Recent Progress in the Field of Polarization, Alignment, and Orientation in Atomic Collisions

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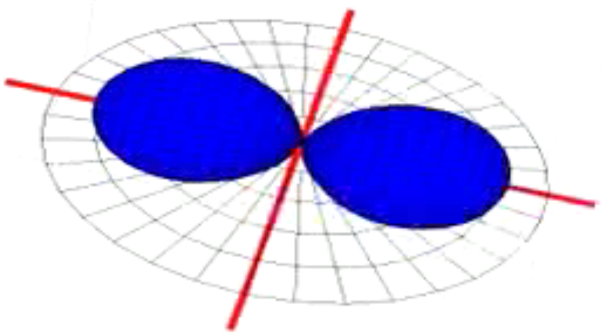
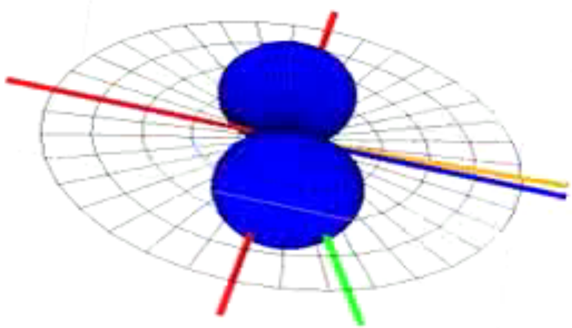




Work supported by the NSF, Univ. of Copenhagen, San Cataldo, the Danish Embassy in Paris, and the Ib Henriksen Foundation.

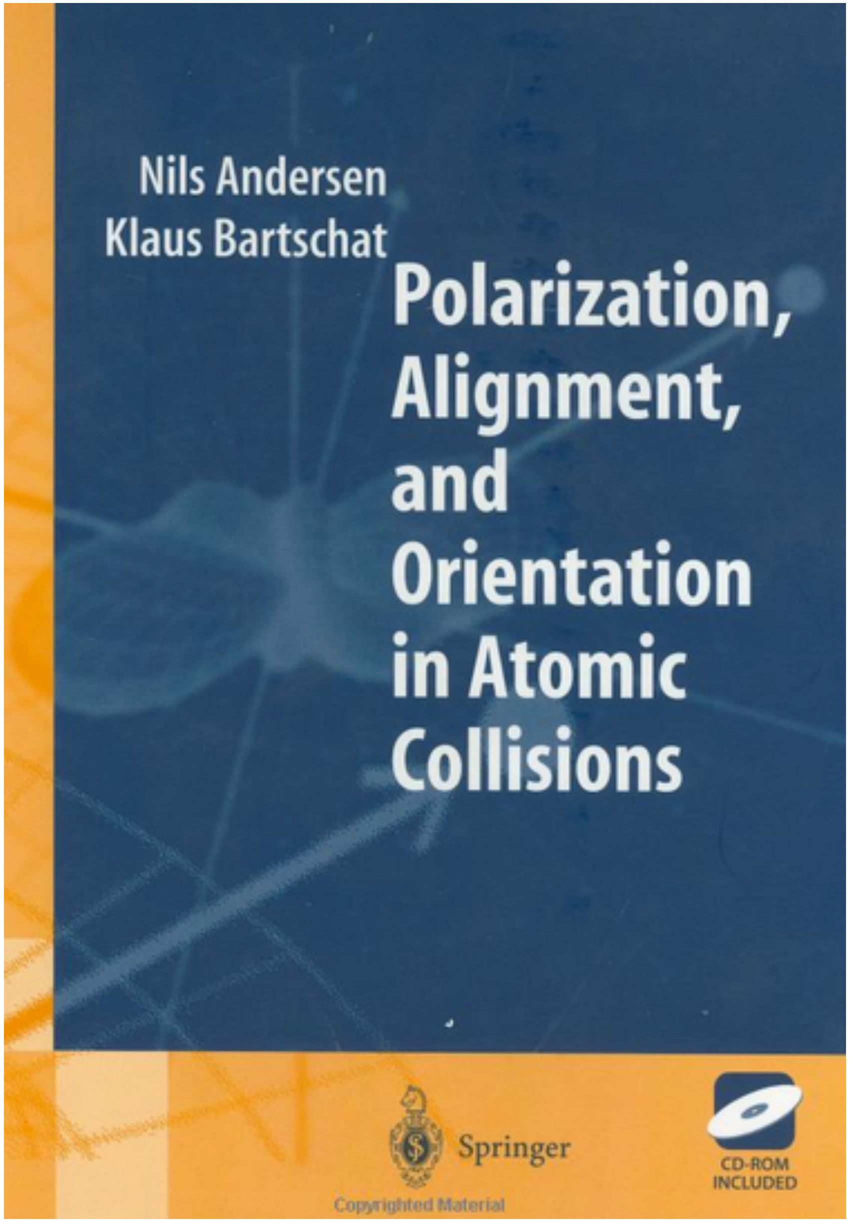
XSEDE

Extreme Science and Engineering Discovery Environment

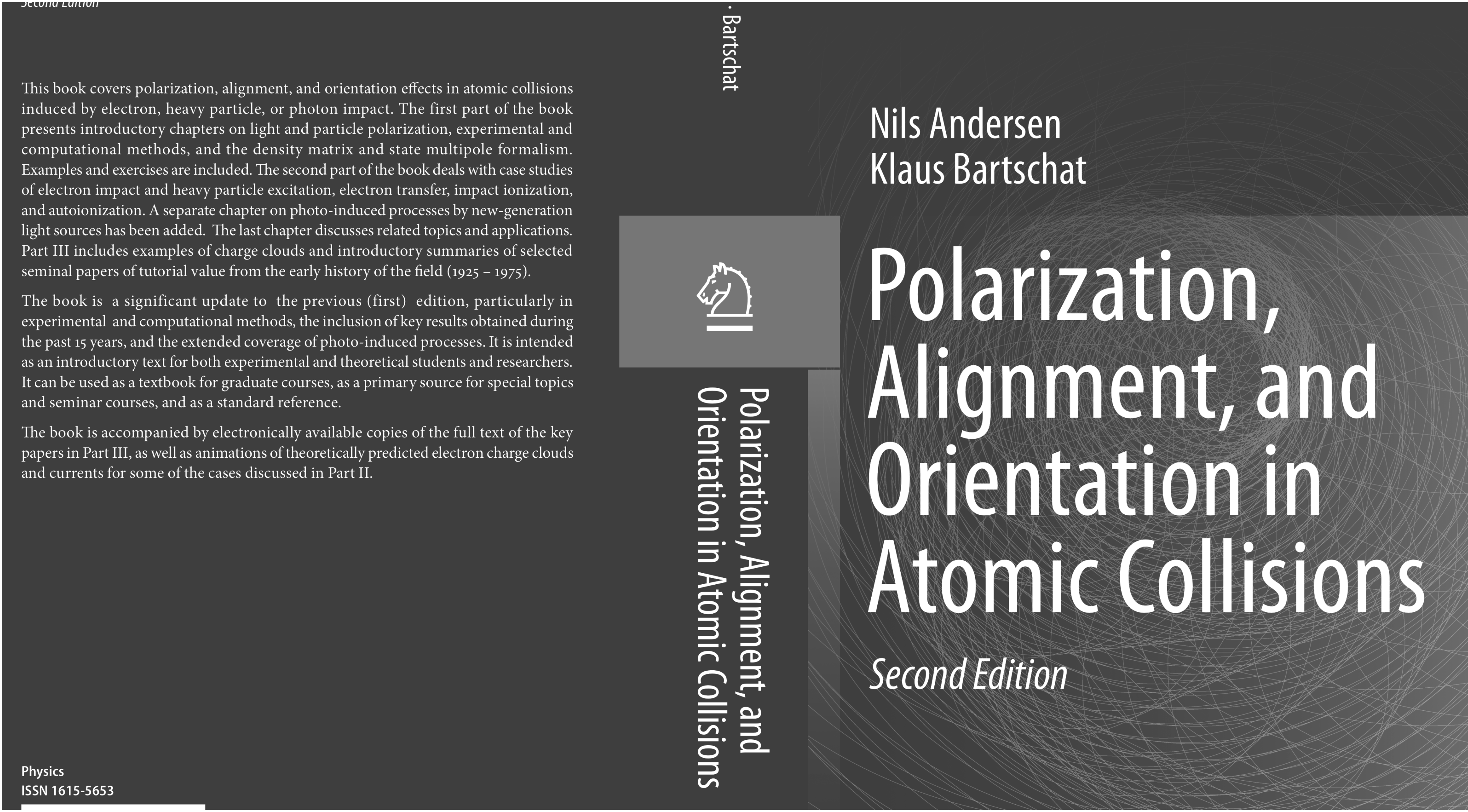
OVERVIEW (there is no hope for a comprehensive review – sorry!)

- Introduction: Motivation and Brief Review
- Update: 2001 –> Today
  - Experimental & Theoretical/Computational Progress
  - Electron Impact (Atoms, Molecules)
  - Heavy-Particle Impact
  - Photon Impact
- One Example of Possible New Directions
- Summary

Hopefully Already in Your Library



Hopefully Coming Soon to Your (E)-Library ....



This book covers polarization, alignment, and orientation effects in atomic collisions induced by electron, heavy particle, or photon impact. The first part of the book presents introductory chapters on light and particle polarization, experimental and computational methods, and the density matrix and state multipole formalism. Examples and exercises are included. The second part of the book deals with case studies of electron impact and heavy particle excitation, electron transfer, impact ionization, and autoionization. A separate chapter on photo-induced processes by new-generation light sources has been added. The last chapter discusses related topics and applications. Part III includes examples of charge clouds and introductory summaries of selected seminal papers of tutorial value from the early history of the field (1925 – 1975).

The book is a significant update to the previous (first) edition, particularly in experimental and computational methods, the inclusion of key results obtained during the past 15 years, and the extended coverage of photo-induced processes. It is intended as an introductory text for both experimental and theoretical students and researchers. It can be used as a textbook for graduate courses, as a primary source for special topics and seminar courses, and as a standard reference.

The book is accompanied by electronically available copies of the full text of the key papers in Part III, as well as animations of theoretically predicted electron charge clouds and currents for some of the cases discussed in Part II.

Physics

ISSN 1615-5653

· Bartschat

Polarization, Alignment, and Orientation in Atomic Collisions

Nils Andersen
Klaus Bartschat

Polarization, Alignment, and Orientation in Atomic Collisions

Second Edition

Motivation/Review

- Polarization, Alignment and Orientation Studies of Atomic Collisions were motivated by the quest for the complete quantum mechanical information about the processes of interest. Hence they are closely related to the idea of "Complete Scattering Experiments" (Bederson).

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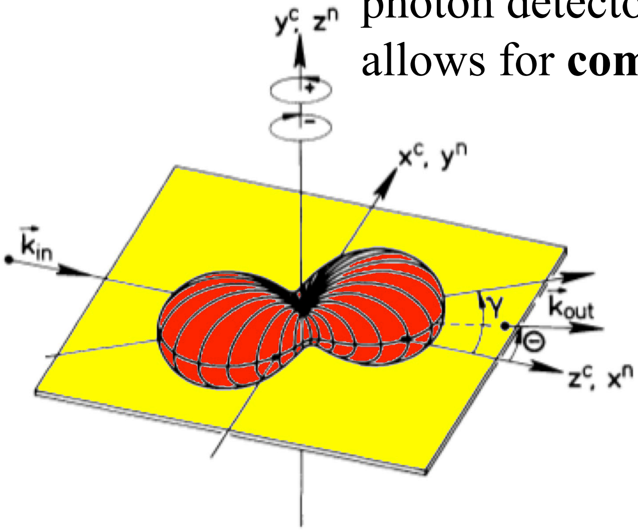
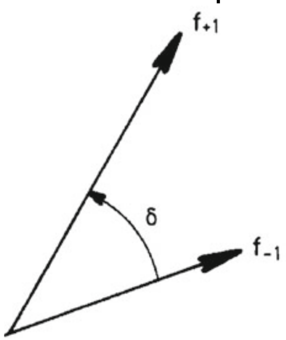
- Data from such experiments often serve as benchmarks for the development of theoretical/computational methods. If a theory can reproduce all the details, it can reasonably expected to predict quantities of high(er) interest for applications (e.g., total cross sections for modeling of plasmas and stars) correctly as well.

He 2^1P excitation: Problem solved in the mid 1990s

photon detector with polarization analysis
allows for **complete experiment!**



length = $(1+P_{lin})/2$
width = $(1-P_{lin})/2$
height = 0; P_4 can measure it



two independent amplitudes in "natural frame"; in collision frame, f_0 and $f_1 = -f_{-1}$

angular-momentum transfer (starts positive)

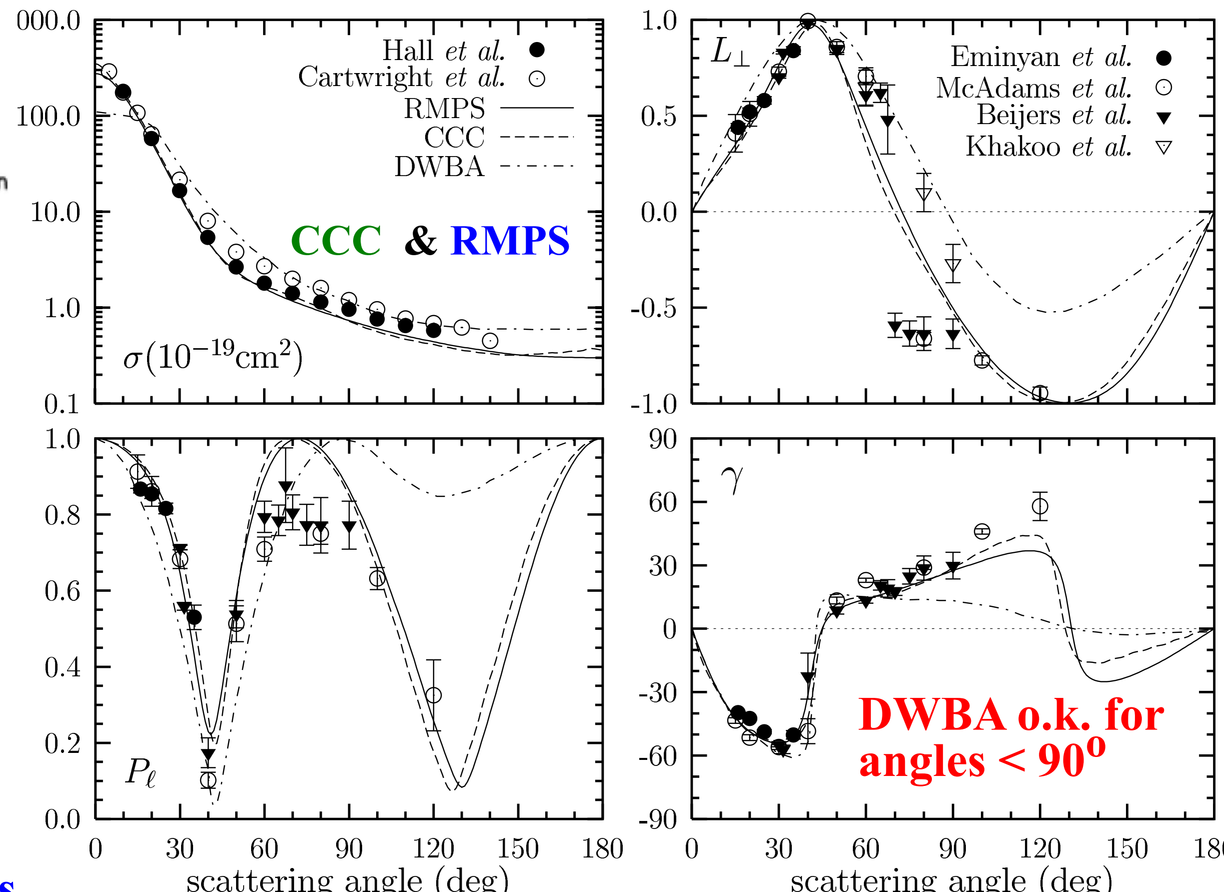


Fig. 7.18 Differential cross section (a) and electron impact coherence parameters $L_\perp$ (b), γ (c), and P_ℓ (d) for electron impact excitation of the 2^1P^o state in helium from the ground state 1^1S at an incident electron energy of 50 eV. The theoretical curves correspond to: RMPS [39], CCC [40], and DWBA [41]. The sources for the experimental data are from [42]– [46].

He 3^1D excitation: Problem solved numerically as well

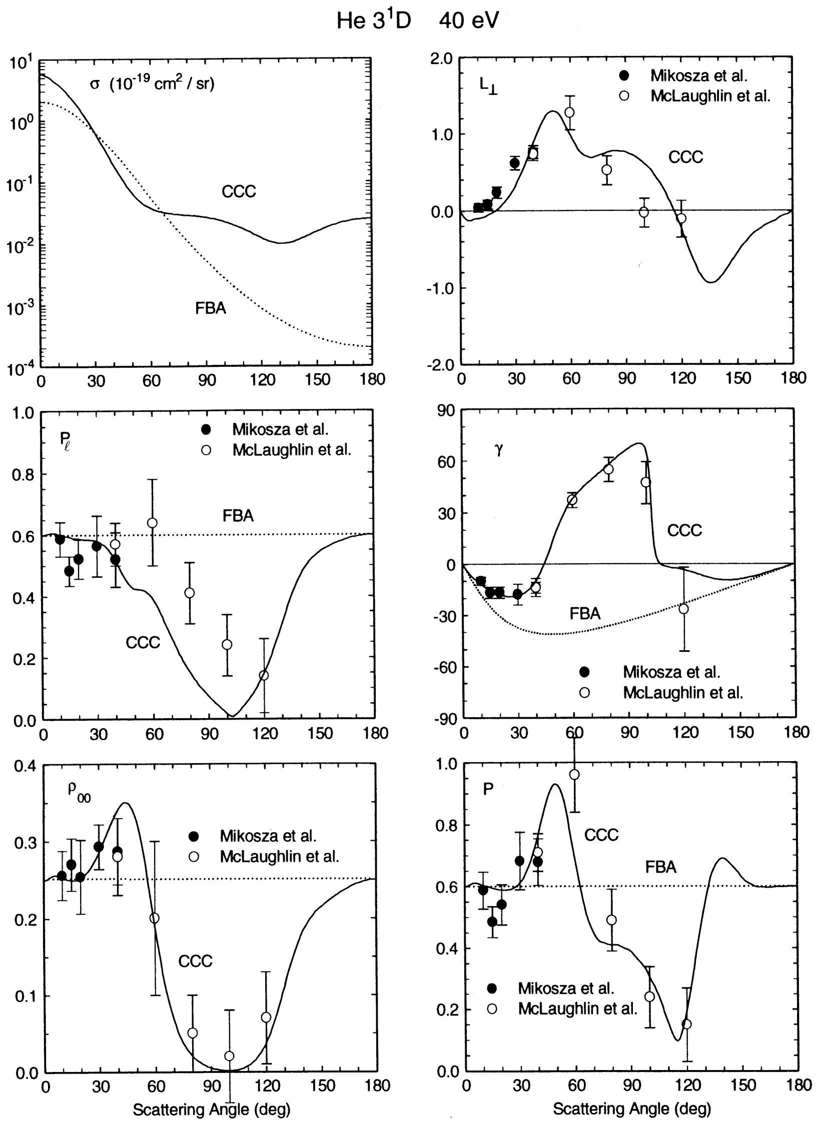
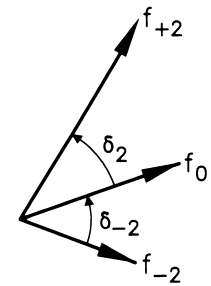
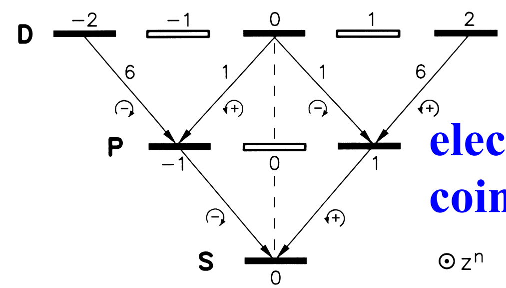


Fig. 7.24 Differential cross section σ and coherence parameters ($L_\perp, P_\ell, \gamma, \rho_{00}, P$) for electron impact excitation of the He 1^1S $\rightarrow$ 3^1D transition at an incident electron energy of 40 eV. The experimental data of Mikosza *et al.* [59] and McLaughlin *et al.* [57] are compared with CCC calculations (solid line) of Fursa and Bray [50] and with the predictions from the First Born Approximation (dotted line).

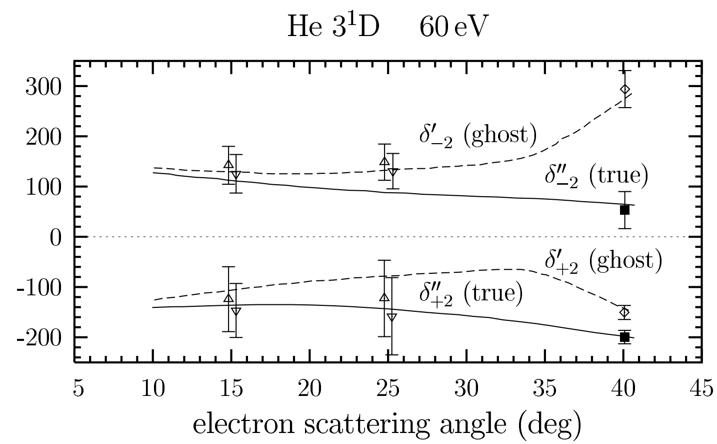
Interpretation is more difficult



three amplitudes



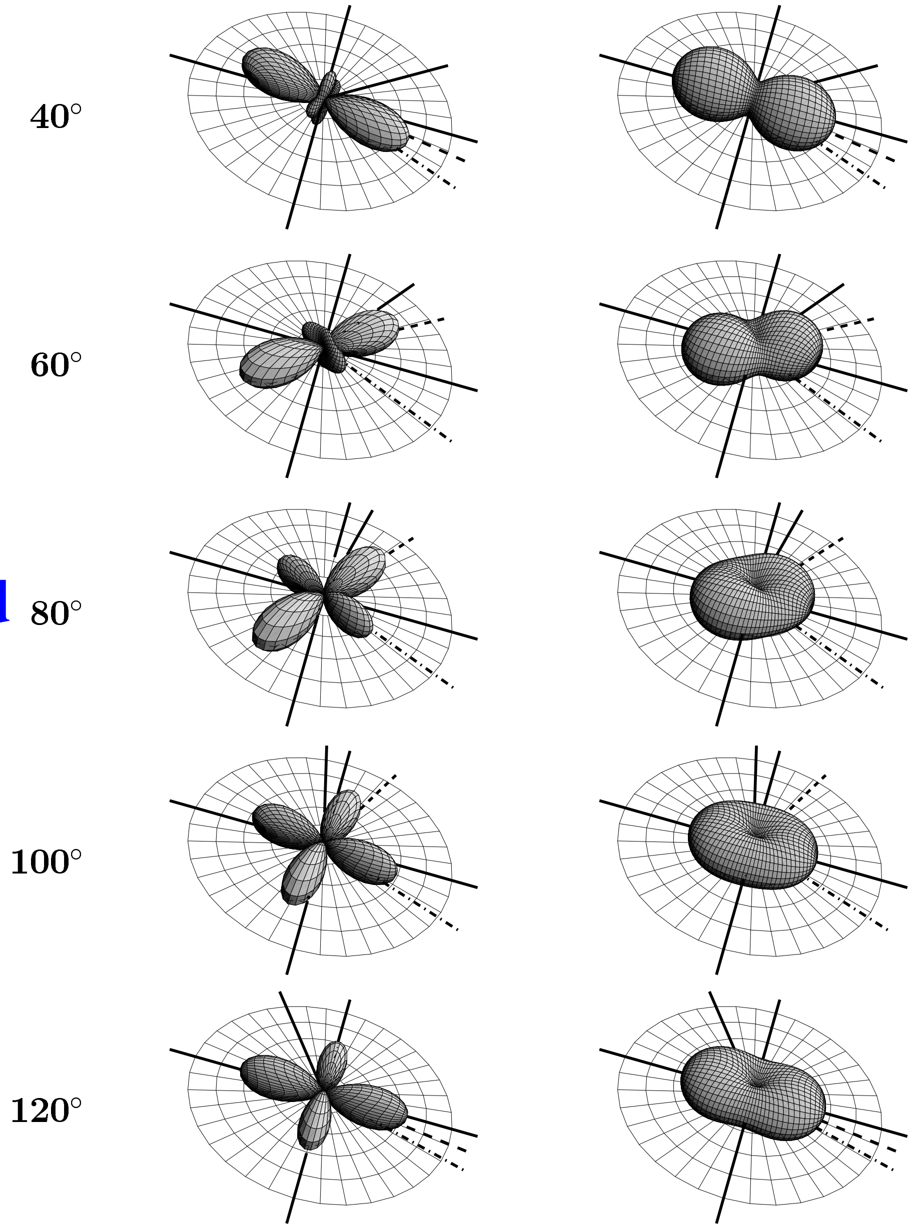
electron-(single)-photon coincidence insufficient



triple coincidence needed (done by Mikosza & Williams); need theory to distinguish true from "ghost" solution

