Intense XUV and X-ray Free Electron Lasers in AMO Physics

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http://www.ncpst.ie http://www.physics.dcu.ie/~jtc

NCPST EXTATIC Seminar Open University
22 June 2017
sfi inspire Integrated NanoScience Platform for Ireland DCU

DCU Laser Plasma-AMO Physics Group

Laser Plasma/AMO Physics @ NCPST - 6 laboratory areas focussed on pulsed laser matter interactions (spectroscopy/ imaging / particle detection)

Principal Investigators (5): John T. Costello, Eugene T. Kennedy (Emeritus), Lampros Nikolopoulos (T), Jean-Paul Mosnier & Paddy Hayden (SFI SIRG PI)

Current Research students (10): Ben Delaney, Stephen Davitt, Hu Lu, Getasew Wubetu, William Hanks, Muhammed Alli, Sadaf Syedah, Lazaros Varvarezos, Tejaswi Katravulapally & Columb Doherty

Recent Int'l Interns (2012-16): K Nishant/R Tejaswi, (LNMIIT, Jaipur), C Hand, (NUIM), S Reddy/R Namboodiri/A Neettiyath (IIT Madras), R Singh/S Gupta (IIT Kanpur), S Howard (Notre Dame), I-M Carrasco Garcia (Malaga), R. Black (Notre Dame), P Colley (Notre Dame)

Recent PhD Grads (2009-2016): Padraig Hough, Conor McLoughlin, Rick O'Haire, Dave Smith, Vincent Richardson, Tommy Walsh, Jack Connolly, Jiang Xi, Leanne Doughty, Eanna MacCarthy, Colm Fallon, Mossy Kelly, D Middleton, Cathal O'Broin, Brian Sheehy, Saikumar Inguva & Nichola Walsh

Recent Past Postdocs (2012-2015): Satheesh Krishnamurthy (Open Univ. UK), Pat Yeates (Elekta Oncology UK) & Subhash Singh (U. Allahabad), Colm Fallon (IC4), Mossy Kelly (Hull Univ.).

Collaboration @ FLASH-DESY & XFEL

XFEL: F. Babies, S. Bakhtiarzadeh, A. Beckmann, J. Buck, A de Fanis, L. Glaser, P. Gessler, M. Ilchen, T. Mazza, A. J. Rafipoor, H Sotoudi & M. Meyer

FERMI: P. O'Keefe

DESY (Hamburg): S. Düsterer, G. Hartmann, F Scholz, J Seltmann & J. Viefhaus

Univ Hamburg: N. Gerken & M. Martins

Crete: E. T. Karamatskos, D. Markellos, P. Lambropoulos (T)

Moscow State University : N. M. Kabachnik

Kurchatov Inst., Moscow: V. L. Nosik

Univ Basque Country: A K Kazansky

DCU: T. J. Kelly(Now Hull), N. Walsh & J. T. Costello

Thanks to AG Photon & AG Machine at FLASH-Hamburg

Collaboration @ FLASH-DESY & FERMI-ELETTRA

XFEL: P. Radcliffe & M. Meyer

Paris (UPMC): R. Taieb (T) & A. Maquet (T)

FERMI: P. O'Keefe, L. Avaldi & K. Prince

DESY (Hamburg): K. Tiedke, S. Düsterer, W. Li, A. Sorokin & P. Juranić,J. Feldhaus

Orsay: D. Cubaynes

Queen's University Belfast: C. L. S. Lewis

Moscow State University : A. N. Grum-Grzhimailo, E. V. Gryzlova, S. I. Strakhova

Crete: P. Lambropoulos (T)

Oulu/GSI: S. Fritzsche (T)

DCU: T. J. Kelly (Hull), N. Walsh, E. T. Kennedy, L Nikolopoulos & J. T. Costello

Thanks to AG Photon & AG Machine at Hamburg and Trieste

Collaboration @ LCLS X-ray FEL (SLAC)

DESY (CFEL): I. Grguras, M Hoffmann & A. Cavalieri

DESY (FLASH): S. Düsterer & J. Feldhaus

DCU: T. J. Kelly (Hull), E. Kennedy, V. Richardson, L. Nikolopoulos (T) & J. T. Costello

MPQ/TU-Munich: A. Maier, W. Helml, W. Schweinberger & R. Kienberger

Ohio (OSU): C. Roedig, G. Doumy* & L. DiMauro

Tohuku University: K. Ueda

Hiroshima University: S. Wada

SLAC: R. Coffee, J. Hastings, C Boestedt, J. Bozek et al.

XFEL Gmbh: P. Radcliffe, T. Tschenscher & M. Meyer

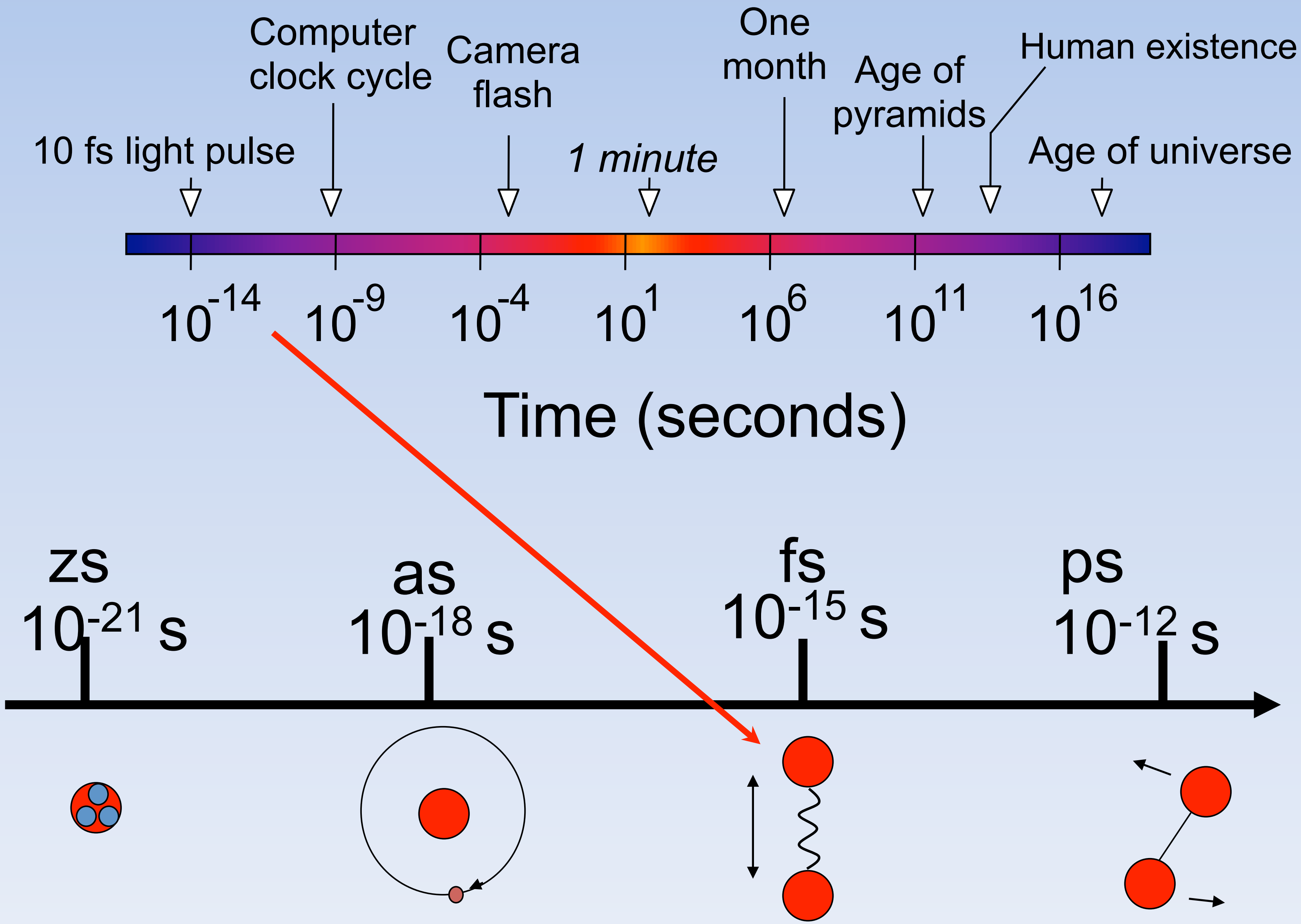
Moscow State University: N. Kabachnik

Thanks to Paul Emma et al.

*Now at Argonne.

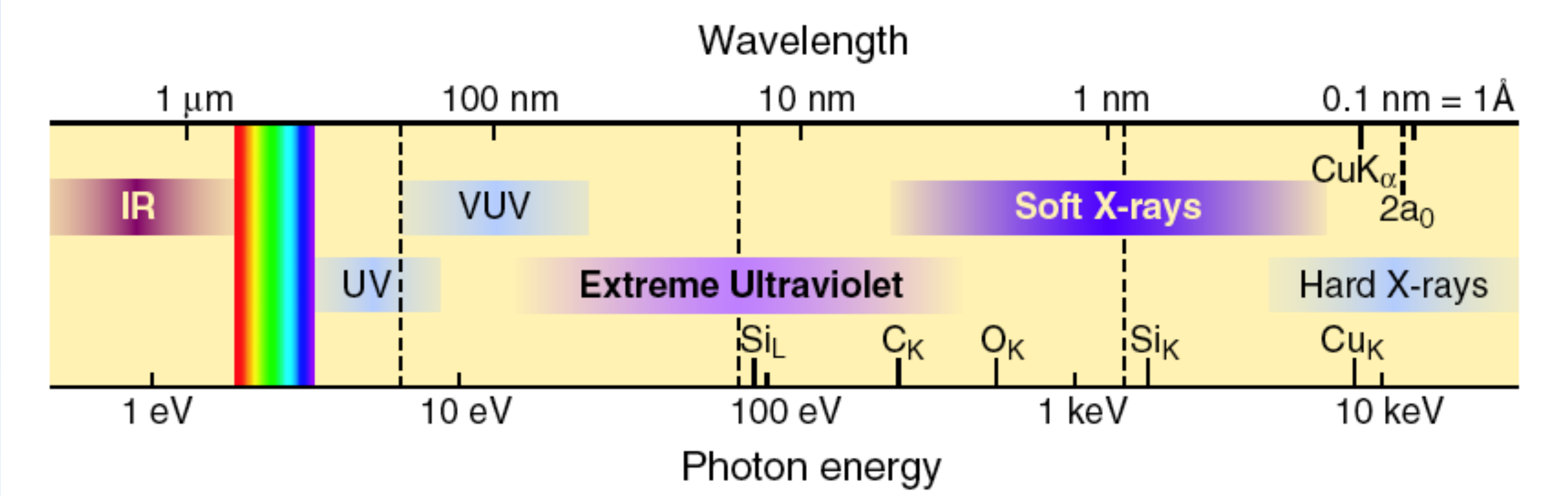
Some members of the LCLS collaboration

TIMESCALES - HOW FAST IS FAST ?



X-ray – How X-ray is X-ray ?

Spectral Range: IR to the X-ray



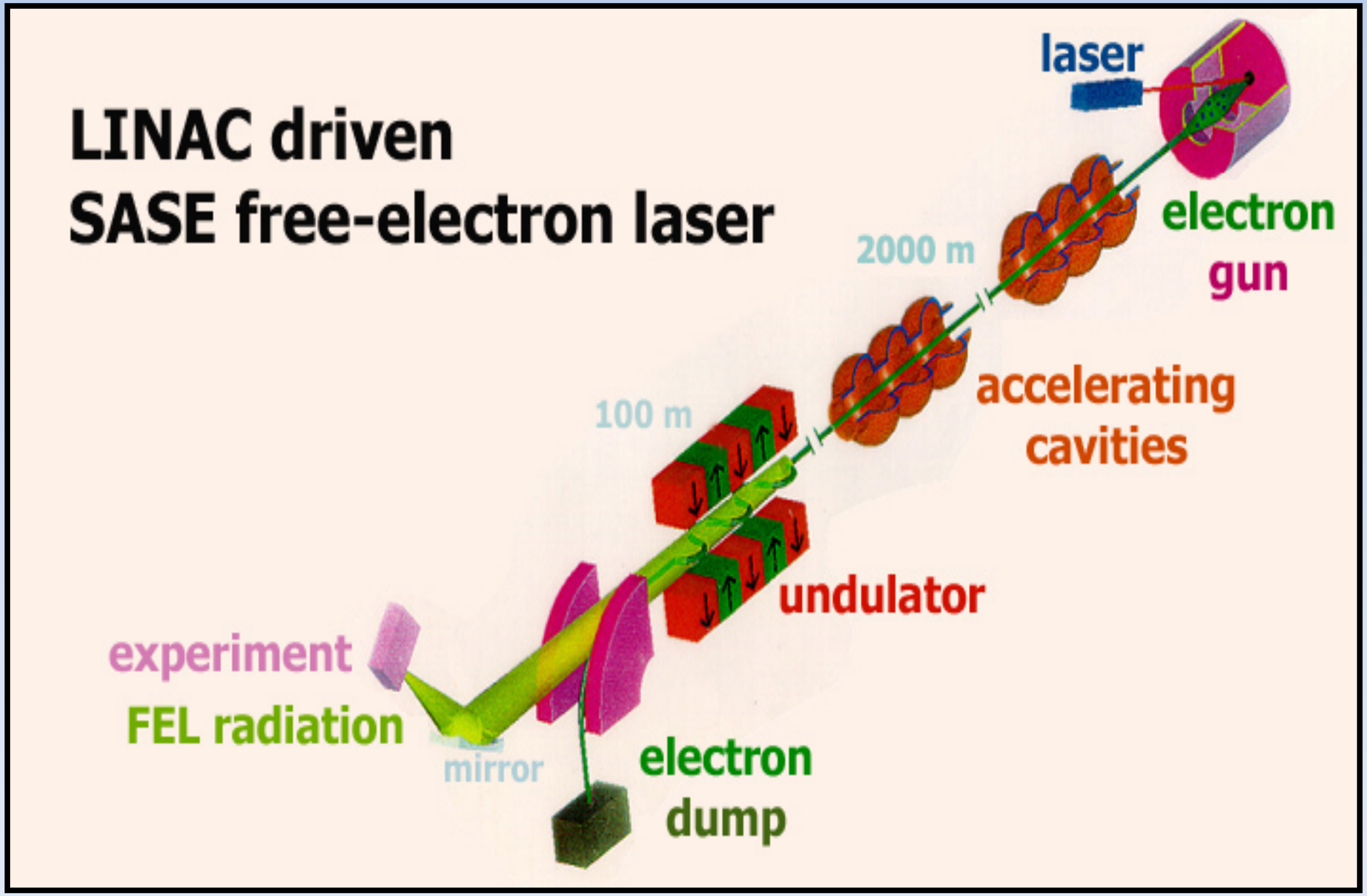
Graphic: Courtesy, Prof. David Attwood (Berkeley)

What do we want in an X-ray laser ?

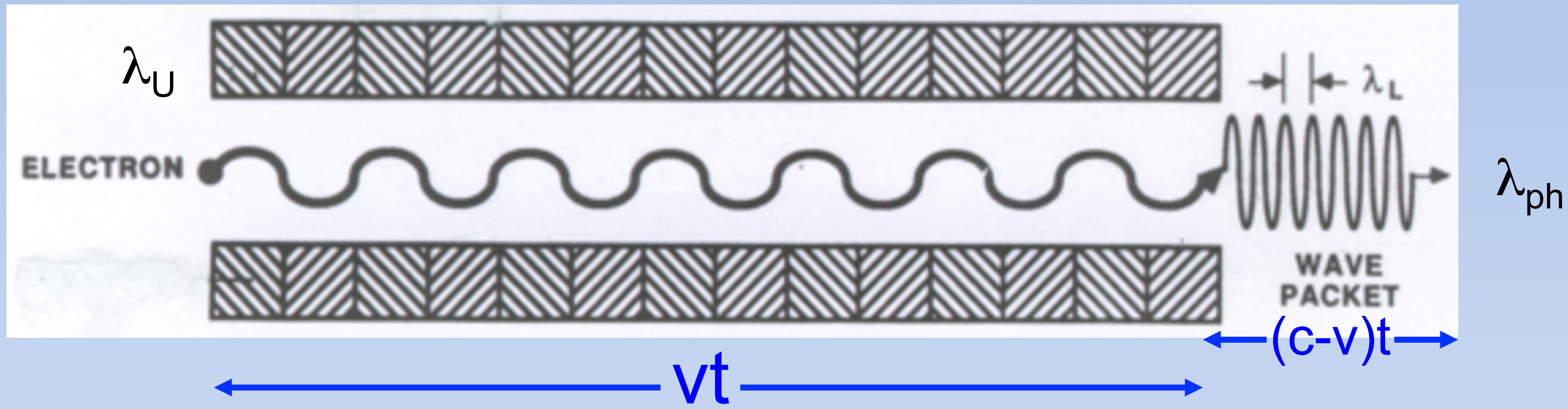
The Holy Grail is an X-ray laser with variable pulse duration on the femtosecond to attosecond timescales with tunable wavelength, variable polarisation and high energy per pulse (few 100 μJ to few 10 mJ)…..

X-ray Free Electron Lasers (FEL)

Main Components of an X-ray FEL



SASE-FEL, Fundamental Principle



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

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

SASE FEL Requirements:
Mono-energetic relativistic electron bunch of very high charge density/low emittance
Ultra-precise long magnetic undulator in perfect alignment

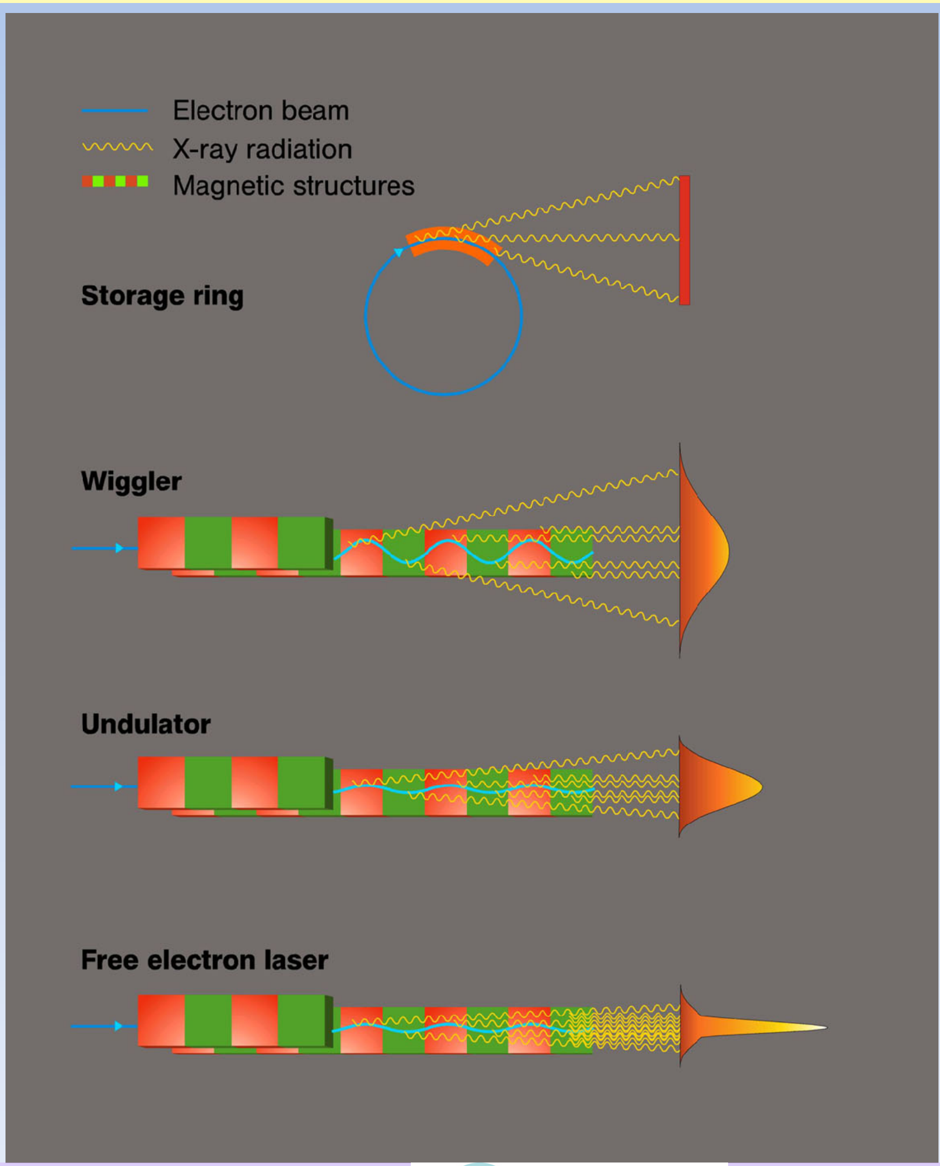
Key concepts

1. Electrons move in slalom-like fashion
2. Electrons emit em waves - forward peaked
3. EM interactions with electrons cause micro-bunching
4. Bunches form 1 λ apart
5. Micro-bunches emit coherently
6. Light grows exponentially with distance along the undulator

Micro-bunching
λ apart

$$\lambda_X = \lambda_s = \lambda_u(1+K^2/2)/2\gamma^2$$

Free Electron Radiation Sources



Josef Feldhaus, DESY, Hamburg

Bending magnet: broad band continuum emission

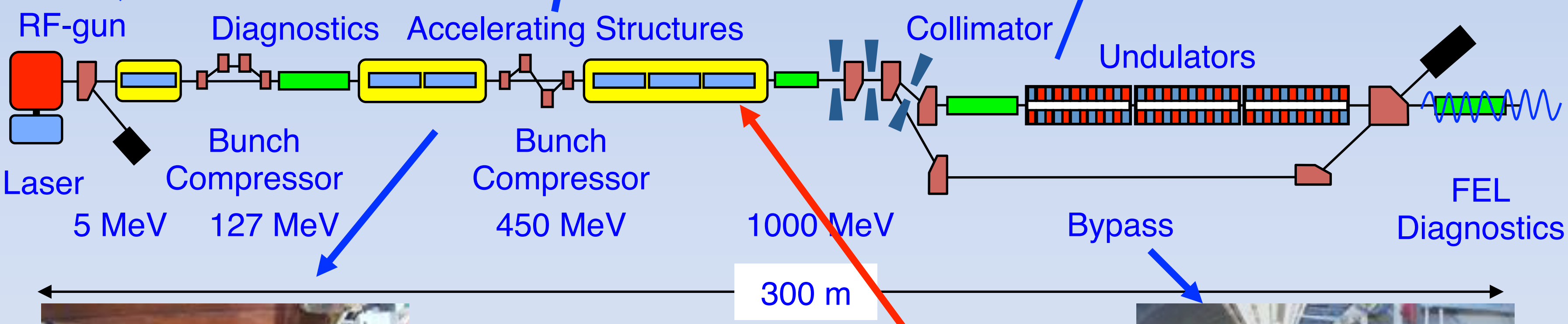
Wiggler:
$I \propto N_W$ x bending magnet

Undulator:
$I \propto N_U^2$ x bending magnet

FEL: $\propto N_U^2 x N_e$xbending magnet

N_U , N_W = # magnetic periods
N_e = # electrons in a bunch

X-ray Free Electron Lasers (FEL)

