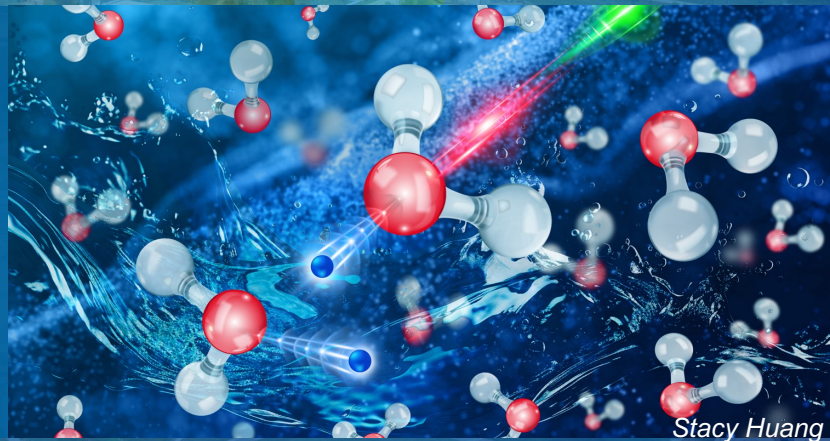
PROMISING APPLICATIONS FROM X-RAY FREE-ELECTRON LASERS



Stacy Huang

LINDA YOUNG
Argonne National Laboratory
The University of Chicago

CAMOS Meeting
Irvine, CA
10 October 2024

U.S. DEPARTMENT OF ENERGY
Argonne National Laboratory is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC.

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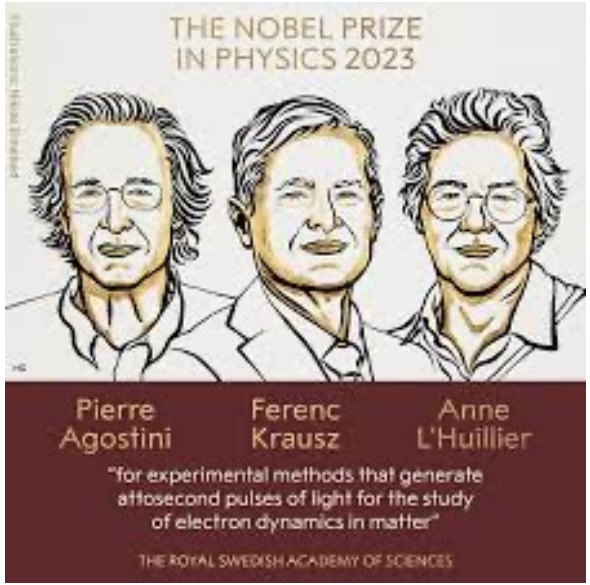
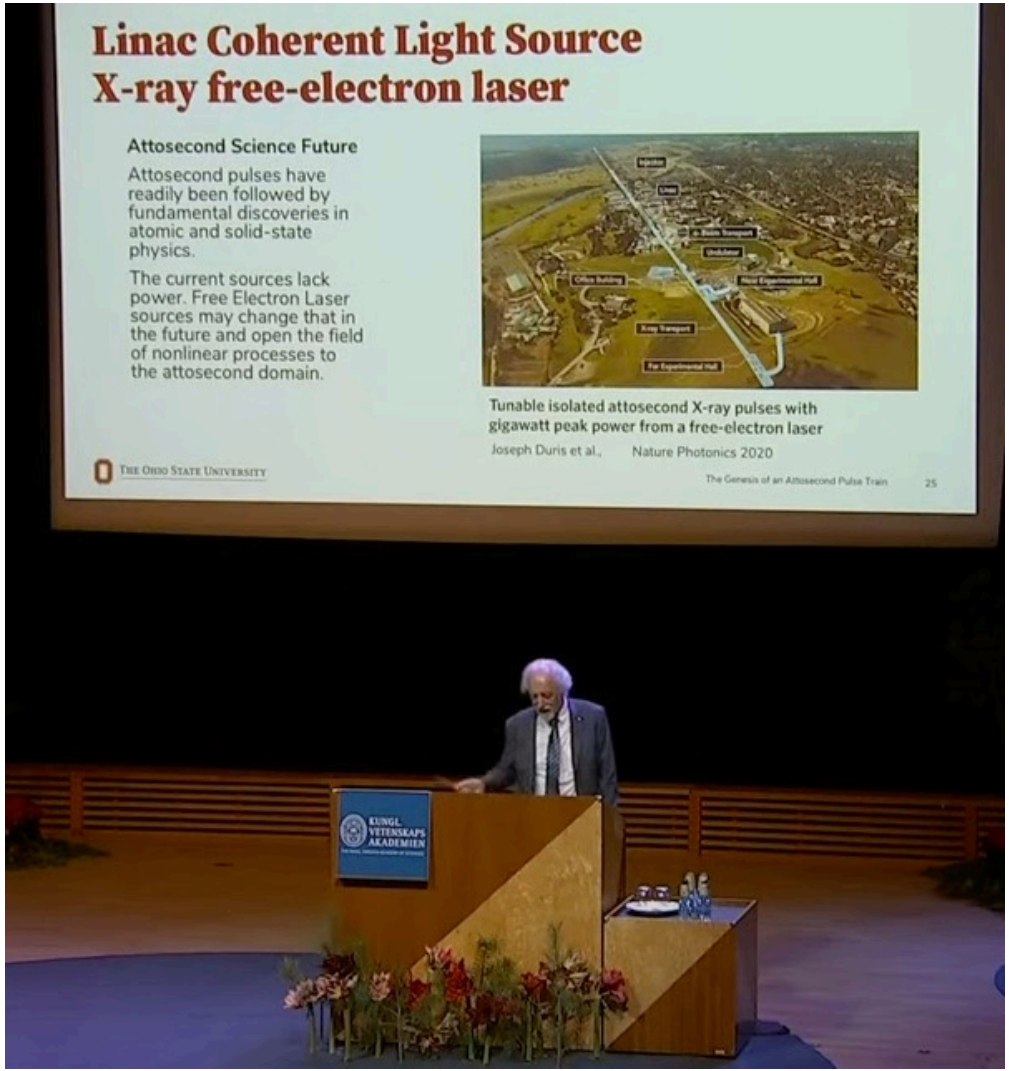


