# Elliptical dichroism in non-linear atomic ionization

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seit 1558

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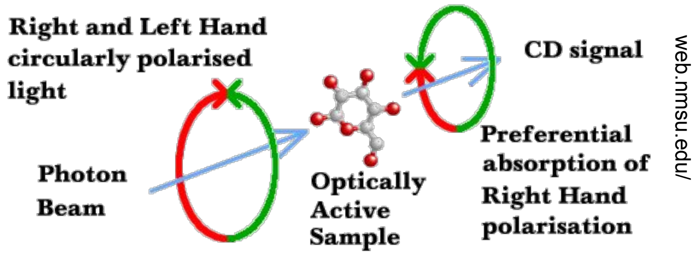
# Circular dichroism (CD)

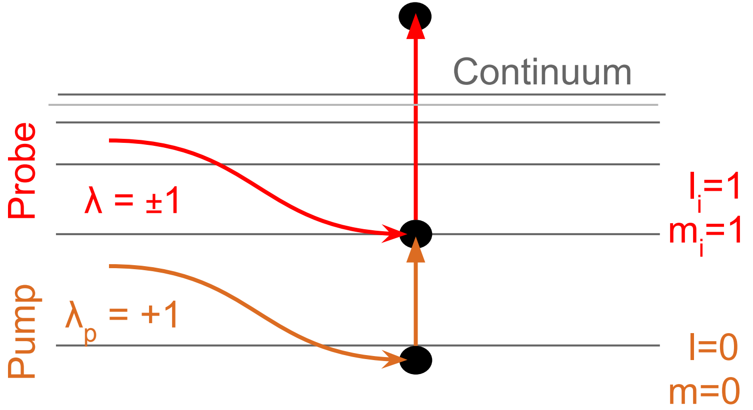



## Widely used in:

- Chemistry
- Biology
- Metamaterials
- Magnetic properties of atoms

## Example from atomic physics

Photoionization of H




### Selection rules

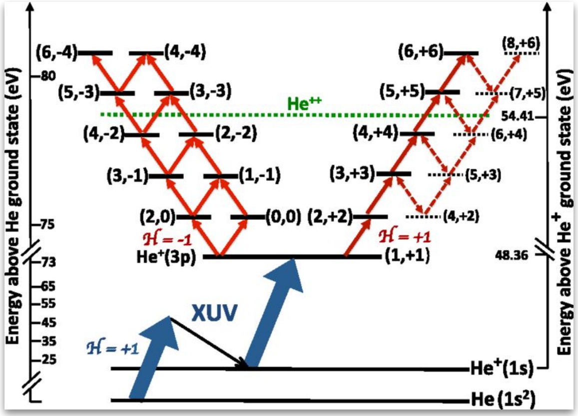
$$m_f = m_i + \lambda$$

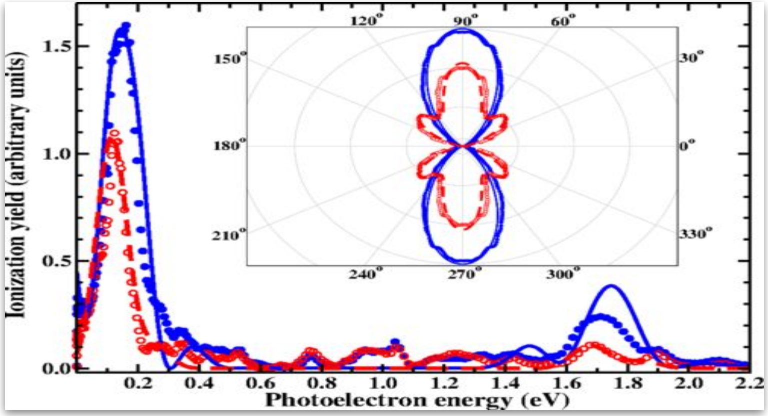
| Left-handedness |  | Right-handedness |
|---|---|---|
| $\lambda = -1$ |  | $\lambda = +1$ |
| $l_f=0$, $m_f=0$ | $l_f=2$, $m_f=0$ | $l_f=2$, $m_f=2$ |

# Circular dichroism - experiment

## Two-color many-photon ionization of He

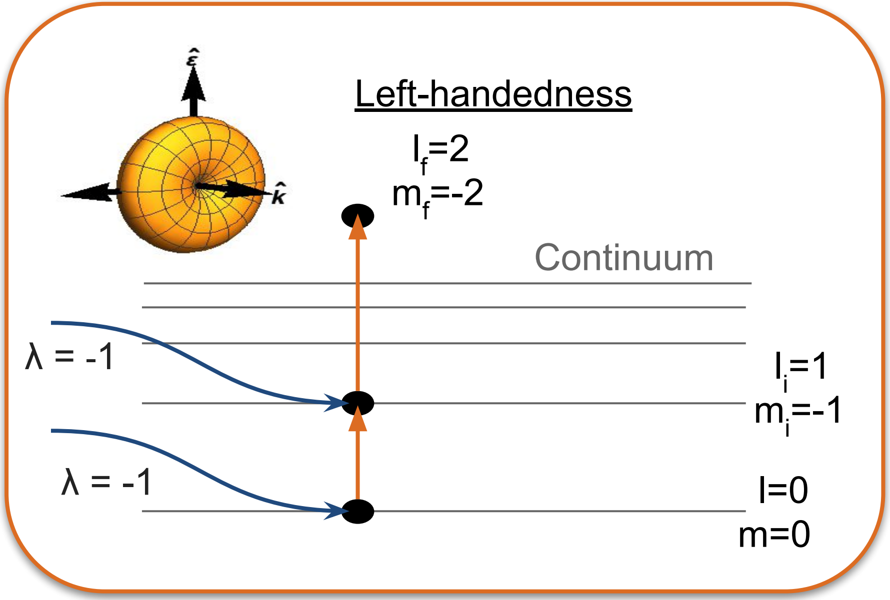
1. Right-handed XUV beam ionizes and orients He atoms
2. Right-/Left- handed IR beam ionizes $He^+$ ion
3. Circular dichroism in both total and differential cross sections

