Ionization Processes in Ultrashort XUV, X-ray and IR Laser Fields

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

CDAMOP, Delhi, Dec 14-16, 2011

Collaboration @ FLASH-DESY, Hamburg

XFEL: P. Radcliffe & M. Meyer

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

PTB (Berlin): A. A. Sorokin (now at IOFFE & DESY) & M. Richter

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

Orsay: D. Cubaynes

Queen's University Belfast: C. L. S. Lewis, *A Delserieys,* H. van der Hart (T)

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

Crete: P. Lambropoulos (T)

Oulu/GSI: S. Fritzsche (T)

DCU: V. Richardson, P. Hayden, T. J. Kelly, E. T. Kennedy, & J. T. Costello

Thanks to AG Photon (R Treusch et al.) & AG Machine (M Yurkov et al.)

Collaboration @ LCLS X-ray FEL (SLAC)

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

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

DCU: T. J. Kelly, 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 & J. Bozek

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

Moscow State University: N. Kabachnik

Thanks to Paul Emma et al. of the machine and diagnostics group.

*Now at Argonne.

DCU Laser Plasma/ AMOP Group

6 laboratory areas focussed on pulsed laser matter interactions (NIR – X-ray/ 30fs – 30 ns, spectroscopy/ imaging/ PLD)

Academic Faculty (5): John T. Costello, Eugene T. Kennedy (Emeritus), Jean-Paul Mosnier, Lampros Nikolopoulos (T) and Paul van Kampen

Current Postdocs (3):
Dr. Patrick Hayden, Dr. Sateesh Krishnamurthy and Dr. Subhash Singh

Funded by:
SFI - Frontiers and Investigator
HEA – PRTLI (Kit)
IRCSET (People)
EU - Marie Curie (People)

Current PhD students (9): Jack Connolly, Leanne Doughy, Colm Fallon, Eanna Mac Carthy, Mossy Kelly, Nichola Walsh, Jiang Xi, Damien Middleton, Cathal O'Broin

Recent (2009-2010) research student graduates:
Brian Doohan, John Dardis, Padraig Hough, Rick O'Haire & Vincent Richardson.

Interns: Thomas Butler, Sarah Feeney, Ronan McCann, Aidan Fallon, Clare Devery, Ciaran Gaffney, Laura Kerr

DCU Int'l Fellows: Prof. Sivanandan Harilal (Purdue) & Dr. Will Bryan (Cardiff)

Outline of Talk

1. SASE-FELs, Operating Principle and Characteristics
2. Rudiments of ionization processes in intense laser fields
3. Photoionization experimental setups (FLASH & DESY)
4. Two photon ionization
5. Two colour Ionization
6. Next Steps

SASE-FELs - Fundamental Principle

$N_u \lambda_U = vt$ $N_u \lambda_{ph} = (c-v)t$ $\Rightarrow \lambda_{ph} \sim \lambda_U (c-v)/v \sim \lambda_u/2\gamma^2$

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

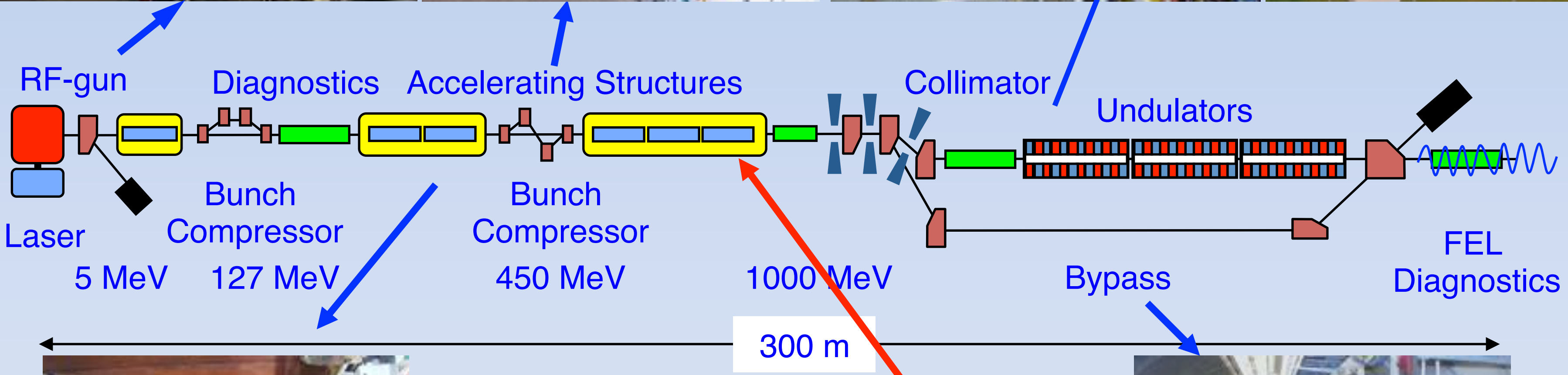
$\lambda = \lambda_u(1+K^2/2)/2\gamma^2$ $\gamma = E/mc^2$

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

Wavelength tunable

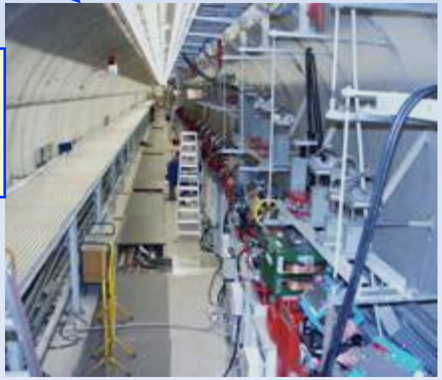
Electron bunch slips behind lightwave by λ per undulator period

FLASH FEL – Physical Layout



LINAC Energy : ~ 1 GeV

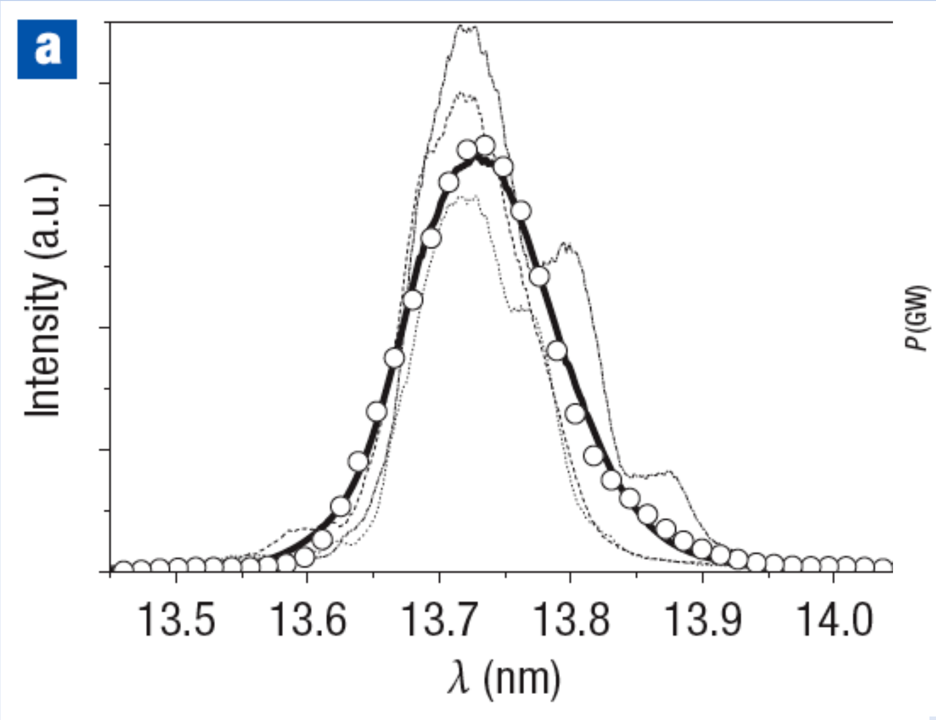
➔ ~ 4 – 60 nm



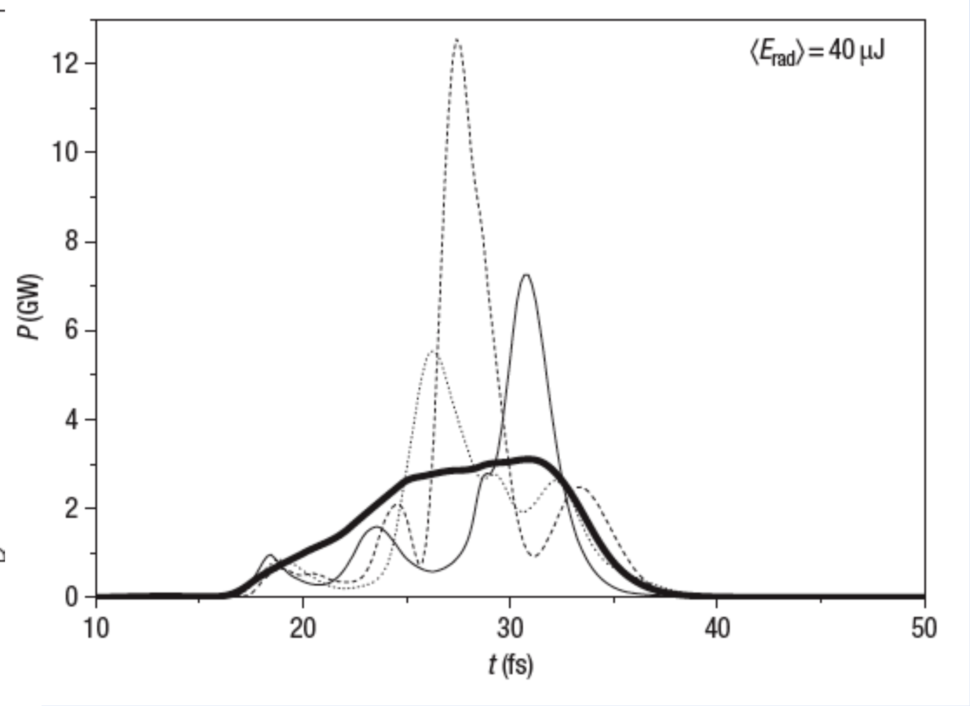
O/P Profile & Spectral Distribution

FEL output builds up from spontaneous emission (photon noise) => ***SASE* – *'Self Amplified Spontaneous Emission'* => *Fluctuations in beam profile, pointing stability, intensity, spectral distribution and pulse duration !!***

Spatially coherent only - 'Seeded FEL projects at FLASH & LCLS'



Spectral Fluctuations

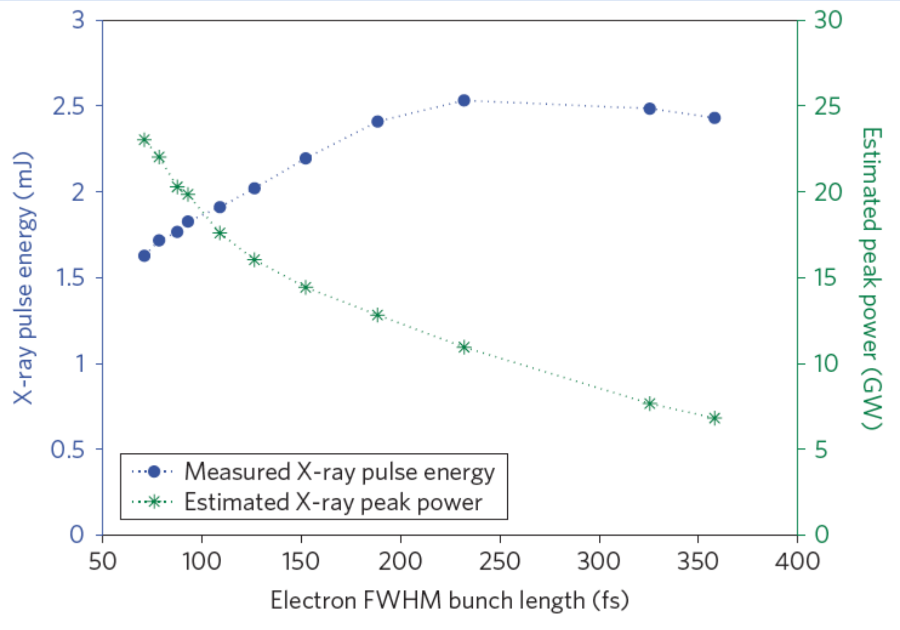


Temporal Fluctuations

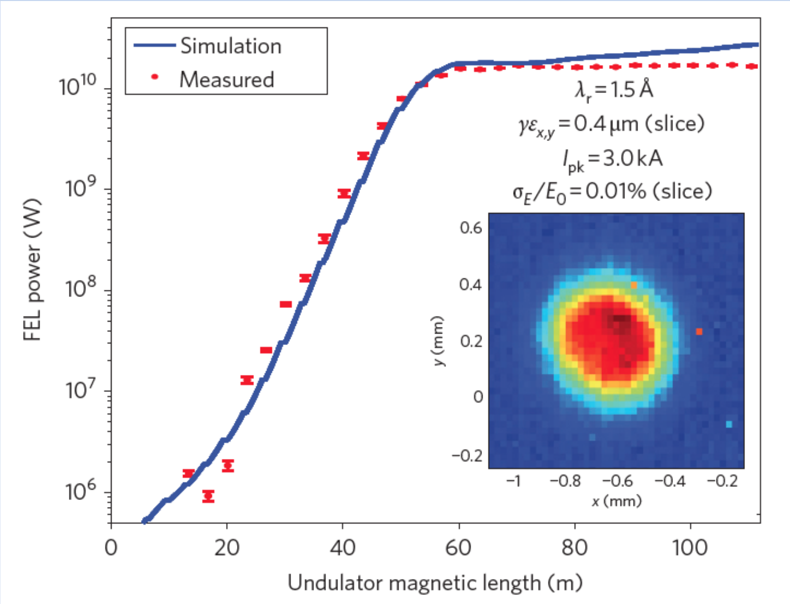
Nature Photonics **1** 336 (2007)

