Soft x-ray absorption and pump-probe experiments of transient states using FEL sources

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XFEL: Ultra-short x-ray pulses

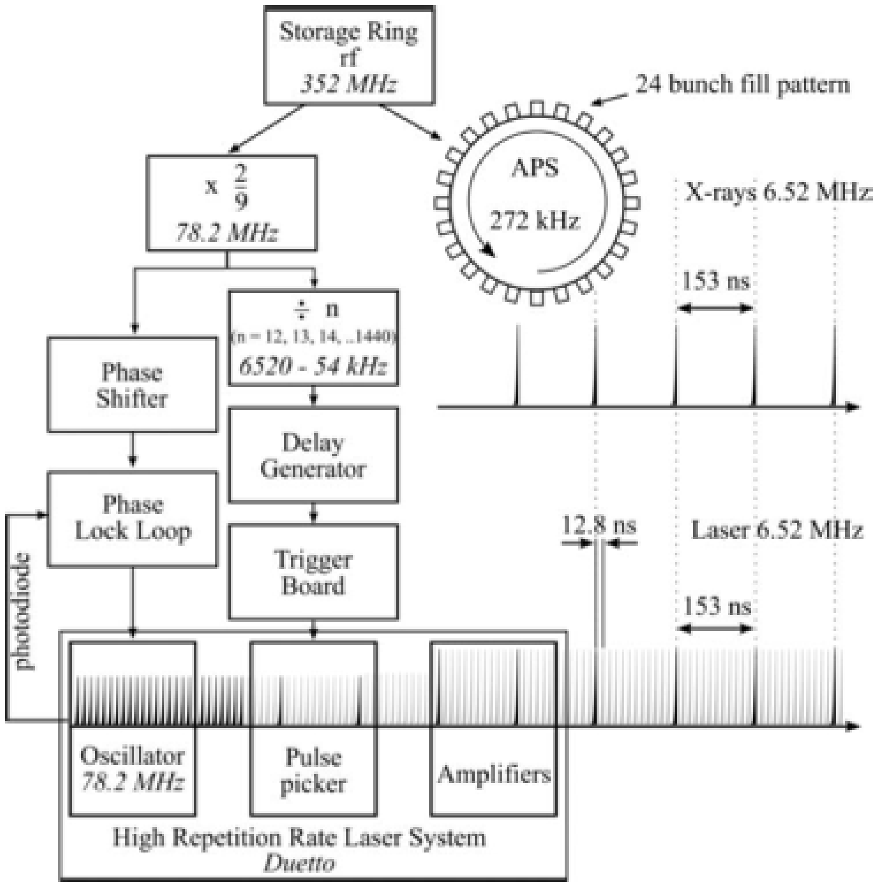


FIG. 1. Schematic of the synchronization and delay control of the laser pulses with respect to the x-ray pulses. The laser is shown operating at 6.52 MHz (78.2 MHz ÷ 12) to match the x-rays in the standard 24 bunch mode. The light gray pulses separated by 12.8 ns illustrate other oscillator pulses that could be chosen for coarse delay of the laser relative to the x-ray pulses.

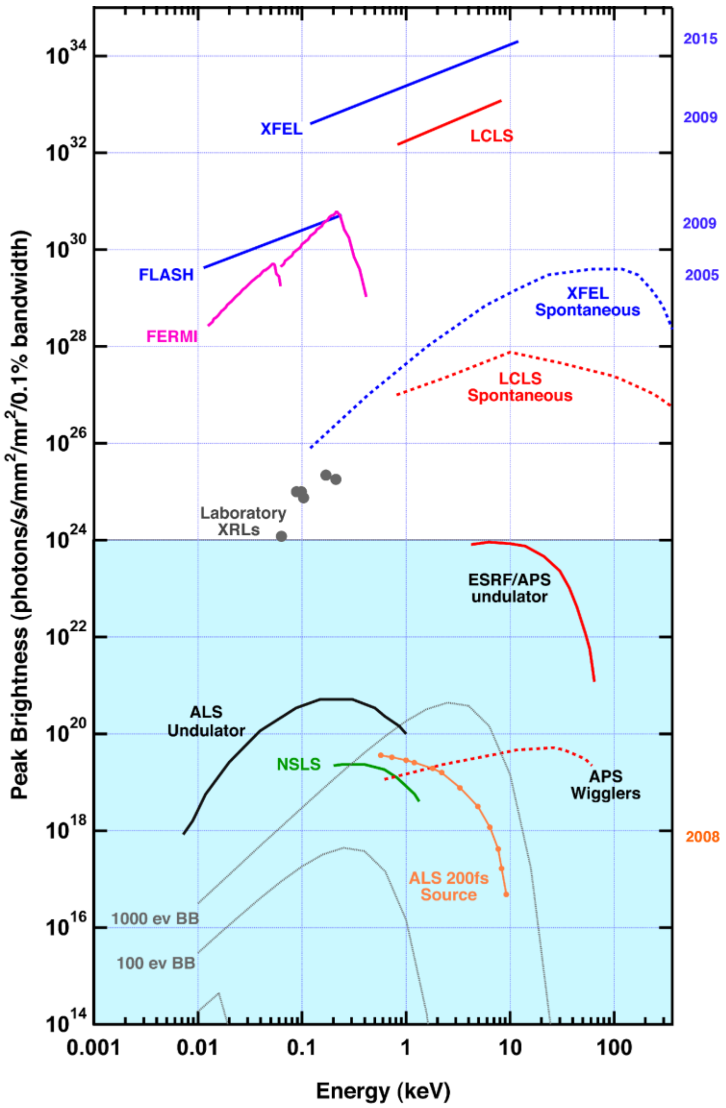
Free Electron Laser

- Usually 10-50 Hz repetition rate (Eupraxia 10-600 Hz)
- XFEL 27 kHz
- Ex. SACLA below 10fs with 10^{18}W/cm2

Faster than electron-phonon interaction, comparable to the lifetime of core-hole

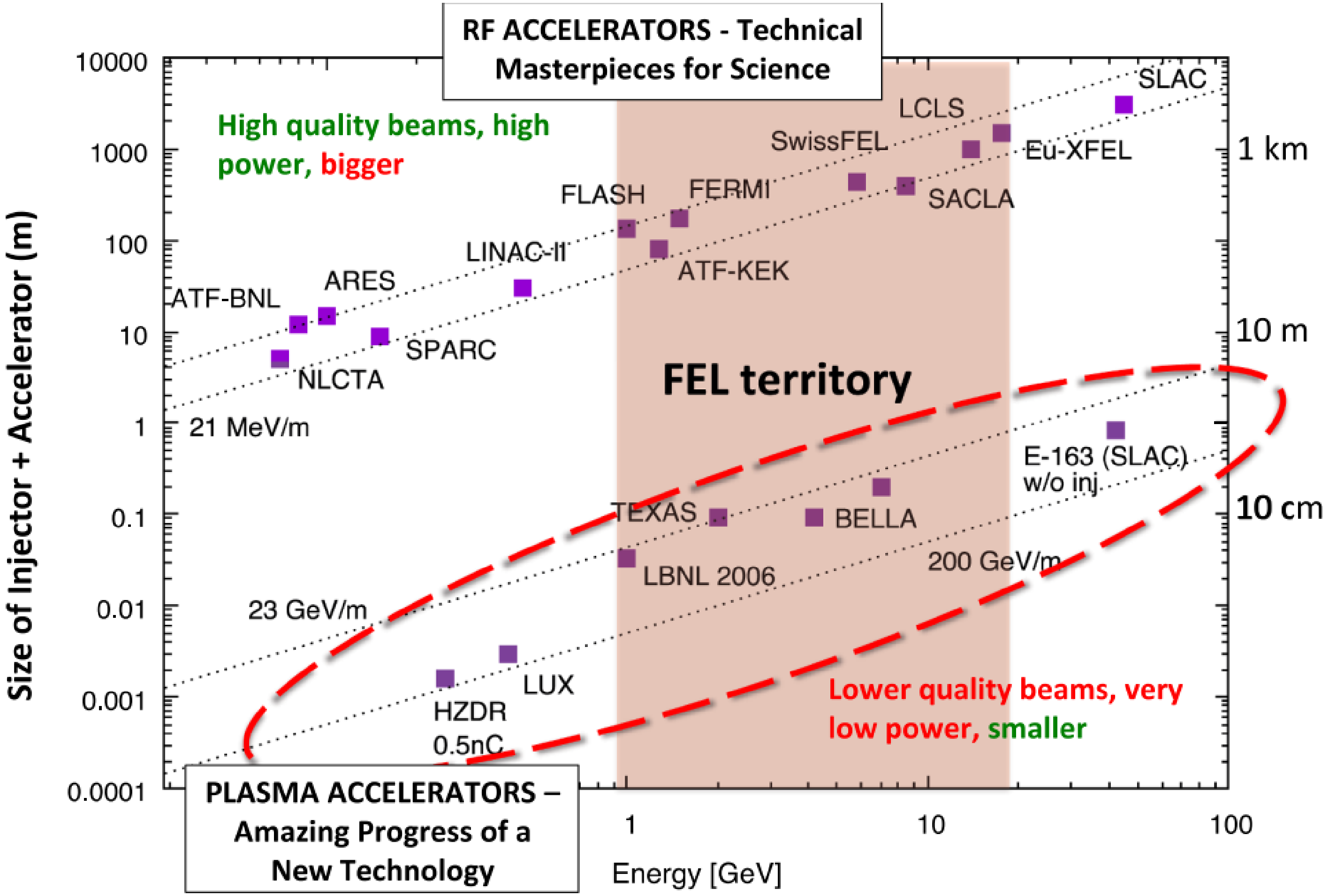
Nonlinear optical effects on ultra short time resolved spectroscopies

Typical peak brightness



Parameter	FEL - 1	FEL - 2	Units	Eupraxia
	Fermi			
Wavelength	100 - 20	20 - 3	nm	100-2 nm
Photon Energy	12 - 62	62 - 413	eV	12-620 eV
Pulse Length	30 - 100	< 100	fs	5–50 fs rms
Bandwidth	~ 20 - 40	~ 20 - 40	meV	0.15–0.23%
Polarization	variable	variable	-	
Repetition Rate	10 - 50	10 - 50	Hz	10-600 Hz
Peak Power	1 ÷ 5	~ 1	GW	
Photons per Pulse	$2 \cdot 10^{14}$ @ 100 nm	$1 \cdot 10^{13}$ @ 10 nm	-	$0.4–2.6 \cdot 10^{12}$
Power Fluctuation	~ 25%	> 50%	-	
Central Wavelength Fluctuation	within bandwidth	within bandwidth	-	
Output Transverse Position Fluctuation	50	50	µm	
Pointing Fluctuation	< 5	< 5	µrad	
Output Spot Size (intensity, FWHM@waist)	290	140	µm	
Divergence (intensity, RMS)	50 @ 40 nm	15 @ 10 nm	µrad	27–51 µrad

