Mulitiphoton and multicolor ionization using FEL and IR Lasers : Achievements & Perspectives

- Introduction
    - -IR vs. XUV laser fields
- Results at FLASH
    - -Two-photon processes in the XUV range
    - -Two-color IR-FEL ionization
- Perspectives

Laboratoire de Chimie Physique-Matière et Rayonnement

CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE

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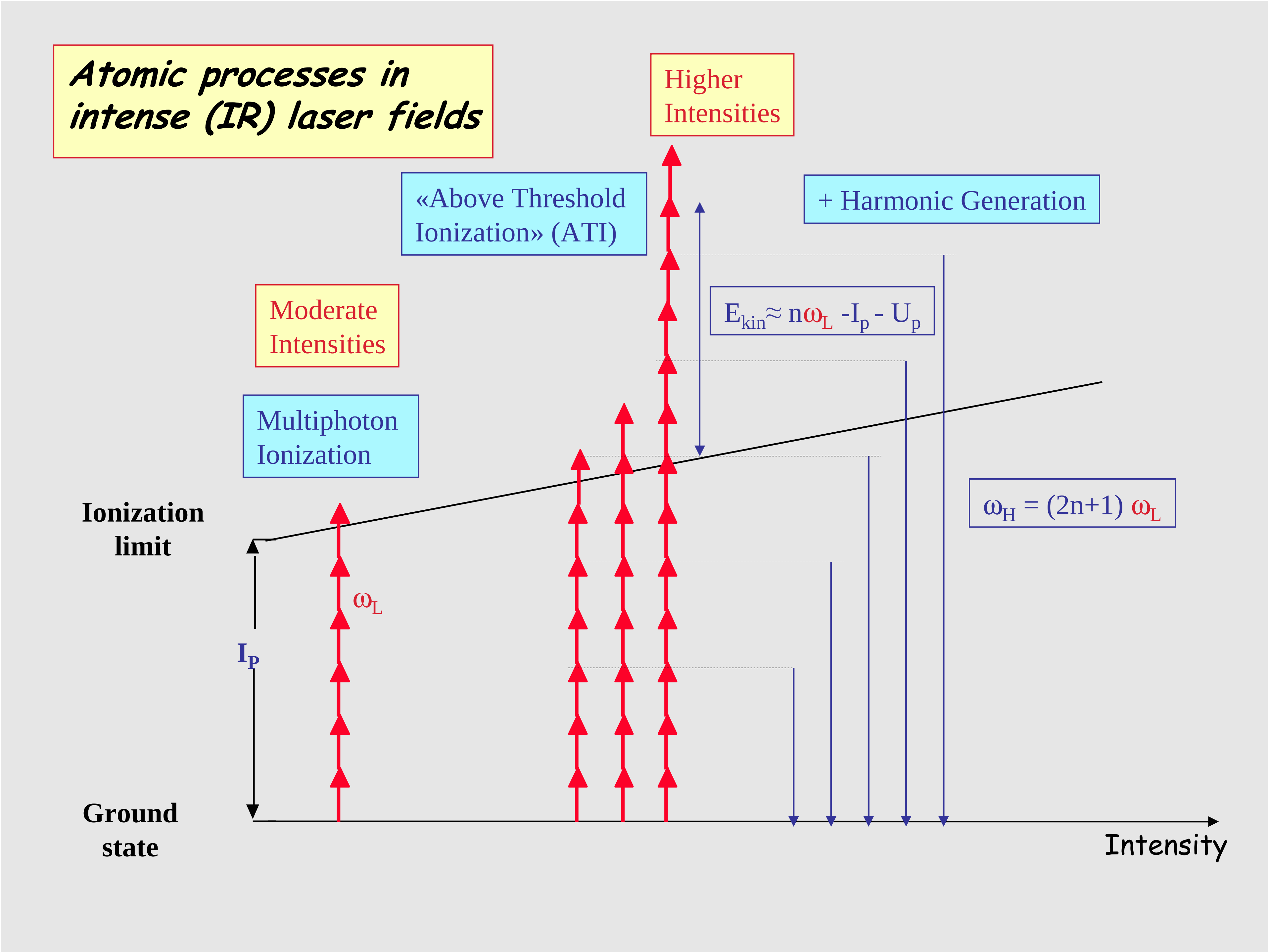


Atomic processes in intense (IR) laser fields
Higher Intensities
«Above Threshold Ionization» (ATI)
+ Harmonic Generation
Moderate Intensities
$E_{kin} \approx n\omega_L - I_p - U_p$
Multiphoton Ionization
Ionization limit
$\omega_H = (2n+1)\,\omega_L$
ω_L
I_P
Ground state
Intensity

Question: Could we observe similar effects in high-frequency (XUV and soft x-ray) fields?

(Are the currently developed sources «strong» enough?)

Strong field atomic physics: what are the differences between IR and XUV fields?

Idea: compare the classical dynamics of a free electron in
XUV vs. IR laser field
$F(t) = F_0\, \varepsilon \sin(\omega t + \phi)$

$$m\ddot{\vec{x}} = qF_0\vec{\varepsilon}\sin(\omega t+\varphi)$$

$$\dot{\vec{x}}(t) = -\frac{qF_0\vec{\varepsilon}}{m\omega}\cos(\omega t+\varphi) + \dot{\vec{x}}(0)$$

$$\vec{x}(t) = -\vec{\alpha}_0\sin(\omega t+\varphi) + \dot{\vec{x}}(0)t + \vec{x}(0)$$

$where: \quad \vec{\alpha}_0 = \dfrac{qF_0\vec{\varepsilon}}{m\omega^2}$ is the excursion length of the oscillatory motion

$U_p = \dfrac{1}{2}m\langle v^2\rangle = \dfrac{q^2F_0^{\,2}}{4m\omega^2}$ is the averaged kinetic energy of the electron in oscillatory motion: ponderomotive energy

for a given field strength, an IR field interacts more strongly than an XUV field with a continuum electron: excursion length and ponderomotive energy vary as ≈ ω^{-2}

Free electron in a laser field

Typical orders of magnitude for the relevant (classical) parameters

Ti: Sapphire laser; $\omega_L \approx 1.55$ eV = 0.057 a.u.

