

# X-ray Free Electron Lasers & ultrafast x-ray science

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# Outline

**Break @ 17h10-17h20**

- What is an X-ray Free Electron Laser and how does it work ?
- Ultrafast science and ultrafast experiments
- X-ray Free Electron Lasers around the world
- What are the new science opportunities ? Examples of recent results
- Ongoing developments: Where are we heading to ? What new science ?
- Outlook

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# 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 installations; applications u. study



→ **X-ray** ultrafast methods using scattering, microscopy, spectroscopy; non-linear methods

## Visible Laser radiation

- Commercial or home-built systems widely distributed, large facilities
- Huge community
- Attosecond & non-linear techn.

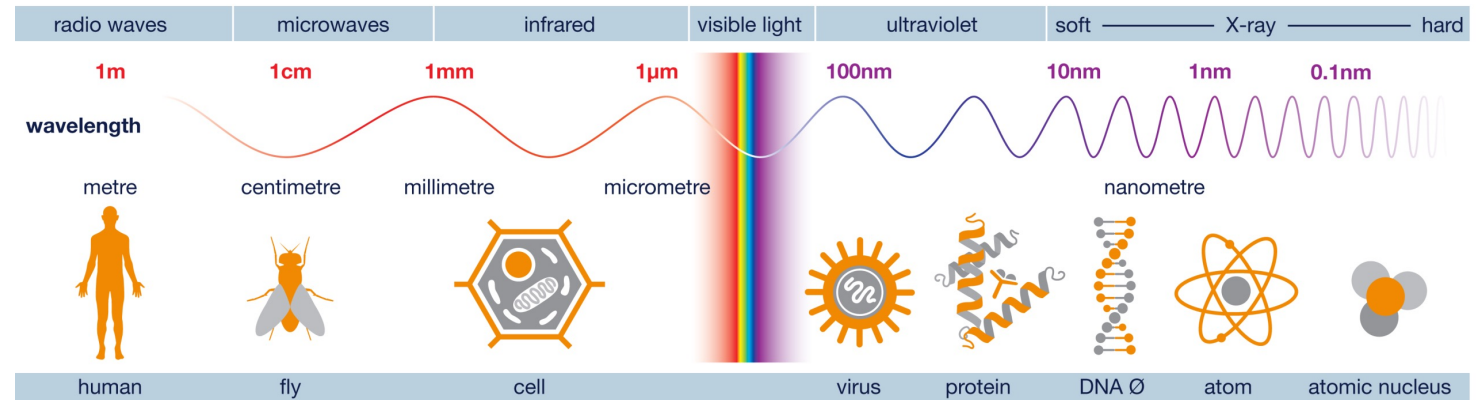


→ **Light** ultrafast and non-linear methods using microscopy or spectroscopy

# XFEL radiation is very different from Laser radiation ...

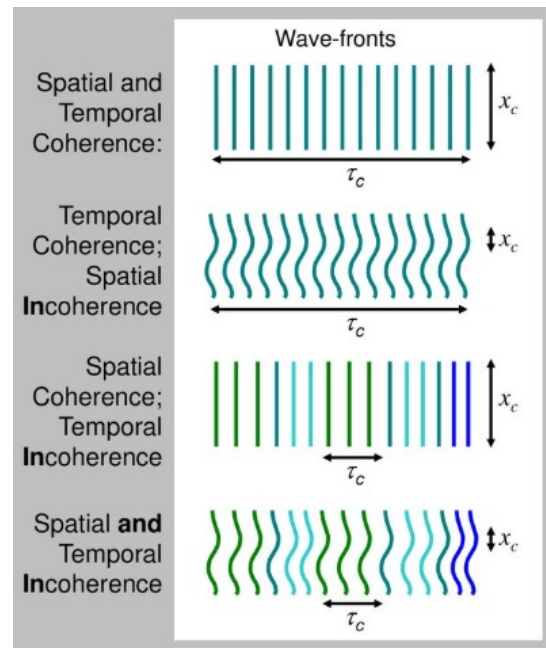
## Wavelength

- orders of magnitude smaller



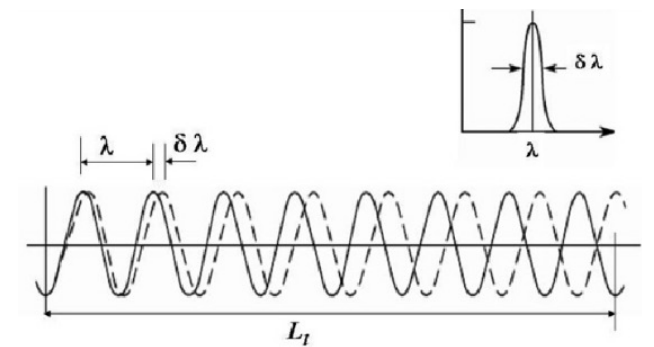
## Coherence

- spatial coherence: similar
- temporal coherence: much lower

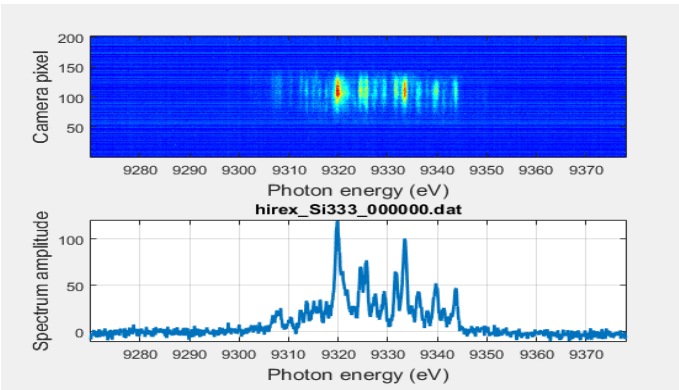


## LASER

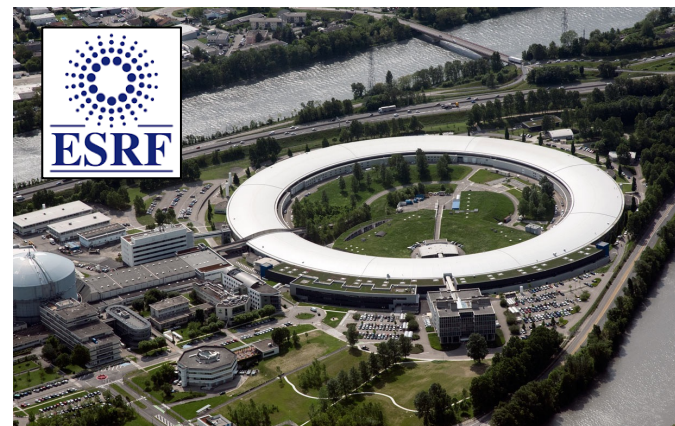
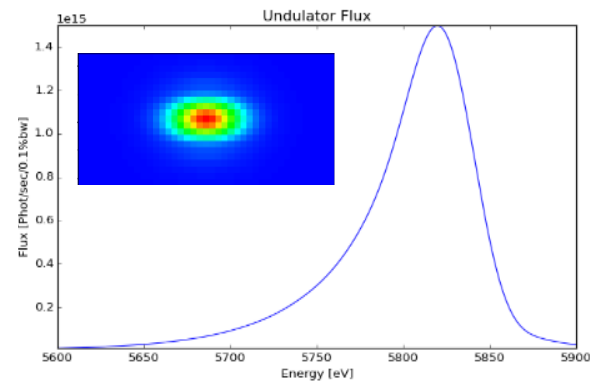
## XFEL



# XFEL radiation is very different from Synchrotron radiation ...

**FEL** $10^{13}$  Photons

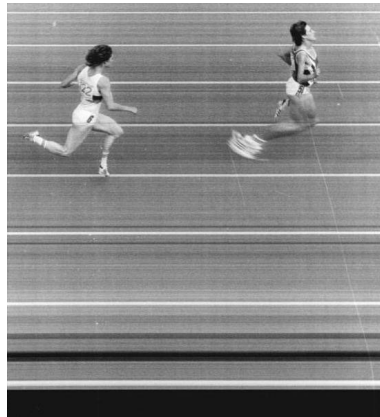
2-100 fs

**Undulator** $10^9$  Photons

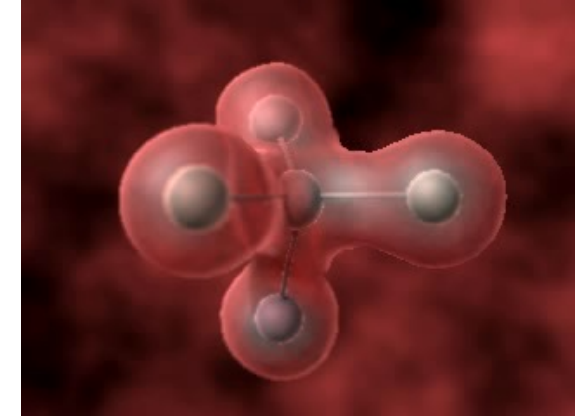
100 ps

time

# XFELs can take snapshots of atomic movements ...



© MIT Museum



|                    |          |               |                                                 |
|--------------------|----------|---------------|-------------------------------------------------|
| Speed of subject   | 10 m/s   | 1000 m/s      | 1000 m/s<br>(rattling speed of atoms)           |
| Picture resolution | 10 mm    | 1 mm          | 1/10 000 000 mm                                 |
| Time resolution    | 1/1000 s | 1/1 000 000 s | 1/10 000 000 000 000 s<br>(or 100 femtoseconds) |

fs pulse: a probe of atomic motion (100fs) .... and charge migration (fs)

hard X-rays: atomic resolution, chemical selectivity, bulk sensitivity



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

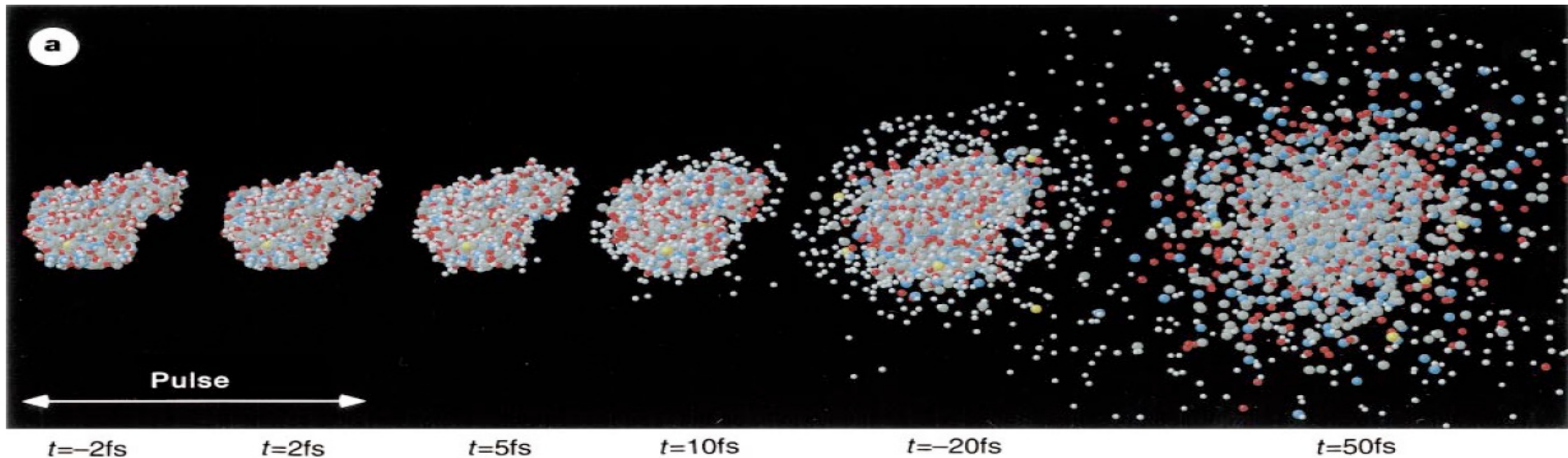
Can we measure phonon dynamics in the time domain ?

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

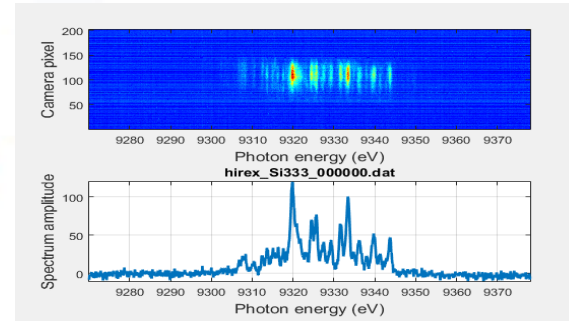
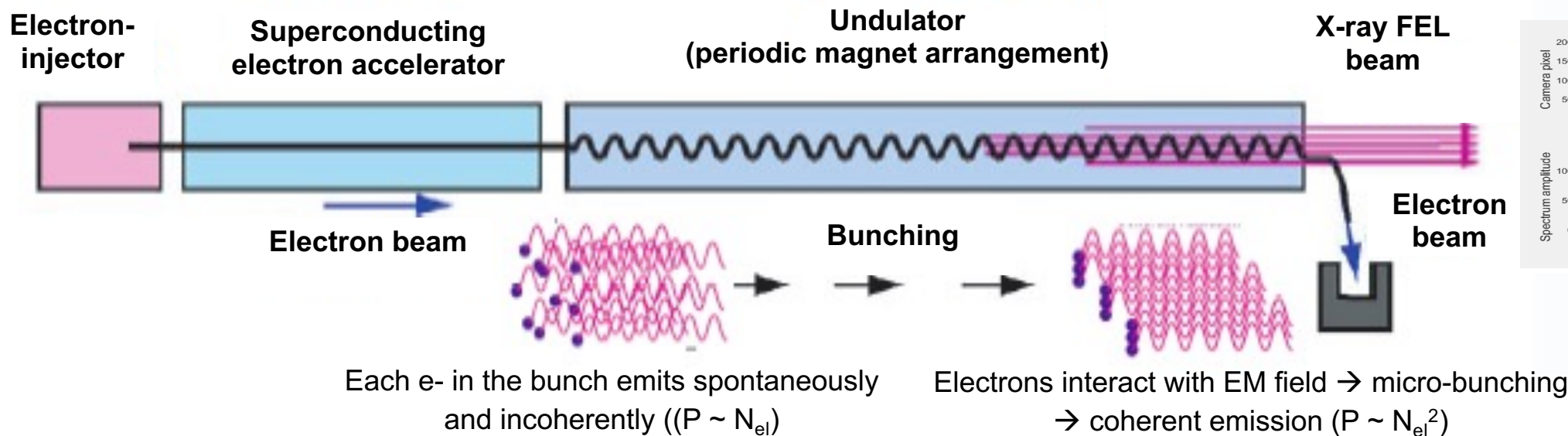
R. Neutze et al, Nature 406, 752 (2000)



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

# X-ray Free-Electron Laser: SASE radiation

## Self-Amplified Spontaneous Emission (SASE)



- ❖ 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 spatial coherence (<10 keV); Limited temporal coherence
  - ❖ Single pass generation / fluctuations

# Differences with respect to undulator/synchrotron radiation

■ Resonance condition is equal

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

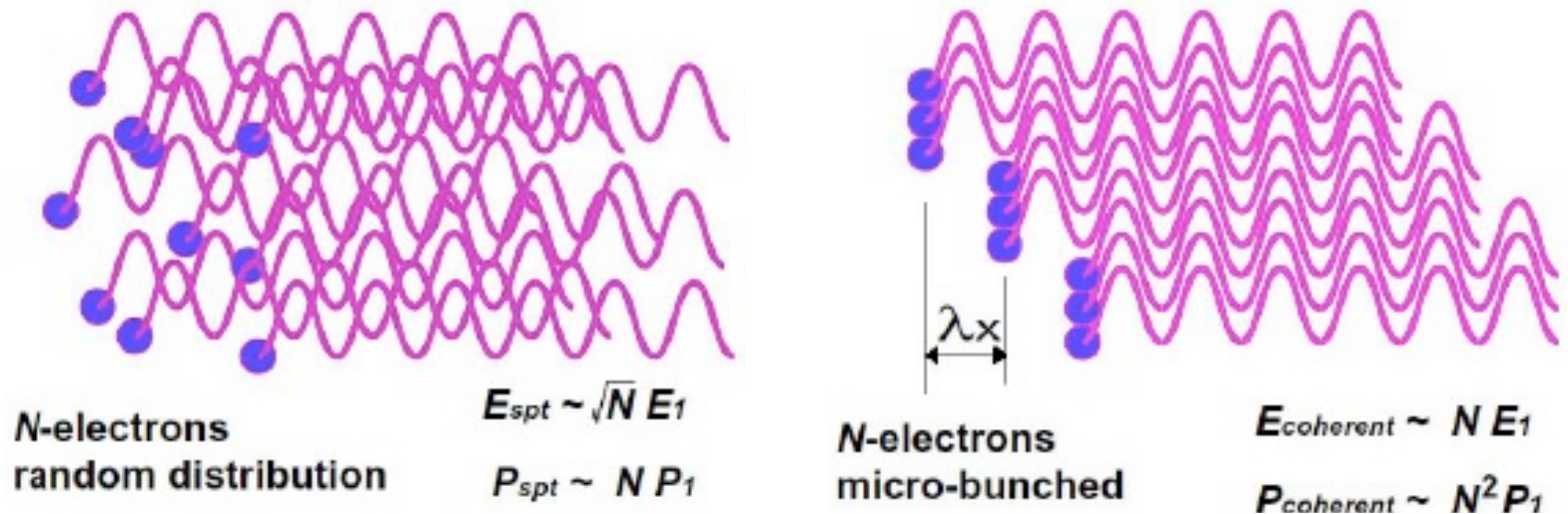
■ Much tighter electron phase space parameters

■ **Extremely small emittance**

■ **Bunch compression**

| Parameter                           | SR                                 | FEL               | factor |
|-------------------------------------|------------------------------------|-------------------|--------|
| Transverse emittance                | 1 nm (x) ( $\rightarrow$ 0.1(x,y)) | 0.015 nm (x,y)    | 40 (7) |
| Longitudinal e <sup>-</sup> density | 10 nC / 30 mm                      | 1 nC / 30 $\mu$ m | 100    |

■ Coherent emission process



# Bunching the electrons

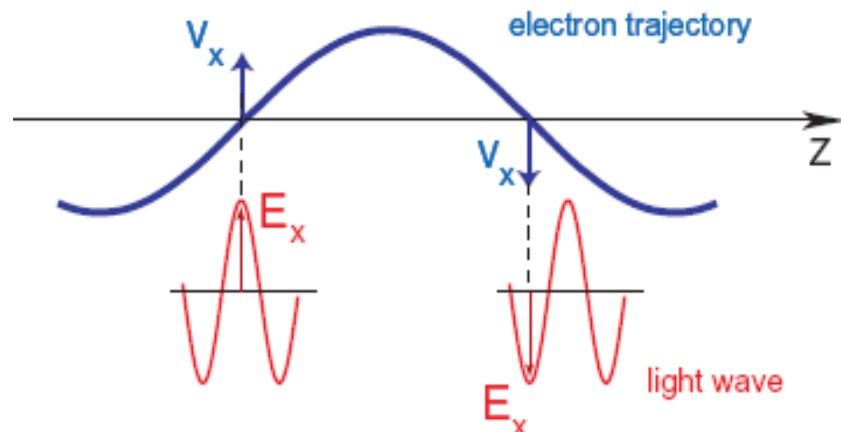
- By electro-magnetic forces arising from interaction with the co-propagating radiation field

- Energy change due to x-ray field:

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

- SASE resonance condition:

