Exploring stochastic nature of X-ray pulses from X-ray Free Electron Laser

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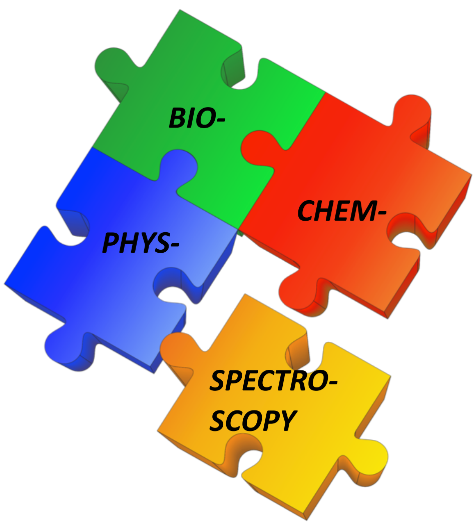
Krakow, Poland

Let us introduce:

Department of Applied Spectroscopy NZ53 (from 11.2019)
Division of Interdisciplinary Research (NO5)

X-ray spectroscopy for Interdisciplinary Research

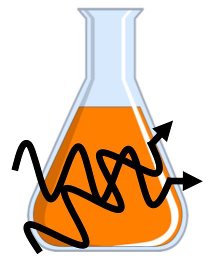


- ➡️ X-ray spectroscopy applied to **biological** and **environmental** science (DNA damage and repair mechanisms, interaction of metal-centred molecules with biological material)
- ➡️ X-ray spectroscopy applied to **chemical** systems (charge transfer processes, chemical transformations in real-time)
- ➡️ X-ray spectroscopy applied to **fundamental** interactions (core-core excitations, nonlinear interactions)

X-rays & X-ray spectroscopy – Why⁉

X-rays:

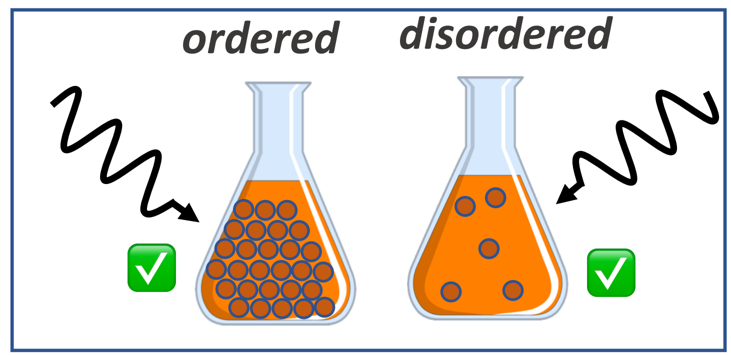
- **penetrating properties**:
  ambient environment
  no sample pre-treatment
  solid & gas & liquid state



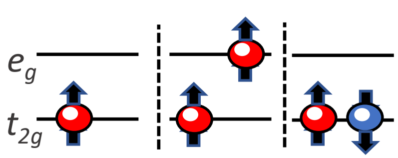
X-rays & Spectroscopy:

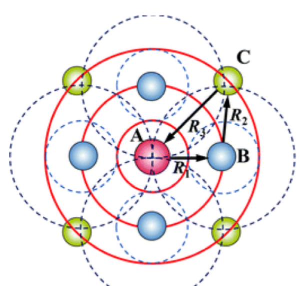
- **element selectivity** by energy

Sample complexity



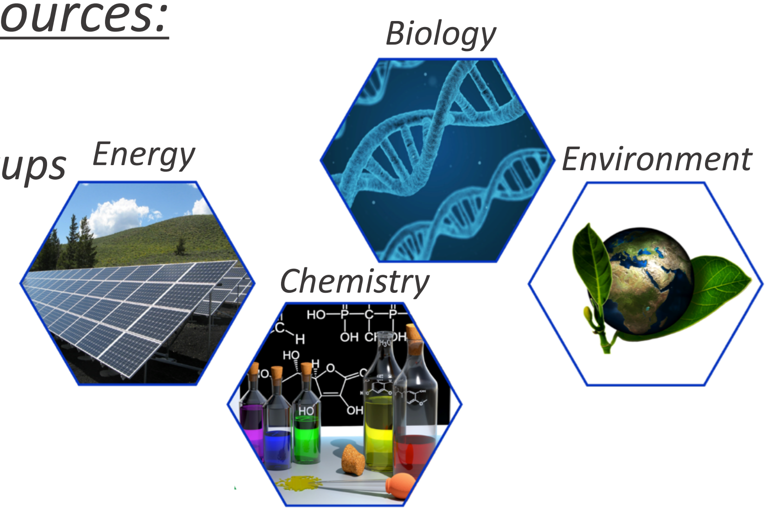
- **structural information** at atomic levels
  electronic properties, oxidation state
  spin configuration, chemical environment





Department of Applied Spectroscopy NZ53
Methods & X-ray Sources:

Laboratory X-ray spectrometers/methods
(X-ray absorption and emission spectrometer setups sample delivery, sample cells development)



3rd and 4th generation X-ray sources:

Synchrotron research
(spectroscopy on biological and chemical systems)

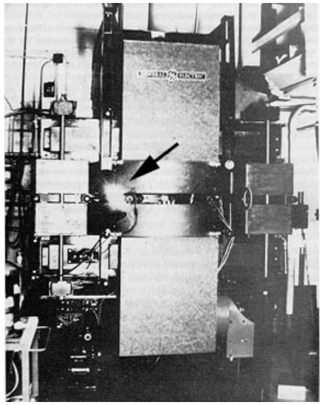
Science with **X-ray Free Electron Lasers**
(dynamics, nonlinear interaction)

X-ray sources:
from Synchrotron Facility to X-ray Free Electron Laser

Four generations of Synchrotron sources

Synchrotron radiation discovered thanks to the experiments in the field of **High Particle Physics (1947)**.

Synchrotron light from the 70-MeV electron synchrotron at GE.



1st generation sources: ➡ exploring storage rings "background"
- electron storage rings
- first X-ray absorption spectra

1950s

2nd generation sources:
➡ towards dedicated sources
- higher electron current
- insertion devices

1970s

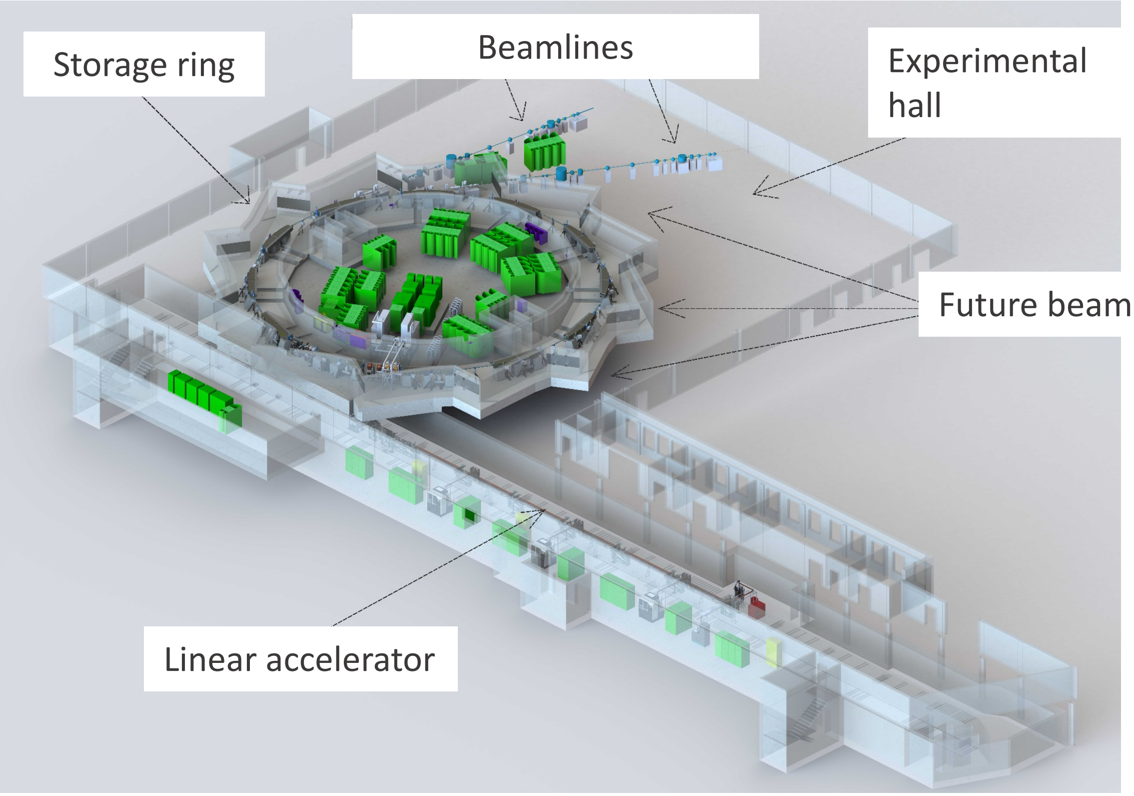
3rd generation sources:
➡ optimized and bright sources
- optimization of ring technology
- high current and low emittance

1990s

2010

4th generation sources:
➡ Hard X-ray Free Electron Laser

Synchrotron facility – 3rd generation storage ring
Solaris Synchrotron in Kraków

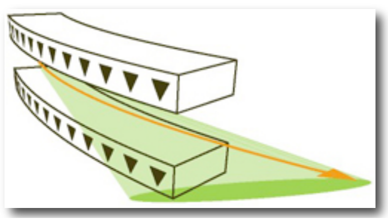


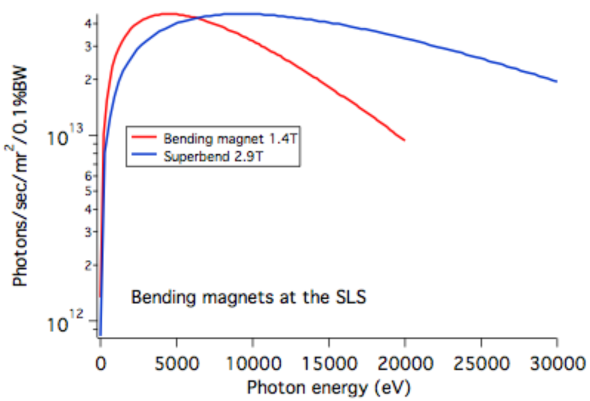
(courtesy of prof. M. Stankiewicz)

Synchrotron radiation

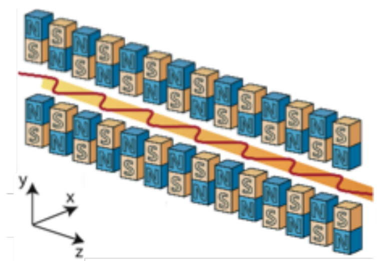
There are two methods of generating light at 3rd generation storage ring sources:

