Atomic Photoionization Dynamics in Intense Radiation Fields

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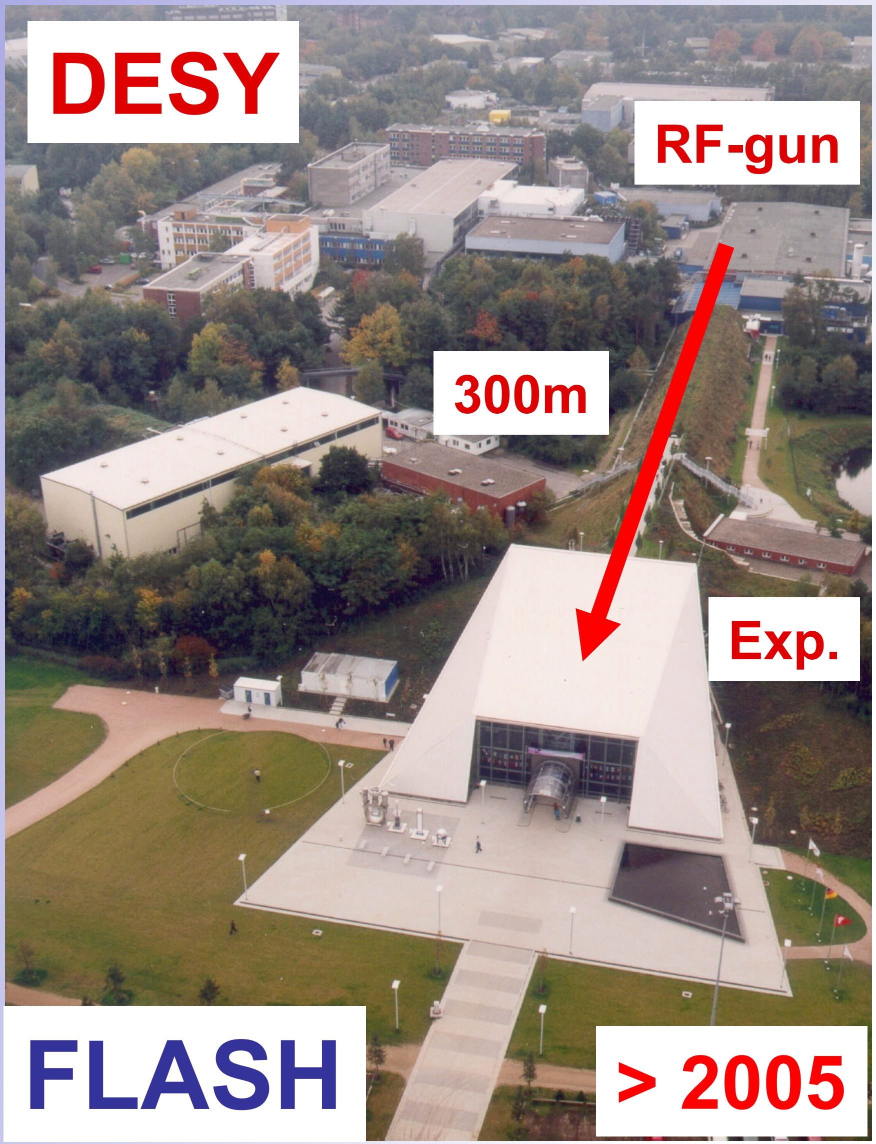
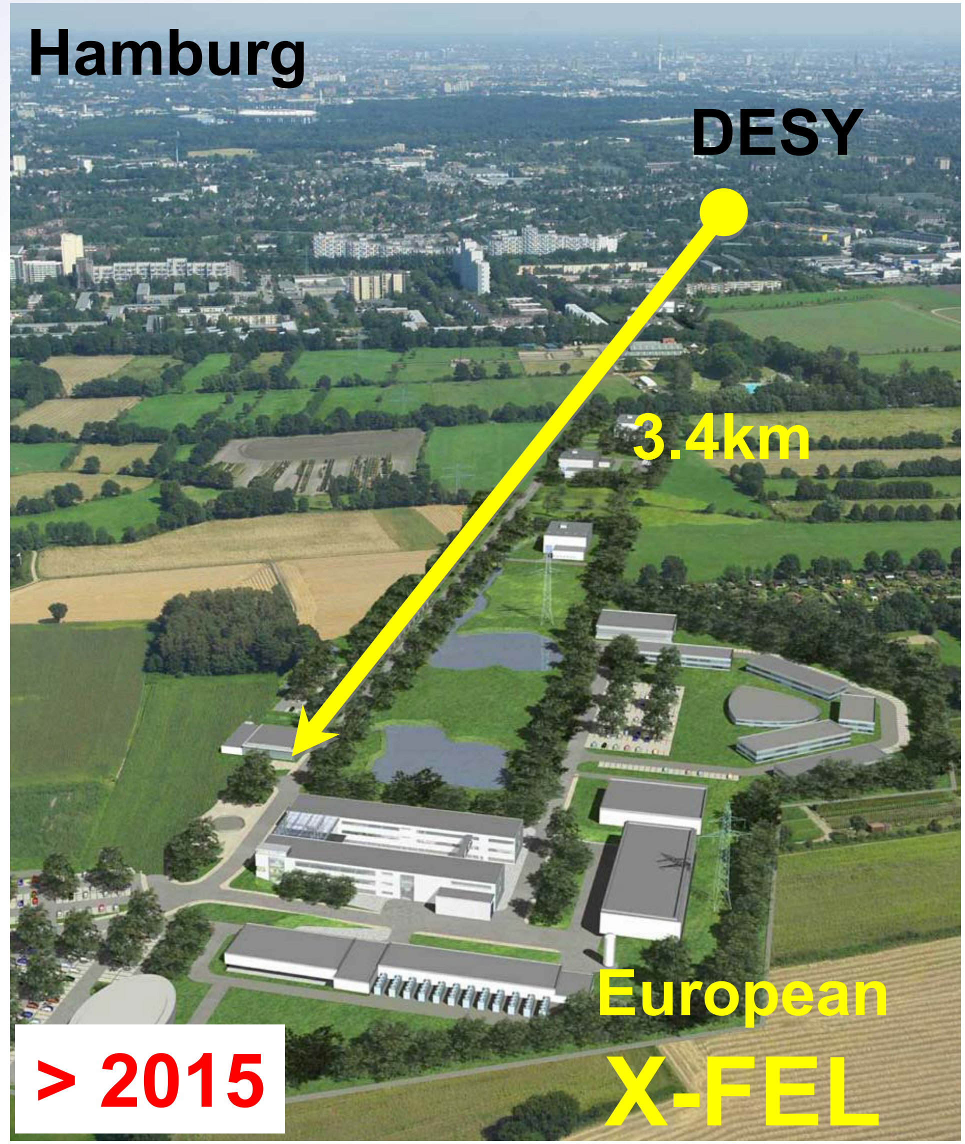
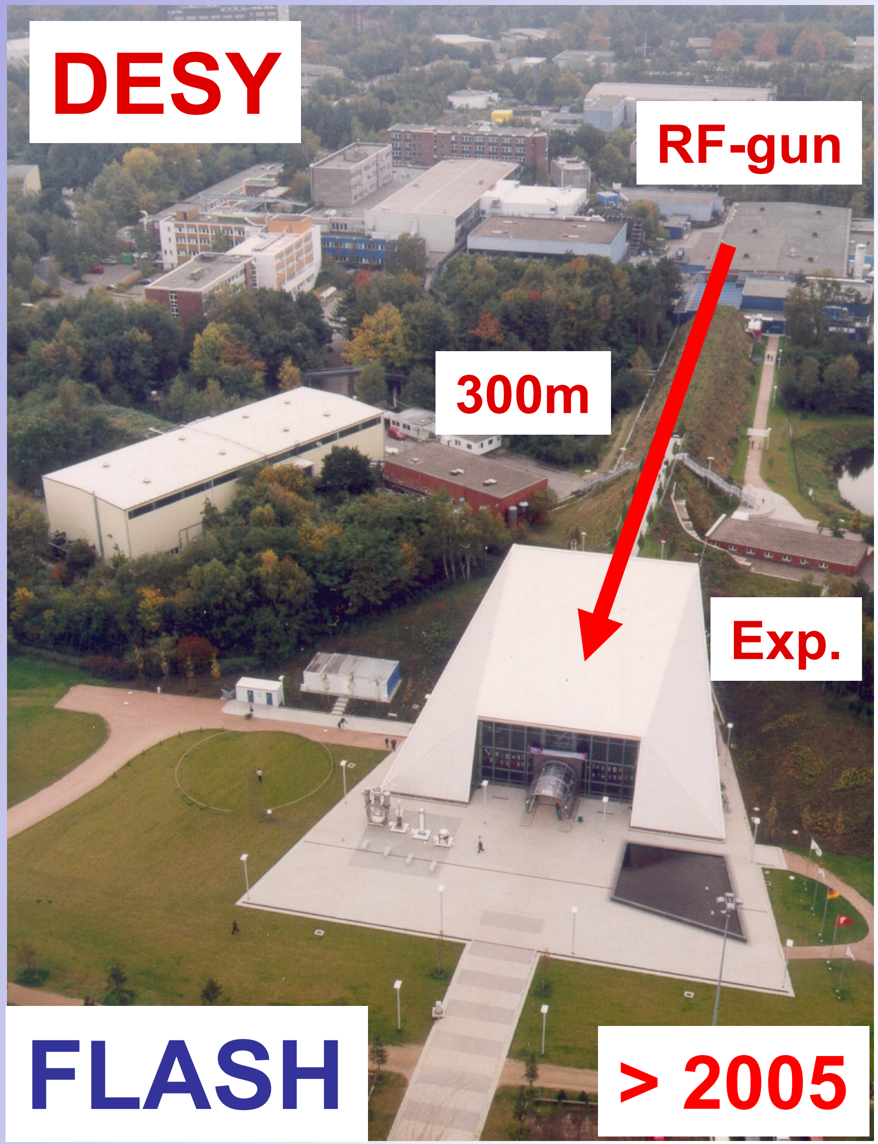
- Introduction
- Two-Color (XUV + NIR) Experiments
  - intense NIR
  - intense XUV
- Time-resolved Pump-Probe Experiments
- Conclusion

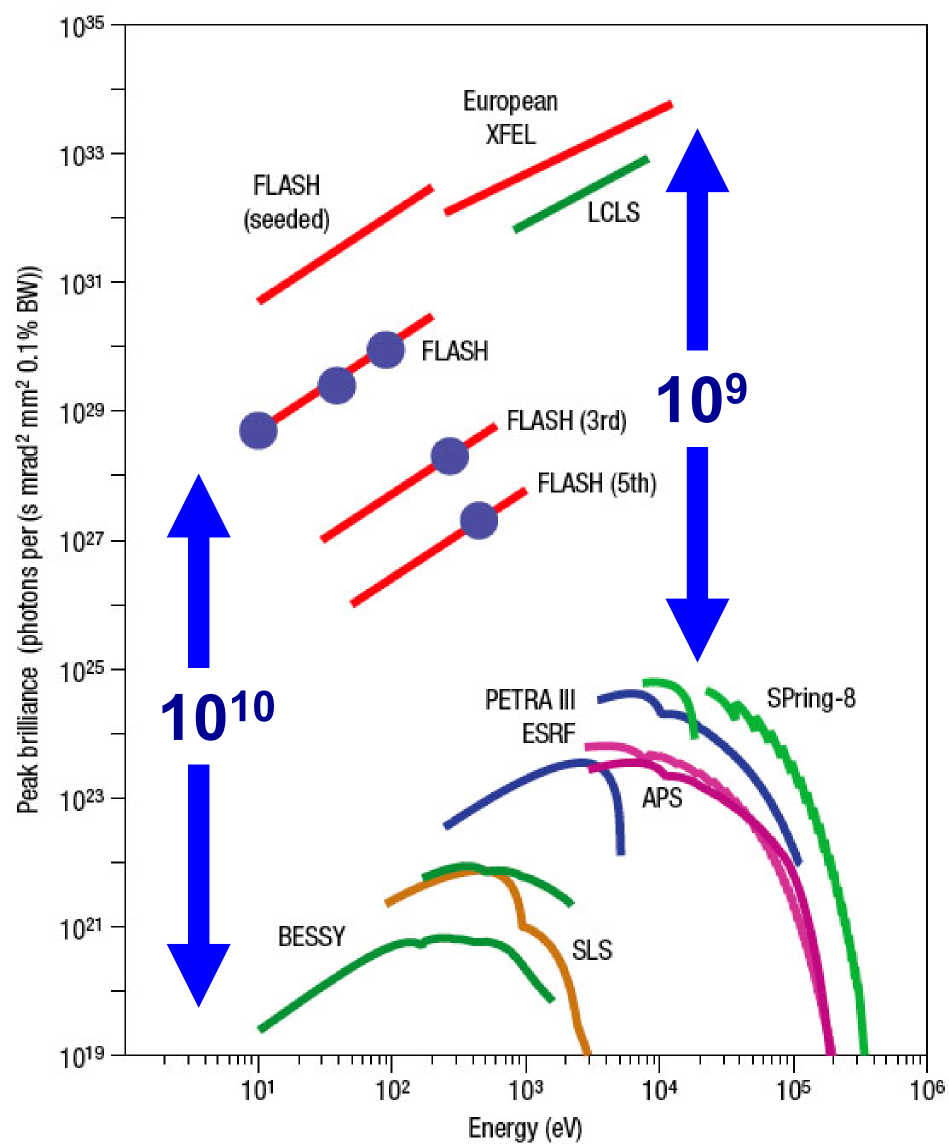
Free Electron Lasers in Hamburg





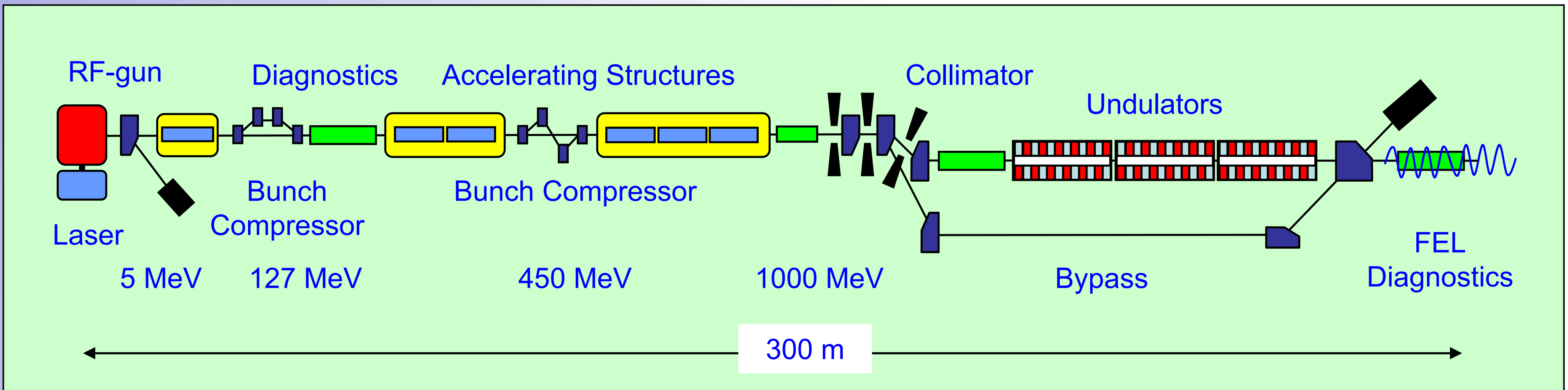
Free Electron Lasers in Hamburg





FLASH (Free electron LASer in Hamburg)

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

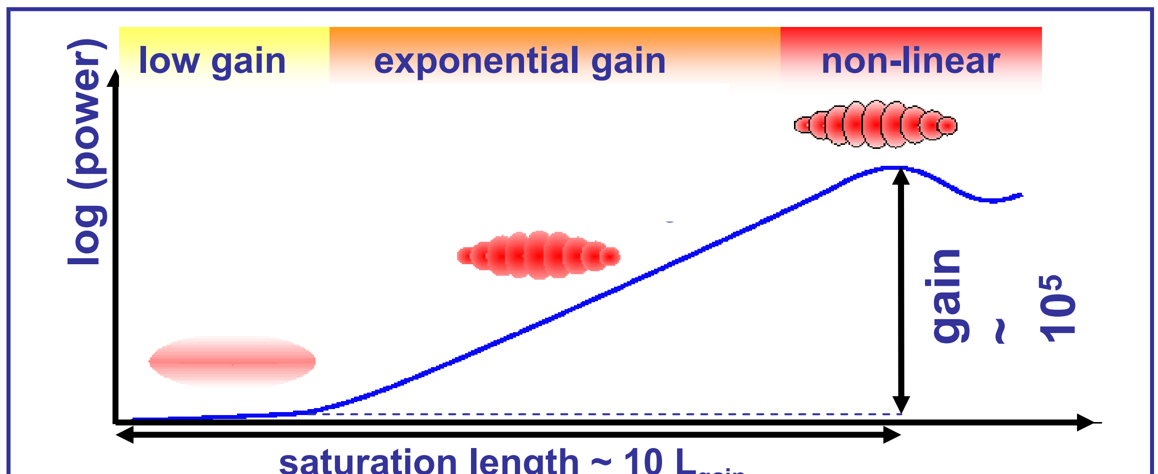


LINAC : 1 GeV

30 m fixed-gap undulator

macro-bunch at 10 Hz

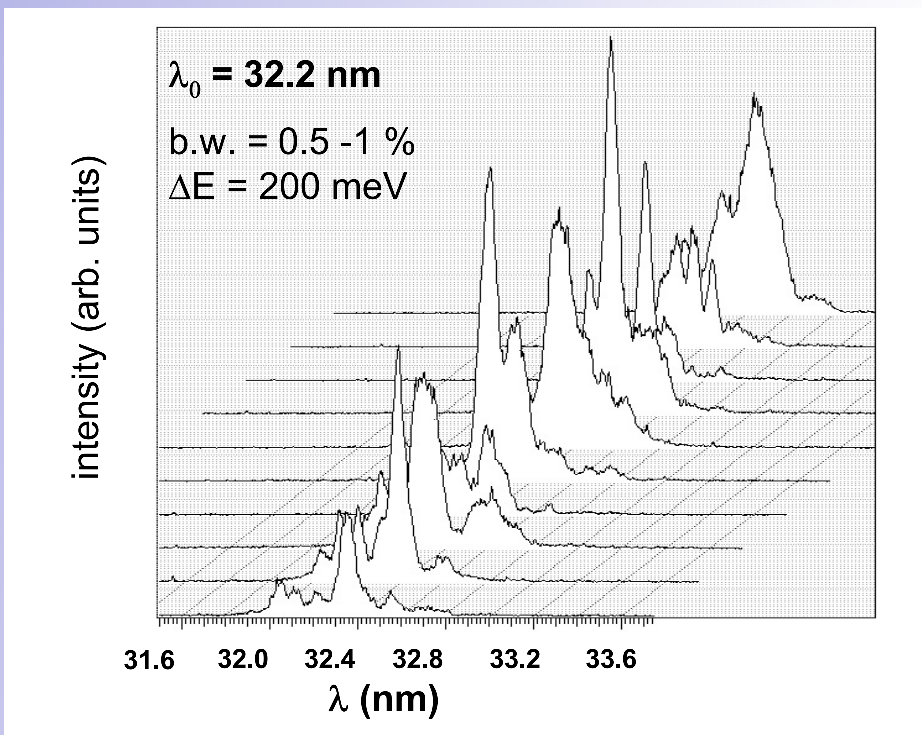
> 30 bunches, 1 μs separation



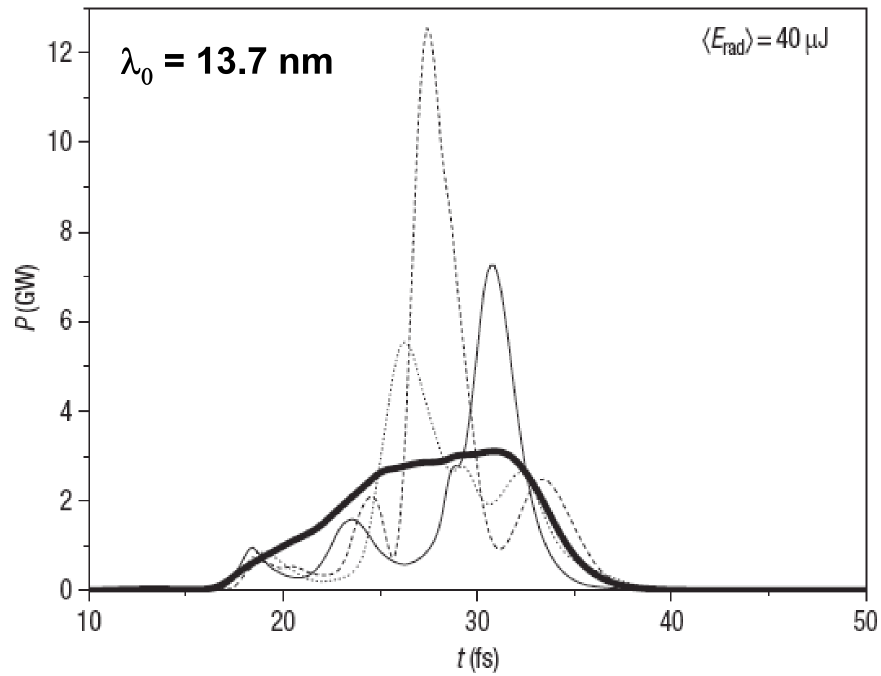
SASE (Self Amplified Spontaneous Emission)

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

FEL output builds up from spontaneous emission (photon noise)



Spectral fluctuation



Temporal fluctuation

Free Electron Laser Sources

FLASH	LCLS (Stanford, CA) SCSS (Japan)	European XFEL (Hamburg)
6 - 60 nm (0.5-1% b.w.) (20 – 200 eV)	0.15 – 1.5 nm (0.8 - 8.3 keV)	0.1 – 4.9 nm (0.25 - 12.4 keV)
5 Hz (up to 60 bunches)	120 Hz / 60 Hz	10 Hz (3000 bunches)
10 - 100 µJ (average)	> 100 µJ	> 100 µJ
10 - 30 fs	1 - 100 fs / 500 fs	< 100 fs
~ 10^{13} photons/pulse	10^{12} – 10^{13} photons/p	10^{12} – 10^{14} photons/p
> 10^{11} W / cm^2	> 10^{11} W / cm^2	> 10^{11} W / cm^2

SCIENCE at FLASH

- Intense Source

$\sim 10^{12} - 10^{13}$ photons/pulse

→ **Studies on dilute targets**
(HCl, m-sel. cluster, HeH+…)

- Short Pulses

$\Delta T = 10 - 20$ fs

$\sim 10 - 100\ \mu J\ \ (> 10^{16}\ W/cm^2)$

→ **Time-resolved studies**
(Two-Color Pump-Probe)

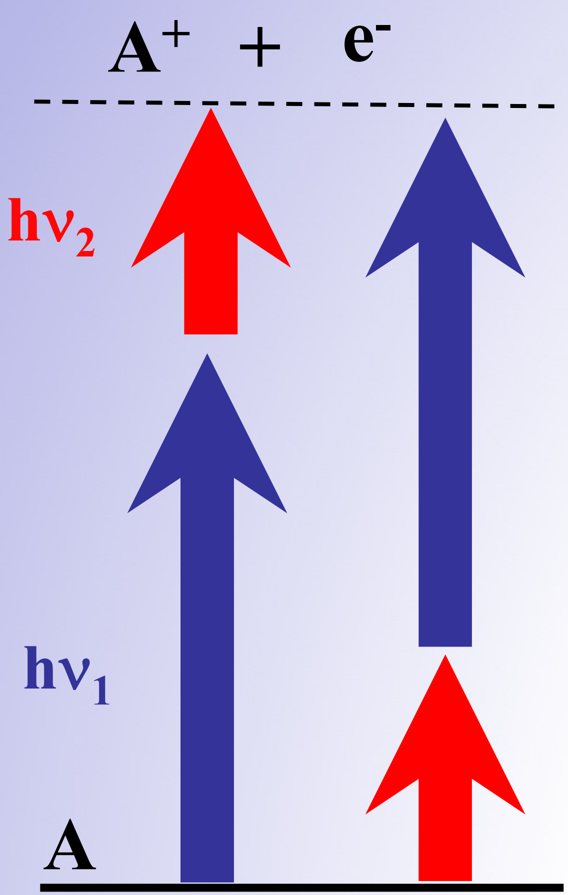
→ **Non-Linear Processes**
(Multi-photon)

- Short Wavelengths

6 - 60 nm (20 - 200 eV)

→ **Innershell Ionization**

Photoionization Dynamics



$h\nu_2$: fs – laser
$>10^{14}$ W / cm^2

$h\nu_1$: FEL
$>10^{12}$ W / cm^2

I) Non-linear processes:

- Two-photon ionization
- Photoionization of dressed atoms

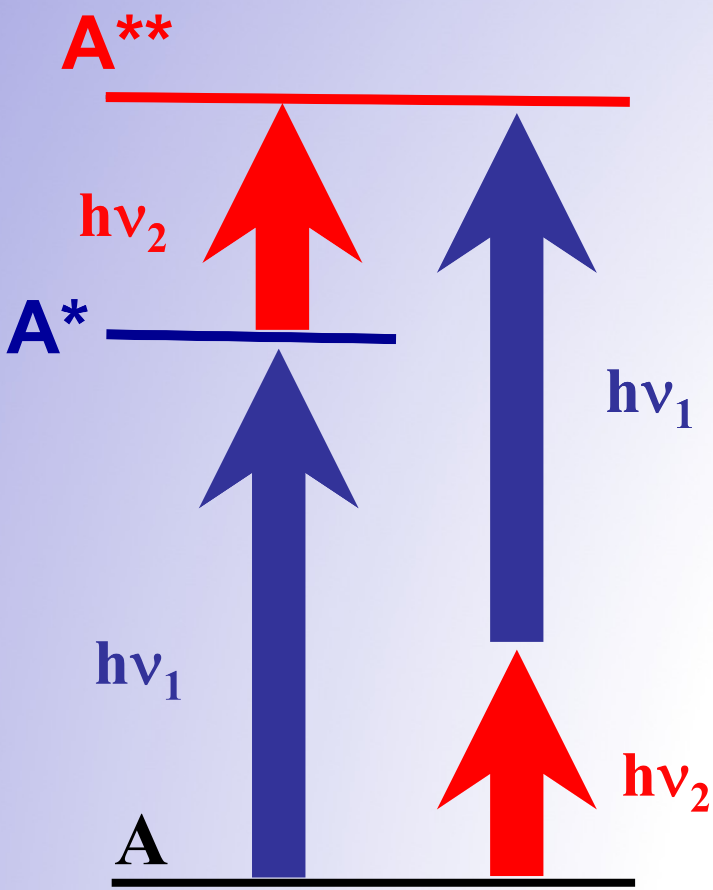
2) Dynamics in strong fields

"Strong" field:

$$A + h\nu_1 + h\nu_2 \rightarrow A^+ + e^-$$

One-step process !!

