AMO Physics with VUV and X-ray FELs

John T. Costello
School of Physical Sciences and NCPST

ISP – CUSAT – Silver Jubilee – Mar 19, 2021

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Photoionization of Ne and Kr in Intense VUV and X-ray Laser Fields

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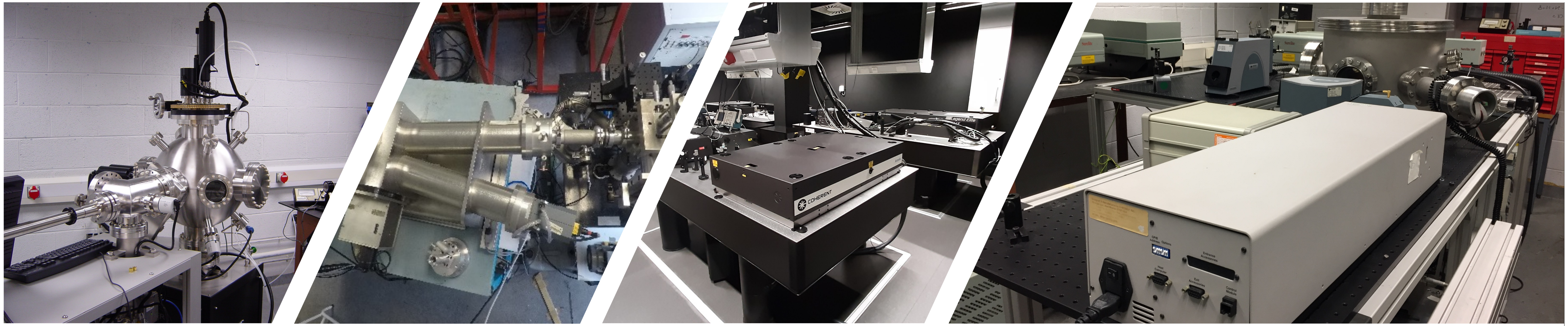
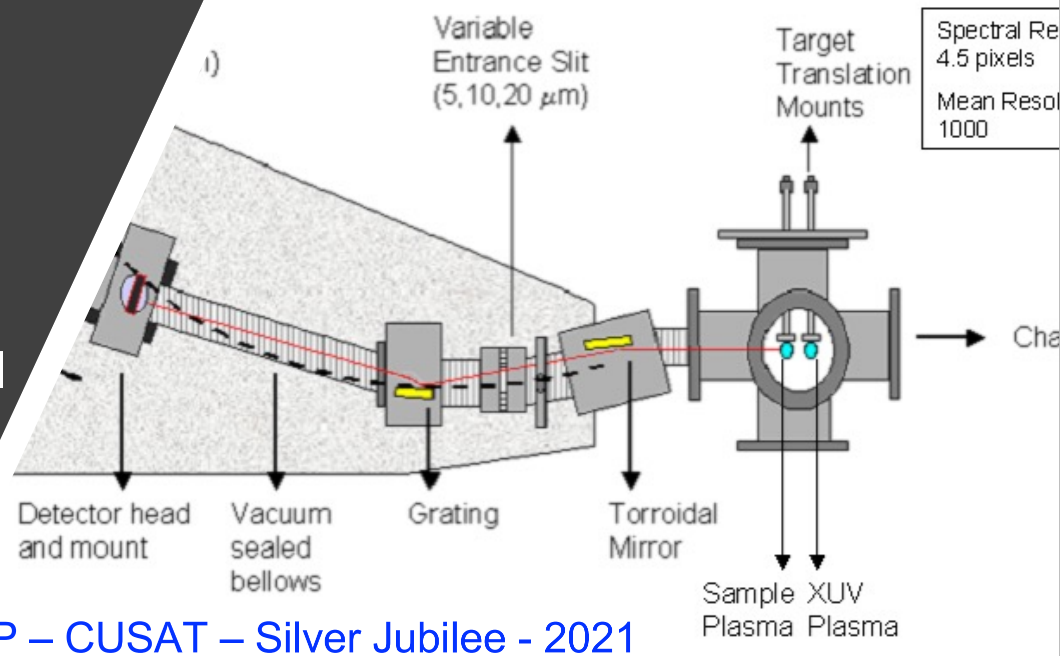


AMO & Laser Plasma Physics @ DCU

Six laboratory areas focused on pulsed laser matter interactions (spectroscopy/ imaging / particle detection) - http://www.physics.dcu.ie/~jtc/expfacil.html



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AMO & Laser Plasma Physics @ DCU

Principal Investigator(s): John T. Costello (LPPs and Free Electron Lasers), Eugene Kennedy+Jean-Paul Mosnier+Paul van Kampen (LPPs and Synchrotrons) and Lampros Nikolopoulos (Theory Group)

Current Research student(s): Muhammed Bilal Alli, Sadaf Syedah Zehra, James Campbell, Séamus Cummins, Stephen Durkan, Andrew Foremski, Ross McGarry and Adam Prior

Current Postdoc(s): Lazaros Varvarezos

Recent PhD Graduate(s) (2017-2020): Nichola Walsh, Ben Delaney, Stephen Davitt, Hu Lu, Getasew Wubetu, William Hanks, Lazaros Varvarezos, Tejaswi Katravulapally & Columb Doherty

Talk Outline

- Short wavelength free electron lasers (FELs)
- Two photon double ionization (TPDI) of krypton
- The 'atomic' streak camera
- Measuring the 'Photo to Auger' electron time delay for K shell ionization of neon
- Next steps

Some members of the AMO@FELs Collaboration

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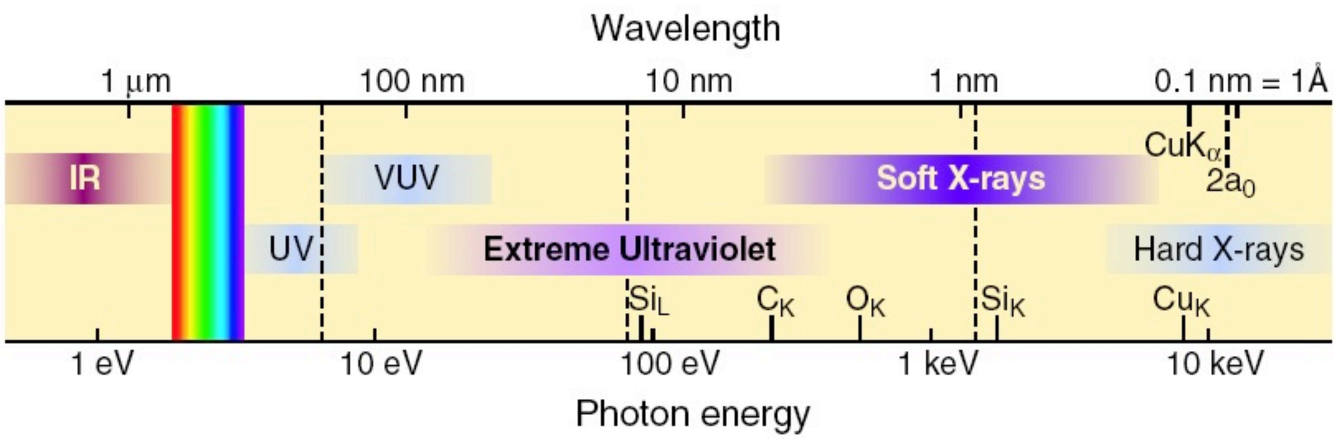
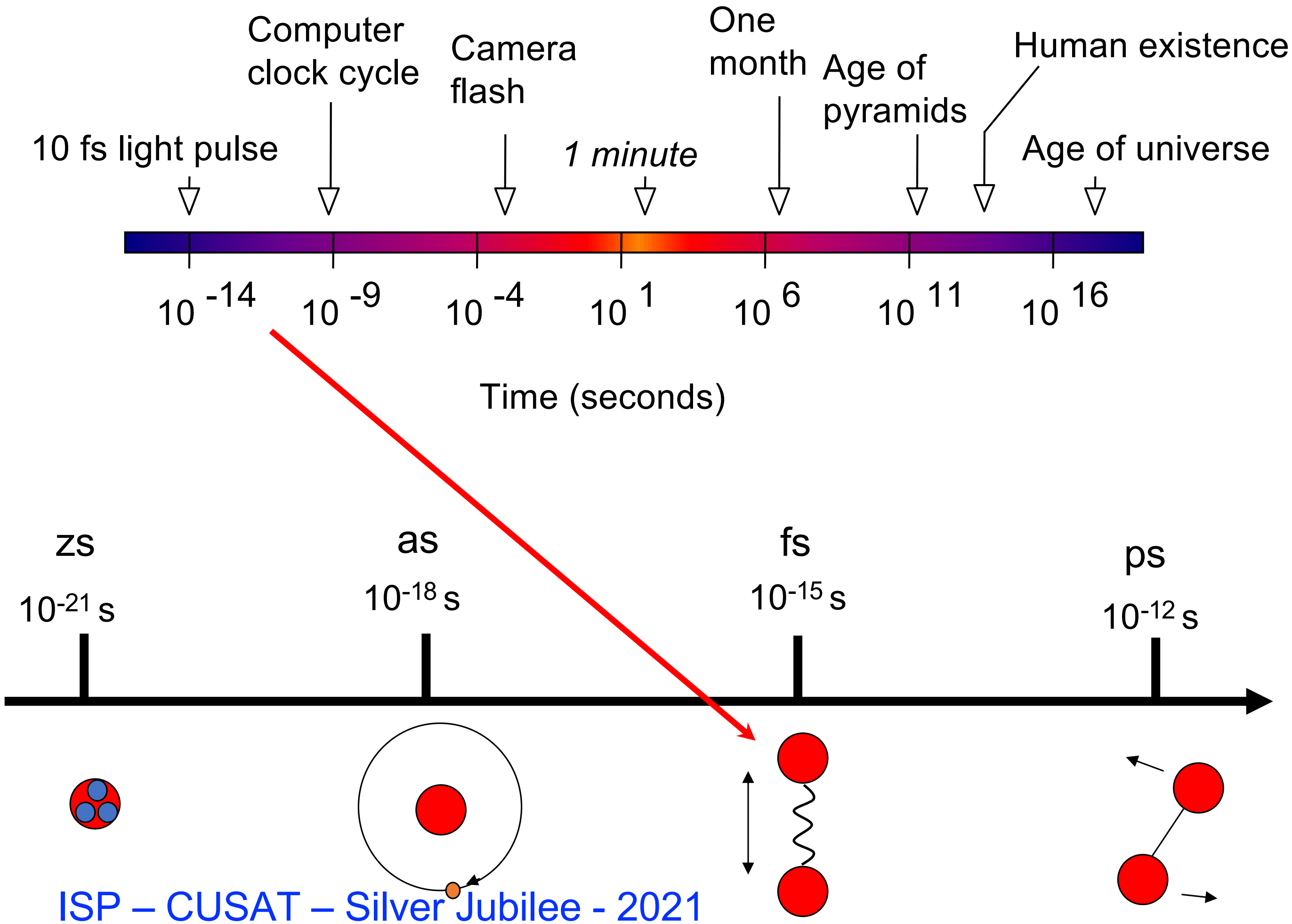
Orientation