I (W/cm^2)	α_0/a_0	v/c	U_p (eV)
3.5×10^{12}	3.08	1.28×10^{-3}	0.209
3.5×10^{14}	30.8	1.28×10^{-2}	20.9
3.5×10^{16} (= I_{at})	308	1.28×10^{-1}	2.09×10^{3}

Soft x-ray source $\omega_X \approx 155$ eV = 5.7 a.u.

I (W/cm^2)	α_0/a_0	v/c	U_p (eV)
3.5×10^{12}	3.08×10^{-7}	1.28×10^{-5}	2.09×10^{-5}
3.5×10^{16} (= I_{at})	3.08×10^{-3}	1.28×10^{-3}	2.09×10^{-1}

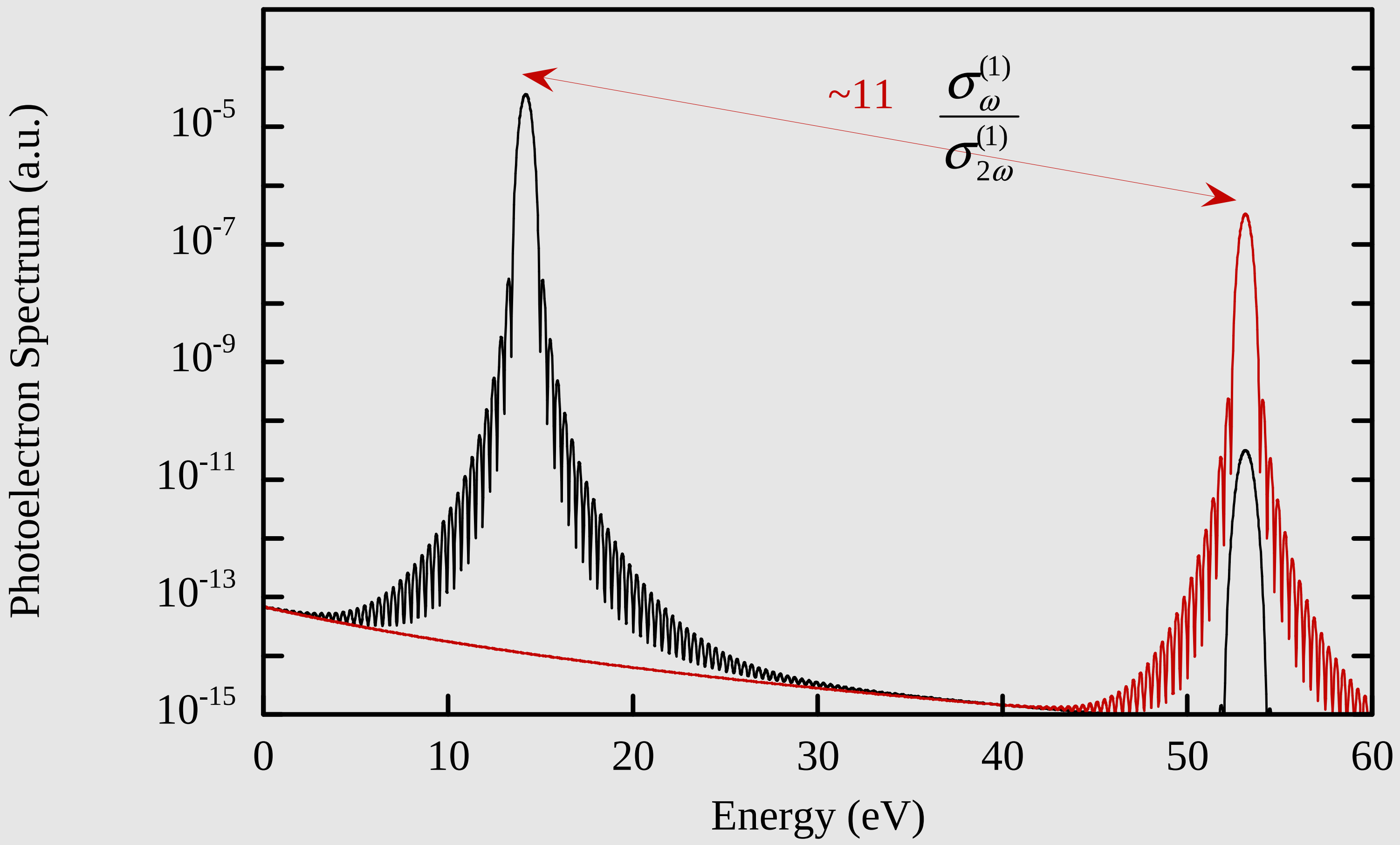
Helium + XFEL

TDSE, Single active e⁻

$$\frac{\sigma_{\omega}^{(2)}}{\sigma_{2\omega}^{(1)}} = 1 \quad I \sim 4\ 10^{14}\ Wcm^{-2}$$

