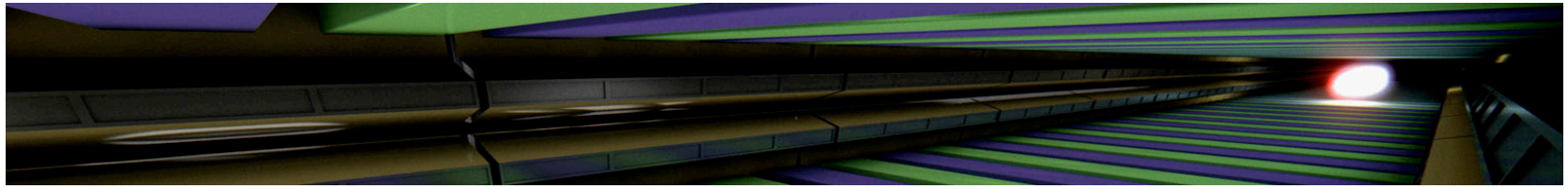


Nonlinear effects in atomic one- and two-color photoionization

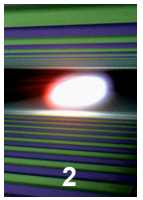
Michael Meyer

European XFEL, Hamburg, Germany

FLASH Users' Meeting
Hamburg, January 27, 2011

European XFEL

Outline



- **Introduction**
  - Nonlinear processes
- **One-color (XUV) experiments**
  - Two-photon resonant excitation in Kr
  - Two-photon Above Threshold Ionization in Xe
- **Two-color (XUV+NIR) experiments**
  - Photoionization in strong NIR dressing fields
  - Laser-assisted sequential two-photon double ionization
- **Summary**

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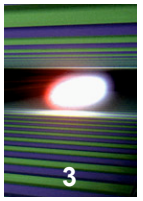
Multi-photon processes



High intensity

FLASH:

10 μJ, 10 fs, 10 μm

~ 10^{15} W/cm^2

Multi-photon processes

Nonlinear: Signal ~ I^N

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

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

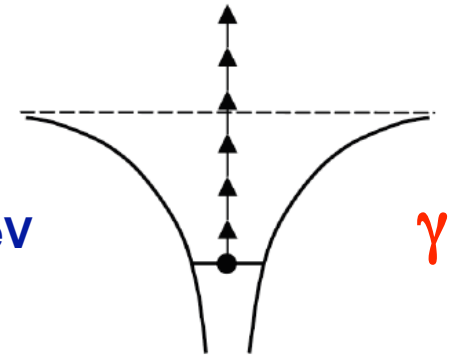
Optical Laser (800nm)

Multi-photon Ionization

8 nm

U_p = 0.006 eV

γ = 45



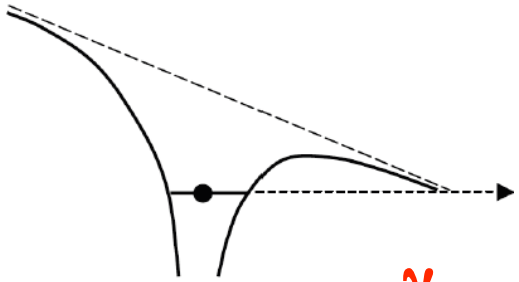
γ >> 1

Tunnel Ionization

800 nm

U_p = 60 eV

γ = 0.45



γ << 1

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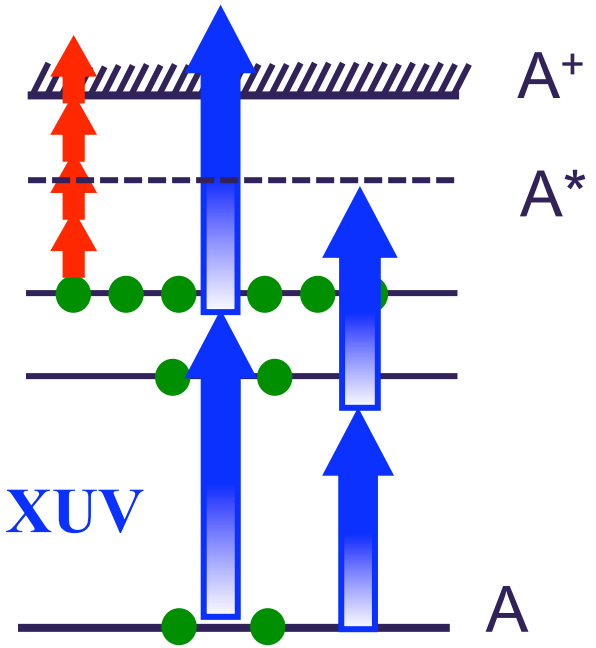
Photoionization in intense fields

One-color processes



XUV photons open access to

- inner-shell electrons (Auger decay)
- two-photon core resonances
- test theoretical models for MPI
  in the short wavelength regime

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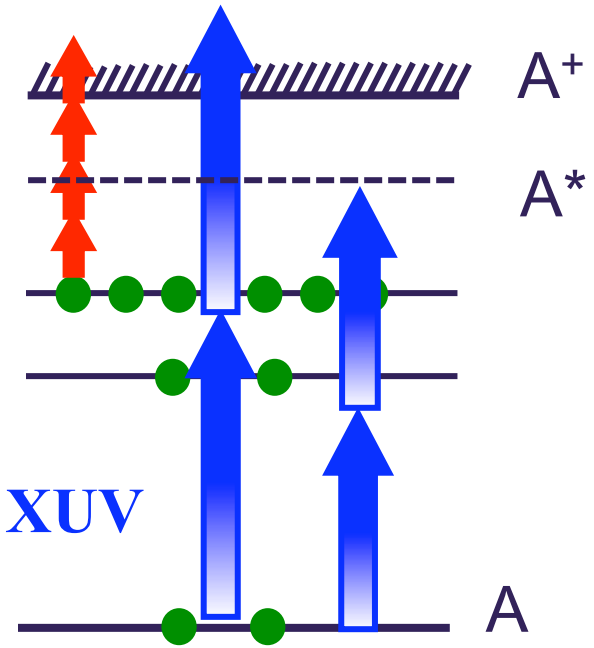
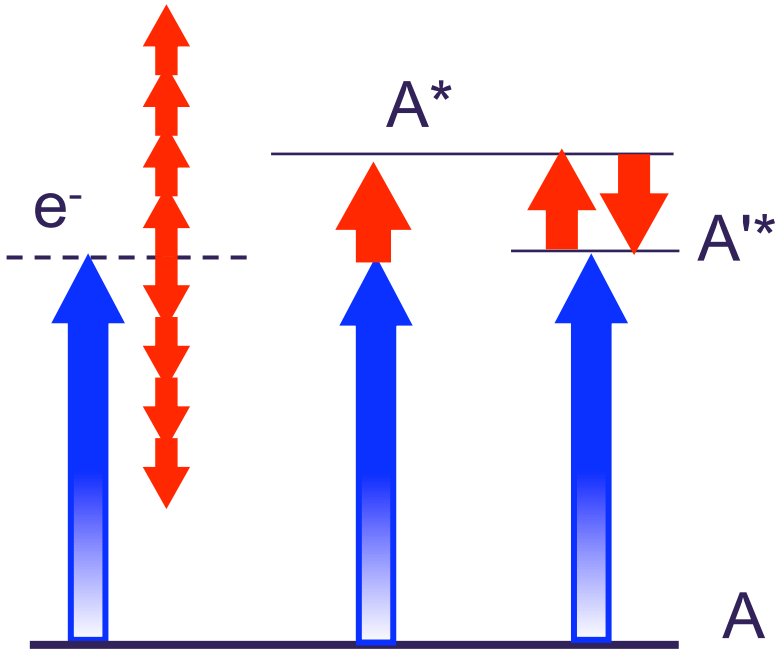
Photoionization in intense fields

One-color processes



Two-color processes



XUV photons open access to

- inner-shell electrons (Auger decay)
- two-photon core resonances
- test theoretical models for MPI in the short wavelength regime

Two-color excitations open access to

- photoionization of dressed atoms
- two-color ATI (Above Threshold Ionization)
- two-photon core-resonances
- coupling of core-resonances

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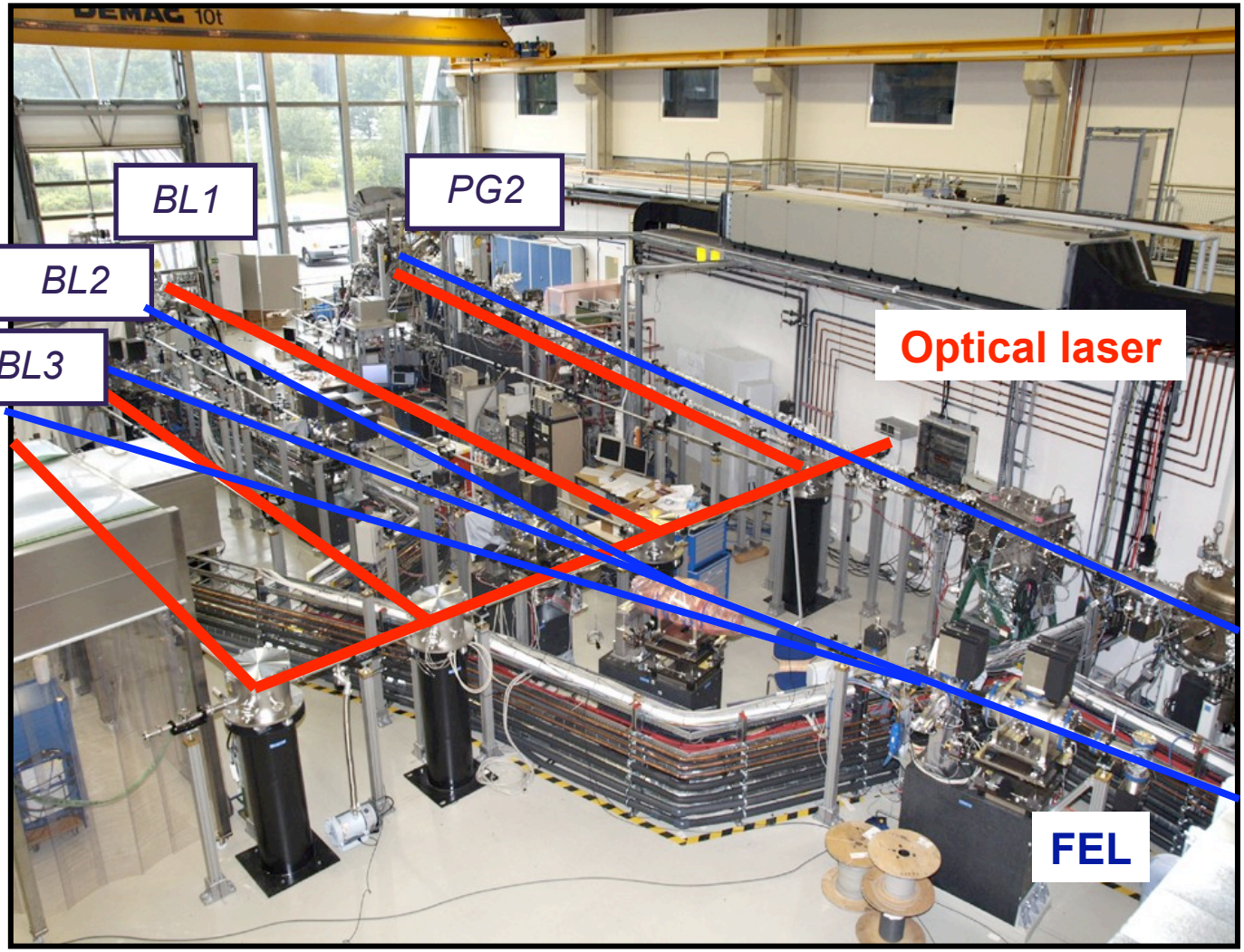
FLASH (Free electron LASer in Hamburg)