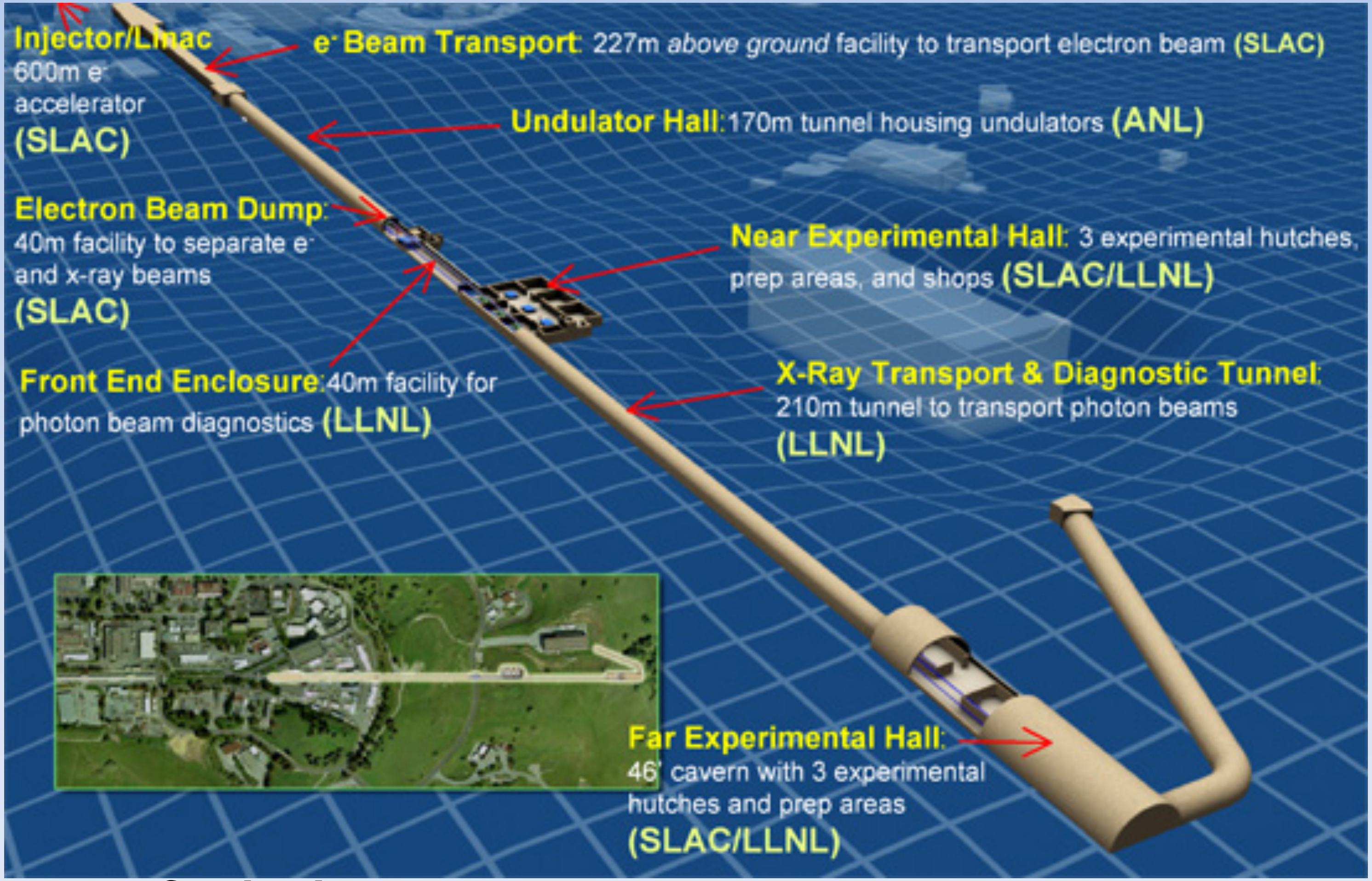


- LINAC Energy : ~ 1 GeV
  ~ 4 – 60 nm

FLASH - Operation & Physical Layout

X-ray Free Electron Lasers (FEL)

LCLS Overview and Specifications



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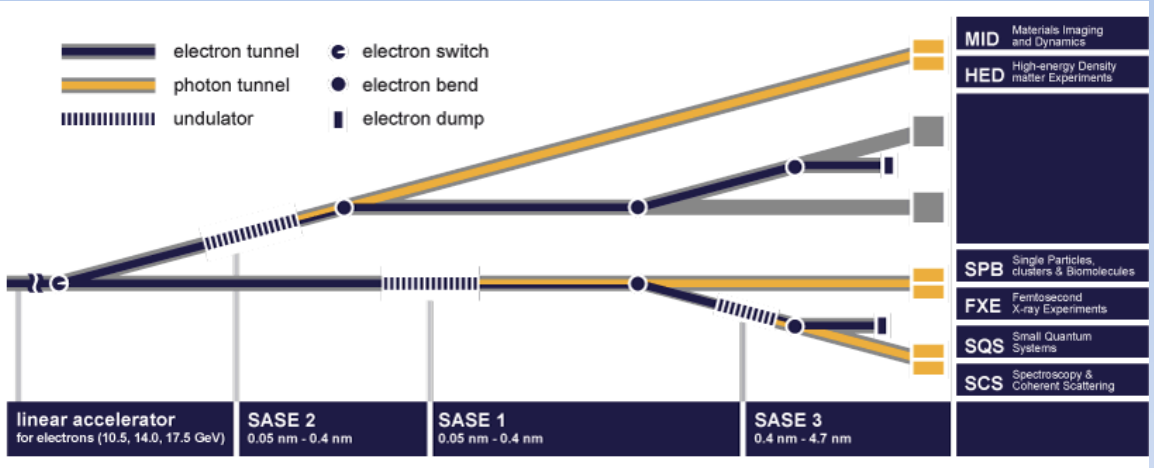
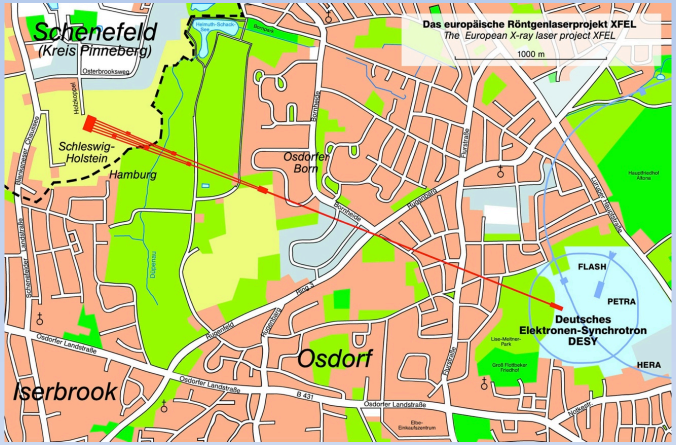
X-ray Free Electron Lasers (FEL)

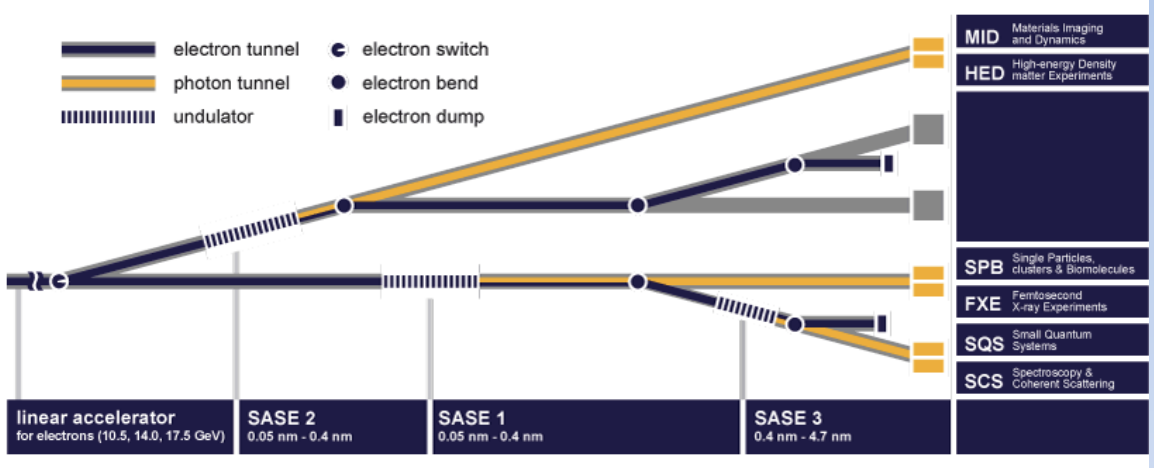
XFEL – Under Construction…… 2017

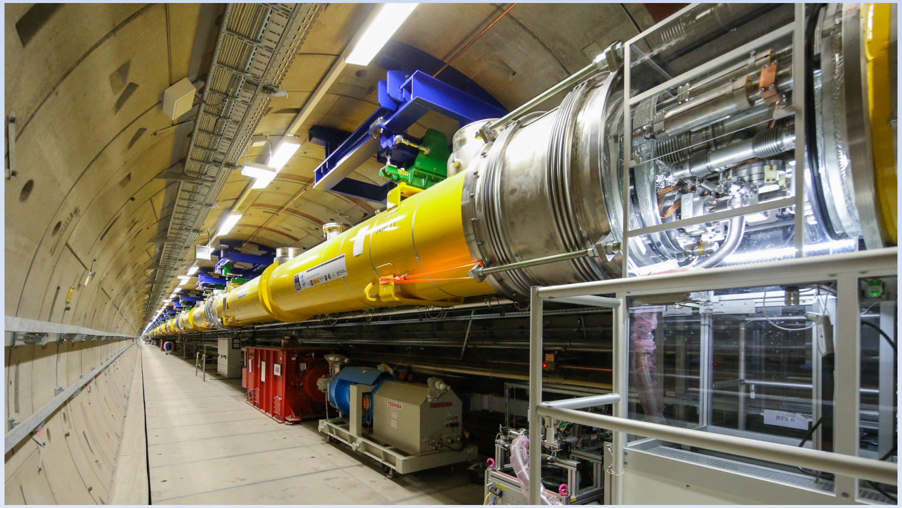




X-ray Free Electron Lasers (FEL)

XFEL – Under Construction…… 2017





USPs of XUV & X-ray FELs (XFELs)?

- *High flux per pulse – typ. 10^{13} photons/pulse*
- *Tunable pulsewidth – from 1 to few 100 fs*
- *Ergo high peak intensity – up to few 10^{20} W.cm^{-2} possible*
- *Seeded and unseeded modes now possible*
- *Unseeded bandwidth – 0.2 – 1.0%*
- *Seeded bandwidth – 0.005% (typ.) / $\lambda/\Delta\lambda \geq 10^4$*
- *Synchronisation to optical fs lasers relatively easy*
- *EUV/EUV and X-ray/X-ray pump-probe possible*

Technology Now…..

So the Holy Grail is now largely realised as the SASE EUV and X-ray FELs at SLAC-Stanford, SCSS & SACLA-RIKEN, FLASH-DESY (+future European XFEL), FERMI@ELETTRA-Trieste, SwissFEL-PSI, Pohang, Shanghai, Dalian, etc……..

Very recently [2012] seeding of LCLS, SCSS and FERMI have resulted in transform limited pulses with $\Delta\lambda/\lambda$ values of ~ 10^{-4}

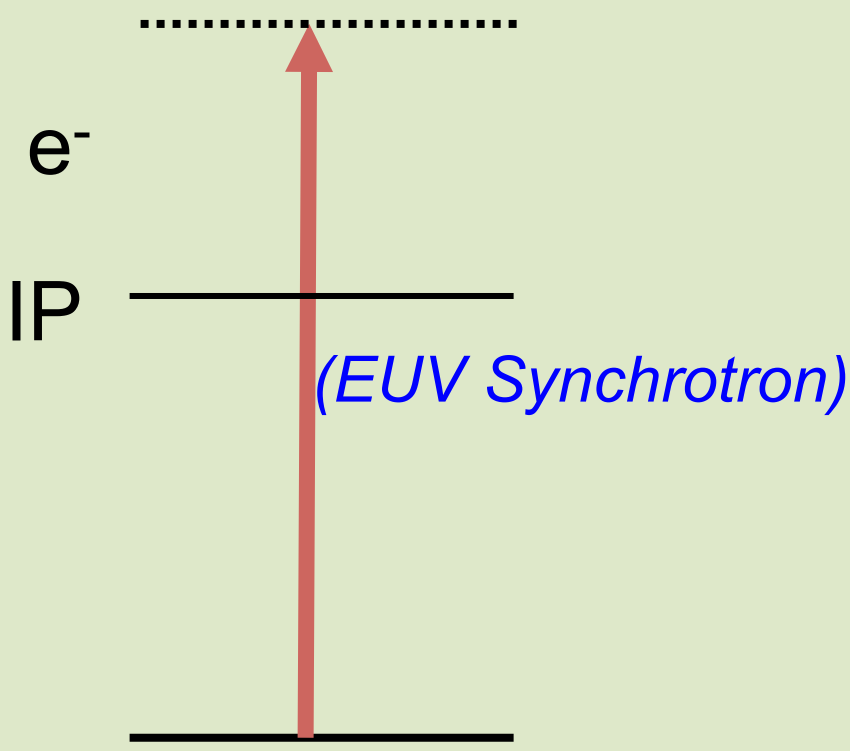
Ionization in Intense Fields

1. **Rudiments of ionization processes in intense laser fields**
2. Photoionization experimental setups (FLASH & DESY)
3. One colour – two photon ionization
4. Two colour Ionization – physics and characterisation
5. Some perspectives

The Atomic Photoelectric Effect

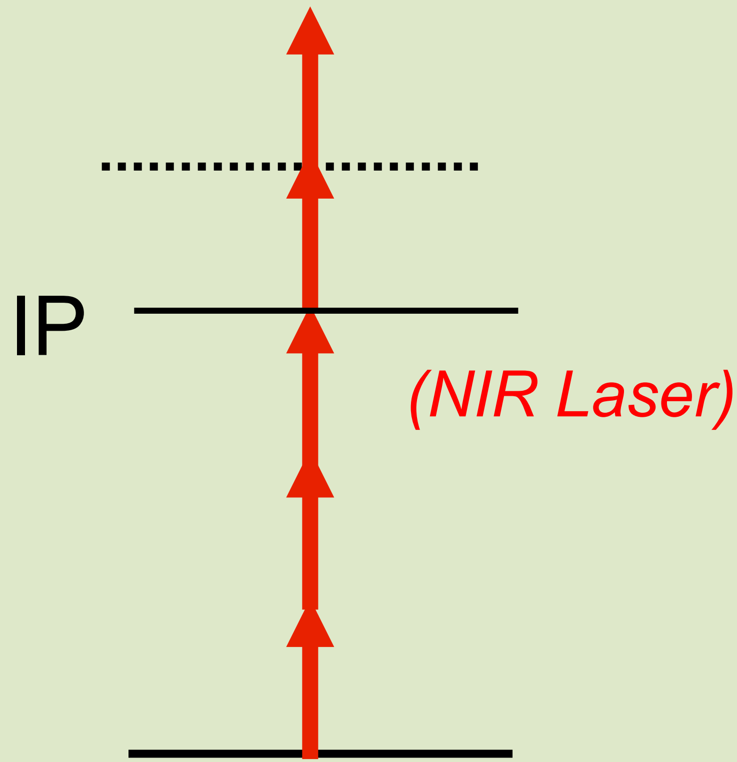
a) Single Photon Ionization (SPI)

$KE(e^-) = h\nu_{EUV} - IP$

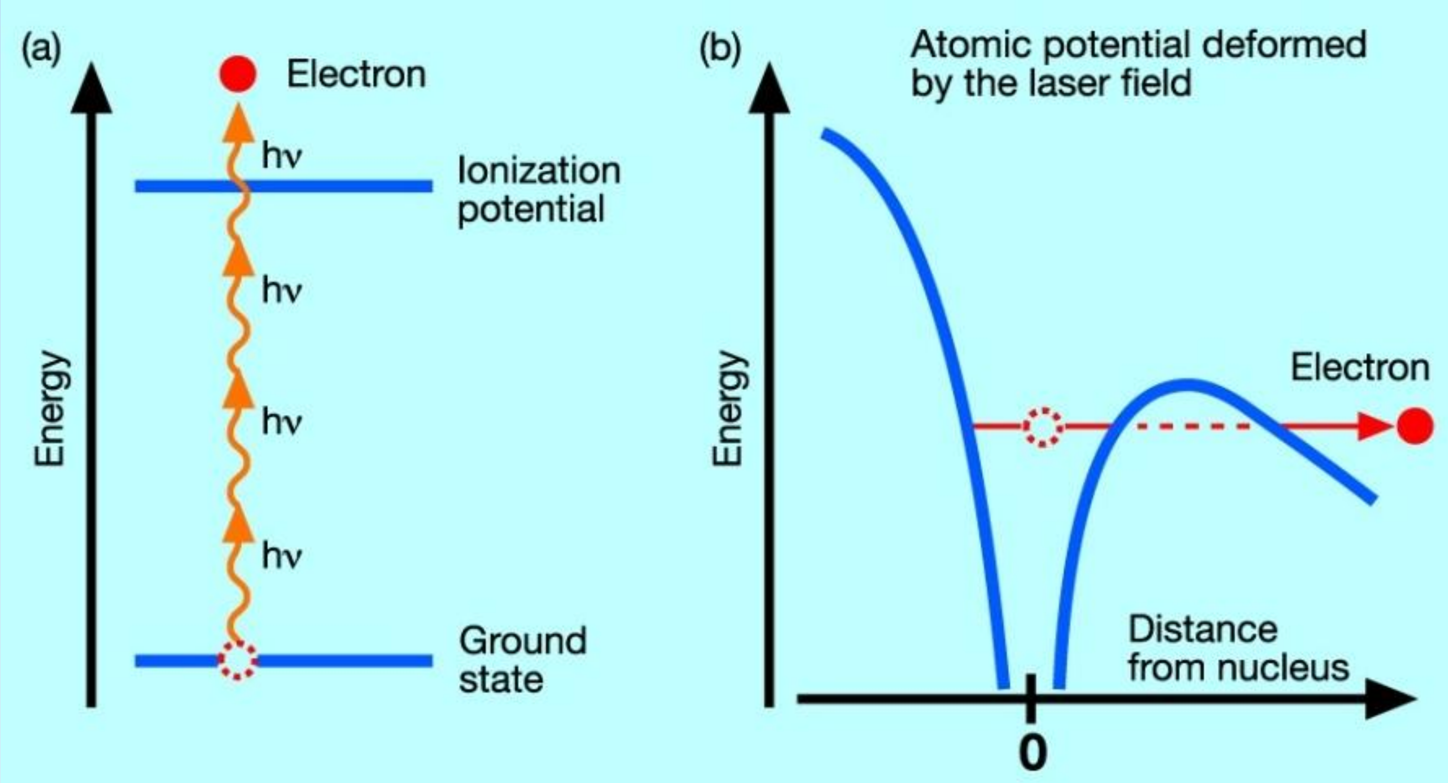


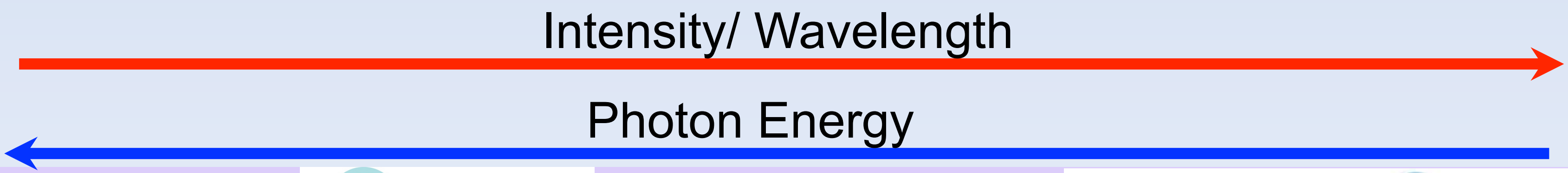
b) Multi Photon Ionization (MPI)

$KE(e^-) = nh\nu_{NIR} - IP$



What happens as the laser intensity (field strength) grows ?





How can you determine in which regime the interaction resides ?

$$\gamma = \sqrt{\frac{IP}{2U_p}}$$

Keldysh Parameter

IP = Ionization Potential
Up = Ponderomotive Pot.

$$U_P = 9.3\times10^{-14} I\left(Wcm^{-2}\right) \lambda^2(\mu m) \quad eV$$

*L V Keldysh, Sov.Phys-JETP 20 1307 (1965)

Keldysh - Ionization Regime

Multiphoton Ionization	Tunnel Ionization	Field Ionization
$\gamma >> 1$	$\gamma \sim 2$	$\gamma << 1$

Example: ***Helium*** in intense laser fields

For Ti-sapphire laser: 800 nm, 10^{15} Wcm^{-2}, γ ~0.45 (TI/FI regime)

For an EUV laser: 8 nm, 10^{15} Wcm^{-2}, γ ~45 (MPI regime)

So for EUV lasers, multi-photon ionization is the primary processs and will involve *few photons* and *potentially few electrons*

USPs of XUV & XFELs in AMO Physics ?

- *Ultra-dilute* targets
- *Photo*-processes with *ultralow cross-sections*
- *Pump and probe* experiments (EUV + EUV or EUV + Opt.)
- *Single shot* measurements
- *Few-photon* single and multiple *ionization processes*

NB1: Makes *inner-shell* electrons key actors in non-linear processes for the first time

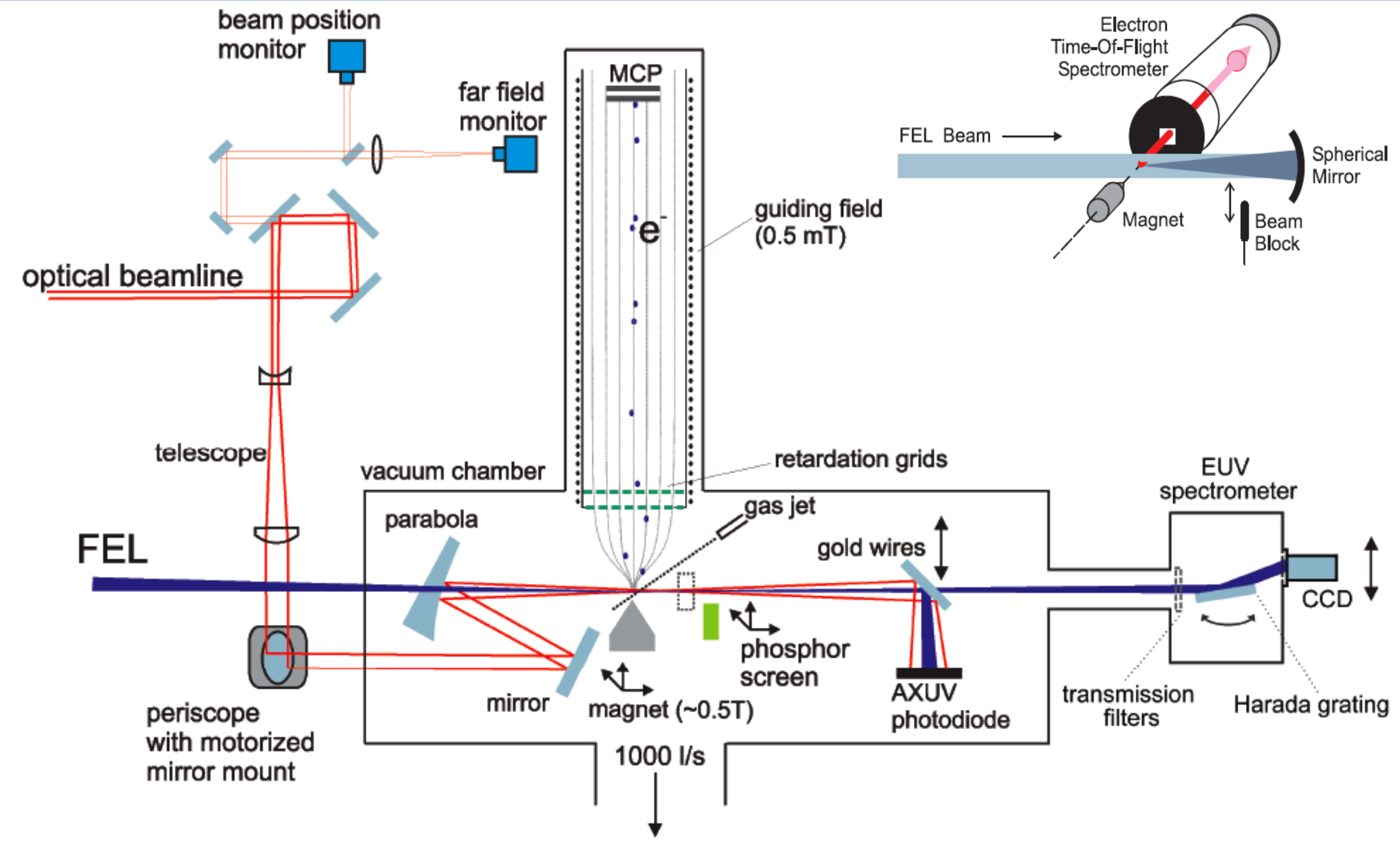
NB2: Re-asserts *primacy of the photon* over field effects !

