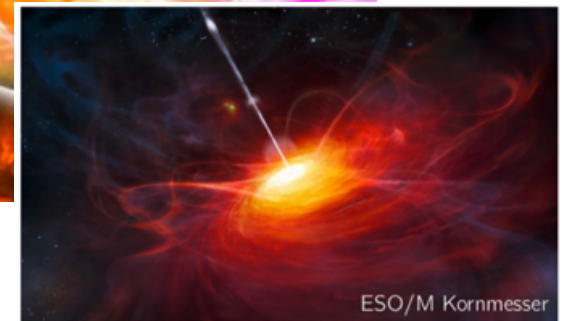
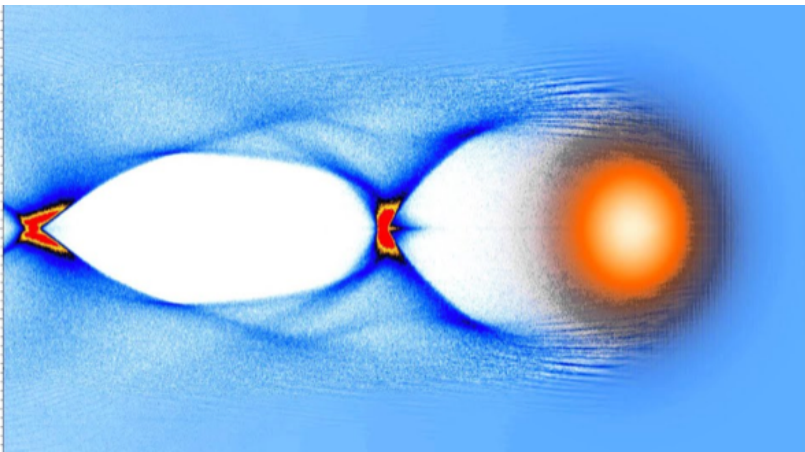


# Using X-rays from a laser wakefield accelerator to perform ultrashort multi-Kev absorption spectroscopy

**Brendan Kettle**

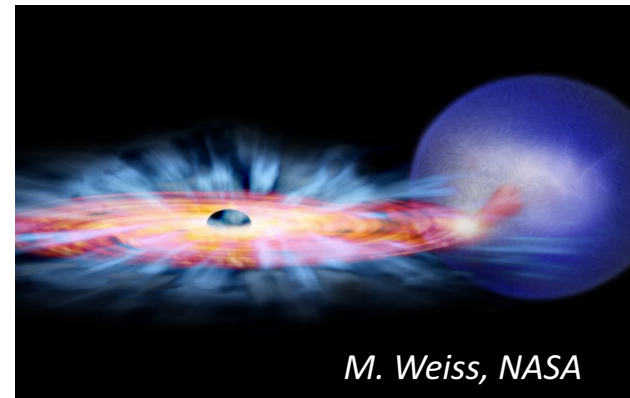
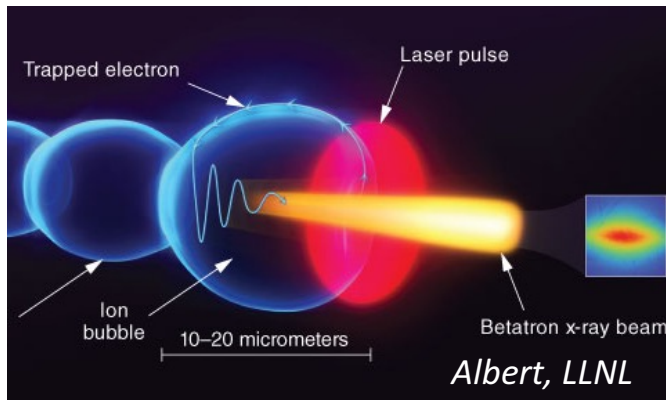
John Adams Institute for Accelerator Science, Imperial College London

JAI Fest 2019, 6<sup>th</sup> December, Oxford University



## The TeXME<sub>x</sub> Project

*Time resolved X-ray probing of Matter under Extr<sub>e</sub>m<sub>e</sub> conditions*

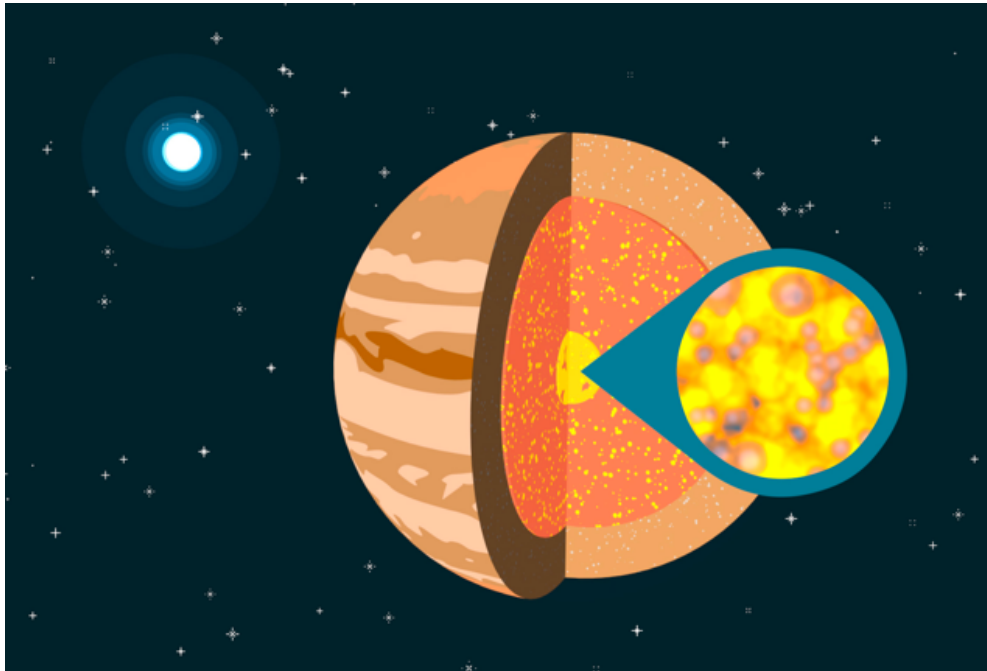


Academics: Stuart Mangles, Steven Rose

Post-docs: Brendan Kettle, Rory Baggott

PhD's: Robbie Watt, Cary Colgan, Eva Los, Wei Wu

## Extreme conditions? .... High Energy Density Matter



Densities  $> 1 \text{ g/cc}$

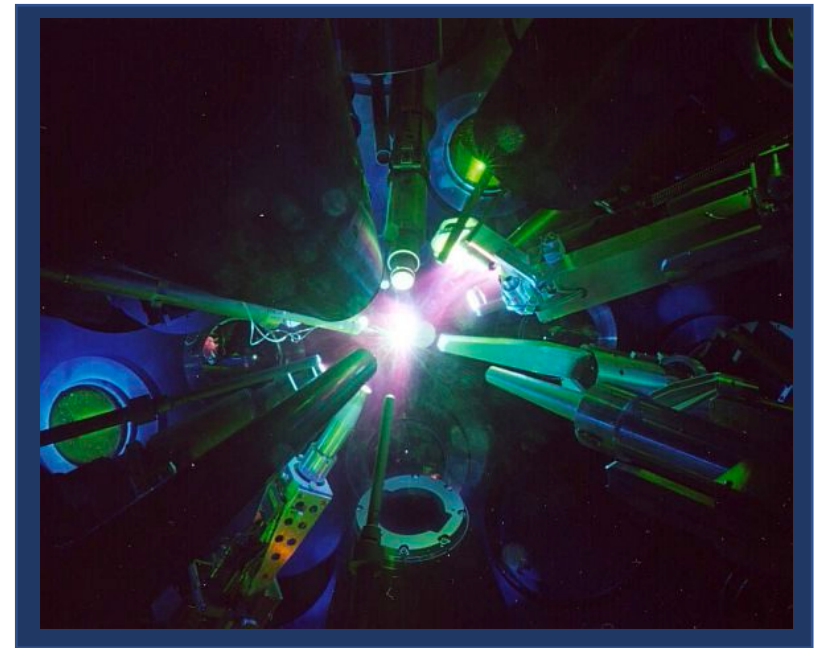
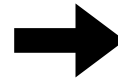
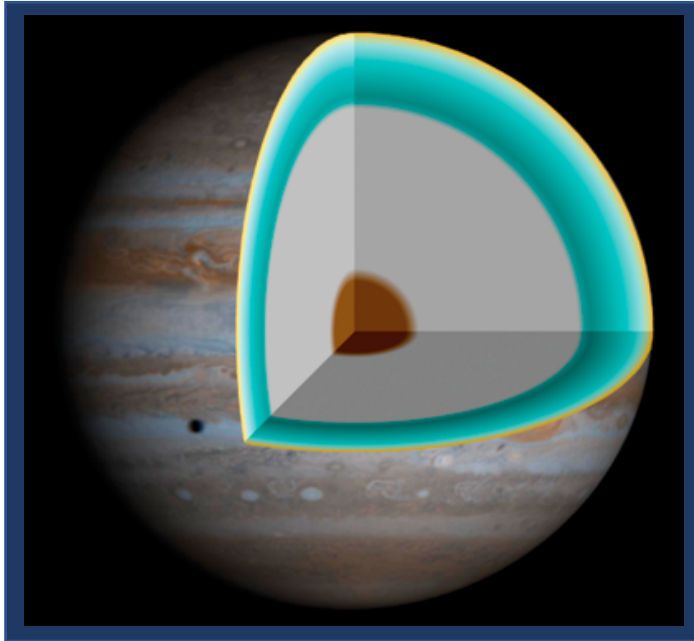
**Warm Dense matter (WDM):**  
1-100 eV. E.g. planets, ICF

$$\Gamma = \frac{(Z^* e)^2}{R_i k T_i}$$

**Hot Dense matter (HDM):**  
 $> 100 \text{ eV}$ . E.g. stars.

Many processes governing dense energetic plasmas remain untested/unquantified due to theoretical complexity and experimental difficulties.

## Why perform ultrafast X-ray absorption measurements?



▷ **X-rays:** Samples too dense for optical probing. Allows access to core electrons.

▷ **Ultrafast:** Transient states (in the lab) and rapid processes; sub-picosecond timescales.