Spectral Range: IR to the X-ray



Graphic: Courtesy of David Attwood (LBL)

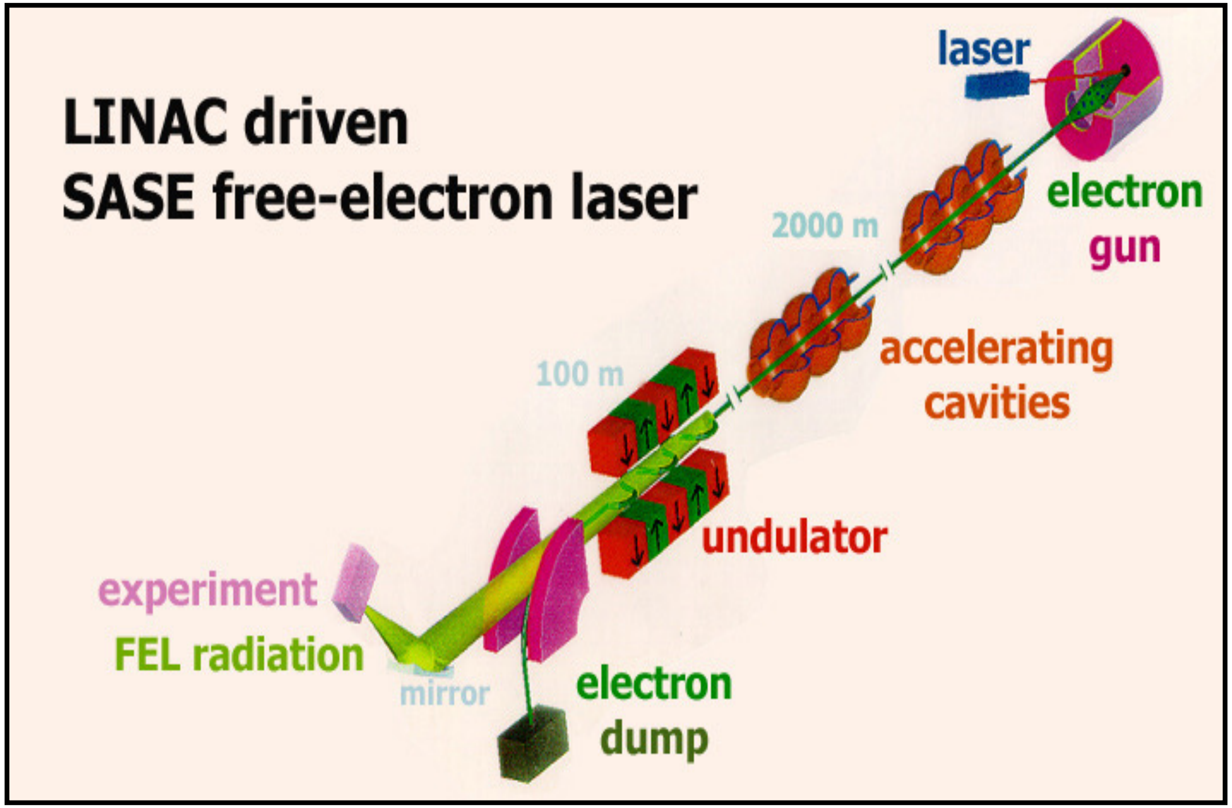
Orientation



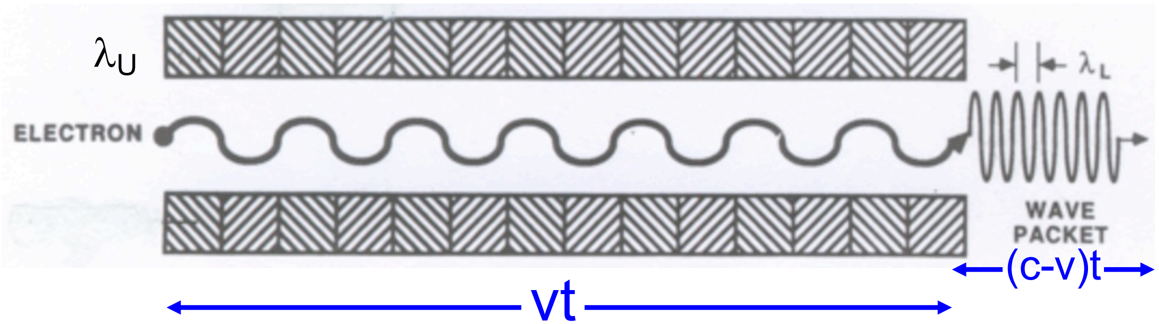
What do we want in a short wavelength (VUV – X-ray) laser?

The Holy Grail is an VUV/X-ray laser with variable pulse duration on the femtosecond to attosecond timescale with tunable wavelength, variable polarisation and high energy per pulse (few 100 μJ to few 10 mJ).

Main Components of an X-ray FEL



SASE-FEL, Fundamental Principle



$N_u\lambda_U = vt$ $\quad N_u\,\lambda_L = (c\text{-}v)t$ $\quad \Rightarrow\ \lambda_L \sim \lambda_U\,(c\text{-}v)/v \sim \lambda_u/2\gamma^2$

1GeV machine $\gamma \sim 2000$
$\lambda_u \sim 2.7$ cm / $\lambda_{laser} \sim 6$nm

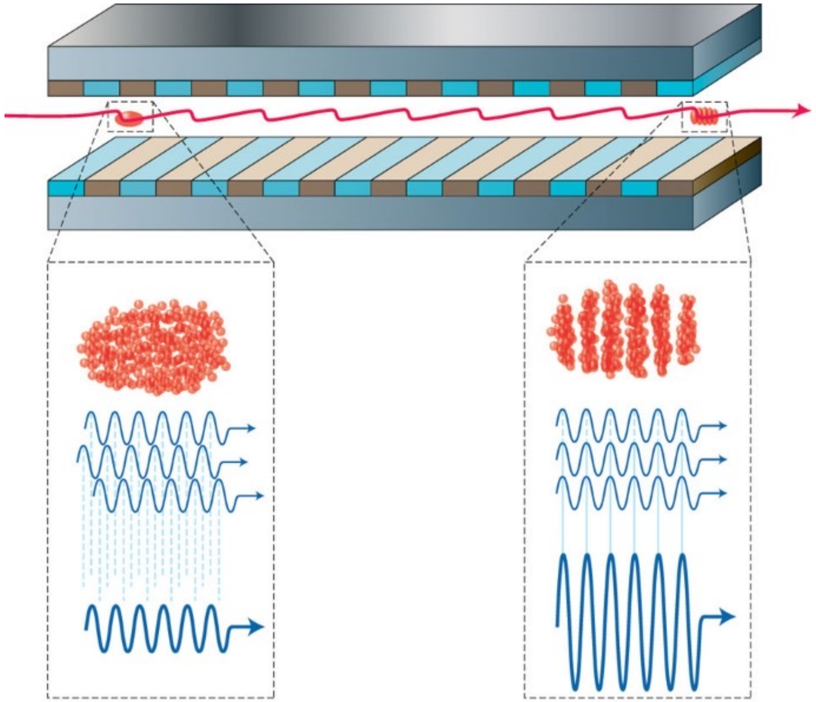
$\lambda_L = \lambda_u(1+K^2/2)/2\gamma^2$ $\quad \gamma = E/mc^2$

$K = eB\lambda_u/2\pi mc$

Wavelength tunable –
by electron beam energy or
by tuning the undulator gap

Electron bunch slips behind the lightwave by λ per undulator period

SASE-FEL, Fundamental Principle



The SASE FEL Instability
(regenerative feedback) –

The more coherent the radiation field, the more spatially coherent the electron bunch charge distribution. The more spatially coherent the electron bunch charge, the more coherent the FEL radiation field.

Nature Photonics 4, pp 814–821(2010)