Experimental Setups (DESY & SLAC)

1. Rudiments of ionization processes in intense laser fields
2. **Photoionization experimental setups (FLASH & LCLS)**
3. One colour – two photon ionization
4. Two colour Ionization
5. Some perspectives

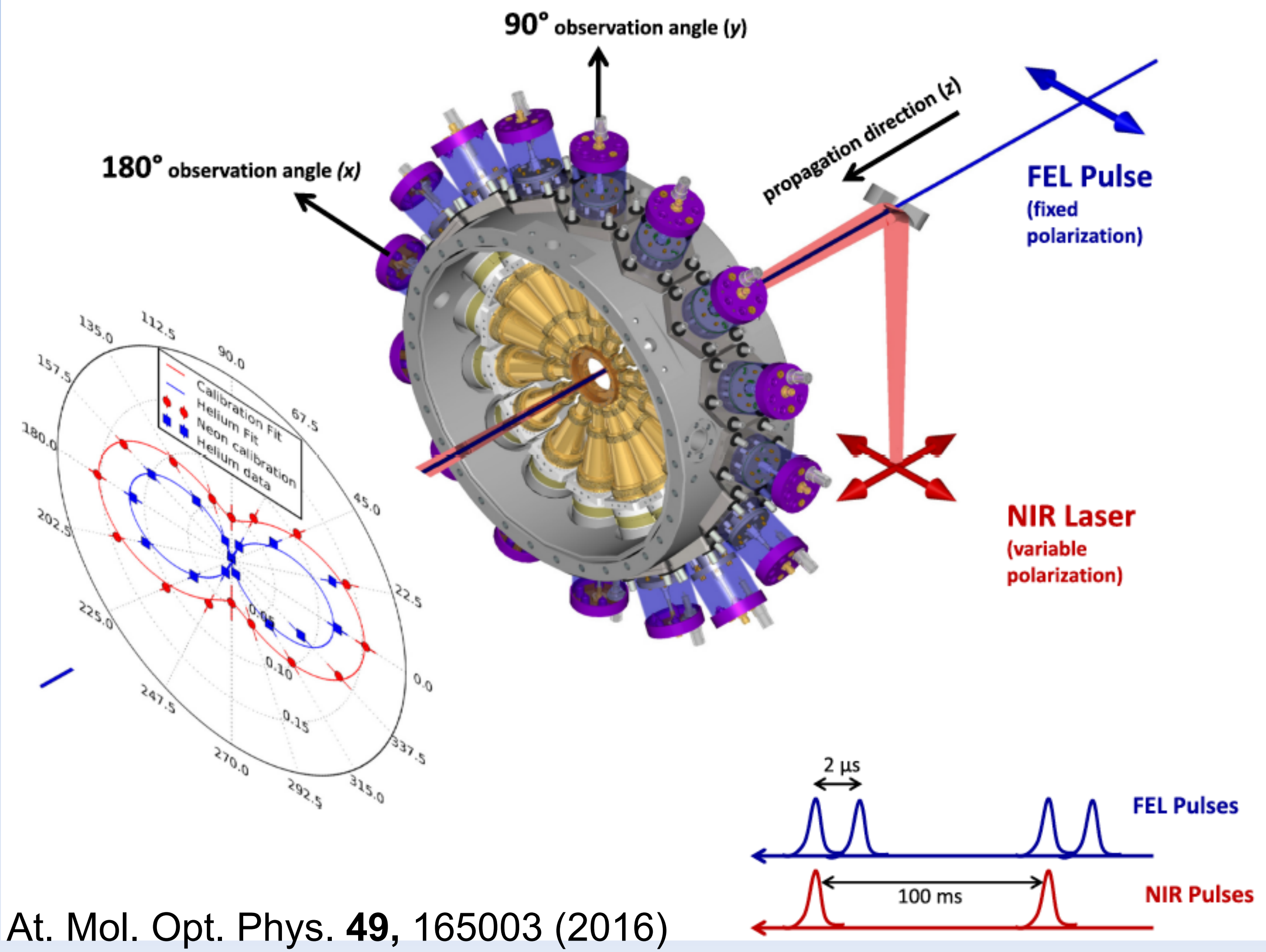
Photoelectron Spectroscopy @ FLASH

Experimental Layout at FLASH - (EU-RTD)



Photoelectron Spectroscopy @ FLASH

ARPES Experimental Layout at FLASH - (Viefhaus et al.)

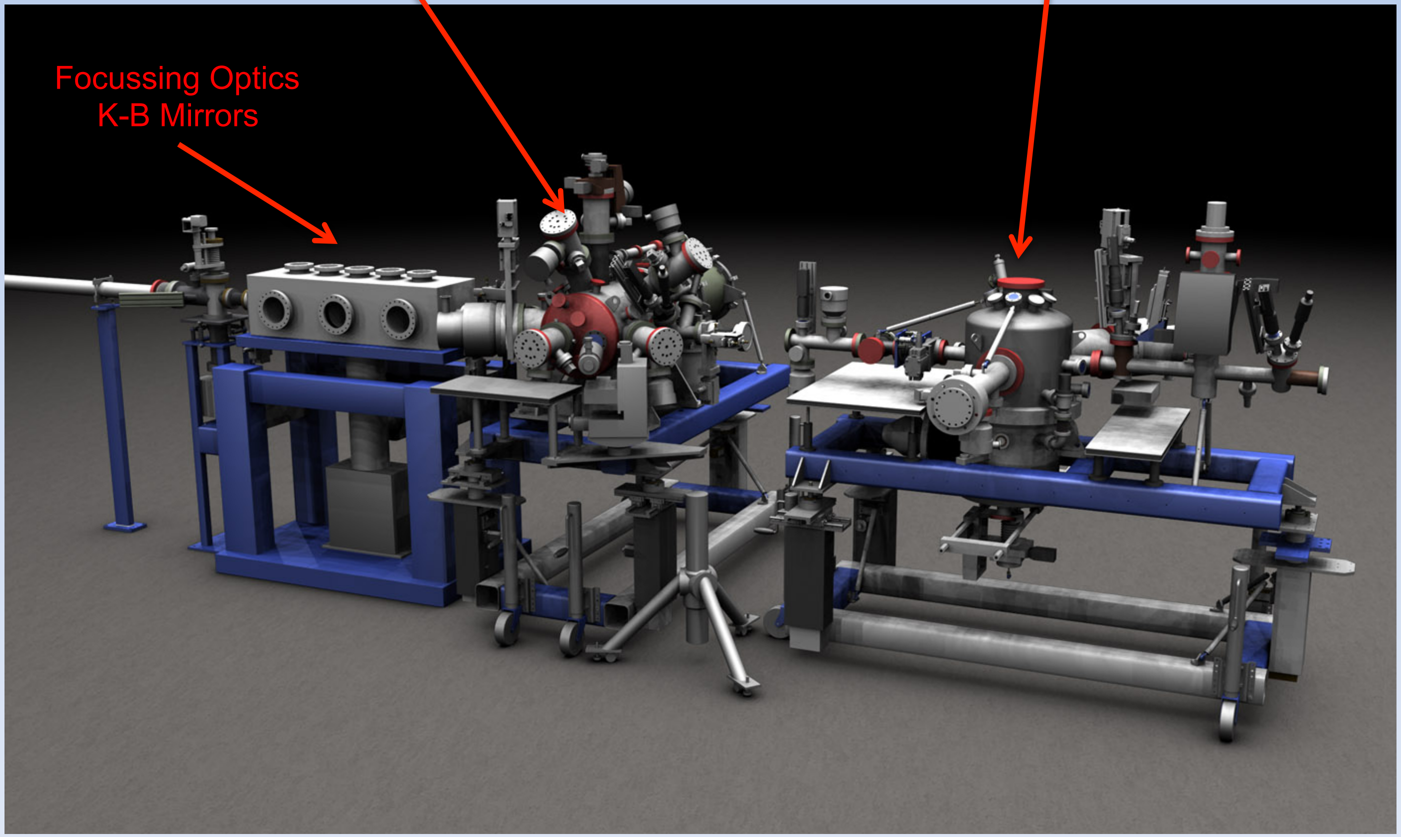


J. Phys. B: At. Mol. Opt. Phys. **49,** 165003 (2016)

AMO PES Chamber at LCLS

Rendered Image:

High Field Chamber (AR-ETOF) and Diagnostics (MBES) Chamber



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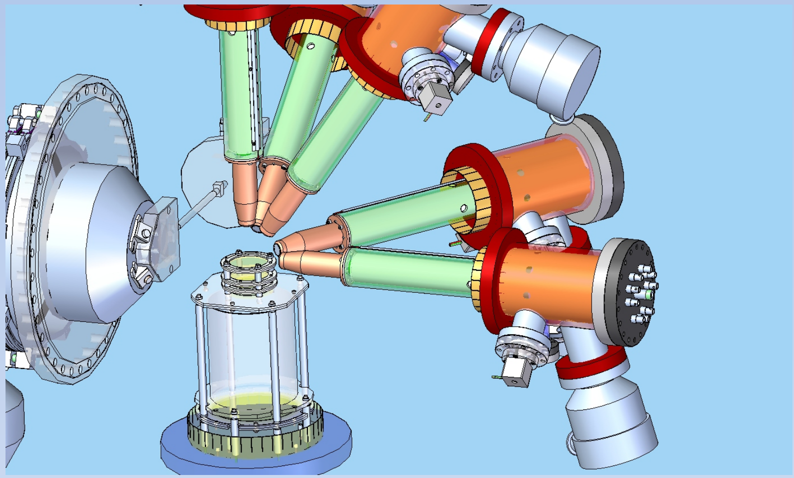
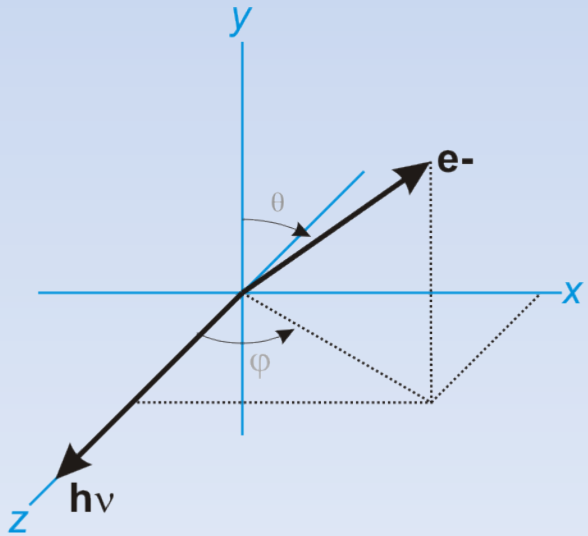
AMO Chamber and Specifications

High Field Chamber (AR-eTOF)

1. Based on a successful design used by the Denis Lindle (RIP) group at ALS – designed for up to 5keV electrons
2. Transmission flat for E_{kin}> 20 eV
3. E/ΔE up to 5,000



•	θ	φ	comment
1	0º	90º	Along y-axis
2	35.3º	90º	Magic angle in xy dipole plane
3	90º	90º	Along x-axis
4	54.7º	0º	Non-dipole
5	90º	35.3º	Non-dipole



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Two Photon Ionization (TPI) of Xe and Kr atoms in an Intense Field

1. Rudiments of ionization processes in intense laser fields
2. Photoionization experimental setups (FLASH & DESY)
3. **One colour - two photon ionization**
4. Two colour Ionization
5. Some perspectives

Non-linear processes in the EUV & X-ray

Question. What is the simplest experiment you can carry out in non-linear optics ? Answer. Either two-photon absorption (TPA) or second harmonic generation (SHG)……..

VOLUME 7, NUMBER 6 PHYSICAL REVIEW LETTERS SEPTEMBER 15, 1961

TWO-PHOTON EXCITATION IN $CaF_2:Eu^{2+}$

W. Kaiser and C. G. B. Garrett
Bell Telephone Laboratories, Murray Hill, New Jersey

(Received August 28, 1961)

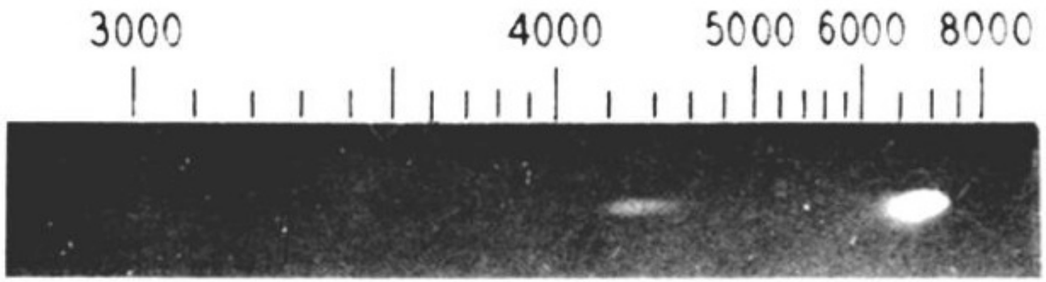
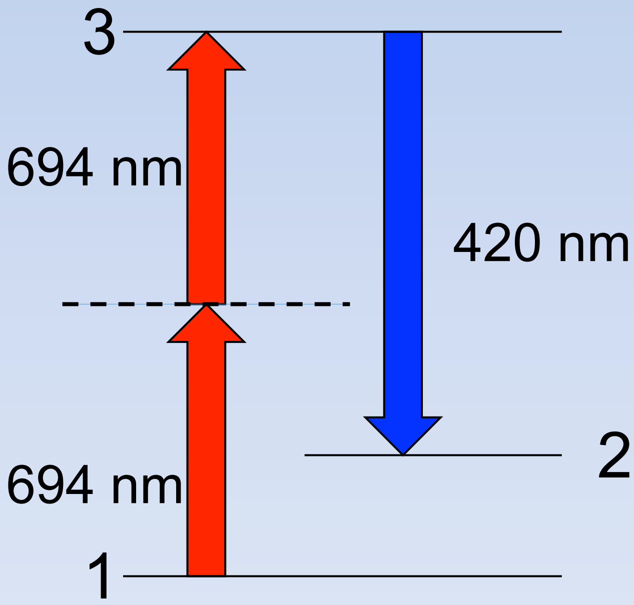
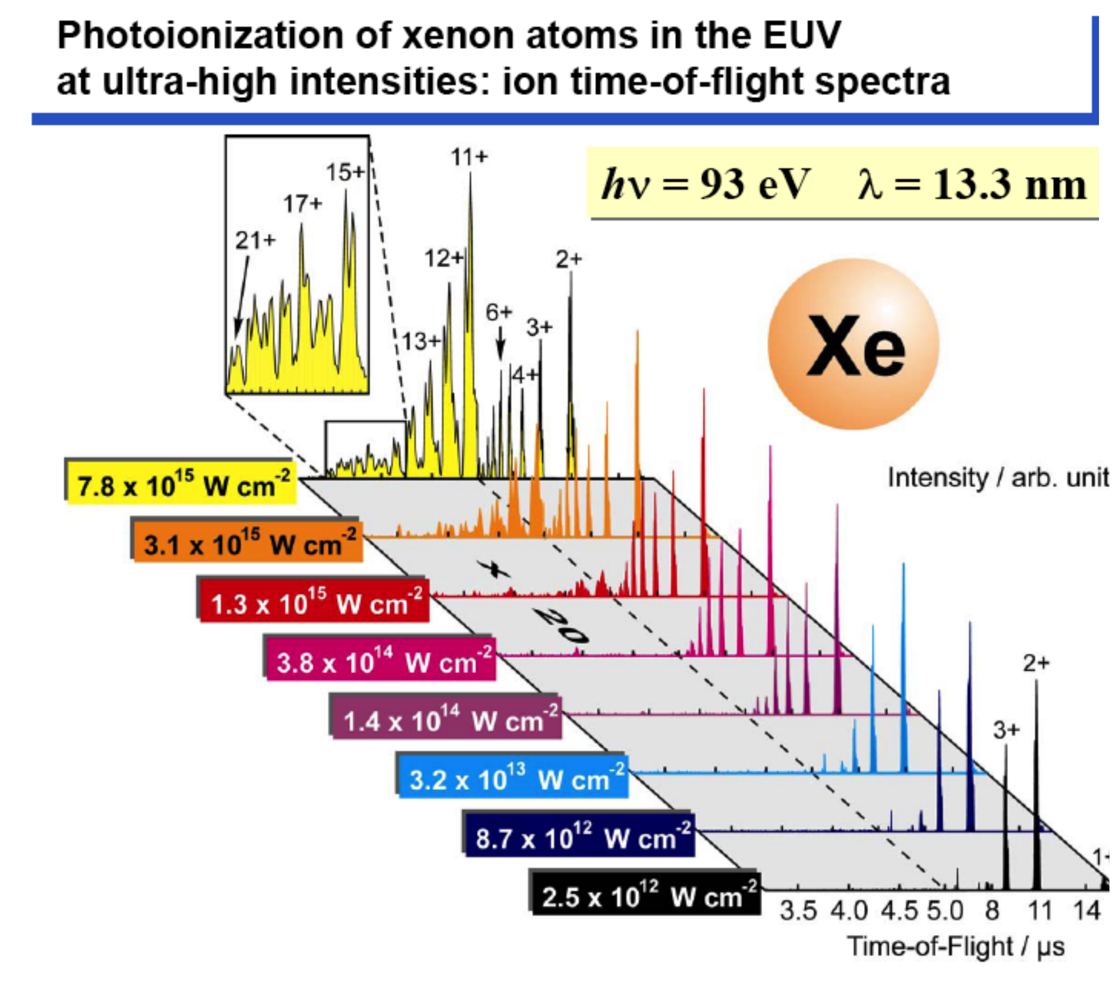


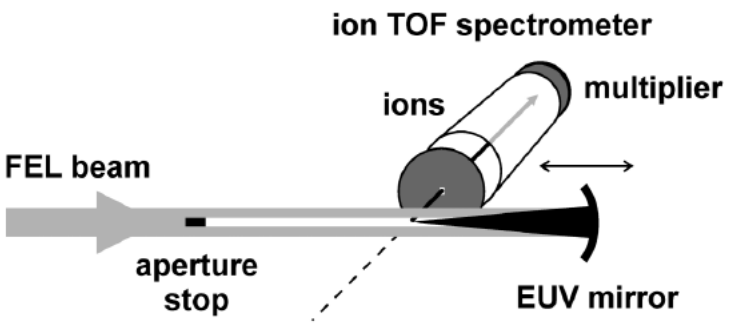
FIG. 1. Positive of photographic plate, indicating the blue emission of a $CaF_2:Eu^{2+}$ crystal under strong illumination with $\lambda_\gamma = 6943$ A.

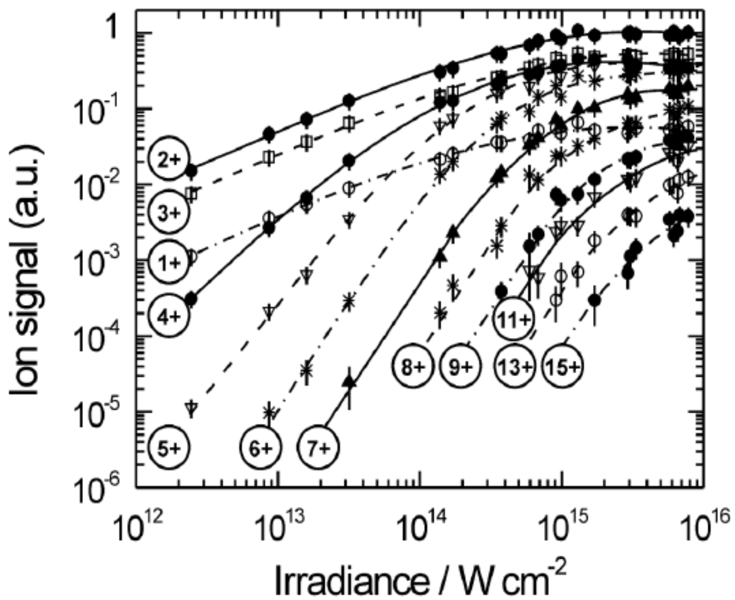


Motivation - Xe TPI in intense EUV fields

Sorokin, Richter et al., PTB, PRL 2007 – Ion Spectroscopy !!



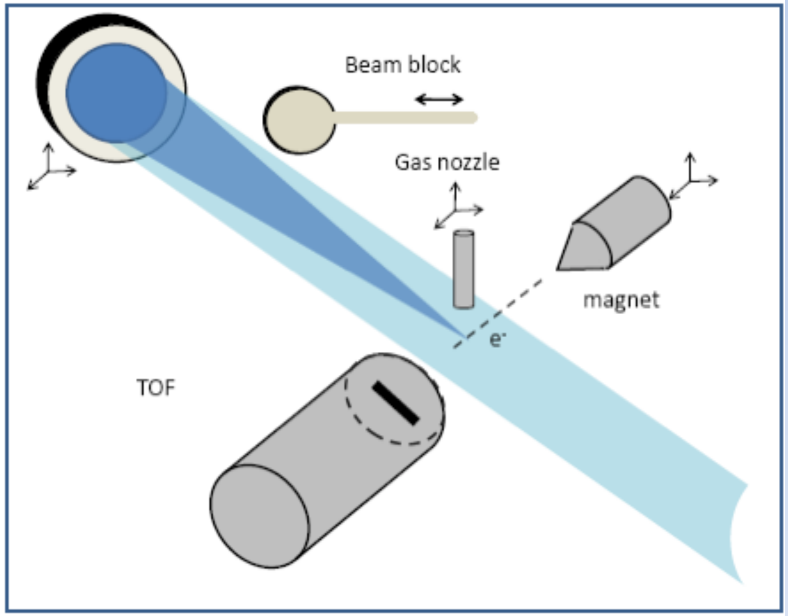




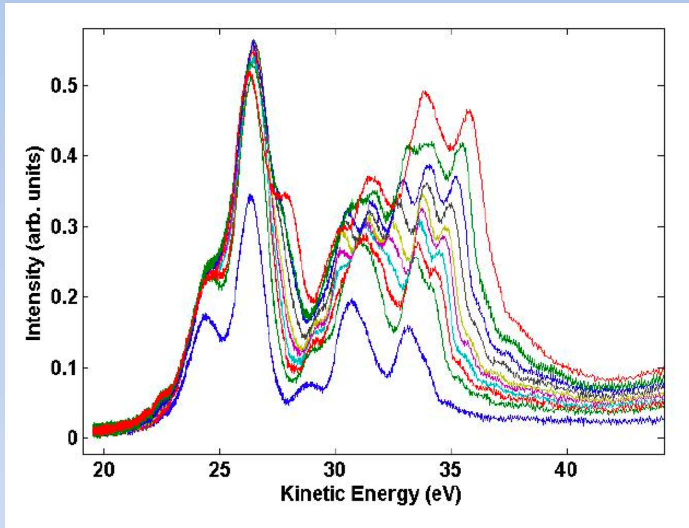
Xe + hν (93 eV) - $Xe^{+}(4d^{-1})$ + e^{-} (~25 eV)

FEL only. hν ~ 93 eV

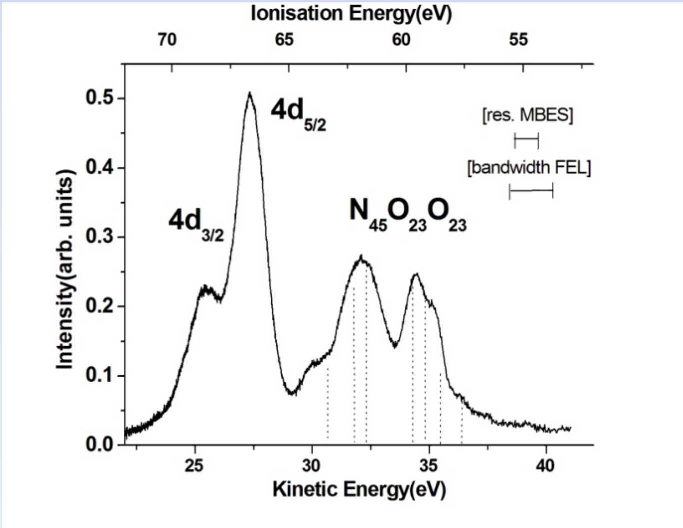
$Xe + h\nu \rightarrow Xe^{n+} + e^{-}$



Replace Ion TOF by MBES – photoelectron spectroscopy



Intensity scaling...

