One- and Two-Colour Photoionization Experiments at FLASH

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- **LCP-MR (Paris, France)**

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Outline

1 - One-colour experiments (FEL)

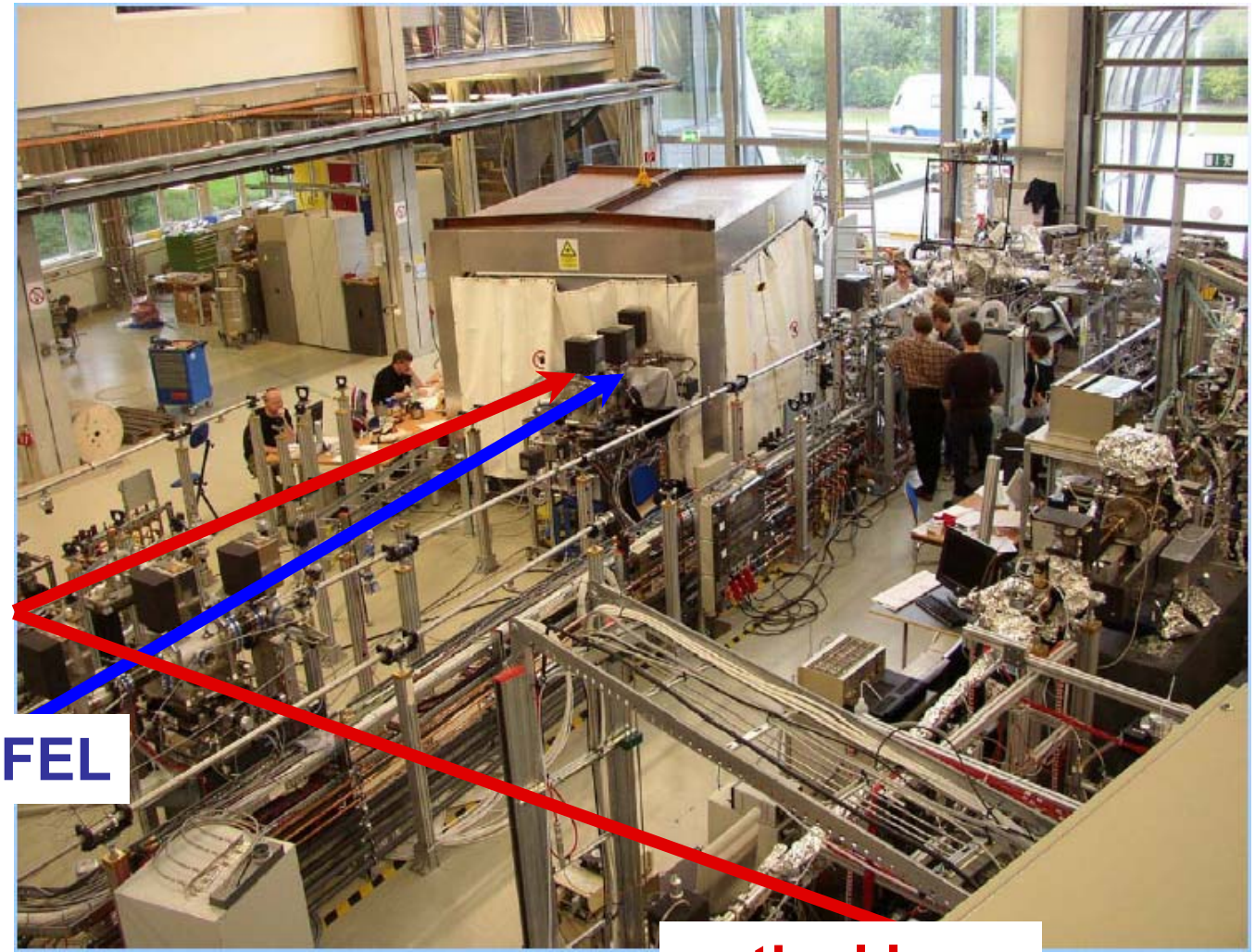
- One-photon ionization (FEL characterization)
- Multi-photon processes (Nonlinear vs. Higher order)

2 - Two-colour experiments (FEL + optical laser)

- Two-photon cross correlation (Temporal characterization)
- Multi-photon ATI (Nonlinear effects)
- Pump-probe applications
  - polarization dependence
  - coupling of autoionization states
  - molecular fragmentation

VUV-FEL Experimental Hall - BL 2

(August 2005, May/June 2006)



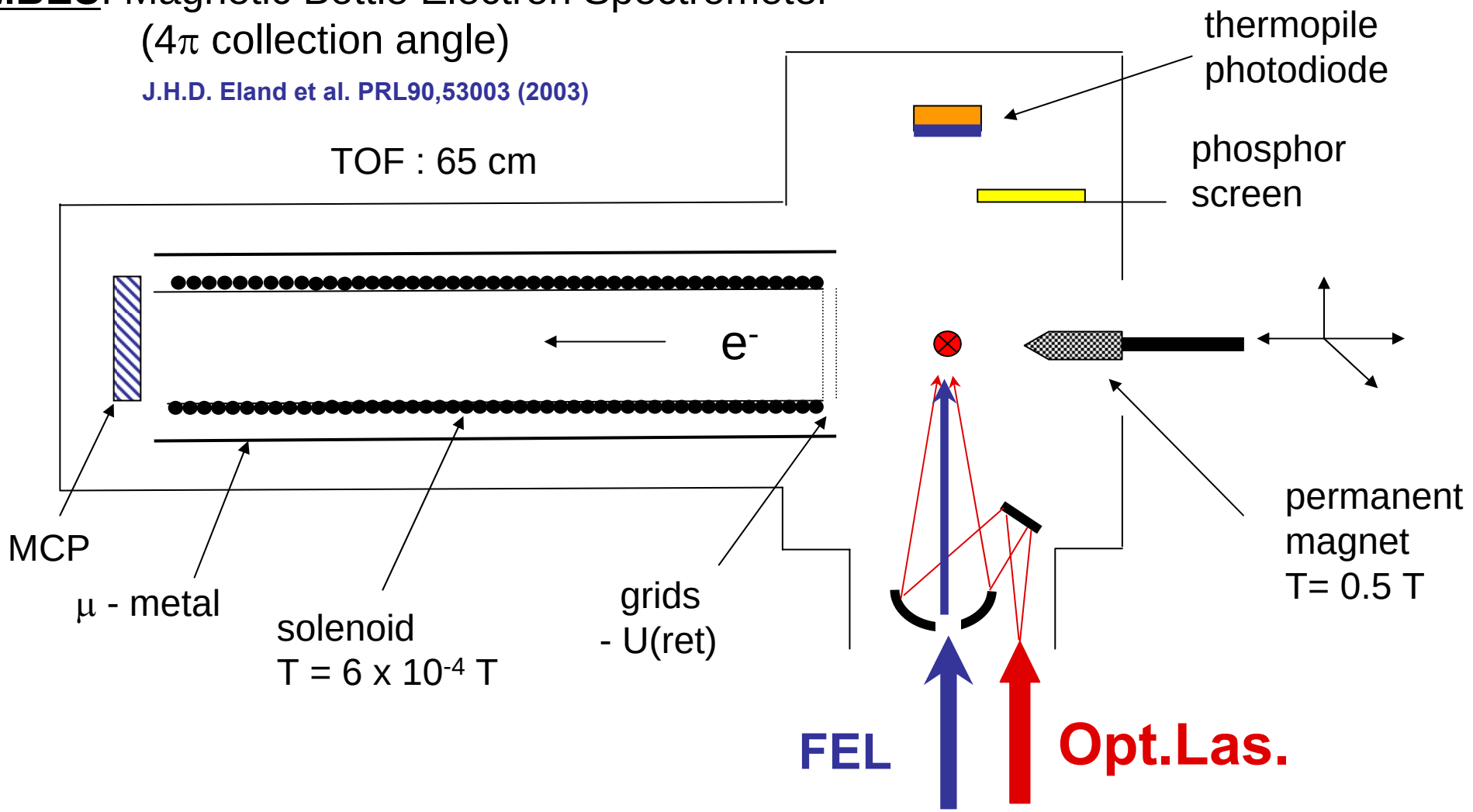
FEL - Parameters

- 32.2 nm (2005)
- 25.5, 13.8 nm (2006)
- 30 - 50 fs
- 1 - 10 µJ (average)
- 2 - 5 Hz (macro)
- ≤ 30 bunches
- 1 µs separation
- B.w. : 0.5 - 1.0 %

