Multiphoton (Inner-Shell) Ionization in Intense EUV FEL Fields

TPI of Xe (93 eV) and Kr (46 eV)

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Outline of Talk

1. FLASH (One slide) Overview

2. Comments on ionization in intense laser fields

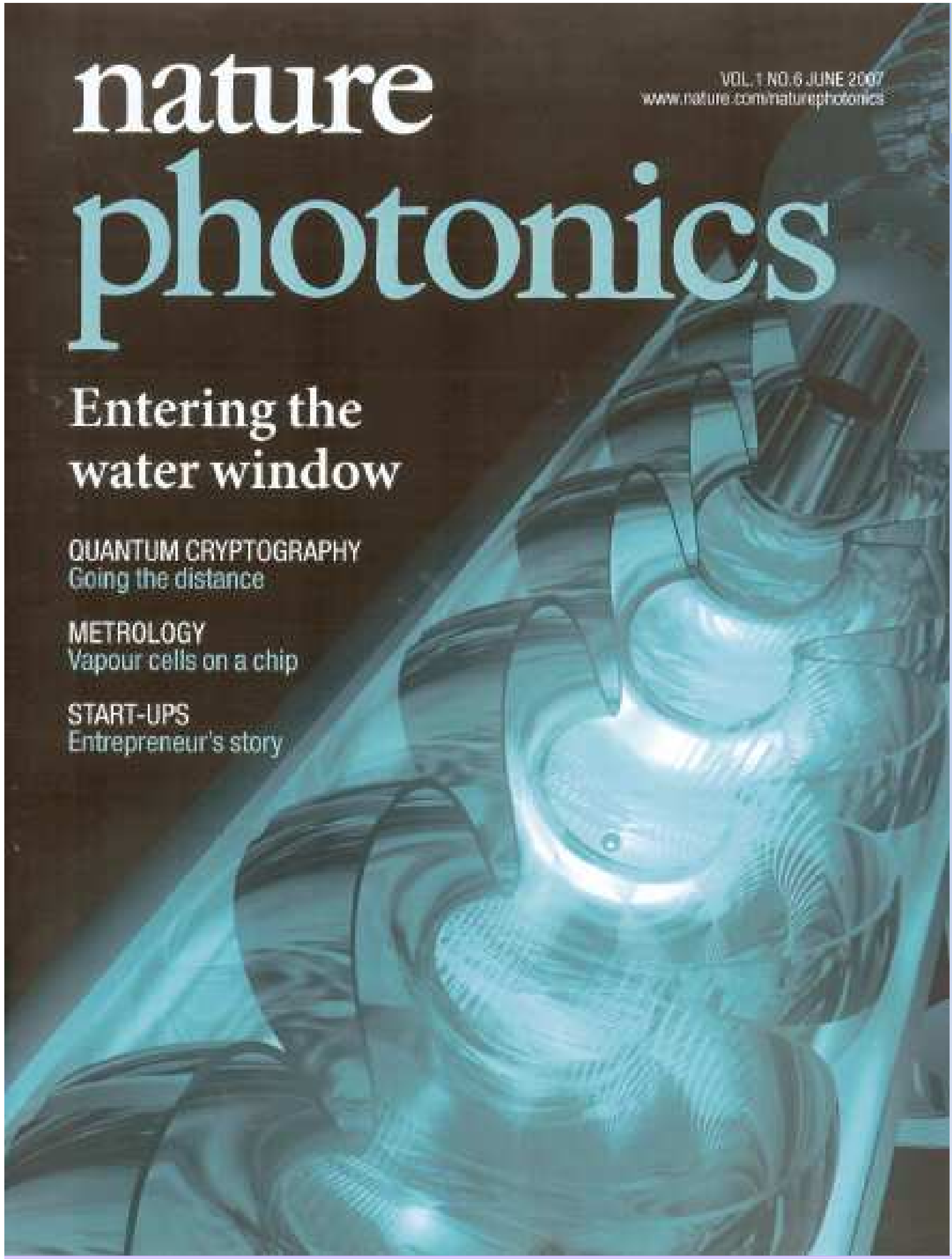
3. Setup for Photoelectron Spectroscopy

4. Two Photon Ionization *inner-shell* of Xe (ATI)

5. *Resonant* Two Photon *inner-shell* Excitation/ Auger Decay in Kr

6. Summary / Conclusions and Next Steps

FLASH:
Key Performance Indicators



BL3 – allows installation of user groups focusing optics

Wavelength – 4.5 nm to 60 nm

Pulse Energy – 10 to 50 µJ

Pulse Length – ~10s fs

Photons per Pulse ~10^{13}

Ackermann et al., Nature Photonics **1** 336 (2007)

What are the USPs of XFELs in AMOP ?

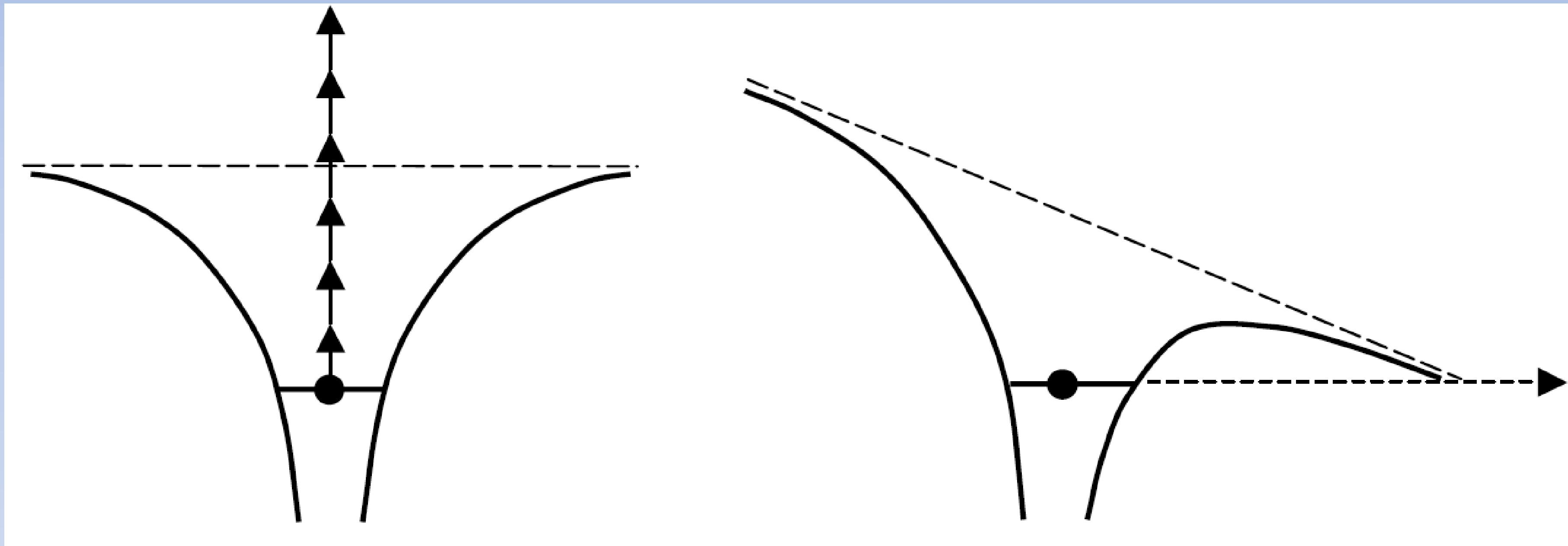
- ***Ultra-dilute*** **targets**
- ***Photo*****-processes with *ultralow cross-sections***
- ***Pump and probe*** **experiments (XUV + XUV or XUV + Opt.)**
- ***Single shot*** **measurements**
- ***Few-photon*** **single and multiple *ionization processes***
- **Makes *inner-shell* electrons key actors in non-linear processes for the first time**
- **Re-asserts *primacy of the photon* over field effects !**

