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John Adams Institute for Accelerator Science

Ultrashort pulse X-ray absorption spectroscopy using laser-plasma accelerators

Laser-Plasma Accelerators Workshop 2025

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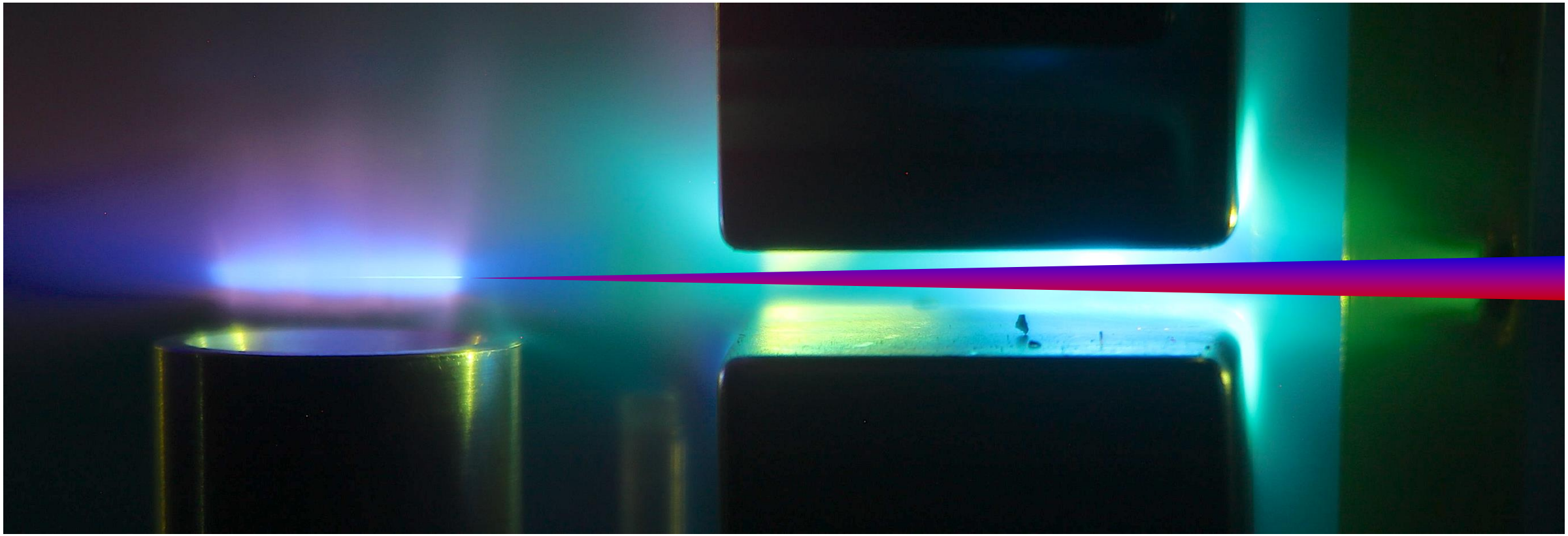
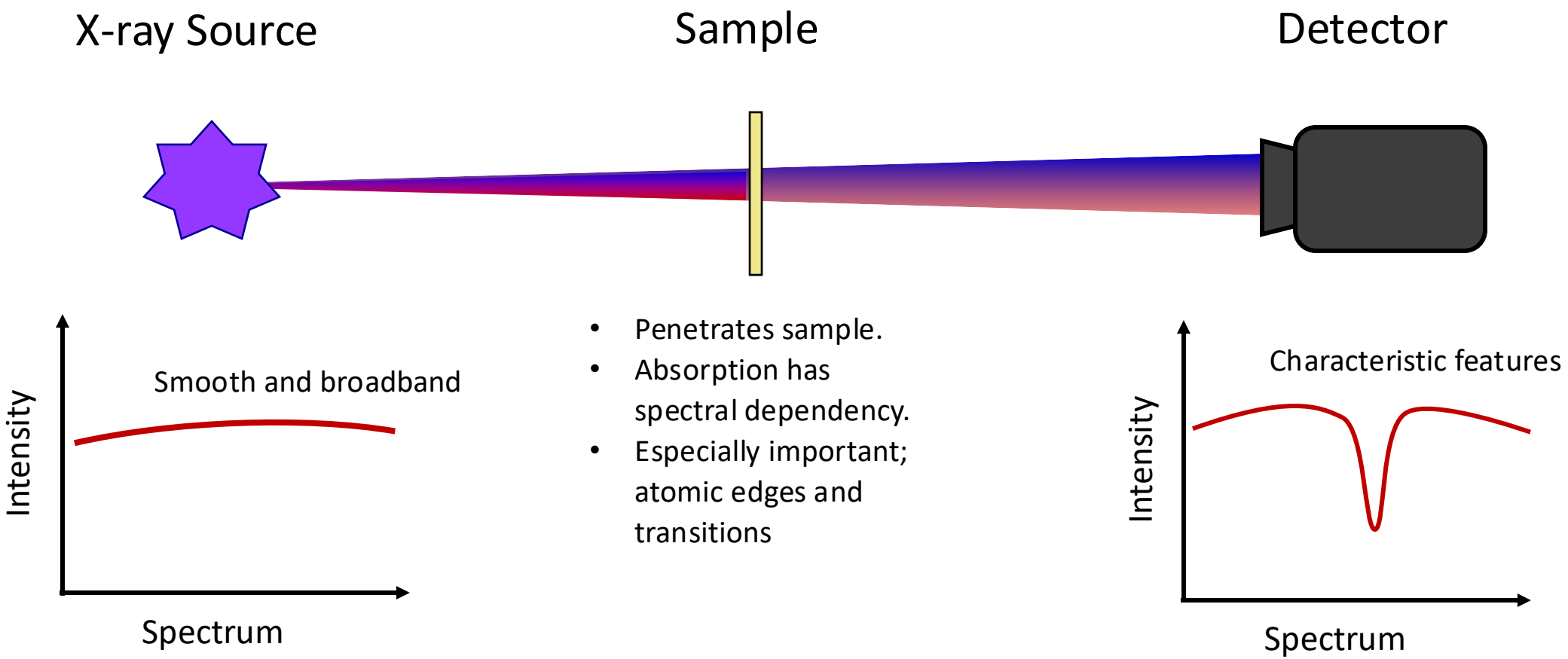
Agenda

- Conventional X-ray absorption spectroscopy
- Moving to ultrashort duration; using LWFA's
- Current state of the art
- Pump-probe studies
- Future & Summary



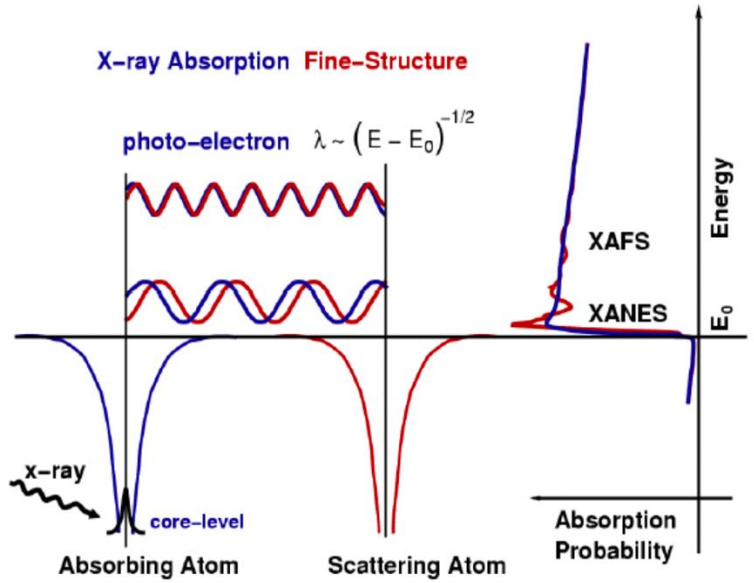
X-ray absorption Spectroscopy
What is it?

