Recent results from the FLASH Free electron LASer (FEL) at Hamburg on one (XUV) and two (XUV+IR) colour photoionisation

John T Costello

National Centre for Plasma Science & Technology (NCPST)/
School of Physical Sciences, Dublin City University

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

NCPST EXTATIC

Japan-Ireland ISCA
UCD, Nov 4 2016

sfi science foundation ireland 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 Postdoc(s): Dr. Pramod Pandey

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

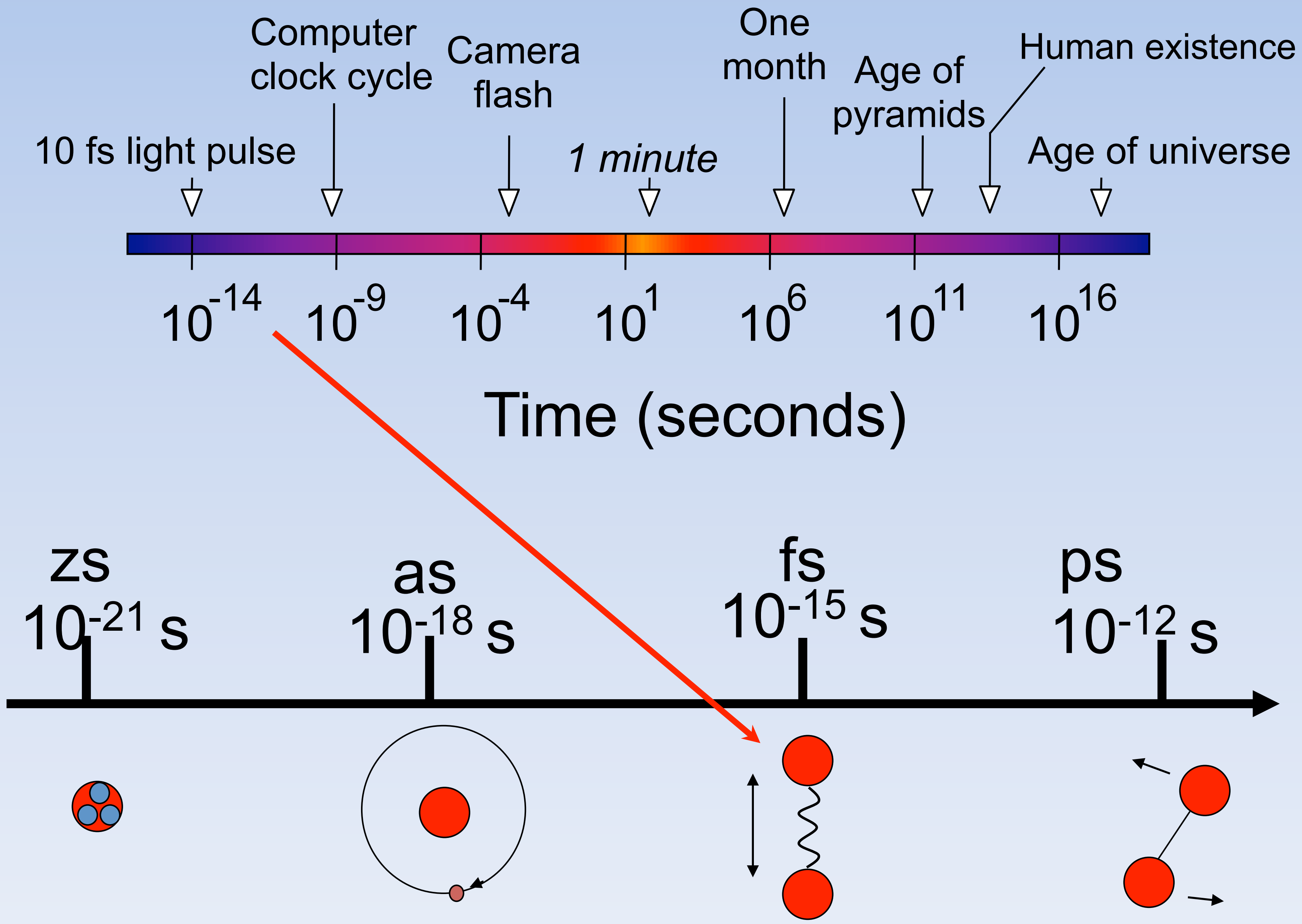
Kurchatov Inst., Moscow: V. L. Nosik

Univ Basque Country: A K Kazansky

DCU: T. J. Kelly, N. Walsh & J. T. Costello

Thanks to AG Photon & AG Machine at FLASH-Hamburg

TIMESCALES - HOW FAST IS FAST ?



10 fs light pulse

Computer clock cycle

Camera flash

1 minute

One month

Age of pyramids

Human existence

Age of universe

10^{-14} 10^{-9} 10^{-4} 10^{1} 10^{6} 10^{11} 10^{16}

Time (seconds)

zs 10^{-21} s

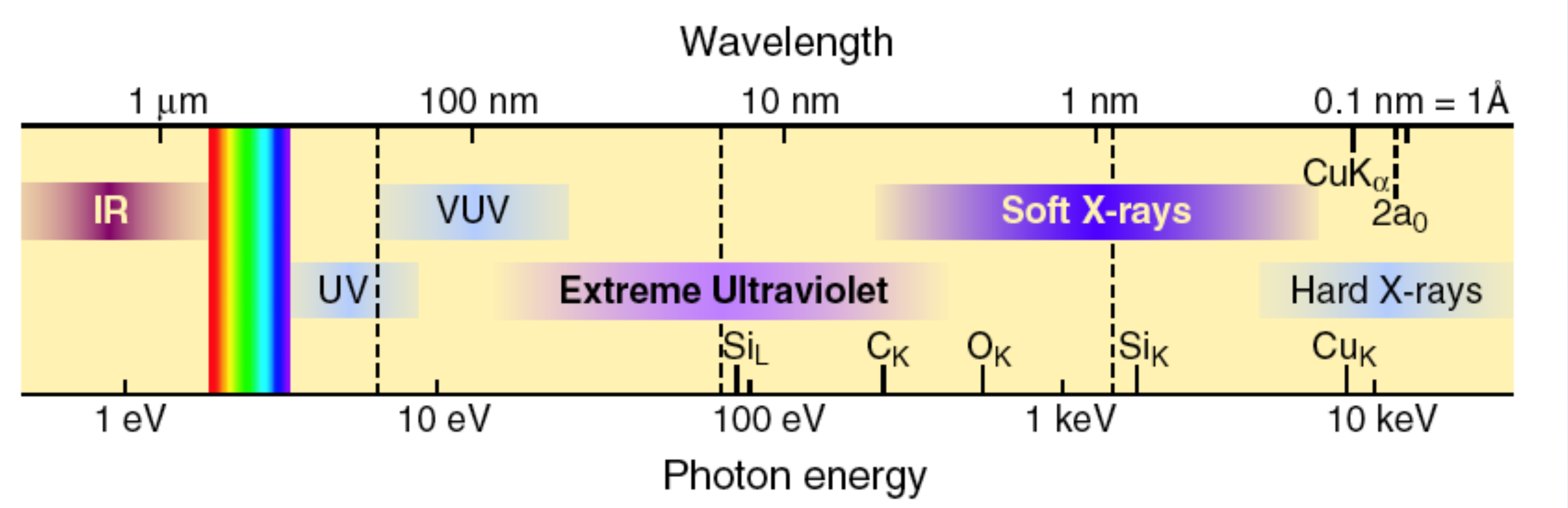
as 10^{-18} s

fs 10^{-15} s

ps 10^{-12} s

X-ray – How X-ray is X-ray ?

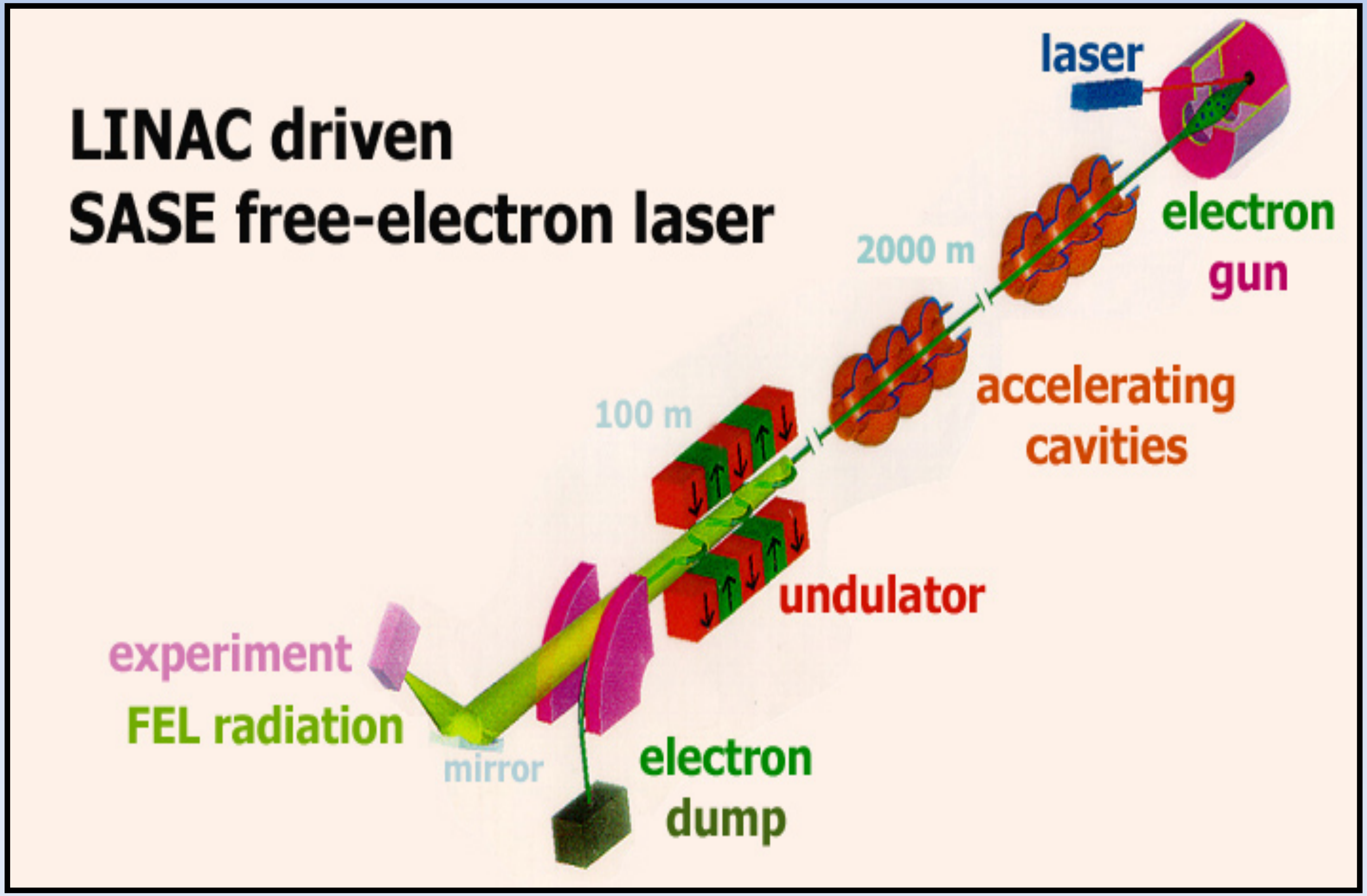
Spectral Range: IR to the X-ray



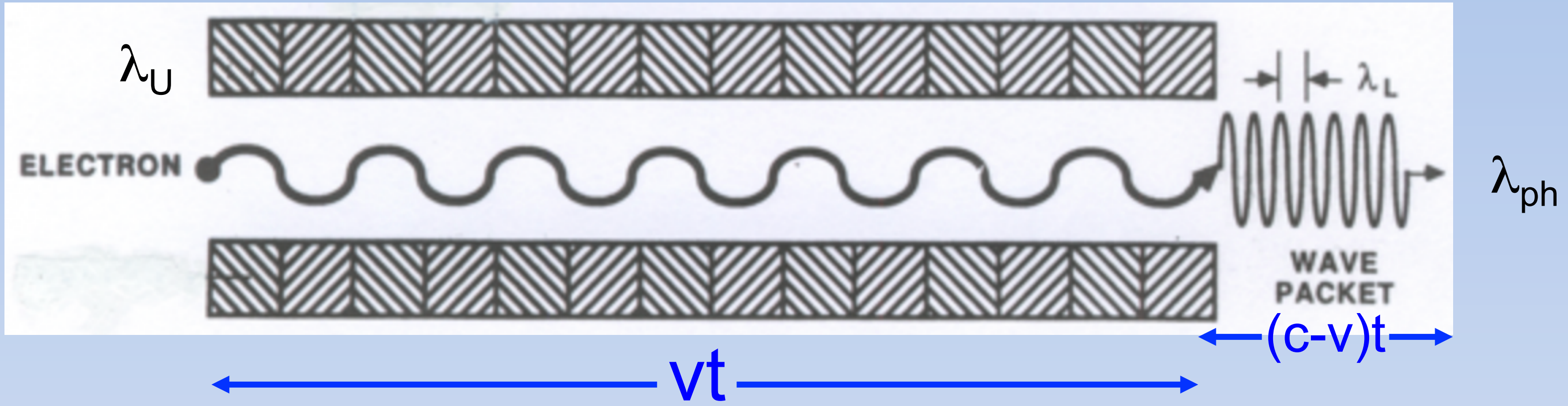
Graphic: Courtesy, Prof. David Attwood (Berkeley)

