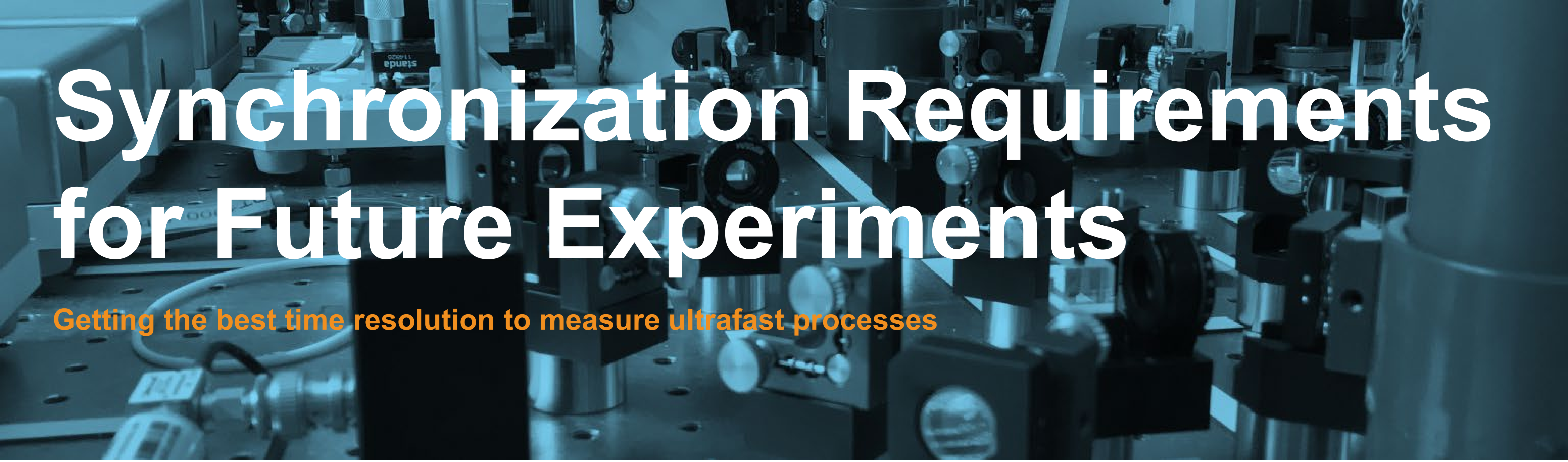


Synchronization Requirements for Future Experiments

Getting the best time resolution to measure ultrafast processes

Stefan Düsterer on behalf of FS-FLASH, FS/LA and **MSK**

Perspectives and Future Challenges in Optical and RF Synchronization Systems
Hamburg, Nov 14, 2023

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

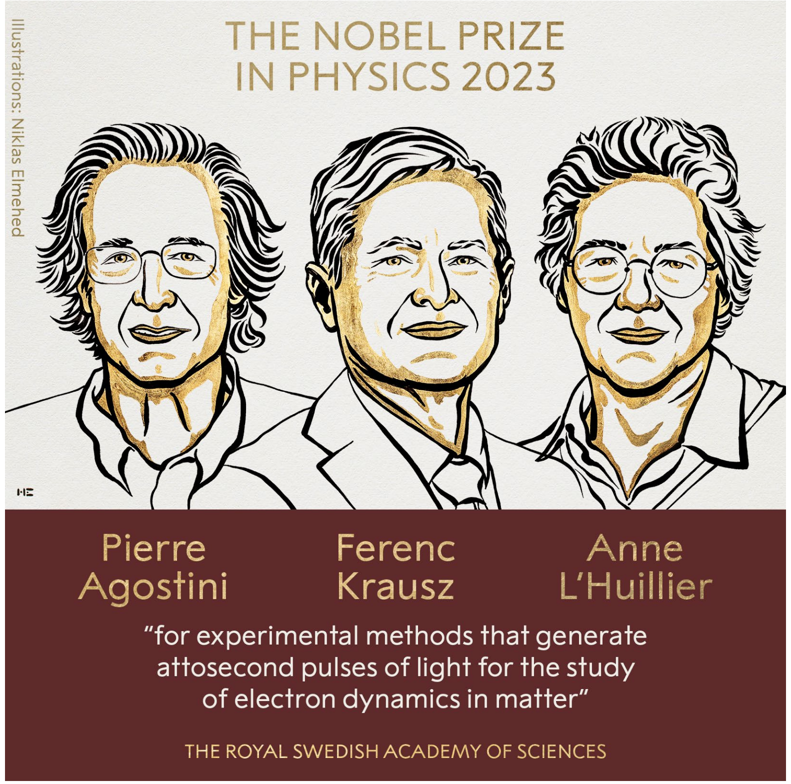
Outline

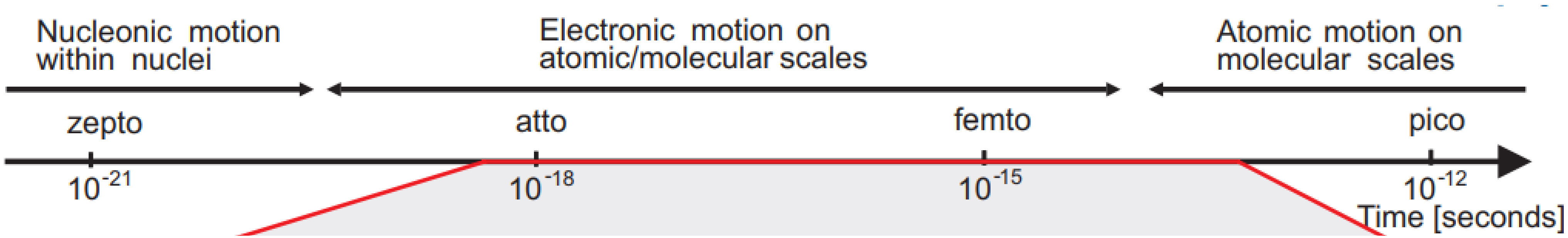
Synchronization requirements at FELs

- What to look at: Time scales and examples
- How to look: methods
- Present status at FLASH
- Thoughts about limits and experimental schemes to overcome them
- Try to answer questions …

Time scales …

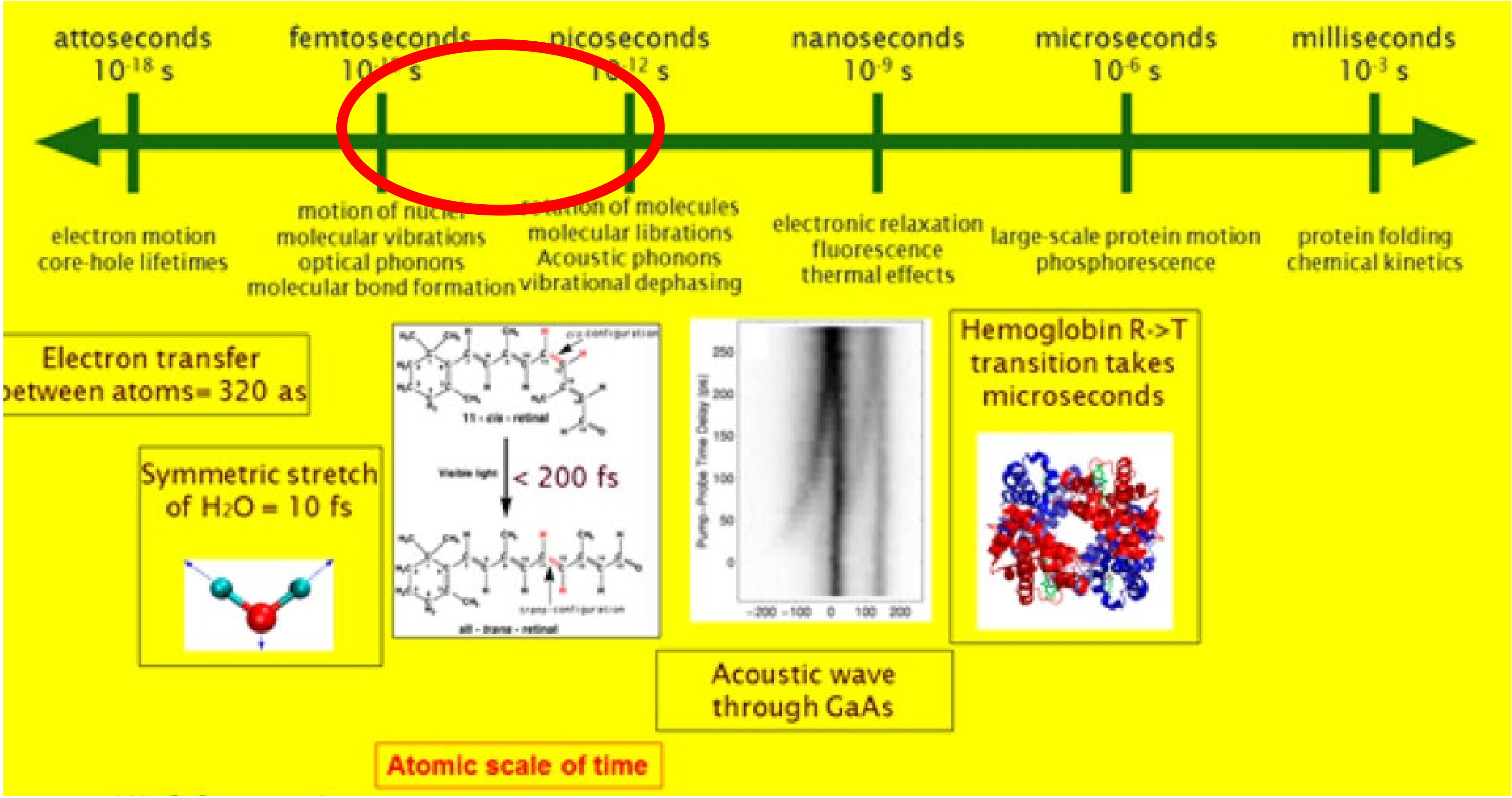
It never can be too short ;)





