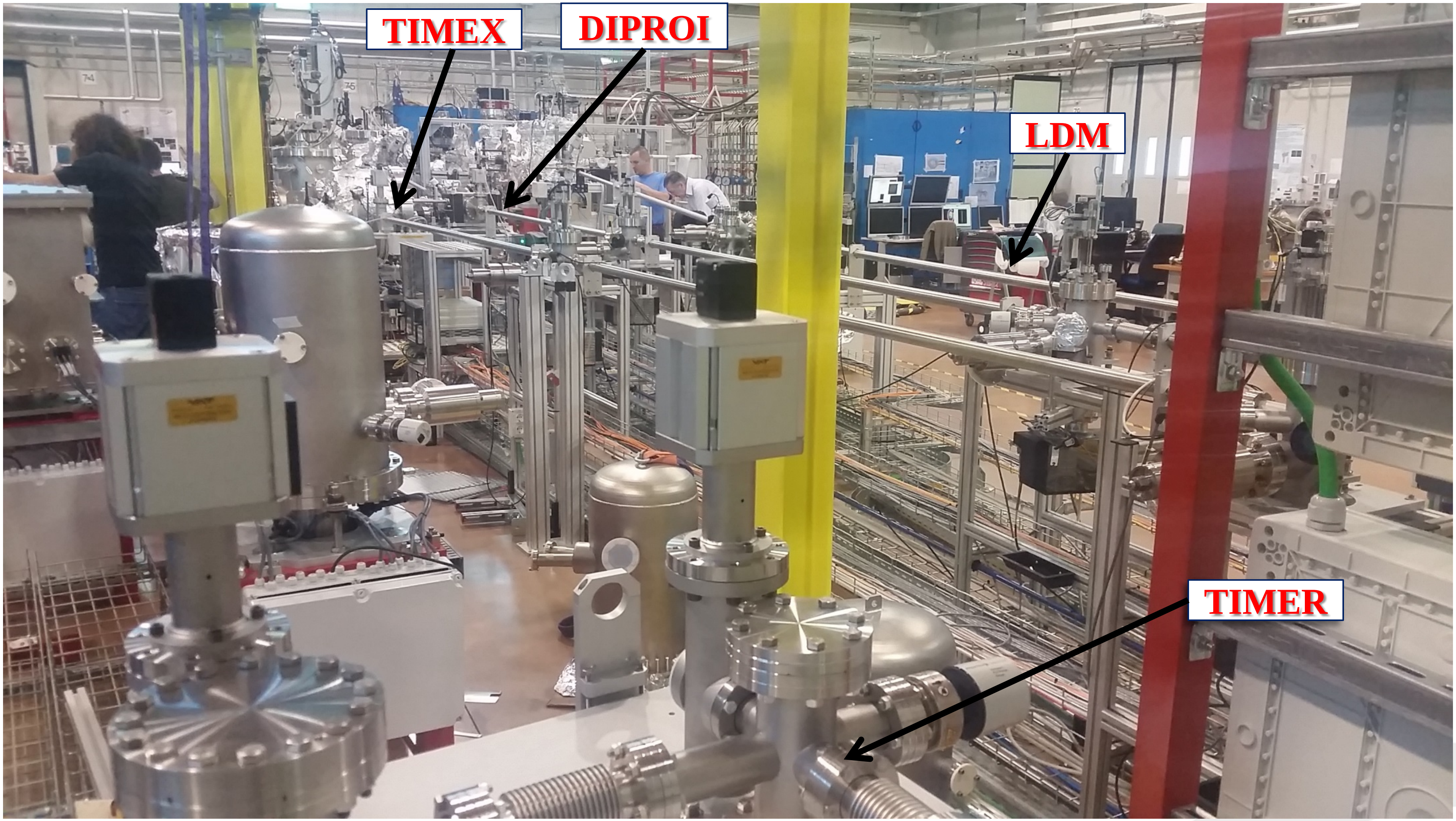
DIPROI (DIffraction & PROjection Imaging)

F. Capotondi et al., J. Synch. Rad. (2015)

MagneDYN (Magnetic Dynamics)

TeraFERMI (THz beramline)

The Experimental Hall



DIPROI Highlight

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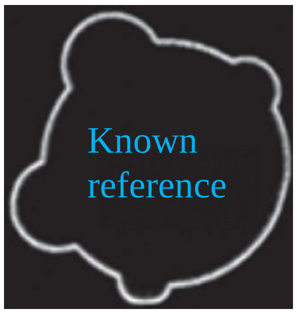
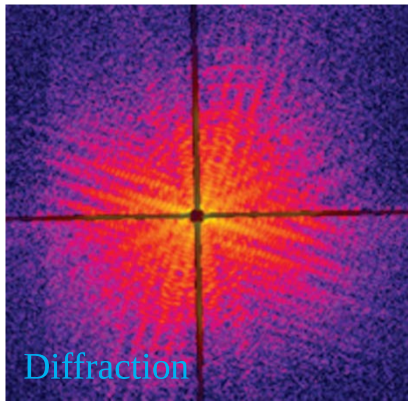
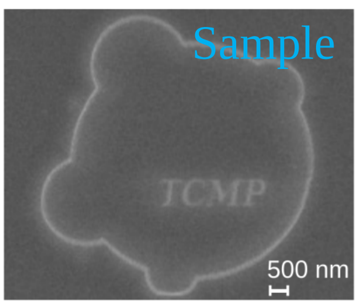
nature COMMUNICATIONS *A. Martin et al. (2014)*

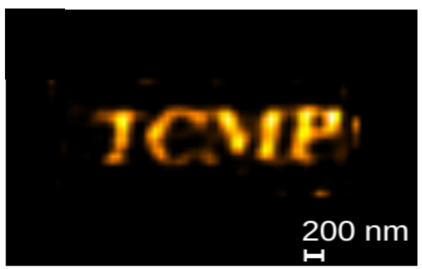
X-ray **holography** with **customizable reference**

Ideal FTH → overcoming restriction due to the reference wave → single-shot imaging





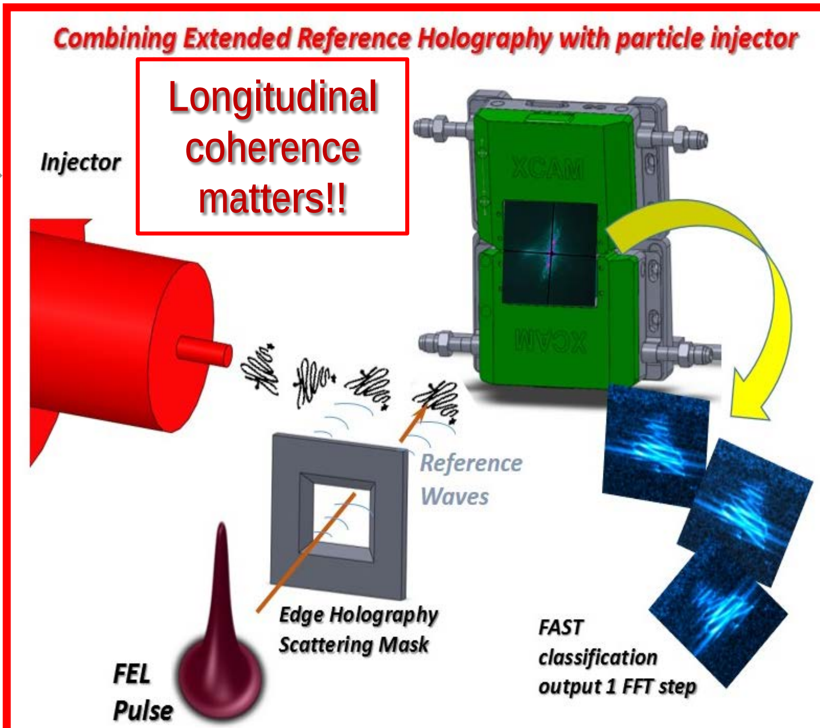




Hologram refined with RAAR phase retrivial

Conjugate-gradient algorithm to recover the image

FTH with an almost **unrestricted choice for the reference**



LDM Highlight

Coherent control at the attosecond time scale

K. Prince et al., submitted

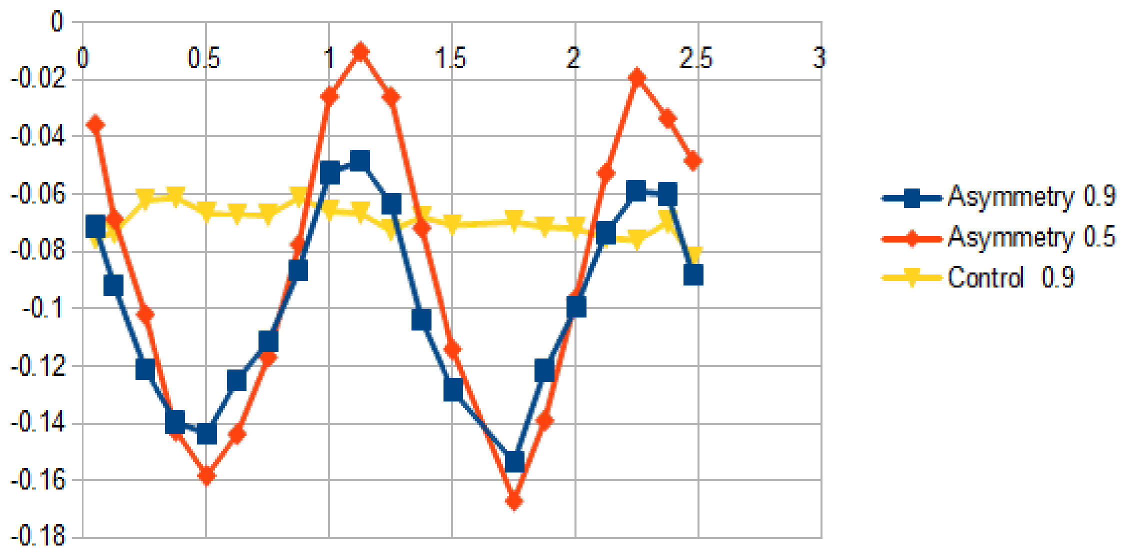
Interference effect among quantum states using single and multiphoton ionization

C. Chen et al., PRL (1990)

$$\text{Intensity} = |\, M1 + M2(\phi)| = |\, M1|^2 + |M2(\phi)|^2 + 2\,\text{Re}(M1\; M2(\phi))$$

Use of first (62.974 eV) and second harmonic on $2p^54s$ resonance of Ne

Change of the phases among the two harmonics ‘invented’ by Allaria et al.,



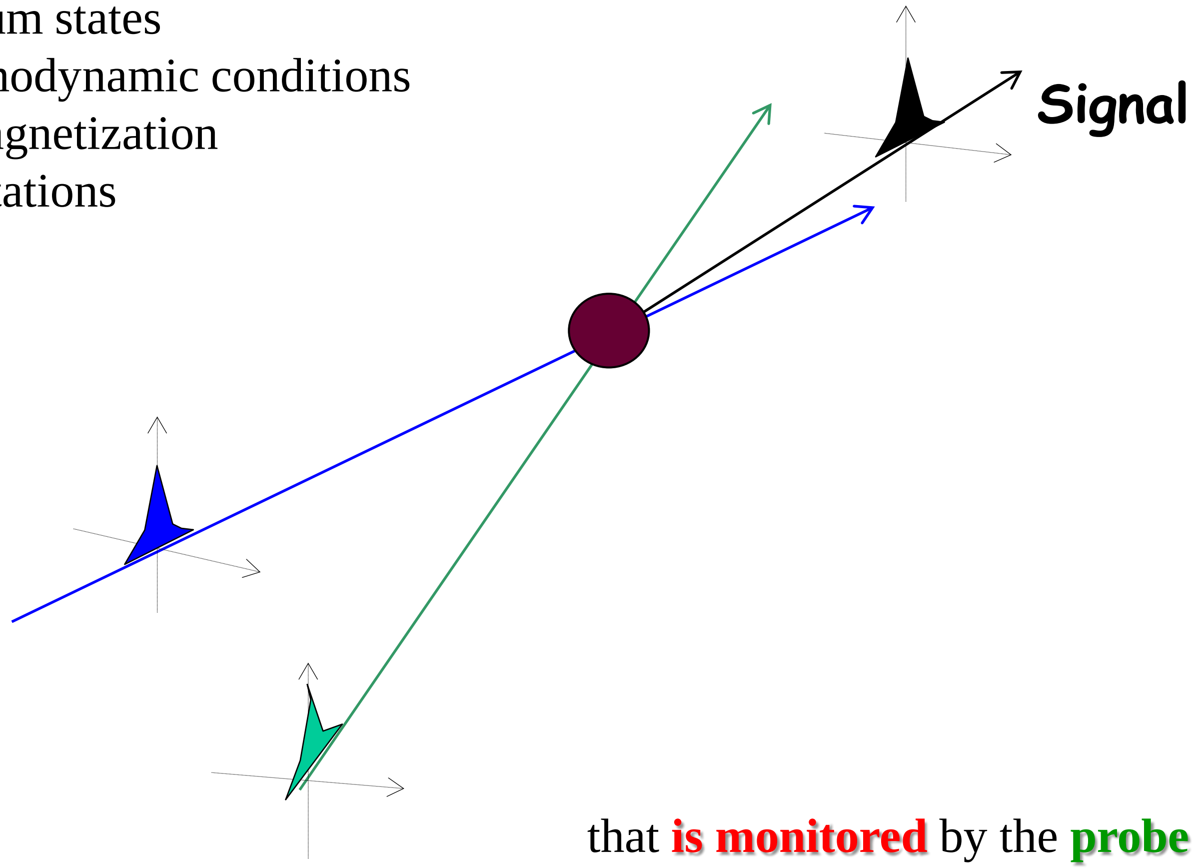
Signal detected as function of phase on the VMI detector

Control of the phase among the two pulses!

Pump & Probe

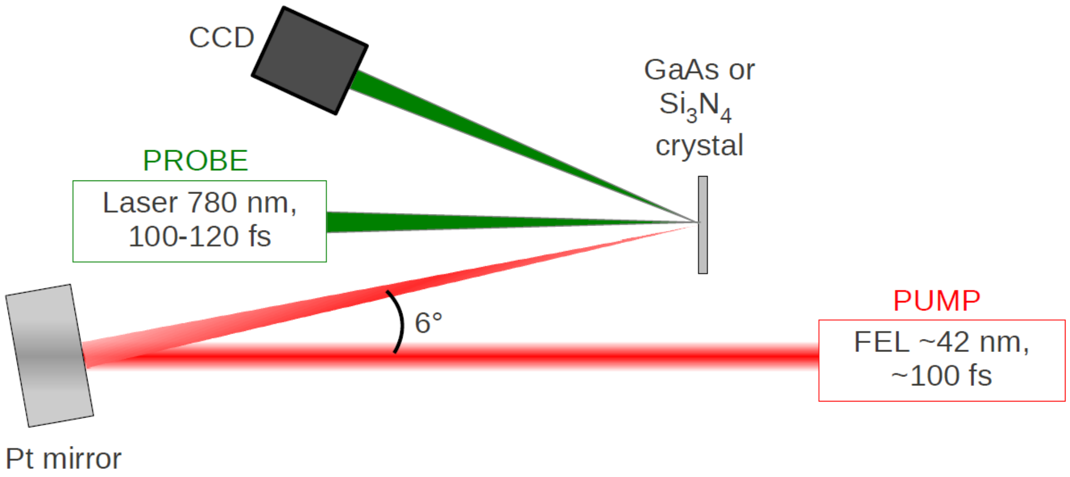
The **pump** pulse produces **a change** in the sample

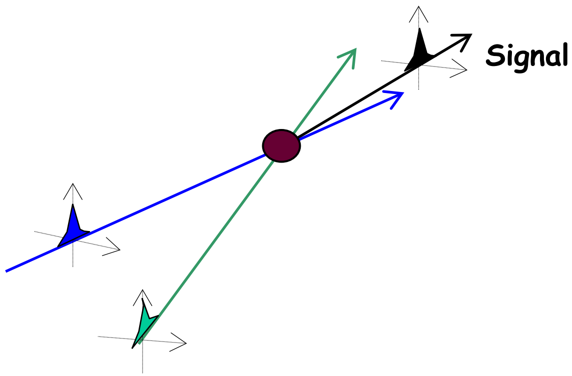
- stimulate a chemical reaction
- non-equilibrium states
- extreme thermodynamic conditions
- ultrafast demagnetization
- coherent excitations
- ....



that **is monitored** by the **probe** pulse

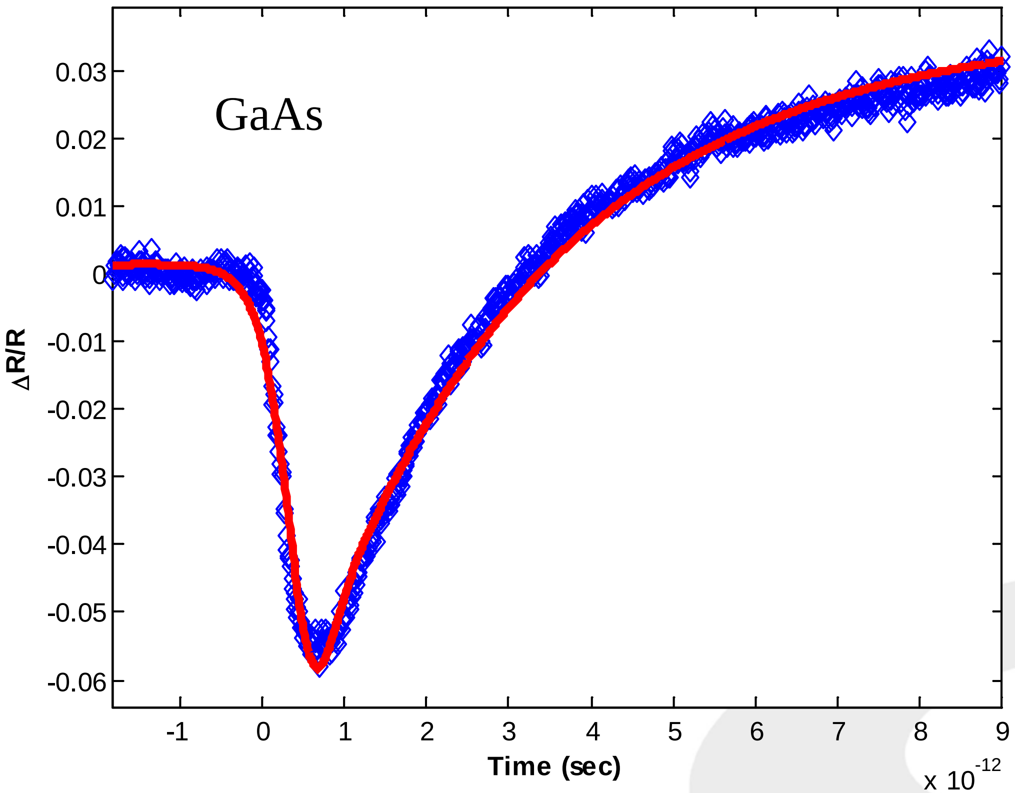
FEL-pump & Laser-probe





Jitter < 7 fs

WORLD RECORD !!



