Short-pulse and polarization diagnostics at FELs based on electron spectroscopy

Dr. Markus Ilchen

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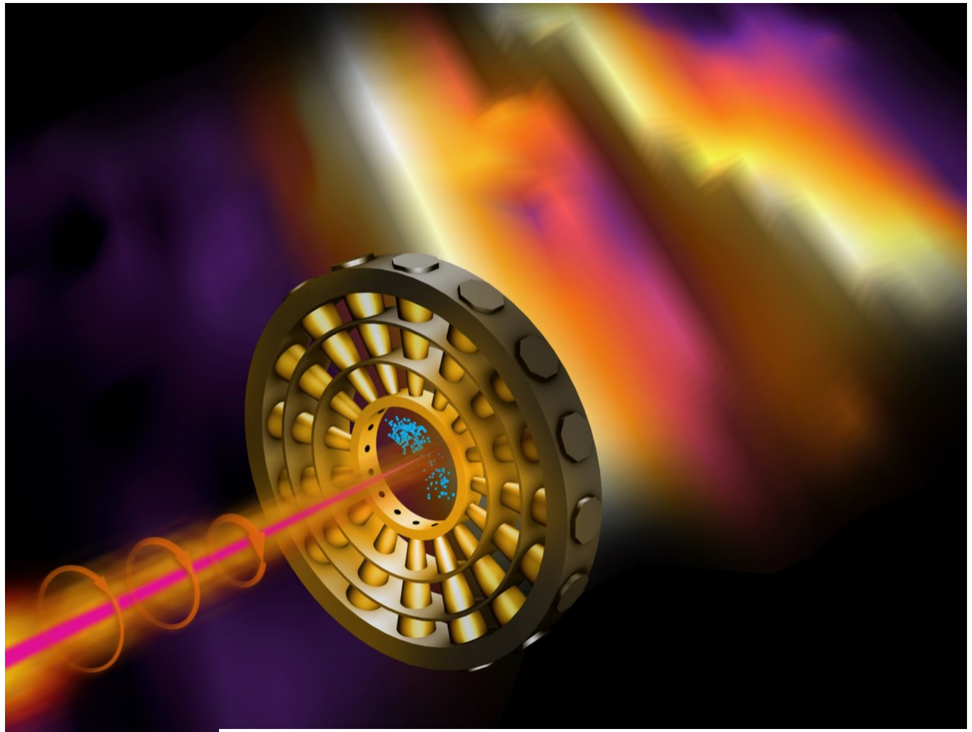
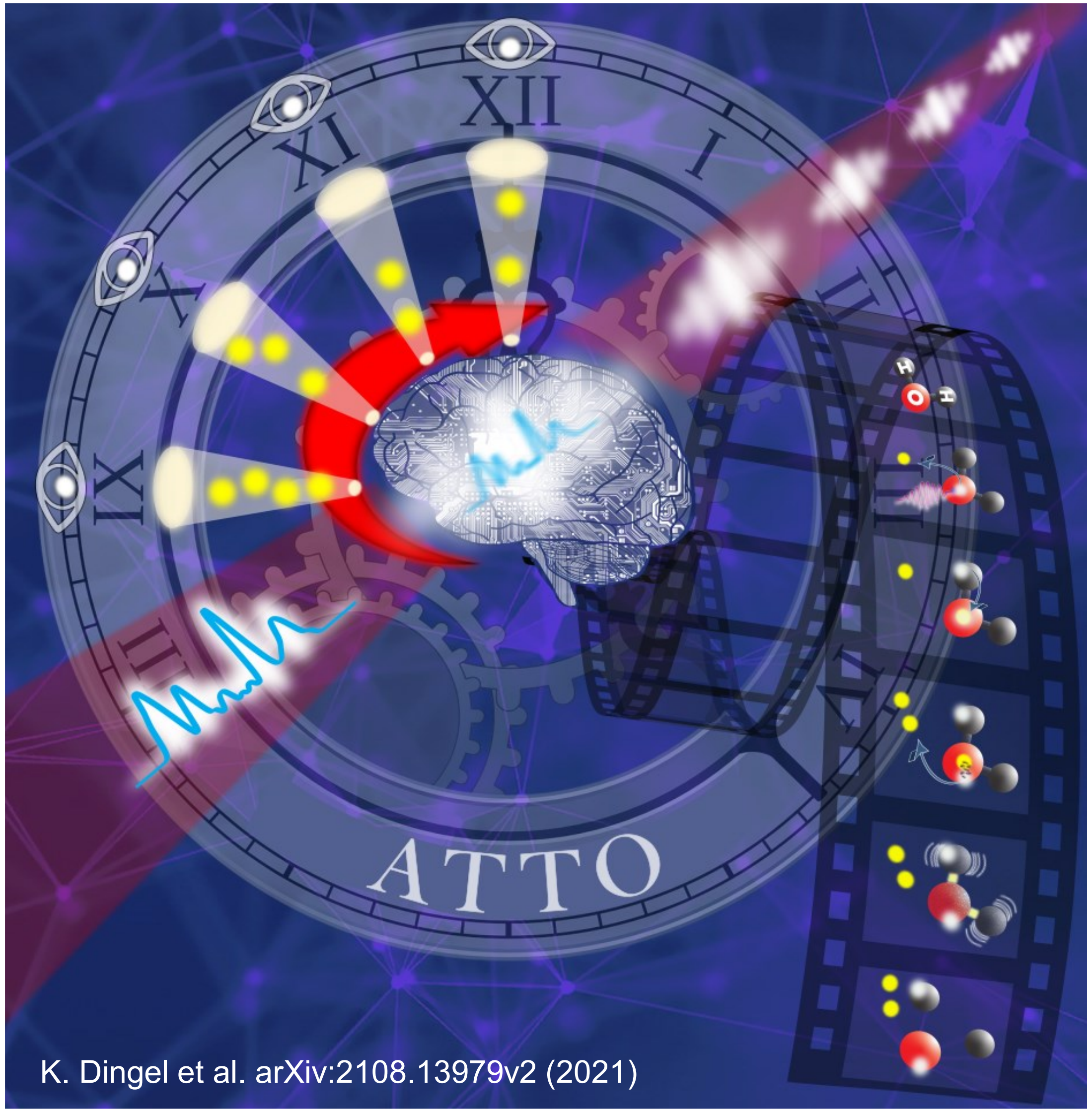
Notkestr. 85
22607 Hamburg





K. Dingel et al. arXiv:2108.13979v2 (2021)

Outline

Diagnostic Developments for (X)FELs based on electron spectroscopy

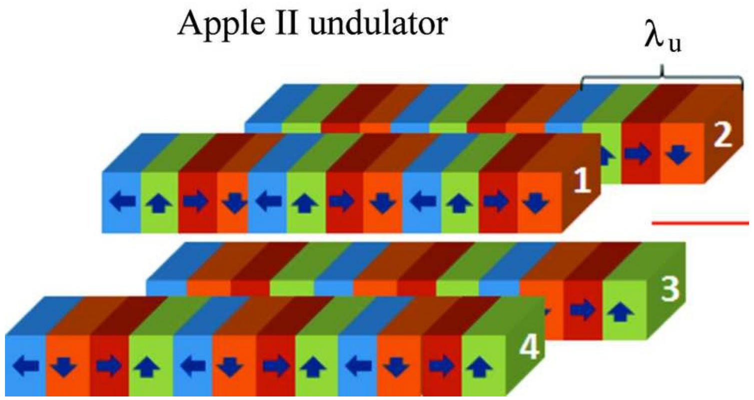
- Polarization Diagnostics
  - Online Diagnostics in the dipole plane
  - Validation of absolute degrees of polarization
- Exemplary Scientific Application of a Polarization-Controlled FEL
  - Nonlinear Circular Dichroism
  - Chirality Science
- Sub-femtosecond Pulse Diagnostics
  - Angular Streaking – Principle
  - Atto-Campaign at European SQS
  - Outlook and Applications
- Challenges and Outlook

Gairhos Herbert, Download flickr.com 2020

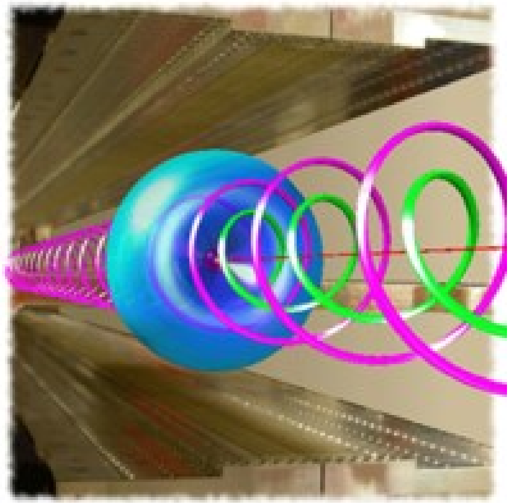
Operation Modes for New Scientific Applications

Ultraintense CPL XUV Pulses from Free-Electron Lasers

- FERMI @ Elettra, Trieste, Italy

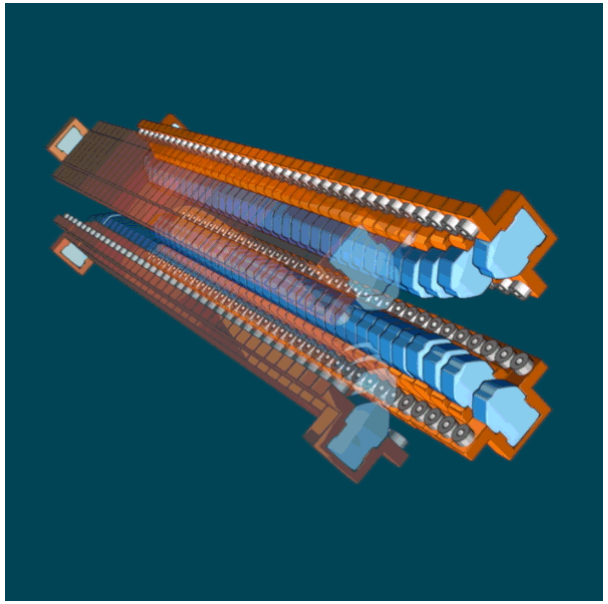


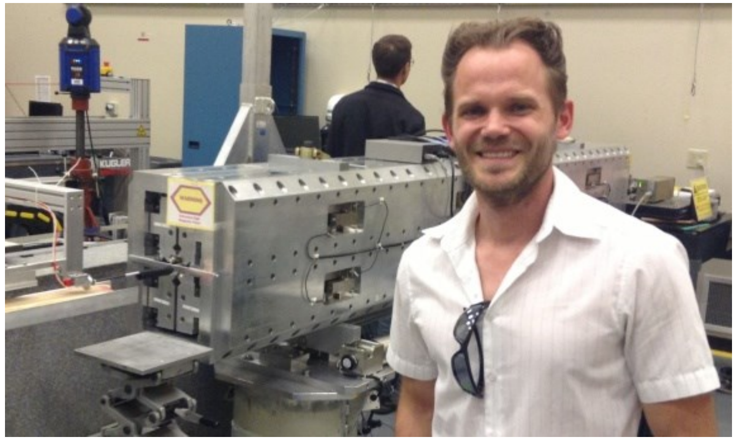
H. Wang et al., J. Synchrotron Rad. **19**, 944-948 (2012)