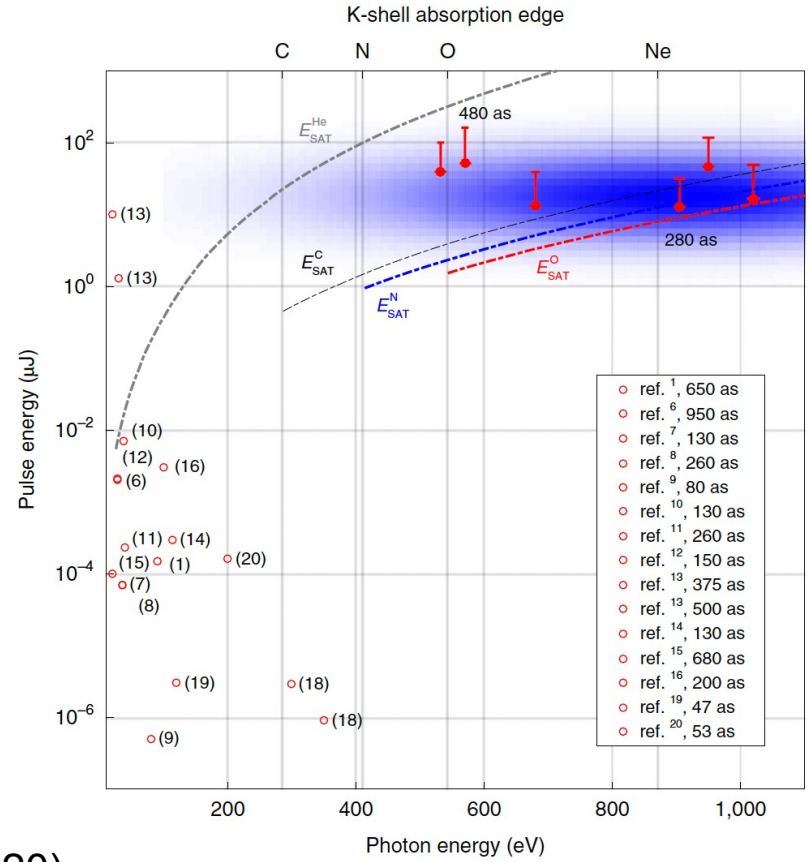
From Nobel Lectures in Physics
8 December 2023

Pierre Agostini: *just a quick look toward the future … so far attosecond pulses are created by visible or infrared lasers…but maybe the future of attoseconds is not in the our lab but with this x-ray free-electron laser…*



Attosecond x-ray pulses: XFELS v HHG

Electron dynamics
+
Atomic spatial resolution



J. Duris *et al*, Nature Photonics (2020)

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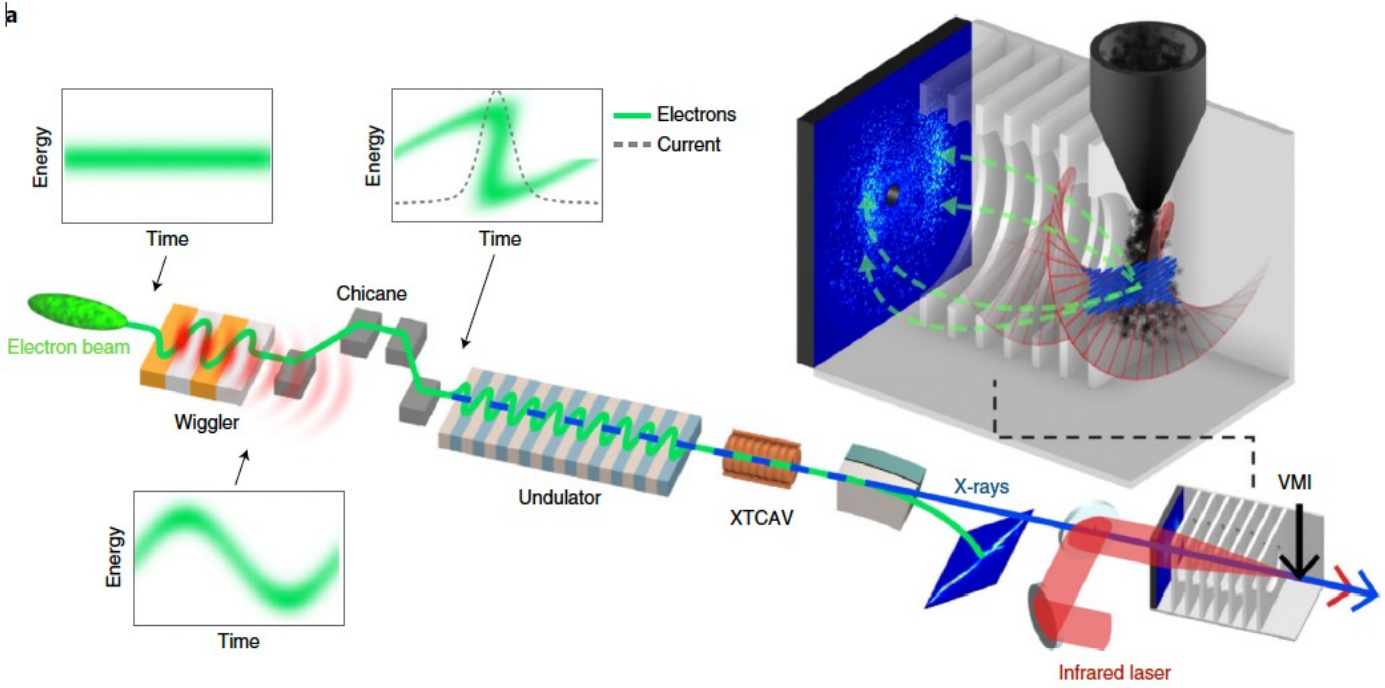
Single-spike attosecond soft x-ray pulses!

XLEAP: With measurement of pulse duration w/ c-VMI streaking

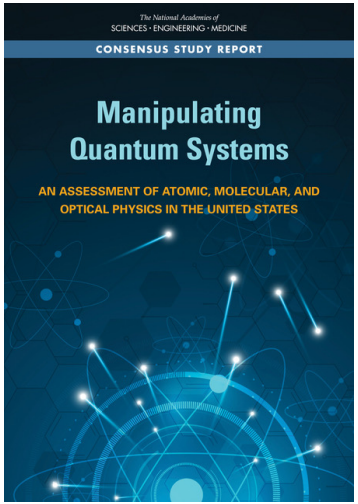


905 eV
$\Delta\tau_{FWHM}$ = 280 as

570 eV
$\Delta\tau_{FWHM}$ = 480 as

Pulse energy
~ 50 μJ

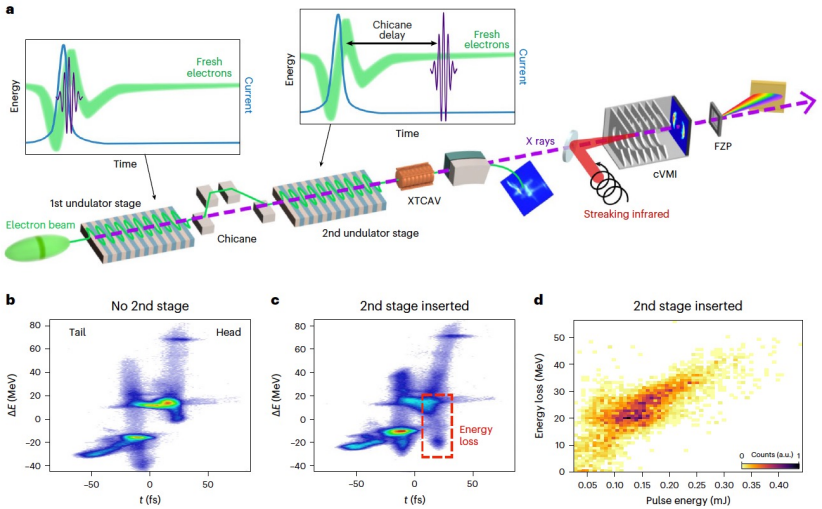
J. Duris…J. Cryan, A. Marinelli, *Nature Photonics* (2020)



