Attosecond x-ray free-electron lasers

Agostino Marinelli
SLAC National Accelerator Laboratory
and Stanford University

Acknowledgements

XLEAP/XLEAP-II Collaboration

SLAC
A. Marinelli, J. Cryan, et al.
ANL
A. Zholents, J. Xu, M. Qian et al.
UCLA
S. Carbajo et al.

Hard X-ray Attosecond Pulses

University of Tokyo/SACLA
I. Inoue
SLAC
A. Marinelli, R. Robles, A. Halavanau

Attosecond Campaign (PIs)

LCLS - Leads
James Cryan
Agostino Marinelli
SLAC - Pulse
Taran Driver
Phil Bucksbaum

Imperial College
Vitali Averbukh
Jon Marangos

UAM
Fernando Martin
Antonio Picon
Alicia Palacios

OSU
Lou Dimauro

LBNL
Oliver Gessner
Daniel Slaughter
Thorsten Weber
Argonne NL
Gilles Doumy
Linda Young
KSU
Daniel Rolles
Artem Rudenko

Univ. Conn.
Nora Berrah
LSU
Ken Lopata

Tohoku Univ.
Kiyoshi Ueda
PSI
Christoph Bostedt
Andre Al-Haddad

Funding:
DOE Basic Energy Sciences
Accelerator and Detector
Research Program

SLAC LDRD and PD programs
Panofsky and Siemann
Fellowships

U.S. DEPARTMENT OF ENERGY | Office of Science | Basic Energy Sciences

Attosecond Core Group at LCLS

Principal Investigators


A. Marinelli

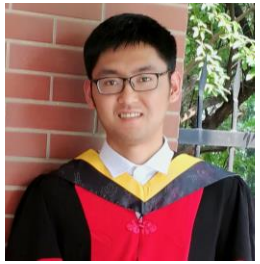

J. Cryan

SLAC

PULSE

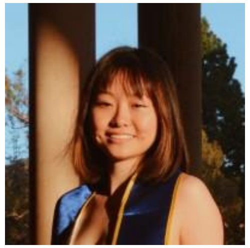
Stanford University

Current Team


Z. Zhang


D. Cesar


N. Sudar


V. Guo

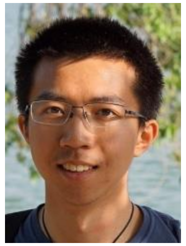

P. Franz


R. Robles


K. Larsen


E. Thierstein


T. Driver


J. Wang

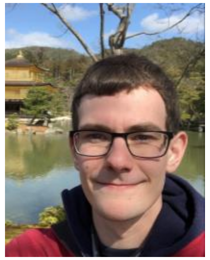

E. Isele

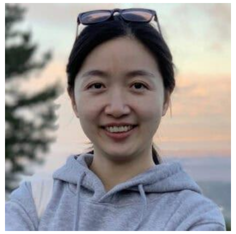

S. O’Tool

Alumni


Z. Guo


A. Wang


J. O’Neal


Siqi Li

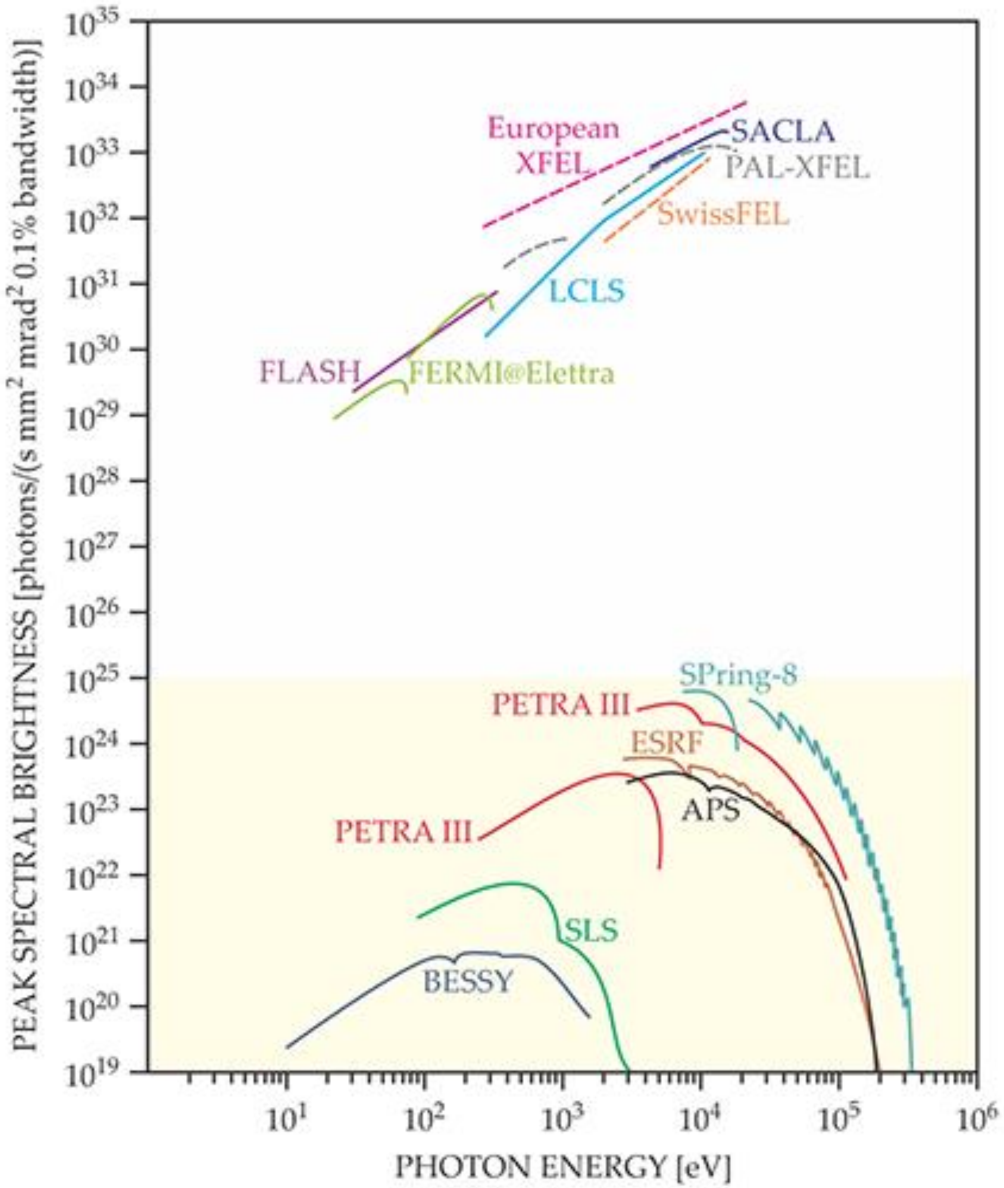

J. Duris


S. Beauvarlet
U. Conn

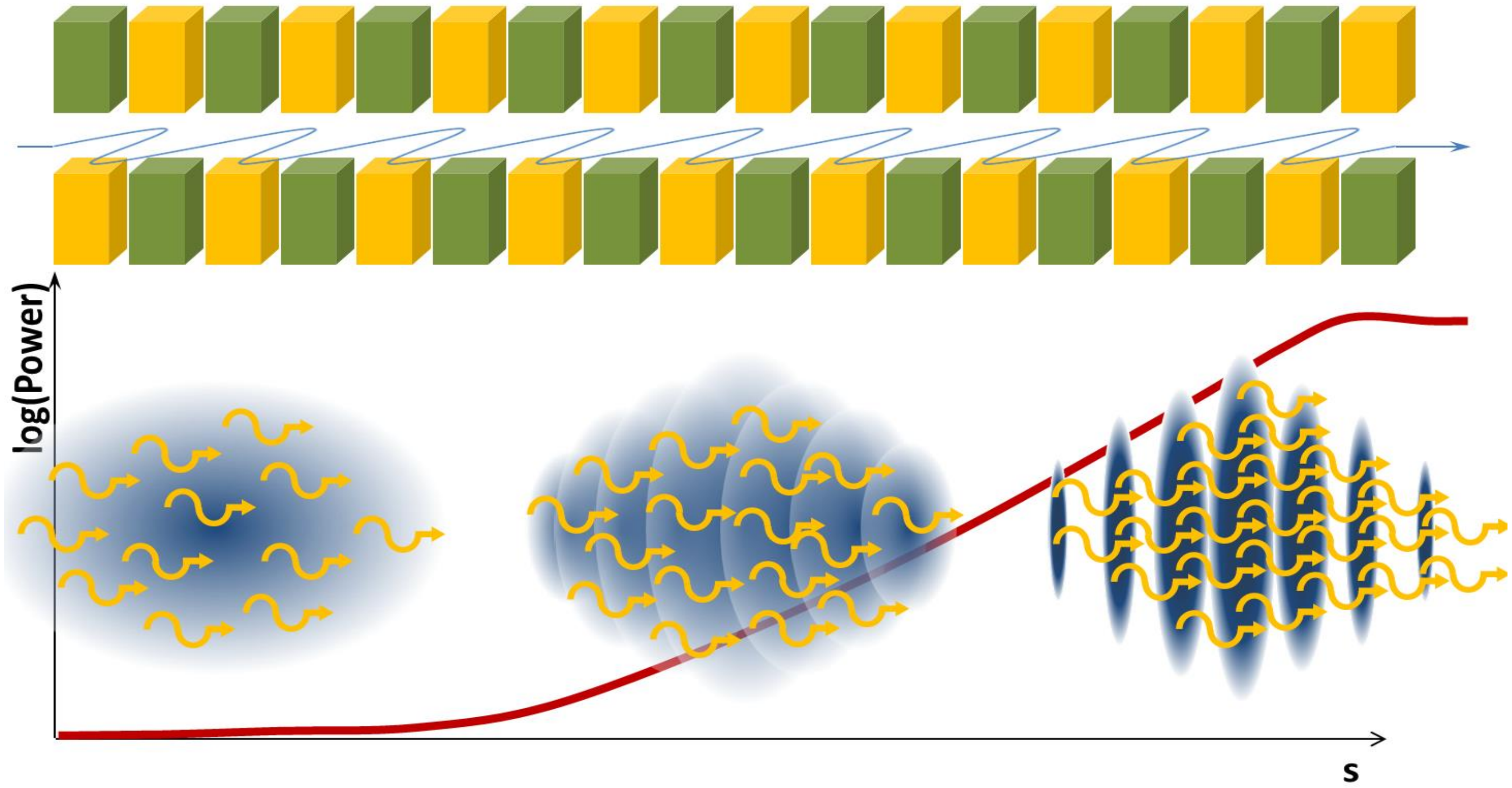
Laser Group


R. Lemons

X-ray Free-Electron Lasers



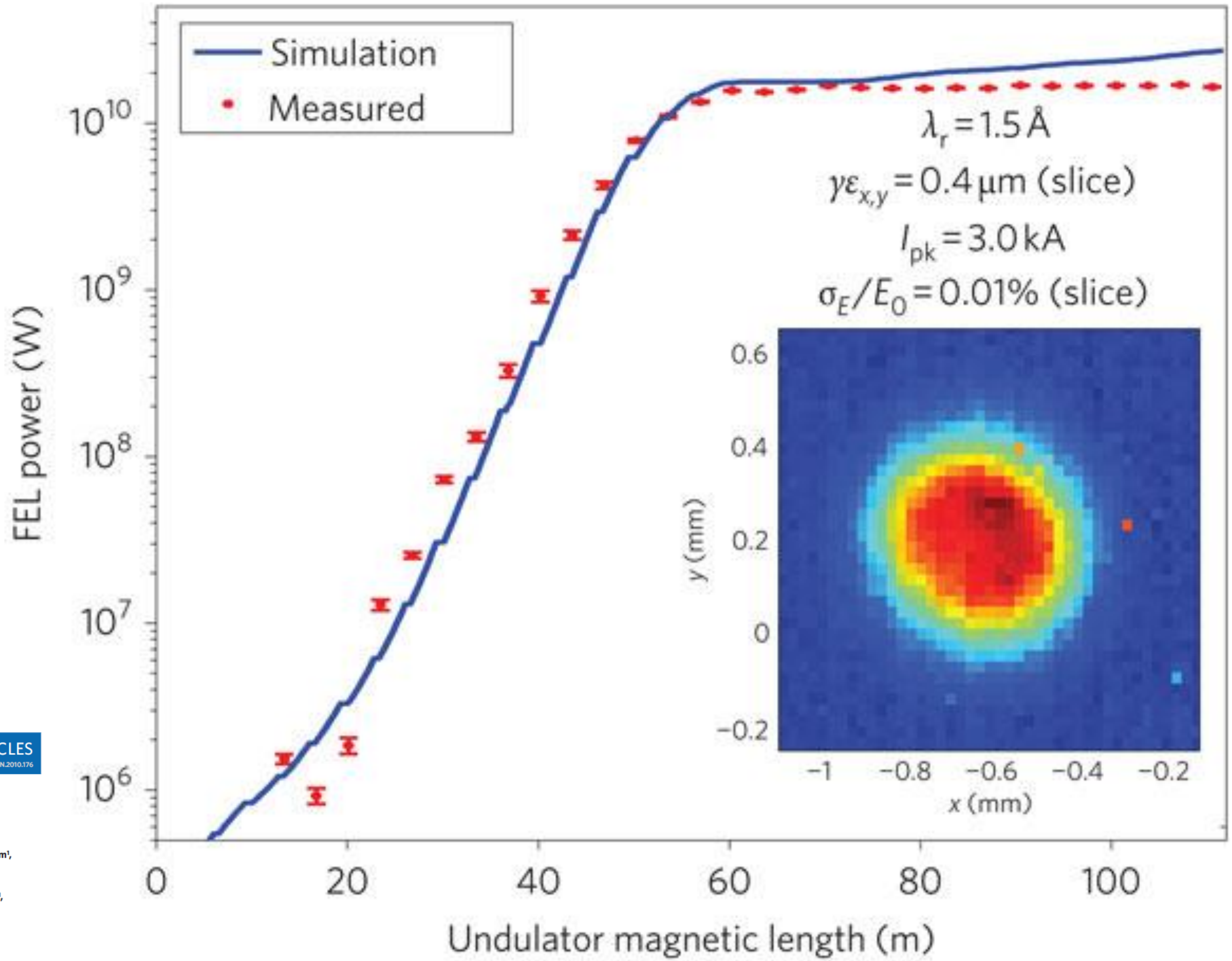
Free-Electron Laser



$$\lambda_r = \lambda_u\left(\frac{1}{\beta_z} - 1\right)$$

Source: https://www.helmholtz-berlin.de

The X-ray Free-Electron Laser

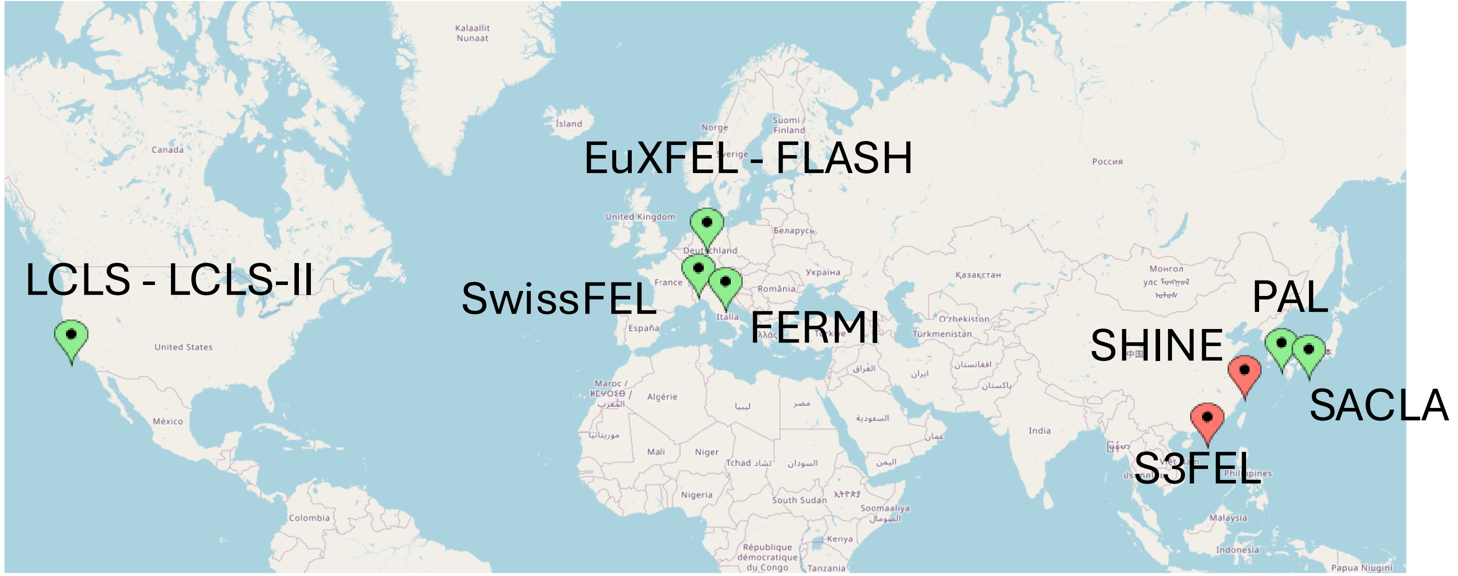


nature photonics ARTICLES
PUBLISHED ONLINE: 1 AUGUST 2010 | DOI: 10.1038/NPHOTON.2010.176

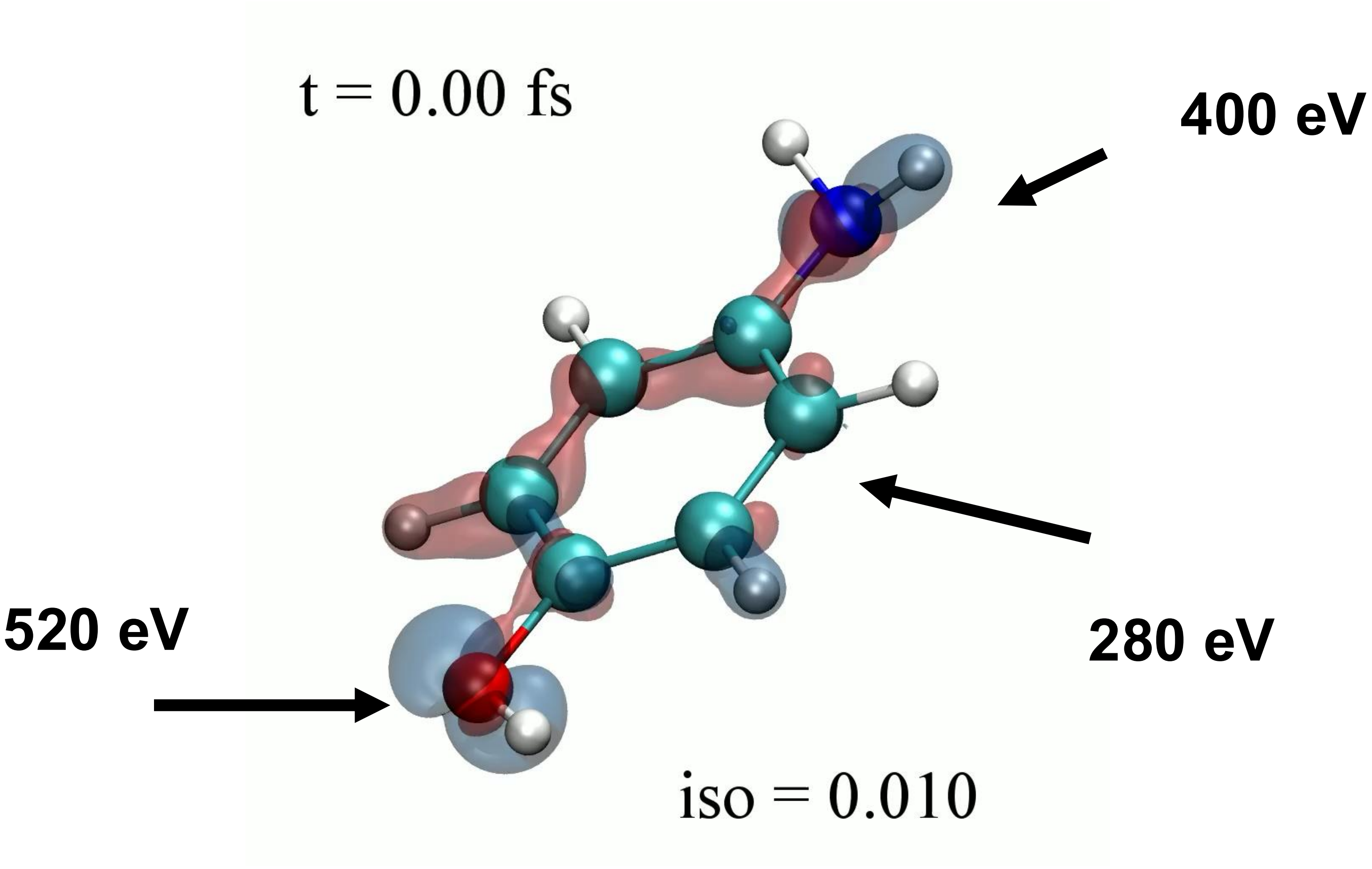
First lasing and operation of an ångstrom-wavelength free-electron laser

P. Emma[1*], R. Akre[1], J. Arthur[1], R. Bionta[2], C. Bostedt[1], J. Bozek[1], A. Brachmann[1], P. Bucksbaum[1], R. Coffee[1], F.-J. Decker[1], Y. Ding[1], D. Dowell[1], S. Edstrom[1], A. Fisher[1], J. Frisch[1], S. Gilevich[1], J. Hastings[1], G. Hays[1], Ph. Hering[1], Z. Huang[1], R. Iverson[1], H. Loos[1], M. Messerschmidt[1], A. Miahnahri[1], S. Moeller[1], H.-D. Nuhn[1], G. Pile[3], D. Ratner[1], J. Rzepiela[1], D. Schultz[1], T. Smith[1], P. Stefan[1], H. Tompkins[1], J. Turner[1], J. Welch[1], W. White[1], J. Wu[1], G. Yocky[1] and J. Galayda[1]

X-ray Free-Electron Lasers



Attosecond Science

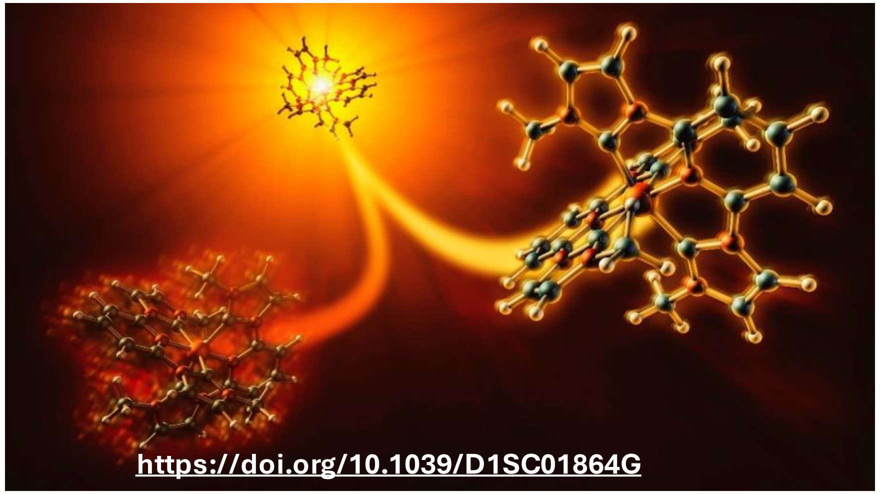


Simulation: G. Grell, F. Martin et al.

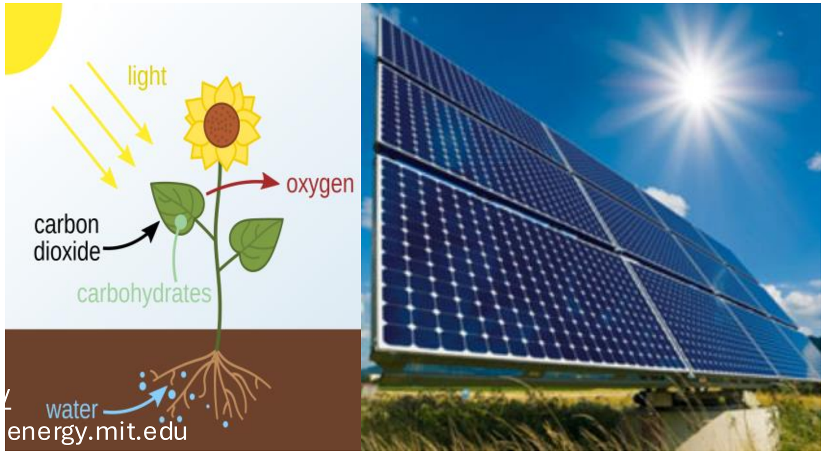
Attosecond Electron Dynamics

Attosecond science =
Study of electron dynamics

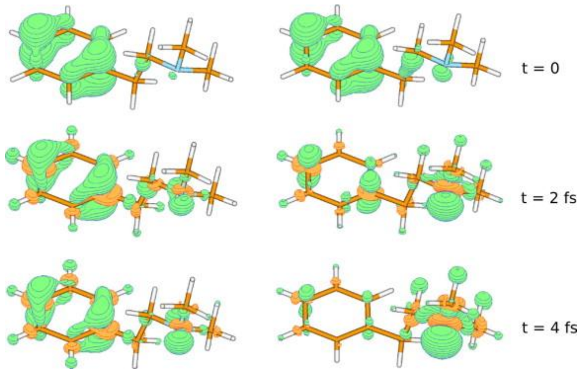
First step in chemistry



Radiation-matter interaction



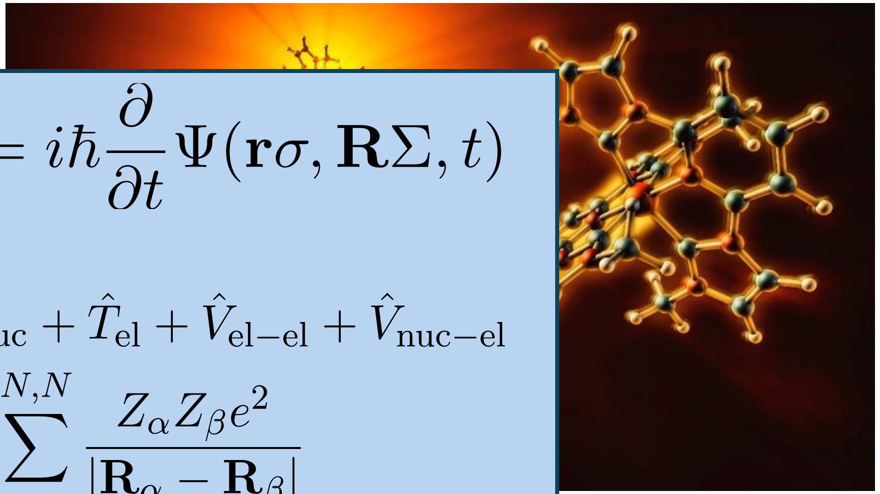
Multi-body quantum mechanics



J. Chem Phys. 8 129(10):104305 (2008)

Attosecond Electron Dynamics

First step in chemistry



Attosec

Study of e

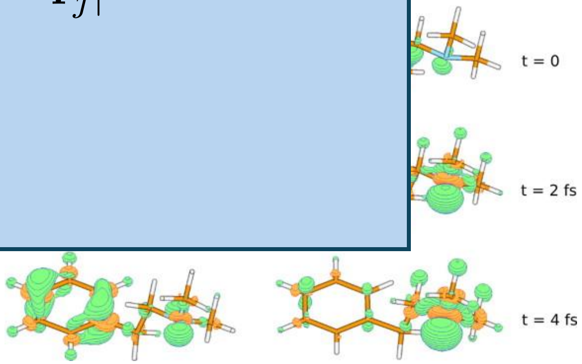
$$\hat{H}_{\text{mol}}\Psi(\mathbf{r}\sigma,\mathbf{R}\Sigma,t) = i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r}\sigma,\mathbf{R}\Sigma,t)$$

$$\begin{aligned}
\hat{H}_{\text{mol}} &= \hat{T}_{\text{nuc}} + \hat{V}_{\text{nuc-nuc}} + \hat{T}_{\text{el}} + \hat{V}_{\text{el-el}} + \hat{V}_{\text{nuc-el}} \\
&= \sum_{\alpha=1}^{N}\frac{\hat{P}_\alpha^2}{2M_\alpha} + \frac{1}{2}\sum_{\alpha,\beta}^{N,N}\frac{Z_\alpha Z_\beta e^2}{|\mathbf{R}_\alpha - \mathbf{R}_\beta|} \\
&\quad + \sum_{i=1}^{n}\frac{\hat{p}_i^2}{2m} + \frac{1}{2}\sum_{i,j}^{n,n}\frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} \\
&\quad - \sum_{\alpha=1}^{N}\sum_{i=1}^{n}\frac{Z_\alpha e^2}{|\mathbf{r}_i - \mathbf{R}_\alpha|}
\end{aligned}$$

Radiation

www.baumeiergroup.com

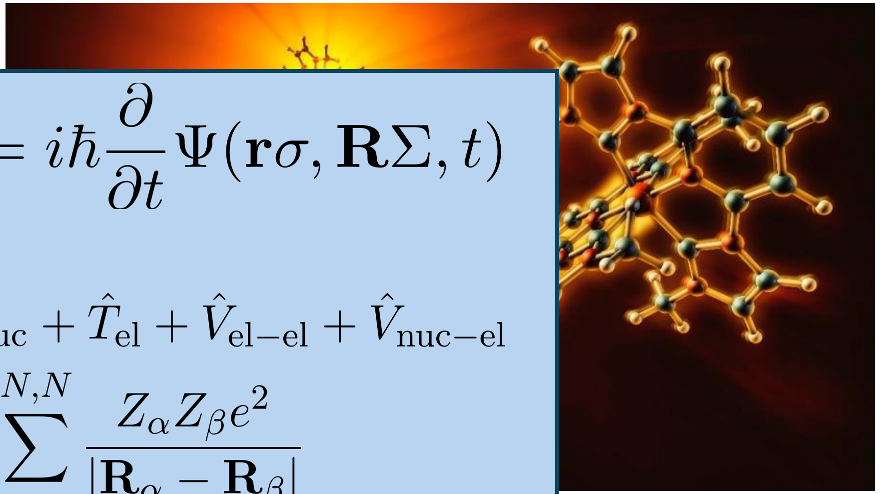
mechanics



J. Chem Phys. 8 129(10):104305 (2008)

Attosecond Electron Dynamics

First step in chemistry



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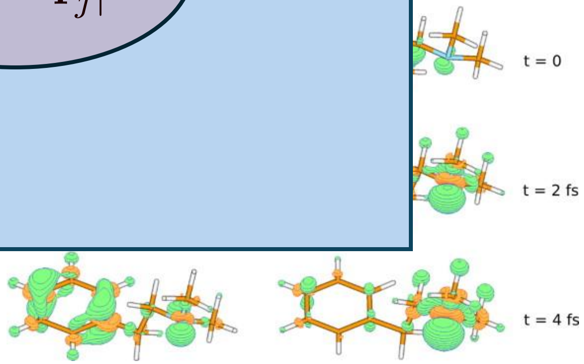
\end{aligned}$$