Eupraxia:



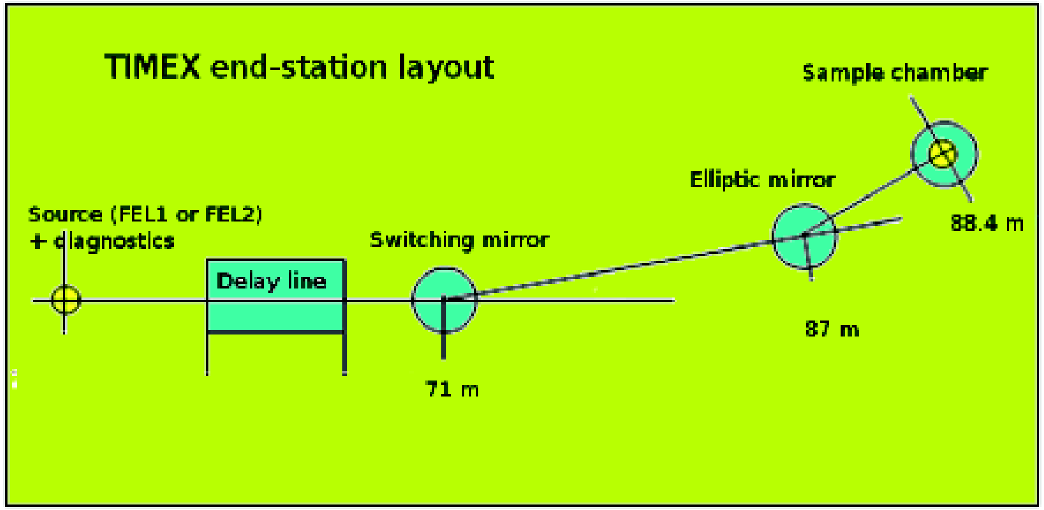
Comparison of size and achievable electron beam energy for different existing radiofrequency and plasma accelerators. It becomes clear that a trade-off between machine size and beam quality at equivalent beam energies distinguished the two technologies to date.

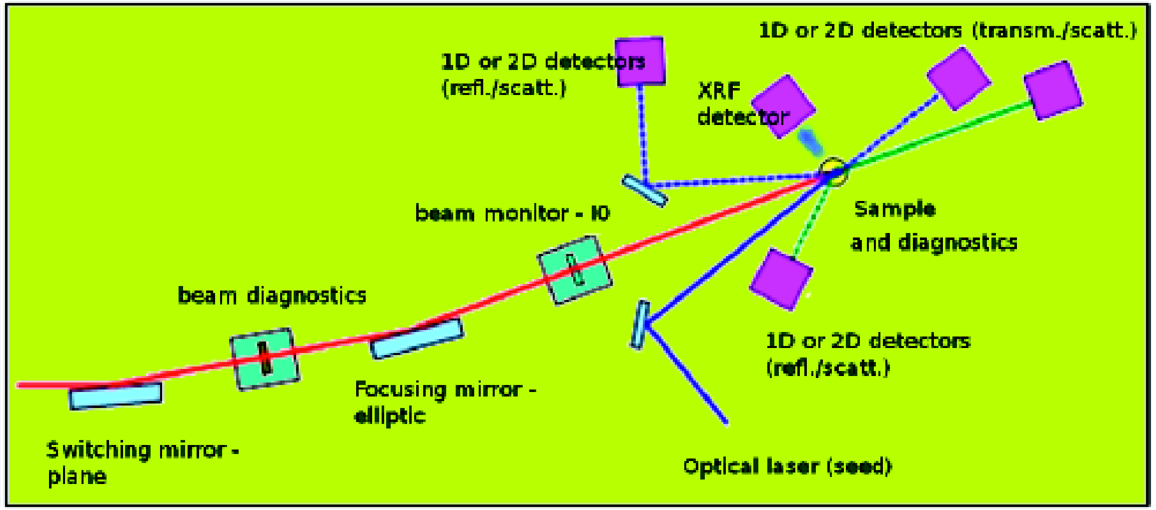
Previous experiences:

TIMEX end-station: TIme-resolved studies of Matter under EXtreme and metastable conditions

Design of the end-station and place of orders completed in June 2010. Vacuum chambers delivered in late October 2010. Pilot experiments (2007- 2010) completed.

Assembling and test of the main components of the TIMEX end-station in the main Fermi hall started in December 2010.





Probing phase transitions under extreme conditions by ultrafast techniques: Advances at the Fermi@Elettra free-electron-laser facility

Journal of Non-Crystalline Solids 357 (2011) 2641–2647

Andrea Di Cicco [a,b,c,*], Francesco D'Amico [b], Goran Zgrablic [b], Emiliano Principi [a], Roberto Gunnella [a], Filippo Bencivenga [b], Cristian Svetina [b], Claudio Masciovecchio [b], Fulvio Parmigiani [b], Adriano Filipponi [d]

Studies on non thermal electron heating and Warm Dense Matter (WDM) dynamics

Warm Dense Matter (WDM) occurs in:

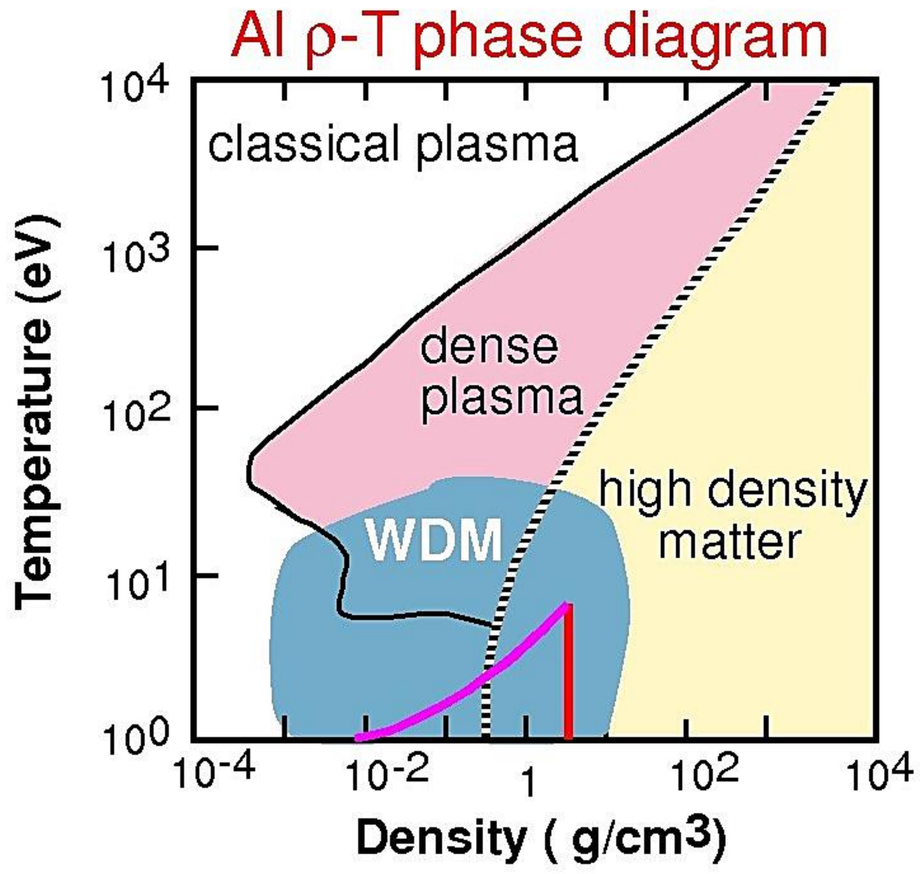
- Cores of large planets
- Systems that start solid and end as a plasma
- X-ray driven inertial fusion implosion

WDM is the regime where neither condensed matter (T = 0) methods nor plasma theoretical methods are valid

Measures the fundamental nature of the matter via equation of state

FEL radiation can create and/or probe WDM in an extended P-T range

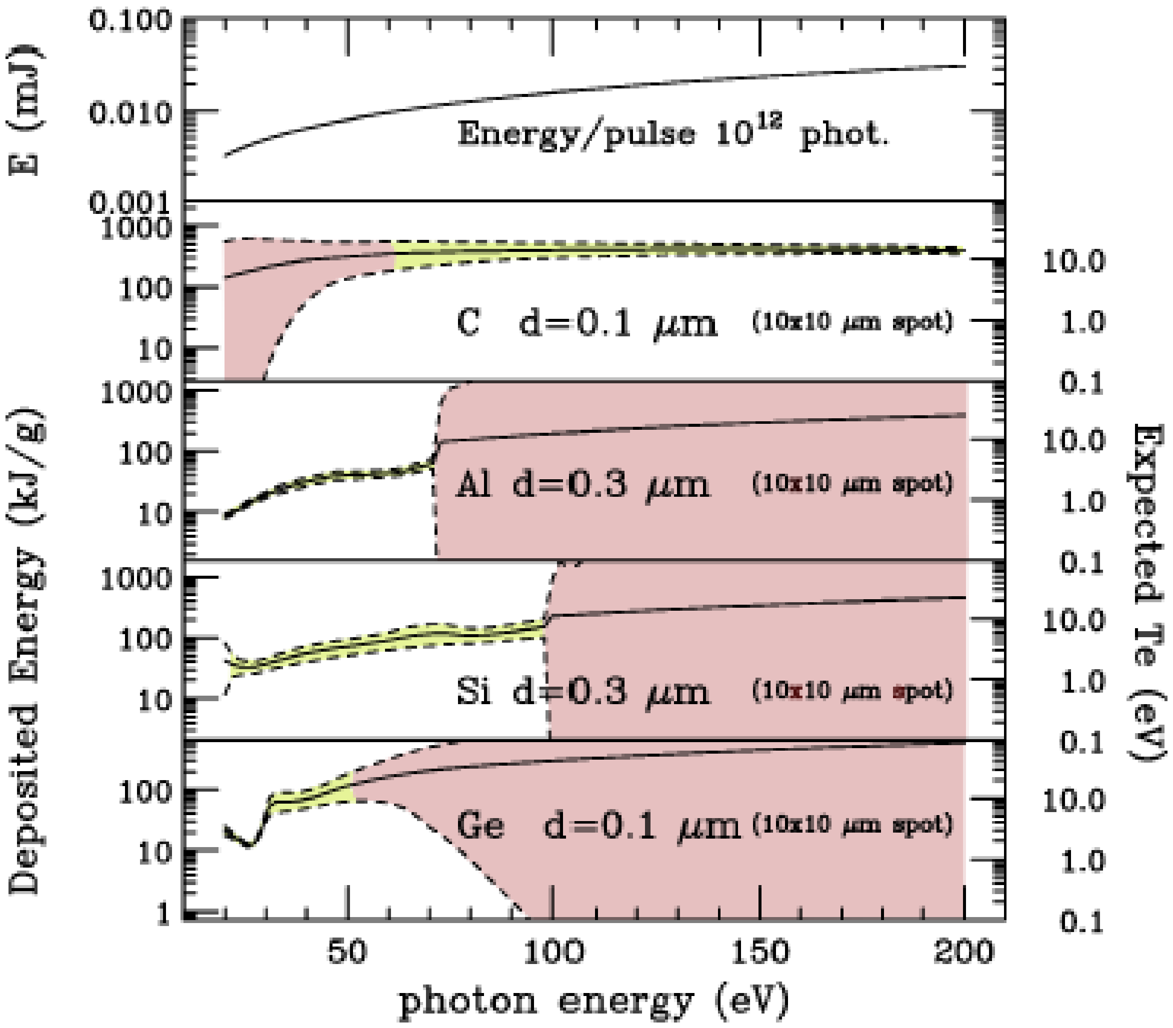
- Lack of knowledge in the Equation of State (EOS) even of simple systems near WDM regime (data largely based on Single-shot shockwave techniques)
- During a typical WDM creation, systems usually start solid and end as a plasma.
- FEL radiation can heat bulk matter rapidly and uniformly to create isochores (constant density) and iso-entropies on release (constant entropy)