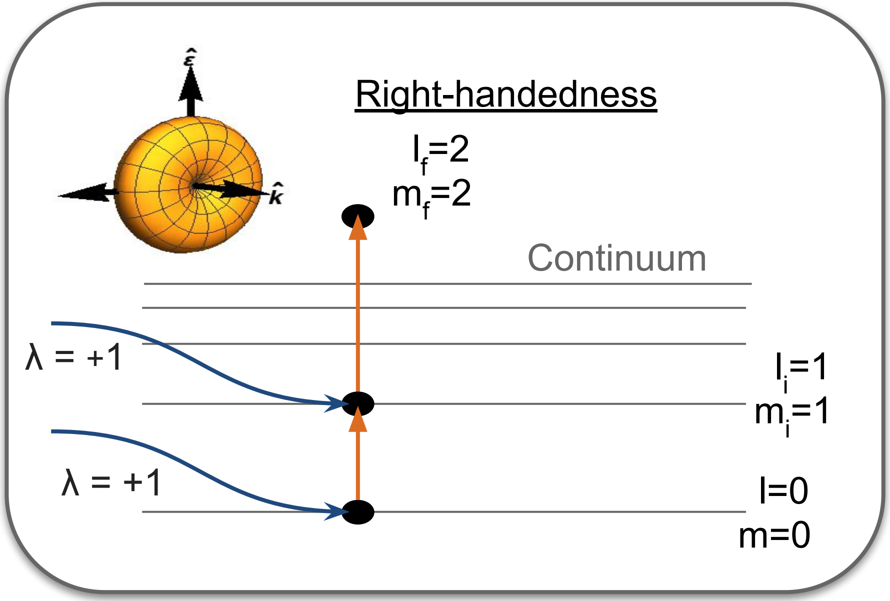



M. Ilchen, et al., PRL **118**, 013002 (2017).

# Circular dichroism with a single pulse?



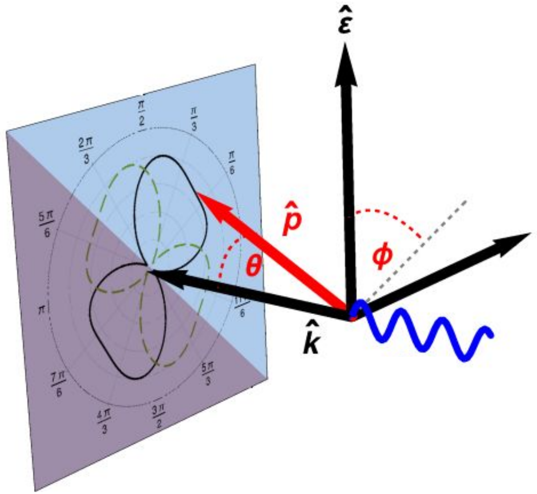



# Discovery of elliptical dichroism (ED)

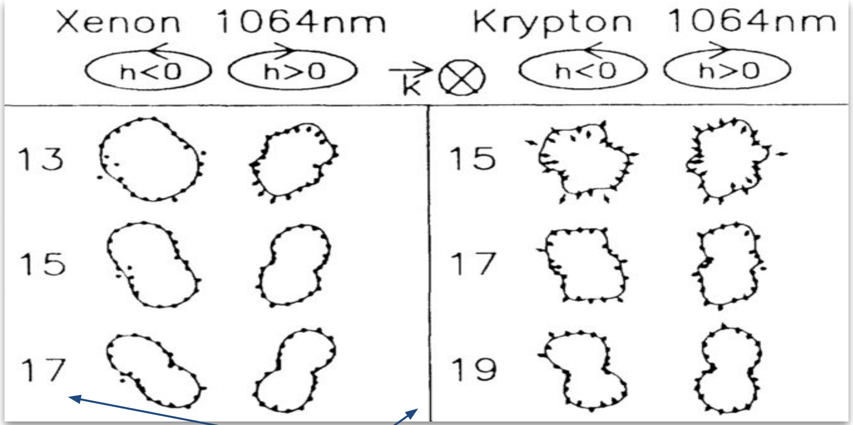
## First detection of ED in photoelectron distributions