X-ray Free Electron Lasers (FEL)

Main Components of an X-ray FEL



SASE-FEL, Fundamental Principle



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

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

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

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

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

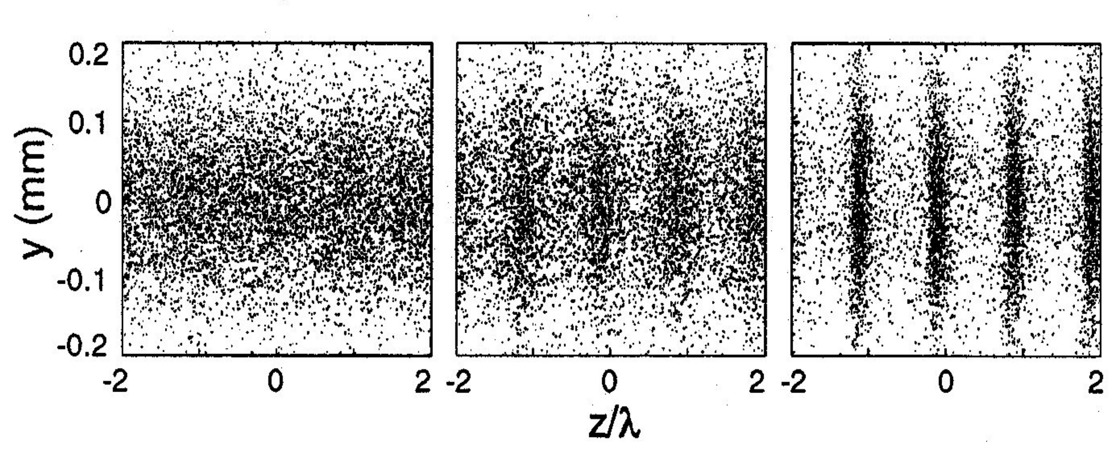
Electron bunch slips behind the lightwave by λ per undulator period

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

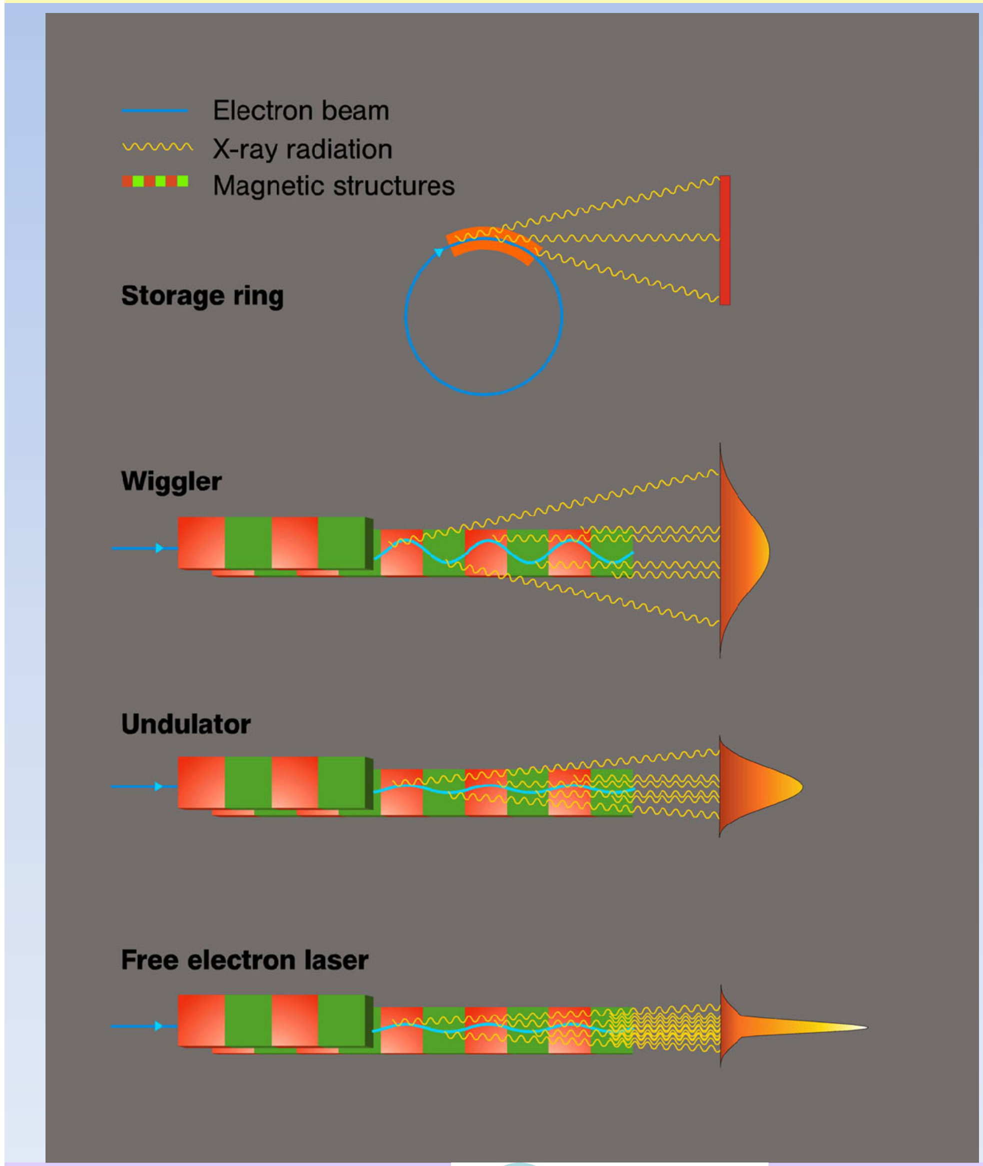
1. Electrons move in slalom-like fashion
2. Electrons emit em waves - forward peaked
3. EM interactions with electrons cause micro-bunching
4. Bunches form 1 λ apart
5. Micro-bunches emit coherently
6. Light grows exponentially with distance along the undulator



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

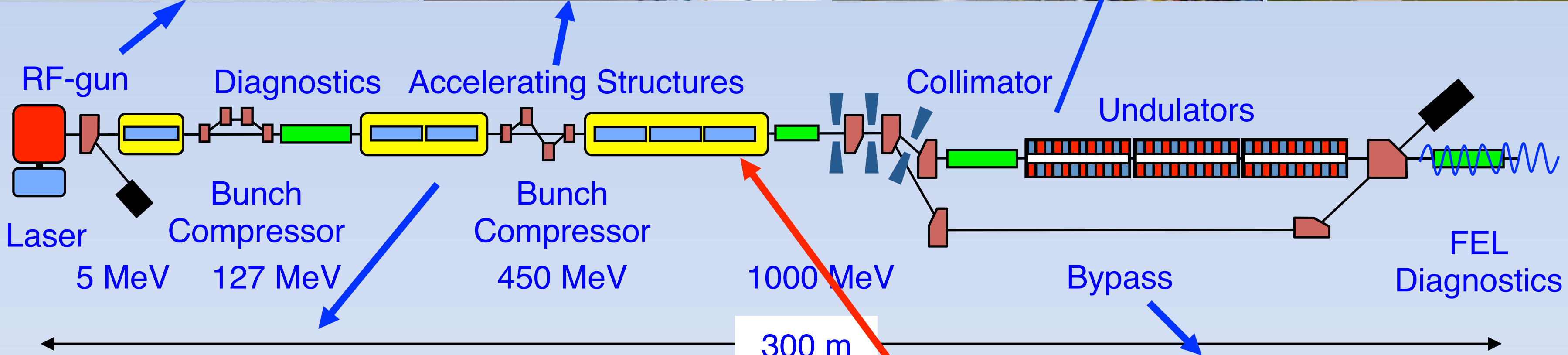
Wiggler:
$I \propto N_W$ x bending magnet

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

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

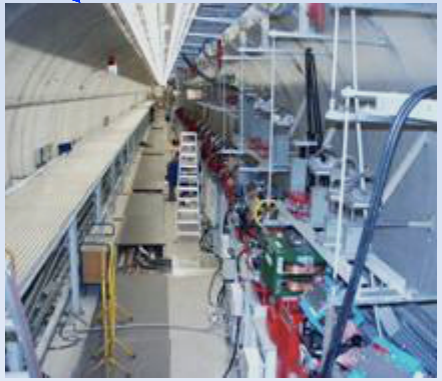
N_U , N_W = # magnetic periods
N_e = # electrons in a bunch