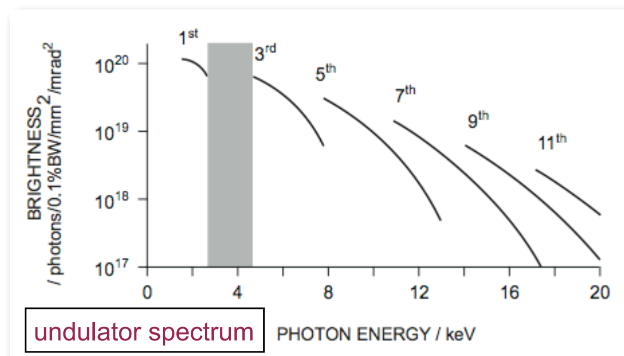
Bending magnet





Insertion devices





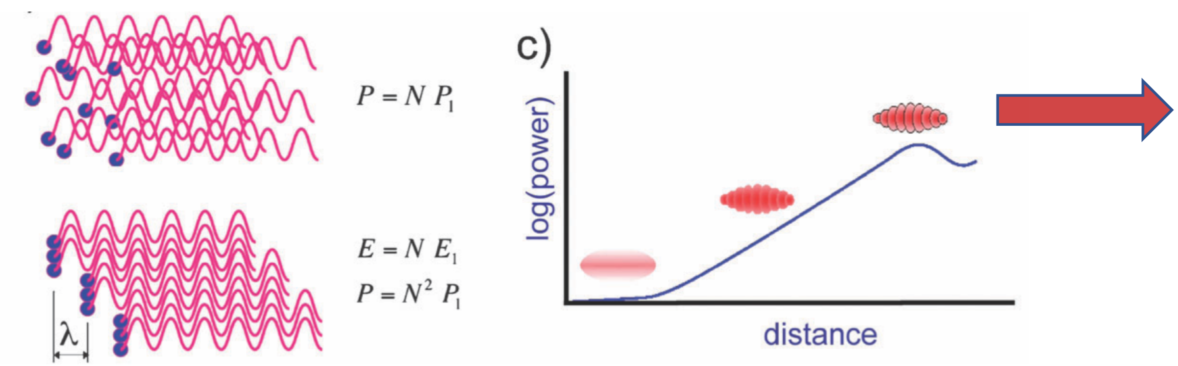
From Synchrotron to X-ray Free Electron Laser

Receipt: relativistic electron beam and long undulator section

A. Kondratenko, E. Saldin, Part. Accelerators10, 207 (**1980**)
Generation of coherent radiation by a relativistic electron beam in an ondulator

R.Bonifacio et al, Opt. Comm.50, 373 (**1984**)
The superradiant regime of a free electron laser

Self Amplified Spontaneous Emission (SASE) and micro-bunching effects

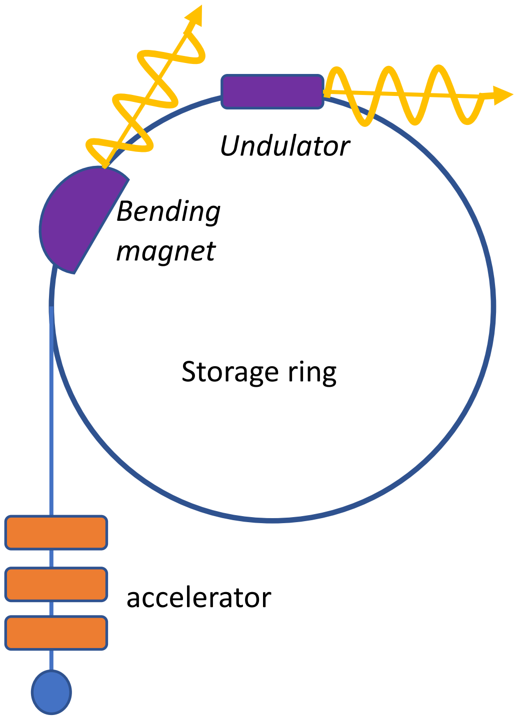


Micro-bunching causes the entire e-bunch to emit coherently:
Extremely intense and short X-ray laser pulse

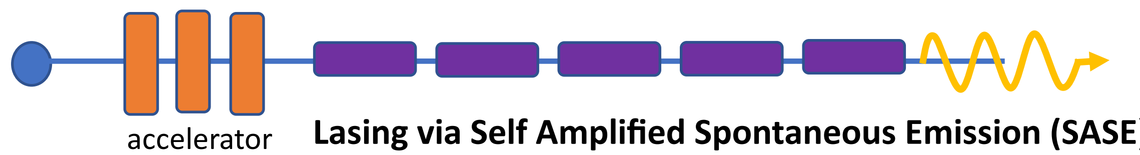
B. D. Patterson and R. Abela Phys. Chem. Chem. Phys., 2010, 12, 5647-5652

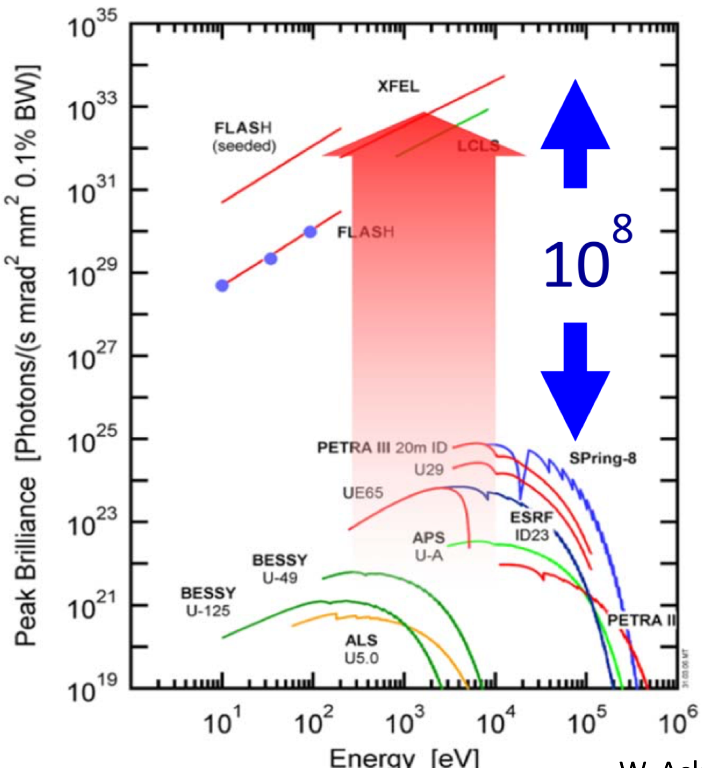
3rd vs. 4th generation X-ray sources

Synchrotron



LINAC – X-ray Free Electron Laser





X-ray flux gain!!

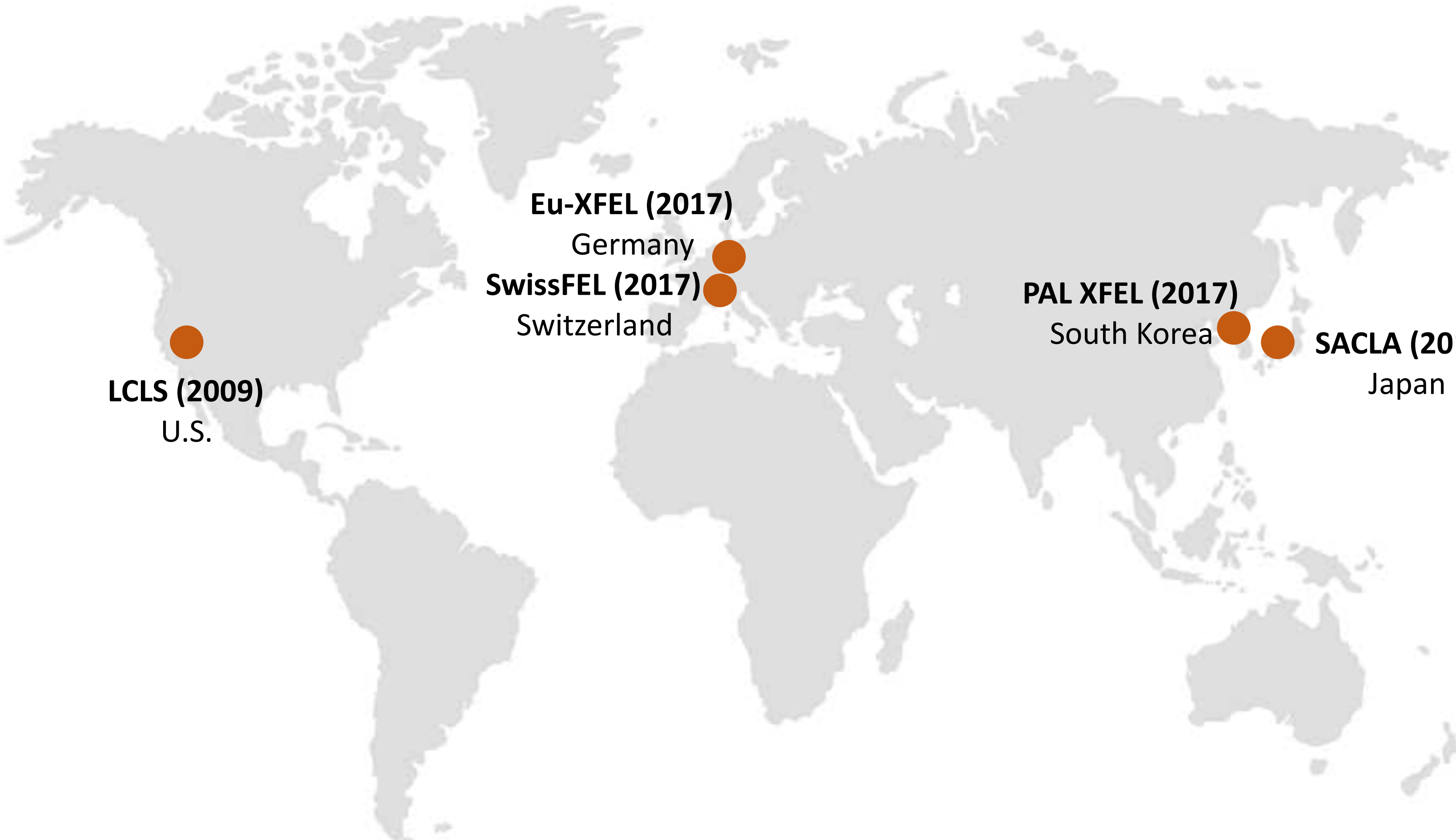
W. Ackermann et al Nat. Photon. 1 336 (2007).

X-ray Free Electron Laser: laser-like pulse properties

*Extremely **intense** and extremely **short** (femtoseconds).*

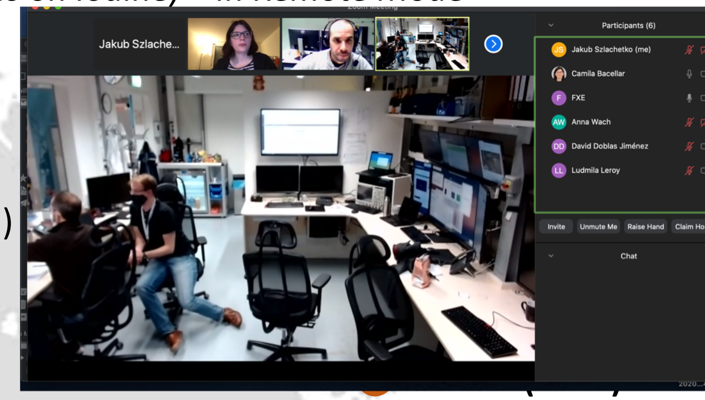
Pulse length defines experimental time resolution

X-ray Laser Guns worldwide



X-ray Laser Guns worldwide

Experiment at FXE station finished on Monday (charge dynamics on Iodine) – In Remote Mode



Nonlinear spectroscopy

(2016-)
J.S, J. C-M.
nature COMMUNICATIONS
scientific reports
LCLS (2009)
U.S.

Eu-XFEL (2017)
Germany
(2017-..)
J.S, W.B. A.W.

SwissFEL (2017)
Switzerland
(2017-..)
J.S, J. C-M., W.K., A.W. W.B.