BL2

$h\nu$ = 46 eV
93 eV

λ = 26.9 nm
13.3 nm

rep. rate: 10 Hz

ΔT = 10 – 30 fs

E = 10 – 30 μJ

d_F = 5 – 30 μm

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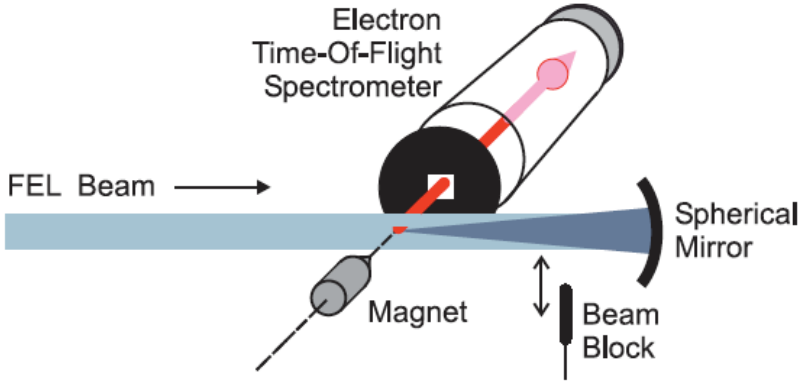
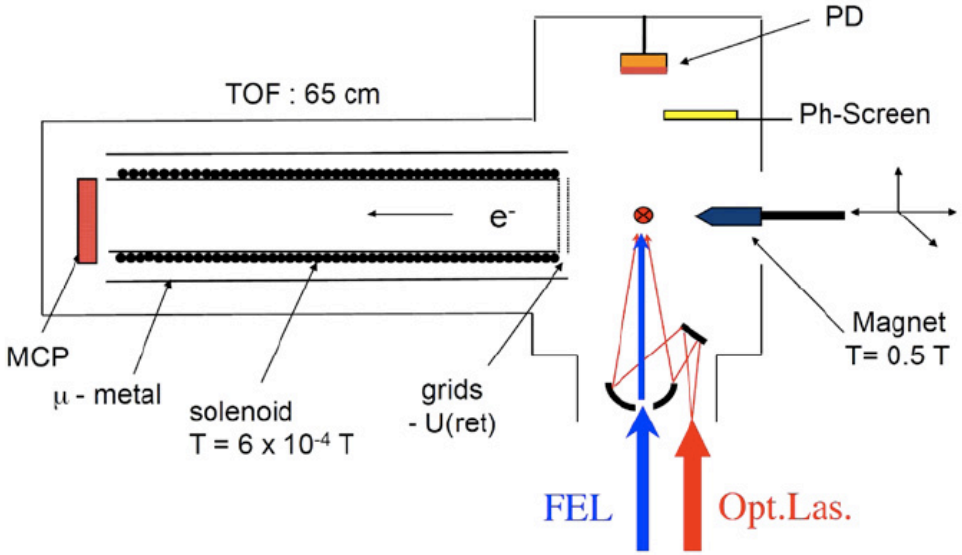
Photoelectron Spectroscopy

One-color set-up

Two-color set-up

Magnetic bottle electron spectrometer



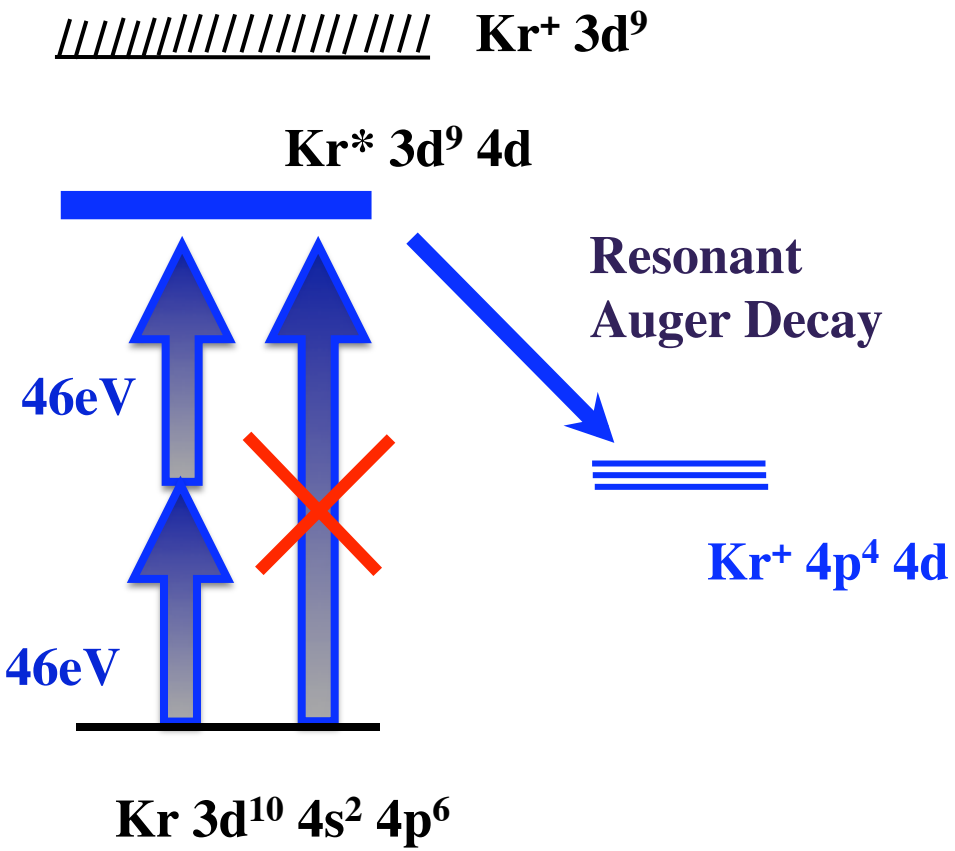


- refocusing ML mirror for XUV
- aperture at MBES entrance

- focusing mirror for optical laser
- diagnostics for spatial and temporal overlap

Two-photon resonant excitation

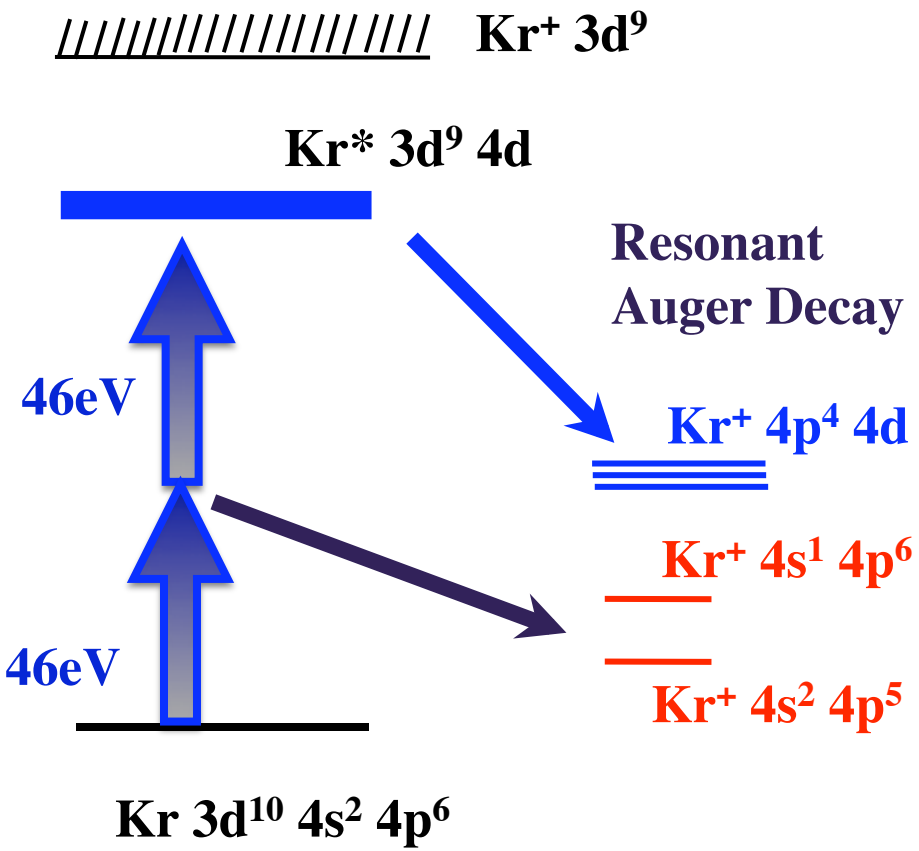
FLASH: 26.9 nm, 15 μJ, 10-30 fs, 5 μm
→ 10^{15} W/cm²