# Ultrafast X-ray absorption measurements

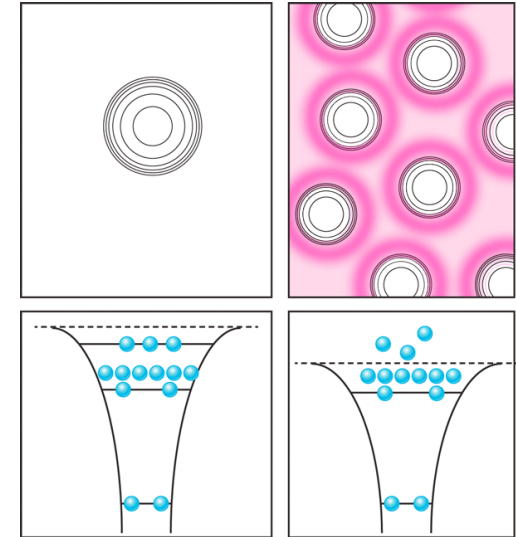
Processes &  
Techniques

- X-ray Absorption Near-Edge Structure (XANES)
  - Extended X-ray Absorption Fine Structure (EXAFS)
  - Ionisation Potential Depression (IPD).
  - Excited state absorption lines.
  - Non-thermal processes.
- Etc.**



Provide

- Temperature
  - Electronic and local atomic structure
  - Ionisation energies/rates
  - Electron-Ion equilibration rates
- Etc.**



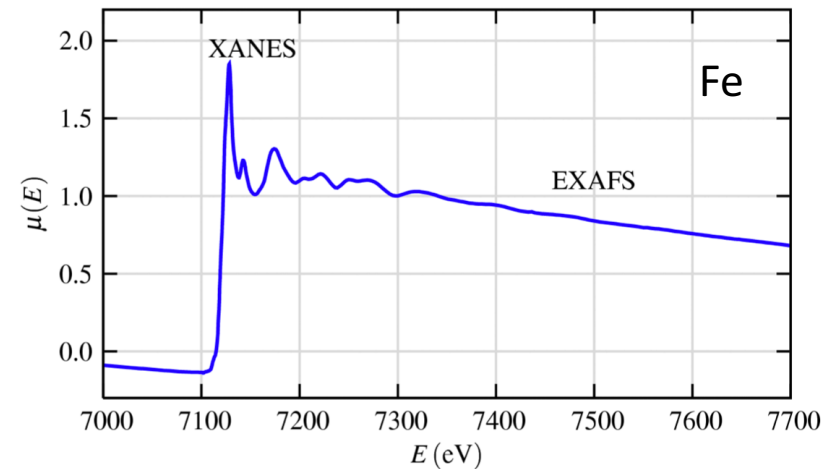
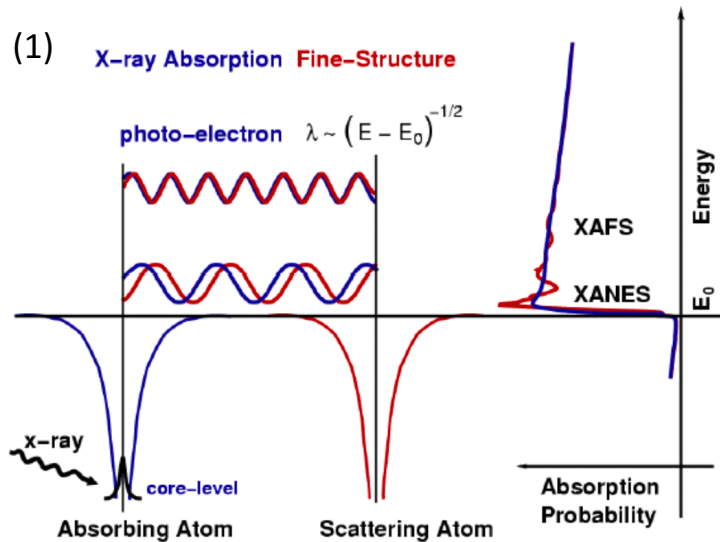
Umstadter, *Physics* 5, 88 (2016)

## Recent publications

Mahieu et al. *Nat. Comms*, 9, 3276 (2018)  
 Dorchie et al. *Phys. Rep.* 657, 1-26 (2016)  
 Cho et al. *PRL* 106, 167601 (2011)  
 Mančić et al. *PRL* 104, 035002 (2010)  
 Engelhorn et al. *PRB* 91, 214305 (2015)

Hoarty et al. *PRL* 110, 265003 (2013)  
 Circosta et al. *PRL* 109, 065002 (2012)  
 Iglesias. *HEDP* 12, 5-11 (2014)  
 Vinko et al. *Nat. Comm* 6, 6397 (2015)

## X-ray Absorption Near Edge Structure (XANES) & Extended X-ray Absorption Fine Structure (EXAFS)



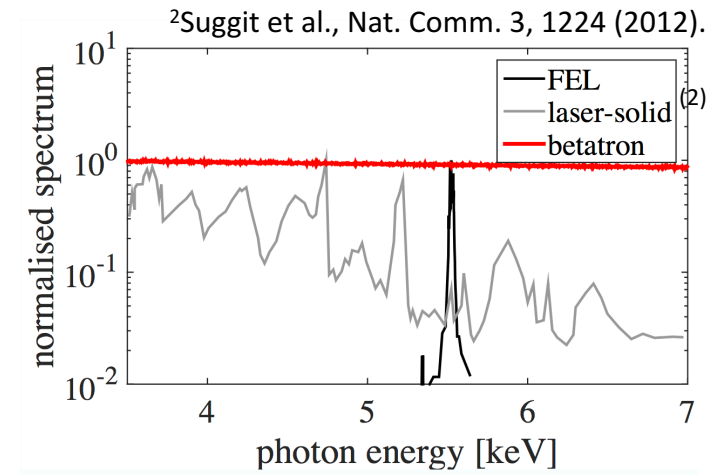
Local scattering of ejected photoelectrons cause modulations in absorption close to resonant edge.

Slope and peak positions detail **electronic & atomic structure** and broadening of features reveal **electron & ion temperature**.

<sup>1</sup> A Practical Introduction to Multiple Scattering Theory, Bruce Ravel, 2005

## What type of X-ray source do we need?

- High photon flux from small source – single shot if possible
- Multi-keV photon energies
- Short sub-picosecond duration
- Broadband and smooth spectrum
- High rep-rate is desirable
- Temporal matching to pump?



	Synchrotrons	XFELs	Laser-plasmas	LWFA
Duration	10 – 100 ps	< 100 fs	1 -10 ps	< 100 fs
Spectrum	Broad and Smooth	Narrow	Broad Not Smooth	Broad and Smooth

## Experiment Results

Stuart Mangles<sup>1</sup>, Steven Rose<sup>1</sup>, Rory Baggott<sup>1</sup>, Robbie Watt<sup>1</sup>, Jason Cole<sup>1</sup>, Elias Gerstmayr<sup>1</sup>,  
Nelson Lopes<sup>1</sup>, Alec Thomas<sup>2,3</sup>, Matthew Streeter<sup>2</sup>, Stephen Dann<sup>2</sup>, Yong Ma<sup>2</sup>, Amina Hussein<sup>3</sup>,  
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Felicie Albert<sup>7</sup>, Nuno Lemos<sup>7</sup>

<sup>1</sup> Imperial College London, U.K.

<sup>2</sup> Lancaster University, U.K.

<sup>3</sup> University of Michigan, USA

<sup>4</sup> Central laser Facility, U.K.

<sup>5</sup> Lund University, Sweden

<sup>6</sup> ELI Beamlines, Czech Republic

<sup>7</sup> Lawrence Livermore National Laboratory, USA



Science & Technology Facilities Council  
Central Laser Facility



LUND  
UNIVERSITY

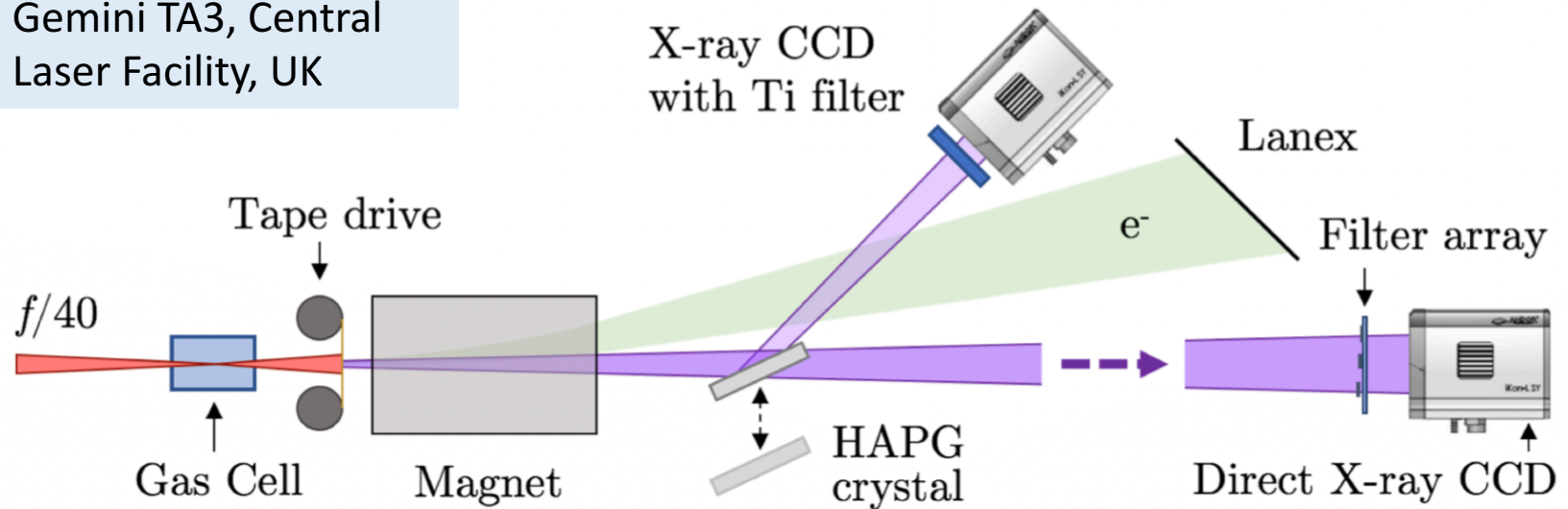


Lawrence Livermore  
National Laboratory

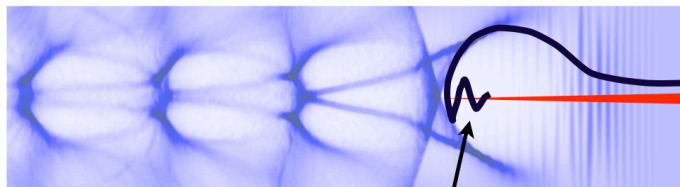


## Campaign in August 2017

Gemini TA3, Central  
Laser Facility, UK



density of high amplitude plasma wave



trapped electron  
trajectory

bright X-ray flash

betatron oscillations

### Drive laser

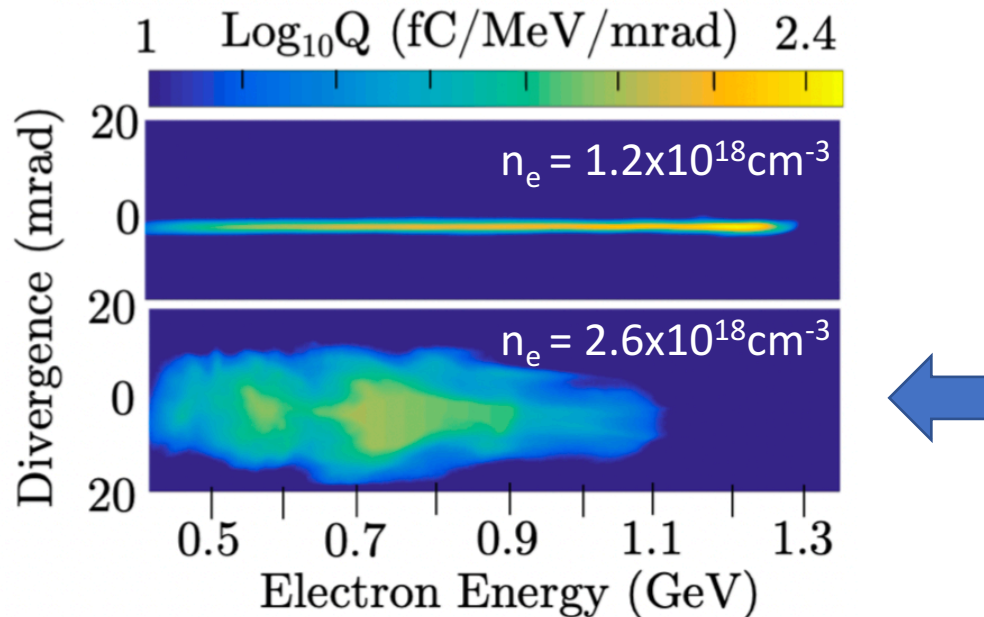
Duration =  $47 \pm 5$  fs

Energy =  $9 \pm 0.3$  J (43% in FWHM)

