Resonant Spectroscopies at Compact FELs

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CompactLight Workshop 27 – 28 November 2018, Genève, Switzerland

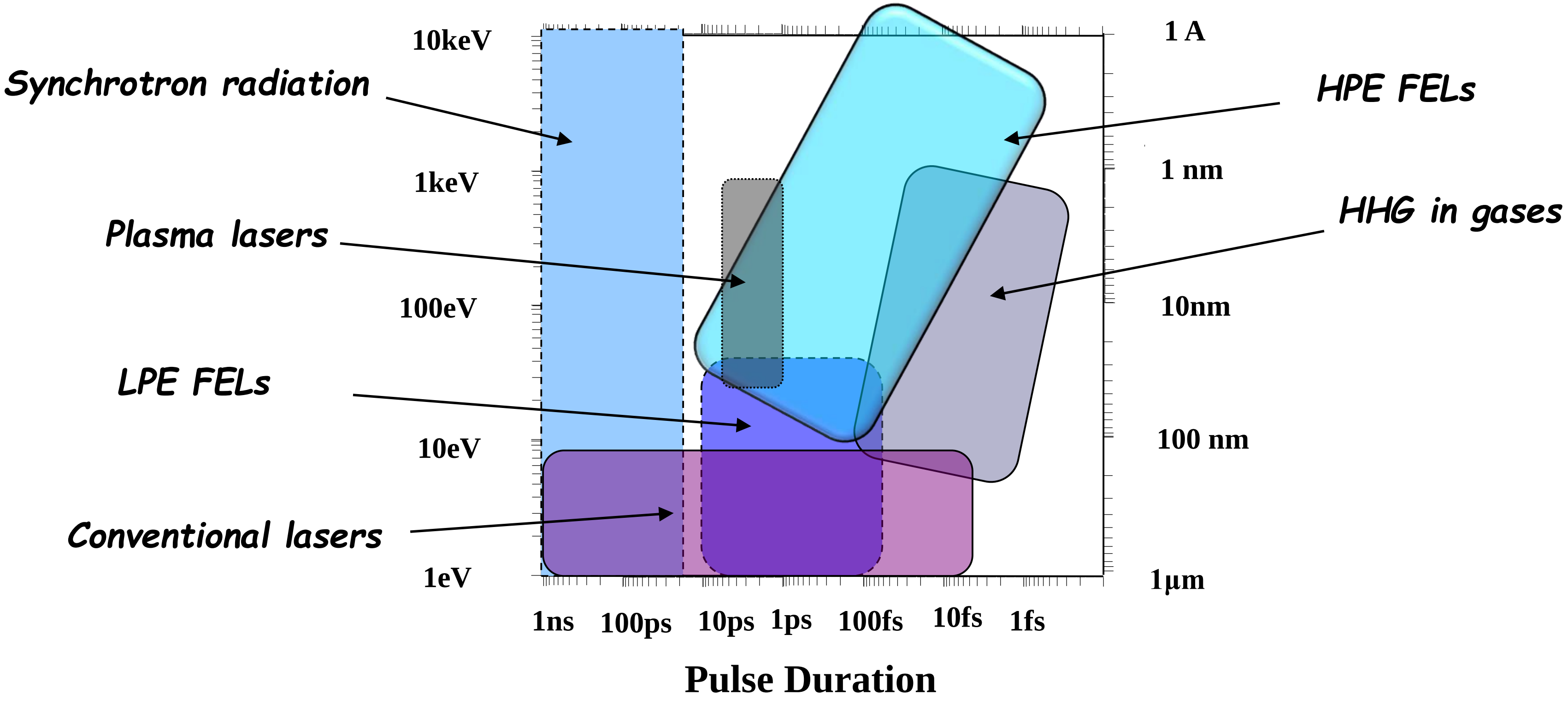
Machine vs Users

Ideal Beamline from the Accelerator Side

Ideal Machine from the Users Side

$\Delta E\ \Delta t \ll \hbar$

Why Free Electron Lasers ?

