Development of Two-Color Sub-Femtosecond Pump/Probe Techniques with X-ray Free-Electron Lasers

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U.S. DEPARTMENT OF ENERGY
Office of Science

SLAC NATIONAL ACCELERATOR LABORATORY

Acknowledgements

SLAC

XLEAP/XLEAP-II Collaboration:

SLAC

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PULSE: J. Cryan, P. Bucksbaum, E. Champenois, T. Driver, J. O'Neal, A. Wang

ANL: A. Zholents, J. Xu, M. Qian

Attosecond Campaign

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PSI
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Attosecond Core Group at SLAC

Principal Investigators


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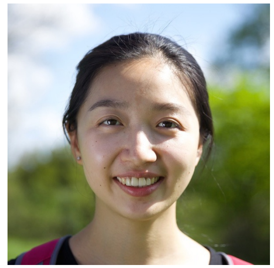
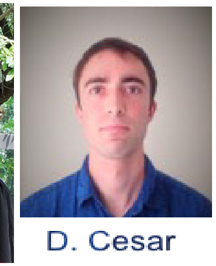
J. Cryan


S. Li


J. Duris

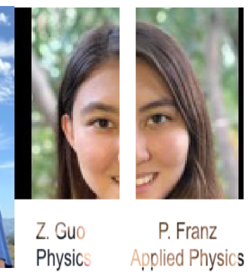

Z. Zhang



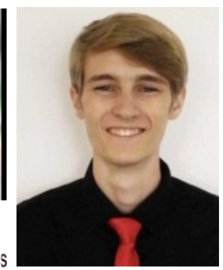
D. Cesar


N. Sudar


Z. Guo



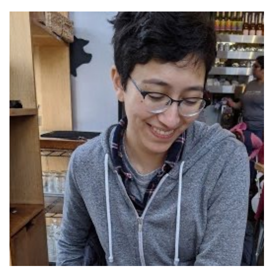
P. Franz


R. Robles

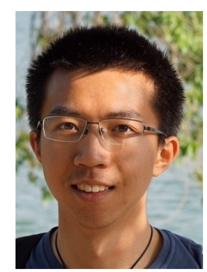

K. Larsen

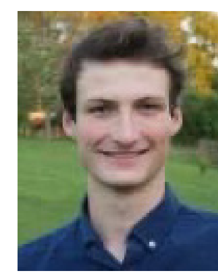

D. Garratt


T. Driver


A. Wang

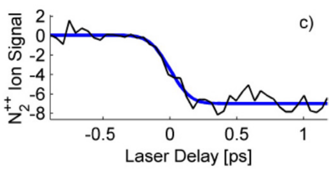

J. O'Neal


J. Wang

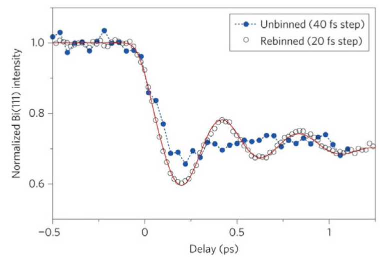

E. Isele

Time Resolution with X-ray FELs

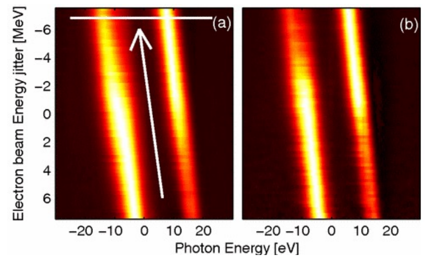
SLAC



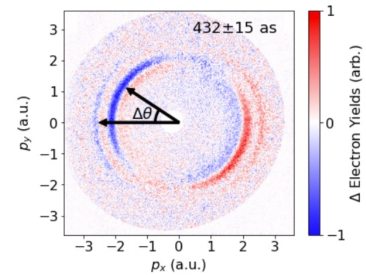
Early experiment
2010
~120 fs



Time sorting
2013~2014
10~30 fs



Two-color FEL
2013~2016
3~10 fs



Attosecond FEL
2017~ongoing
< 1 fs

1 ps | 100 fs | 10 fs | 1 fs | 100 as

Time Resolution

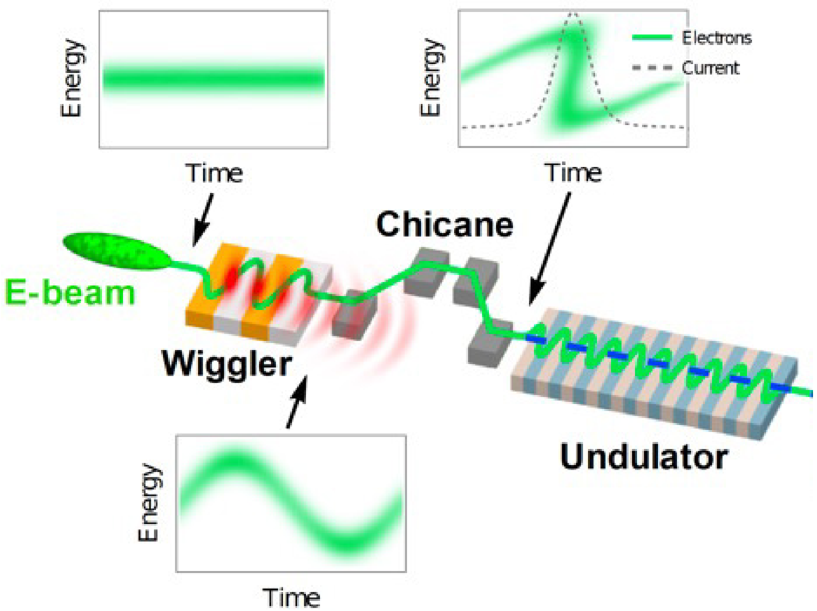
1. Glownia et al. Opt. Express 18. 17 (2010): 17620-17630
2. Cryan, James P., et al. Physical review letters 105.8 (2010): 083004.

1. Harmand, M., et al. Nature Photonics 7 (2013): 215-218
2. Hartmann, Nik, et al. Nature photonics 8.9 (2014): 706-709

1. Lutman, A. A., et al. Physical review letters 110.13 (2013): 134801.
2. Hara, Toru, et al. Nat Commun 4, 2919 (2013)
3. Marinelli, A., et al. Physical review letters 111.13 (2013): 134801.
4. Marinelli, A., et al. Nat Commun 6, 6369 (2015).
5. Lutman, Alberto A., et al. Nature Photonics 10.11 (2016): 745-750.

1. Duris, J., et al. Nature Photonics 14.1 (2020): 30-36.
2. Huang, S., et al. Physical review letters 119.15 (2017): 154801.
3. Malyzhenkov, A., et al. Physical Review Research 2.4 (2020): 042018.
4. Maroju, P. K., et al. Nature 578.7795 (2020): 386-391.

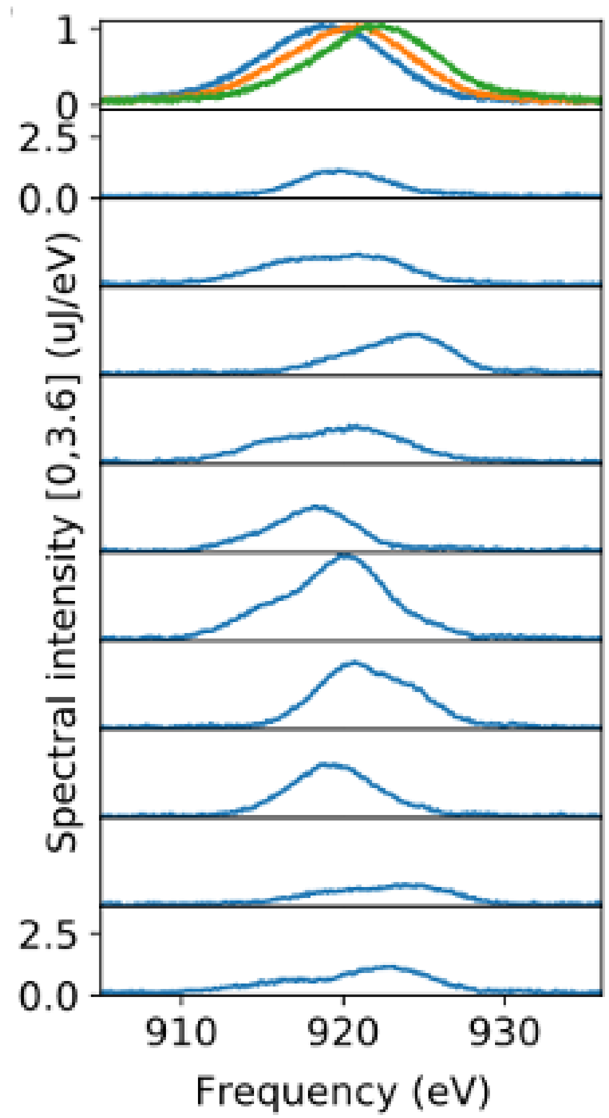
Attosecond Pulses: Enhanced SASE



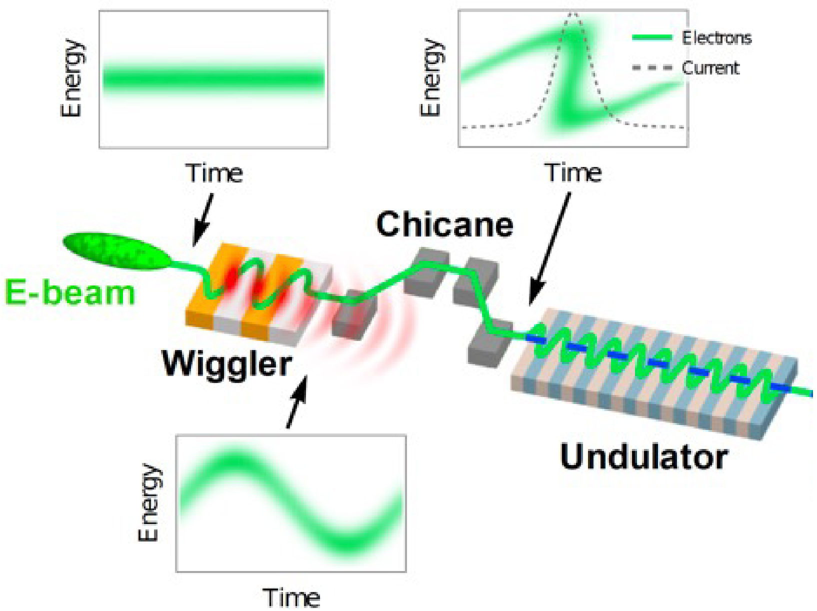
Original concept using laser: Zholents PRSTAB 8, 040701 (2005)

XLEAP Project
SLAC/ANL Collaboration

J. Duris, S. Li et al. *Nature Photonics* 14.1 (2020): 30-36.
J. Duris et al. Phys. Rev. Lett. 126, 104802 (2021)
J. MacArthur., et al. *Physical review letters* 123.21 (2019): 214801.
Zhang, Zhen, et al. *New Journal of Physics* 22.8 (2020): 083030.



