Free Electron Lasers
&
ultrafast x-ray science

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European XFEL

XVII School on Synchrotron Radiation: Fundamentals, Methods and Applications
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Outline

Different types of lightsources

Storage rings (SR)

- High perf. x-ray source
- Very stable; highly efficient
- Many user installations for large variety of applications

→ X-ray scattering, microscopy, spectroscopy

X-ray FEL radiation

- Peak brightness x-ray source
- Single-pass; few sources
- So far only few user installa-tions; applications u. study

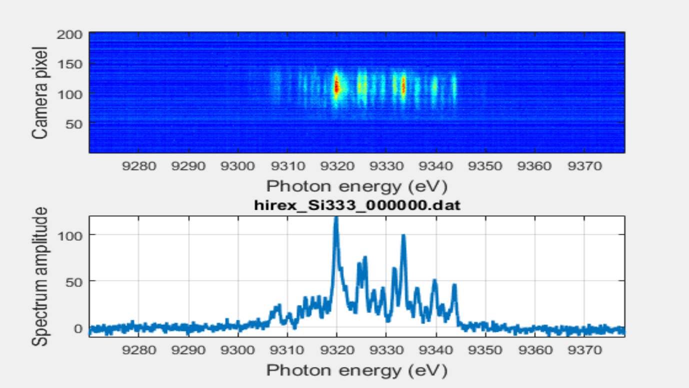
→ X-ray ultrafast methods using scattering, microscopy, spec-troscopy; non-linear methods

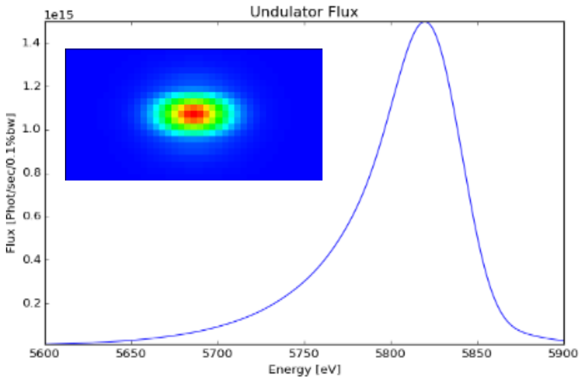
Visible Laser radiation

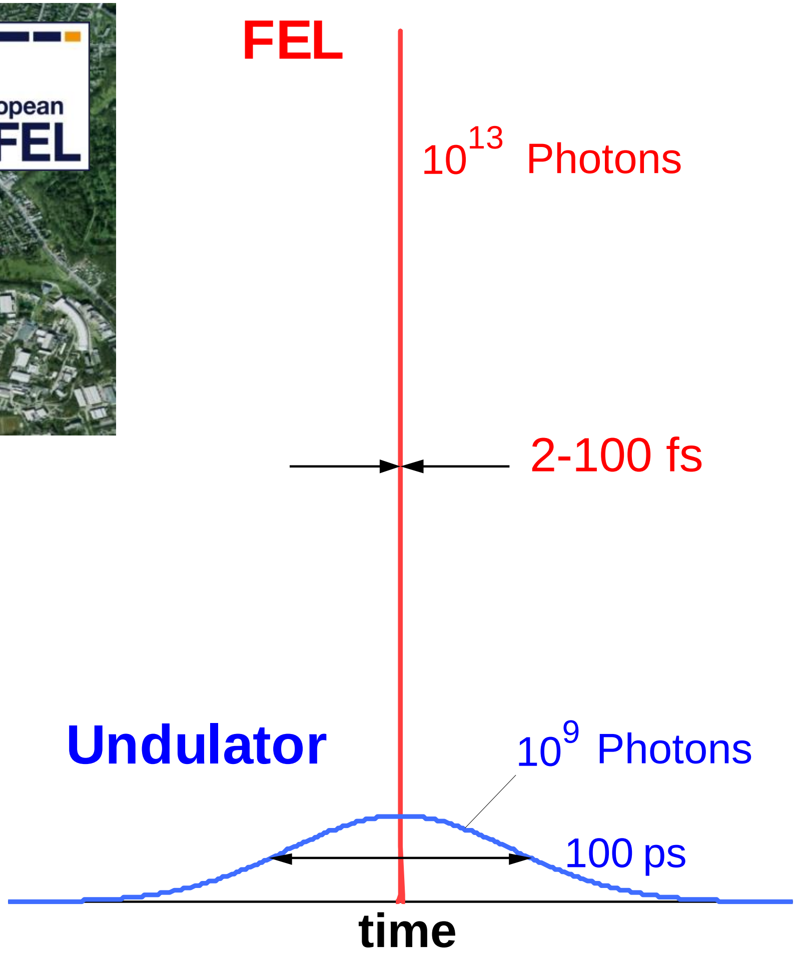
- Commercially or home-built systems widely distributed
- Huge community
- Attosecond & non-linear techn.

→ Light ultrafast and non-linear methods using microscopy or spectroscopy

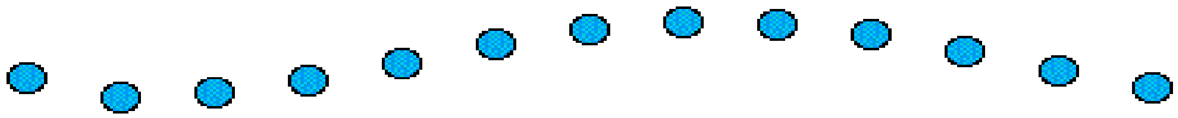
XFEL radiation is very different from synchrotron radiation







- fs pulse: a probe of atomic motion (100fs) …. and charge migration (fs)
- hard X-rays: atomic resolution, chemical selectivity, bulk sensitivity



Acoustic vibration

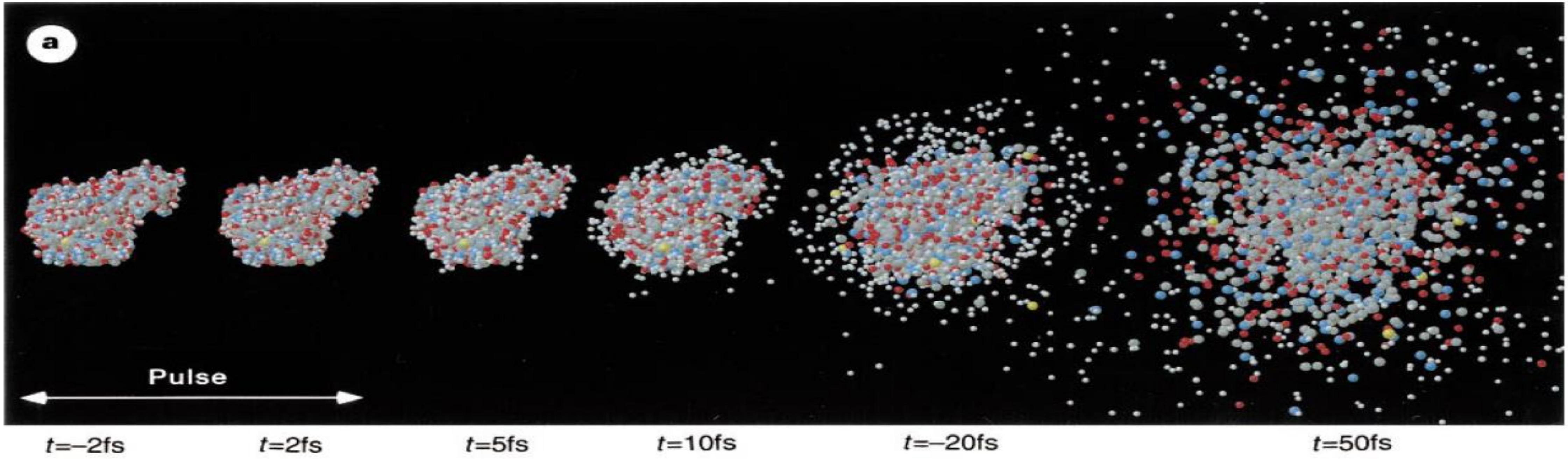
Speed of sound: $v = 3000\, m/s\ ,\ a = 3Å \ \Rightarrow \delta t \leq 10^{-13} s = 100\ fs$

- Can we measure phonon dynamics in the time domain ?
- Can we see how charge migrates from one atom to another ?

Diffract before destruction: a totally new approach to structural determination with X-rays

Free Electron Lasers : The exploding protein

R. Neutze et al, Nature 406, 752 (2000)



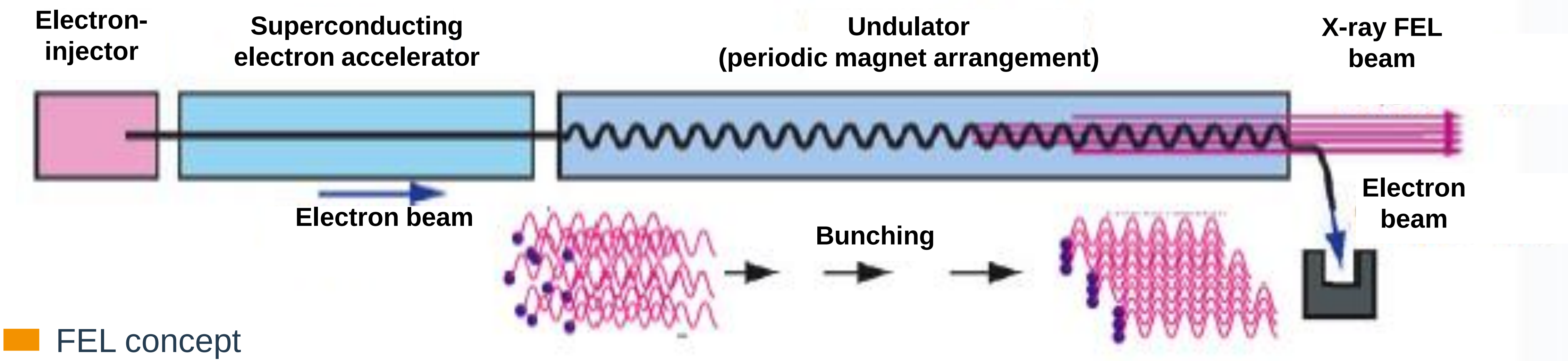
- Can we beat radiation damage ?
- Can we measure at room temperature ?
- Can we make movies of proteins at work ?

Outline

- Introduction
- **Free-electron laser basics**
- X-ray FEL facilities

  – break –
- Ultra fast processes in chemistry and materials
- Biological and soft matter
- Extreme states of matter
- Future

X-ray Free-Electron Laser (X-ray FEL)



- FEL concept
  - J.M.J. Madey, J. Appl. Phys. 42, 1906(1971)
- SASE FEL radiation
  - **High Gain Single Pass regime:** A.M. Kondra-tenko, E.L. Saldin, Part. Accel. 10, 207 (1980)
  - **Collective instability and self-organization**: R. Bonifacio, C. Pellegrini, L.M. Narducci, Opt. Communications 50, 373 (1984)

- Properties of SASE x-ray FEL radiation
  - **Ultrashort duration (fs – 200 fs)**
  - **Very high pulse energies (0.1 – 1 mJ)**
    - ▶ 10^{12} (10^{13}) photons at 0.1 (1) nm
  - **Almost full transverse coherence (<10 keV)**
  - **Limited longitudinal coherence**
  - **Single pass generation / fluctuations**

Differences with respect to undulator/synchrotron radiation

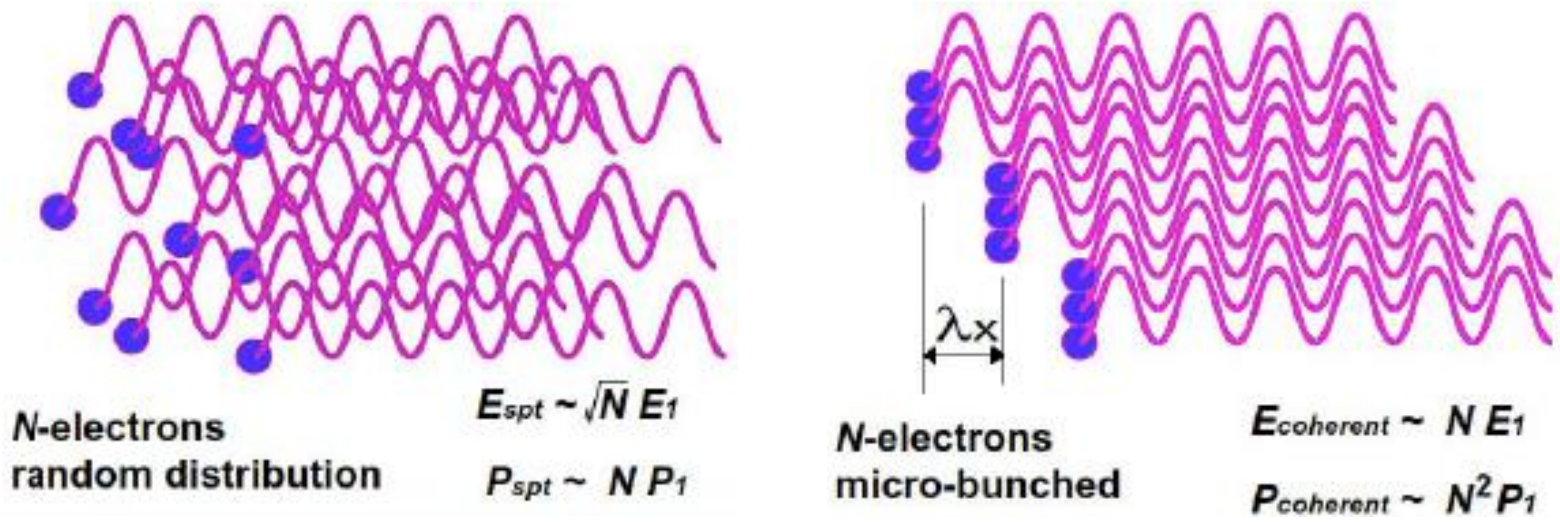
- Resonance condition is equal

$$\lambda = \frac{\lambda_u}{2\gamma^2}\left(1 + \frac{K^2}{2}\right) \quad K = 0.934\lambda_u B_0$$

- Much tighter electron phase space parameters
  - **Extremely small emittance**
  - **Bunch compression**

Parameter	SR	FEL	factor
Transverse emittance	1 nm (x) (→ 0.1(x,y))	0.015 nm (x,y)	40 (7)
Longitudinal e$^-$ density	10 nC / 30 mm	1 nC / 30 µm	100

- Coherent emission process



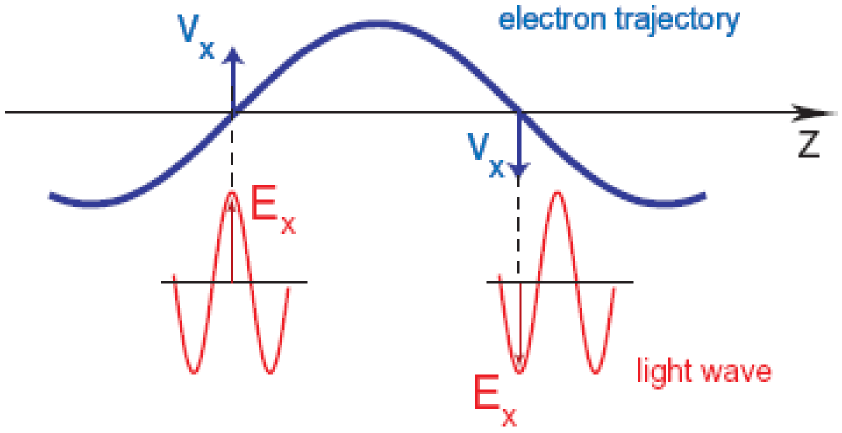
Bunching the electrons

- By electro-magnetic forces arising from interaction with the co-propagating radiation field
  - **Energy change due to x-ray field:**

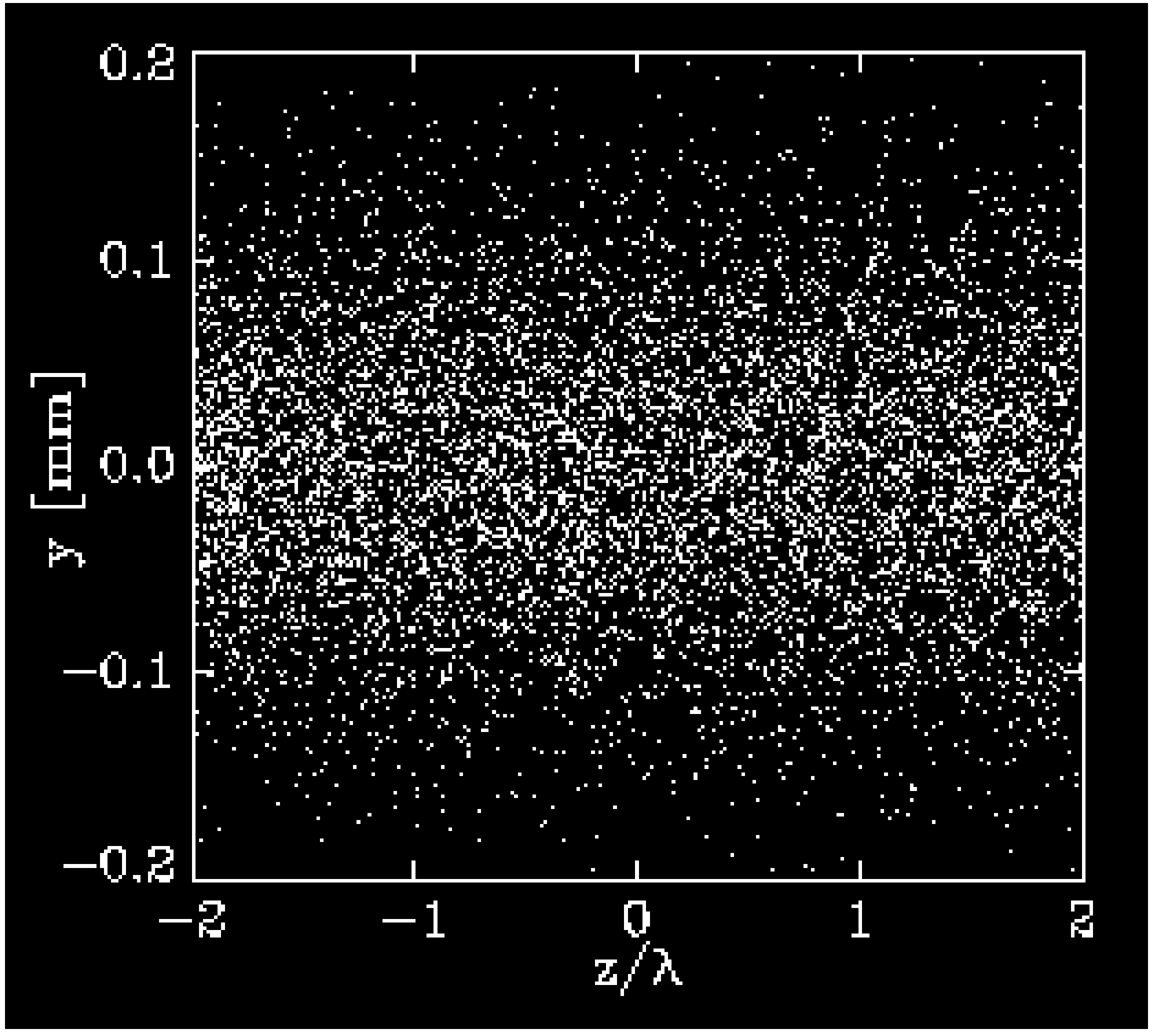
$$dE = -e\, v_x(t) \times E_x(t)\, dt$$

  - **SASE resonance condition:**

Keep phase between $v_x(t)$ and $E_x(t)$ const.