M. Danailov et al., Opt. Express (2014)

Elastic and Inelastic Scattering (EIS)

The **Sample Side**

Short pulses with very high peak power   Δt ~ 100 fs ; Peak Power ~ 5 GW ; E ~ 100 eV

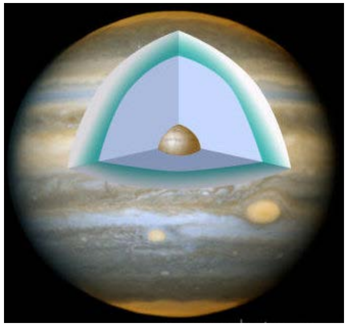
What happens to the **Sample?**   Non-equilibrium distribution of electrons

Converge (electron-electron & electron-phonon collisions) to equilibrium (Fermi-like)

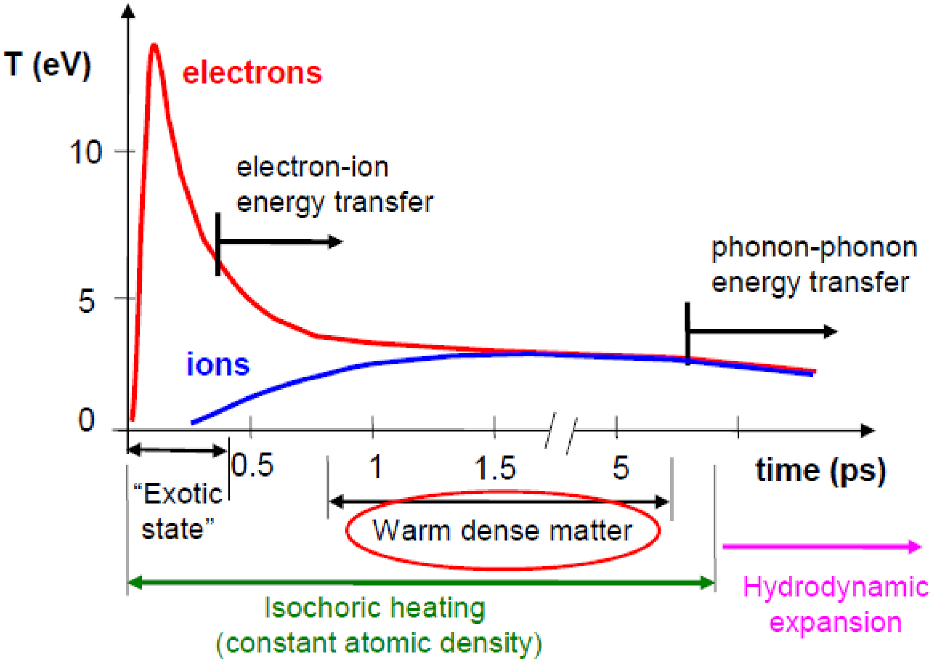
The intensity of the FEL pulses will determine the process to which the sample will undergo: simple heating, structural changes, ultrafast melting or ultrafast ablation

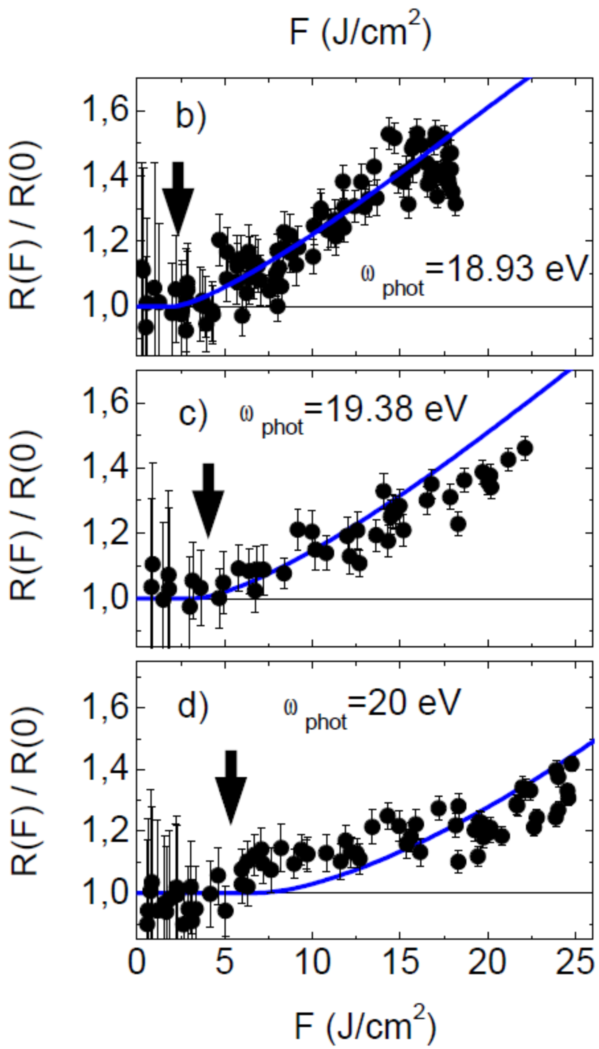
TIMER

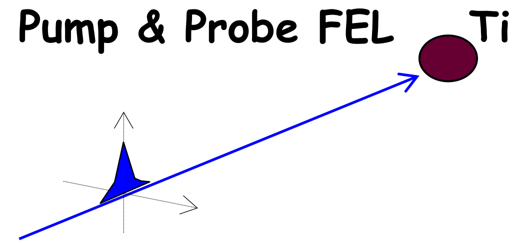
TIMEX



interior of **large planets** and stars







$$R \approx \left| \frac{\sqrt{\epsilon} - 1}{\sqrt{\epsilon} + 1} \right|^2$$

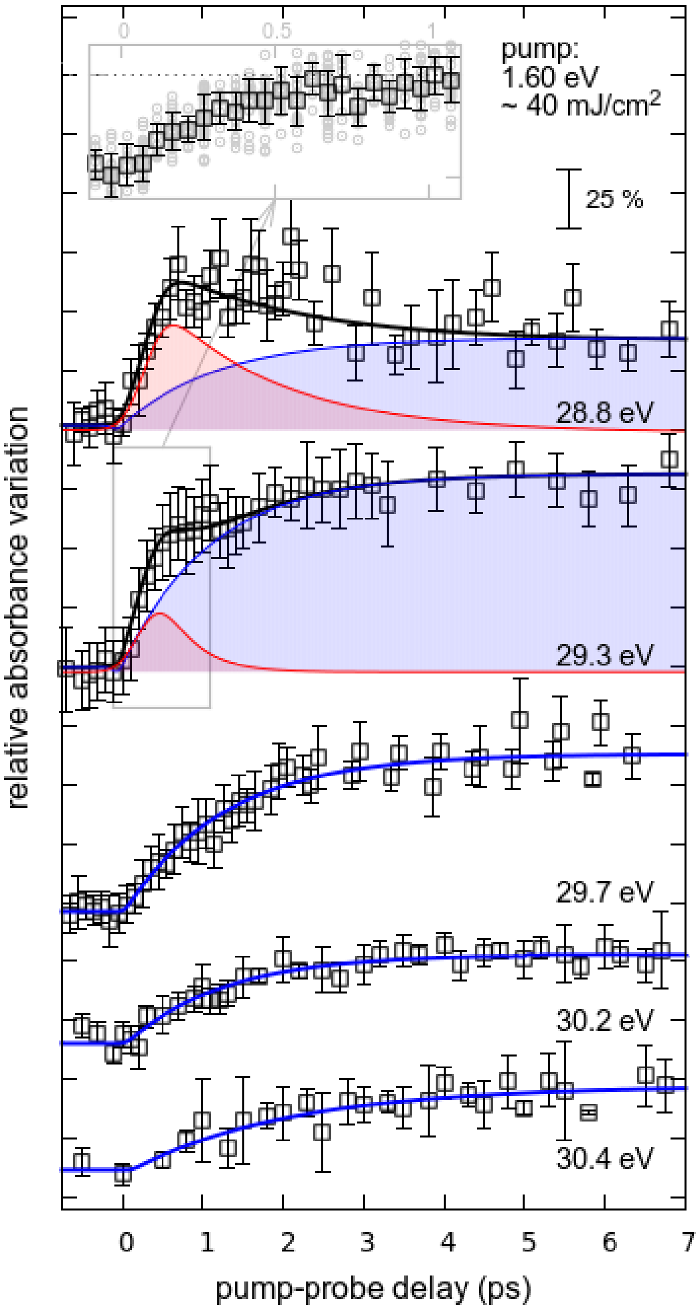
$$Z(F) = \frac{N_e(F)}{N_a} = \frac{m_e \epsilon_0}{e^2 N_a} \int_{-\infty}^{+\infty} \omega_p^2(F, t') G(t') dt'$$

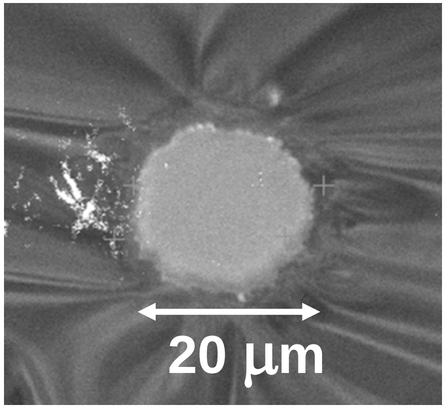
$$\overline{E}(F, t) = \frac{F}{L} \int_{-\infty}^{t} (1 - R(F, t'))\ G(t') dt'$$

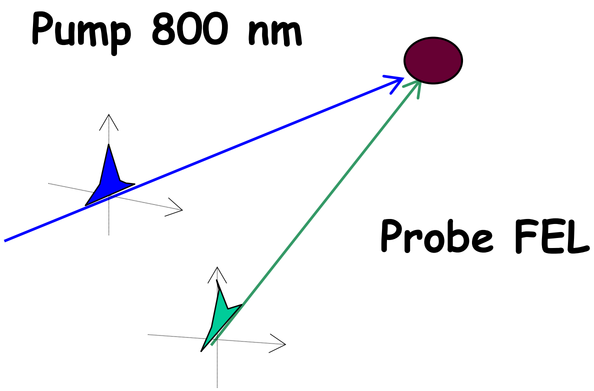
$$\Downarrow$$

$$T_e(F)$$

Pump & Probe on Germanium







Upon the absorption edge (Fermi level) the spectroscopy is sensitive both to the **Fermi function smearing** (red curve) and to the shift of the edge due to the **metallization** of the sample (blue curve)

E. Giangrisostomi et al., in preparation

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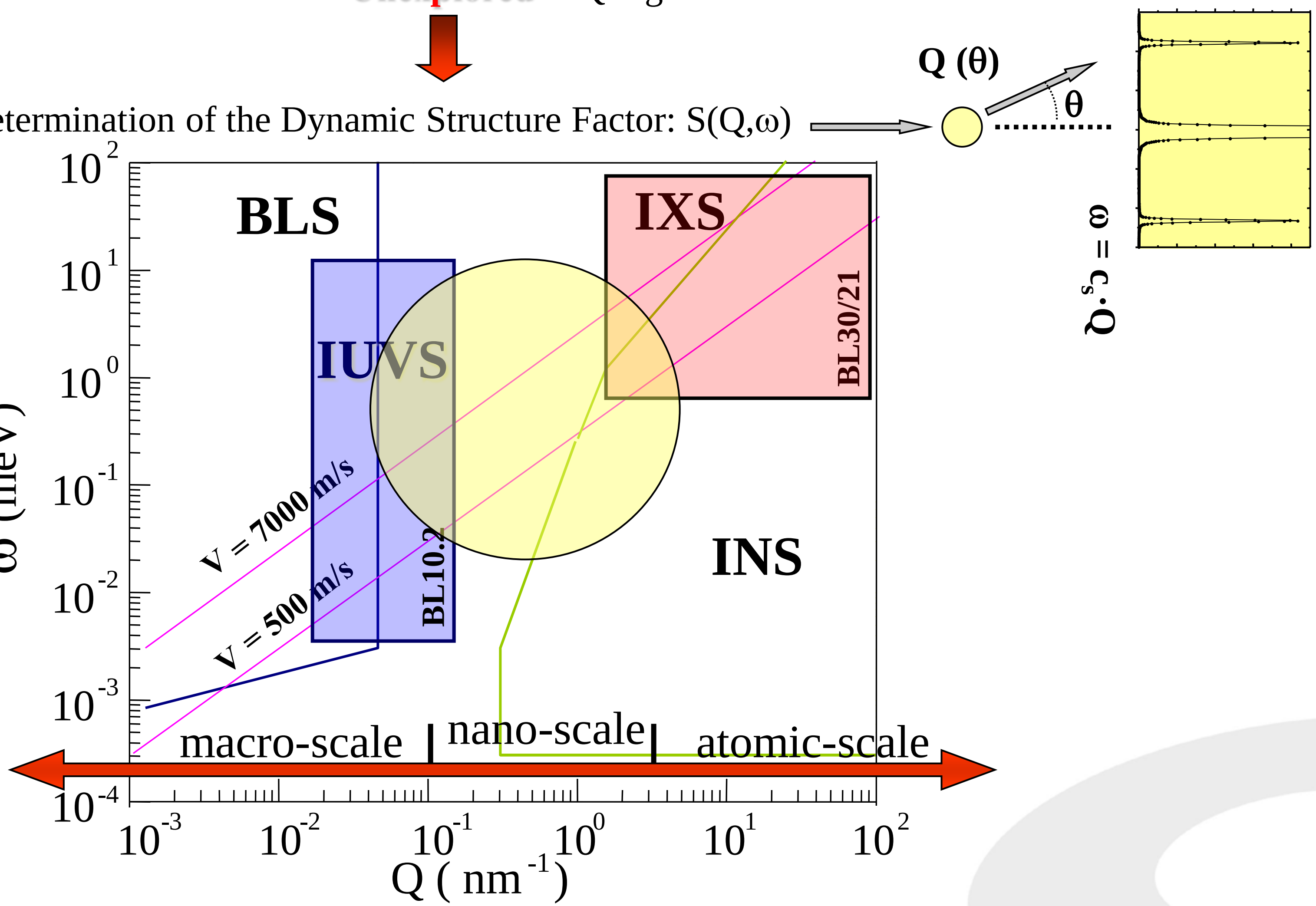
TIMER **TIME-Resolved spectroscopy of mesoscopic dynamics in condensed matter**

Challenge: Study Collective Excitations in **Disordered Systems**
in the **Unexplored** ω-Q region

Determination of the Dynamic Structure Factor: S(Q,ω)



Why Disordered Systems ?

UNSOLVED PROBLEMS IN PHYSICS

Condensed matter physics

Amorphous solids

What is the nature of the transition between a fluid or regular solid and a glassy phase? What are the physical processes giving rise to the general properties of glasses?

High-temperature superconductors

What is the responsible mechanism that causes certain materials to exhibit superconductivity at temperatures much higher than around 50 Kelvin?

