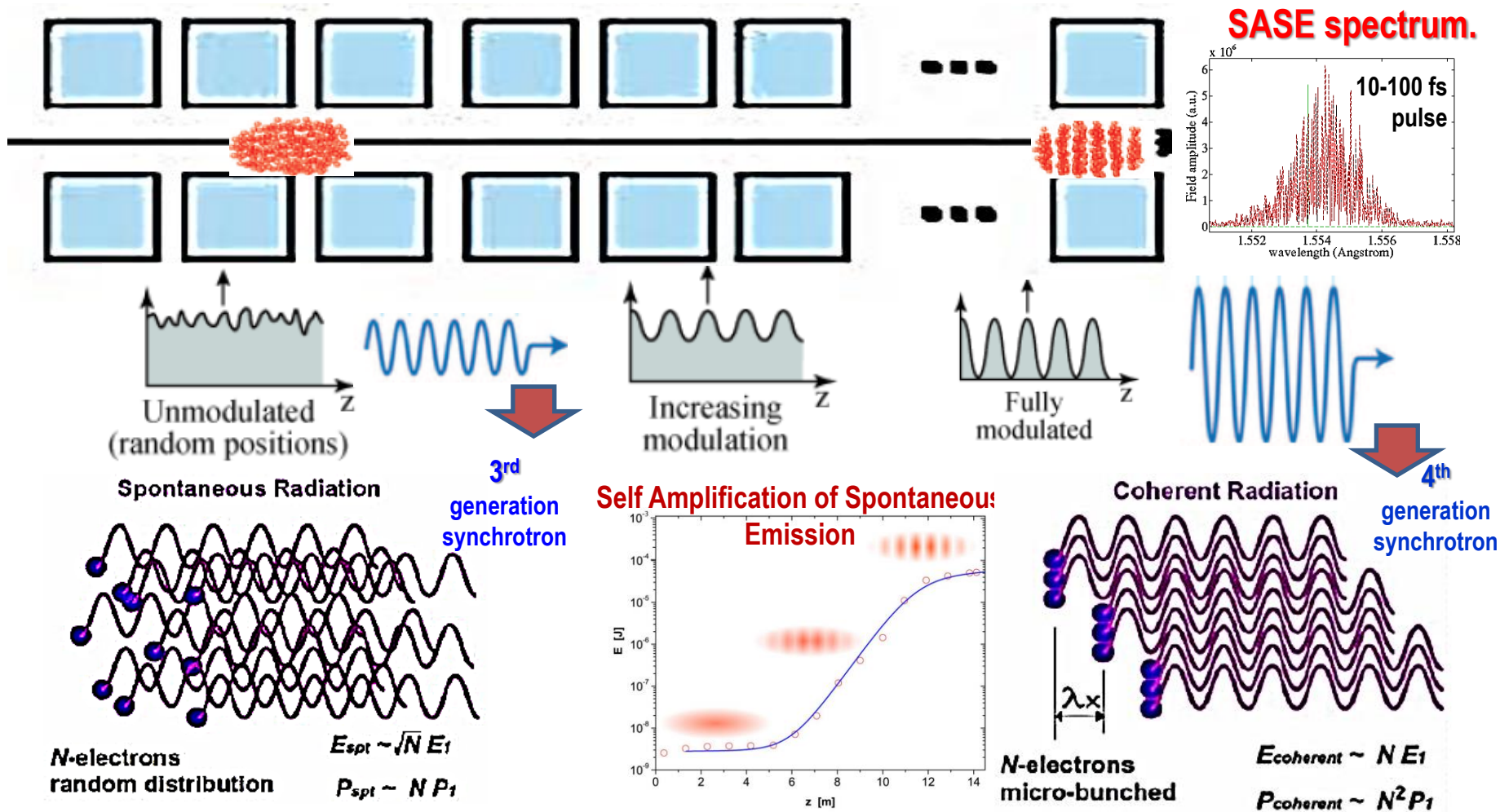


COHERENT PHOTON SOURCES: FREE ELECTRON LASER AND THEIR MULTIDISCIPLINARY APPLICATIONS

- FEL production, characteristics and properties of FEL light.
- Selected application examples.
- What is next?

FEL PRODUCTION: Electron motion in a very long undulator evolves to a free electron laser

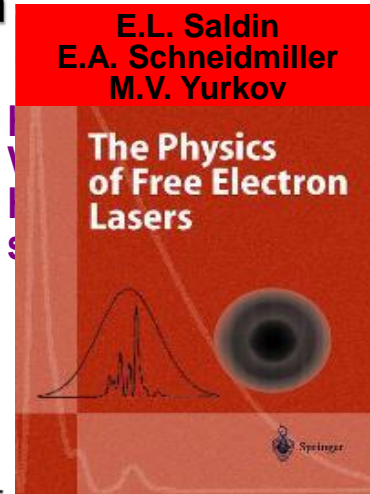
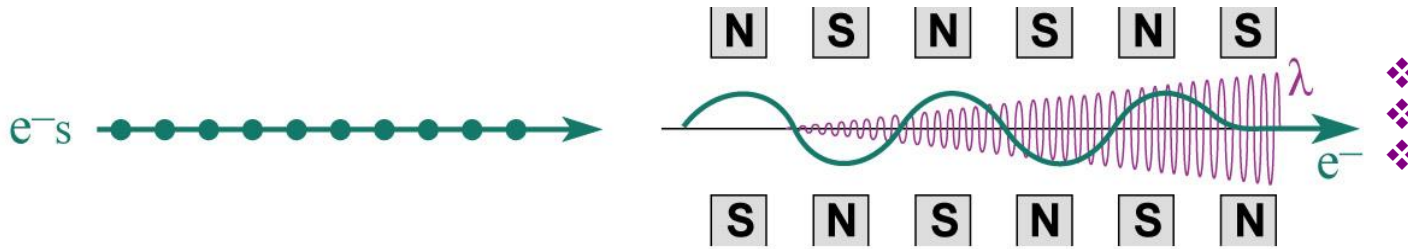
With a very long undulator the radiated fields become stronger and lead to microbunching, i.e. transform the random positions and motions of electrons into correlated waves of electrons, emitting radiation in phase.



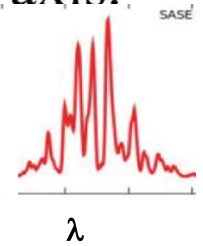
Courtesy of K-J. Kim

Gen_Saturator_FEL_graph.e

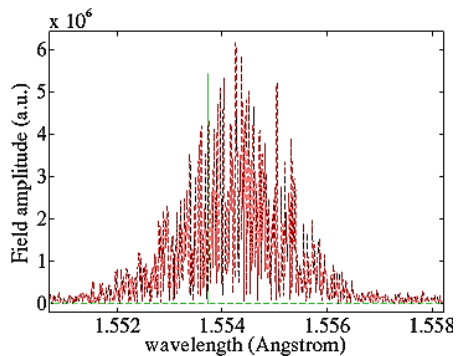
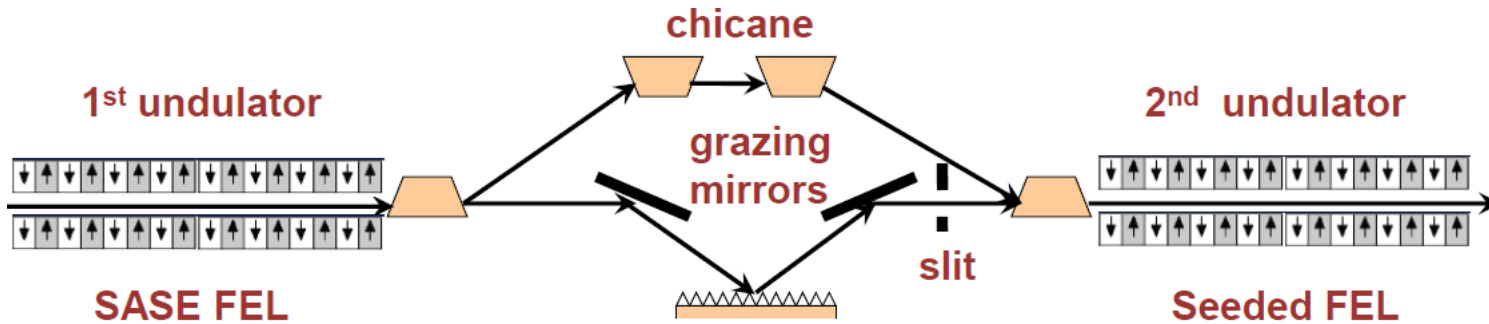
SASE-FEL Physics: increase of coherence power N as result of constructive interference of emitted radiation



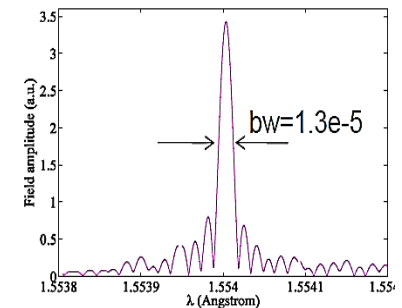
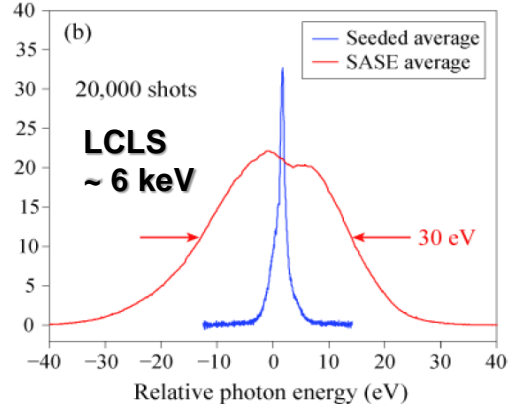
- Uniformly distributed particles (beam) into undulator.
- Emission of radiation (“spontaneous” emission).
- Wave grows enough (undulator radiation) to begin affecting particle dynamics through $m\mathbf{a} = -e\mathbf{E}$ radiation.
- Transverse coupling between \mathbf{E}_{rad} and transverse velocity \mathbf{v}_x (in undulator) leads to energy exchange between fields and particle (zero net at first) $\frac{dE_e}{dt} = mc^2 \frac{d\gamma}{dt} = \mathbf{F} \cdot \mathbf{v} = -e \mathbf{E} \cdot \mathbf{v}_x$.
- Modulated velocities with increments in \mathbf{v}_x lead to bunching on axis.
- Electron density modulation leads to stronger radiation, $P_{\text{Tot}} \propto \frac{Q^4}{M^2} \sim N^2 \frac{e^4}{m^2}$. **Time/energy structure: envelope of a series of sub-pulses with random intensity, time duration, bandwidth and phase.**
- Stronger fields (wave) drive stronger transverse velocity.
- Stronger \mathbf{v}_x drives stronger bunching, . . . stronger fields, . . . FEL action.



- First undulator generates SASE.
- X-ray monochromator (grating for soft X-rays or Si/diamond crystal for hard X-rays) filters SASE and generates seed.
- Chicane delays electrons and washes out SASE micro-bunching.
- Second undulator amplifies seed to saturation.



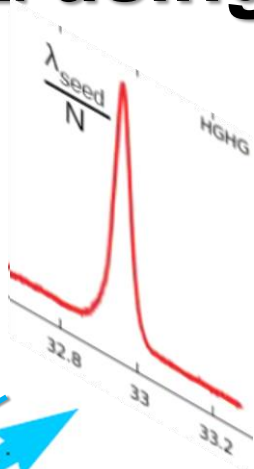
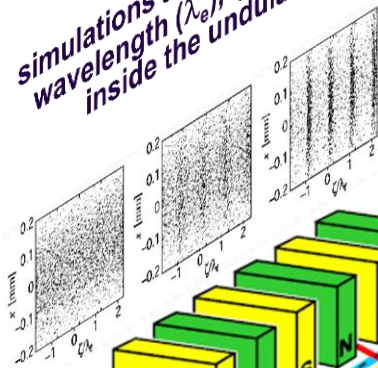
**narrow bandwidth
enhanced peak power**



Seeding SASE-FEL using optical laser

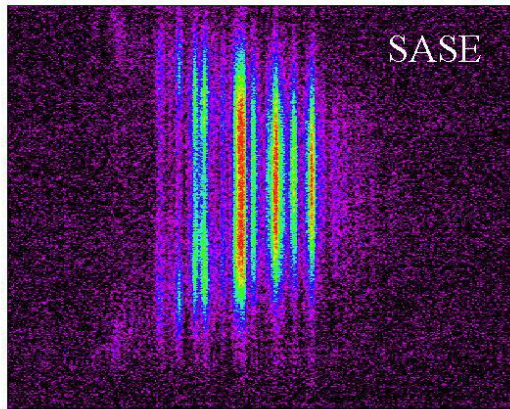
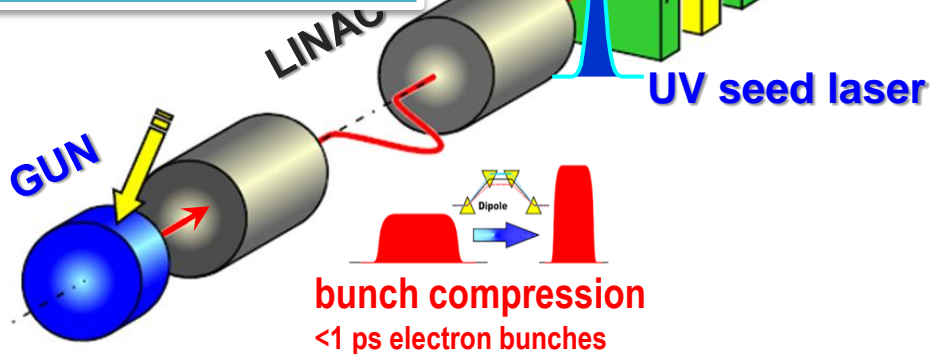
High Gain Harmonic Generation (HGHG): seeding (modifying) the emitting electron bunch with an external laser pulse controlled in all the relevant photon parameter

simulations at the radiation wavelength (λ_e), ζ - distance inside the undulator



“SASE” FEL – several separate “waves” of electrons with uncorrelated phase. Less peak power, broader spectrum.

The properties of the FEL radiation are entangled with those of the seed laser. Defined energy-time profile.



31.8 32 32.2 32.4 32.6 32.8 33
 λ (nm)



Free Electron Lasers in operation and coming 2016

Elettra Sincrotrone Trieste

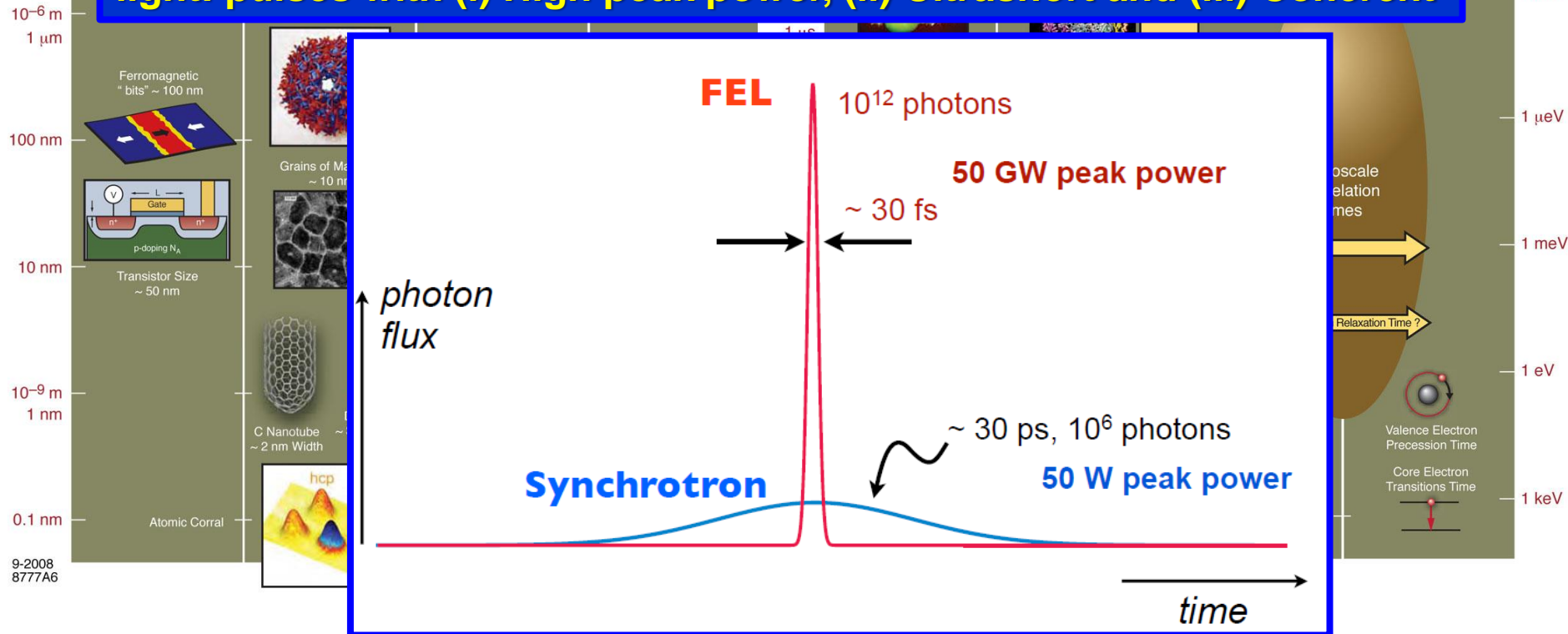
	LCLS	LCLS II	Eu-XFEL	SACLA	FLASH	FLASH II	FERMI	SwissFEL	PAL XFEL	Shanghai XFEL
Shortest wavelength	1.5 Å	1 Å	0.5 Å	1 Å	40 Å	40 Å	40 Å	1 Å	1 (0.6) Å	1 Å
Undulator type hard X-ray.	Fixed gap	Variable gap	Variable gap	In-vacuum Var. gap	n.a.	n.a.	n.a.	In-vacuum var. gap	Variable gap	Variable gap
Undulator type soft X-ray.	n.a.	Variable gap	Variable gap	n.a.	Fixed gap	Variable gap	Apple II	Apple II	Apple II	?
Injector	S-band RF gun	S-band RF gun	L-band RF gun	Pulsed Diode	L-band RF gun	L-band RF gun	S-band RF gun	S-band RF gun	S-band RF gun	S-band RF gun
Cathode	Cu	Cu	Cs ₂ Te	CeB ₆ (thermionic)	Cs ₂ Te	Cs ₂ Te	Cu	Cu	Cu	Cu
Main linac technology	n.c. Pulsed	n.c. pulsed	s.c. pulsed	n.c. pulsed	s.c. pulsed	s.c. pulsed	n.c. pulsed	n.c. pulsed	n.c. pulsed	n.c. pulsed
RF frequency	S-band	S-band	L-band	C-band	L-band	L-band	S-band	C-band	S-band	C-band
RF Rep. rate	120 Hz	120 Hz	10 Hz	60 Hz	10 Hz	10 Hz	10-50 Hz	100 Hz	120 Hz	60 Hz
FEL pulses/RF pulse	1	1	2700	1	2700	2700	1	2	1	1
max. bunch charge	0.25 nC	0.25 nC	1 nC	0.2 nC	1 nC	1 nC	0.5 nC	0.2 nC	0.2 nC	0.2 nC
max. electron energy	13.6 GeV	14 GeV	17.5 GeV	8 GeV	1.2 GeV	1.2 GeV	1.5 GeV	5.8 GeV	10 GeV	6.4 GeV
No. RF stations	81	81	29	69	5	5	15	34	49	?
Approx. facility length	1.7 km	1.7 km	3.4 km	0.8 km	0.32 km	0.32 km	0.5 km	0.7 km	1.1 km	0.6 km
Start operation	2009	2017	2015	2011	2005	2013	2010	2016	2015	2019

