4th Satellite Workshop on Photon Beam Diagnostics 28th January 2016
DESY Campus, Hamburg

Molecular polarimetry: a mean to determine the complete polarization state of the ionizing light (Stokes parameters s1, s2, s3) in the VUV-soft Xray range, based on molecular frame photoemission

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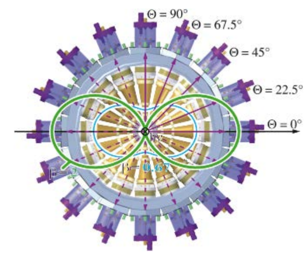
OPT2X Lidex Project Paris-Saclay University

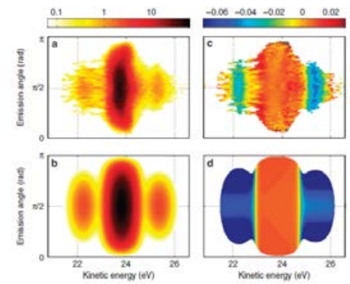
Control and characterization of polarization in the XUV and X-ray range

Need for circularly polarized, bright, coherent « chiral » light to study the chiral properties of matter : biomolecules, magnetic materials…
particular timely to produce and control ultra-short CP pulses to investigate time resolved dynamics

- Synchrotron radiation: X-ray range, VUV
  F. Schäfers et al., Appl. Opt. **38**, 4074 (1999); L. Nahon and C. Alcaraz, Appl. Opt. **43**, 1024 (2004)….

- Free Electron Lasers: FERMI FEL
  E. Allaria et al Phys. Rev. X 4, 041040 (2014)
  Mazza et al Nat. Comm. 5 3648 (2014):
  AR-PES on IR+XUV ATI of He





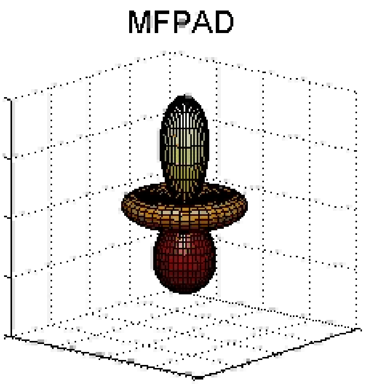
- Circularly polarized plasma-based soft X-ray laser
  A. Depresseux et al Phys. Rev. Lett. 115, 083901 (2015) /B. Vodungbo et al Opt. Expr. 19, 4346 (2011)

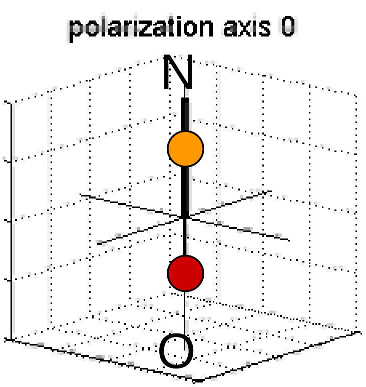
- Elliptically polarized high-order harmonics (HHG)
  O. Kfir et al Nature photonics 9, 99 (2015)
  X. Zhou et al Phys. Rev. Lett. 102, 073902 (2009)
  A. Ferré et al Nat. photon. 9, 93 (2015)
  G. Lambert et al Nat. Comm. 6 6167 (2015)

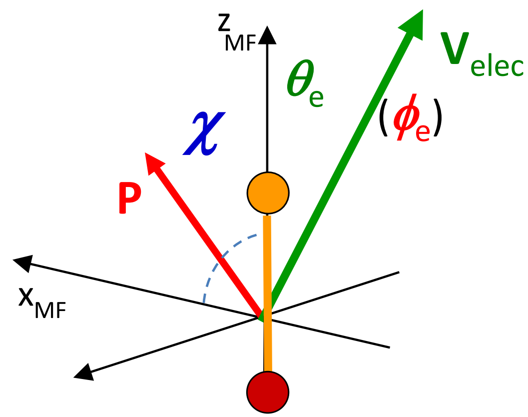
Complete characterization of the polarization state of the ionizing light based on MFPADs, including the degree of polarization

Molecular Frame Photoelectron Angular Distributions: MFPADs most complete observable in photoionization dynamics

NO ($X^2\Pi$, $\mathbf{4\sigma^2}$ $5\sigma^2$ $1\pi^4$ $2\pi^1$) + $h\nu$ $\rightarrow$ NO^+($c^3\Pi$ $\mathbf{4\sigma^{-1}}$) + e (**l,λ**)   ($h\nu \geq 22$ eV, XUV)







$$I_{lm\mu}^{M_i M_f} = \left\langle \psi_{M_i}^i \middle| d_\mu \middle| \phi_{M_f}^f \psi_{lm}^{(-)} \right\rangle$$

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

Polar and azimuthal dependence of MF photoemission for each orientation χ of the polarisation relative to the molecular axis

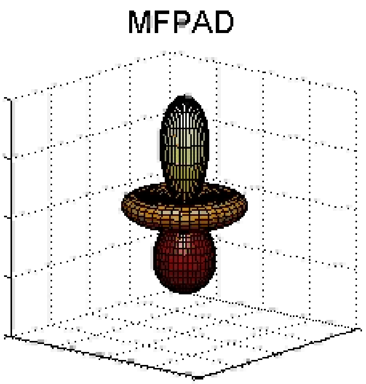
Linear polarization

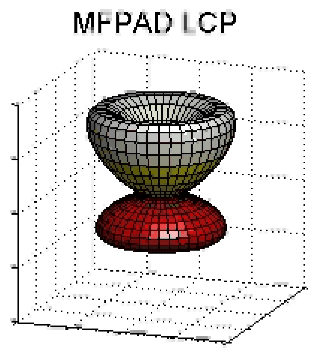
$h\nu$ = 23.15 eV

Lucchese et al PRA 65, 020702 (2002); Lafosse et al PRL 84, 5987 (2000)

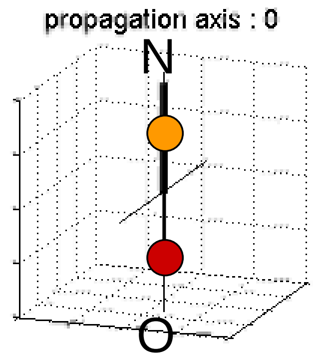
Circular dichroism in the molecular frame: MF-CDAD

