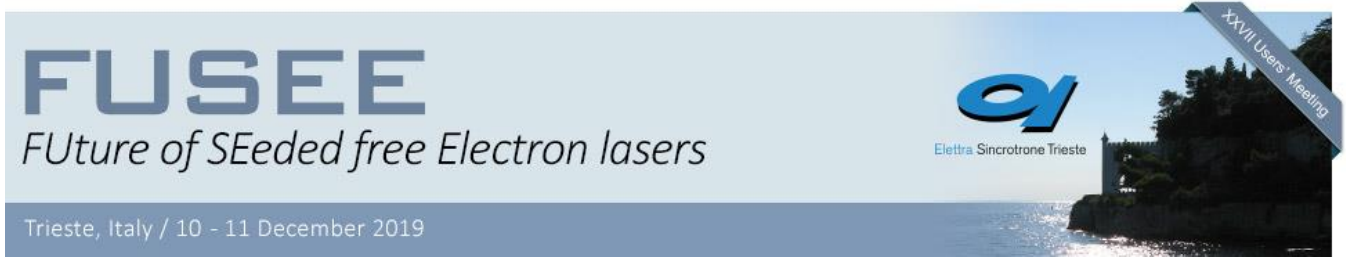

Complete reconstruction of bound and unbound electronic wavefunctions in two-photon double ionization

P. A. Carpeggiani [1,2,11], E. V. Gryzlova [3], M. Reduzzi[1,2], A. Dubrouil[1], D. Faccialá [1,2], M. Negro [1,2], K. Ueda [4], S. M. Burkov [5], F. Frassetto [6], F. Stienkemeier [7], Y. Ovcharenko [8,9], M. Meyer [9], O. Plekan [10], P. Finetti [10], K. C. Prince [10], C. Callegari [10], A. N. Grum-Grzhimailo [3] and G. Sansone[7].

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ARTICLES
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Outline

1. Intro: complete experiment & two photon double ionization

2. FERMI FEL and Low Density Matter beamline

3. Neon, non resonant case:
   - excitation scheme & detection
   - yield ratio and PAD (photoelectron angular distribution)
   - complete experiment: theory vs experiment

4. Resonant case: the role of AIS (auto ionizing states)

5. Complete experiment: reconstruction of all the variables

6. Conclusions

Motivations

Complete experiment in photoionization

- ✓ *Complete characterization of the process*
- ✓ *Information on all observables (test of theory)*
- ✓ *Three dimensional photoelectron distribution (Amplitudes A_n and phases φ_n of the partial waves)*

Two-photon double ionization

- ✓ *Dominant nonlinear mechanism intensity range 10^{13}-10^{15} W/cm^2*
- ✓ *High XUV intensities are required*
- ✓ *Complete experiment in ions (intermediate polarized state)*

H. Klar and H. Kleinpoppen J. Phys. B: At. Mol. Opt. Phys. **15**, 933 (1982)

FERMI @ ELETTRA
& LDM (Low Density Matter) Beamline

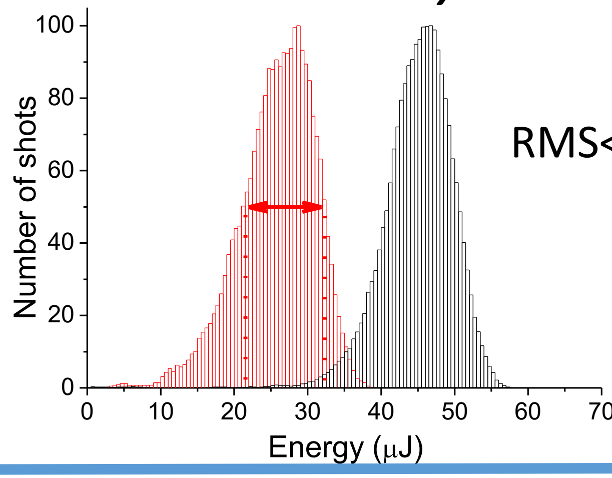
FERMI and ELETTRA

Seeded FEL XUV pulses:

- Polarization control
- Spectral and intensity stability
- Spectral tunability

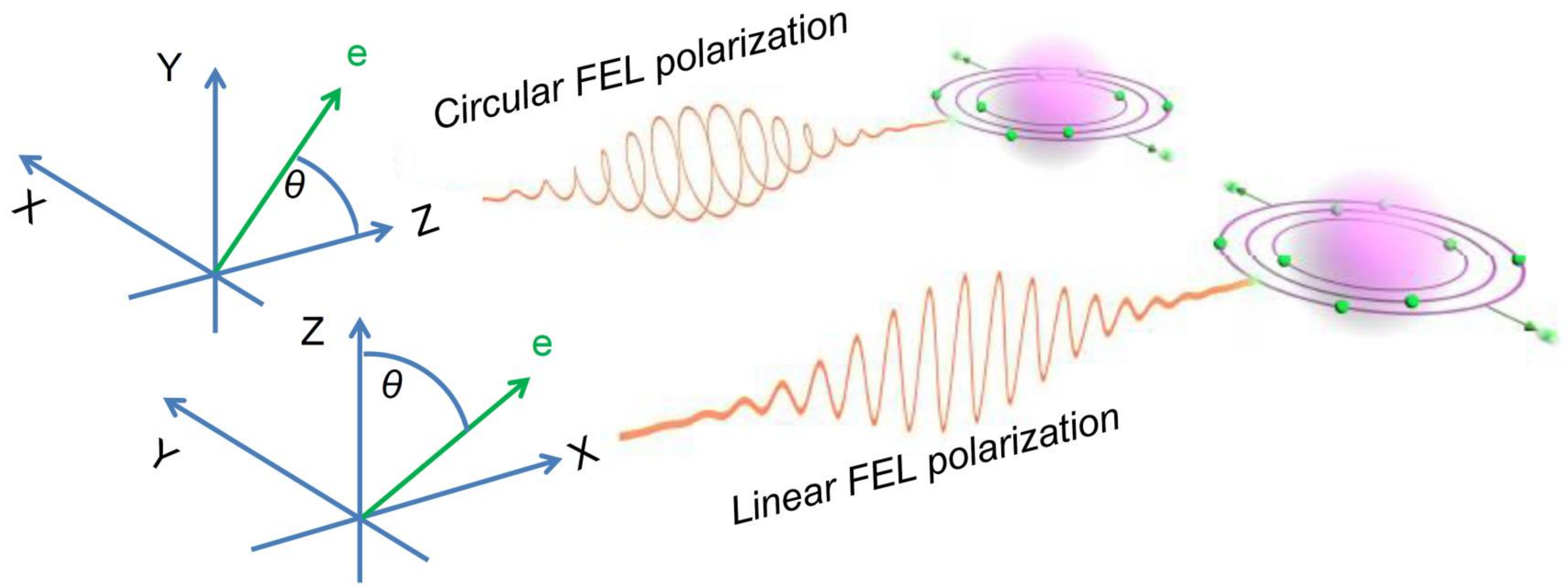
- Pulse energy ~10µJ
- $\Delta\omega$ = 1.5% FWHM (90meV @ 60eV)

Pulse energy stability

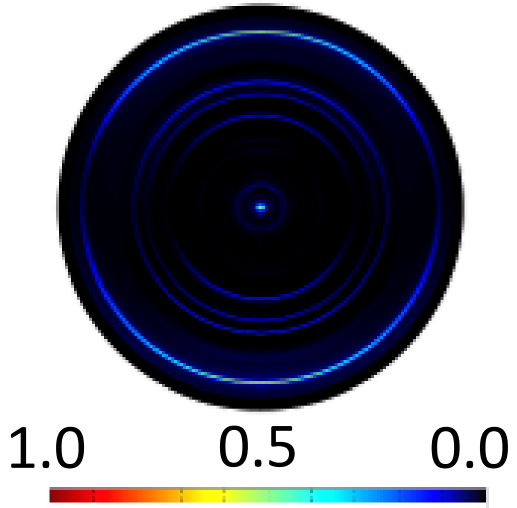


RMS<10%

The experiment:
linearly and circularly polarized intense XUV pulses



Detection:
Velocity Map Imaging (VMI)