FLASH FEL Hamburg - Physical Layout - 2007

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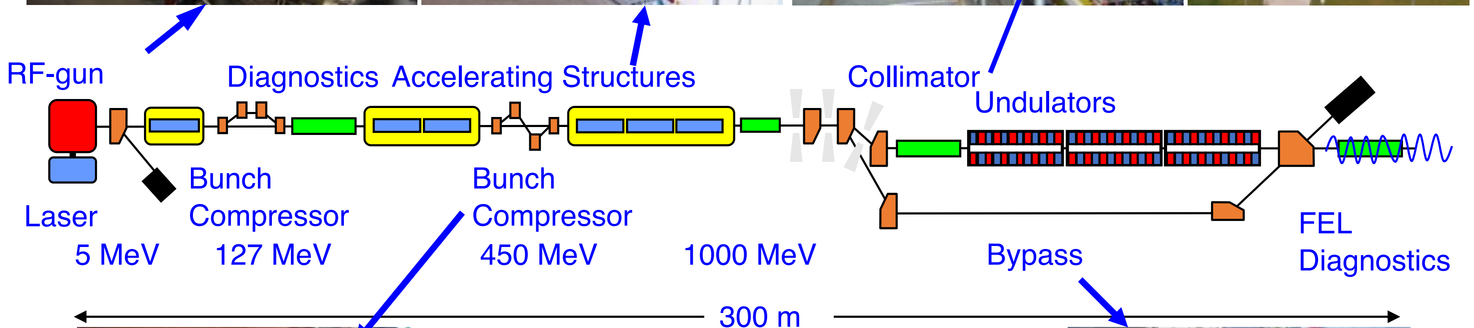
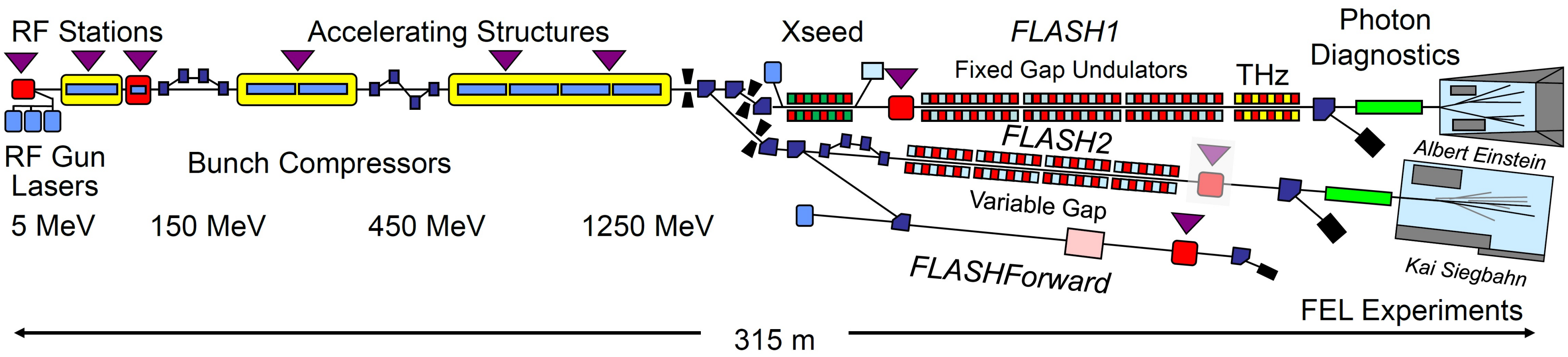
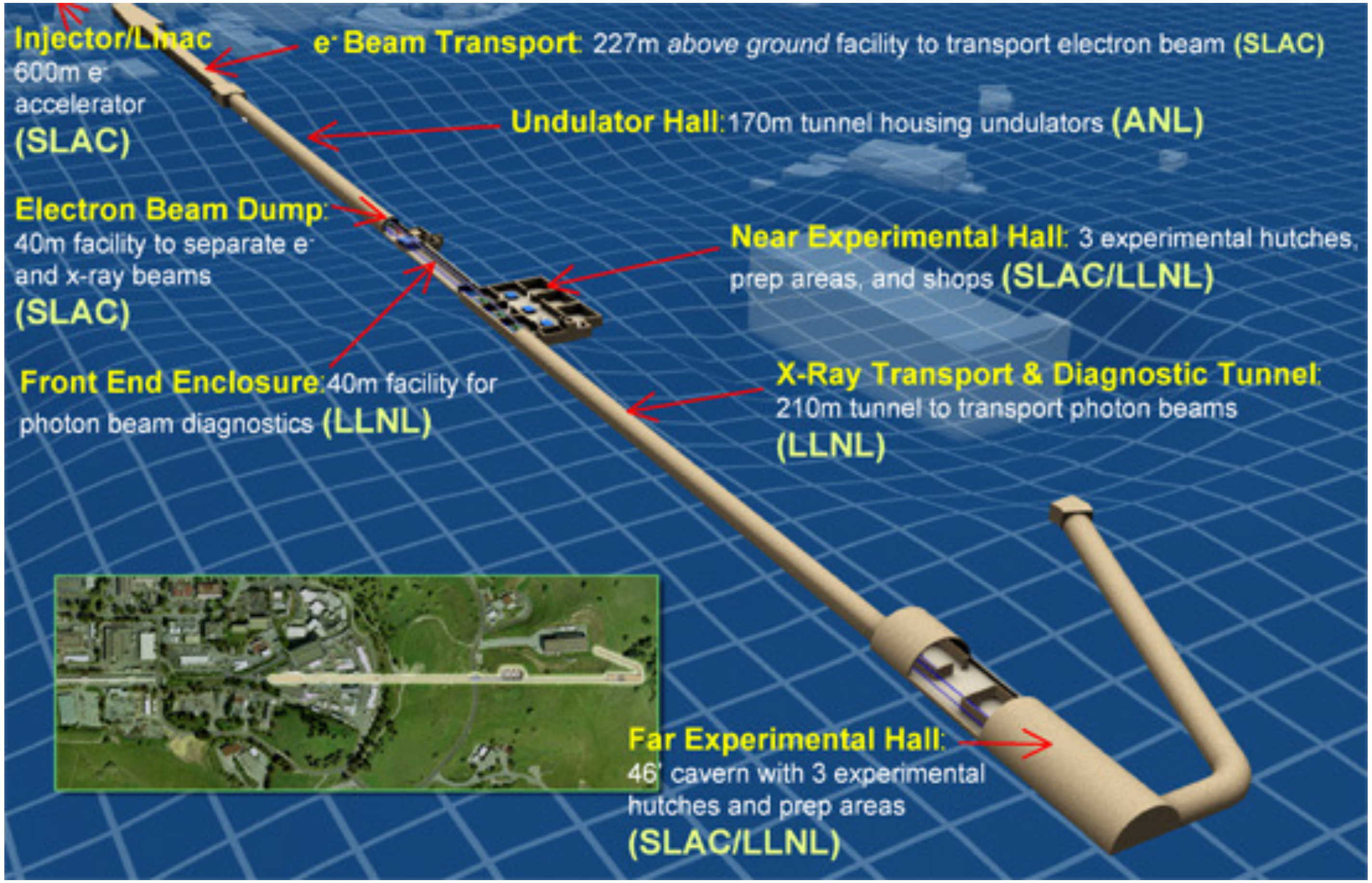


- LINAC Energy : ~ 1 GeV
  ~ 4 – 60 nm

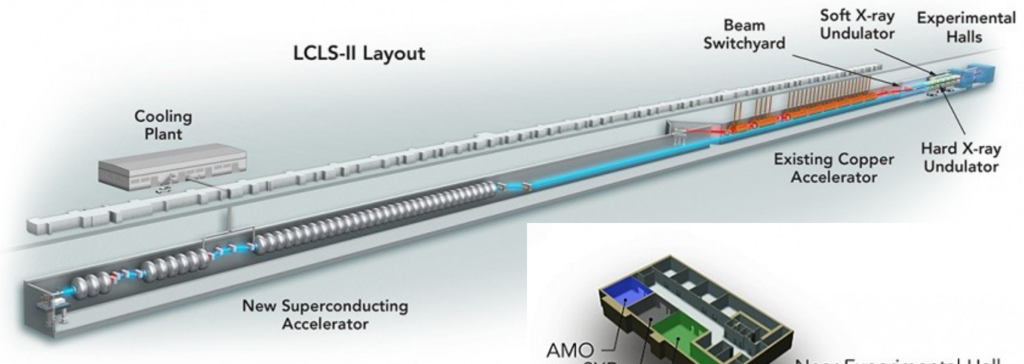
FLASH FEL Hamburg - Physical Layout 2020

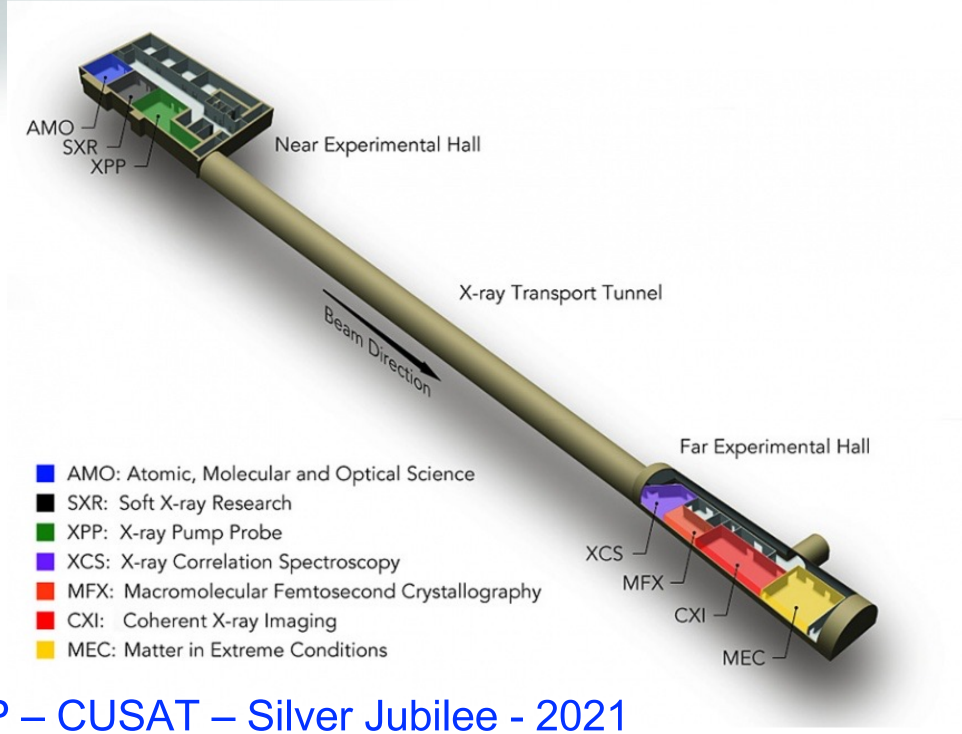


LCLS FEL SLAC Stanford - Physical Layout - 2011



LCLS II FEL SLAC Stanford – June 2022





TPDI of Kr at FLASH - Hamburg

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PHYSICAL REVIEW A **103**, 022832 (2021)