1988 - Above threshold ionization of noble gases

Unclear origin of the effect to the authors



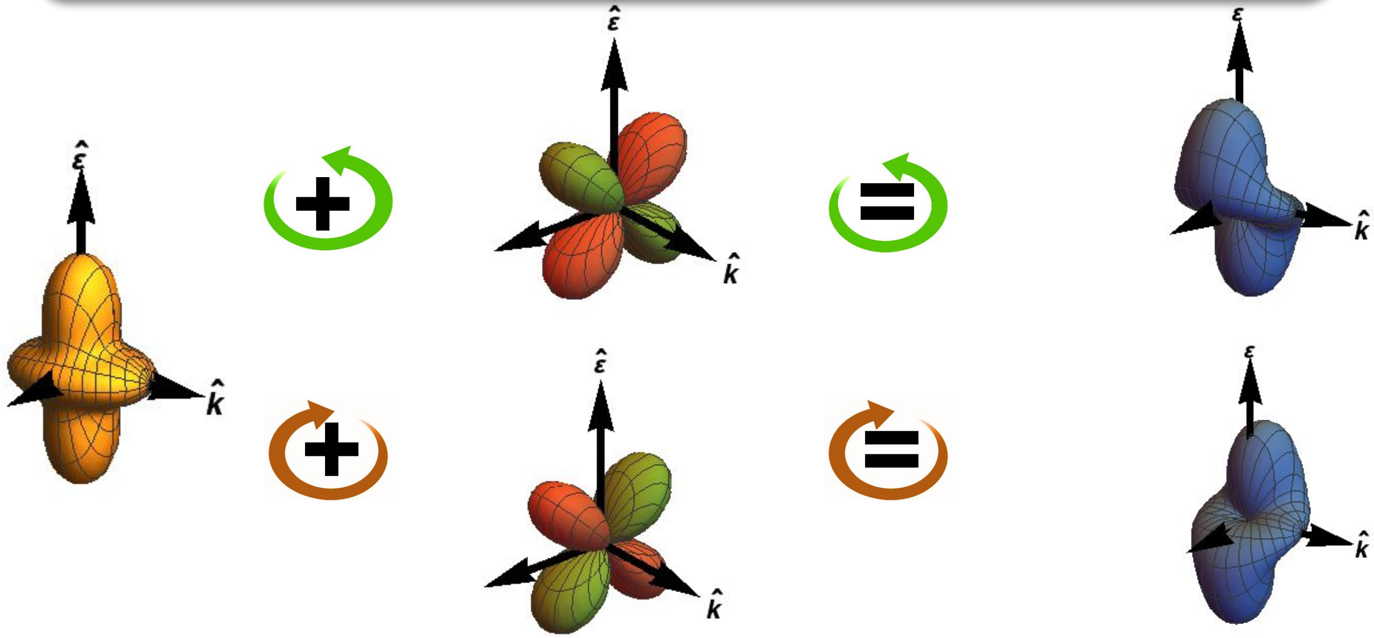

M. Bashkansky, P. H. Bucksbaum, and D. W. Schumacher, PRL **60**, 2458 (1988).




Number of absorbed photons

# Understanding of ED

A. Kassaee, M. L. Rustgi, and S. A. T. Long, Phys. Rev. A **37**, 999 (1988).
P. Lambropoulos, and X. Tang, Phys. Rev. Lett. **61**, 2506 (1988).
H. G. Muller, G. Petite, and P. Agostini, Phys. Rev. Lett. **61**, 2507 (1988).

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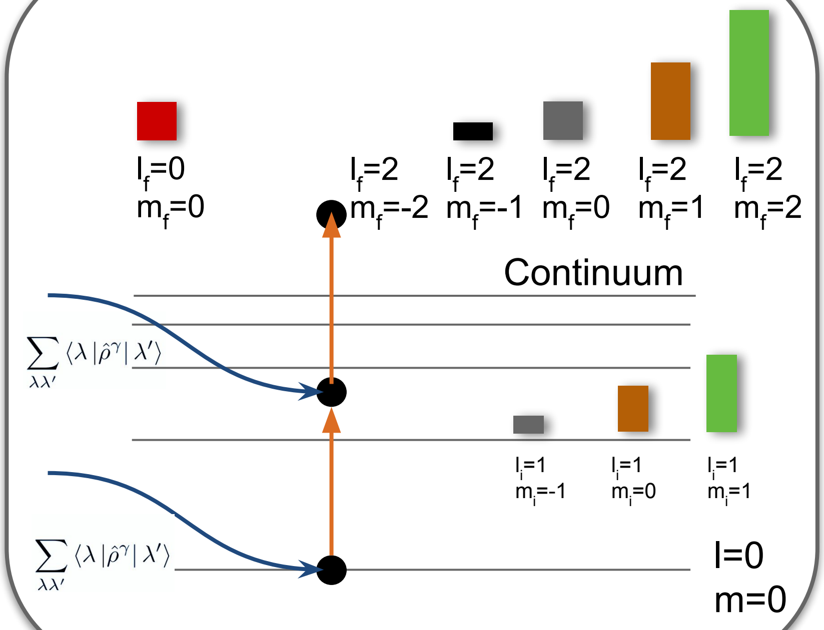
# Physical understanding

Consider a resonant two-photon ionization




<u>Two steps:</u>

Step 1:

Excitation of the neutral unpolarized atom

Step 2:

Ionization of the excited atom

# Further efforts

**Asymmetric angular distributions in multiphoton ionisation of helium by elliptically polarised light**

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Asymmetries in the angular distributions of above threshold ionization in an elliptically polarized laser field

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ember 1998; received in revised form 18 February 1999; accepted 4 March 1999

VOLUME 84, NUMBER 17 PHYSICAL REVIEW LETTERS 24 APRIL 2000

**Above-Threshold Ionization by an Elliptically Polarized Field: Interplay between Electronic Quantum Trajectories**

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R. Kopold and W. Becker‡

*Max-Born-Institut, 12489 Berlin, Germany*

(Received 9 December 1999)