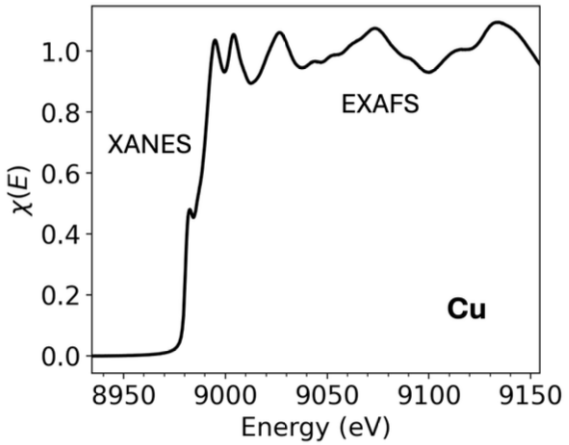


- Penetrates sample.
- Absorption has spectral dependency.
- Especially important; atomic edges and transitions

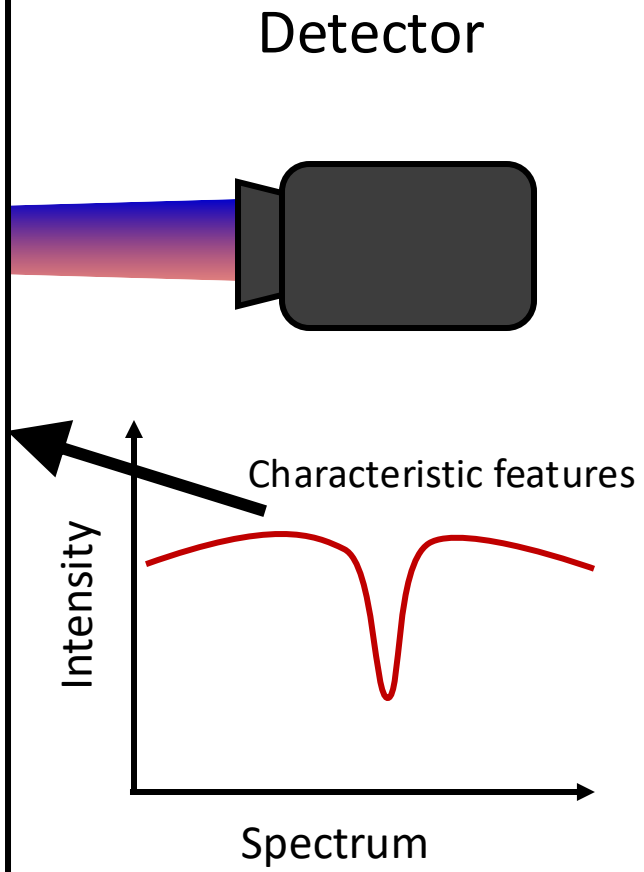
X-ray absorption Spectroscopy
What is it?

XANES & EXAFS





- Local scattering of ejected photoelectrons cause modulations in absorption close to resonant edge.
- Slope and peak positions = **electronic & atomic structure**
- Broadening of features = **electron & ion temperature**.



A Practical Introduction to Multiple Scattering Theory, Bruce Ravel, 2005

X-ray absorption Spectroscopy

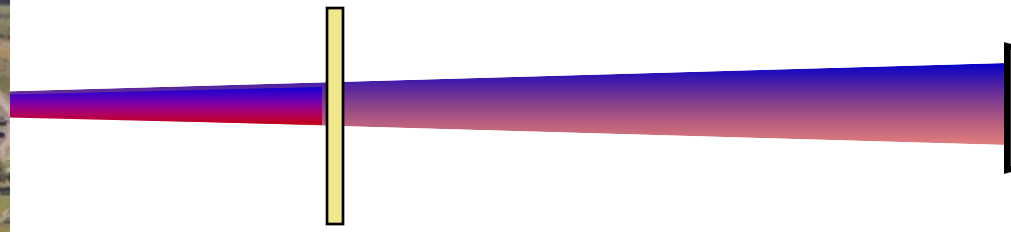
Synchrotrons are ideal for “ambient” measurements

Synchrotron

- **Broadband**, 10’s keV X-rays
- MHz rep. rate
- 10’s – 100’s ps duration

Spectrum

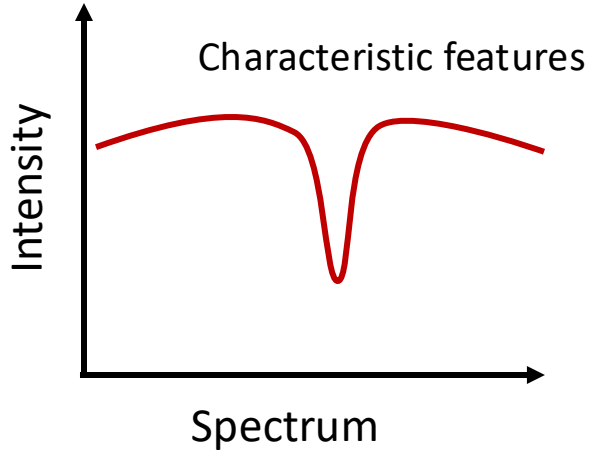
Sample



Success over 4 decades;

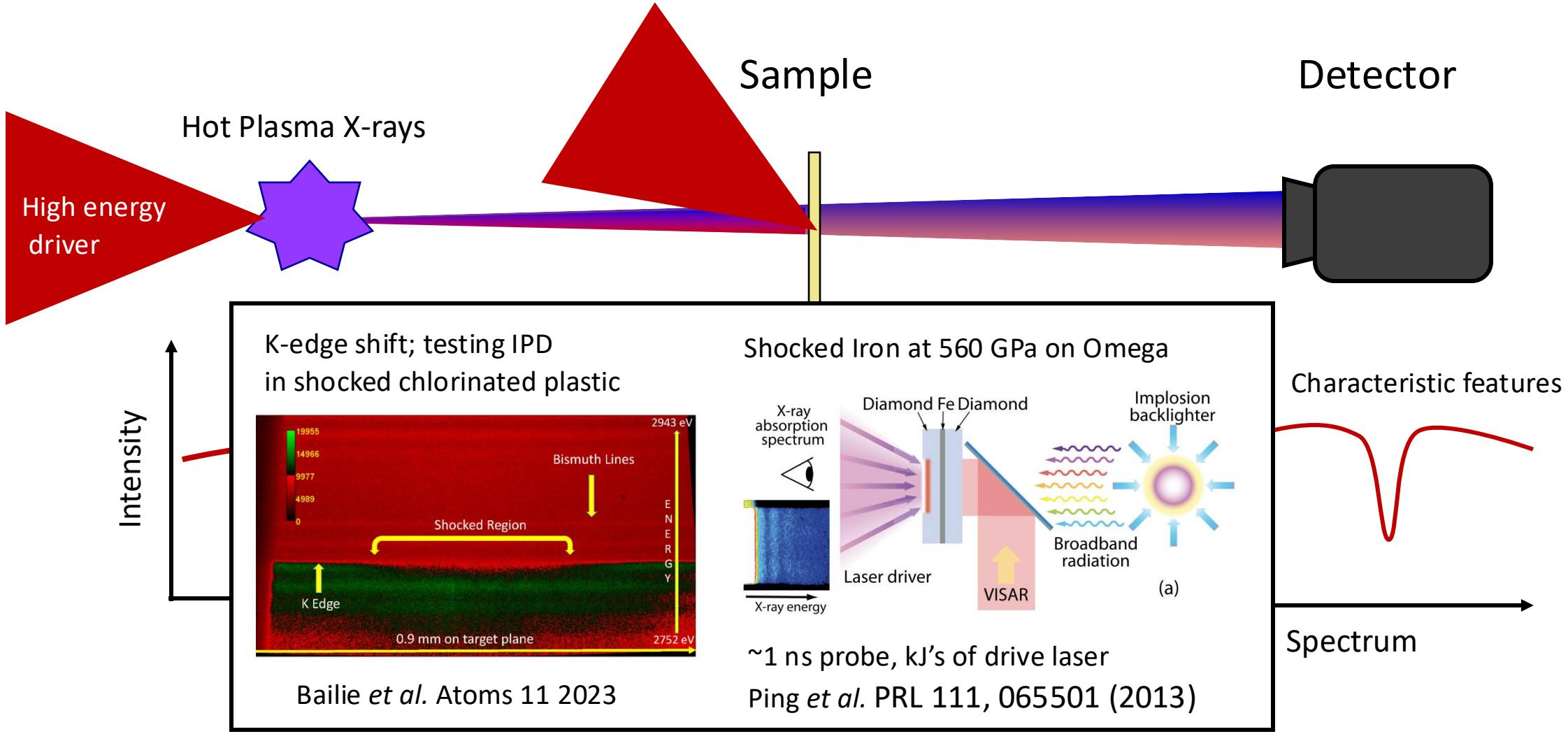
- Material science
- Nanomaterials
- Semiconductors
- Biology
- Protein structure
- Chemistry & Catalysis
- Energy Storage
- Environmental
- Etc….

