Multicolor Experiment with FEL: FWM

C. Masciovecchio
Elettra - Sincrotrone Trieste, Basovizza, Trieste I-34149

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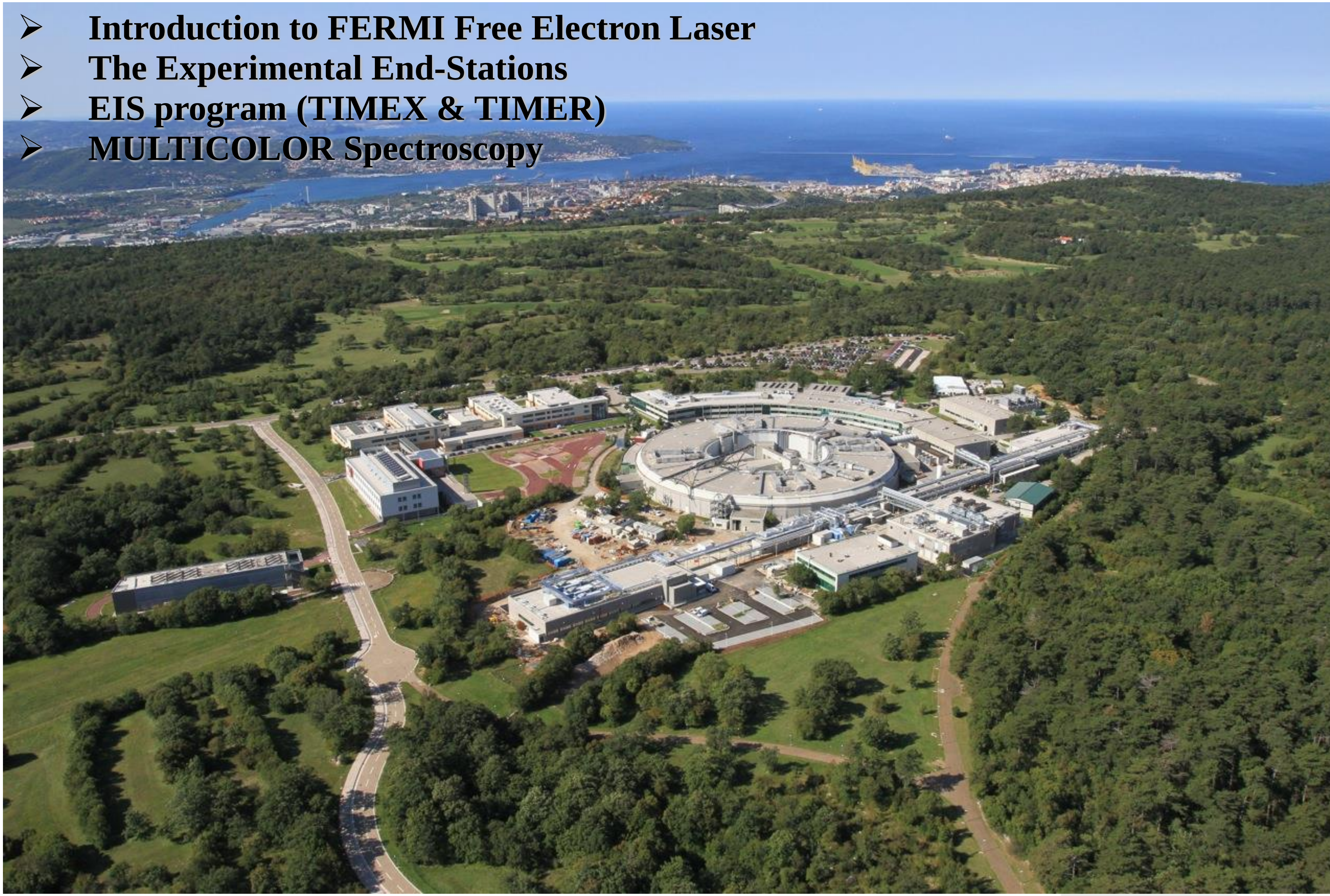

- **Introduction to FERMI Free Electron Laser**
- **The Experimental End-Stations**
- **EIS program (TIMEX & TIMER)**
- **MULTICOLOR Spectroscopy**

Why Free Electron Lasers?

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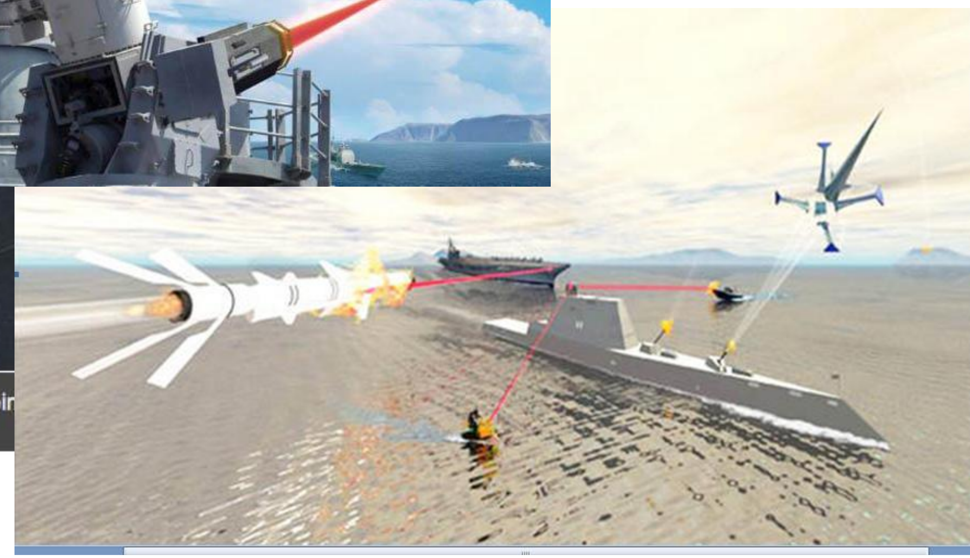
MILITARY

Boeing to develop Free Electron Laser for US Navy

By David Greig
05:42 April 21, 2009 PDT

Boeing to develop Free Electron Laser for US Navy (Photo: Frank Buck/Boein



“ Boeing has won a U.S. Navy contract worth up to $163 million to develop the Free Electron Laser (FEL), a weapon system that the company says "will transform naval warfare in the next decade by providing an ultra-precise, speed-of-light capability and unlimited magazine depth to defend ships against new, challenging threats, such as hyper-velocity cruise missiles." The envisioned level of precision would enable U.S. Navy ships to deliver nonlethal or lethal force to targets with power and minimal collateral damage………

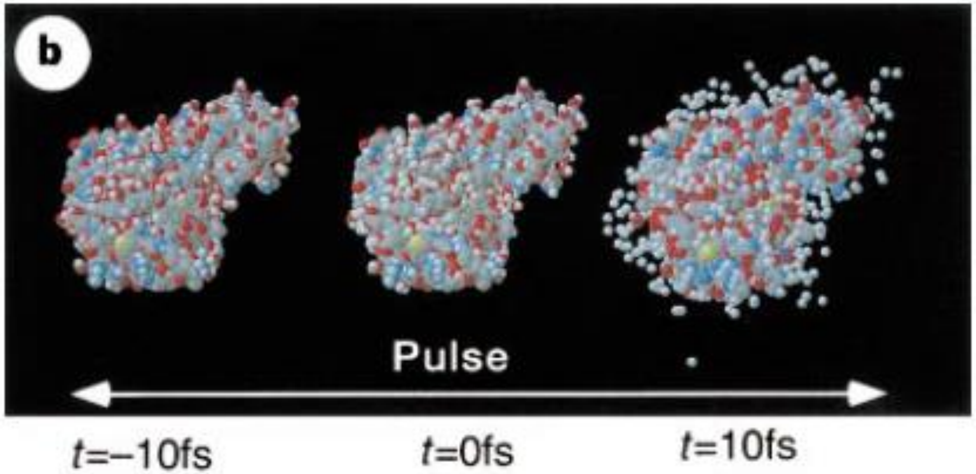
FEL technology is considered by the US Navy as a promising technology for an antimissile directed-energy weapon. A directed-energy weapon (DEW) is a type of weapon that emits energy in an aimed direction without the means of a projectile- like an intense emission of laser light. FEL’s are capable of achieving the megawatt power level the Navy requires for ship defense. “

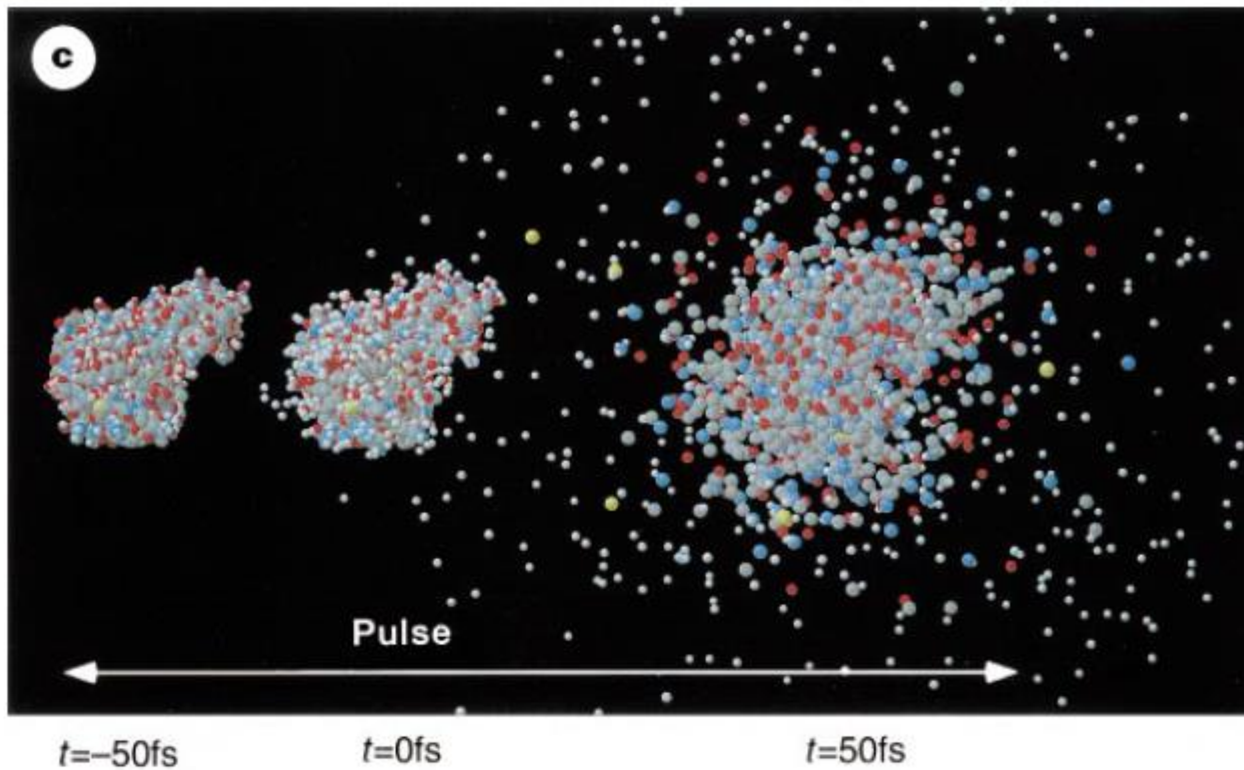
Why Free Electron Lasers?

Leonard comes home...and promptly passes out on the couch. The reason wasn't a good night on the town, however. It was the fact that he's working nights using the free-electron laser to conduct his X-ray diffraction experiment.

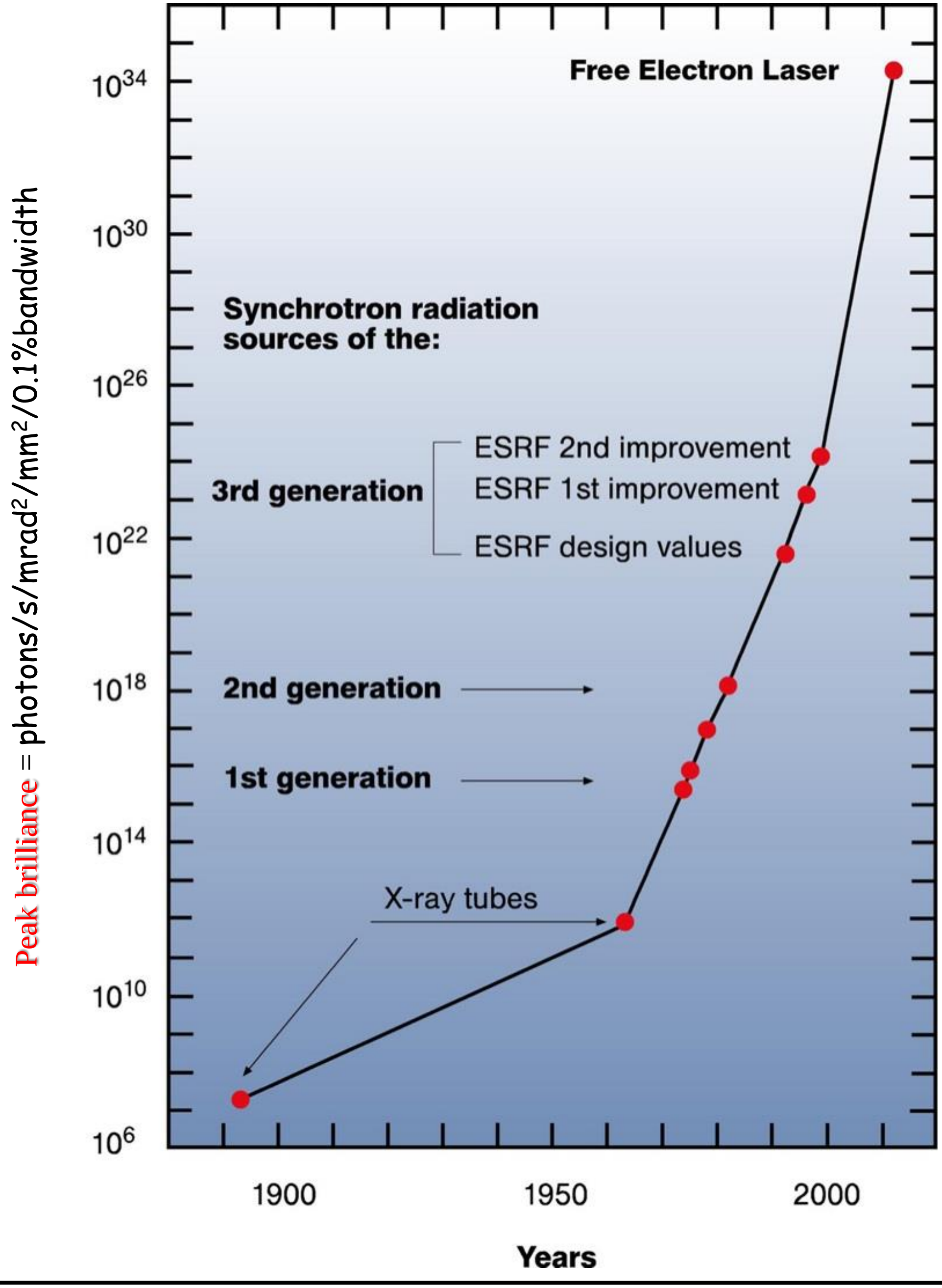
Why Free Electron Lasers ?

R. Neutze et al, Nature (2000)





Why Free Electron Lasers ?



Exercise 1

Calculate average flux at the undulator exit

elettra typical undulator working at 50 eV:

10^{15} photons/s/0.1%bandwidth

$\Delta\lambda/\lambda \sim n/N$

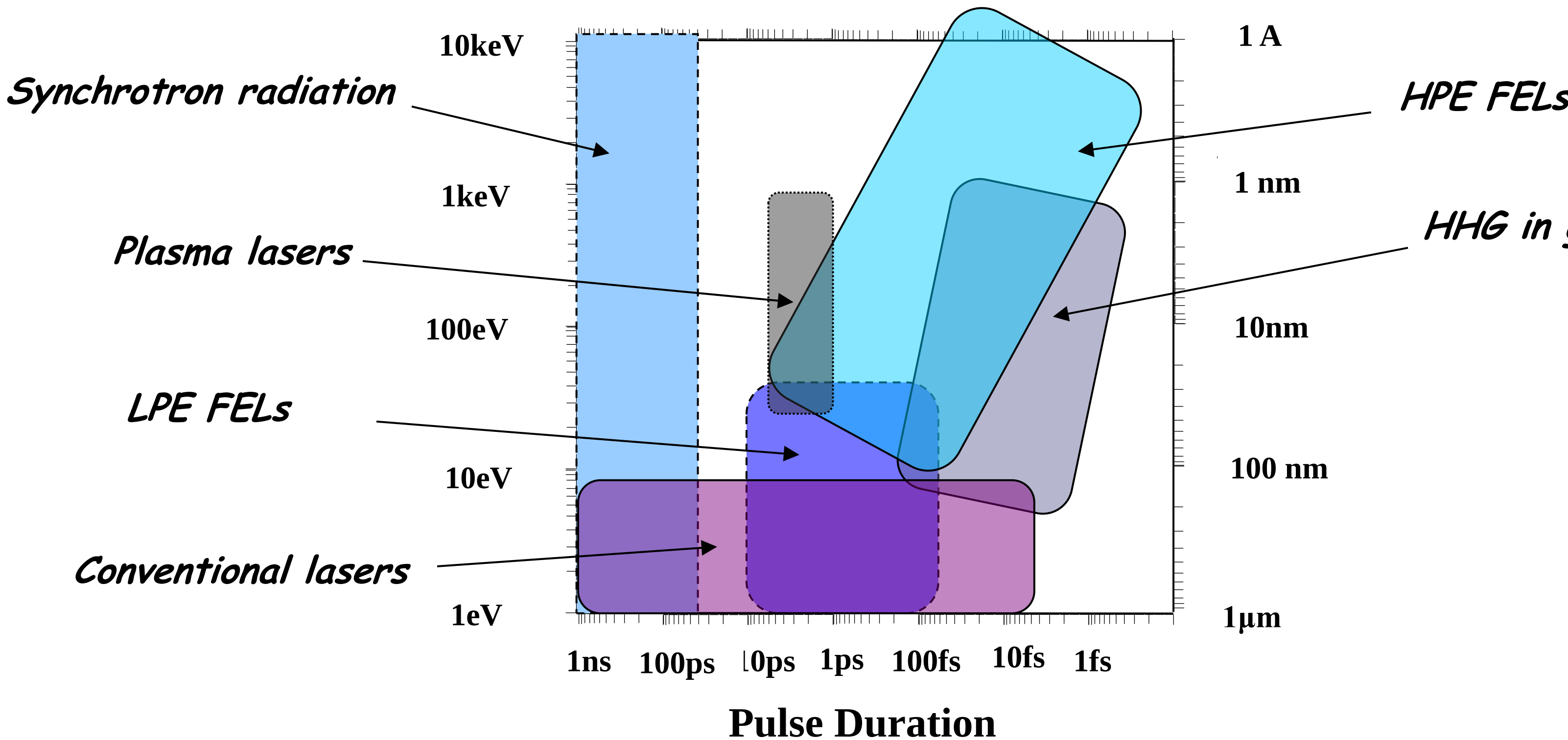
(n = harmonic number ~1, N = number of magnetic periods ~ 50)

FERMI@elettra parameters at 50 eV

$2 \cdot 10^{14}$ photons per pulse (100 fs)

Rep rate ~ 50 Hz

Why Free Electron Lasers ?