Keldysh - Ionization Regimes

Multiphoton Ionization — $\gamma >> 1$

Tunnel Ionization — $\gamma \sim 2$

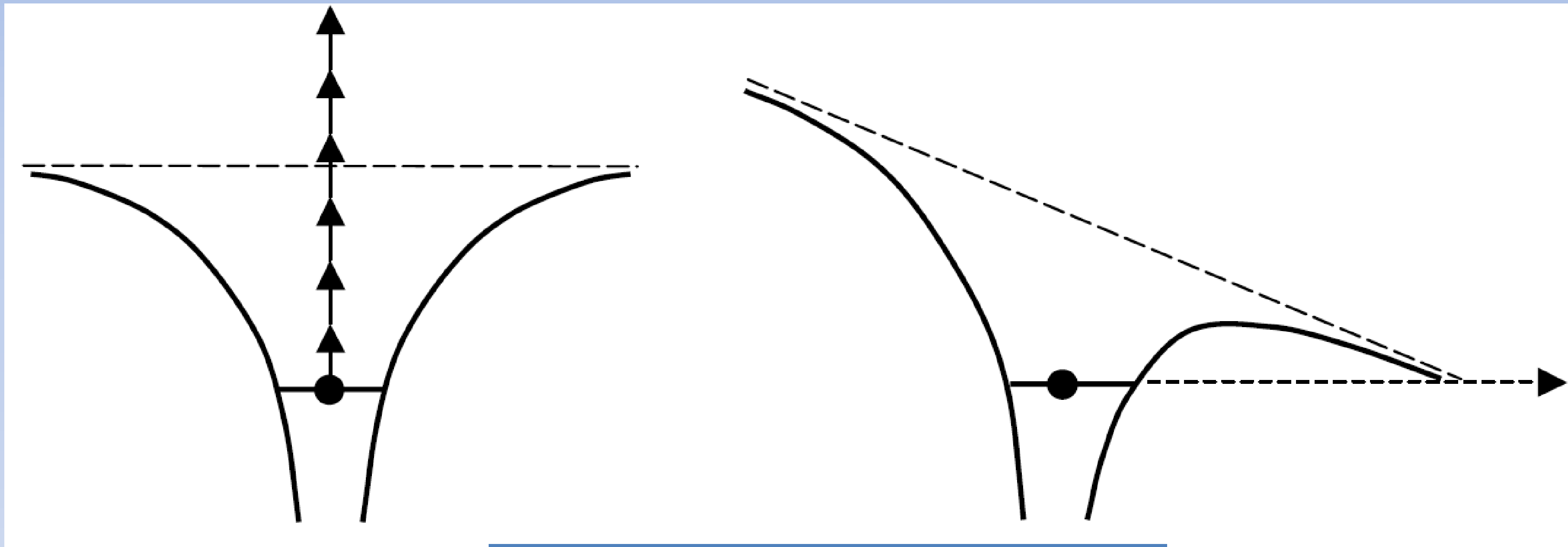
Field Ionization — $\gamma << 1$



$$\gamma = \sqrt{\frac{IP}{2U_p}} \quad \text{where} \quad U_P = 9.3\times10^{-14} I\left(Wcm^{-2}\right)\lambda^2(\mu m) \quad eV$$

Keldysh - Ionization Regimes

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



Intensity/ Wavelength →

← **Photon Energy**

FLASH@DESY offers….

High intensity - 100μJ/10fs/10μm ~ 10^{16} W/cm^2

Interaction with matter

Drive non-linear photoionization processes…..?

At high (EUV) photon energy - up to 300 eV

Keldysh - γ

$$\gamma = \sqrt{\frac{IP}{2U_p}} \quad \text{where} \quad U_P = 9.3\times10^{-14}\, I\left(Wcm^{-2}\right)\lambda^2(\mu m) \quad eV$$

Keldysh - Ionization Regime

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

I (Units of 10^{14} W.cm^{-2})	*U_p (eV)-800 nm*	*U_p (eV)-8 nm*	*γ (800 nm)*	*γ (8 nm)*
0.1	0.59	0.00006	4.50	454
0.5	2.98	0.0003	2.03	203
1	5.95	0.0006	1.44	143
5	29.7	0.003	0.64	64
10	***59.5***	***0.006***	***0.45***	***45***
100	595.2	0.06	0.14	14

Ti-Sapphire in the NIR
Non-Pertubative (TI) Regime

FLASH in the EUV -
Pertubative (MPI) Regime:

So these non linear photoionization processes will involve *predominantly* ***few photons*** and inner and/or ***few electron***....

Consequence - ion yield scales with intensity as I^n

What's really important about NLO/S with EUV/X-ray Lasers ?

- Importantly - EUV/X-ray FELs *bring inner shell electrons into the non-linear interaction* of radiation with matter for the first time…..

- So Autoionising states (with femtosecond lifetimes) can play a key role in the process…. This will lead to a *complex dynamical interaction* between X-ray excitation and decay which means that simple 'Single Active Electron - SAE' models will no longer suffice……

- Models that can combine and capture the physical competition between pumping and rapid (mainly) non-radiative decay of small quantum systems, along with a gamut of other parasitic/competitive NL processes (e.g., ATI) in intense EUV/X-ray fields are now needed…….

General EUV / XFEL AMOP Refs

Summary of AMOP@FLASH
http://hasylab.desy.de/science/user_collaborations/amopflash

