Opportunities for X-ray pump – X-ray probe attosecond science

Dr. Laurent Mercadier
SCS

European XFEL / DESY Users' meeting 2025
Satellite meeting: Status and development of SCS Instrument
Schenefeld
Thursday, 23rd January 2025

Outlook

- Introduction
- Transient XAS of warm dense matter
- Perspectives for all X-ray pump-probe spectroscopy and attosecond pulse characterization at SCS
- Conclusion

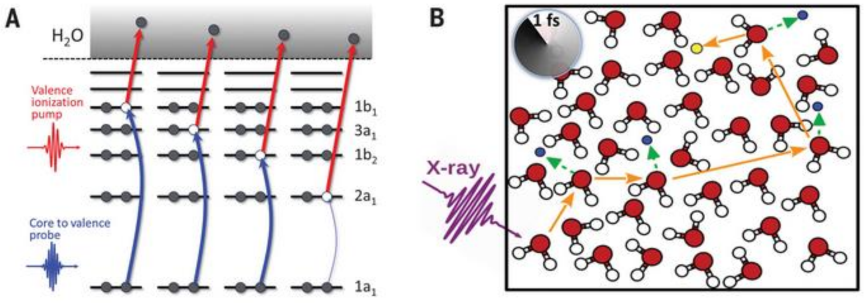
Attosecond, all X-ray science holds many promises

- X-ray attosecond pulses by XFELs

  Nat. Photonics **18**, 691 (2024)
  Nat. Photonics **18**, 698 (2024)
  Nat. Photonics **14**, 30 (2020)
  Appl. Sci. **10**, 2728 (2020)
  Opt. Express **26**, 4531 (2018)

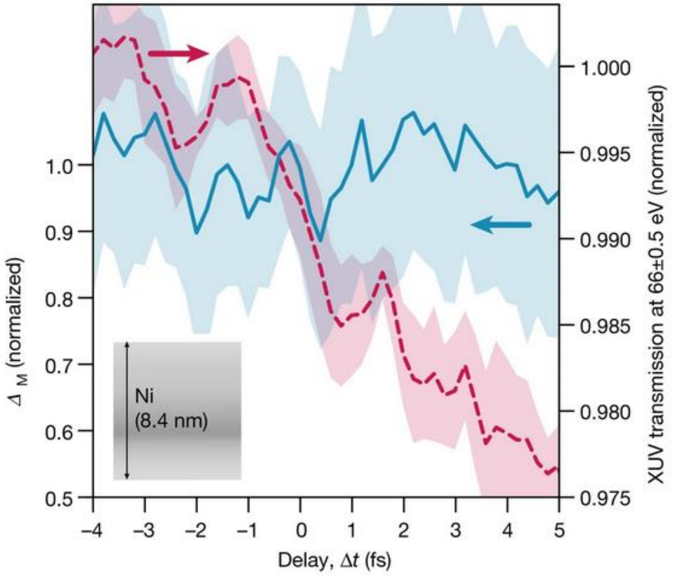
- Solvation dynamics and water science

  Science **383**, 1118 (2024)



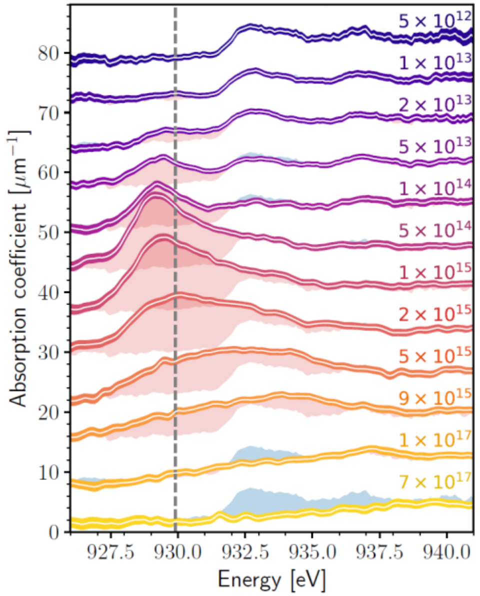
- Magnetism

  Nature **571**, 240 (2019)



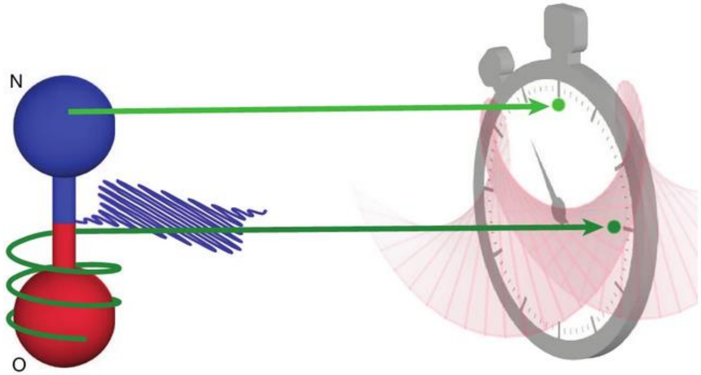
- Nonlinear X-ray / warm dense matter

  PNAS **113**, 1492 (2016)
  Nature Physics **20**, 1564 (2024)



- AMO

  Science ***375,*** *285* (2022)
  Nature **632**, 762 (2024)



Warm Dense Matter (WDM) and XFELs

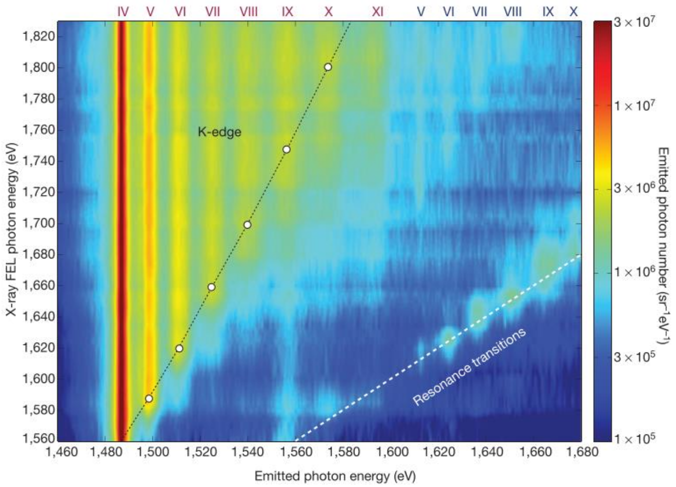
- First demonstration of XFEL-created WDM (FLASH):

Turning solid aluminium transparent by intense soft X-ray photoionization

Bob Nagler et al.

Nature Physics **5**, 693–696 (2009) | Cite this article

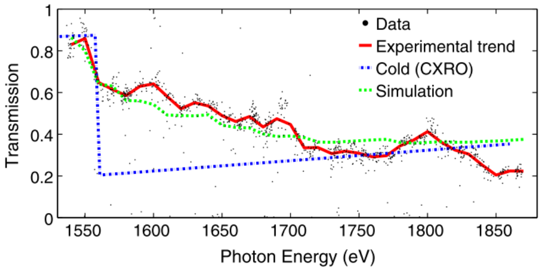
- LCLS experiments (J. Wark et al.):
  X-ray Emission Spectroscopy of WDM Aluminum



Vinko et al., *Nature* **59** (2012)
Ciricosta et al., PRL **109** (2012)
many more…

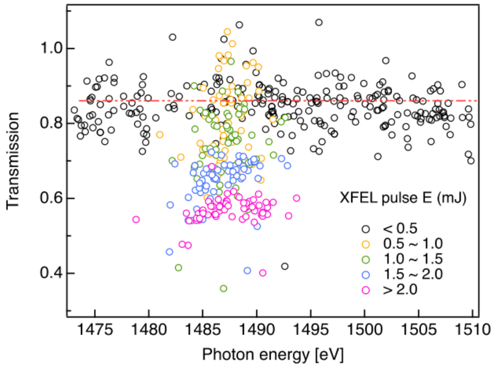
- Measurement of Ionization Potential Depression
- Estimation of collisional rates
- Development of theoretical tools

- Saturable Absorption (SA)
  - Depletion of the absorbing state by the front of the pulse causes absorption to saturate → **Decrease of absorption with intensity**



Rackstraw et al., PRL **114** (2015)
Yoneda et al., *Nat. Commun.* **5** (2014)

- Reverse Saturable Absorption (RSA)
  - Excited state created by the front of the pulse is more absorbing than the ground state → **Increase of absorption with intensity**



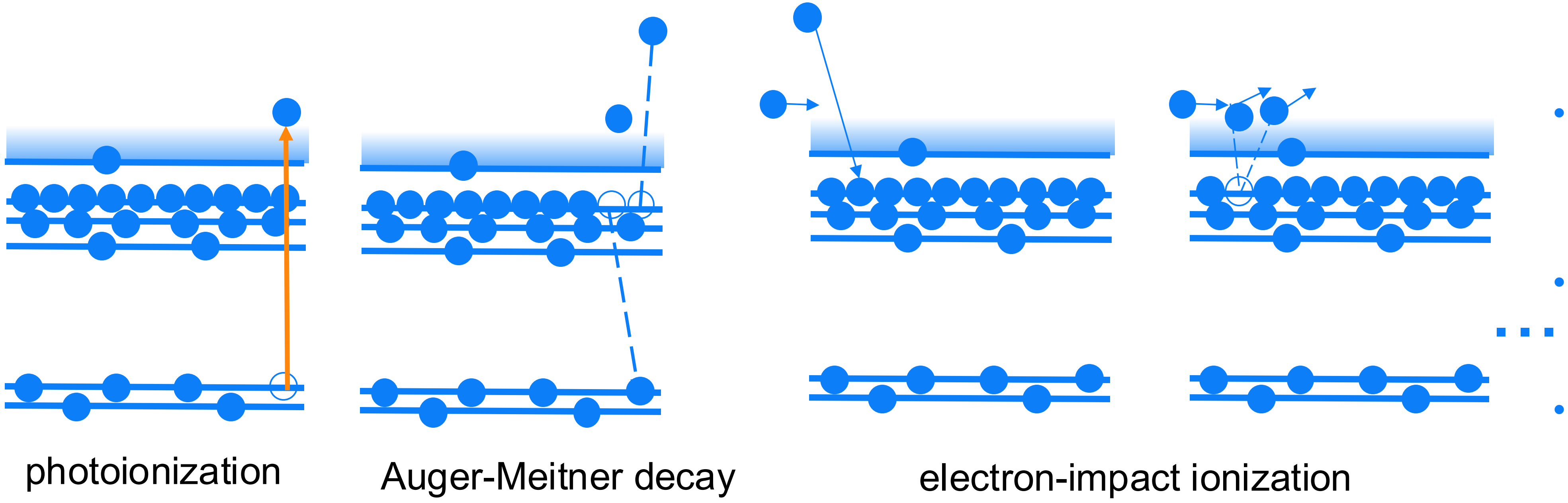
Cho et al., PRL **119** (2017)