Near-threshold two-photon double ionization of Kr in the vacuum ultraviolet

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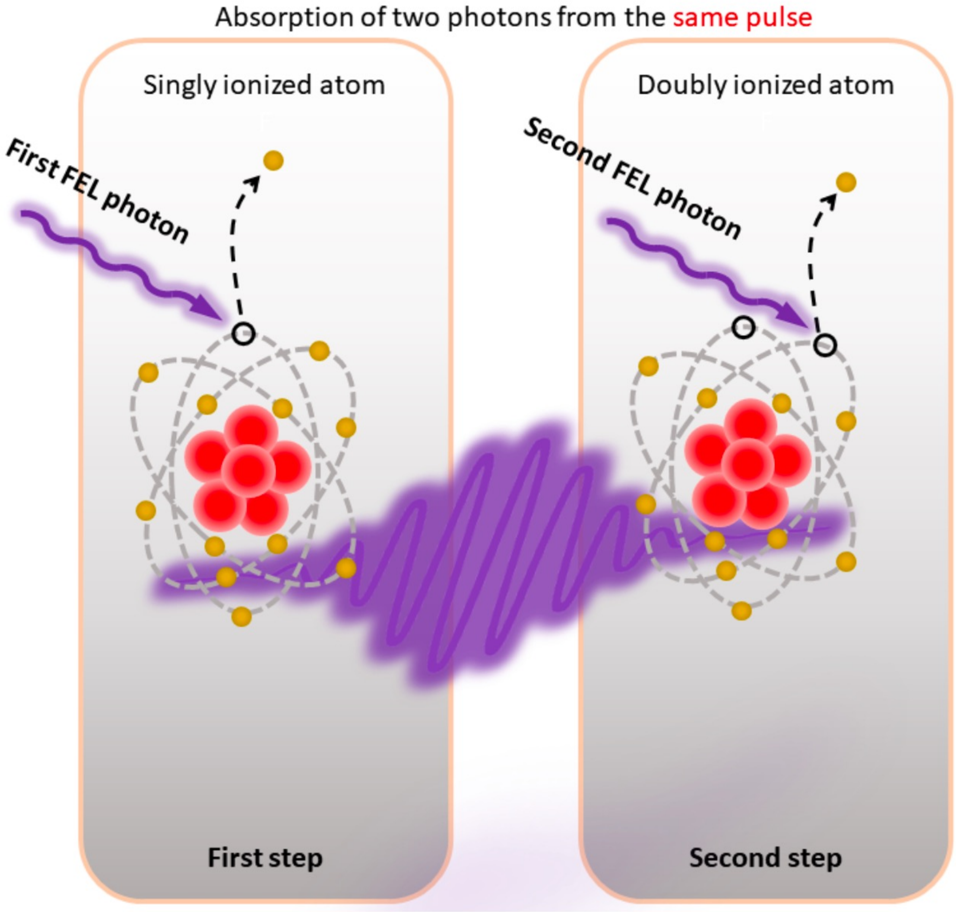
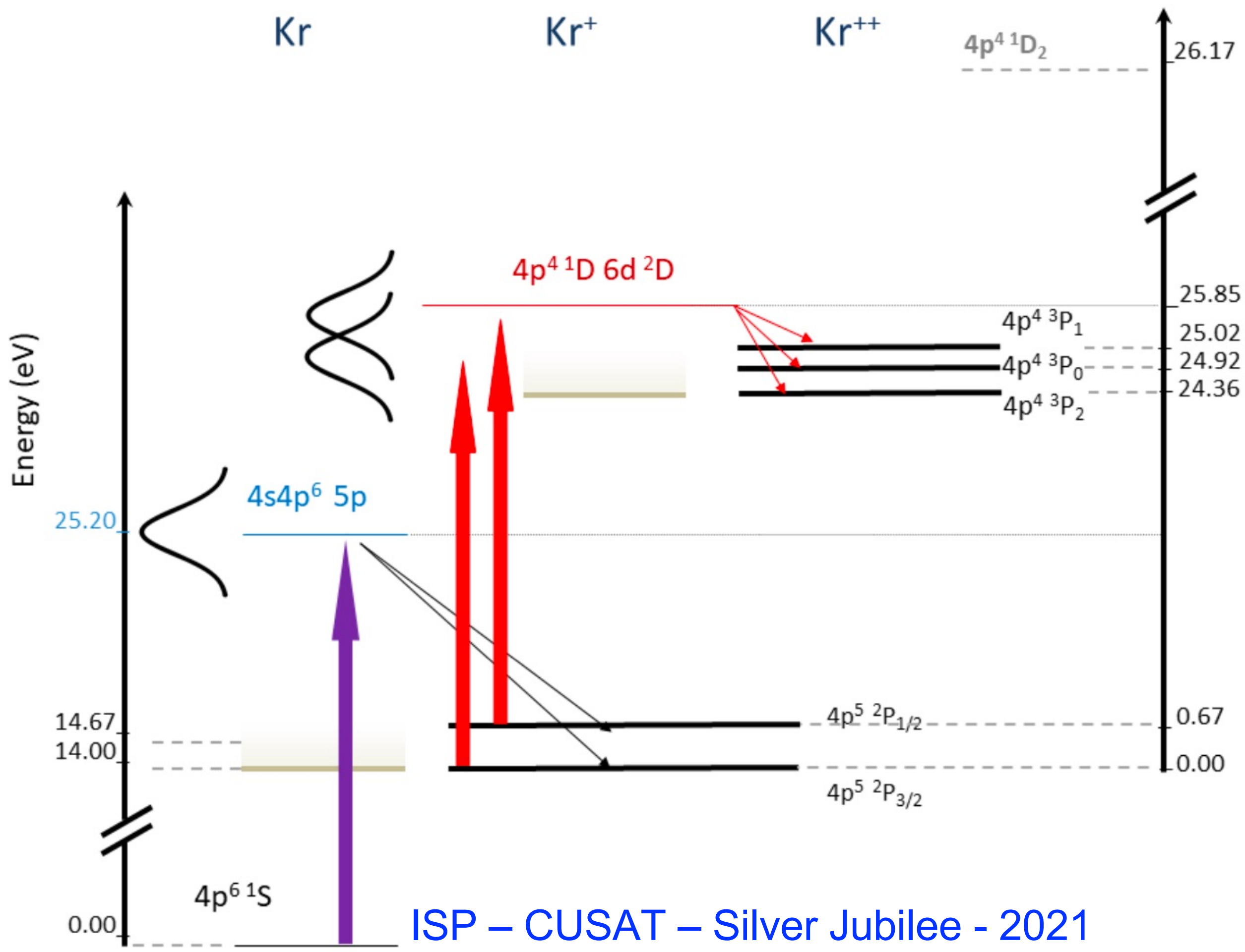
TPDI of Kr at FLASH - Hamburg



Source: L Varvarezos, PhD Thesis, DCU 2020

TPDI of Kr – Energy Level Scheme



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TPDI of Kr within ~40 fs VUV pulse with hν =25.2 eV

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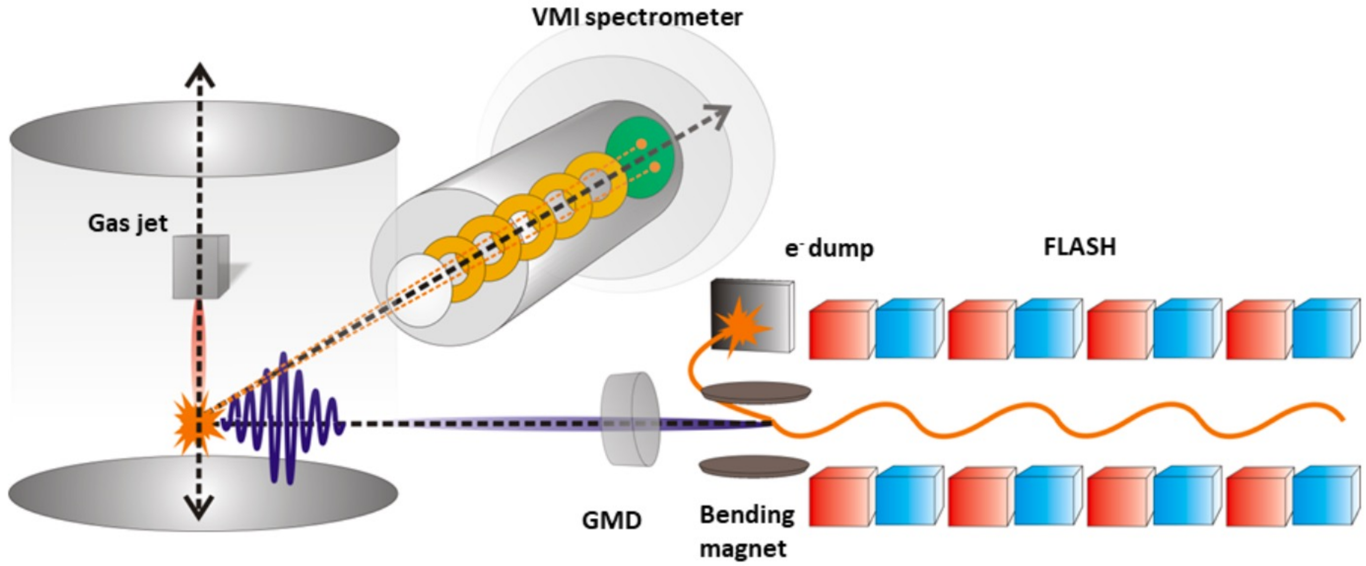
So near resonant excitation in both steps:

Kr (4p^6 $^1S_0$) + hν – Kr (4s4p^{6}5p $^1P_1$) – Kr^+ (4p^5 $^2P_{1/2,3/2}$) + e^-

followed by

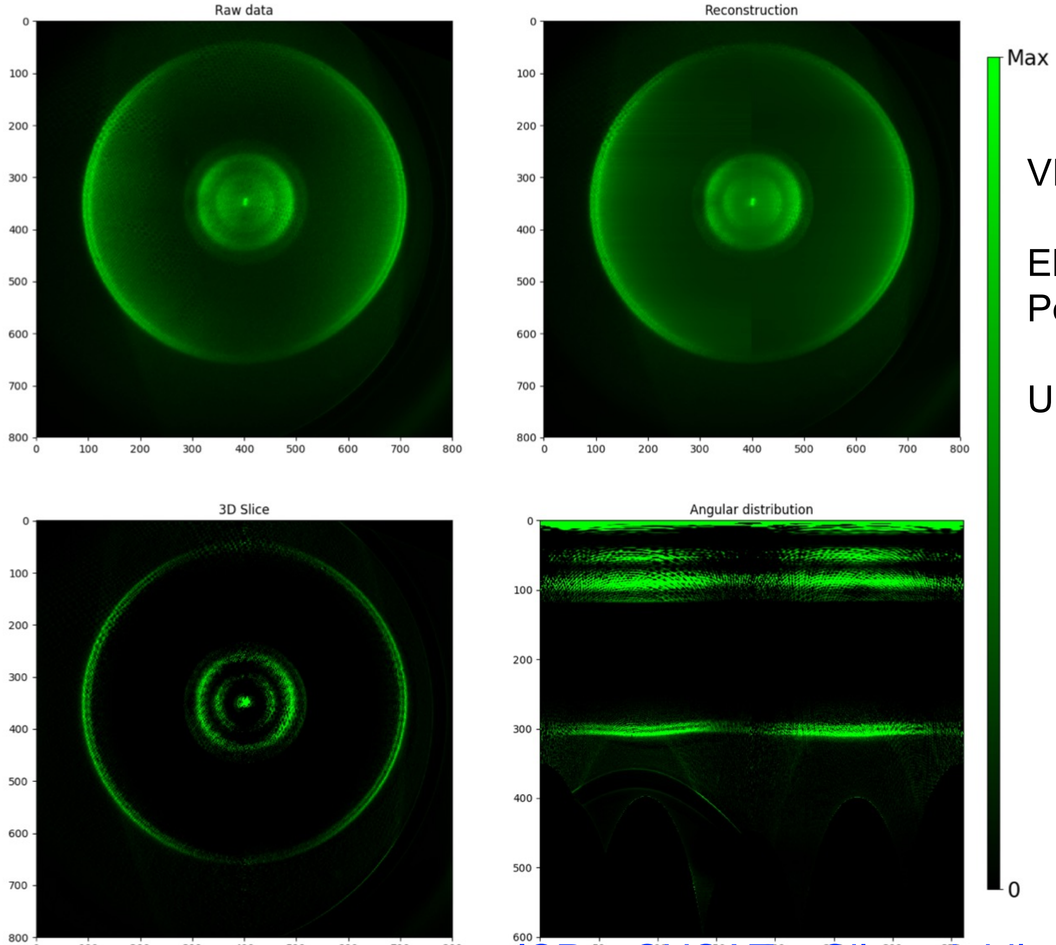
Kr^+ (4p^5 $^2P_{1/2,3/2}$)+ hν – Kr^+ (4p^{4}4d $^2D_{3/2,5/2}$) – Kr^{2+} (4p^4 $^3P_{2,1,0}$)+ e^-