Relaxation Dynamics of Core Resonances



$h\nu_2$: fs – laser
$>10^{14}$ W / cm²

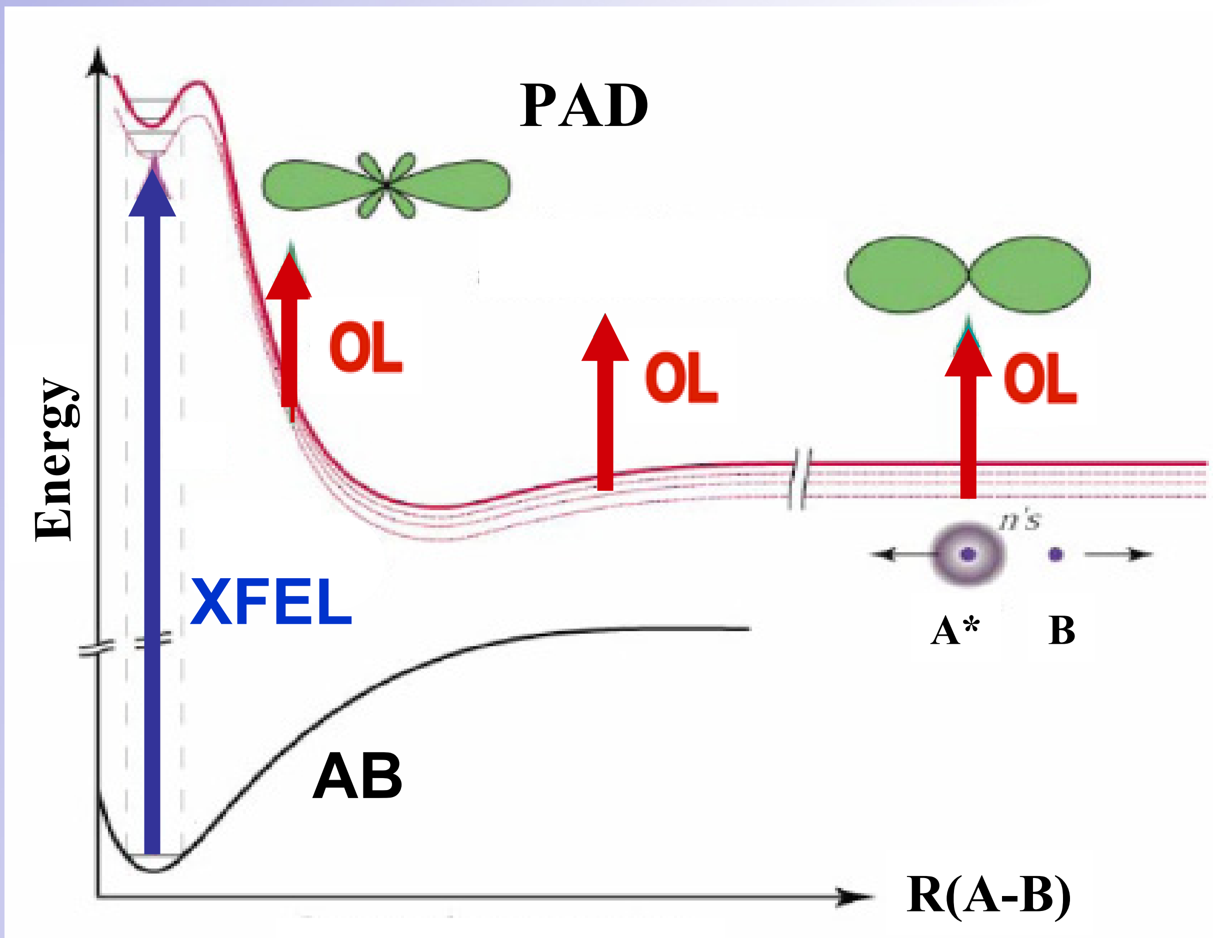
$h\nu_1$: FEL
$>10^{12}$ W / cm²

3) Non-linear processes:

- Two-photon resonances
- Coupling of Autoionizing States

Photodissociation Dynamics

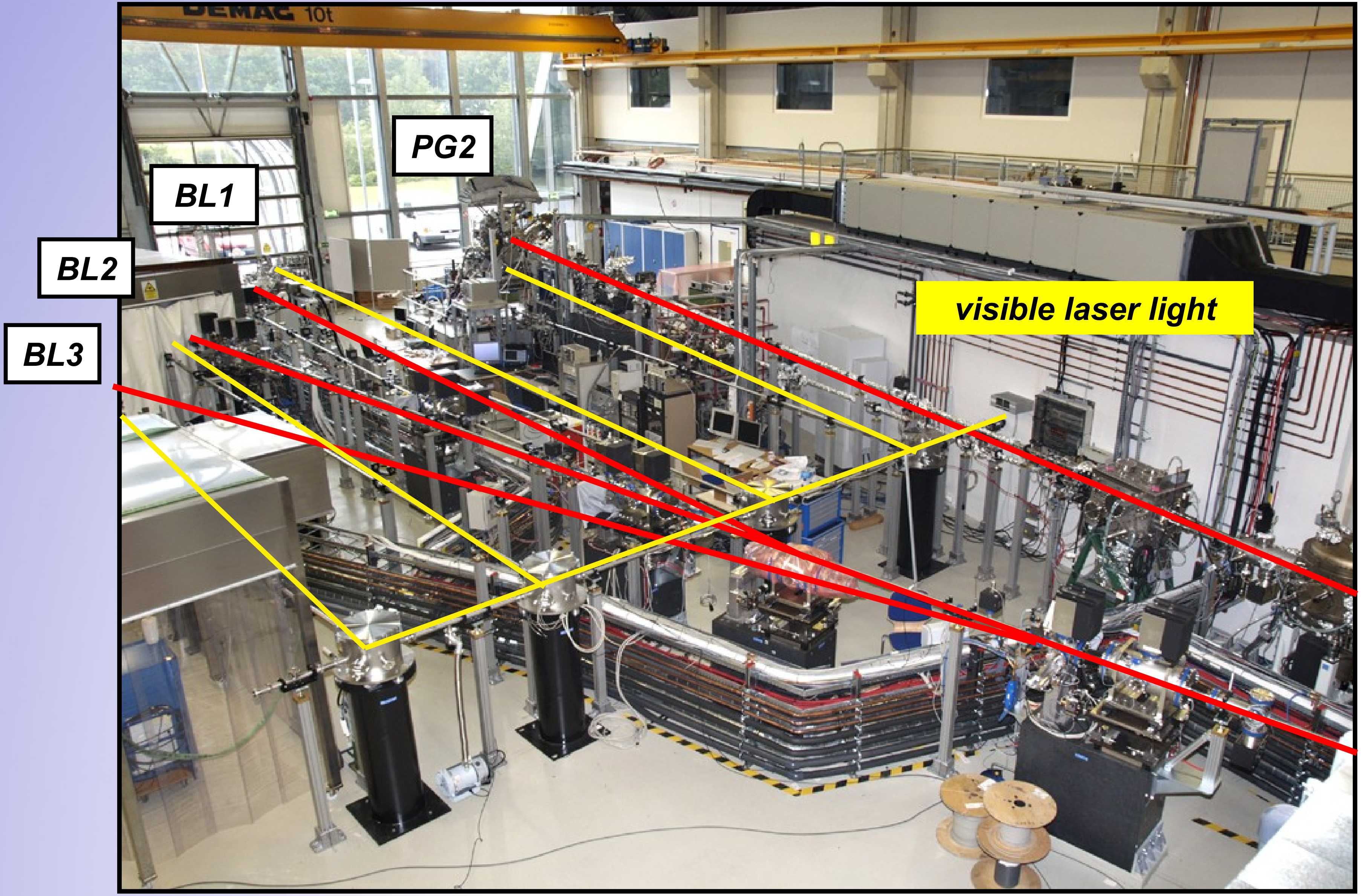
Time-resolved development of the electronic structure during dissociation or chemical reactions



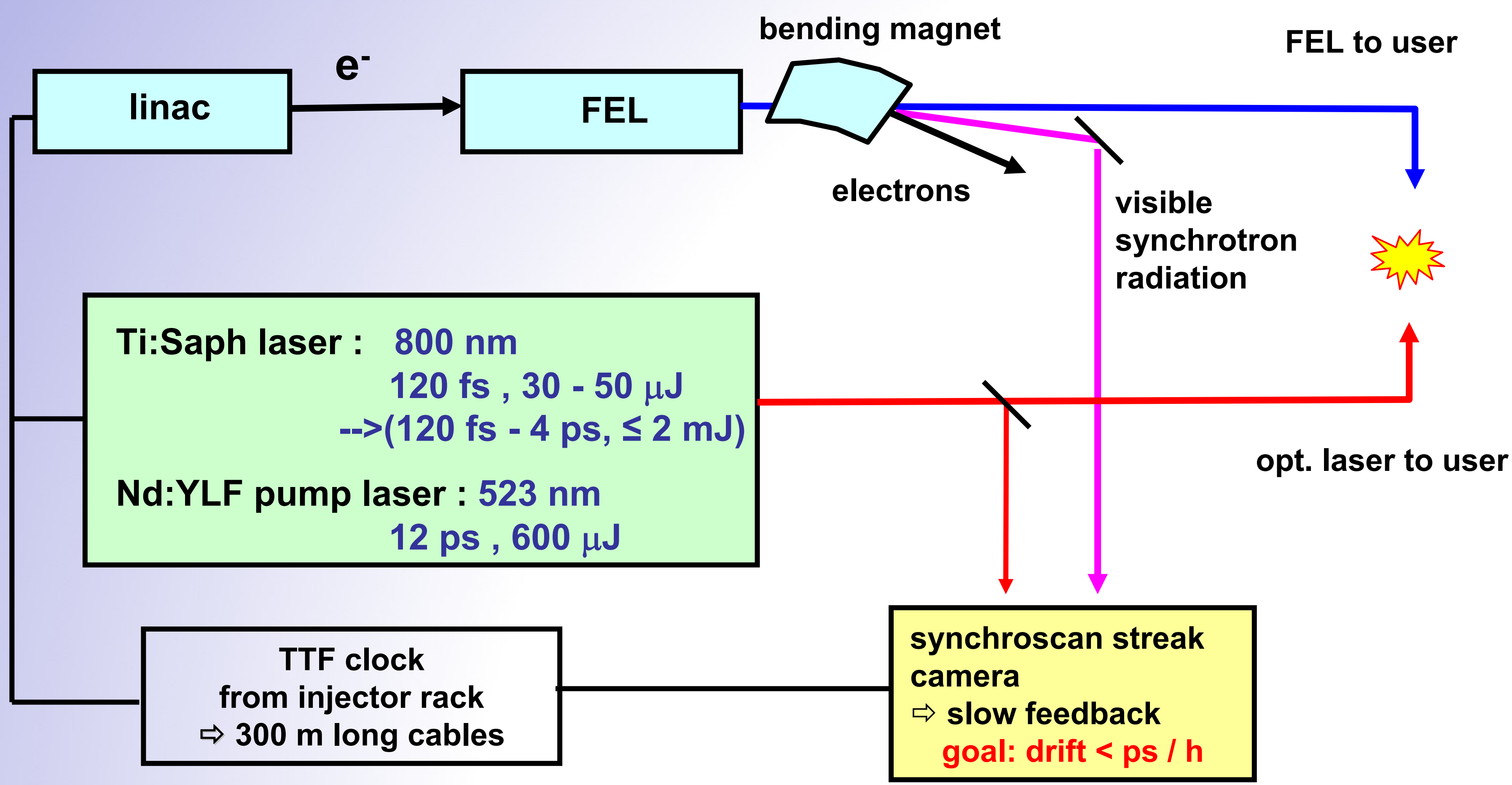
Excitation of core electrons

element specific « Pump » or « Probe »

Experiments at FLASH



FLASH + Optical Laser

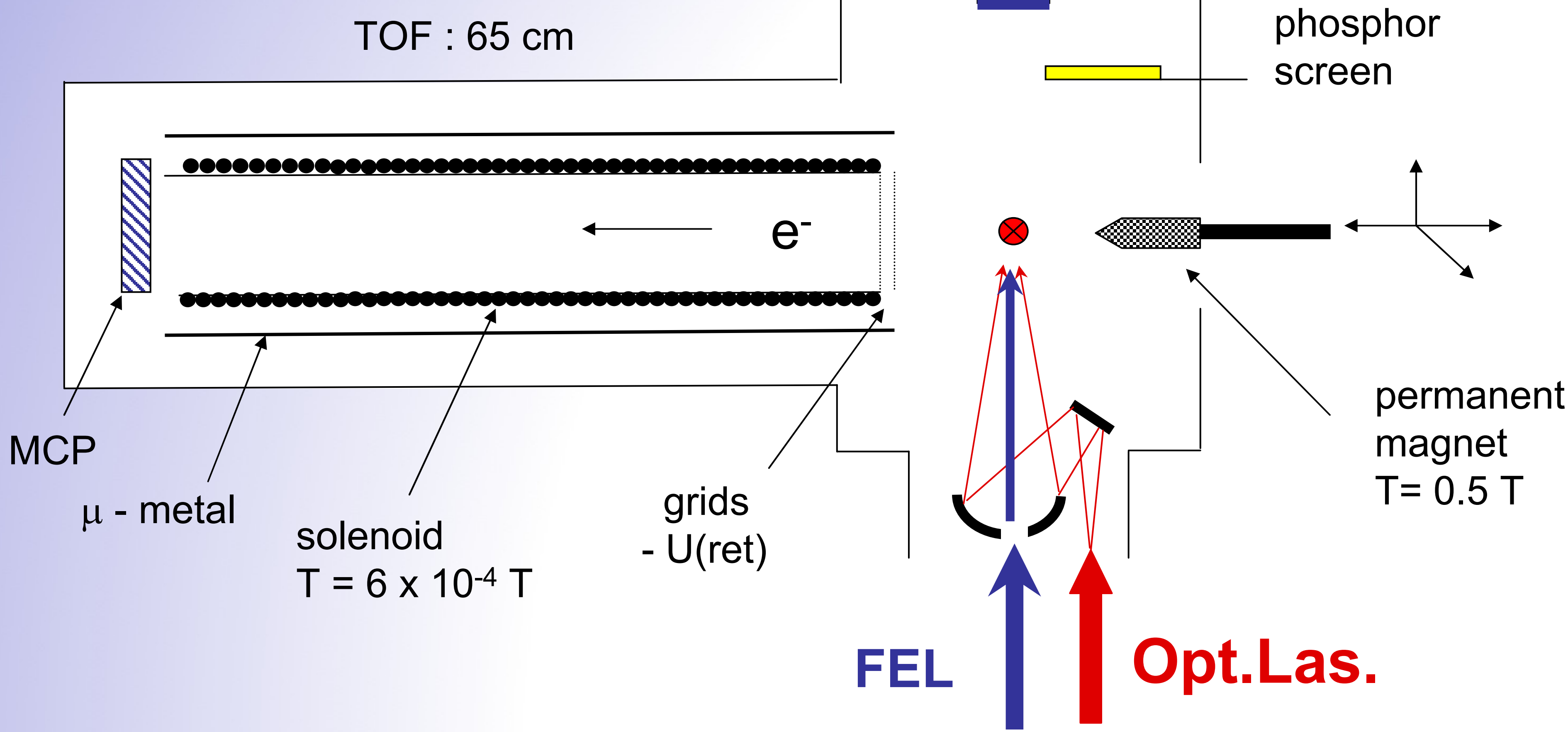


Max-Born-Institut, Berlin
I. Will, H. Redlin

Two-Color Photoelectron Spectroscopy

MBES: Magnetic Bottle Electron Spectrometer
(4π collection angle)

J.H.D. Eland et al. PRL90,53003 (2003)



Two-color experiments

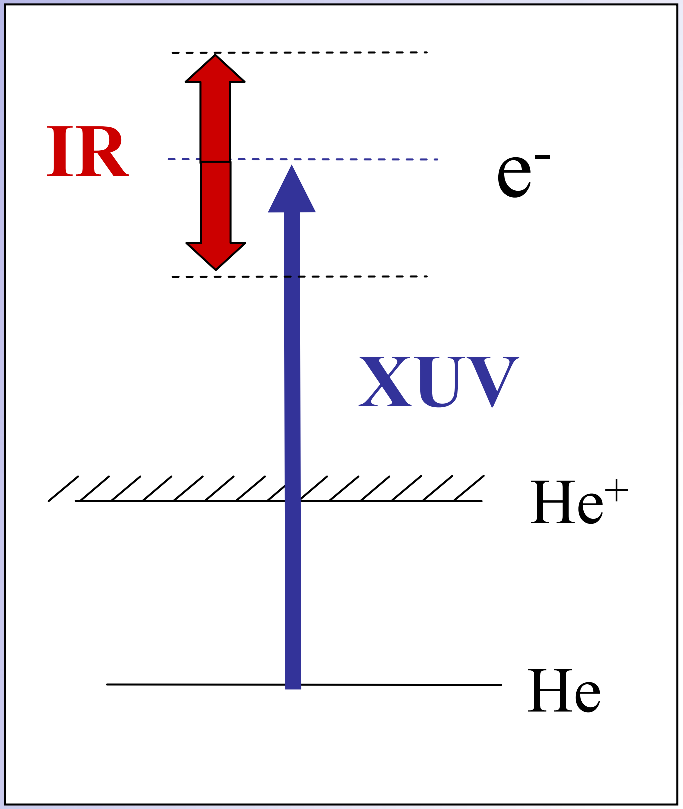
Intense optical field: Above Threshold Ionization of rare gases

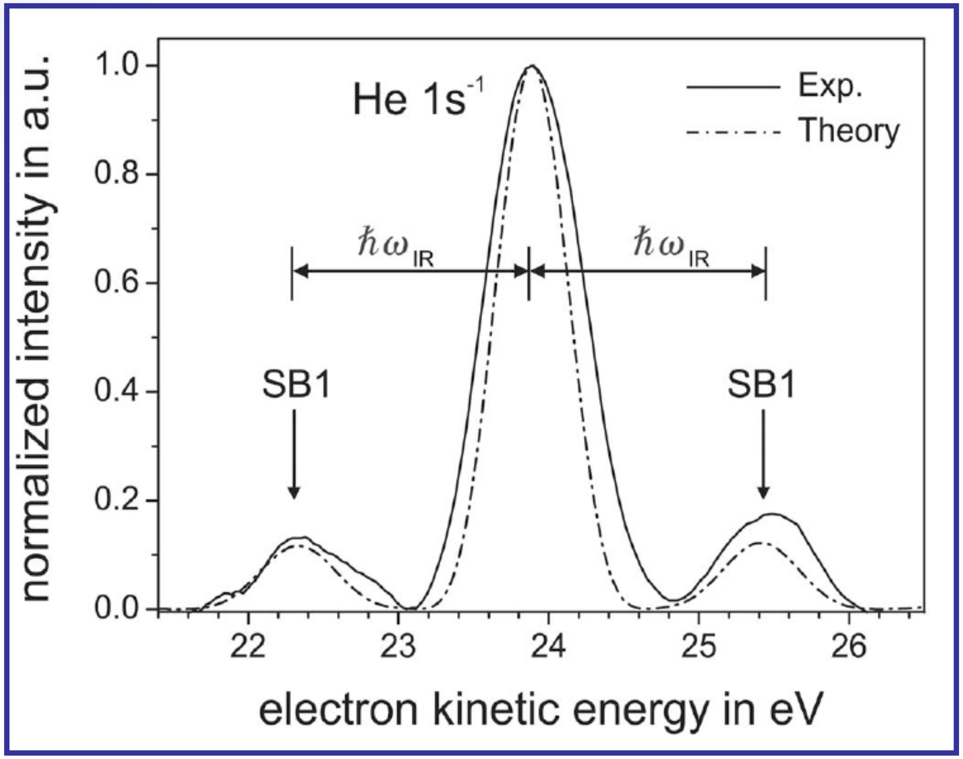
Above Threshold Ionization of Rare Gases

FLASH: 25.5 nm, 20 μJ, 50 μm focus, 20 fs

Opt. Laser : 800 nm, 20μJ, 50 μm focus, 12 ps

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





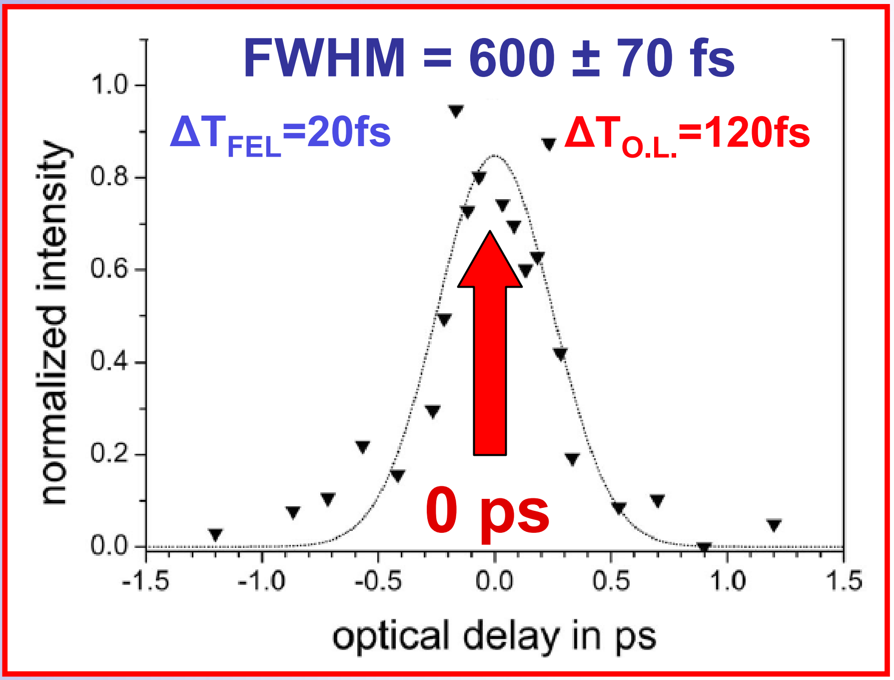
Toma et al. PRA 62, 0618015 (2000)

Radcliffe et al. APL 90, 131108 (2007)

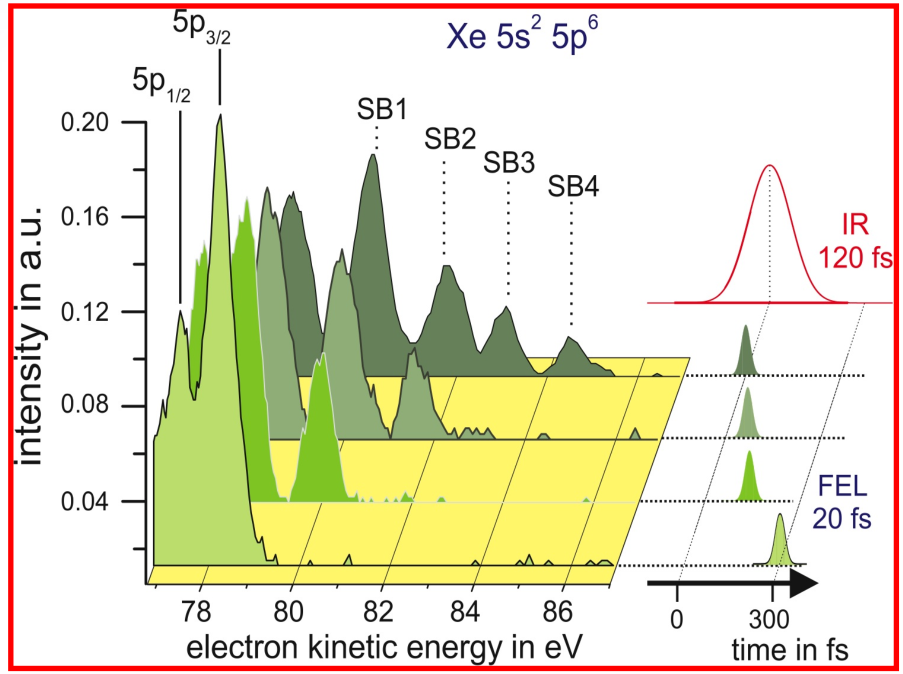
Maquet/Taieb, J.Mod.Opt. 54, 1847 (2007)

Temporal resolution / Synchronization : ATI

Average Mode:
Cross Correlation Curve



Single Shot Mode:
Photoelectron spectra



temporal resolution $\Delta T = 600$ fs

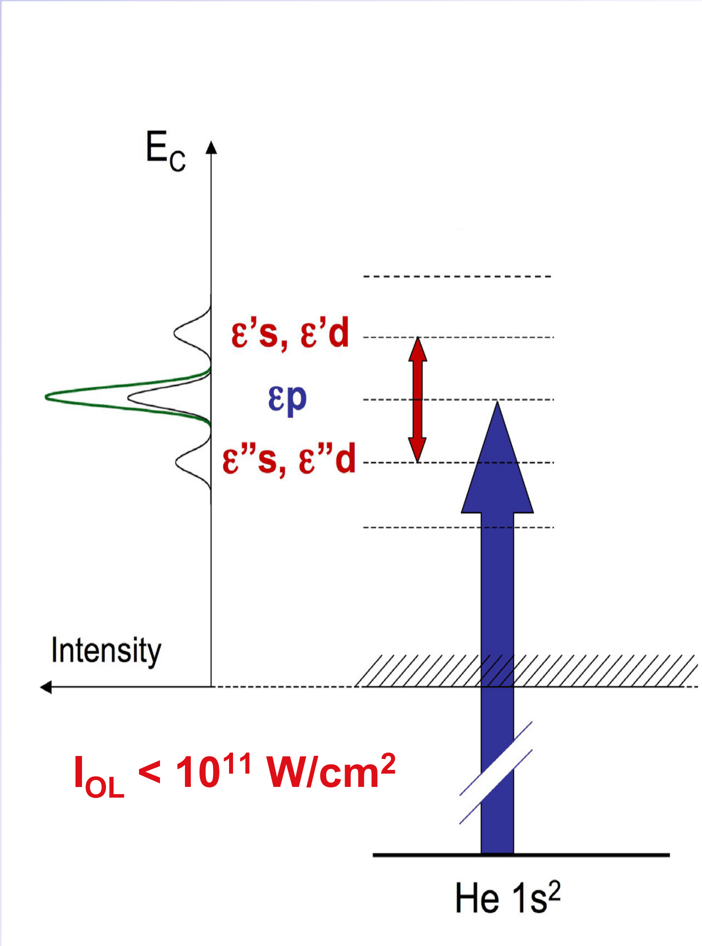
→ Jitter (FEL) = 550 ± 80 fs

temporal resolution $\Delta T \leq 50$ fs

→ only for overlapping pulses

M. Meyer et al. PRA 74 011401R (2006), P. Radcliffe, et al., APL 90 131108 (2007)
A. Maquet, R. Taïeb, J. Mod. Opt. 54 1847 (2007)

Polarization control in two-color photoionization



M. Meyer et al., PRL 101, 193002 (2008)

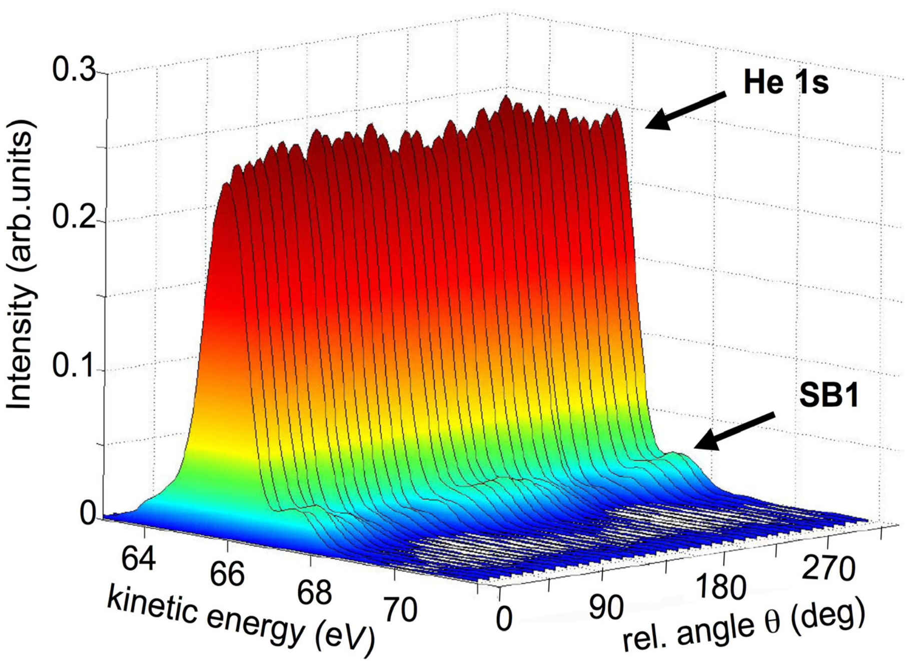
He $1s^2$ + $h\nu_{XUV}$ ----> He^+ 1s + εp