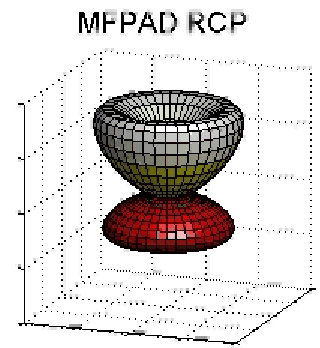
$NO\ (X^2\Pi,\ \mathbf{4\sigma^2}\ 5\sigma^2\ 1\pi^4\ 2\pi^1) + h\nu \rightarrow NO^+(c^3\Pi\ \mathbf{4\sigma^{-1}}) + e\ (\mathbf{l,\lambda})$ $(h\nu \geq 22\ \text{eV, XUV})$



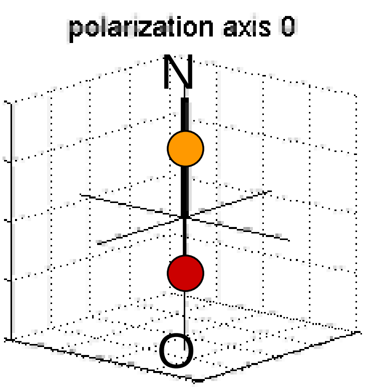


S_3=-1

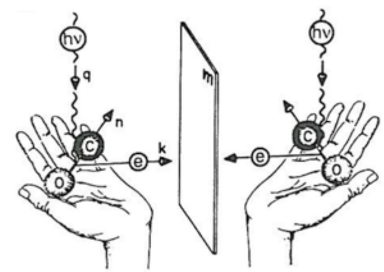




S_3=1



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



Circular polarization

Linear polarization

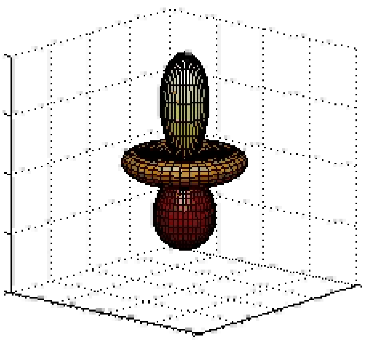
$h\nu$ = 23.15 eV

Dubbs et al, PRL, 54, 1249 (1985)
Cherepkov et al Z. Phys. D 7 721 (1987)
Schönhense Phys. Scripta 31, 255 (1989)
Jahnke et al, PRL 88, 073002(2002)
…

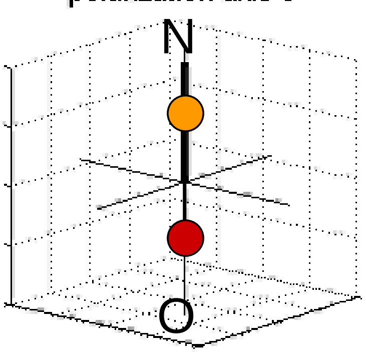
Circular dichroism in the molecular frame: MF-CDAD

NO ($X^2\Pi$, $\mathbf{4\sigma^2}$ $5\sigma^2$ $1\pi^4$ $2\pi^1$) + $h\nu$ $\rightarrow$ NO^+($c^3\Pi$ $\mathbf{4\sigma^{-1}}$) + e (**l,λ**)   ($h\nu \geq 22$ eV, XUV)

MFPAD

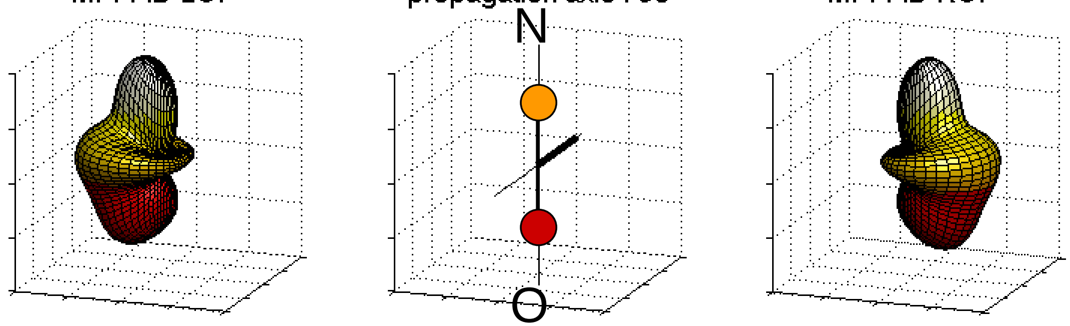


polarization axis 0



Linear polarization

$h\nu$ = 23.15 eV

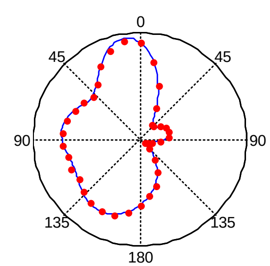


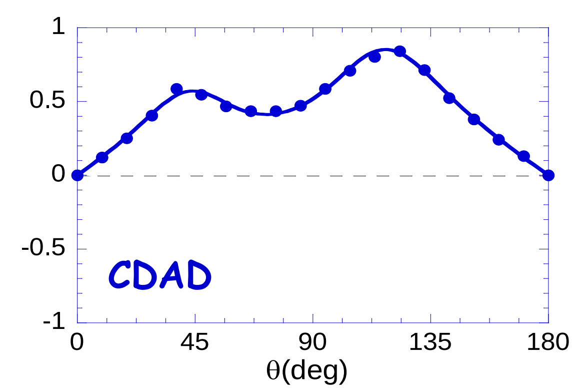
S_3=-1

S_3=1

Circular dichroism

$$CDAD(\theta_e) = \frac{I_{LCP}(\theta_e) - I_{RCP}(\theta_e)}{I_{LCP}(\theta_e) + I_{RCP}(\theta_e)}$$



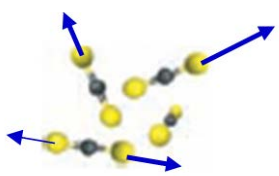


Gessner et al PRL 88 193002 (2002); Lebech et al JCP 118, 9653 (2003);

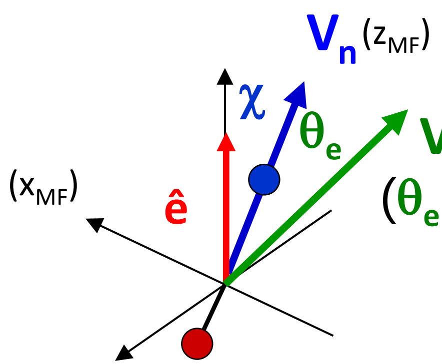
Electron-ion 3D coincidence momentum spectroscopy in Dissociative PhotoIonization of small molecules