AMO 2020 decadal survey, p. 158

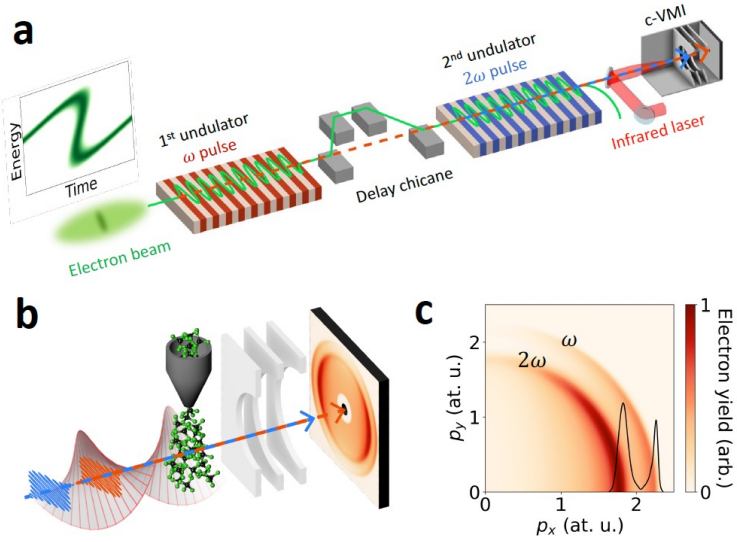
Another exciting prospects for attosecond science is the impending availability of intense attosecond pulses in the soft and hard X-ray regime from XFELs, which would enable the initiation of electron dynamics (e.g., charge migration) from inner valence or core electrons that are in general highly localized on specific atoms within a molecule. In combination with pump-and-probe capabilities, such pulses would thus allow for both spatial and temporal resolution of attosecond electron dynamics.

Terawatt-scale attosecond X-ray pulses from a cascaded superradiant free-electron laser



P. Franz … J. Cryan, A. Marinelli,
Nature Photonics (2024) May

Synchronized attosecond x-ray pulse-pair generation



Z. Guo … J. Cryan, A. Marinelli,
Nature Photonics (2024) May

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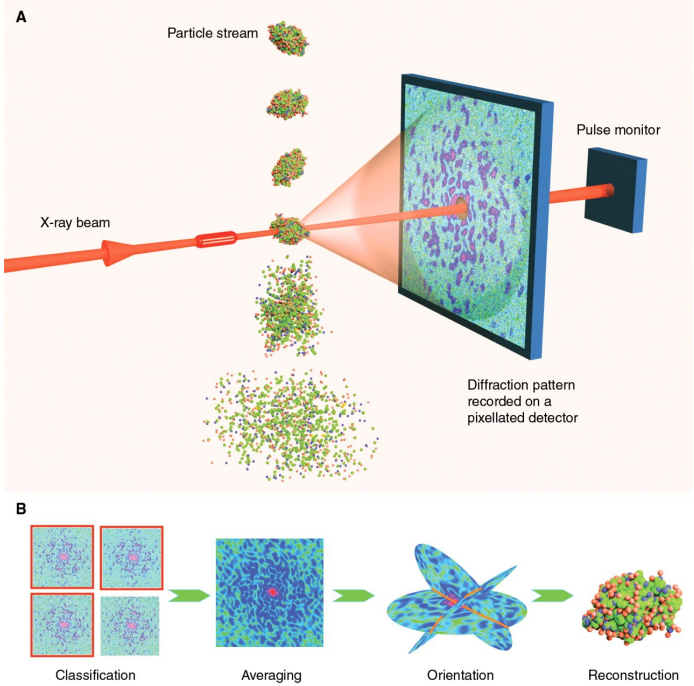
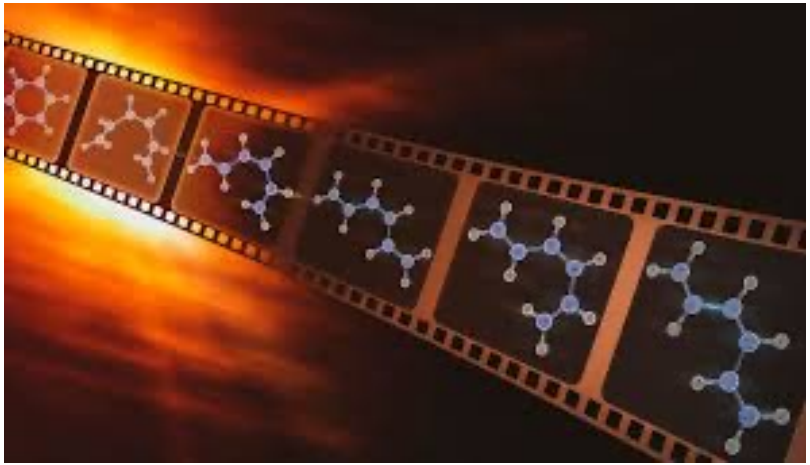
Original applications of XFELs – enhanced w/attosecond pulses

Single Particle Imaging



K. J. Gaffney & H.N. Chapman, *Science (2007)*
Neutze… Hajdu, *Nature* (2000)
Chapman...Spence, *Nature* (2011)
Seibert… Hajdu, *Nature* (2011)

Molecular Movies

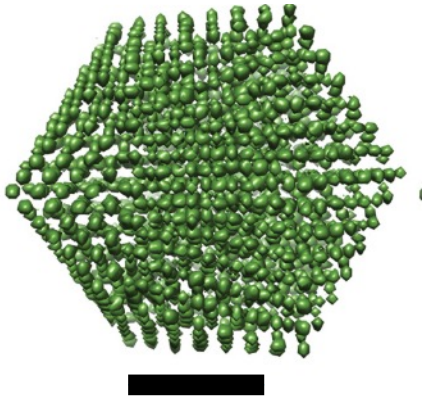


Minitti et al, *PRL* **114**, 255501 (2015) – XRD
Attar et al, *Science* **356**, 54 (2017) - XAS
Wolf et al, *Nat Chem*. **11**, 504 (2019) - UED

Spatial resolution with CDI at FELs

Theory

Ar

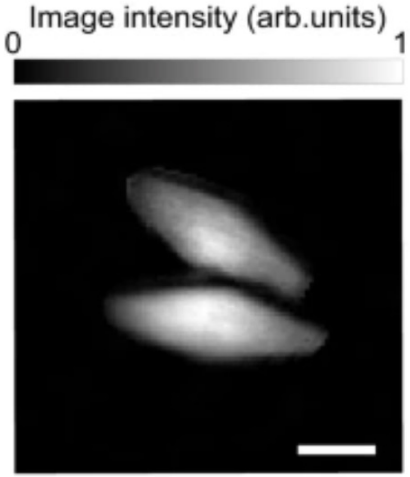


4 nm

Ho, Phay J., et al., *Physical Review A* 94.6 (2016): 063823.