He 3^1D excitation: 40 eV (CCC predictions)

left: actual charge cloud



right: apparent charge cloud from electron-single-photon coincidence setup

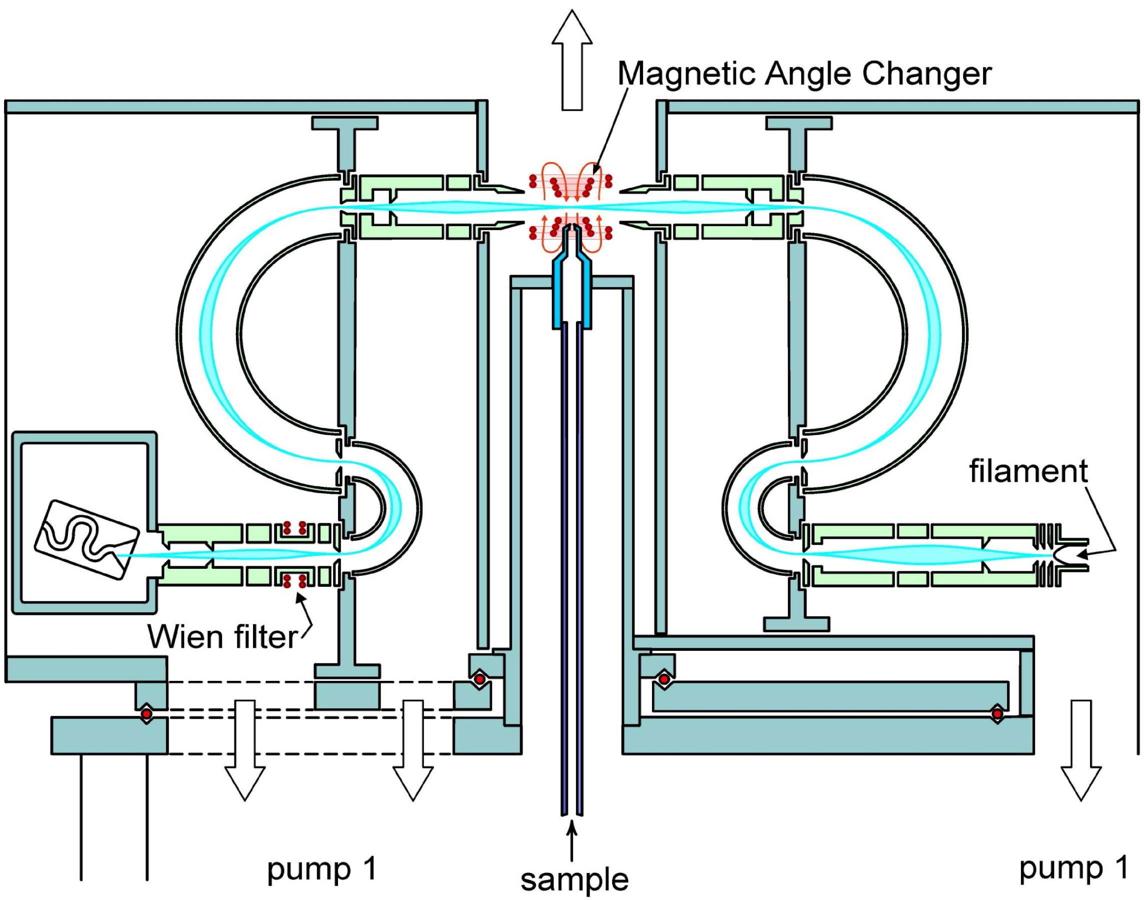
Two Examples of Experimental Progress:

Magnetic Angle Changer (MAC) and Reaction Microscope (REM)

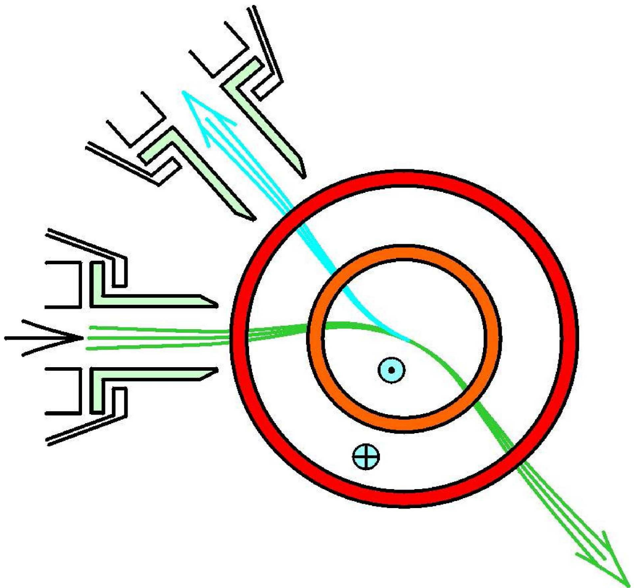
(There are many more, e.g., FELs, HHG, ...)

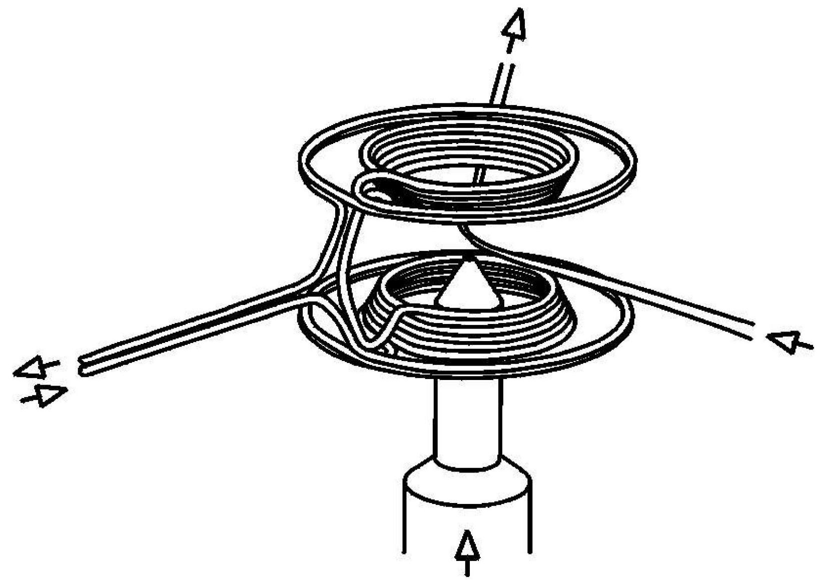
Magnetic Angle Changer (MAC)

Michael Allan's **high-resolution spectrometer to measure:**
- **specific angles**
- **specific transitions (energy selection)**



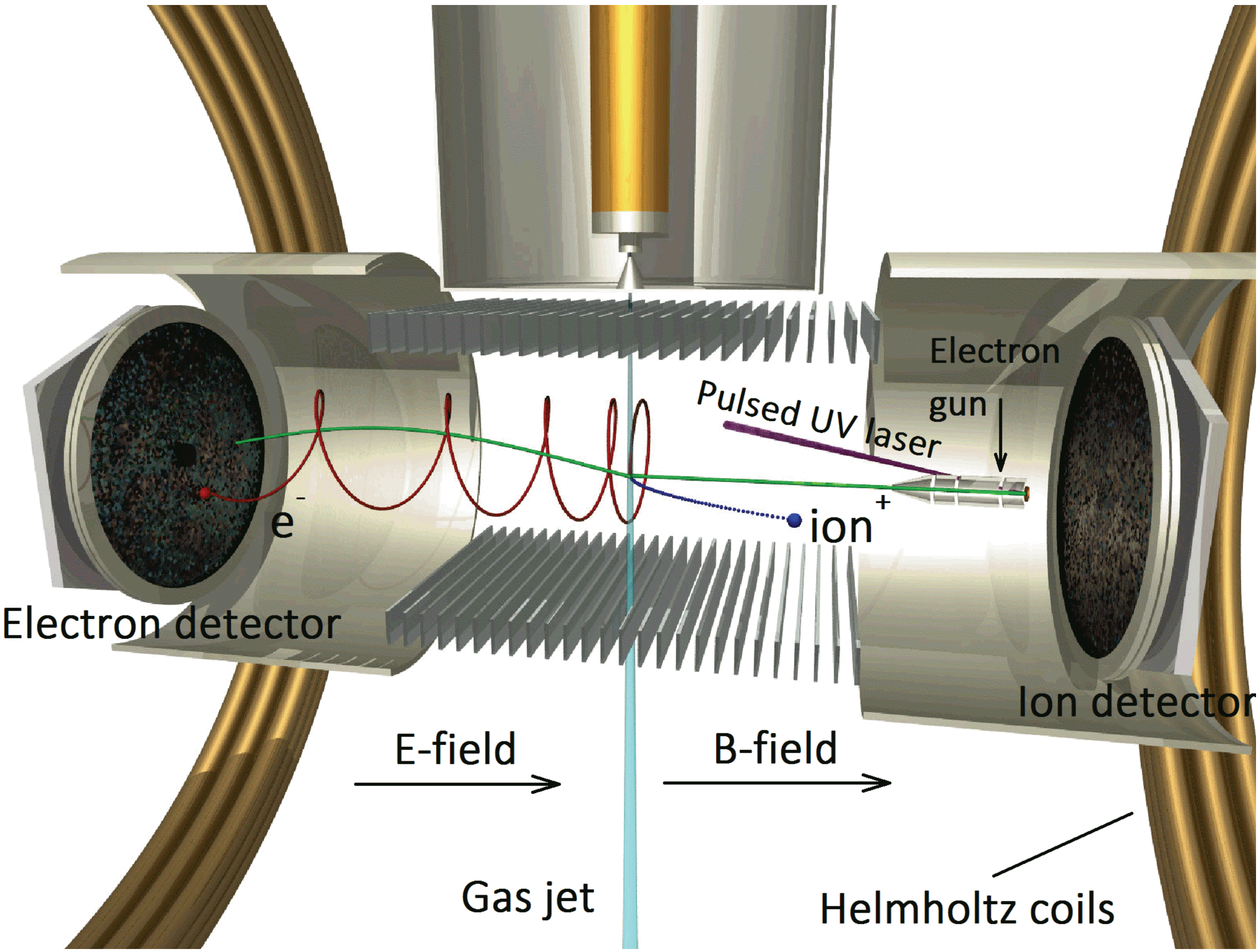
The "Magnetic Angle Changer" (MAC), developed by Reid and Channing, makes it possible to measure the full angular range, including 180º and other angles for which stainless steel may be in the way.





The Reaction Microscope

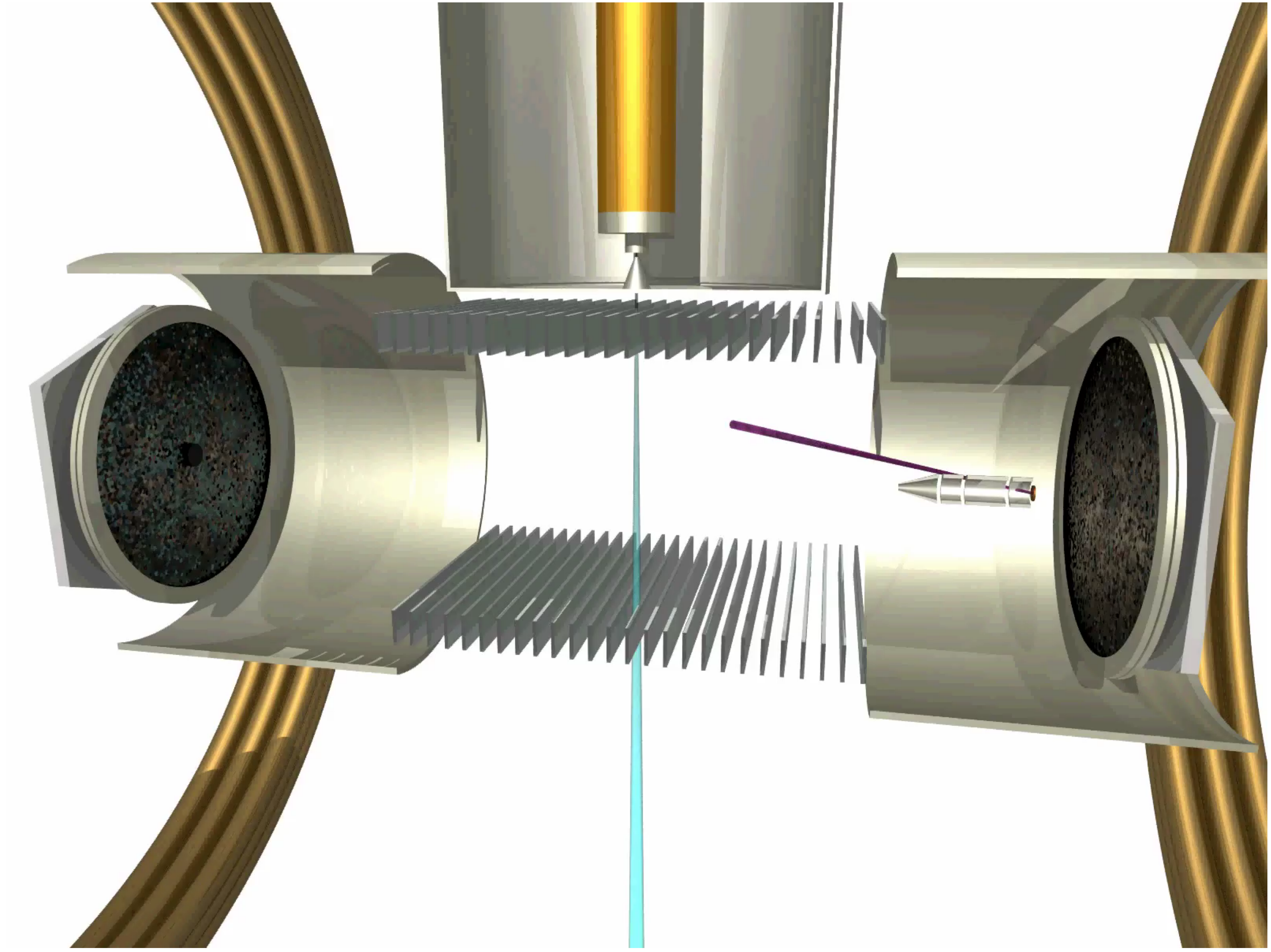
Ullrich, Moshammer, Dorn, Schmidt, Cocke, Schmidt-Böcking, (+ Cocke), Rep. Prog. Phys. 66 (2003) 1463



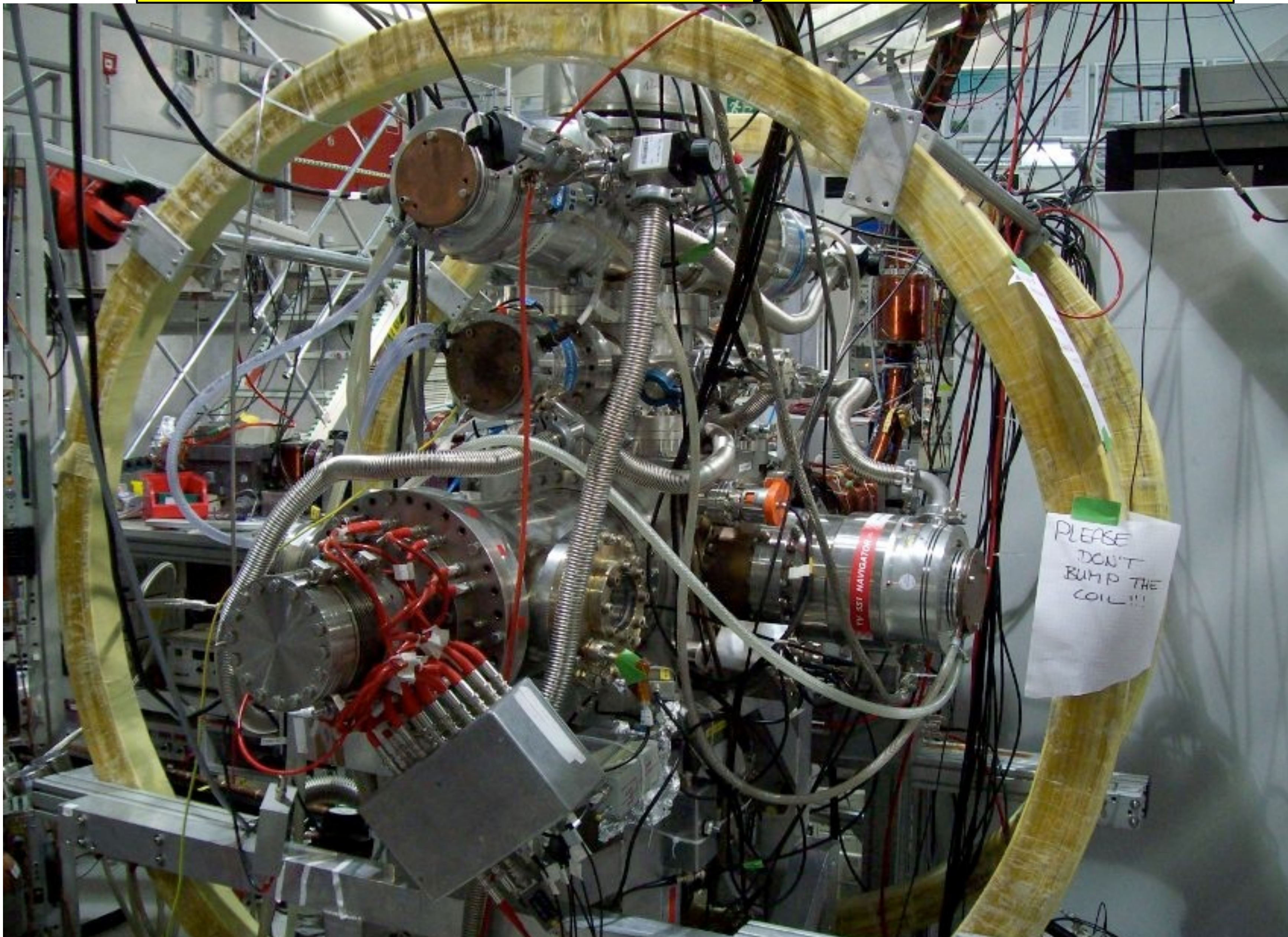
Used in A. Dorn's group for (e,2e) and even (e,2eγ)

They can get the full 3D-picture in a single shot!

This is how it works ...



This is how it really looks like!



Examples of Theoretical/Computational Progress

- **Theory:** Some of formulations have been extended (e.g., P – P transitions, advanced angular momentum gymnastics, collisions in fields, ...)

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- **Computation:** The enormous increase of computational power has led to the development of methods that can solve the Schrödinger (even Dirac) Equation with high accuracy for "simple systems" (H, He, quasi-one and quasi-two electron targets). They include:
  - Convergent close-coupling (**CCC**)
  - R-matrix with pseudostates (**RMPS**)
  - Exterior complex scaling (**ECS**)
  - Time-dependent close-coupling (**TDCC**, also for heavy-particle impact)

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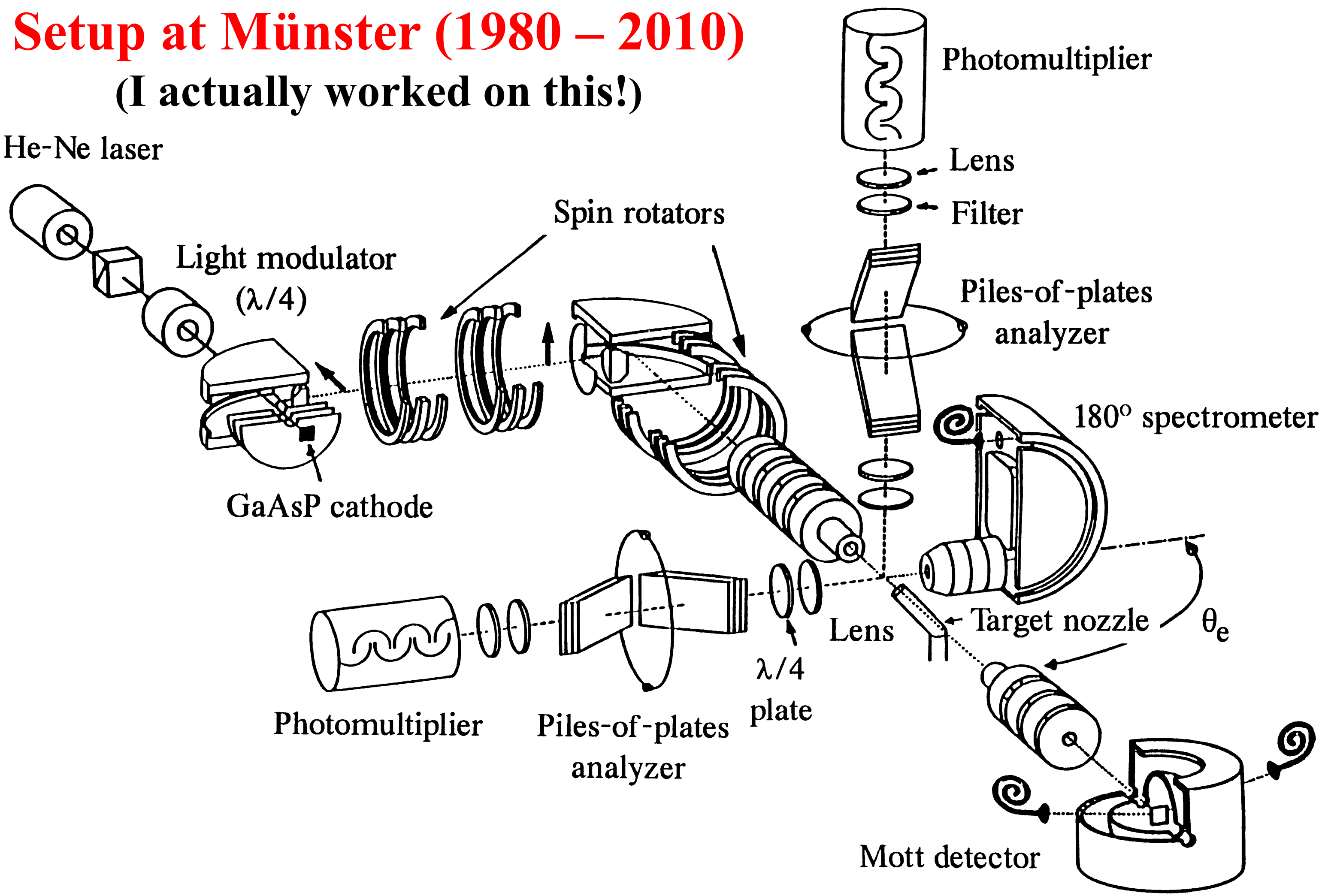
  - Convergent close-coupling (**CCC**)
  - R-matrix with pseudostates (**RMPS**)
  - Exterior complex scaling (**ECS**)
  - Time-dependent close-coupling (**TDCC**, also for heavy-particle impact)
- For more complex targets and processes, such as the heavy noble gases, ionization with excitation, fully-differential ionization, molecular targets, ...
  - B-spline R-matrix (**BSR**) has had significant success
  - DWBA with proper 3-body Coulomb boundary condition (**3C, 3DW, M3DW**, ...)

Selected Examples for Electron Impact:

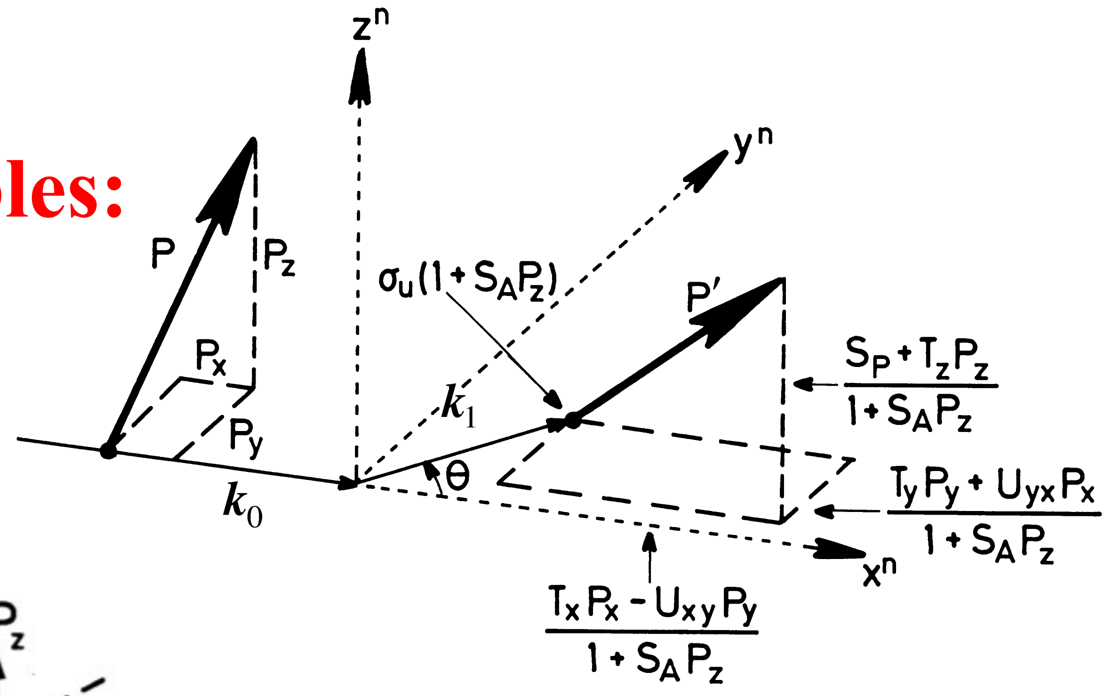
Spin Polarization, Propensities, P_2 Controversy, Atoms & Molecules

Setup at Münster (1980 – 2010)

(I actually worked on this!)