$AB + h\nu\,(\hat{e}) \rightarrow AB^{+} + e$

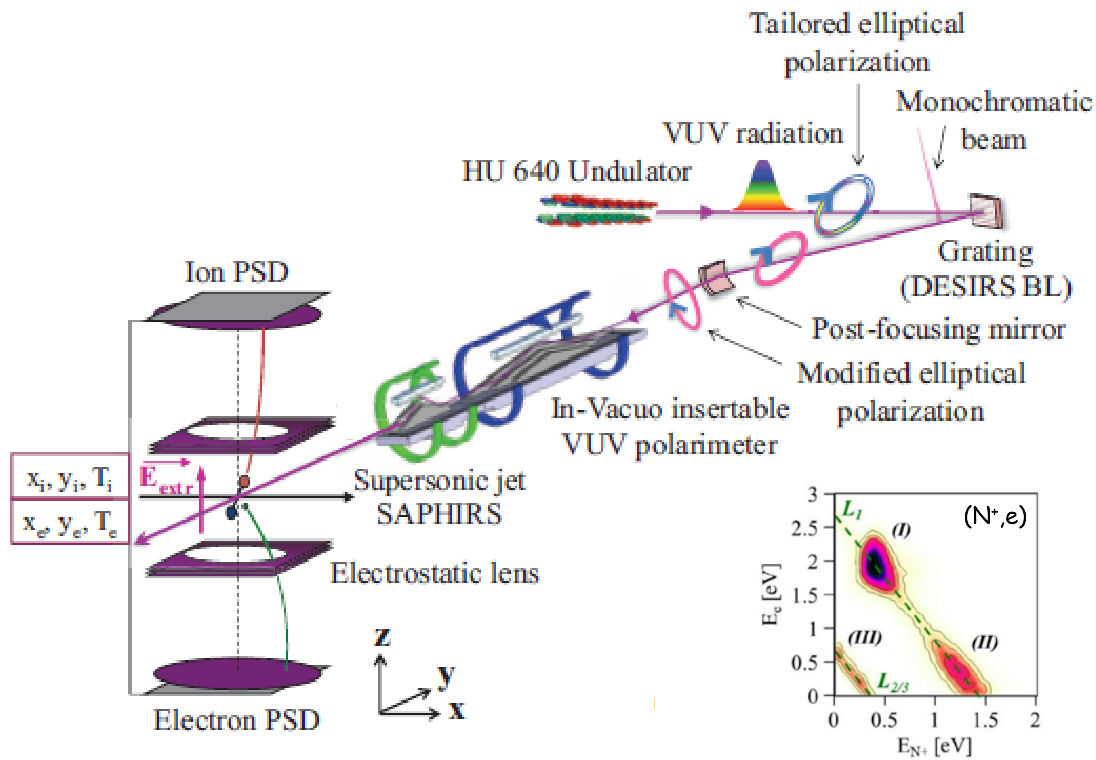
$\rightarrow A^{+} + B + e$



$(V_e, V_{A+}, \hat{e})$



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



DESIRS beam-line 5-40 eV valence & inner-valence PI

PLEIADES inner-shell PI

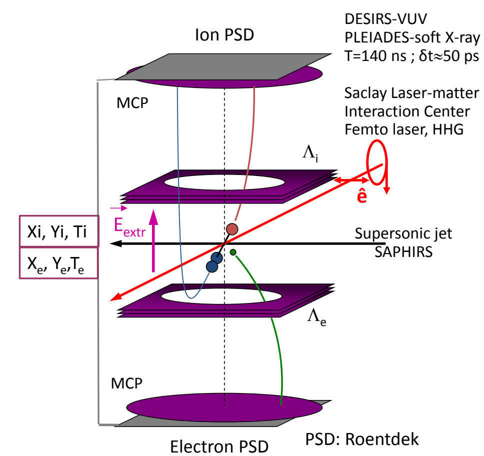
Rep-rate of the light source: SOLEIL (few bunch mode T=147 ns) ~ 7MHz
Coincidences: n events/pulse << 1

Lafosse et al PRL 84 5987 (2000) ; Lebech et al RSI 73 1866 (2002); L. Nahon et al J. Synch. Rad. 19, 508 (2012)

Electron-ion 3D coincidence momentum spectroscopy in Dissociative PhotoIonization of small molecules: soft X-rays and light sources

$$AB + h\nu\,(\hat{e}) \rightarrow AB^{++} + e_{ph} + e_{Au} \rightarrow A^{+} + B^{+} + e_{ph} + e_{Au}$$

$$(V_e, V_{A+}, V_{B+}, \hat{e})$$



K-shell ionization of CO and NO….

C(1s): 284 eV, N(1s) 410 eV, O(1s) 543 eV …

W.B. Li et al PRA 75, 052718 (2007);
Jahnke et al, PRL 88, 073002(2002)

Sources of ultrashort XUV & X-ray pulses:
Femtosecond lasers and HHG XUV sources x 1 kHz

FELs : European XFEL 27000 pulses/s

Efficient methods for the multidimensional data analysis:

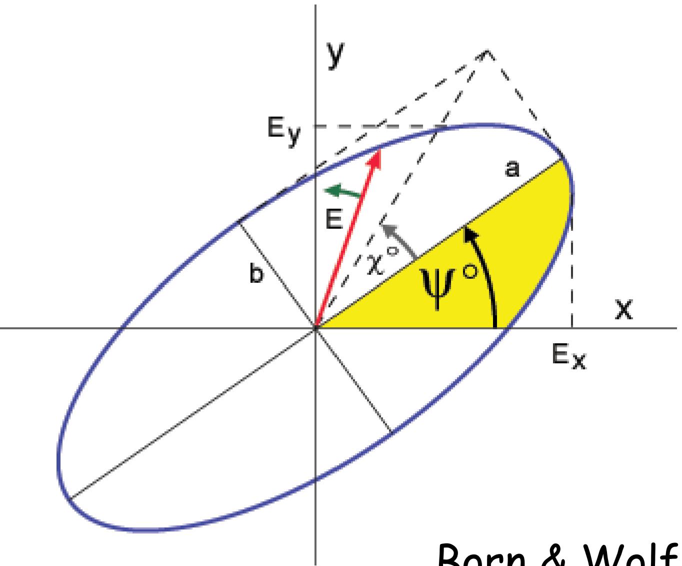
Lebech et al JCP 118, 9653 (2003)
Billaud et al J. Phys. B 45 (2012)