Attosecond Pulses: Enhanced SASE



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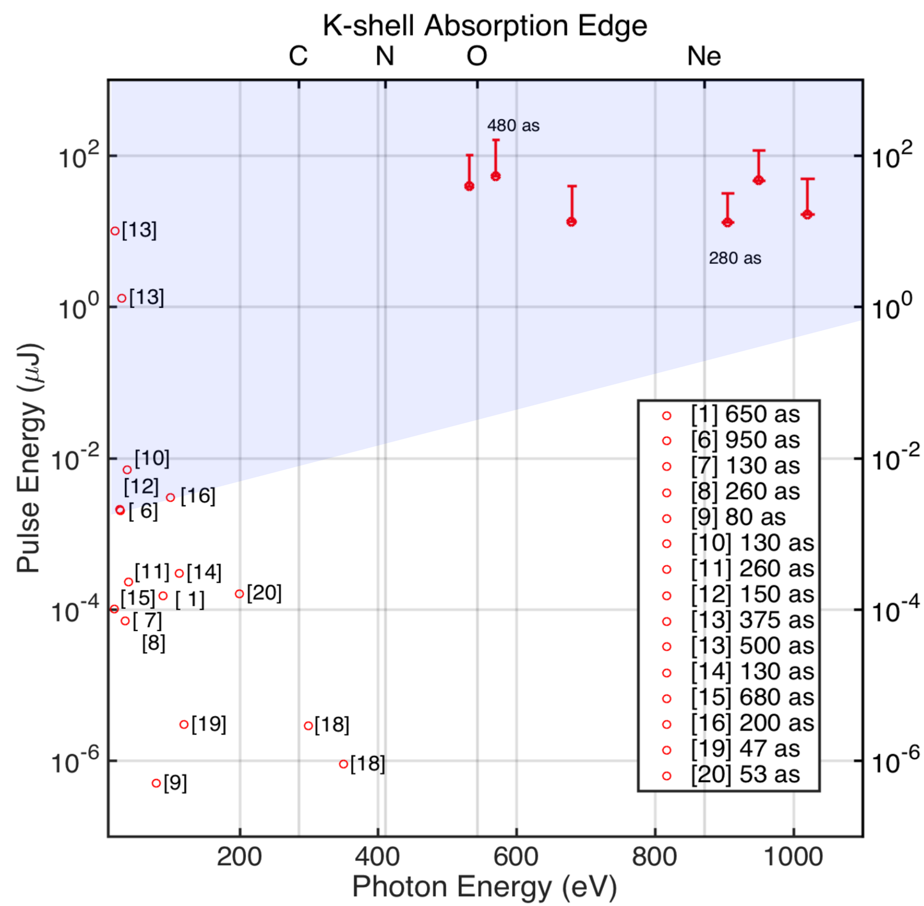
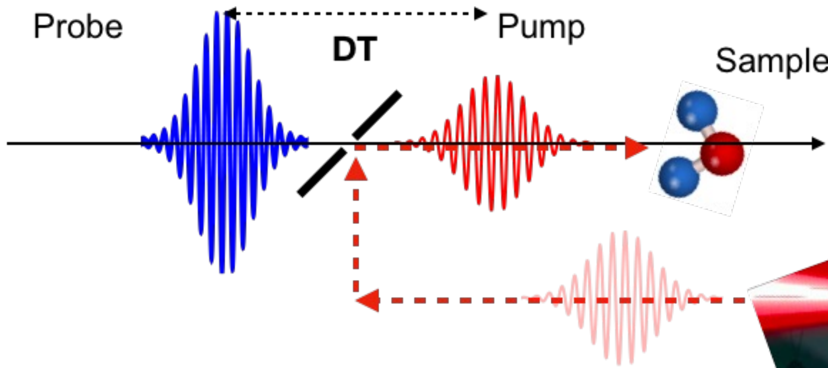
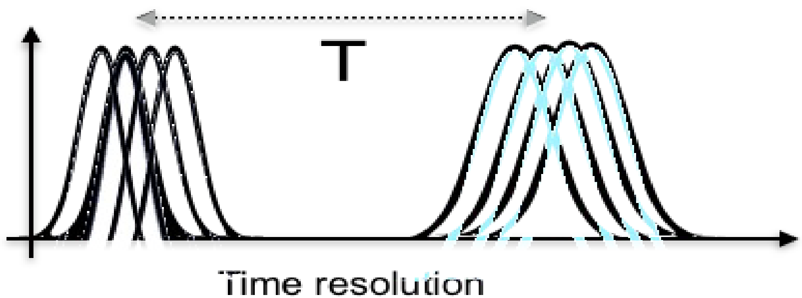


Attosecond Pump/Probe Experiment



Arrival time jitter/uncertainty ~ 10s – 100s fs!



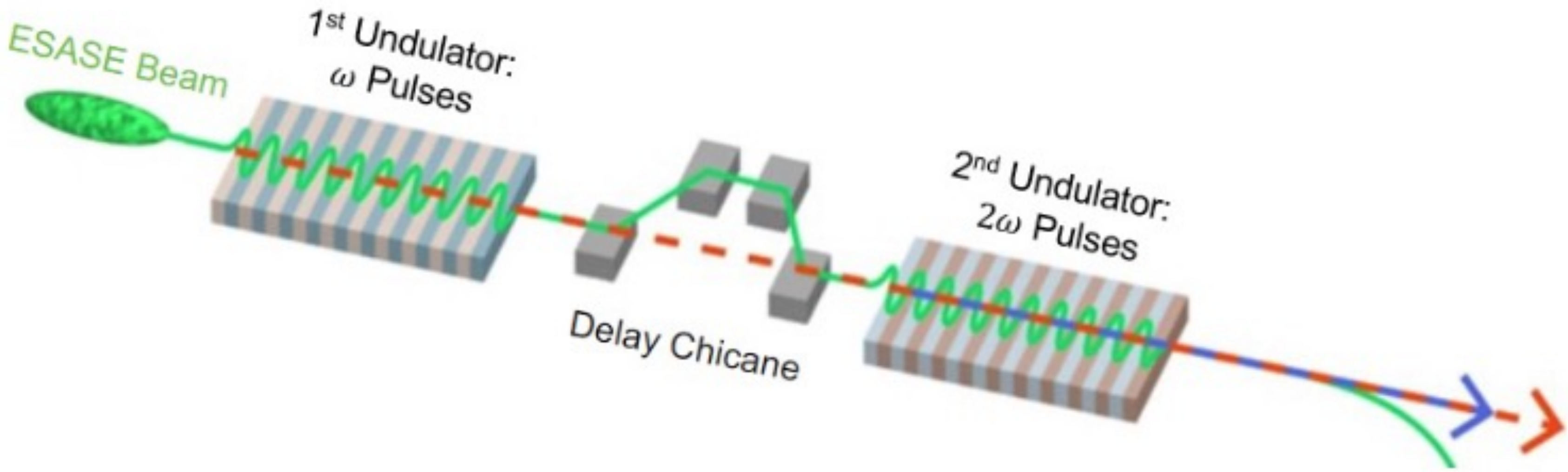
$$\sqrt{\Delta t_1^{\,2} + \Delta t_2^{\,2} + \sigma_T^2}$$

~20 fs RMS Uncertainty at XFELs!

Kang, Heung-Sik, et al. Nature Photonics 11.11 (2017): 708.
Harmand, M., et al. Nature Photonics 7.3 (2013): 215

Attosecond Two-Color Pump/Probe Setup

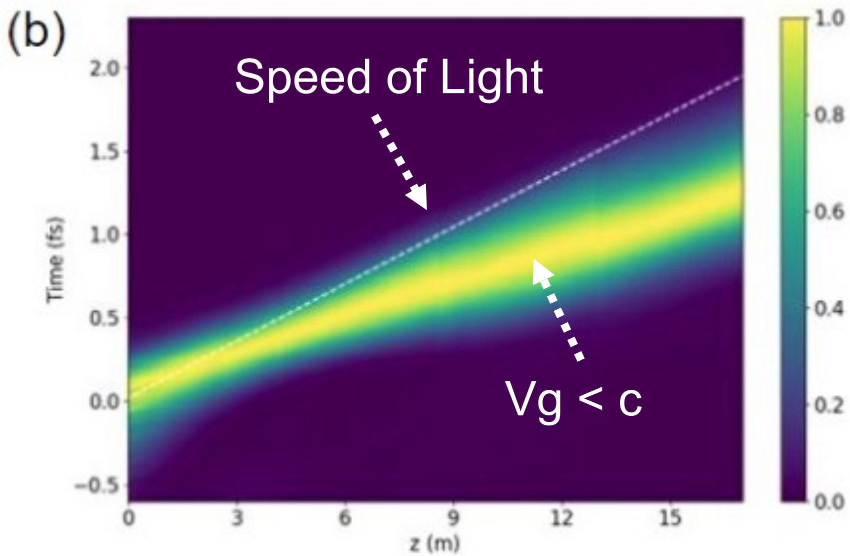
Attosecond stability -> pump and probe from e-beam!