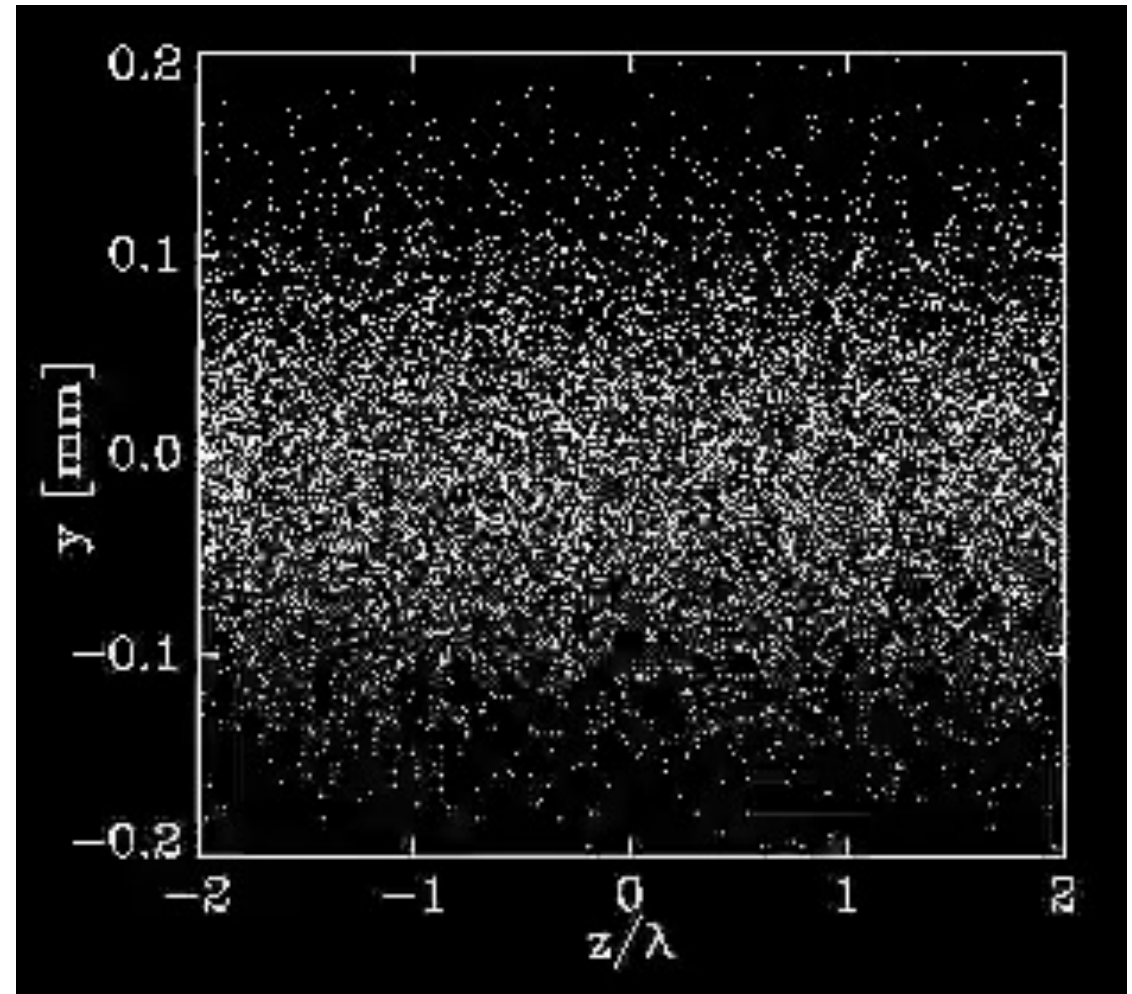
Keep phase between  $v_x(t)$  and  $E_x(t)$  const.



*GENESIS* -

simulation for TTF parameters

Courtesy - Sven Reiche (DESY, now PSI)



# Outline

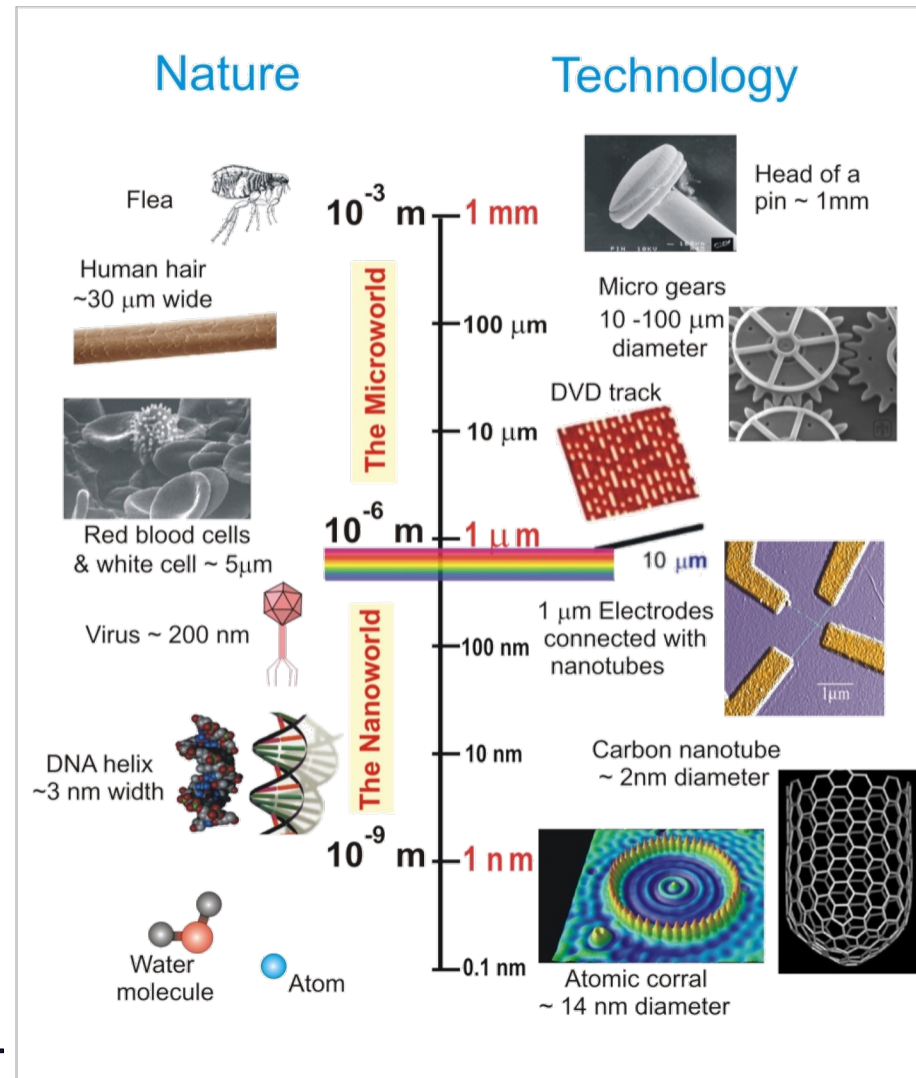
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# Ultrafast x-ray science

## Ultra-Small

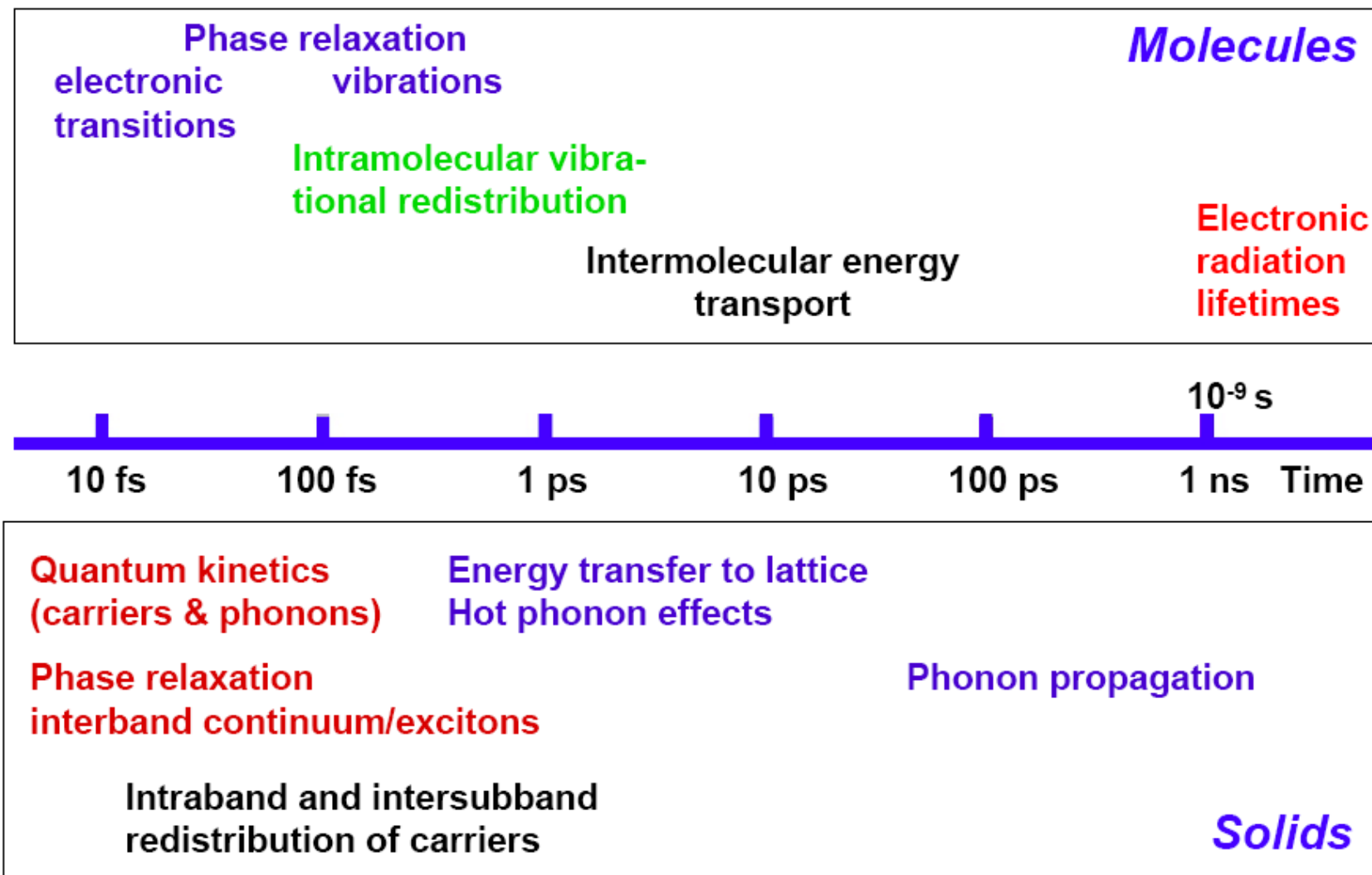
- Connection of length and time scales



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

Resolution elements

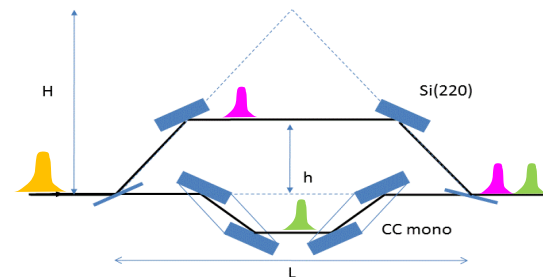
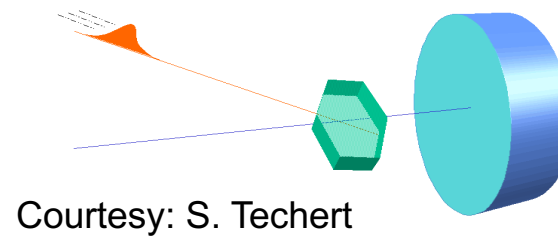
Exciting **pump** pulse: Optical laser, x-ray, others

**Probe** pulse: typ. x-ray

Precision of time-difference (jitter)

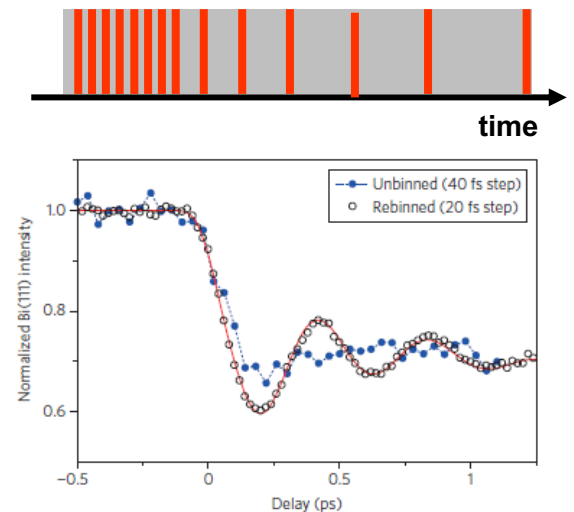
Synchronization of different light sources

Application range down to  $\sim 50$  fs  $\rightarrow 10$  fs  $\rightarrow 1$  fs

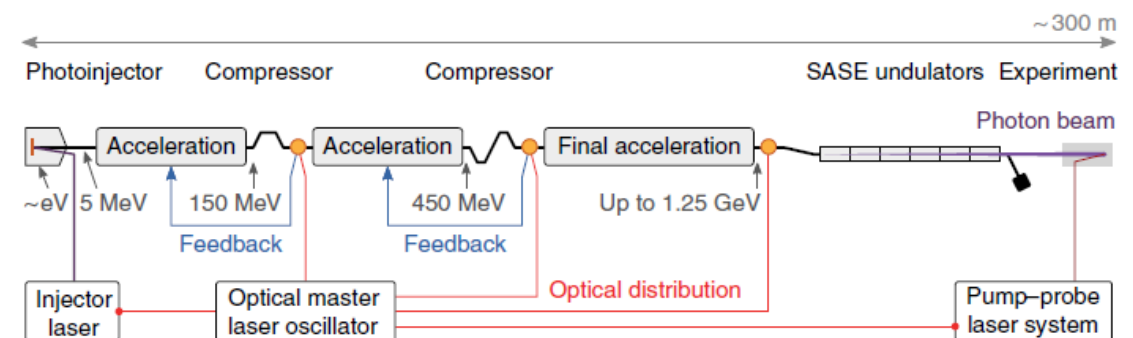


European XFEL pulse train

$\leftarrow 600 \mu\text{s}$  (1 bunch train)  $\rightarrow$

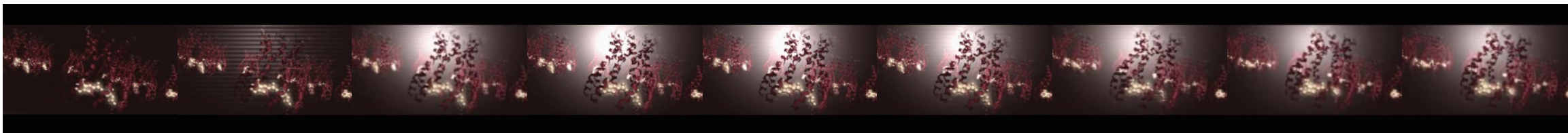


M. Harmand et al., Nature Phot. **7**, 215 (2013)

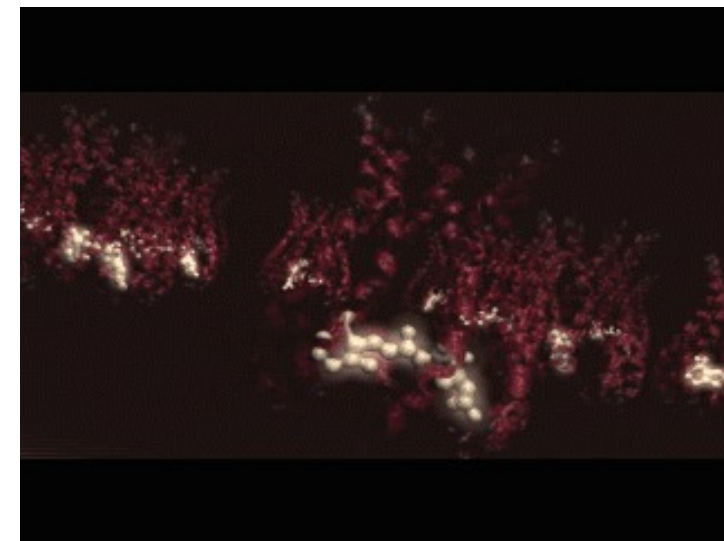
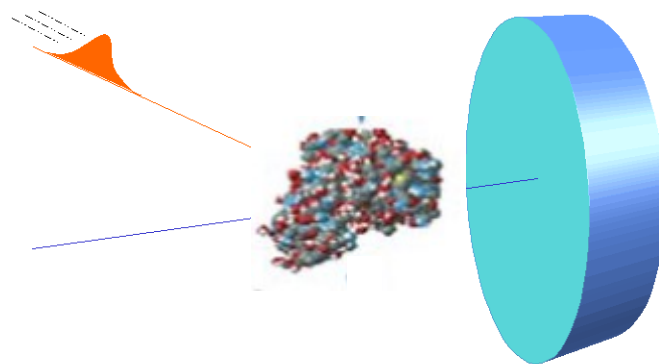


