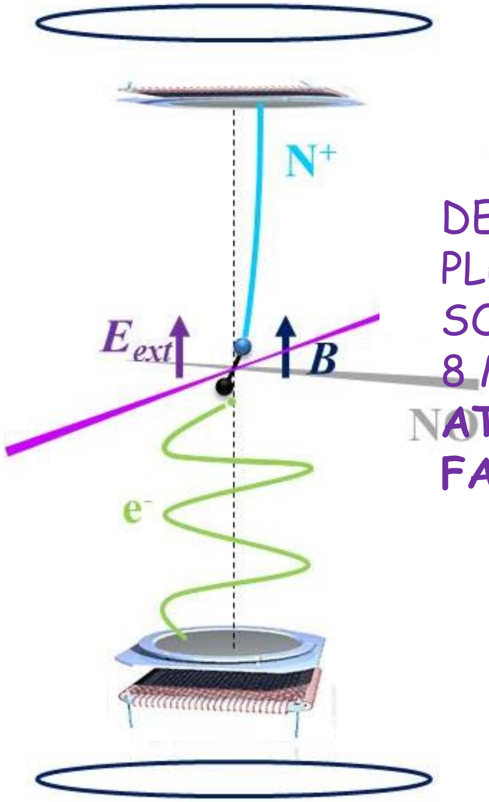
Angularly resolved RABBITT experiments in the gas phase on FAB 10

Danielle Dowek, Institut des Sciences Moléculaires d'Orsay (ISMO)

(V_{A+}, V_e, P)



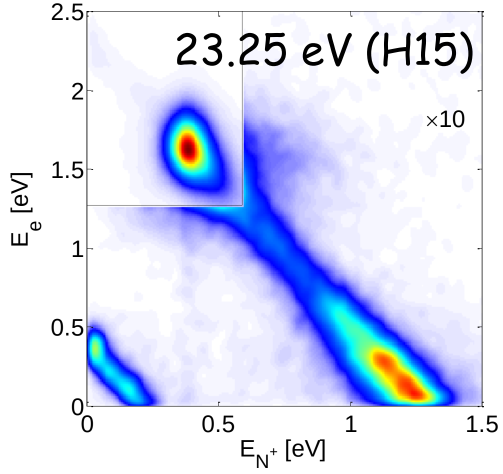
DESIRS
PLEIADES
SOLEIL
8 MHz
ATTOLAB
FAB10

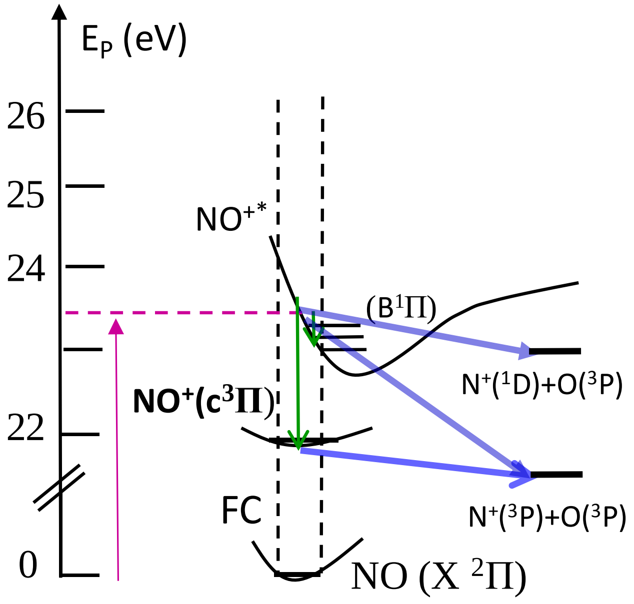
Dissociative photoionization of simple molecules

$ABC(X) + h\nu\ (XUV, P) \rightarrow ABC^{+*} + e\ (V_e) \rightarrow A^{+*}(V_e) + BC + e\ (V_e)$

Energy and angular **observables** (laboratory and Molecular Frame)

Kinetic energy correlation diagram
(e,A^+) **KECD** resolving power





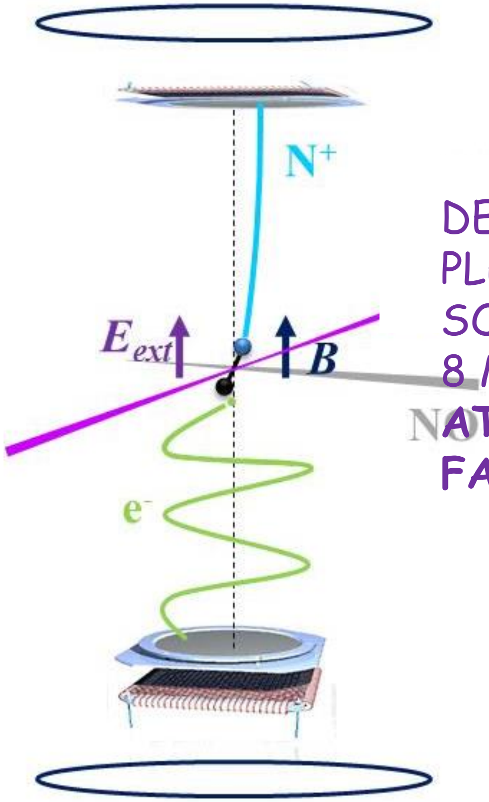
Identify ionic states and dissociation limits
Branching ratios

Gisselbrecht et al., RSI 76, 013105 (2005)
Lebech et al., RSI 73, 1866 (2002)
S.J. Weber et al RSI 86, 033108 (2015)