Experiment

Au



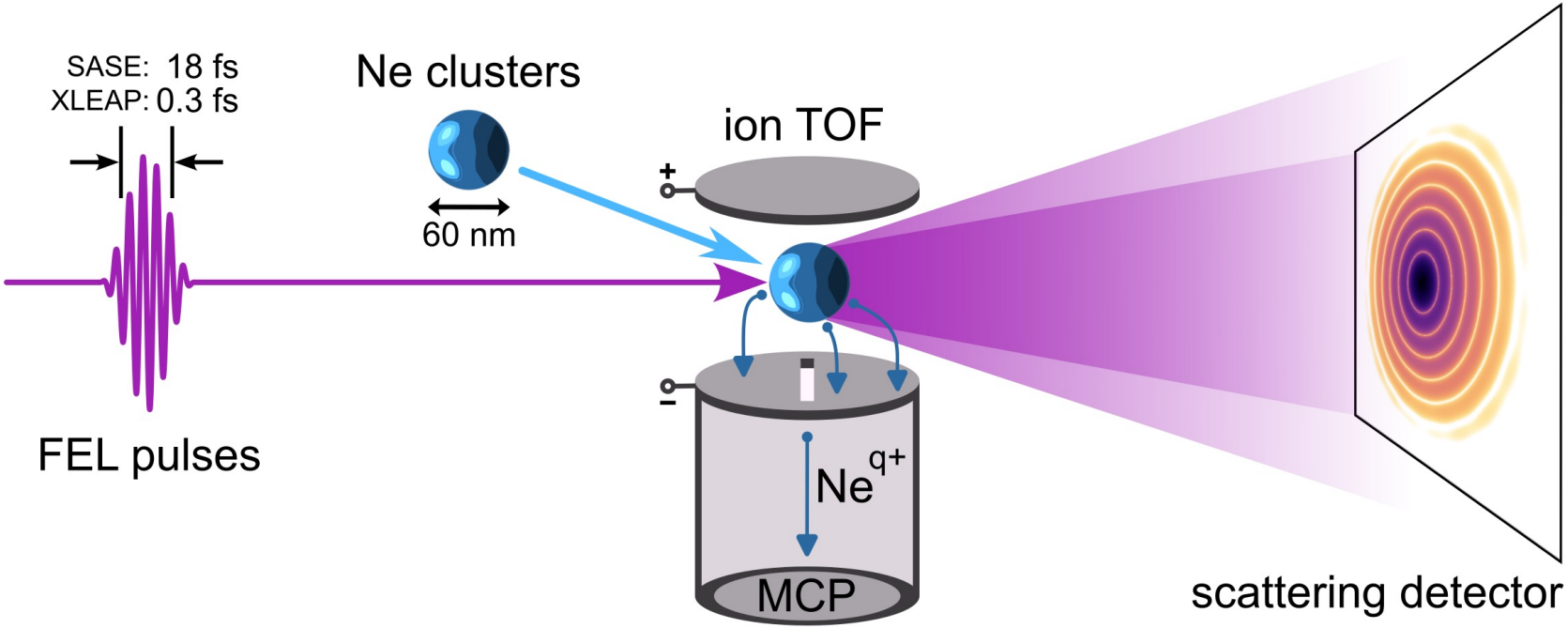
20 nm

Yumoto, Hirokatsu, et al. *Nat. Comm.* 13.1 (2022): 1-8.

Courtesy Tais Gorkhover

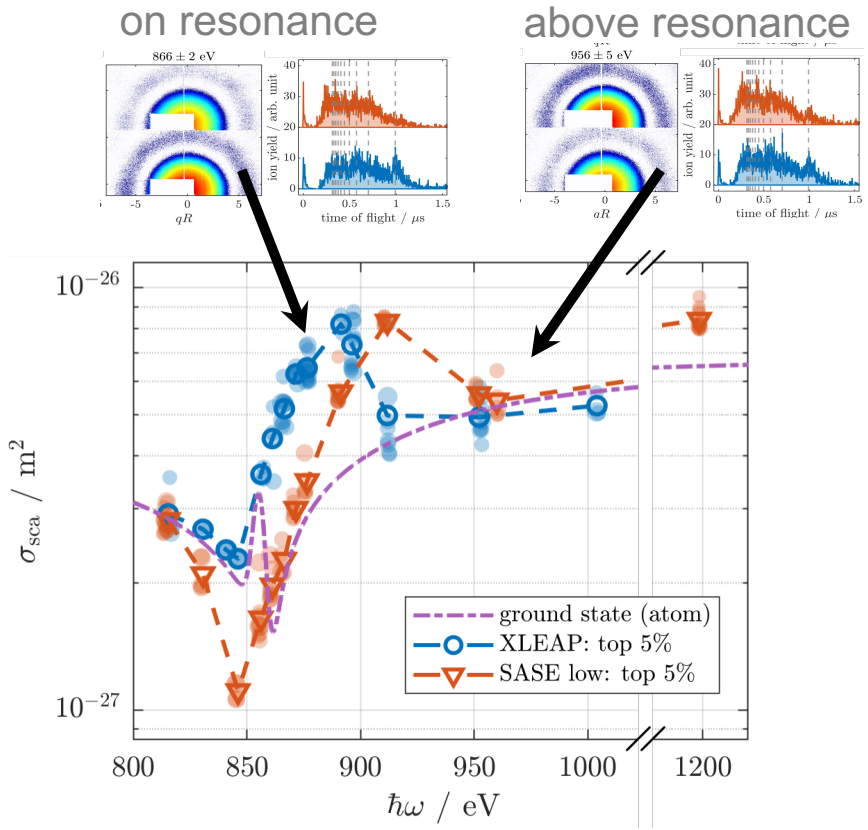
Optimizing single particle imaging

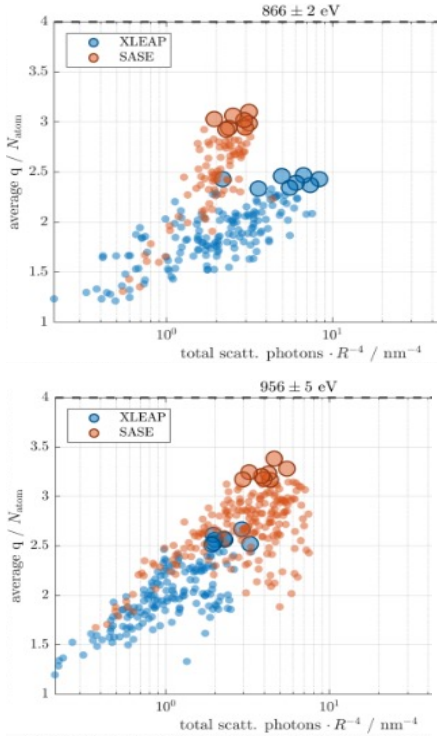
SASE (fs pulses) v XLEAP (attosecond pulses)