Radiation

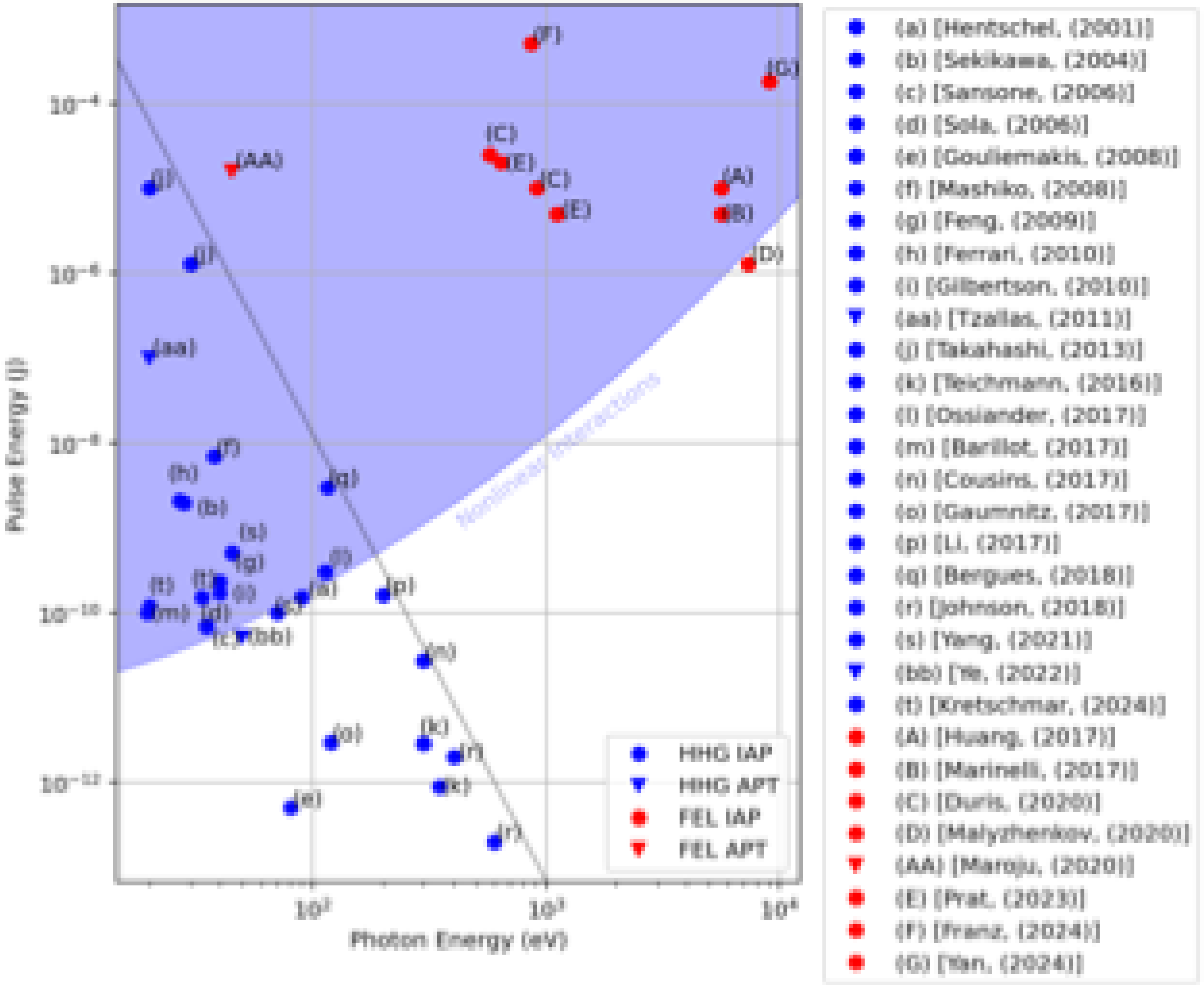
mechanics

www.baumeiergroup.com



J. Chem Phys. 8 129(10):104305 (2008)

Attosecond X-ray FELs

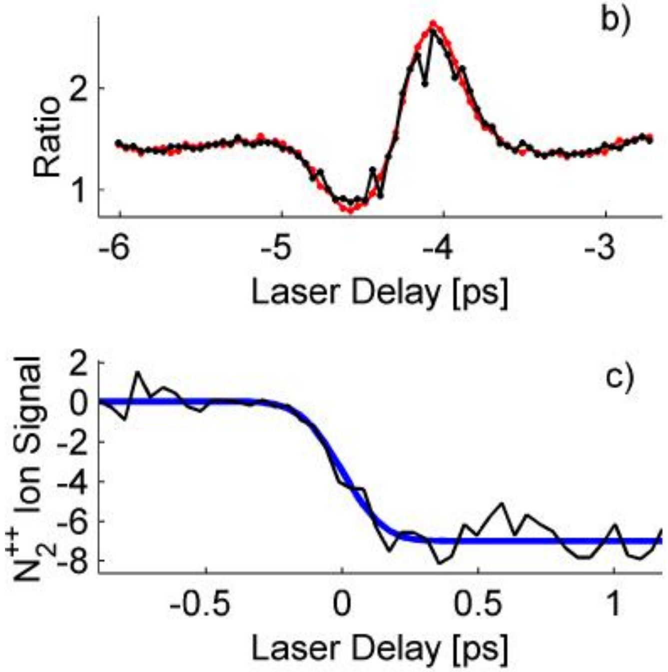


Three Orders of Magnitude Improvement in Time Resolution

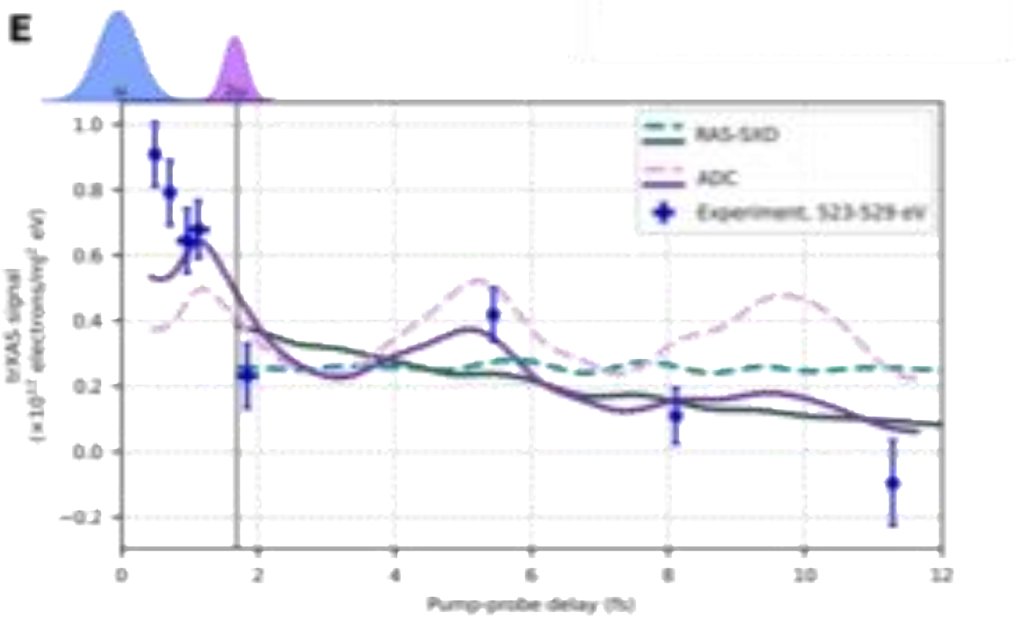
Time Resolution

Early Days	Time sorting	2-Color FELs	Attosecond FELs
2010	**2013-14**	**2013-16**	**2020-ongoing**

1 ps — **100 fs** — **10 fs** — **1 fs** — **0.1 fs**

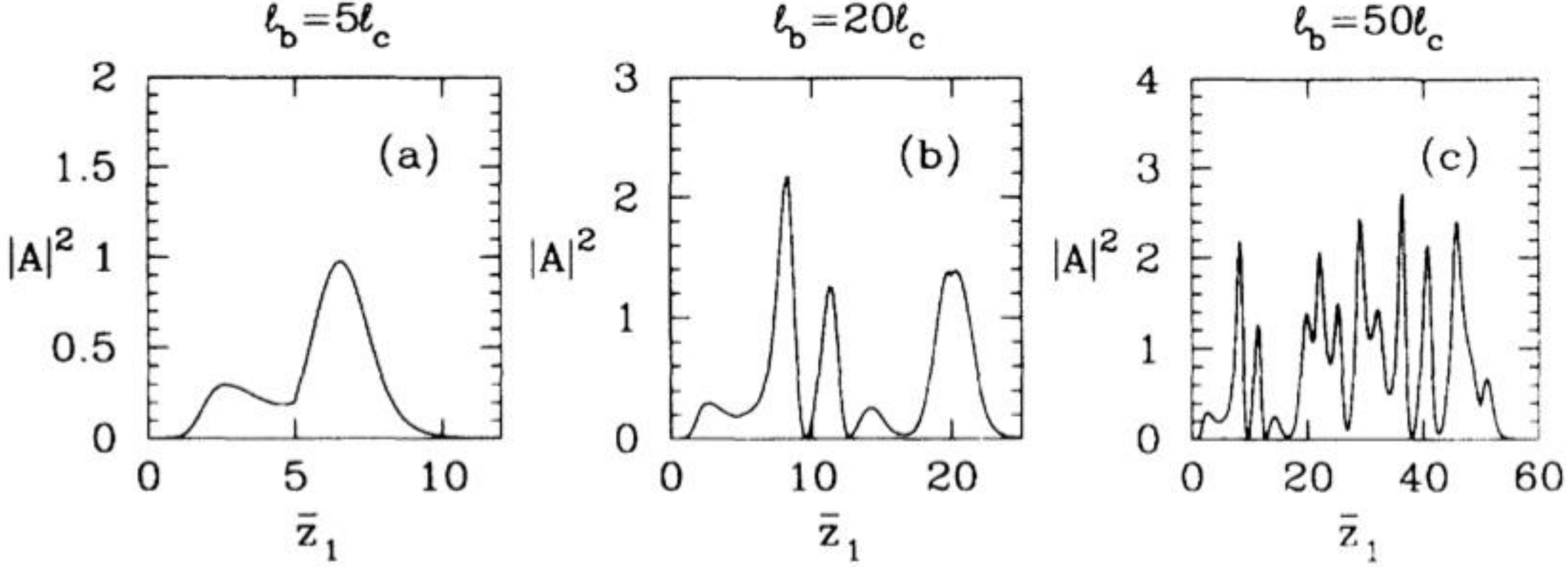


J. M. Glownia., et al *Optics express* 18.17 (2010): 17620-17630.



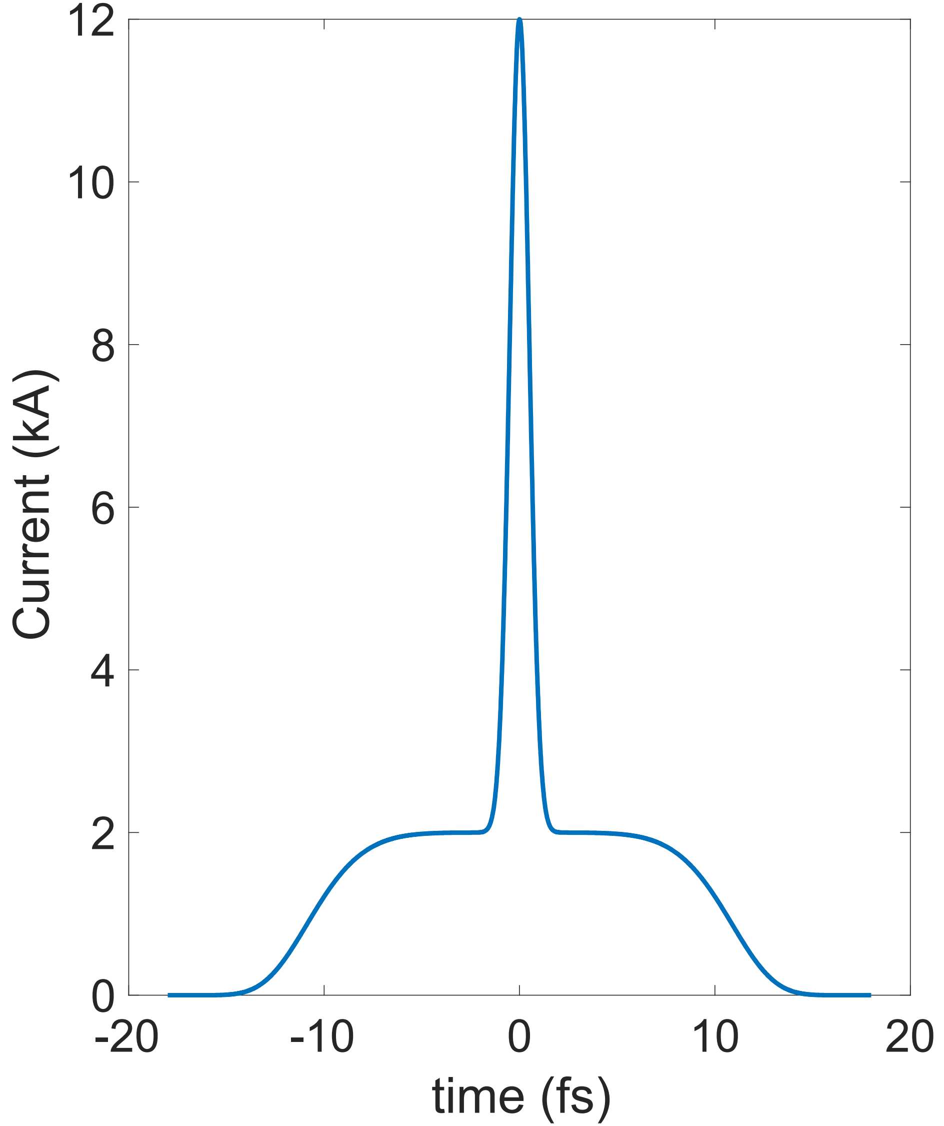
T. Driver et al. arXiv:2411.01700

SASE Spikes

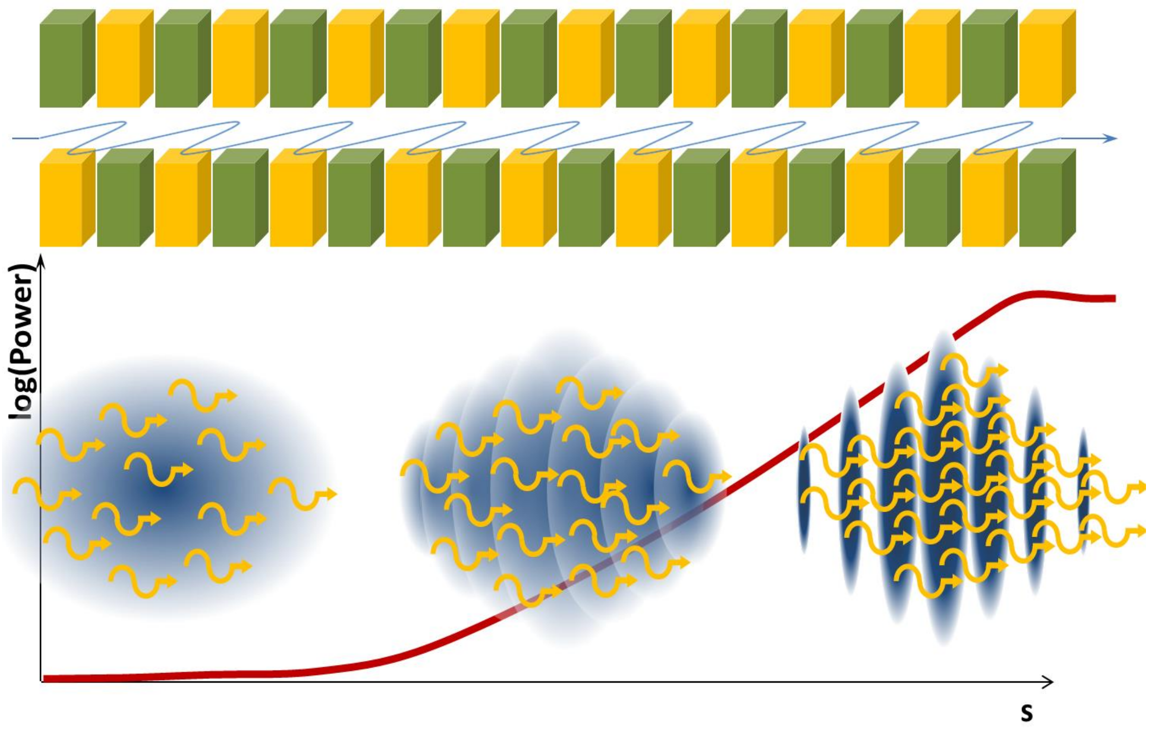


Bonifacio, R., et al. *Phys. Rev. Lett.* 73.1 (1994): 70.

Attosecond FELs

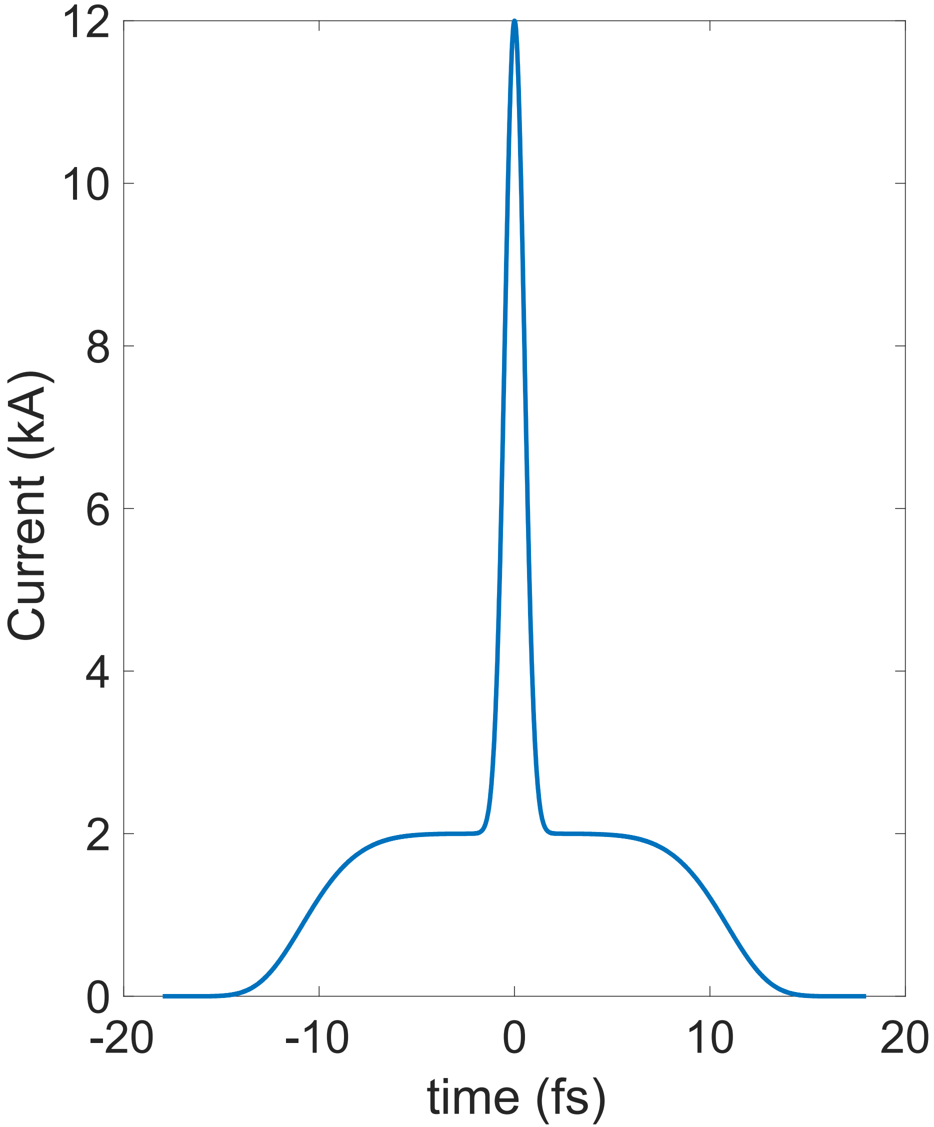


Attosecond FELs

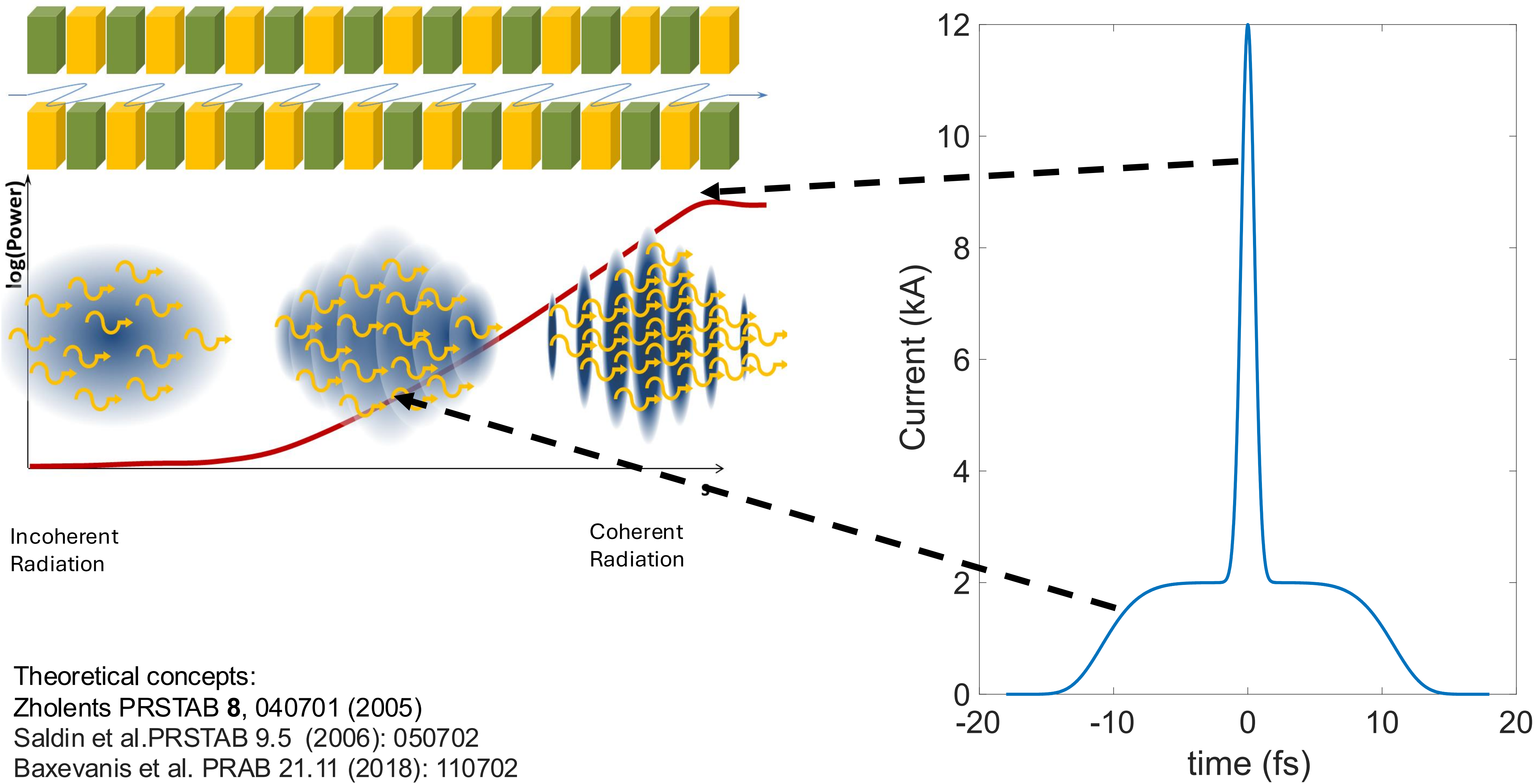


Incoherent
Radiation

Coherent
Radiation



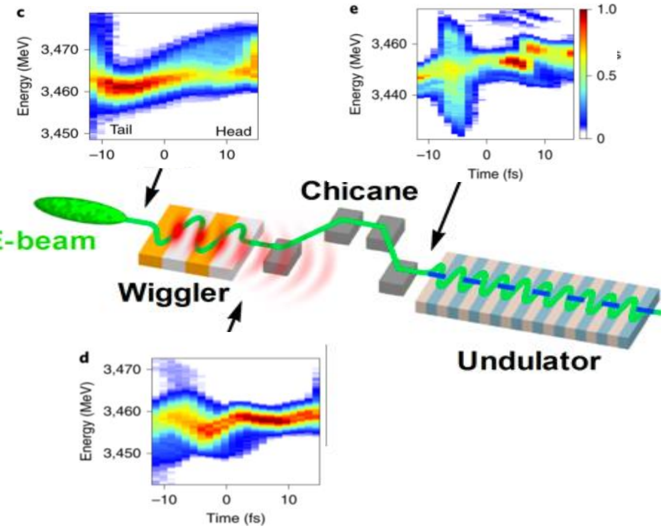
Attosecond FELs



Theoretical concepts:
Zholents PRSTAB **8**, 040701 (2005)
Saldin et al.PRSTAB 9.5 (2006): 050702
Baxevanis et al. PRAB 21.11 (2018): 110702

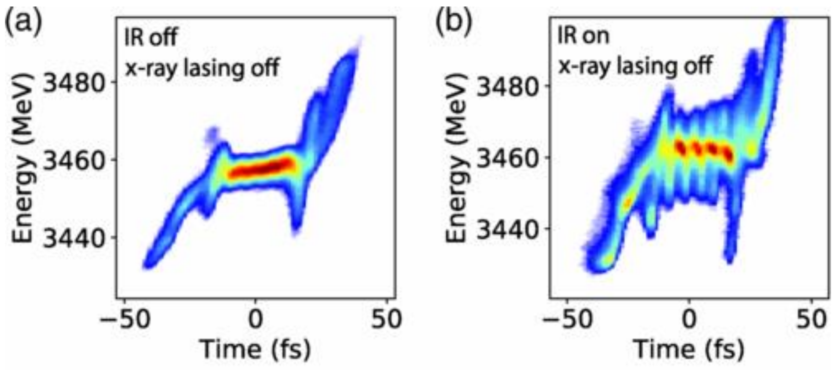
E-Beam Shaping

Self-modulation



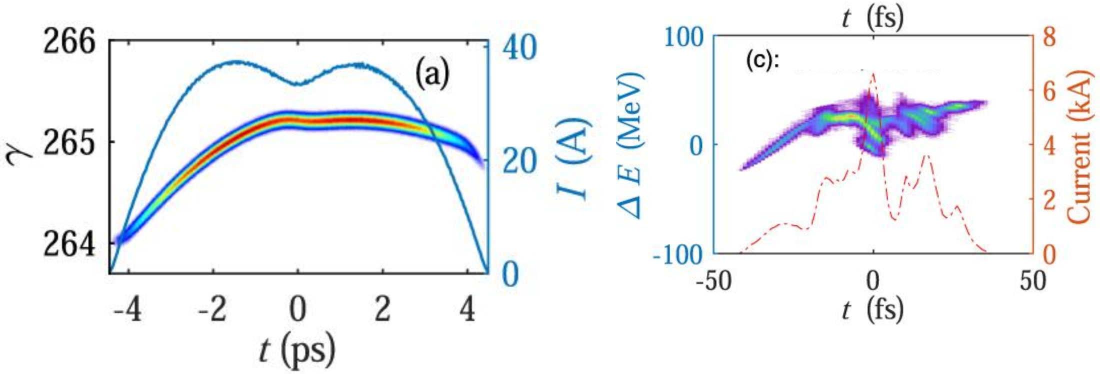
J. Duris, S. Li et al. *Nat. Photonics* 14.1 (2020): 30-36.

Laser modulation



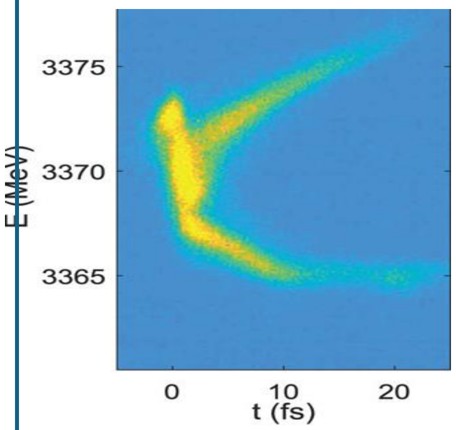
J. Duris et al. Phys. Rev. Lett. 126, 104802 (2021)

Seeded microbunching instability



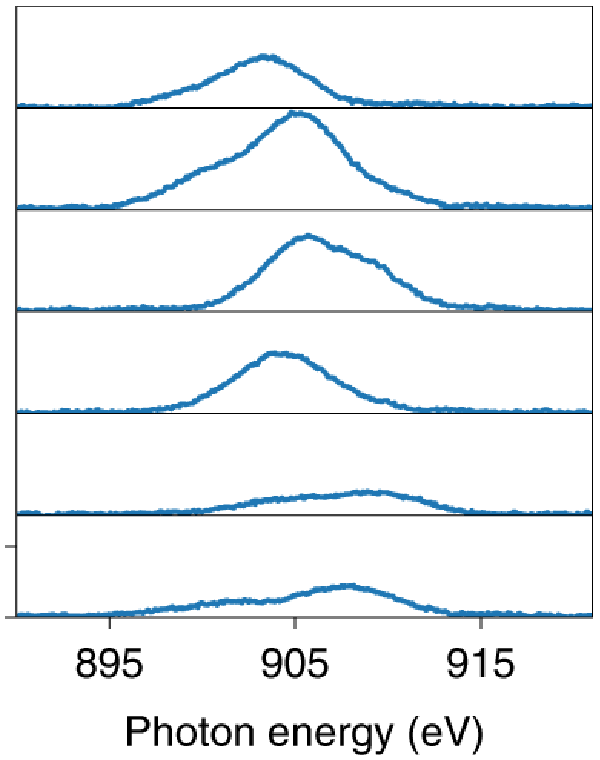
Zhang, Zhen, et al. *New Journal of Physics* 22.8 (2020): 083030.
S. Li et al. APL 125.19 (2024).

Non-linear compresion



S. Huang et al. PRL 119, 154801 (2017)
E. Prat et al. APL Photonics 8, 111302 (2023)
Yan et al. *Nat. Photonics* 18.12 (2024): 1293-1298.