Intensity =  $4.9 \pm 0.6 \times 10^{18}$  W/cm<sup>2</sup>  
( $a_0 \approx 1.5$ )

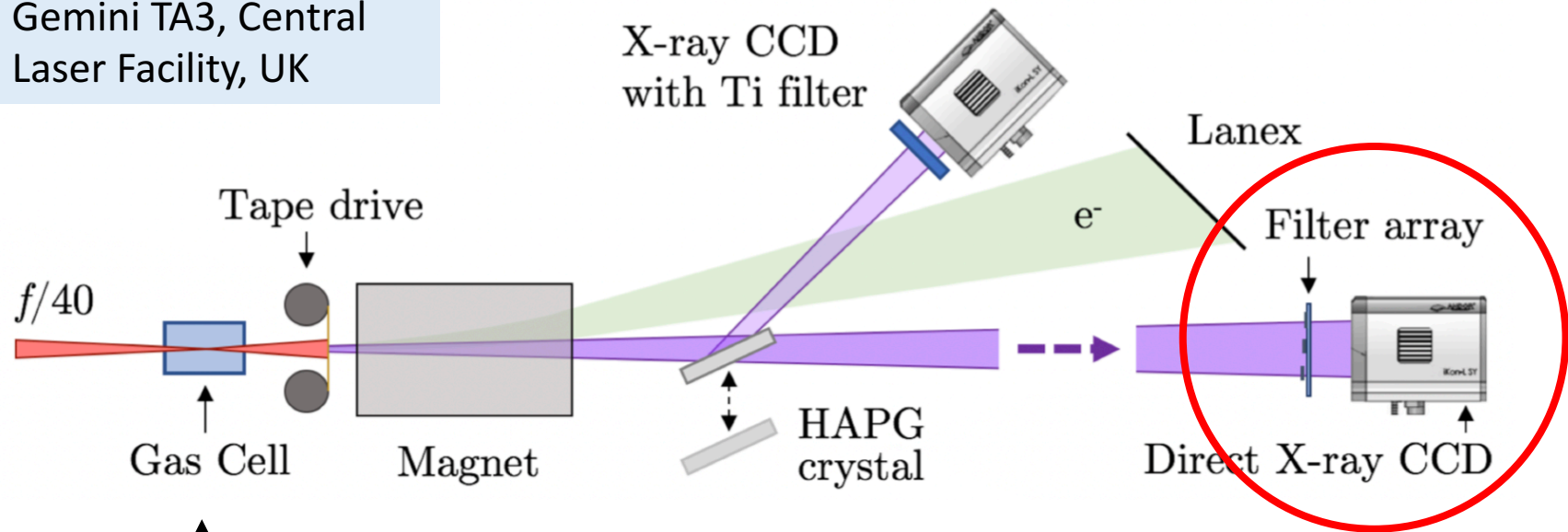
## Laser-wakefield accelerator performance

- Two stage gas cell. First stage (3mm long) 2% Nitrogen mix for **ionisation injection**, second stage  $\approx 20$ mm long, pure helium.
- Went for **high charge** (and **high X-ray flux**) “messy” electron beams (using increased density in first cell).

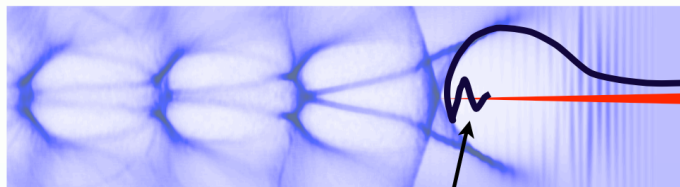


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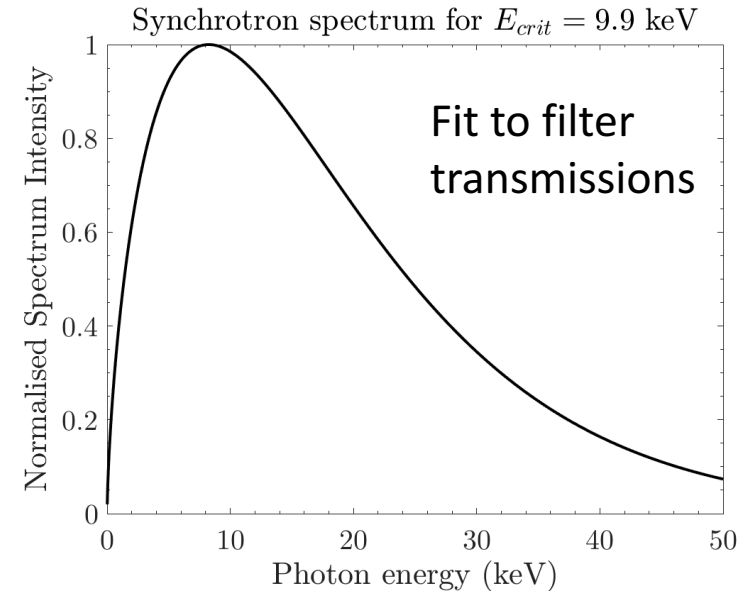
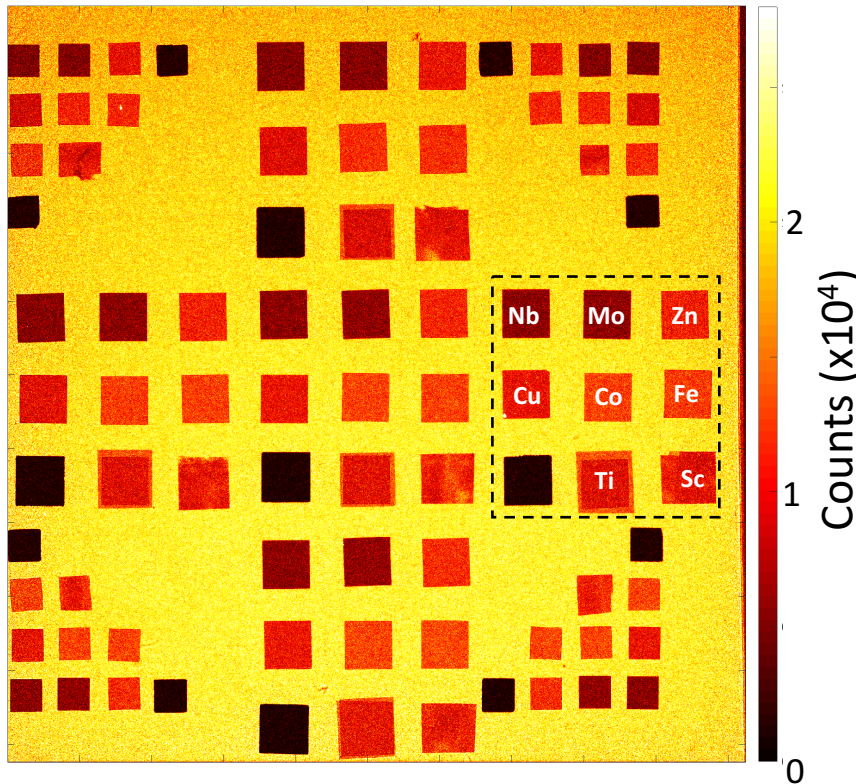
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( $a_0 \approx 1.5$ )

## LWFA X-ray results – broad spectrum using filter array



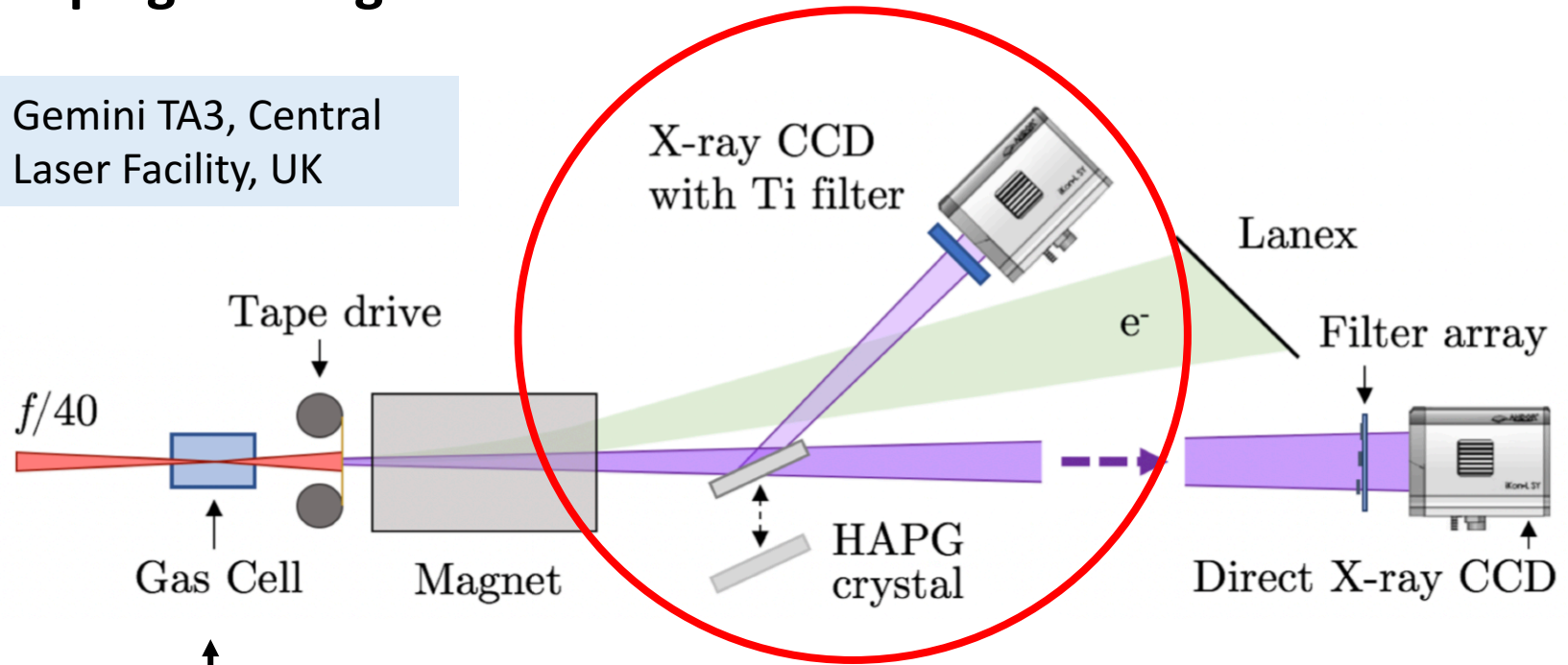
$$\left. \frac{d^2I}{dE d\Omega} \right|_{\theta=0} \propto \gamma_e^2 \left( \frac{E}{2E_{crit}} \right)^2 \mathcal{K}_{2/3}^2 \left( \frac{E}{2E_{crit}} \right)$$