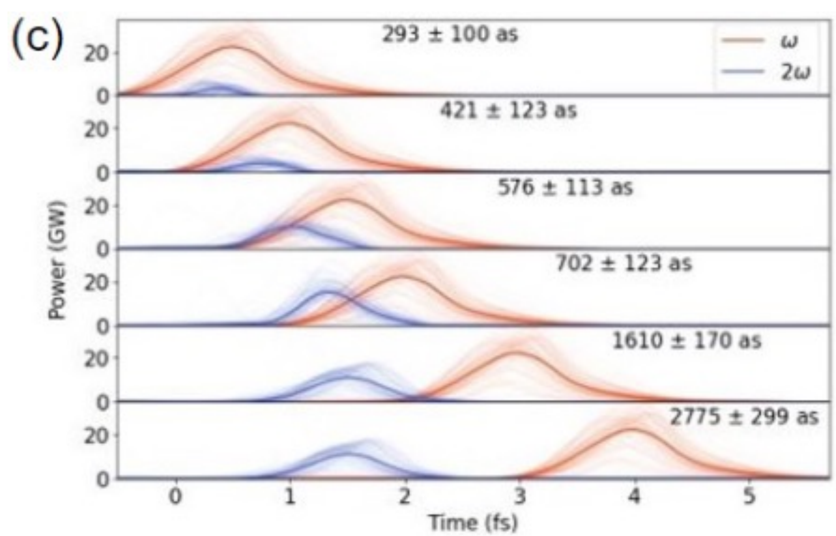


Minimal delay constrained by **the slippage of the 1st pulse in the 2nd undulator section**

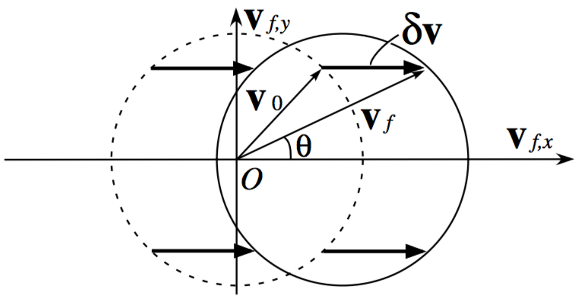
Use **harmonic configuration** to achieve **sub-femtosecond delay**

Attosecond Delay Control

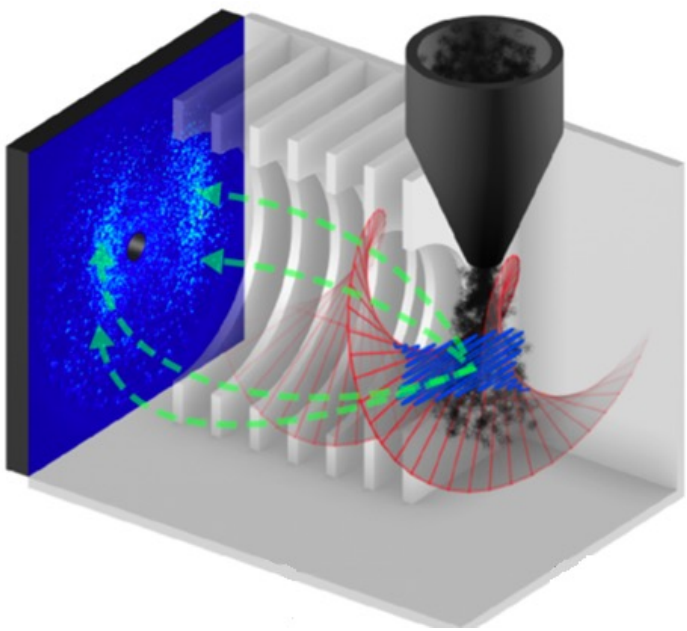


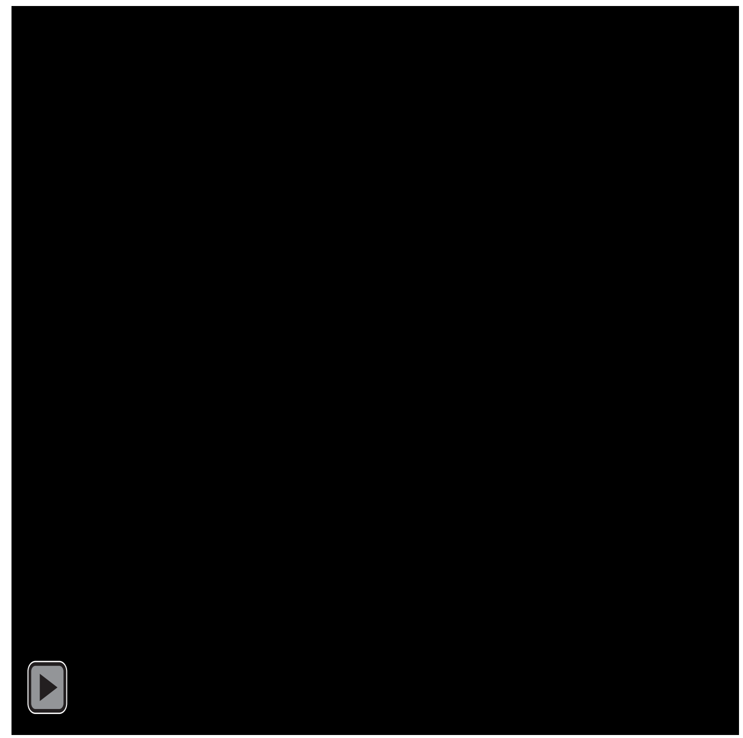


Attosecond Delay Measurement



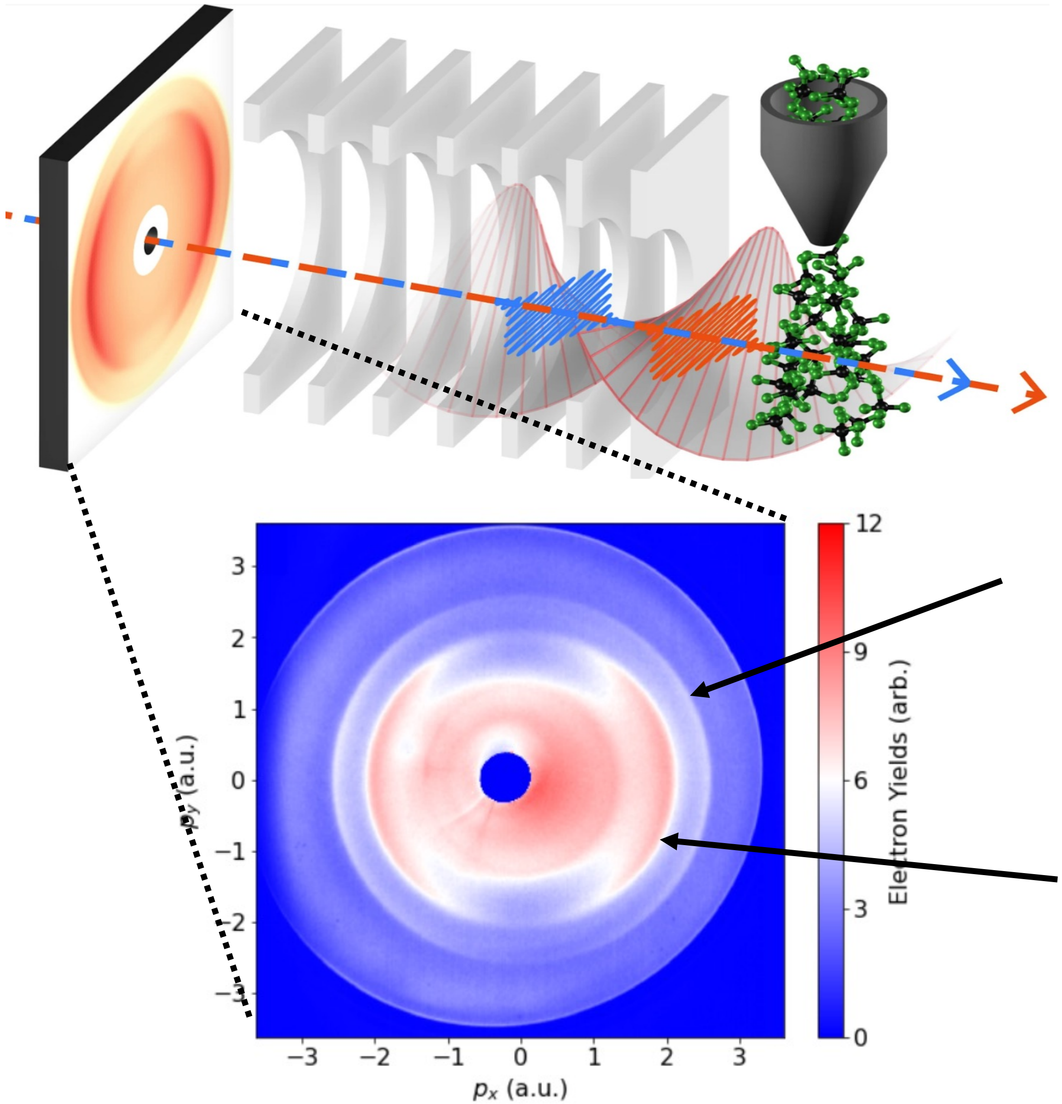
$$\vec{v}_f = \vec{v}_i + \frac{e}{m_e}\vec{A}(t_0)$$





J. Itatani, et al. Physical Review Letters 88.17 (2002): 173903.
M. Kitzler, et al. Physical review letters 88.17 (2002): 173904.
Hartmann, N., et al. Nature Photonics 12.4 (2018): 215-220.
J. Duris, S. Li, et al. Nature Photonics 14.1 (2020): 30-36.