Experimental Setup – VMI e^- Detection



Source: L Varvarezos, PhD Thesis, DCU 2020

Angle Resolved PES of Kr and Kr^+ for $h\nu$ = 25.2 eV



VMI = Velocity Map Imaging

Electron Kinetic Energy = Radial Position

Unfold to get ARPES

Source: L Varvarezos, PhD Thesis, DCU 2020

Photoelectron spectra (PES) of Kr & Kr^+ for $h\nu$ = 25.2 eV

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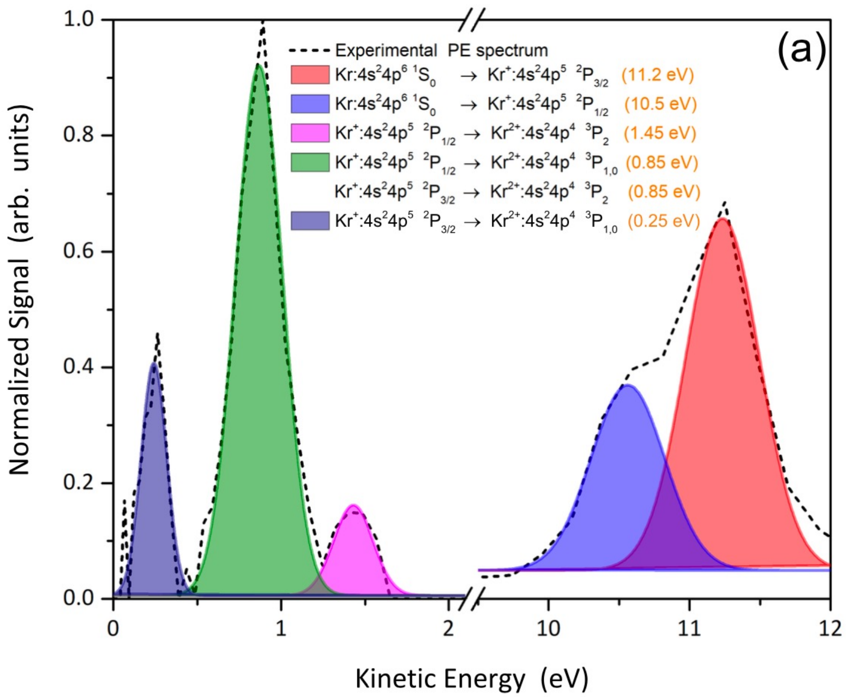
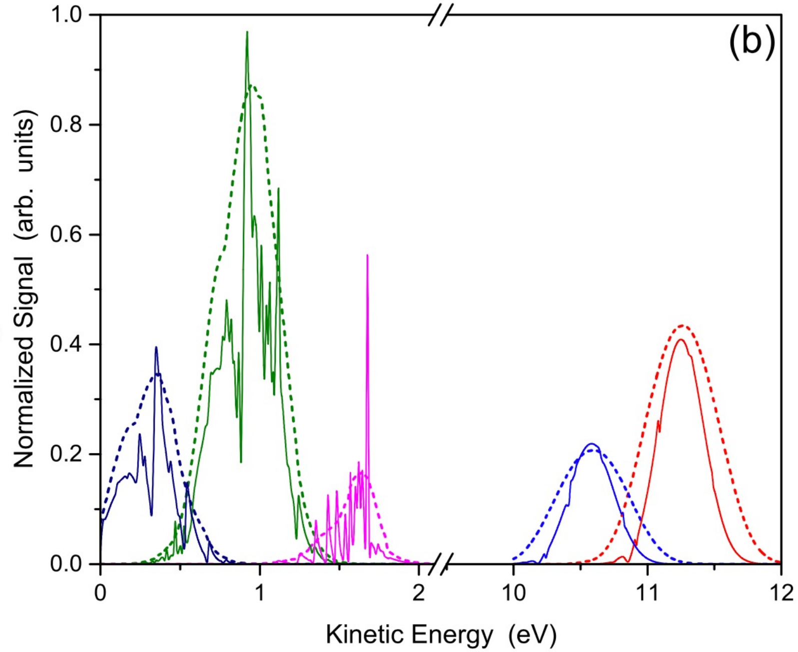


Experiment

Theory



One and Two Photon Angle Resolved PES (ARPES)

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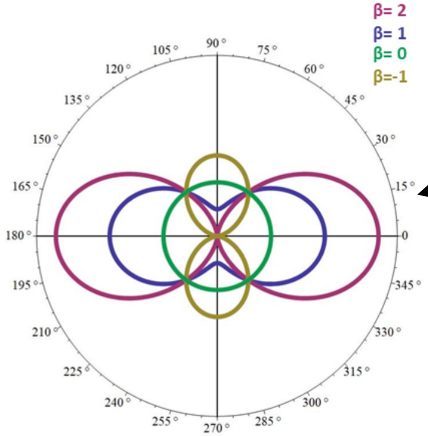
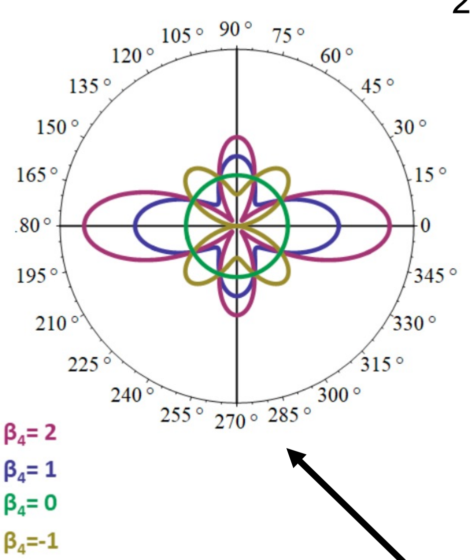
1 Photon PAD



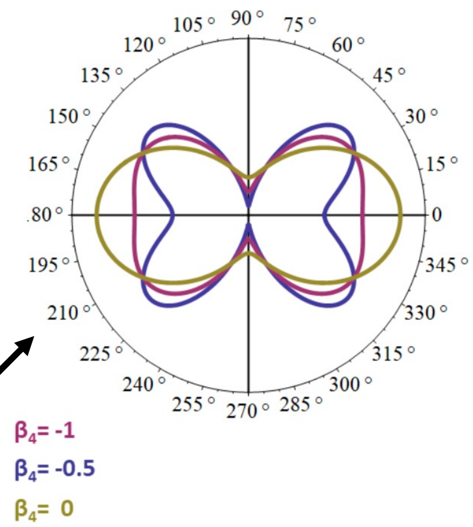
$$\frac{d\sigma(E)}{d\omega} = \frac{\sigma(E)}{4\pi}(1 + \beta_2(E) * P_2(cos\theta))$$

2 Photon PAD

β_2= 0



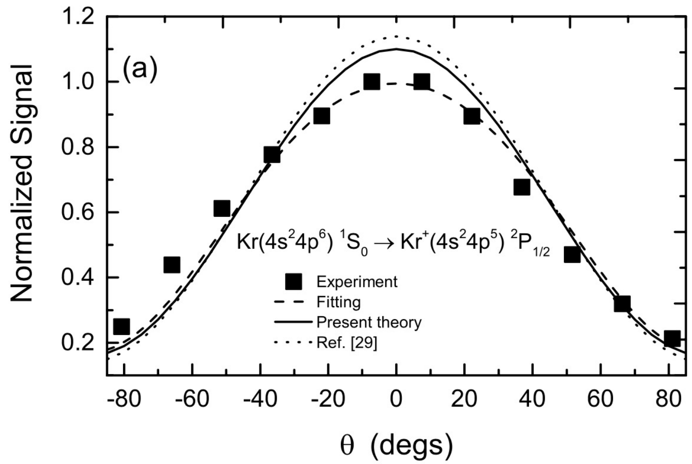
β_2= 1



$$\frac{d\sigma(E)}{d\omega} = \frac{\sigma(E)}{4\pi}(1 + \beta_2(E) * P_2(cos\theta) + \beta_4(E) * P_4(cos\theta))$$

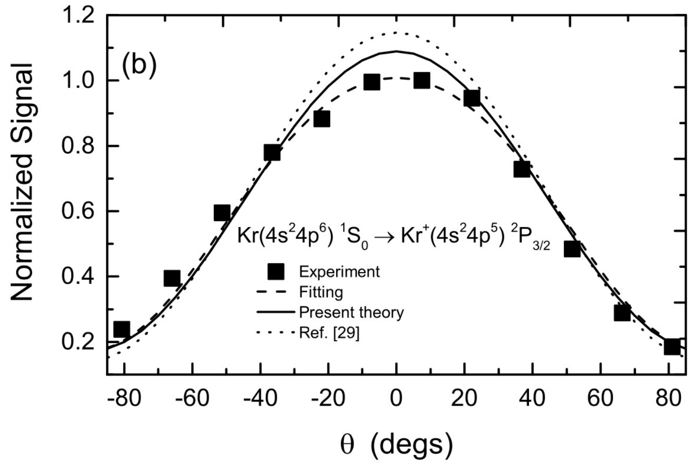
Source: L Varvarezos, PhD Thesis, DCU 2020

ARPES of Kr (1st Step) for hν = 25.2 eV



Kr – Kr$^+$ ($^2P_{1/2}$)

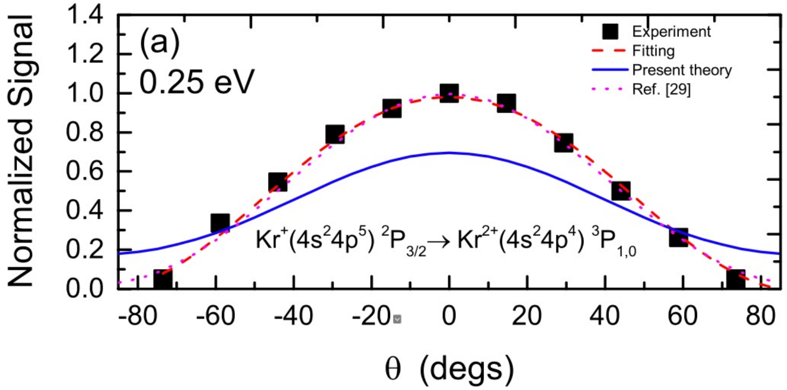
$$\frac{d\sigma(E)}{d\omega} = \frac{\sigma(E)}{4\pi}\left(1 + \beta_2(E) * P_2(cos\theta)\right)$$



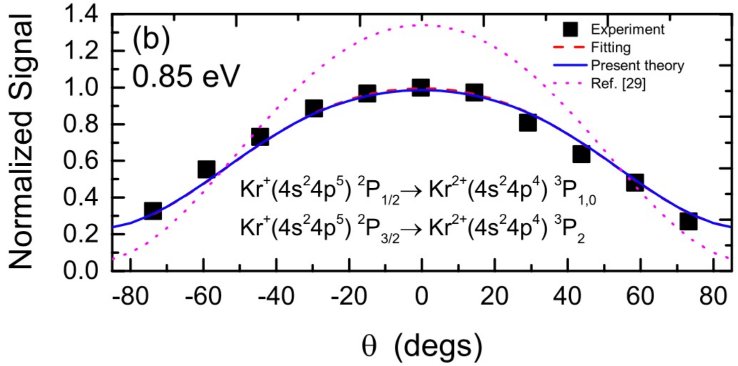
Kr – Kr$^+$ ($^2P_{3/2}$)