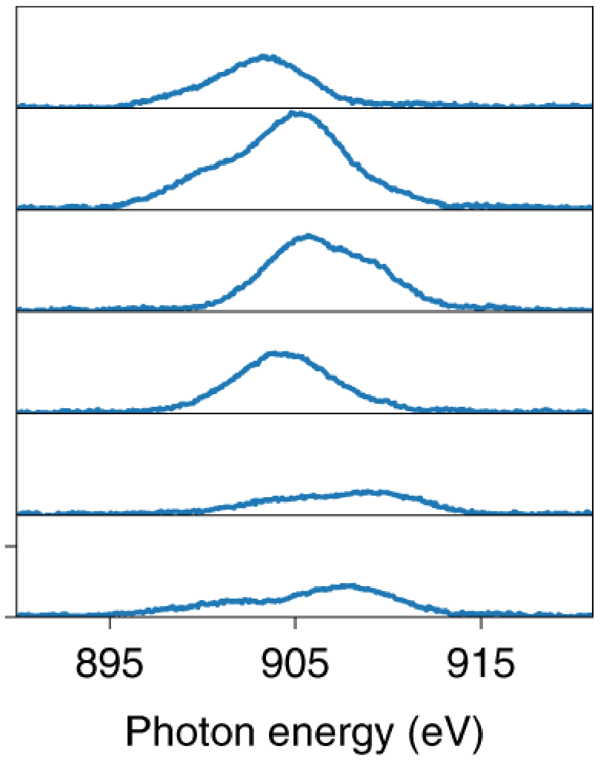
Typical Results

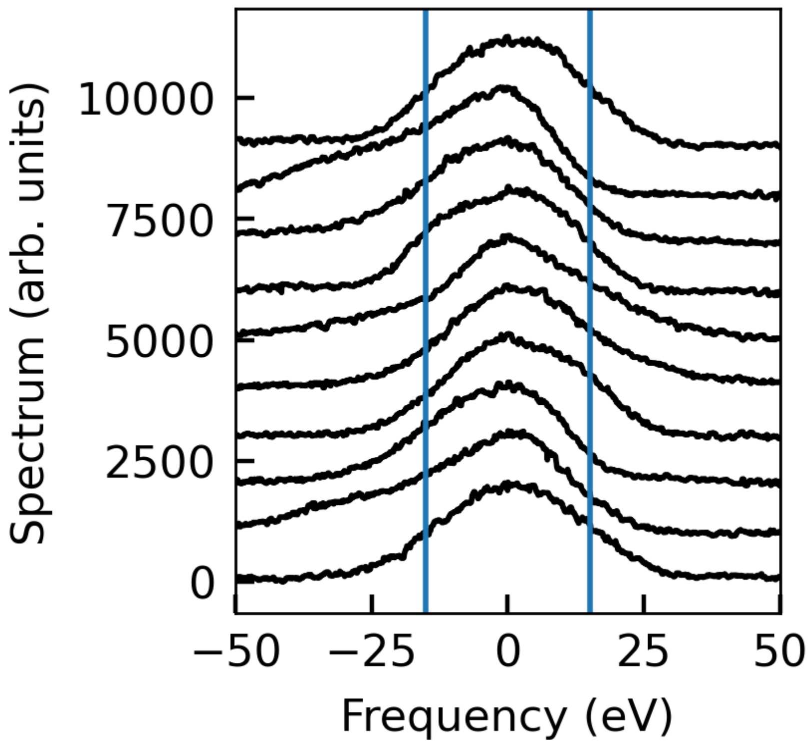


900 eV
<BW> = 7.1 eV
<Ep> = 10 uJ

J. Duris, S. Li et al. *Nat. Photonics* 14.1 (2020): 30-36.

Typical Results



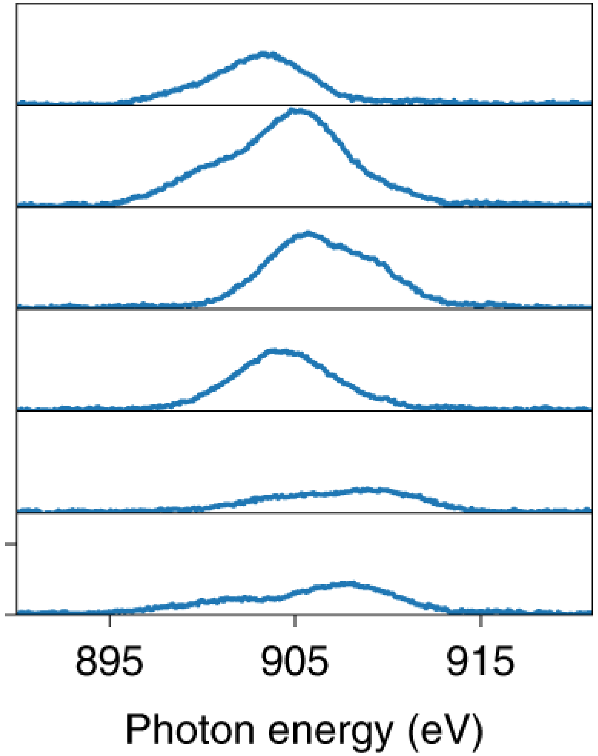


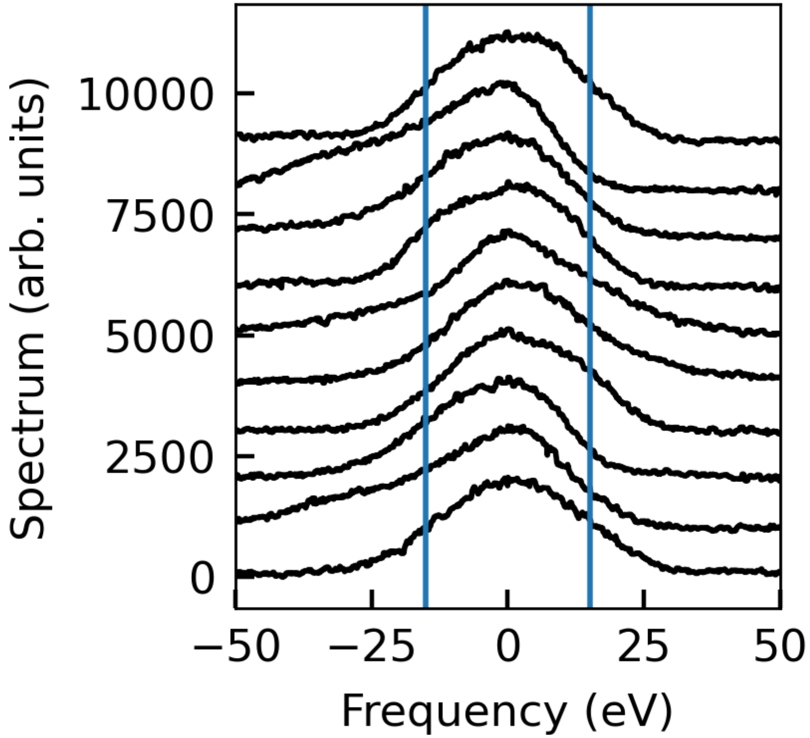
900 eV
<BW> = 7.1 eV
<Ep> = 10 uJ

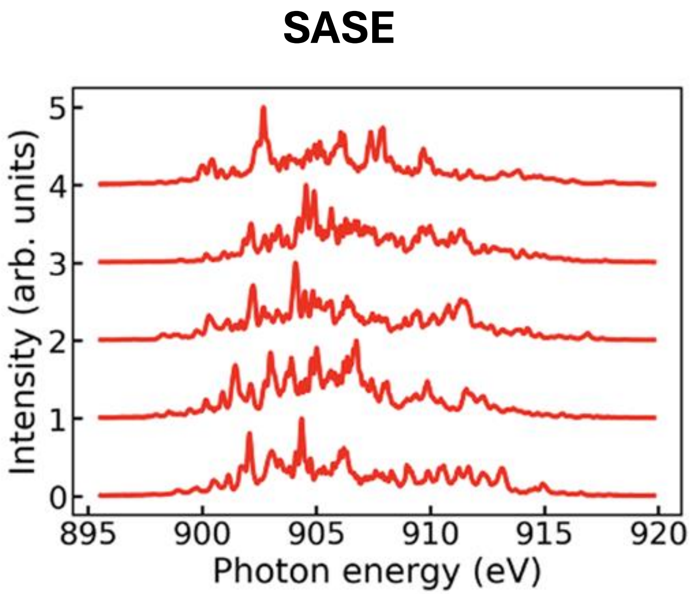
10 keV, unpublished
<BW> = 30 eV
<Ep> = 20 uJ

J. Duris, S. Li et al. *Nat. Photonics* 14.1 (2020): 30-36.

Typical Results







K. Li et al. Communications Physics **volume 5**, 191 (2022)

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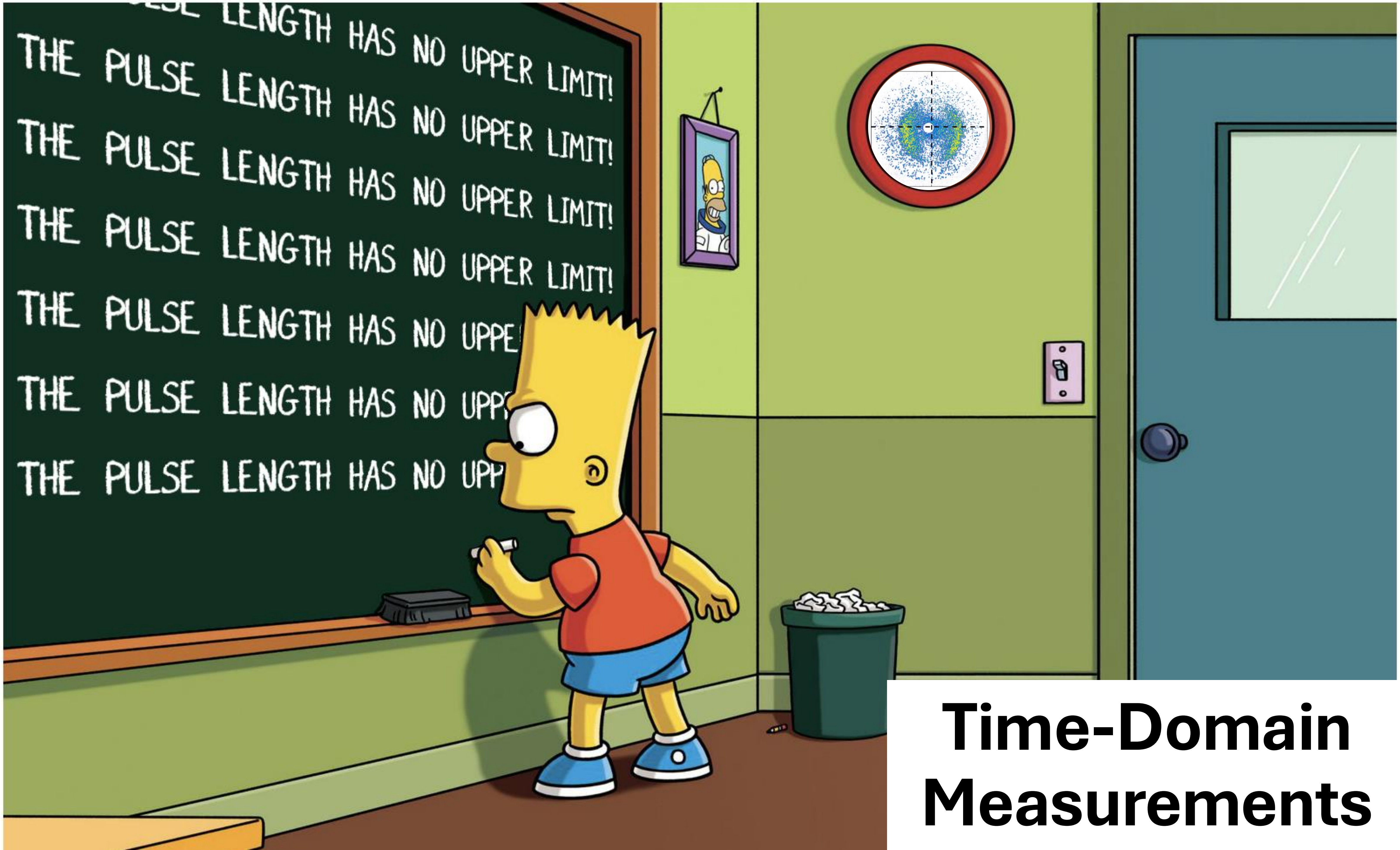

Time-Domain Measurements

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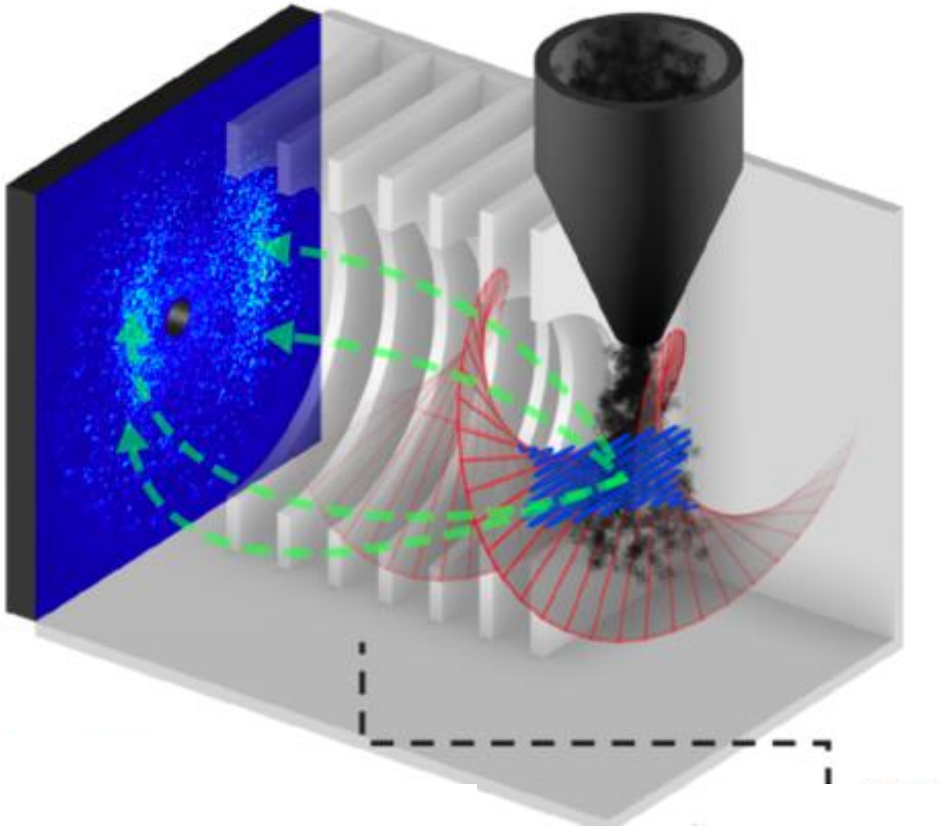
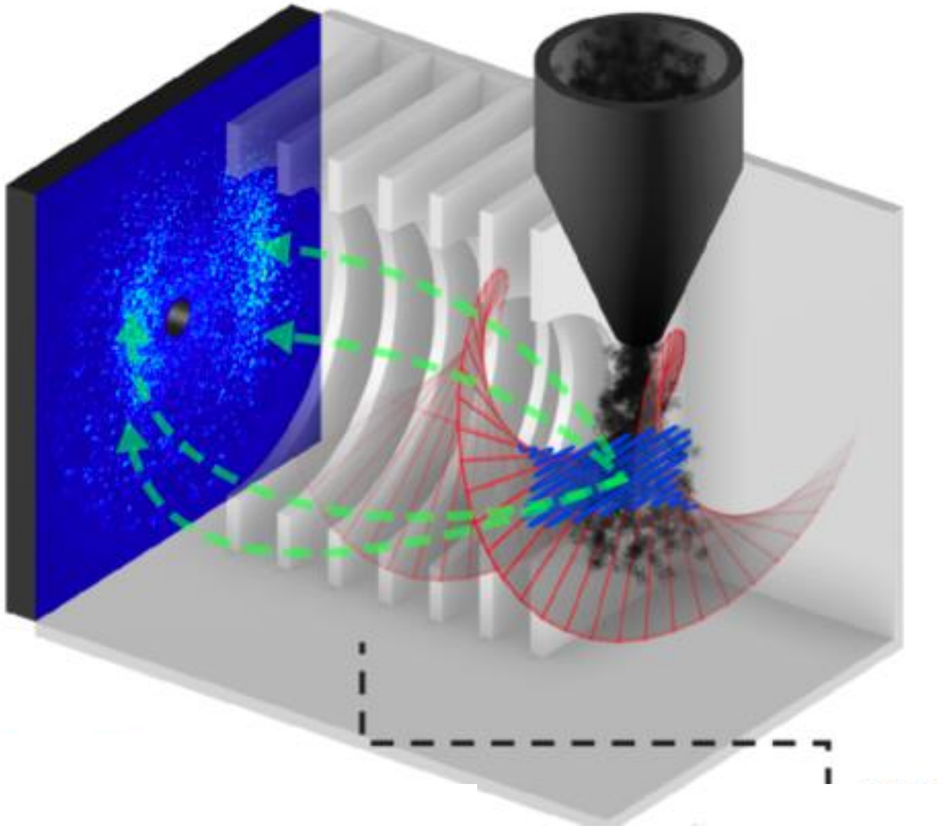


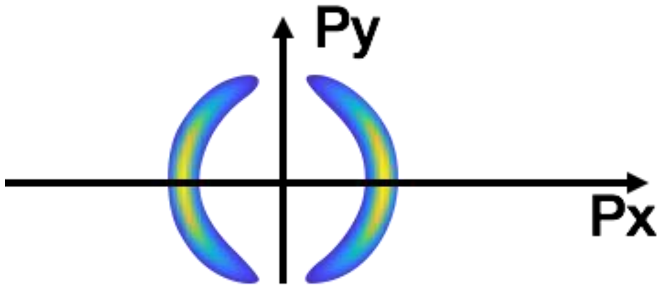
Original concept
P. Eckle et al. *Nature Physics* 4.7 (2008): 565-570.

Application to femtosecond XFELs
N. Hartmann
Nature Photonics 12.4 (2018): 215-220.

Application to attosecond XFELs:
S. Li *et al.* Optics Express 26 4531 (2018)
J. Duris, S. Li et al. *Nat. Photonics* 14.1 (2020): 30-36.

Time-Domain Measurements





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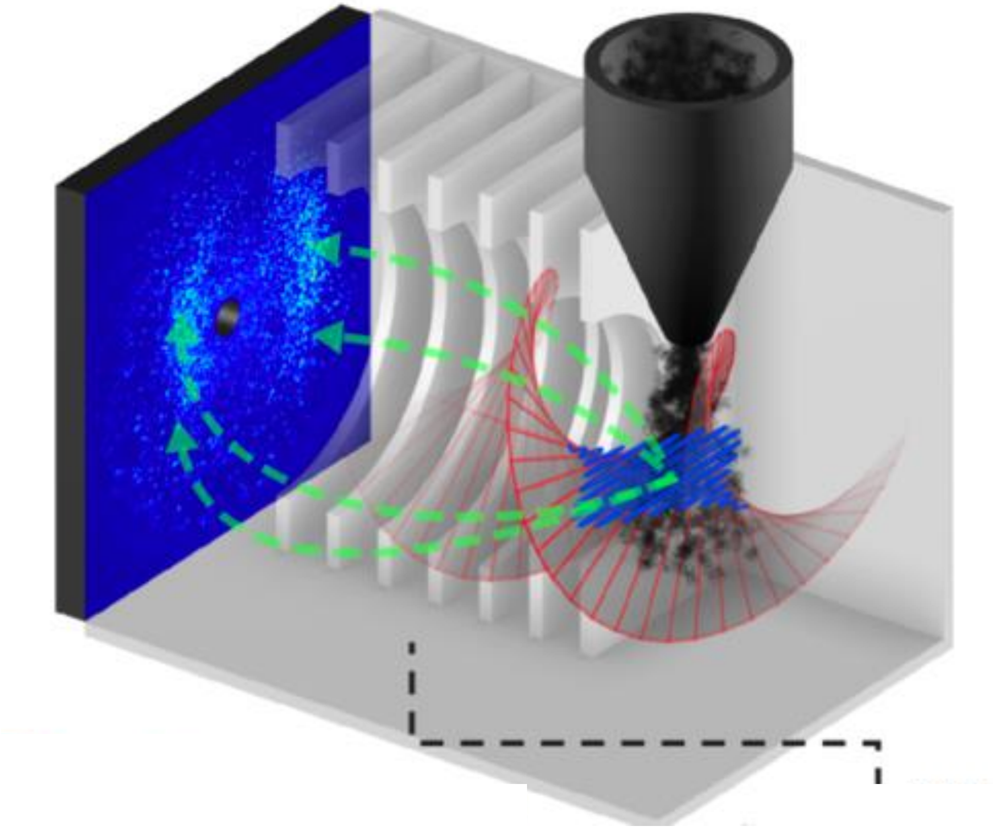

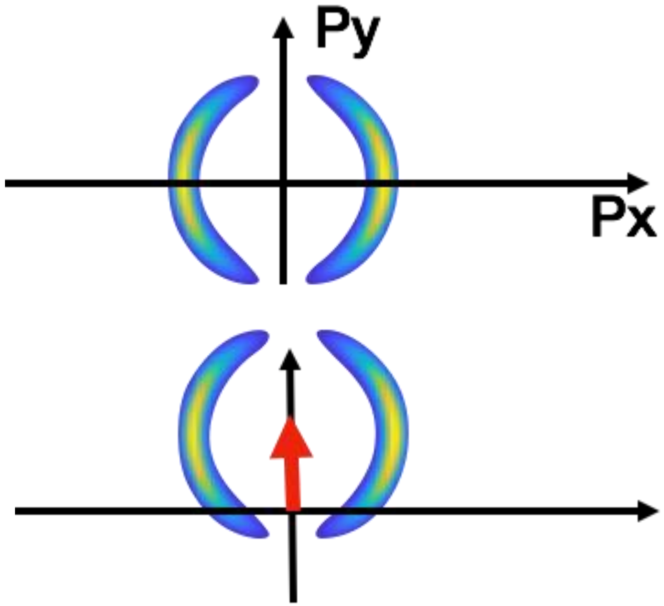

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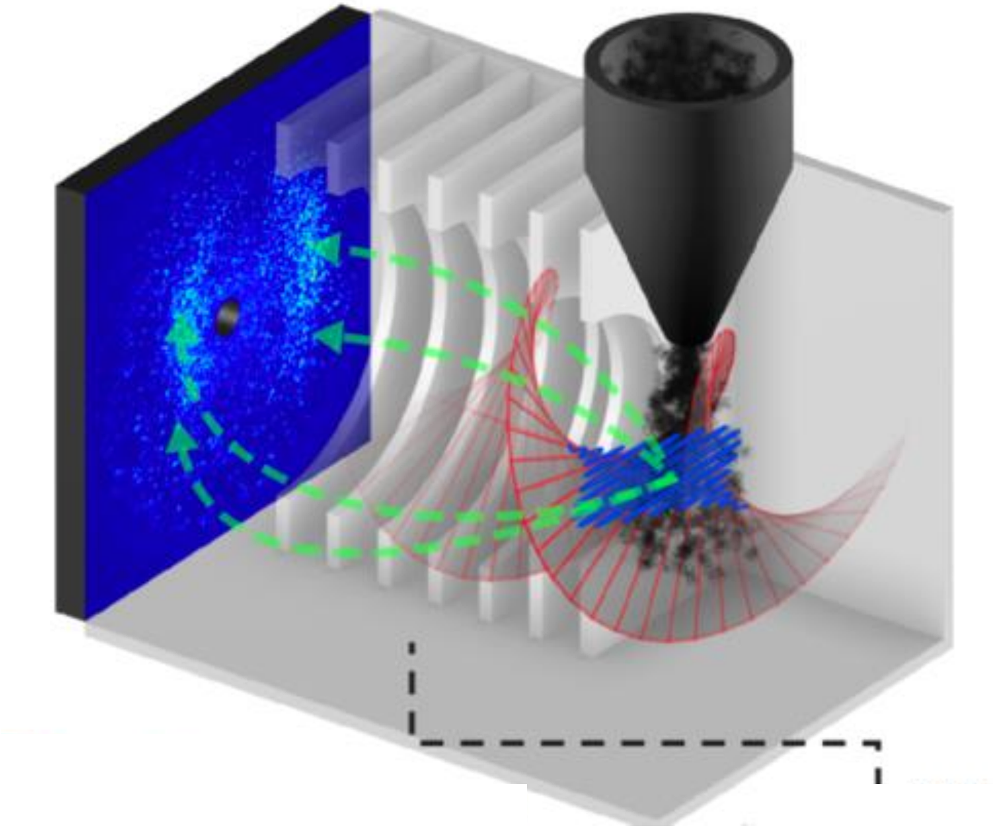

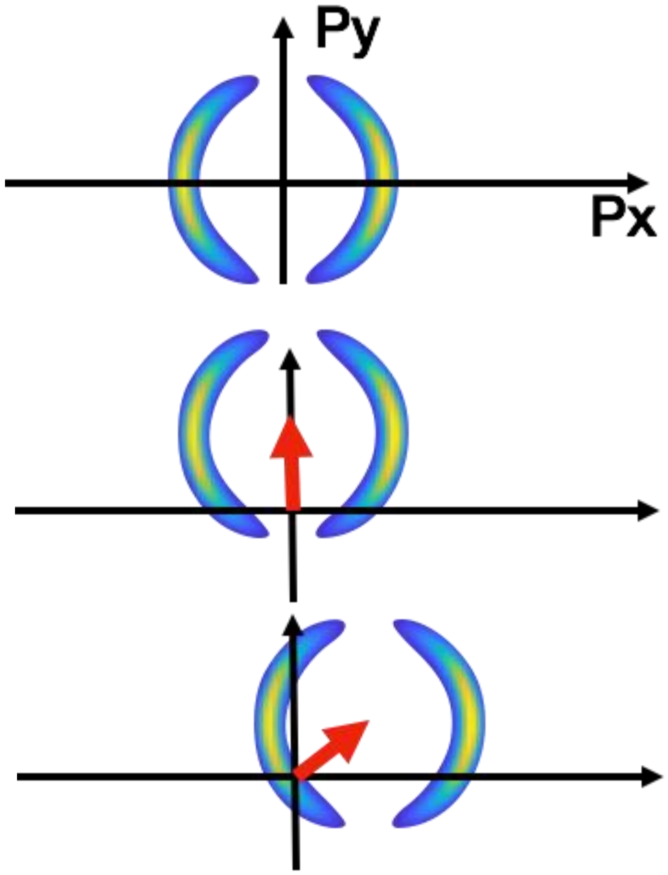

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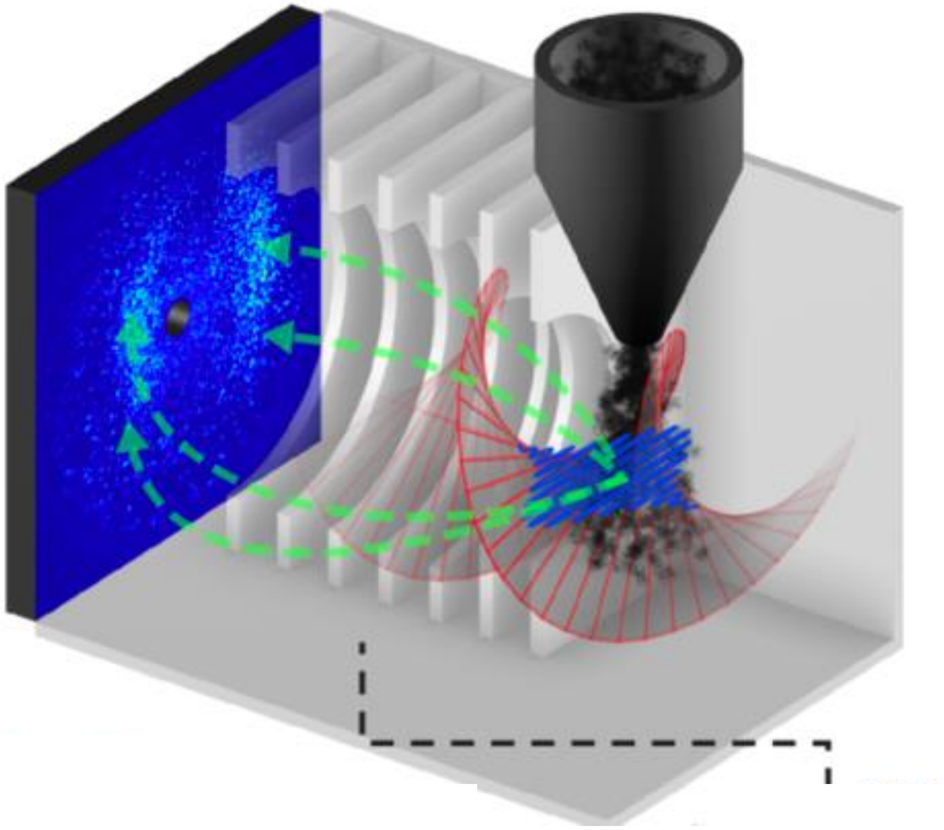
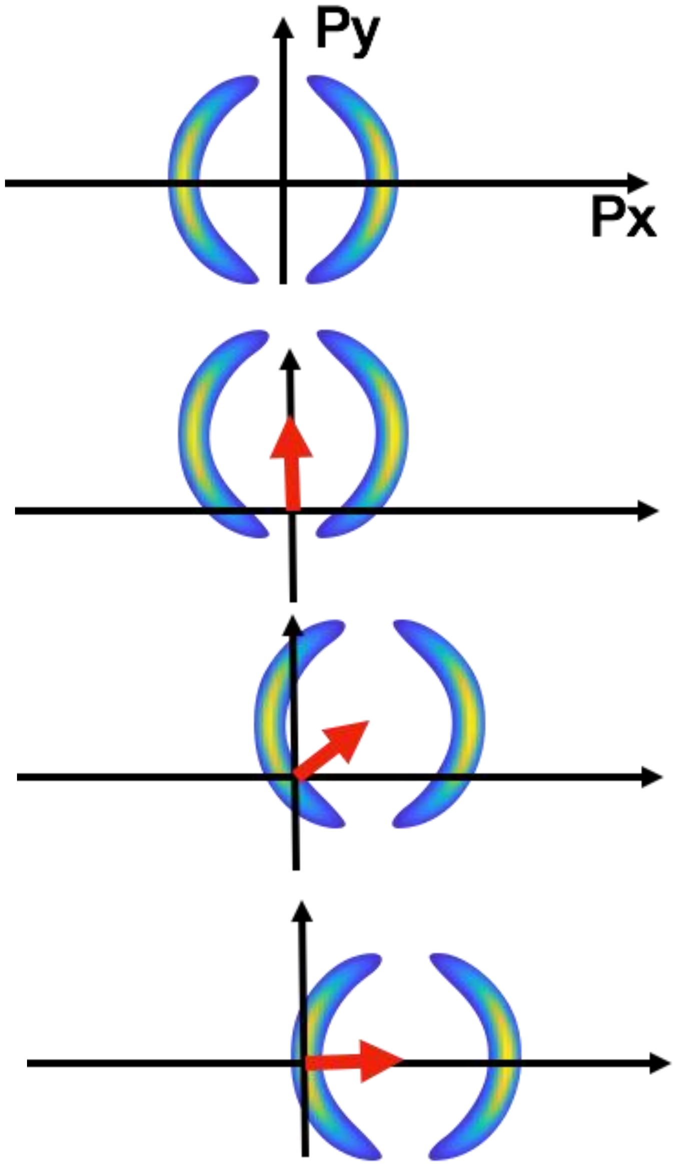
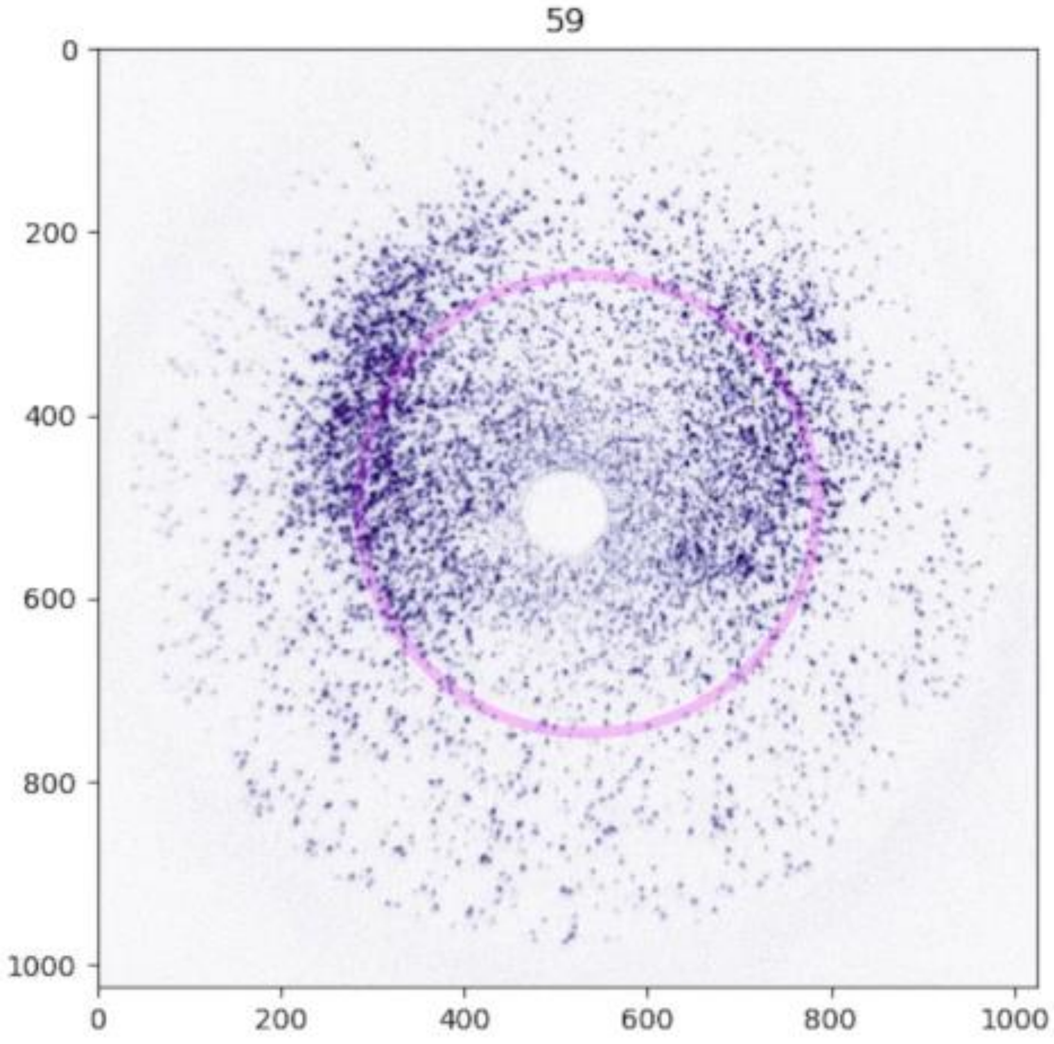