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

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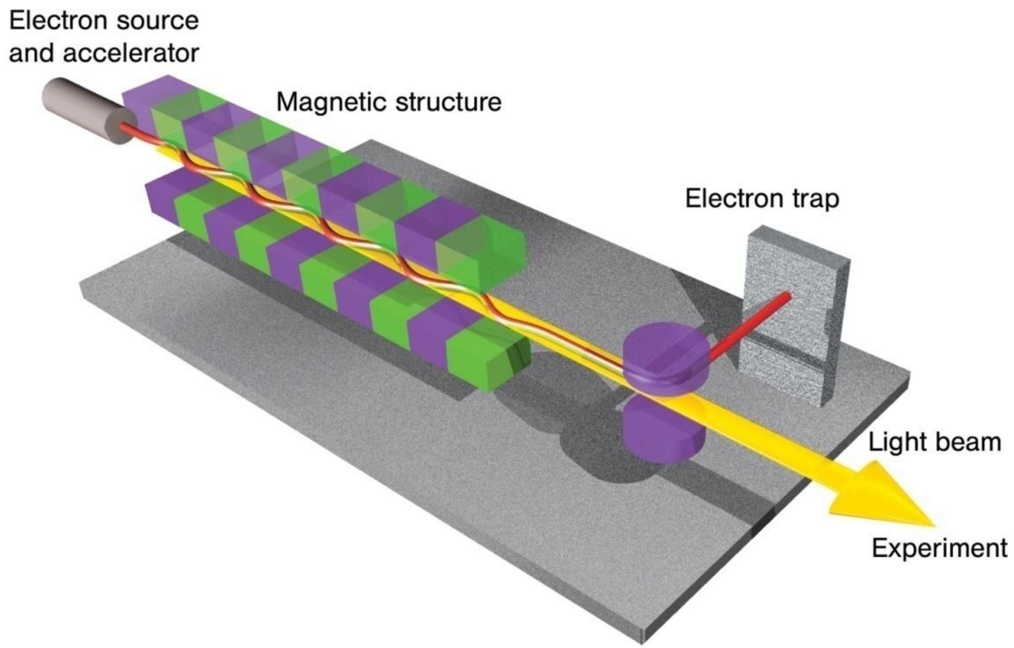

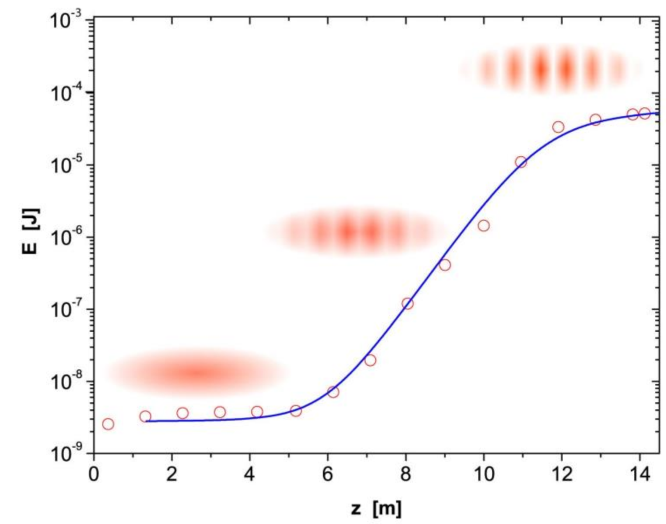

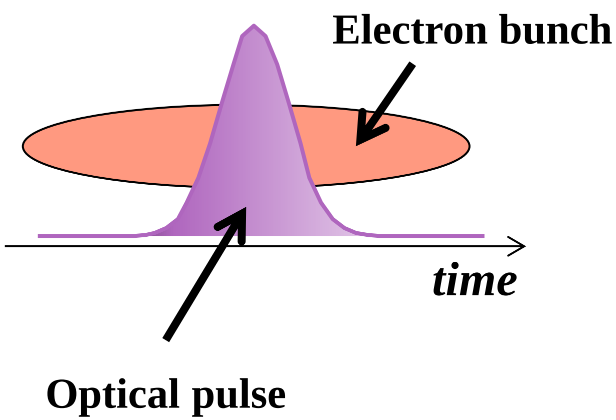

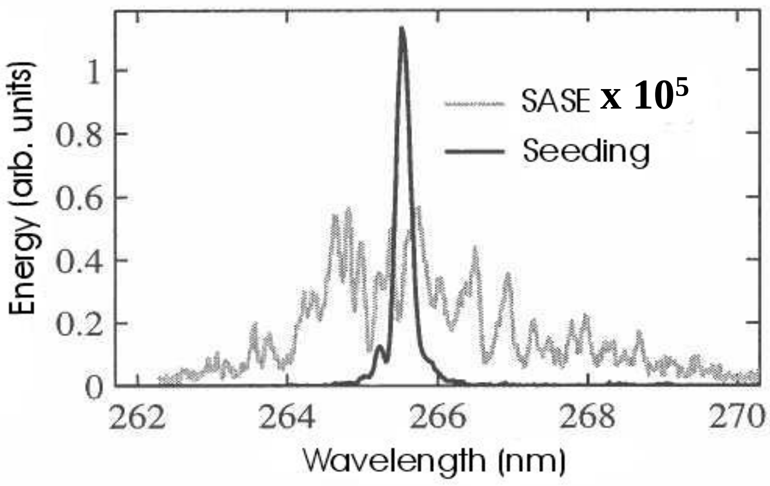

L. H. Yu et al., PRL (2003)

nature photonics

ARTICLES

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

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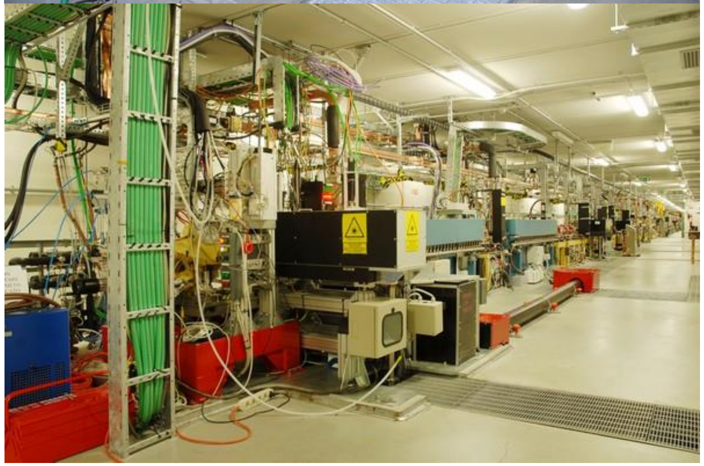
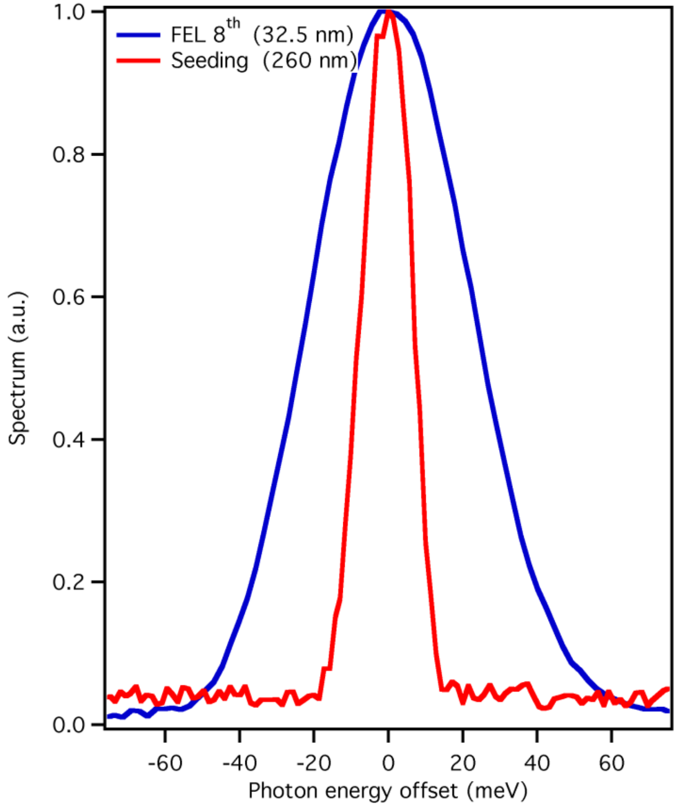
Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet

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

FERMI – Photons/Pulse

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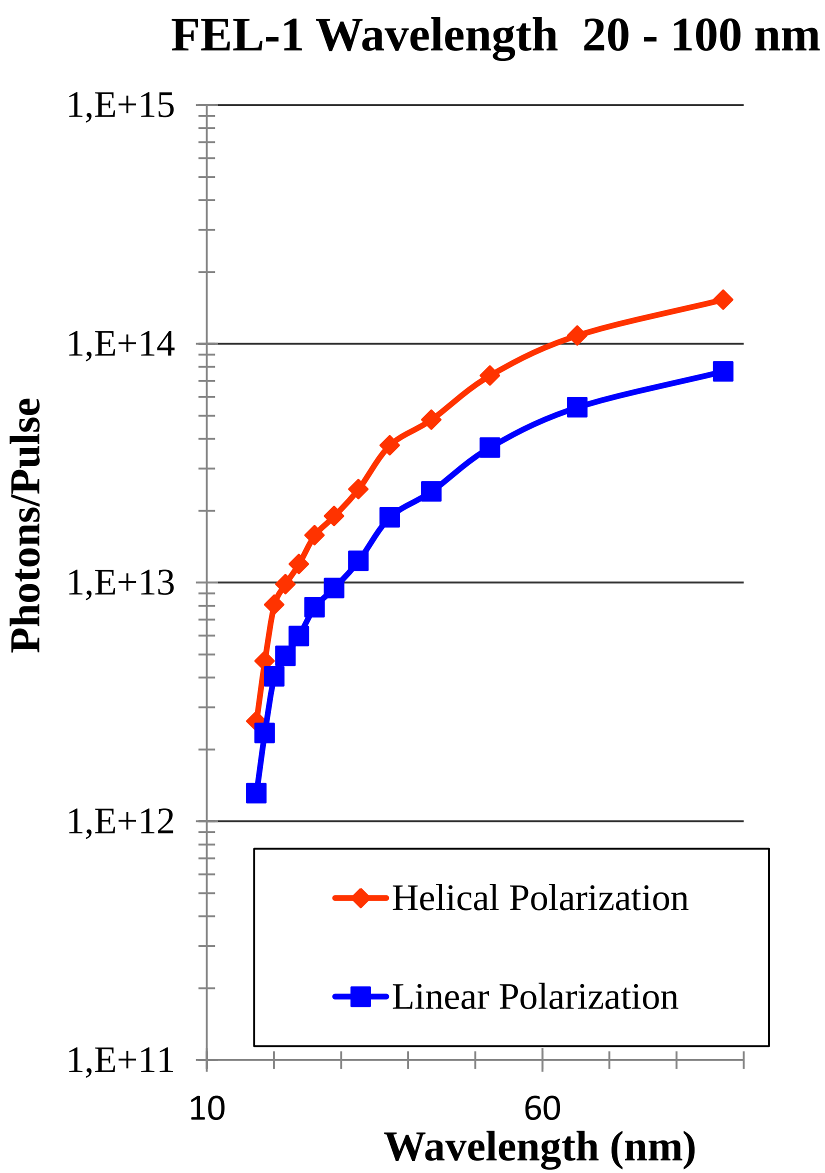

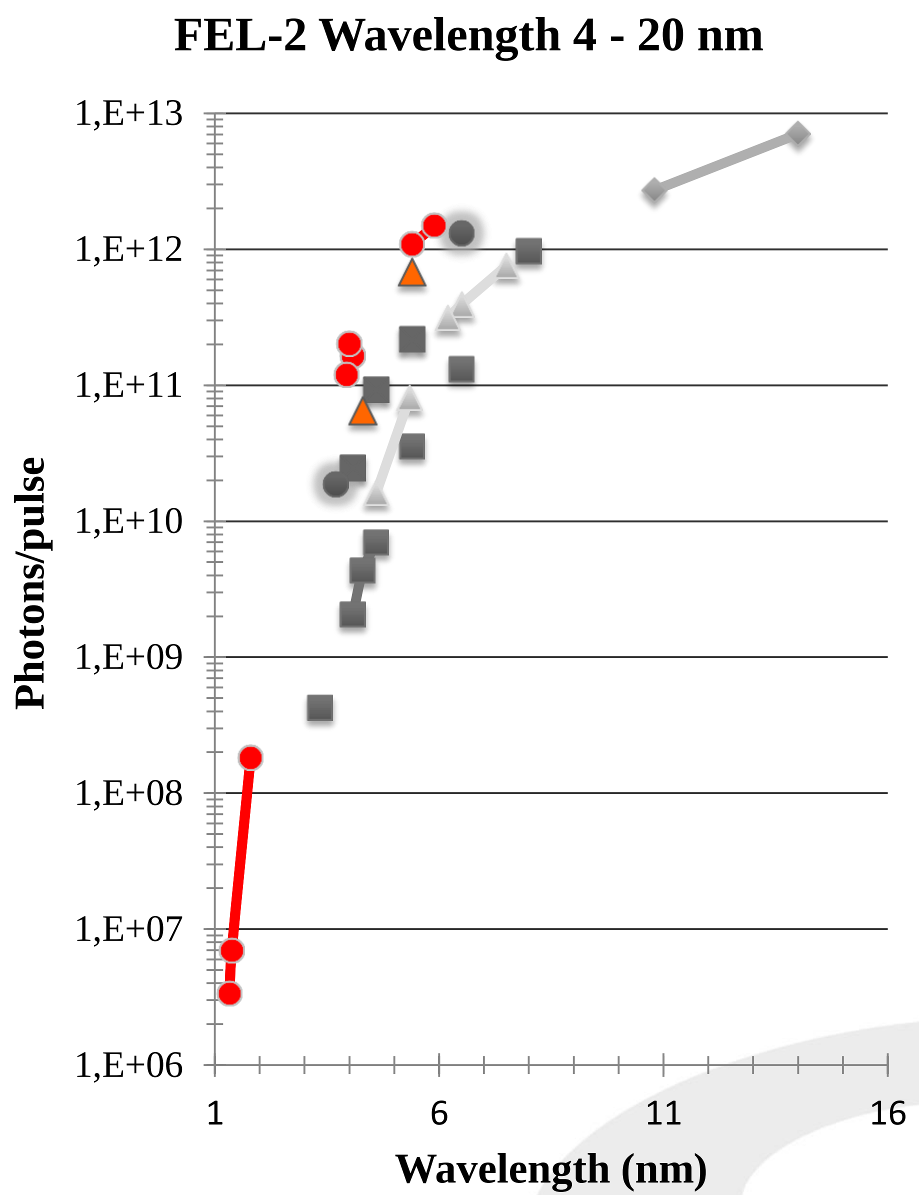

The Experimental Hall

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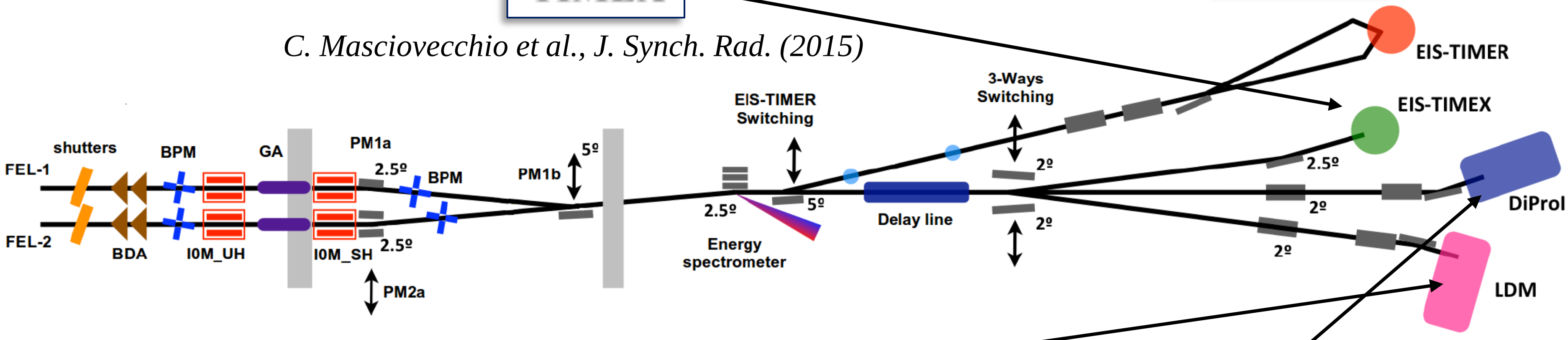
Commissioning

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

TIMER

TIMEX

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



LDM (Low Density Matter)

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

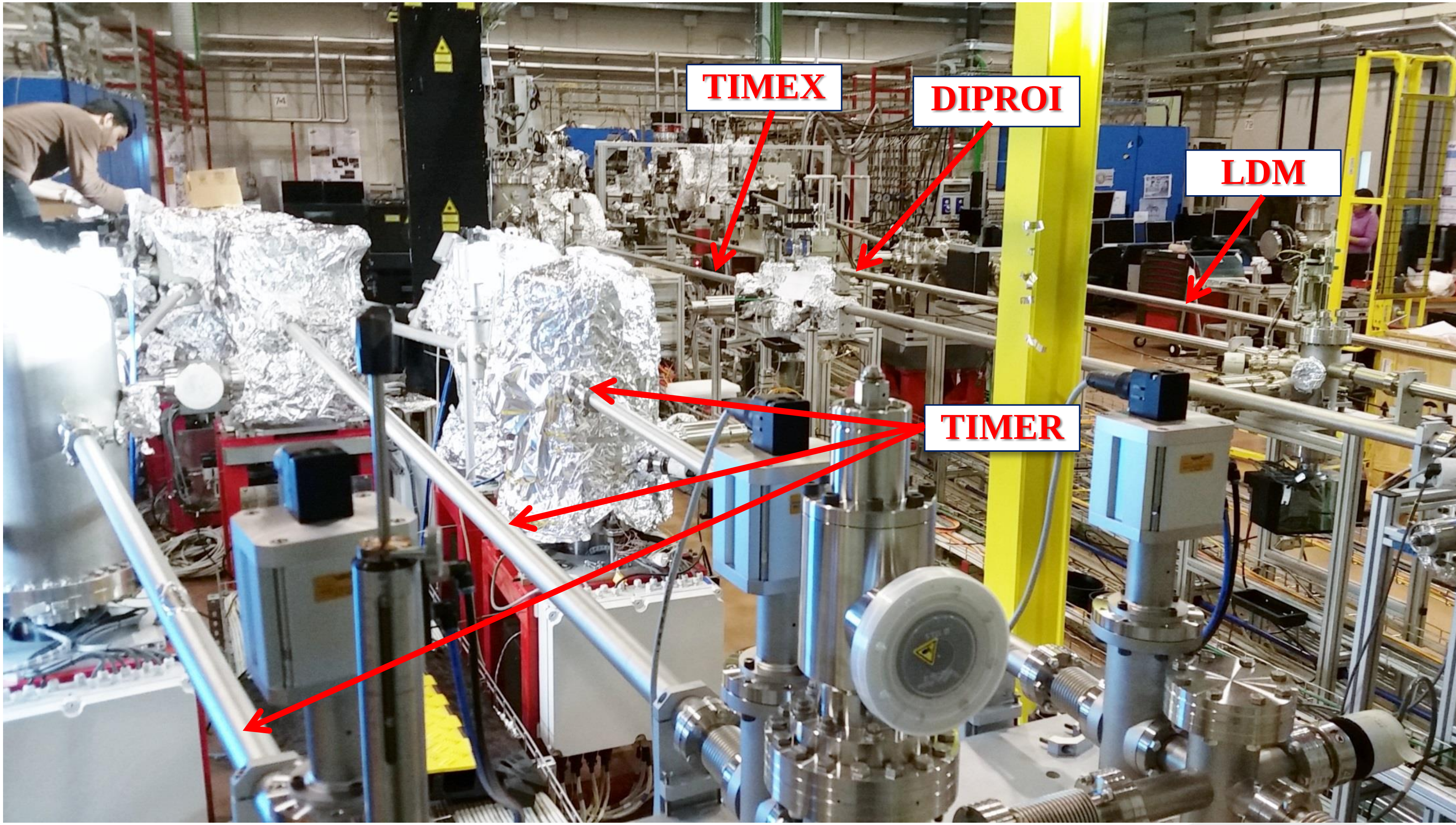
DIPROI (DIffraction & PROjection Imaging)

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

MagneDYN (Magnetic Dynamics)

TeraFERMI (THz beamline)