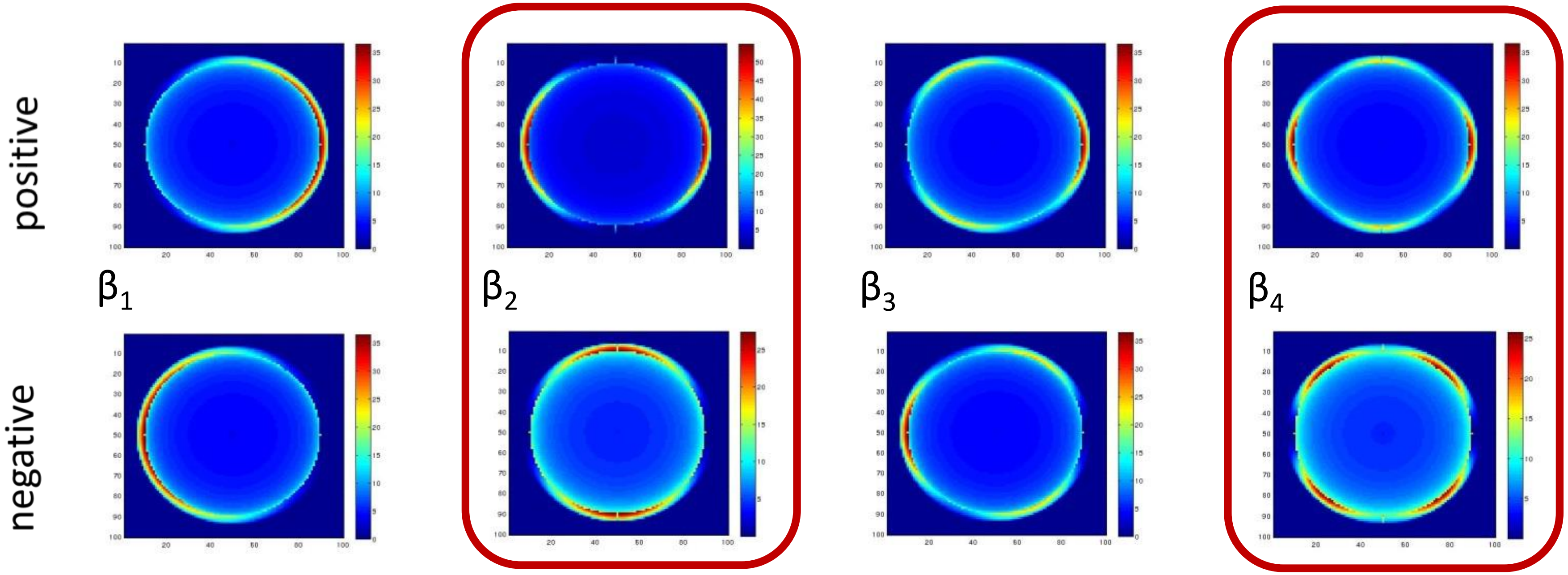


1) *Complete experiment*: 44-62 eV in steps of 0.5 eV

2) *Role of autoionizing states*: 56.2-56.7 eV in steps of 20 meV

β_k parameters:
definition and physical meaning

$$\frac{dW}{d\Omega} = \frac{W_0}{4\pi}\left[1 + \sum_{k=1}^{K} \beta_k P_k(\cos\theta)\right]$$



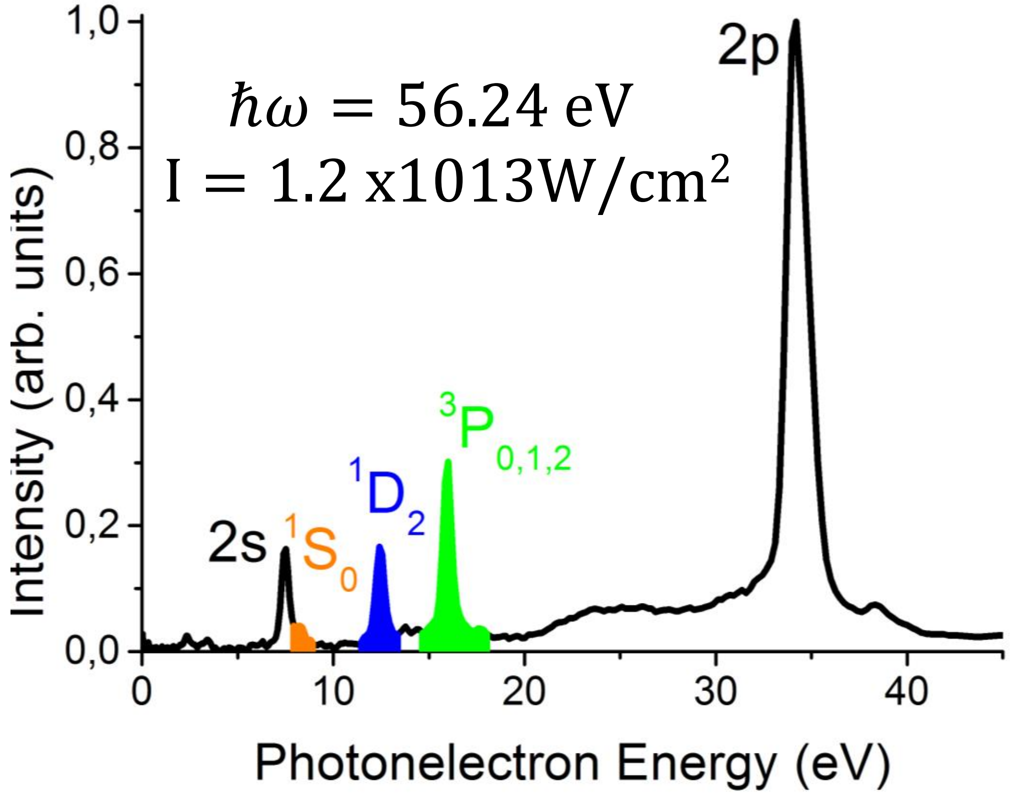
Calculated VMI images for PAD with a $\pm\beta_{k=1\ldots4}$ behaviour

Ne, 1 color, 1 photon ionization: $1+ \beta_2 P_2$

Ne, 1 color, 2 photon double sequential ionization: $1+ \beta_2 P_2 + \beta_4 P_4$

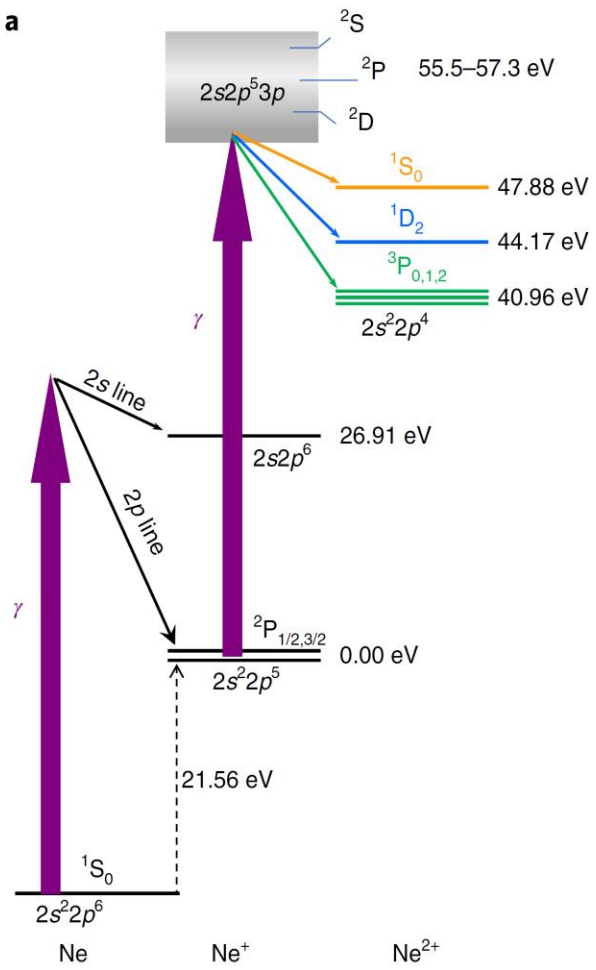
(pulse duration >> optical cycle)