Sonoluminescence

What causes the emission of short bursts of light from imploding bubbles in a liquid when excited by sound?

Turbulence

Is it possible to make a theoretical model to describe the statistics of a turbulent flow (in particular, its internal structures)? Also, under what conditions do smooth solution to the Navier-Stokes equations exist?

Glass is a very general state of condensed matter → a large variety of systems can be transformed from liquid to glass

The liquid-glass transition cannot be described in the framework of classical phase transitions since T_g depends on the quenching rate → one cannot define an order parameter showing a critical behaviour at T_g

Why at the nanoscale ?

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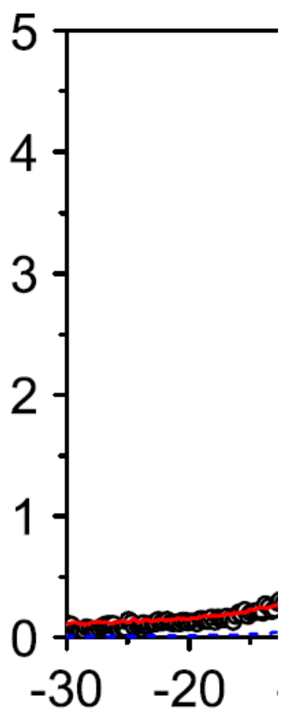
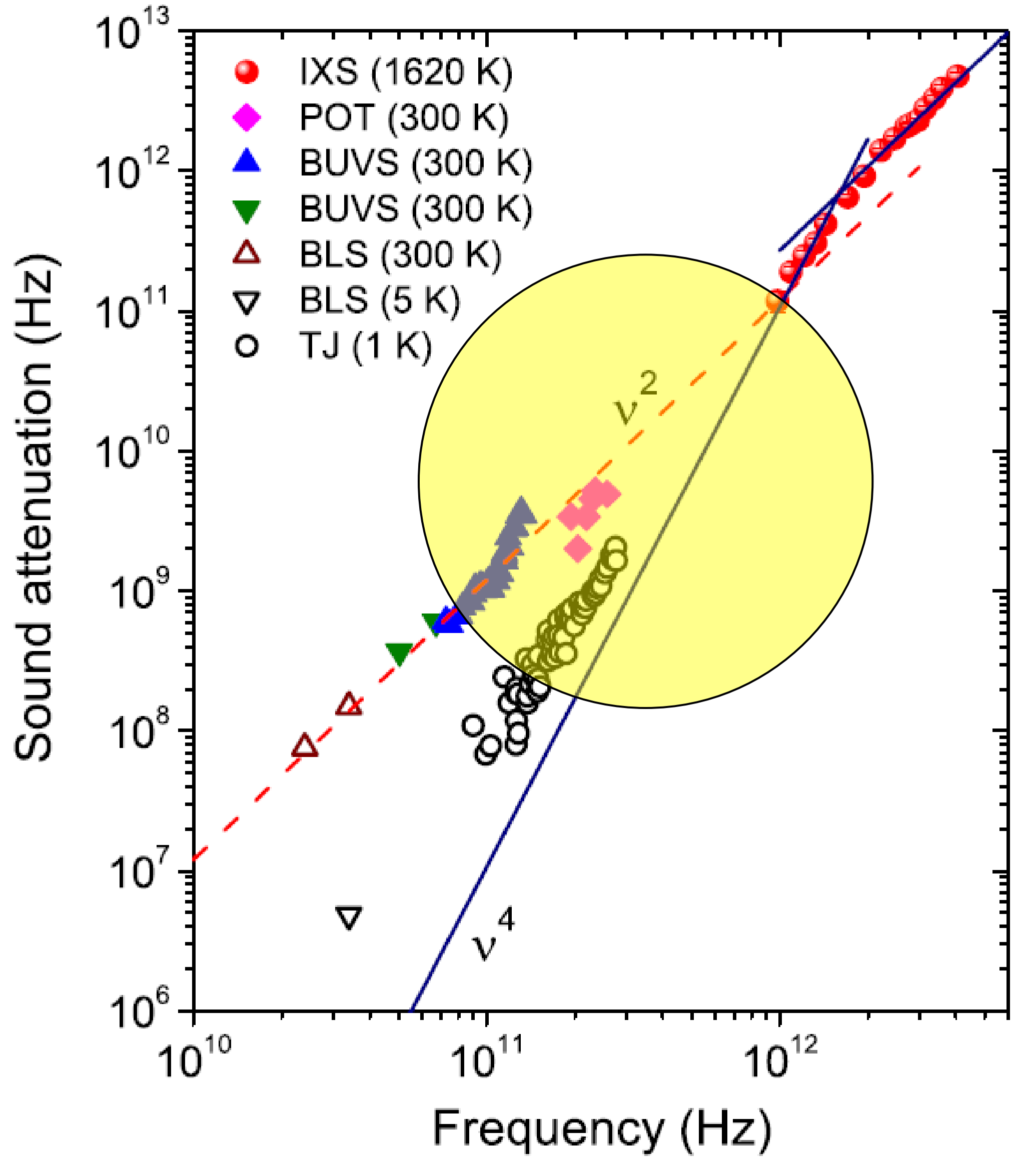
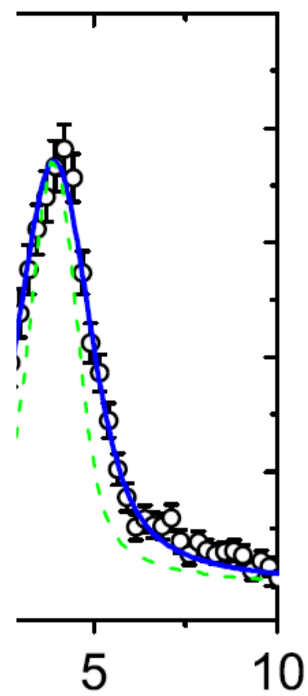
Trieste

The nature of ear ($V-SiO_2$)







Funda

Why at the nanoscale ?

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Water exhibits very unusual properties:

- Negative volume of melting
- Density maximum in the normal liquid range
- Isothermal compressibility minimum in the liquid
- Increasing liquid fluidity with increasing pressure

"...water's puzzling properties are not understood and 63 anomalies that distinguish water from other liquids remain unsolved...." H. E. Stanley, Physica A (2007)

T_g T_H T_M

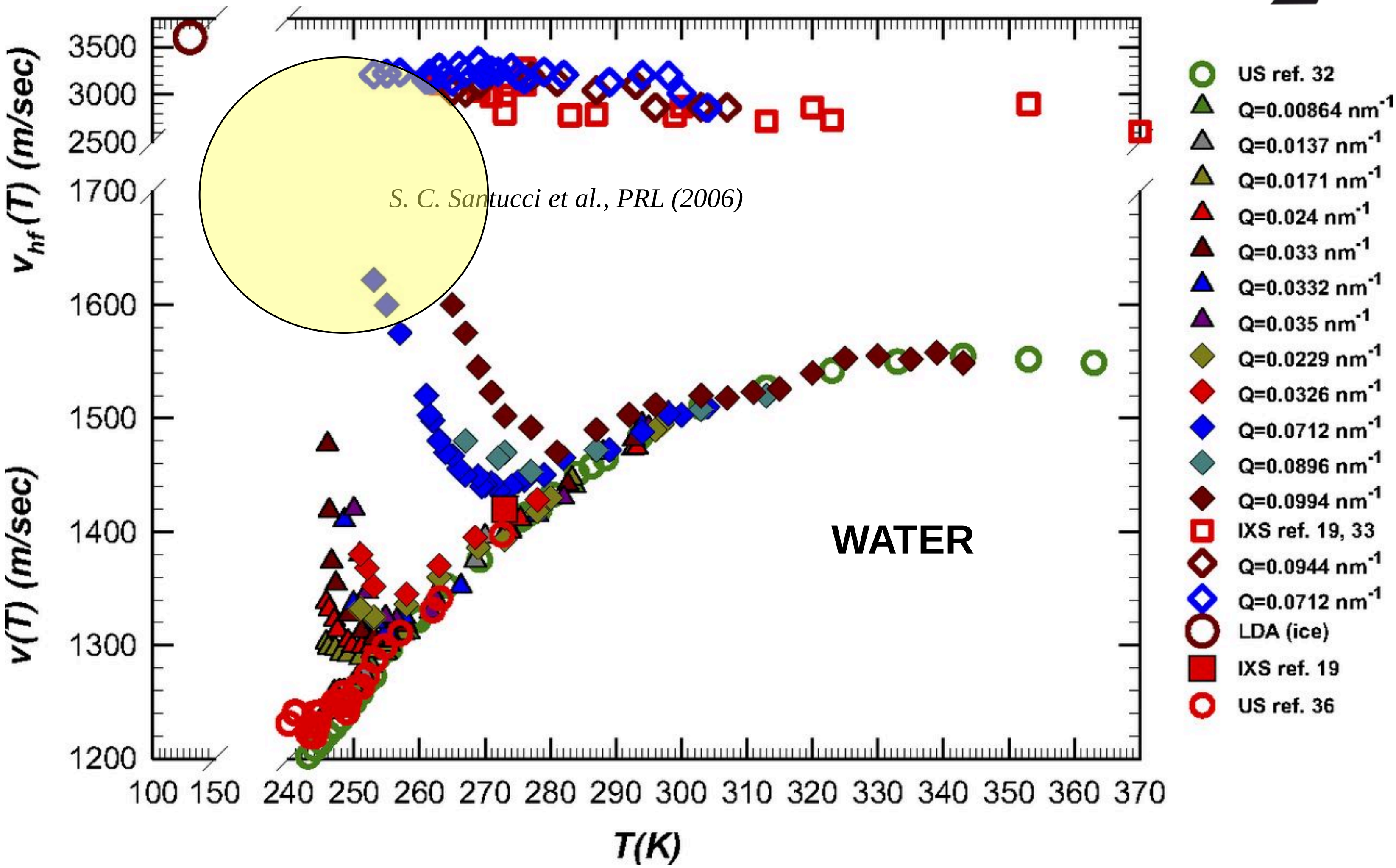
nature International weekly journal of science

Ultrafast X-ray probing of water structure below the homogeneous ice nucleation temperature

J. A. Sellberg et al., Nature (2014)

The hope now is that these observations and our detailed structural data will help identify those theories that best describe and explain the behaviour of water.

Why at the nanoscale ?



……only by treating water as a viscoelastic system is it possible to understand the microscopic origin of water anomalies. The proposed scenario here has been developed using a dynamic approach……

F. Mallamace et al., PNAS (2013)

EIS beamline - TIMER

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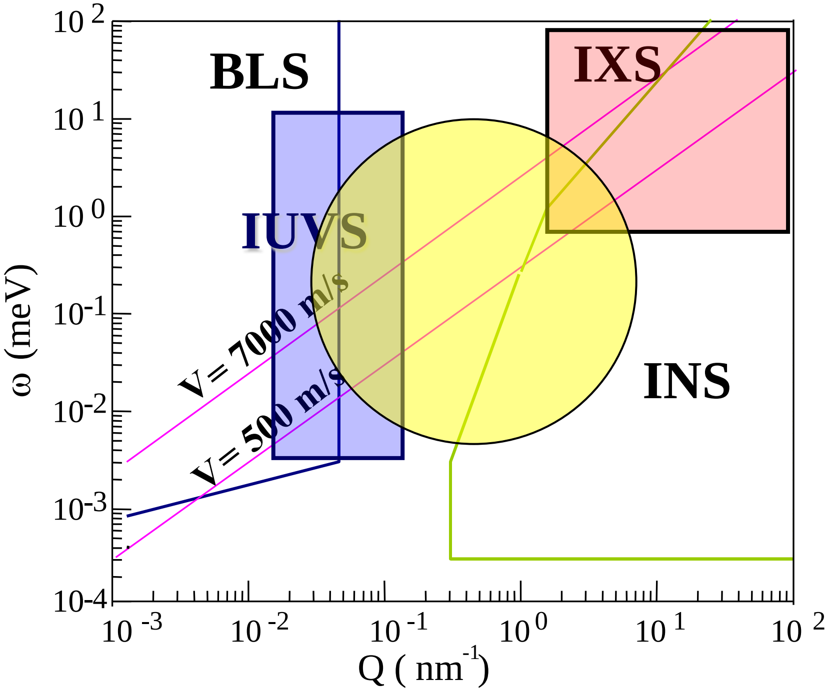
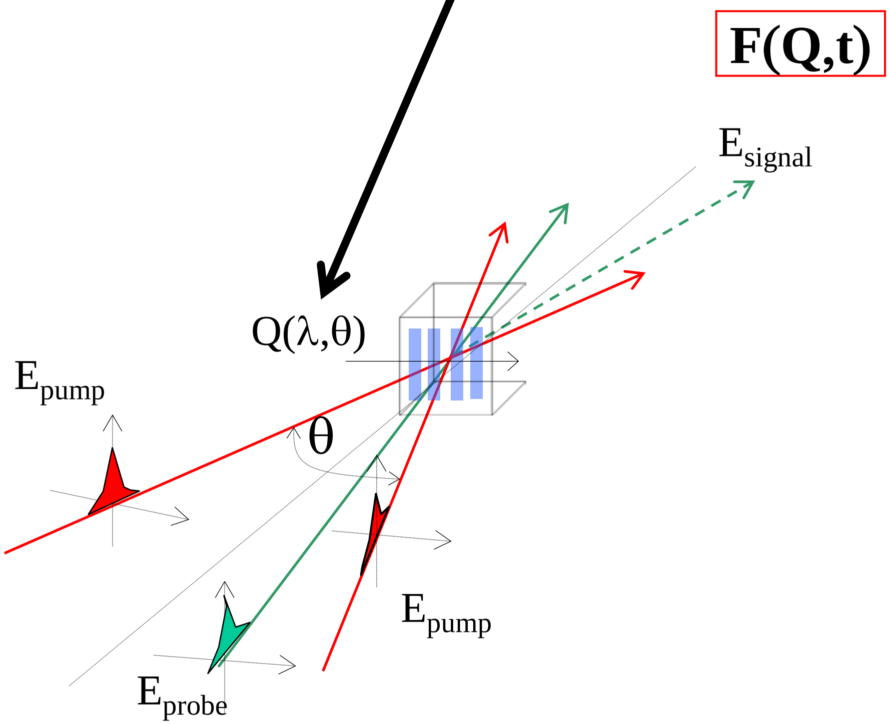


Solution: Free Electron Laser based Transient Grating Spectroscopy



European Research Council

Funded Grant: **1.8 M€**

The Spectrum

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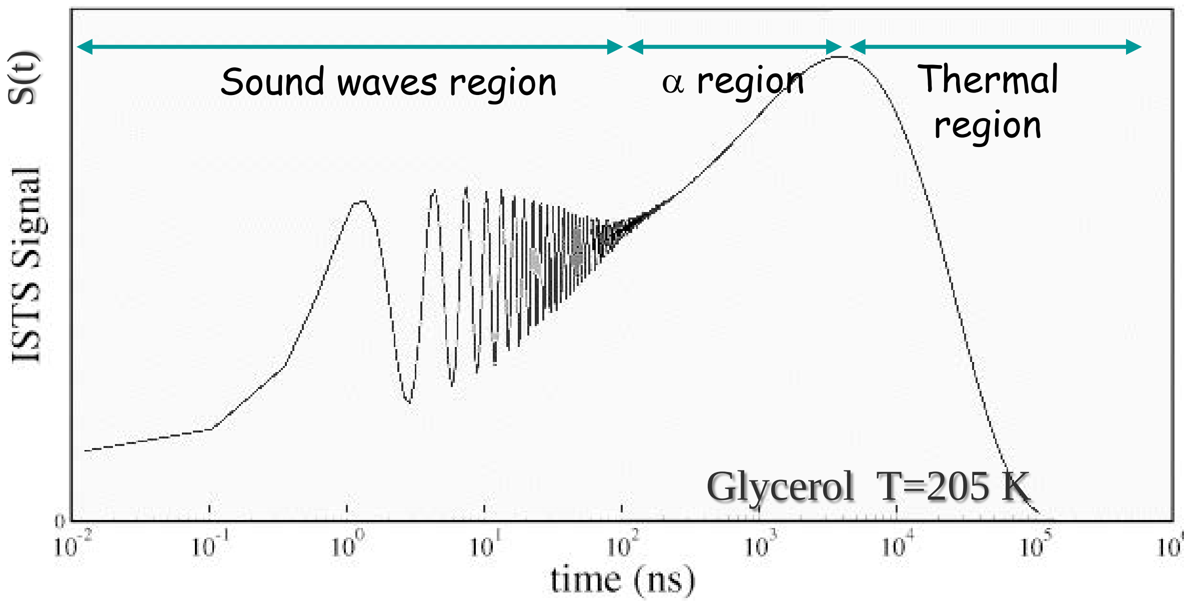
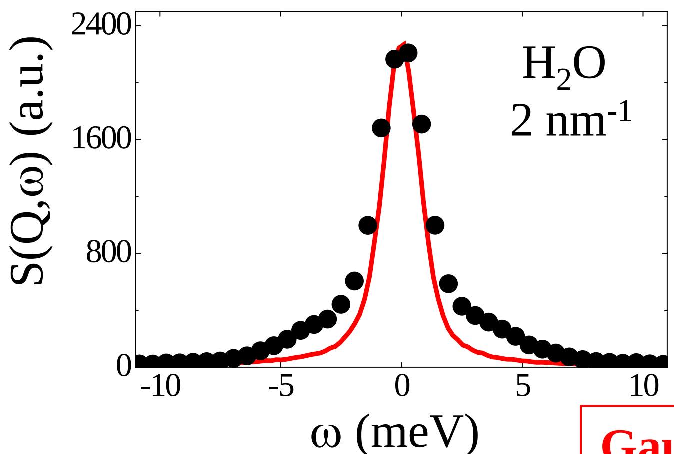
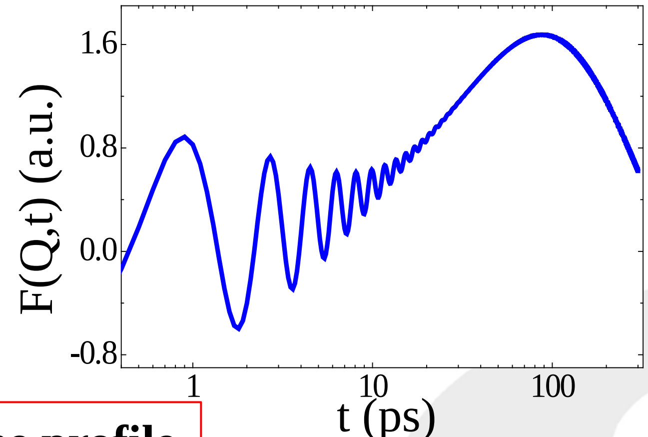
Optical absorption → Temperature Grating →Time-dependent Density Response