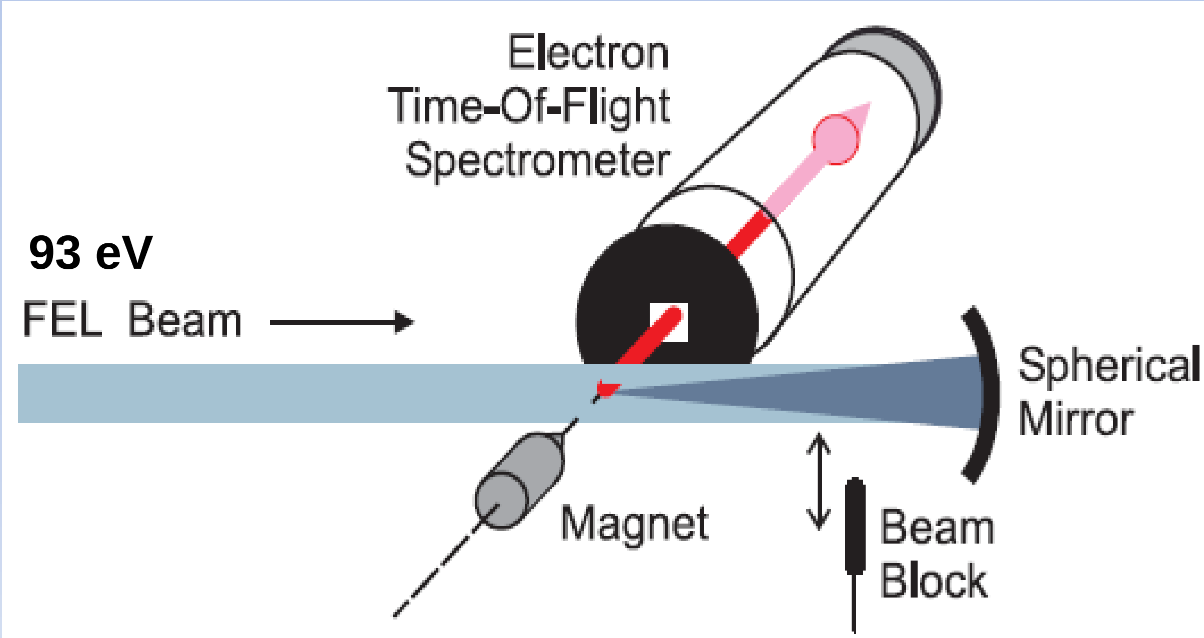
1. Photoionization Experiments with the Ultrafast XUV Laser FLASH
J T Costello, J Phys Conf Ser **88** Art No 012057 (2007)

2. Experiments at FLASH
C. Bostedt et al., Nucl. Inst. Meth. in Res. A **601** 108 (2009)

3. Non-linear processes in the interaction of atoms and molecules with intense EUV and X-ray fields from SASE free electron lasers (FELs),
N. Berrah et al., Journal of Modern Optics (*in Press 2010*)

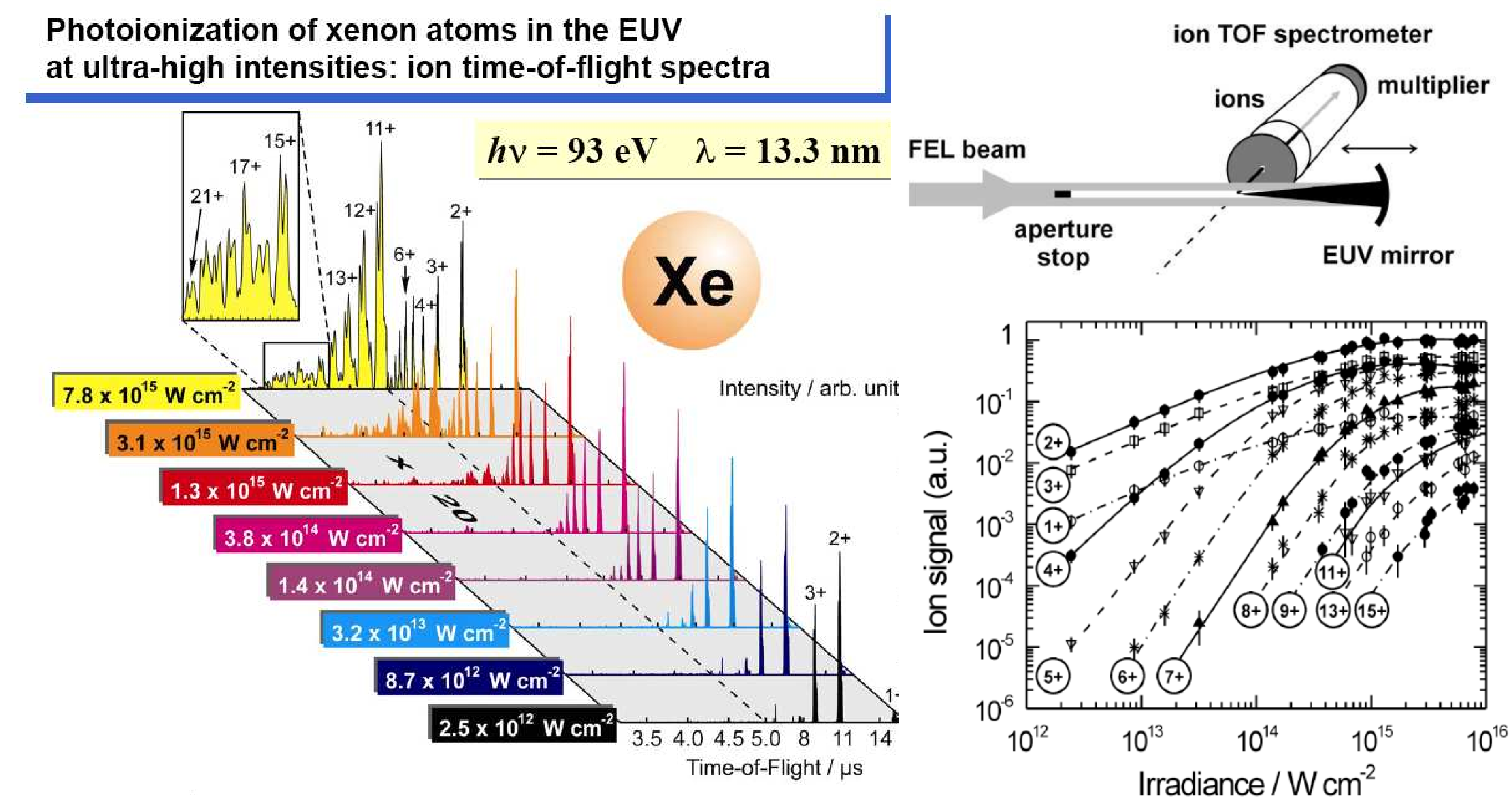
3. Photoelectron Spectroscopy Setup

- High quality, Si/Mo multi-layer mirror employed – spot size at focus ~ 4µm → 10^{16}Wcm^{-2} (IOF-Jena)
- Gas Monitor Detector (GMD) provides shot-to-shot measure of FEL pulse intensity
- 0.65m TOF e^{-} - spectrometer - directional magnetic field to maximise collection efficiency