Probing matter on nm length scales and fs time scales

Characteristic Nanoscales in Matter

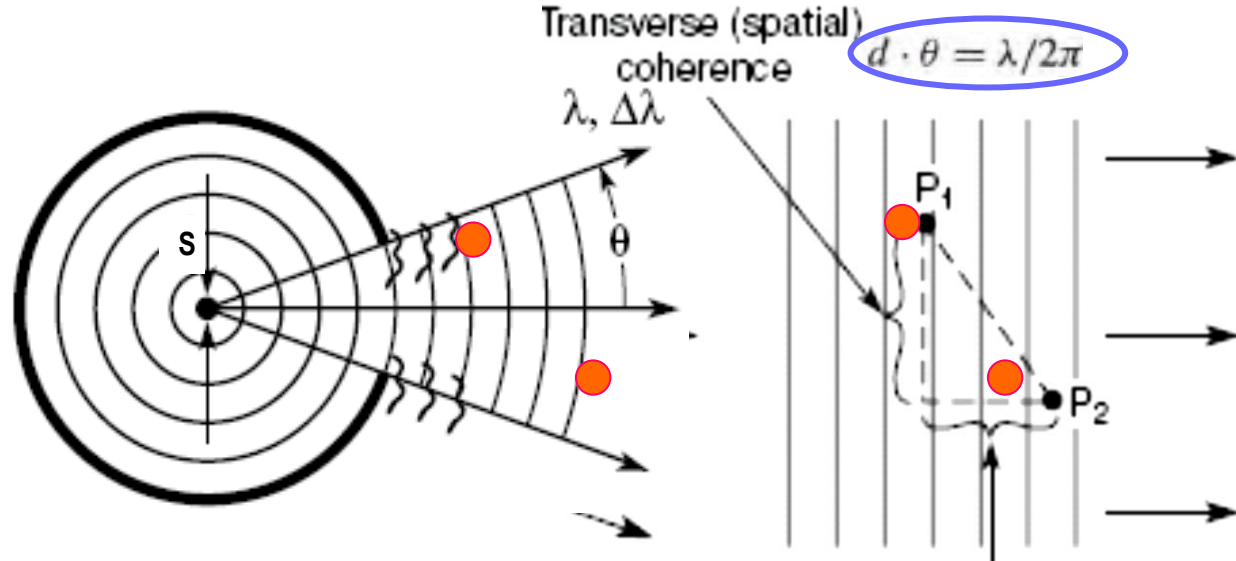
Characteristic Times in Matter

Unique new opportunities thanks to the distinct properties of FEL light: pulses with (i) High peak power, (ii) Ultrashort and (iii) Coherent



FEL: temporal and spatial coherence

A source with finite size and spectral bandwidth, restricted to radiate over a narrow solid angle, generates fields with strong phase and amplitude correlation to a limited extent

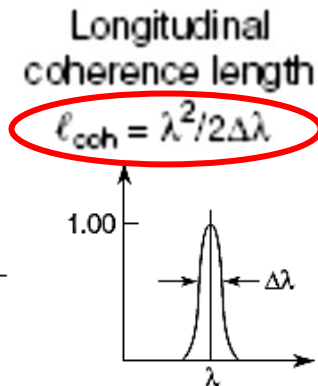
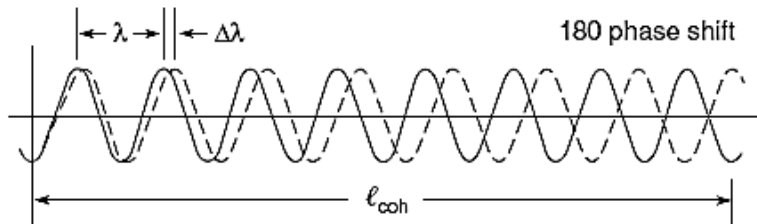


Source of finite size divergence & duration

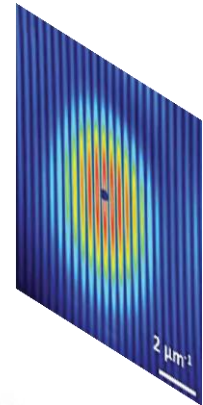
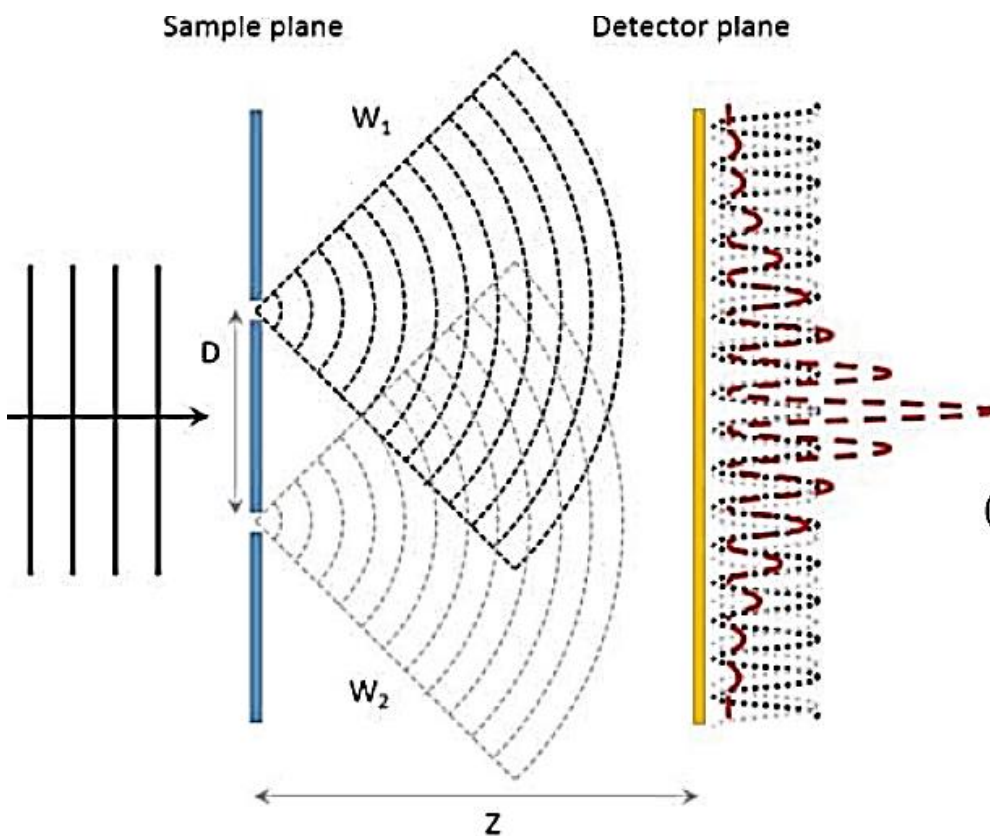
Transverse (spatial) Coherence:
depends on the source size, S , and angle of emission, Θ .

Longitudinal (temporal) Coherence:
depends on the finite spectral band width, $\Delta\lambda/\lambda$.

These requirements become very stringent for shorter wavelengths.

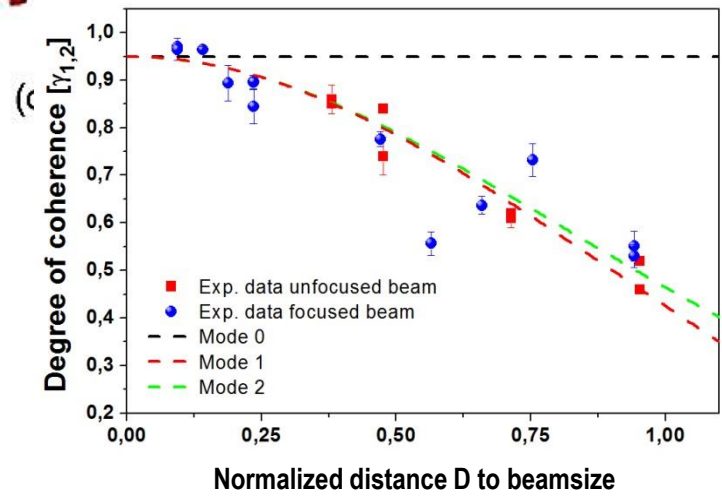


Young's double slit experiments as measure of spatial coherence



$$L_{coh}^T = \frac{2 \cdot \lambda \cdot Z}{\pi \cdot S}$$

the spatial distance along the two generated wavefronts out of phase by a factor $\pi/2$.



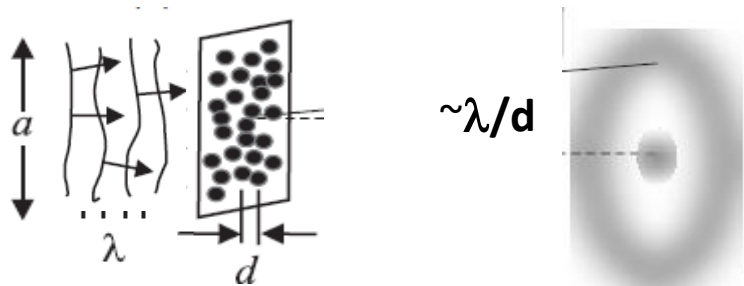
- Self-interference only
- Electric fields chaotic
- Intensities add



- Phase coherent electric fields
- Electric fields from all particles interfere constructively

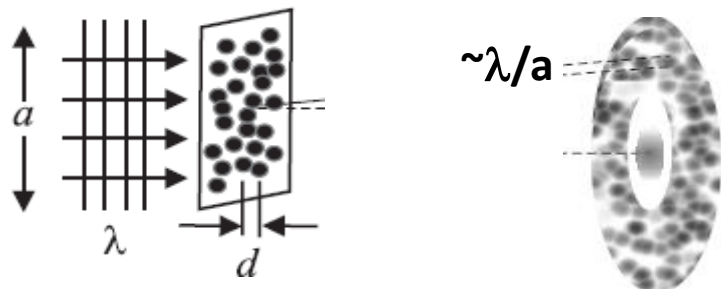
Coherent Diffraction Imaging (CDI):

based upon the principle of coherent scattering in combination with a method of direct phase recovery called oversampling



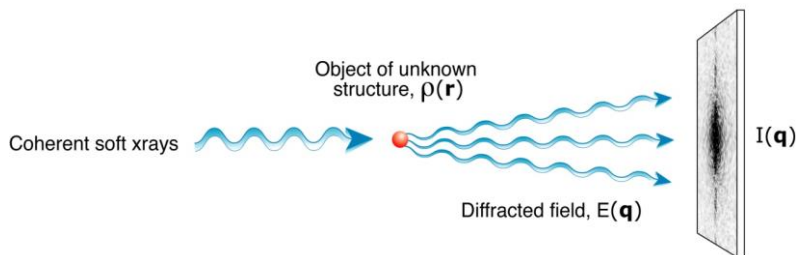
Incoherent illumination: coherence length larger than the sample structures

Diffuse scattering that averages over all features resulting from slightly different wavefronts



Coherent illumination: coherence length larger than the sample

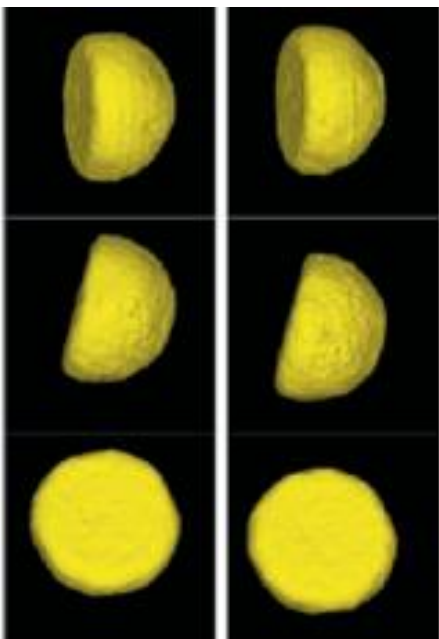
Speckles due to interference of wavefronts scattered from the features - information on the positions of each feature, obtained inverting the pattern.



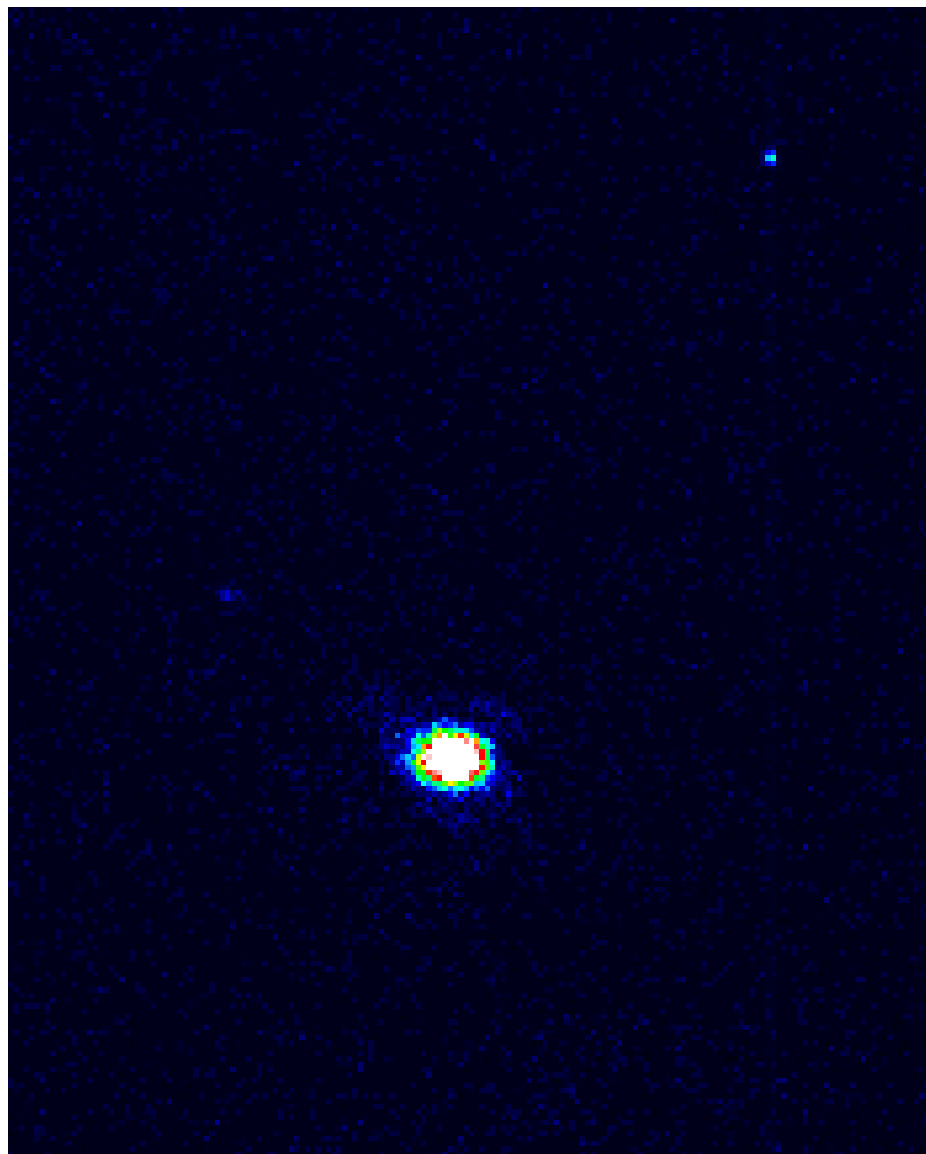
The scattered amplitude is Fourier transform of real space electron density $f(r)$ of the object: $F(k) = \int f(r) e^{-2\pi i k \cdot r} dr$