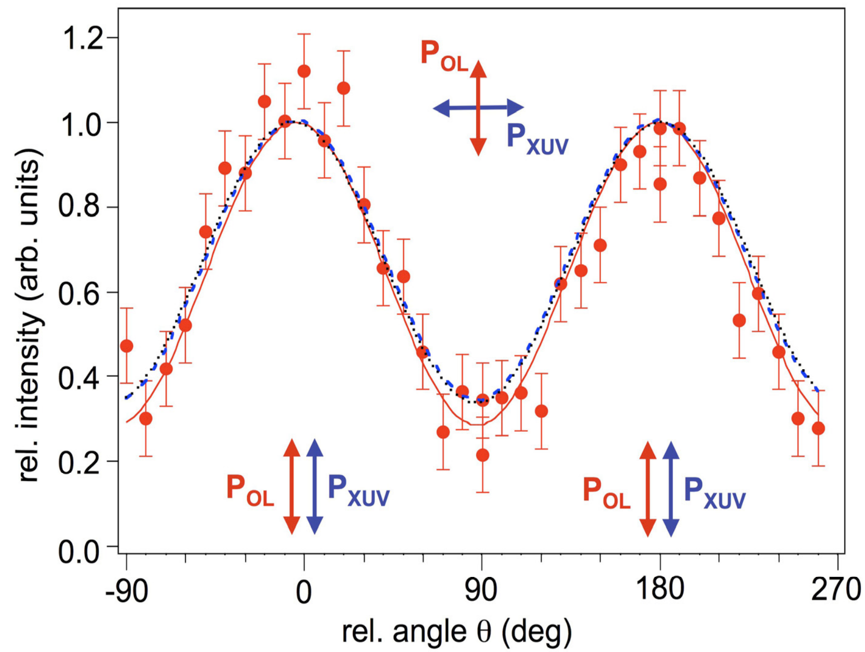
He $1s^2$ + $h\nu_{XUV}$ + $h\nu_{OL}$ ---> He^+ 1s + εs, εd

Polarization control: two-color **two-photon**

FLASH: 13.7 nm, 10-20 fs, 20µJ
OL: 800nm, 4ps, 70µJ, 8 x 10^{10} W/cm^2

$$\sigma(\theta) = 3S_d + (5S_s + S_d)\cos^2\theta$$





Two-photon ionization

He 1s^2 + hν_{XUV} + hν_{OL} ---> He$^+$ 1s + εs, εd

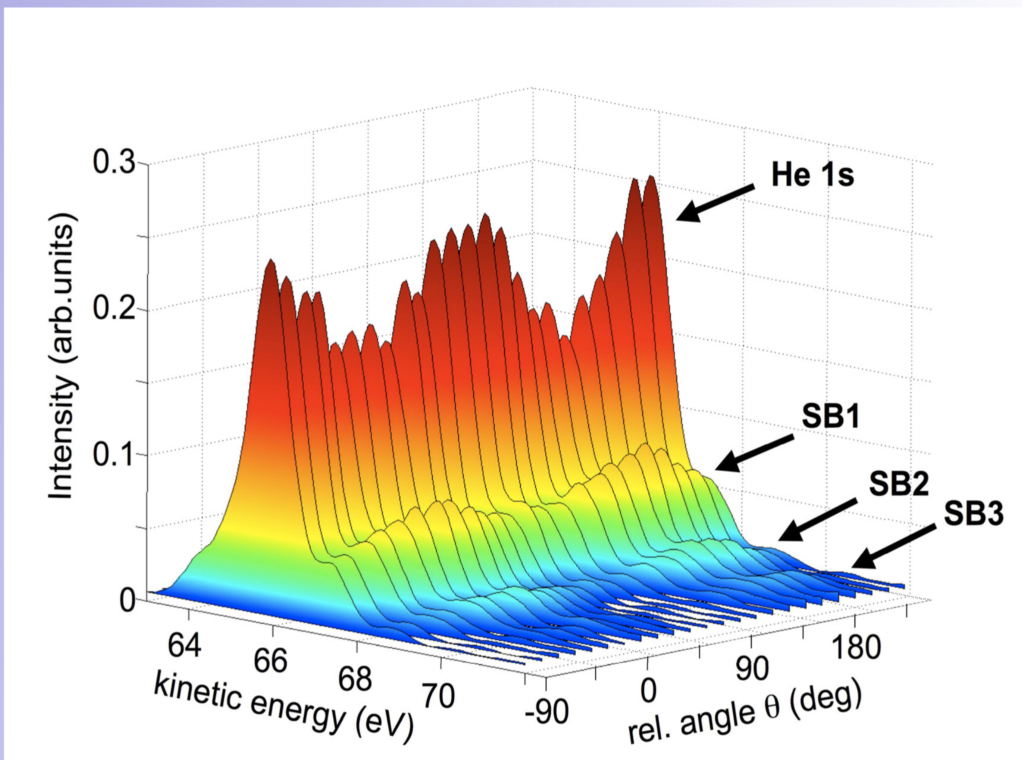
$$S_s / S_d = 1.25 \pm 0.3$$

90° → only 'd' - emission
0° → 1.5 times more 's' than 'd'

Polarization control: two-color multi-photon

FLASH: 13.7 nm, 10-20 fs, 20µJ

OL: 800nm, 4ps, 400µJ, 6 x 10^{11} W/cm^2

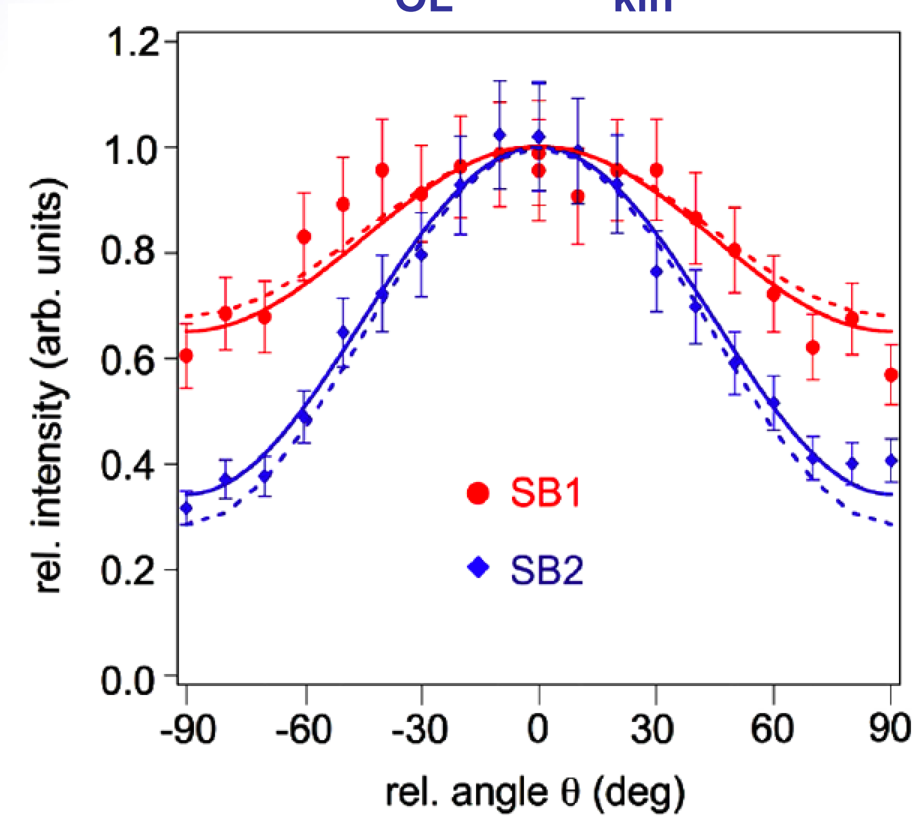


He $1s^2$ + $h\nu_{XUV}$ + $h\nu_{OL}$ ---> He^+ 1s + εl

higher l ≥ 0(s), 2(d)

"Soft-Photon Approximation"

$h\nu_{OL} \ll E_{kin}$

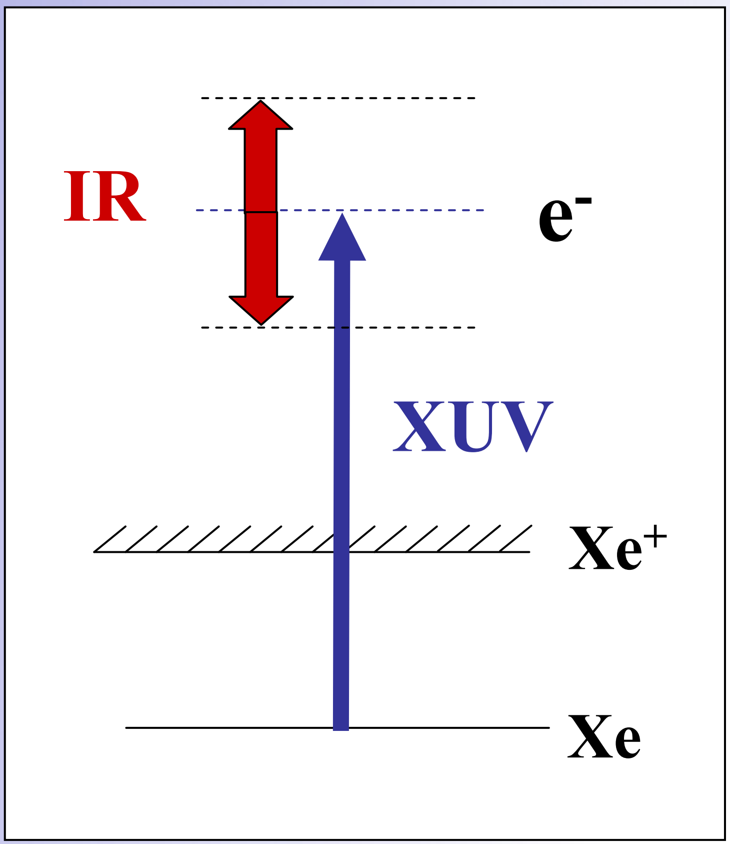


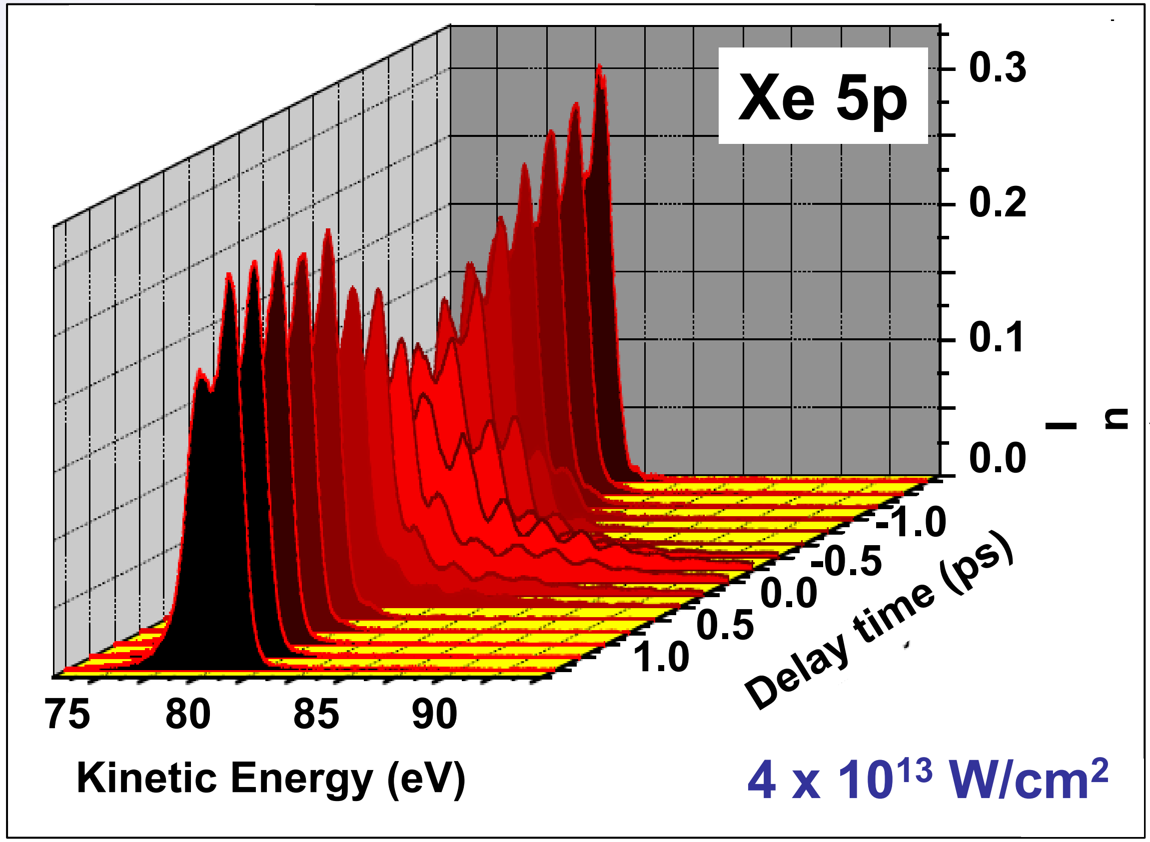
A. Maquet, R. Taieb, J. Mod. Opt. 54, 1847 (2007)

Temporal Control of ATI

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

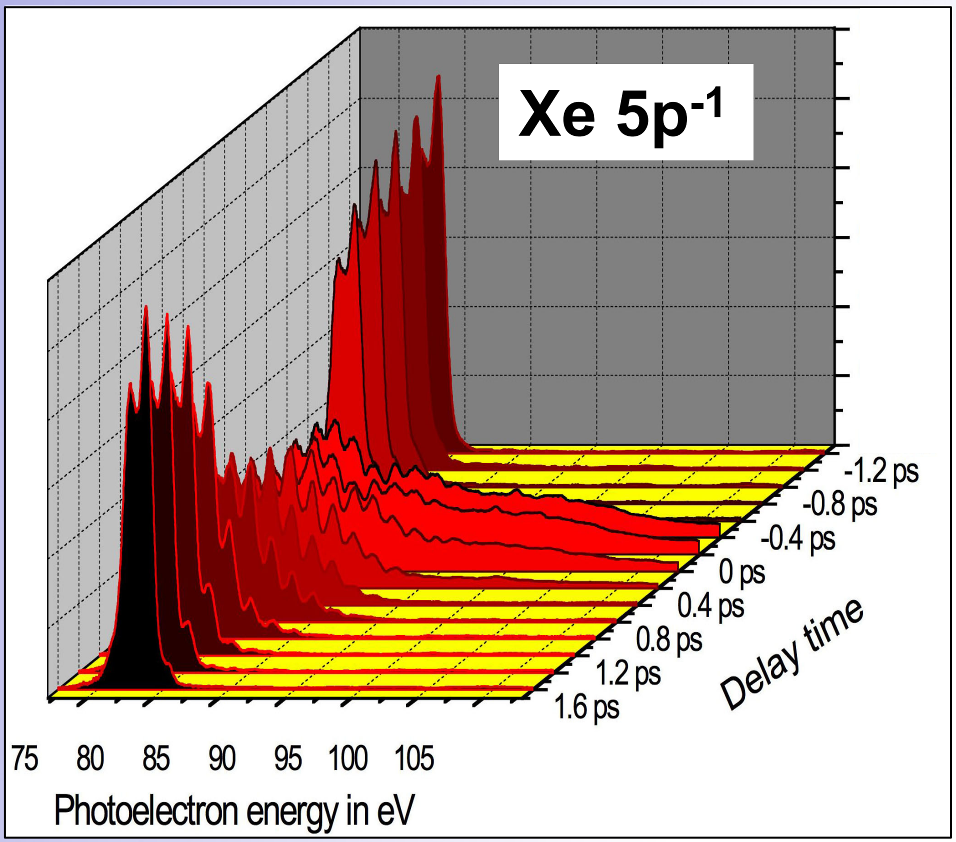
Opt. Laser : 800 nm, ≤ 4 mJ, 50 μm focus, 120 fs - 4 ps

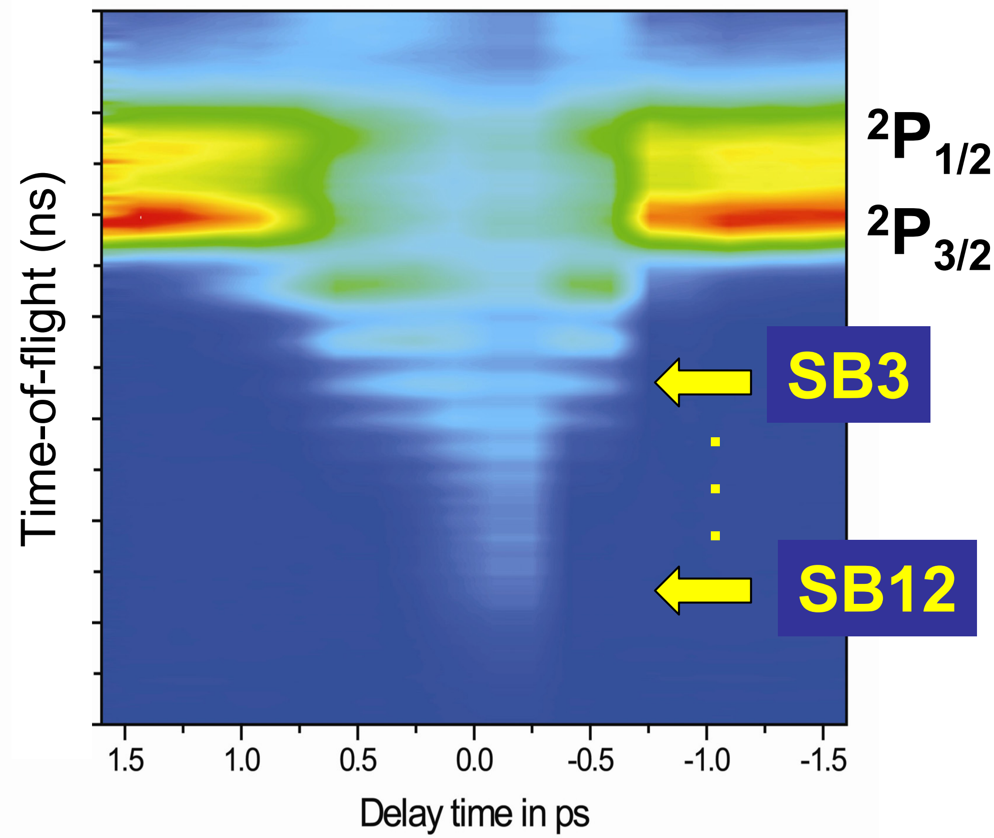




ATI : Strong NIR Dressing Field (Xe)

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





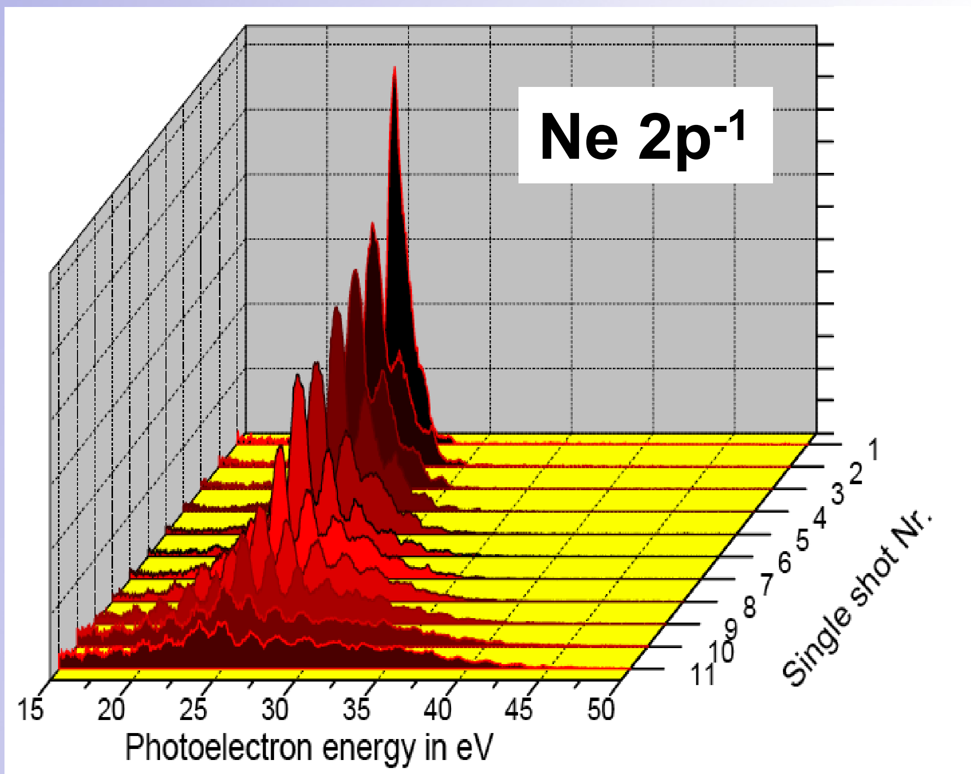
Multi-photon processes

hν(800nm) = 1.5eV

$\Delta E(^2P_{3/2} - {}^2P_{1/2}) = 1.3$eV

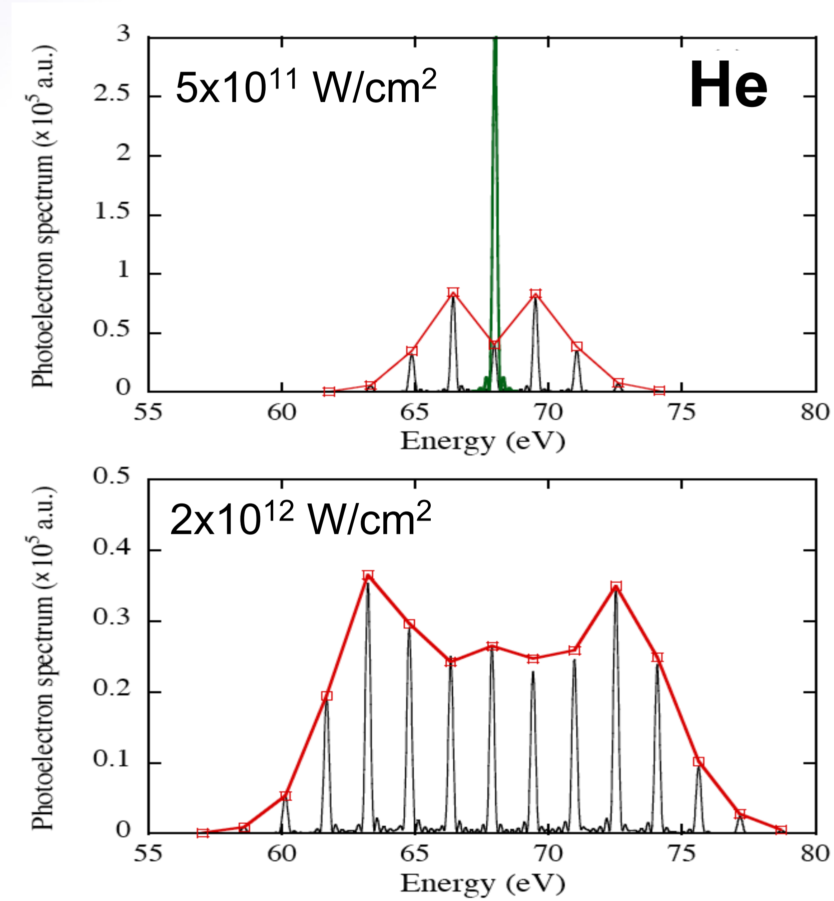
ATI : Strong NIR Dressing Field (Ne)

FEL: 26.9nm (46 eV)
Laser: 800nm, 1.8mJ, 100fs
$3x10^{13}$ W/cm^2



Multi-photon processes

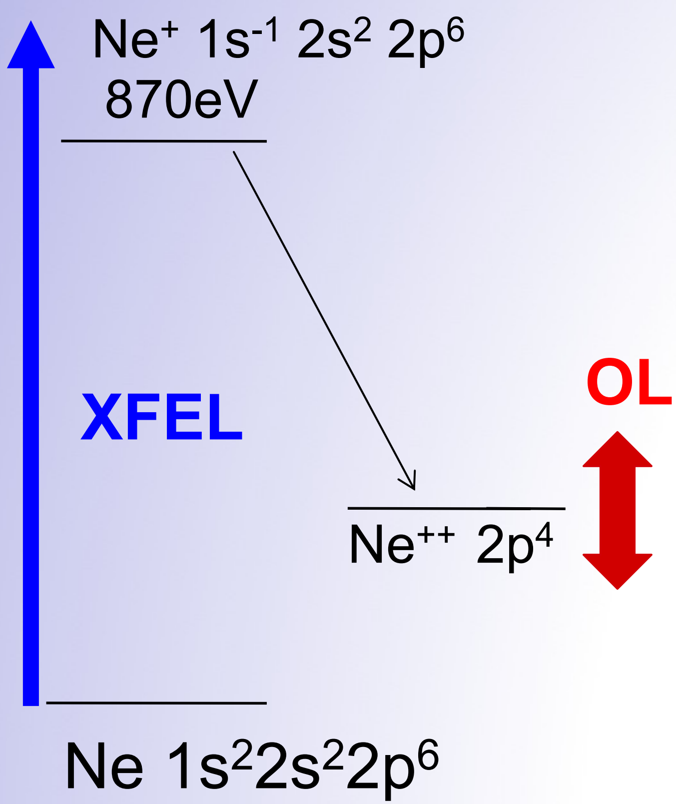
"Soft-Photon Approximation"