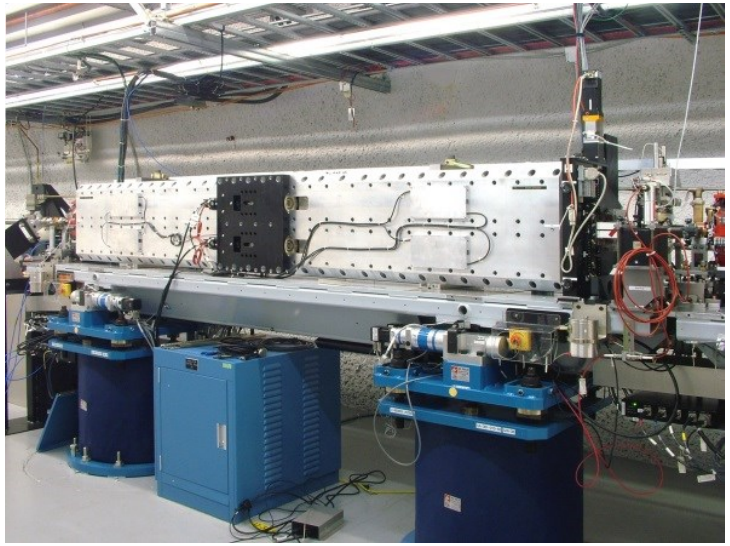


- LCLS @ SLAC, Menlo Park, USA

SLAC News 2016 – *A. Lutman et al., Nature Photonics,* ***10***, *468, 2016*







Diagnostic Aspects to Tackle for Attoscience with Polarization Control

Road towards FEL Applications

Max von Laue Fest at DESY – © DESY

DORIS III and PETRA III, DESY, Hamburg

in the Group of J. Viefhaus, (now HZB Berlin)

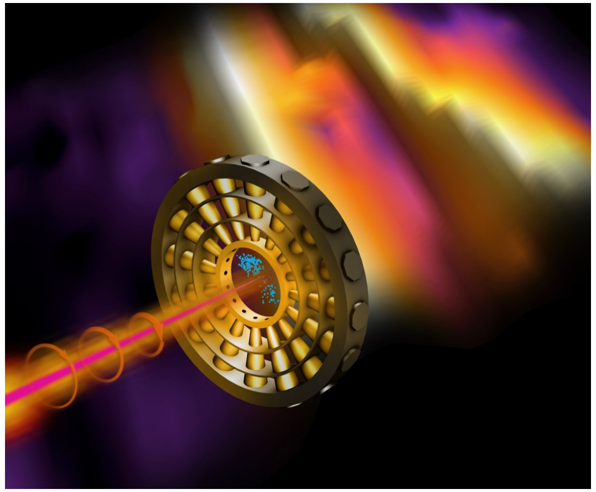
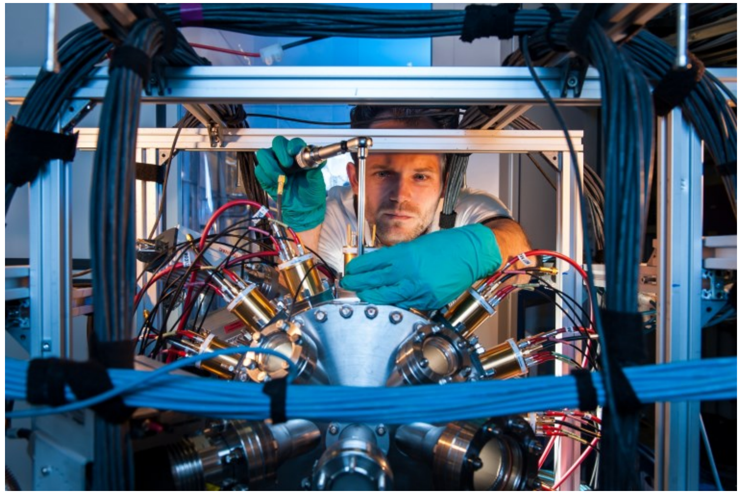
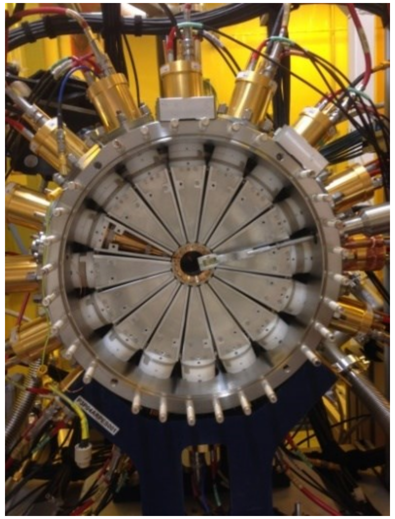
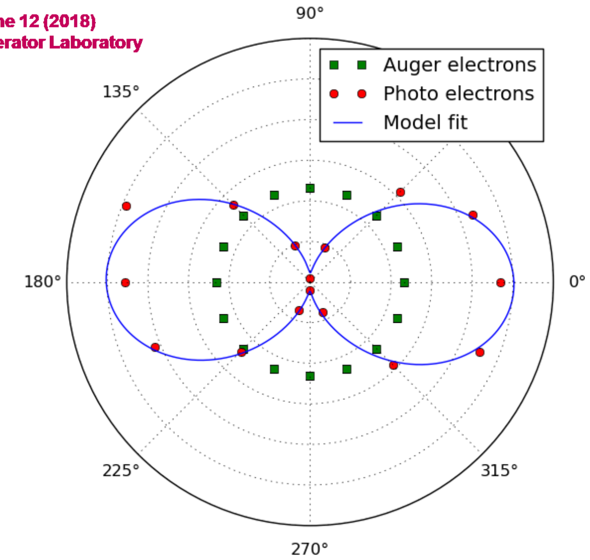
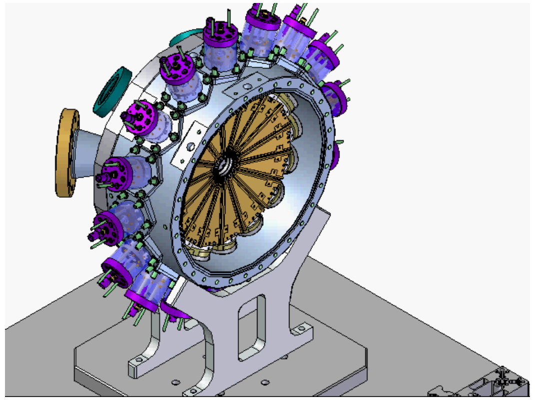


Photo: Cover Page Nature Photonics, volume 12 (2018)





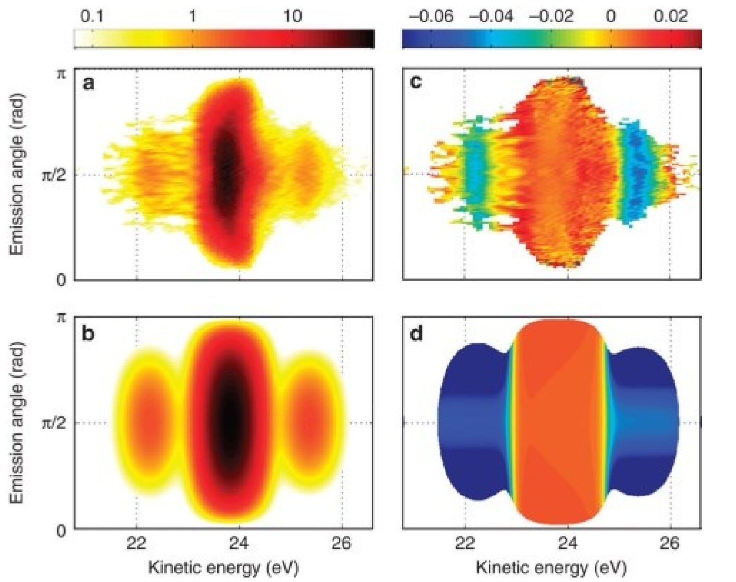




$$P(\theta) = 1 + \frac{\beta}{4} \cdot \left[1 + 3 \cdot P_{lin} \cdot \cos\left(2 \cdot (\theta - \psi)\right) \right]$$

The First Circularly Polarizing Short-Wavelength FEL - FERMI

Polarization Commissioning in the



nature COMMUNICATIONS

ARTICLE

Received 13 Feb 2014 | Accepted 14 Mar 2014 | Published 16 Apr 2014 DOI: 10.1038/ncomms4648

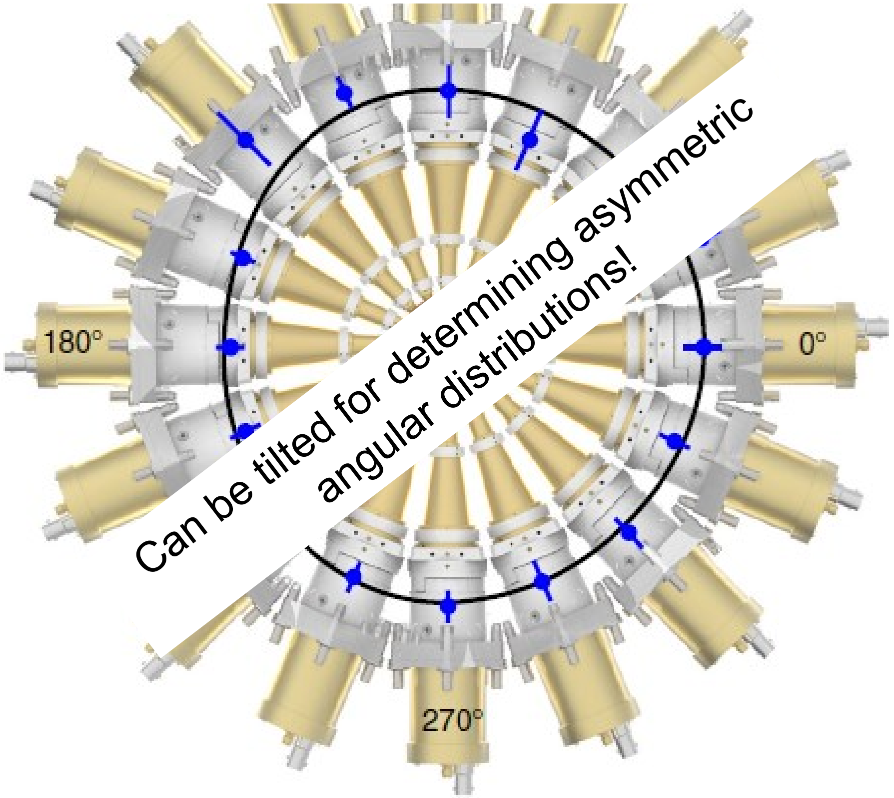
Determining the polarization state of an extreme ultraviolet free-electron laser beam using atomic circular dichroism

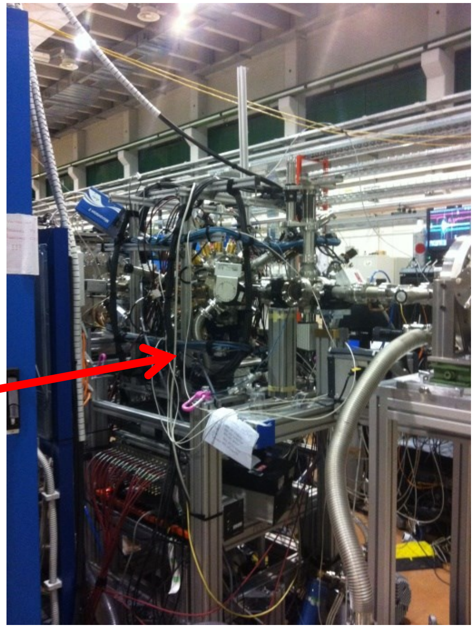
T. Mazza[1], M. Ilchen[1], A.J. Rafipoor[1], C. Callegari[2], P. Finetti[2], O. Plekan[2], K.C. Prince[2,3,4], R. Richter[2], M.B. Danailov[2], A. Demidovich[2], G. De Ninno[2,5], C. Grazioli[2], R. Ivanov[2,5], N. Mahne[2], L. Raimondi[2], C. Svetina[2,6], L. Avaldi[7], P. Bolognesi[7], M. Coreno[7], P. O'Keeffe[7], M. Di Fraia[8], M. Devetta[9], Y. Ovcharenko[10], Th. Möller[10], V. Lyamayev[11], F. Stienkemeier[11], S. Düsterer[12], K. Ueda[13], J.T. Costello[14], A.K. Kazansky[15,16,17], N.M. Kabachnik[1,13,17,18] & M. Meyer[1]