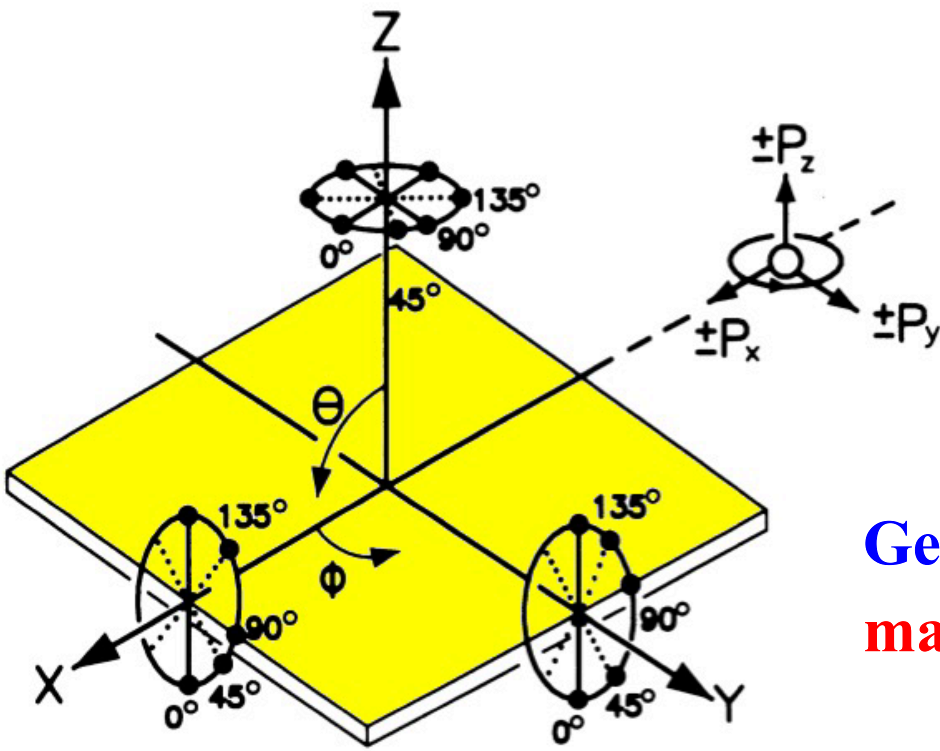


Generalized STU Parameters



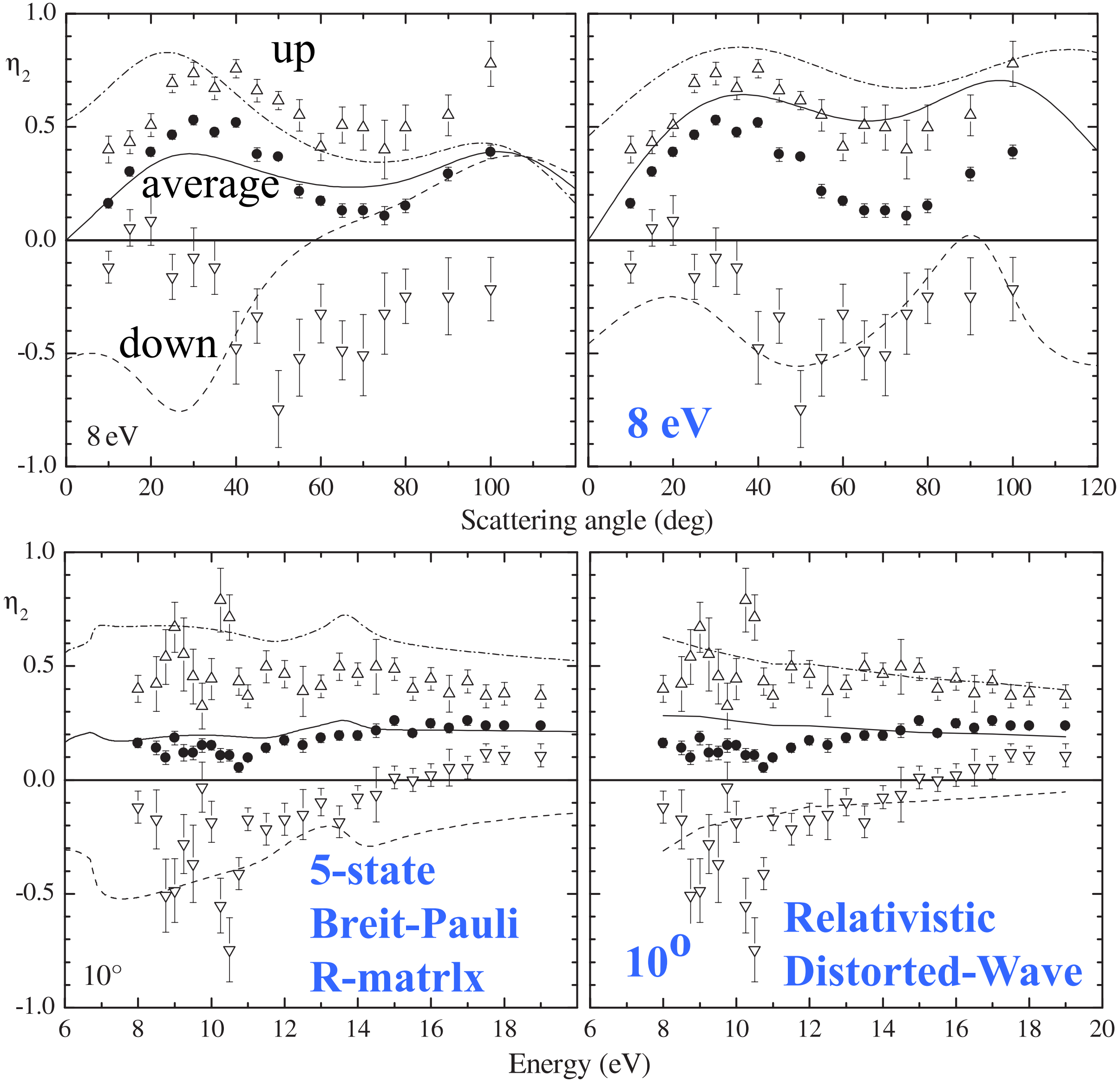
Two Sets of Observables:

P_1: 0°/90° linear
P_2: 45°/135° linear
$P_3 = -\eta_2$: left/right circular



Generalized Stokes Parameter matrix with elements $Q_{ij}(\hat{\mathbf{n}})$.

Spin-Dependent Angular Momentum Transfer and Propensities ($\eta_2 = -P_3 = L_{perp}$)

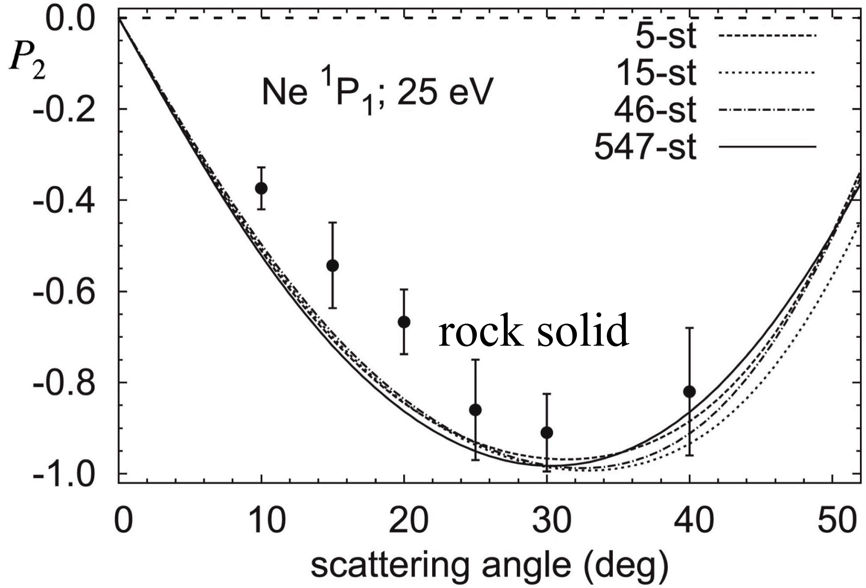


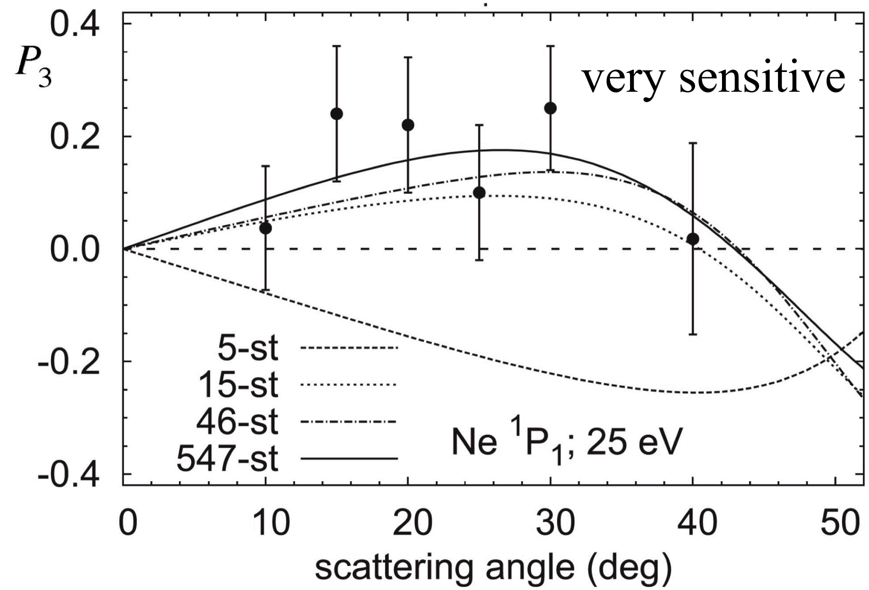
Experiment: Hg $^3P_1$
Herting *et al.*, 2002

The average value is positive at small angles.

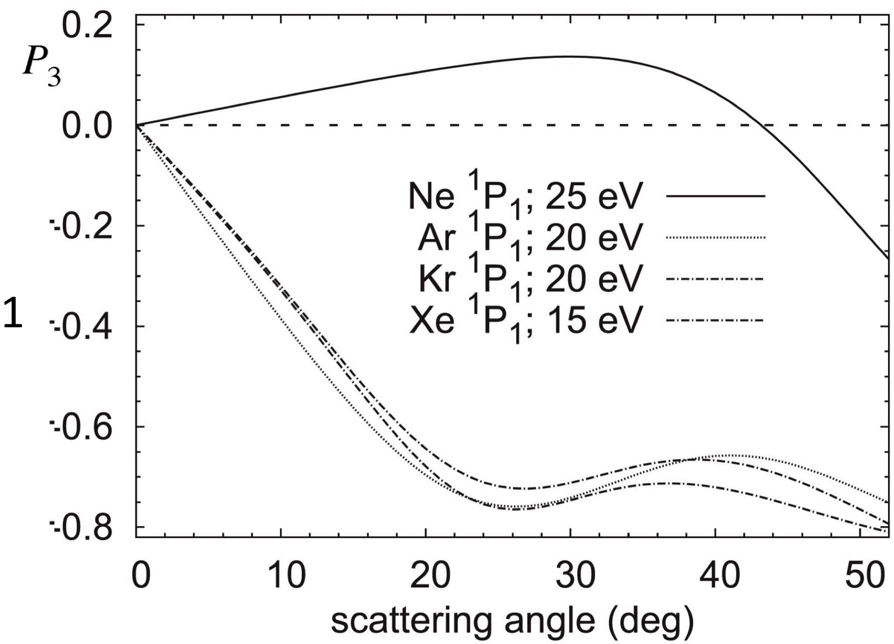
Many attempts have been made to explain this propensity, with varying levels of success.

Propensities are just that ...





Experiment: Hargreaves, Khakoo *et al.*
Phys. Rev. A **85** (2012) 050701
Theory: BSR



e-Ne at 25 eV
is an exception.
Why???

The P_2 Controversy

PHYSICAL REVIEW A **85**, 022701 (2012)

Topological angular momentum in electron exchange excitation of a single atom

J. F. Williams, L. Pravica, and S. N. Samarin

ARC Centre of Excellence for Antimatter and Matter Studies Centre for Atomic, Molecular and Surface Physics (CAMSP), School of Physics, M013, University of Western Australia, Perth 6009, Australia

(Received 15 July 2011; published 6 February 2012)

In a single free two-valence-electron atom, the motion of the electron spin is a consequence of quantum statistics and the Pauli exclusion principle. Subsequently, during an electron impact exchange excitation from a ${}^1S_0\, M_S = 0$ to a ${}^3S_1\, M_S = 0$ state, the electron spin is "parallel transported" around a closed path with a geometrical Berry phase of π radians creating an aligned exchange spin angular momentum. This alignment is observed via the Stokes parameter P_2 of the photon decay into a ${}^3P$ state. The geometric phase is in addition to the dynamic phase. Measurements from zinc and mercury atoms in different laboratories show the effect close to the excitation threshold where there are no competing excitation processes. Similar effects are expected in other atomic and molecular quantum scattering processes where comparable geometrical or topological paths exist. Electron quantum scattering theories use antisymmetrized wave functions but none include this geometrical exchange angular momentum.

DOI: 10.1103/PhysRevA.85.022701 PACS number(s): 34.80.Dp

"The task remains for theory to include a topological nondynamical phase."

Theorists did not agree (and still don't!)

PHYSICAL REVIEW A **87**, 016701 (2013)

Comment I on "Topological angular momentum in electron exchange excitation of a single atom"

Christopher J. Bostock,* Dmitry V. Fursa, and Igor Bray
ARC Centre for Antimatter-Matter Studies, Curtin University, GPO Box U1987, Perth, Western Australia 6845, Australia
(Received 4 April 2012; published 9 January 2013)

In their recent paper, Williams *et al.* [Phys. Rev. A **85**, 022701 (2012)] report on the apparatus and experimental method for the measurement of the Stokes parameter P_2 associated with spin-polarized electron impact $(3d^{10}s^2)\,{}^1S_0 \rightarrow (3d^{10}4s5s)\,{}^3S_1$ excitation of zinc. On the basis of a qualitative semiclassical argument, they make the following claim regarding the discrepancy between theory and experiment for P_2: "The task remains for theory to include a topological nondynamical phase." We analyze the validity of this assertion.

DOI: 10.1103/PhysRevA.87.016701 PACS number(s): 34.80.Dp

Last sentence: "We analyze the validity of this assertion."

Experimental Setup: (Angle)-Integrated Stokes Parameters after Impact Excitation by Spin-Polarized Electrons

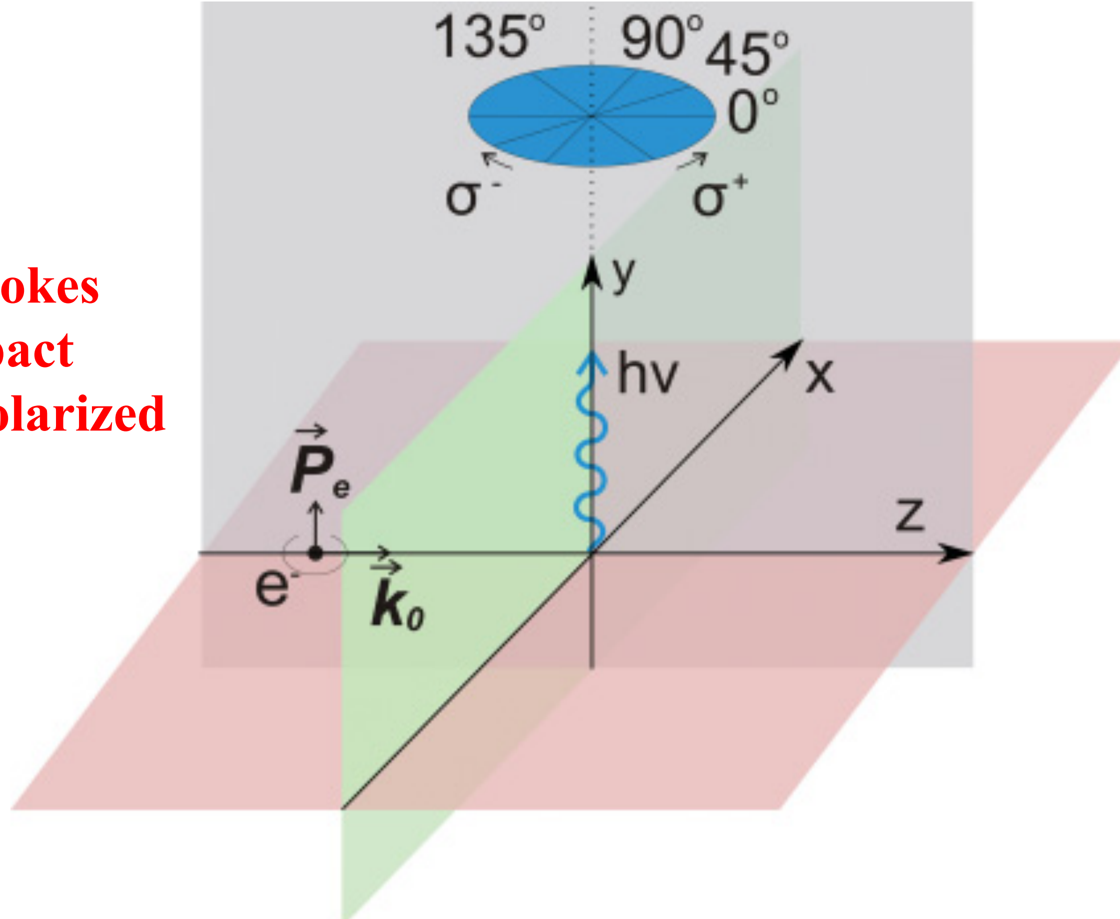


FIG. 1. (Color online) The geometrical (xyz) reference frame and scattering geometry. The spin $\boldsymbol{P_e}$ momentum $\boldsymbol{k_0}$ vectors of the incident electron beam define the scattering (yz) planar symmetry with the target atoms at the origin. Photons emitted along the y axis are analyzed with wavelength filters and linear and circular polarizers before detection with a photomultiplier.

e + Zn ($4s^2$) —> e + Zn ($4s5s$)$^3S_1$ —> e + Zn ($4s4p$)$^3P_{0,1,2}$ + hν

Farago & Wykes (1969); Wykes (1971) suggested optical P_e measurement

Eminyan & Lampel (1980): P_3 = factor(J_f) x P_e (confirmed experimentally)

K.B. & K. Blum (1982): $P_1 = P_2 = 0$ (independent of P_e for this transition)

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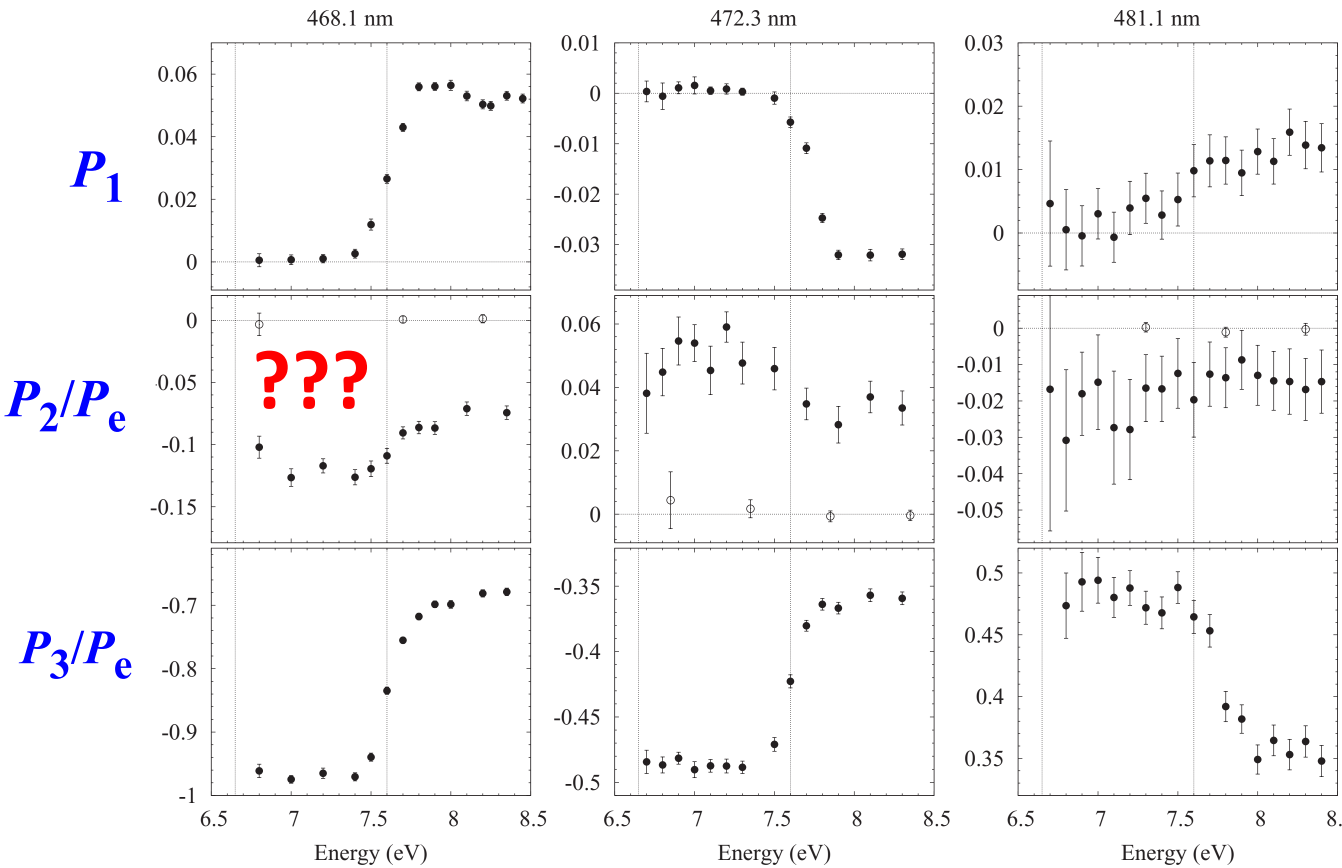
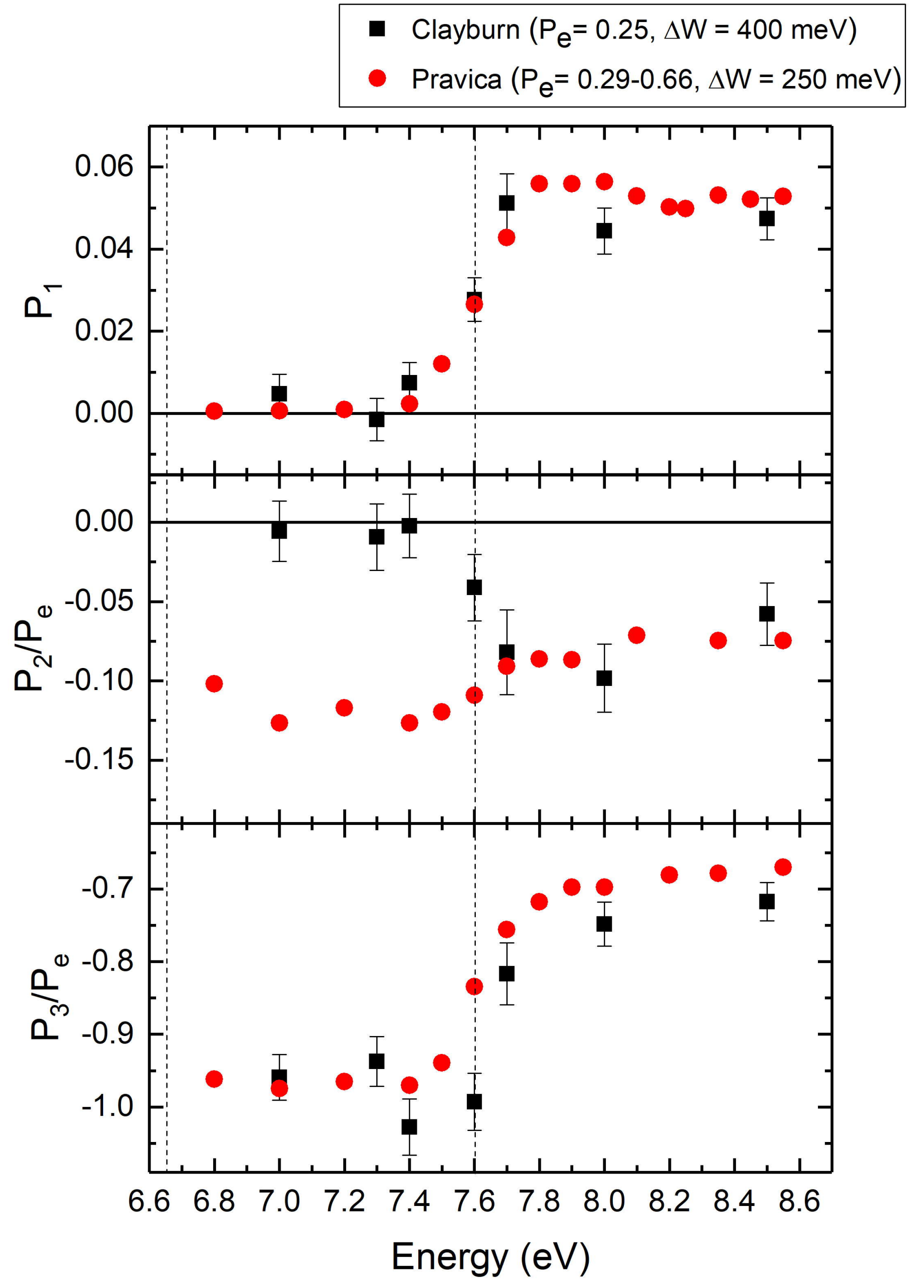