FLASH-Free electron LASer at Hamburg

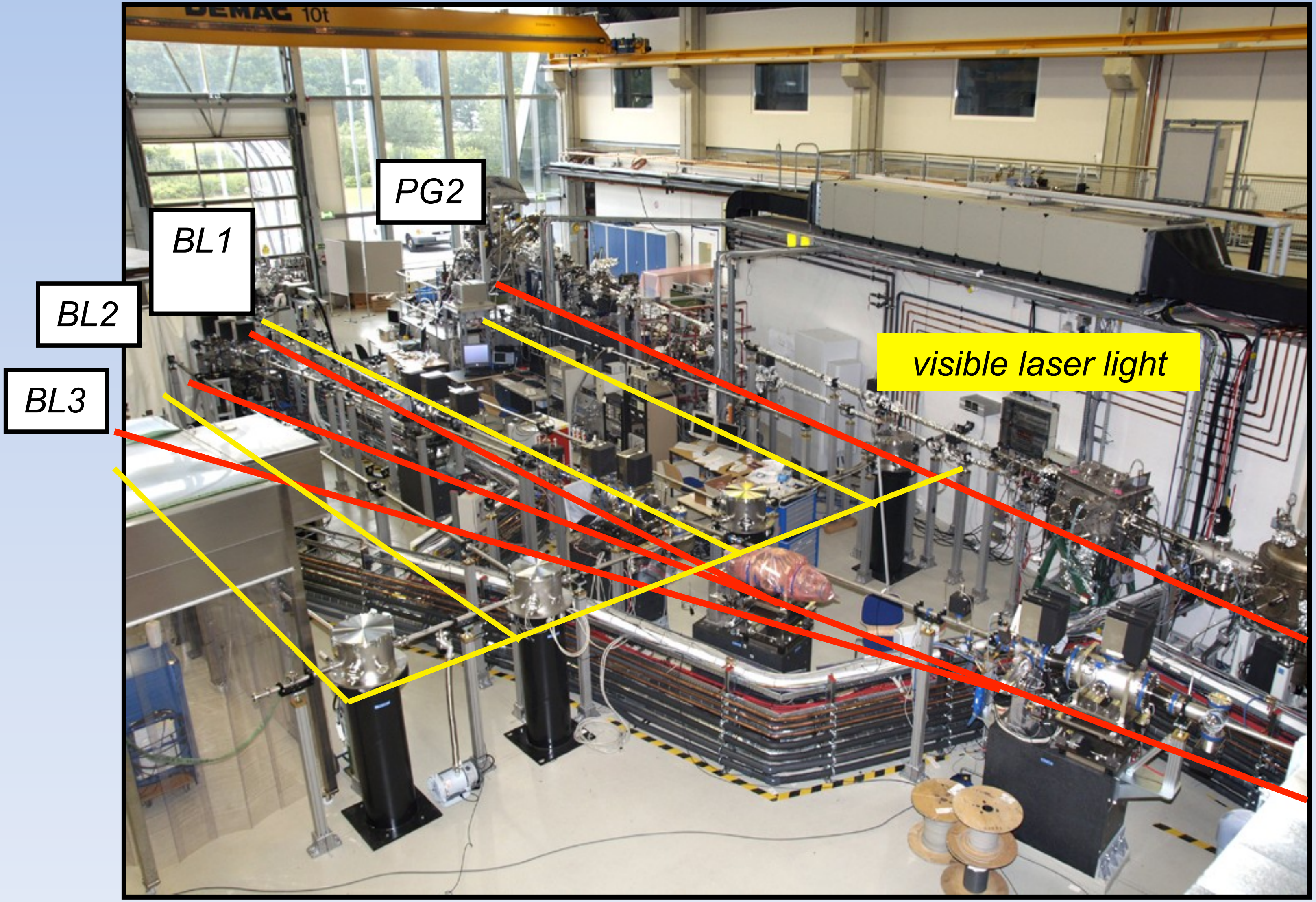


- LINAC Energy : ~ 1 GeV
  ~ 4 – 60 nm

FLASH - Operation & Physical Layout

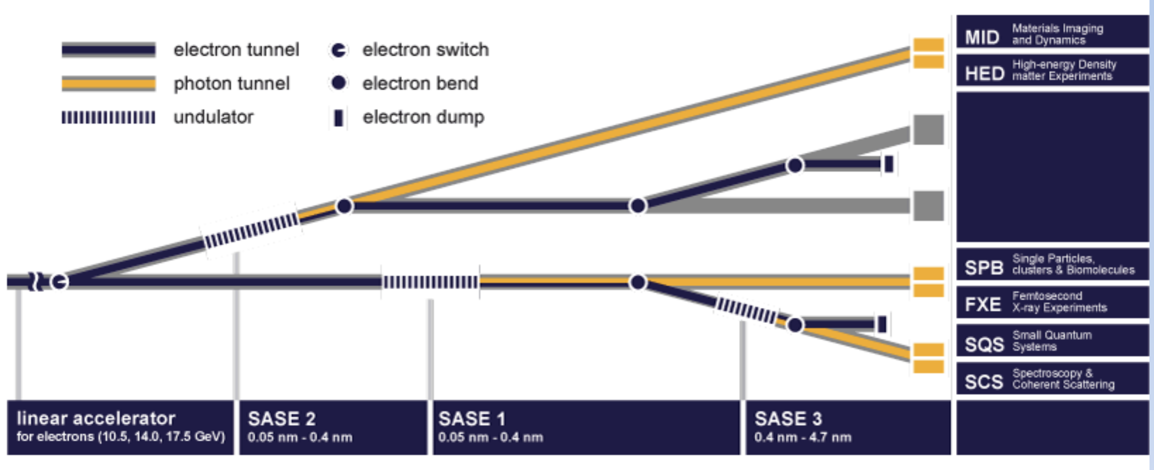


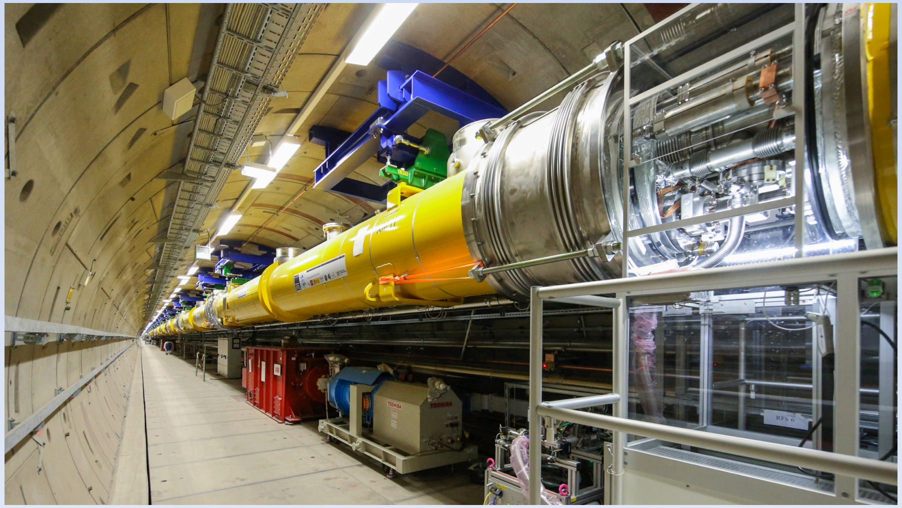
FLASH NIR and EUV Beams (Layout)



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 – 0.5 – 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*

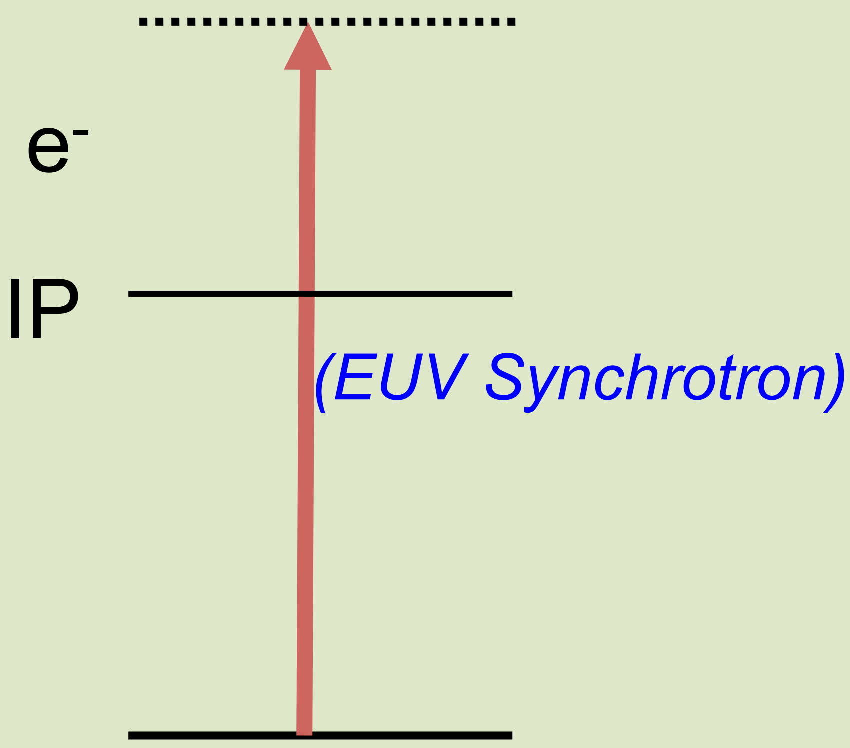
Ionization in Intense Fields

1. **Rudiments of ionization processes in intense laser fields**
2. Photoionization experimental setup (FLASH@DESY)
3. One colour (Ar) – Role of two electrons processes
4. Two colour (He) – ARPES with polarisation control
5. Next Step

The Atomic Photoelectric Effect

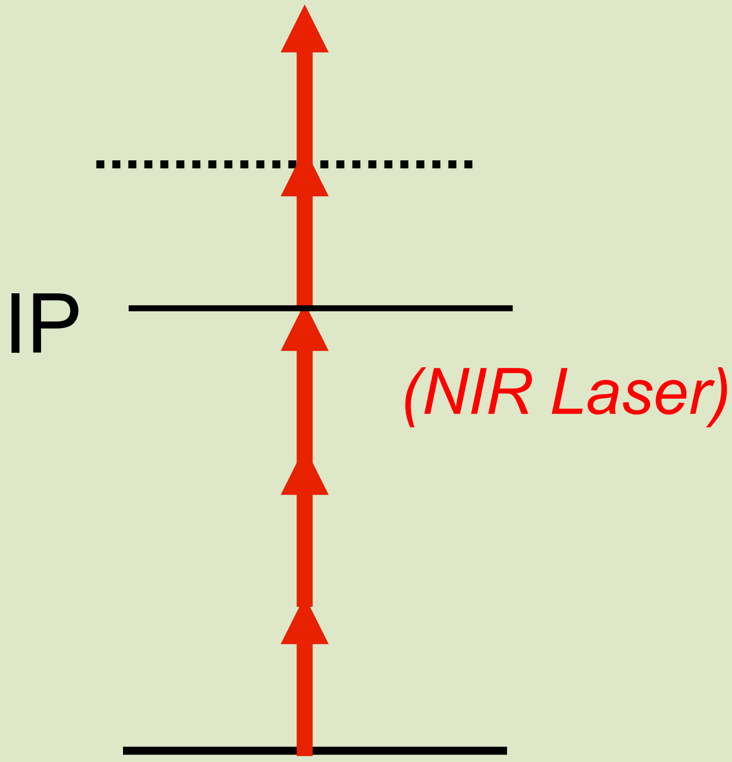
a) Single Photon Ionization (SPI)

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

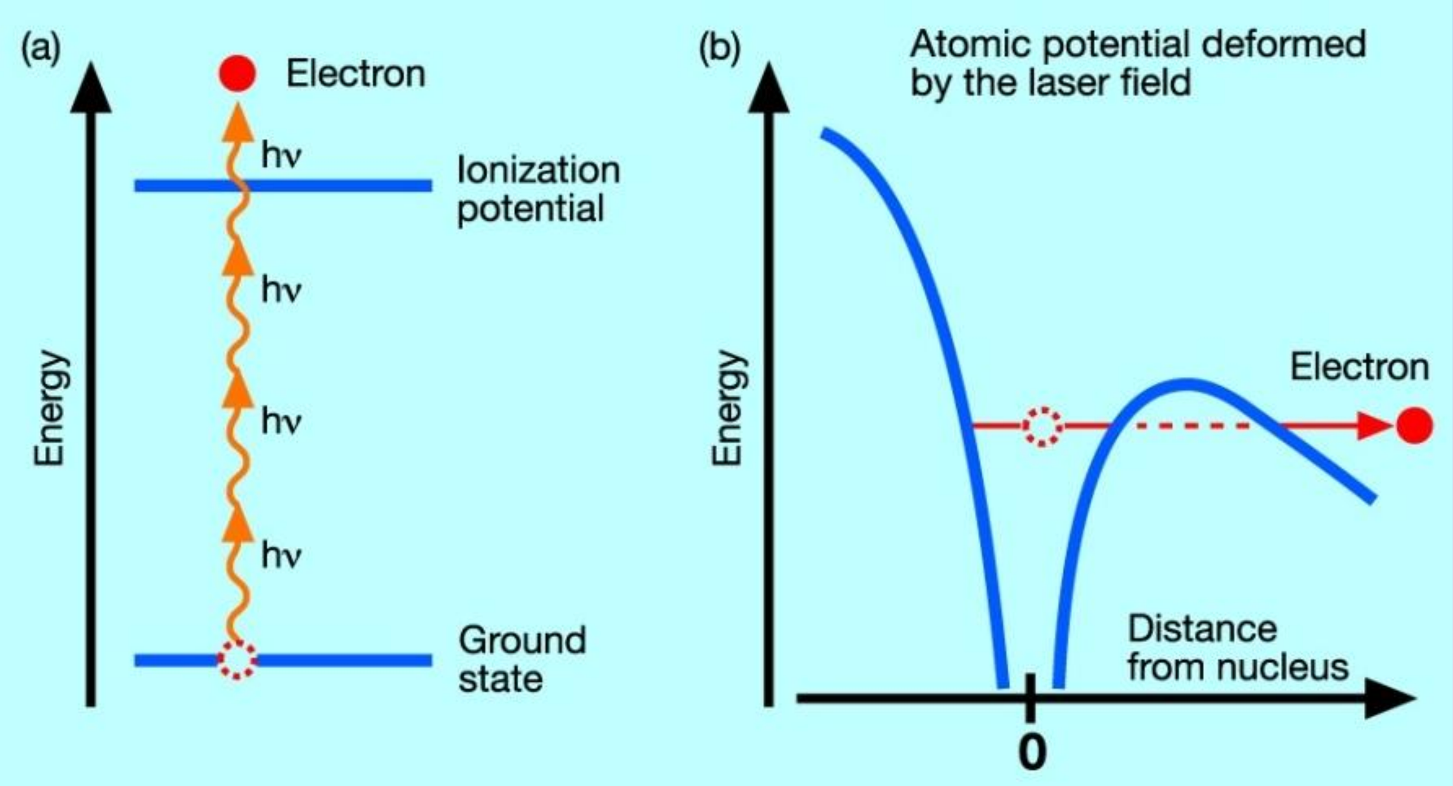


b) Multi Photon Ionization (MPI)

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



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



Intensity/ Wavelength

Photon Energy

How can you determine in which regime the interaction resides ?

Keldysh Parameter

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

IP = Ionization Potential

Up = Ponderomotive Pot.

$$U_P = 9.3\times 10^{-14} I\left(Wcm^{-2}\right) \lambda^2\left(\mu m\right) \quad eV$$

*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**

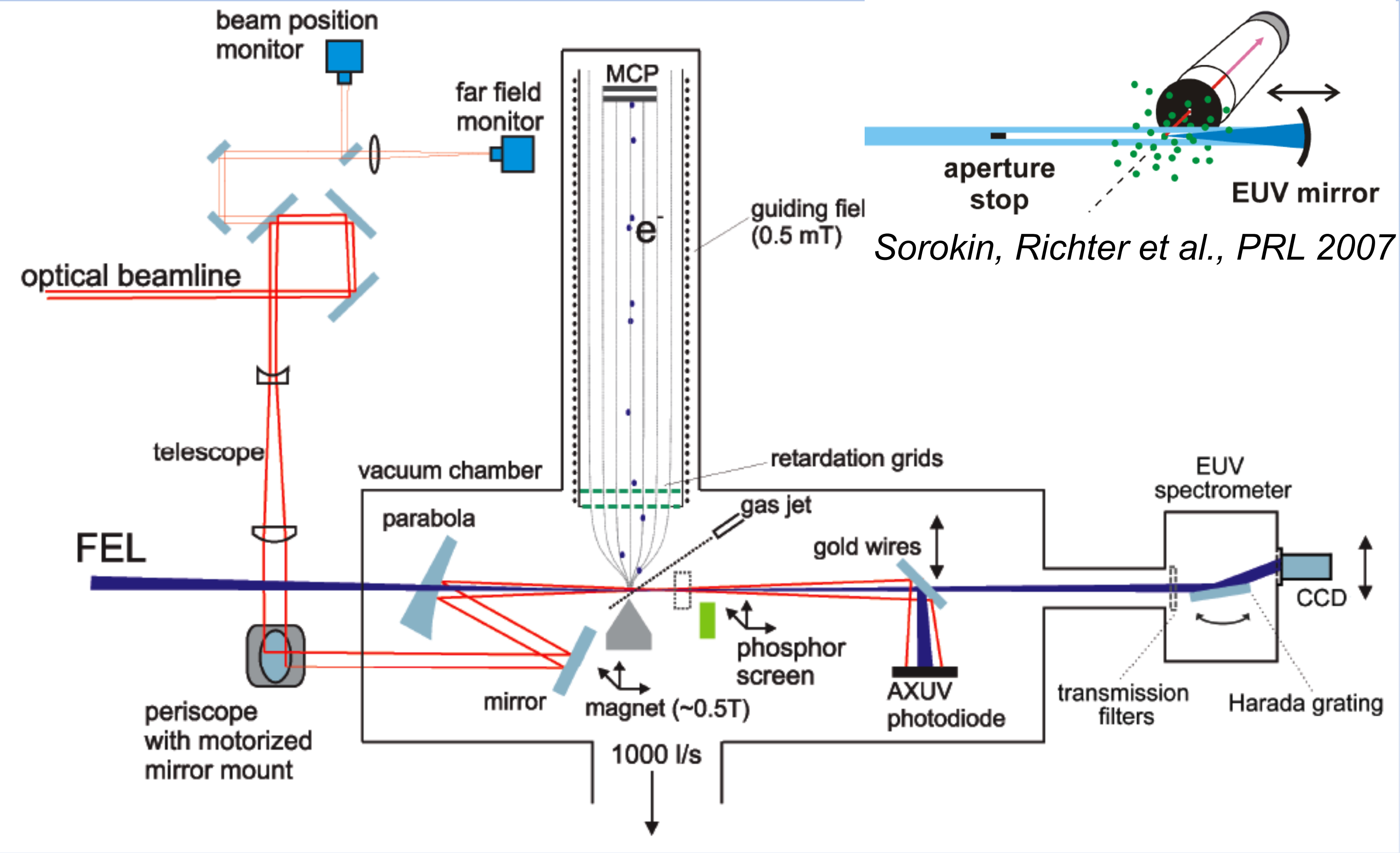
NB2: Re-asserts *primacy of the photon* **over field effects !**