- Proposed by Sayre to visualize the electron-density distribution in non-crystalline materials (1980)
- Pioneering experiments: Kirz, Miao, Chapman, Spence, Robinson, (Nature 400, 342; ibid. 442, 63; 448, 679; MRS Bull 29, 177, PNAS 102, 15343), Science, 316, 5830 etc)

3D CDI of Ag cubes using synchrotron



Surface plots of
reconstructed shapes



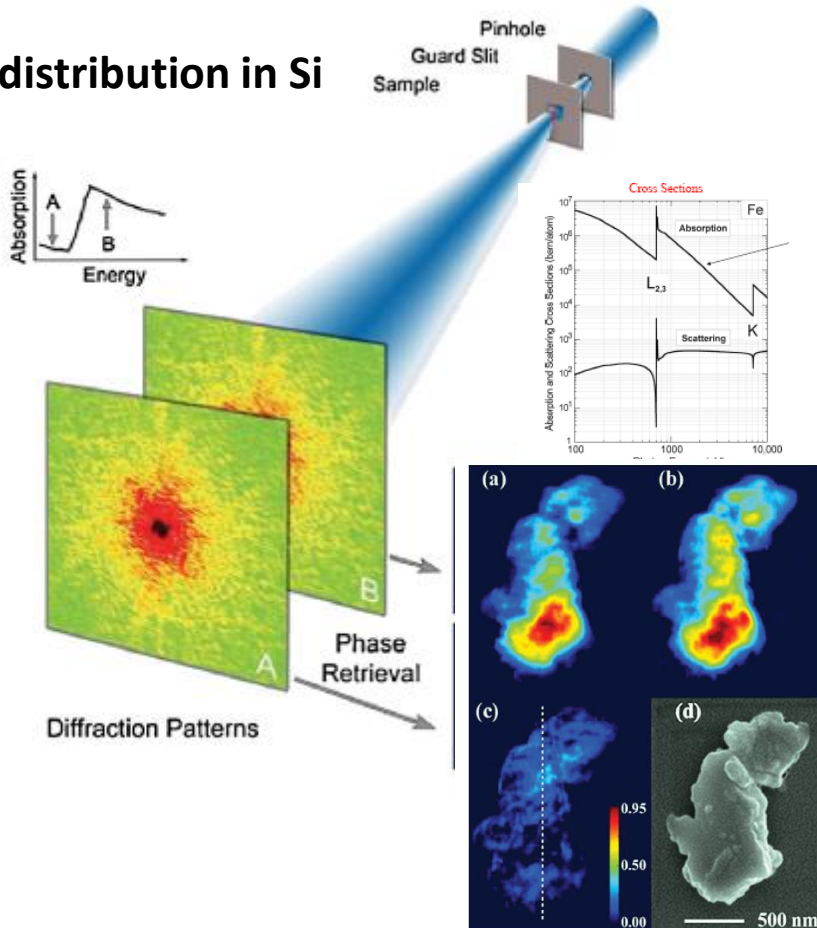
Rocking scan of Ag
cubes with 0.01° steps,

courtesy K. Robinson, PRL 87, 195505

Nanoscale Imaging of Buried Structures with Elemental Specificity Using Resonant X-Ray Diffraction Microscopy

Changyong Song,¹ Raymond Bergstrom,¹ Damien Ramunno-Johnson,¹ Huaidong Jiang,¹ David Paterson,² Martin D. de Jonge,³ Ian McNulty,³ Jooyoung Lee,⁴ Kang L. Wang,⁴ and Jianwei Miao^{1,*}

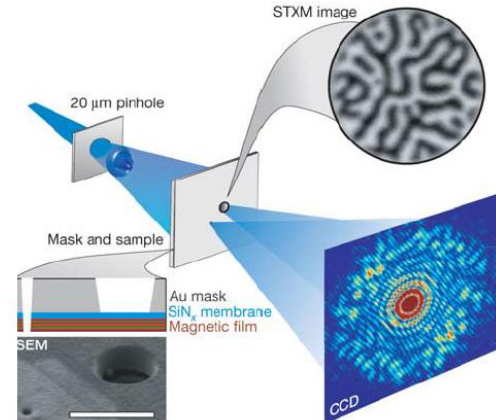
Bi distribution in Si



CDI is also sensitive to chemical states via near-edge resonances and can be extended to exploit other contrast mechanisms depending on resonant transitions such as **x-ray magnetic circular dichroism, electronic orbital** as well as **chemical state**.

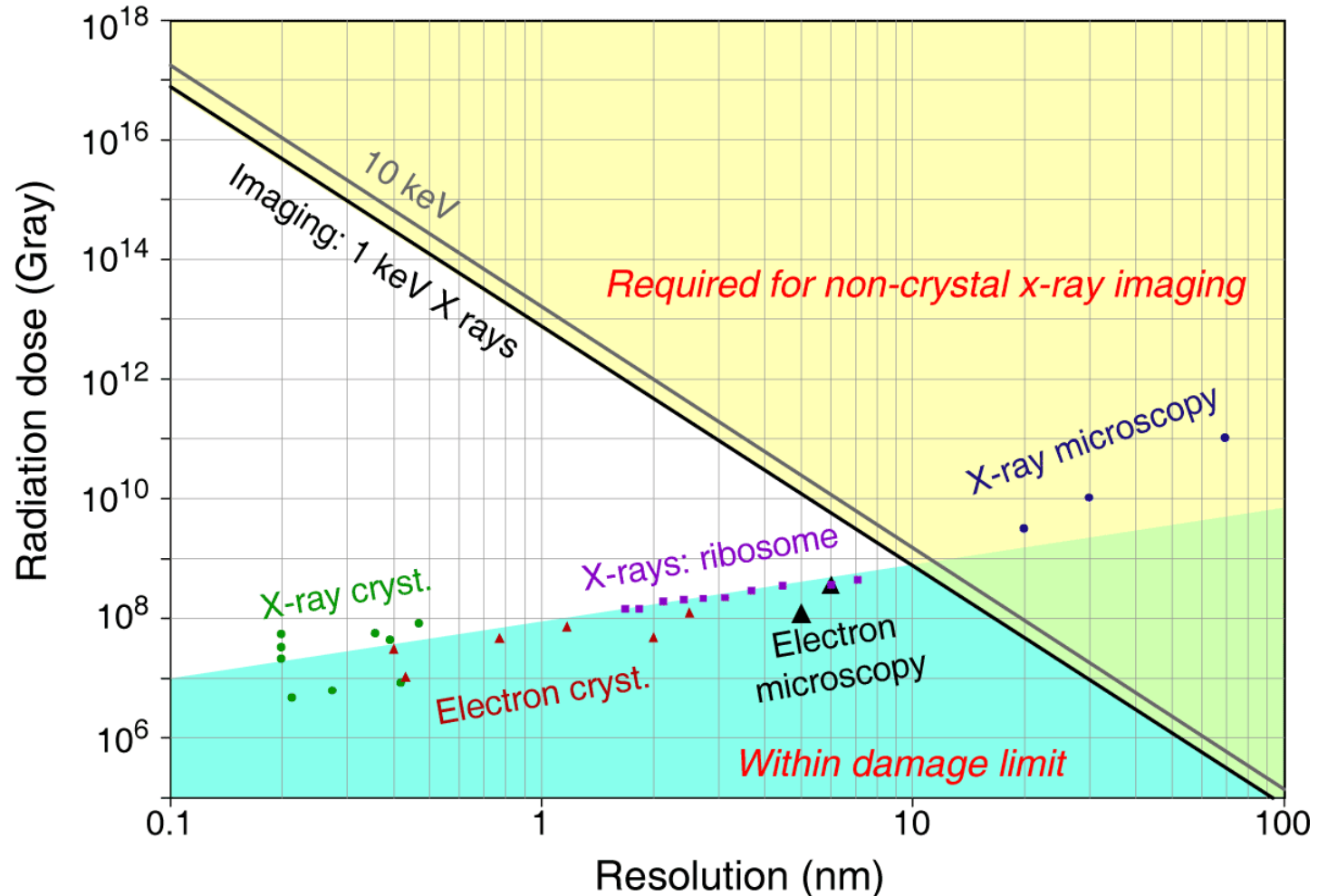
Holographic image of random magnetic domains in a Co/Pt ML sample, Co L₃-edge absorption edge.

S. Eisebitt¹, J. Lüning², W. F. Schlotter^{2,3}, M. Lörger¹, O. Hellwig^{1,4}, W. Eberhardt¹ & J. Stöhr² NATURE, 432, 885 (2004)



X-ray exposure determines the achievable resolution. Radiation damage sets the dose

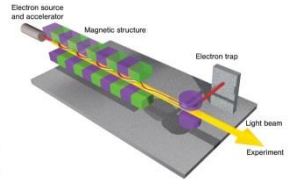
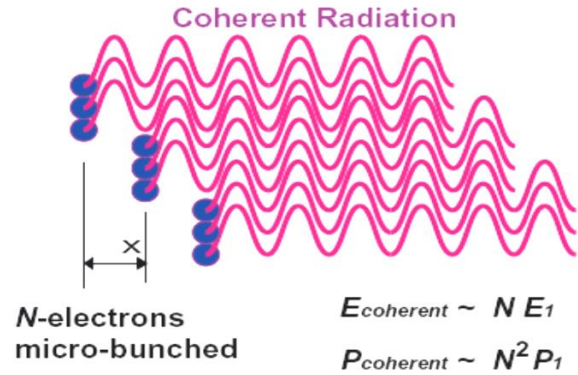
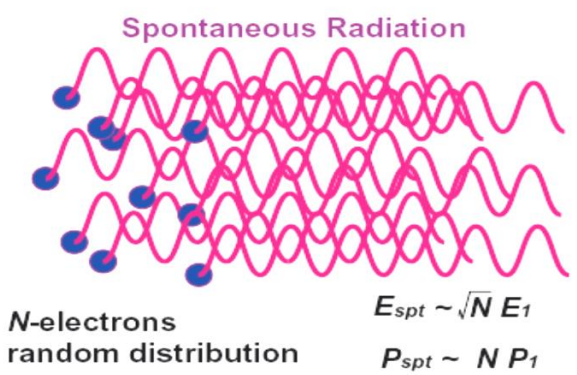
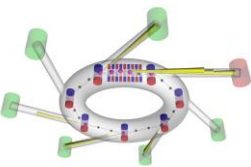
For each scattered photon that contributes to the diffraction pattern there are about 10 x-ray photons absorbed. This absorption deposits energy into the sample and leads to sample degradation.



Coherent Diffraction Imaging: Synchrotron vs FEL Radiation

long pulses (sub-ns) max $\sim 10^8$ photons/pulse

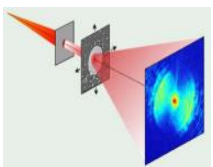
Fs pulses $> 10^{11}$ photons/pulse



$\times 10^6 \dots 10^8$ times higher

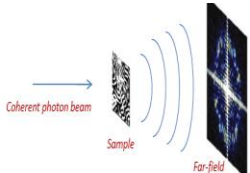
Synchrotron radiation:

pinhole and monochromators for spatial and spectral filtering, but at the expense of intensity!

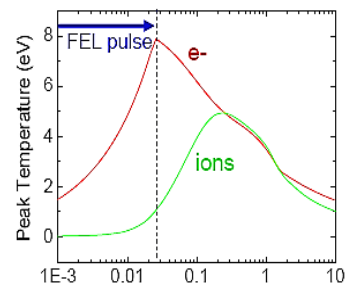


FEL (FLASH, LSLs, SACLA, FERMI):

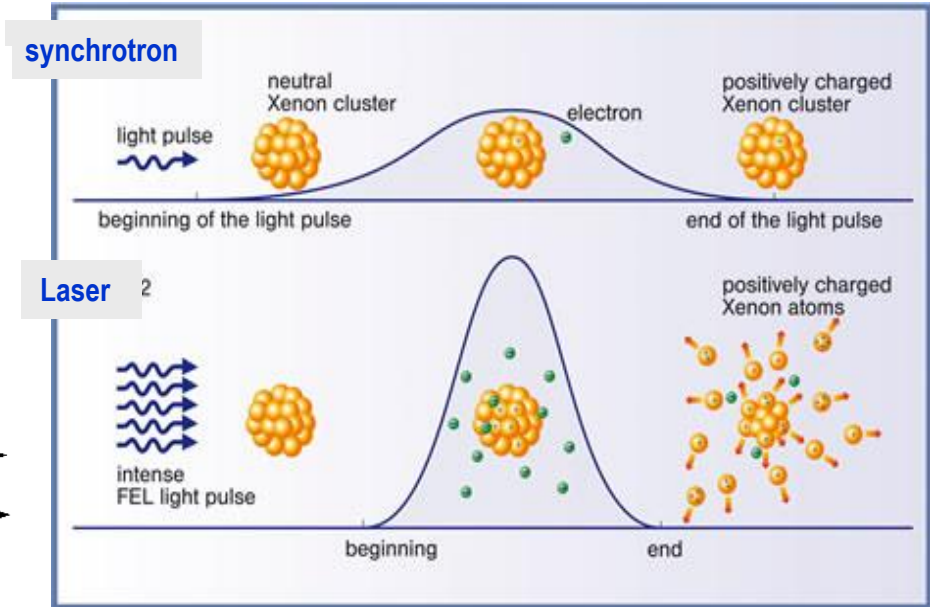
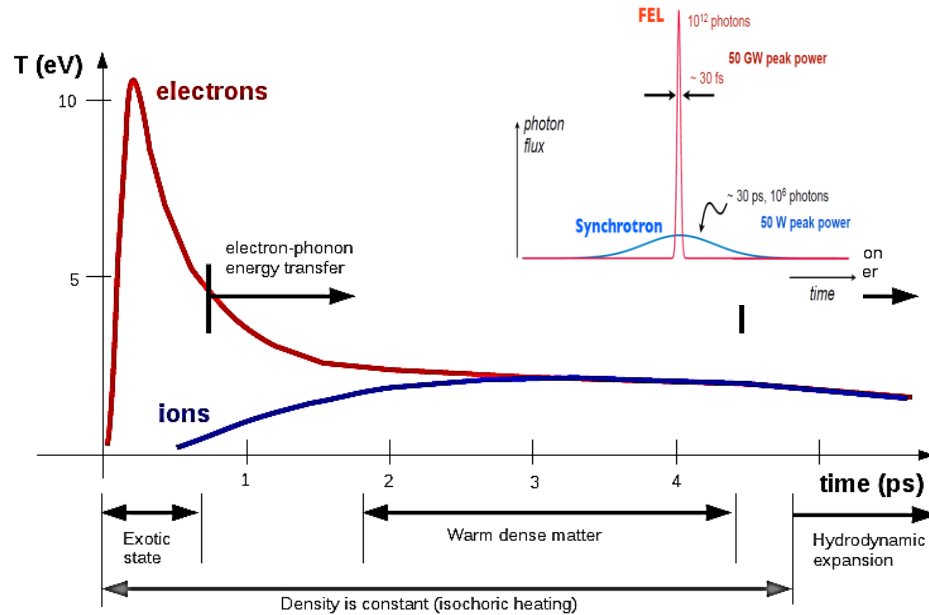
natural space coherence: each electron - spontaneous emission that overlap each other in phase



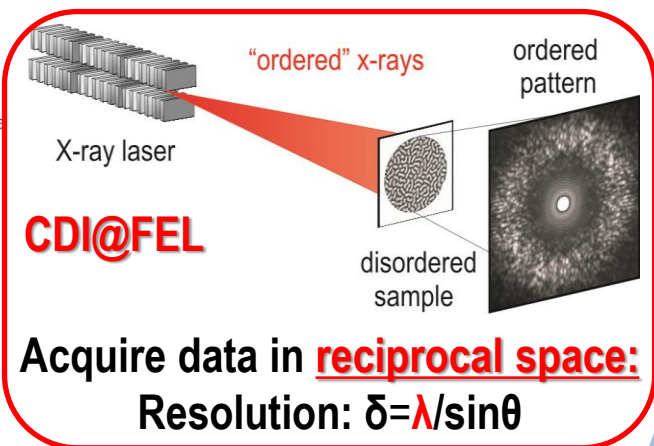
Ultra-short (fs) and ultra-bright coherent FEL pulses allow imaging with single pulse before the radiation damage manifests itself !