- Efficient, isochoric X-ray excitation
  - Core shell ionization + Auger decay
  - Impact ionization
- Non-linear effects transferred to the X-ray domain

XFEL as a tool to create WDM

Secondary ionization processes following inner-shell ionization

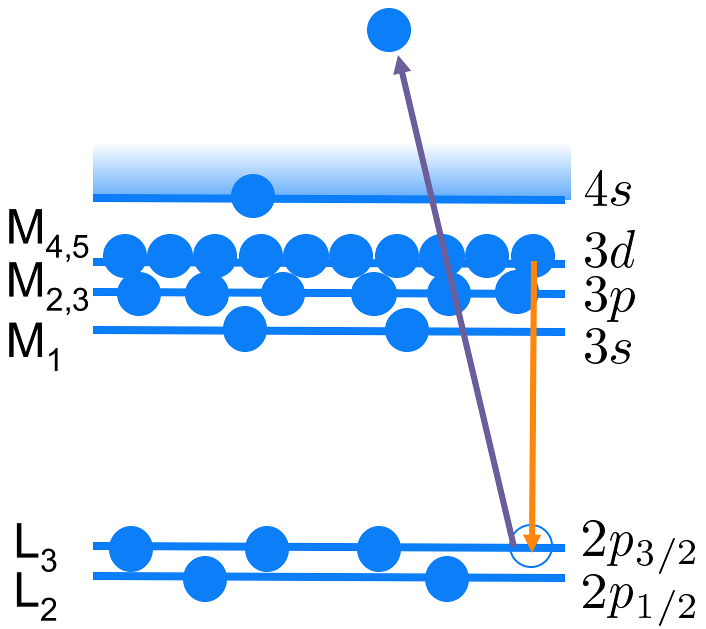
Build-up of high charge states and hot continuum electrons



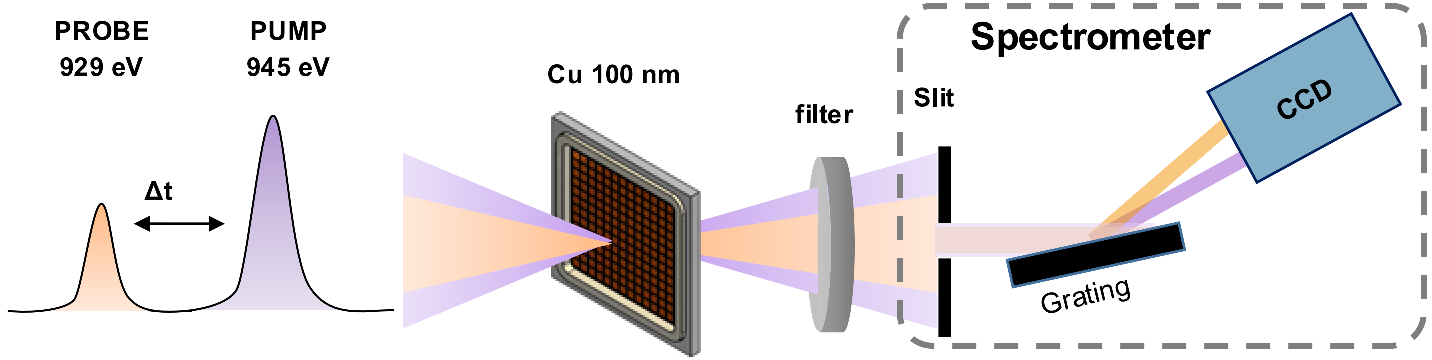
- elastic and inelastic collisions of electrons and ions,
- recombination processes,
- short range electron–electron scattering

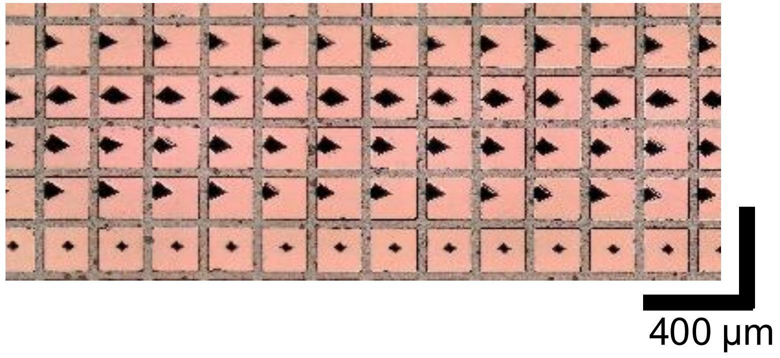
- Highly excited exotic state of matter
  - Role of non-thermal electrons?
  - Ionization dynamics?
  - Modelling: solid state vs plasma state

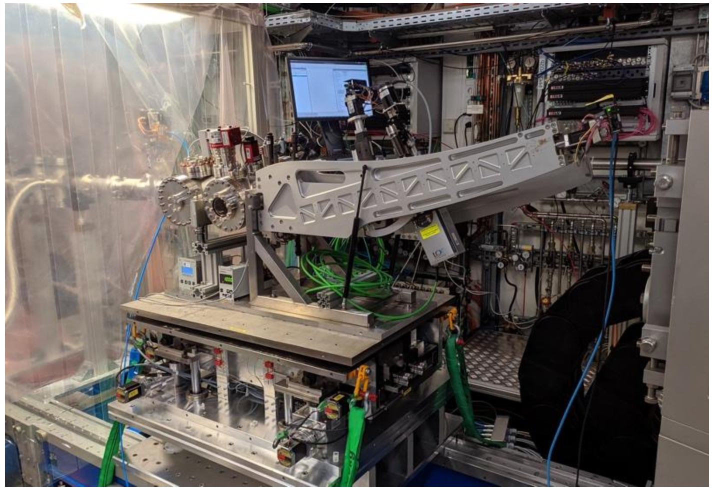
Measurement scheme



- Cu L_3-edge: 932 eV
- Cu L_2-edge: 952 eV

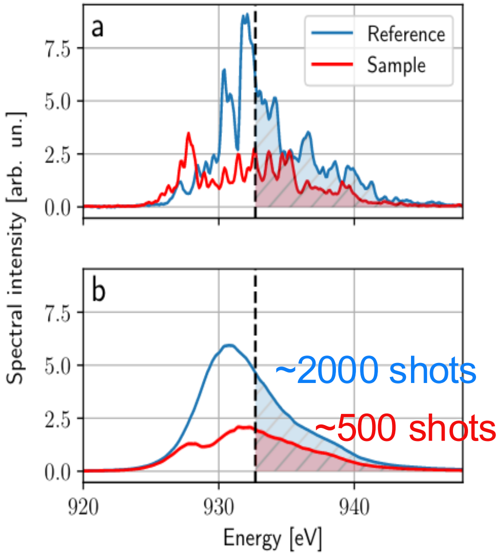


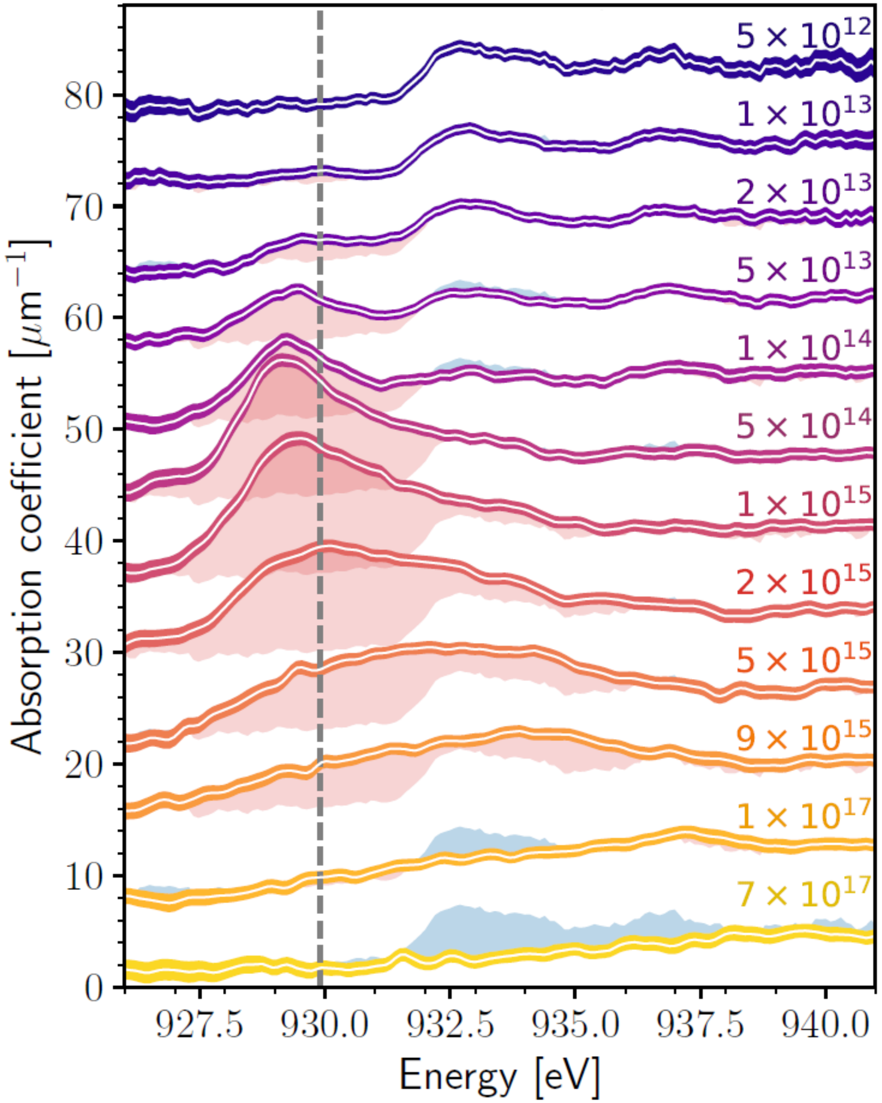




- 4 µm FWHM beam size
- 15 fs
- Intensity range: 10^{12} to 7×10^{17} W cm^{-2}
- 100 nm Cu film on Ni mesh
- Irradiation at 10 Hz