**Elliptic dichroism and angular distribution of electrons in two-photon ionization of atoms**

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VOLUME 87, NUMBER 13 PHYSICAL REVIEW LETTERS 24 SEPTEMBER 2001

**Threshold Effects on Angular Distributions for Multiphoton Detachment by Intense Ell**

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[1]*Department of Physics and Astronomy, Th*

[2]*Department of Physics, Voron*

(Received 7 May 20

PRL **93,** 233002 (2004) PHYSICAL REVIEW LETTERS week ending 3 DECEMBER 2004

**Coulomb Asymmetry in Above-Threshold Ionization**

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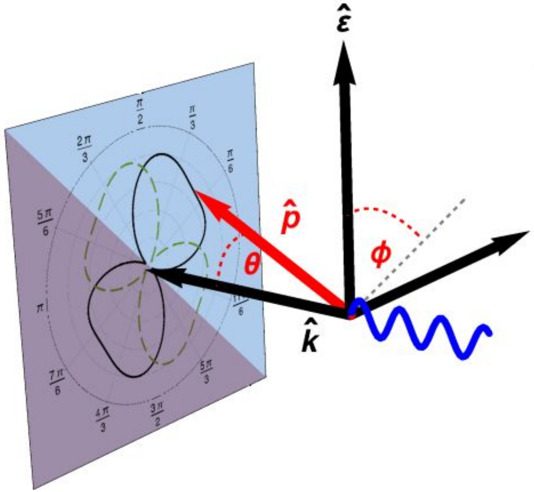
Can we predict elliptical dichroism without calculations?

Can we find the maximum effect without calculations?

Yep!

# Can we find maximum ED?




What do we mean by maximum?

$$\Delta_{ed}(\pi/2, \phi) = \frac{\sigma_+(\pi/2, \phi) - \sigma_-(\pi/2, \phi)}{\sigma_+(\pi/2, \phi) + \sigma_-(\pi/2, \phi)} = \pm 1$$

Zero emission at a given angle

$$\sigma_\pm(\pi/2, \phi) = 0$$

$$\sigma_\mp(\pi/2, \phi) \neq 0$$

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# Two-photon ionization of *s* electron

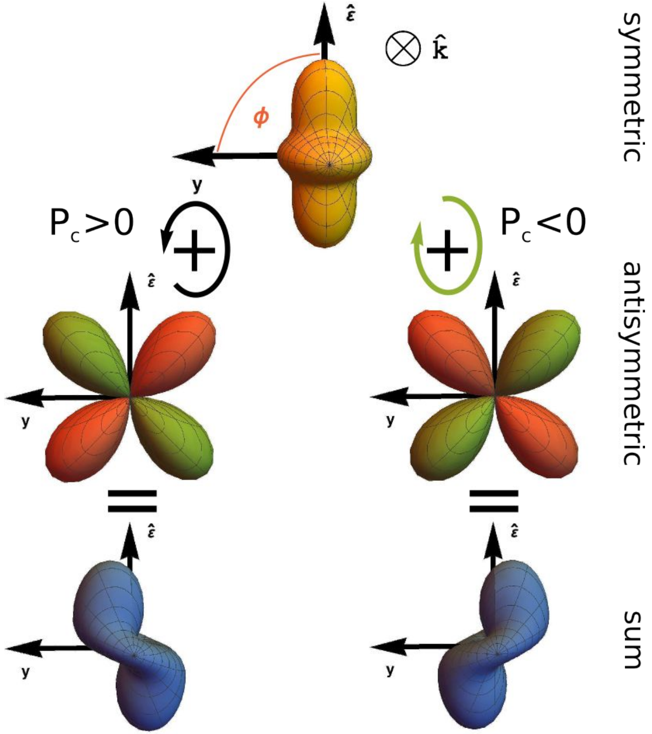
$$\frac{d\sigma}{d\Omega} = \frac{8\pi^3\alpha^2}{\omega^2}\frac{1}{[j_a]}\sum_{\substack{\lambda_1\lambda_2\\ \lambda_1'\lambda_2'}} \langle \boldsymbol{k}\lambda_1 |\hat{\rho}| \boldsymbol{k}\lambda_1'\rangle \langle \boldsymbol{k}\lambda_2 |\hat{\rho}| \boldsymbol{k}\lambda_2'\rangle$$

$$\times \sum_{m_e m_a} M^{\lambda_1\lambda_2}_{m_e m_a} M^{\lambda_1'\lambda_2' *}_{m_e m_a}.$$

$$M^{\lambda_1\lambda_2}_{m_e m_a} = \sum\!\!\!\!\!\!\int_n \frac{\langle \boldsymbol{p}_e m_e | \boldsymbol{\alpha}\cdot\boldsymbol{A}_{\lambda_2} | n\rangle\langle n | \boldsymbol{\alpha}\cdot\boldsymbol{A}_{\lambda_1} | n_a\kappa_a m_a\rangle}{E_{n_a\kappa_a} + \omega - E_{n_n\kappa_n}}$$

$$\frac{d\sigma^{(nonrel.)}}{d\Omega} = \frac{9\pi^2\alpha^2}{2\omega^2}\Big\{|U_s|^2\mathcal{P} + |U_d|^2\Big[\mathcal{P} - 3\sin^2\theta\big(\mathcal{P} + 2P_l\cos(2\phi)\big) + \frac{9}{2}\sin^4\theta\big(1 + P_l\cos(2\phi)\big)^2\Big] + 2\mathrm{Re}\Big[U_s U_d^* e^{i(\Delta_s-\Delta_d)}\big[\mathcal{P} - \frac{3}{2}\sin^2\theta\big(\mathcal{P} + 2P_l\cos(2\phi) + 2iP_lP_c\sin(2\phi)\big)\big]\Big]\Big\},$$