Experimental set-up

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

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

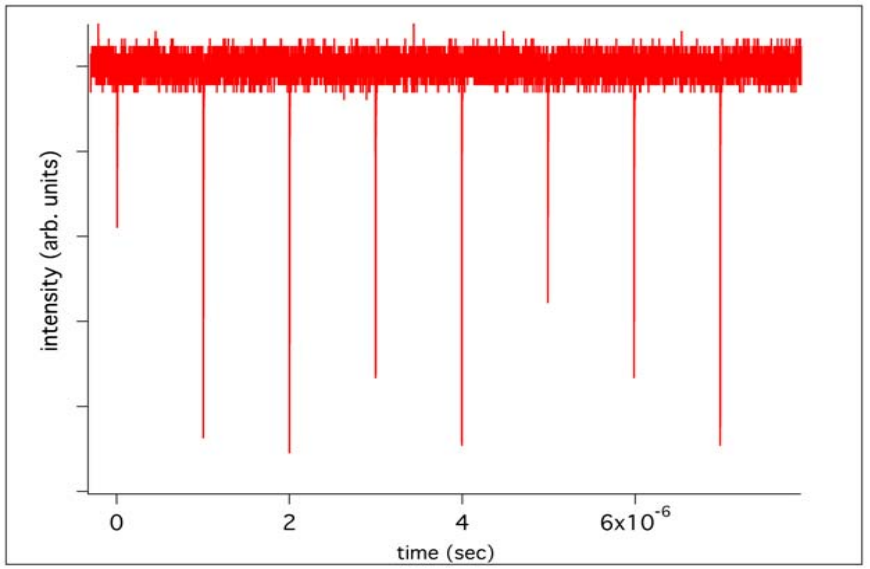


One - Colour / One - Photon

Photoionization of rare gazes

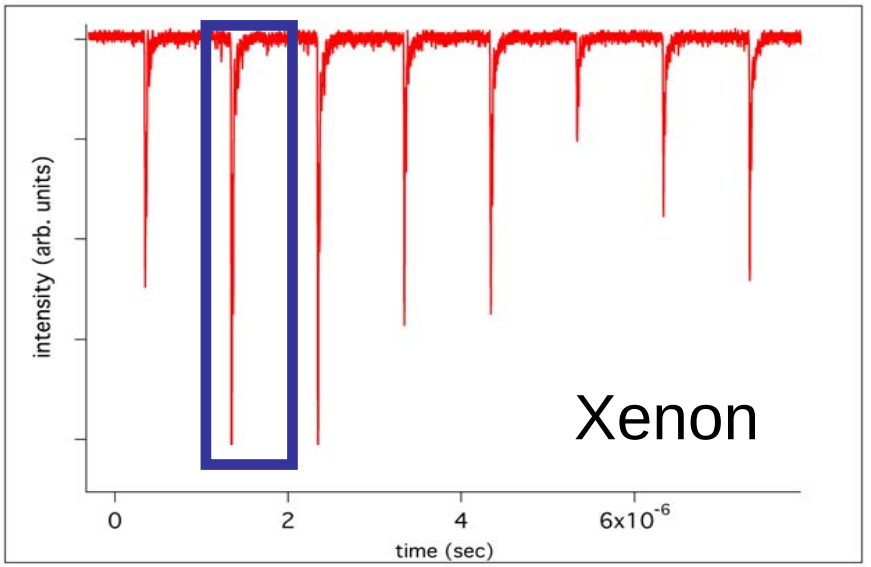
One-photon ionization of Xenon

thermopile - signal



- ΔT (bunch) = 1 μsec
- relative intensity
- definition of T_0

photoelectron - signal

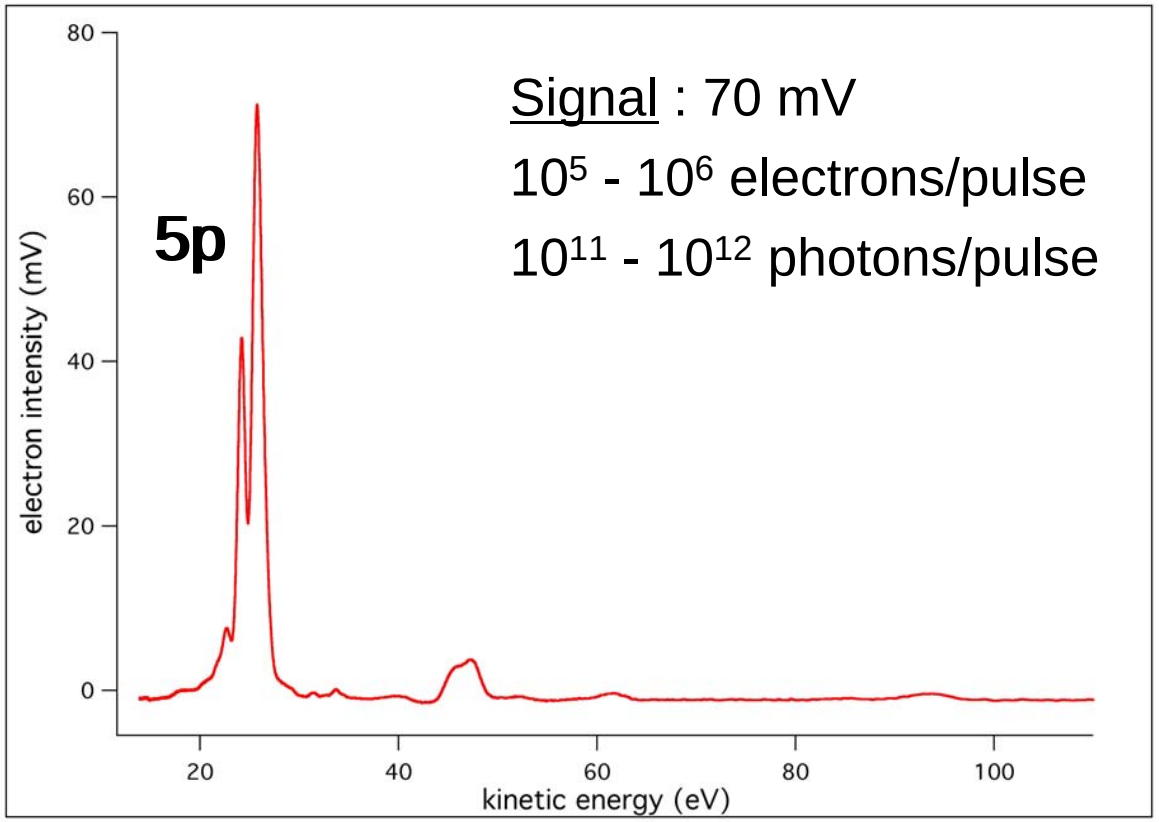


- photoionization of Xe
- time-of flight
- single-shot spectra

One-photon ionization of Xe

U(ret) = -15V

Xenon



hν (FEL) = 38.5 eV

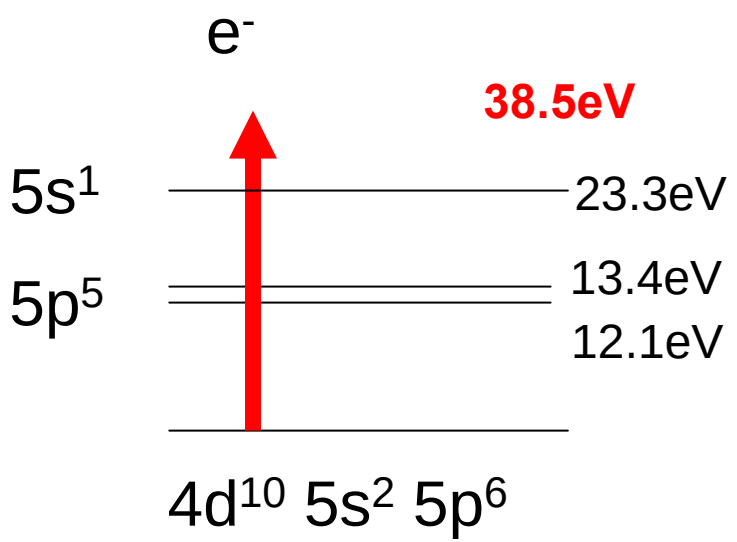
- single shot analysis

- nonlinear effects

- space charge

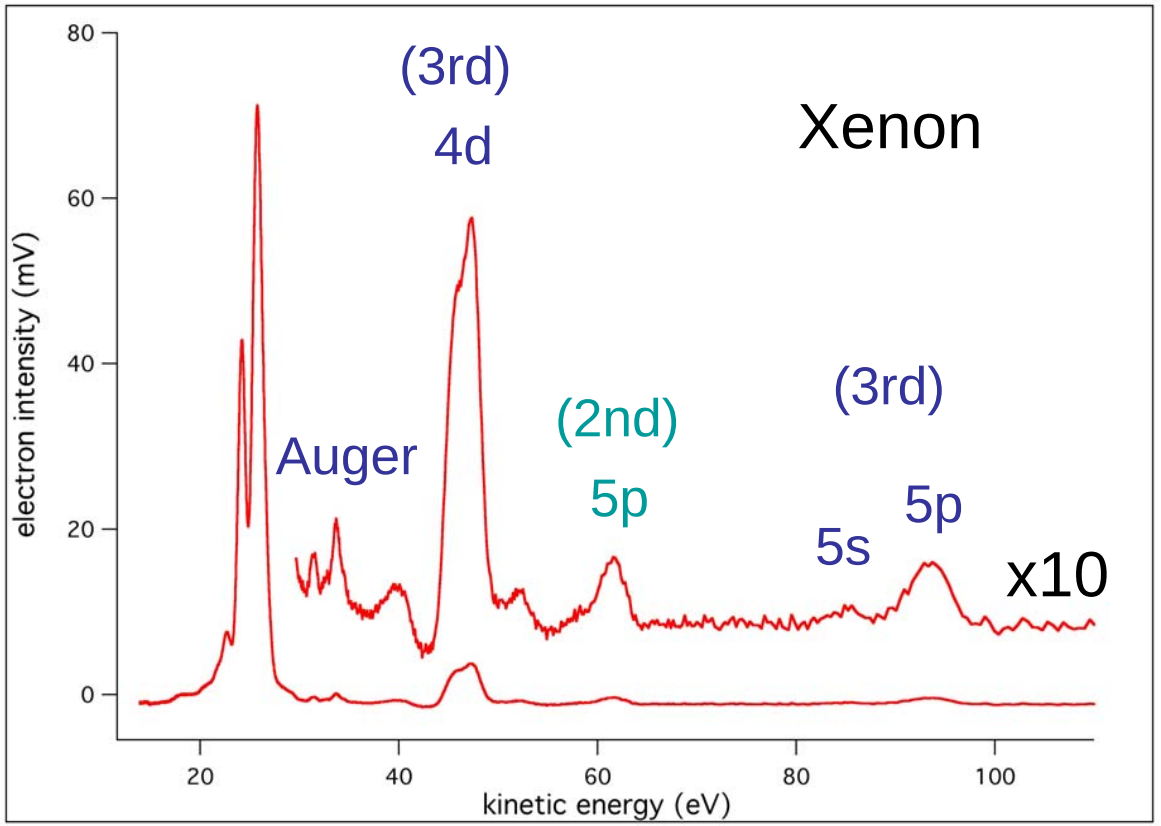
- ΔE = 300 meV

ΔE(max.) = 50 meV

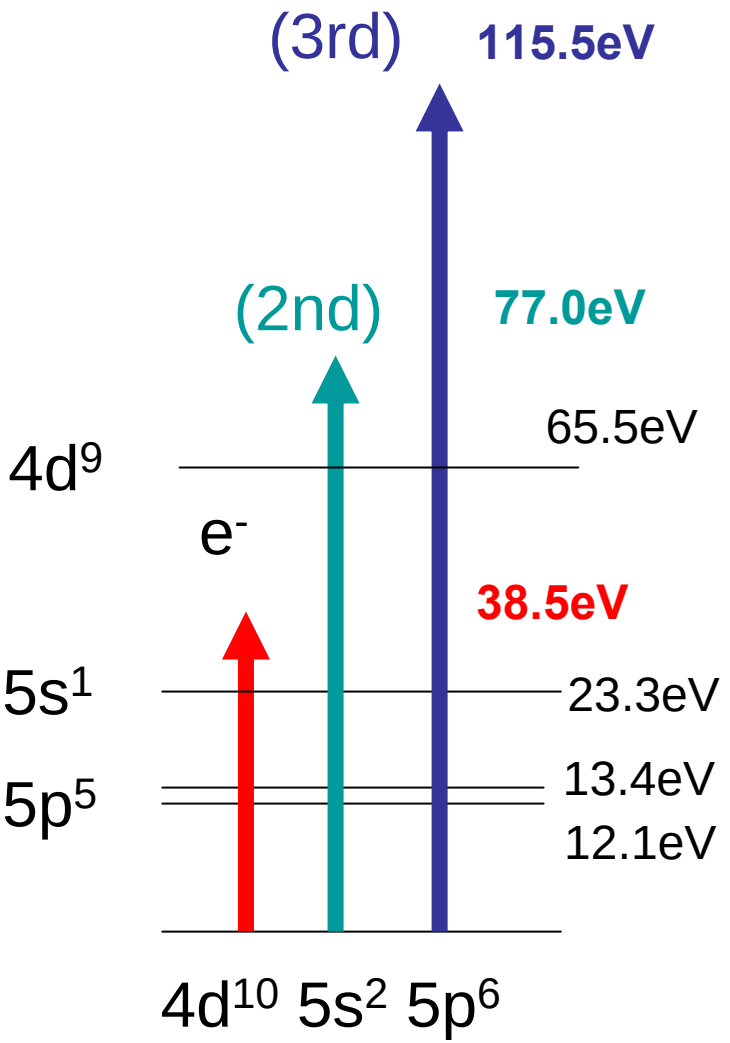


One-photon ionization of Xe

U(ret) = -15V



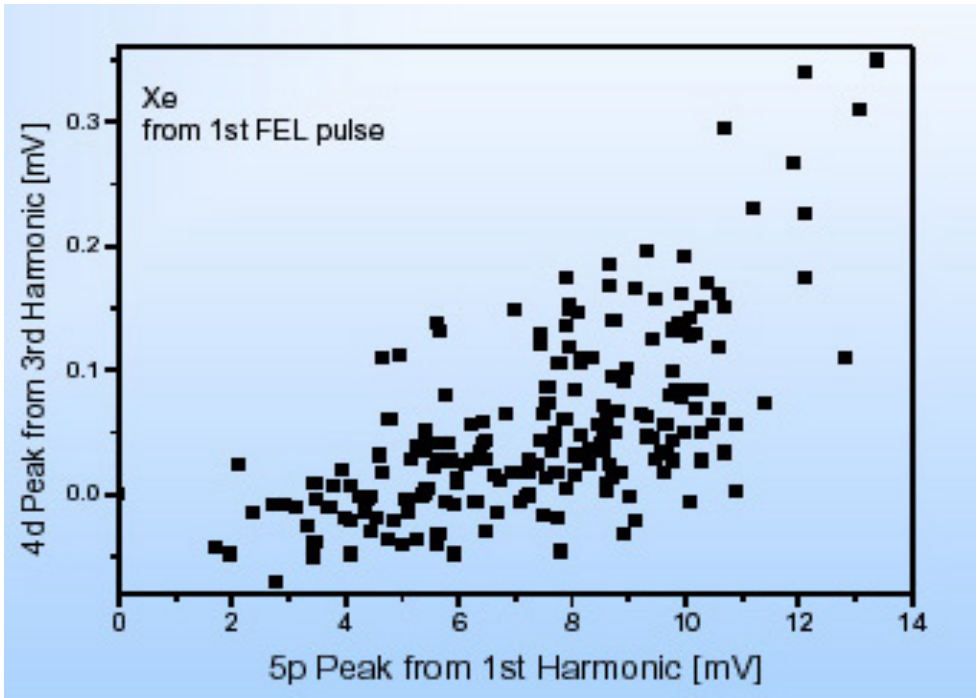
hν (FEL) = 38.5 eV

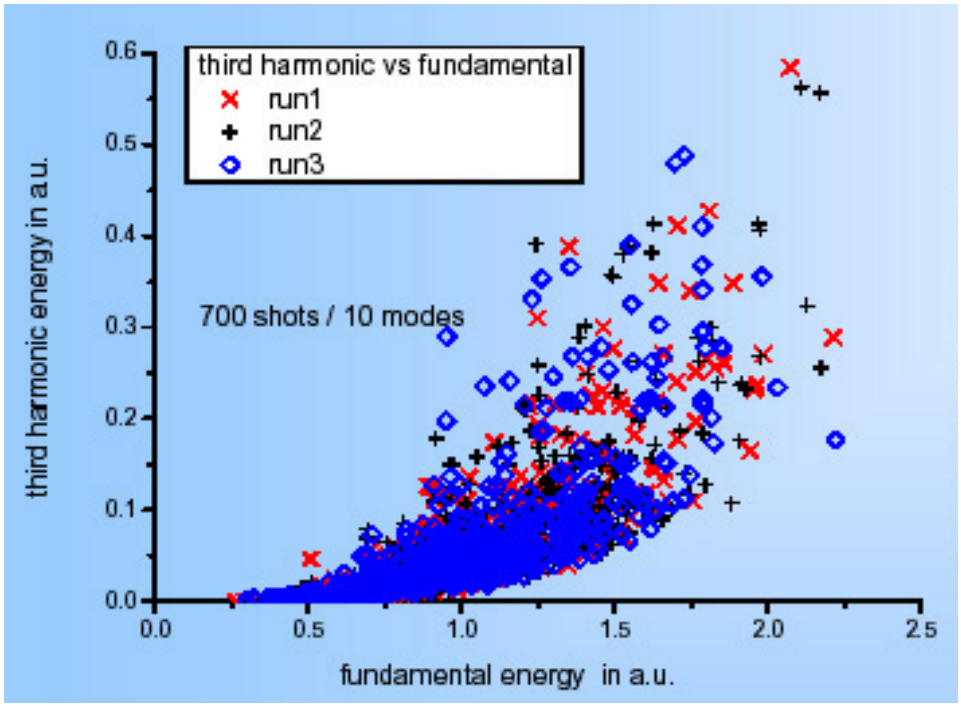


Statistical behavior of higher harmonics

Single shot correlation fundamental - third harmonic