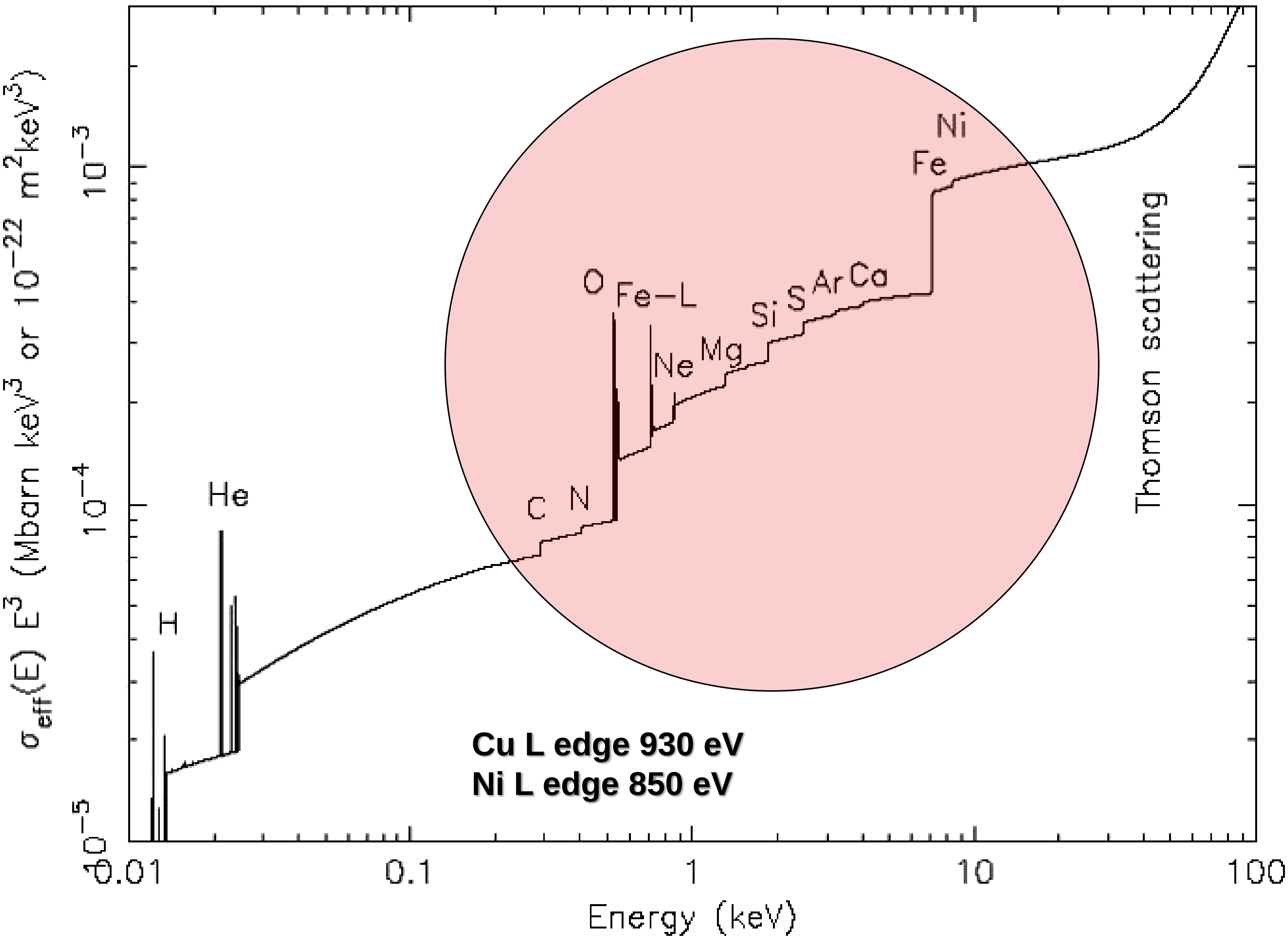


Imaging with high Spatial Resolution (~ λ): fixed target imaging, particle injection imaging,..

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

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

Resonances



Cu L edge 930 eV
Ni L edge 850 eV

nature photonics

ARTICLES

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

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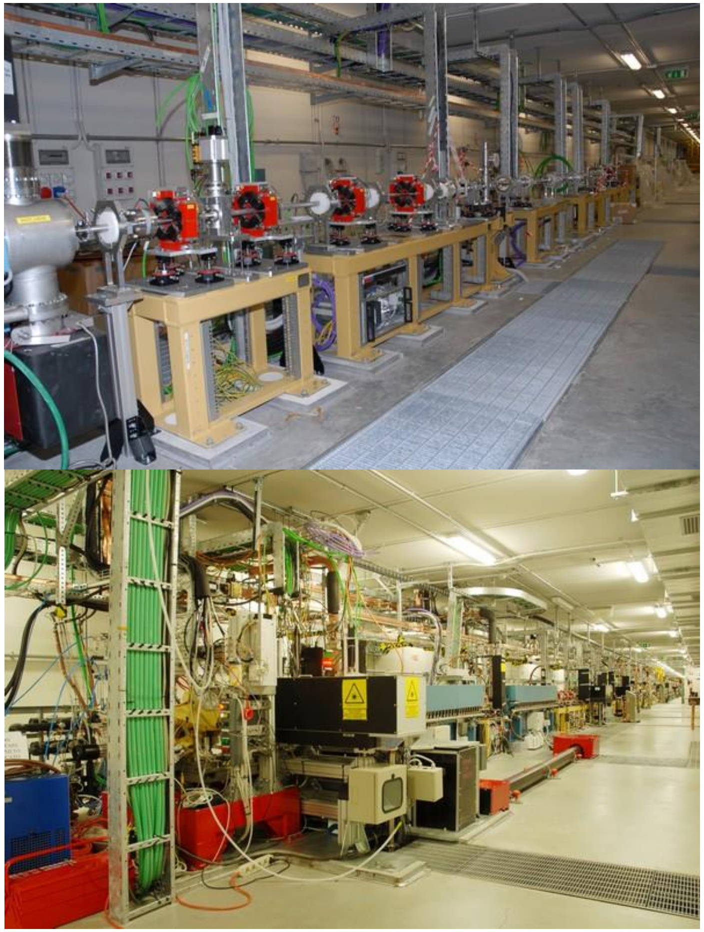
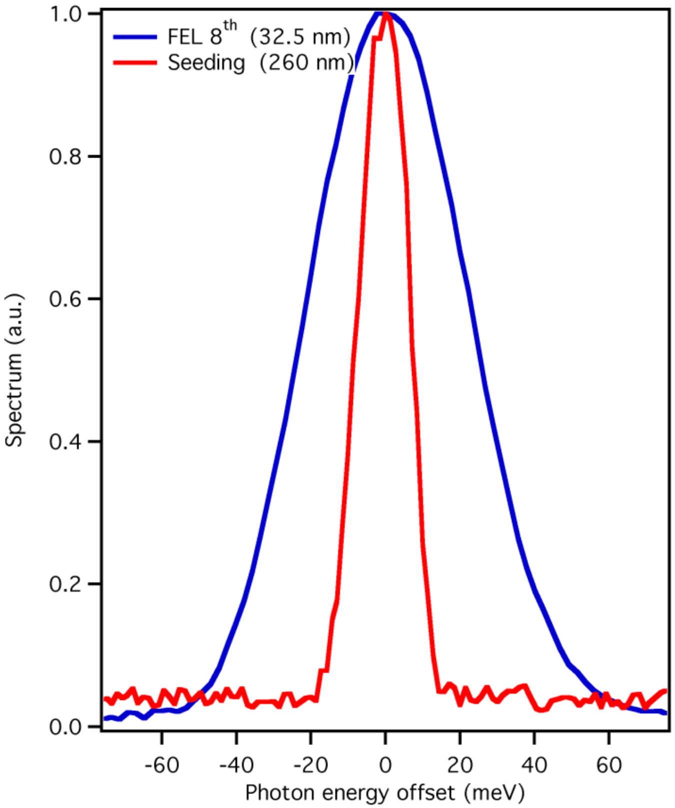
Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet

E. Allaria et al (2012)





$\Delta t < 100$ fs

Flux $< 10^{14}$ ph/pulse

E ~ 20 - 400 eV

Total Control on

Pulse Energy

Time Shape

Polarization

Ultrafast Demagnetization Imaging

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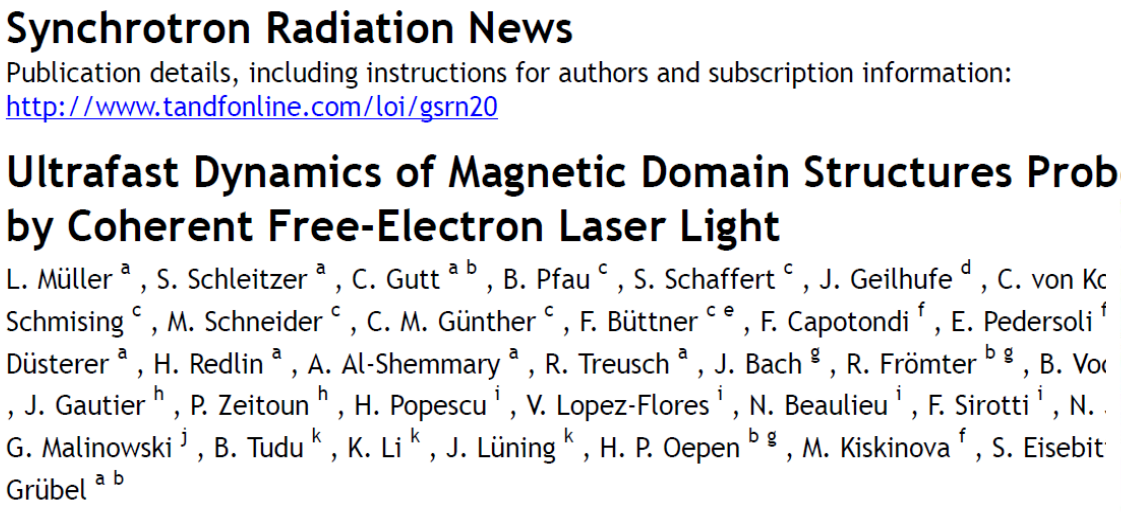
Ultrafast Magnetic Dynamics