B. Nagler et al., Nat. Phys. 5, 693 (2009)

Bulk heating 20-200 eV photons

- Quasi-isotherm bulk heating can be obtained by using FEL pulses on several films (Al, Si, Ge, and more)
- Electron temperatures are estimated to be in the range 110 eV (WDM regime)
- Large sized self-standing films are robust (0.1-0.3 microns thickness) and can be used for shot to shot Experiments at the FEL repetition rate
- Useful for bulk heating of some important material (C for example)
- Soft X-ray FELs: a path to efficient bulk heating!

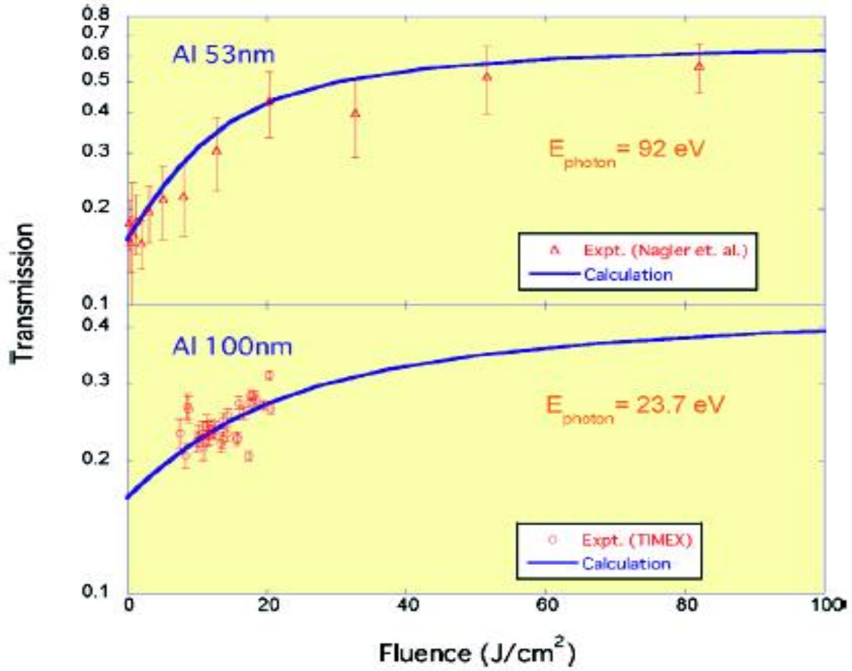


nature physics
ARTICLES
PUBLISHED ONLINE: 26 JULY 2009 | DOI: 10.1038/NPHYS1341

Turning solid aluminium transparent by intense soft X-ray photoionization

Absorption saturation effect

Bob Nagler *et al.**



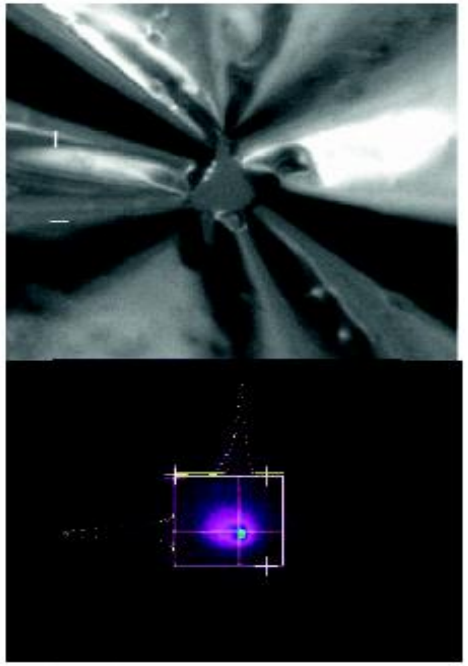
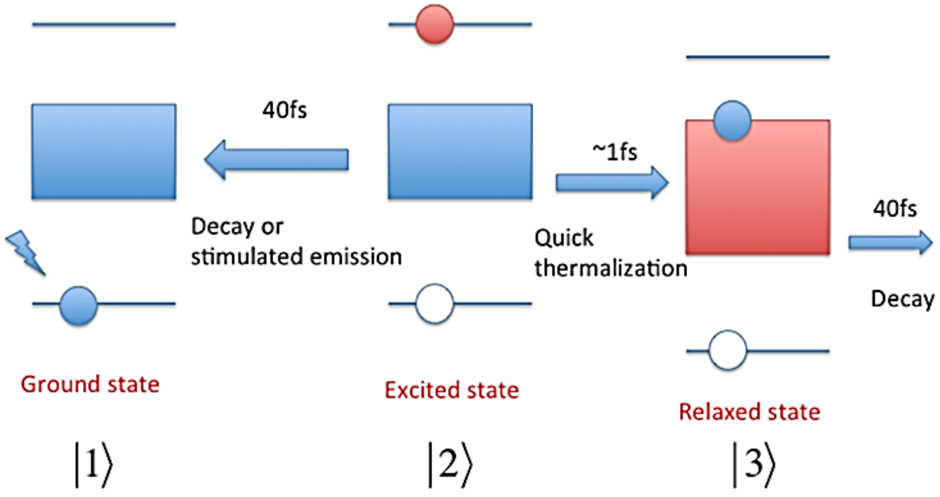


Figure 4

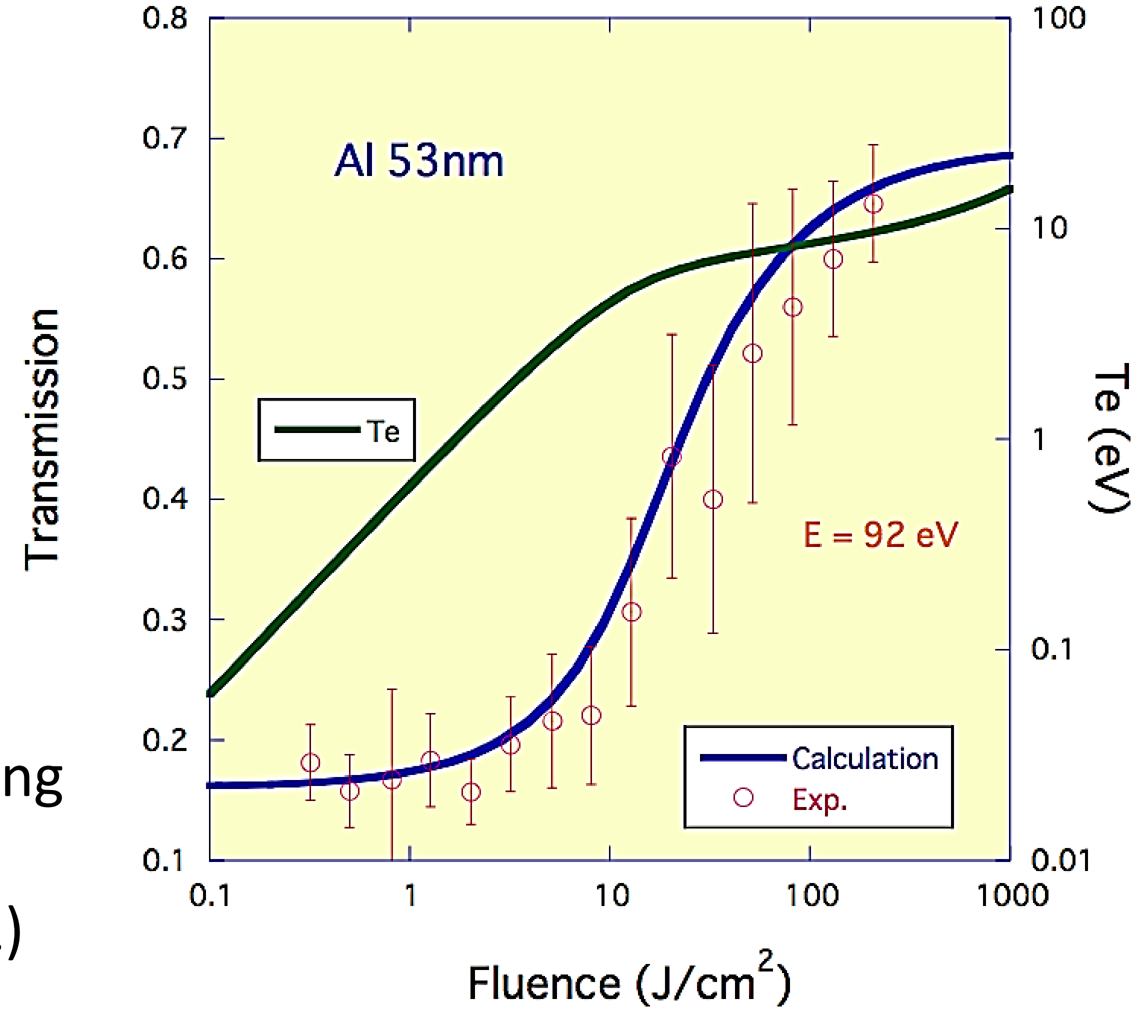
Left side: transmission of Al ultrathin foils as a function of the incident fluence of FEL pulses. The result of transmission measurements at the FLASH facility (see ref.[7]) and the first results obtained at TIMEX are compared with calculations (see text). Right side: the lower panel shows the lateral dimensions of the FEL pulses (10x10 μ FWHM) at focus (as observed on a YAG screen by the TIMEX telemicroscope), the upper panel shows the effect of about 100 repeated FEL shots (fluence 10-20 J/cm^2) on a 100 nm ultrathin Al foil. The pulses of seed laser were not filtered and concur to the damage of the foil.