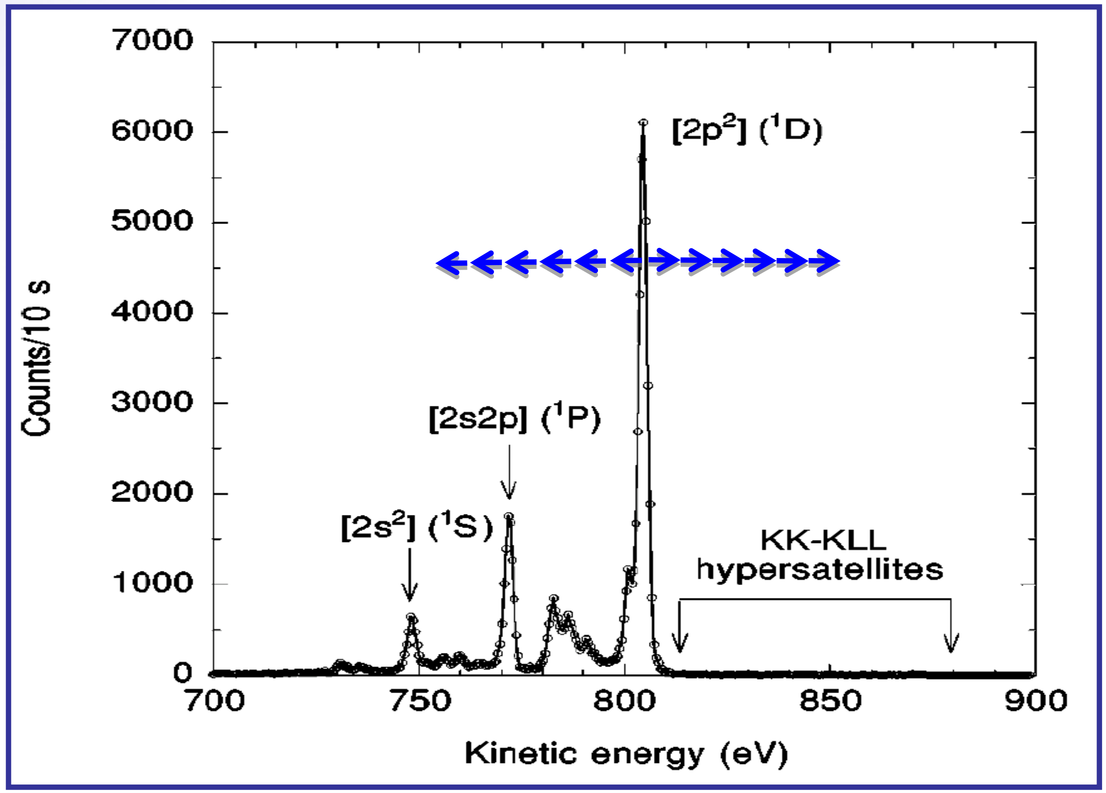


A. Maquet, R. Taieb, J. Mod. Opt. 54, 1847 (2007)

XFEL: Laser-assisted resonant Auger decay

Laser Coupling of Final Ionic States

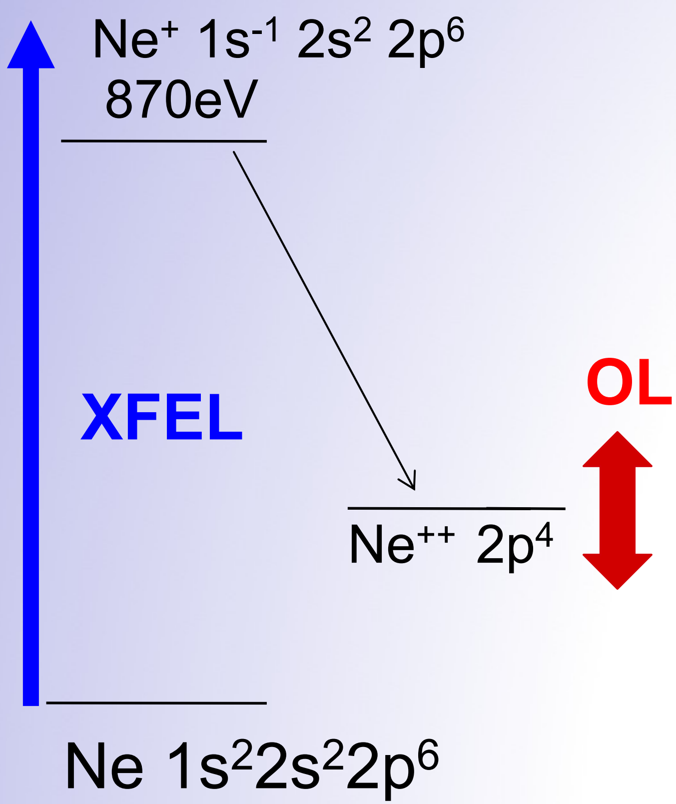




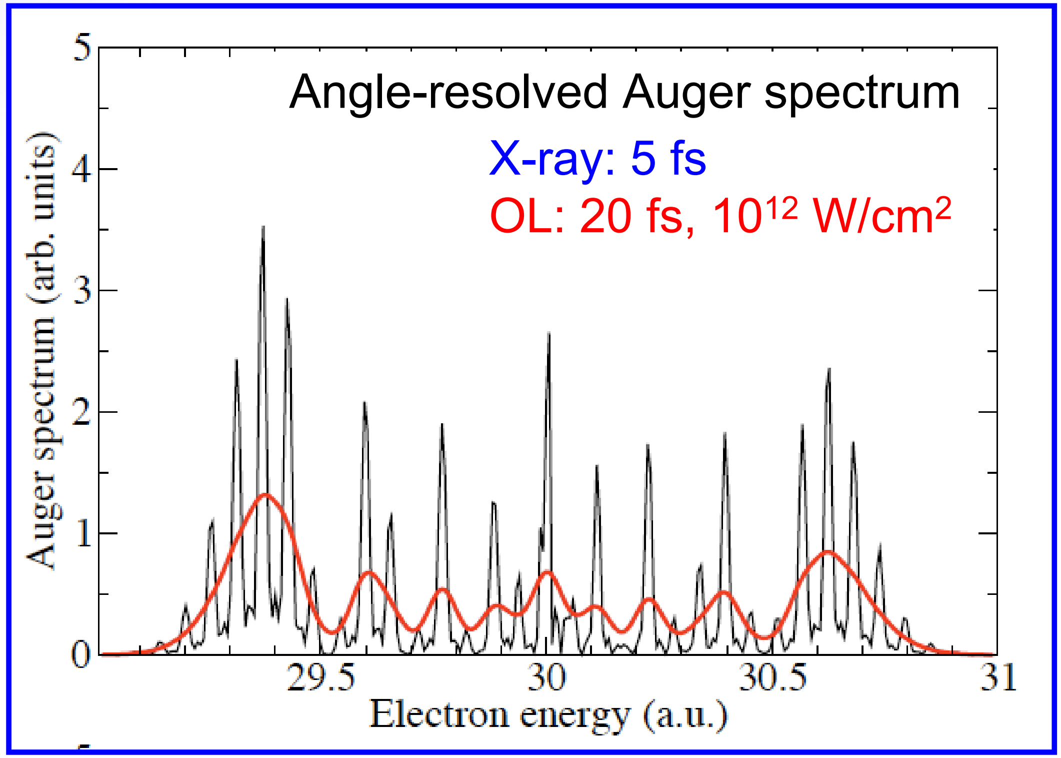
Southworth et al. PRA 52, 1272 (1995)

XFEL: Laser-assisted resonant Auger decay

Laser Coupling of Final Ionic States



A. Kazansky, N. Kabachnik, J. Phys. B (2009)



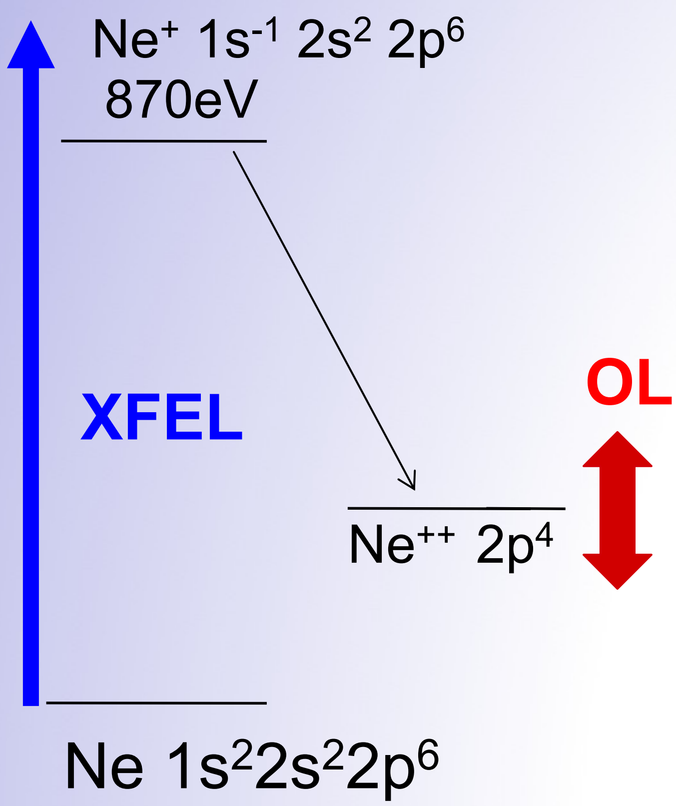
Interference of electron emission within one cycle of the optical laser field!!!

XFEL: Laser-assisted resonant Auger decay

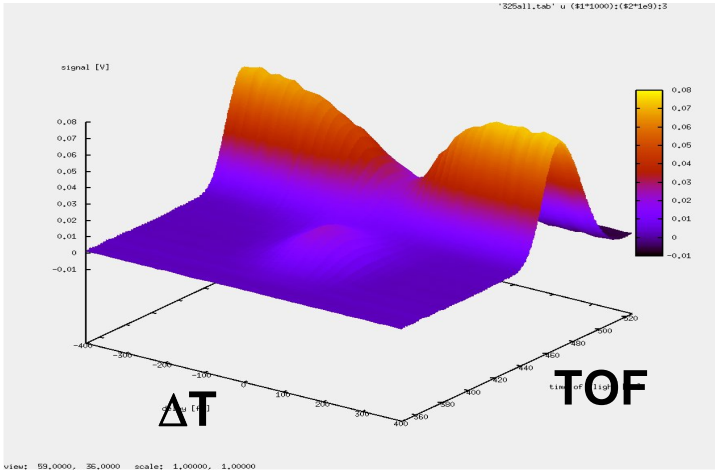
LCLS_2009

X-ray: 950 eV, 2 fs

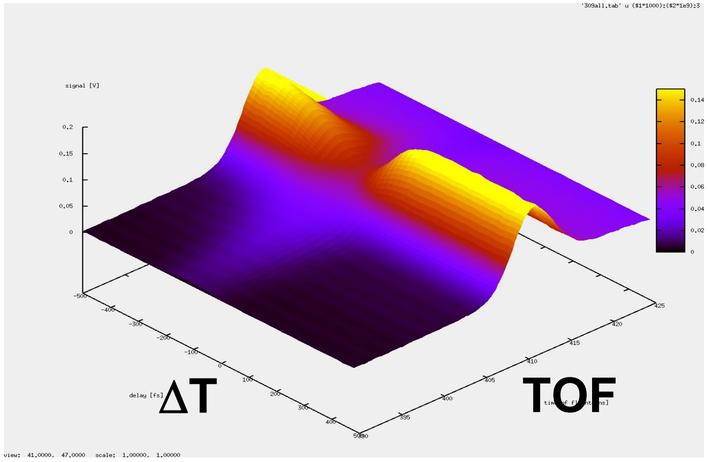
OL : 800nm, 30fs



Ne 1s - photoline



Ne 1s – Auger line



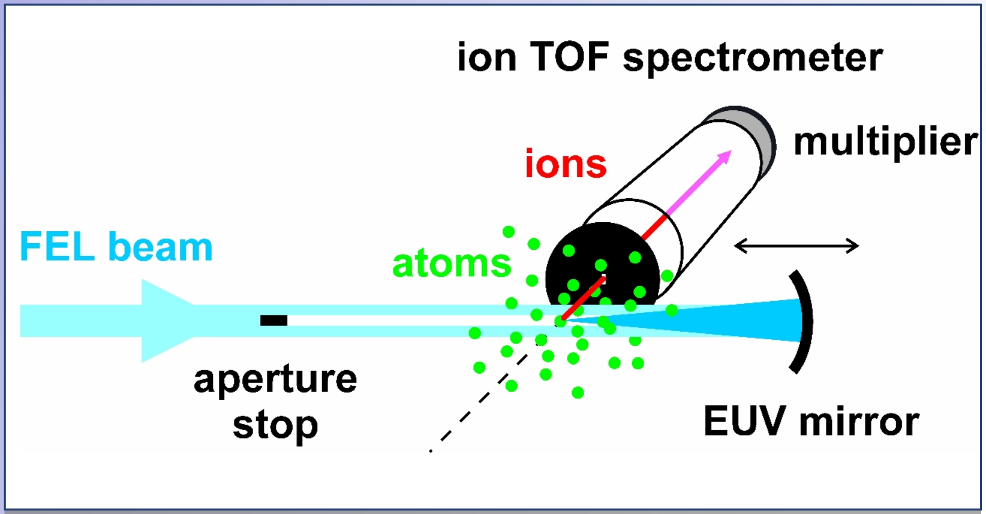
Two-color experiments

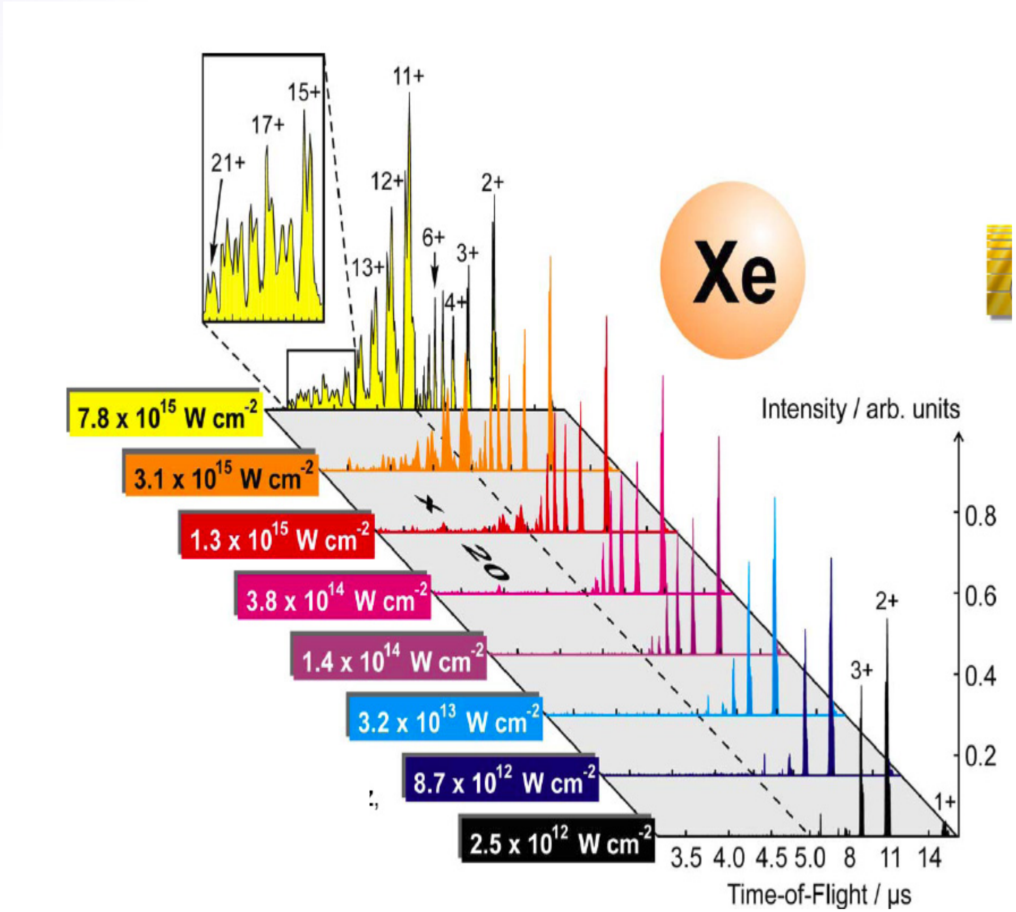
Intense XUV field: Multi-photon Ionization

FLASH: Non-linear Processes

Ion spectroscopy in strong FEL fields

λ(FEL) = 13.3 nm





Sorokin, Bobashev, Feigl, Tiedke, Wabnitz, Richter, Phys. Rev. Lett. 99 213002 (2007)

FEL : 93 eV, focus 2.6 µm

----> 7.8 x 10^{15} W / cm^2

----> Xe^+ …… Xe^{21}

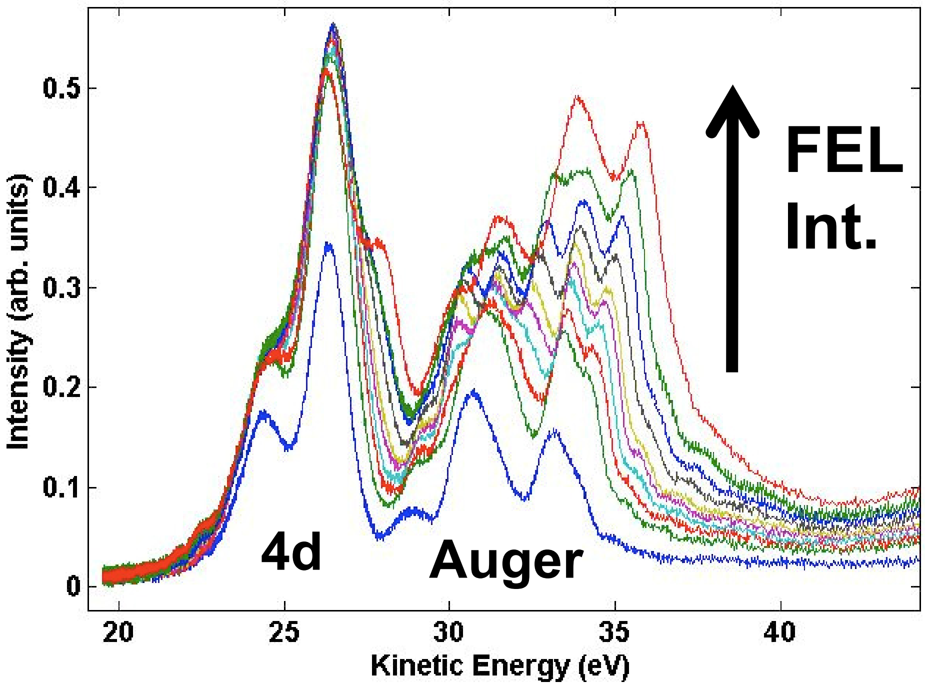
IP(Xe 21+) ≈ 5 keV

Electron Spectroscopy on atomic Xe

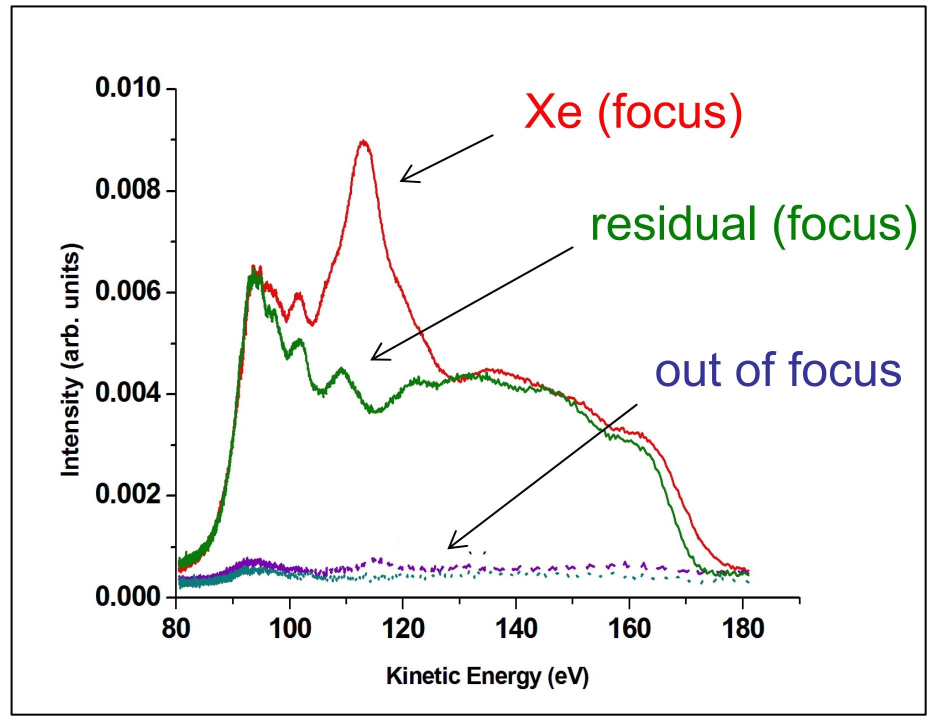
Costello, Düsterer, Meyer, Richter et al.

hν (FEL) 93.3 eV ; □ 10^{15} W / cm^2

One-photon Process



Two-photon Process



Xe^+ $4d^9 5s^2 5p^6$ $^2D_{5/2}$ = 67.5 eV

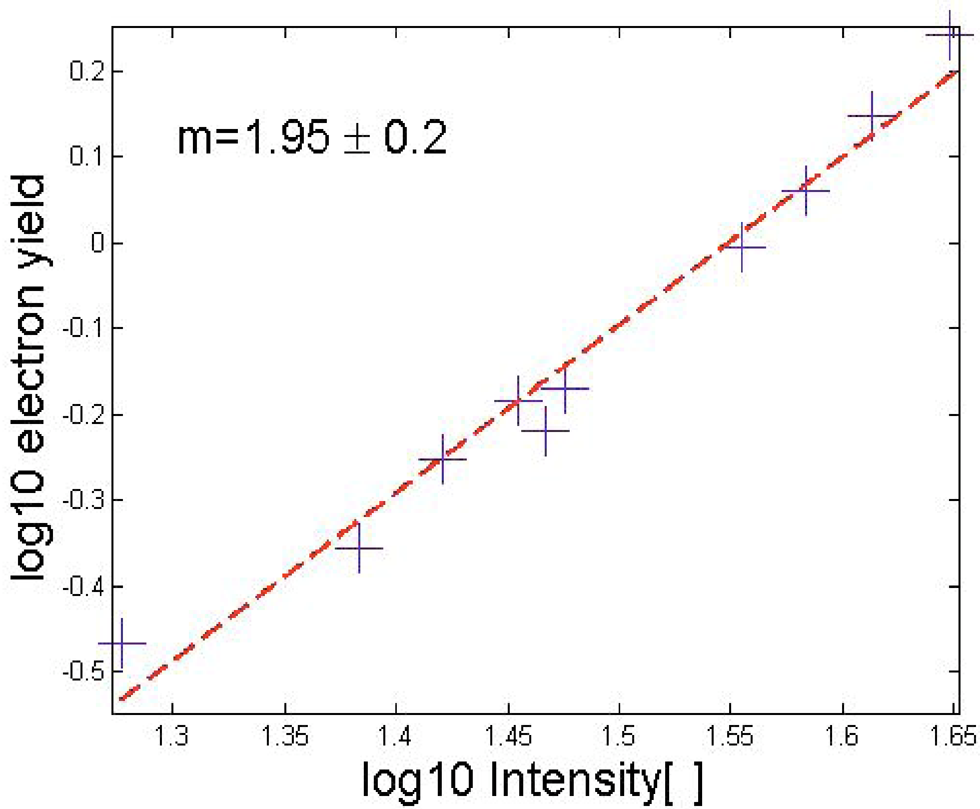
$^2D_{3/2}$ = 69.5 eV

Electron Spectroscopy on atomic Xe

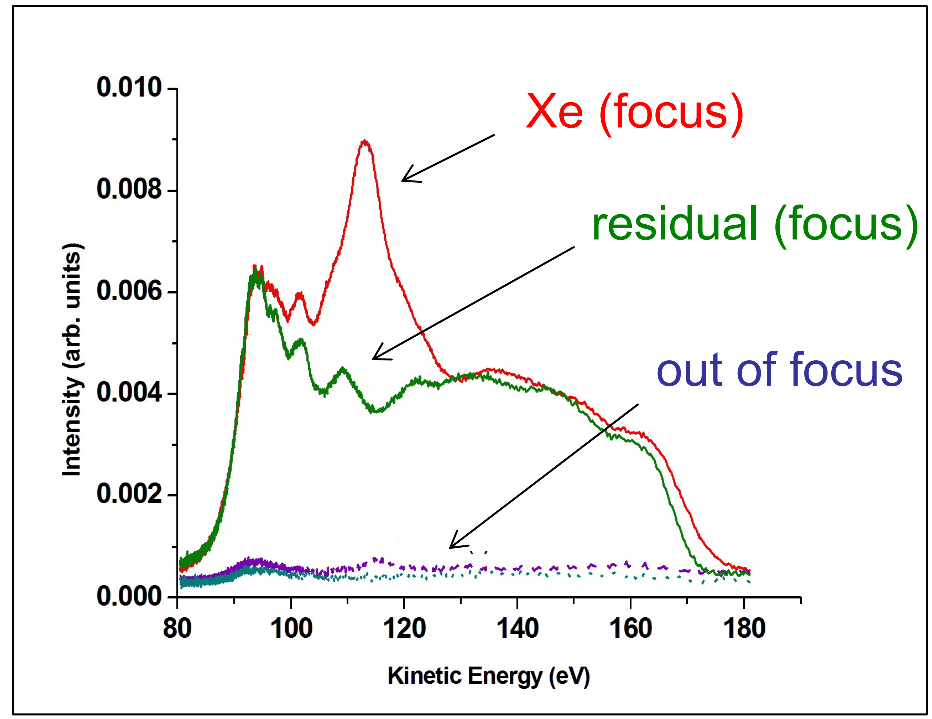
Costello, Düsterer, Meyer, Richter et al.