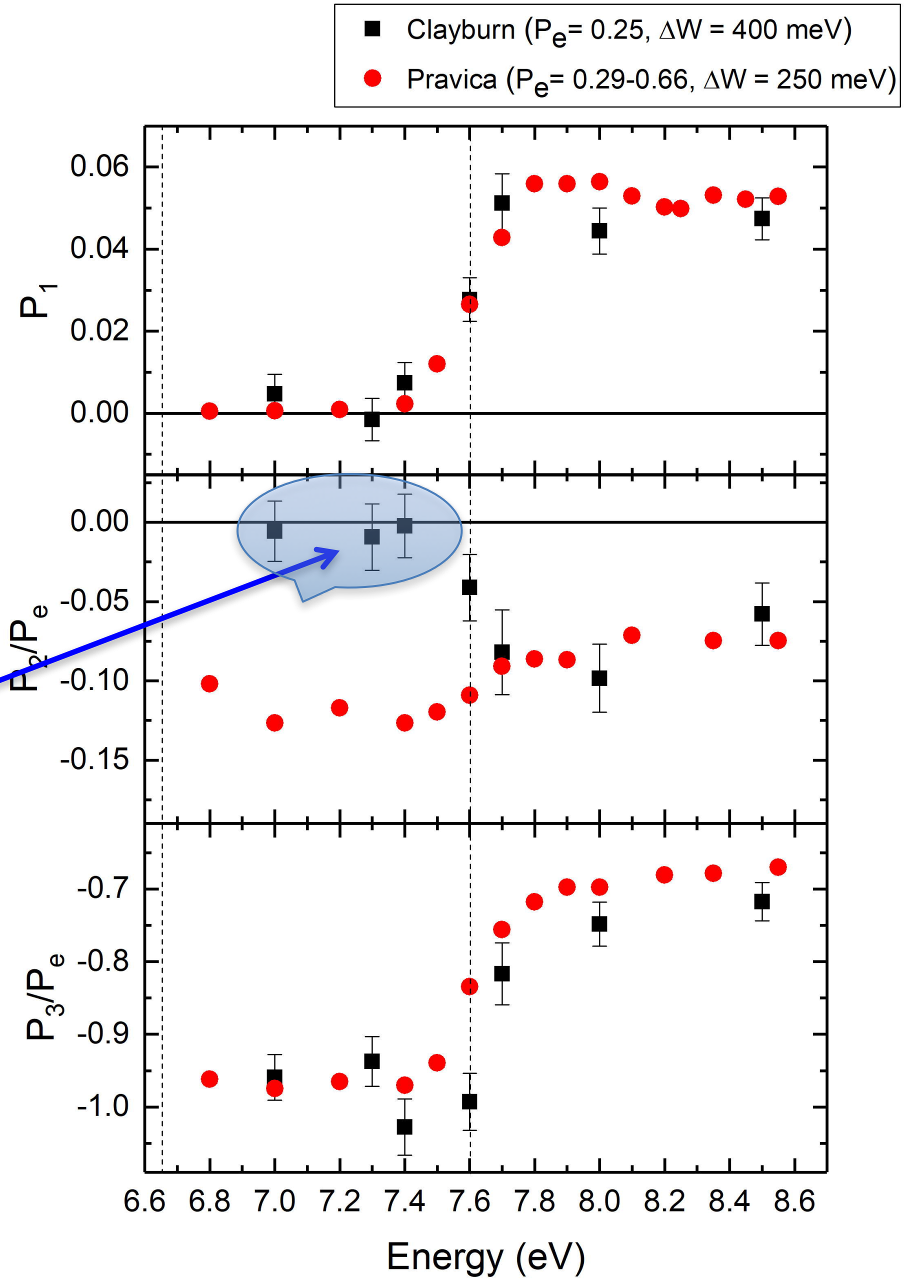
FIG. 2. The integrated Stokes parameters $P_{i=1,2,3}$ for zinc atoms excited from the ground $4s^1S_0$ state to the $5s^3S_1$ state and observed by the subsequent radiative decay to the $4p\,^3P_{0,1,2}$ states with photon wavelengths for $J = 0,1,2$ of 468.1, 472.3, and 481.1 nm, respectively. The data were normalized to an electron beam polarization which varied for different measurements but was normally of the order of 66 ± 0.5%. The threshold excitation energy for the $4s5s\ ^3S_1$ state is 6.65 eV and for the first cascading $5p\ ^3P$ state at 7.6 eV, as shown by the vertical lines. The open circles indicate measurements using unpolarized electrons and the closed circles using polarized incident electrons and normalized to the average incident spin P_e..

The latest development (Clayburn and Gay, ICPEAC 2017)



More in T.J. Gay's talk.

The latest development (Clayburn and Gay, ICPEAC 2017)



I like these three points a lot! ☺

Comment II on "Topological angular momentum in electron exchange excitation of a single atom"

Klaus Bartschat and Oleg Zatsarinny
Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA
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A recent article by Williams *et al.* [Phys. Rev. A **85**, 022701 (2012)] highlights a discrepancy between experiment and theory for the linear light polarization P_2 measured after impact excitation of zinc atoms by a spin-polarized electron beam. The claim is made that current collision theories must be modified by including a geometric (Berry) phase in the calculations in order to reproduce the experimental data for Zn and similar data from the Münster group for Hg. We show that the e-Hg data can be qualitatively reproduced by our fully relativistic B-spline R-matrix approach *without* any further modification.

DOI: 10.1103/PhysRevA.87.016702 PACS number(s): 34.80.Dp

BSR gets nonzero P_2/P_e for Hg, but not for Zn.

A serious discrepancy between experimental data and theoretical predictions was recently reported [1] for spin-polarized electron-impact excitation of the $(4s5s)\,^3S_1$ state in Zn atoms. The linear light polarization P_2, measured for optical decays to the $(4s4p)\,^3P_{0,1,2}$ states with a photon detector aligned along the direction of the spin polarization P_e of the incident electron beam, was found to be significantly (nearly 10% for the final state $^3P_0$) different from zero, whereas, all available numerical calculations predicted an effect of less than 0.01% in the cascade-free region just above the excitation threshold. In 1982, Bartschat and Blum [2] predicted a zero

The experimental data from the Münster group (Goeke, Wolcke, Hanne, Keßler) were never published! Why???

interaction with terms of different L's and S's is negligible. The $(4s5s)\,^3S_1$ state in Zn seems to be a very good candidate for such a case, and Zn is sufficiently light that spin-orbit effects during the excitation process are likely small. Hence,

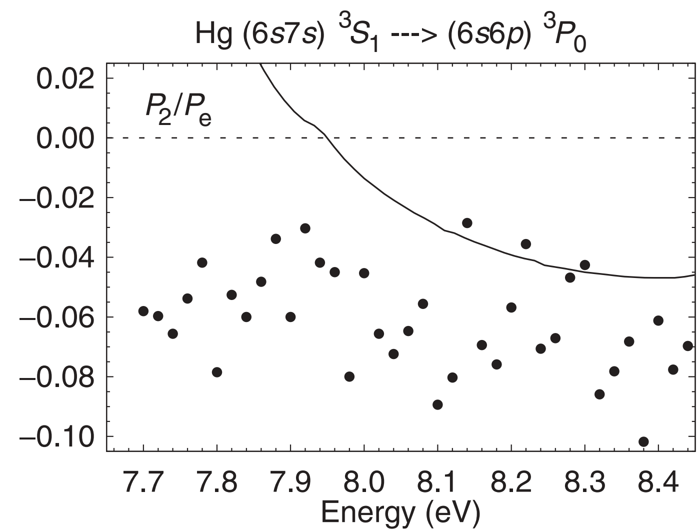

FIG. 1. P_2/P_e for spin-polarized electron-impact excitation of the $(6s7s)\,^3S_1$ state in Hg with subsequent optical decay to the $(6s6p)\,^3P_0$ state. The experimental data of Goeke [5] are compared with the DBSR prediction based on the model described in Ref. [6].

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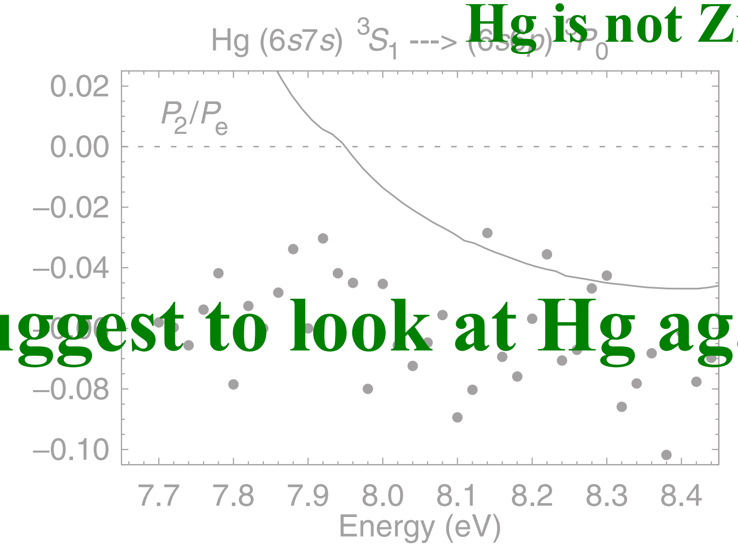
Hg is not Zn!



I suggest to look at Hg again!

FIG. 1. P_2/P_e for spin-polarized electron-impact excitation of the $(6s7s)\,^3S_1$ state in Hg with subsequent optical decay to the $(6s6p)\,^3P_0$ state. The experimental data of Goeke [5] are compared with the DBSR prediction based on the model described in Ref. [6].

Theorists did not agree (and still don't!)

PHYSICAL REVIEW A **87**, 016701 (2013)

Comment I on "Topological angular momentum in electron exchange excitation of a single atom"

Christopher J. Bostock,* Dmitry V. Fursa, and Igor Bray
ARC Centre for Antimatter-Matter Studies, Curtin University, GPO Box U1987, Perth, Western Australia 6845, Australia
(Received 4 April 2012; published 9 January 2013)

In their recent paper, Williams *et al.* [Phys. Rev. A **85**, 022701 (2012)] report on the apparatus and experimental method for the measurement of the Stokes parameter P_2 associated with spin-polarized electron impact $(3d^{10}s^2)\,^1S_0 \rightarrow (3d^{10}4s5s)\,^3S_1$ excitation of zinc. On the basis of a qualitative semiclassical argument, they make the following claim regarding the discrepancy between theory and experiment for P_2: "The task remains for theory to include a topological nondynamical phase." We analyze the validity of this assertion.

DOI: 10.1103/PhysRevA.87.016701 PACS number(s): 34.80.Dp

Theorists' Conclusion:

A geometrical phase may be used to interpret the results from a full quantum calculation, but it won't give any new results (somewhat similar to Bohmian Mechanics).

Ionization in the Close-Coupling Formalism

- **Recall: We are interested in the ionization process**

$$e_0(\boldsymbol{k}_0, \mu_0) + A(L_0, M_0; S_0, M_{S_0}) \rightarrow e_1(\boldsymbol{k}_1, \mu_1) + e_2(\boldsymbol{k}_2, \mu_2) + A^+(L_f, M_f; S_f, M_{S_f})$$

- **We need the ionization amplitude**

$$f(L_0, M_0, S_0; \boldsymbol{k}_0 \rightarrow L_f, M_f, S_f; \boldsymbol{k}_1, \boldsymbol{k}_2)$$

- **We employ the B-spline R-matrix method of Zatsarinny (CPC 174 (2006) 273) with a large number of pseudo-states:**
  - **These pseudo-states simulate the effect of the continuum.**

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- We need the ionization amplitude

$$f(L_0, M_0, S_0; \boldsymbol{k}_0 \to L_f, M_f, S_f; \boldsymbol{k}_1, \boldsymbol{k}_2)$$

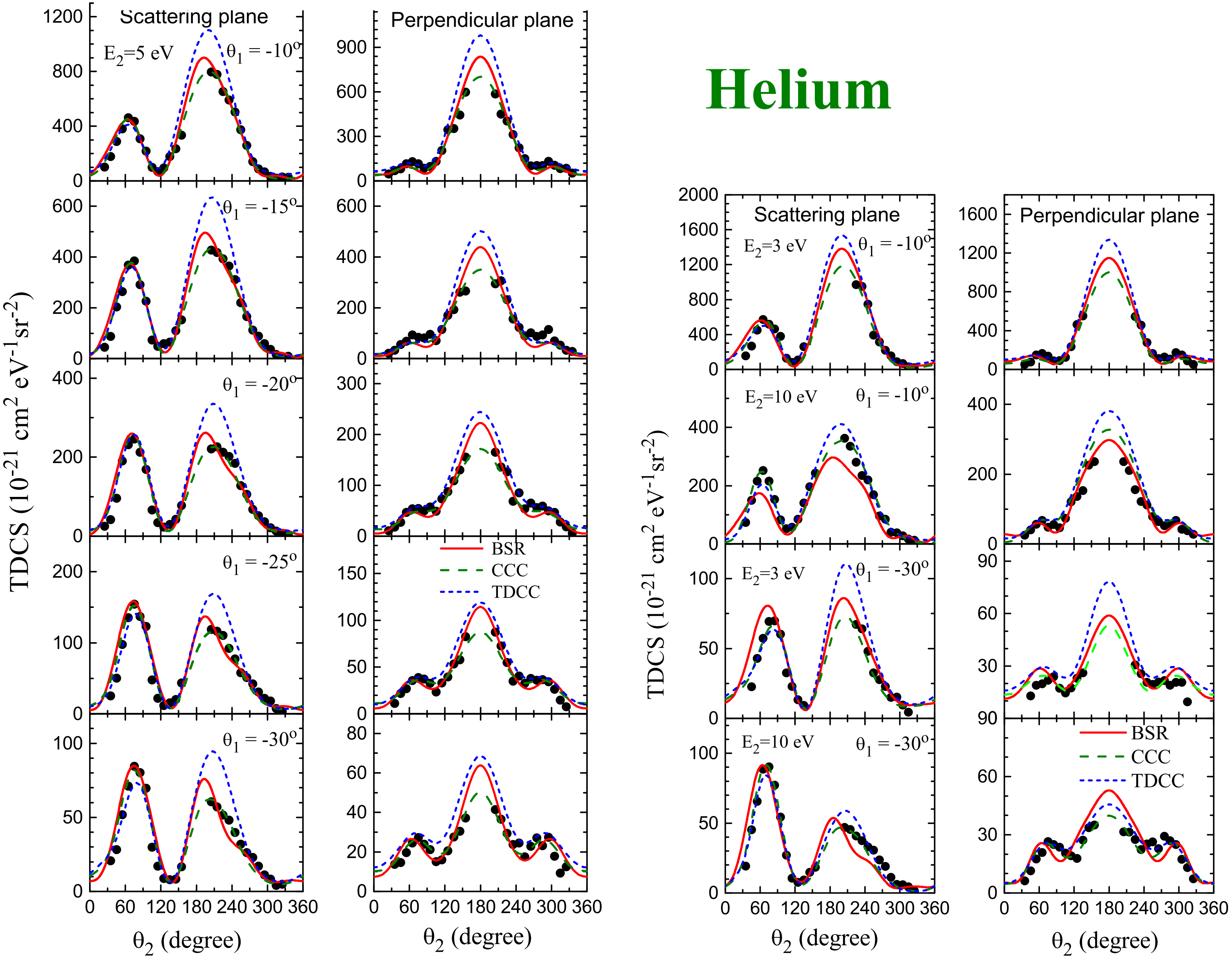
- We employ the B-spline R-matrix method of Zatsarinny (CPC 174 (2006) 273) with a large number of pseudo-states:
  - These pseudo-states simulate the effect of the continuum.
  - The scattering amplitudes for excitation of these pseudo-states are used to form the ionization amplitude: **This direct projection is the essential idea; it doesn't come from first principles, but it works.**

$$f(L_0, M_0, S_0; \boldsymbol{k}_0 \to L_f, M_f, S_f; \boldsymbol{k}_1, \boldsymbol{k}_2) = \sum_p \langle \Psi_f^{\boldsymbol{k}_2 -} | \Phi(L_p S_p) \rangle \, f(L_0, M_0, S_0; \boldsymbol{k}_0 \to L_p, M_p, S_p; \boldsymbol{k}_{1p}).$$

- Both the true continuum state $|\Psi_f^{\boldsymbol{k}_2 -}\rangle$ (with the appropriate multi-channel asymptotic boundary condition) and the pseudo-states $|\Phi(L_p S_p)\rangle$ are consistently calculated with the same close-coupling expansion.

Triple-Differential Cross Section for Direct Ionization

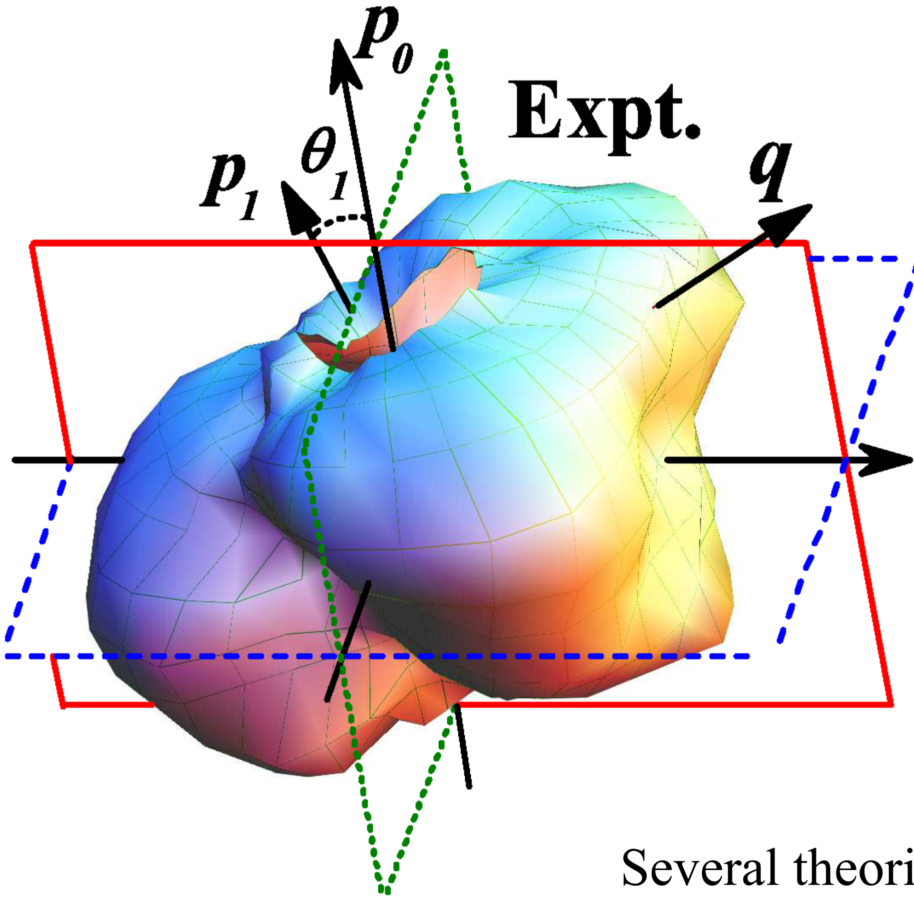
A Benchmark Test: E_0 = 195 eV; Phys. Rev. A **83** (2011) 052711

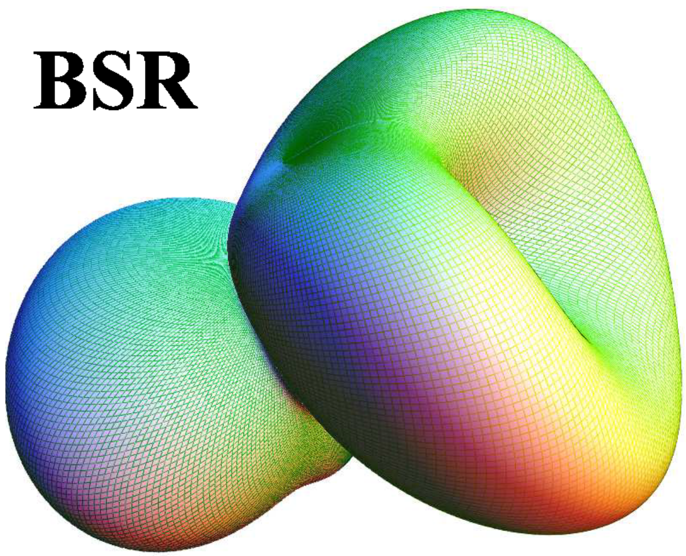


X. Ren *et al*. (Phys. Rev. A **93** (2016) 062704)

(e,2e) on Ar $(3p^6)$

E_0 = 66 eV; E_1 = 47 eV; E_2 = 3 eV; θ_1 = 15°





Several theories worked o.k. for Ne (65 eV), but only BSR reproduced the Ar data consistently well.