Modeling Non-Equilibrium Dynamics and Saturable Absorption Induced by Free Electron Laser Radiation

Keisuke Hatada *,† and Andrea Di Cicco



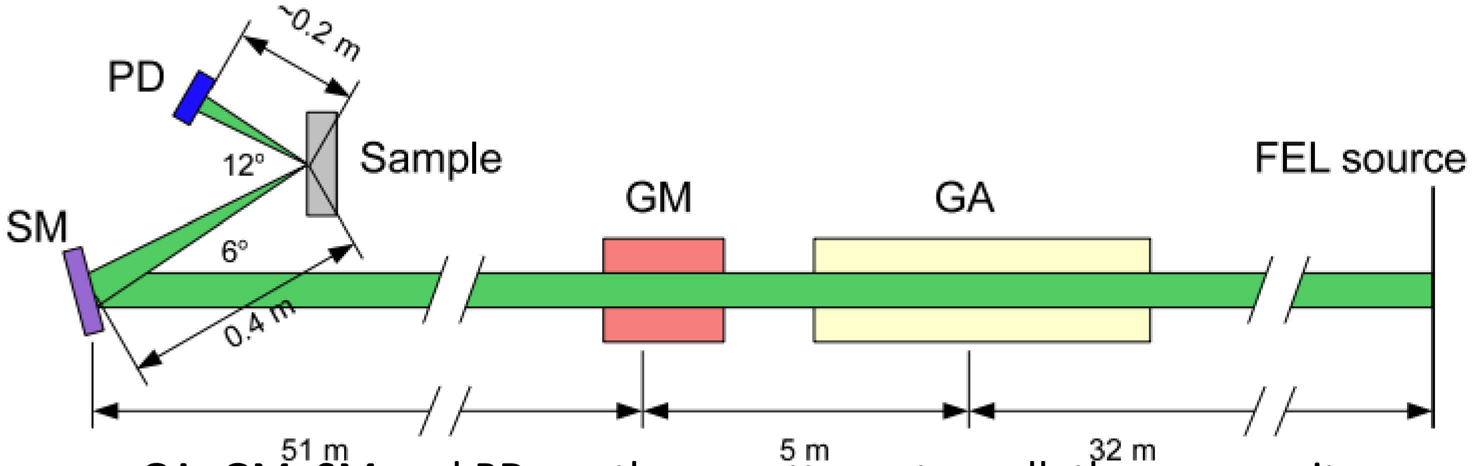
0.85~1.7 fs Electron thermalization time



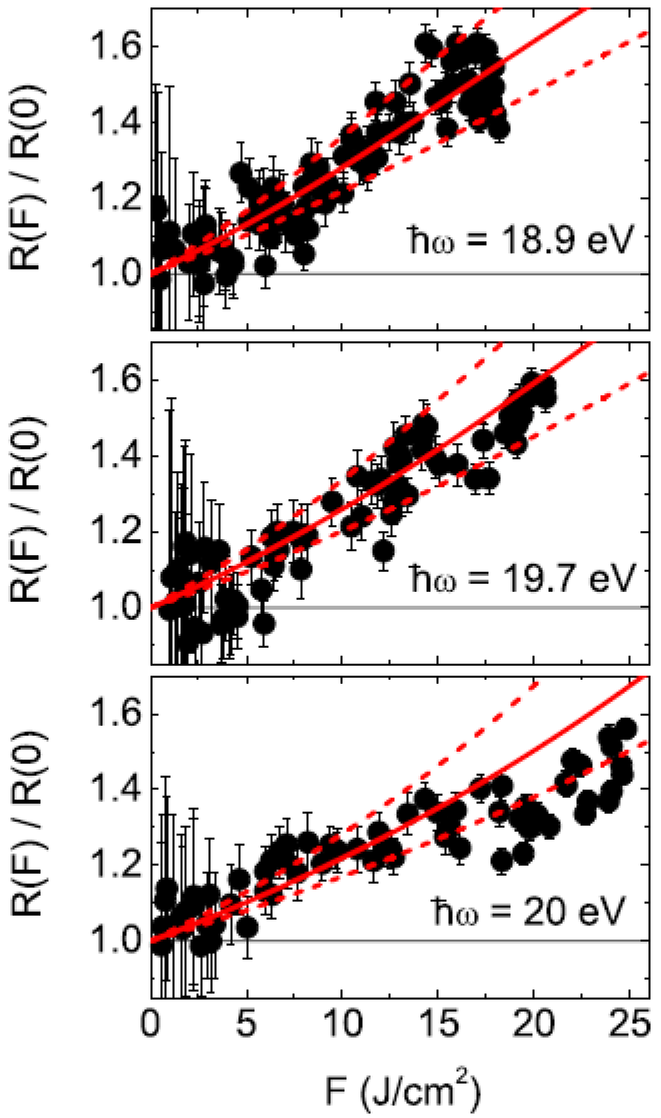
- In the low fluence limit the attenuation is decreasing exponentially with the thickness (Lambert-Beer)
- Above the saturation threshold (about 1-10 J cm-2) linearly constant.

Appl. Sci. 2017, *7*, 814; doi:10.3390/app7080814 Journal of Electron Spectroscopy and Related Phenomena 196 (2014) 177–180

Reflectivity increase by Flux



GA, GM, SM and PD are the gas attenuator cell, the gas monitor, the spherical focusing mirror and the photodiode, respectively



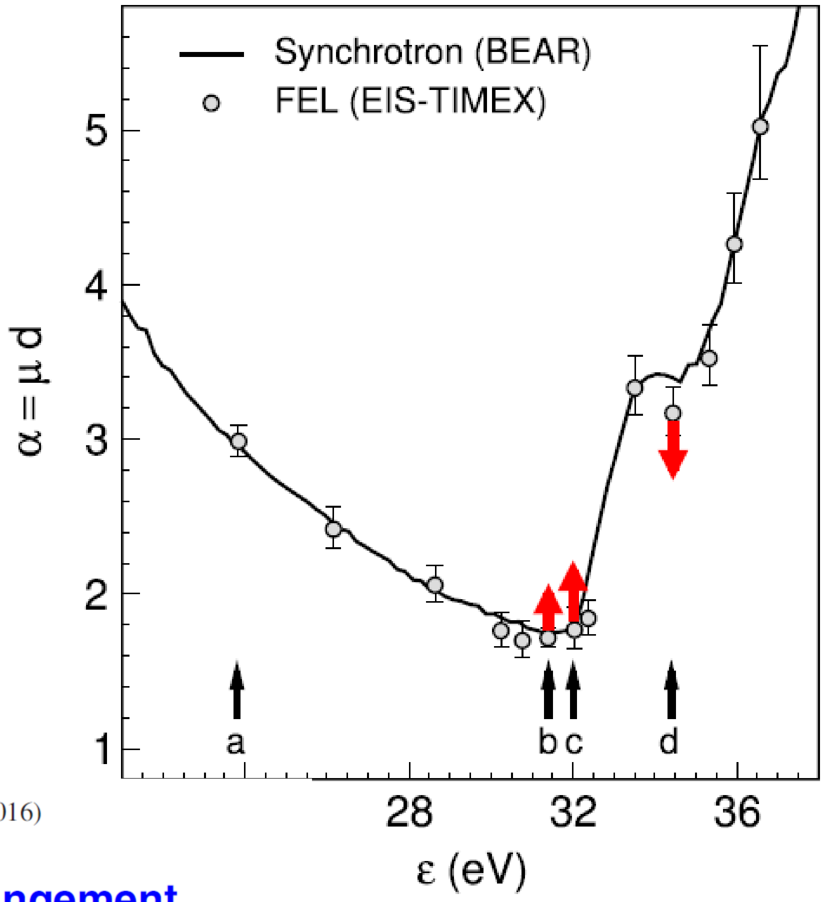
New phenomenon observed in the range 18.9-20 eV, interpreted as a shift of the plasmon energy (Drude model, ~17 eV in Ti) as a consequence of the injection of free electrons (increase of the electron density) within the pulse duration.

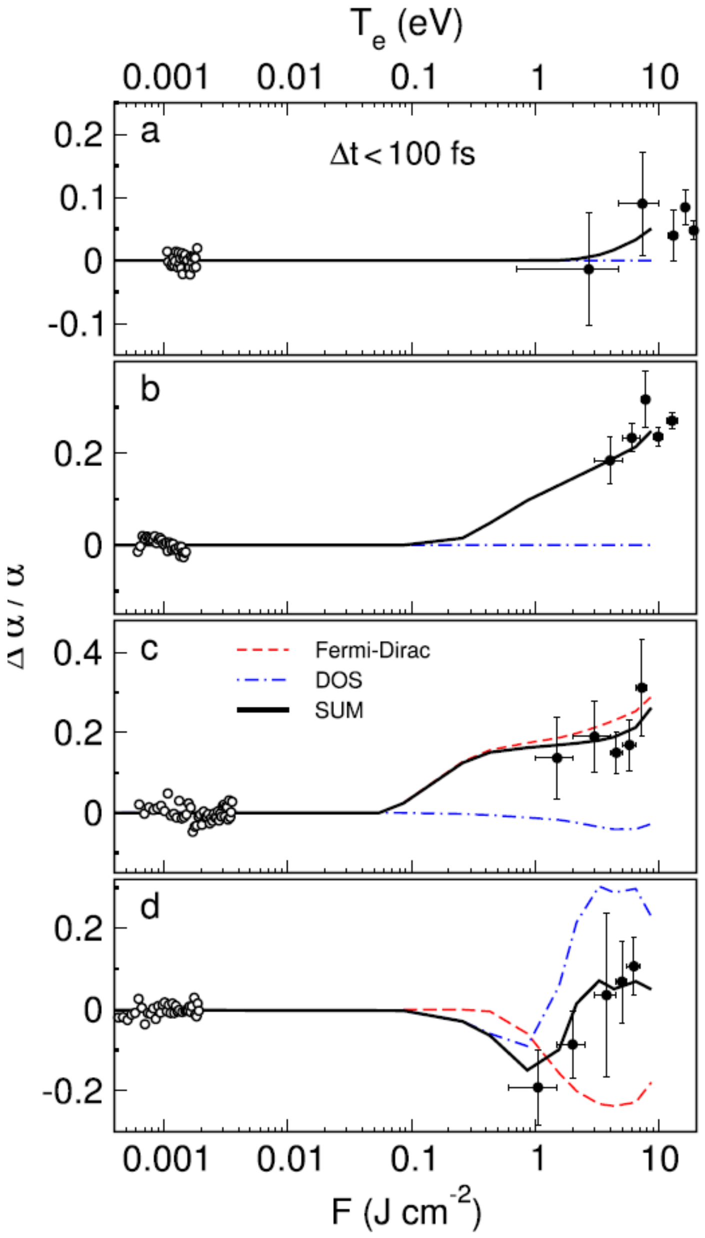
FEL will allow one to probe the condition where the metal reflectivity is almost insensitive to plasma frequency variations but strongly depends on the plasmon lifetime.