F. Krausz, M. Ivanov, REVIEWS OF MODERN PHYSICS, VOLUME 81, 2009

Time scales … overview

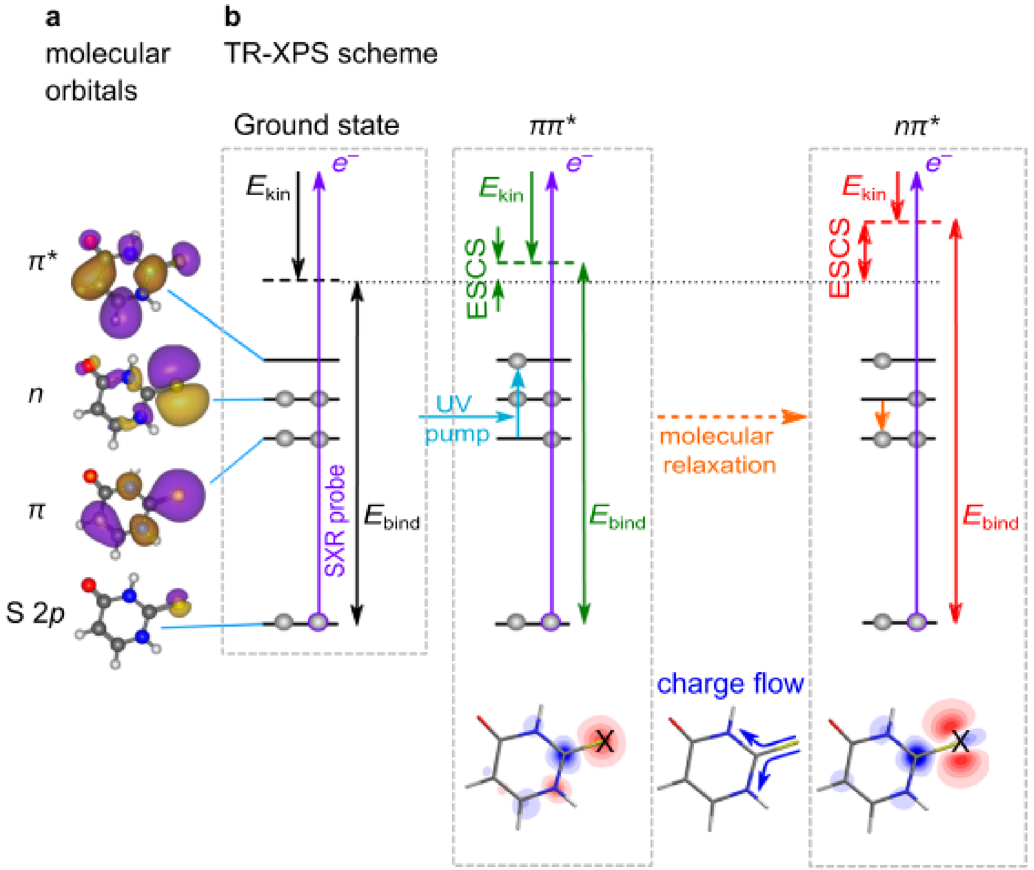


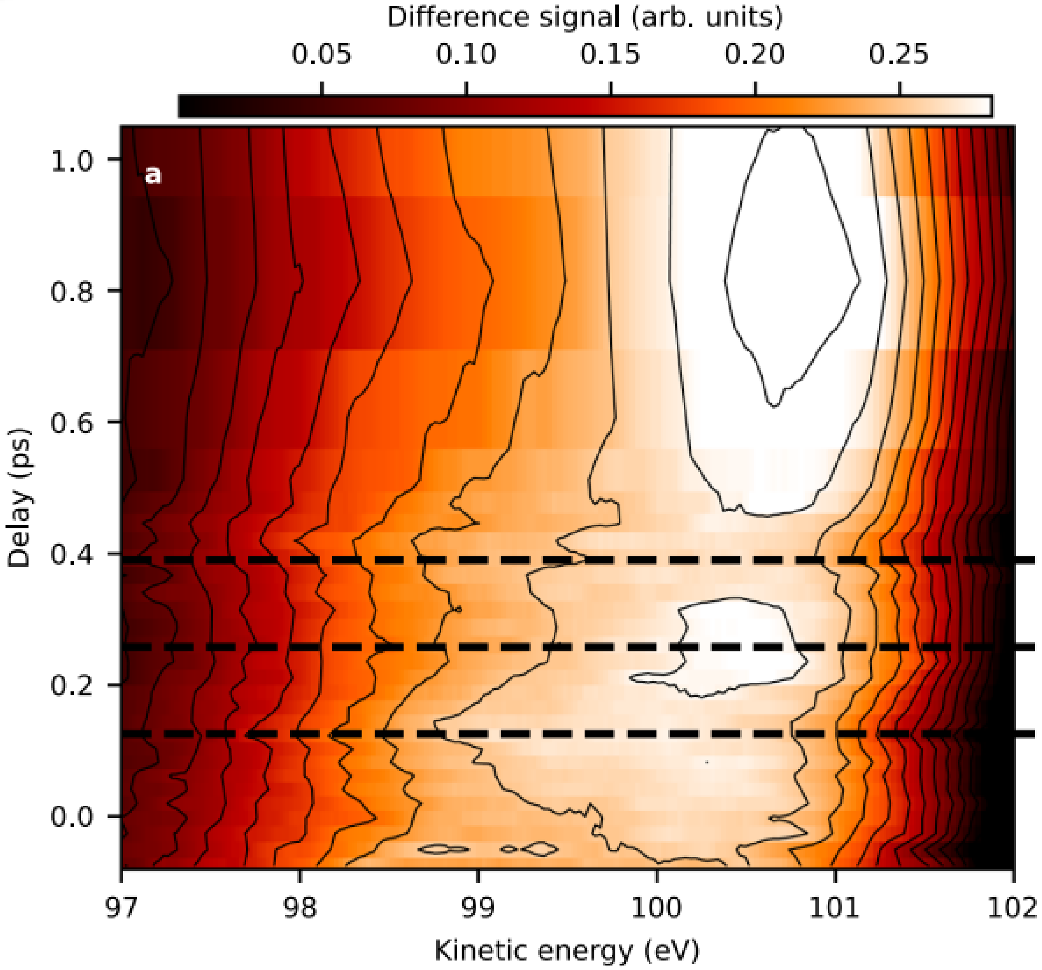
- *Chem. Rev.* 2017, 117, 16, 11025–11065

Examples from FLASH

Charge movement inside excited molecules

250 fs oscillations …



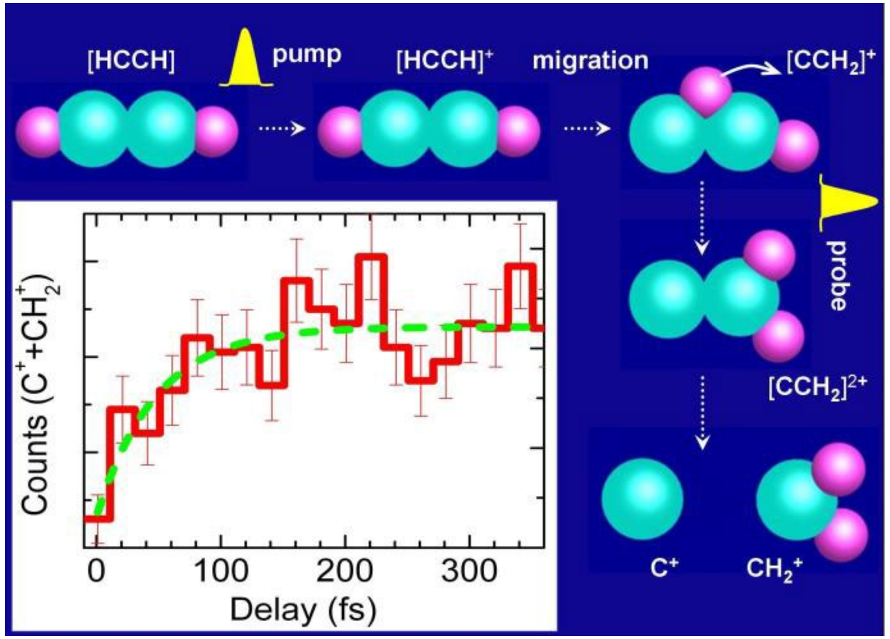


Nature Communications **13**, 198 (2022);

Examples from FLASH

Isomerization and electron coherence

~ 50 fs

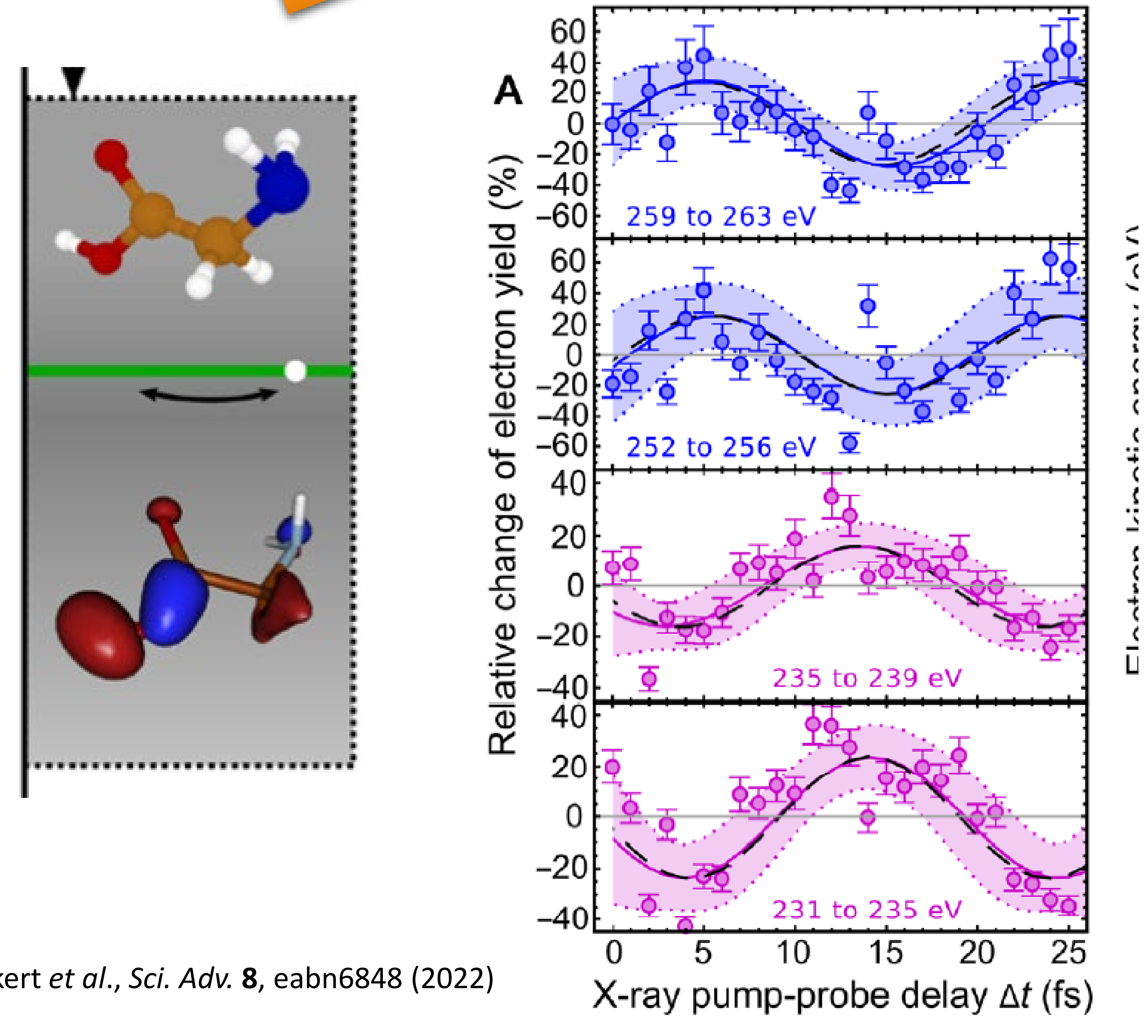


Isomerization from Acetylene to Vinylidene

(https://doi.org/10.1103/PhysRevLett.105.263002)

electronic coherence in photoionized biomolecules

~ 20 fs



Schwickert *et al.*, *Sci. Adv.* **8**, eabn6848 (2022)

Time resolved experiments … pump-probe

How to measure fast processes …

Pump: initiating the reaction
Probe: looks what happened

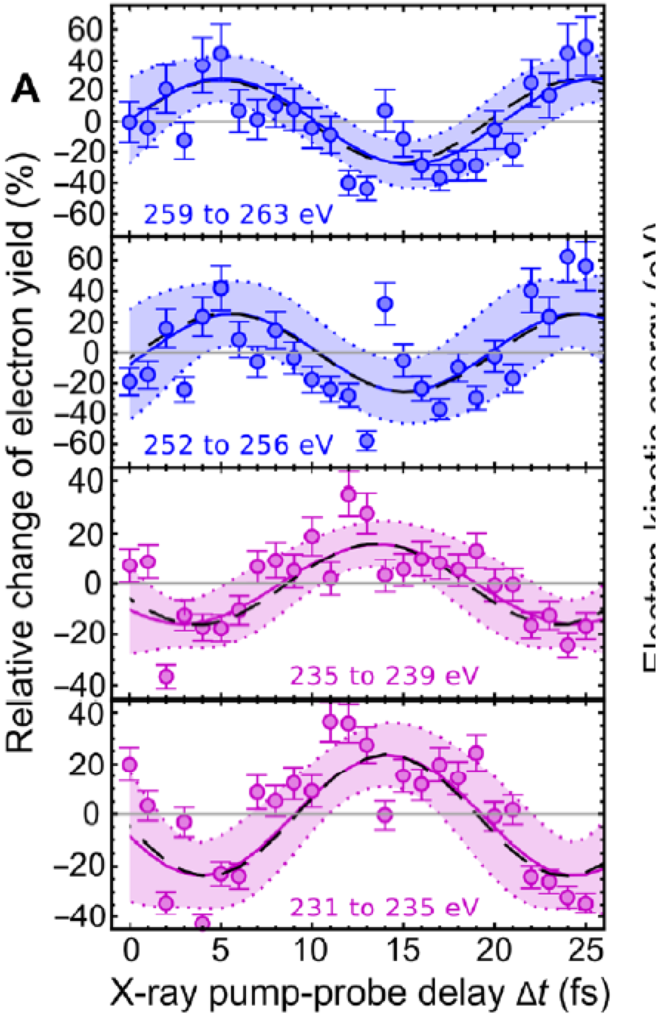
- FEL + FEL
- FEL + optical laser
- FEL + THz
- …

$$t_{res} \sim \sqrt{t_1^2 + t_2^2 + \sigma_{jitter}^2}$$

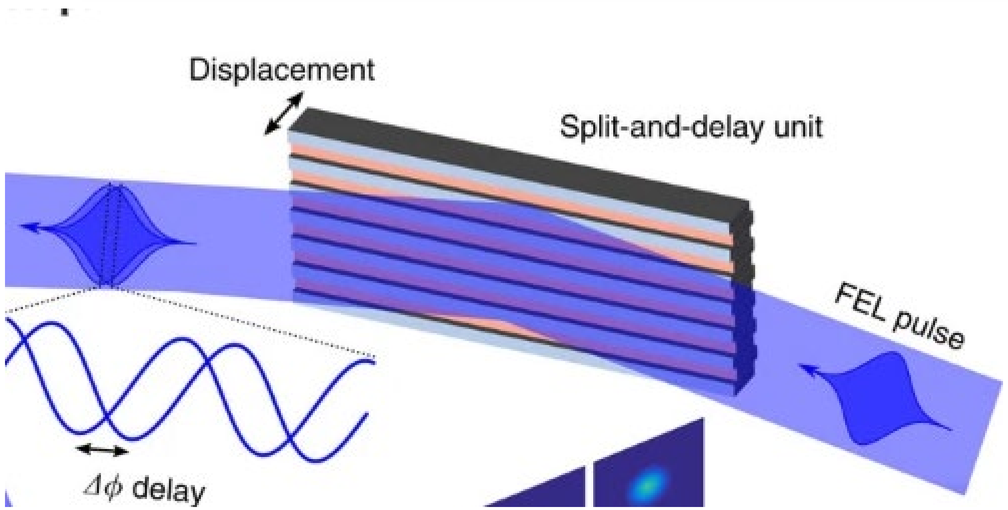
https://www.sciencephoto.com/media/618549/view/bullet-hitting-an-apple