→ **Molecular Frame Photoelectron Angular Distribution**

Photoionization of molecules: Electron-ion coincidence 3D momentum Spectroscopy

(V_{A+}, V_e, P)



DESIRS
PLEIADES
SOLEIL
8 MHz
ATTOLAB
FAB10

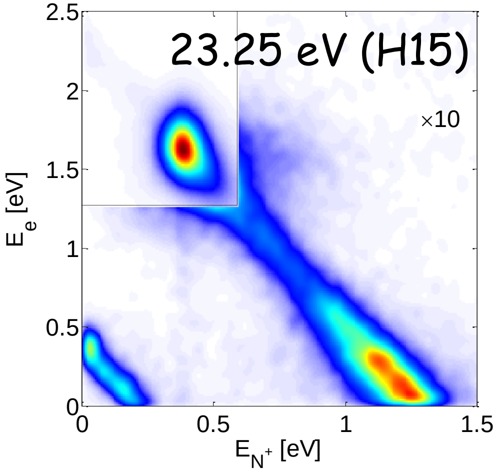
n events/pulse << 1

Dissociative photoionization of simple molecules

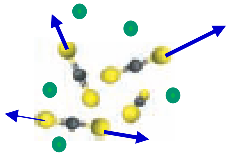
$ABC(X) + h\nu\ (XUV, P) \rightarrow ABC^{+*} + e\ (V_e) \rightarrow A^{+*}(V_e) + BC + e\ (V_e)$

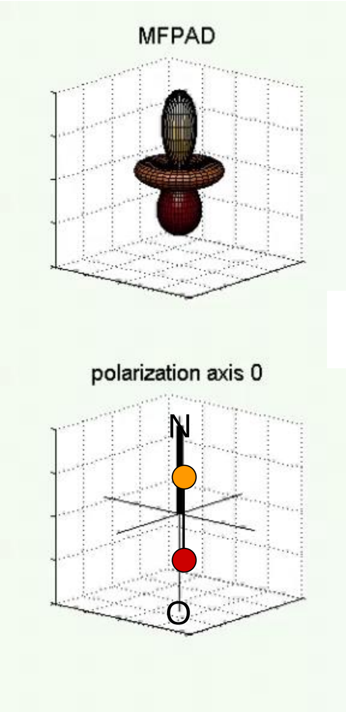
Energy and angular **observables** (laboratory and Molecular Frame)

Kinetic energy correlation diagram
(e,A⁺) **KECD** resolving power



MFPAD





$T_{fi}(\theta_e, \phi_e, \chi)$

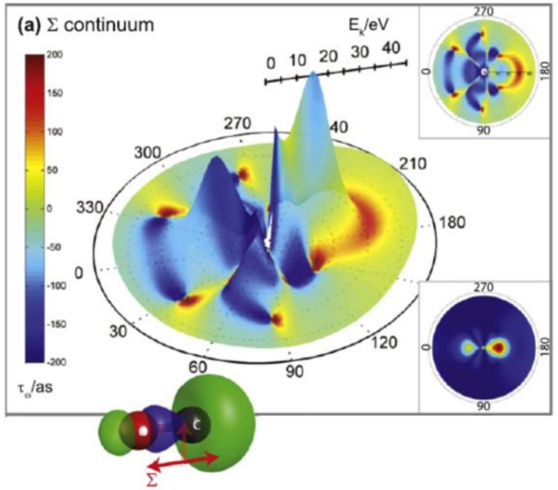
$$I^{M_i M_f}_{lm\mu} = \langle \psi^i_{M_i} | d_\mu | \phi^f_{M_f} \psi^{(-)}_{lm} \rangle$$

$\approx d_{lm\mu} \exp(-\eta_{lm\mu})$

Complete experiments: access to the complex dipole matrix elements
Valence and inner shell photoionization
Photoionization and photo dissociation dynamics

Motivation: Time delays in molecular photoionization, angularly resolved in the MF

CO : 5σ → kσ



V(r, ϑ, φ)

Hockett et al., *J.Phys.B* 49 095602 (2016)

D. Baykusheva and H.J. Wörner., *JCP. 146, 124306 (2017)*

Vos et al., *Science* 360 1326 (2018)

Electron wave packet: $\Psi_g = \Sigma_{l,m}\, \Psi_{l,m}$

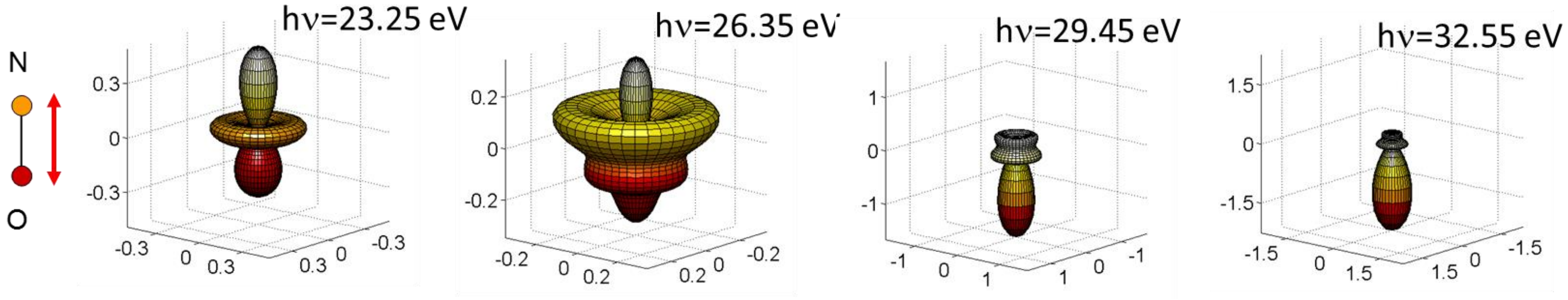
$$I_{lm\mu}^{M_i M_f} = \langle \psi_{M_i}^i | d_\mu | \phi_{M_f}^f \psi_{lm}^{(-)} \rangle \approx d_{lm\mu} \exp(-\eta_{lm\mu})$$