$$T_{fi\pm1}(\chi,\theta_e,\phi_e) = F_{00}(\theta_e) - 0.5\,F_{20}(\theta_e)\,P_{20}(\cos\chi) - 0.5\,F_{21}(\theta_e)\,P_{21}(\cos\chi)\cos(\phi_e)$$
$$- 0.5\,F_{22}(\theta_e)\,P_{22}(\cos\chi)\cos(2\phi_e)$$
$$\pm\,F_{11}(\theta_e)\,P_{11}(\cos\chi)\sin(\phi_e)$$

$$F_N^L(\theta_e) = \sum_{L'} C_{L'LN}\,P_{L'}^N(\cos\theta_e)$$

STOKES PARAMETERS $\hat{e} = (s_1, s_2, s_3)$ and Polarization Ellipse

Polarisation ellipse



Born & Wolf

Linear component: s_1, s_2 orientation Ψ

$$\mathrm{tg}\, 2\Psi = \frac{s_2}{s_1}$$

Ellipticity: $\varepsilon = \mathrm{tg}\, \chi$

$$\sin 2\chi = \frac{s_3}{\sqrt{s_1^2 + s_2^2 + s_3^2}}$$

Degree of polarization:

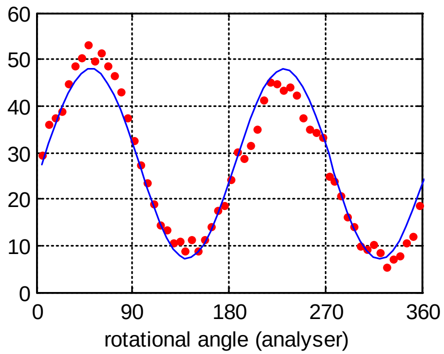
$$P = \sqrt{s_1^2 + s_2^2 + s_3^2} = 1 - s_4$$

Optical polarimetry without dephaser:
Malus law

$$\Psi = \frac{1}{2}\mathrm{Arctg}\frac{s_2}{s_1} \qquad s = \sqrt{s_1^2 + s_2^2}$$

$$s_{3ub} = \sqrt{1 - s^2}$$

Assuming P = 1: $\varepsilon_{ub} = \mathrm{tg}\ 0.5\ \mathrm{Arcsin}(s_{3ub})$



MFPADs INDUCED BY ELLIPTICALLY POLARIZED LIGHT

$\hat{e} = (s_1, s_2, s_3)$

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

$$F_{00}(\theta_e) + F_{20}(\theta_e)\{-0.5\, P_{20}(\cos\chi) + 0.25\, t_1(\gamma)\, P_{22}(\cos\chi)\}$$

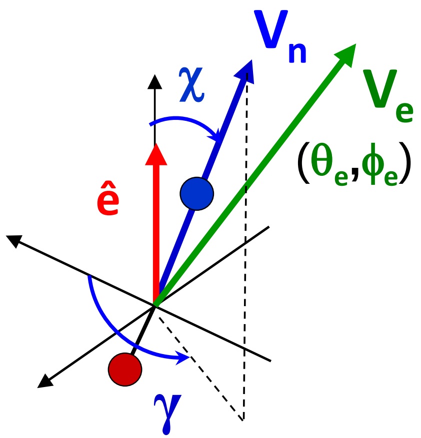
$$+ F_{21}(\theta_e)\{[-0.5\, P_{21}(\cos\chi) - 0.5\, t_1(\gamma)\, P_{21}(\cos\chi)]\cos(\phi_e) - 1.5\, t_2(\gamma)\, P_{11}(\cos\chi)\sin(\phi_e)\}$$

$$+ F_{22}(\theta_e)\{[-0.5\, P_{22}(\cos\chi) + t_1(\gamma)\,(2+P_{20}(\cos\chi))]\cos(2\phi_e) + 3\, t_2(\gamma)\, P_{10}(\cos\chi)\sin(2\phi_e)\}$$

$$-\ s_3 F_{11}(\theta_e)\{P_{11}(\cos\chi)\sin(\phi_e)\}$$

with $t_1(\gamma) = -s_1\cos(2\gamma) + s_2\sin(2\gamma)$

$t_2(\gamma) = -s_2\cos(2\gamma) - s_1\sin(2\gamma)$



Extraction of the 5 $F_{LN}(\theta_e)$ functions: PI dynamics

Extraction of s_3: Molecular frame circular dichroism

Extraction of s_1 and s_2:
Lab frame ion fragment (or photoelectron) (χ, γ) angular distribution

Veyrinas et al., PRA 88, 063411 (2013)

Determination of the STOKES PARAMETERS (s_1,s_2,s_3): s_3

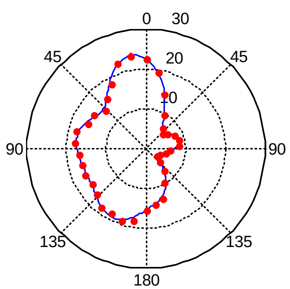
NO ($X^2\Pi$, $\mathbf{4\sigma^2}$ $5\sigma^2$ $1\pi^4$ $2\pi^1$) + $h\nu$ $\rightarrow$ $NO^+(c^3\Pi$ $\mathbf{4\sigma^{-1}})$ + e ($\mathbf{l},\lambda$)

Dominant DPI reaction,

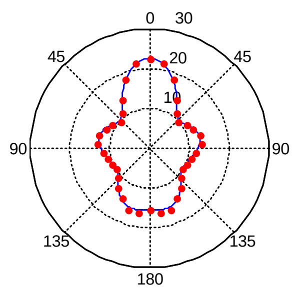
Large β_{ion} anisotropy: $\beta_{ion} \approx 1$

Large circular dichroism (F_{11})