(driven by thermal expansion)

$S(t) \approx (\text{cost} - F(Q,t))$

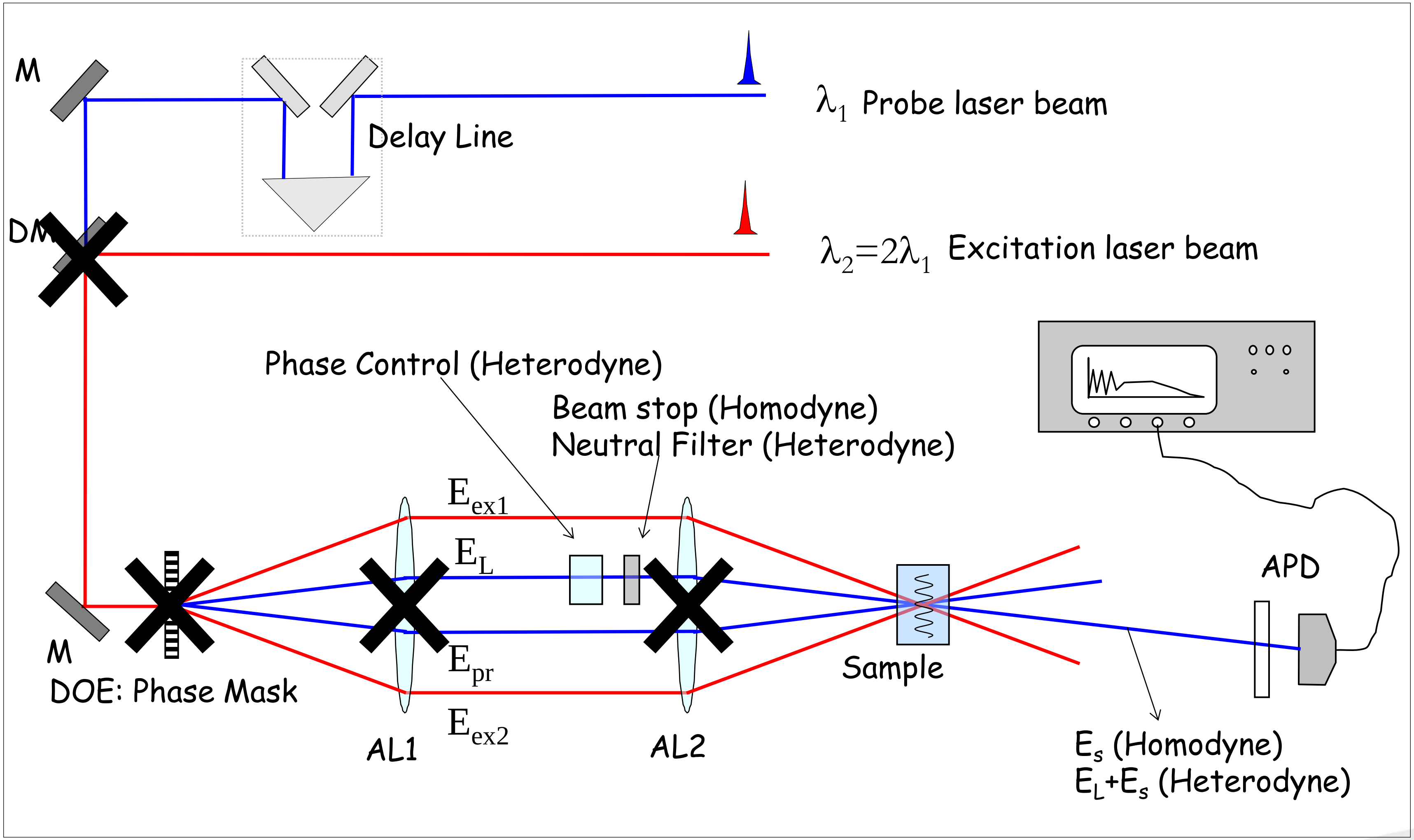






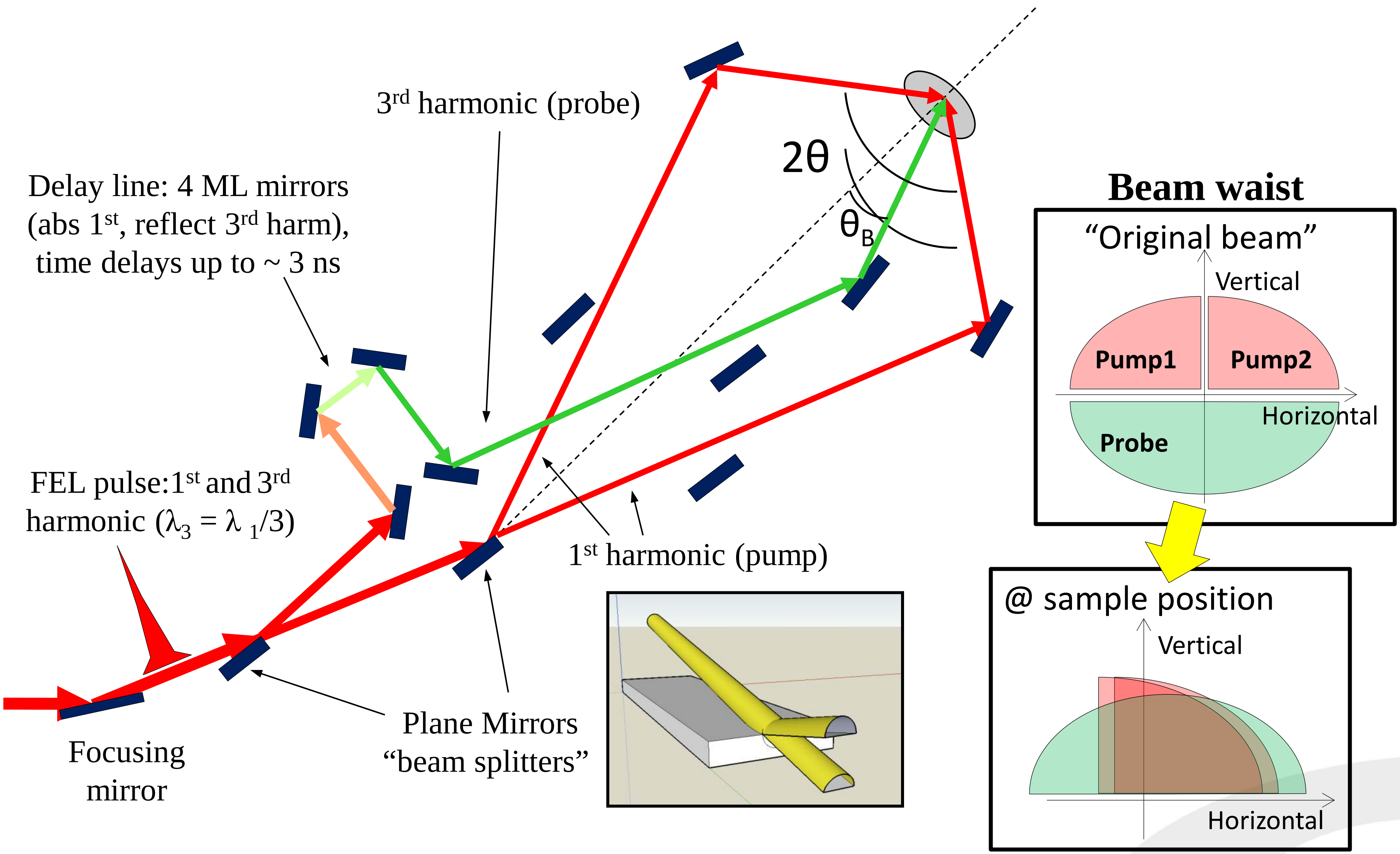
Gaussian-like time profile

Typical Infrared/Visible Set-Up



Challenge: Extend and modify the set-up for UV Transient Grating Experiments

TIMER Layout



R. Cucini et al et al., NIMA (2011)

TG @ EIS laser lab

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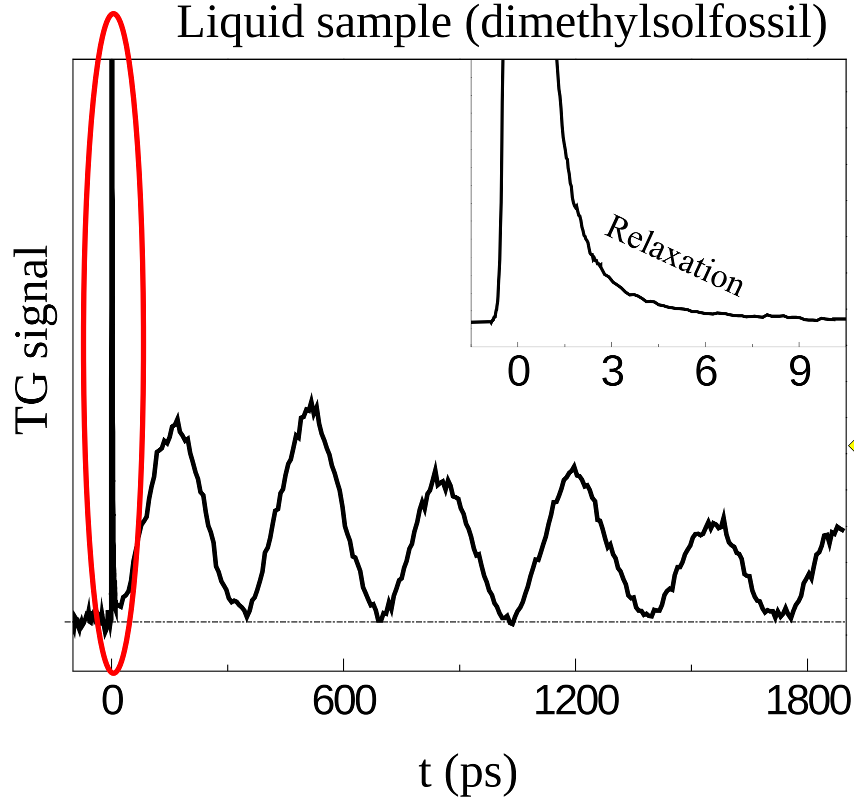
Liquid sample (dimethylsolfossil)



Pump energy ~ 25 μJ/pulse

Probe energy ~ 5 μJ/pulse

Beam dimensions ~ 50÷200 μm

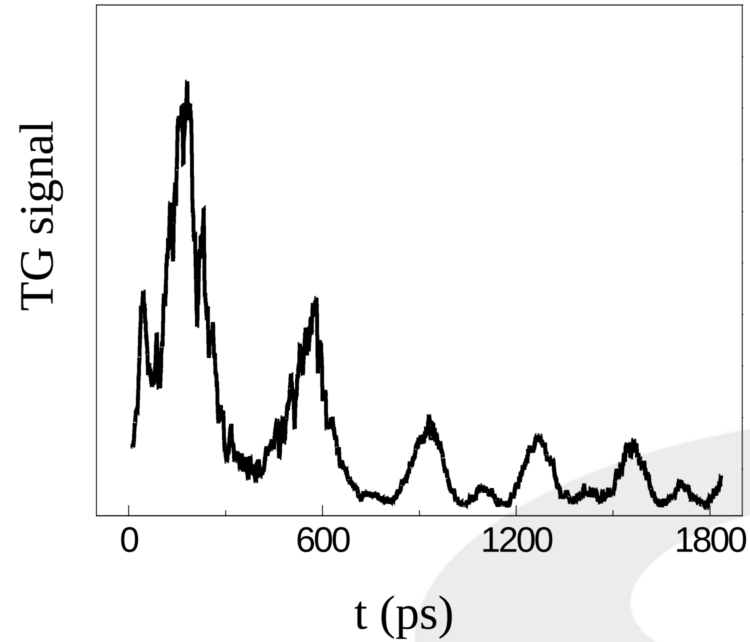
θ = 26°, Δt=150 fs, λ=800 nm

Signal ~ 0.1÷1 pJ → I_{signal}/I_{probe} ~10^{-7}

Signal ~ $[A \cdot \sin(\omega t) e^{-\Gamma_1 t} + B(1-\cos(\omega t)) e^{-\Gamma_2 t}]^2$

Sound velocity = 1486 +/- 2 m/s (OK)

Surface (200 nm Au on Si)



F. Bencivenga et al., NIMA (2010), R. Cucini et al., NIMA (2011), R. Cucini et al., Opt. Lett. (2011), R. Cucini et al., Opt. Lett. (2014)

Expected TG signal

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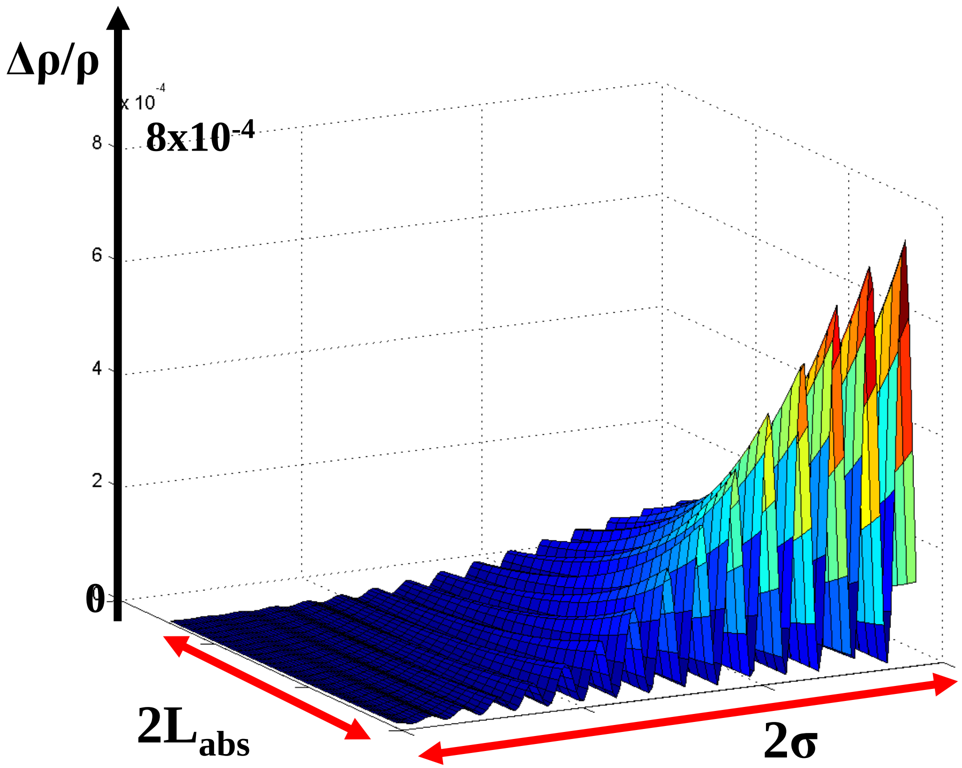
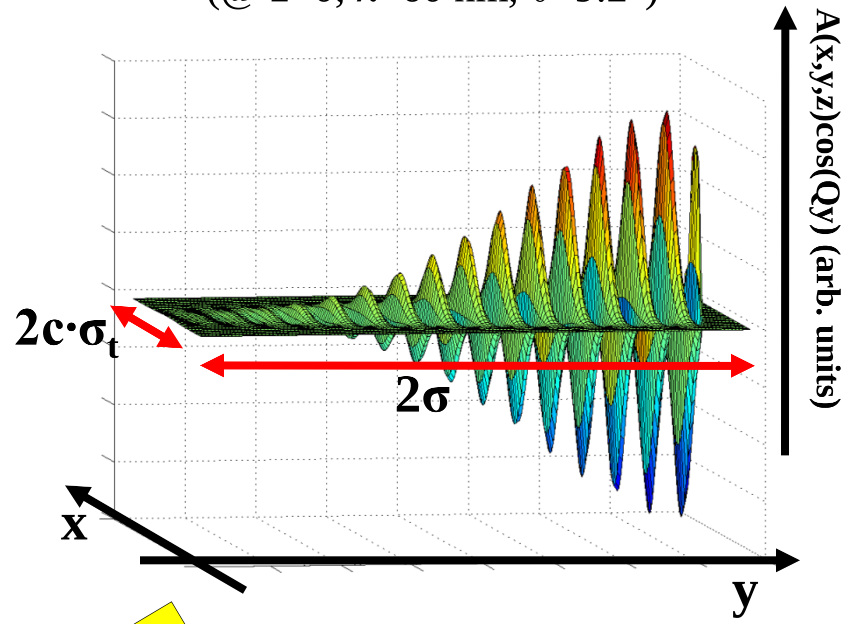
SiO_2 sample