hν (FEL) 93.3 eV ; □ 10^{15} W / cm^2

Intensity dependence



Two-photon Process



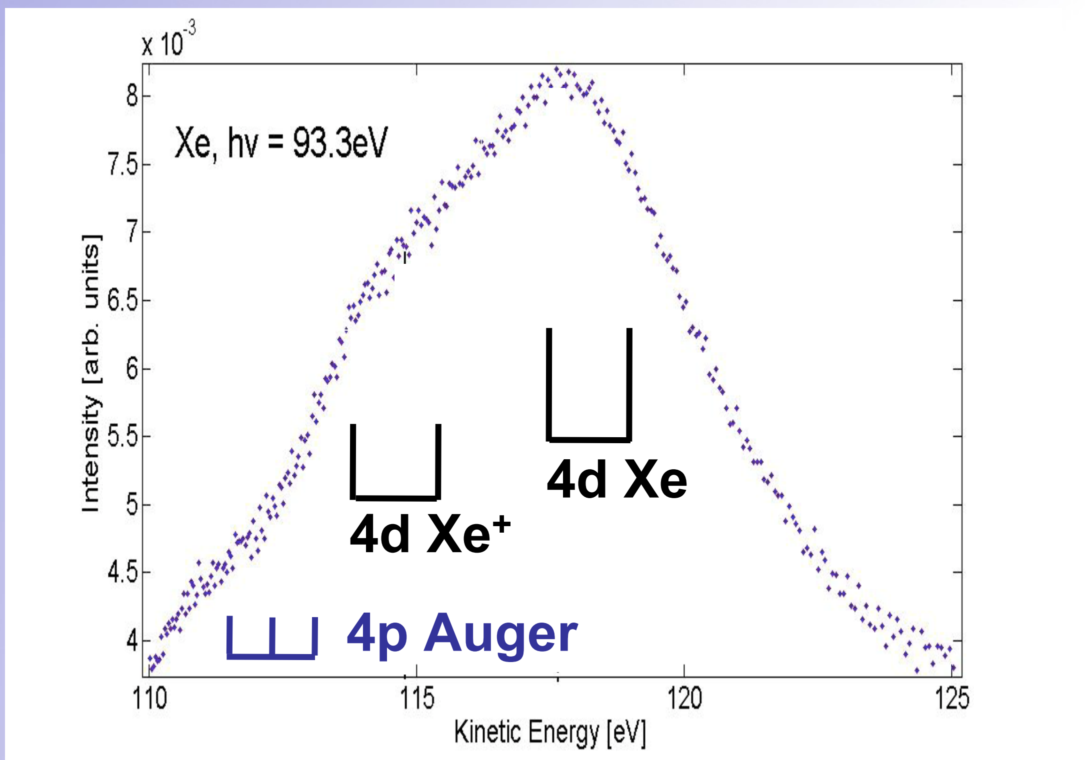
Electron Spectroscopy on atomic Xe

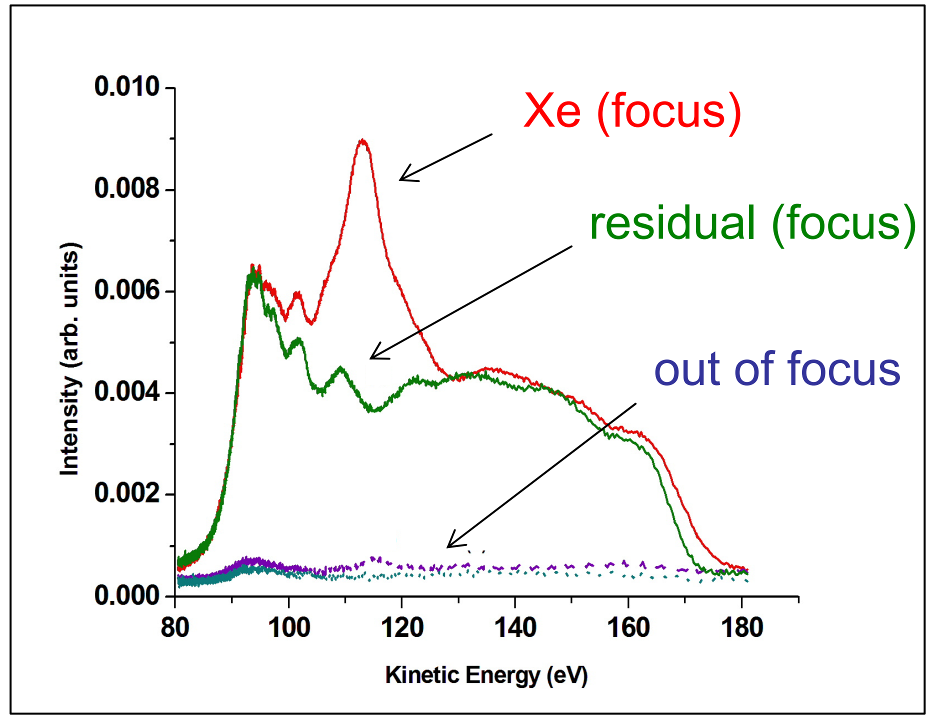
Costello, Düsterer, Meyer, Richter et al.

hν (FEL) 93.3 eV ; □ 10^{15} W / cm²

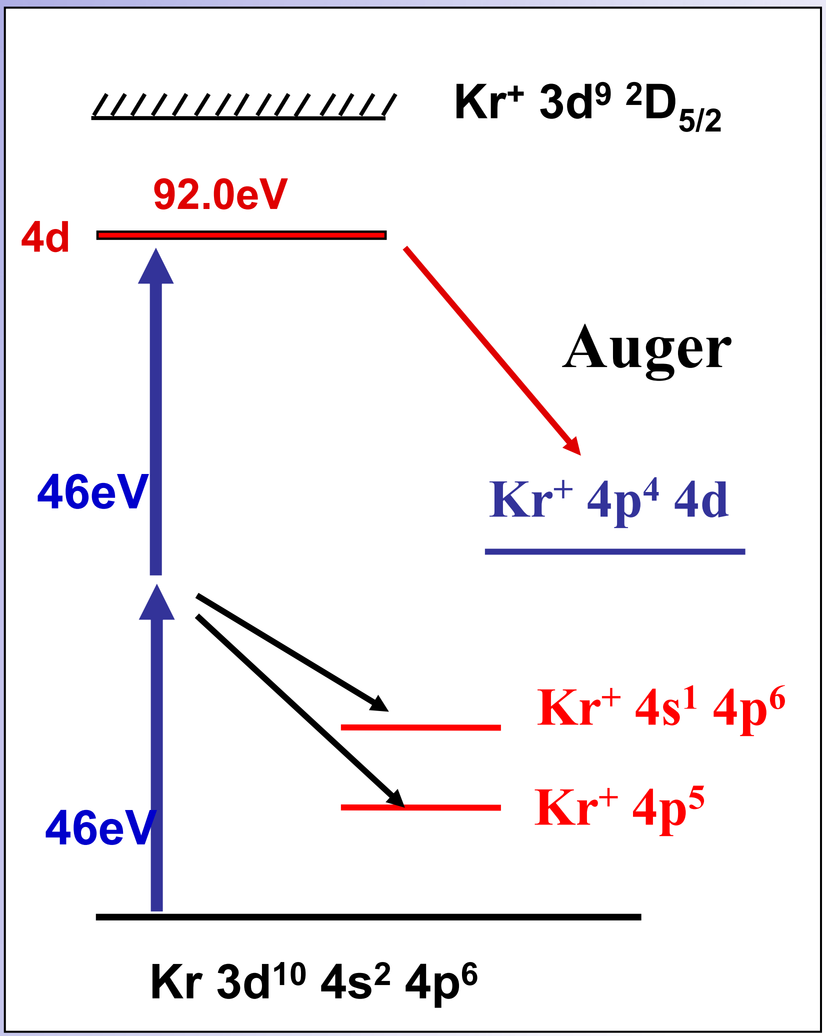
2 x hν (FEL) 93.3 eV

Two-photon Process





Two-photon one-color excitation : Kr* 3d^9 4d

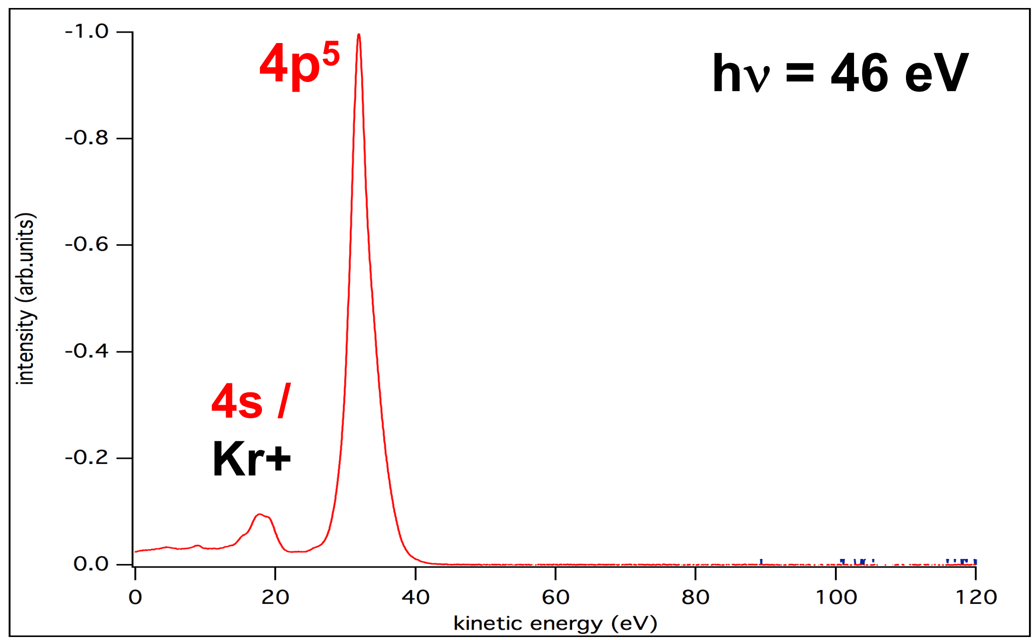


One-photon ionization:

Kr + hν → Kr$^+$ 4p^5 / 4s^1

Two-photon ionization:

Kr + 2 hν → Kr$^+$ 4p^5 / 4p^{4}4d

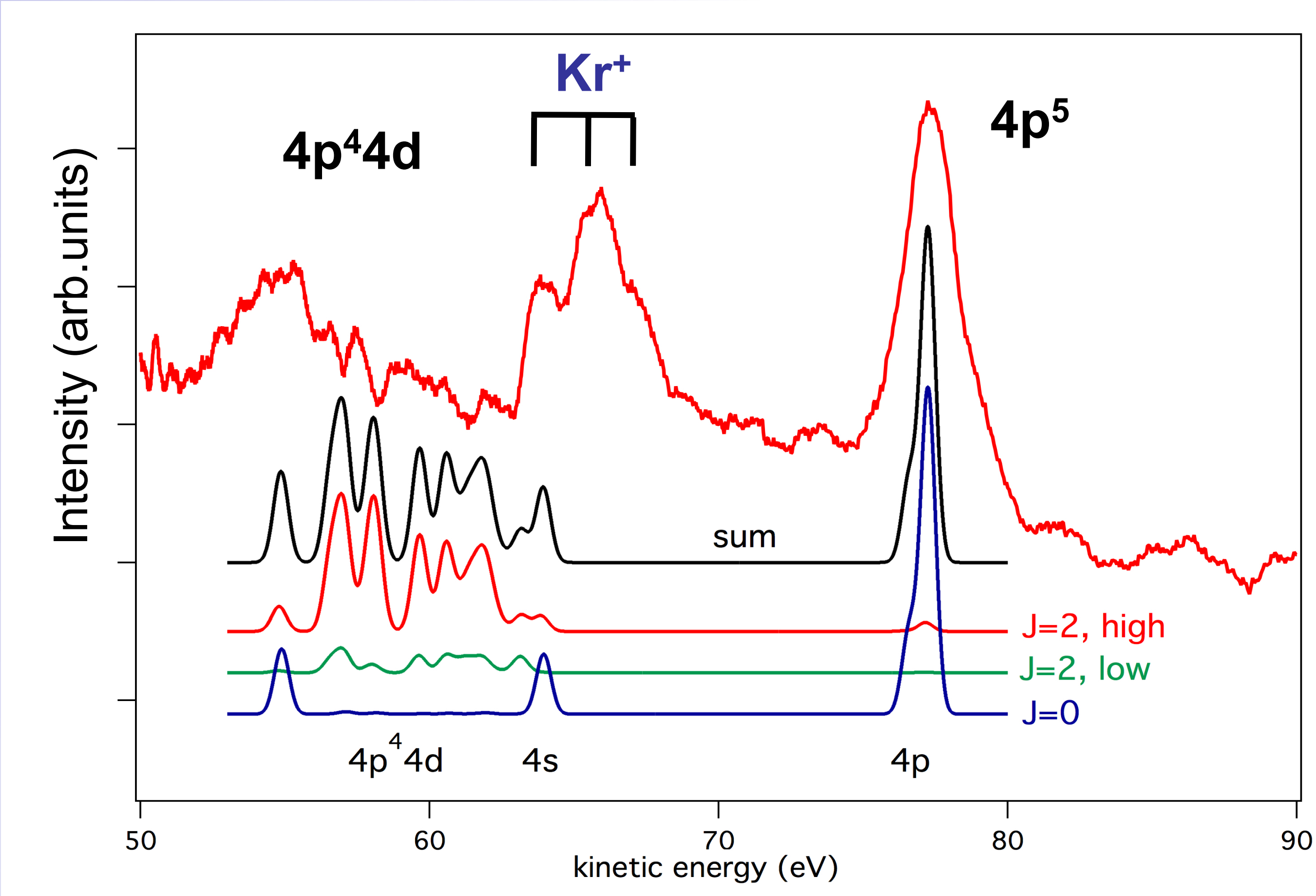


5 μm; >10^{14} W/cm^2

Resonant Auger Decay: Kr* 3d^9 **4d**

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

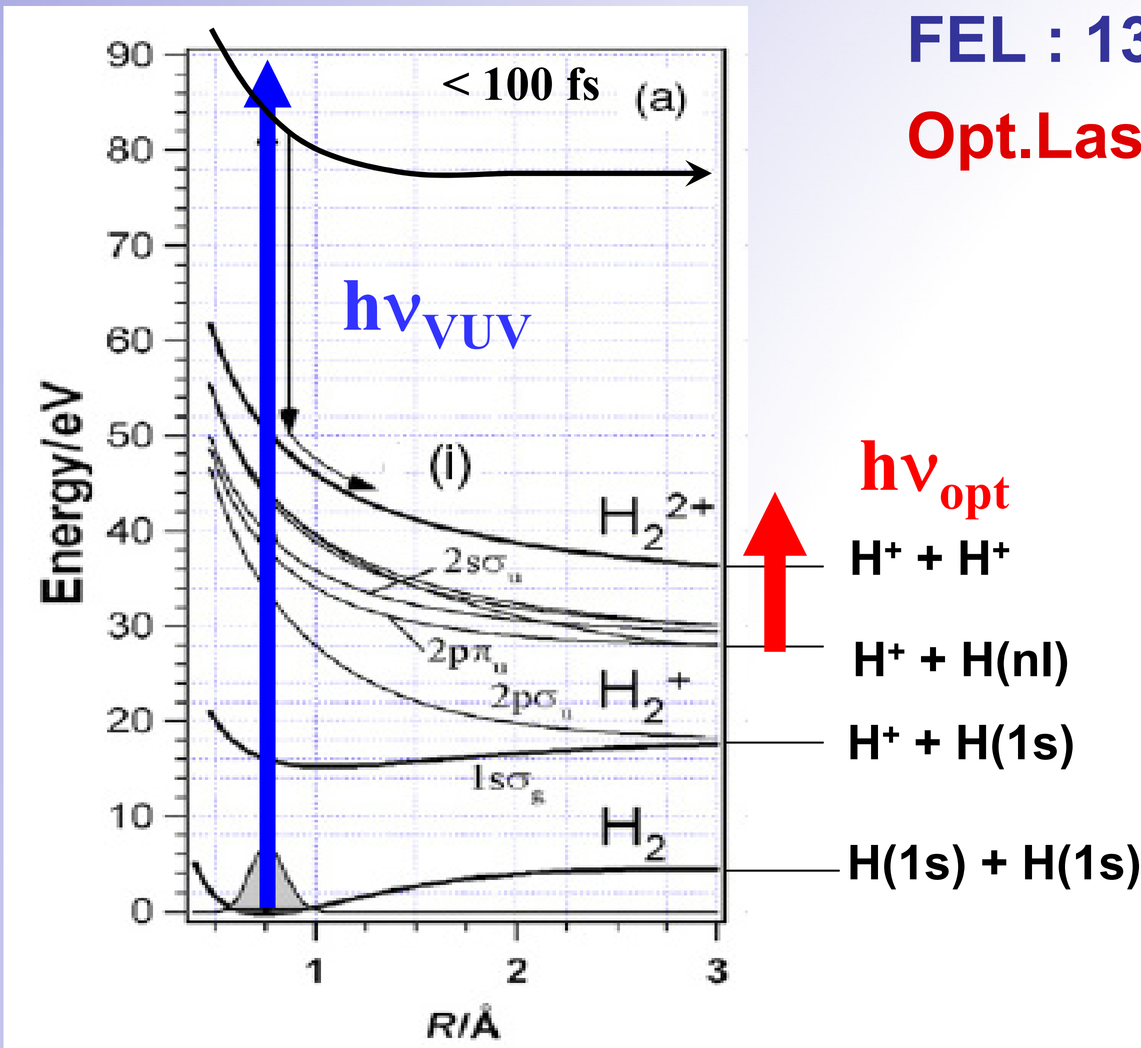
Kr 3d^{9}4s^{2}4p^{6}4d (J=0,2) ---> Kr$^+$ 3d^{10}4s^{2}4p^{4}4d + e$^-$**



Two-color experiments

Dissociation dynamics of H_2

Dissociation Dynamics in H_2



FEL : 13.7 nm, 90.5 eV

Opt.Las. : 800 nm, 1.55 eV

400 nm, 3.1 eV

H*(n=2) : E(bind) = 3.4 eV

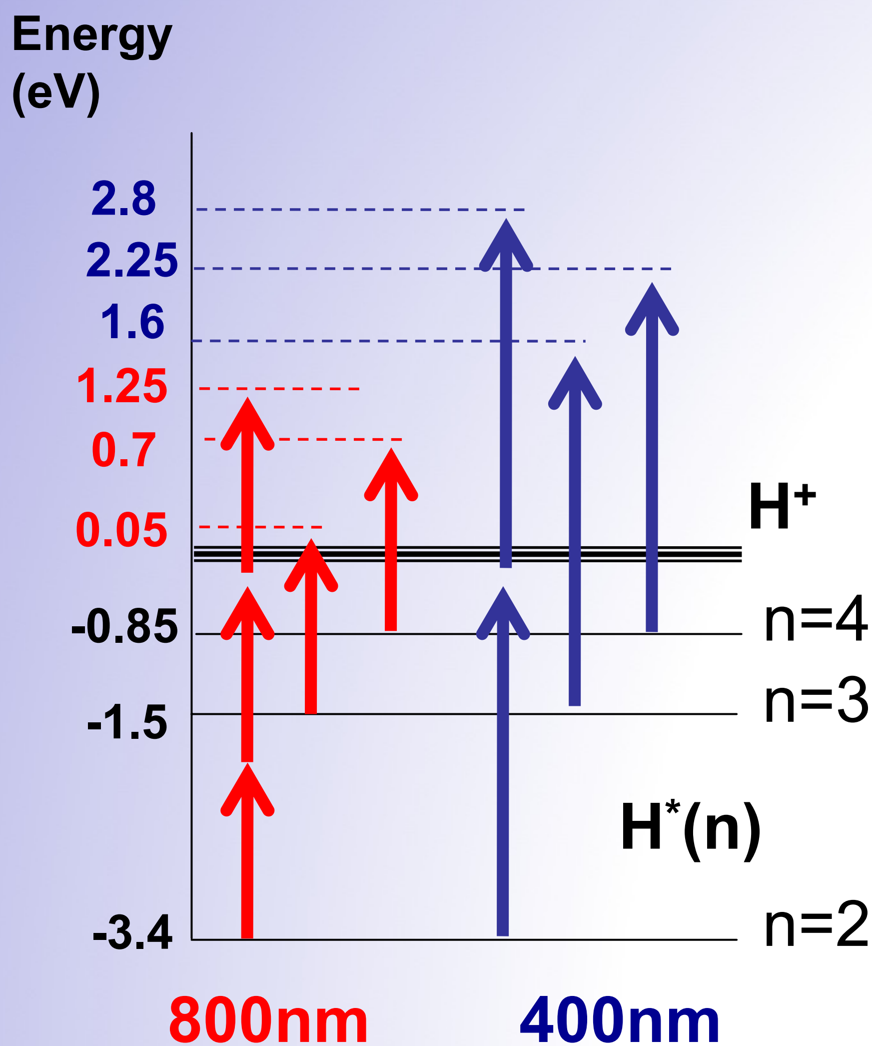
H*(n=3) : E(bind) = 1.5 eV

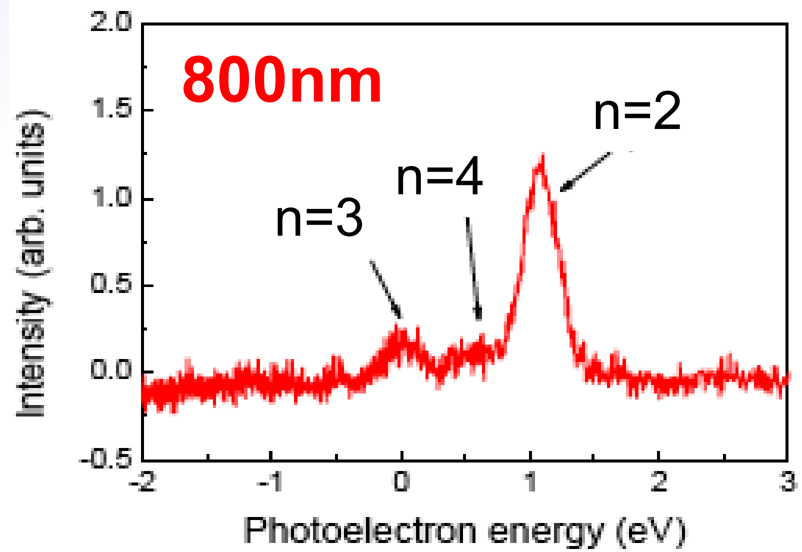
H*(n=4) : E(bind) = 0.8 eV

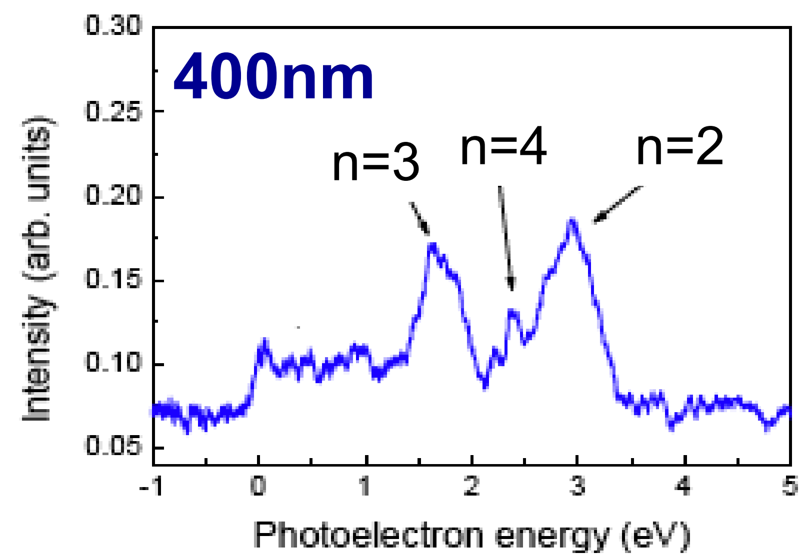
⋮

E (kin.) < 1.5 eV

Photoionization of excited atomic fragments

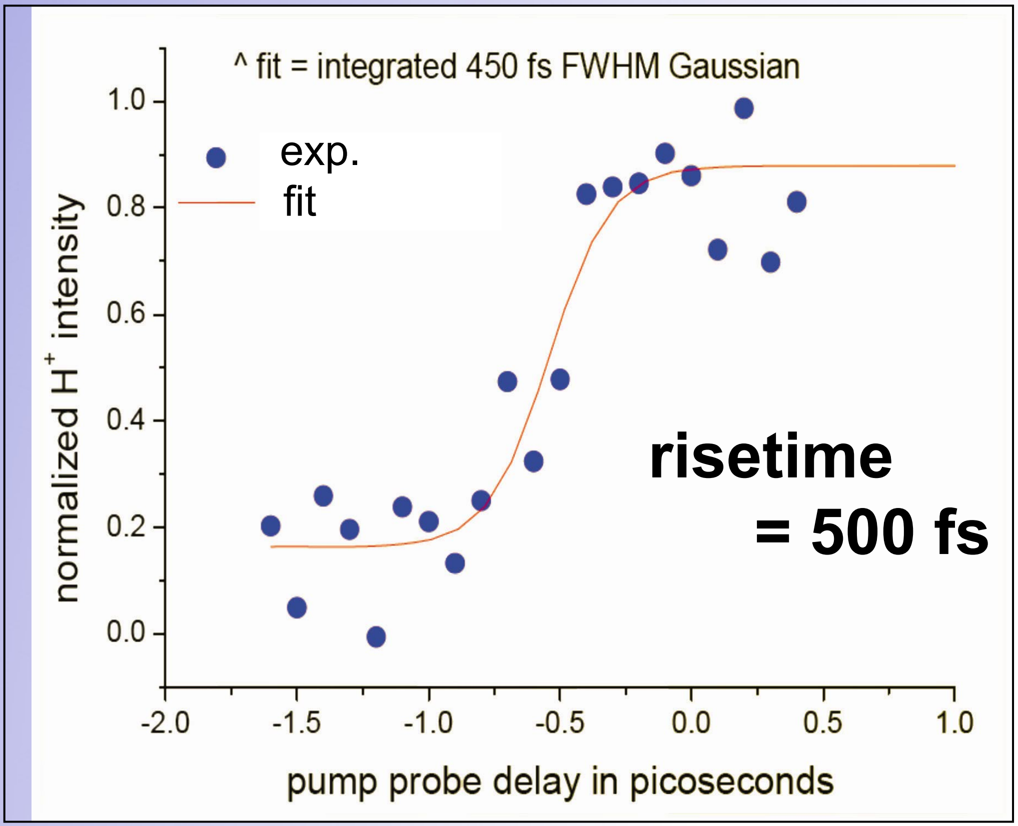




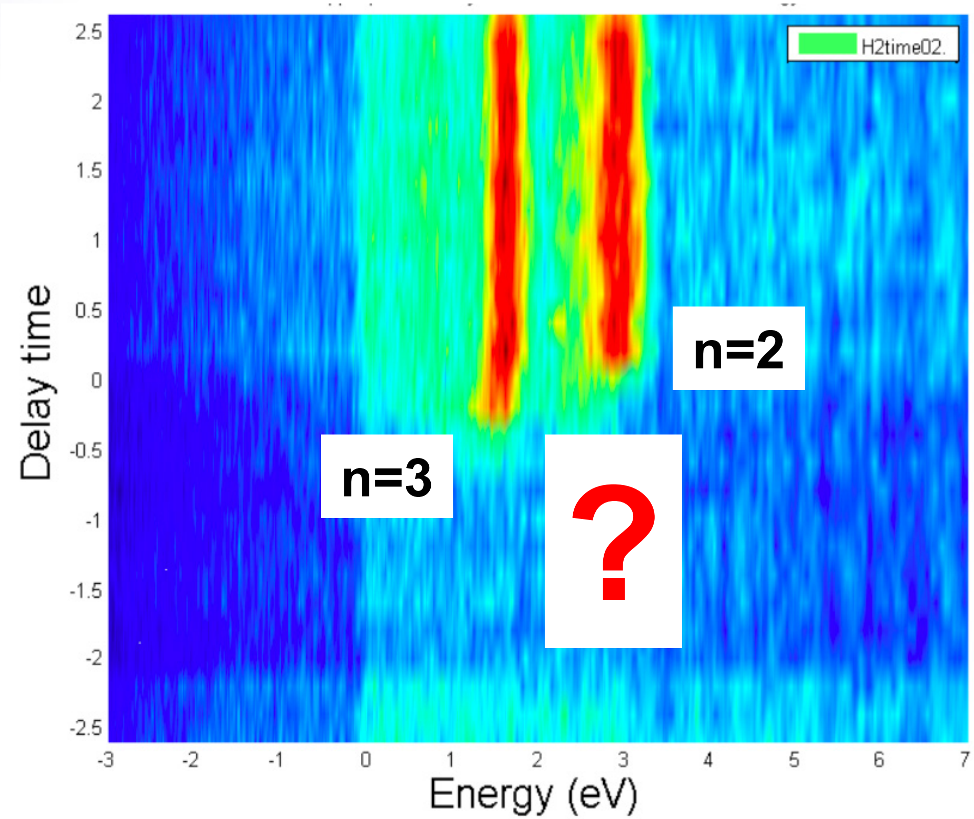


Ultra-fast molecular dissociation

800 nm Laser



400 nm Laser



Fast fragmentation < 100 fs

Summary

- **Above threshold ionization (ATI) of rare gases**
    - **Beyond Soft-Photon Approximation**
- **Non-linear (multi-photon) processes**
    - **Auger dynamics in dressed atoms (2-colour)**
    - **Ionization mechanisms (1-colour)**
- **Resonant two-photon excitation**
    - **1-colour vs. 2-colour**
- **Molecular dissociation dynamics**
    - **Excitation of core resonances**