Transient XAS of Warm Dense Copper

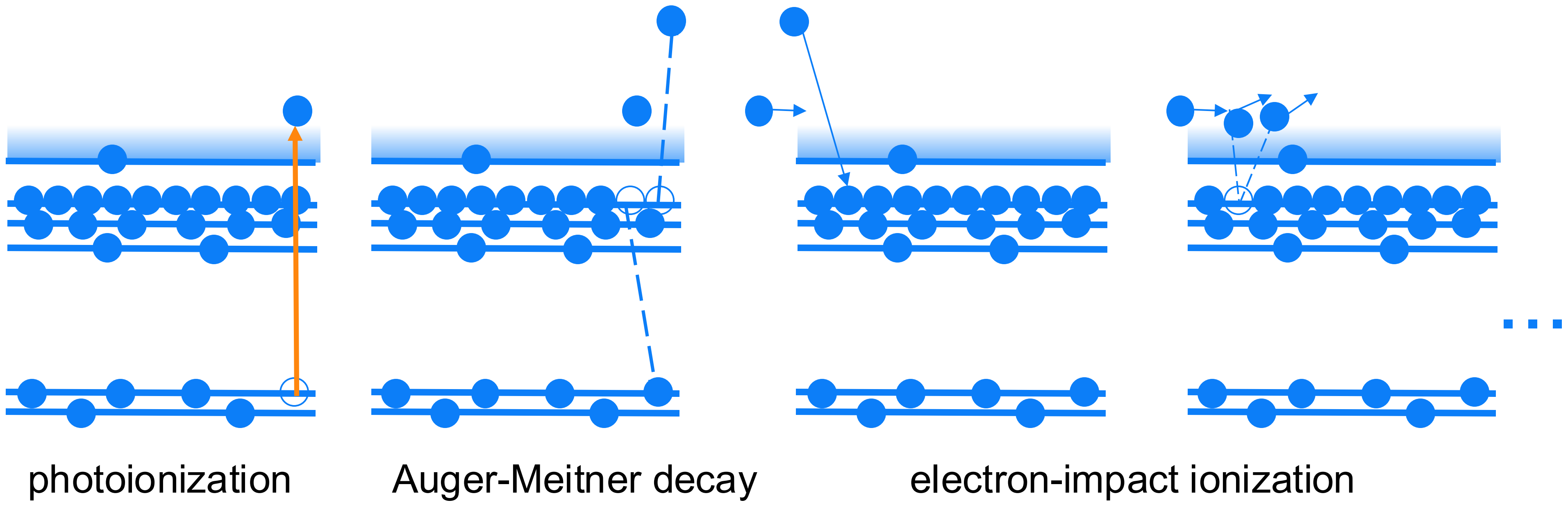




- Drastic spectral modifications within the 15 fs pulse duration
- Reverse Saturable Absorption: pre-edge peak
  - Creation of 3d vacancies
  - Opening of absorption channel $2p_{3/2} \rightarrow 3d$: RSA
  - Red-shift of the peak: signature of shift of the 3d band
- Flattening and blue-shift of the pre-edge peak
  - Opening of 2p→3d absorption channels of higher charge states
- Saturable Absorption at 10^{17} W cm^{-2}

Mercadier et al., *Nature Physics* **20**, 1564 (2024)

Modeling x-ray heated warm dense copper with Boltzmann equations



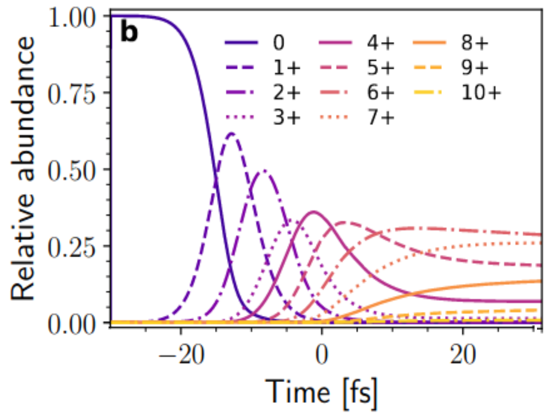
Prof. Dr. Beata Ziaja

Dr. Michal Stransky

- elastic and inelastic collisions of electrons and ions,
- recombination processes,
- short range electron–electron scattering.

- Follow the evolution of true atomistic configurations resulting from photoionization, photoexcitation, Auger decay, collisional processes (impact ionization, recombination, short range e-e interaction, …)
- Simplified with the “predominant excitation and relaxation path” (PERP)
- Well suited to capture non-equilibrium dynamics in the early stages of WDM formation

- Outputs
  - Time-resolved occupation of atomic/ionic configurations
  - Time-resolved free electron energy distribution
  - Kinetic temperature of free electrons
  - Time-resolved ionization degree



B. Ziaja et al., Eur. Phys. J. D 40, 465 (2006); Phys. Rev. E 93, 053210 (2016)

Modeling XFEL-created WDM: Boltzmann equations + FEFF10

- **Combining plasma physics and solid-state physics descriptions**

John Rehr

Josh Kas

Boltzmann kinetic equations solver

non-equilibrium electronic distribution

Relative abundances of various atomic configurations

3d occupation vs time and intensity

Andrei Benediktovitch

T_{eff}

Account of environment

FEFF10 real-space multiple scattering code

Band structure and 3d occupation

- Broadening effects
- Edge of ionized atoms position
- …

Absorption cross-section

Space and time averaging

XANES spectra

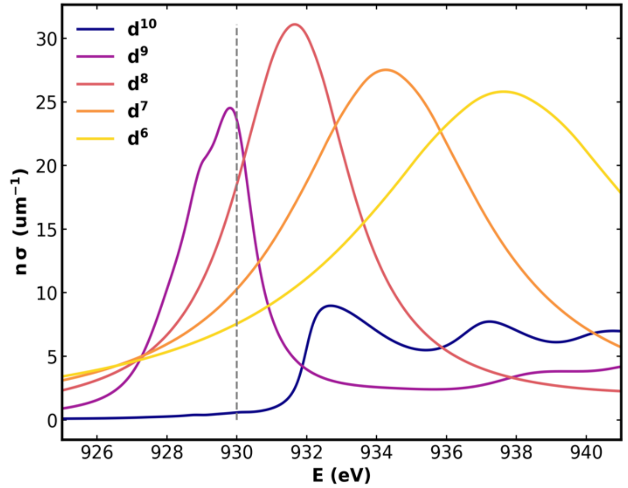
J.J. Rehr, R.C. Albers, Rev. Mod. Phys. 72, 621 (2000)

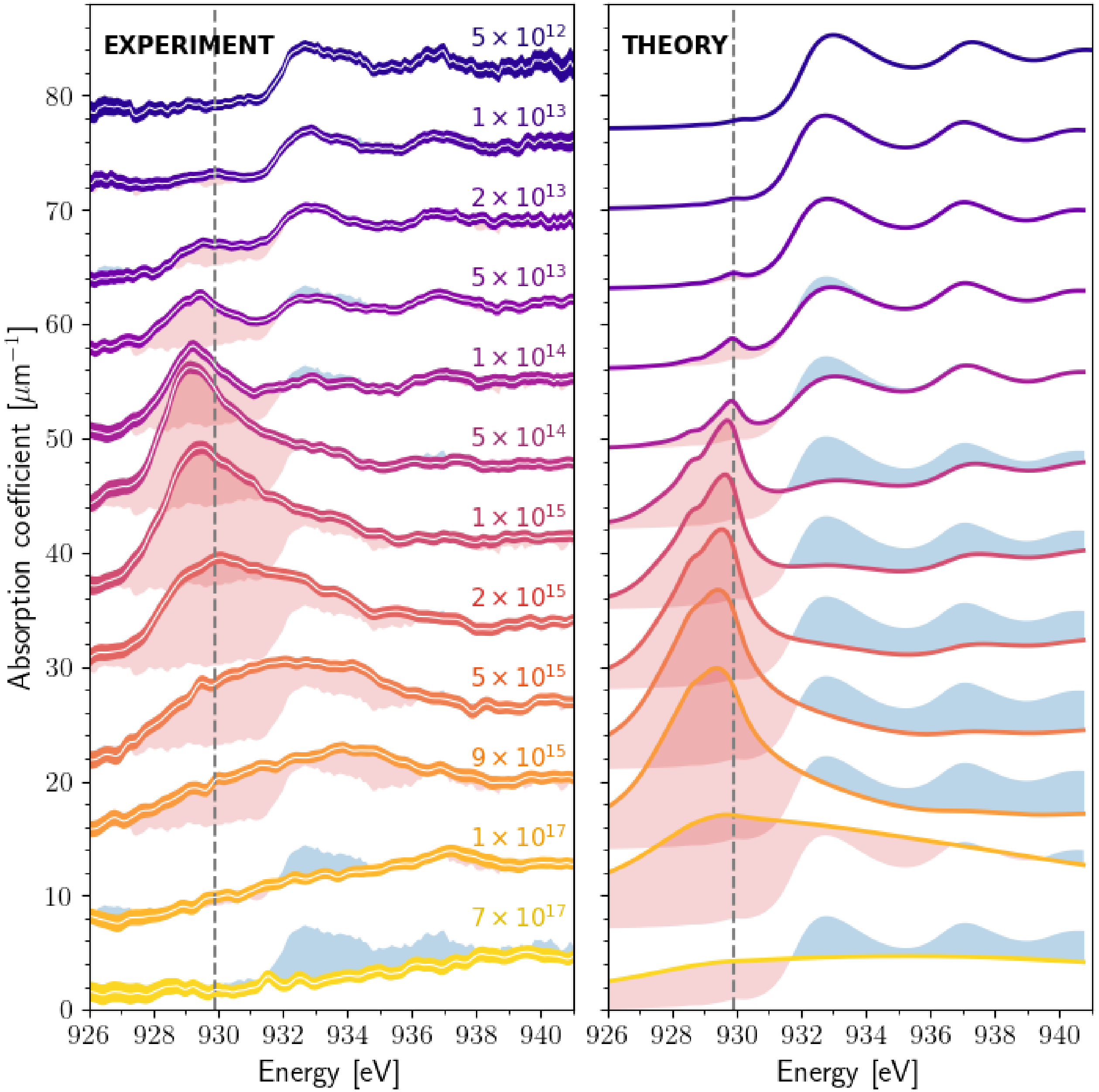
T.S. Tan, J.J. Kas, J.J. Rehr, PRB 104, 035144 (2021)