FEL, ω = 38.8 eV, $I_X = 7\ 10^{11}$ W cm^{-2}

2FEL, ω = 77.6 eV, $I_{2X} = 7\ 10^{10}$ W cm^{-2}



Multiphoton multi-ionization of rare gases with FLASH

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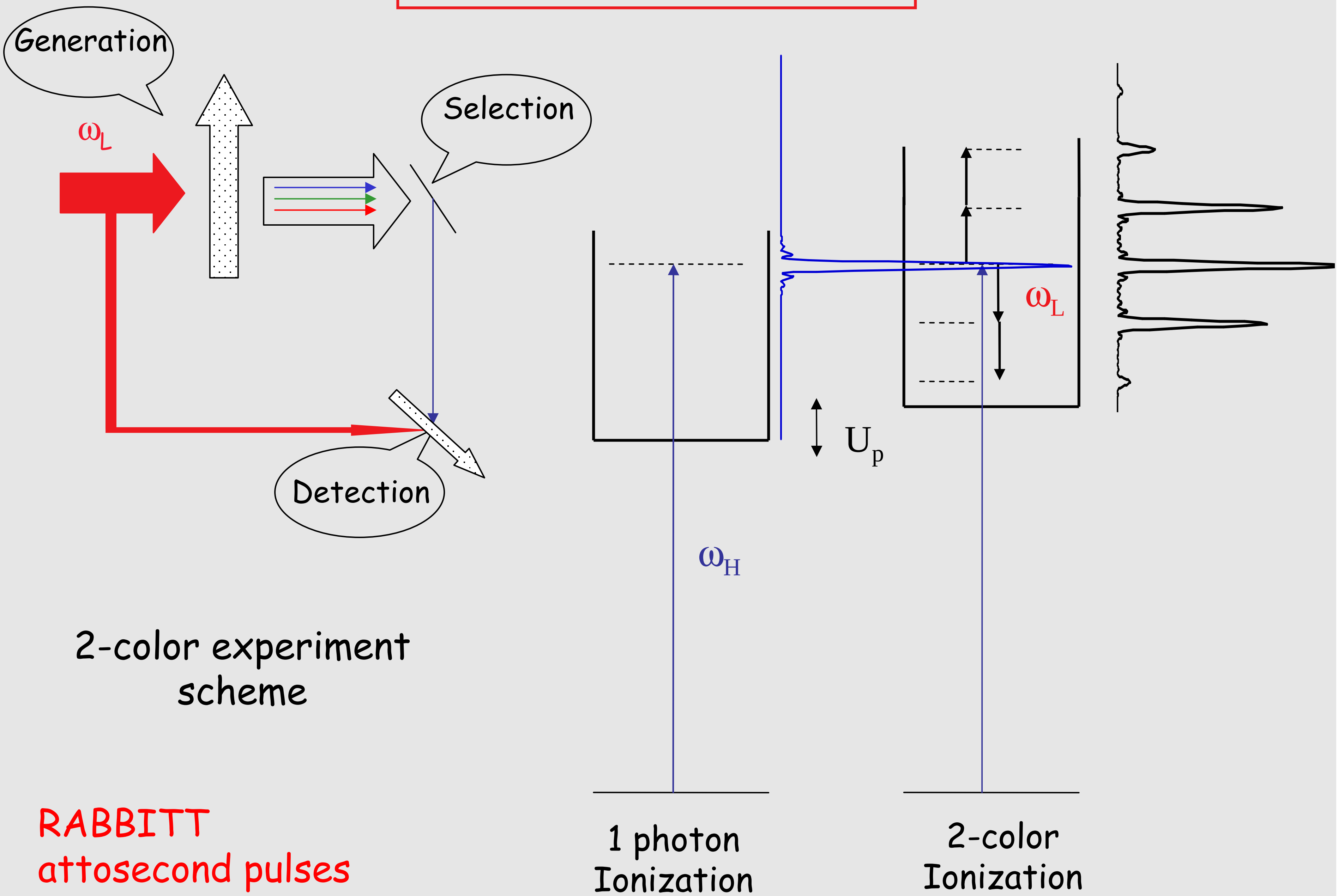
M. Richter

2-color « Experiment »