LCLS Overview and Specifications

LCLS pulse energy and peak power



LCLS lasing saturation at ~ 8.25 keV

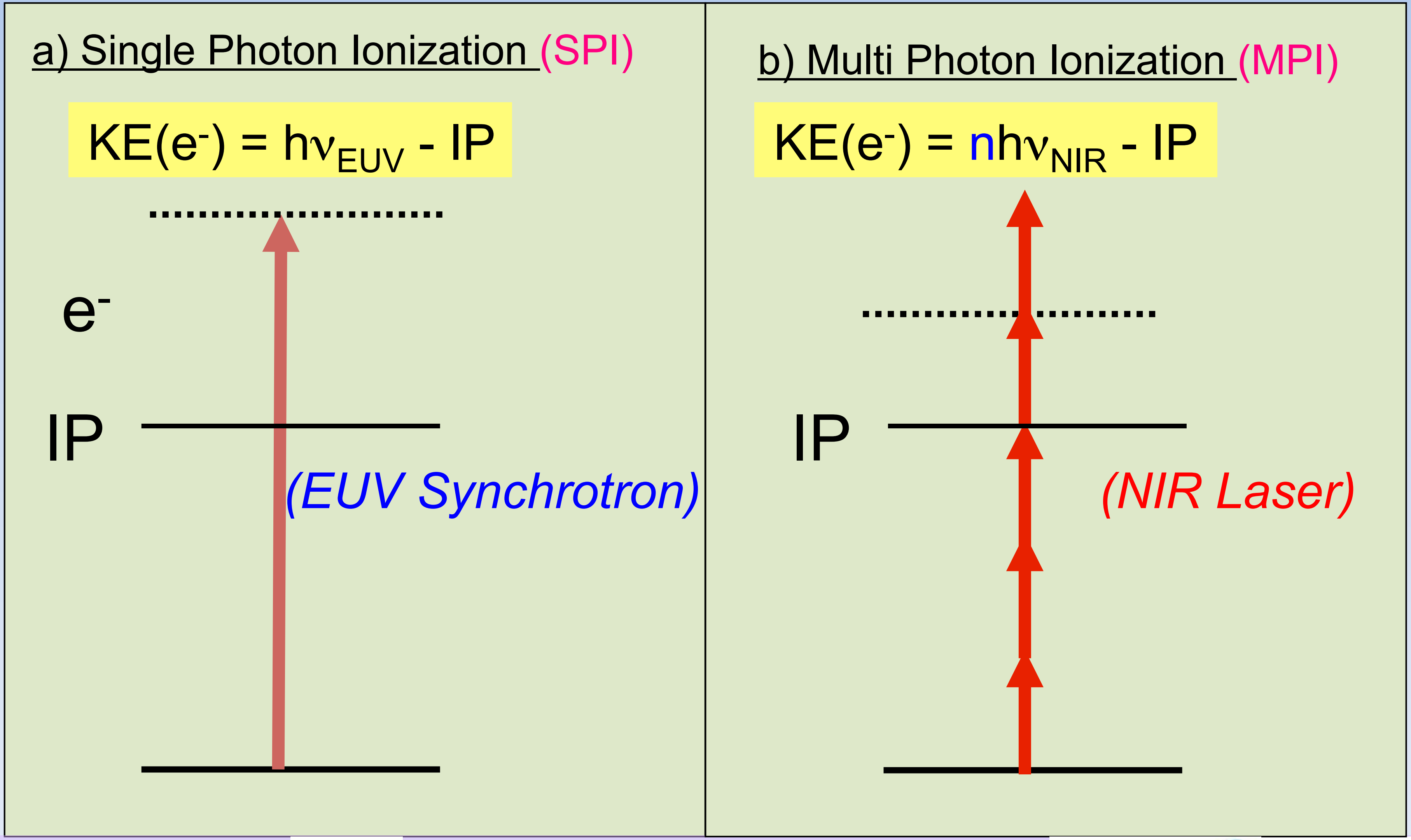


P. Emma et al., Nature Photonics **4**, pp 641-647 (2010)

Ionization in Intense Fields

1. SASE-FELs, Operating Principle and Characteristics
2. **Rudiments of ionization processes in intense laser fields**
3. Photoionization experimental setups (FLASH & DESY)
4. Two photon ionization
5. Two colour Ionization
6. Next Steps

The Atomic Photoelectric Effect



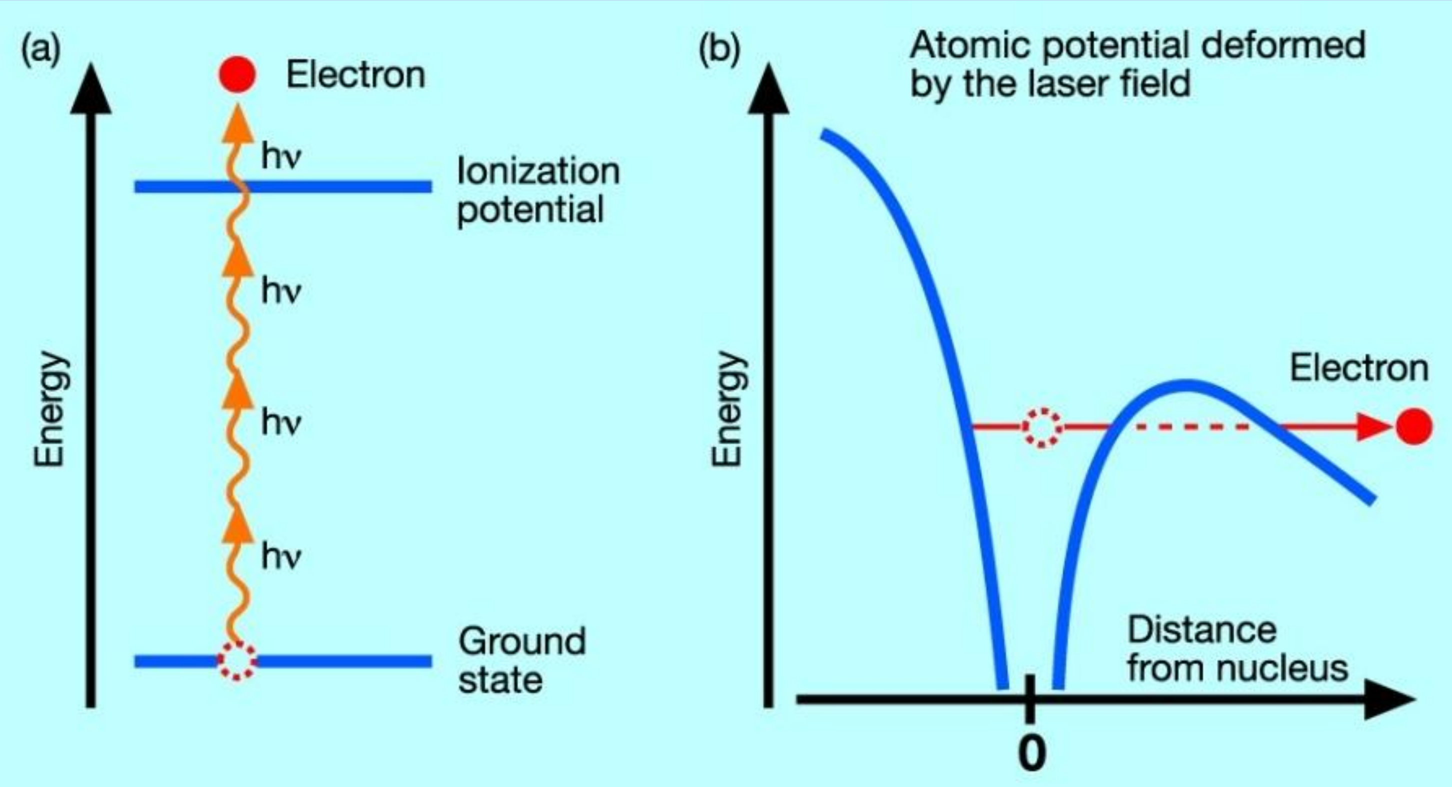
a) Single Photon Ionization (SPI)

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

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 ?

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

Keldysh Parameter

IP = Ionization Potential
Up = Ponderomotive Pot.

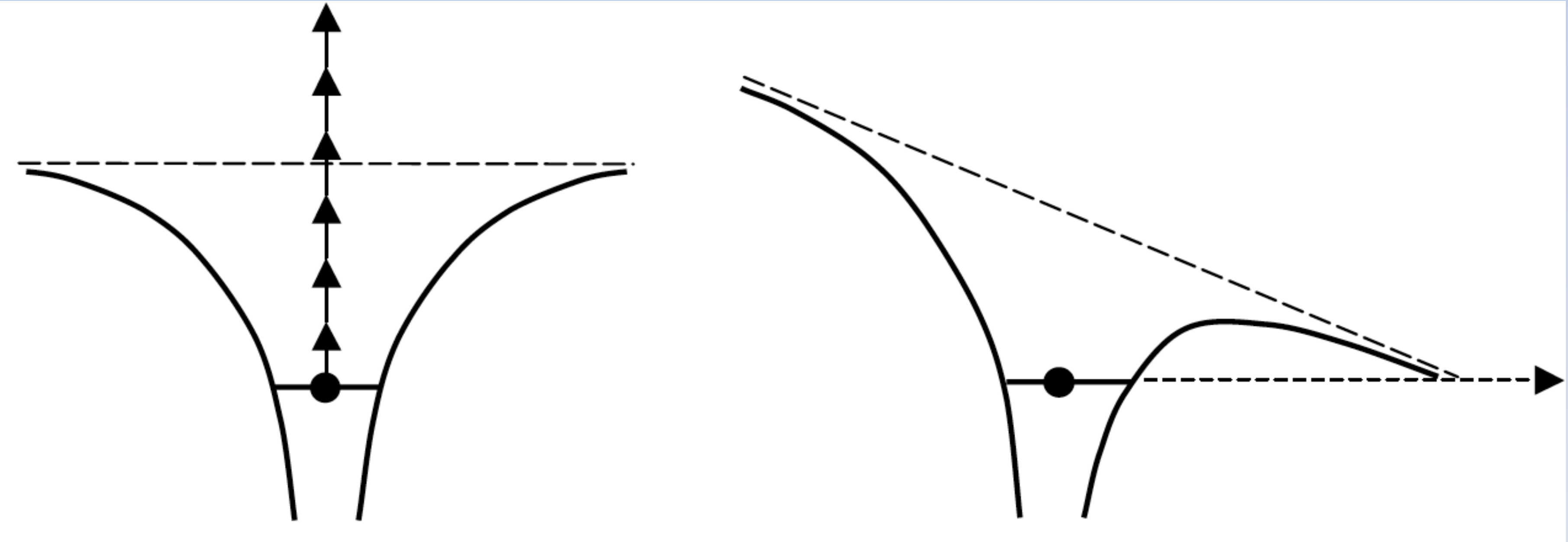
$$U_P = 9.3\times10^{-14} I\left(Wcm^{-2}\right)\,\lambda^2\left(\mu m\right) \quad eV$$