$E_{crit} = 9.9 \pm 1.5$  keV, and the entire beam contained  $7.2 \pm 2.8 \times 10^5$  photons/eV at 5 keV

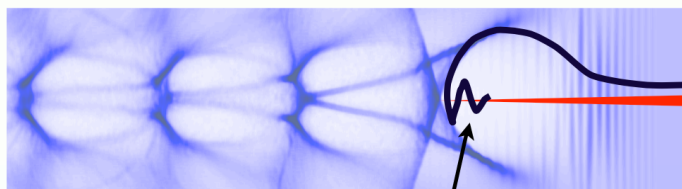
( $\approx 10^{22}$ - $10^{23}$  photons/s/mm<sup>2</sup>/mrad<sup>2</sup>/0.1% BW)

## Campaign in August 2017

Gemini TA3, Central  
Laser Facility, UK



density of high amplitude plasma wave



betatron oscillations

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### Drive laser

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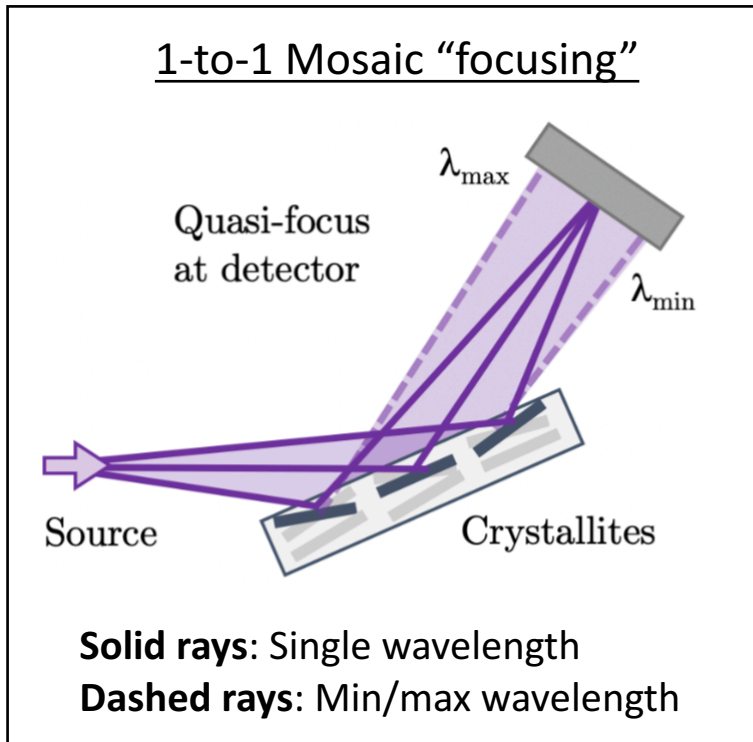
Intensity =  $4.9 \pm 0.6 \times 10^{18}$  W/cm<sup>2</sup>  
( $a_0 \approx 1.5$ )

## LWFA X-rays – High resolution using crystal spectrometer

Bragg's law  
 $2d \sin \theta = n\lambda$

$2d$  = Crystal lattice spacing (fixed)  
 $\theta$  = Incident ray angle  
 $\lambda$  = Diffracted X-ray wavelength

Graphite  $2d = 6.708 \text{ \AA}$   
Range of energies  $\approx 2 - 10 \text{ keV}$

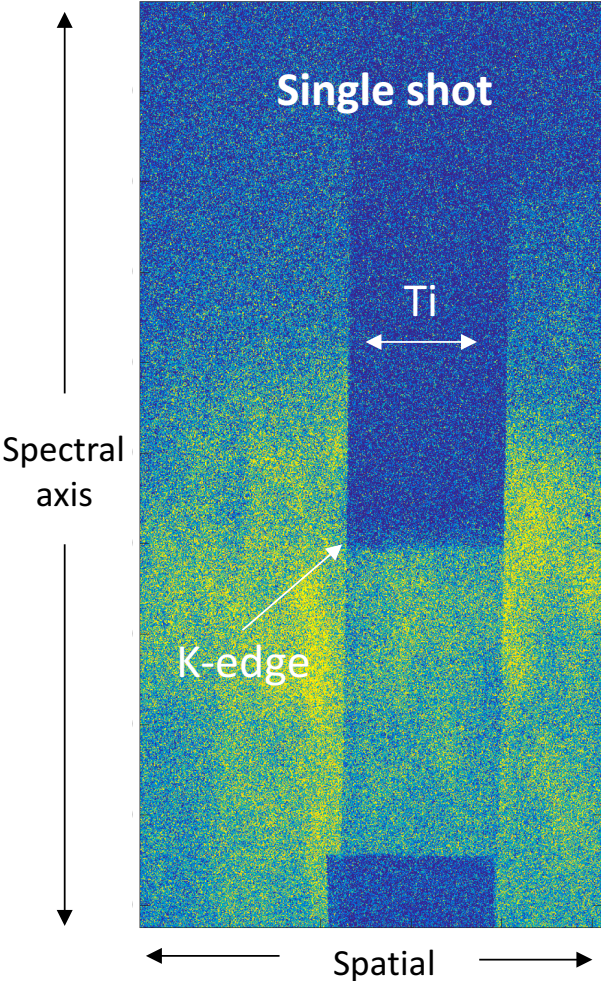


- Bragg reflections provide sweep of X-ray energies on detector.
- **Mosaic:** Random orientation of crystallite planes allow more rays to be diffracted to detector - **10 times more efficient** than "perfect" crystal, with small sacrifice in resolution.

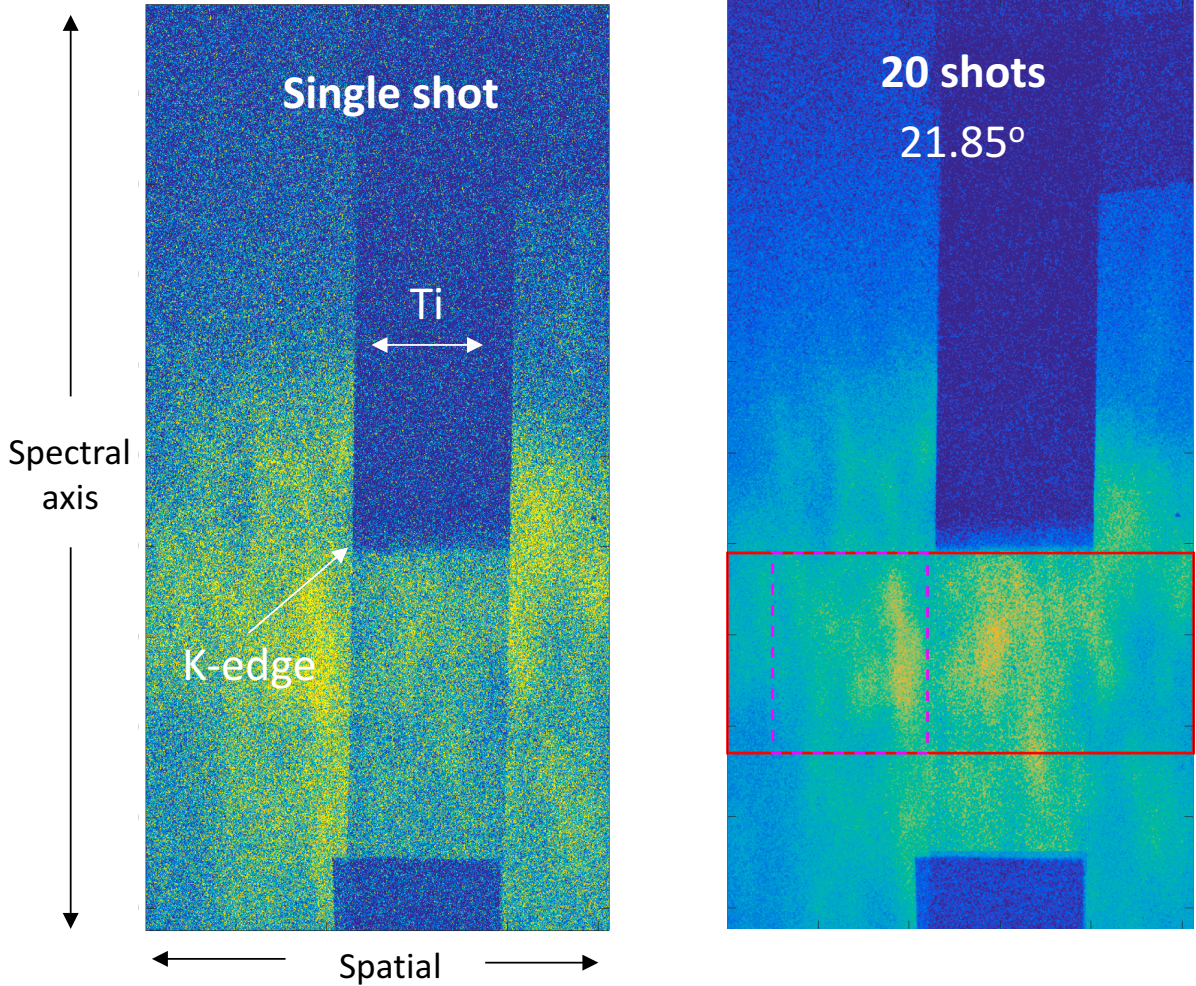
Resolution:  
 $dE/E \approx 2000$

Zastrau et al. J.Inst. 8 P10006 (2013)