S. Schulz et al., Nat. Comm. **6**, 5938 (2015)

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

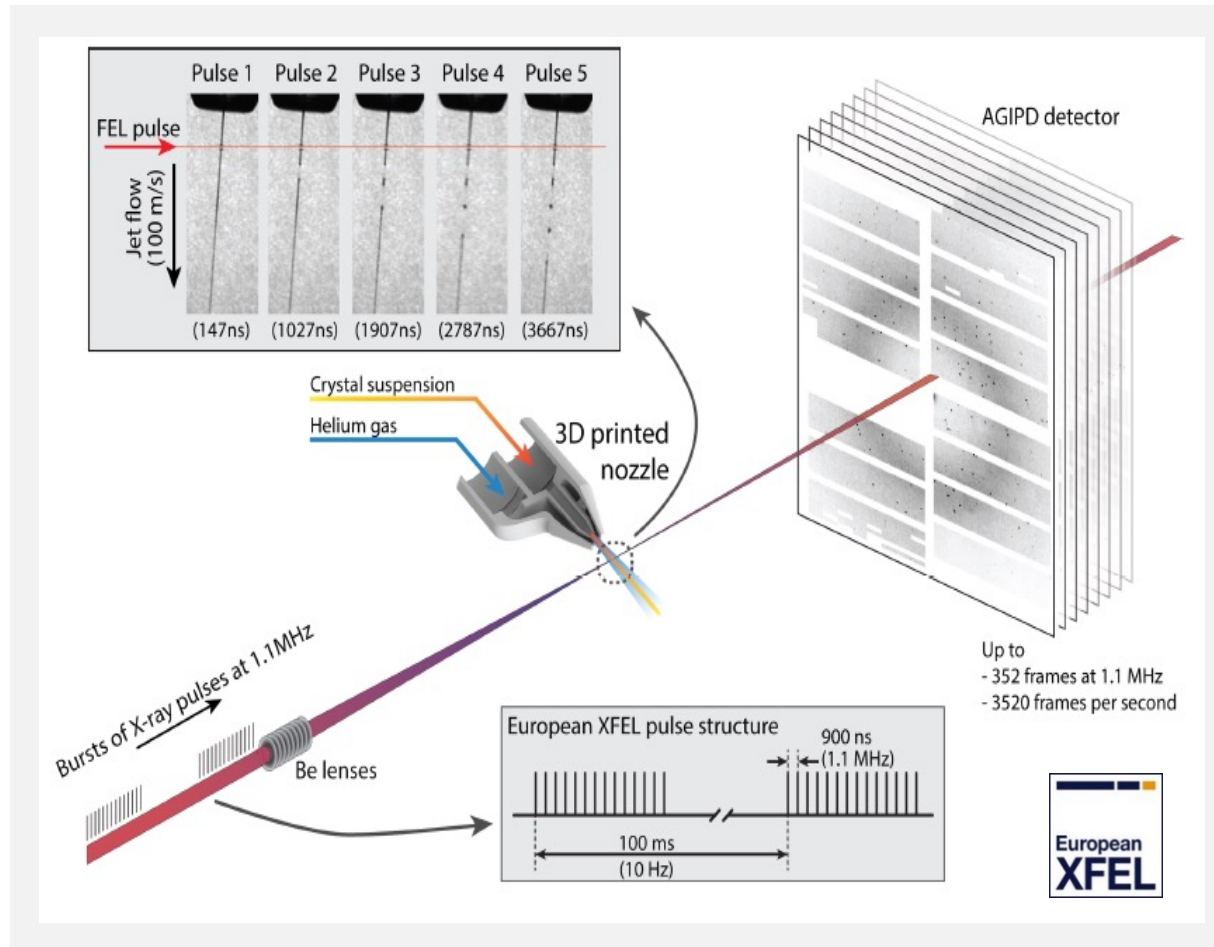


## ■ Making molecular movies

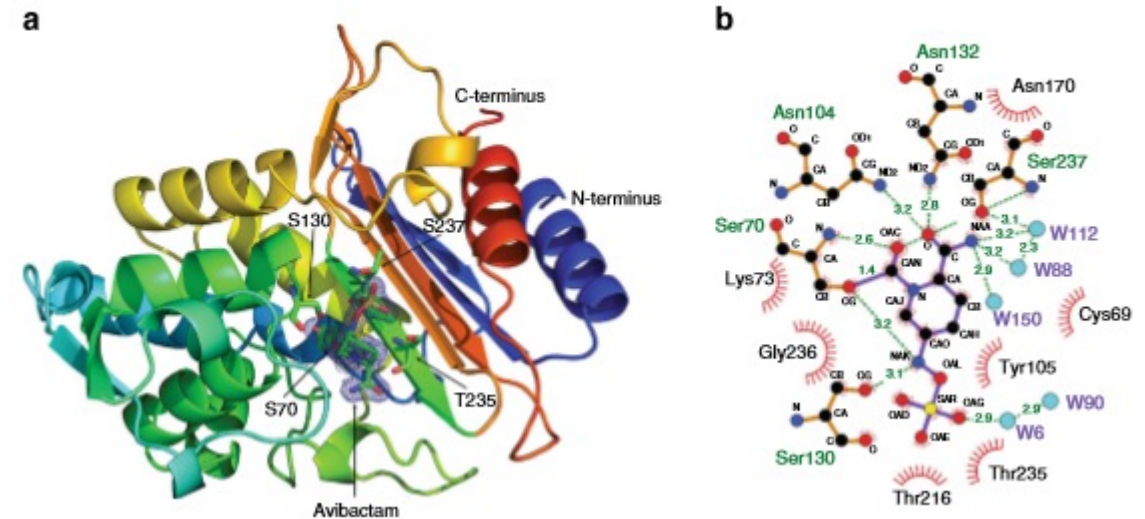


# Ad-hoc sample delivery methods have been developed

## ■ MHz Serial Femtosecond Crystallography (SFX)



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



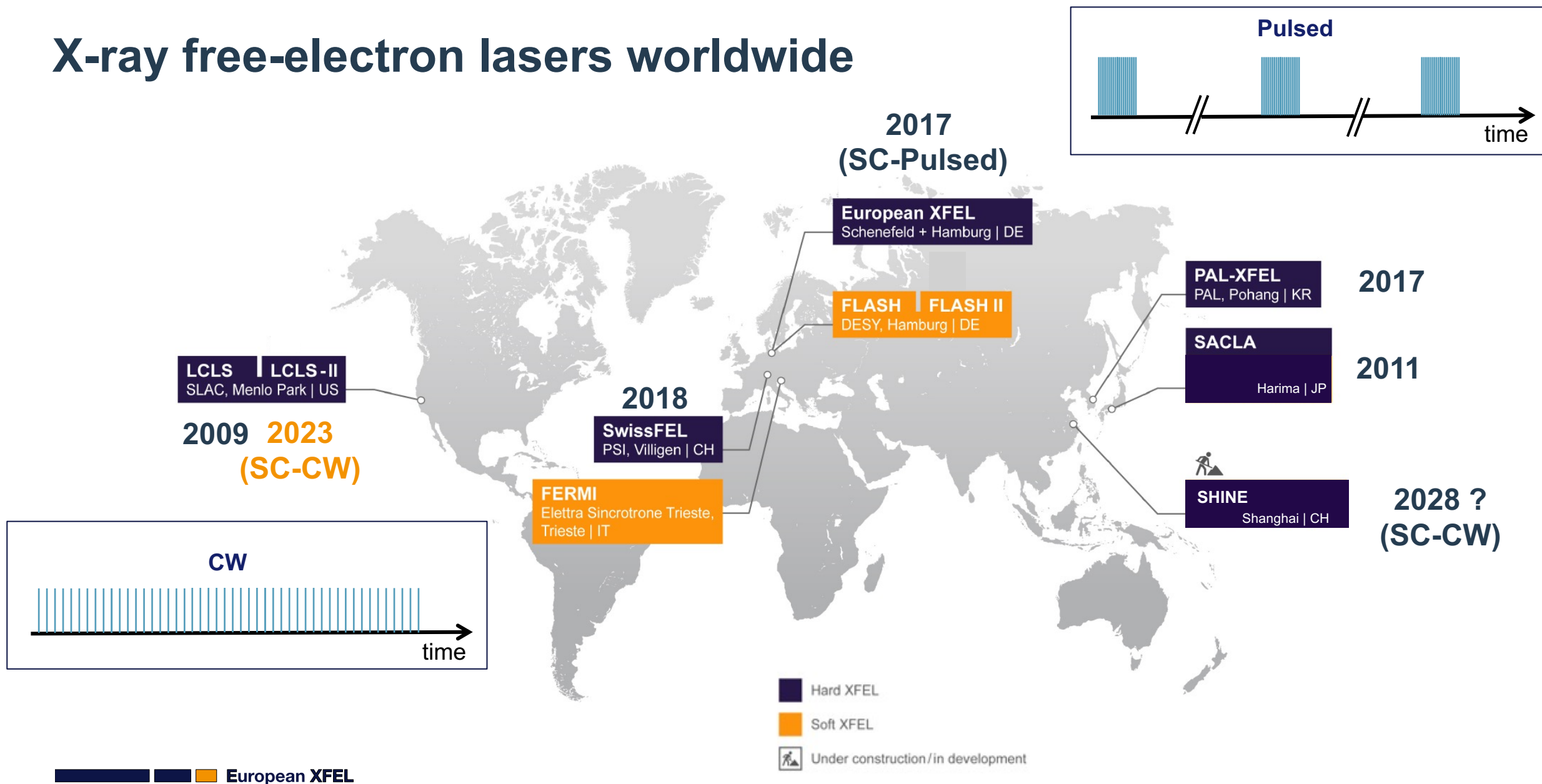
~  $10^5$  good patterns per structure

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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



# About the European XFEL

## International user facility for FEL research

- Providing soft & hard X-ray FEL radiation
- Photon energy 300 eV to  $\geq 20$  keV
- High pulse energies
- MHz repetition rates

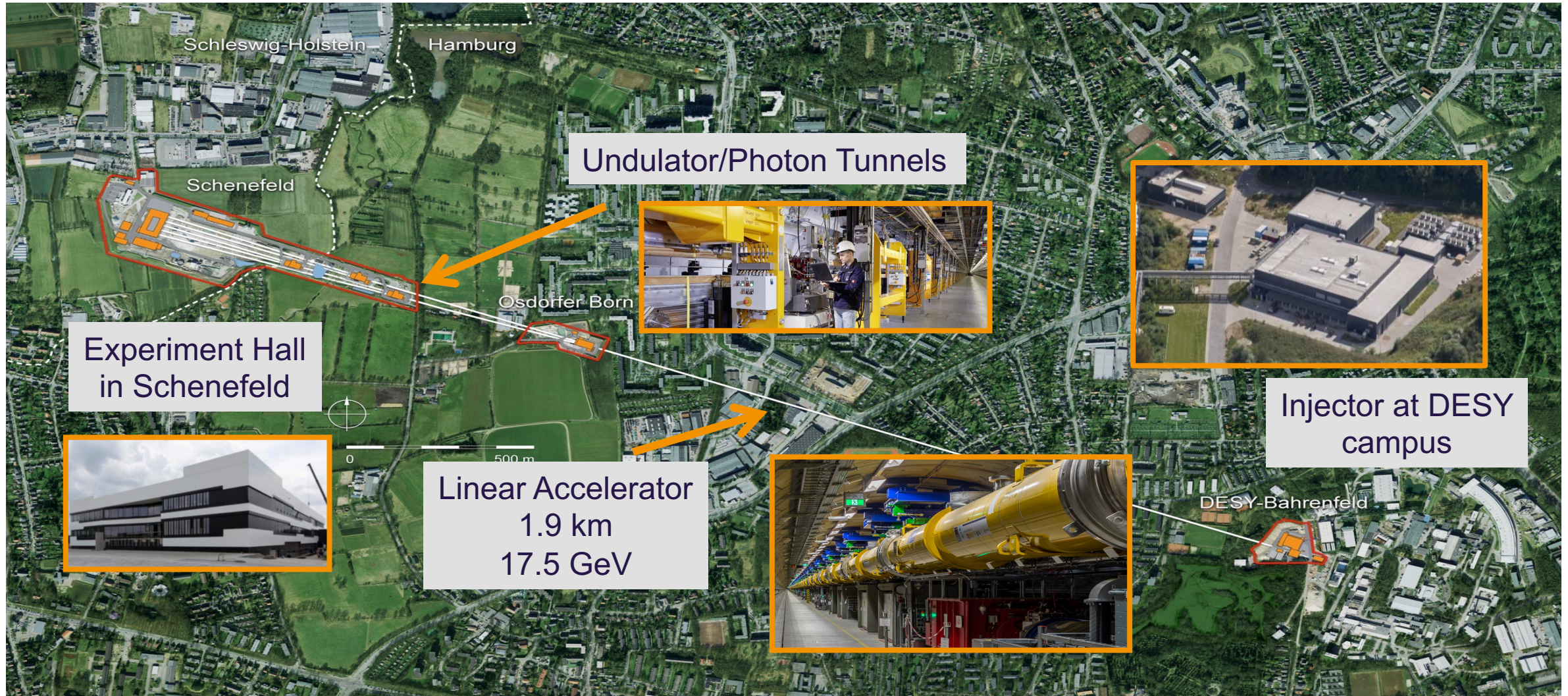
## Multi-disciplinary science community

- Physics
- Chemistry
- Biology
- Geosciences
- Materials

## Status

- Construction 2009-2017
- First experiments 2017
- 6 instruments operating since 2019 (+ 1 in 2023)

# Layout of the European XFEL





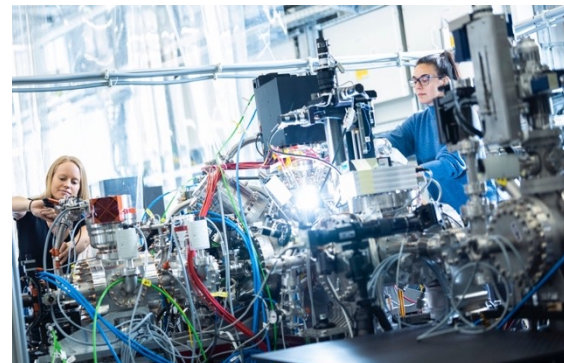
# Seven scientific instruments



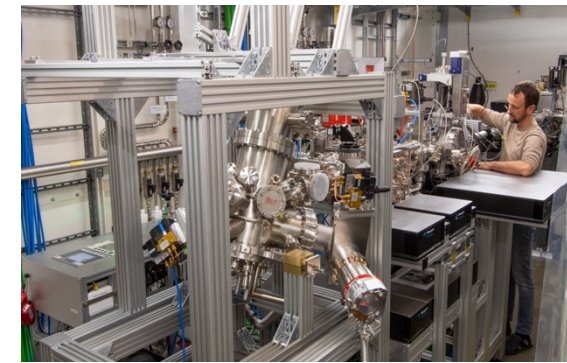
**SPB/SFX** (start Sep 2017)



**MID** (start Apr 2019)



**SQS** (start Nov 2018)



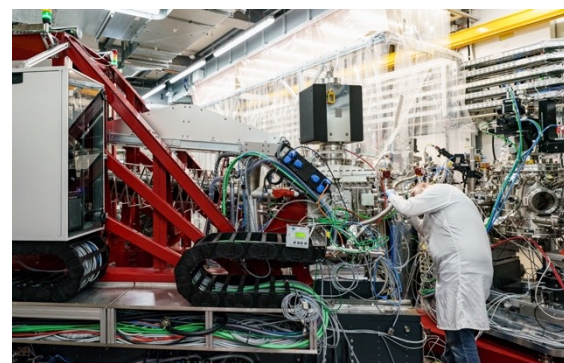
**SXP** (start summer 2023)



**FXE** (start Sep 2017)



**HED** (start May 2019)



**SCS** (start Nov 2018)

# Scientific instruments

SASE1

## FXE (Femtosecond X-ray Experiments)

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

SASE2

## MID (Materials Imaging & Dynamics)

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

SASE3

## SQS (Small Quantum Systems)

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

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

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

## HED (High Energy Density science)

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

## SCS (Spectroscopy & Coherent Scattering)

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

## SXP (Soft X-ray Port)

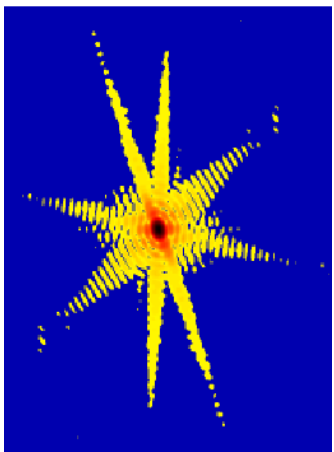
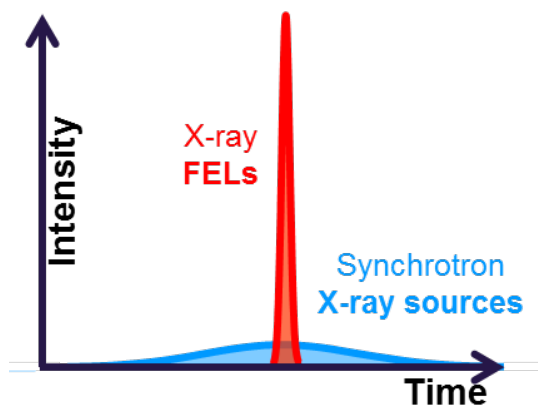
- Time-resolved X-ray photoelectron spectroscopy

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# New opportunities offered by X-ray FELs



## Ultrashort pulses

**1 – 100 fs**

## Coherence

**Almost full transverse**

**Partially temporal**

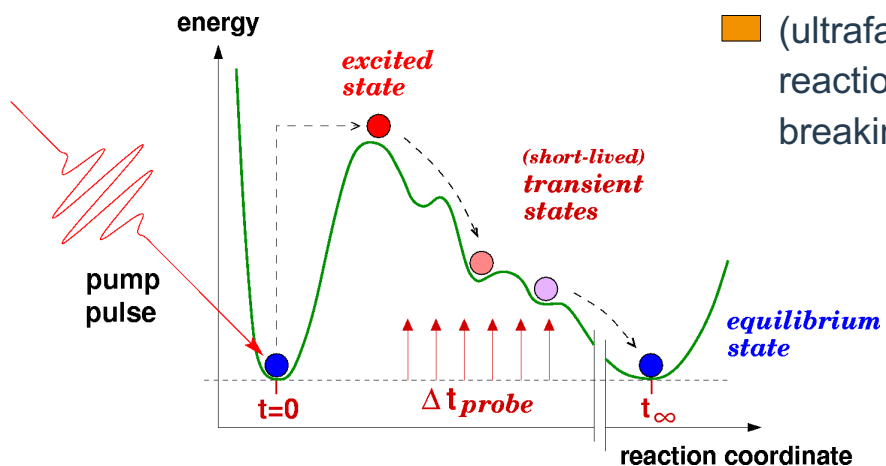
## Intensity/power

**up to few mJ**

**up to  $>10^{20}$  W/cm<sup>2</sup>**

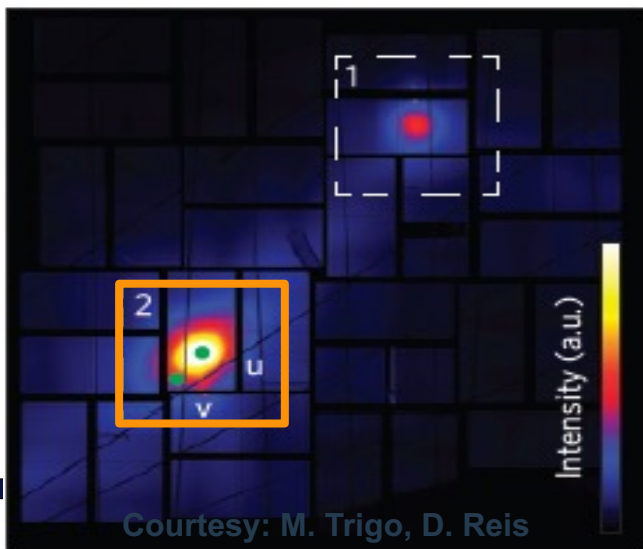
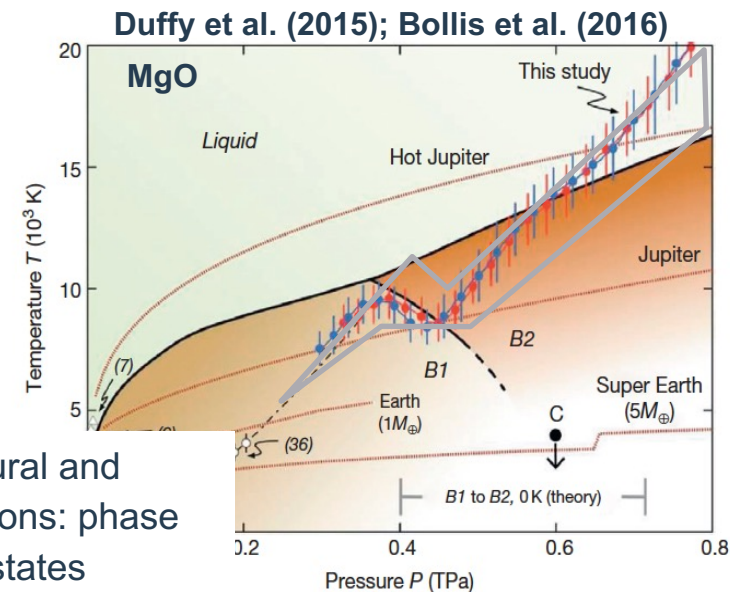
- Structural dynamics
  - Measurement of atomic and electron dynamics with high spatial [0.1 nm] and temporal [10 fs] res.
  - physics, materials sciences, chemistry, life science
- Imaging at the nanoscale
  - Imaging experiments on confined and extended objects with atomic to mesoscale resolution [0.1–1000 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



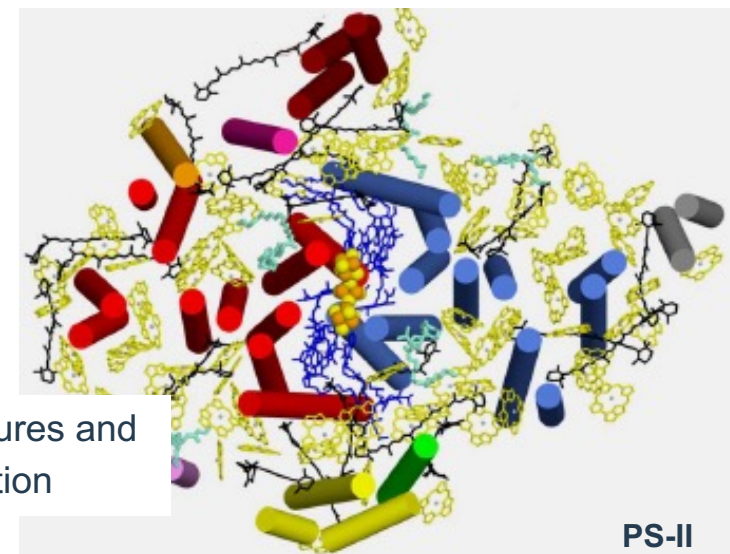
(ultrafast) (bio-)chemical processes:  
reactions, phase transitions, bond  
breaking & forming