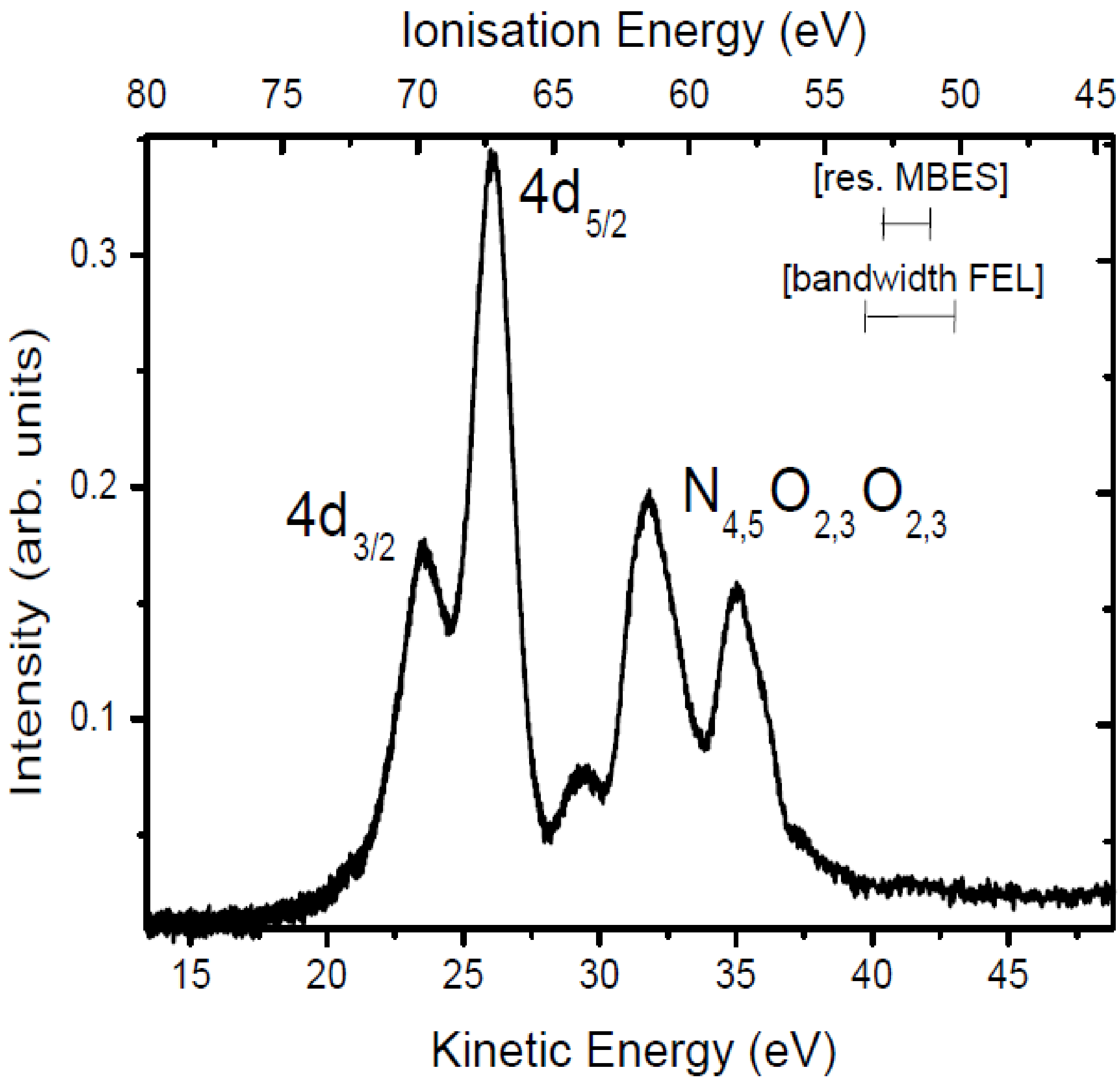


4. Xe ionization in intense XUV fields

Motivation - *Sorokin, Bobashev, Richter et al., PTB, PRL 2007*



One Photon Ionisation at 93 eV



- For low intensities ($<10^{14} Wcm^{-2}$), one photon processes are dominant
- Salient features – 4d photoelectron line with s/o split + Auger electron spectrum
- Single shot……
- Not shown – $5s^{-1}$ and $5p^{-1}$ lines at higher KEs

Two Photon *Inner Shell* Ionisation in Xe

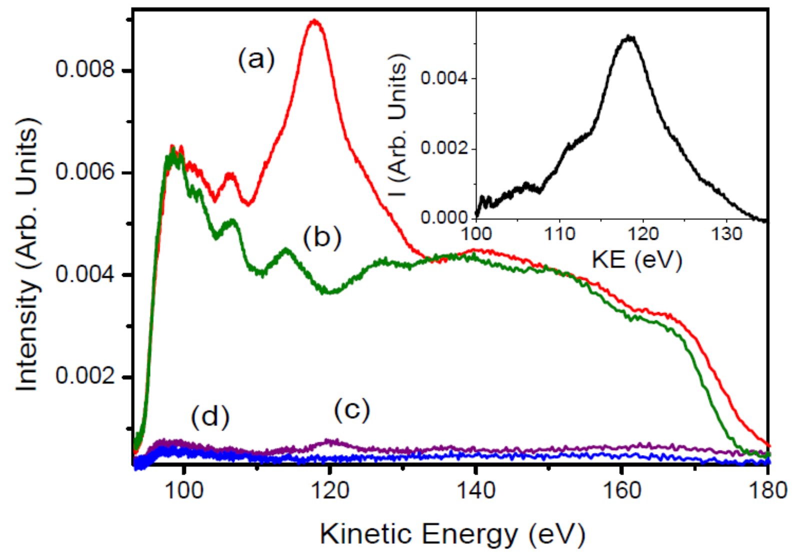
- Using MBES, first evidence of two photon ***inner*** shell ionisation, in this case of 4d electron – $Xe + 2h\nu \rightarrow Xe^{+}\ 4d^{9} + e^{-}$

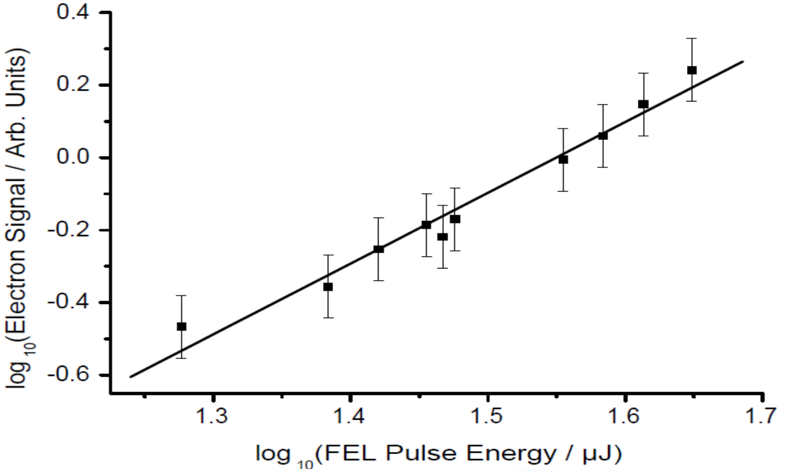
- 'Retardation field' applied to suppress low KE electrons (one photon processes)– electron detected due solely to multiphoton events