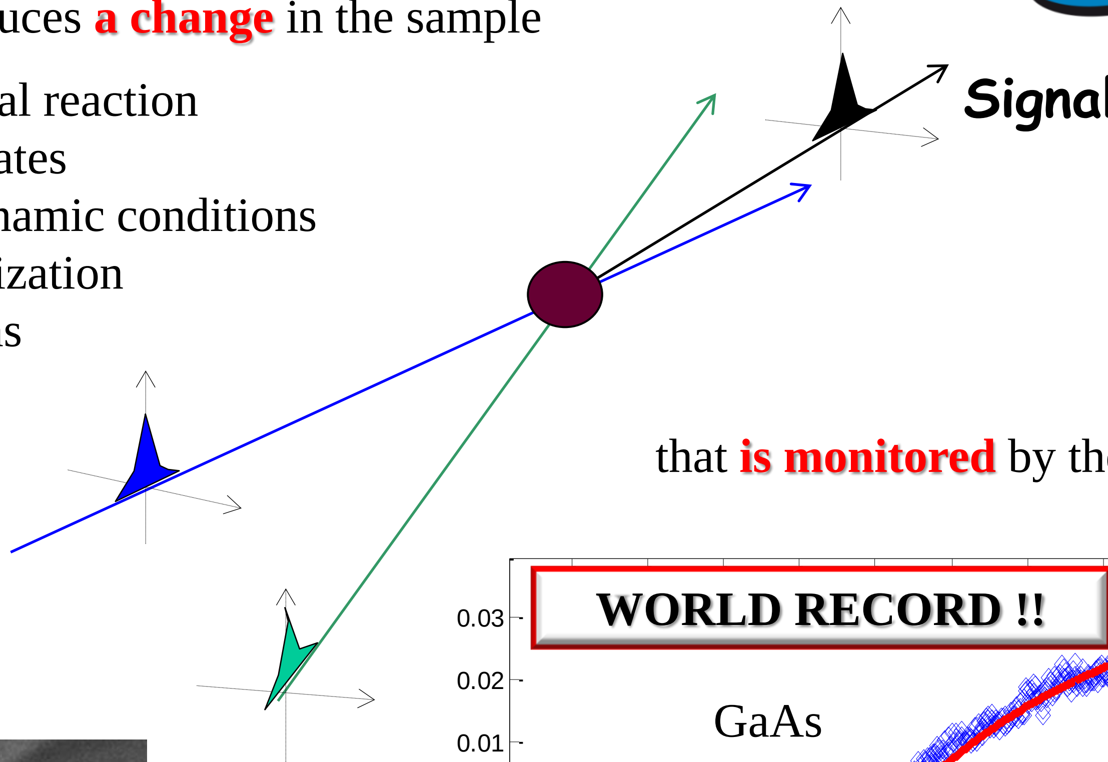
The Experimental Hall



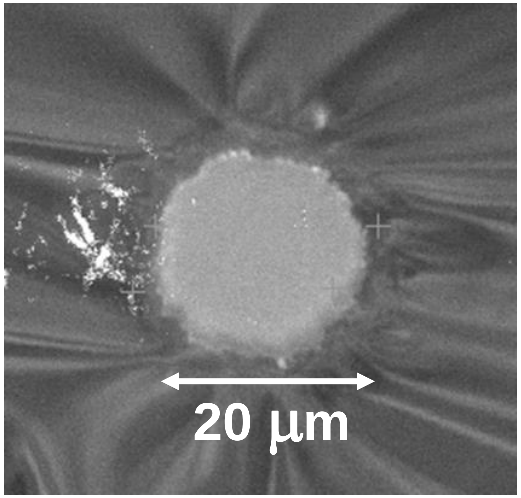
Pump & Probe

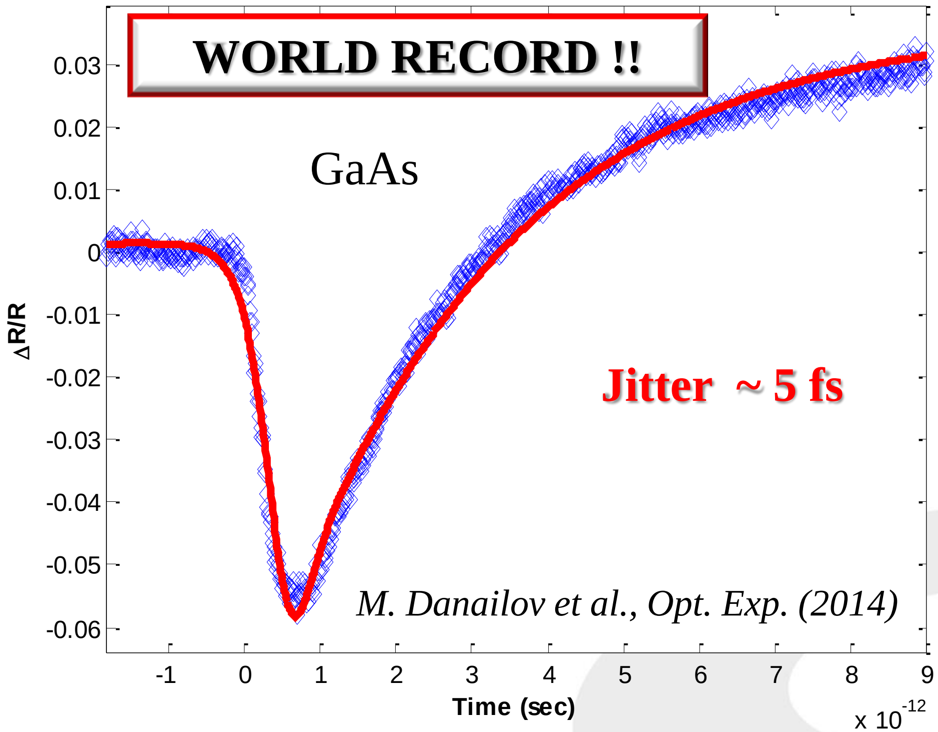
The **pump** pulse produces **a change** in the sample

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



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





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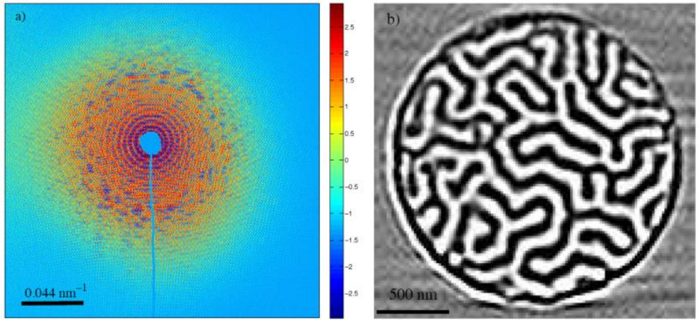
Ultrafast Magnetic Dynamics

Synchrotron Radiation News

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gsrn20

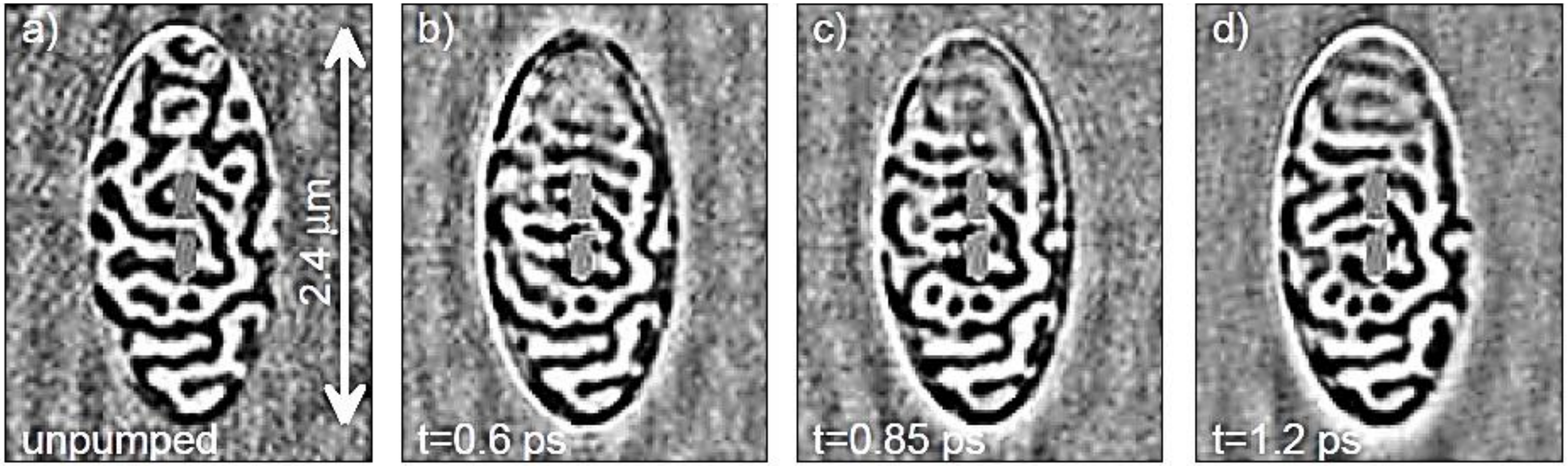
Ultrafast Dynamics of Magnetic Domain Structures Probed by Coherent Free-Electron Laser Light

L. Müller [a], S. Schleitzer [a], C. Gutt [a b], B. Pfau [c], S. Schaffert [c], J. Geilhufe [d], C. von Kc
Schmising [c], M. Schneider [c], C. M. Günther [c], F. Büttner [c e], F. Capotondi [f], E. Pedersoli [f]
Düsterer [a], H. Redlin [a], A. Al-Shemmary [a], R. Treusch [a], J. Bach [g], R. Frömter [b g], B. Voi
, J. Gautier [h], P. Zeitoun [h], H. Popescu [i], V. Lopez-Flores [i], N. Beaulieu [i], F. Sirotti [i], N. .
G. Malinowski [j], B. Tudu [k], K. Li [k], J. Lüning [k], H. P. Oepen [b g], M. Kiskinova [f], S. Eisebit
Grübel [a b]



Co/Pt ML

Controlling ultrafast demagnetization using localized optical excitation



C. von Korff Schmising, S. Eisebitt et al, PRL (2014)

LDM beamline

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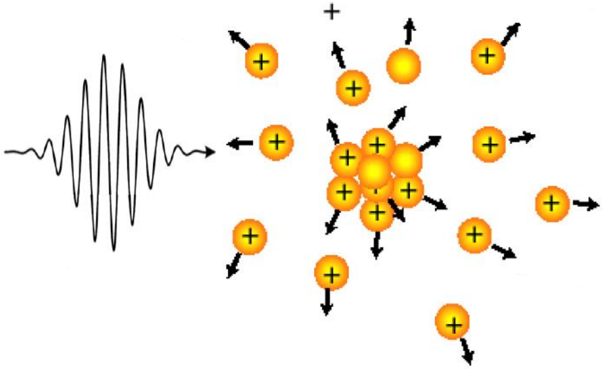
Fundamental physics:

- Structure of nano clusters
- Ionization dynamics
- Superfluidity – relaxation dynamics
- Non-linear optics
- Chirality



Material science:

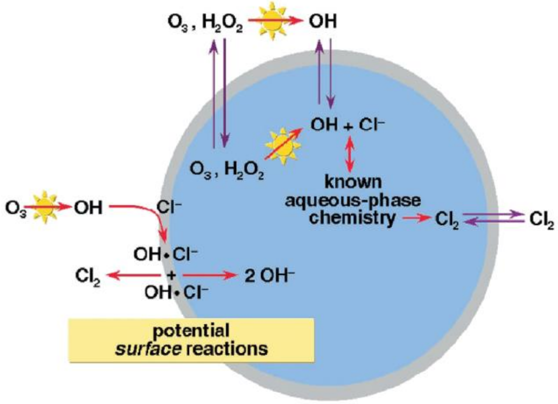
- Electronic properties of organic nanostructures
- Charge transfer dynamics in heterogeneous structures
- Magnetism of nanoparticles
- Catalysis in nanomaterials

Biochemistry:

- Micro solvation of bio-molecules

Aerosol / Atmospheric chemistry:

- Reactions at microscopic water interfaces



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Coherent control at the attosecond time scale

K. Prince et al., Nat. Phot. (2016)

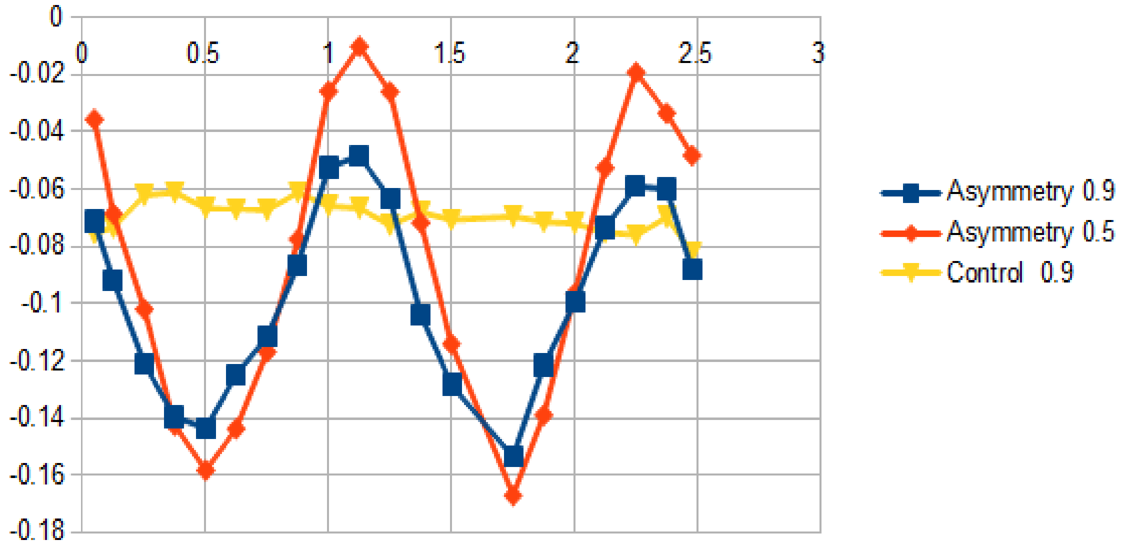
Interference effect among quantum states using single and multiphoton ionization

C. Chen et al., PRL (1990)

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

Use of first (62.974 eV) and second harmonic on $2p^5 4s$ 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!

Elastic and Inelastic Scattering (EIS)

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The **Sample Side**

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

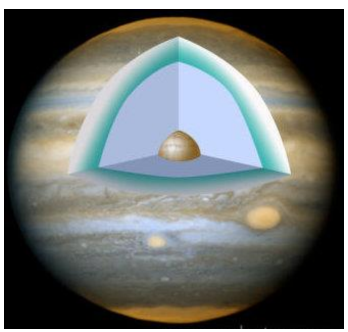
What happens to the **Sample?** — Non-equilibrium distribution of electrons

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

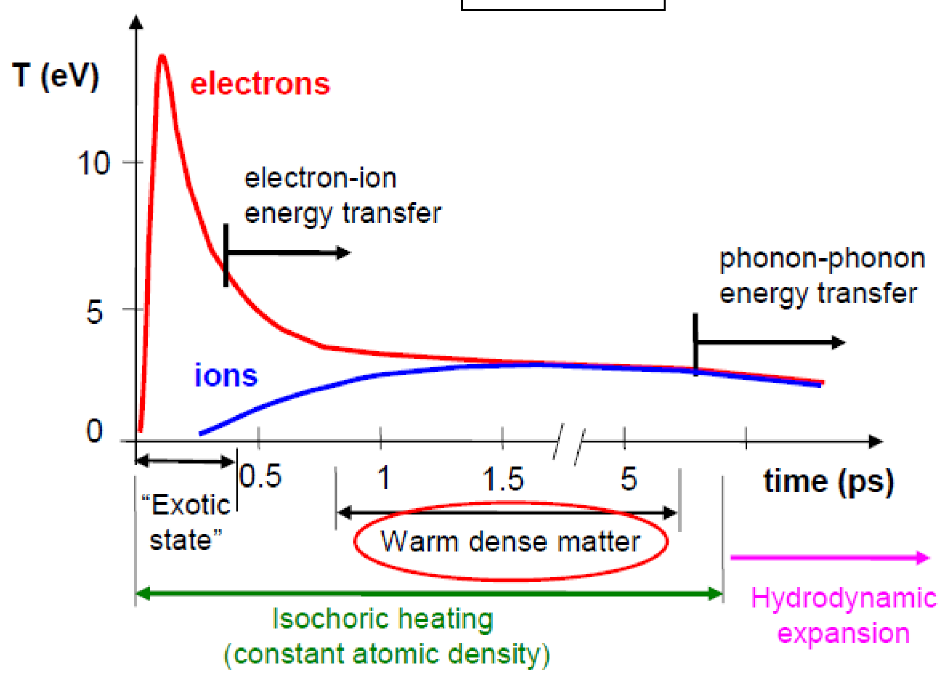
The intensity of the FEL pulses will determine the process to which the sample will undergo: simple heating, structural changes, ultrafast melting or ultrafast ablation

TIMER

TIMEX



interior of **large planets** and stars



Exercise 2

Calculate sample temperature assuming total absorption from surface

N = number of photons per pulse

E = photon energy (100 eV)

d = beam size (100 μm)

σ_{abs} = absorption cross section per atom (Al ~ $7 \cdot 10^{-18}$ cm^2)

k_B = Boltzmann constant $1.3 \cdot 10^{-23}$ J K^{-1}

$$\mathbf{T_{SAMPLE} \sim N/3(k_B/E)\,(d^2/\sigma_{abs})}$$

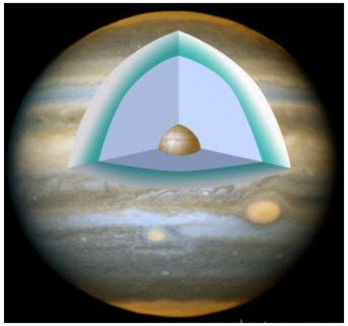
EIS - TIMEX

TIMEX

TIme-resolved studies of Matter under EXtreme and metastable conditions

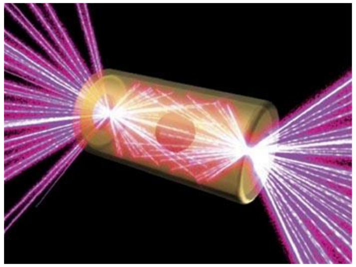
• Generation of warm dense matter (WDM) in simple metals (Al,Au, etc…): looking for EoS studying the FEL-induced isochoric heating followed by isoentropic expansion

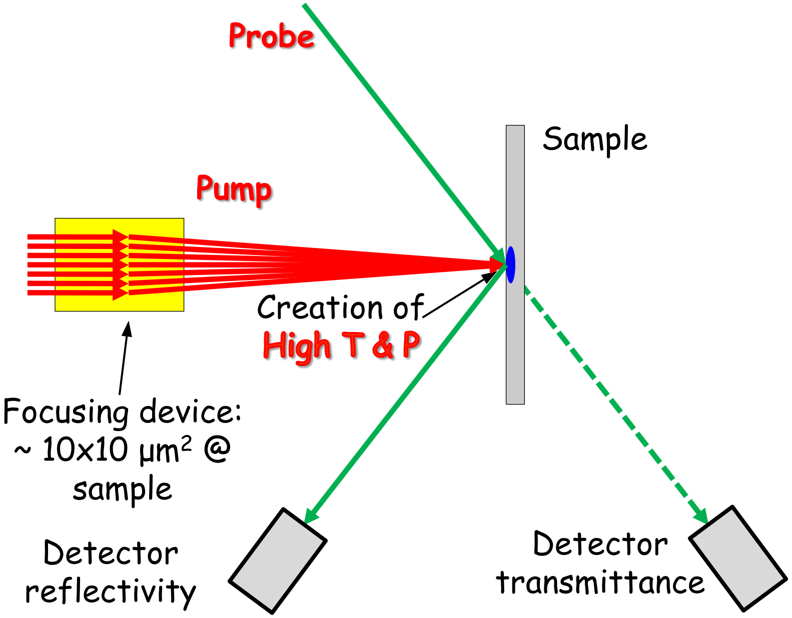
• Generation of metastable liquids in a "no man's land" through ultrafast heating of amorphous states (Si, Ge...)



interior of **large planets** and stars

inertial confinement **fusion**





• Pump → Table-top Laser, FEL

• Probe1 → Table-top Laser (single energy or Super-continuum (CaF_2 crystal))

• Probe2 → FEL (first or third harmonics (tunability))

• Probe3 → Electron Diffraction (100 fs, 200 keV)

FEL pulses allow **uniform** heating:

300 Å Al uniformly heated at 1 eV (10000 K)
Δt = 100 fs, λ = 60 nm, d = 10 μm^2, 10^{12} ph/pulse

C. Masciovecchio et al., JSR (2015)

SCIENTIFIC REPORTS

F. Bencivenga et al., (2014)

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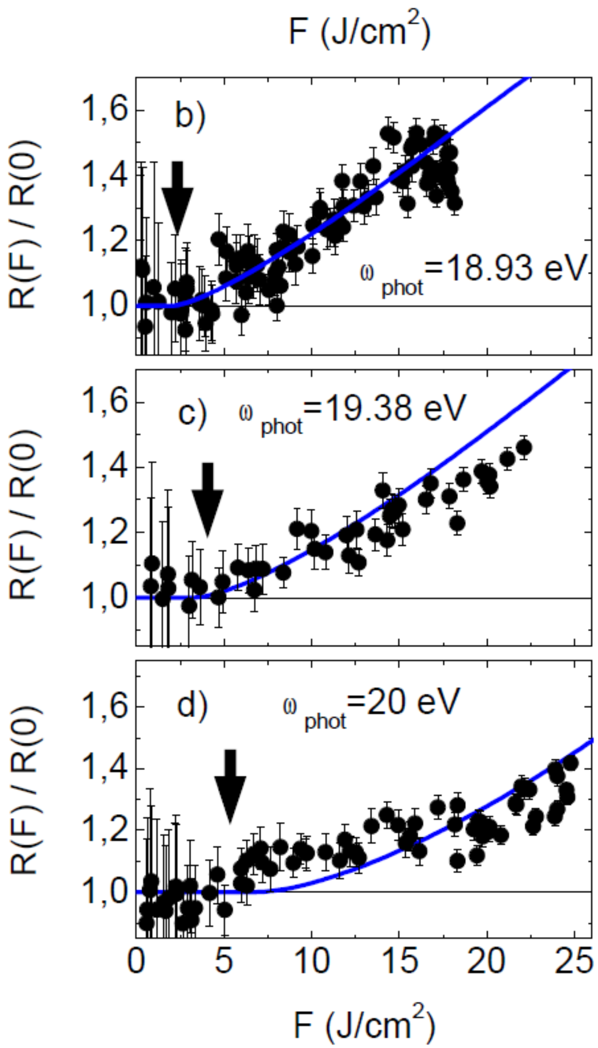
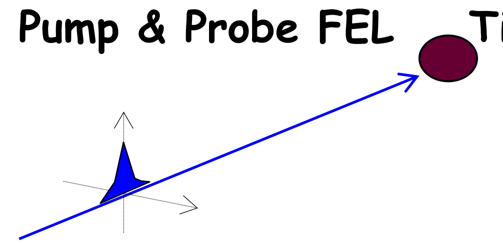




$$R \approx \left| \frac{\sqrt{\epsilon} - 1}{\sqrt{\epsilon} + 1} \right|^2$$

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

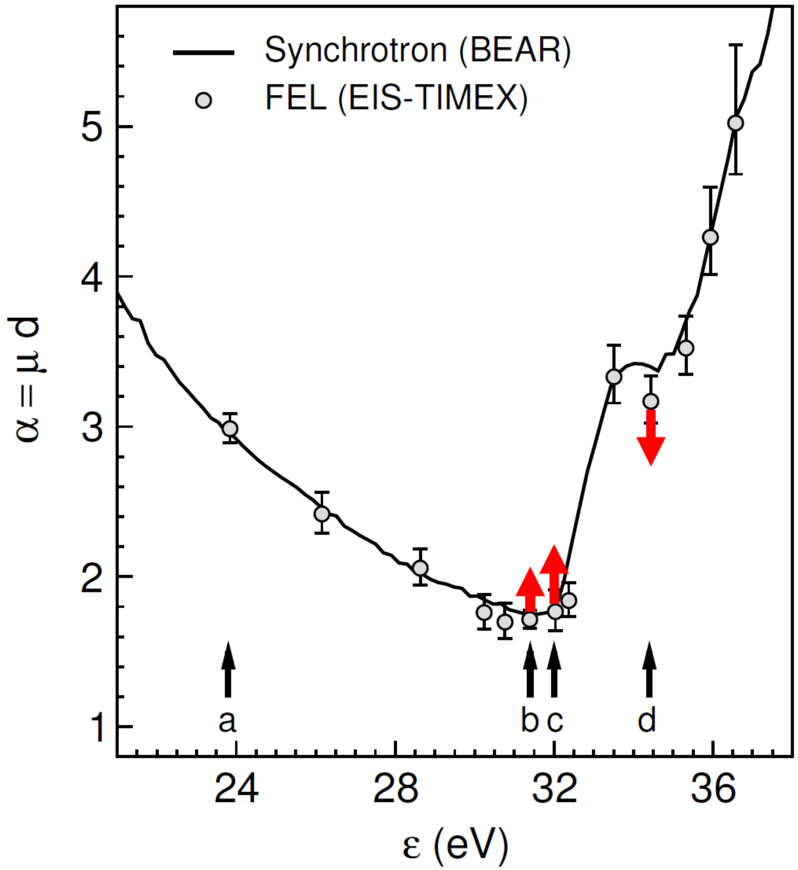
$$\overline{E}(F, t) = \frac{F}{L} \int_{-\infty}^{t} (1 - R(F, t'))\, G(t') dt'$$