Attosecond Delay Measurement

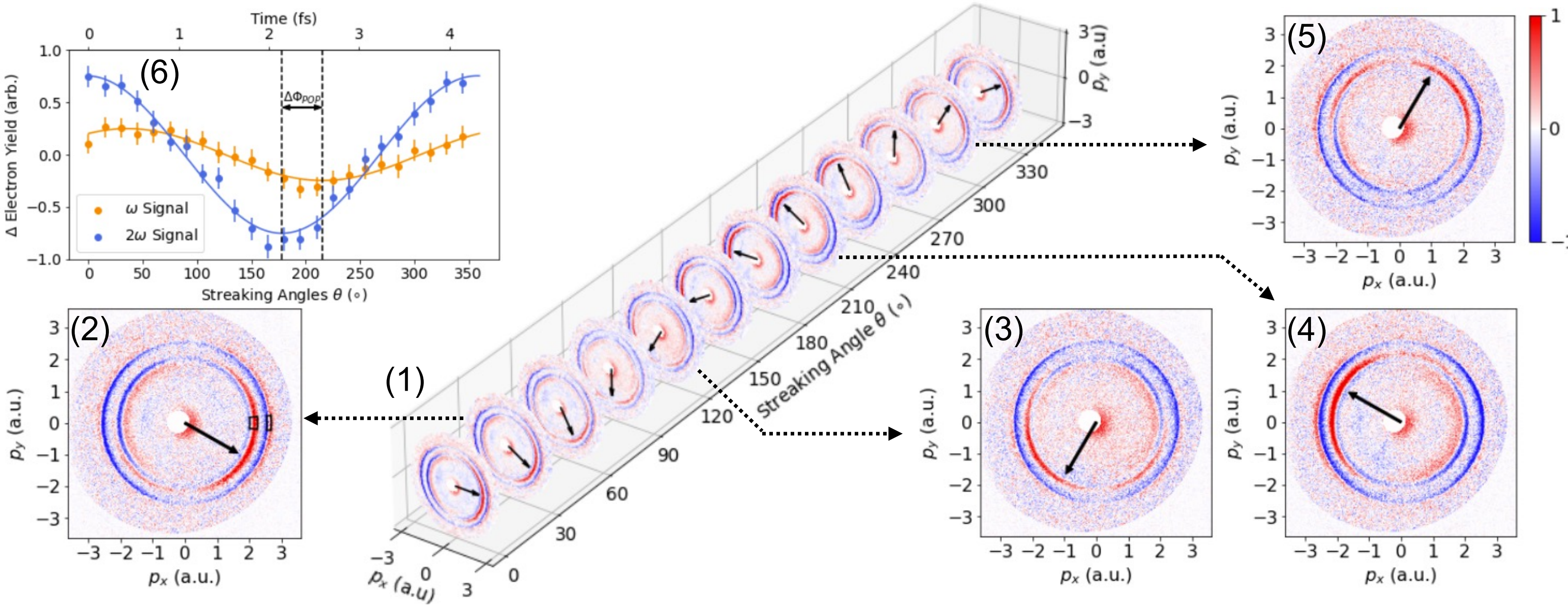


Averaged IR-mistimed Image

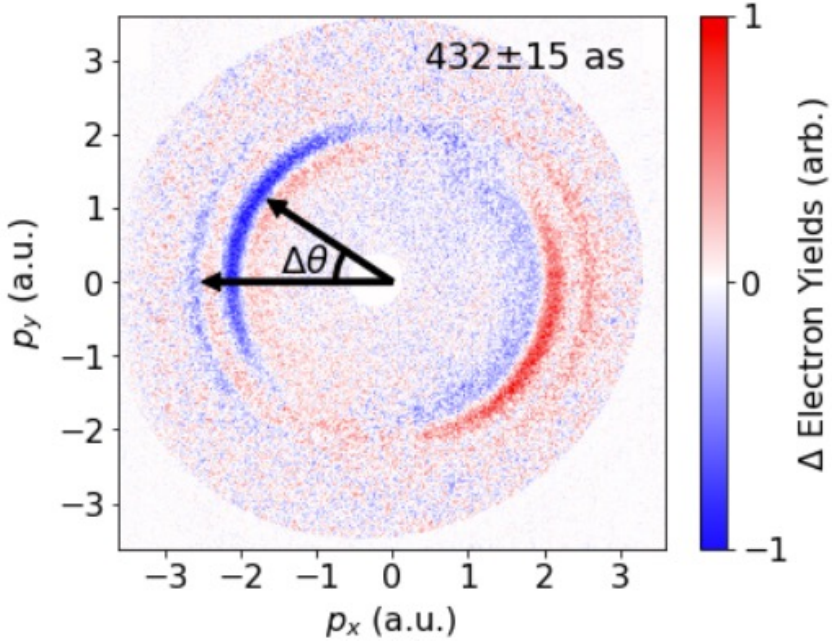
ω (370 eV) pulses ionize carbon 1s:
370 eV – 284 eV = 86 eV