DNA damage & repair
OLED devices

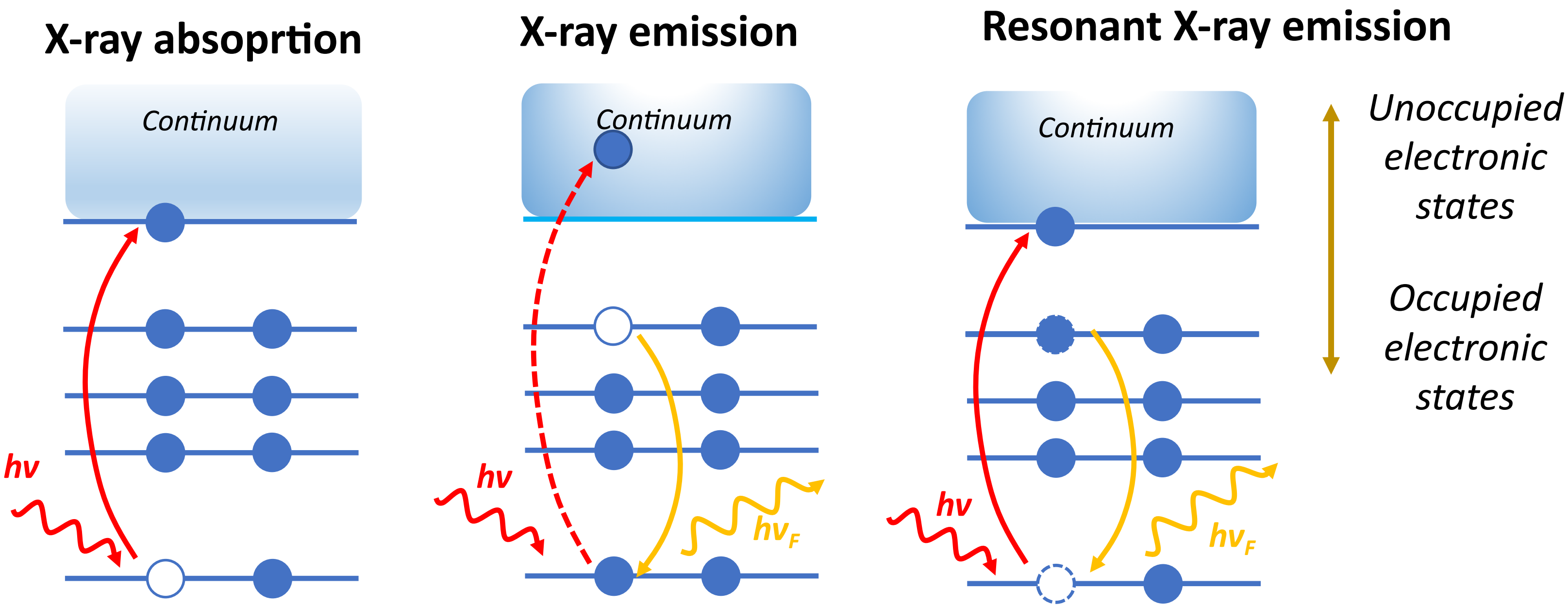
Japan
(2018)
J.S
nature COMMUNICATIONS
Molecular dynamics


PSI Newsletter, IFJ News
Lightsources News, Fels-of-Europe,
nature COMMUNICATIONS
PNAS

J.S. – Jakub Szlachetko
W.K. – Wojciech Kwiatek
J.C-M. – Joanna Czapla Masztafiak
W.B. – Wojciech Błachucki
A.W. – Anna Wach

X-ray Free Electron Lasers: The Jitter challenge

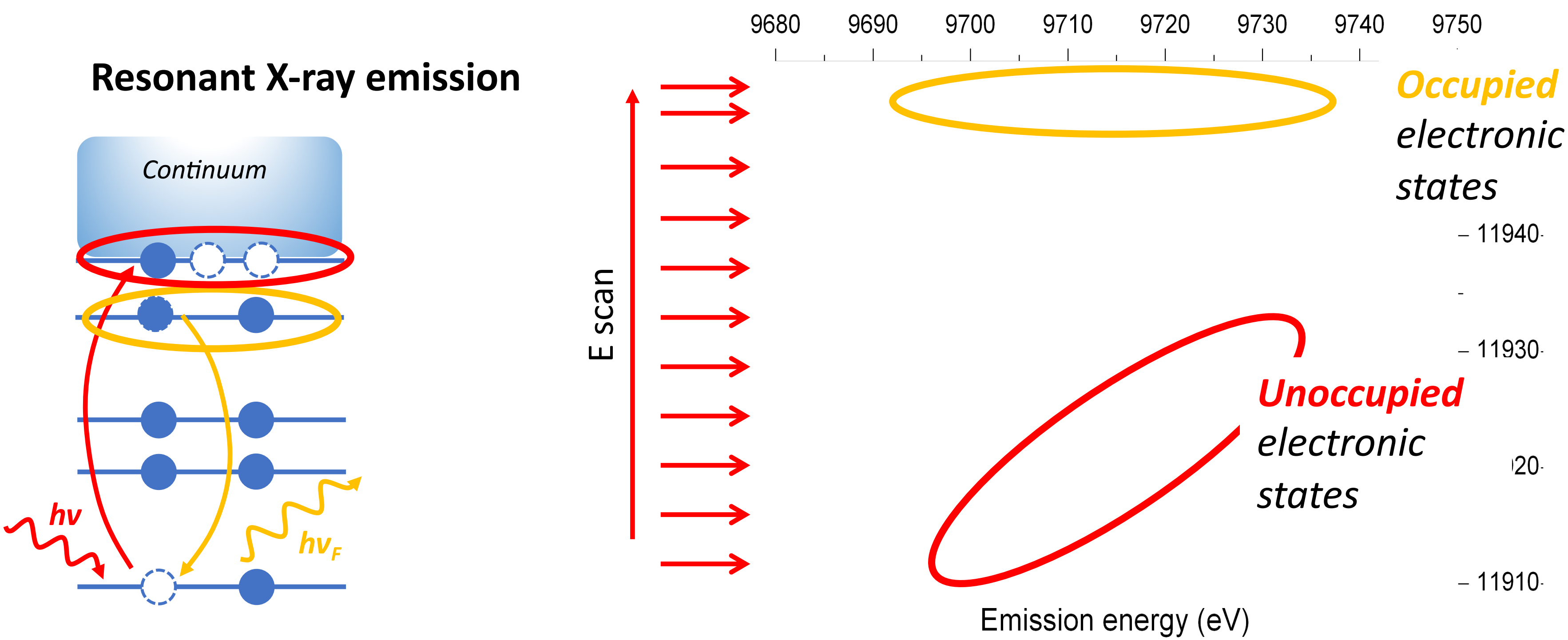
X-ray spectroscopy methods requirements
High energy resolution X-ray absorption and X-ray emission



Full electronic picture in single experiment
Electronic states in few eV range round Fermi
Multi-electron interactions

Resonant X-ray emission spectroscopy:

Use of stable, monochromatic incidence X-rays from 3rd generation sources



Recent highlights on RXES: J. Szlachetko et al., *Mater.Horiz., 5, 905—911 (2018). J. Czapla-Masztafiak et al. J. Phys. Chem. Lett. (2018),* W. Błachucki et al., JAAS 34, 1409 (2019), J. Szlachetko et al., Nat. Comm. (2019)., W. Błachucki et al., Struct. Dyn. 6, 024901 (2019).

X-ray Free Electron Lasers: The Jitter challenge

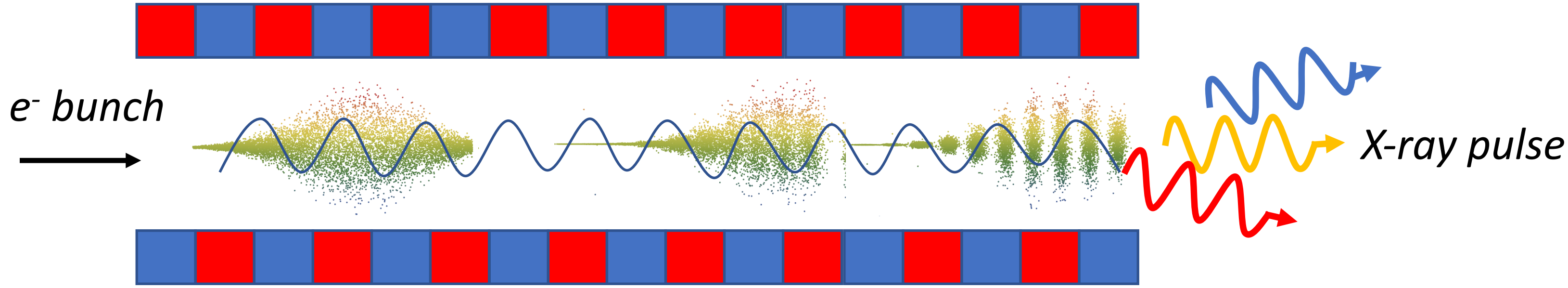
Linear Accelerator based **S**elf **A**mplification of **S**pontaneous **E**mission (**SASE**)

Electron bunch modulated with its own synchrotron radiation filed:

➡️ micro-bunching

➡️ more and more electrons radiate in phase (until saturation)

Electron beam quality (initial conditions) is reflected and enhanced in emitted X-ray pulse:



Stochastic nature of SASE results in X-ray beam jitter:

➡️ undefined **position** (tens of um)

➡️ undefined **arrival time** (100s of fsec)

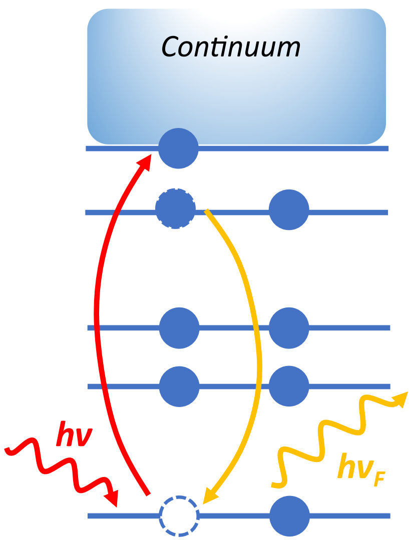
➡️ undefined **energy distribution** (tens of eV)

X-ray Free Electron Lasers: The Jitter challenge

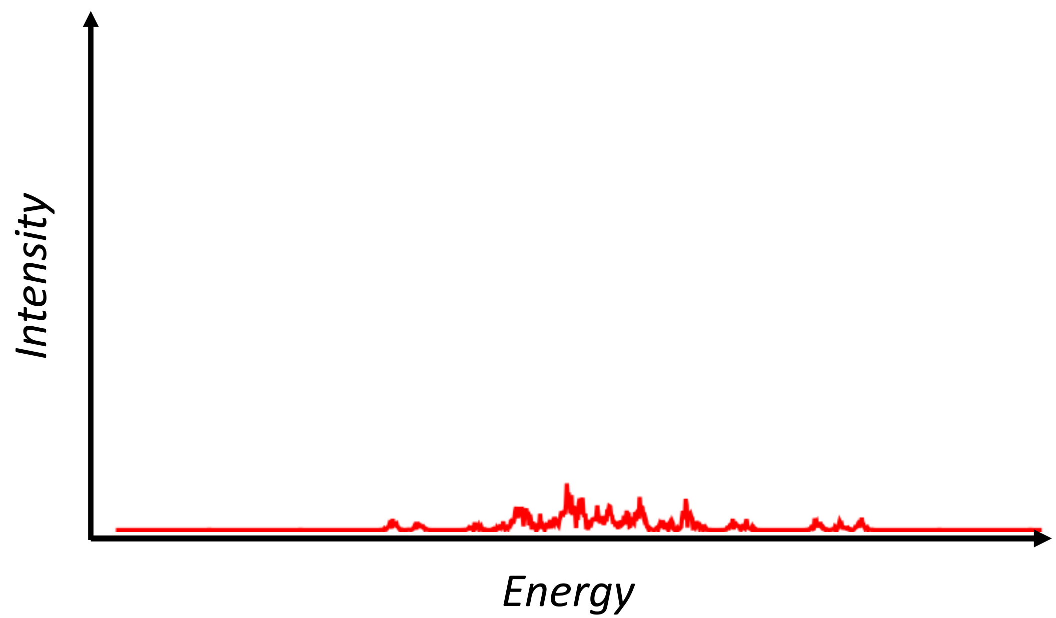
Linear Accelerator based **S**elf **A**mplification of **S**pontaneous **E**mission (**SASE**)

