

Two-colour IR-XUV non-linear processes in atoms

- Introduction
- Free-free transitions
- Laser-assisted electron-atom excitation
- Two-colour IR-UV ionisation
- Laser-assisted Auger transitions
- Circular dichroism from unpolarized targets
- Harmonic sources:
 - Quantum interferences and phase effects
 - Characterization of « attosecond » pulses trains
 - Towards time-resolved inner-shell spectroscopy

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Strong IR laser + Synchrotron radiation

Tunable lasers used to transfer population in an excited state:
⇒ inner-shell ionisation dynamics of excited species.
Demonstration of the importance of correlations.
(*Bizau, Wulleumier et al. early eighties...*)

With strong IR lasers, new «two-colour», non-resonant processes can be explored, notably, those involving continuum states!

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

Idea: compare the classical dynamics of a free electron in

- XUV radiation field vs.
- IR laser field

$$F(t) = F_0 \varepsilon \sin(\omega t + \phi)$$

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

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

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

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

$$U_p = \frac{1}{2} m \langle v^2 \rangle = \frac{q^2 F_0^2}{4m\omega^2} \text{ is the averaged kinetic energy of the electron}$$

in oscillatory motion : ponderomotive energy.

\Rightarrow for a given field strength, an IR field interacts more strongly than an UV field with a continuum electron: excursion length and ponderomotive energy vary as $\approx \omega^{-2}$

Free electron in a laser field

Typical orders of magnitude for the relevant (classical) parameters

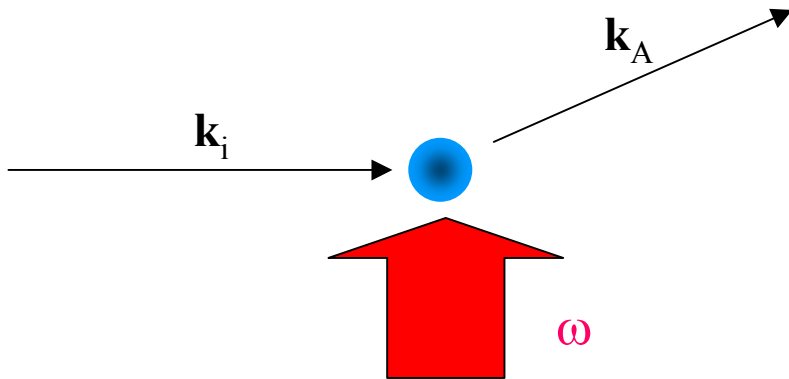
Ti: Sapphire laser; $\omega_L \approx 1.55 \text{ eV} = 0.057 \text{ a.u.}$

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
3.5×10^{18}	3.08×10^3	-	2.09×10^5

Soft x-ray source $\omega_x \approx 155 \text{ eV} = 5.7 \text{ a.u.}$

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^{14}	3.08×10^{-5}	1.28×10^{-4}	2.09×10^{-3}
3.5×10^{16} (= I_{at})	3.08×10^{-3}	1.28×10^{-3}	2.09×10^{-1}

A typical example: **Laser**-Assisted Electron-**Atom** Collisions:
«Free-Free Transitions»



$$e^-(\mathbf{k}_i) + A + N\gamma(\omega) \rightarrow e^-(\mathbf{k}_A) + A + (N \pm n)\gamma(\omega)$$

$$\mathbf{k}_i^2/2 = \mathbf{k}_A^2/2 \pm n\omega \quad ; \quad \text{NB: } N \gg n$$

In the course of the scattering event, the projectile-target system can either:

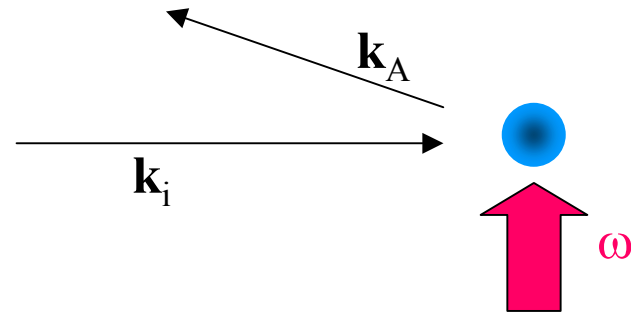
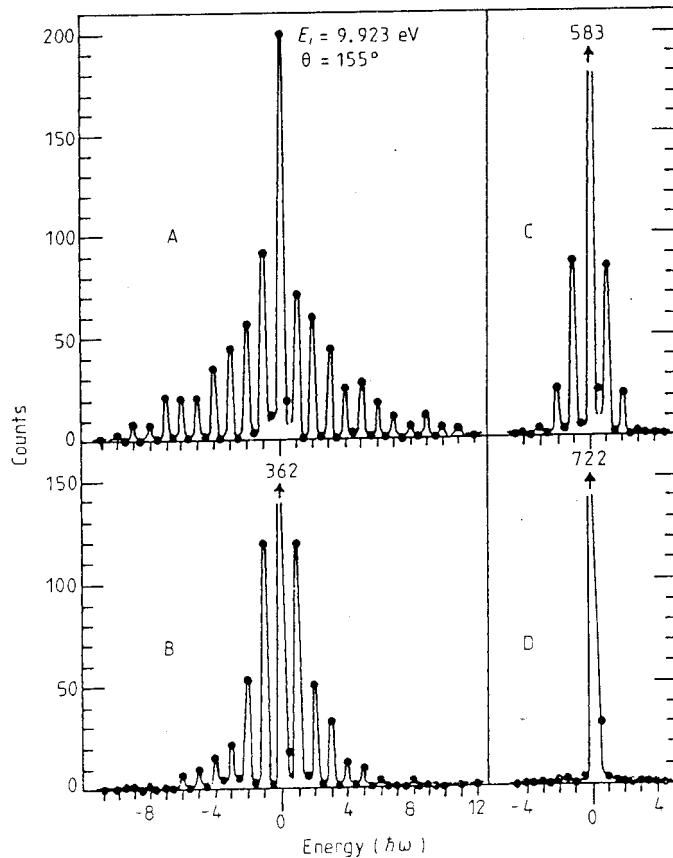
- **absorb** (-n) = «inverse bremsstrahlung», or
- **emit** (+n, stimulated emission) photons from the field = «stimulated bremsstrahlung».
- If the atom remains in its ground state the process is dubbed: «free-free transition».
- The process plays an important role when discussing plasma heating, etc.

NB. When $\omega \rightarrow 0$, the bremsstrahlung cross-section varies as (Low, 1958):

$$d\sigma_{\text{bremss}}(\omega) \approx \omega^{-1} d\sigma_{\text{elast}}$$

(Exp.: Weingartshofer et al, 1979)

A Weingartshofer et al



$(e^-, \text{Ne}) + \text{CO}_2 \text{ laser}$

NB: With IR lasers, the so-called «Soft-Photon Approximation» provides a good estimate of the differential cross sections for the exchange of n photons:

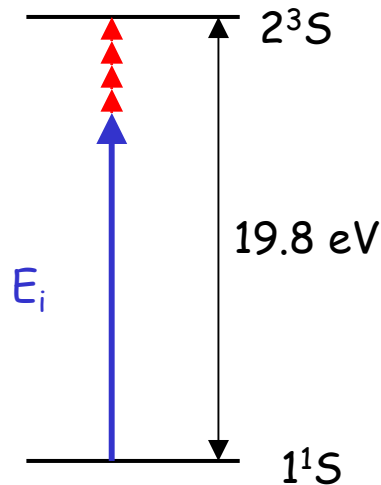
$$d\sigma_n(E_i) \approx |J_n(\alpha, \mathbf{K})|^2 d\sigma_{\text{exact}}(E_i \pm n\hbar\omega),$$

where $\mathbf{K} = \mathbf{k}_i - \mathbf{k}_A$ and $d\sigma_{\text{exact}}$ is the (exact!) elastic cross section in the absence of the laser, [Kroll & Watson (1973)].

Laser-Assisted Electron-Atom Excitation: $1^1S \rightarrow 2^3S$ transition in He

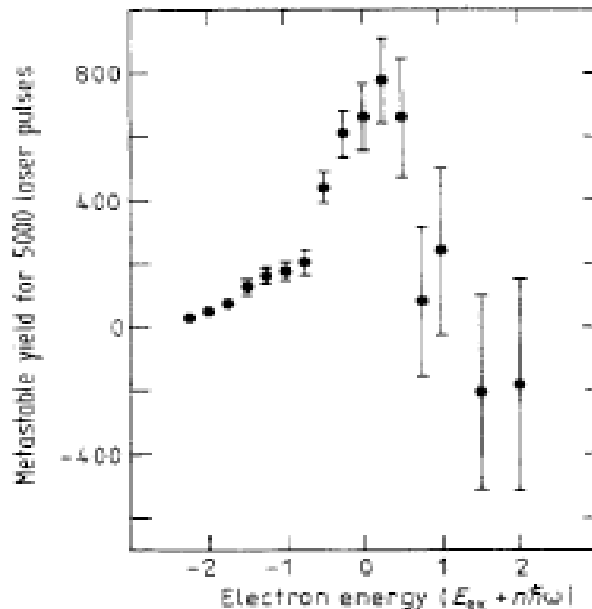
If $E_i < 19.8$ eV, the laser photons can supply the difference...

Experiments: • Mason and Newell (1987...) • Wallbank et al. (1988...) CO_2
• Hippler et al. (1991) Nd-Yag



Wallbank et al. (1989):