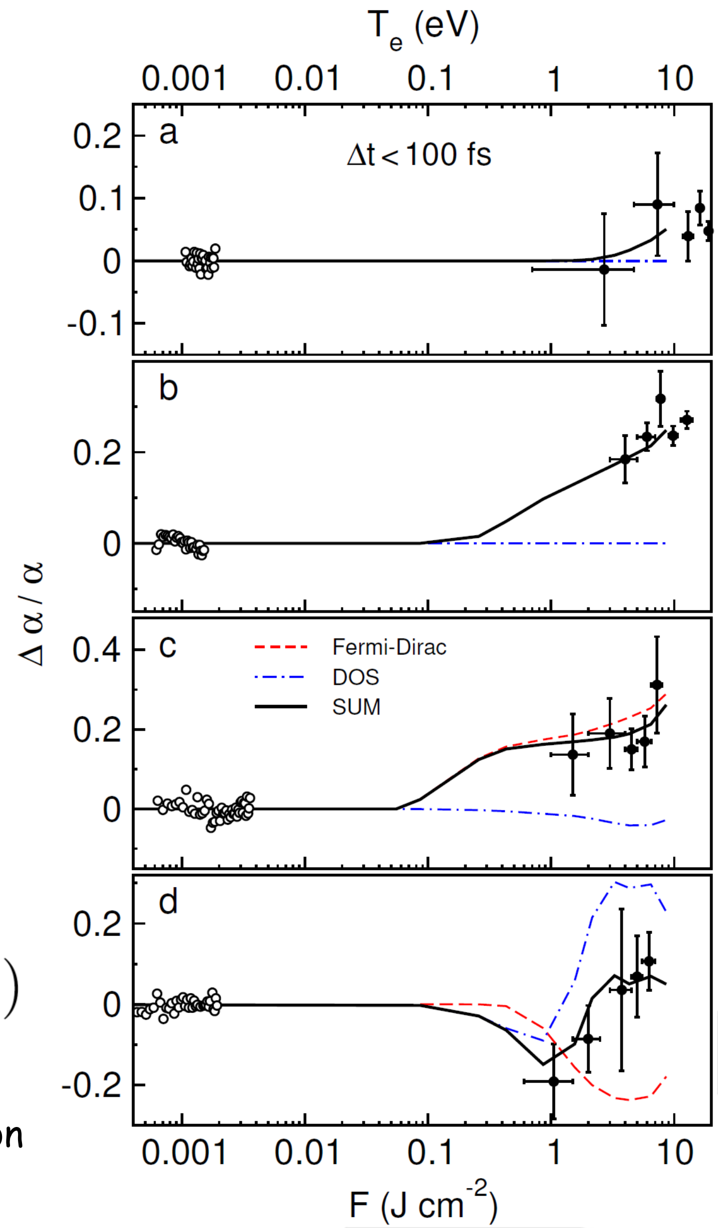
$$\Downarrow$$

$$T_e(F)$$

TIMEX

Ultrafast changes in the **Electronic Structure** of Ti



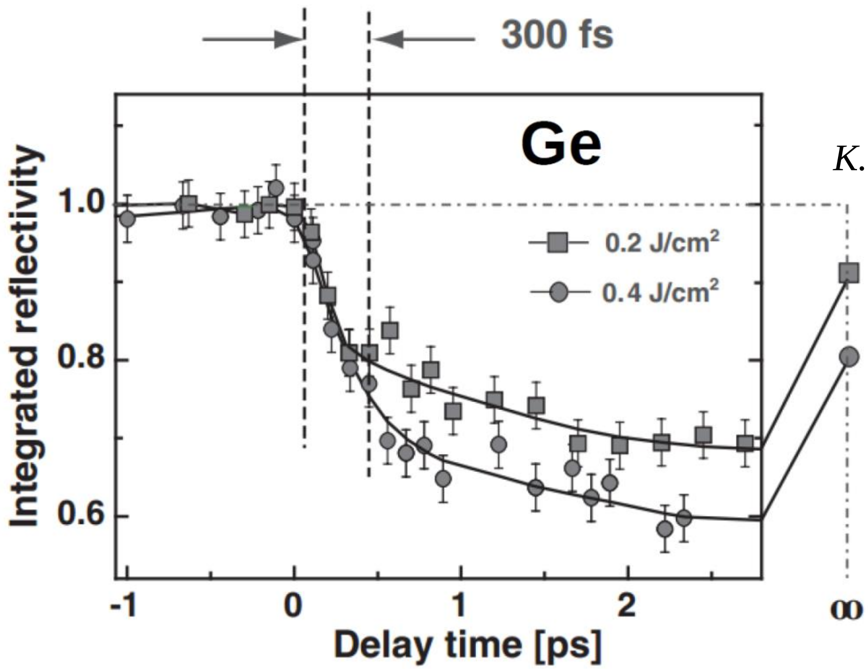


$$\frac{\Delta\alpha}{\alpha}(\varepsilon, T_e) = c_g(\varepsilon)\, g(\varepsilon, T_e) + c_\phi(\varepsilon)\, \phi(\varepsilon, T_e)$$

DOS variation

Empty Electronic States Probability Distr. variation

Principi et al., Struct. Dynamics (2016)



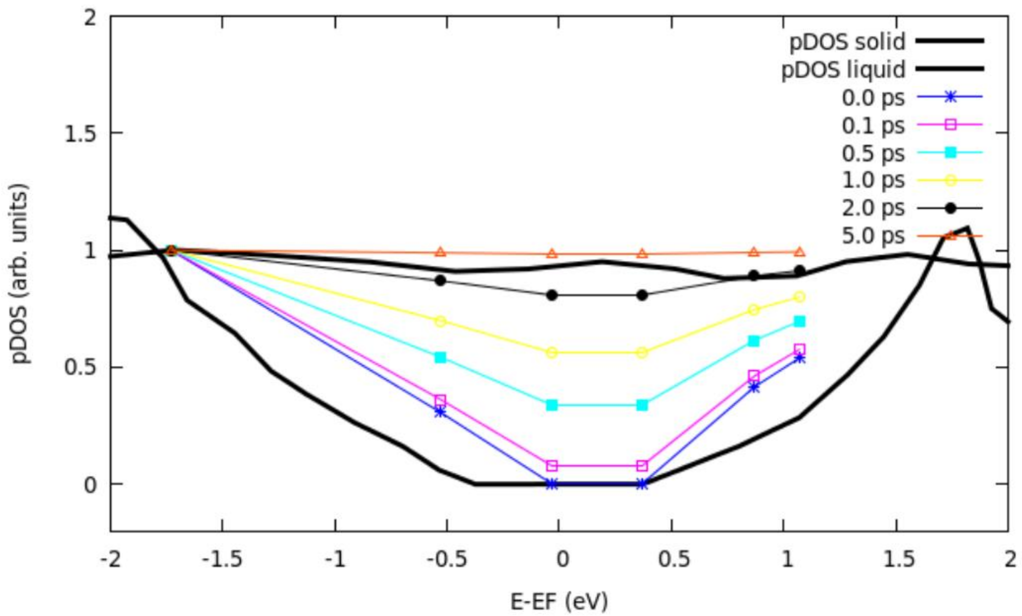
K. Sokolowski-Tinten et al., J of Phys: Cond. Matt. (2004)

Time-resolved **X-ray diffraction** is sensitive to the **long-range order** and therefore to the melting

but **does not** probe the **short range order** that finally affects the **DOS**

Time-resolved **X-ray absorption does** probe the **short range order**

low density liquid (**LDL**) phase with a **tetrahedral local structure** appears after 300 fs and survives for about 1 ps



E. Giangrisostomi et al., to be submitted

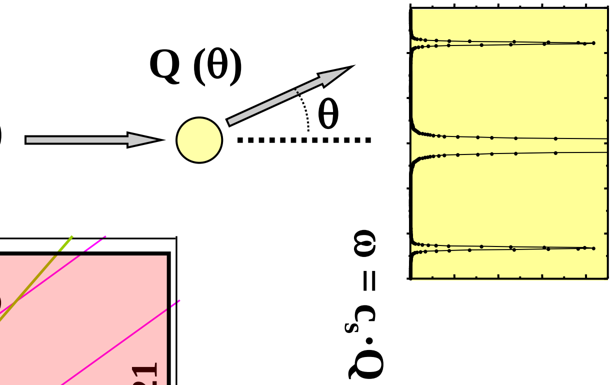
EIS - TIMER

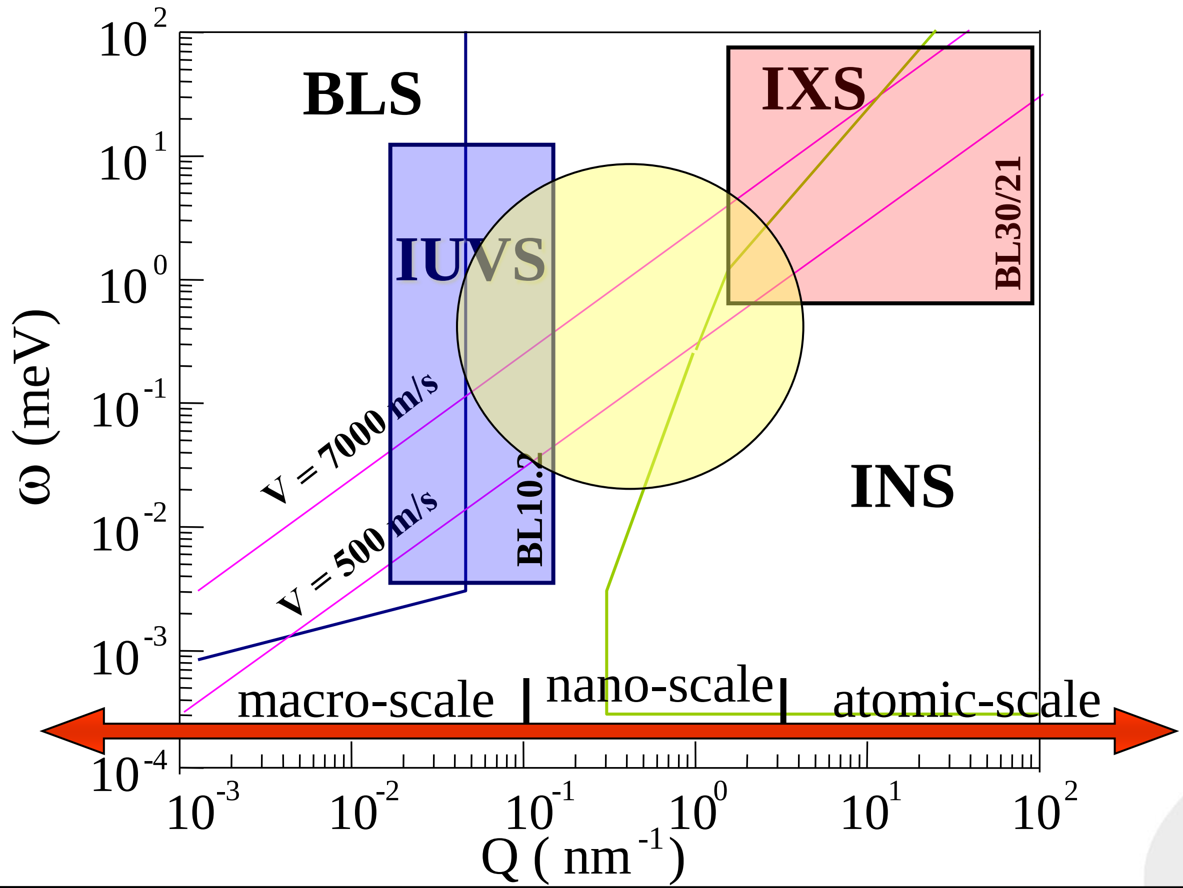
TIMER

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?

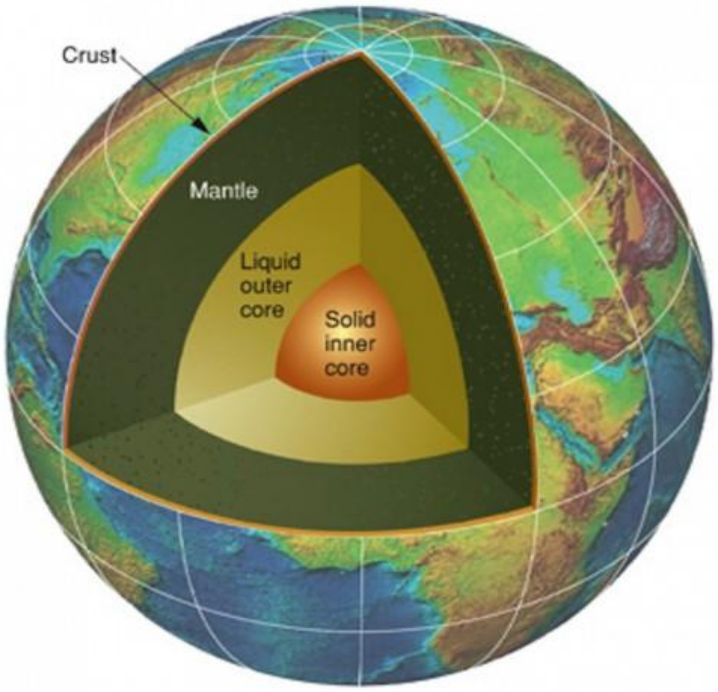
Sonoluminescence
What causes the emission of short bursts of light from imploding bubbles in a liquid when excited by sound?

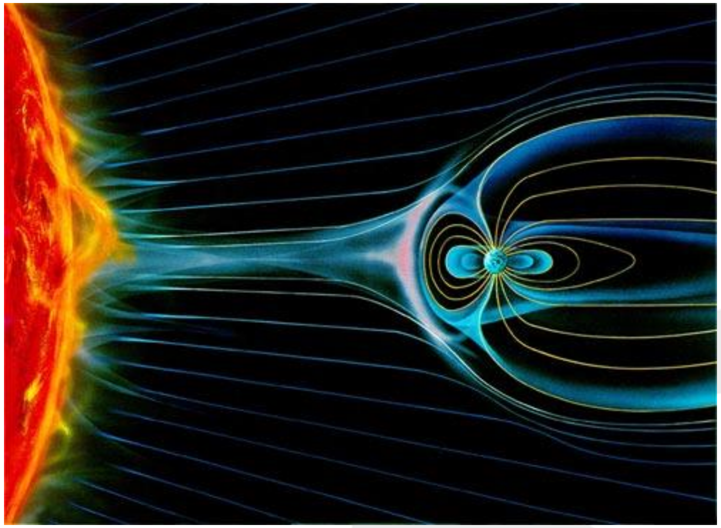
Turbulence
Is it possible to make a theoretical model to describe the statistics of a turbulent flow (in particular, its internal structures)? Also, under what conditions do smooth solution to the Navier-Stokes equations exist?

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 Disordered Systems ?





Convection of liquid metals creates the Earth's magnetic field. It deflects the solar wind. Without this field, the solar wind would directly strike the Earth's atmosphere. This would strip the Earth's atmosphere away slowly. This is hypothesized to have happened to the Martian atmosphere, rendering the planet incapable of supporting life.

Why at the nanoscale ?

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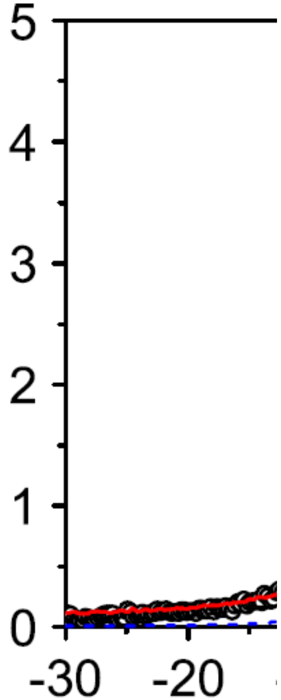
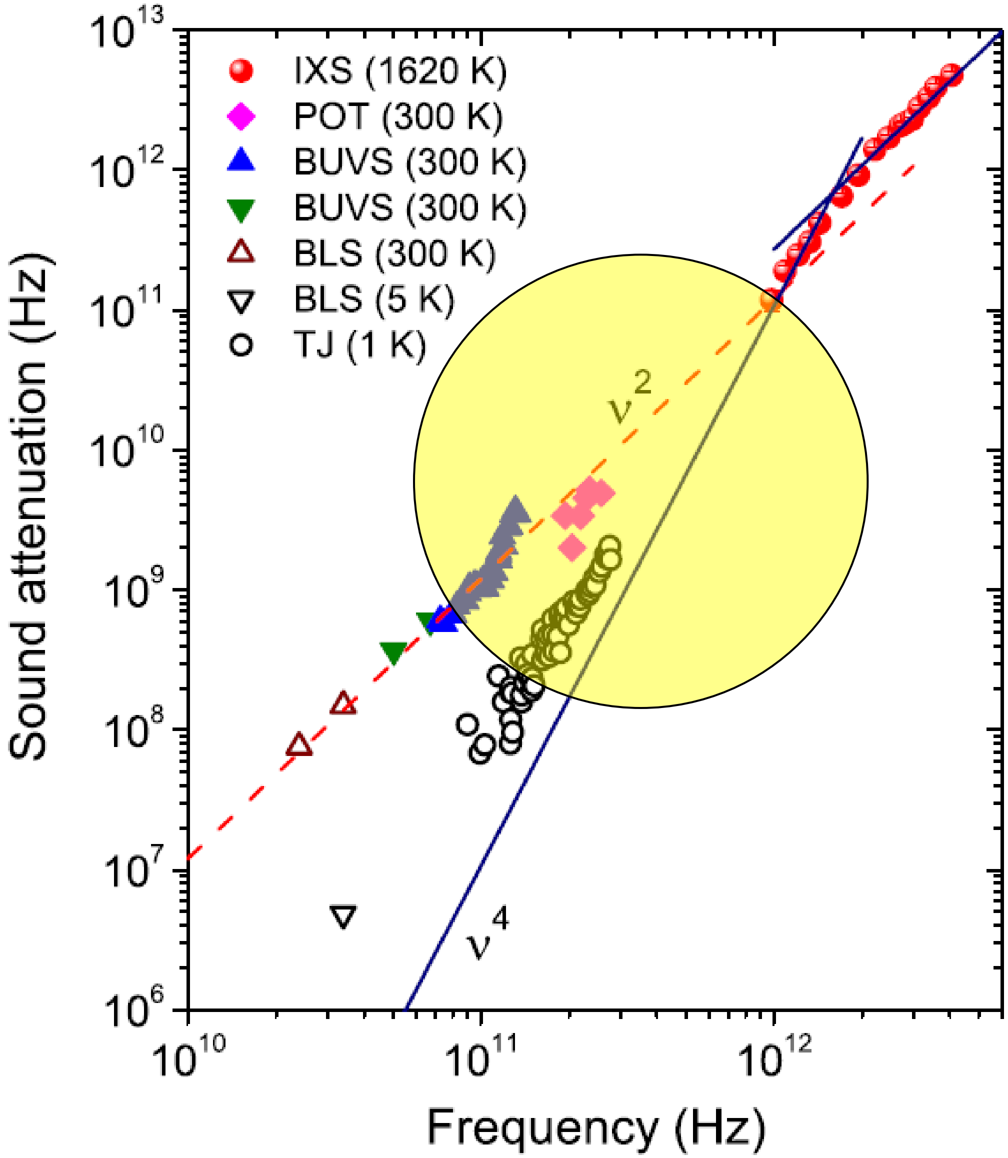
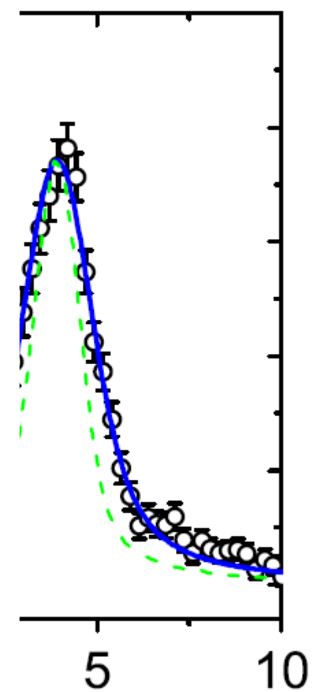
The nature of ... ear (V-SiO_2)







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.