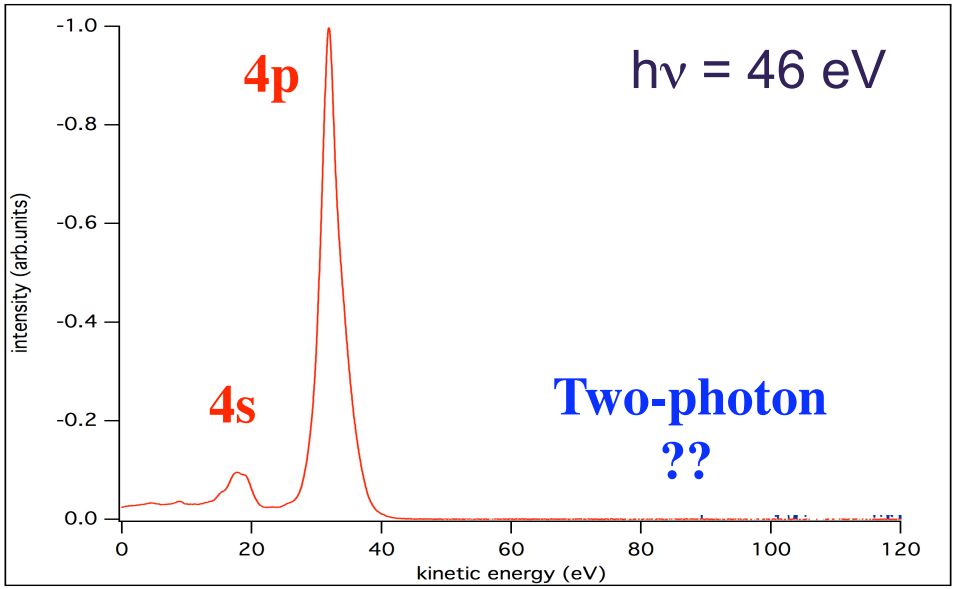
European XFEL

Two-photon resonant excitation

FLASH: 26.9 nm, 15 μJ, 10-30 fs, 5 μm
→ 10^{15} W/cm²



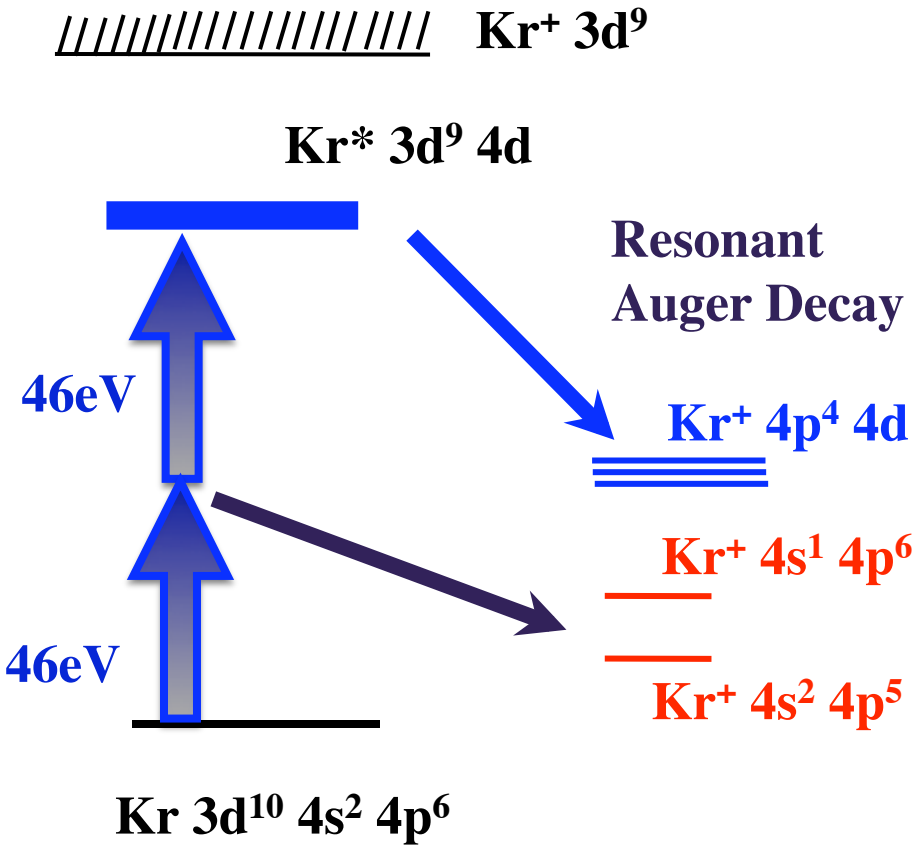
One-photon ionization



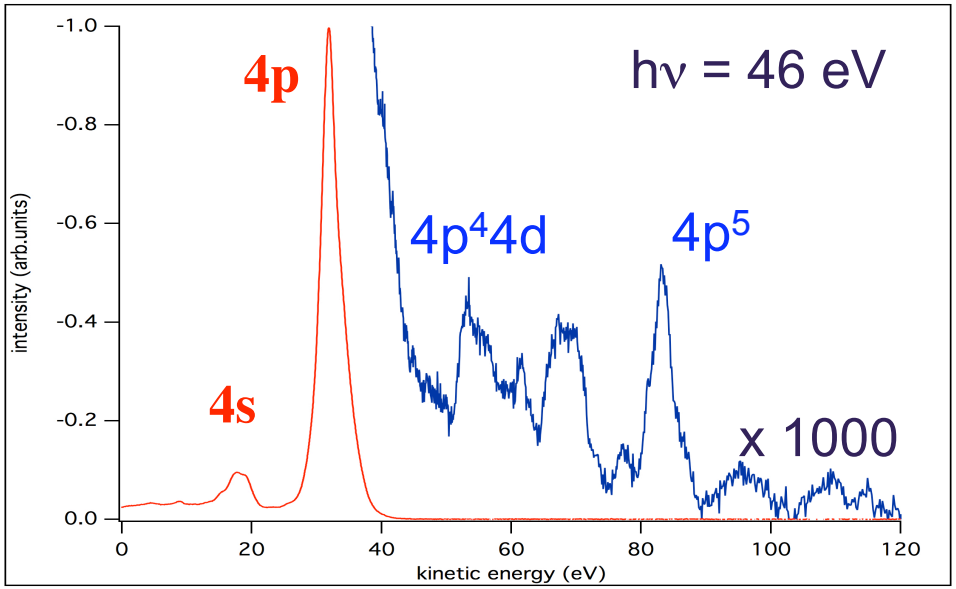
European XFEL

Two-photon resonant excitation

FLASH: 26.9 nm, 15 μJ, 10-30 fs, 5 μm
→ 10^{15} W/cm²



One-photon ionization



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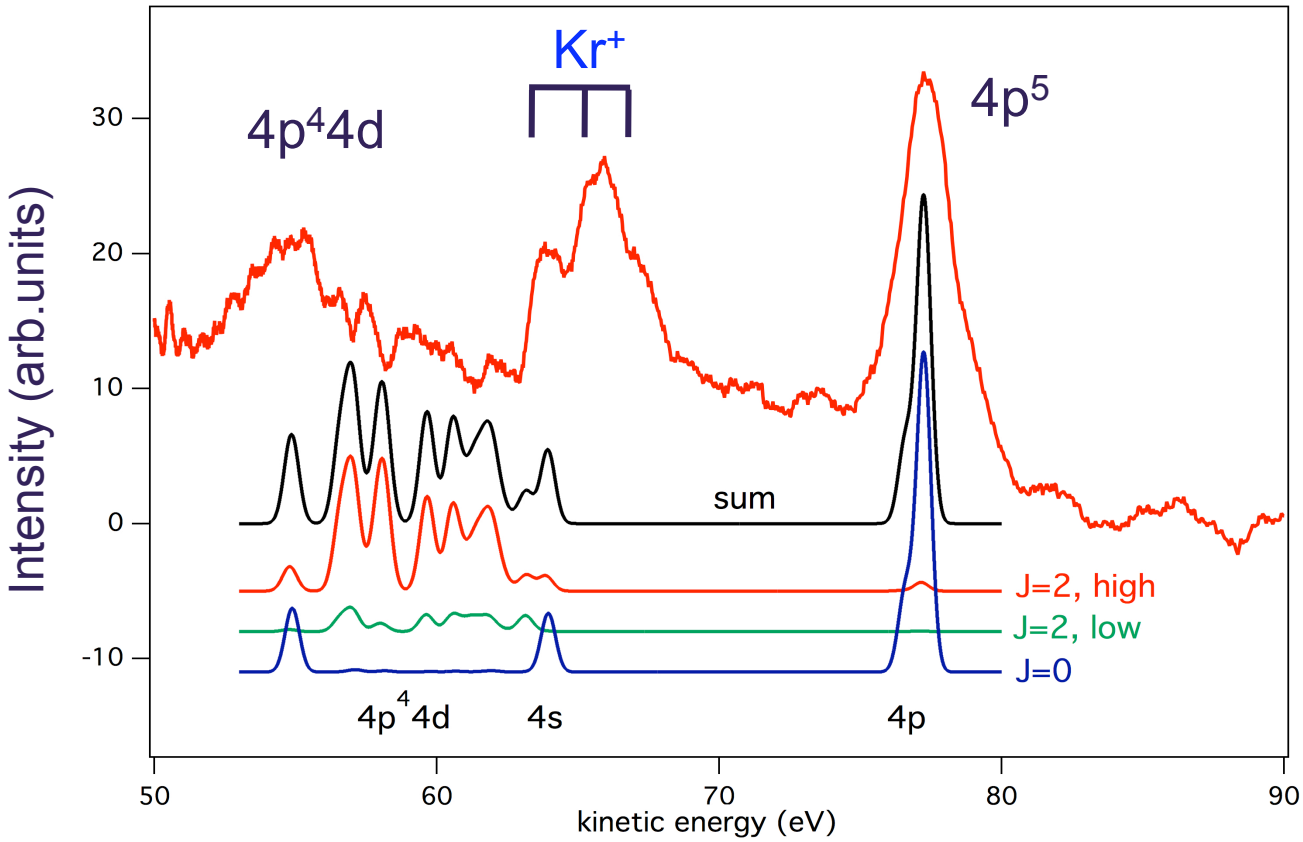
Resonant Auger decay

Theory: S. Fritzsche, P. Lambropoulos, A. Mihelic,

$Kr^{**}\ 3d^9 4s^2 4p^6 4d\ (J=0,2) \quad \rightarrow Kr^{+}\ 3d^{10} 4s^2 4p^4 4d + e^-$



Two-photon processes:

- Resonant excitation

- 4p ATI from neutral Kr

- 4p ATI from ionic Kr

M. Meyer, D. Cubaynes, V. Richardson, J. T. Costello, P. Radcliffe, W. B. Li, S. Düsterer, S. Fritzsche, A. Mihelic, K. G. Papamihail, and P. Lambropoulos
PRL 104, 213001 (2010)

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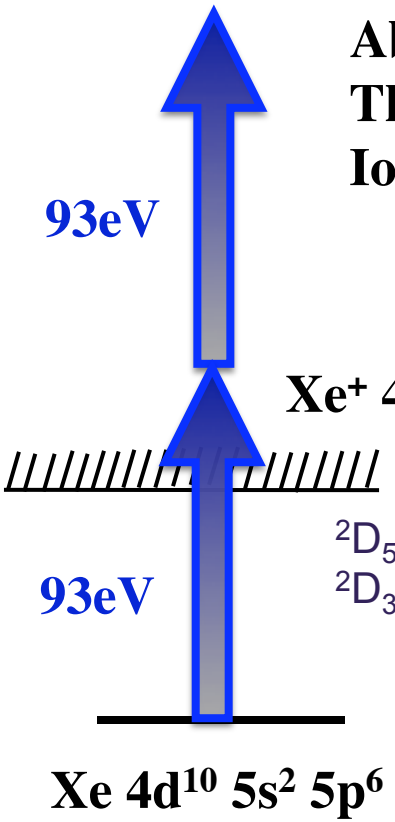
Two-photon excitation of Xe 4d

hν (FEL) 93 eV ; ~ 10^{15} W / cm^2



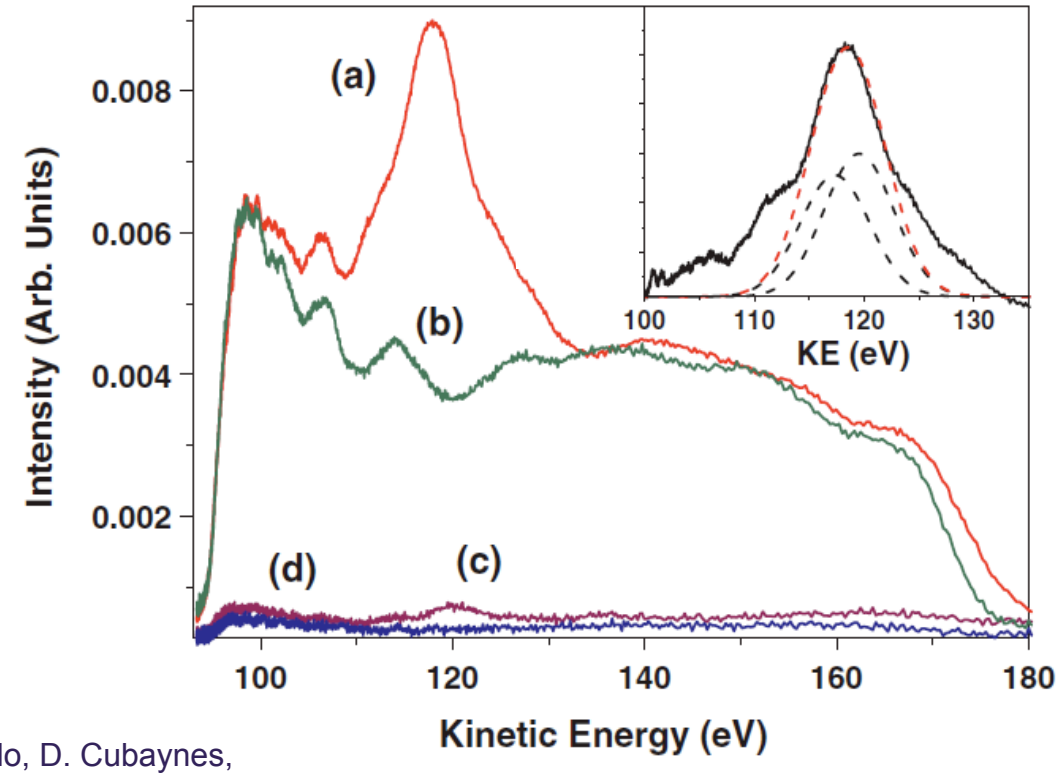
Above Threshold Ionization

$Xe^+ 4d^9$

$^2D_{5/2}$: 67.5 eV
$^2D_{3/2}$: 69.5 eV

$Xe\ 4d^{10}\ 5s^2\ 5p^6$

U_{Ret} = - 90 eV



V. Richardson, J. T. Costello, D. Cubaynes, S. Düsterer, J. Feldhaus, H. W. van der Hart, P. Juranić, W. B. Li, M. Meyer, M. Richter, A. A. Sorokin, K. Tiedke
PRL 105, 013001 (2010)

Two-photon ATI of Xe 4d
2 x hν – 118 eV = 68 eV

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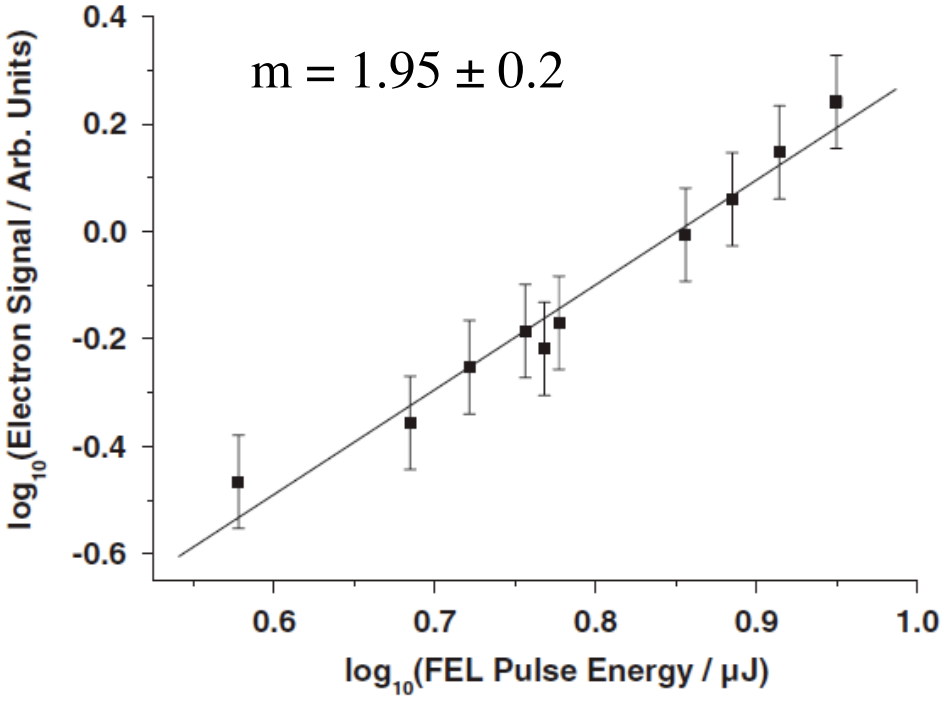
Two-photon excitation of Xe 4d