Simultaneous electron-photon excitation of He 2^3S

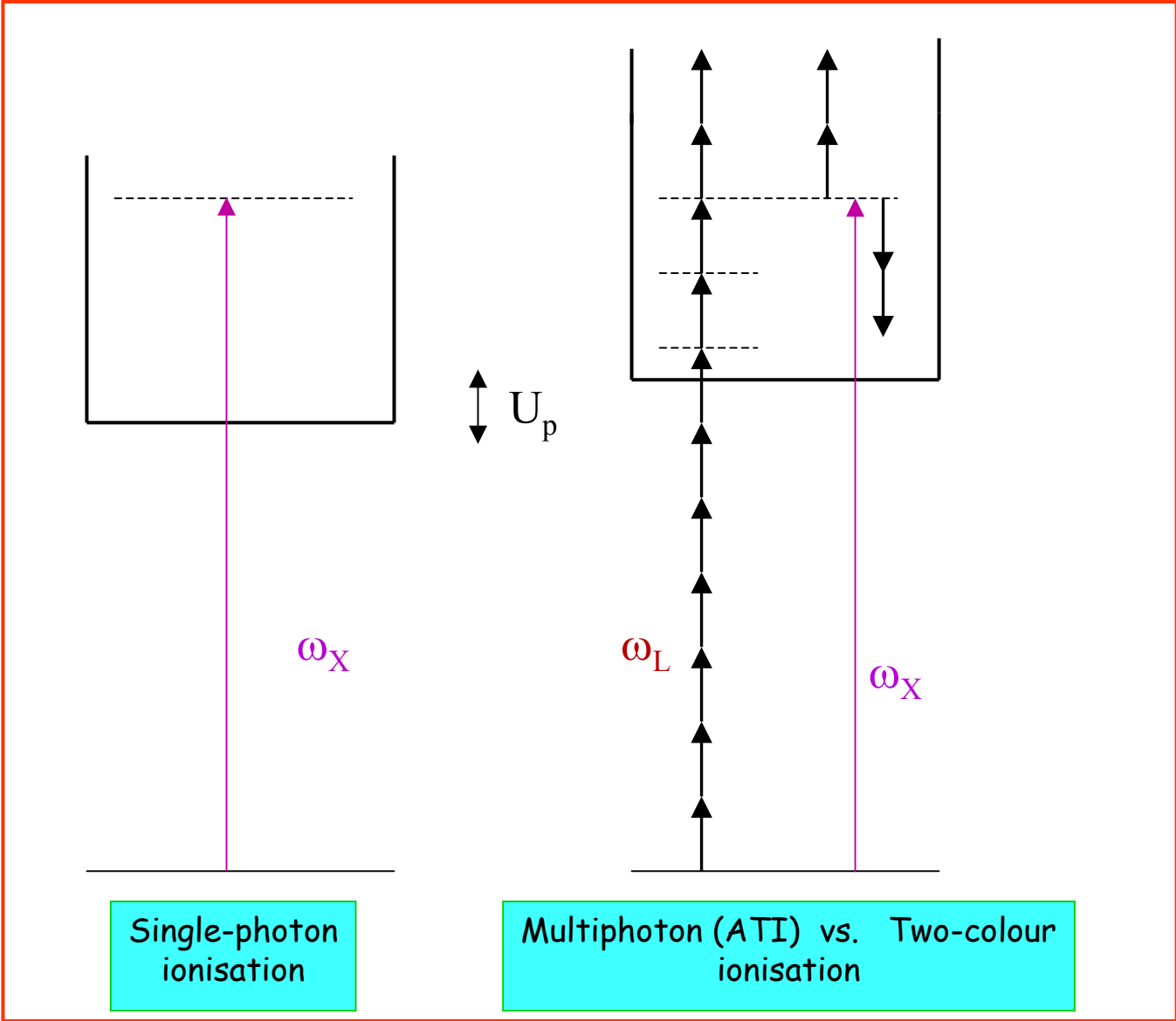


NB: Soft-photon approximation \approx OK for total cross-sections.

See: Geltman & Maquet (1989), Maquet & Cooper (1990), Fainstein & Maquet (1996).

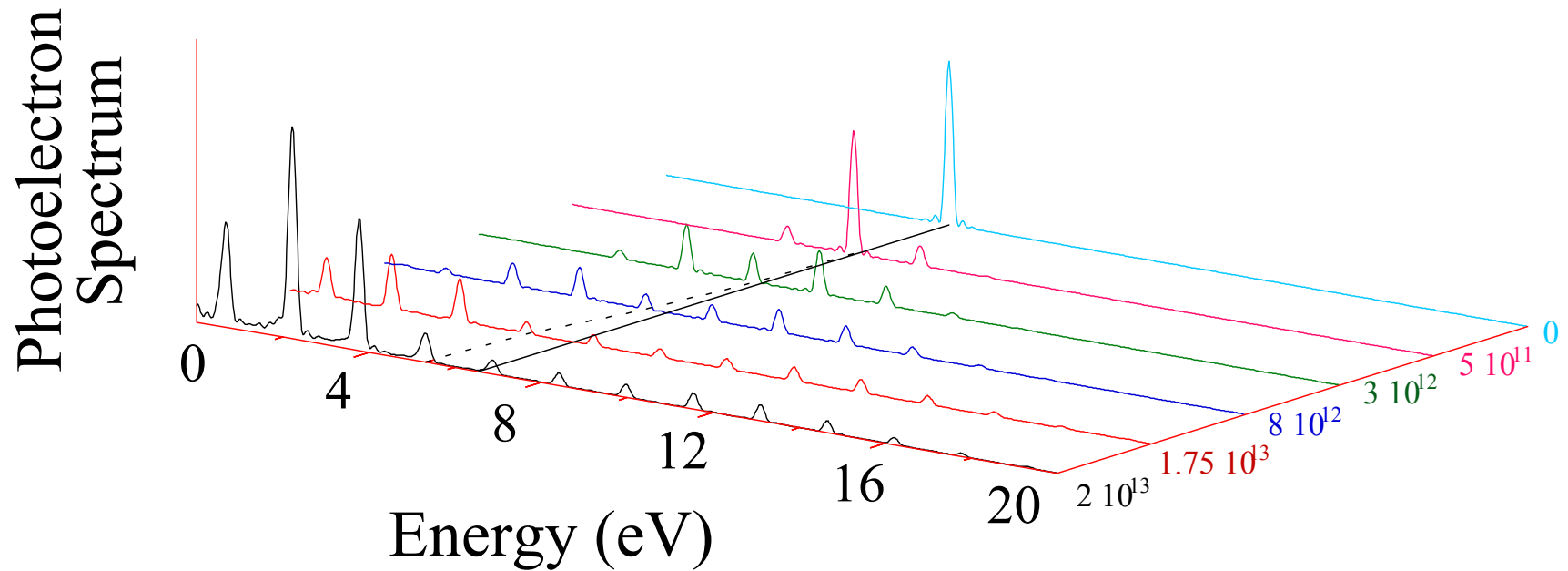
However: no consensus amongst theoreticians regarding angular distributions... Lack of experiments: COLTRIMS?

Another scenario: laser-assisted single-photon ionisation



Simulation in H atom: two-colour IR-UV photoelectron spectra
 (V. Vényard, R. Taïeb & A. Maquet, PRL, 74, 4161, 1995)

$$\omega_L = 0.057 \text{ au} ; \omega_H = 13 \omega_L ; I_H = 3 \cdot 10^8 \text{ W/cm}^2$$

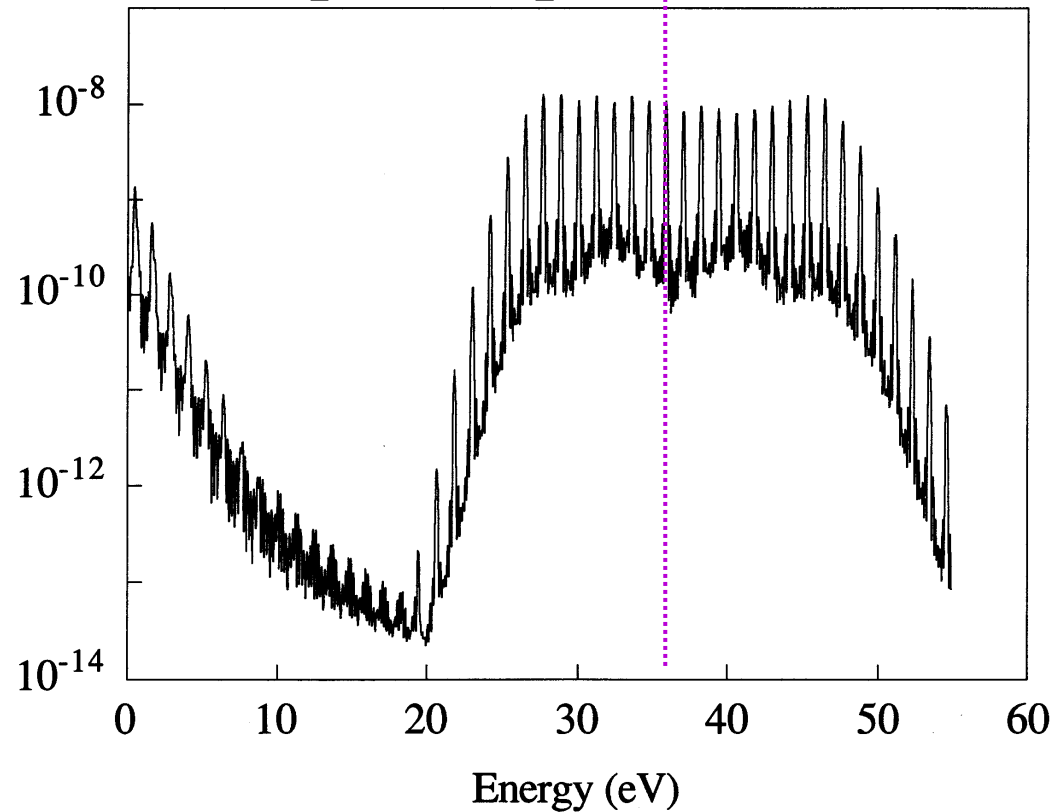


Taïeb *et al.* JOSA B, 13 363, (1996)

Simulation in H atom: two-colour IR-UV photoelectron spectrum
(R. Taïeb *et al.* 1996)

$$\omega_X = 50 \text{ eV} \quad I_X = 3 \cdot 10^9 \text{ W/cm}^2$$

$$\omega_L = 1.17 \text{ eV} \quad I_L = 5 \cdot 10^{12} \text{ W/cm}^2$$



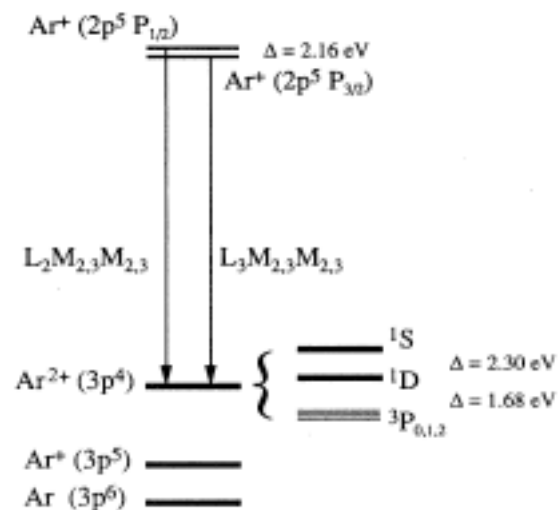
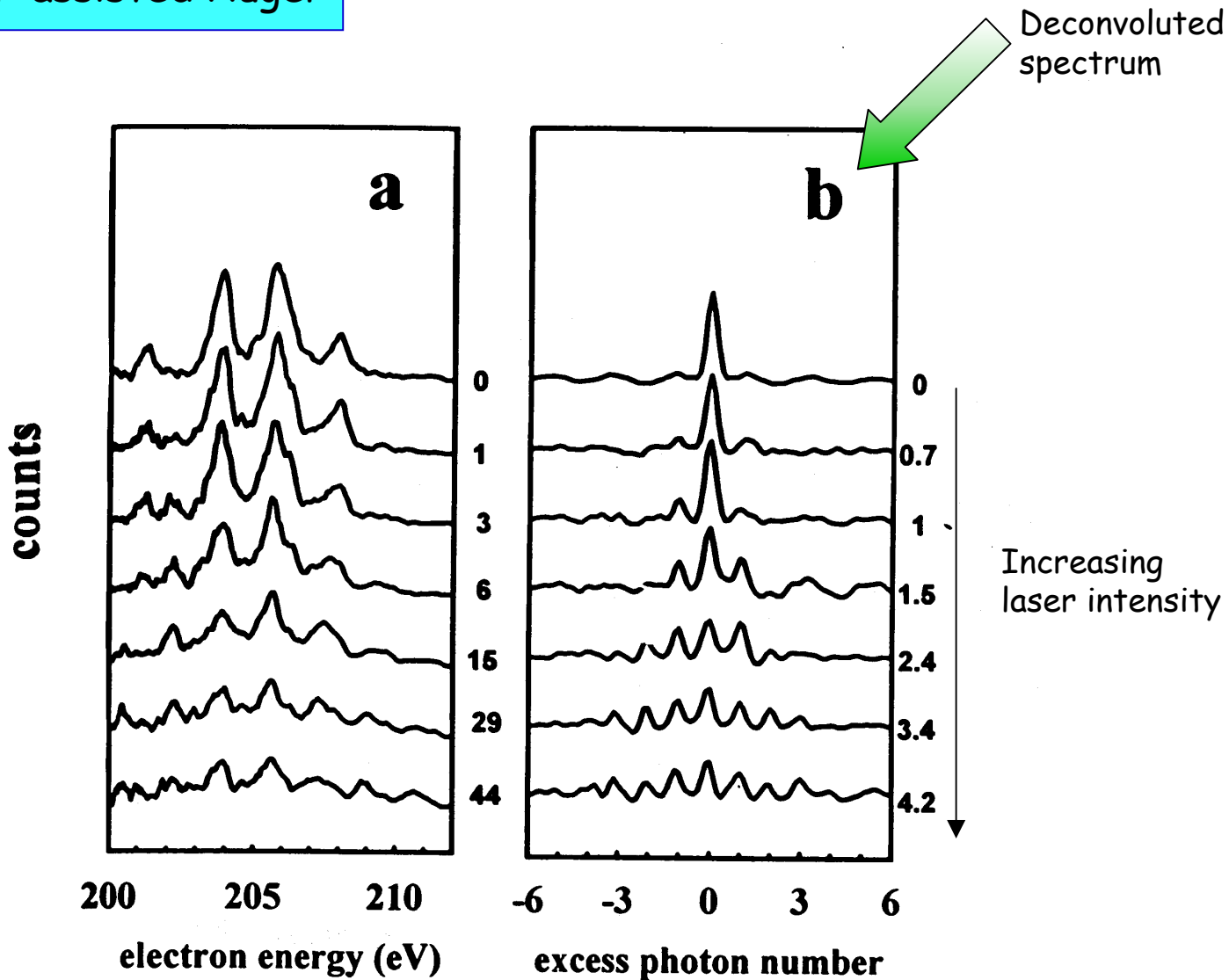