Water Dynamical Structuring

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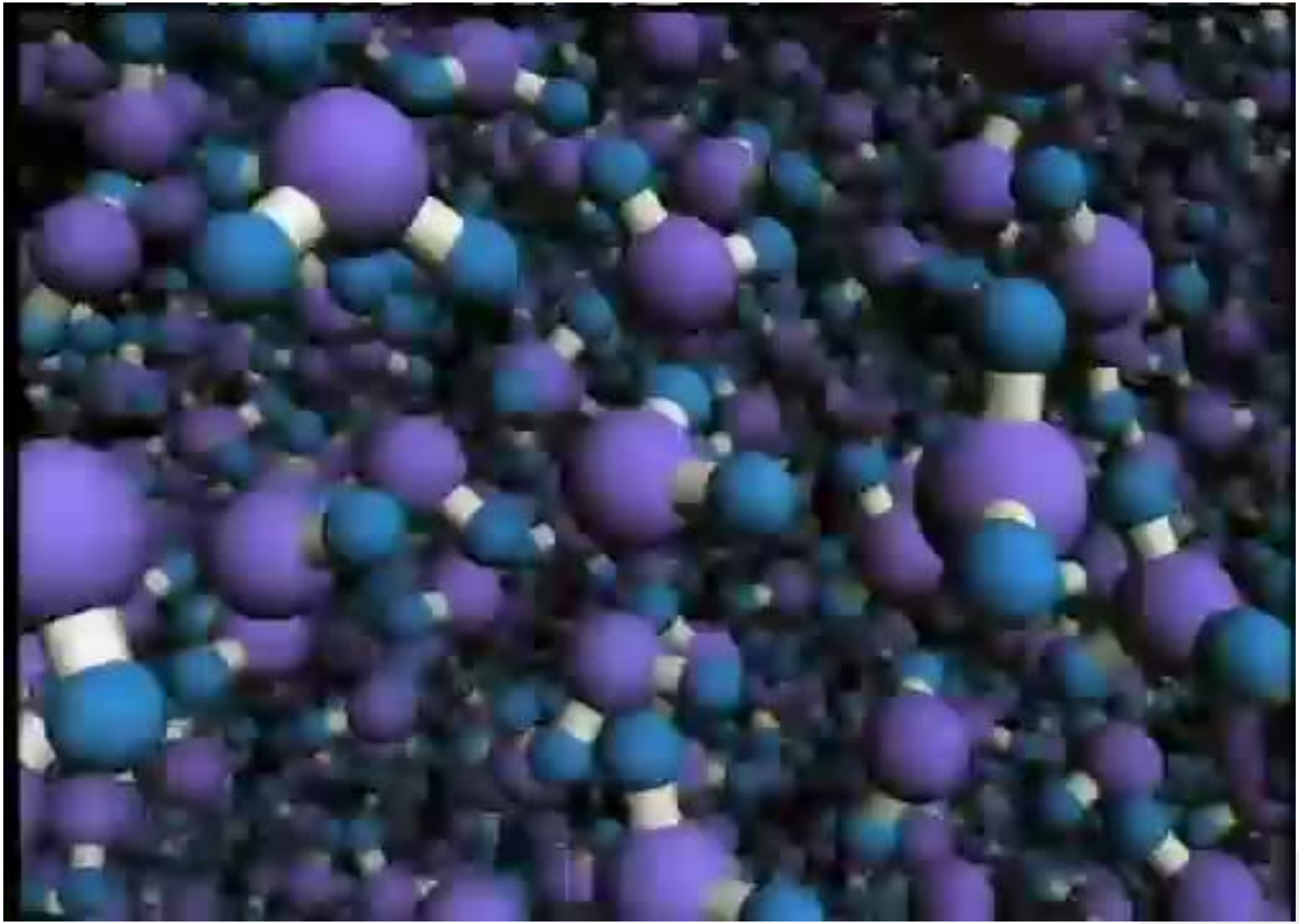
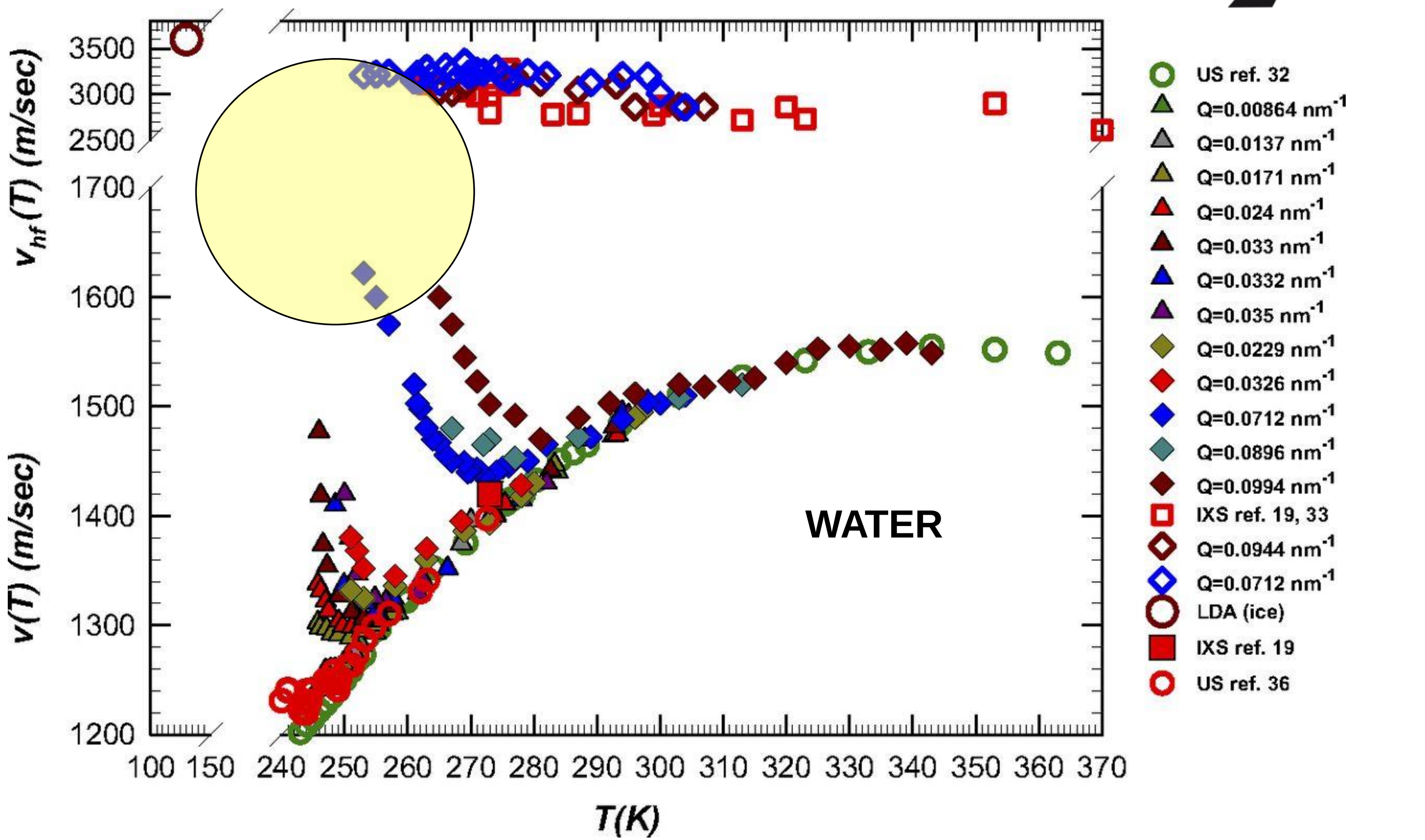
Sincrotrone
Trieste



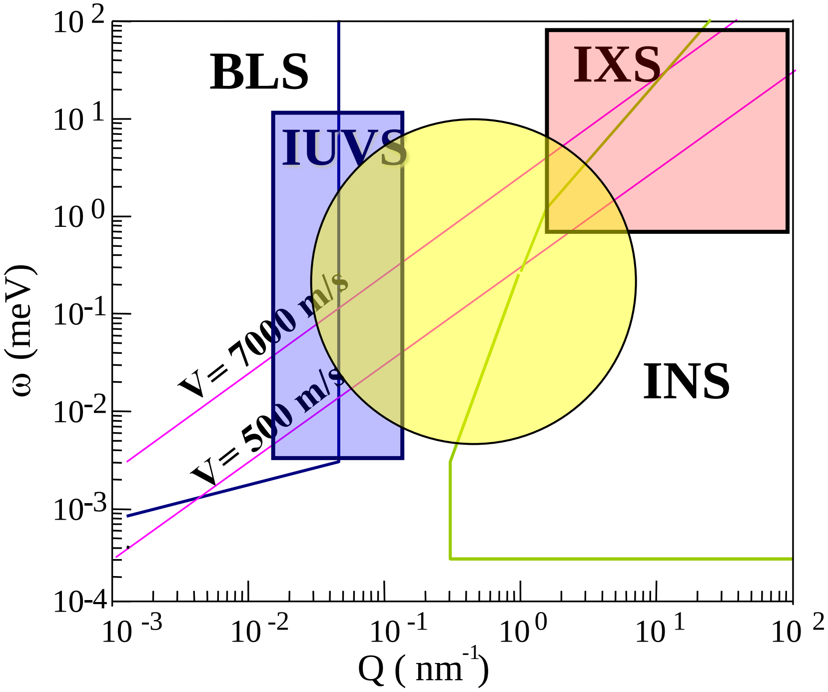
Water Structural Relaxation



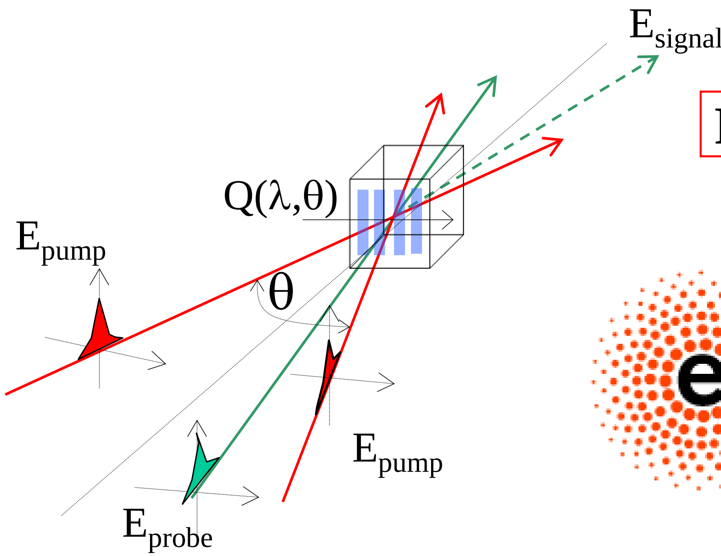
……only by treating water as a viscoelastic system is it possible to understand the microscopic origin of water anomalies. The proposed scenario here has been developed using a dynamic approach……

F. Mallamace et al., PNAS (2013)

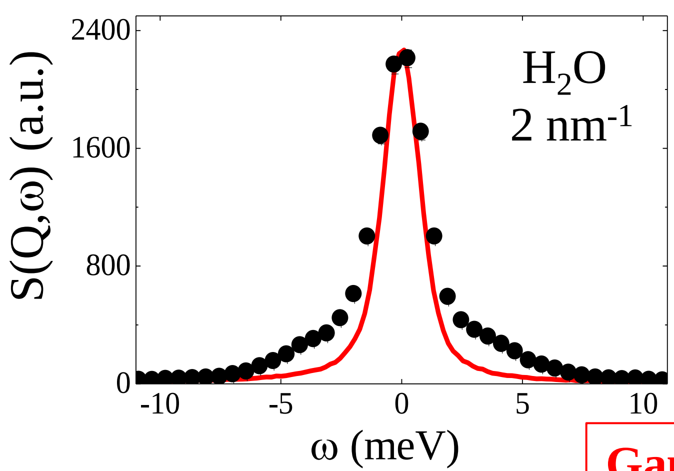
TIMER

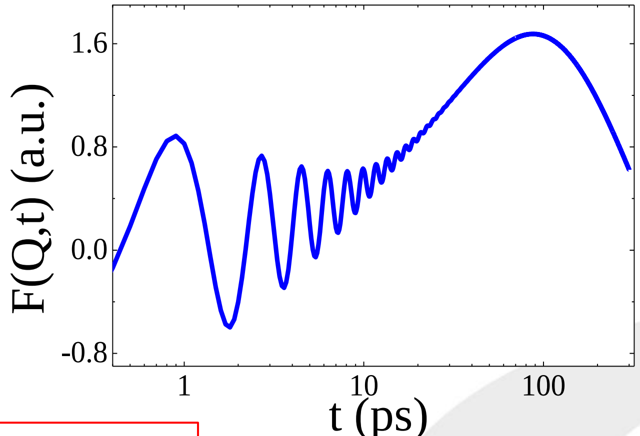


Solution: Free Electron Laser based Transient Grating Spectroscopy



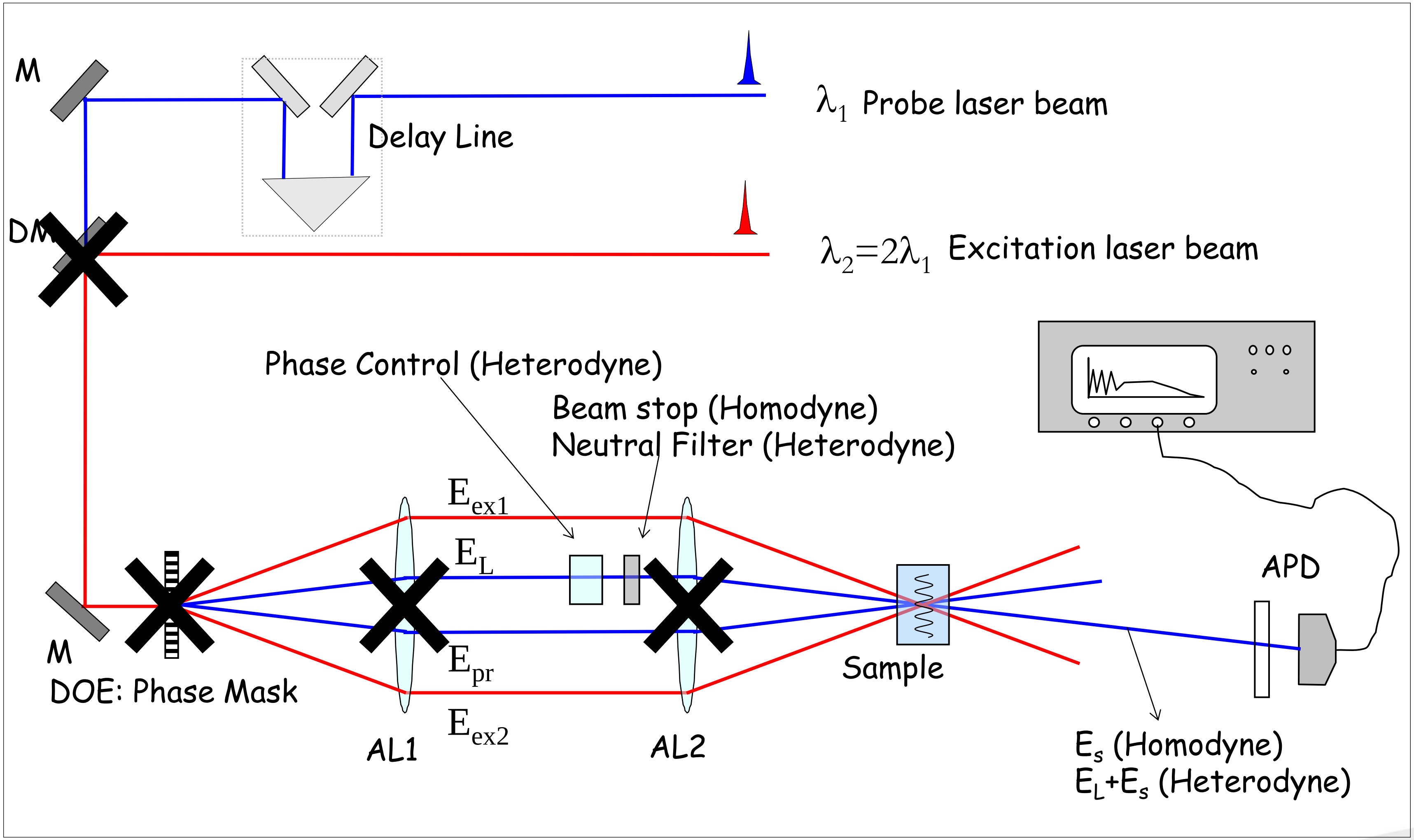
F(Q,t)