Two-photon process

Experimental evidence

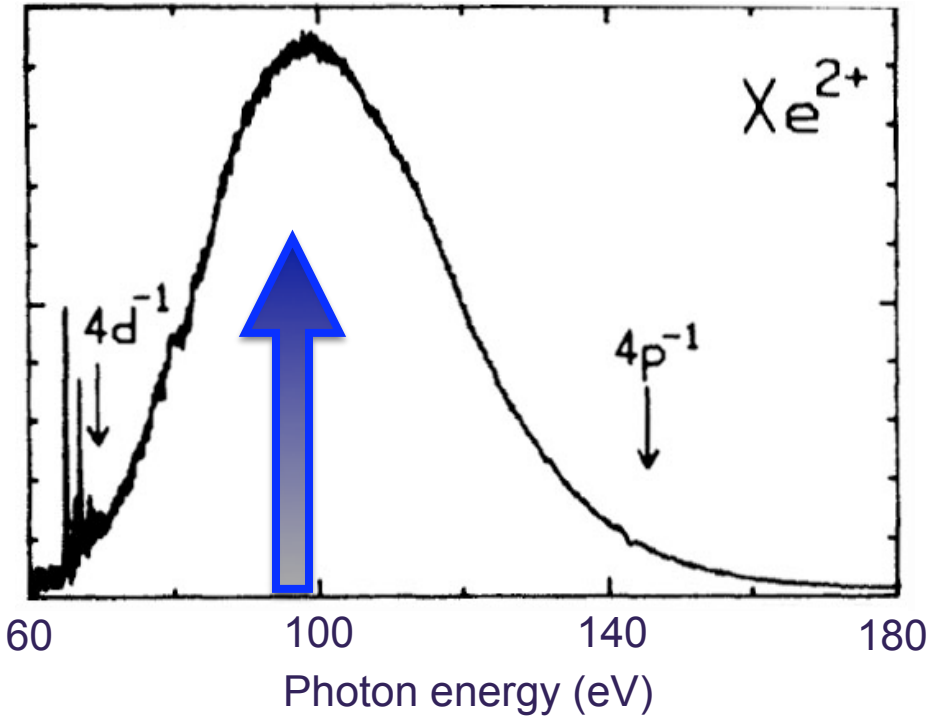
Signal ~ I^N (N=2)



Resonant enhanced two-photon process

Theory : H. van der Hardt

Xe: 4d → εf (giant resonance) → ε'g



10^{16} W/cm² : 4d (two-photon) = 0.5% 4d (total)

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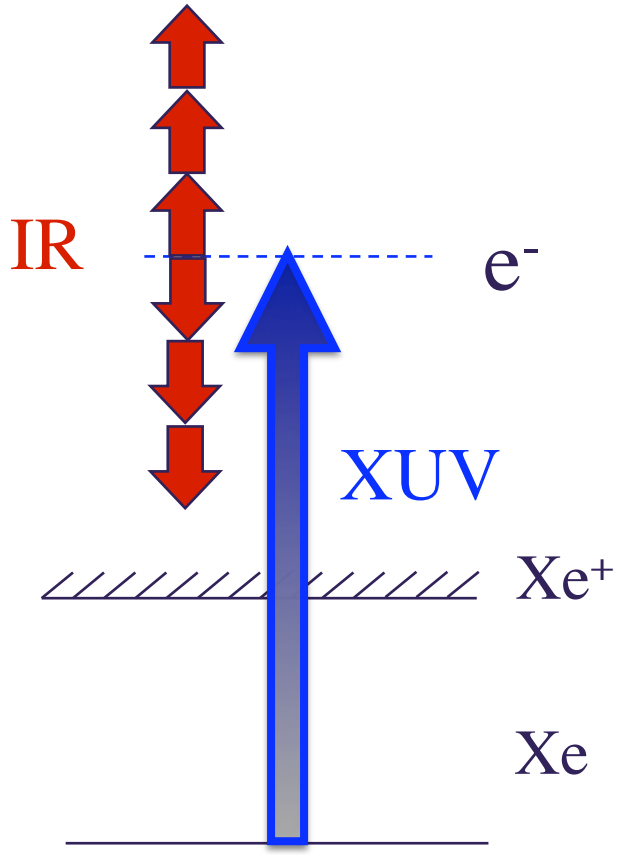
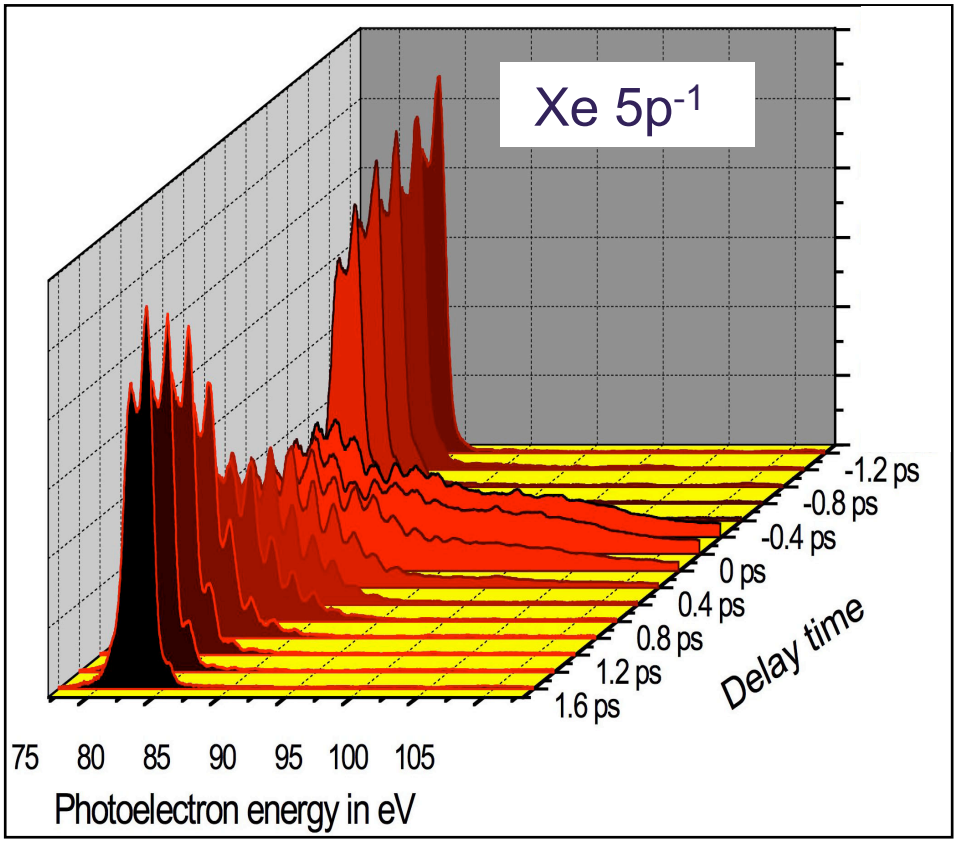
Two-color multi-photon processes

Above Threshold Ionization



Toma et al. PRA 62, 0618015 (2000)

Optical laser: > 10^{14} W/cm^2



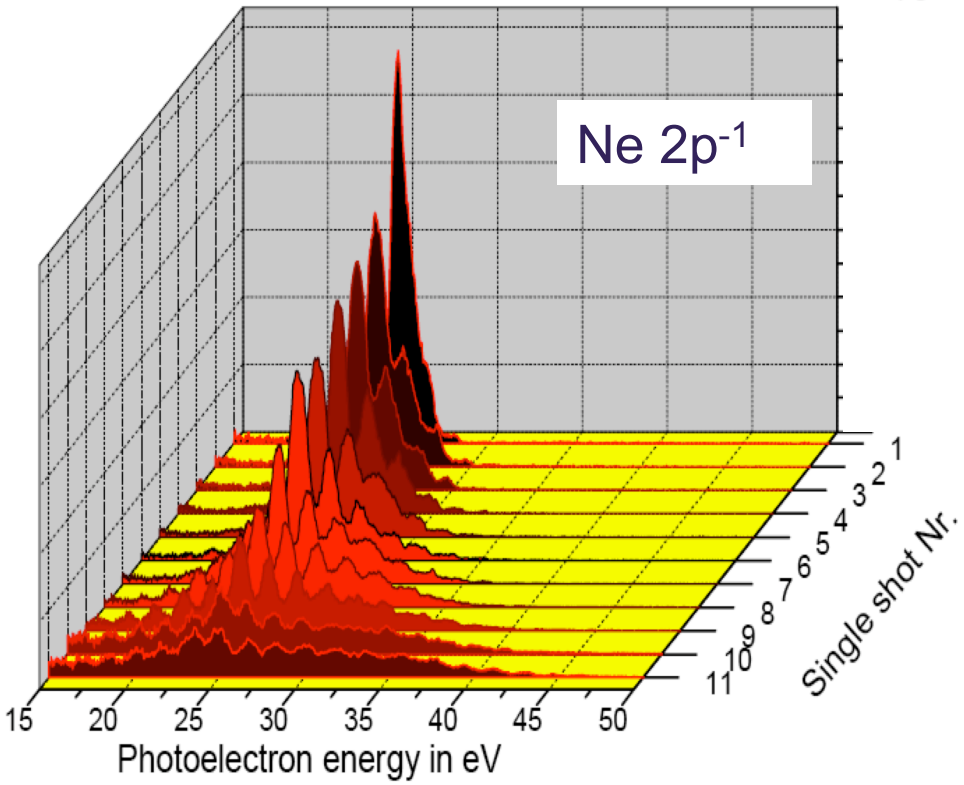
Multi-photon processes

European XFEL

ATI : Strong NIR Dressing Field (Ne)

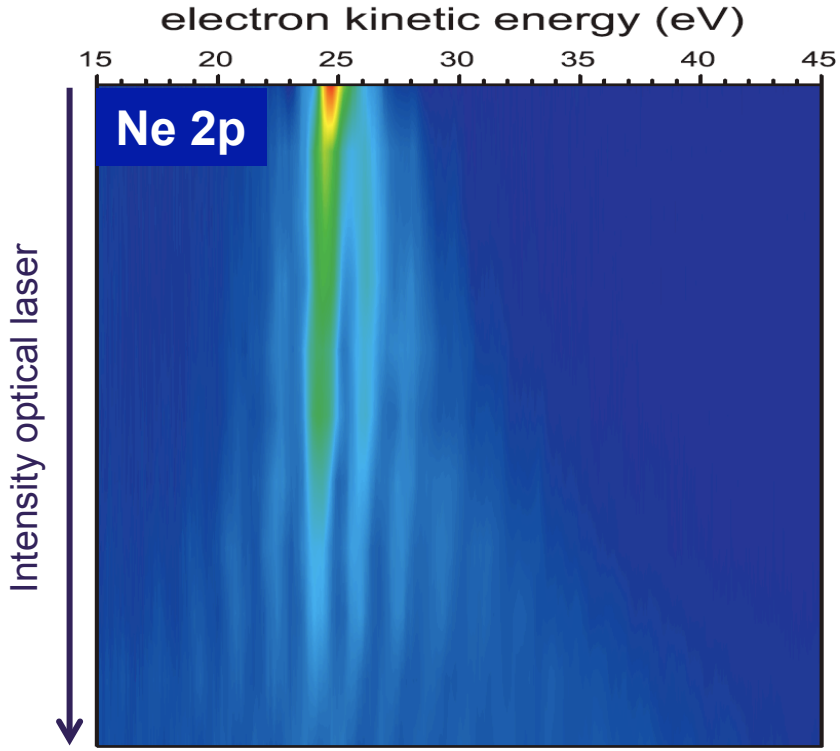
FLASH: 26.9nm (46 eV)

Opt.Las.: 800nm, 1.8mJ, 100fs



Single shot spectra for overlap

Optical laser: 2 x 10^{13} W/cm^2



Sorted single shot spectra

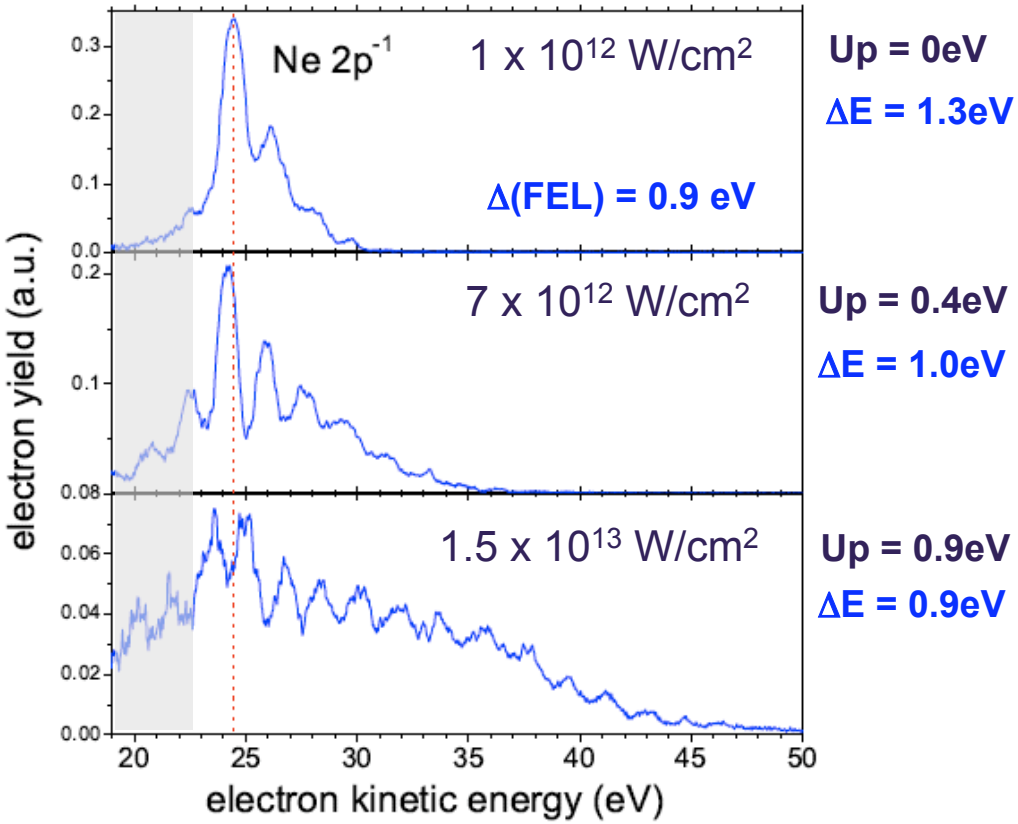
"Jitter-free" → Defined field strength

European XFEL

ATI : Strong NIR Dressing Field (Ne)