S. Düsterer et al. Opt.Lett. 31, 1750 (2006)





Experiment average 3 μJ
max. 10 μJ

Simulations (G. Geloni, E. Saldin)
10 modes

Higher harmonics of FEL radiation

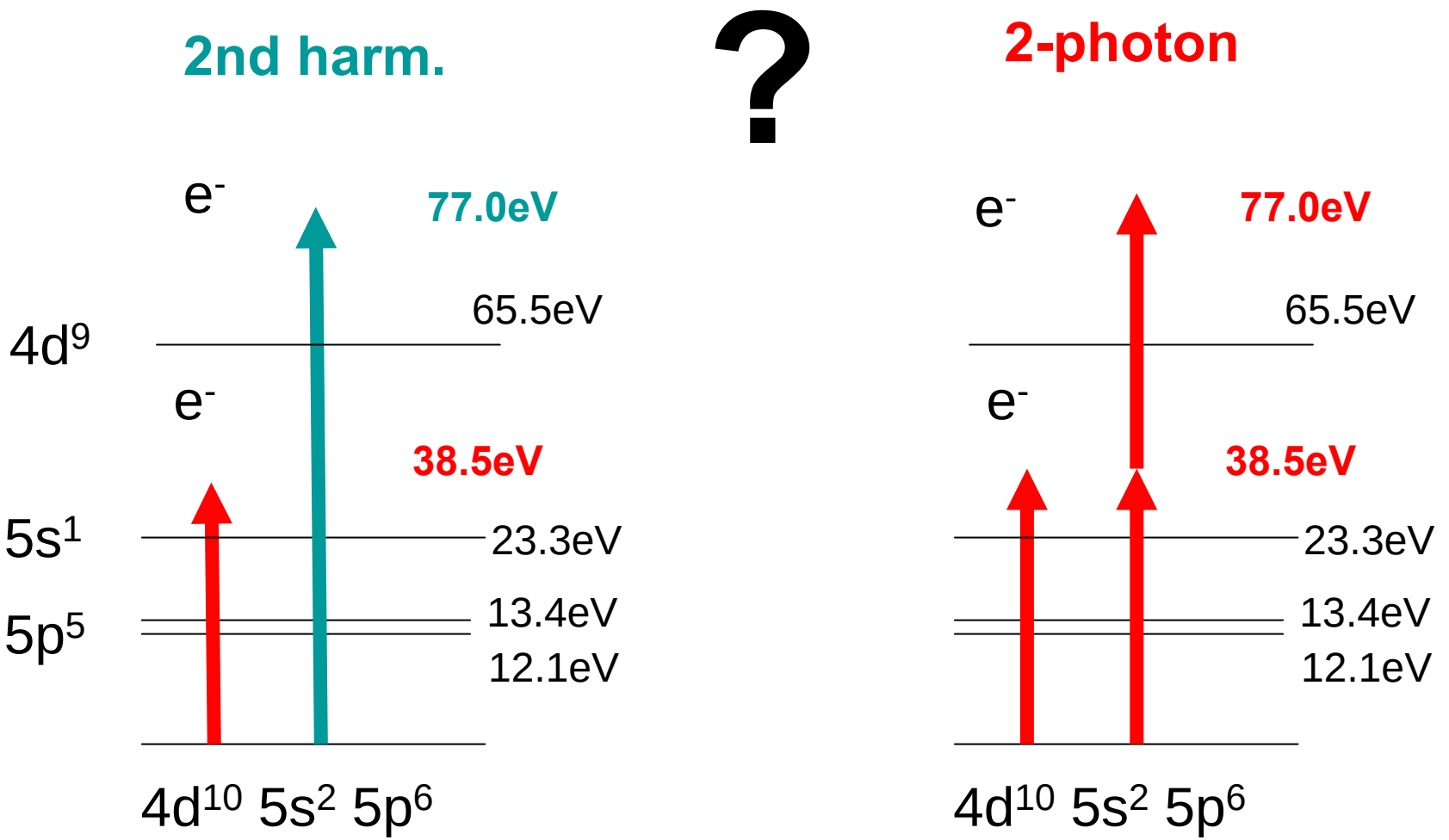
32.2 nm
August 2005

Measured harmonic ratios

gas	2nd / 1st	3rd / 1st	I (FEL)
Xe	0.6 (2) %	0.5 (2) %	5 - 7 μJ
	0.3 (2) %	0.2 (2) %	3 μJ
Ar	0.3 (2) %	0.35 (20) %	3 μJ
He	0.5 (2) %	0.85 (2) %	5 - 7 μJ

average over 254 single-shot spectra

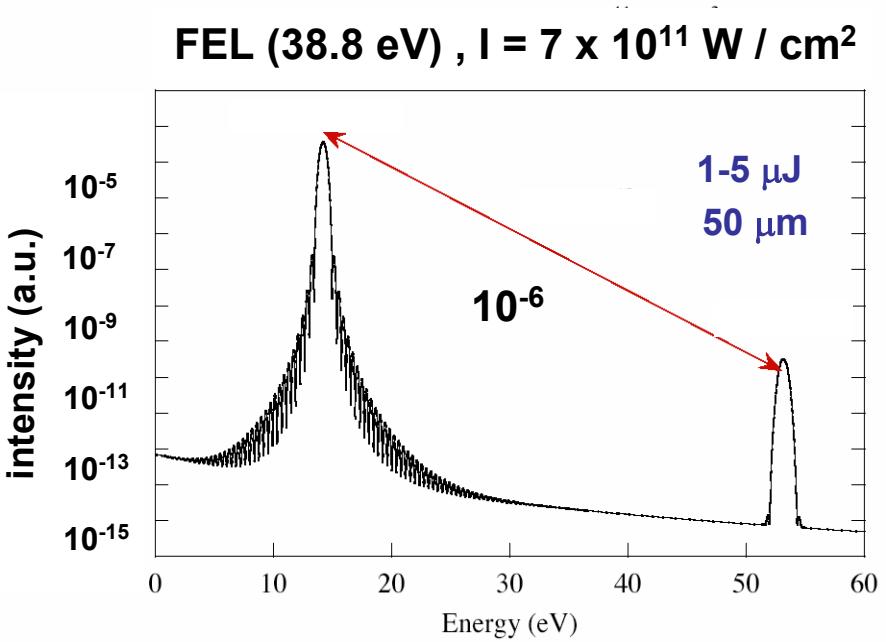
Higher harmonics vs. multi-photon



Higher harmonics vs. multi-photon

Harmonics (August 2005):

- similar values for He, Ar, Xe
- observed in focused and unfocused beam
- observed in the spectral distribution
- TDSE calculations
  (R. Taïeb, A. Maquet, LCP-MR, Paris)



Multi-photon (August 2006):

- FEL pulse energy **> 50 μJ**
- focus size **30 μm**
- FEL intensity **> 3 x 10^{13} W/cm^2**

Non-linear processes:

Two-photon double-ionization

Two-photon double-ionization in He

Lambropoulos, Nikolopoulos, Makris, Phys.Rev. A 72, 013410 (2005)