Relative density variation
@ z=0, considering only optical absorption



Interference @ sample position

(@ z=0, λ=60 nm, θ=9.2°)



840 J kg^{-1} K^{-1}

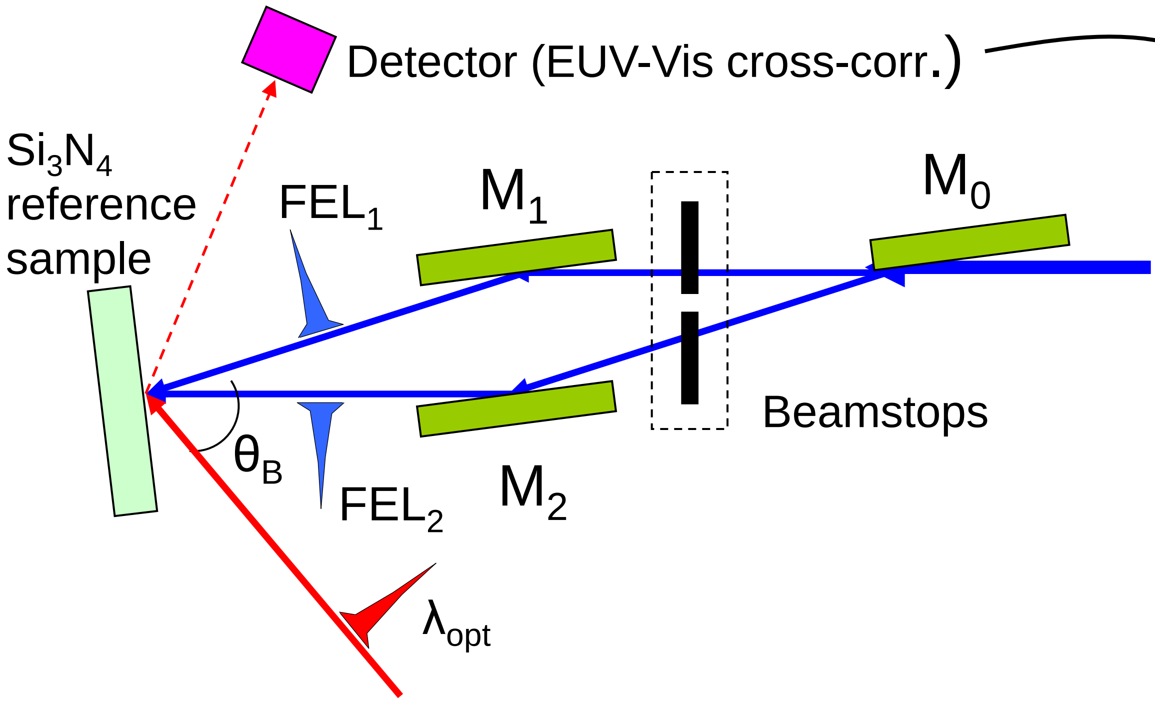
$$\Delta\rho/\rho(x,y,z) \sim \alpha\Delta E(x,y,x)/(\Delta V c_v \rho)$$

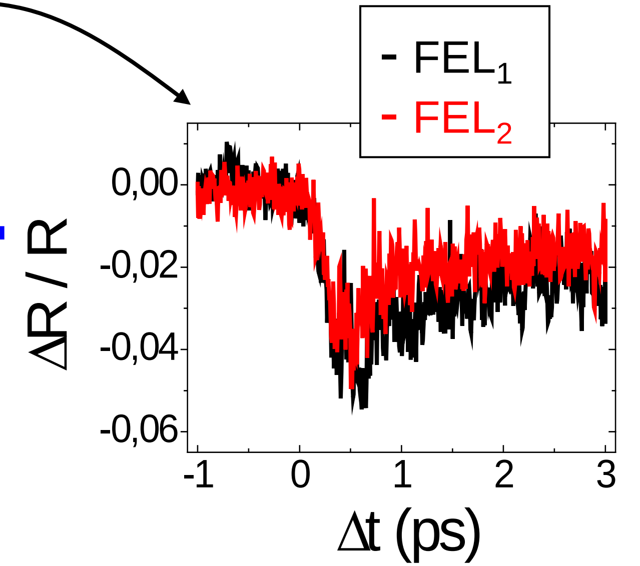
25.5·10^{-6} K^{-1}

2200 kg m^{-3}

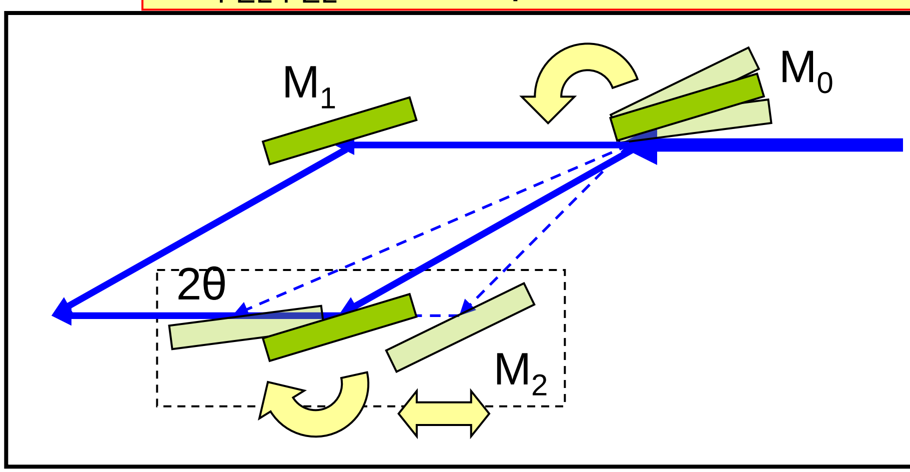
Adiabatic heating of the sample in the illuminated region < 3 °C

Transient Grating Experiments on V-SiO_2



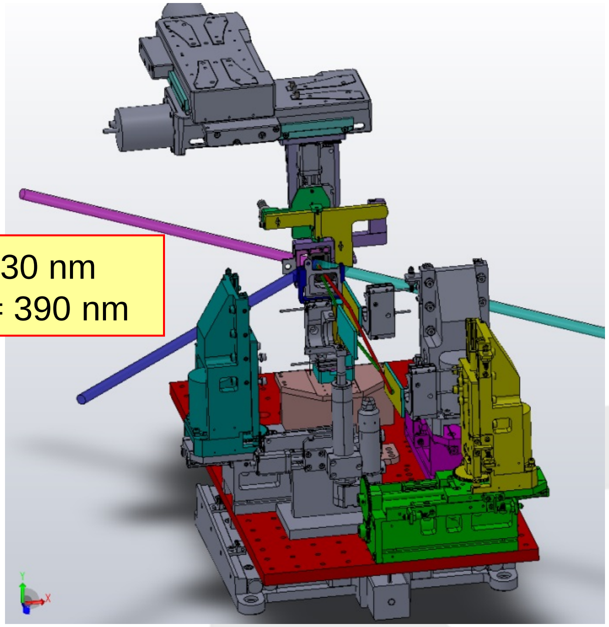


$\Delta t_{FEL\text{-}FEL} = \pm\ 0.5$ ps at 2θ = constant



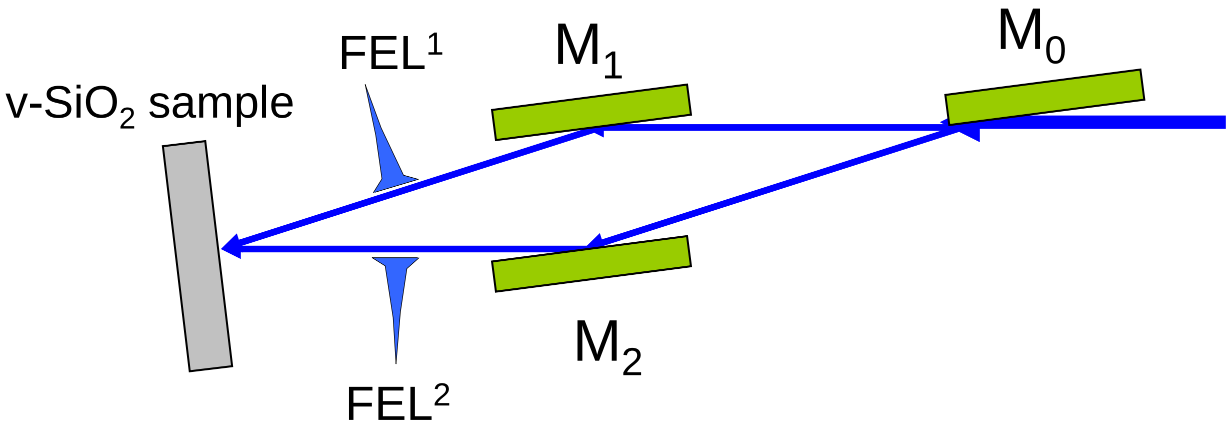
$\theta = 3^o$; $\lambda_{FEL} \approx 30$ nm

$\theta_B \approx 45^o$; $\lambda_{laser} = 390$ nm



Transient Grating Experiments on V-SiO_2

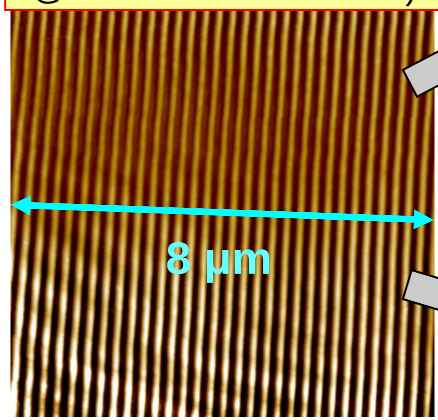
Elettra Sincrotrone Trieste

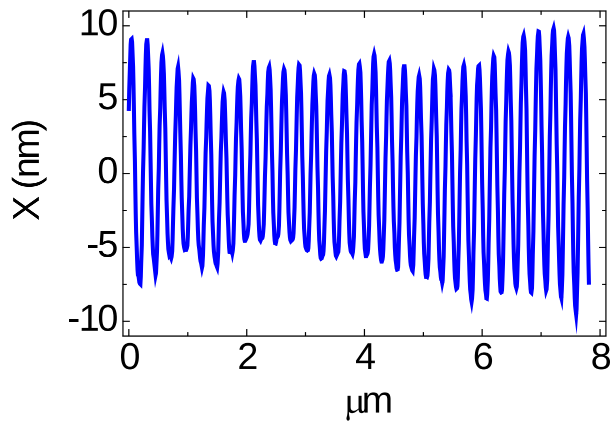


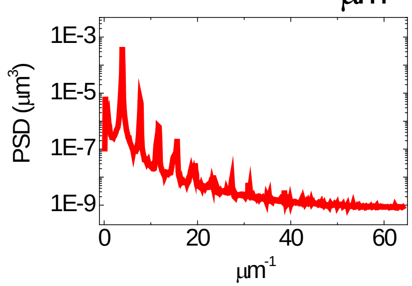
Inprints on SiO_2
→ $2\theta = 5.975^o$

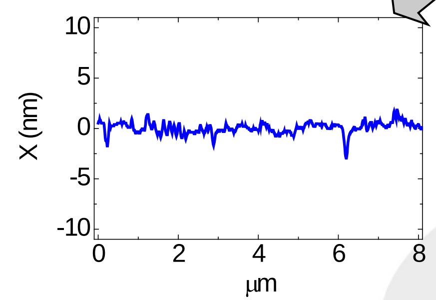
Grating visibility after multi-shot explosure
→ FEL^1-FEL^2 optical path difference < λ_{FEL}

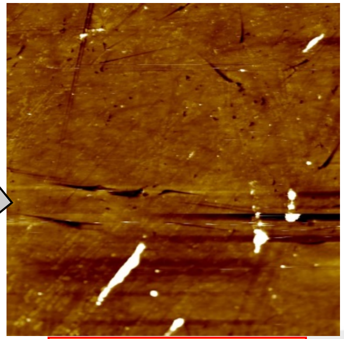
Permanent gratings on SiO_2 (after 1000's shots @ FEL flux > 50 mJ/cm^2)