SCIENTIFIC REPORTS | 4 : 4952 | DOI: 10.1038/srep04952

Absorption modification:

- Ti 3p near-edge ultrafast absorption data were collected for fluences up to about 10 Jcm-2 corresponding to electron temperatures in the 1-10 eV range.
- Clear changes in the shape at high fluence are assigned to ultrafast electron heating within 100 fs (no lattice heating)





STRUCTURAL DYNAMICS 3, 023604 (2016)

Free electron laser-driven ultrafast rearrangement of the electronic structure in Ti

E. Principi,[1,a)] E. Giangrisostomi,[1] R. Cucini,[1] F. Bencivenga,[1] A. Battistoni,[1] A. Gessini,[1] R. Mincigrucci,[1] M. Saito,[1] S. Di Fonzo,[1] F. D'Amico,[1] A. Di Cicco,[2] R. Gunnella,[2] A. Filipponi,[3] A. Giglia,[4] S. Nannarone,[4,5] and C. Masciovecchio[1,b)]

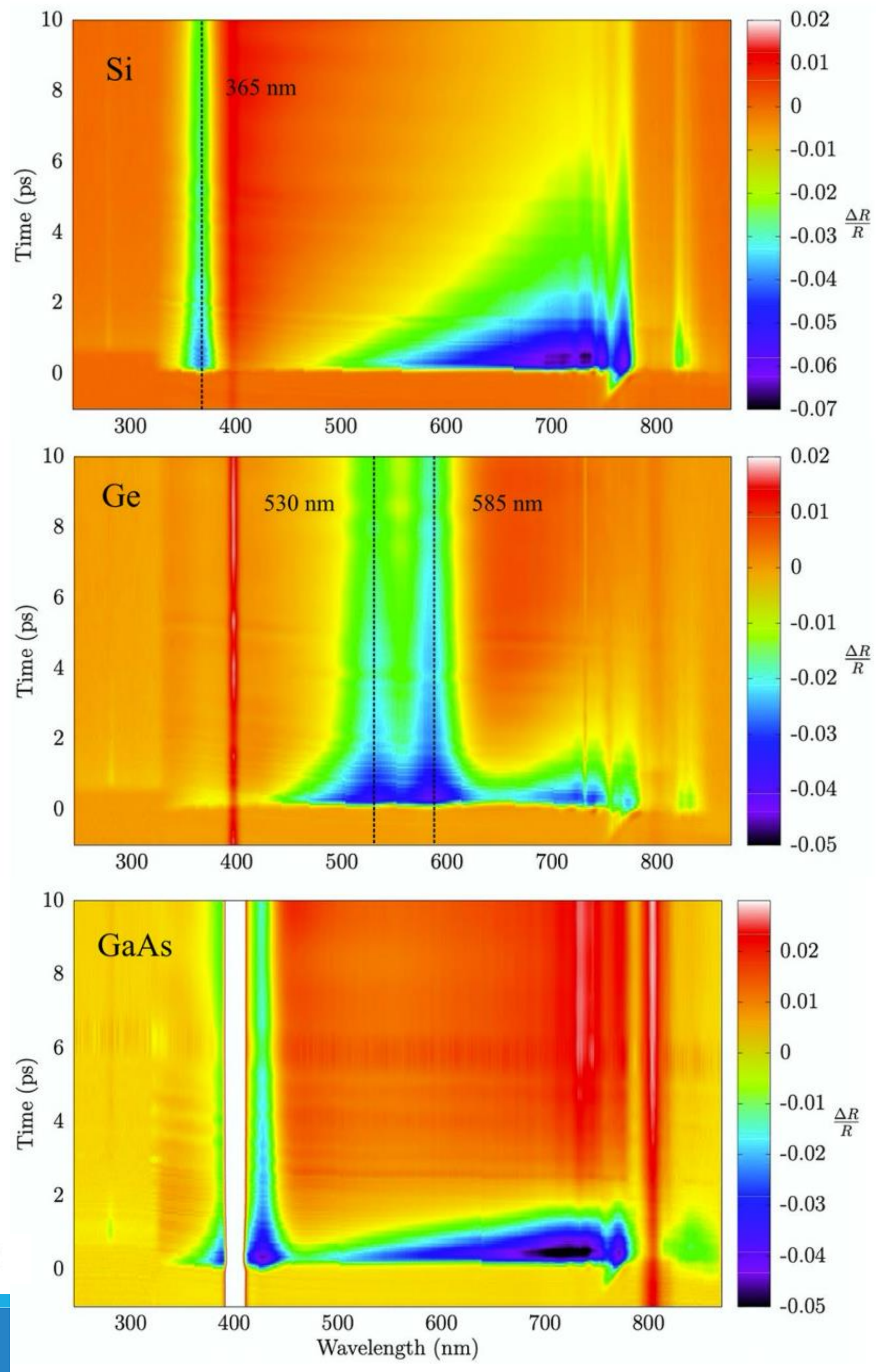
Pump and Probe: Pilot ultrafast experiments with a laser source

Pump-and-probe reflectivity measurements on Si, Ge and GaAs surface.

Laser source:
Pump: at 400 nm, pulse width FWHM 80 fs, 500 Hz. Typical pump spot size 100 µm (pump) and 50 µm (probe).
Probe: Super-continuum 350-800 nm probe, probing depth around 10 nm.

Ultrafast pump-and-probe pilot reflectivity experiments on Si show that useful information about the dynamics of phase transitions can be obtained. Non-thermal melting of Si takes place within 300 fs and is followed by lattice heating or melting within 3 ps, as a function of the pump fluence.

Diverse time frame in case of different DOS configuration of the investigated sample.



scientific reports

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Broadband optical ultrafast reflectivity of Si, Ge and GaAs

A. Di Cicco[1,2], G. Polzoni[1], R. Gunnella[1], A. Trapananti[1,2], M. Minicucci[1], S. J. Rezvani[1], D. Catone[3], L. Di Mario[3], J. S. Pelli Cresi[3], S. Turchini[3] & F. Martelli[4]

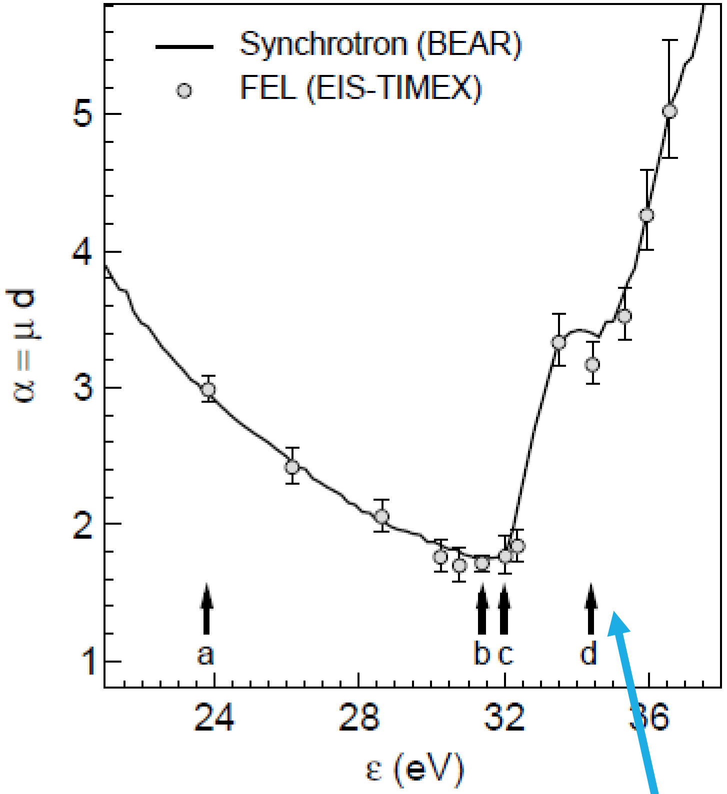
Scientific Reports | (2020) 10:17363 |

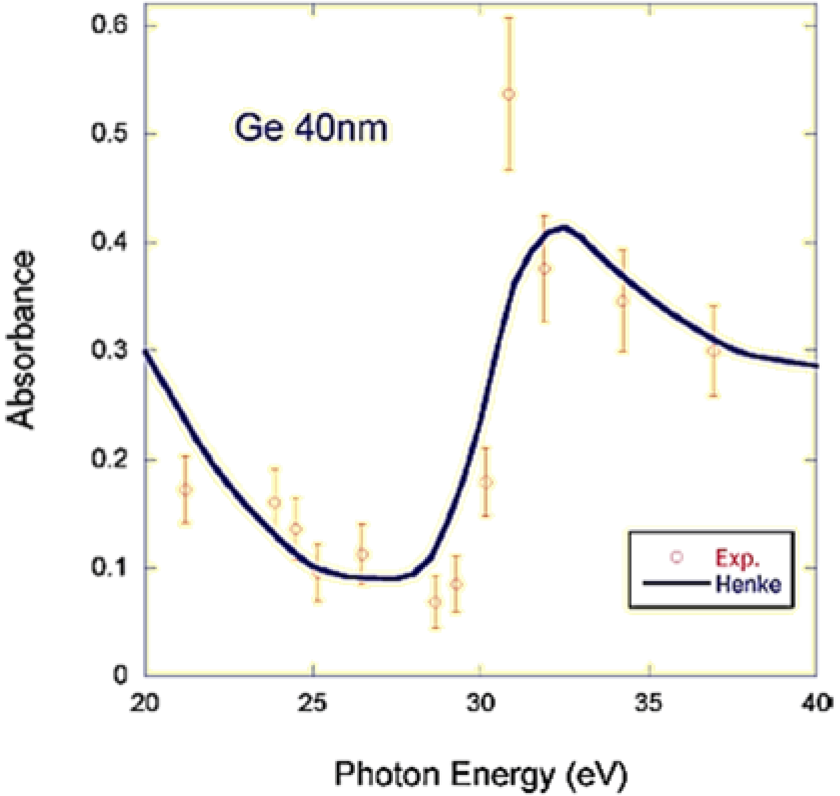
nature research

X-ray Absorption Spectroscopy at X-ray Free Electron Lasers

X-ray Absorption Spectroscopy (XAS) is an extremely powerful x-ray diagnostic for simultaneously probing the electronic structure and the local atomic arrangement.