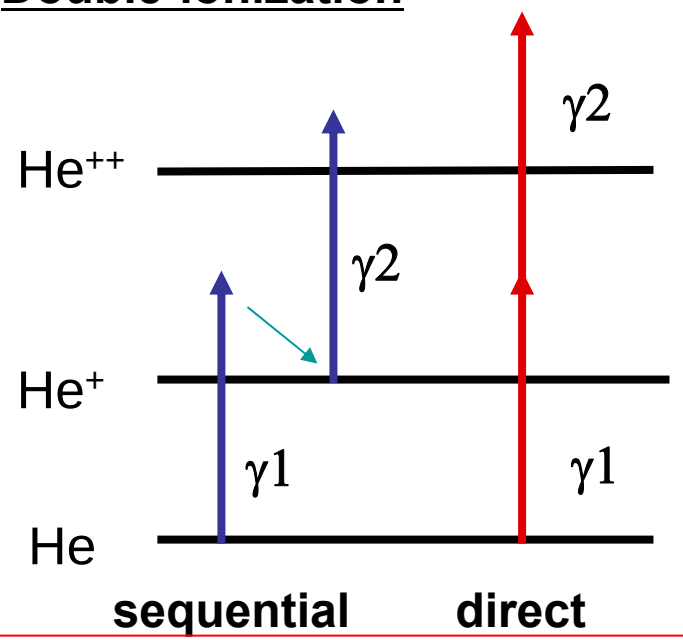
Theory: $h\nu = 45$ eV

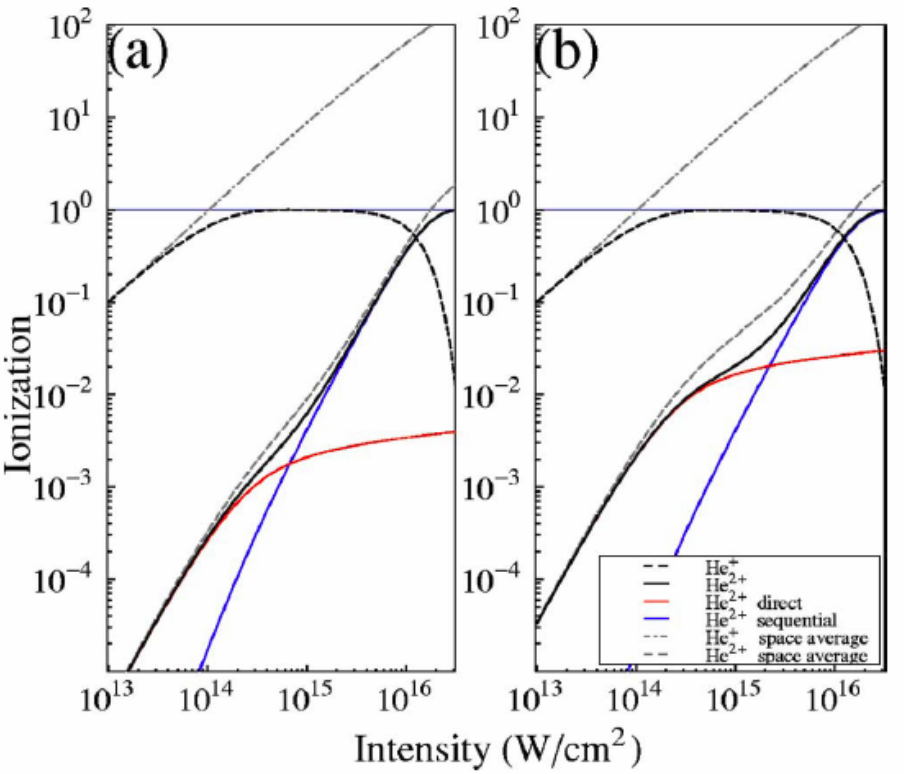
$I = 10^{13} - 10^{16}$ W/cm^2

direct

sequential

Double-ionization





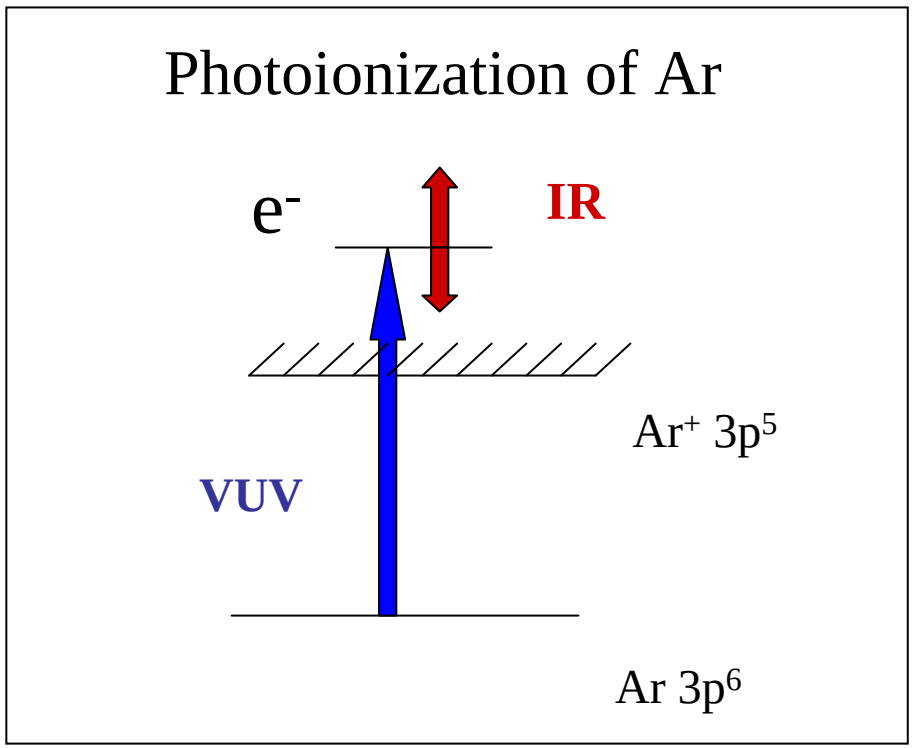
Two - Colour / Two - Photon

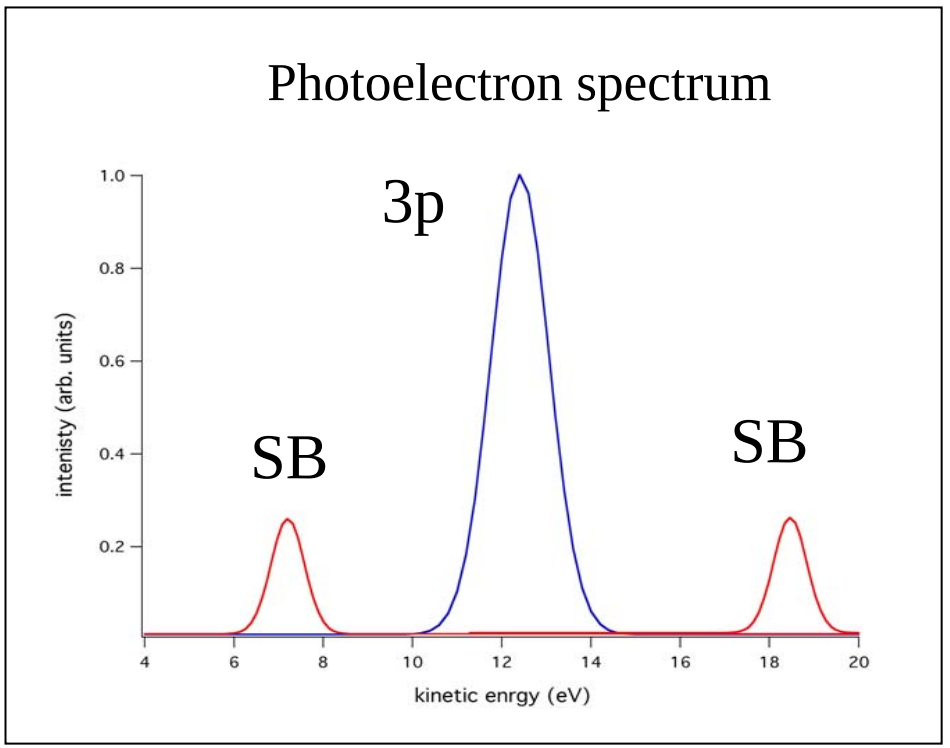
Photoionization of dressed atoms

Two-photon (two-color) ionization

General idea: photoionization in a strong field (dressed atoms)

E.S. Toma et al.
Phys. Rev. A 62, 061801 (2000)