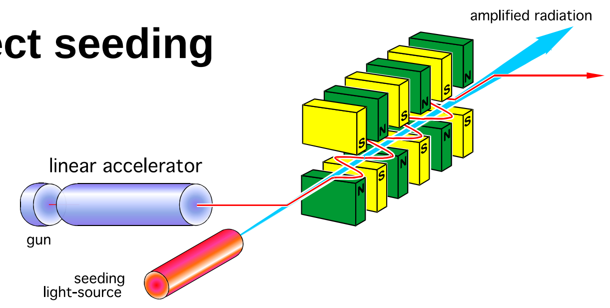
GENESIS -
simulation for TTF parameters
Courtesy - Sven Reiche (DESY, now PSI)



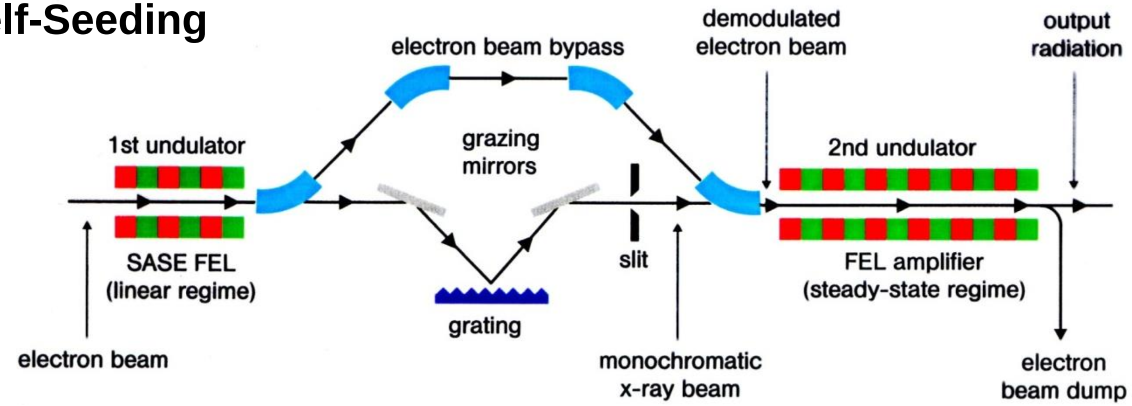
Seeded FEL radiation

- Manipulate electron bunch using seeding pulse
  - **improve start condition compared to noise (SASE case)**
  - **improved radiation properties**
    - temporal, spectrum, coherence, fluctuations
  - **shorter gain length**
  - **typically needs more setup time**
- Various methods (ħω dependent)
- Note: in general easier for smaller ħω

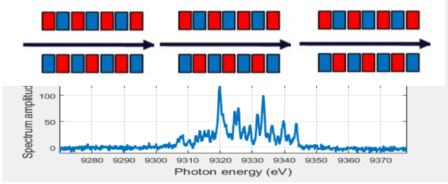
HHG direct seeding



Self-Seeding

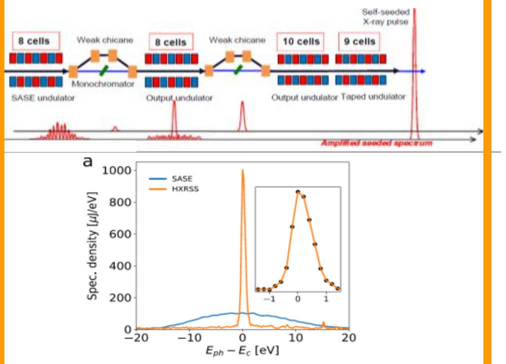


SELF AMPLIFIED SPONTANEOUS EMISSION (SASE)



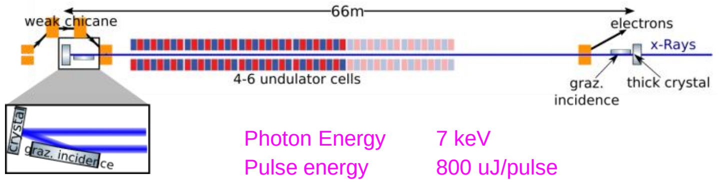
Photon Energy	9 keV
Pulse energy	2.2 mJ/pulse
BW (FWHM)	20 eV
Spectral density	**100 uJ/eV**

HARD X-RAY SELF SEEDING (HXRSS)

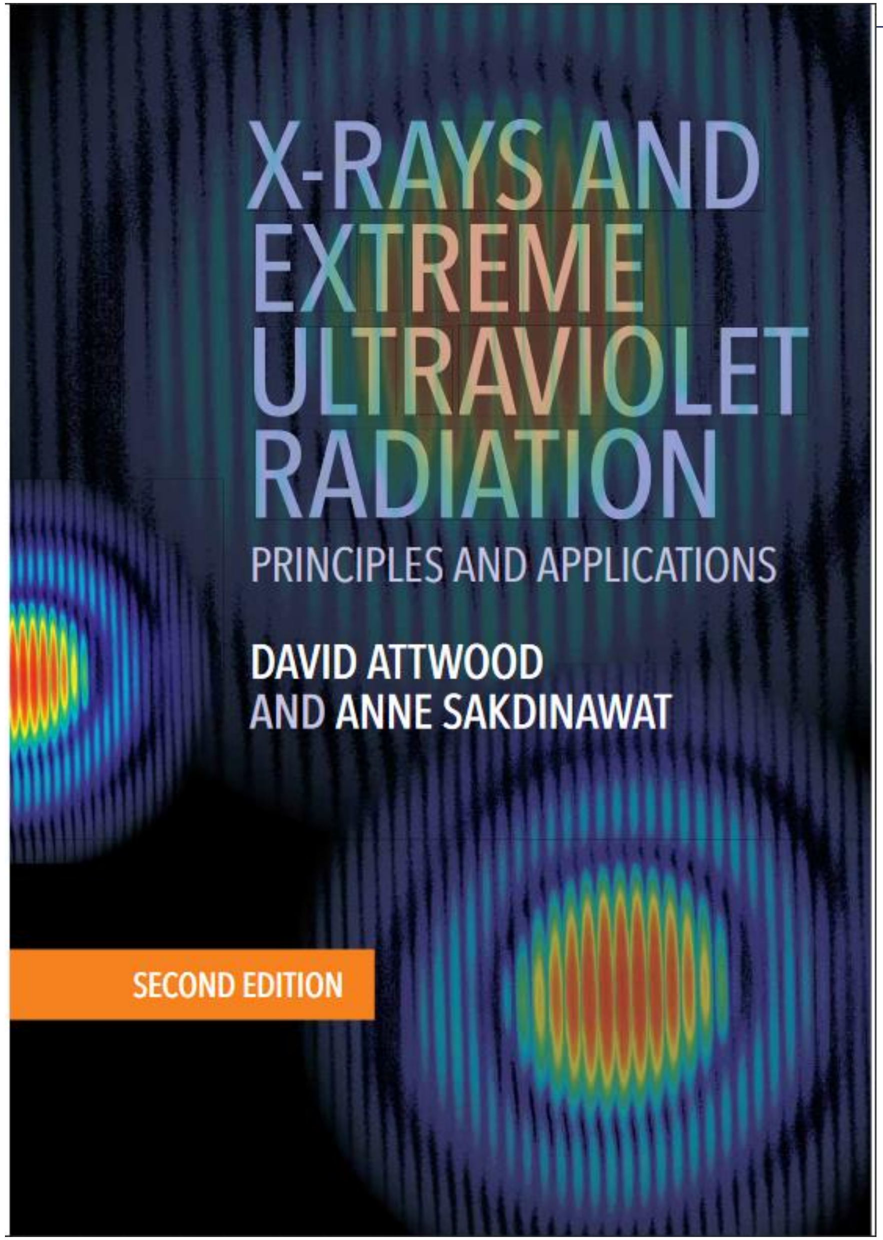


Photon Energy	9keV
Pulse energy	1.2 mJ (bkg 0.4 mJ)
BW (FWHM)	0.8 eV
Spectral density	**1000 uJ/eV**

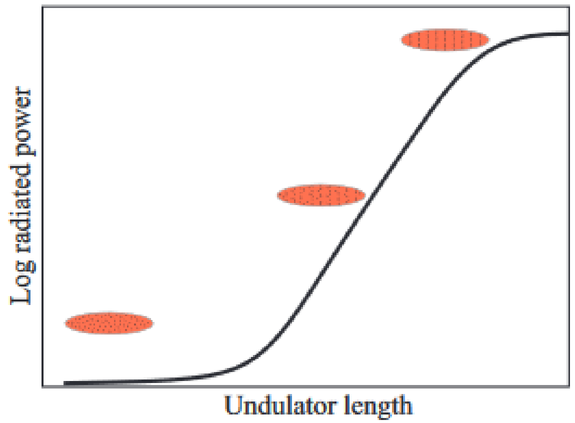
X-RAY FREE ELECTRON LASER OSCILLATOR (XFELO)



Photon Energy	7 keV
Pulse energy	800 uJ/pulse
BW (FWHM)	20 meV/pulse
Spectral density	**40000 uJ/eV**



6 X-Ray and EUV Free Electron Lasers



$$\rho_{FEL} = \left(\frac{\hat{K}^2 r_e n_e \lambda_u^2}{32\pi\gamma^3}\right)^{1/3} \qquad (6.9)$$

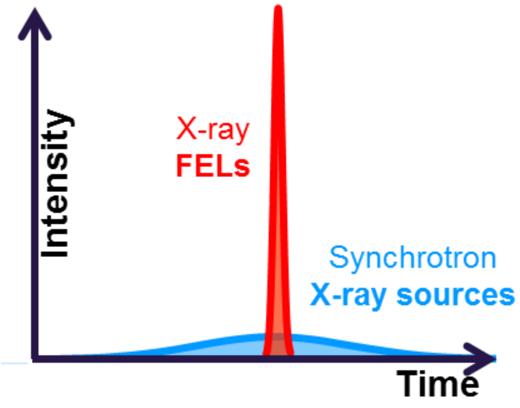
$$L_G = \lambda_u / 4\pi\sqrt{3}\rho_{FEL} \qquad (6.12)$$

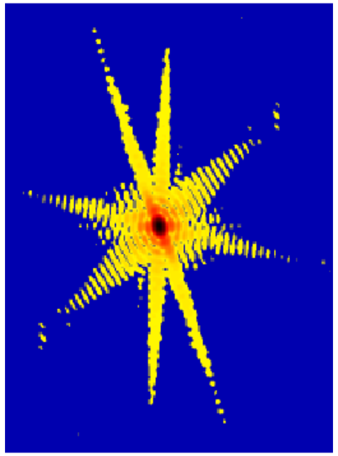
$$\hat{P}_{sat} \simeq \rho_{FEL} \cdot \underbrace{\gamma mc^2 \cdot \hat{I}/e}_{\hat{P}_e} \qquad (6.13)$$

$$2\sigma\sigma' \cong \lambda/2\pi \qquad (6.2)$$

www.cambridge.org/xrayeuv

New opportunities offered by X-ray FELs





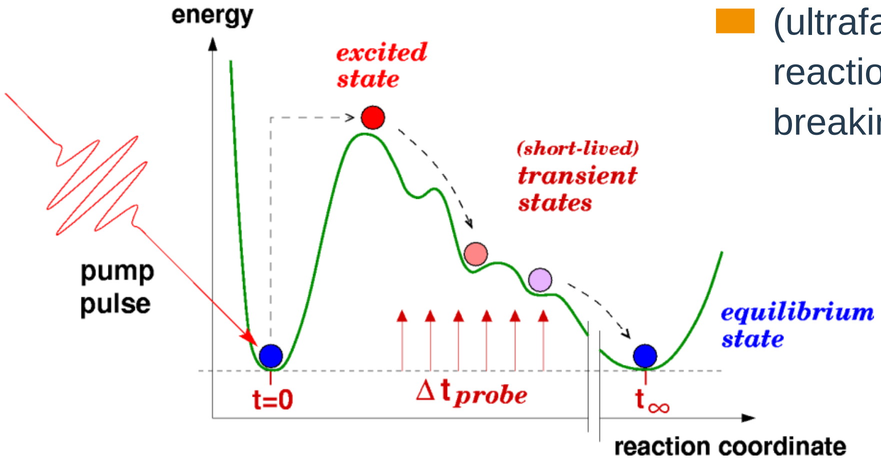
Ultrashort pulses
1 – 100 fs

Coherence
Almost full transverse
Partially temporal

Intensity/power
up to few mJ
up to >10^{20} W/cm^2

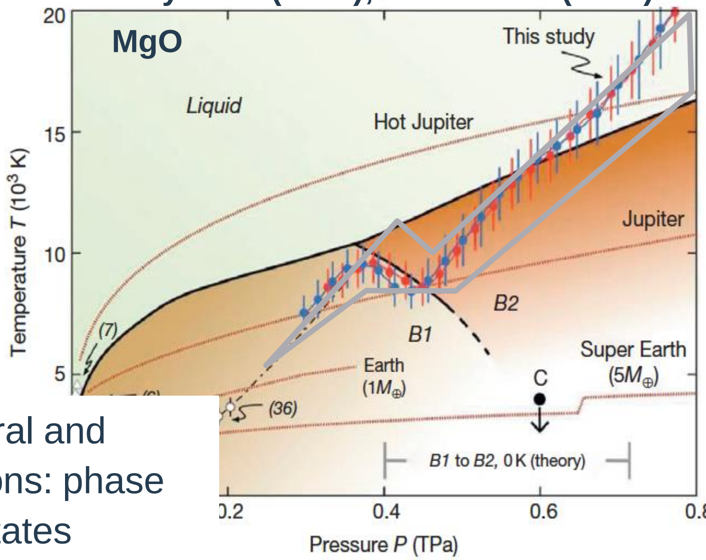
- Structural dynamics
  - **Measurement of atomic and electron dynamics with high spatial [0.1 nm] and temporal [10 fs] res.**
  - **physics, materials sciences, chemistry, life science**
- Imaging at the nanoscale
  - **Imaging experiments on confined and extended objects with atomic to mesoscale resolution [0.1–1000 nm]**
  - **physics, materials sciences, chemistry, life science**
- Non-linear x-ray science
  - **Start using non-linear techniques to obtain hidden information (off-diagonal elements in reaction matrices)**
  - **physics, chemistry**

Open science problems

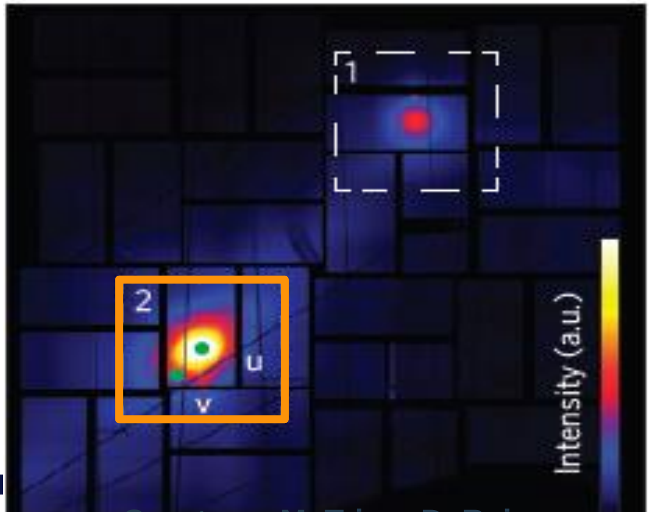


- (ultrafast) (bio-)chemical processes: reactions, phase transitions, bond breaking & forming

Duffy et al. (2015); Bollis et al. (2016)



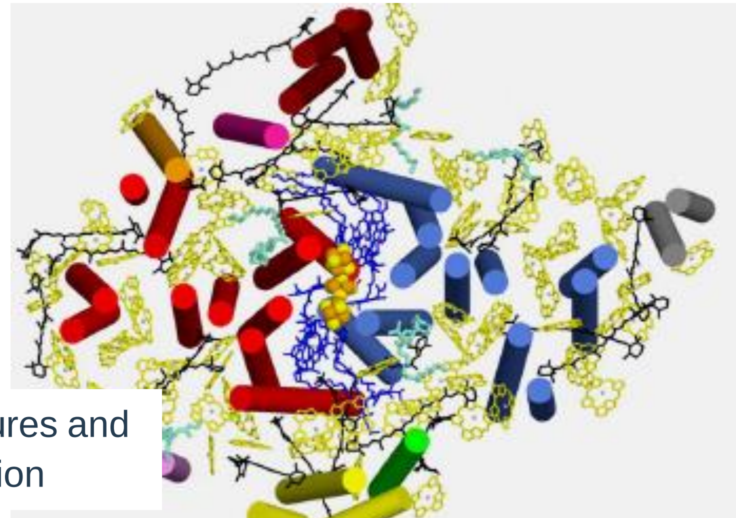
- (ultrafast) structural and electronic transitions: phase transitions, new states



Courtesy: M. Trigo, D. Reis

- Systems ‚in function‘: excited states, non-reversible processes

- complex (bio-)structures and their temporal evolution



PS-II

Outline

- Introduction
- Free-electron laser basics
- **X-ray FEL facilities**

  – break –
- Ultra fast processes in chemistry and materials
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- Extreme states of matter
- Future

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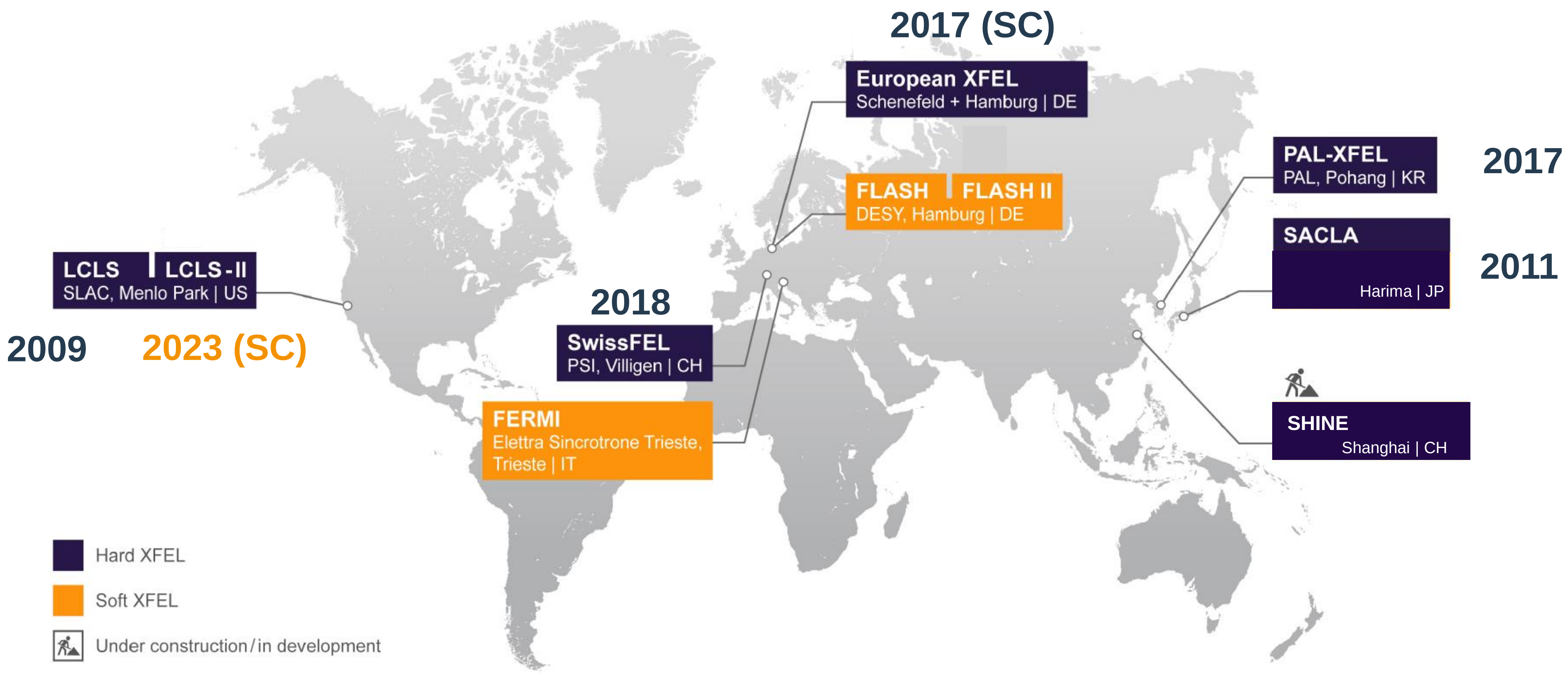
X-ray free-electron lasers worldwide



Some specifics about FEL facilities

- FLASH – first user facility (2005) for XUV and soft x-ray FEL radiation
- FERMI – first user facility to successfully employ laser seeding (XUV to soft x-rays)
- LCLS – first user facility (2009) for hard x-ray FEL radiation and later self-seeding
- SACLA, PAL, SwissFEL – compact hard x-ray FELs
- European XFEL – First international user facility (2017) for soft and hard x-ray FEL radiation

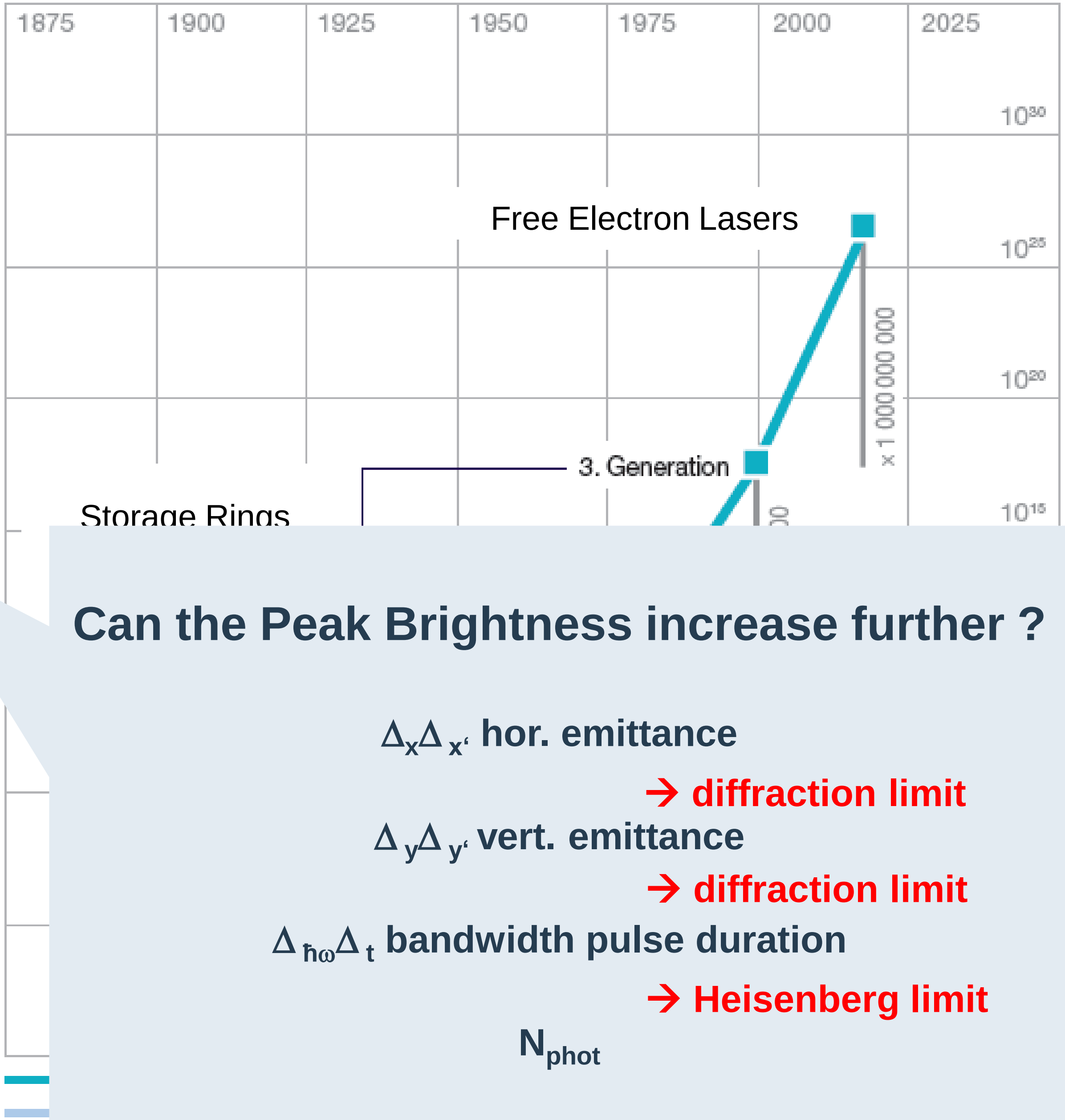
- LCLS-II(-HE), SHINE – first cw-type soft and hard x-ray FELs