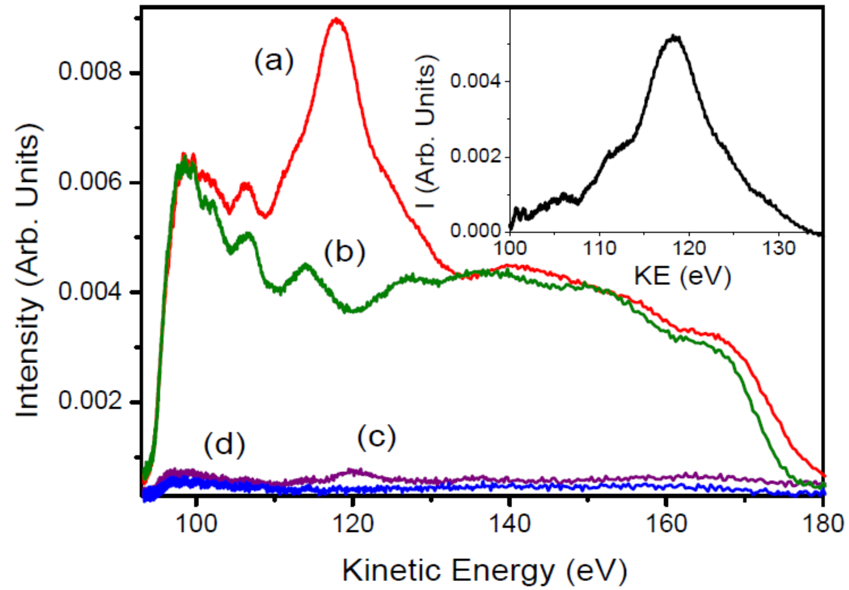


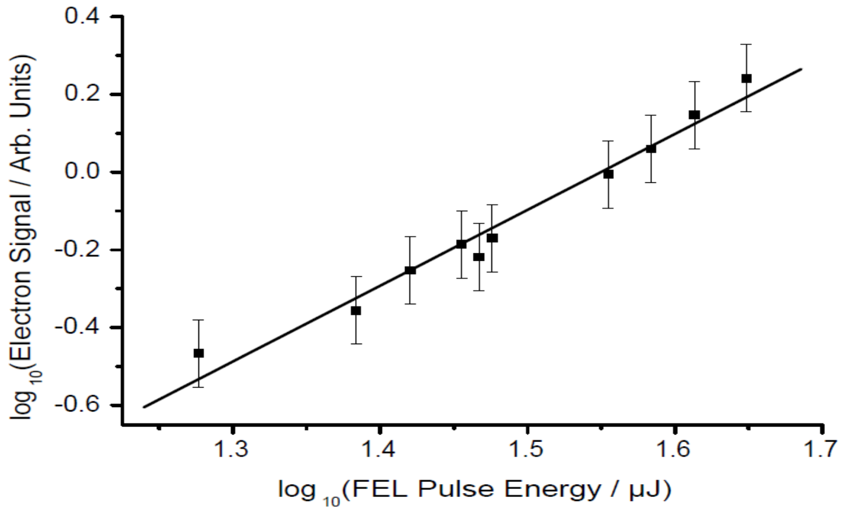
Weakest field…

$Xe + 2h\nu$ (93 eV) - $Xe^{+}(4d^{-1}) + e^{-}$ (~ 118 eV)

Now ramp up the intensity to > 10^{15} W.cm^{-2}…………..

- Using MBES, first evidence of two photon ***inner*** shell ionisation, (in this case) of 4d electron –
  $Xe + 2h\nu \rightarrow Xe^{+}\ 4d^{9} + e^{-}$
- ‘Retardation field’ applied to suppress low KE electrons (one photon processes) – hence electrons detected are due solely to multiphoton events
- Energetically –
  2 × (93) eV – 118 eV = 68 eV
- Yield scales quadratically, n=1.95 ± .2





Summary - One Colour

- Xenon – Demonstration of an 'above threshold absorption-ionization' two-photon process involving an *inner shell electron*.

- It is clear that the although single photon ionization processes dominate, they are sufficiently important at high irradiance that, for a given intensity, much higher ionization stages can be reached compared to optical lasers.

- The strength and the nature of the $4d \rightarrow \varepsilon f$ resonance may open up, at high irradiance, additional ionization channels, namely the *simultaneous multiphoton / multi-electron from the inner 4d shell, 'inside-out ionization' or 'peeling the onion from the inside out'*

- *Kr (Not Shown) – was the first step on the road to resonant NL processes with EUV/X-rays…. **REMPI at X-rays.***

Xe - Richardson et al. PRL (July 2 – 2010), Kr - Meyer et al., PRL (May 28 - 2010)

XUV (X-ray) + IR Ionization

1. Rudiments of ionization processes in intense laser fields
2. Photoionization experimental setups (FLASH & DESY)
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Atoms in Intense Superposed *X-ray* + IR Laser Fields

Main objective

Study the effect of X-ray pulse width on fundamental photoionization processes in intense and ultrashort ionizing (FEL) and dressing (NIR) fields

Fringe benefit: Can help one develop techniques to measure X-ray pulse duration, X-ray-to-optical laser jitter, etc. on a femtosecond timescale….

Two Extremes:

X-ray pulse duration is ‘many’ optical cycles (800 nm => 2.5 fs)

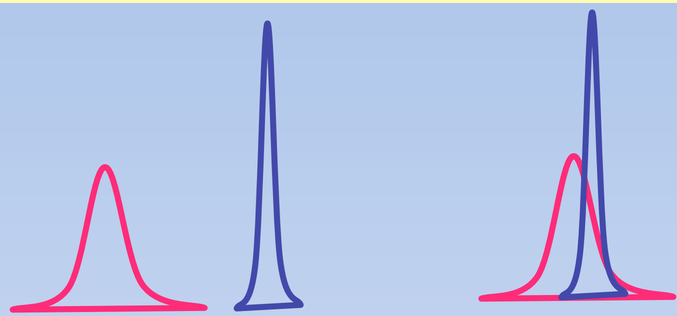
X-ray pulse duration is less than ½ optical cycle (< 1.25 fs)

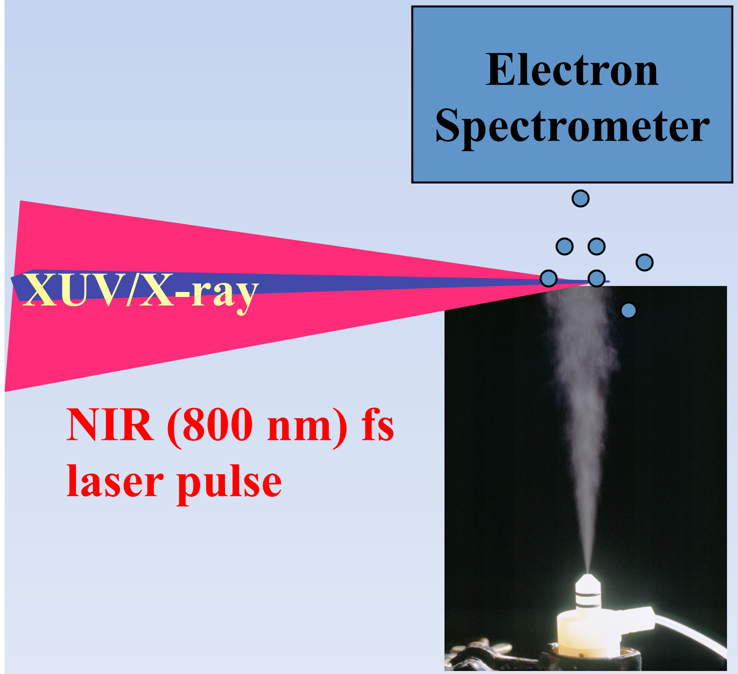
Two colour ATI/ Laser Assisted PES

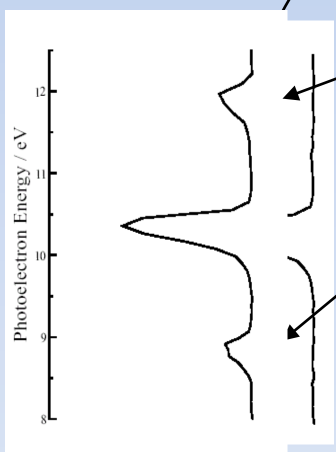
Superposition of visible and XUV pulses in a noble gas jet

Schins et al. PRL ***73****, 2180 (1994)*

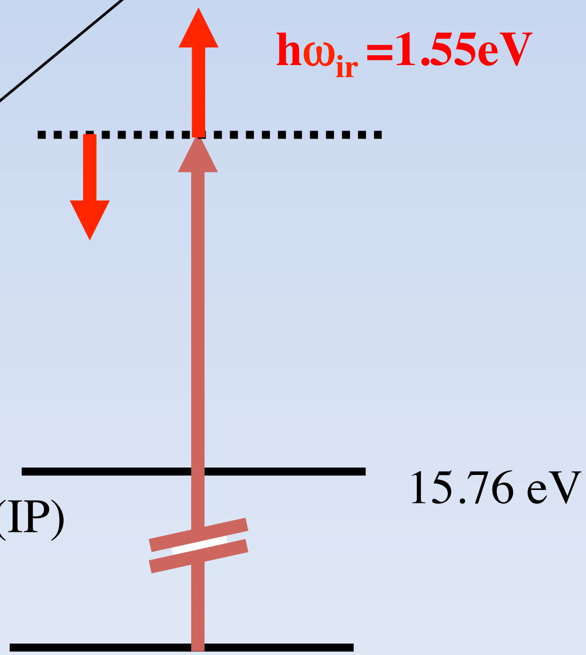
E.S. Toma et al. PRA **62** 061801 (2000)





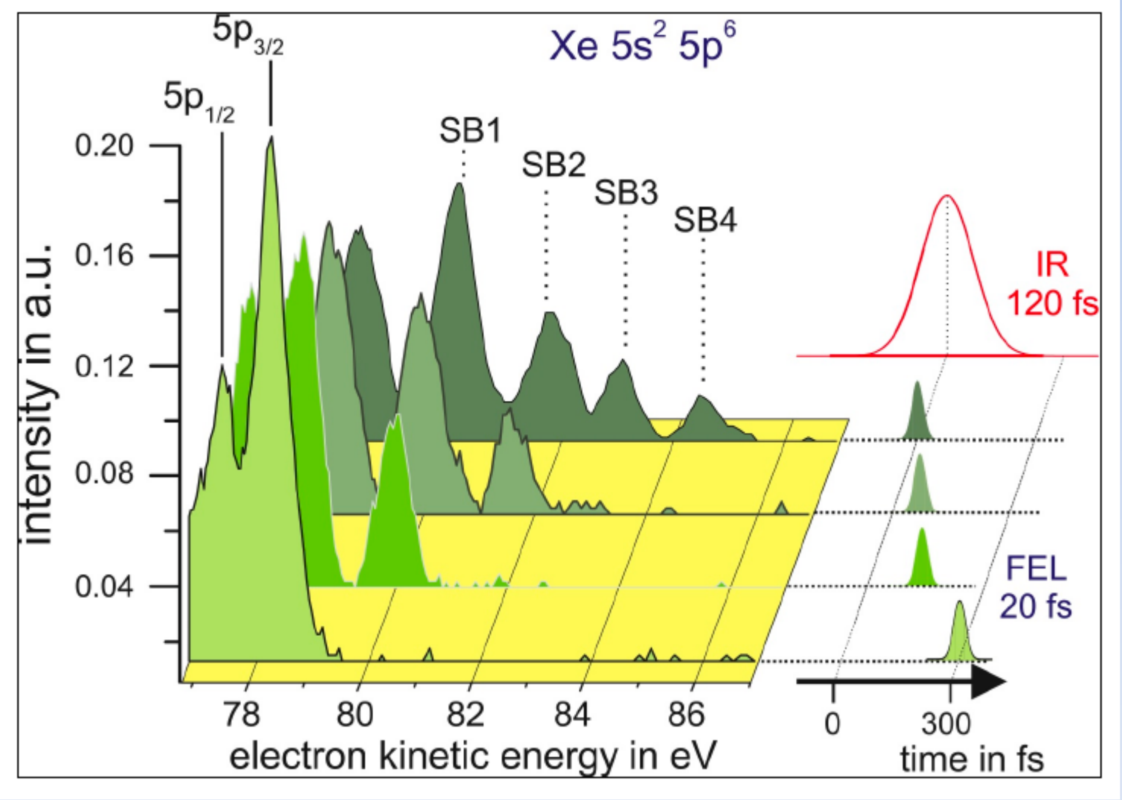


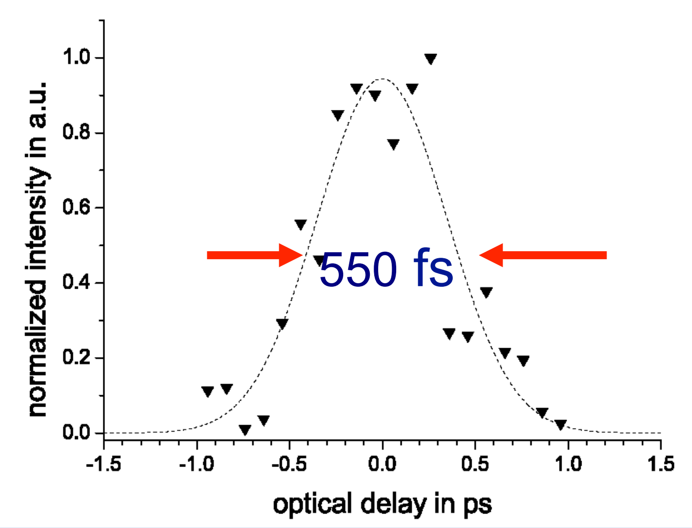
Sideband intensity *very sensitive* to XUV-IR pulse area overlap.
- Cross Correlation…



Atoms in 'Long' XUV (X-ray) + IR Fields

Sideband number/intensity depend strongly on XUV/NIR overlap ⇒ by comparison with theory ***we are able to determine relative time delay to better than 100 fs***





1. Ultrafast XUV-modulated optical-reflectivity methods
C. Gahl et al., Nature Photonics **2** 165-169 (2008)
T. Maltezopoulos et al., New J Phys **10** Art. No. 033026 (2008)

2. 'TEO'
A. Azima et al., APL, **94** 144102 (2009)

NCPST sfi science foundation ireland

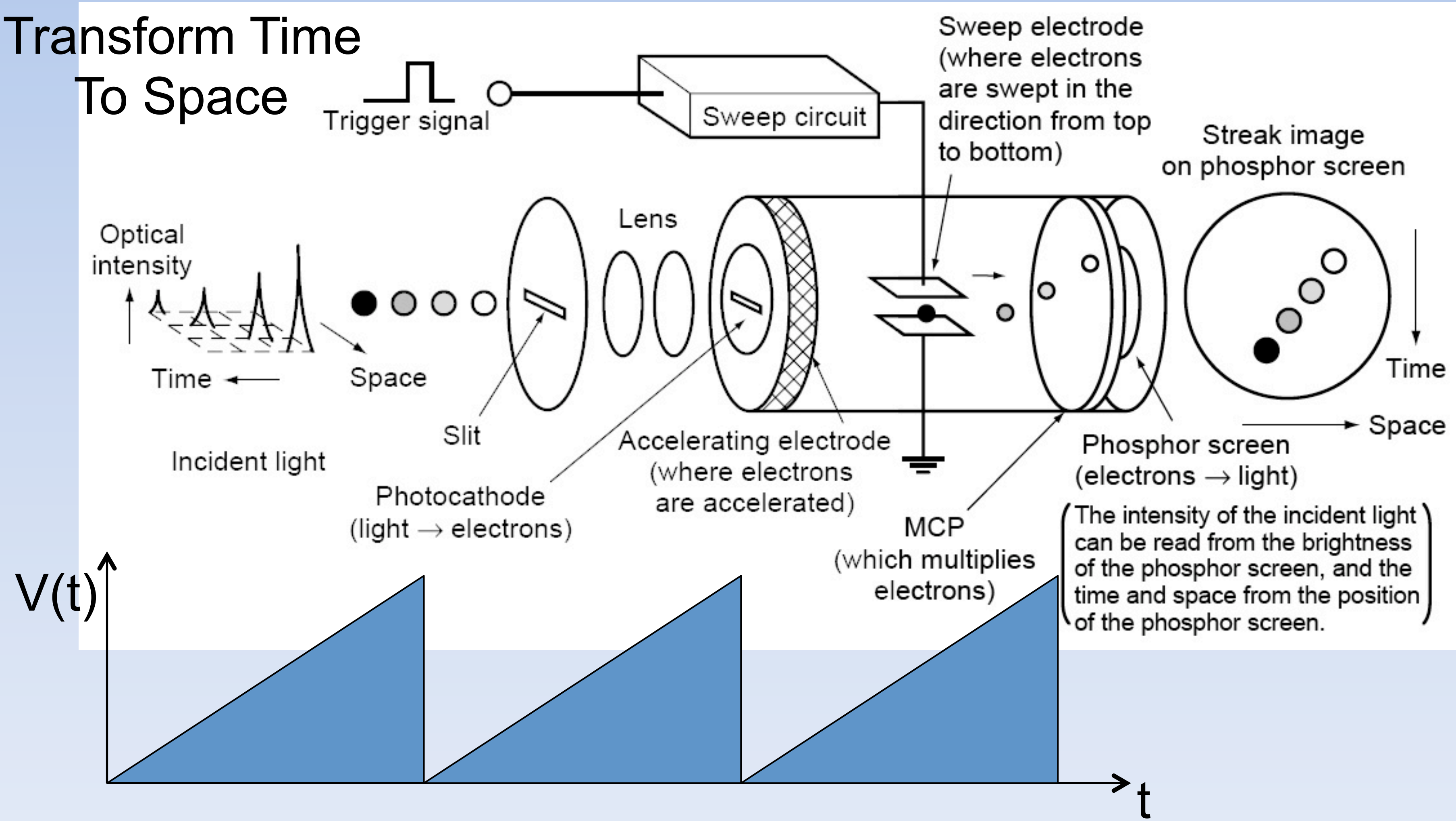
NIMA **83**, 516-525 (2007)
Appl. Phys. Lett **90** 131108 (2007)

inspire Integrated NanoScience Platform for Ireland DCU

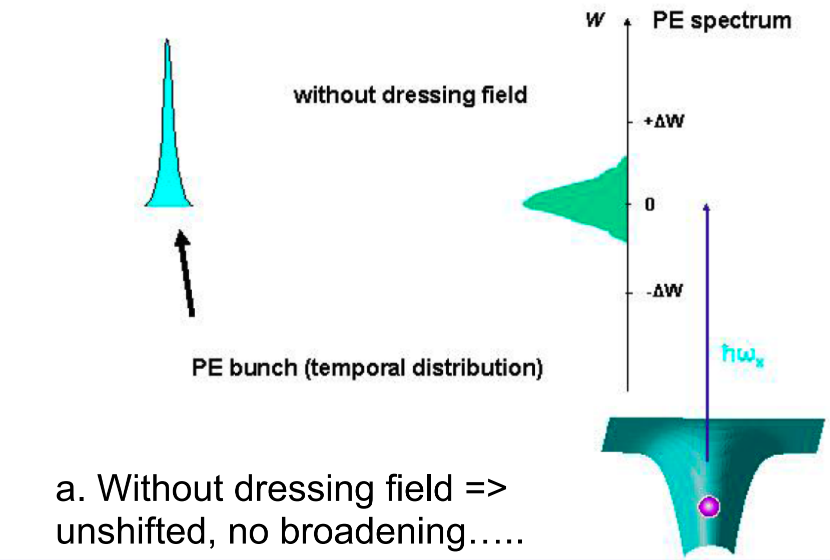
Atoms in 'Short' XUV (X-ray) + IR Fields

Streak Camera Operation – Courtesy Hamamatsu Corp.

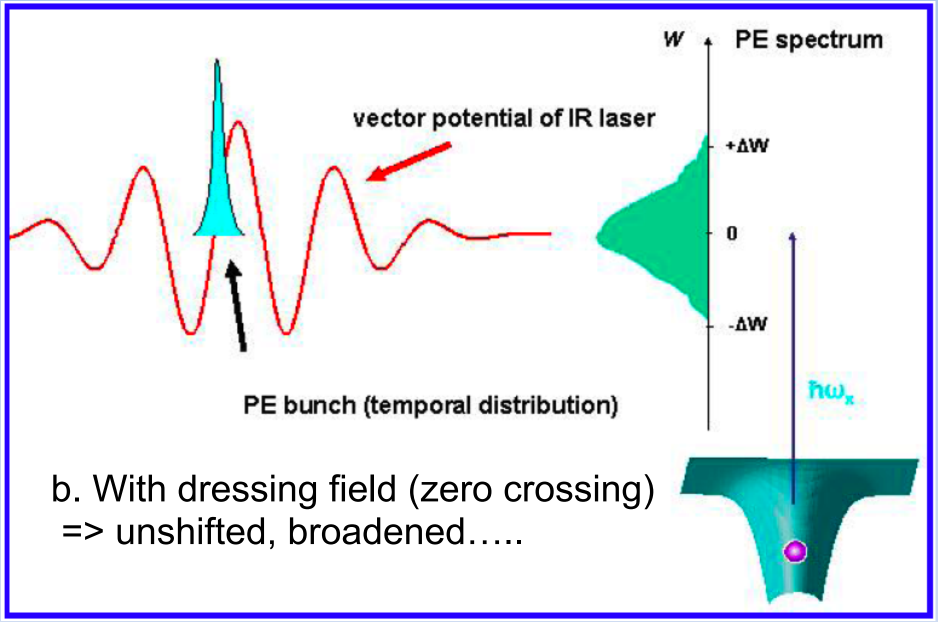
Transform Time
To Space



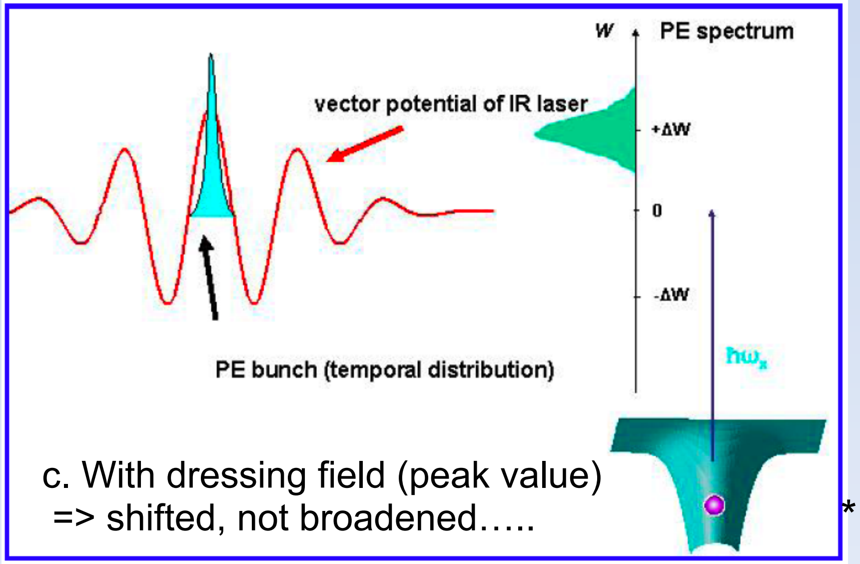
Atoms in 'Short' XUV (X-ray) + IR Fields



a. Without dressing field => unshifted, no broadening…..



b. With dressing field (zero crossing) => unshifted, broadened…..



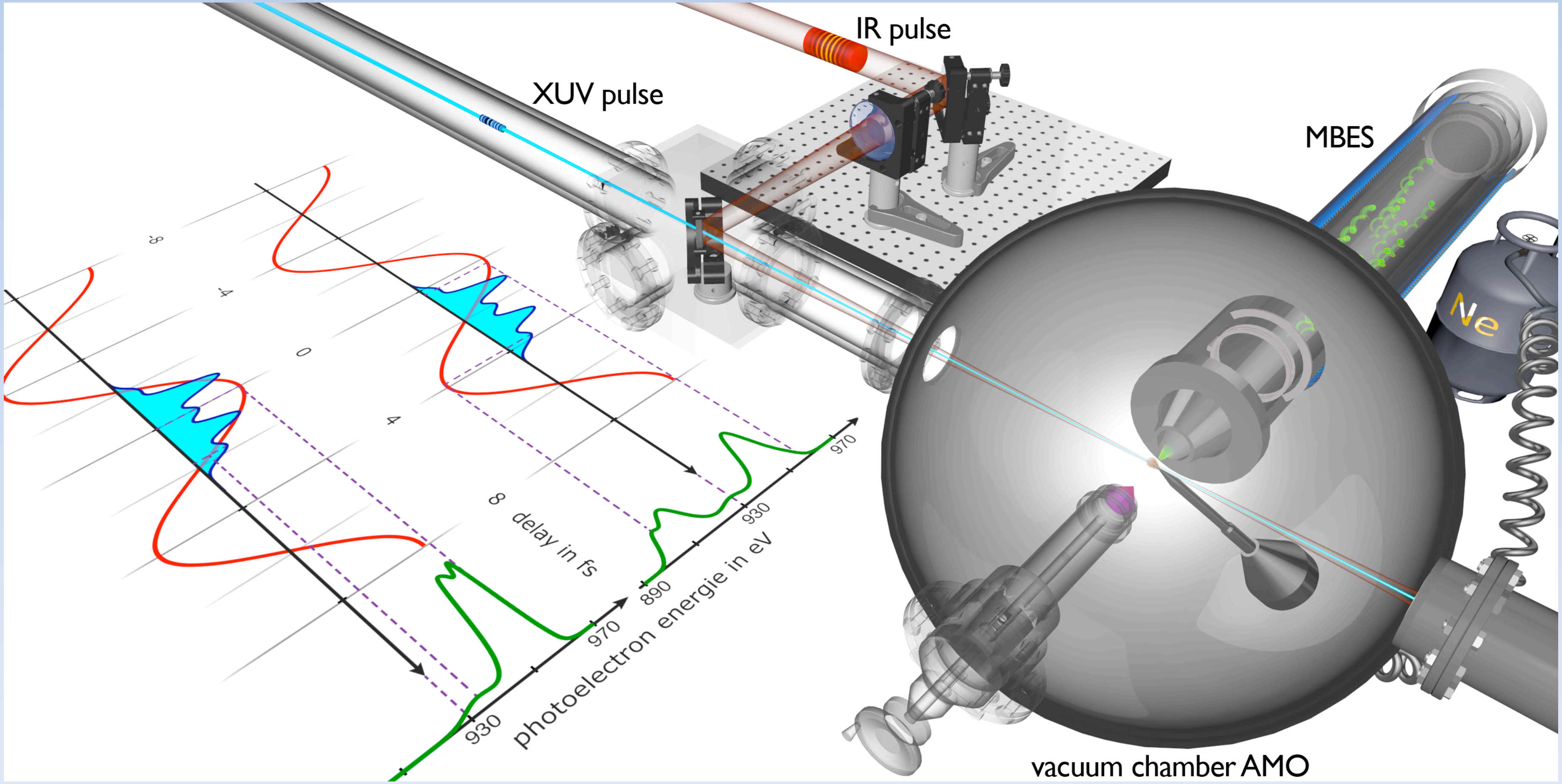
c. With dressing field (peak value) => shifted, not broadened…..