Stochastic nature of SASE results in X-ray beam jitter:
- undefined **energy distribution** (tens of eV)

Resonant X-ray emission



30 fsec long X-ray pulse



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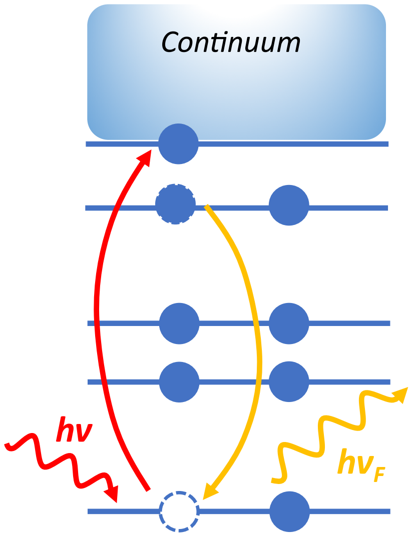
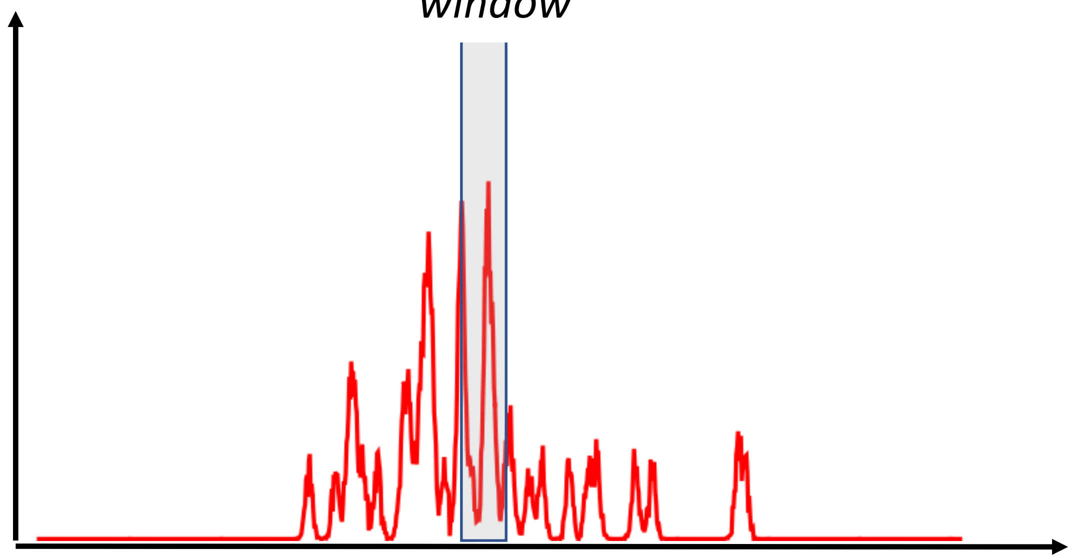
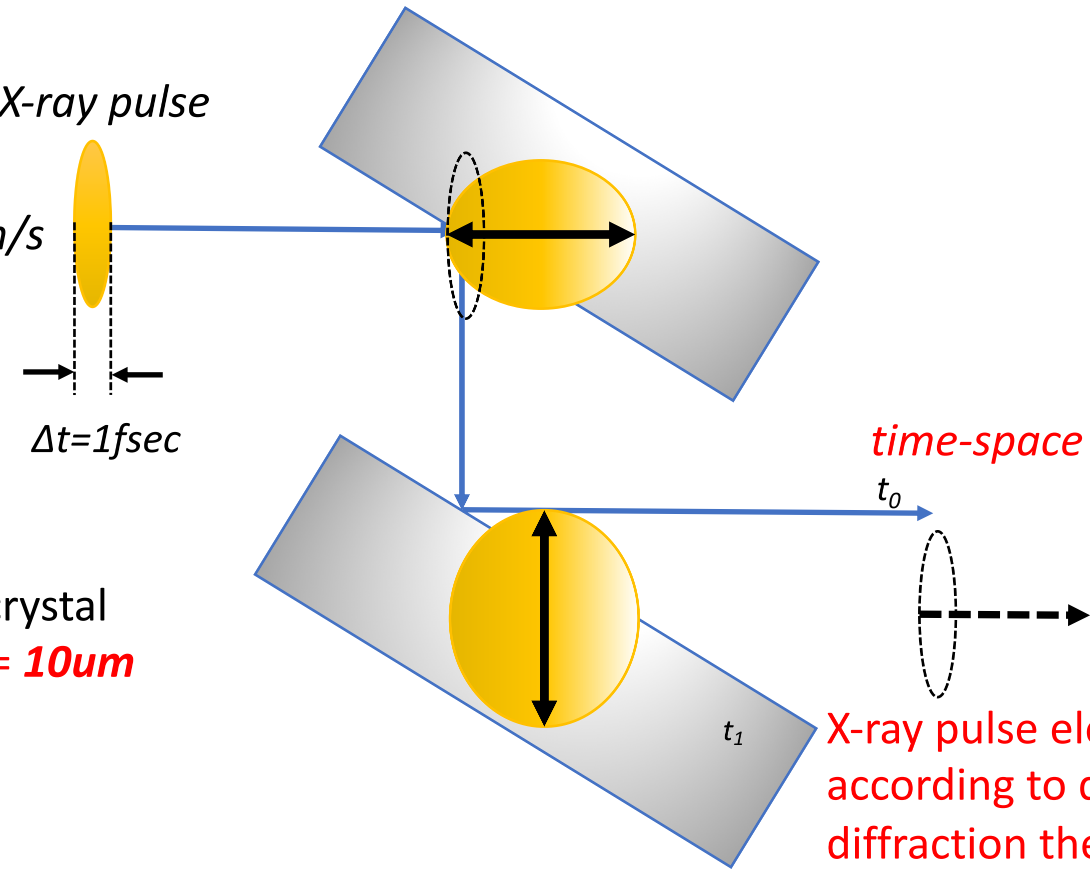




X-ray Free Electron Lasers: The Jitter challenge
Temporal diffraction limit in X-ray optics (time-space relation)

Time domain:

Speed of light = 300 000 000 m/s
= 0.3um / fsec



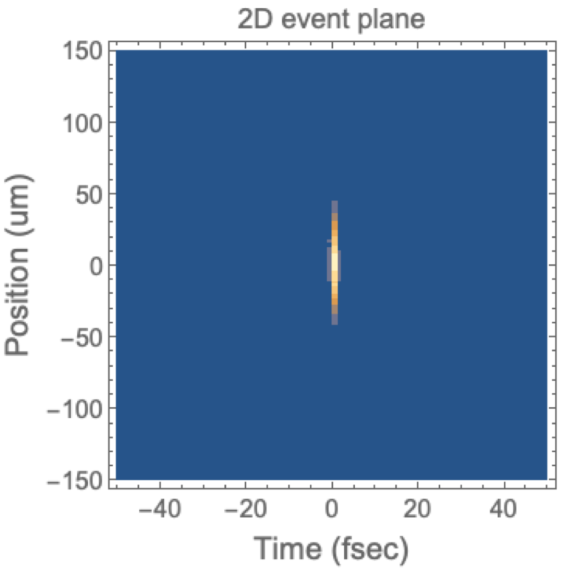
Monochromator crystal
Extinction length = ***10um***

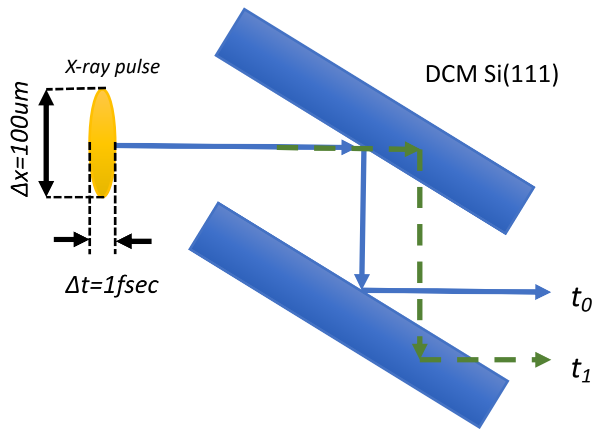
X-ray pulse elongation according to dynamical diffraction theorem

X-ray Free Electron Lasers: The Jitter challenge
Temporal diffraction limit in X-ray optics

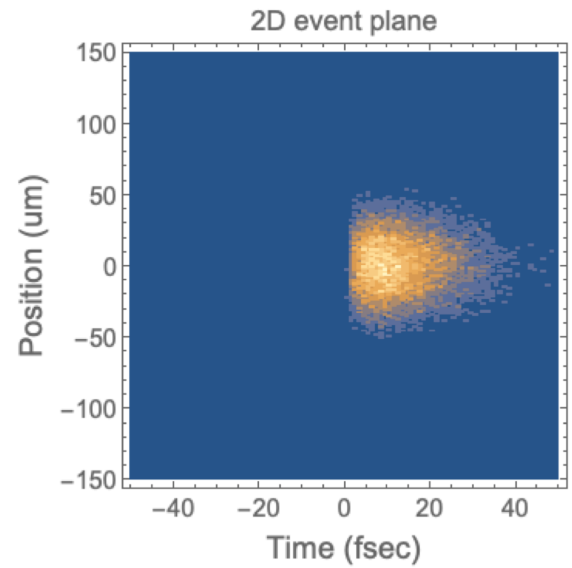
Geometrical considerations

before monochromator





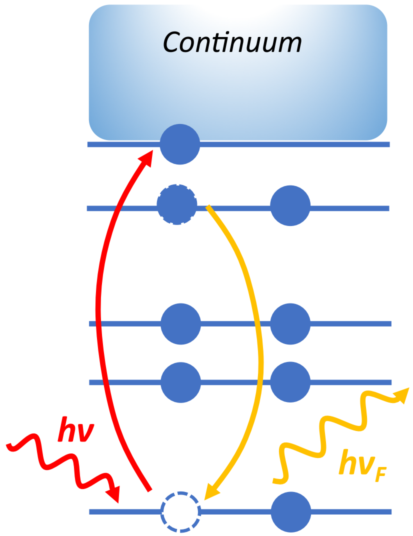
after monochromator

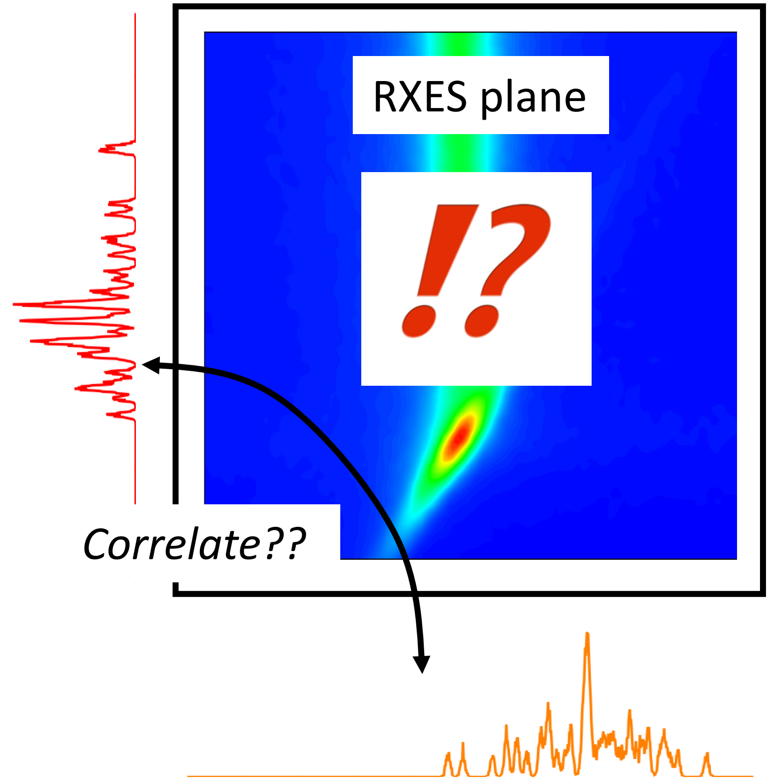


Bushuev, V. et al., Proc. SPIE 8141, 8141 (2011).
J.S. Wark and H. H. E,Laser and Particle Beams 12, 507-513 (1994).