Circular dichroism in the molecular frame $\mathbf{s_3}$ $F_{11}(\theta_e)$

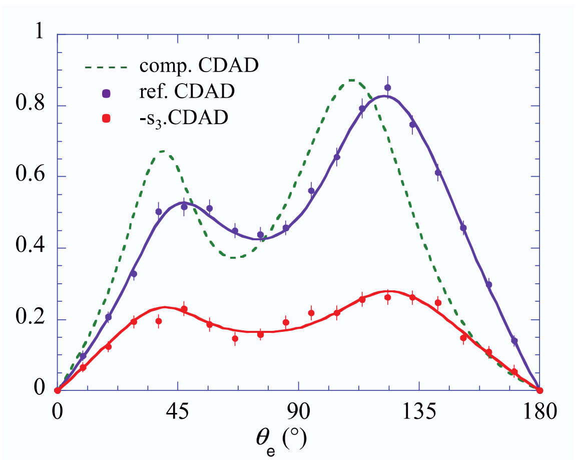


S_3=-1



S_3=0

$CDAD_{meas} = -\ \mathbf{s_3}\ CDAD_{ref}$



S_3 = -0.36 (0.02)

Establish reference values of the CDAD (F_{11})

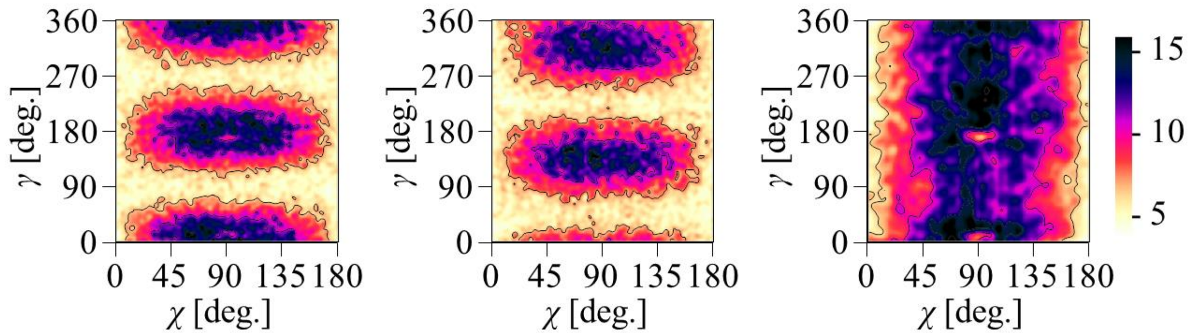
Veyrinas et al., PRA 88, 063411 (2013): MCSCI calculations R.R. Lucchese

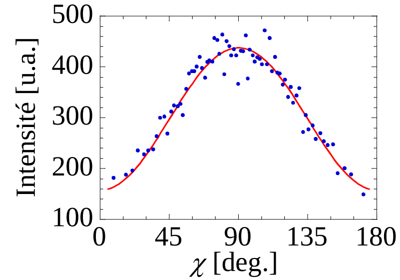
Determination of the STOKES PARAMETERS (s_1,s_2,s_3): s_1,s_2

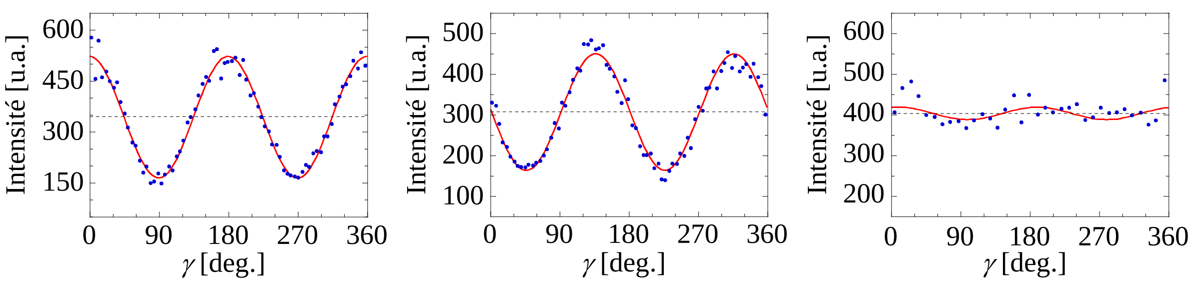
Photoion angular distribution in the lab frame 2D histogram (χ,γ): s_1 and s_2

$$T(\chi,\gamma)=C\left(1+\beta_i\left[-\frac{1}{2}P_2^0(\cos\chi)+\frac{1}{4}\left(s_1\cos(2\gamma)-s_2\sin(2\gamma)\right)P_2^2(\cos\chi)\right]\right)$$

Integration (θ_e,ϕ_e)



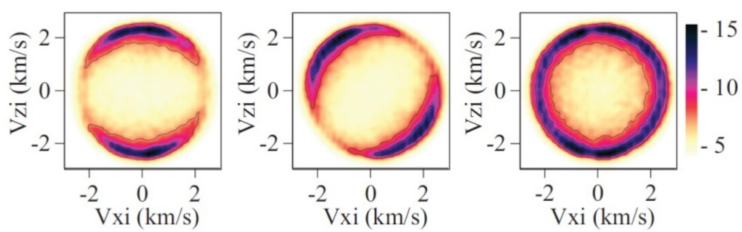




$$T(\gamma)=C\left(1+\frac{\beta_{N+}}{2}(s_1\cos 2\gamma-s_2\sin 2\gamma)\right)$$

s_1 = **-0.96**, s_2 = -0.03 | s_1 = -0.01, s_2 = **-0.86** | s_1 = -0,07 s_2 = 0.02

$\Delta s \sim \pm\ 0.01$



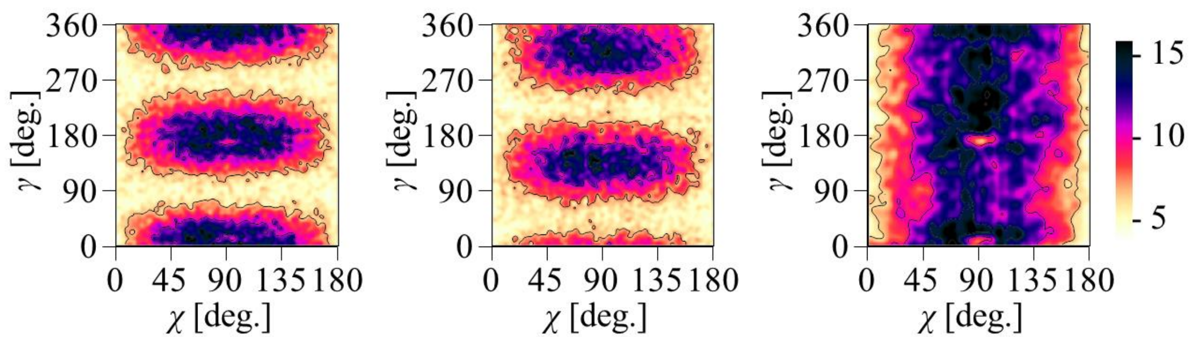
Veyrinas et al., PRA 88, 063411 (2013)