FLASH → LCLS → XFEL

Atomic Photoionization Dynamics in Intense Radiation Fields

Experiment

- **LIXAM (Orsay, France)**
  D. Cubaynes, M. Meyer

- **DESY (Hamburg, Germany)**
  S. Düsterer, W.-B. Li, A. Azima,
  P. Radcliffe, H. Redlin, J. Feldhaus

- **Dublin City University (Dublin, Ireland)**
  J. Dardis, P. Hayden, P. Hough, M. Kelly,
  V. Richardson, E.T. Kennedy, J.T. Costello

Theory

- **LCP-MR (Paris, France)**
  R. Taïeb, A. Maquet

- **State University Moscow (Russia)**
  E.V. Gryzlova, S.I. Strakhova,
  A.N. Grum-Grzhimailo

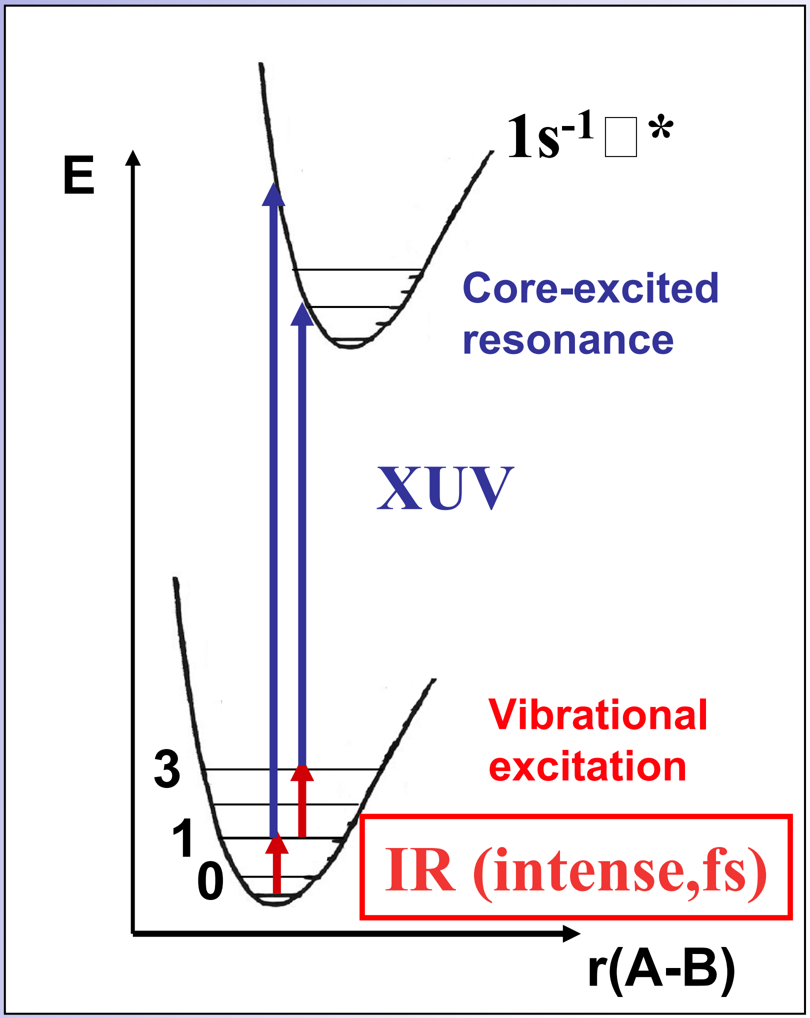
- **FORTH (Heraklion, Crete)**
  P. Lambropoulos

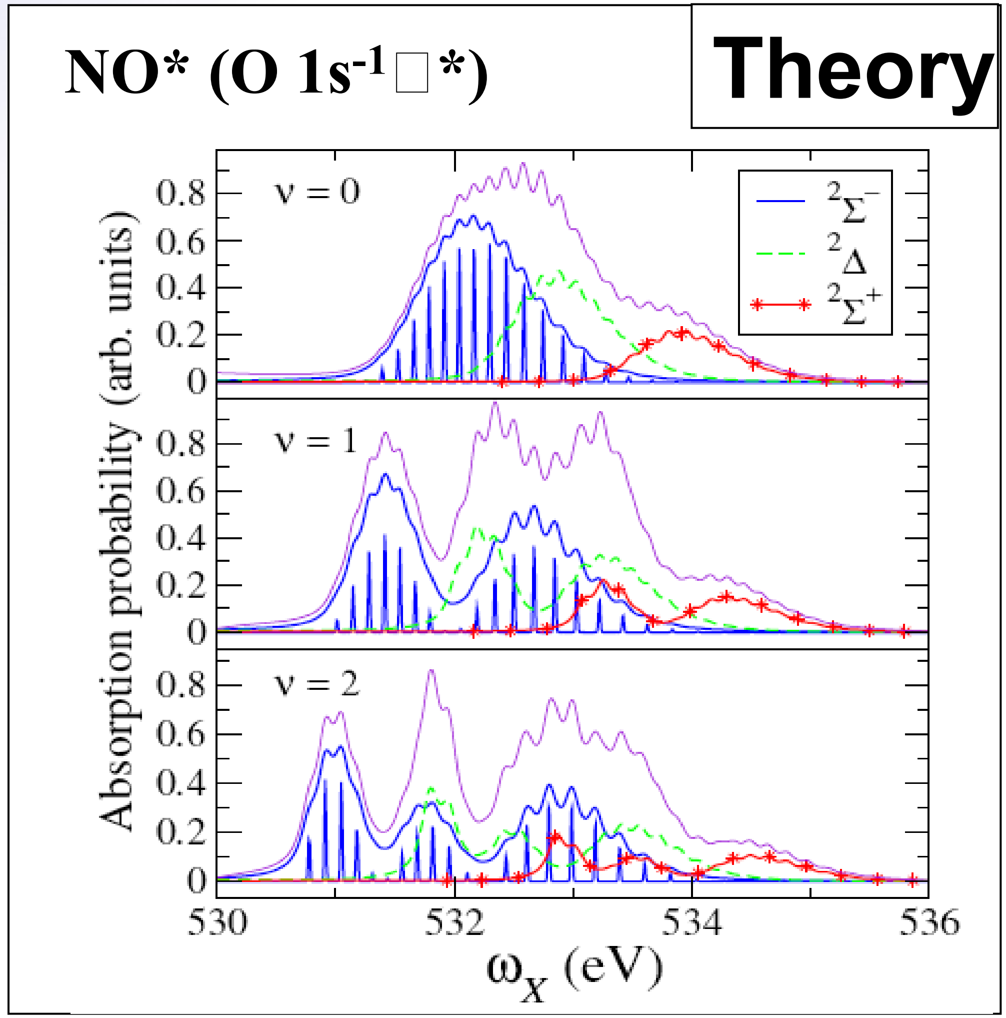
- **Jozef Stefan Institute (Ljubljana, Slov.)**
  A. Mihelic

- **GSI (Darmstadt, Ger.) / Univ. Oulu (Finl.)**
  S. Fritzsche

Photoexcitation of excited molecules

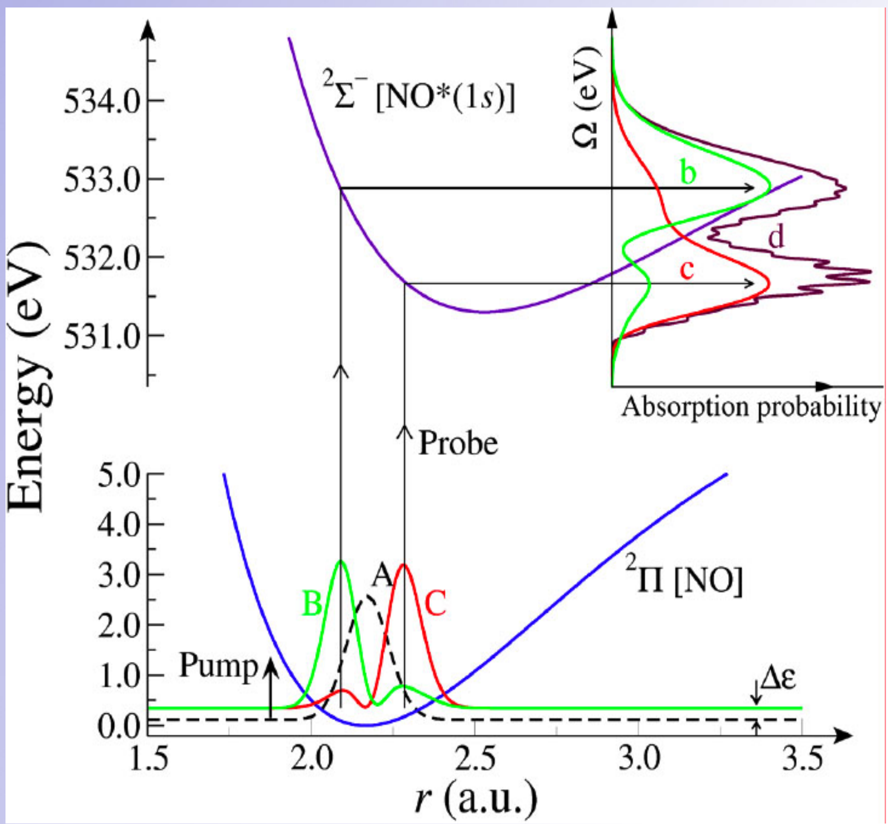
F. Guimaraes, F. Gel'mukhanov, H. Agren et al. (KTH, Stockholm)

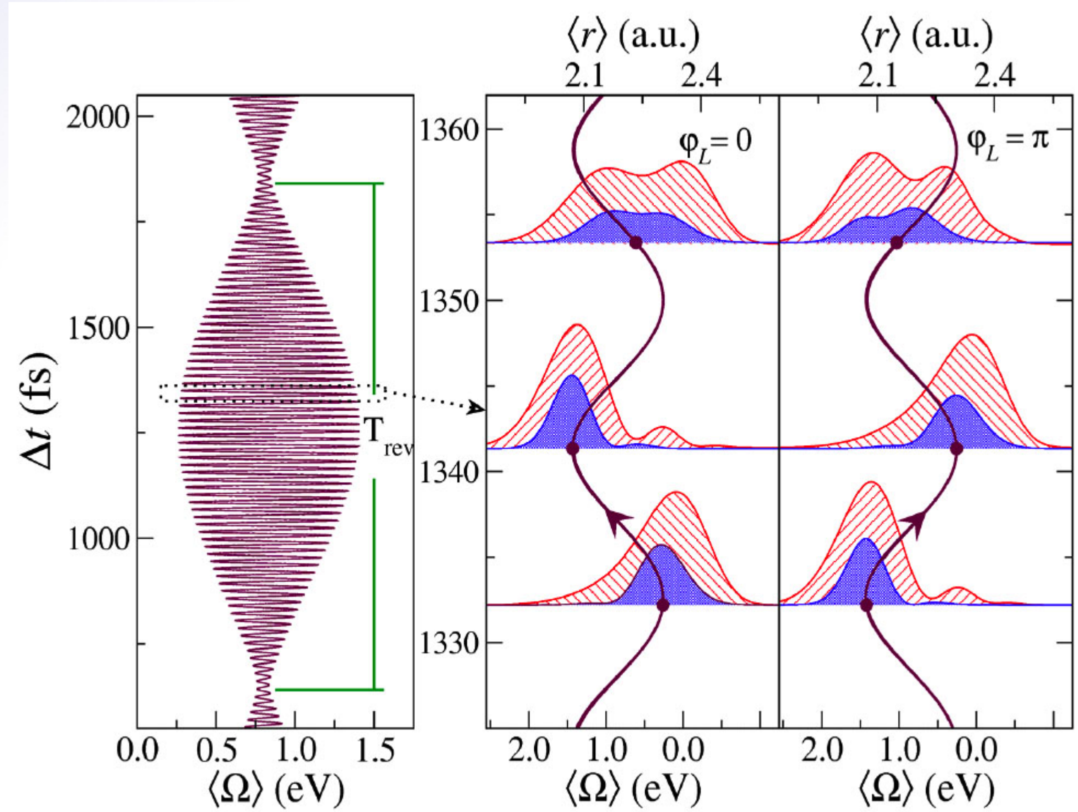




Vibrational wavepackets

F. Guimaraes, F.Gel'mukhanov et al. Phys. Rev. A72, 12714 (2005)





Pump: laser **Probe: FEL**

OL: 2 x 10^{12} W/cm^2 , 100fs
X-ray: 5 fs

Two-Color Pump-Probe Studies

FEL

Atoms

Molecules

Opt. laser

VUV – XUV

- photoionization
- core electrons

Short fs pulses

Intense pulses

Model systems

- electron correlation
- electron/nuclear dynamics

Basic processes

Fundamental laws

UV/Vis/NIR

400-800nm

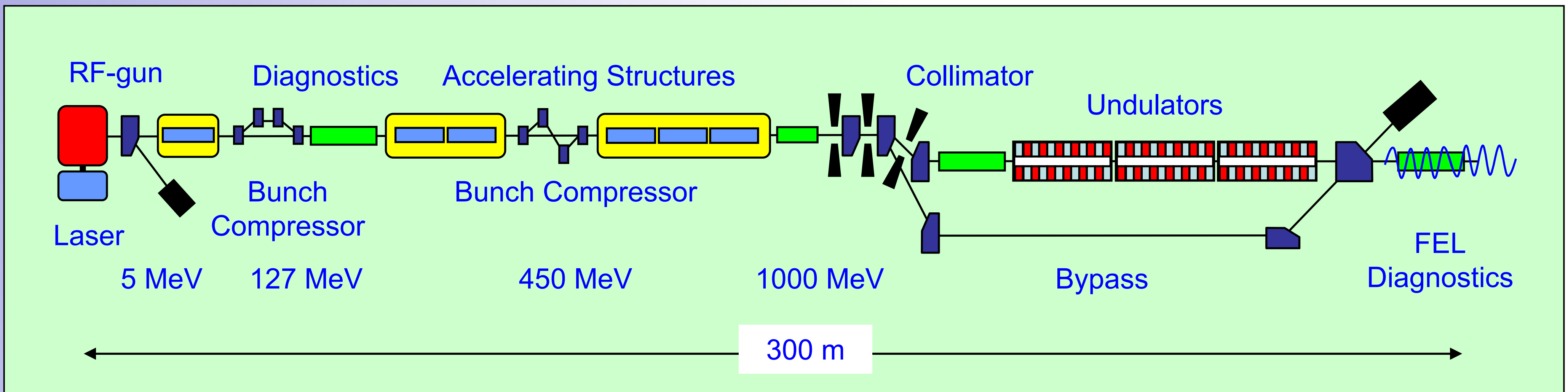
< 100 fs

1-5 mJ

$< 10^{15}$ W/cm^2

FLASH (Free electron LASer in Hamburg)

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

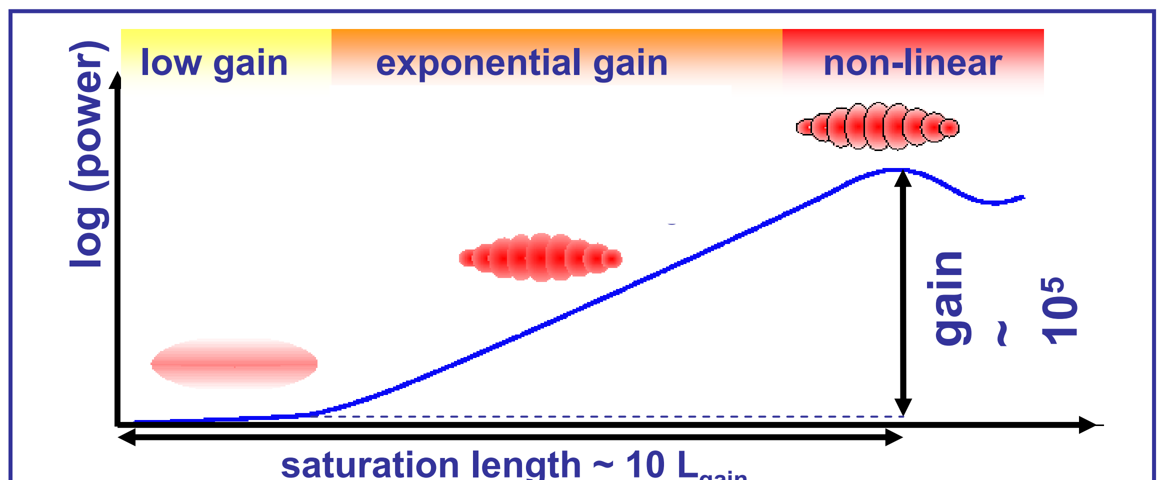


LINAC : 1 GeV

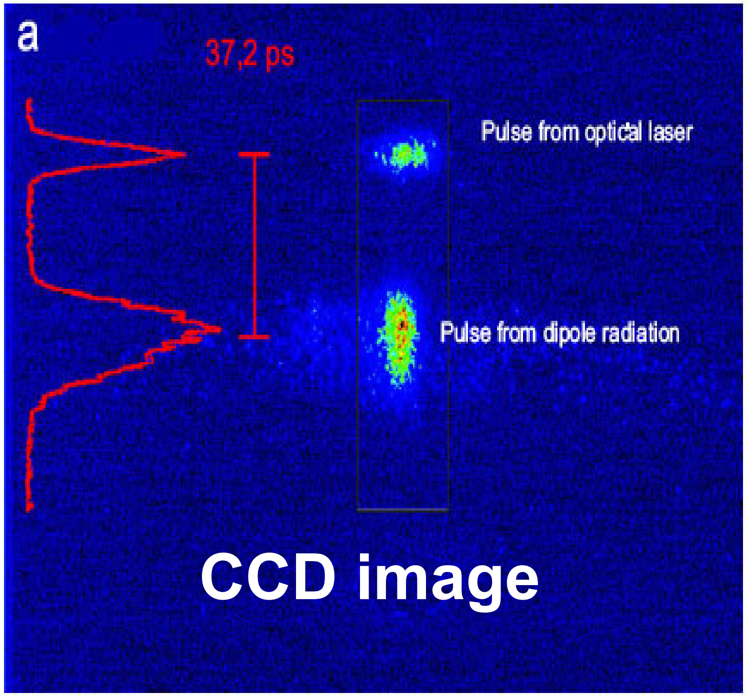
30 m fixed-gap undulator

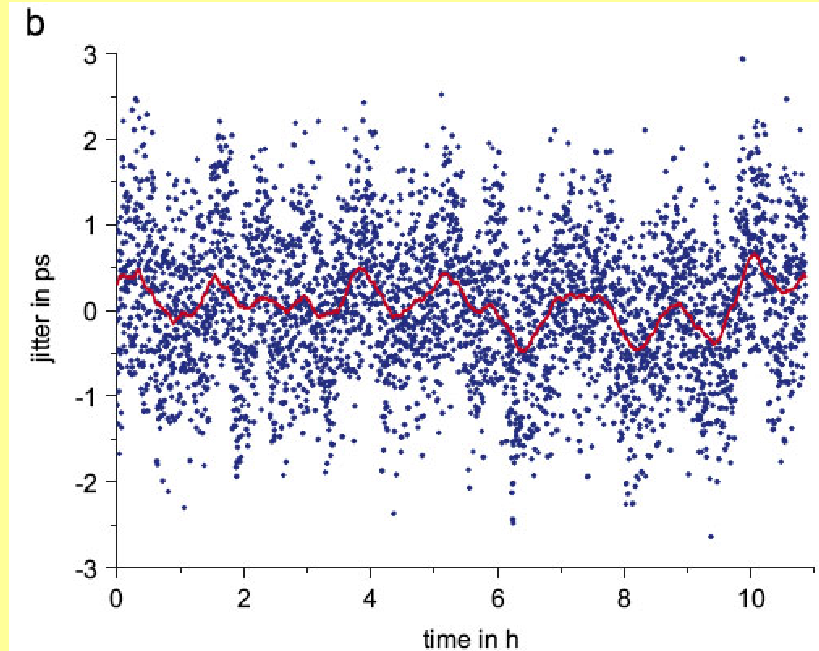
macro-bunch at 10 Hz

> 30 bunches, 1 µs separation



FLASH + Optical Laser





Long term stability : 800 fs r.m.s

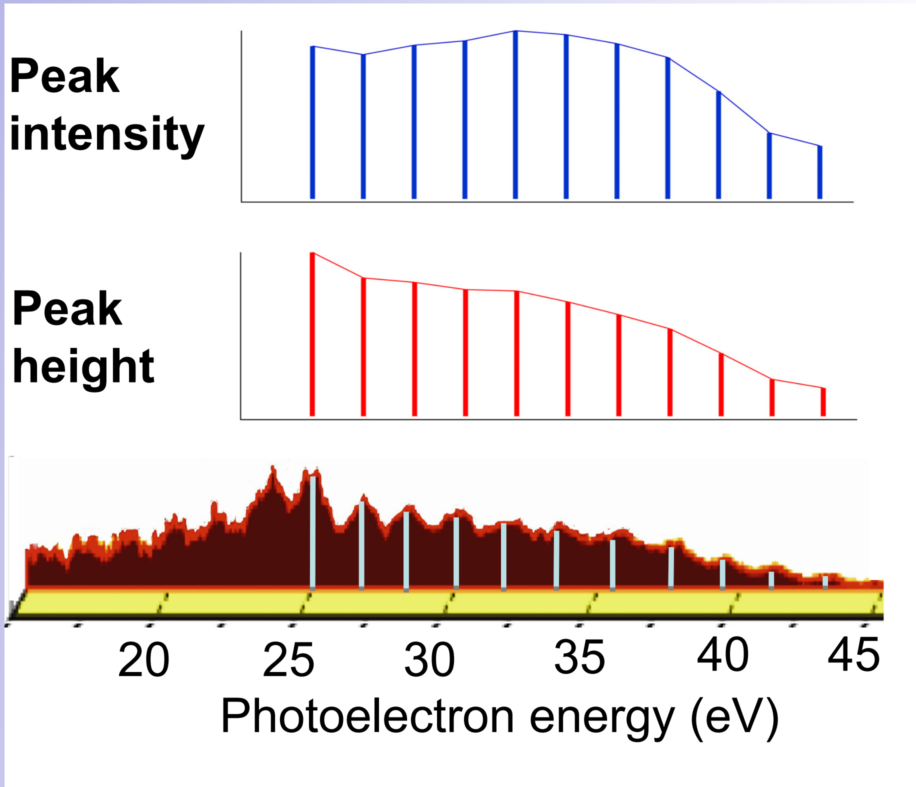
TTF clock
from injector rack
⇨ 300 m long cables

synchroscan streak camera
⇨ slow feedback
goal: drift < ps / h

Max-Born-Institut, Berlin
I. Will, H. Redlin

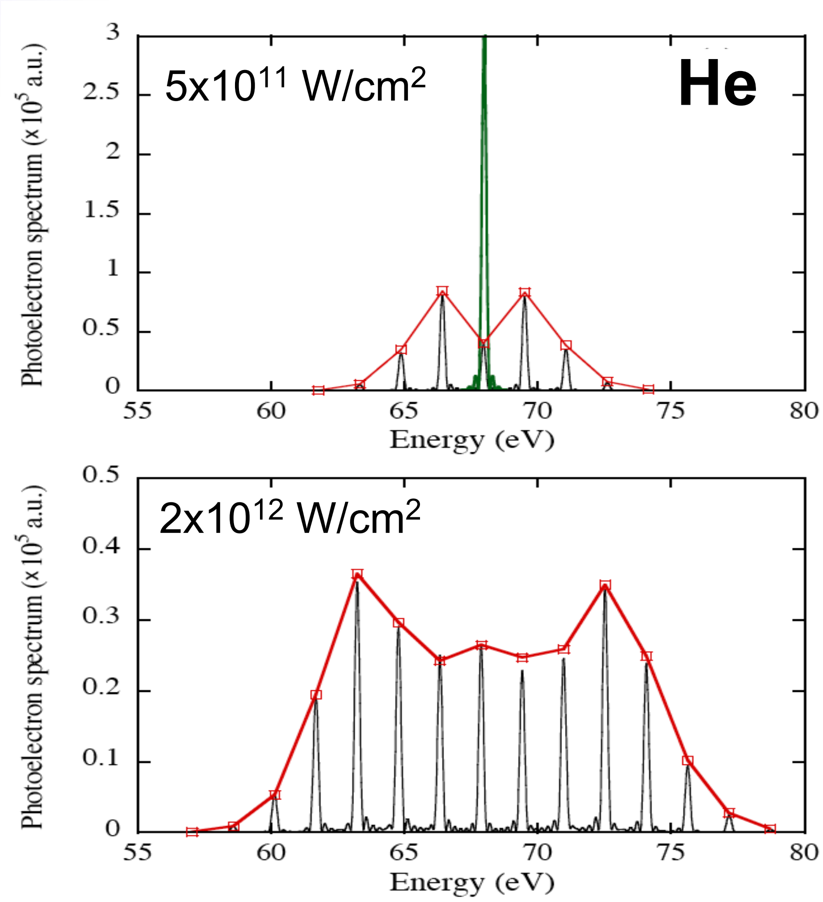
ATI : Strong NIR Dressing Field (Ne)

FEL: 26.9nm (46 eV)
Laser: 800nm, 1.8mJ, 100fs
$3x10^{13}$ W/cm²



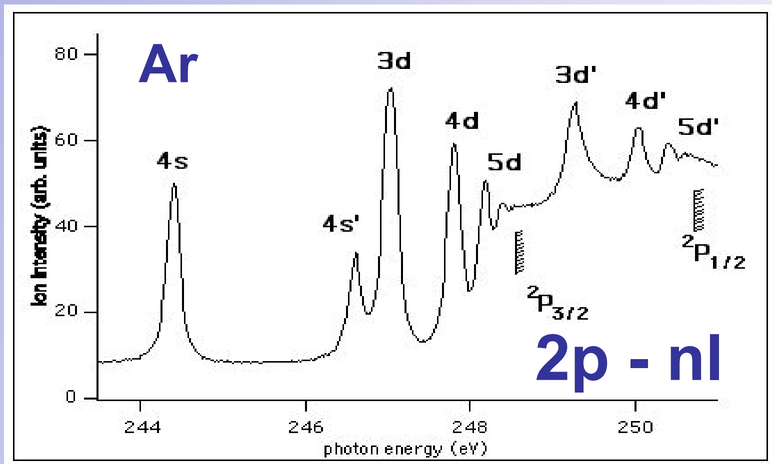
Multi-photon processes

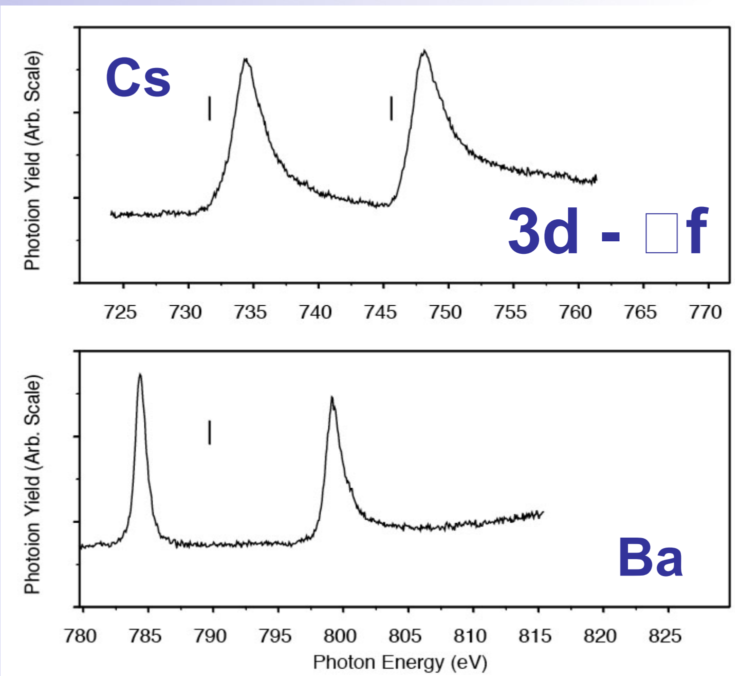
"Soft-Photon Approximation"