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



Systems 'in function': excited states,  
non-reversible processes

complex (bio-)structures and  
their temporal evolution

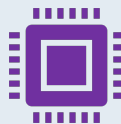


# Research at the European XFEL

## Digitalization

Structural dynamics of functional and quantum materials

- Magnetism at nm/fs scales
- Promotion of AI/data technologies



## Climate & Energy

Structural dynamics of new materials, processes, and molecular reactions

- Photocatalysis
- Microphysics of Fusion



## Environment & Sustainability

Properties of durable materials, sustainable agriculture, and planetary processes

- Structure of natural insecticides
- Hard materials under load



## Health

Structural dynamics and reaction pathways of biomolecules, viruses, and cells

- Structure & dynamics of biomolecules
- New therapies and diagnostics



# Research at the European XFEL

## Climate & Energy

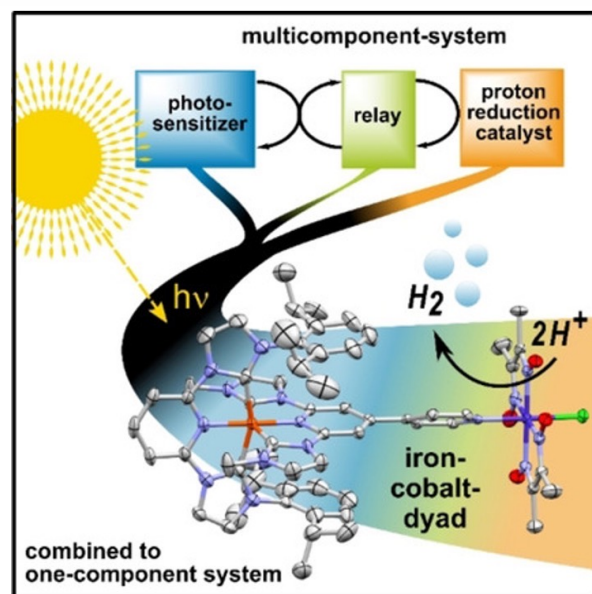
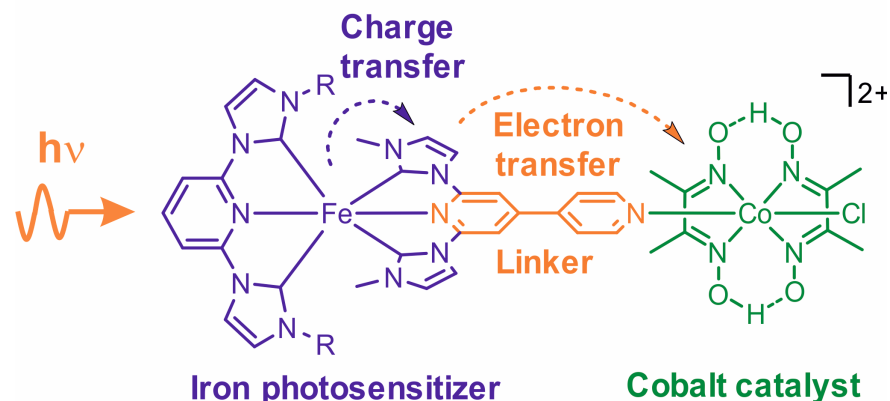
Structural dynamics of new materials,  
processes, and molecular reactions

- Photocatalysis
- Microphysics of Fusion

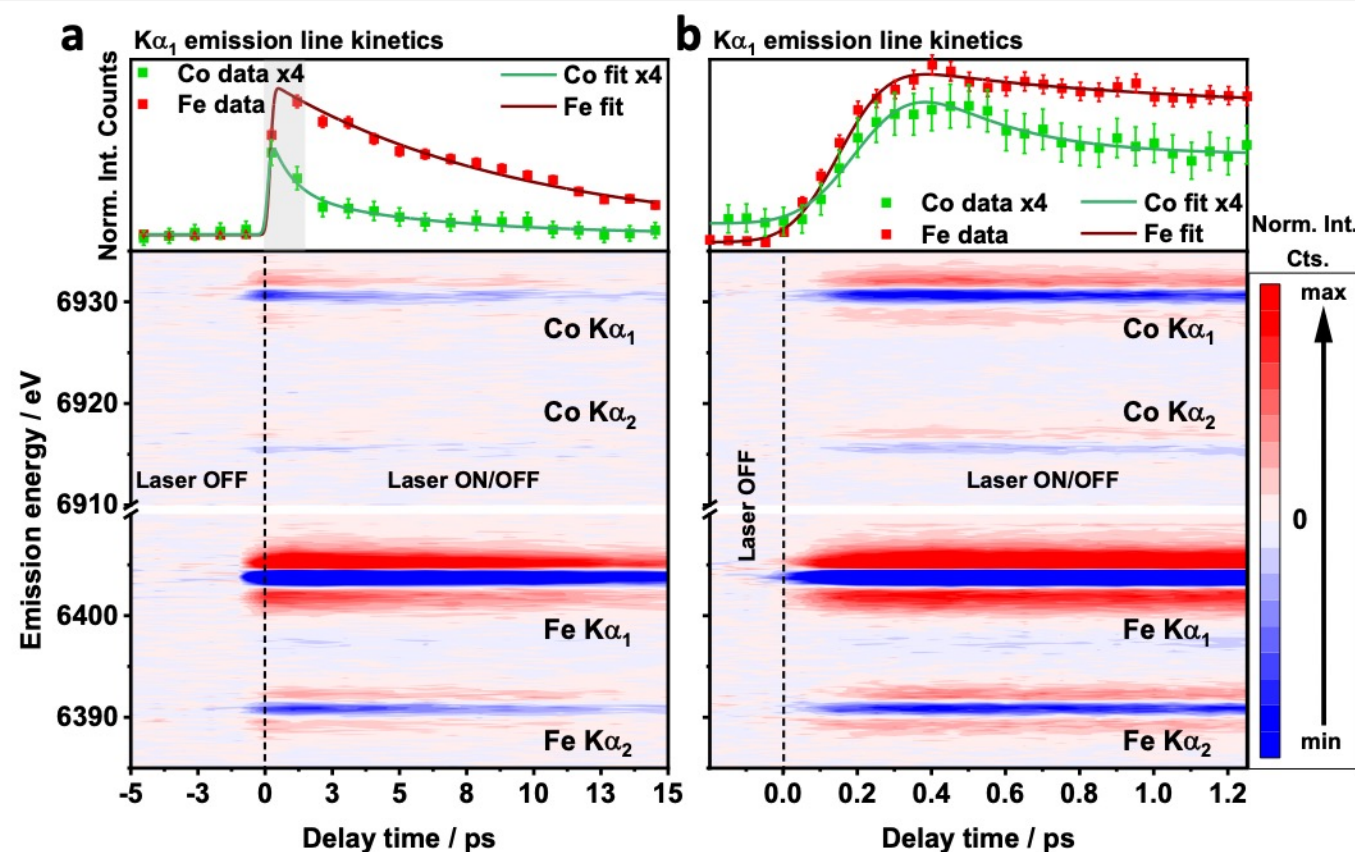


# Excited State Landscape in a Base Metal Dyad

Using light for a sustainable production of molecular hydrogen



## Ultrafast Two-Color X-Ray Emission Spectroscopy

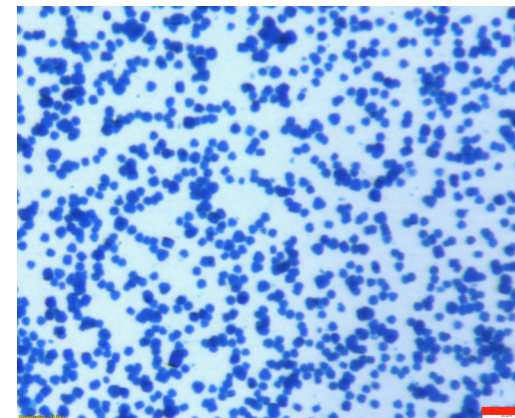
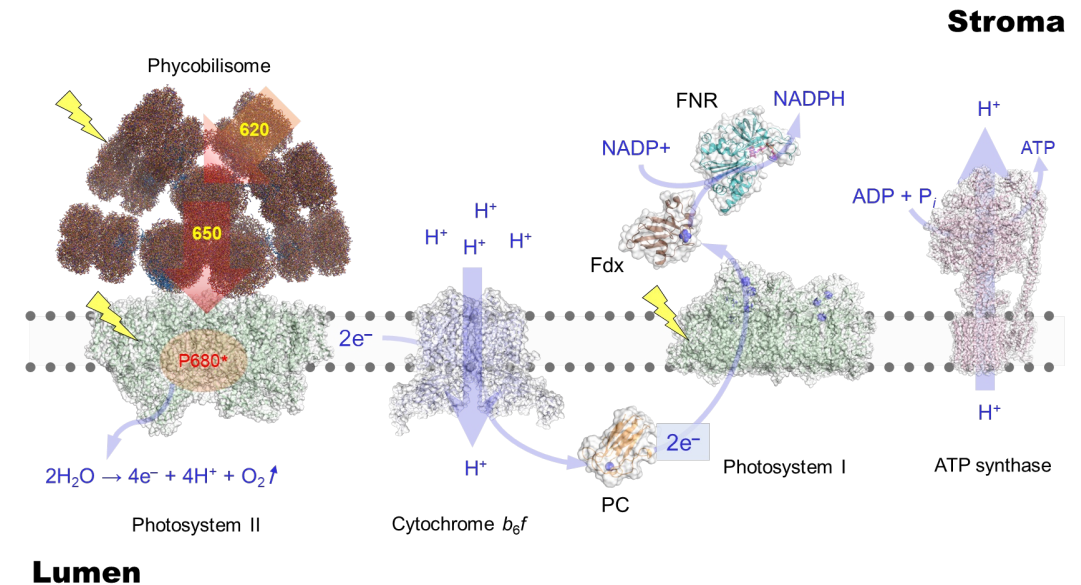


# Energy transfer mechanism in cyanobacteria

Towards more efficient photosynthetic light-harvesting

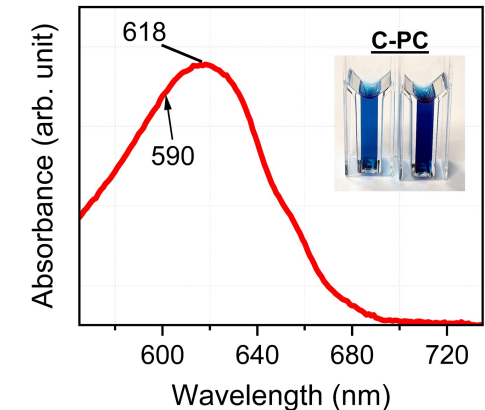
- Specialized bacteria which use photosynthetic energy generation
- Light absorbing-molecules (Phycocyanobilin "PCB") channel high energy photons and convert to lower energy photons suitable for photosystem II
- Funnels energy to photosynthetic reaction center (unidirectional down-hill energy transfer) through 3 chromophores
- Our study investigates the energy transfer mechanism via time resolved SFX of the Phycobilisome complex

SPB/SFX



Crystals for SFX

## Wavelength response

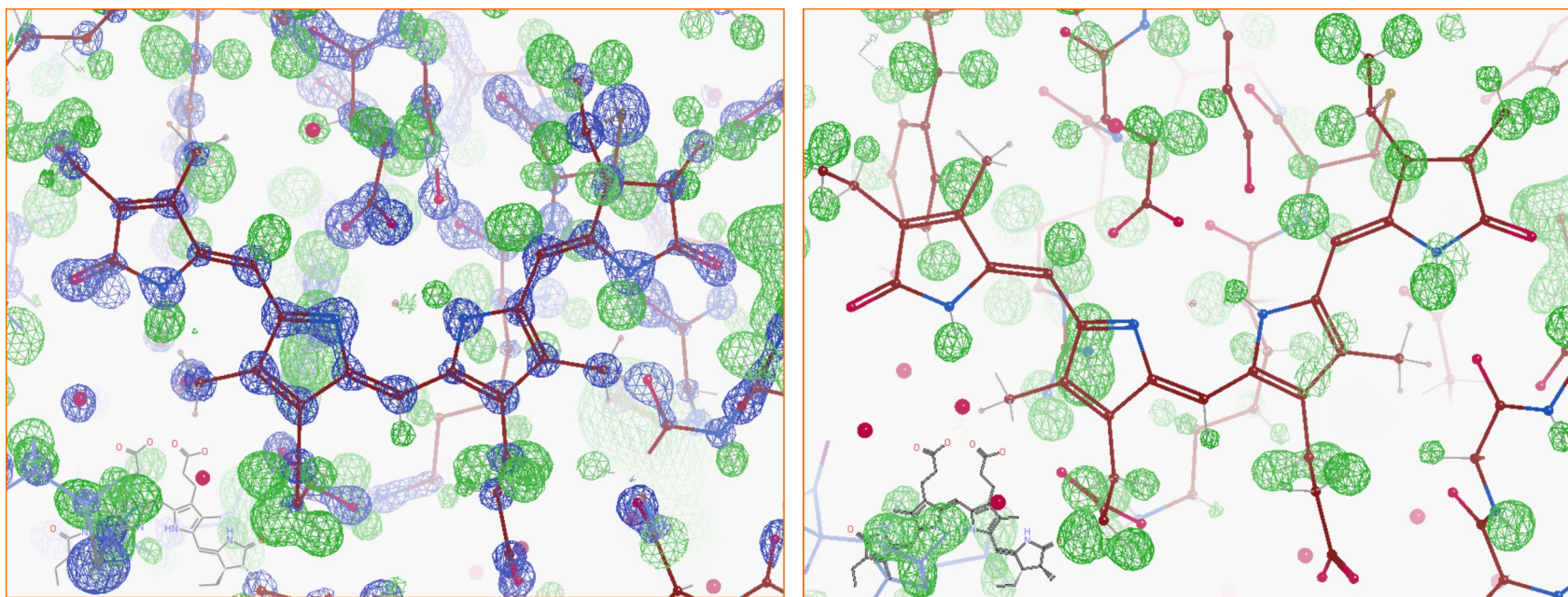


Faisal Koua et al., in preparation

# Energy transfer mechanism in cyanobacteria

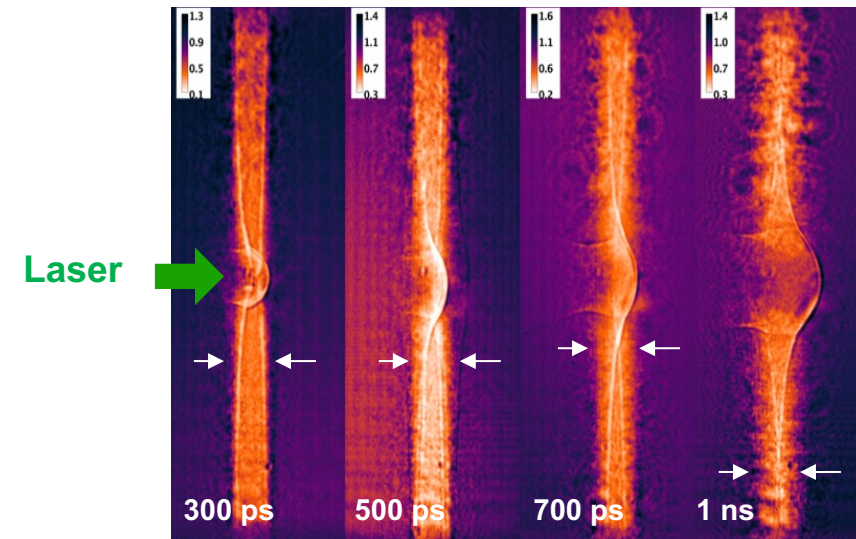
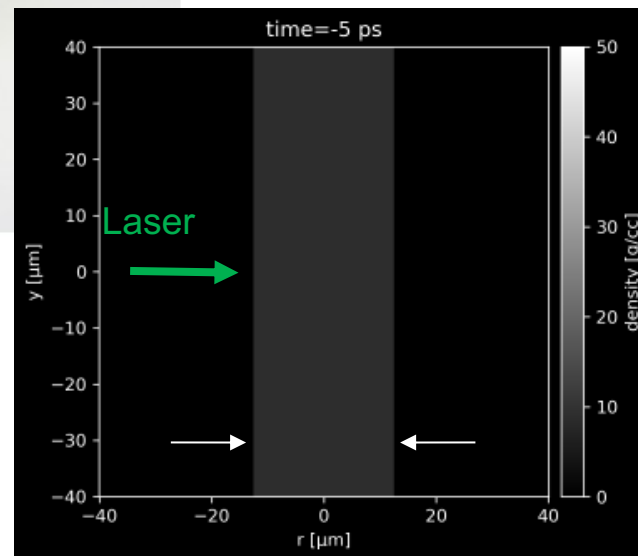
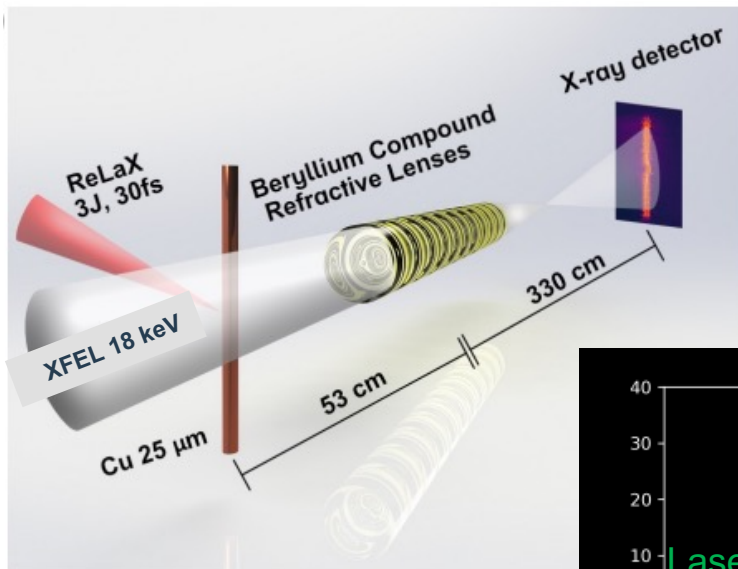
Towards more efficient photosynthetic light-harvesting

- High resolution structures allow accurate attribution of hydrogens (green mesh) in the structural model
- Observed change in chromophore electron densities (pentamer ring structures) and hydrogens over time sheds light on the energy transfer pathway through the harvesting molecule.