2ω (740 eV) pulses ionize fluoride 1s:
740 eV – 685 eV = 55 eV

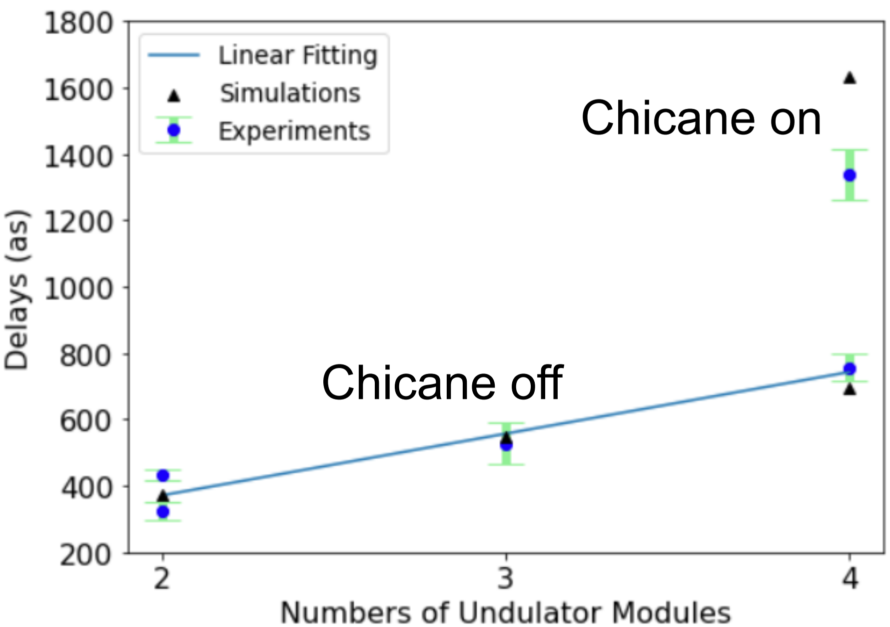
Attosecond Delay Measurement



Attosecond Delay Measurement



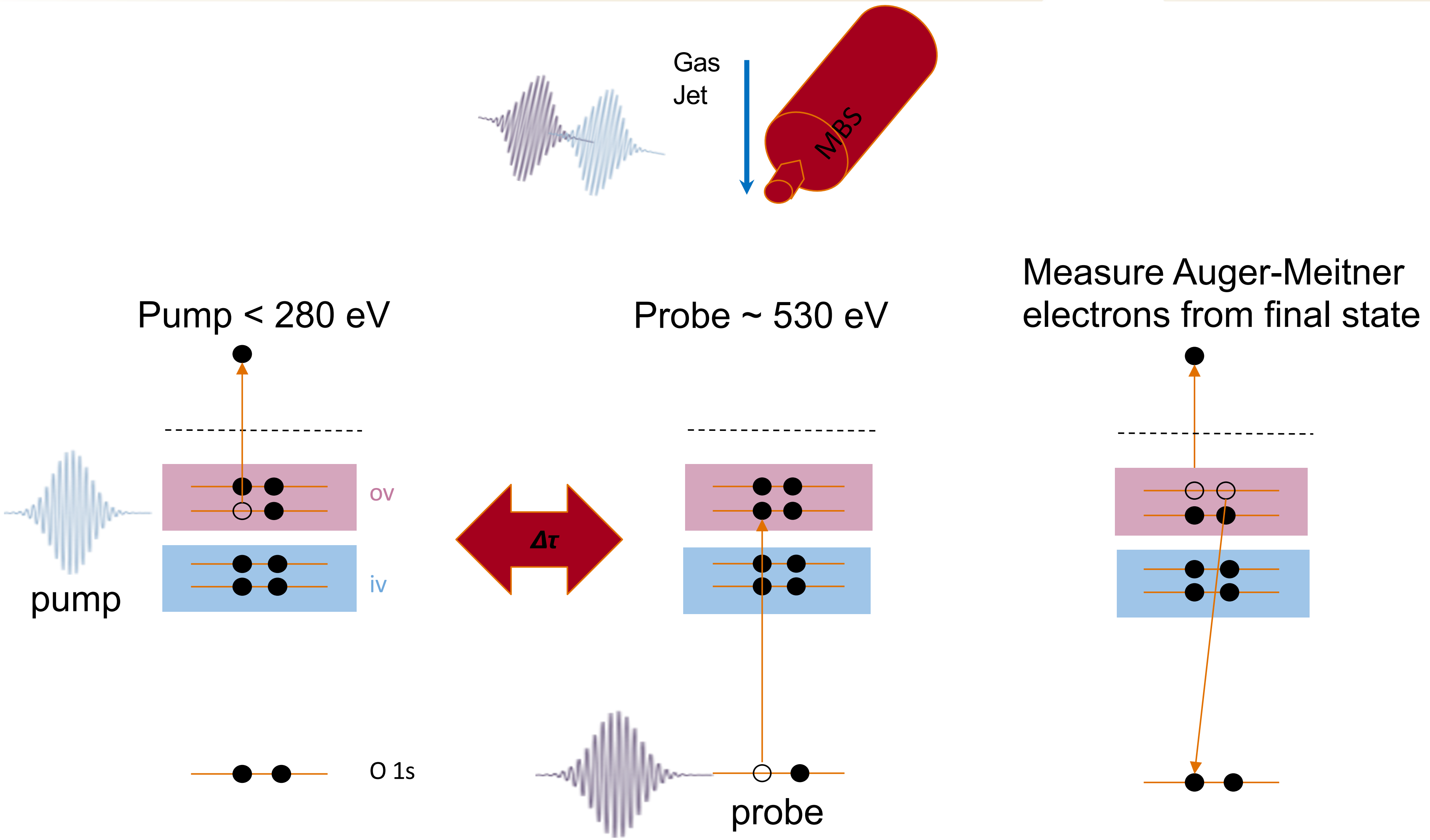
Chicane off + 2 Undulator Modules



$\Delta\tau$ per und ~150 as

Full slippage: 87×5.6 as (740 eV) = 486 as

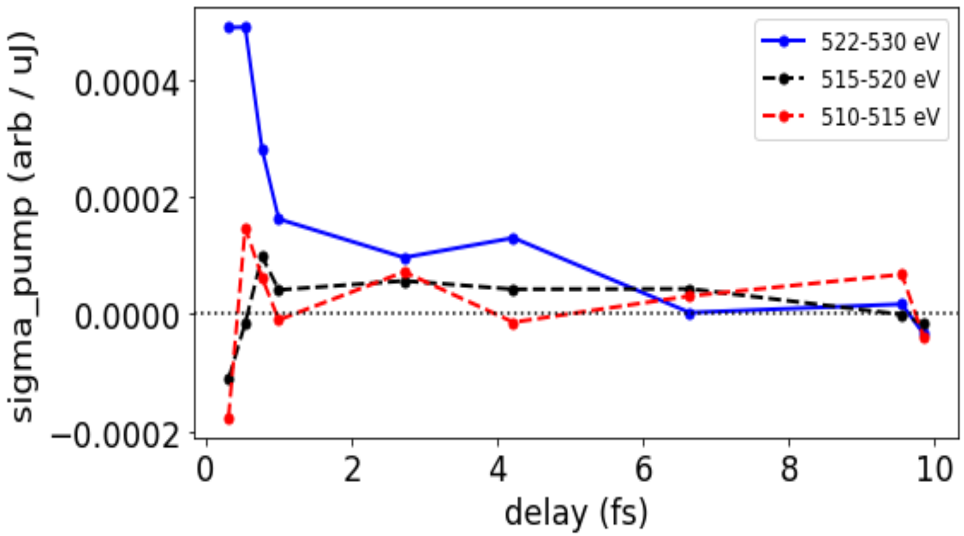
Pump-Probe Scheme



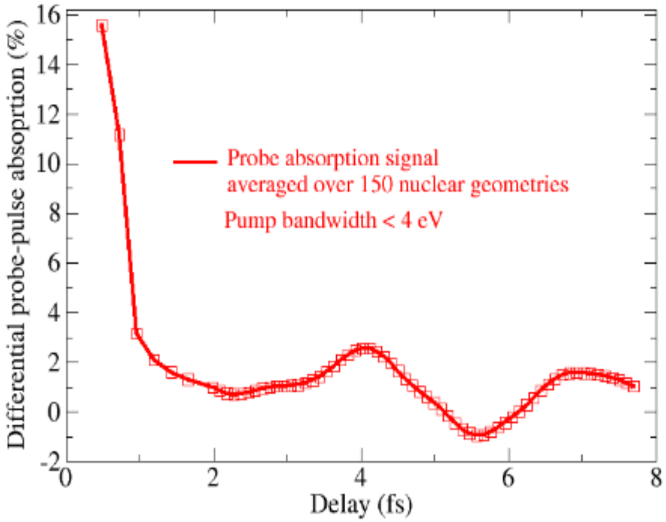
Time-Delay Scan (Preliminary Analysis)

SLAC

Experiment



Theory (XAS)

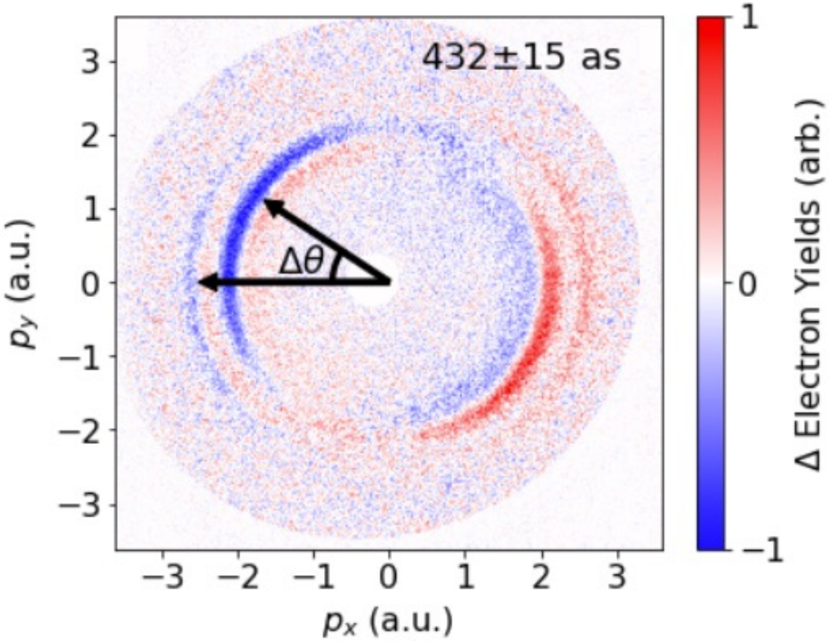


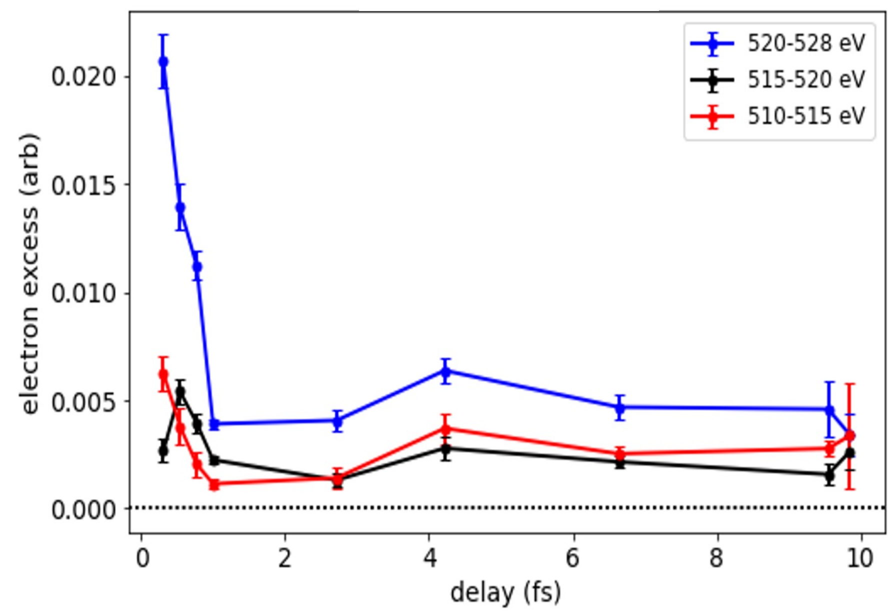
Data analysis: J. O'Neal, O. Alexander, Z. Guo, T. Driver et al.
Theory: M. Ruberti

Conclusions

- Attosecond pump/probe XFEL capabilities are now a reality!
- Sub-femtosecond pump/probe delays are measured with angular streaking. Pump/probe delays can be controlled with 150 as accuracy.
- Sub-femtosecond two-color pump/probe technique has been successfully applied to observe attosecond charge migrations.

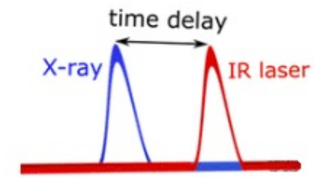
Questions?

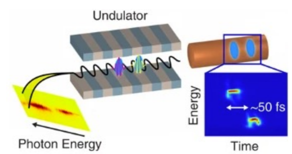

432±15 as
Δθ
p_y (a.u.)
p_x (a.u.)
Δ Electron Yields (arb.)
3
2
1
0
−1
−2
−3

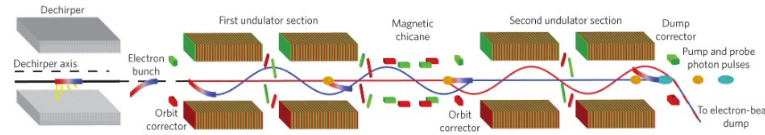

520-528 eV
515-520 eV
510-515 eV
electron excess (arb)
delay (fs)
0.020
0.015
0.010
0.005
0.000
0
2
4
6
8
10