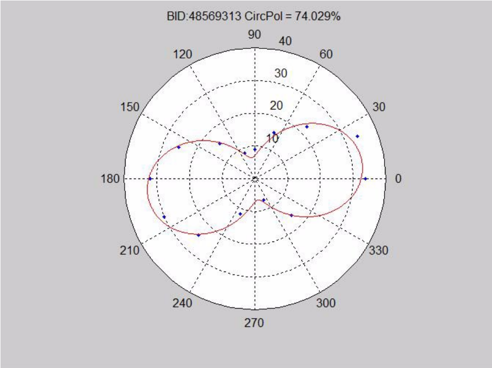
nature COMMUNICATIONS

Symmetry breakdown of electron emission in extreme ultraviolet photoionization of argon

M. Ilchen[1,2], G. Hartmann[2,3], E.V. Gryzlova[4], A. Achner[1], E. Allaria[5], A. Beckmann[6], M. Braune[3], J. Buck[1,3], C. Callegari[5], R.N. Coffee[7], R. Cucini[5], M. Danailov[5], A. De Fanis[1], A. Demidovich[5], E. Ferrari[8], P. Finetti[5], L. Glaser[3], A. Knie[2], A.O. Lindahl[9], O. Plekan[5], N. Mahne[5], T. Mazza[1], L. Raimondi[5], E. Roussel[5], F. Scholz[3], J. Seltmann[3], I. Shevchuk[3], C. Svetina[10], P. Walter[3,7], M. Zangrando[5,11], J. Viefhaus[3], A.N. Grum-Grzhimailo[1,4] & M. Meyer[1]





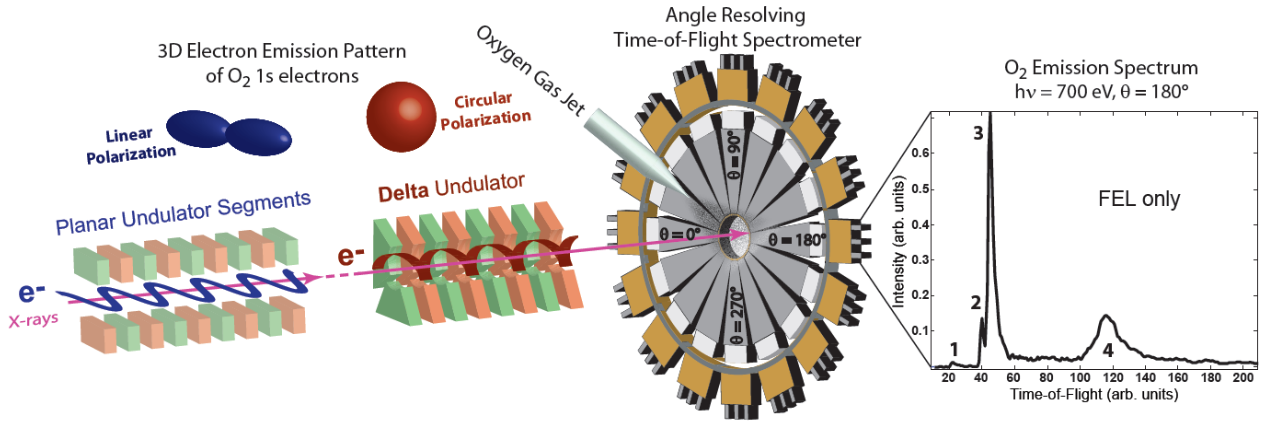


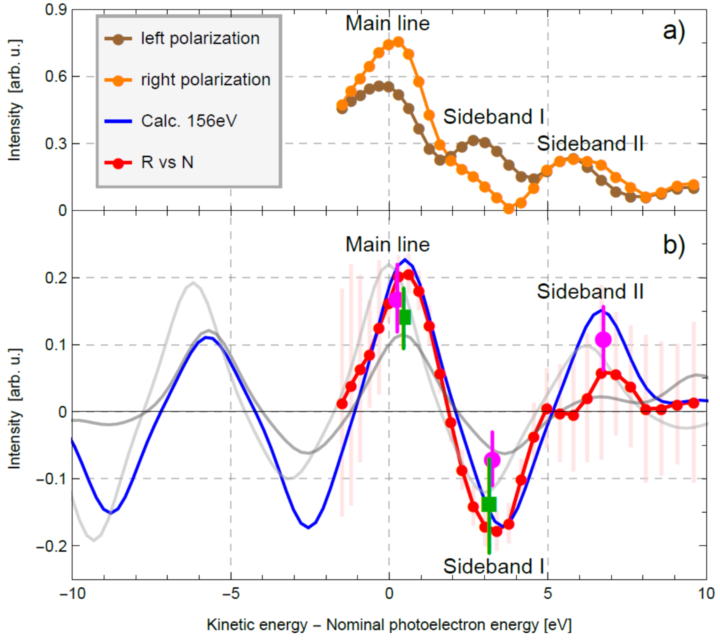
E. Ferrari et al.

Scientific Reports, **5**, 13531 (2015)

Diagnostic Aspects to Tackle for Attoscience with Polarization Control

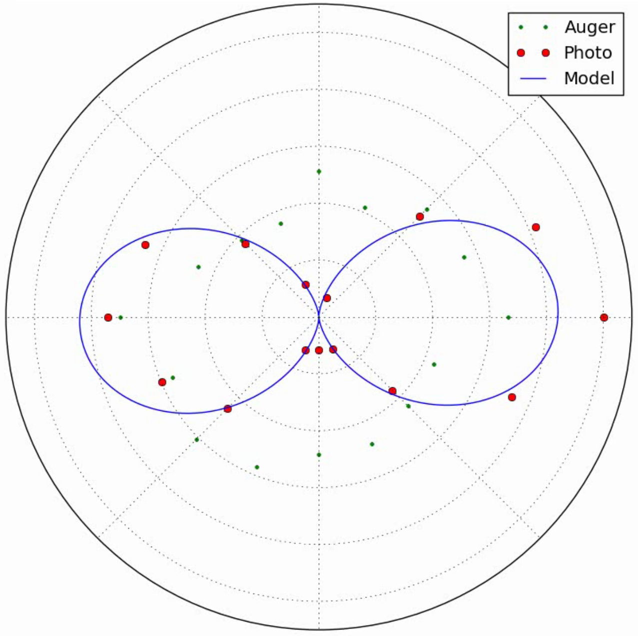
Confirmation about Absence of Randomly Polarized Light is Imperative!



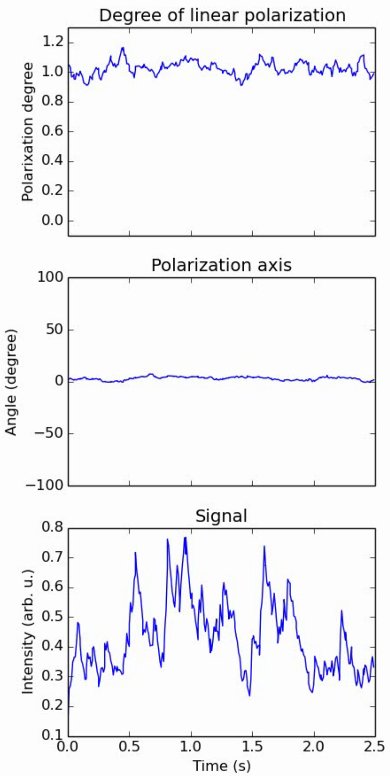


Hartmann et al.
REVIEW OF SCIENTIFIC INSTRUMENTS **87**, 083113 (2016)

Online Analysis by A. Lindahl

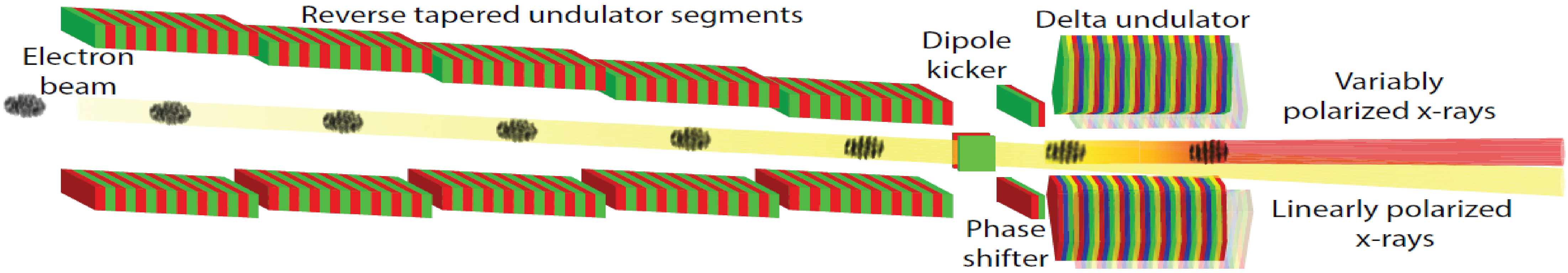


$$P(\theta) = 1 + \frac{\beta}{4} \cdot \left[1 + 3 \cdot P_{lin} \cdot \cos\left(2 \cdot (\theta - \psi)\right)\right]$$



Operation Modes for New Scientific Applications

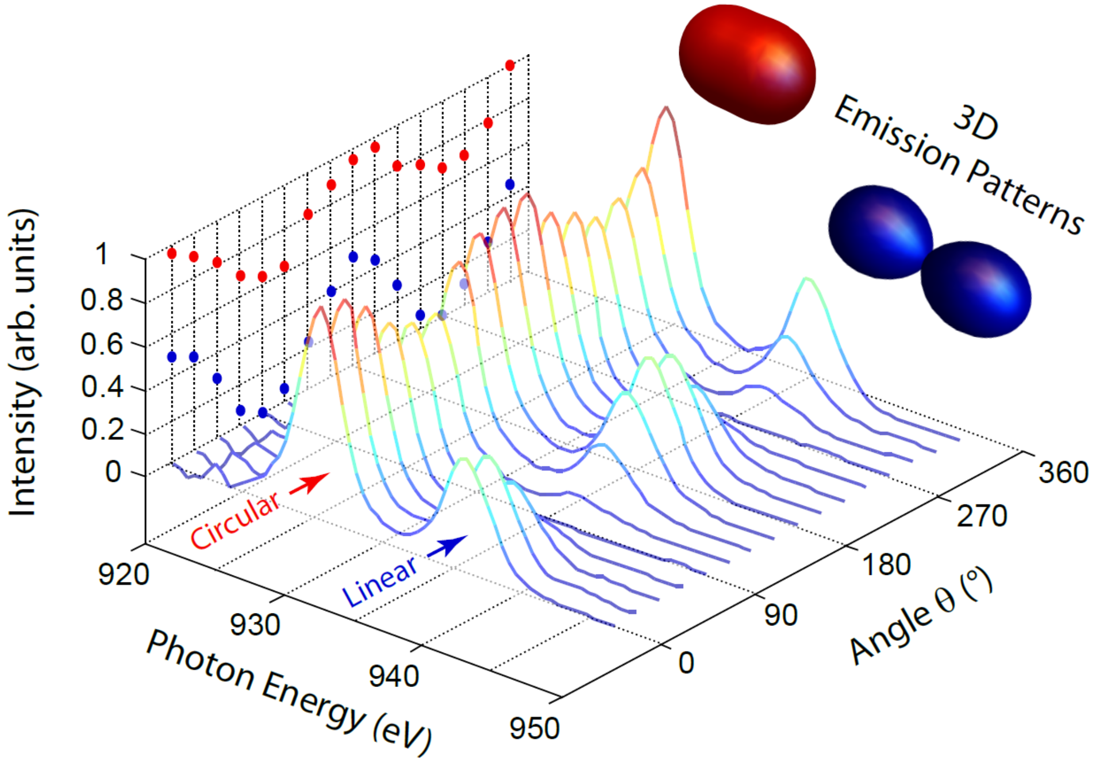
Ultraintense CPL XUV Pulses from Free-Electron Lasers



The pulses were controllable in:

1. Polarization (one is always linear)
2. Energy separation (~2%),
3. Time delay (-10 fs to 800 fs),
4. Intensity distribution (max. achieved so far is a total pulse energy of 300 µJ, certain limitations apply)