Possible Tunability across the edges





New Journal of Physics **14** (2012) 113009 (19pp)
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STRUCTURAL DYNAMICS **3**, 023604 (2016)

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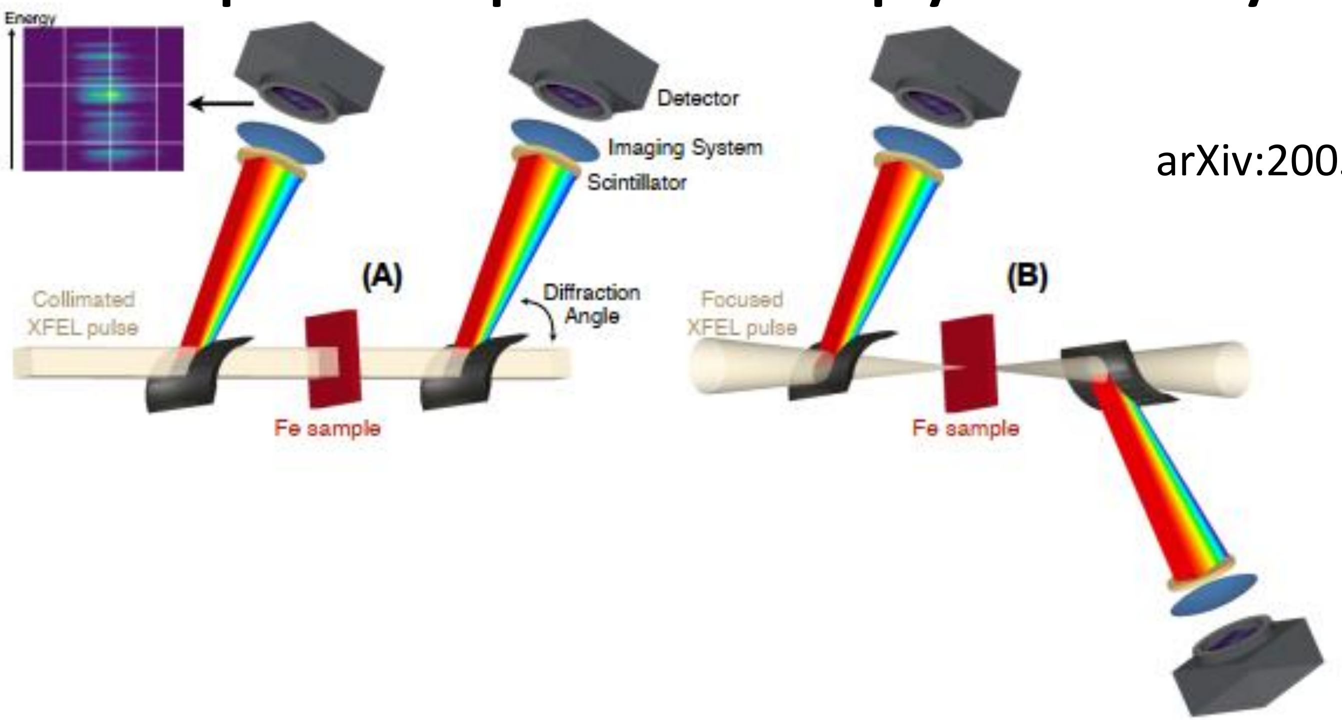
High-quality Ti $M_{2,3}$-edge (3p) absorption spectra with FEL ultrashort pulses: near-edge changes at high fluence (high-energy density).

Single-shot X-ray Absorption Spectroscopy at X-ray Free Electron Lasers



arXiv:2005.01572v1

Energy dispersive experimental setup at LCLS-SLAC with the unfocused collimated (A) and the focused (B) XFEL beam. Two identical spectrometers allow simultaneous measurement of the incident and transmitted spectra.

- Energy resolution better than the ~1%
- FEL bandwidth (grating mono)
- Normalization on a shot-to-shot basis (spikes and FEL intensity fluctuations).

Normalized single-shot X-ray absorption spectroscopy at a free-electron laser

GÜNTER BRENNER,[1,*] SIARHEI DZIARZHYTSKI,[1] PITER S. MIEDEMA,[1] BENEDIKT RÖSNER,[2] CHRISTIAN DAVID,[2] AND MARTIN BEYE[1]
Vol. 44, No. 9 / 1 May 2019 / *Optics Letters* 2157

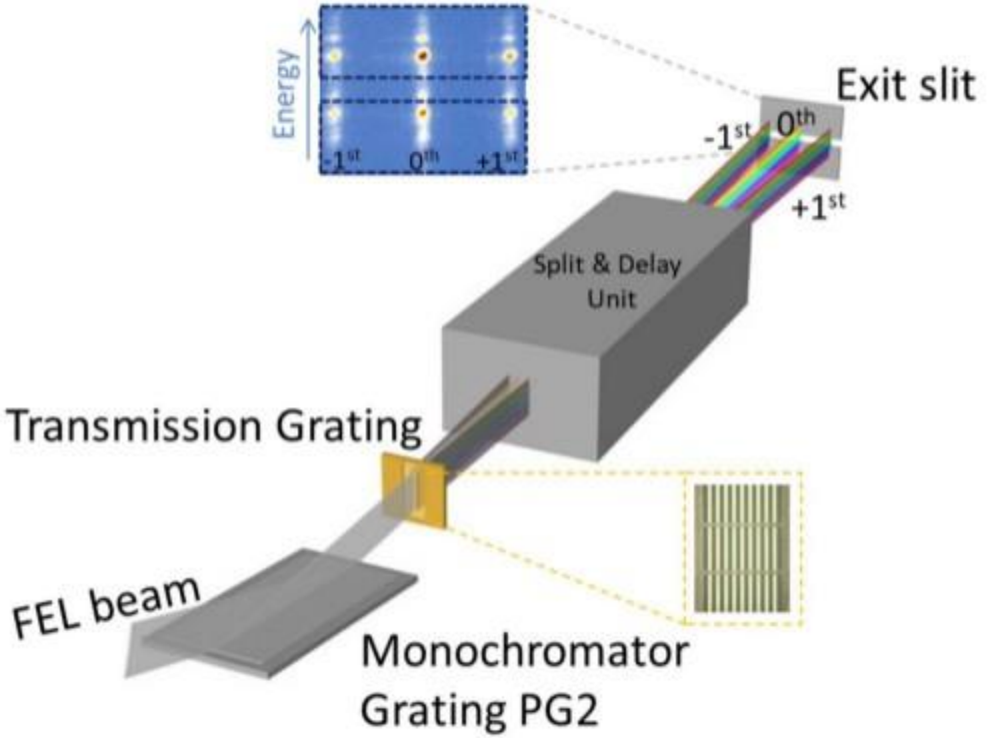
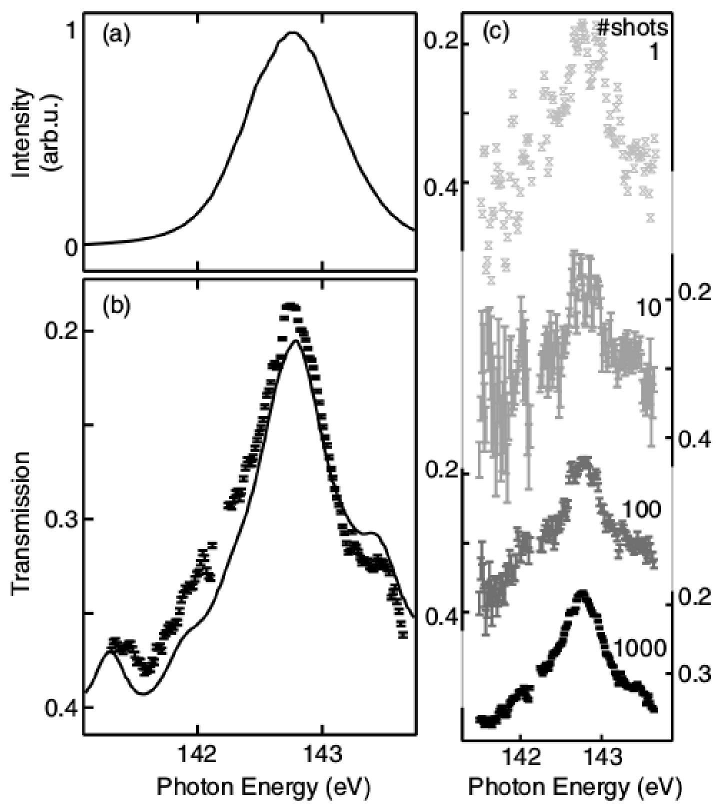
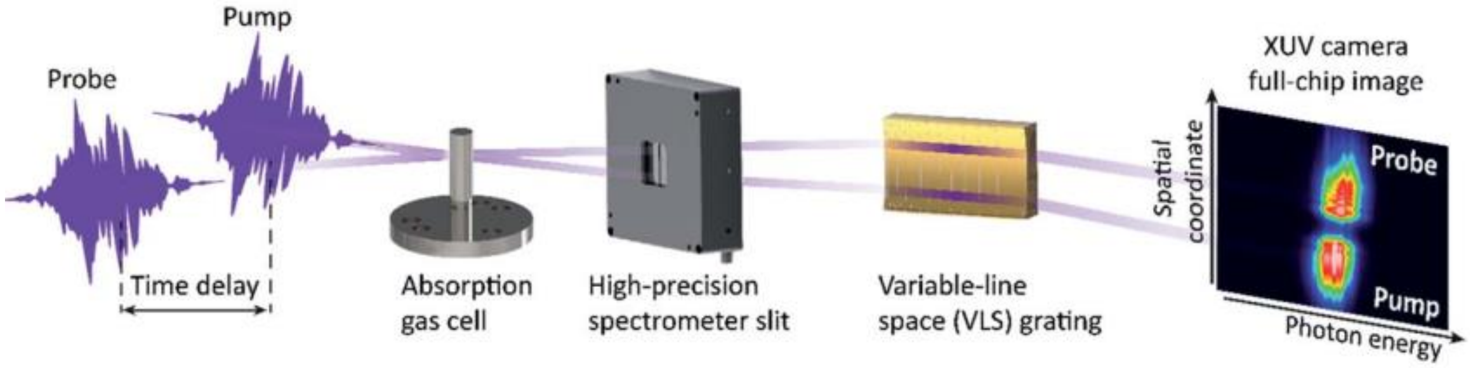


Fig. 1. Schematic of the setup for XAS at FLASH using a TG to split the FEL beam into a signal and reference beam (+1st and −1st order, respectively).