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

Keldysh - Ionization Regime

Multiphoton Ionization   Tunnel Ionization   Field Ionization

$\gamma>>1$ $\gamma \sim 2$ $\gamma <<1$



Intensity/ Wavelength →

← Photon Energy

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

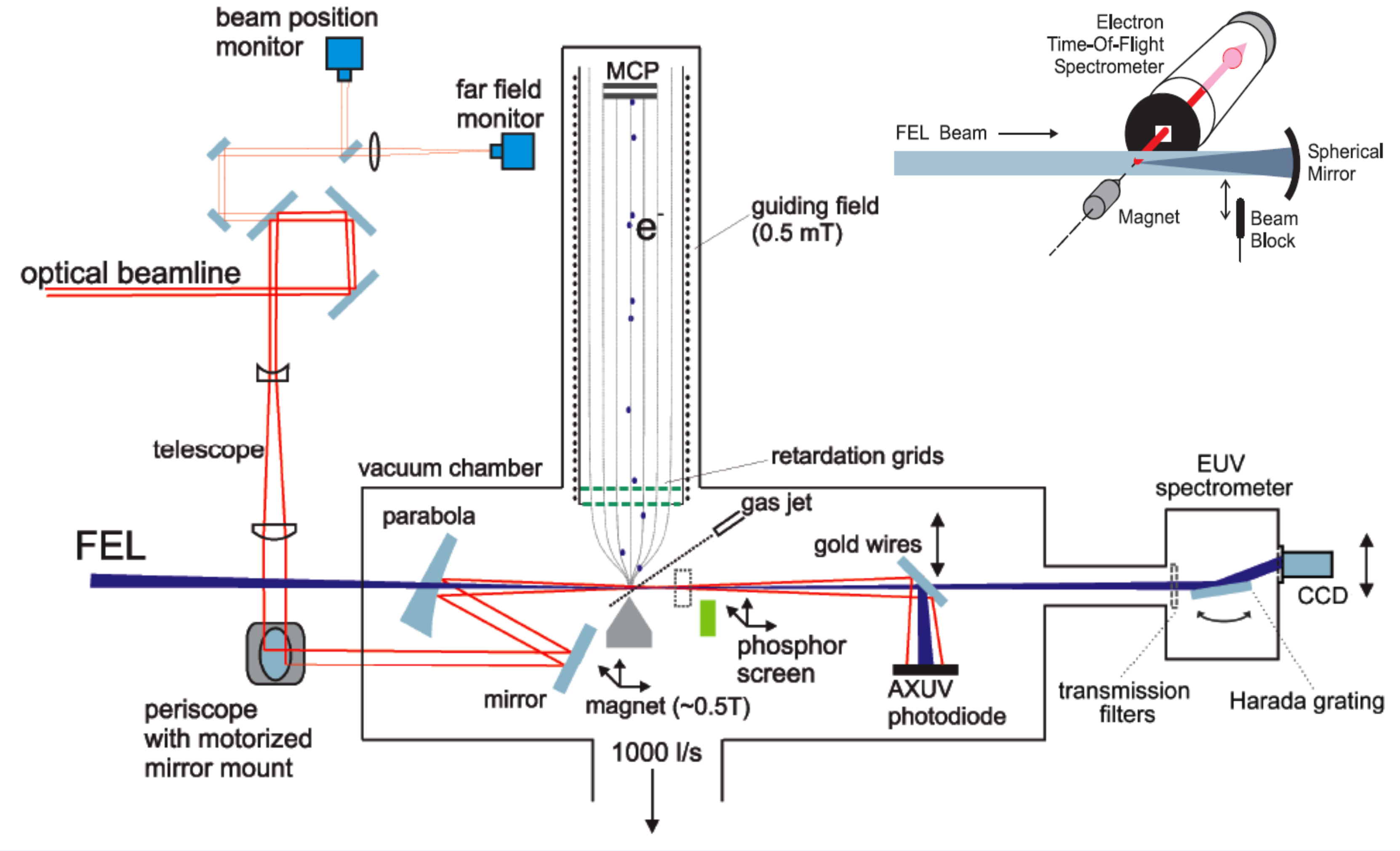
NB2: Re-asserts *primacy of the photon* over field effects !

Experimental Setups (DESY & SLAC)

1. SASE-FELs, Operating Principle and Characteristics
2. Rudiments of ionization processes in intense laser fields
3. **Photoionization experimental setups (FLASH & LCLS)**
4. Two photon ionization
5. Two colour Ionization
6. Next Steps

Photoelectron Spectroscopy @ FLASH

Experimental Layout at FLASH - (EU-RTD)

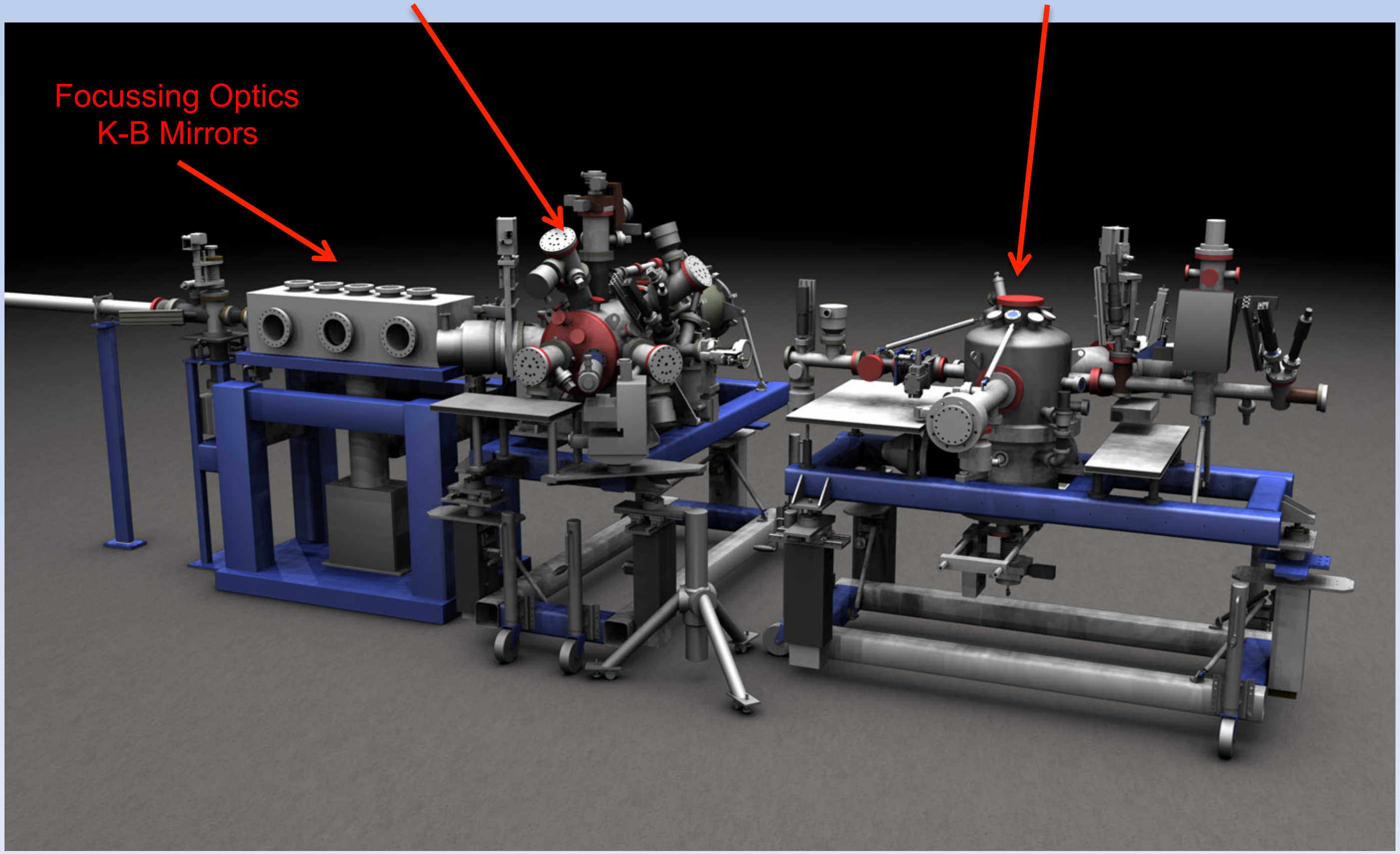


Nucl. Instr. & Meth. A **83**, 516-525 (2007)

AMO PES Chamber at LCLS

Rendered Image:

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



http://lcls.slac.stanford.edu

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

1. SASE-FELs, Operating Principle and Characteristics
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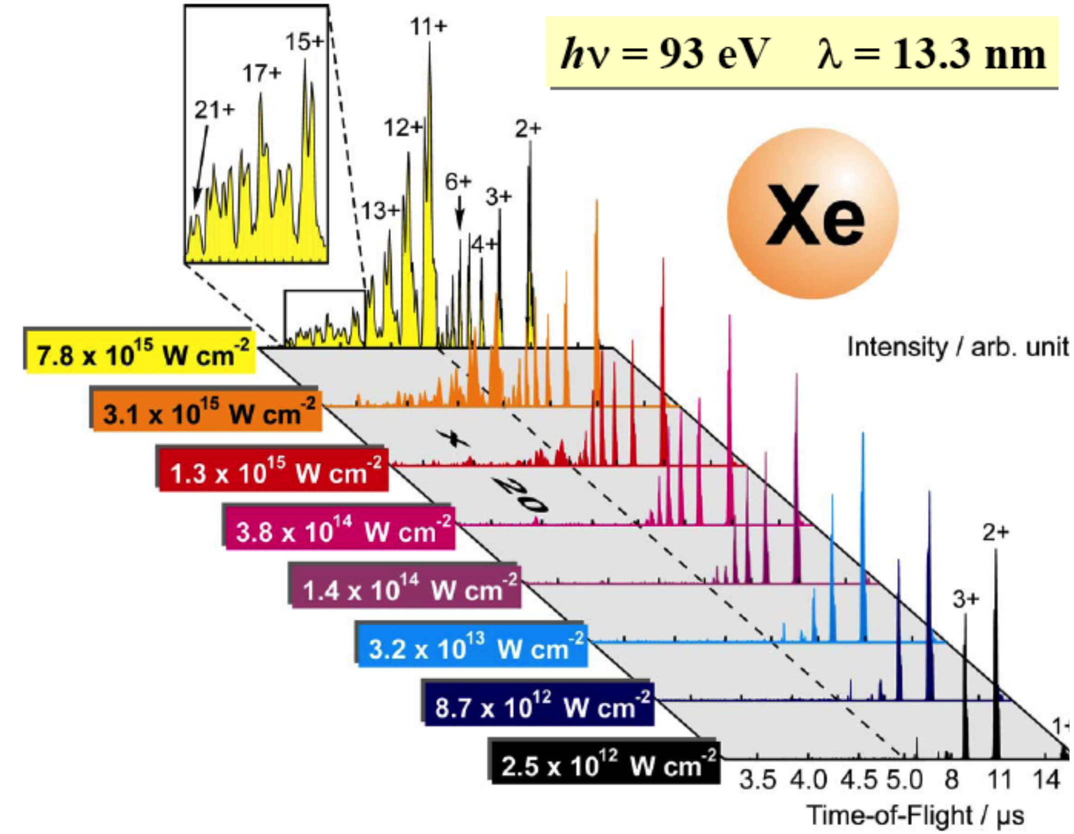
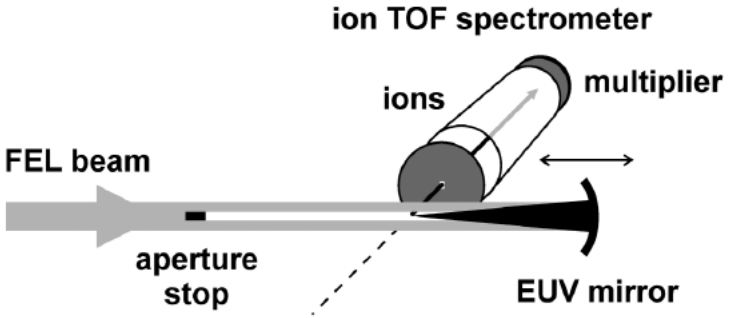
Motivation - Xe TPI in intense EUV fields

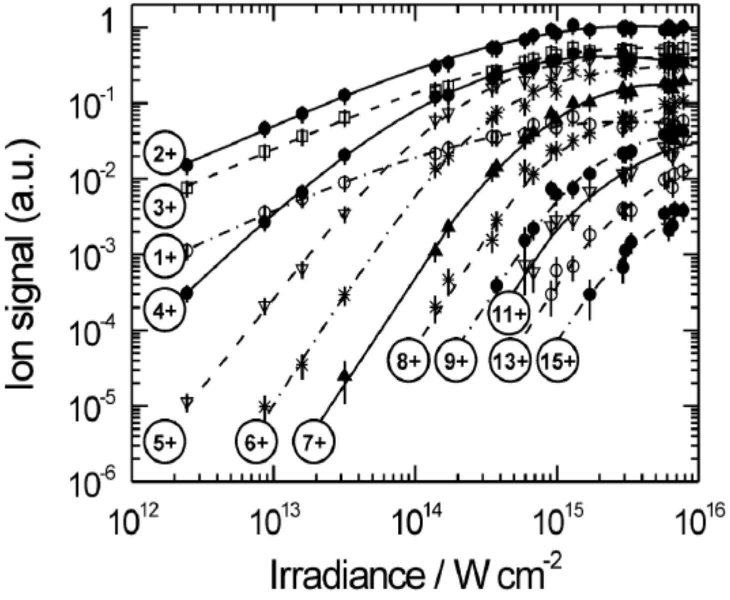
Sorokin, Richter et al., PTB, PRL 2007 – Ion Spectroscopy !!