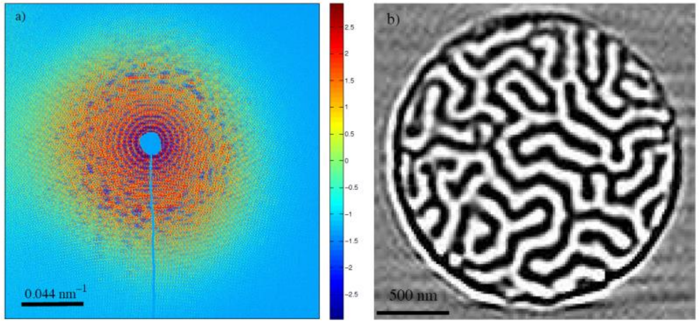


Synchrotron Radiation News

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

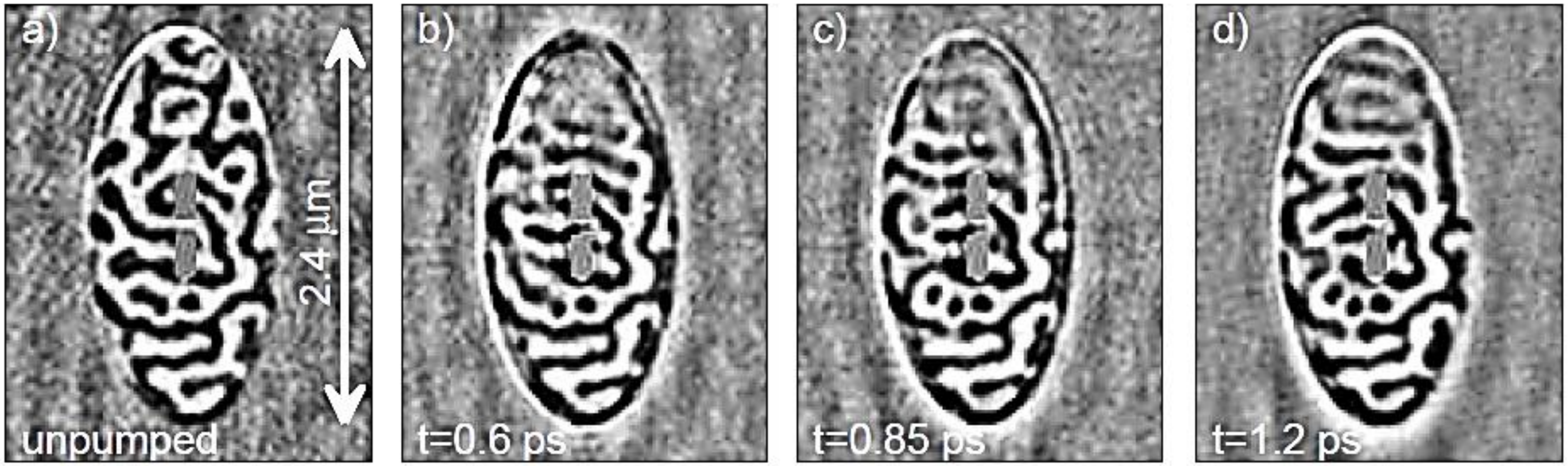
Ultrafast Dynamics of Magnetic Domain Structures Probed by Coherent Free-Electron Laser Light

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. Vo
, 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, PRL (2014)

Non-linear Optics

N. Bloembergen 1981

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

Non linear techniques are **powerful** when one wants

to measure **sample properties** that cannot be addressed by ***conventional*** linear optical spectroscopy or to obtain spectroscopic information with a higher **sensitivity** than that associated with linear spectroscopy

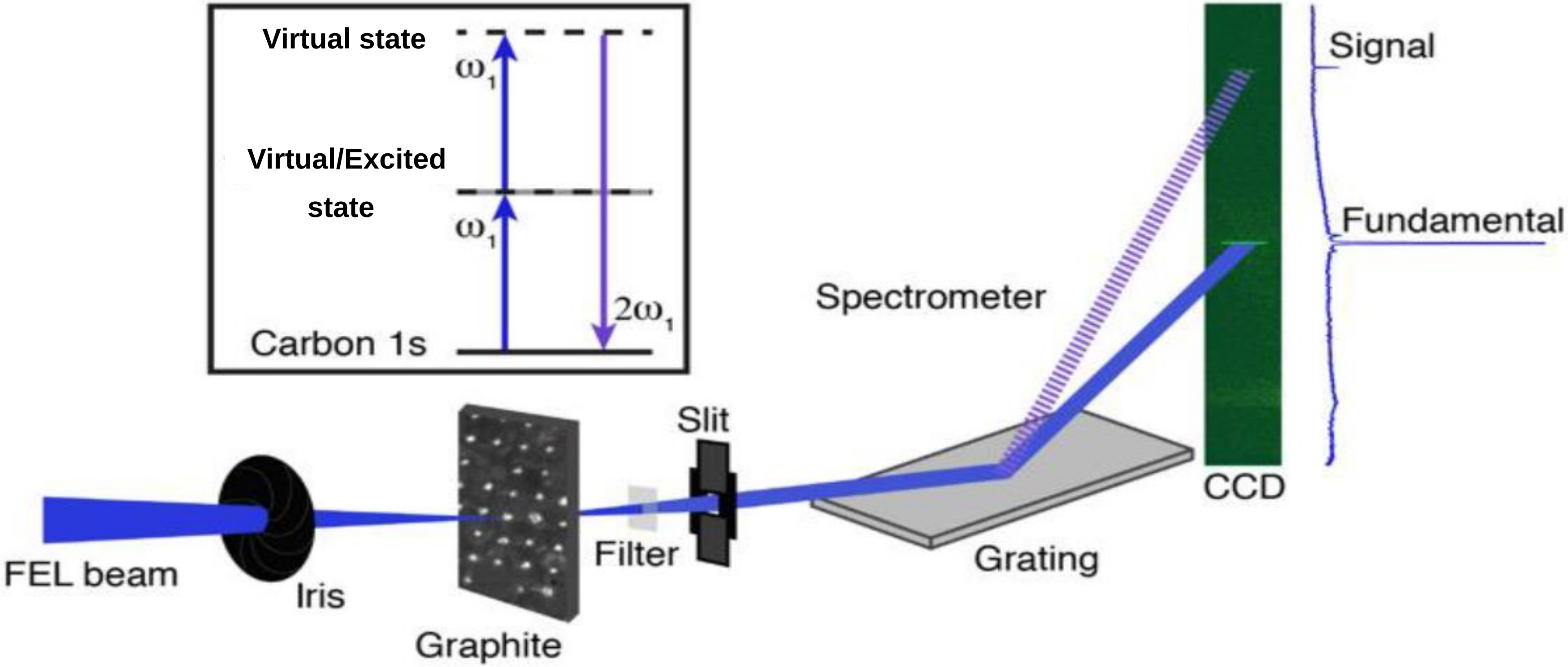
$$\begin{aligned}\frac{\boldsymbol{P}^{(2)}}{\varepsilon_0} = {} & [\chi^{(2)}\boxed{(2\cdot\omega_1)}\cdot \boldsymbol{E}_1'^2 \cdot \exp(-2\cdot i\cdot\omega_1\cdot t) + \chi^{(2)}\boxed{(2\cdot\omega_2)}\cdot \boldsymbol{E}_2'^2 \cdot \exp(-2\cdot i\cdot\omega_2\cdot t) \\ & + 2\cdot\chi^{(2)}\boxed{(\omega_1+\omega_2)}\cdot \boldsymbol{E}_1'\cdot \boldsymbol{E}_2'\cdot \exp(-i\cdot(\omega_1+\omega_2)\cdot t) \\ & + 2\cdot\chi^{(2)}\boxed{(\omega_1-\omega_2)}\cdot \boldsymbol{E}_1'\cdot \boldsymbol{E}_2'\cdot \exp(-i\cdot(\omega_1-\omega_2)\cdot t) + \mathrm{C.C.}] \\ & + 2\cdot\chi^{(2)}(\omega=0)\cdot[\boldsymbol{E}_1'\cdot \boldsymbol{E}_2'^* + \boldsymbol{E}_1'^*\cdot \boldsymbol{E}_2']\end{aligned}$$