Photoionization amplitude $$A_{MF}(\varepsilon, \hat{k}_e, \hat{R}_\chi) = \sum_{lm\mu} I_{lm\mu} Y_{lm}(\hat{k}_e) D_{\mu m_p}(\hat{R}_\chi)$$

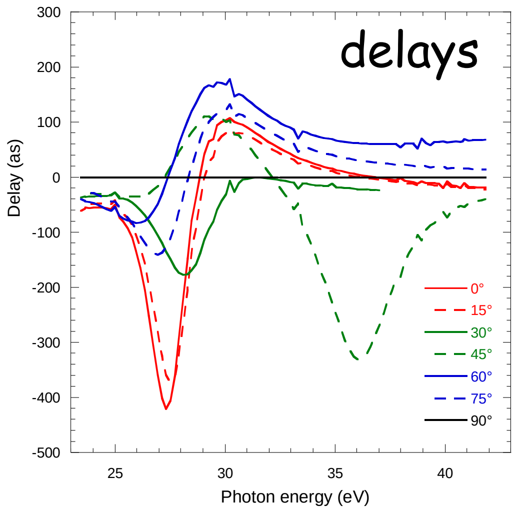
Time delay $$\tau_{W-MF}(\varepsilon, \hat{k}_e, \hat{R}_\chi) = \frac{\partial}{\partial \varepsilon} \arg\left\{ \sum_{lm\mu} I_{lm\mu} Y_{lm}(\hat{k}_e) D_{\mu m_p}(\hat{R}_\chi) \right\}$$

NO ($X^2\Pi$) + **hν (APT)→** $NO^+(c^3\Pi$ **$4\sigma^{-1}$**)

Résonance de forme : 4σ → kσ



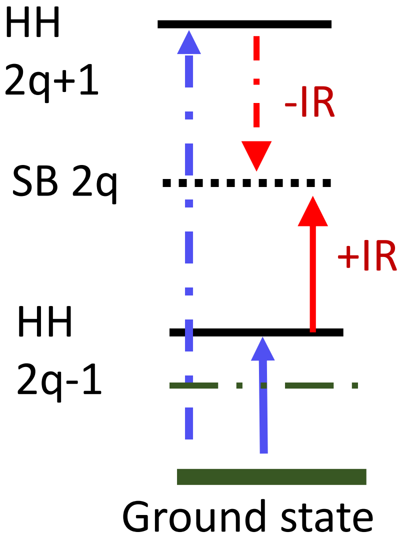
Veyrinas et al., *Faraday Discussions*, 2016, **194**, 161 - 183

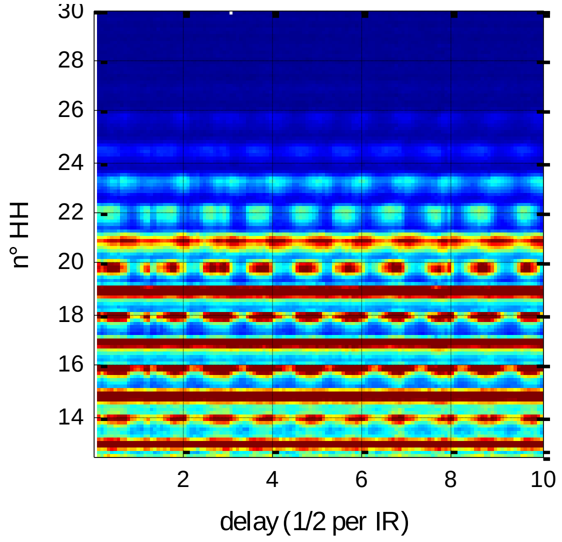


R.R. Lucchese private com.

Two-photon XUV-IR photoionization in the RABBITT scheme

Reconstruction of Attosecond Beating by Two-photon Transition: XUV APT (comb of HHs) and IR





$$I_{SB}(\tau) = A + B\cos(2\omega\tau + C)$$

$$C = \Delta\phi_{HH} + \Delta\phi_{A}$$

$$\Delta\phi_{A} = \Delta\phi_{W} + \Delta\phi_{cc}$$

$$\tau_{W} = \Delta\phi_{W}/2\omega$$

$$\tau_{W}(\varepsilon) = \frac{\partial \delta_l}{\partial \varepsilon}$$

Wigner Phys. Rev. 98 145 (1955)
Smith Phys. Rev. 118 349 (1960)

$$I_{SB}(\theta,\tau) = A(\theta) + B(\theta)\cos\left(2\omega\tau + C(\theta)\right)$$

Véniard, V. *et.al*. (1996). *Physical Review A*, *54* (1), 721.
Paul, P. M., et al. *Science* (2001) 292, 1689.