T.S. Tan, J.J. Kas, et al, PRB 107, 115122 (2023)

Modeling: comparison with experiment

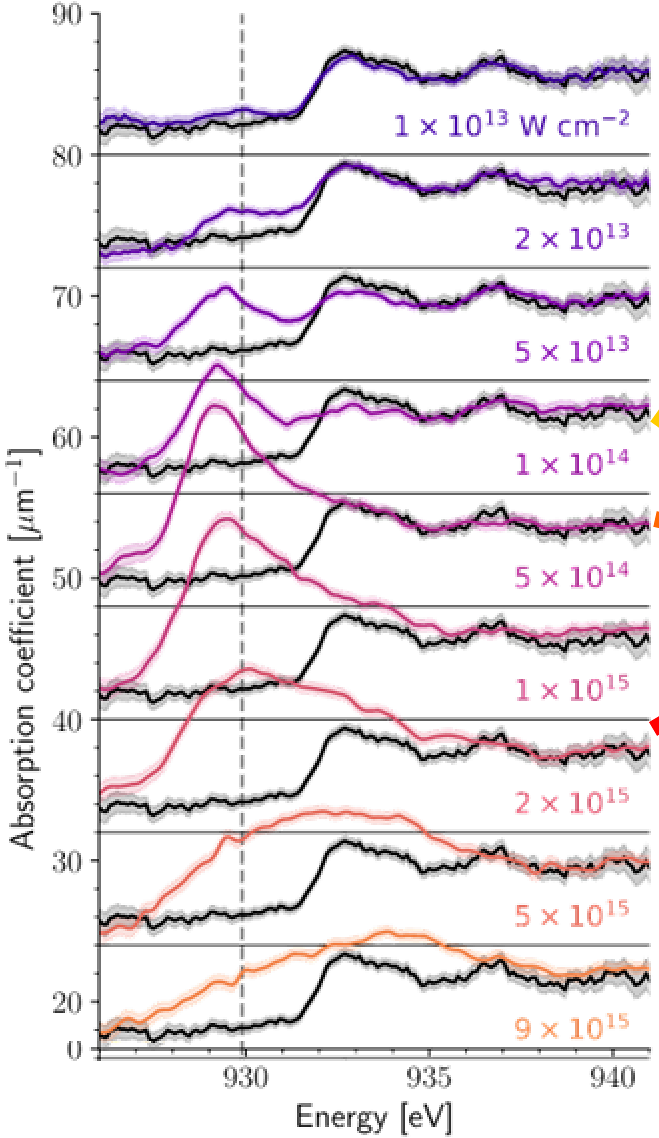
- Qualitative agreement
  - Transition from RSA to SA
  - Red-shift of pre-edge peak at low to moderate intensities: movement of the 3d band
  - Blue-shift and SA at higher intensities: deeper holes in the d-band

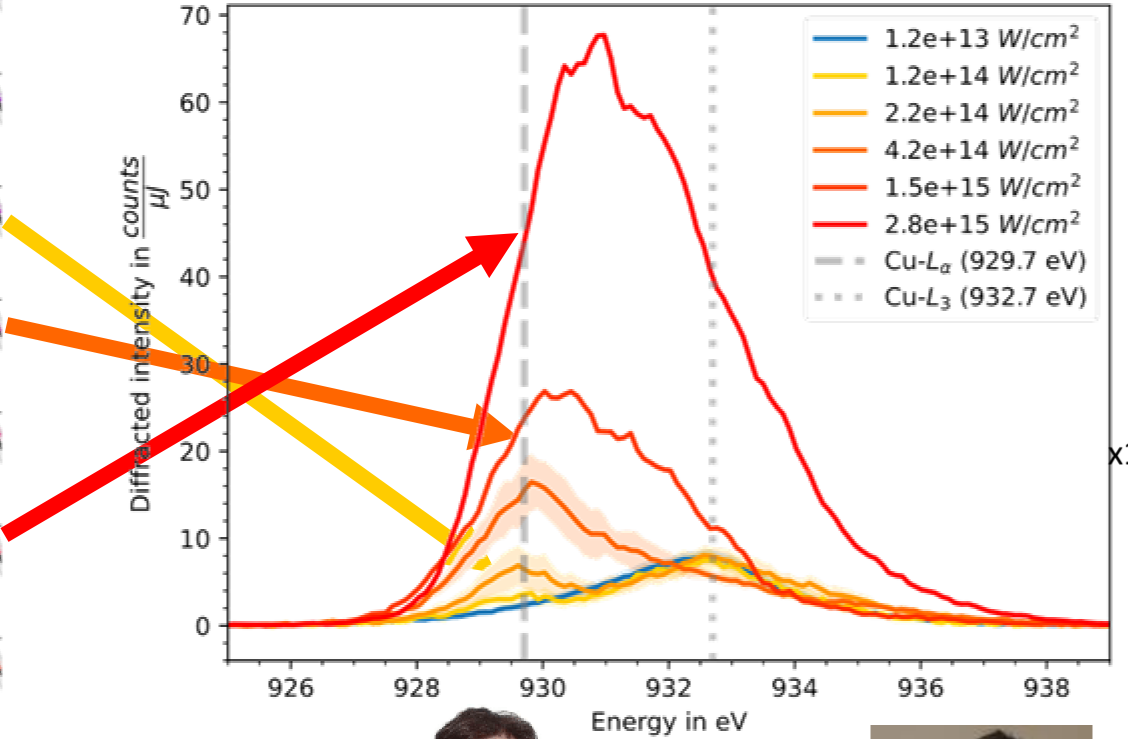


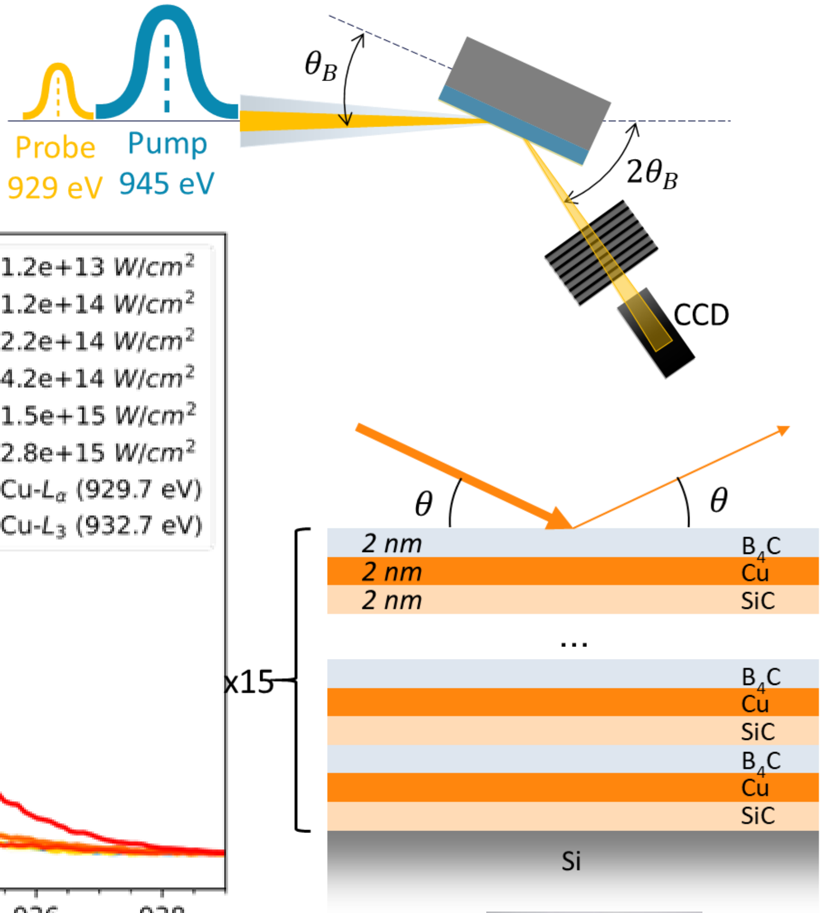


Mercadier et al., *Nature Physics* **20**, 1564 (2024)