SHG, SFG, DFG

Non-linear Optics

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

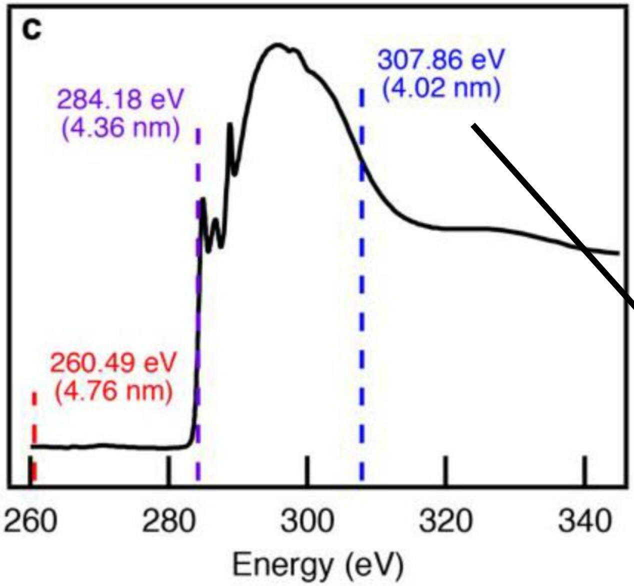
SHG



R. Lam et al., PRL (2018)

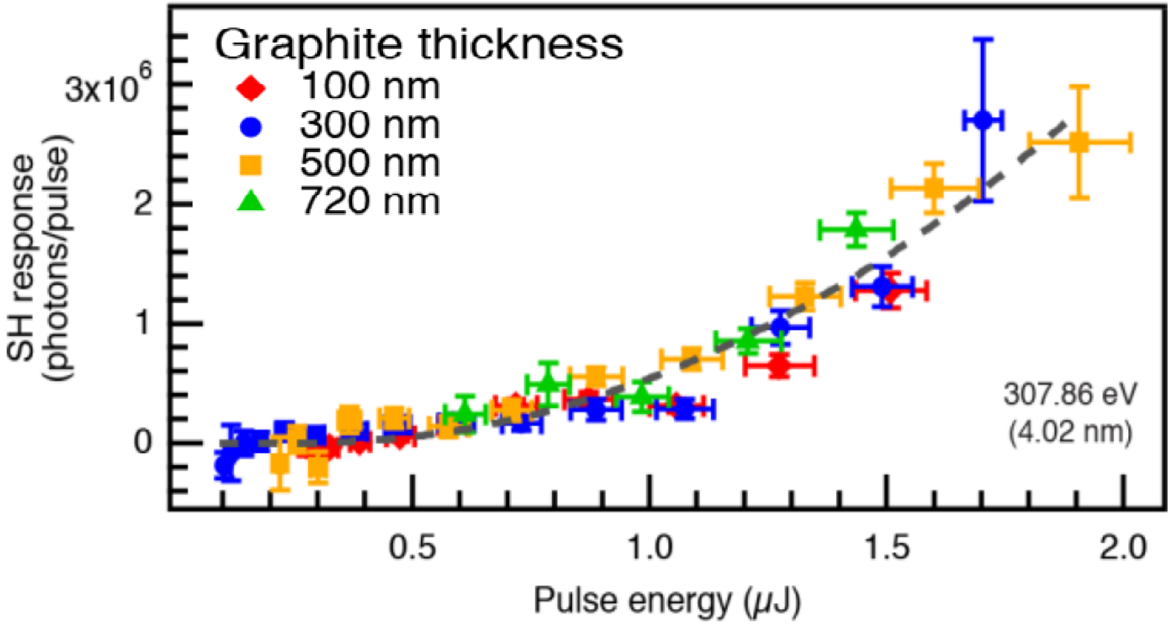
Second Harmonic Generation at FERMI

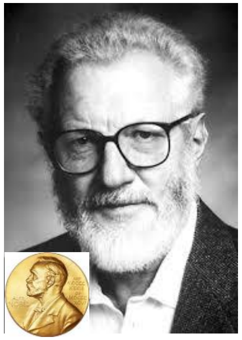
Elettra Sincrotrone Trieste



NO thickness dependence of SHG signal

Measure local geometric and/or electronic structure of a **single atomic layer**

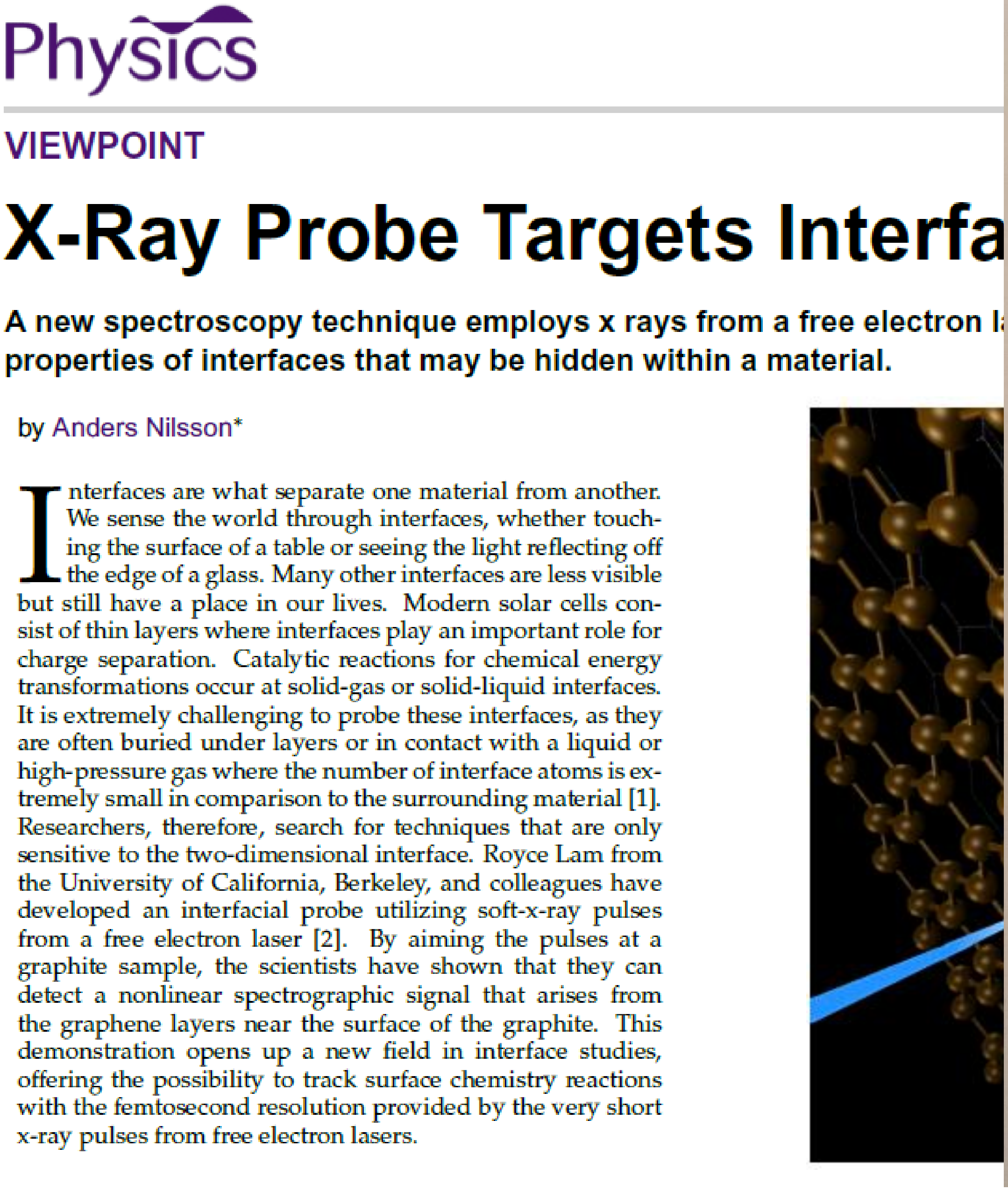




H. Kroemer 2000

<u>The interface is the device</u>!

Second Harmonic Generation



Physics

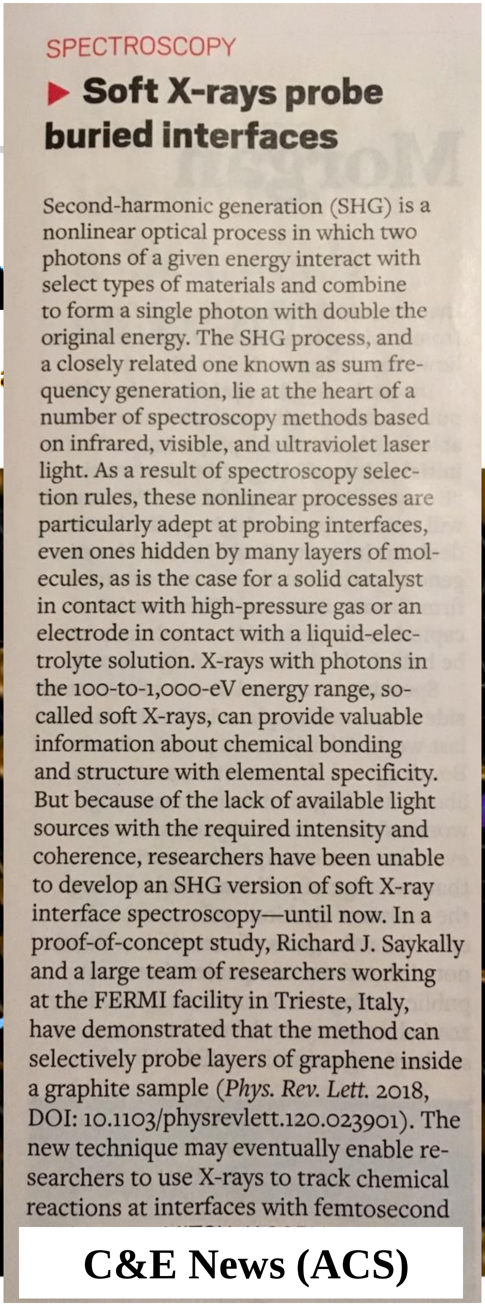
VIEWPOINT

X-Ray Probe Targets Interfa

A new spectroscopy technique employs x rays from a free electron l properties of interfaces that may be hidden within a material.

by Anders Nilsson*

Interfaces are what separate one material from another. We sense the world through interfaces, whether touching the surface of a table or seeing the light reflecting off the edge of a glass. Many other interfaces are less visible but still have a place in our lives. Modern solar cells consist of thin layers where interfaces play an important role for charge separation. Catalytic reactions for chemical energy transformations occur at solid-gas or solid-liquid interfaces. It is extremely challenging to probe these interfaces, as they are often buried under layers or in contact with a liquid or high-pressure gas where the number of interface atoms is extremely small in comparison to the surrounding material [1]. Researchers, therefore, search for techniques that are only sensitive to the two-dimensional interface. Royce Lam from the University of California, Berkeley, and colleagues have developed an interfacial probe utilizing soft-x-ray pulses from a free electron laser [2]. By aiming the pulses at a graphite sample, the scientists have shown that they can detect a nonlinear spectrographic signal that arises from the graphene layers near the surface of the graphite. This demonstration opens up a new field in interface studies, offering the possibility to track surface chemistry reactions with the femtosecond resolution provided by the very short x-ray pulses from free electron lasers.