# Crystal spectrometer results

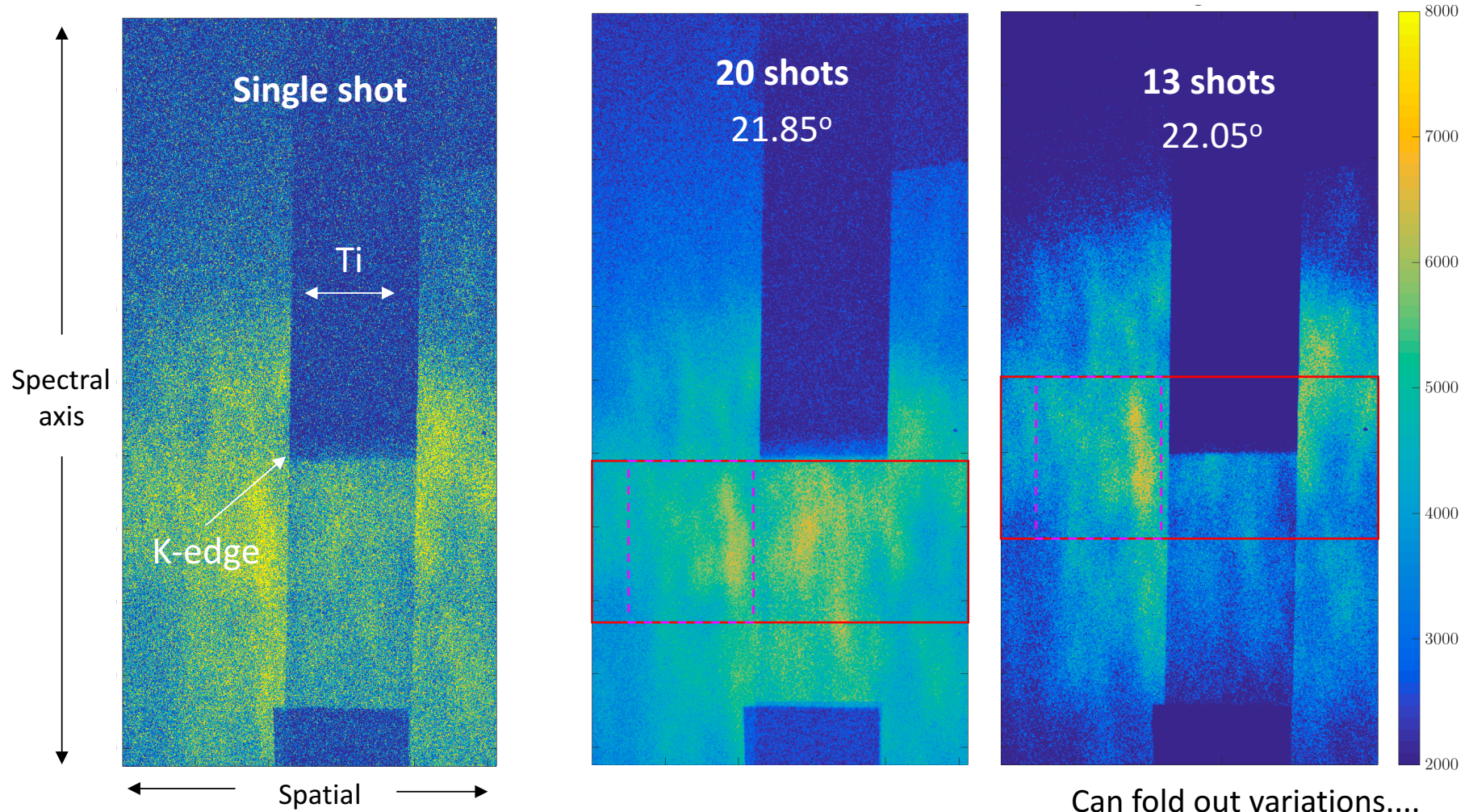


# Crystal spectrometer results

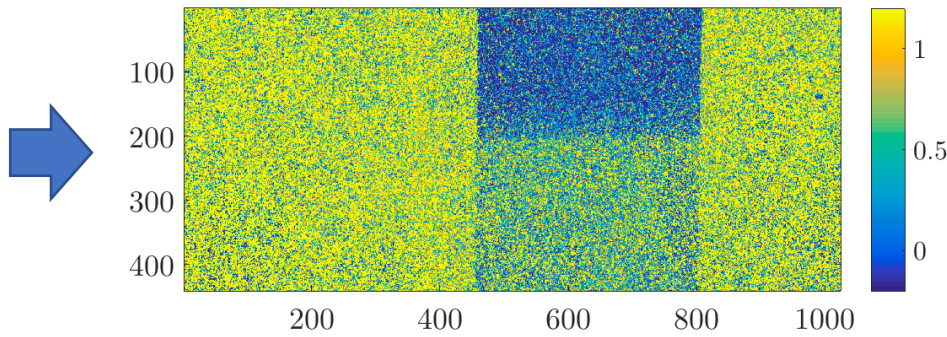




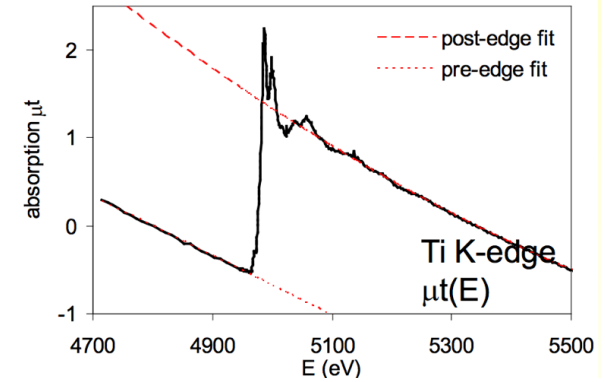
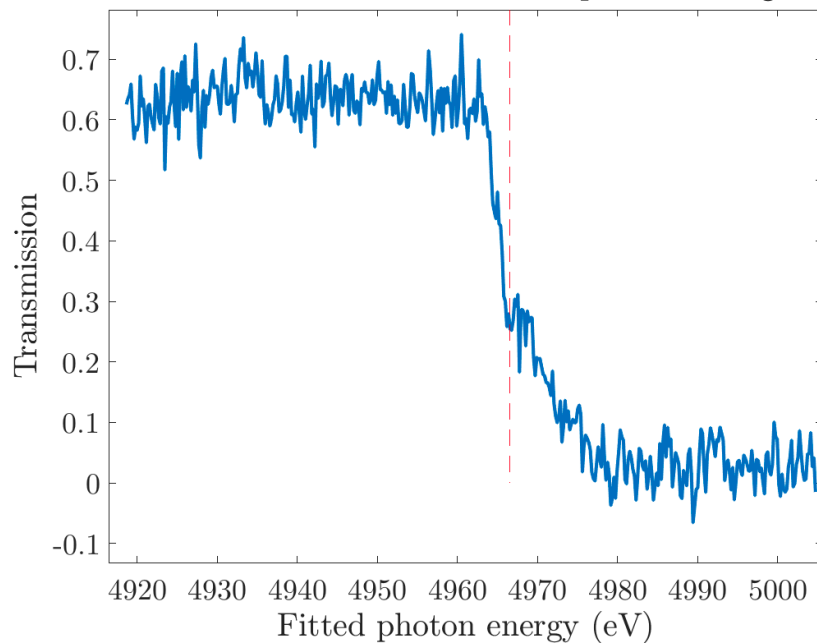
# Crystal spectrometer results