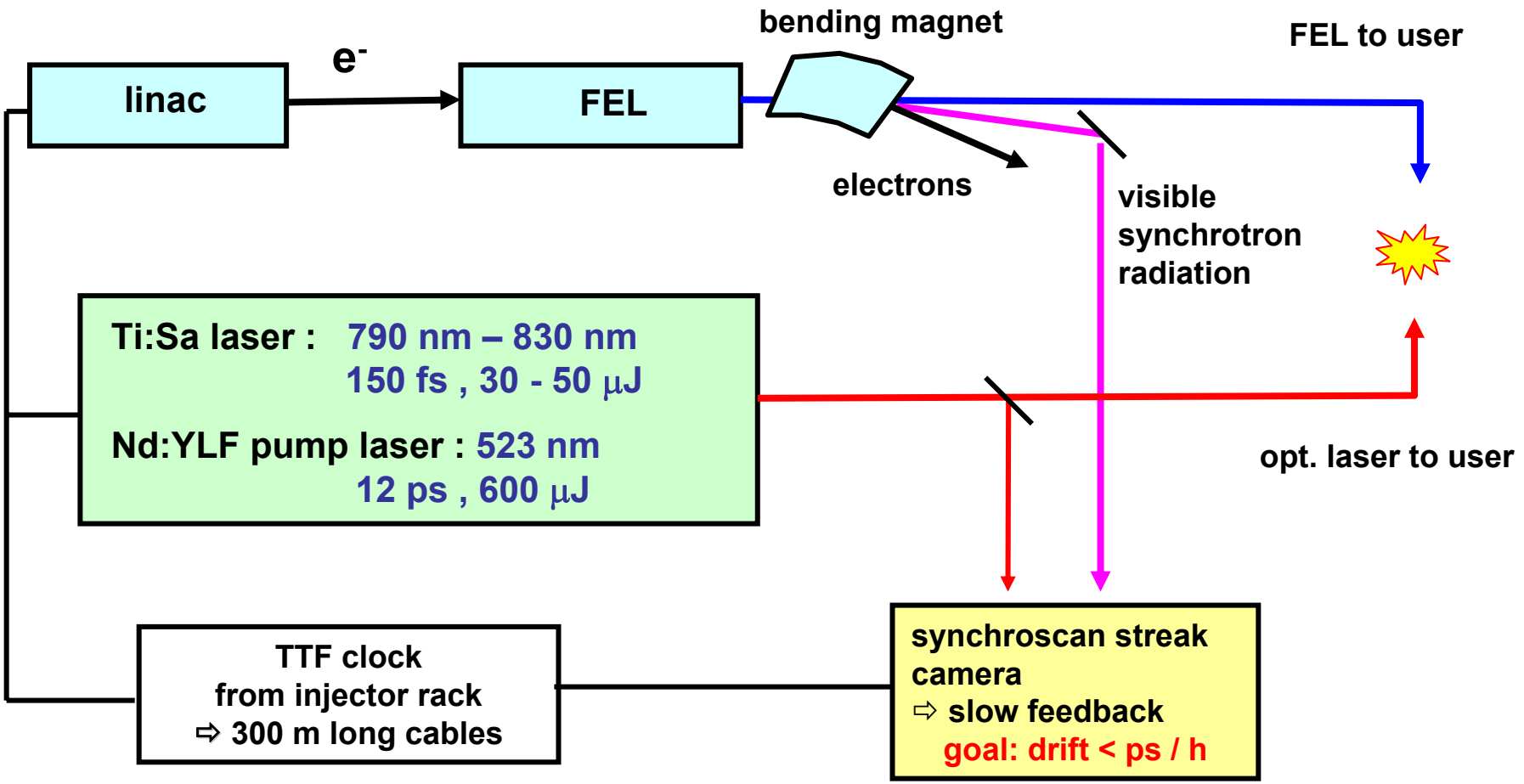




Ti:Saph: 800nm, 40fs, 1 kHz
dressing beam: up to 50μJ --> 13 TW/cm^2

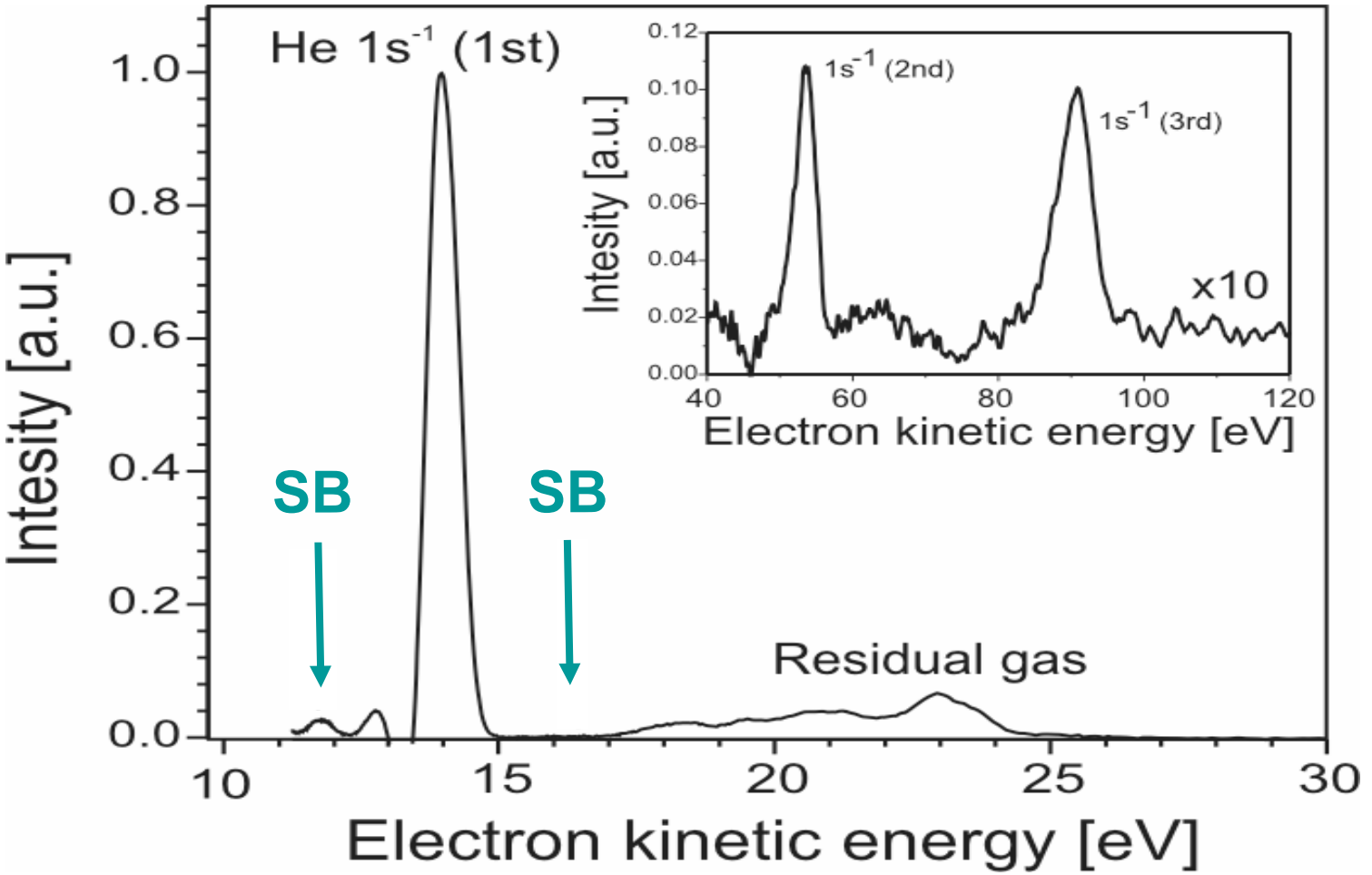
Sideband intensity very sensitive to temporal overlap

FEL + Optical Laser



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

One-photon ionization of He

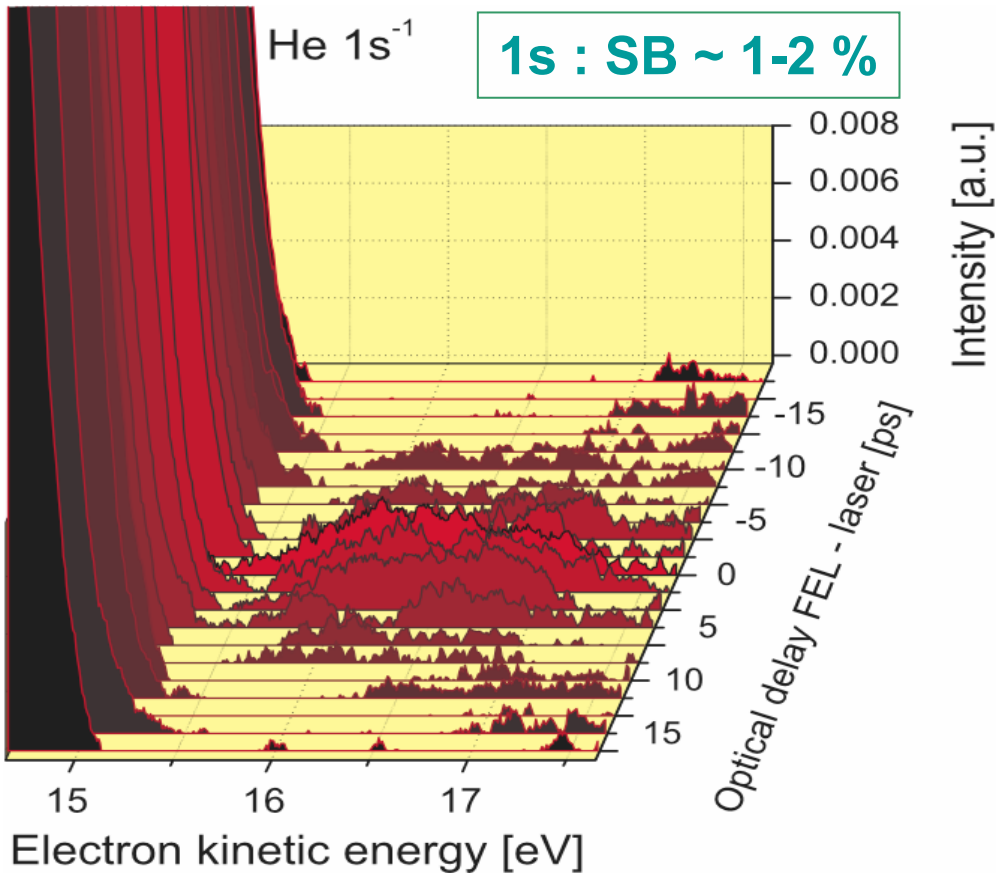


hν (FEL) = 38.5 eV

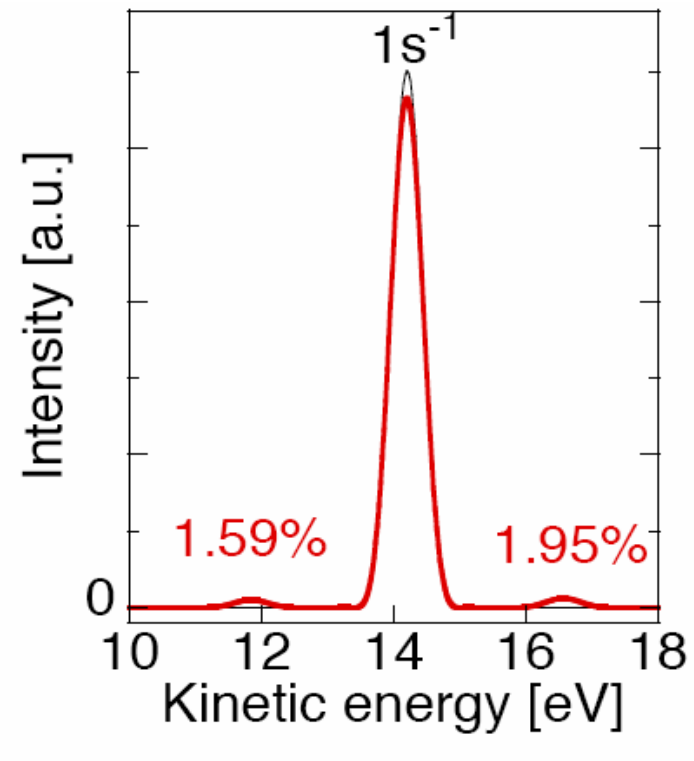
Two-photon ionization of He

FEL: 32 nm, 1 - 3 μJ, 50 μm focus, 50 fs, ~ 10^{12} W/cm^2

Laser : 523 nm, 250 μJ, 50 μm focus, 12 ps, ~ 10^{11} W/cm^2



TDSE calculation (R. Taïeb, A. Maquet)

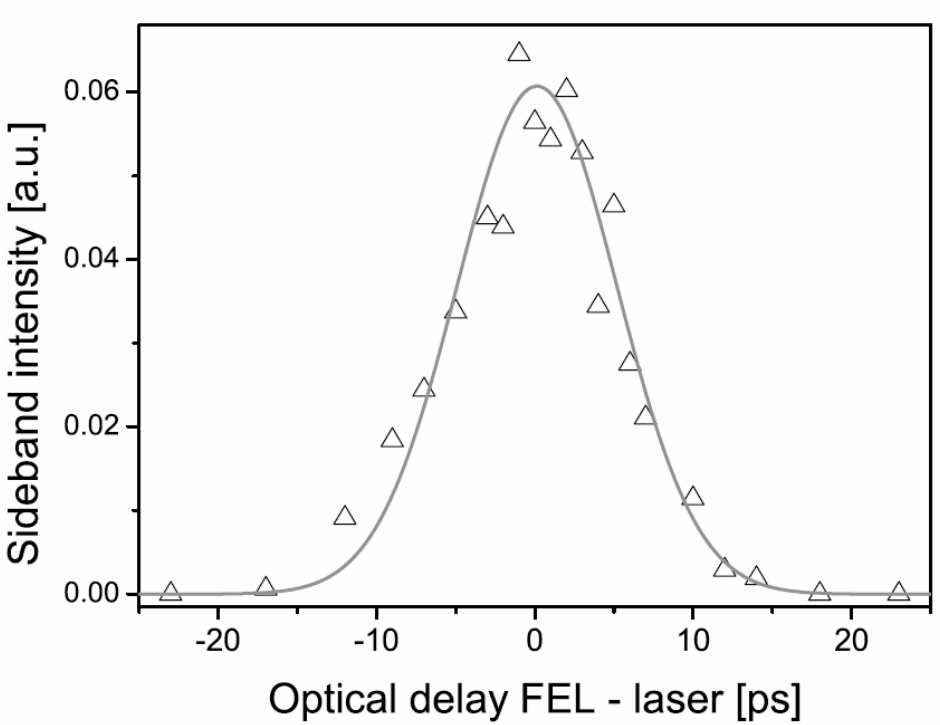


M. Meyer et al., Phys.Rev. A74, 11401(R) (2006)

Two-photon (FEL + ps laser) ionization of He

Cross correlation FEL (32.2 nm) - optical laser (523 nm)