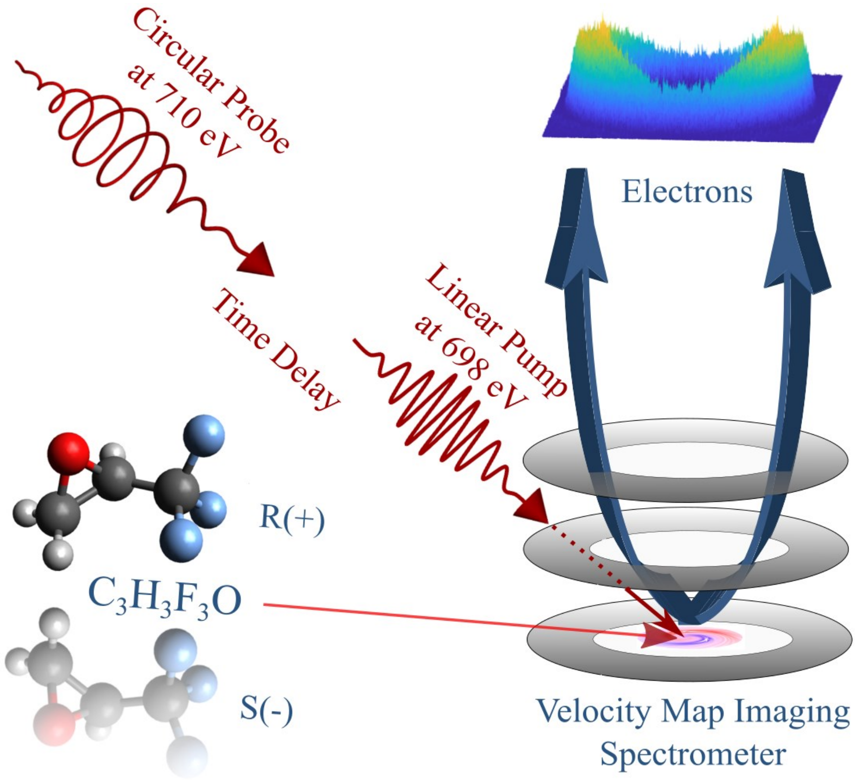
Lutman et al. NATURE PHOTONICS | VOL 10 | JULY 2016

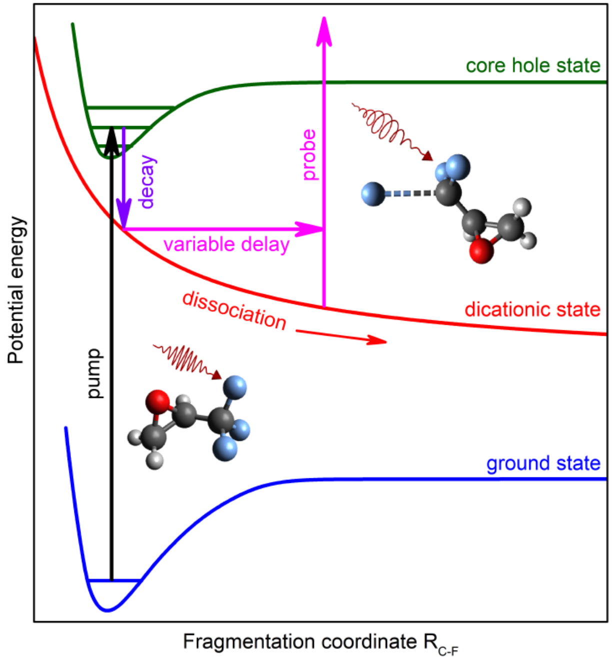


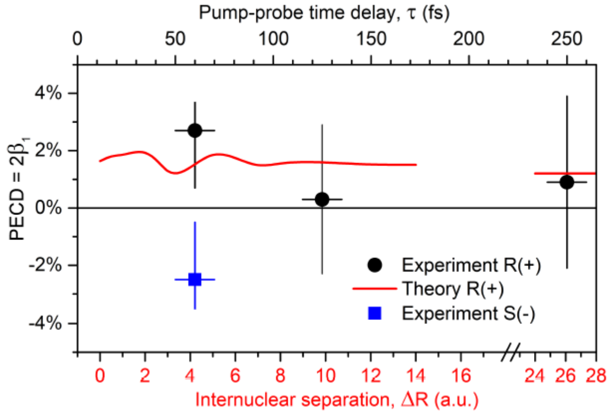
Ultrafast Circular Dichroism Experiments

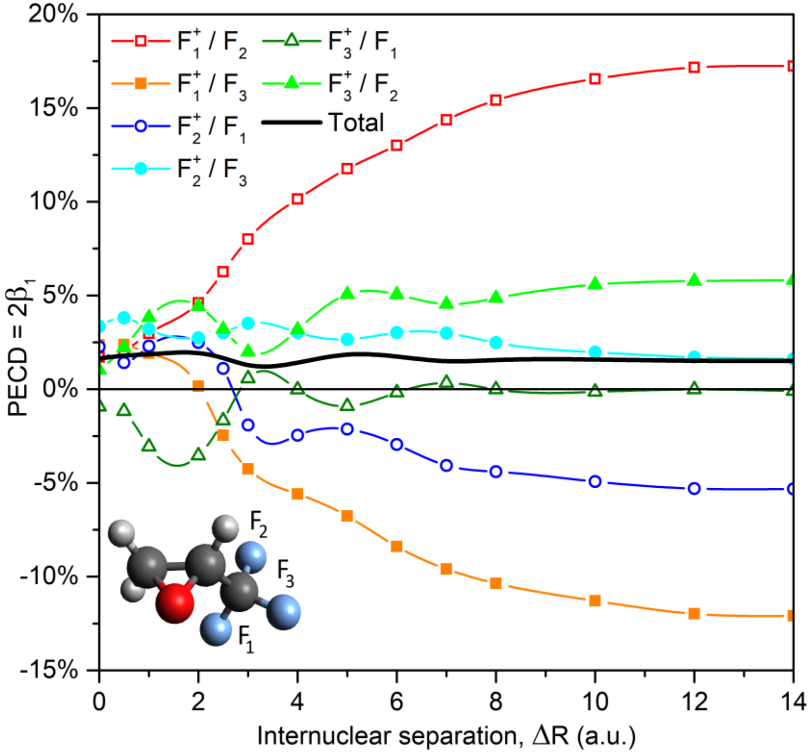
Example of a Kick-Off Chirality Experiment at LCLS

- PECD as tool for chiral recognition!
- Double site-specificity possible!
- Ultrafast chirality at XFELs in reach.





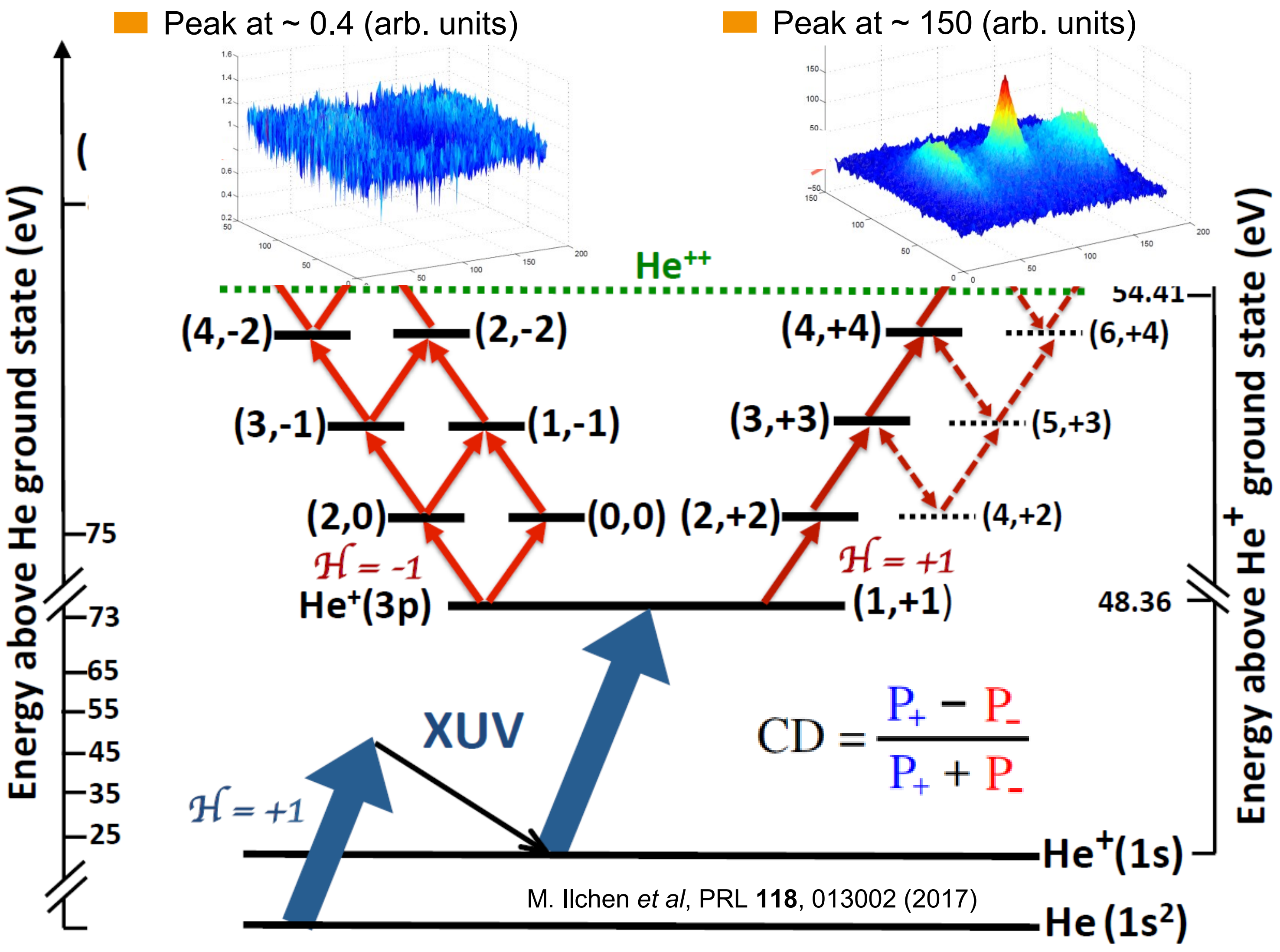




M. Ilchen *et al*,
Communications Chemistry **4**, 119 (2021)

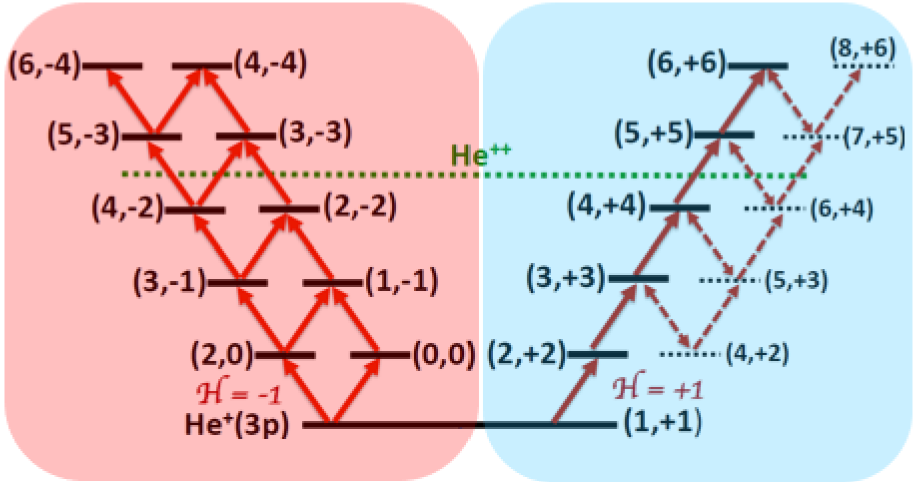
Nonlinear Circular Dichroism Experiments