SPECTROSCOPY

▶ Soft X-rays probe buried interfaces

Second-harmonic generation (SHG) is a nonlinear optical process in which two photons of a given energy interact with select types of materials and combine to form a single photon with double the original energy. The SHG process, and a closely related one known as sum frequency generation, lie at the heart of a number of spectroscopy methods based on infrared, visible, and ultraviolet laser light. As a result of spectroscopy selection rules, these nonlinear processes are particularly adept at probing interfaces, even ones hidden by many layers of molecules, as is the case for a solid catalyst in contact with high-pressure gas or an electrode in contact with a liquid-electrolyte solution. X-rays with photons in the 100-to-1,000-eV energy range, so-called soft X-rays, can provide valuable information about chemical bonding and structure with elemental specificity. But because of the lack of available light sources with the required intensity and coherence, researchers have been unable to develop an SHG version of soft X-ray interface spectroscopy—until now. In a proof-of-concept study, Richard J. Saykally and a large team of researchers working at the FERMI facility in Trieste, Italy, have demonstrated that the method can selectively probe layers of graphene inside a graphite sample (*Phys. Rev. Lett.* 2018, DOI: 10.1103/physrevlett.120.023901). The new technique may eventually enable researchers to use X-rays to track chemical reactions at interfaces with femtosecond

C&E News (ACS)

Liquid Carbon

NEWS AND VIEWS

CONDENSED STATE

First light on fluid carbon

Nicolaas Bloembergen

iversity of
caught the
carbon. D.
C. Downer
(B45, 2677–
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tures of ab-
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vessel, be-
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perature re-
of carbon is
ture of this
se studying
but because
these conditions can be found in planetary interiors, it is also important for geophysics and astrophysics.

The first evidence for this phase came from experiments using pulsed ohmic heating of graphite electrodes. Shock-wave studies and pulsed-laser heating of graphite and diamond have also been tried (see F. B. Bundy's review in *Physica* **156**A, 169–178; 1989). Downer and colleagues used laser pulses lasting less than a picosecond (10^{-12} s) in their new experiments, both to create and to probe their fluid carbon.

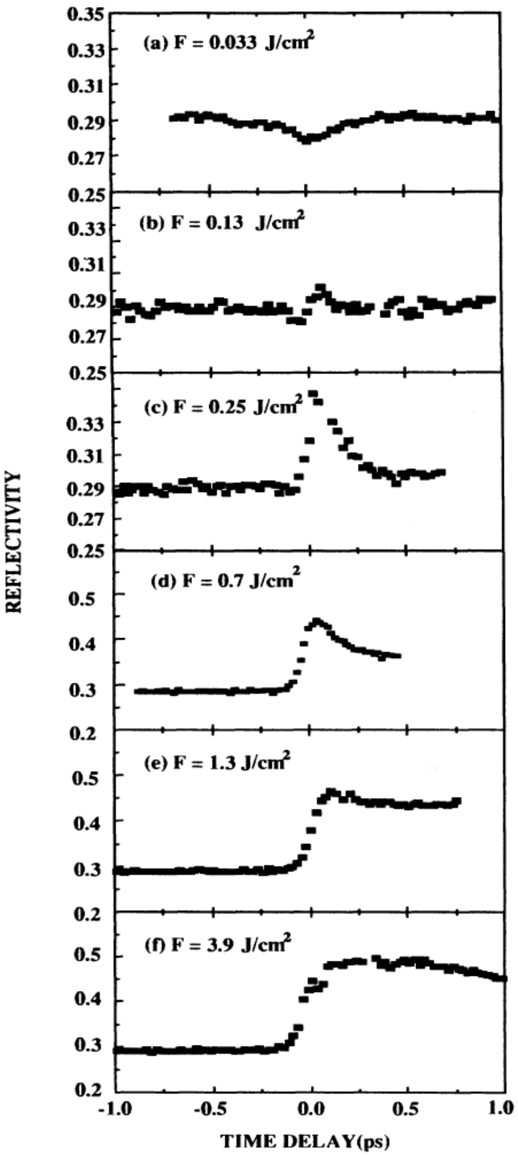
It is well known that short, intense laser pulses are capable of turning any substance into a high-temperature, high-density plasma. By careful control of the energy fluence in the pump pulse it is possible to determine a threshold for melting as the first stage in the process of plasma formation. A second, probe pulse will be reflected or will generate second-harmonic (or frequency-doubled) light, and the change in either of these can be followed as a function of the pump fluence, the time elapsed, and the wavelength, polarization and angle of incidence of the probe. This parameterization gives detailed information on the changes in electronic structure of the

Snap judgements

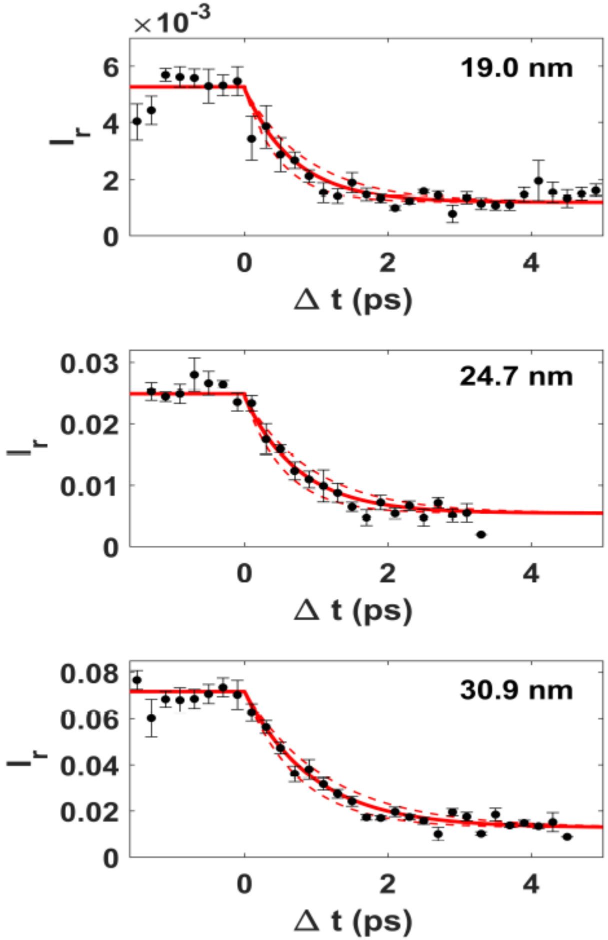
THIS remarkable photograph was taken by a remarkable man, Eadweard Muybridge, a pioneer of sequence photography. Muybridge — born Edward Muggeridge (he changed his name), died Eadweard Maybridge (a misspelling on his tombstone) — was English by birth but in 1851, at the age of 21, moved to the United States where he gained a high reputation as a photographer. Among his commissions was one from Leland Stanford, a former governor of California, to photograph a trotting horse and settle the controversy as to whether all four hooves are lifted off the ground at the same time. The answer, revealed by images such as that reproduced here, was that they are. The work of Muybridge and others is celebrated in a exhibition which opened this week at the Museum of the Moving Image in London. Entitled "Catching the Action: Muybridge and the Chronophotographers", it runs until 31 May. **T.L.**