Detector



X-ray absorption Spectroscopy

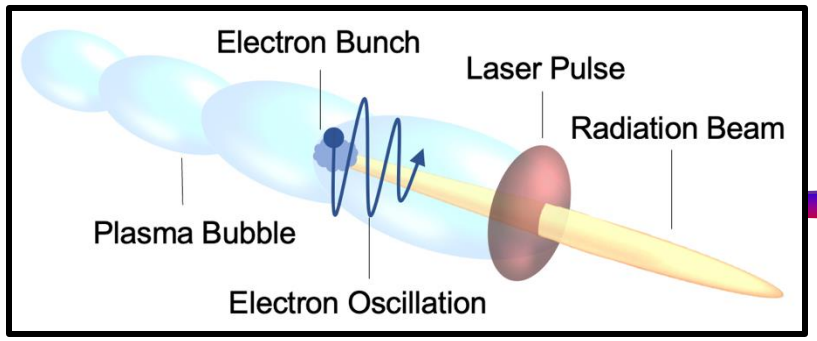
Multi-picosecond laser-plasma probes



Ultrafast X-ray absorption Spectroscopy

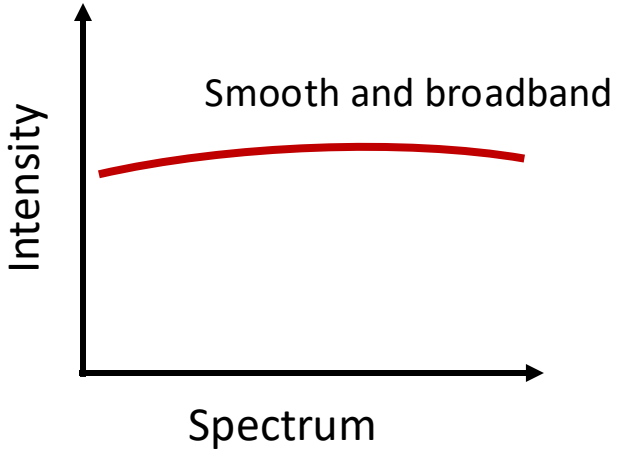
Moving to femtosecond resolution

LWFA driven

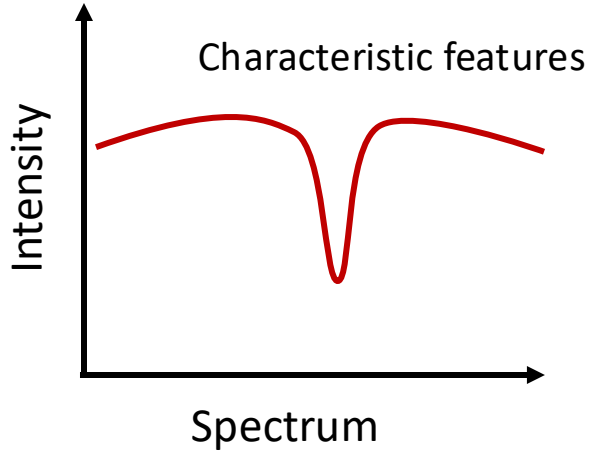


Sample

Detector



- Bright keV X-rays
- Broadband
- **10's femtosecond scale**
- ≈ mrad divergence
- ≈ micron source size
- Synchronised to pump



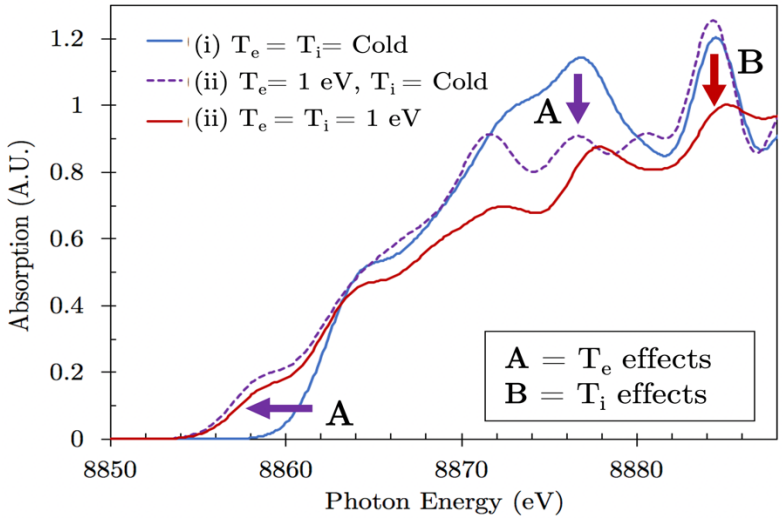
Ultrafast X-ray absorption Spectroscopy

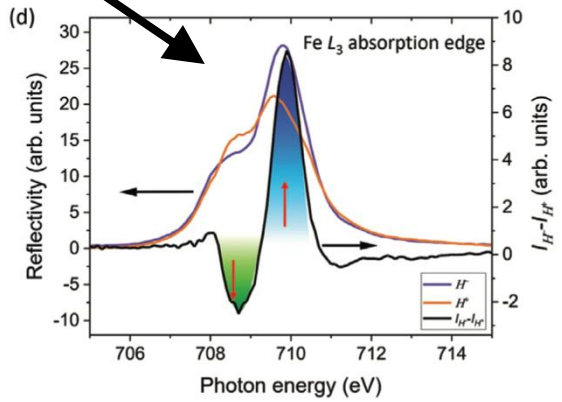
Moving to femtosecond resolution – why?

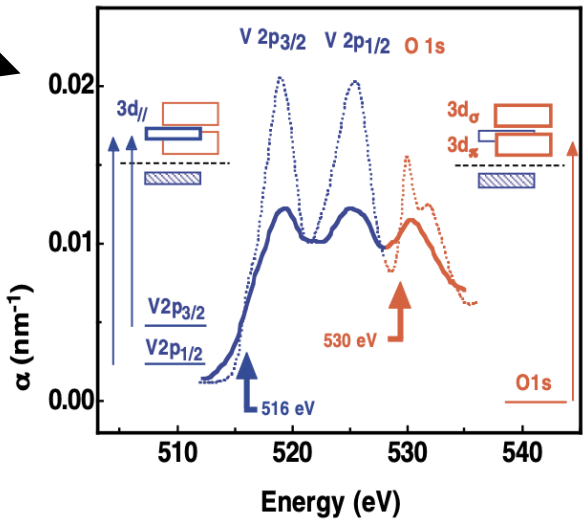
Allows interrogation of sub-picosecond processes, especially electron dynamics

Examples of interest:

- Electron-ion equilibration in plasma
- Non-thermal melting / Bond hardening / Electron-phonon coupling[1,2,3]
- Insulator-metal transitions[4]; e.g. <100 fs in VO_2
- Spintronics/antiferromagnets for electronics[5]
- Optical-magnetic switches[6]; E.g. YIG:Co
- … Etc.