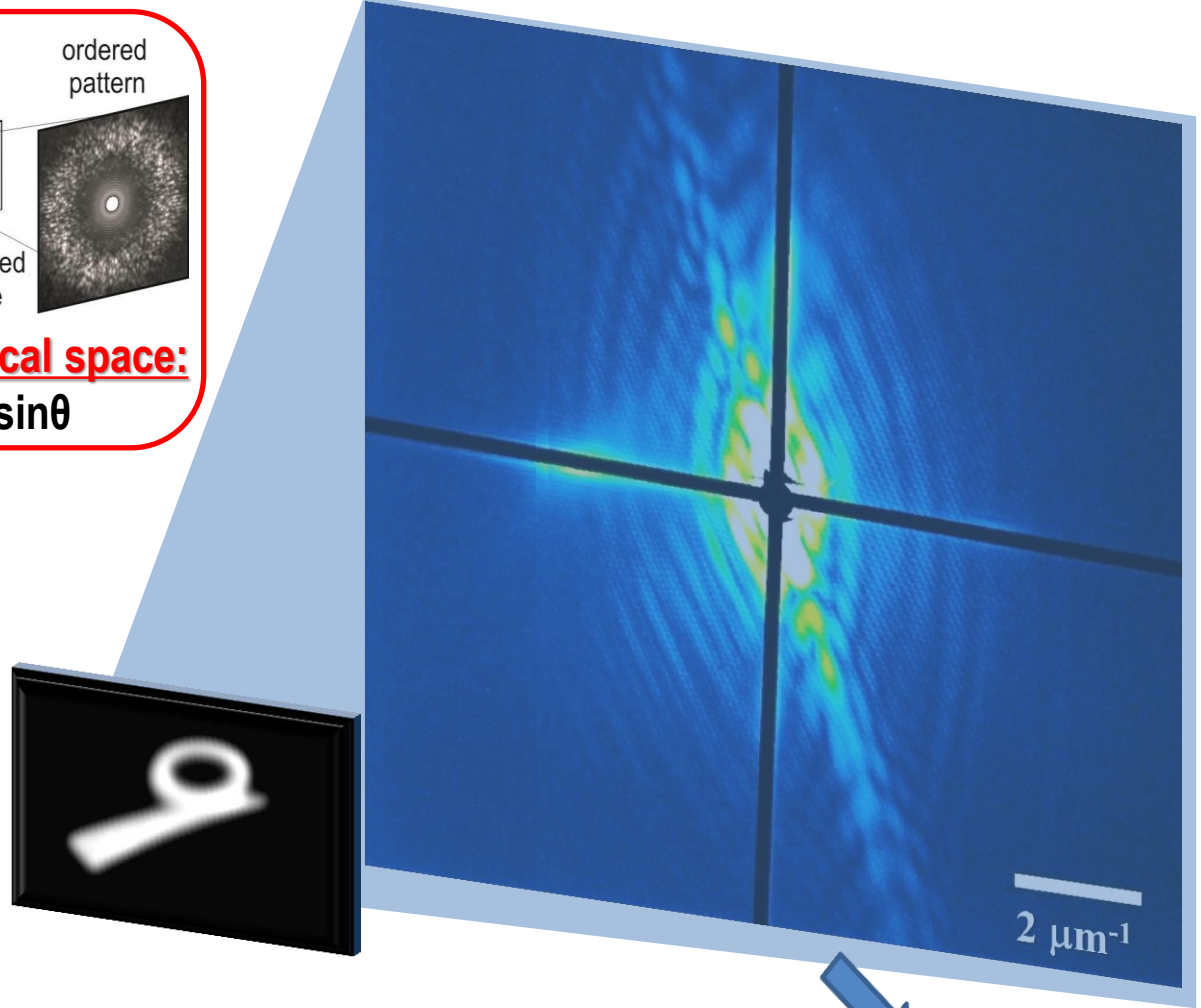
How matter will respond when exposed to a very high power short fs pulses



When matter is irradiated with very intense light, exciting deeper electronic levels, unusual processes occur which do not happen upon irradiation with less intense light: exotic non-equilibrium state with electrons at temperatures tens eV) and ions at RT (< 100 fs), electron-phonon energy transfer leading to warm dense matter (>1 ps), lattice expansion (> 5 ps) Coloumb explosion..

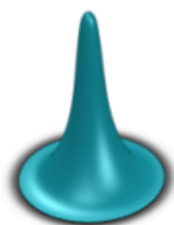


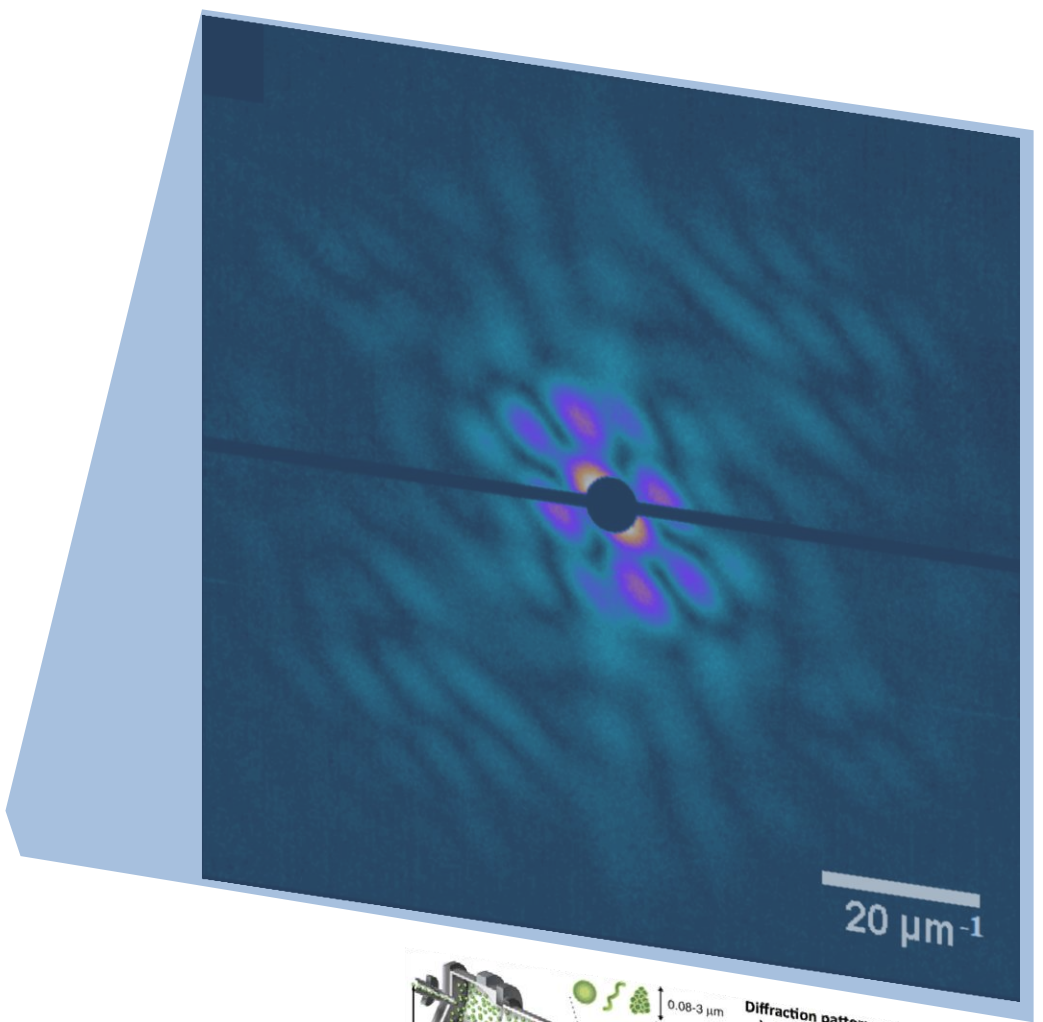
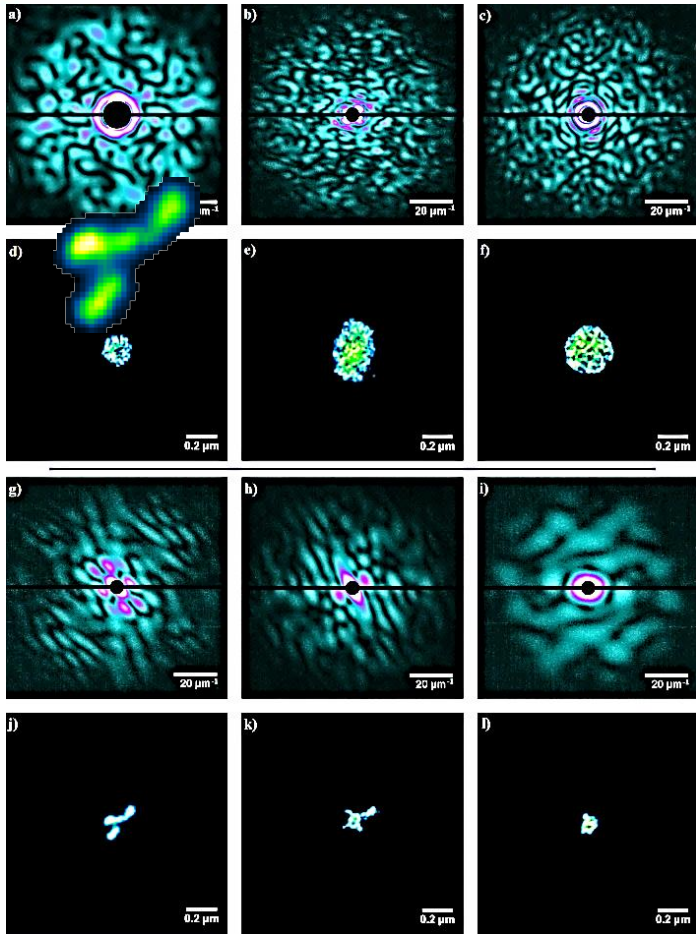
Chapman et al, *Nature Phys* 2 839 (2006)



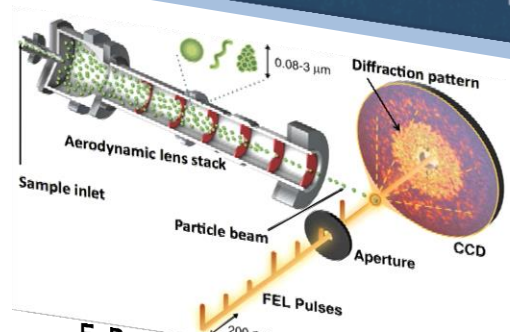
RECONSTRUCTED OBJECTS using phase retrieval

- ❖ Structure and dynamic phenomena in morphologically complex, disordered or particulate matter.
- ❖ Serial Crystallography





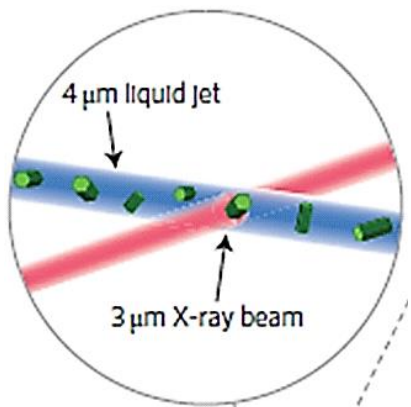
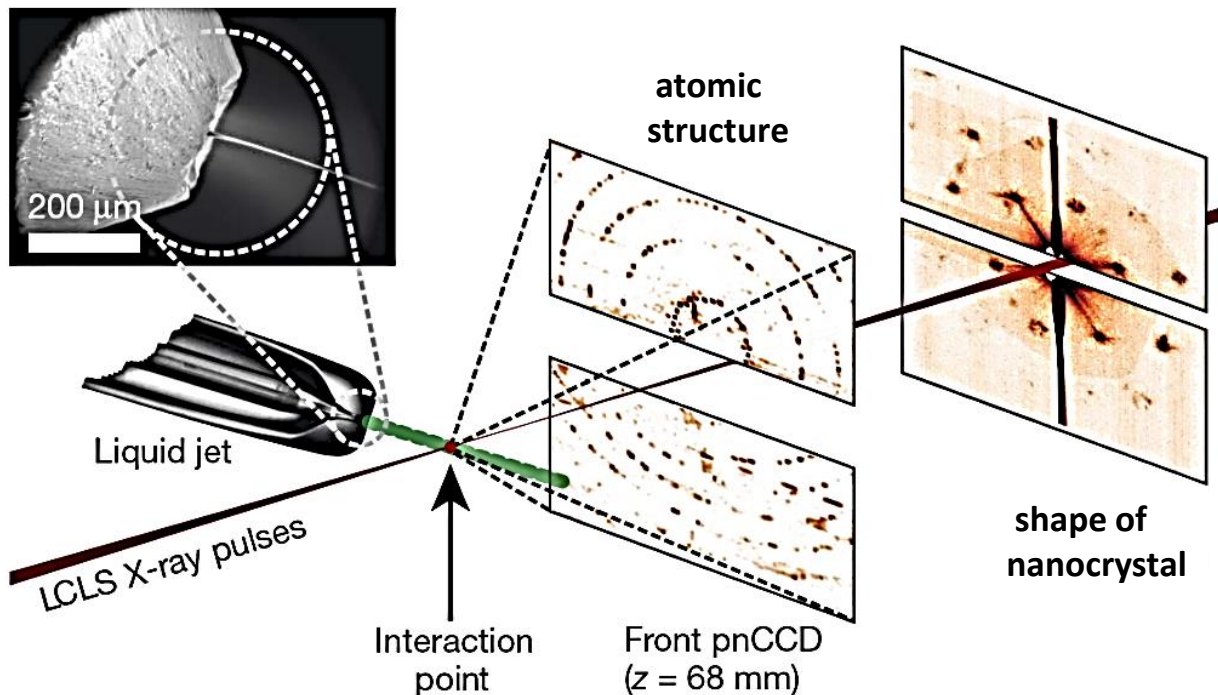
Appealing to explore the new collective properties resulting from the secondary structures of the assembled NP



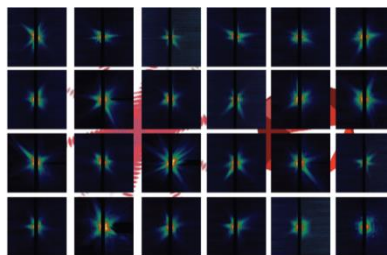
E. Pedersoli et al,
J. Phys. B: At. Mol. Opt. Phys. 46 (2013) 164033

Maya Kiskinova

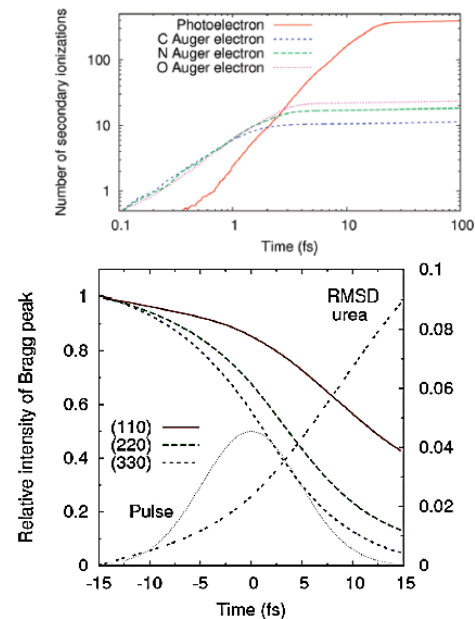




Randomly oriented patterns to be assembled to recover 3D image



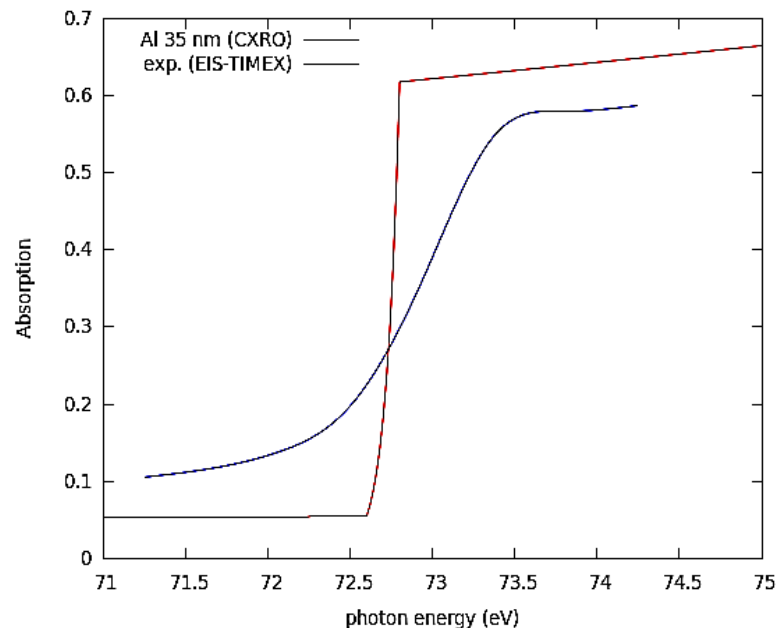
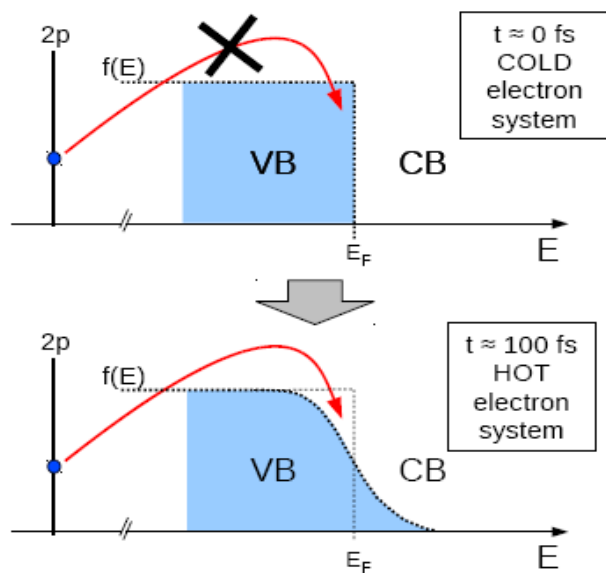
Need very short pulses



FEL \sim 6-8 keV: radius of gyration of the photoelectron cloud can reach 300 nm, and that of the Auger electron cloud - 8 nm: photoelectron cascade becomes bigger than a typical nanocrystal under consideration.