Reverse saturable absorption and enhanced scattering







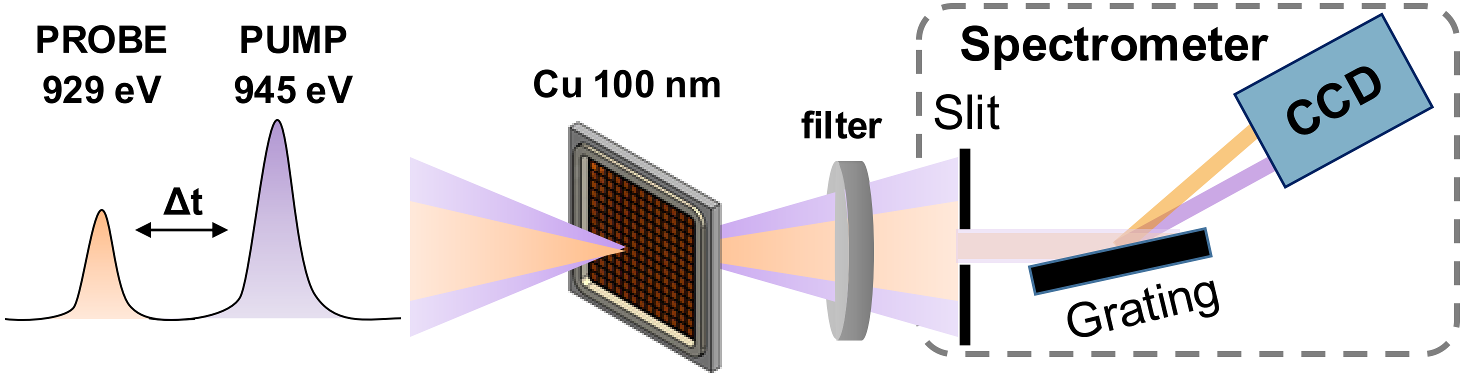

Andrei Benediktovitch


Daniele Ronchetti

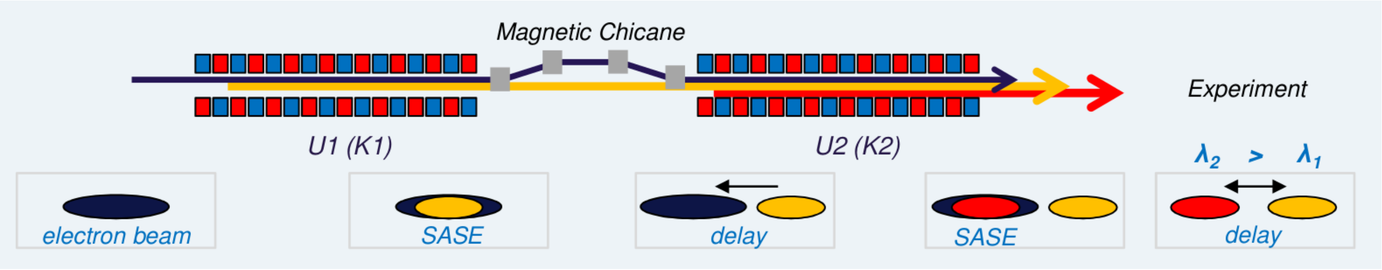

Nina Rohringer

All of this happened within the pulse duration (15 fs)…

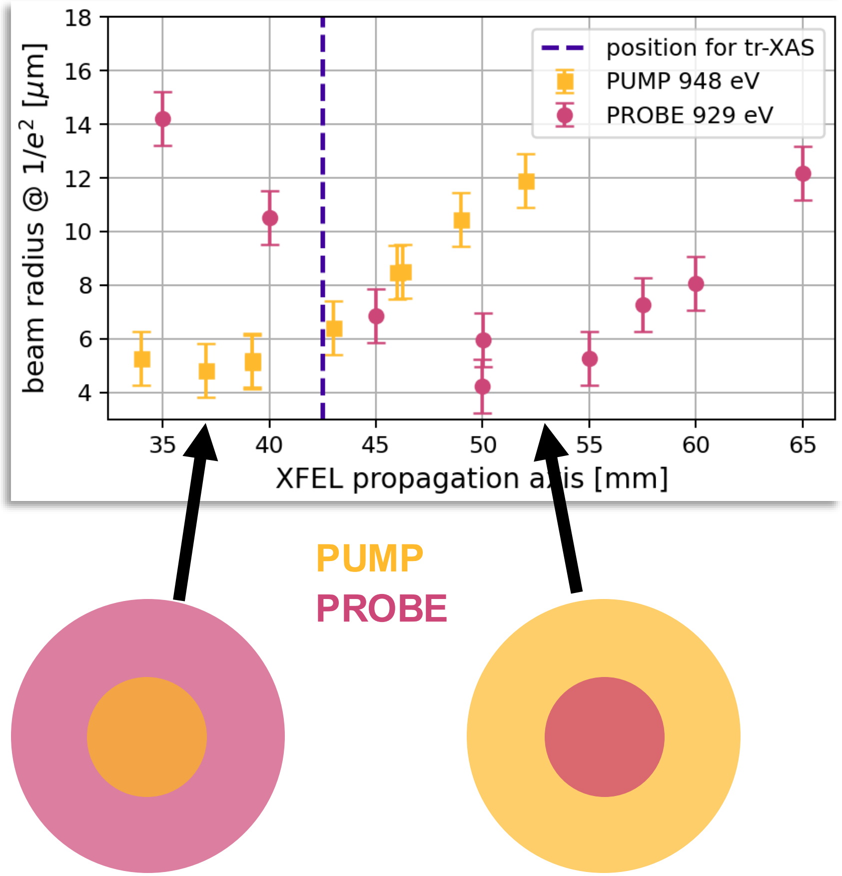
Let’s look at the dynamics after XFEL-heating



Two colors operation



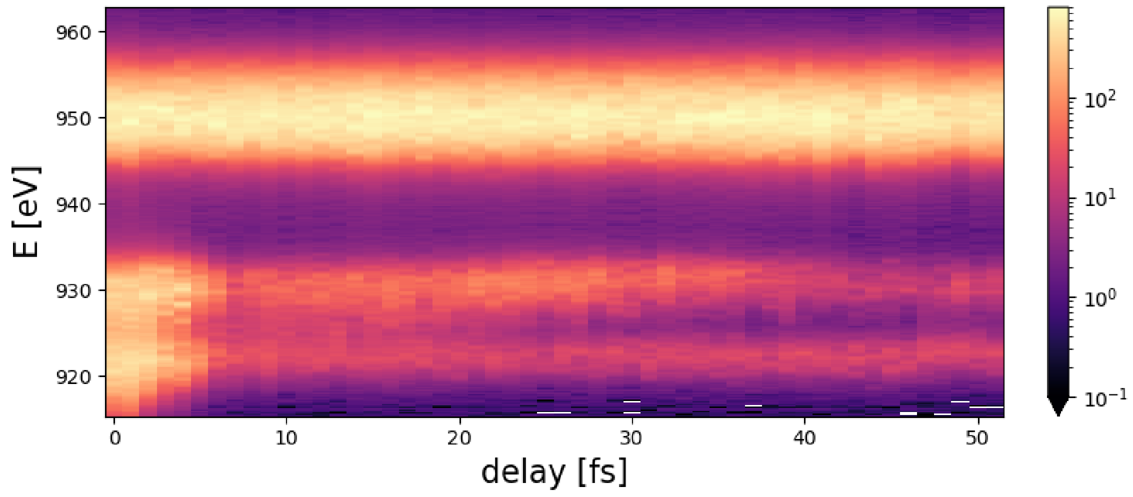
S. Serkez et al., Appl. Sci. 10, 2728 (2020)



- **Pulse duration:** 10-15 fs (spectral analysis of g_2 correlation of SASE spikes)
- **Beam size** measurement with knife-edge
- two foci separated by 12 mm because of different source position

Delay scan: cross-talk between the two colors

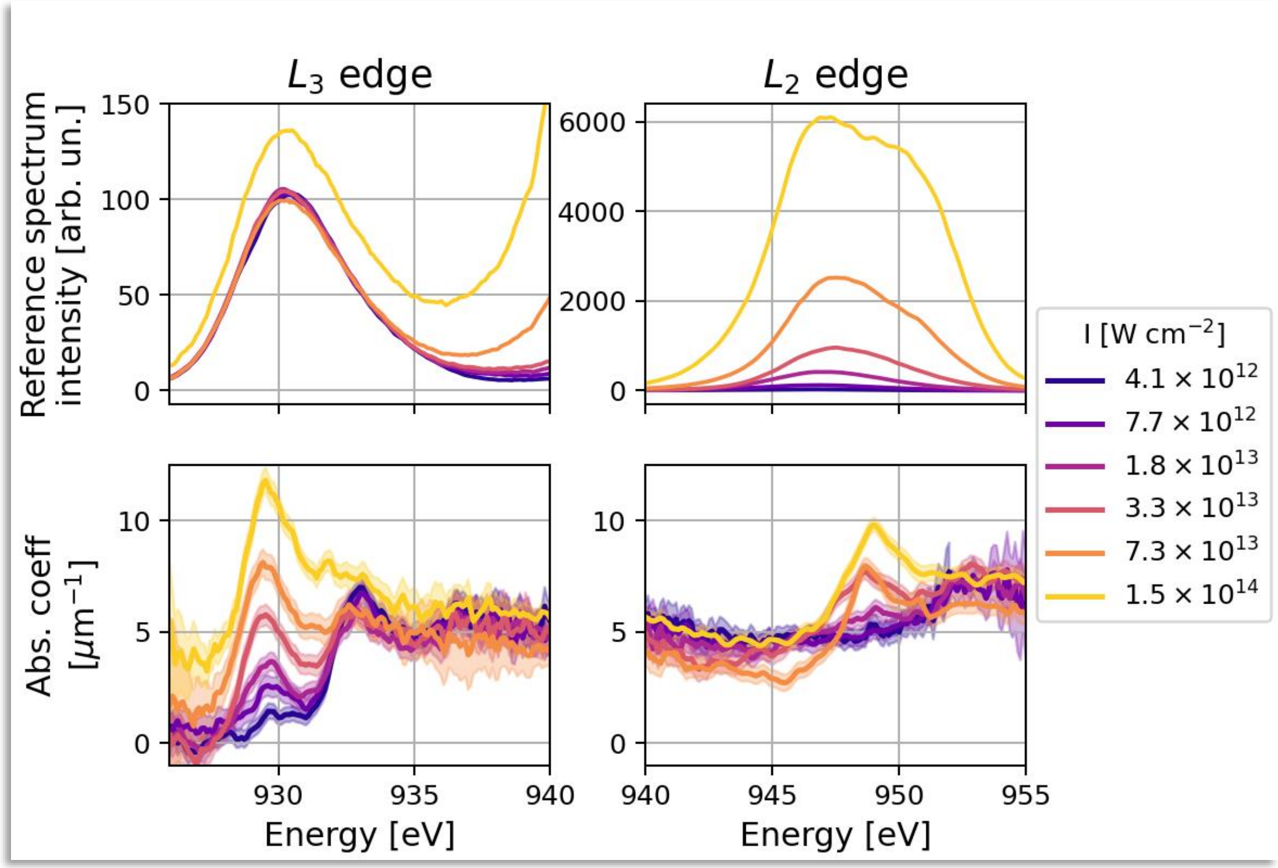
- Intensity and spectral shape of probe changes with delay
- No clear definition of time zero
- No access to negative time delays
- Influence on pulse duration / chirp?