Raw Data



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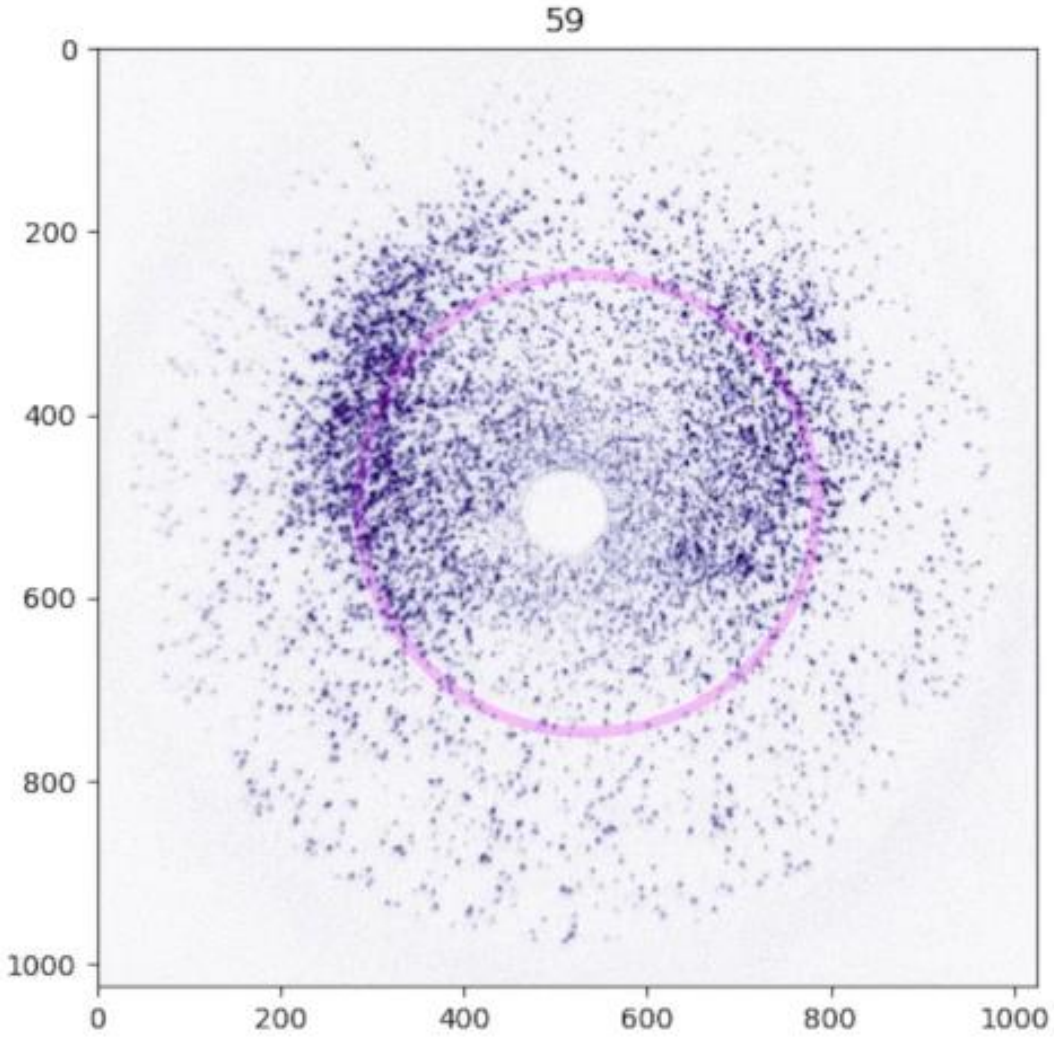
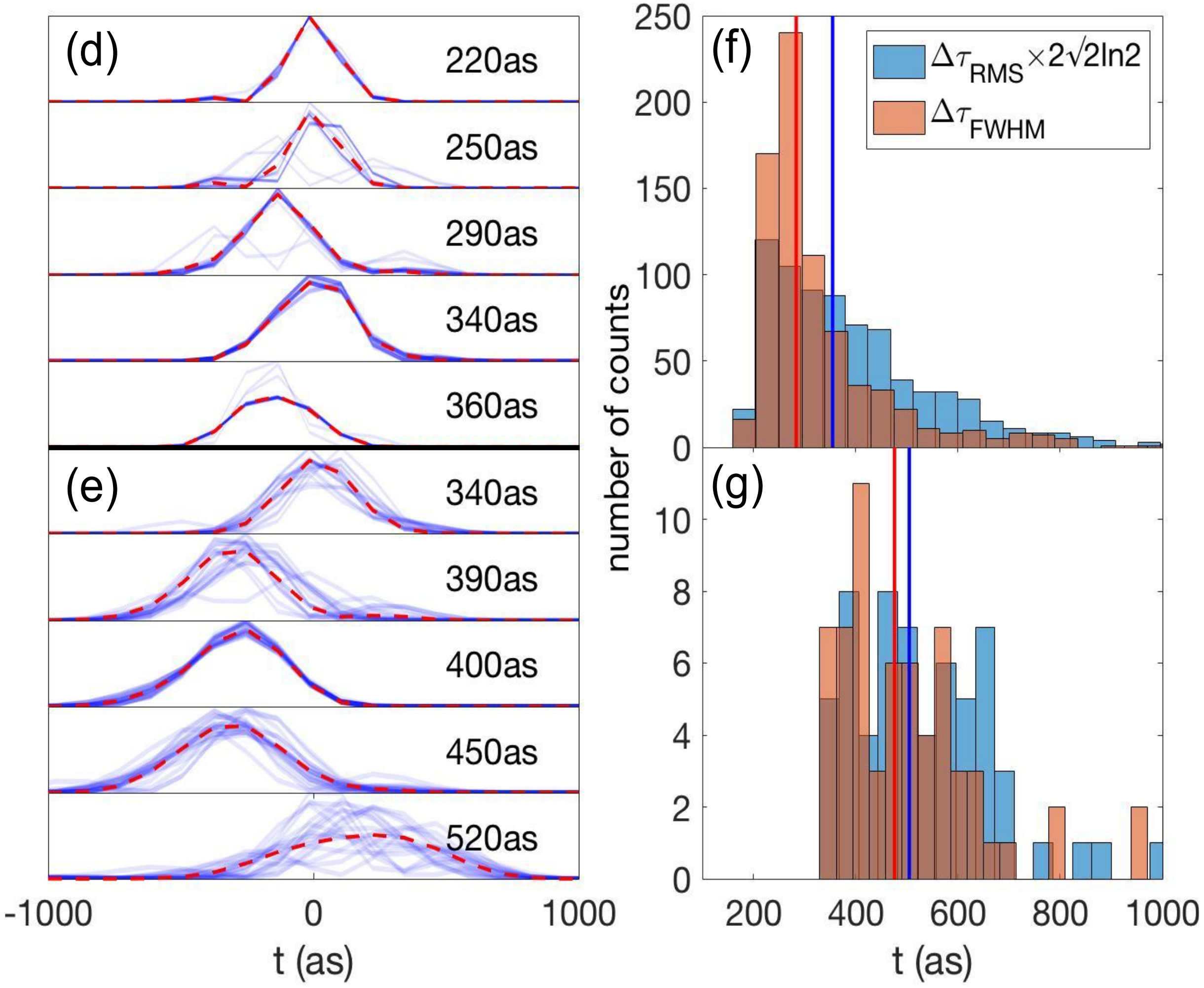




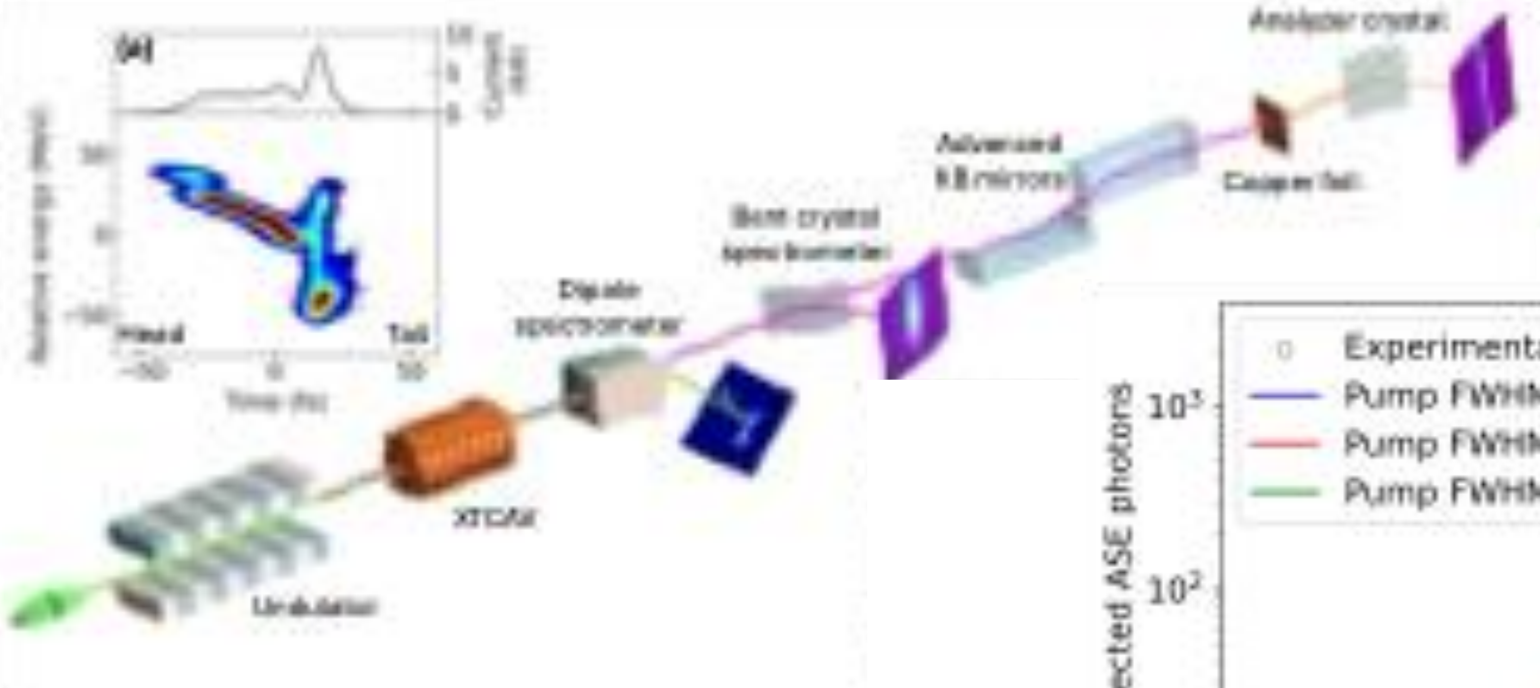
J. Duris, S. Li et al. *Nature Photonics* 14.1 (2020): 30-36.

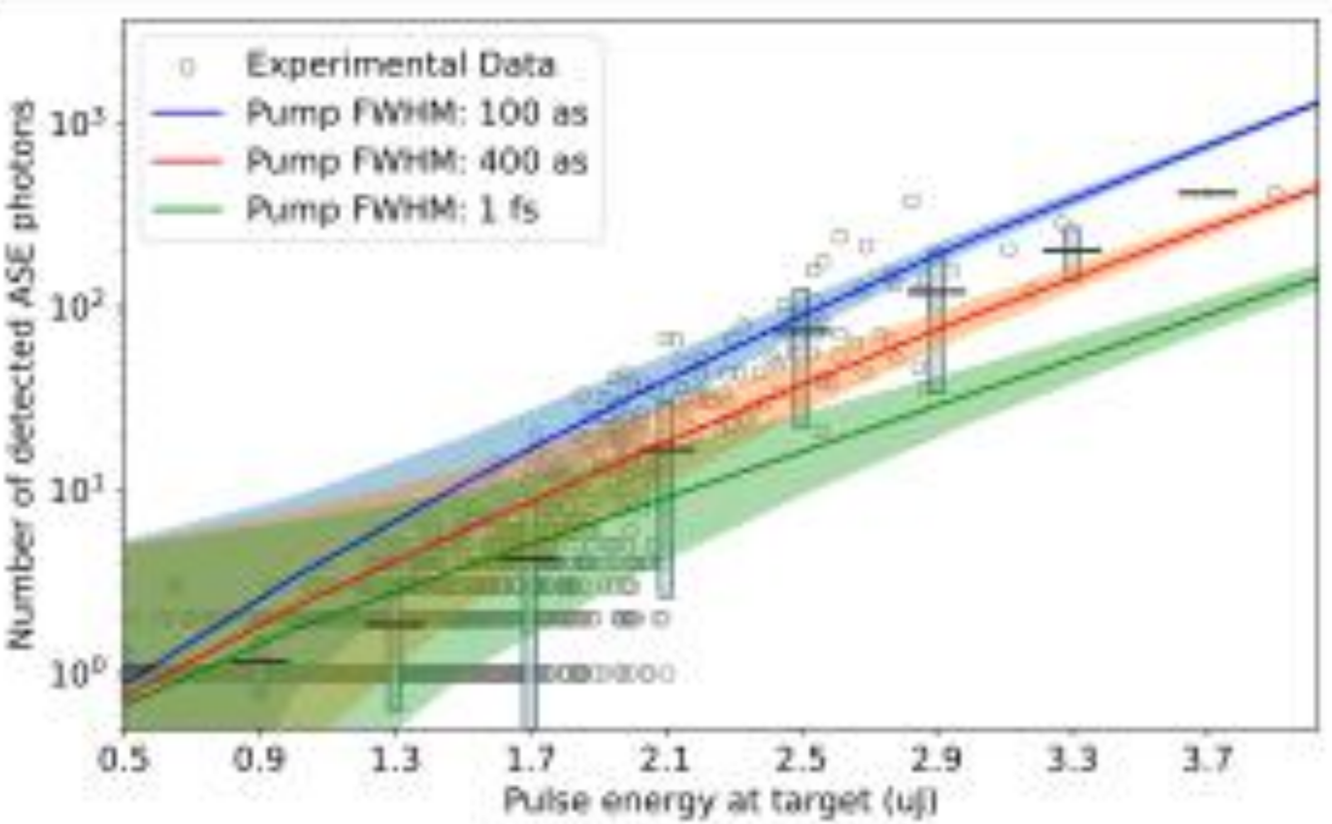
S. Li et al. Optics Express Vol. 26, [Issue 4](), pp. 4531-4547 (2018)

First Experimental Evidence of Sub-Fs Hard X-ray Pulses

I. Inoue, R. Robles,
A. Halavanau … A. Marinelli
arXiv preprint arXiv:2506.07968

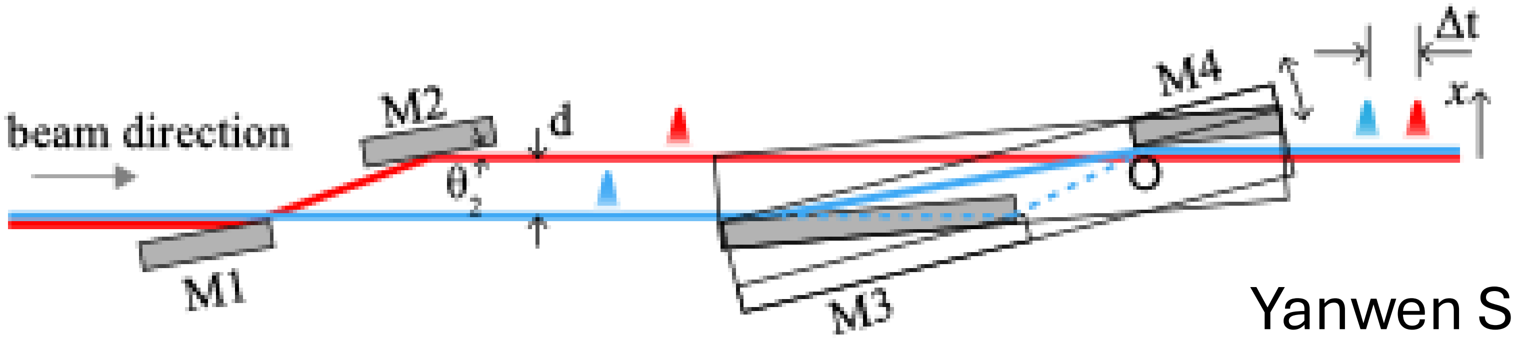
Under Review, Nature Photonics





Comparison with 3-D Model of ASE
AND Literature value of ASE threshold
Pulse Duration ~ 100-400 as FWHM

Attosecond Autocorrelation Measurement



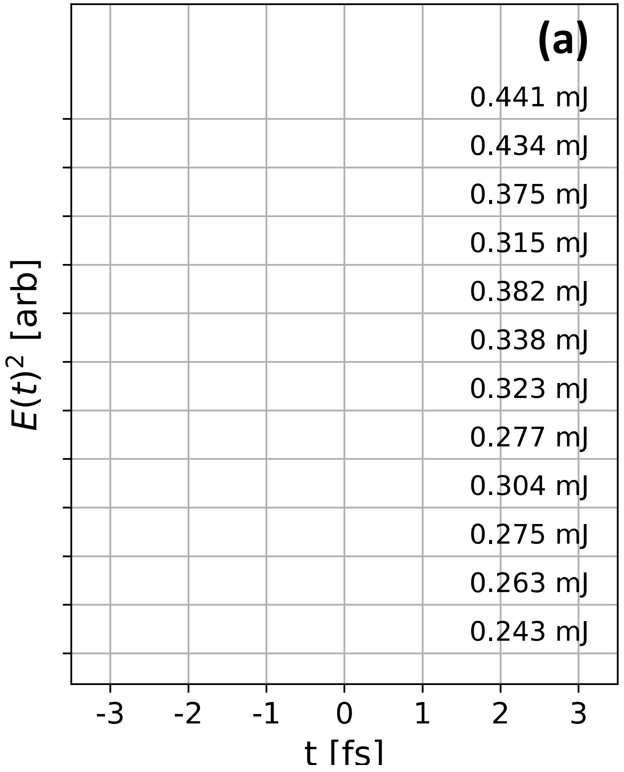
Yanwen Sun et al. arXiv:25

130 as FWHM
40-50 uJ pulse energy
30 eV Bandwidth

Advanced Attosecond Modes

TW-scale Pulses

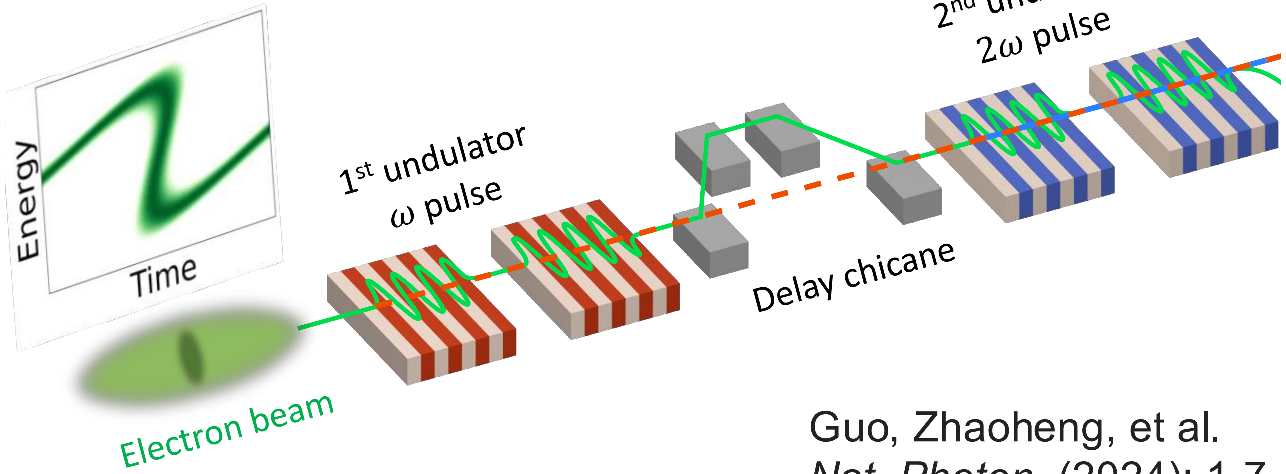
P. Franz et al. *Nat. Photonics* 18.7 (2024): 698-703



$$\frac{d^2 b}{dx^2} - b + 3b|b|^2 = 0$$

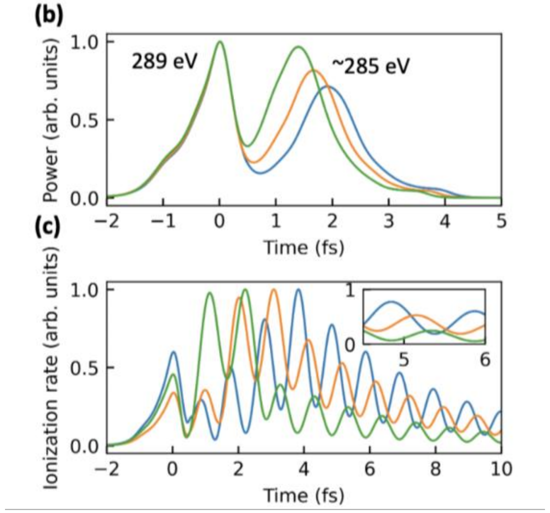
R. Robles et al. Phys. Rev. Research 6, 033158

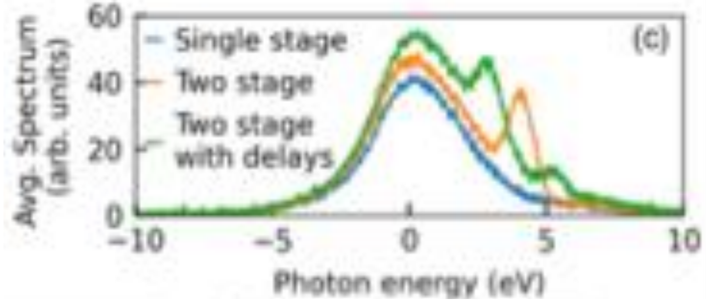
Attosecond Pulse Pairs



Guo, Zhaoheng, et al. *Nat. Photon.* (2024): 1-7.

Pulse shaping





R. Robles et al. Phys. Rev. Lett. **134**, 115001 (2025)

Soliton-Like Superradiance

R. Bonifacio et al. / The superradiant regime of a FEL 363

The solution of eqs. (28) does not depend on z_1 and δ, but only on the initial conditions in $y = 0$: $A_1(0) = \langle \exp(-i\theta_{10}) \rangle = b_0$, $p_{1j}(0) = 0$. It follows that $|A|^2 \propto z_1^2 \propto \rho^2$; since $\rho \propto N^{1/3}$ and $|E|^2 \propto \rho N |A|^2$, then $|E|^2 \propto N\rho^3 \propto N^2$, and the peak intensity scales as the square of the electron current; the width of the pulse scales as $z_1^{-1/2}$, i.e. as the inverse of the square root of the peak field. Furthermore, the factor $\exp(i\delta z_1)$ moves the signal to the resonance: in fact, from eq. (2), $E \exp[ik(z - ct)] \propto A \exp[ik(z - ct) - i\delta\bar{z}] \propto A_1 \exp[ik(z - ct) - i\delta z_2]$, where $z_1 + z_2 = \bar{z}$ has been used. From eqs. (5, 7) it follows that $\delta z_2 = (k_s - k)(ct - z)$, so that $E \propto \exp[ik_s(z - ct)]$.

Setting $x \equiv y^2$, eq. (28c) reads

$$(d/dx)(xA_1) = \langle \exp(-i\theta_1) \rangle \equiv b_1,$$

so that by integration

$$A_1 = \frac{1}{x}\int_0^x dx' b_1(x') \equiv \bar{b}_1, \tag{29}$$

where $\bar{b}_1$ is just the average of the bunching parameter with respect to x. Eq. (29) is a generalization of the adiabatic expression $A_1 = b_1$ of ref. [4].

In fig. 2 the intensity $|A_1|^2$, from the numerical integration of eqs. (28), is plotted vs y (dashed line); the result is compared with the output (continuous line) of the numerical code [5], integrating the system (9) in the short-pulse limit (see the captions for the parameters): $|A|^2$ is divided by z_1^2 and plotted vs $y = \sqrt{z_1}(\bar{z} - z_1)$, for $z_1 = 1/K$ (at the pulse head). The agreement turns out to be very satisfactory for nearly the first three peaks of the intensity, demonstrating the validity of the similarity transformation (27). It is interesting that the first pulse of the intensity in fig. 2 is very well described by the expression

$$|A_1|^2 = \frac{|b_0|^2}{3\pi} \frac{\exp\left[3\sqrt{3}(y/2)^{2/3}\right]}{\left\{y + \left(|b_0|^2/12\pi\right)\exp\left[3\sqrt{3}(y/2)^{2/3}\right]\right\}^2} = \frac{1}{y}\,\mathrm{Sech}^2\left[\frac{3\sqrt{3}}{2}\left(\frac{y}{2}\right)^{2/3} + \log\left(\frac{|b_0|}{2\sqrt{3\pi y}}\right)\right]. \tag{30}$$

In the next section we will discuss the validity of the similarity equations (28) also for the long-pulse case.