Development of peak brightness

- Peak brightness is the quantity best describing performance of pulsed coherent (laser) x-ray sources

$$\text{Peak brightness} = \frac{Number\ of\ \ photons}{\Delta_x \Delta_{x'} \Delta_y \Delta_{y'} \times bandwidth \times \Delta_t}$$

- SR sources use average brightness (brightness scaled to 1 second)
- Tremendous improvement opens route to new methods & applications



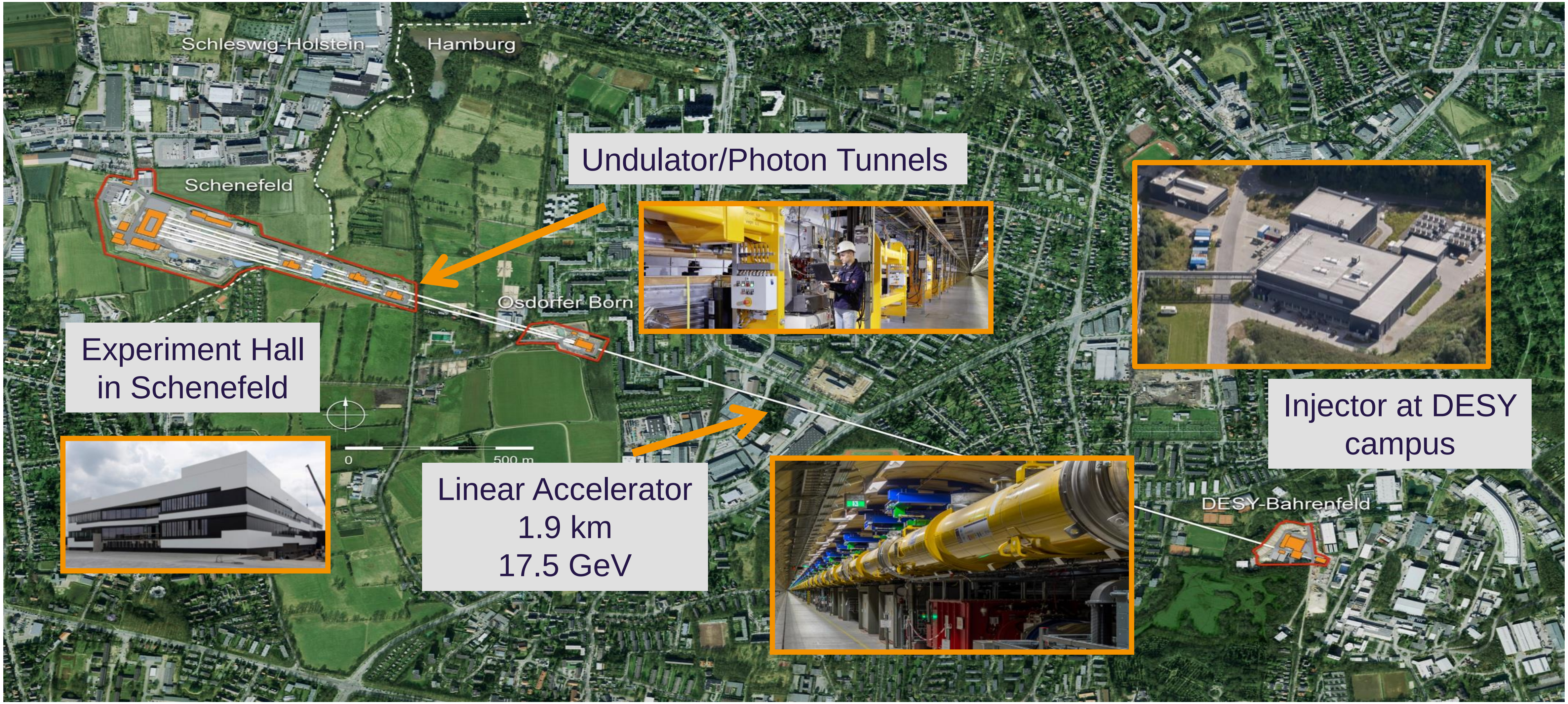
Can the Peak Brightness increase further ?

$\Delta_x \Delta_{x'}$ hor. emittance
→ diffraction limit
$\Delta_y \Delta_{y'}$ vert. emittance
→ diffraction limit
$\Delta_{\hbar\omega} \Delta_t$ bandwidth pulse duration
→ Heisenberg limit
N_{phot}

About the European XFEL

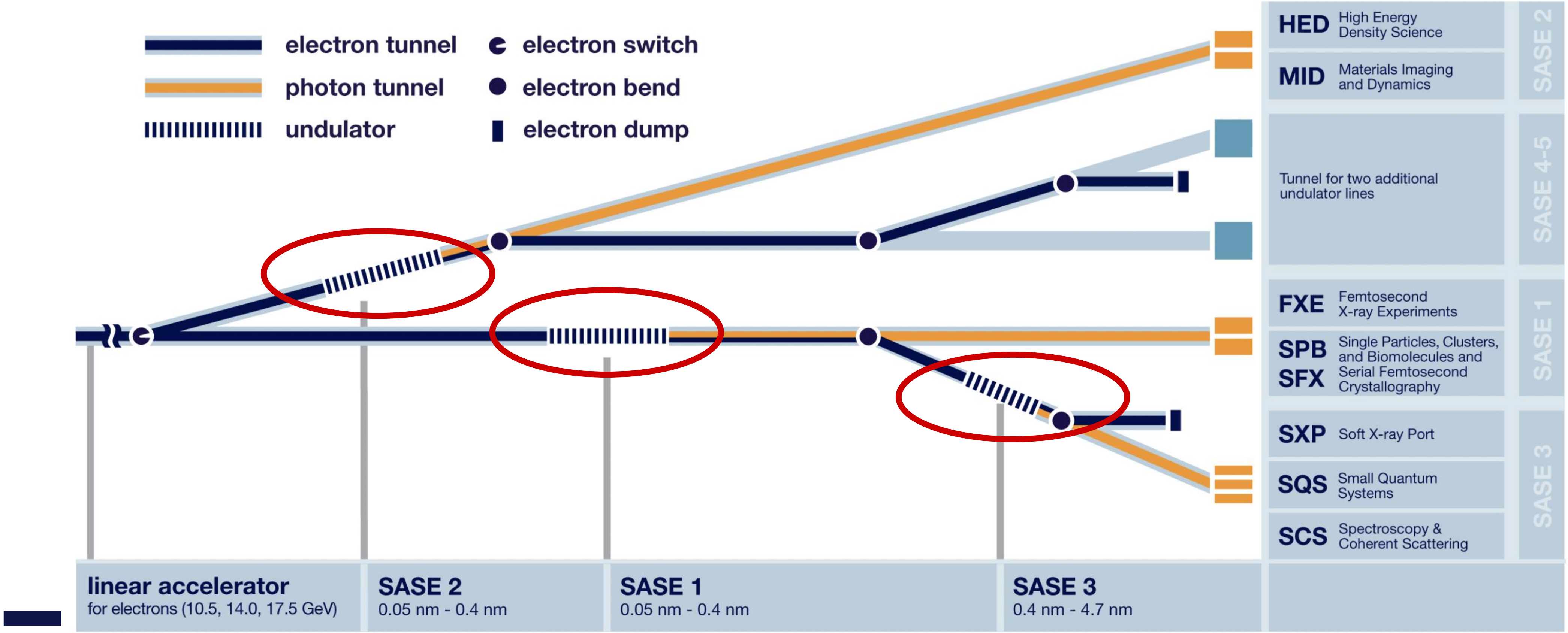
- International user facility for FEL research
  - **Providing soft & hard X-ray FEL radiation**
  - **Photon energy 300 eV to ≥ 20 keV**
  - **High pulse energies**
  - **MHz repetition rates**
- Multi-disciplinary science community
  - **Physics**
  - **Chemistry**
  - **Biology**
  - **Geosciences**
  - **Materials**
- Status
  - **Construction 2009-2017**
  - **First experiments 2017**
  - **6 instruments operating since 2019 (+ 1 in 2023)**
  - **Enter now “Harvesting” phase**

Layout of the European XFEL



Beam distribution & instruments

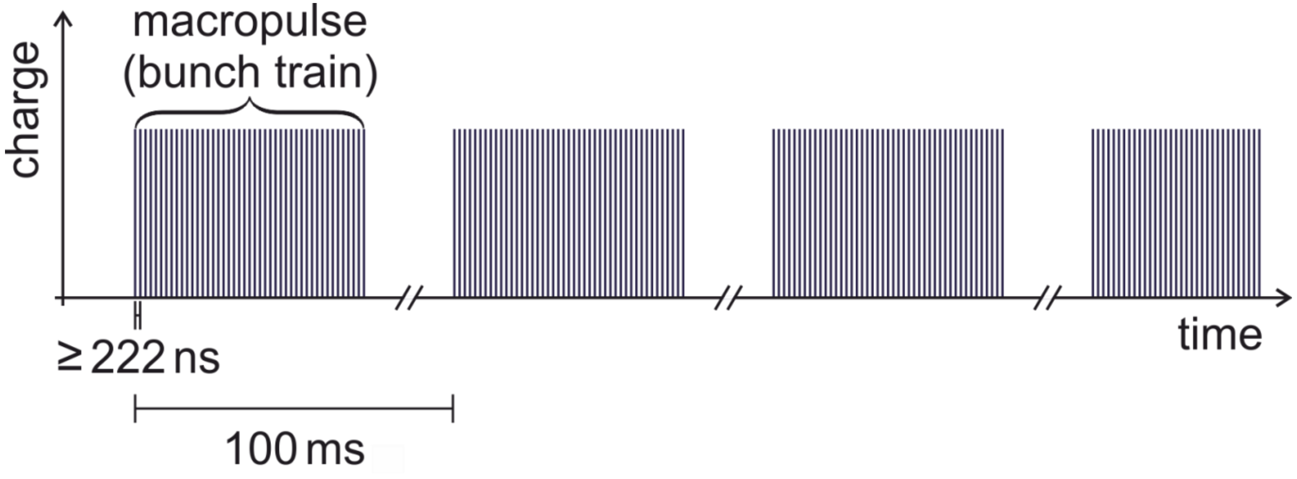
Undulator Segment	FEL radiation energy [keV]	Wavelength [nm]
SASE 1	3 - over 24	0.4 - 0.05
SASE 2	3 - over 24	0.4 - 0.05
SASE 3	0.27 – 3	4.6 – 0.4

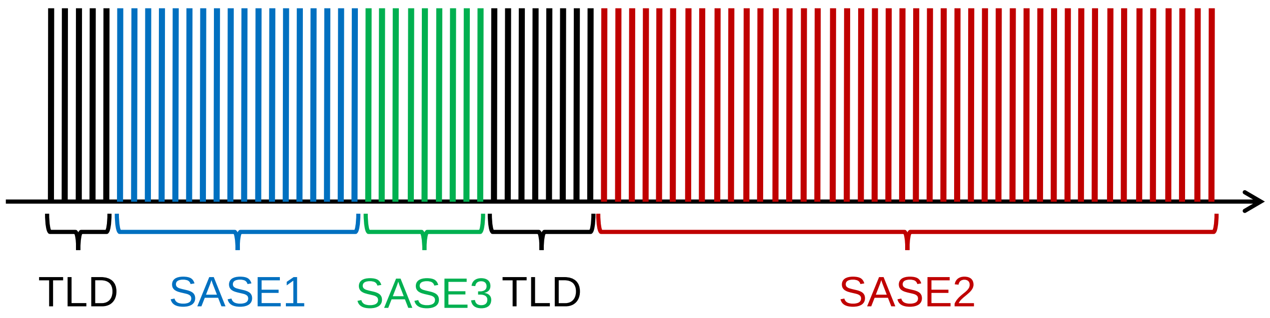


Key parameters of EuXFEL

Parameter	Value
Electron Energy	8.5 – 17.5 GeV
Photon energy	0.26 - 25 keV
Pulse duration	2 – 100 fs
Self Seeding	Operational SA2
# of pulses	27000 /s
# of FELs	3
# of instruments	7
Start of operation	2017

- Specific electron & x-ray beam delivery pattern
  - **RF repetition rate: 10 Hz**
  - **RF flat-top length: ~ 600 μs**
  - **Up to 2700 bunches/train**
  - **Bunch spacing: Up to 4.5 MHz**



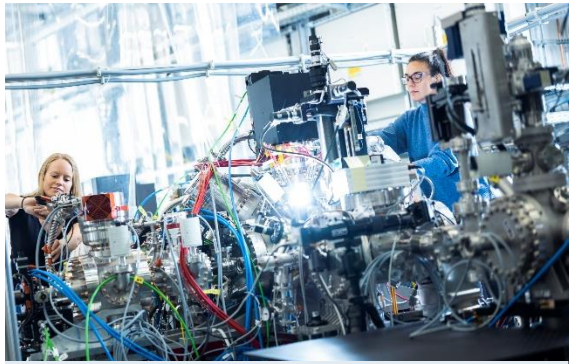


Seven instruments are in user operation

FXE (start Sep 2017)

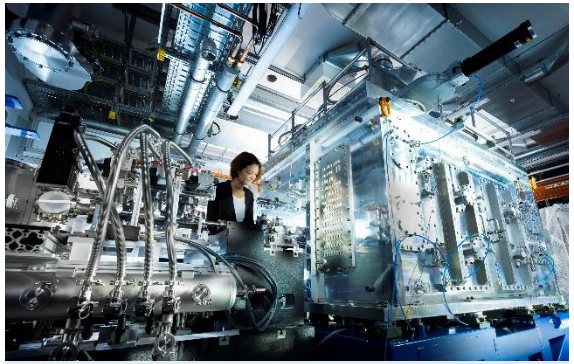
SPB/SFX (start Sep 2017)

SCS (start Nov 2018)

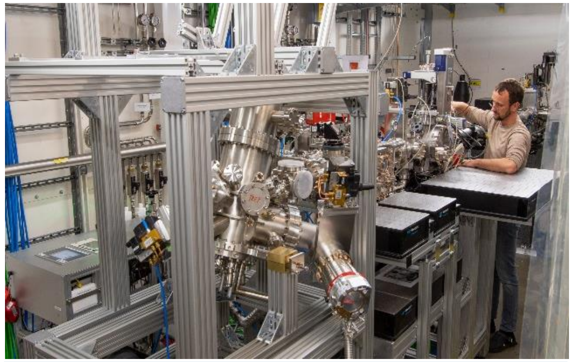


SQS (start Nov 2018)

MID (start Apr 2019)



HED (start May 2019)



SXP (start summer 2023)

Scientific instruments

SASE1

FXE (Femtosecond X-ray Experiments)

- Ultrafast dynamics of liquids and solid matter
- Combination of spec. & scat. techniques

SPB/SFX (Single Part., Bioimaging, & SFX)

- Coherent diffraction imaging from single part.
- Serial fs nano-crystallography

SASE2

MID (Materials Imaging & Dynamics)

- CDI from nano-structured samples
- XPCS of nanoscale dynamics

HED (High Energy Density science)

- Ultrafast dynamics of highly excited matter
- Combinations of scattering, diff. & spectroscopy

SASE3

SQS (Small Quantum Systems)

- Ultrafast dynamics of atoms, ions & clusters
- Combination of spec. & coh. scat. techniques

SCS (Spectroscopy & Coherent Scattering)

- Ultrafast dynamics of complex solids
- Combination of hr-inelastic spec. & coh.scattering

SXP (Soft X-ray Port)

- Time-resolved X-ray photoelectron spectroscopy
- Open port

Examples of X-ray FEL scientific applications

- Ultrafast processes in chemistry and materials

  Make use of fs time resolution w/pump-probe methods

- Biological and soft matter

  Make use of coherence and high intensity

  Serial fs crystallography

  Photon correlation spectroscopy

- Extreme states of matter

  Make use of very intenste and short x-ray pulses

  to study short- lived states

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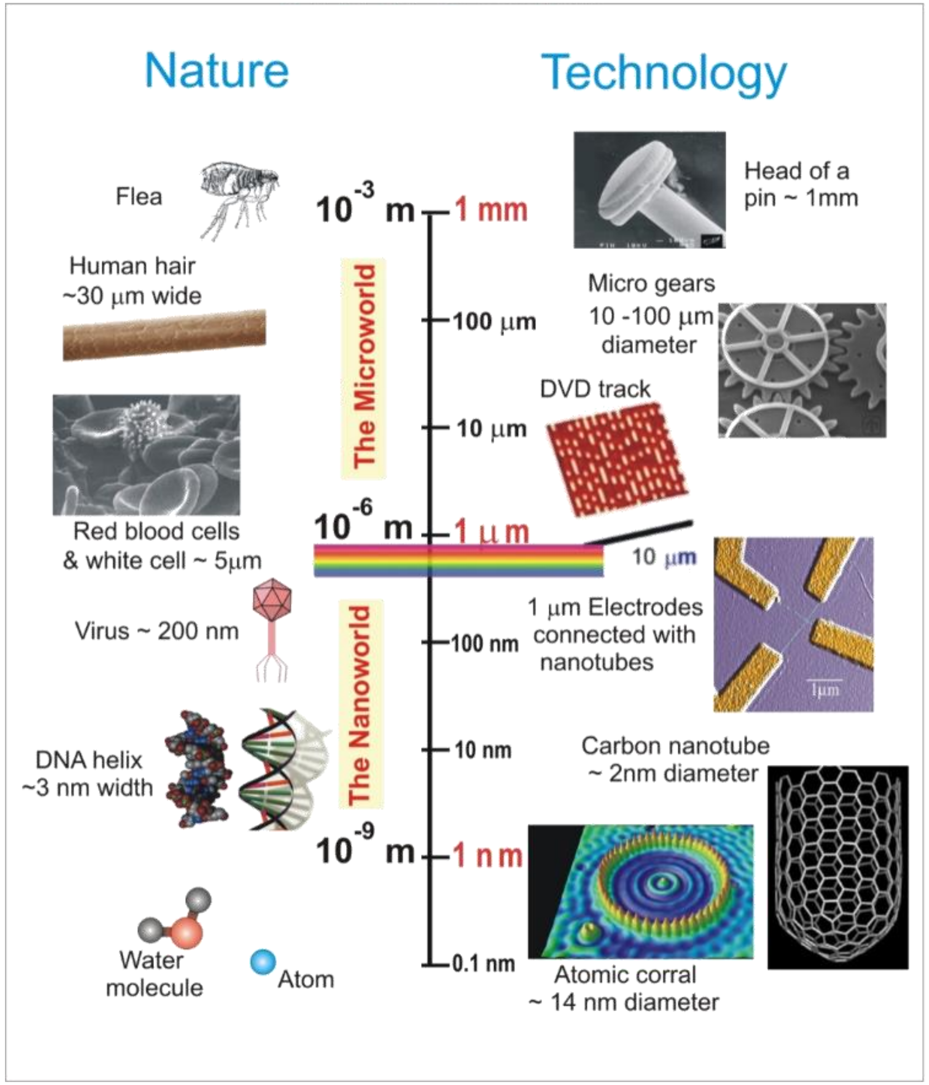
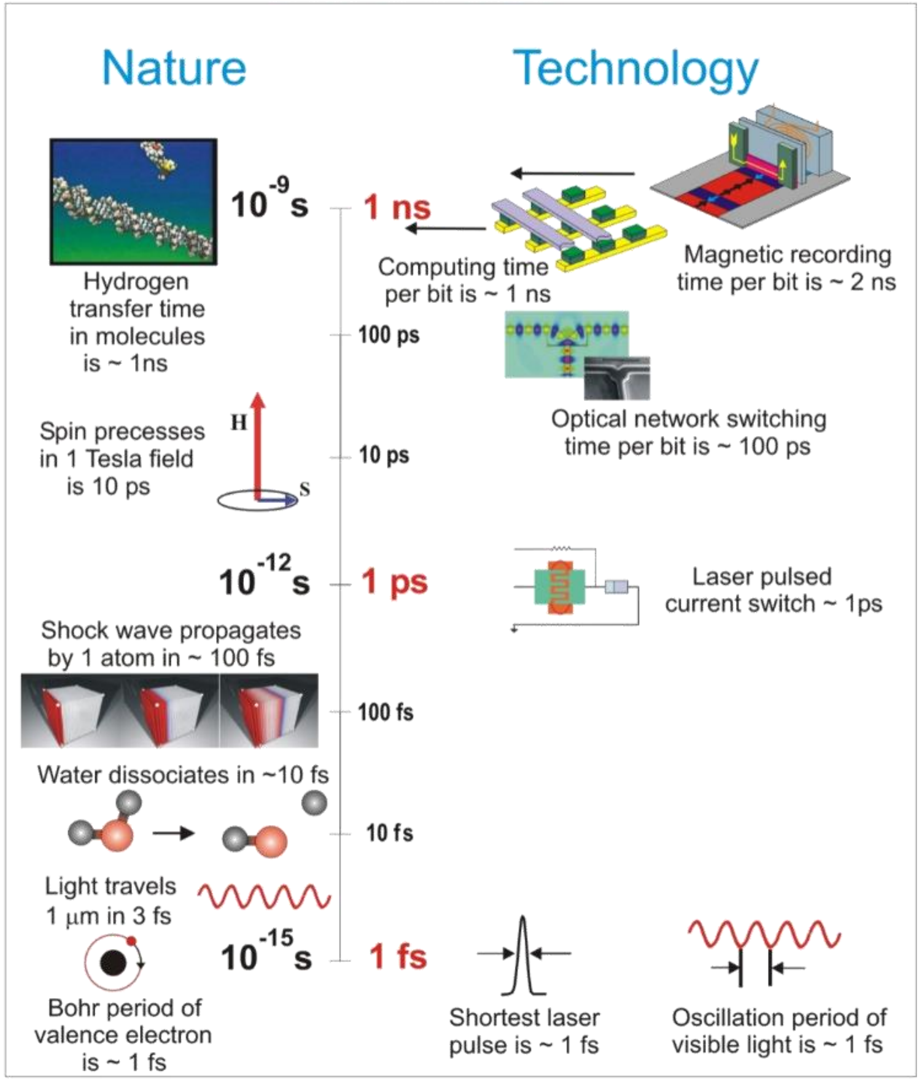
Ultrafast x-ray science