C. Caleman et al, ACS NANO 5, 136, 2011

Response of solid state matter exposed to a very high power short fs pulses: Al

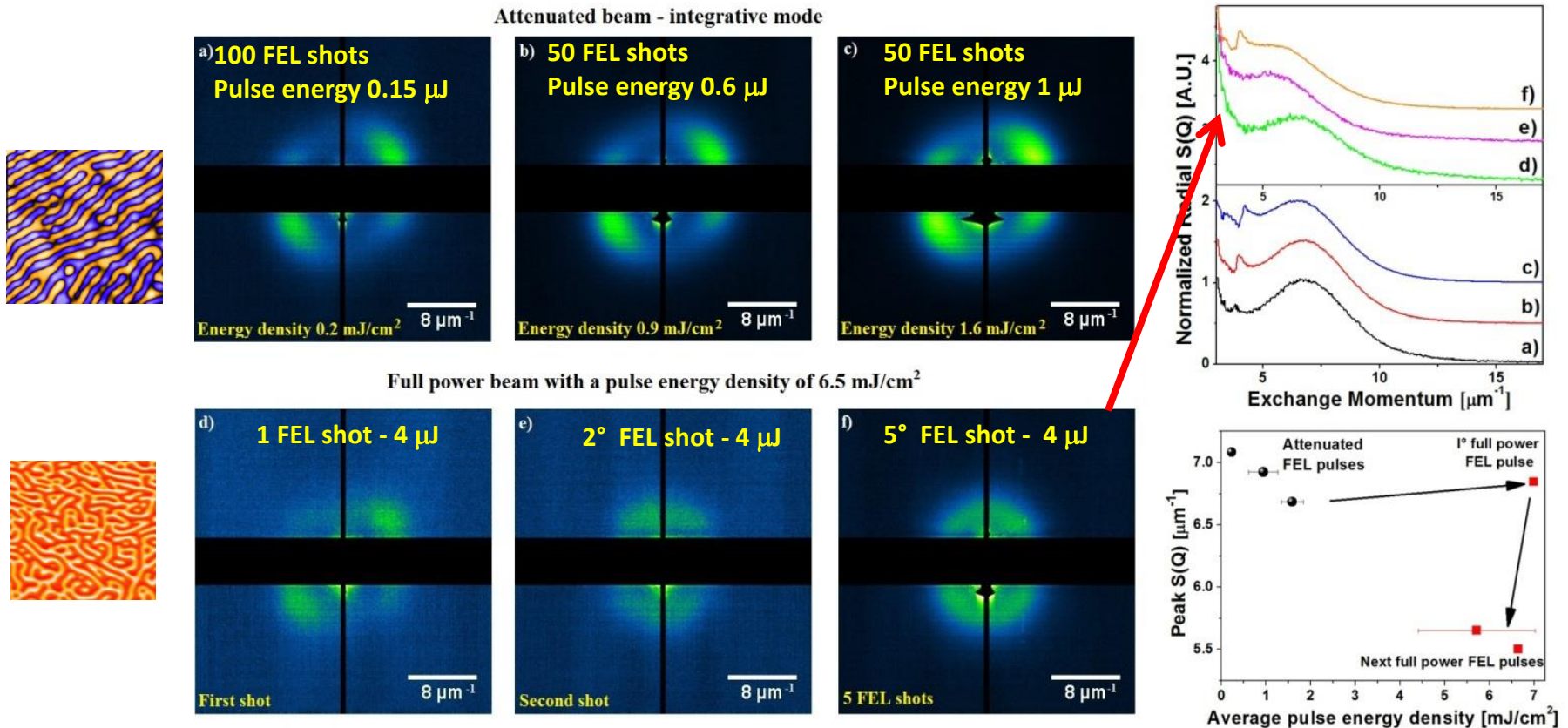


The smearing of the absorption edge indicates an ultrafast rearrangement of the electron population around the Fermi level, driven by the sudden electron temperature change. The thermalization of conduction band electrons occurs within ~ 70 fs pulse duration, time shorter than phonon-phonon scattering.

For E_{photon} above Al 2p edge, the temperature of the electron sub-system is estimated to be ~ 0.5 eV, well above the Al melting point ~ 0.05 eV.

Magnetic domains exposed to a very high power short fs pulses

Reorganization from aligned to labyrinth domain structure & change in the average domain period with increasing pulse power:
? is 'damage' threshold for dynamic magnetic studies.

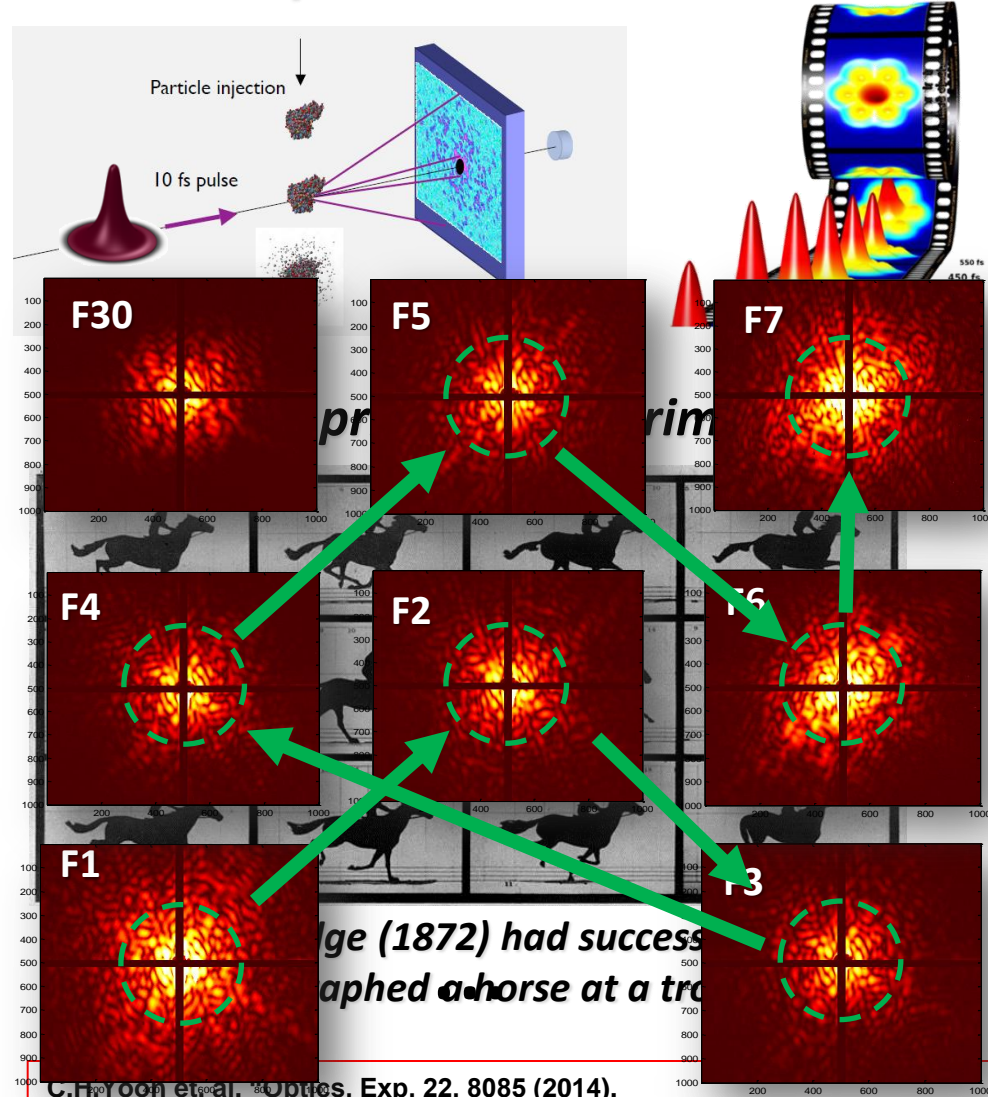




Towards fs-movie with single shot CDI

Elettra Sincrotrone Trieste

Microscopists dream



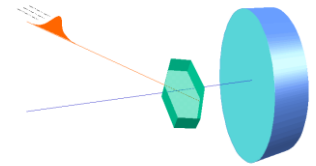
Challenging computational task:

- a) Collection of a speckle pattern not a real space image. Phase retrieval algorithm.
- b) Reconstruction algorithm has to catalog orientation and "recognize" the frames temporal evolution.

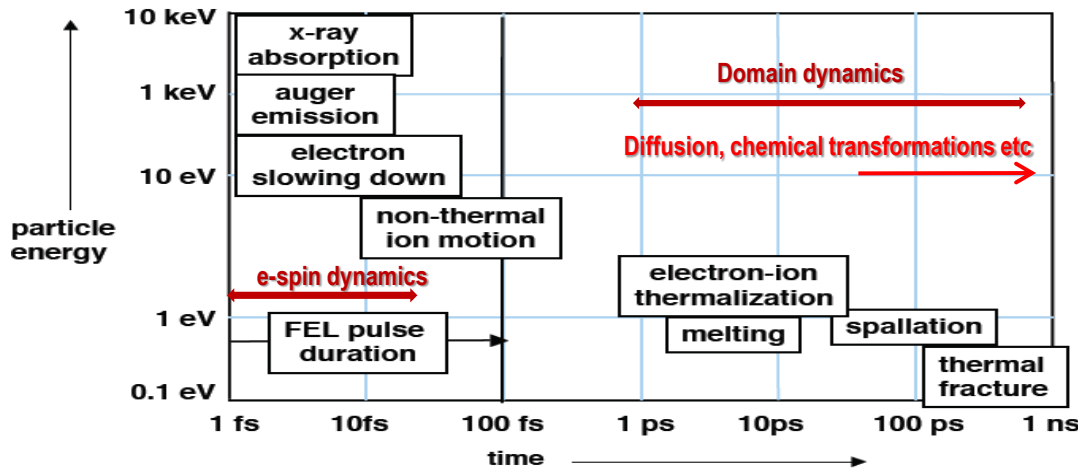
Using phase retrieval algorithm:
galloping horse movie can be
reconstructed



C.H. Roen et al. Optics. Exp. 22, 8085 (2014).



Energy-time picture of x-ray material interaction



- ✓ Temporal resolution of the sample response is determined by the duration of the probe.
- ✓ For larger particles CDI spatial resolution becomes depended on longitudinal coherence and SEEDING is highly desirable.

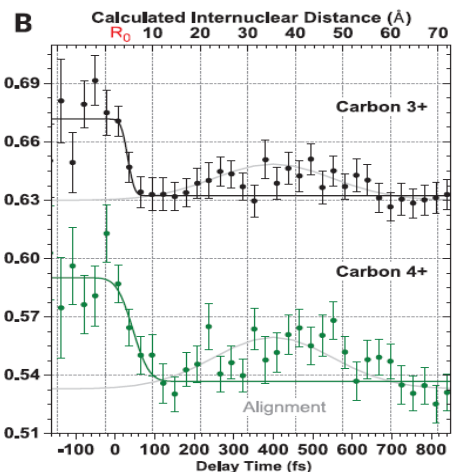
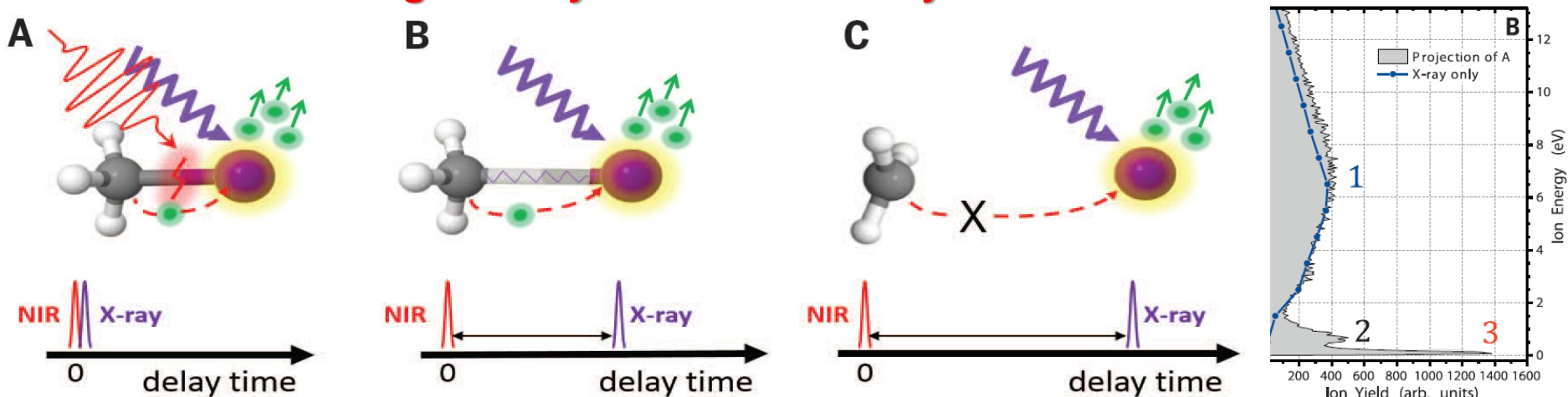
1. IR-UV pump/X-ray probe: X-ray pulse duration determines temporal resolution: – dynamic study of transient phenomena – fluctuating and fast evolving systems, structural dynamics on nanoscale resulting from chemical or physical changes induced by IR and UV laser.
2. Ultrafast FEL pulse as a pump: X-ray or IR probe: a unique way of depositing energy into materials and to create states of strong electronic excitation, high temperature and pressure.

Tunability allows working above and below resonances, probing the selectively the effect of excitation on the target constituents.