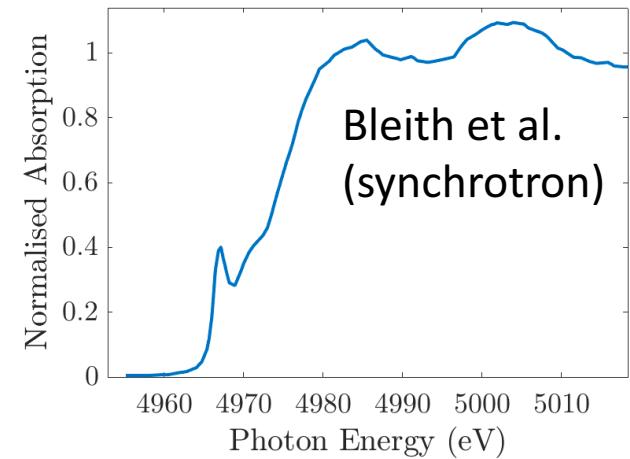
Corrected map



Transmission Lineout - with spectral fitting



$$\chi(E) = \frac{\mu(E) - \mu_0(E)}{\mu_1(E)}$$

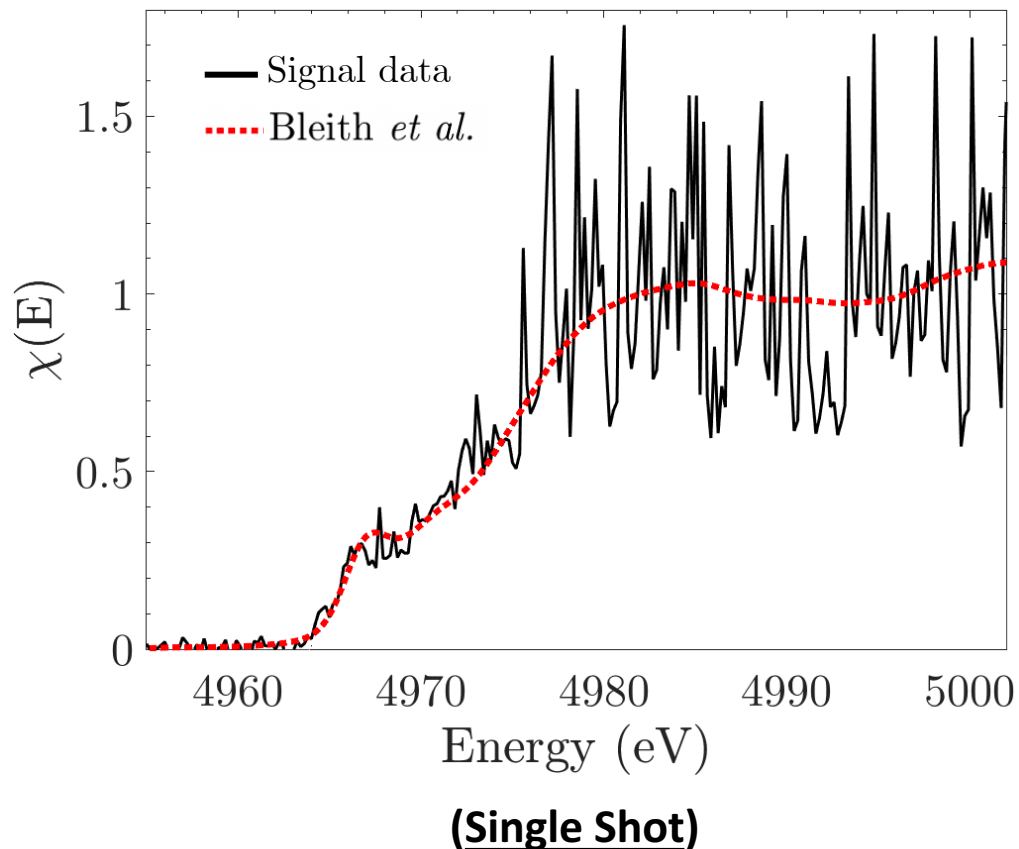


<sup>1</sup> *A Practical Intro. to Multiple Scattering Theory*, B. Ravel, 2005

<sup>2</sup> *Bleith et al. J. Mater. Chem. A*, 2014, 2, 12513

## Titanium K-edge XANES measurements

Normalised absorption, 10 $\mu$ m Ti Foil

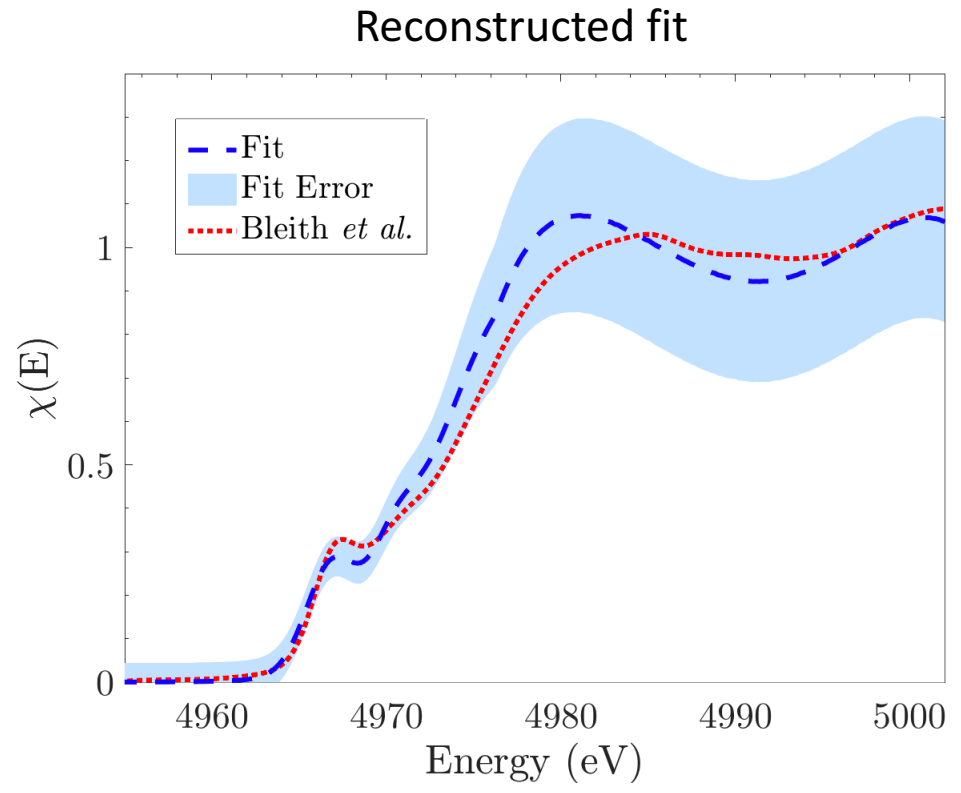
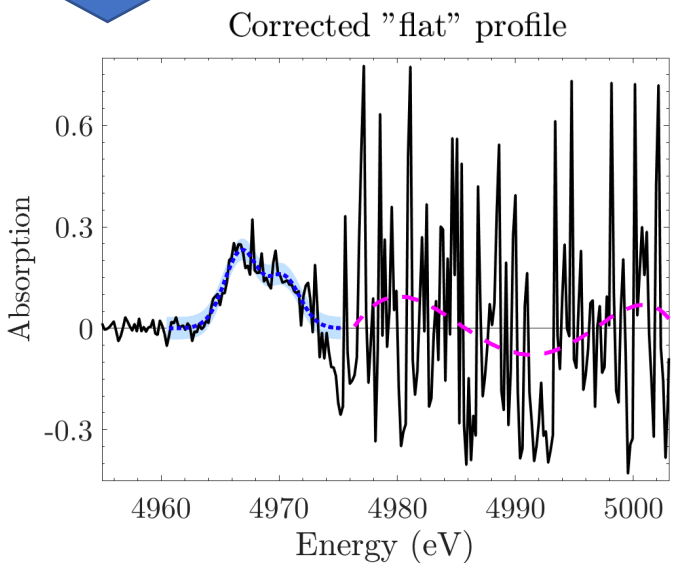
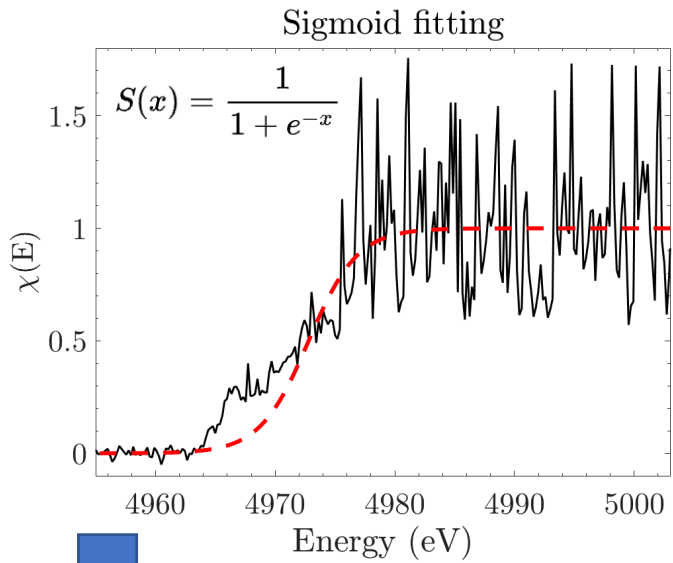


Direct spectral measurement of X-rays with approximately 2.2 eV resolution over an 80 eV window.

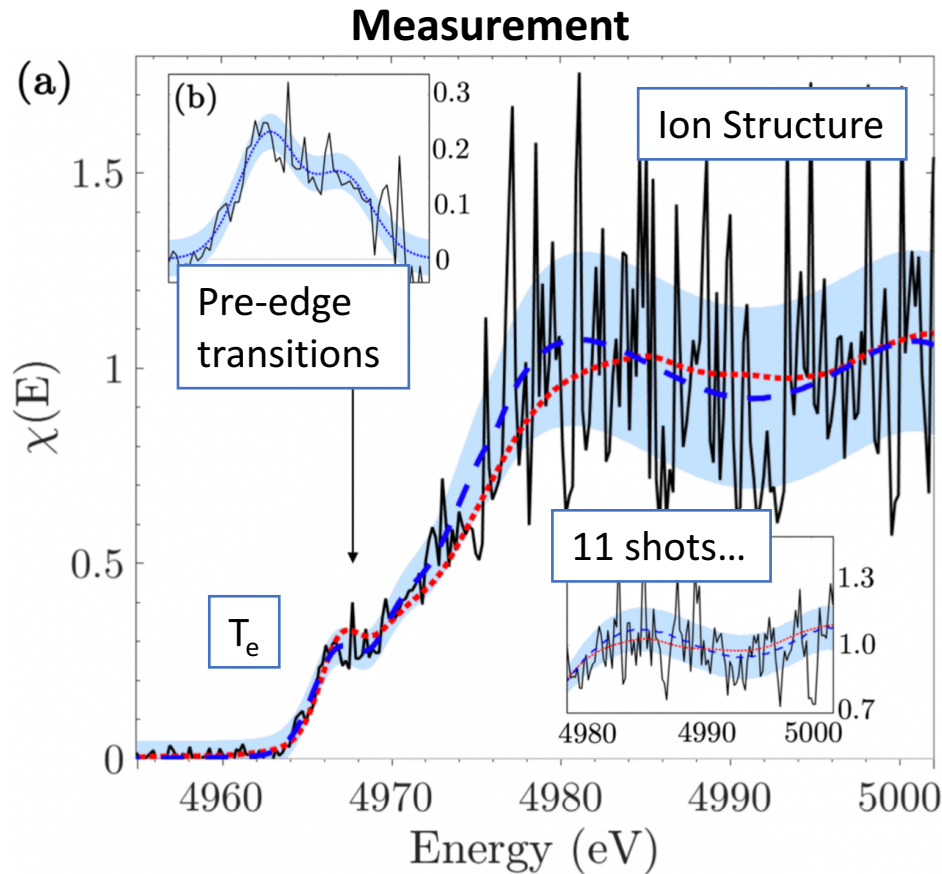
**Signal-to-noise = 300:1**

Poisson? .... should be 1100:1

... Noise is mostly from electron dump, not X-ray source and can be improved...



# Single Shot XANES measurement



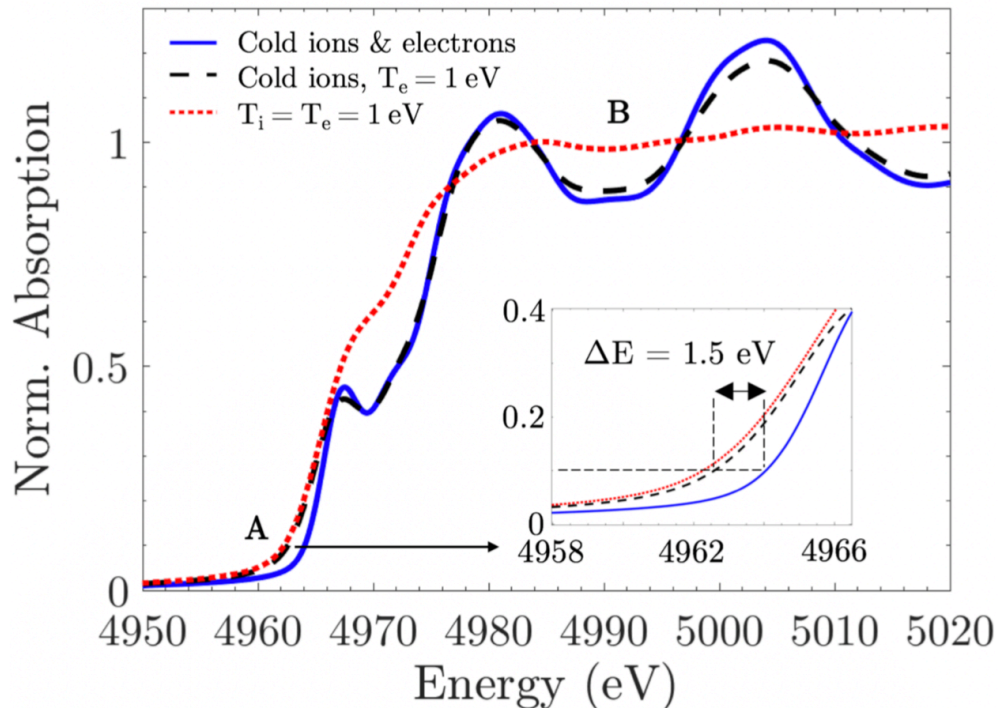
— Signal Data  
 - - - Signal Fit  
 ■ Fit error  
 ..... Sync. Ref.