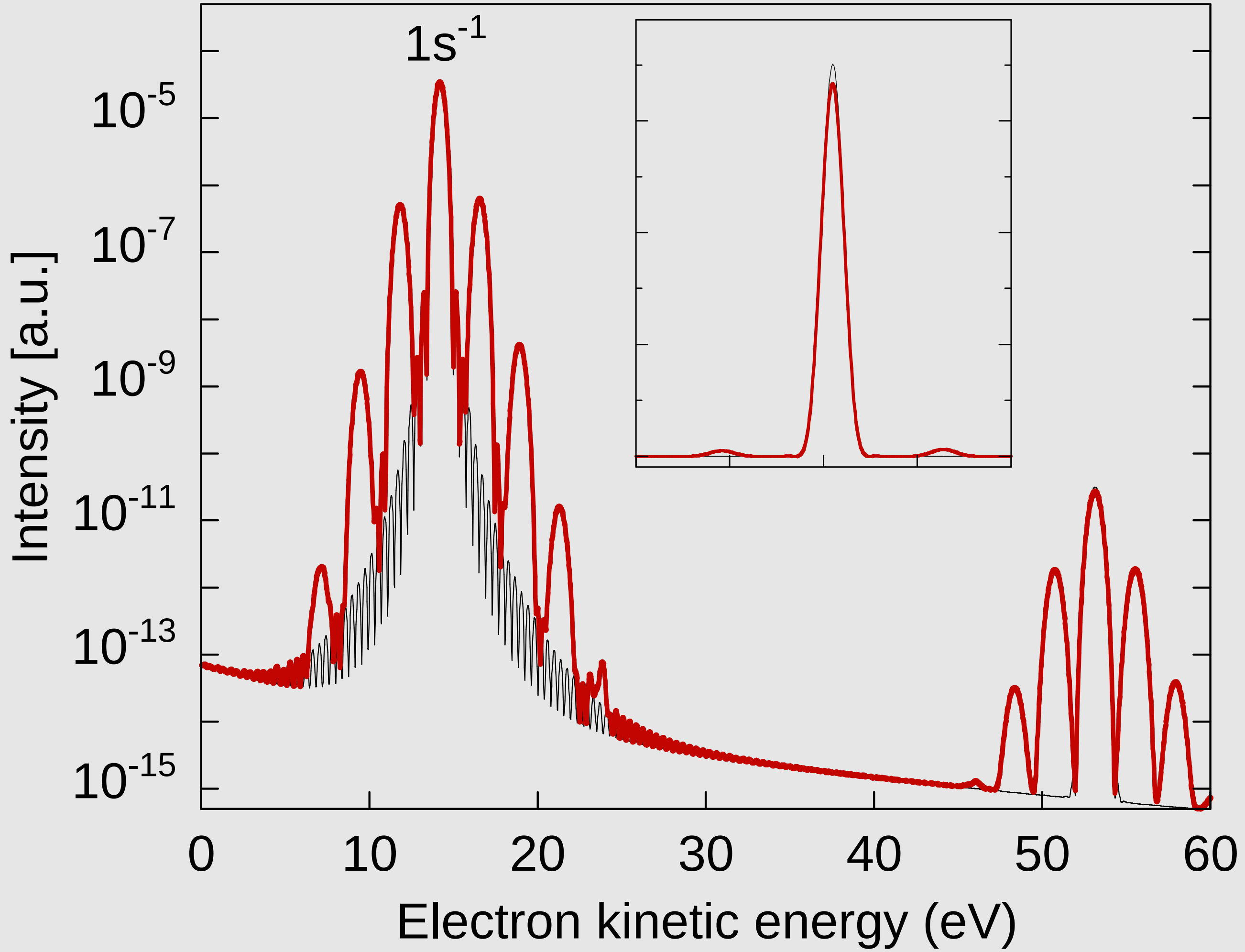
2-color experiment scheme

RABBITT
attosecond pulses

Helium + XFEL

TDSE, Single active e^-

$I_L = 2\ 10^{11}\ Wcm^{-2}$

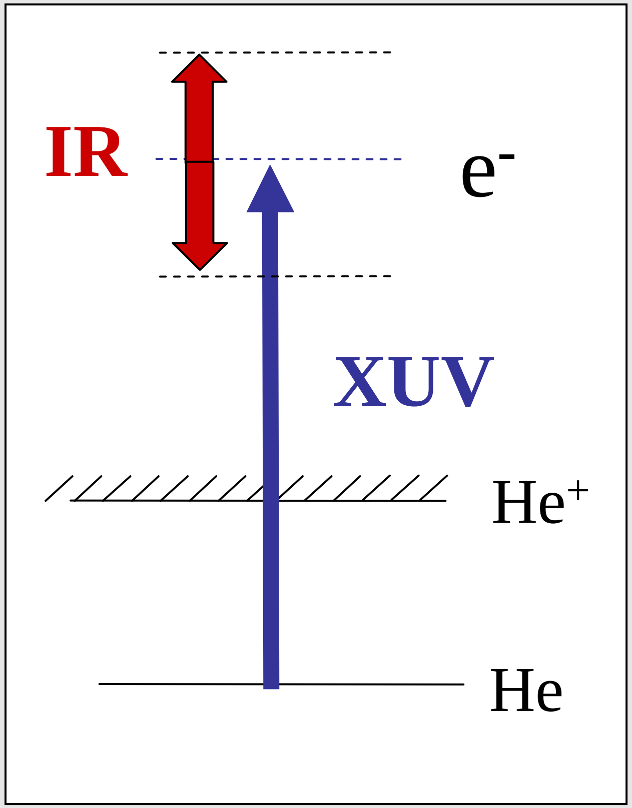


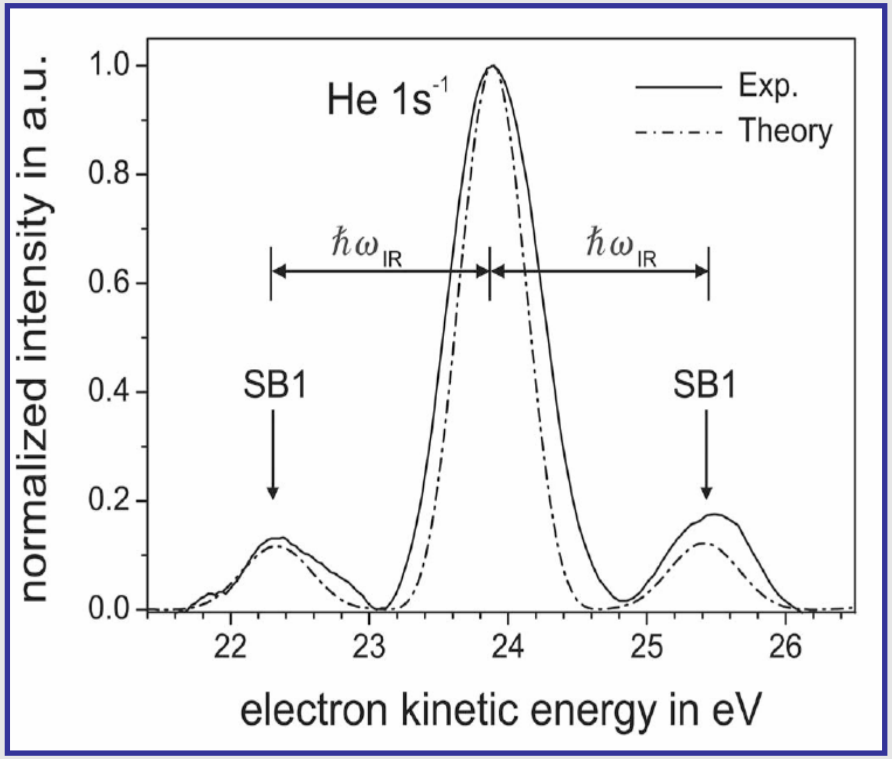
Above Threshold Ionization of Rare Gases

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





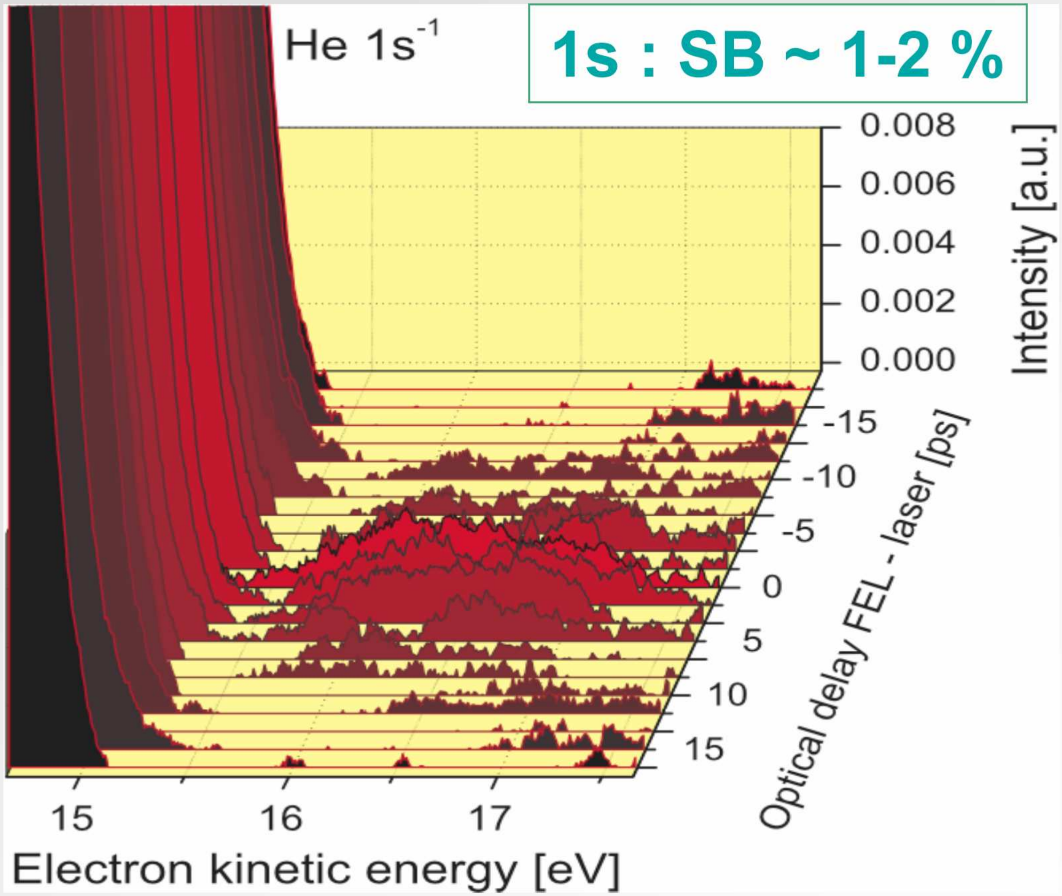
Radcliffe et al. APL 90, 131108 (2007)

