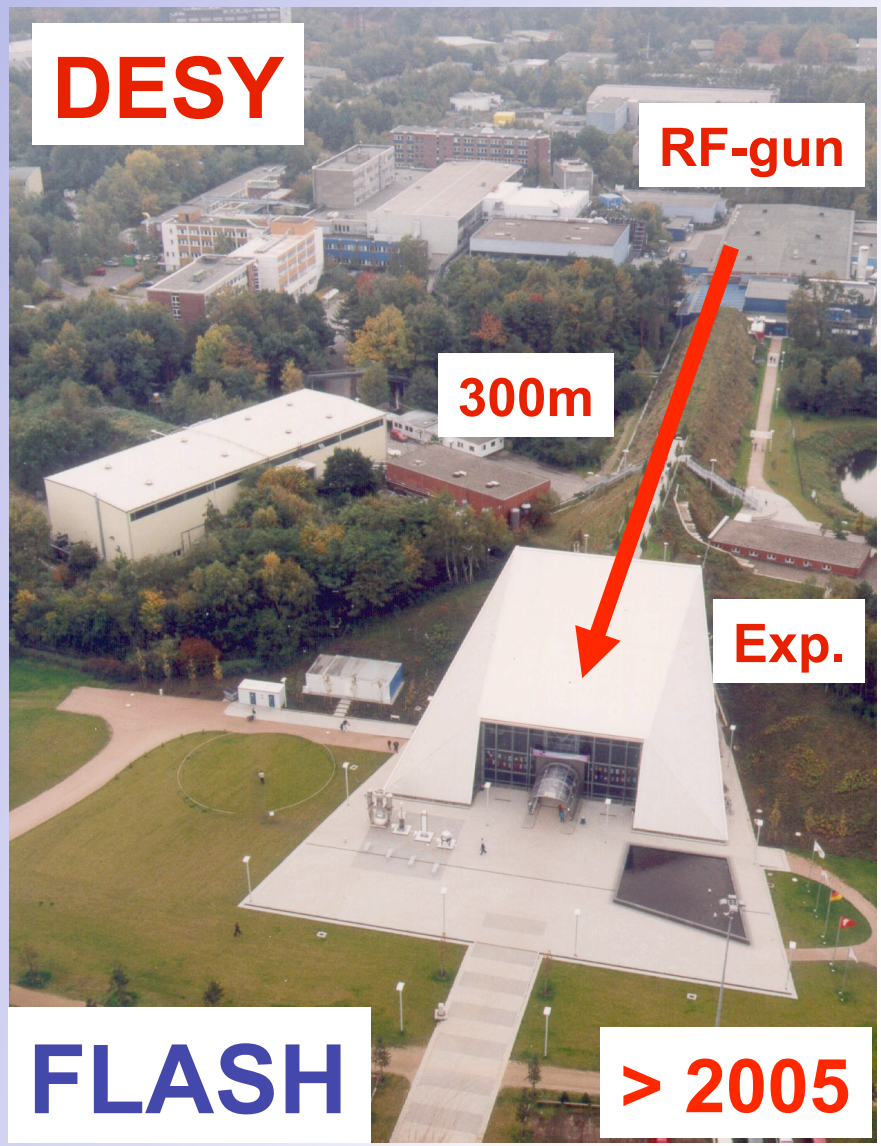
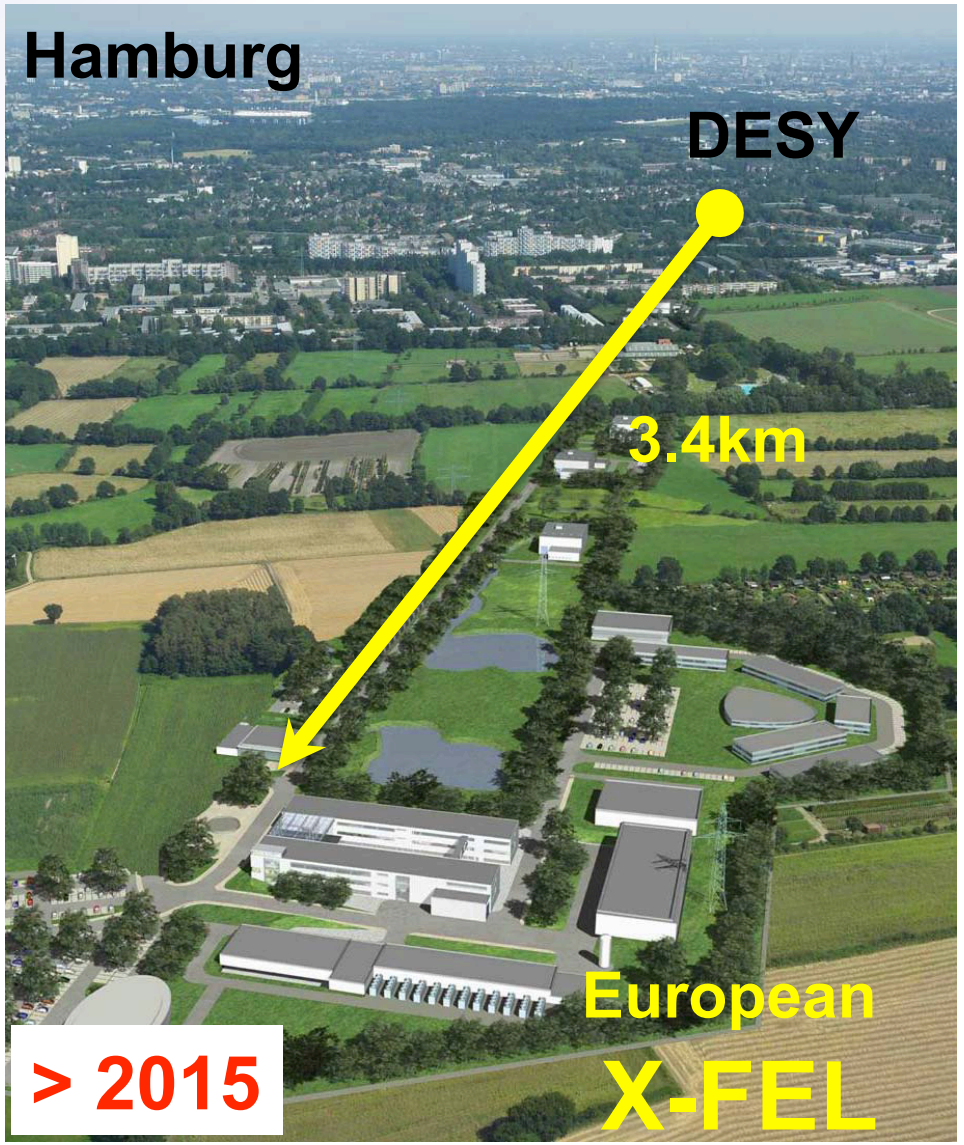
Atomic Photoionization Dynamics in Intense Radiation Fields

M. Meyer
LIXAM, Centre Universitaire Paris Sud, Orsay France

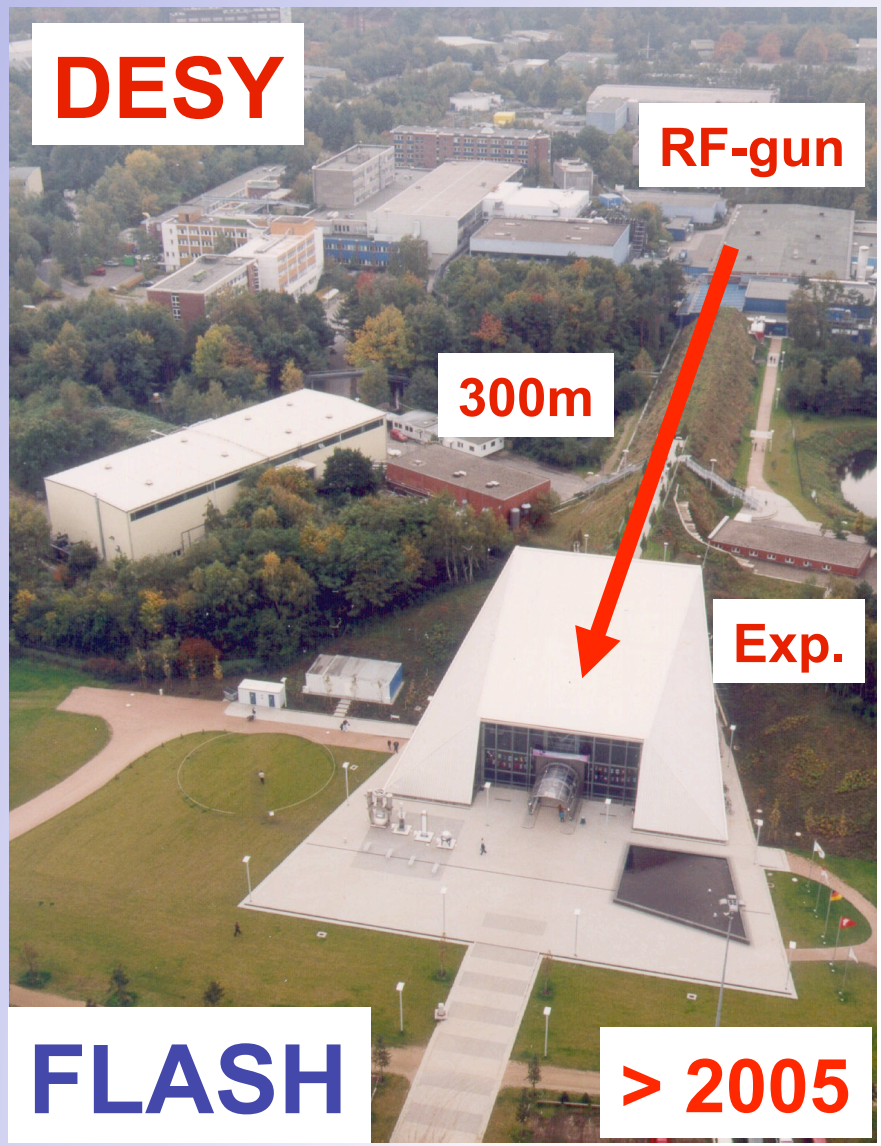
- Introduction
- Two-Color (XUV + NIR) Experiments
  - intense NIR
  - intense XUV
- Time-resolved Pump-Probe Experiments
- Conclusion

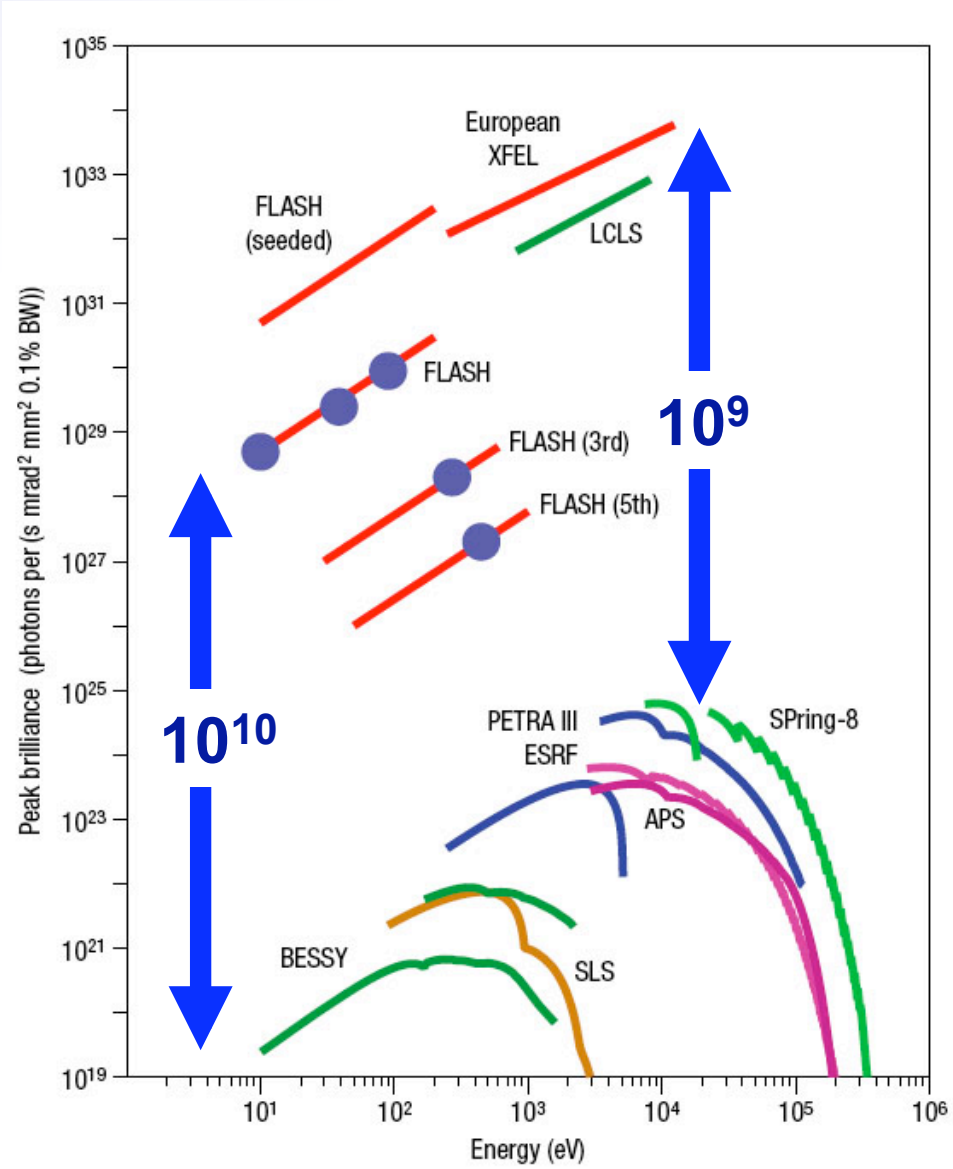
Free Electron Lasers in Hamburg





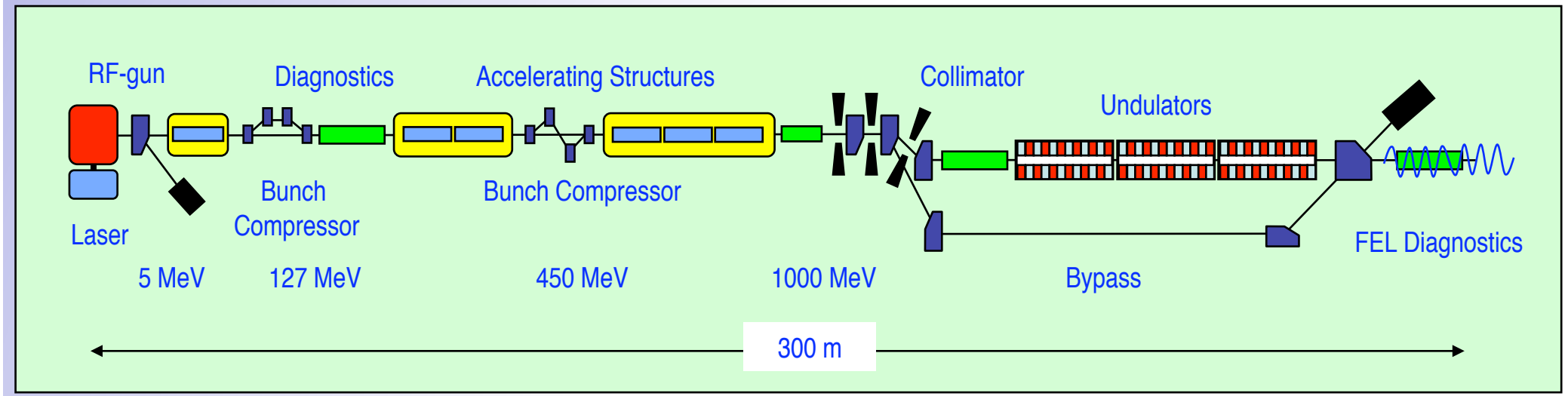
Free Electron Lasers in Hamburg





FLASH (Free electron LASer in Hamburg)