Superradiant self-similar solution:

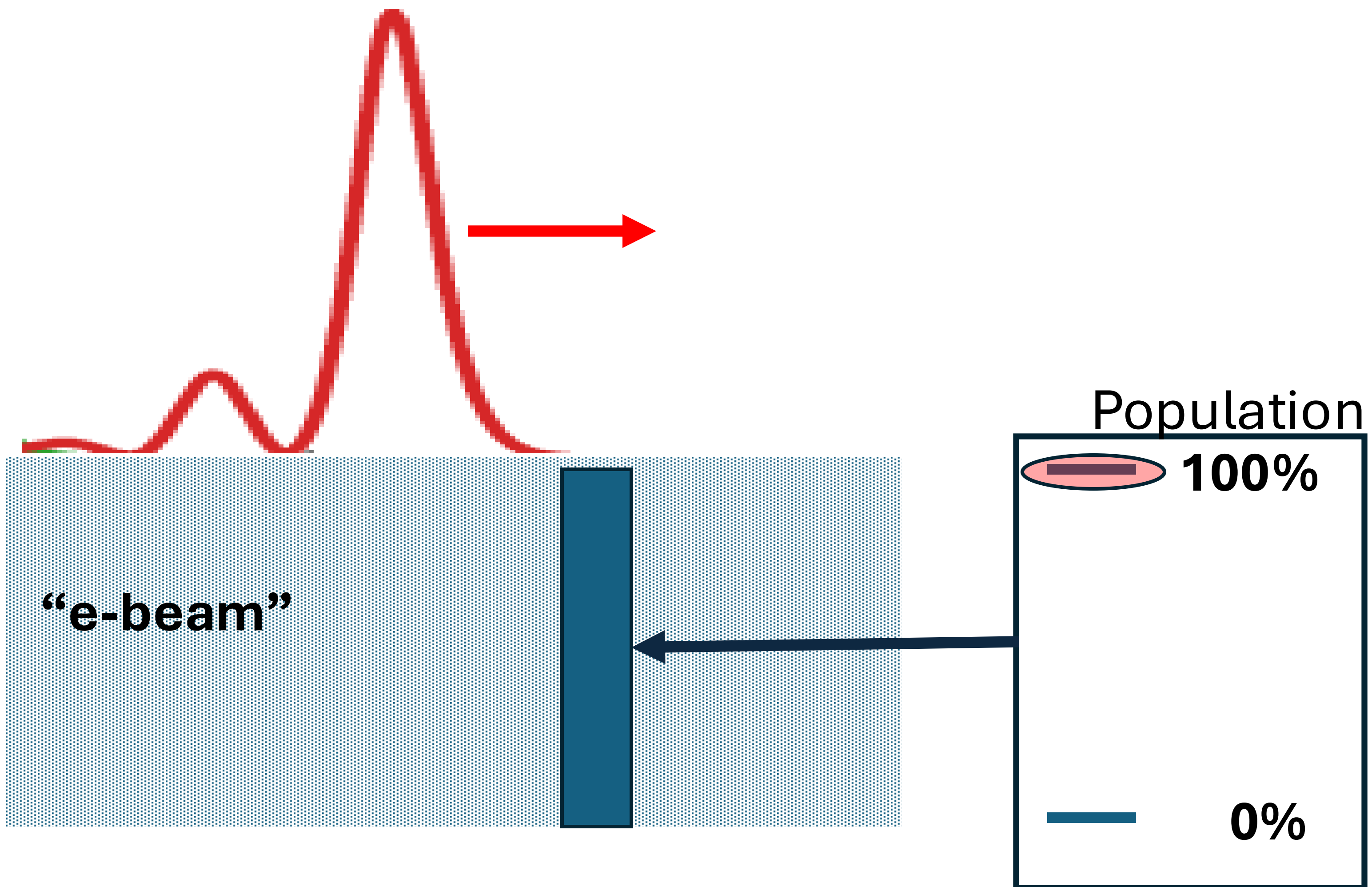
$P \sim z^2$
$\Delta t \sim z^{1/2}$

In practice pulse shrinking effect saturates due to diffraction…

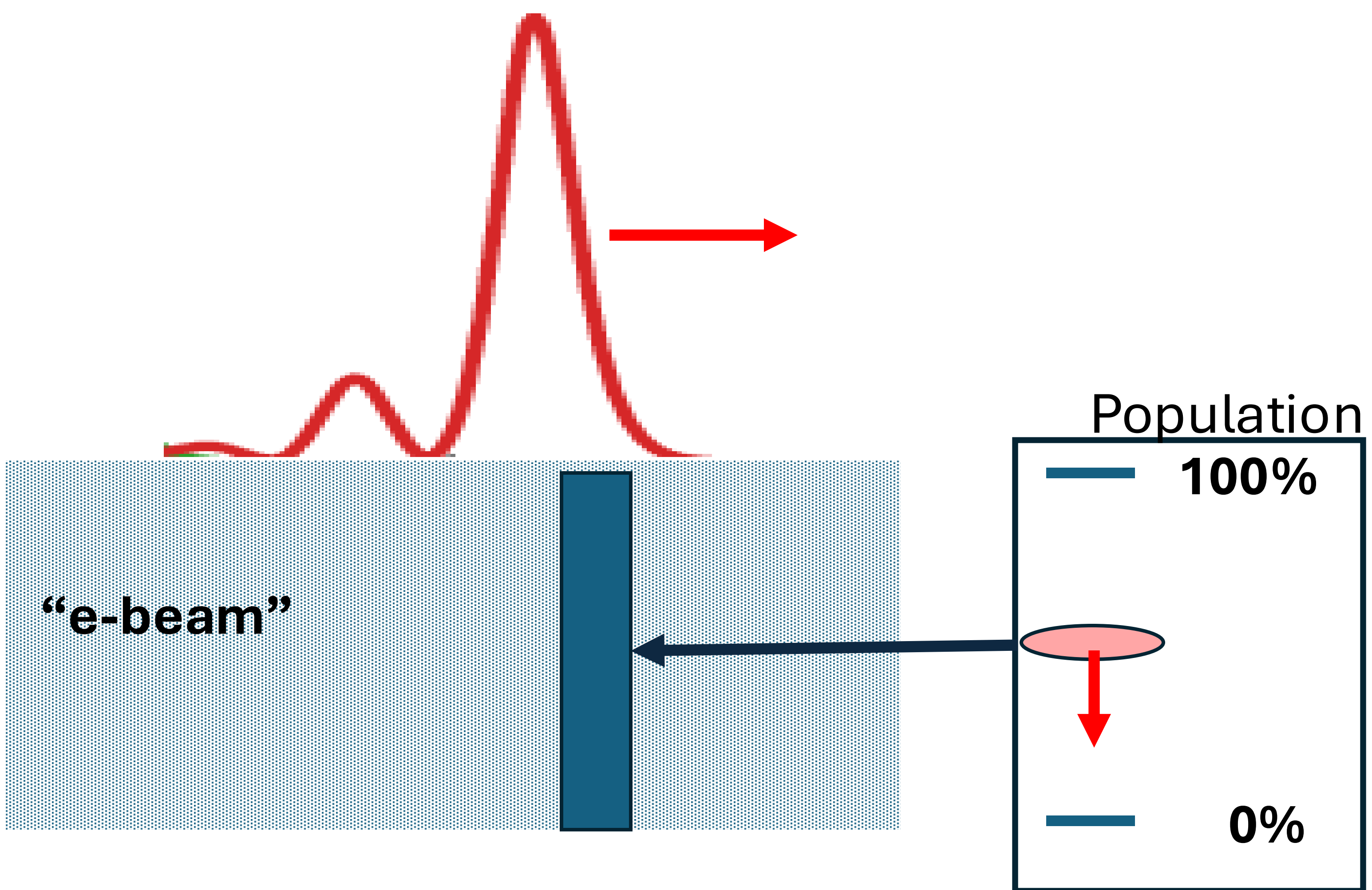
Robles et al. (2024)

Bonifacio et al.
Nuclear Instruments and Methods inPhysics Research A296 (1990) 358-367

Superradiance



Superradiance



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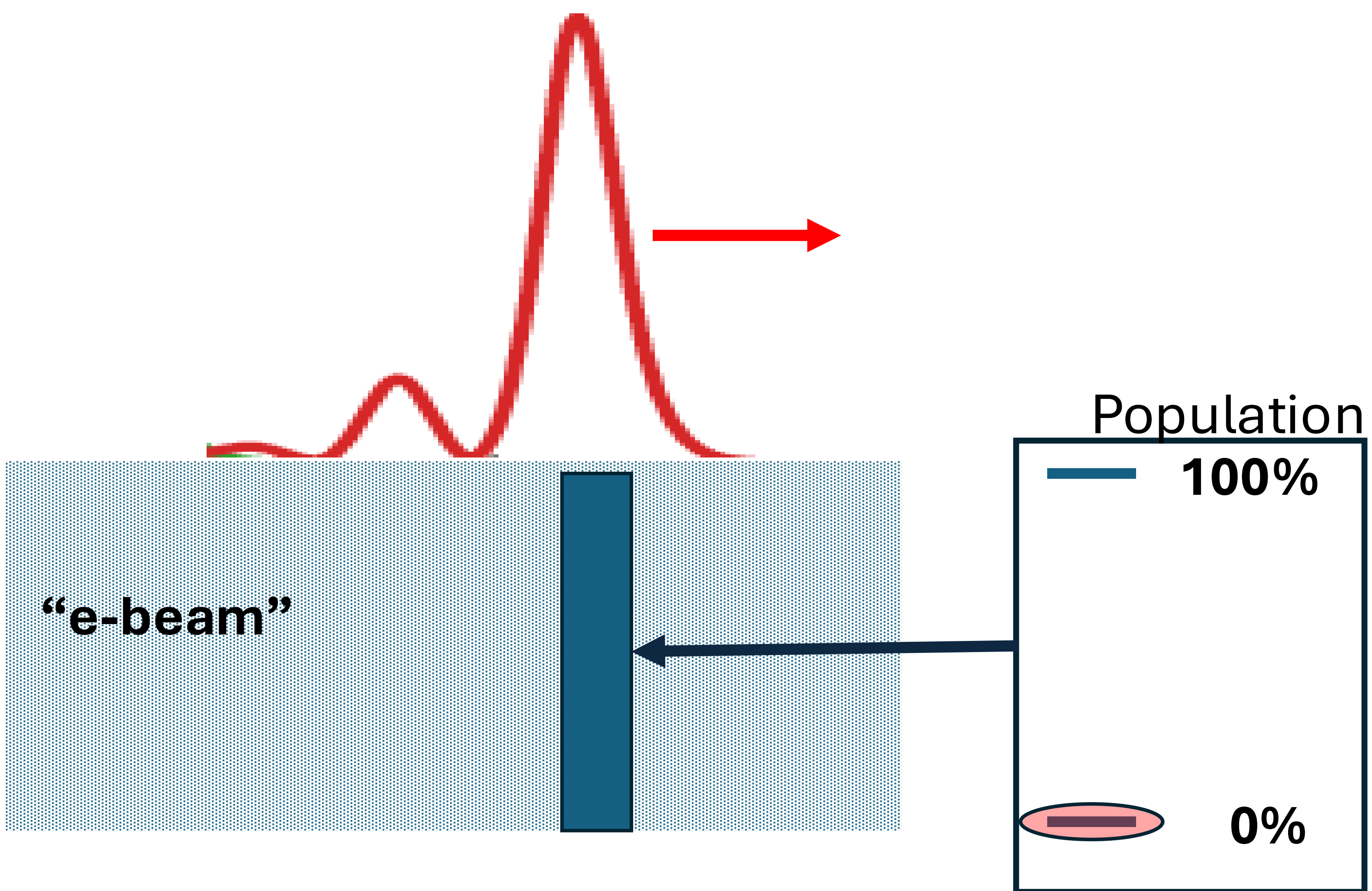

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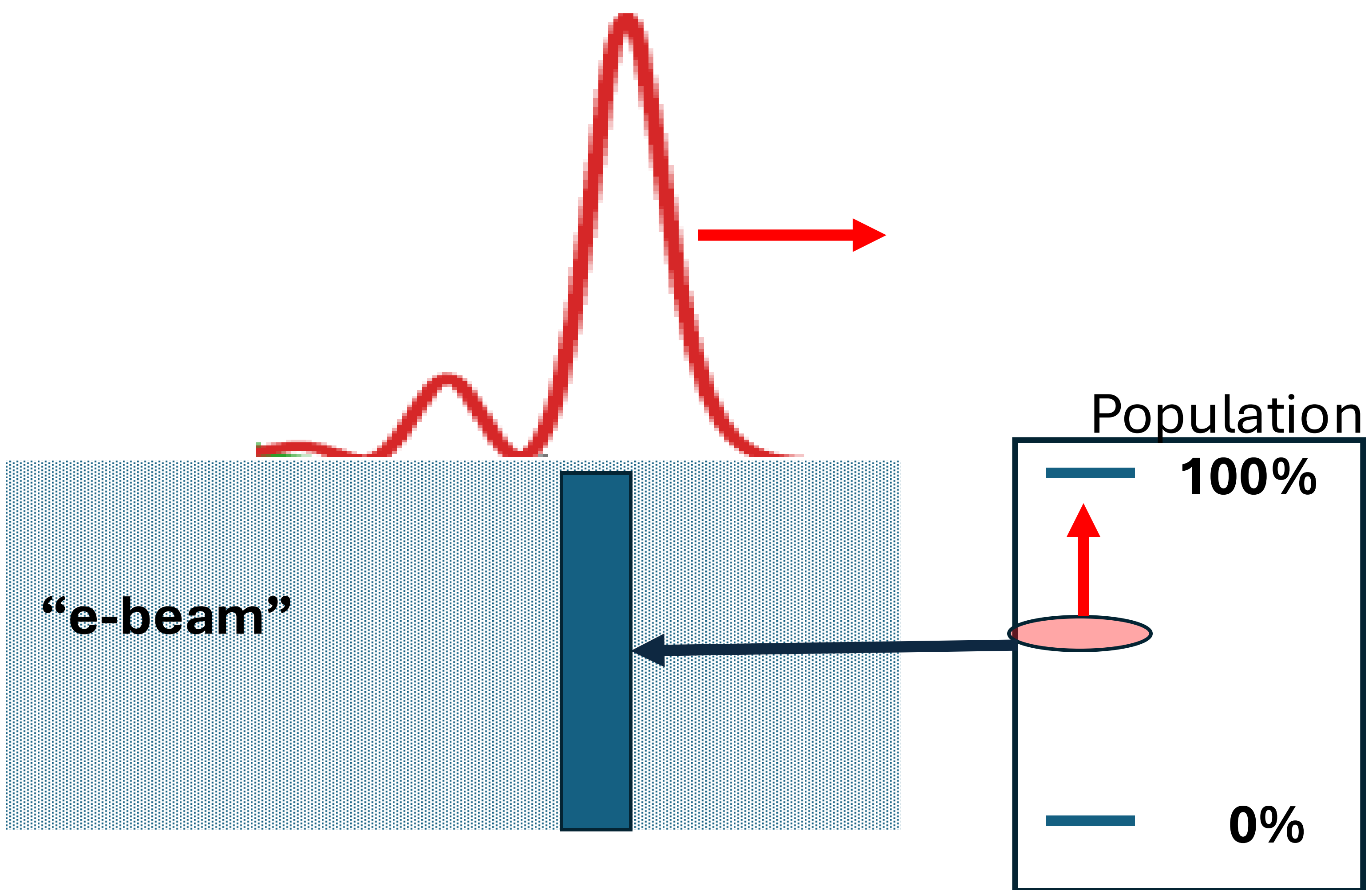

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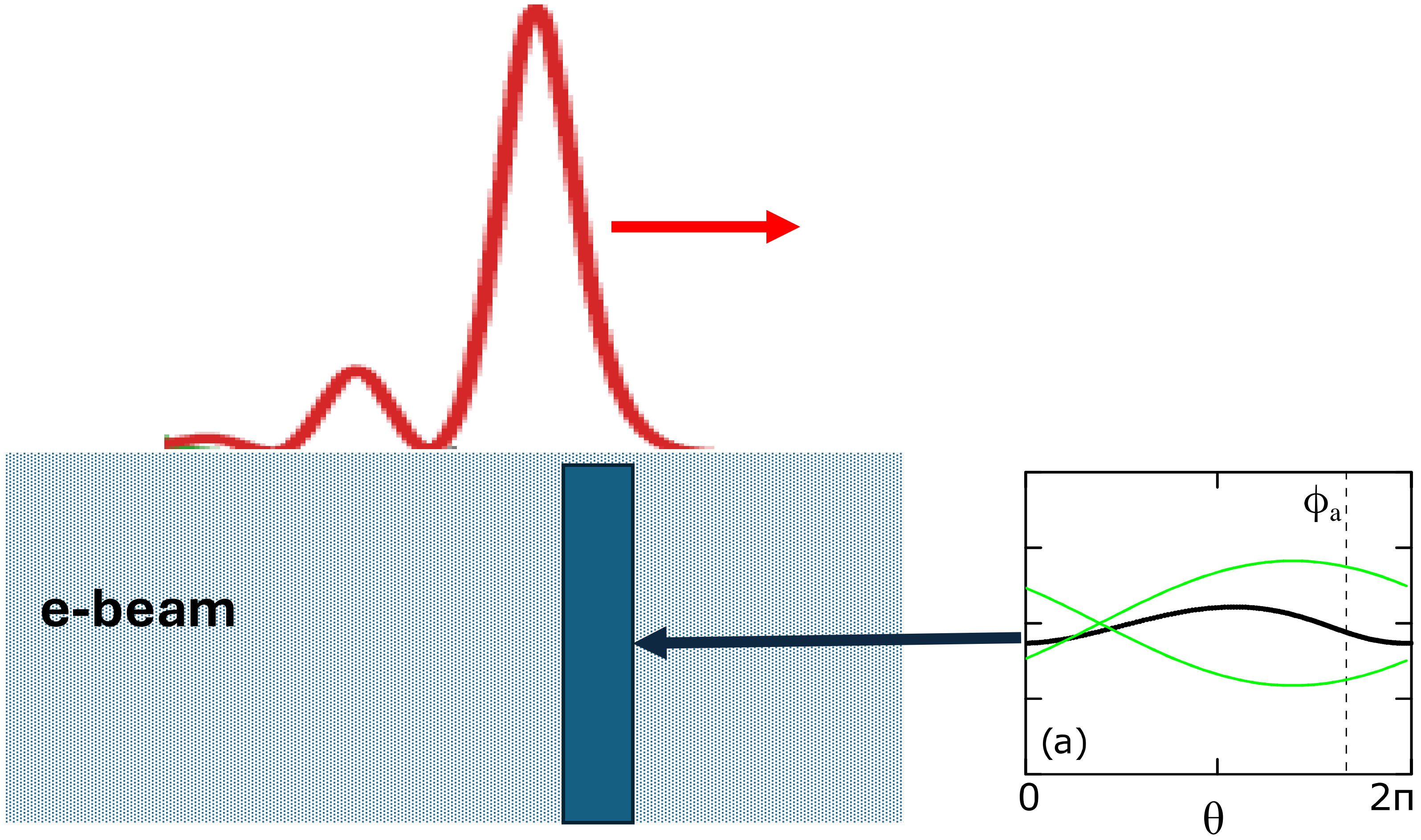

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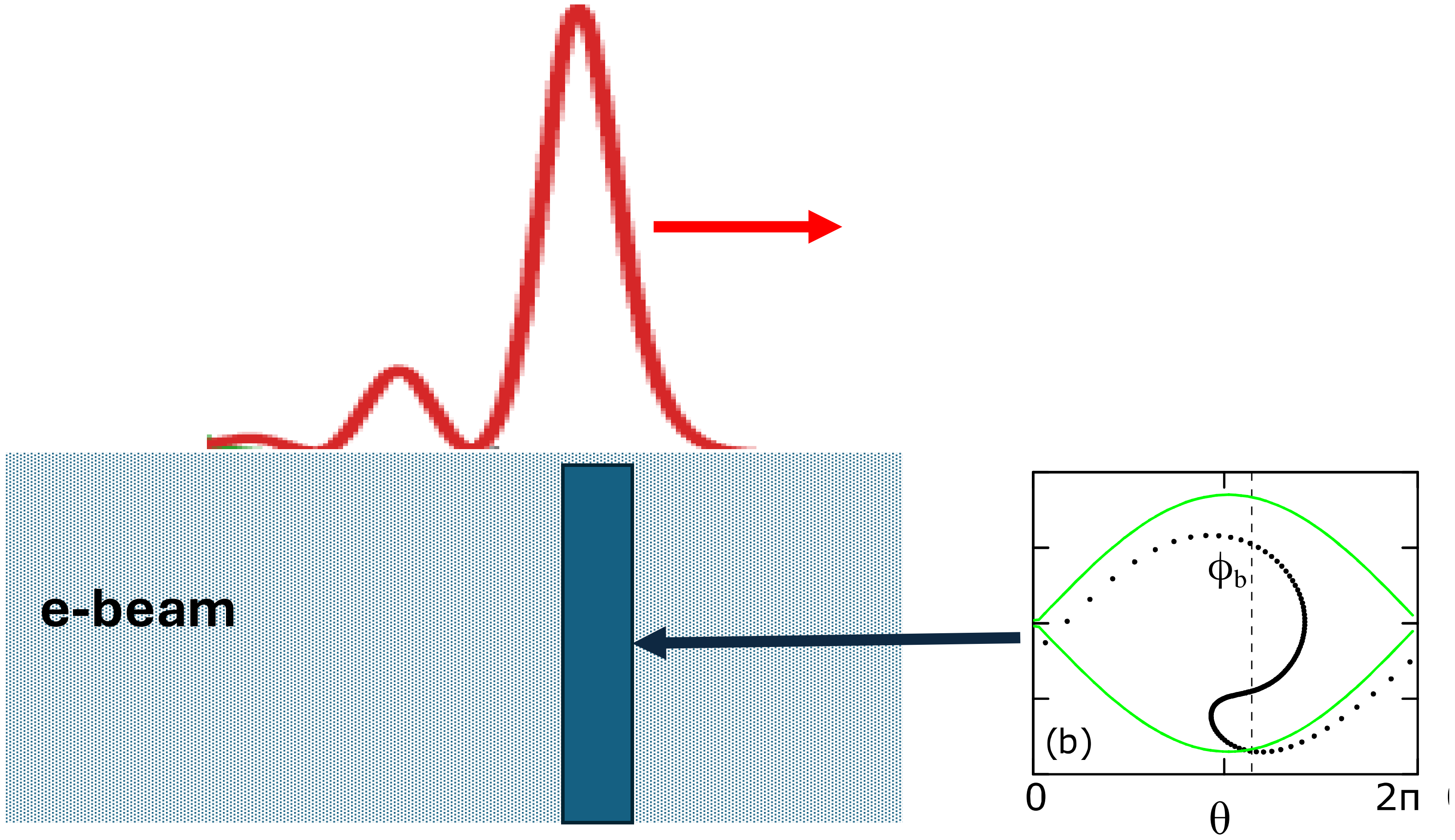

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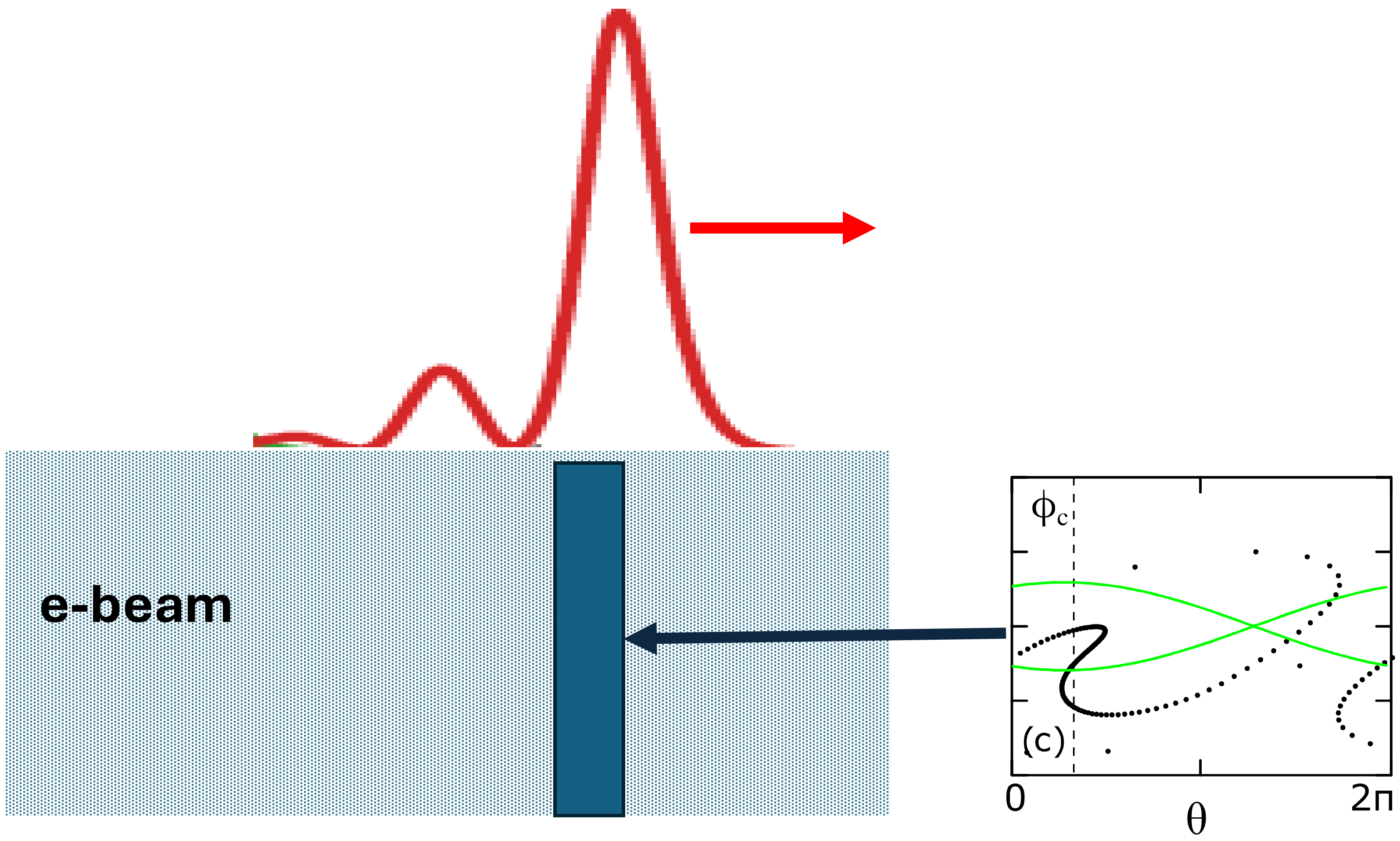

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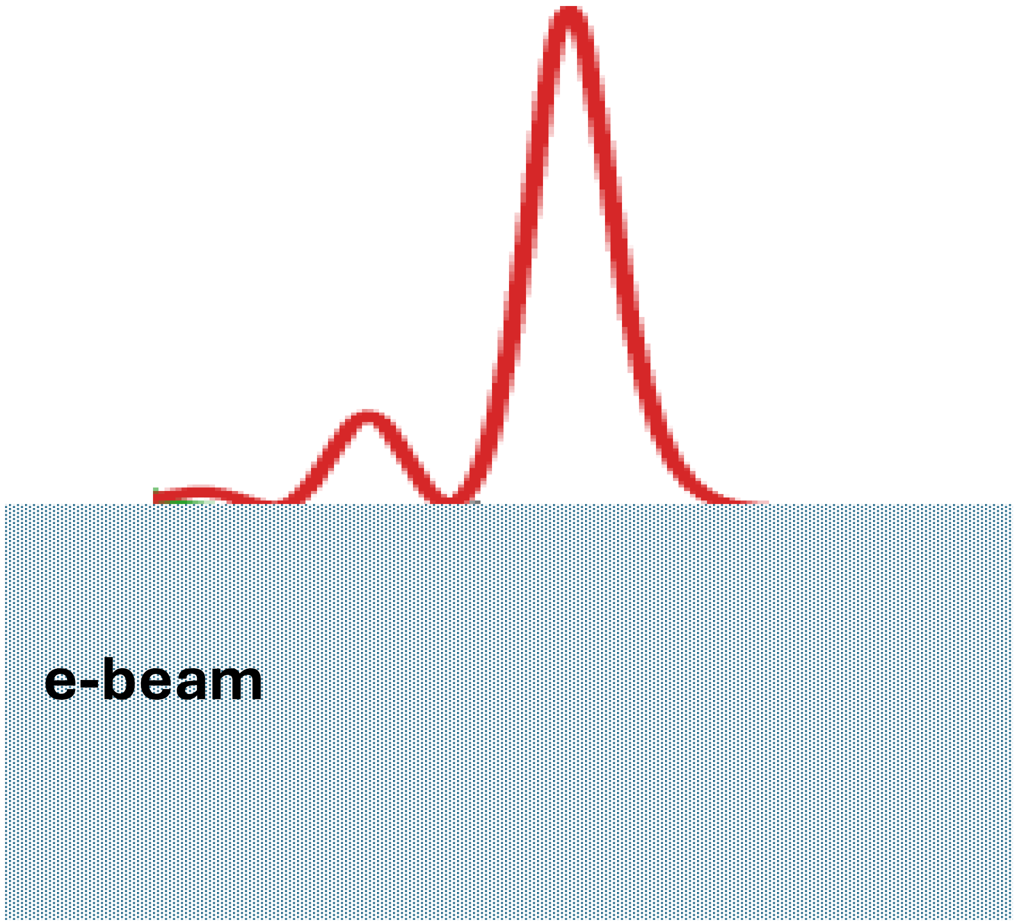

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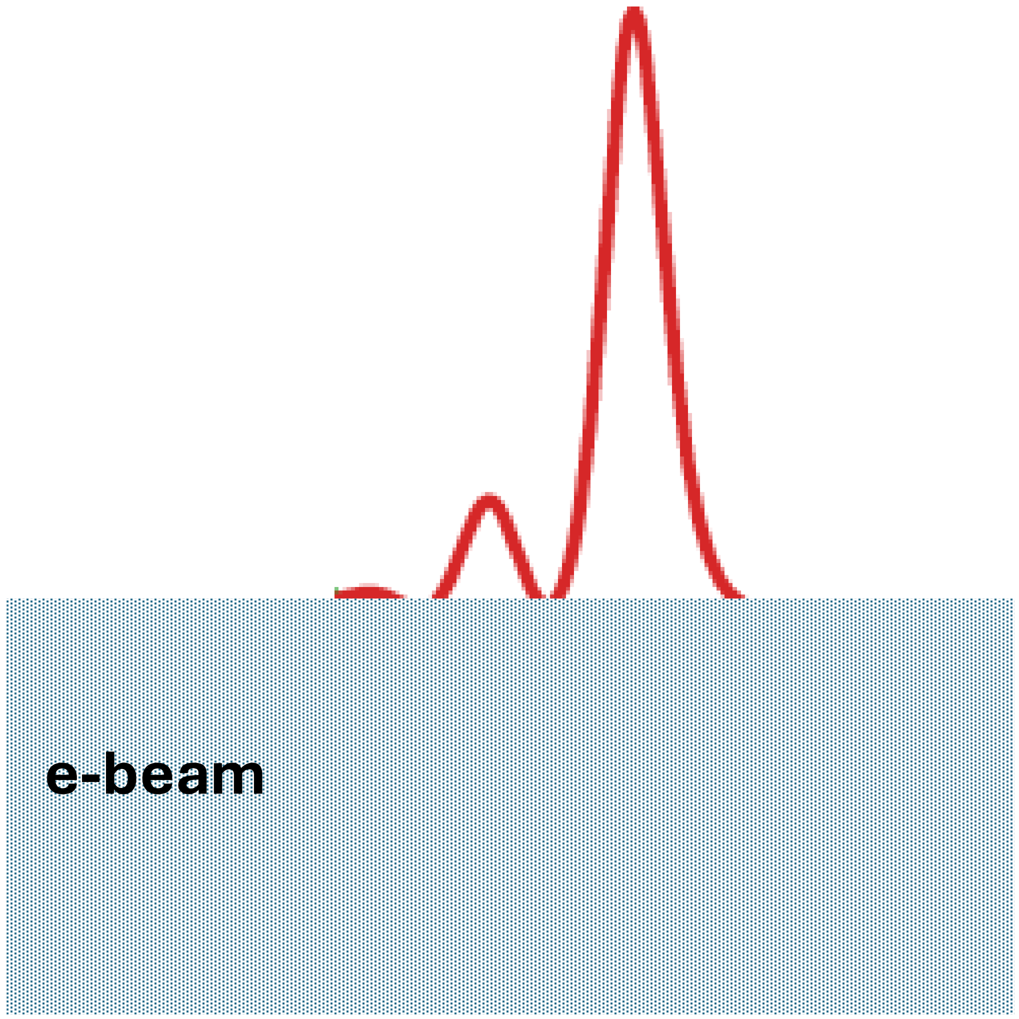