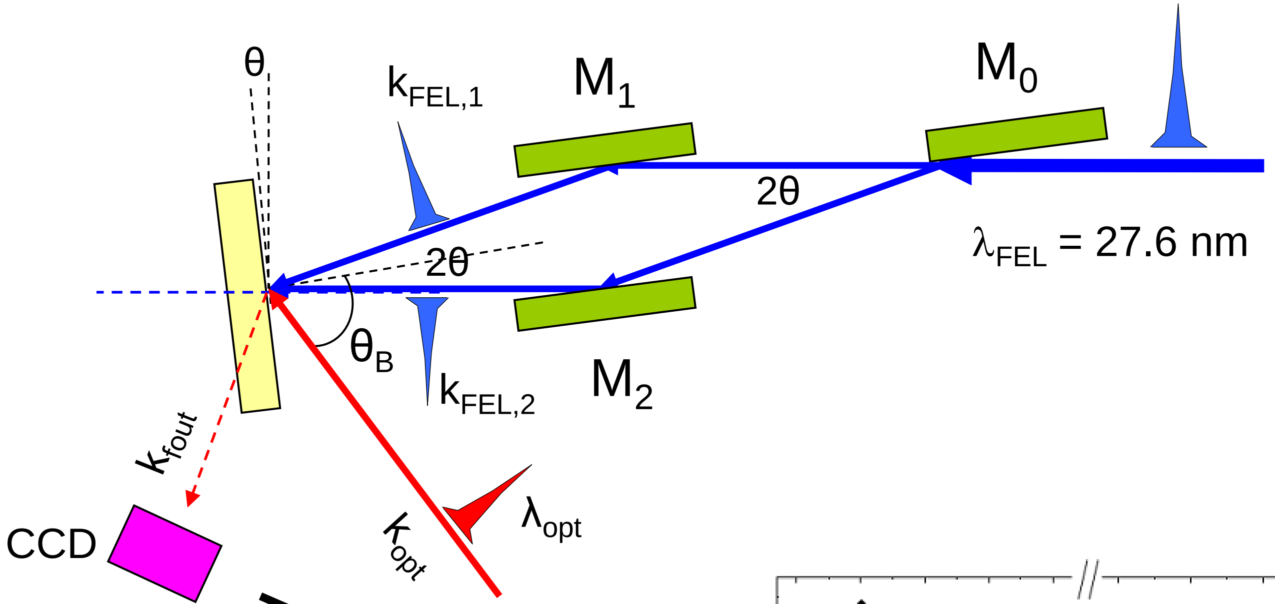
Clean sample

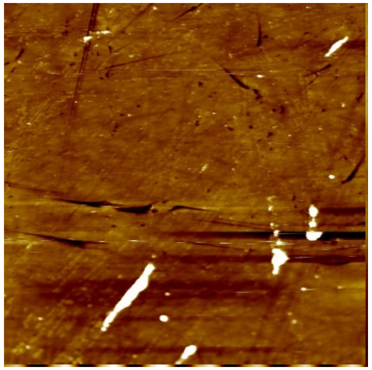
doi:10.1038/nature1434

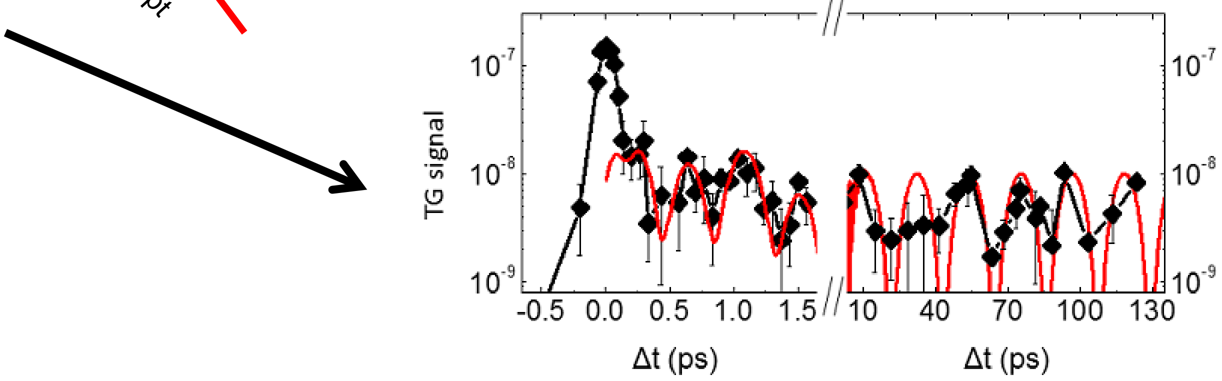
Elettra Sincrotrone Trieste

Four-wave mixing experiments with extreme ultraviolet transient gratings

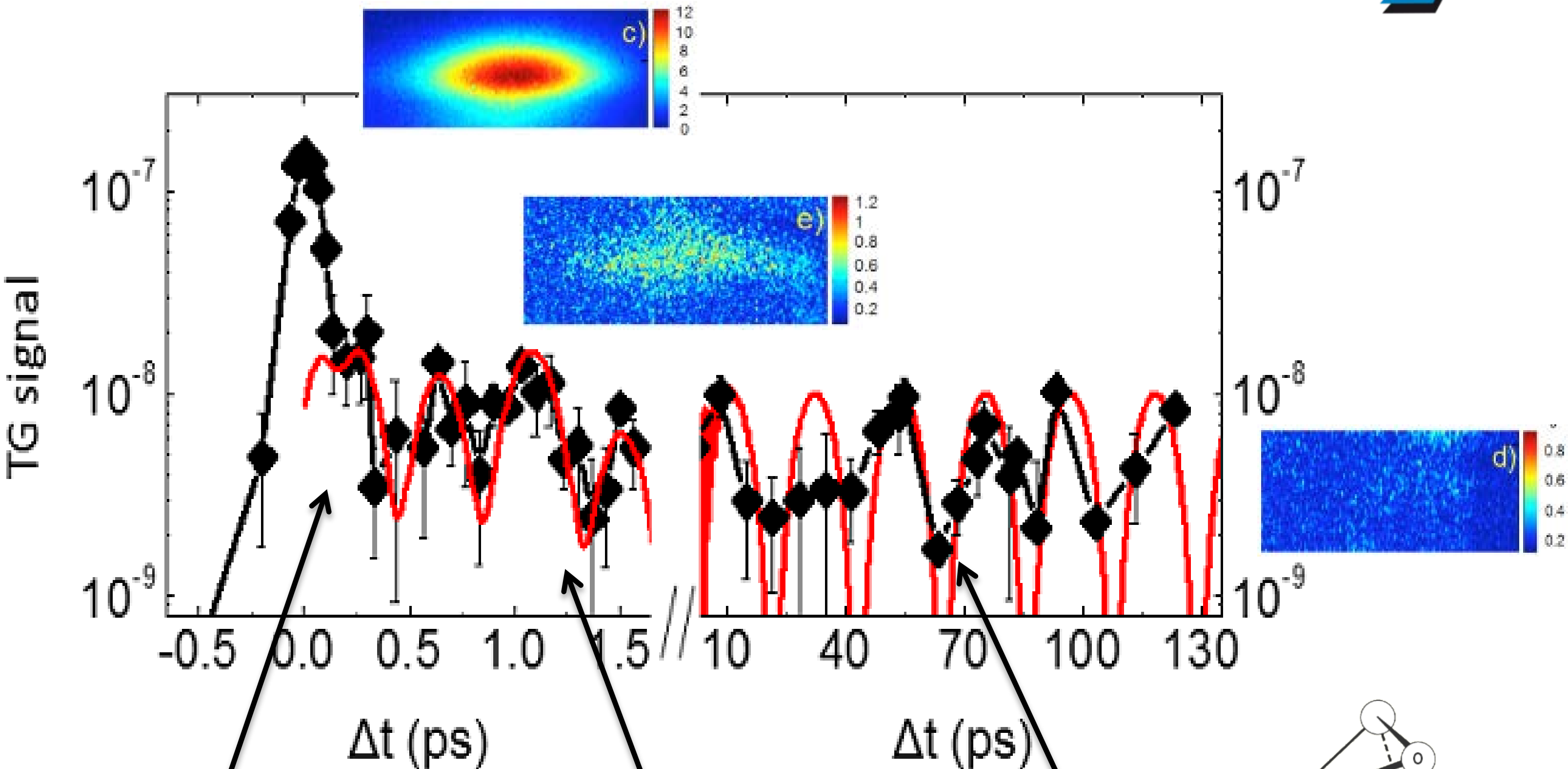
F. Bencivenga et al., Nature 2015

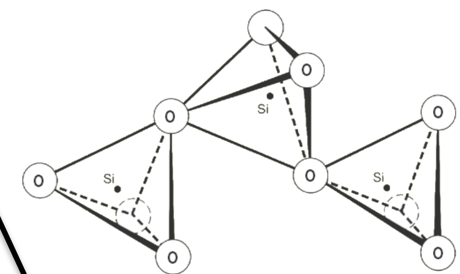






Transient Grating Experiments on V-SiO_2





Hyper - Raman modes due to coupled tetrahedral rotations $\nu_2 \approx 4.1$ THz

Raman modes due to tetrahedral bending $\nu_1 \approx 1.2$ THz

Acoustic-like excitations

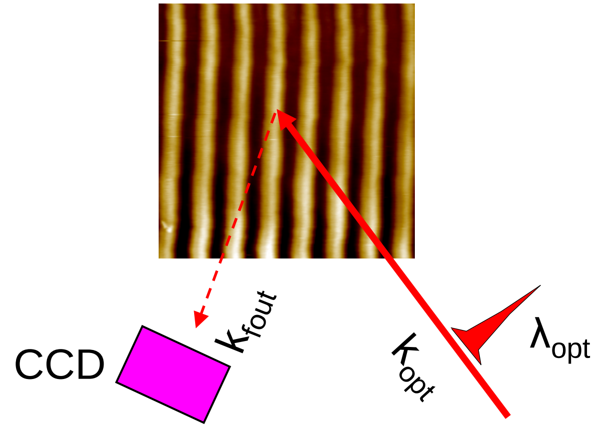
Heterodyning with FEL

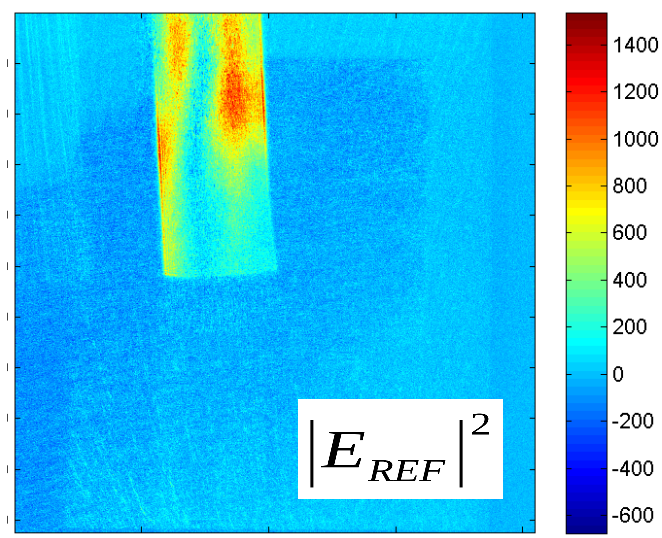
Elettra Sincrotrone Trieste

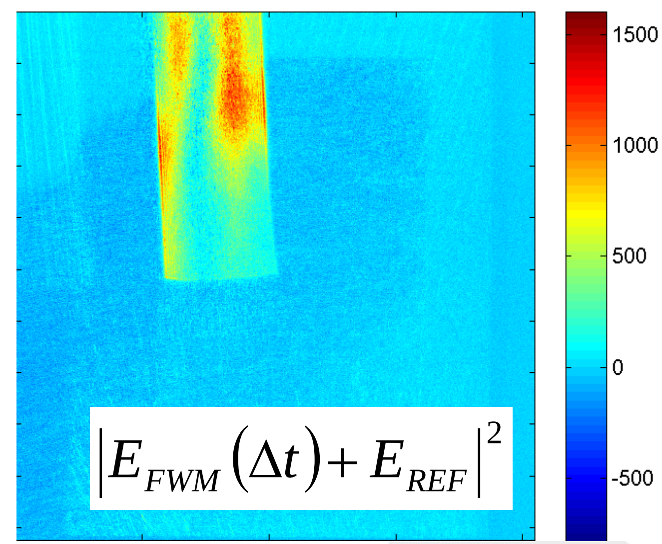
Heterodyning is a signal processing technique invented in 1901 by R. Fessenden

$$I = |E_{\mathrm{ref}}|^2 + |E_{\mathrm{s}}(t)|^2 + 2|E_{\mathrm{ref}} E_{\mathrm{s}}(t)|\cos(\phi_{\mathrm{ref}} - \phi_{\mathrm{s}})$$

Time independent local field

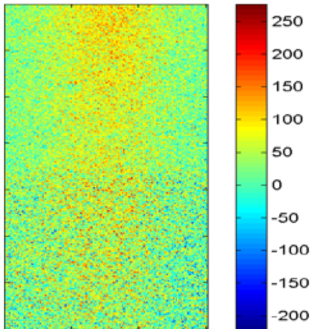






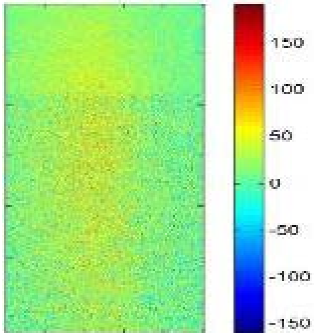
Heterodyning with FEL

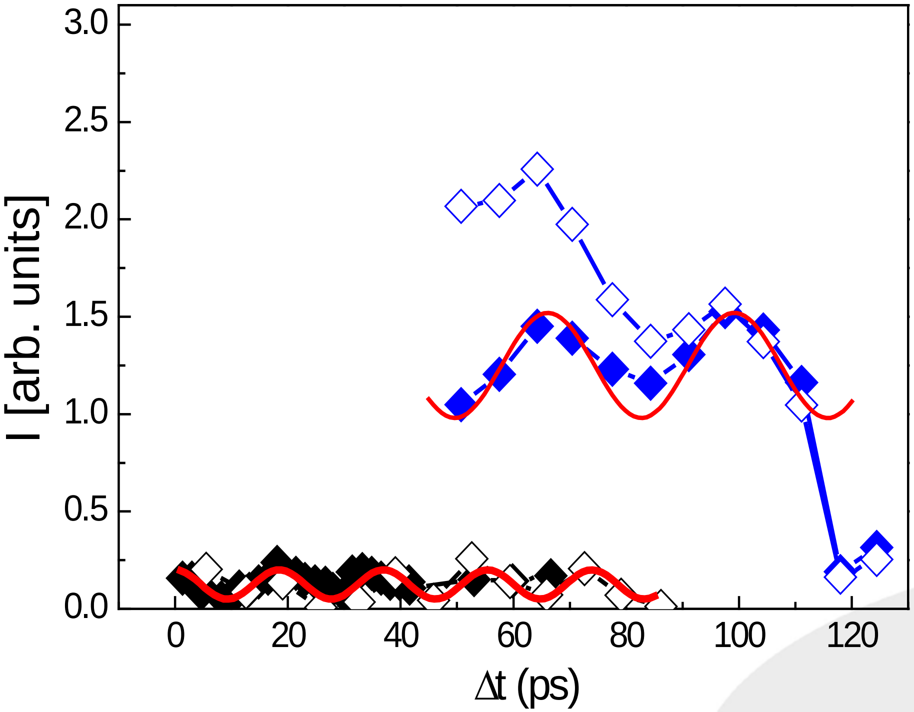
Heterodyne



VS

Homodyne





Clear intensity **increase !!!**

Nonlinear Optics

Elettra Sincrotrone Trieste

N. Bloembergen 1981

$$P_i = \chi_{ij}^{(1)} E_i + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \cdots$$

is the lowest nonlinear order for centrosymmetric materials →
→ **all materials** have a third-order nonlinear response.

FOUR-WAVE MIXING SPECTROSCOPY

The nonlinearity $\chi^{(3)}$ describes a coupling between four light waves, and some

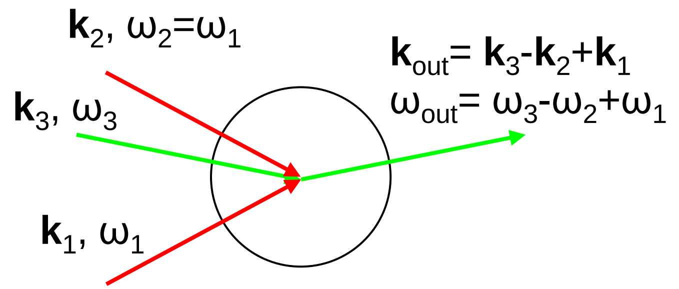
N. Bloembergen Nobel Lecture 1981

Phase Matching

Clean technique

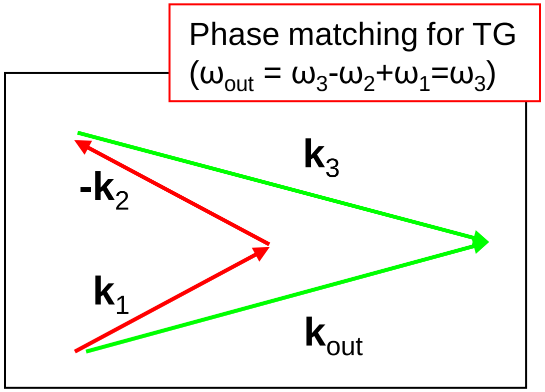
$$I_{FWM} \sim \chi^{(3)} I_1 I_2 I_3 \cdot \text{sinc}(\Delta \mathbf{k}_{FWM} \cdot L/2)$$

$$\Delta \mathbf{k}_{FWM} = \mathbf{k} - \mathbf{k}_{out}$$



Phase matching → non linear emission from N elementary emitters placed at different sample locations within $2\delta k^{-1}$ (coherence length of the non-linear process) **adds in phase** (intensity grows up as N^2) **along k_{out}**

→ $\delta k > 0$ because, e.g., bandwidth and divergence

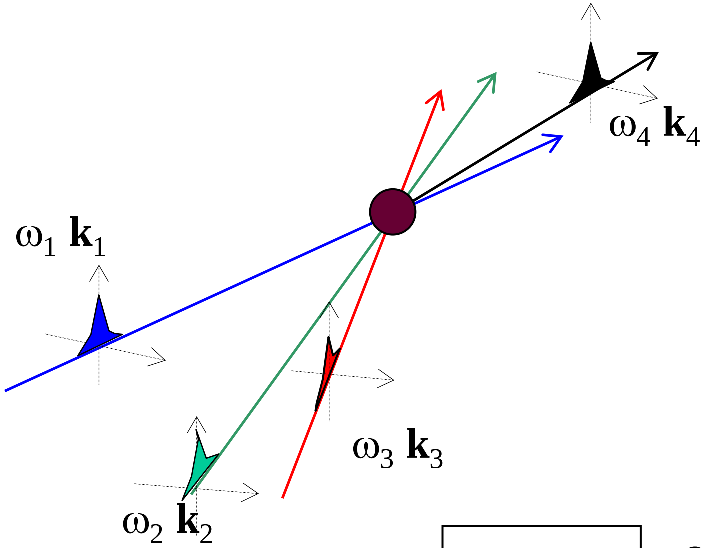


Directionality + **coherent addition** may lead to a dominating non linear signal

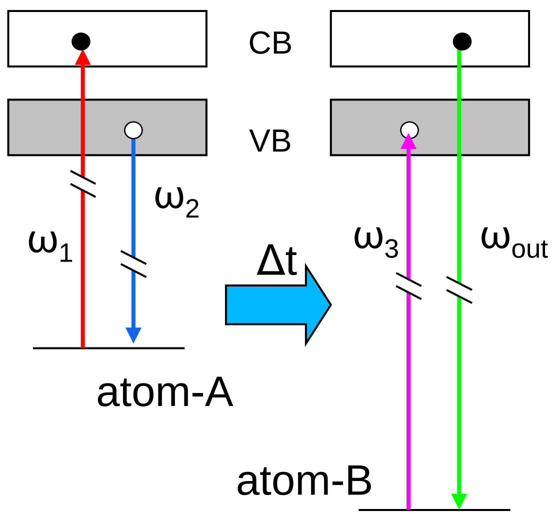
FERMI: pulses with Fourier-limited bandwidth → increase in δk^{-1} → substantial ($\sim N^2$) increase of I_{FWM} along k_{out}