# Phase contrast imaging of cylindrical ablation-driven compression

Developing high resolution imaging of dense plasma



■ HED

- Relativistic laser drives 200 kA surface return current
- Surface heating to  $\sim 400$  eV  $\rightarrow$  ablative compression
- Cu compressed  $\sim 10 \times \rho_0 \sim 800$  Mbar
- New technique to reach stellar conditions with 1000-fold lower laser energy than with direct laser compression

# Research at the European XFEL

## Health

Structural dynamics and reaction pathways of biomolecules, viruses, and cells

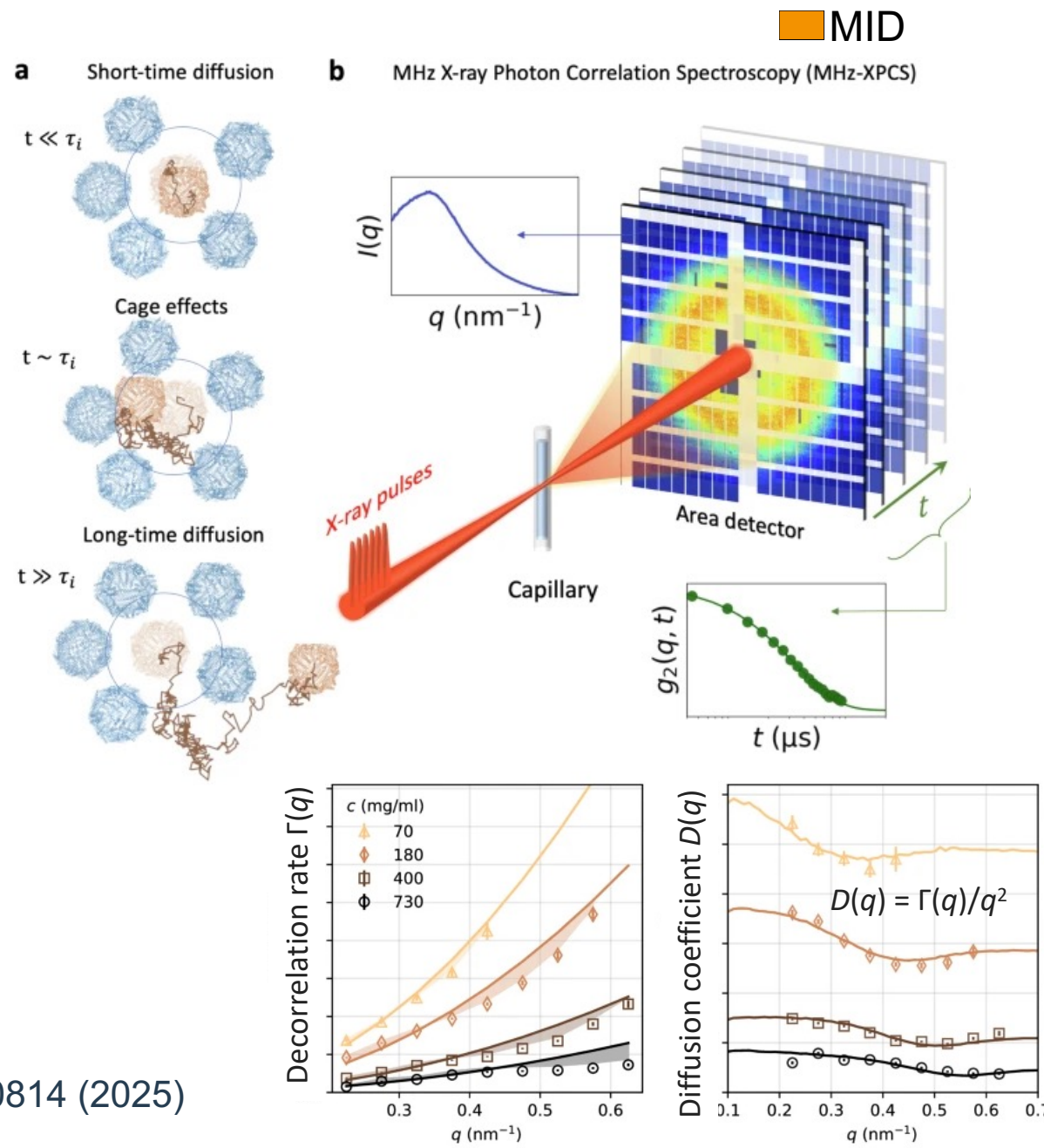
- Structure & dynamics of biomolecules
- New therapies and diagnostics



# Anomalous molecular diffusion in crowded protein solutions

Optimizing protein-based drug delivery

- Understanding protein motion within the cell is crucial for predicting reaction rates and macromolecular transport in the cytoplasm
- How do crowded environments, like inside the cell, affect protein dynamics ?
- The results reveal anomalous diffusion on microsecond time scales
- Behaviour consistent with the presence of cage-trapping in-between the short- and long-time protein diffusion regimes.



# Research at the European XFEL

## Environment & Sustainability

Properties of durable materials, sustainable agriculture, and planetary processes

- Structure of natural insecticides
- Hard materials under load

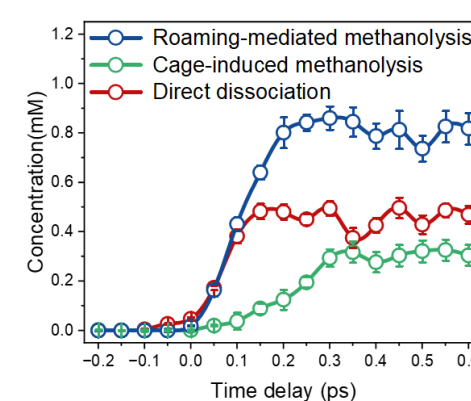
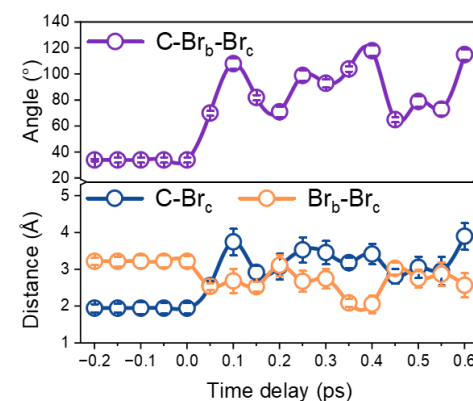
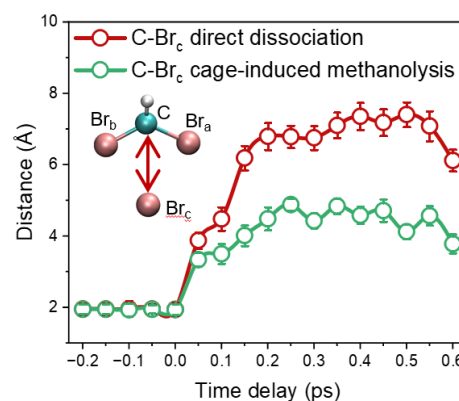
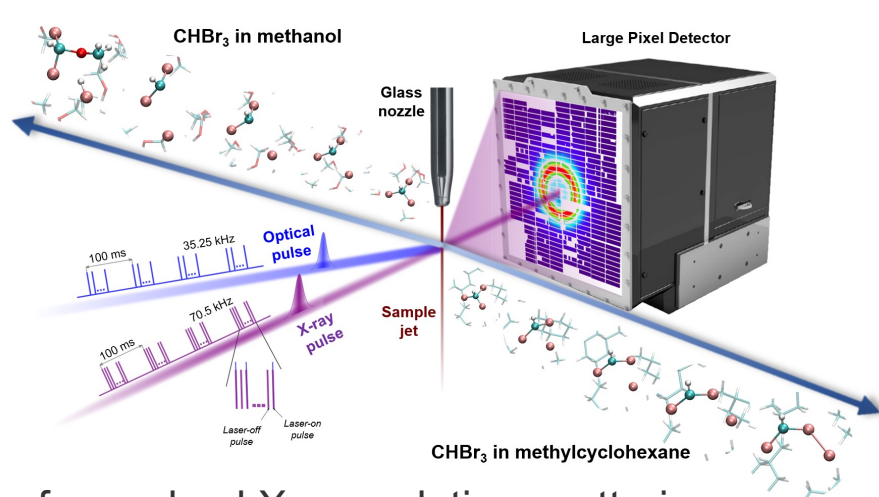
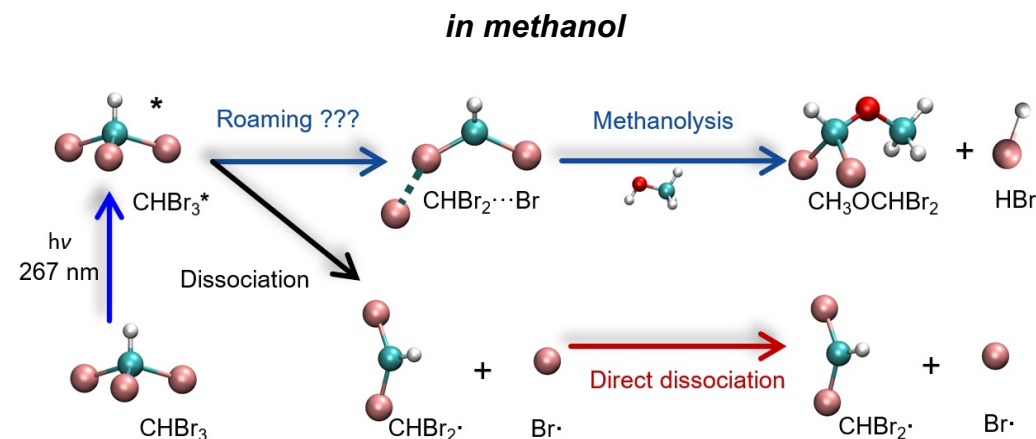


# Ultrafast solvent-modulated roaming mechanism in bromoform

How small molecules in the atmosphere dissociate under the effect of UV irradiation

FXE

- UV irradiation of poly-halomethanes releases reactive halogens, driving catalytic ozone depletion in the atmosphere
- Bromoform ( $\text{CHBr}_3$ ) is the most abundant natural source of bromine ( $\text{Br}_2$ ), >100 times more destructive to ozone than other halogens.
- Solvent-dependent reaction pathways of very short-lived roaming intermediates remain poorly understood, especially for environmentally relevant compounds involved in ozone depletion.



fs-resolved X-ray solution scattering

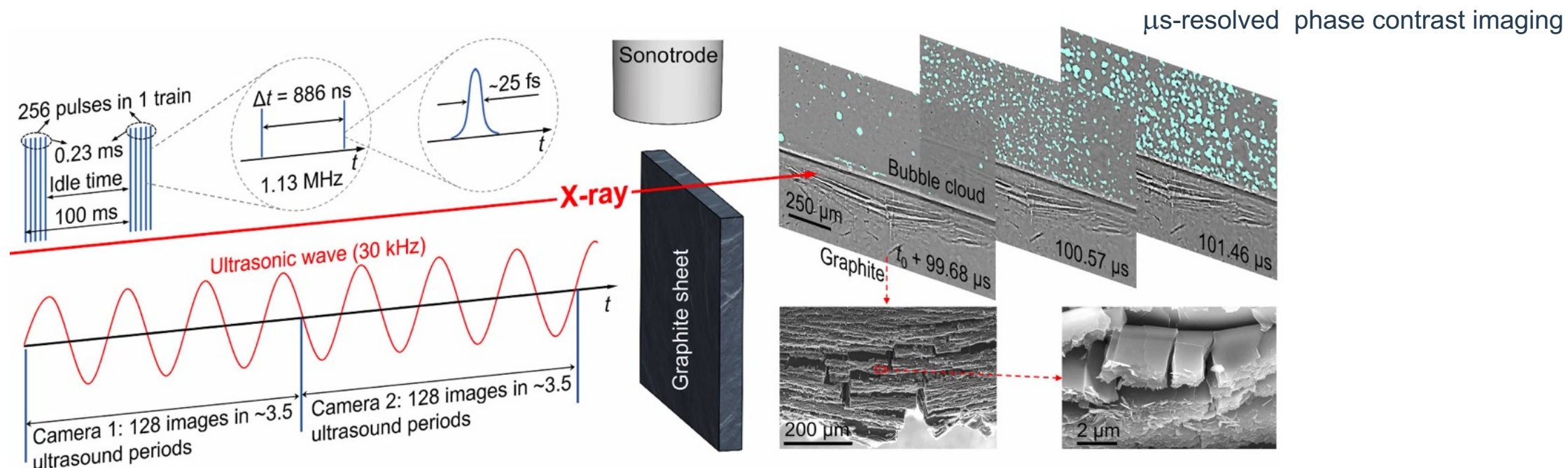
Peiyuan Su, ... & Qingyu Kong, Nature Comm 17, 2514 (2026)

# Ultrasonic exfoliation of graphene

SPB/SFX

Environmentally friendly processes to produce large surfaces of 2D functional materials

- Ultrasound Liquid Phase Exfoliation has been identified as a promising low cost and environmentally friendly technical route for producing large size multilayers from bulk materials.
- Investigation of the dynamics of cavitation bubbles and the subsequent exfoliation of graphene layers from graphite provides important insights for tuning the technique for commercial processes

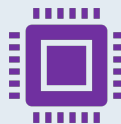


# Research at the European XFEL

## Digitalization

Structural dynamics of functional and quantum materials

- Magnetism at nm/fs scales
- Promotion of AI/data technologies

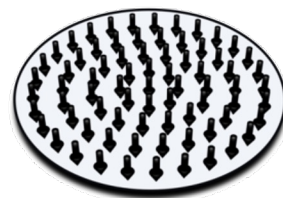


# Picosecond localization dynamics following ultrafast nanoscale topological phase transition

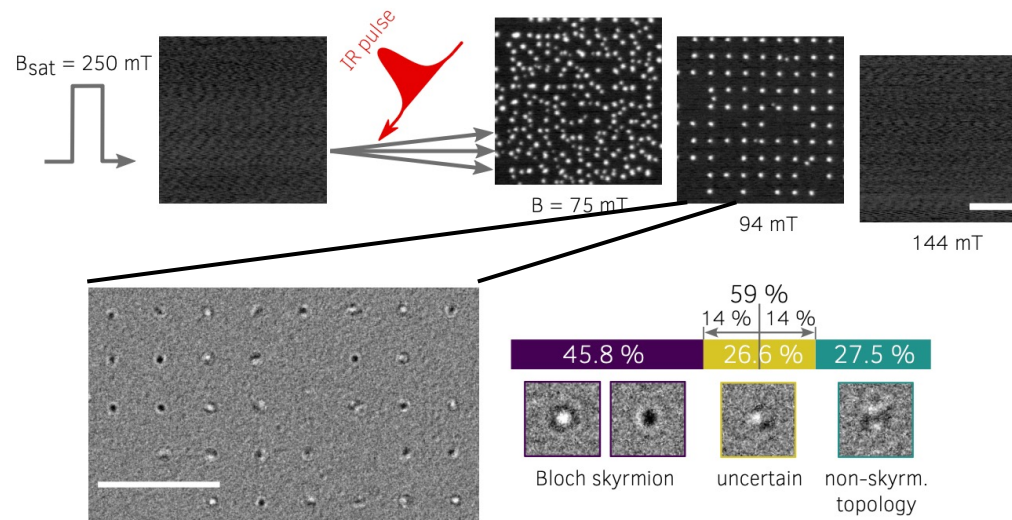
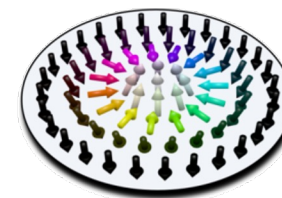
Advancing ultrafast control in nanoscale technologies

- Track, in real time, the laser-driven nucleation and localization of nm-scale spin textures
- Deterministic spatial control of skyrmion formation after laser excitation through ion-beam patterning of the underlying magnetic anisotropy
- Experiment reveals that nucleation and localization are two distinct processes, occur on different timescales
- Ultrafast phase-transition dynamics fundamentally differ from conventional transitions

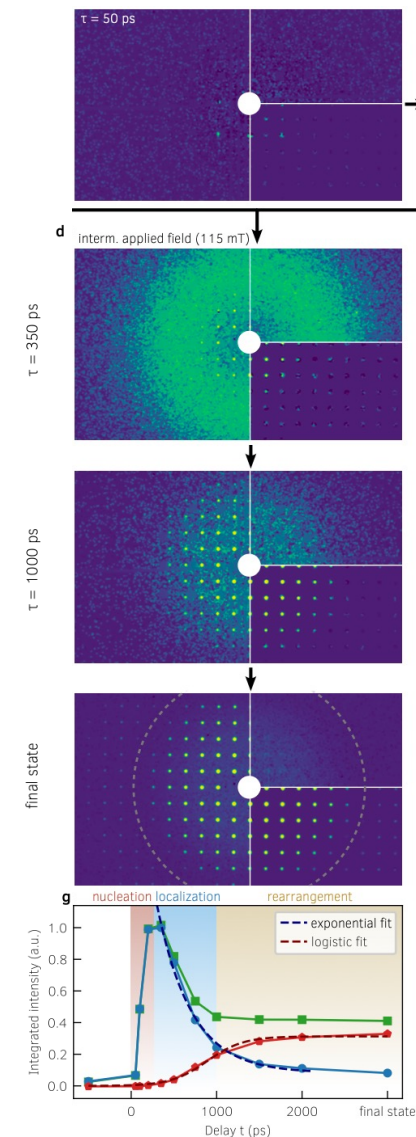
Uniform state



Magnetic Skyrmion



Shot-resolved resonant X-ray scattering



# Outline

**Break @ 17h10-17h20**

- What is an X-ray Free Electron Laser and how does it work ?
- Ultrafast science and ultrafast experiments
- X-ray Free Electron Lasers around the world
- What are the new science opportunities ? Examples of recent results
- Ongoing developments: Where are we heading to ? What new science ?
- Outlook

# Infrastructure: developments, challenges and strategy

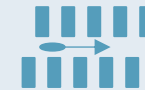
## Accelerator

- Operation and development of superconducting accelerator
- Enabling technologies (e.g. RF)
- Superconducting gun



## X-ray Sources

- X-ray cavity
- Superconducting undulators
- Polarization and angular momentum control



## Advanced Operation Modes

- Hard X-ray Self Seeding (HXRSS)
- Attosecond pulses
- High photon energies



## Detectors and Data Science

- Detectors: CW & hard X-ray compatible
- Data reduction, processing, analysis
- Machine learning, artificial intelligence
- High-performance computing



# Infrastructure: developments, challenges and strategy

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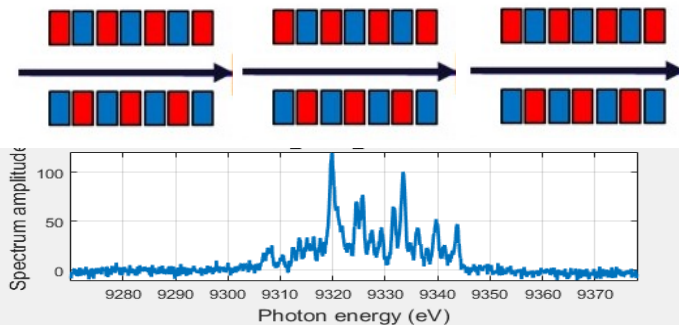
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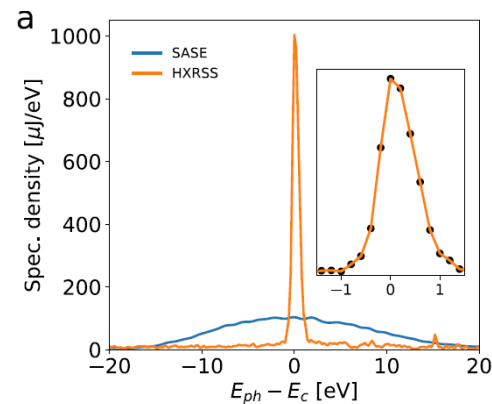
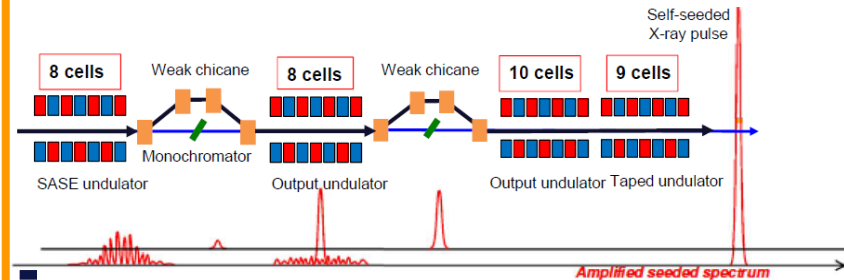
# Increasing spectral brilliance, reducing shot-shot instability

## SELF AMPLIFIED SPONTANEOUS EMISSION (SASE)



Photon Energy 9 keV  
 Pulse energy 2.2 mJ/pulse  
 BW (FWHM) 20 eV  
 Spectral density 100  $\mu\text{J}/\text{eV}$