Few tens of fsec!!!

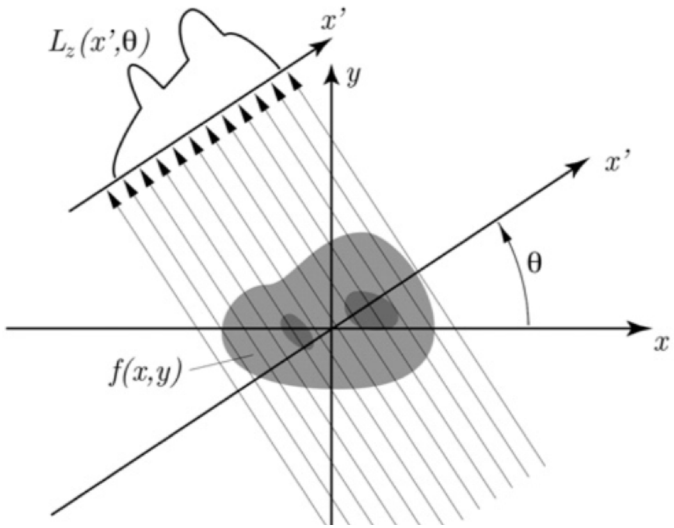
Can we get 2D image of electronic state with stochastic X-ray pulses?
Without monochromator optics?



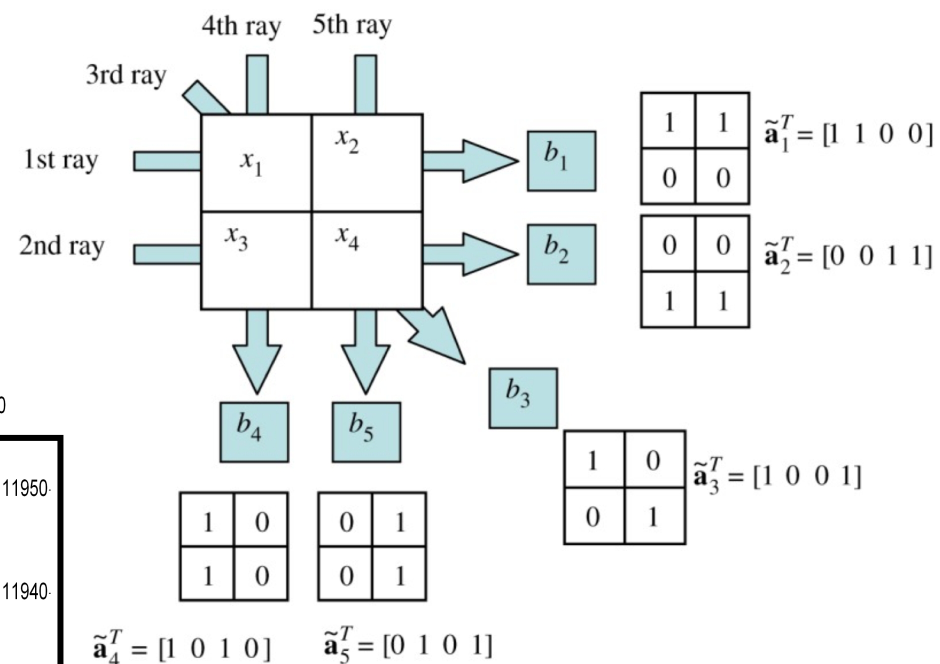


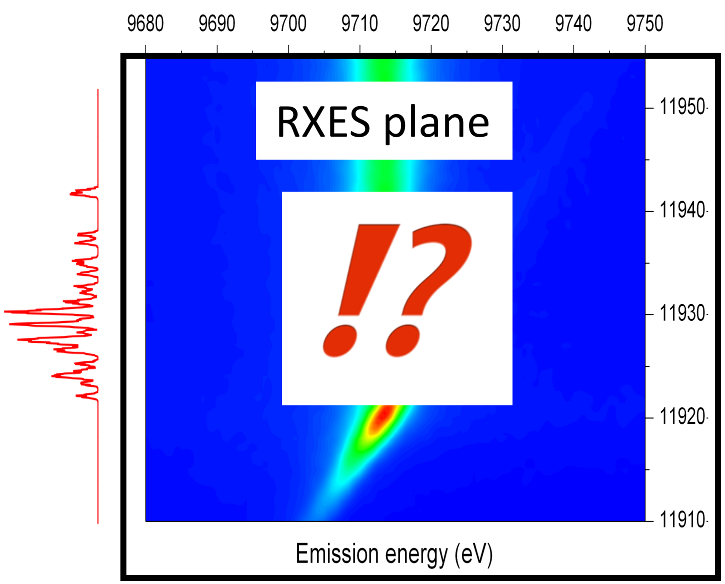
***YES:** precise monitoring of incidence and emission spectra*
all measured on shot-to-shot basis
with sufficient number of X-ray pulses

Reconstruction procedures in X-ray tomography



Iterative reconstruction algorithm

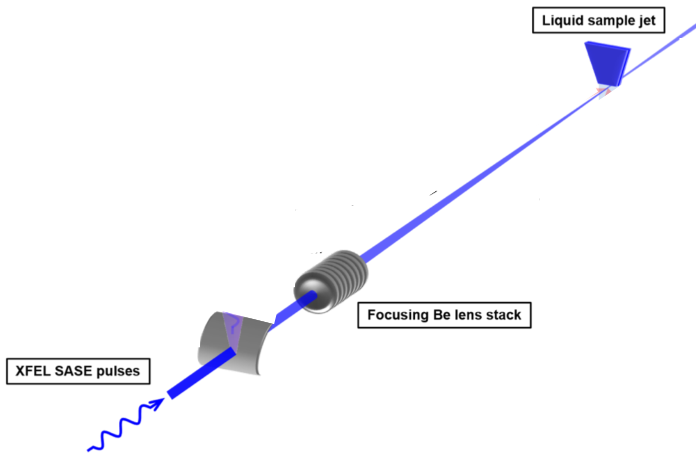




https://www.ncbi.nlm.nih.gov/

Experimental setup at XPP station of LCLS

Linac Coherent Light Source (LCLS)
X-ray pump-probe (XPP) instrument

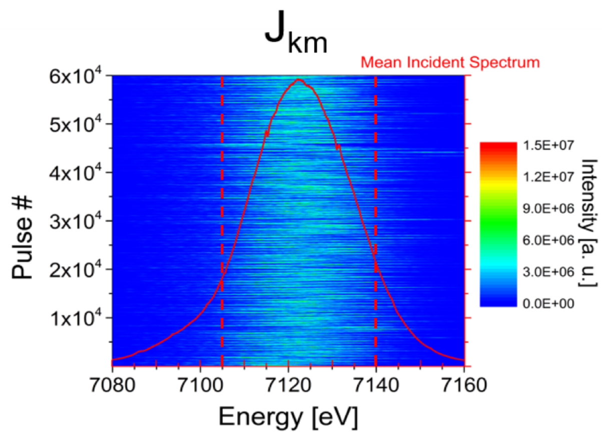


X-ray beam properties:
- self-amplified spontaneous emission (SASE)
- repetition rate: 120 Hz
- mean photon energy: 7100 eV (around Fe K-edge)
- pulse duration: 5fs & 30 fs
- number of photons per pulse: 10^{12}
- beam spot area on the target changed from 6 to 241 μm^2 with Be lenses

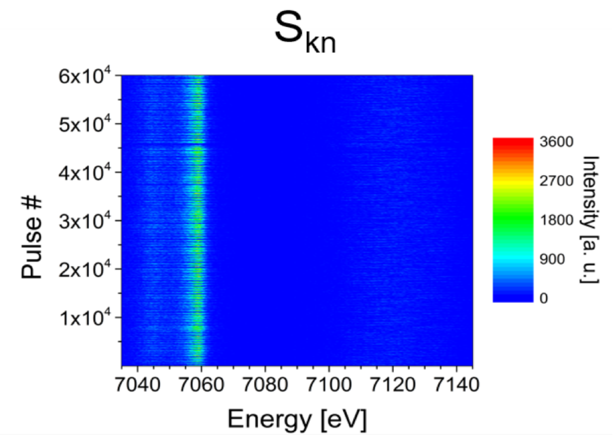
Target:
aqueous dispersion of Fe_2O_3 nanoparticles

Experimental results – use of stochastic X-rays for 2D imaging of electronic states