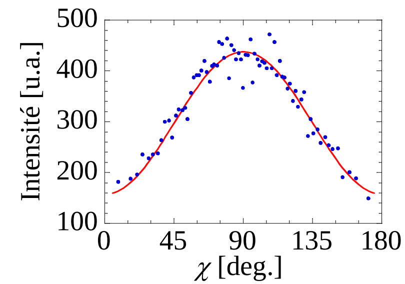
DETERMINATION of the 3 STOKES PARAMETERS

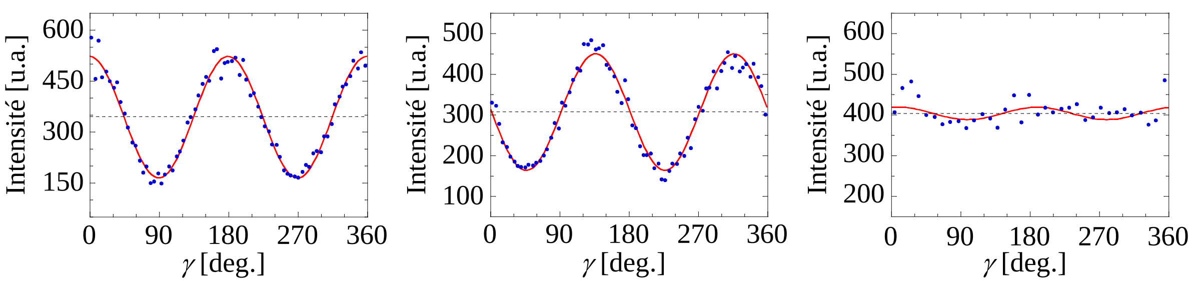
Photoion angular distribution in the lab frame 2D histogram (χ,γ): s_1 and s_2

$$T(\chi,\gamma) = C\left(1+\beta_i\left[-\frac{1}{2}P_2^0(\cos\chi)+\frac{1}{4}\left(s_1\cos(2\gamma)-s_2\sin(2\gamma)\right)P_2^2(\cos\chi)\right]\right)$$

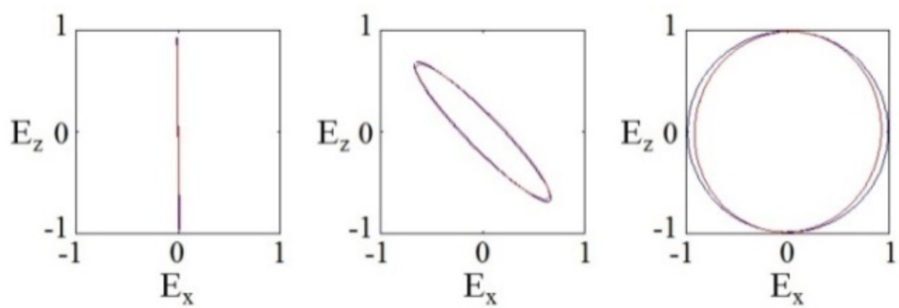
Integration
(θ_e,ϕ_e)





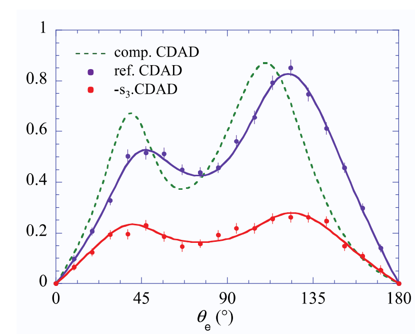


$$T(\gamma) = C\left(1+\frac{\beta_{N+}}{2}\left(s_1\cos 2\gamma - s_2\sin 2\gamma\right)\right)$$



OP & MP
(s_1,s_2,s_3)

$P = (1\text{-}s_4)$

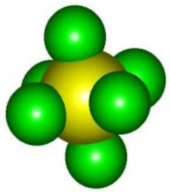


Veyrinas et al., PRA 88, 063411 (2013)

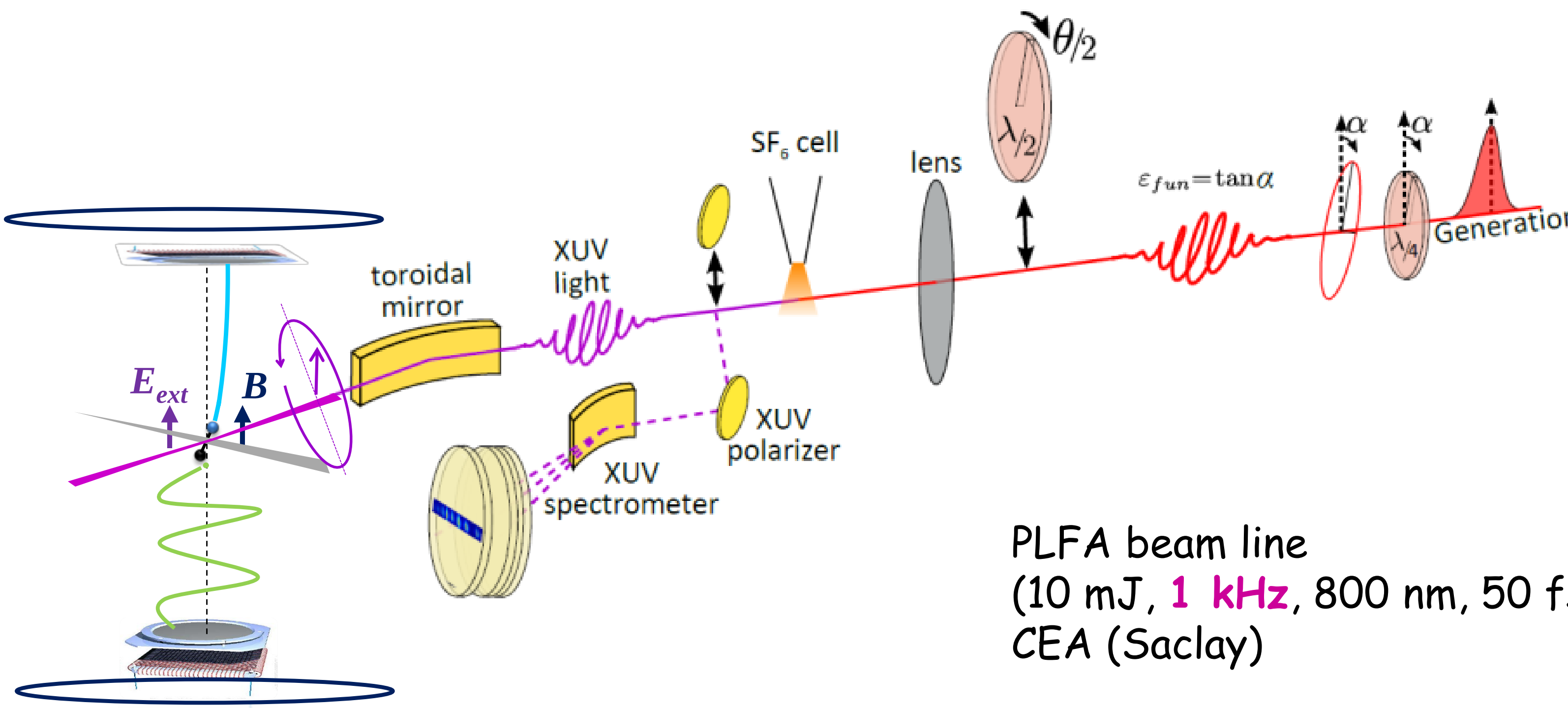
Polarization state of HHG XUV Attosecond Pulse Train