FWHM = 12.0 ± 0.4 ps



Δt(opt.las) = 11.8 ± 0.4 ps

(ΔT (FEL) = 50 fs)

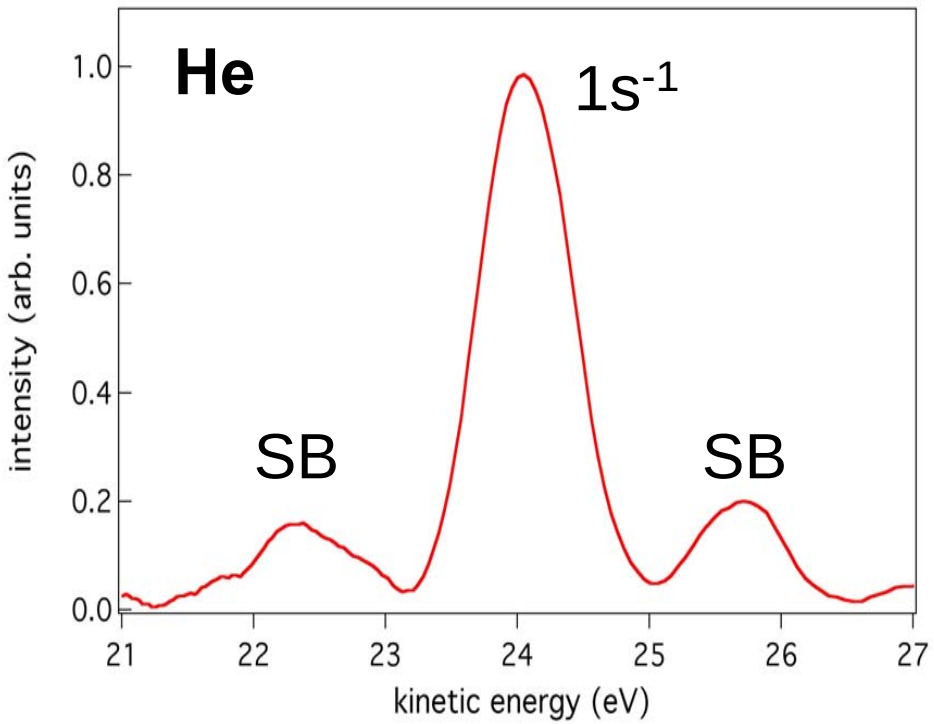
1) Determination ΔT = 0 ps

2) Jitter (FEL) < 1 ps

Two-photon (FEL + fs laser) ionization of He

FEL: 25 nm, 5 μJ, 50 μm focus, 30 fs

Laser : 800 nm, 20 μJ, 50 μm focus, 150 fs

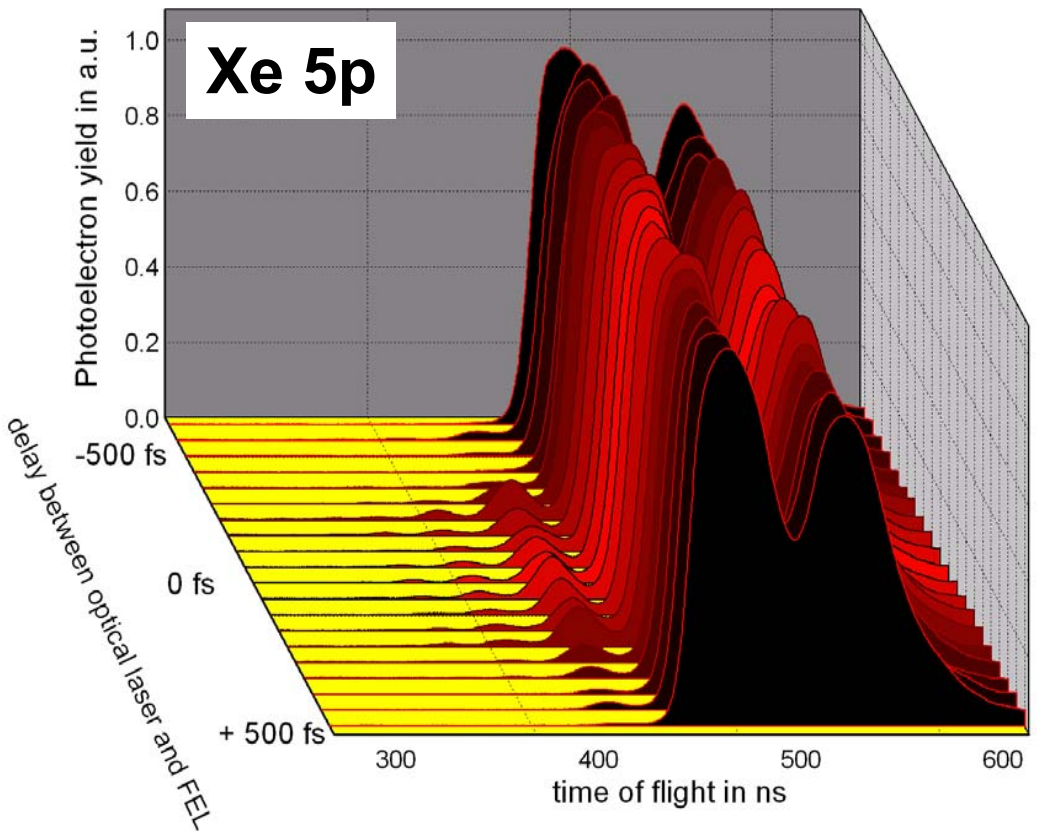


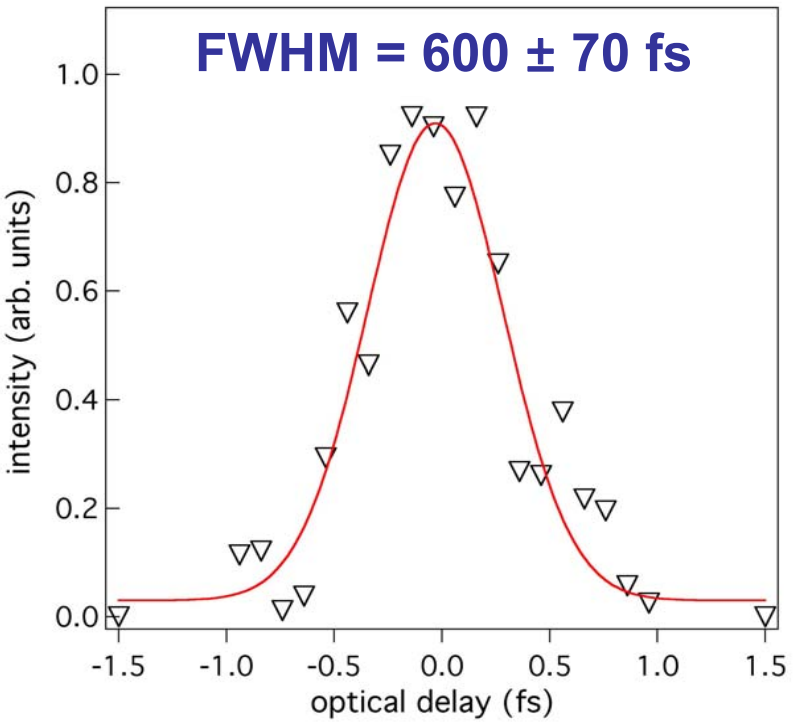
SB / $1s^{-1}$ = 0.20

Two-photon (FEL + fs laser) ionization of He

FEL: 25 nm, 5 μJ, 50 μm focus, 30 fs