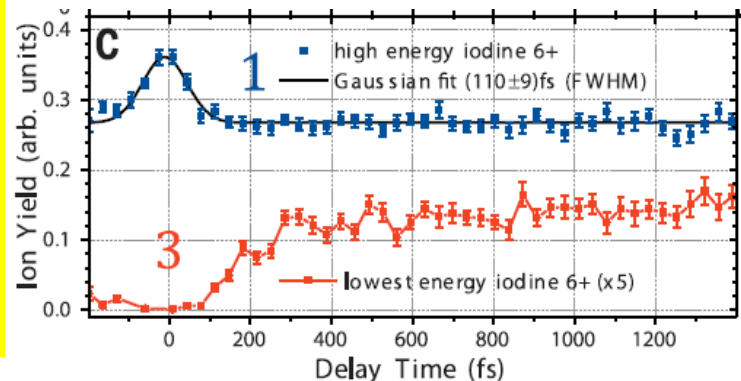
Stroboscopic schemes for time resolved FEL experiments: two color IR pump/FEL probe

IR-induced dissociation of CH_3I following the energy of emitted I ions

Localized charge on I atom, created by X-rays may transfer to the methyl group via Auger decay: event affected by the I-C distance

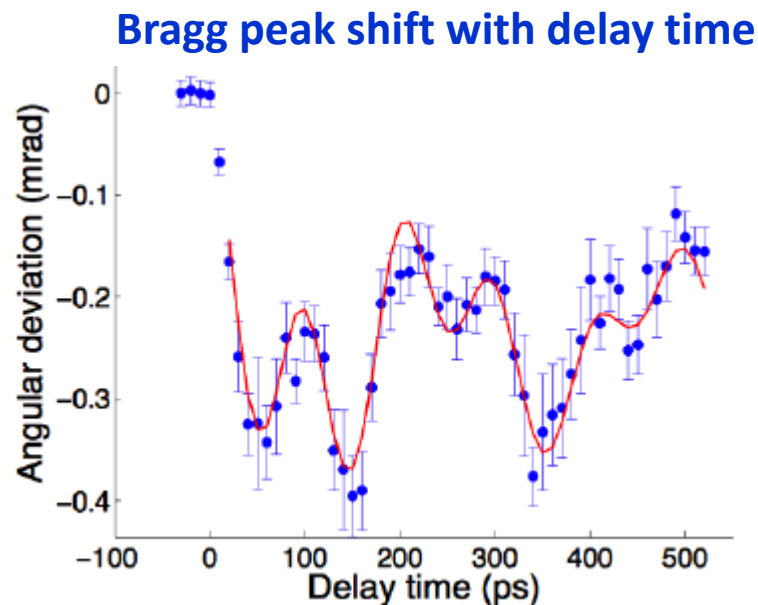
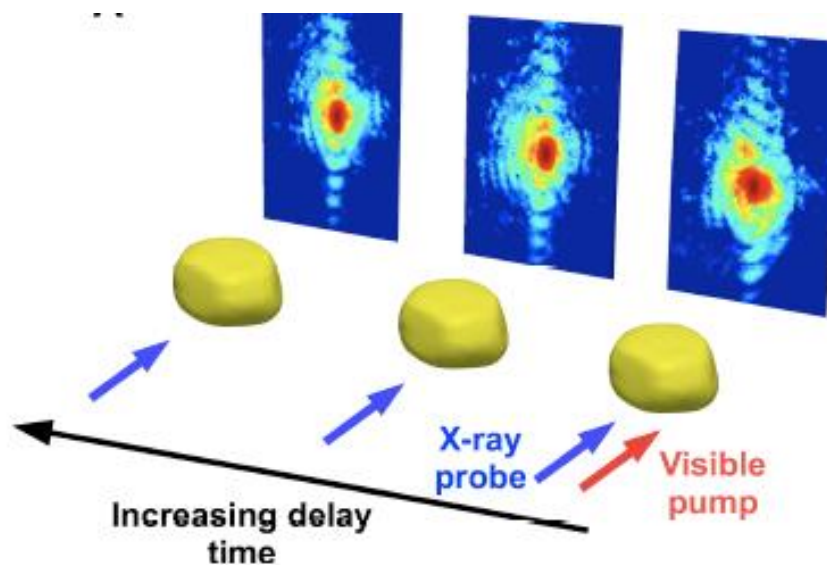


C ions mainly acquire their charge via electron transfer to initially ionized iodine. After ~ 100 fs the molecule the charge becomes determined by FEL interaction with CH_3 and I



Stroboscopic schemes for time resolved FEL experiments: two color IR pump/FEL probe

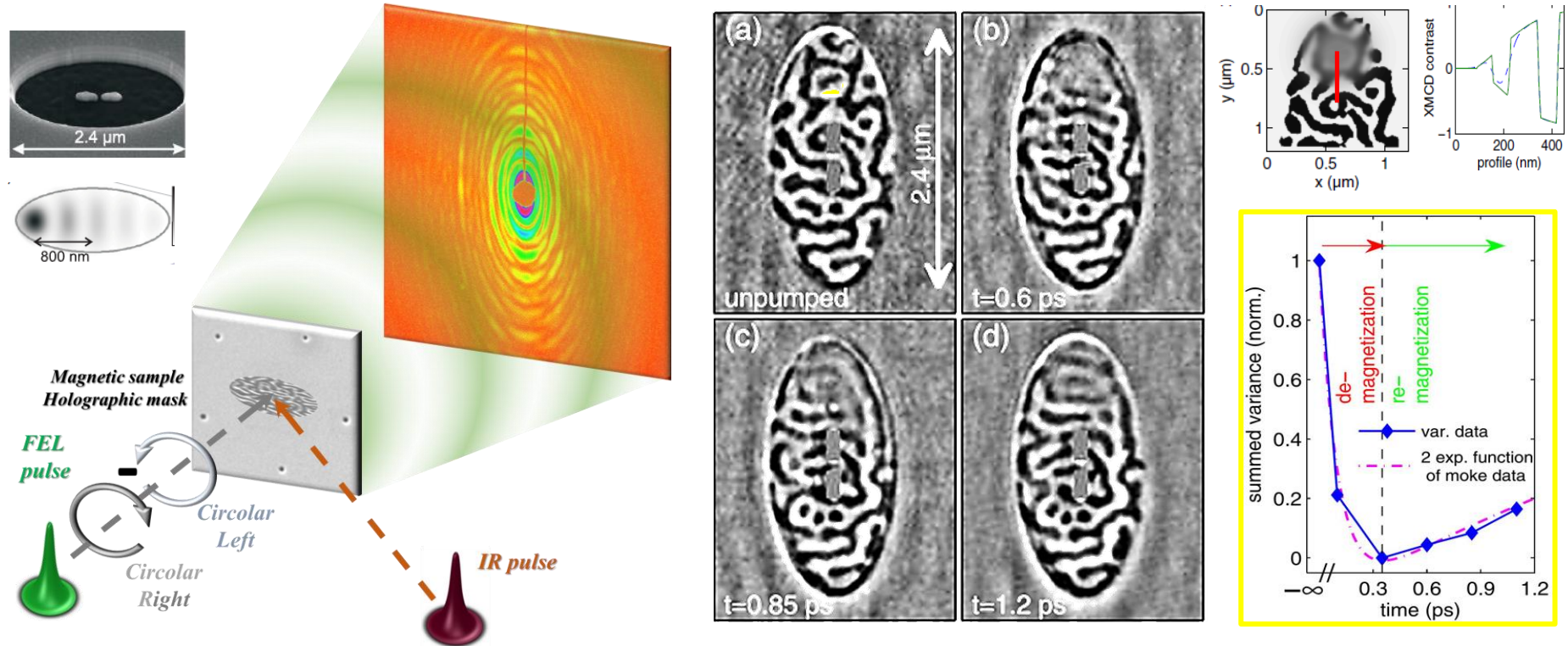
Shedding light on lattice dynamics in individual gold nanocrystals via coherent diffraction



The evolution of the coherent acoustic phonons within the nanocrystal through the Bragg peak shift: can be modelled as a harmonic oscillator with two modes.

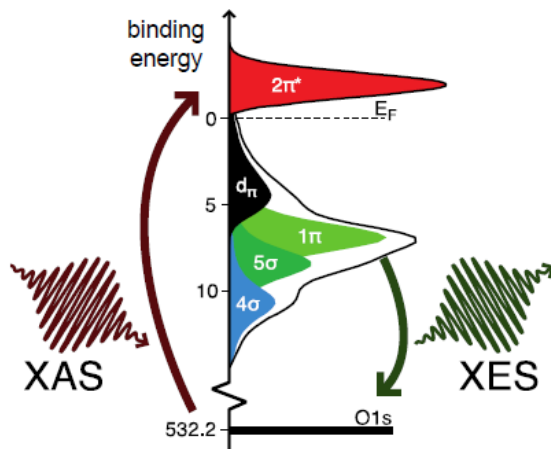
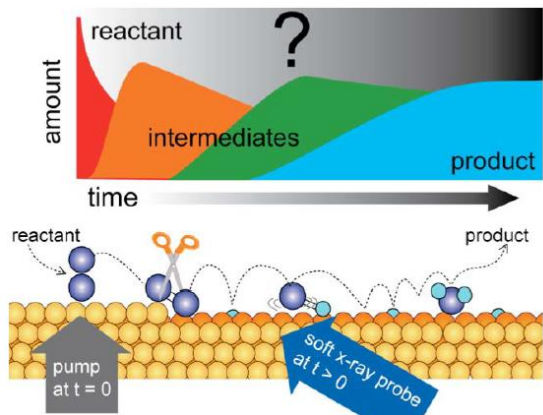
J. Clark, *Science* 341, 56–59 (2013)

Time resolved resonant magnetic holography with sub-100 nm spatial and 100-fs temporal resolution

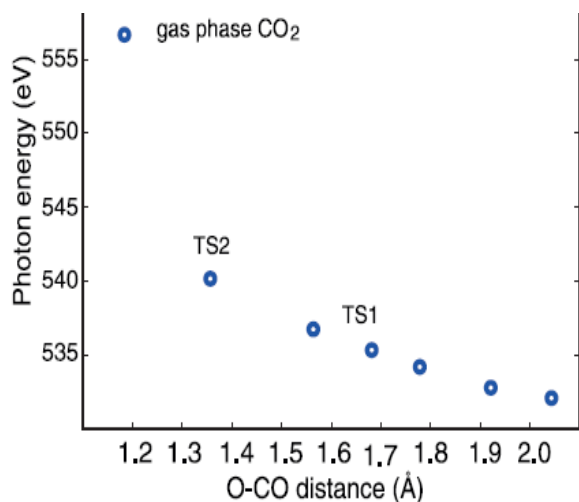


Ultrafast de-magnetization within 250 fs, spatially localized via a tailored micro-resonator. Point out the important role of ultrafast spin-electron transport.

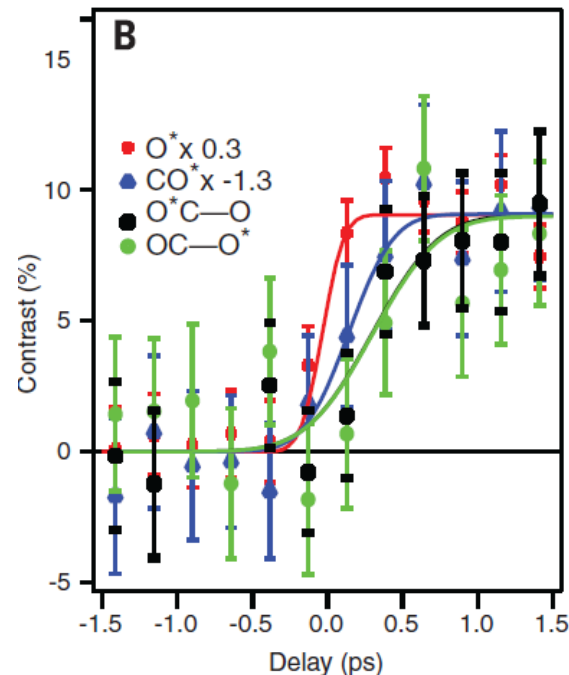
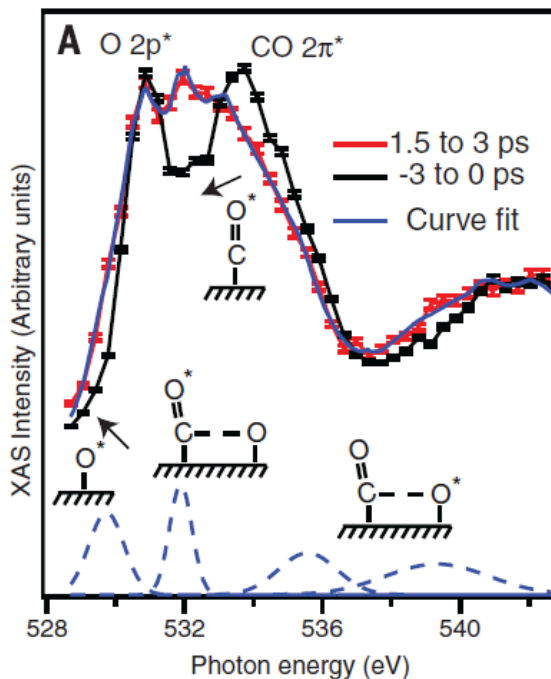
IR / FEL: Laser-induced surface reactions: map transient changes of electronic structure by trRIXS (x-ray emission/absorption spectroscopy - XES/XAS)



O becomes activated on a time scale below 300 fs, whereas CO is activated on a 500-fs time scale and beyond the transient states 1 and 2 are formed leading to CO₂

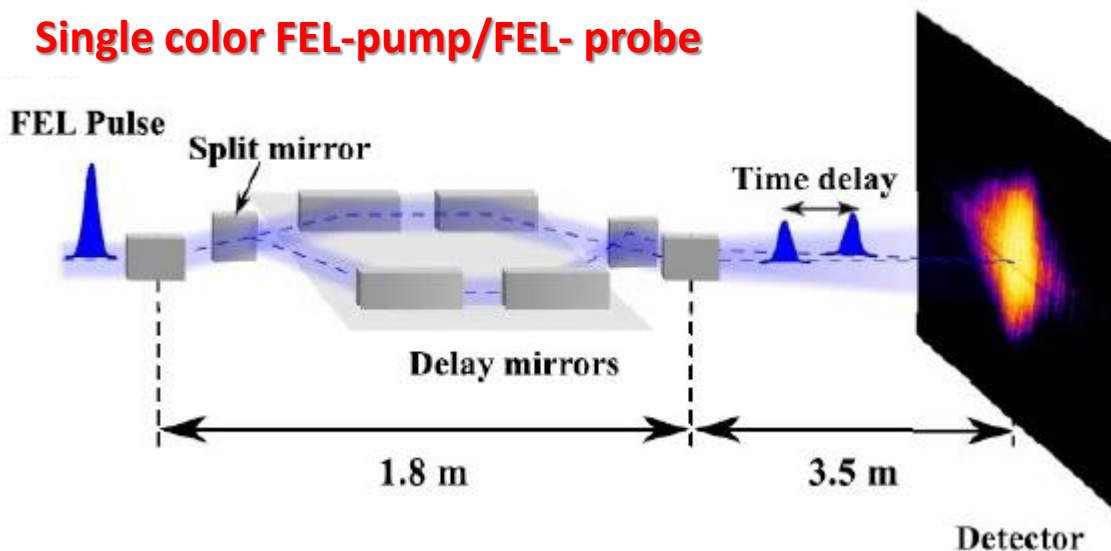


H. Öström et al. Science 347, 978 (2015)

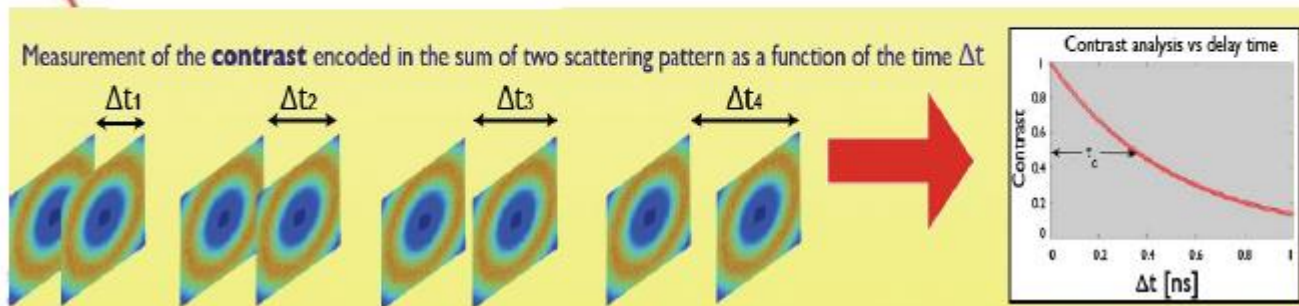


Stroboscopic schemes for FEL/FEL experiments: **single color FEL-pump/probe**

Single color FEL-pump/FEL- probe

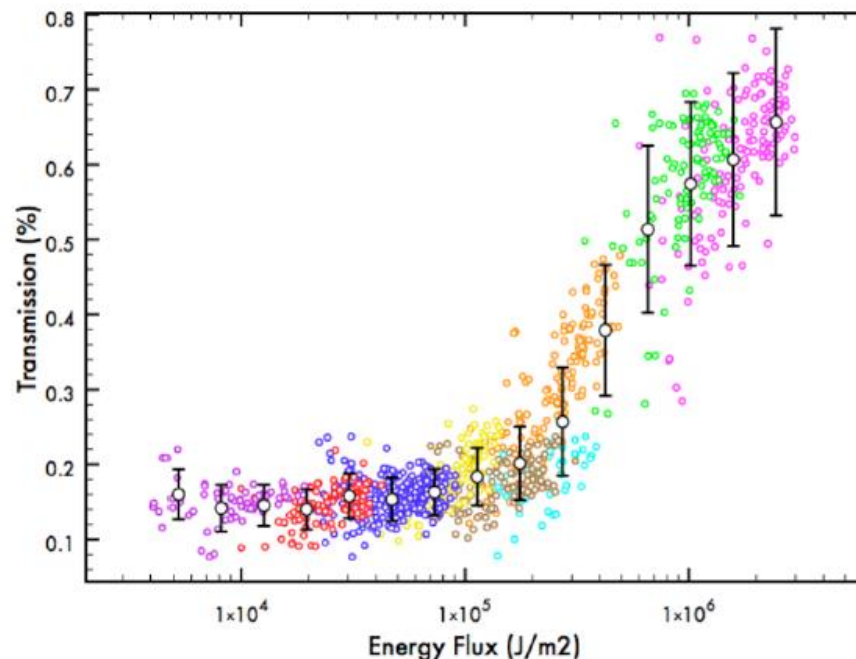
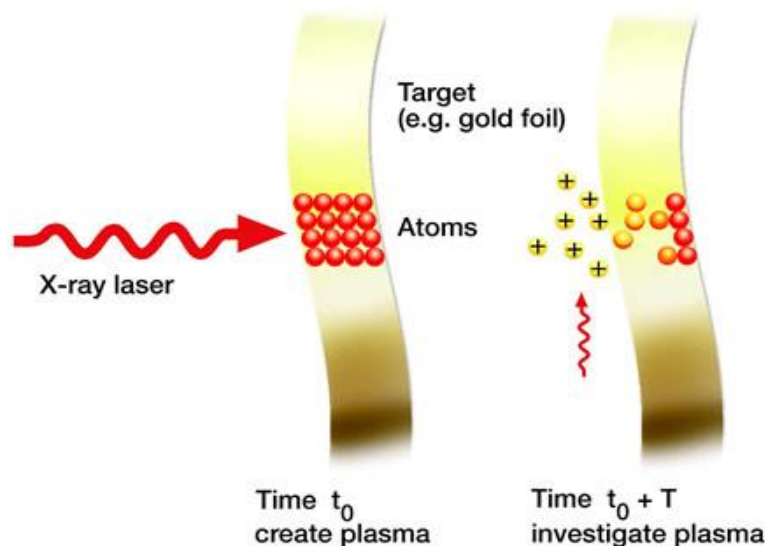


X-ray Photon Correlation Spectroscopy (XPCS)



Producing plasma – highly ionized state of matter and monitoring its evolution via **FEL pump-probe**

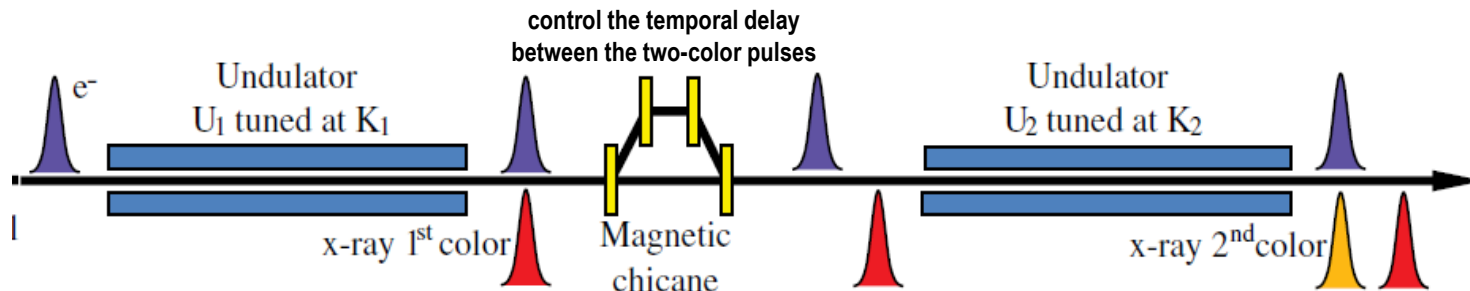
With an X-ray laser, plasmas can be created that are as hot as the interiors of giant stars. At the same time, it will be possible to investigate the status of created plasmas at varying intervals with another part of the laser beam and thus to conduct research into the plasma state.