***Single Shot Atomic Streak Camera – SSASC => few fs pulse widths.* Target:** Neon, **LCLS: >**870 eV**,** ~1 - 4 fs**, Laser:** *OPA (2000 nm, ~ 7 fs),*

* *R. Kienberger et al., J. Mod. Opt **52** 261-275 (2005)*

Measurement of few fs pulses @ LCLS

Experimental Layout at LCLS



NCPST EXTATIC

Nature Photonics **8**
pp950 - 957 (2014)

sfi inspire Integrated NanoScience Platform for Ireland DCU

Measurement of few fs pulses @ LCLS

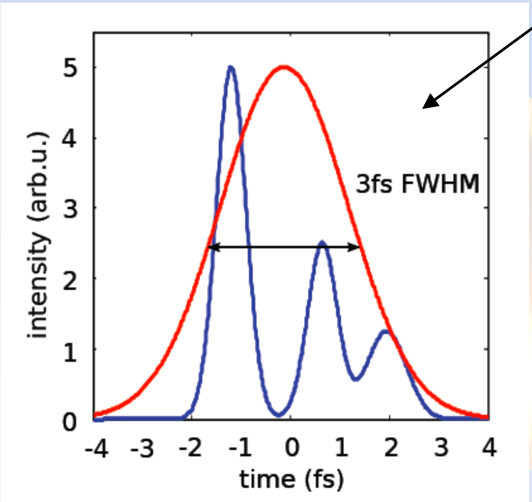
LCLS low current/ slotted spoiler/ few fs mode -

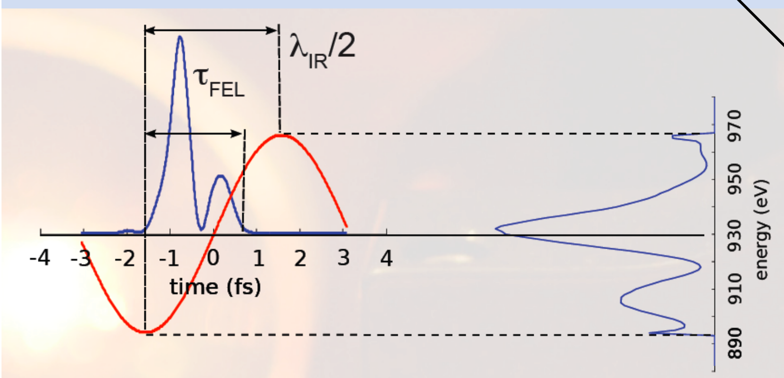
Data still under analysis……

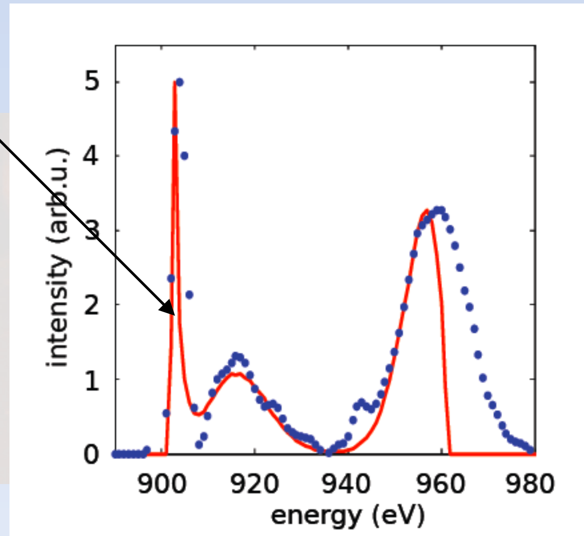
Process. Ne + hν (1.8 keV) -> Ne^{+} ($1s^{-1}$) + e- +I_L (10^{14} $W.cm^{-2}$)

Essentially mapping time (fs) to energy in (eV) allows one to measure X-ray (and EUV) pulse widths to attosecond accuracy provided the *X-ray (EUV) pulse width is less than one one half cycle of the optical laser in duration* !!

3fs case - simulation and experiment………



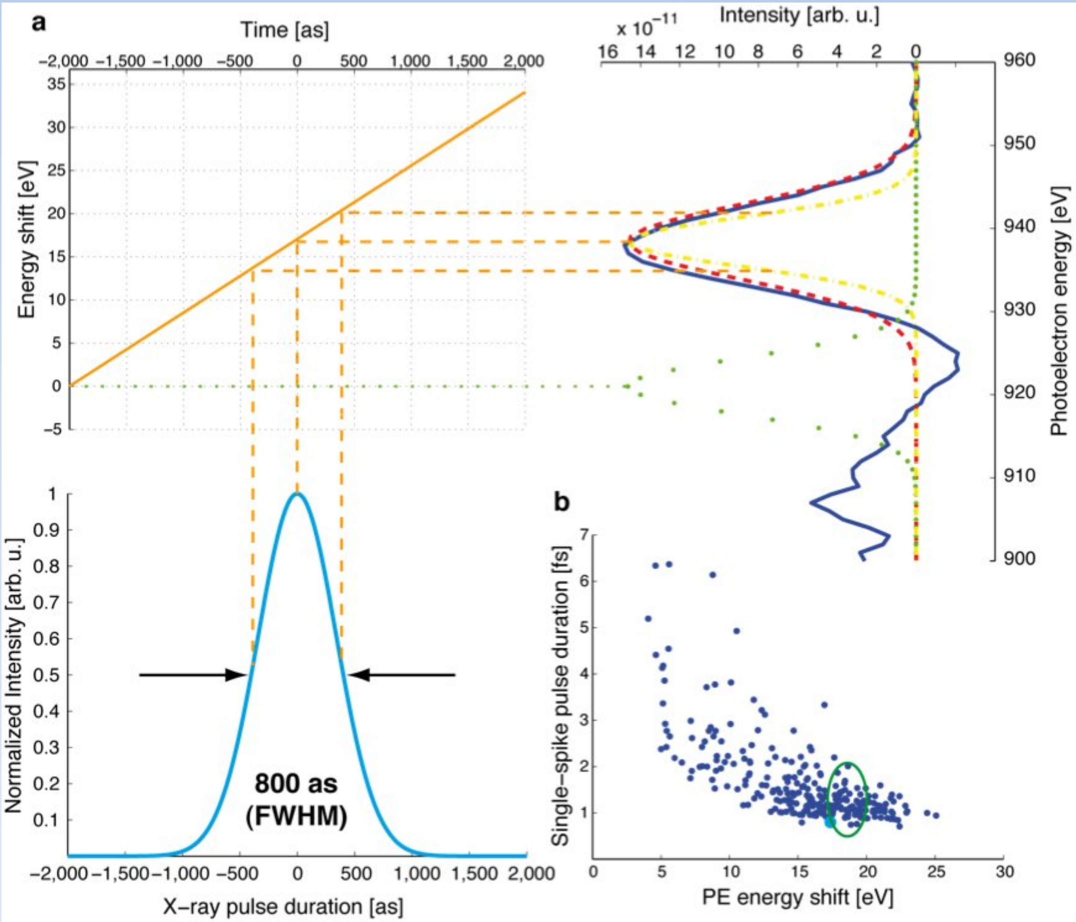




Sub-femtosecond pulses @ LCLS

800 as X-ray pulse !!

Process. Ne + hν (1.8 keV) -> Ne^+ ($1s^{-1}$) + e- +I_L (10^{14} $W.cm^{-2}$)



200 uJ in 800 as =

2 x 10^{-4} J/ 8 x 10^{-16} s =

0.25 TW peak power

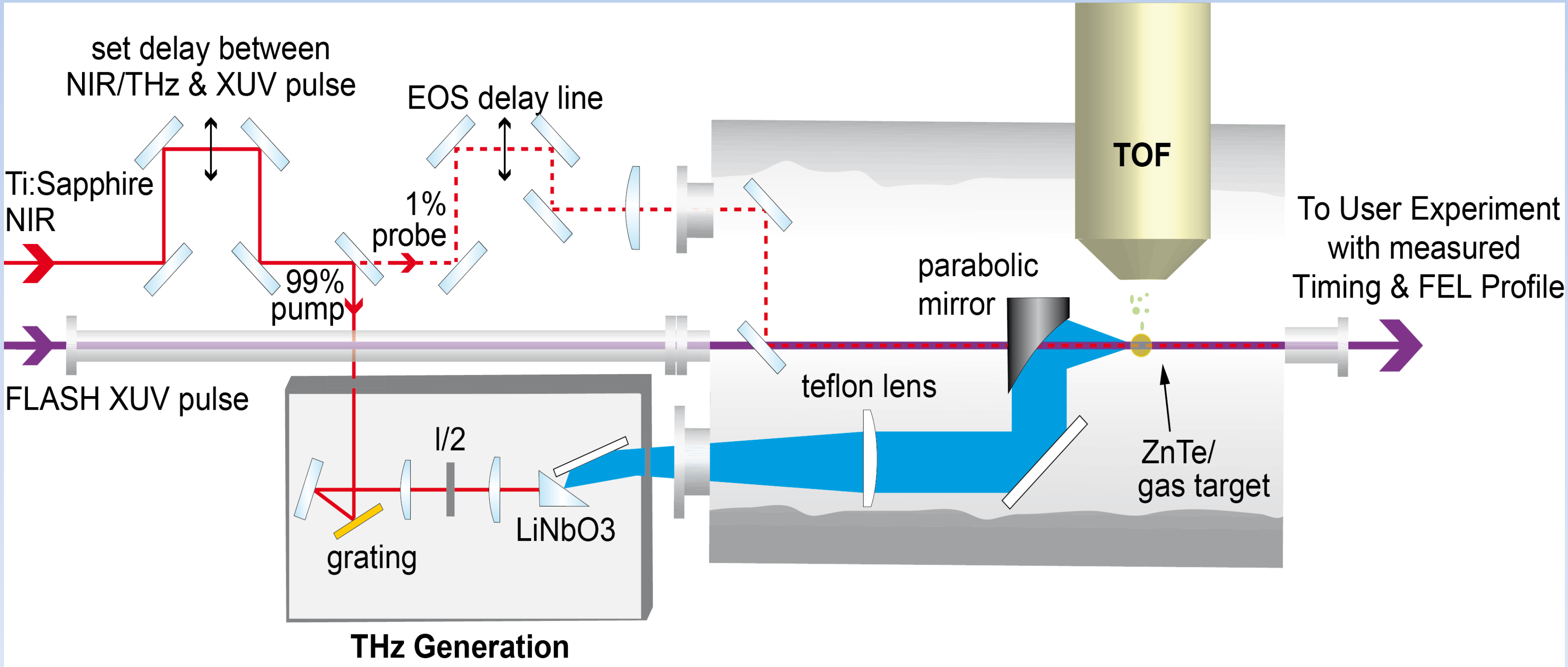
Focused to a spot of
10 um = 10^{-6} cm^2 =>

An irradiance of:
2.5 x 10^{17} $W.cm^{-2}$!!!

Single Cycle THz Streaking @ FLASH

Femtosecond Atomic Streak Camera

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

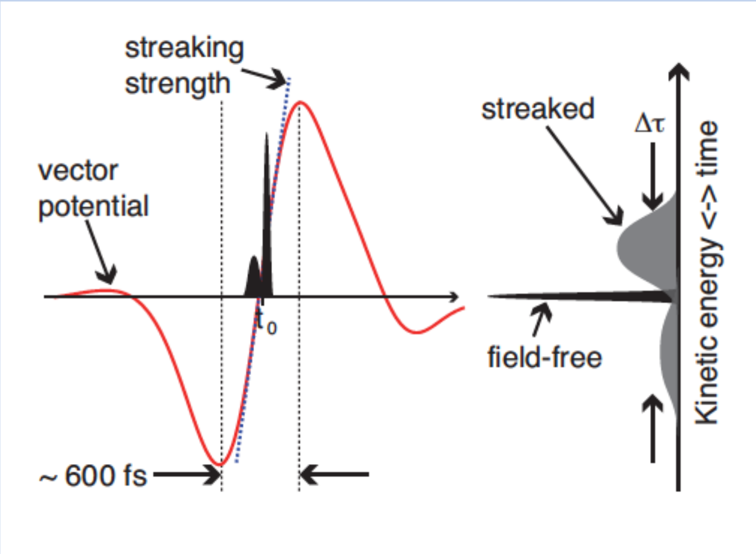


Schematic layout of the THz Streaking Experiment at FLASH

Single Cycle THz Streaking @ FLASH

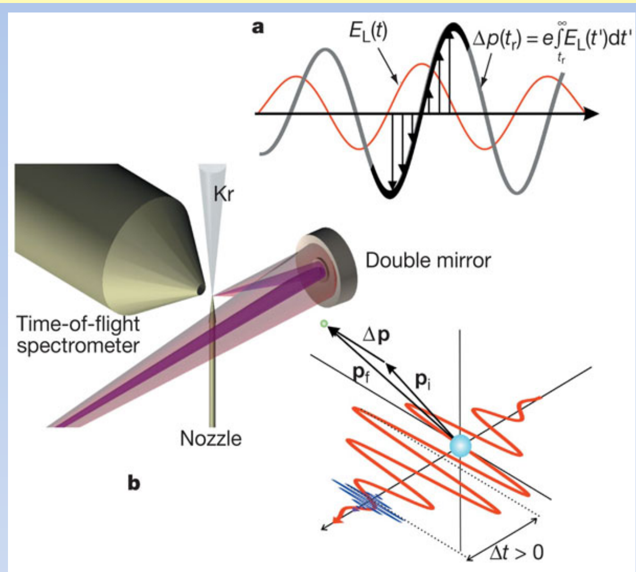
Femtosecond Atomic Streak Camera

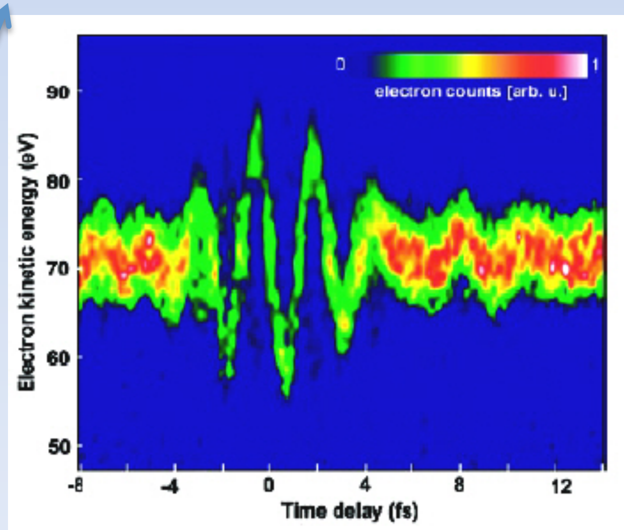
Generate single (picosecond) cycle pulse using optical rectification of Ti-Sappire laser pulses – field ~ 50MV/m maximum



Principle of the experiment

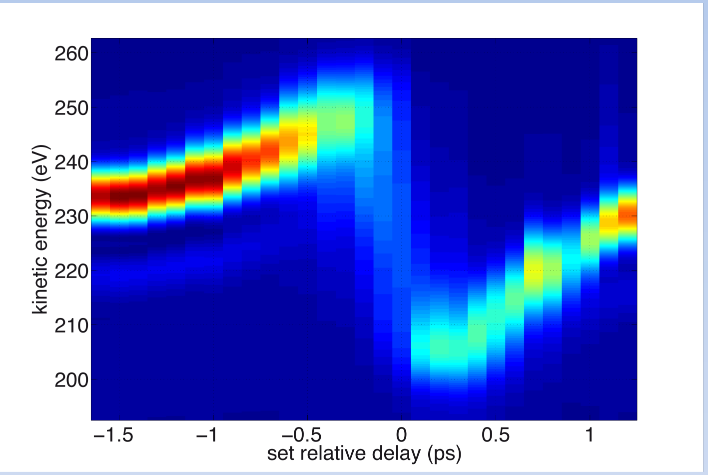
Attosecond Photoelectron Streaking showing how the E-field of a few cycle fs laser pulse can be mapped – MPI-Q.



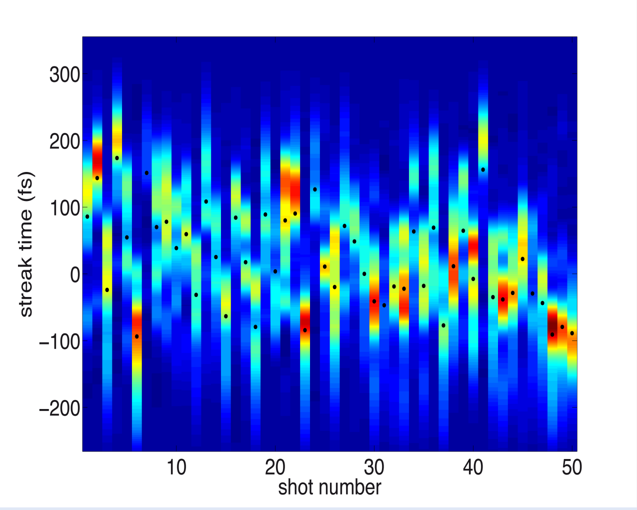


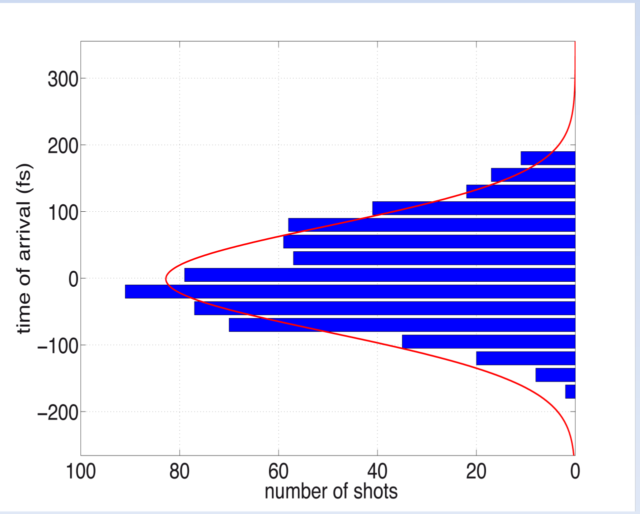
Single Cycle THz Streaking @ FLASH

A Cavalieri et al. from CFEL, DCU, XFEL & DESY



Single cycle THz Photoelectron Streaking showing how the E-field of a single cycle ps laser pulse can be mapped



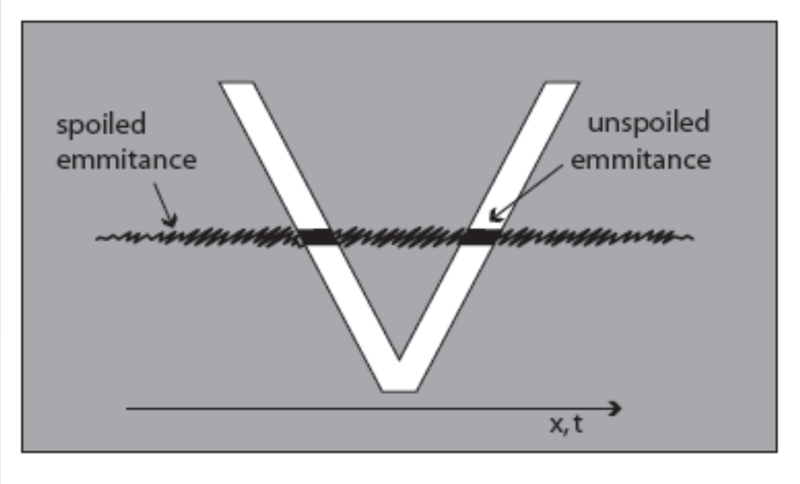


Jitter measurements on 50 consecutive streak traces

Nature Photonics **6** pp852-857 (2012)

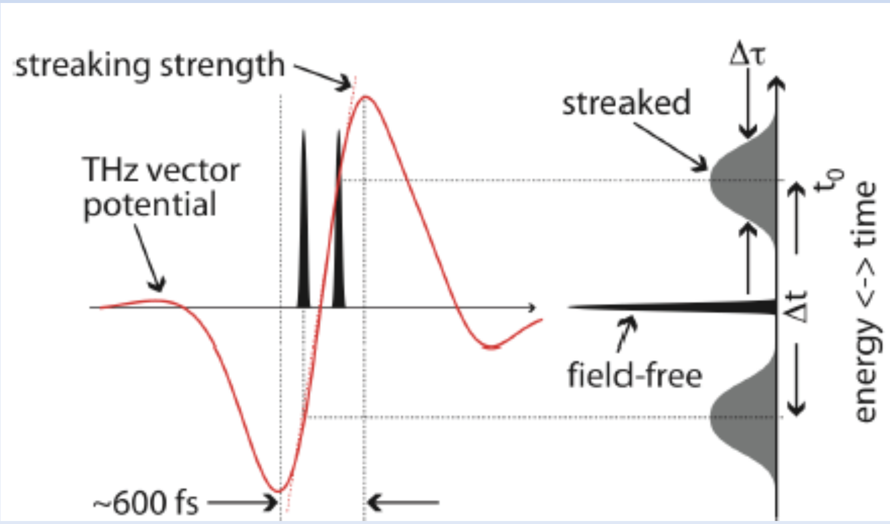
LCLS - Single Cycle THz Streaking

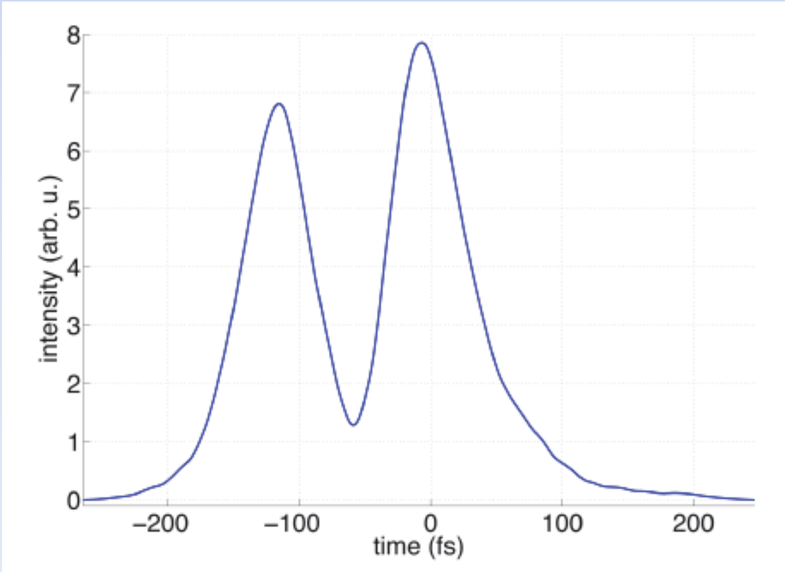
A Cavalieri et al. from CFEL, DCU, XFEL & SLAC



If the dispersed bunch is intercepted by a 'V-shaped' vertical slot, then **the emittance of the all but TWO small parts in space (time) of the bunch is 'spoiled'** -=> 2 X 'few fs' pulses of variable separation result.