Photoelectron spectrum & ionization channels



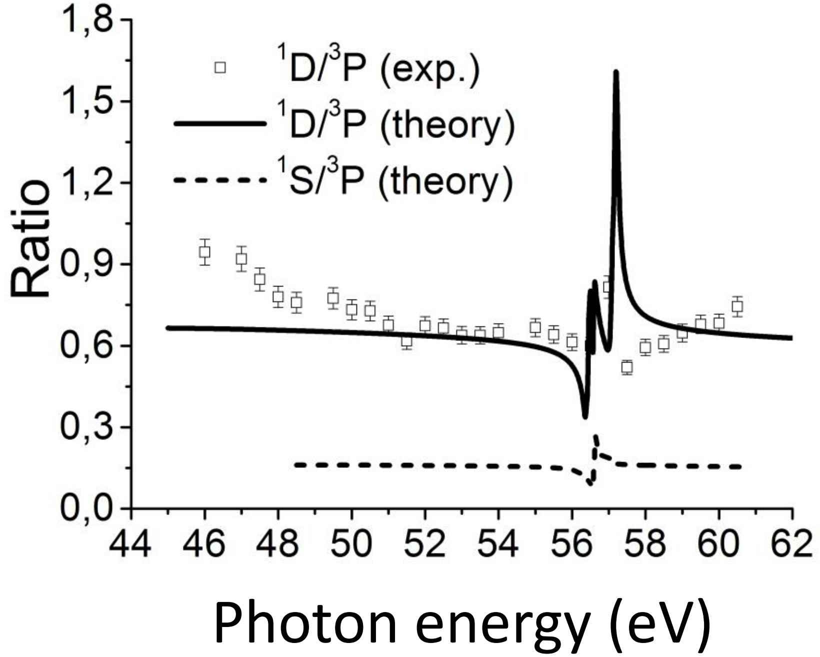
$$I^{\upsilon}(\theta) = (I_0^{\upsilon}/4\pi)[1 + B_2^{\upsilon} P_2(\cos\theta) + B_4^{\upsilon} P_4(\cos\theta)]$$

$$\upsilon = L\ (linear)\ B_2^{\nu} = \beta$$
$$\upsilon = C\ (circular)\ B_2^{\nu} = -\beta/2$$

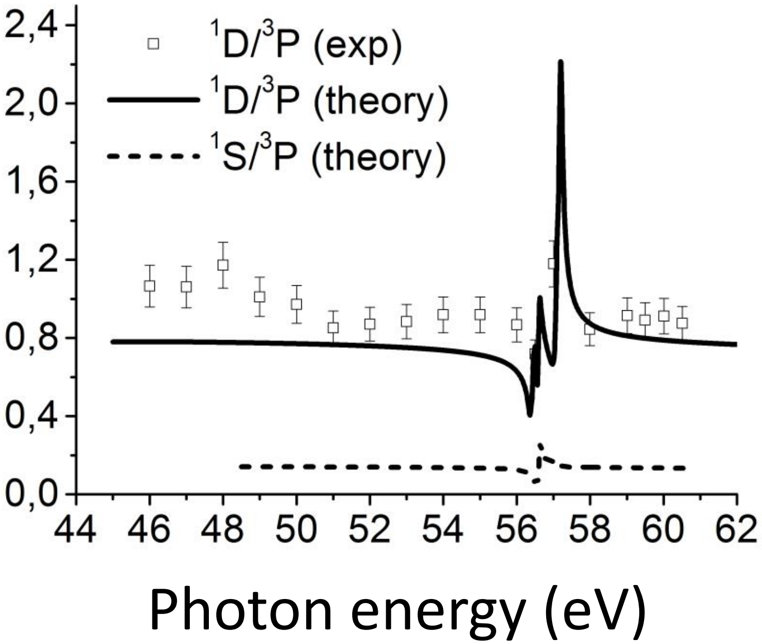


PES (Photo Electron Spectra):
Peak Ratios

Linear Polarization



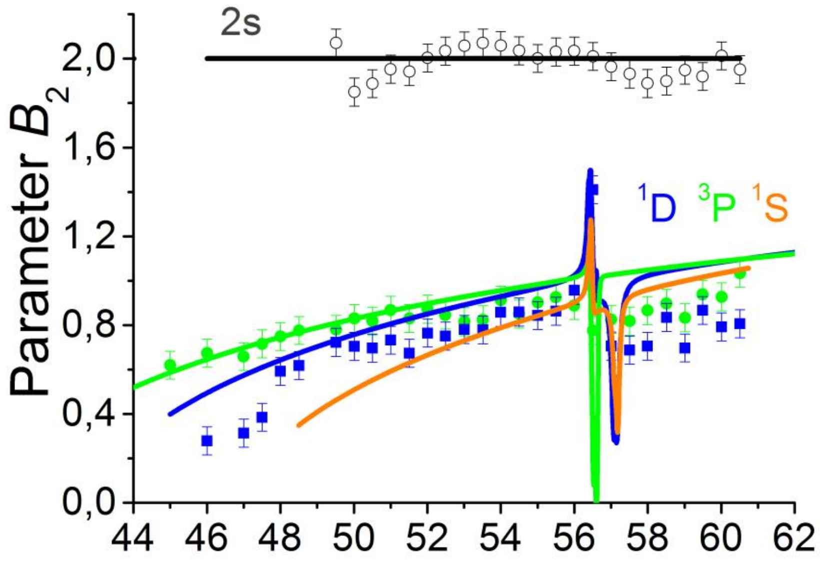
Circular Polarization



- ✓ *Good agreement between experimental and theoretical ratios*
- ✓ *Presence of sharp resonances around 56 eV (autoionizing states)*
- *Overlap between 2s and ^1S photoelectrons*

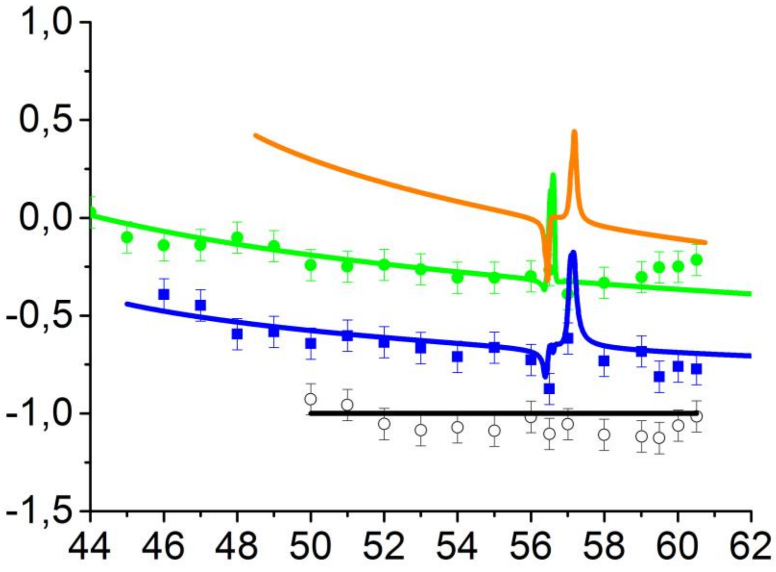
PES (Photo Electron Spectra): Angular Distribution (β_2 parameter)

Linear Polarization



Photon energy (eV)

Circular Polarization

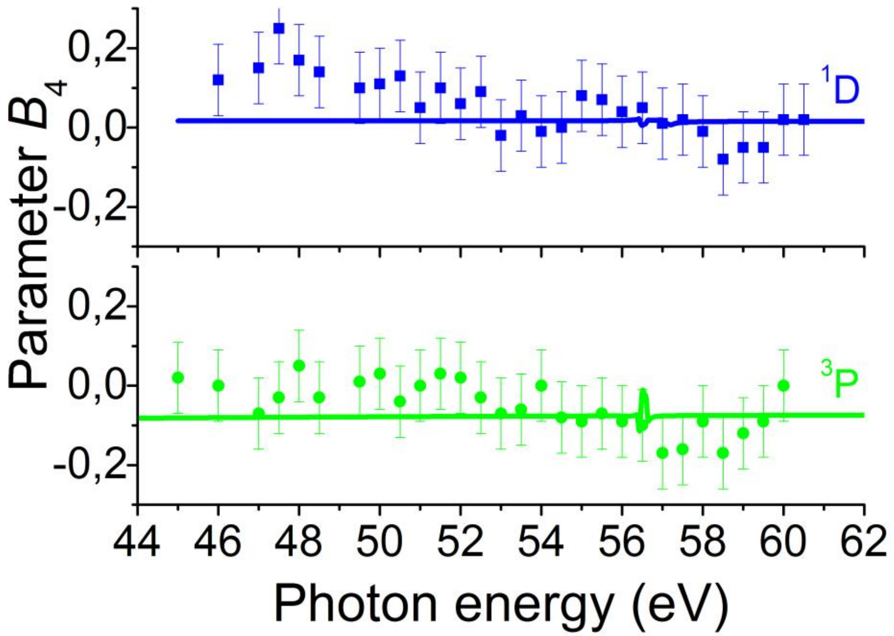


Photon energy (eV)