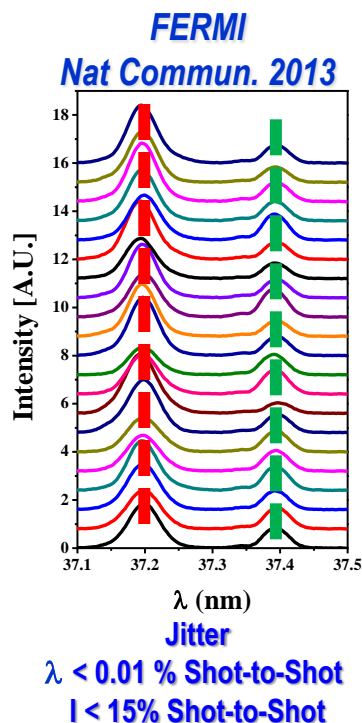
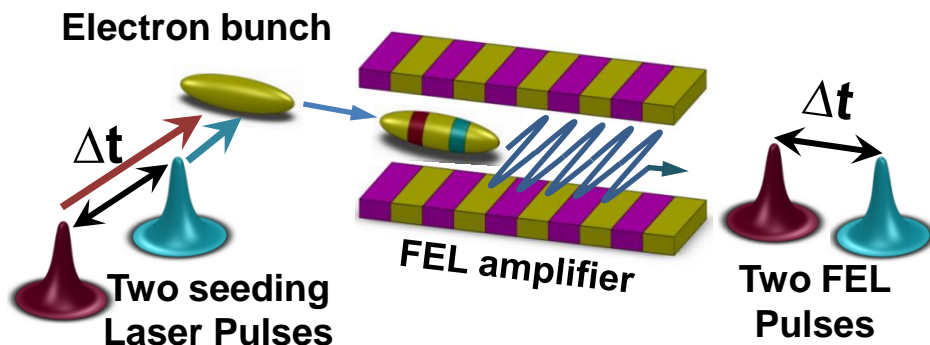
Al foil irradiated with high energy 92 eV FEL photons becomes transparent because both electrons from the 2p state are ejected and no more photoionization of electrons is possible – blue shift in the L edge

Generation of 2-color FEL pulses separated in time

SASE: dividing the radiator in two detuned sections



FERMI: using the same electron bunch via two independent seed pulses two FEL pulses (λ_1 and λ_2) are generated



LCLS SASE
PRL 110, 134801

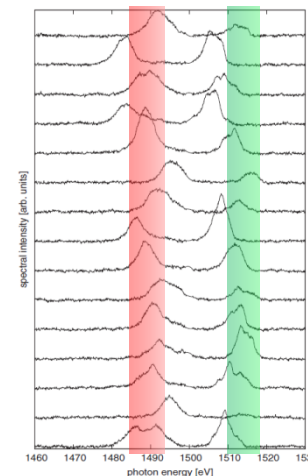


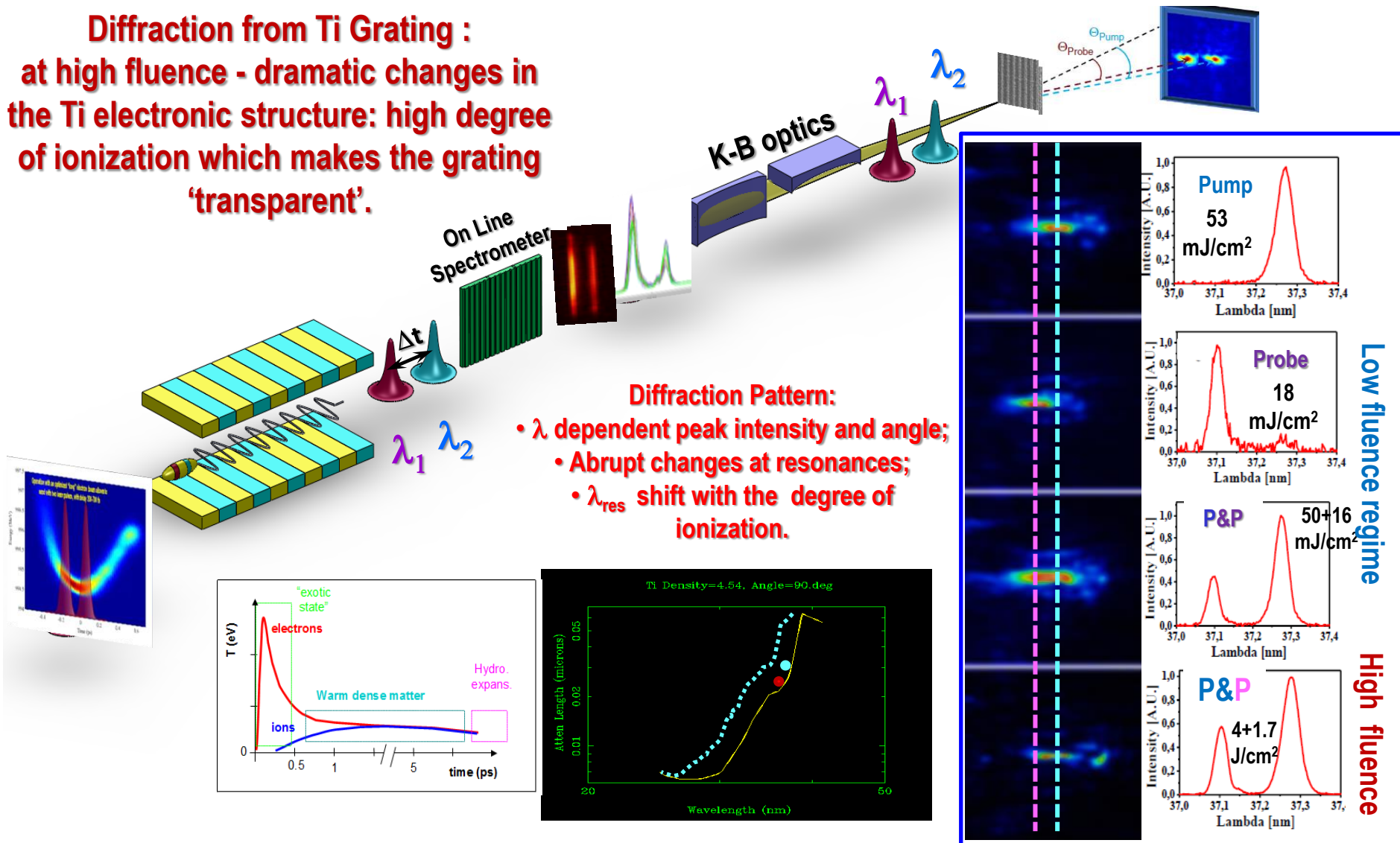
FIG. 2. Fifteen consecutive x-ray spectra produced under scheme 1 with chicane delay set to the nominal 0 fs.

Jitter
 $\lambda < 0,5\%$ Shot-to-Shot
 $I < 40\%$ Shot-to-Shot

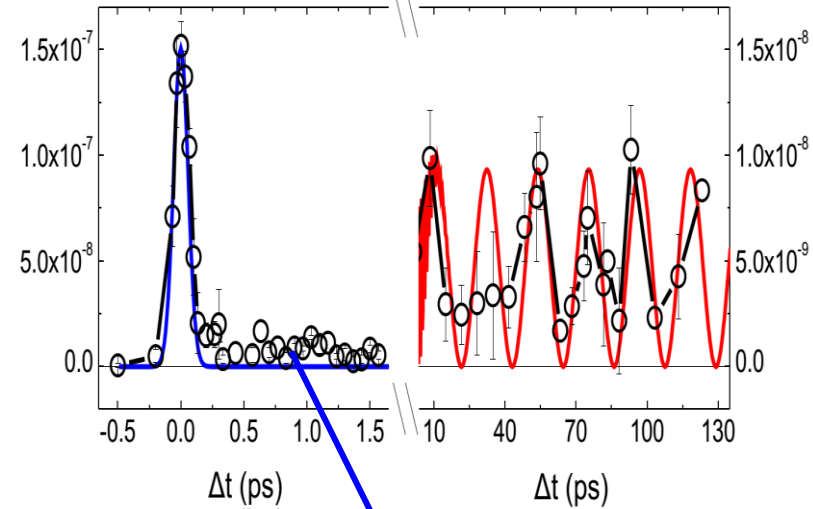
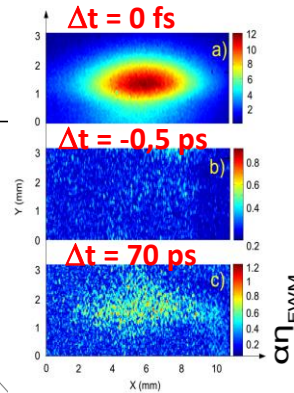
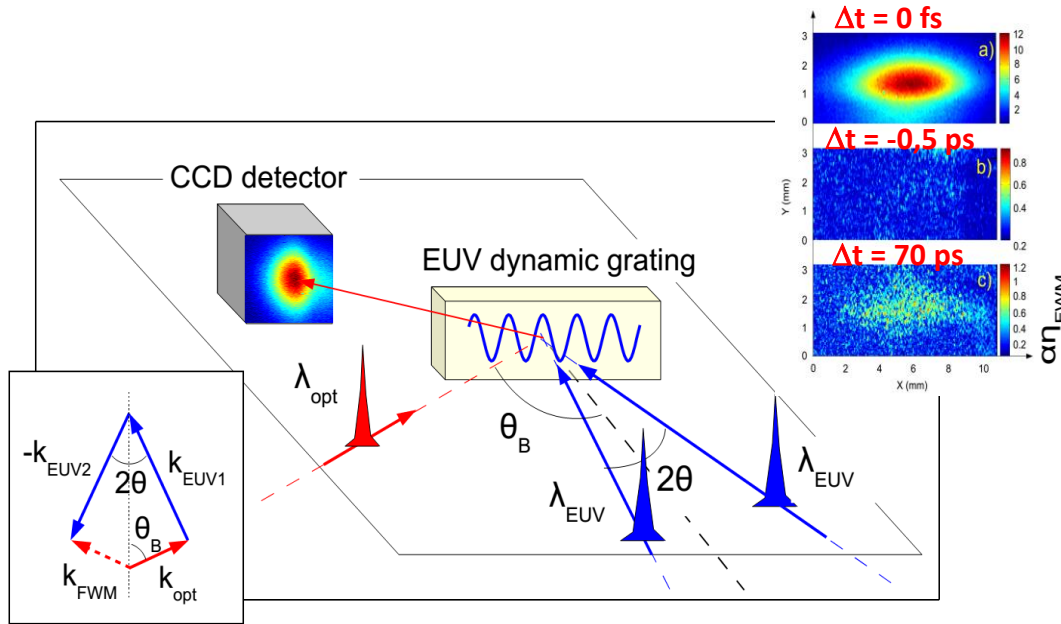
- $\Delta\lambda$ (0.5%) limited by undulator bandwidth;
- Δt -0.2 – 0.8 ps (bunch compression),
- $I_{\lambda_1}/I_{\lambda_2}$ variable.

Diffraction from Ti grating using two color FEL pulses

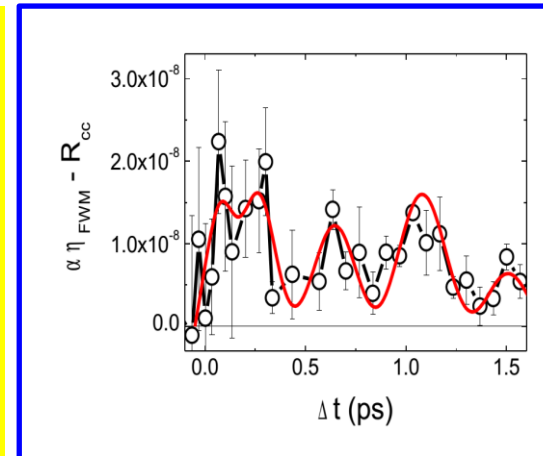
Diffraction from Ti Grating :
at high fluence - dramatic changes in the Ti electronic structure: high degree of ionization which makes the grating 'transparent'.



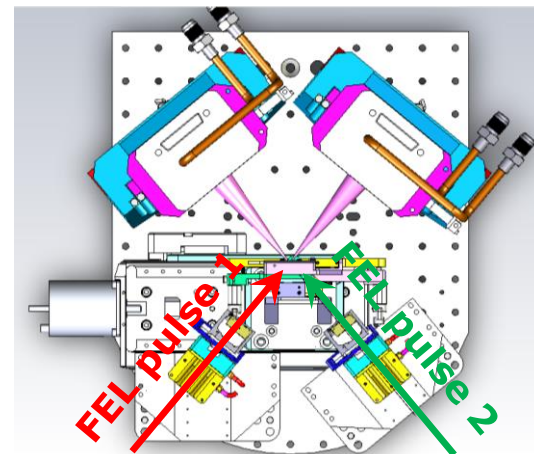
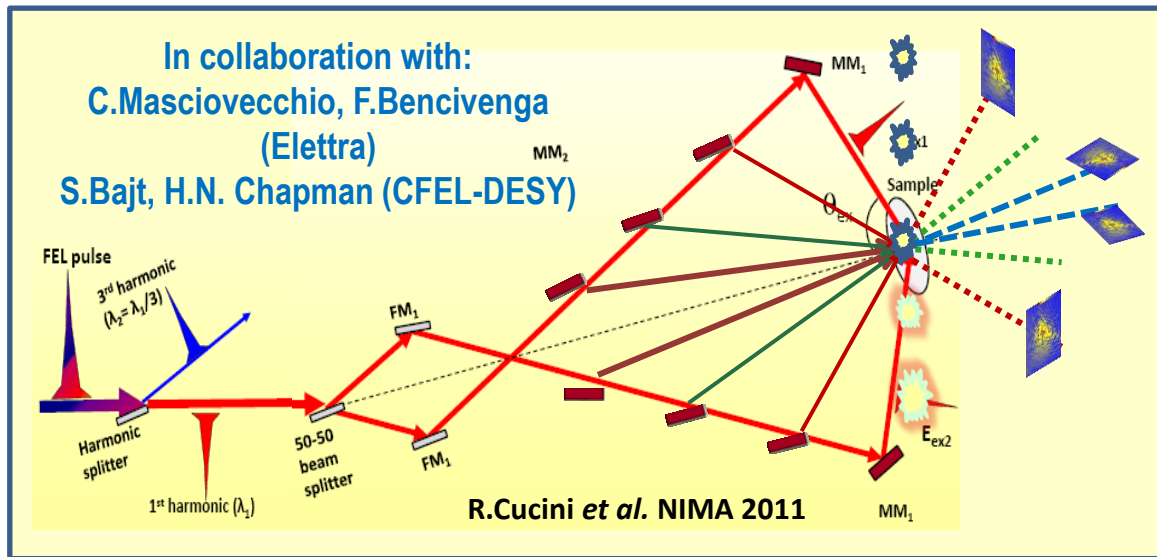
Four wave mixing: generation of VUV transient grating in the SiO₂ using two coincident FEL pulses



When the three beams arrive on the sample at the same time ($\Delta t = 0$) a FWM signal is recorded, showing the occurrence of the wave mixing process. With time delay of the optical pulse, intensity modulation of the scattered signal are observed compatible with the excitation of Raman modes ($\Delta t < 1.5$ ps) and longitudinal acoustic modes ($\Delta t > 10$ ps).