HHG in SF_6 molecules induced by elliptically polarized IR (moderate)

A. Ferré et al Nat. Phot. 9 (2014)



Production of large « ellipticities » $\varepsilon_{ub} = 0.6\text{-}0.8$



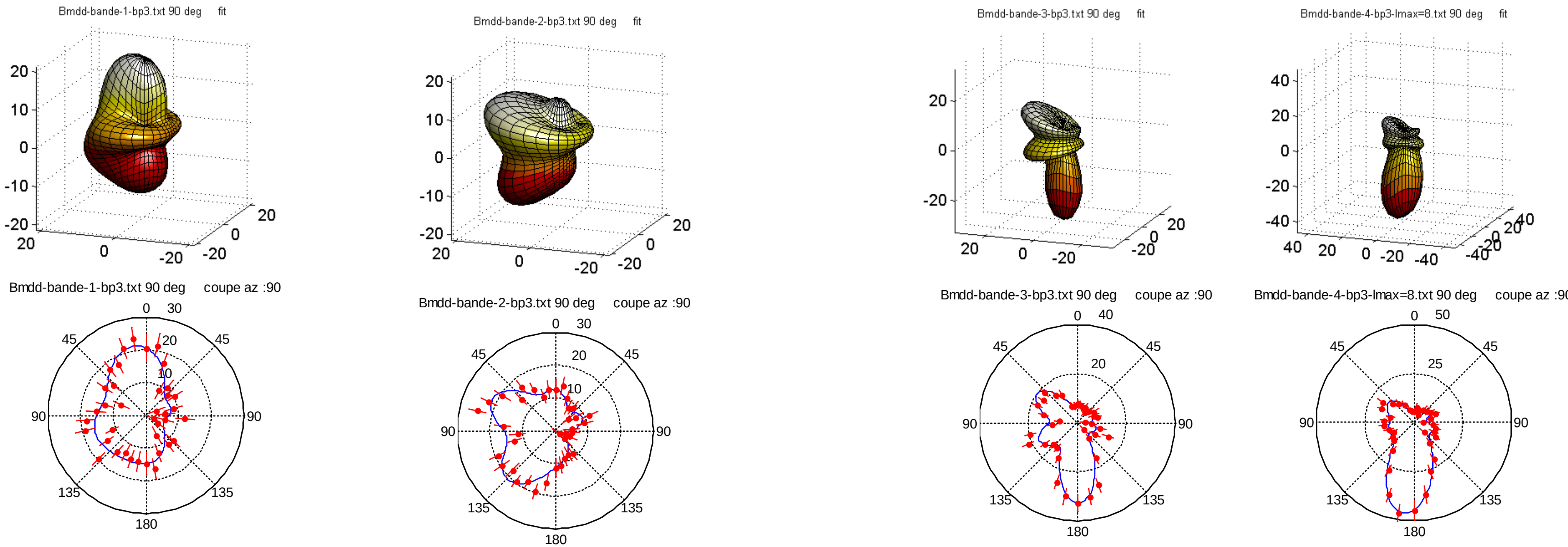
PLFA beam line
(10 mJ, **1 kHz**, 800 nm, 50 fs)
CEA (Saclay)

S.J. Weber et al RSI 86, 033108 (2015)

CIEL COLTRIMS

Gisselbrecht et al., RSI 76, 013105 (2005)
Lebech et al., RSI 73, 1866 (2002)

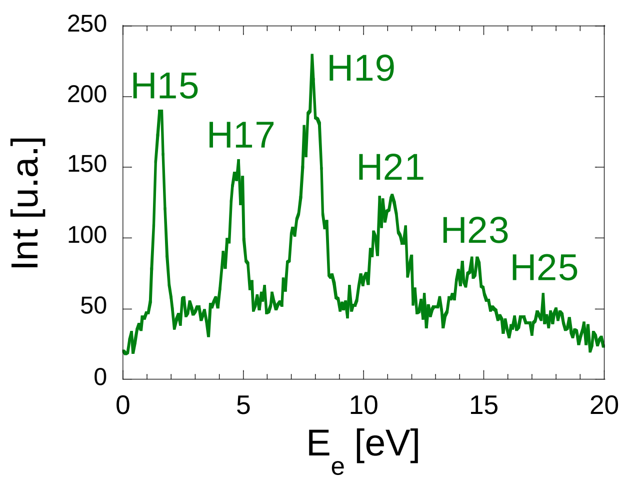
Results: HHG from SF_6 molecules ε (IR) = 0.2



$\varepsilon \approx 0.84(0.07)$ $\varepsilon \approx 0.65(0.05)$ $\varepsilon \approx 0.26(0,04)$ $\varepsilon \approx 0.11\ (0,04)$

Reference
DESIRS SOLEIL $S_3 \sim 1$

Veyrinas, Gruson *et al* in preparation (2016)