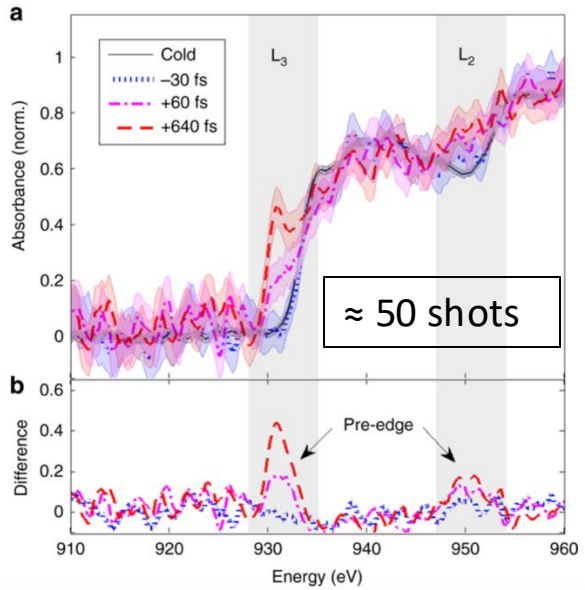
[1]Rousse et al. Nature 410, 65 (2001)
[2]Ernstorfer et al. Science 323,1033 (2009)
[3]Fernandez-Pañella et al. PRB 101, 184309 (2020)
[4]Cavalleri et al. PRL 95, 067405 (2005)
[5]Hirohata et al. J. Mag. Mat. 509 166711 (2020)
[6]Parchenko et al. Adv. Sci. 202302550 (2023)

Ultrafast X-ray absorption Spectroscopy

Pump-probe really demonstrates strengths

Synchronised femtosecond laser drivers an obvious strength

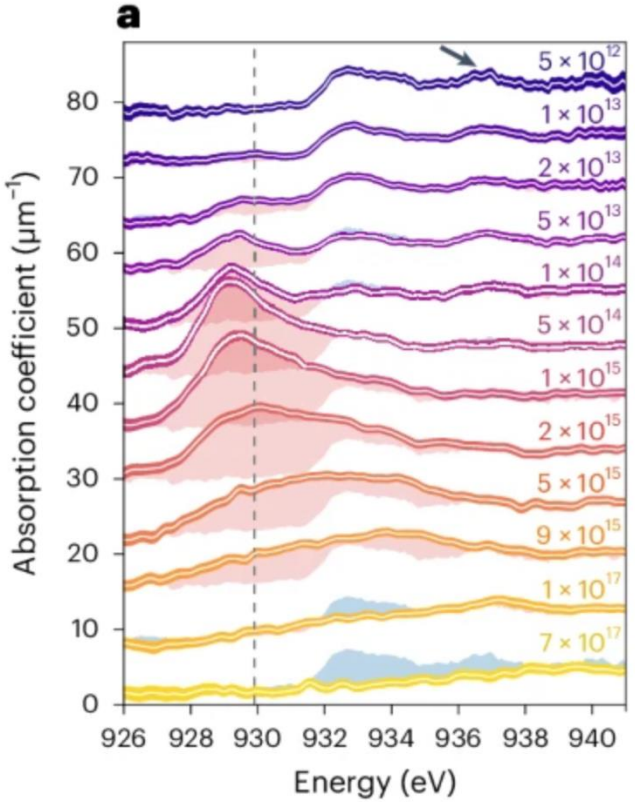
- Non-equilibrium dynamics of a laser driven WDM copper foil studied.
- Sub- 100 fs rise-time of the electron temperature measured using pre-edge peak feature at L-edge.



Mahieu *et al.* Nat. Comms. 9, 3276 (2018)

- XFEL investigations possible, but narrow bandwidth and spikey spectrum and spectral range is challenging
- Here, XFEL was both pump and probe;
- Non-linear absorption effects seen from reverse saturable absorption, to saturable absorption at higher intensities.

(200-1000 shots per scan)



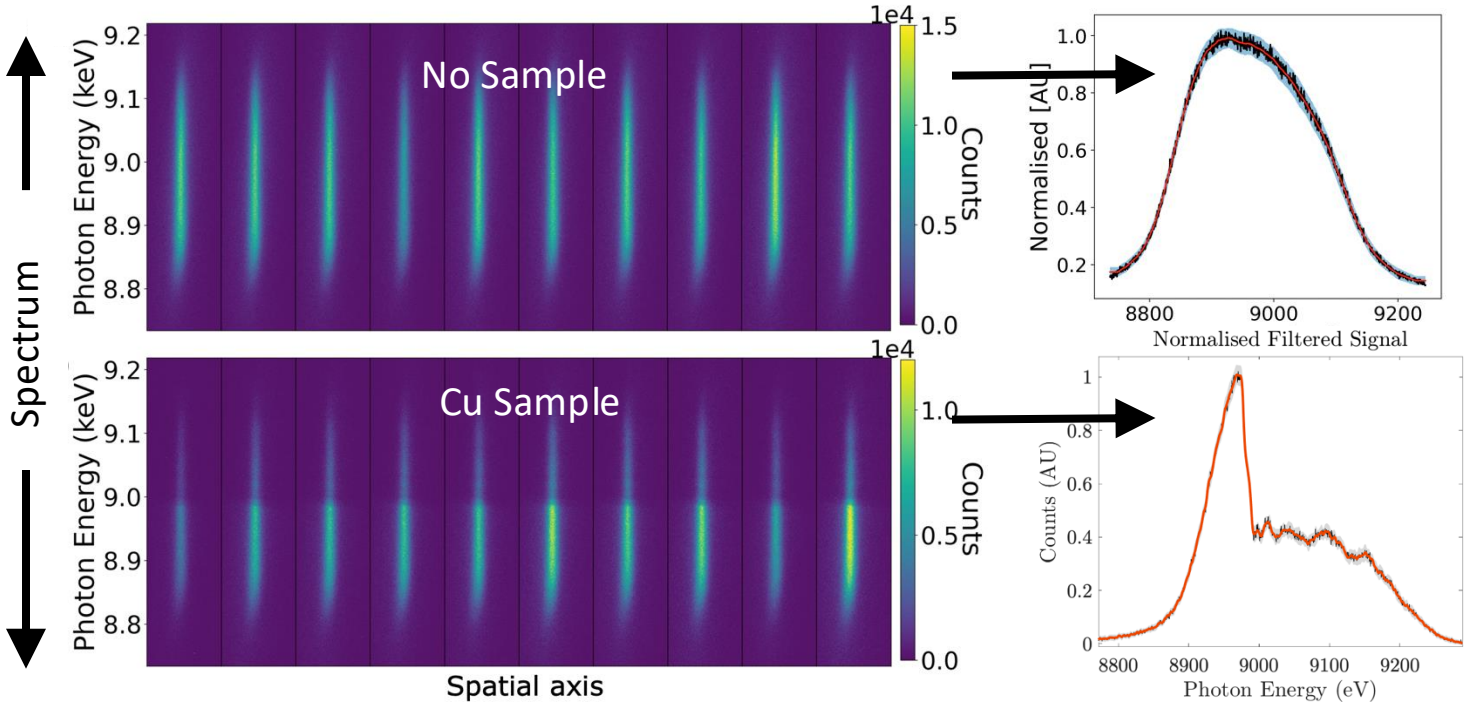
Mercadier *et al.* Nat Phys 20 1564 (2024)