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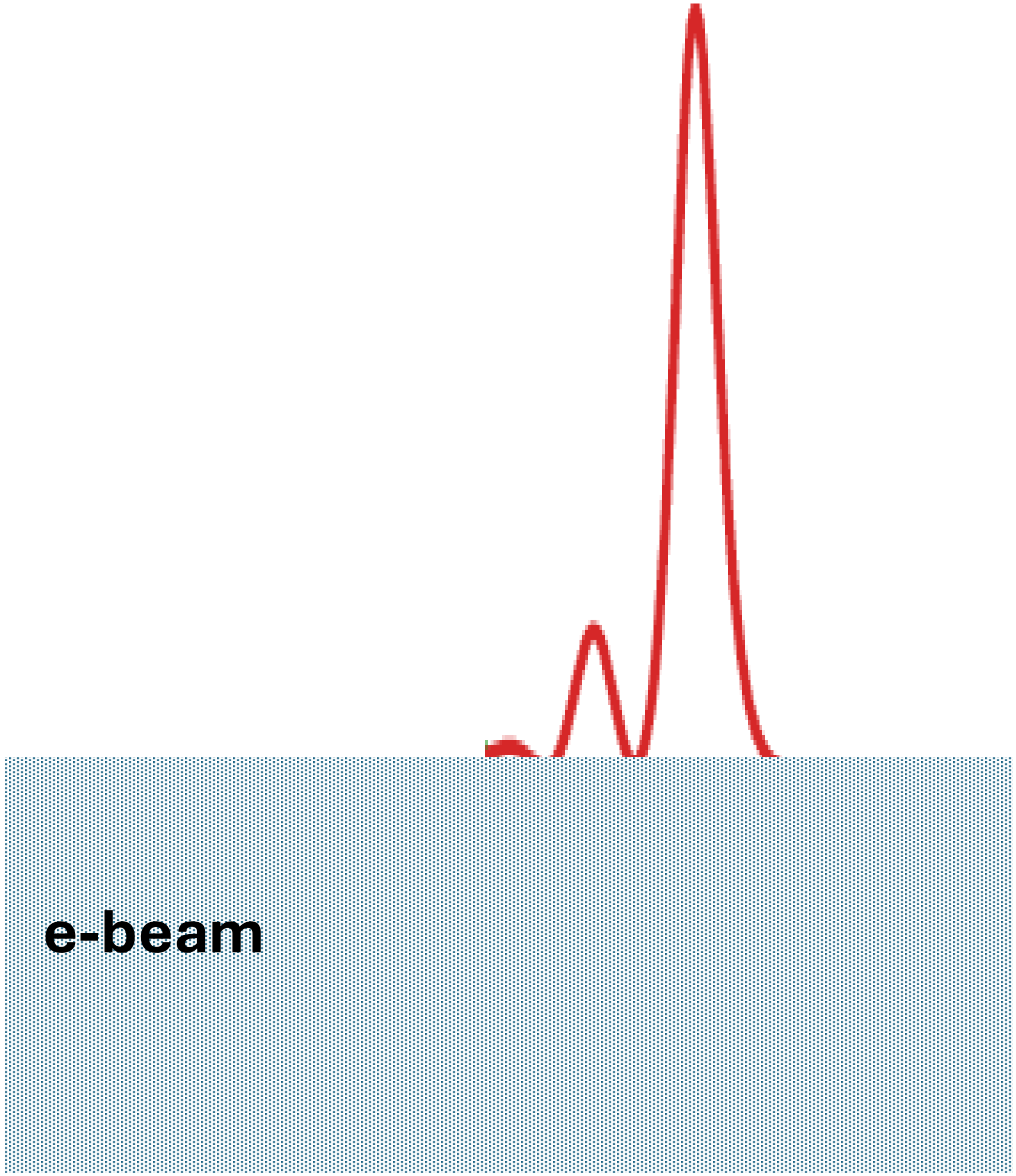
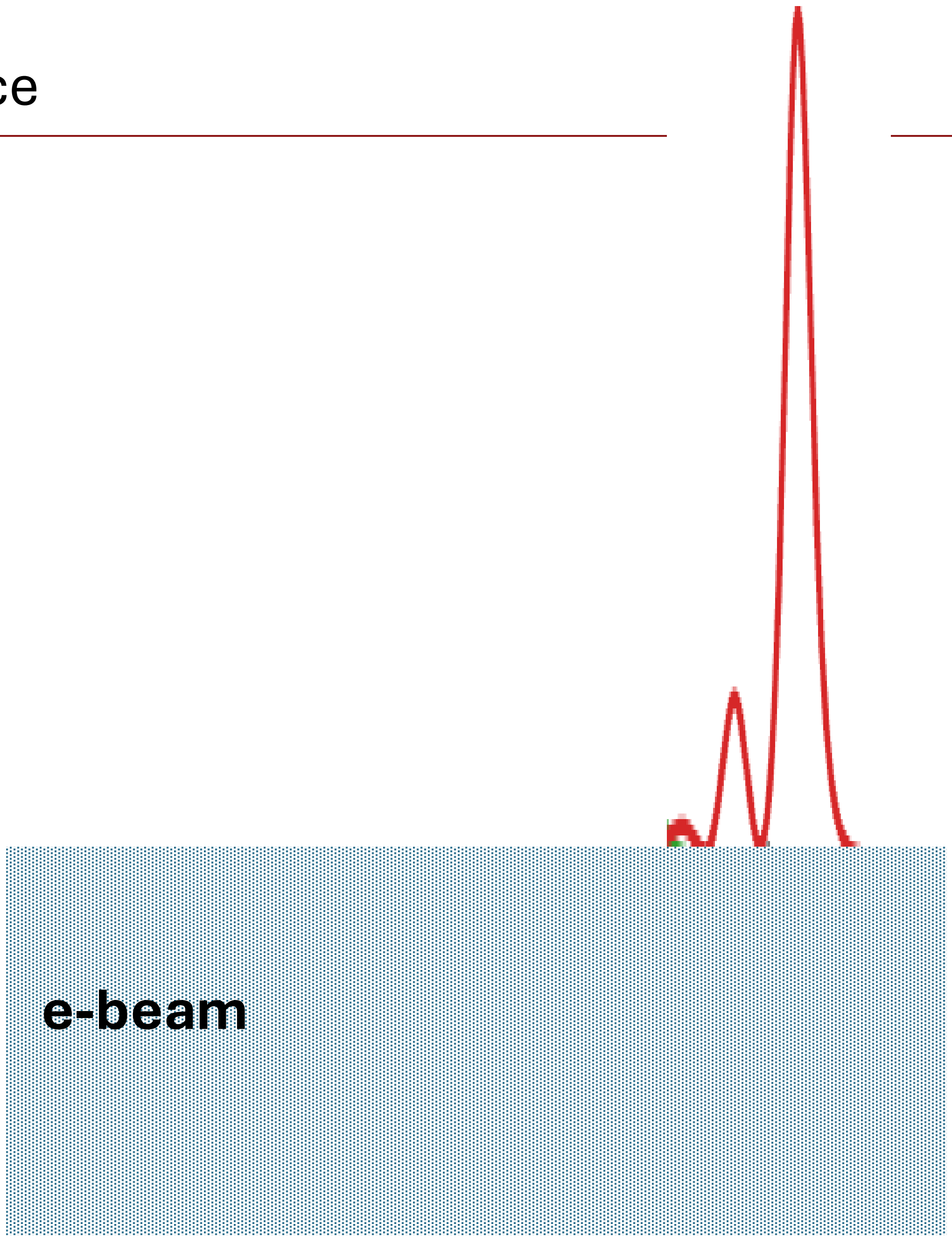
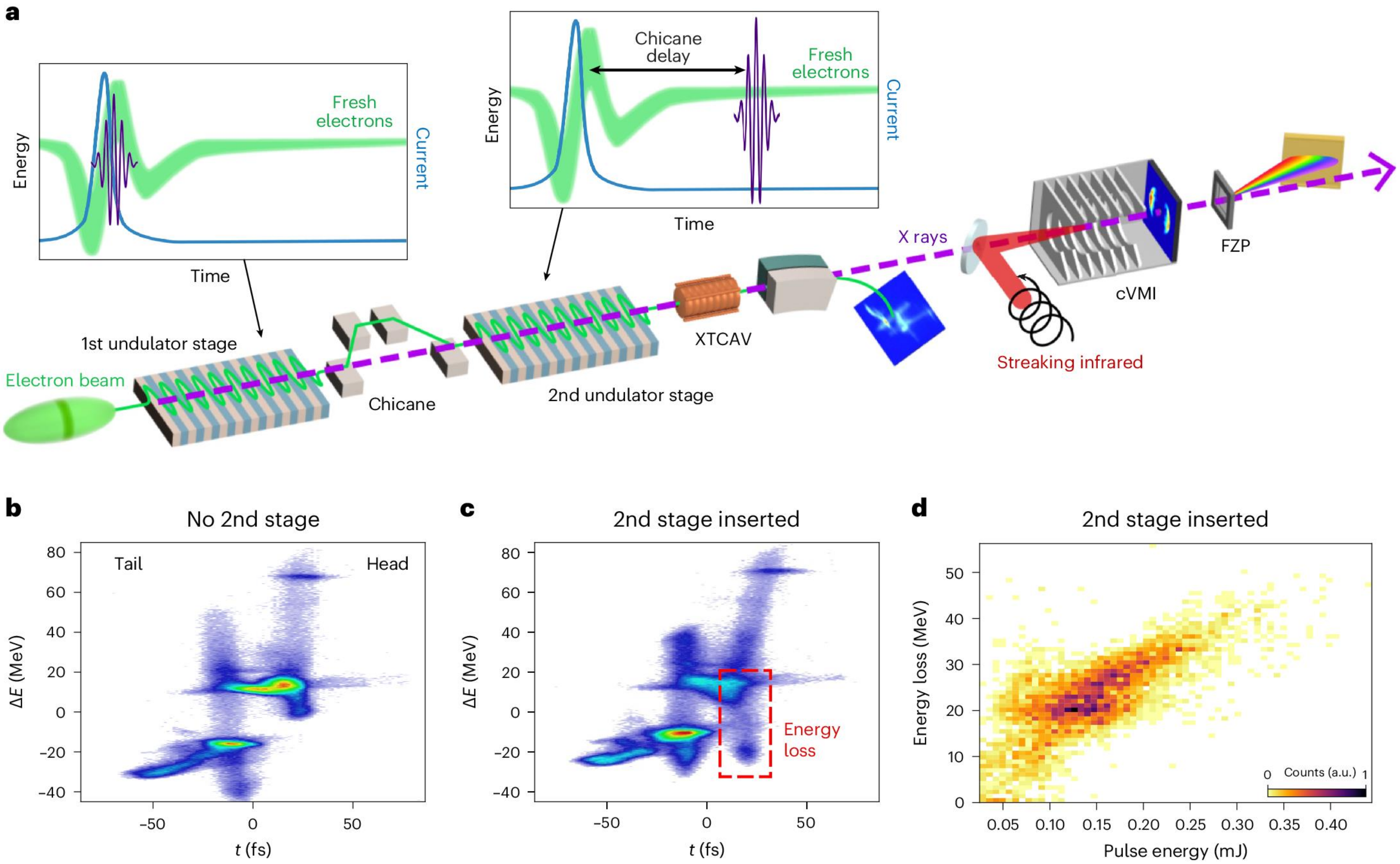


Superradiance



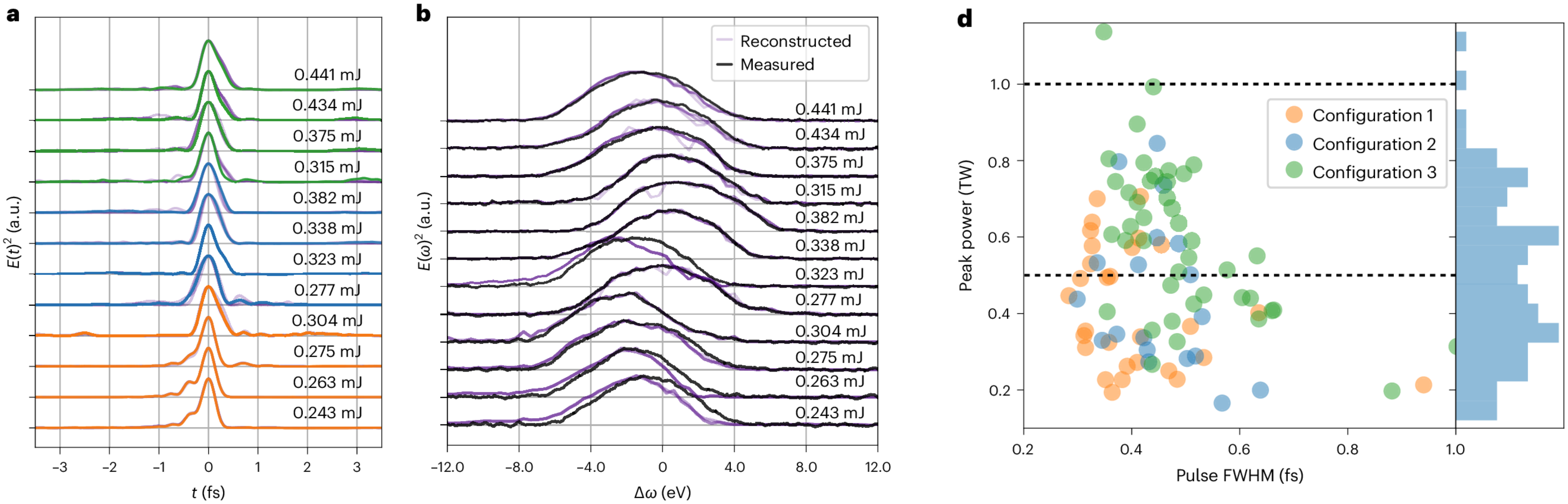
Cascaded Amplification

P. Franz



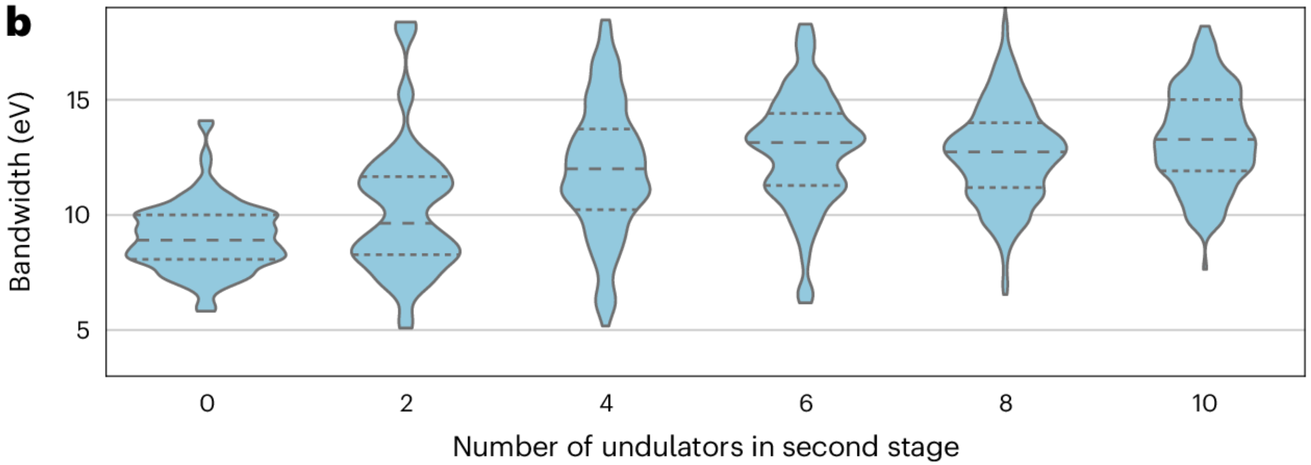
Paris Franz et al. "*Nature Photonics* 18.7 (2024): 698-703.

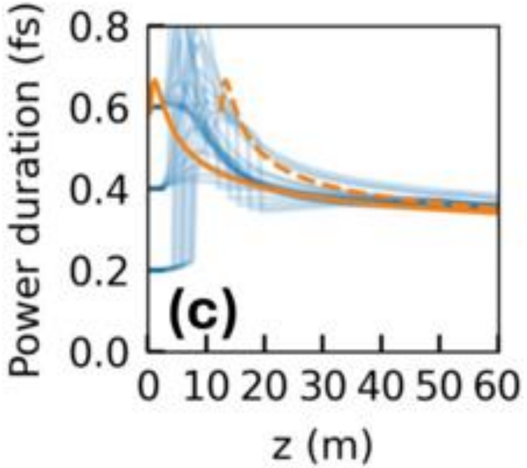
TW-Scale Pulses



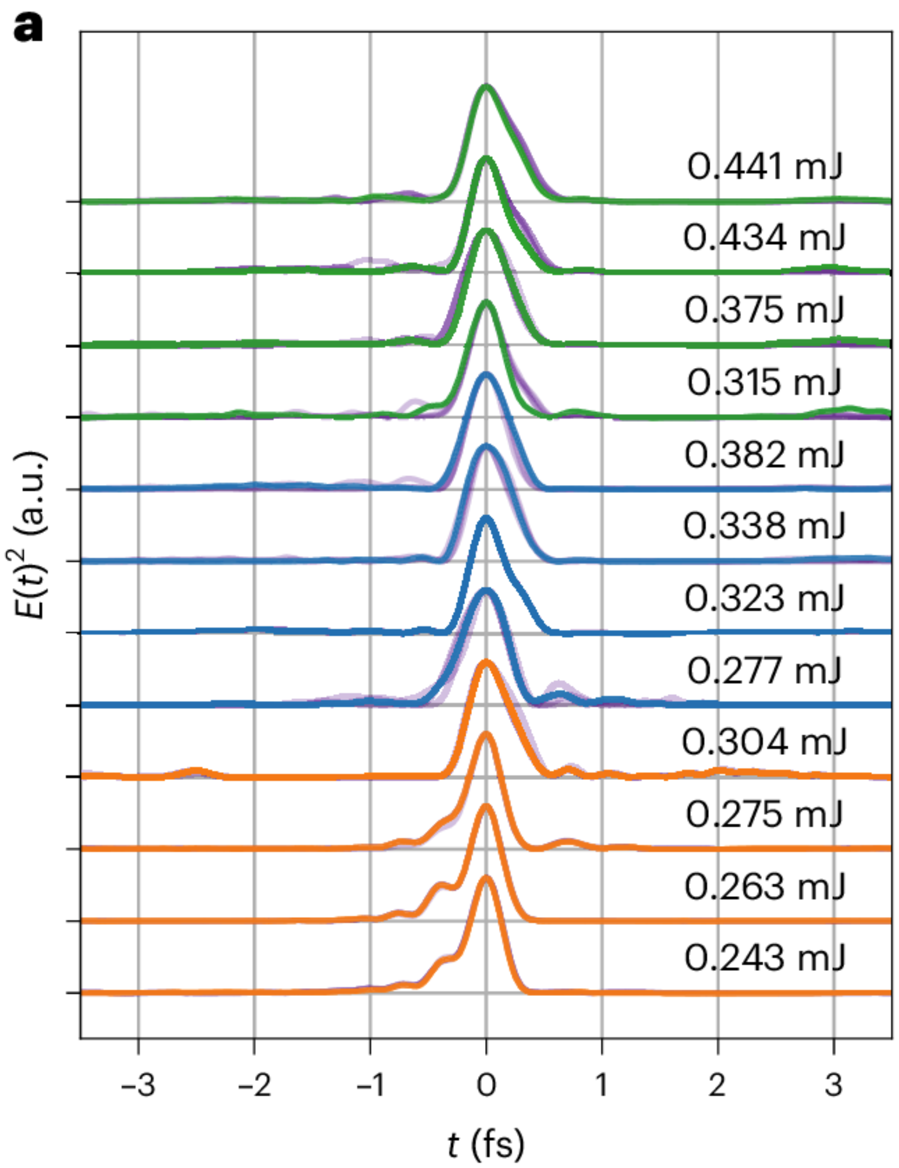
Paris Franz et al. "*Nature Photonics* 18.7 (2024): 698-703.

Is This Superradiance?





R. Robles



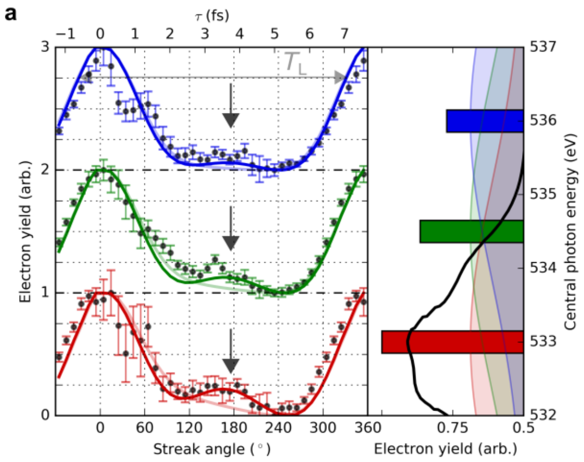
Paris Franz et al. "*Nature Photonics* 18.7 (2024): 698-703.

R. Robles et al. Phys. Rev. Research **6**, 033158 (2024)

See also results from FERMI: Mirian et al. *Nature Photonics* 15.7 (2021): 523-529

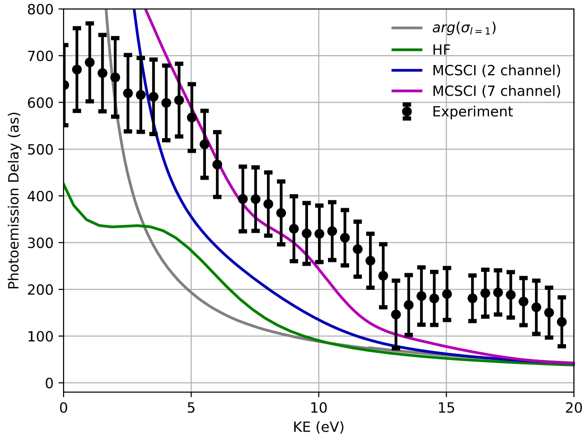
Attosecond Science at LCLS

Electronic Coherence in Auger-Meitner Decay



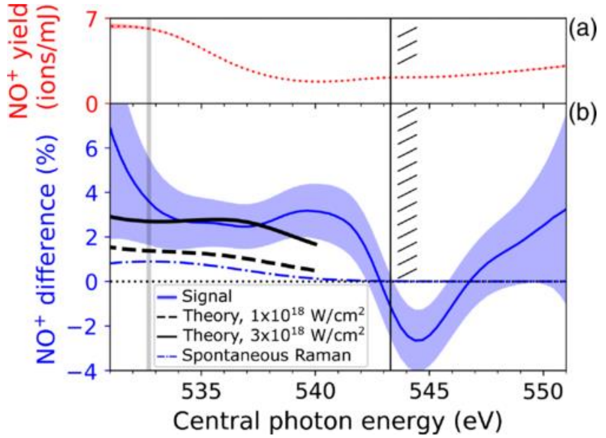
S. Li, T. Driver et al. *Science* 375.6578 (2022): 285-290.

Photoemission delays



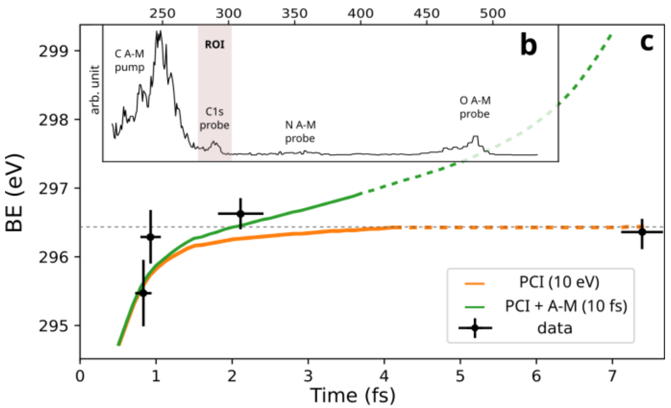
T. Driver et al. Nature 632 (8026), 762-767

Non-linear methods



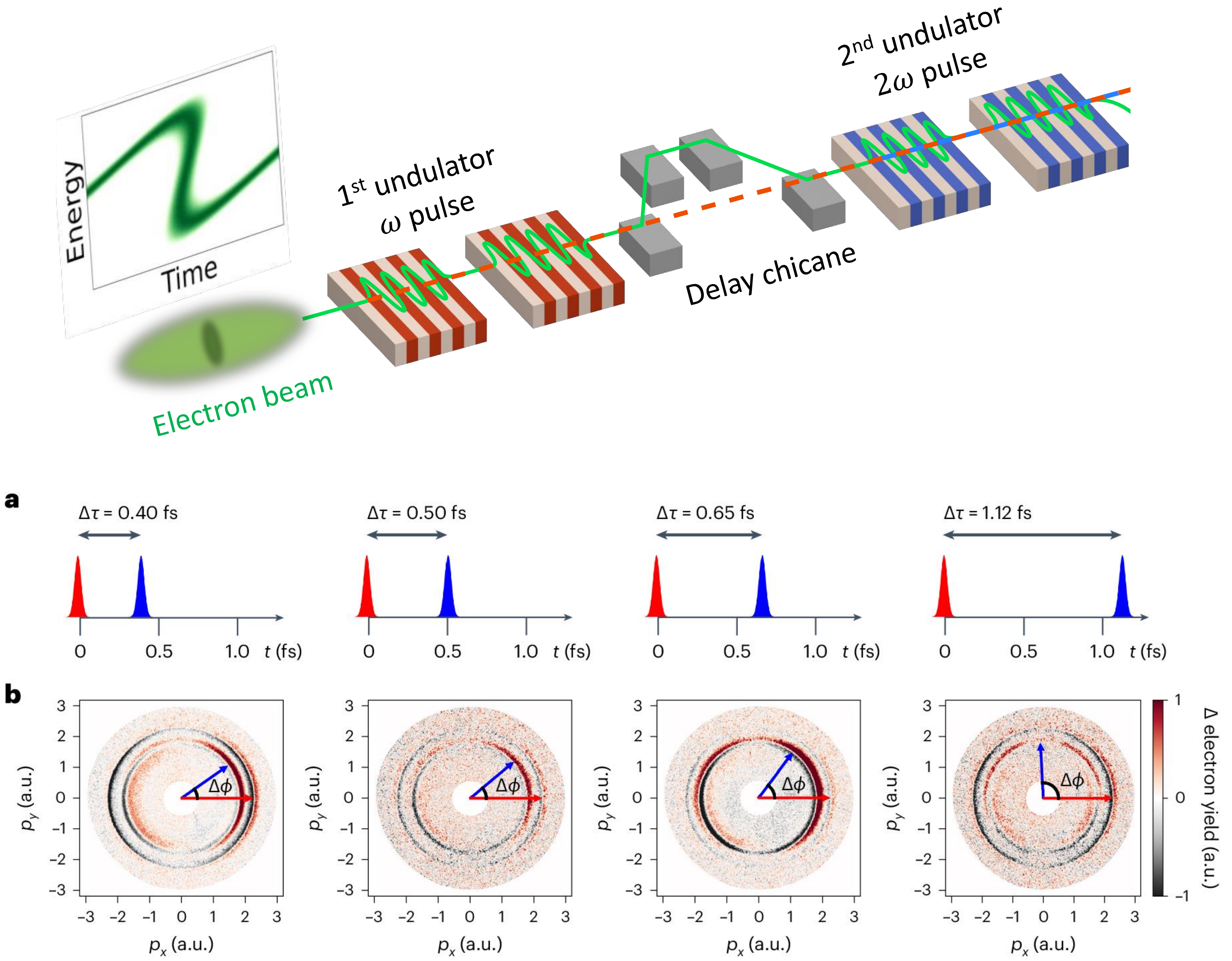
J. O'Neal et al. *Phys. Rev. Lett.* 125.7 (2020): 073203

Pump/Probe Experiments

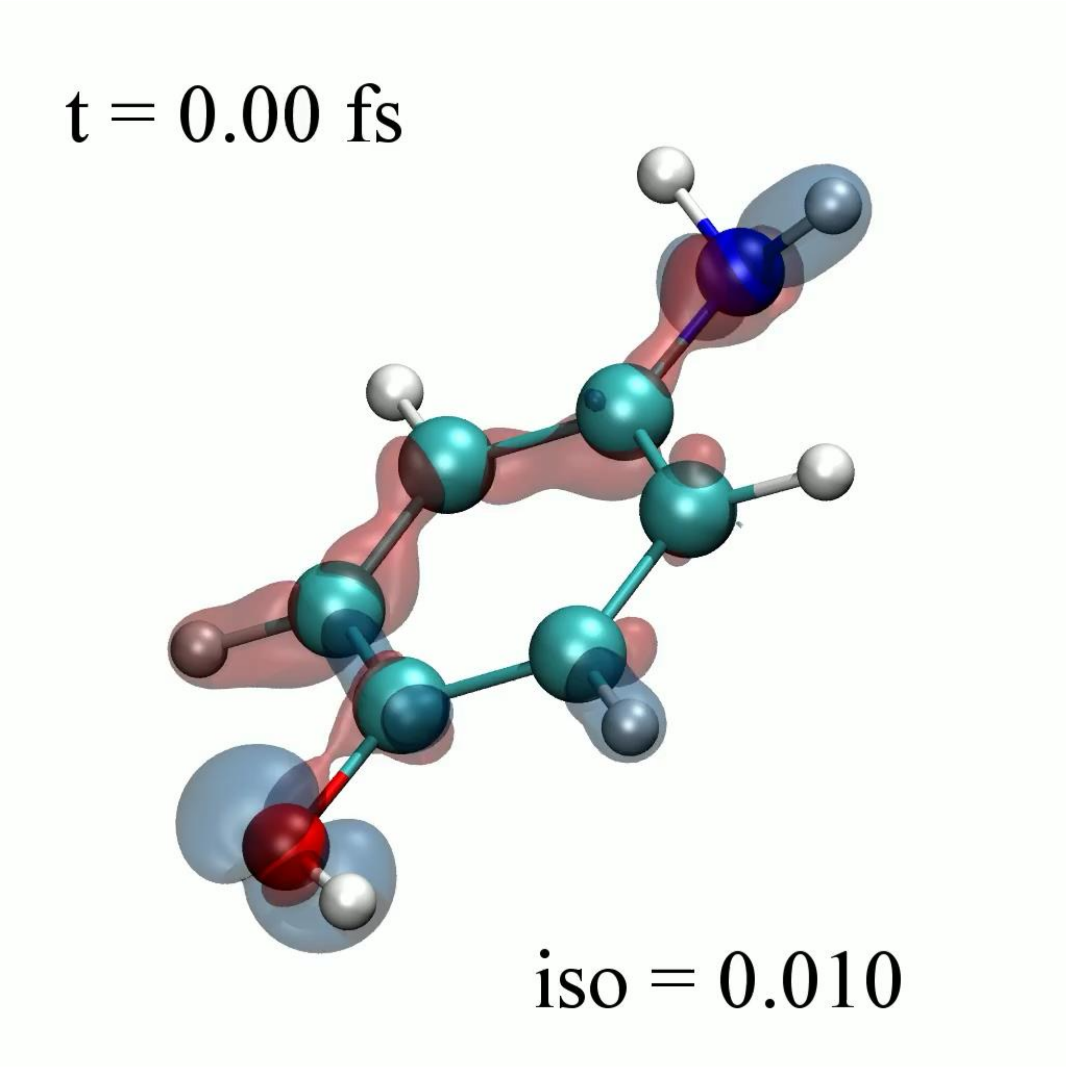


Z. Guo et al. *Nature Photonics* (2024): 1-7.

Attosecond Pump/Probe Setup



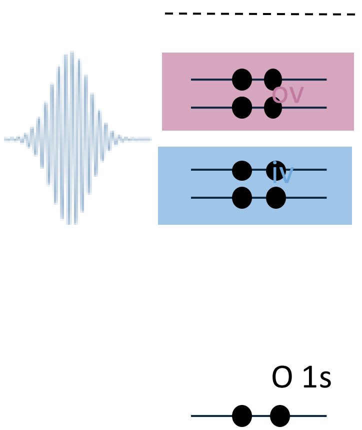
Pump/Probe Demonstration

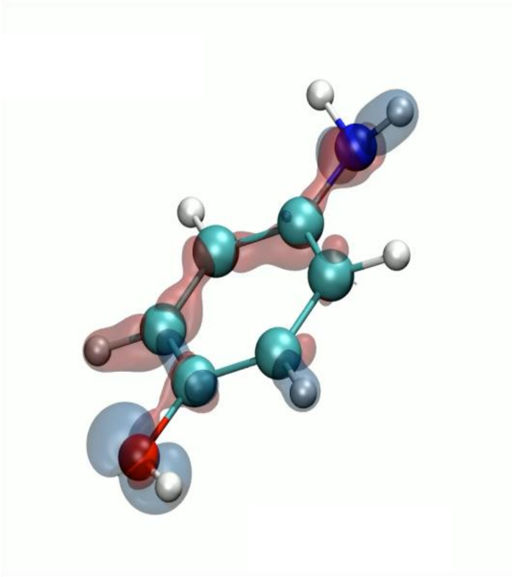


Simulation: G. Grell, F. Martin et al.