X-ray incidence beam



X-ray emission spectra

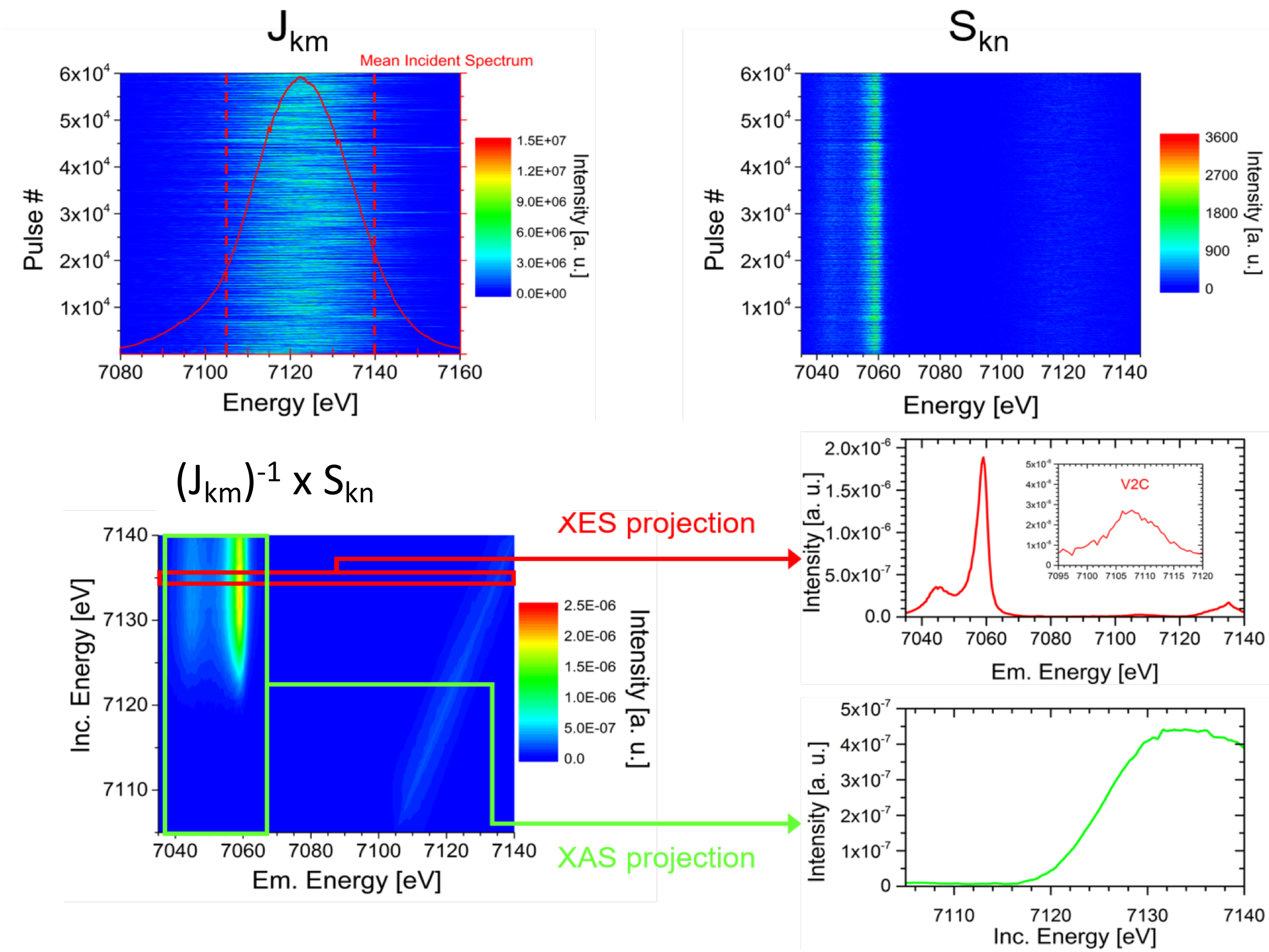


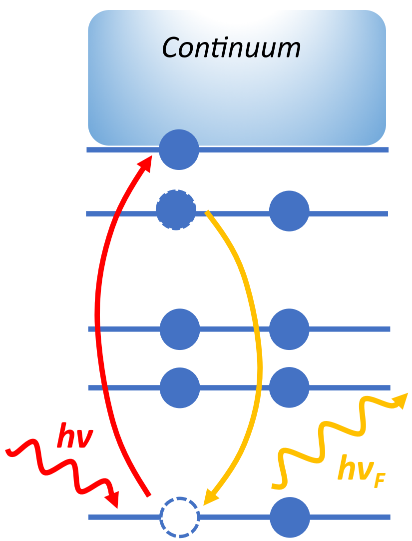
Deliberate use of stochastic process: **measure** instead of **control**!

Nature Communications 10, 4761 (2019) & EurekAlert! AAAS *(2020).*

Experimental results – use of stochastic X-rays for 2D imaging of electronic states

X-ray incidence beam *X-ray emission spectra*





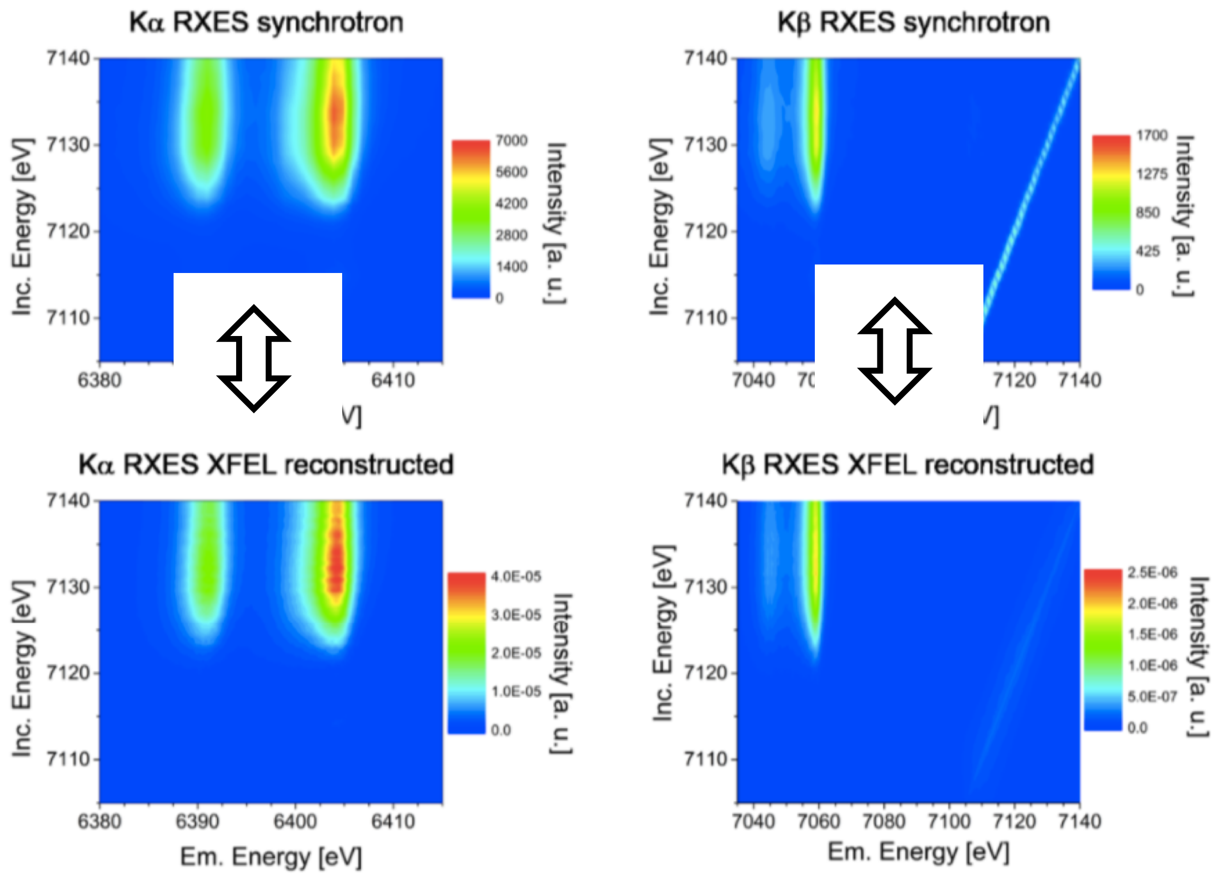
Experimental results – comparison to synchrotron data

Synchrotron

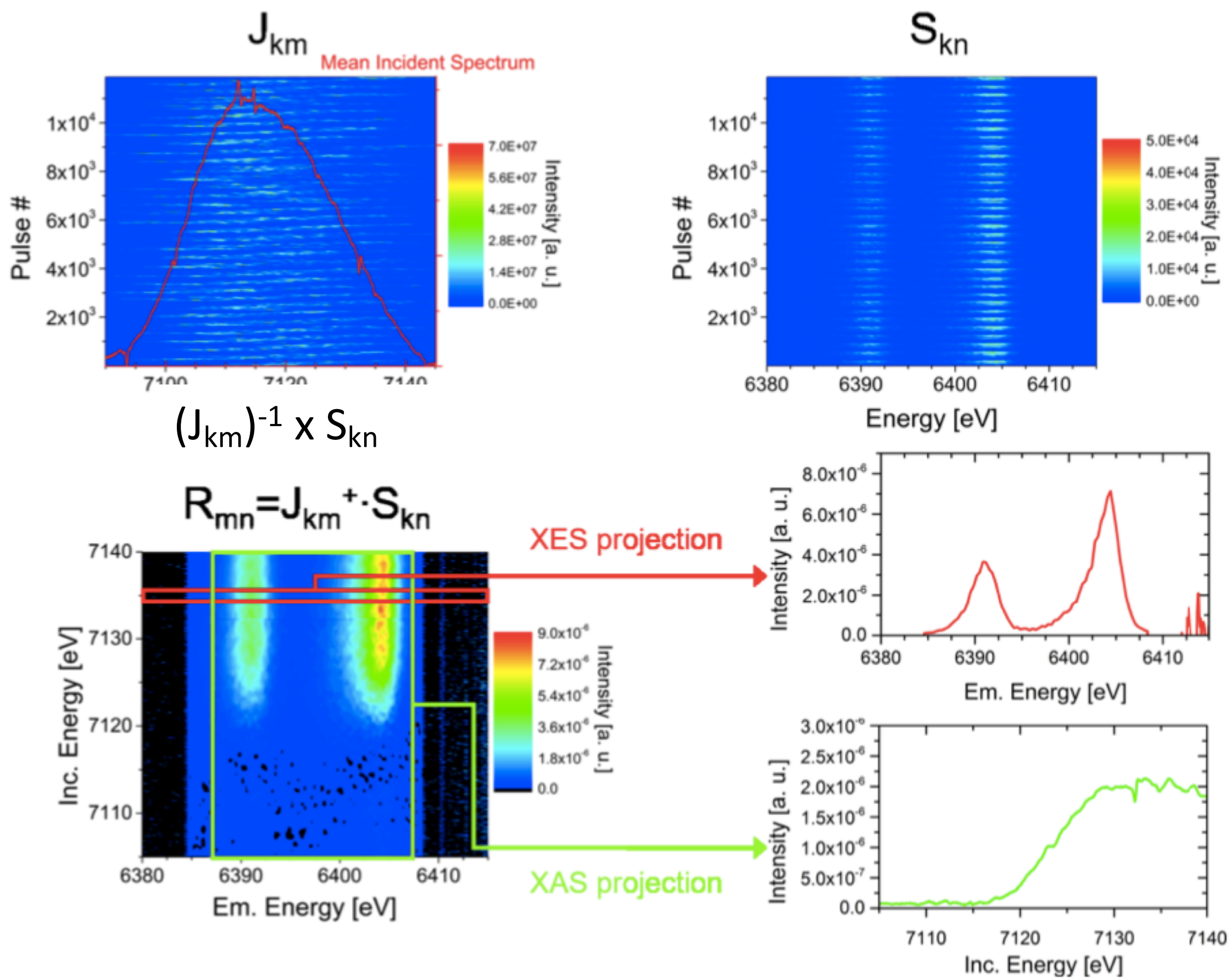
Monochromatic excitation

XFEL

Stochastic excitation



Experimental results – going beyond diffraction limit
Electronic states determination with 5fsec-long X-ray pulse



Towards nonlinear 2D spectroscopy with stochastic X-ray pulses

What types of experiments are X-ray Free Electron Lasers good at ?

XFELs are defined by lots of photons in a very short pulse but the average flux isn't that different from a 3rd-generation synchrotron

Only three types *of experiments benefit from the high peak flux from an XFEL:*

Single-shot *experiments that need lots of photons in a short pulse*
Pump-probe *measurements where the short pulse for measurement of fast dynamics*
Nonlinear *X-ray experiments that depend nonlinearly on the incident X-ray flux*

X-ray spectroscopy

Linear X-ray spectroscopy (absorption & emission):

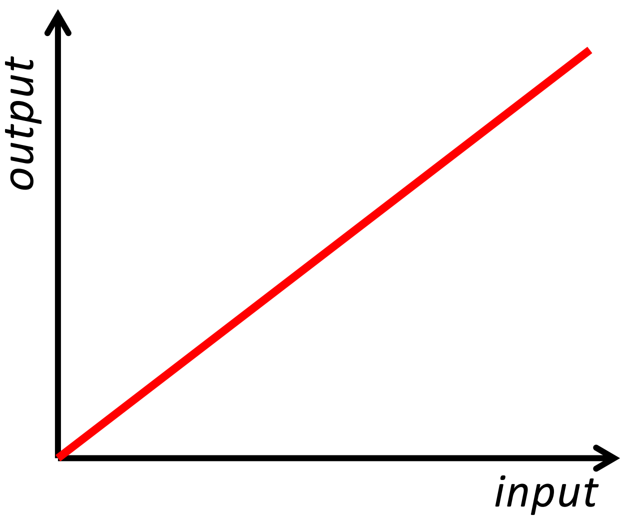
- weak limit interaction of X-rays with matter
- measurements are performed in *one-photon-in one-photon-out* manner

Nonlinear X-ray spectroscopy

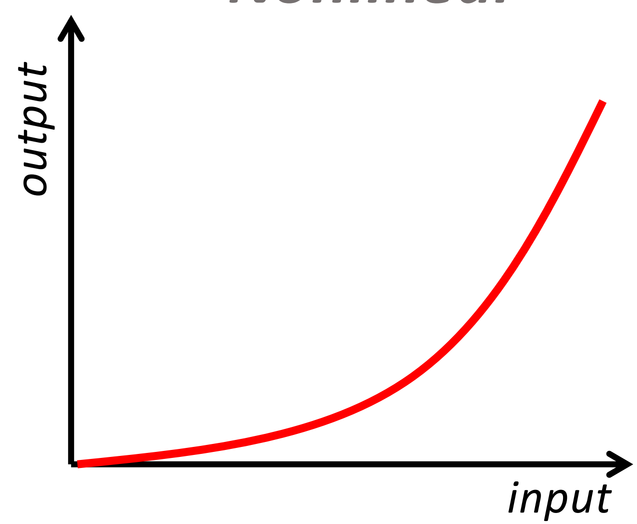
- strong X-ray fields
- X-ray signals are not correlated by linear dependence
- processes are based on *multi-photon-in multi-photon-out* interactions

Nonlinear system is a system in which the change of the output is not proportional to the change of the input
The **nonlinearities** in the meaning of **photon - matter** interaction, may be described as a change in optical property of the material induced by the high intensity of the beam.