Photoionization of xenon atoms in the EUV at ultra-high intensities: ion time-of-flight spectra

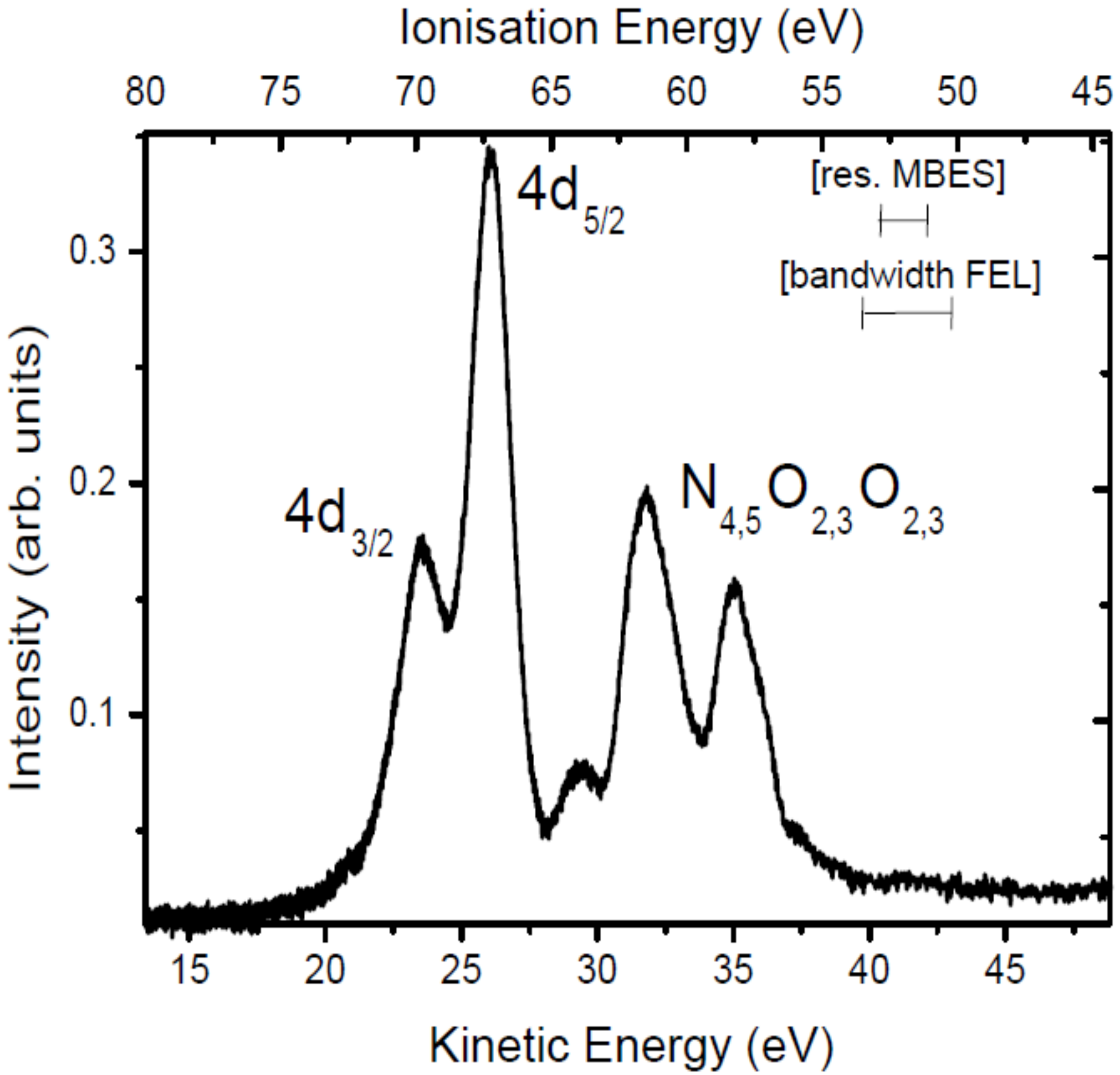
$h\nu = 93$ eV $\lambda = 13.3$ nm







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



Electrons !!!

- Single shot spectrum….
- For low intensities ($<10^{13}$ W.cm^{-2}), ONE PHOTON processes dominate
- **Salient features** – spin orbit split 4d photoelectron line + Auger electron spectrum
- **Not shown** – $5s^{-1}$ and $5p^{-1}$ lines at higher KEs

Xe + 2hν (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 –

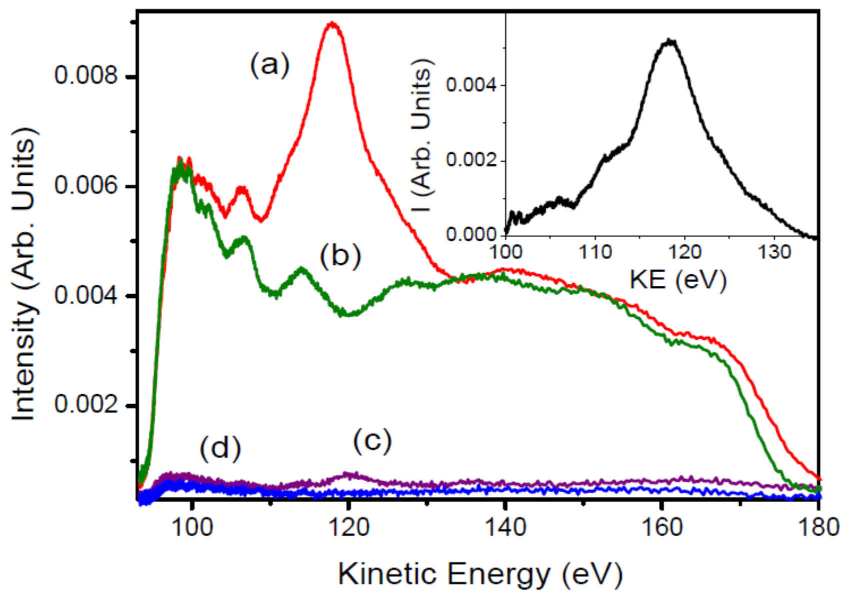
  Xe + 2*hv* → Xe$^+$ 4d^9 + e$^-$

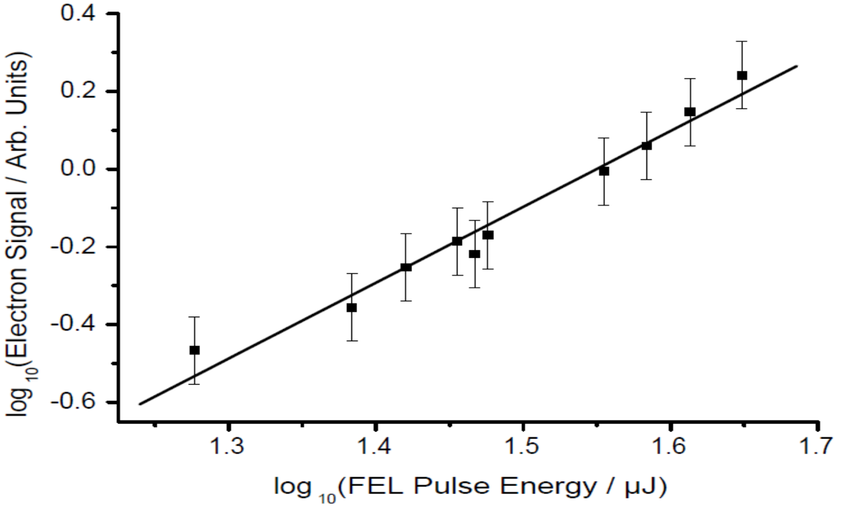
- ‘Retardation field’ applied to suppress low KE electrons (one photon processes) – hence electrons detected are due solely to multiphoton events

- Energetically –

  2 × (93) eV – 118 eV = 68 eV

- Yield scales quadratically, n=1.95 ± .2





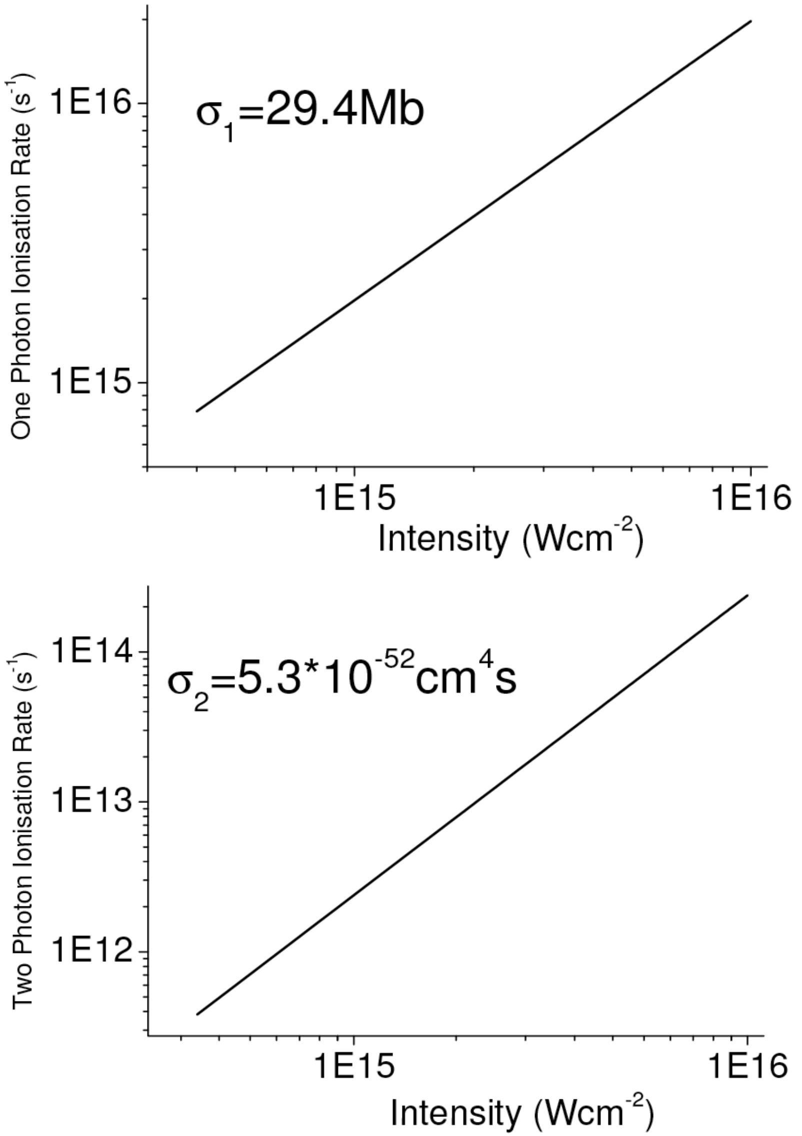
1 to 2 Photon Ionization Branching Ratio

1. R-Matrix (H.W. Van der Hart) – one and two photon 4d emission cross sections

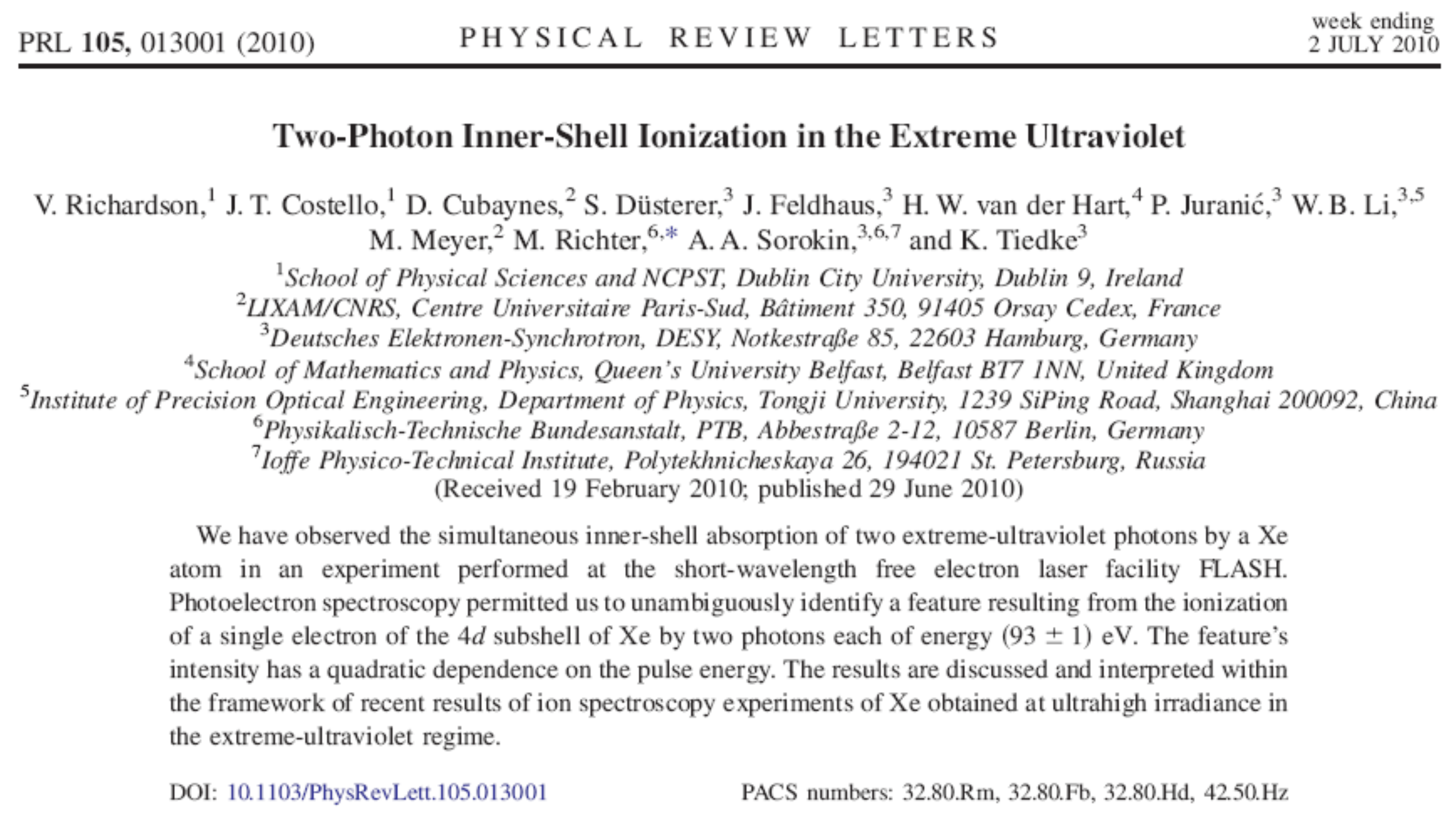
2. Dominant process is one photon ionization – 93 eV can remove next 4d as well - or maybe excite 4p – 4d. High rate of 'inside-out' ionization…….