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

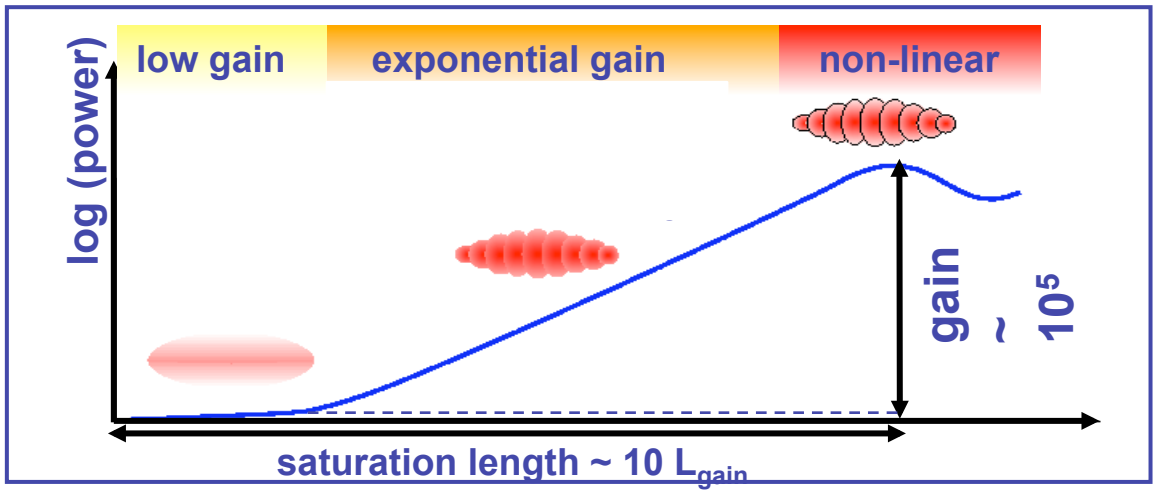


LINAC : 1 GeV

30 m fixed-gap undulator

macro-bunch at 10 Hz

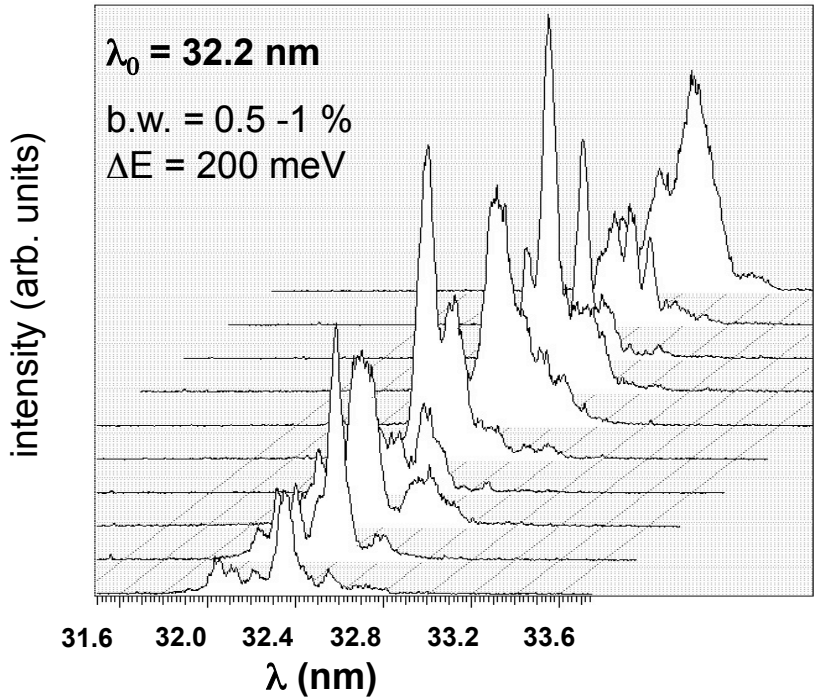
> 30 bunches, 1 μs separation



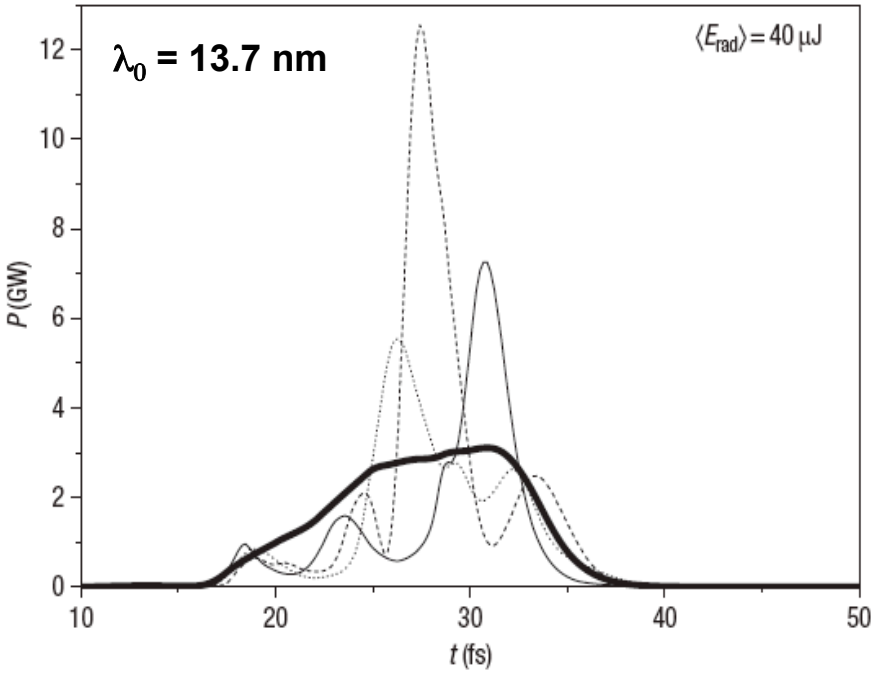
SASE (Self Amplified Spontaneous Emission)

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

~ 10^{12} - 10^{13} photons/pulse

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

- Short Pulses

ΔT = 10 - 20 fs

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

~ 10 - 100 μJ (> 10^{16} W/cm^2)

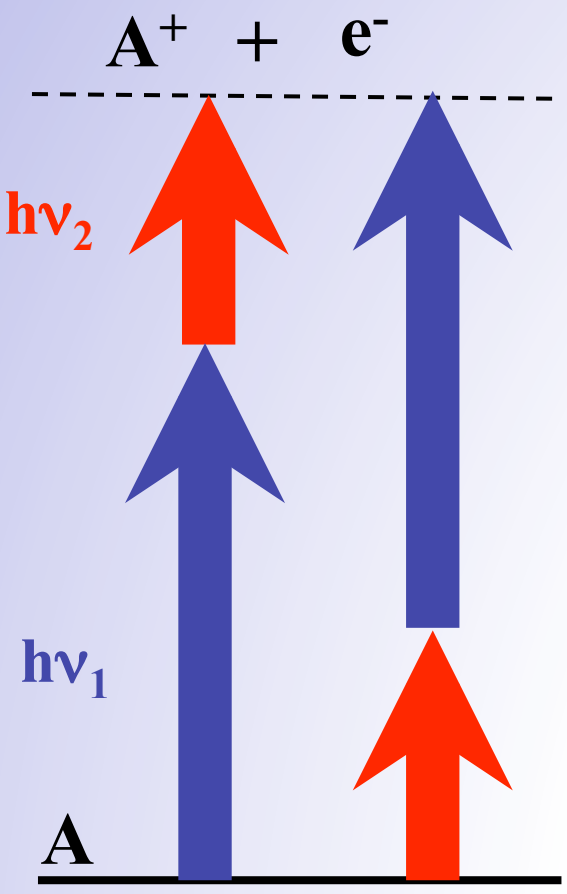
→ **Non-Linear Processes**
(Multi-photon)

- 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

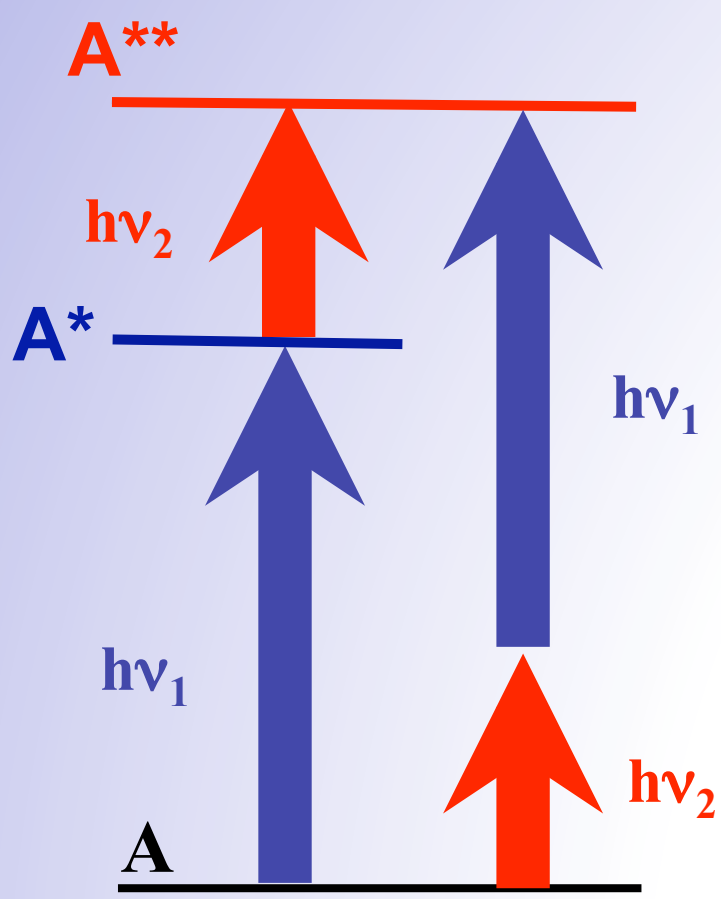
2) Dynamics in strong fields

"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 / cm^2

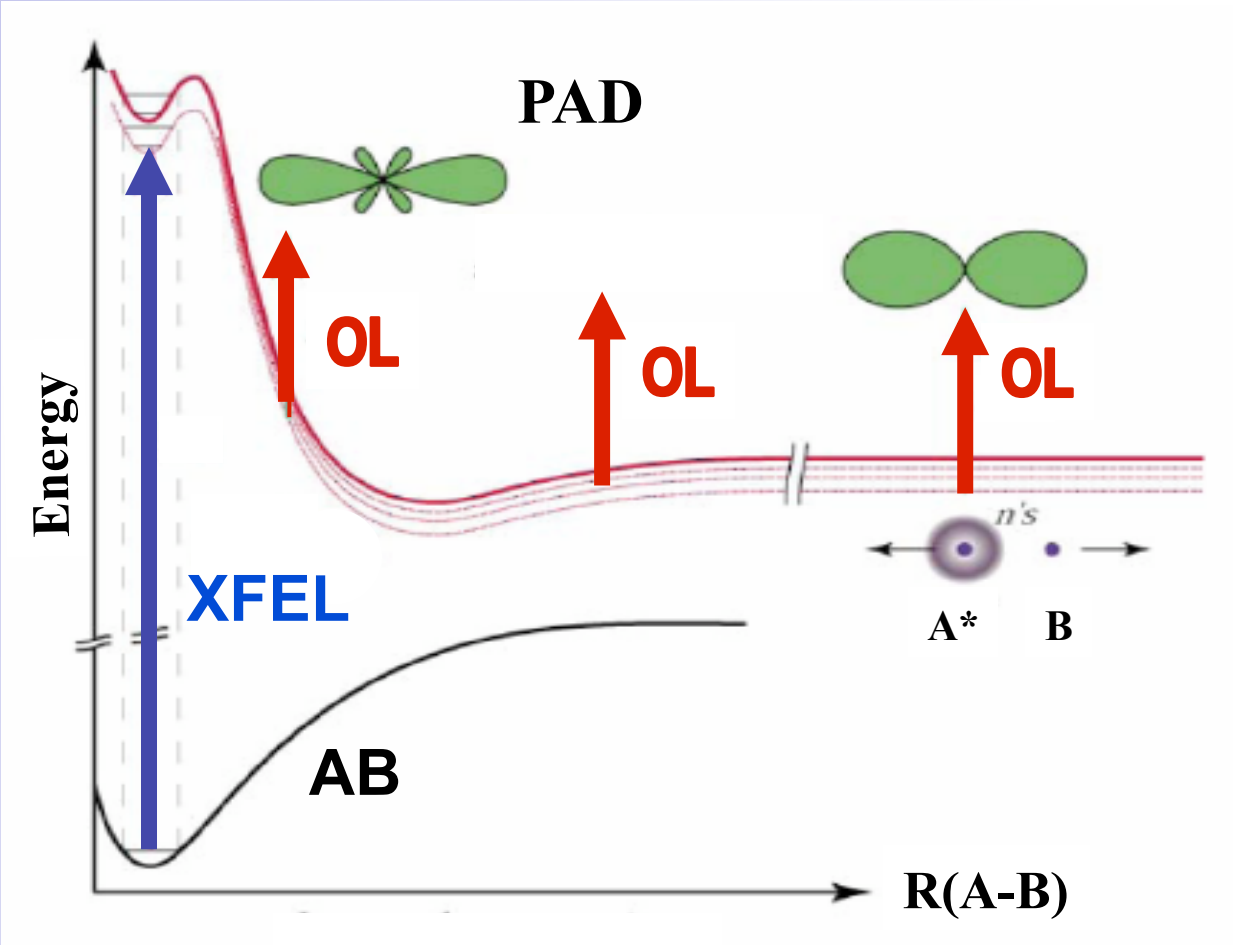
$h\nu_1$: FEL
$>10^{12}$ W / cm^2

3) Non-linear processes:

- Two-photon resonances
- Coupling of Autoionizing States

Photodissociation Dynamics

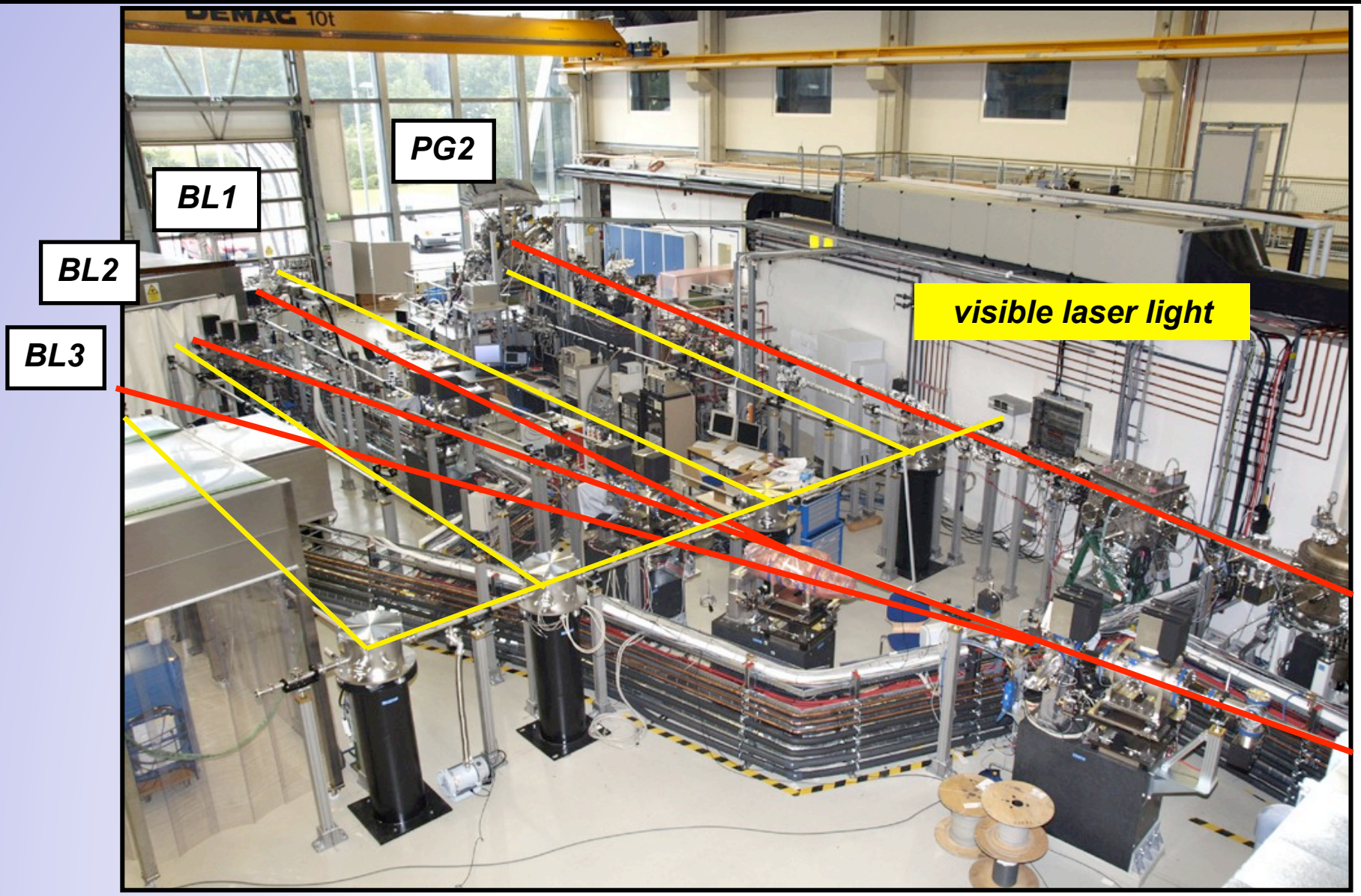
Time-resolved development of the electronic structure during dissociation or chemical reactions