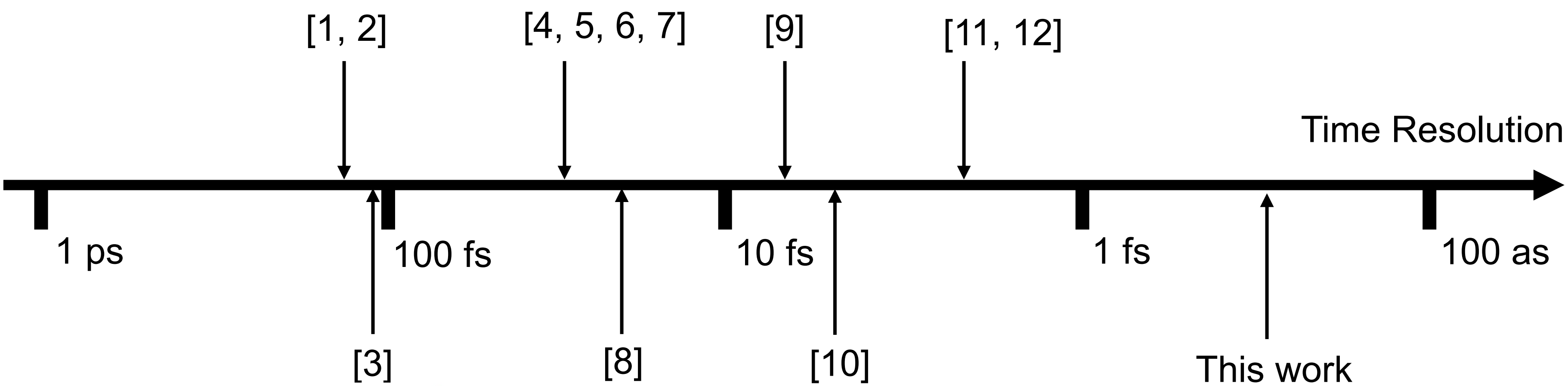
Backup Slides

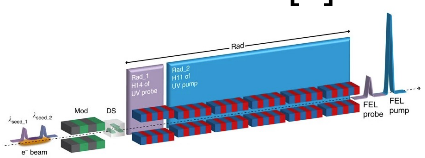
Time Resolution with X-ray FELs

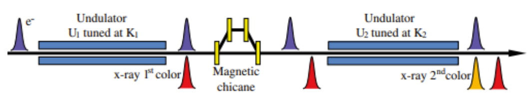










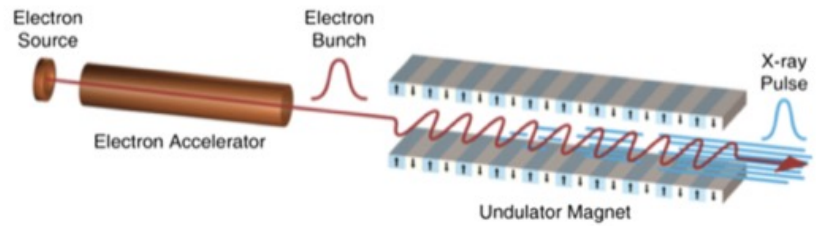


1. Glownia et al. Opt. Express 18. 17 (2010): 17620-17630
2. Cryan, James P., et al. Physical review letters 105.8 (2010): 083004.
3. Ferrari, Eugenio, et al. Nat Commun 7, 10343 (2016).
4. Harmand, M., et al. Nature Photonics 7 (2013): 215-218
5. Hartmann, Nik, et al. Nature photonics 8.9 (2014): 706-709.
6. Prat, Eduard, et al. Nat. Photonics 14, 748–754 (2020).
7. Kang, Heung-Sik, et al. Nature Photonics 11.11 (2017): 708-713.
8. Lutman, A. A., et al. Physical review letters 110.13 (2013): 134801.
9. Marinelli, A., et al. Nat Commun 6, 6369 (2015).
10. Hara, Toru, et al. Nat Commun 4, 2919 (2013)
11. Lutman, Alberto A., et al. Nature Photonics 10.11 (2016): 745-750.
12. Barillot, T., et al. Physical Review X 11.3 (2021): 031048.

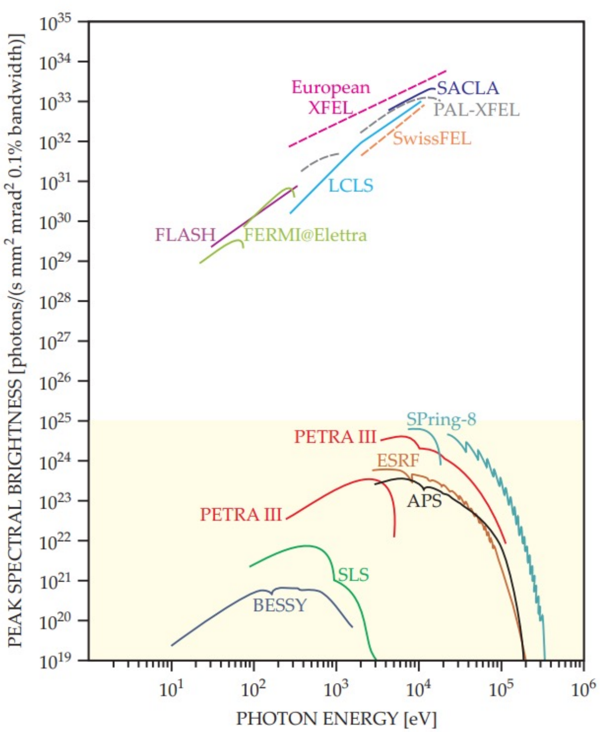
X-ray Free-Electron Laser (XFEL)

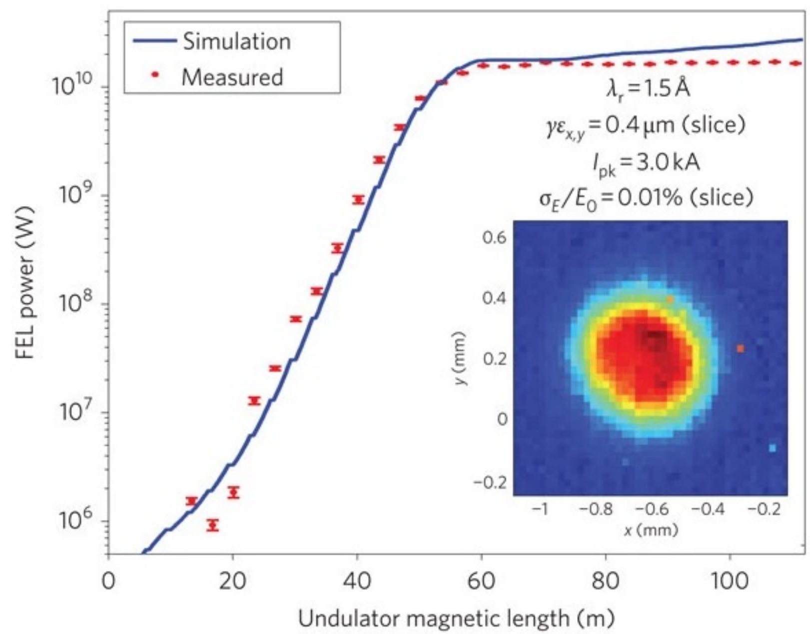
XFEL shares properties of conventional ultrafast lasers:

- High power (up to 100s GW)
- Short pulse (sub-hundred fs)
- Narrow bandwidth (~0.1% to 0.005%)
- Transverse coherence



$$\lambda_{FEL} = \frac{\lambda_u}{2\gamma^2}\left(1 + \frac{K^2}{2}\right)$$

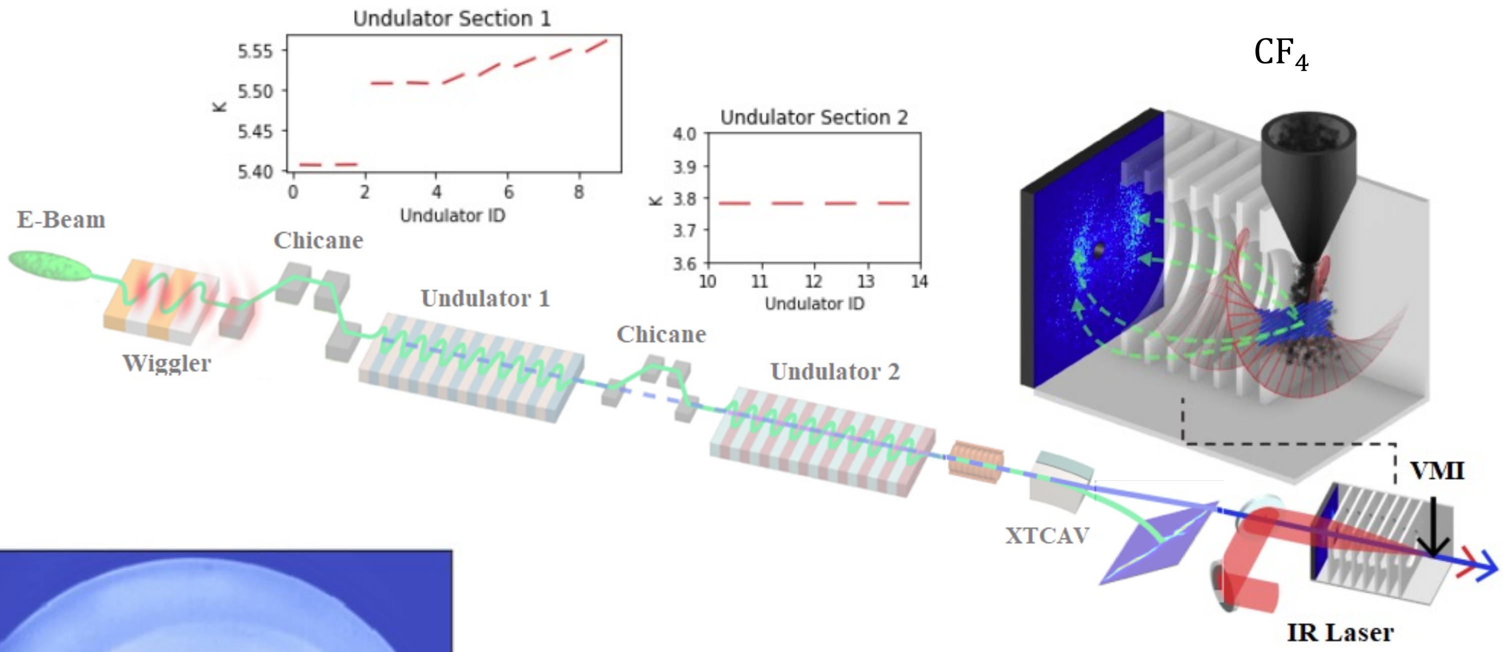


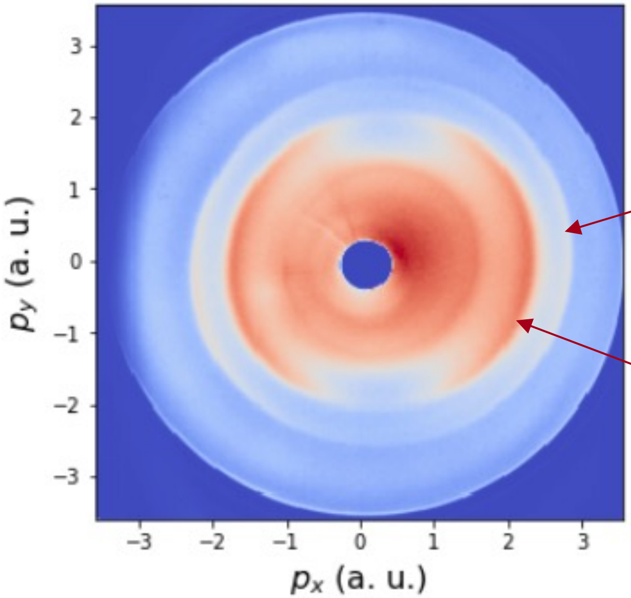


Bucksbaum, Philip H., and Nora Berrah. *Physics Today* 68.7 (2015): 26.