3. Accurate calculation requires a far more rigorous description of the atomic structure than at present

4. Estimated two photon $4d^{-1}$ emission is ~1% of total at ~ 7 x 10^{15} $W.cm^{-2}$



Read the full story here…….



PRL **105,** 013001 (2010) PHYSICAL REVIEW LETTERS week ending 2 JULY 2010

Two-Photon Inner-Shell Ionization in the Extreme Ultraviolet

V. Richardson,[1] J. T. Costello,[1] D. Cubaynes,[2] S. Düsterer,[3] J. Feldhaus,[3] H. W. van der Hart,[4] P. Juranić,[3] W. B. Li,[3,5]
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We have observed the simultaneous inner-shell absorption of two extreme-ultraviolet photons by a Xe atom in an experiment performed at the short-wavelength free electron laser facility FLASH. Photoelectron spectroscopy permitted us to unambiguously identify a feature resulting from the ionization of a single electron of the 4*d* subshell of Xe by two photons each of energy (93 ± 1) eV. The feature's intensity has a quadratic dependence on the pulse energy. The results are discussed and interpreted within the framework of recent results of ion spectroscopy experiments of Xe obtained at ultrahigh irradiance in the extreme-ultraviolet regime.

DOI: 10.1103/PhysRevLett.105.013001 PACS numbers: 32.80.Rm, 32.80.Fb, 32.80.Hd, 42.50.Hz

PRL **105** 013001 2010

Resonant 2-photon, 3d Excitation of Kr

1. To date we have looked at a **non-resonant** two photon process (sort of ATI really)

2. FELs are wavelength tunable - one can also explore **resonant two photon** processes

Kr $3d^{10}4s^24p^6$ ($^1S_0$) + 2 x hν (46 eV) -> $3d^94s^24p^64d$ (J=0,2)

i.e., 3d - 4d two photon resonant excitation

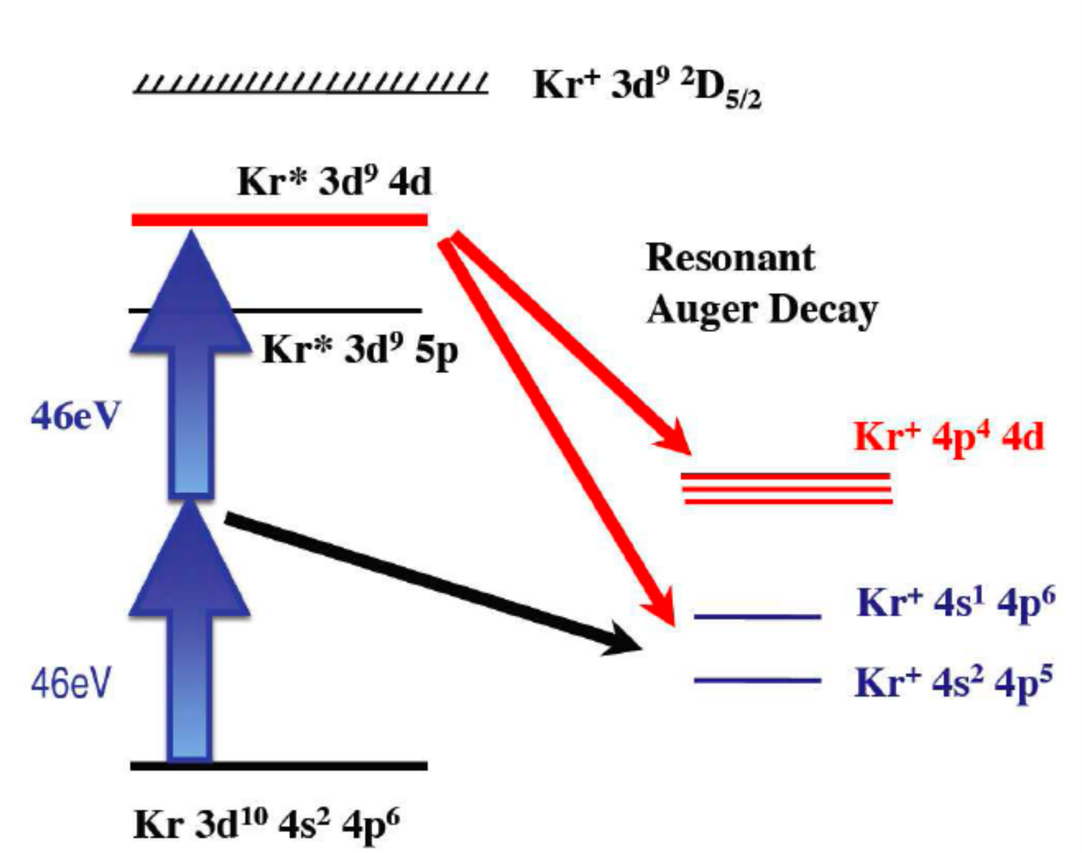
Kr - *Resonant* Two Photon Excitation

1. ***Kr $3d^{10}4s^24p^6$ ($^1S_0$) + 2 x hν (46 eV) -> $3d^94s^24p^64d$ (J=0,2) i.e.,*** 3d - 4d two photon excitation

2. Of course there is a direct ionization path and the usual interference results - manifested as asymmetric resonance profiles (Fano/ Fano-Mies)

3. But here the $3d^94s^24p^64d$ (J=0,2) resonance undergoes Auger decay to Kr^+ on a femtosecond timescale - similar to the FLASH pulse duration - so competition between excitation and decay (ergo, in addition to simple ATI, this case makes for an intriguing, problem for theory)..

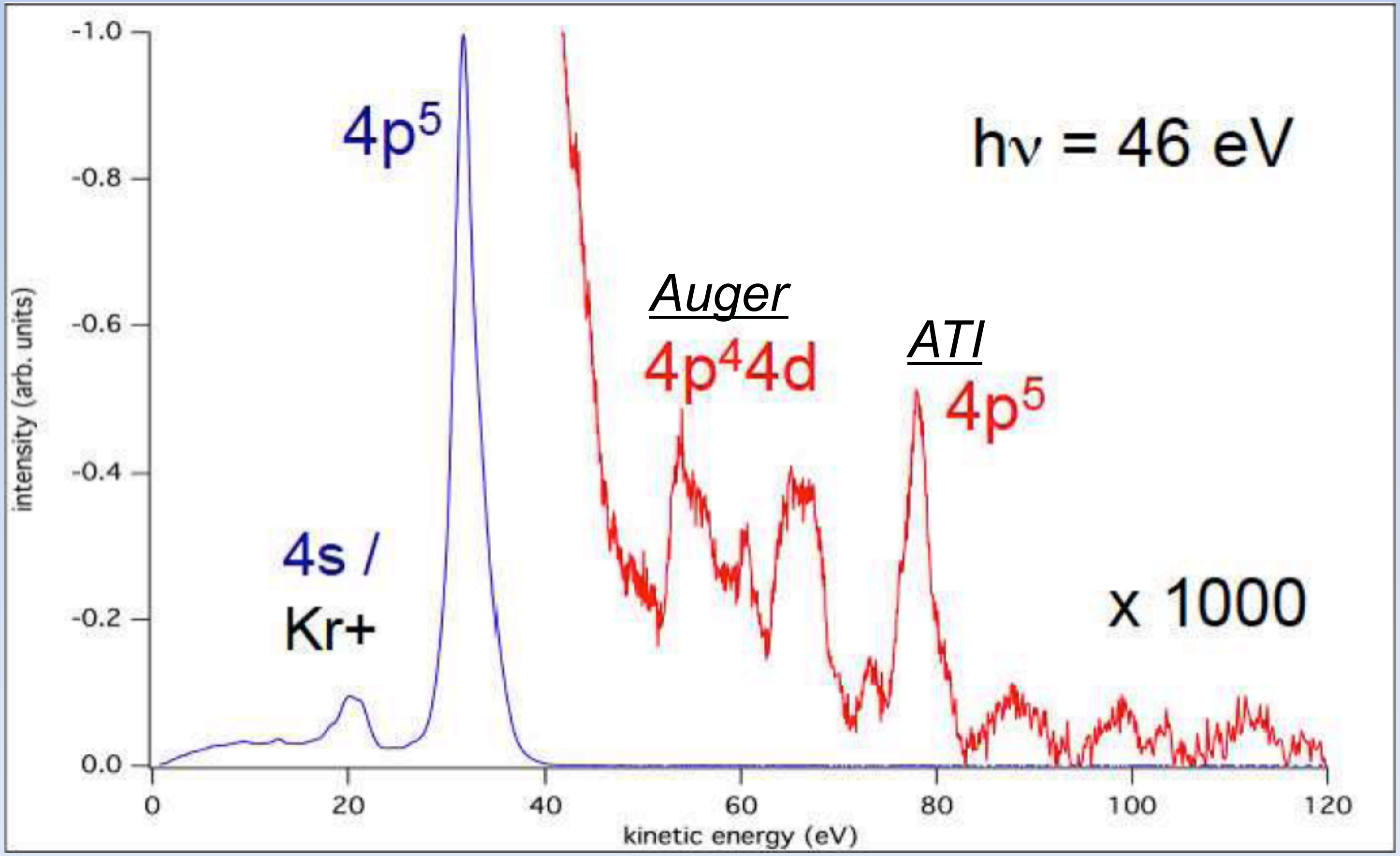
hν = 46 eV (~27 nm)



Meyer et al., PRL **104** 213001 (2010)