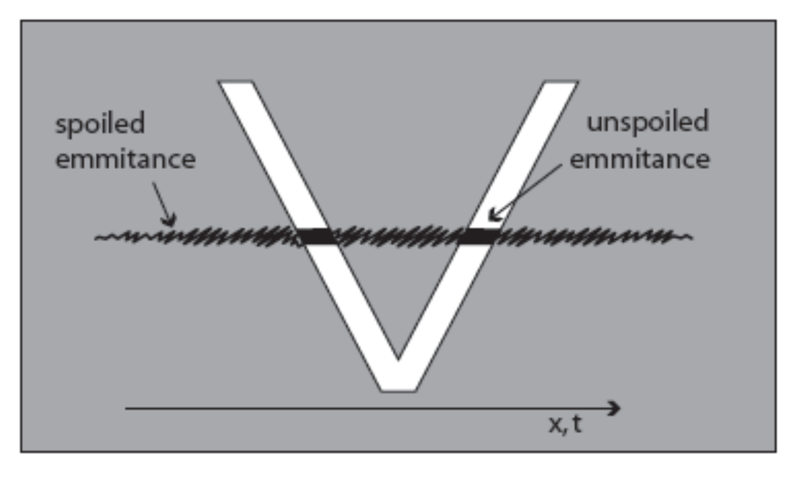
P Emma et al., PRL **109** 254802 (2012)

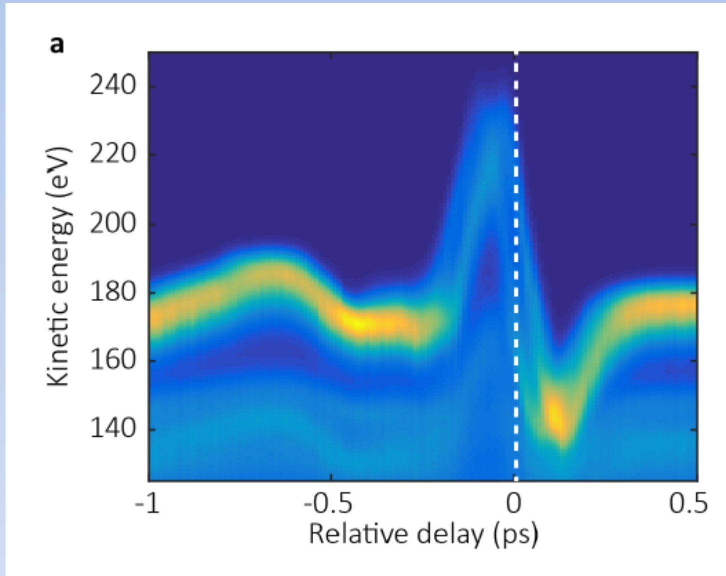


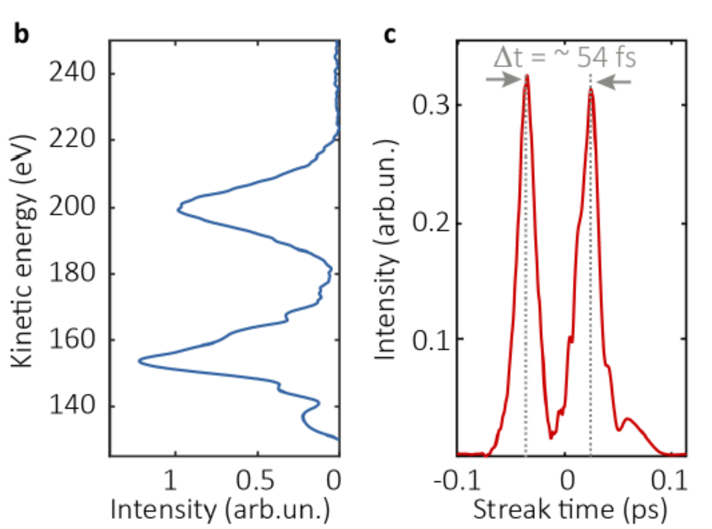


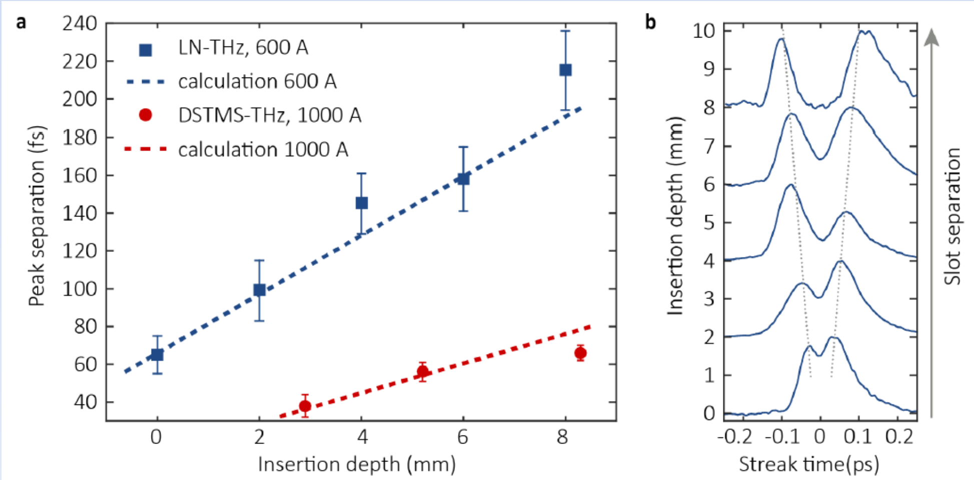
LCLS - Single Cycle THz Streaking

A Cavalieri et al. from CFEL, DCU, XFEL & SLAC









NEW !! All Optical Synchronisation - FLASH

A Cavalieri et al. from CFEL, DCU, MPI (SDM), SLAC & XFEL

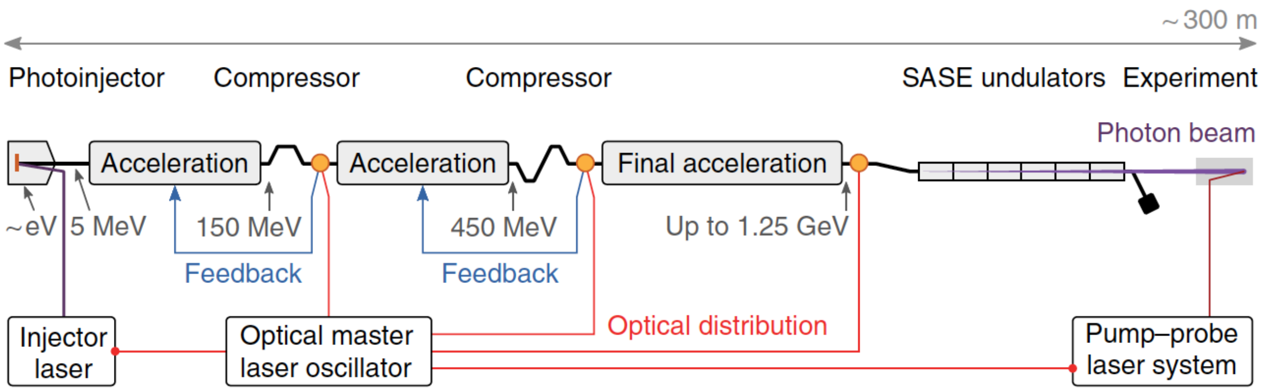

nature COMMUNICATIONS

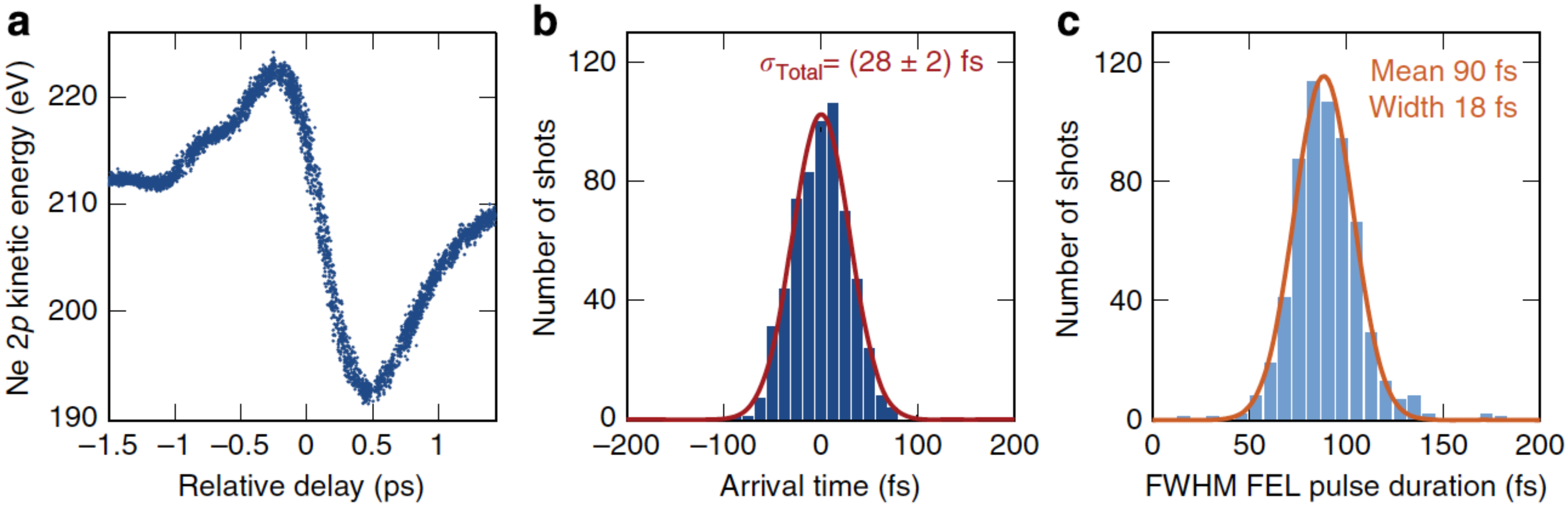
ARTICLE

Received 10 Apr 2014 | Accepted 24 Nov 2014 | Published 20 Jan 2015 DOI: 10.1038/ncomms6938 OPEN

Femtosecond all-optical synchronization of an X-ray free-electron laser

S. Schulz[1], I. Grguraš[2,3,4], C. Behrens[1,5], H. Bromberger[2], J.T. Costello[6], M.K. Czwalinna[1], M. Felber[1], M.C. Hoffmann[5], M. Ilchen[7], H.Y. Liu[2], T. Mazza[7], M. Meyer[7], S. Pfeiffer[1], P. Prędki[8], S. Schefer[4], C. Schmidt[1], U. Wegner[1], H. Schlarb[1] & A.L. Cavalieri[2,3,4]





NCPST EXTATIC

Nature Comms **6**
Art. No. 5938 (2015)

sfi inspire DCU

Two Colour ARPES – AR Sidebands

FEL:
PG2 Beamline – Zero Order
hν=44 eV (28.2nm), 50μJ/pulse, 100 fs, 150 μm

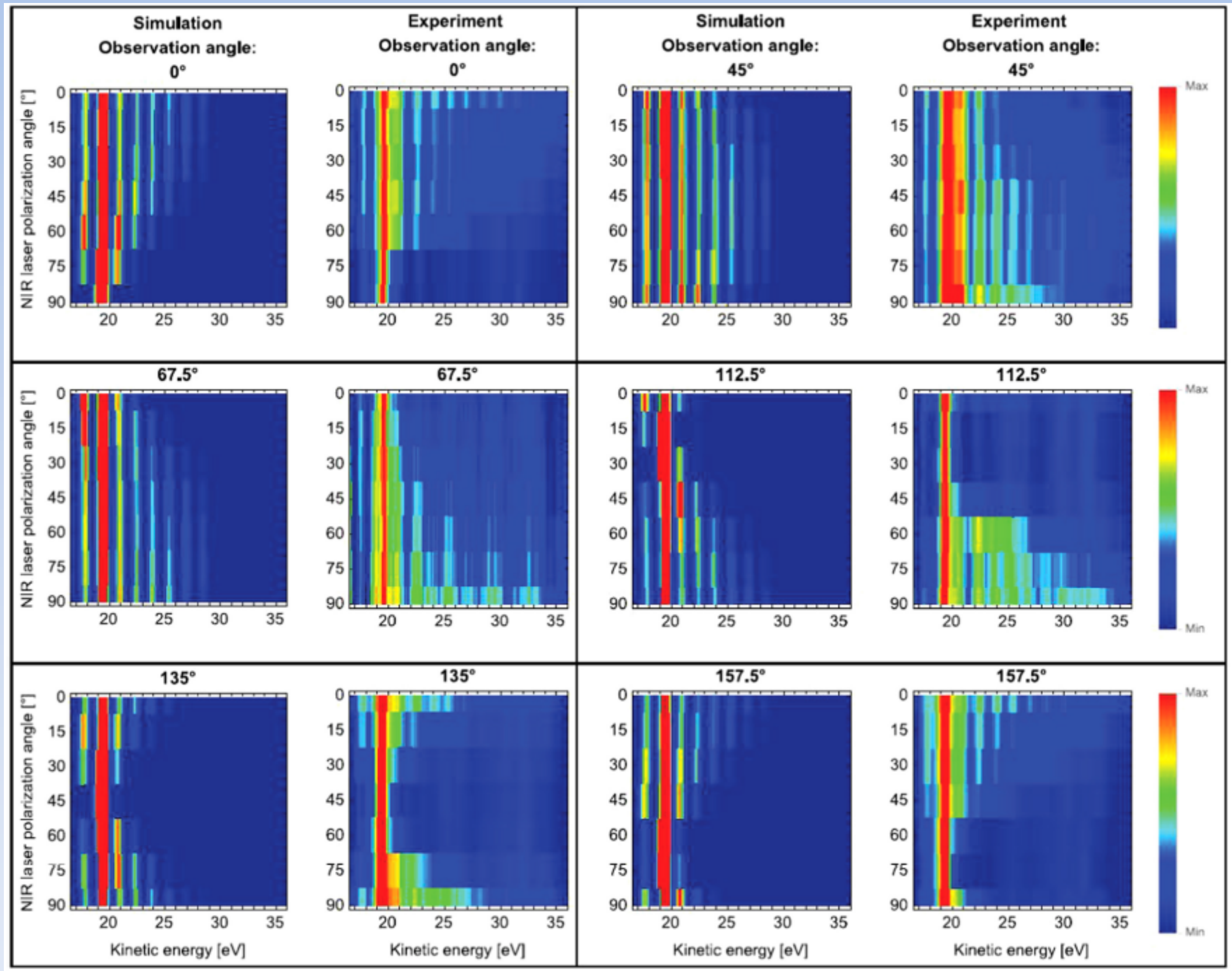
Laser:
Ti-Sa. 800 nm, 0.5 mJ, 120 fs, 200 μm
Polariser/Waveplate: 15^{O} increments

TOF:
16 TOF channels at 22.5^{O} increments
4GS/sec/channel sampling rate / 12 bits

Timing jitter: 100 fs (re-sort by number of SBs)

J. Phys. B: At. Mol. Opt. Phys. **49,** 165003 (2016)

Two Colour ARPES – AR Sidebands - He



Main photoline at 19.4 eV = 44 eV – IP (He)

Each panel corresponds to an angle between the XUV and NIR polarisation directions

Each shows count rate versus electron emission angle

Two Colour ARPES – AR Sidebands - He

The intensity of the m^th^ sideband is given by:

$$\sigma^{(m)} \sim |J_m(\bar{q})|^2 \sin^2\vartheta \cos^2\varphi$$

Where:

$$\bar{q} = \frac{A_L k}{\omega_L} \sin\vartheta \cos(\phi - \chi)$$

- θ and φ are electron emission angles
- A_L and ω_L are the NIR vector potential and angular frequency
- k is the free electron linear momentum
- χ is the angle between the XUV and NIR polarisation directions

A K Kazansky et al. Phys Rev A **85,** 053409 (2012)

Two Colour ARPES - AR Sidebands - He

- FEL operated in two bunch (pulse) mode – every second pulse used to obtain a 'FEL only' spectrum to check operation of the detectors – e.g., He $\beta_2 = 2.0$….
- Photoelectron spectra for opposite detectors were all checked to be identical
- The relative sideband intensities vary considerably with polarisation direction of the NIR field
- For aligned polarisations (XUV and NIR) and observation direction one sees the highest number of SBs. For orthogonal polarisations the SB number is smallest
- In the latter case the electron is emitted normal to the NIR field and does not interact with it
- In the case high NIR fields the SBs depend only on it
- The process is easily understood from the SFA model….

Two Colour ARPES - Circ. Dichroism - He

- What happens when we change from plane polarised XUV and NIR to circularly polarised XUV and NIR ?

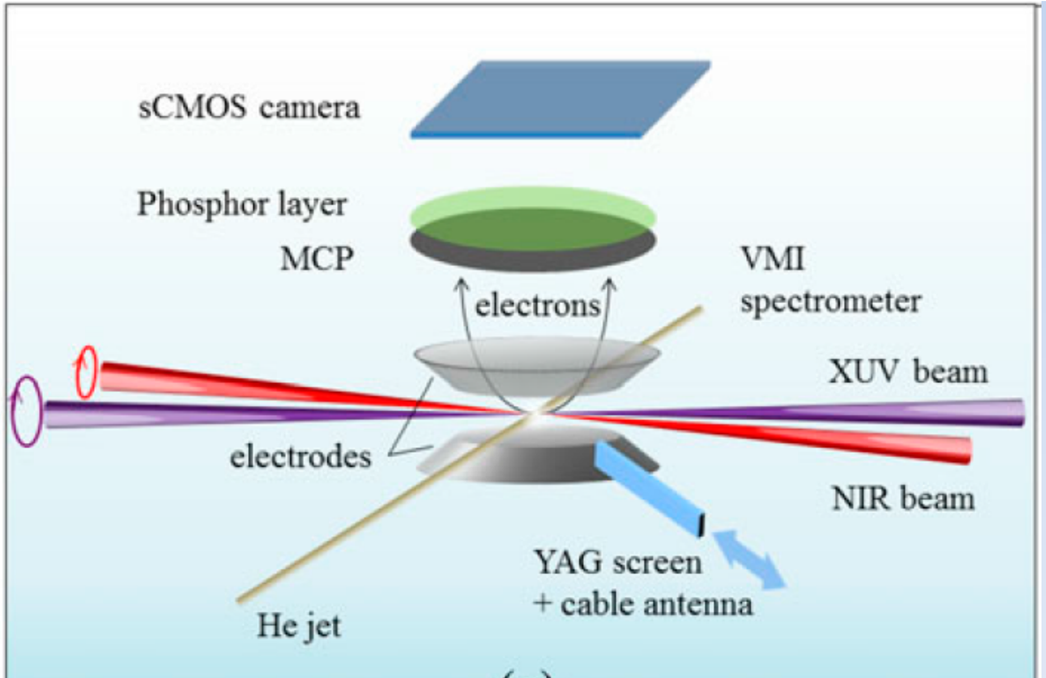
1. In single colour MPI of non-oriented atoms by circularly polarised light the cross section and angular distribution of emitted electrons do not depend on the helicity of the laser.
2. In two colour ionisation of non-oriented atoms, where both beams are circularly polarised, it is found that the photoelectron yield and the angular distribution are slightly different for co-rotating and counter-rotating XUV and NIR beams and so the system exhibits induced Circular Dichroism [e.g., J. Phys. B: At. Mol. Opt. Phys. 1999, 32, 3747– 3767]

Two Colour ARPES - Circ. Dichroism - He

Angular distribution and circular dichroism in the two-colour XUV+NIR above-threshold ionization of helium

J. Mod. Opt. 63, pp367-382 (2016)

T. Mazza[a], M. Ilchen[a], A.J. Rafipoor[a], C. Callegari[b], P. Finetti[b], O. Plekan[b], K.C. Prince[b,c,d], R. Richter[b], A. Demidovich[b], C. Grazioli[b], L. Avaldi[e], P. Bolognesi[e], M. Coreno[e], P. O'Keeffe[e], M. Di Fraia[f], M. Devetta[g], Y. Ovcharenko[h], V. Lyamayev[a,i], S. Düsterer[j], K. Ueda[k], J.T. Costello[l], E.V. Gryzlova[m], S.I. Strakhova[m], A.N. Grum-Grzhimailo[m], A.V. Bozhevolnov[n], A.K. Kazansky[o,p,q], N.M. Kabachnik[a,m,q] and M. Meyer[a]



(a)

FERMI@ELETTRA
FEL: 48.4 eV, 80 μJ, 100fs,
Focus: 50 μm, 10^{13} W.cm^{-2}

Laser: 800 nm, 30 to 750 μJ, 175 fs,
Focus: 200 μm, 3 x10^{11} - 7 x10^{12} W.cm^{-2}

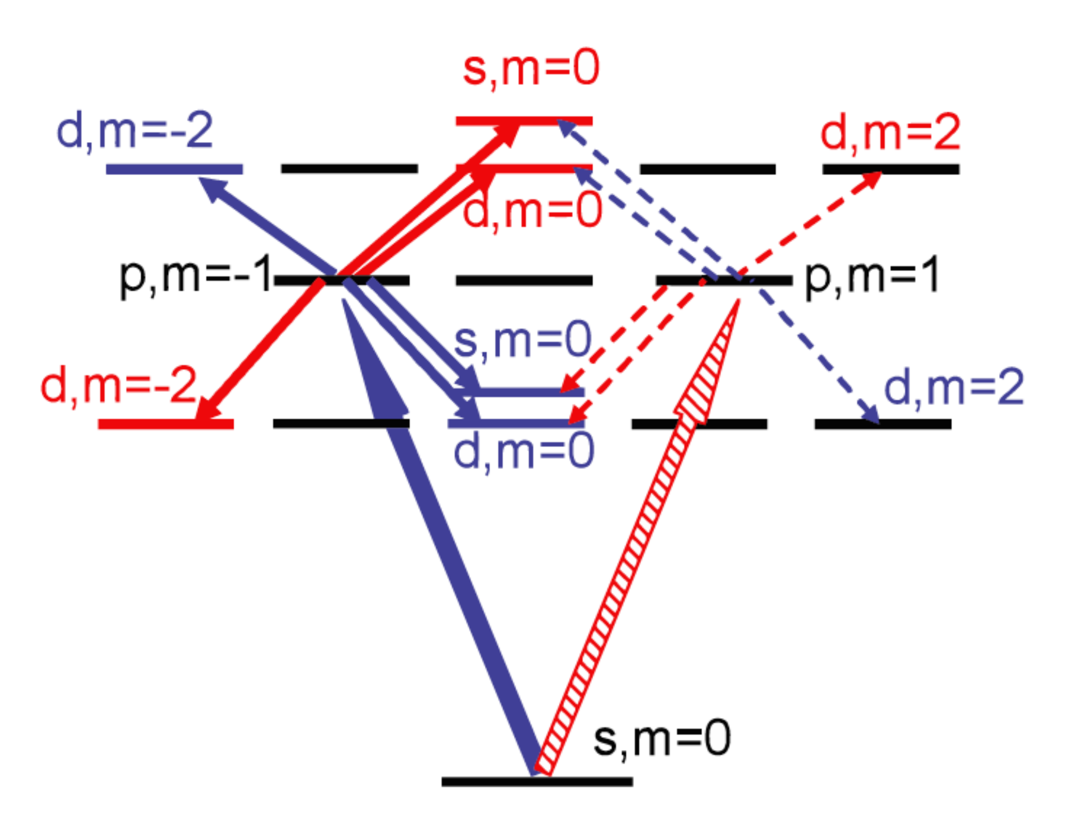
Timing Jitter: 25 fs (rms)

Camera: CMOS, 2560x2160, 6.5 μm pixel

Two Colour ARPES - Circ. Dichroism - He

He: 2-colour, 2-photon excitation scheme

Blue: Left Circularly Polarised (LCP) XUV
Red: Right Circularly Polarised (RCP) XUV



J. Mod. Opt. 63, pp367-382 (2016)

Special Issue: *Short Wavelength Free Electron Lasers*, Journal of Modern Optics, 63:4, pp285-287 (2016)
Editors: John Costello, Eugene Kennedy & Lampros Nikolopoulos

Two Colour ARPES - Circ. Dichroism - He

Theory: SFA (strong NIR fields) and Perturbation Theory (weak NIR fields)

SFA:

Photoionization amplitude =

$$\mathcal{A}_{\vec{k}}^{++} = -i\int_{-\infty}^{\infty} \mathrm{d}t\tilde{\mathcal{E}}_X(t) d_{sp} Y_{1,+1}(\theta_0(t),\phi_0(t)) \times e^{i\Phi(\vec{k},t)} e^{i(E_b-\omega_X)t},$$

where

$$\Phi(\vec{k},t) = -\frac{1}{2}\int_t^{\infty} \mathrm{d}t' \left[\vec{k} - \vec{A}_L(t')\right]^2$$

$$k_0^2(t) = (\vec{k} - \vec{A}_L(t))^2$$

$$\theta_0(t) = \arccos\left(k_z/k_0(t)\right),$$

$$\exp\left(i\phi_0(t)\right) = \frac{(k_x - A_{Lx}(t)) + i(k_y - A_{Ly}(t))}{(k_0^2(t) - k_z^2)^{1/2}}$$

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\nu\nu'} \sim |\mathcal{A}_{\vec{k}}^{\nu\nu'}|^2$$

CD Angle Resolved

$$\mathrm{CDAD} = \frac{|\mathcal{A}_{\vec{k}}^{++}|^2 - |\mathcal{A}_{\vec{k}}^{+-}|^2}{|\mathcal{A}_{\vec{k}}^{++}|^2 + |\mathcal{A}_{\vec{k}}^{+-}|^2}$$

CD Angle `integrated

$$\mathrm{CD} = \frac{\int \mathrm{d}\Omega|\mathcal{A}_{\vec{k}}^{++}|^2 - \int \mathrm{d}\Omega|\mathcal{A}_{\vec{k}}^{+-}|^2}{\int \mathrm{d}\Omega|\mathcal{A}_{\vec{k}}^{++}|^2 + \int \mathrm{d}\Omega|\mathcal{A}_{\vec{k}}^{+-}|^2}$$

Two Colour ARPES - Circ. Dichroism - He

Theory: SFA (strong NIR fields) and Perturbation Theory (weak NIR fields)

PT:

Differential Cross-section = $$\left(\frac{d\sigma}{d\Omega}\right)_{\nu\nu'} = \frac{\sigma_{\nu\nu'}}{4\pi}\left(1 + \beta_2^{\nu\nu'} P_2(\cos\theta) + \beta_4^{\nu\nu'} P_4(\cos\theta)\right)$$

$$\sigma_{+-} = 2\pi(2\pi\alpha)^2\omega_X\omega_L\left(\frac{1}{9}|D_s|^2 + \frac{1}{45}|D_d|^2\right)$$

$$\beta_2^{+-} = \frac{2}{7}\frac{|D_d|^2 + 7\,\Re[e^{i\Delta_{ds}}D_sD_d^*]}{|D_s|^2 + \frac{1}{5}|D_d|^2},$$

$$\beta_4^{+-} = \frac{18}{35}\frac{|D_d|^2}{|D_s|^2 + \frac{1}{5}|D_d|^2},$$

$$\sigma_{++} = 2\pi(2\pi\alpha)^2\omega_X\omega_L\frac{2}{15}|D_d|^2$$

where $\Delta_{ds} = \delta_d - \delta_s$ and D_l (l=0 or 2) are the two photon matrix elements

$$CD = \pm\frac{5}{7}\frac{|D_d|^2 - |D_s|^2}{|D_d|^2 + \frac{5}{7}|D_s|^2}$$

$$D_l(E) = \int \frac{\int P_{El}(r) r P_{E'p}(r)\,dr \int P_{E'p}(r) r P_{1s}(r)\,dr}{E' - \omega_X + i0}\,dE'$$

$$E = E_{gr} + \omega_X \pm \omega_L$$

Two Colour ARPES - Circ. Dichroism - He

He at two NIR intensities – ratio ~ 25……….