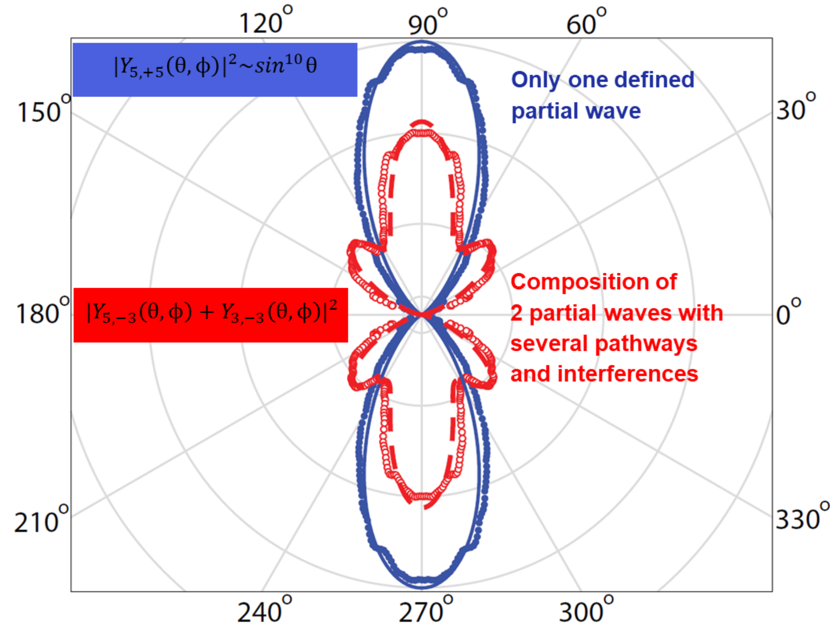
Example of Experiments at FERMI



M. Ilchen *et al*, PRL **118**, 013002 (2017)

NIR Intensity = 1.4*10¹² W/cm²





NIR Intensity = 1.4*10¹² W/cm²

Entering the Attosecond Regime

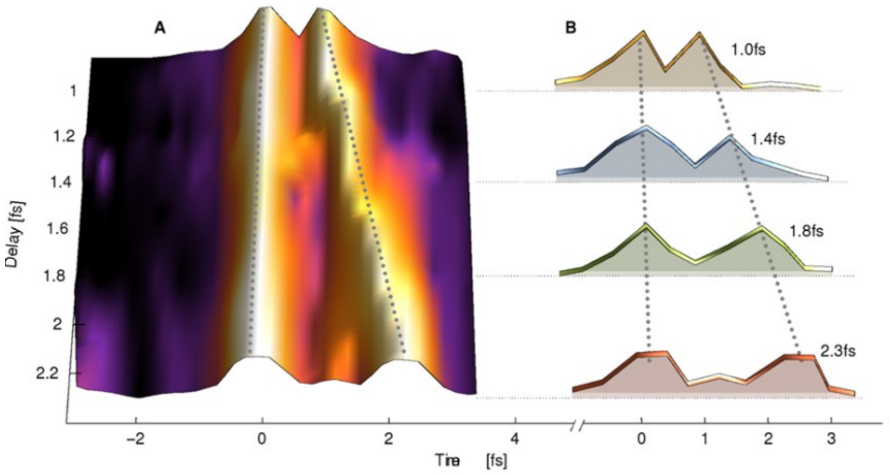
- First investigation of single-spike lasing at ~1000 eV via a new "angular streaking" technique at LCLS, USA (upper right) enables sorting for 500 as pulses or specific pulse shapes.

- Perspectives for sub-fs pump-probe schemes within a single XFEL pulse (lower right):
  - ► Electron dynamics such as relaxation and migration can be investigated for the first time.
  - ► A project at European XFEL was conducted in June 2022.

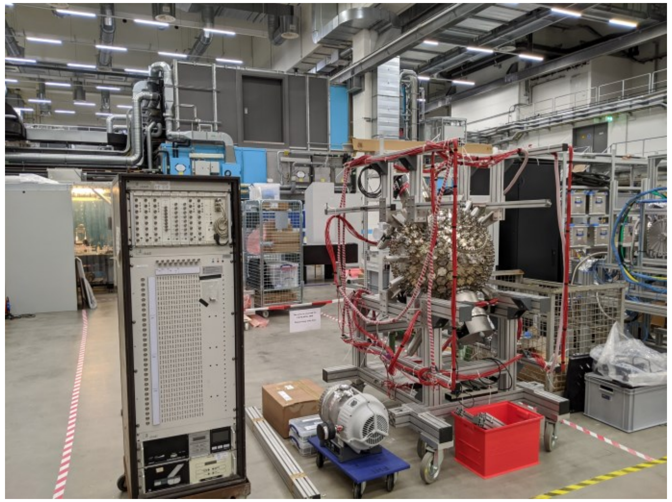
nature photonics ARTICLES
https://doi.org/10.1038/s41566-018-0107-6

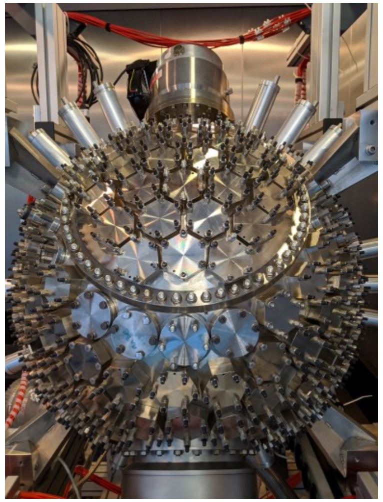
Attosecond time-energy structure of X-ray free-electron laser pulses

N. Hartmann[1,2,17]*, G. Hartmann[3,4], R. Heider[5], M. S. Wagner[5], M. Ilchen[1,6,7], J. Buck[3,6], A. O. Lindahl[1,8,9], C. Benko[10], J. Grünert[6], J. Krzywinski[1], J. Liu[6], A. A. Lutman[1], A. Marinelli[1], T. Maxwell[1], A. A. Miahnahri[1], S. P. Moeller[1], M. Planas[6], J. Robinson[1], A. K. Kazansky[11,12,13], N. M. Kabachnik[6,14], J. Viefhaus[3], T. Feurer[2], R. Kienberger[5,15], R. N. Coffee[1,7] and W. Helml[1,5,16]*



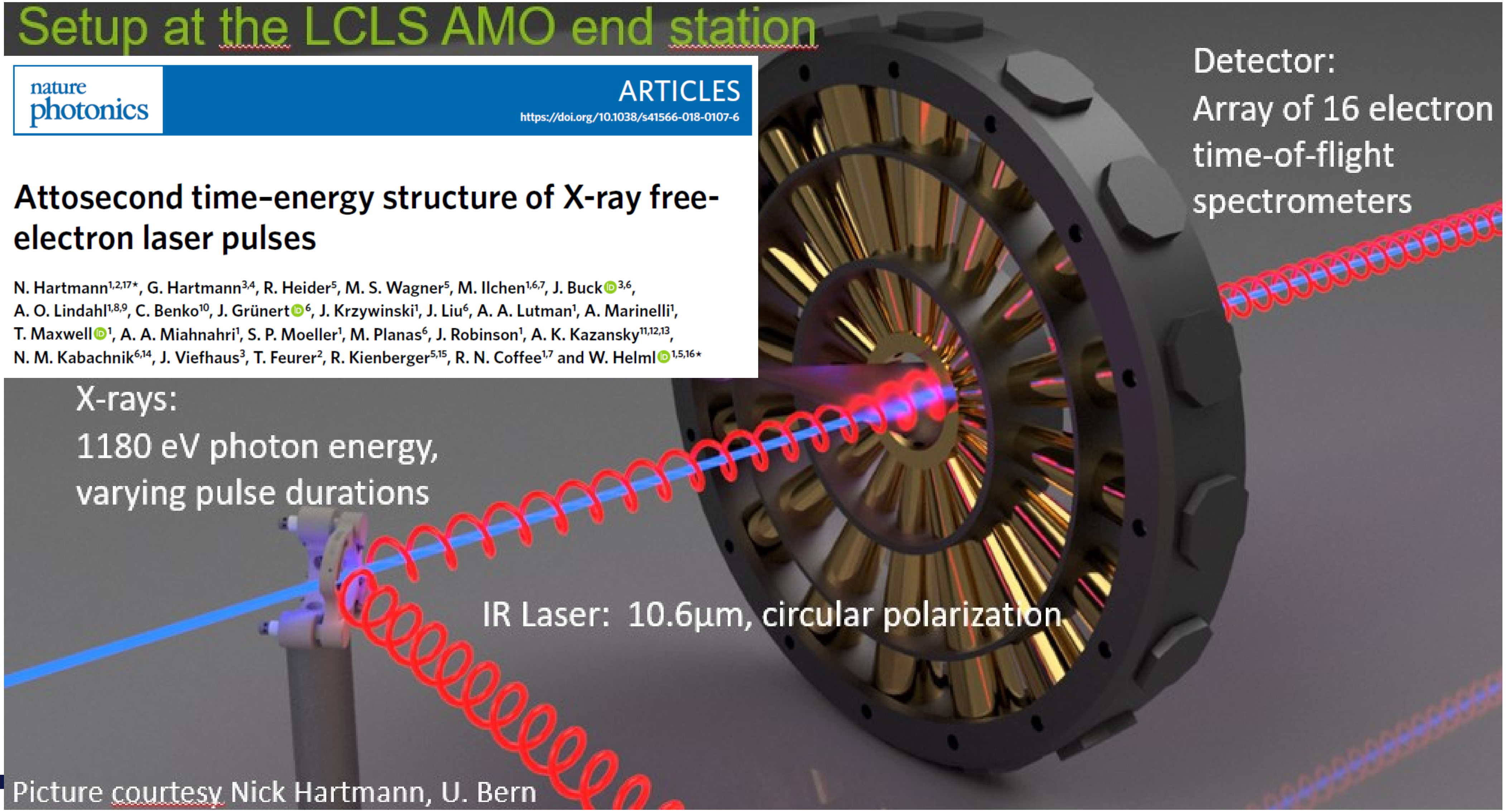
Sorted pulses demonstrating the principal capability of single-pulse pump-probe with attosecond pulses.





Left: Experimental equipment for the attosecond-campaign at SQS: Angle-resolving time-of-flight spectrometer setup for studying nonlinear electron dynamics in the soft X-ray regime.