Linear



Nonlinear



Nonlinear interactions

Saturable absorption.

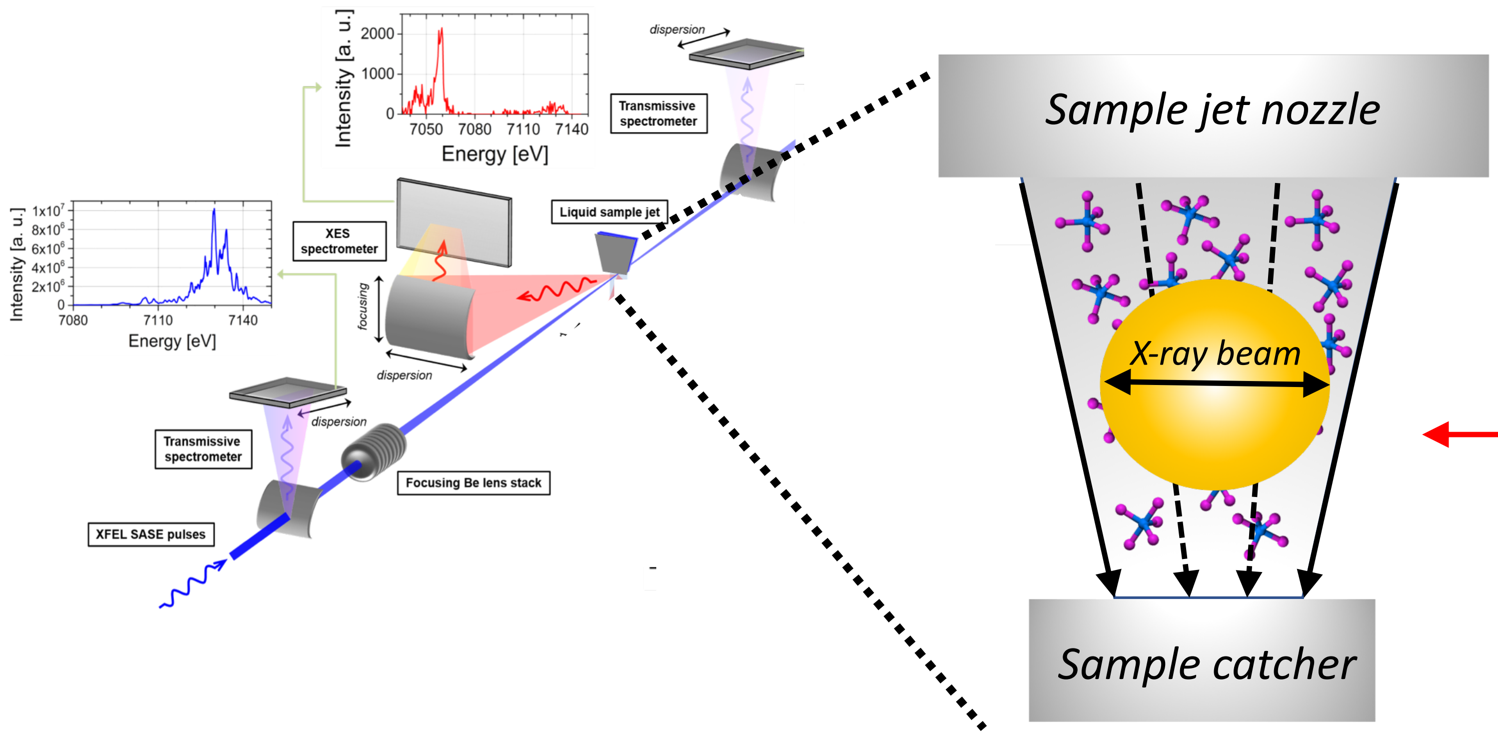
In the linear regime, the absorption of electromagnetic radiation is well described by the linear absorption coefficient $\boldsymbol{\mu}$.

However, with the growth of the intensity, the material properties changes, and the linear absorption coefficient u value start to depend on beam intensity :

$$\mu = \frac{\mu_0}{1 + \frac{I}{I_s}}$$

where the Is is the intensity at which the system reaches the saturation of absorption.

Experimental setup at XPP station of LCLS

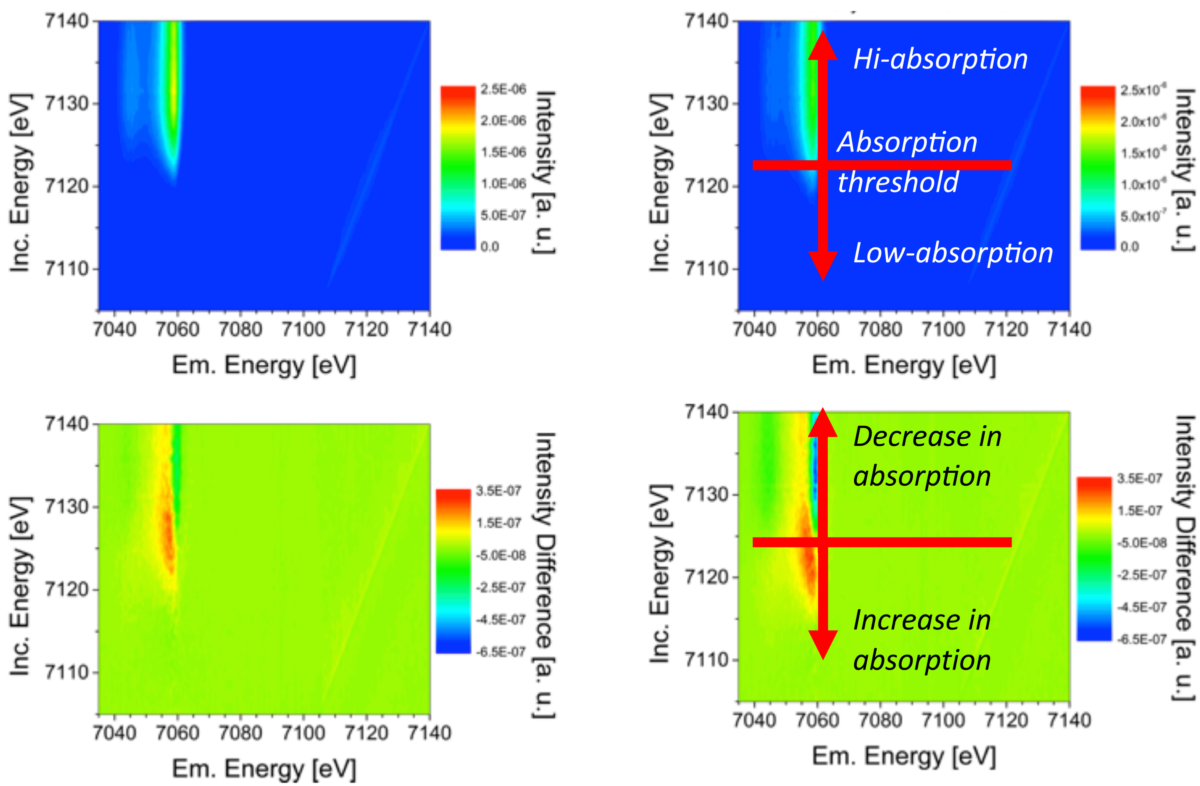


x200 increase in photon density!!

<u>X-ray beam properties</u>:

- self-amplified spontaneous emission (SASE)
- repetition rate: 120 Hz
- mean photon energy: 7100 eV (around Fe K-edge)
- pulse duration: 5fs & 30 fs
- number of photons per pulse: 10^{12}
- beam spot area on the target changed from 6 to 241 μm^2 with Be lenses

Nonlinear atomic response around ionization threshold

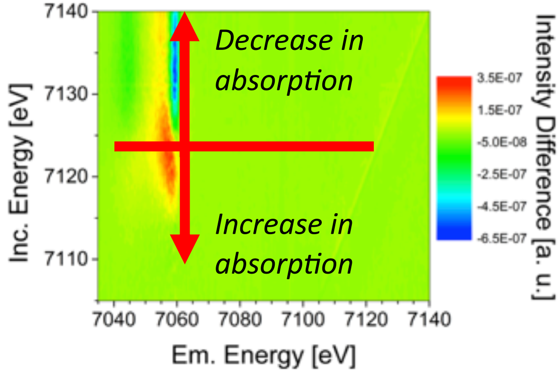


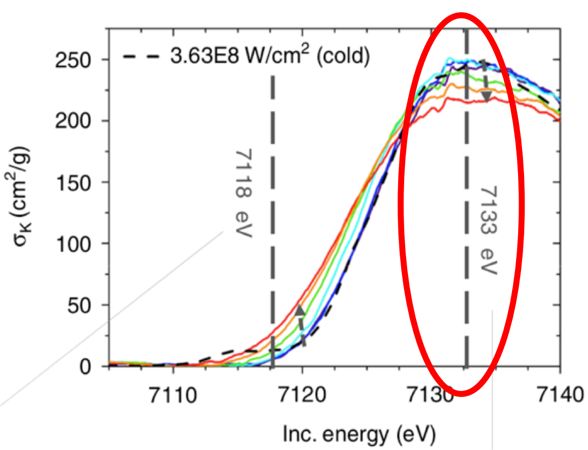
increase in X-ray density

Properties of material become inversed !!