No More Spectators:

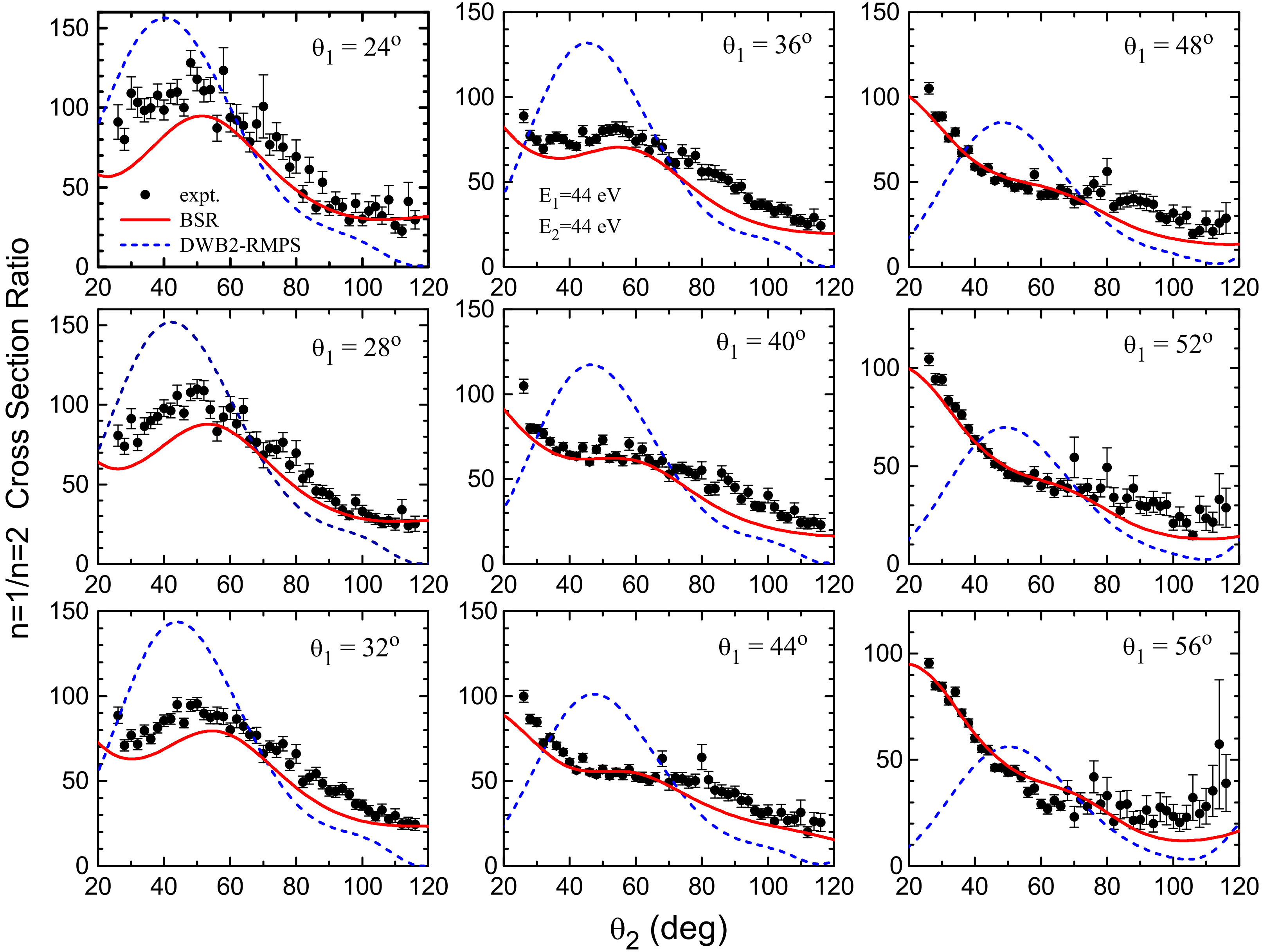
Ionization with Excitation of Helium

Three Electrons Change Their Quantum State (Movie by A. Harris)

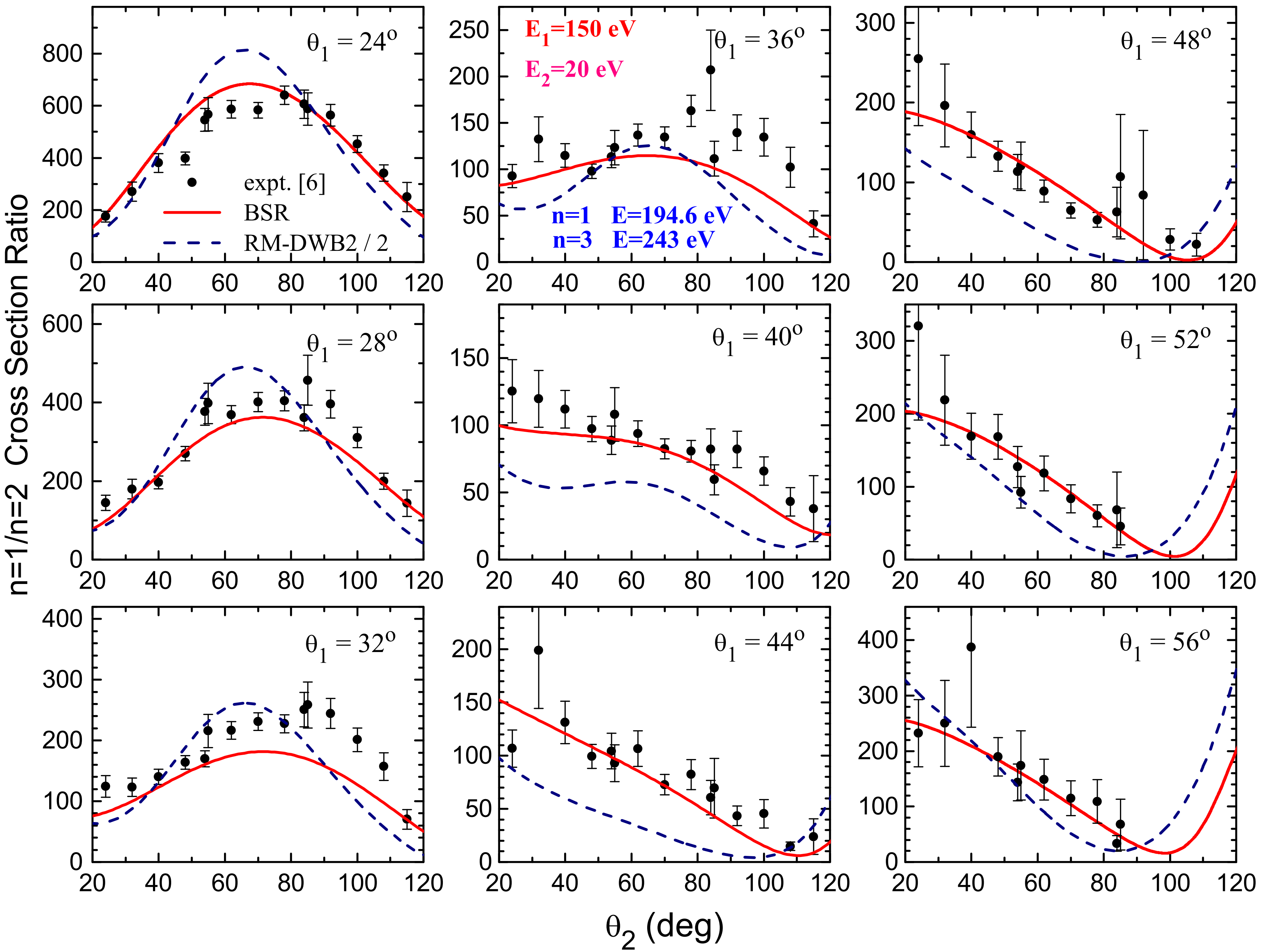
Helium (*n* = 2; symmetric energy sharing) Phys. Rev. Lett. **107** (2011) 023203

Triple-Differential Cross Section Ratio

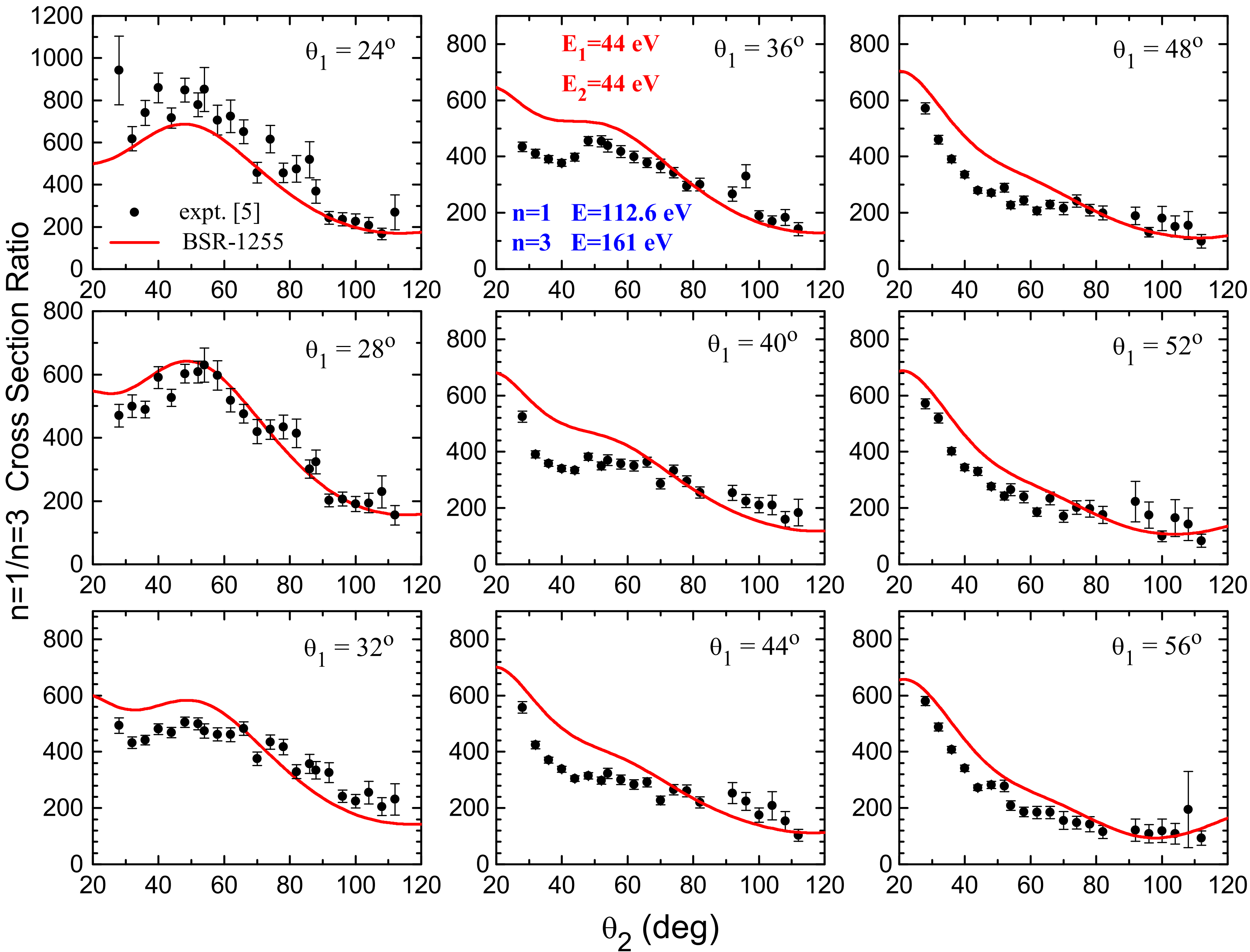
experiment: Bellm, Lower, Weigold; measured directly



Helium (*n* = 2; asymmetric energy sharing) Phys. Rev. Lett. **107** (2011) 023203

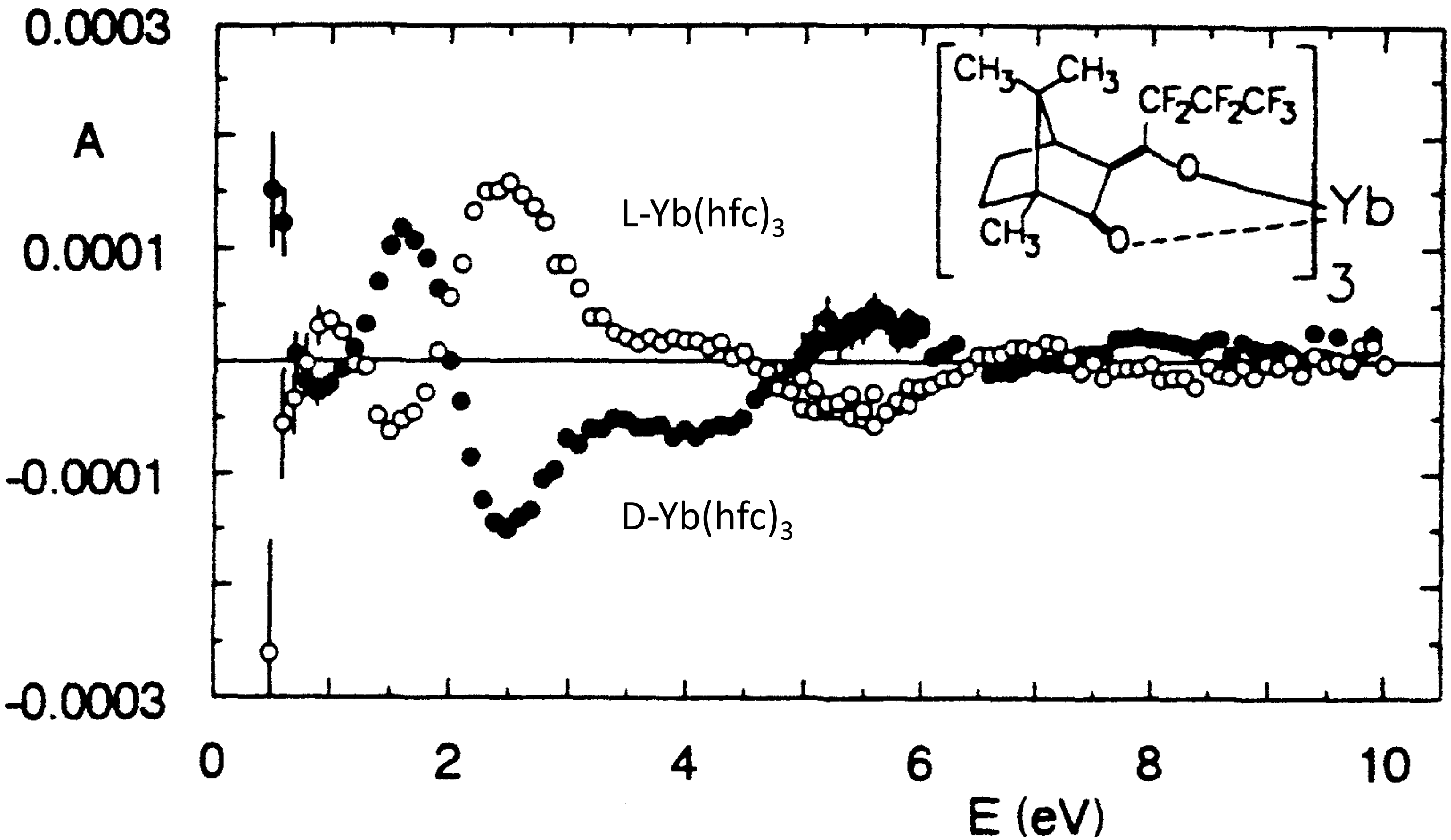


Helium: *n*=3; it still works ☺☺☺ ... Phys. Rev. A **93** (2016) 012712



Transmission Asymmetry for Longitudinally Polarized Electrons in Chiral Molecules

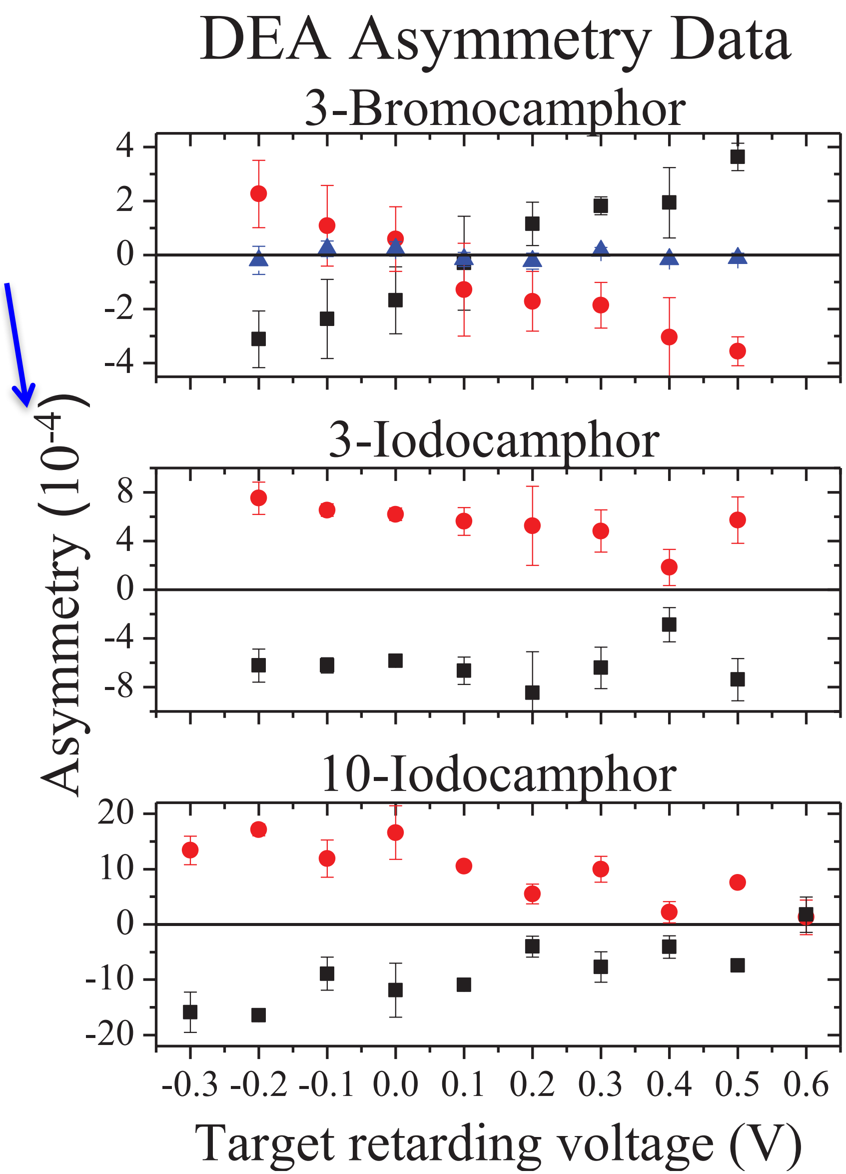
S. Mayer & J. Kessler, Phys. Rev. Lett. **74**, 4803 (1995)

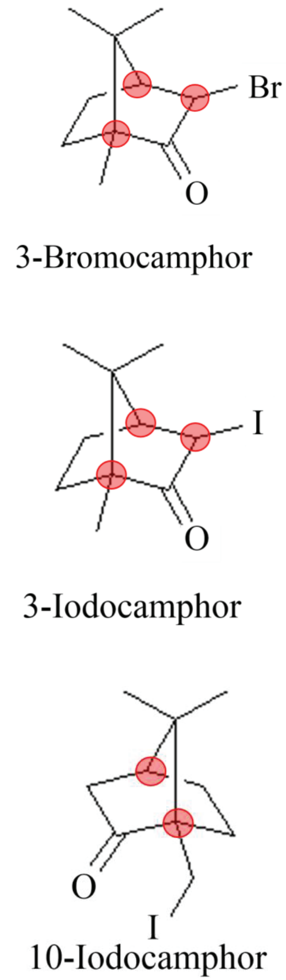


$$A = [I(P) - I(-P)] / [I(P) + I(-P)]$$

Dissociative Electron Attachment of Longitudinally Polarized Electrons in Chiral Molecules

Dreiling & Gay, Phys. Rev. Lett. **116**, 093201 (2016)





This work is relevant for the Vester-Ulbricht model on the origin of biological homochirality.

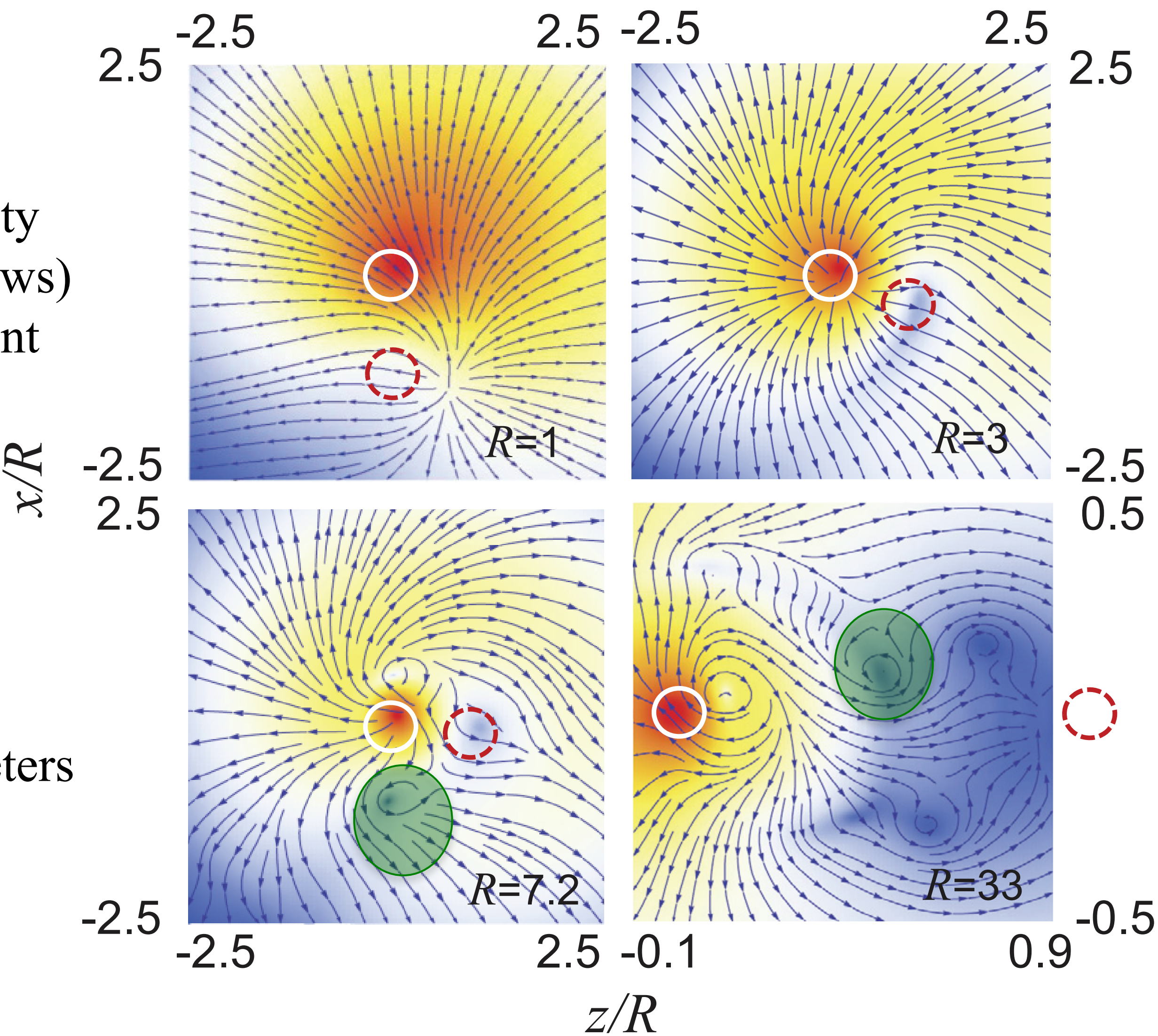
Two Examples on Heavy-Particle Impact:

Vortex formation in antiproton impact on atomic hydrogen

Ovchinnikov, Macek, Schultz; Phys. Rev. A **90**, 062713 (2014)

Electronic probability density (contours) and current (arrows) for 5 keV impact for different impact parameters.

Note the **vortex formation** for the large impact parameters



Ionization of a Li MOT target by a 24 MeV pulsed O^{8+} ion beam

Hubele *et al*., Phys. Rev. Lett. **110** (2013) 133201

An "**orientational dichroism**" in the angular distribution of the ejected electrons due to the oriented 2p (m = -1) state was observed.

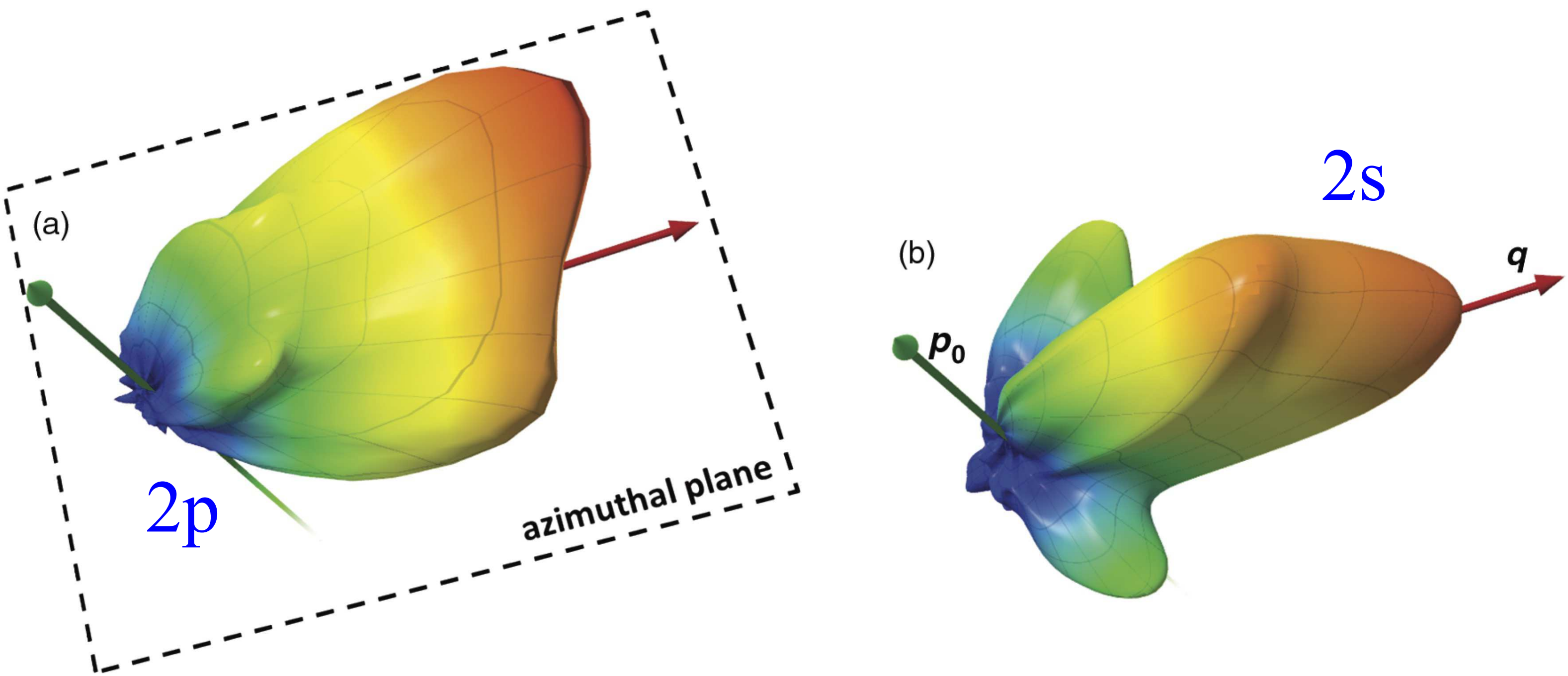
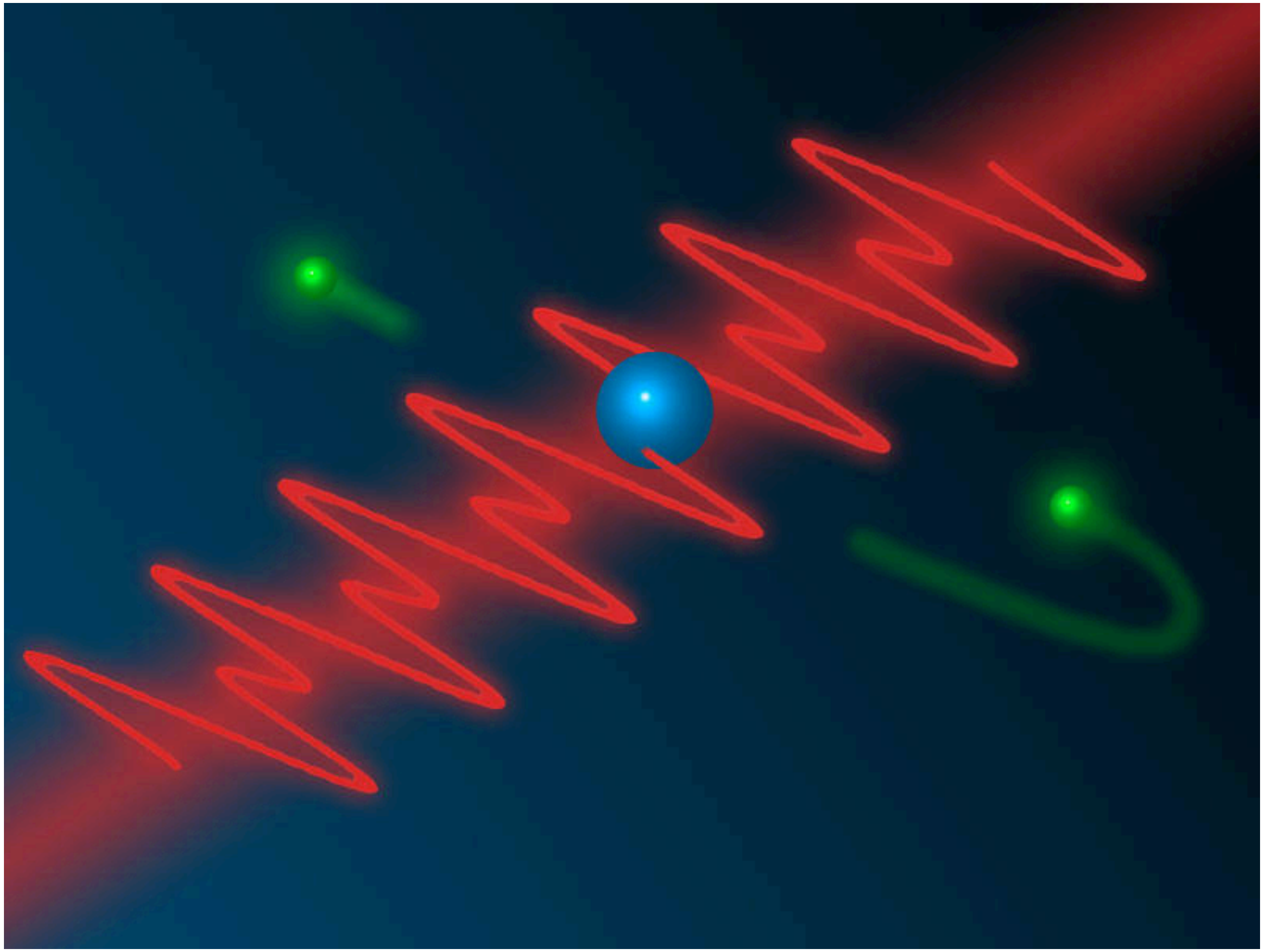


Fig. 10.18 Three-dimensional, fully-differential angular distributions of electrons ejected from the (a) Li(2p) and (b) Li(2s) state by 24 MeV O^{8+} impact. The electron energy is fixed at 1.5 eV and the momentum transfer $\boldsymbol{q}$ at 0.3 a.u. for the Li(2p) state and at 1.0 a.u. for the Li(2s) state [54].