Experimental Setups (DESY & SLAC)

1. Rudiments of ionization processes in intense laser fields
2. **Photoionization experimental setup (FLASH@DESY)**
3. One colour (Ar) – Role of two electrons processes
4. Two colour (He) – ARPES with polarisation control
5. Next Step

Photoelectron Spectroscopy @ FLASH

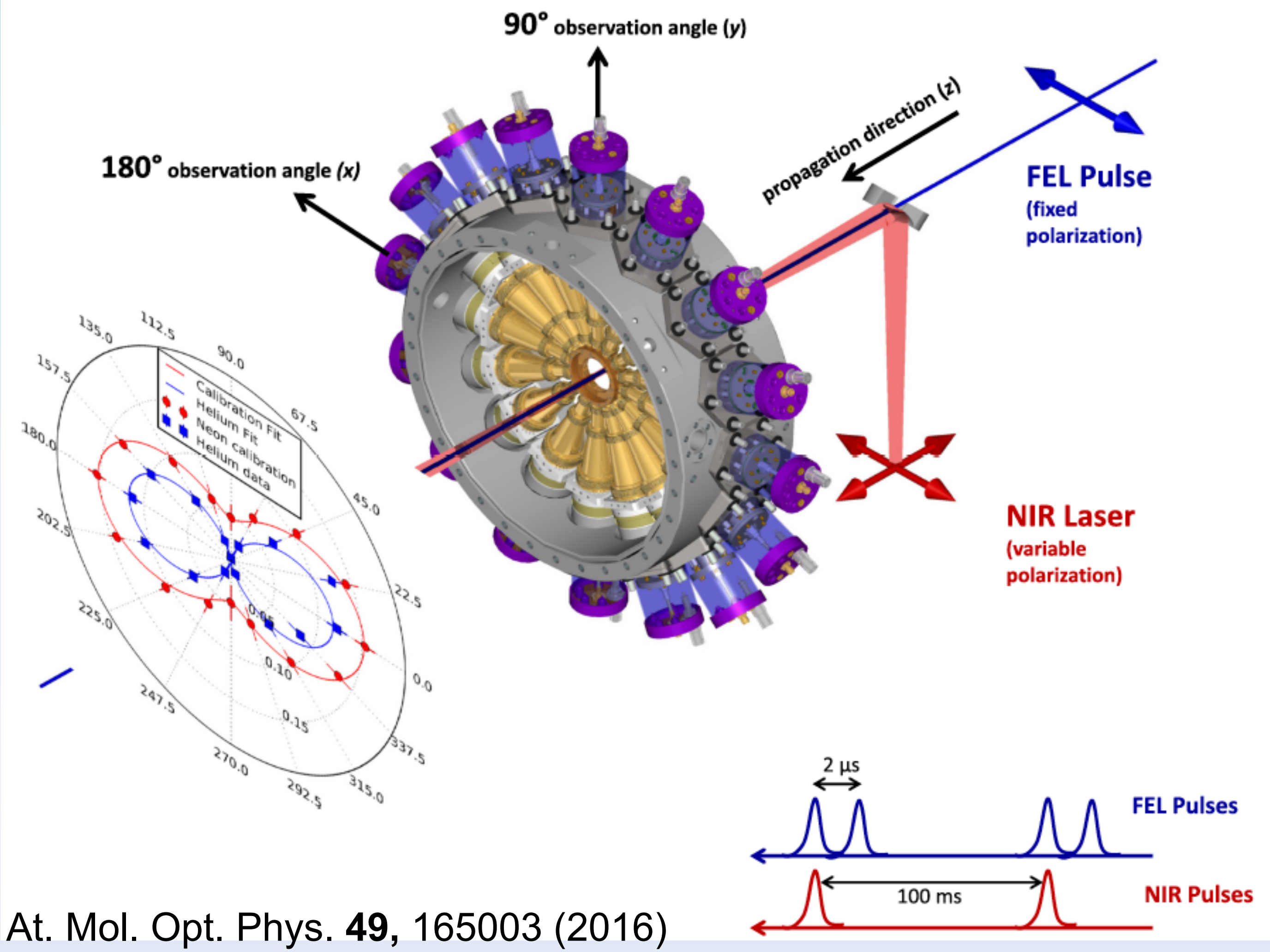
Experimental Layout at FLASH - (EU-RTD)



Sorokin, Richter et al., PRL 2007

Photoelectron Spectroscopy @ FLASH

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



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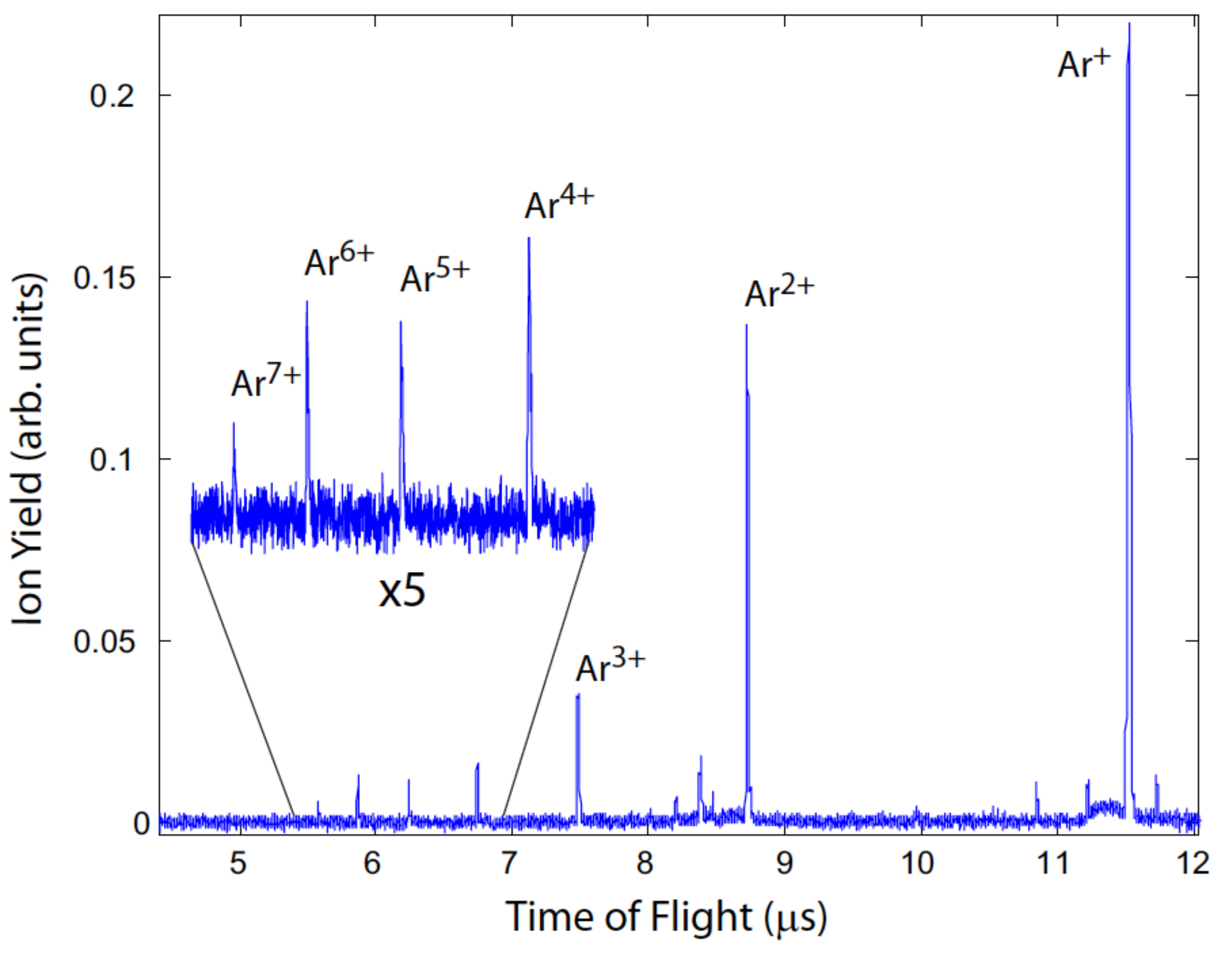
Ionization Ar atoms in an Intense XUV (hν = 105 eV) Field

1. Rudiments of ionization processes in intense laser fields
2. Photoionization experimental setup (FLASH@DESY)
3. **One colour (Ar) – Role of two electron processes**
4. Two colour (He) – ARPES with polarisation control
5. Next Step

Ar ion TOF Trace



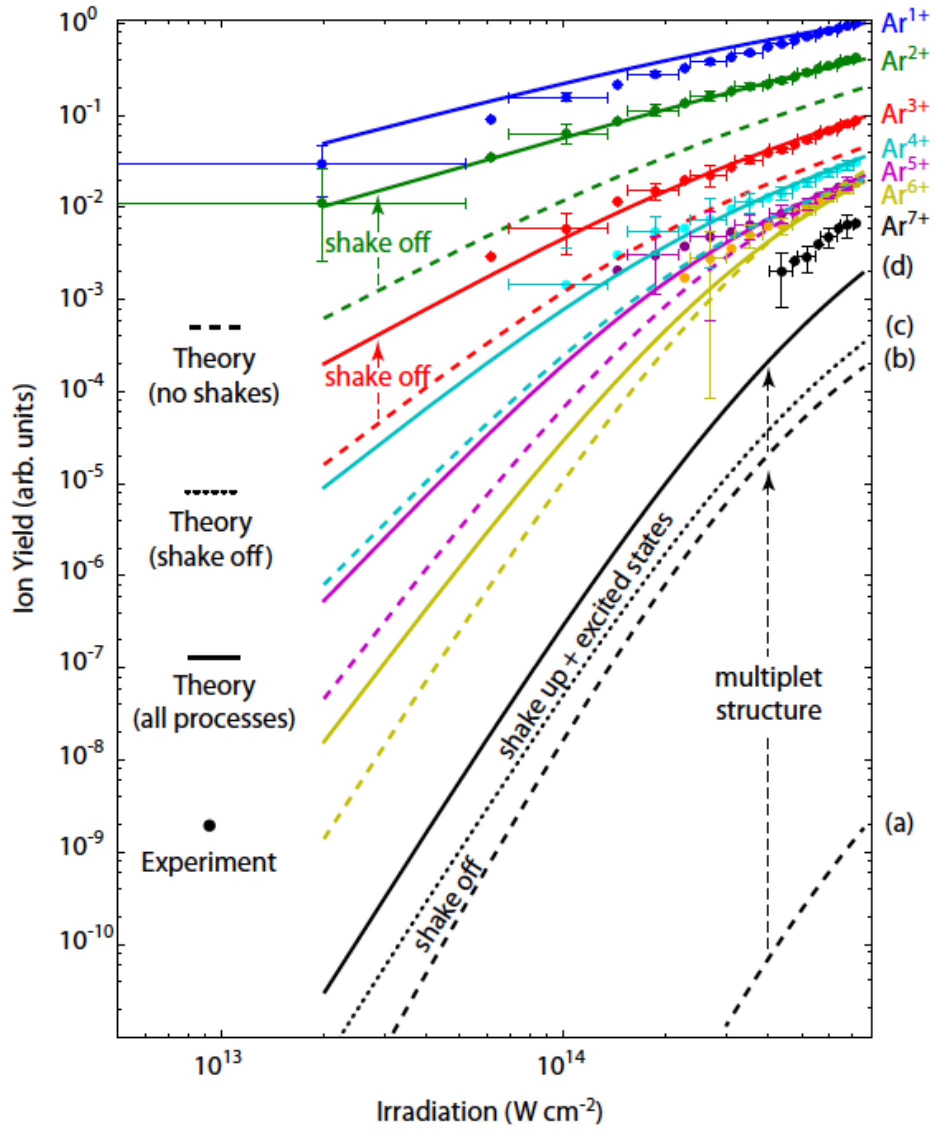
$h\nu$ = 105 eV
BW = 1%
Pulse:
Energy = 16 μJ
Duration = 80 fs
Spot size = 4 μm
P = 0.2 GW
I = 1.25 PW.cm^{-2}
R = 40%
I_{corr} = 0.3 PW.cm^{-2}

Ar ion yield – Experiment and Theory

Experimental (symbols) and theoretical results {model, including (solid lines), and excluding (dashed lines), shake processes} of the relative ionic charge state yield of atomic Ar.