110

E. Principi et al., in preparation



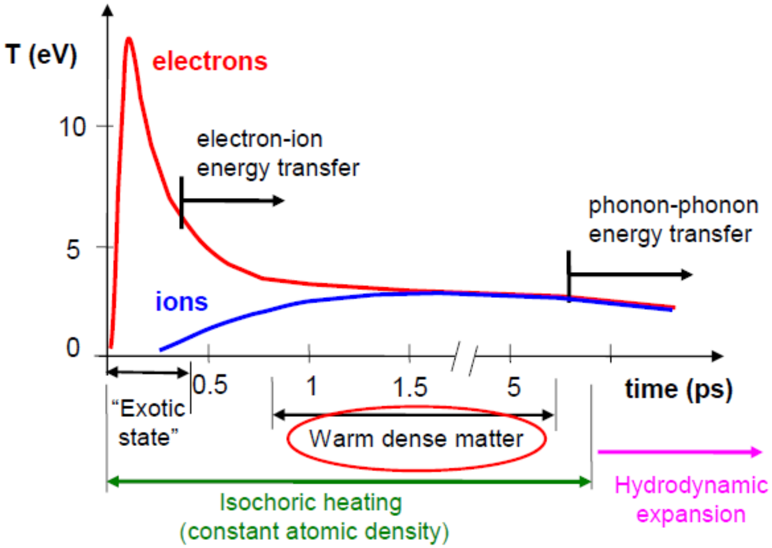
D. H. Reitze et al., Phys. Rev. B (1992)

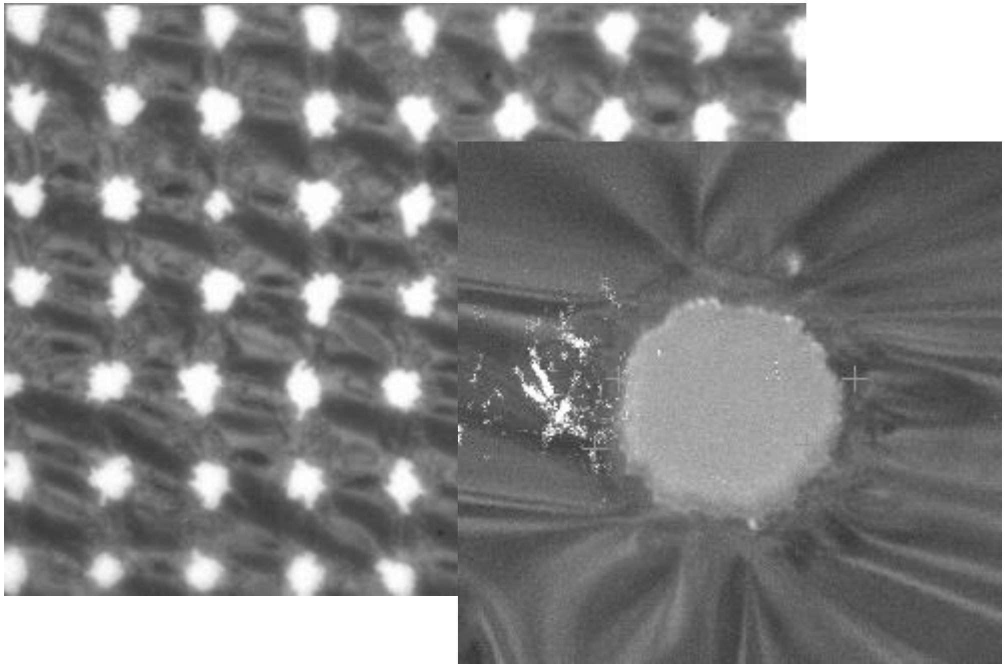


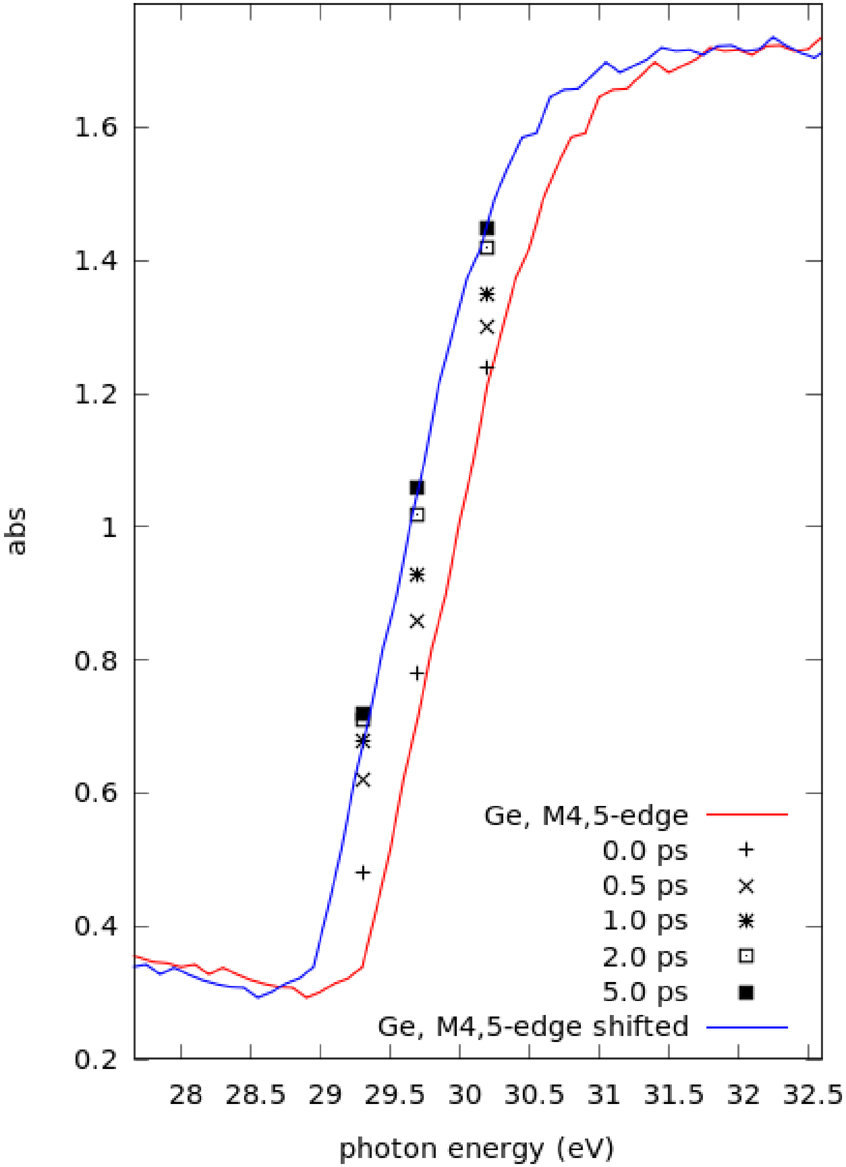
R. Mincigrucci et al., Photonics (2017)