- **Extracted features** - forbidden transitions into 3d shell (allowed by 3d-4p mixing)
- Estimate  $\Delta T_e \approx 0.4$  eV resolution in electron temperature change
- Ion temperature/structure in post edge structure

Recently accepted for PRL: <https://arxiv.org/abs/1907.10167>

## Model predictions for electron-ion equilibration in dense plasmas

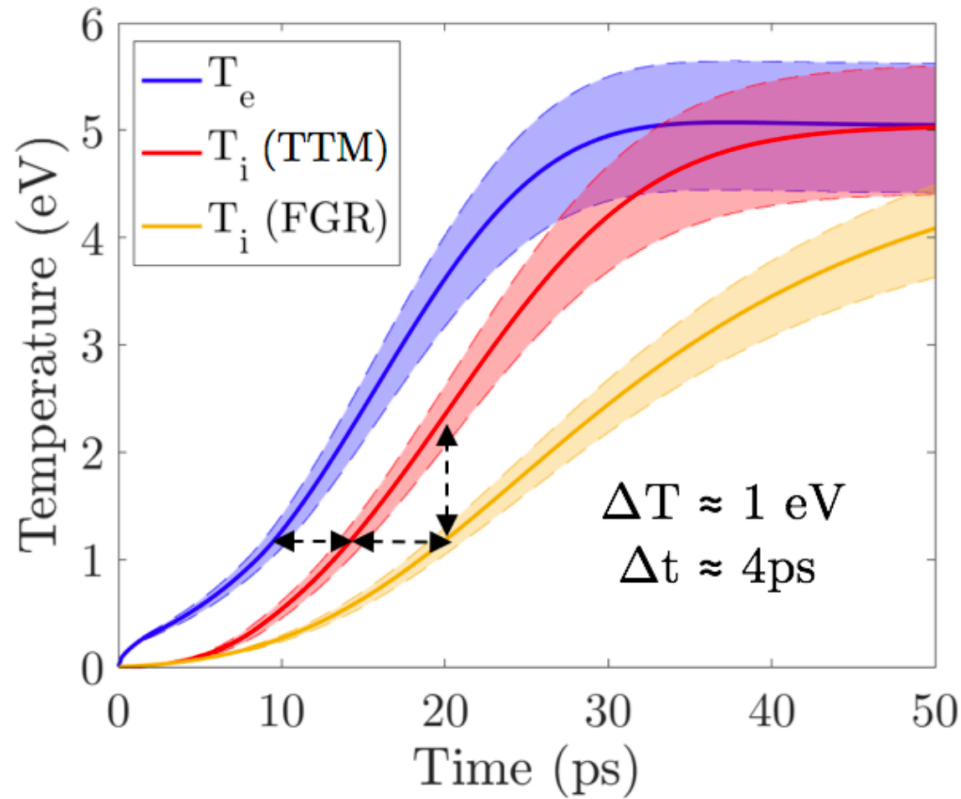
### Copper DFT simulations (GPAW)



- Electron and ion temperatures have individual signatures
- Can **independently measure the electron and ion temperature** on a single shot.
- Very interesting for high energy density samples due to ultrashort pulse duration of X-rays.

Simulations by Rory Baggott

## Model predictions for electron-ion equilibration in dense plasmas

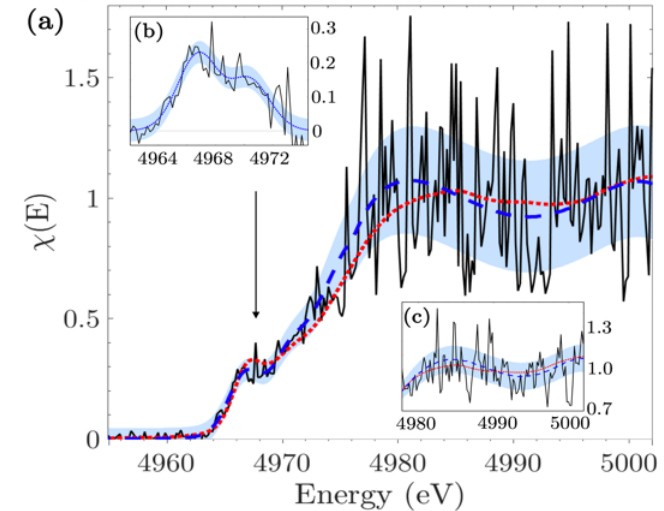


- Temperature equilibration estimates for Copper
- 20ps X-ray driver
- Comparing Two-temperature model with plasma based model

**Possible to measure equilibration rates of WDM and HDM.**

## In conclusion

- High resolution direct spectral measurement of LWFA X-rays
- Single-shot multi-keV absorption of Ti K-edge.
- With shielding/improved geometry this can be even better – post-edge leads to ion information.
- With appropriate heating scheme; Measurements of ultrafast processes in high energy density matter.



Single-shot multi-keV X-ray absorption spectroscopy using an ultrashort laser wakefield accelerator source

B. Kettle,<sup>1,\*</sup> E. Gerstmayr,<sup>1</sup> M.J.V. Streeter,<sup>2,†</sup> F. Albert,<sup>3</sup> R.A. Baggott,<sup>1</sup> N. Bourgeois,<sup>4</sup> J.M. Cole,<sup>1</sup> S. Dann,<sup>2,‡</sup> K. Falk,<sup>5,6</sup> I. Gallardo González,<sup>7</sup> A.E. Hussein,<sup>8</sup> N. Lemos,<sup>3</sup> N.C. Lopes,<sup>9</sup> O. Lundh,<sup>7</sup> Y. Ma,<sup>2,§</sup> S.J. Rose,<sup>1</sup> C. Spindloe,<sup>4</sup> D.R. Symes,<sup>4</sup> M. Šmíd,<sup>5</sup> A.G.R. Thomas,<sup>8,2</sup> R. Watt,<sup>1</sup> and S.P.D. Mangles<sup>1</sup>

<sup>1</sup>The John Adams Institute for Accelerator Science, Imperial College London, London, SW7 2AZ, UK

<sup>2</sup>Physics Department, Lancaster University, Lancaster LA1 4YB, United Kingdom

<sup>3</sup>Lawrence Livermore National Laboratory (LLNL), Livermore, California 94550, USA

<sup>4</sup>Central Laser Facility, STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, UK

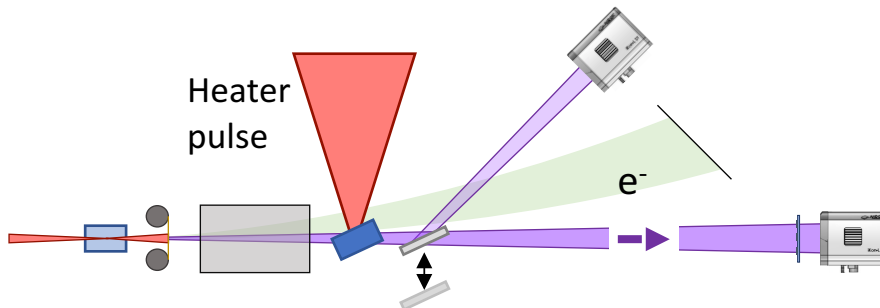
<sup>5</sup>Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstrasse 400, 01328 Dresden, Germany

<sup>6</sup>Institute of Physics of the ASCR, Na Slovance 1999/2, 182 21 Prague, Czech Republic

<sup>7</sup>Department of Physics, Lund University, P.O. Box 118, S-22100, Lund, Sweden

<sup>8</sup>Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, Michigan 48109-2099, USA

<sup>9</sup>GoLP/Instituto de Plasmas e Fusão Nuclear, Instituto Superior Técnico, U.L., Lisboa 1049-001, Portugal

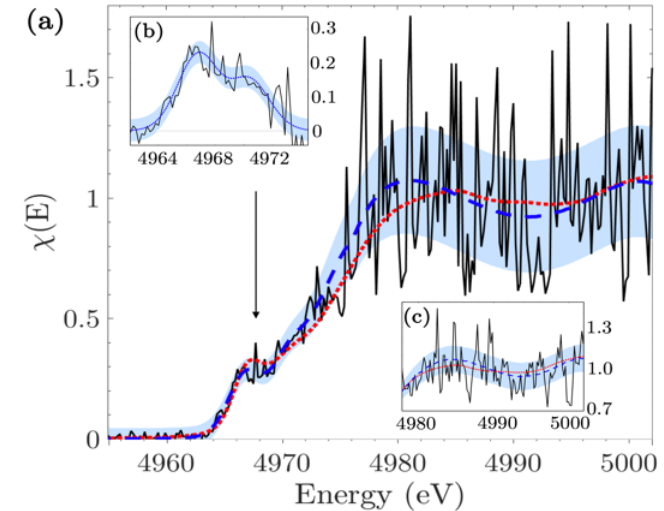


Single-shot absorption measurements have been performed using the multi-keV X-rays generated by a laser wakefield accelerator. A 200 TW laser was used to drive a laser wakefield accelerator in a mode which produced broadband electron beams with a maximum energy above 1 GeV and a broad divergence of  $\approx 15$  milliradians FWHM. Betatron oscillations of these electrons generated  $1.2 \pm 0.2 \times 10^6$  photons/eV in the 5 keV region, with a signal-to-noise ratio of approximately 300:1. This was sufficient to allow high-resolution XANES measurements at the K-edge of a titanium sample in a single shot. We demonstrate that this source is capable of single-shot, simultaneous measurements of both the electron and ion distributions in matter heated to eV temperatures by comparison with DFT simulations. The unique combination of a high-flux, large bandwidth, few femtosecond duration X-ray pulse synchronised to a high-power laser will enable key advances in the study of ultra-fast energetic processes such as electron-ion equilibration.



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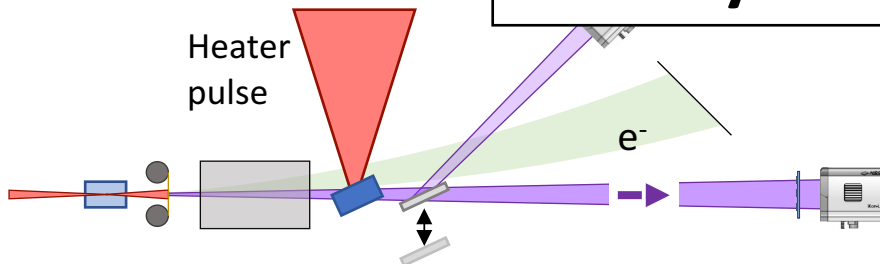


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<sup>2</sup>Lancaster University, Lancaster LA1 4YB, United Kingdom  
<sup>3</sup>Livermore National Laboratory (LLNL), Livermore, California 94550, USA  
<sup>4</sup>STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, UK  
<sup>5</sup>Leibniz-Zentrum für Plasma- und Raumforschung, Bautzner Landstrasse 400, 01328 Dresden, Germany  
<sup>6</sup>STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, UK  
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Thank you for listening



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## Acknowledgements

Stuart Mangles<sup>1</sup>, Steven Rose<sup>1</sup>, Rory Baggott<sup>1</sup>, Robbie Watt<sup>1</sup>, Jason Cole<sup>1</sup>, Elias Gerstmayr<sup>1</sup>,  
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<sup>2</sup> Lancaster University, U.K.