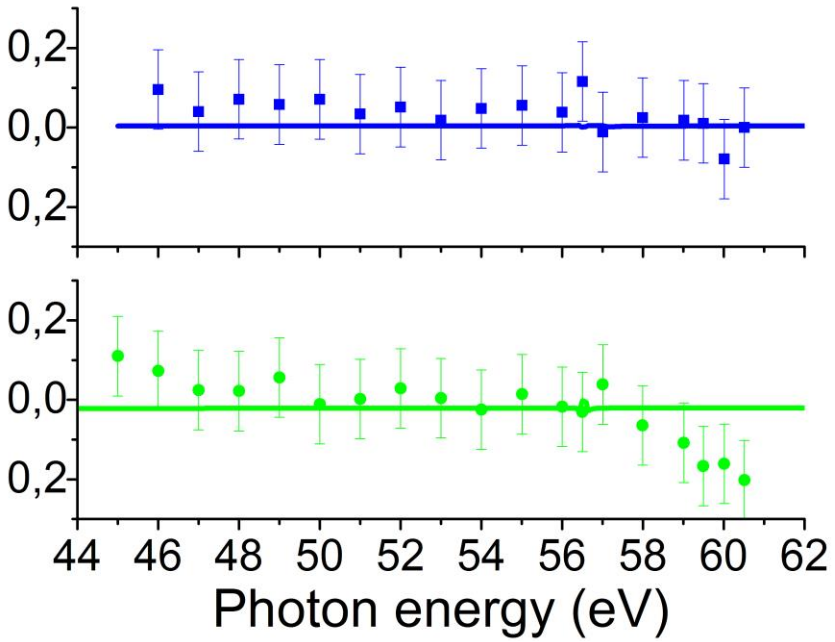
- ✓ *Good agreement between experiment and theory*
- ✓ *Presence of sharp resonances around 56 eV (autoionizing states)*
- ▪ *Overlap between 2s and* ^{1}*S photoelectrons*

PES (Photo Electron Spectra): Angular Distribution (β_4 parameter)

Linear Polarization



Circular Polarization



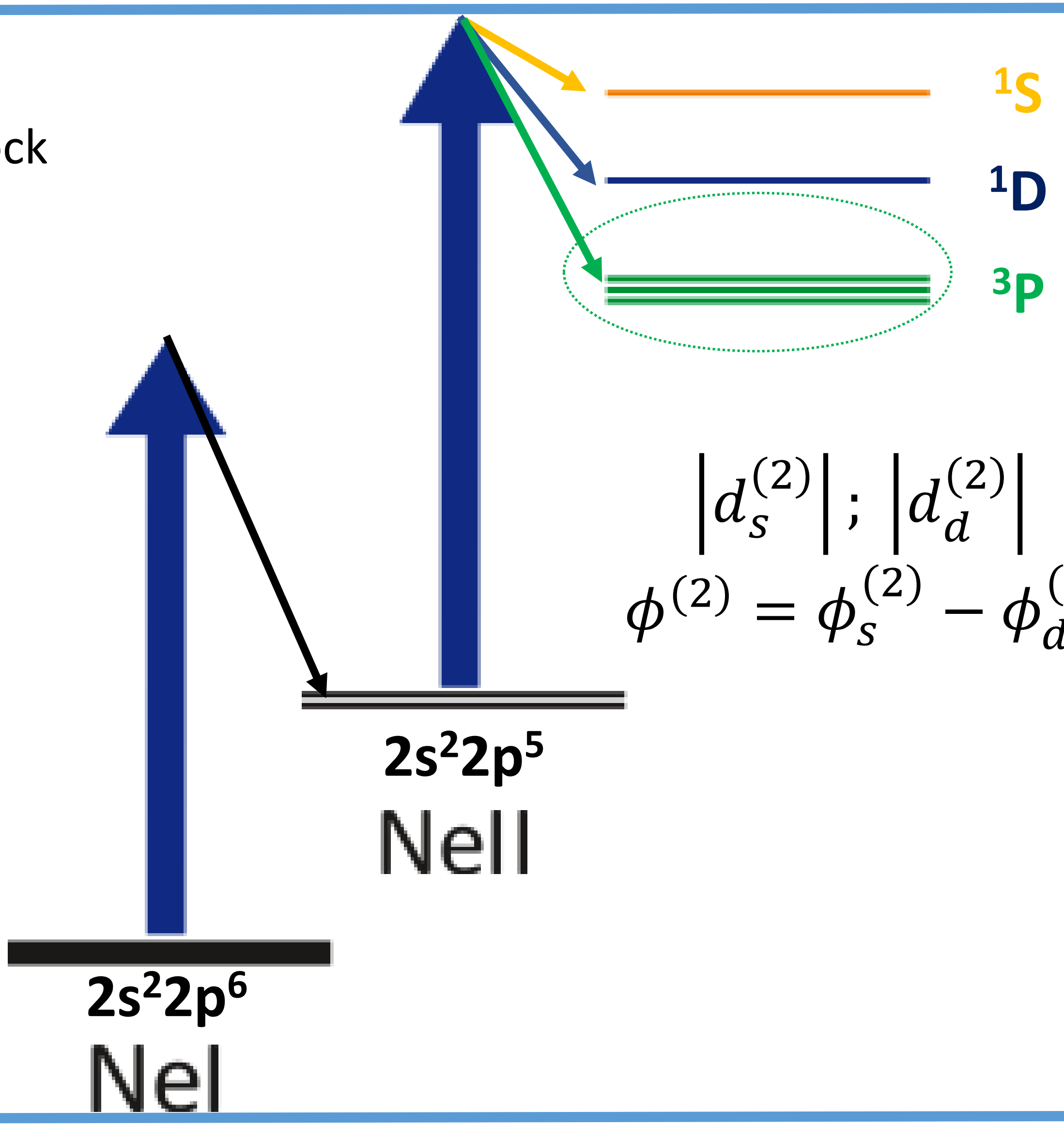
- ✓ *Good agreement between experiment and theory*
- ✓ *Small alignment of the intermediate ion (Ne^+)*

Complete experiment: the model

1) Two-step process: sequential ionization
2) Matrix elements multiconfiguration Hartree-Fock (MCHF) approach
3) Incoherent excitation of intermediate states (due to XUV pulse duration)

Cooper-Zare model:
Each photoionization step is described by three quantities:

$$\left|d_s^{(1)}\right|;\ \left|d_d^{(1)}\right|$$
$$\phi^{(1)} = \phi_s^{(1)} - \phi_d^{(1)}$$



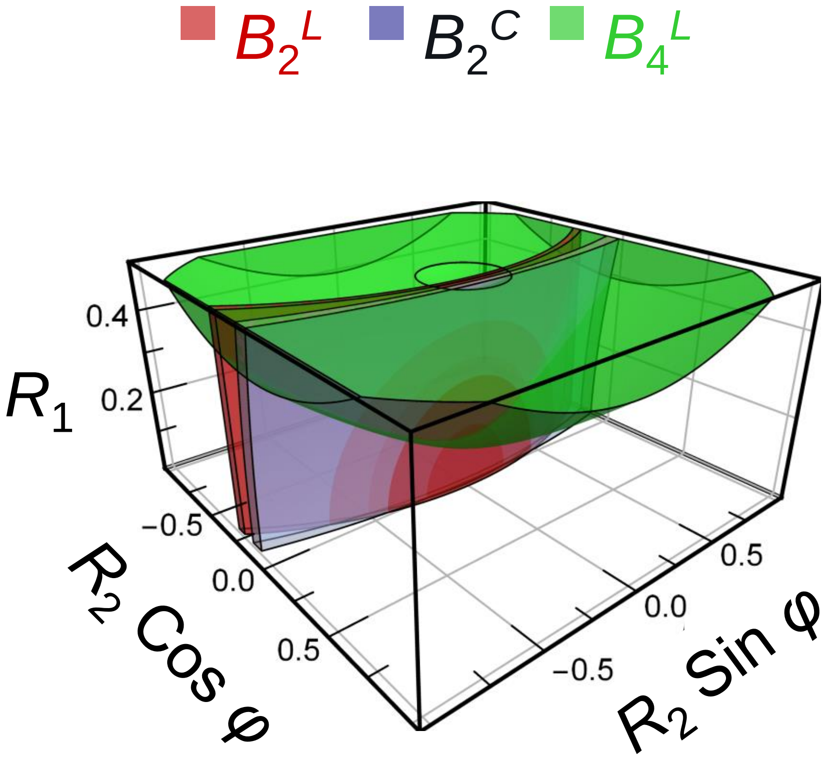
$$\left|d_s^{(2)}\right|;\ \left|d_d^{(2)}\right|$$
$$\phi^{(2)} = \phi_s^{(2)} - \phi_d^{(2)}$$