FIG. 1. Level scheme of argon, including only the states that intervene in the present experiment. Pulsed, broadband x radiation excites argon from its ground state to beyond the core-hole states Ar⁺(2p⁵), the photoelectron carrying away any excess energy. Auger transitions to the ground state of the doubly excited species yield different lines due to the fine-structure of the upper and lower states. The fine-structure splittings are given in the figure. The energy separating the members of the triplet of Ar²⁺(3p⁴) is less than the lifetime broadening of the Auger transitions.

Laser-assisted Auger

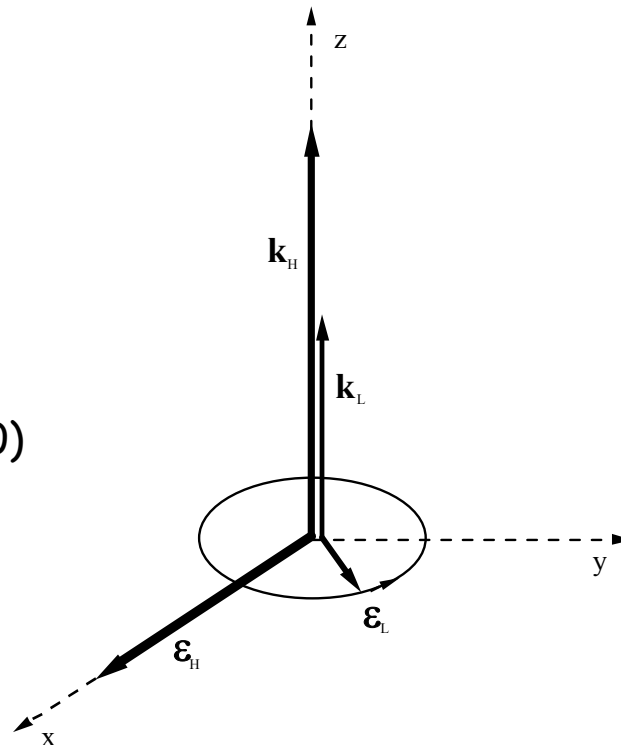


Circular dichroism from unpolarized atom in multiphoton 2-color ionization



Two-photon absorption

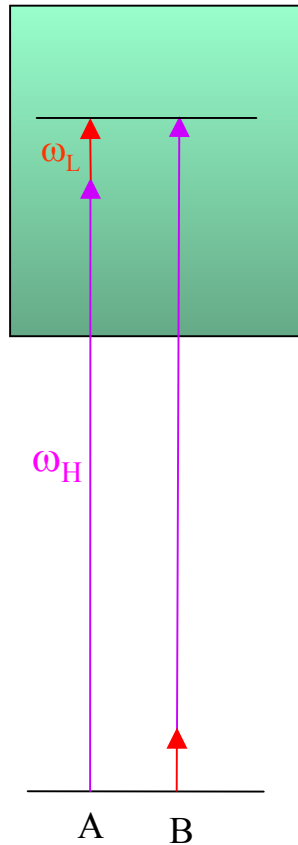
Taieb *et al.* PRA, 62 013402 (2000)
exp: Agostini *et al.*



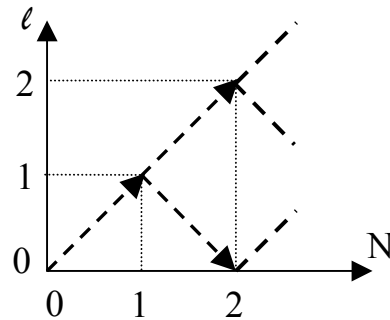
Dichroism and angular momenta

Two-photon transitions and selection rules :

$\Delta l = \pm 1$; $\Delta m = 0$ (lin. pol.), $\Delta m = +1$ (left circ. pol.), $\Delta m = -1$ (right circ. pol.),



From an s state, : accessible final states : s and d



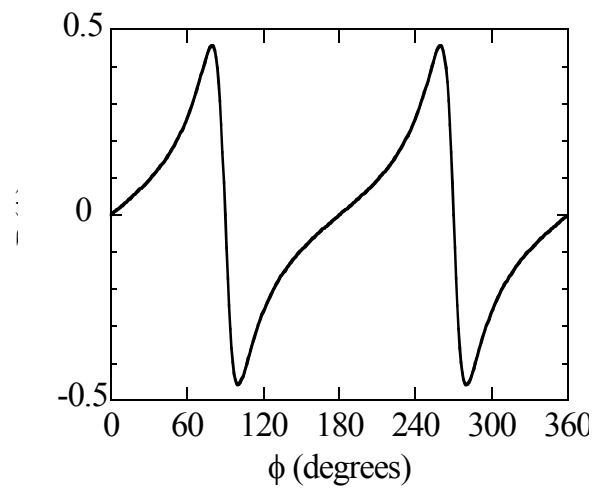
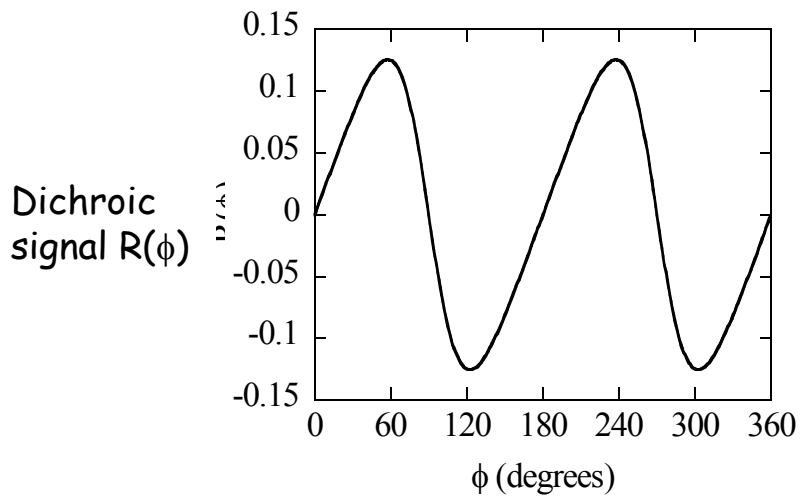
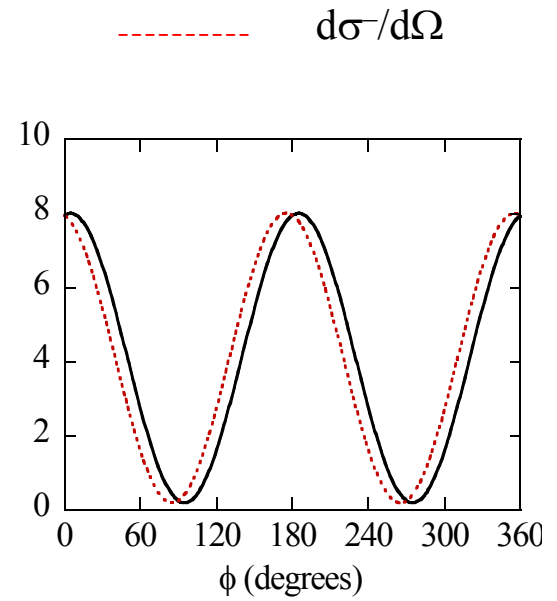
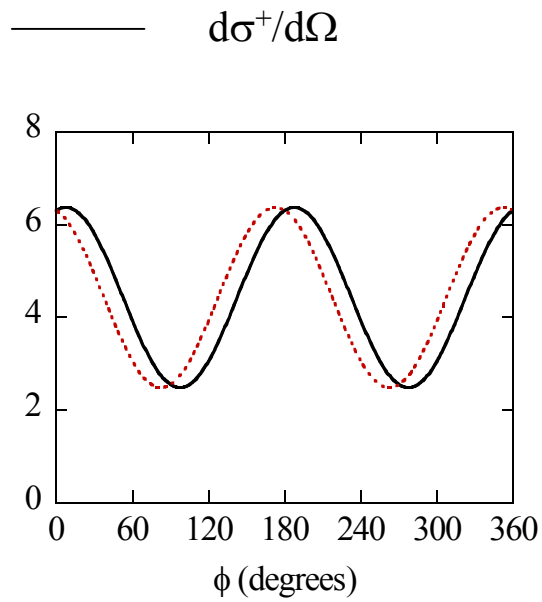
transition mplitude : $M^\pm = M_A^\pm + M_B^\pm$

with $M_{A,B}^\pm \approx e^{i\eta(0)} T_0 Y_{0,0}(\theta, \phi) + e^{i\eta(2)} T_2 Y_{2,\pm 1}(\theta, \phi)$

where: $\eta(L)$ = phase-shift of continuum states

$$d\sigma^+ - d\sigma^- \approx T_0 T_2 \sin[\eta(0) - \eta(2)] \sin(2\phi)$$

$$R(\phi) = \frac{d\sigma^+ - d\sigma^-}{d\sigma^+ + d\sigma^-} = \frac{\sin(2\phi)}{\alpha + \beta \cos(2\phi)}$$