Few fs dynamics observed at FLASH

FEL + FEL pulse



Split and delay units: down to few as resolution



Schwickert *et al.*, *Sci. Adv.* **8**, eabn6848 (2022)

$$t_{res} \sim \sqrt{t_1^2 + t_2^2 + \sigma_{jitter}^2}$$

Pump probe with 2 independent sources

FEL - Optical Laser Experiments

$$t_{res} \sim \sqrt{t_{FEL}^2 + t_{Opt-Las}^2 + \sigma_{jitter+drift}^2}$$

- **Pulse duration:**
  - PP Laser: ~15fs – 100 fs … relatively easy to diagnose
  - FEL: < 10 fs – 300 fs … well .. THz streaking / TDS

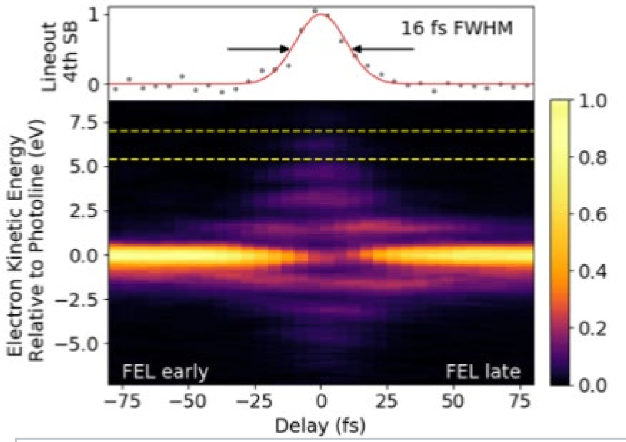
- **Jitter + Drift**
  - Synchronization system
    - Produce synchronized electrons and laser pulses
  - Electron acceleration … BAM (Beam arrival time monitor)
  - Laser amplification and transport … LAM (Laser arrival time monitor)
  - FEL process + XUV transport
  - Environmental changes (temp, humidity, air pressure …)

Temporal resolution at different FELs

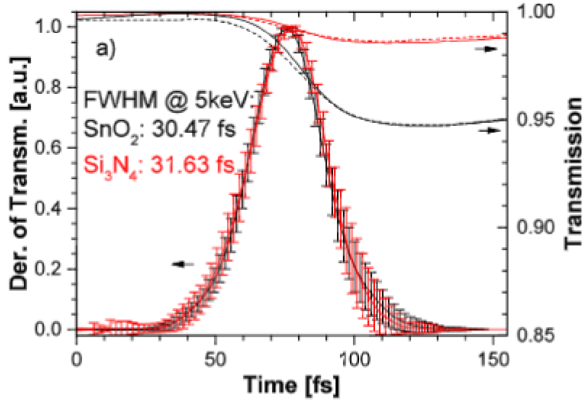
Few examples

- **XFEL: sub 20 fs** very short FEL and optical pulses

  (no jitter sorting)



Rivas et al Vol. 9, No. 4 / April **2022** / Optica 430

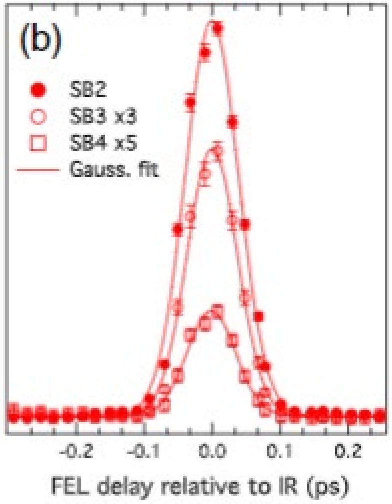


Appl. Phys. Lett. **113**, 114102 (2018)

- **LCLS: usually ~60fs** (with sorting)

  with very short optical pulses **30 fs:**

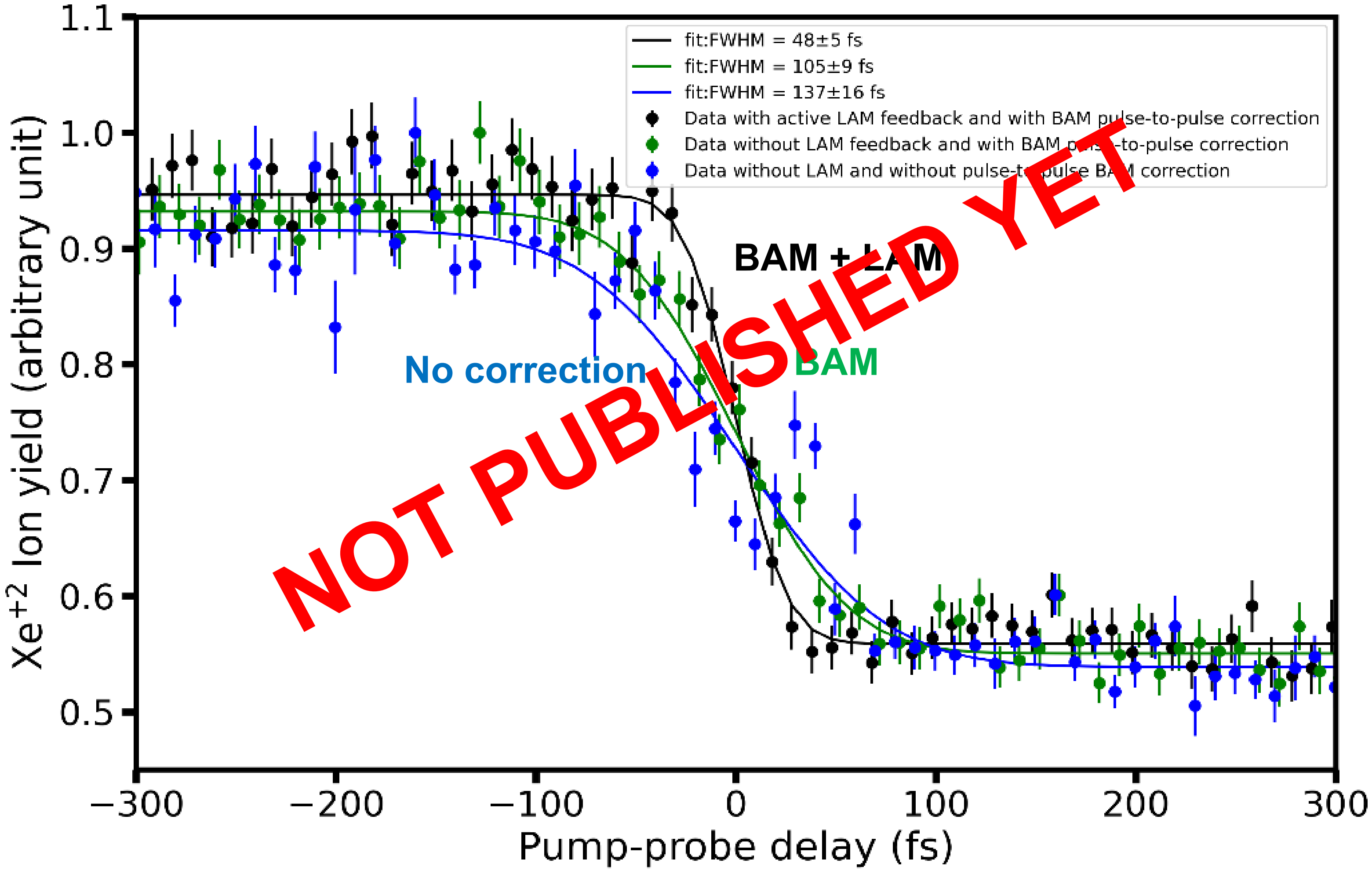
- **Fermi: ~ 100 fs** … very low jitter (5 fs), limited by optical pulse duration



P FINETTI et al. PHYS. REV. X 7, 021043 (**2017**)

Resolution measurement at FLASH

"Towards Pulse-Length Limited Temporal Resolution"



FEL: 7.7 nm, ~ 20 fs (FWHM)
Laser 800 nm , 18 fs (FWHM)

-> pulse duration limit ~ 30 fs

+ Jitter: ~ 30 fs (FWHM) / 12 fs (rms)

-> ~ 50 fs FWHM resolution
(~ 20 fs rms)

$$\sigma_{jitter} = \sqrt{(\sigma^2_{OptRef} + \sigma^2_{BAM} + \sigma^2_{SASE} + \sigma^2_{LAM-jitter})}$$

Submitted – curtesy A. Tul Noor

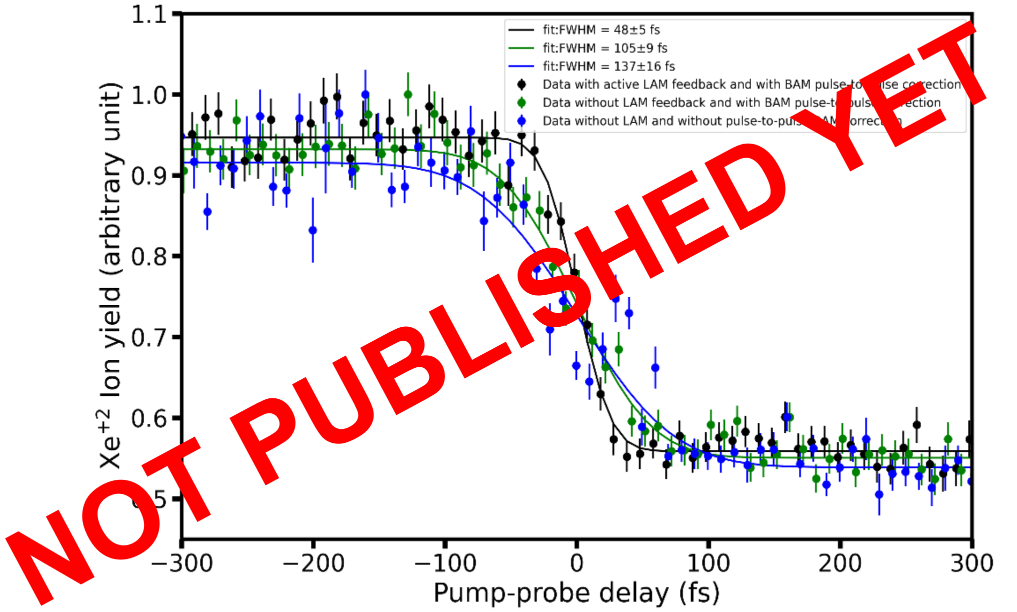
How to achieve good temporal resolution

Different steps of correction

- **~140 fs** with **no correction** (measurement time 1 h) … already very good …

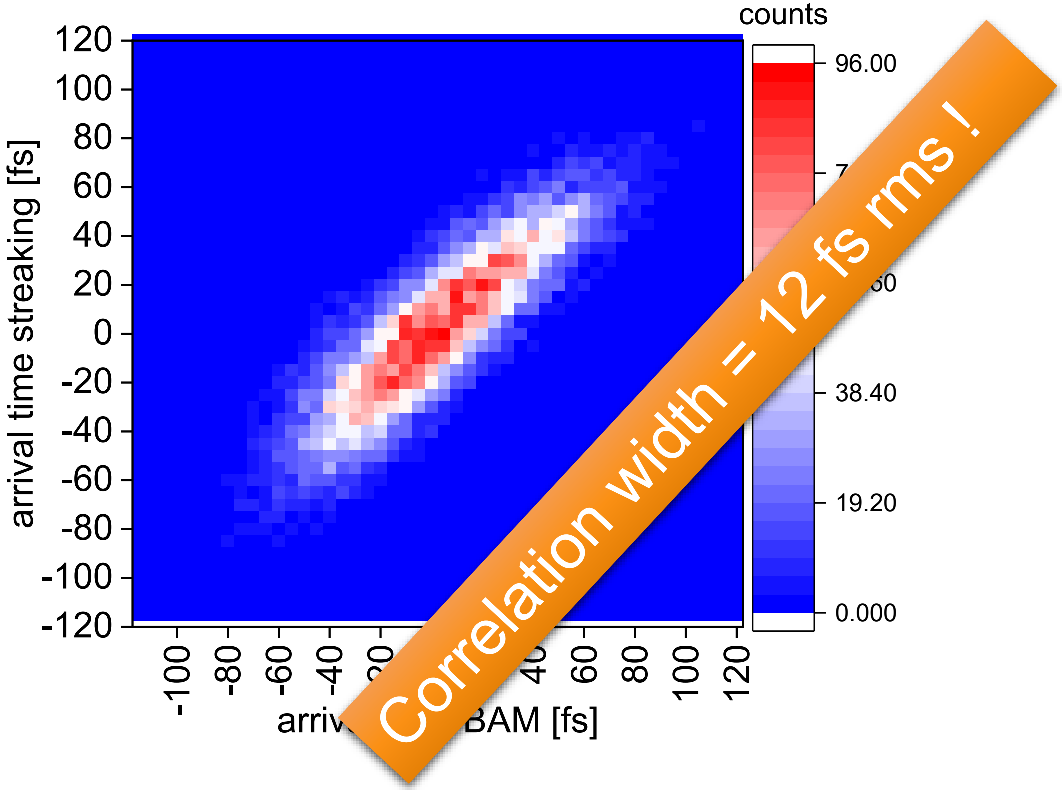
- **~100 fs** with **FEL arrival time** correction (BAM)
  - The arrival time of each electron bunch is corrected (post processing)