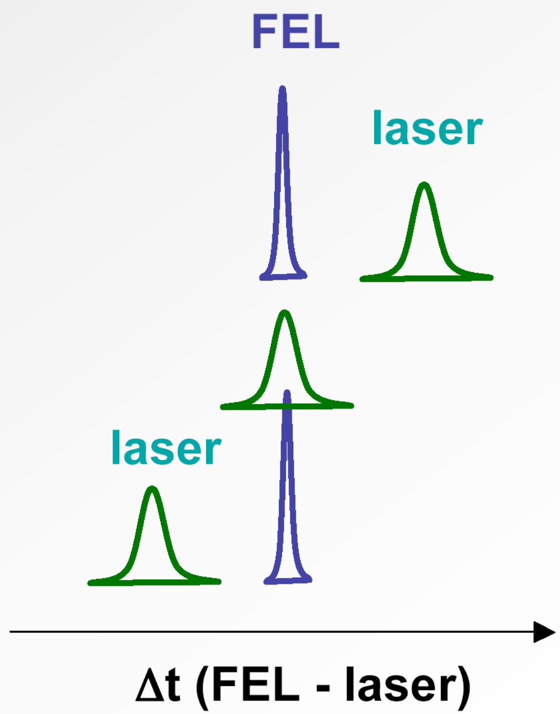
Maquet/Taieb, J.Mod.Opt. 54, 1847 (2007)

Two-photon ionization

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

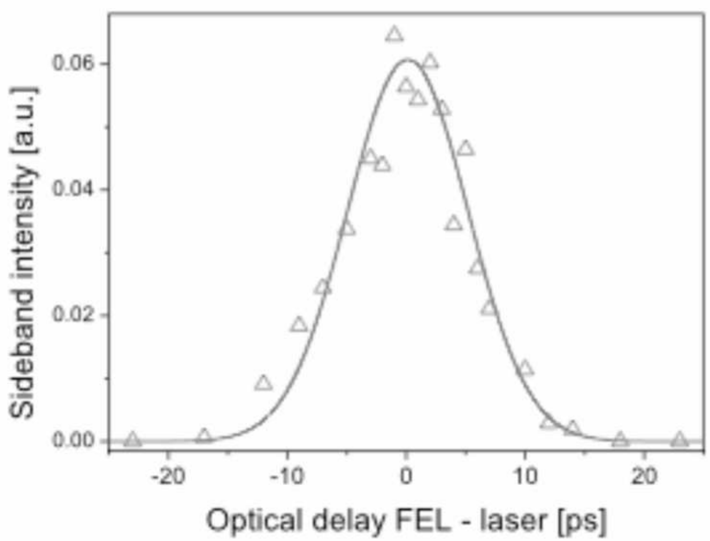




Two-photon ionization

Cross correlation FEL - optical laser

FWHM = 12.0 ± 0.4 ps



Δt(opt.las) = 11.8 ± 0.4 ps

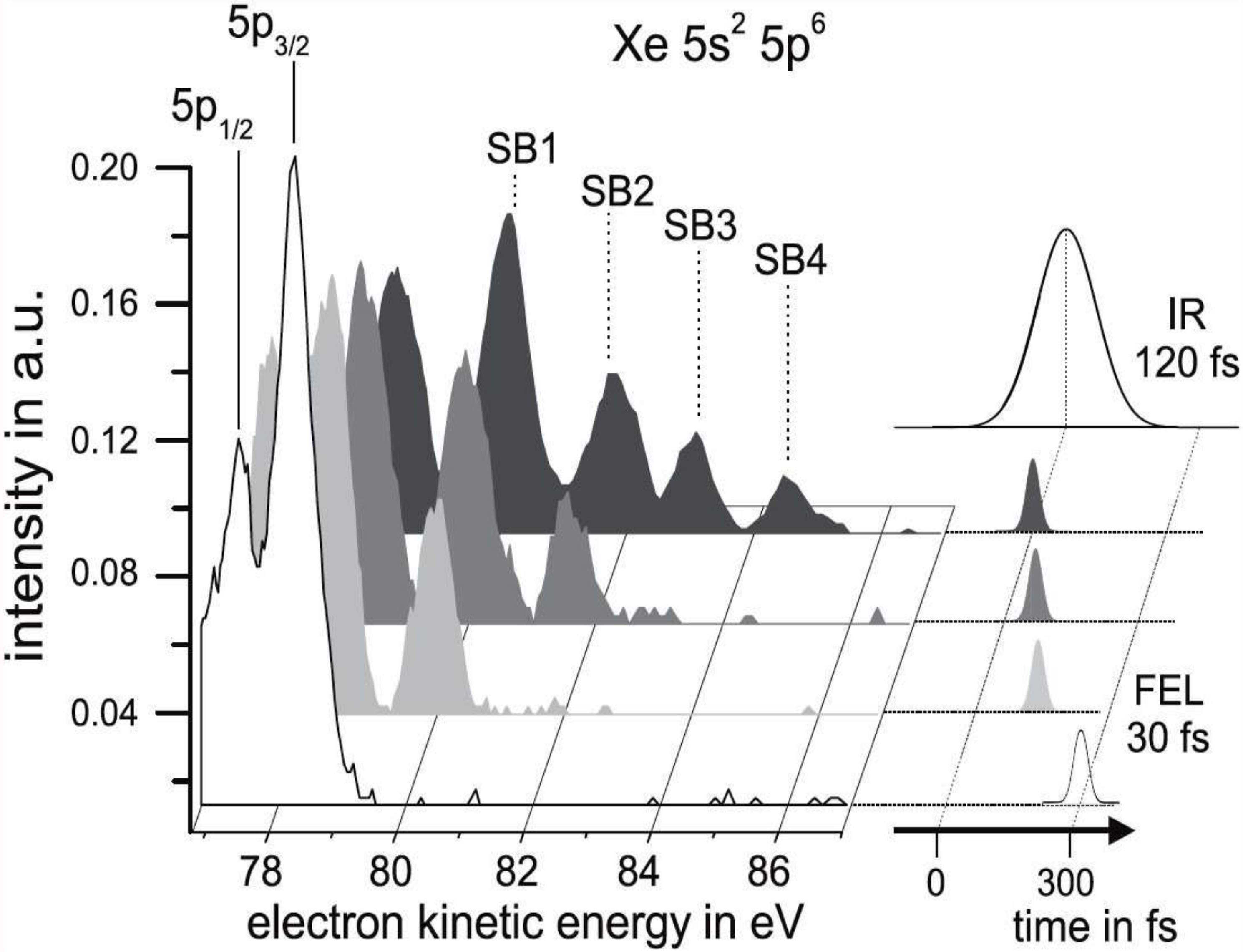
(ΔT (FEL) = 50 fs)