## Second order perturbation theory



# Two-photon ionization of *s* electron

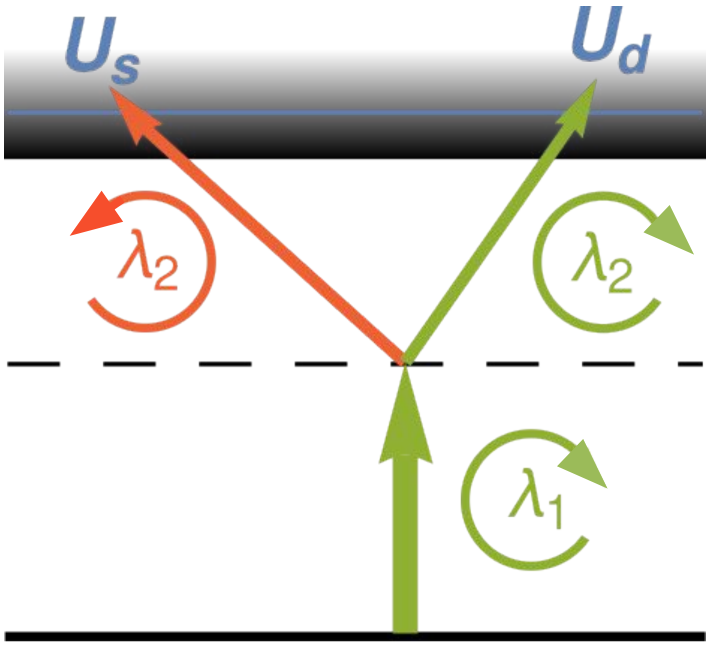
Analytical analysis gives:

$$\sigma_\pm(\pi/2,\phi) \propto \delta_s - \delta_d\,,\; U_{rat} = \frac{U_s - U_d}{U_s + U_d}$$

For any phase difference:

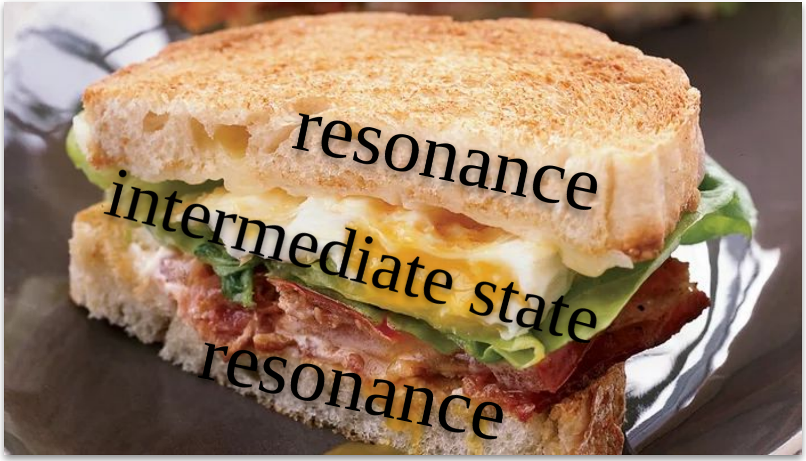
$$U_{rat} \approx 1/3$$

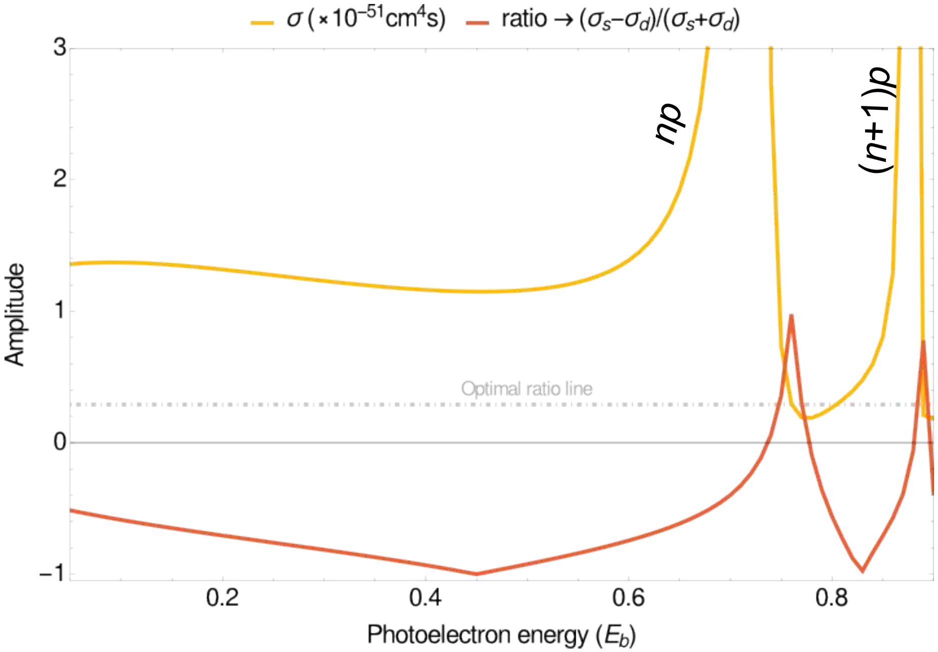
guarantees a maximum effect



# Below threshold ionization

Sandwiching intermediate state between two resonances guarantees maximum elliptical dichroism for all atoms