- Energetically –

  2 × (93) eV – 118 eV = 68 eV

- Yield scales quadratically, n=1.95 ± .2





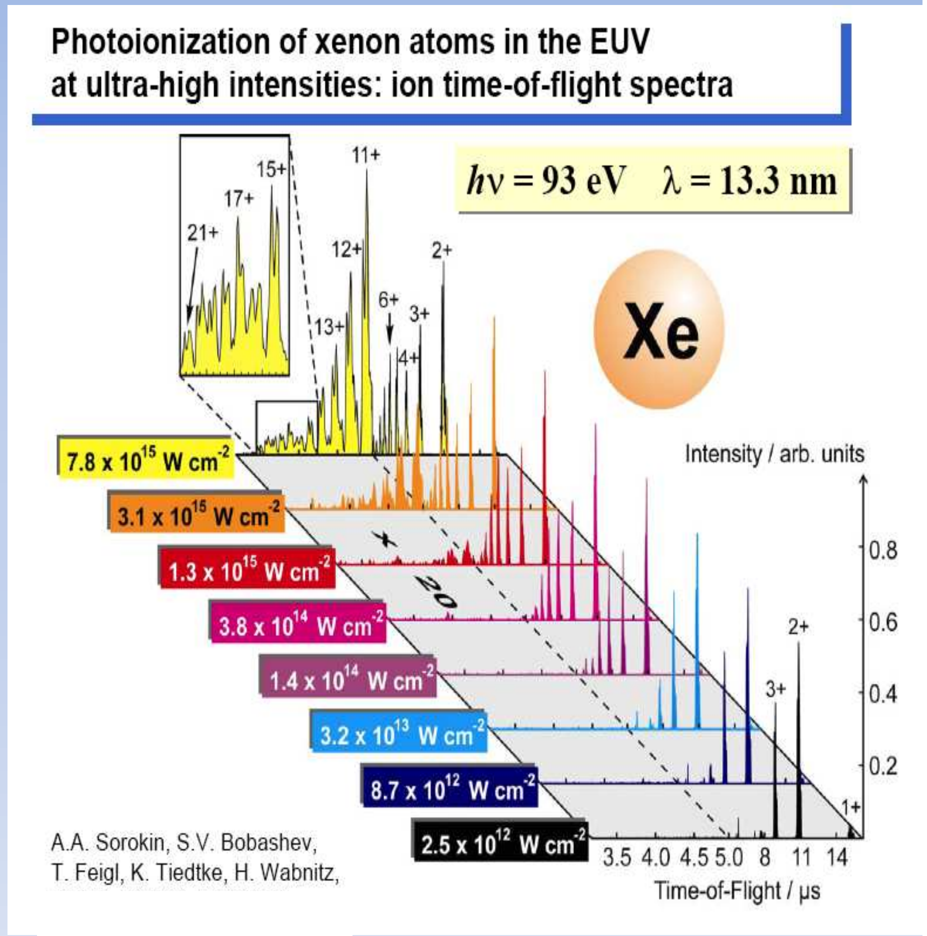
Two Photon *Inner Shell* Ionisation in Xe

1. However, from PRL 99 (2007) 213002 one may conclude that the FEL field produces and interacts with a highly ionized target.

2. Xe^{+} has four $4d^{-1}$ ionization thresholds at 71.6 eV, 72.9 eV, 74.9 eV, and 76.2eV - yield photolines with KE from 110 to 115eV. *However, Xe^{+} appears only weakly in the ion spectra even at very high FEL intensity.*

3. $4d^{-1}$ from higher charge states also possible – outside KE region of interest

4. Additionally, two photon O-shell ionisation cross section expected to be weak from Xe^{4+} & Xe^{5}



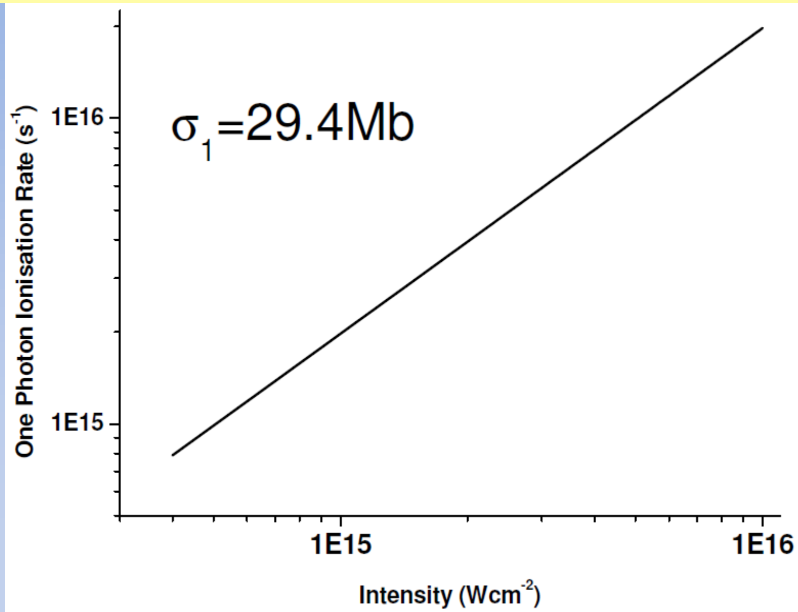
Two Photon Inner Shell Ionisation in Xe

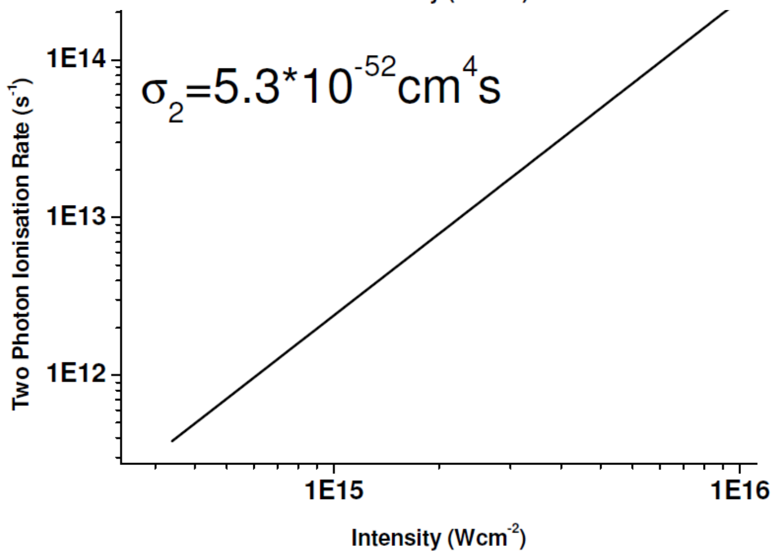
1. Using R-Matrix calculations H.W. Van der Hart) – can calculate one and two photon 4d emission partial cross sections

2. Dominant process at this KE is still one photon – ejection of next 4d – 93 eV can remove next 4d as well - or maybe 4p – removal of the second 4d electron may lead to excitation over a wide range of states.

3. Accurate calculation requires a far more substantial description of the atomic structure

4. Early estimation puts two photon $4d^{-1}$ emission at 0.5-1% of total





V Richardson et al. under review at PRL

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5. *Resonant* two photon Excitation of Kr