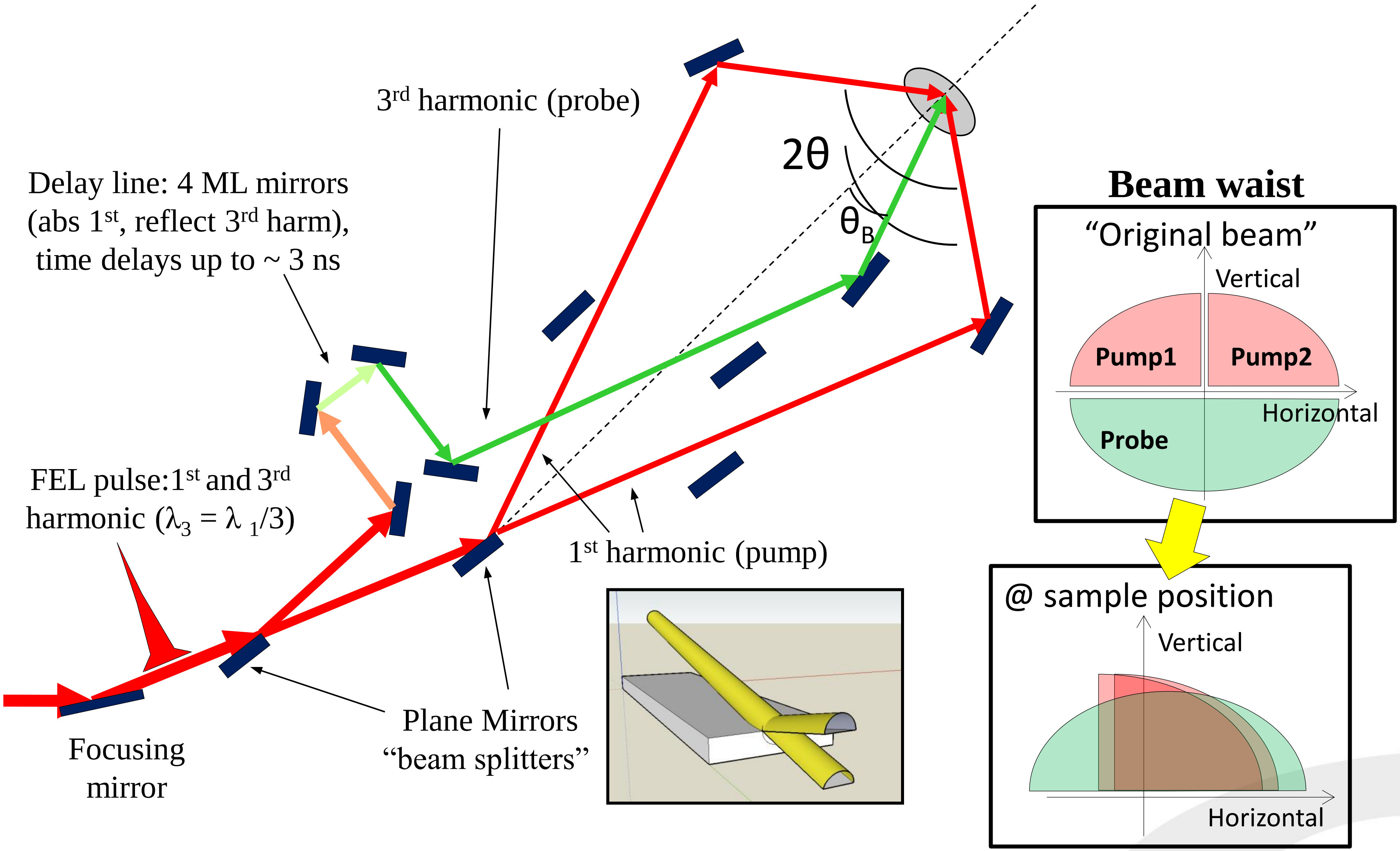
Gaussian-like time profile

Typical Infrared/Visible Set-Up



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

TIMER Layout

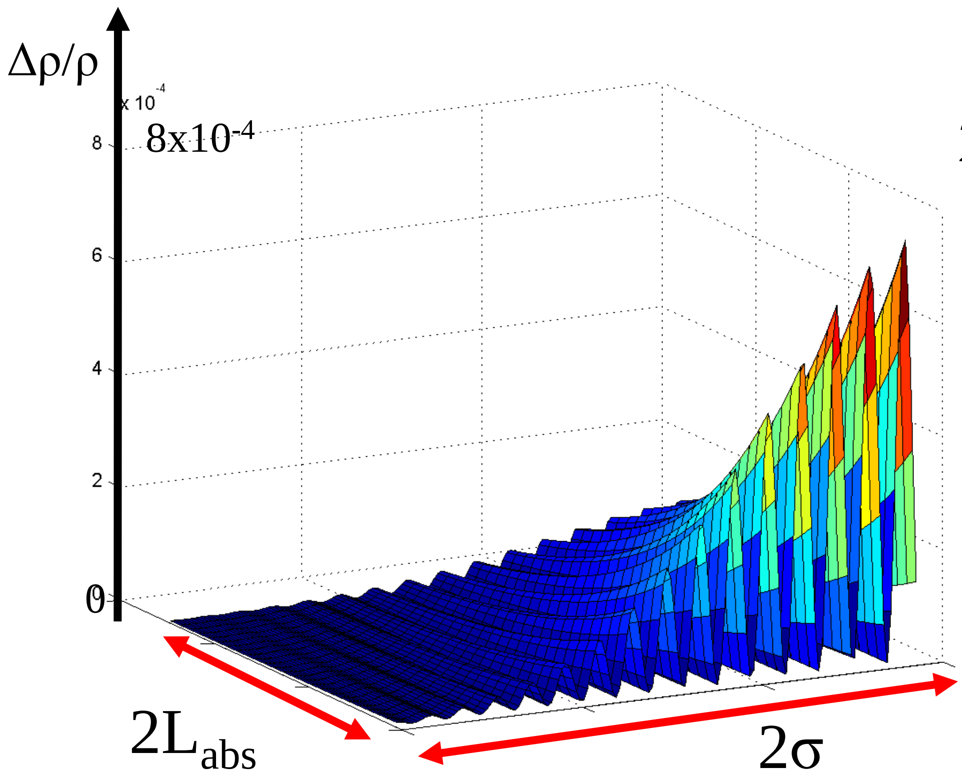


R. Cucini et al et al., NIMA (2011)

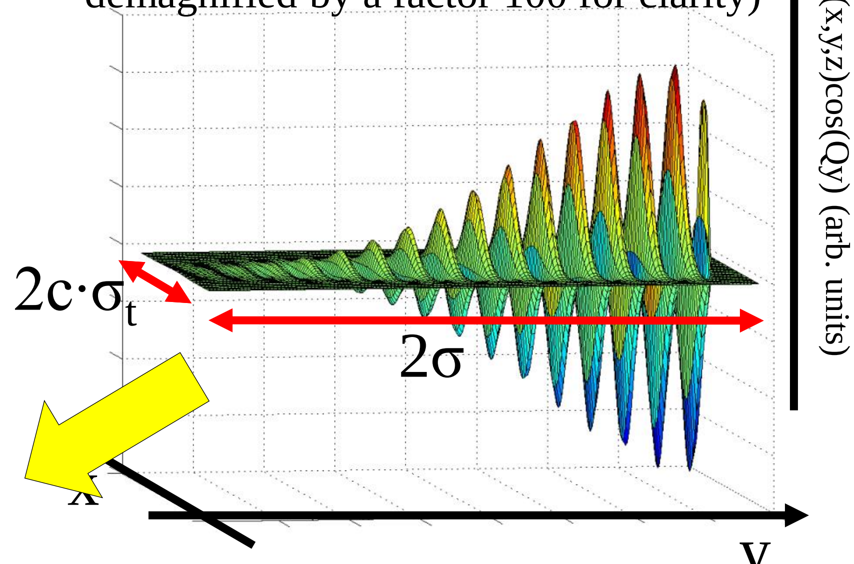
Expected TG formation

SiO_2 sample

Relative density variation (@ z=0, considering only the optical absorption of pump radiation)



Interference @ sample position (@ z=0, λ=60 nm, θ=9.2°, periodicity demagnified by a factor 100 for clarity)



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

α: $25.5\cdot10^{-6}$ K^{-1}

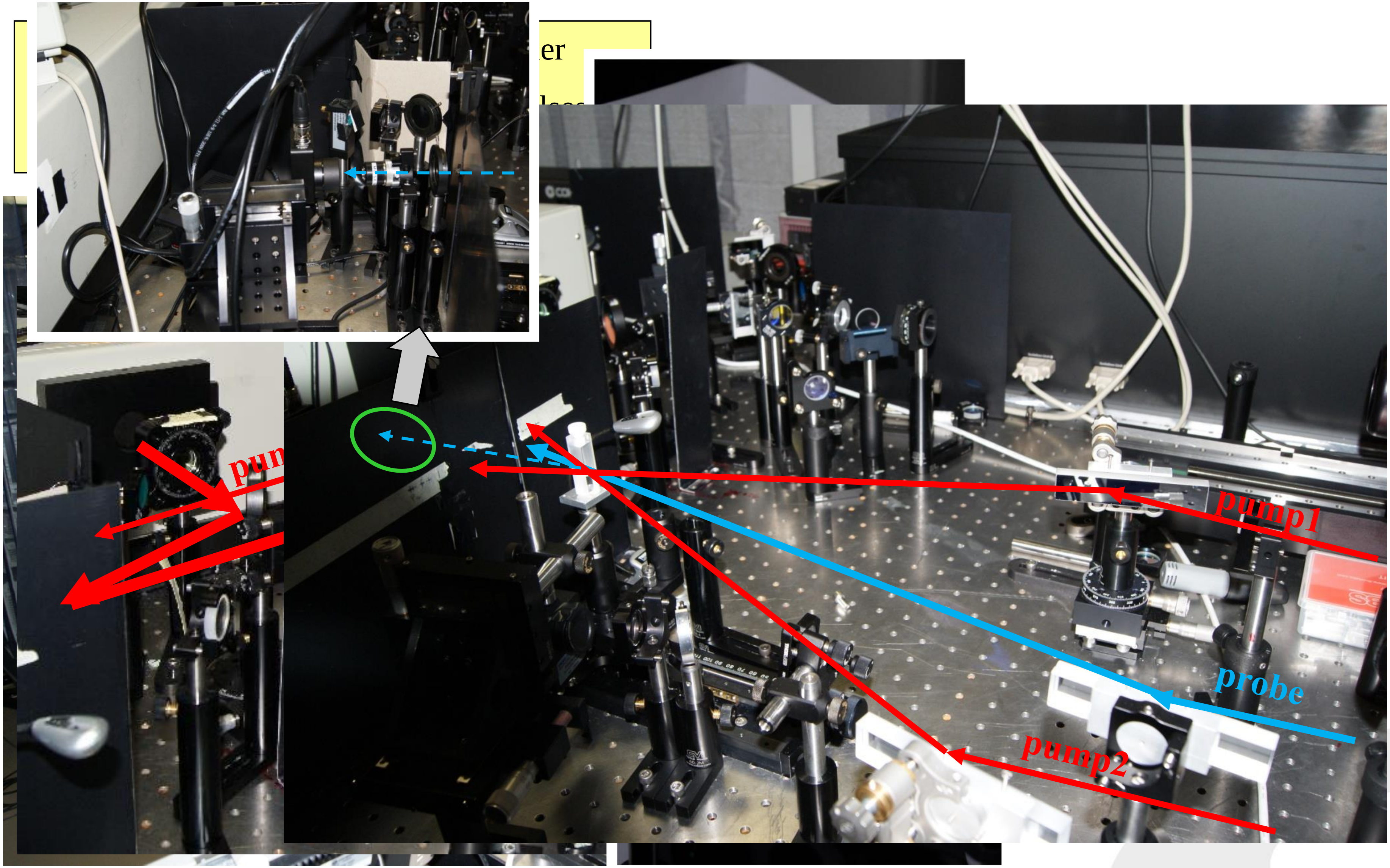
c_v: 840 J kg^{-1} K^{-1}

ρ: 2200 kg m^{-3}

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

Expected count rate: 1 – 10^4 counts per pulse

TG @ EIS laser lab



Layout tests @ laser lab

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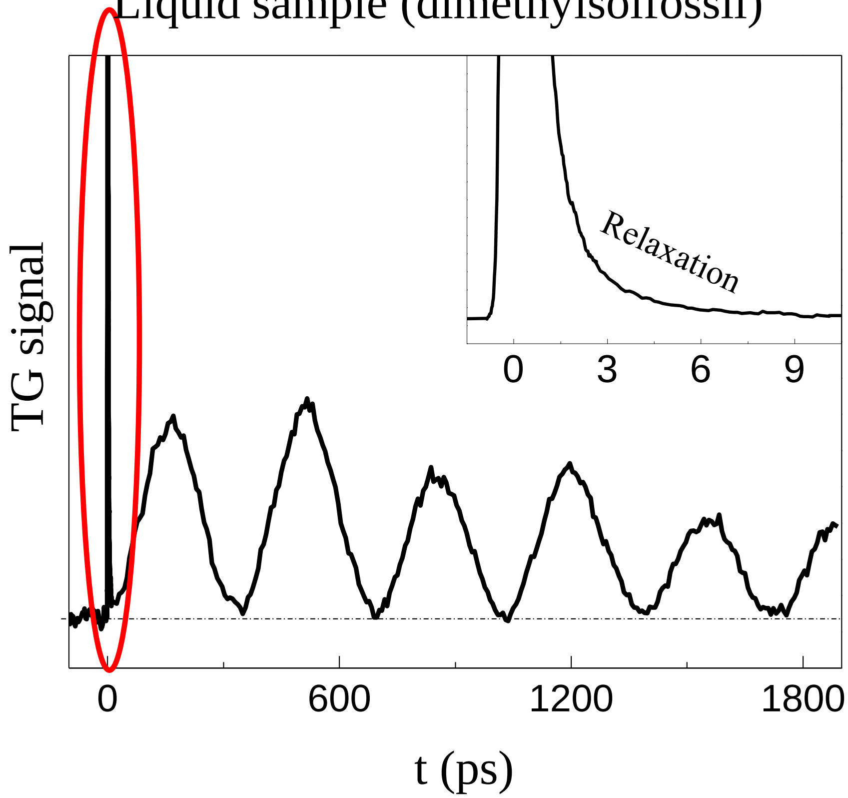
Liquid sample (dimethylsolfossil)



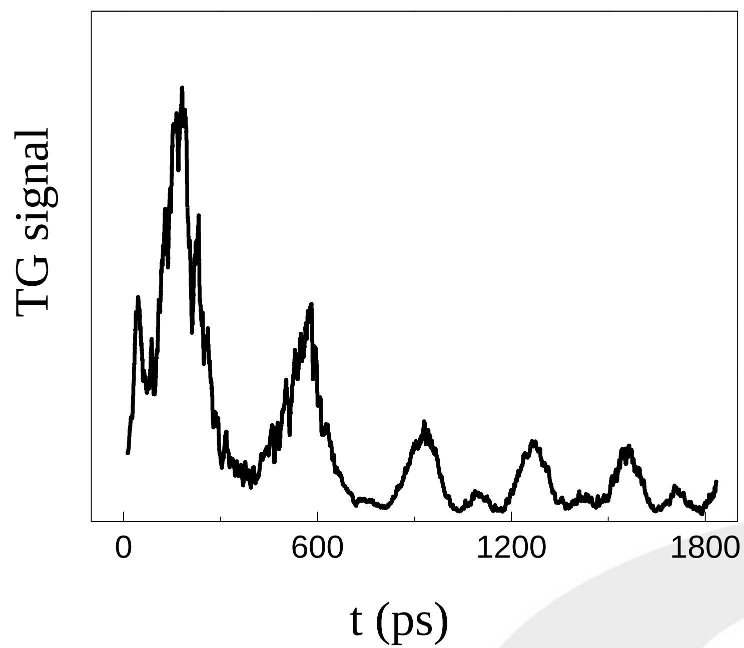
Pump energy ~ 25 μJ/pulse

Probe energy ~ 5 μJ/pulse

Beam dimensions ~ 50÷200 μm

$\theta = 26°$, Δt=150 fs, λ=800 nm

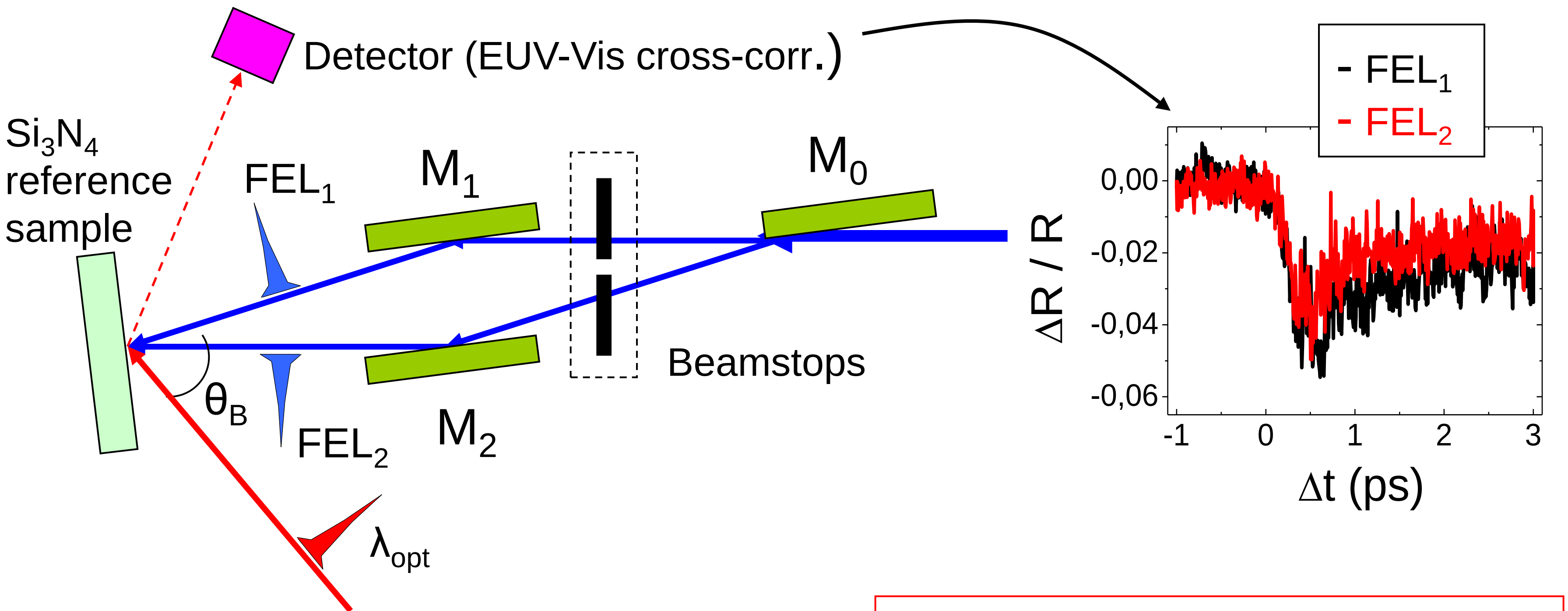
Surface (200 nm Au on Si)



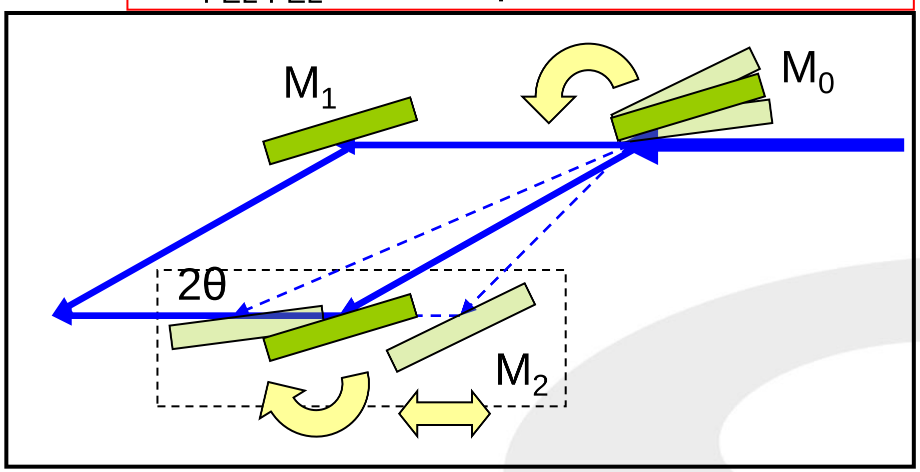
Sound velocity = 1486 +/- 2 m/s

R. Cucini et al et al., Opt. Lett (2011)

FEL Transient Grating Experiments on $V\text{-}SiO_2$

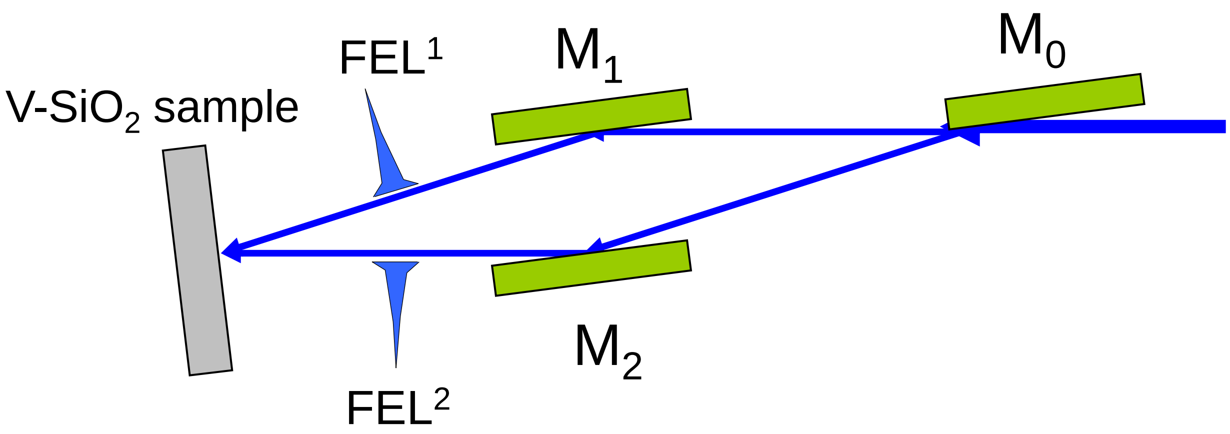


$\Delta t_{FEL\text{-}FEL} = \pm\ 0.5$ ps at 2θ = constant



F. Bencivenga et al., NIMA (2010)
R. Cucini et al., NIMA (2011)
R. Cucini et al., Opt. Lett. (2011)
F. Casolari et al., Appl. Phys. (2014)
M. Danailov et al. Opt. Express (2014)
R. Cucini et al., Opt. Lett. (2014)

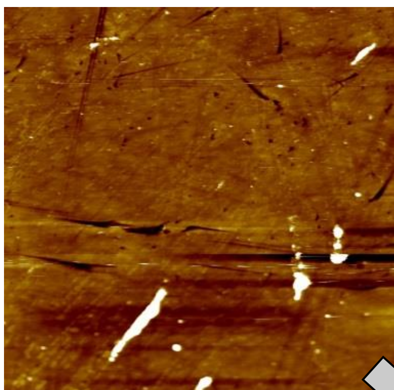
FEL Transient Grating Experiments on V-SiO_2



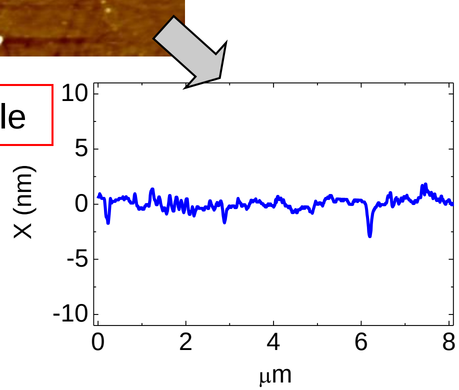
λ_{FEL} = 27.6 nm

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

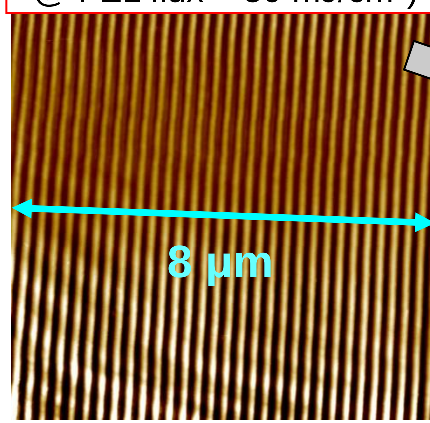
Grating visibility after multi-shot explosure
→ FEL^1-FEL^2 optical path difference < λ_{FEL}