FLASH: 26.9nm (46 eV)

Opt.Las.: 800nm, 1.8mJ, 120fs



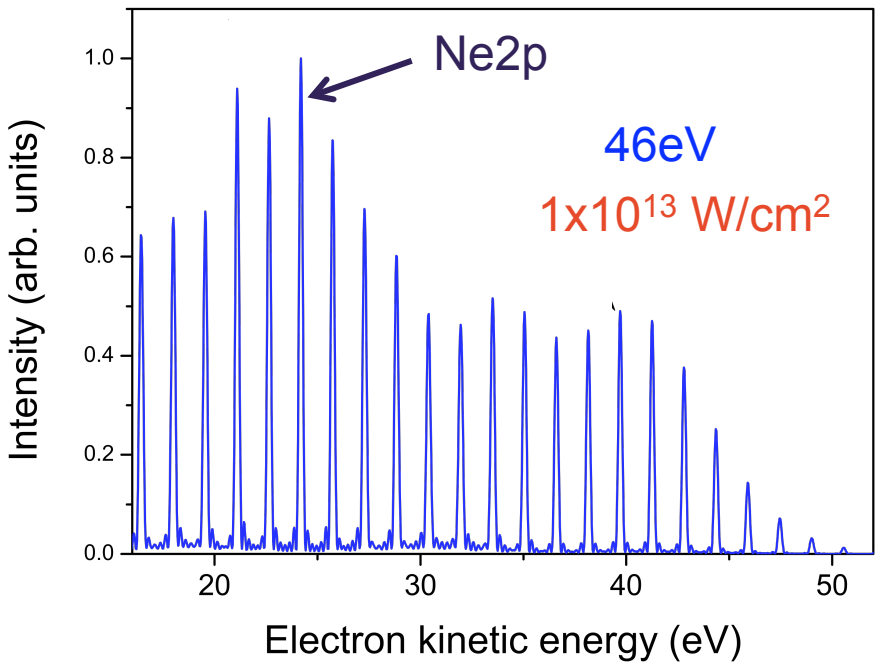
Up = 0eV
ΔE = 1.3eV

Up = 0.4eV
ΔE = 1.0eV

Up = 0.9eV
ΔE = 0.9eV

TDSE: Time-dependent Schrödinger Equation
Maquet, Taieb, J. Mod. Opt. 54, 1847 (2007)

Simulated theoretical spectrum



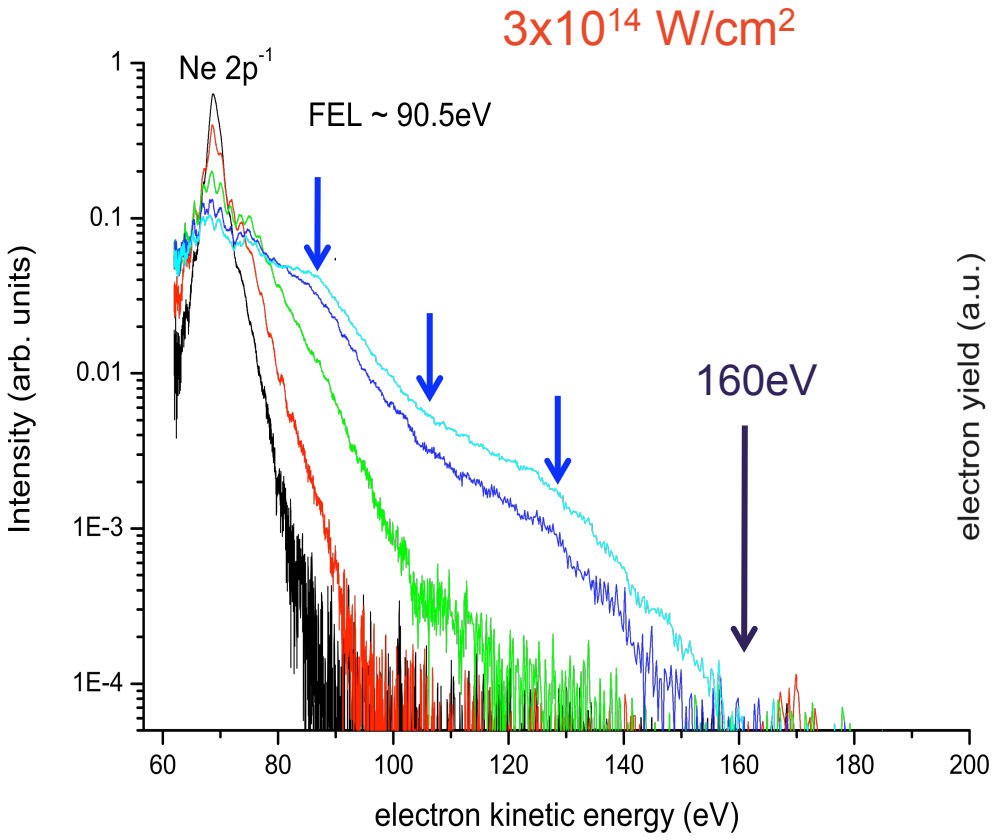
Experiment: **bandwidth** broadening and **jitter (intensity)** broadening

European XFEL

Photoionization in strong NIR fields (Ne)

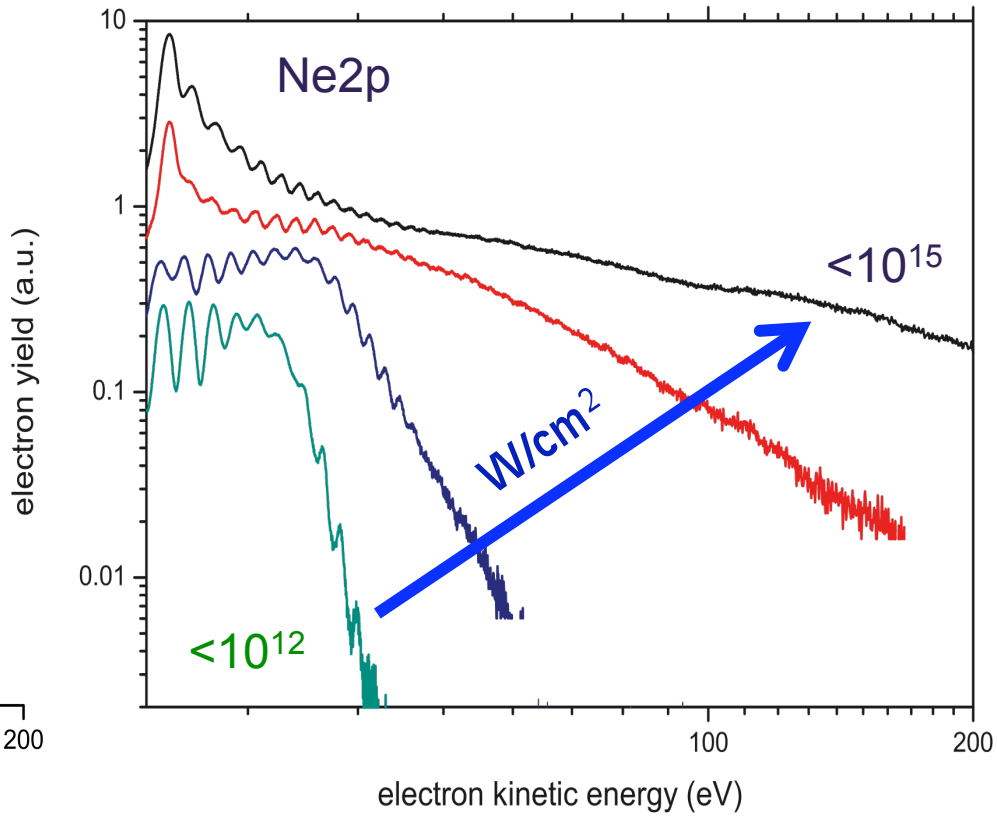
FLASH: 13.5 nm (90.5 eV)

Opt.Las.: 800nm, <1.8mJ, 120fs



FLASH: 26.9 nm (46 eV)

Opt.Las.: 800nm, < 1.8mJ, 120fs



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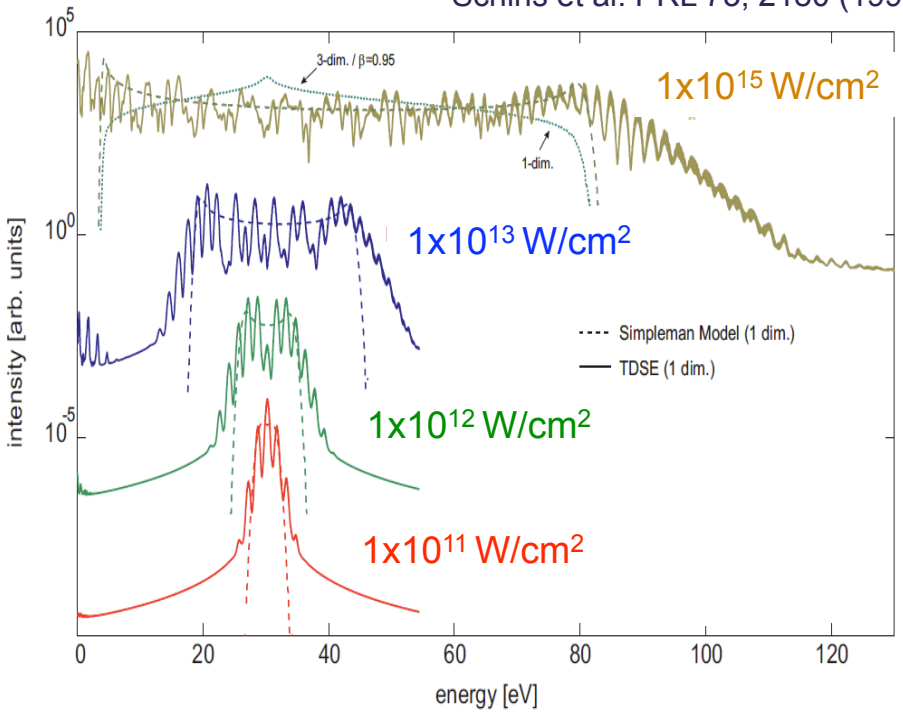
Photoionization in strong NIR fields (Ne)

Photoionization in a combined XUV – IR field

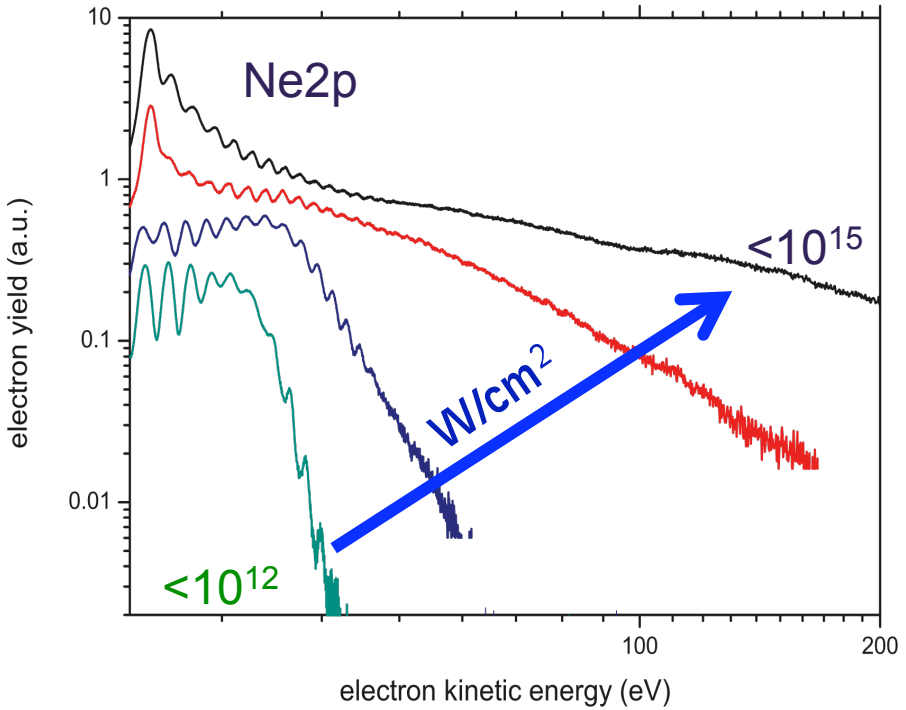
TDSE and "Simpleman Model"