Laser : 800 nm, 20 μJ, 50 μm focus, 150 fs





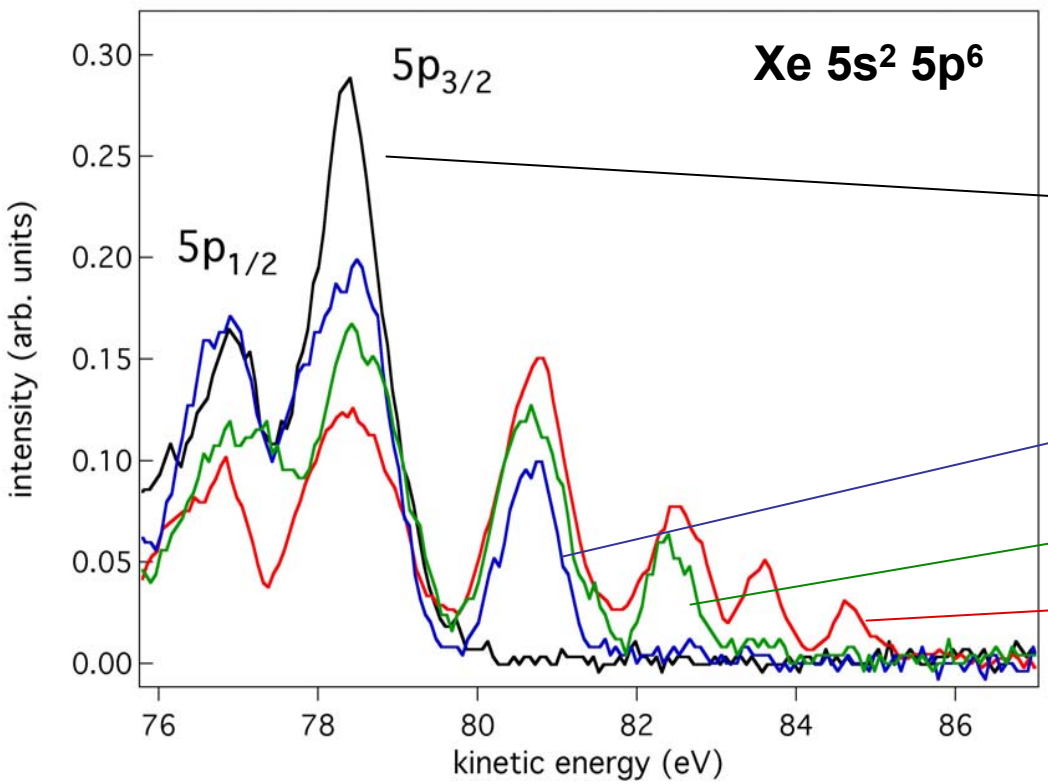
Jitter (FEL) = 550 ± 80 fs

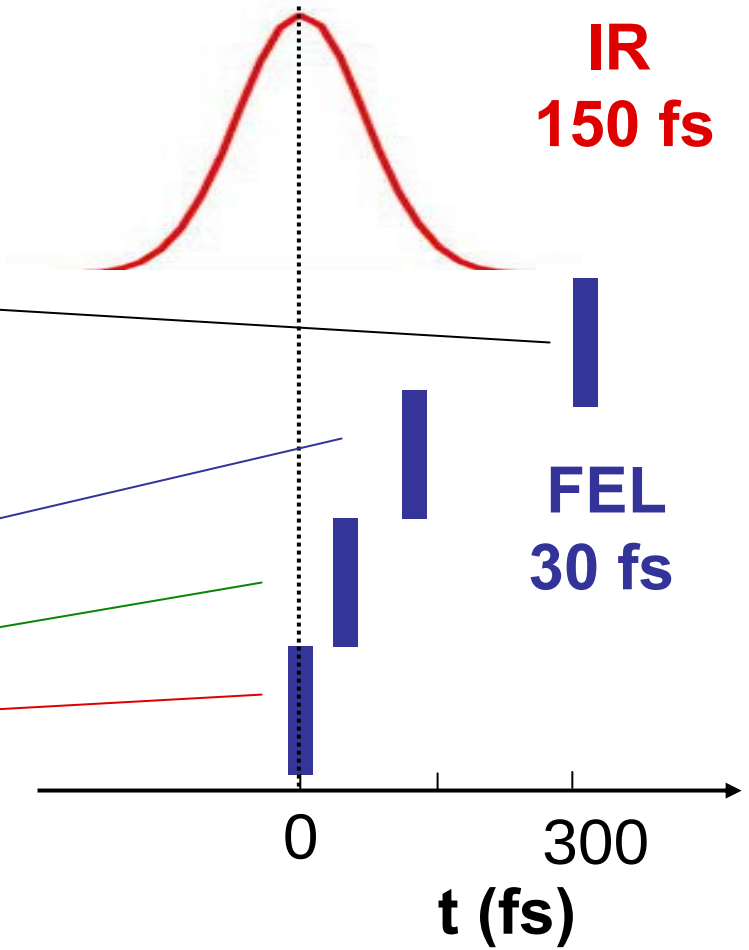
Two-photon (FEL + fs laser) ionization of Xe

hν(FEL) = 90.6 eV

IR : 150fs, 20 μJ

ΔT = 0 fs

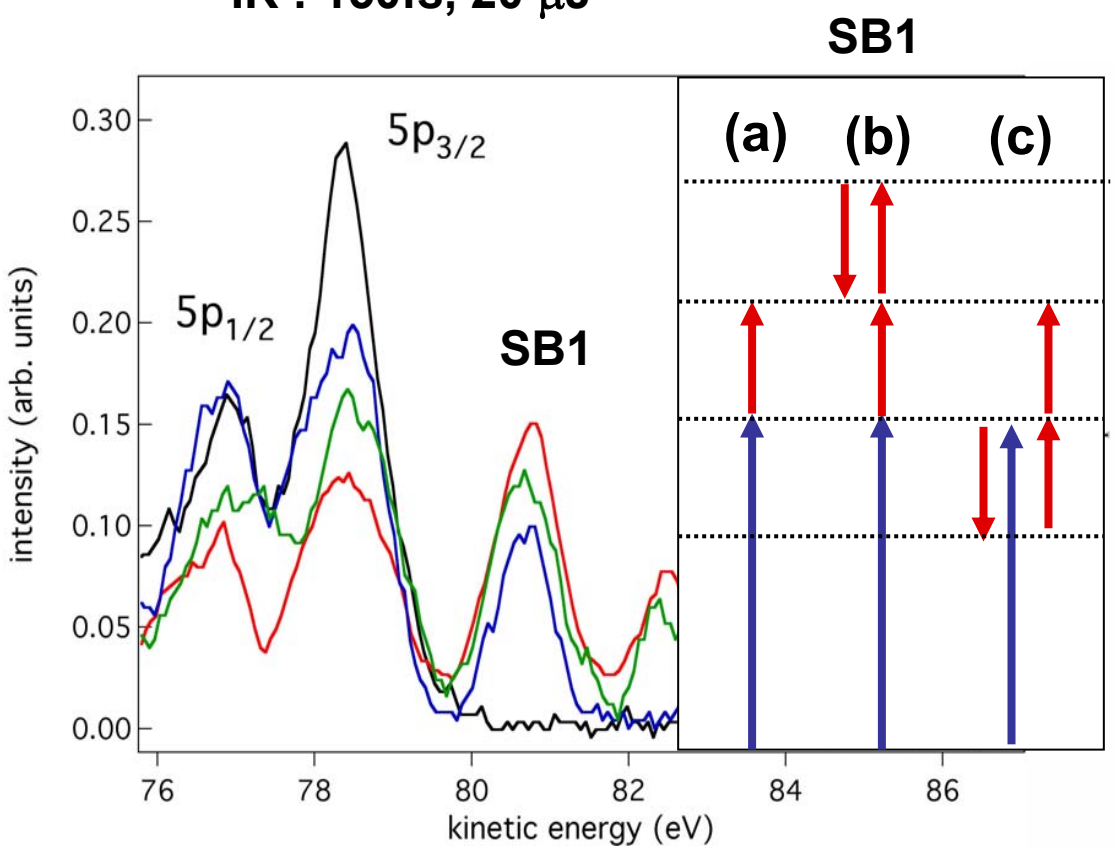




Two-photon (FEL + fs laser) ionization of Xe

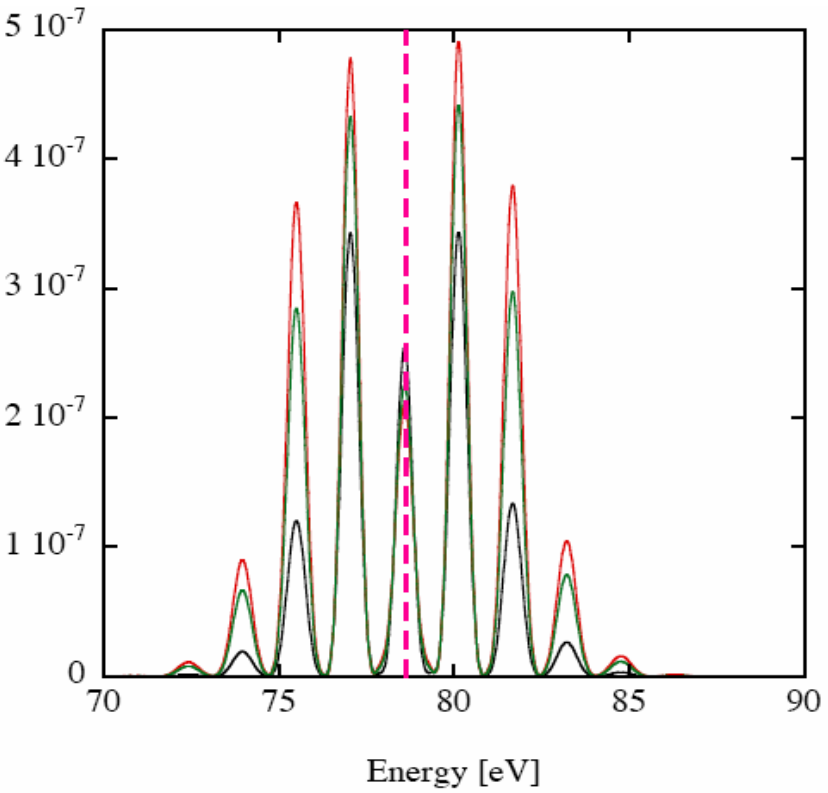
hν(FEL) = 90.6 eV

IR : 150fs, 20 μJ



Theory

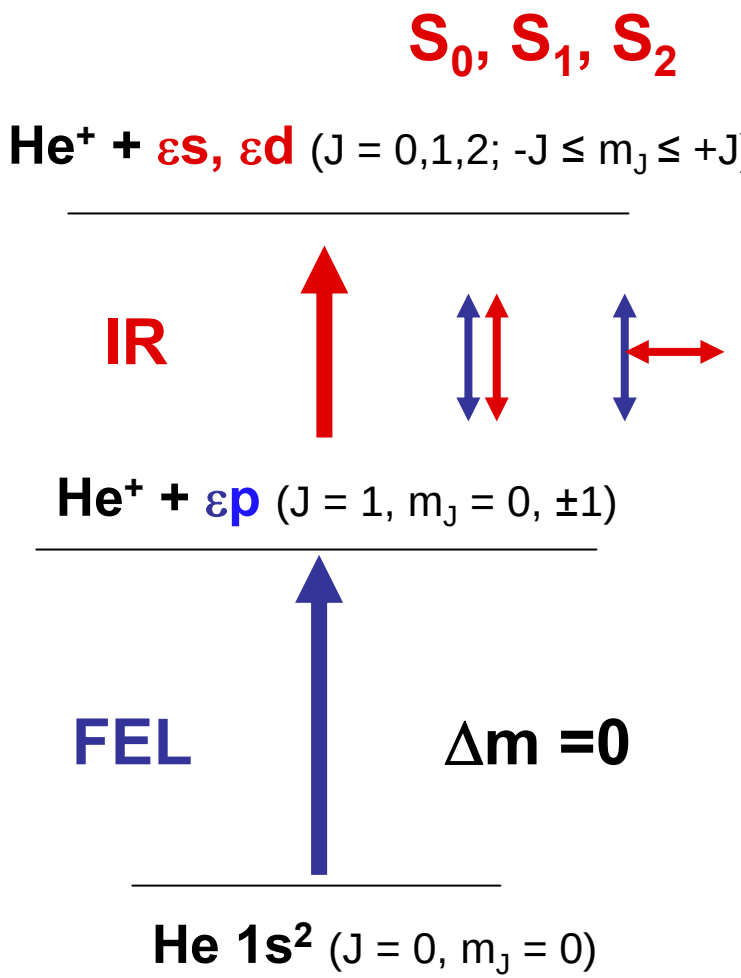
Xe + 90.6 eV (~13.7 nm)
+
TiS (I=6 10^{11} W cm^{-2})