ARPES of Kr^+ (2^{nd} Step) for $h\nu$ = 25.2 eV

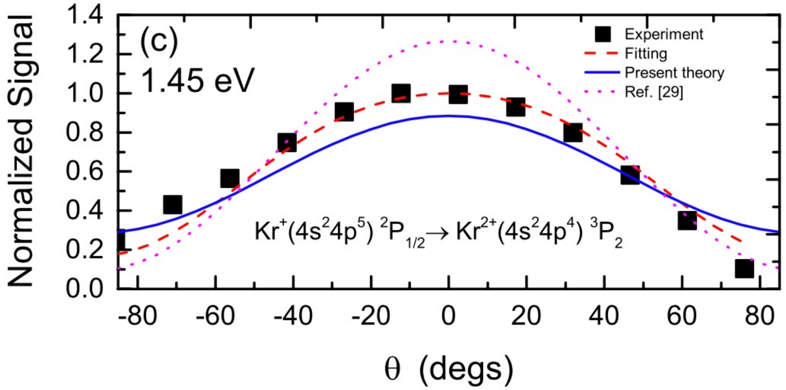
Kr^+ ($^2P_{3/2}$) – Kr^{2+} ($^3P_{1,0}$)



Kr^+ ($^2P_{1/2}$) – Kr^{2+} ($^3P_{1,0}$)
Kr^+ ($^2P_{3/2}$) – Kr^{2+} ($^3P_2$)



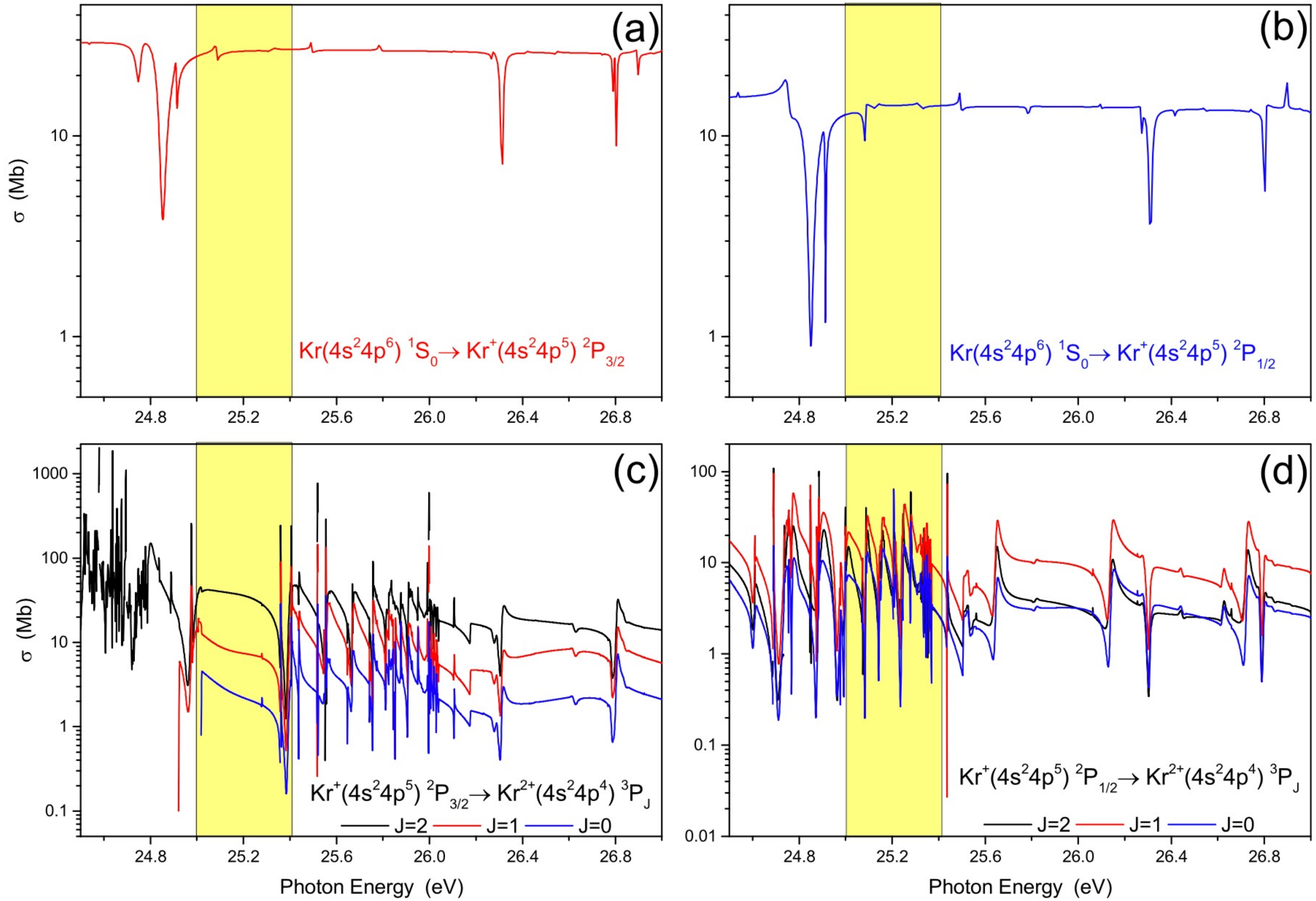
Kr^+ ($^2P_{1/2}$) – Kr^{2+} ($^3P_2$)



Extracted β_2 & β_4 values

	1st step		2nd step		
E_e, eV	11.2	10.5	1.4*	0.8	0.2
β_2	1.29	1.32	0.93	1.0	1.03
β_4	-	-	−0.02	−0.13	0.08

Resonances within the FEL BW complicates matters



Summary Kr TPDI

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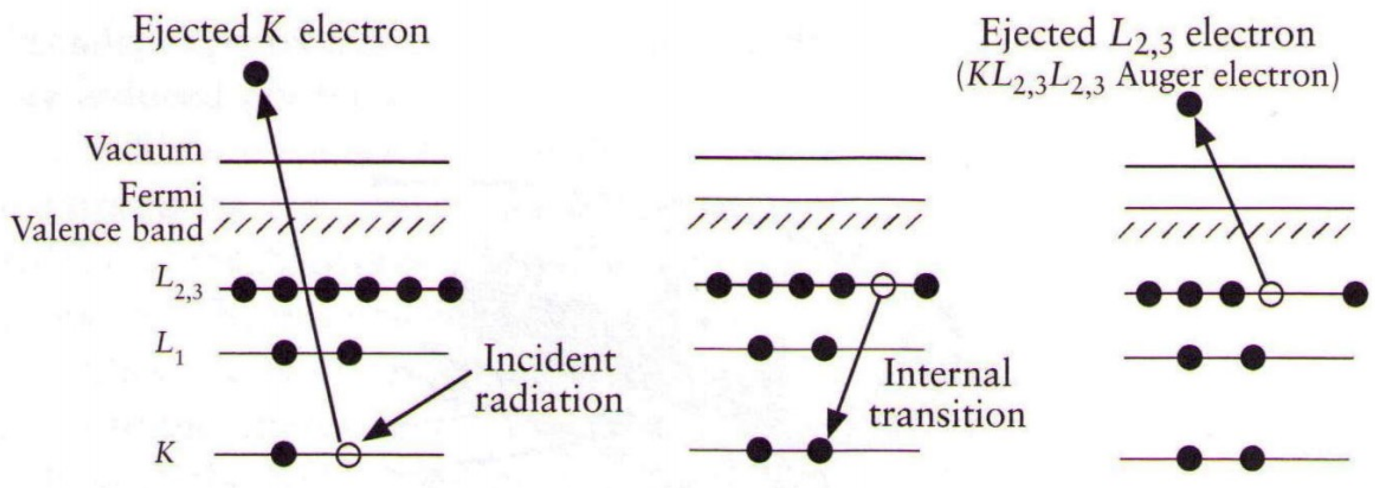
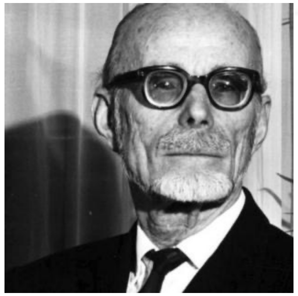
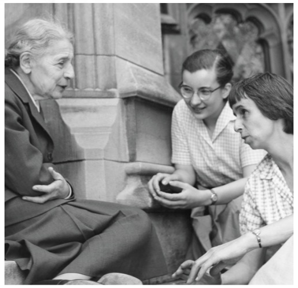
- Photoelectron angular distributions (PADs) have been measured for sequential two-photon double ionization of Kr at an FEL photon energy close to the second ionization threshold.
- The fine structure in the initial and final states in the first step ionization pathways and (partially) in the second step has been resolved for the first time in TPDI
- The (angle-integrated) photoelectron spectra exhibit good agreement with theory (R-Matrix)
- The inclusion of resonances (4s - nl and $4p^m$ - $4p^{m-2}$n'l,"nl") in the calculations is essential for reproducing the PADs (especially in the second ionization step, Kr^+ - Kr^{2+})

Photo to Auger electron emission delay time?



Primary
Photoelectron
Emission





Secondary
Auger electron
Emission

What is the time delay between photo and Auger electron emission?

P. Auger, *C. R. Hebd. Séances Acad. Sci.* **177**, 169 (1923)

L. Meitner, *Z. Phys.* **9**, 145 (1922)

Photo to Auger electron emission delay time?

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nature physics

ARTICLES

https://doi.org/10.1038/s41567-020-01111-0

Check for updates

Clocking Auger electrons

D. C. Haynes[1,2,3] ✉, M. Wurzer[4], A. Schletter[4], A. Al-Haddad[5,6], C. Blaga[7,8], C. Bostedt[5,6,9], J. Bozek[10], H. Bromberger[2,11], M. Bucher[6], A. Camper[7], S. Carron[12], R. Coffee[12], J. T. Costello[13], L. F. DiMauro[7], Y. Ding[12], K. Ferguson[12], I. Grguraš[1,2], W. Helml[4,14], M. C. Hoffmann[12], M. Ilchen[15,16], S. Jalas[17], N. M. Kabachnik[15,18], A. K. Kazansky[19,20,21], R. Kienberger[4], A. R. Maier[2,17], T. Maxwell[12], T. Mazza[15], M. Meyer[15], H. Park[7], J. Robinson[12], C. Roedig[7], H. Schlarb[11], R. Singla[1,2], F. Tellkamp[1,2], P. A. Walker[2,17], K. Zhang[7], G. Doumy[6], C. Behrens[11] and A. L. Cavalieri[1,2,3,5,22] ✉

Intense X-ray free-electron lasers (XFELs) can rapidly excite matter, leaving it in inherently unstable states that decay on femtosecond timescales. The relaxation occurs primarily via Auger emission, so excited-state observations are constrained by Auger decay. In situ measurement of this process is therefore crucial, yet it has thus far remained elusive in XFELs owing to inherent timing and phase jitter, which can be orders of magnitude larger than the timescale of Auger decay. Here we develop an approach termed 'self-referenced attosecond streaking' that provides subfemtosecond resolution in spite of jitter, enabling time-domain measurement of the delay between photoemission and Auger emission in atomic neon excited by intense, femtosecond pulses from an XFEL. Using a fully quantum-mechanical description that treats the ionization, core-hole formation and Auger emission as a single process, the observed delay yields an Auger decay lifetime of $2.2^{+0.2}_{-0.3}$ fs for the KLL decay channel.