Four Wave Mixing at FEL's

Elettra Sincrotrone Trieste



Four Wave Mixing techniques are: Stimulated Raman Gain Spectroscopy, Photon Echo and Raman Induced Kerr Effect Spectroscopy, Femtosecond Stimulated Raman Scattering and Coherent Antistokes Raman Scattering (**CARS**)

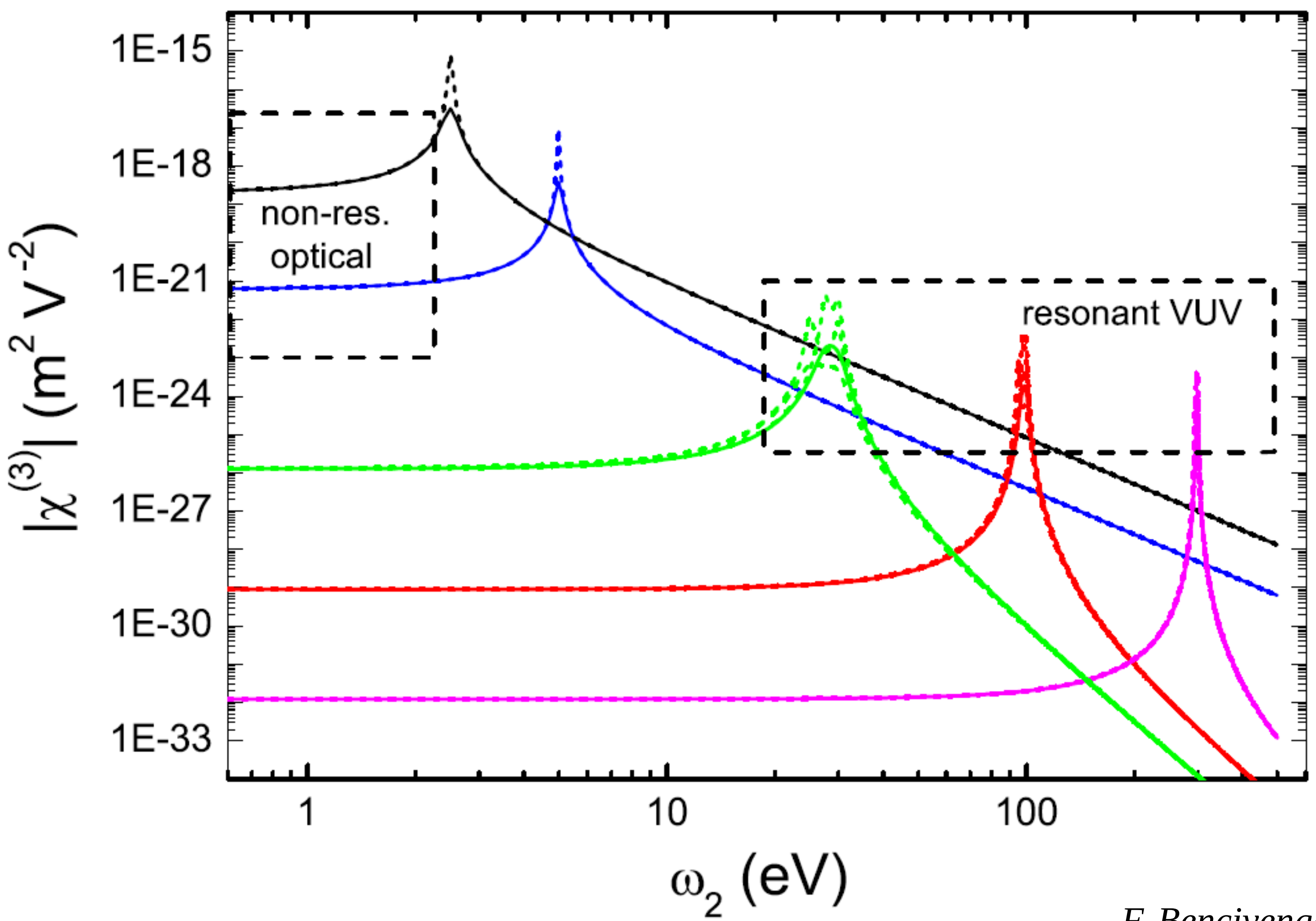


Measure the coherence between the two different sites → tuning energies and time delay makes possible to chose where a given excitation is created, as well as where and when it is probed

delocalization of electronic states and charge/energy transfer processes.

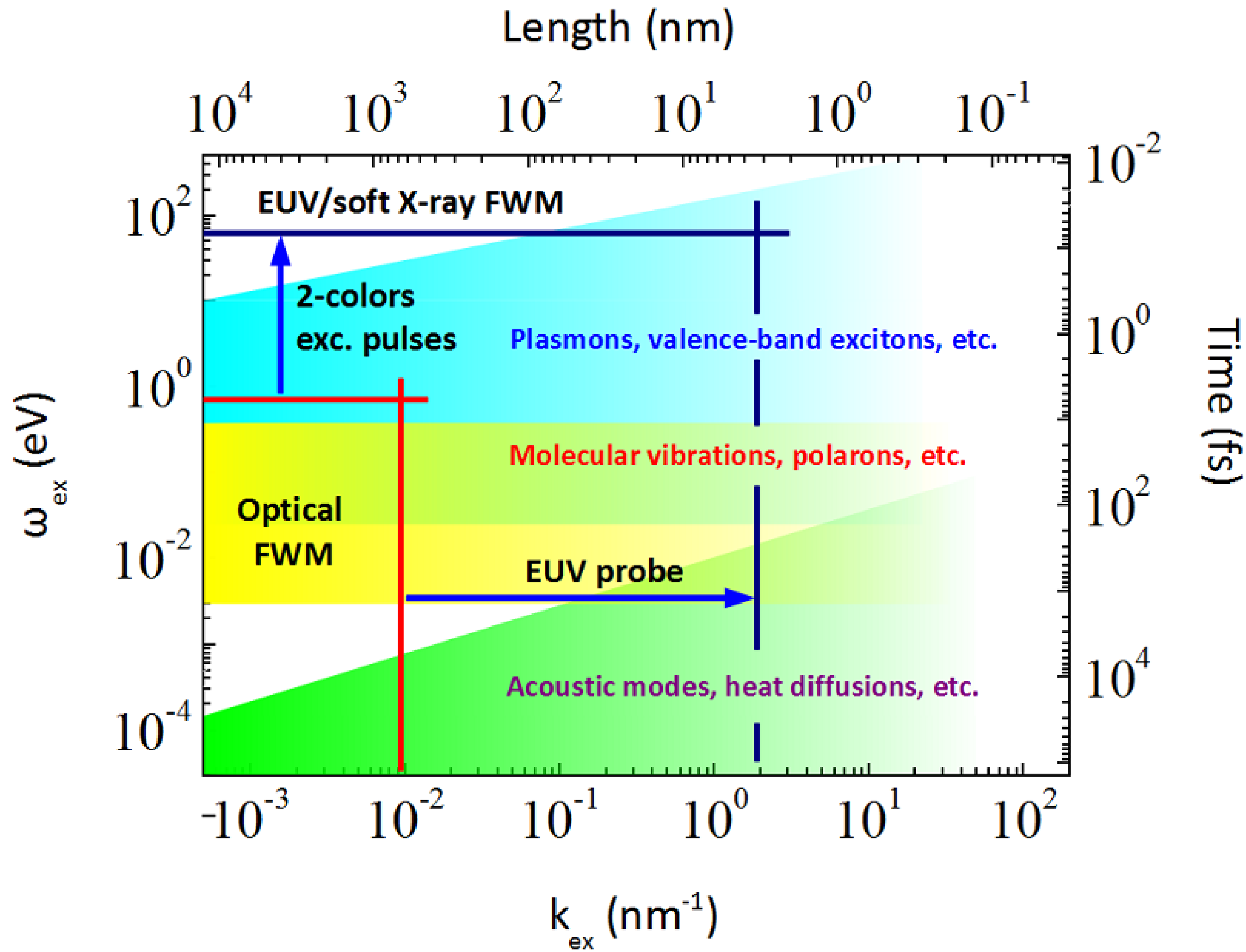
S. Tanaka and S Mukamel, PRL (2002)

Four Wave Mixing at FEL's



F. Bencivenga et al., New. J. Phys. 2013

Energy - Momentum transfer region "Invasion"

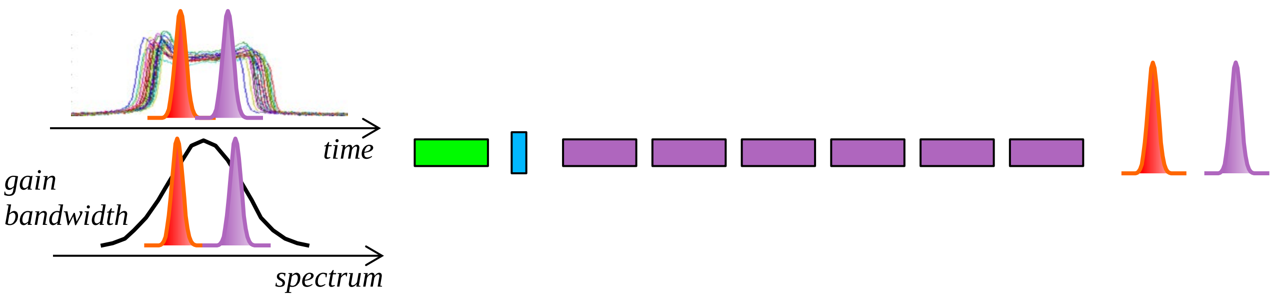


F. Bencivenga et al., Adv. In Phys. 2015

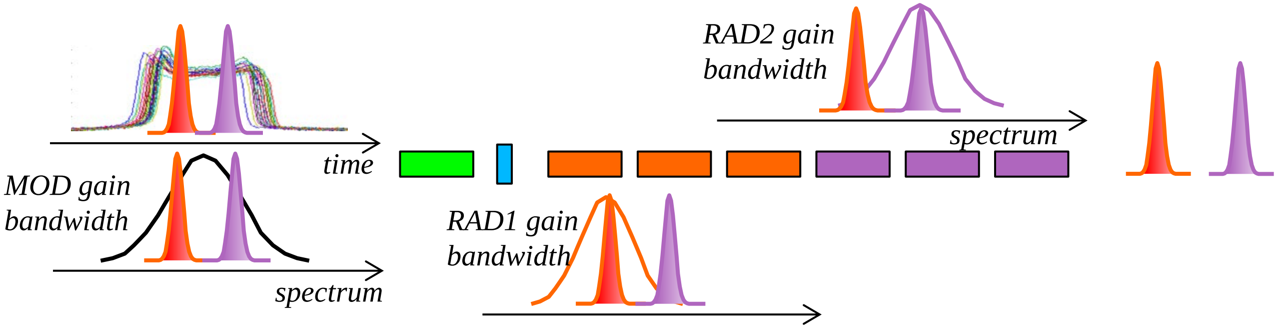
Multiple pulse configurations

Elettra Sincrotrone Trieste

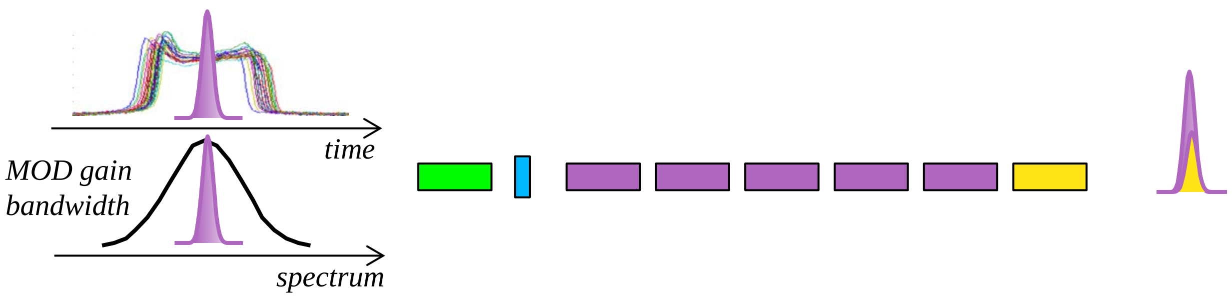
Multiple pulses can be generated by **double pulse seeding**
Temporal separation between 25-300 and 700-800 fs.
Shorter separations are accessible via FEL pulse splitting. *Mahieu et al. Optics Express (2013)*
Larger separations require the **split & delay line**.



Spectral separation 0.4-0.7%
(*E. Allaria et al., Nat. Comm 2013*)



Spectral separation 2-3%
or much larger if the two radiators are tuned at different harmonics
(Sacchi et al., in preparation)



Two (almost) temporally superimposed pulses at harmonic wavelengths of the seed. They are correlated in phase that can be controlled with the phase shifter
(K. Prince et al., submitted)

Multicolor at FEL's

Elettra Sincrotrone Trieste

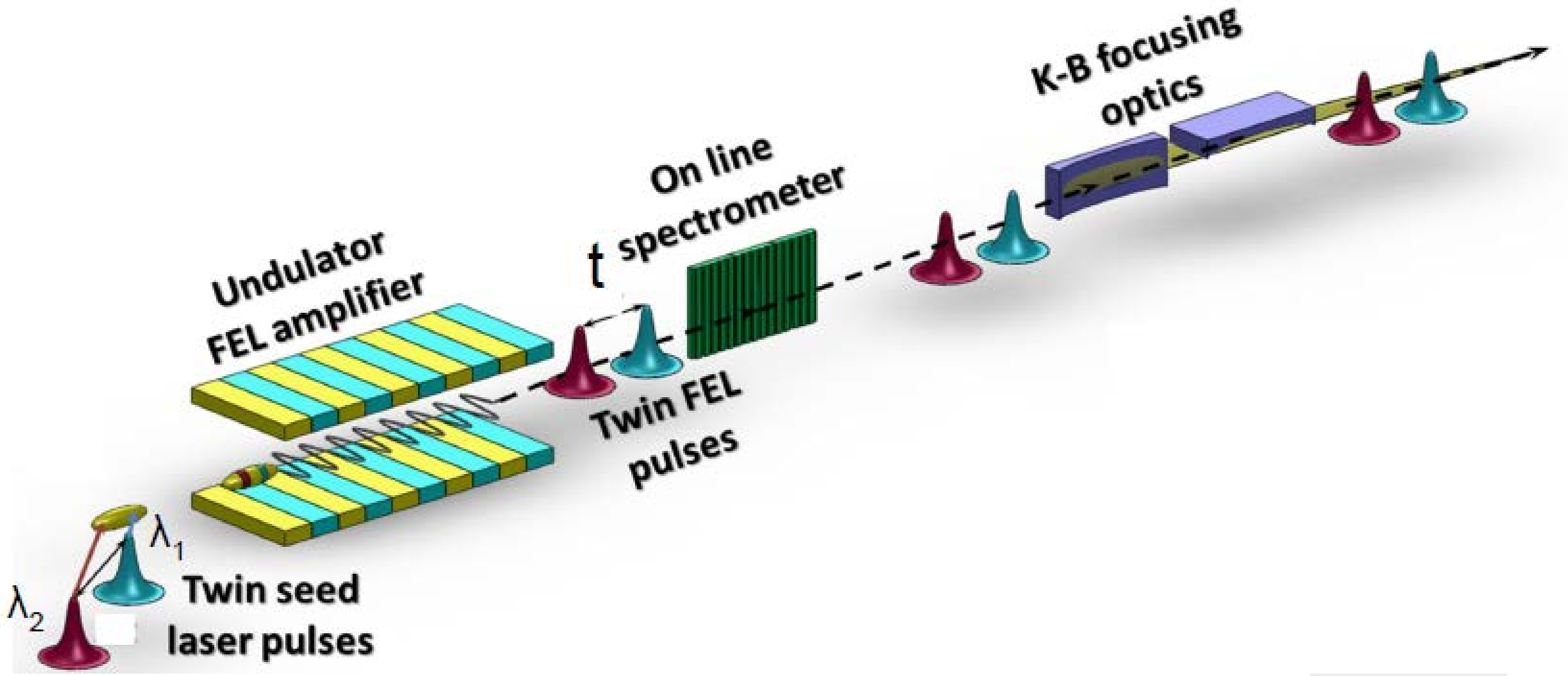
nature COMMUNICATIONS

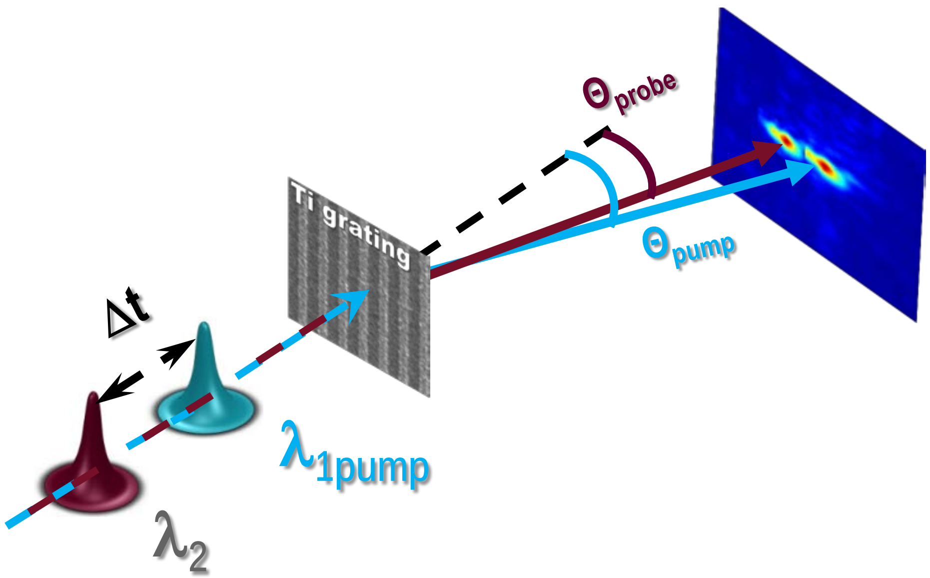
Received 24 May 2013 | Accepted 21 Aug 2013 | Published 18 Sep 2013