Diagnosing sub-femtosecond pulses

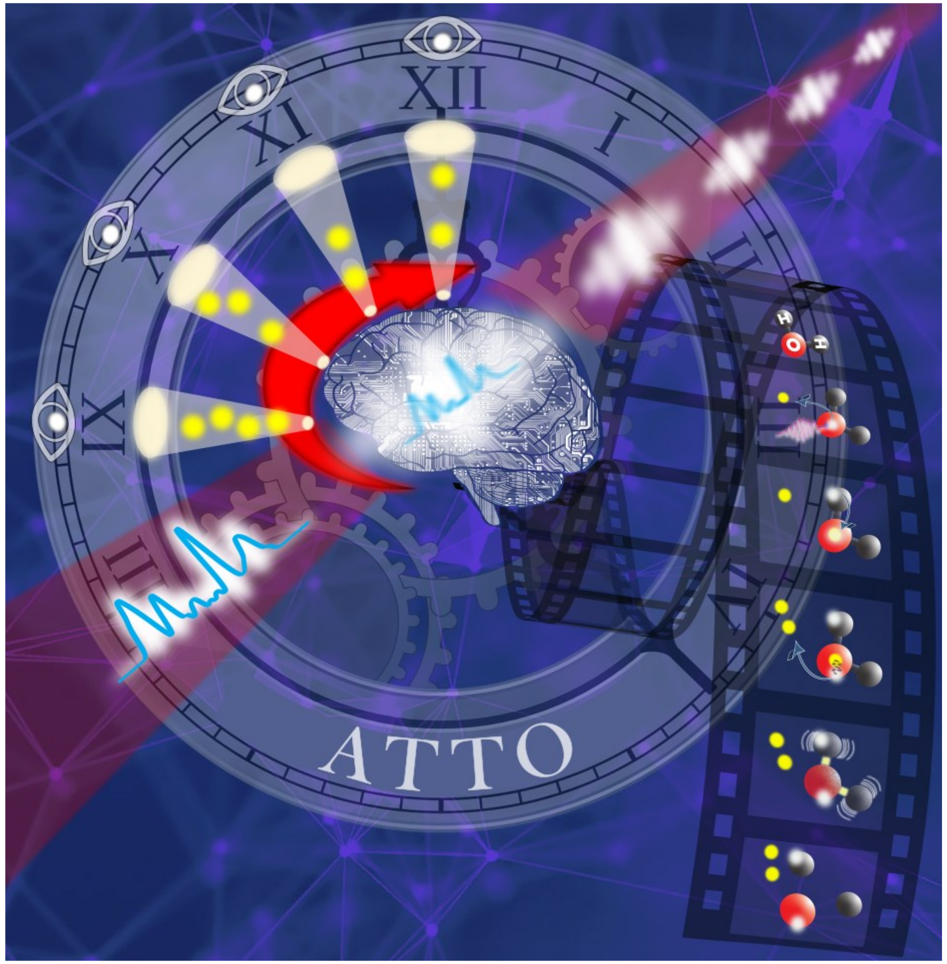


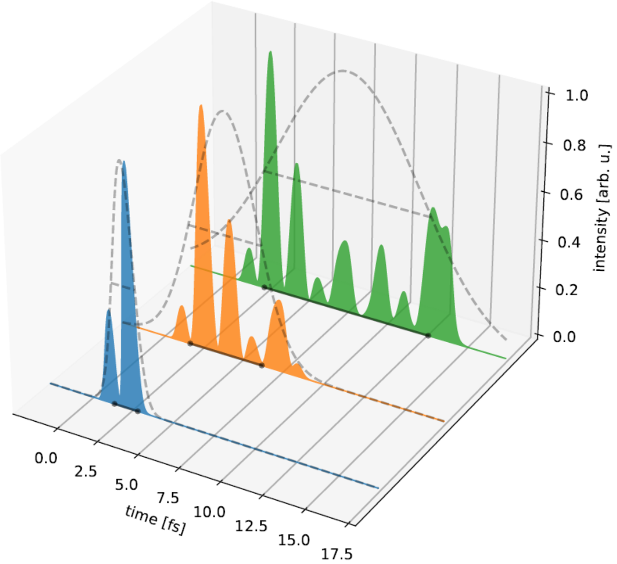
Angular Streaking at SQS – Helml/Ilchen et al.

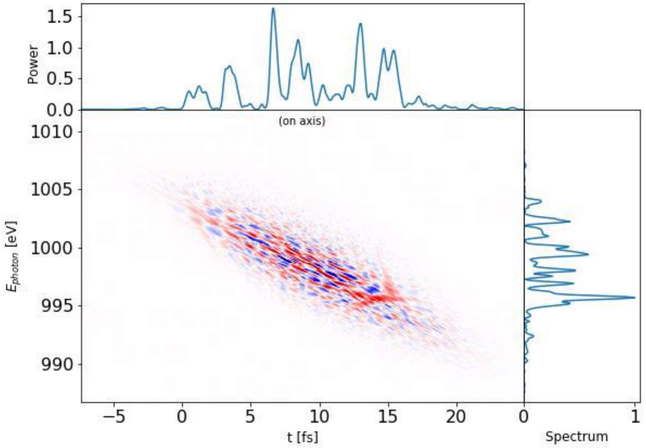
Aims: 1) Resolve the full time-energy structure of every incoming pulse online with attosecond resolution

2) Discover a single-spike lasing event aka an isolated attosecond pulse

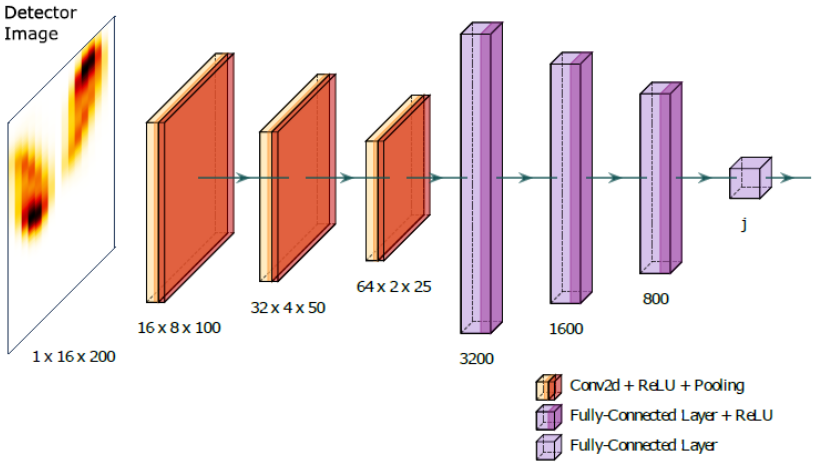
3) Track electron dynamics in an atom – Lifetime of resonant double-core-hole Auger-Meitner







Courtesy of S. Serkez, XFEL



K. Dingel et al. arXiv:2108.13979v2 (2021)

Angular Streaking at SQS – Helml/Ilchen et al.

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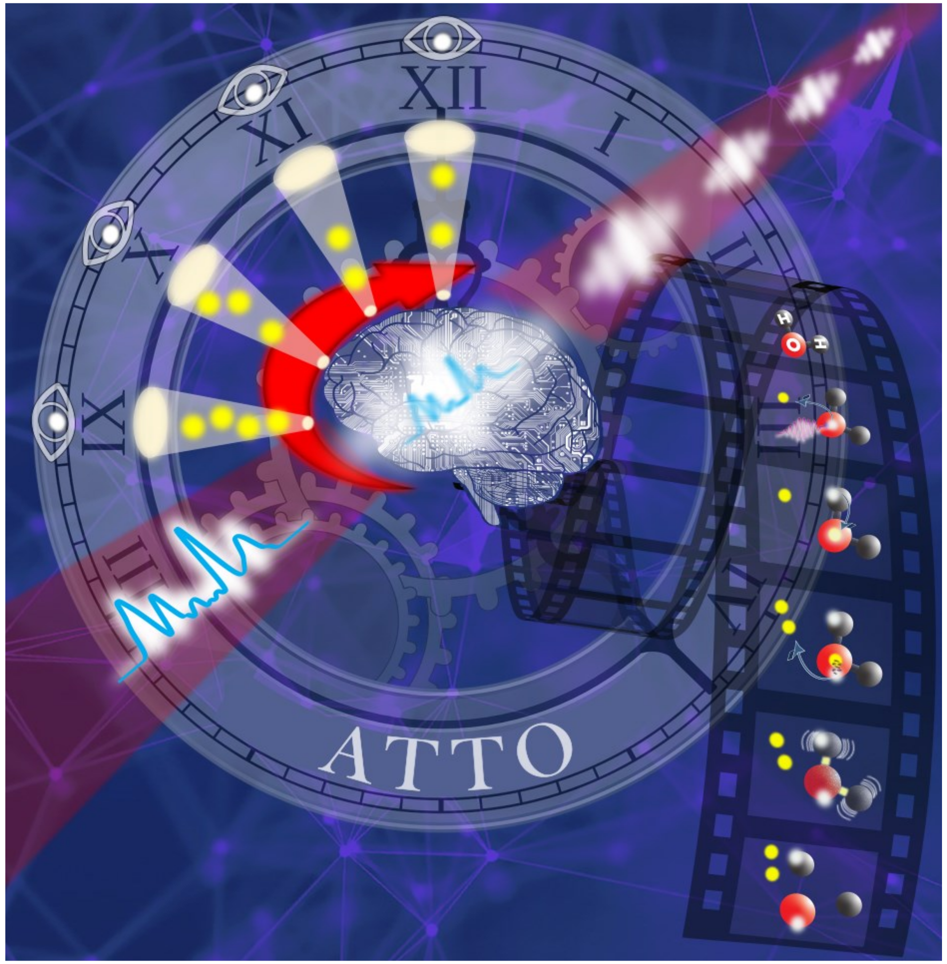
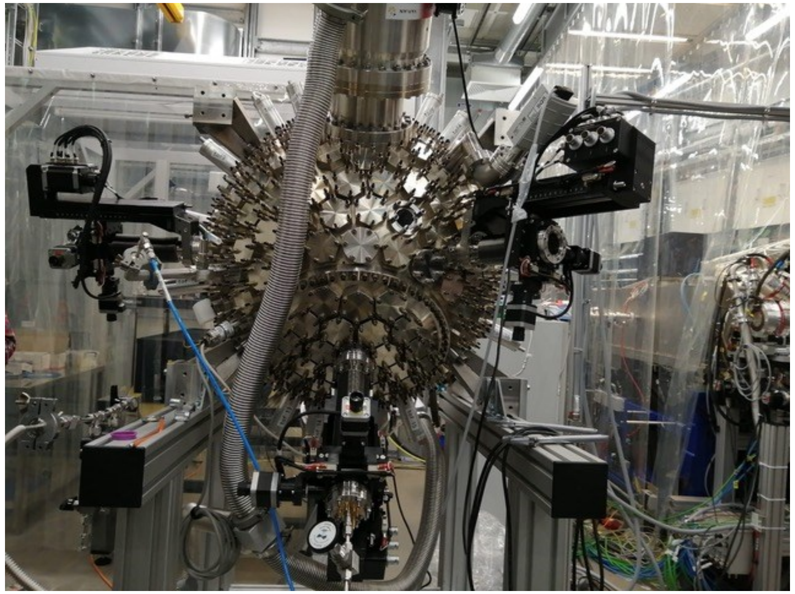
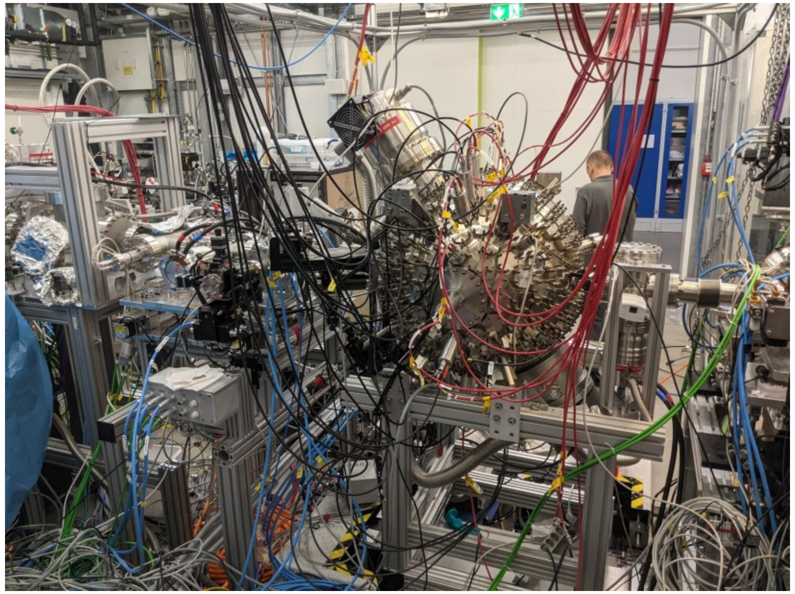


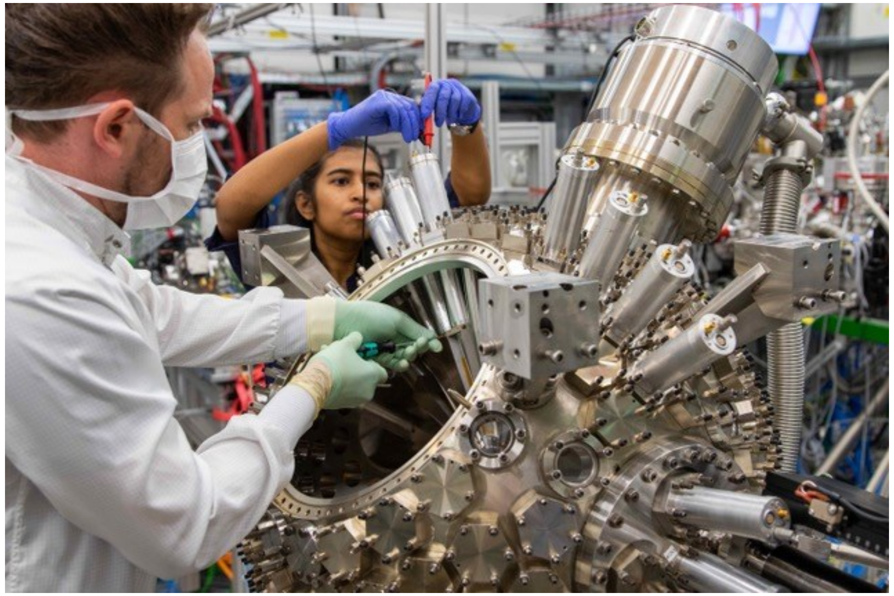
K. Dingel et al. arXiv:2108.13979v2 (2021)