1) Determination ΔT = 0 ps

2) Jitter (FEL) < 1 ps

Toma et al. PRA (2000)

Single-shot spectra

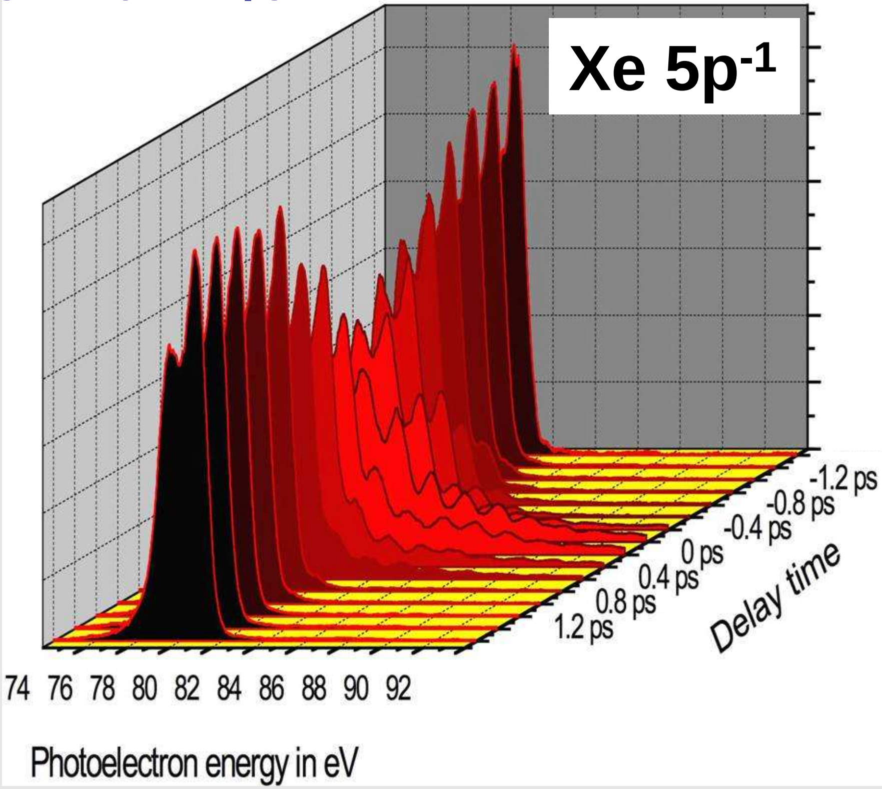


Above Threshold Ionization of Rare Gases

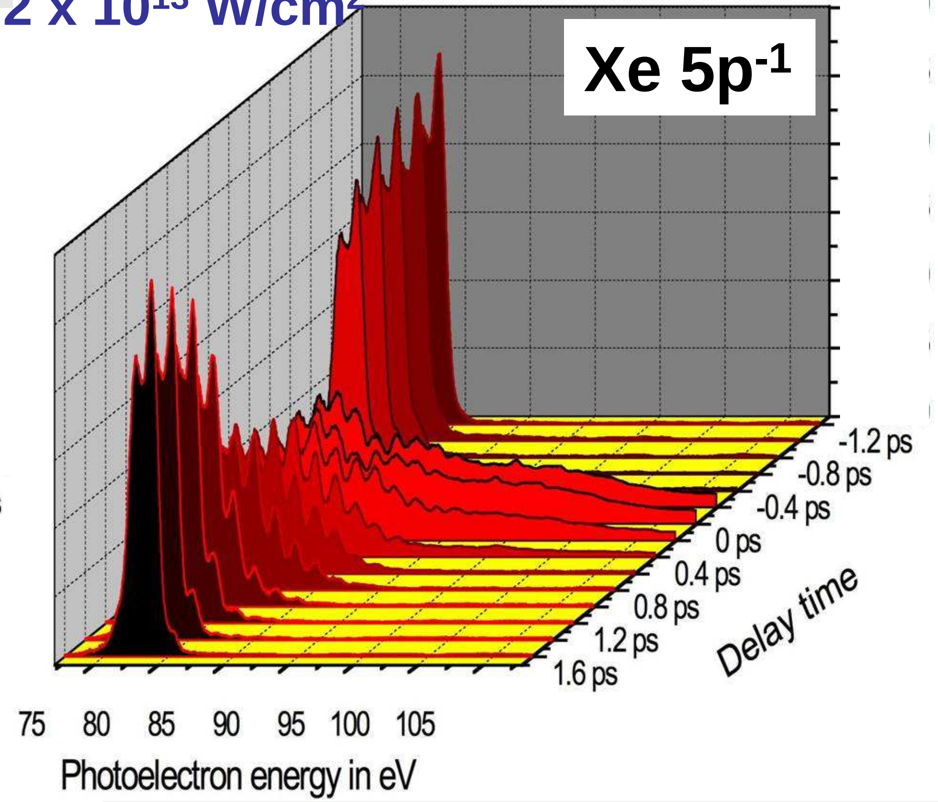
FLASH: 13.7 nm, 30 μJ, 50 μm focus, 20 fs