- Connection of length and time scales

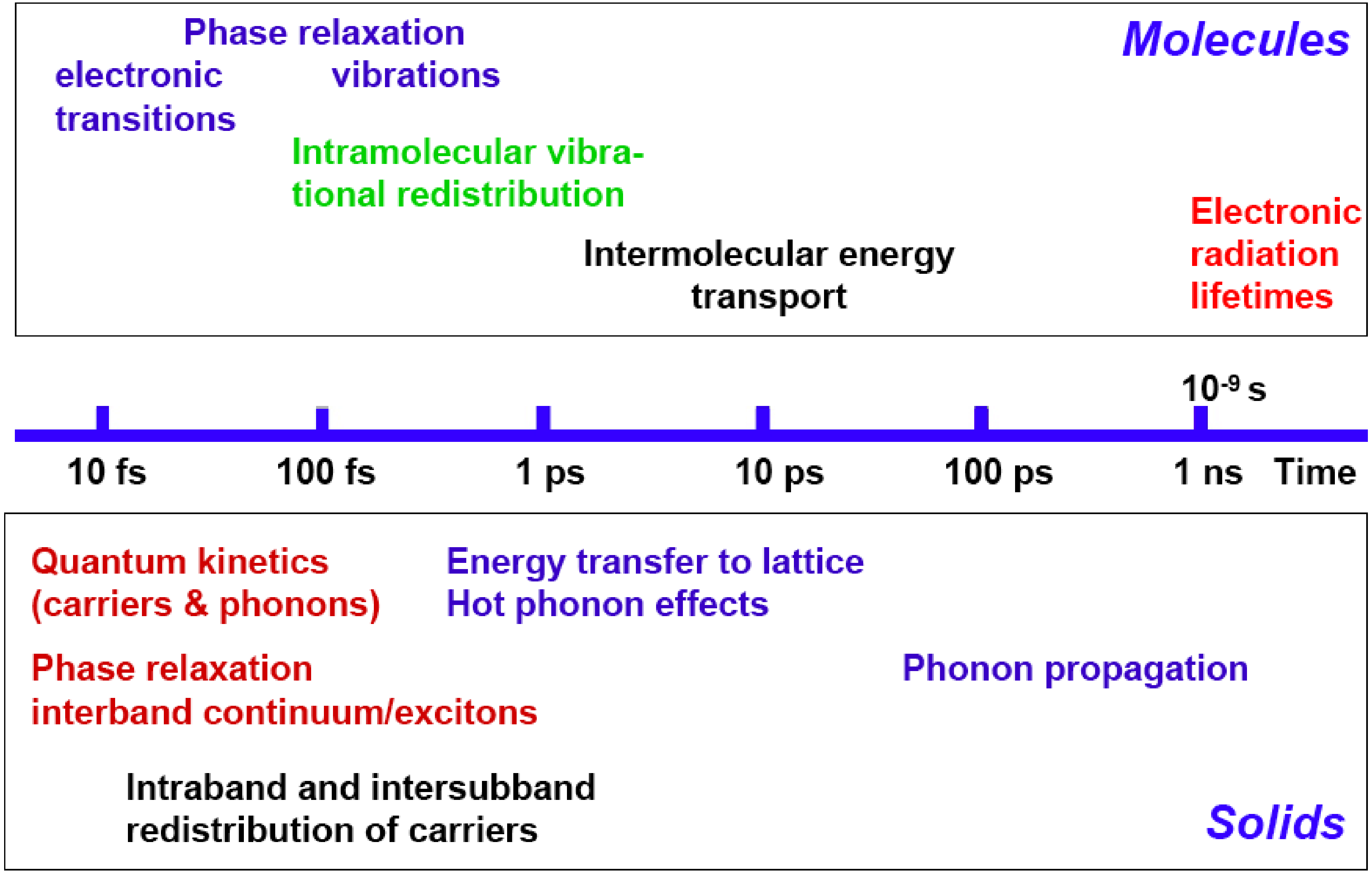




Courtesy: SLAC

Investigations of structural dynamics

- Atoms, molecules, clusters
- Solids, materials
- Complex matter
- Extreme states



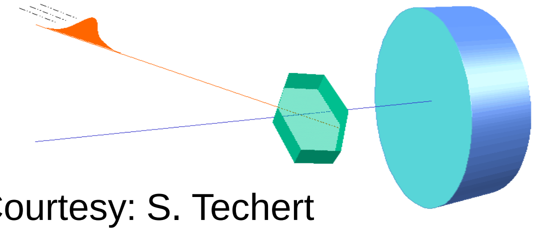
Performing ultrafast x-ray experiments using FELs

- Usually performed in pump-probe mode
  - **Lack of ultrafast detection schemes**
  - **Exception are high rep. rate FELs**

- Resolution elements
  - **Exciting pump pulse: OL, x-ray, others**
  - **Probe pulse: typ. x-ray**
  - **Precision of time-difference (jitter)**

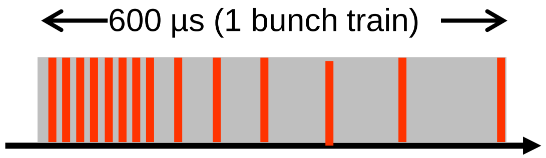
- Synchronization of different light sources

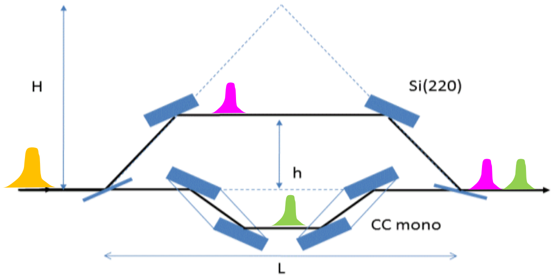
- Application range: fs-to-ns or even fs-to-µs
  - **Signal-to-noise excellent**
  - **Single measurement capability**

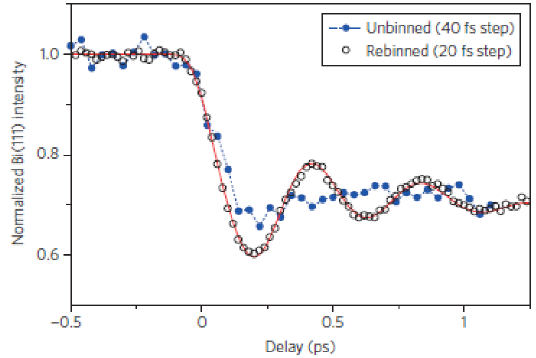


Courtesy: S. Techert

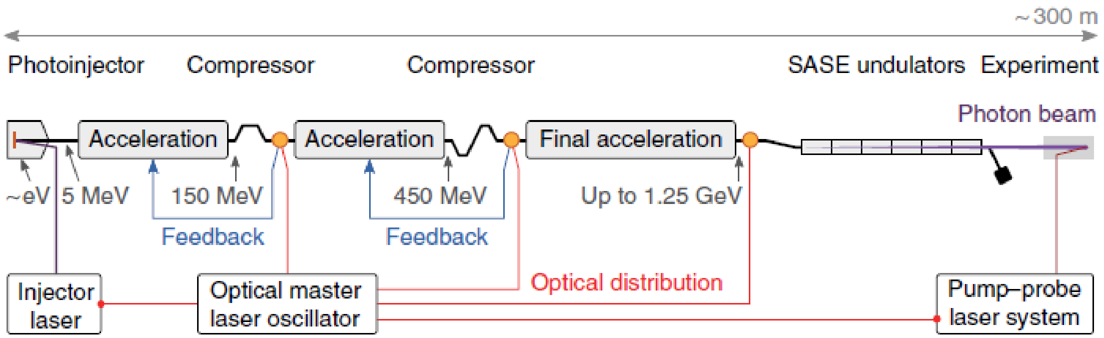
European XFEL pulse train







M. Harmand et al., Nature Phot. 7, 215 (2013)



S. Schulz et al., Nat. Comm. 6, 5938 (2015)

Chemical Dynamics

LETTER

doi:10.1038/nature14163

PRL 114, 255501 (2015)

PHY

Probing the transition stat
catalytic CO oxidation on

ARTICLE

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Visualizing the non-equilibrium dynamics o
photoinduced intramolecular electron trans
with femtosecond X-ray pulses

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The Journal of Physical Chemistry Letters

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Letter

Revealing Hot and Long-Lived Metastable Spin States in the Photoinduced Switching of Solvated Metallogrid Complexes with Femtosecond Optical and X-ray Spectroscopies

Maria Naumova, Aleksandr Kalinko, Joanne W. L. Wong, Mohamed Abdellah, Huifang Geng, Edoardo Domenichini, Jie Meng, Sol Alvarez Gutierez, Pierre-Adrien Mante, Weihua Lin, Peter Zalden, Andreas Galler, Frederico Lima, Katharina Kubicek, Mykola Biednov, Alexander Britz, Stefano Checchia, Victoria Kabanova, Michael Wulff, Jennifer Zimara, Dirk Schwarzer, Serhiy Demeshko, Vadim Murzin, David Gosztola, Martin Jarenmark, Jianxin Zhang, Matthias Bauer, Max Latevi Lawson Daku, Wojciech Gawelda, Dmitry Khakhulin, Christian Bressler, Franc Meyer, Kaibo Zheng, and Sophie E. Canton*

Cite This: https://dx.doi.org/10.1021/acs.jpclett.9b03883

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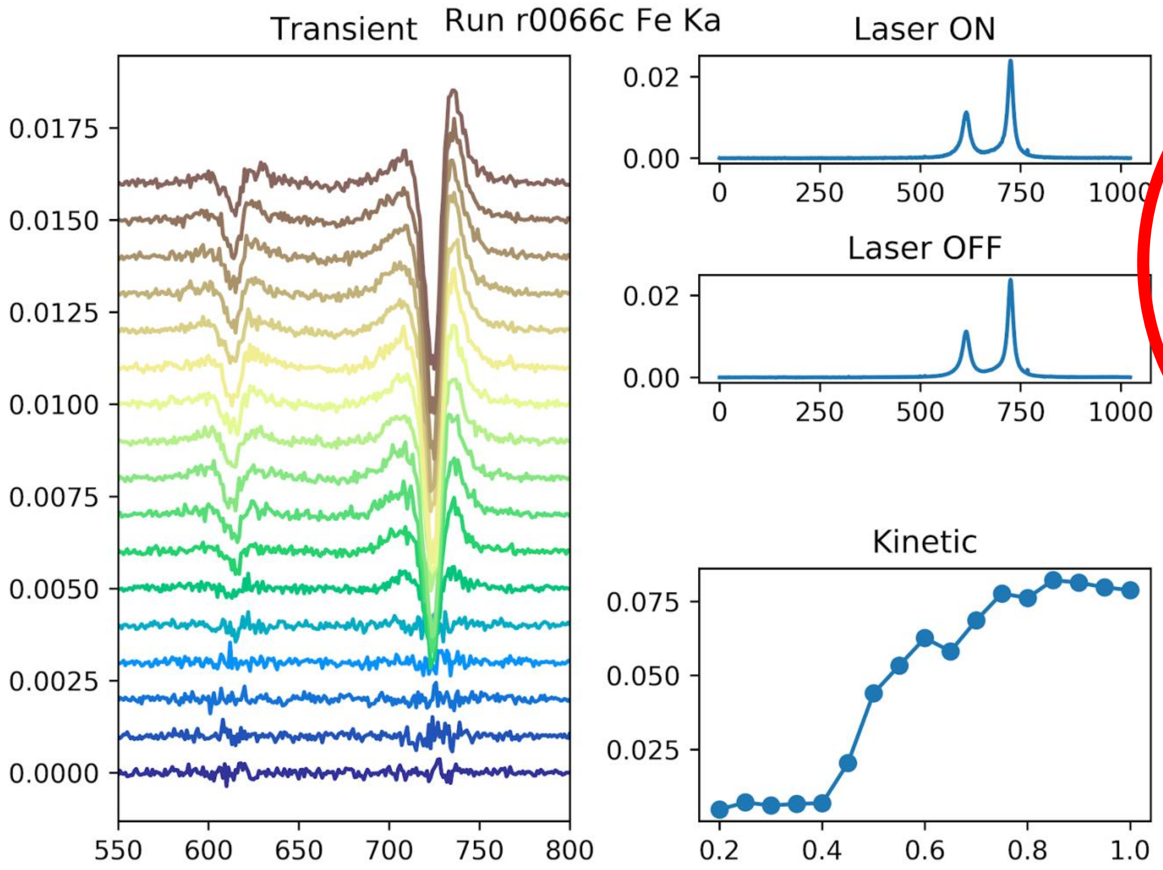
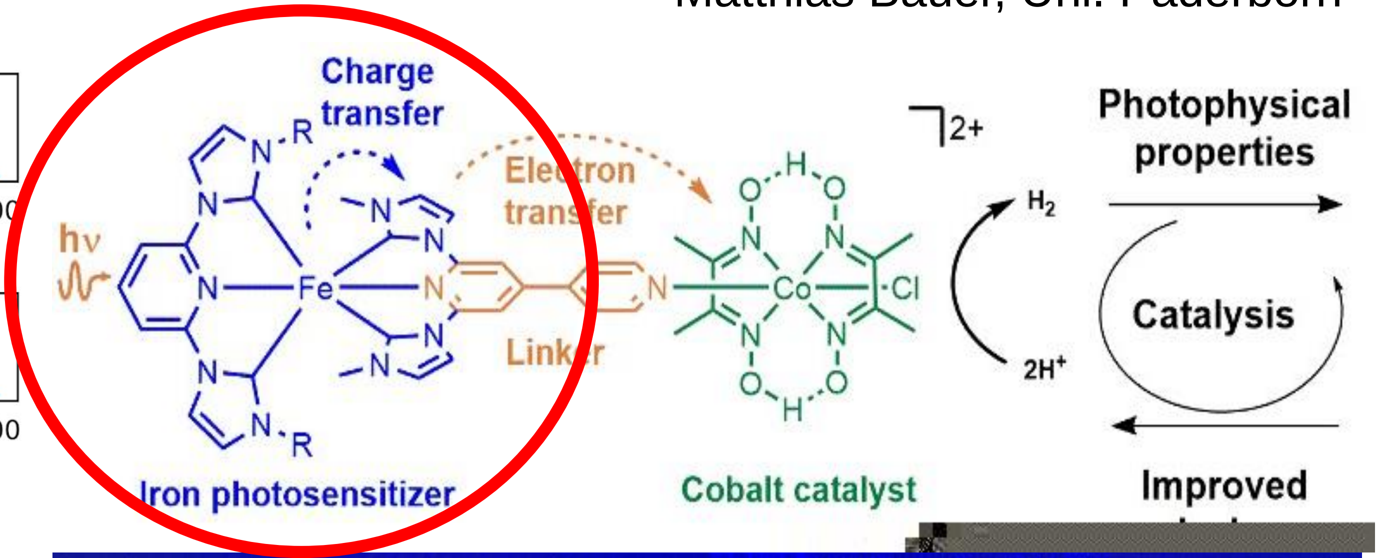
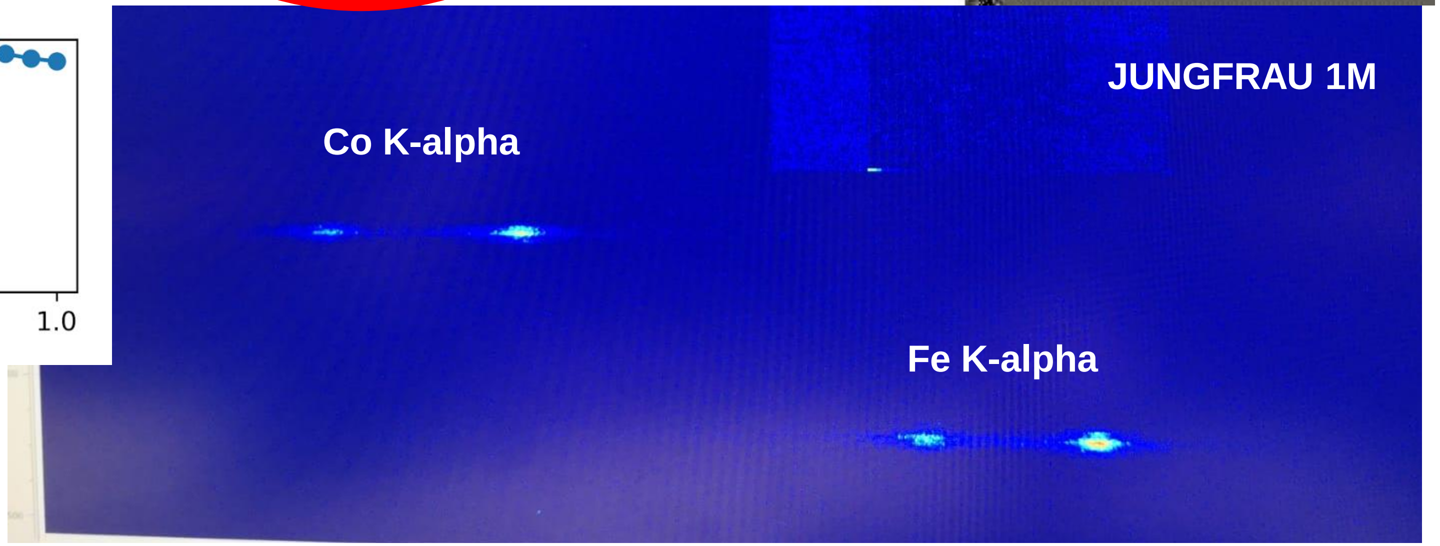
Charge transfer dynamics in photocatalysts

Matthias Bauer, Uni. Paderborn



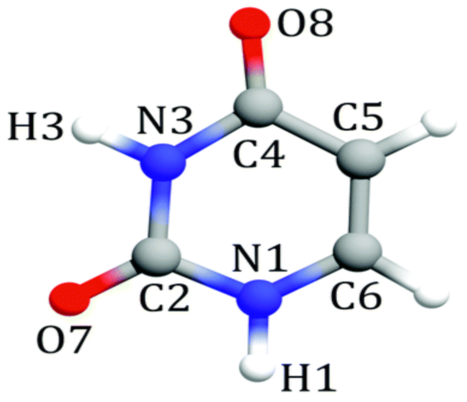




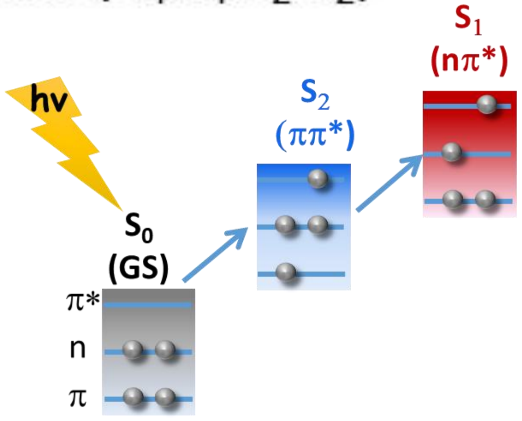
FXE

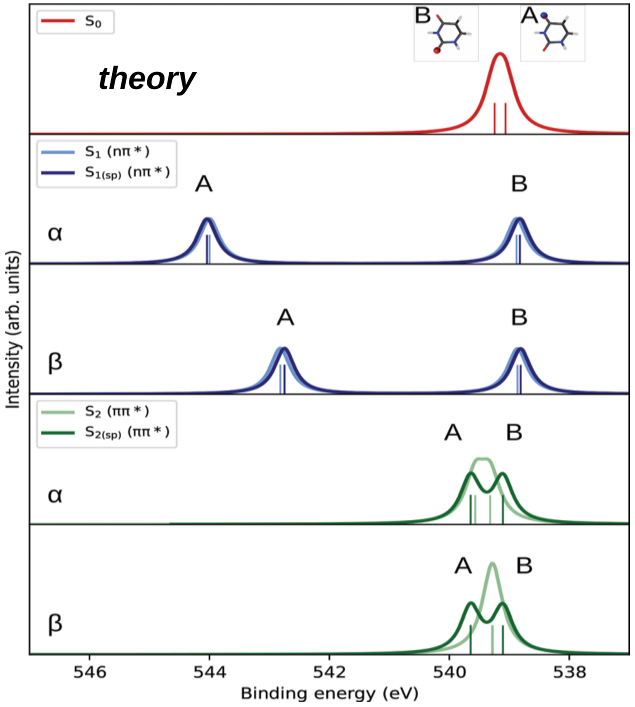
Understanding the ultrafast dynamics of DNA/RNA

- Investigating the ultrafast photochemistry of nucleobases to address the photodamage and photoprotection mechanisms
- Photoelectron spectroscopy as a marker of the photoexcited states