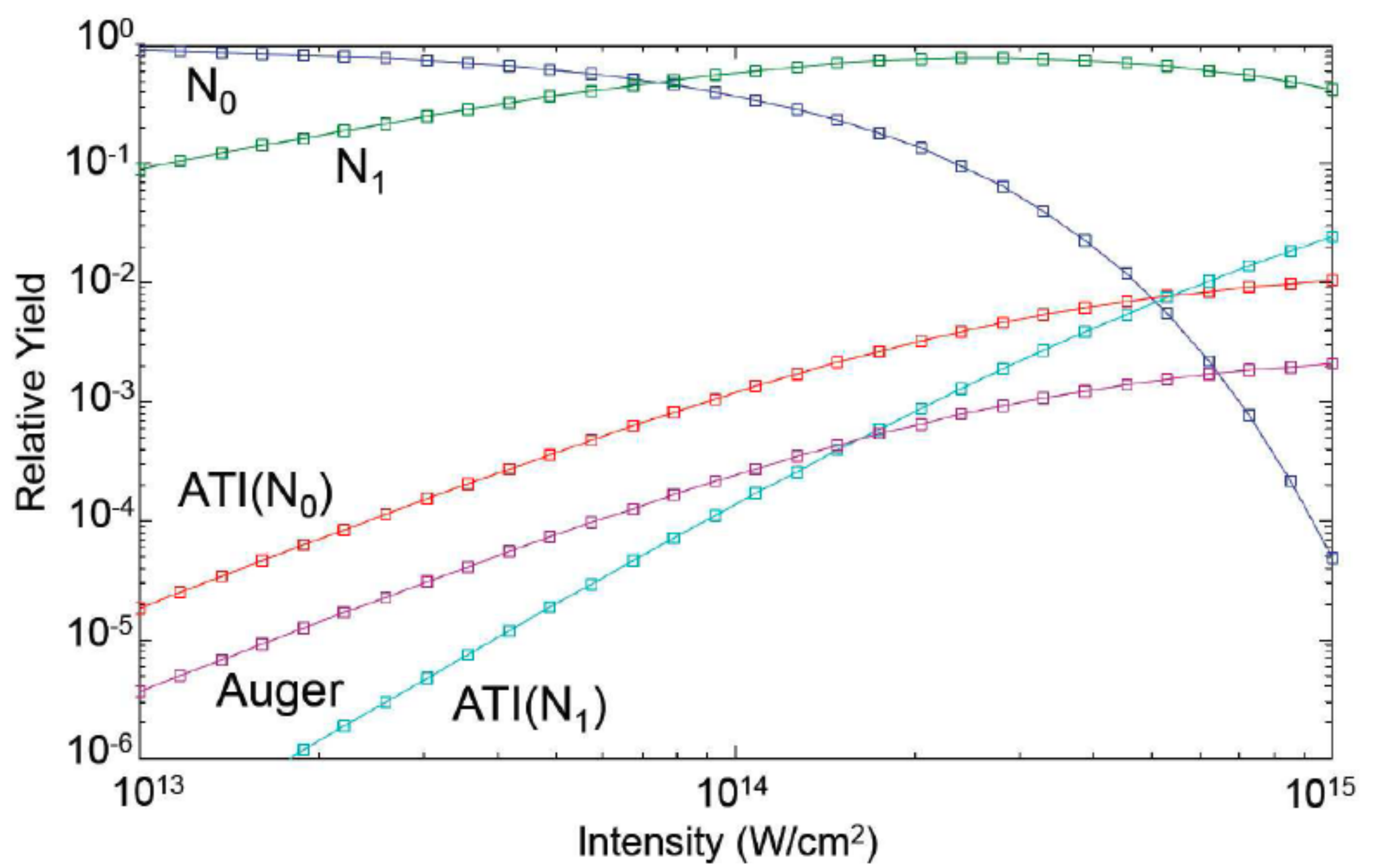
Kr ($3d^94d$) 2 Photon Resonance Auger

MBES Photoelectron spectrum - ~ 5 x 10^{14} W.cm^{-2}

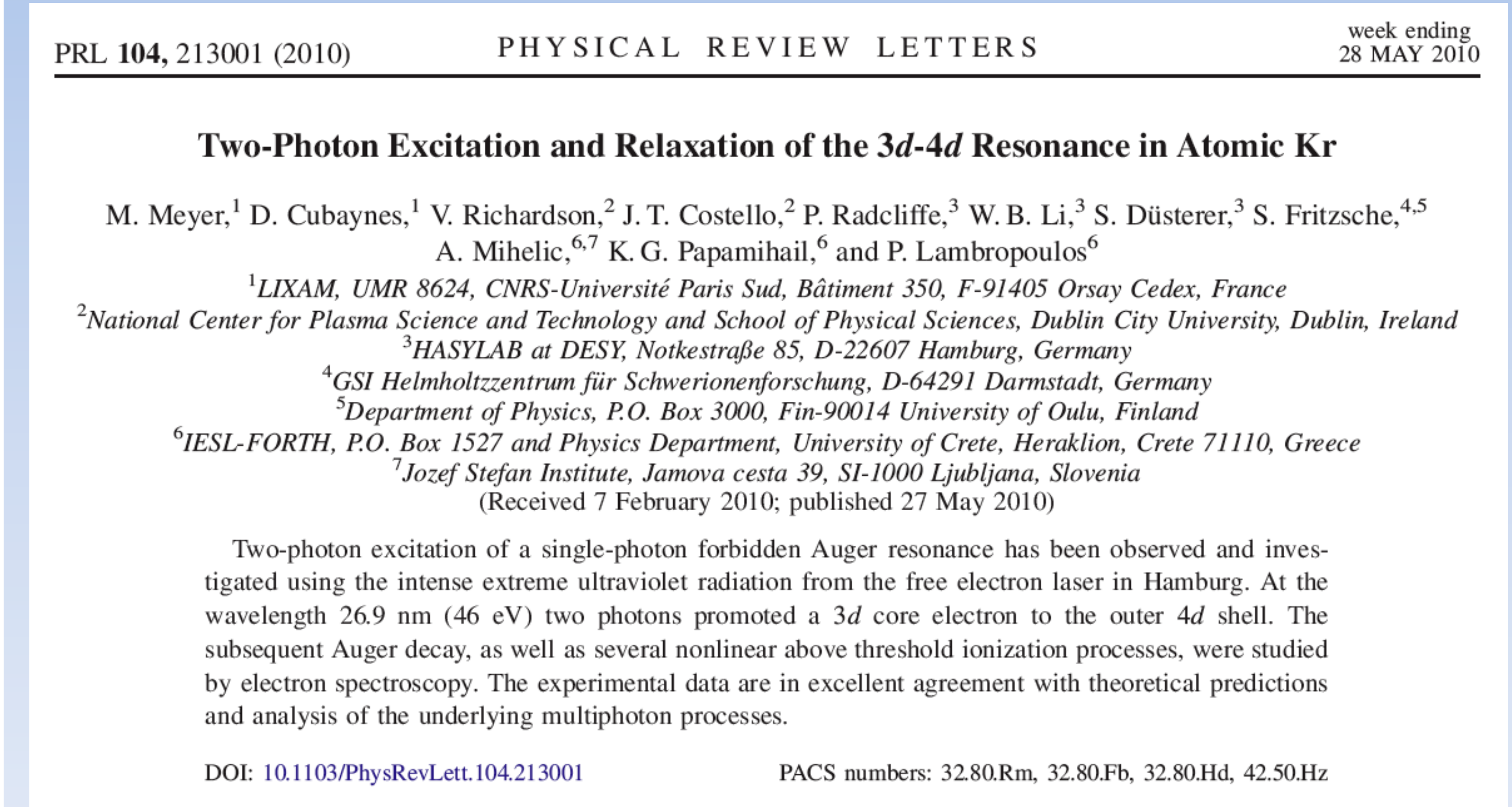


Kr ($3d^94d$) 2 Photon Resonance Auger

Ionization rates – P. Lambropoulos, Crete



Details here……..



PRL **104,** 213001 (2010) PHYSICAL REVIEW LETTERS week ending 28 MAY 2010

Two-Photon Excitation and Relaxation of the 3*d*-4*d* Resonance in Atomic Kr

M. Meyer,[1] D. Cubaynes,[1] V. Richardson,[2] J. T. Costello,[2] P. Radcliffe,[3] W. B. Li,[3] S. Düsterer,[3] S. Fritzsche,[4,5]
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(Received 7 February 2010; published 27 May 2010)

Two-photon excitation of a single-photon forbidden Auger resonance has been observed and investigated using the intense extreme ultraviolet radiation from the free electron laser in Hamburg. At the wavelength 26.9 nm (46 eV) two photons promoted a 3*d* core electron to the outer 4*d* shell. The subsequent Auger decay, as well as several nonlinear above threshold ionization processes, were studied by electron spectroscopy. The experimental data are in excellent agreement with theoretical predictions and analysis of the underlying multiphoton processes.

DOI: 10.1103/PhysRevLett.104.213001 PACS numbers: 32.80.Rm, 32.80.Fb, 32.80.Hd, 42.50.Hz

PRL **104** 213001 (2010)

Summary - One Colour

- Xenon - First detection of a so-called ‘above threshold ionization’ (ATI) two-photon process involving an *inner shell electron* shell.

- 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 - first step on the road to resonant NL processes with EUV/X-rays….* ***REMPI at X-ray.***

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

XUV (X-ray) + IR Ionization

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LCLS Experiments: 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 and dressing fields

Two Extremes:

X-ray pulse duration is ‘many’ optical cycles
X-ray pulse duration is less than ½ optical cycle

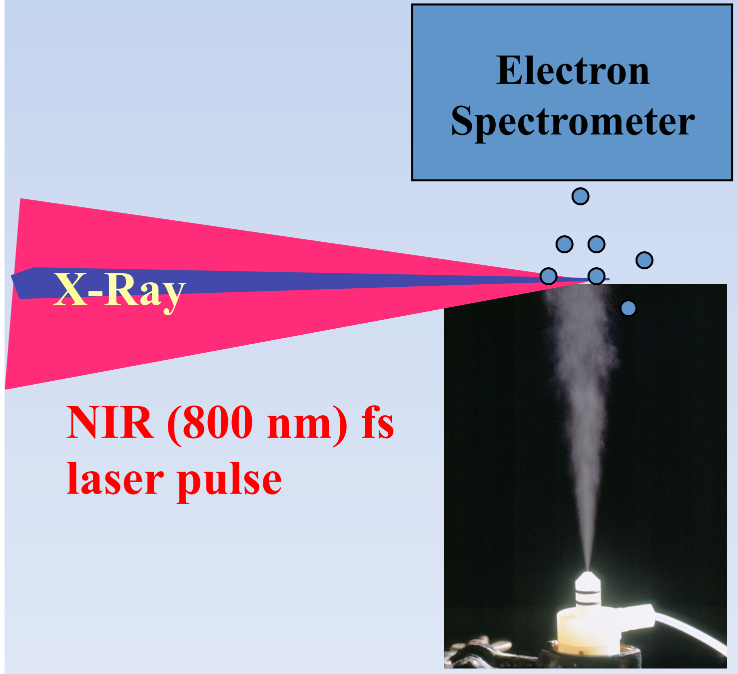
What about the intermediate (few optical cycle) regime ?

Atoms in 'Long' X-ray + IR Fields

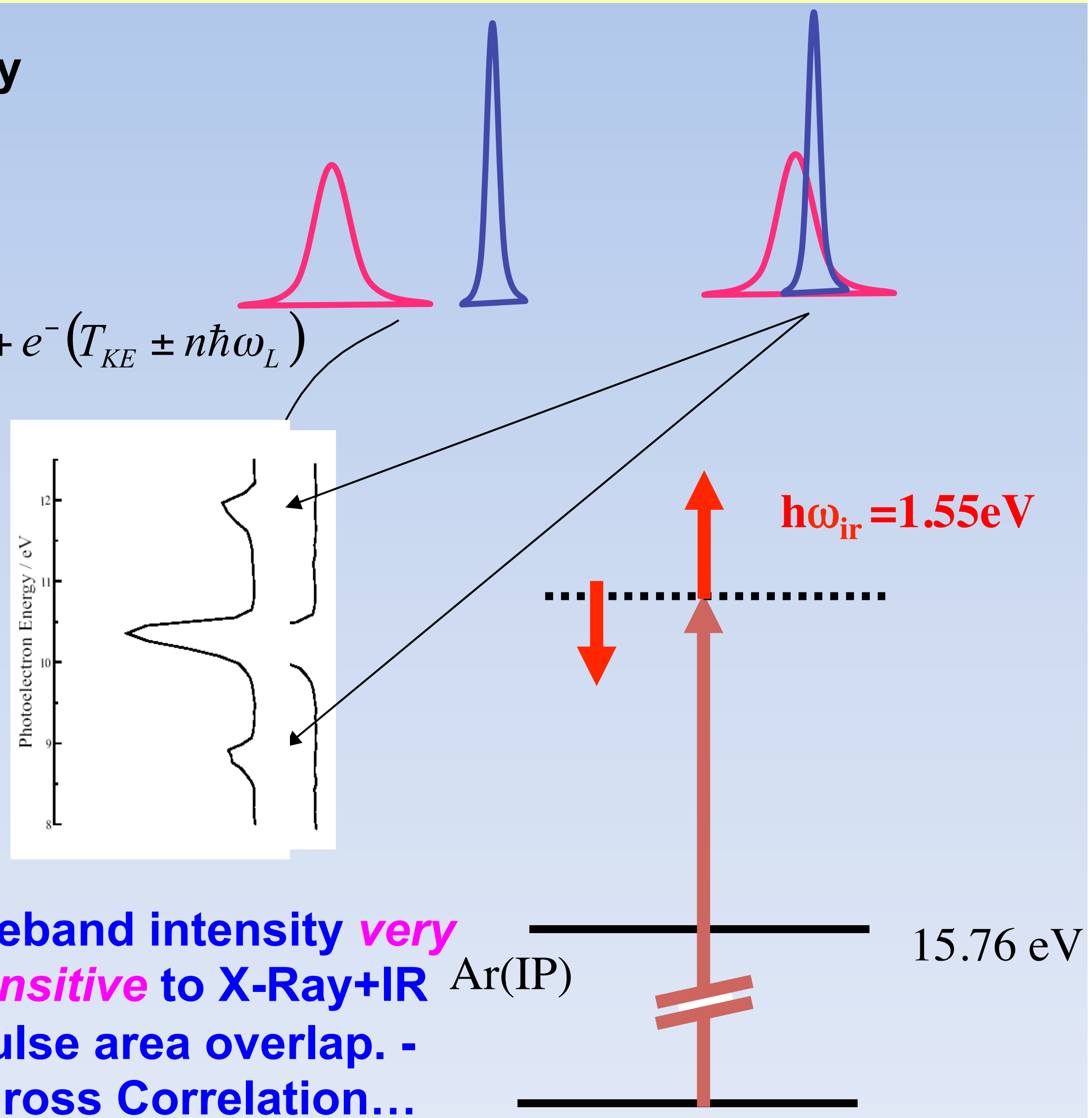
Superposition of visible and X-Ray pulses in a noble gas jet

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

$$A+\hbar\omega_{XUV} \rightarrow A^{+}+e^{-}(T_{KE}) \pm n\hbar\omega_{L} \rightarrow A^{+}+e^{-}(T_{KE} \pm n\hbar\omega_{L})$$