ES Toma HG Muller J. Phys.B (2002) 35, 3435

Haessler et al., *PRA* 80, 011404 (2009)
Klünder et al., *PRL* 106, 143002 (2011)
Dahlström et al Chem. Phys. 414, 53 (2013)

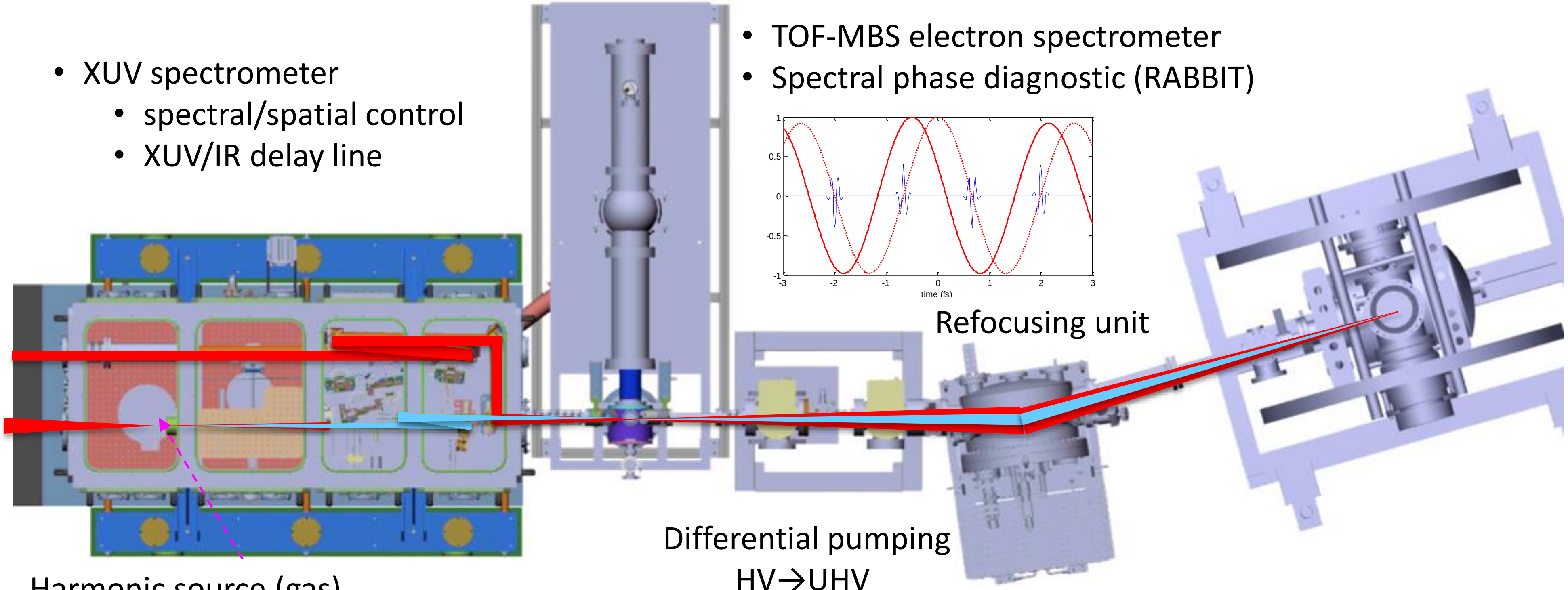
Atomic target:

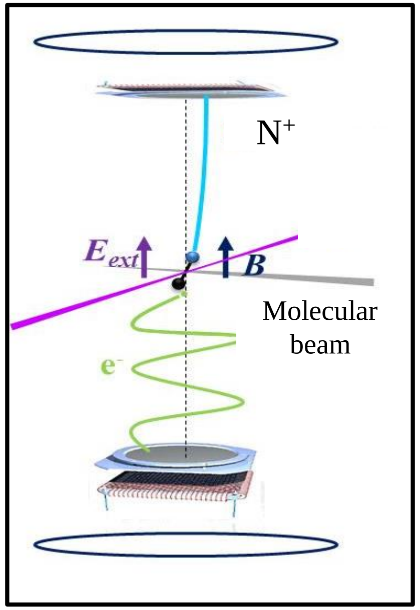
Aseyev, S. A., et al. *PRL* 91.22 (2003): 223902
Picard, Y. J. et al. *Phys. Rev. A* 89, 031401 (2014)
Heuser, S. et al. Phys. Rev. A 94, 063409 (2016)
Cirelli. et al. Nat. Commun. 9, 955 (2018)
Bray, A. W.et.al.Phys. Rev. A 97, (2018).
Ivanov, I. A. & Kheifets, A. S. *Phys. Rev. A* 96, (2017).
Loriot et al J. Opt. 19, 114003 (2017)
Fuchs et al arXiv: 1907.03607 v1 [physics. atom-ph]

Electron-ion coincidence momentum spectroscopy @ ATTOLAB-FAB10 XUV-IR beamline

- XUV spectrometer
  - spectral/spatial control
  - XUV/IR delay line

- TOF-MBS electron spectrometer
- Spectral phase diagnostic (RABBIT)





- 3 modes of XUV spectral / temporal selection

Type	E	ΔE	Δτ	XUV / IR delay
Very broadband	10-100 eV	10-30 eV	100 as	< 100 as
Broadband	32, 54, 91 eV	1-5 eV	1 fs	100 as
Narrowband	5-35 eV 30-60 eV 50-85 eV	100 meV	10 fs	fs

NO and O_2

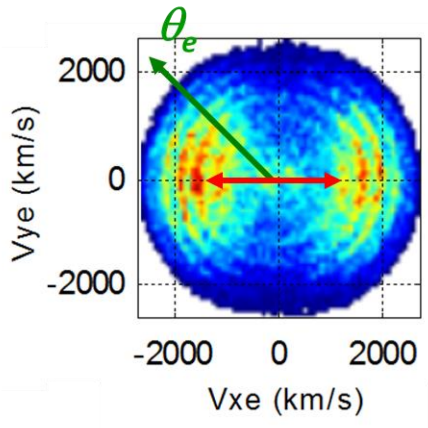
Ar and Ne

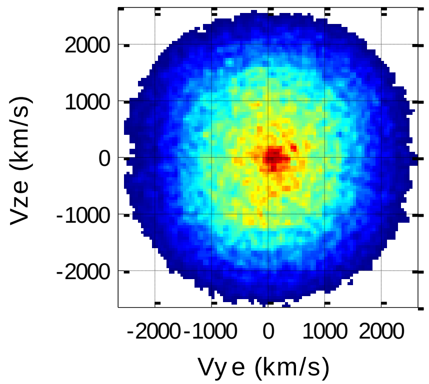
Angle resolved RABBITT scheme: Photoemission time delays & XUV-IR photoionization