E. Gryzlova *et al.* J. Phys. B: At. Mol. Opt. Phys. **43**, 225602 (2010)
L. A. A. Nikolopoulos Phys. Rev. Lett. 111, 093001 (2013)

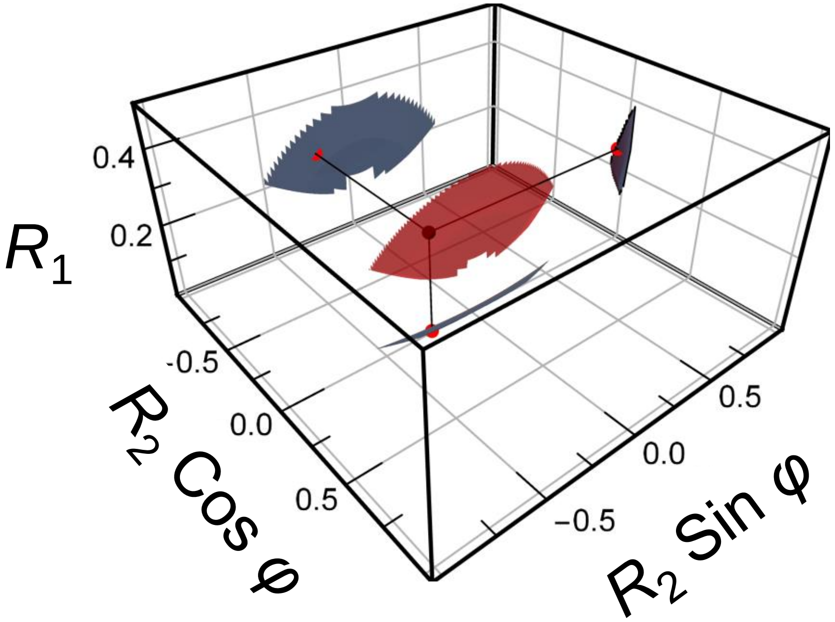
Complete experiment: experiment vs theory

- $Ratio\ 1^{st}\ step\ R_1 = \left|d_s^{(1)}/d_d^{(1)}\right|$
- $Ratio\ 2^{nd}\ step\ R_2 = \left|d_s^{(2)}/d_d^{(2)}\right|$
- $Relative\ phase\ 2^{nd}\ step\ \phi = \phi_s^{(2)} - \phi_d^{(2)}$

$$B_2^L = f(R_1, R_2, \phi)$$
$$B_2^C = g(R_1, R_2, \phi)$$
$$B_4^L = h(R_1, R_2, \phi)$$