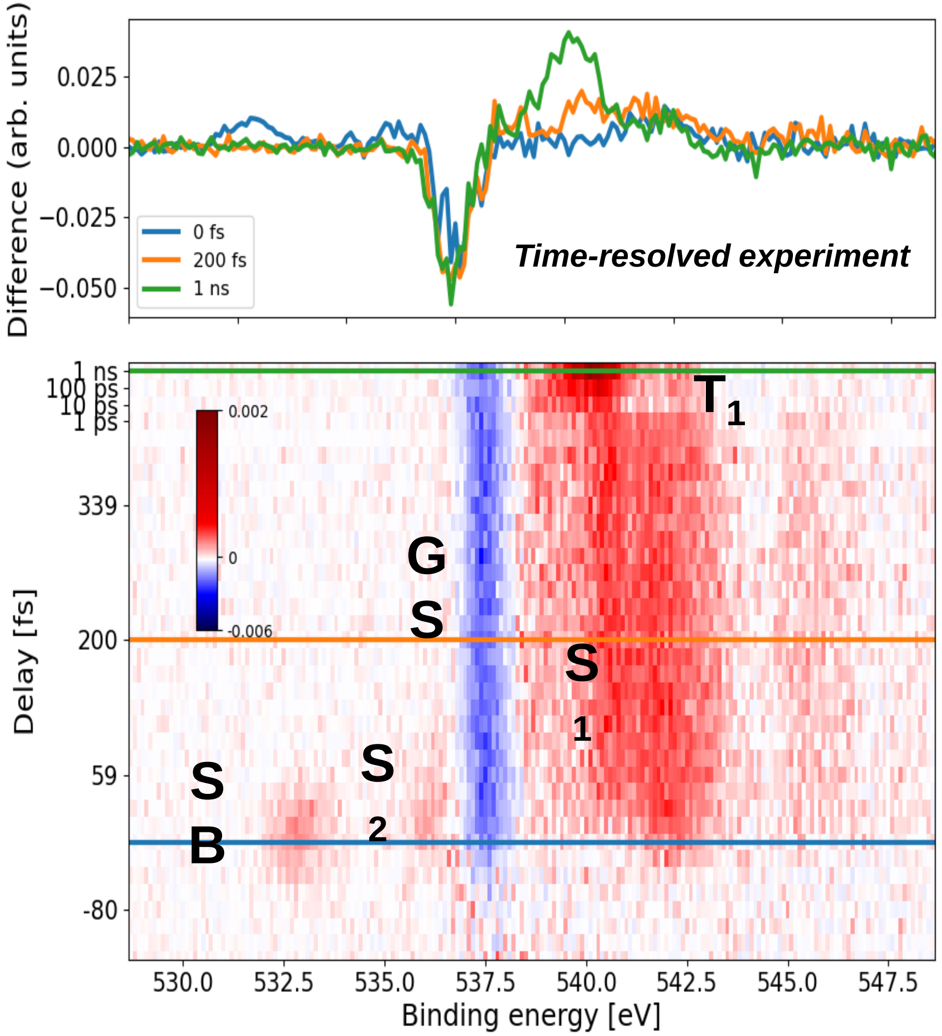


Uracil ($C_4H_4N_2O_2$)





SQS

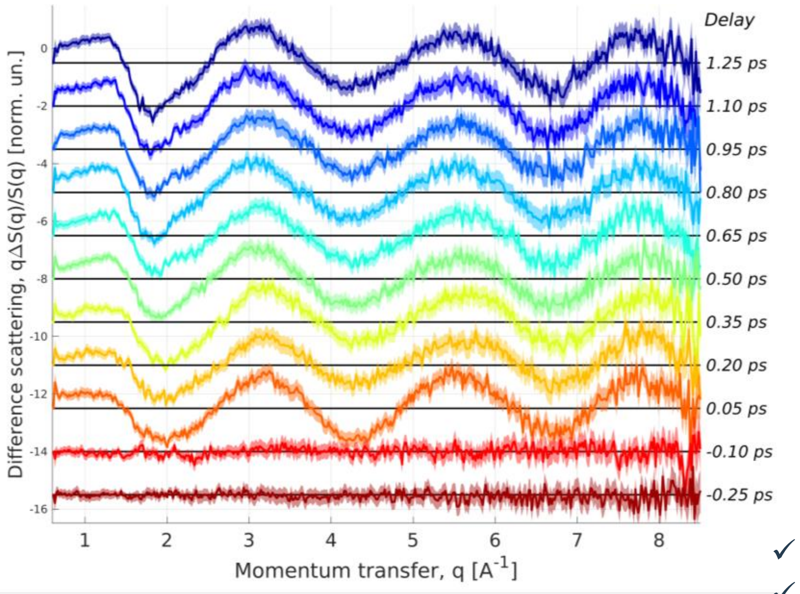


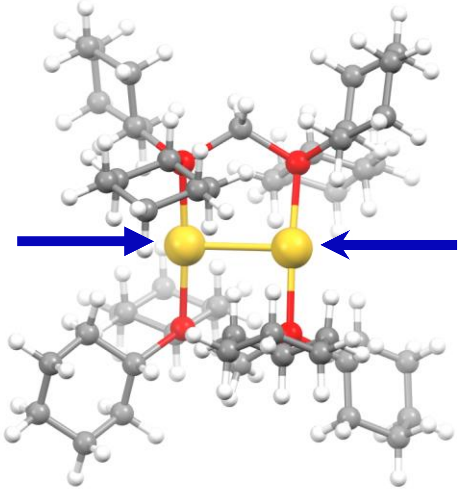
PI: Oksana Plekan, Elettra

European XFEL

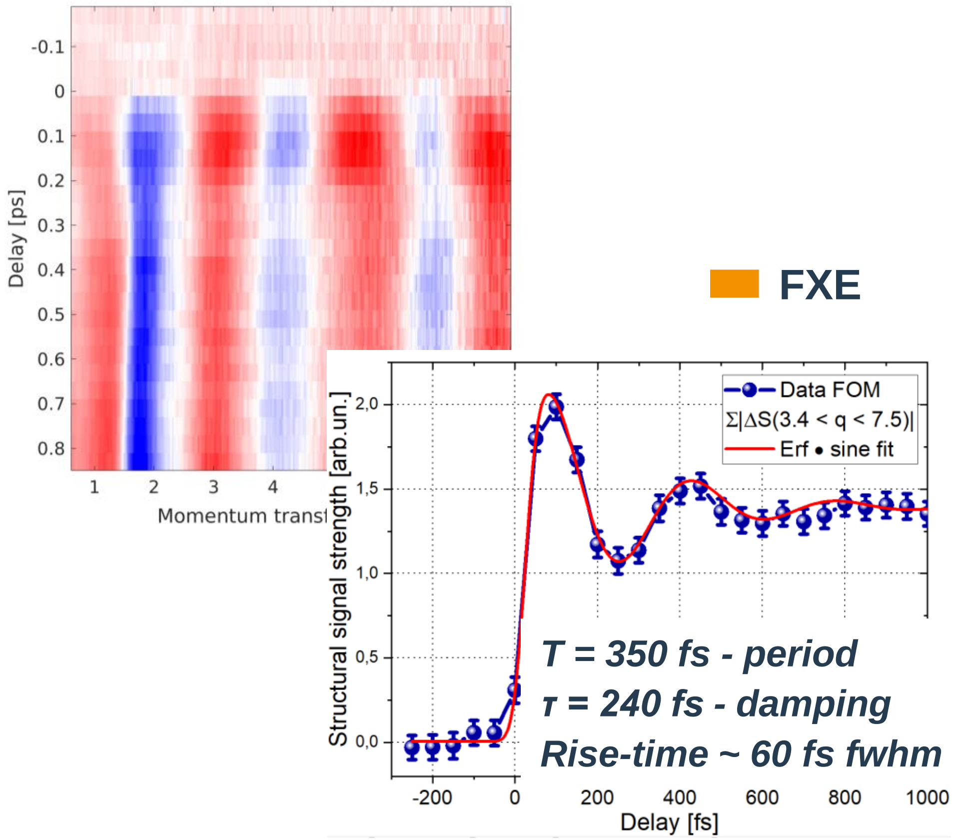
Aurophilicity in Au(I) complexes by fs X-ray solution scattering

- Applications in chemical sensing, OLED devices, bio-medical imaging
- Metal-metal interaction; equivalent to strong H bonding
- Study of excited state structural and electronic dynamics
- With high photon energy (high q), Au-Au distance can be measured





- ✓ **Au-Au distance decrease = 0.08 Å**
- ✓ Vibrational wave packet formation ~**300 fs**



FXE

Ph.D. thesis: Sharmistha Paul Dutta
PI: Dmitry Khakhulin
Local Contact: Mykola Biednov

Materials

LETTERS
PUBLISHED ONLINE: 27 OCTOBER 2013 | DOI: 10.1038/NPHYS2788
nature physics

Fourier-transform inelastic X-ray scattering from
-phonon

itz[3], K. Gaffney[2],
,
, G. Wang[10],

PRL 112, 217203 (2014) PHYSICAL REVIEW LETTERS week ending 30 MAY 2014

Imaging Ultrafast Demagnetization Dynamics after a Spatially

lla,[1,2] J. Perron,[3,4] B. Vodungbo,[3,4]
and S. Eisebitt[1,7,8]
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zza, Trieste, Italy
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Fast-track Communication

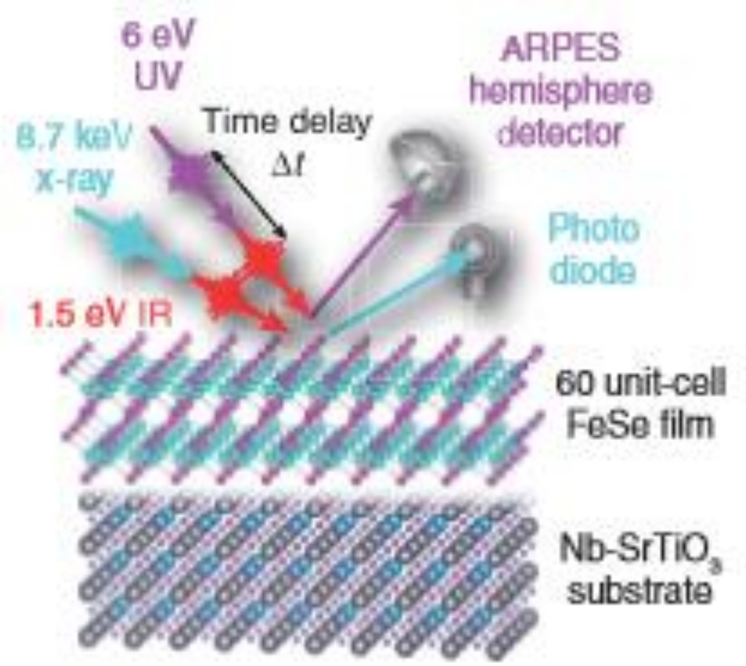
llet[b],

LETTER
doi:10.1038/nature13266

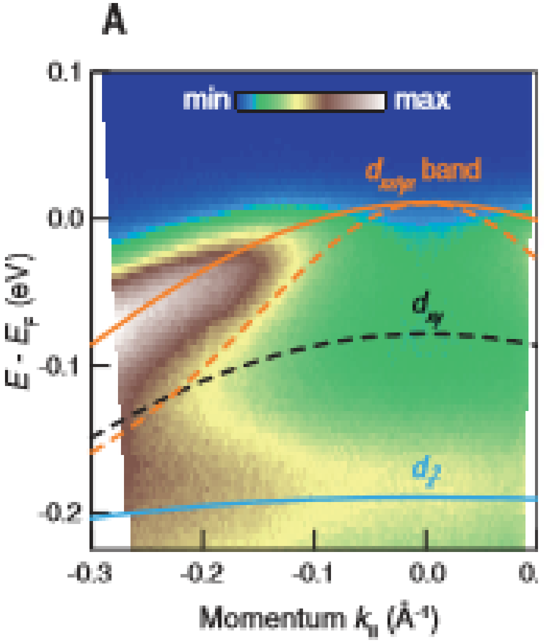
Ultrafast X-ray probing of water structure below the homogeneous ice nucleation temperature

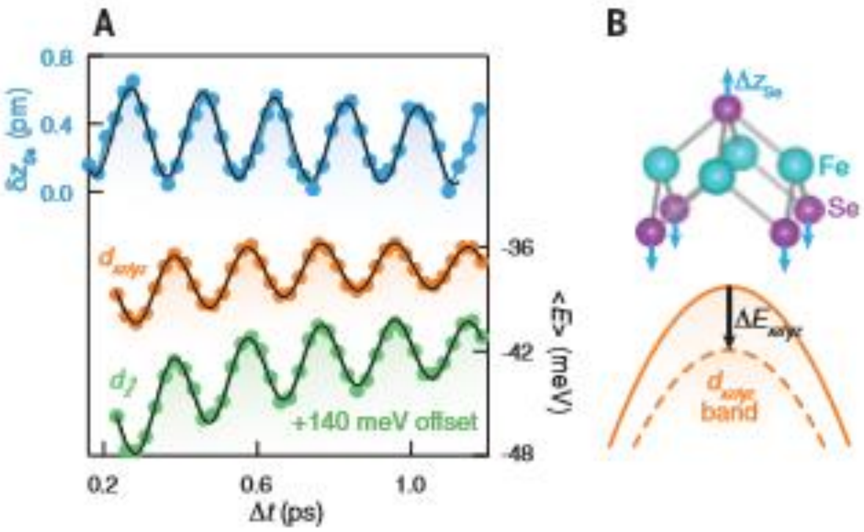
J. A. Sellberg[1,2], C. Huang[3], T. A. McQueen[1,4], N. D. Loh[5], H. Laksmono[5], D. Schlesinger[2], R. G. Sierra[5], D. Nordlund[3], C. Y. Hampton[5], D. Starodub[5], D. P. DePonte[6,7], M. Beye[1,8], C. Chen[1,4], A. V. Martin[6], A. Barty[6], K. T. Wikfeldt[2], T. M. Weiss[3], C. Caronna[7], J. Feldkamp[7], L. B. Skinner[9], M. M. Seibert[7], M. Messerschmidt[7], G. J. Williams[7], S. Boutet[7], L. G. M. Pettersson[2], M. J. Bogan[5] & A. Nilsson[1,2,3]

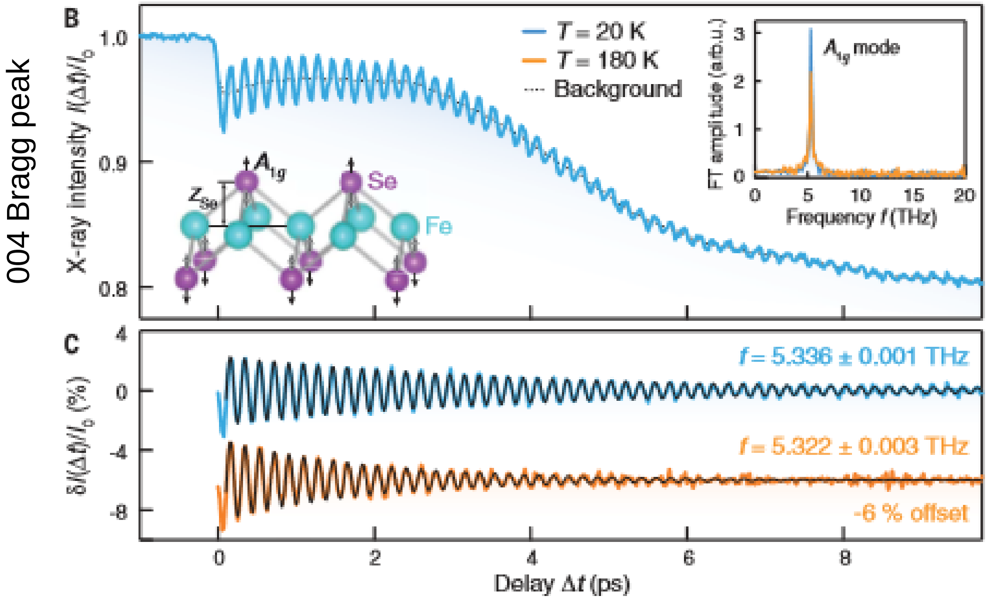
Electron-phonon coupling in superconductors



1. Initiate coherent phonon mode through photo-excitation of electrons.
2. Light induced coherent dynamics of crystal lattice → XRD
3. Electronic band energy → ARPES







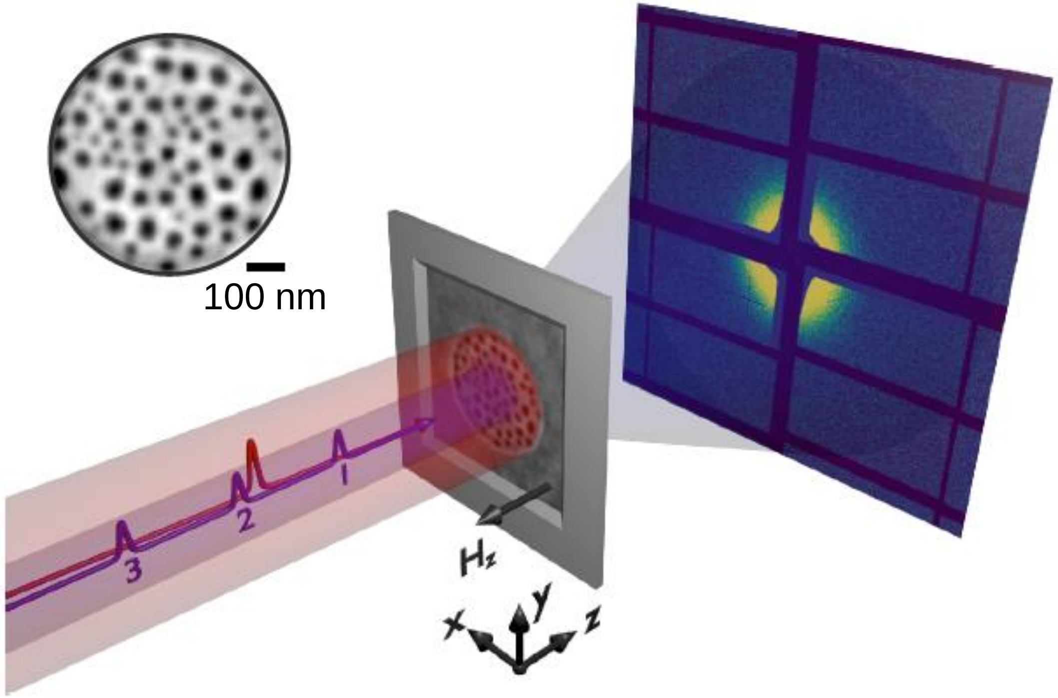
European XFEL

Gerber, Science 2017 (LCLS)

LCLS

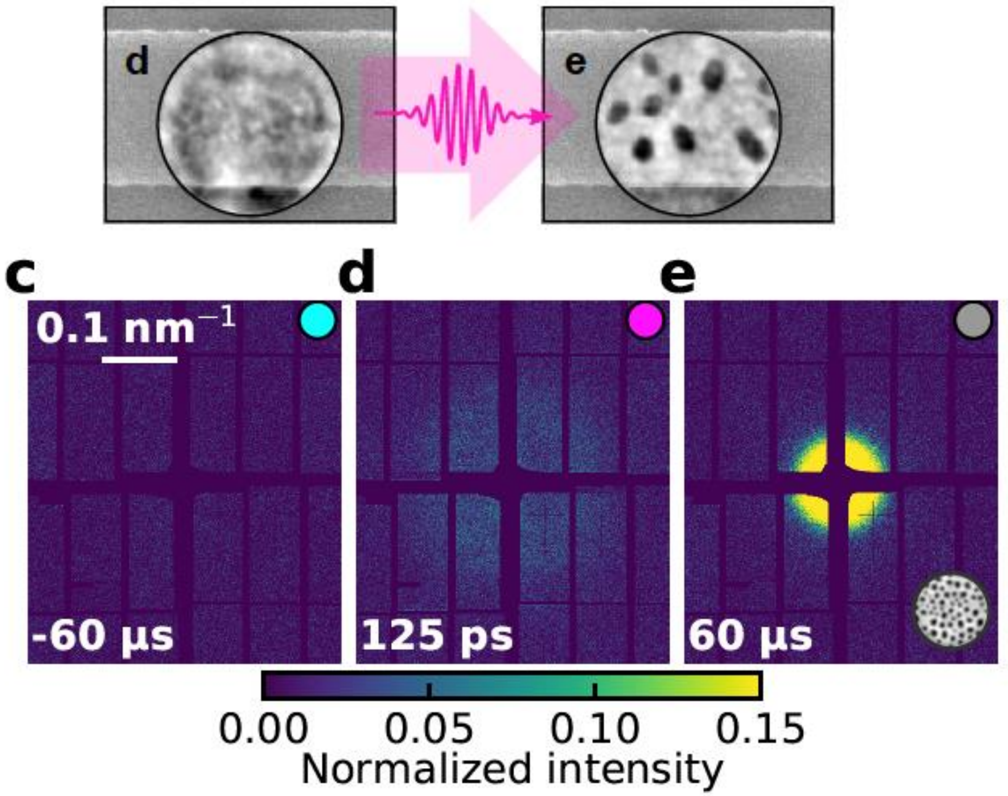
Nucleation and annihilation of magnetic skyrmions

- SCS



nature materials
ARTICLES
https://doi.org/10.1038/s41563-020-00807-1
Check for updates

Observation of fluctuation-mediated picosecond nucleation of a topological phase

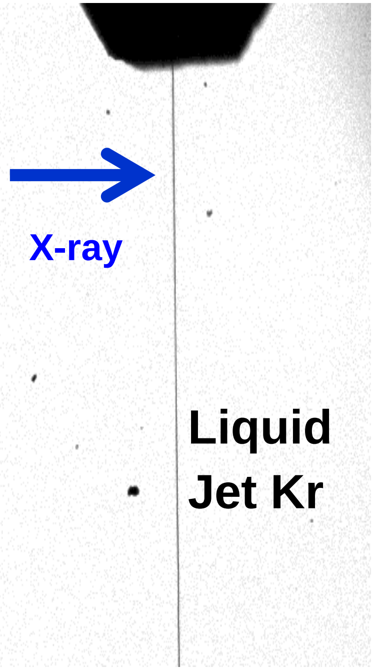


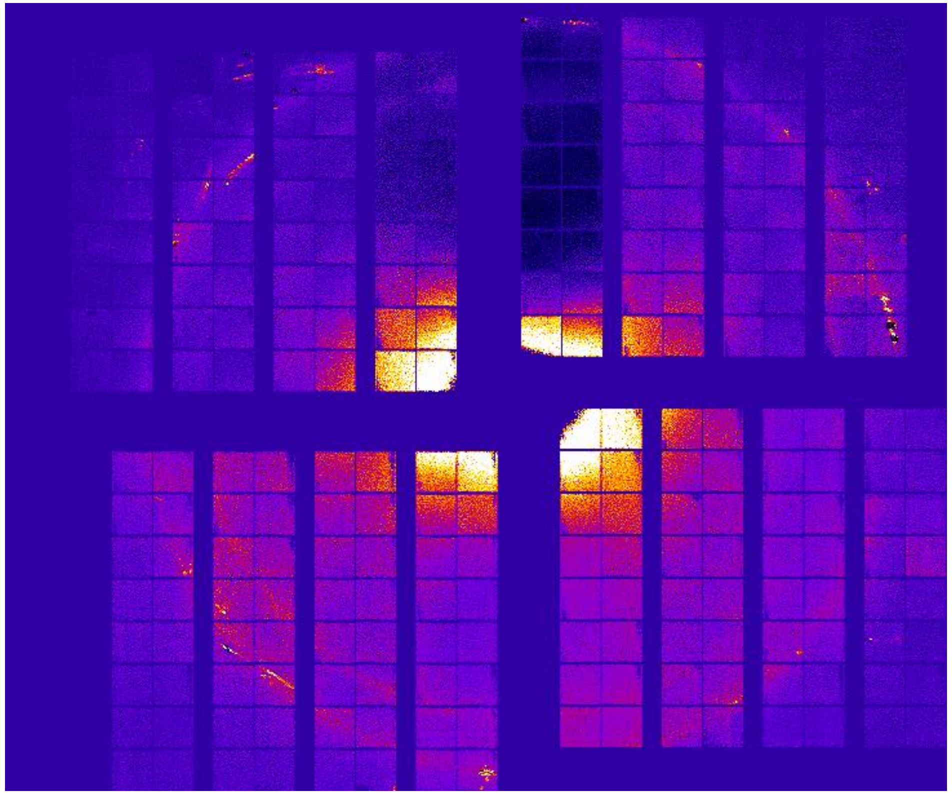
Büttner et al. Nature Materials 20, 30–37 (2021)