Hint: We have two posters on this next door☺

Photons and Pulses are coming to PAO...

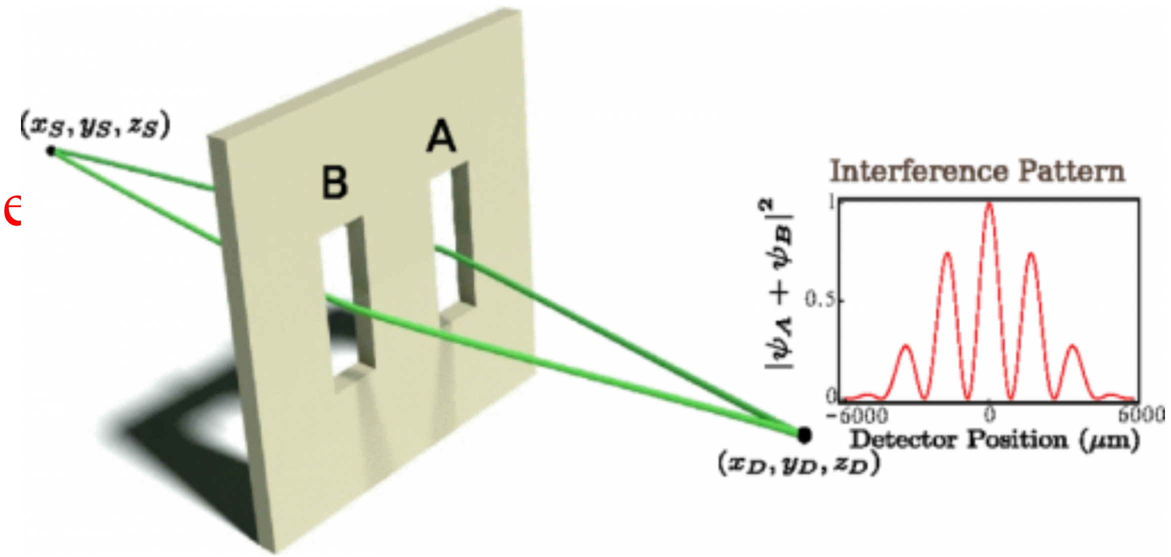
Example: Light-induced Coherent Quantum Control



The Basics

- One of the goals of "quantum control" is to steer electrons into specific directions or locations (e.g., selected bond breaking in a molecule).

- Two-pathways interference is a way to achieve coherent control.



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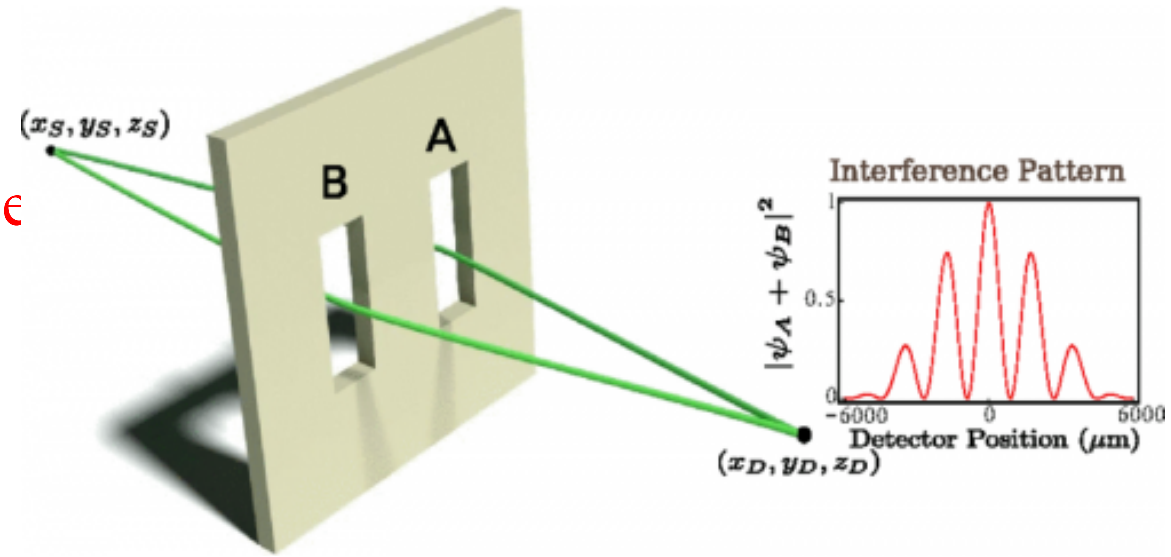
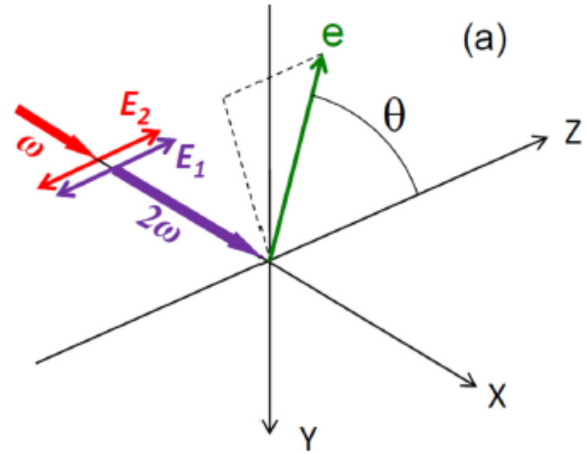


- Photoionization of an atomic system by the fundamental and the second harmonic (ω + 2ω) of a femtosecond VUV pulse is an example of coherent control of the photoelectron angular distribution.

Bichromatic Atomic Ionization with Linearly Polarized Light

- In the case of linearly polarized light, the electric field is expressed as

$$E(t) = F(t)\left[\cos\omega t + \eta\cos(2\,\omega t + \phi)\right]$$



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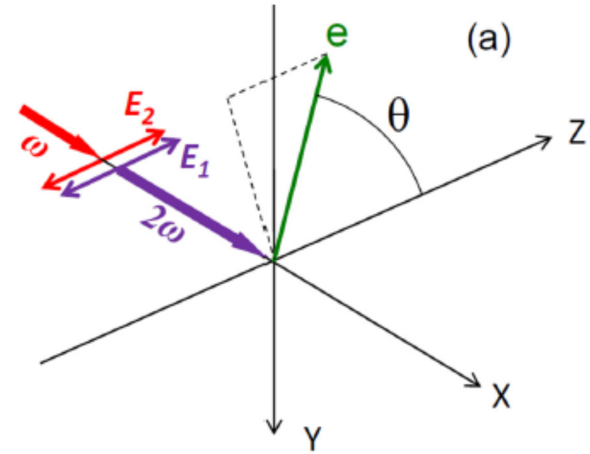

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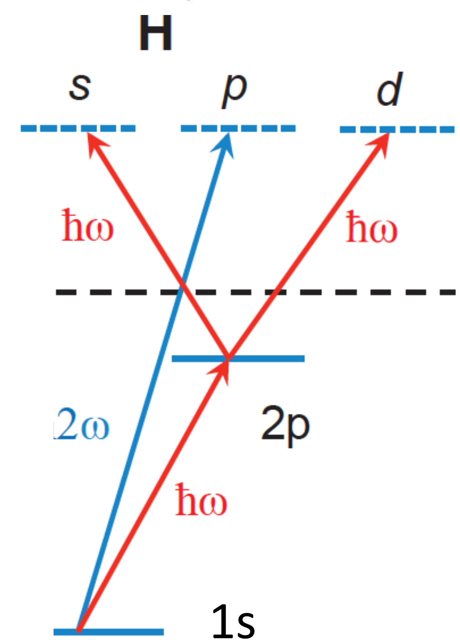
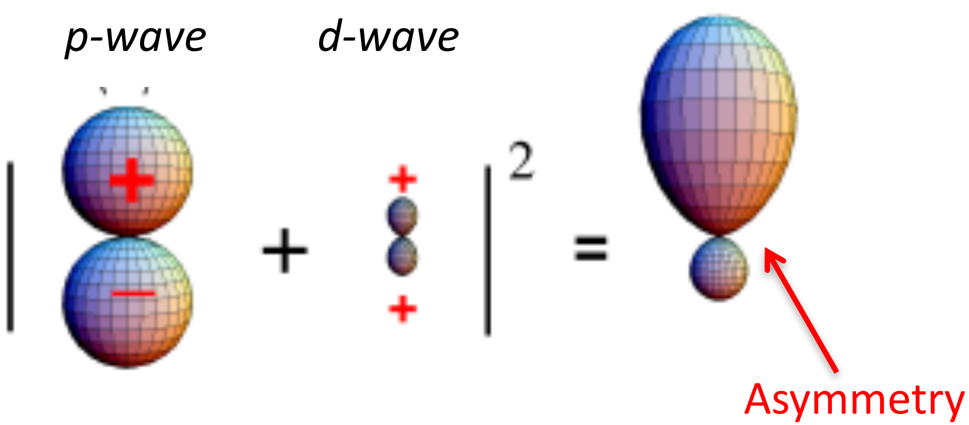
- Two-pathways interference is enhanced by tuning the first harmonic near an intermediate state (e.g. 2p in H).

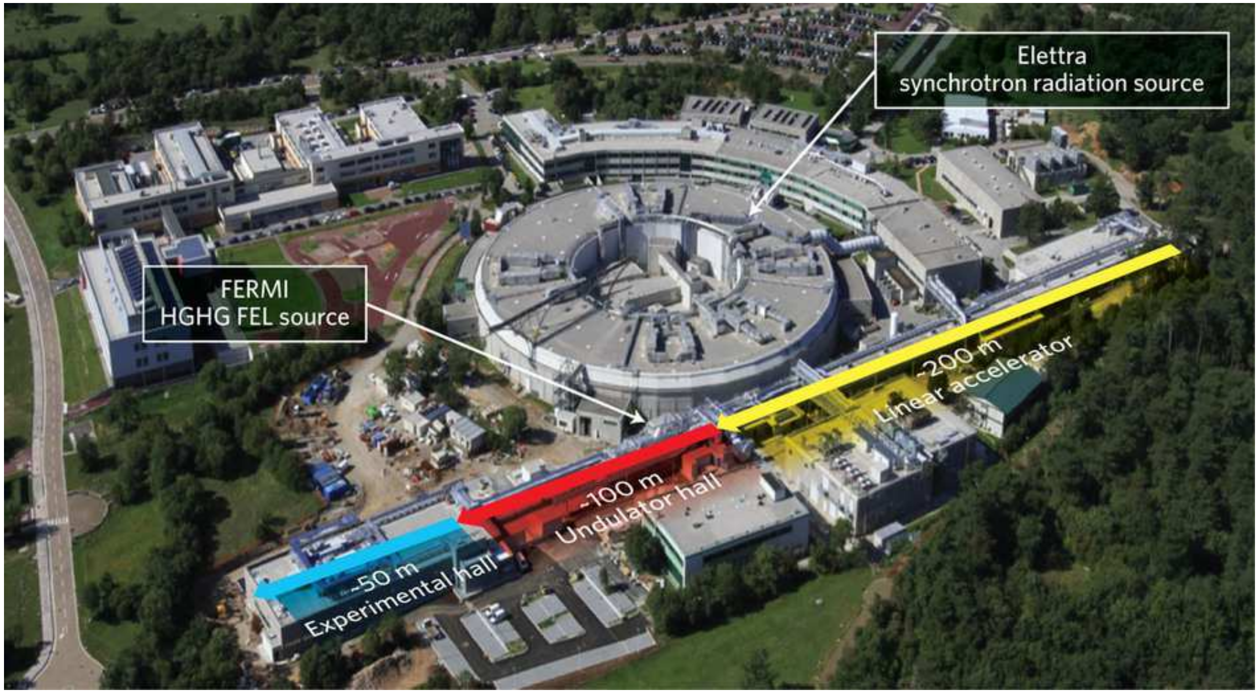


- Ionization leading to partial waves with different parities can cause an asymmetry in the angular distribution.



Without interference

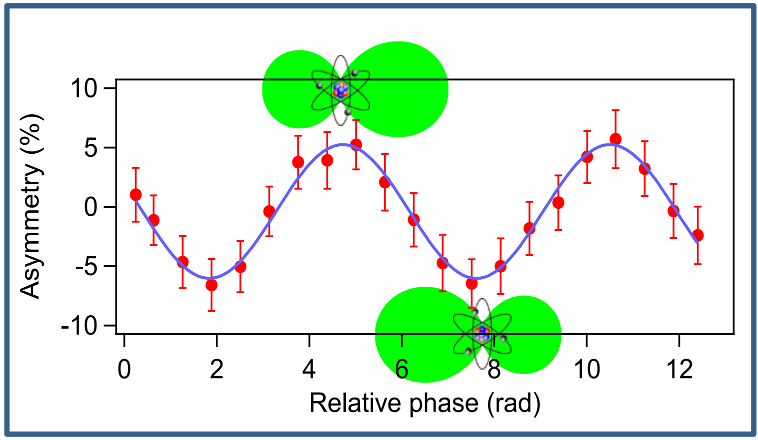
Experimental Setup at FERMI (Trieste, Italy)



<u>Basic idea:</u> Use Ne($2p^6$) as target and tune the fundamental to one of the $(2p^5 4s)_{J=1}$ states.

<u>Results:</u> (more details at K.C. Prince's ICPEAC Talk)

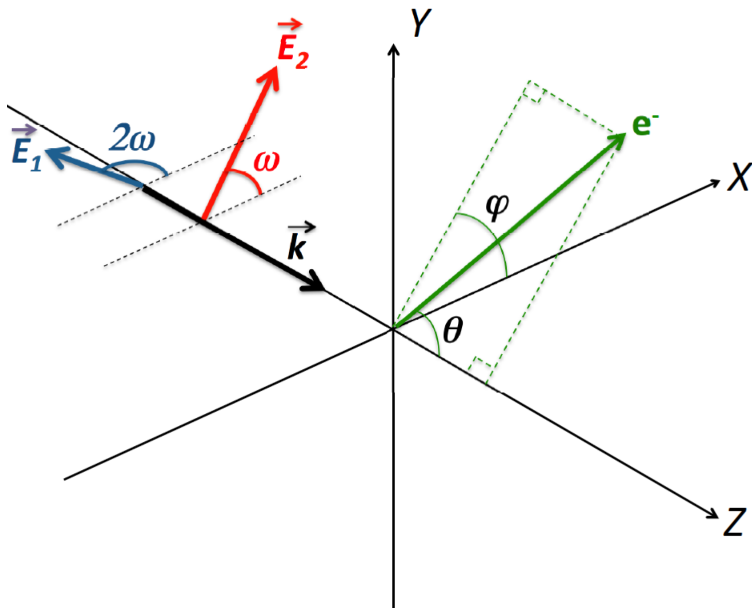
The delay between the two pulses was controlled to a precision better than 3.1 attoseconds (as). This is equivalent to controlling the phase ϕ to high precision [K.C. Prince *et al.*, Nat. Phot. **10** (2016) 176-179]

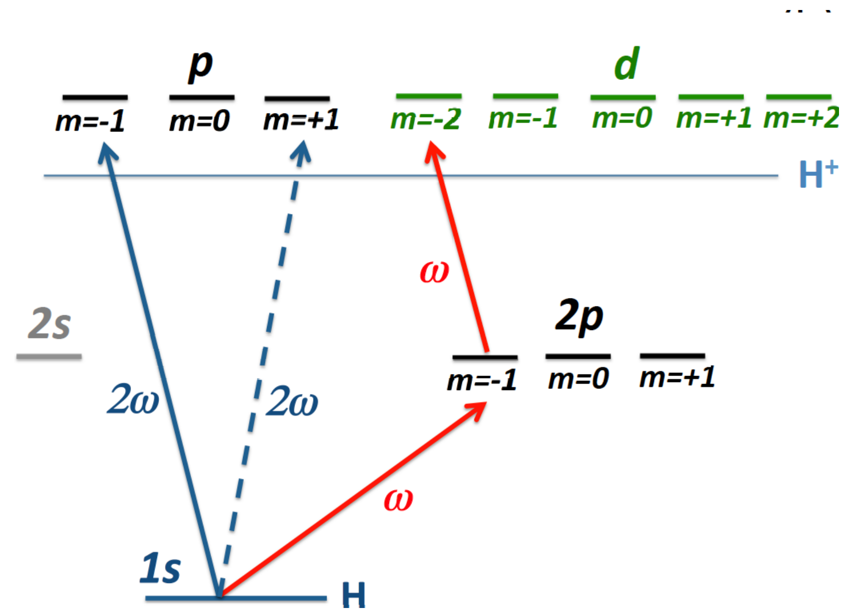


→ ***The asymmetry oscillates as a function of ϕ as predicted theoretically.***

Photoionization Scheme with Circularly Polarized Light in Atomic Hydrogen

- The electric field is in the *XY* plane and propagates along the *Z* axis.





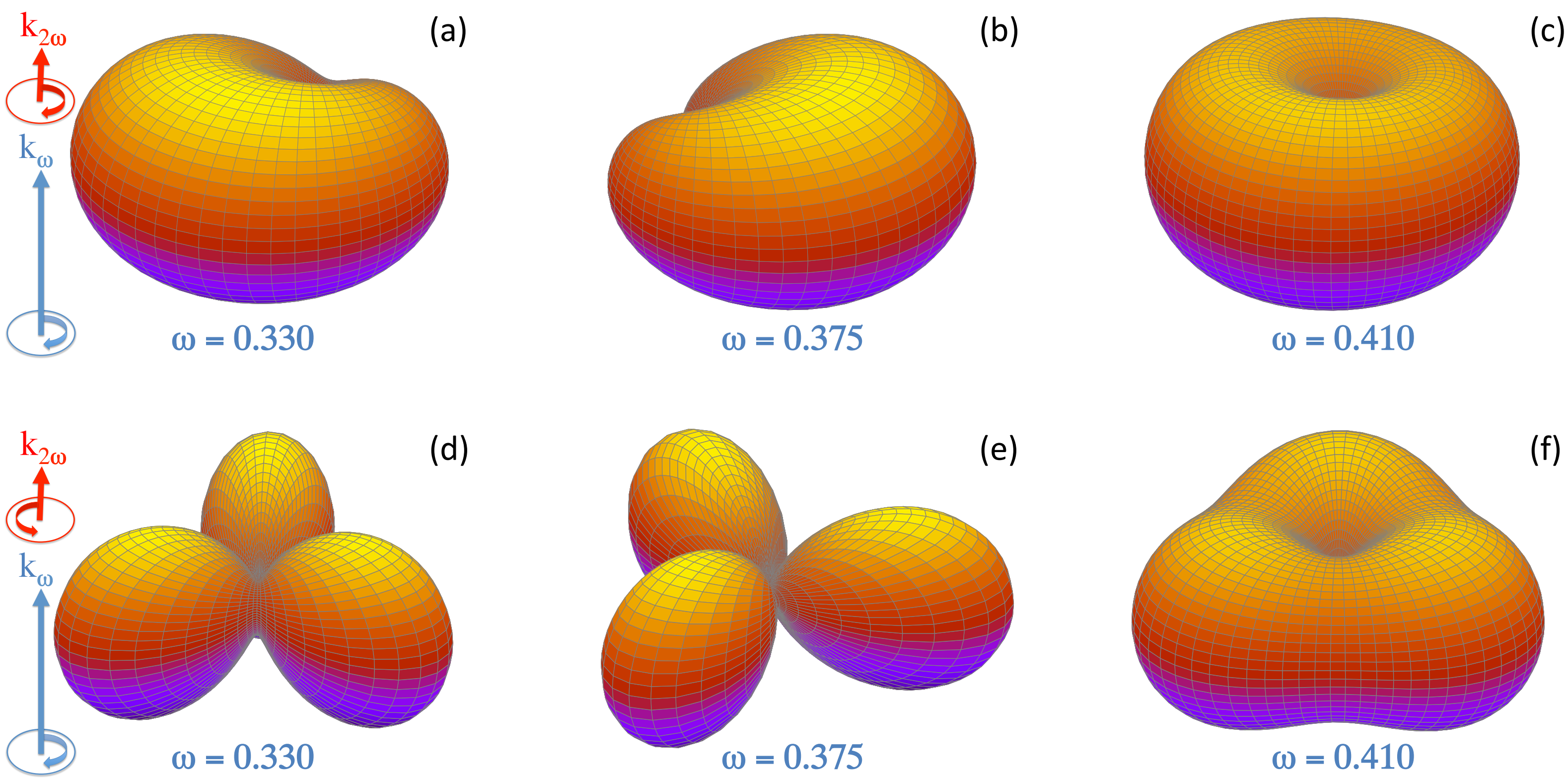
Pulse envelope　Amplitude ratio　CEP　Helicity

$$\mathcal{E}(t) = F(t)\Big[\underbrace{\cos(\omega t)\hat{\boldsymbol{x}} - \sin(\omega t)\hat{\boldsymbol{y}}}_{\text{First Harmonic}} + \eta\underbrace{\{\cos(2\omega t+\phi)\hat{\boldsymbol{x}} + \mathscr{H}\sin(2\,\omega t+\phi)\hat{\boldsymbol{y}}\}}_{\text{Second Harmonic}}\Big]$$