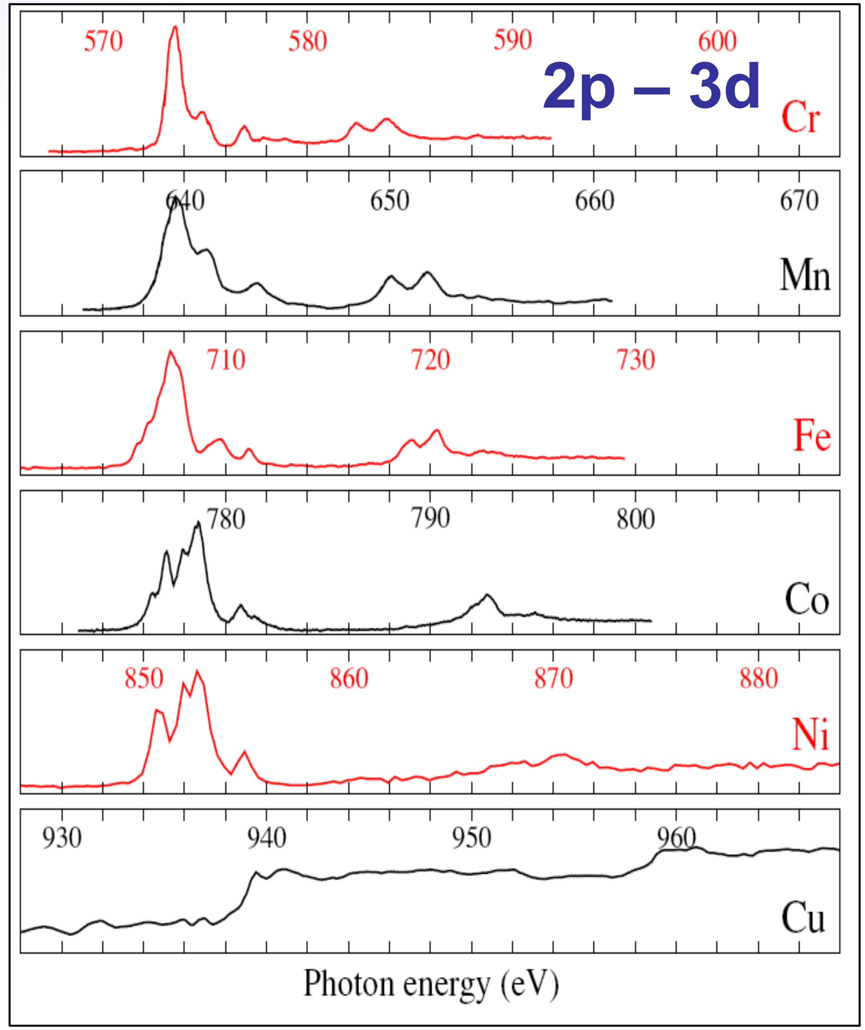


A. Maquet, R. Taieb, J. Mod. Opt. 54, 1847 (2007)

XFEL : Dynamics of Innershell Resonances



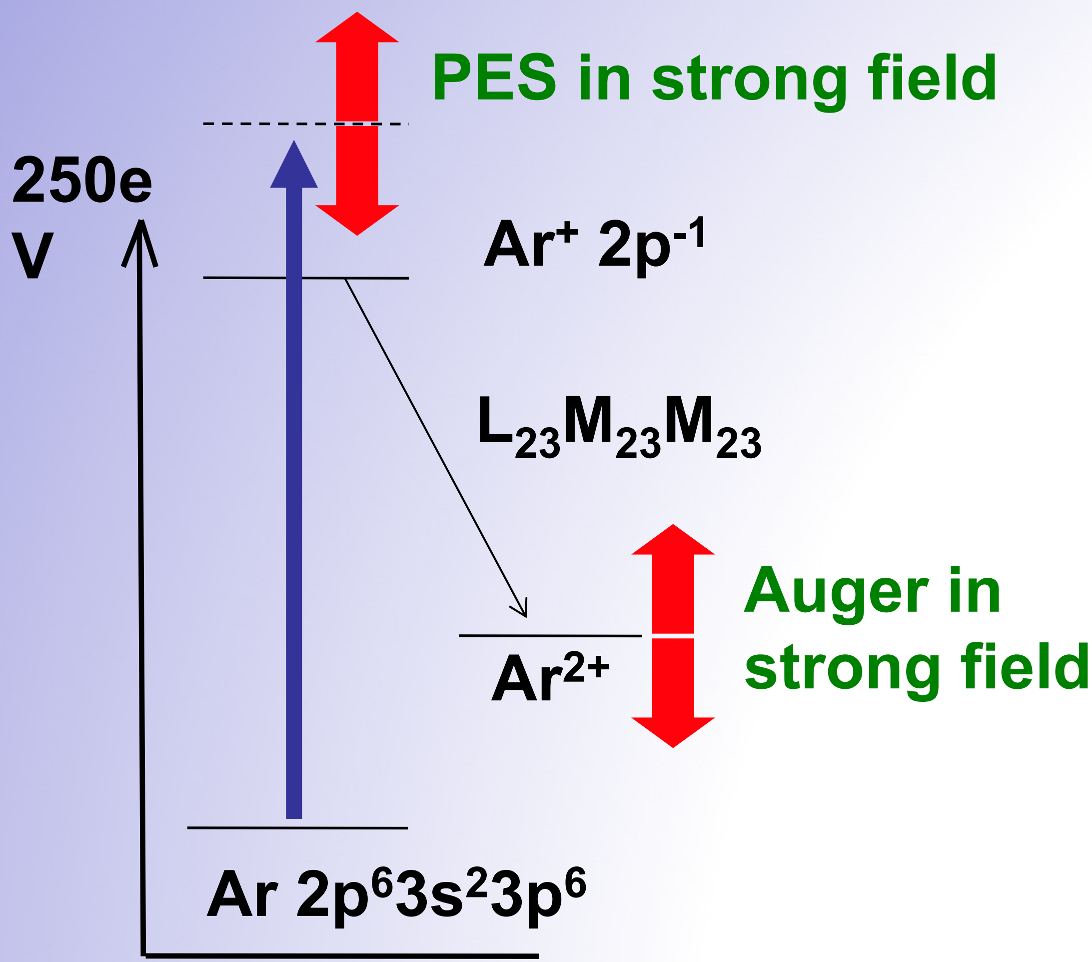




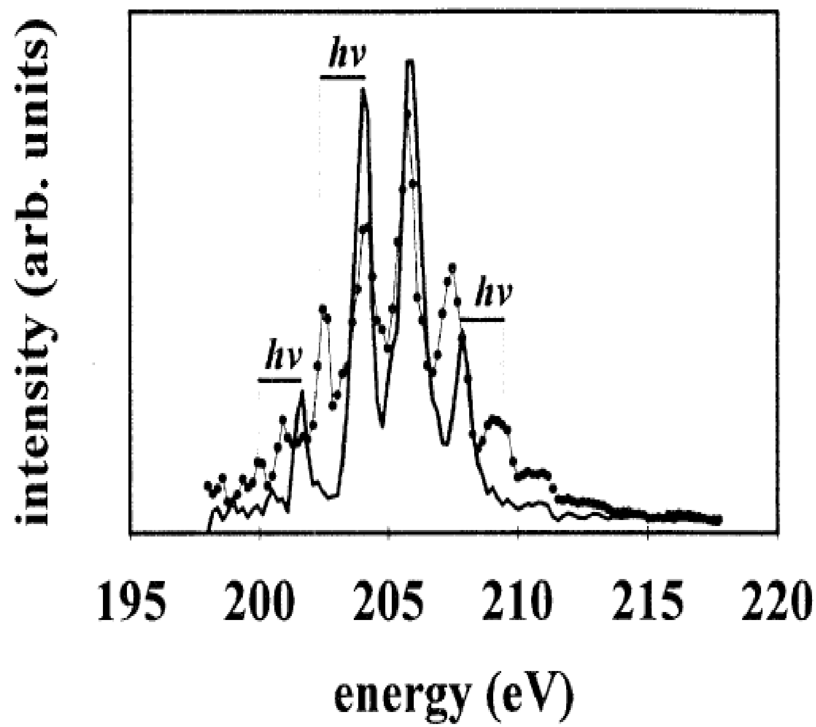
Arp et al., JPB32 (1999)

M. Martins et al. JPB 39, 2006

Next: Laser-assisted Auger decay

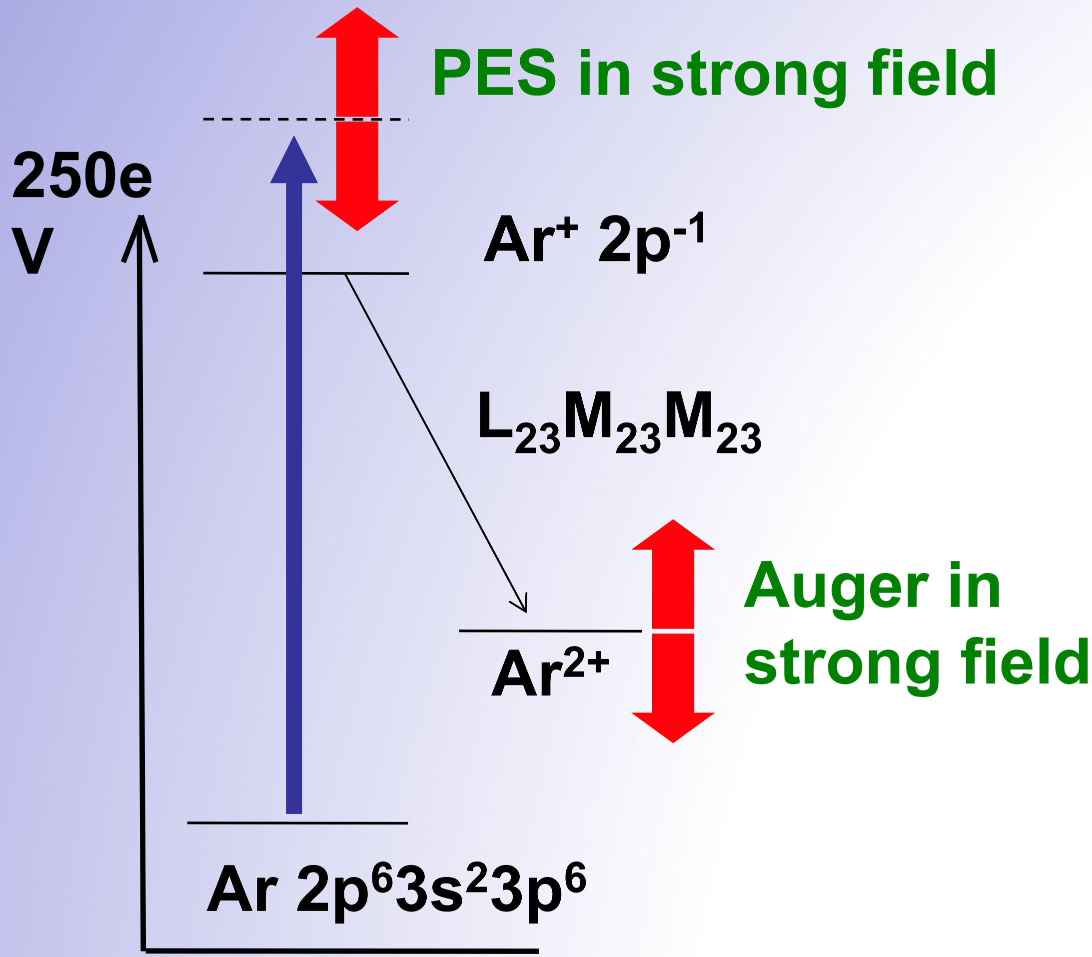


Schins et al. PRA 52, 1272 (1995)



Nov. 2009 LCLS : Ne 1s at hν > 870 eV

Next: Laser-assisted Auger decay



Dynamics in strong fields

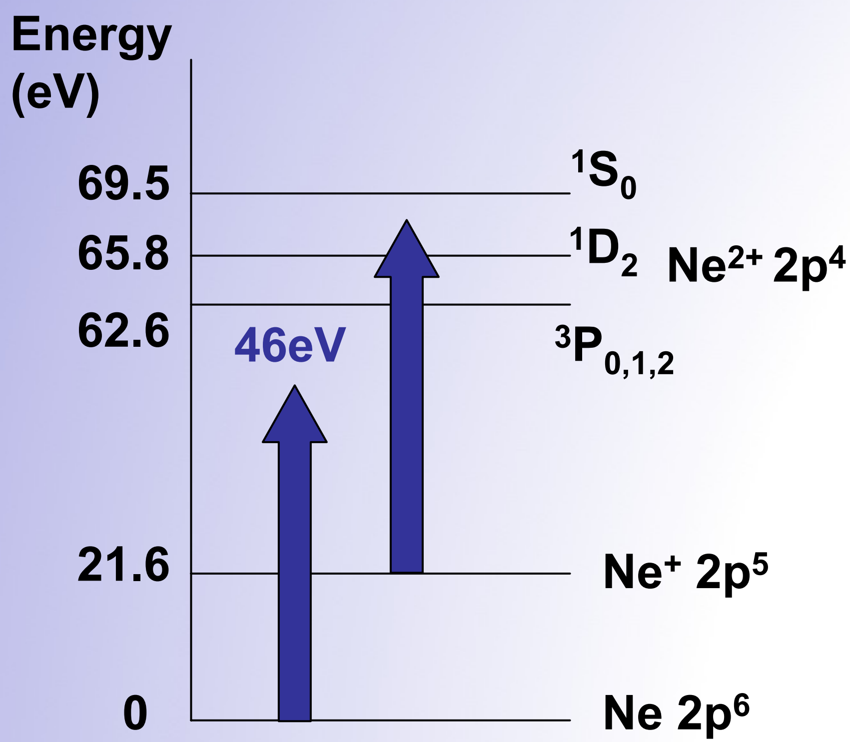
- angular distribution
- $\Delta T(XUV) \approx \Gamma(Nat)$
- resonant Auger
- coupling of ionic states
- control of decay

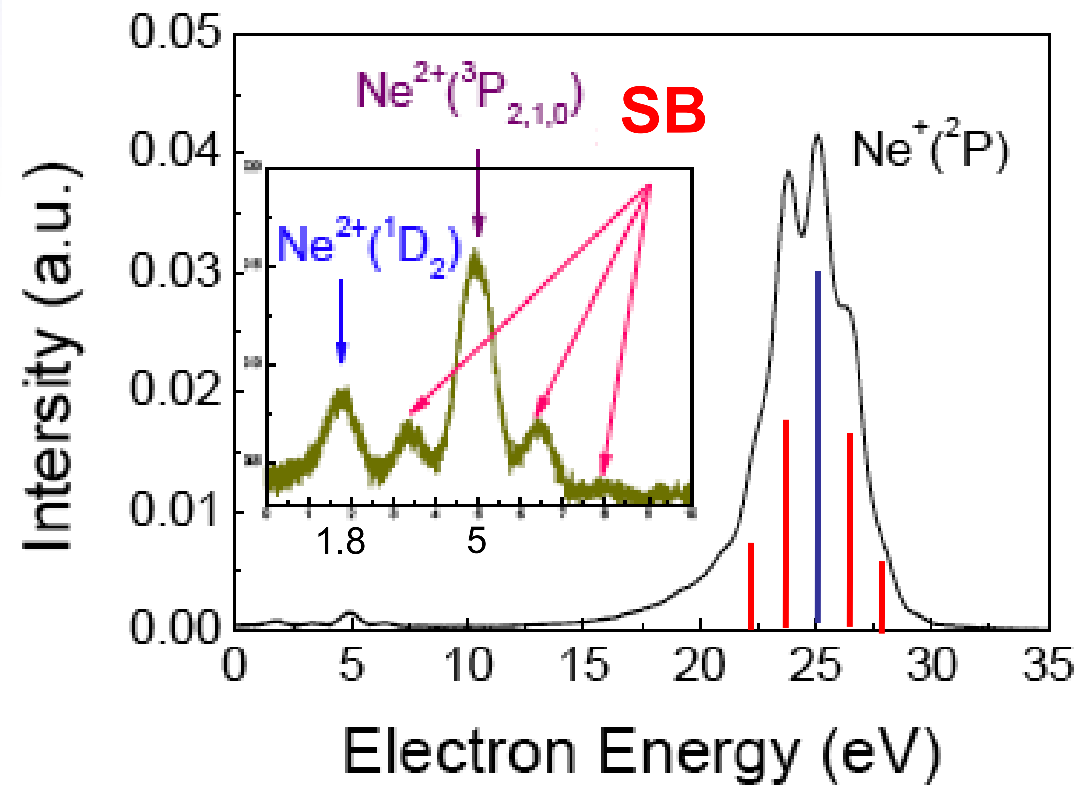
Nov. 2009 LCLS : Ne 1s at $h\nu > 870$ eV

Strong XUV + NIR Fields

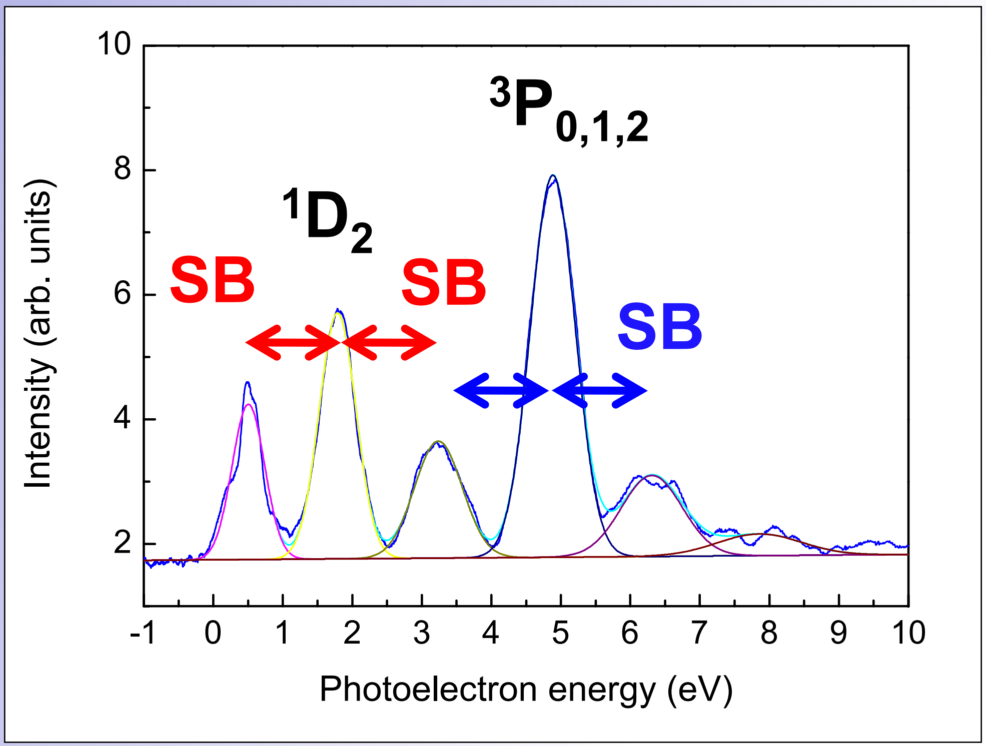
Sequential Two-Photon Double-Ionization

FEL: 46 eV, 20 μJ, 30fs
OL: 800nm, 1.8mJ, 3ps



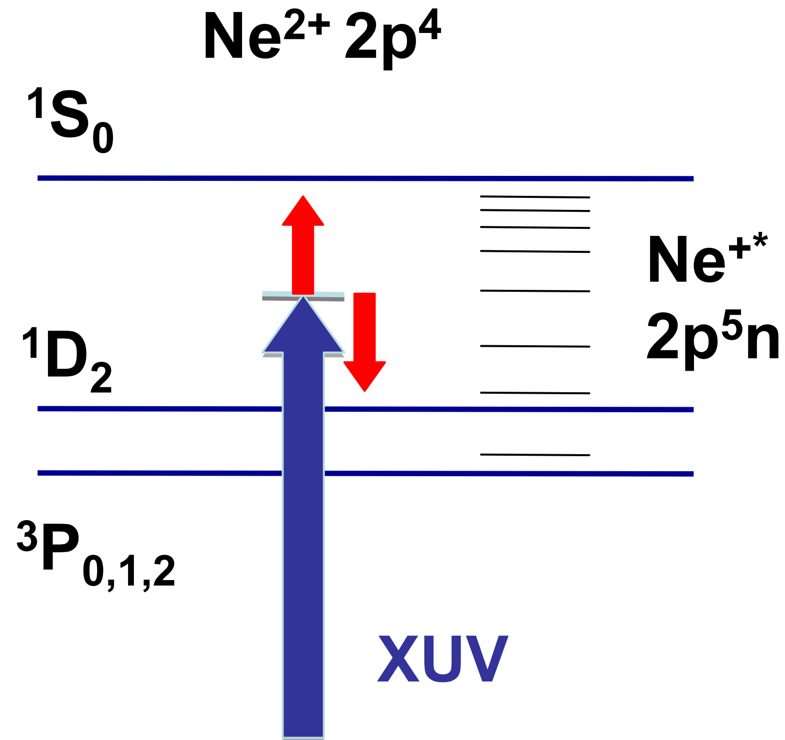


Photoionization of dressed ionic states



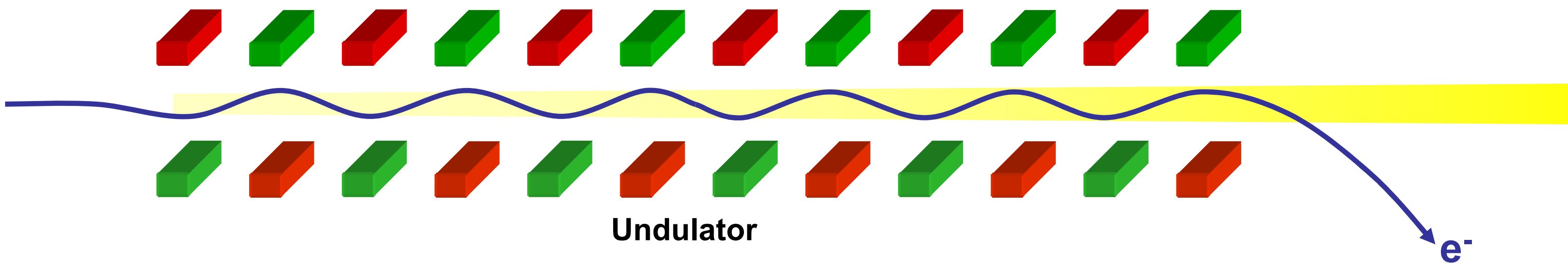
$I(^1D_2) / I(^3P_{0,1,2}) = 0.67$

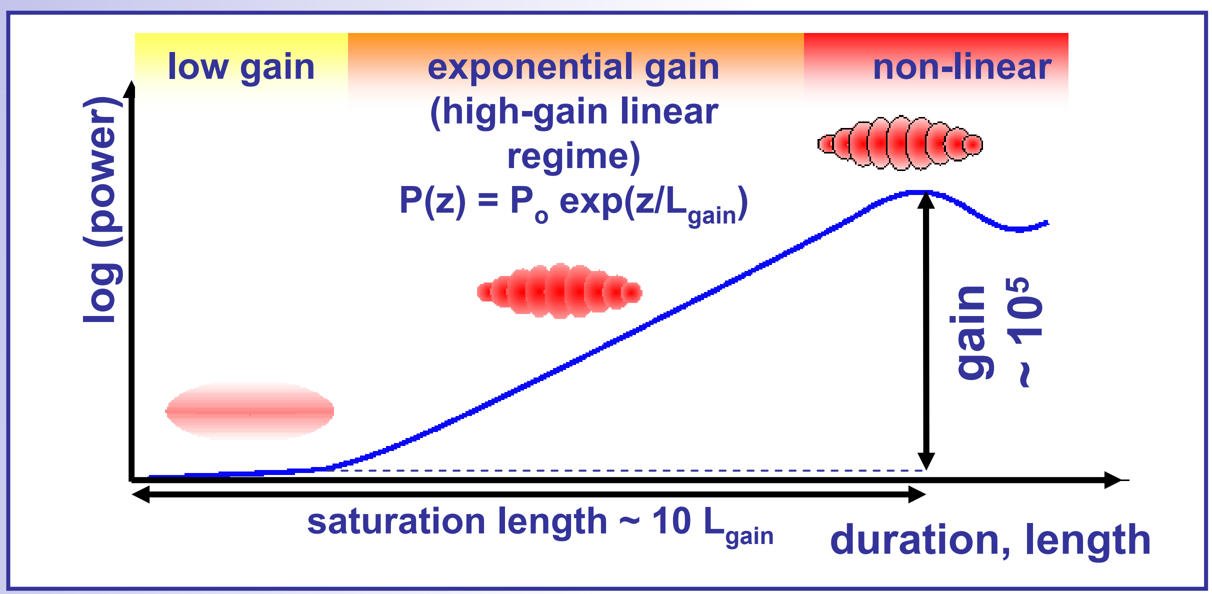
Two-photon resonances



Self-Amplified Spontaneous Emission

SASE FEL





... for X-ray wavelength

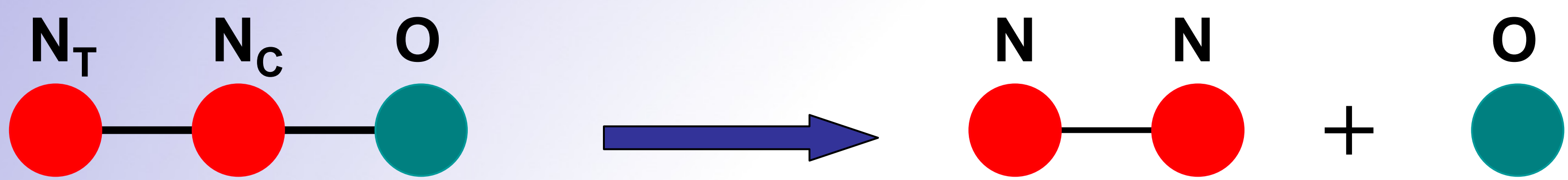
- no efficient reflectors exist
- lasing in a ‚single-pass‘
- amplified spontaneous emission