Schins et al. PRL 73, 2180 (1994)



FLASH: 26.9 nm (46 eV)

Opt.Las.: 800nm, 1.8mJ, 100fs



T. Fennel / M. Arbeiter
Uni Rostock, Germany

Differences with one-color MPI
Interferences between MPI processes

European XFEL

Summary

One-color (XUV) two-photon processes

Resonant Auger decay of two-photon resonance (Kr)

- Excitation of core-electrons
- ATI of ions formed within the same pulse

Above Threshold Ionization of core-electrons (Xe)

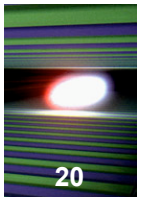
- Resonantly enhanced two-photon process

Two-color (XUV) two-photon processes

Photoionization in intense NIR dressing fields

- Above Threshold Ionization
- High kinetic energy electrons

Collaborations



- **European XFEL**
  P. Radcliffe, M. Meyer
- **DESY**
  H. Redlin, **S. Düsterer**, P. Juranic, K. Tiedtke,
  A.A. Sorokin, J. Feldhaus
- **Dublin City University**
  V. Richardson, P. Hayden, M. Kelly, E. Kennedy,
  J. Costello
- **Tongji University**
  W.B. Li
- **ISMO, Orsay**
  D. Cubaynes
- **PTB Berlin**
  M. Richter
- **IESL-FORTH**
  K.G. Papamihail, P. Lambropoulos
- **Jozef Stefan Institute**
  A. Mihelic
- **Moscow State University**
  A. N. Grum-Grzhimailo, N. M. Kabachnik
- **LCPMR, Paris**
  A. Maquet, R. Taïeb
- **GSI / University of Oulu**
  S. Fritzsche
- **University of Rostock**
  M. Arbeiter, T. Fennel
- **Queen's University Belfast**
  H. van der Hardt

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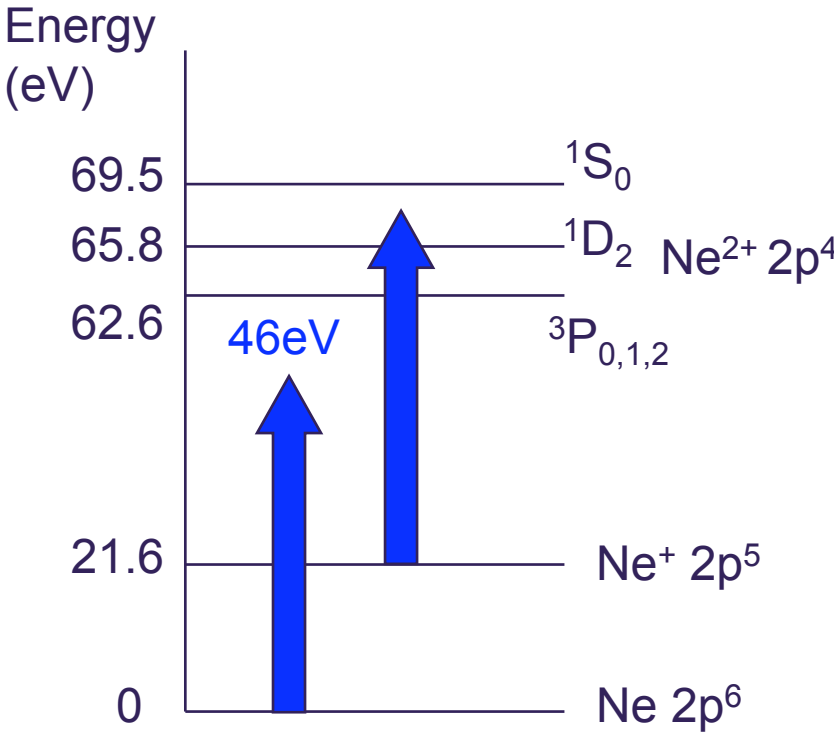
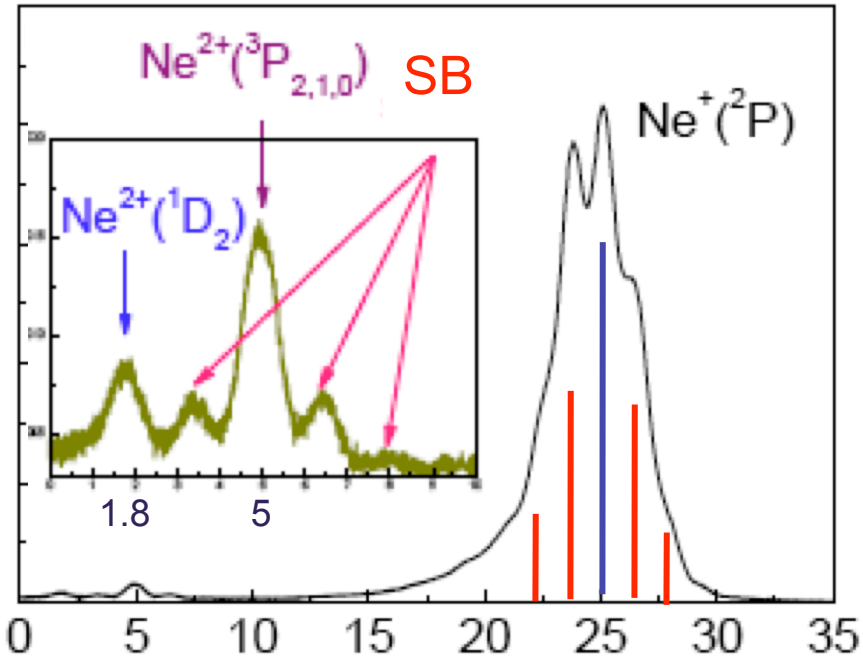
Strong XUV + NIR Fields

Sequential Two-Photon Double-Ionization



FEL: 46 eV, 20 μJ, 30fs

OL: 800nm, 1.8mJ, 3ps



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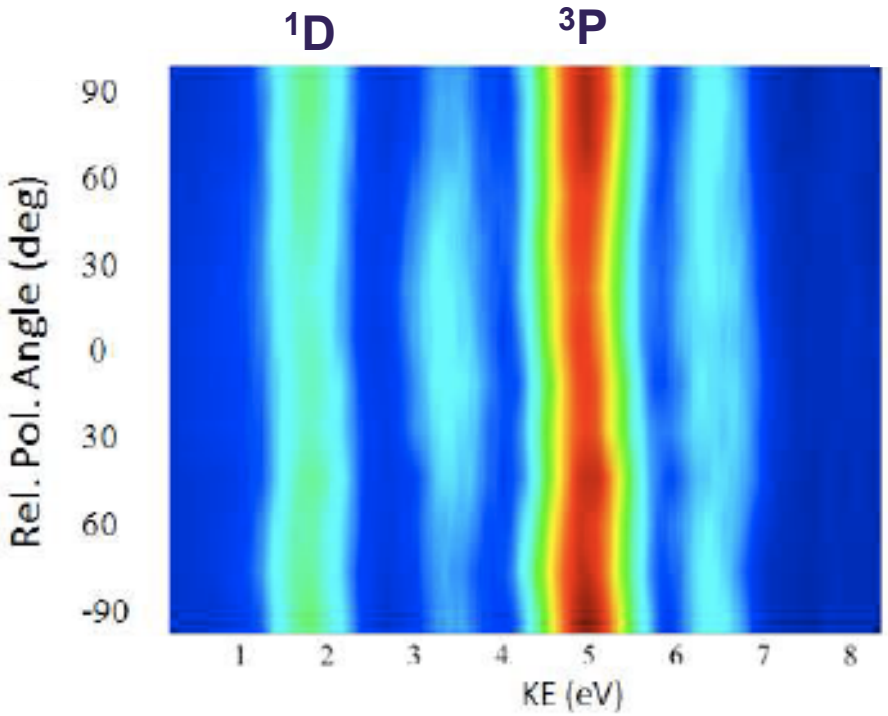
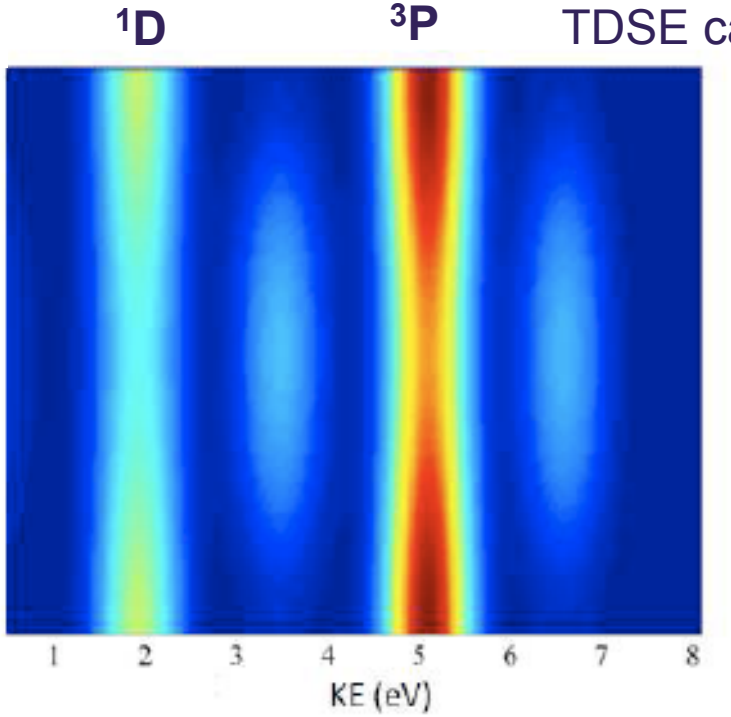
Polarization dependence of two-color ATI

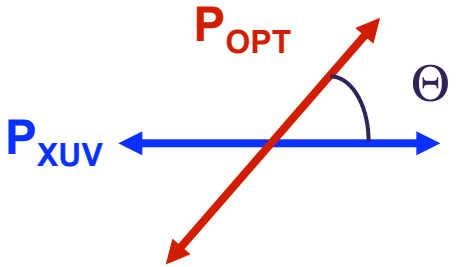
Experiment



Theory

L. Nikolopoulos:
TDSE calculation





$$\sim \quad A_{||}^{(C)}(\epsilon)\cos^2\theta_p + A_{\perp}^{(C)}(\epsilon)\sin^2\theta_p$$

$A_{||}$ and $A_{\perp}$ contain
dipol matrix elements for **p → s / d** (first step)
s → p / d → p / f (second step)

European XFEL

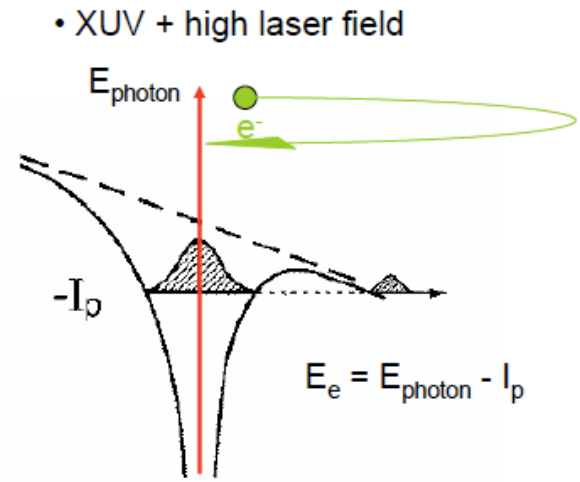
Strong NIR Dressing Field (Ne)

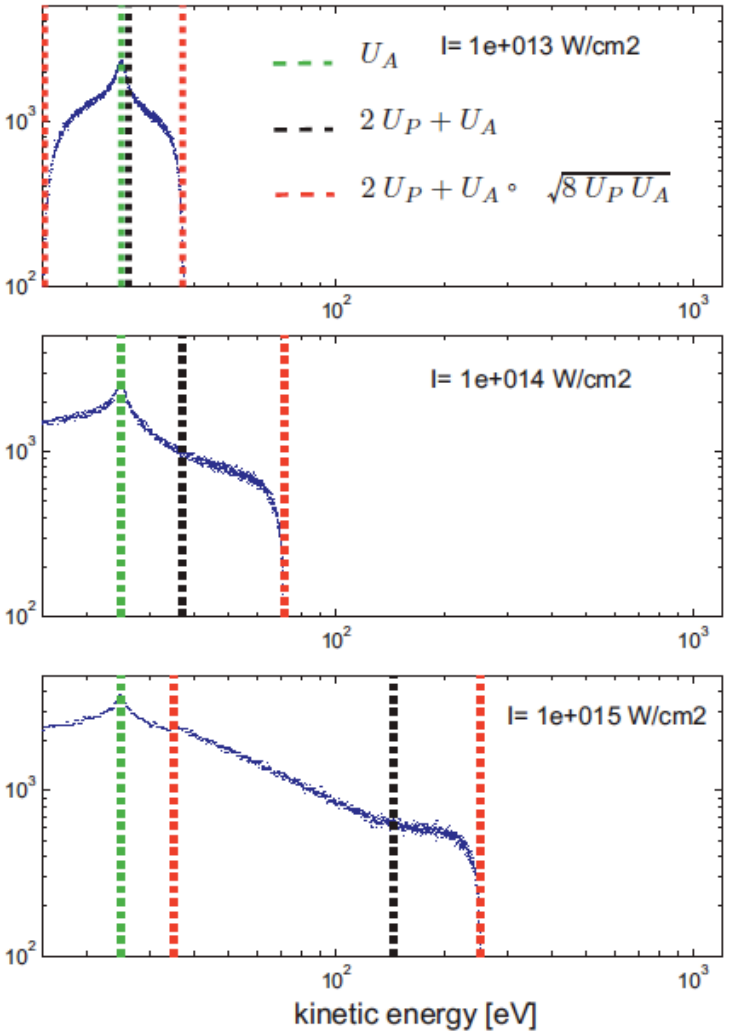
Photoionization in a combined XUV – IR field