Courtesy Tais Gorkhover

Resonant as pulses increase scattering & decrease damage





Courtesy Tais Gorkhover

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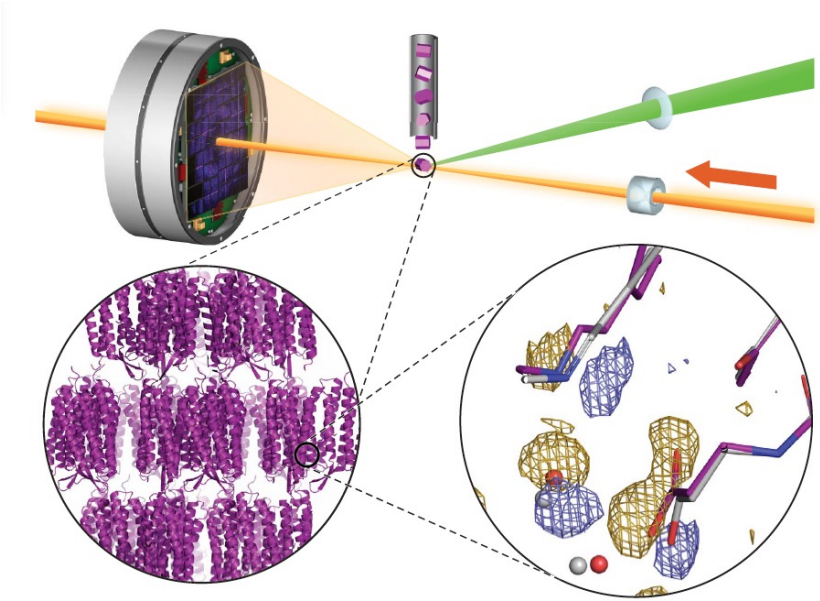
STRUCTURAL BIOLOGY

Advances and challenges in time-resolved macromolecular crystallography

Gisela Brändén and Richard Neutze*



Diffract before destroy – SFX
+ optical pump laser

~770 SFX deposits in PDB
~200,000 total deposits

Photosystem I, Photosystem II, Photoactive yellow protein, human rhodopsin, bacteriorhodopsin, light-activated ion channels, fluorescent proteins, myoglobin-CO, cytochrome c oxidase-CO

Science ***373***, *980 (2021)*

Science 1 Dec (2023)

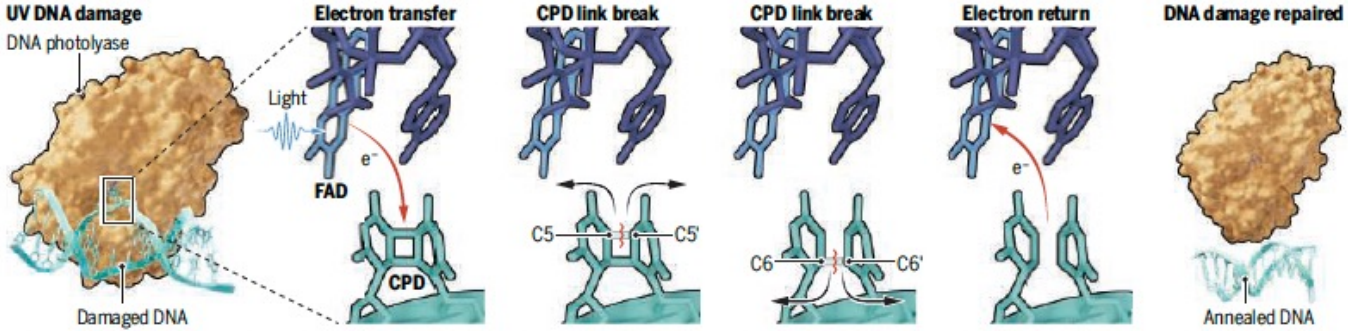
MOLECULAR BIOLOGY

Filming DNA repair at the atomic level

Dissection of multistep catalysis by a photoenzyme could inspire green chemistry applications

Light-induced DNA repair

Cyclobutane pyrimidine dimer (CPD) DNA photolyase repairs ultraviolet (UV) light–induced CPDs that arise between two adjacent pyrimidine bases. Four temporally distinct chemical steps occur that involve the flavin-adenine dinucleotide (FAD) coenzyme of the photolyase and the bases to break the two bonds that form the CPD.



By **Marten H. Vos**

ENZYMOLOGY

Visualizing the DNA repair process by a photolyase at atomic resolution

Maestre-Reyna *et al.*, *Science* **382**, eadd7795 (2023) 1 December 2023

ENZYMOLOGY

Time-resolved crystallography captures light-driven DNA repair

Christou *et al.*, *Science* **382**, 1015–1020 (2023) 1 December 2023

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Article

Influence of pump laser fluence on ultrafast myoglobin structural dynamics

https://doi.org/10.1038/s41586-024-07032-9

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Check for updates

Thomas R. M. Barends[1]✉, Alexander Gorel[1,10], Swarnendu Bhattacharyya[2,10], Giorgio Schirò[3], Camila Bacellar[4], Claudio Cirelli[4], Jacques-Philippe Colletier[3], Lutz Foucar[1], Marie Luise Grünbein[1], Elisabeth Hartmann[1], Mario Hilpert[1], James M. Holton[5], Philip J. M. Johnson[4], Marco Kloos[6], Gregor Knopp[4], Bogdan Marekha[7], Karol Nass[4], Gabriela Nass Kovacs[1], Dmitry Ozerov[4], Miriam Stricker[8], Martin Weik[3], R. Bruce Doak[1], Robert L. Shoeman[1], Christopher J. Milne[4], Miquel Huix-Rotllant[2]✉, Marco Cammarata[9] & Ilme Schlichting[1]✉

Nature 626, 905 (2024)

NEWS & VIEWS FORUM | 14 February 2024

Energetic laser pulses alter outcomes of X-ray studies of proteins

Cutting-edge X-ray sources have enabled the structural dynamics of proteins to be tracked during biochemical processes, but the findings have been questioned. Two experts discuss the implications of a study that digs into this issue.