Emma, Paul, et al. *Nature Photonics* 4.9 (2010): 641-647.

Angular Streaking Measurements of Two-Color Attosecond Pump/Probe Pulse Pairs on LCLS-II



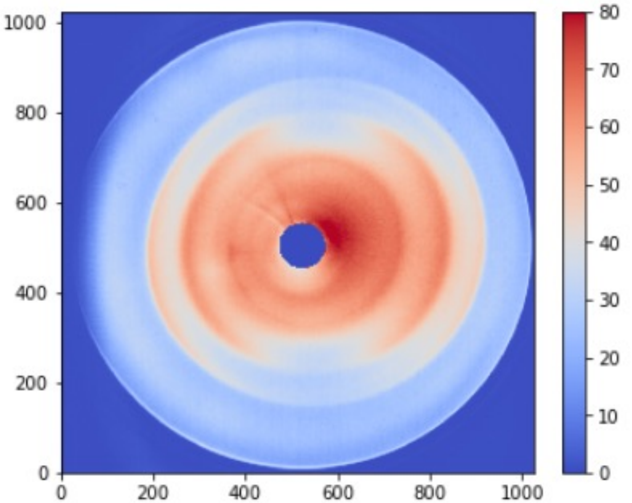


375 eV pulses ionize carbon
Kinetic energy: 375 eV – 283 eV = 92 eV

750 eV ionize ionize fluoride
Kinetic energy: 750 eV – 687eV = 63 eV

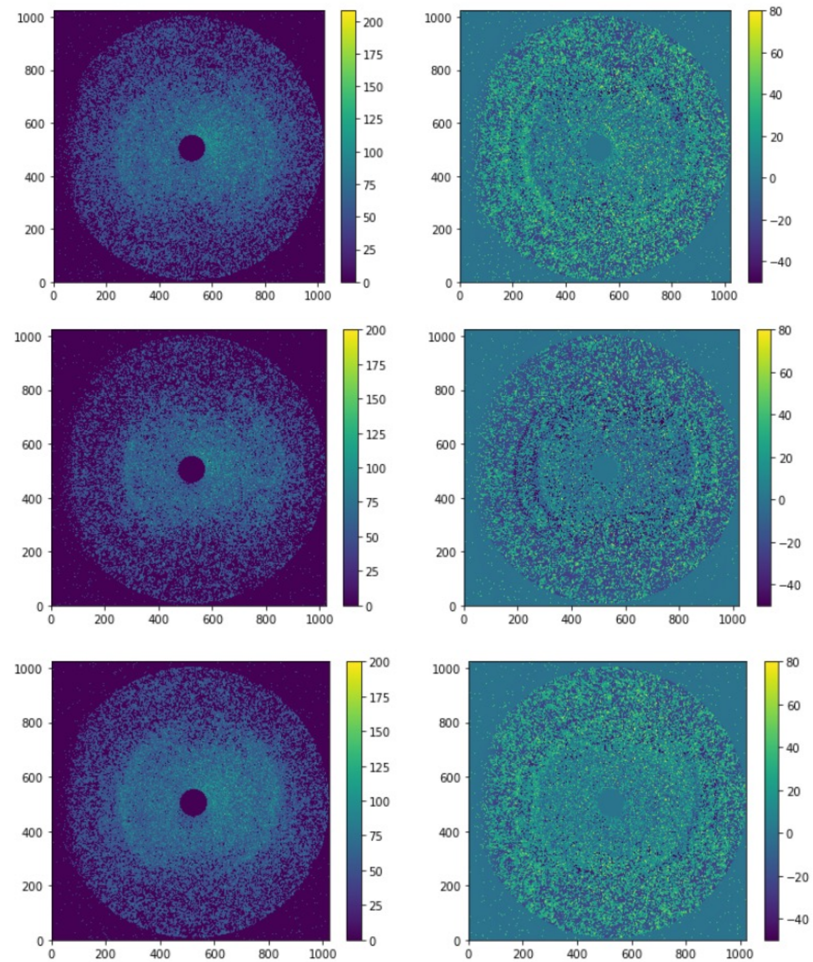
Backup Slide: Single-Shot Image of 2-Color Streaking

Averaged IR-Mistimed cVMI Image



Single Shot Image

Single Shot - Unstreaked



Backup Slide: Streaking Analysis

Workflow of streaking analysis:

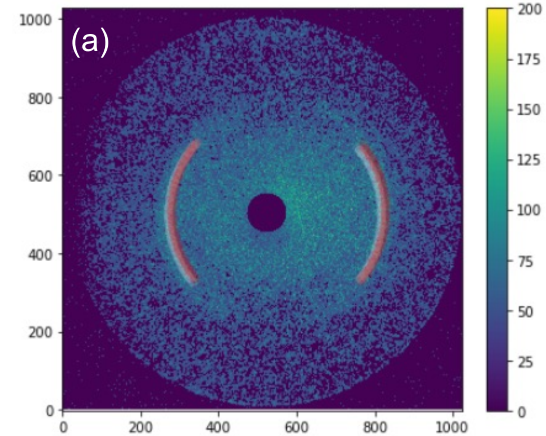
1.Apply single-shot based analysis algorithm to the reference photoline in each VMI image.

2. Estimate the streaking angle and amplitude for each single shot.

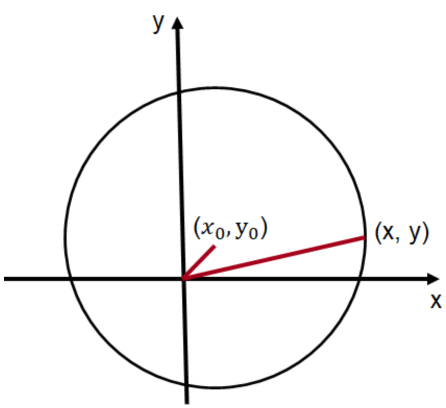
3.Sum up shots with the same streaking angle.

4.Compare averaged streaked images to the averaged unstreaked image.

5.Analyze the delay.



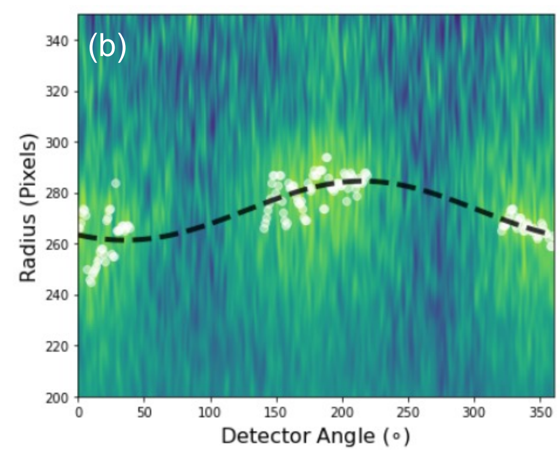
Red blob: Unstreaked photoline
White blob: Streaked photoline



$$x_0 = r_0\cos(\theta_0),\ y_0 = r_0\sin(\theta_0)$$

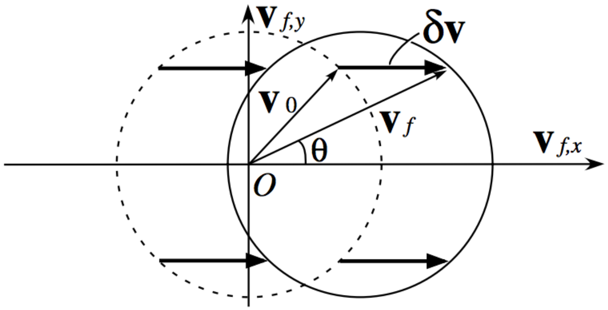
$$x = r\cos(\theta), y = r\sin(\theta)$$

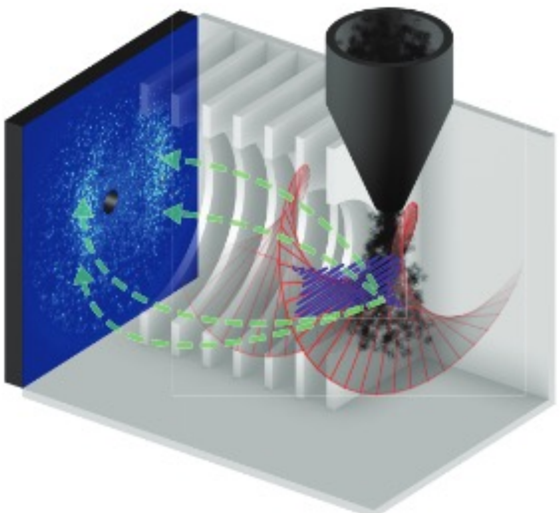
$$r = r_0\cos(\theta - \theta_0) + \sqrt{R^2 - r_0^2\sin^2(\theta - \theta_0)}$$



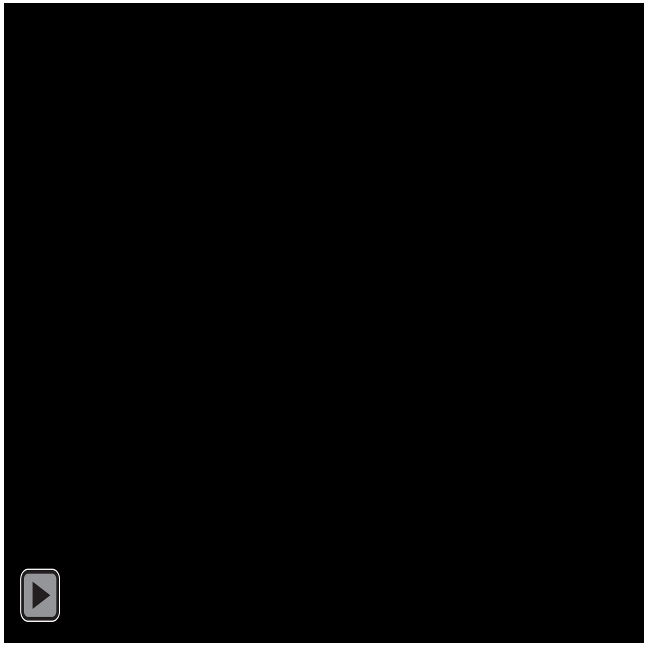
Angular Streaking Measurements of Isolated Attosecond XFEL Pulses