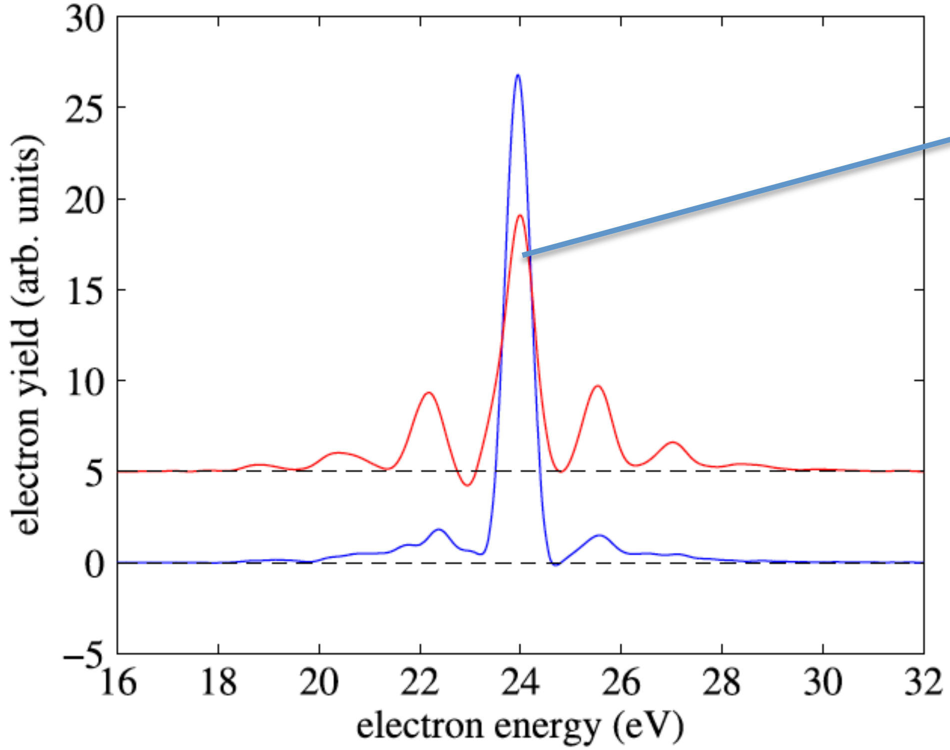


Figure 4. High intensity (red curve) and low intensity (blue curve) spectra at $\pi/2$ emission angle when both XUV and NIR pulses are left-hand circularly polarized. NIR intensities are 7.2×10^{12} and 3×10^{11} W/cm^2, respectively. (The colour version of this figure is included in the online version of the journal.)

XUV and NIR both LCP '- -'

Decrease in main photoline..

Integrated signals ~ equal

3 SBs visible at 7 x 10^{12} $W.cm^{-2}$

'Weak': at 3 x 10^{11} $W.cm^{-2}$

'Strong': at 7 x 10^{12} $W.cm^{-2}$

Two Colour ARPES - Circ. Dichroism - He

Table 1. The anisotropy parameters β_2 and β_4 measured and calculated for the low-energy (SB_{-1}) and high-energy (SB_{+1}) sidebands in two-colour photoionization of He at low NIR intensity of 3×10^{11} W/cm^2. Theoretical values are calculated using the SFA and PT.

Weak NIR Field….

		β_2			β_4		
Case	Sideband	Exp	SFA	PT	Exp	SFA	PT
LL	SB_{+1}	−1.39 ± 0.02	−1.40	−1.43	0.41 ± 0.02	0.40	0.43
	SB_{-1}	−1.37 ± 0.04	−1.33	−1.25	0.38 ± 0.04	0.33	0.35
LR	SB_{+1}	−1.43 ± 0.02	−1.47	−1.30	0.43 ± 0.02	0.47	0.40
	SB_{-1}	−1.39 ± 0.04	−1.41	−1.43	0.40 ± 0.05	0.40	0.43

Table 2. The anisotropy parameters β_2, β_4, and β_6 measured and calculated for the low-energy (SB_{-n}) and high-energy (SB_{+n}) sidebands in two-colour photoionization of He at high NIR intensity of 7.2×10^{12} W/cm^2. Theoretical values are the result of simulation using the SFA as described in the text.

Strong NIR Field….

		β_2		β_4		β_6	
Case	Sideband	Exp	SFA	Exp	SFA	Exp	SFA
LL	SB_{-3}	−0.90 ± 0.09	−1.62	0.29 ± 0.04	0.72	−0.07 ± 0.10	−0.09
	SB_{-2}	−1.19 ± 0.07	−1.38	0.45 ± 0.16	0.34	−0.24 ± 0.20	0.05
	SB_{-1}	−1.15 ± 0.13	−1.09	0.17 ± 0.23	0.08	−0.02 ± 0.10	$-1 \cdot 10^{-5}$
	SB_{+1}	−1.17 ± 0.03	−1.17	0.16 ± 0.07	0.06	−0.02 ± 0.07	0.09
	SB_{+2}	−1.41 ± 0.07	−1.44	0.45 ± 0.12	0.39	−0.14 ± 0.10	0.08
	SB_{+3}	−1.46 ± 0.07	−1.68	0.55 ± 0.15	0.83	−0.22 ± 0.20	−0.12
LR	SB_{-3}	−0.81 ± 0.10	−1.72	0.16 ± 0.08	0.89	−0.01 ± 0.20	−0.18
	SB_{-2}	−1.09 ± 0.05	−1.49	0.31 ± 0.12	0.48	−0.17 ± 0.10	0.04
	SB_{-1}	−1.17 ± 0.08	−1.21	0.11 ± 0.11	0.11	−0.0 ± 0.001	0.09
	SB_{+1}	−1.27 ± 0.05	−1.25	0.24 ± 0.06	0.12	−0.00 ± 0.04	0.13
	SB_{+2}	−1.48 ± 0.08	−1.54	0.57 ± 0.11	0.52	−0.16 ± 0.10	0.07
	SB_{+3}	−1.47 ± 0.08	−1.77	0.52 ± 0.14	0.98	−0.05 ± 0.20	−0.20

Two Colour ARPES - Circ. Dichroism - He

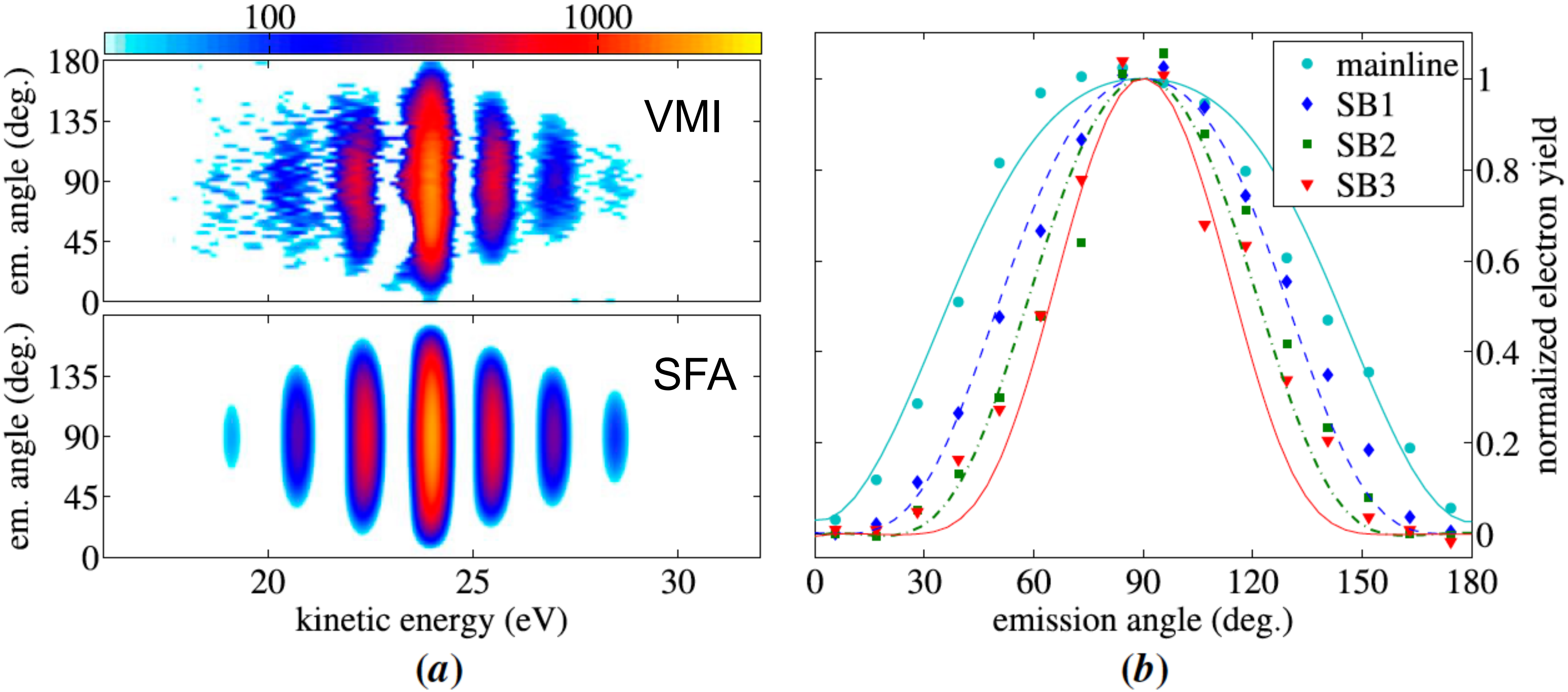
SFA and Perturbation Theory (weak NIR fields)

- The good agreement between experimental values of the asymmetry parameters and both theoretical models for the weak NIR field is remarkable since the SFA and the PT have different initial premises: the SFA completely ignores the ionic field assuming a free photoelectron moving in the NIR field, while the PT treats the NIR field as a perturbation to the intra-atomic interactions.

- From β^{+-}_4 we get the ratio $|D_s|/|D_d| = 1.00 \pm 0.04$ for SB_{+1} and $|D_s|/|D_d| = 1.07 \pm 0.06$ for SB_{-1}, in excellent agreement with the theoretical (PT) values of 1.04 and 1.12 respectively. The result accords with our earlier (angle integrated) measurements at FLASH* for $\omega_X = 48.6$eV (25.5 nm) and $\omega_L = 2.47$ eV (523 nm), where the corresponding extracted ratio was $|Ds|/|Dd| = 0.95 \pm 0.15$ for the SB_{+1} - *Phys. Rev. Letts* **101** 193002 (2008)

Two Colour ARPES - Circ. Dichroism - He

SFA and Perturbation Theory (strong NIR fields) - 7 x 10^{12} $W.cm^{-2}$



Co-rotating XUV and NIR fields

$Sin^{2n}(\theta)$ dependence* – Ang. distribution peaked at $\theta=90^{O}$ and narrows 'n'

*P. Lambropoulos Phys. Rev. Lett **28** pp585–587 (1972)

Two Colour ARPES - Circ. Dichroism - He

Circular Dichroism at emission angle θ = 90°: SFA (weak and strong NIR fields)

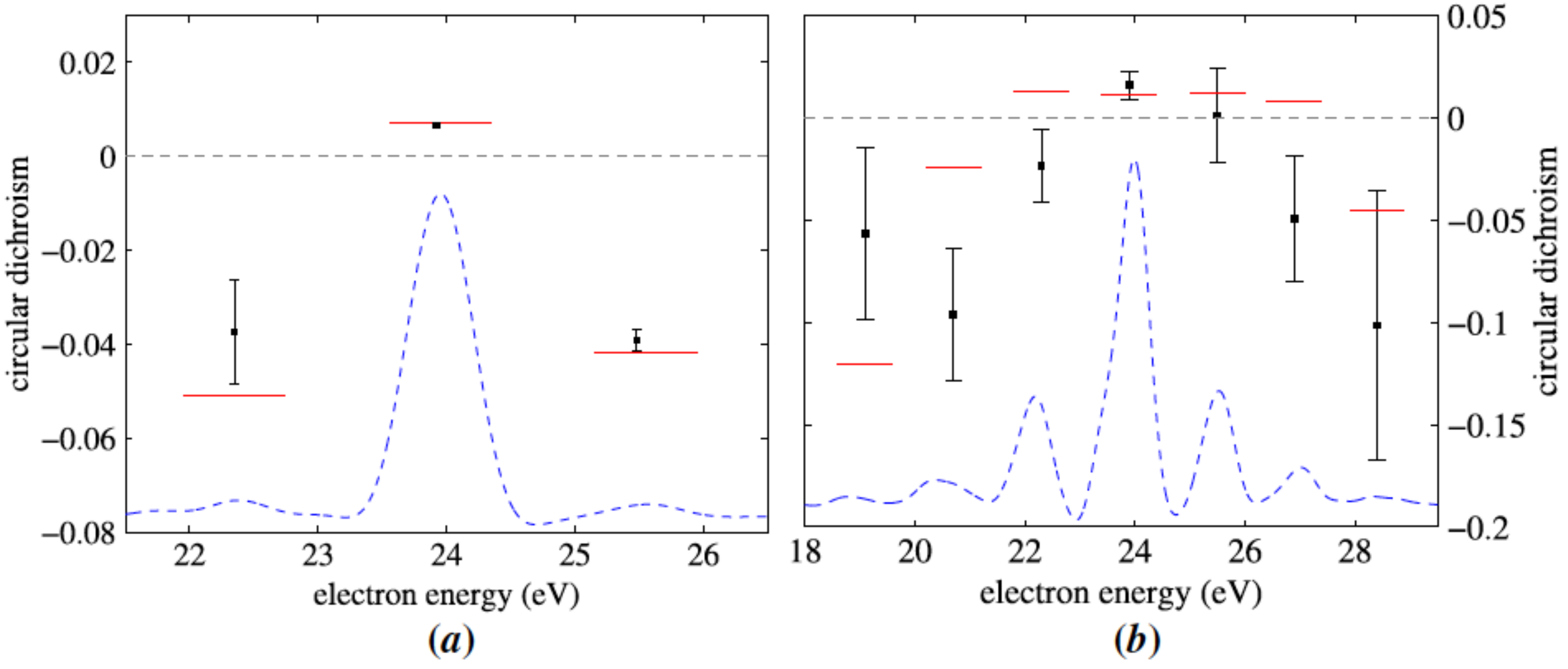


Figure 8. Experimentally determined CDADs at 90° emission angle for the central line and the sidebands for the NIR intensity of 3×10^{11} W/cm^2 (*a*) and 7.2×10^{12} W/cm^2 (*b*) are shown by dots with error bars. The dashed lines represent the experimental electron spectra (the zero-line is shifted for clarity). The CDADs extracted from the simulated spectra are shown by red bars (see text for details).

$$\mathcal{A}_{\vec{k}}^{++} = -i\int_{-\infty}^{\infty} \mathrm{d}t\,\tilde{\mathcal{E}}_X(t)\,d_{sp}\,Y_{1,+1}(\theta_0(t),\phi_0(t)) \times e^{i\Phi(\vec{k},t)}e^{i(E_b-\omega_X)t},$$

$$\mathrm{CDAD} = \frac{|\mathcal{A}_{\vec{k}}^{++}|^2 - |\mathcal{A}_{\vec{k}}^{+-}|^2}{|\mathcal{A}_{\vec{k}}^{++}|^2 + |\mathcal{A}_{\vec{k}}^{+-}|^2}$$

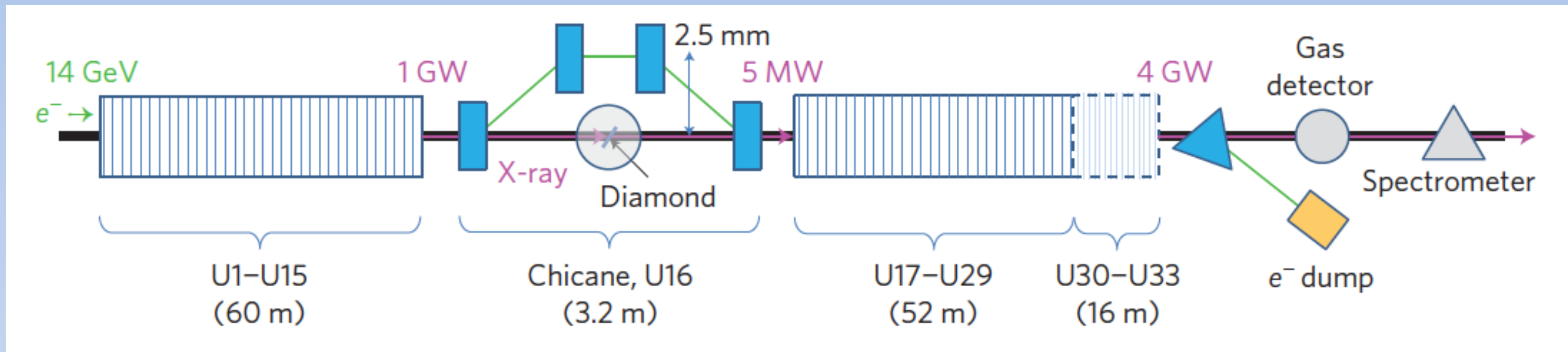
2C ARPES - Circ. Dichroism - Summary

- PAD measurements at low intensity provide the partial s- and d-amplitudes and their relative phases, including the dynamic phase induced by the multi-photon mechanism of the process - complete experiment in two-colour two-photon above threshold ionization.
- By changing the helicity of the NIR pulses, we have confirmed the existence of CD in angular distributions in two-colour multi-photon ionization.
- Calculated CDs agree well with the experiment.
- In the future, investigations can be extended to resonant phenomena in the multi-photon regime as well as to processes in the near-threshold region.

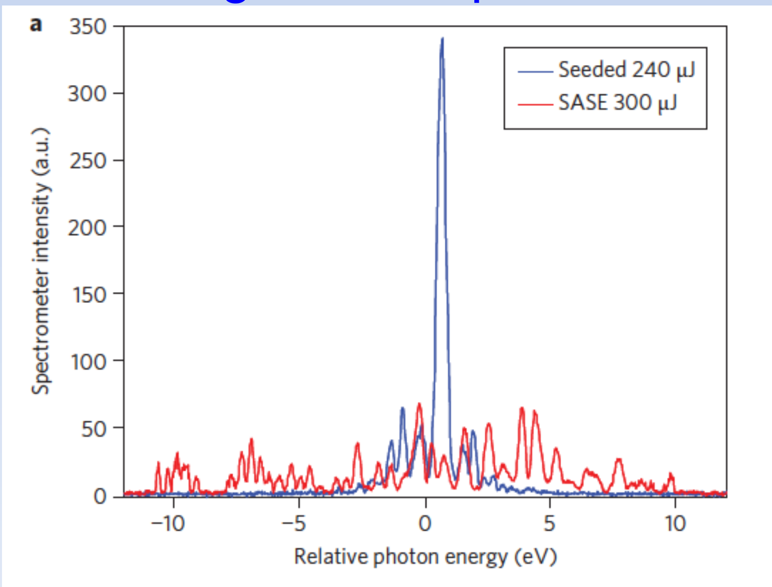
Next Steps

1. Rudiments of ionization processes in intense laser fields
2. Photoionization experimental setups (FLASH & DESY)
3. One colour – two photon ionization
4. Two colour Ionization
5. **Some perspectives**

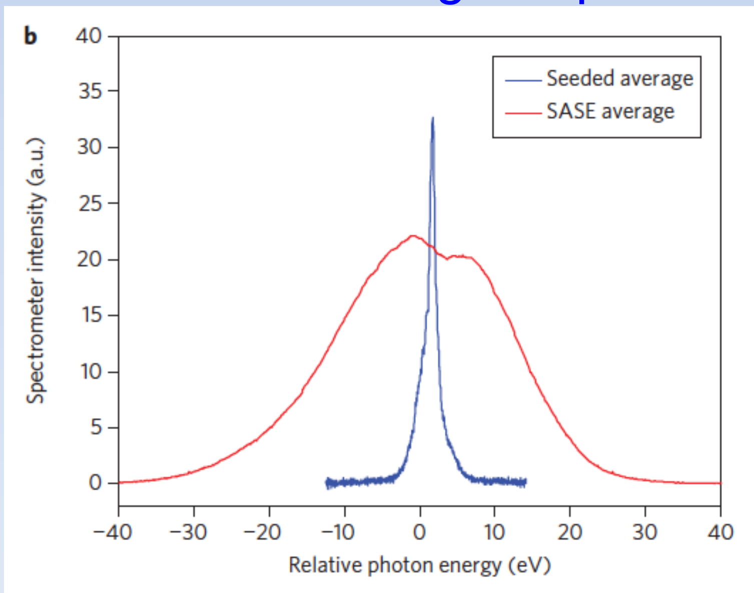
Self - Seeded FELs, e.g., LCLS…..