Electric field

H with 2p as stepping stone: Visualizing the PAD in 3D

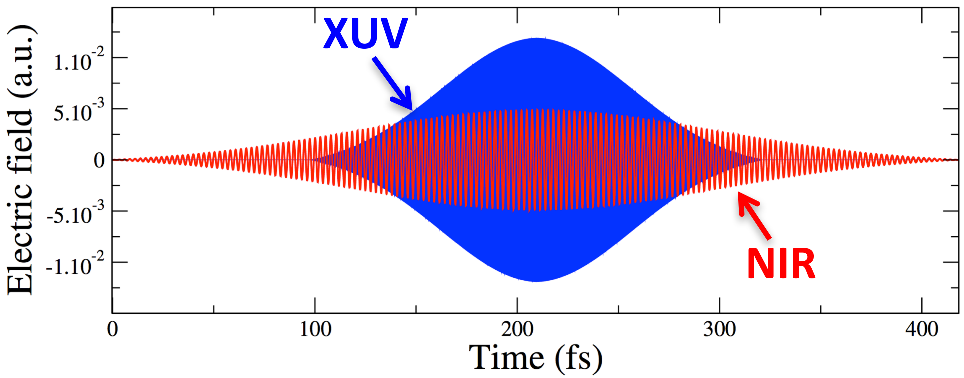
$I = 10^{14}$ W/cm^2



N. Douguet *et al.* Phys. Rev. A **93** (2016) 033402

Circular Dichroism in Oriented He^+

- An overlapping circular XUV + NIR field is created at the FEL at FERMI



- The circularly polarized XUV pulse (FWHM = 100 fs and I = 10^{13}W/cm^2 with positive helicity ($\mathcal{H} = +1$) creates oriented He^+(3p ; m = +1) via sequential absorption of two XUV photons:

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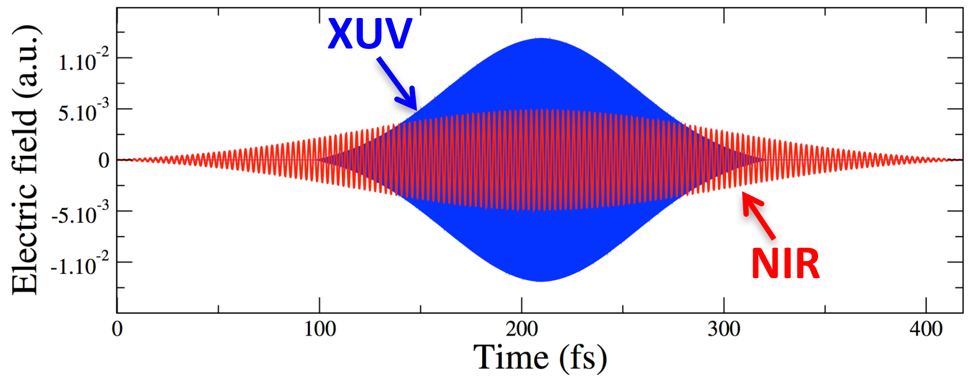

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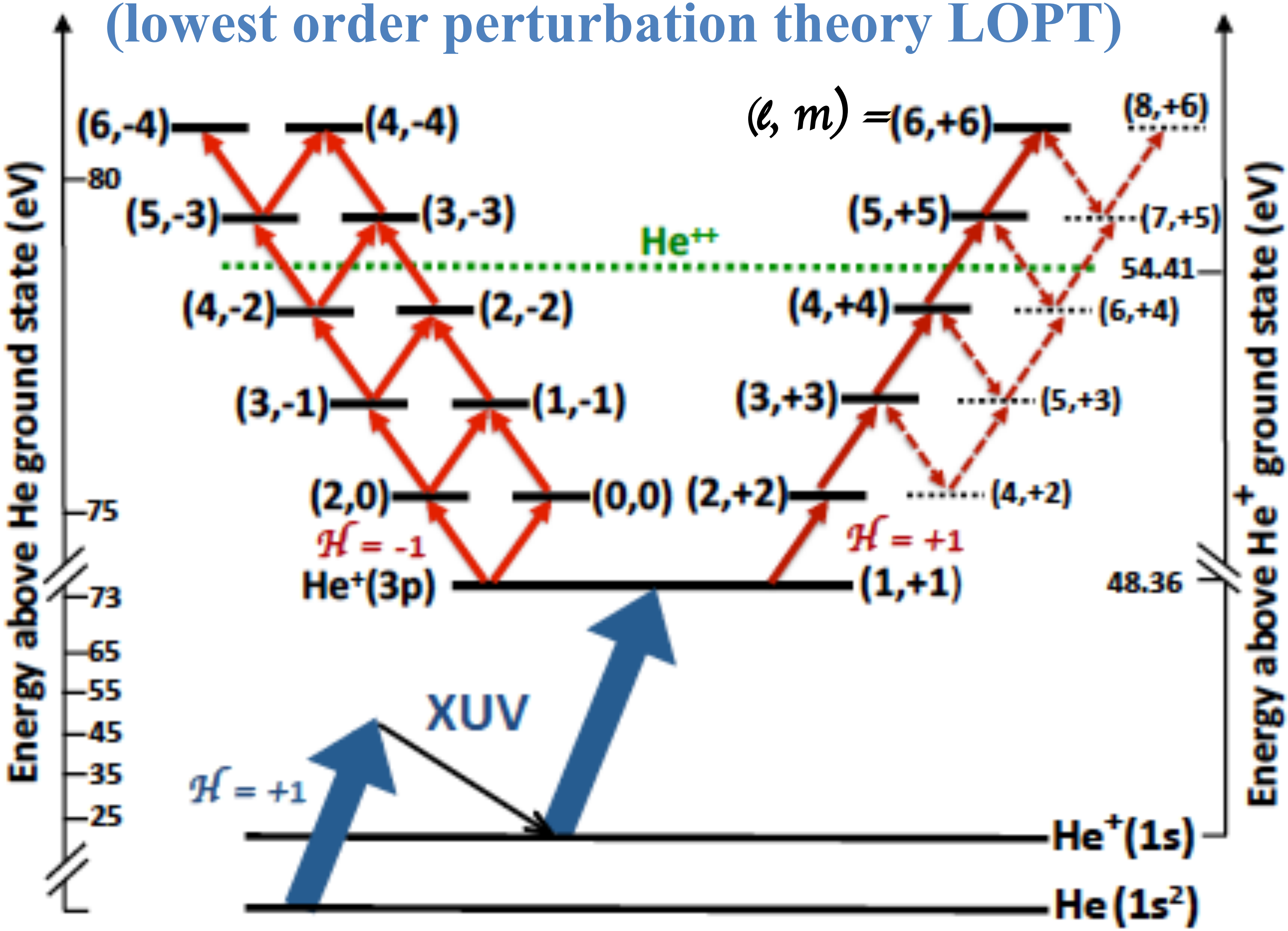
(1) **Ionization** : $He\ (1s^2) + h\nu\ (48.37\ eV) \rightarrow He^+(1s) + e^-$

(2) **Pumping** : $He^+(1s) + h\nu\ (48.37\ eV) \rightarrow He^+(3p;\ m = +1)$

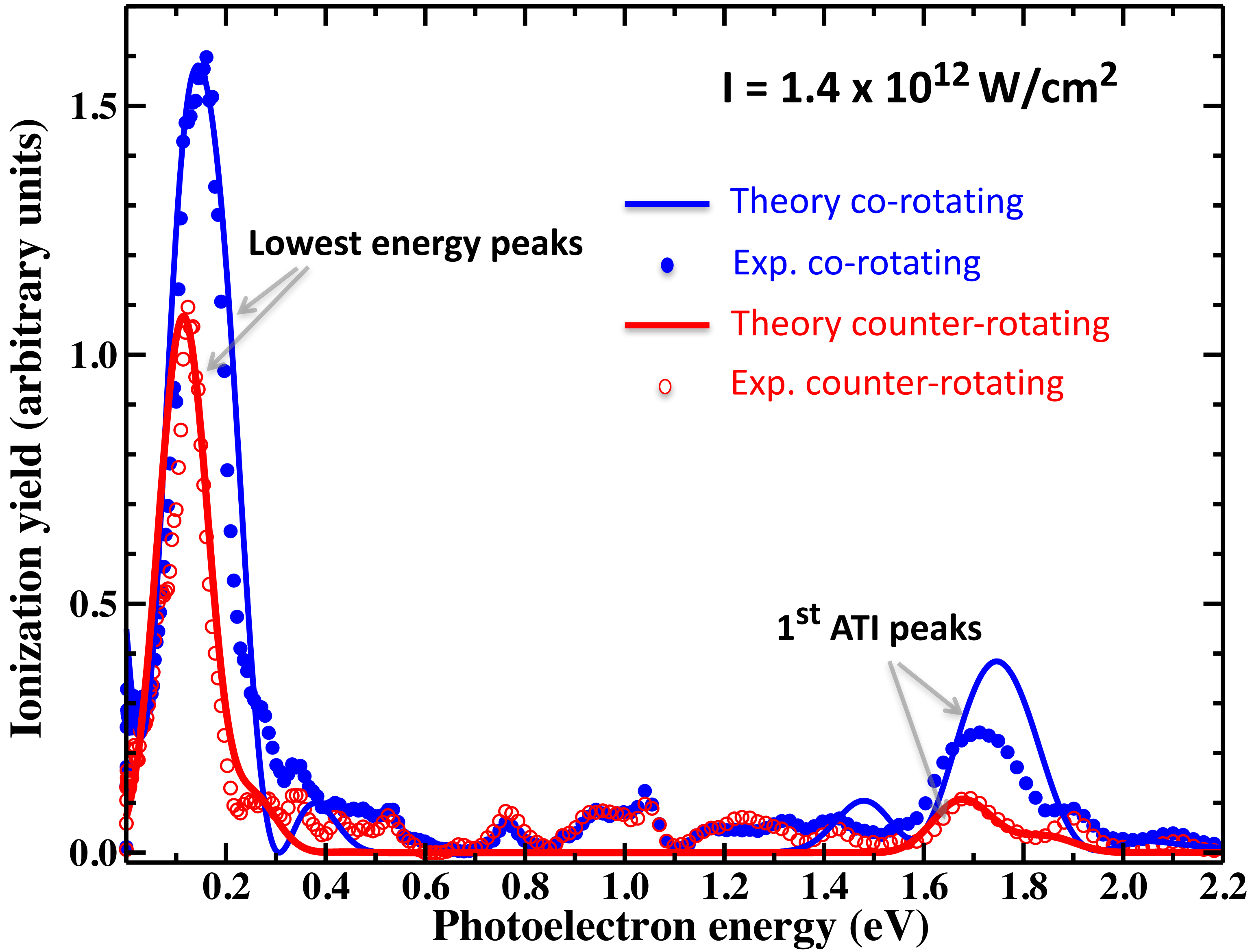
- The overlapping circularly polarized optical laser pulse (FWHM = 170 fs) with ($\mathcal{H} = +1$) or ($\mathcal{H} = -1$) ionizes the oriented $He^+(3p\ ;\ m = +1)$ ion.

(3) **Multiphoton ionization:** $He^+(3p;\ m = +1) + 4\ h\nu\ (1.58\ eV) \rightarrow He^{++} + e^-$

Multiphoton ionization scheme
(lowest order perturbation theory LOPT)

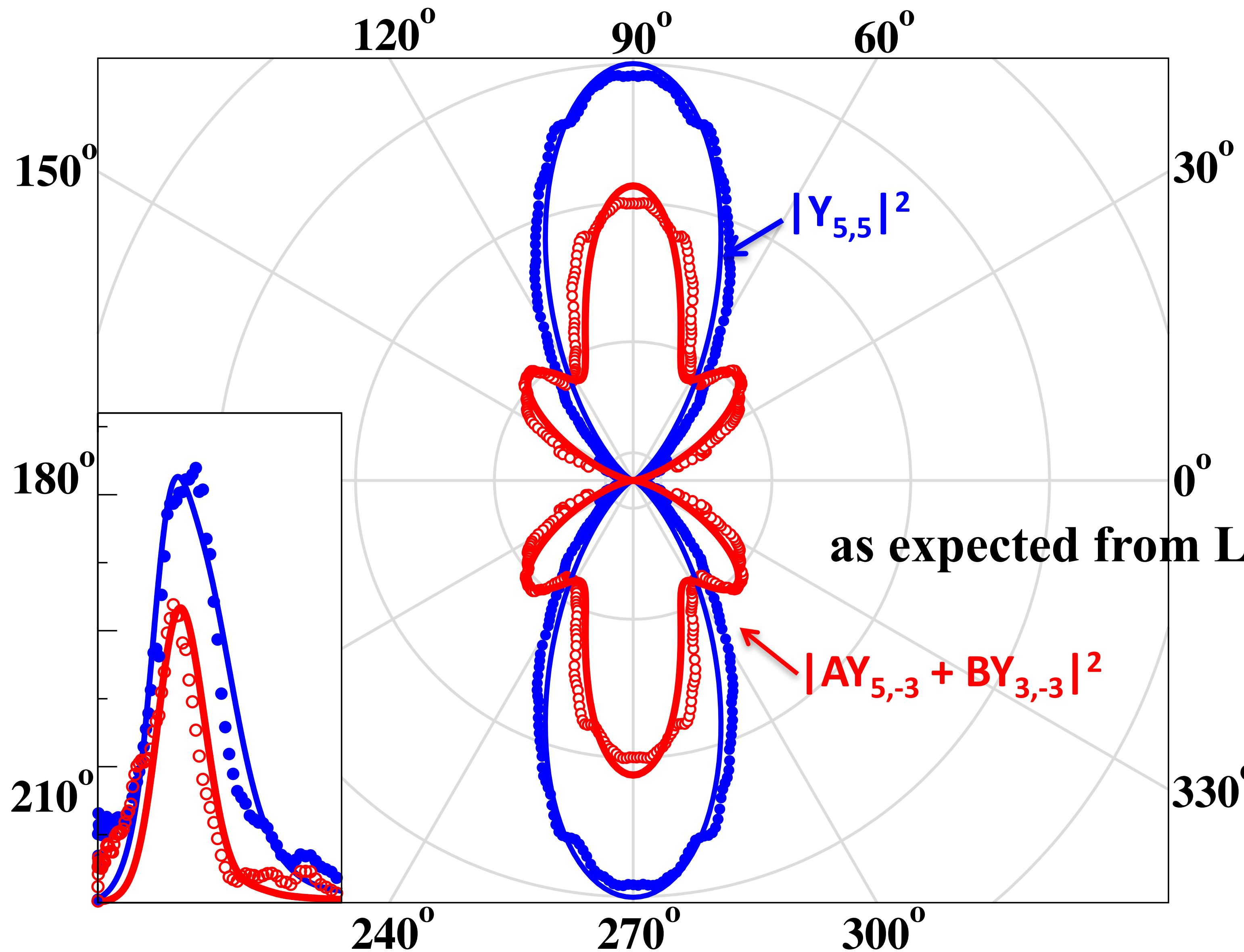


Photoelectron spectrum



M. Ilchen, N. Douguet *et al.*, Phys. Rev. Lett. **118** (2017) 013002

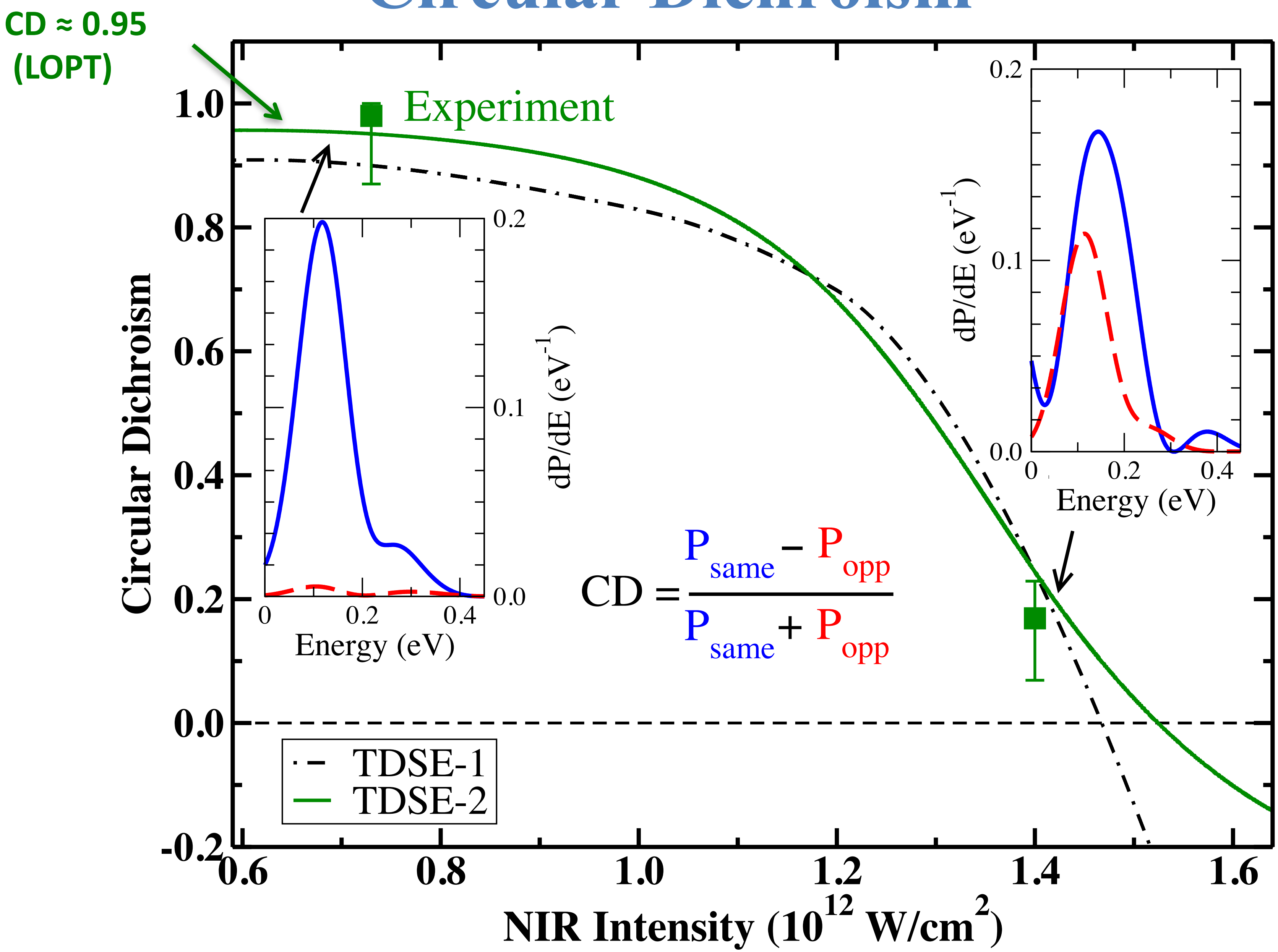
Photoelectron angular distribution



as expected from LOPT

M. Ilchen, N. Douguet *et al.*, Phys. Rev. Lett. **118** (2017) 013002

Circular Dichroism



M. Ilchen, N. Douguet *et al.*, Phys. Rev. Lett. **118** (2017) 013002

A Possible New Direction

PRL **116,** 033201 (2016) PHYSICAL REVIEW LETTERS week ending 22 JANUARY 2016

Entanglement and Bell Correlation in Electron-Exchange Collisions

K. Blum and B. Lohmann*
Institut für Theoretische Physik, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm-Straße 9, D-48149 Münster, Germany
(Received 14 June 2015; published 21 January 2016)

Elastic collisions between initially unpolarized electrons and hydrogenlike atoms are discussed, aiming to analyze the entanglement properties of the correlated final spin system. Explicit spin-dependent interactions are neglected and electron exchange only is taken into account. We show the final spin system to be completely characterized by a single spin correlation parameter depending on scattering angle and energy. Its numerical value identifies the final spins of the collision partners to be either in the separable, entangled, or Bell correlated regions. We emphasize explicit examples for the mixed spin system in order to illustrate the abstract concepts. The analysis of published experimental and numerical data reveals the possibility to create tunable pairs of collision partners with any desired degree of spin entanglement.

DOI: 10.1103/PhysRevLett.116.033201

Another hint: We also have a poster on this next door☺

PHYSICAL REVIEW A **94**, 032331 (2016)

Tunable entanglement resource in elastic electron-exchange collisions out of chaotic spin systems

B. Lohmann,[1,*] K. Blum,[1] and B. Langer[2]
[1] *Institut für Theoretische Physik, Westfälische Wilhelms-Universität Münster, Wilhelm-Klemm-Strasse 9, 48149 Münster, Germany*
[2] *Physikalische Chemie, Freie Universität Berlin, Taku-Strasse 3, 14195 Berlin, Germany*
(Received 5 July 2016; published 29 September 2016)

Elastic collisions between initially unpolarized electrons and hydrogenlike atoms are discussed aiming to analyze the entanglement properties of the correlated final spin system. Explicit spin-dependent interactions are neglected and electron exchange only is taken into account. We show the final spin system to be completely characterized by a single spin correlation parameter depending on scattering angle and energy. Its numerical value identifies the final spins of the collision partners to be either in the separable, entangled, or Bell correlated regions. The symmetry of the scattering process allows for the construction of explicit examples applying methods of classical communication and local operations for illustrating the concepts of nonlocality versus separability. It is shown that strong correlations can be produced violating Bell's inequalities significantly. Furthermore, the degree of entanglement can be continuously varied simply by changing either the scattering angle and/or energy. This allows for the generation of tunable spin pairs with any desired degree of entanglement. It is suggested to use such nonlocally entangled spin pairs as a resource for further experiments, for example in quantum information processes.

DOI: 10.1103/PhysRevA.94.032331

General Theory

- Typically, Bell correlations are discussed for two particles forming a **pure state of zero spin or orbital angular momentum**.
- Examples are:
  - **two electrons** (or other spin-1/2 particles) starting from a $^1\mathbf{S}_0$ **state**;
  - **two-photon decay** for an $\mathbf{S} \rightarrow \mathbf{S}$ optical transition.

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- The situation proposed by Blum and Lohmann is **different from a pure state**. They consider the scattering of an **unpolarized electron beam from an also unpolarized beam of (quasi-)one-electron atoms (H, Li, Na, ...)**
- **After the collision**, the projectile and the target valence electron are **correlated due to the possibility of exchange**.
- The total spin of the system is an energy- and angle-dependent **mixture of singlet and triplet states**.

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- **After the collision**, the projectile and the target valence electron are **correlated due to the possibility of exchange**.
- The total spin of the system is an energy- and angle-dependent **mixture of singlet and triplet** states.
- The **degree of correlation is determined by the spin correlation parameter** $\boldsymbol{P}$, which depends on the collision energy and the scattering angle.
- The limiting values are $P = +1/3$ for pure triplet and $P = -1$ for pure singlet scattering.

General Theory (continued)

- One can go further and **analyze the density matrix for the combined projectile + target spin system**.
- According to criteria derived by Peres (Phys. Rev. Lett. **77** (1996) 1413) and Horodecki *et al.* (Phys. Lett. A **223** (1996) 1), the system can be classified as
  - **separable (S)** if $P > -1/3$,
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"Spukhafte Fernwirkung = spooky action at a distance"

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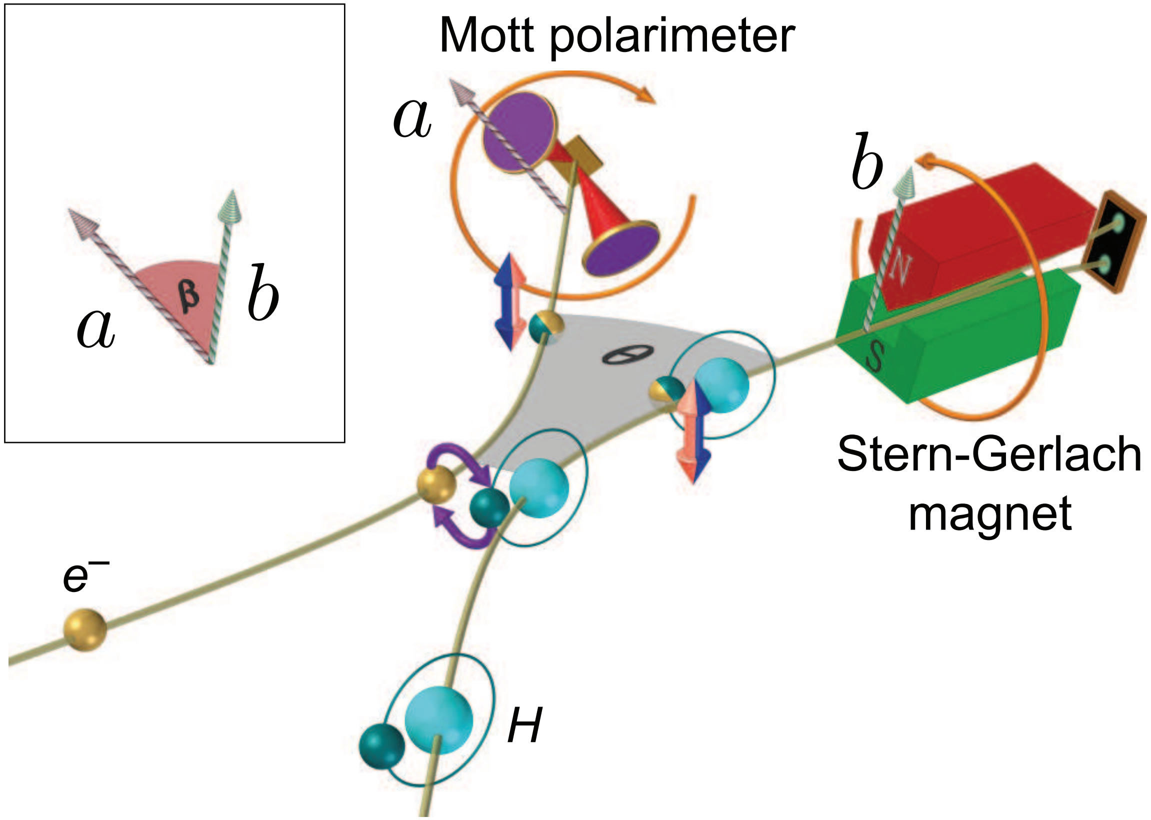
- In the Bell-correlated regime, any further manipulation of the electron that remains in the target would also affect (in a nonlocal way) the continuum electron that is long gone!
- **Due to the energy- and angle-dependence of $\boldsymbol{P}$, the degree of entanglement is tunable!**
- The electron−atom system after the collision can thus serve as a **source to provide the desired degree of entanglement** in (hopefully) forthcoming sophisticated experiments on **quantum information processes**.