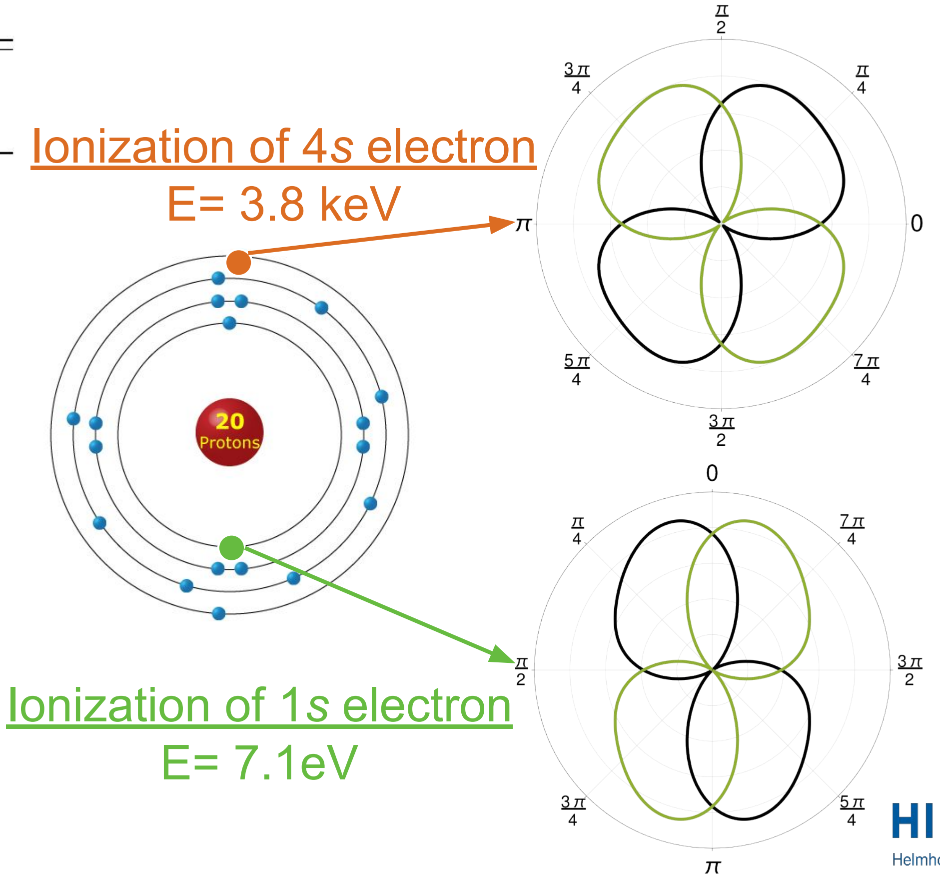



$$\sigma \propto \frac{\langle f|D|n_1\rangle\langle n_1|D|i\rangle}{E_i+\omega-E_{n_1}} + \frac{\langle f|D|n_2\rangle\langle n_2|D|i\rangle}{E_i+\omega-E_{n_2}}$$

# Example: Two-photon ionization of Ca

| Atom | Ionized electron | Intermediate resonances | $\omega$ (eV) | $\phi_{max}$(deg) |
|---|---|---|---|---|
| Ca | $1s$ | $2p - 3p$ | 3807 | 52.1 |
| | | | 3898 | 52.7 |
| | | $3p - 4p$ | 4030 | 49.8 |
| | | | 4036 | 50.4 |
| | $2s$ | $3p - 4p$ | 434 | 87.6 |
| | | | 436 | 87.1 |
| | | $4p - 5p$ | 446 | 86.2 |
| | | | 447 | 86.2 |
| | $3s$ | $3p - 4p$ | below threshold | — |
| | | | 39.2 | 68.2 |
| | | $4p - 5p$ | 54.4 | 49.8 |
| | | | 55.5 | 51.0 |
| | $4s$ | $4p - 5p$ | 3.50 | 44.1 |
| | | | 3.52 | 61.3 |
| | | $5p - 6p$ | 4.18 | 34.4 |
| | | | 4.23 | 69.3 |

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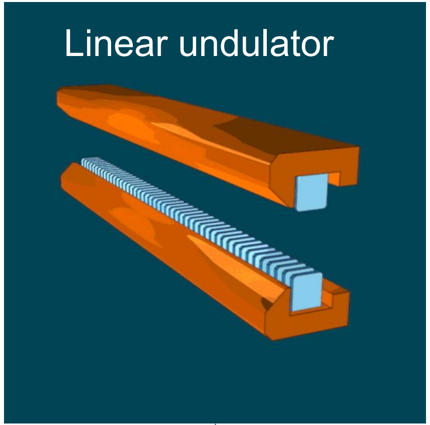
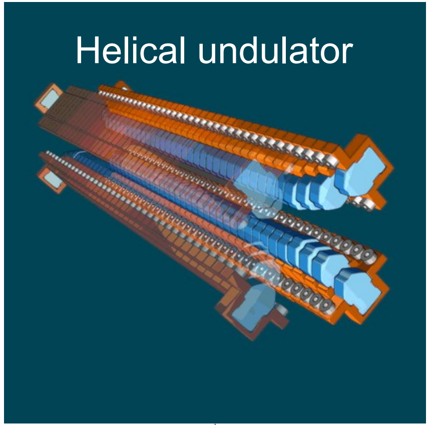
Can we verify it experimentally?