Experimental and theoretical curves are normalized to a relative yield of 1 for Ar^{+} at the highest FEL intensity.

For the Ar^{7+} yield the results of the different theoretical models are indicated (a) including only the electronic ground state, (b) including the Ar^{6+}(3s3p) multiplet structure, (c) adding shake-off processes (dotted line), and (d) adding shake-up processes.



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Ar ion yield – Experiment and Theory

Population rate equations for the various charge state of Ar

$$\frac{dN_0}{dt} = -\left[(\sigma_{10} + \sigma_{10\mathrm{exc}} + \sigma_{10\mathrm{sh}} + \sigma_{20\mathrm{so}})F(t) + \left(\sigma^{(2)}_{\mathrm{ATI}+} + \sigma^{(2)}_{20}\right)F^2(t)\right]N_0,$$

$$\frac{dN_1}{dt} = \left[(\sigma_{10} + \sigma_{10\mathrm{exc}} + \sigma_{10\mathrm{sh}})F(t) + \sigma^{(2)}_{\mathrm{ATI}+}F^2(t)\right]N_0 - \left[\left(\sigma_{21} + \sigma_{21\mathrm{exc}} + \sigma_{21d} + \sigma_{21\mathrm{sh}} + \sigma_{31\mathrm{so}}\right)F(t) + \sigma^{(2)}_{\mathrm{ATI2}+}F^2(t)\right]N_1,$$

$$\frac{dN_2}{dt} = \left[\sigma_{20\mathrm{so}}F(t) + \sigma^{(2)}_{20}F^2(t)\right]N_0 + \left[(\sigma_{21} + \sigma_{21\mathrm{exc}} + \sigma_{21d} + \sigma_{21\mathrm{sh}})F(t) + \sigma^{(2)}_{\mathrm{ATI2}+}F^2(t)\right]N_1 - (\sigma_{32} + \sigma_{32\mathrm{exc}} + \sigma_{32d} + \sigma_{32\mathrm{sh}})F(t)N_2,$$

$$\frac{dN_3}{dt} = \sigma_{31\mathrm{so}}F(t)N_1 + (\sigma_{32} + \sigma_{32\mathrm{exc}} + \sigma_{32d} + \sigma_{32\mathrm{sh}})F(t)N_2 - (\sigma_{43} + \sigma_{43\mathrm{exc}} + \sigma_{43d} + \sigma_{43\mathrm{sh}})F(t)N_3,$$

$$\frac{dN_4}{dt} = (\sigma_{43} + \sigma_{43\mathrm{exc}} + \sigma_{43d} + \sigma_{43\mathrm{sh}})F(t)N_3 - (\sigma_{54} + \sigma_{54\mathrm{exc}} + \sigma_{54d} + \sigma_{54\mathrm{sh}})F(t)N_4,$$

$$\frac{dN_5}{dt} = (\sigma_{54} + \sigma_{54\mathrm{exc}} + \sigma_{54d} + \sigma_{54\mathrm{sh}})F(t)N_4 - (\sigma_{65} + \sigma_{65\mathrm{exc}} + \sigma_{65d} + \sigma_{65\mathrm{sh}})F(t)N_5,$$

$$\frac{dN_6}{dt} = (\sigma_{65} + \sigma_{65\mathrm{exc}} + \sigma_{65d} + \sigma_{65\mathrm{sh}})F(t)N_5 - \left[(\sigma_{76d} + \sigma_{76\mathrm{sh}})F(t) + \sigma^{(2)}_{76}F^2(t) + \sigma^{(3)}_{86}F^3(t)\right]N_6,$$

$$\frac{dN_7}{dt} = \left[(\sigma_{76d} + \sigma_{76\mathrm{sh}})F(t) + \sigma^{(2)}_{76}F^2(t)\right]N_6 - \sigma^{(2)}_{87}F^2(t)N_7,$$

$$\frac{dN_8}{dt} = \sigma^{(3)}_{86}F^3(t)N_6 + \sigma^{(2)}_{87}F^2(t)N_7,$$

N_i denotes the population of an ion with charge +i.

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Ar ion yield – Experiment and Theory

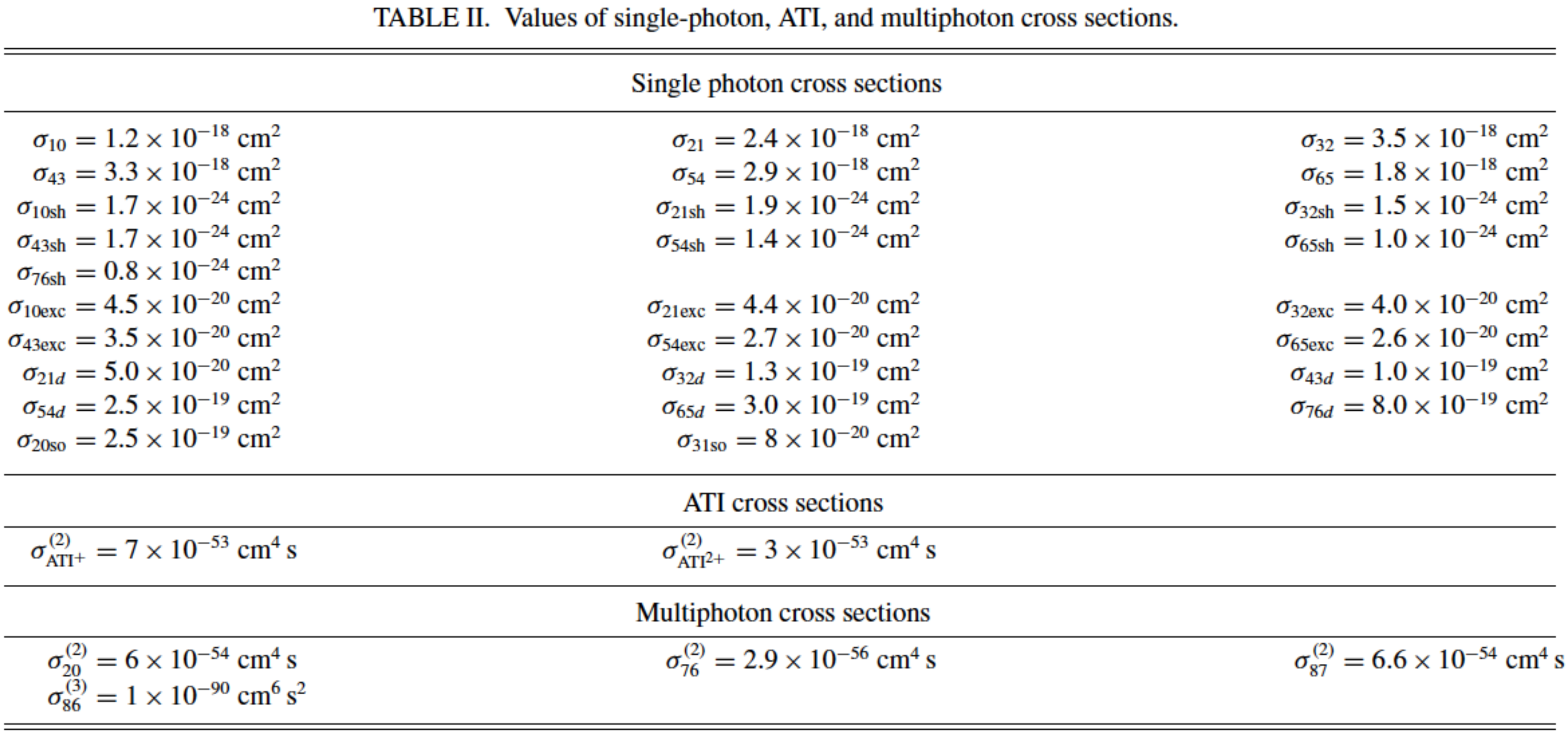

TABLE II. Values of single-photon, ATI, and multiphoton cross sections.

Single photon cross sections		
$\sigma_{10} = 1.2 \times 10^{-18}$ cm^2	$\sigma_{21} = 2.4 \times 10^{-18}$ cm^2	$\sigma_{32} = 3.5 \times 10^{-18}$ cm^2
$\sigma_{43} = 3.3 \times 10^{-18}$ cm^2	$\sigma_{54} = 2.9 \times 10^{-18}$ cm^2	$\sigma_{65} = 1.8 \times 10^{-18}$ cm^2
$\sigma_{10\mathrm{sh}} = 1.7 \times 10^{-24}$ cm^2	$\sigma_{21\mathrm{sh}} = 1.9 \times 10^{-24}$ cm^2	$\sigma_{32\mathrm{sh}} = 1.5 \times 10^{-24}$ cm^2
$\sigma_{43\mathrm{sh}} = 1.7 \times 10^{-24}$ cm^2	$\sigma_{54\mathrm{sh}} = 1.4 \times 10^{-24}$ cm^2	$\sigma_{65\mathrm{sh}} = 1.0 \times 10^{-24}$ cm^2
$\sigma_{76\mathrm{sh}} = 0.8 \times 10^{-24}$ cm^2		
$\sigma_{10\mathrm{exc}} = 4.5 \times 10^{-20}$ cm^2	$\sigma_{21\mathrm{exc}} = 4.4 \times 10^{-20}$ cm^2	$\sigma_{32\mathrm{exc}} = 4.0 \times 10^{-20}$ cm^2
$\sigma_{43\mathrm{exc}} = 3.5 \times 10^{-20}$ cm^2	$\sigma_{54\mathrm{exc}} = 2.7 \times 10^{-20}$ cm^2	$\sigma_{65\mathrm{exc}} = 2.6 \times 10^{-20}$ cm^2
$\sigma_{21d} = 5.0 \times 10^{-20}$ cm^2	$\sigma_{32d} = 1.3 \times 10^{-19}$ cm^2	$\sigma_{43d} = 1.0 \times 10^{-19}$ cm^2
$\sigma_{54d} = 2.5 \times 10^{-19}$ cm^2	$\sigma_{65d} = 3.0 \times 10^{-19}$ cm^2	$\sigma_{76d} = 8.0 \times 10^{-19}$ cm^2
$\sigma_{20\mathrm{so}} = 2.5 \times 10^{-19}$ cm^2	$\sigma_{31\mathrm{so}} = 8 \times 10^{-20}$ cm^2	
ATI cross sections		
$\sigma^{(2)}_{\mathrm{ATI}+} = 7 \times 10^{-53}$ cm^4 s	$\sigma^{(2)}_{\mathrm{ATI}2+} = 3 \times 10^{-53}$ cm^4 s	
Multiphoton cross sections		
$\sigma^{(2)}_{20} = 6 \times 10^{-54}$ cm^4 s	$\sigma^{(2)}_{76} = 2.9 \times 10^{-56}$ cm^4 s	$\sigma^{(2)}_{87} = 6.6 \times 10^{-54}$ cm^4 s
$\sigma^{(3)}_{86} = 1 \times 10^{-90}$ cm^6 s^2		

$\sigma(n)_{ij}$ denotes the ionization cross section for ionization of an ion with charge +j via an n-photon process thereby creating an ion with charge +i. Other processes are denoted through additional indices, where **exc** stands for the process of simultaneous ionization of an electron while exciting another to a d-state orbital, **d** denotes the ionization cross section of these excited d states, **sh** denotes ionization through radiation of the second harmonic, and **so** denotes the shake-off processes.