MID

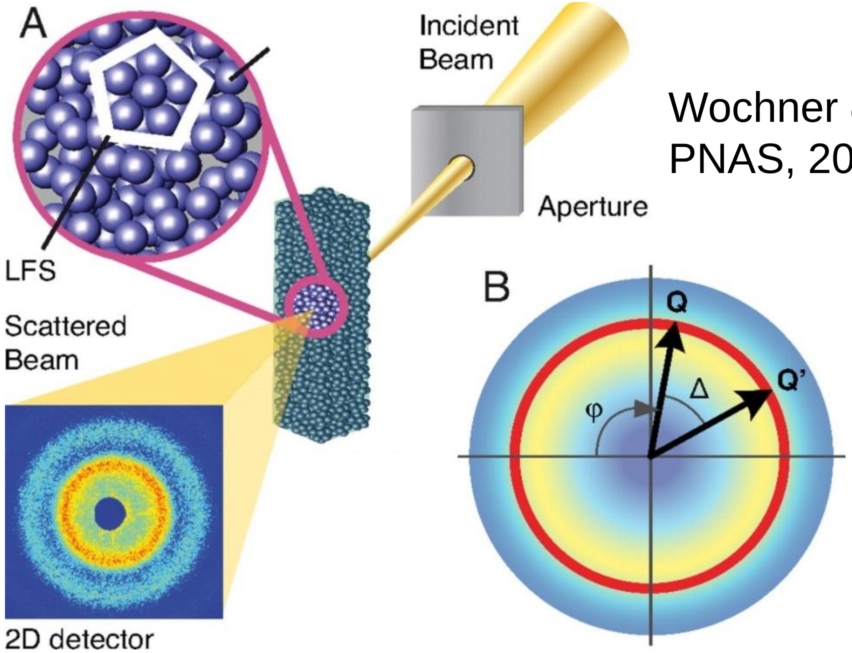
Capturing early stages of crystallization

XCCA: X-ray Cross-Correlation Analysis





4.5 MHz, single shots, 50fs, 9keV, 300nm beam



Wochner *et al.* PNAS, 2009

Spatial Cross-Correlation function

$$\left\langle C^{i,j}(q,\Delta)\right\rangle_M = \left\langle\left\langle I^i(q,\varphi) I^j(q,\varphi+\Delta)\right\rangle_\varphi\right\rangle_M$$

Searching for pre-crystallization features in the data

Johannes Möller *et al.* Phys. Rev. Lett. 132, 206102 (2024)

Examples of X-ray FEL scientific applications

- Ultrafast processes in chemistry and materials

  Make use of fs time resolution w/pump-probe methods

- Biological and soft matter

  Make use of coherence and high intensity

  Serial fs crystallography

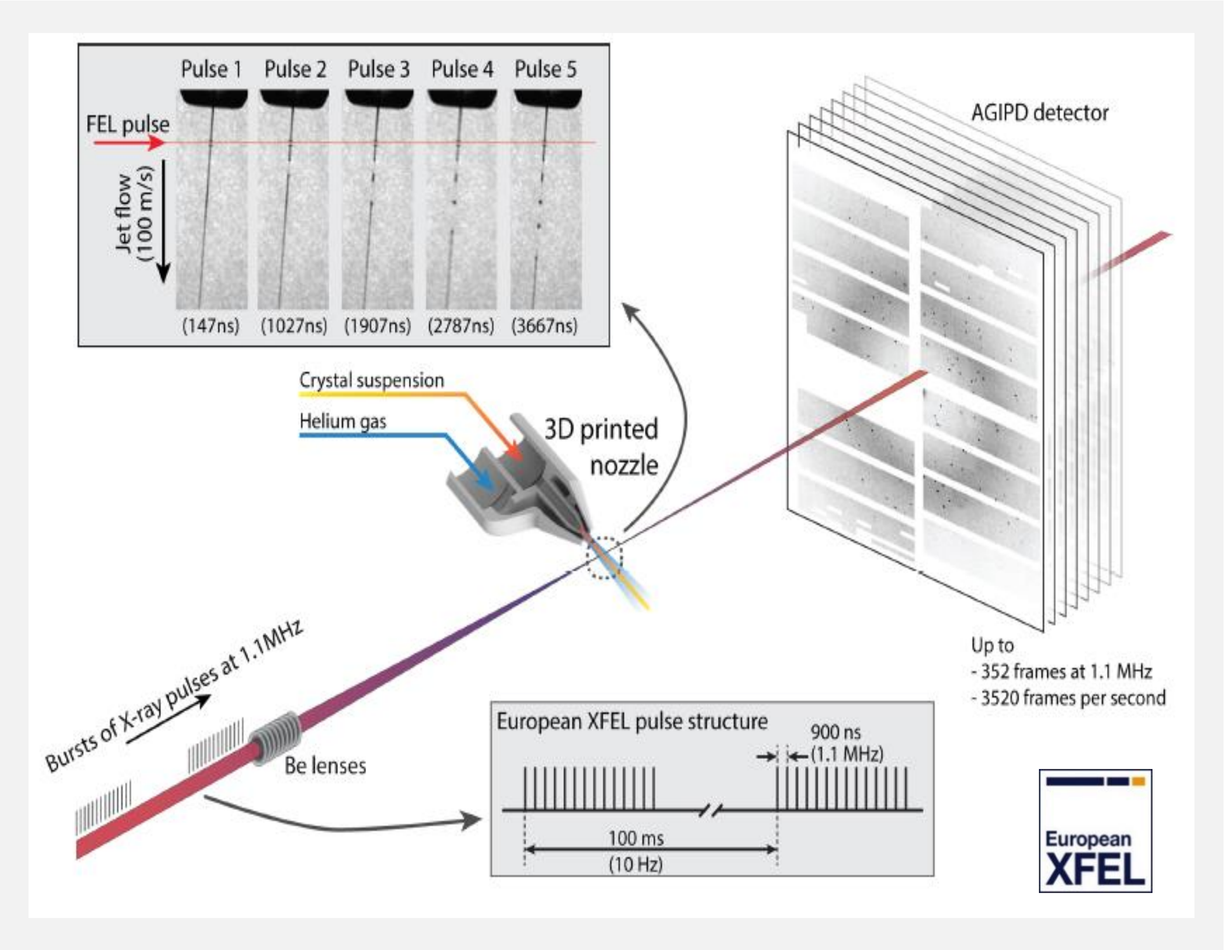
  Photon correlation spectroscopy

- Extreme states of matter

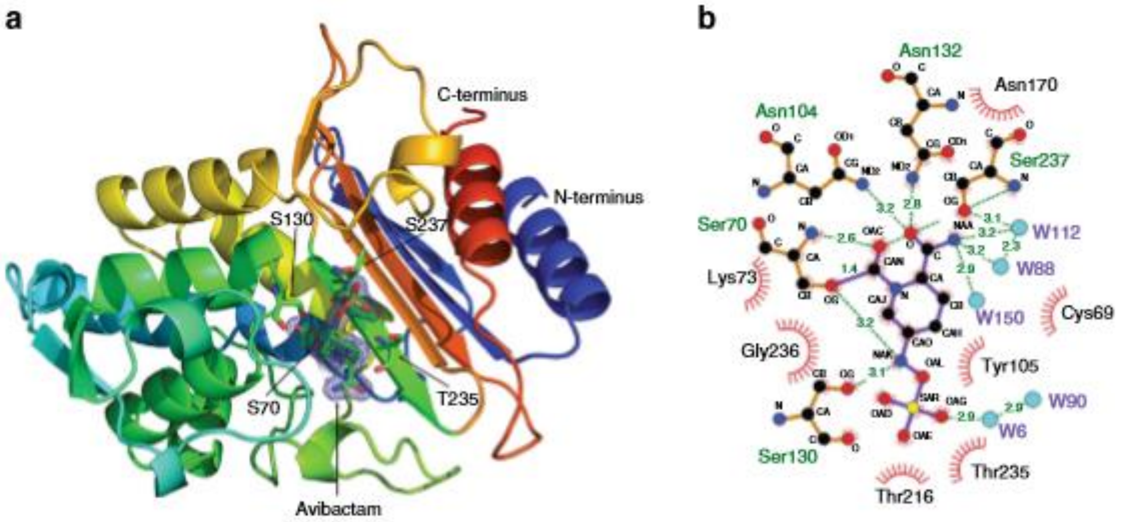
  Make use of very intenste and short x-ray pulses to study short- lived states

MHz serial femtosecond crystallography

SPB-SFX



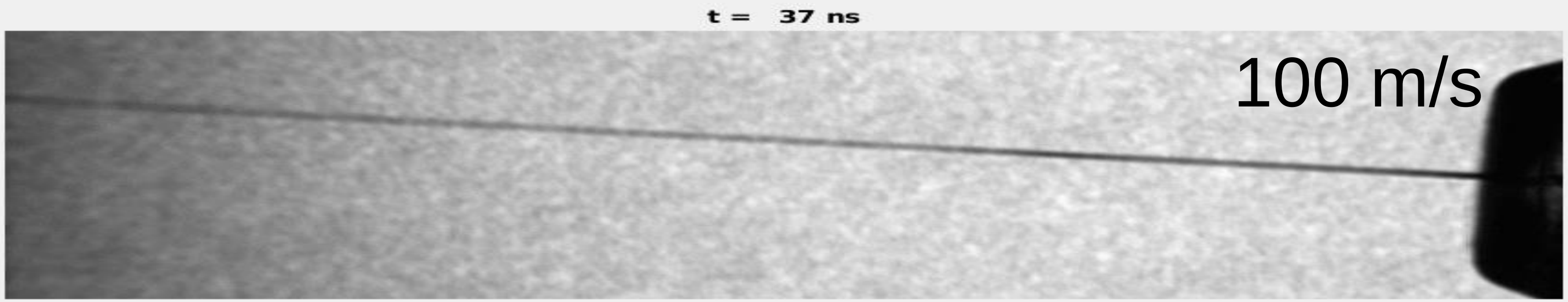
High resolution structure determination from very small, “radiation sensitive” or “dynamic” crystals

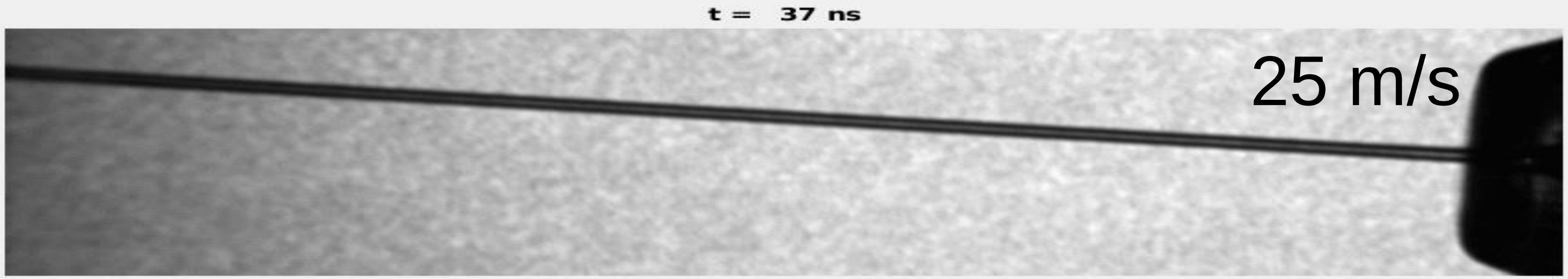


~ 10^5 good patterns per structure

M.O. Wiedorn et al. Nature Comm 9, 4025 (2018)

CFEL-designed fast jets recover in time for the next pulse at 1.1 MHz repetition rate





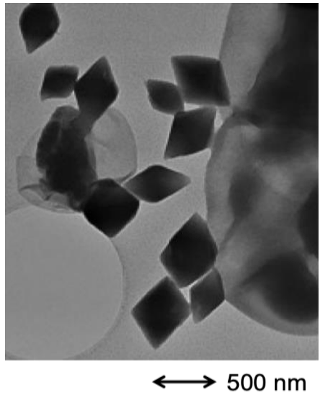
Max Wierdorn, Claudio Stan

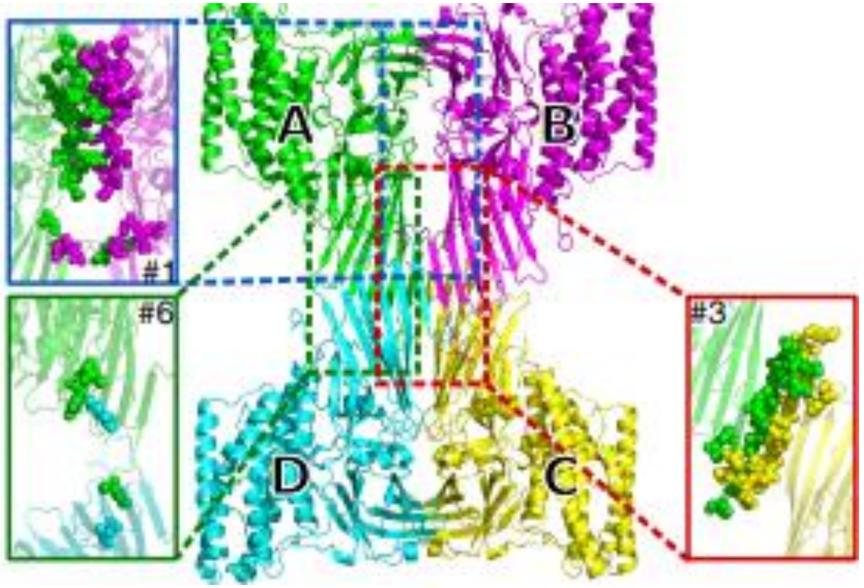
courtesy A. Barty, H. Chapman

nature COMMUNICATIONS

ARTICLE
Check for updates
https://doi.org/10.1038/s41467-022-31746-x
OPEN

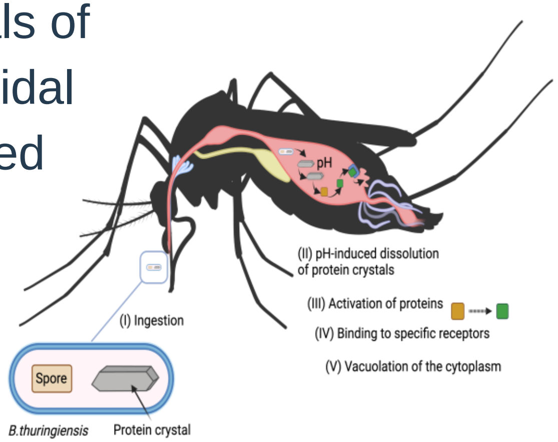
De novo determination of mosquitocidal Cry11Aa and Cry11Ba structures from naturally-occurring nanocrystals





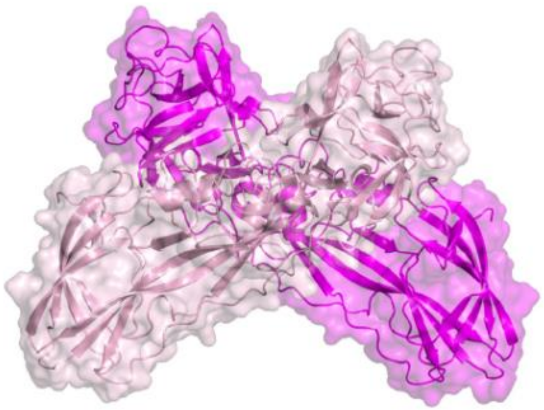
Tetreau Nat. Comm. (2022) 13:4376

- The structure of natural crystals of toxins produced by mosquitocidal Bacillus thuringiensis elucidated using SFX



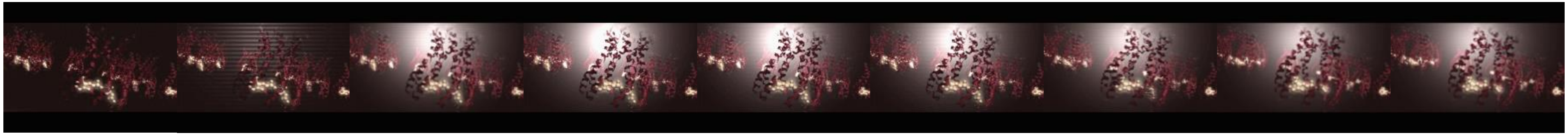
PNAS RESEARCH ARTICLE | BIOCHEMISTRY OPEN ACCE

Structure of the *Lysinibacillus sphaericus* Tpp49Aa1 pesticidal protein elucidated from natural crystals using MHz-SFX

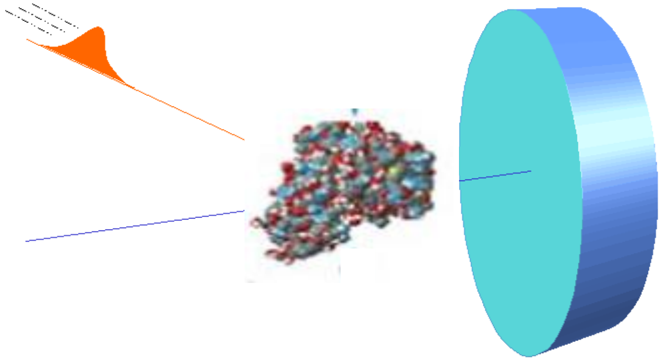


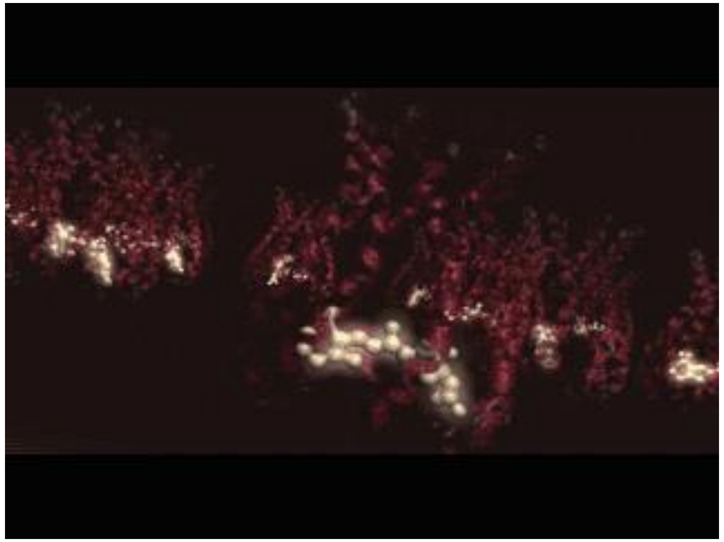
Williamson PNAS (2023), Vol. 120, no. 49

Real time molecular dynamics in native state & transient structures of proteins



Making molecular movies

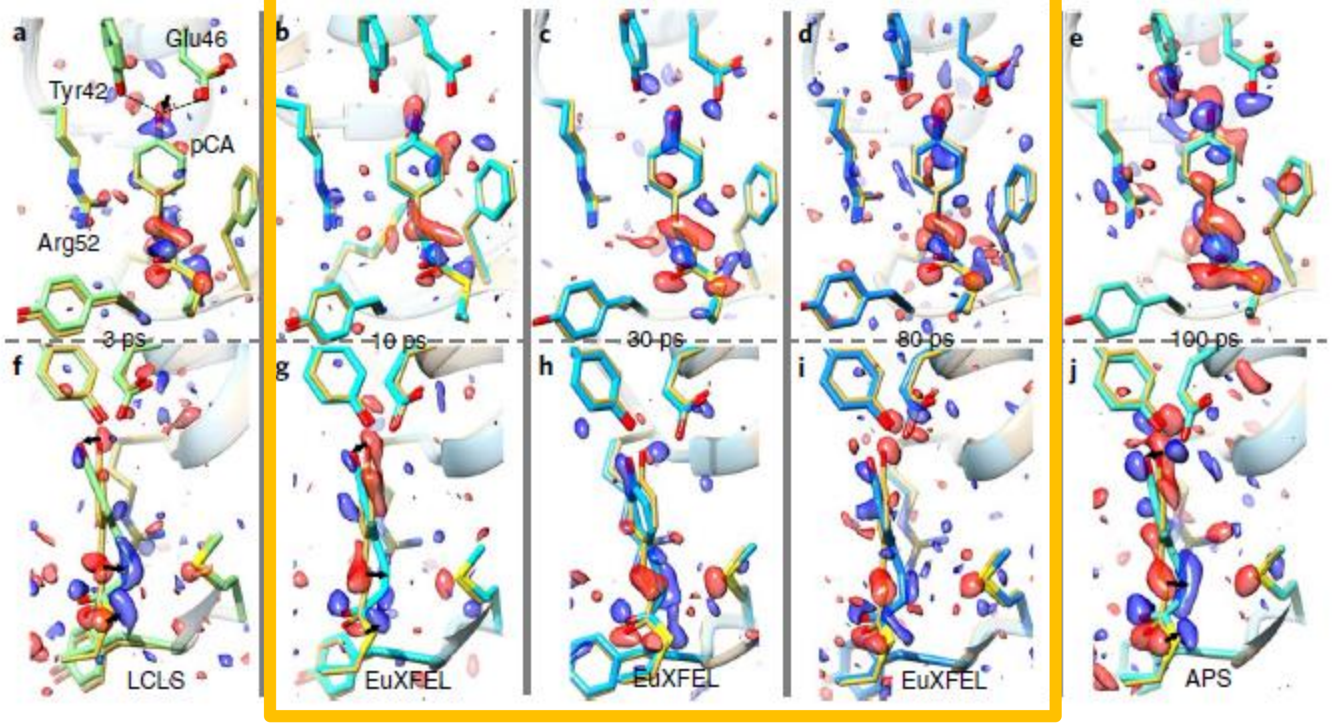




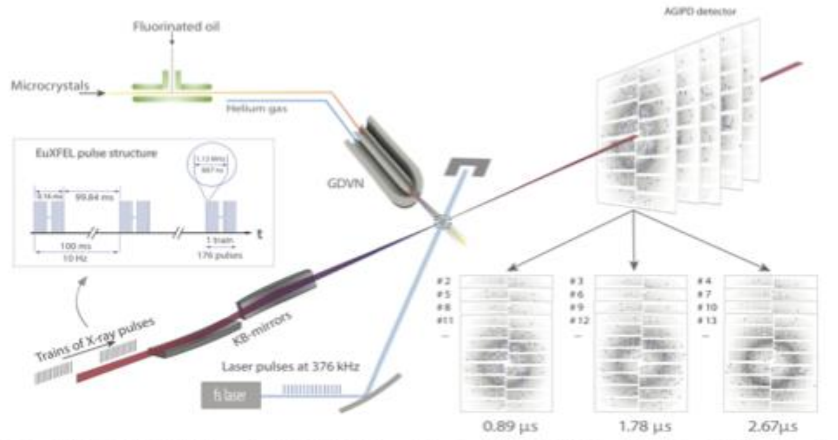
A pump-probe experiment at an XFEL

Start FXE animation

NATURE METHODS **ARTICLES**



■ **SPB-SFX**



Extended Data Fig. 1 | Setup of a MHz TR-SFX experiment at the EuXFEL (modified from Wiedorn et al., 2018). X-ray pulses arrive in 1.13 MHz bursts which repeat every 100 ms. There are 176 X-ray pulses in the burst. The KB-mirror system focuses the X-ray beam to a 2 – 3 µm focal spot. The fs-laser delivers 376 kHz pulses (λ=420 nm, blue) synchronized to the X-ray pulses. The laser focus is 42 µm Ø in the X-ray interaction region (dotted circle). The microcrystals are mixed with fluorinated oil and injected by a GDVN. The jet produced by the GDVN, the laser beam as well as the X-ray pulses precisely intersect. The time-resolved diffraction patterns are collected by the AGIPD. Diffraction patterns with common time-delays were separated based on the pulse ID (see also Fig. 2b) and combined to datasets.