# Polarization control of FELs




https://www6.slac.stanford.edu




Common linear undulator

Delta undulator

Further polarization control upgrade plans in other facilities

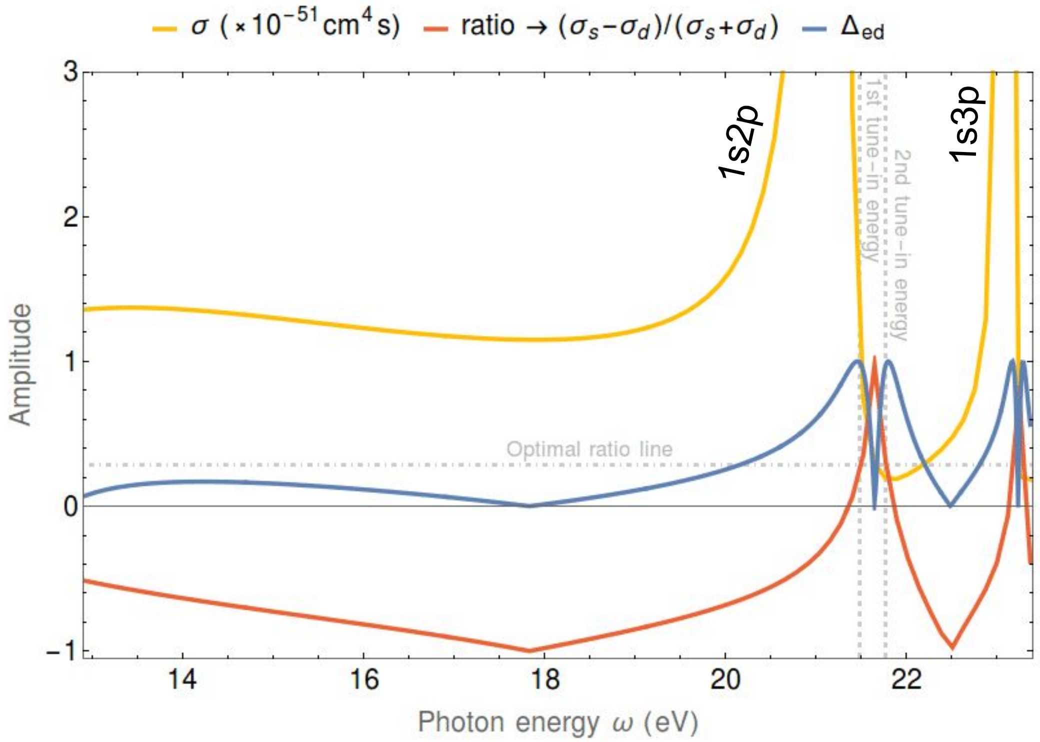
**Apple II undulator at FERMI**

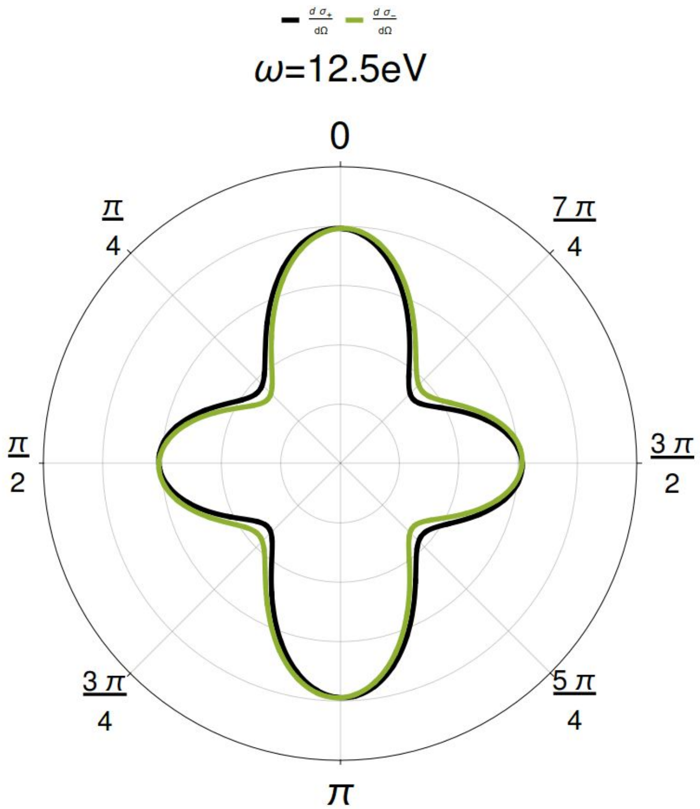
20 - 120 eV
~$10^{12}$ photons/pulse
~0.1 eV Energy resolution

**Delta undulator at LCLS**

500 - 1200 eV
~$10^{11}$ photons/pulse
~1 eV Energy resolution

# Experimental verification: He



# Take home message

Elliptical dichroism requires no asymmetry of target

In two-photon ionization of s-state, maximal ED is always present

Promising experimental possibilities

# Acknowledgement

Many thanks to:

Markus Ilchen
and
Michael Meyer

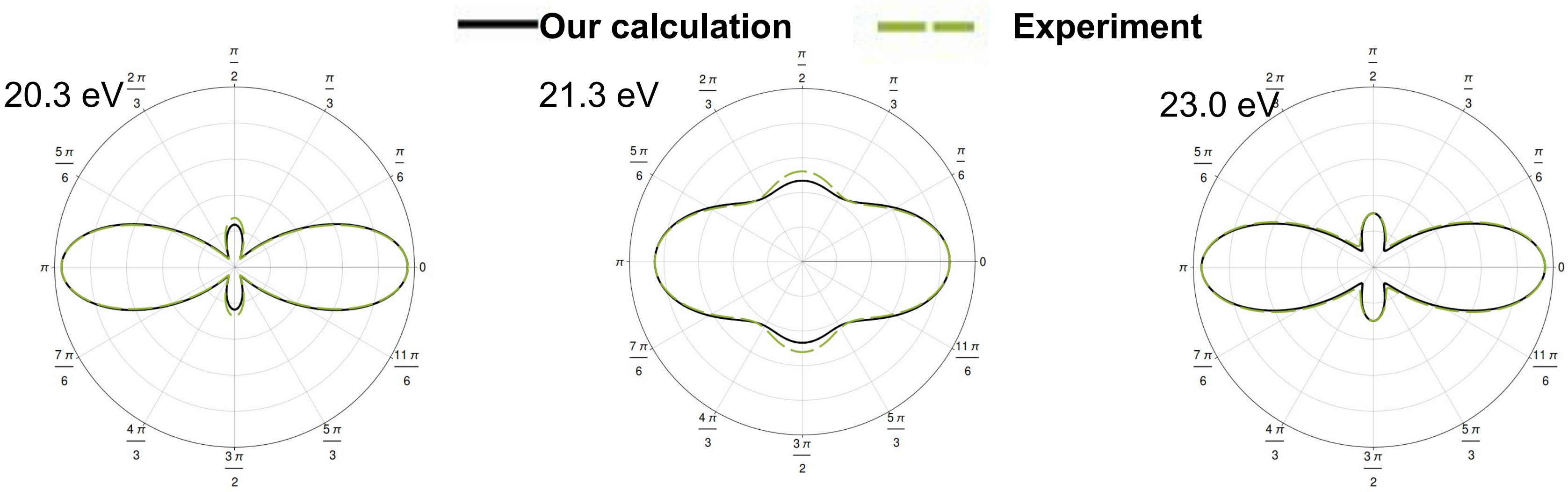
From the European XFEL

# Thank you for your attention!

# Comparison with experiment

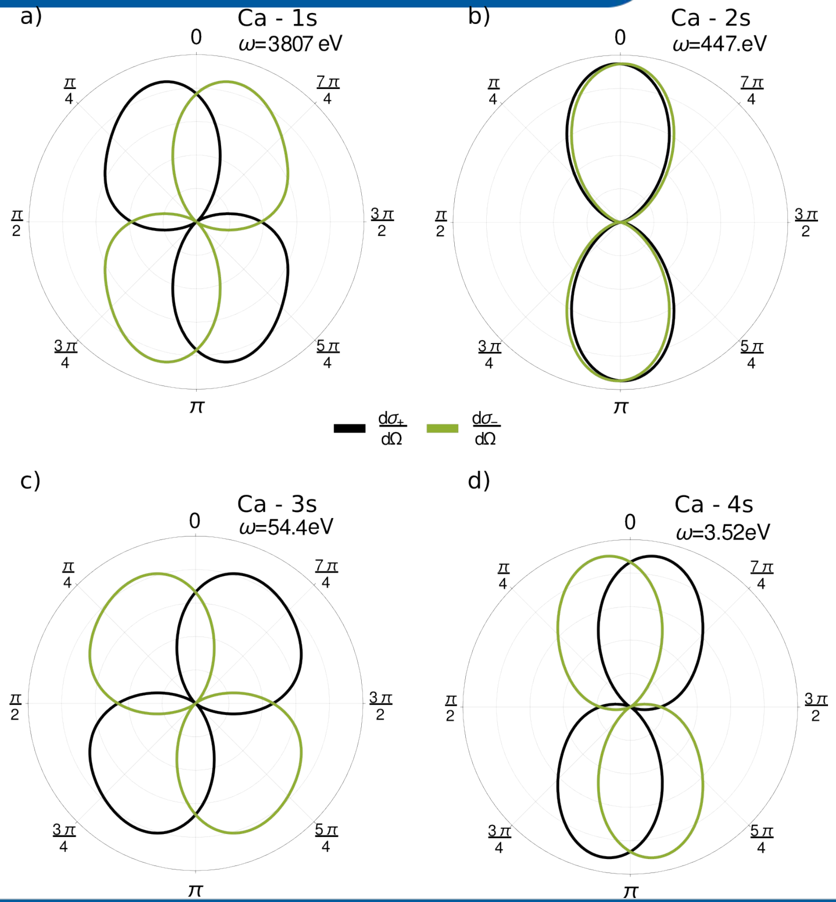
Two-photon ionization of He by linearly polarized light

Very good agreement with existing experiment




R. Ma, et al., J. Phys. B **46** , 164018 (2013).

# Phase importance




Phase difference of π/2 leads to minimal overlap of the distributions, see c)

Phase difference of ~0 leads to maximal overlap of the distributions, see b)