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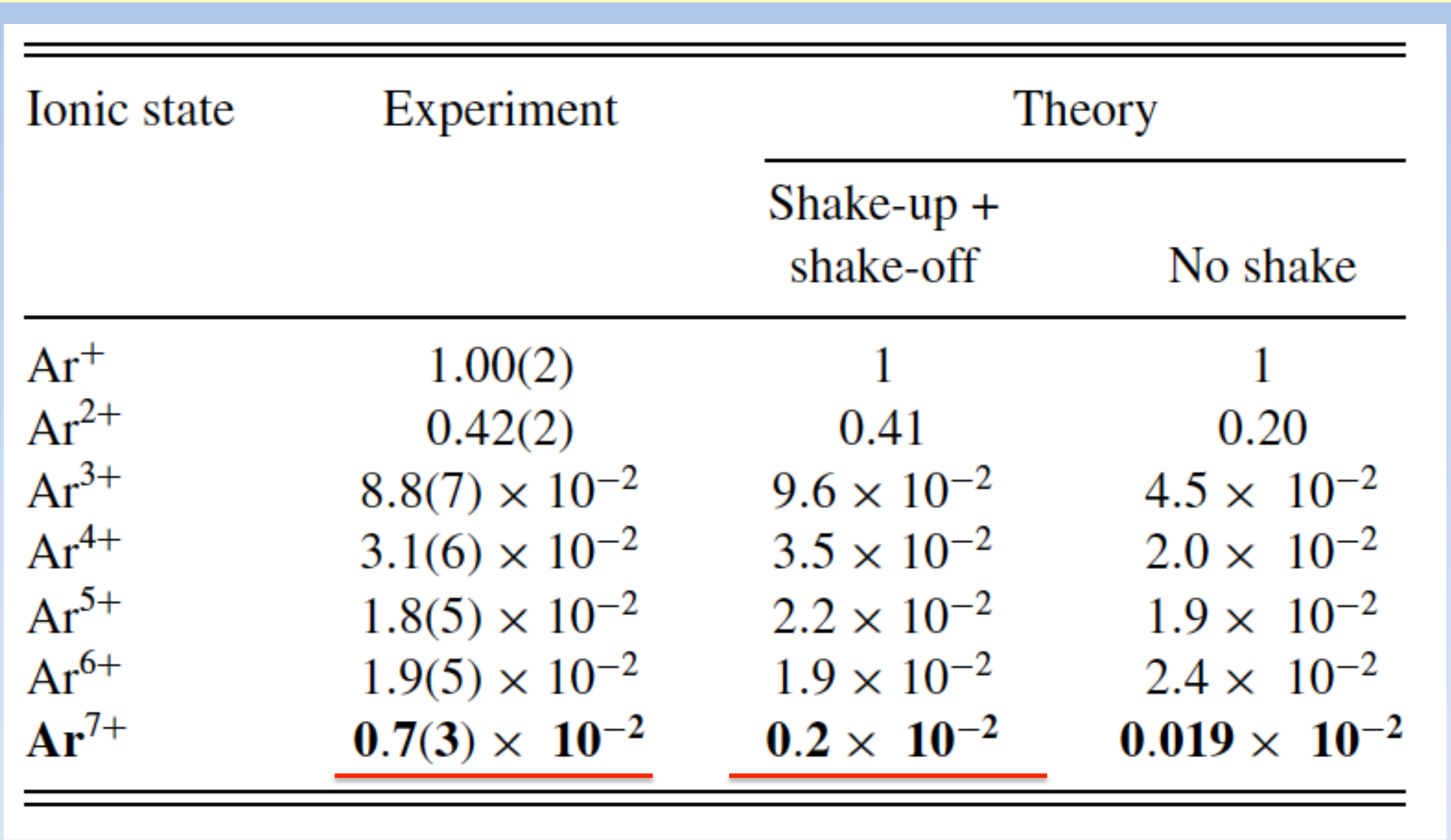
Ar ion yield – Experiment and Theory



Ionic state	Experiment	Theory	
		Shake-up + shake-off	No shake
Ar^{+}	1.00(2)	1	1
Ar^{2+}	0.42(2)	0.41	0.20
Ar^{3+}	$8.8(7) \times 10^{-2}$	9.6×10^{-2}	4.5×10^{-2}
Ar^{4+}	$3.1(6) \times 10^{-2}$	3.5×10^{-2}	2.0×10^{-2}
Ar^{5+}	$1.8(5) \times 10^{-2}$	2.2×10^{-2}	1.9×10^{-2}
Ar^{6+}	$1.9(5) \times 10^{-2}$	1.9×10^{-2}	2.4×10^{-2}
Ar^{7+}	**$0.7(3) \times 10^{-2}$**	**0.2×10^{-2}**	**0.019×10^{-2}**

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Summary - One Colour

- The work highlights the importance of processes governed by electron correlation in particular, namely, ionization with excitation and shake-off, to be important in intense FELs fields and not just synchrotron studies
- Such processes are usually accessible only by measurements of the photoelectrons but we have uncovered strong indications of ionization plus excitation in the relative abundance of ionic species, without recourse to photoelectron energy spectra, in the case of FELs.
- The Ar^{7+} yield shows a clear deviation from the predictions of the commonly used model of sequential ionization via single-electron processes. The observed signal can only be explained by taking into account the full multiplet structure of the electron configurations involved and by inclusion of two-electron processes
- For a photon energy of 105 eV, the ejection of one electron from the $3s^2$ ground state of Ar^{6+} is a two-photon process, while ionization of the highest 3s3p multiplet state or the ejection of a 3d or another outer electron from another excited state [e.g., Ar^{6+}(3s3d)] is a single-photon process
- 3d cross section is low for low charges states. $\sigma_{3p} \sim \sigma_{3d}$ at Ar^{5+} and $\sigma_{3d} > \sigma_{3p}$ at Ar^{6+} as the 3d becomes more tightly bound.
- Two-photon ionization from the ground state of Ar^{6+} is calculated to be orders of magnitude smaller than σ_{3p} & σ_{3d}

XUV (X-ray) + IR Ionization

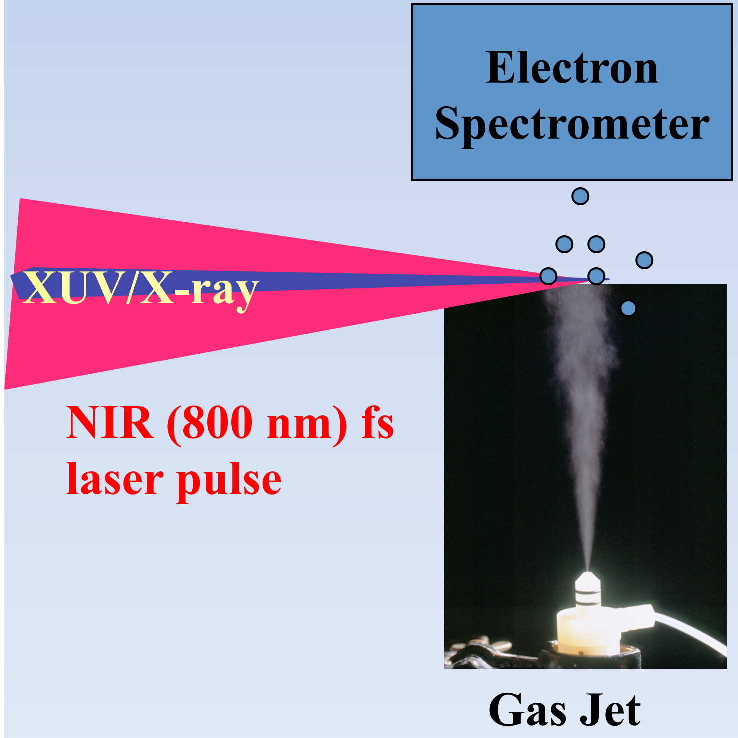
1. Rudiments of ionization processes in intense laser fields
2. Photoionization experimental setup (FLASH@DESY)
3. One colour (Ar) – Role of two electrons processes
4. **Two colour (He) – ARPES with polarisation control**
5. Next Step

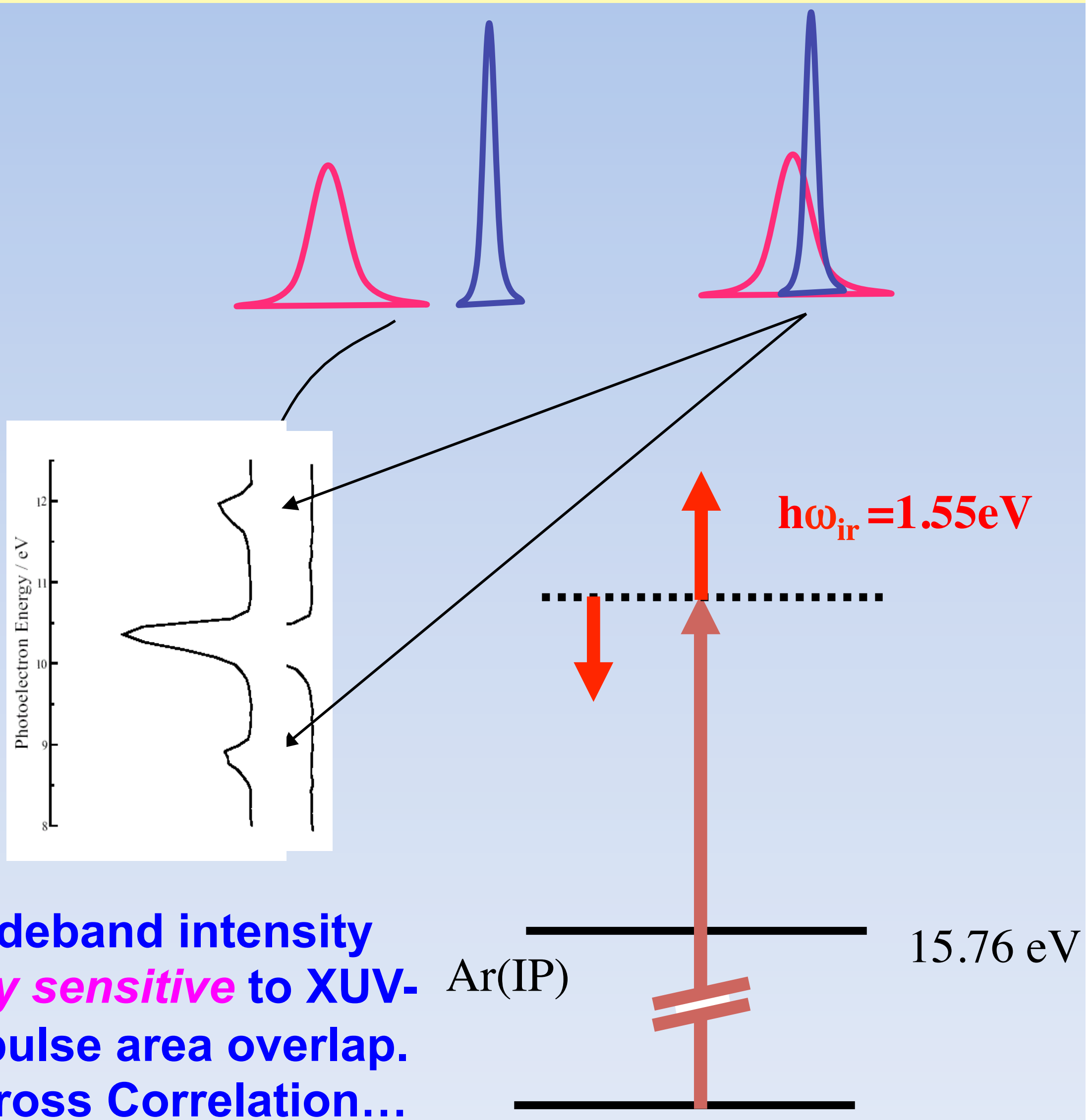
Two colour ATI/ Laser Assisted PES

Superposition of visible and XUV pulses in a noble gas jet

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

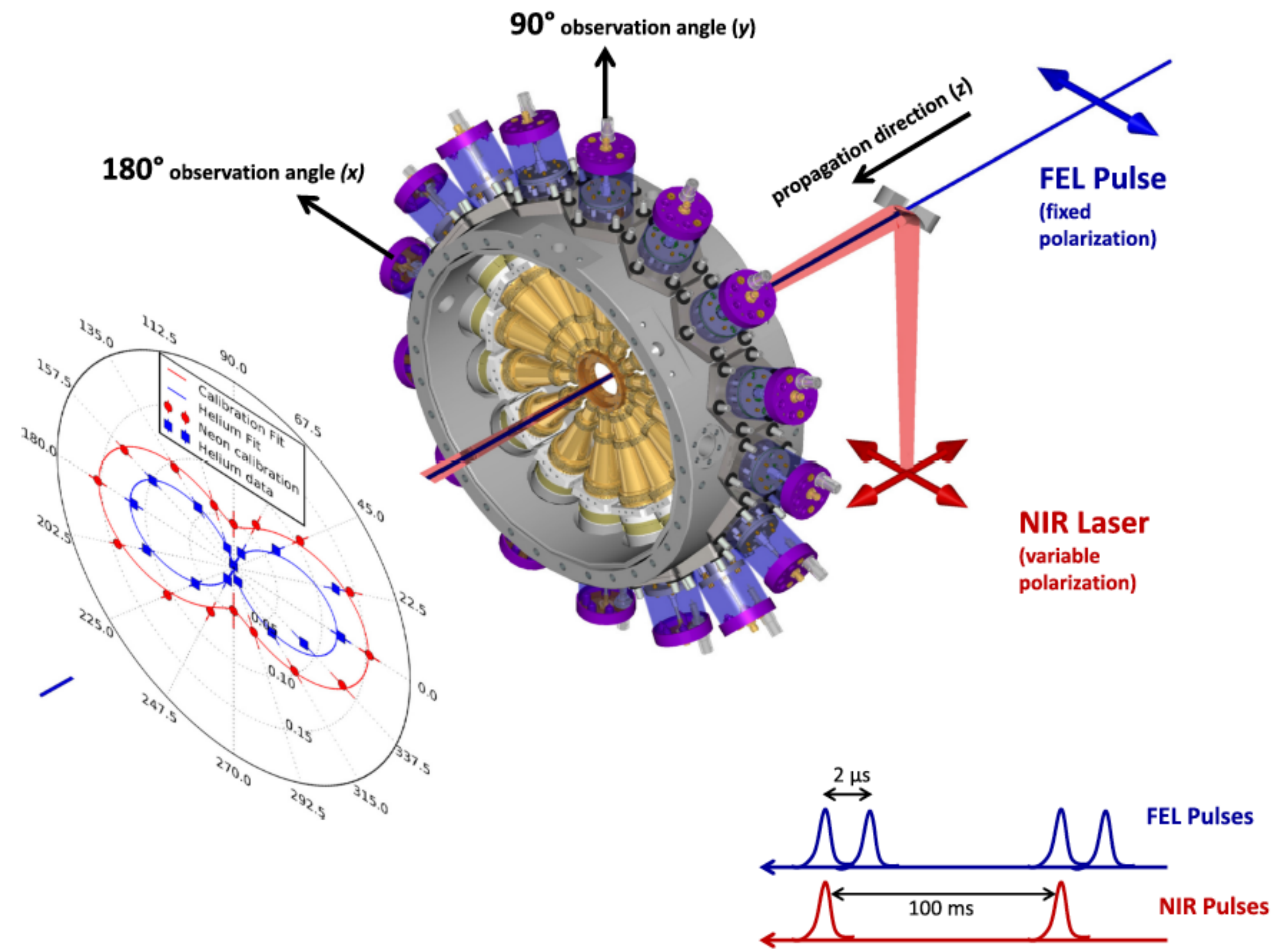
E.S. Toma et al. PRA **62** 061801 (2000)