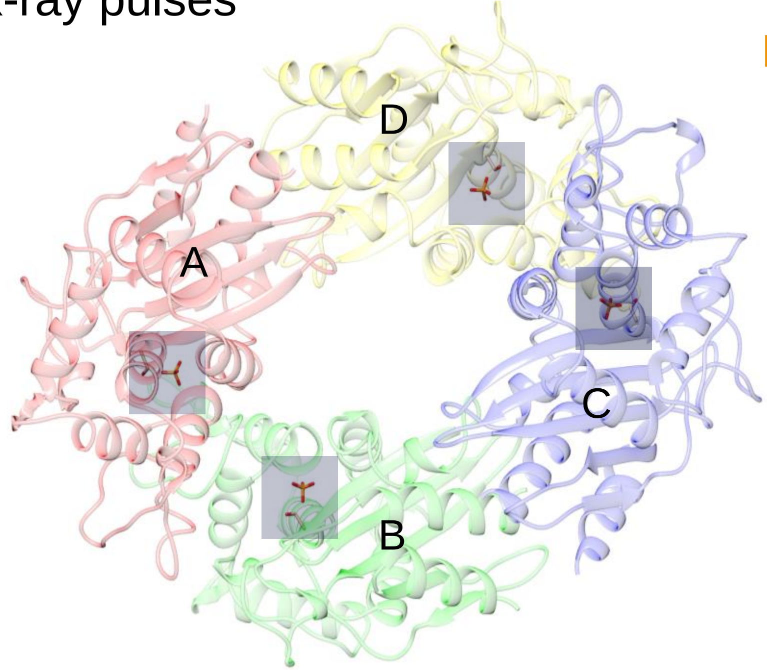
Time series of TRX data from 3 ps to 100 ps at LCLS, EuXFEL and APS. Structures and difference electron density of the photocycle of the photoactive yellow protein

Pandey Nat. Methods (2020), 17, p.73–78

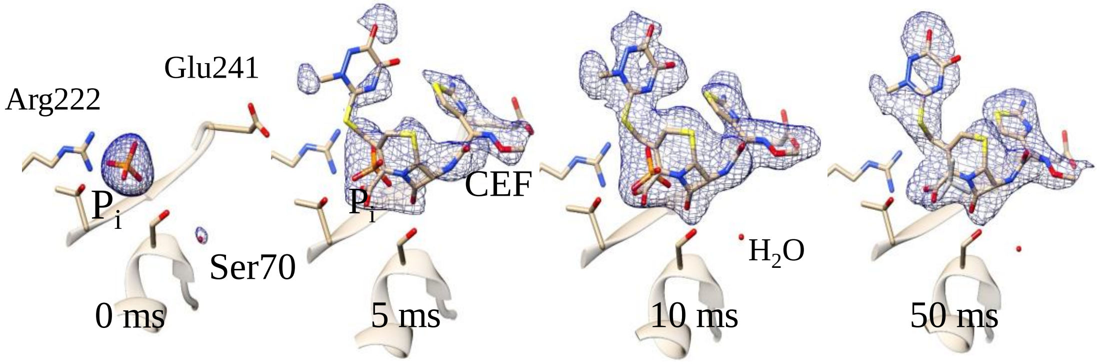
IUCrJ
ISSN 2052-2525
BIOLOGY | MEDICINE

Observation of substrate diffusion and ligand binding in enzyme crystals using high-repetition-rate mix-and-inject serial crystallography

- Substrate binding study to investigate antibiotic resistance in β-lactamase tuberculosis
- Small crystals allow substrate diffusion across crystals in short time and can be interrogated with short, powerful x-ray pulses



- M. tuberculosis β-lactamase
  - chemically modifies β-lactam antibiotics
  - reaction with cephalosporin antibiotics ceftriaxone



formation of the enzyme-substrate complex

SPB-SFX

Pandey IUCrJ (2021) 8, 878–895

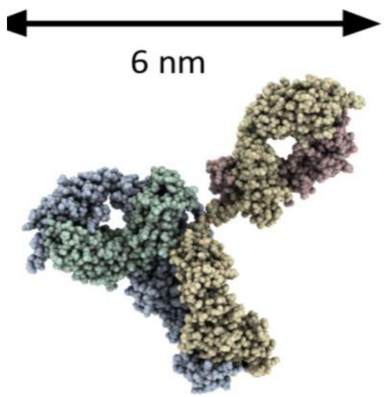
nature communications

Article

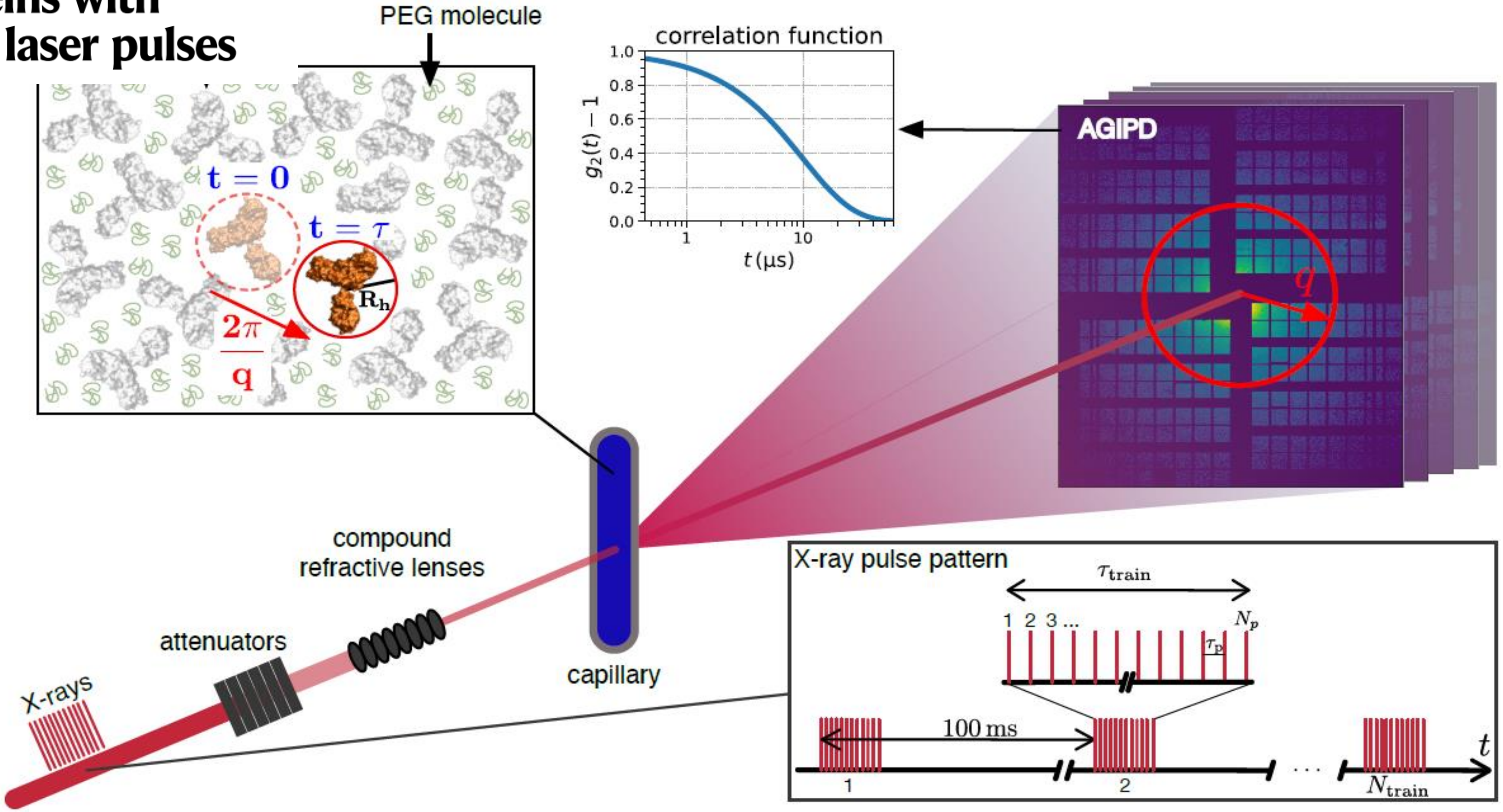
https://doi.org/10.1038/s41467-022-33154-7

Resolving molecular diffusion and aggregation of antibody proteins with megahertz X-ray free-electron laser pulses

MID



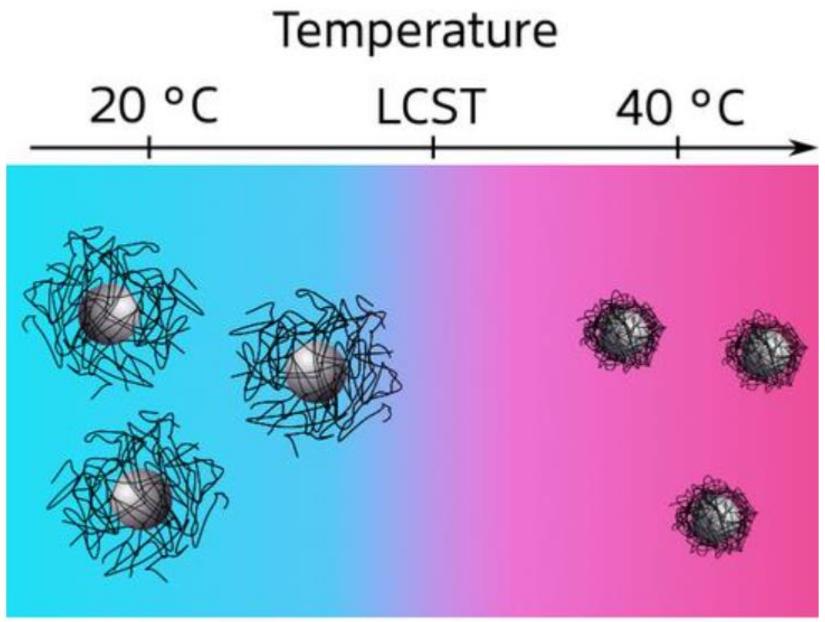
Immunoglobulin

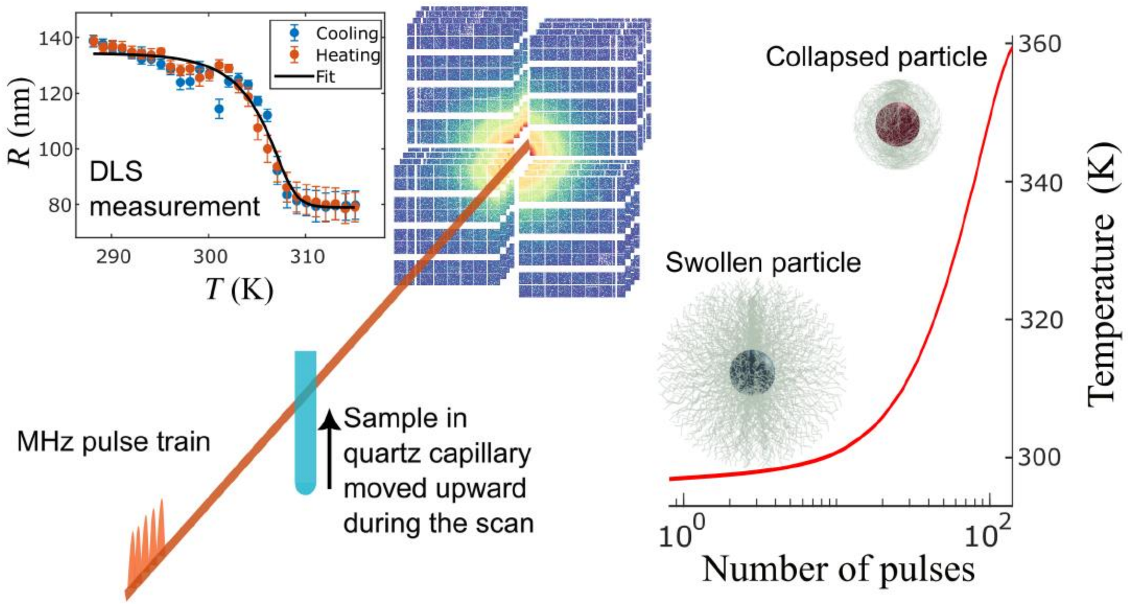


Reiser Nat Comm. (2022) 13, 5528

Real-time swelling-collapse kinetics of nanogels

- PNIPAm: An important nanogel used in medicine to release drugs in a targeted and controlled manner at the desired location in a patient’s body
- Undergoes temperature-induced swelling and collapsing





- Rapid, temperature-dependent changes investigated by X-ray photon correlation spectroscopy (XPCS)
- In contrast to previous studies, the nanogel shrinks significantly faster in the range of 100 nanoseconds but takes two to three orders of magnitude longer to swell

MID

Dallari et al.: Sci. Adv. 10, eadm7876 (2024)

francesco.dallari@ unipd. it

Examples of X-ray FEL scientific applications

Ultrafast processes in chemistry and materials	Make use of fs time resolution w/pump-probe methods
Biological and soft matter	Make use of coherence and high intensity Serial fs crystallography Photon correlation spectroscopy
Extreme states of matter	Make use of very intenste and short x-ray pulses to study short- lived states

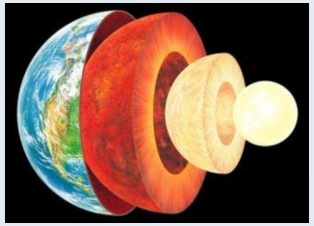
High Energy Density Science at X-ray FELs

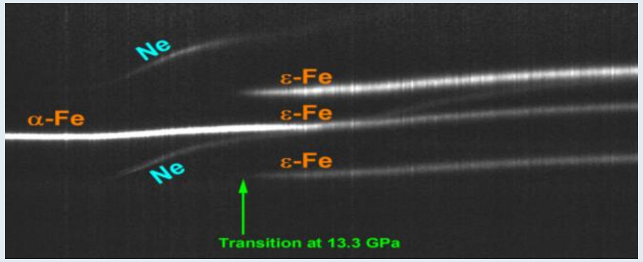
high density $\rho>\rho_0$, T < few eV

solid density $\rho=\rho_0$, T up to keV

High pressure science

Planetary/geo science
Melting curves
Thermal conductivity
Chemical synthesis
Strain-rate dependence
High T superconductivity
Novel materials

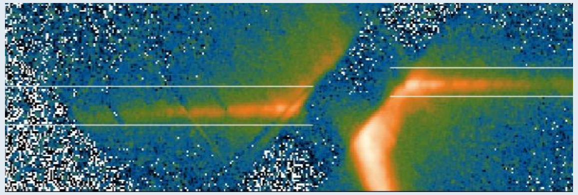




Nanosecond 50 - 100 J laser
DAC, d-DAC, Pulsed laser heated DAC

Relativistic Laser-Plasmas

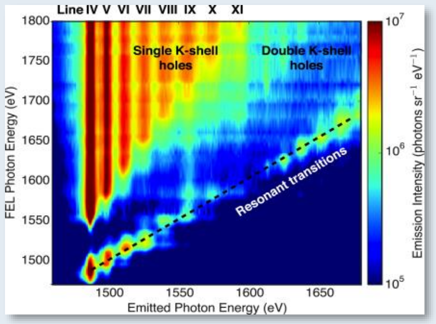
Electron transport,
Instabilities and filamentation,
Ionization dynamics
Particle acceleration



Multi-100 TW fs laser

Intense radiation matter interaction

Transport properties,
Hollow atoms, rates

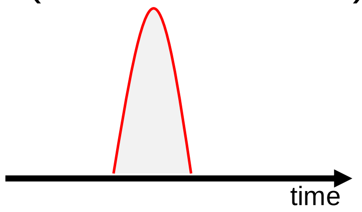


Isochoric X-ray excitation

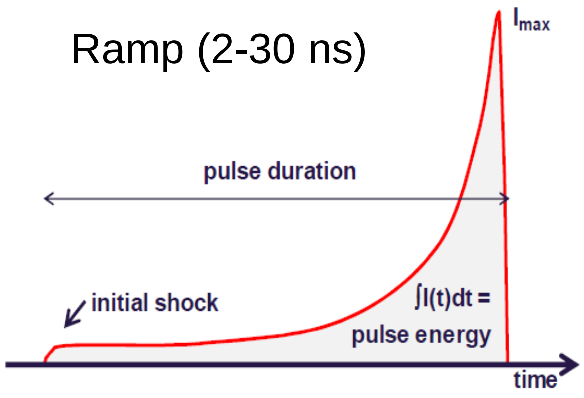
Dynamic compression using high energy lasers

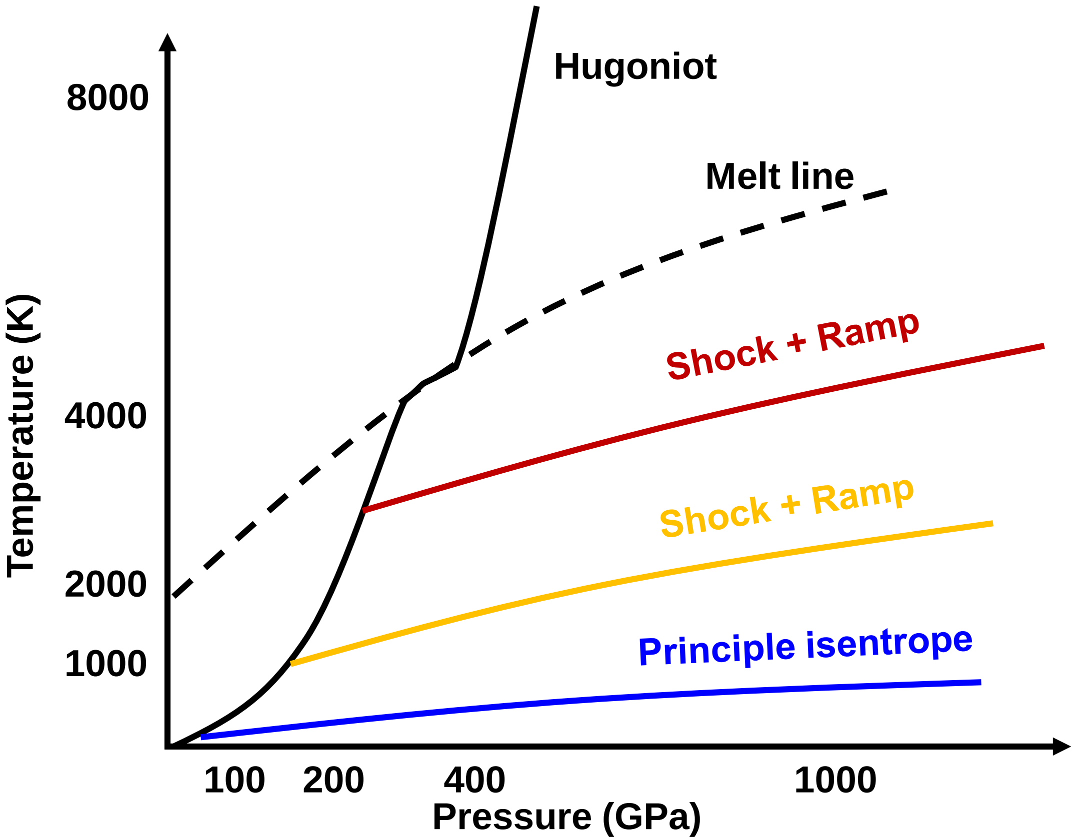
- Use laser to drive a 'shock'
  - **Ablation of matter creates shockwave**
  - **Shaped laser pulses to approach quasi-isentropic ('shockless') compression**
  - **Can reach higher pressures**
  - **Dynamics: strain rate can be varied**
  - **Short-lived (10s of ns)**