## HARD X-RAY SELF SEEDING (HXRSS)



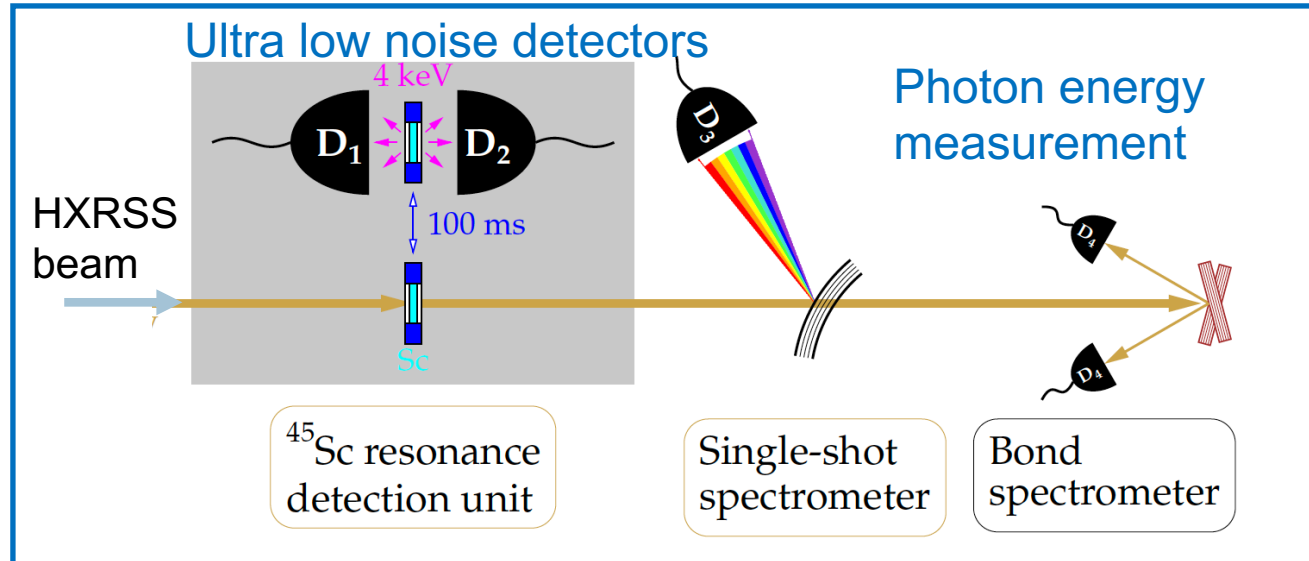
Photon Energy 9 keV  
 Pulse energy 1.2 mJ (bkg 0.4 mJ)  
 BW (FWHM) 0.8 eV  
 Spectral density 1000  $\mu\text{J}/\text{eV}$

Liu et al., Nature Photonics 17, 984 (2023)

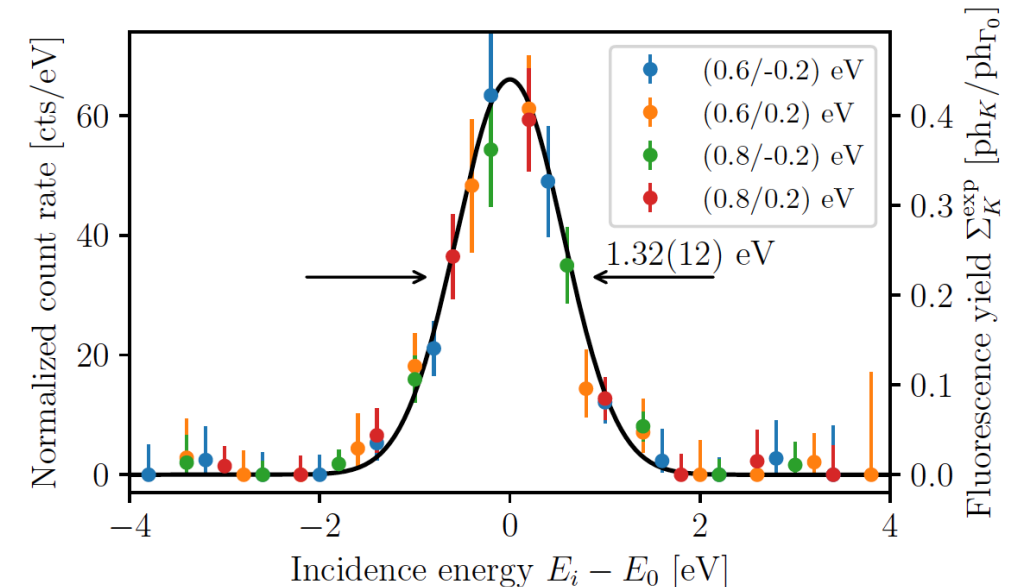
MID

# Resonant excitation of the nuclear clock isomer $^{45}\text{Sc}$

- Resonant oscillators with stable frequencies and large quality factors determine our ability to keep track of time with high precision
- Nuclear oscillators surpass their atomic counterparts by their naturally higher quality factors and higher resilience against external perturbations
- The 12.4 keV nuclear transition in  $^{45}\text{Sc}$ 
  - Ultranarrow transition linewidth: 1.4 femto-eV
  - Extreme metrology and ultraprecise frequency standards



$$E_0 = 12,389.59^{+0.15(\text{stat})}_{+0.12(\text{syst})} \text{ eV}$$



# Infrastructure: developments, challenges and strategy

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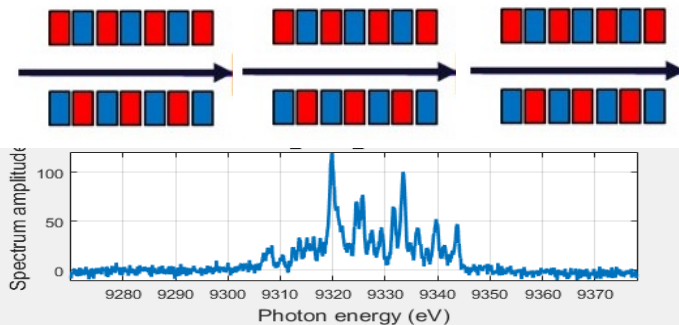
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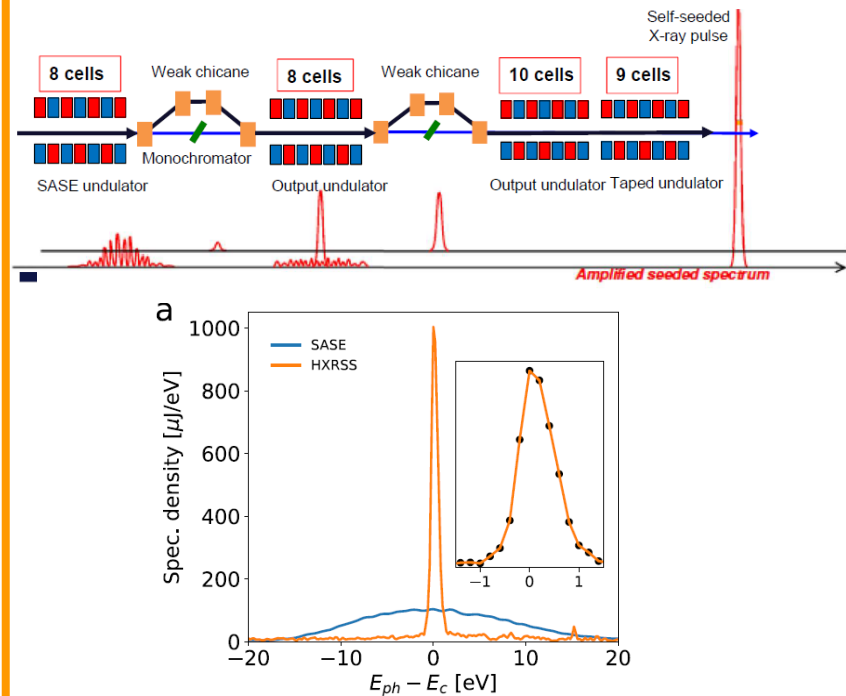
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 BW (FWHM) 20 eV  
 Spectral density 100 uJ/eV

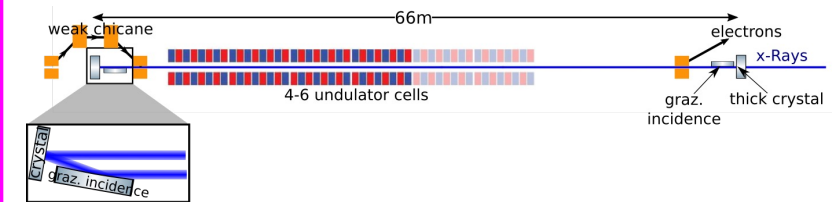
## HARD X-RAY SELF SEEDING (HXRSS)



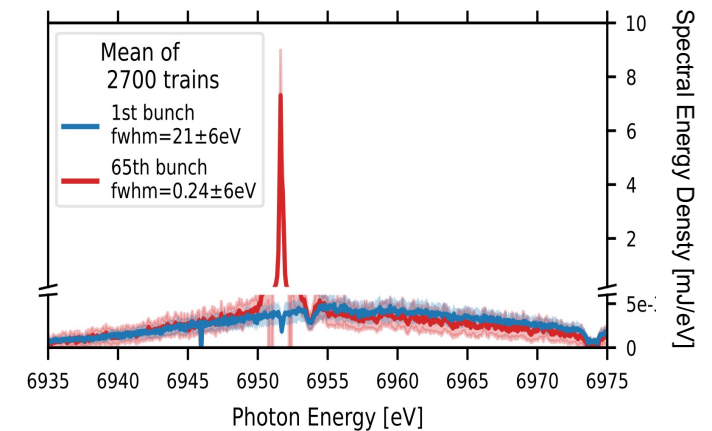
Photon Energy 9 keV  
 Pulse energy 1.2 mJ (bkg 0.4 mJ)  
 BW (FWHM) 0.8 eV  
 Spectral density 1000 uJ/eV

Liu et al., Nature Photonics 17, 984 (2023)

## CAVITY BASED X-RAY SOURCE



Sunday 18 May 2025



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

Rauer et al., Nature 650, 93 (2026)

# Infrastructure: developments, challenges and strategy

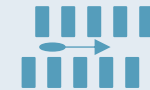
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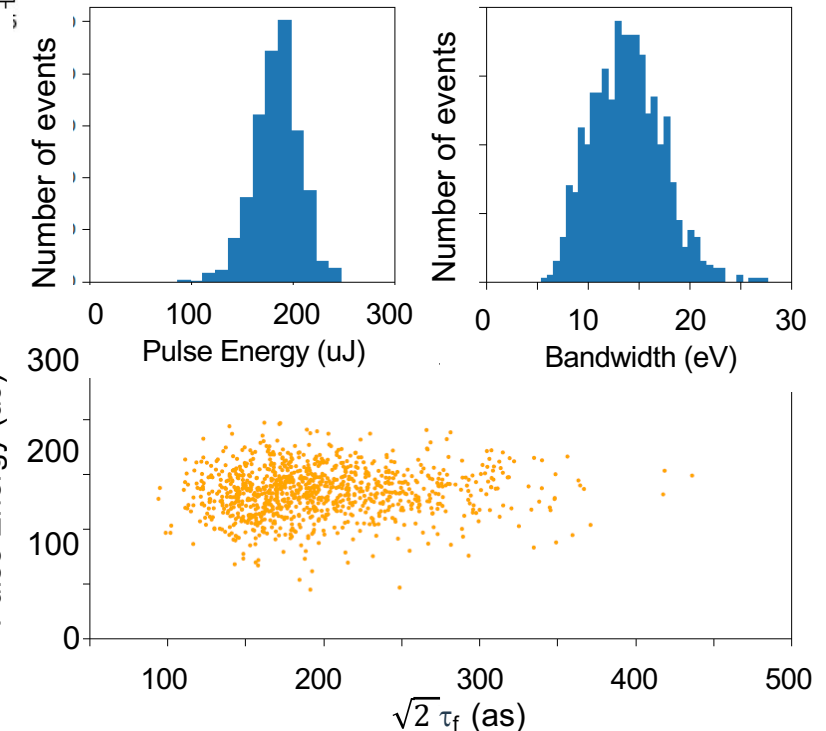
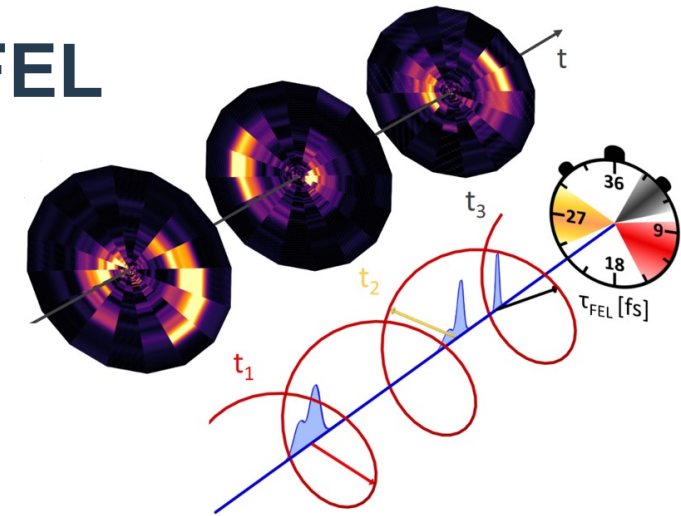
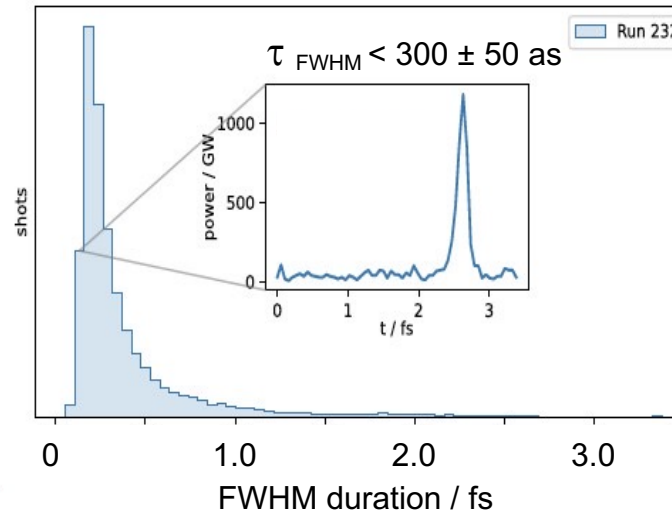
# Current generation capabilities at the European XFEL

## Soft X-rays

- up to mJ level
- ~ keV photon energy
- few 100 as
- 2-color pulses up to few 100 uJ each

## Hard X-rays

- up to few 100 uJ
- ~ 9keV photon energy
- few 100 as
- 2-color pulses - preliminary demonstration



## Terawatt-attosecond hard X-ray free-electron laser at high repetition rate

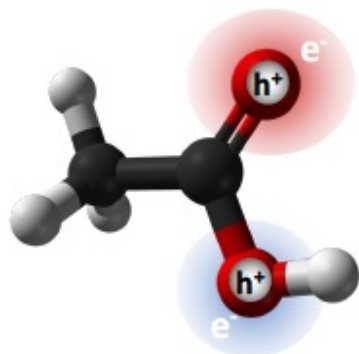
Jiawei Yan , Weilun Qin, Ye Chen, Winfried Decking, Philipp Dijkstal, Marc Guetg, Ichiro Inoue, Naresh Kujala, Shan Liu, Tianyun Long, Najmeh Mirian & Gianluca Geloni

*Nature Photonics* **18**, 1293–1298 (2024) | [Cite this article](#)

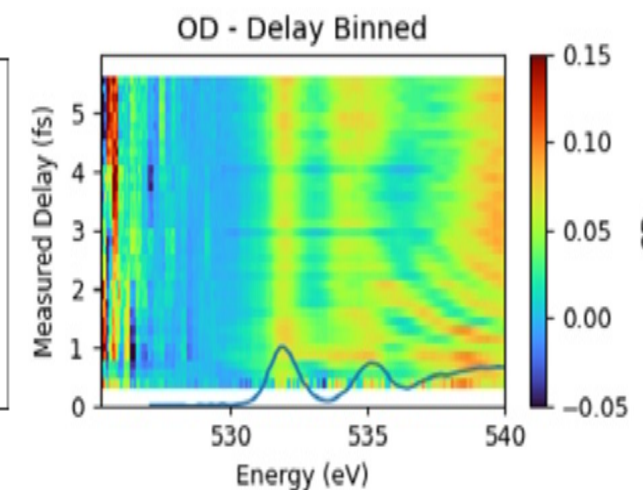
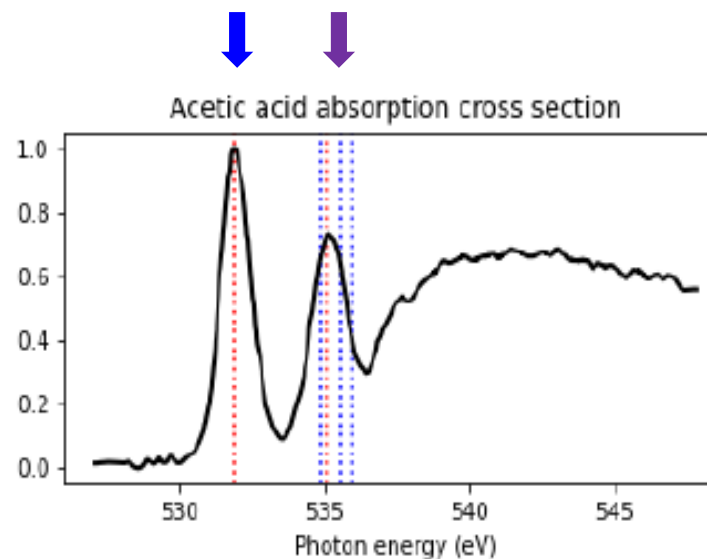
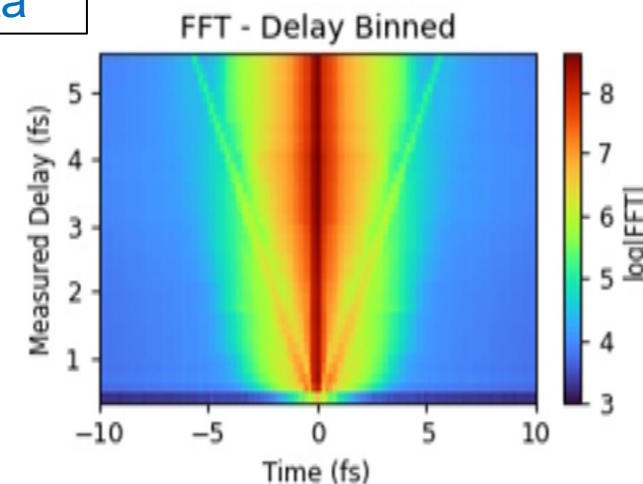
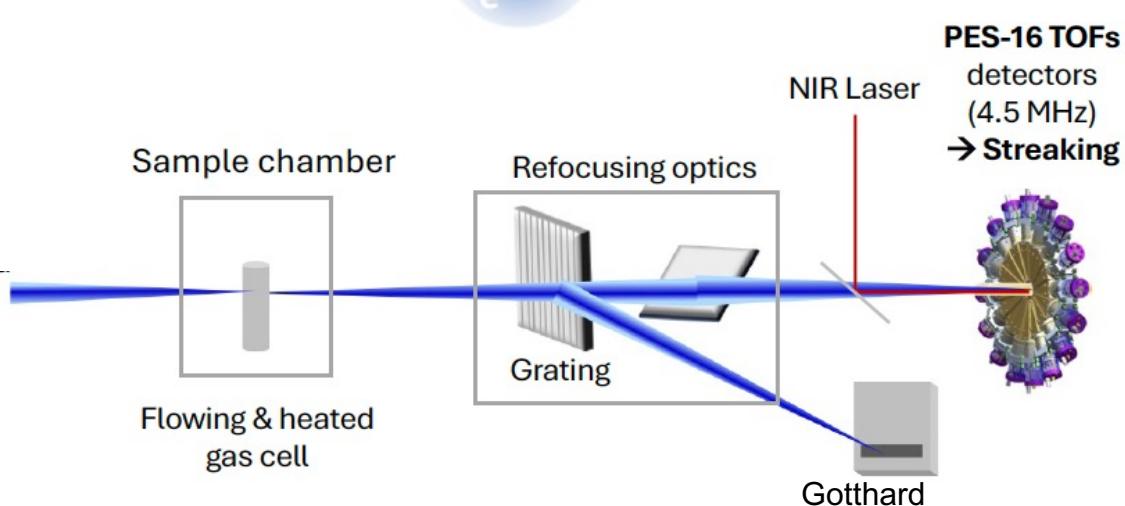
# Electron Dynamics in Molecules