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

Two Colour ARPES – AR Sidebands



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Two Colour ARPES – AR Sidebands

FEL:

PG2 Beamline – Zero Order

$h\nu$=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:

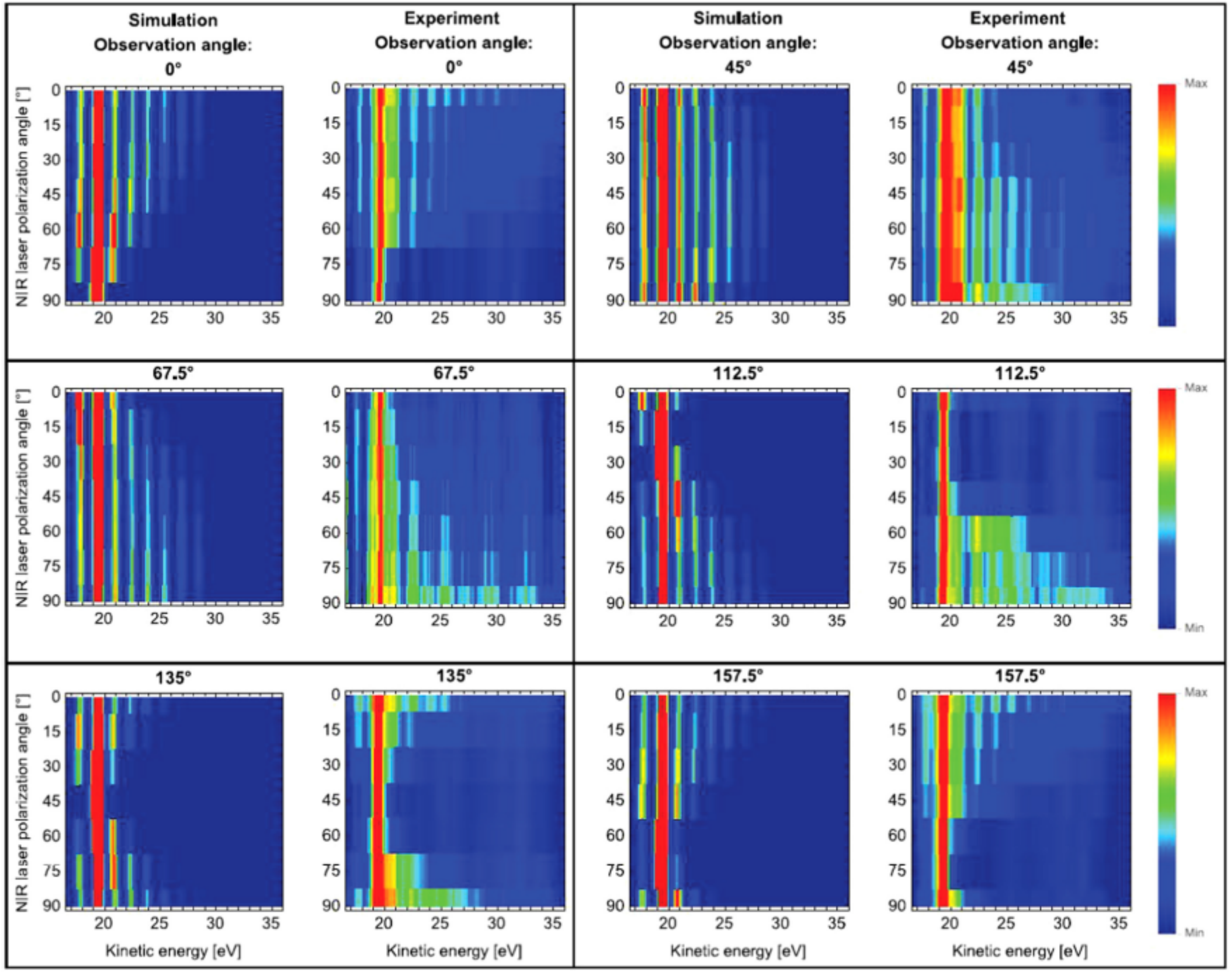
16 TOF channels at 22.5^{O} increments

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 mth 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

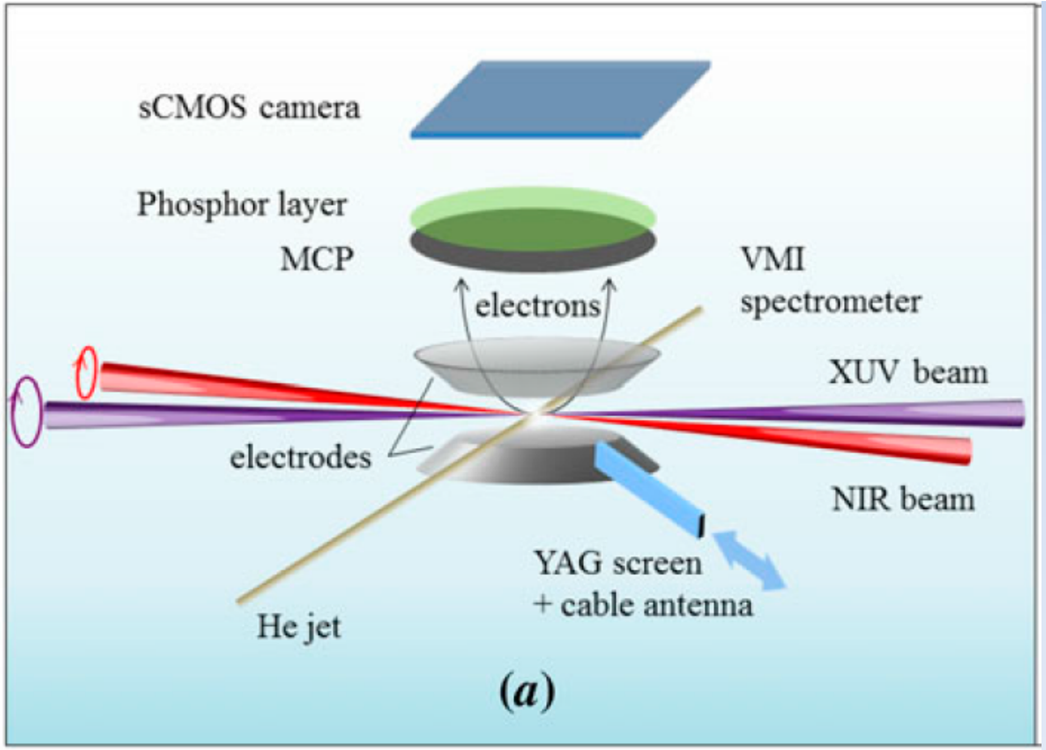
- What happens when we change from plane polarised XUV and NIR to circularly polarised XUV and NIR ?

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]



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}

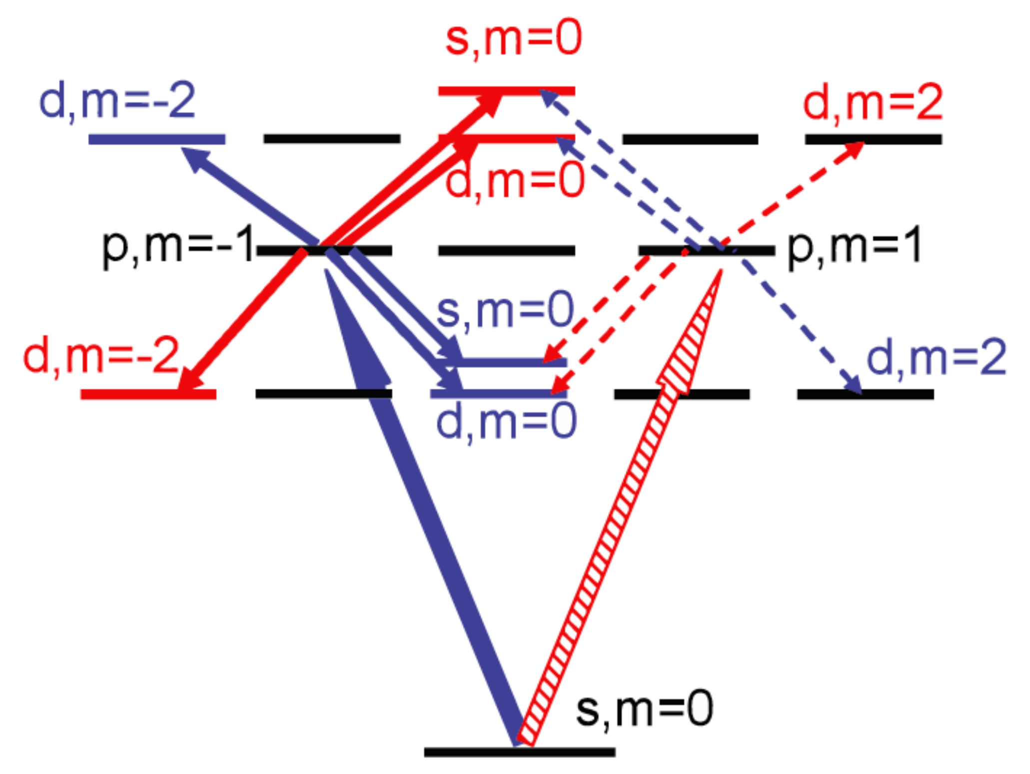
Timing Jitter: 25 fs (rms)

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

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

$$D_l(E) = \int\frac{\int P_{El}(r)rP_{E'p}(r)\mathrm{d}r\int P_{E'p}(r)rP_{1s}(r)\mathrm{d}r}{E'-\omega_X+i0}\mathrm{d}E'$$

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

Two Colour ARPES - Circ. Dichroism - He

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

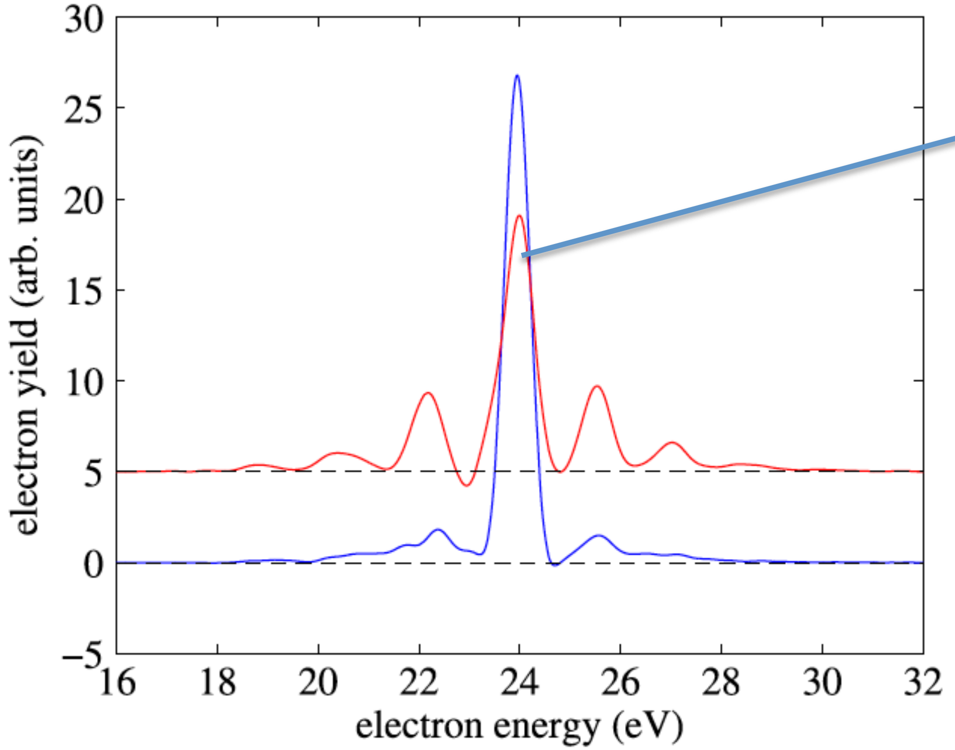


Figure 4. High intensity (red curve) and low intensity (blue curve) spectra at $\pi/2$ emission angle when both XUV and NIR pulses are left-hand circularly polarized. NIR intensities are 7.2×10^{12} and 3×10^{11} $\mathrm{W/cm^2}$, respectively. (The colour version of this figure is included in the online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