Intersection

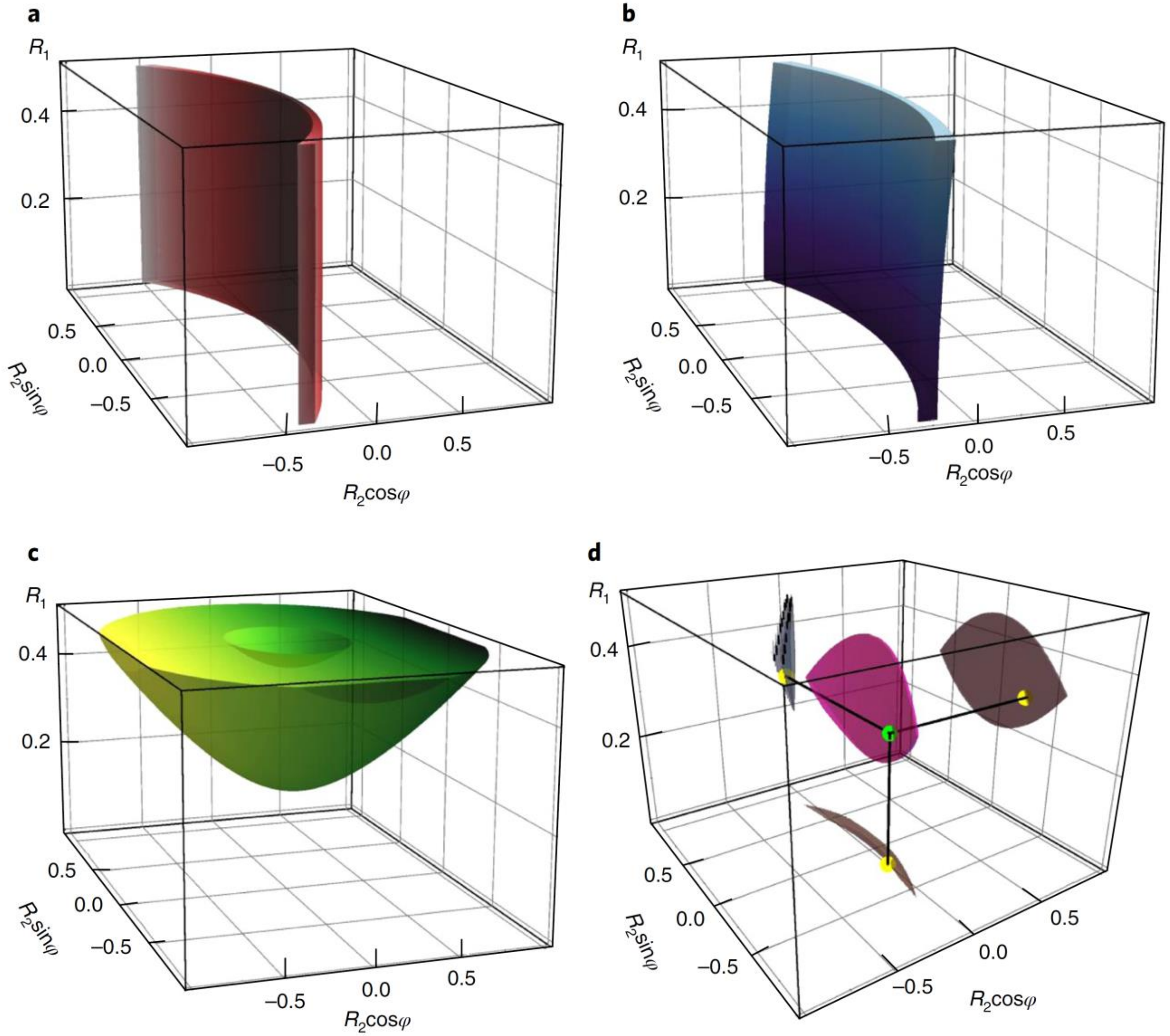


● Simulations

Complete experiment: experiment vs theory

β_2^L

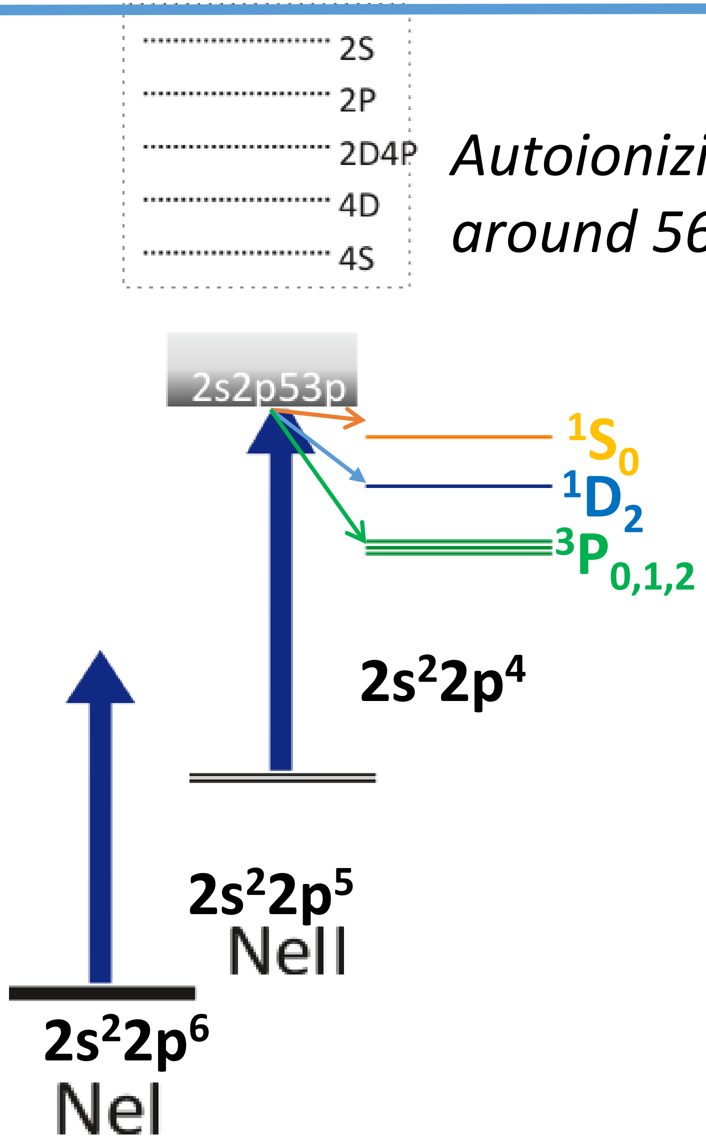
β_2^C



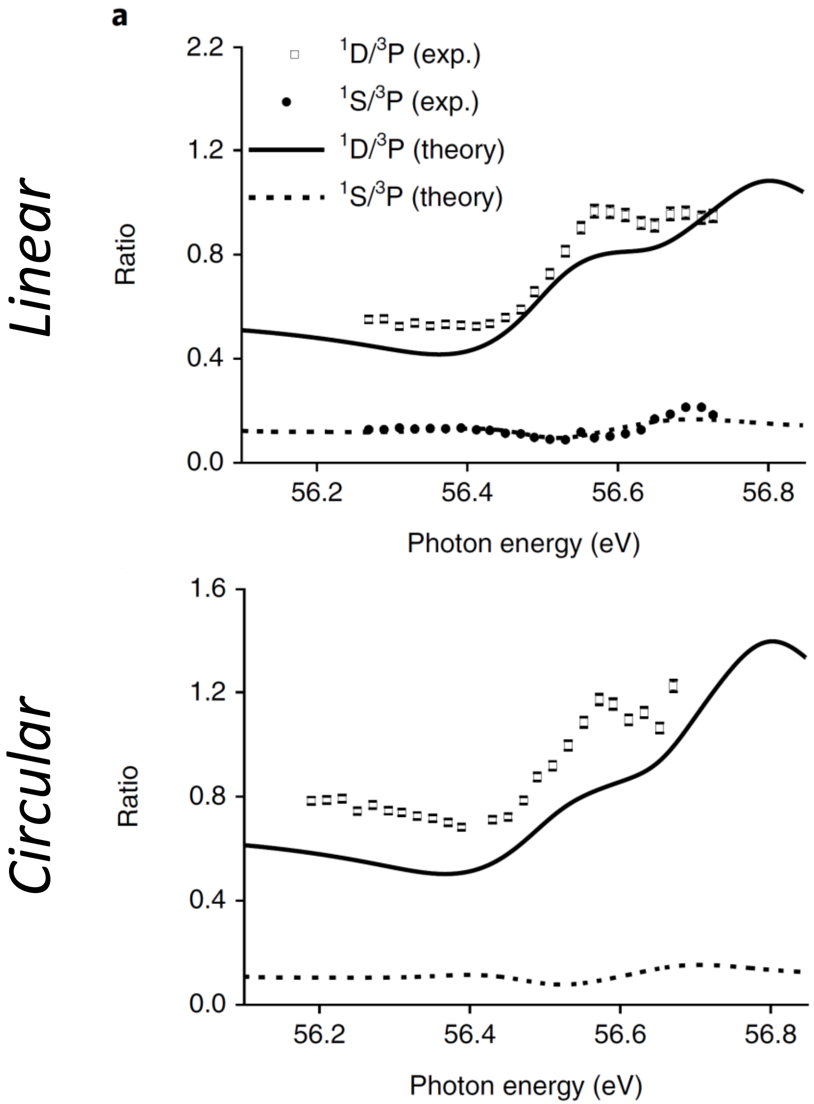
β_4^L

intersection

Autoionizing states (AIS) in TPDI: ratios

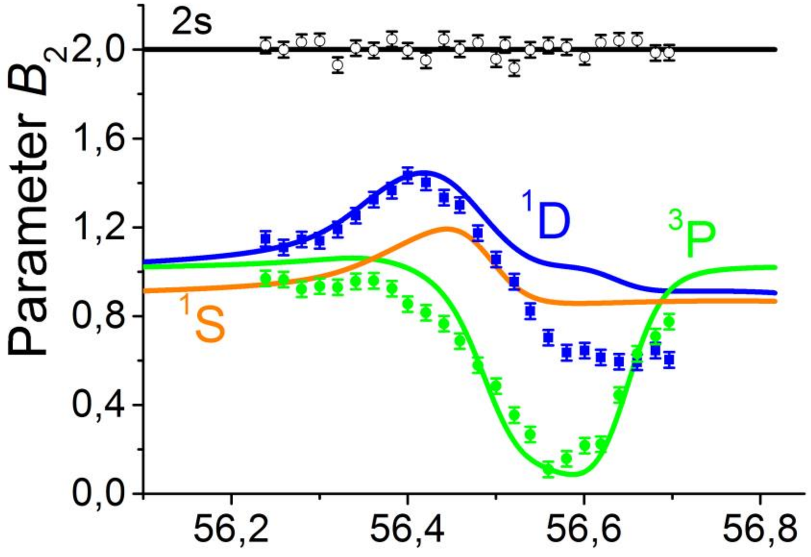


Autoionizing states around 56.4 eV



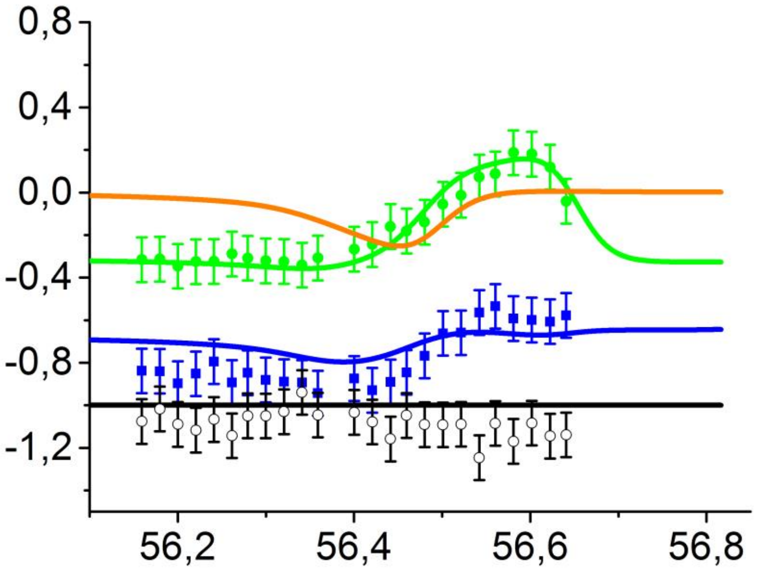
Autoionizing states (AIS) in TPDI: angular Distribution (β_2 parameter)

Linear Polarization



Photon energy (eV)