Single-Shot Spectra

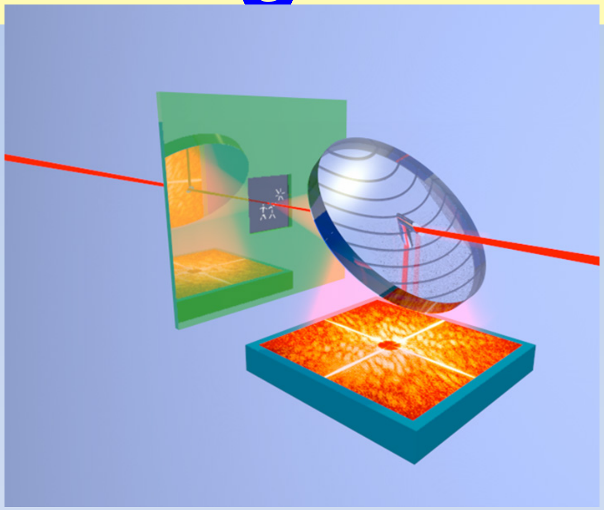


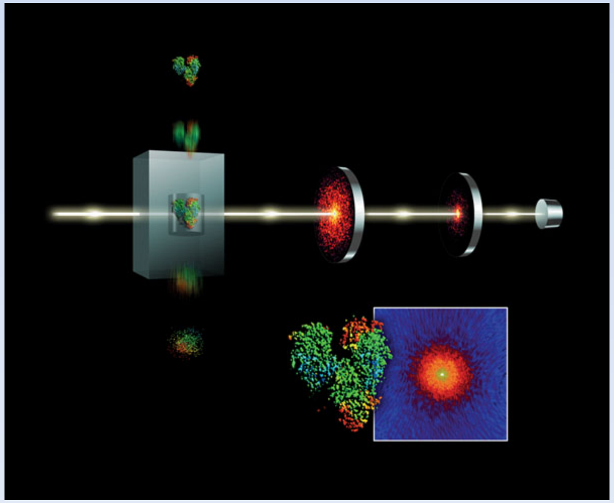
Multi-Shot Averaged Spectra

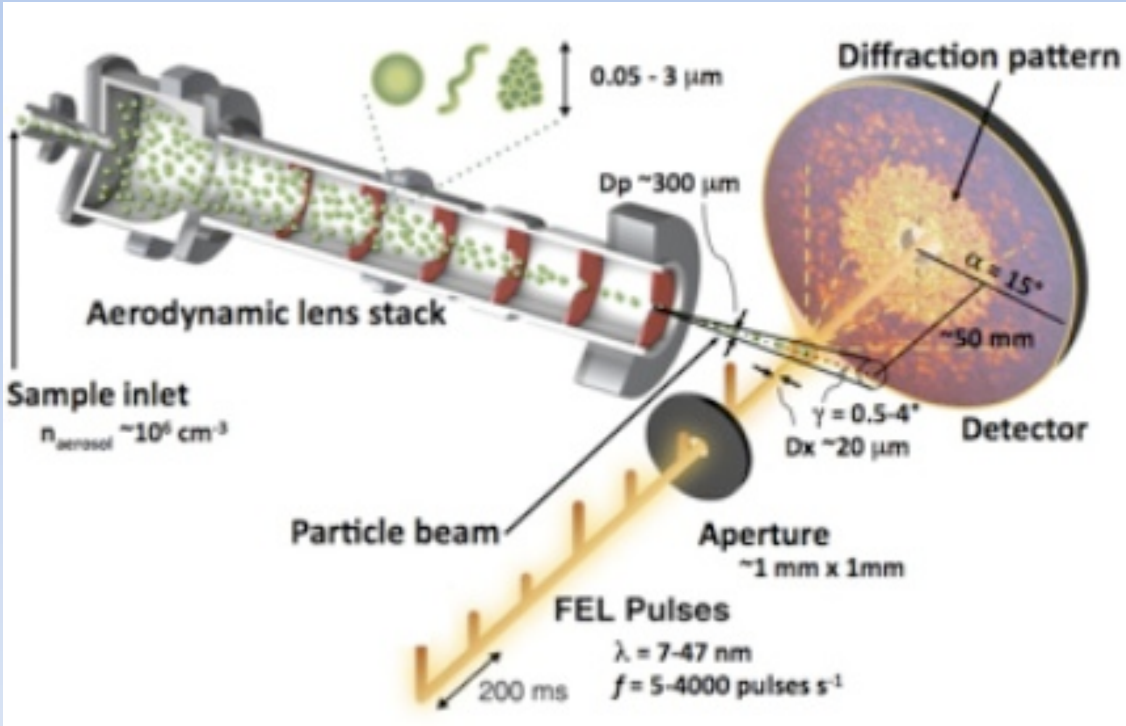


Lutman et al., PRL 113 Art. No. 254801 (2014)/Amann et al. *Nature Photonics* **6**, 693 (2012)

Imaging, micro-protein crystals, single viruses, even molecules !!!







Single shot dynamic coherent diffraction imaging on femtosecond timescales - soon to be used in single biomolecular imaging to make molecular movies !!!

Cf: CFEL, DESY, LCLS and PULSE-Stanford

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Science Foundation Ireland – Investigator Programme – 12/IA/1742 & 07/IN.1/I1771

Irish Research Council (PhD Scholarships / Postdoctoral Fellowships)

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In Conclusion

1. To date we have looked only at one and two colour non-resonant photoionization processes
2. Now – FELs seeded and easily tunable - we can explore resonant processes where inner shell electrons dominate

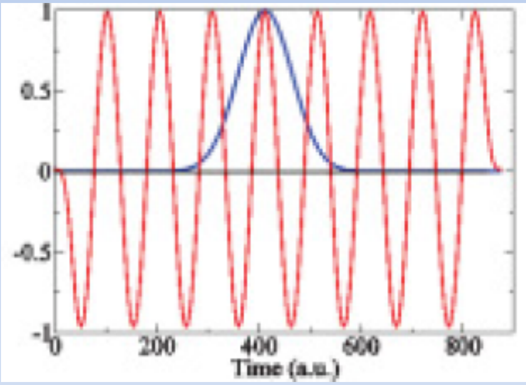
Next steps (XFEL Technology): X-CPA

XFELs are finally becoming real lasers – truly monochromatic, fully phase coherent, collimated…... If it can be done with an optical laser – we can now propose it for XFELs….

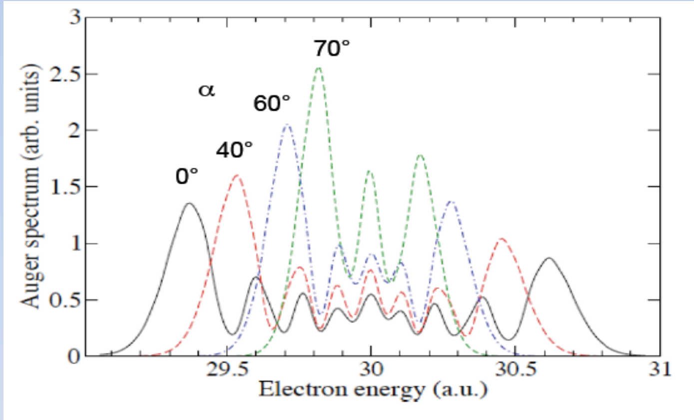
But what about the intermediate (few optical cycle) regime ?

Based on theoretical work by: Nikolay Kabachnik et al., Moscow State Univ.

Auger lifetime similar to optical (800 nm) cycle



Angle Resolved Sideband Spectra



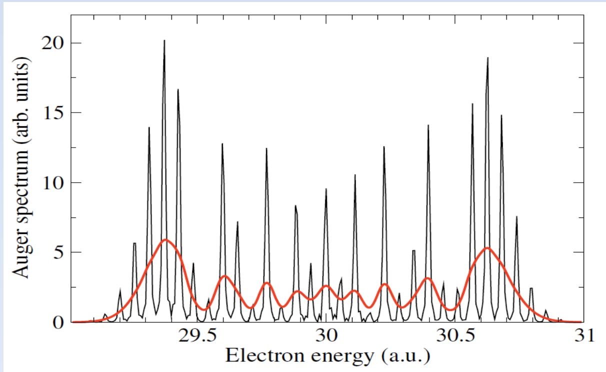
Core hole lifetime
τ (Ne 1s) = 2.4fs

Optical cylce
T (800nm) = 2.6fs

LCLS: 1 keV, 2-5 fs

NIR: 800 nm,
1 x 10^{12} W/cm^2

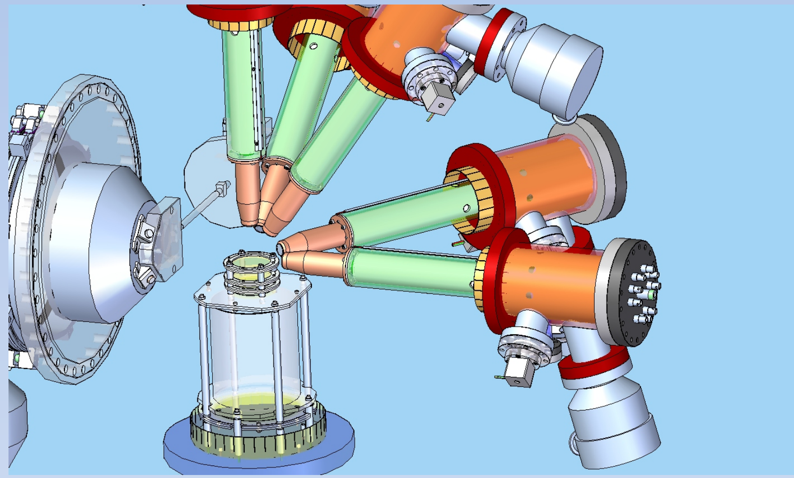
Simulated spectrum for electron emission in the direction of the field (0°)



A.K. Kazansky, N.M. Kabachnik, JPB 42, 121002 (2009)
A.K. Kazansky, N.M. Kabachnik, JPB 43, 035601 (2010)

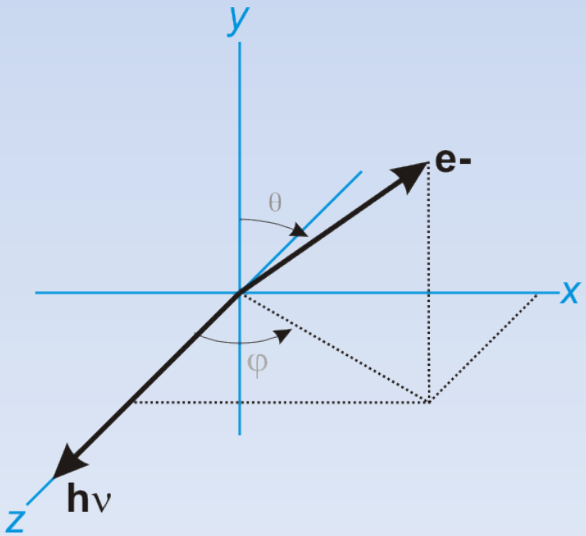
AMO Chamber and Specifications

High Field Chamber (AR-eTOF)



1. Based on a successful design used by the Denis Lindle (RIP) group at ALS – designed for up to 5keV electrons
2. Transmission flat for E_{kin}> 20 eV
3. E/ΔE up to 5,000

•	θ	φ	comment
1	0º	90º	Along y-axis
2	35.3º	90º	Magic angle in xy dipole plane
3	90º	90º	Along x-axis
4	54.7º	0º	Non-dipole
5	90º	35.3º	Non-dipole



lcls.slac.stanford.edu

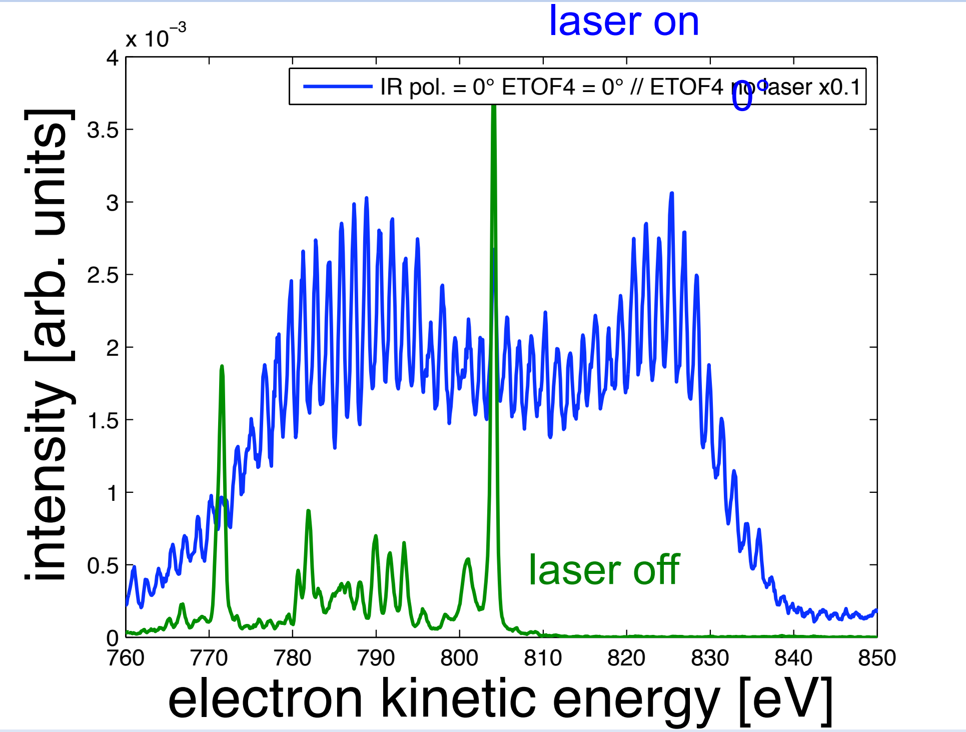
SB modulation – few/sub-optical cycle effects

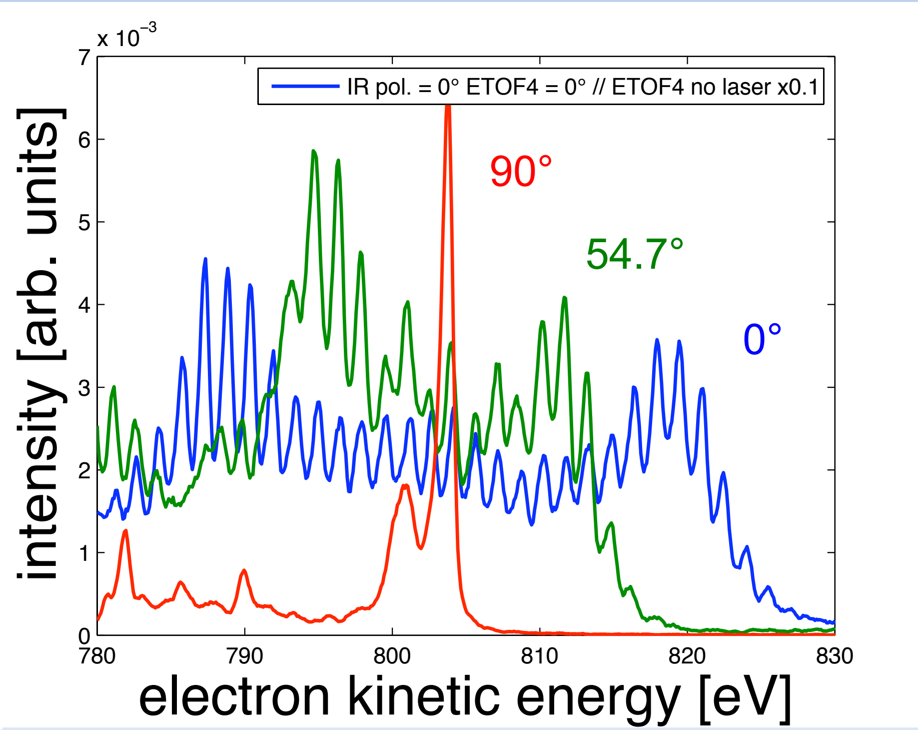
LCLS: 1 keV, “4fs”, 20pC bunch current

NIR : 800nm, 1 mJ, 3ps

1 x 10^{12} W/cm^2

6 x 10^{11} W/cm^2





Strong sideband structure

Strong angular effect

NCPST

EXTATIC

Phys. Rev. Lett **108** 063007 (2012)

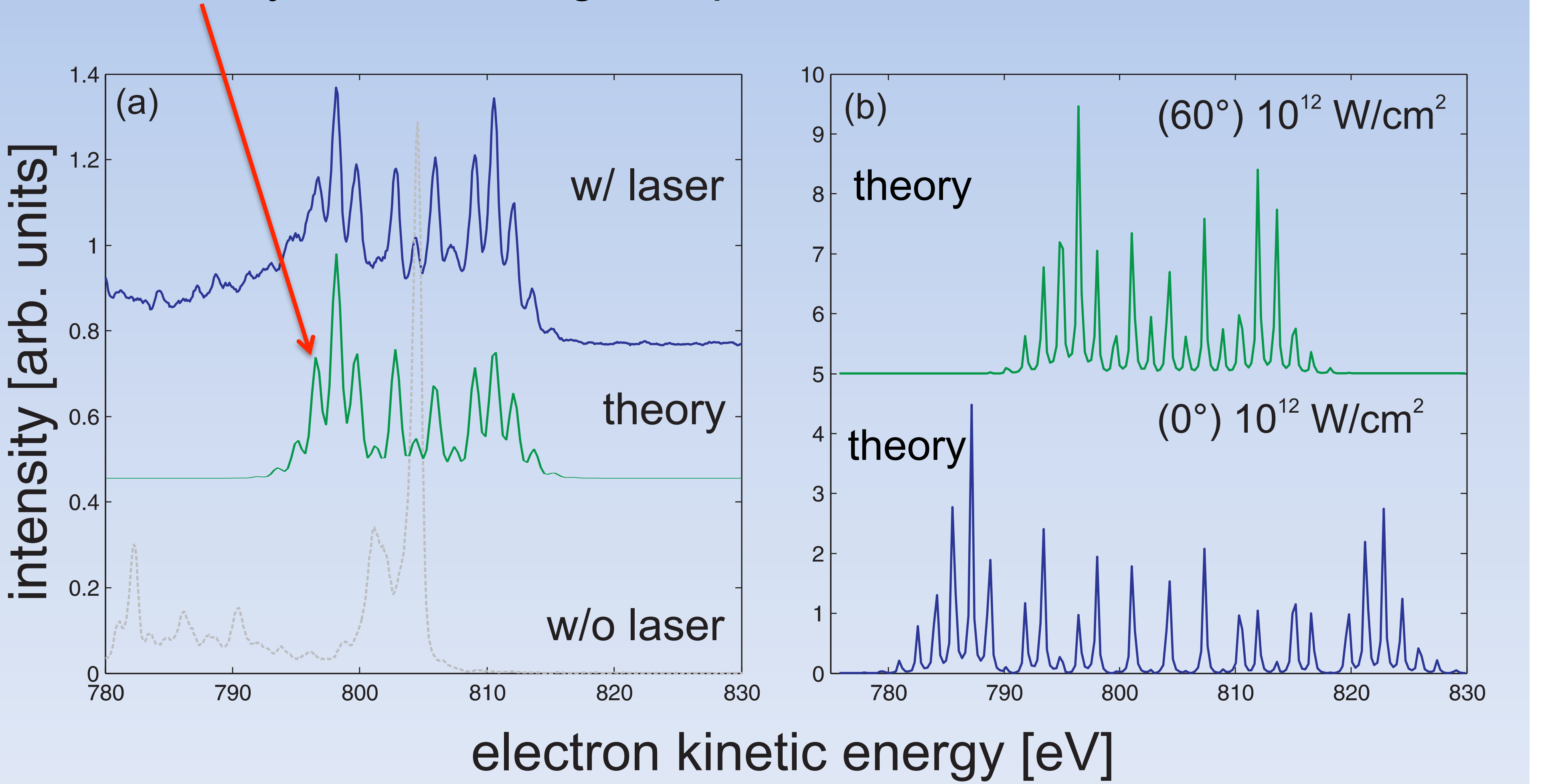
sfi inspire

science foundation ireland Integrated NanoScience Platform for Ireland

DCU

SB modulation – few/sub-optical cycle effects

Theory – accounting for spatial variation of the laser field



NCPST EXTATIC

Phys. Rev. Lett **108** 063007 (2012)

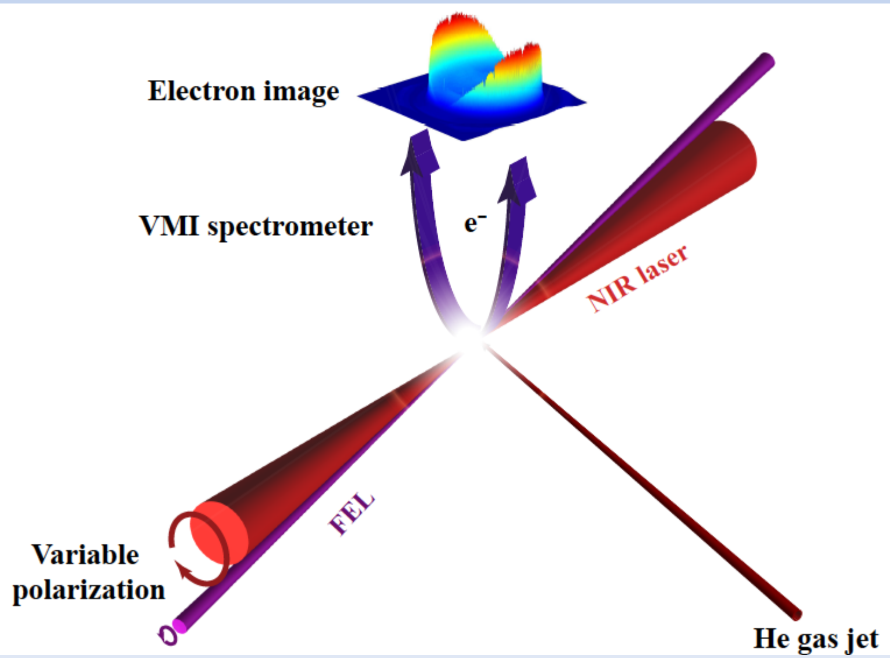
sfi inspire DCU

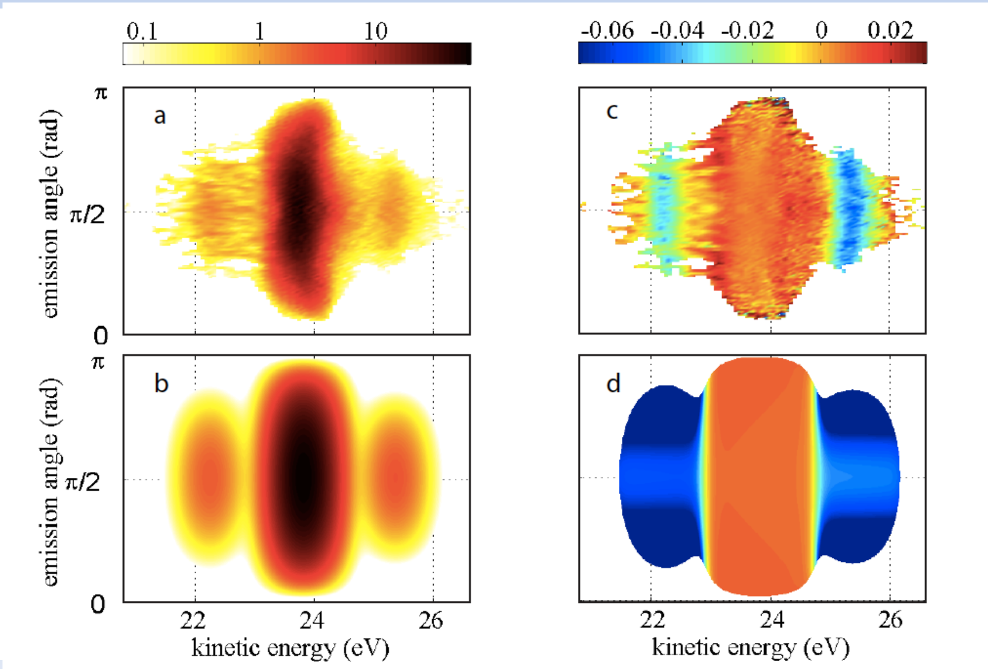
Measuring Polarisation of XFELs

T Mazza et al. (XFEL GmbH, DESY, FERMI@ELECTTRA, DCU, MSU, etc)

Theory - Kazansky, A. K., Grigorieva, A. V. and Kabachnik, N. M. Circular Dichroism in Laser-Assisted Short Pulse Photoionization. Phys. Rev. Lett. 107, 253002 (2011).

DDCS (Expt./Th.) | DDCS (Expt./Th.) | CD (L-R/L+R.)





Nature Comms **5** Art. No. 3628 (2014)