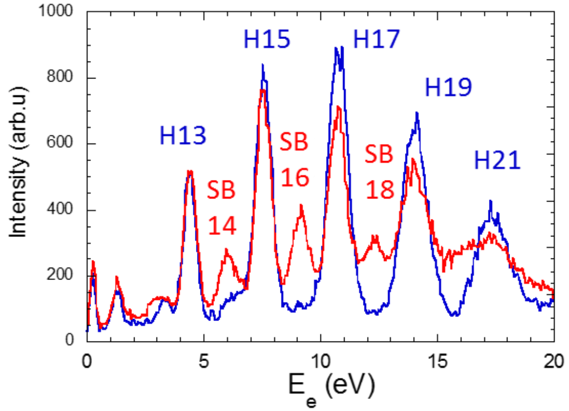
XUV-IR photoionization of Ar(3p)

$Ar\,(3p^6)$ + APT (+IR, τ) → $Ar^+(3p^5) + e^-$

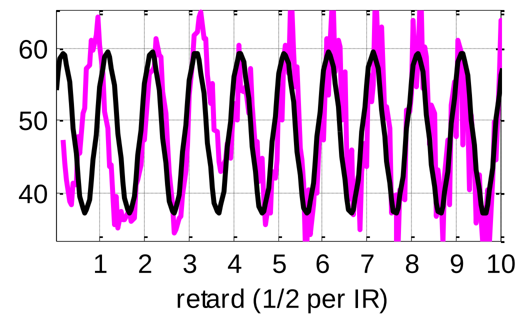
$e\,(v_x, v_y, v_z)$







SB 16



$$I_{SB}(\theta,\tau) = A(\theta) + B(\theta)\cos\left(2\omega\tau + C(\theta)\right)$$

$$I_{SB}(\theta,\tau) = h_0(\tau) + h_2(\tau)P_2(cos\theta) + h_4(\tau)P_4(cos\theta)$$

$$\beta_2 = \frac{a_2}{a_0} \qquad \beta_4 = \frac{a_4}{a_0}$$

$$h_0(\tau) = a_0 + b_0\cos\left(2\omega\tau + \phi_0\right) = I_{SB}(\tau)$$

$$h_2(\tau) = a_2 + b_2\cos\left(2\omega\tau + \phi_2\right)$$

$$h_4(\tau) = a_4 + b_4\cos\left(2\omega\tau + \phi_4\right)$$

- ❑ Set of nine coefficients

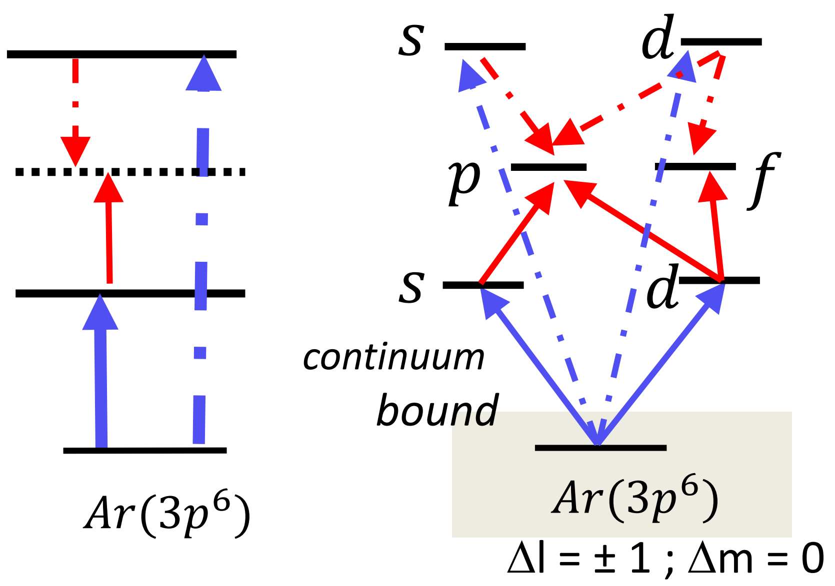
$$(a_i, b_i, \phi_i)$$

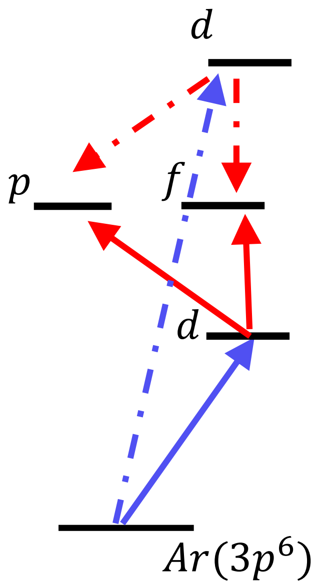
$$\gamma_2 = \frac{b_2}{b_0} \qquad \gamma_4 = \frac{b_4}{b_0}$$

$$\phi_{20} = \phi_2 - \phi_0 \qquad \phi_{40} = \phi_4 - \phi_0$$

$I_{SB}(\theta,\tau)$ PAD: formal expansion in state-to-state transition matrix elements

Ar $(3p^6)$ + APT (+IR) $\rightarrow$ $Ar^+(3p^5) + e^-$





pdf - σ^-_{pdf} δ^-_{pdf}

pdp- σ^-_{pdp} δ^-_{pdp}

pdf+ σ^+_{pdf} δ^+_{pdf}

pdp+ σ^+_{pdp} δ^+_{pdp}

Table 1. The m-independent radial parts $(l_i l' l_f)$ of the matrix elements.