- Optical chicane will allow negative delays and mitigate the cross-talk
- Large energy separation can help minimize the cross-talk

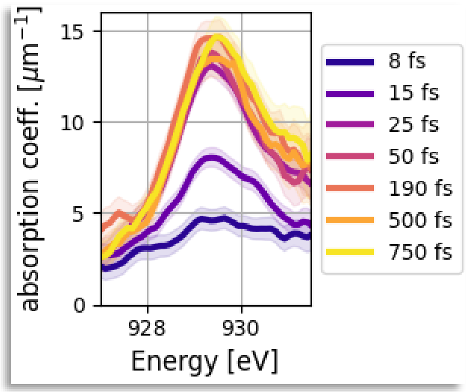
Two colors: pump intensity scan at time overlap

- Ensure “cold” probe
- Adjust ration between pump and probe by opening/closing cells
- Absorption feature appears at both edges
- Comparable to single color but more efficient pumping

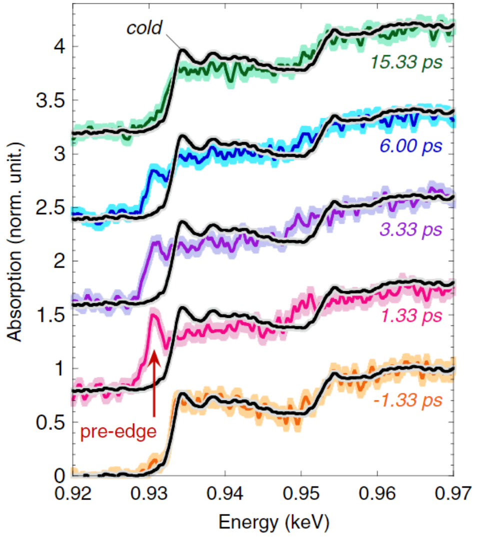


Two colors: delay scan at fixed pump intensity

- Appearance of pre-edge feature within the pulse duration
- Presence of 3d holes up to 750 fs
- Improved time-resolution

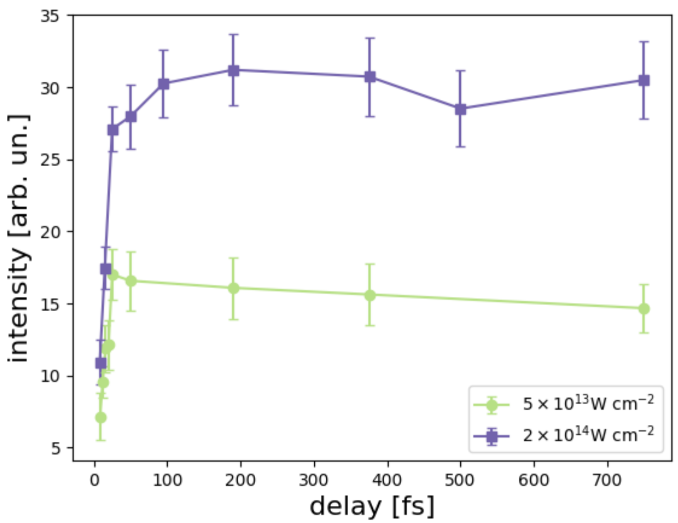


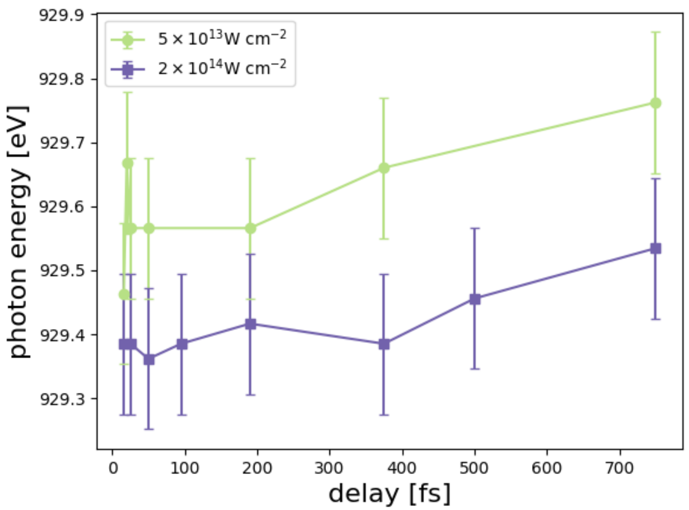
Optical excitation



Jourdain et al., Phys. Rev. B **97**, 075148 (2018)

- Smaller probed energy window
- Small redshift with time
  - Cooling down
- Modelling in progress…



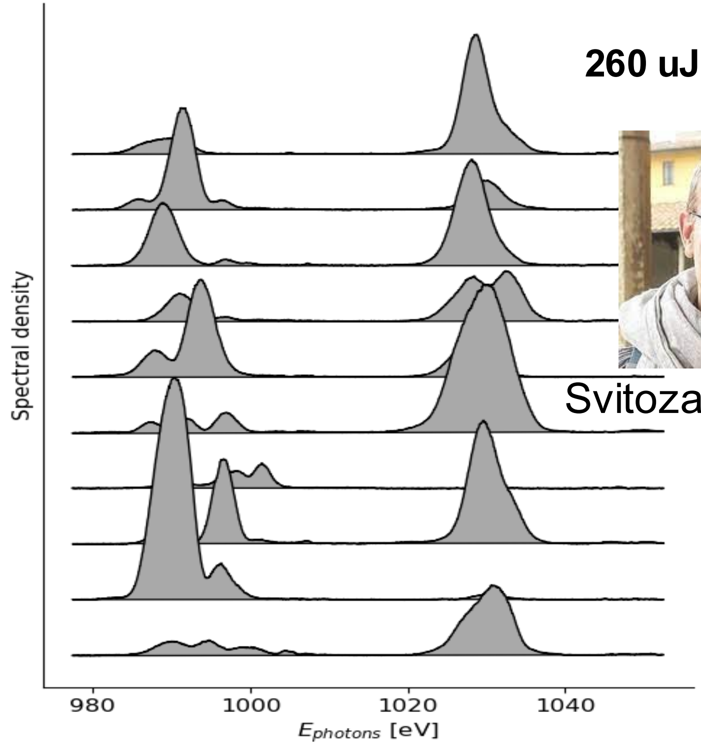


Outlook

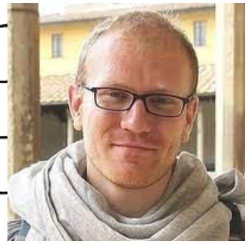
- Introduction
- Transient XAS of warm dense matter
- **Perspectives for all X-ray pump-probe spectroscopy and attosecond pulse characterization at SCS**
- Conclusion

Current challenges for attosecond transient XAS at SCS: X-ray delivery

- Setup of two-colors is a special mode
  - Requires long tuning time, some during the beamtime
  - Tests ahead are difficult to plan (machine in different conditions than for the beamtime)
  - Pump on / off has strong influence on probe
- Cross-talk between the two colors around time overlap
- Where is time zero?
  - Magnetic chicane does not allow for negative delays
  - Optical chicane to be installed in fall 2025
- Pulse duration? Chirp?
  - Spike analysis of high resolution spectra (invasive and not pulse-resolved)



260 uJ



Svitozar Serkez

Current challenges for attosecond transient XAS at SCS: Beam properties

- Spectrum normalization
  - BOZ only applicable to monochromatic and weak beam
  - Sequential recording of sample spectra + reference spectra does not capture machine fluctuations
  - Development of self-referencing grating

- No SCS spectrometer
  - Viking spectrometer (Uppsala University, J.-E. Rubensson)
    - ► strong background, source unknown
    - ► Resolution limited to ~2000
  - SCS spectrometer
    - ► Improved resolution
    - ► Expected delivery fall 2026

- Micrometer beam size measurement
  - Limits of knife-edge
  - Imprints can only be analyzed a posteriori
  - Development of focus characterization with curved grating

Please contact SCS staff to plan a two-color experiment!

EuXFEL Facility Development Programme (2030): Enabling attosecond science at the European XFEL

PI: Gianluca Geloni

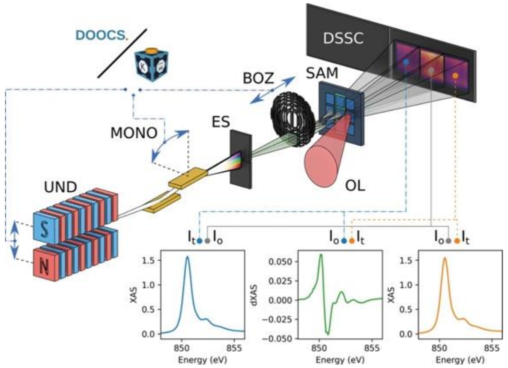
- Aim: deliver in a reliable, verified and usable way well characterized, high power attosecond pulses at high repetition rate

- Electron diagnostics: online, high resolution measurement of the longitudinal electron beam phase space

- Photon diagnostics:
  - Temporal characterization of attosecond pulses (XPD group)
  - Spectral characterization of attosecond pulses at SCS with forward spectrometer
    ► Self-referencing allows for single shot characterization
  - Reliable beam size characterization based on focus imaging by diffractive optics

- Test experiments at SCS
  - Transient XAS of WDM and magnetic materials
  - Liquid jets at a later stage