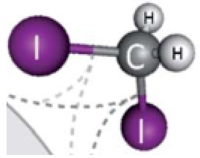
Gd2O3 sample: Gd N edge



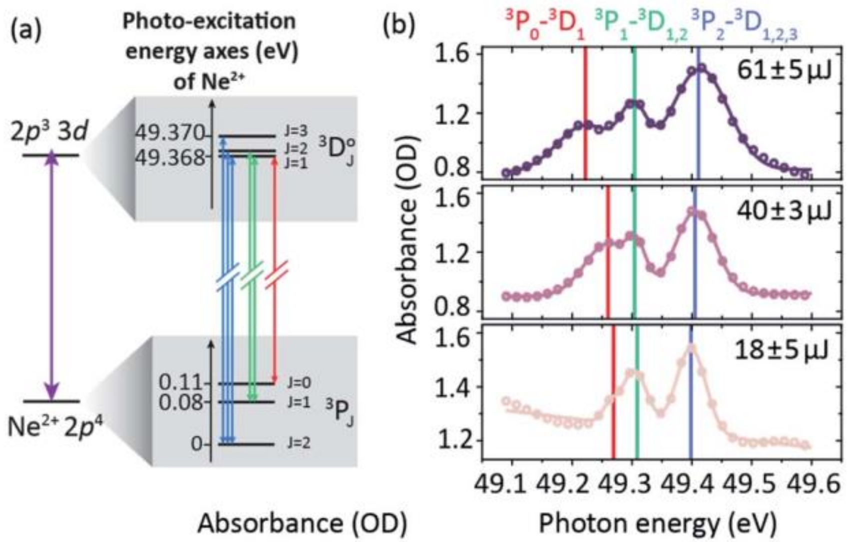
a) Average spectral distribution of FLASH. A total energy bandwidth of 0.9 eV FWHM was measured. (b) Gd N4,5-edge x-ray absorption spectrum measured in transmission of a Gd2O3 thin film. The symbols represent data recorded at FLASH, while the solid line displays a reference measurement of the sample from a synchrotron. (c) Single-shot and averaged spectra for 10, 100, and 1000 FEL pulses



Measurement principle of split-beam XUV-pump XUV-probe transient-absorption spectroscopy. After focusing the pulses into the absorption gas cell filled with a moderately dense gas medium the transmitted pulses are coupled into a grating spectrometer consisting of a high precision slit, a VLS grating, and an XUV-sensitive camera which simultaneously detects the spatially separated pump and probe pulse spectra on a single shot basis



diiodomethane CH_2I_2



Faraday Discussions

Cite this: *Faraday Discuss.*, 2021, **228**, 519

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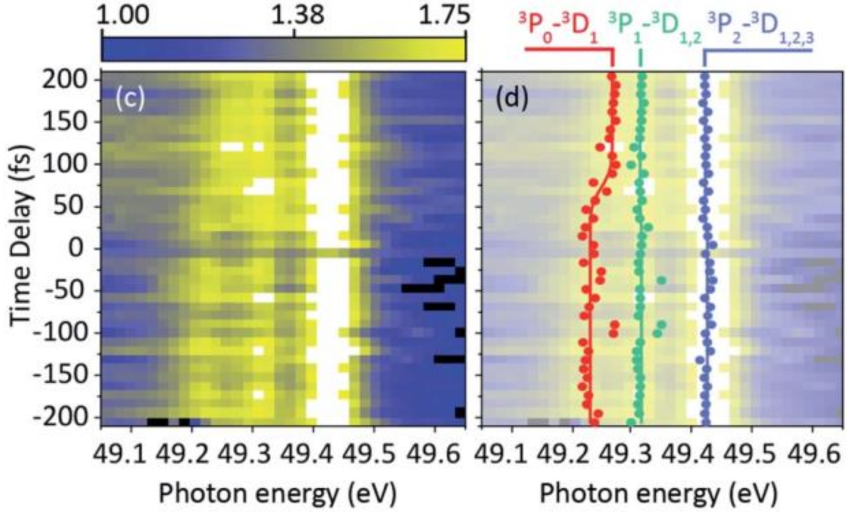
XUV pump–XUV probe transient absorption spectroscopy at FELs

Thomas Ding,*[a] Marc Rebholz,[a] Lennart Aufleger,[a] Maximilian Hartmann,[a] Veit Stooß,[a] Alexander Magunia,[a] Paul Birk,[a] Gergana Dimitrova Borisova,[a] Carina da Costa Castanheira,[a] Patrick Rupprecht,[a] Yonghao Mi,[a] Thomas Gaumnitz,[b] Zhi-Heng Loh,[c] Sebastian Roling,[d] Marco Butz,[e] Helmut Zacharias,[e] Stefan Düsterer,[f] Rolf Treusch,[f] Christian Ott[a] and Thomas Pfeifer[a]

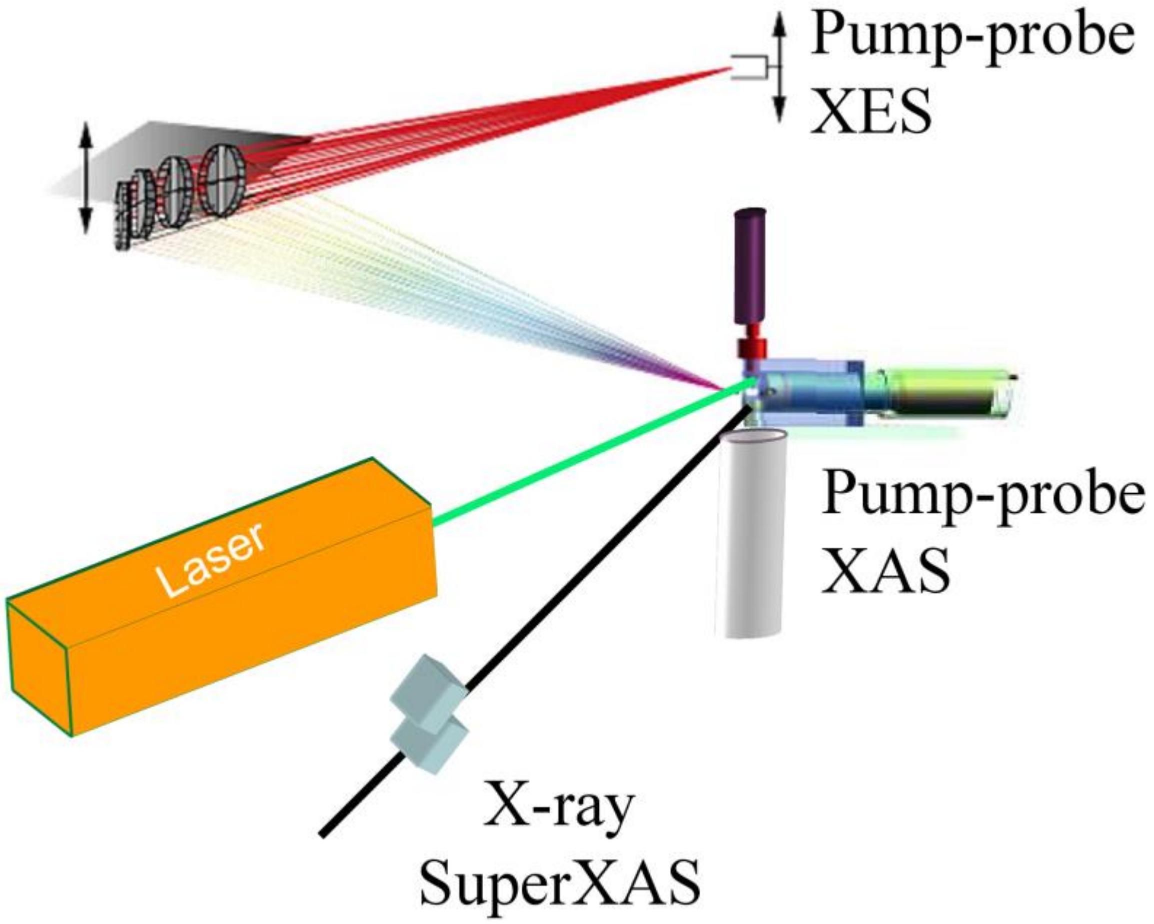
Level scheme of the resonant coupling mechanism of the Ne2+ ion. Observed absorption lines correspond to the ground-state splitting, while the 1 meV-scale excited state splitting is unresolved. The observed sets of multiplet transitions are indicated in blue, green, and red.



Other Possible Scenarios:

Pump:FEL
Probe: Optical laser
Problems to overcome:
Reference
Synchronization (jitter effect)
Overlap control (spatial resolution)
Beam fluctuations

Pump: Optical laser
Probe: FEL
Problems:
Tuning to the required edge (monochromation)
Overlap
Fluctuations



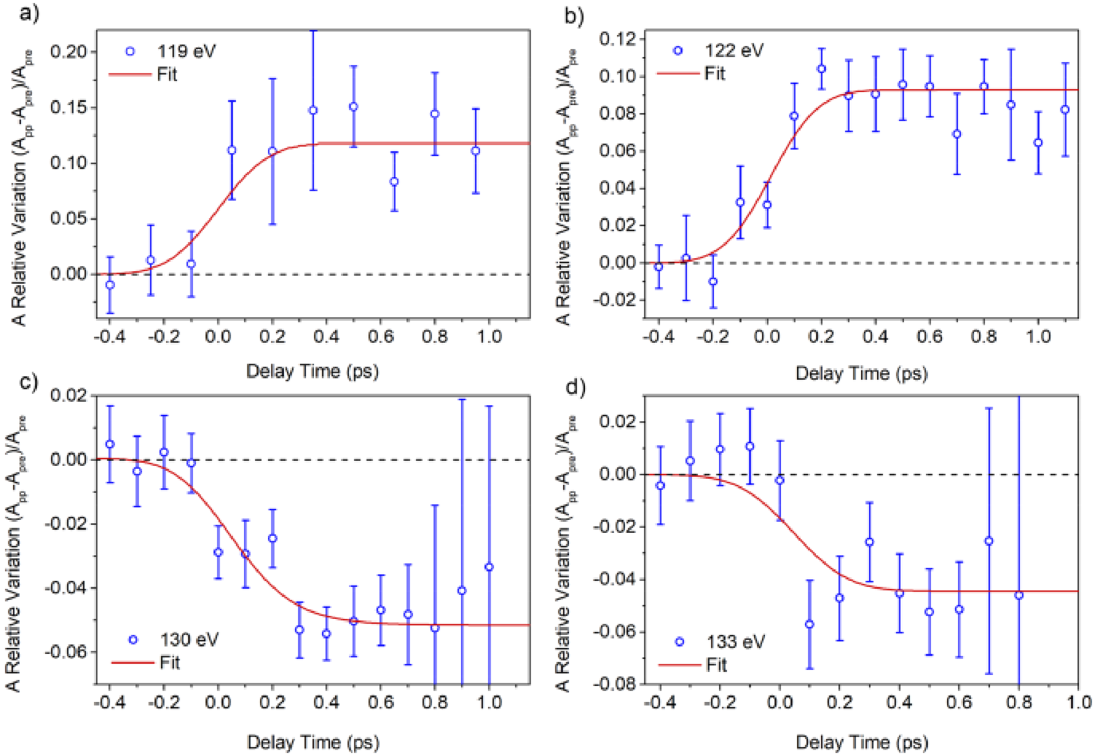
Nano science FEL

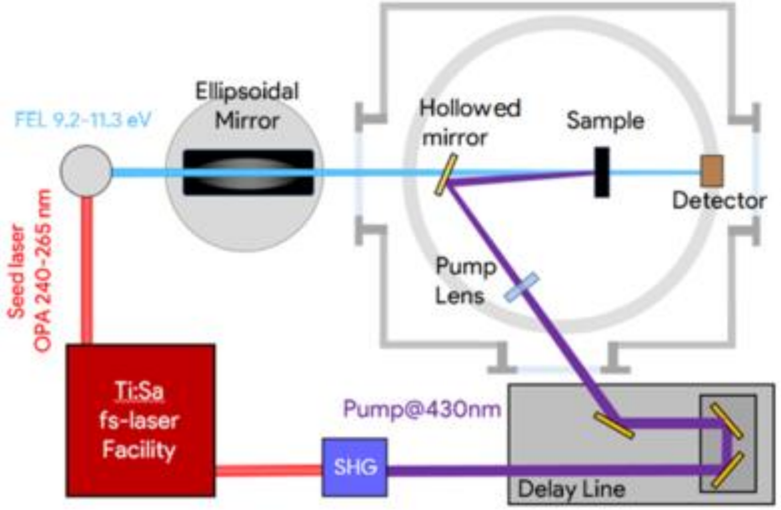
Free carriers
State filling
lattice heating
Band Modification
Free carrier absorption
Shape and size (Confinement)