Instrument installation at SQS

Perspectives of Polarization Control in the Attosecond Regime in Europe

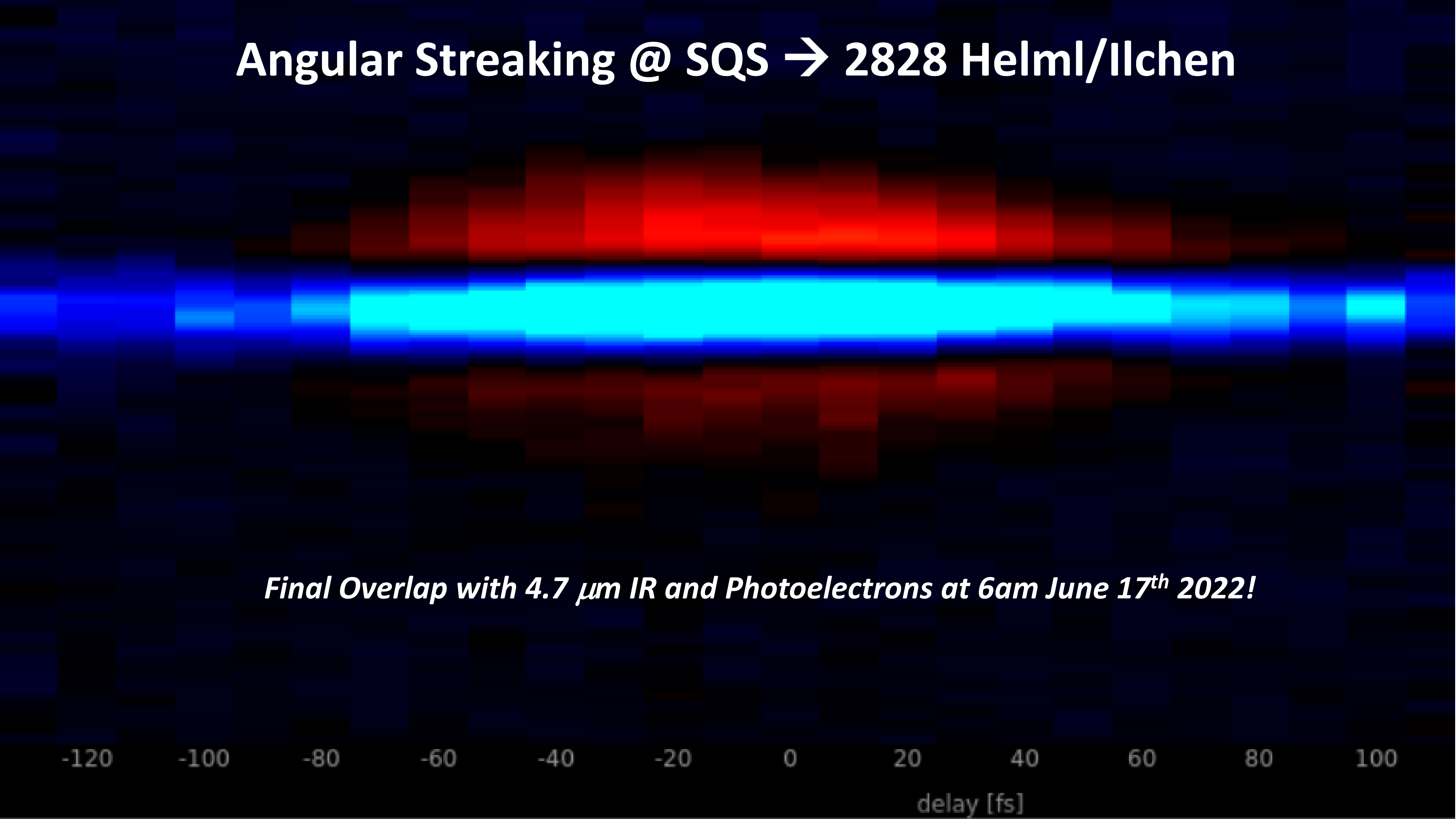






Our beamtime to reveal the full time-energy structure of every incoming pulse at European XFEL (SQS) was three weeks ago (Helml/Ilchen)! Observing the first isolated attosecond pulse at European XFEL and directly measuring the lifetime of highly transient resonances were the ultimate goals!

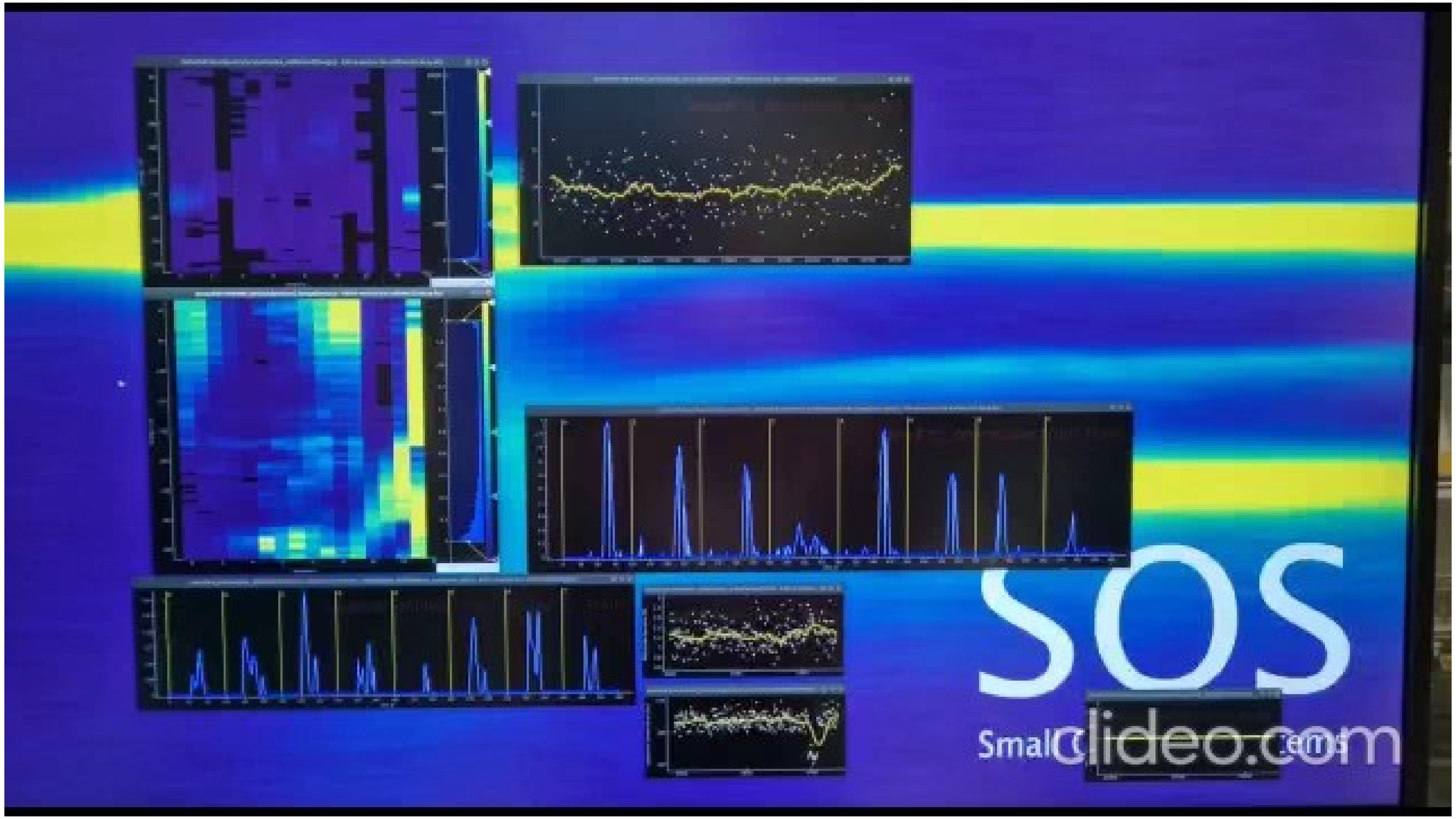
Angular Streaking @ SQS → 2828 Helml/Ilchen



Final Overlap with 4.7 μm IR and Photoelectrons at 6am June 17th 2022!

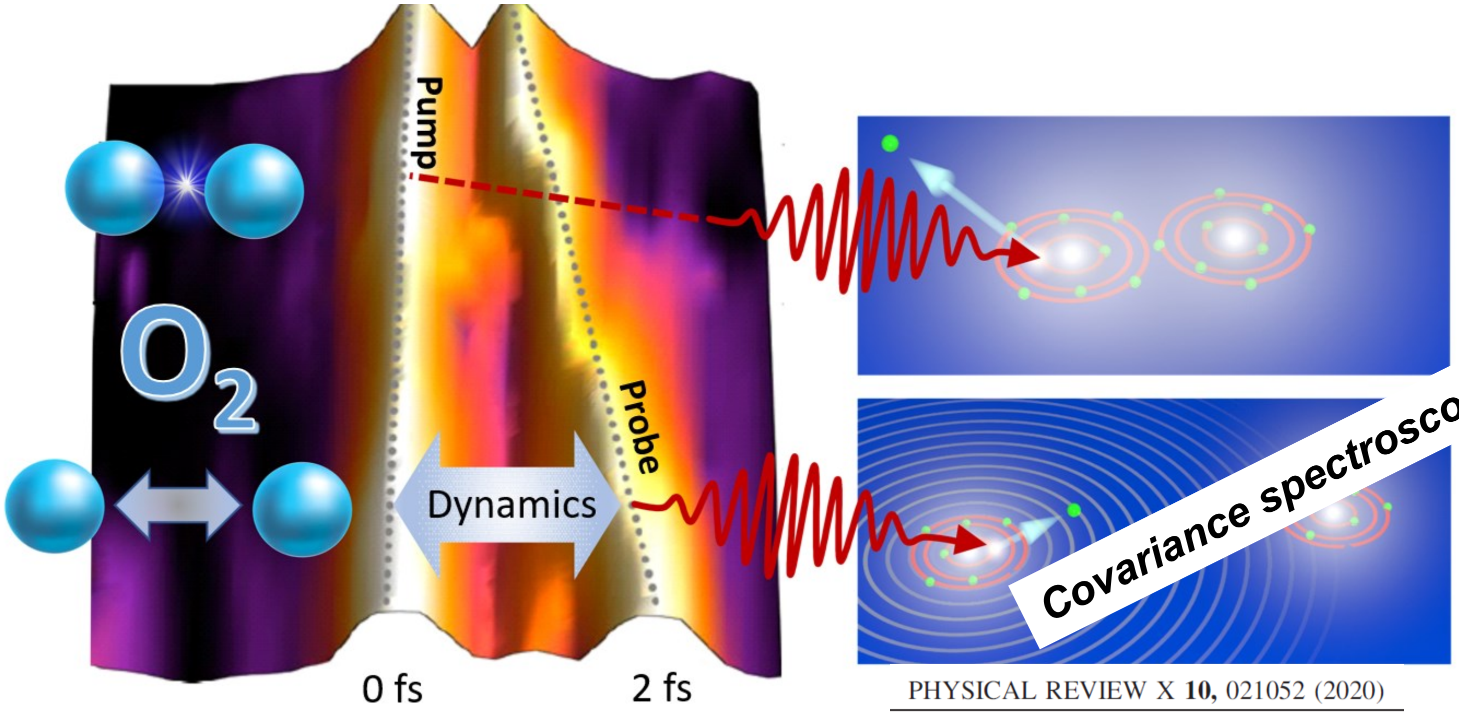
Online Diagnostics in Real Time

Aims: 1) Resolve the full time-energy structure of every incoming pulse online with attosecond resolution
2) Discover a single spike lasing event aka an isolated attosecond
3) Track electron dynamics in an atom – Lifetime of resonant double-core-hole Auger-Meitner