1. To date we have looked only at **non-resonant** processes

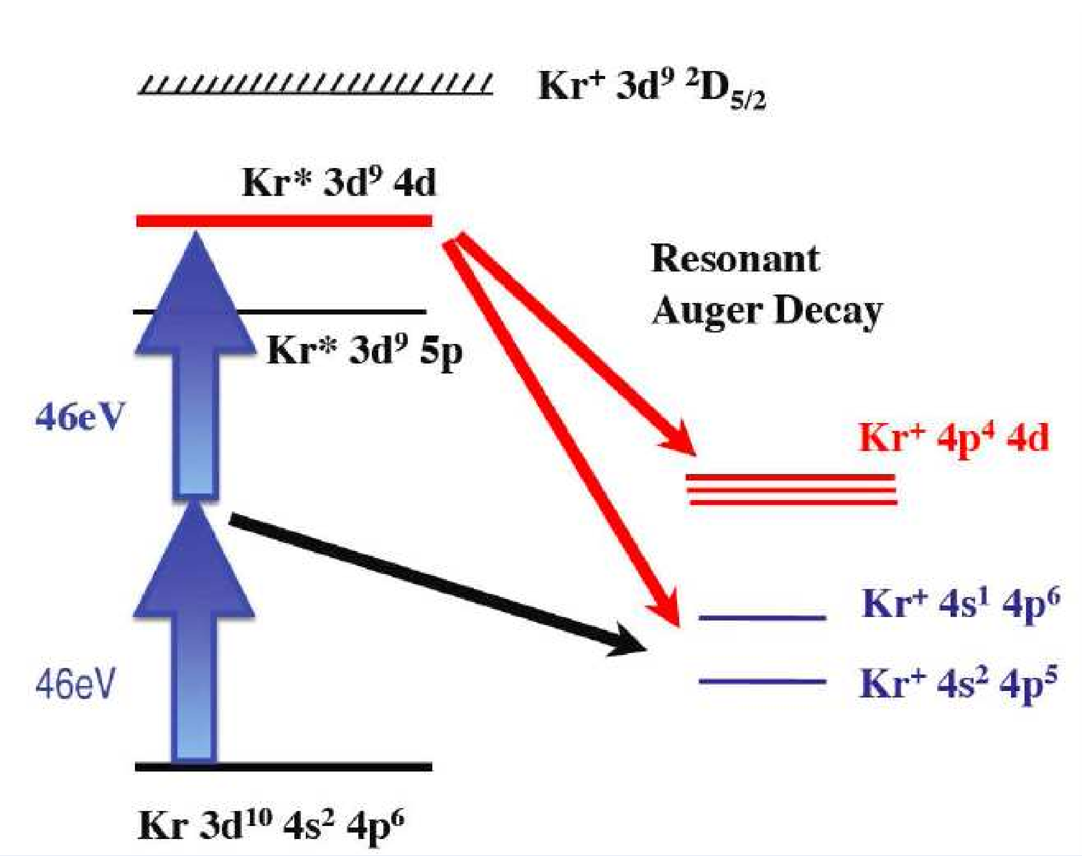
2. Next phase - FEL more easily tunable, so we can now explore **resonant two photon** processes…….

Kr - *Resonant* Two Photon Excitation

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

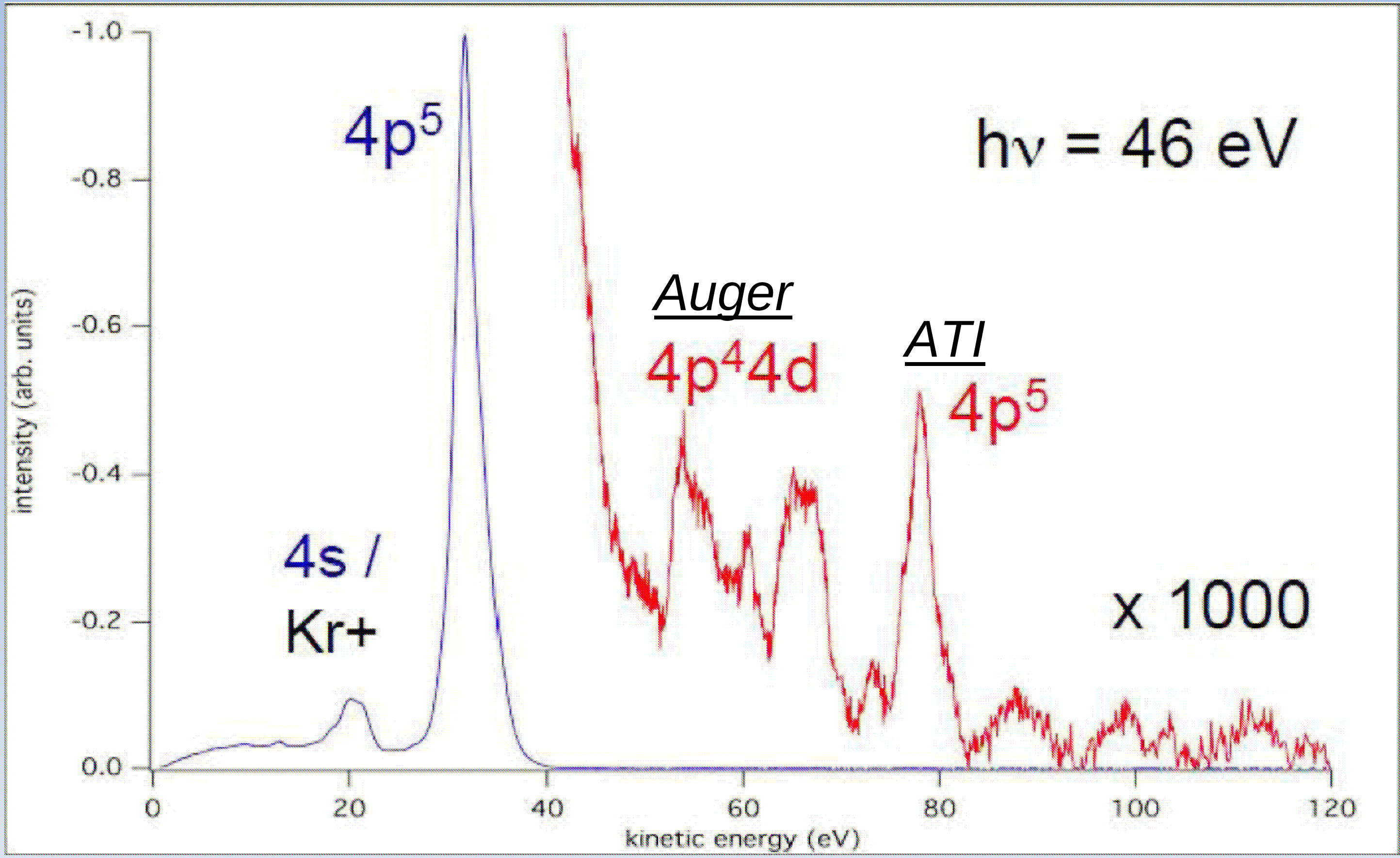
2. Of course there is a direct ionization path and the usual interference results - manifested in resonance profiles (Fano/Fano-Mies)

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 (as well as ATI makes for a complex, but intriguing, problem for theory)……