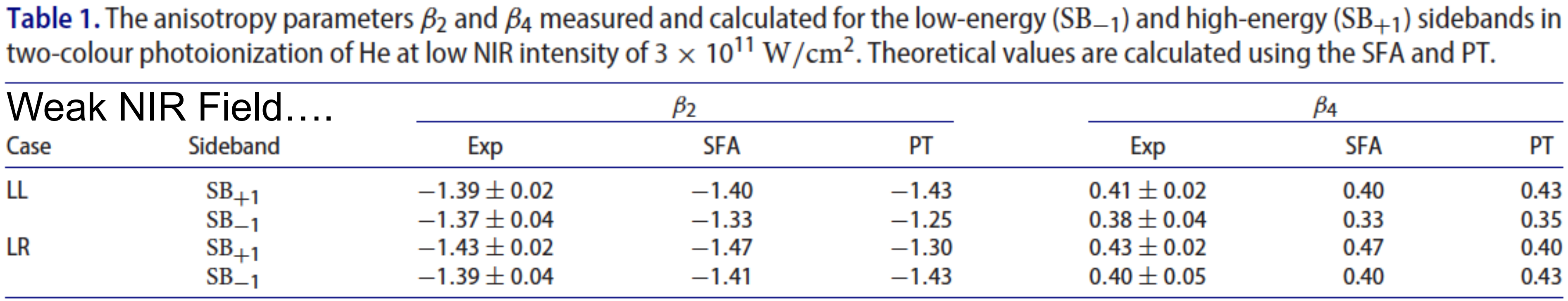


Table 1. The anisotropy parameters β_2 and β_4 measured and calculated for the low-energy (SB_{-1}) and high-energy (SB_{+1}) sidebands in two-colour photoionization of He at low NIR intensity of 3×10^{11} W/cm^2. Theoretical values are calculated using the SFA and PT.

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

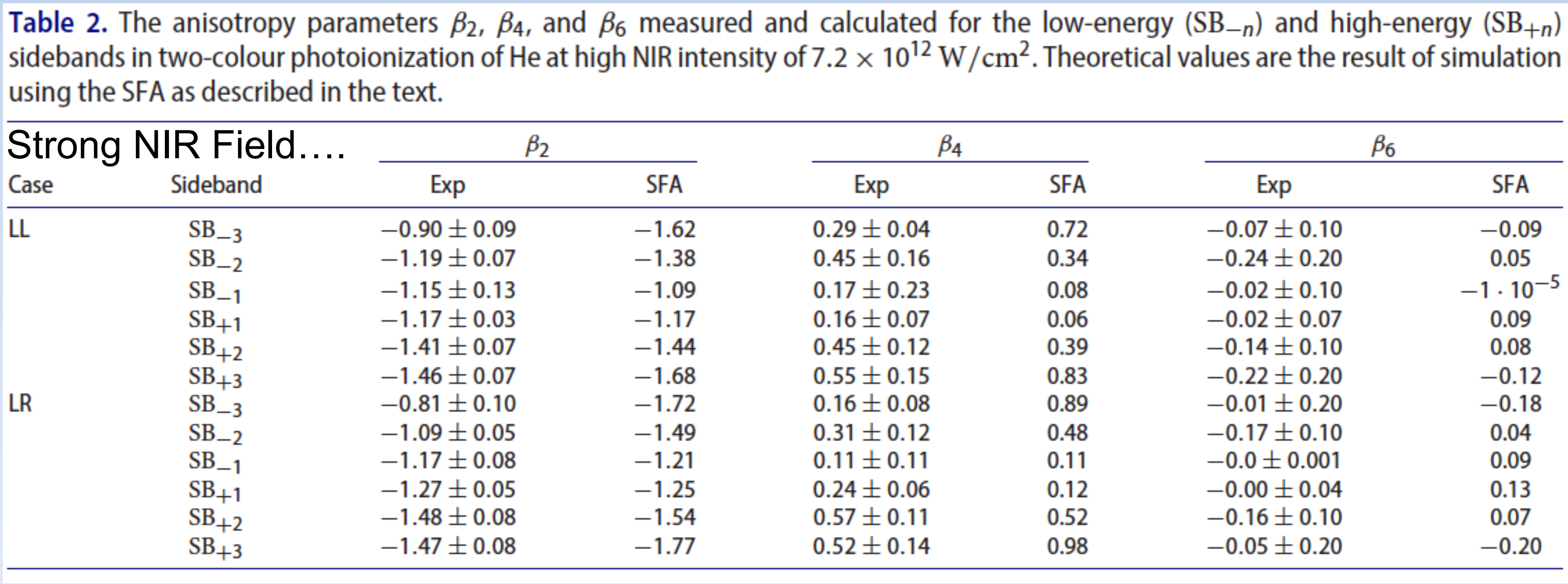


Table 2. The anisotropy parameters β_2, β_4, and β_6 measured and calculated for the low-energy (SB_{-n}) and high-energy (SB_{+n}) sidebands in two-colour photoionization of He at high NIR intensity of 7.2×10^{12} W/cm^2. Theoretical values are the result of simulation using the SFA as described in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

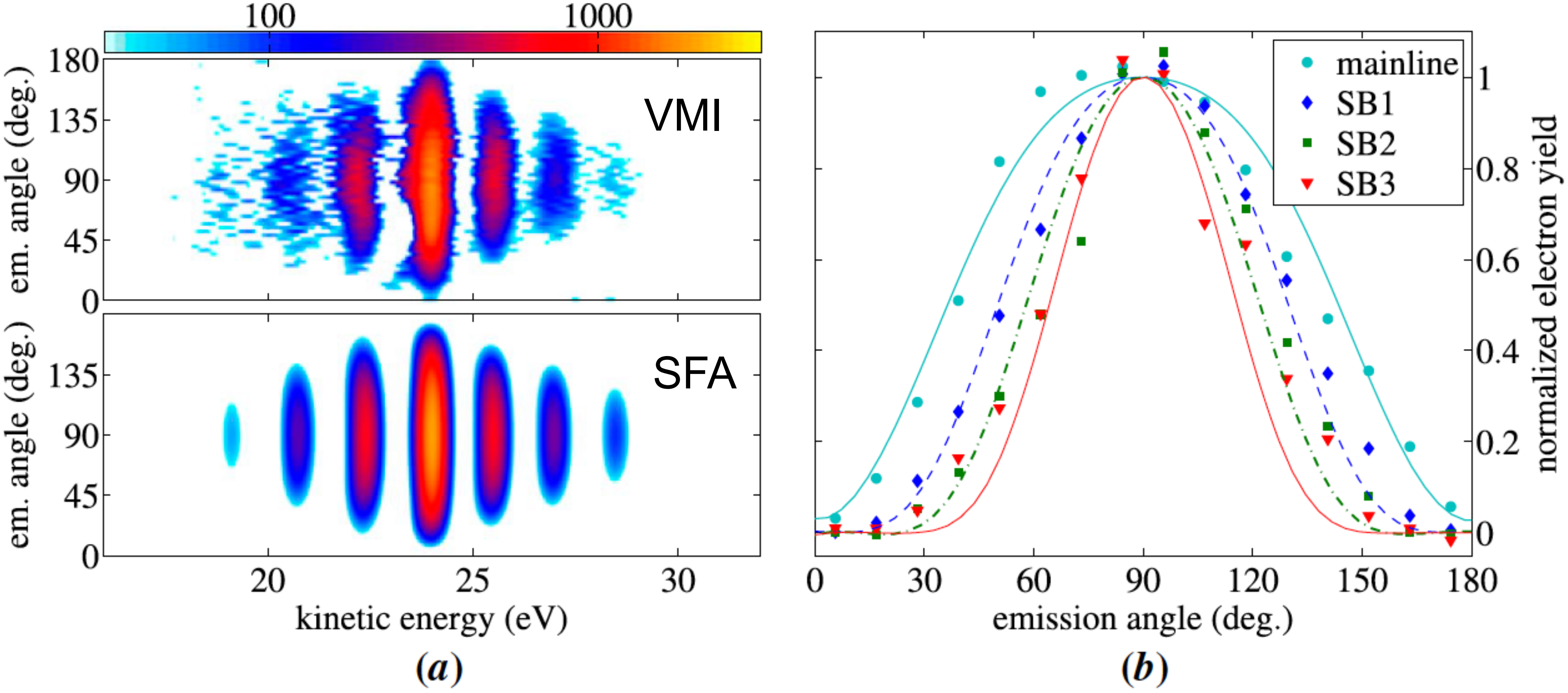
SFA and Perturbation Theory (weak NIR fields)

- 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}(θ) dependence* – Ang. distribution peaked at θ=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)

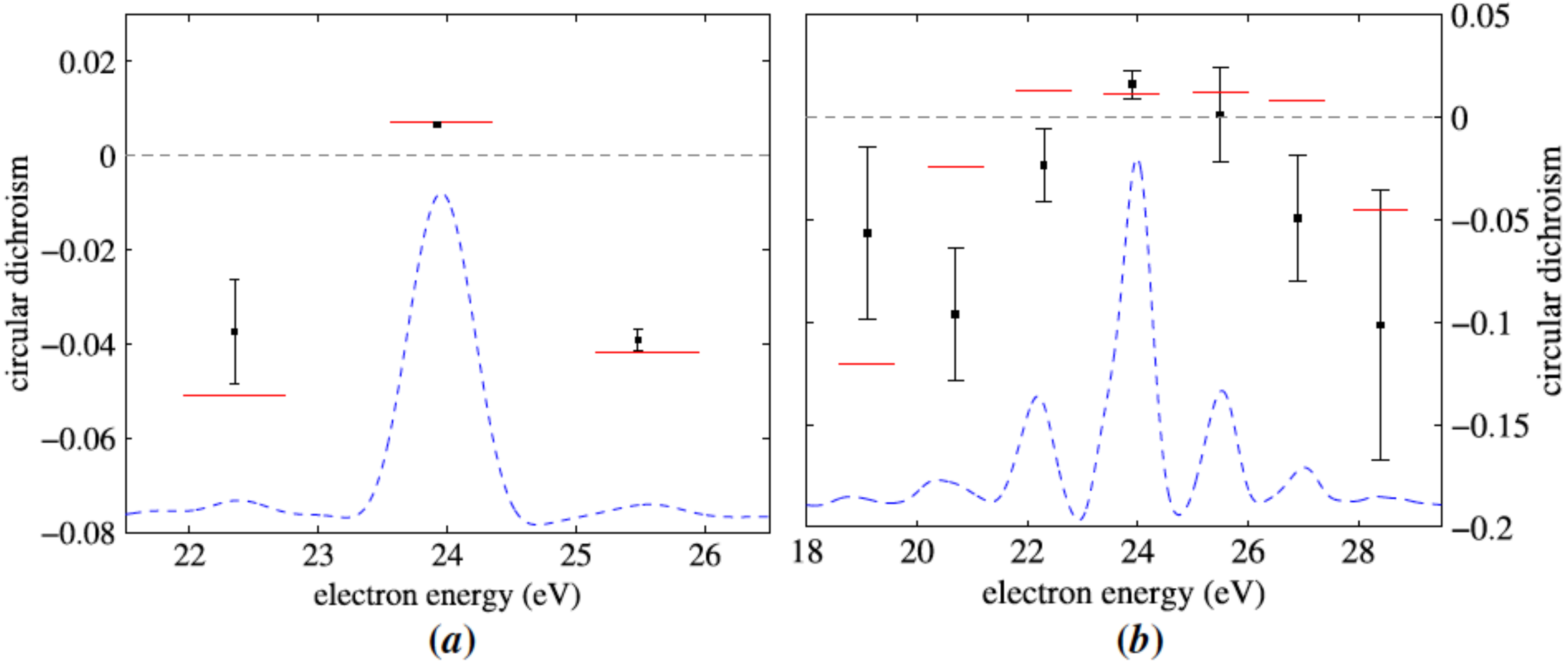


Figure 8. Experimentally determined CDADs at 90° emission angle for the central line and the sidebands for the NIR intensity of 3×10^{11} W/cm^2 (*a*) and 7.2×10^{12} W/cm^2 (*b*) are shown by dots with error bars. The dashed lines represent the experimental electron spectra (the zero-line is shifted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

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. **Next Step**

Next Step..

- As already demonstrated in numerous studies in the linear regime (weak field synchrotrons) at low intensities, the sensitivity of the CD to details of the electron dynamics and to the atomic/molecular structure will also provide unique and valuable information on non-linear photoemission processes, which are accessible now with the new FEL sources.

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