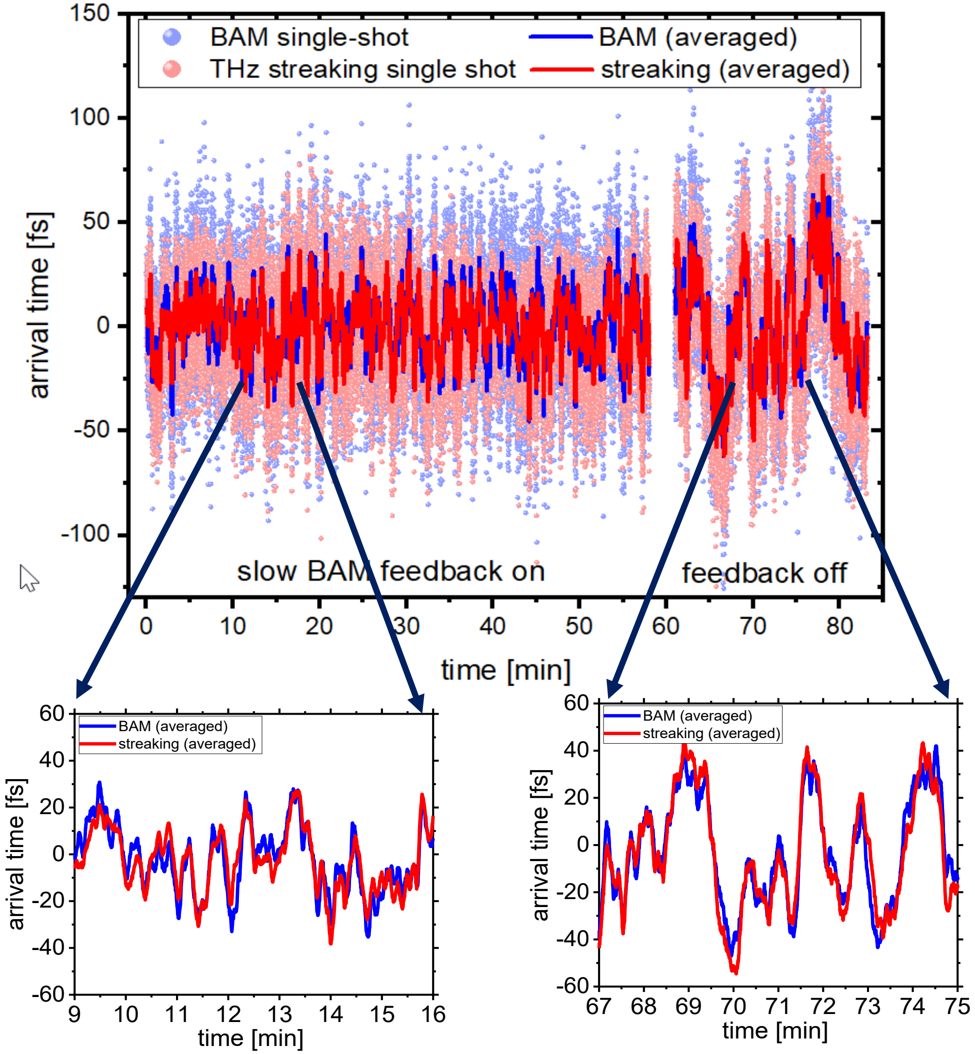
- **~ 50 fs** with BAM and **laser arrival time** correction (LAM)
  - The laser drifts / jitter are measured and corrected for (feedback)



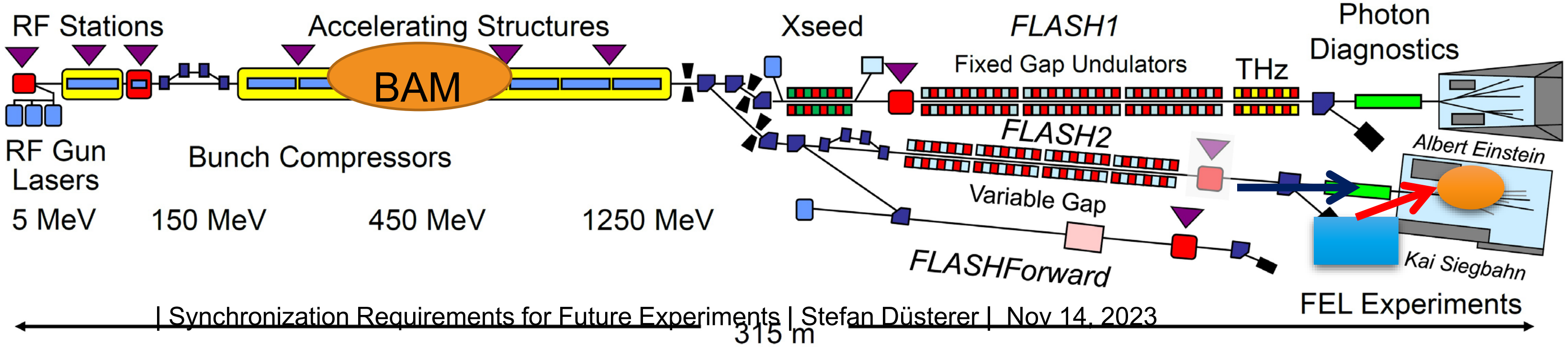
Arrival time measurements - BAM

Photons and electrons are well correlated



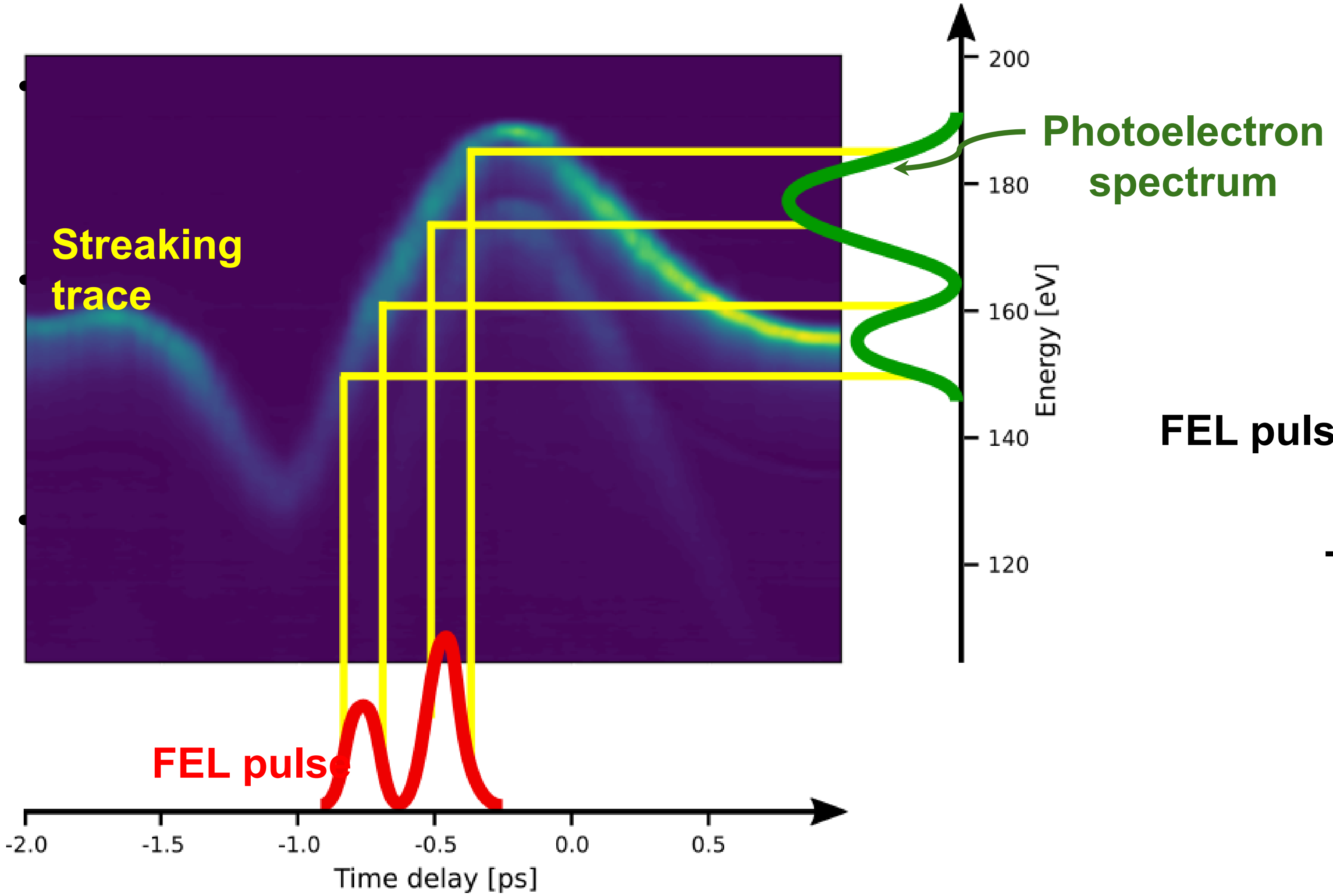


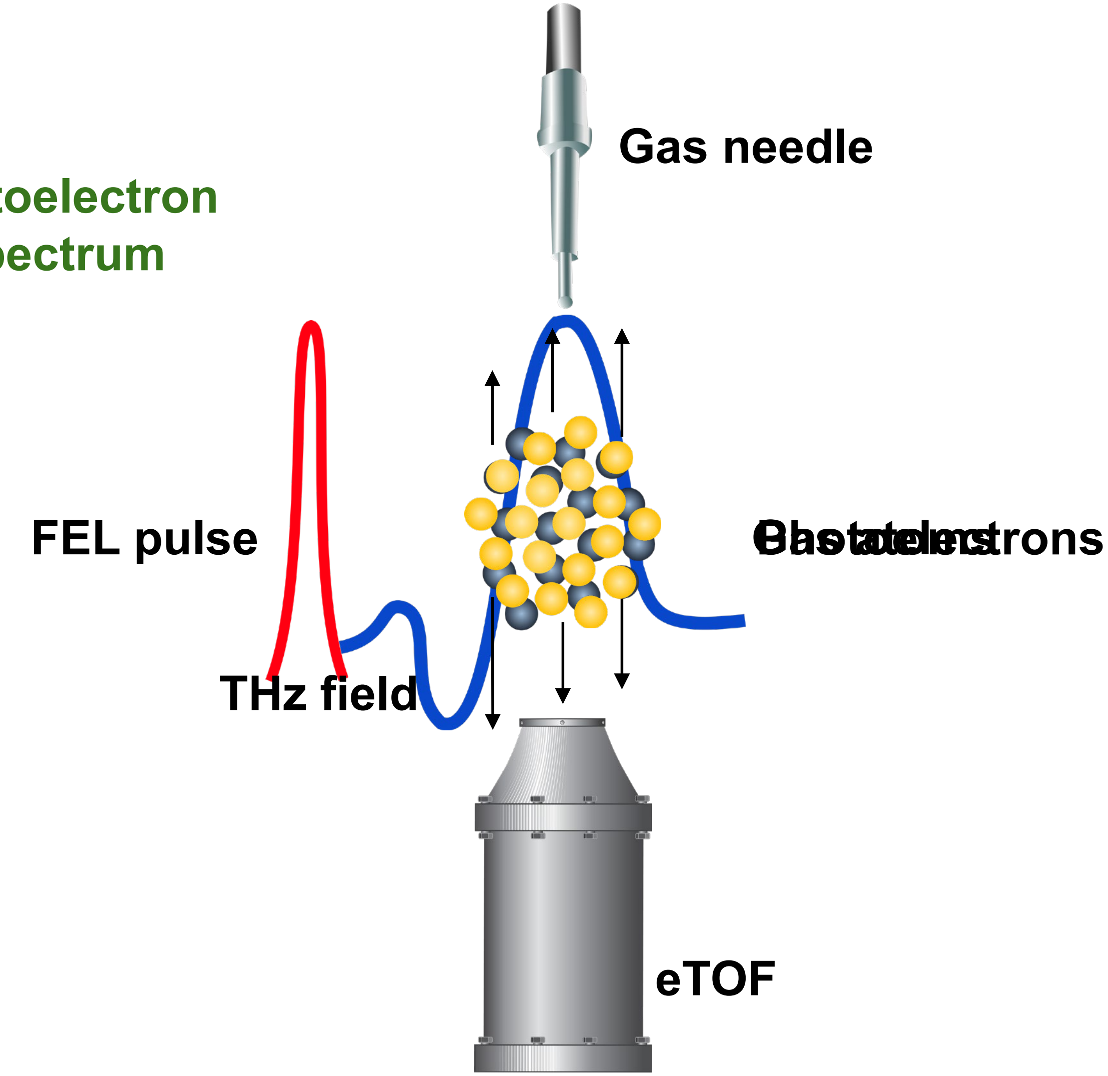
R.Ivanov et al, Opt. Express **31**, 19146 (2023)