## Transient Absorption on Acetic Acid

Preliminary data



Site-resolved electron dynamics  
on two chemically distinct  
oxygen atoms

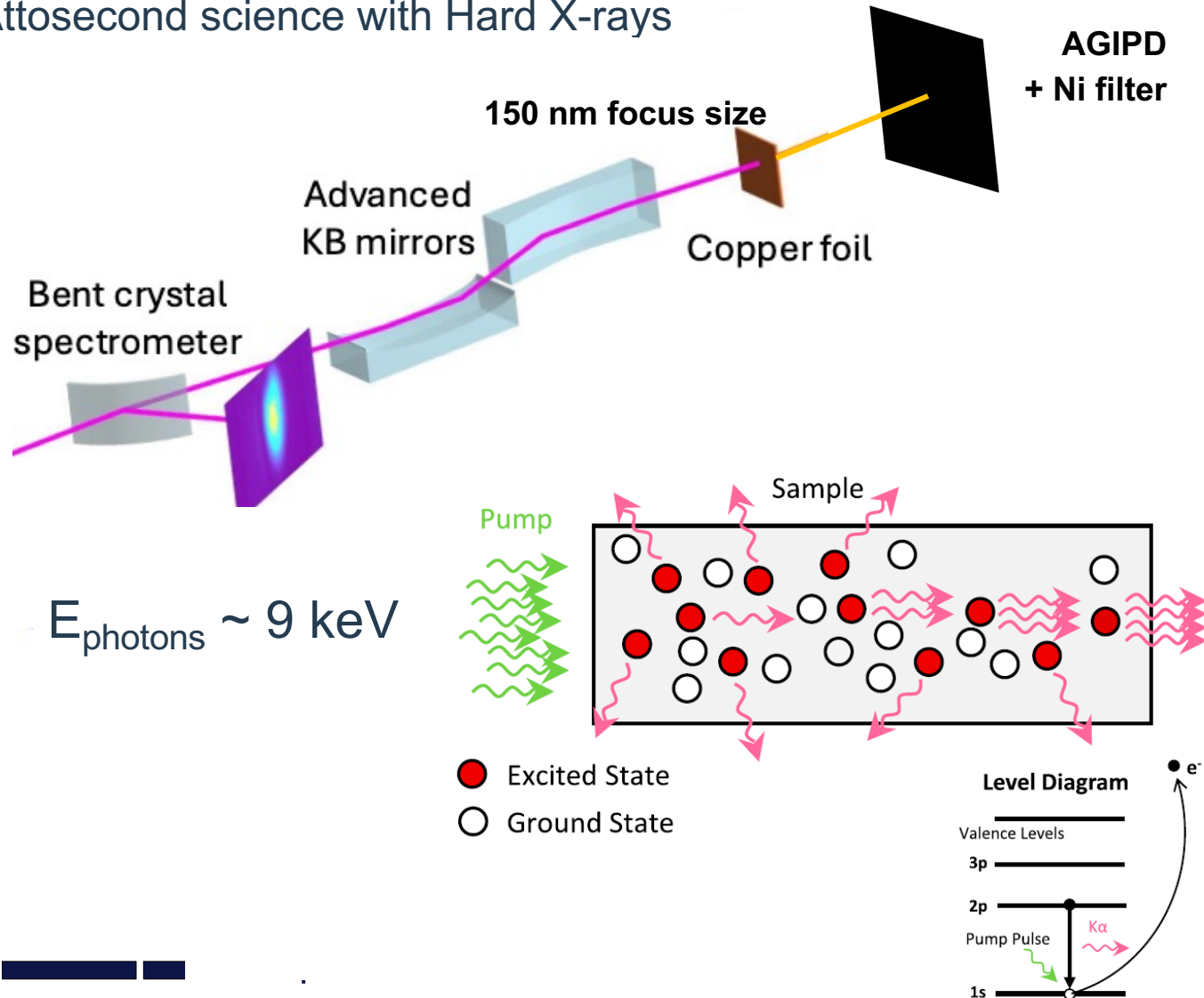


## Two-Color (X-ray / X-ray) Spectroscopy at SQS

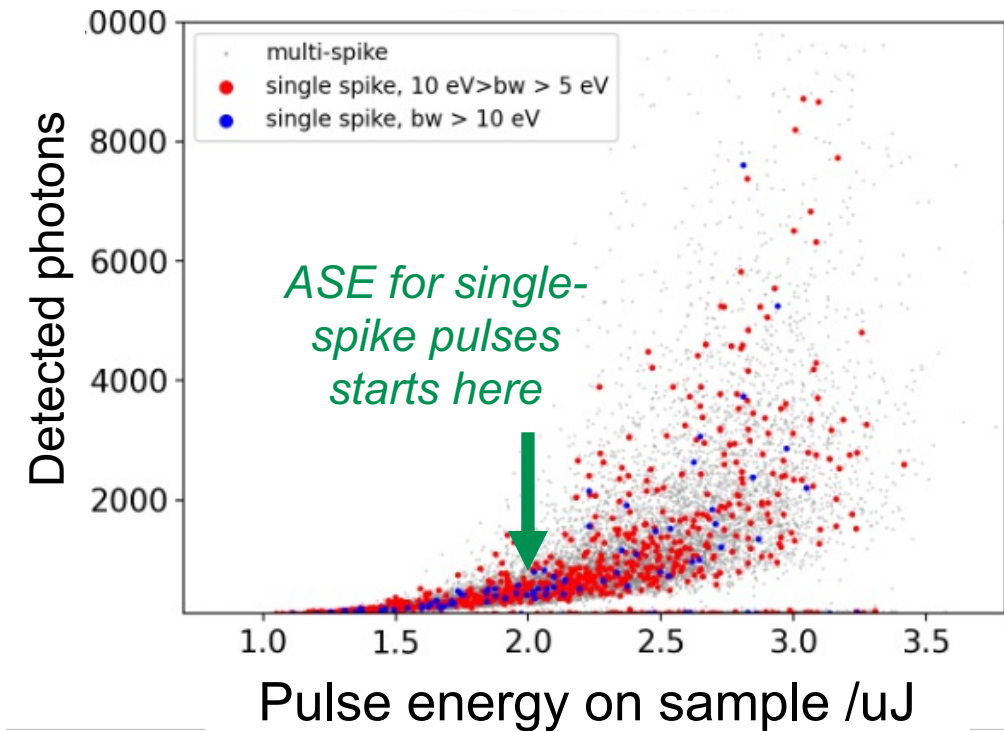
# Amplified Spontaneous Emission in Cu

 MID

Attosecond science with Hard X-rays



ASE: Amplified Spontaneous Emission  
(onset of non-linear behaviour at  $\sim 2 \times 10^{19} \text{ W/cm}^2$ )



Ichiro Inoue *et al.* (in preparation)

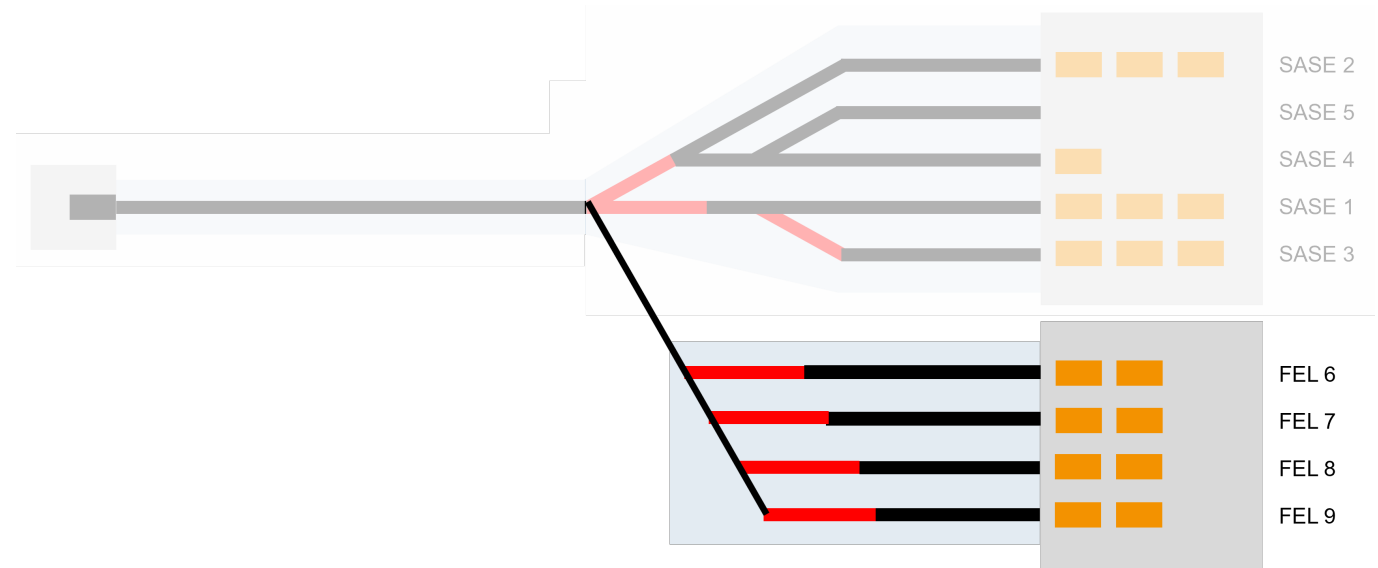
# Outline

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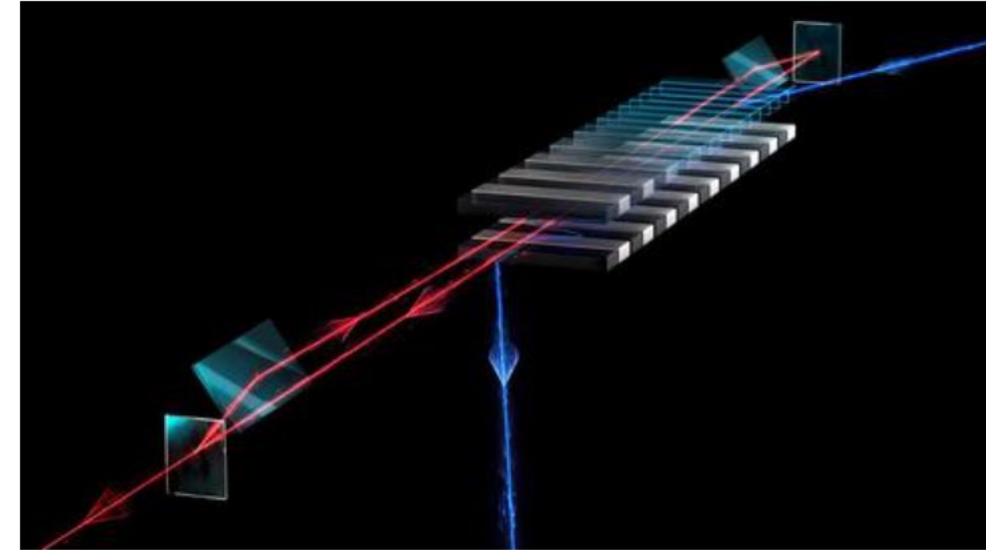
# Outlook I: Making an impact

- X-ray FELs are new research infrastructures providing research opportunities complementary to well-established SR sources.
- Very few facilities; the community is still small. Experiments gradually transitioning from “heroic” to “application oriented”.
- Future facilities designed for beam distribution to multiple beamlines & instruments w/standardized platforms
- **With more research opportunities, including for non-expert and smaller groups, XFEL user communities will reach the critical mass for XFEL science to make real impact on society.**

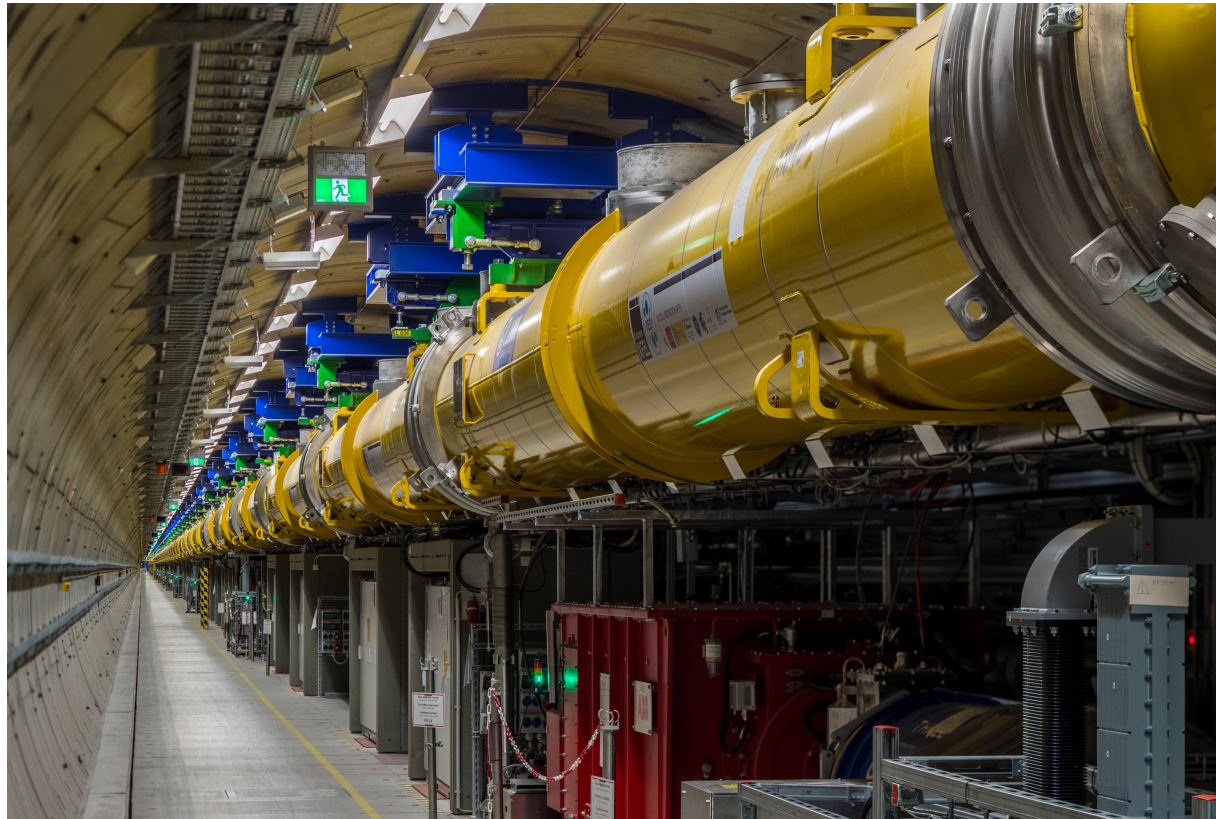


## Outlook II: X-ray science of the future

- With the lasing of our X-ray cavity, the EuXFEL and DESY have achieved one of the most ambitious goals in accelerator-based light source research.
- **Cavity based FEL may represent for X-ray science what the invention of the optical laser represented for optics: the transition from bright but incoherent light bulb to fully coherent laser radiation**
- Stable, fully coherent, spectrally pure hard X-ray pulses will **unlock regimes in quantum X-ray optics and enable transformative experiments**
  - meV-resolved inelastic and nuclear resonance spectroscopy
  - ultrafast pump–probe resonant inelastic X-ray scattering in the hard X-ray regime
  - nonlinear wave-mixing
  - .....



# Thank you for your attention !



# Additional slides

# Literature I – FEL theory, FEL schemes & first papers

## Firsts & proposals:

J.M.J. Madey, J. Appl. Phys. 42, 1906(1971)  
?. Deacon et al., Phys. Rev. Lett. (1977)  
J. Andruszkow et al., Phys. Rev. Lett. 85, 3825(2000)  
W. Ackermann et al., Nature Phot. 1, 336(2007)  
P. Emma et al., Nature Phot. 4, 641(2010)  
T. Ishikawa et al., Nature Phot. 6, 540(2012)

## Theory:

A.M. Kondratenko, E.L. Saldin, Part. Accel. 10, 207 (1980)  
R. Bonifacio, C. Pellegrini, L.M. Narducci, Opt. Communications 50, 373 (1984)

## Seeding & beam manipulation:

J. Feldhaus et al., Opt. Communications 140, 341(1997)  
P. Emma et al., Phys. Rev. Lett. 92, 074801 (2004)  
A.A. Zholents et al., N. Journal Phys. 10, 025005(2008)  
G. Geloni et al., J Mod Opt 58, 1391 (2011)  
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E. Allaria et al., Nature Phot. 6, 699 (2012)  
E. Allaria et al., Nature Phot. 7, 913 (2013)  
D. Ratner et al., Phys. Rev. Lett. 114, 054801 (2015)  
A.Lutman et al., Nature Phot. 10, 745 (2016)  
S. Liu et al. Nature Phot. 17, 984 (2023)  
P. Rauer et al., Phys. Rev. Accel. Beams 26, 020701 (2023)

# Literature II – FEL facilities & instrumentation

## Facilities:

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E. Allaria et al., J. Synchrotron Rad. 22, 485 (2015)

M. Yabashi et al., J. Synchrotron Rad. 22, 477(2015)

In Soo Ko et al., Appl. Sci. 7, 479 (2017)

Th. Tschentscher et al., Appl. Sci. 7, 592 (2017)

W. Decking et al., Nature Phot. 14, 391 (2020)

## Diagnostic:

K. Tiedtke et al., J Appl. Phys. 103, 094511 (2008)

Y. Feng et al., Appl. Phys. Lett. 101, 034103 (2012)

M. Beye et al., Appl. Phys. Lett. 100, 121108 (2012)

I. Grguras et al., Nature Phot. 6, 852 (2012)

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

J. Viefhaus et al., Nucl. Instruments Meth. A710, 151 (2013)

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

T. Guest et al., Journal of Synchrotron Radiation 29, 939-946 (2022)

## Optics & Damage:

T. Koyama et al., Optics Exp. 21, 015382 (2013)

I. Bahns et al., Communications Physics 7, 95 (2024)

# Literature III – FEL scientific applications (a selection)

## AMO:

- A.A. Sorokin et al., PRL 99, 213002 (2007)  
 L. Young et al., Nature 466, 56 (2010)  
 U. Eichmann et al., Science, 369, 1630 (2020)  
 R. Boll et al., Nature Physics, 18, 423–428 (2022)  
 I. Ismail et al., Physical Review Letters, 131, 253201 (2023)

## Chemistry:

- K.H. Kim et al., Nature 518, 790 (2015)  
 M. Minetti et al., Phys. Rev. Lett. 114, 255501 (2015)  
 S.E. Canton et al., Nature Commun. 6, 6359 (2015)  
 H. Öström et al., Science 347, 978 (2015)  
 M. Naumova et al. The Journal of Physical Chemistry Letters, 11, 2133 (2020)  
 R. Sension et al., Journal of the American Chemical Society, 145, 14070 (2023)

## Condensed-matter:

- M. Trigo et al., Nature Phys. 9, 790 (2013)  
 C. Weninger et al., Phys. Rev. Lett. 111, 233902 (2013)  
 M. Först et al., Solid State Commun. 169, 24 (2013)  
 J.A. Sellberg et al., Nature 510, 381 (2014)  
 C. Von Korff Schmising et al., Phys. Rev. Lett. 112, 217203 (2014)  
 F. Büttner et al., Nature Materials 20, 30 (2021)  
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 J. Möller et al., Physical Review Letters, 132 (20), 206102 (2024)

## Extreme states & plasmas

- B. Nagler et al., Nature Phys. 5, 693 (2009)  
 R.F. Fäustlin et al., Phys. Rev. Lett. 104, 125002 (2010)  
 S. Vinko et al., Nature 482, 59 (2012)  
 D. Kraus et al., Nature Commun. 7, 10970 (2016)  
 R. Husband et al. Journal of Synchrotron Radiation, 30 (4), 671-685 (2023)  
 L. Mercadier et al., Nature Physics, 20 (7) (2024)  
 M. Frost et al., Nat Astron 8, 174–181 (2024)  
 A. Laso Garcia et al. Nature Comm., 15 (1), 7896 (2024)