Future developments (stereo/strobo-CDI)



Possible experiments

FEL 1st beam
FEL 2nd pulse

1.5 μm
 0.5 μm
 1.5 μm

Different view angles reveal different features, easier to reconstruct and perform a stereo-CDI

1st FEL
Delayed FEL

Co

Res-CDI,
 Strobo FEL-FEL P&P
 experiment recorded by
 different CCD.
 Strobo-CDI

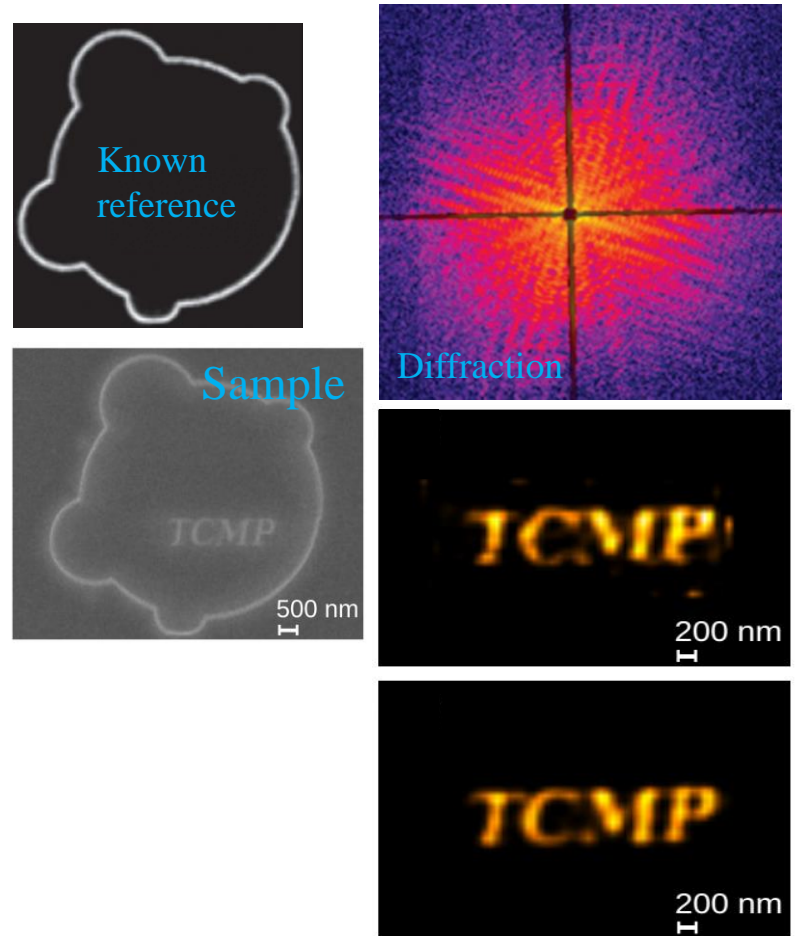
Extended reference holography

FTH ideal case is to optimize the experimental geometry without restriction due to the reference wave, permitting to optimize signal-to-noise and resolution.

Seed FEL pulse has an high degree of transversal and longitudinal coherence.

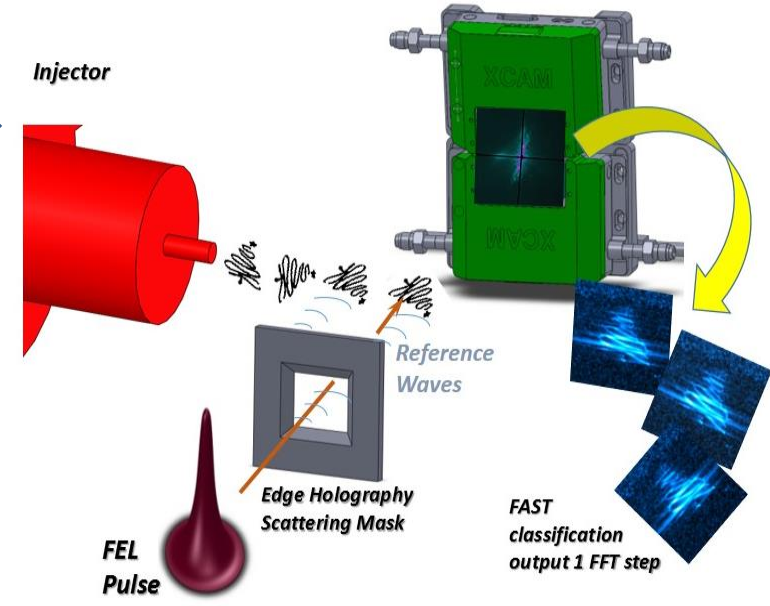
- 1. Transversal coherence is the key element in CDI.*
- 2. Longitudinal coherence is important in holography when mask is decoupled from the sample.*

Combining Extended Reference Holography with particle injector



Hologram obtained solving iteratively
 $A_{data} - A_{ref} = C_{ref,obj}$
 where $C_{ref,obj} = \Psi_{ref} * \Psi_{obj}$

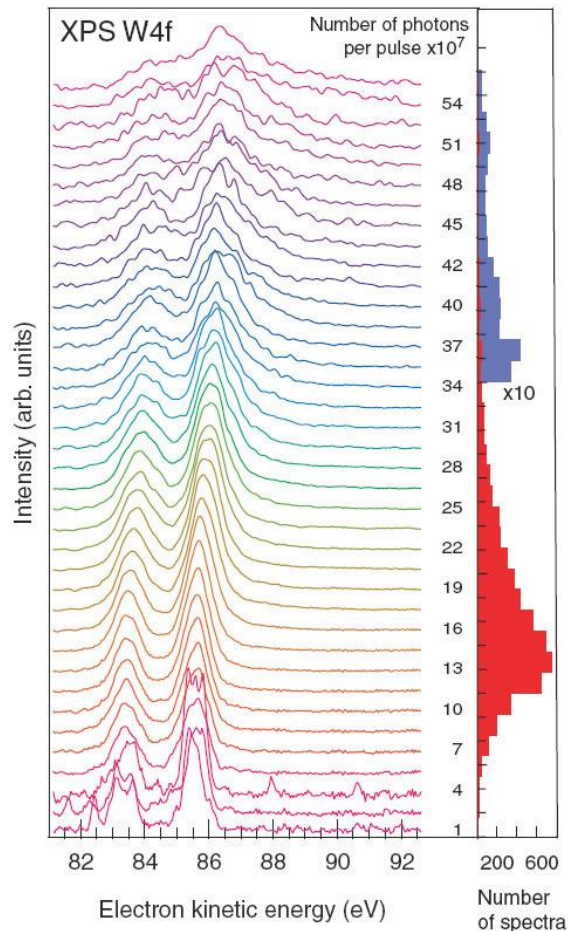
Hologram refined with RAAR phase retrieval



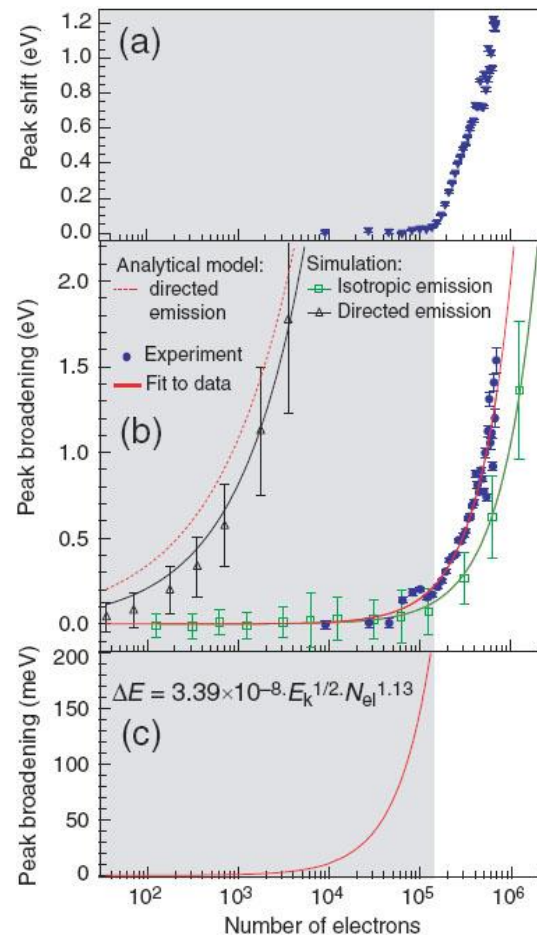
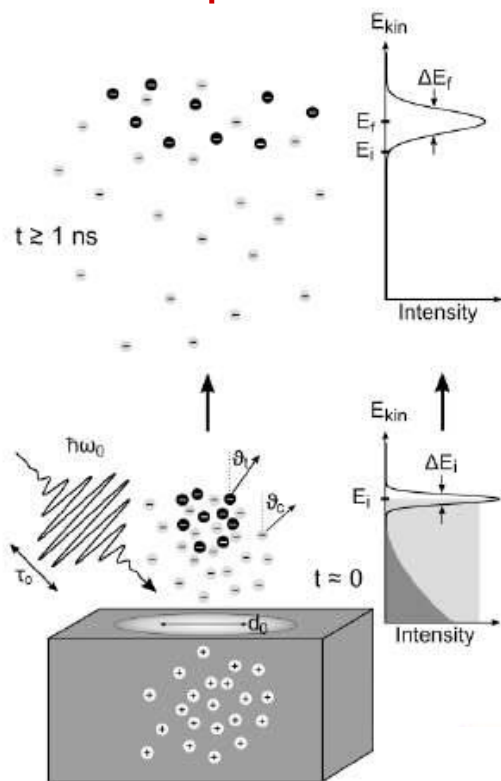
A.V. Martin. et al. "X-ray holography with customizable reference" Nature Comm. 4, 2476 (2014).

PES with FELs???

Core-level PE was proven to be extremely useful tool for time-resolved studies but FELs are too bright!.....



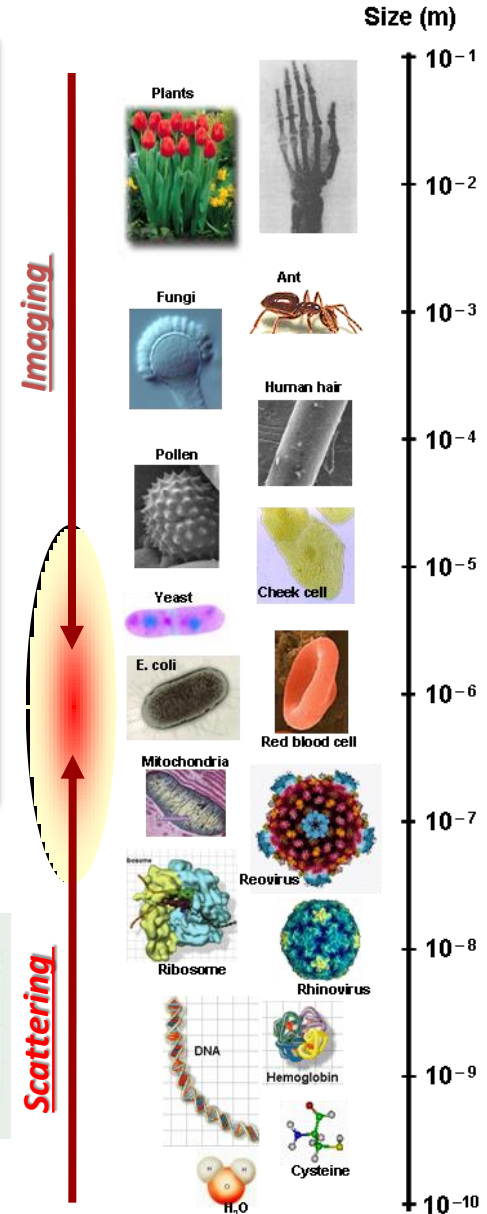
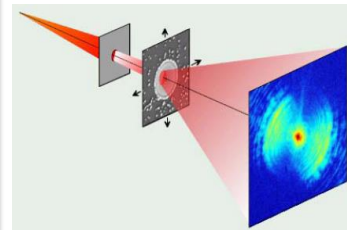
space charge (1mm spot)
 $> 10^8$ phot/pulse
 $> 10^4$ el/pulse



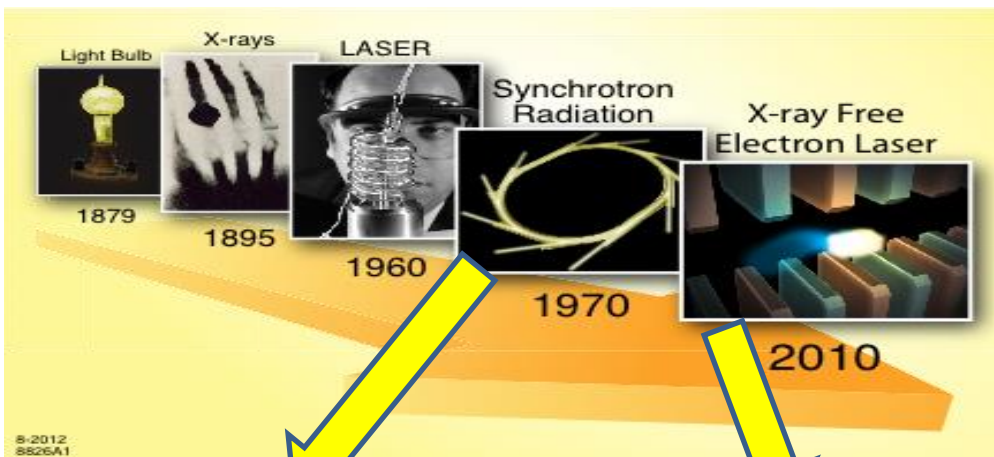
Momentum-Time resolved resonant inelastic x-ray scattering with high spectral resolution is feasible and complementary.

- Scanning microscopes monitoring electrons - limited to surfaces.
- Transmission electron microscopes can resolve even atoms but are limited in penetration (samples thinner than ~ 30 nm).
- X-ray crystallography reveals the globally averaged 3D atomic structures based on the diffraction phenomenon, but requires crystals.
- Classical x-ray microscopy – limited in resolution and focal depth by the optical elements. Temporal resolution - \geq ns

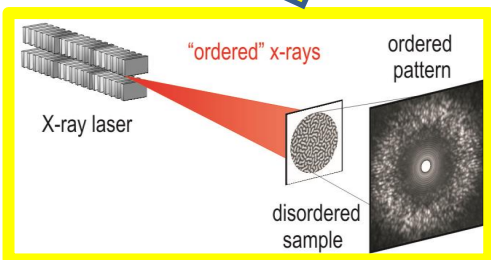
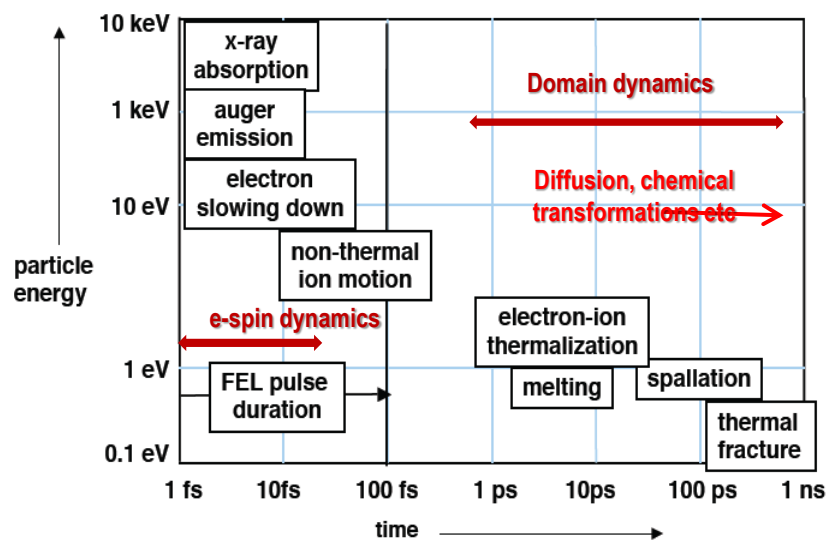
The optics depth and resolution limitations can be overcome by image reconstruction from measured coherent X ray scattering pattern visualizing the electron density of non-crystalline sample.



X-ray sources complementary used in material science: from static to dynamics



Energy-time picture of x-ray material interaction



- ❖ Micro-nanoscale order
- ❖ Equilibrium States
- ❖ Slower processes > ps

new paradigm →

- Nanoscale order
- Fast dynamics < ps
- Excited transient states