THz streaking principle

How to measure x-ray pulse duration and arrival times





U. Frühling et al. Nat. Photonics 3, 523 (2009)
I. Grguraš et al. Nat. Photonics 6, 852 (2012)

Laser transport drift correction

LaserArrivaltimeMonitor LAM

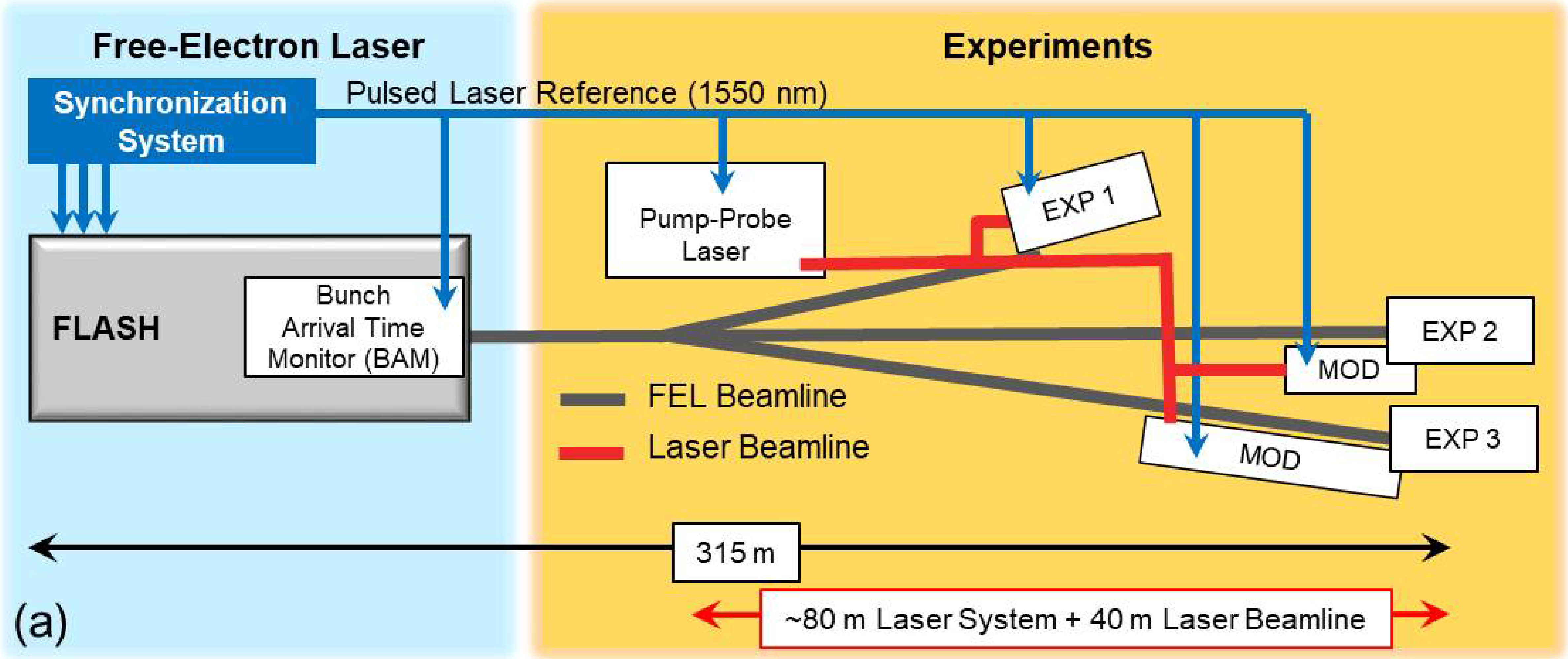
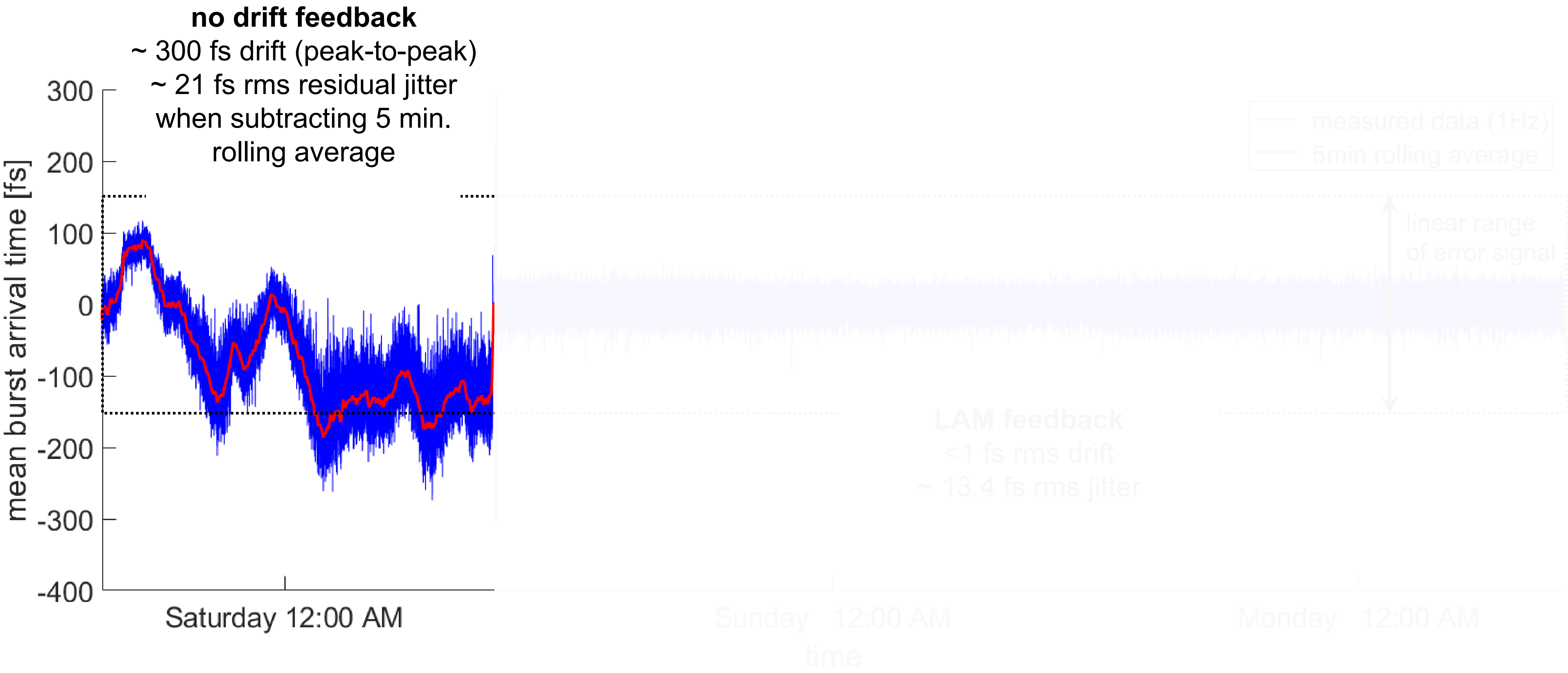


Image courtesy of N. Schirmel

LAM Feedback

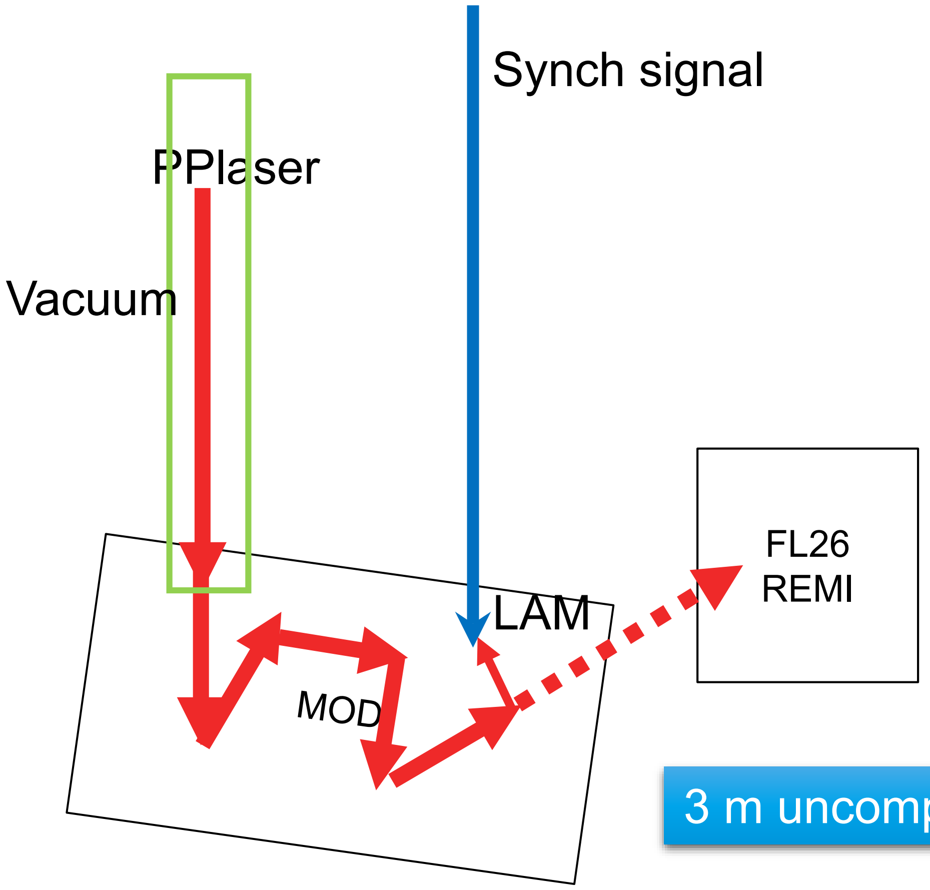
Significant improvement of the timing stability

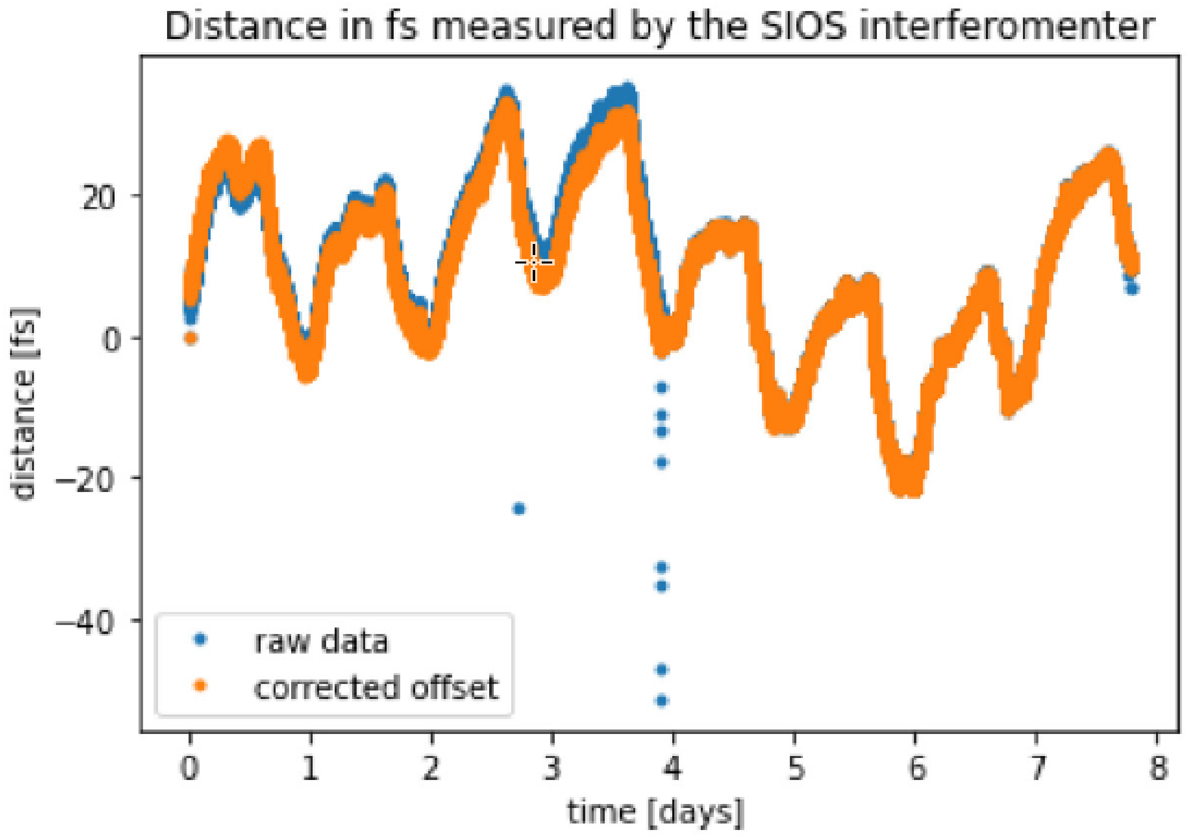


slide courtesy of Nora Schirmel

Remaining „few meters“

Understanding residual drifts





3 m uncompensated to Endstation

Thoughts about limits

ALL 3 parameters have to be looked at !!!