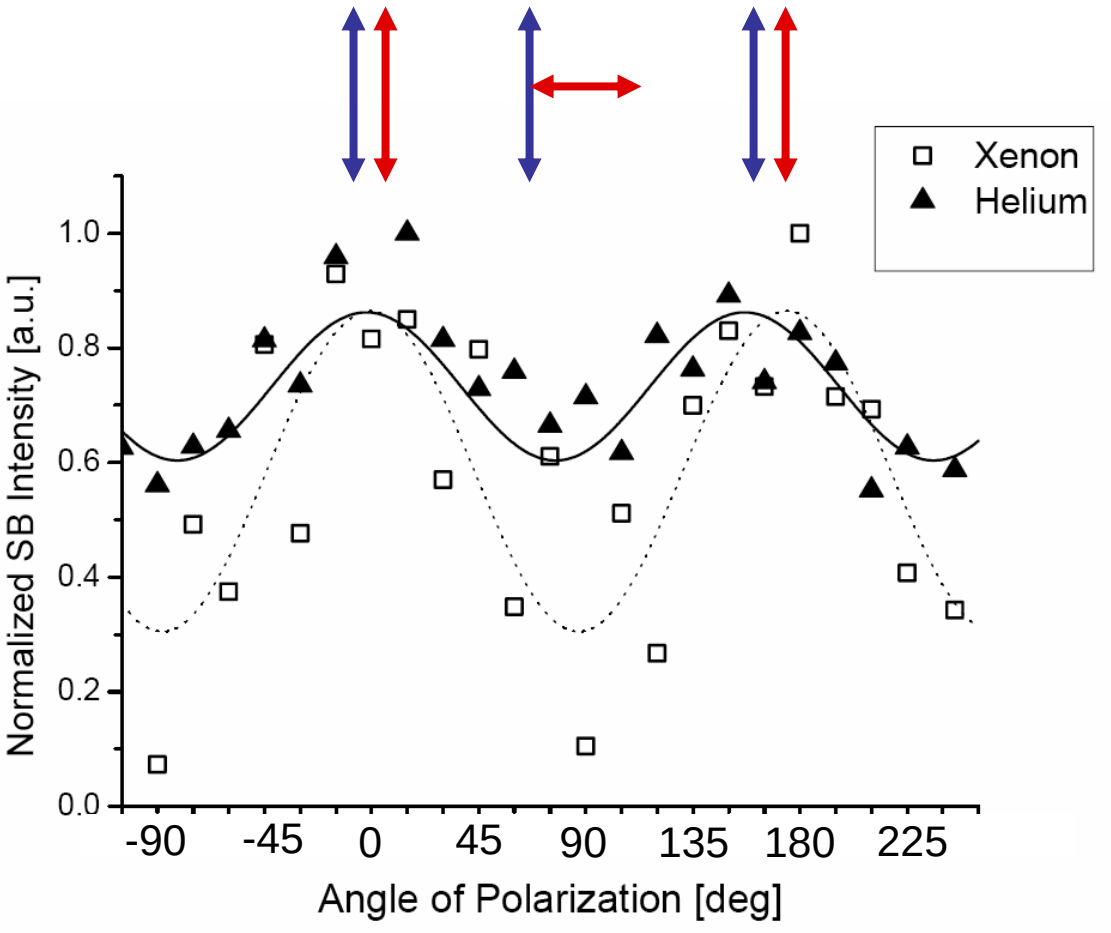


Two - Colour / Two - Photon

First applications

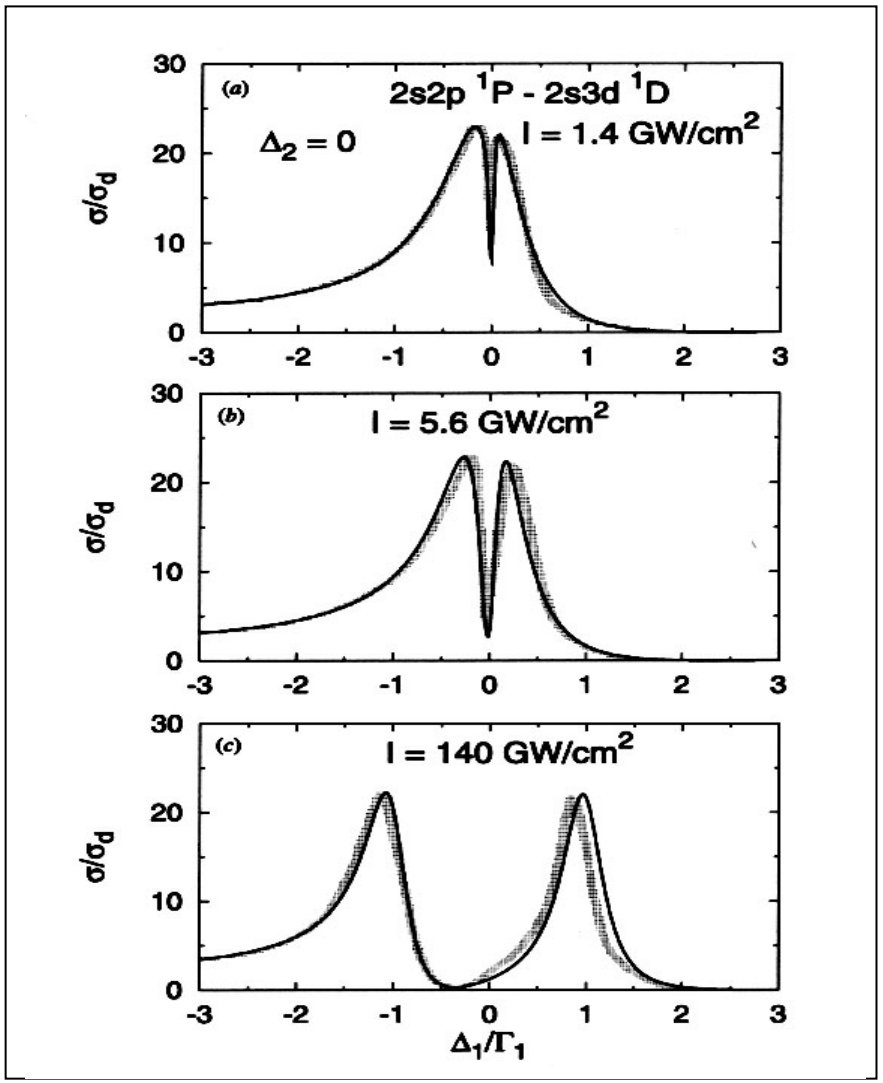
Two-photon (FEL + fs laser) : Polarization effects

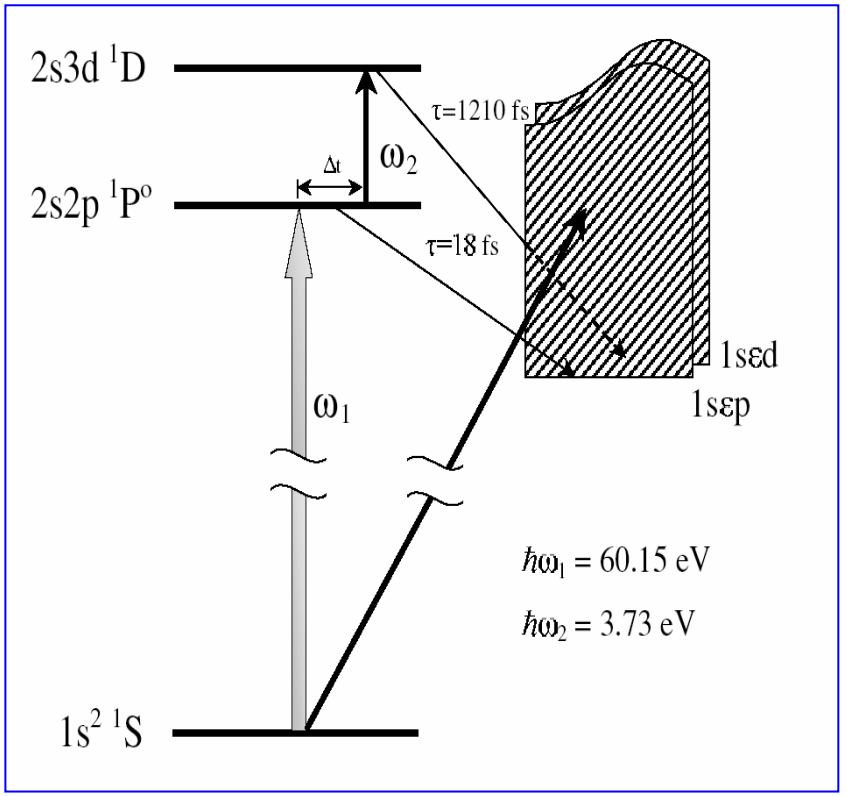




$$W(E_c) = C(3(S_1+S_2) + (2S_0-3S_1+S_2)\cos^2\Theta)$$

Dynamics of autoionizing states : coupling between two AIS by a strong laser field

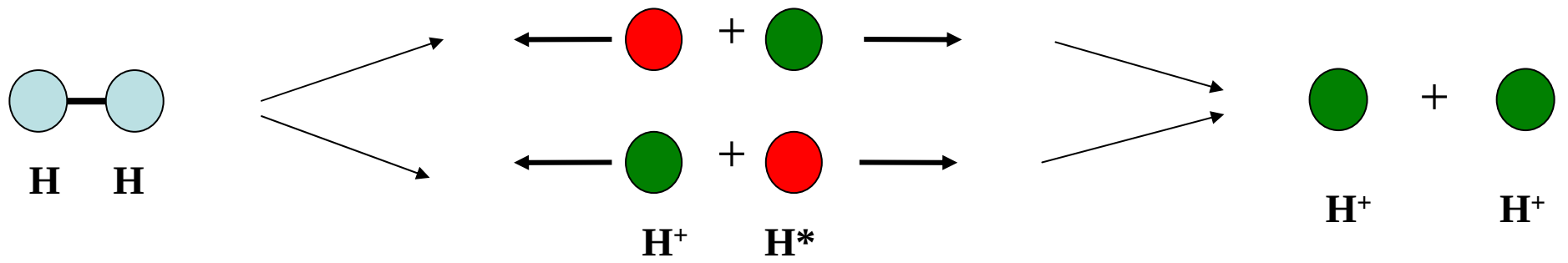




- S.I. Themelis, P. Lambropoulos, M. Meyer J. Phys. B 37, 2832 (2004)
- H. Bachau, Lambropoulos, Shakeshaft, PRA 34, 4785 (1986)

Interference effects in molecular fragmentation

H_2 + **hν (FEL)** ---> H^+ + H* (2l , 3l …) **hν (laser)** ---> H^+ + H^+



Experiment:

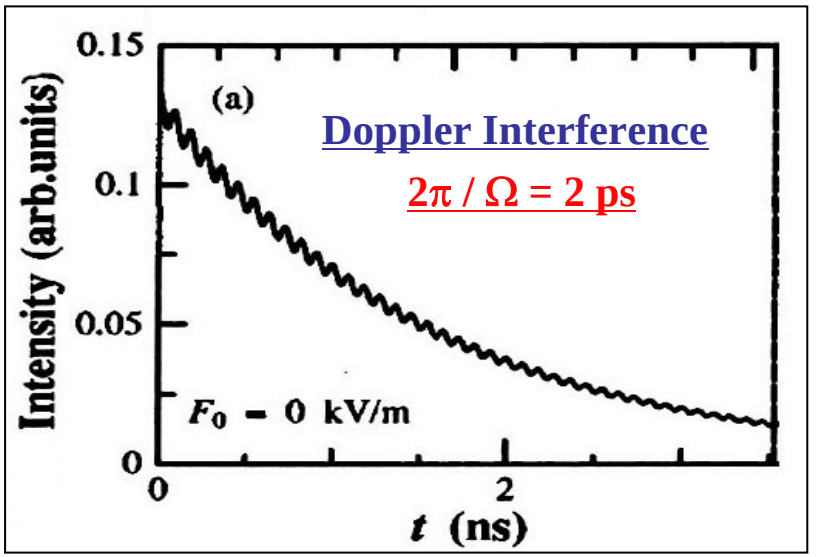
H*(3l) + hν (laser) --> H^+ + e^-

Doppler Interference

λ(las) ≈ 800 nm --> E_{kin} (e^-) = 0.1 eV

hν(FEL) = 16.8 eV --> E_{kin} (H*) = 0.3 eV

2π / Ω = 2 ps



Summary

One-color experiments

- **single shot spectra** (high photon flux)

  ---> **statistical behavior of the FEL**

- **two- and multiphoton processes** (short pulses)

  ---> **non-linear processes** **(coming up!)**

Two-color experiments

- **sideband spectra of He, Xe and Kr** (ps and fs optical laser)

  ---> **temporal stability of FEL**

- **polarization effects**

  ---> **two-photon ionization continua**

- **pump-probe applications**

  ---> **electron and nuclear dynamics in atoms and molecules**