The Atomic Streak Camera (time delays in photoionization)

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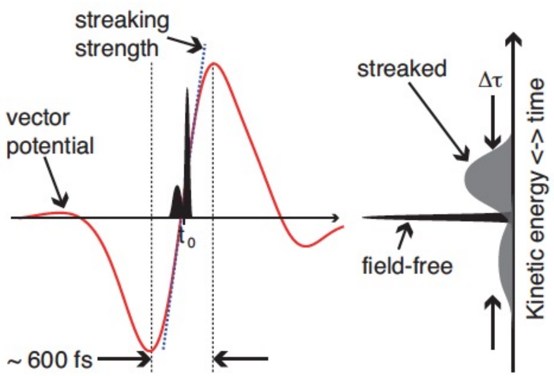
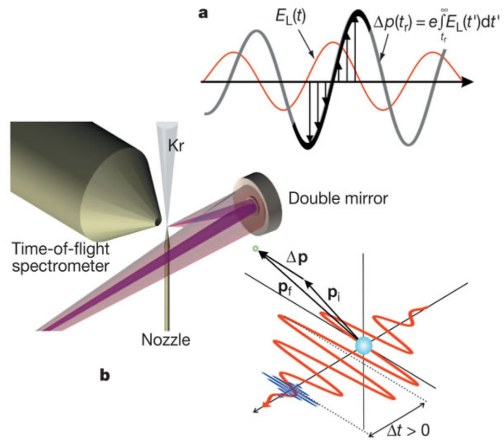
Generate single (picosecond) cycle pulse using optical rectification of Ti-Sapphire laser pulses – field ~ 50MV/m maximum

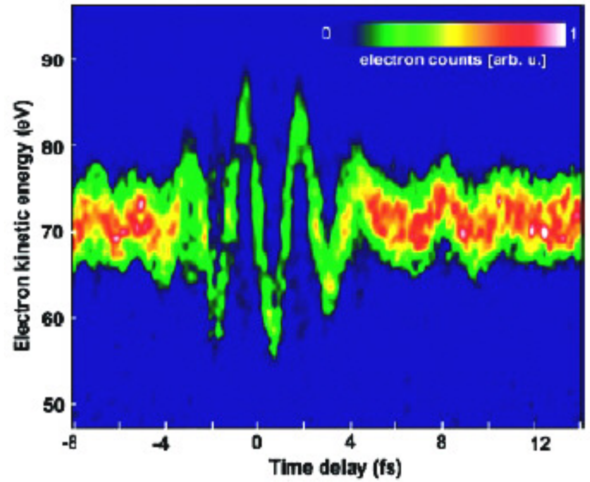


Principle of the experiment

Attosecond Photoelectron Streaking showing how the E-field of a few cycle fs laser pulse can be mapped – MPI-Q.

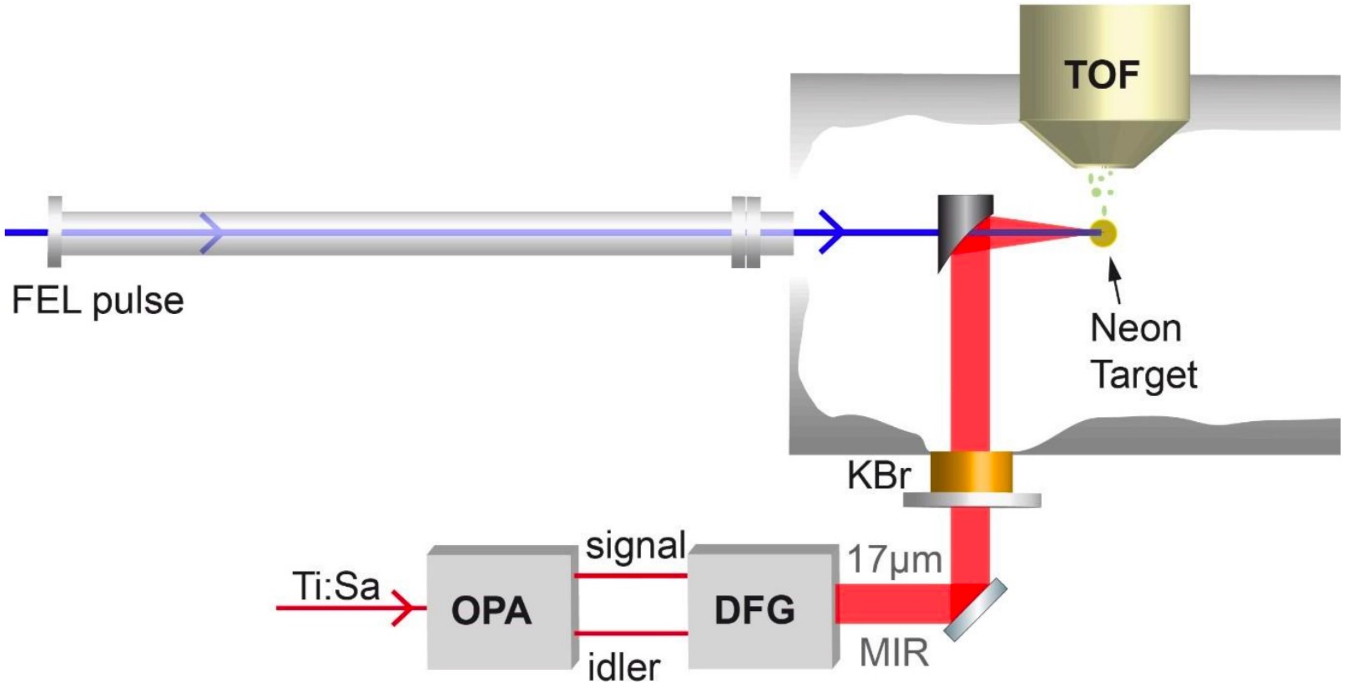
I Grguras et al. *Nature Photonics* **6** pp852-857 (2012)





Nature **419,** pp 803–807(2002)

The Atomic Streak Camera Experiment



Haynes, D.C. *et al.* Clocking Auger electrons. *Nat. Phys.* (2021). https://doi.org/10.1038/s41567-020-01111-0

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The Atomic Streak Camera (computing the streaked energy)

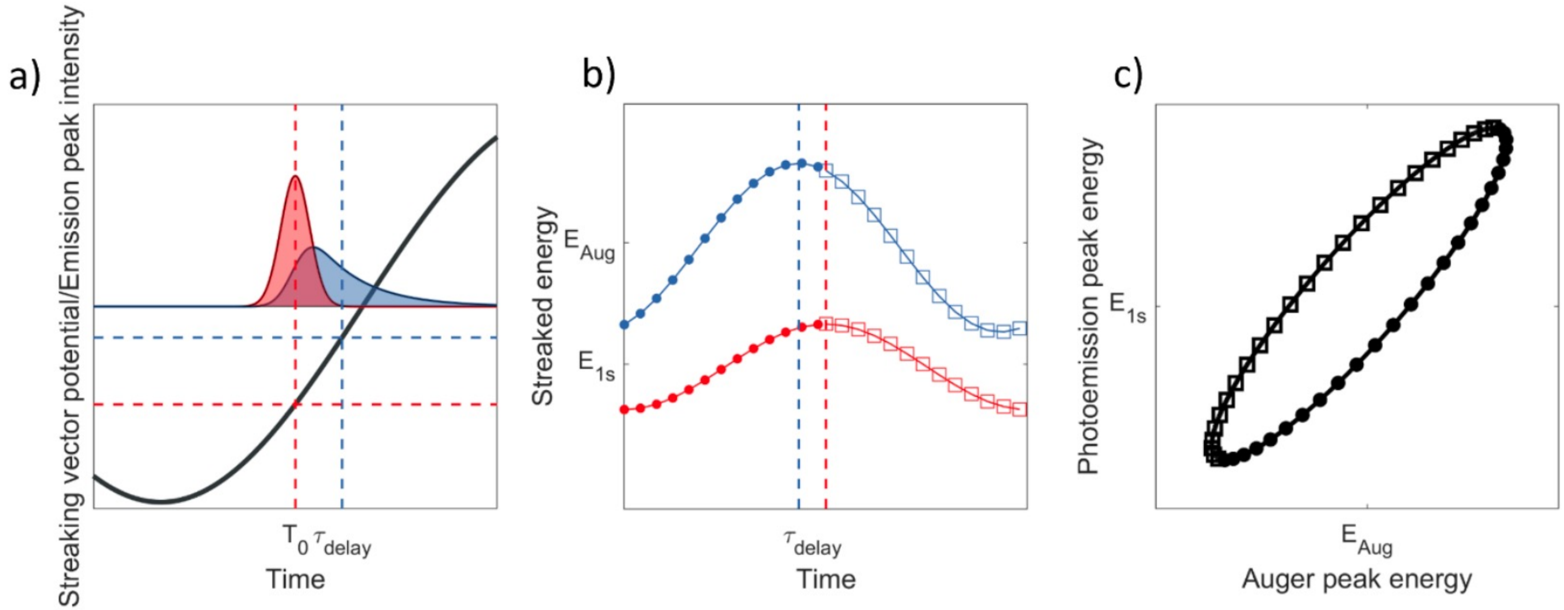
Shift in electron KE due to the IR laser field

$$\Delta E \approx \sin(\emptyset_i)\sqrt{8E_{el}U_p}$$

$\boldsymbol{\phi_i}$ is the phase at the instant in time $\boldsymbol{t_i}$ that the electron with KE $\boldsymbol{E_{el}}$ is bor*n…*

$$U_p = \frac{e^2 E_0^2}{4 m_e \omega_{IR}^2}$$

Photoelectron – Auger electron time delay



Primary
Photoelectron
Emission (red)

Secondary
Auger electron
Emission (blue)

Photo vs Auger
'streaked' energy =
ellipse

What is the time delay between photo and Auger electron emission?