"Simpleman's Model"

$$E_{fin} = 2\,U_P\,f^2(t_0)\sin^2(\omega t_0) + U_A + \sqrt{8\,U_P\,U_A}\,\cos(\theta_{\vec{v}_0,\vec{E}_0})\,\sin(\omega t_0)$$

U_A : excess energy U_p : ponderomotive enrgy





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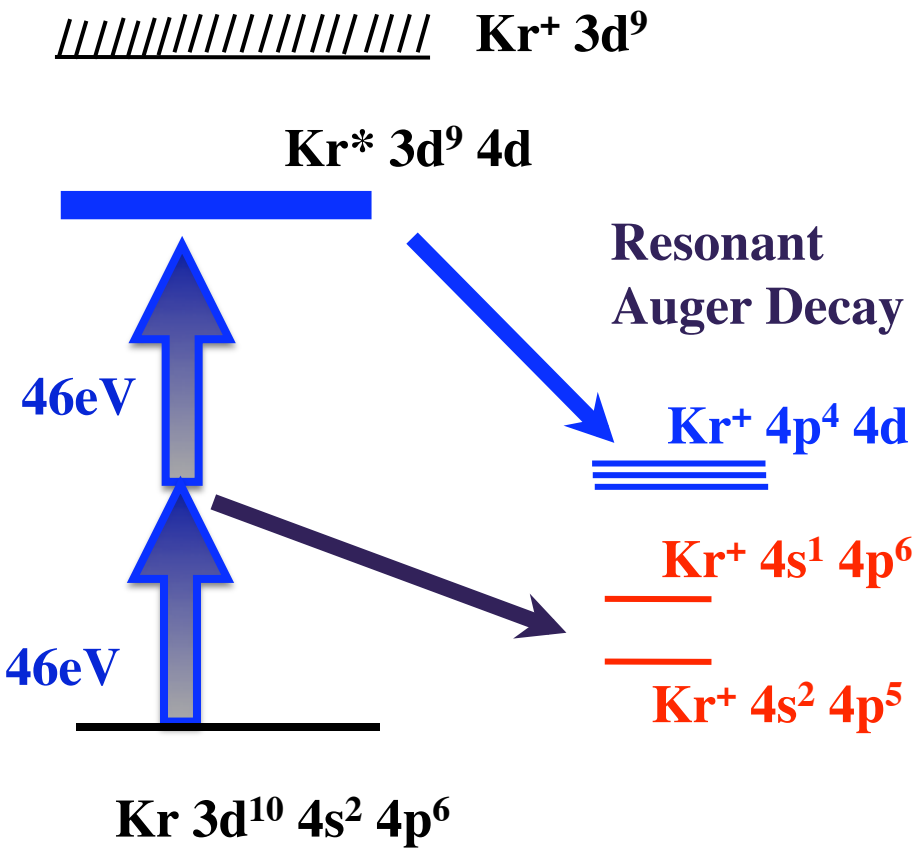
Uni Rostock, Germany

Two-photon resonant excitation

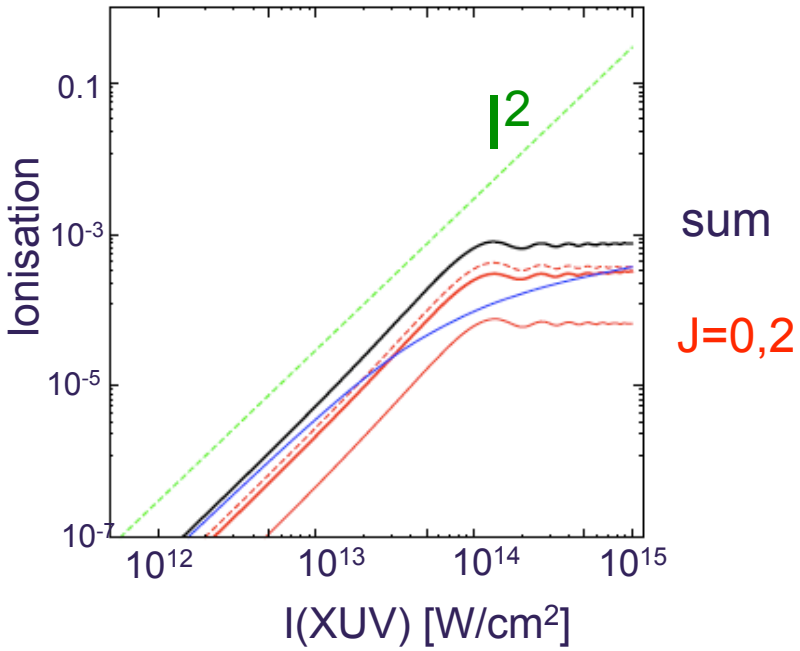
FLASH: 26.9 nm, 15 μJ, 10-30 fs, 5 μm
→ 10^{15} W/cm²



P. Lambropoulos, A. Mihelic et al.

Kr $3d^{10}4s^24p^6$ (J=0) + 2 hν(46eV)

---> Kr** $3d^94s^24p^64d$ (J=0,2)