Improving normalization scheme

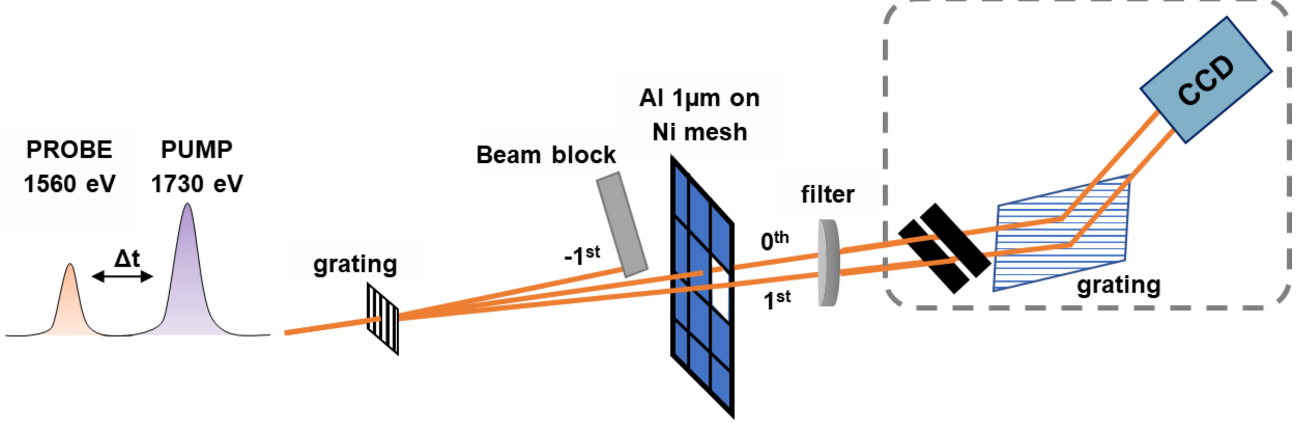


L. Le Guyader et al., J. Synchrotron Rad. ***30****, 284-300 (2023)*

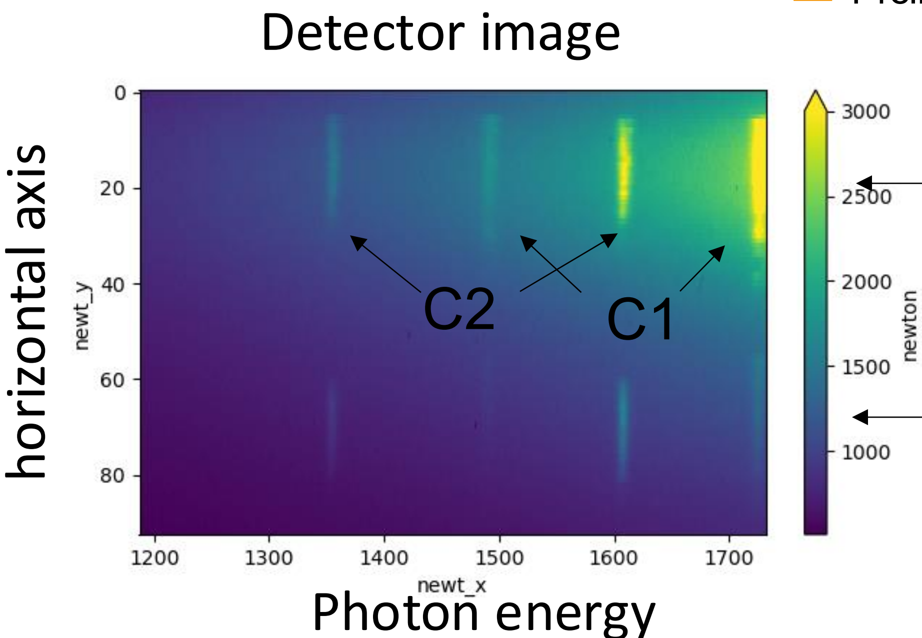
- Beam-splitting off-axis zone plate (BOZ)
- Photon shot-noise limit at MHz rep. rate with DSSC
- Limited to monochromatic beam
  - Stretched pulses (> 30 fs)
- Limited to moderate fluence

Sarlota Birnsteinova

First attempt with transmission grating



- Preliminary results: partially successful, fluence limited



0^{th} order on sample

1^{st} order through hole

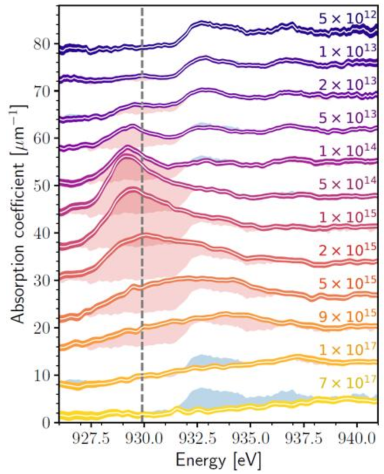
Devesh Chopra

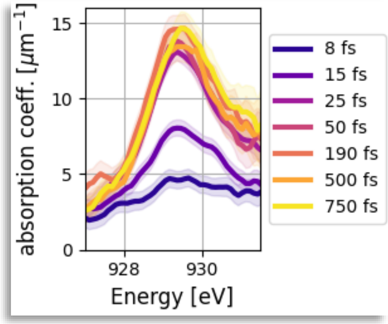
See Devesh's poster on WDM AI

Conclusions

- XAS setup to study XFEL-created WDM at the SCS instrument of EuXFEL
  - Single color: transition from RSA to SA at Cu L_3
  - Two colors: Long-lived excitation. Need of as pulses to follow ionization dynamics

- Moving towards all-x-ray attosecond XAS requires several improvements
  - EuXFEL strategy programme
    - ► X-ray pump / probe delivery, beam temporal properties diagnostics
  - At SCS
    - ► New spectrometer with increased resolution and reduced background
    - ► Self-referencing normalization scheme
    - ► Robust beam size characterization





Team

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Michal Stransky (EuXFEL)

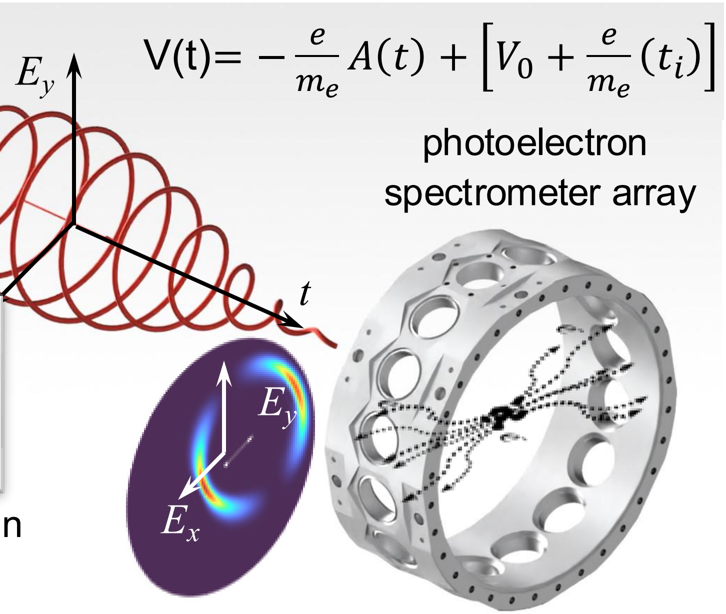
ETH zürich

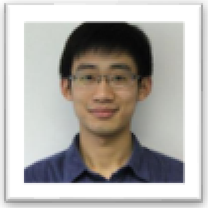
Giuseppe Fazio

John J. Rehr
Joshua Kas

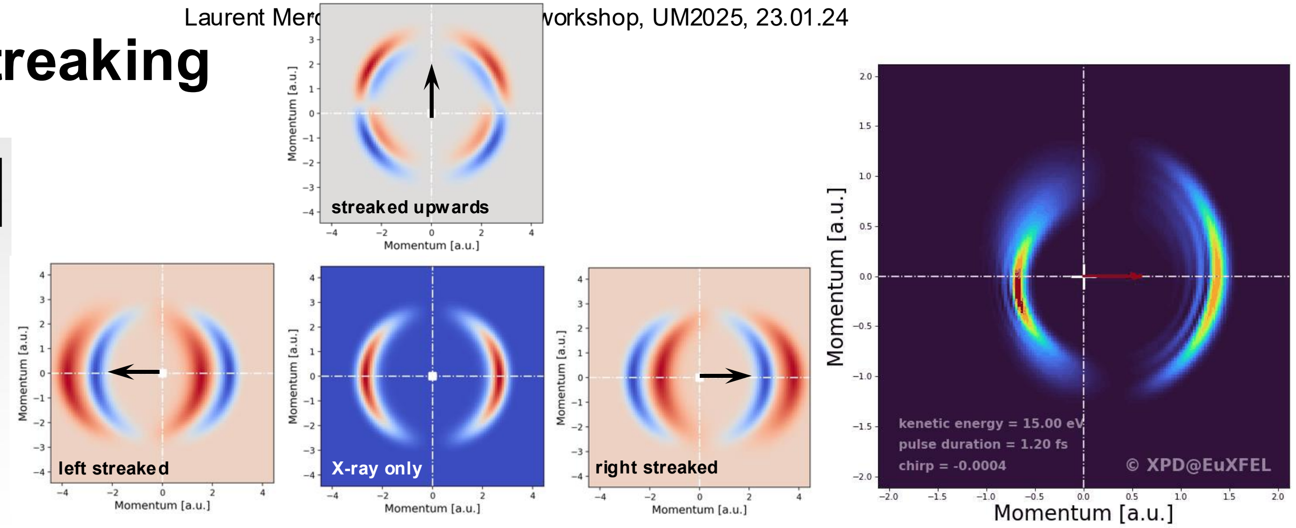
Thank you for your attention!

The photoelectron attoclock: Angular streaking

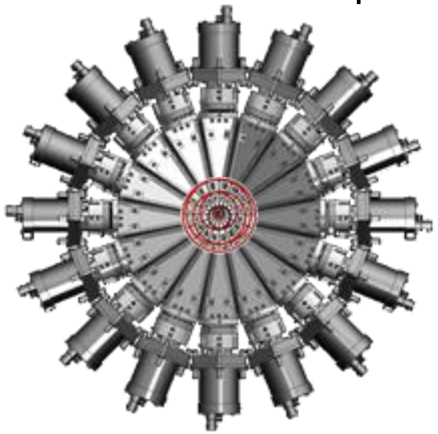



J. Liu

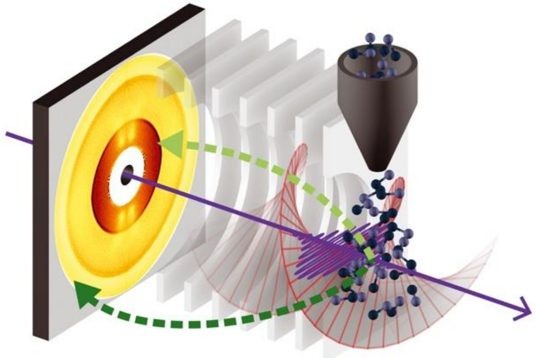

J. Laksmann



- angular streaking with:
  - an cookie-box-type eTOF spectrometer array
  - co-axial VMI spectrometer, new development by **XPD**



Nat. Photonics **12**, 215 (2018)
Phys. Rev A **100**, 053420 (2019)



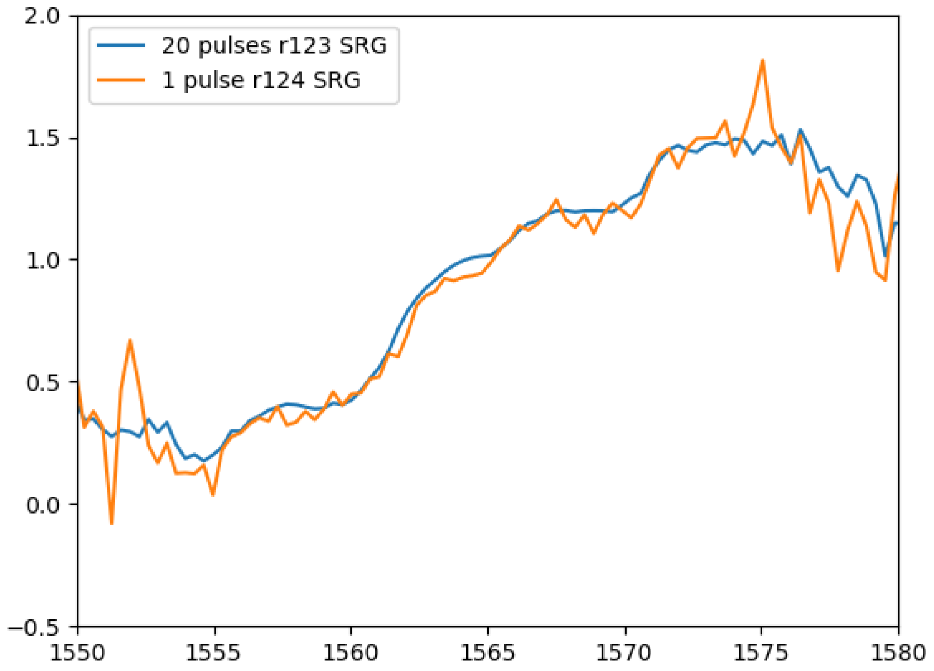
Nature **632**, 762 (2024)
Nat. Photonics **18**, 698 (2024)
Nat. Photonics **18**, 691 (2024)
Science***, 375,** 285* (2022)
Nat. Photonics **14**, 30 (2020)
Opt. Express **26**, 4531 (2018)