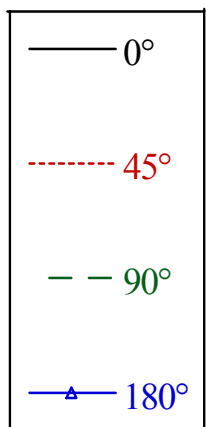
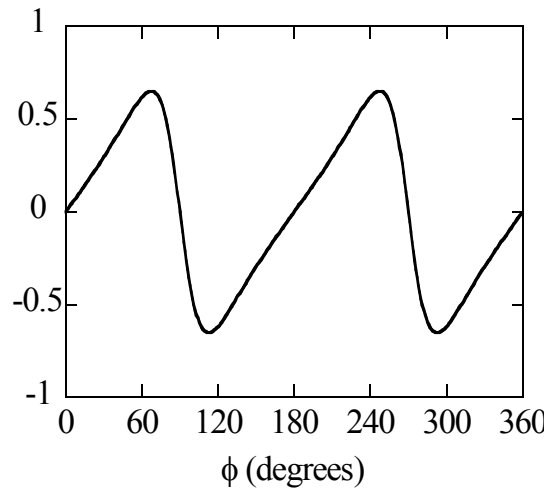
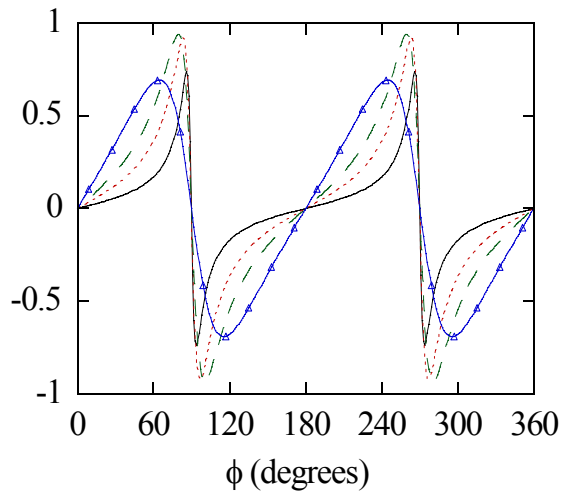
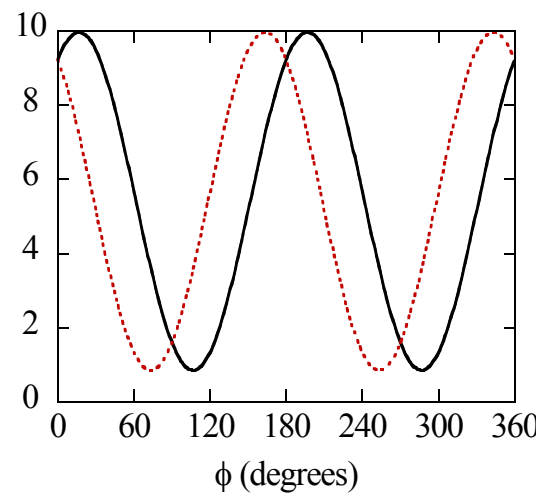
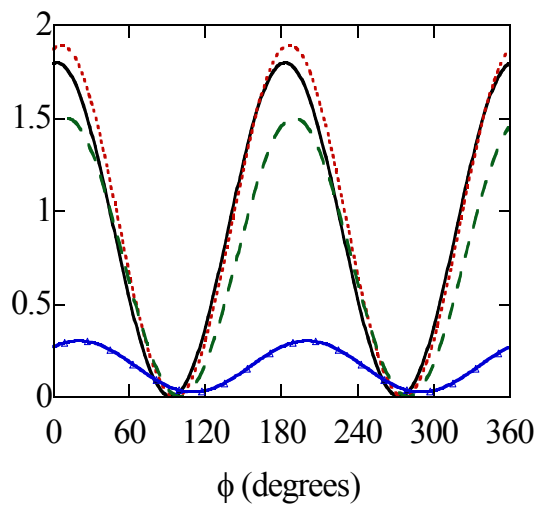


$\hbar\omega_L = 1.361 \text{ eV}; \quad \omega_H = 9\omega_L$

$\hbar\omega_L = 1.55 \text{ eV}; \quad \omega_H = 9\omega_L$

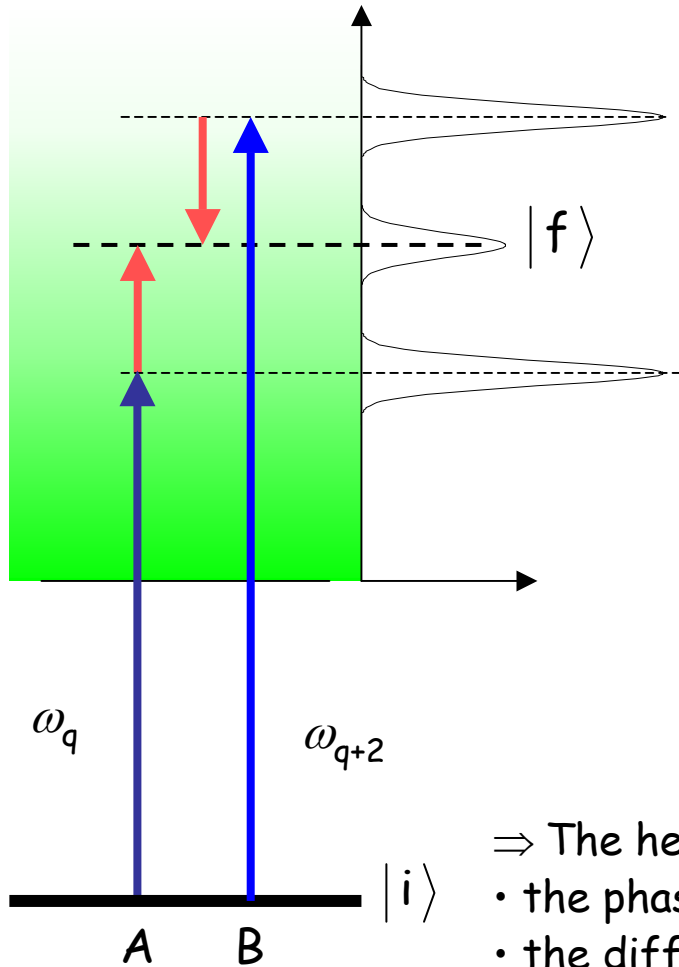
$$\hbar\omega_L = 1.55 \text{ eV}; \quad \omega_{H1} = 9\omega_L; \quad \omega_{H2} = 11\omega_L$$

$$2 \times (\hbar\omega_L = 1.55 \text{ eV}); \quad \omega_H = 9\omega_L$$



QuickTime™ et un décompresseur
GIF sont requis pour visualiser
cette image.

Quantum interferences in multicolor ir-uv ionization



Ex: two harmonics + pump laser
Véniard *et al.* PRA **54**, 721 (1996)

Transition probability amplitude (perturbative regime): $T_f \approx T_A + T_B +$ (small, non-interfering contributions)...

$$|T_f|^2 \approx |T_A|^2 + |T_B|^2 + 2 |T_A| |T_B| \cos(2\epsilon_{\text{IR}} + \epsilon_q - \epsilon_{q+2} + \varphi_A - \varphi_B)$$

where:

ϵ_{IR} = pump laser phase

$\epsilon_q, \epsilon_{q+2}$ = intrinsic harmonic phases

φ_A, φ_B = « atomic » phases

⇒ The height of the peak $|f\rangle$ depends on:

- the phase difference $\Delta\epsilon = \epsilon_q - \epsilon_{q+2}$ between the harmonics;
- the difference $\varphi_A - \varphi_B$ between the «atomic phases»;

If the latter are known, one can have access to the harmonic phases difference $\Delta\epsilon$, by monitoring the height of the peak $|f\rangle$, as a function of the pump-probe delay...

$$\omega_q = q \omega_{\text{IR}} ; q = (2n+1)$$

$$\omega_{q+2} = (q+2) \omega_{\text{IR}}$$

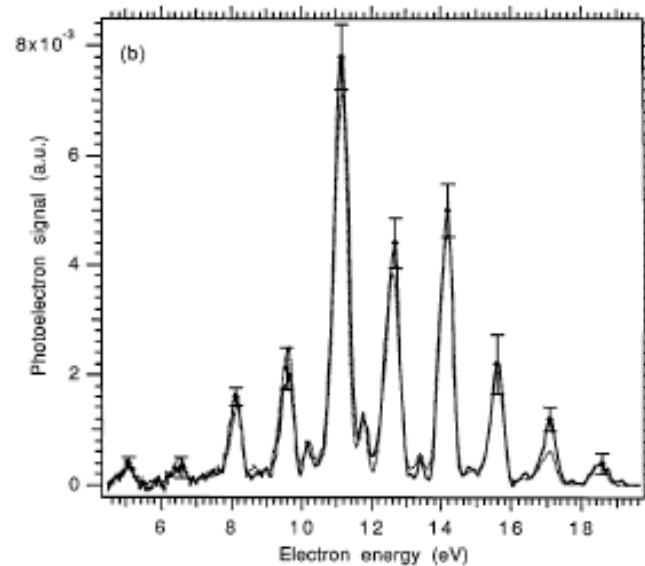
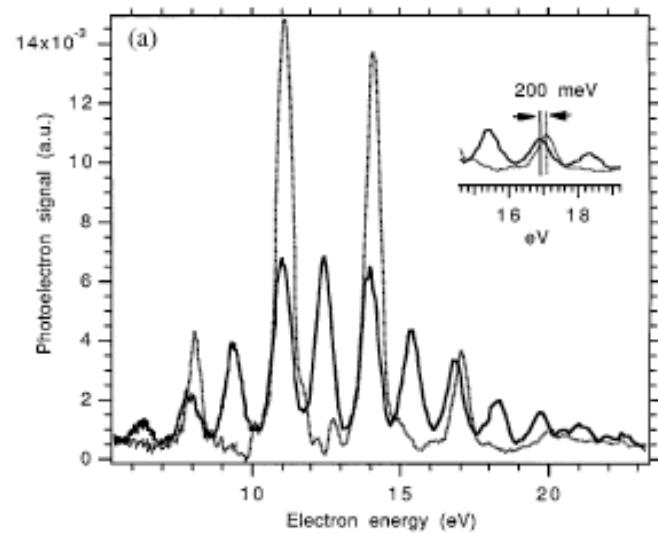
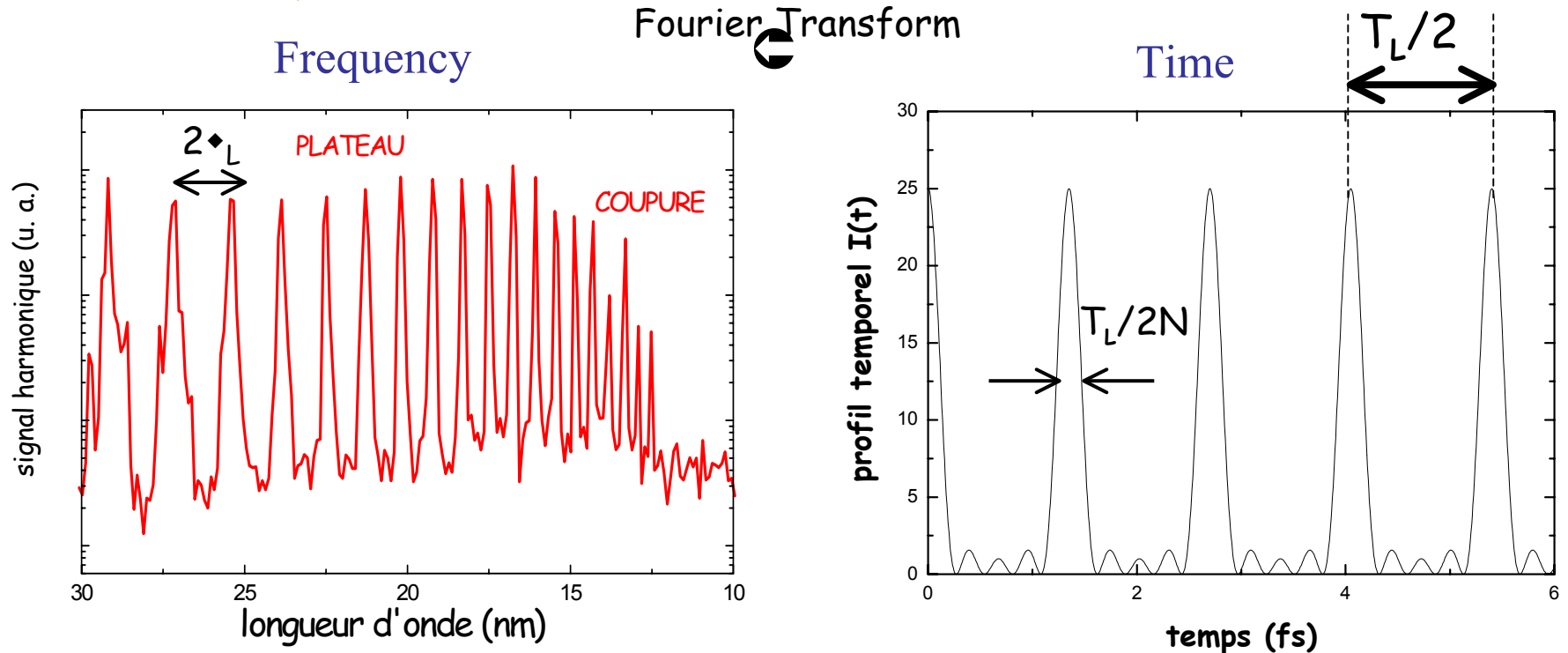