Ultrafast X-ray absorption Spectroscopy

LWFA X-ray source status

Gemini, CLF (~200 TW)
~700 MeV electrons, ~100 pC

~10^6 photons/eV/shot



10 consecutive shots; great spectral stability, no on-shot reference required

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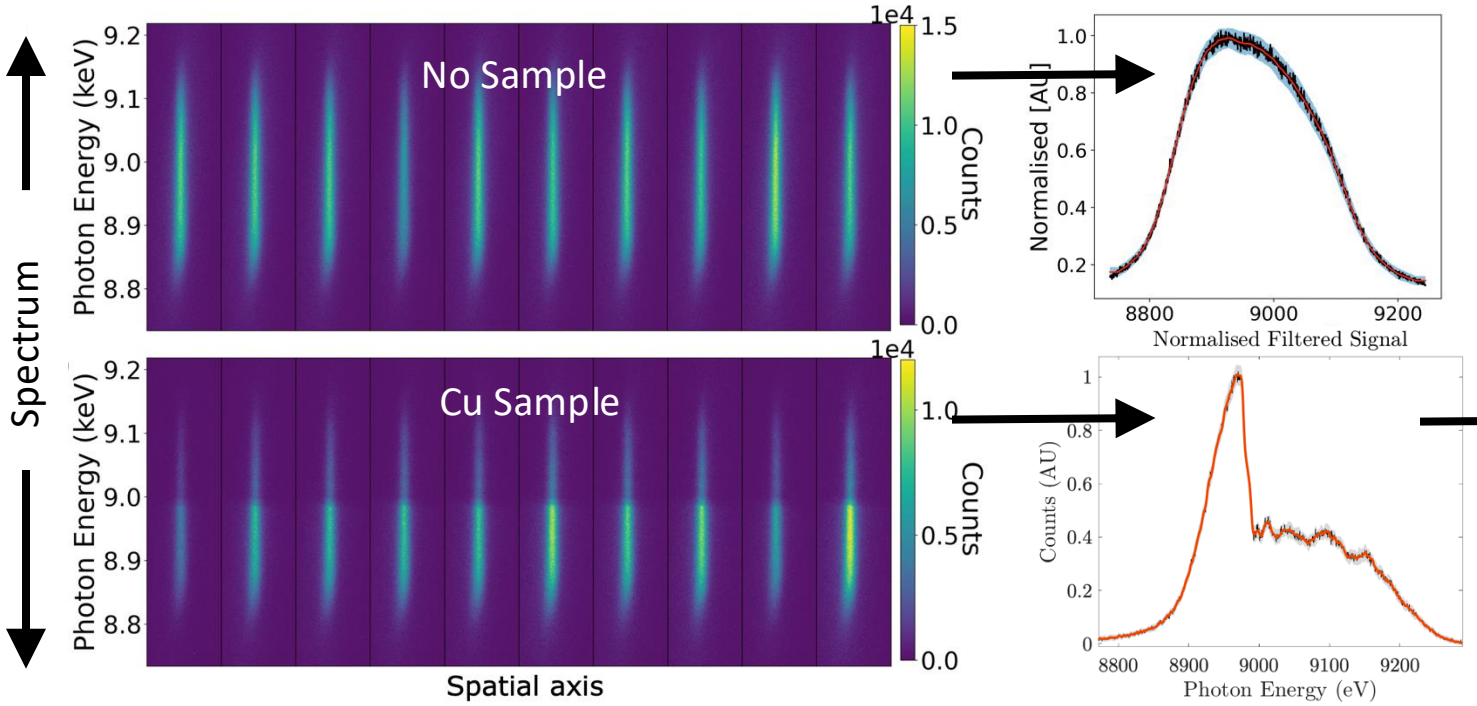
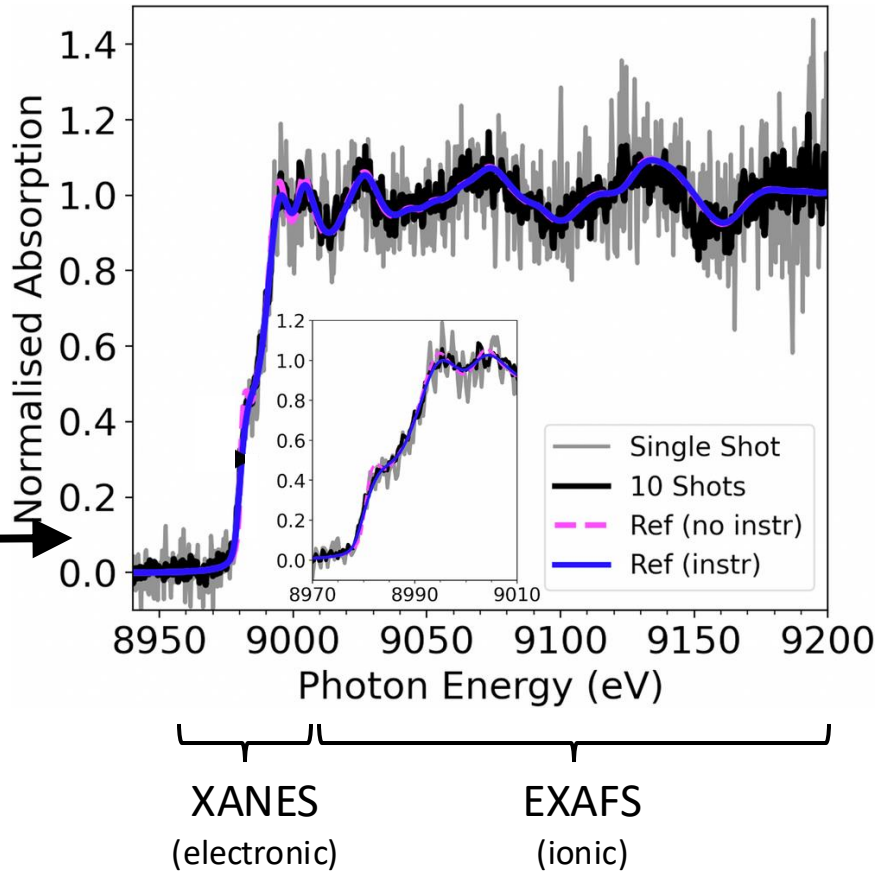


Single shot demonstration



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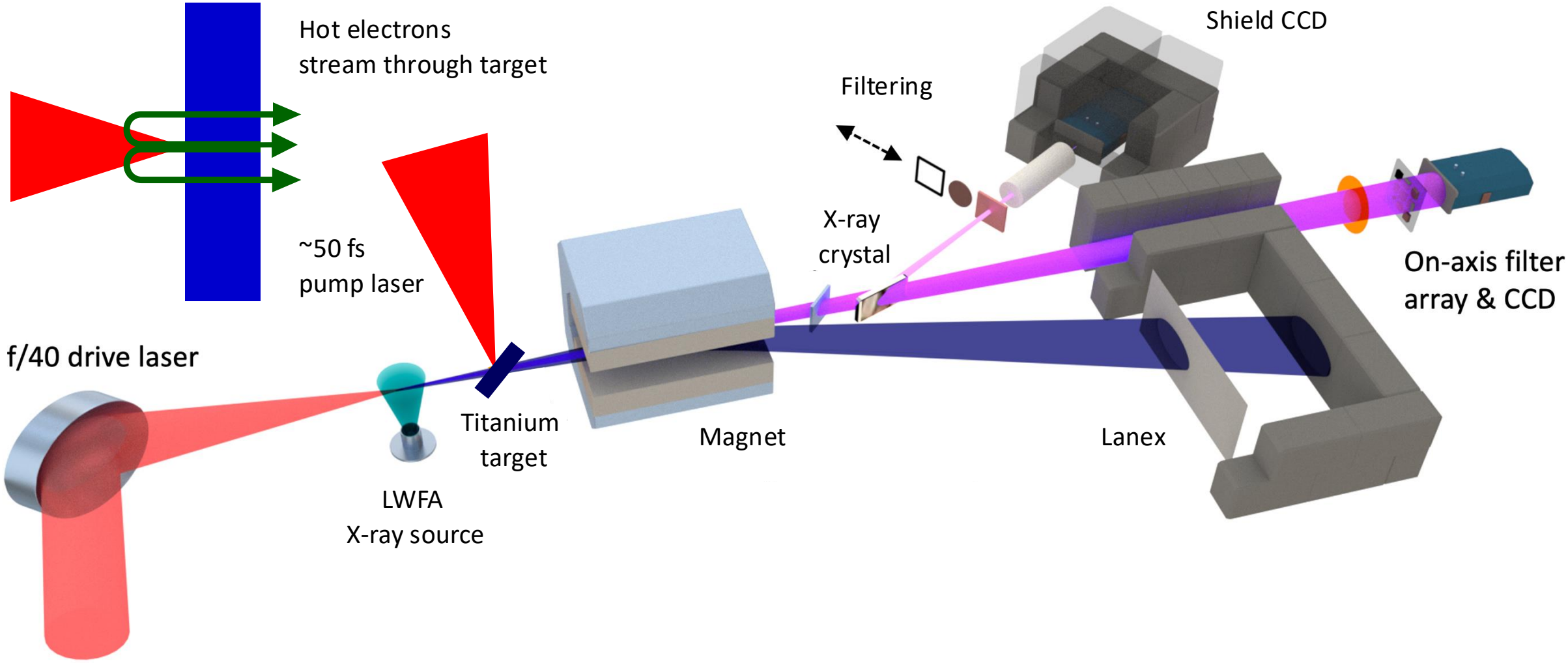
Kettle *et al.* Comms Phys 7, 247 (2024)