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

5 x 10^{12} W/cm²



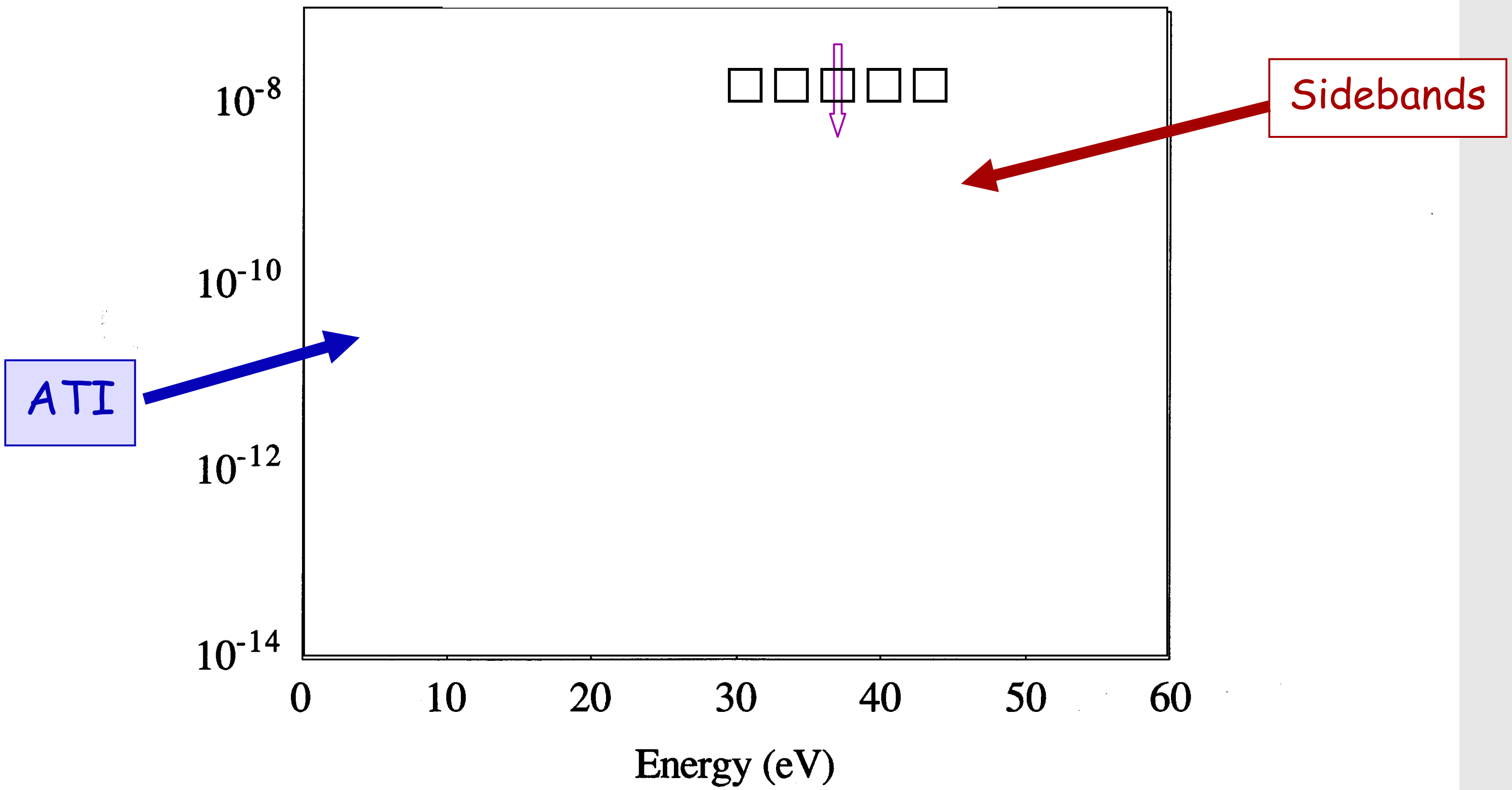
2 x 10^{13} W/cm²



H(1s) + 50 eV XUV + IR laser

TDSE

ω_X=50 eV I_X=3 10^9 W/cm^2

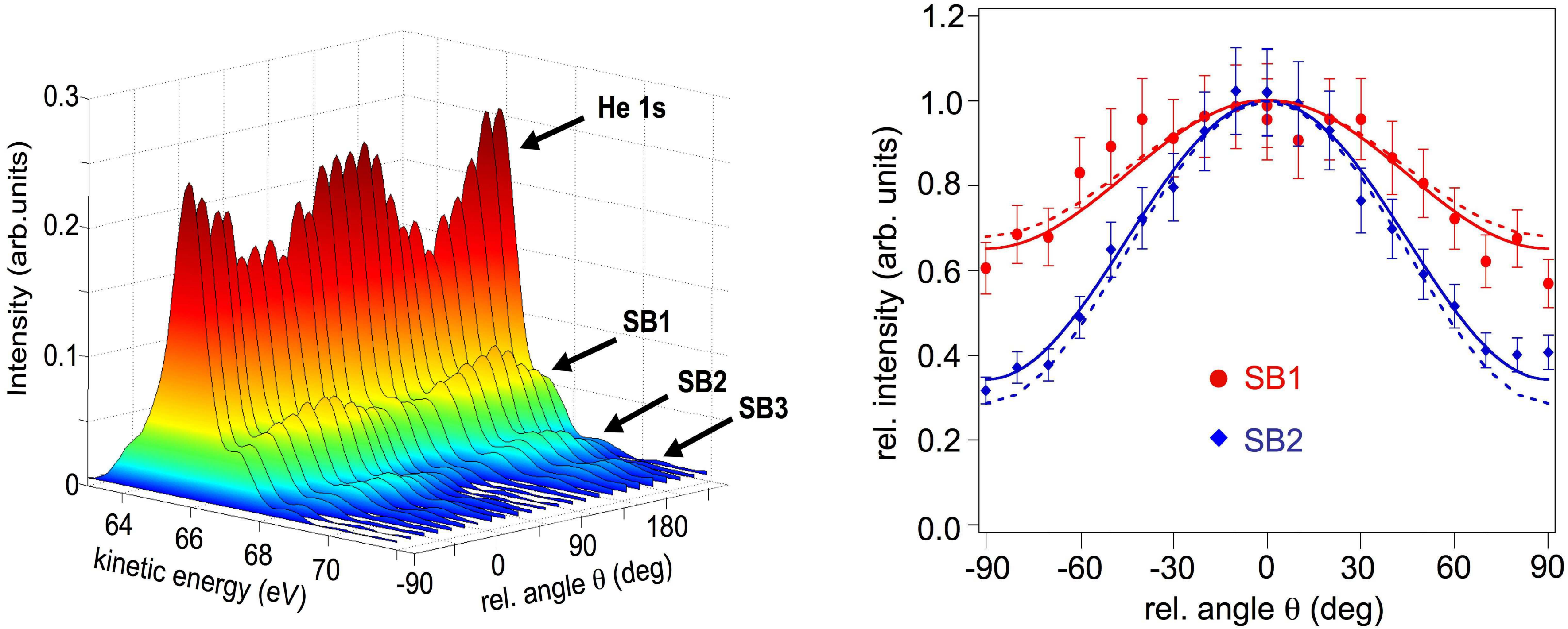


R. Taïeb *et al.* J. Opt. Soc. Am. B (1996)

Polarization control in two-color photoionization (He)

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)

PRL (08)

Higher frequencies: Time-resolved dynamics in the soft x-ray range

The advent of soft x-ray sources with pulse durations in the femto- and attosecond ranges opens new perspectives for pump-probe spectroscopies involving atomic inner-shells!

two-color ionization (x-ray + ir laser):
- the ejected (either Auger or photo-)electrons are "dressed" by the infrared laser;
- one can monitor the magnitude of the side-bands in terms of the time delay between the two pulses (phase effects)

• CHALLENGES (for theory)
- Treatment of electron correlations in a time-dependent context,
- At higher frequencies: dynamics of inner-shell processes.