<sup>3</sup> University of Michigan, USA

<sup>4</sup> Central laser Facility, U.K.

<sup>5</sup> Lund University, Sweden

<sup>6</sup> ELI Beamlines, Czech Republic

<sup>7</sup> Lawrence Livermore National Laboratory, USA



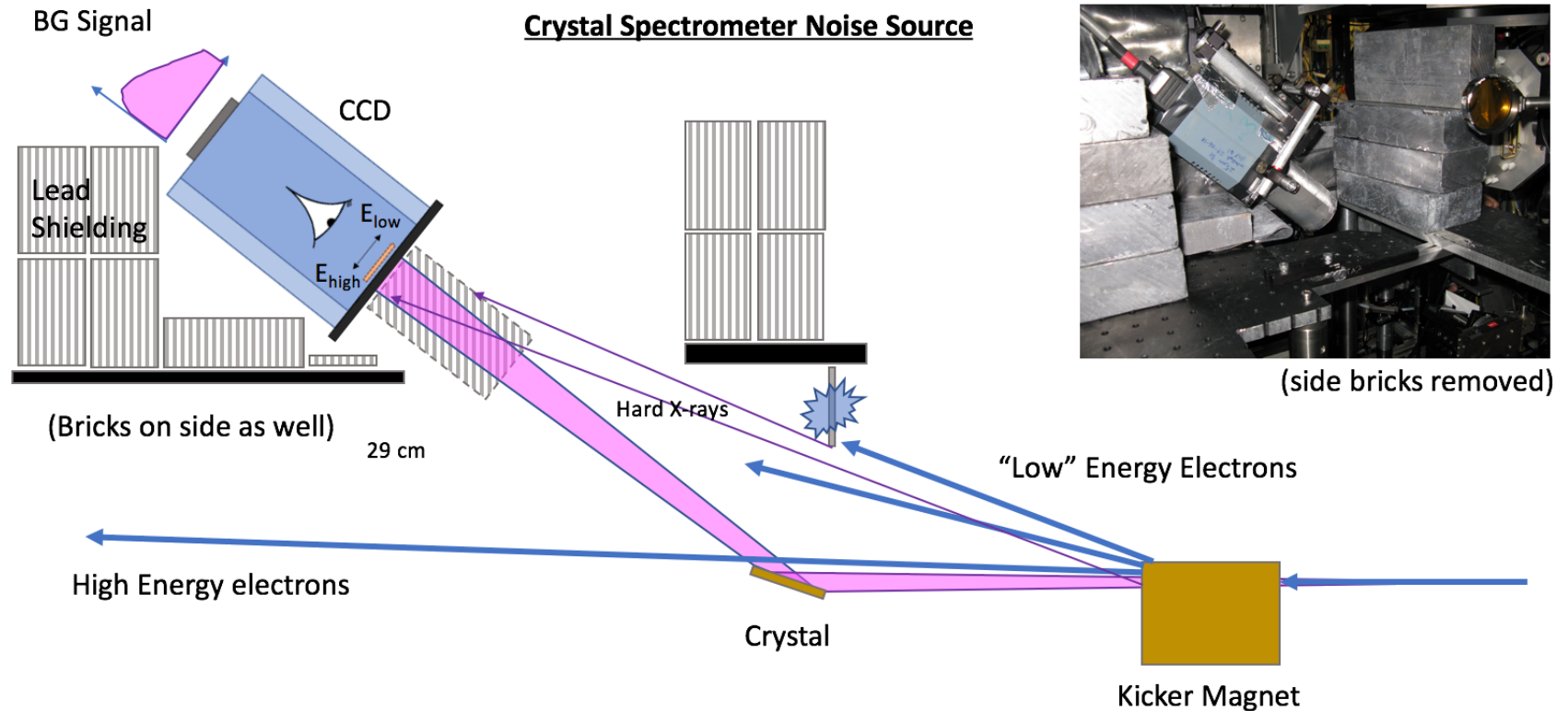
Part funded by  
ERC TeXMex Grant  
(Ref: 682399)





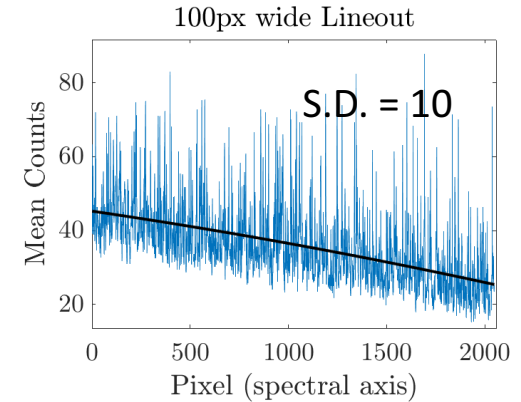
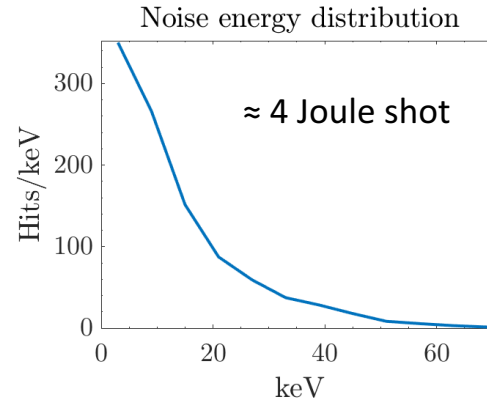
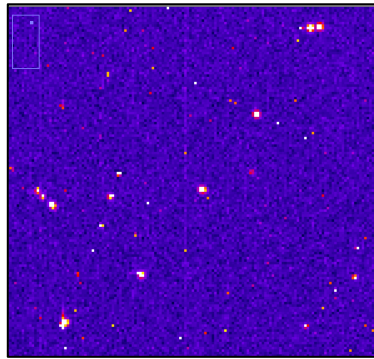
## Error/Noise contributions

- Hard photon background from electron dump. Dominates. Can be significantly reduced by sweeping electrons in other direction and improving shielding.
- Reference map errors. Give larger scale fluctuations. Can be reduced by a full crystal characterisation.
- Inherent source fluctuations.

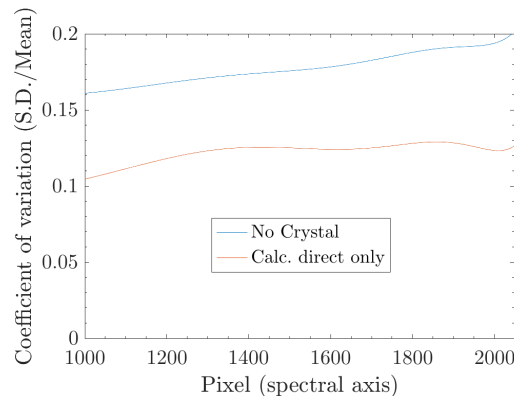
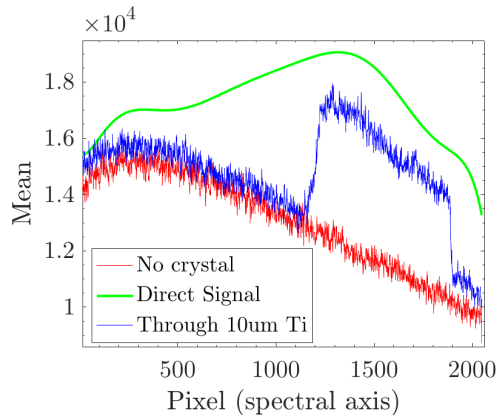


## ▷ Crystal spectrometer noise issue

Looking at low power shots with "single hit" noise.  
Run charge recombination routine and gauge spectrum.



On full power shots, this becomes  $\approx \times 200$



Can improve

- Shielding
- Electron dumping
- Detector geometry  
(including curved crystal)

To reduce noise and move  
closer to single shot  
measurements.

## BACKGROUND NOISE

We assess the background level on the crystal spectrometer by summing the CCD counts on shots where the crystal is moved out of the direct path of the X-ray beam, and hence no signal is being reflected towards the detector. For each of these shots we also sum the total electron charge simultaneously detected on the electron spectrometer. Fig. 3 (a) depicts a linear correlation for the number of CCD counts as a function of electron charge.

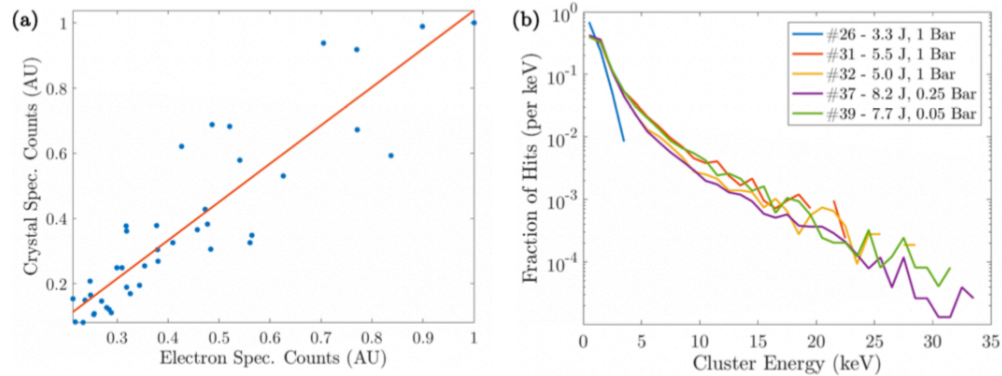


FIG. 3: (a) Crystal spectrometer CCD counts as a function of measured electron charge for shots with no crystal in place. (b) Single-hit cluster energies for various background shots at low background flux.

In an attempt to further characterise this background signal we perform a single-hit cluster analysis of the individual CCD strikes on shots with a low enough level of flux (thus avoiding strike pile-up). Less than 3% of CCD pixels registered a value above threshold for all shots used in the single-hit analysis. A cluster combining algorithm was used to calculate the total freed energy for each individual identifiable strike on the CCD. Fig. 3 (b) depicts the number of strikes as a function of total contained energy in each cluster. For the shots depicted, various plasma densities and laser energies were used, corresponding to varying levels of electron charge being generated from the LWFA. Shot 26 in the blue has no detectable electron charge driven, and represents the dark current of the CCD. All shots apart from 26 show a broadband spectra of cluster energies tailing out to 30 keV (which is also where the quantum efficiency of the CCD falls off).

These results indicate that in the current configuration the background noise is being produced by the accelerated electrons interacting with the target chamber and creating secondary noise sources. Importantly, it should therefore be possible to significantly reduce the background with improved shielding and appropriate electron beam dumping, away from the CCD.