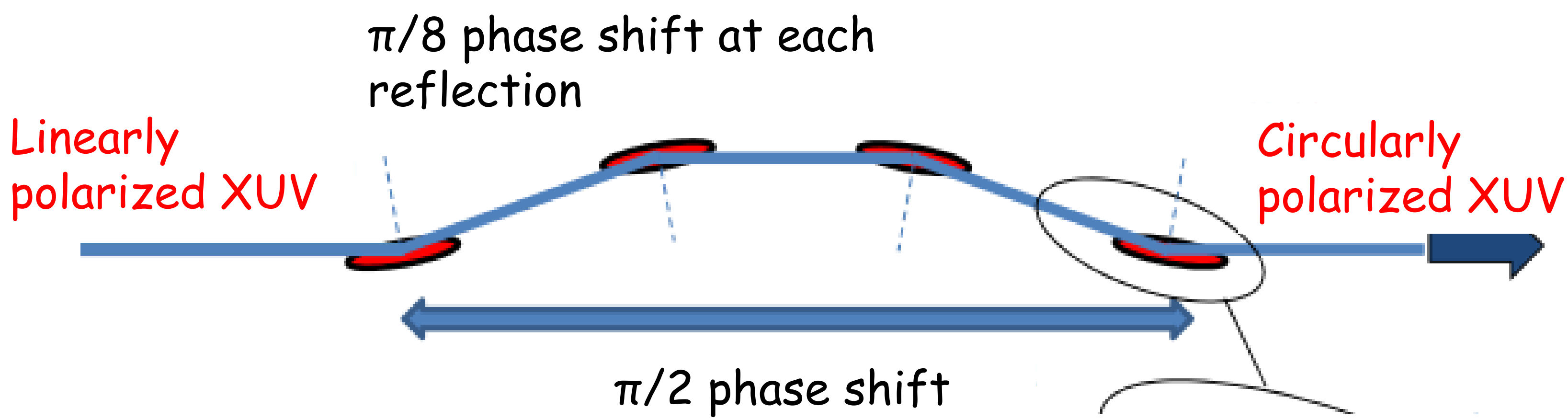
OPT2X project: 7 labs
Université Paris-Saclay
XUV Beamline instruments

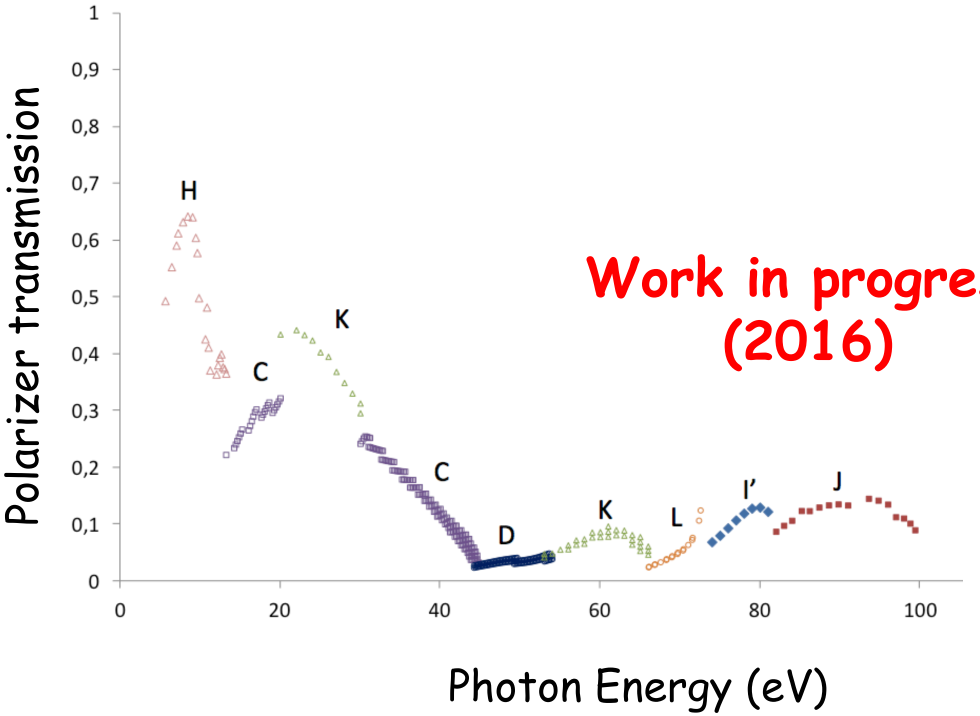
ATTOLAB: FAB 10 (10kHz)

Multilayer optical polarimeters

Task: « polarization diagnostic for probing HHG mechanisms ? »

OPT2X: Polarization control Development of a versatile ML based optical polarizer for ultrashort XUV pulses





Work in progress (2016)

ML → photon energies 8-100eV
Reflectivity: 3 - 64%
S3 99,7% for monochromatic XUV

M. Dehlinger, F. Delmotte (LCF)
Optics characterization: SOLEIL
L.Nahon WG1 OPT2X

Conclusion and perspectives

Molecular frame photoemission:

A sensitive "in situ" probe of the complete polarization state of XUV and X-ray radiation in one-XUV or X-ray photon ionization:

Full polarization ellipse (s_1, s_2, s_3), including the degree of polarization
disentanglement of s_3 and s_4
Need to establish reference value for the CDAD (exp and/or theory)

Extended photon energy range, tunability with the phototelectron energy
Inner-valence and K /L shell ionization CO, NO, CS2….
C(1s), O(1s), N(1s), S(2p), F(1s)…

Electron-ion coincident momentum spectroscopy
Take advantage of High rep rate of XFEL and the existence of COLTRIMS instruments

Characterization of high harmonics elliptically polarized XUV attosecond pulses when symmetry breaking occurs:

- HHG from aligned molecules: origin of depolarization ?
- Role of shape resonance n PI of SF6: amplification of ellipticity
- Counter rotating bichromatic fields (O. Cohen et al)

Integration in beamlines…

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L. Nahon, G. Garcia et al DESIRS beamline (SOLEIL)

J. Bozek, C. Miron, C. Nicolas, M. Patanen PLEIADES beamline (SOLEIL)

V. Gruson, S. Weber, L. Barreau, A. Camper, T. Ruchon, B. Carré, P. Salières et al
J.F. Hergott, F. Lepetit et al
Attophysics and SLICS (LIDyL, CEA, Saclay)

M. Simon, L. Journel, R. Guillemin (LCPMR, UPMC)

F. Martín et al (UAM Madrid)

ATTOLAB, OPT2X

Thank you for your attention