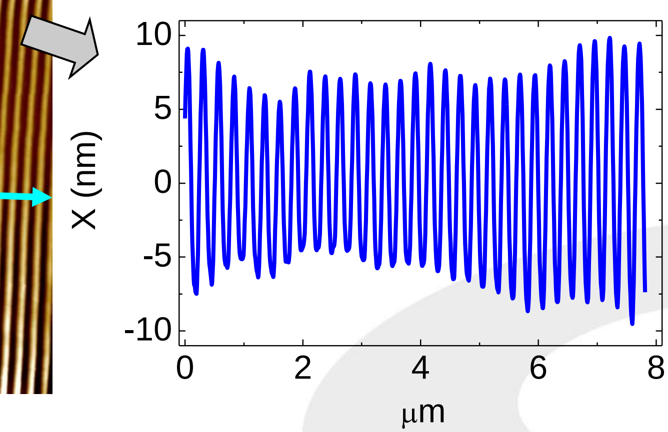


Clean sample



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





LETTER

doi:10.1038/nature1434

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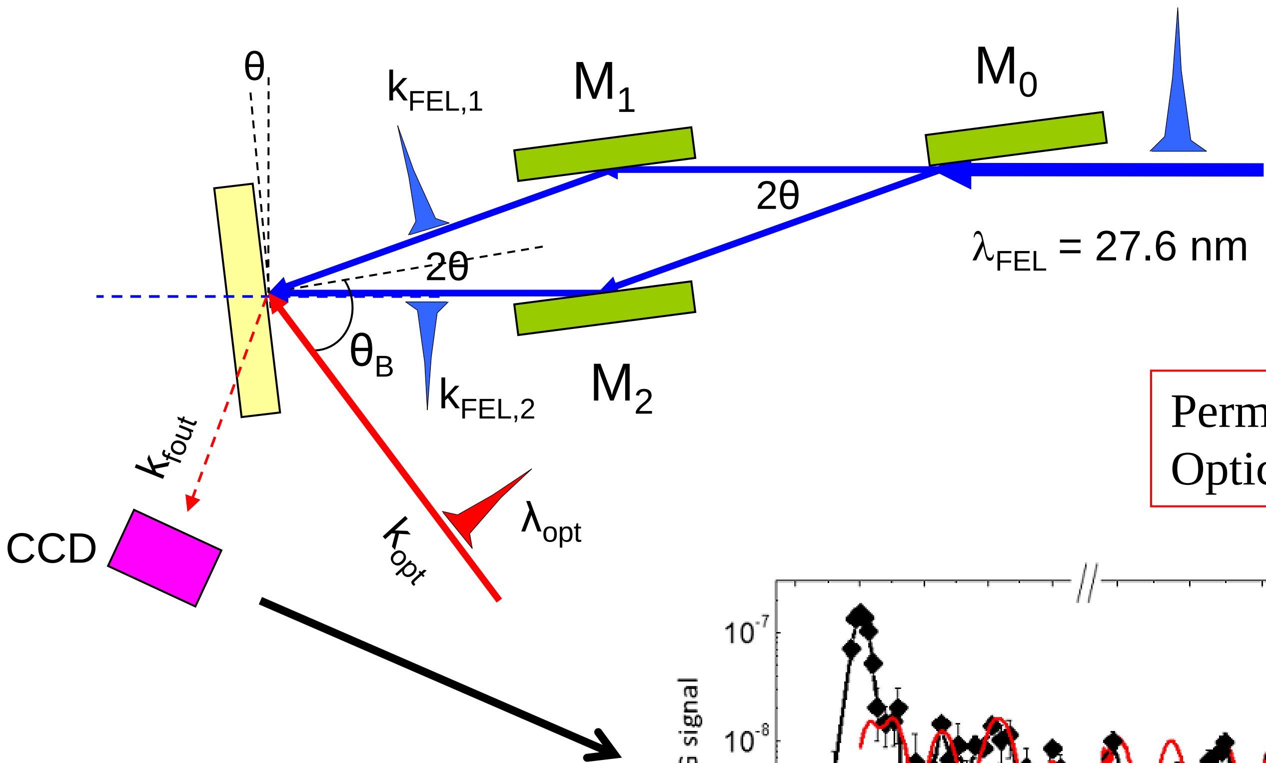
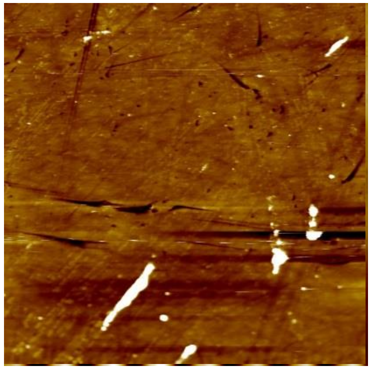
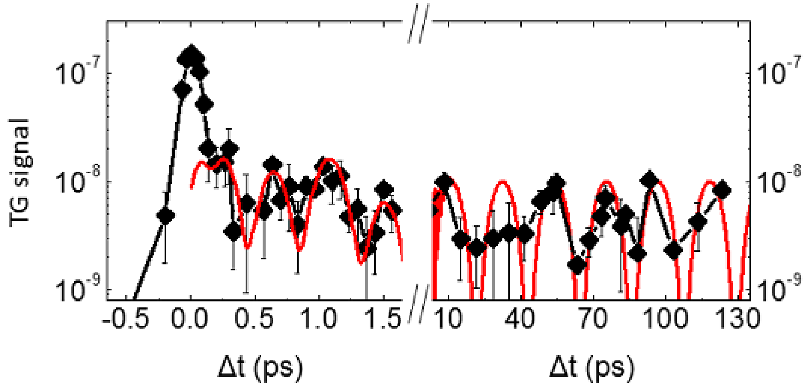
Four-wave mixing experiments with extreme ultraviolet transient gratings

F. Bencivenga et al., Nature 2015

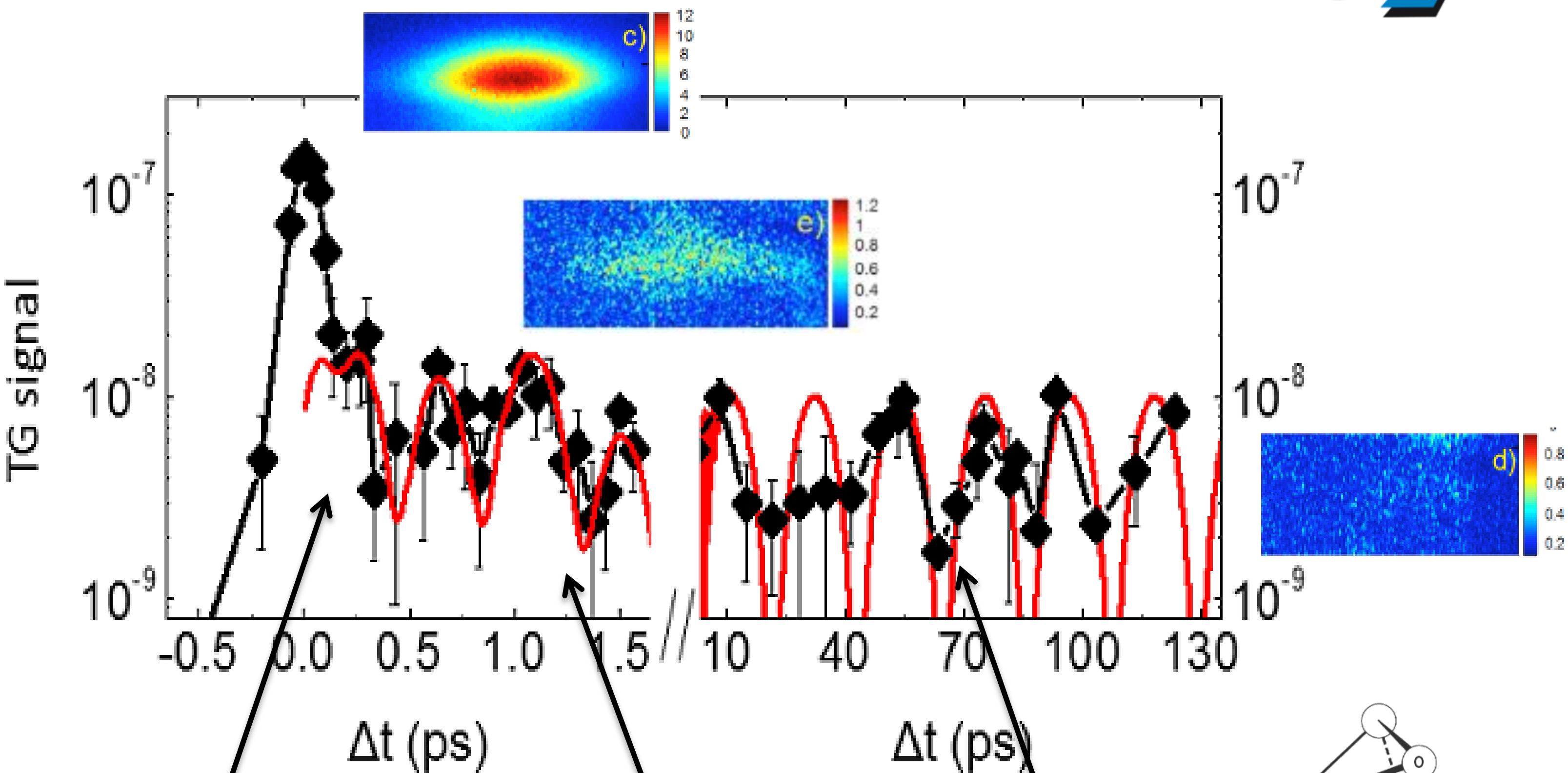


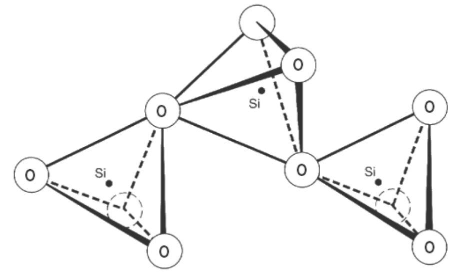


Permanent Grating after 1k-shots
Optical path difference < λ_{FEL}



Transient Grating Experiments on V-SiO_2





Hyper - Raman modes due to coupled tetrahedral rotations

Raman modes due to tetrahedral bending

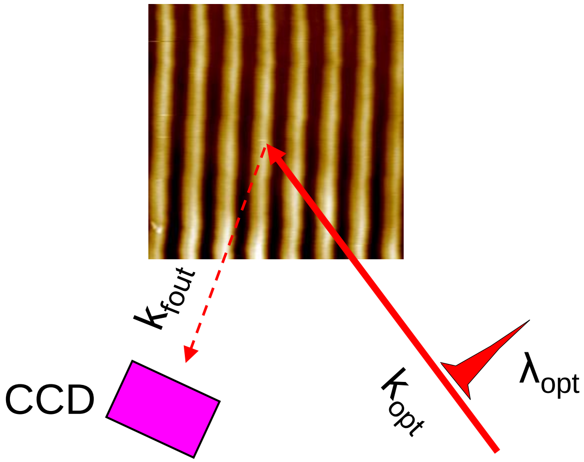
Acoustic-like excitations

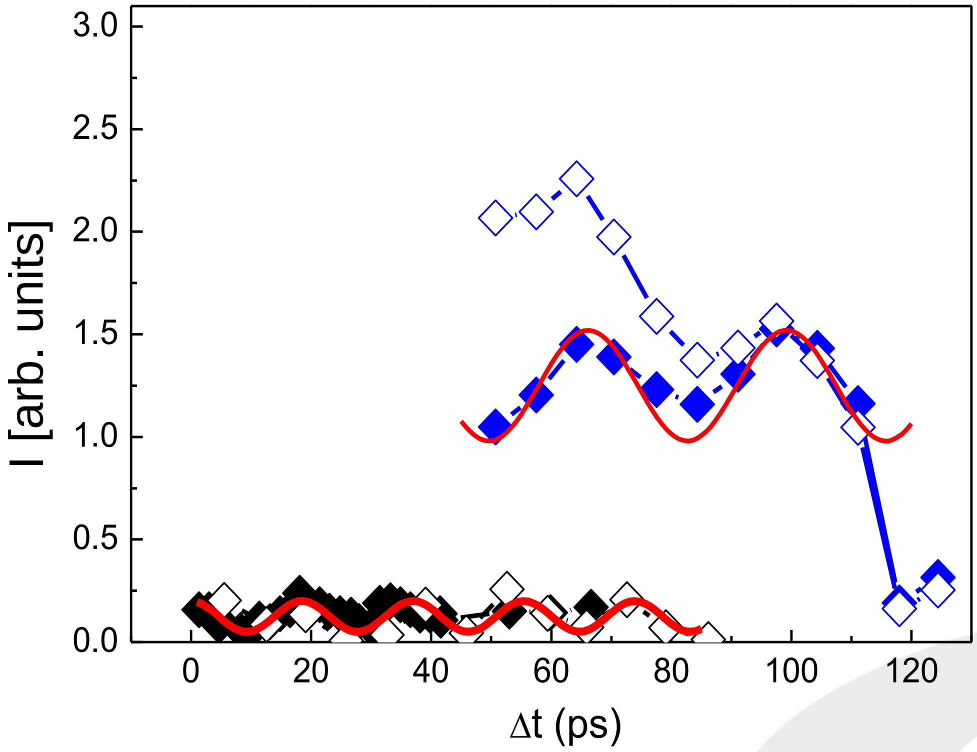
Heterodyning with FEL

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

$$I = |E_{\text{ref}}|^2 + |E_s(t)|^2 + 2|E_{\text{ref}} E_s(t)|\cos(\phi_{\text{ref}} - \phi_s)$$

Time independent local field





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.

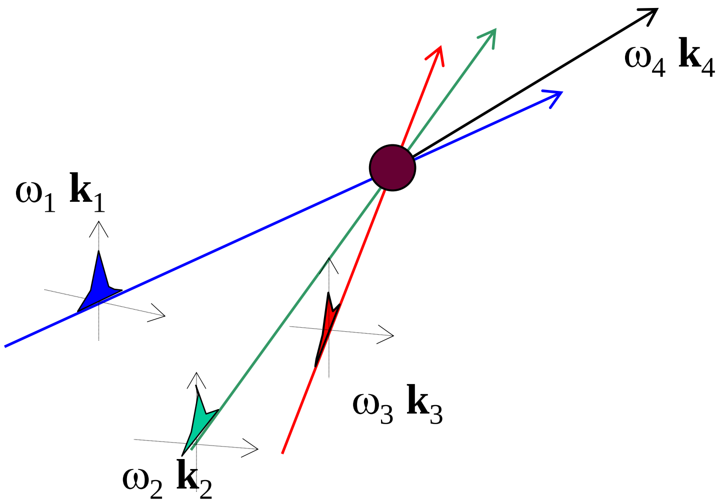
FOUR-WAVE MIXING SPECTROSCOPY

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

N. Bloembergen Nobel Lecture 1981

Elettra Sincrotrone Trieste

Transient grating is one of the possible application of **Four Wave Mixing** techniques:



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

VOLUME 89, NUMBER 4 PHYSICAL REVIEW LETTERS 22 JULY 2002

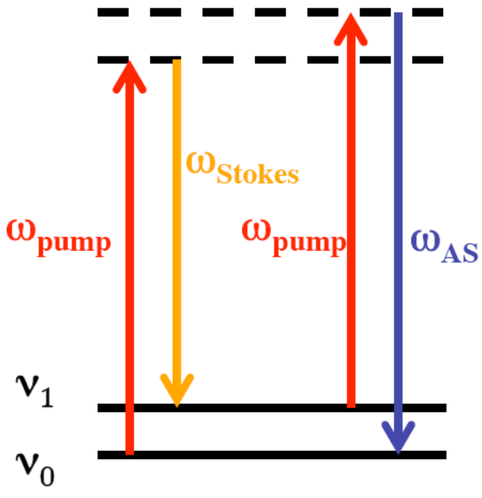
Coherent X-Ray Raman Spectroscopy: A Nonlinear Local Probe for Electronic Excitations

Satoshi Tanaka[1,3] and Shaul Mukamel[1,2]

[1]*Department of Chemistry, University of Rochester, Rochester New York, 14627*
[2]*Department of Physics and Astronomy, University of Rochester, Rochester New York, 14627*
[3]*College of Integrated Arts and Sciences, Osaka Prefecture University, Sakai 599-8531, Japan*
(Received 27 November 2001; published 9 July 2002)

Nonlinear x-ray four-wave mixing experiments are becoming feasible due to rapid advances in high harmonic generation and synchrotron radiation coherent x-ray sources. By tuning the difference of two x-ray frequencies across the valence excitations, it is possible to probe the entire manifold of molecular electronic excitations. We show that the wave vector and frequency profiles of this x-ray analogue of coherent Raman spectroscopy provide an excellent real-space probe that carries most valuable structural and dynamical information, not available from spontaneous Raman techniques.

VUV - CARS



Energy diagram

From **Vibrational** → **Core Hole** excitations

The **VUV-CARS** signal is a measure of 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.

VUV/soft-X-ray wavelength is comparable to the molecular size → the **dipole approximation** which imposes strict selection rules in optical spectroscopy **does not apply**, allowing the direct probing of the entire manifold of electronic transitions.

By tuning the VUV/soft-X-ray frequencies across the core excitation of various atoms, it becomes possible to **investigate** precisely the **nonlocal nature** of the **valence electronic excitation**.

VUV/soft-X-ray based CARS experiments allow the **direct observation** of excitation **energy transfer between atoms within a molecule**. This is an advantage over spontaneous X-ray Raman scattering which takes place at the same atomic site and cannot probe spatial coherence over different atoms.