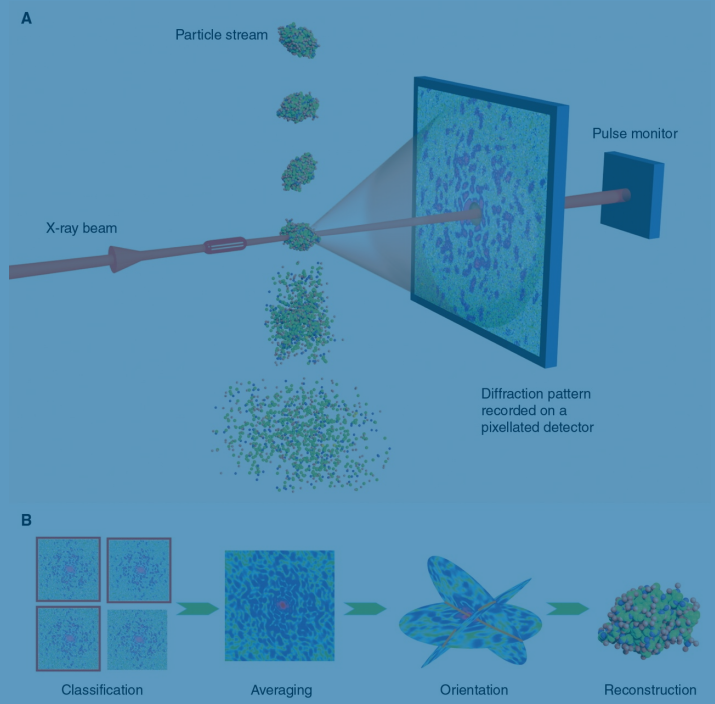
By Richard Neutze ✉ & R. J. Dwayne Miller ✉

Nature 626, 720 (2024)

Grand challenge: structure & fcn at atomic resolution at physiological conditions

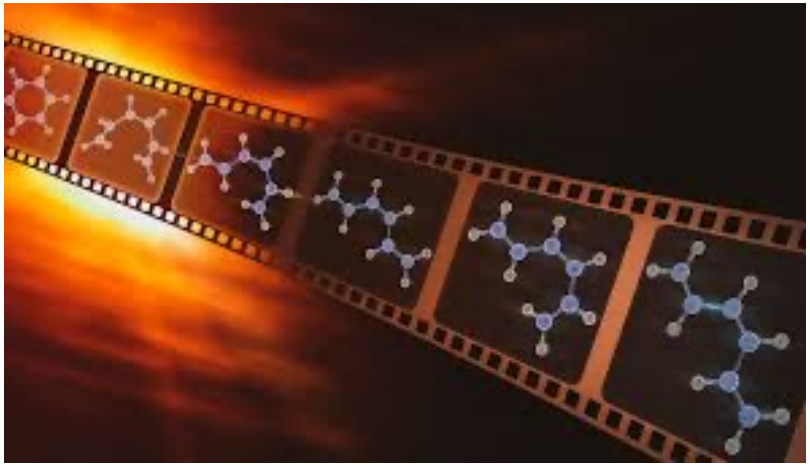
Original applications of XFELs – enhanced w/attosecond pulses

Single Particle Imaging



K. J. Gaffney & H.N. Chapman, *Science (2007)*

Molecular Movies



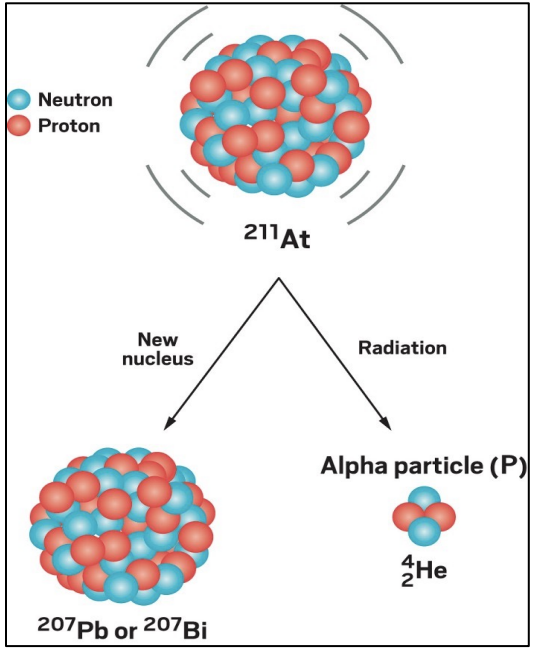
Minitti et al, *PRL* **114**, 255501 (2015) – XRD
Attar et al, *Science* **356**, 54 (2017) - XAS
Wolf et al, *Nat Chem.***11**, 504 (2019) -UED

Radiolysis: radiation effects in liquid water

Space Travel

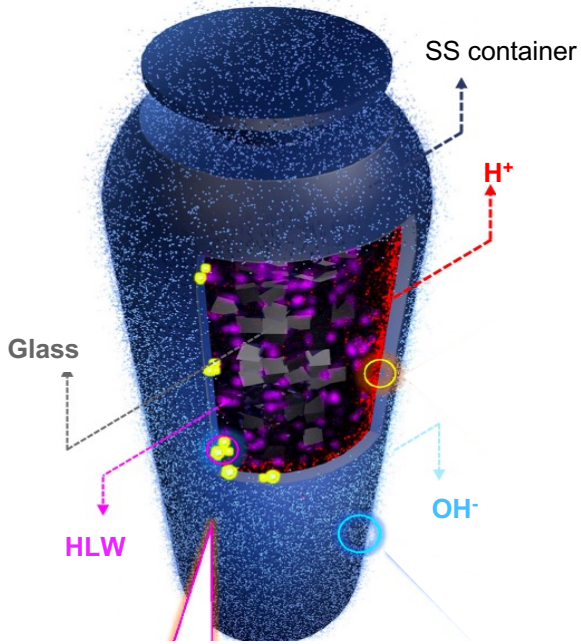
Science – Apr 12, 2019

Cancer Therapeutics



C&E News – 2020

Corrosion in nuclear power plants & repositories

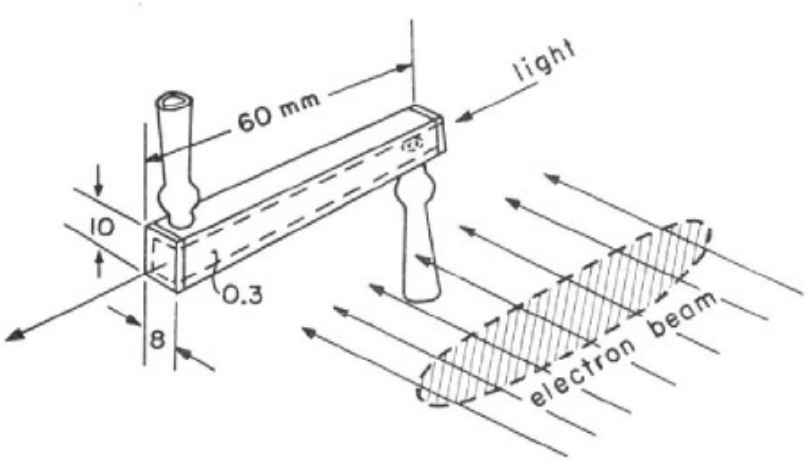


Nat. Mater. 19, 310 (2020)
C&E News Mar & Sep (2020)