- **attosecond pulse + gas**
  - electron wavepacket (replica)
- **electron wavepacket in *streaking field***
  - the electric field of the streaking field acts as an ultrafast phase modulator
  - the spectral ***amplitude*** and ***phase*** of the as pulse are encoded in the modulated photoelectron spectrum
    - ► *pulse duration & profile, chirp, relative time delay*

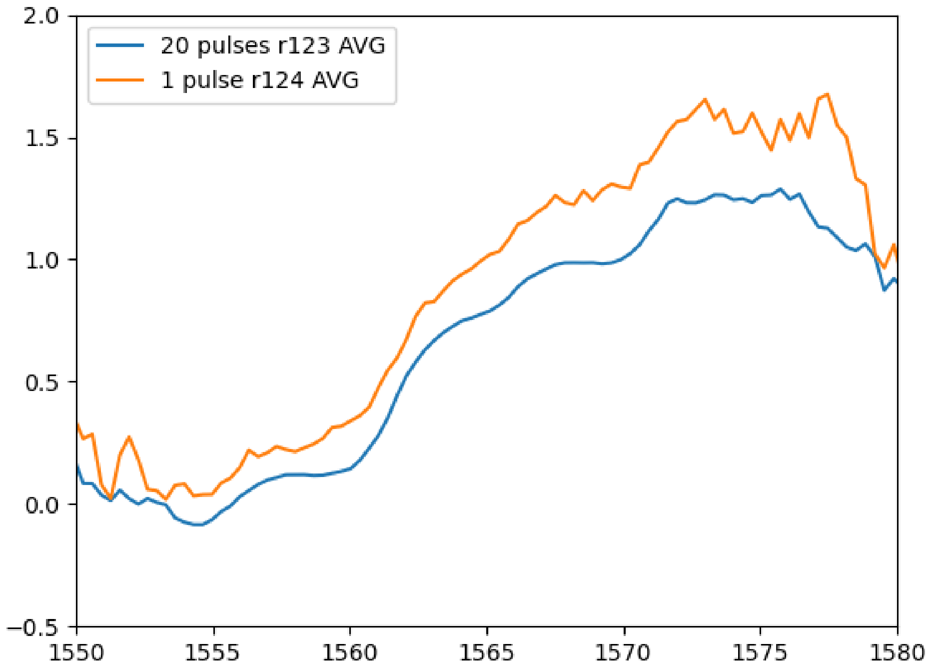
Self-referencing improves quality

Comparing 1 and 20 pulses / train, color 2 only, low intensity (cold absorption)

Self-referencing



Sequential averages sample / no sample

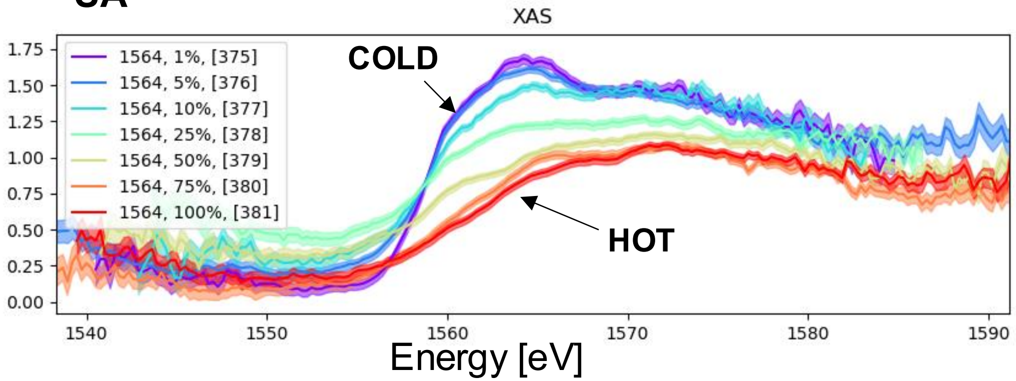


Note: PES normalization is also attempted (J. Laksman)

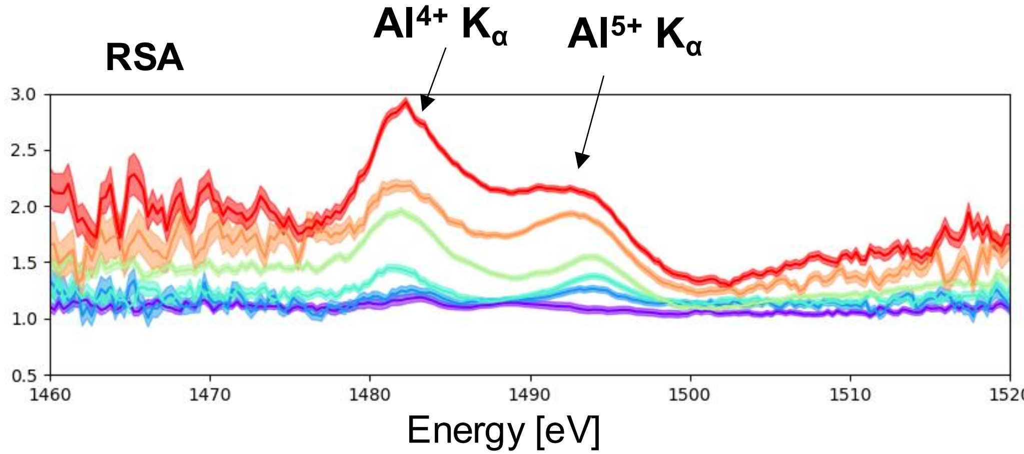
Spectrally-resolved SA and RSA in Aluminum

SA

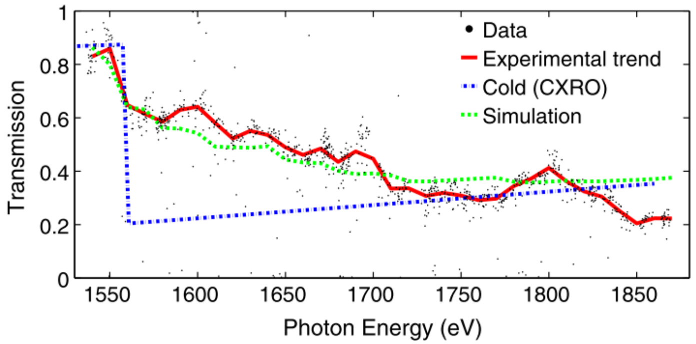
THIS WORK



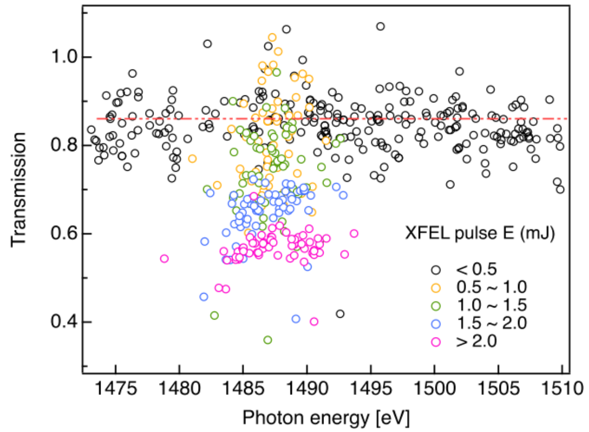
RSA



Rackstraw et al., PRL **114** (2015)



Cho et al., PRL **119** (2017)



- Edge bending → slope gives indication of temperature
- Visualization of ionic charge states through RSA
- Towards pump-probe experiment to study ionization dynamics

Beam size determination with curved gratings

- Original publication: M. Schneider et al., *Nature Communications* **9**, 214 (2018)
- Applied at SCS for KB mirrors characterization:
  G. Mercurio et al, *Optics Express* **30**, 20980 (2022)

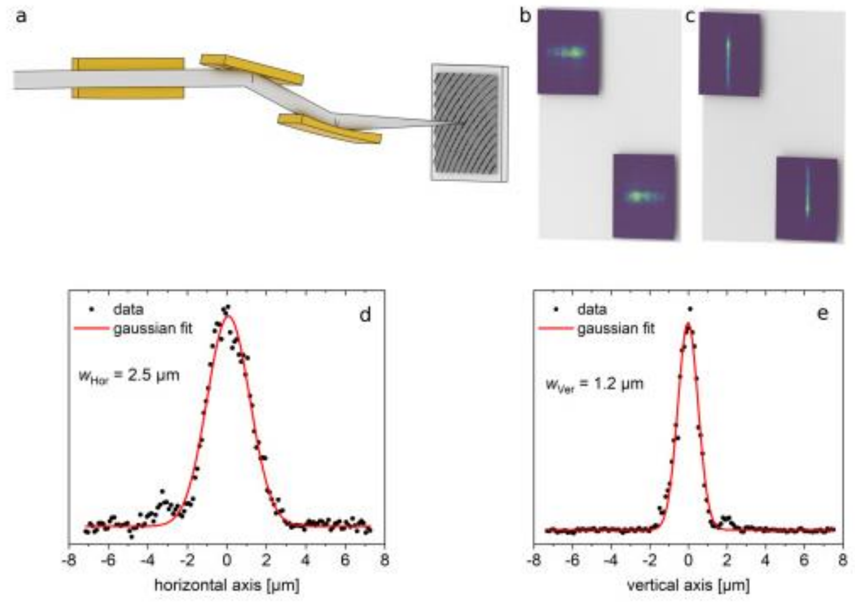


Fig. 2. Sketch of curved grating experimental setup (a), with HFM, VDM, VFM and grating (not in scale). Detector image of horizontally (b) and vertically (c) focused beam with the respective projections and Gaussian fits (d) and (e.) Note that the hyperbolic curved grating used rotates the image by 90°.

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