Shock (sub-ns to few ns)



Ramp (2-30 ns)





Observing new states of matter

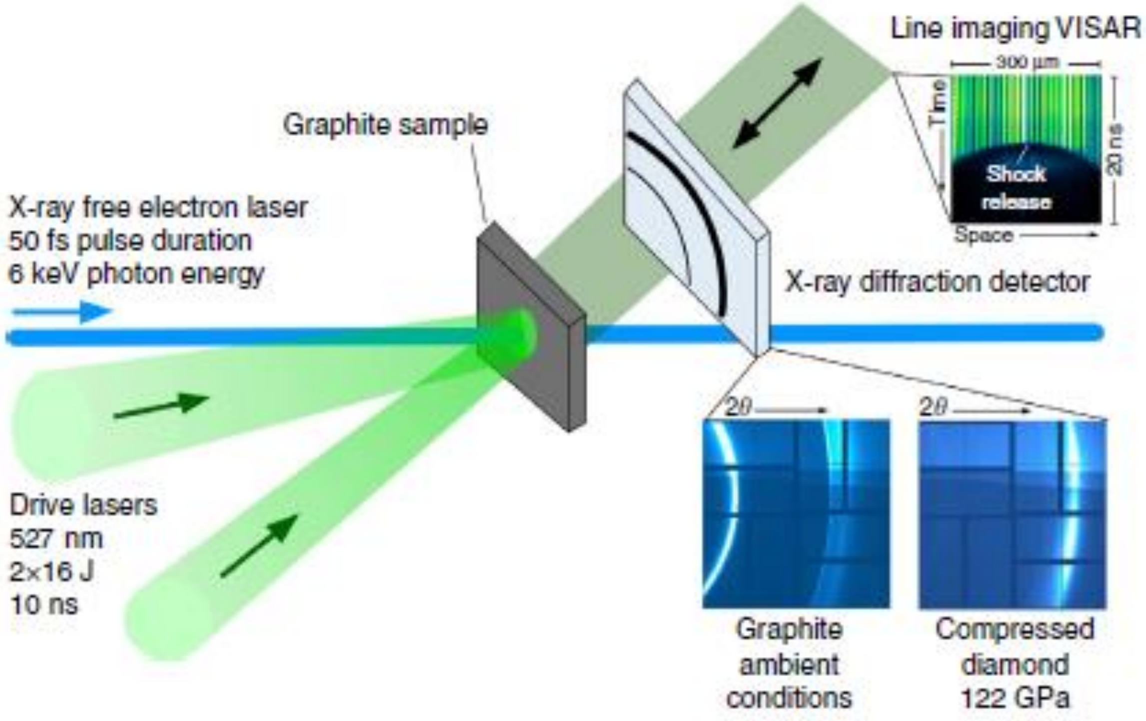
ARTICLE

Received 5 Apr 2015 | Accepted 5 Feb 2016 | Published 14 Mar 2016 DOI: 10.1038/ncomms10970 OPEN

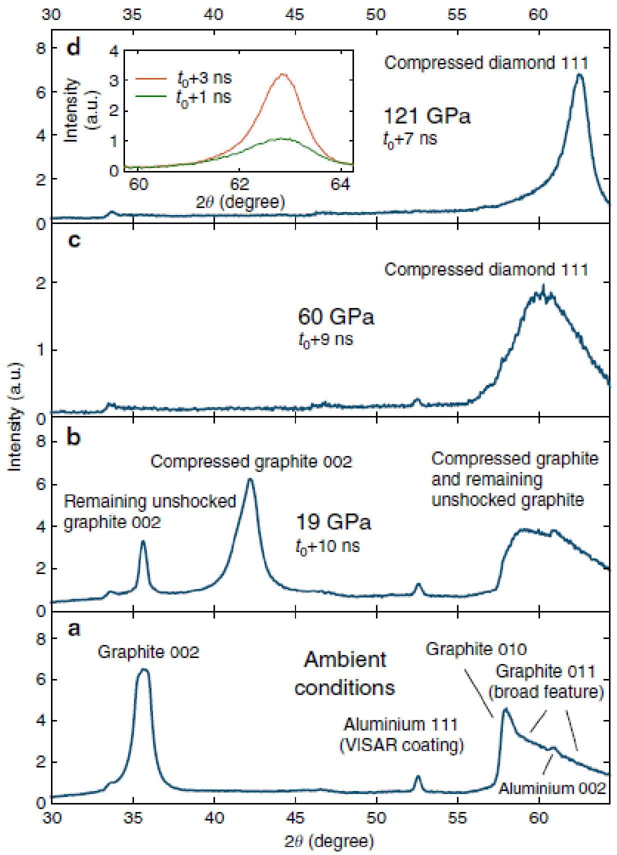
Nanosecond formation of diamond and lonsdaleite by shock compression of graphite

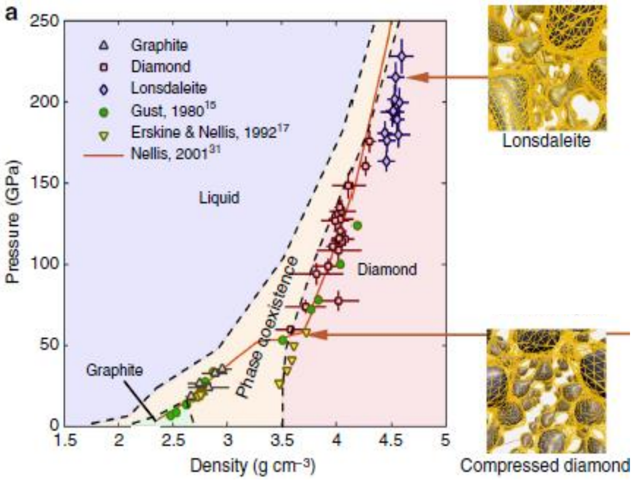
D. Kraus[1], A. Ravasio[2], M. Gauthier[2], D.O. Gericke[3], J. Vorberger[4,5], S. Frydrych[6], J. Helfrich[6], L.B. Fletcher[2], G. Schaumann[6], B. Nagler[2], B. Barbrel[1], B. Bachmann[7], E.J. Gamboa[2], S. Göde[2], E. Granados[2], G. Gregori[8], H.J. Lee[2], P. Neumayer[9], W. Schumaker[2], T. Döppner[7], R.W. Falcone[1], S.H. Glenzer[2] & M. Roth[6]

D. Kraus et al., Nat. Comm. 7:10970 (2016)



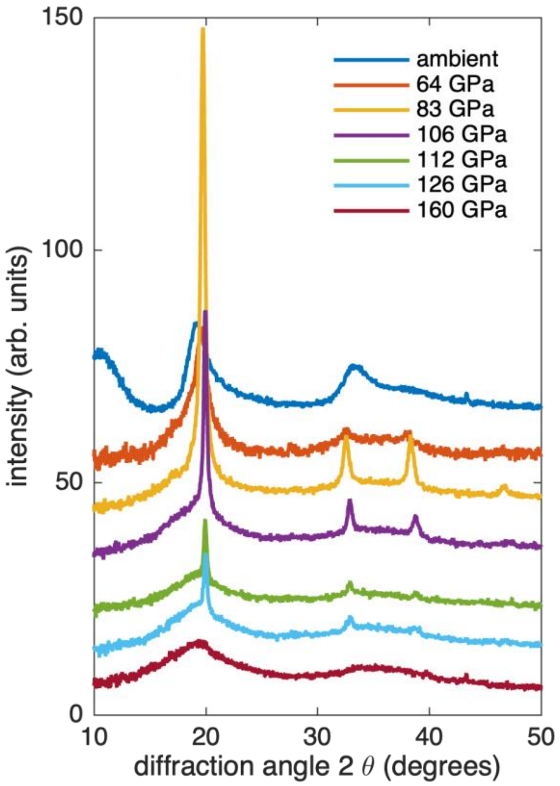
- LCLS





- Observed diamond and lonsdaleite formation under dyn. shock conditions

HED/HIBEF@EuXFEL – first data and results

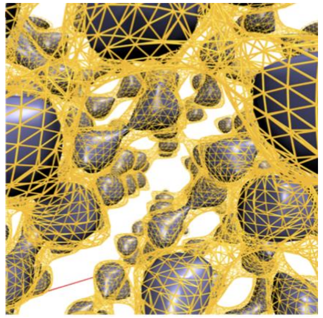


Glassy carbon

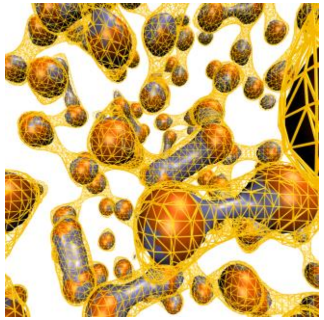
Diamond

Liquid + diamond

Liquid



Compressed diamond



Liquid carbon

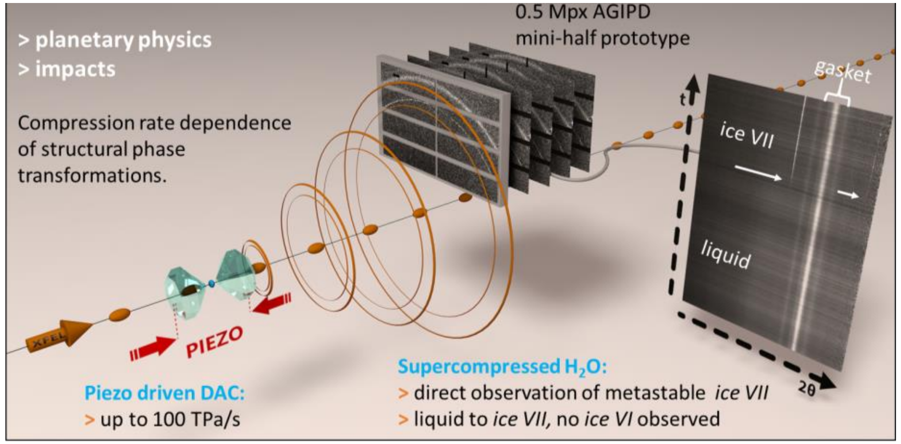
Evolution of carbon's structure under laser-driven shock compression for laser power of increasing intensity

PI Dominik Kraus, Rostock Uni

HED

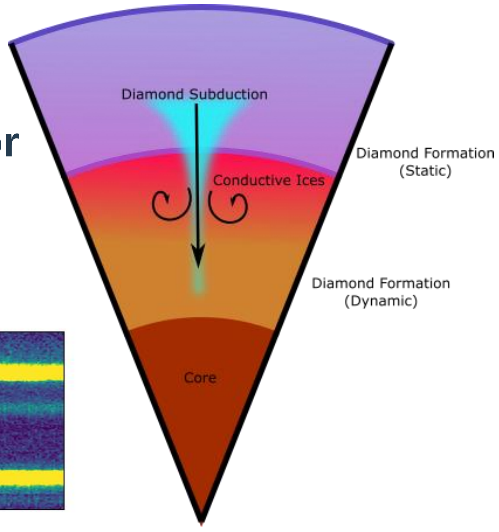
European XFEL

Metastable phases of Ice – compression rate dependence of structural phase transitions

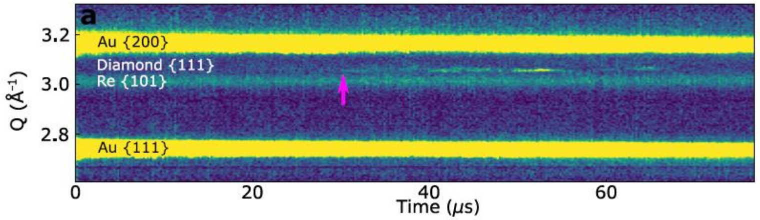


PI Amy Jenei, LLNL and Hanns Peter Liermann, DESY

Diamond Precipitation from Hydrocarbons at Icy Planet Interior Conditions



Frost et al. Nature Astronomy, 2023



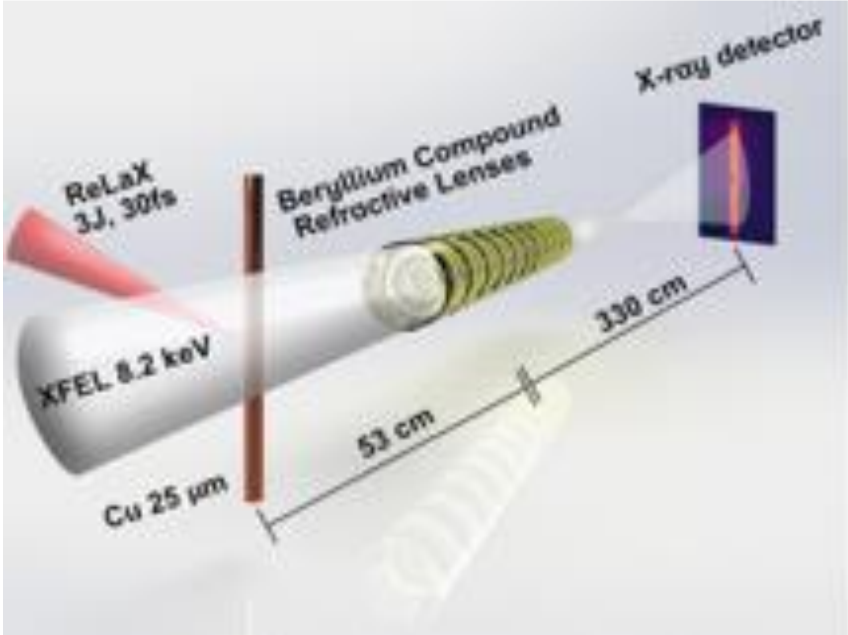
HED-HIBEF High-Energy Density Science

HIBEF HELMHOLTZ INTERNATIONAL BEAMLINE FOR EXTREME FIELDS

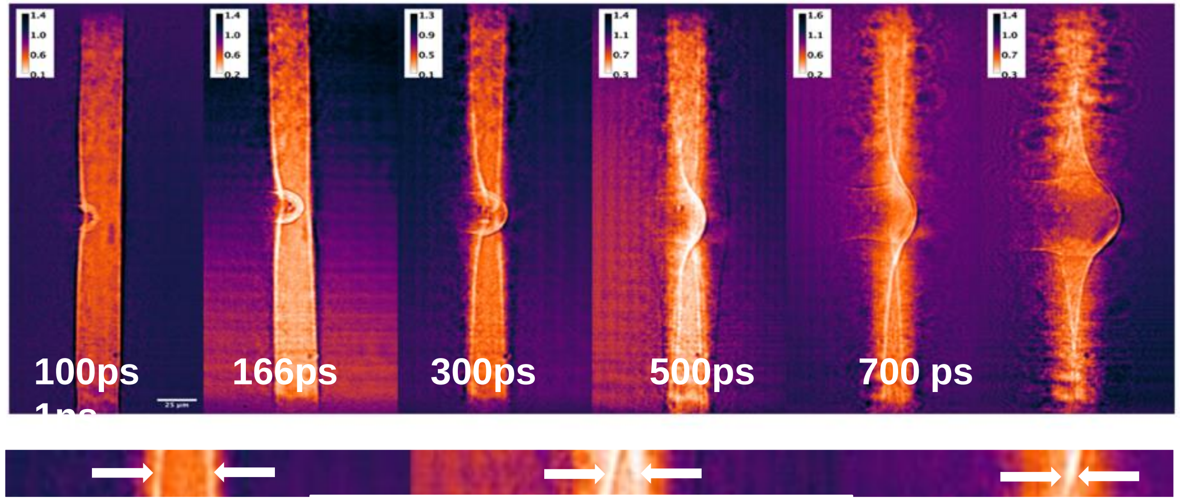
HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

Imaging cylindrical compression of thin metal wires at fusion relevant conditions

Driven by ReLaX



Wire implosion driven by return current heating



Cylindrical shock wave

25 µm Cu wire by a 3 J, 30 fs laser pulse

- Irradiation of plain thin Cu wire generates a converging cylindrical shock
- Shock travels towards the wire axis, reaches at the convergence point a compression factor of 9 and P > 800 Mbar (simulations)

Garcia et al., Nature Comm, in print

Outline

- Introduction
- Free-electron laser basics
- X-ray FEL facilities

  – break –

- Ultra fast processes in chemistry and materials
- Biological and soft matter
- Extreme states of matter
- **Future**

From ‘first’s’ to applications

- X-ray FELs are operational since ~ 15 yrs.
- Many new observations for the interaction of intense (energy & ultrashort) x-ray pulses with matter have been made. Large number of highly visible papers published.
- A next step will be to develop applications which can be used by a broader science community. Potential candidates are:
  - **Structure and dynamics of proteins and similar bio-molecules**
  - **Chemical reaction dynamics : Intermediates & pathways**
  - **Materials science: visualization of irreversible, stochastic, rare, processes**
- FEL experiments are complex → still small community, how can we make experiments simpler
- One can expect still lot of new results and new methods to come out of the (still young) FEL research

Most important science drivers

- Capability to measure ultrafast dynamics will allow to determine the response of photo-active catalysts to light excitation → develop new compounds/materials for solar energy conversion

- Capability to determine atomic structures of bio and biochemical objects will allow to better understand their function → develop new drugs and disease treatments

- Capability to observe new and yet undiscovered states of matter and their structural properties will allow to better understand fundamental physical properties of matter and to detect new materials properties → develop new materials and processes

- Possibility to observe non-linear x-ray processes will allow to develop new x-ray spectroscopy methods → enable new methods to study excited matter

Summary

- X-ray FELs are new research infrastructures providing research opportunities complementary to well-established SR sources.
  - **Femtosecond time resolution**
  - **High pulse energies enabling single shot experiments and non-linear x-ray scattering**
  - **High coherence facilitating imaging and correlation spectroscopy experiments**
- Since 2017 5 hard x-ray facilities are operational. This will broaden the experiment and user base and will provide better access and research opportunities.
- Many applications so far concern basic science, many are exploratory. With a better under-standing of what is possible using FELs and an increase in available beam-time an expansion of scientific applications is expected.
- The field is still very young and dynamic → new developments are very frequent: Stay tuned !

Additional slides

FEL parameters

- Full 3-dimensional FEL theory is extremely demanding
- Simplified 1-dimensional approach
- Pierce (FEL) parameter ρ

$$\rho = \left(\frac{I A_{JJ}^2 K^2 \lambda_{rad}^2}{I_A 32\pi^2 \gamma^2 \varepsilon_n \beta_f} \right)^{1/3}$$

$\rho \sim 2\text{-}5\times10^{-4}$ (0.1 nm)

- **Number of cooperating electrons N_c**

$$N_c = (N_e \lambda_{rad}/L)/2\pi\rho$$

$N_c \sim 1/500 \times N_e$ (0.1 nm)

- **Power gain length L_G**

$$L_G \approx \lambda_{und}/4\pi\rho$$

- **Saturation length L_{sat}**

$$L_{sat} \approx 10 \times L_G$$

- **Radiated energy E_{sat}**

$$E_{sat} = \rho N_e E_{e,kin,total}$$

- **Radiation bandwidth $\Delta\lambda/\lambda$**

$$\Delta\lambda/\lambda \approx 2\rho$$

FEL process is critically sensitive to electron parameters

- Transverse emittance (diffraction limit)

$$\varepsilon < \lambda_{rad} / 4\pi$$

ε~8 pm (λ=0.1 nm) → ε_n~0.22×10^{-6} mrad (15 GeV)

- Energy spread

$$\sigma_e / E < \rho$$

→ ~10^{-4}

- Gain length

$$L_G < L_{Rayleigh} = 2\pi\sigma_{rad}^2 / \lambda_{rad}$$

In general,
small emittance,
small energy spread,
and
high peak current
are needed
simultaneously.

FELs vs. storage rings

Storage rings

- Electron bunches are circulated
- Stable operation conditions
- Many science instruments, often dedicated to specific methods or science applications
- Large number of exp.s and users per year
- Average spectral brightness is key property
- Specialization on low/medium/high energy facilities with resp. spectral coverage
- ~50 yrs development

FELs

- Normally electron bunch is used only once
- Single path means instable operation conditions
- Few science instruments, often designed for multi-purpose applications
- Typ. only many 10s of experiments per year
- Peak spectral brightness is key property
- Specialization on low/medium/high energy facilities with resp. spectral coverage
- High repetition and multi-beamline facilities will broaden access and average flux properties
- ~10 yrs development

Comparison of SR and FEL x-ray properties

X--ray pulse properties	SR x-rays	X-ray FELs
Pulse energy	pJ - nJ	µJ - mJ
Photons per pulse (~10 keV)	10^5 – 10^8	10^{10}- 10^{13}
Pulse duration / time-resolution	30 - 100 ps	2 - 100 fs
Coherence degree (~10 keV)	< 10^{-3}	0.1 - 1
Rel. bandwidth (Undulator)	10^{-2}	10^{-3}
Spectral characteristics	continuous	spiked
Divergence (hor. × vert.)	1000 × 10 µrad^2	< 2 × 2 µrad^2
Repetition rate	~100 MHz	~MHz | 10 - 100 Hz
Peak brilliance	~10^{24}	~10^{33}
Average brilliance	~10^{20}	~10^{24} | ~10^{22}

Comparison of SR and FEL experiments

Synchrotron experiments	FEL experiments
Typically continuous illumination until desired S/N-ratio is reached (ms to hrs)	Typically single pulse illumination and collection of few – 100s pulses to reach desired S/N
Continuous illumination facilitated by high stability and linear intensity dependence	Single-shot illumination due to strong fluctuations of pulse properties and possibility of non-linear dependence
Single pulses typically not meaningful	Single pulses typically give meaningful results
Coherence degree small and requires to limit phase space/flux to reach good degree	High coherence degree enables use of full pulse
50 ps- to µs- time resolution; ps to hrs probing	fs- to ps- time resolution; fs to ms probing
Samples typically probed non-destructively	Samples often degrade and need to be refreshed (OL-pumping; irreversible; damage) Can use X-ray pulse to pump sample
OL most often cw; few sync. OL systems	Complex OL for pump-probe experiments
Many (O(10-30)) parallel experiments	Few (O(1-5)) parallel experiments