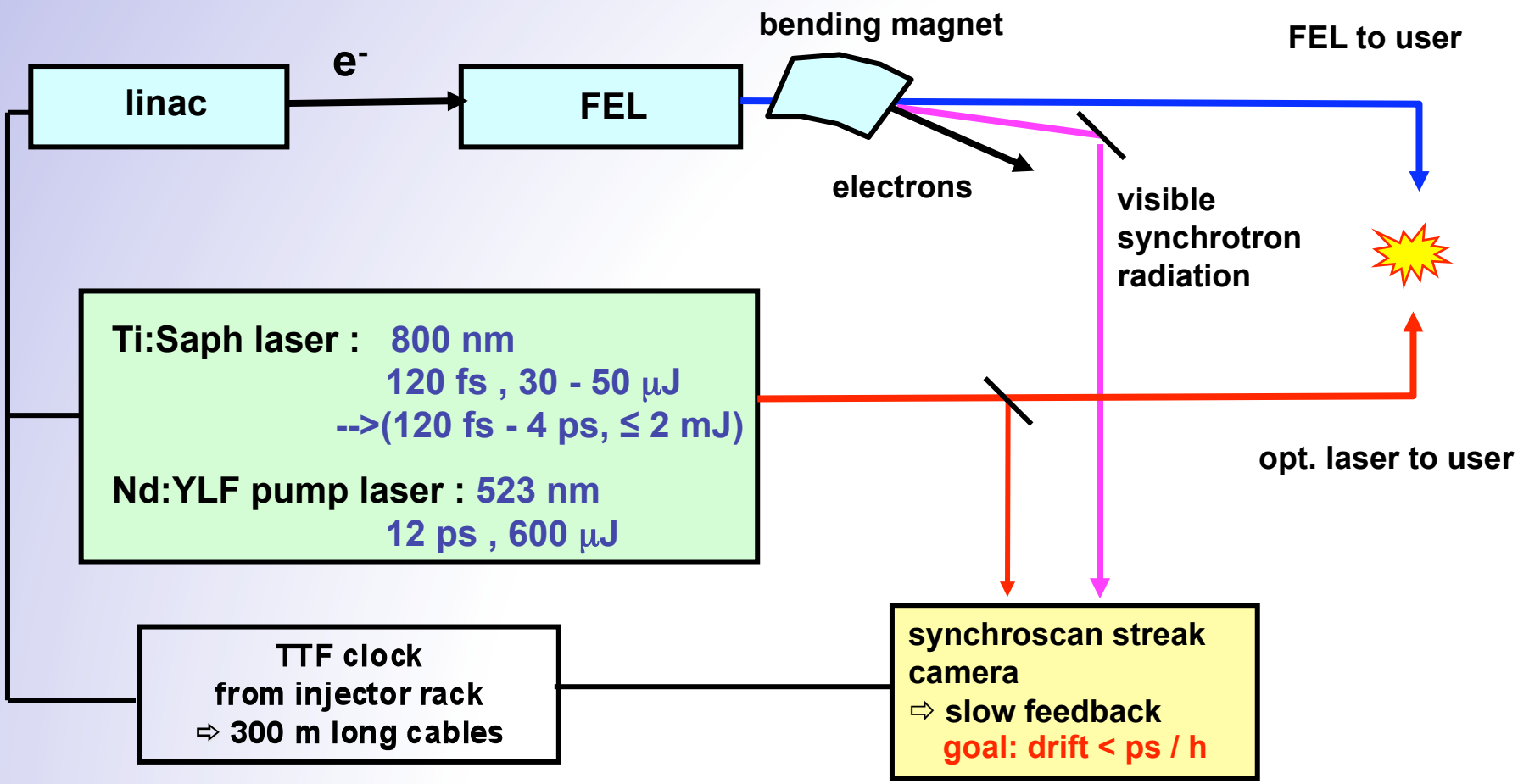
Excitation of core electrons

element specific
« Pump » or « Probe »

Experiments at FLASH



FLASH + Optical Laser

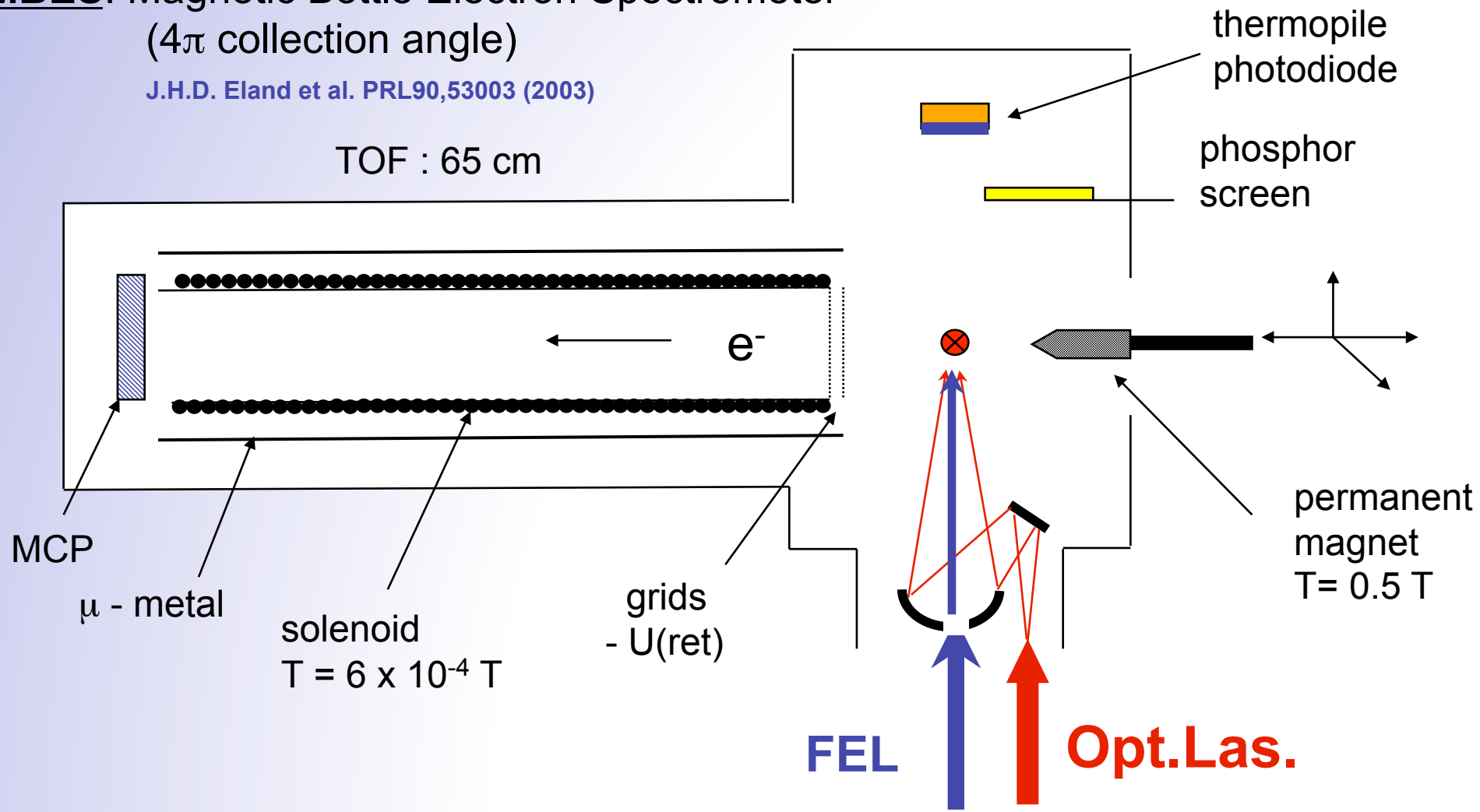


Max-Born-Institut, Berlin
I. Will, H. Redlin

Two-Color Photoelectron Spectroscopy

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

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



Two-color experiments

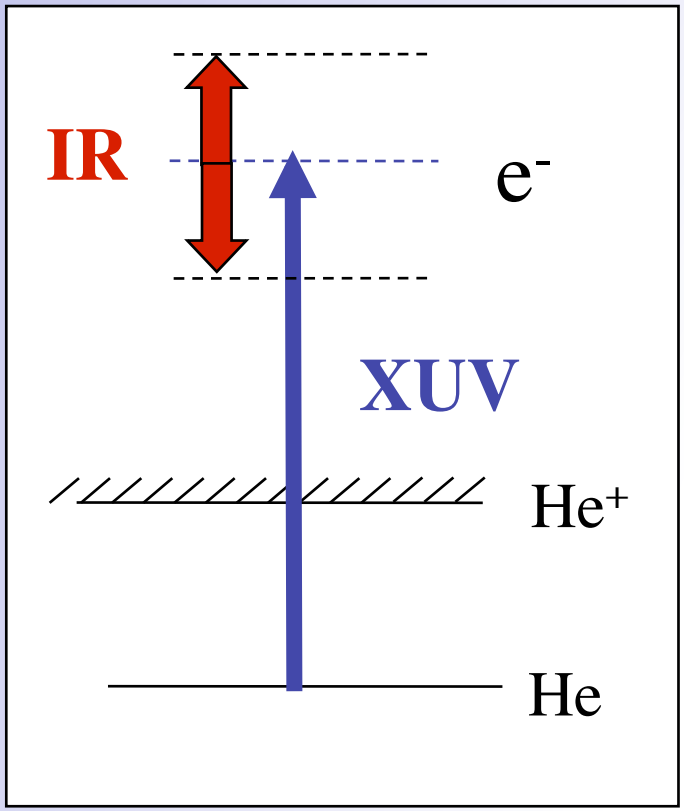
Intense optical field: Above Threshold Ionization of rare gases

Above Threshold Ionization of Rare Gases

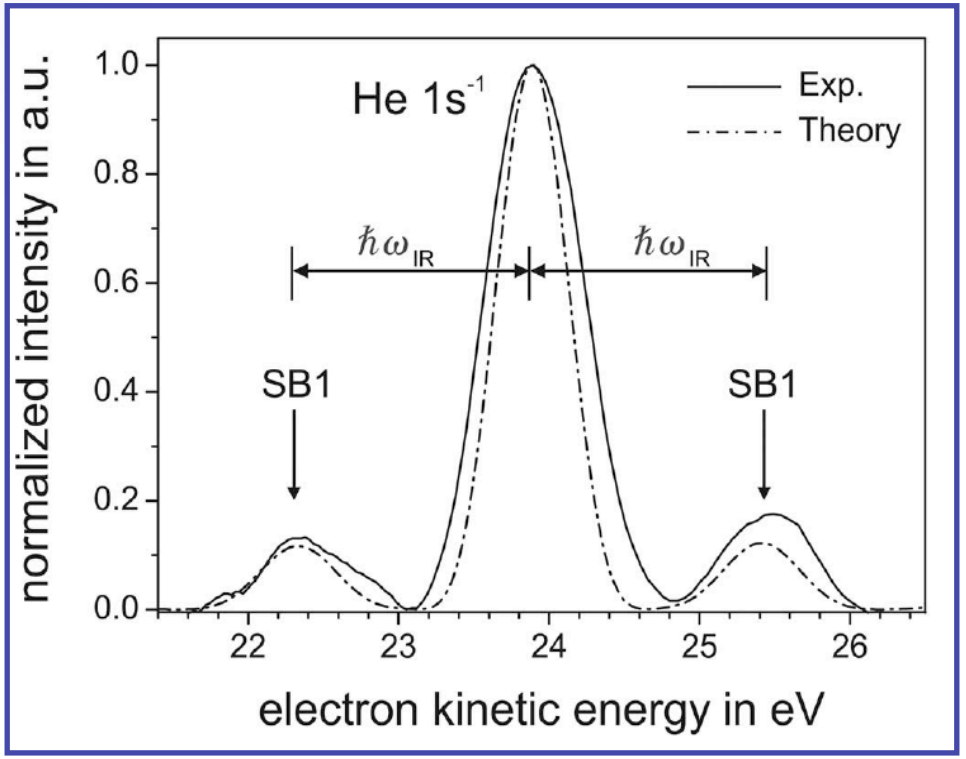
FLASH: 25.5 nm, 20 μJ, 50 μm focus, 20 fs

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

2×10^{11} W/cm^2



Toma et al. PRA 62, 0618015 (2000)



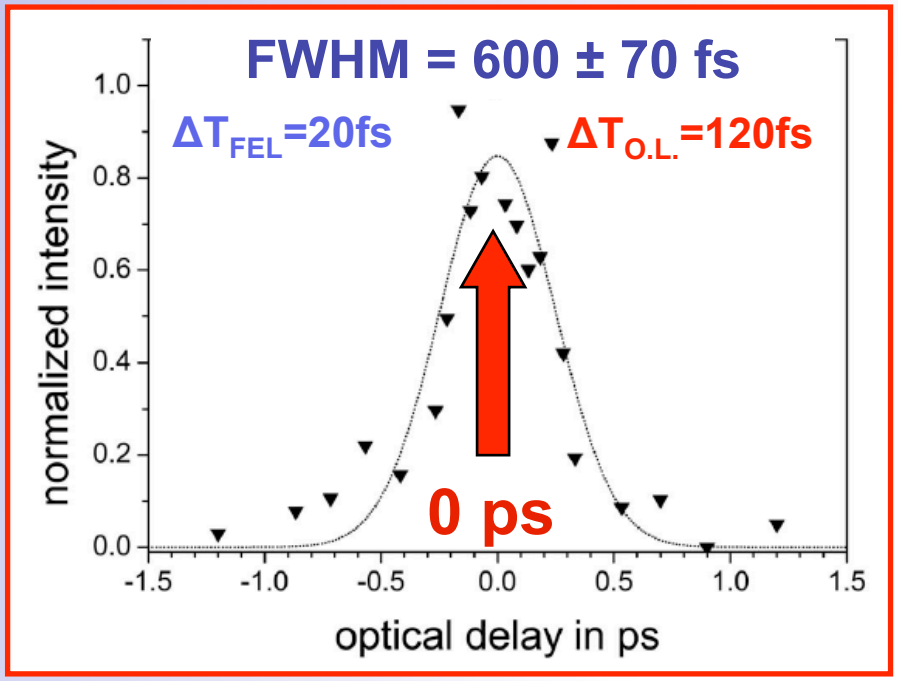
Radcliffe et al. APL 90, 131108 (2007)

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

Temporal resolution / Synchronization

Average Mode:

Cross Correlation Curve

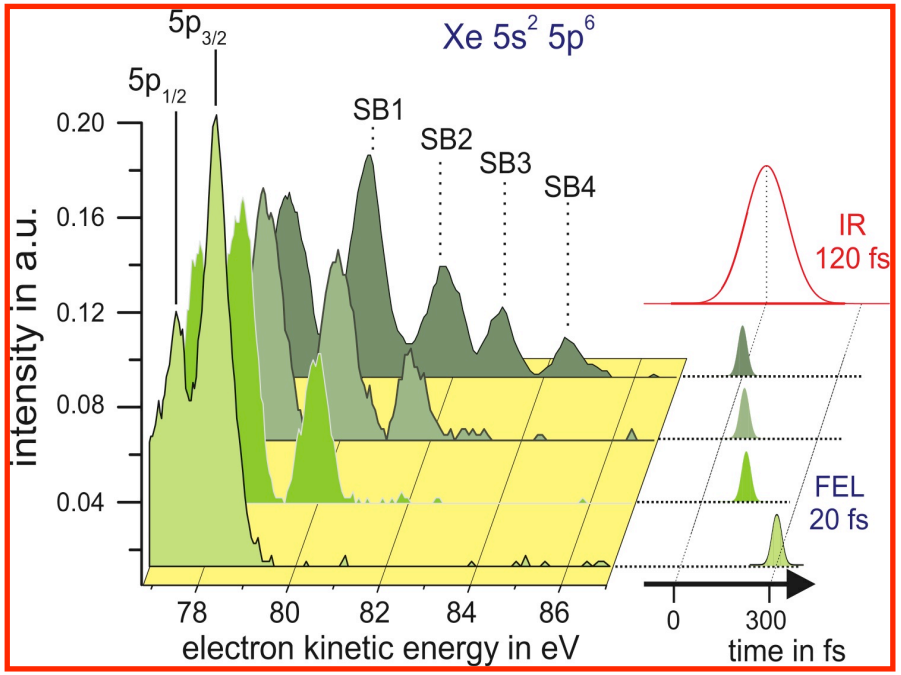


temporal resolution ΔT = 600 fs

→ **Jitter (FEL) = 550 ± 80 fs**

Single Shot Mode:

Photoelectron spectra

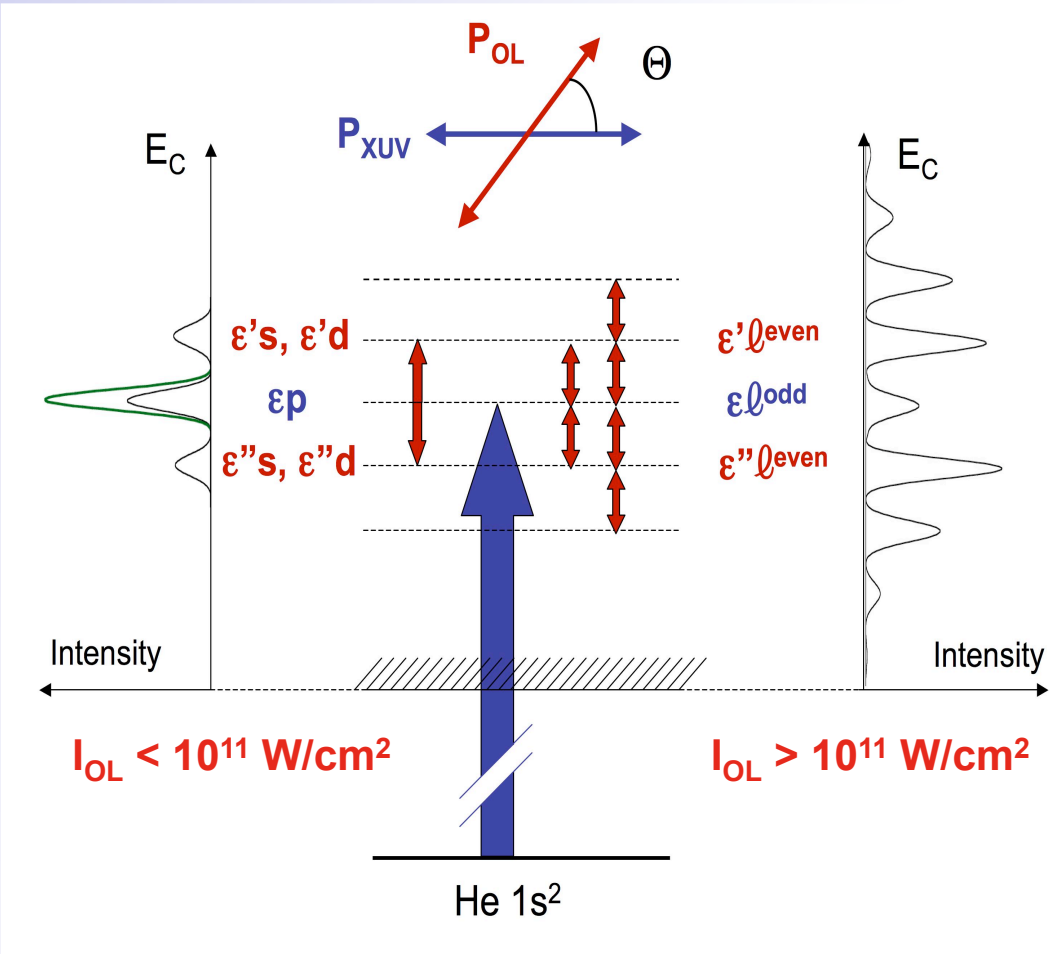


temporal resolution ΔT ≤ 50 fs

→ **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

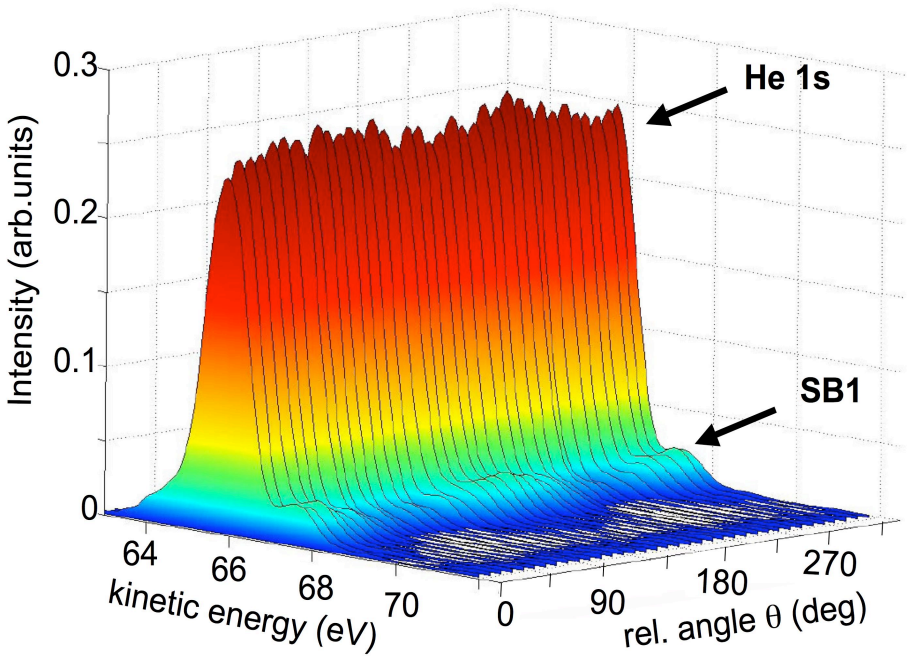
He $1s^2$ + $h\nu_{XUV}$ + $h\nu_{OL}$ ---> He^+ 1s + εs, εd

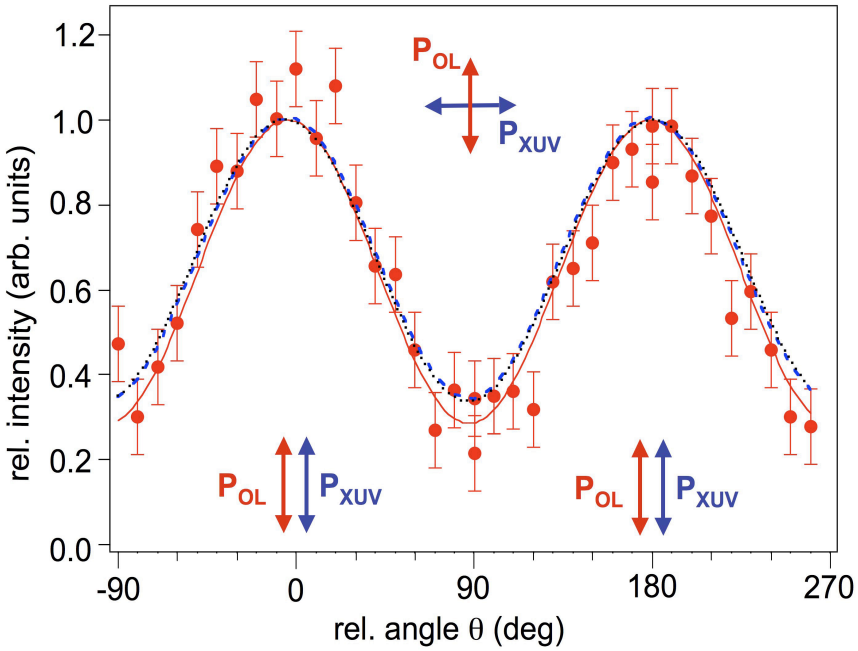
Polarization control: two-color **two-photon**

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

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

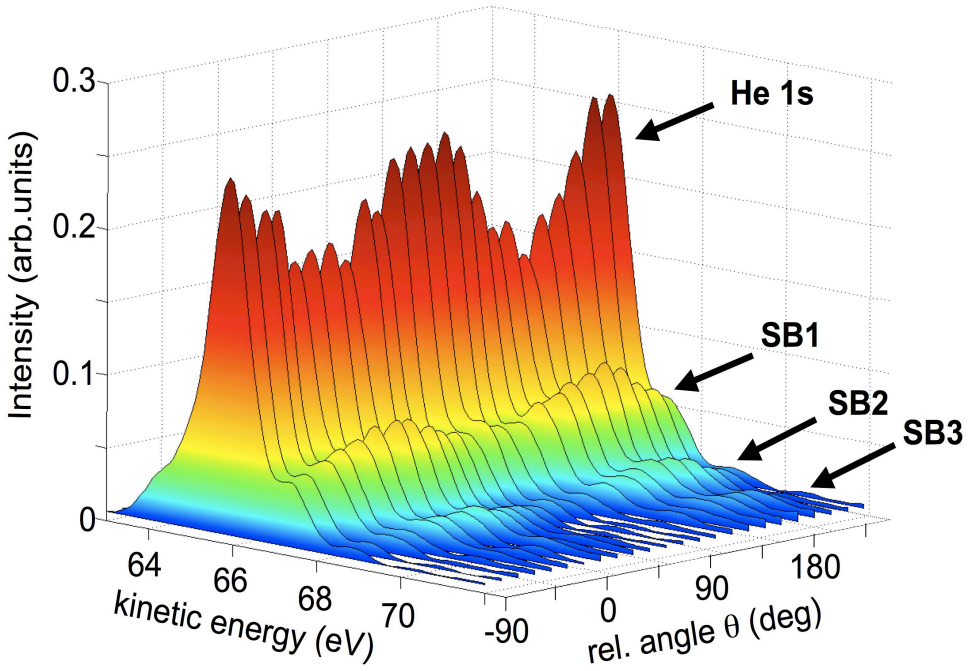
90° → only 'd' - emission

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

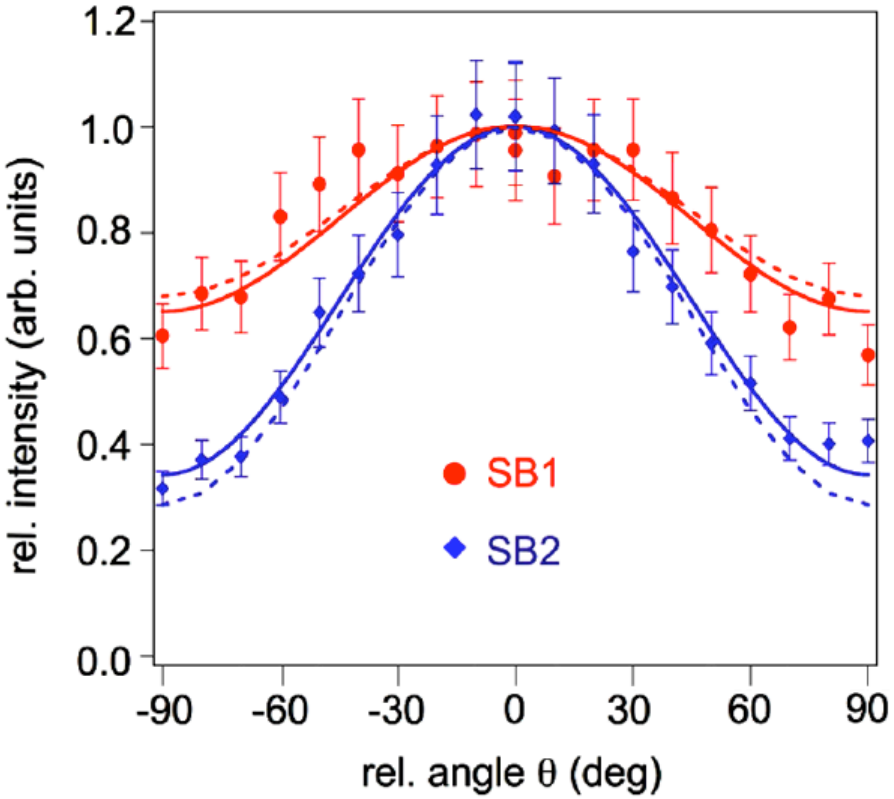


He $1s^2$ + $h\nu_{XUV}$ + $h\nu_{OL}$ ---> He^+ 1s + εl

higher l ≥ 0(s), 2(d)

"Soft-Photon Approximation"