Standard method for e- beam radiolysis

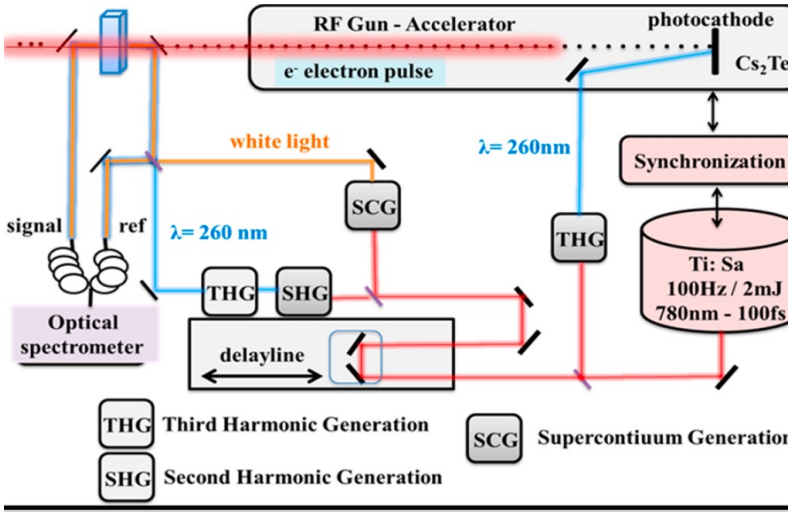
Optical detection of radiolysis products

Hart & Boag JACS 1962



e^-: 1.8 MeV, 2 μs, 0.5 A peak
probe: continuum 4 μs

Picosecond pulse radiolysis: ELYSE



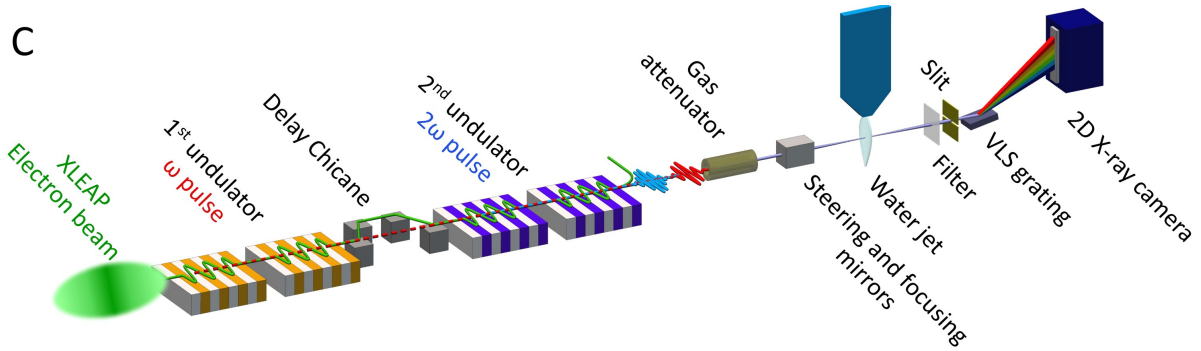
e^-: 7–8 MeV, 7 ps, 4 nC
Broadband fs probe: 380-1500 nm
Absorption cell ~ 5 mm

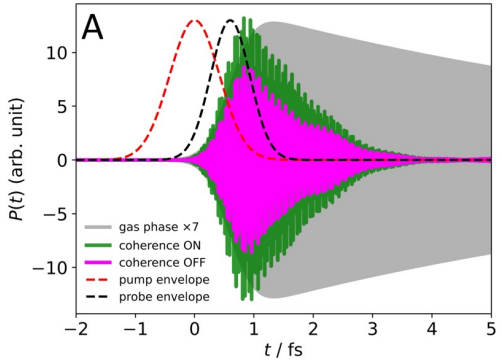
All x-ray attosecond pump/probe expts

A new tool to understand radiolysis —› mechanistic origin of reactive species

AX-ATAS: All X-ray Attosecond Transient Absorption

Polzn response – sub fs





AX-ATAS spectral snapshots freeze all nuclear motion

Shuai Li *et al. Science* **383**, 1118 (2024)

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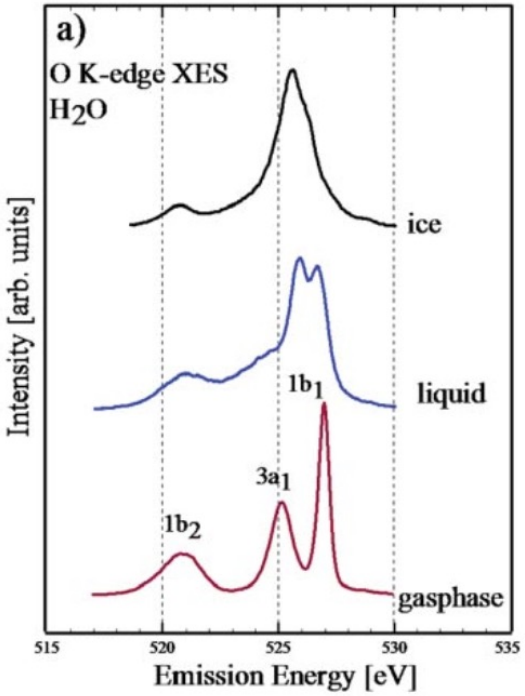
Equilibrium properties from our attosecond pump-probe experiments

resolving a debate in liquid water structure

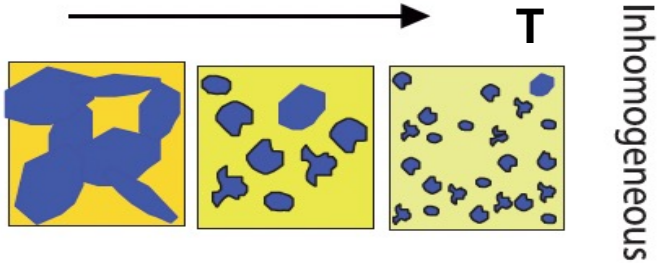
See Physics Today, April 2024 issue, Johanna Miller
Physics, April 2024, Charles Day