Circular Polarization

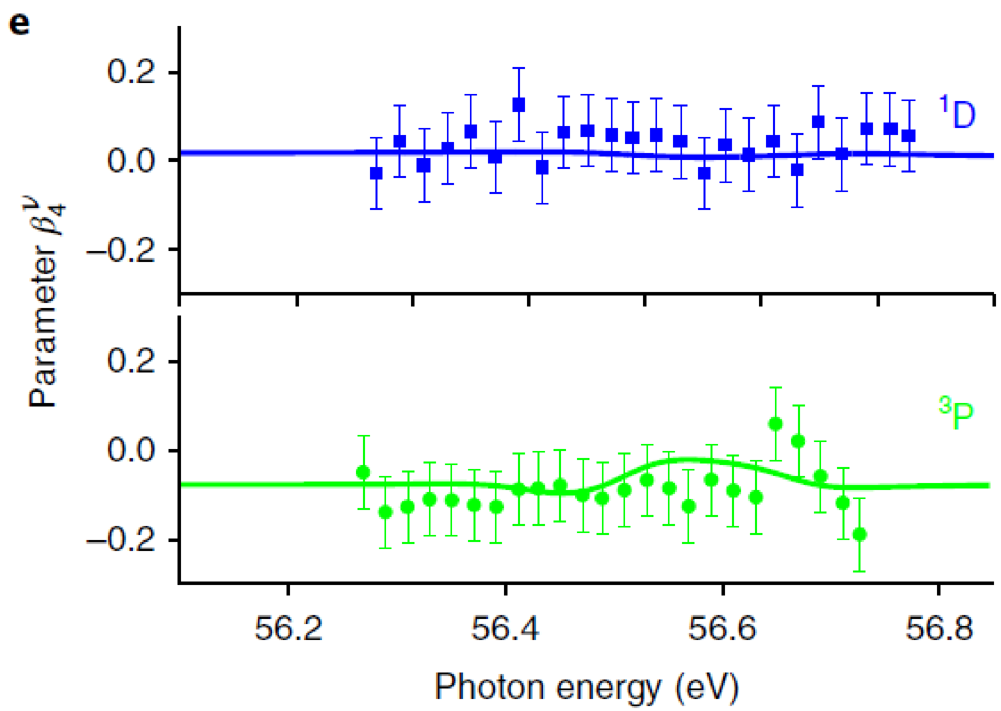


Photon energy (eV)

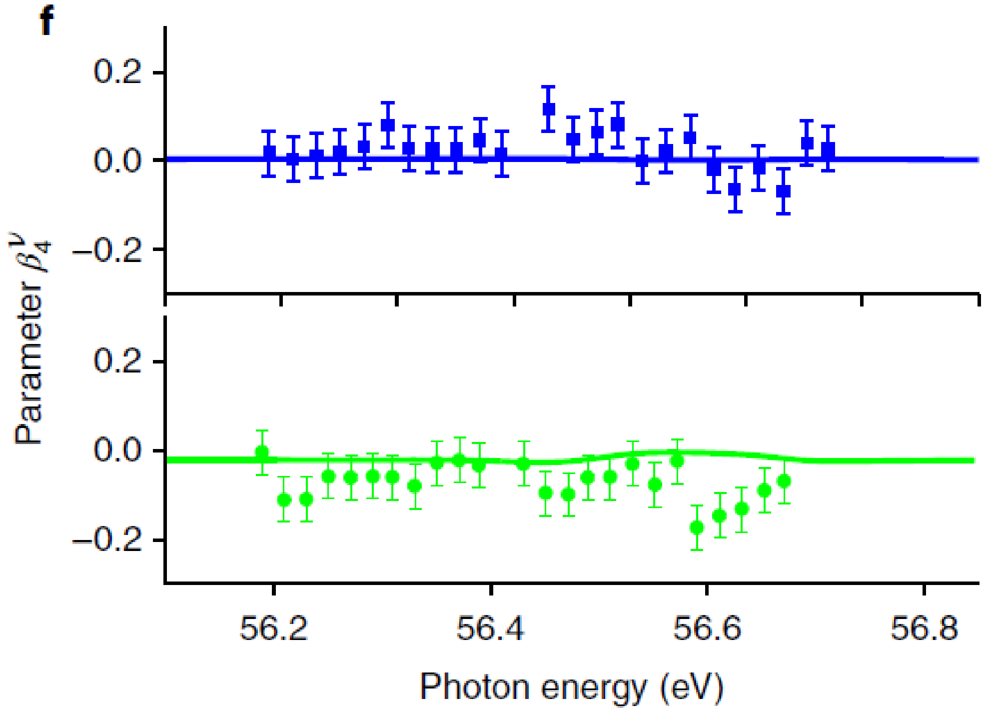
- ✓ *Effect of the autoionizing states on the PADs*
- ✓ *Good agreement between experiment and theory for circular and linear polarization*
- ✓ *2s PAD as a benchmark of the data quality*

Autoionizing states (AIS) in TPDI: Angular Distribution (β_4 parameter)

Linear Polarization



Circular Polarization



Complete experiment for AIS: theory vs experiment

Converging to the non-resonant model

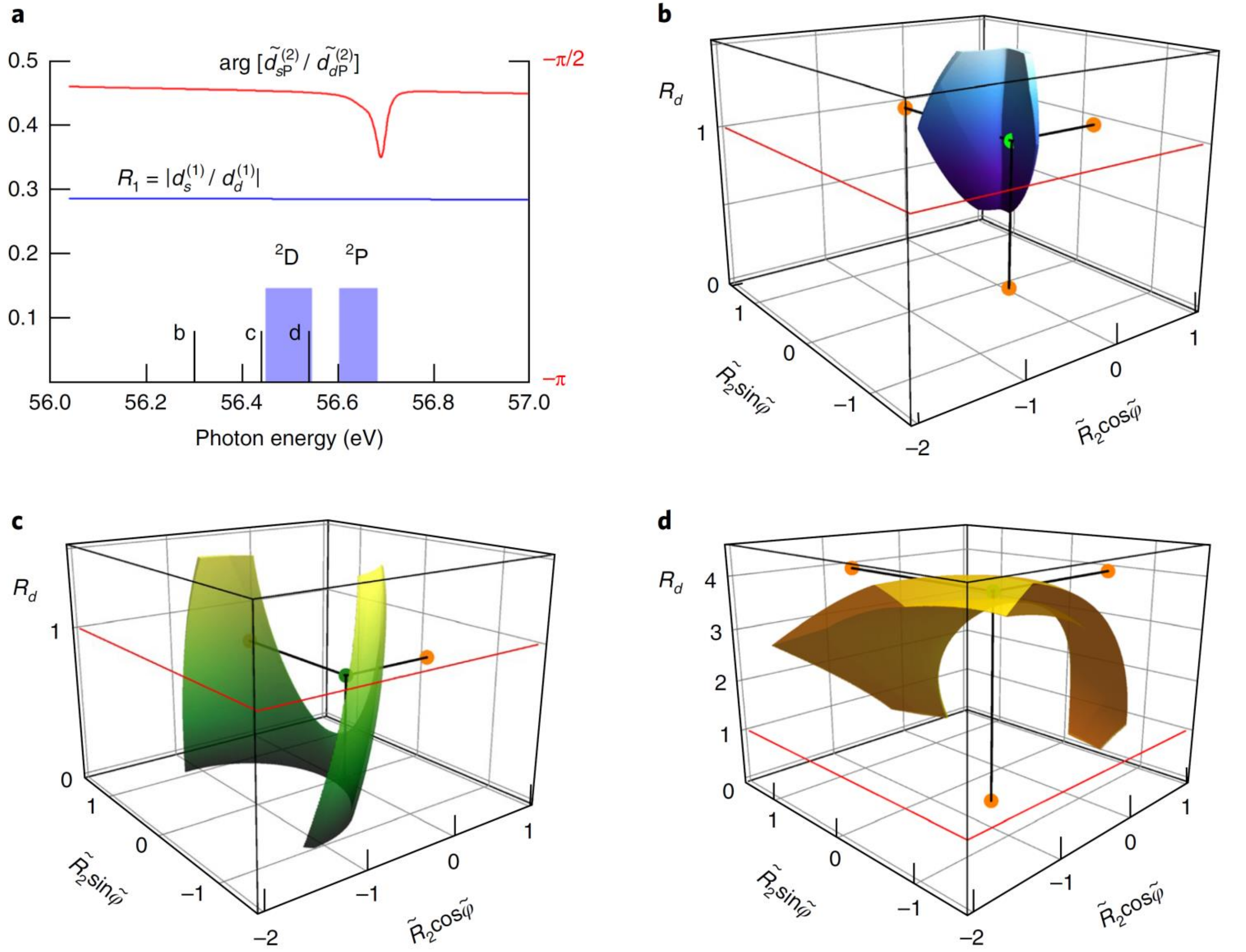
$$\gamma + \mathrm{Ne}^{+}(2s^2 2p^5\ {}^2\mathrm{P}) \rightarrow \mathrm{Ne}^{2+}(2s^2 2p^4\ {}^3\mathrm{P}) + \begin{Bmatrix} e^-(Es): & {}^2\mathrm{P} \\ e^-(Ed): & {}^2\mathrm{P} \\ e^-(Ed): & {}^2\mathrm{D} \end{Bmatrix}$$