FIG. 1. (a) Photoelectron spectra obtained in the presence (solid line) and absence (dashed line) of the laser pulse. The inset shows an expanded view indicating a ponderomotive shift in the photoelectron peak positions. (b) Measured photoelectron spectrum obtained in the presence of the laser field (solid line). The dashed curve is a theoretical prediction discussed in the text.

First experiment using
harmonics + ir laser in
photoemission spectra:
Glover et al. PRL, 76, 2468
(1996)

Pump-probe characterization of subfemtosecond («attosecond») harmonic pulses

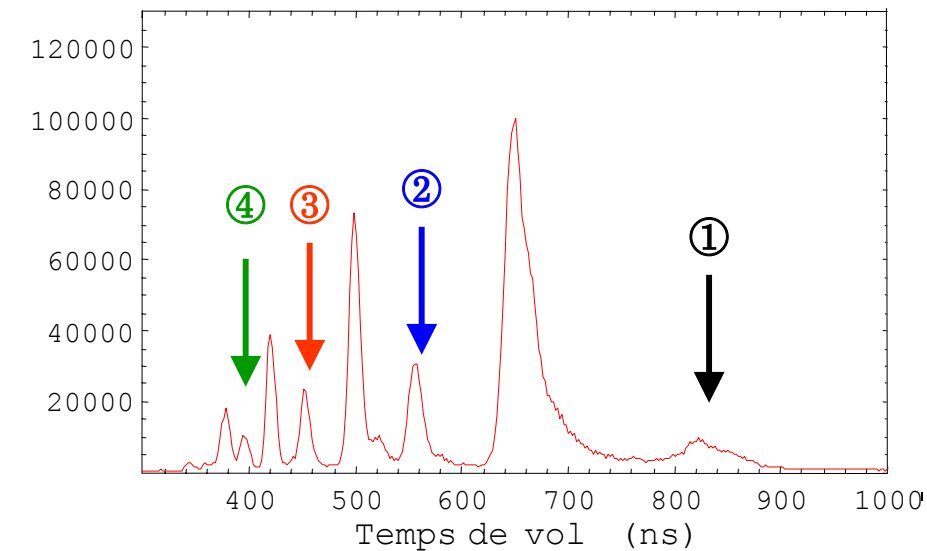
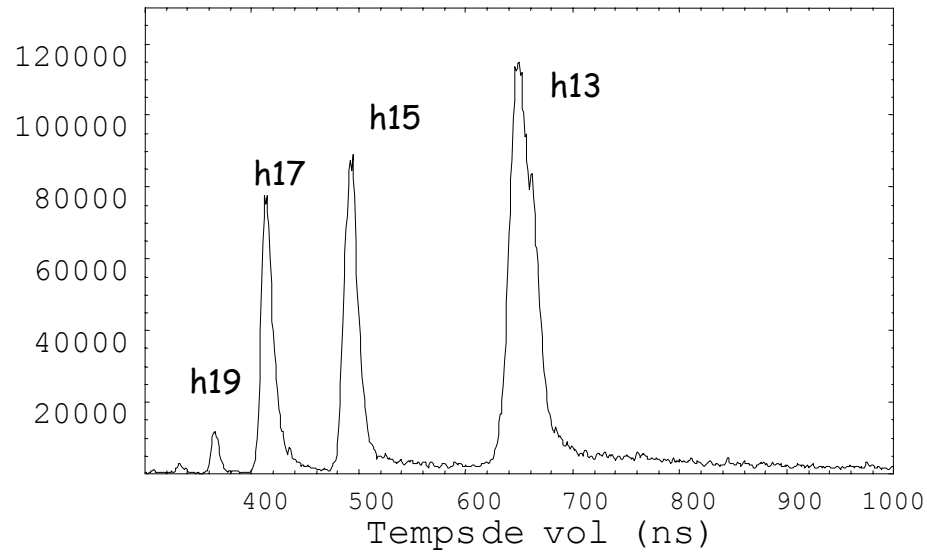
Farkas and Toth (*Phys. Lett. A* 1992),
 Harris et al. (*Opt. Commun.* 1993)



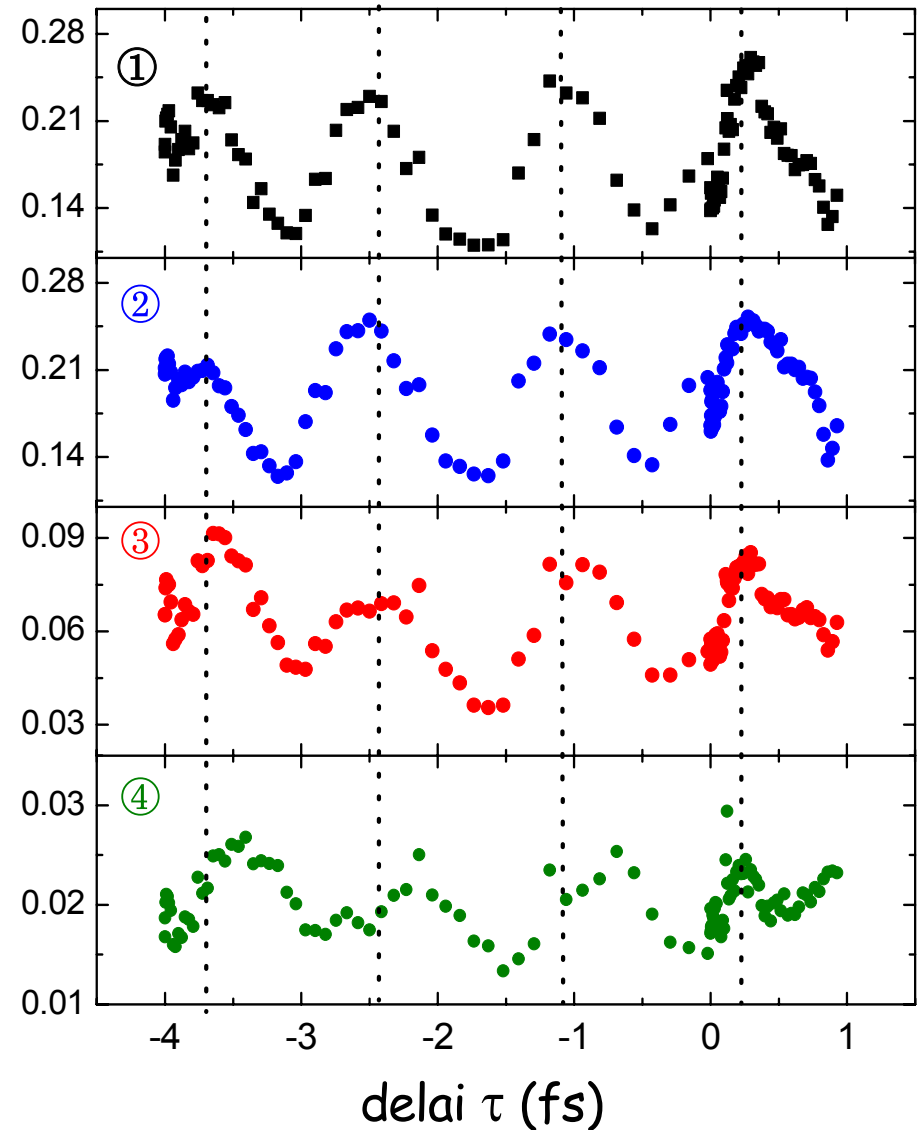
$$I(t) = \left| \sum_{q_0}^{q_0+N-1} A_q e^{-i[(2q+1)\omega t + \varphi_q]} \right|^2 = \left(\frac{\sin[N(\omega t + \varphi)]}{\sin(\omega t + \varphi)} \right)^2$$

Experiment: P-M Paul *et al.* Science 292, 1689 (2001)