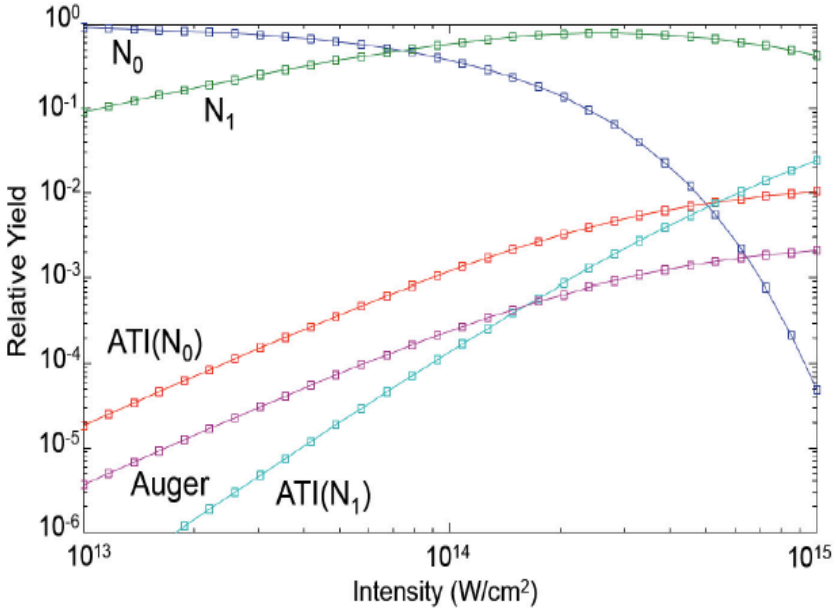


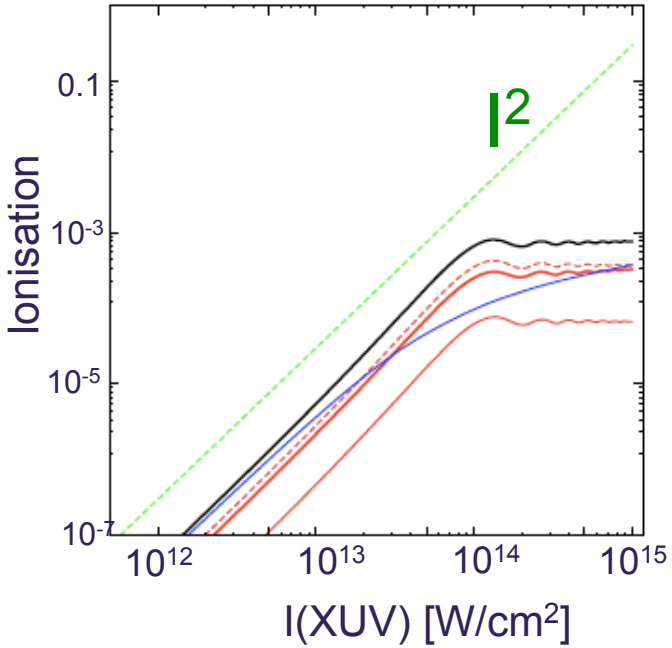
European XFEL

Two-photon resonant excitation

P. Lambropoulos, A. Mihelic et al.

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



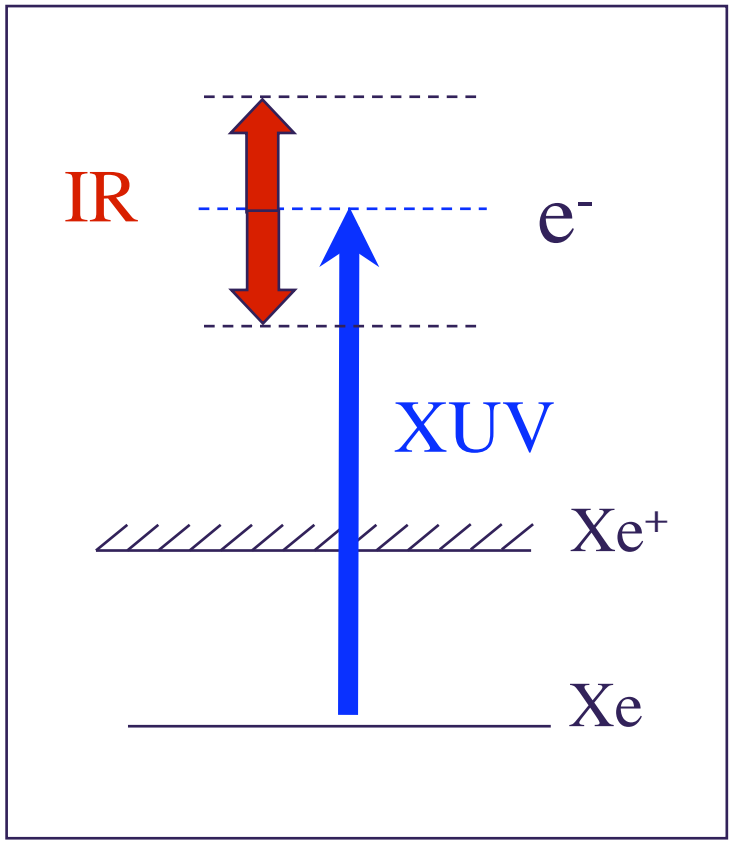


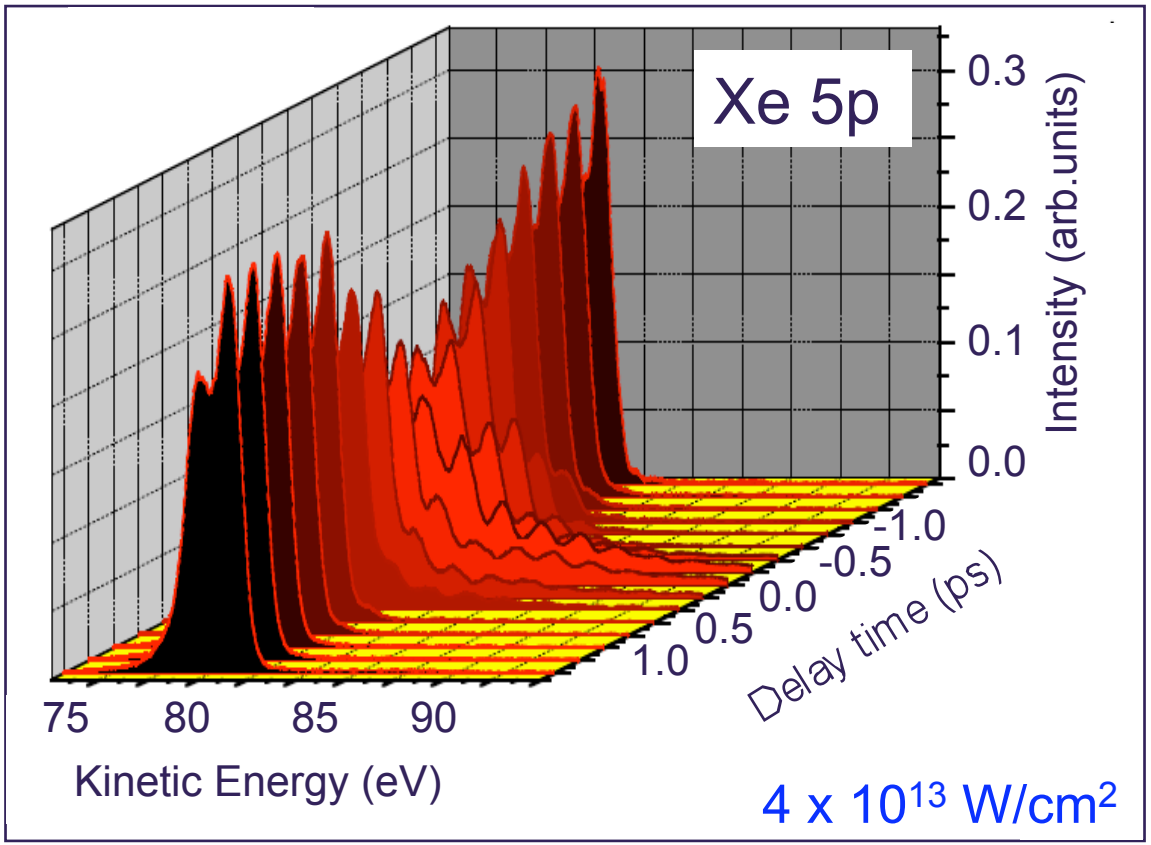
European XFEL

Characterization of FLASH pulses: ATI

FLASH: 13.7 nm, 30 μJ, 50 μm focus, 20 fs

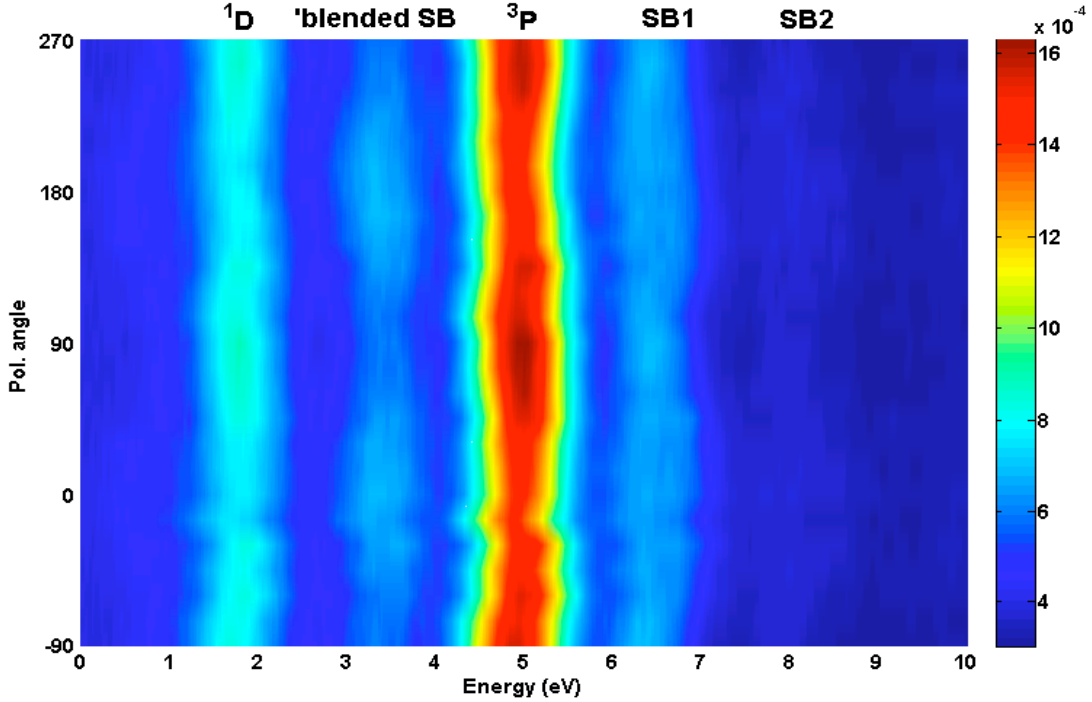
Opt. Laser : 800 nm, ≤ 4 mJ, 50 μm focus, 120 fs

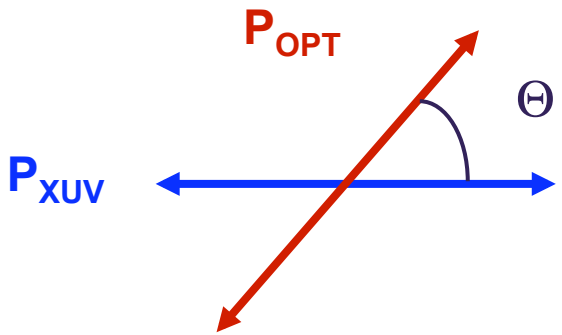




European XFEL

Strong XUV + NIR Fields



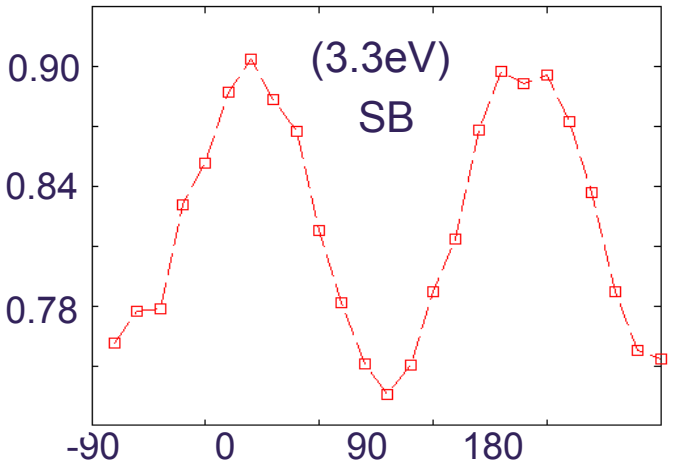


Soft Photon Approx.

O'Keeffe et al. PRA 69, 051401(R) (2004)

$$I(SB) = 1 - \frac{3\beta}{5 + 2\beta} \sin^2\Theta$$

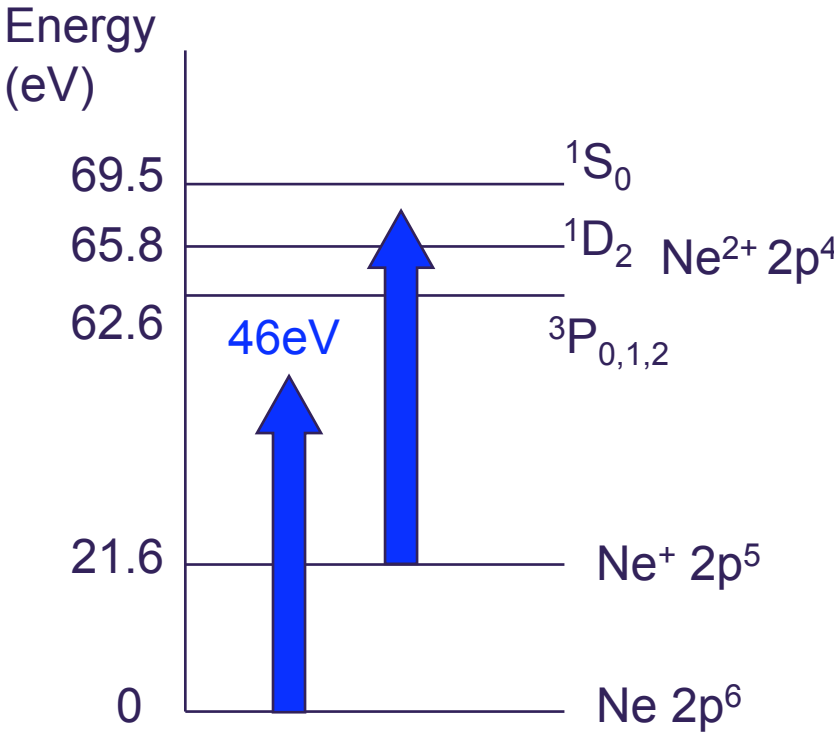
β : asymmetry parameter for TPDI



European XFEL

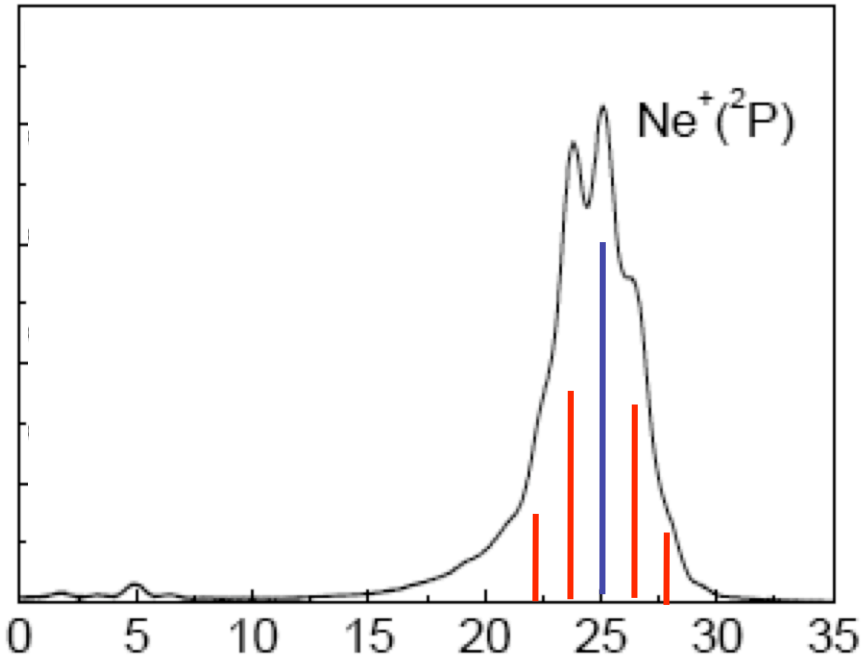
Photoionization in strong XUV + NIR Fields

Sequential Two-Photon Double-Ionization



FEL: 46 eV, 20 μJ, 30fs

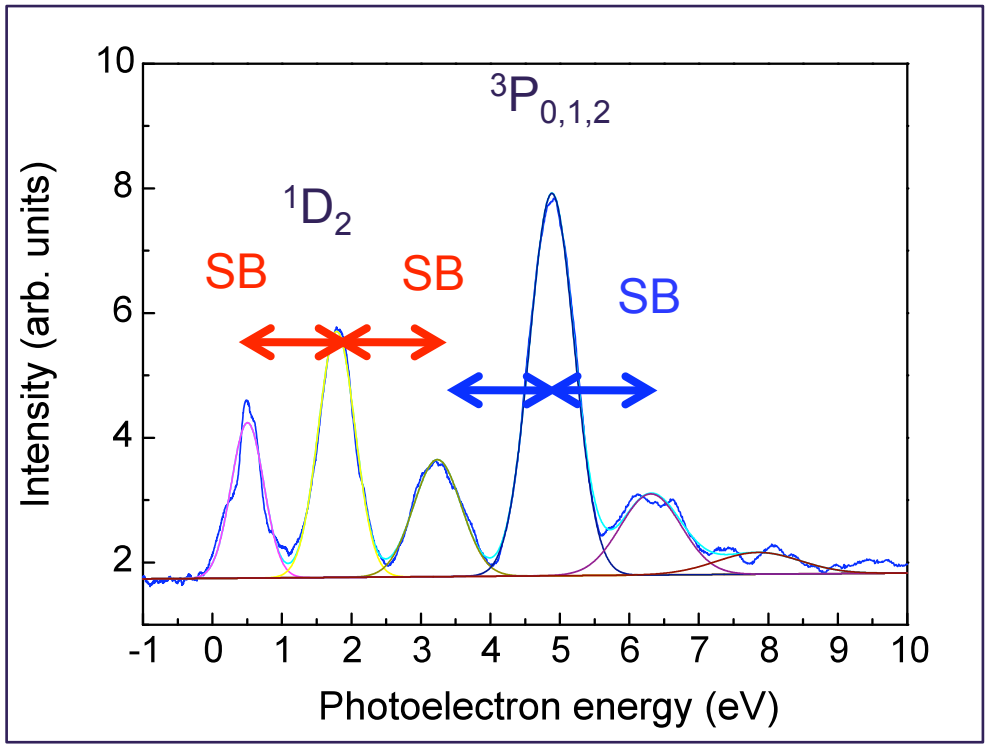
OL: 800nm, 1.8mJ, 3ps



European XFEL

Strong XUV + NIR Fields

Sequential Two-Photon Double-Ionization



$I(^1D_2) / I(^3P_{0,1,2}) = 0.67$

2-Photon Process