Transition	(psp)	(pdp)	(pdf)
11 + IR	$218.61 - i\pi 12$	$98.85 - i\pi 131.71$	$-105.92 + i\pi 300.93$
13 + IR	$184.21 - i\pi 23.20$	$228.06 - i\pi 100.71$	$-109.65 + i\pi 204.17$
15 + IR	$158.12 - i\pi 28.15$	$239.46 - i\pi 64.57$	$-69.98 + i\pi 145.4$
17 + IR	$137.63 - i\pi 29.74$	$216.63 - i\pi 37.66$	$-39.08 + i\pi 104.49$
13 − IR	$89.16 - i\pi 36.48$	$61.55 - i\pi 178.13$	$-14.76 + i\pi 89.25$
15 − IR	$89.79 - i\pi 36.02$	$196.89 - i\pi 114.78$	$-17.47 + i\pi 82.34$
17 − IR	$86.06 - i\pi 34.67$	$208.93 - i\pi 68.41$	$-11.3 + i\pi 67.71$
19 − IR	$80.78 - i\pi 34.07$	$179.56 - i\pi 38.23$	$-4.73 + i\pi 52.10$

Table 2. The l-dependent phase shifts for final continuum states p ($l_f = 1$) and f ($l_f = 3$).

Sideband	δ_p (rad)	δ_f (rad)
12IR	−6.17	−3.25
14IR	−6.12	−2.66
16IR	−6.10	−2.30
18IR	−6.10	−2.05

ES Toma HG Muller J. Phys.B 2002

Access to the complex matrix elements in a RABBITT experiment

Example: d intermediate state

$$I_{SB}(\theta,\tau)=\sum_{m'=-1,0,1}\left|(M^{m}_{pdp})^{+}_{2q-1}e^{i\omega\tau}e^{i\delta^{+}_{pdp}}Y_{1m}+(M^{m}_{pdf})^{+}_{2q-1}e^{i\omega\tau}e^{i\delta^{+}_{pdf}}Y_{2m}+(M^{m}_{pdp})^{-}_{2q+1}e^{-i\omega\tau}e^{i\delta^{-}_{pdp}}Y_{1m}+(M^{m}_{pdf})^{-}_{2q+1}e^{-i\omega\tau}e^{i\delta^{-}_{pdf}}Y_{2m}\right|^{2}$$

m=0 contribution:

$$h_0(\tau)=\frac{16}{9}\left(\left(\sigma^{+}_{pdp}\right)^2+\left(\sigma^{-}_{pdp}\right)^2\right)+\frac{12}{7}\left(\left(\sigma^{+}_{pdf}\right)^2+\left(\sigma^{-}_{pdf}\right)^2\right)+\frac{32}{9}\sigma^{+}_{pdp}\sigma^{-}_{pdp}\cos(2\omega\tau+(\delta^{+}_{pdp}-\delta^{-}_{pdp}))+\frac{24}{7}\sigma^{+}_{pdf}\sigma^{-}_{pdf}\cos(2\omega\tau+(\delta^{+}_{pdf}-\delta^{-}_{pdf}))$$

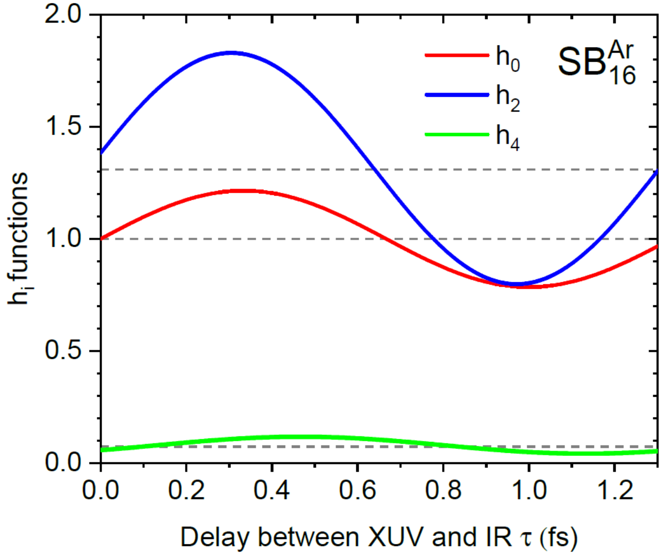
$$h_2(\tau)=\frac{16}{7}\left[\sigma^{+\,2}_{pdf}+\sigma^{-\,2}_{pdf}\right]+\frac{32}{9}\left[\sigma^{+\,2}_{pdp}+\sigma^{-\,2}_{pdp}\right]+\frac{48}{7}\left[\sigma^{+}_{pdf}\sigma^{+}_{pdp}\cos\left(\delta^{+}_{pdf}-\delta^{+}_{pdp}\right)+\sigma^{-}_{pdf}\sigma^{-}_{pdp}\cos\left(\delta^{-}_{pdf}-\delta^{-}_{pdp}\right)\right]$$
$$+\frac{32}{7}\sigma^{+}_{pdf}\sigma^{-}_{pdf}\cos\left(2\omega\tau+\delta^{+}_{pdf}-\delta^{-}_{pdf}\right)+\frac{64}{9}\sigma^{+}_{pdp}\sigma^{-}_{pdp}\cos\left(2\omega\tau+\delta^{+}_{pdp}-\delta^{-}_{pdp}\right)+\frac{48}{7}\left[\sigma^{+}_{pdf}\sigma^{-}_{pdp}\cos\left(2\omega\tau+\delta^{+}_{pdf}-\delta^{-}_{pdp}\right)+\sigma^{-}_{pdf}\sigma^{+}_{pdp}\cos\left(2\omega\tau-\delta^{-}_{pdf}+\delta^{+}_{pdp}\right)\right]$$