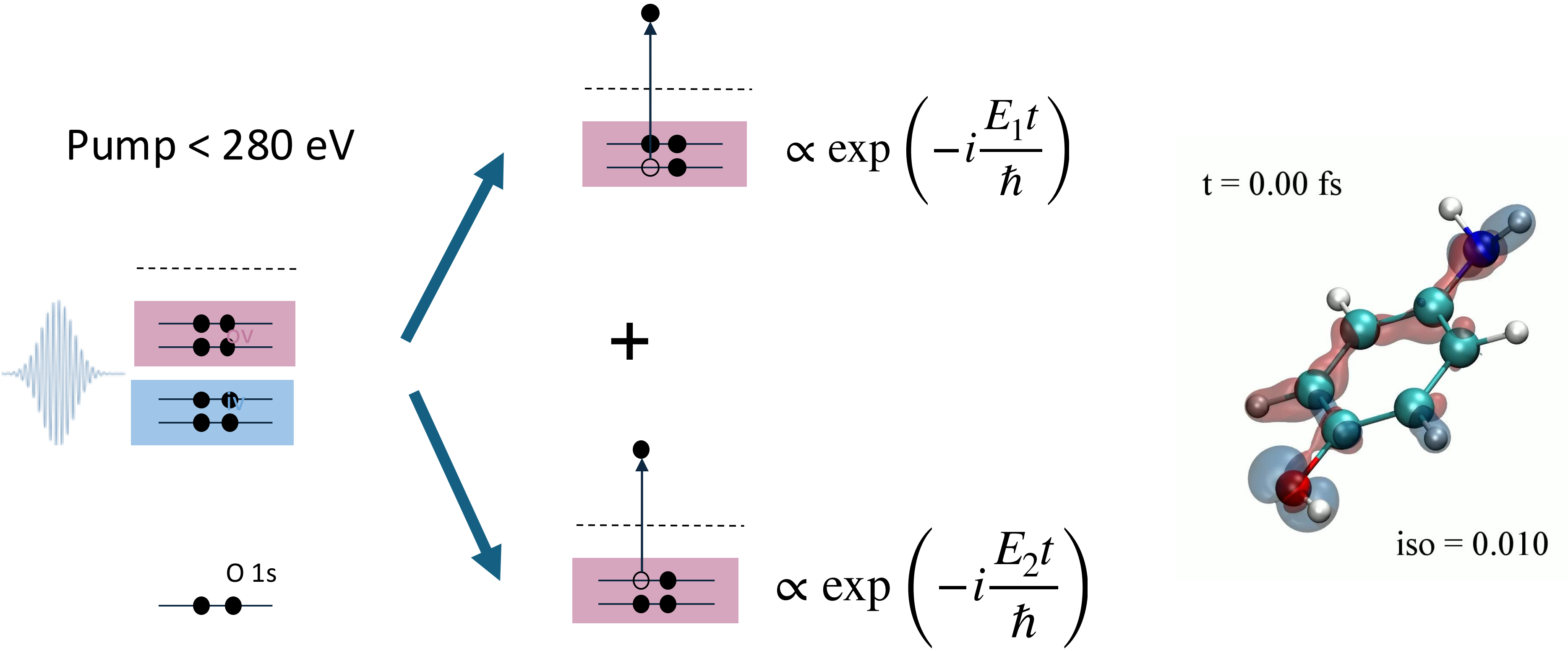
Pump-Probe Scheme

Pump < 280 eV

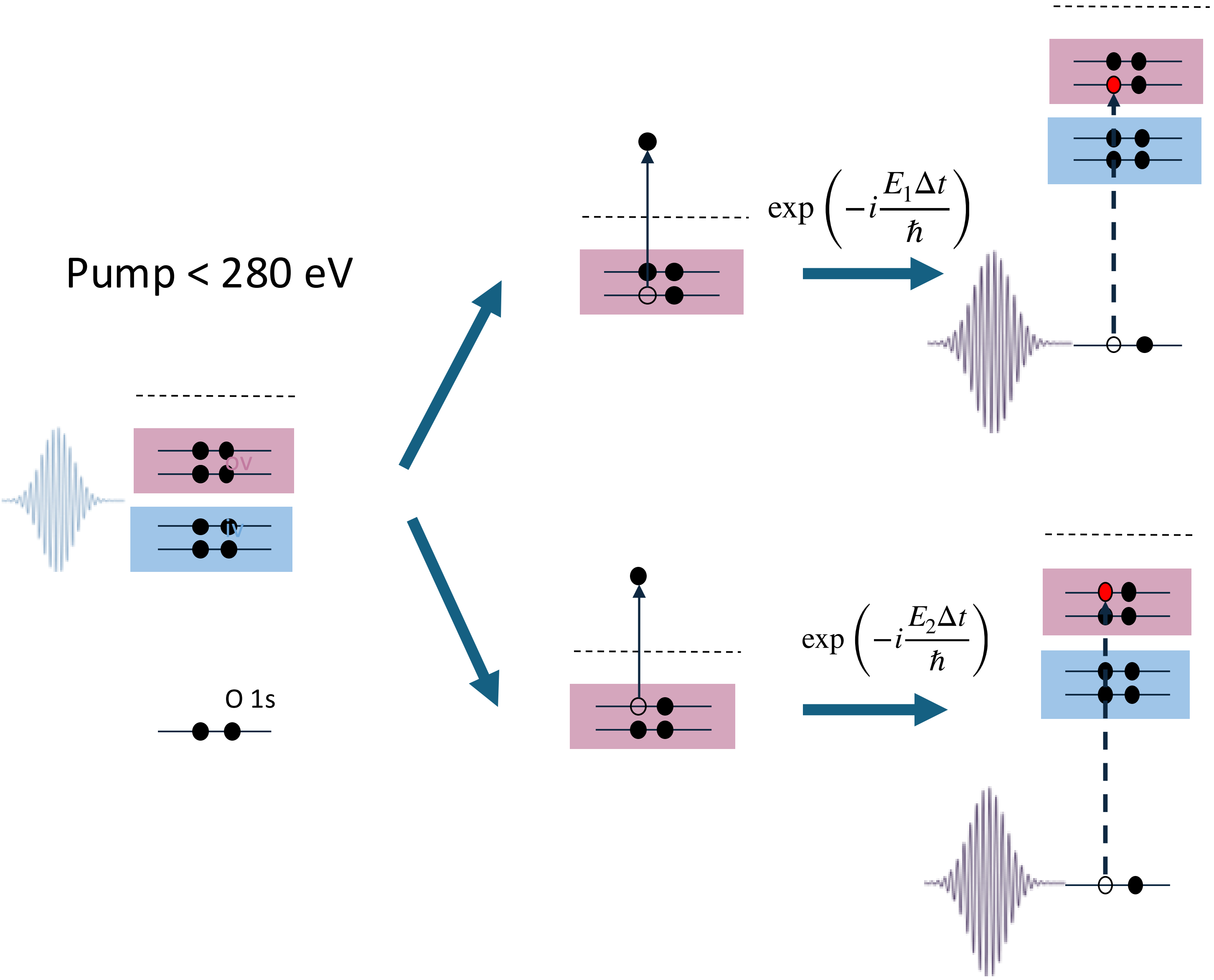




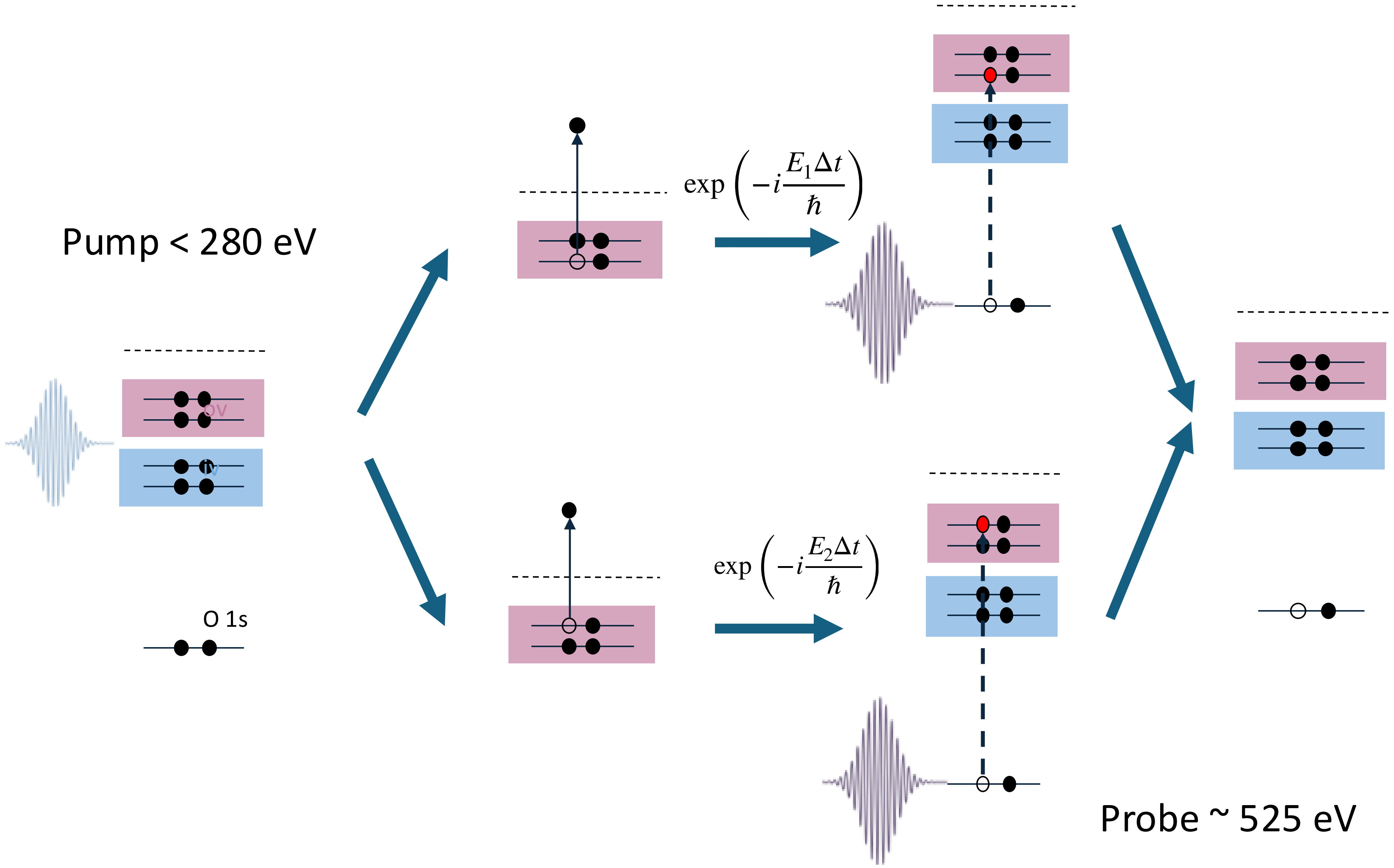
Pump-Probe Scheme



Pump-Probe Scheme

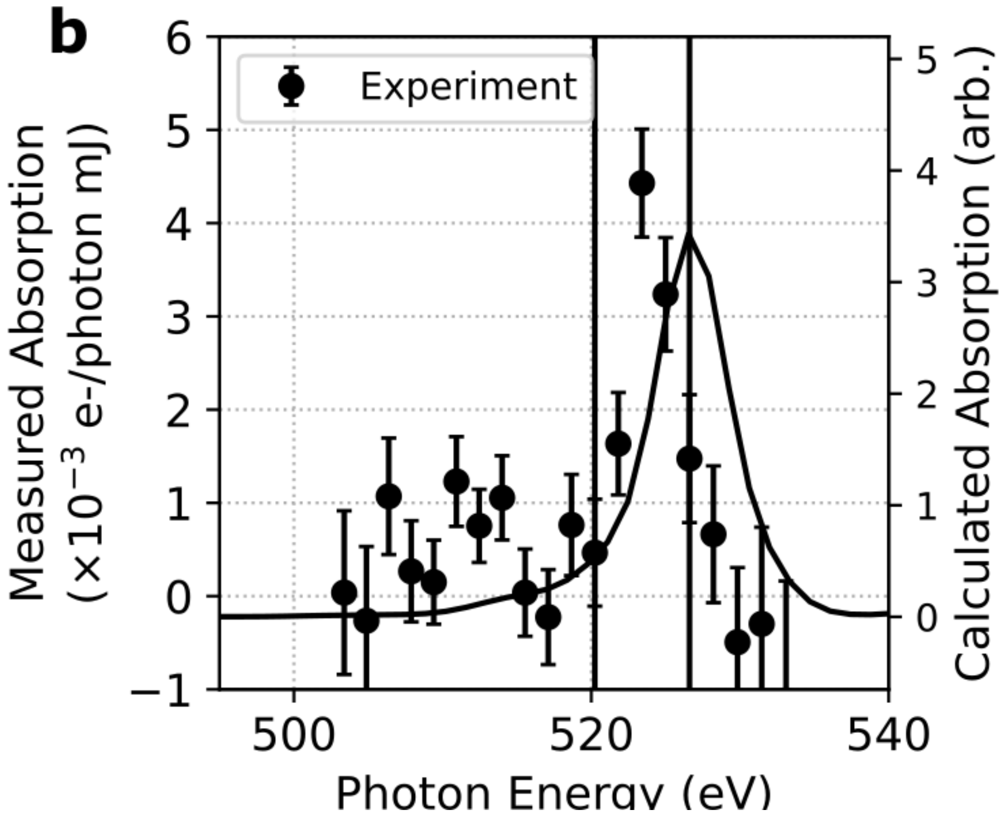


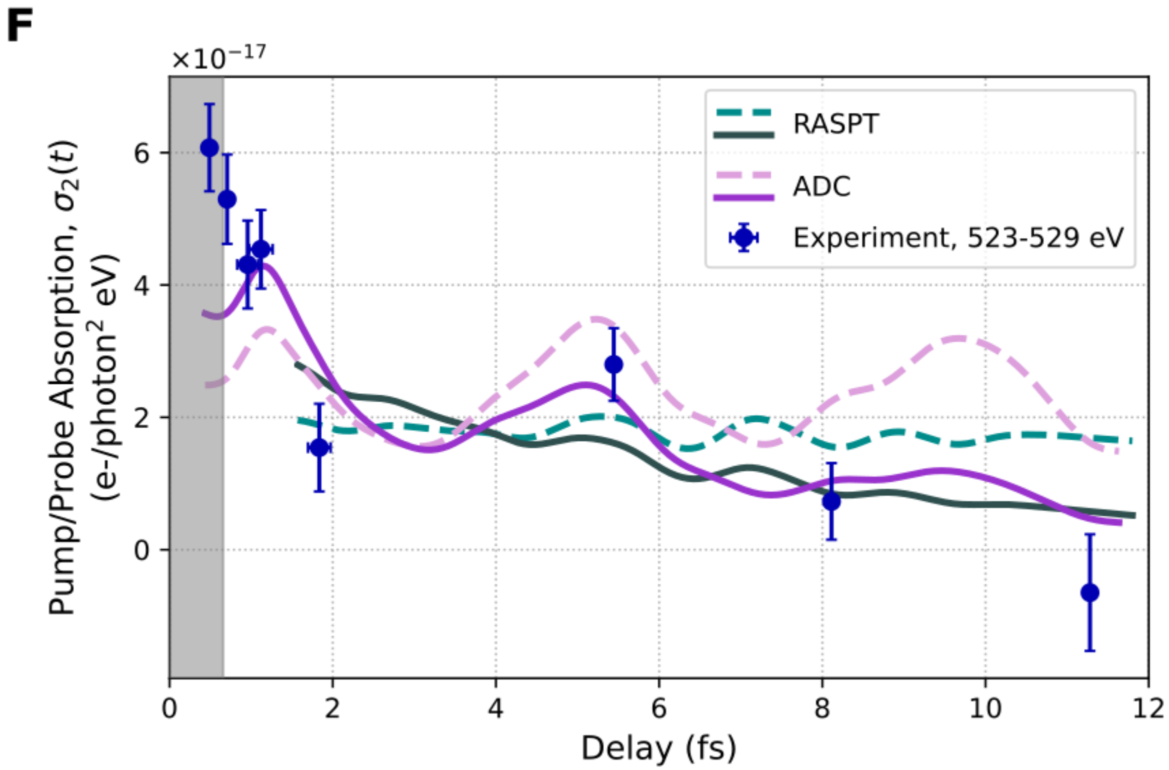
Pump-Probe Scheme



Pump/Probe Demonstration

T. Driver et al. arXiv:2411.01700

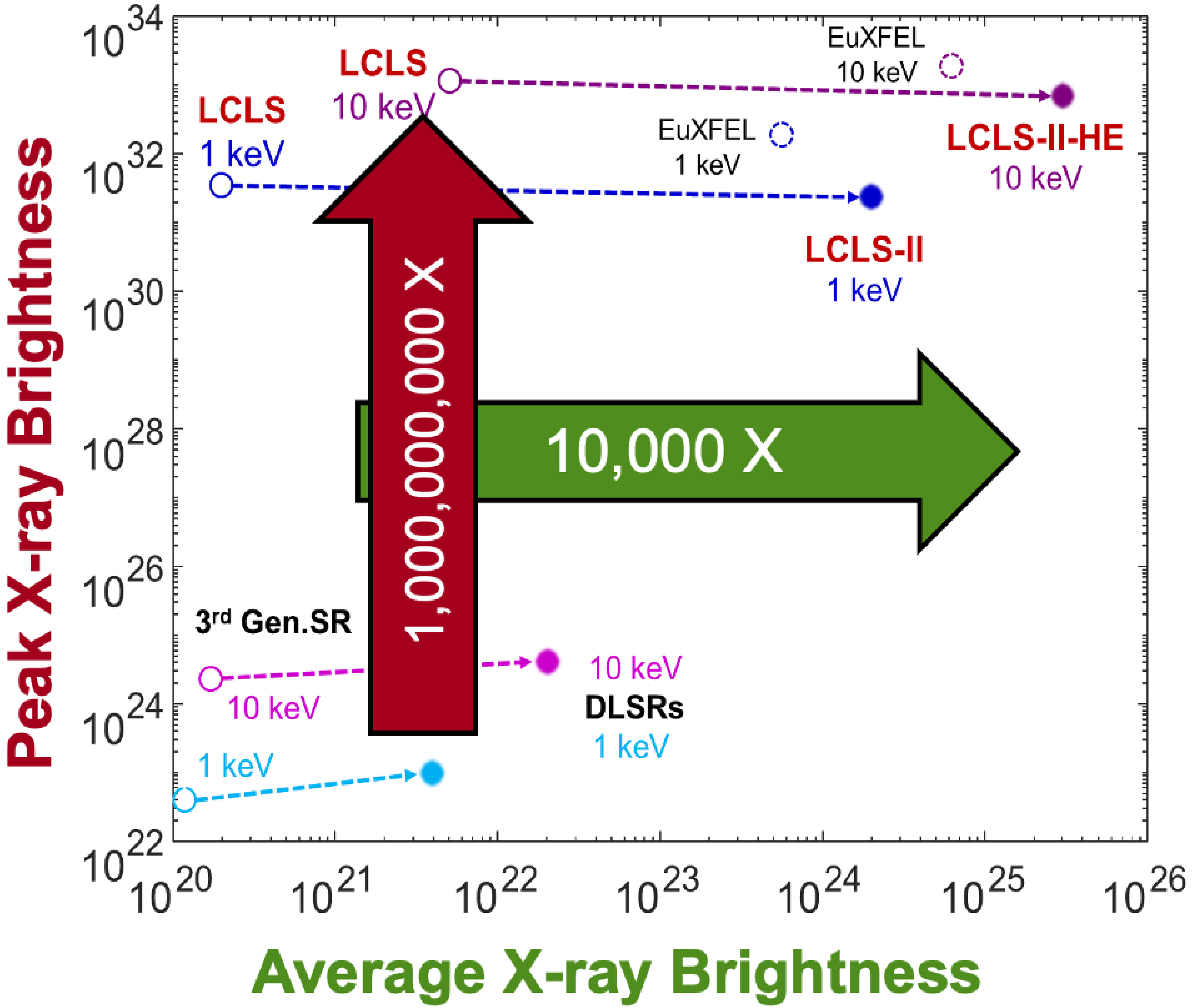




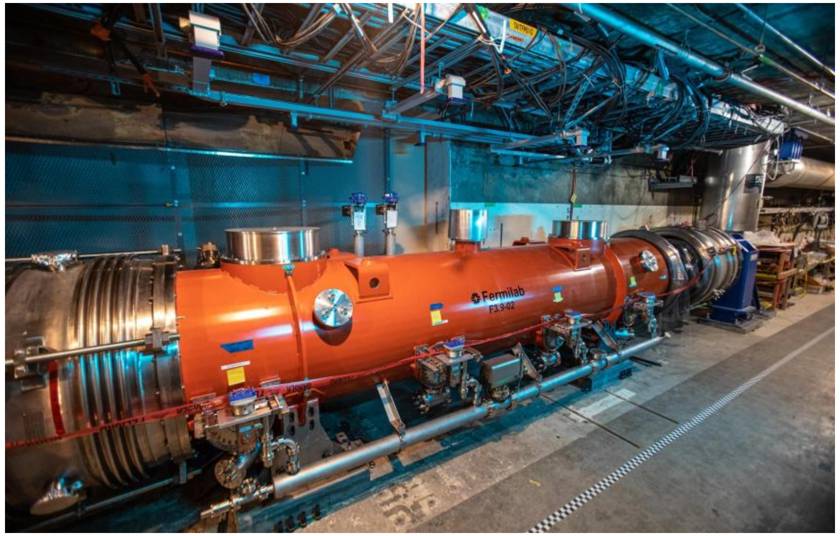
Analysis:
T. Driver, Z. Guo E. Isele
(SLAC/Stanford)
D. Tuthill (OSU)

Simulations:
M. Ruberti and
V. Averbukh (Imperial College)

Attosecond Science in the LCLS-II Era



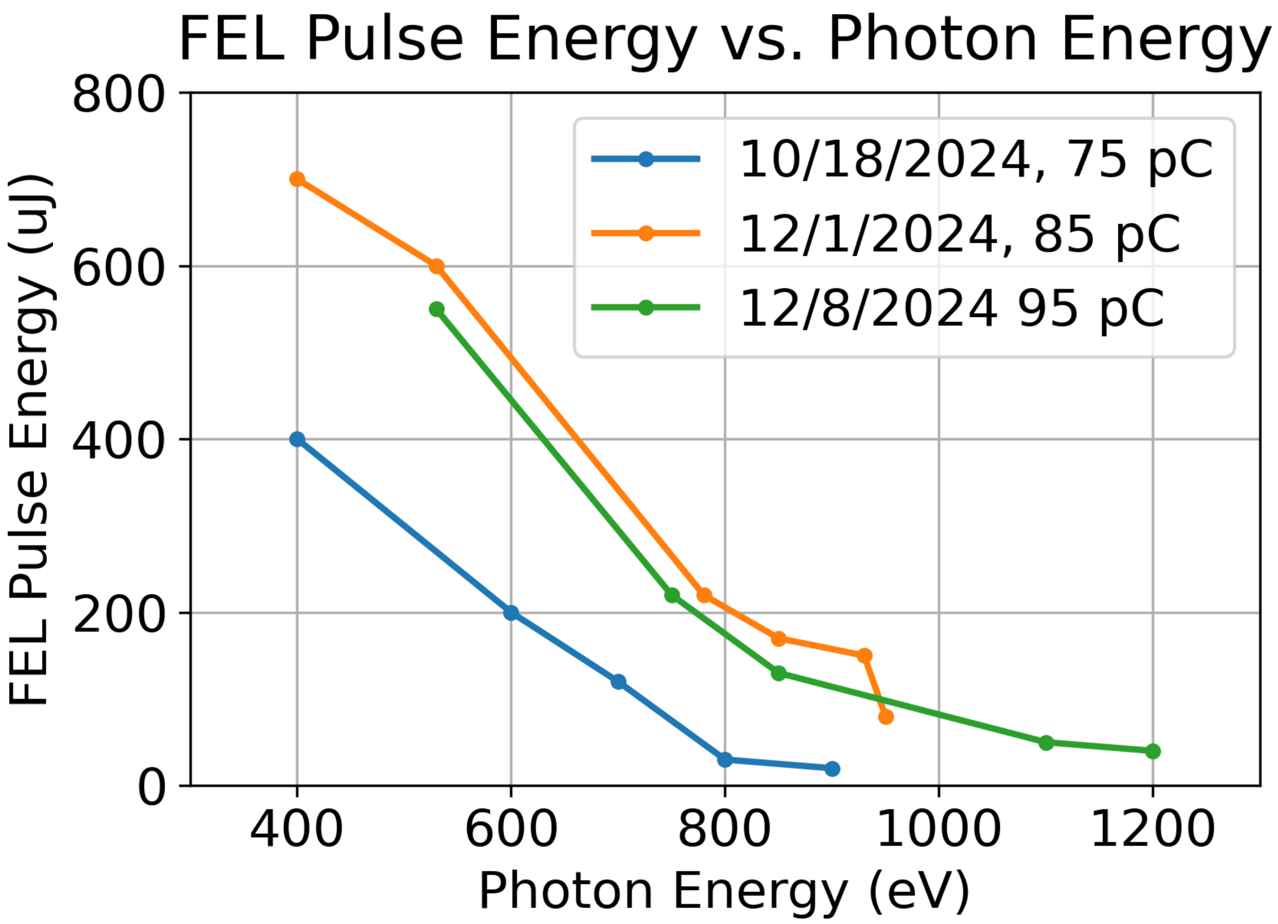
LCLS-II

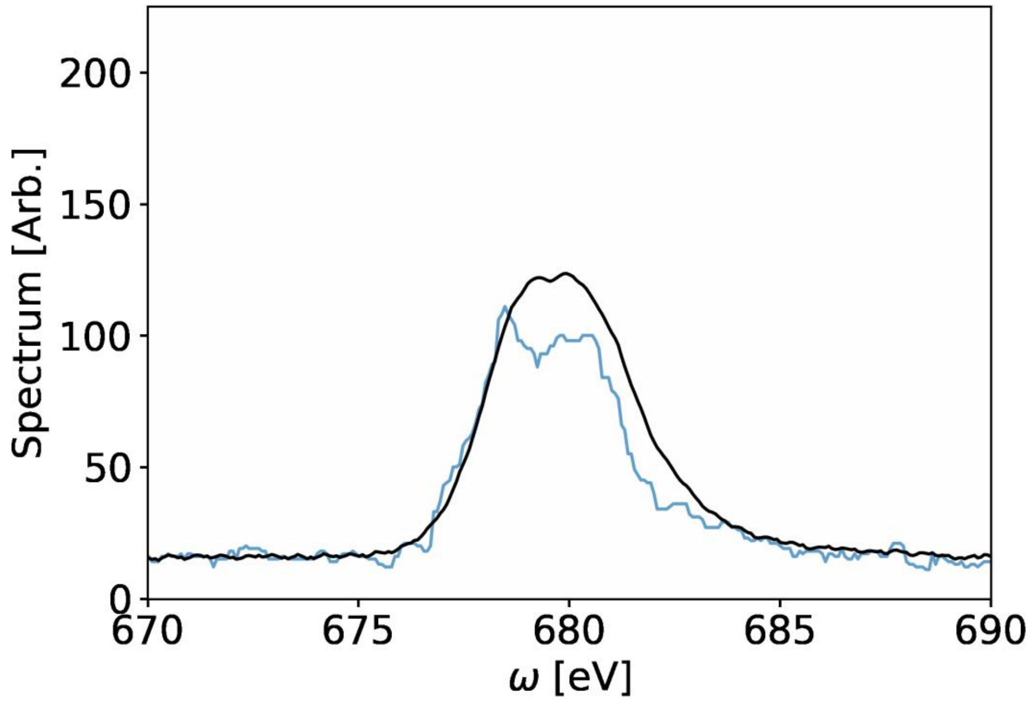


- Superconducting RF accelerator in CW mode
- Up to 1 MHz repetition rate

Currently operating in soft x-ray region.
Ongoing HE upgrade will extend to hard x-rays (2027)