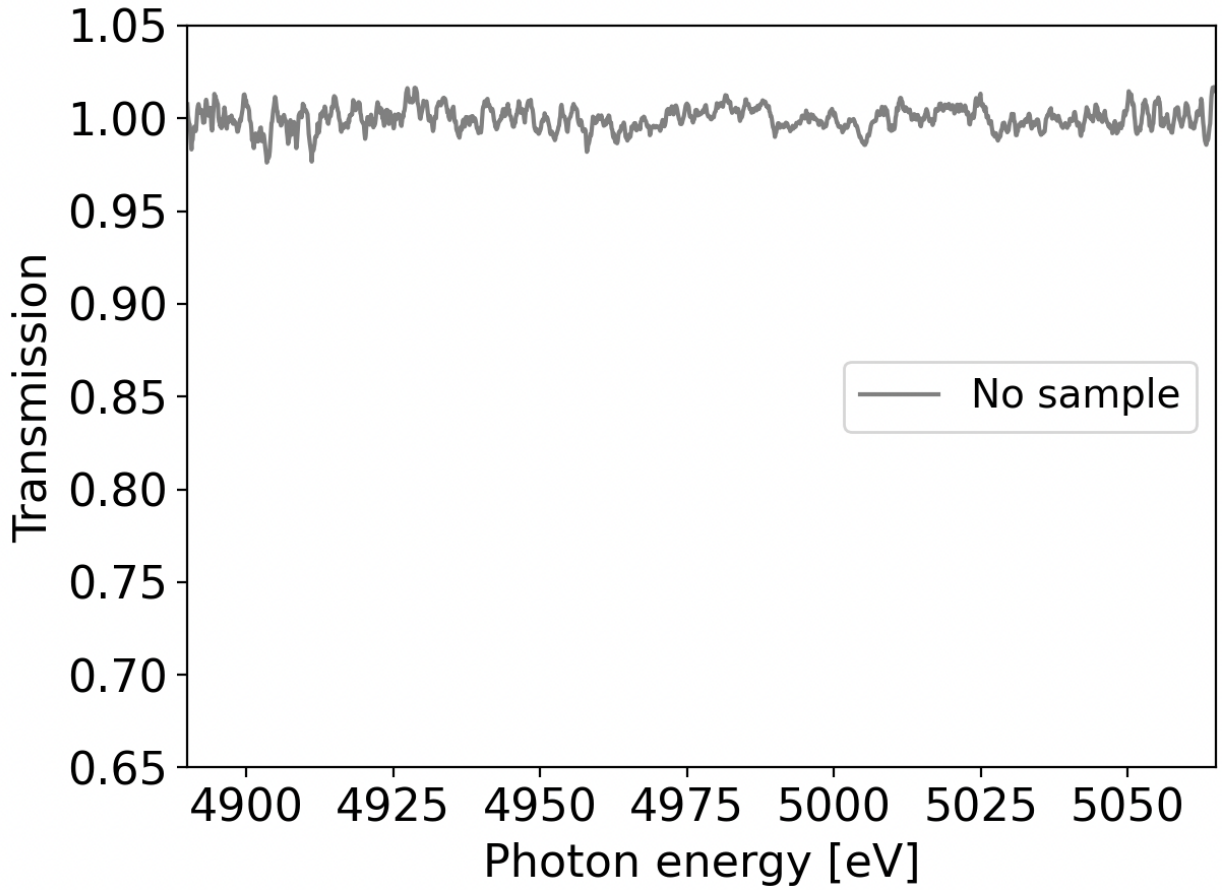
Ultrafast X-ray absorption Spectroscopy

Gemini February 2025 – pump probe investigation



Ultrafast X-ray absorption Spectroscopy

Gemini February 2025 – pump probe investigation



Ultrafast X-ray absorption Spectroscopy

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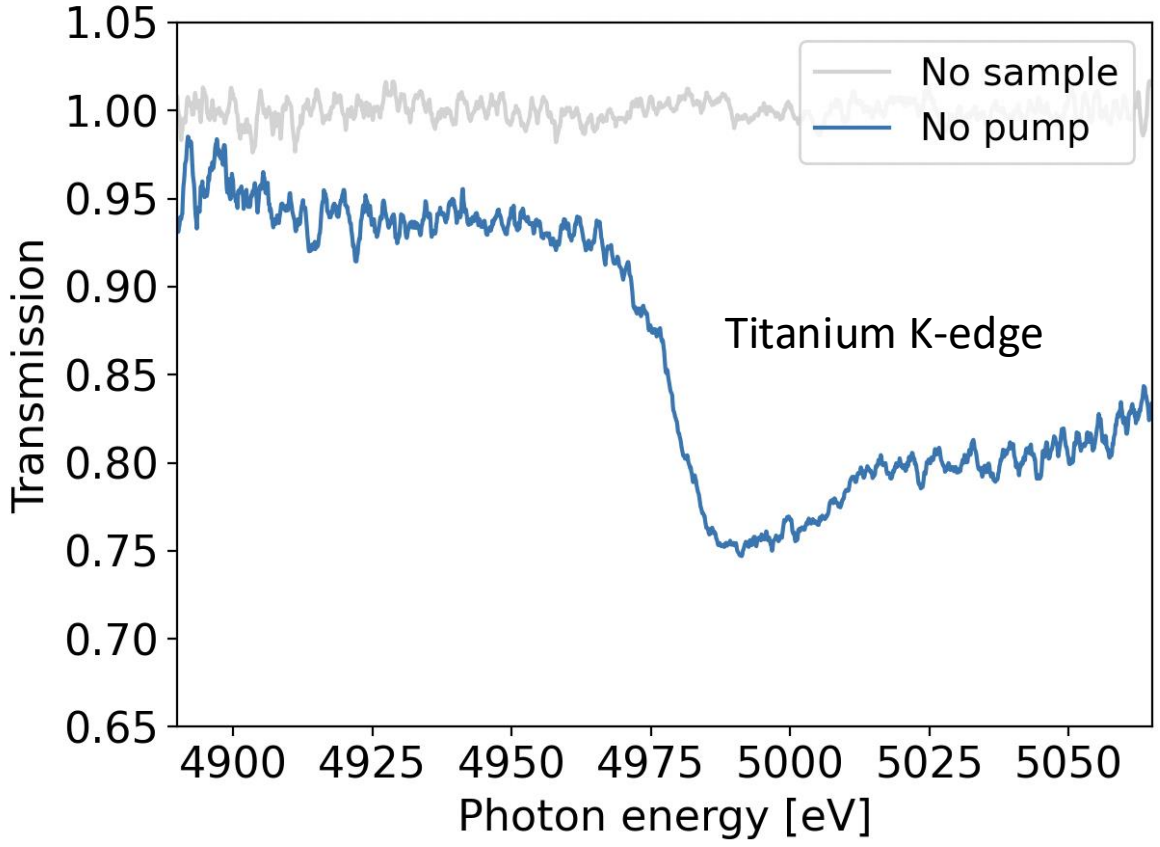
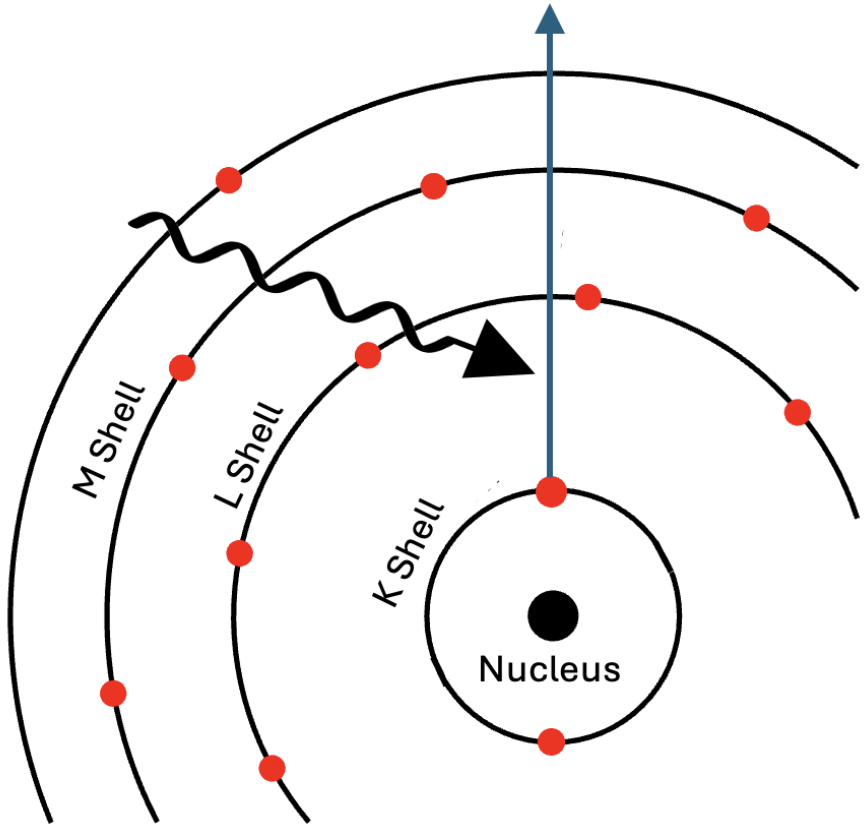