## Life sciences:

- R. Neutze et al., Nature 406, 752 (2000)  
 H.N. Chapman et al., Nature. Phys. 2, 839 (2006)  
 M.R. Howells et al., J. Elec. Spec. Rel. Phen. 170, 4 (2009)  
 H.N. Chapman et al., Nature 470, 73 (2011)  
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 J. Tenboer et al., Science 346, 1242 (2014)  
 T. Ekeberg et al., Phys. Rev. Lett. 114, 098102 (2015)  
 A. Aquila et al., Struct. Dyn. 2, 041701 (2015)  
 K. Ayer et al., Nature 530, 202 (2016)  
 M.L. Grunbein et al., Nature Communications, 9 (1), 3487 (2018)  
 M. Reiser et al. Nature Communications, 13 (1), 5528 (2022)  
 G. Tetreau et al. Nature Communications, 13 (1), 4376 (2022)

# FEL parameters

Full 3-dimensional FEL theory is extremely demanding

Simplified 1-dimensional approach

Pierce (FEL) parameter  $\rho$

$$\rho = \left( \frac{IA_{JJ}^2 K^2 \lambda_{rad}^2}{I_A 32 \pi^2 \gamma^2 \varepsilon_n \beta_f} \right)^{1/3} \quad \rho \sim 2-5 \times 10^{-4} (0.1 \text{ nm})$$

Number of cooperating electrons  $N_c$

$$N_c = (N_e \lambda_{rad} / L) / 2\pi\rho \quad N_c \sim 1/500 \times N_e (0.1 \text{ nm})$$

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)

$$\varepsilon < \lambda_{rad} / 4\pi \quad \curvearrowright \quad \varepsilon \sim 8 \text{ pm } (\lambda = 0.1 \text{ nm}) \quad \curvearrowright \quad \varepsilon_n \sim 0.22 \times 10^{-6} \text{ mrad } (15 \text{ GeV})$$

## ■ Energy spread

$$\sigma_e / E < \rho \quad \curvearrowright \quad \sim 10^{-4}$$

## ■ 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                          | $\mu\text{J}$ - mJ              |
| Photons per pulse ( $\sim 10$ keV) | $10^5 - 10^8$                    | $10^{10}$ - $10^{13}$           |
| Pulse duration / time-resolution   | 30 - 100 ps                      | 2 - 100 fs                      |
| Coherence degree ( $\sim 10$ keV)  | $< 10^{-3}$                      | 0.1 - 1                         |
| Rel. bandwidth (Undulator)         | $10^{-2}$                        | $10^{-3}$                       |
| Spectral characteristics           | continuous                       | spiked                          |
| Divergence (hor. $\times$ vert.)   | $1000 \times 10 \mu\text{rad}^2$ | $< 2 \times 2 \mu\text{rad}^2$  |
| Repetition rate                    | $\sim 100$ MHz                   | $\sim \text{MHz}$   10 - 100 Hz |
| Peak brilliance                    | $\sim 10^{24}$                   | $\sim 10^{33}$                  |
| Average brilliance                 | $\sim 10^{20}$                   | $\sim 10^{24}$   $\sim 10^{22}$ |

# Comparison of SR and FEL experiments

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

# Short recap – FEL applications focus on ...

- Ultrashort pulses (fs)
- High pulse energies (mJ)
- High transverse coherence (~100%)
- High peak & average brightness

| Field                               | Application | FEL property required |
|-------------------------------------|-------------|-----------------------|
| Condensed-matter                    |             |                       |
|                                     |             |                       |
|                                     |             |                       |
| Atoms, molecules, optical phenomena |             |                       |
|                                     |             |                       |
|                                     |             |                       |
| Materials                           |             |                       |
|                                     |             |                       |
|                                     |             |                       |
| Chemistry                           |             |                       |
|                                     |             |                       |
| Life science                        |             |                       |
|                                     |             |                       |

# Damage by intense x-ray pulses

While most SR experiments are performed in a regime in which the x-ray pulse is minimally interacting with the sample, FEL experiments often depart from this regime.

Need to estimate x-ray – matter interaction

Experiment specific sample response

Ultrafast (fs): change of probed system

Fast (ns-μs): diffusion, expansion, ablation

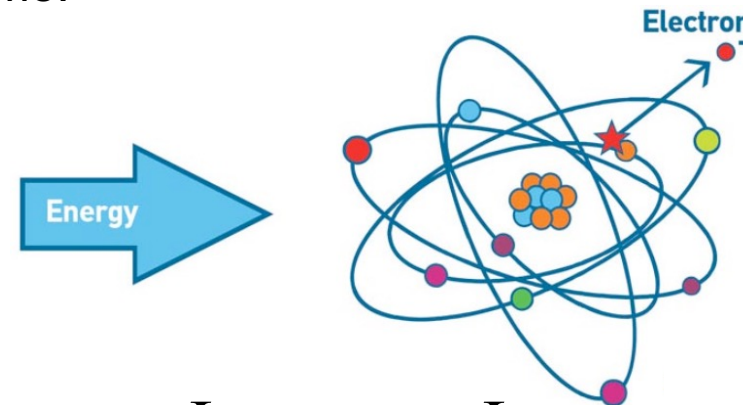
Minimize absorption !

Use low Z materials (optics, absorbers)

Optimize sample geometry

Reduce pulse intensity

Reduce pulse duration



$$I_{scatter} \ll I_{absorb}$$

At ~12 keV for bio-samples (C, H, N, O)  $I_{scatter} \sim 0.1 I_{absorb}$

Absorption generates photo-electrons → damage

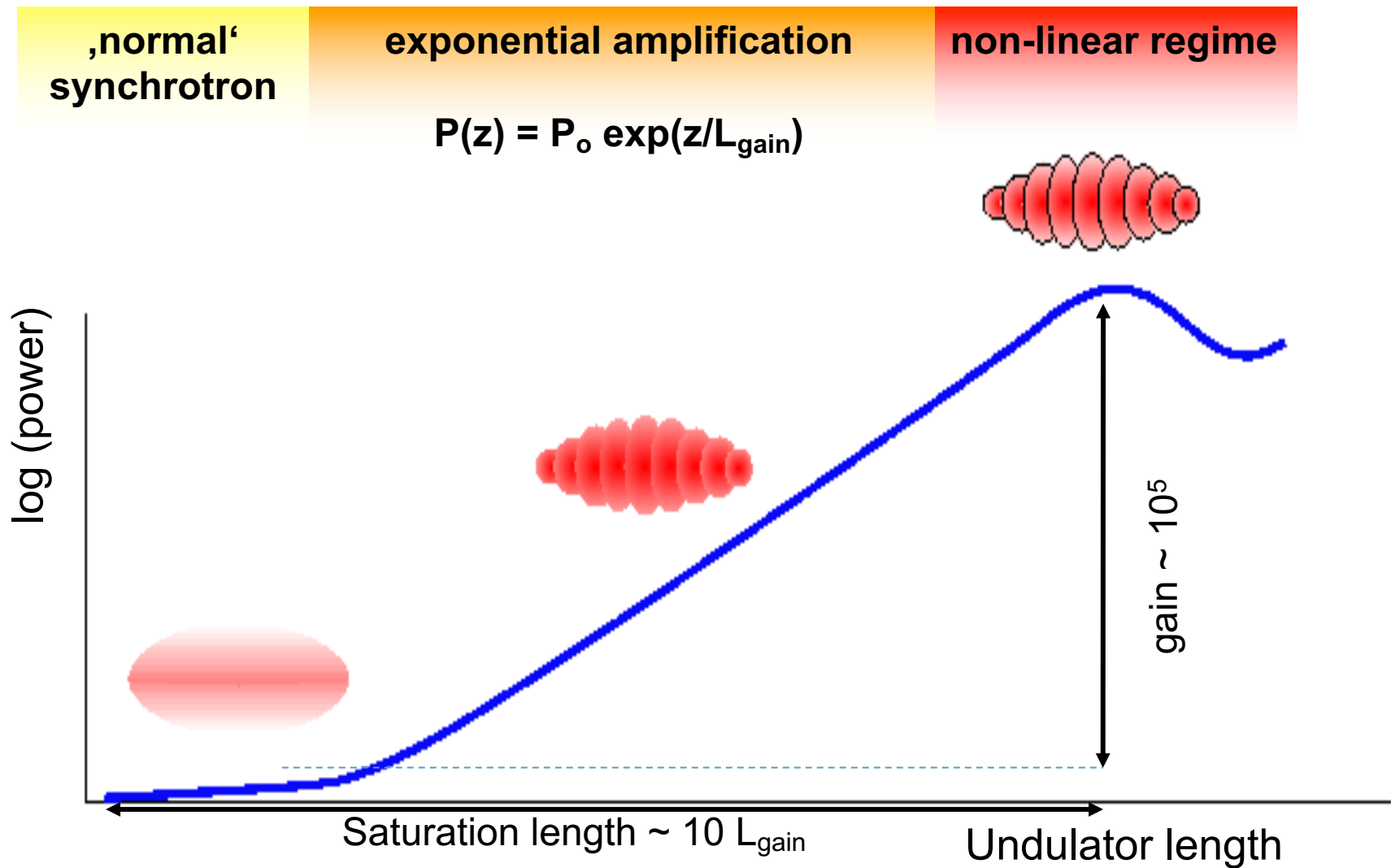
Cascading generates add. electrons → more damage

1<sup>st</sup> photo-electron

2<sup>nd</sup> Auger-electron

Many electrons by inelastic electron scattering

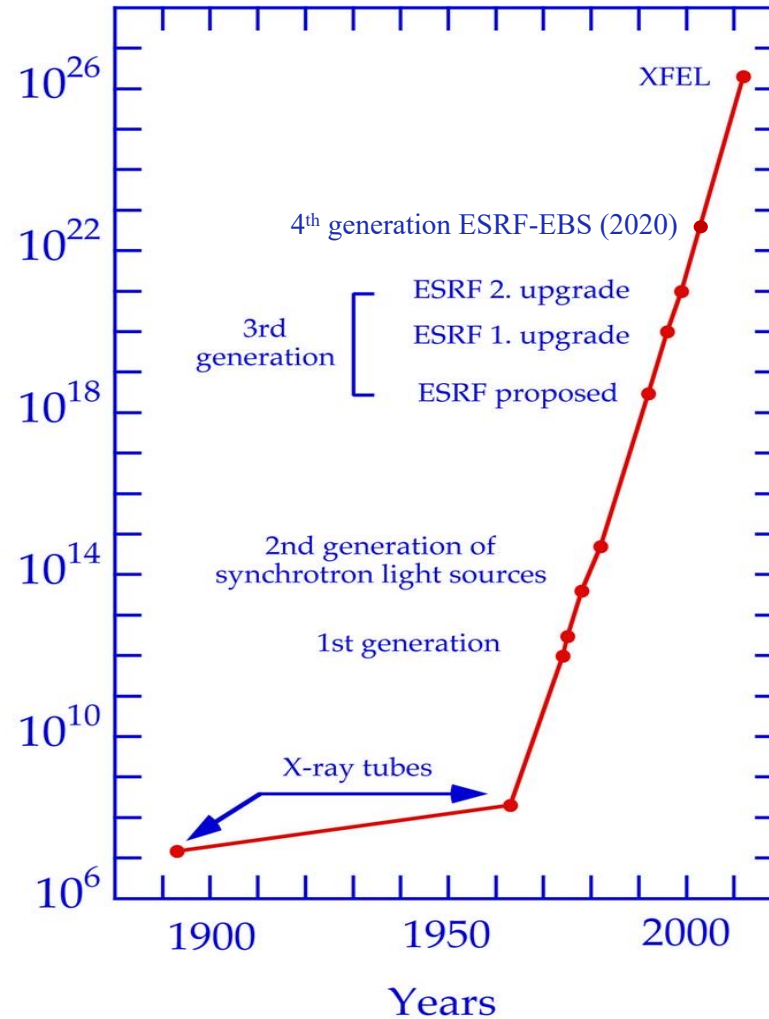
# FEL gain



# Accelerator based X-ray facilities



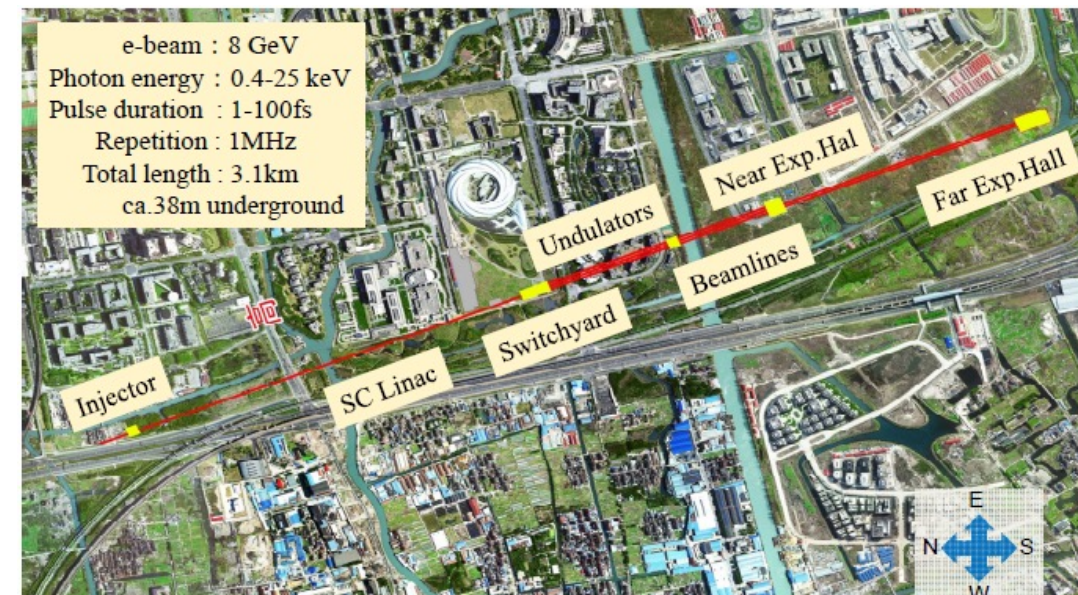
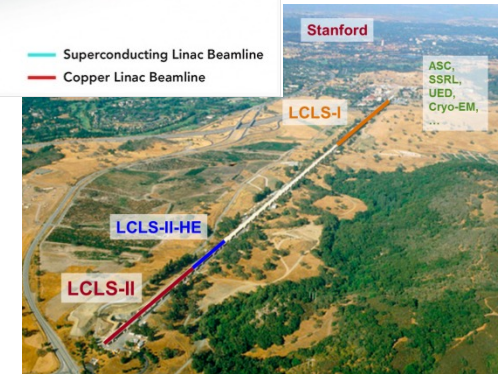
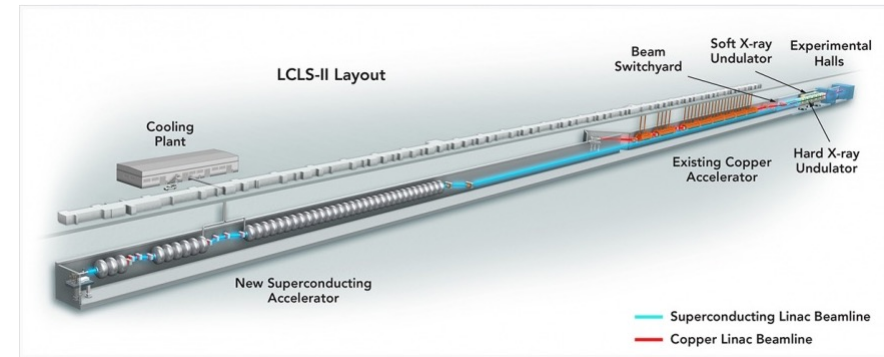
Average Brilliance [Phot./ (sec · mrad<sup>2</sup> · mm<sup>2</sup> · 0.1% bandw.)]



# Towards CW-type FELs

- CW == high repetition rate continuous wave accelerators
  - Superconducting RF to deal with power losses
  - High rep. rate low emittance injectors
  
- First project is LCLS-II – operating now
  - Using know-how and technology developed for European XFEL
  - 4 GeV → soft x-rays
  - 8 GeV → coming
  
- Second project is Shanghai Coherent Light Source (SHINE)
  - 8 GeV → soft and hard x-rays
  
- European XFEL design enables upgrade to CW-operation

■ ■ European XFEL

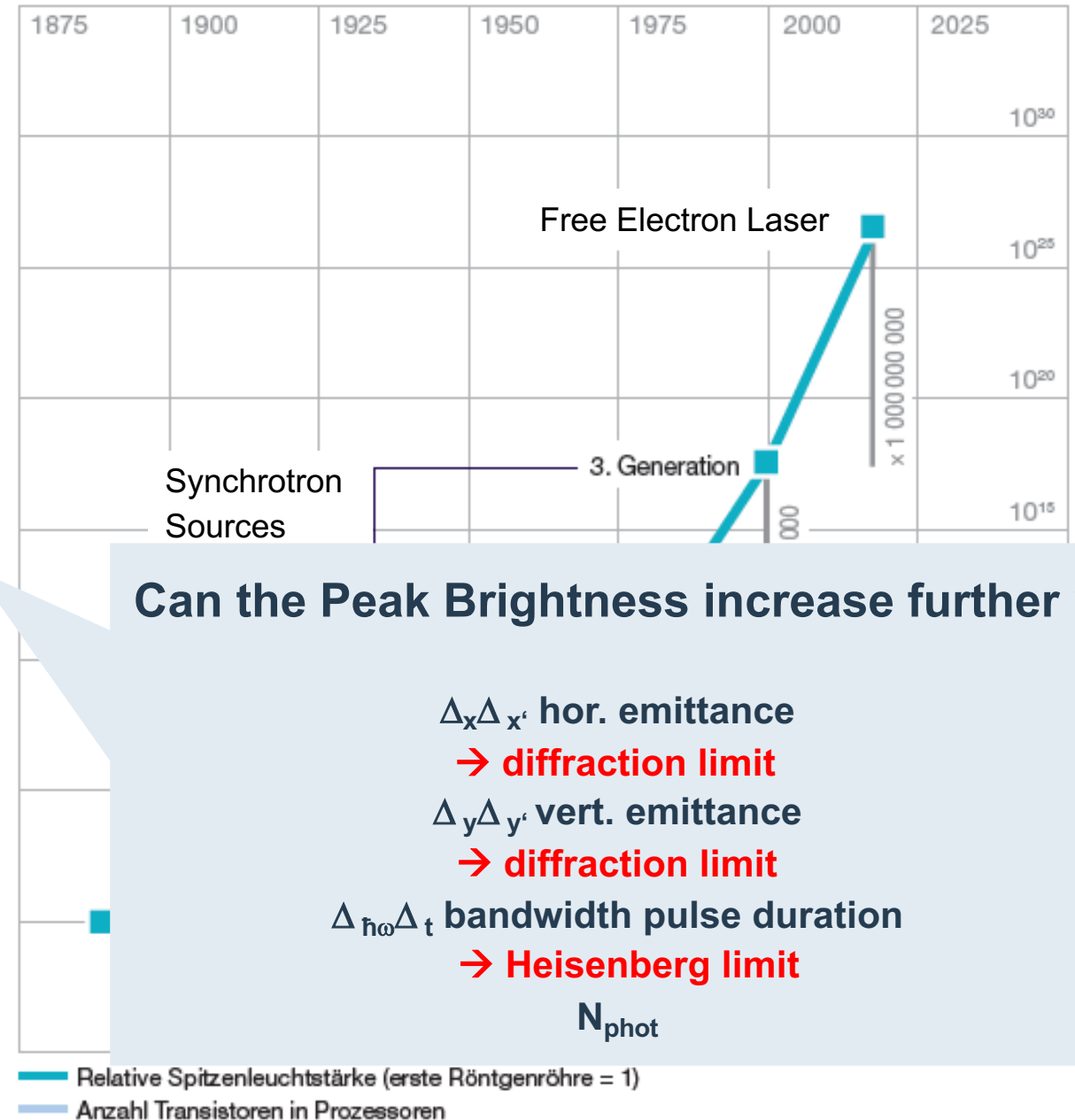


# Development of peak brightness

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

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

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



# 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 understanding 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 !