Nonlinear atomic response around ionization threshold

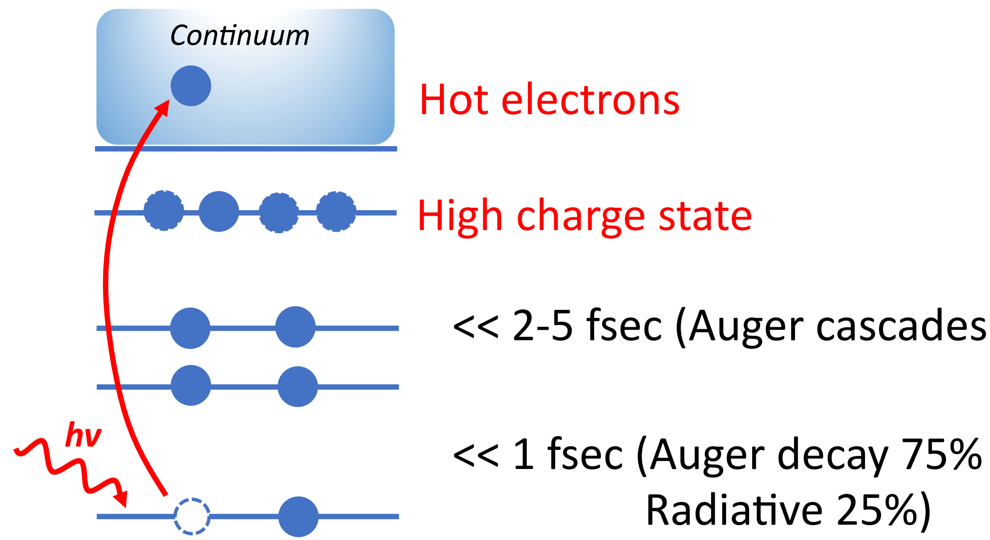
Experimental





Decrease in absorption for energies above threshold suggest saturable absorption effect

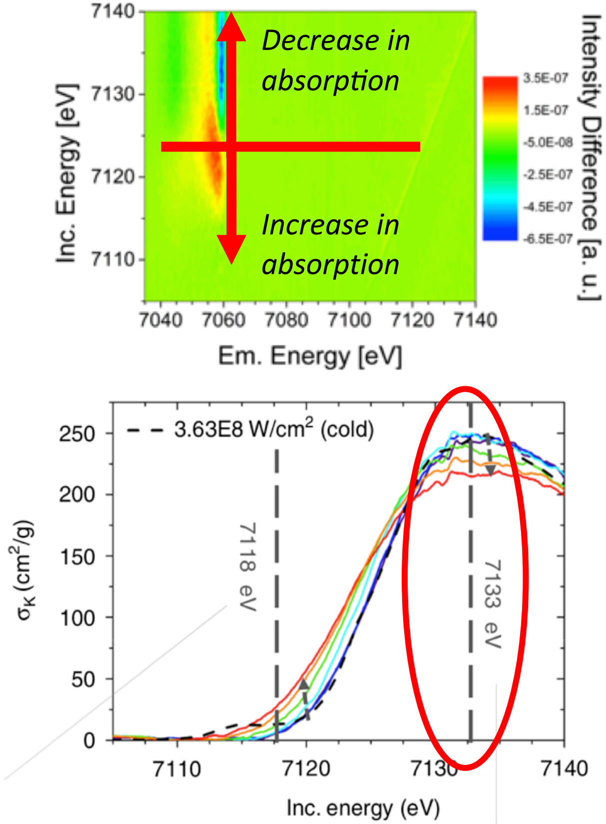
Ionization and decay processes

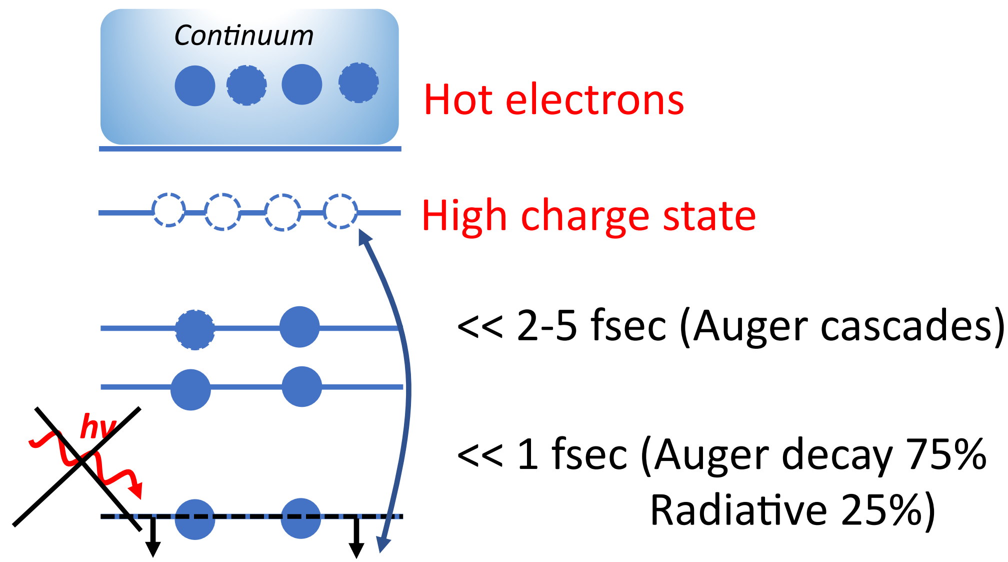


Pulse length 30fsec

Nonlinear atomic response around ionization threshold

Experimental





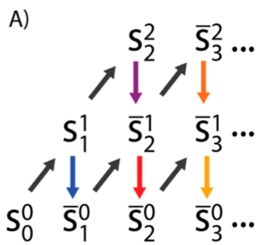
Change in charge state ➡️ change in screening ➡️ change in binding

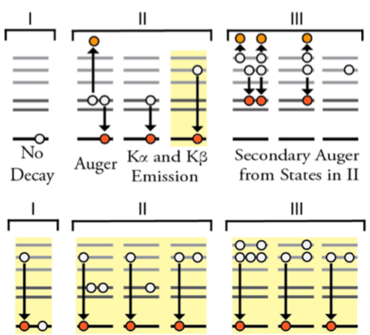
Pulse length 30fsec

Decrease in absorption for energies above threshold suggest saturable absorption effect

Nonlinear RXES around ionization threshold – saturable absorption

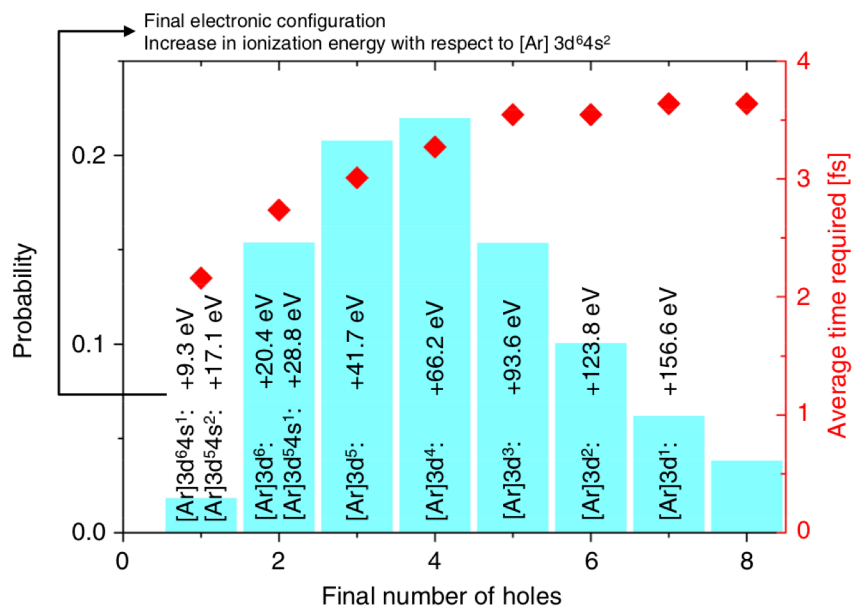
Multiple decay paths after single photo-ionization





S. C. Jensen et al, J. Phys. Chem. Lett. 2019, 10, 441–446.

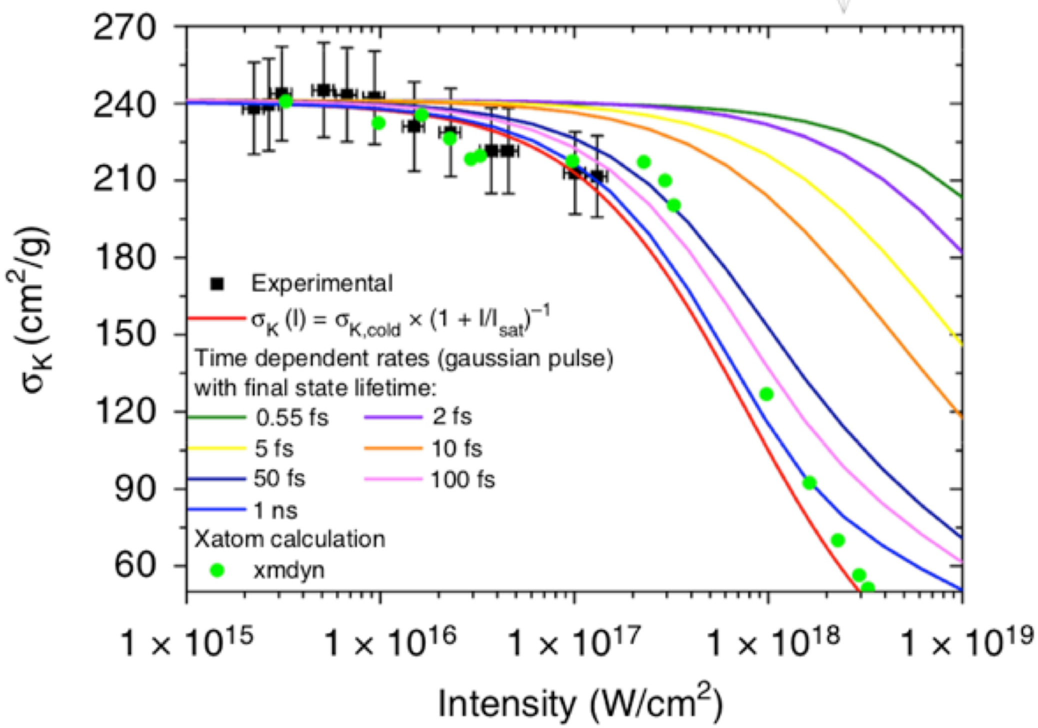
- 1s core–hole ionization is followed by multiple decay paths and the core-hole is transferred to higher atomic orbitals
- Domination of Auger decay mechanisms leads to multiplication of number of holes at higher states
- Inner-core hole transfer is very fast (sub fsec)

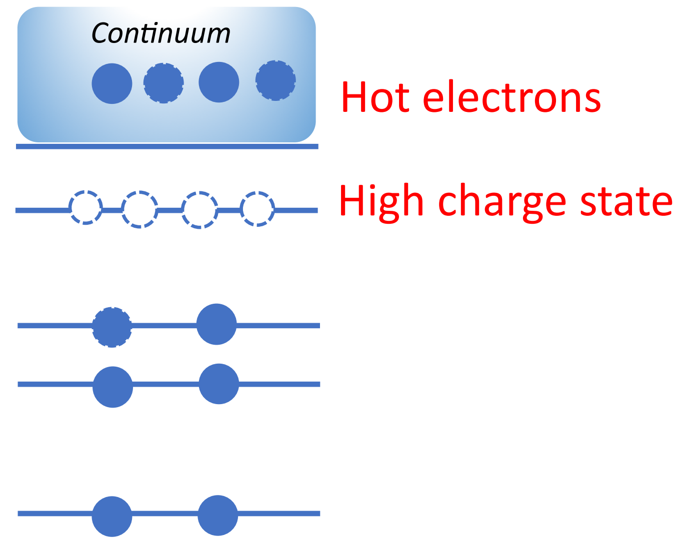


Nature Communications 10, 4761 (2019) & EurekAlert! AAAS (2020).

Nonlinear RXES around ionization threshold – saturable absorption

Rate equation calculations + xmdyn code





Nature Communications 10, 4761 (2019) & EurekAlert! AAAS (2020).

Summary:

✅ *deliberate use of stochastic process for X-ray spectroscopy applications*
✅ *overcoming diffraction limit -> toward attosecond spectroscopy*
✅ *strong X-ray filed induce inverted properties of matter*

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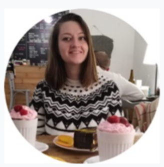

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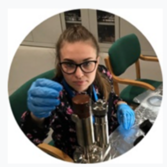

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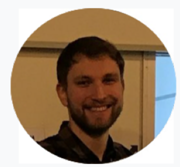

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Acknowledgments

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2017/27/B/ST2/01890
2015/19/B/ST2/00931
2015/18/E/ST3/00444

C. Milne, J. Sa, M. Nachtegaal, Y. Kayser
SwissFEL team, SuperXAS team MicroXAS team, CXI team @ LCLS, XPP team @ LCLS

PAUL SCHERRER INSTITUT PSI

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Thank you for your attention!