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

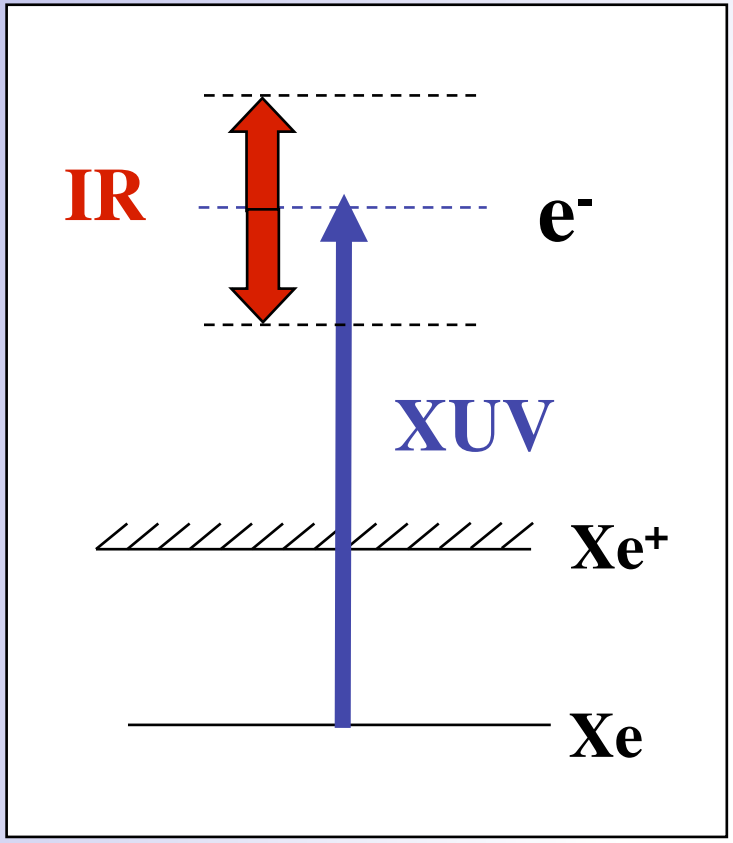


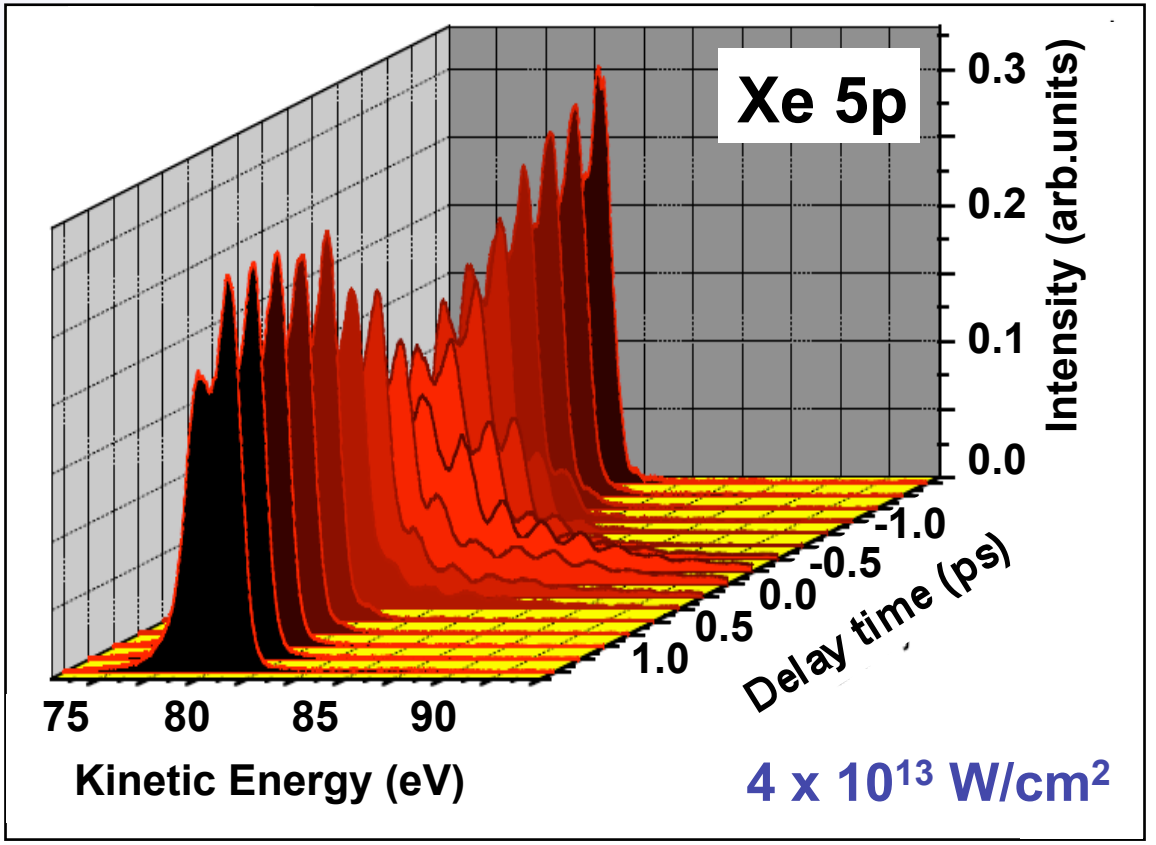
A. Maquet, R. Taieb, J. Mod. Opt. 54, 1847 (2007)

Temporal Control of ATI

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

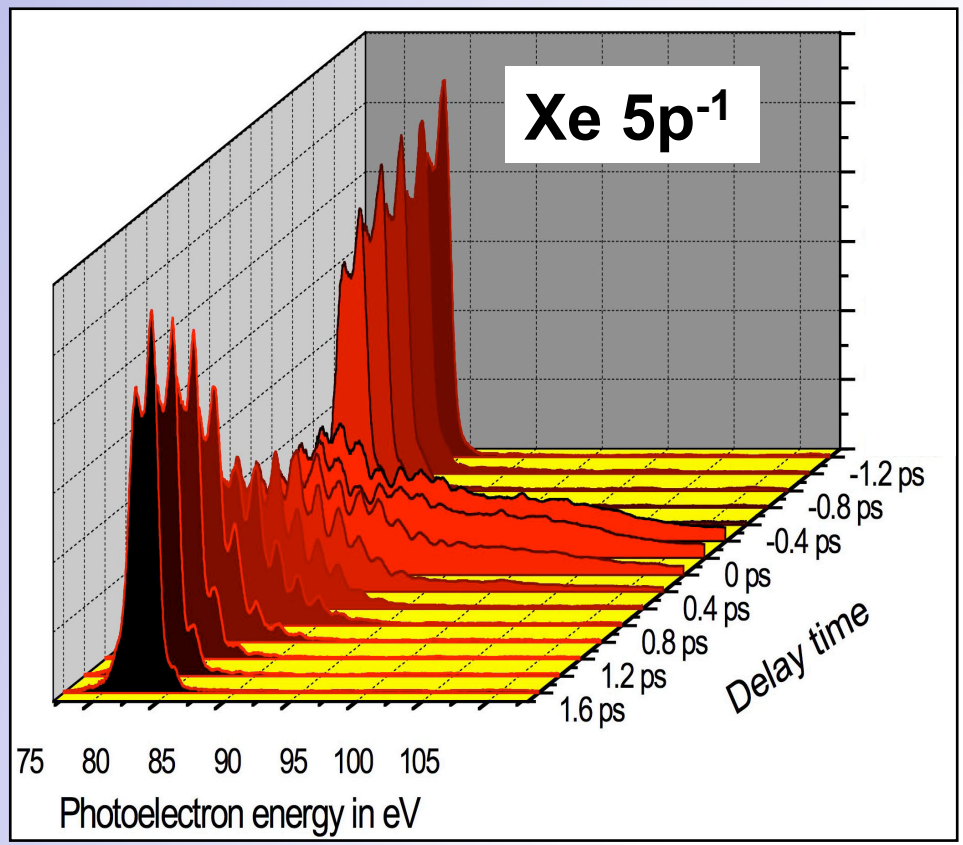
Opt. Laser : 800 nm, ≤ 4 mJ, 50 μm focus, 120 fs - 4 ps

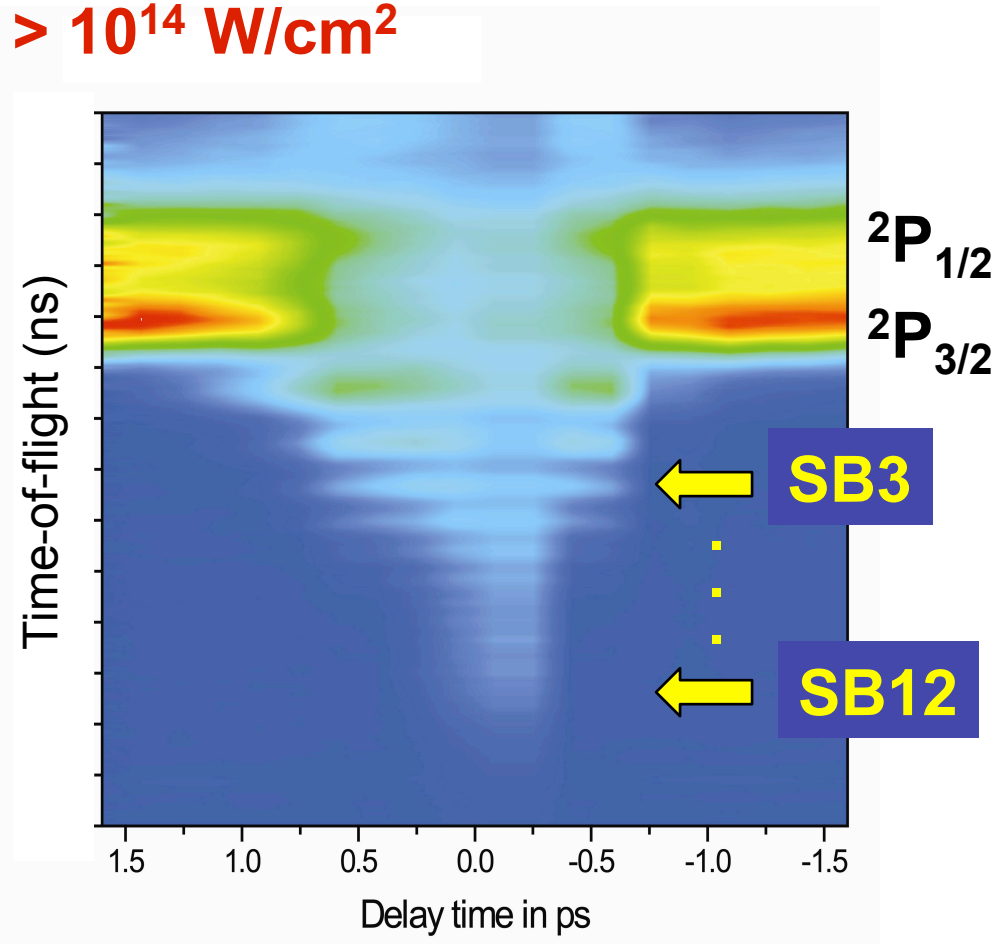




ATI : Strong NIR Dressing Field (Xe)

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





Multi-photon processes

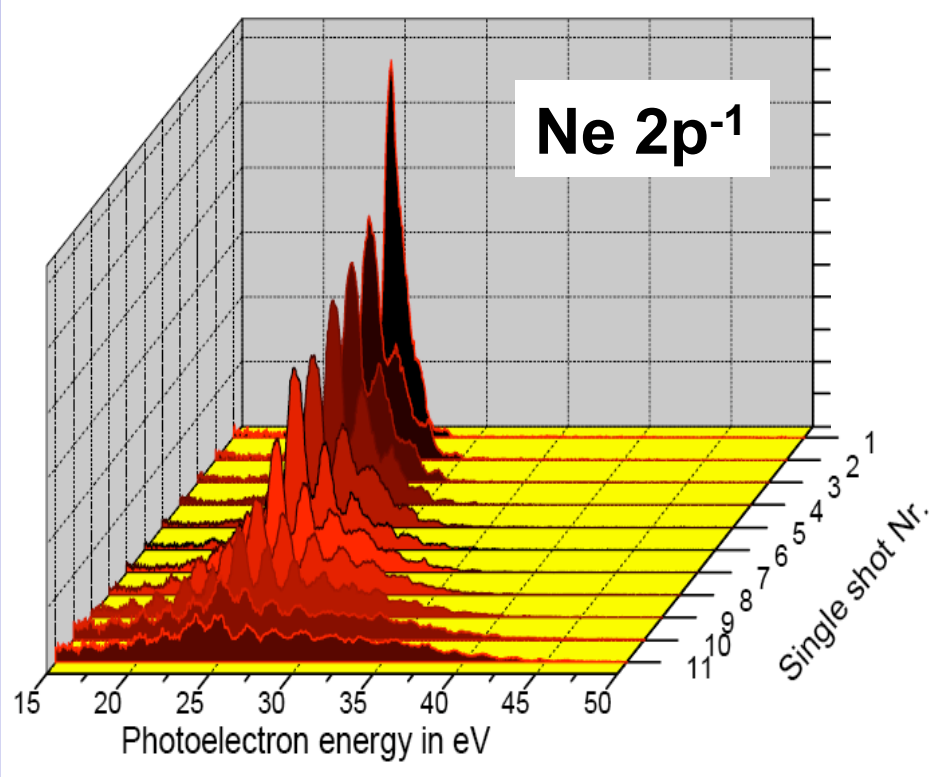
hν(800nm) = 1.5eV

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

ATI : Strong NIR Dressing Field (Ne)

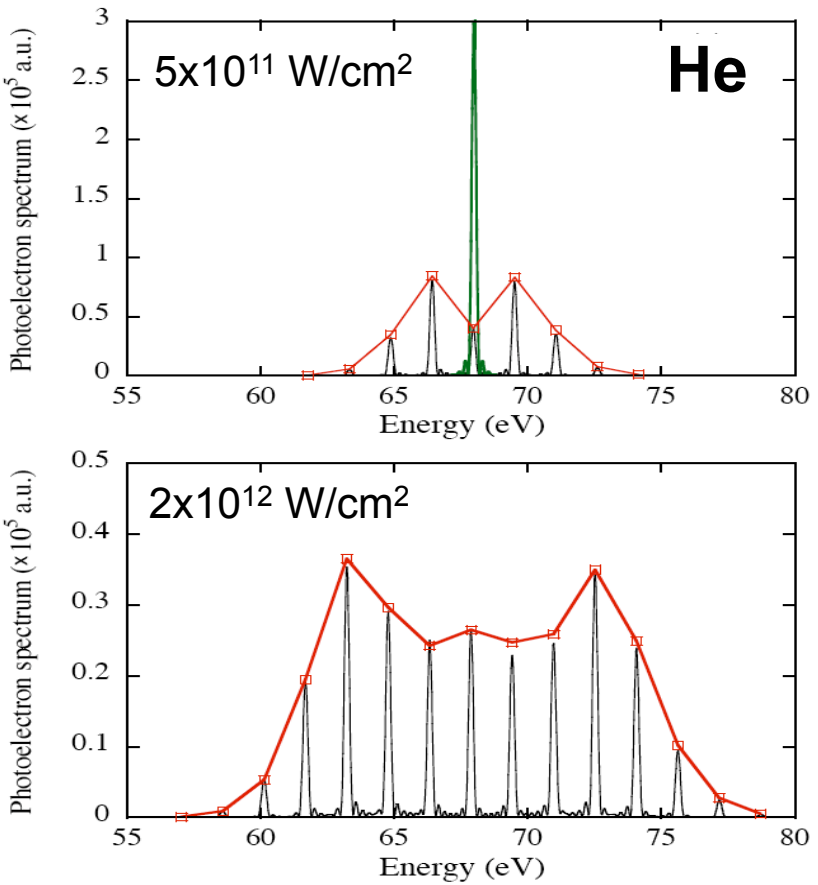
FEL: 26.9nm (46 eV)
Laser: 800nm, 1.8mJ, 100fs

$3x10^{13}$ W/cm^2



Multi-photon processes

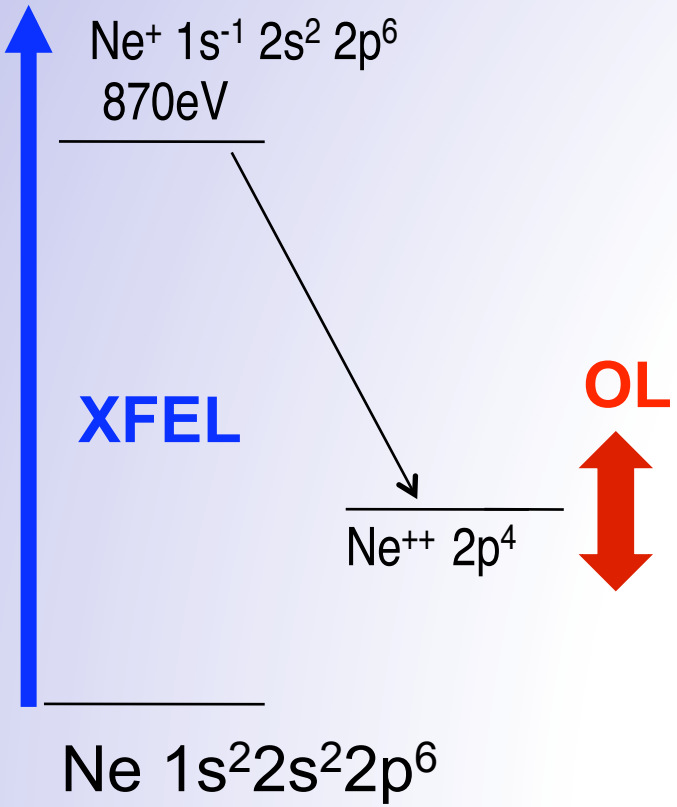
"Soft-Photon Approximation"