Next steps for attosecond investigations

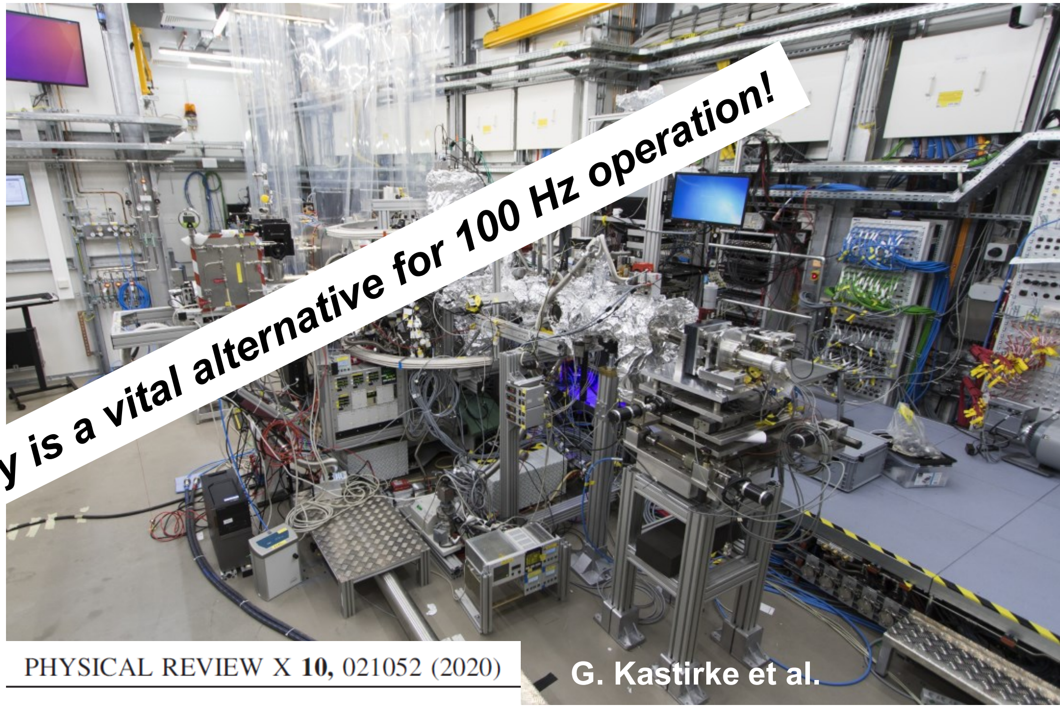
Combination experiments of angular streaking and other instrumentation



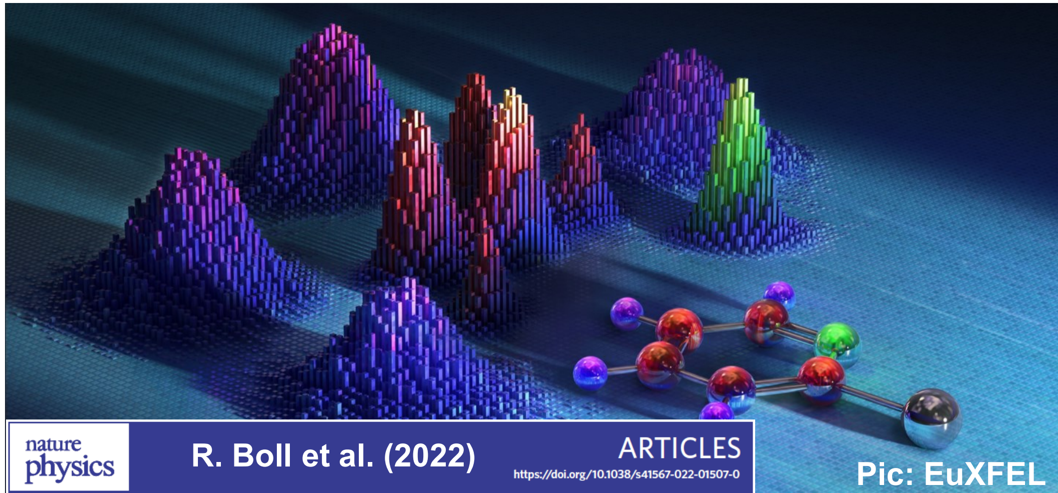
Time between Pulses

Nature Photonics, **12**, 215 (2018)

Covariance spectroscopy is a vital alternative for 100 Hz operation!



PHYSICAL REVIEW X **10**, 021052 (2020) G. Kastirke et al.



R. Boll et al. (2022)

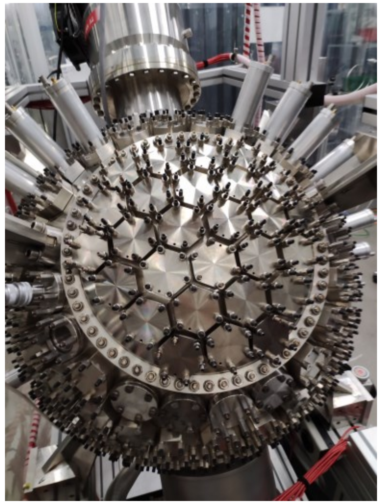
Pic: EuXFEL

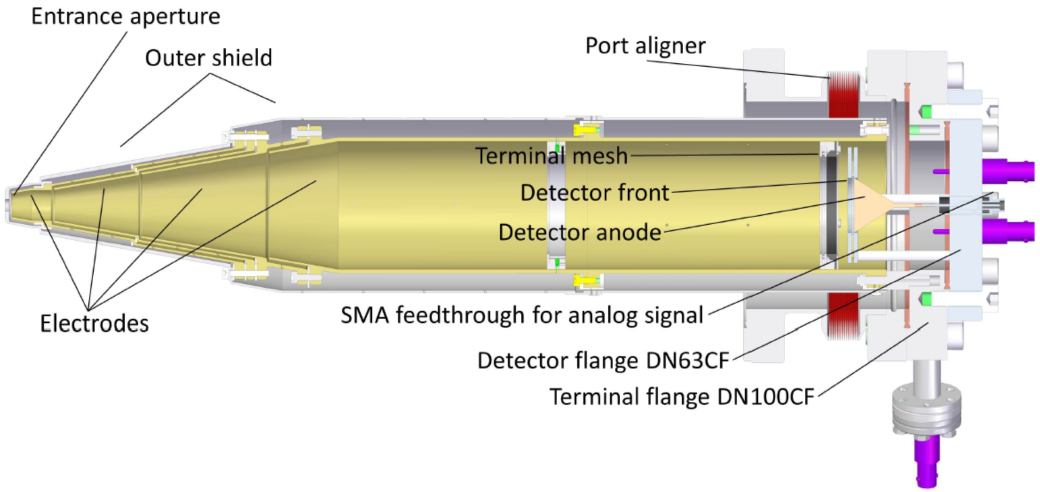
Electron-Ion coincidence spectroscopy at high-repetition-rate FELs has opened new avenues for monitoring molecular structures and can, in combination with atto-streaking imminently access electron dynamics with dissociation channel specificity!

Part of the Team of the Attocampaign

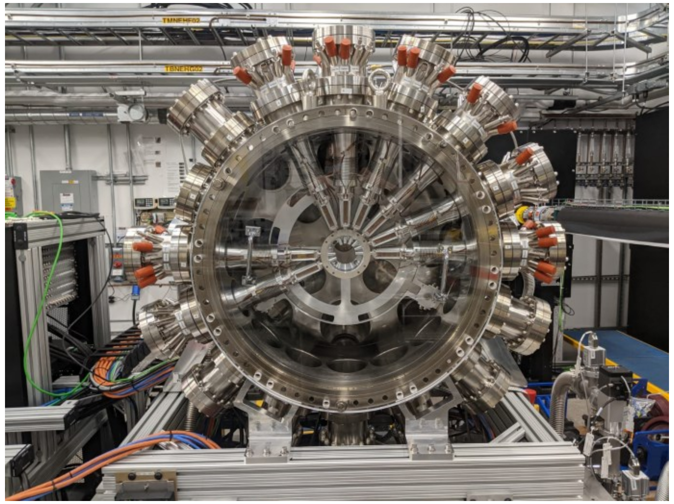
Plans for instrumentation upgrades

From diagnostic achievements to chirality science at the attosecond frontier in gas and liquid phase

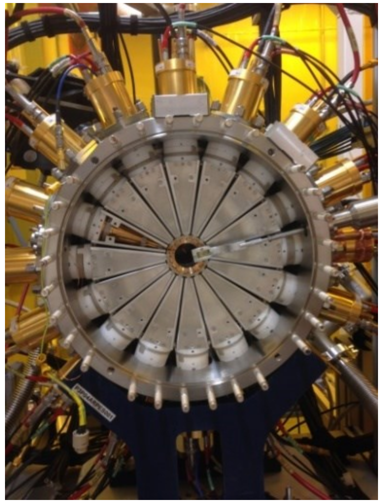


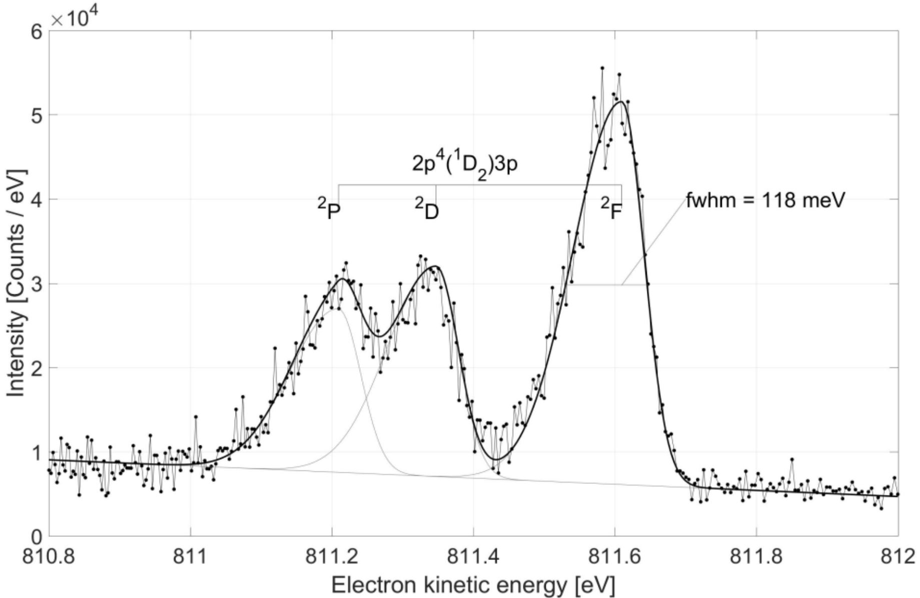


A. DeFanis et al. J. Synchrotron Rad. 29, 755–764 (2022)



P. Walter *et al*, J. Synchrotron Rad. **28**, 1364 (2021)





Next Generation:
SpeAR Project for atto-streaking
W. Helml et al.

Scientific Road of (X)FELs with Undulator-Based Polarization Control (for gas- and liquid phase studies)

- Technological state-of-the-art and future technological perspectives of polarization control at (X)FELs
  - FERMI
  - LCLS
  - FLASH
  - SwissFEL
  - SHINE/SXFEL
  - European XFEL
- Diagnostics and instrumentation
- Opportunities for ultrafast und nonlinear stereochemistry
- Nonlinear circular dichroism studies
- Ultrafast dichroic phenomena explored with reaction microscopes
- Exploring orbital angular momenta with twisted photons
- Perspectives for stereochemistry in complex molecules
- Theoretical Perspectives for Polarization-Controlled FELs
  - Atoms - Perturbative Methods, Single-Active-Electron Approximation, i.e., solve the TDSE in some potential, Multi-Electron Approaches like RMT, and Molecules
- Summary and Outlook