Photoelectron spectra

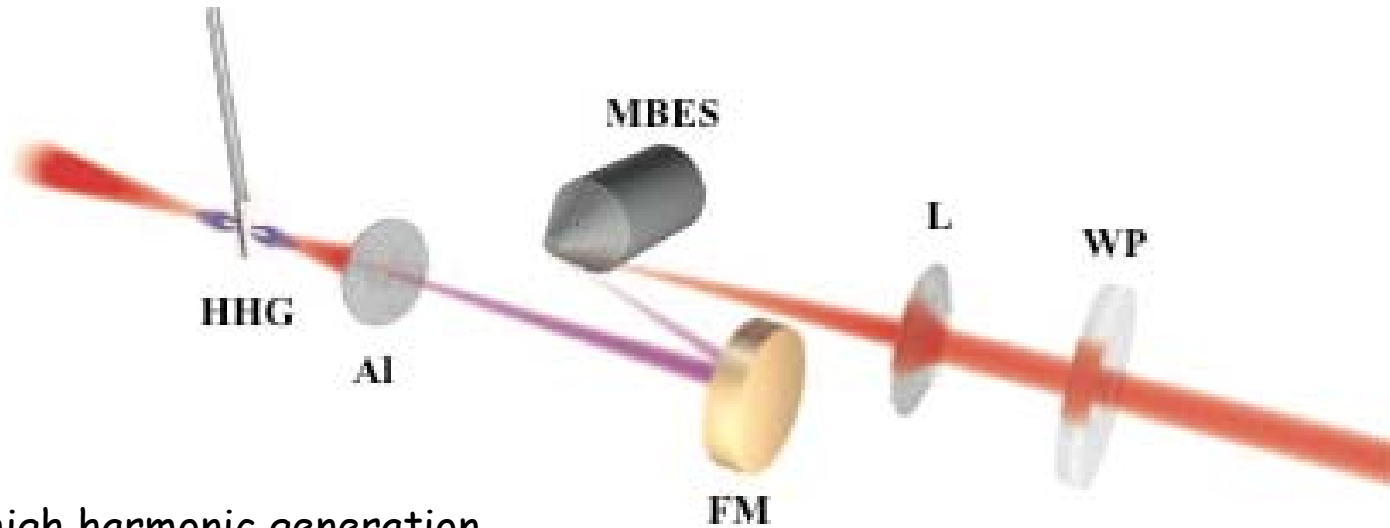


Normalized amplitudes of the sidebands



Polarization and phase effects in two-photon UV-IR ionization

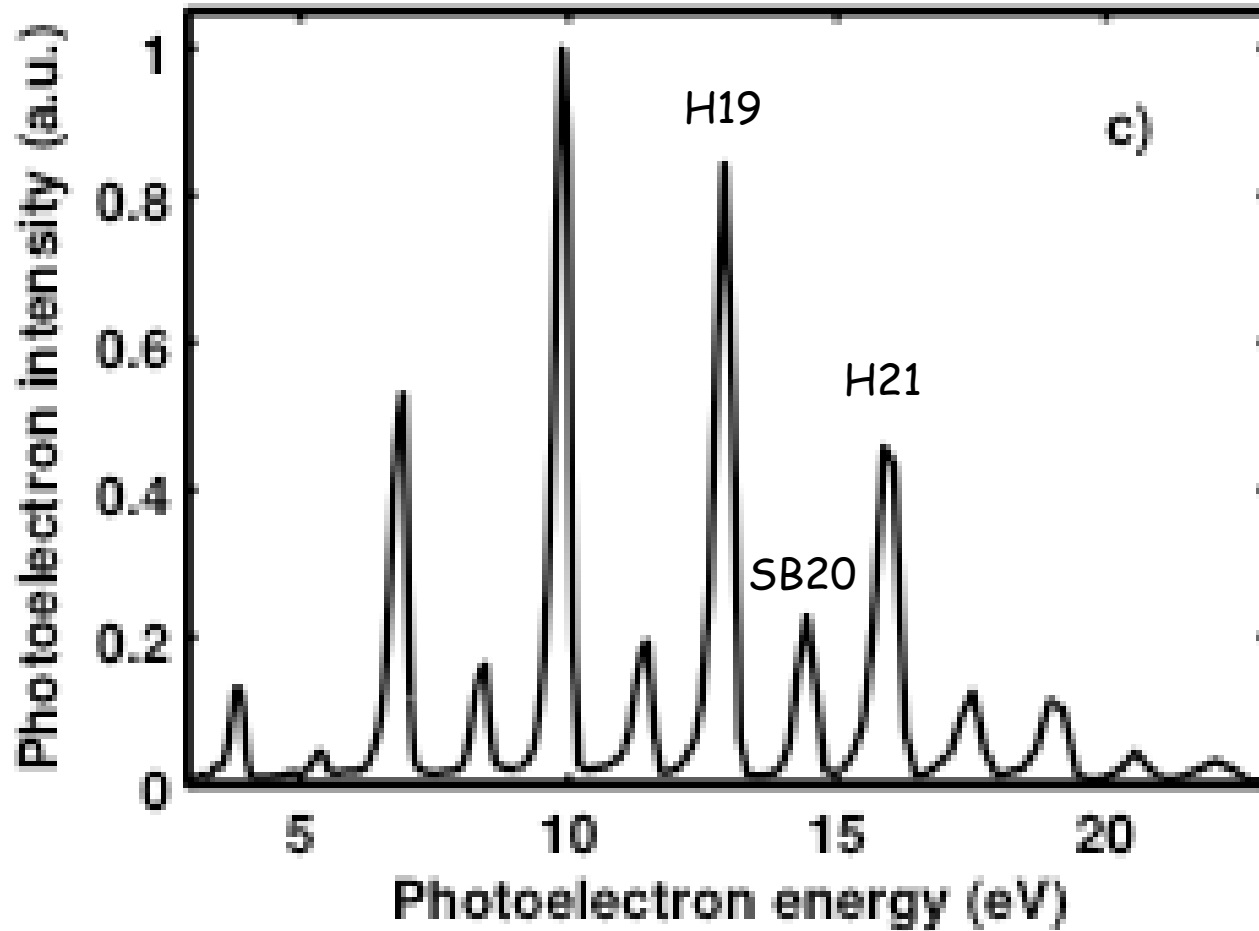
Using polarization effects for determining the harmonic and «atomic» phases?



HHG = high harmonic generation
MBES = electron spectrometer
FM = focussing mirror
WP = retarding wave plate

LURE (Orsay), Lund, LCP-MR (Paris), SPAM (Saclay) Collaboration

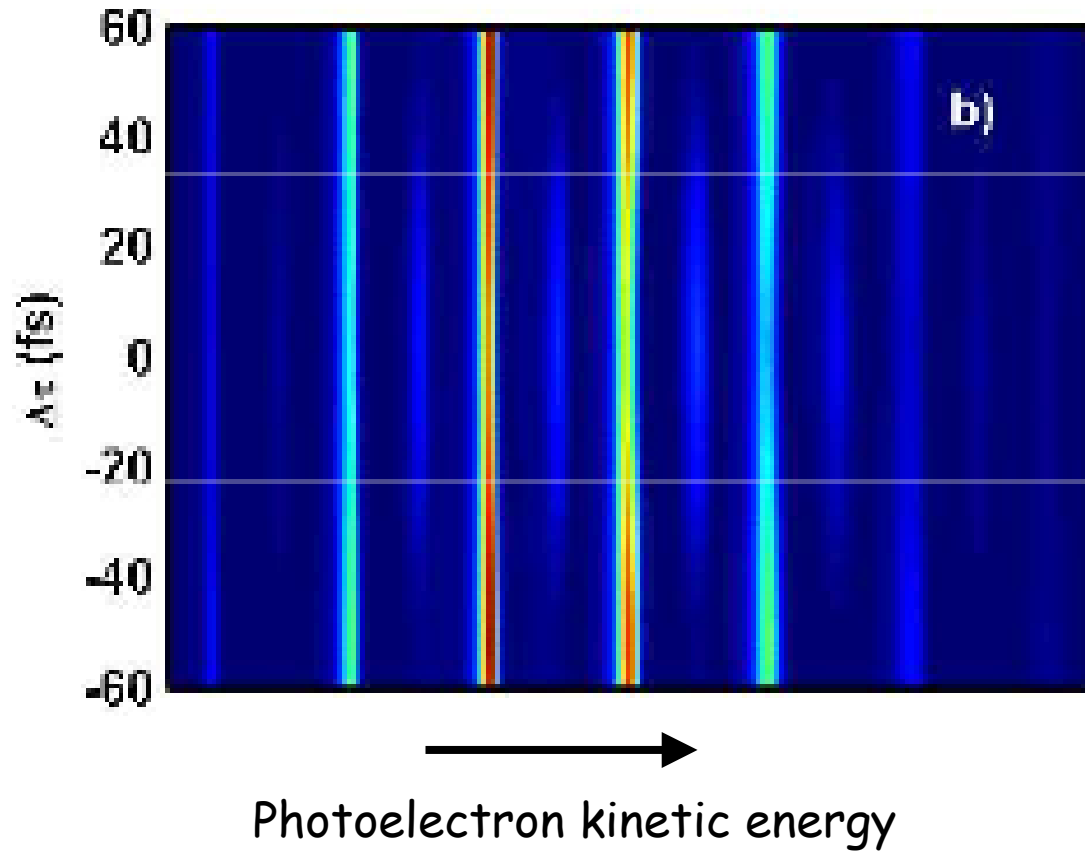
A typical photoelectron spectrum in Ar



The magnitudes of the sidebands depend on:

- the time delay between the IR and UV pulse
- and also on the respective polarization orientations of the fields

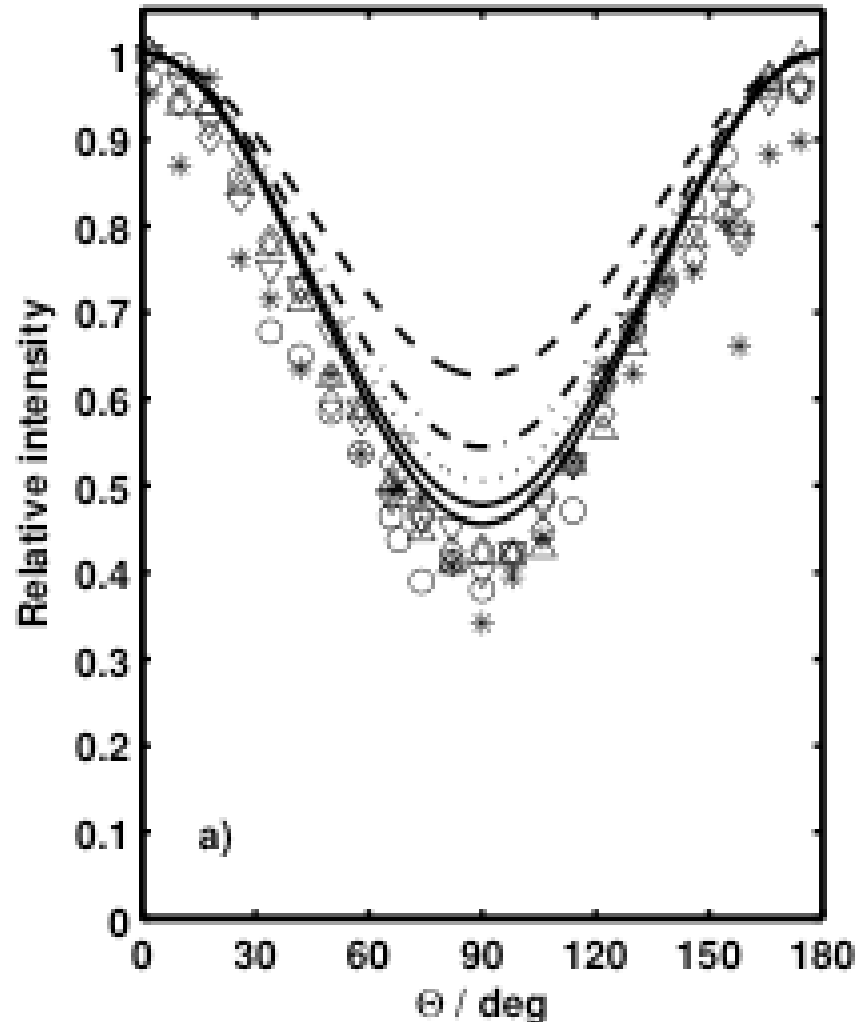
Role of the time delay on the magnitudes of the sidebands



Influence of the angle between the polarizations of the two fields on the magnitudes of the sidebands

Case of two linear polarizations with different orientations:
Comparison between theory (soft-photon approximation) and experiment.