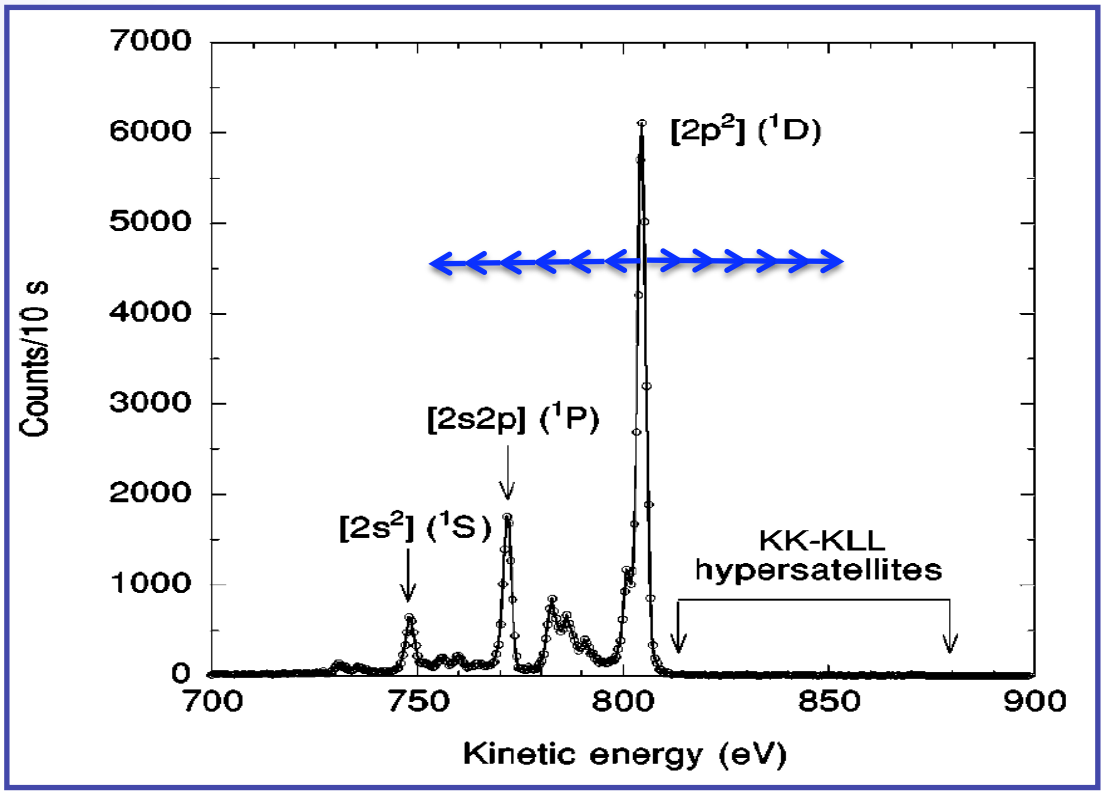


A. Maquet, R. Taieb, J. Mod. Opt. 54, 1847 (2007)

XFEL: Laser-assisted resonant Auger decay

Laser Coupling of Final Ionic States

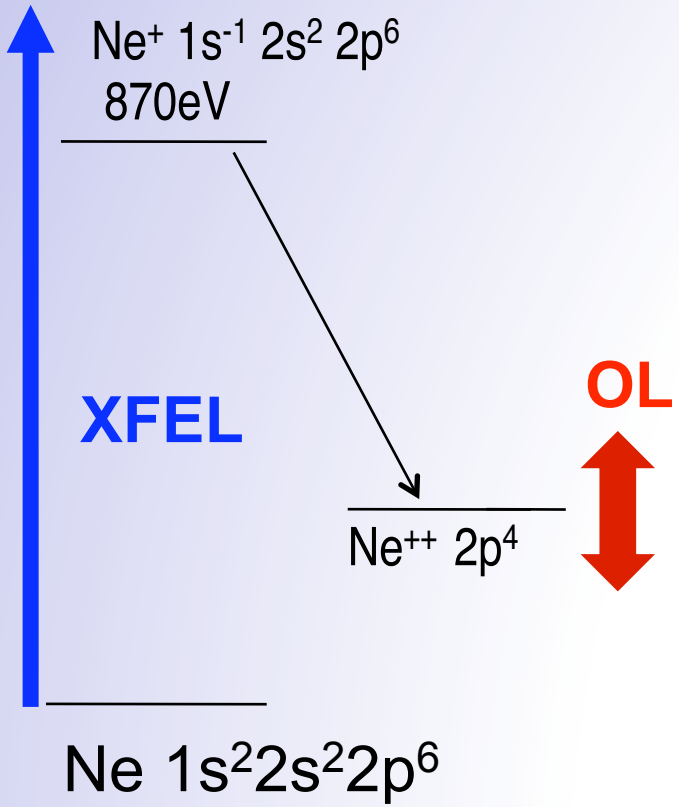




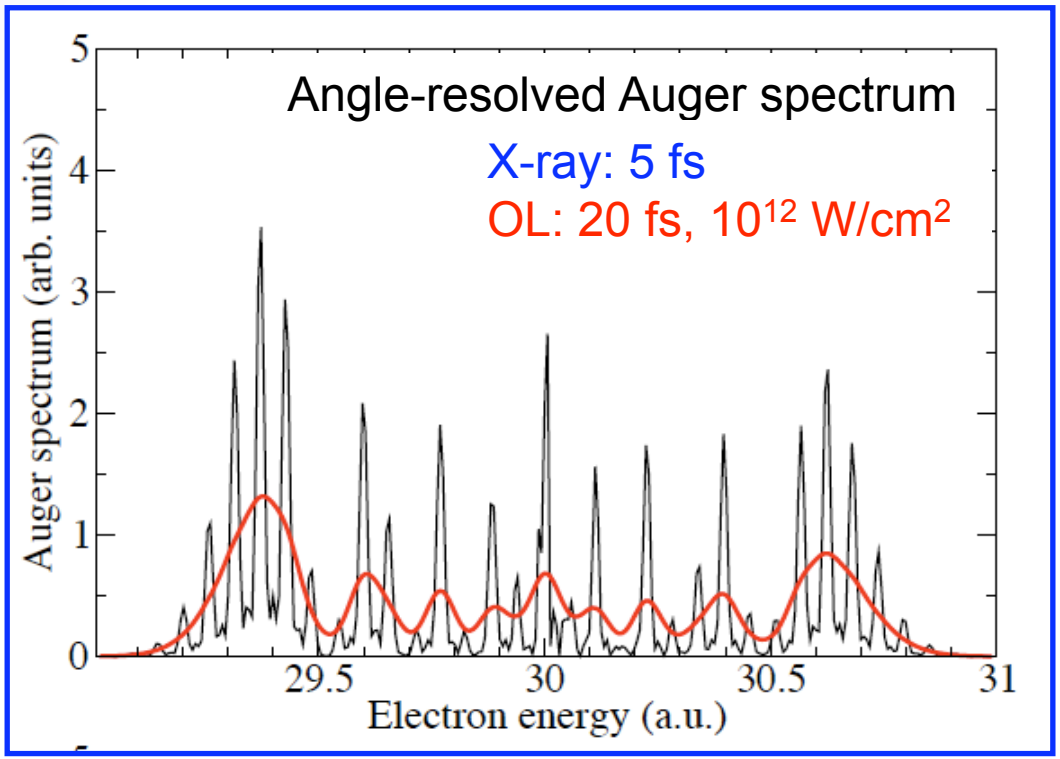
Southworth et al. PRA 52, 1272 (1995)

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!!!

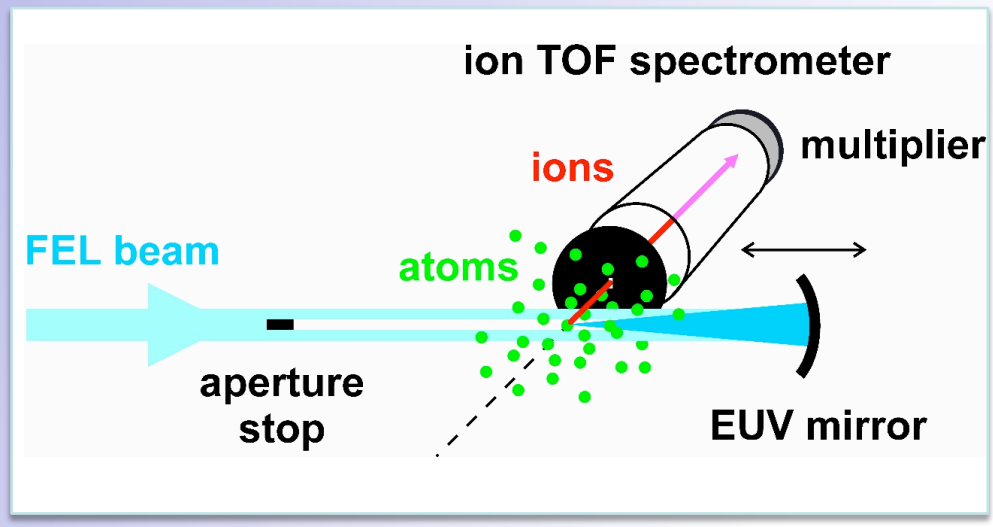
Two-color experiments

Intense XUV field: Multi-photon Ionization

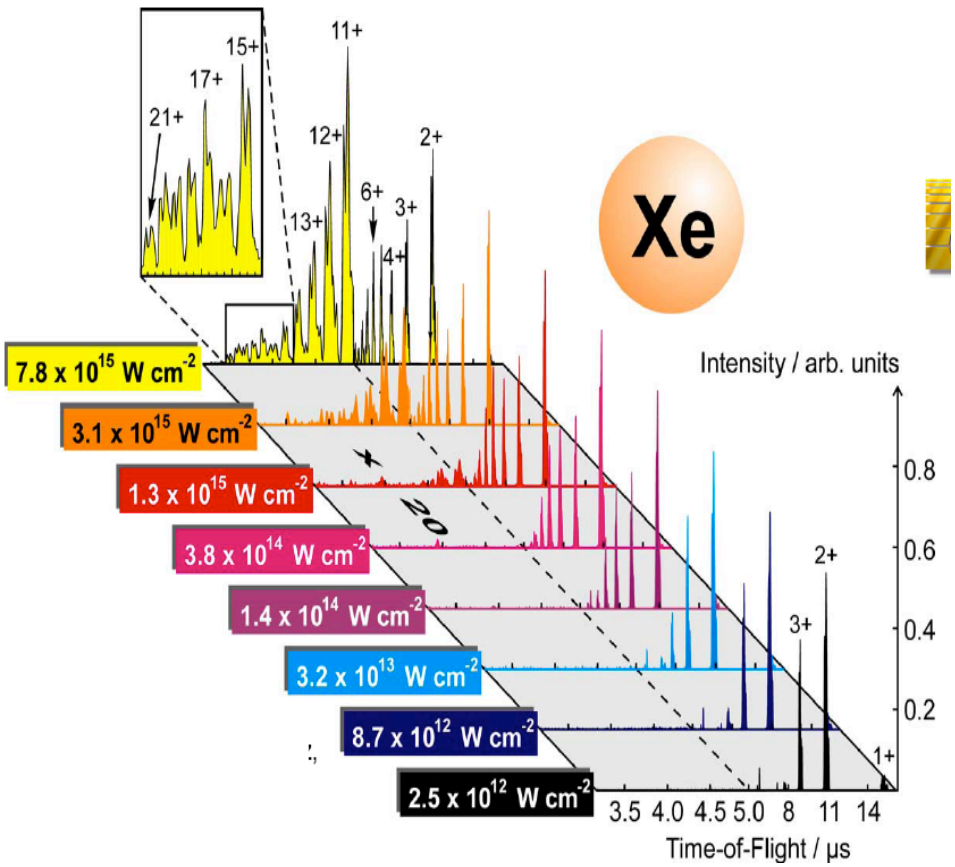
FLASH: Non-linear Processes

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}

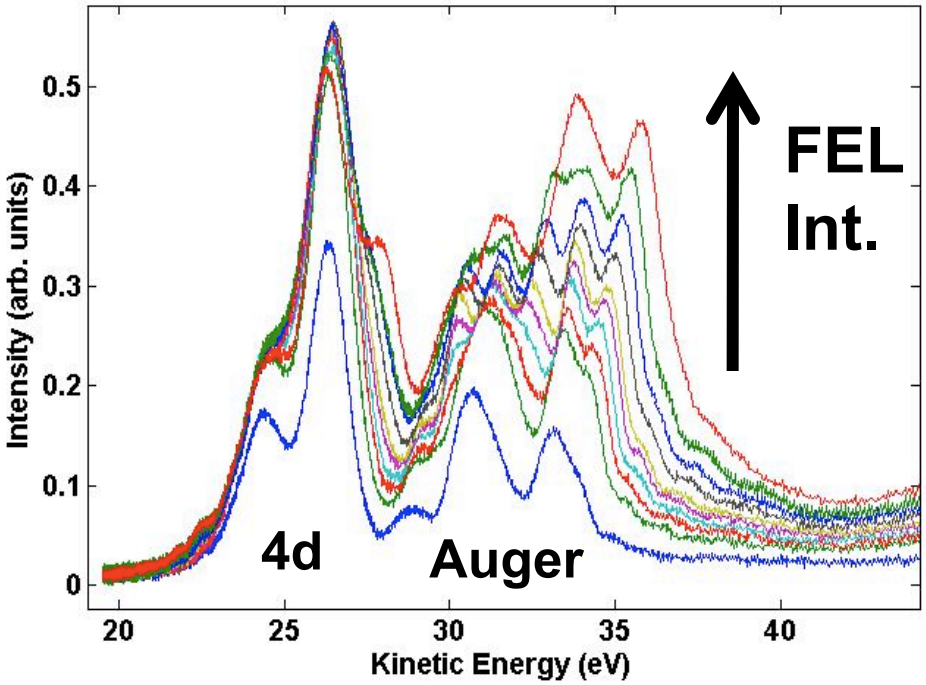
IP(Xe 21+) ≈ 5 keV

Electron Spectroscopy on atomic Xe

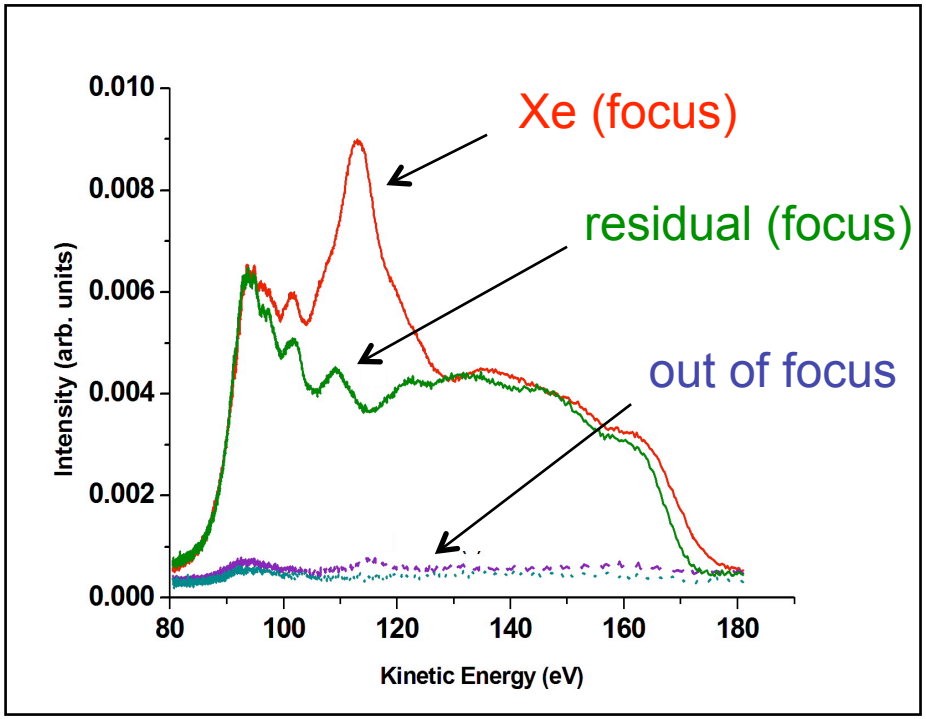
Costello, Düsterer, Meyer, Richter et al.