Ultrafast X-ray absorption Spectroscopy

Gemini February 2025 – pump probe investigation

Work in progress…

Ultrafast X-ray absorption Spectroscopy

Gemini February 2025 – pump probe investigation

Work in progress…

Ultrafast X-ray absorption Spectroscopy

Gemini February 2025 – pump probe investigation

Work in progress…

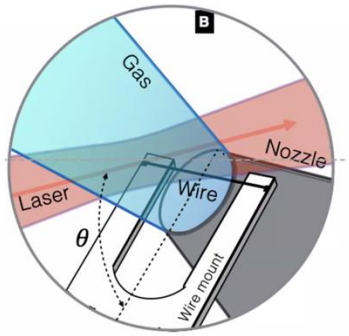
Ultrafast X-ray absorption Spectroscopy

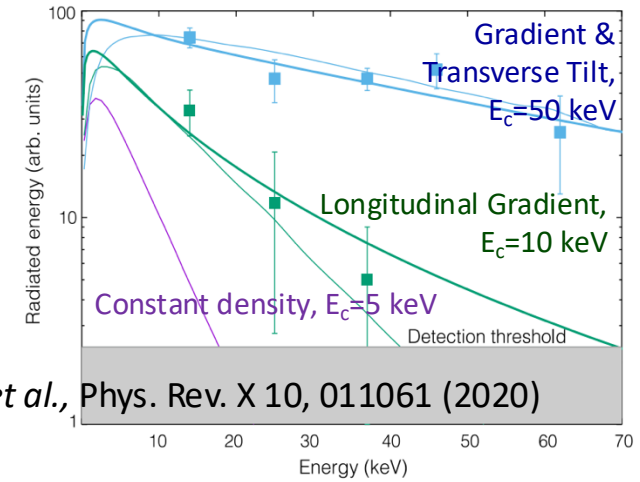
Future prospects

- Exploit temporal resolution of LWFA X-rays further; 10's fs or less.
- More facilities coming online with suitable LWFA driver in tandem to other high-power lasers.
- Given they can be inherently synchronised, this will provide access to wider parameter space to study. HEDP and extreme states.
- Tailored densities and increased drive laser power can improve X-ray source further.

https://www.clf.stfc.ac.uk/Pages/Vulcan-2020.aspx

E.g. Vulcan 20-20 at the CLF
10 kJ ns + 20 PW + 1 PW (LWFA)



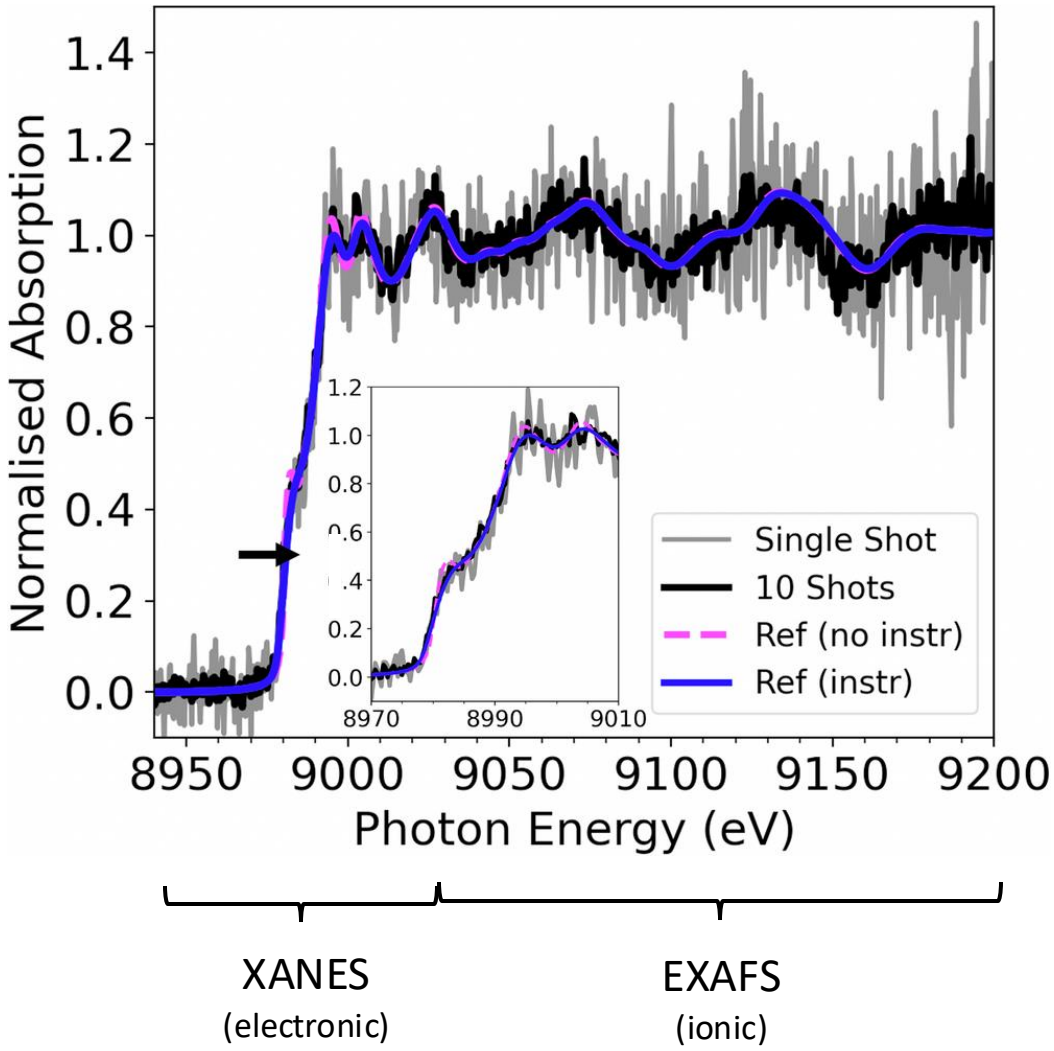


Kozlova *et al.*, Phys. Rev. X 10, 011061 (2020)

In Summary

- Multi-keV single-shot X-ray absorption spectroscopy using LWFA X-rays
- Allows investigation of electron and ion properties (simultaneously) on ultrafast timescales (10's fs).
- Inherent synchronisation and coupling to pump-probe systems.
- Powerful tool for studying many rapid processes, including HEDP and non-equilibrium studies.
- Important probe capability for future facilities.