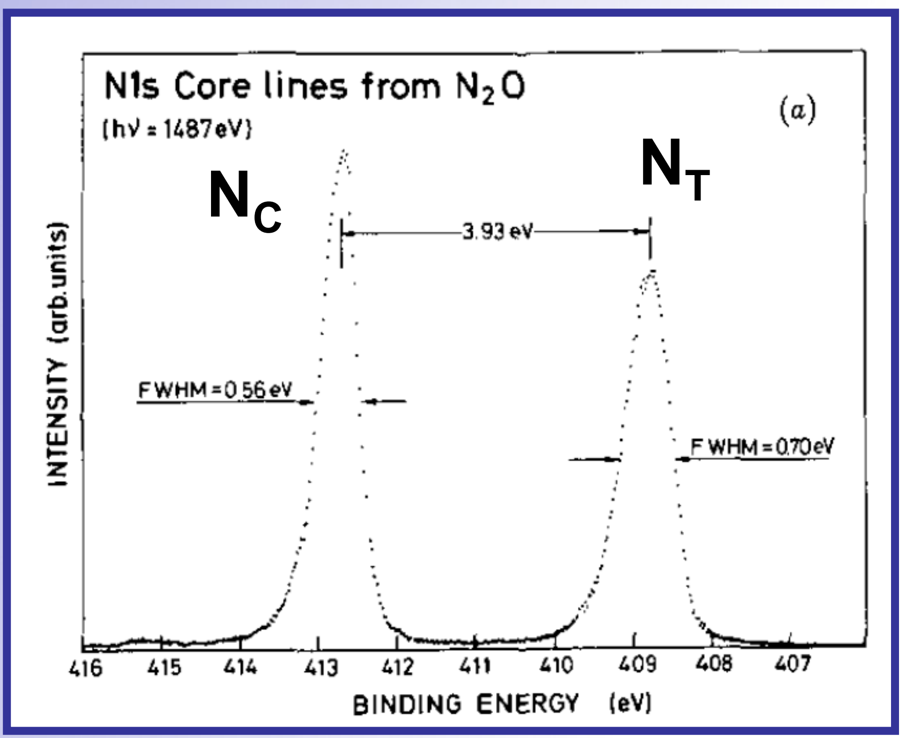
Two-color experiments

Two-photon resonant excitation

Next: Photodissociation dynamics

Innershell pump-probe experiment

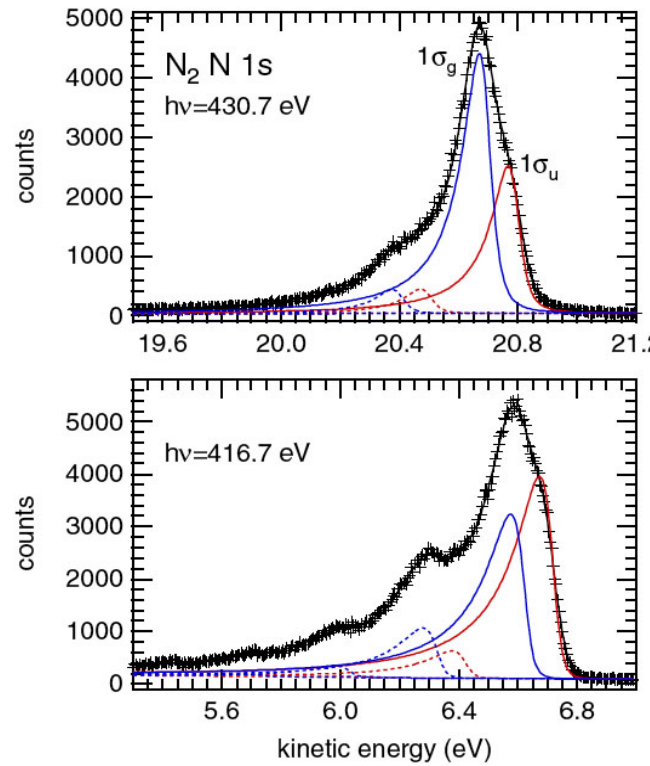




Giffiths et al., J.Phys.B24, 4187 (1991)

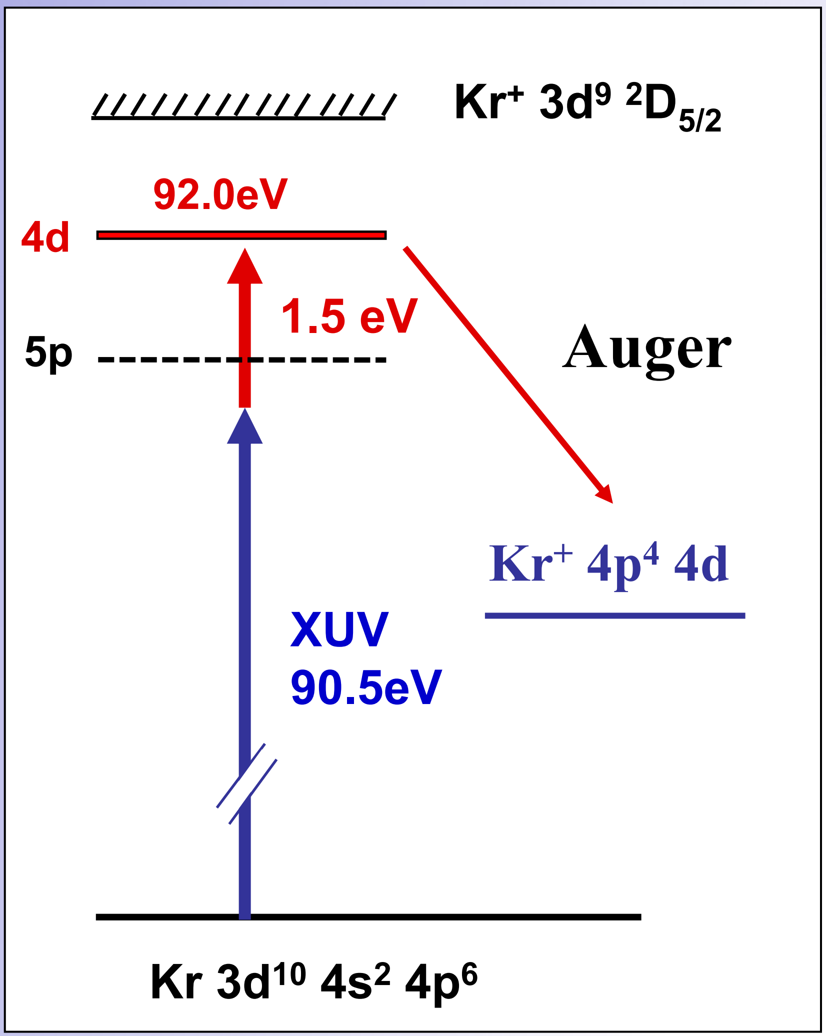
□T

I(□)

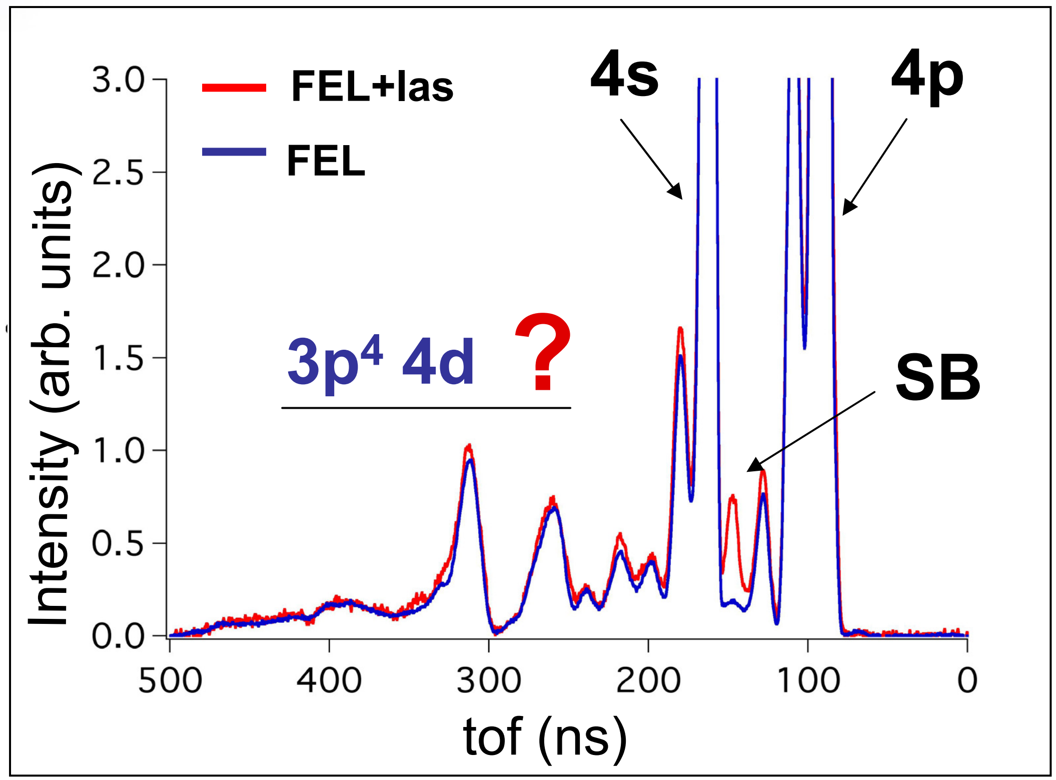


U. Hergenhahn, J.Phys.B37, R89 (2004)

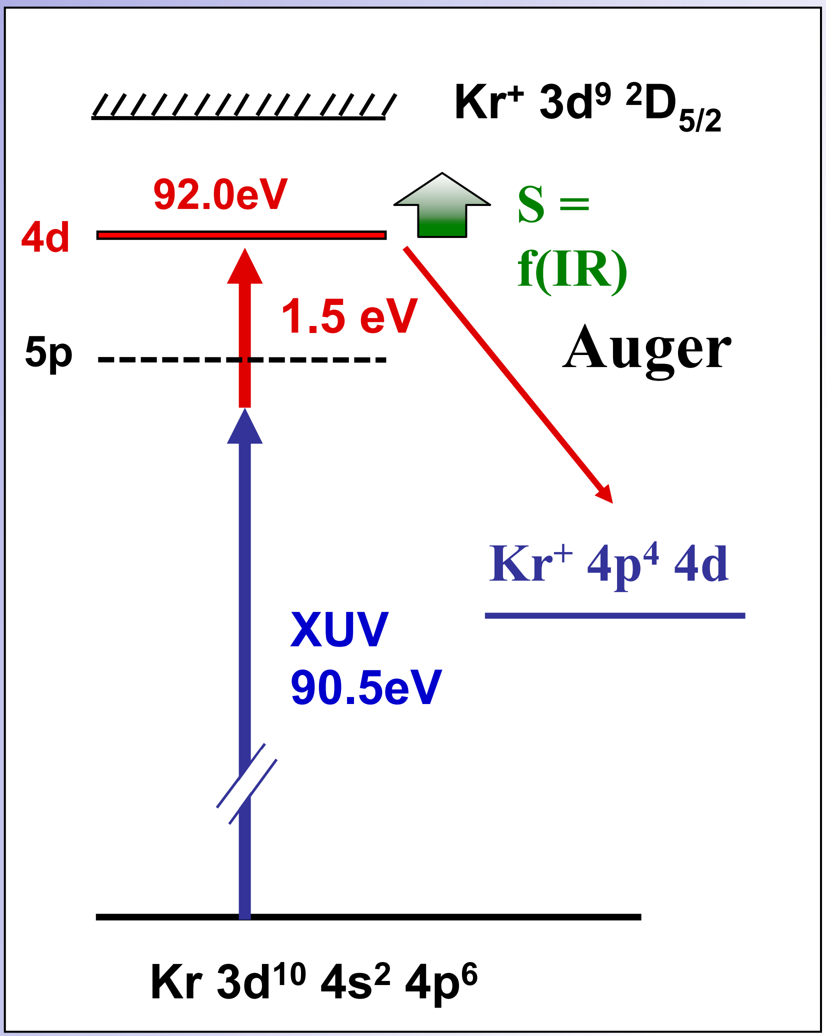
Two-photon resonance : Kr* $3d^9$ 4d



Resonant Auger Spectrum (Two-Photon)

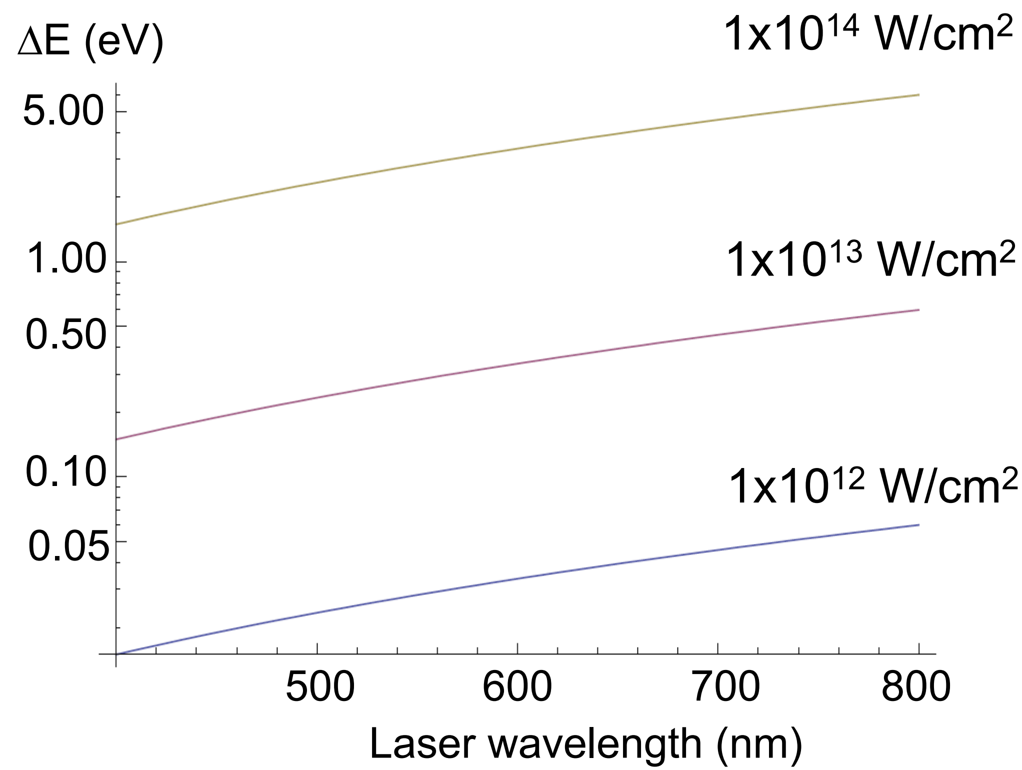


Two-photon resonance : Kr* 3d⁹ 4d

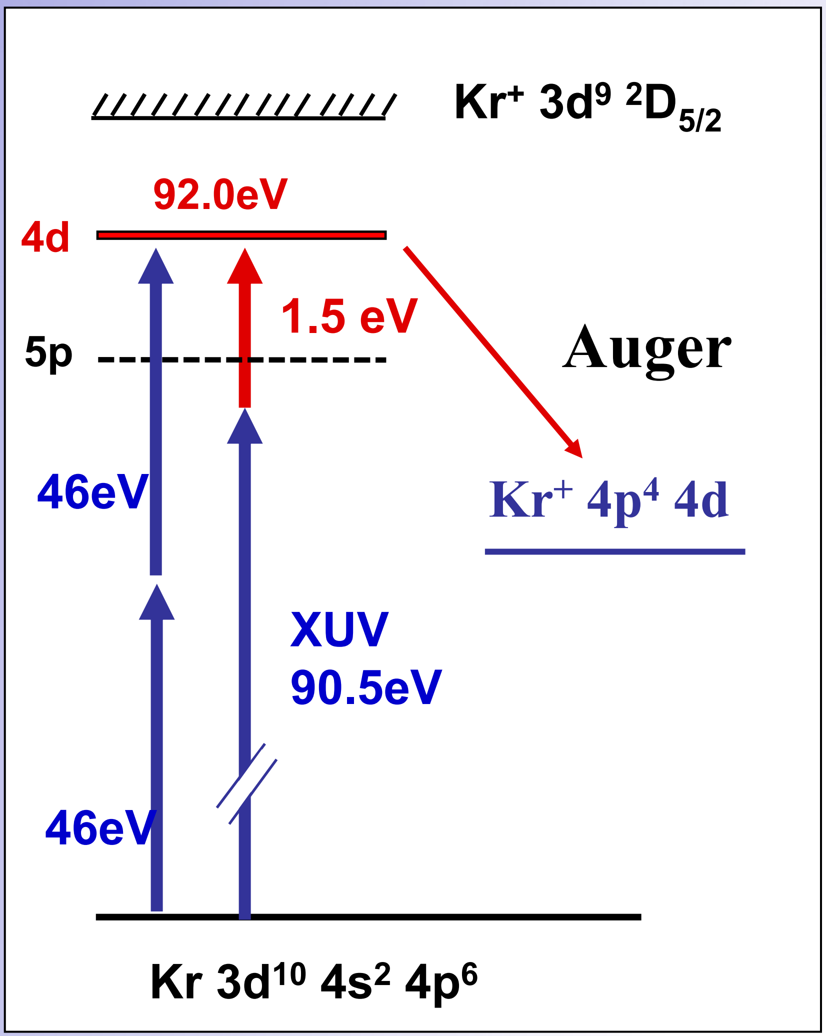


P. Lambropoulos, A. Mihelic

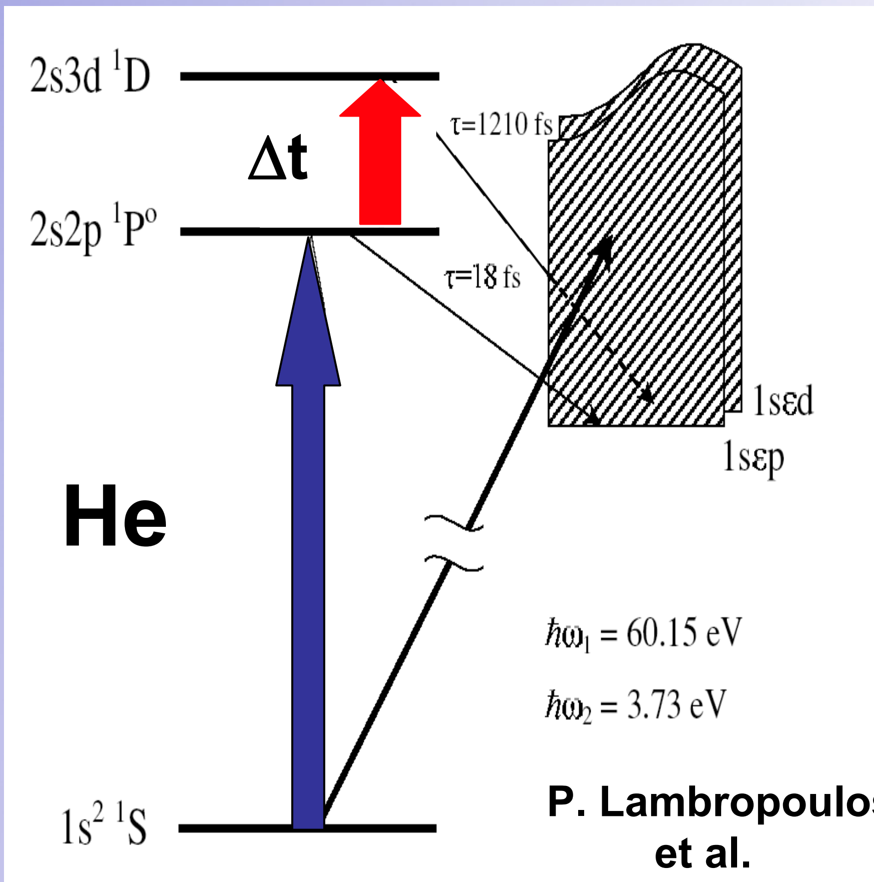
Ponderomotive shift "S"
of resonance positions



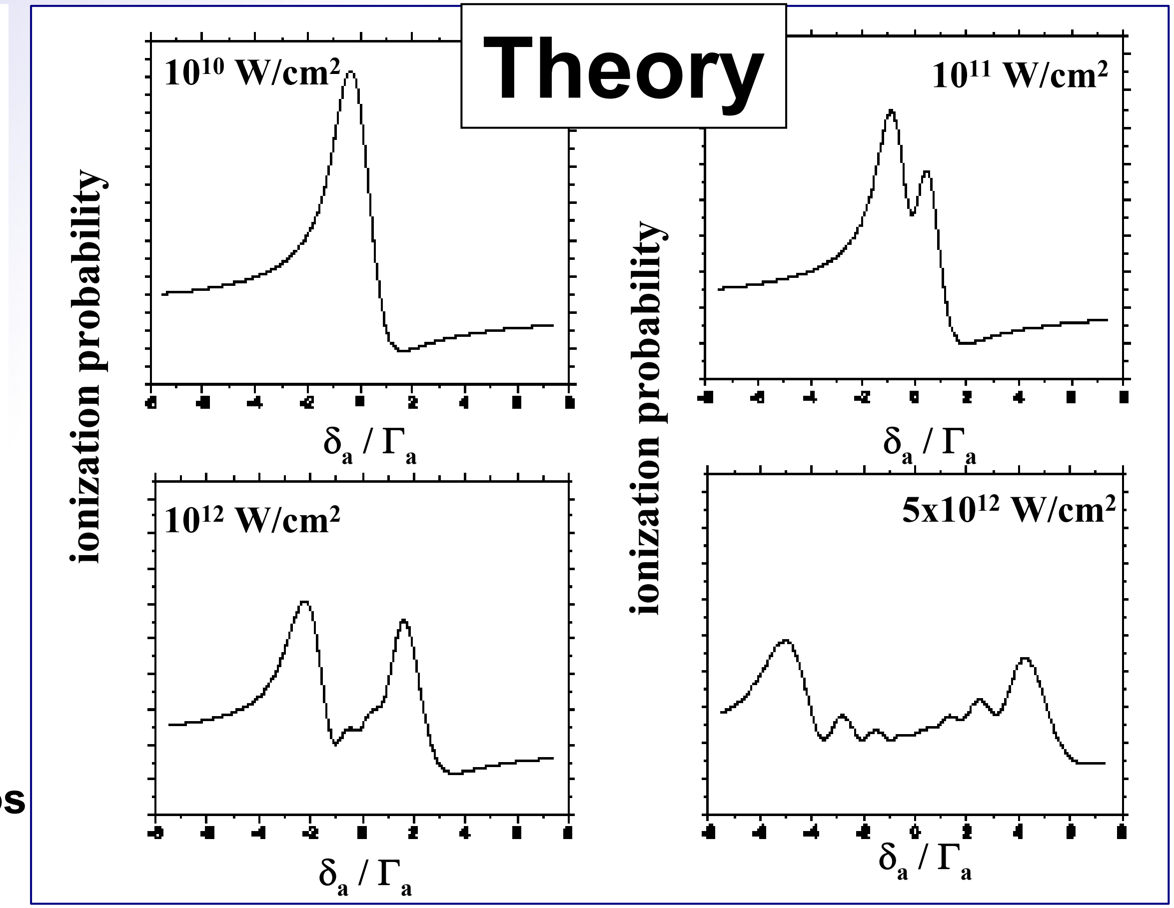
Two-photon resonance : Kr* $3d^9$ **4d**



Next: Laser-Coupling of Autoionization States



P. Lambropoulos et al.

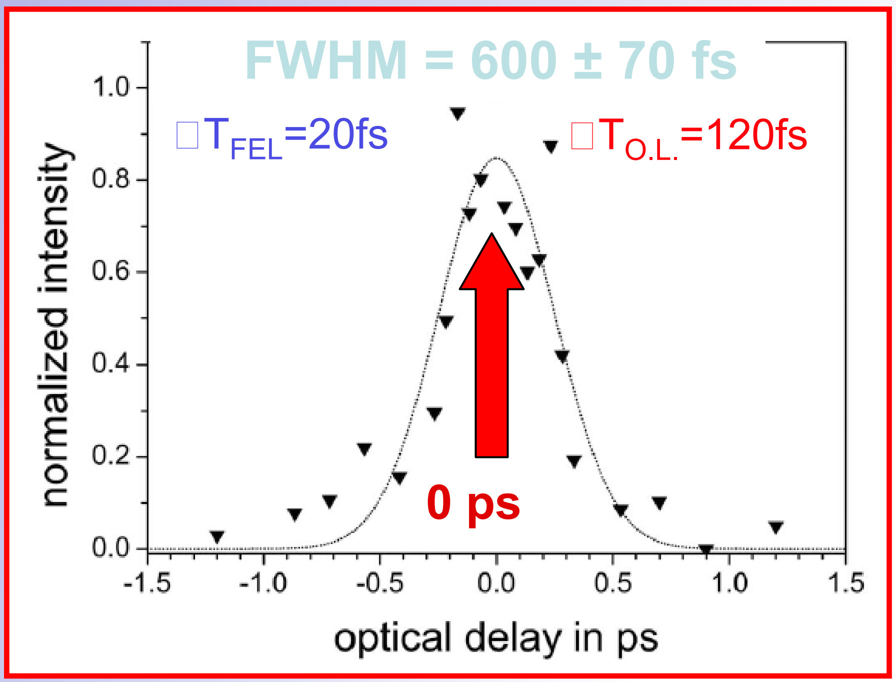


S.I. Themelis, P. Lambropoulos, M. Meyer
J. Phys. B 37, 2832 (2004)

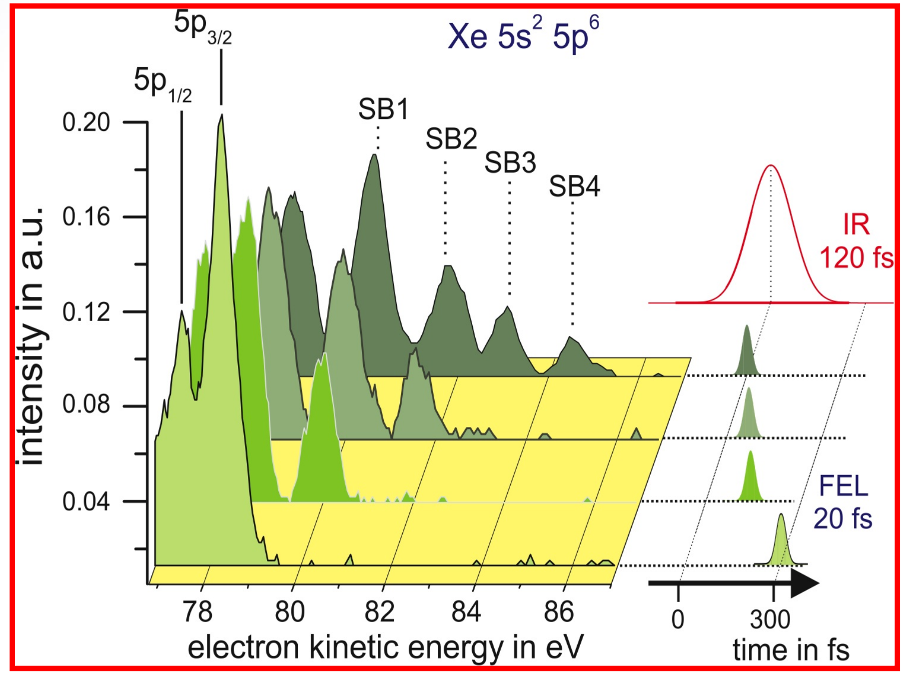
Time-dependent calculations
Dynamics of electronic relaxation

Temporal resolution / Synchronization

Average Mode:
Cross Correlation Curve



Single Shot Mode:
Photoelectron spectra



temporal resolution □T = 600 fs

➡ **Jitter (FEL) = 550 ± 80 fs**

temporal resolution □T ≤ 50 fs

➡ **only for overlapping pulses**

M. Meyer et al. PRA **74** 011401R (2006), P. Radcliffe, et al., APL **90** 131108 (2007**)**
A. Maquet, R. Taïeb, J. Mod. Opt. **54** 1847 (2007)