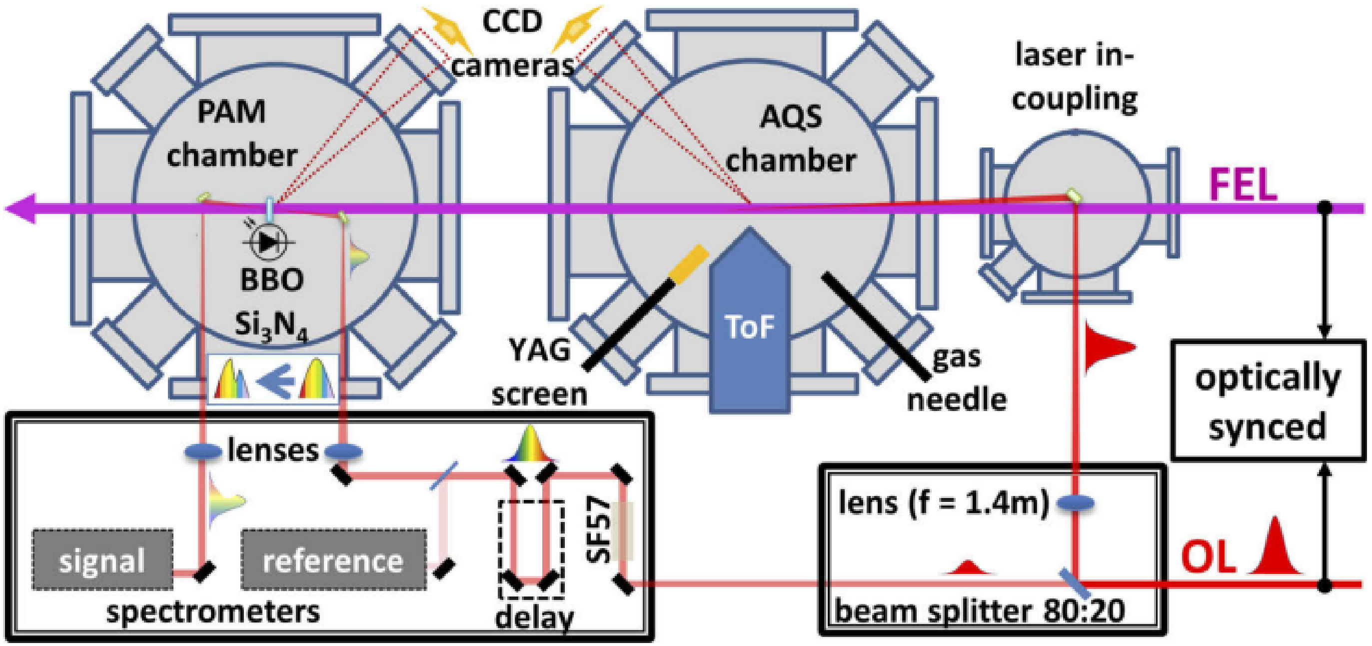
$$t_{res} \sim \sqrt{t_{FEL}^2 + t_{Opt-Las}^2 + \sigma_{jitter}^2}$$

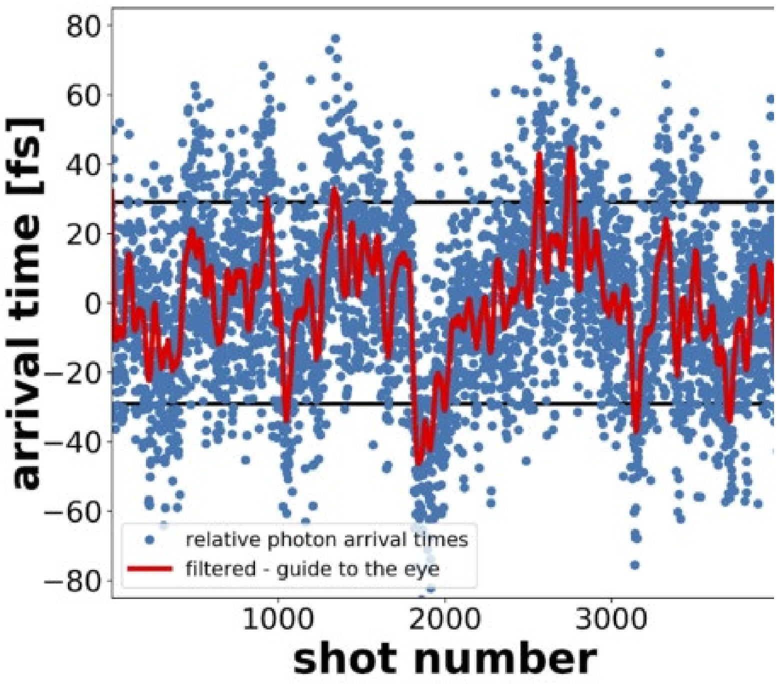
- Pulse duration:
  - PP Laser: **< 10-15 fs** is hard for vis / IR
  - FEL (FLASH): shortest pulses **few fs**
    - possible to beat the coherence time (Photonics **10**, 653 (2023))
    - Needs reliable measurement

- "Timing"
  - Synchronization – already in the **few fs** range
  - **Use BAM and LAM** … will be hard to get to sub 10 fs timing stability for longer times
  - To get better: Measure **IN or very close to the experiment** the actual relative timing
    - Use info to improve "timing"
    - No general solution for all experiments in the XUV ☹
    - "self referencing experiments"

Direct measurement of X-ray and IR arrival times

Improve the resolution even more …



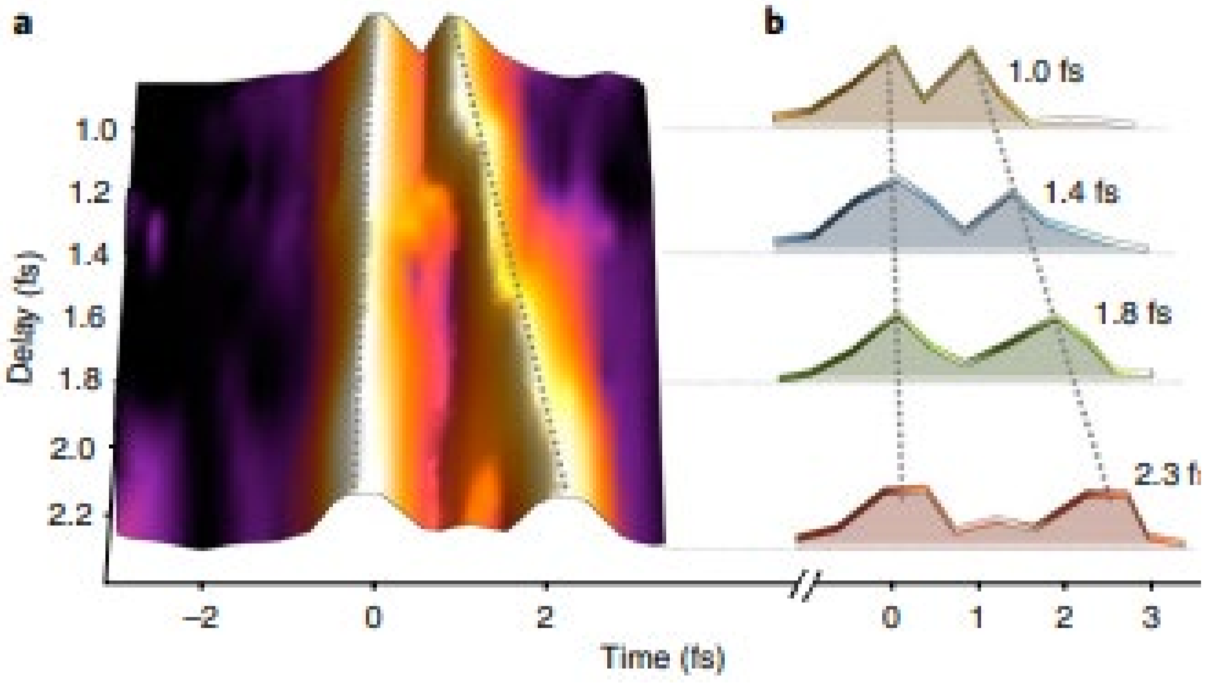


- Current resolution few fs
- Often used at X-ray FELs
- Problematic at XUV

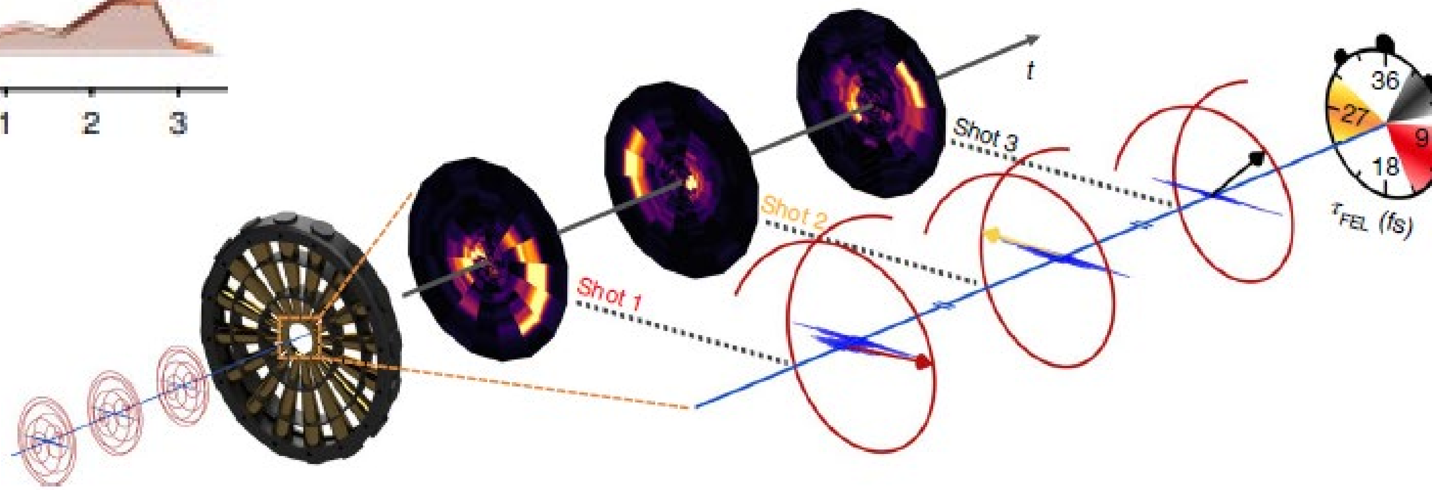
Vol. 29, No. 23 / 8 Nov 2021 / Optics Express 37429

Self referencing experiments – measure SASE pulse shapes

Example from LCLS: Measure and sort …



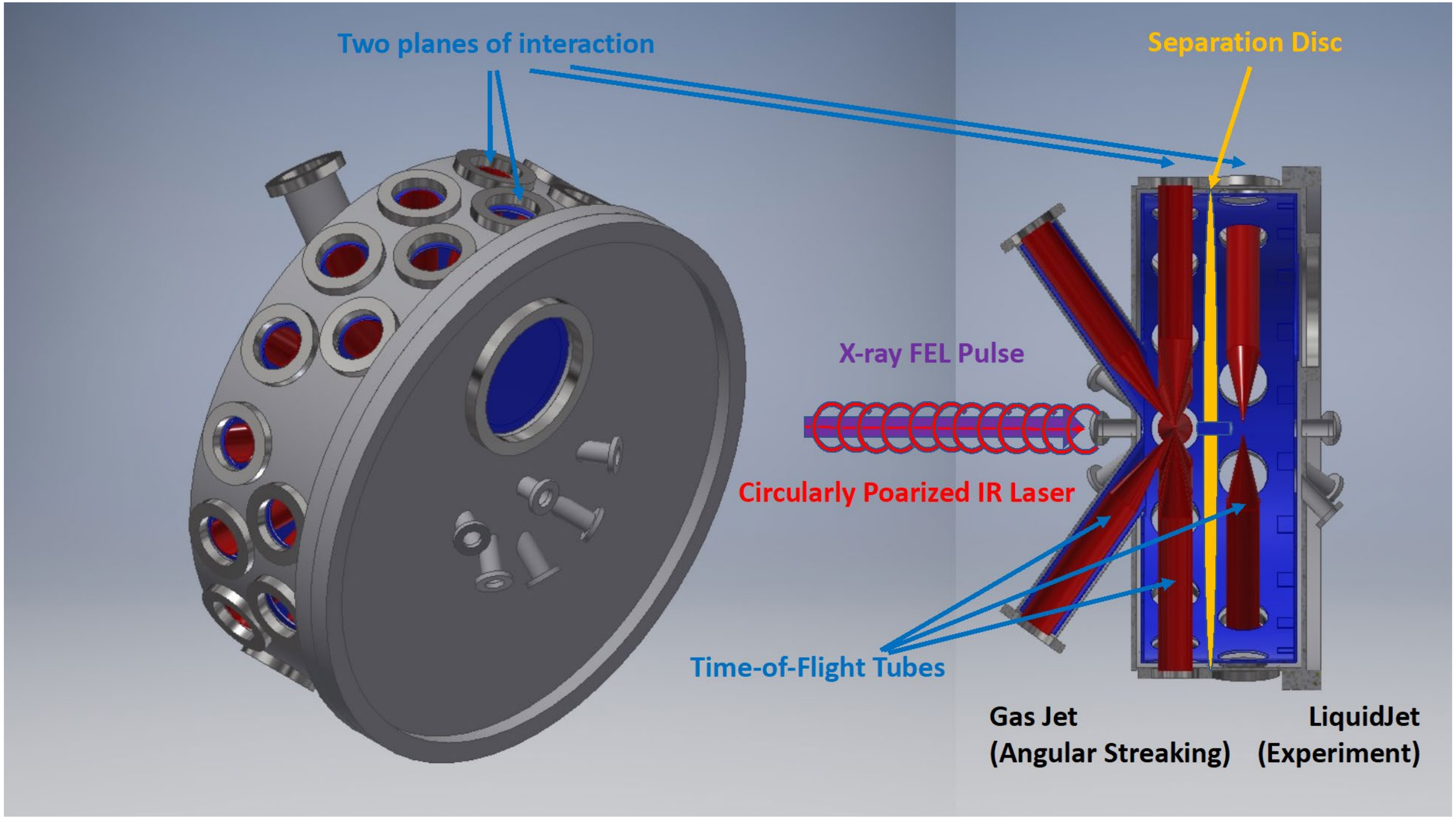
Angular streaking – attosecond resolution