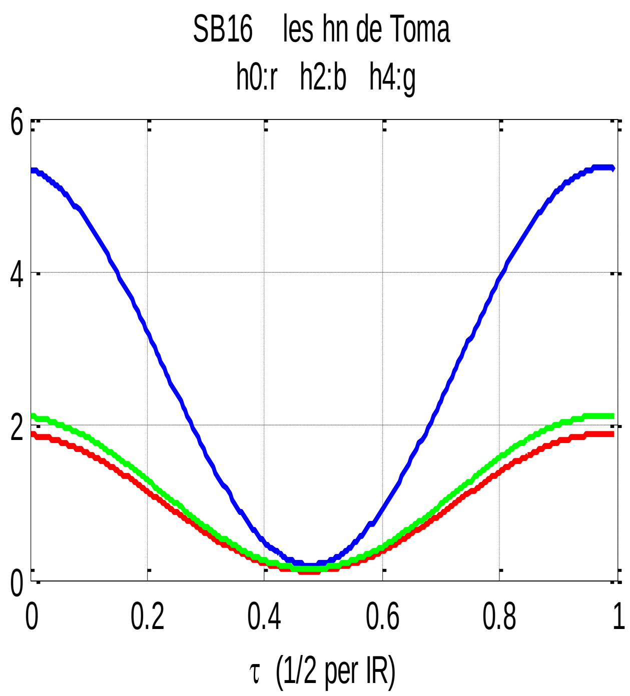
$$h_4(\tau)=\frac{216}{77}\left[\sigma^{+\,2}_{pdf}+\sigma^{-\,2}_{pdf}\right]+\frac{64}{7}\left[\sigma^{+}_{pdf}\sigma^{+}_{pdp}\cos\left(\delta^{+}_{pdf}-\delta^{+}_{pdp}\right)+\sigma^{-}_{pdf}\sigma^{-}_{pdp}\cos\left(\delta^{-}_{pdf}-\delta^{-}_{pdp}\right)\right]$$
$$+\frac{432}{77}\sigma^{+}_{pdf}\sigma^{-}_{pdf}\cos\left(2\omega\tau+\delta^{+}_{pdf}-\delta^{-}_{pdf}\right)+\frac{64}{7}\left[\sigma^{+}_{pdf}\sigma^{-}_{pdp}\cos\left(2\omega\tau+\delta^{+}_{pdf}-\delta^{-}_{pdp}\right)+\sigma^{-}_{pdf}\sigma^{+}_{pdp}\cos\left(2\omega\tau-\delta^{-}_{pdf}+\delta^{+}_{pdp}\right)\right]$$

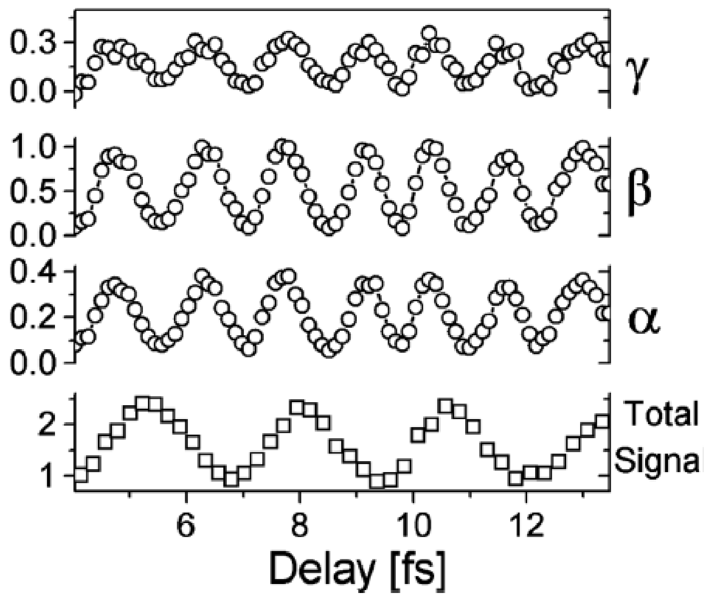
$$h_6(\tau)=\frac{400}{77}\left[\sigma^{+\,2}_{pdf}+\sigma^{-\,2}_{pdf}\right]+\frac{800}{77}\sigma^{+}_{pdf}\sigma^{-}_{pdf}\cos\left(2\omega\tau+\delta^{+}_{pdf}-\delta^{-}_{pdf}\right)$$

J. Joseph et al (in preparation)

Example of comparison with calculations or experiments







Ar(3p)

$h_0 = 1 + 0.21\cos(2\omega\tau)$

$h_2 = 1.31 + 0.52\cos(2\omega\tau + 0.134)$

$h_4 = 0.074 + 0.038\cos(2\omega\tau - 0.626)$

Ar(3p) (Toma)