$$\tilde{R}_2 = |\tilde{d}^{(2)}_{sP} / \tilde{d}^{(2)}_{dD}|$$

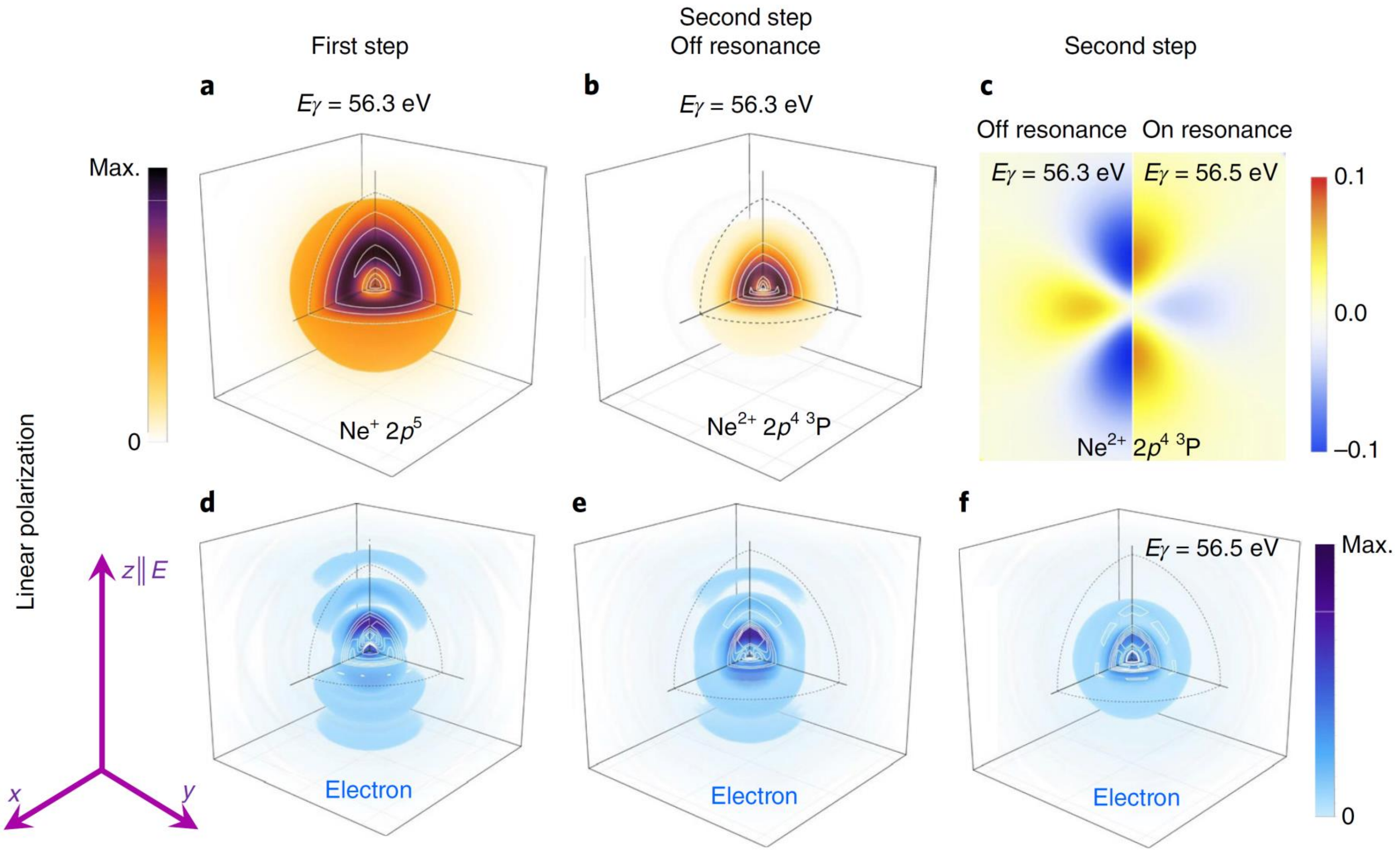
$$R_d = |\tilde{d}^{(2)}_{dP} / \tilde{d}^{(2)}_{dD}|$$

$$\tilde{\varphi} = \arg(\tilde{d}^{(2)}_{sP} / \tilde{d}^{(2)}_{dD})$$

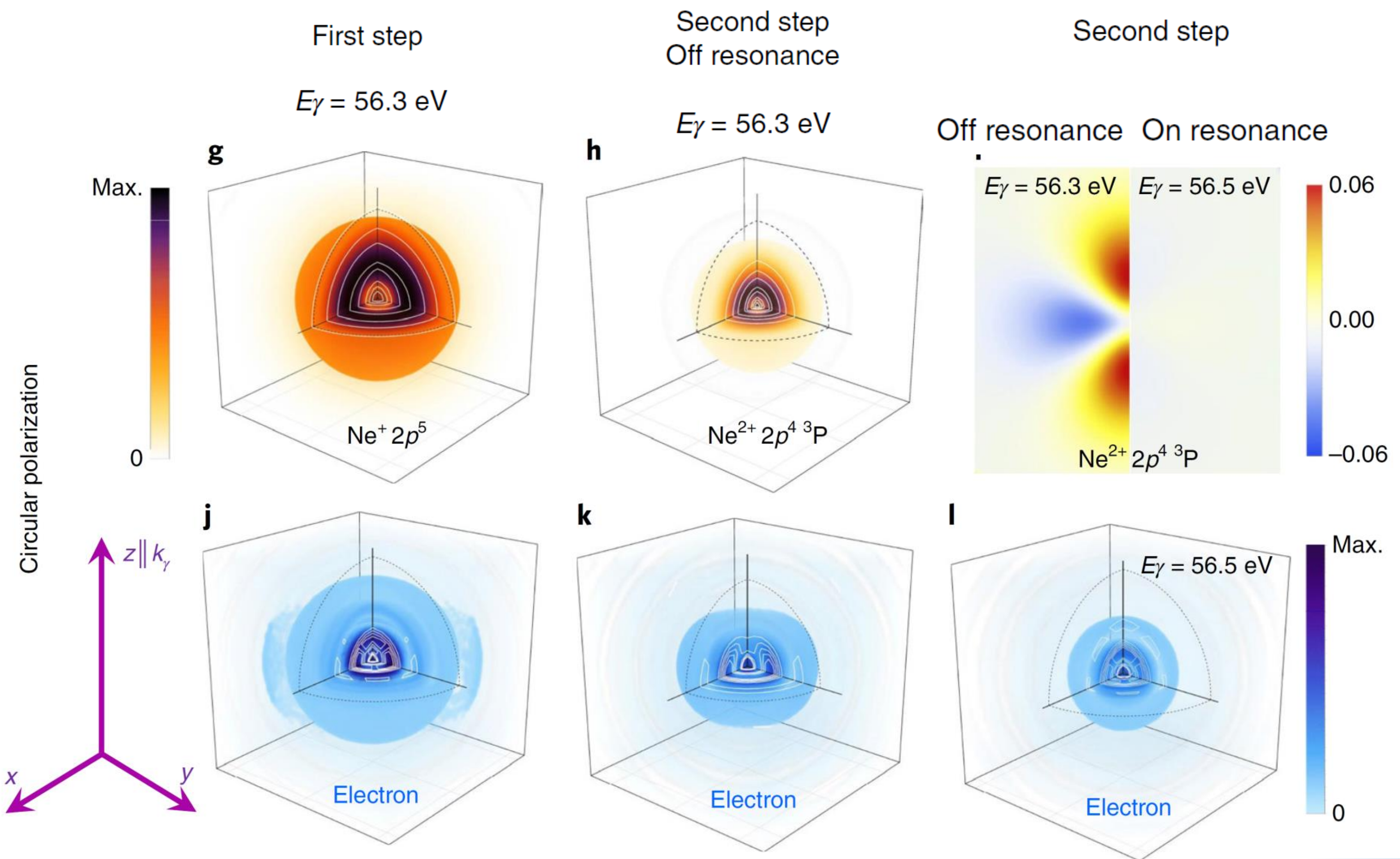
Parameters in the CE at the ^{2}D resonance



Complete experiment (linear polarization): residual ion polarization and electron scattering



Complete experiment (circular polarization): residual ion polarization and electron scattering



Conclusions

- Intense, tunable XUV pulses from FERMI
- Sequential two photon double ionization
  ➔ alignment of Ne^{+}
- Observation of PE peak intensity and angular distribution with VMI
- First complete experiment of photoionization in an ion
- Reconstruction for resonant and non-resonant ionization
- Determination of the observable quantities in photoionization