X-ray emission spectroscopy has been used to infer the structure of liquid water



A. Nilsson *et al.*, N Cimento (2016)

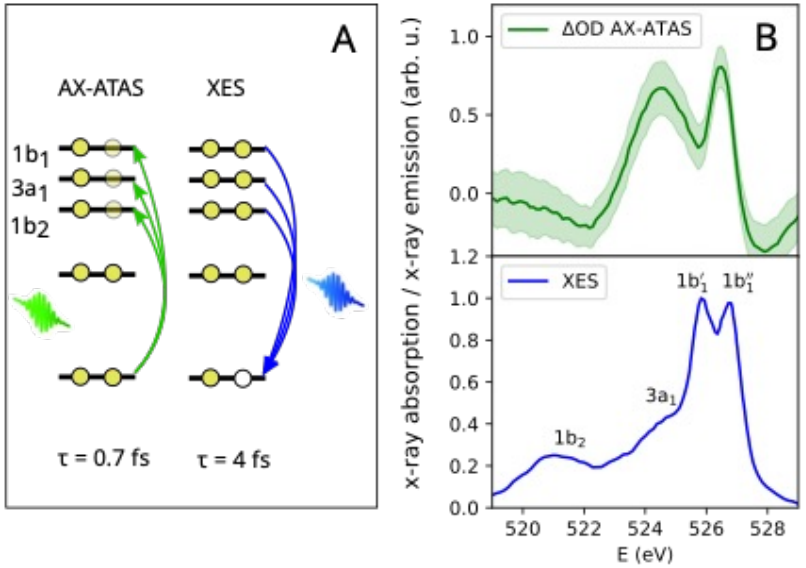
"*High resolution X-ray emission spectroscopy of liquid water: The observation of two structural motifs*"
Tokushima … Chem Phys Lett (2008)



"*Isotope and Temperature Effects in Liquid Water Probed by X-Ray Absorption and Resonant X-Ray Emission Spectroscopy*"
Fuchs …Phys Rev Lett (2008)

Ultrafast dissociation in the core-excited state

AX-ATAS compared to X-ray Emission Spectroscopy



AX-ATAS @ 0.7 fs shows no evidence for two structural motifs in ambient liquid water

Shuai Li *et al. Science* **383**, 1118 (2024)

OPPORTUNITIES w/XFELs

Tunable, synchronized attosecond pulse pairs +

- **High repetition rate —› big data —› rare events**

  ExaFEL: extreme-scale real-time data processing for X-ray free electron laser science
  J. P. Blaschke *et al.*, *Frontiers in High Performance Computing Oct 2024*

- **Extreme focusing —› nonlinear phenomena**

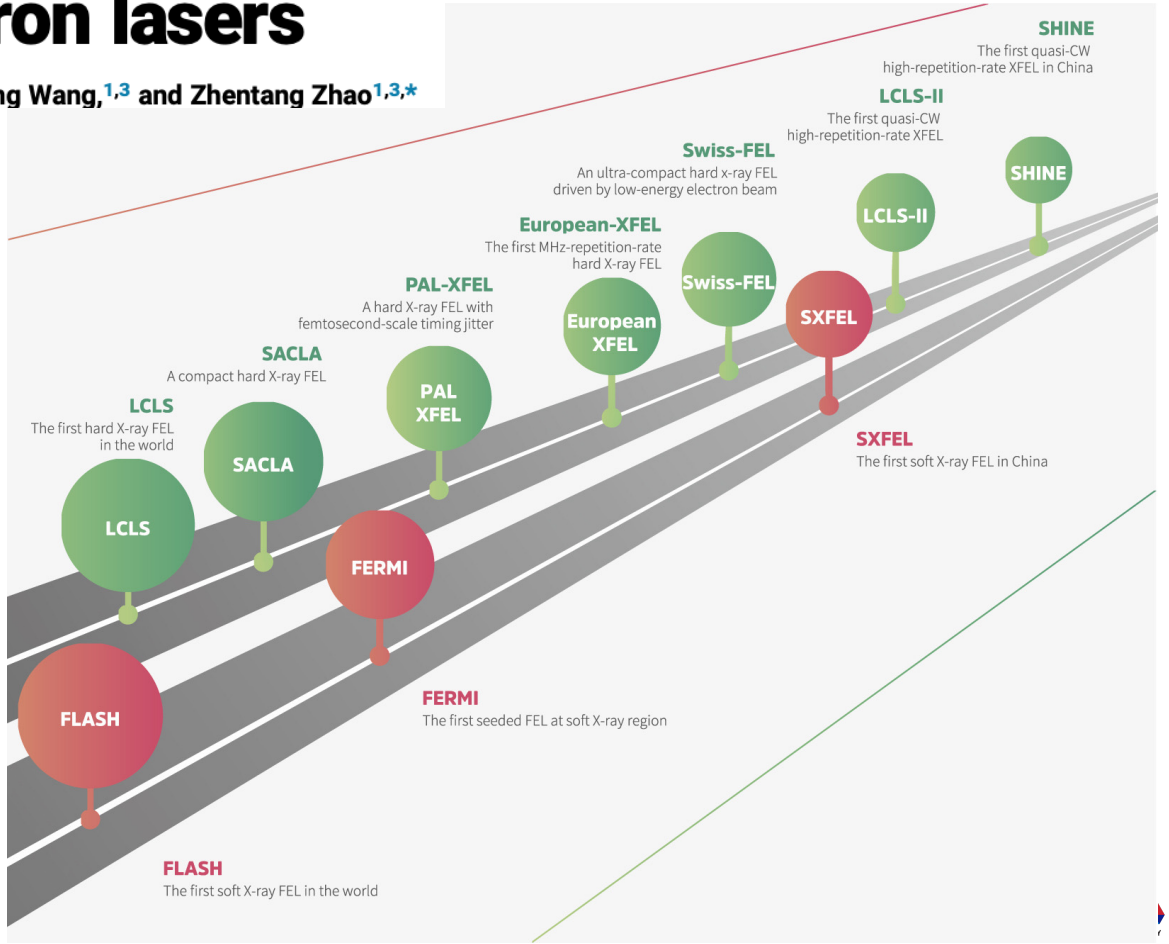
  Extreme focusing of hard X-ray free-electron laser pulses enables 7 nm focus width and 10^{22} W cm^{-2} intensity
  J. Yamada *et al.*, *Nature Photonics* **18**, 685–690 (2024)

- **More accessibility: biology, chemistry, materials, condensed matter physics, warm dense matter**

Features and futures of X-ray free-electron lasers

Nanshun Huang,[1,2] Haixiao Deng,[1,3,*] Bo Liu,[1,3] Dong Wang,[1,3] and Zhentang Zhao[1,3,*]

The Innovation 2(2), 100097 (2021).



SHINE
8 GeV, SC accel, 1 MHz
3 undulators, 0.4-25 keV
10 endstations
User expts 2027

young@anl.gov