Photoelectron – Auger electron time delay

For an ellipse once can write that:

$$x(\theta) = A\sin(\theta + \phi_A)\,, \qquad y(\theta) = B\sin(\theta).$$

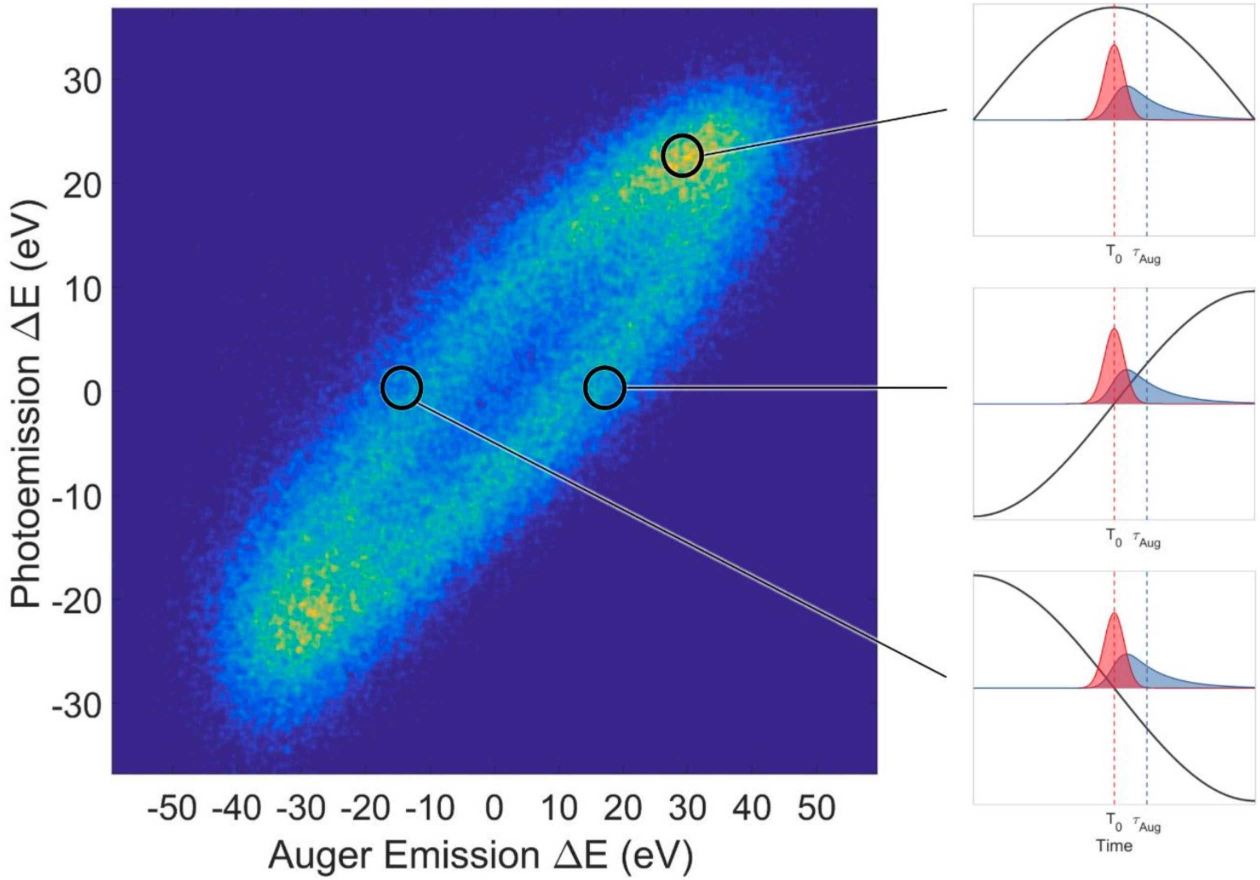
where x = Auger electron energy and y = photoelectron energy.

ϕ_A is the phase advance that occurs between the two instants of electron emission.

$$\phi_A = \sin^{-1}\left(\frac{y_1}{y_2}\right)$$

where y_1 is the ellipse's y-intercept, and y_2 is the maximum value of y.

Photoelectron – Auger electron time delay



$1s^{-1}$ Photoelectron – KLL Auger electron time delay for Ne

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$$2.2\,{}^{+\,0.2}_{-\,0.3}\ \text{fs}$$

Haynes, D.C. *et al.* Clocking Auger electrons. *Nat. Phys.* (2021).
https://doi.org/10.1038/s41567-020-01111-0

Summary Auger delays and next steps.

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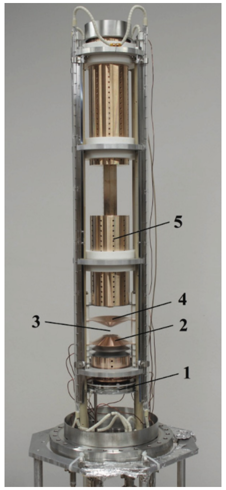
- ‘Self Referenced’ streaking can work with all SASE FELs, delivering a resolution that can reach < 1 femtosecond

- Future experiments with better KE resolution and CEP stabilized IR (MIR) lasers could (in principle) permit timing measurements down to 10-100 attoseconds

- X-rays yield ‘site specificity’ and so in principle it can be applied to atoms in molecules to determine the molecular environmental impact on Auger hole lifetimes/delays

- Combine with IR pump and X-ray probes in molecules to measure non-Born-Oppenhemier electron-nuclear coupling and dynamics (e.g., wavepacket speading at conical intersections in real time).

Next steps–FERMI–Impulsive Molecular Alignment



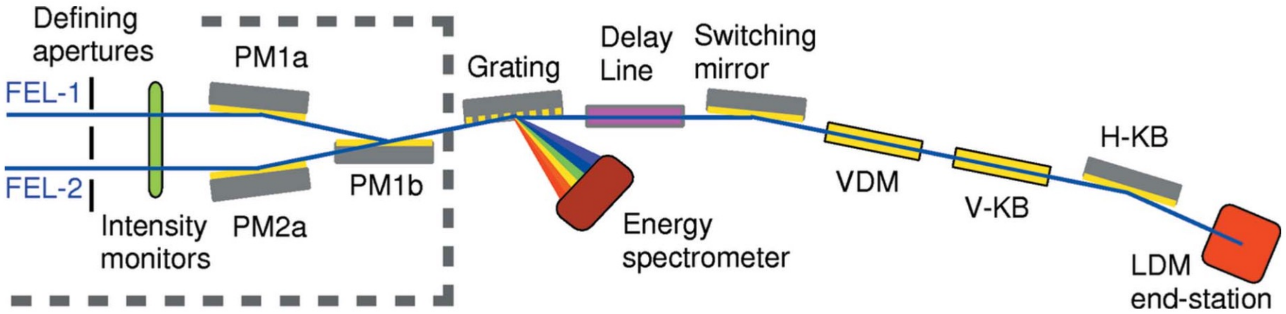


Figure 1
The photon beam transport and diagnostics system of FERMI. The two FEL undulator lines are visible on the left, inside the safety hutch (dashed line). The LDM endstation is in the bottom-right corner. The parameters of the optics are reported in Table 2.

J. Synchrotron Rad. (2015). **22**, 538–543 Cristian Svetina

Next steps–FERMI–Impulsive Molecular Alignment

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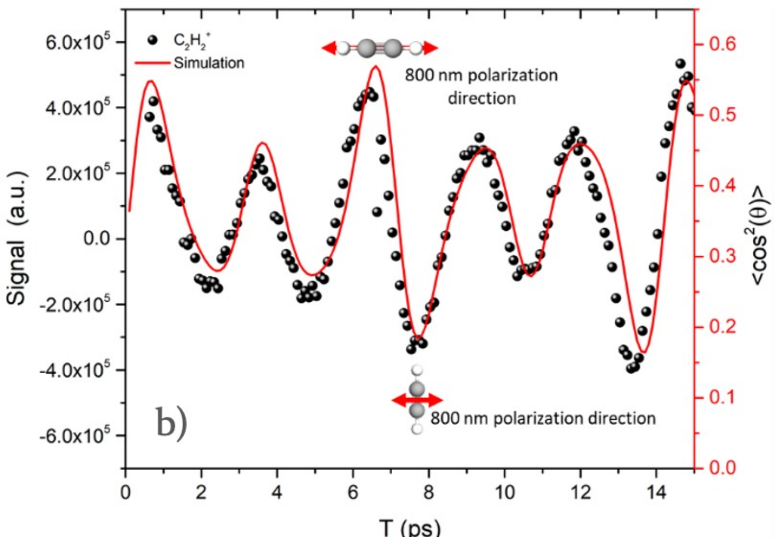

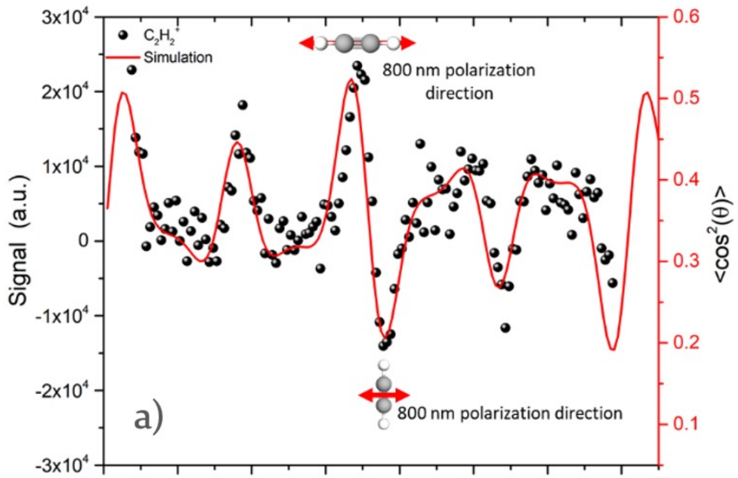

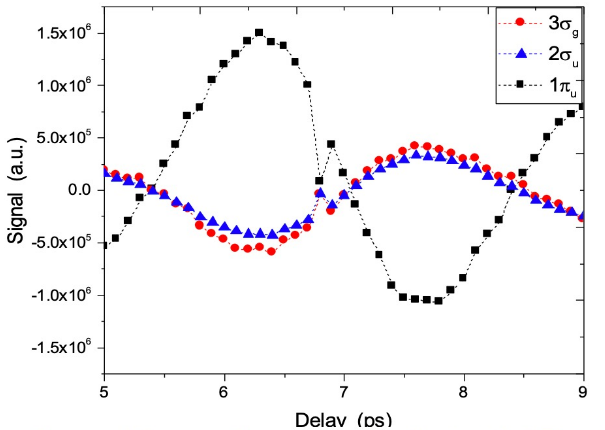

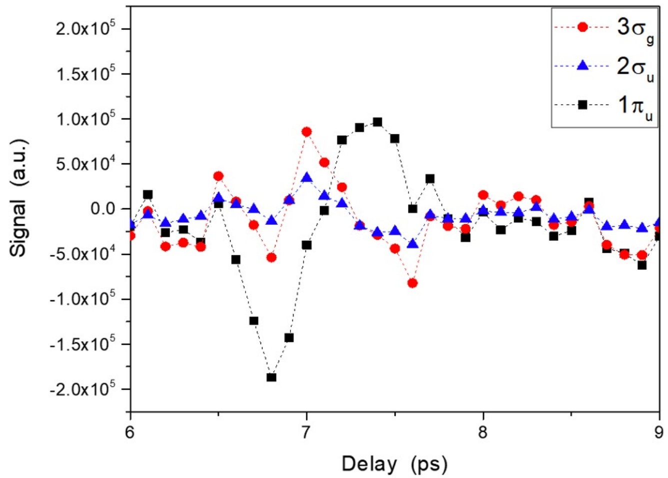

People (Lots of them) & Funders

FLASH - Kr TPDI

Lazaros Varvarezos (DCU), Stefan Düesterer (FLASH-DESY), G Hartmann (Uni Kassel).

LCLS - Auger delays

Dan Haynes (MPSD-Hamburg), Adrian Cavalieri (Uni Bern and SwissFEL), Reini Kienberger (TU-Munich).

FERMI – C_2H_2 alignment

Carlo Callegari/Michele di Fraia/Oksana Plekan/Luca Gianessi/Kevin prince (FERMI), Kiyoshi Ueda (Tohuku)

Congratulations on the first 25 years

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COCHIN UNIVERSITY OF SCIENCE AND TECHNOLOGY

All the very best for the next 25 years

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