(As expected, the soft-photon approximation provides better results for higher sidebands).

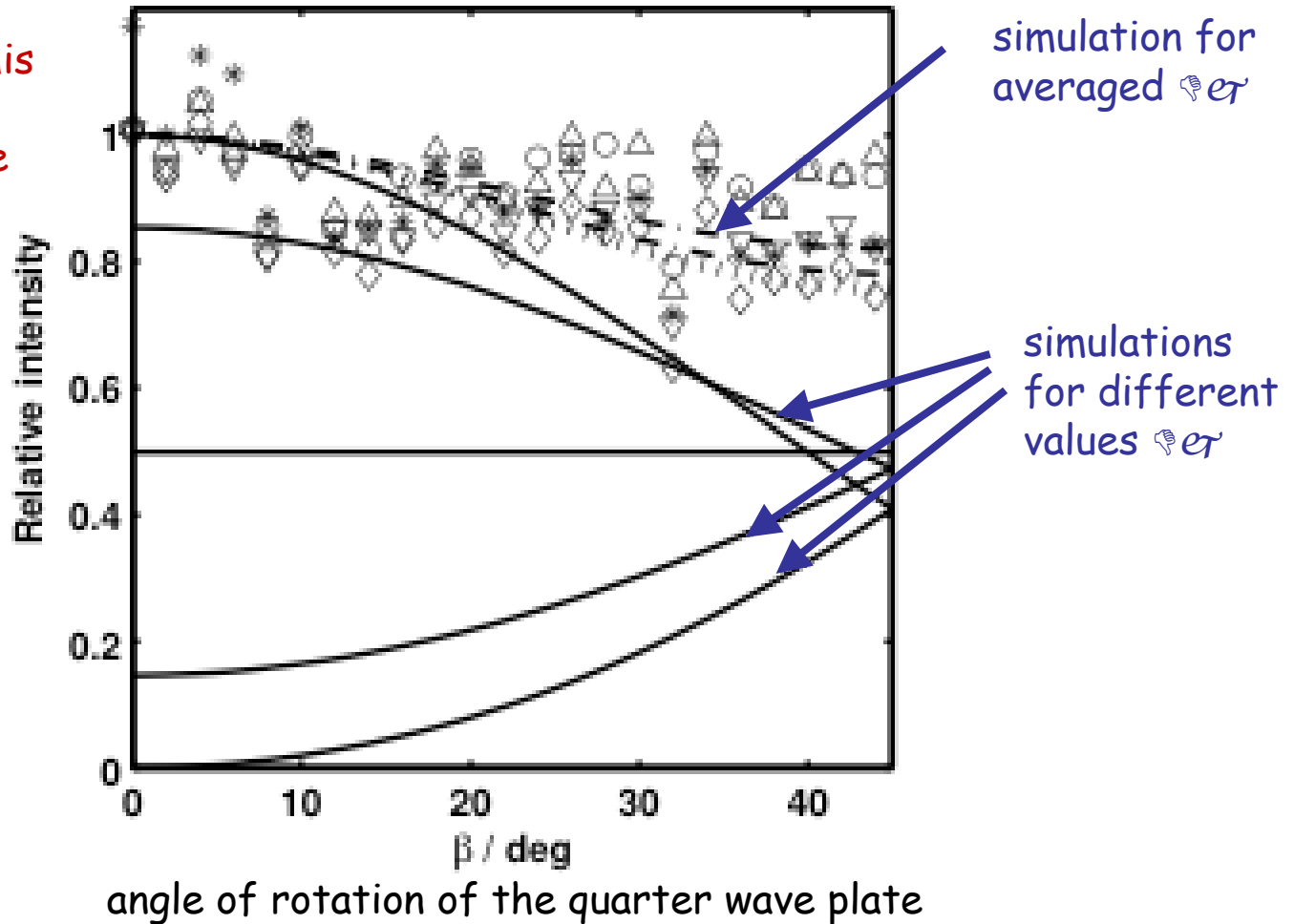


$$\theta = (\varepsilon_L, \varepsilon_H)$$

Influence of the ellipticity of the laser field on the magnitudes of the sidebands

With the help of this scenario, one can in principle, determine the atomic and harmonic phases, (O'Keefe *et al.* to appear).

= a challenge for theory!



On the «atomic phases»

Two-photon matrix element for «Above-Threshold Ionization» (ATI):

$$T_A = \langle \varphi_{f,\bar{k}} | D_L | G(E_i + \omega_q) | D_q | \varphi_i \rangle$$

As the argument of the Green's function $E_i + \omega_q > 0$, $G(E_i + \omega_q)$ reads:

$$G(E_i + \omega_q) = \sum_n \frac{|\varphi_n\rangle\langle\varphi_n|}{E_i + \omega_q - E_n} + P \int_0^{+\infty} dE \frac{|\varphi_E\rangle\langle\varphi_E|}{E_i + \omega_q - E} - i\pi\delta(E_i + \omega_q - E)$$

and T_A becomes:

$$T_A = \sum_n \frac{\langle \varphi_{f,\bar{k}} | D_L | \varphi_n \rangle \langle \varphi_n | D_q | \varphi_i \rangle}{E_i + \omega_q - E_n} + P \int_0^{+\infty} dE \frac{\langle \varphi_{f,\bar{k}} | D_L | \varphi_E \rangle \langle \varphi_E | D_q | \varphi_i \rangle}{E_i + \omega_q - E} - i\pi \langle \varphi_{f,\bar{k}} | D_L | \varphi_{E_i + \omega_q} \rangle \langle \varphi_{E_i + \omega_q} | D_q | \varphi_i \rangle$$

Here φ_f and φ_E are continuum wave functions with phase-shift factors $\exp(i\eta_\nu)$, that are energy and angular momentum dependent.

[See, for instance: Kennedy and Manson, PRA **5**, 227 (1972)]

There is, in addition, a phase depending on the respective magnitude of the $-i\pi\delta$ term, as compared to the remaining.

⇒ The determination of θ_A in $T_A = |T_A| \exp(i\theta_A)$ is a challenge for theory!

Time-resolved atomic inner-shell spectroscopy with IR laser + «attosecond» XUV pulses

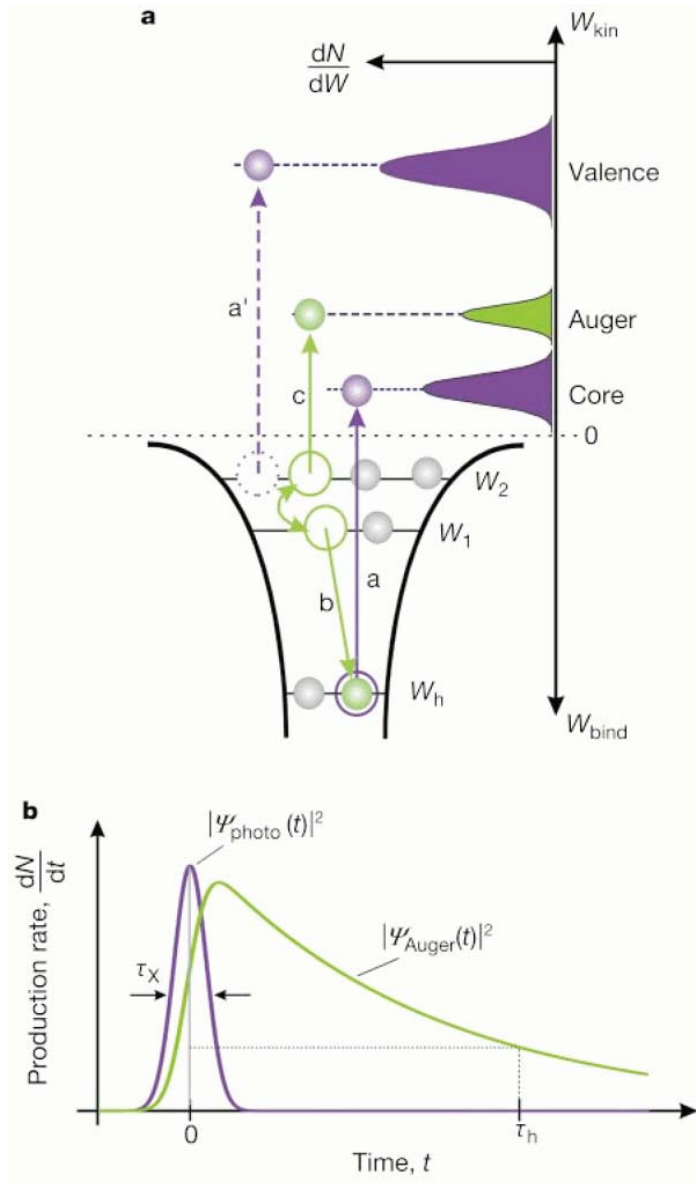
Drescher *et al.* Nature, **419**, 803 (2002)

two-colour ionisation (x-ray + ir laser):

- an «attosecond» x-ray pulse creates a core-hole

- the ejected electrons are "dressed" by the infrared laser;