Gas Jet

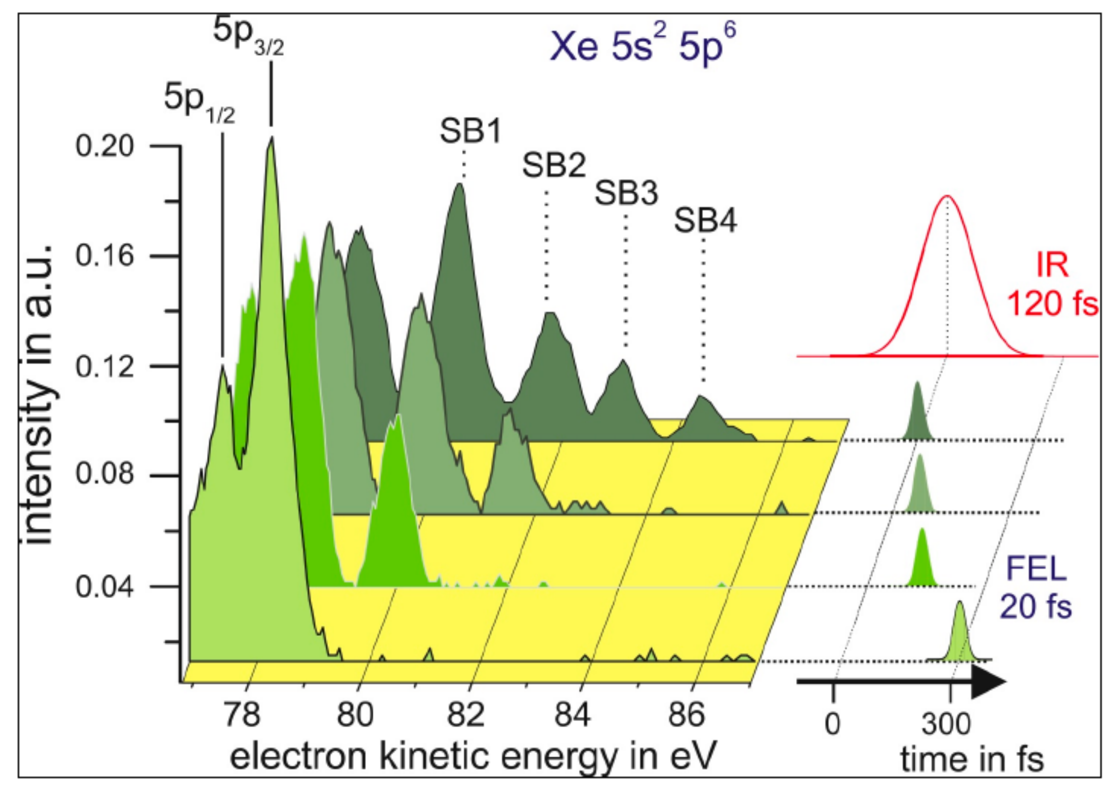


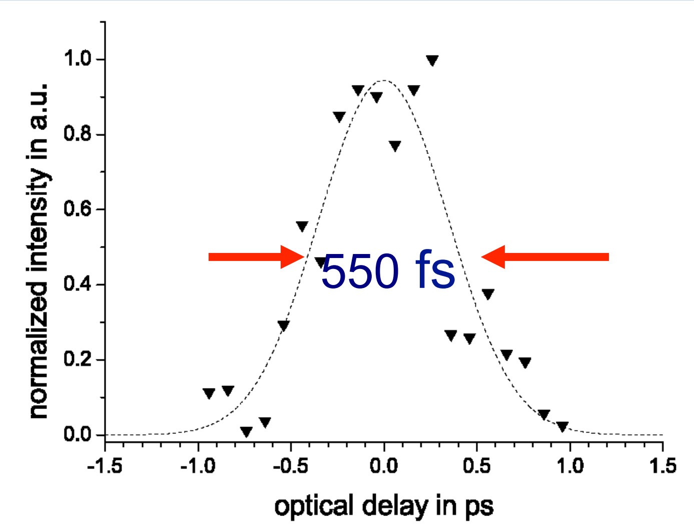
Sideband intensity *very sensitive* to X-Ray+IR pulse area overlap. - Cross Correlation…

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

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)

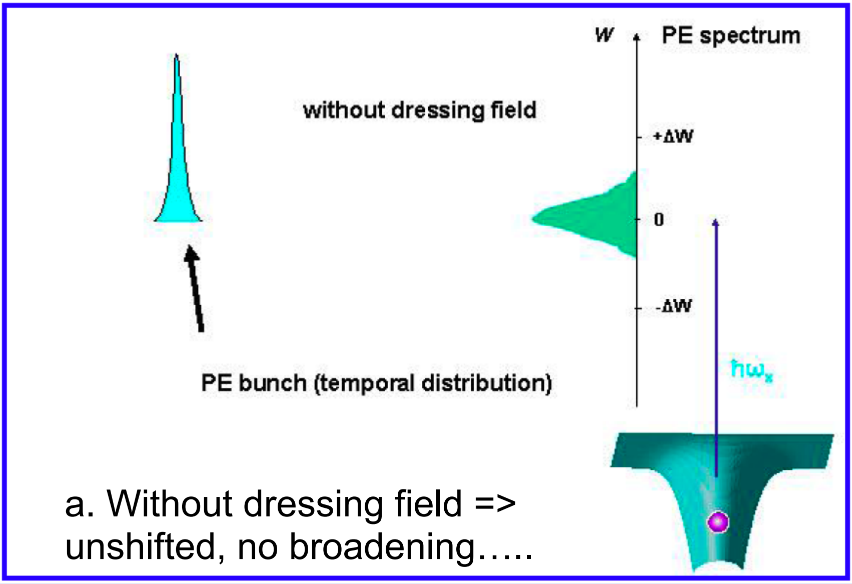
2. 'TEO'

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

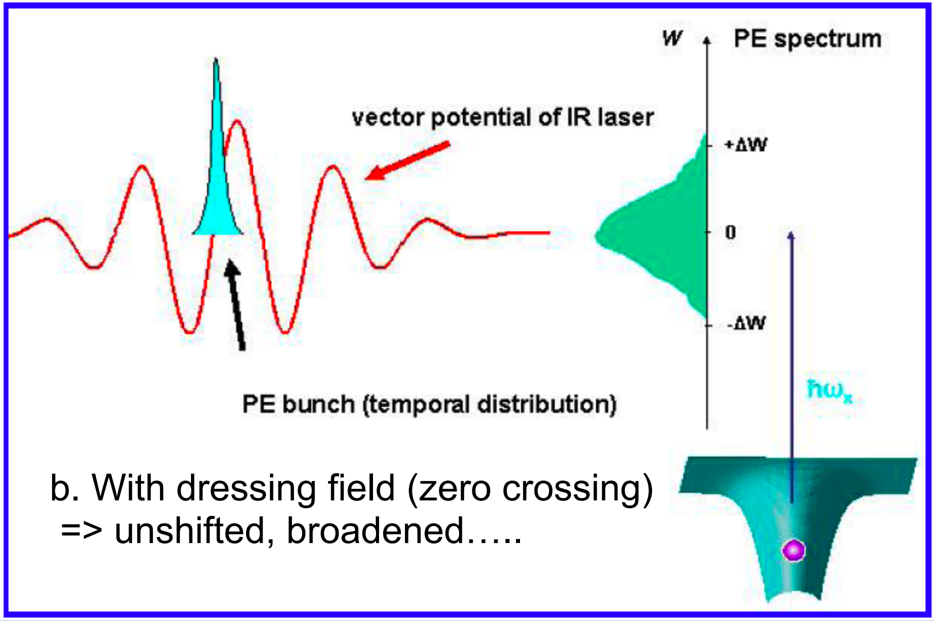
NIMA **83**, 516-525 (2007)

Appl. Phys. Lett **90** 131108 (2007)

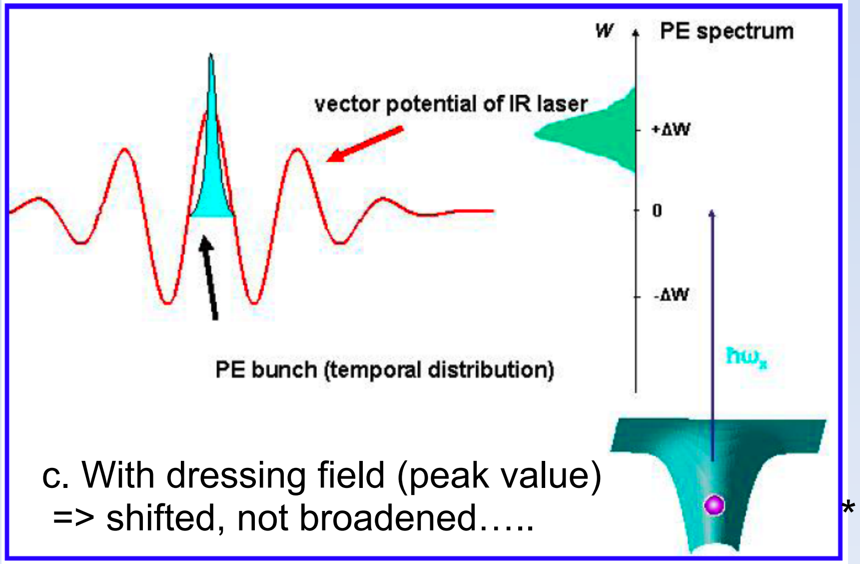
Atoms in ‘Short’ XUV (X-ray) + IR Fields



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)*

Aside: Measurement of few fs pulses @ LCLS

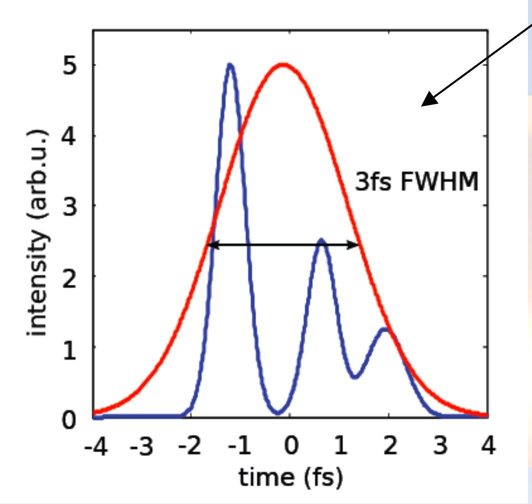
LCLS low current/slotted spoiler/ few fs mode -

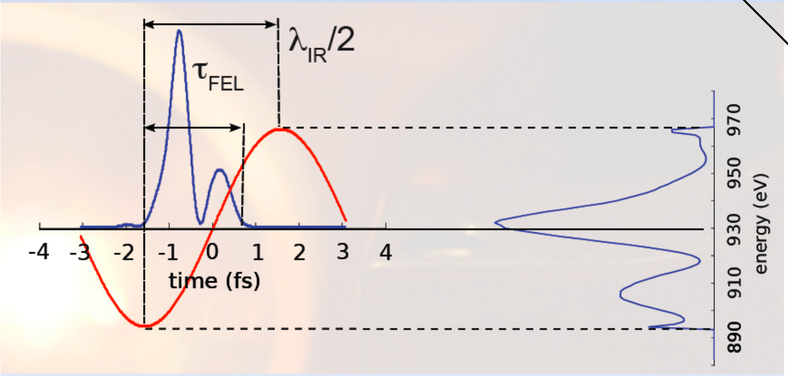
Data still under analysis……

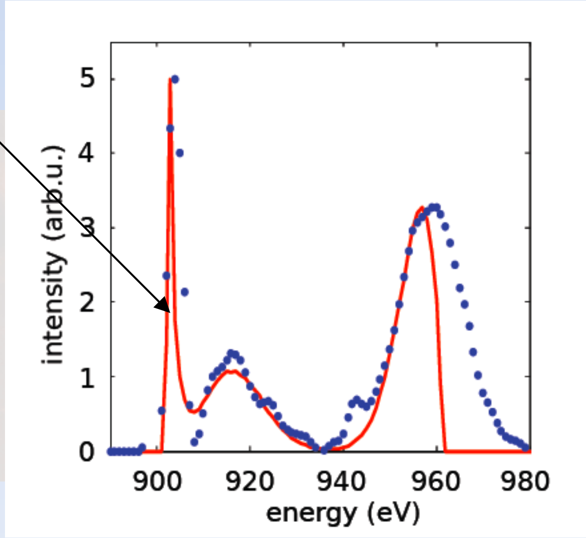
Process. Ne + hν (1.8 keV) -> Ne$^+$ (1s^{-1}) + e- +I$_L$ (10^{14} W.cm^{-2})

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………



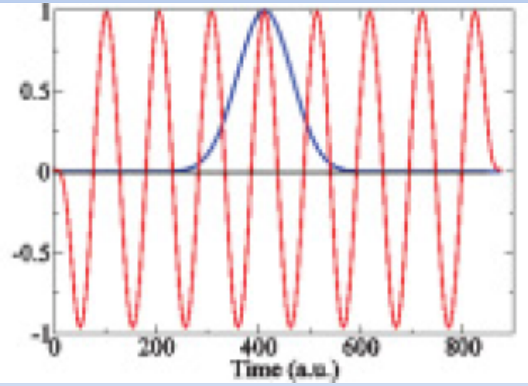




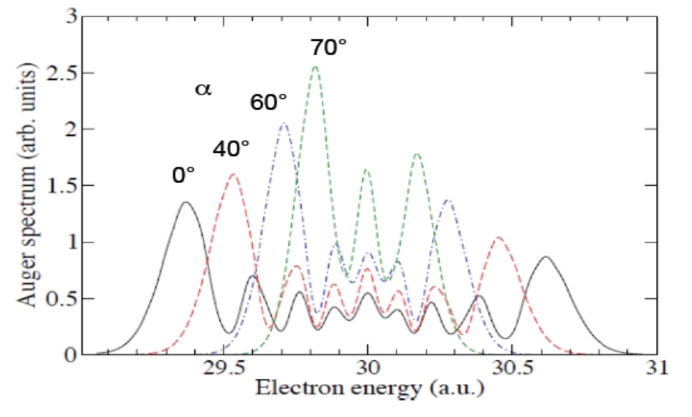
Intermediate regime–sub-optical cycle effects