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

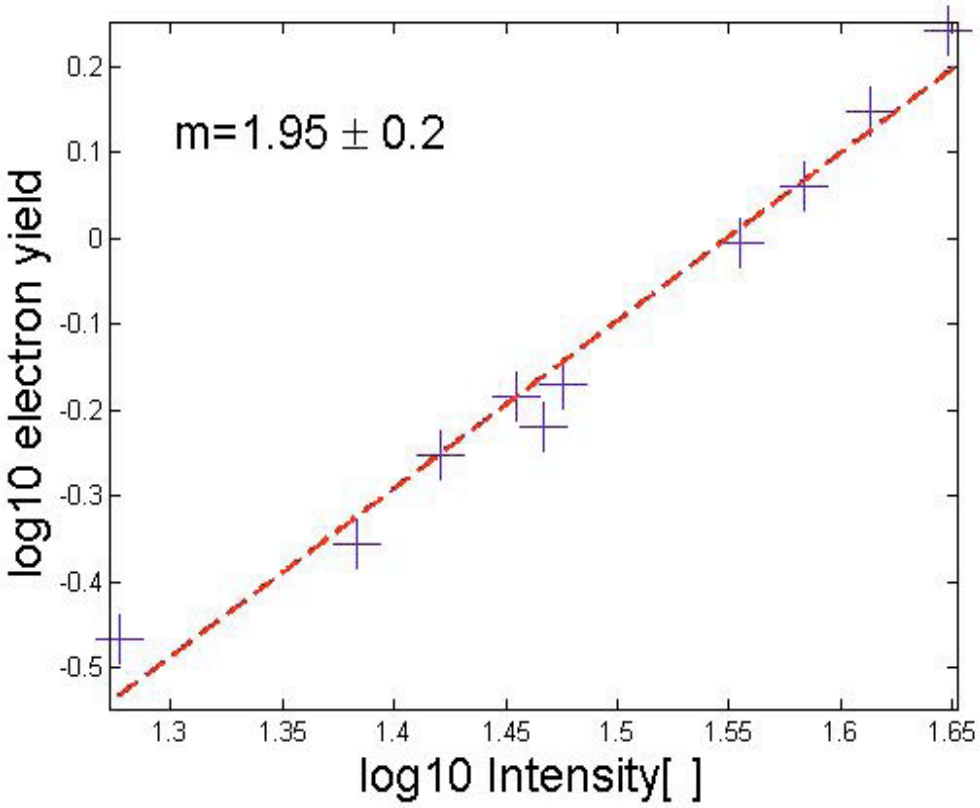
$^2D_{3/2}$ = 69.5 eV

Electron Spectroscopy on atomic Xe

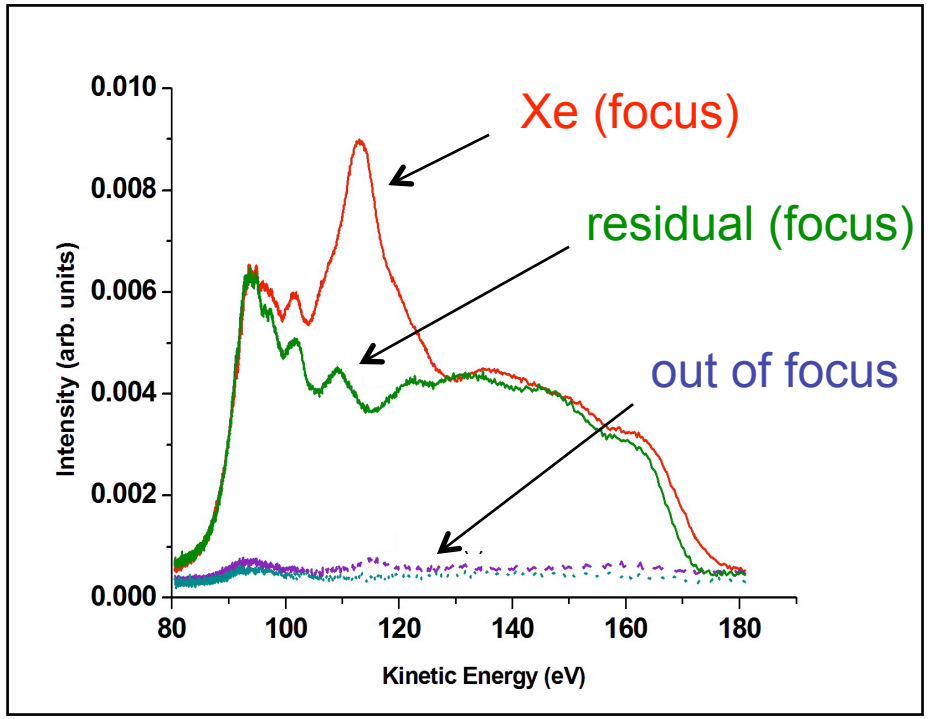
Costello, Düsterer, Meyer, Richter et al.

hν (FEL) 93.3 eV ; ~ 10^{15} W / cm^2

Intensity dependence



Two-photon Process



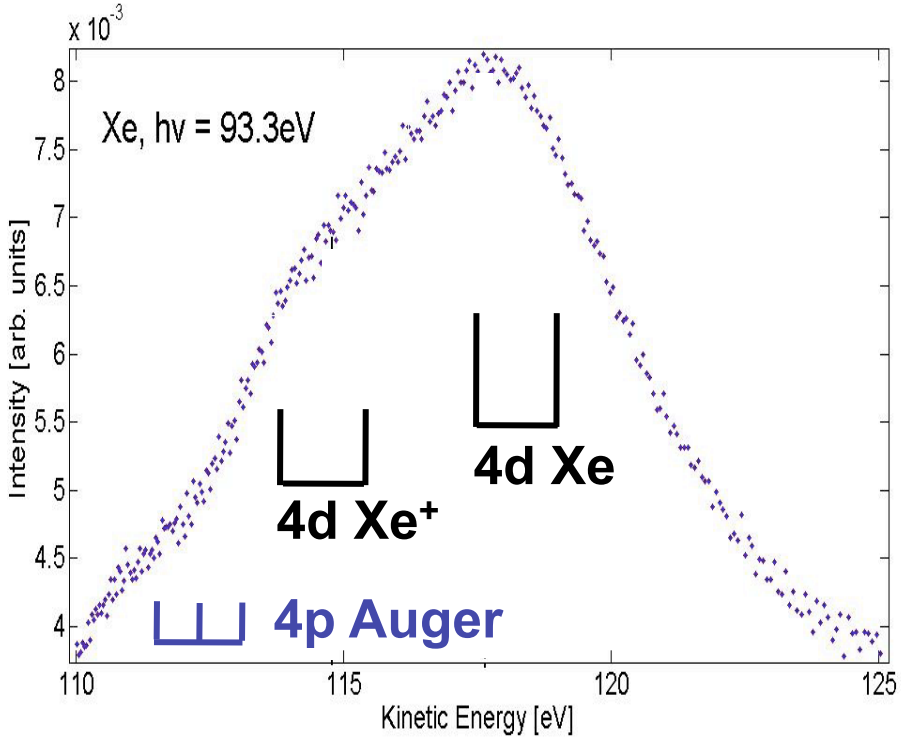
Electron Spectroscopy on atomic Xe

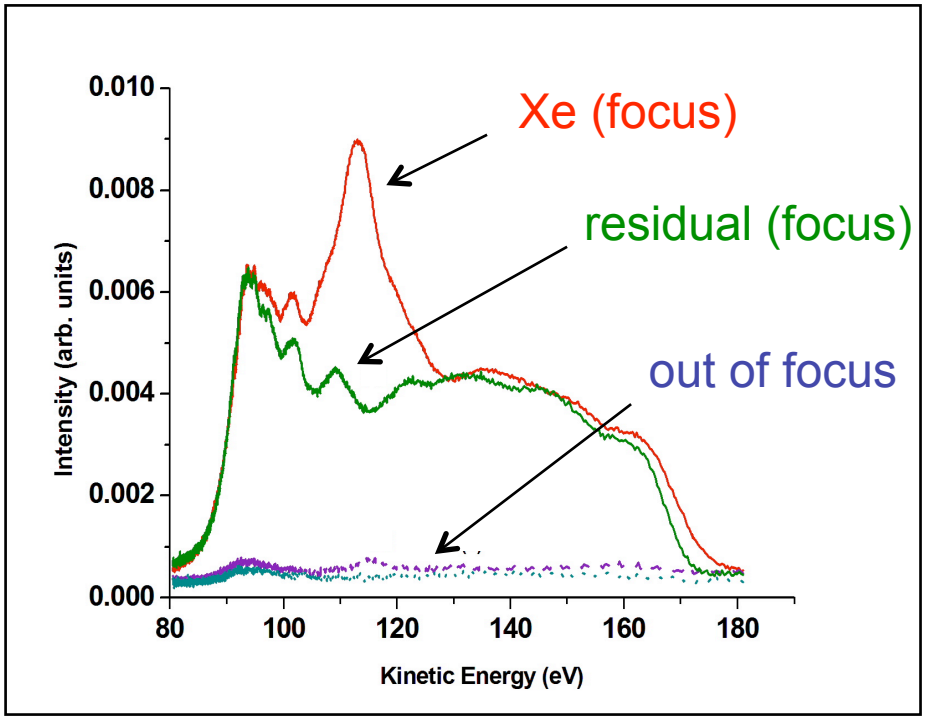
Costello, Düsterer, Meyer, Richter et al.

hν (FEL) 93.3 eV ; ~ 10^{15} W / cm^2

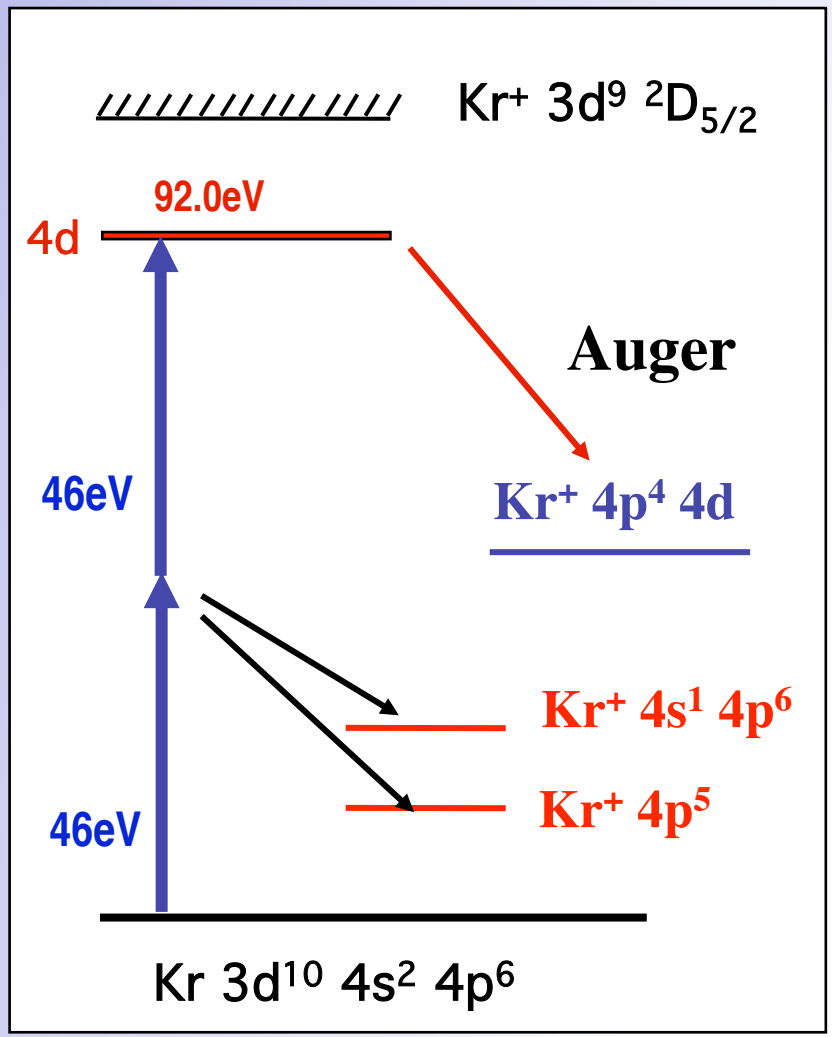
2 x hν (FEL) 93.3 eV

Two-photon Process





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

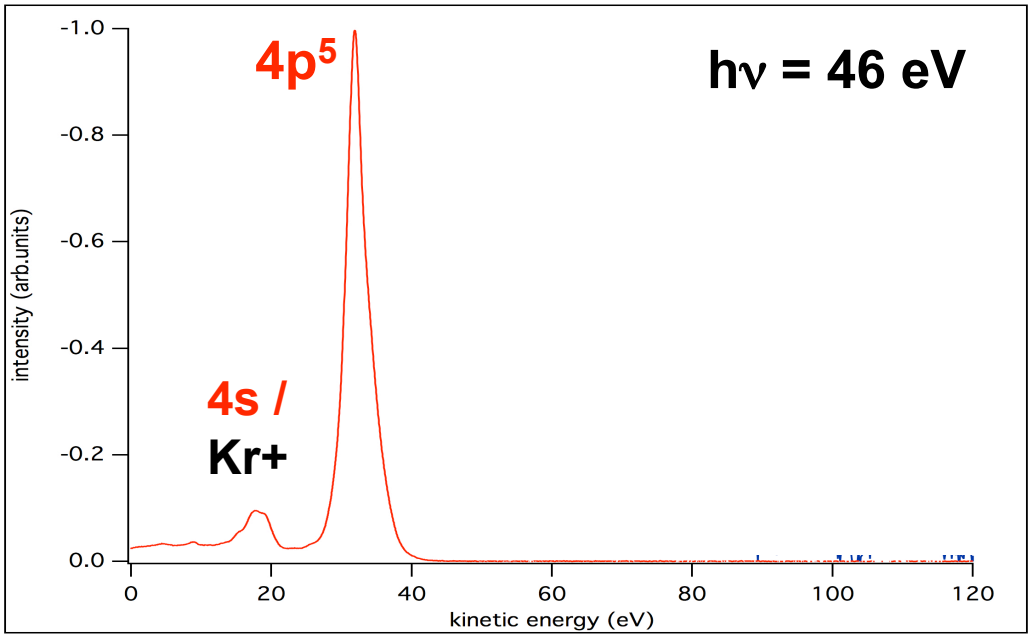


One-photon ionization:

Kr + hν → Kr^+ $4p^5$ / $4s^1$

Two-photon ionization:

Kr + 2 hν → Kr^+ $4p^5$ / $4p^4$4d

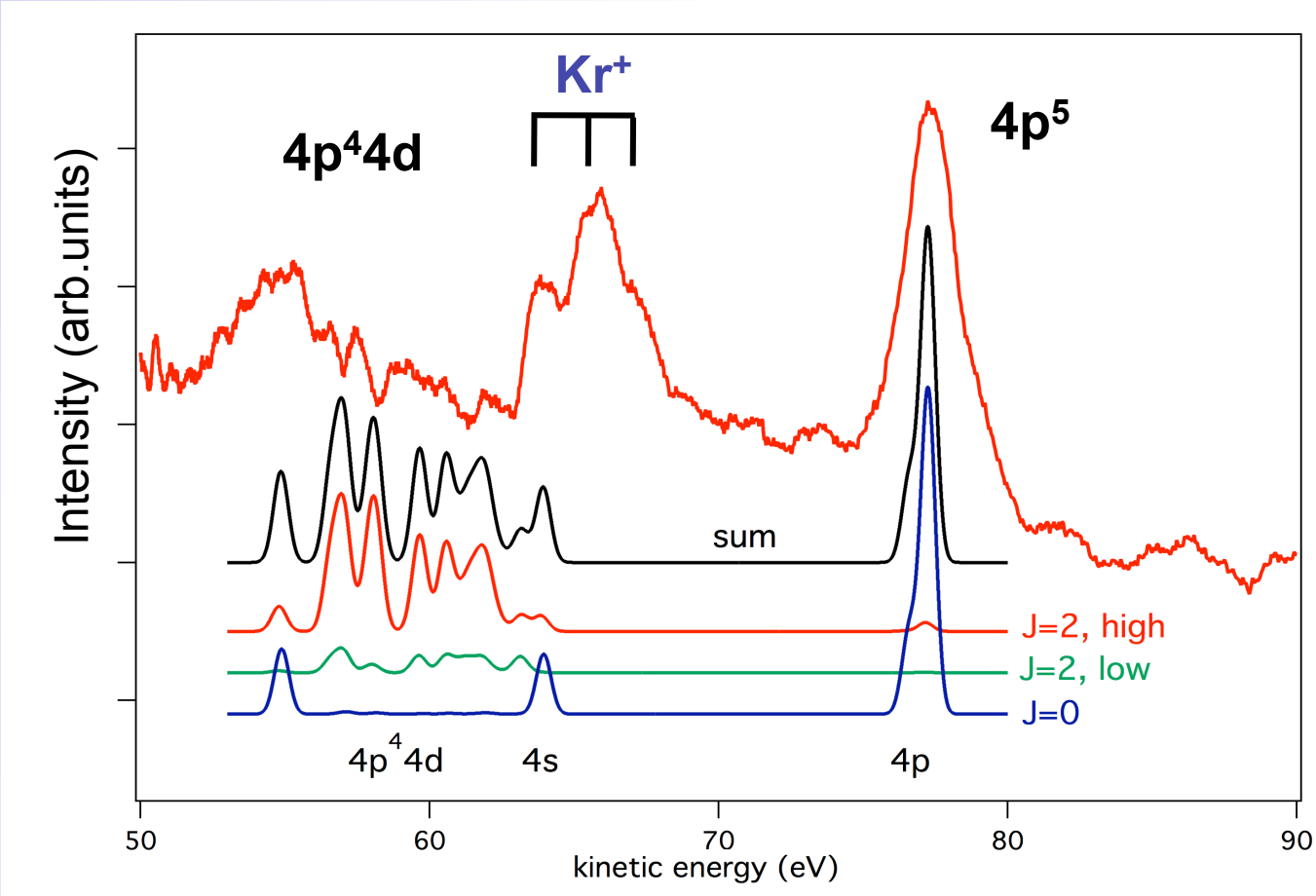


5 μm; >10^{14} W/cm^2

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

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

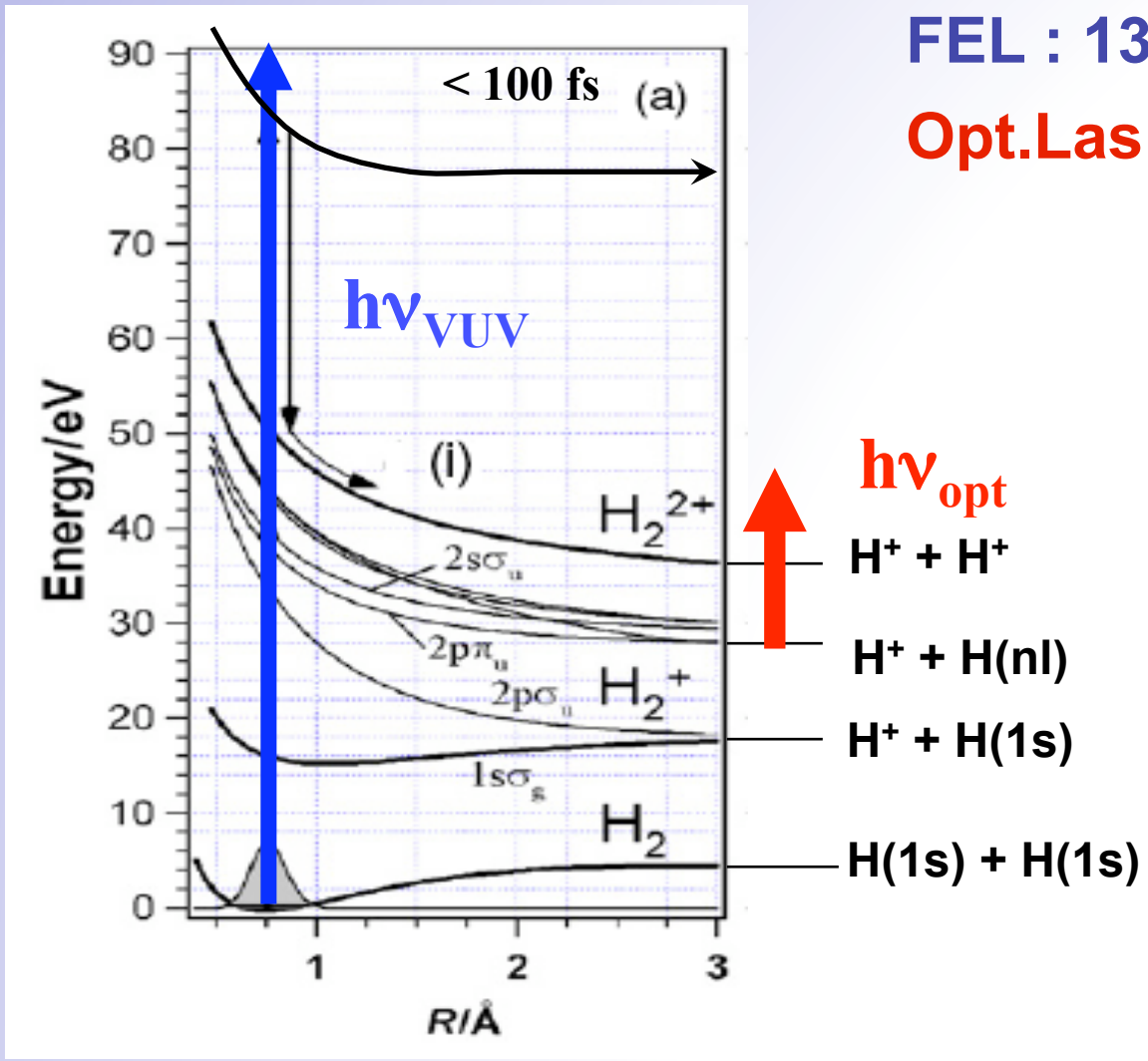
Kr^{**} $3d^9 4s^2 4p^6 4d$ (J=0,2) ---> Kr^+ $3d^{10} 4s^2 4p^4 4d$ + e^-



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

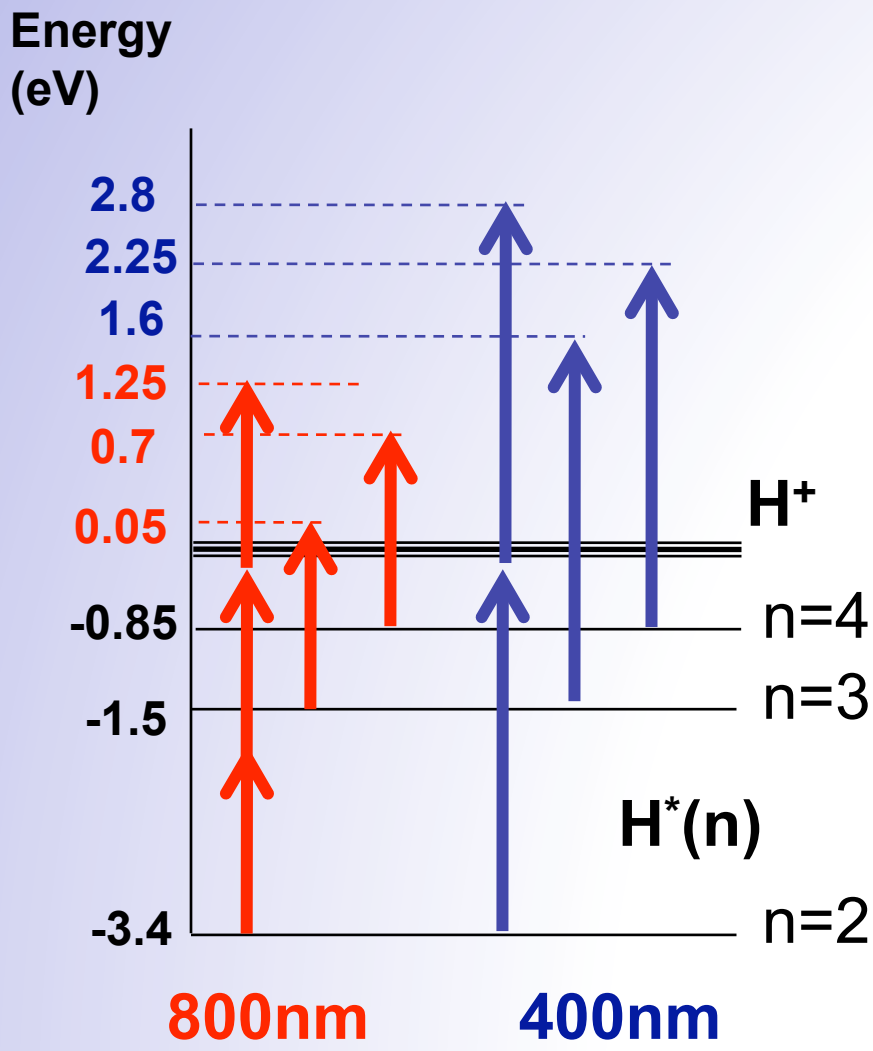
H*(n=3) : E(bind) = 1.5 eV

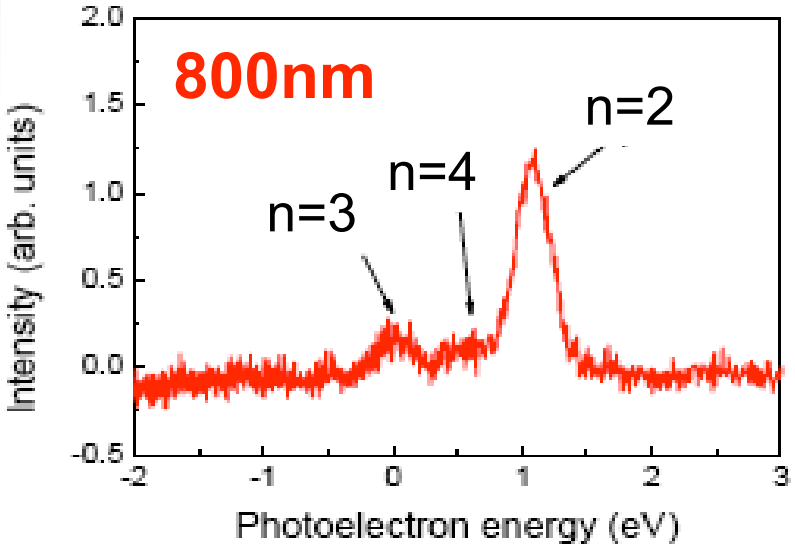
H*(n=4) : E(bind) = 0.8 eV

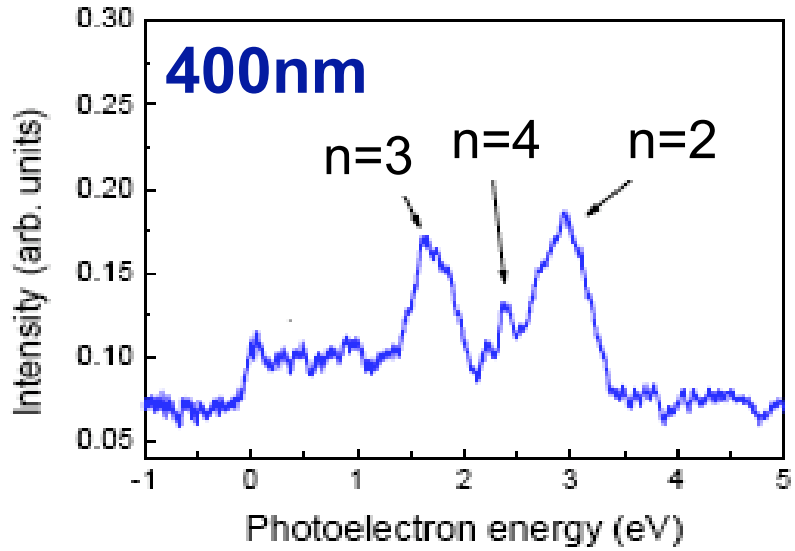
⋮

E (kin.) < 1.5 eV

Photoionization of excited atomic fragments

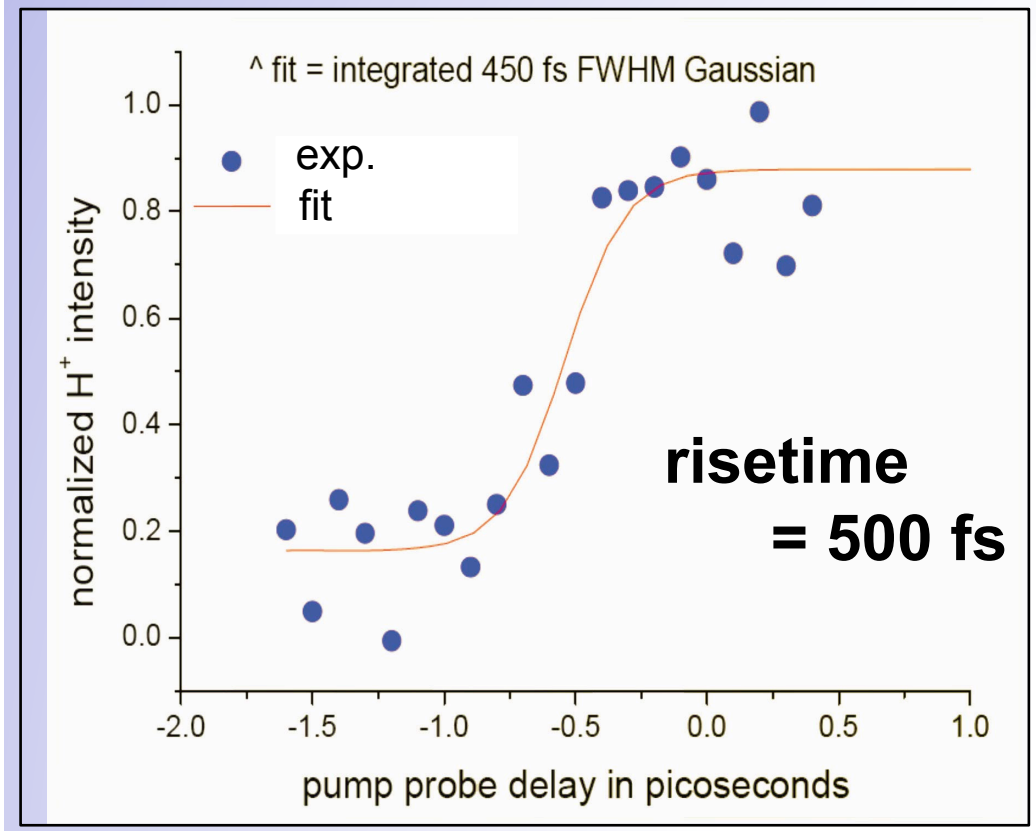




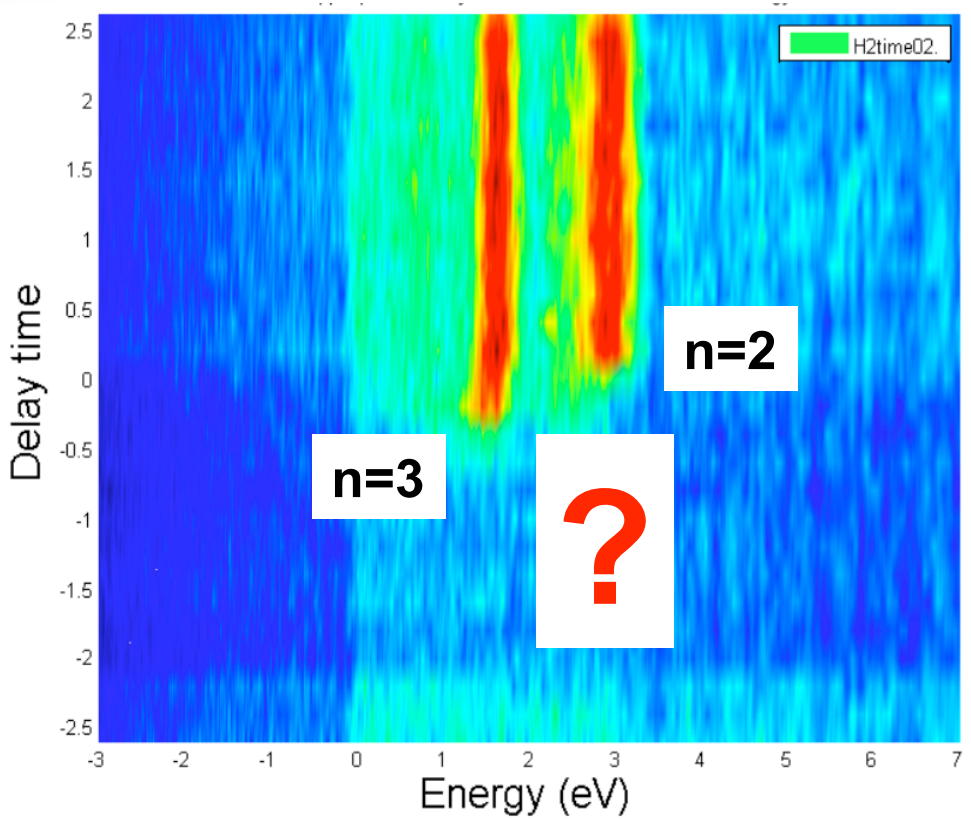


Ultra-fast molecular dissociation

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