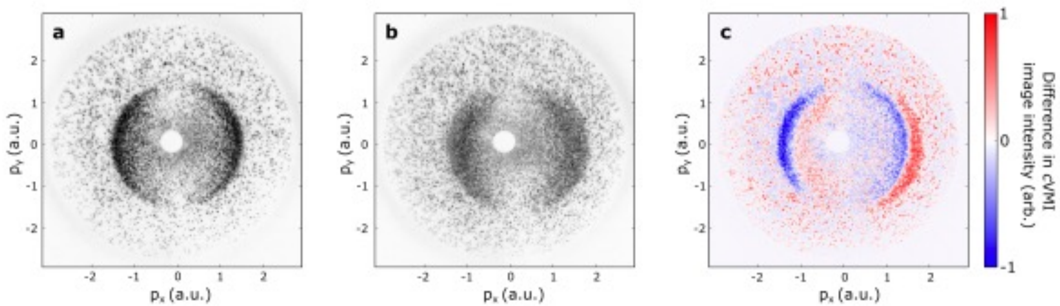
$$\vec{v}_f = \vec{v}_0 + \frac{e}{m_e}\vec{A}(t_0)$$





Movie made by Philipp Rosenberger





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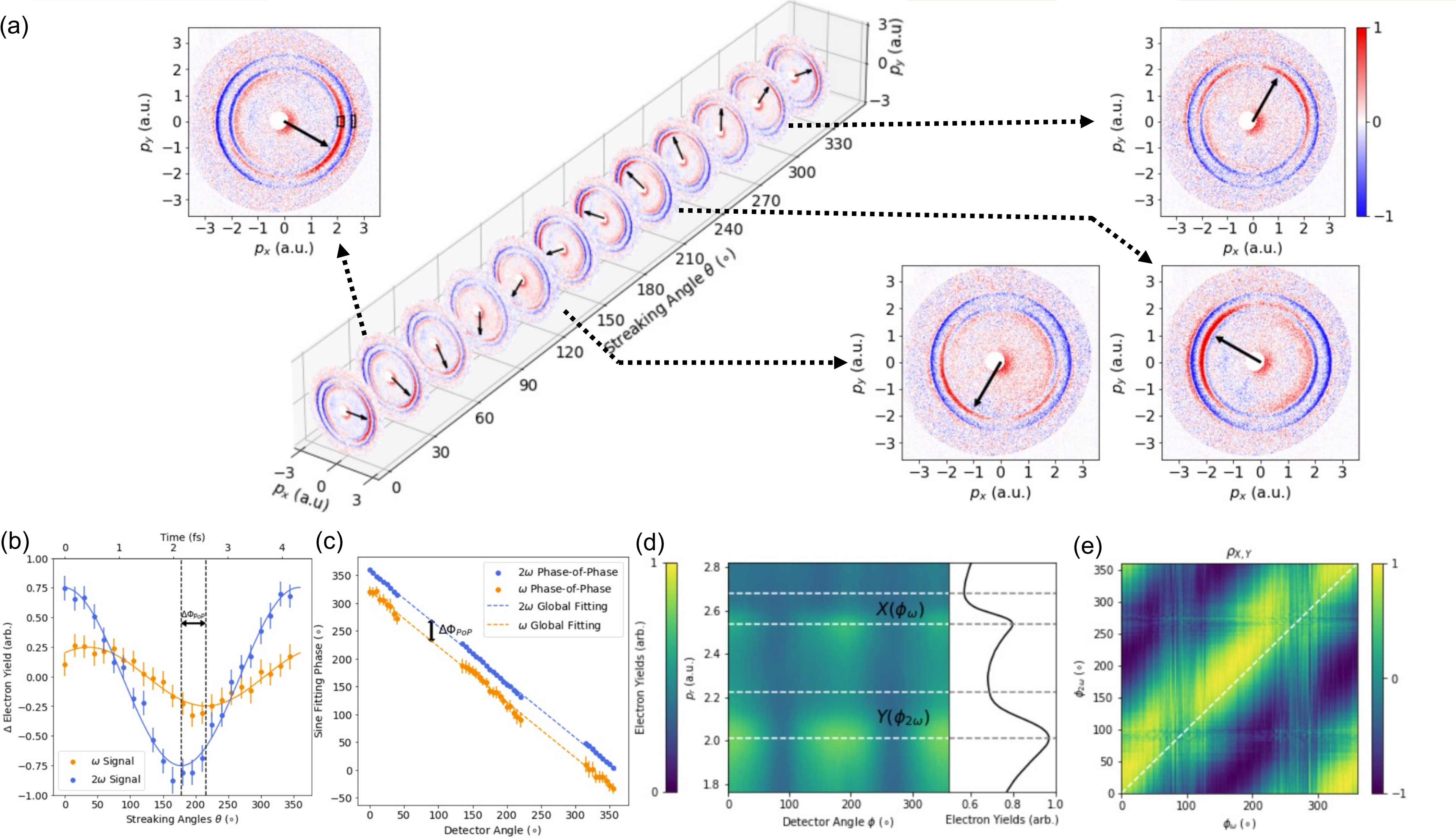
M. Kitzler, et al. Physical review letters 88.17 (2002): 173904.
Hartmann, N., et al. Nature Photonics 12.4 (2018): 215-220.
J. Duris, S. Li, et al. Nature Photonics 14.1 (2020): 30-36.

Angular Streaking Measurements of Two-Color Attosecond Pump/Probe Pulse Pairs

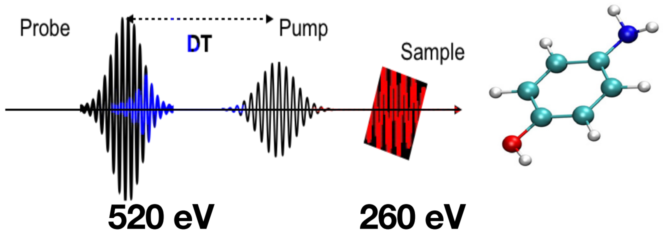


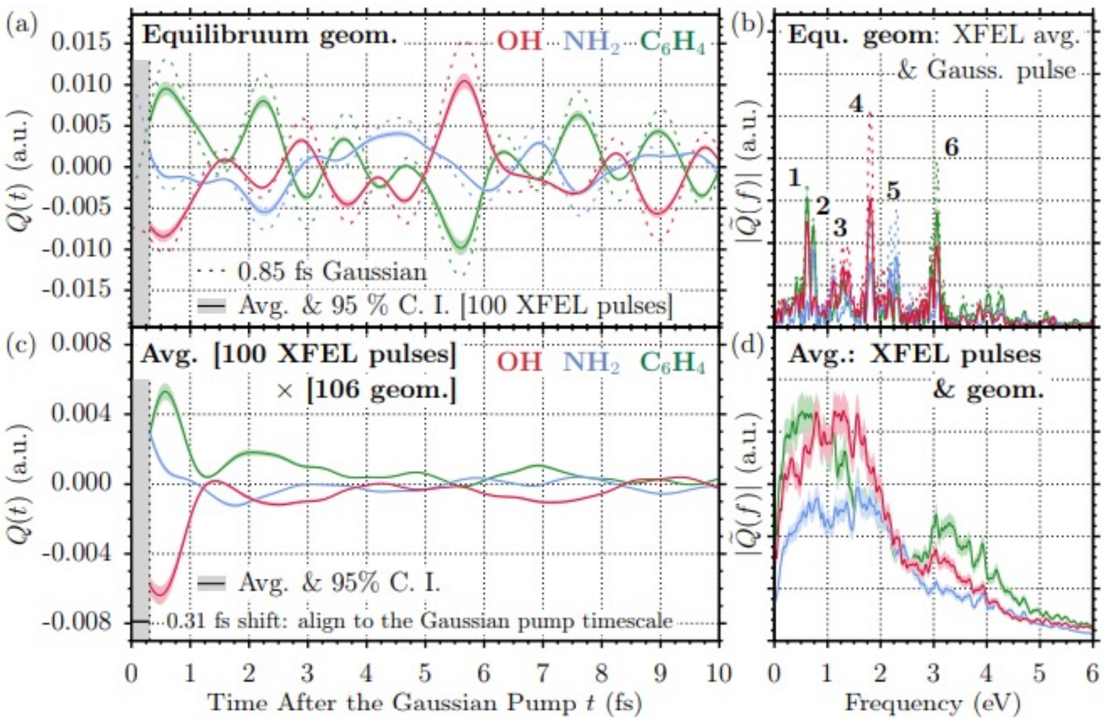
Real-Time Observation of Attosecond Charge Migration

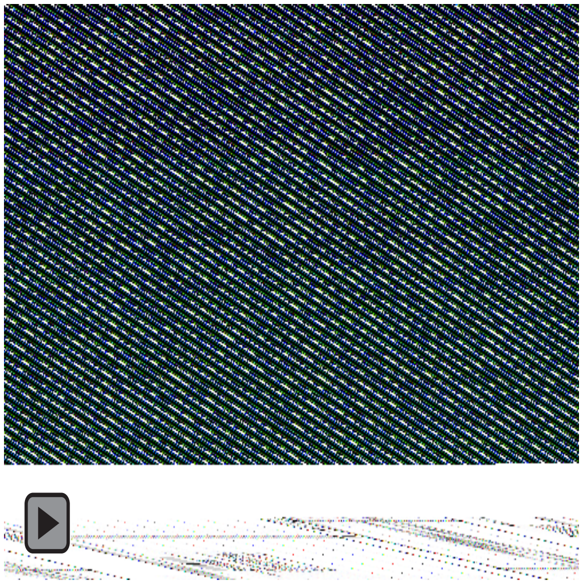
SLAC

Impulsive removal of valence electrons with XLEAP:

- Create coherent superposition of ionic states with pump pulse
- Probe around oxygen edge with probe pulse







Movie by G. Grell

G. Grell, Z. Guo et al., submitted to Optica

Real-Time Observation of Ultrafast Electron Motion

Attosecond Campaign @ LCLS:
Cryan, Walter, and Marinelli