NANO LETTERS
pubs.acs.org/NanoLett
Letter

Ultrafast Dynamics of Plasmon-Mediated Charge Transfer in Ag@CeO_2 Studied by Free Electron Laser Time-Resolved X-ray Absorption Spectroscopy

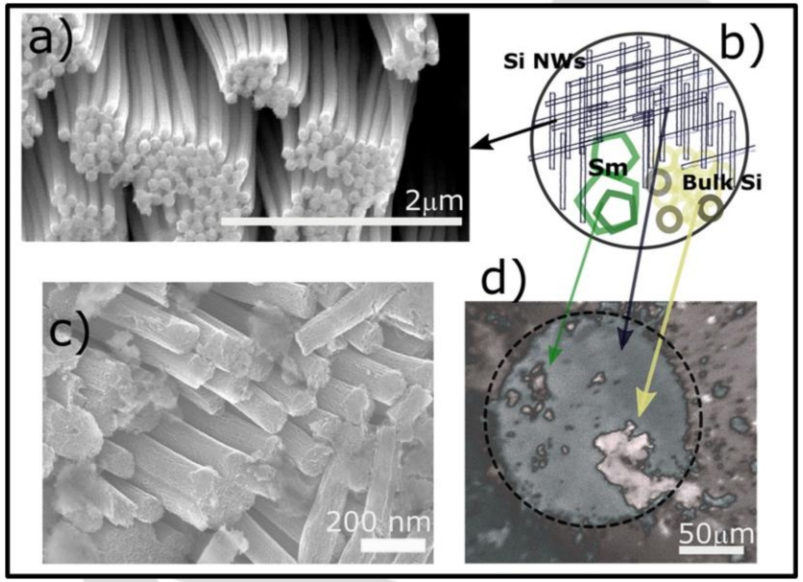
Jacopo Stefano Pelli Cresi,* Emiliano Principi, Eleonora Spurio, Daniele Catone, Patrick O'Keeffe, Stefano Turchini, Stefania Benedetti, Avinash Vikatakavi, Sergio D'Addato, Riccardo Mincigrucci, Laura Foglia, Gabor Kurdi, Ivaylo P. Nikolov, Giovanni De Ninno, Claudio Masciovecchio, Stefano Nannarone, Jagadesh Kopula Kesavan, Federico Boscherini, and Paola Luches

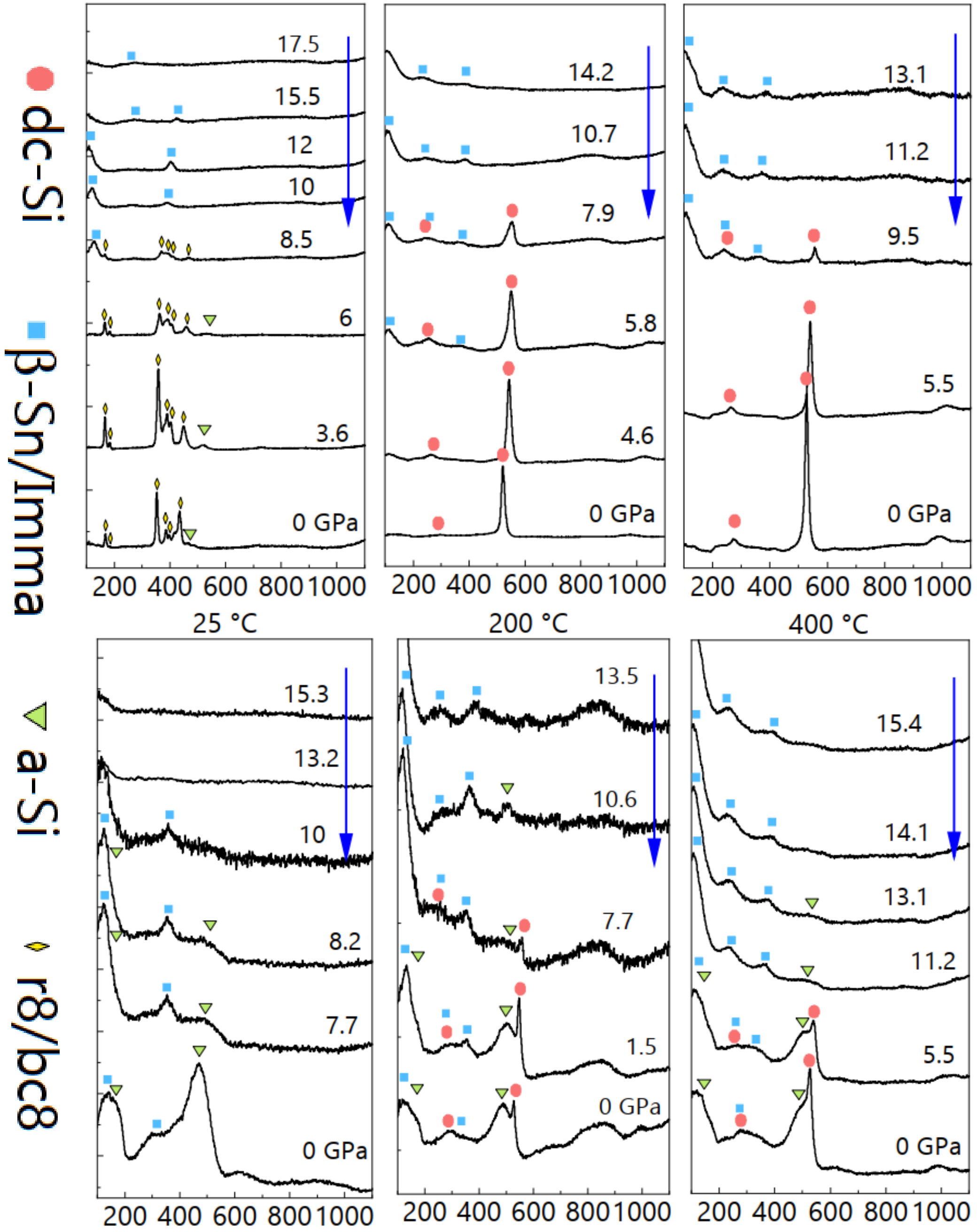




- Ce atoms undergoes an ultrafast change following photoexcitation of the localized surface plasmon resonances in the Ag NPs.
- The sign and the amplitude of the observed variations at the different energies and their ultrafast nature demonstrate that the decay of the LSPR in the Ag NPs involves electron transfer processes, which is the dominant process below 1 ps.

Π-Silicon nanowires with distinct structural dynamics





Porous Silicon nanowires show distinct structural dynamics compared with the bulk one due to the Size and shape affecting electronic structure of the material.

Rezvani et.al. Applied Physics Letters 119 (5), 053101

Comparison of the phase diagram:

Porous silicon nanowires phase transformations at high temperatures and pressures

Cite as: Appl. Phys. Lett. **119**, 053101 (2021); doi: 10.1063/5.0057706
Submitted: 24 May 2021 · Accepted: 19 July 2021 ·
Published Online: 2 August 2021

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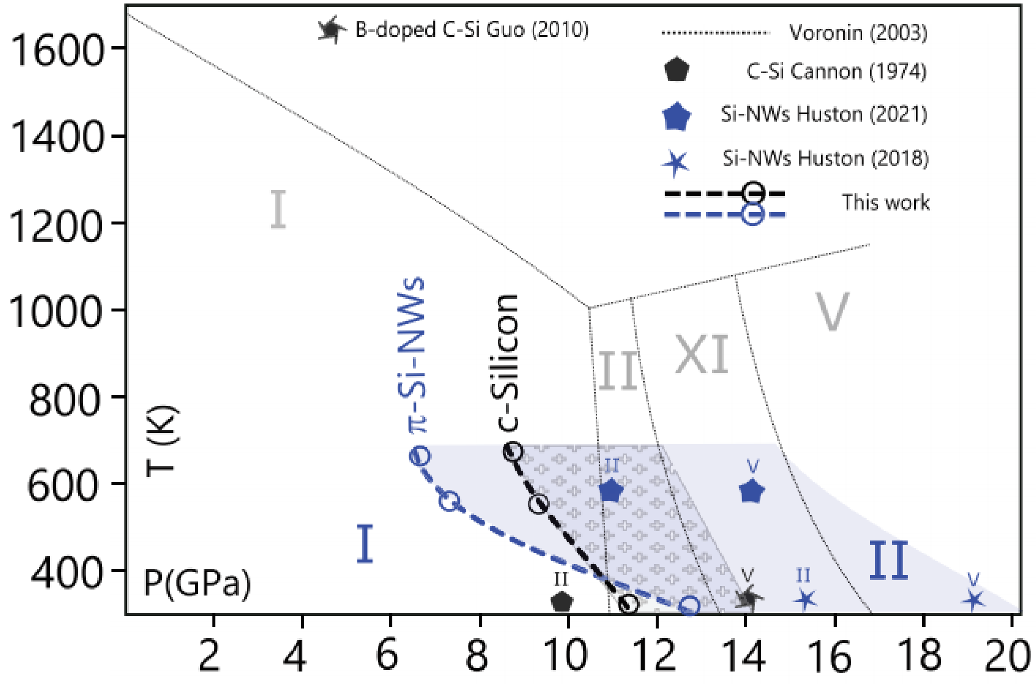
S. J. Rezvani,[1,a)] Y. Mijiti,[2] and A. Di Cicco[2,a)]



- Π- Silicon nanowires phase diagram.
- Large phase persistence band
- Co-existence of the phases
- Modified DOS

Conclusions:

- FEL sources open new opportunities for investigating extreme and non-equilibrium states in condensed matter.

- The XAS technique is shown to be particularly useful and ultrafast single-shot are possible.

- Present results at the available EUV and soft x-ray facilities show that challenging single-shot XAS experiments can be done also using pump-probe schemes. Proper diagnostics for sample control and data normalization for XAS is still under development.

- The realization of challenging XAS experiments at the new Eupraxia facility would need a dedicated beamline including diagnostics and solutions carried out in other FEL facilities.

- FEL opens a new road to investigate the effect of DOS dynamics at low dimensions.