Nature Photonics | VOL 12 | APRIL 2018 | 215–220

Combine experiment and diagnostic

SPEAR chamber in preparation for FLASH

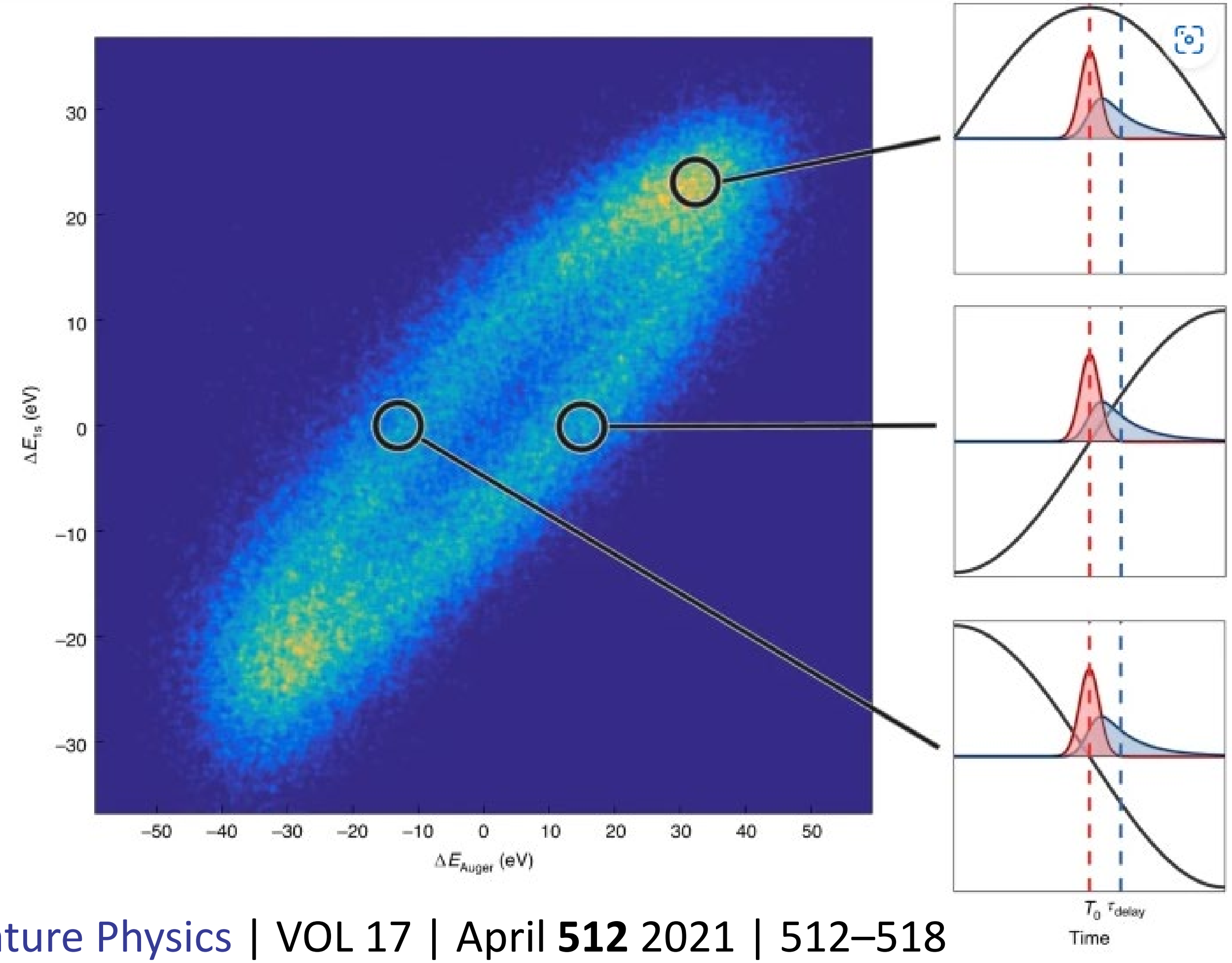


Courtesy M. Ilchen and W. Helml

Self-referenced streaking

Get the relative timing between FEL and an IR pulse by the experiment …

Clocking Auger electrons



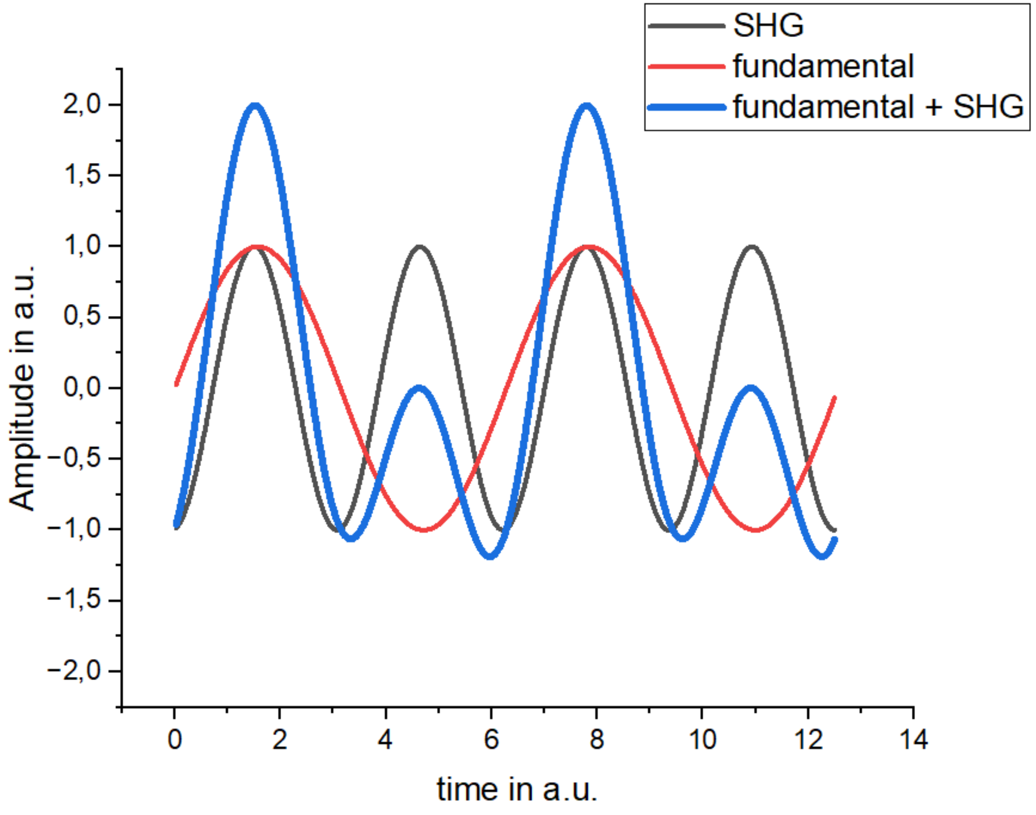
Nature Physics | VOL 17 | April **512** 2021 | 512–518

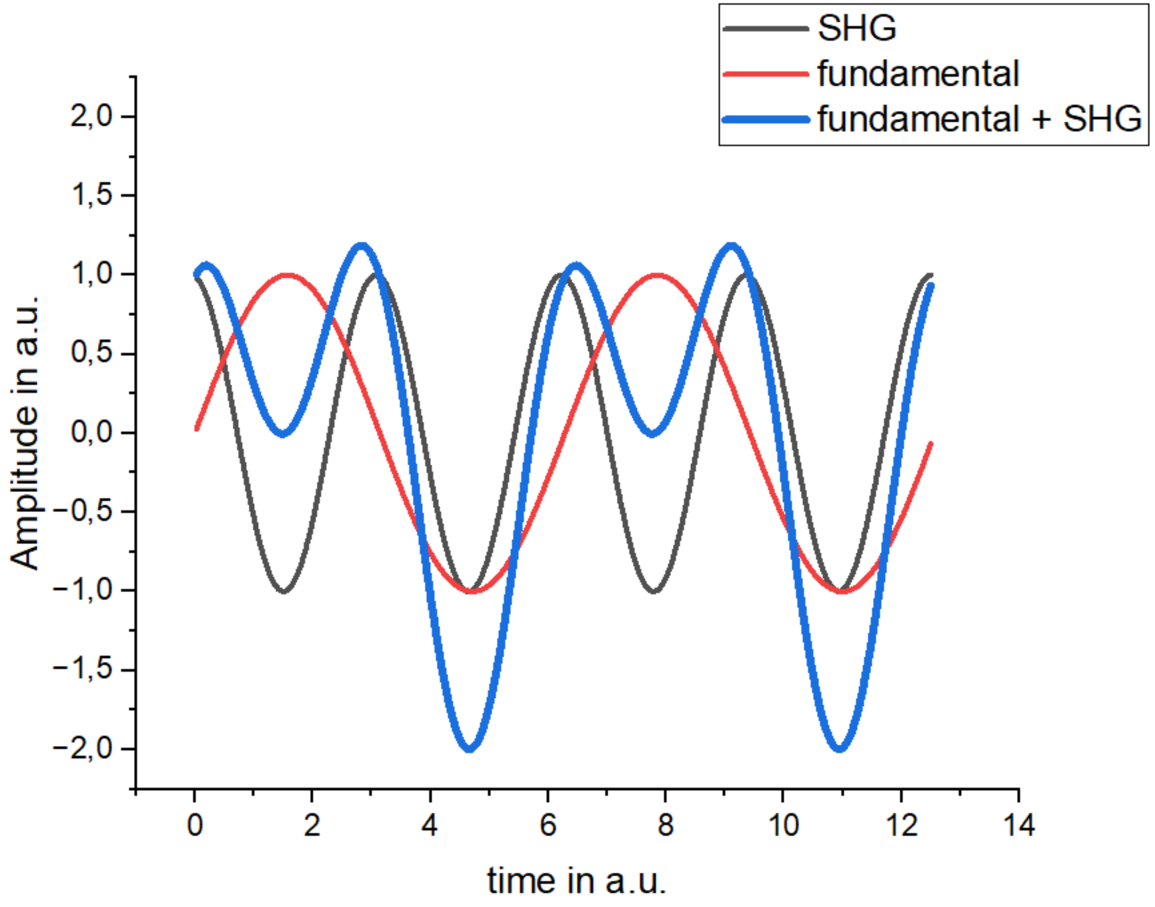
"self-referenced streaking will enable experiments to take advantage of the extreme-intensity X-ray pulses at XFELs while simultaneously exploiting the unrivalled time resolution provided by attosecond streaking spectroscopy."