Kettle *et al.* Comms Phys 7, 247 (2024)



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Thank you

Stuart Mangles[1], Steve Rose[1], Laurence Bradley[1], Rory Baggott[1], Cary Colgan[1], Annabel Gunn[1], Eva Los[1], Greg Christian[1], Adam Hughes[1], Elias Gerstmayr[2], Matthew Streeter[2], Shane McManus[2], Dave Riley[2], Niall Cavanagh[2], Gianluca Sarri[2], Oli Finlay[3], Chris Thornton[3], Dan Symes[3], Sam Astbury[3], Chris Spindloe[3], Rajeev Pattathil[3], Alec Thomas[4], Olle Lundh[5], Kristoffer Svendsen[5], Cornelia Gustafsson[5], Kateřina Falk[6], Michal Šmíd[6], Chris Murphy[7], Owen Lawrence[7], Joe Grieves[8], John Neeley[8]

[1] Imperial College London, U.K.
[2] Queens University Belfast, U.K.
[3] Central laser Facility, U.K.
[4] University of Michigan, USA
[5] Lund University, Sweden
[6] HZDR, Germany
[7] University of York, U.K.
[8] AWE, U.K.

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Central Laser Facility

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of York

Ultrashort pulse X-ray absorption spectroscopy using laser-plasma accelerators

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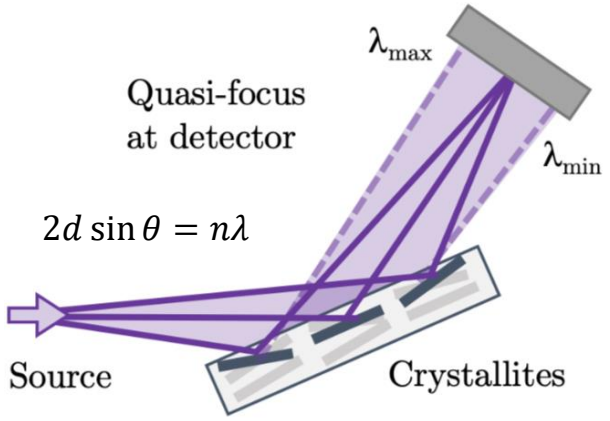
Mosaic crystals; High efficiency Bragg reflection



Solid rays: Single wavelength
Dashed rays: Min/max wavelength

- Bragg reflections provide sweep of X-ray energies on detector.
- **Mosaic**: Random orientation of crystallite planes provides **approximately x10 in efficiency** compared to "perfect" crystals, with small sacrifice in resolution.

Graphite 2d = 6.708 Å
Energy Range ≈ **2-10 keV**

Resolution:
dE/E ≈ 2000

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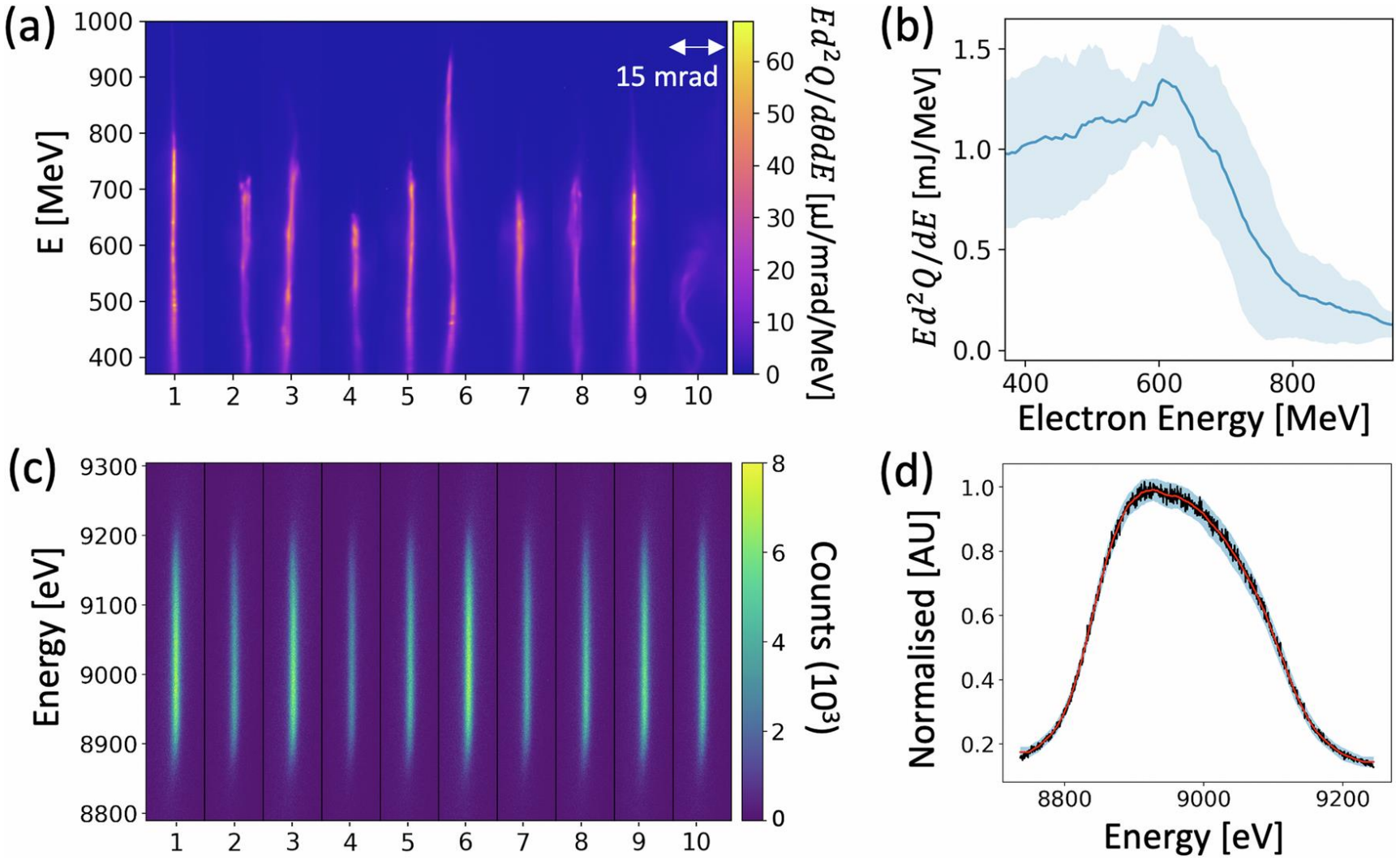
(a)
15 mrad
E [MeV]
$Ed^2Q/d\theta dE$ [μJ/mrad/MeV]
(b)
Ed^2Q/dE [mJ/MeV]
Electron Energy [MeV]
(c)
Energy [eV]
Counts (10^3)
(d)
Normalised [AU]
Energy [eV]