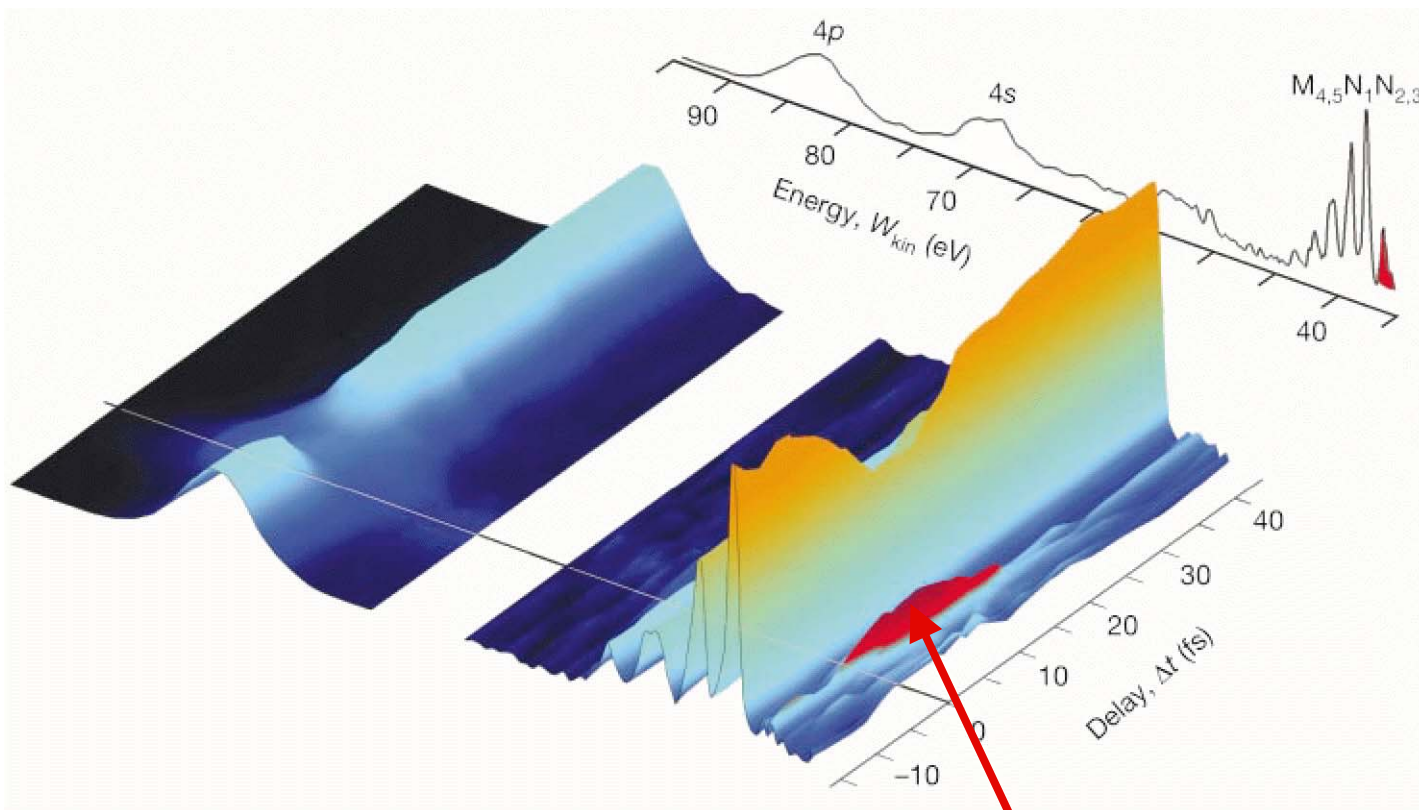
one monitors the side-bands seen in the Auger electron spectrum in terms of the time delay between the two pulses ($\tau_x \ll \tau_{ir}$)



Soft x-ray atomic ionisation (Xe M shell).
 Population density in continuum:
 (a) inner-shell
 (a') outer-shell
 Core-hole relaxation through
 Auger decay:
 (b) vacancy filling
 (c) Auger electron

Typical time dependence of the photoelectron and Auger wavepackets:
 τ_x = x-ray pulse duration,
 τ_h = core-hole decay time

Dependence of the ejected electron spectrum (photoelectrons and Auger) on the time delay between the x-ray and ir pulses



sideband

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

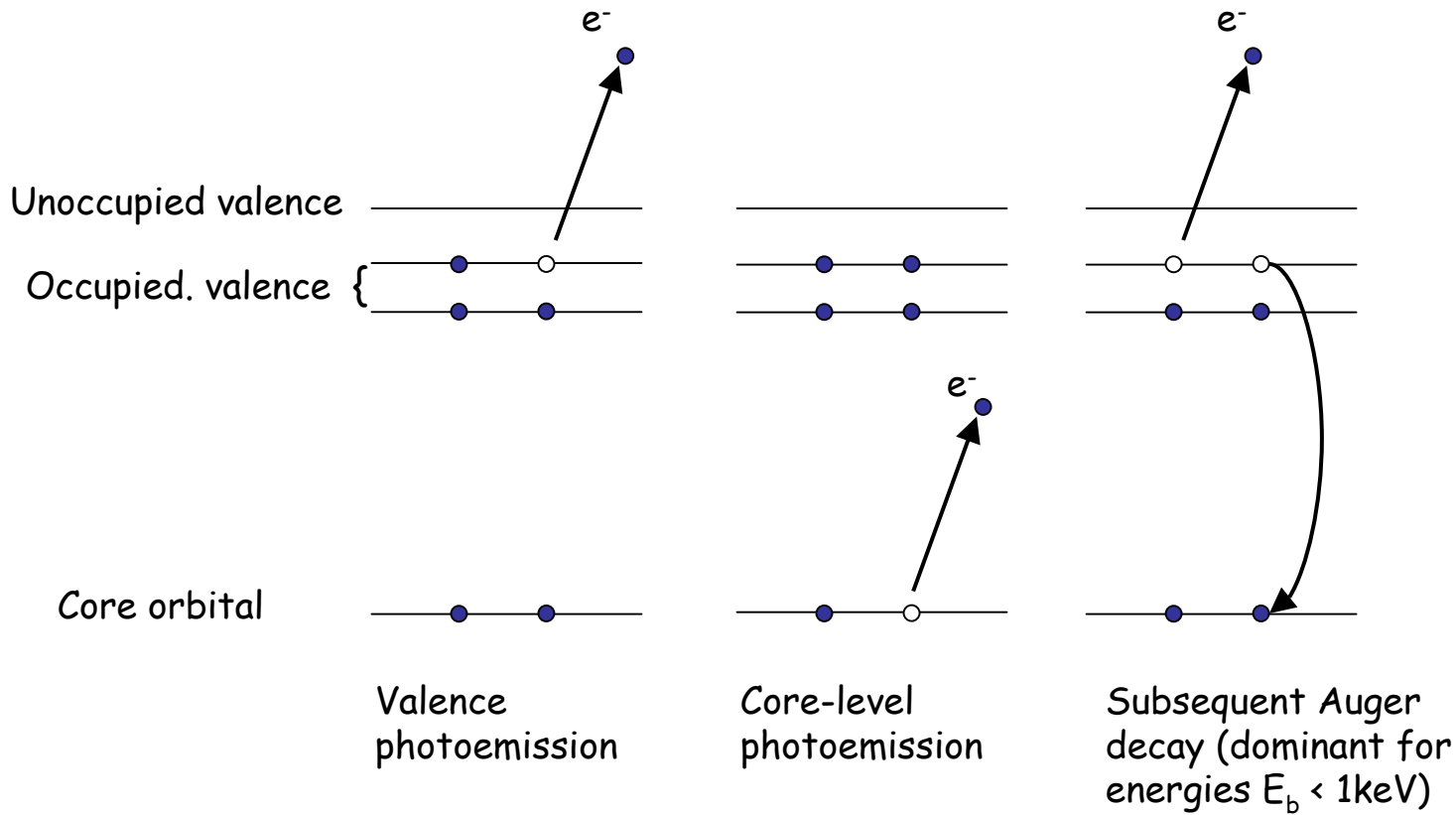
The advent of soft x-ray sources with pulse durations in the femto- and attosecond ranges opens new perspectives for **pump-probe** spectroscopies involving atomic inner-shells!

However, new resonance spectroscopies are currently developed on 3rd generation synchrotron sources:

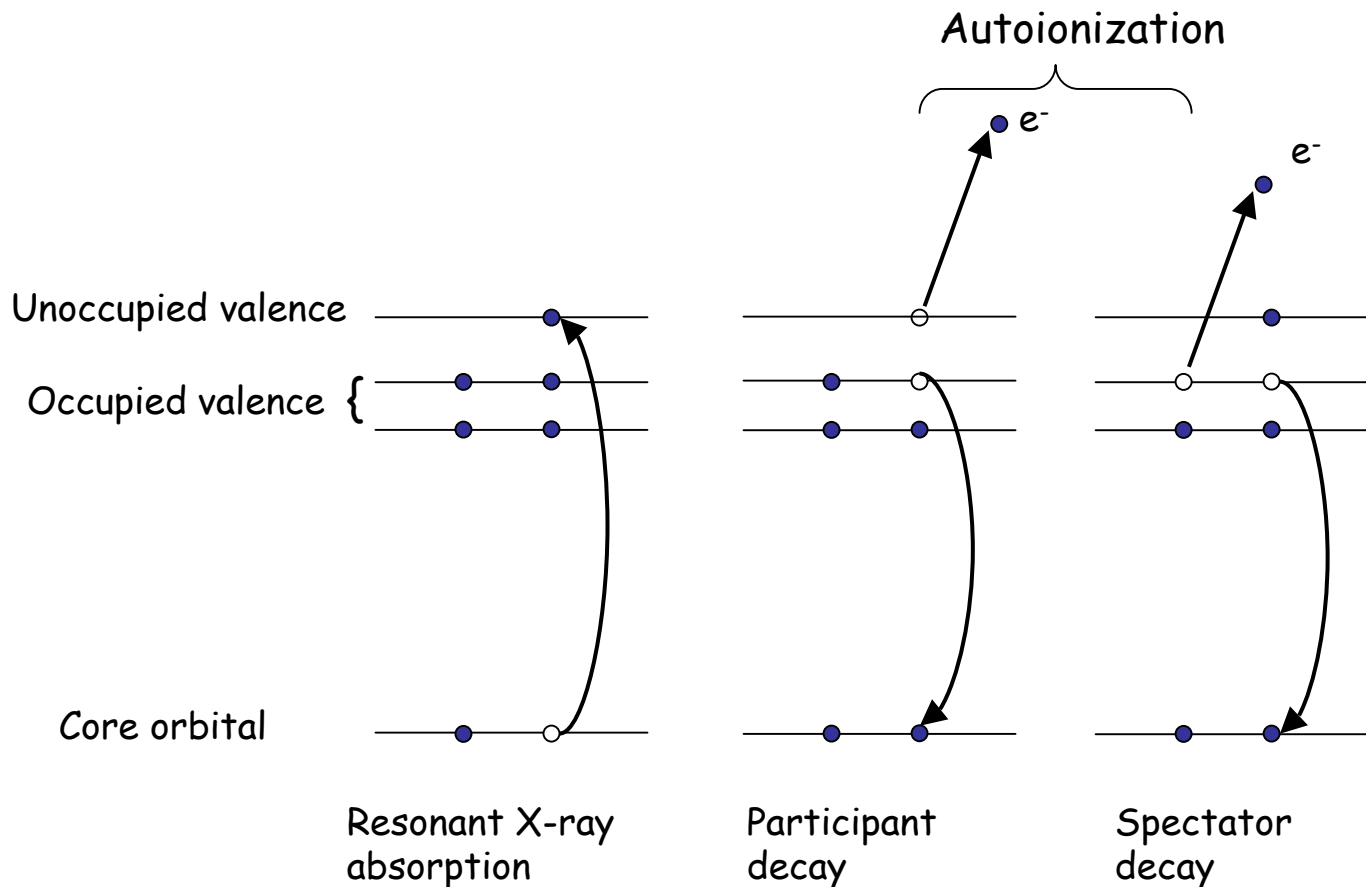
- Resonant Inelastic X-ray Scattering (RIXS) (photon in- photon out)
- Resonant Auger Spectroscopy, etc.

They give access to sub-femtosecond lifetimes via **«excitation-decay»** measurements in the energy domain. The time scale is set by the **«core-hole clock»**, i.e. by the intrinsic lifetime of the intermediate state.

Higher frequencies: Core-electron excitation and deexcitation



Higher frequencies: Core-electron excitation and deexcitation (follows)



In condensed matter (cluster, solids, surfaces, etc), the relative importance of these processes depends also on the strength of the coupling of the valence states with continua, See for instance Brühwiler et al. RMP **74**, 703 (2002)