XANES on Germanium

Elettra Sincrotrone Trieste

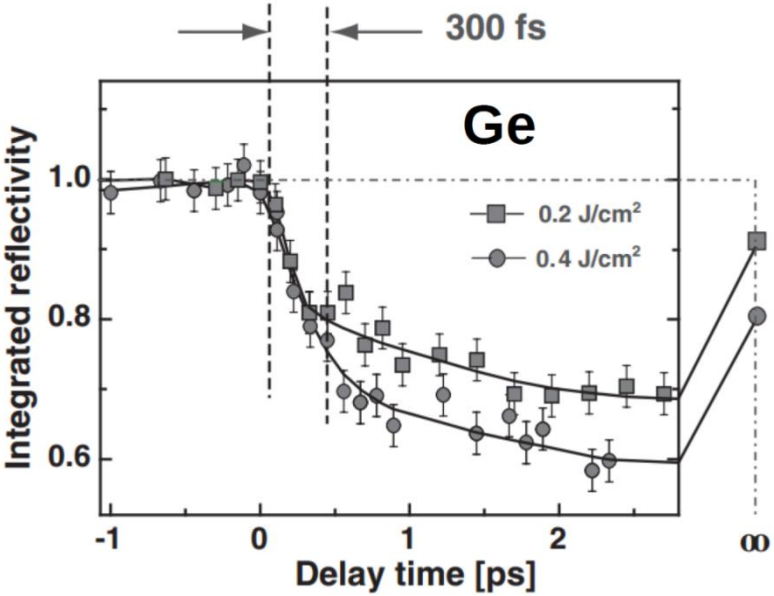






HDL – LDL transition in Germanium

Elettra Sincrotrone Trieste



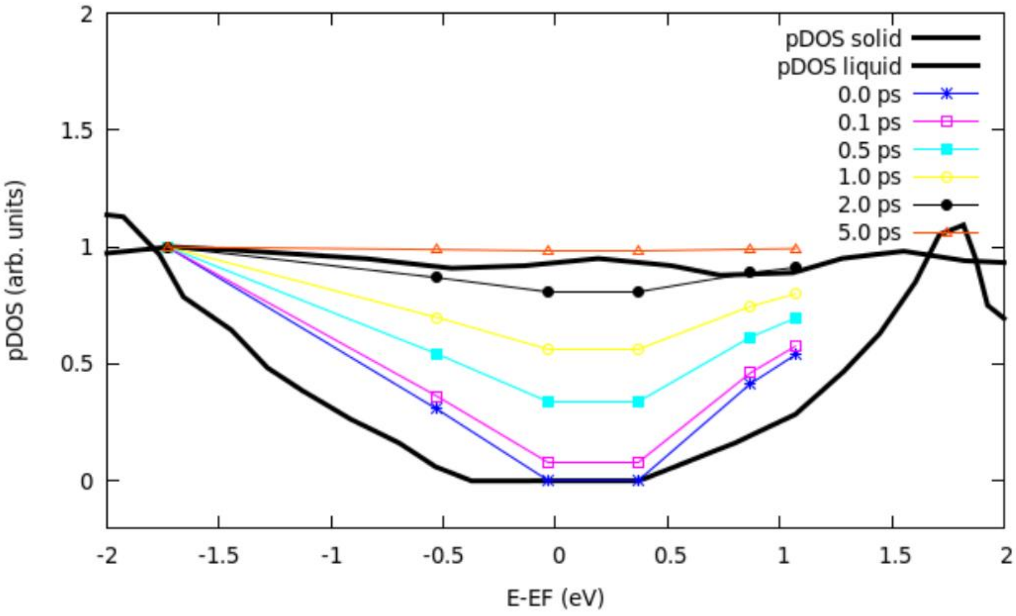
K. Sokolowski-Tinten et al., J of Phys: Cond. Matt. (2004)

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

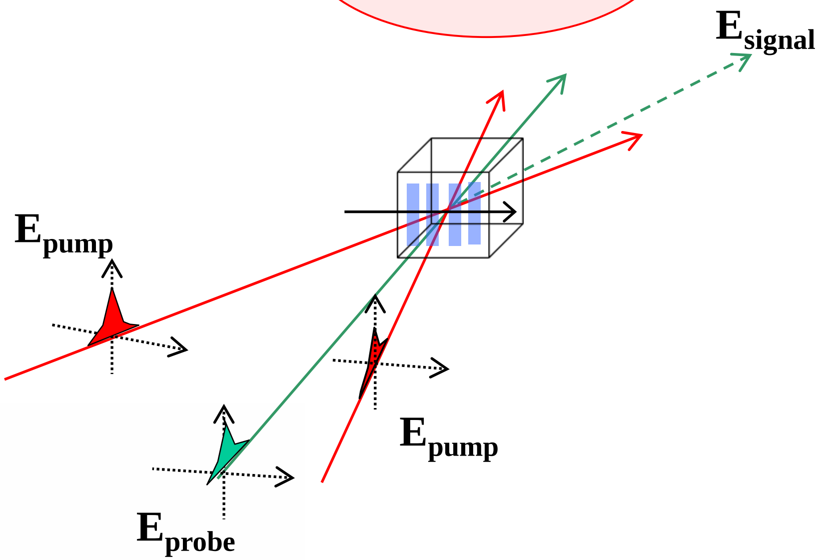


E. Principi et al., PRB (2018)

Third Order Processes

N. Bloembergen 1981

$$P_i = \chi^{(1)}_{ij}E_j + \chi^{(2)}_{ijk}E_jE_k + \chi^{(3)}_{ijkl}E_jE_kE_l + \dots$$



$$\omega_1 + \omega_2 + \omega_3$$

$$3\omega_j\,,\ \omega_j \qquad j=1,2,3$$

$$2\omega_i + \omega_j\,,\ 2\omega_i - \omega_j\,,\ \omega_i - 2\omega_j$$

$$\boxed{\omega_i + \omega_j - \omega_k}\,,\ \omega_i - \omega_j - \omega_k$$

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R. Cucini

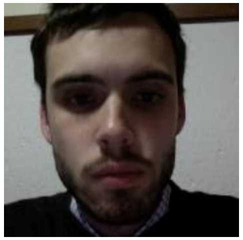

E. Pedersoli


L. Foglia


A. Gessini


F. Bencivenga


E. Principi


R. Mincigrucci