$h_0 = 1 + 0.888\cos(2\omega\tau)$

$h_2 = 2.770 + 2.585\cos(2\omega\tau - 0.009)$

$h_4 = 1.131 + 0.993\cos(2\omega\tau + 0.006)$

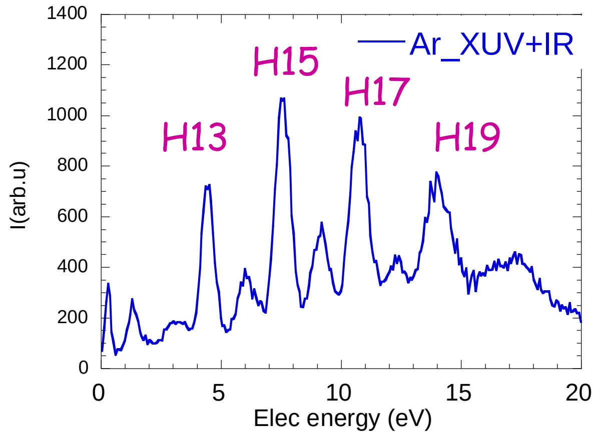
$$S(n, \theta) = \alpha + \beta P_2(\cos\theta) + \gamma P_4(\cos\theta).$$

ES Toma HG Muller J. Phys.B 2002

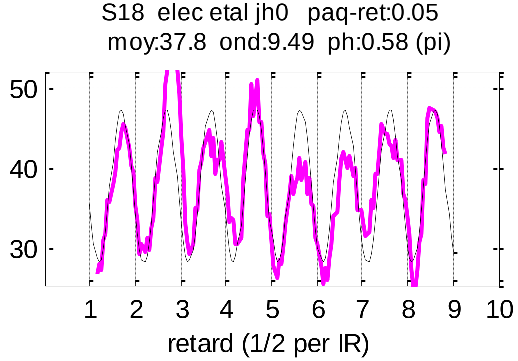
Aseyev, S. A., et al. *PRL* 91.22 (2003) 223902

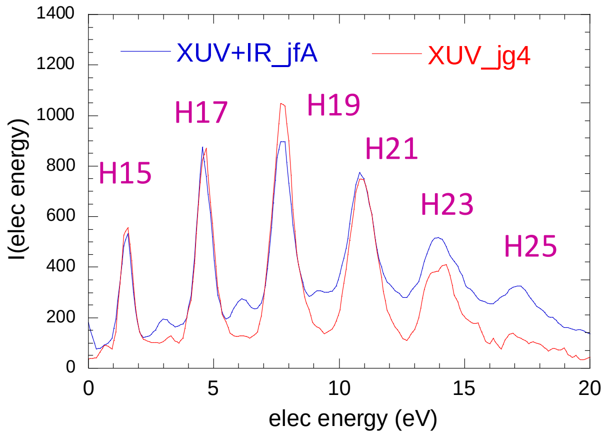
10s

Preliminary results : XUV-IR RABBITT interference: PI NO, O_2

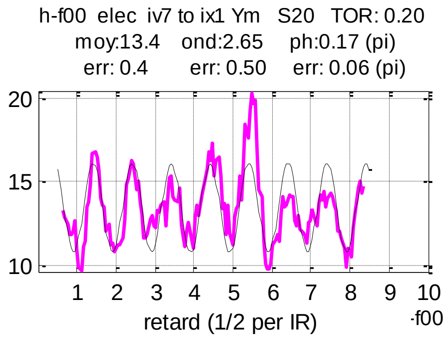


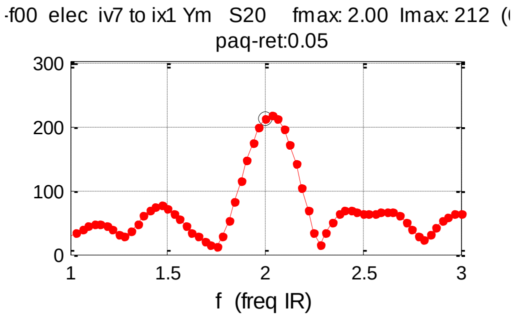
Ar(3p)

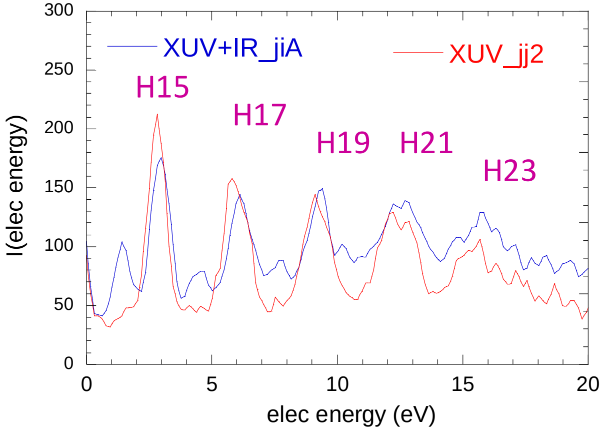




$NO^+(c^3\Pi)$







$O_2^+(B^2\Sigma_g^-)$...

$$SB_{2q}(\theta,\tau) = A(\theta) + B(\theta)cos[2\omega\tau - C(\theta)]$$

Acknowledgements


F. Holzmeier, J. Joseph, JC Houver

E. Bouisset, J. Guigand, S. Lupone, A. Marié, N. Tournier…

T. Ruchon, D. Bresteau, B. Carré et al (Attophysics group)
J F Hergott, O Tcherbakoff, F Lepetit, P D'Oliveira (SLIC)
M. Billon, I. Vadillo-Torre

C. Spezzani, J. Lenfant (OPT2X)

F. Polack, D. Dennetières (Groupe Optique)

L. Nahon, G. Garcia, JF Gil et al DESIRS beamline

J. Bozek, C. Nicolas, A Milosavljevic, E. Robert PLEIADES beamline

R.R. Lucchese (LBNL)

Merci de votre attention