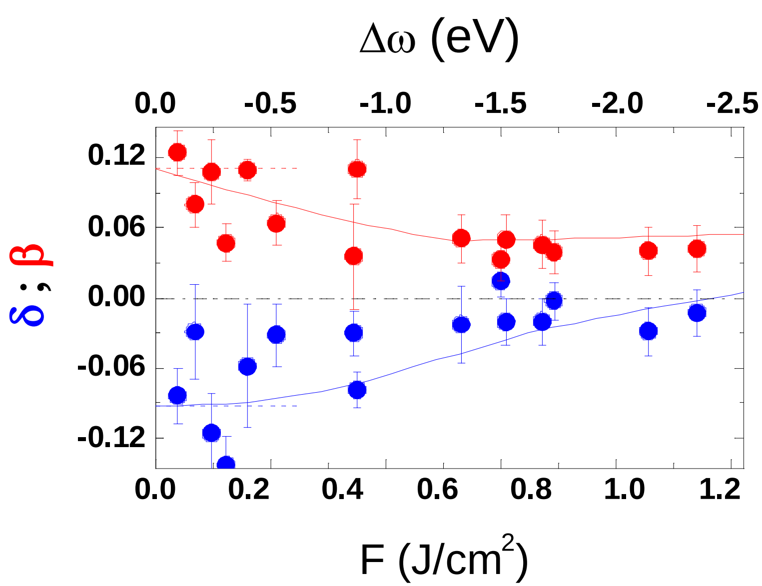
DOI: 10.1038/ncomms3476

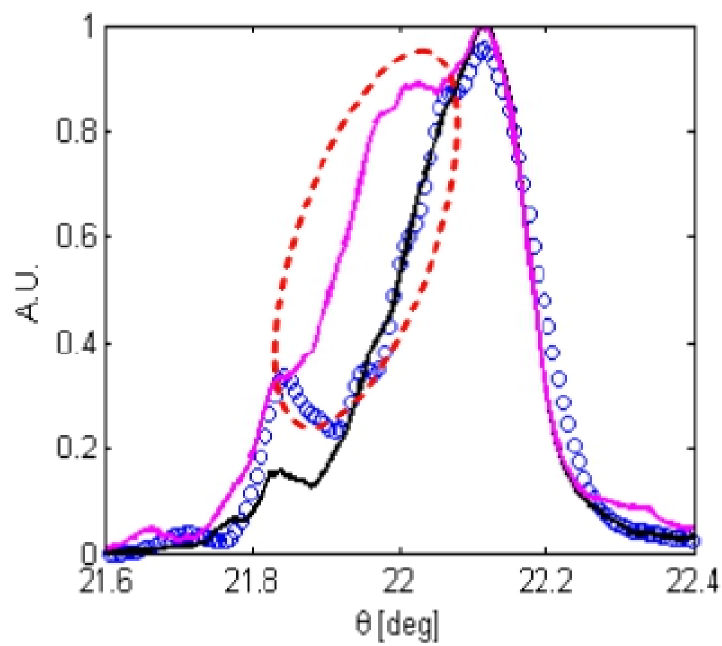
OPEN

Two-colour pump-probe experiments with a twin-pulse-seed extreme ultraviolet free-electron laser



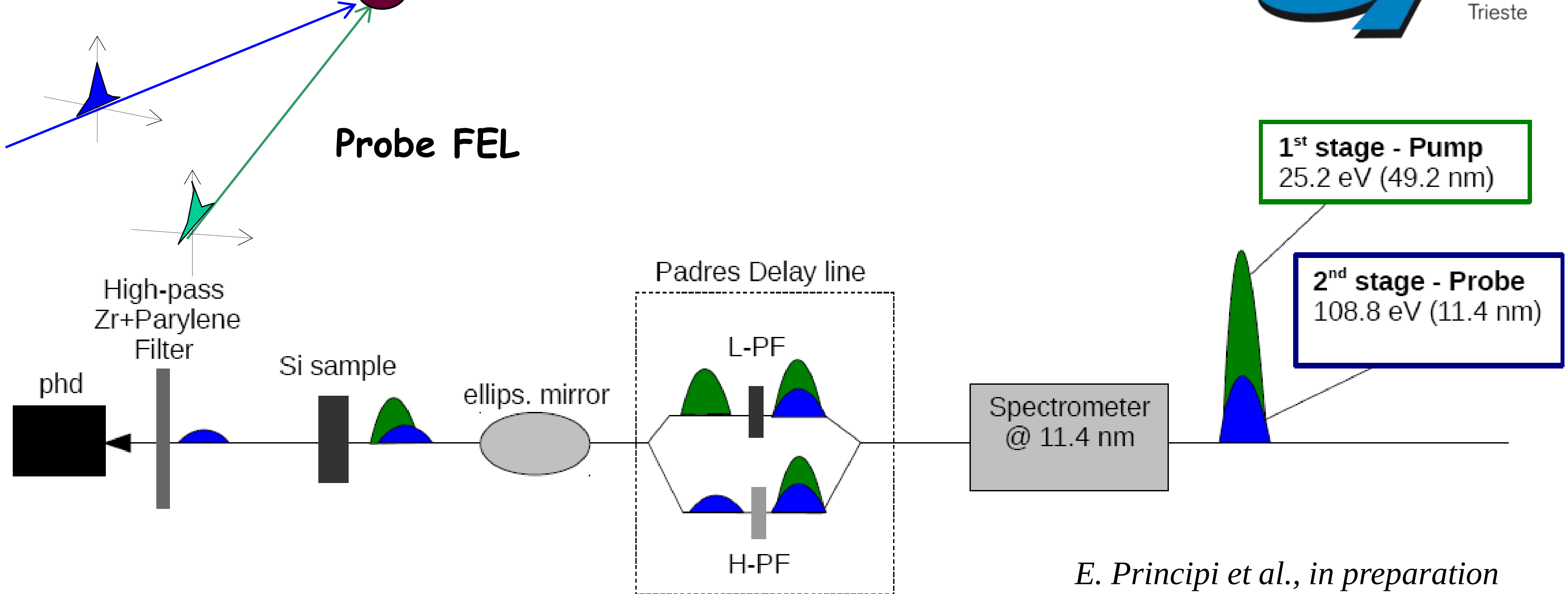




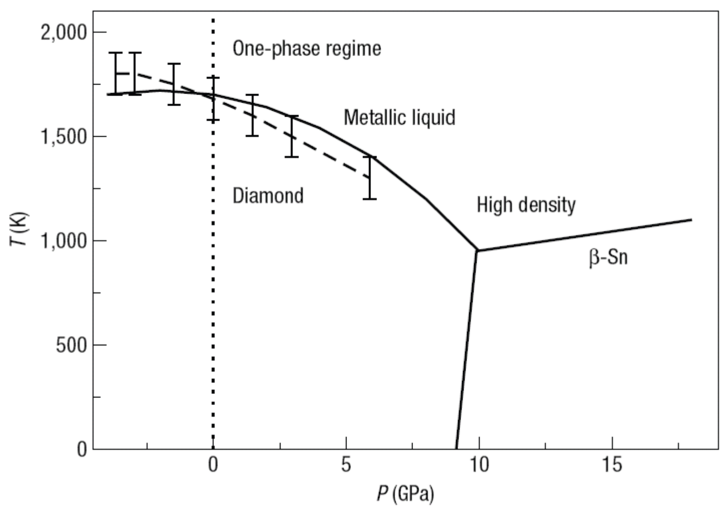


F. Bencivenga et al., Farad. Discussion (2014)

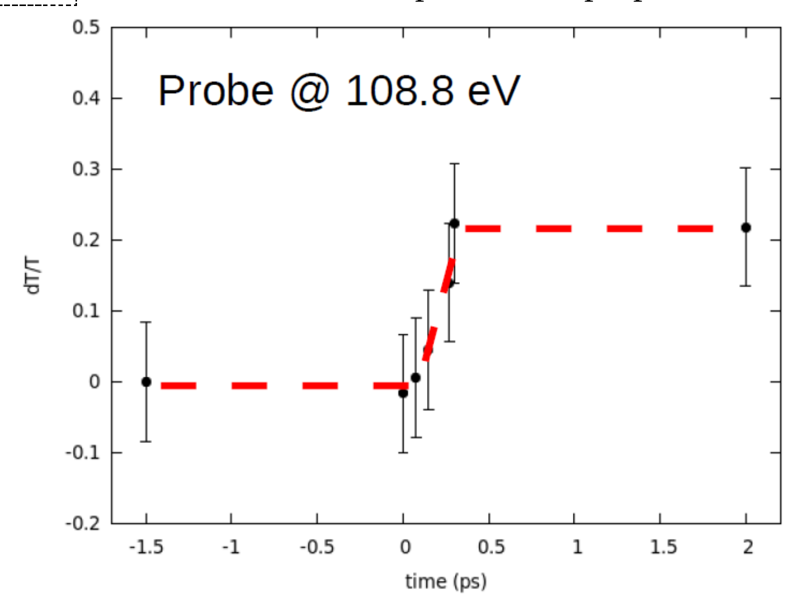
Pump & Probe on Silicon



E. Principi et al., in preparation



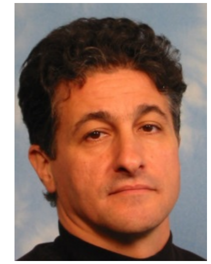
P. F. McMillan et al., Nature (2005)



Conclusions

T. Scopigno

K. Nelson



M. Chergui

- Charge transfer dynamics in **metal complexes**
- Charge injection and transport in **metal oxides nanoparticles**
- Vibrational modes in **Glasses**
- Charge **Density Wave**
- Quasiparticle diffusion (**Polarons**)
- Sound velocity in **Graphene**

G. Knopp

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G. Monaco

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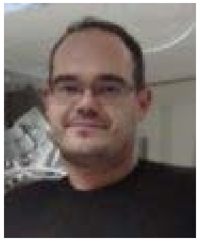


F. Capotondi

A. Gessini

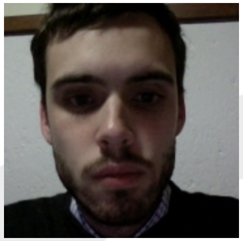
E. Pedersoli

R. Cucini

E. Giangrisostomi

F. Bencivenga

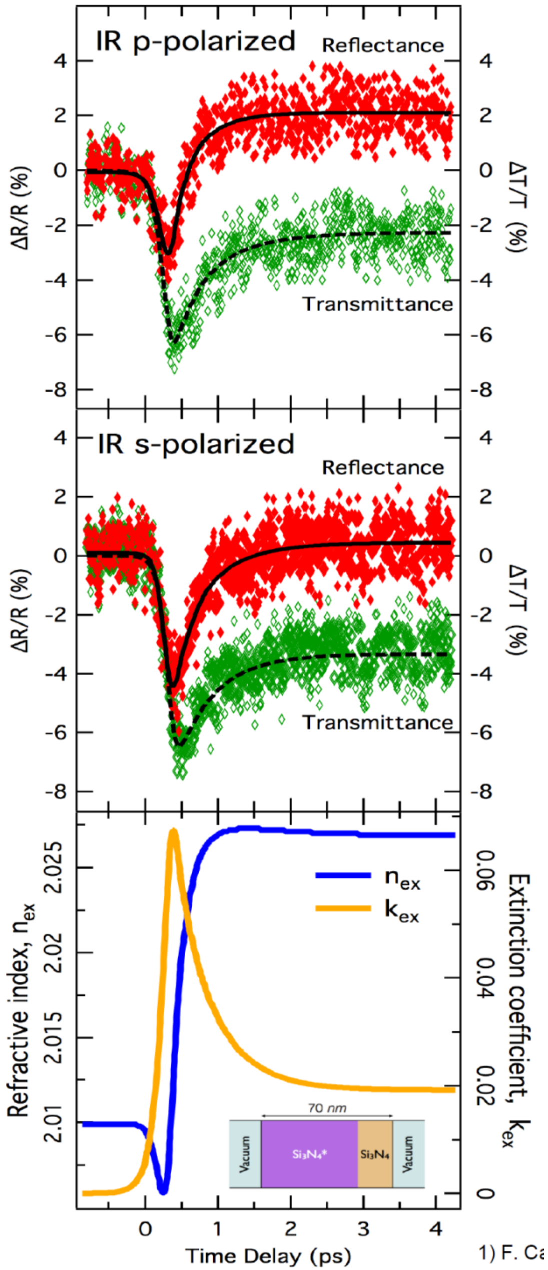
E. Principi

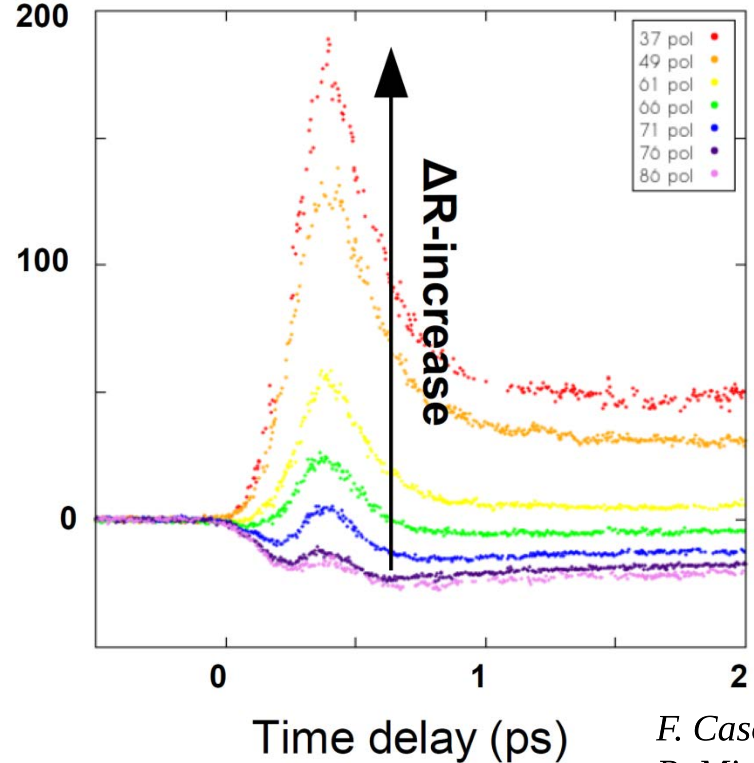


R. Mincigrucci

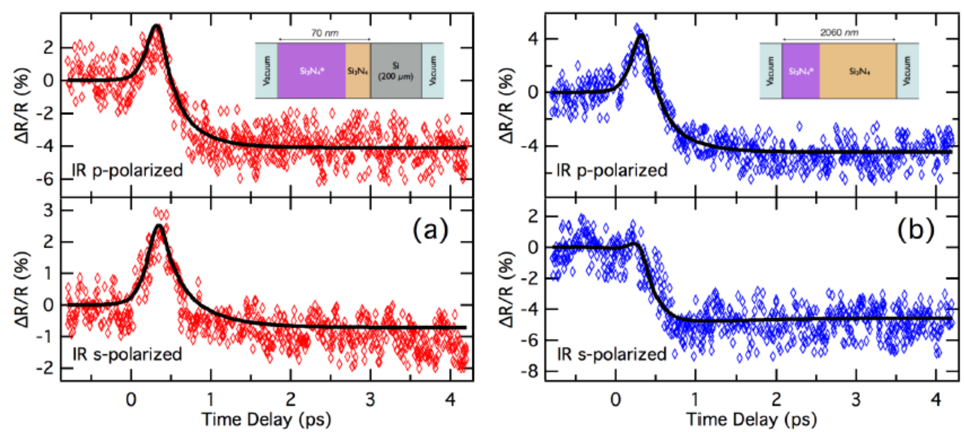
SiN reflectivity

Elettra
Sincrotrone
Trieste





F. Casolari et al., APL (2014)
R. Mincigrucci et al., in preparation



TG @ EIS laser lab