nature COMMUNICATIONS

Elettra Sincrotrone Trieste

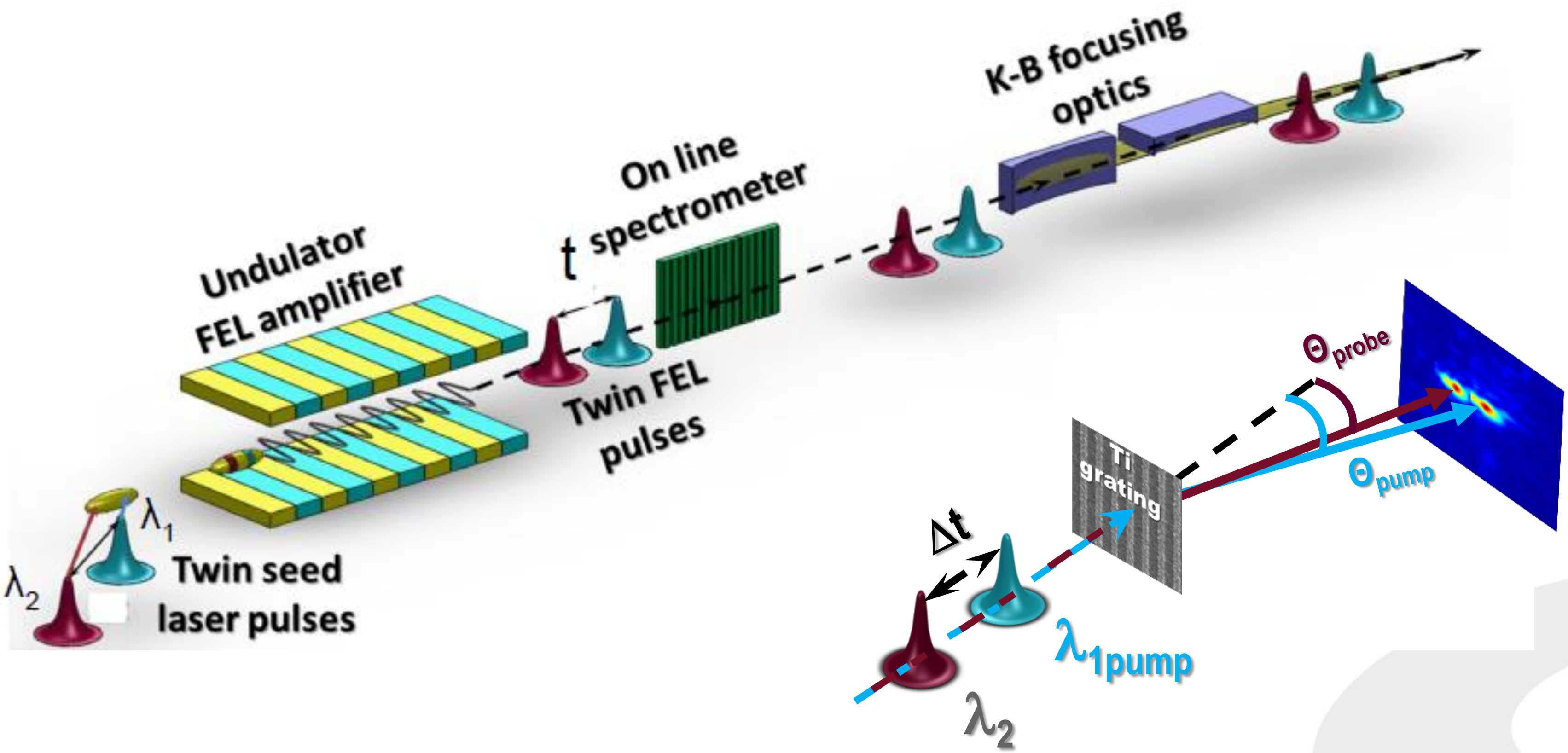
ARTICLE

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



Element selective magnetization dynamics

Elettra Sincrotrone Trieste

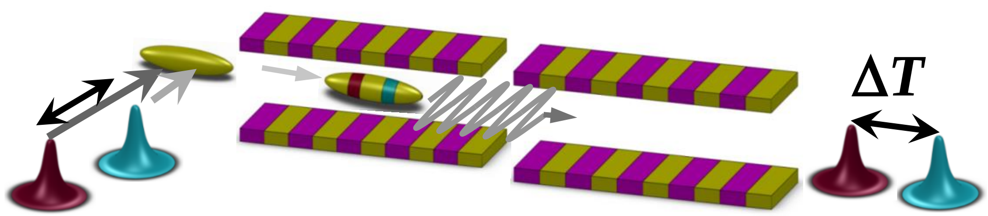
nature COMMUNICATIONS

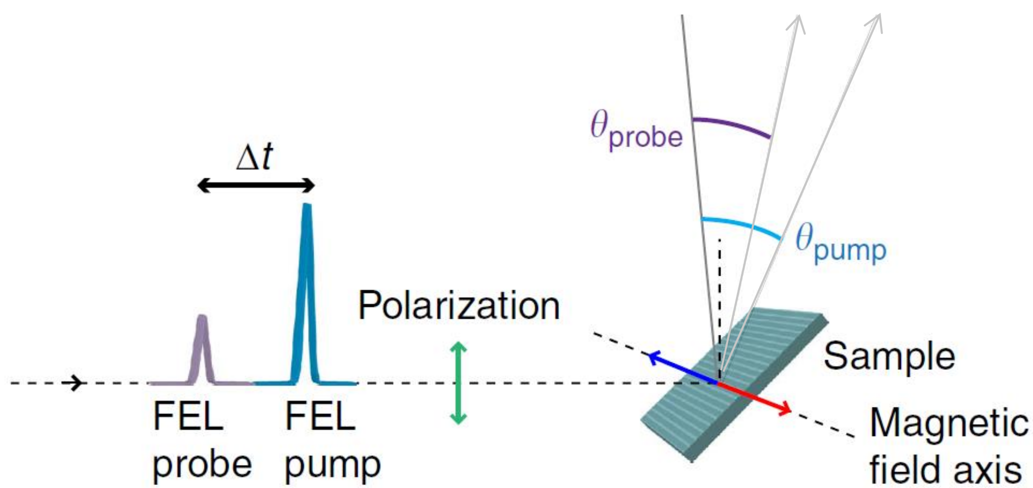
E. Ferrari et al., (2016)

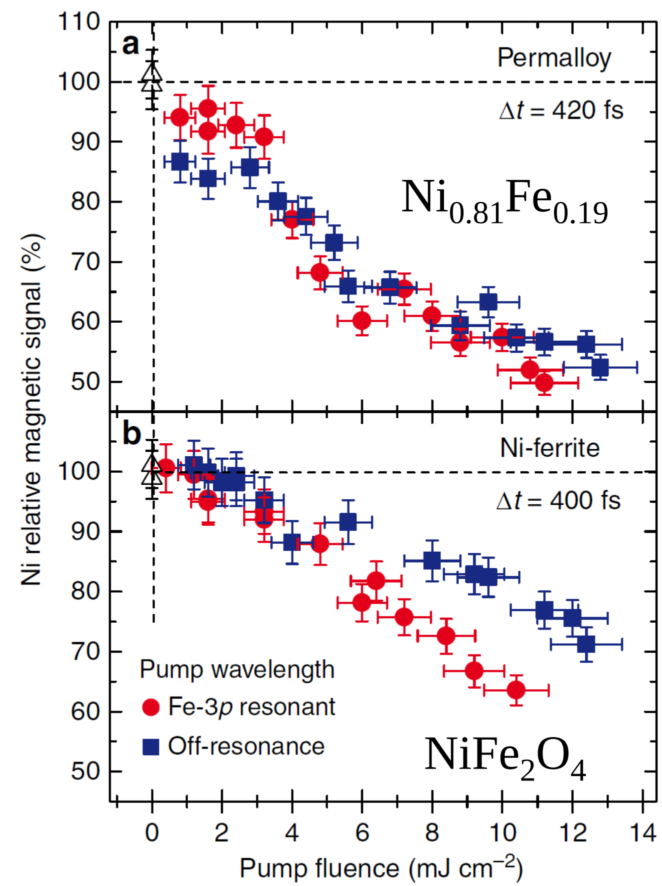
Received 21 Aug 2015 | Accepted 3 Dec 2015 | Published 13 Jan 2016 DOI: 10.1038/ncomms10343 OPEN

Widely tunable two-colour seeded free-electron laser source for resonant-pump resonant-probe magnetic scattering

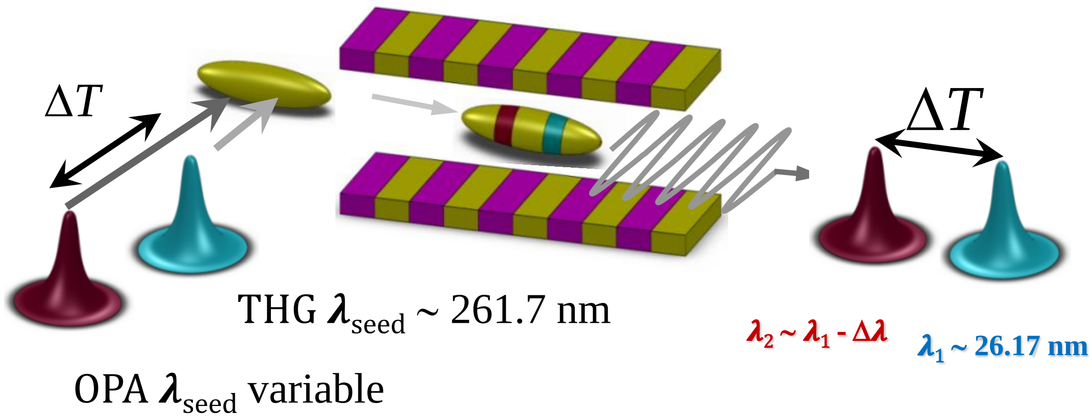
Radiators at different harmonics

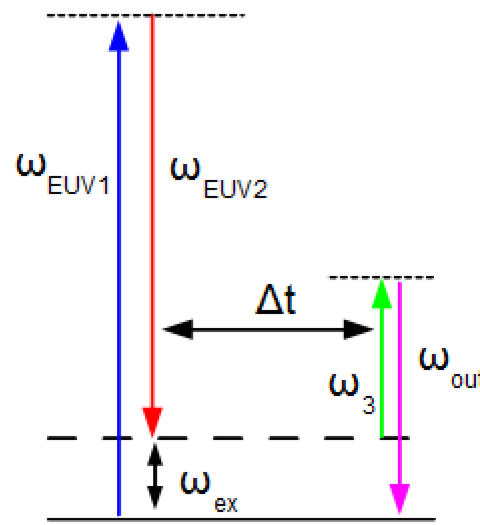


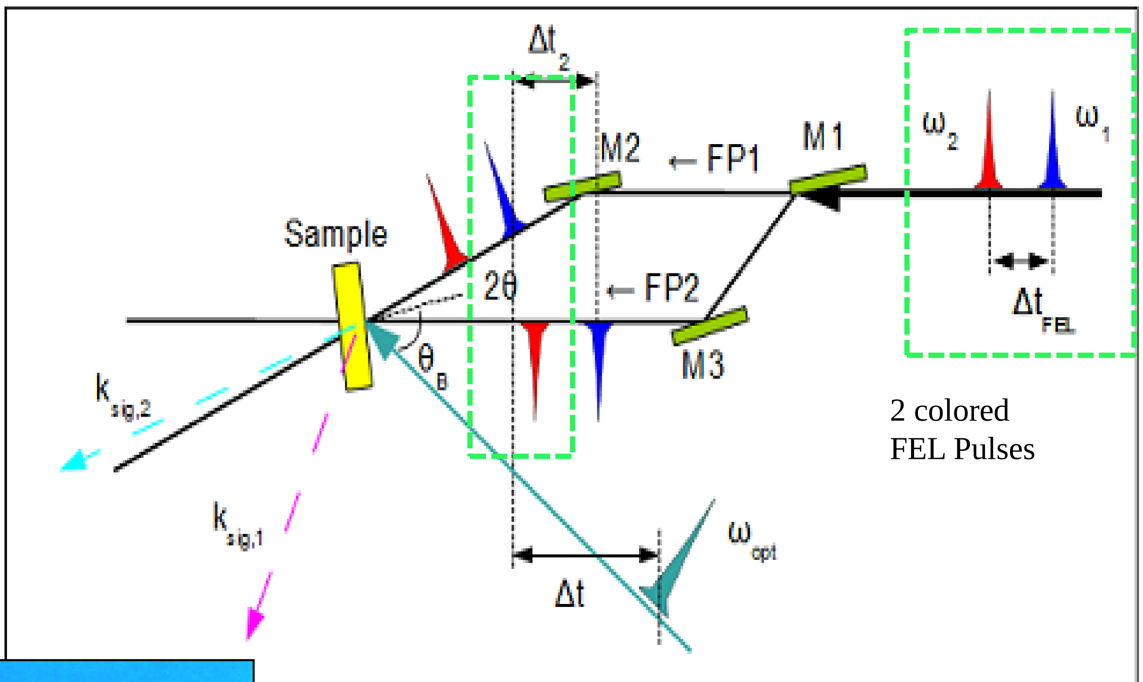


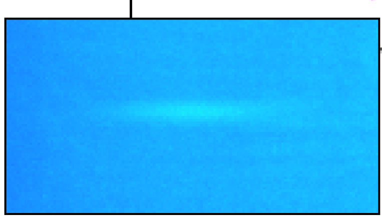


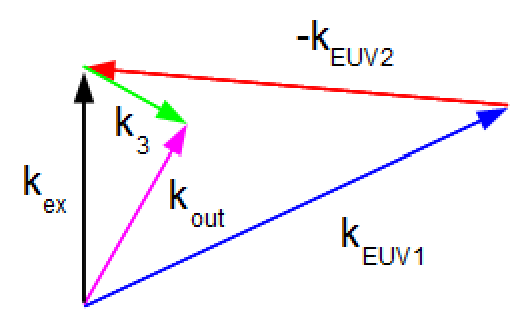
FERMI based CARS





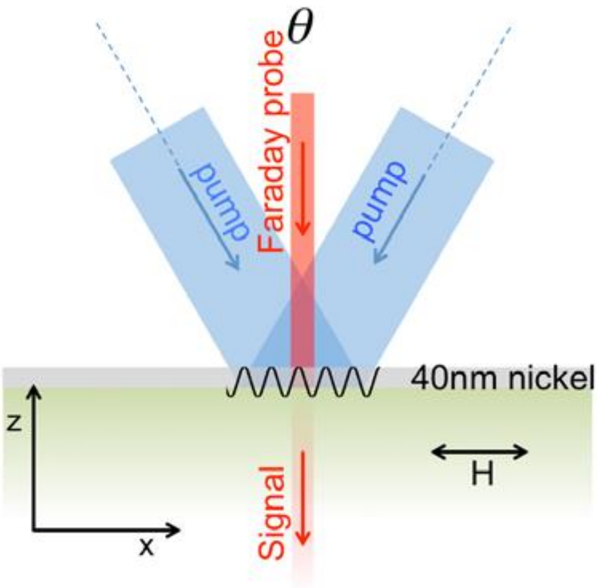




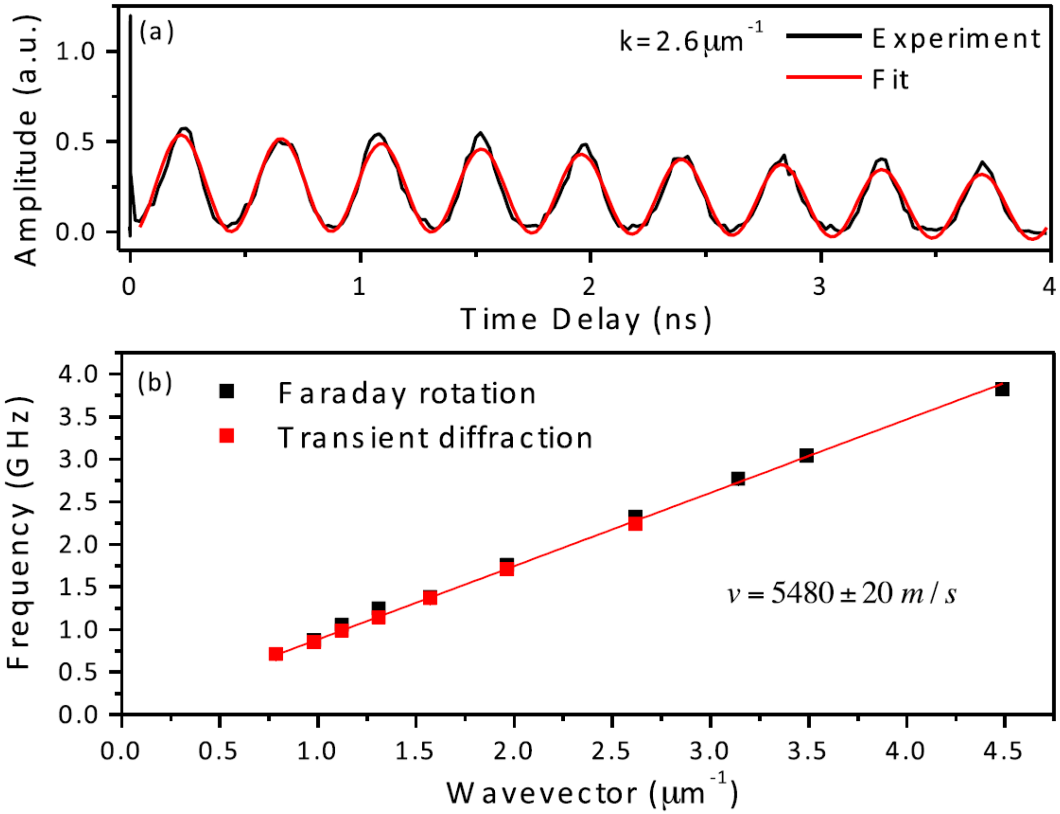


F. Bencivenga et al., in preparation

Optical TG in FM thin films



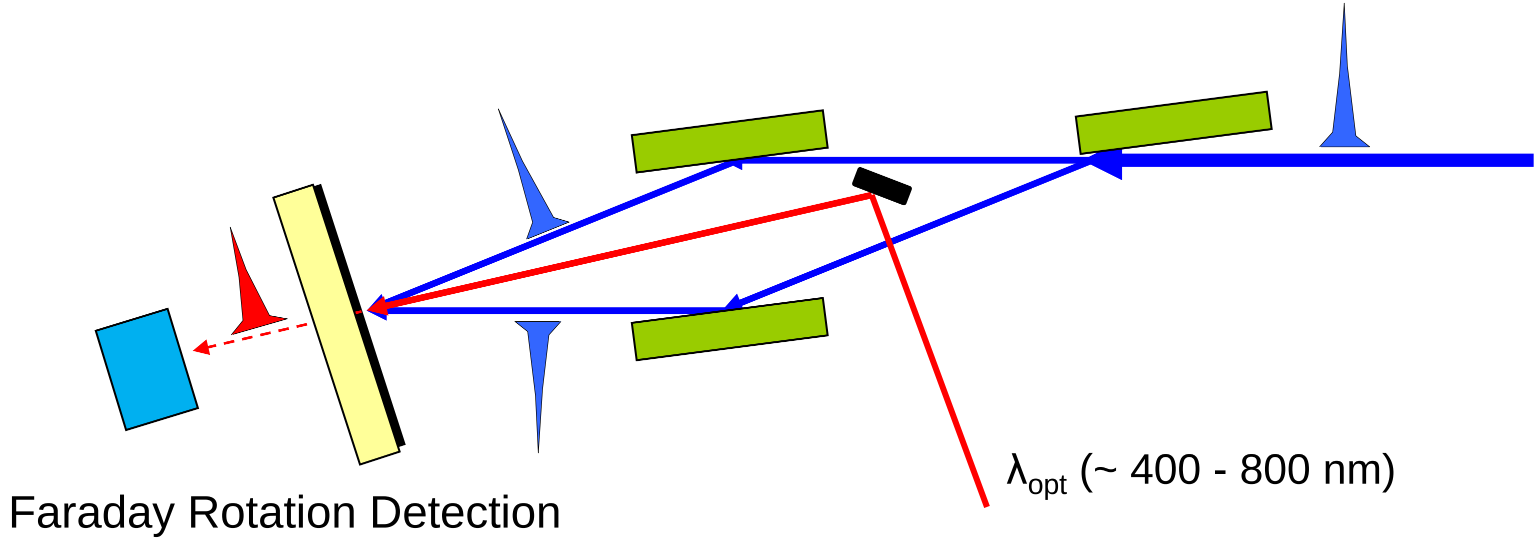
Optical TG used for probing magneto-elastic waves in Ni
J.Janusonis et al., APL (2015)

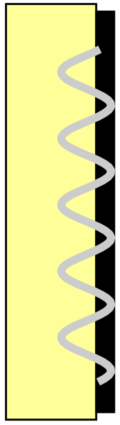


Faraday and TG exhibits the **same** dispersion

one-to-one correspondence
between **acoustic** and **magnetic** response

Sub-Optical TG in FM thin films





Study of

1) Magneto – Acoustic **coupling**
2) Q – dependence of Magnetic Waves
3) Excite with FEL short λ and probe with Faraday detector to *see* the acoustic waves

Conclusions

T. Scopigno

K. Nelson

M. Chergui

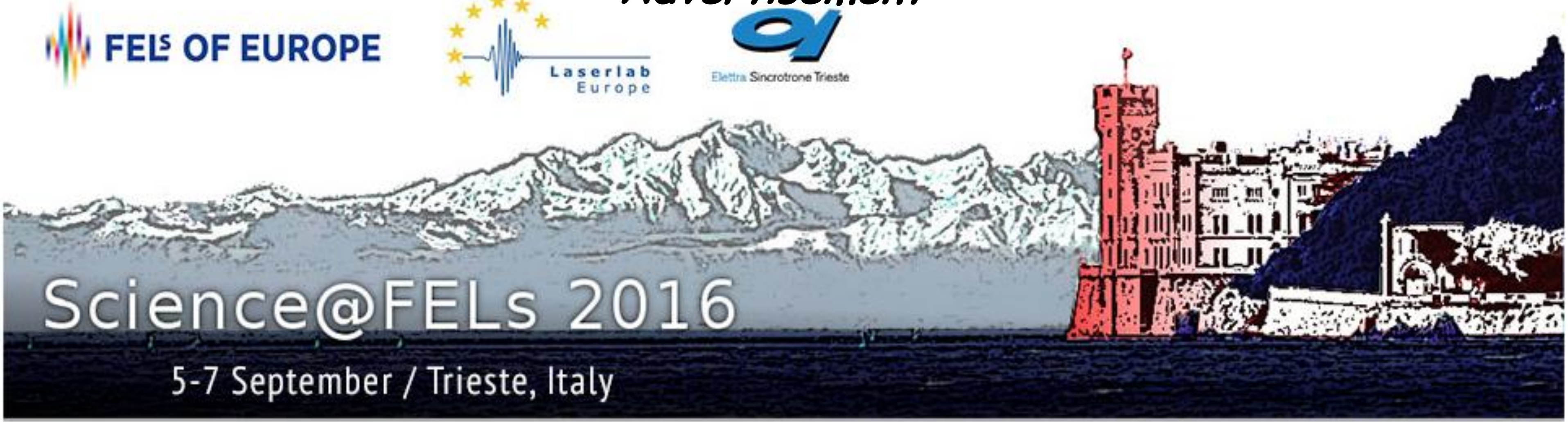
- Charge transfer dynamics in **metal complexes**
- Charge injection and transport in **metal oxides nanoparticles**
- Vibrational modes in **Glasses**
- Charge **Density Wave**
- Quasiparticle diffusion (**Polarons**)
- Sound velocity in **Graphene**

G. Knopp

S. Mukamel

G. Monaco

Advertisement



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Shaul Mukamel (UCI)

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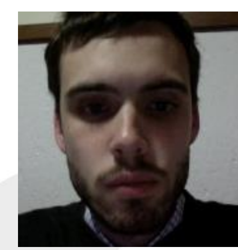
E. Principi


R. Mincigrucci