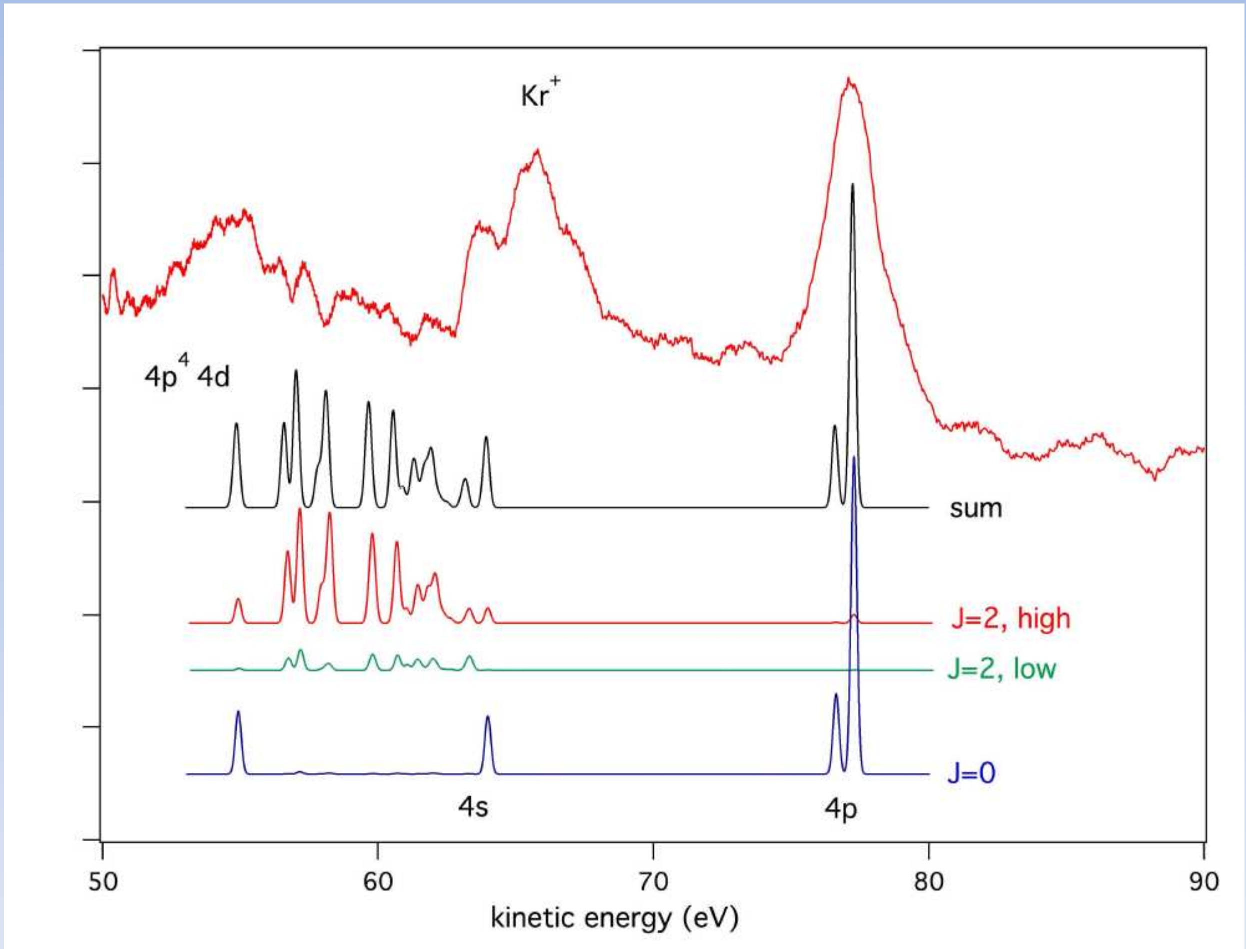
Kr (3d^{9}4d) 2 Photon Resonance Auger

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



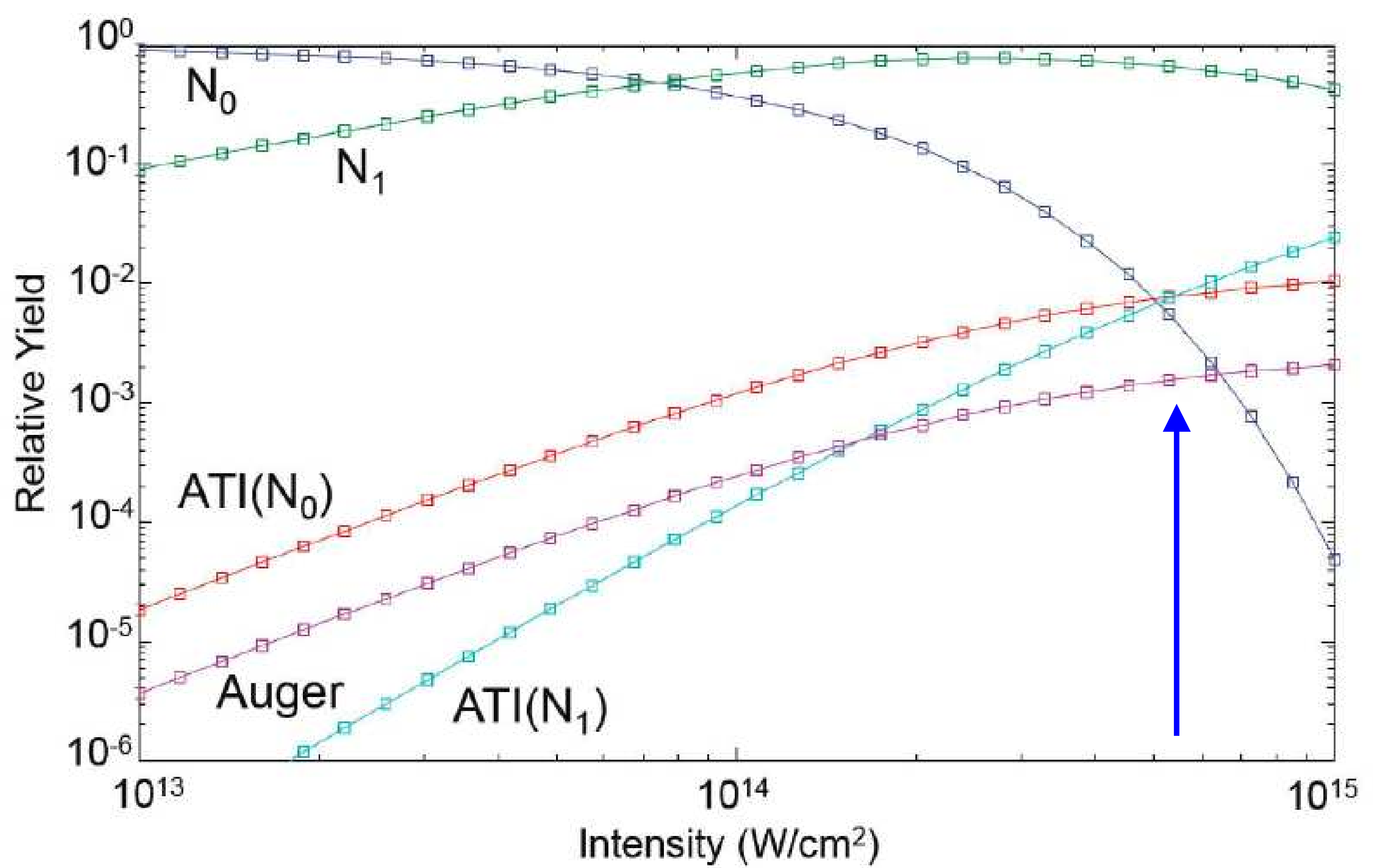
Kr ($3d^9 4d$) 2 Photon Resonance Auger

Theoretical Spectra – Stefan Fritzsche (GSI & Oulu)



Kr ($3d^9 4d$) 2 Photon Resonance - Ion Yield

Ionization rates – P. Lambropoulos, Crete



Kr ($3d^9 4d$) 2 Photon Resonance

Just out…………..