Seeding – from fs to sub wavelength shifts: play with phases

Phase shift between fundamental and second harmonic

- Changing the phase between fundamental and second harmonic (10 nm ~ 33 as)

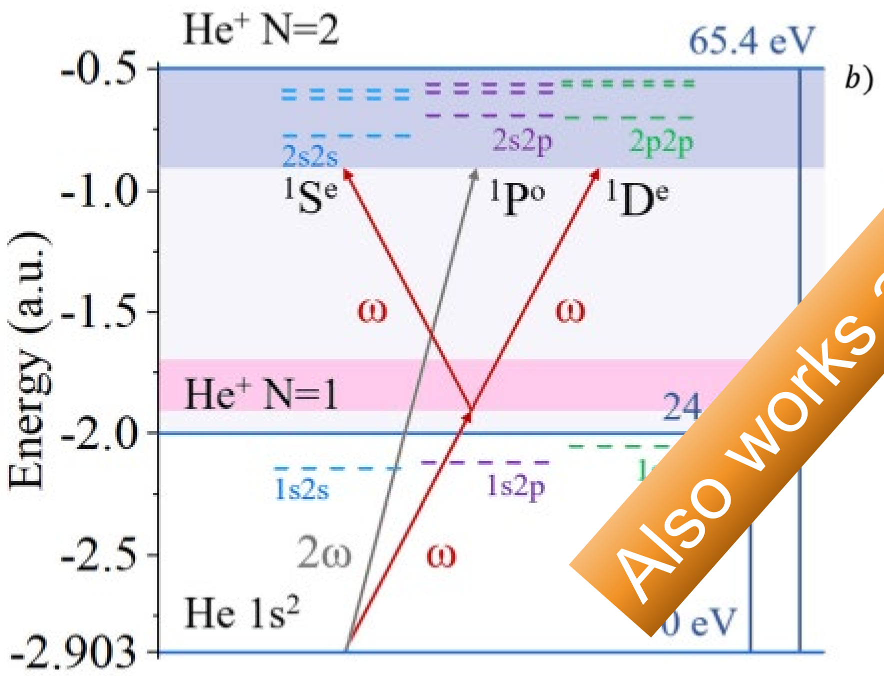


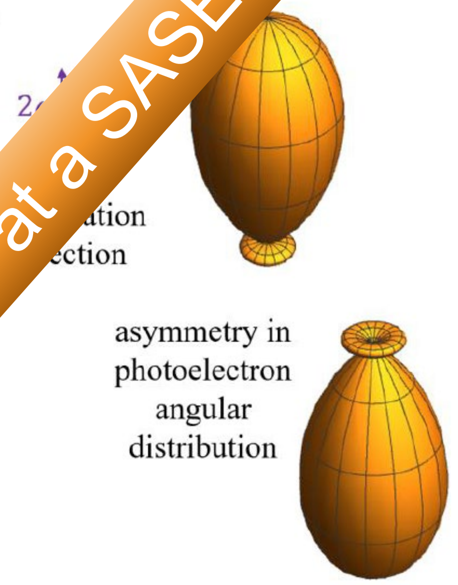


Seeding – play with harmonics

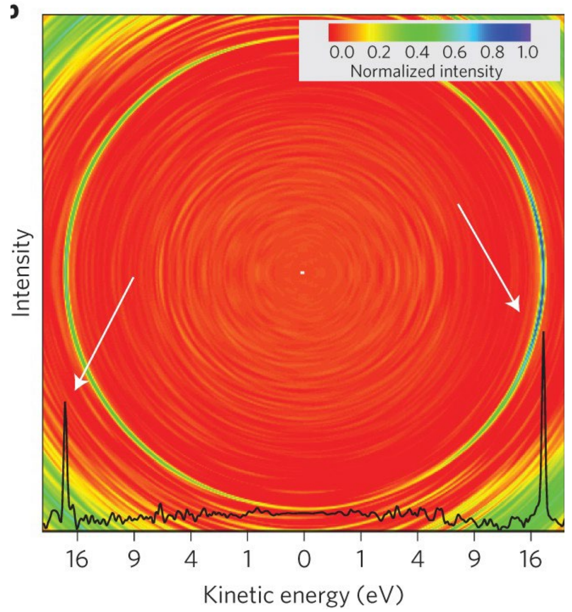
Phase shift between fundamental and second harmonic

Also works at a SASE FEL !!!

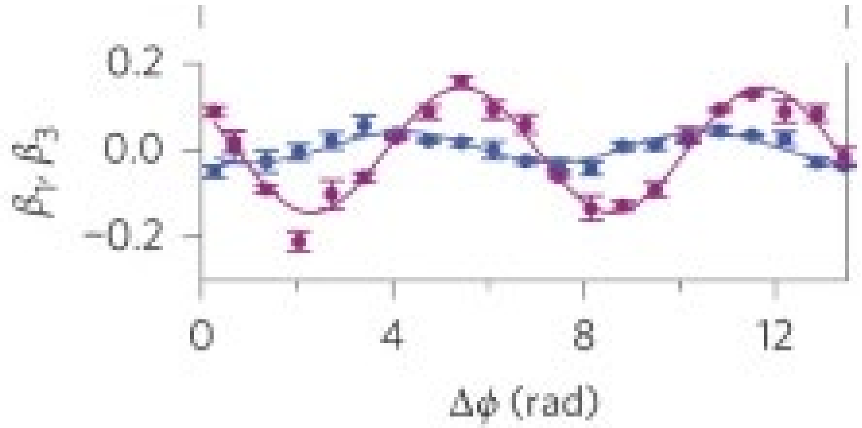


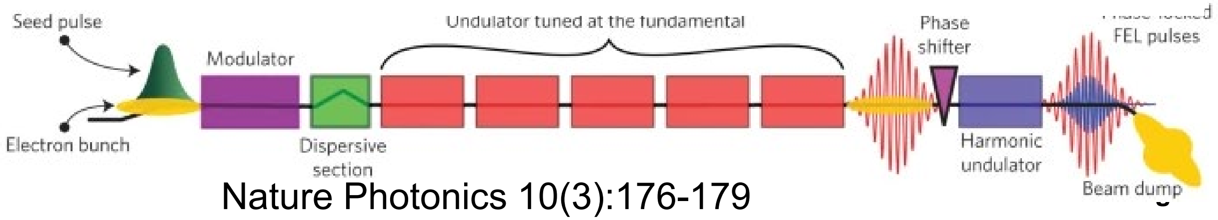


PHYSICAL REVIEW A 103, 053118 (2021)



Step size: 0.056 rad (900 zs)





Nature Photonics 10(3):176-179

Harmonics and optical laser

attosecond timing tool

Single-shot technique to determine the relative synchronization between an attosecond pulse train and a near-infrared field,
with a resolution of one atomic unit (24 as).

Nature Photonics volume 17, pages200–207 (2023)

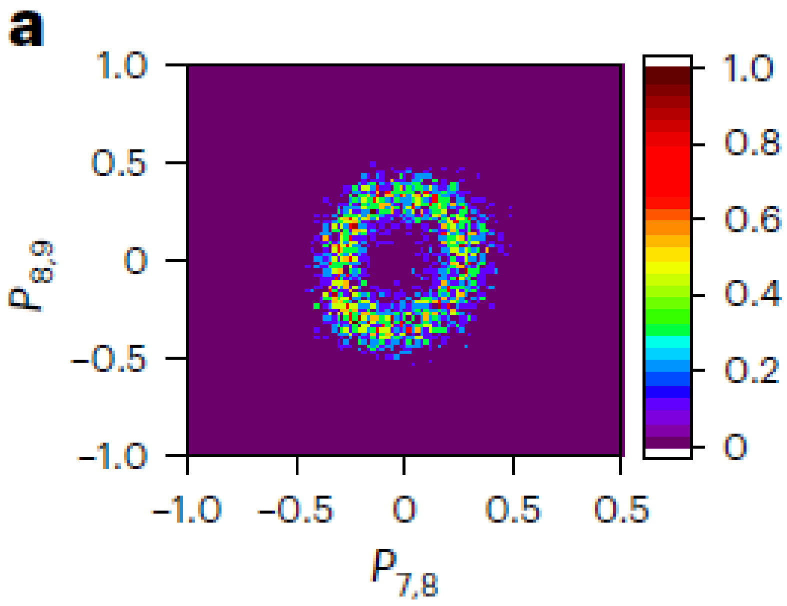
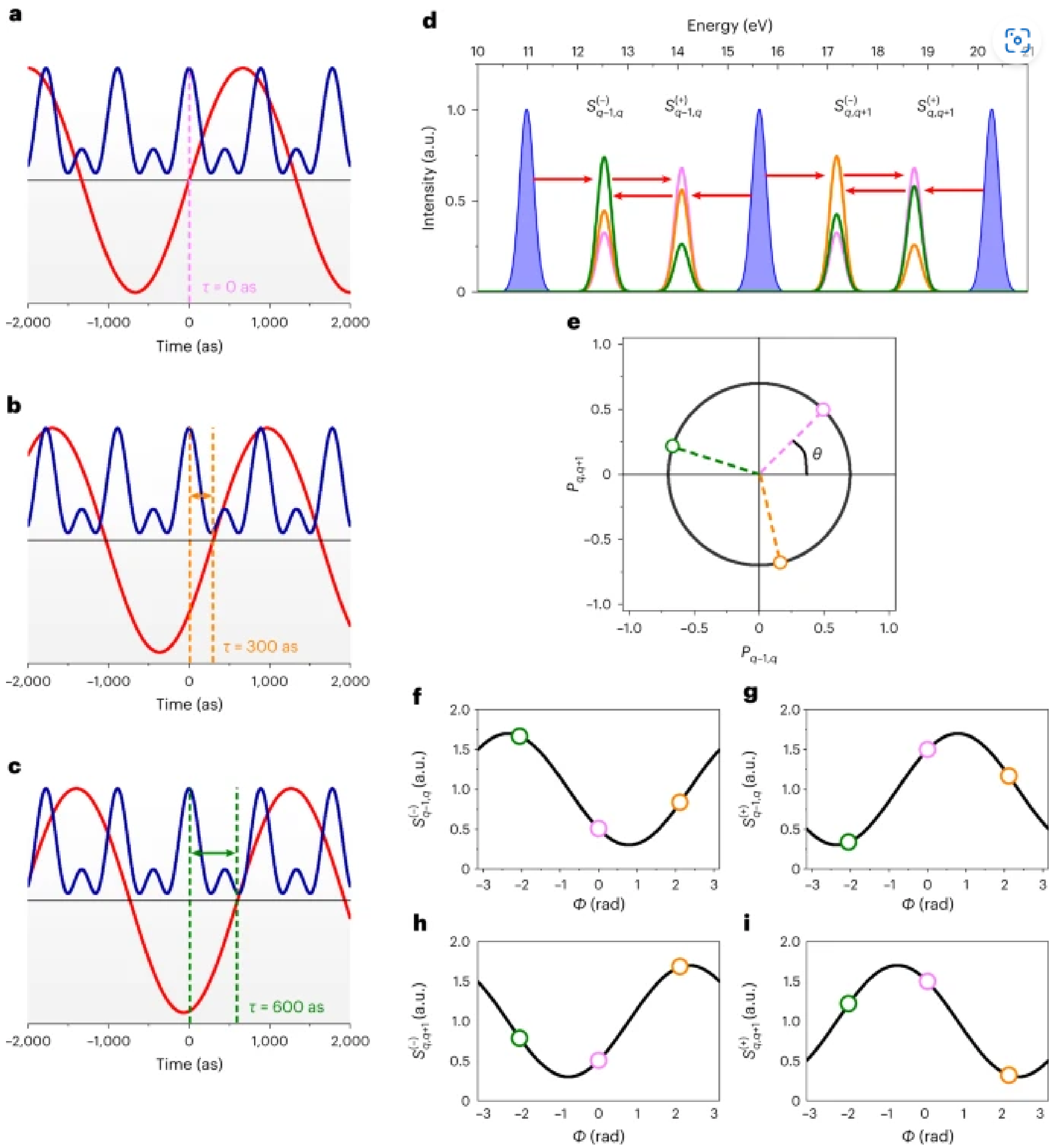


Fig. 1: Principle of the attosecond timing tool.



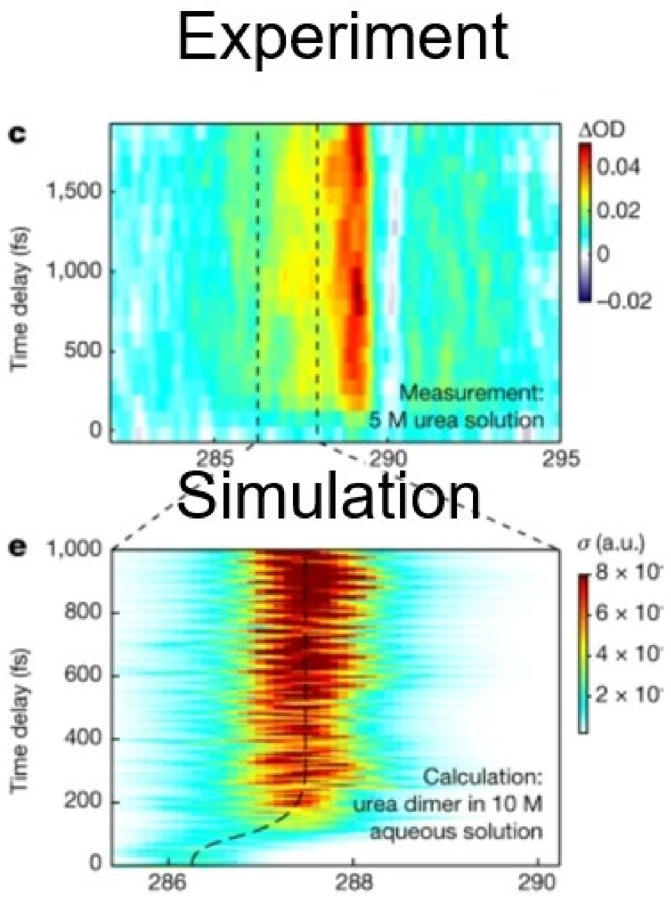
Resolution wish list – experiment view

Results from a quick poll …

- AMO:
  - 10 – 50 fs resolution (FWHM) look at femto-chemistry
  - Few fs or better for atomic processes …

- Solid state: typically 100 fs
  - 20-50 fs e.g. resolve optical phonons
  - 10-50 fs to look at molecules on surfaces …

Note: for “optical” pulses:
1 oscillation at 500nm are 1.7 fs
10 fs short pulse has already ~0.1 eV bandwidths



Nature volume 619, pages749–754 (2023)

Questions for the workshop

And some ideas for answers

- Current limits …
  - FEL + optical laser : ~ 20-30 fs (FWHM) - special cases much better …
  - FEL + FEL … well … few fs to sub fs
- Requirements for future synchronization
  - Jitter and drift in the few fs range …

feedback is much better than post sorting

  - Look at pulse durations AND synchronization
  - Robust …
- New developments needed
  - Accurate measurements of the relative arrival time at the experiment – non invasive …
  - Measure accessible parameters and predict timing -> ML
- Cooperation
  - Closer cooperation between “photon” and “electron” side

Thanks

for your

attention