Practical Considerations

- A **direct measurement of *P*** would require a **coincidence experiment with polarization analysis** of both electrons.



- This seems **(nearly) impossible with current technology**.

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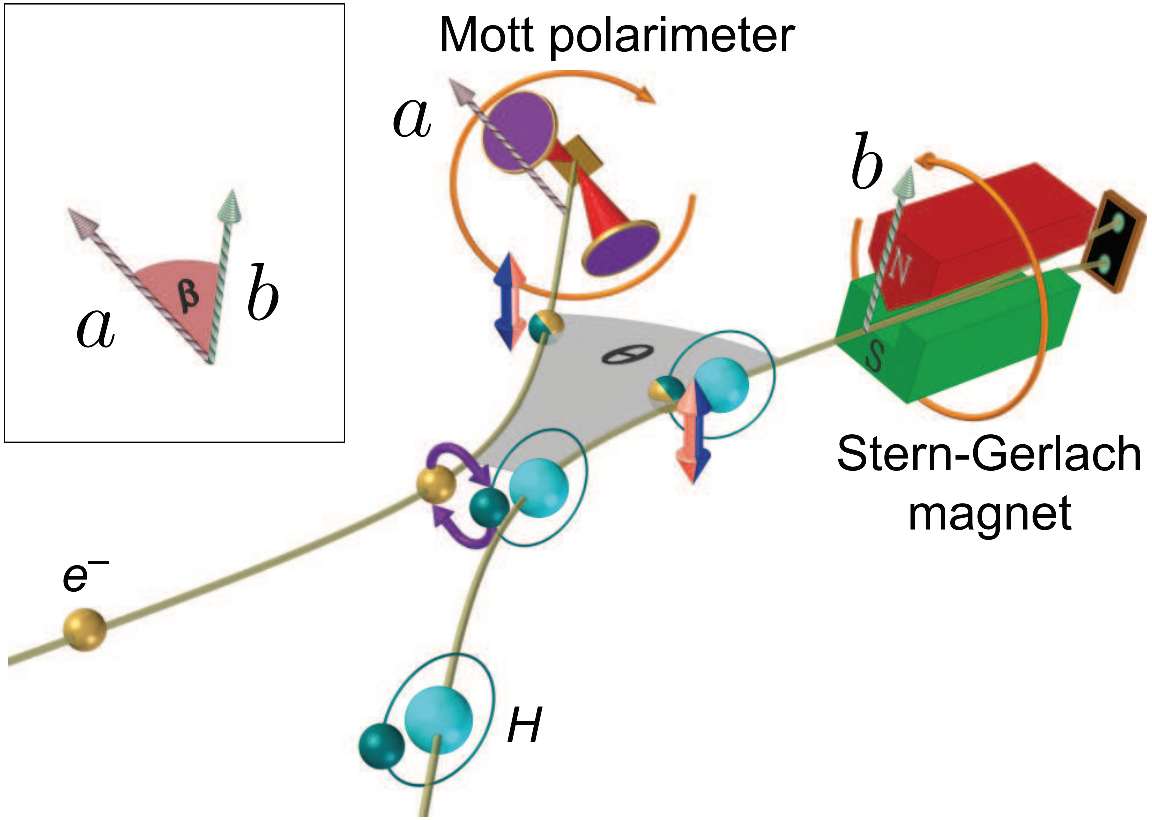

- This seems **(nearly) impossible with current technology**.
- **Fortunately, one can substitute this experiment by a setup using initially spin-polarized beams.**

Basic Setup (Alternative Experiment)

- Consider the elastic scattering of an electron beam with degree of spin polarization P_e from a spin-polarized target with spin polarization P_A.
- One now measures the asymmetry

$$\frac{1}{P_\mathrm{e} P_\mathrm{A}} \, \frac{N^{\uparrow\uparrow} - N^{\uparrow\downarrow}}{N^{\uparrow\uparrow} + N^{\uparrow\downarrow}}$$

where $N^{\uparrow\uparrow}$ ($N^{\uparrow\downarrow}$) are the count rates for parallel (anti-parallel) spin orientations of the projectile and target spins.

Baum *et al.*, Phys. Rev. Lett. **57** (1986) 1855

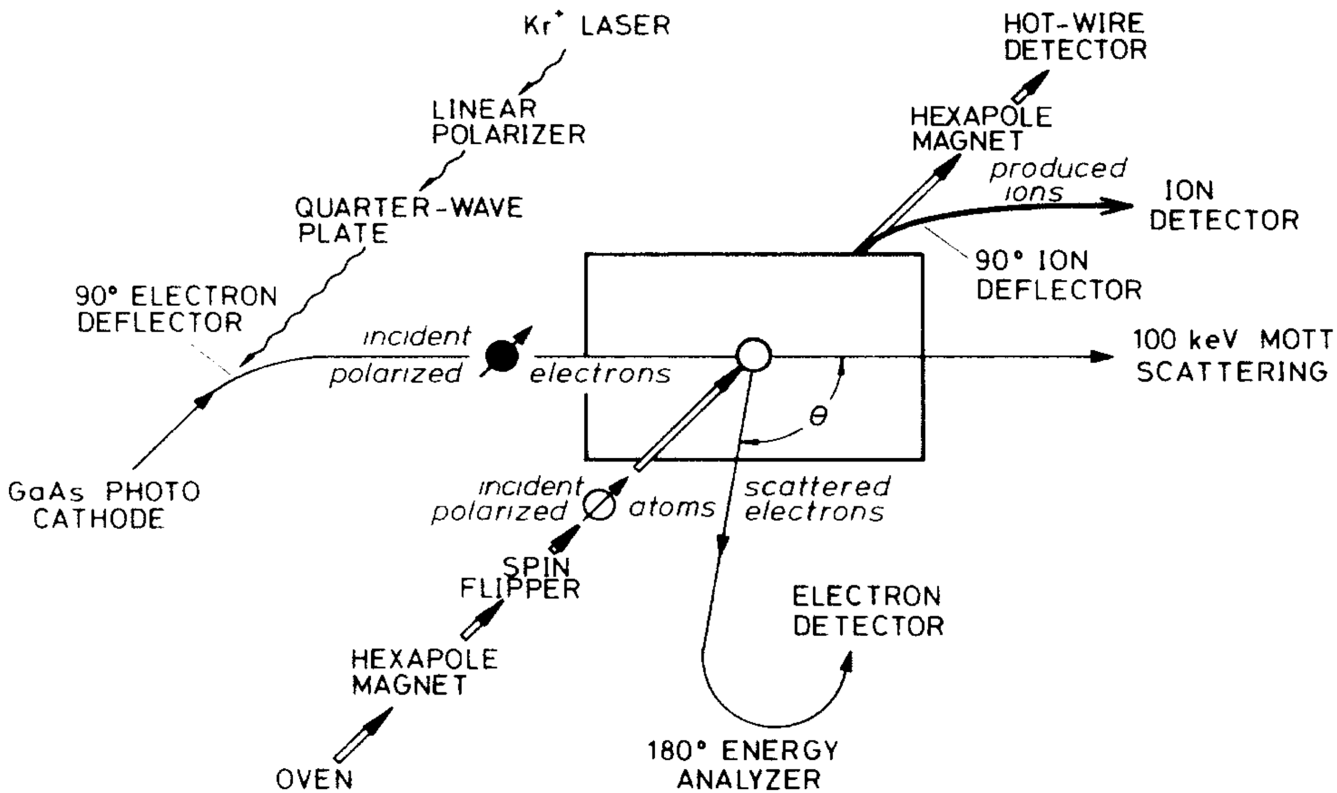

FIG. 1. Schematic diagram of experiment.

Basic Setup (Alternative Experiment)

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  where $N^{\uparrow\uparrow}$ $(N^{\uparrow\downarrow})$ are the count rates for parallel (anti-parallel) spin orientations of the projectile and target spins.
- This asymmetry can also be written as

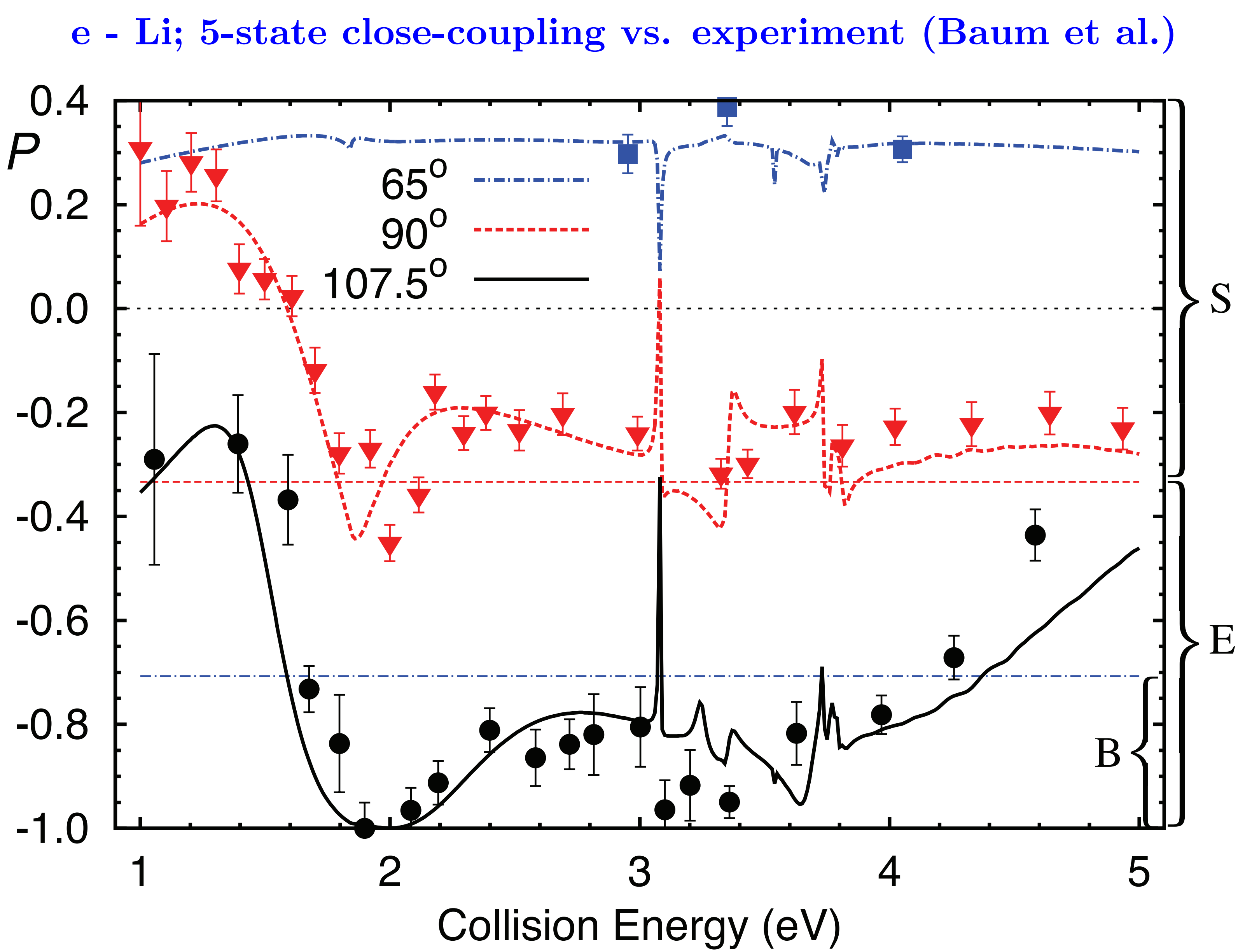
$$P = -A_{\mathrm{ex}} = \frac{\sigma^{\uparrow\uparrow} - \sigma^{\uparrow\downarrow}}{\sigma^{\uparrow\uparrow} + \sigma^{\uparrow\downarrow}} = \frac{\sigma^{t} - \sigma^{s}}{3\,\sigma^{t} + \sigma^{s}}$$

  where $\sigma^{\uparrow\uparrow}$ $(\sigma^{\uparrow\downarrow})$ and σ^{s} (σ^{t}) denote the angle-differential cross sections for parallel (anti-parallel) spin orientations of the projectile and target spins or triplet (singlet) scattering.
- The DCS for unpolarized projectile and target beams is given by

$$\sigma_u = \frac{1}{4}\sigma^{s} + \frac{3}{4}\sigma^{t}.$$

Practical Considerations

- A **direct measurement of *P*** would require a **coincidence experiment with polarization analysis** of both electrons.
- This seems **(nearly) impossible with current technology**.
- **Fortunately, one can substitute this experiment by a setup using initially spin-polarized beams**.
- Such experiments were performed about 20 years ago in Bielefeld (Baum and collaborators) and at NIST (McClelland, Kelley, and collaborators), with the main motivation being to **provide benchmark data for electron-atom collision theories**.
- For such systems, very accurate calculations based on convergent close-coupling formulations such as **CCC** and R-matrix with pseudo-states (**RMPS**) have now become possible.
- Blum and Lohmann used the existing results, which were limited to particular energies and angles, for their analysis.



e - Li; 5-state close-coupling vs. experiment (Baum et al.)
P
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
-1.0
65°
90°
107.5°
S
E
B
1
2
3
4
5
Collision Energy (eV)

Practical Considerations

- A **direct measurement of *P*** would require a **coincidence experiment with polarization analysis** of both electrons.
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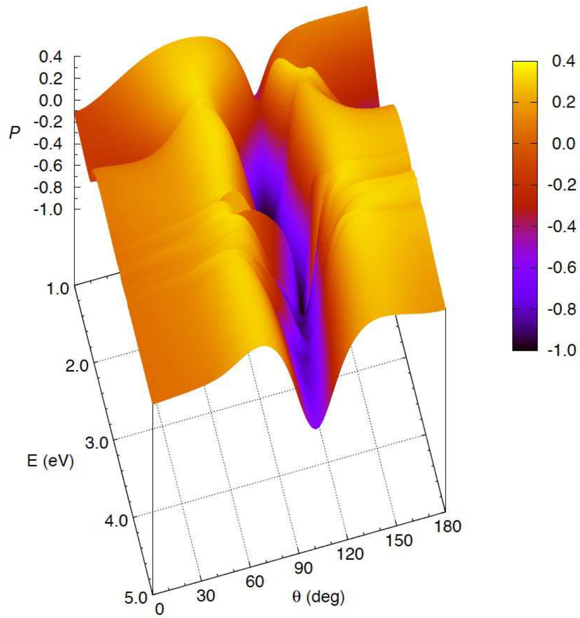
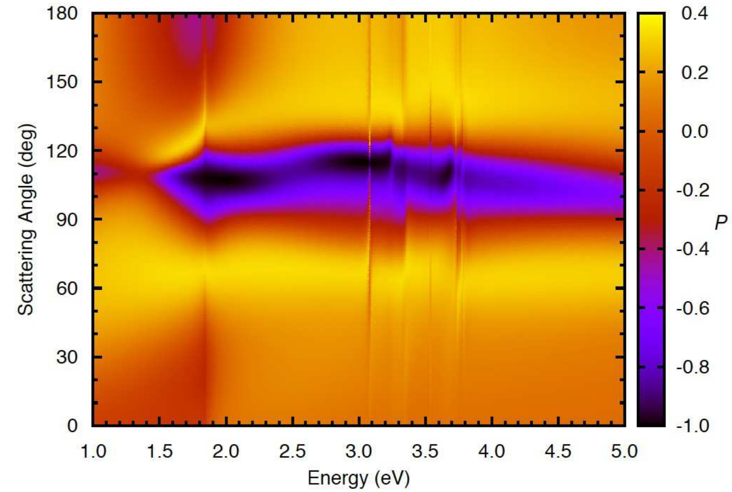
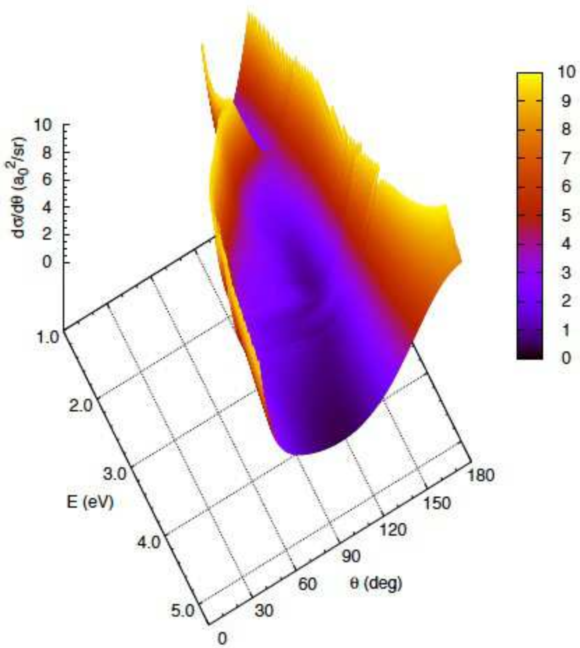
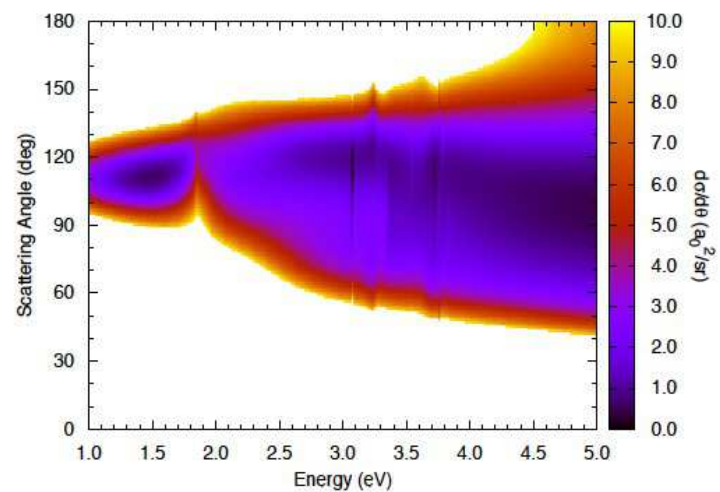
- For such systems, very accurate calculations based on convergent close-coupling formulations such as **CCC** and R-matrix with pseudo-states (**RMPS**) have now become possible.
- Blum and Lohmann used the existing results, which were limited to particular energies and angles, for their analysis.
- Because of **possible applications in quantum information**, it would be very useful to have **accurate and comprehensive numerical data available over a dense energy-angle grid**.

e - Li; 5-state close-coupling









Recent Publication

PHYSICAL REVIEW A **95**, 042707 (2017)

Spin entanglement in elastic electron scattering from lithium atoms

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Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA
(Received 21 November 2016; published 19 April 2017)

In two recent papers [Blum and Lohmann, Phys. Rev. Lett. **116**, 033201 (2016); Lohmann *et al.*, Phys. Rev. A **94**, 032331 (2016)], the possibility of continuously varying the degree of entanglement between an elastically scattered electron and the valence electron of an alkali-metal target was discussed. To estimate how well such a scheme may work in practice, we present results for elastic electron scattering from lithium in the energy regime of 1–5 eV and the full range of scattering angles 0°–180°. The most promising regime for Bell correlations in this particular collision system are energies between about 1.5 and 3.0 eV, in an angular range around 110° ± 10°. In addition to the relative exchange asymmetry parameter, we present the differential cross section that is important when estimating the count rate and hence the feasibility of experiments using this system.

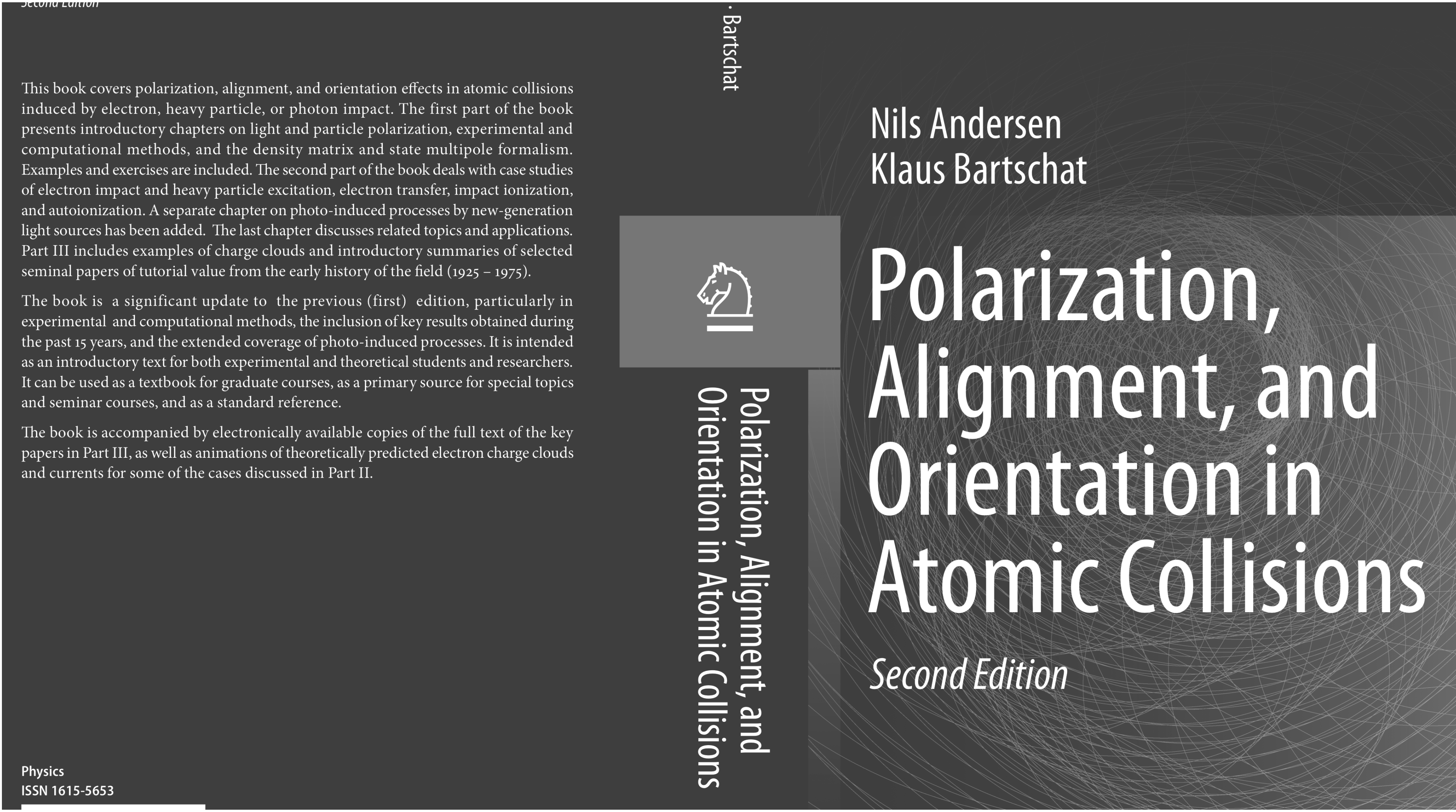
DOI: 10.1103/PhysRevA.95.042707

We also have results for H, Na, K, Rb, and Cs.
The best targets seem to be Li and Na.

Summary

- Using a few selected examples, I tried to give you an impression of where the field of polarization, alignment, and orientation in atomic (and molecular) collisions went during the past 20 years.
- If you want to know the long story, please read .....

THIS!


This book covers polarization, alignment, and orientation effects in atomic collisions induced by electron, heavy particle, or photon impact. The first part of the book presents introductory chapters on light and particle polarization, experimental and computational methods, and the density matrix and state multipole formalism. Examples and exercises are included. The second part of the book deals with case studies of electron impact and heavy particle excitation, electron transfer, impact ionization, and autoionization. A separate chapter on photo-induced processes by new-generation light sources has been added. The last chapter discusses related topics and applications. Part III includes examples of charge clouds and introductory summaries of selected seminal papers of tutorial value from the early history of the field (1925 – 1975).

The book is a significant update to the previous (first) edition, particularly in experimental and computational methods, the inclusion of key results obtained during the past 15 years, and the extended coverage of photo-induced processes. It is intended as an introductory text for both experimental and theoretical students and researchers. It can be used as a textbook for graduate courses, as a primary source for special topics and seminar courses, and as a standard reference.

The book is accompanied by electronically available copies of the full text of the key papers in Part III, as well as animations of theoretically predicted electron charge clouds and currents for some of the cases discussed in Part II.

Physics
ISSN 1615-5653

· Bartschat

Polarization, Alignment, and Orientation in Atomic Collisions

Nils Andersen
Klaus Bartschat

Polarization, Alignment, and Orientation in Atomic Collisions

Second Edition

Thank You for Your Attention!