- Spectral analysis of the continuous spectrum components of a multielectron wave function.

Multi-electron systems

Multiconfigurational Time Dependant Hartree-Fock (MCTDHF)

Wavefunction is developed on time-dependant Slater determinants

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décompresseur TIFF (LZW)
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System of nonlinear coupled equations for c_J and ϕ□

Compact representation of the multi-electron wavefunction

A. Scrinzi (U. Vienne)

Multiphoton transitions with XFEL

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16 keV

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Multiphoton transitions with XFEL

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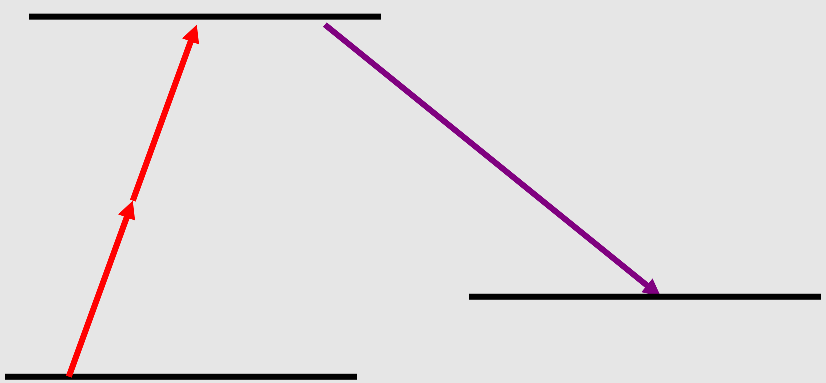
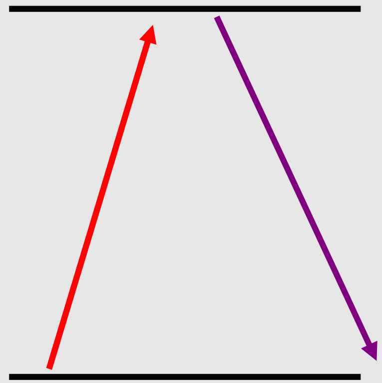
Multiphoton transitions with XFEL

Fluorescence

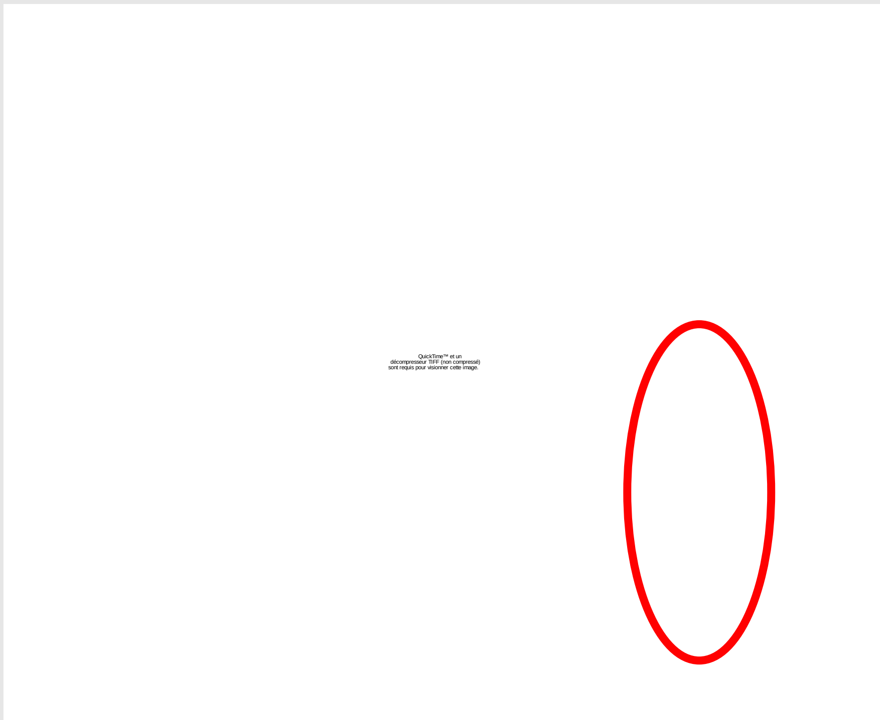




Multiphoton RIXS

Influence of high field ?

Theory ?



Possible with XFEL and core-hole level ?

Mollow (1965)

LCP-MR (Paris, France)

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