LCLS-II

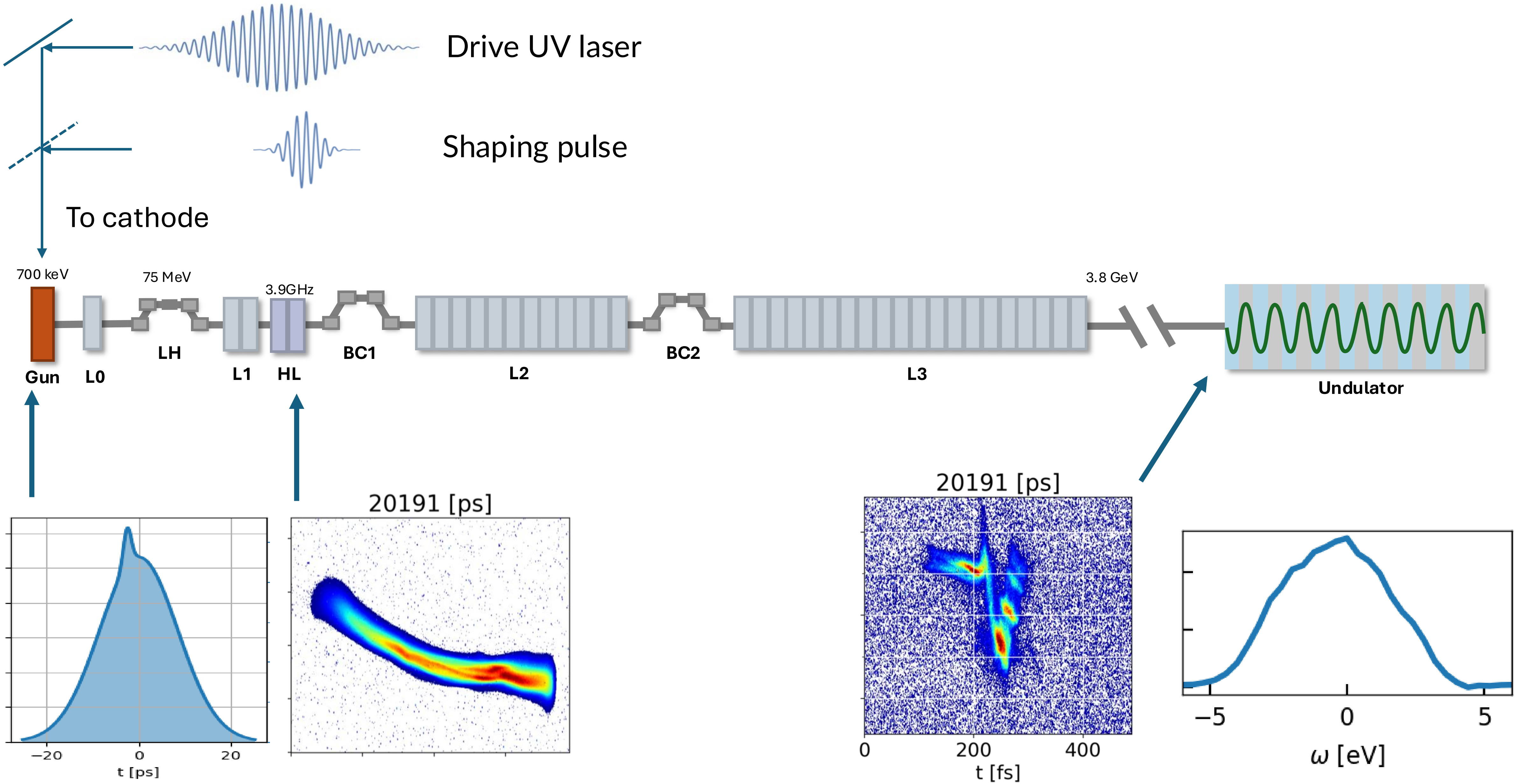




Demonstrated up 93 kHz

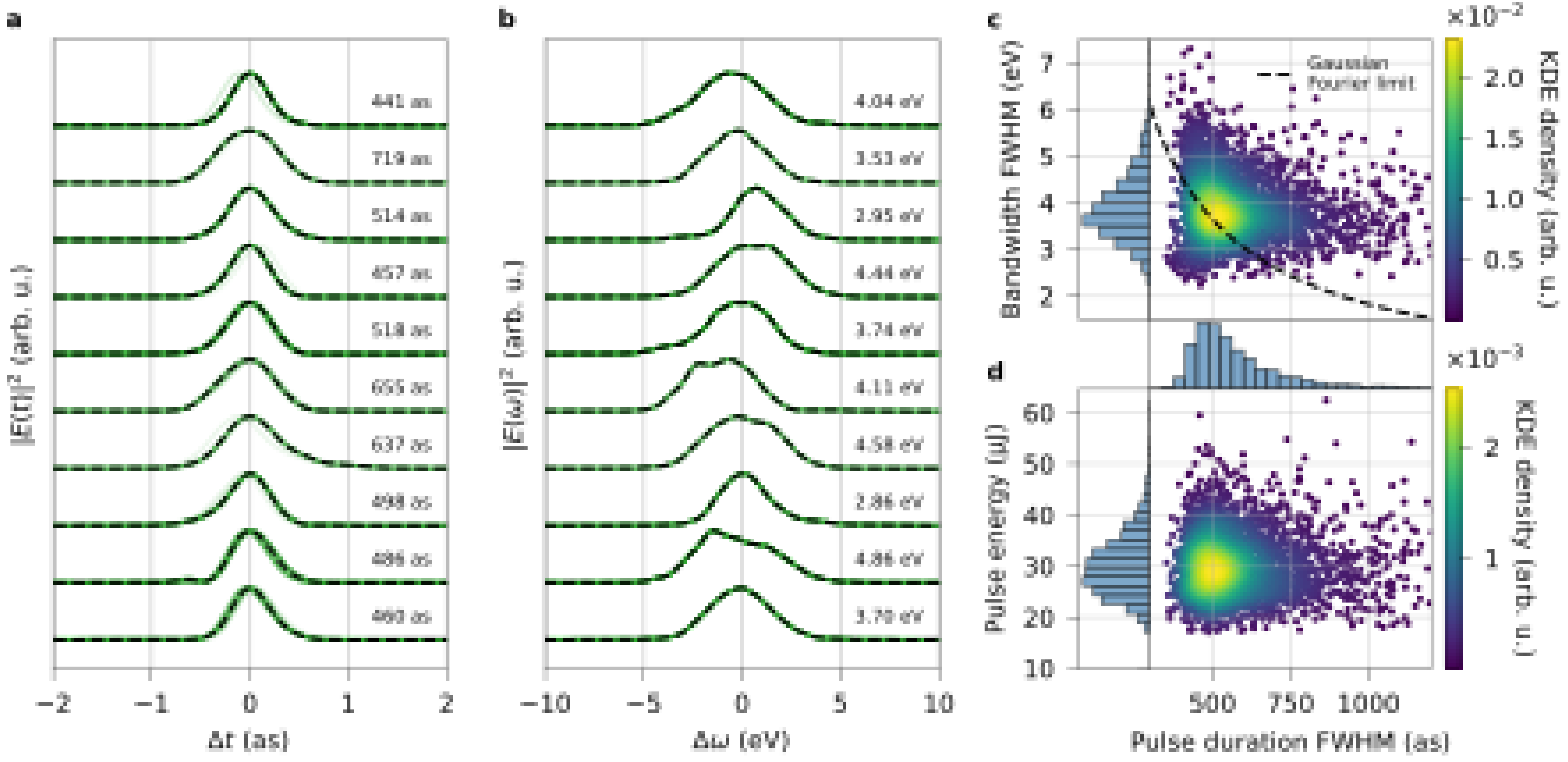
energy jitter: 1e-4,
peak current jitter: 4%,
arrival time jitter: 20 fs.

Attosecond Pulses at LCLS-II



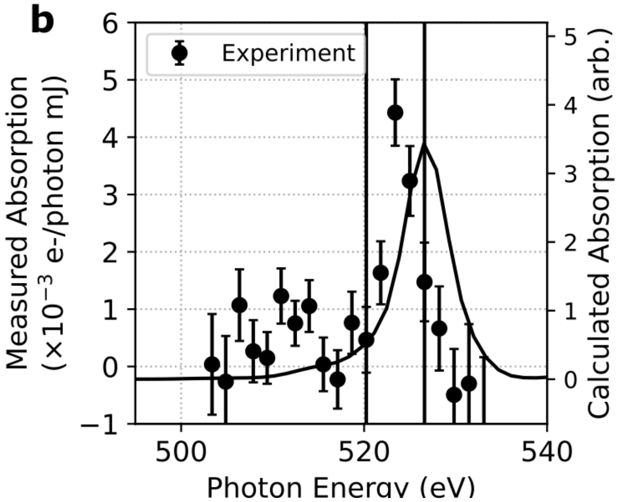
Angular Streaking Measurements

Demostrated at 33 kHz
User operation at 16 kHz

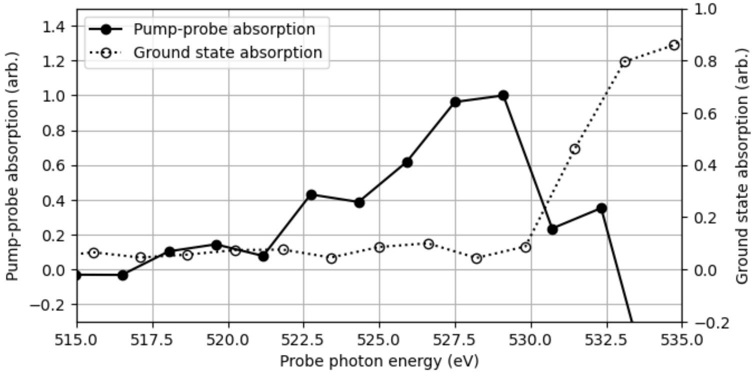


Recent Results at LCLS-II

Impulsive ionization in para-aminophenol

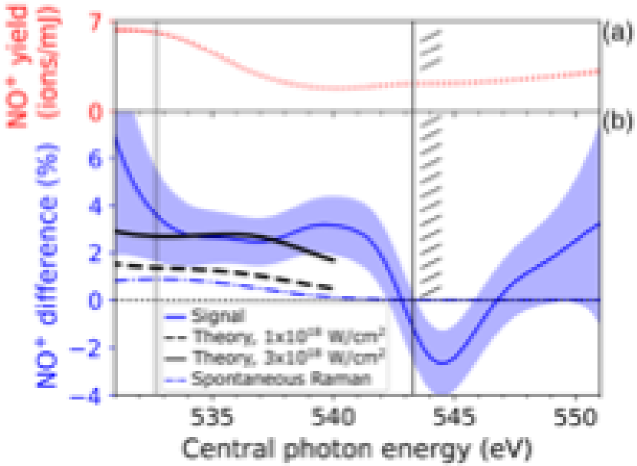


LCLS: 1-2 hours



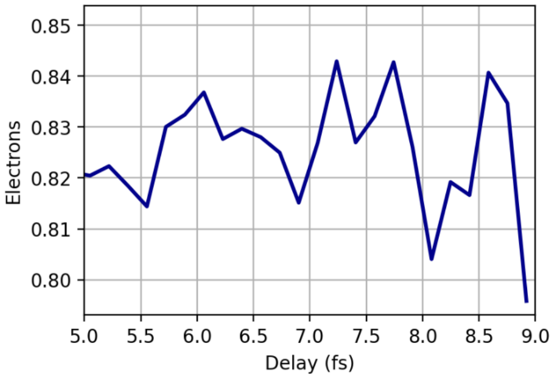
LCLS-II: ~1-2 minutes

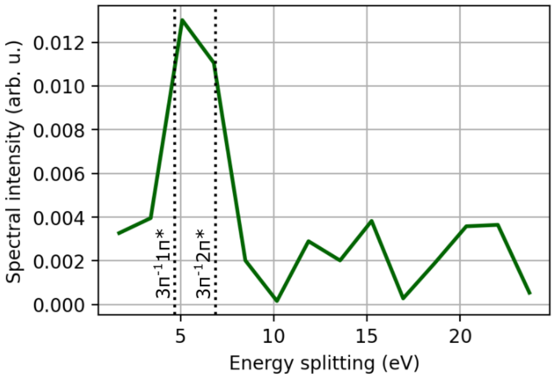
Stimulated Impulsive Raman Scattering



J. O'Neal et al. *PRL.* 125.7 (2020): 073203

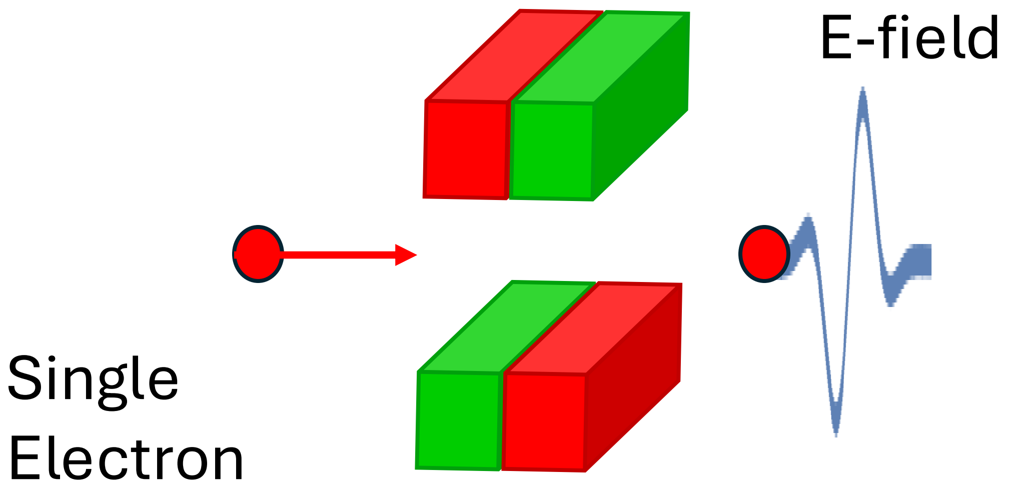
LCLS: 5 days for 1 data set (no time delay scan)



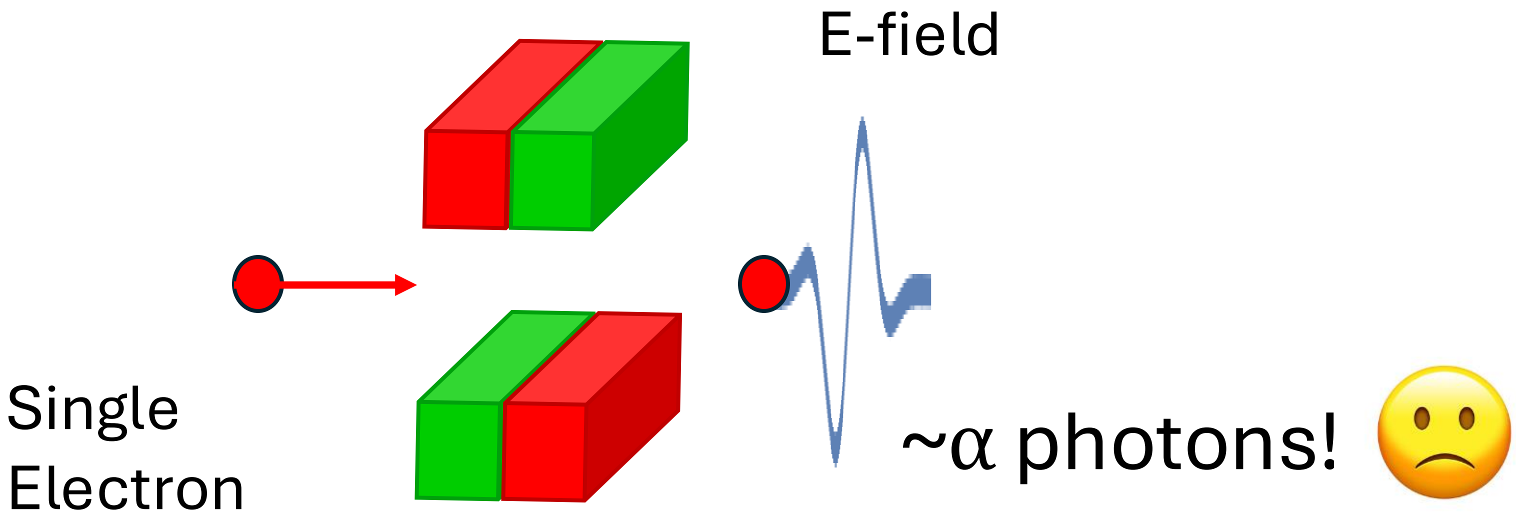


LCLS-II: 1 day, 18 data sets (pump/probe scan)
(Preliminary analysis)

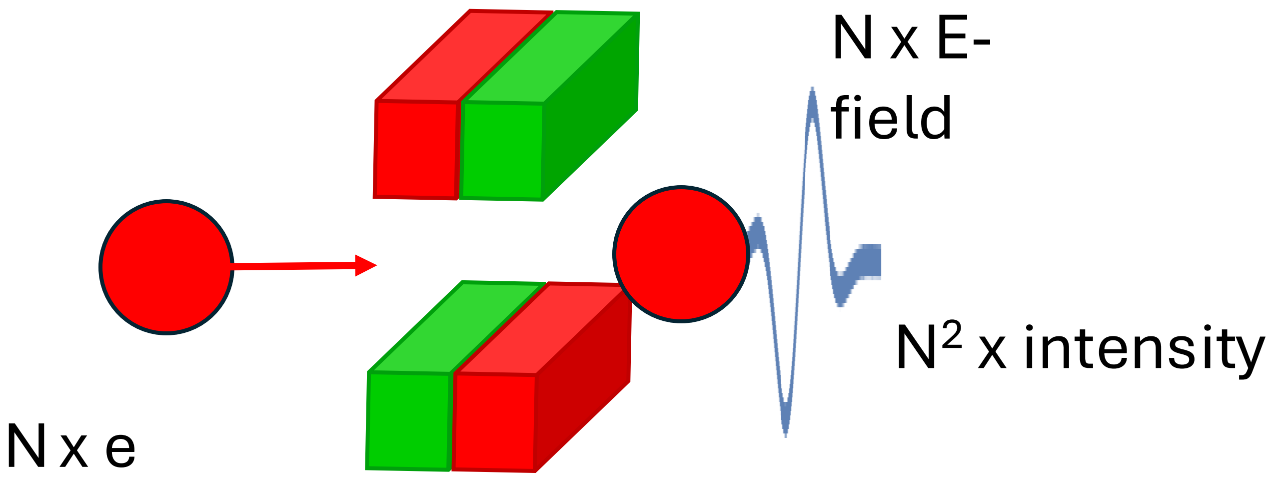
Looking Way Into the Future…



Looking Way Into the Future...

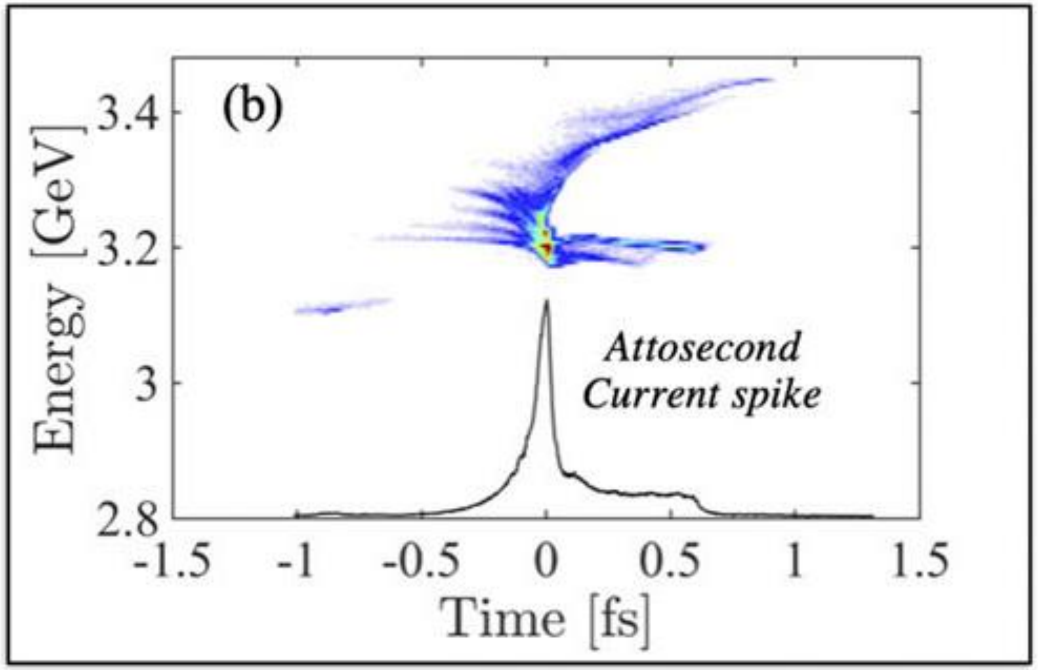


Looking Way Into the Future…



Plasma-Based Attosecond X-ray Pulses

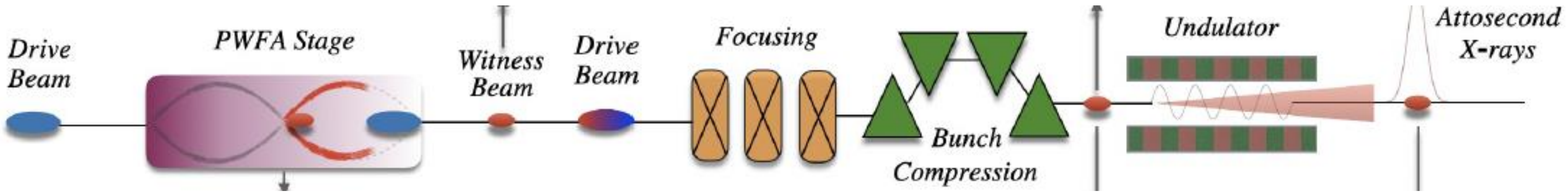
SIMULATION

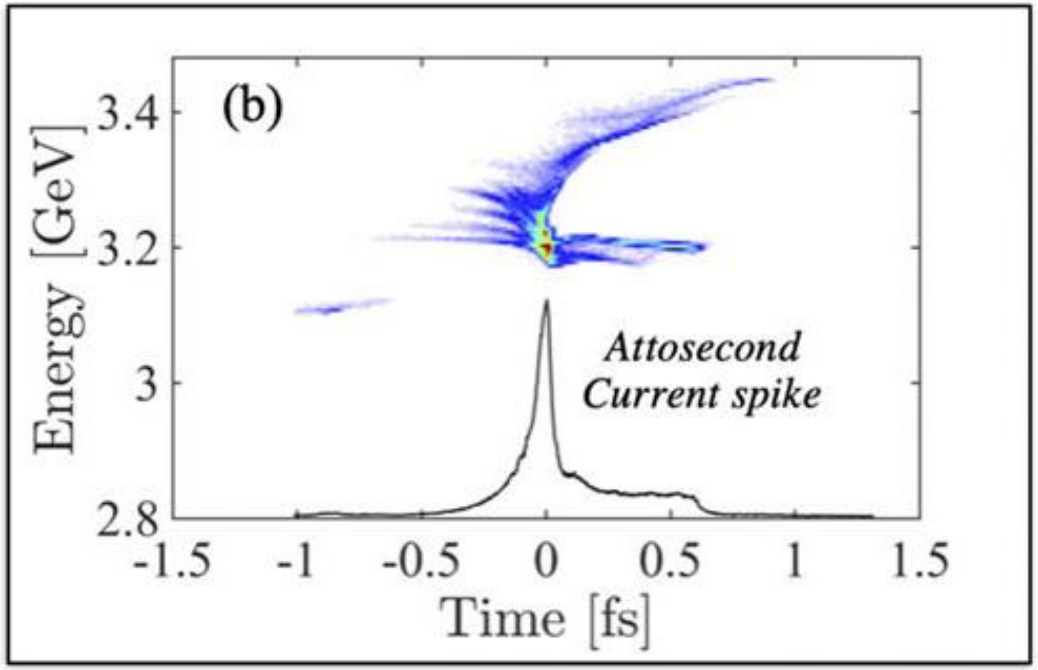


C. Emma et al. APL Photonics 6.7 (2021): 076107

C. Emma, X. Xu, R. Hessami,
R. Robles, K. Larsen
M. Hogan, **A. Marinelli**

Plasma-Based Attosecond X-ray Pulses



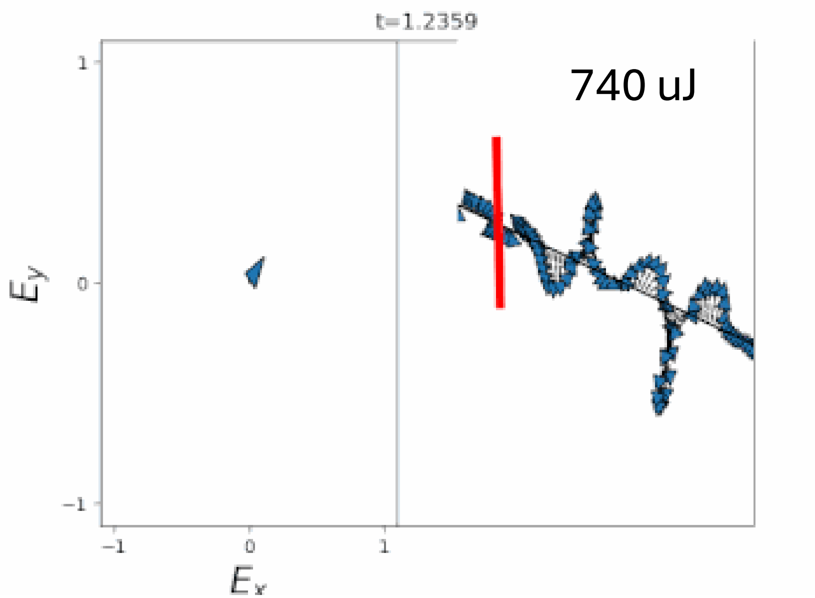


C. Emma et al. APL Photonics 6.7 (2021): 076107

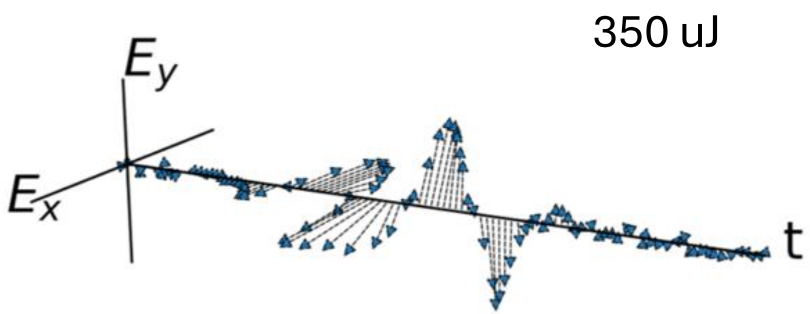
C. Emma, X. Xu, R. Hessami,
R. Robles, K. Larsen
M. Hogan, **A. Marinelli**

Single-Period Shaping (Sims at 10-30 nm)

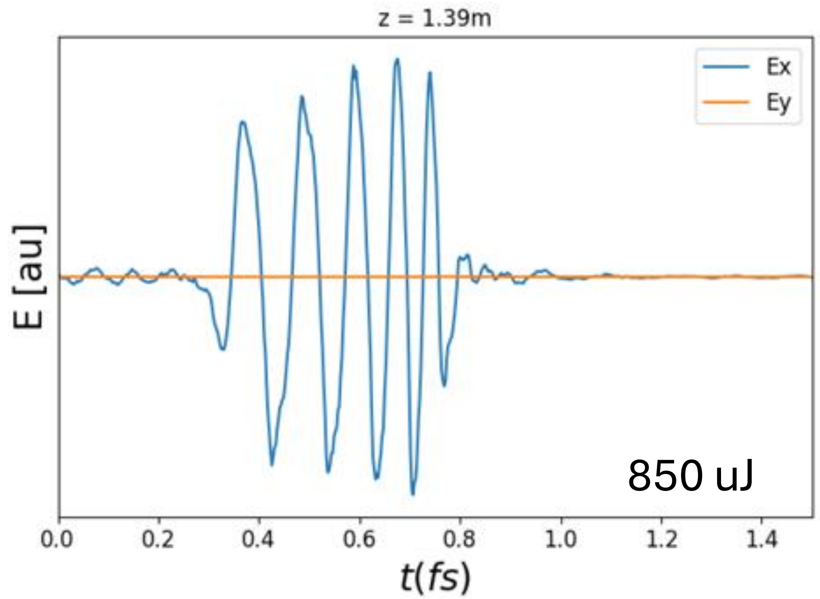
Circular polarization switching



Linear polarization switching



Extreme Chirp

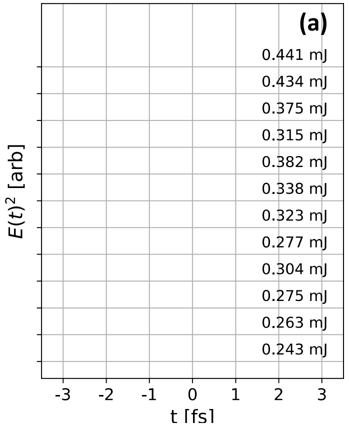


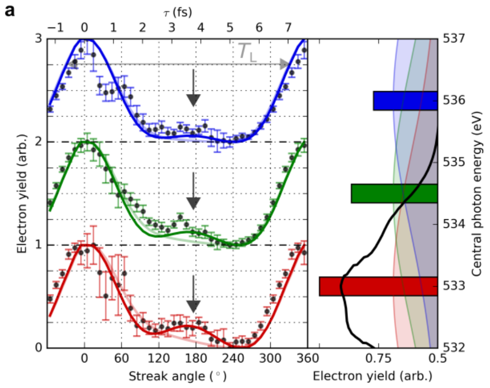
Non-SVEA simulations
Jenny Morgan (SLAC)

Stay Tuned…

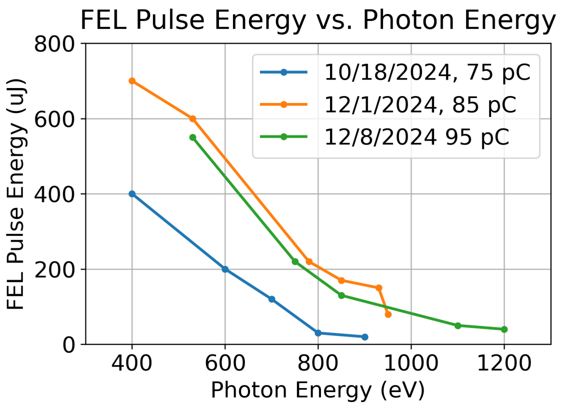
Summary

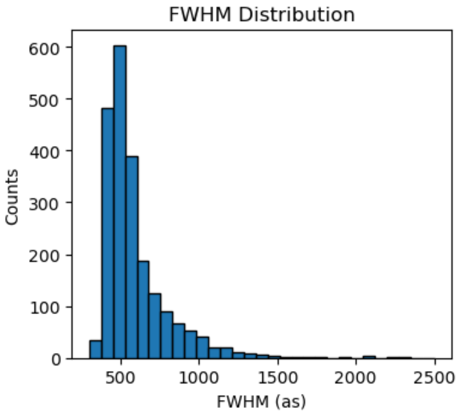
Attosecond or single spike pulses are now a reality at most XFEL facilities!



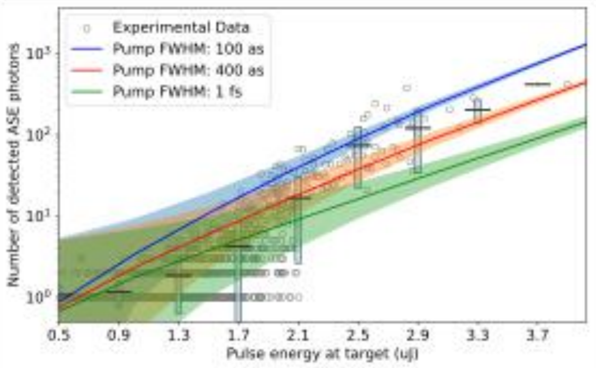


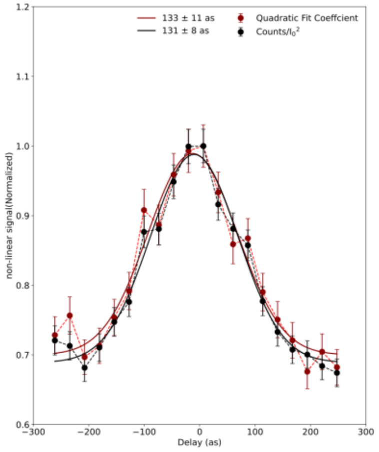
LCLS-II is up and running. Attosecond pulses at high rep-rate.





Hard X-ray Attosecond Spectroscopy and Metrology





Path to Single-Cycle Waveform Shaping with Plasma Wakefield Acceleration