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

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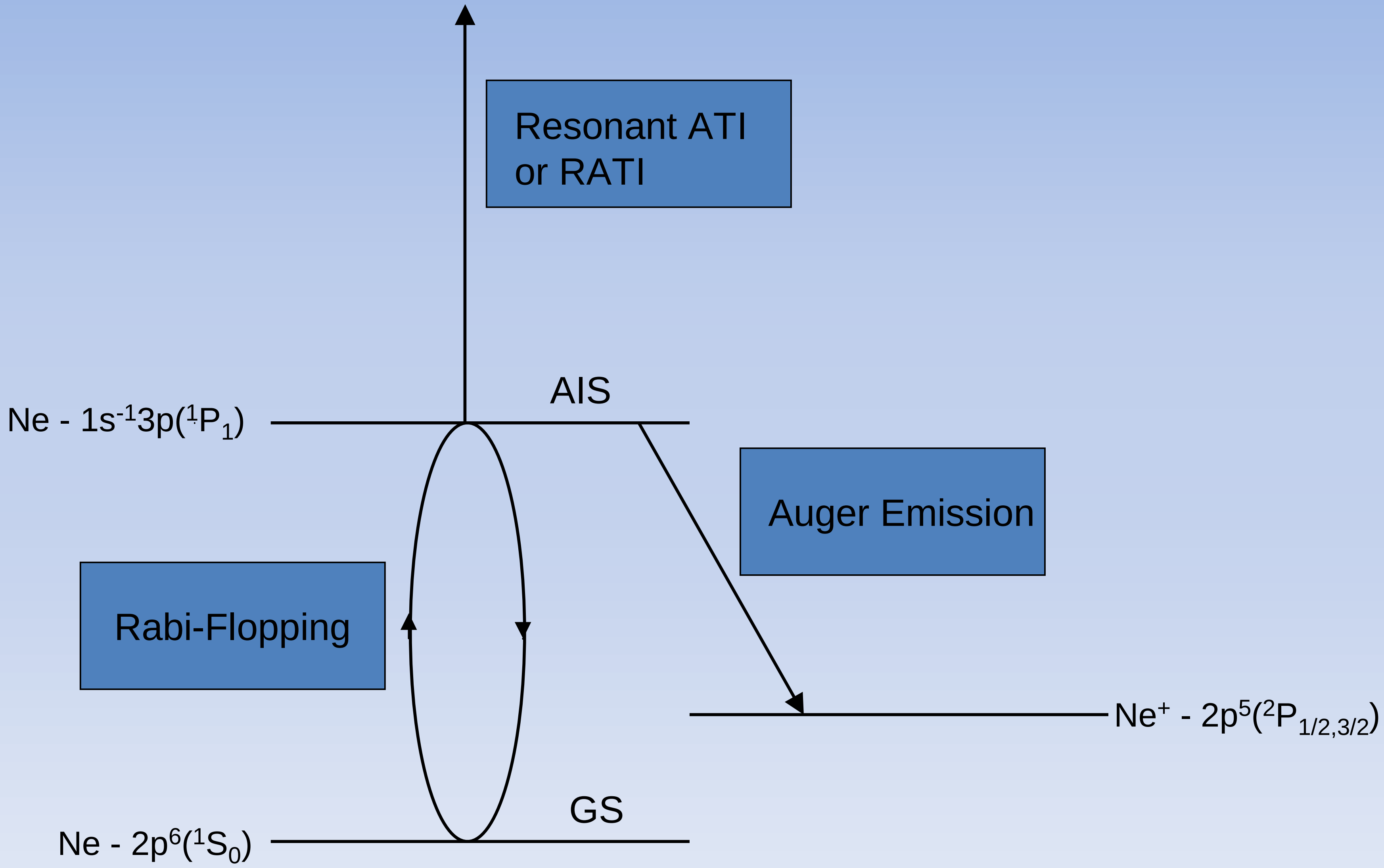
Two-photon excitation of a single-photon forbidden Auger resonance has been observed and investigated using the intense extreme ultraviolet radiation from the free electron laser in Hamburg. At the wavelength 26.9 nm (46 eV) two photons promoted a 3*d* core electron to the outer 4*d* shell. The subsequent 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

6. Next steps: X-ray coherent control ?



Next steps: X-ray coherent control ?

PHYSICAL REVIEW A **77**, 053404 (2008)

Resonant Auger effect at high x-ray intensity

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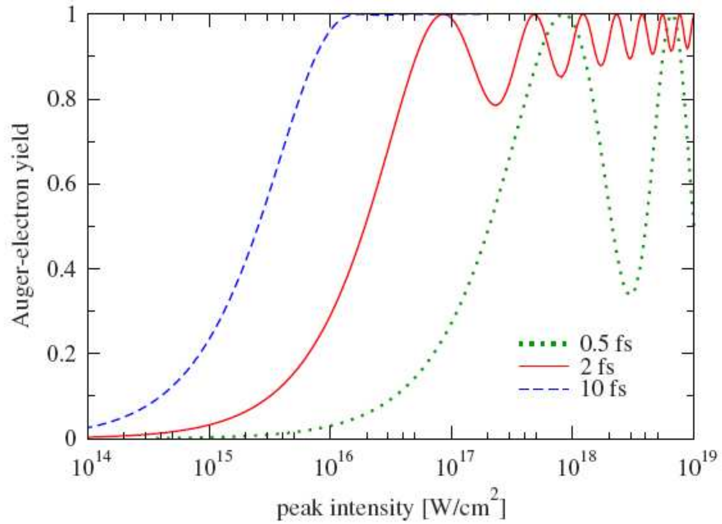

FIG. 2. (Color online) Total resonant Auger electron yield after exposure to a Gaussian-shaped pulse of duration σ=0.5 fs (green dotted line), σ=2 fs (red solid line), and σ=10 fs (blue dashed line) as a function of the x-ray peak intensity.

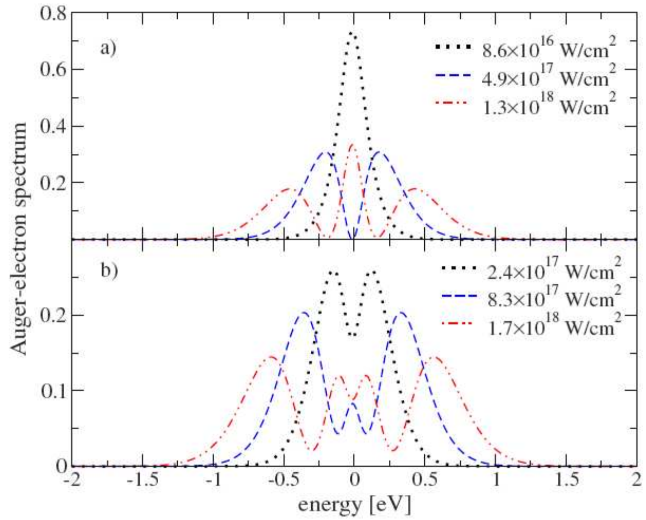

FIG. 3. (Color online) Resonant Auger electron line profile for a Gaussian-shaped pulse of duration σ=2 fs for different peak intensities. The peak intensities chosen correspond, respectively, to the first three maxima (a) and the first three minima (b) of the total resonant Auger electron yield depicted in Fig. 2.

Summary

- First detection of a so-called ‘above threshold ionization’ (ATI) two-photon process in an *inner electron* shell. Strong-field Multiple Ionization in the Inner 4d Shell by EUV Radiation

- 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’*

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

- Next step - Optical pumping / coherent control at X-rays @ XFEL/LCLS

Refs:
Xe - Richardson et al. under review at PRL, Kr - Meyer et al., PRL (May 28 - 2010)