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

Auger lifetime similar to optical (800 nm) cycle



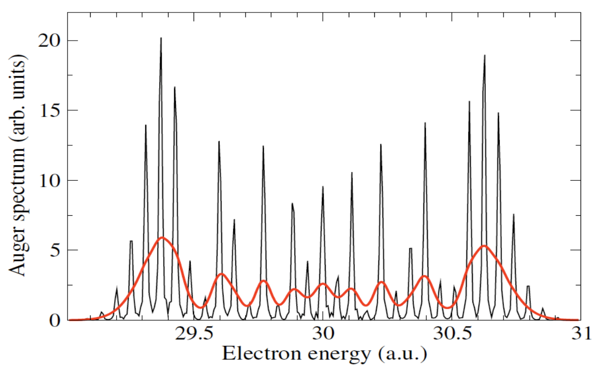
Angle Resolved Sideband Spectra



Core hole lifetime
τ (Ne 1s) = 2.4fs

Optical cylce
T (800nm) = 2.6fs

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



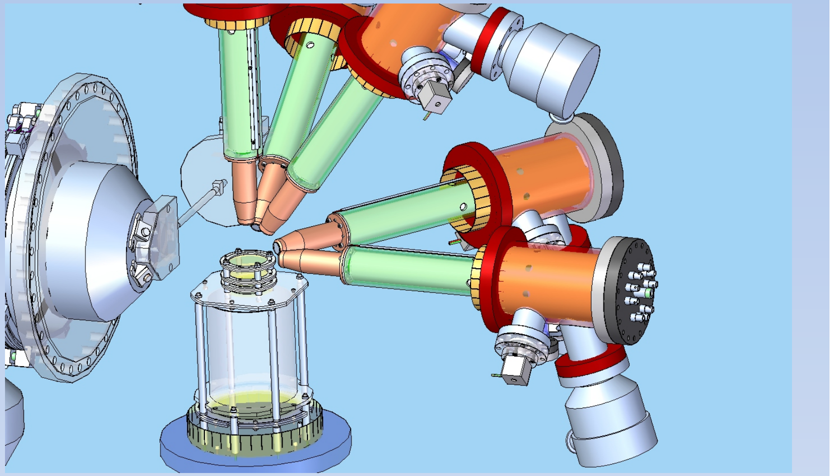
LCLS: 1 keV, 2-5 fs

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

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

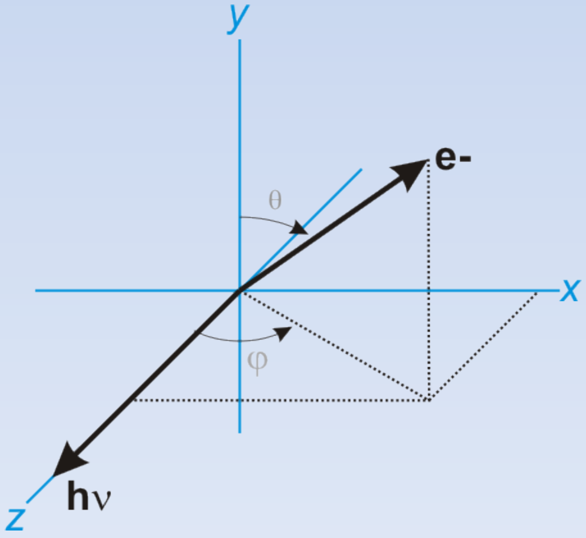
AMO Chamber and Specifications

High Field Chamber (AR-eTOF)



1. Based on a successful design used by Denis Lindle's 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

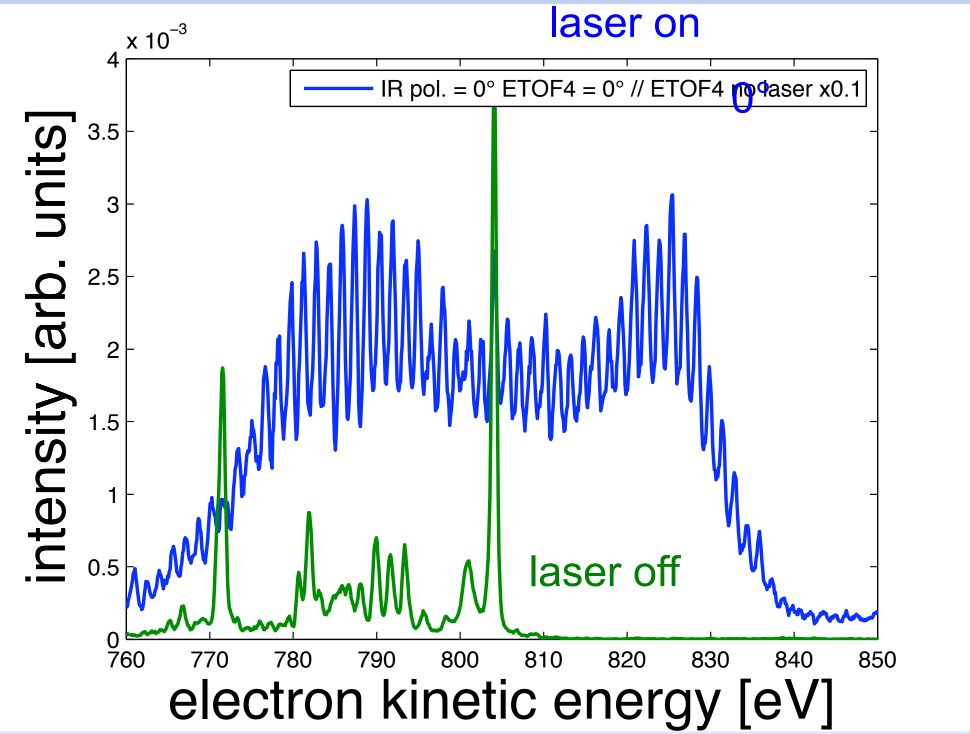
SB modulation – few/sub-optical cycle effects

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

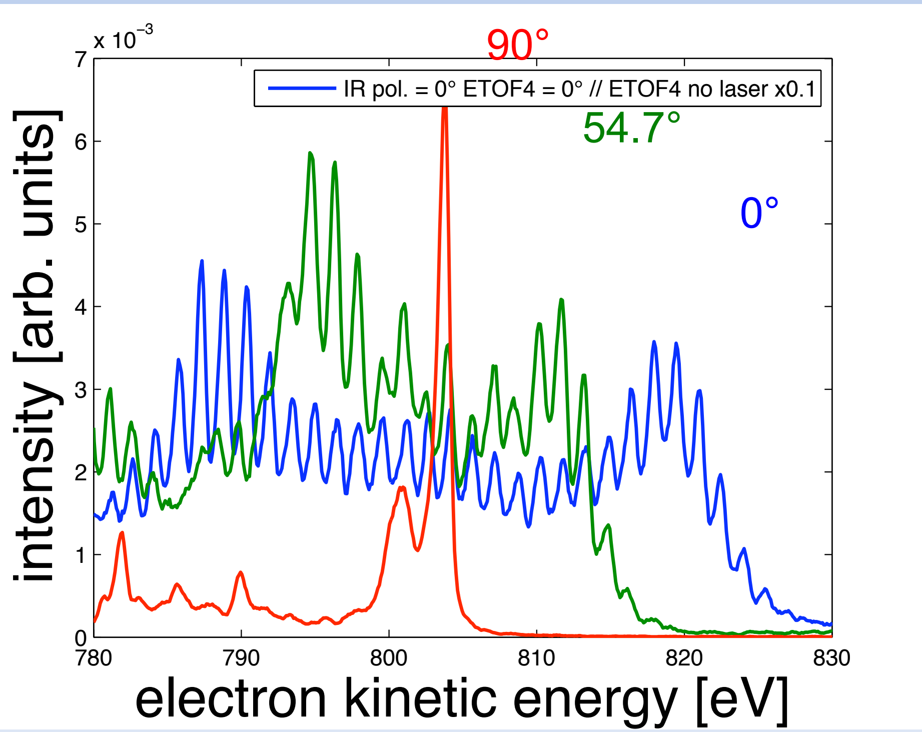
NIR : 800nm, 1 mJ, 3ps

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

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



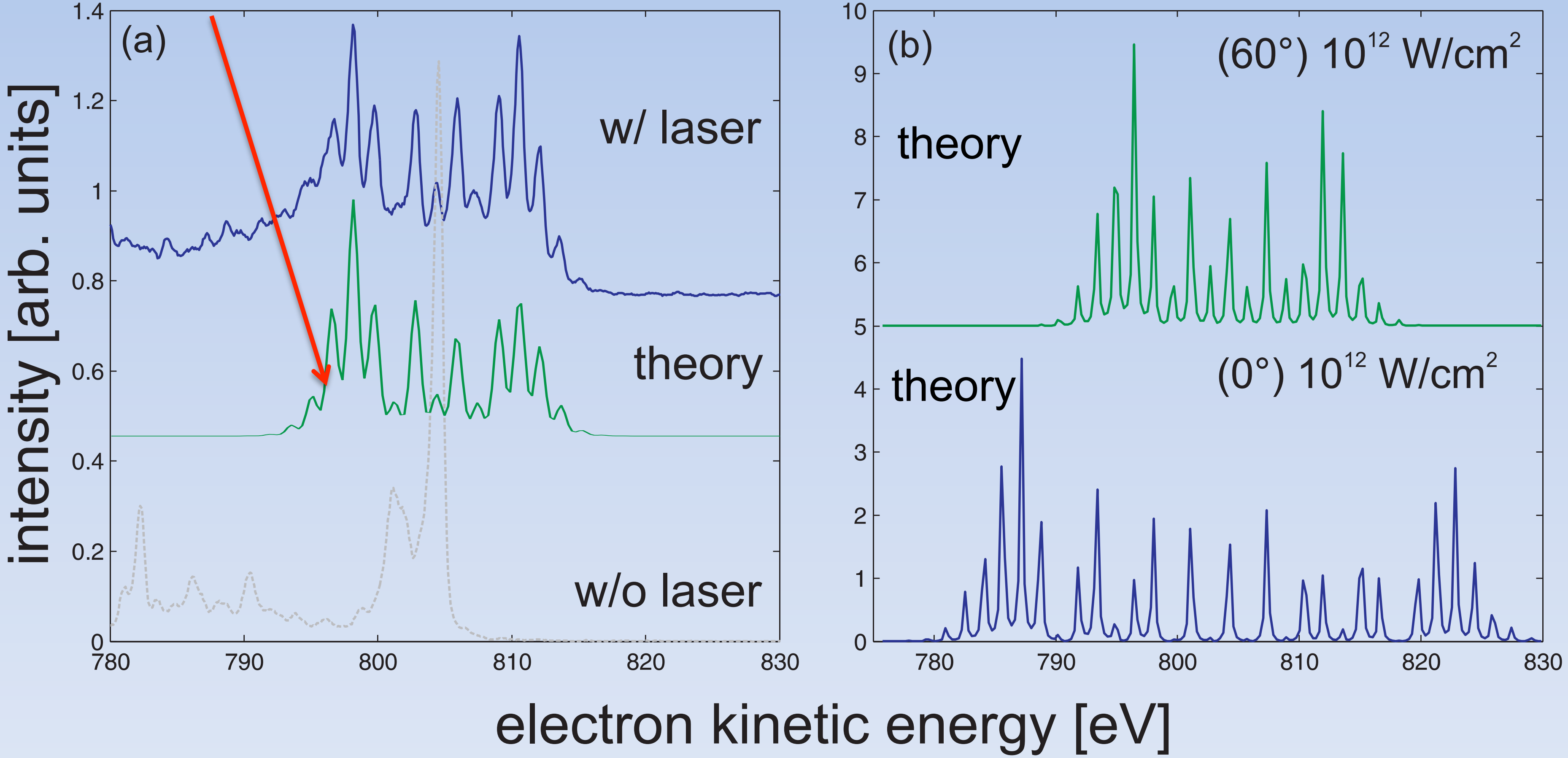
Strong sideband structure



Strong angular effect

SB modulation – few/sub-optical cycle effects

Theory – accounting for spatial variation of the laser field



PRL, Just accepted, out early 2012…….

Next Steps

1. SASE-FELs, Operating Principle and Characteristics
2. Rudiments of ionization processes in intense laser fields
3. Photoionization experimental setups (FLASH & DESY)
4. Two photon ionization
5. Two colour Ionization
6. **Next Steps**

Erasmus Mundus Joint Doctorate

‘EXTATIC’ – Extreme-ultraviolet & X-ray Technology And Training for Interdisciplinary Cooperation

http://www.extatic.eu

Closing date: Mid January 2012……….

http://www.extatic.eu

Academic Partners include:

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- University College Dublin
- Kings College London
- The University of Southampton
- Czech Technical University-Prague
- The University of Padua
- The Military University of Warsaw
- RWTH-Aachen University

Current associated academic partners include:

- Colorado State University (USA)
- Purdue University (USA)
- Tongji University (China)

Current associated industry/ private laboratory partners include:

- Silson (UK)
- Prevac (Poland)
- *XENOCS (France)*
- EPPRA (France)
- Rigaku Innovative Tech. (Prague)
- Bruker (Germany)
- Fraunhofer-ILT (Germany)
- *XFEL Gmbh (Germany)*

Conclusions – Two Colour

1. We have demonstrated interference free *sidebands (SB) to high order*,
2. Especially useful as an *X-ray-Optical cross correlation technique*
3. Experiments started at LCLS (summer 2009) - SBs have been used to measure 'long' (> 100 fs) pulse widths
4. First 'few fs' streak measurements at LCLS
5. First data on *angular distribution of SB* electrons at LCLS –
6. LCLS will test the limits of UF X-ray pulse width and X-ray optical jitter measurement techniques – 'Tandem streaking' and single cycle THz streaking
7. Next step (physics) - *X-ray laser pumping, coherent control, Rabi flops*,…
8. 2012 and beyond: FLASH - seeding (sFLASH), *LCLS/XFEL - aluminium 'slotted spoiler' for cleaner pulses, seeding (Echo 7 project),…..*