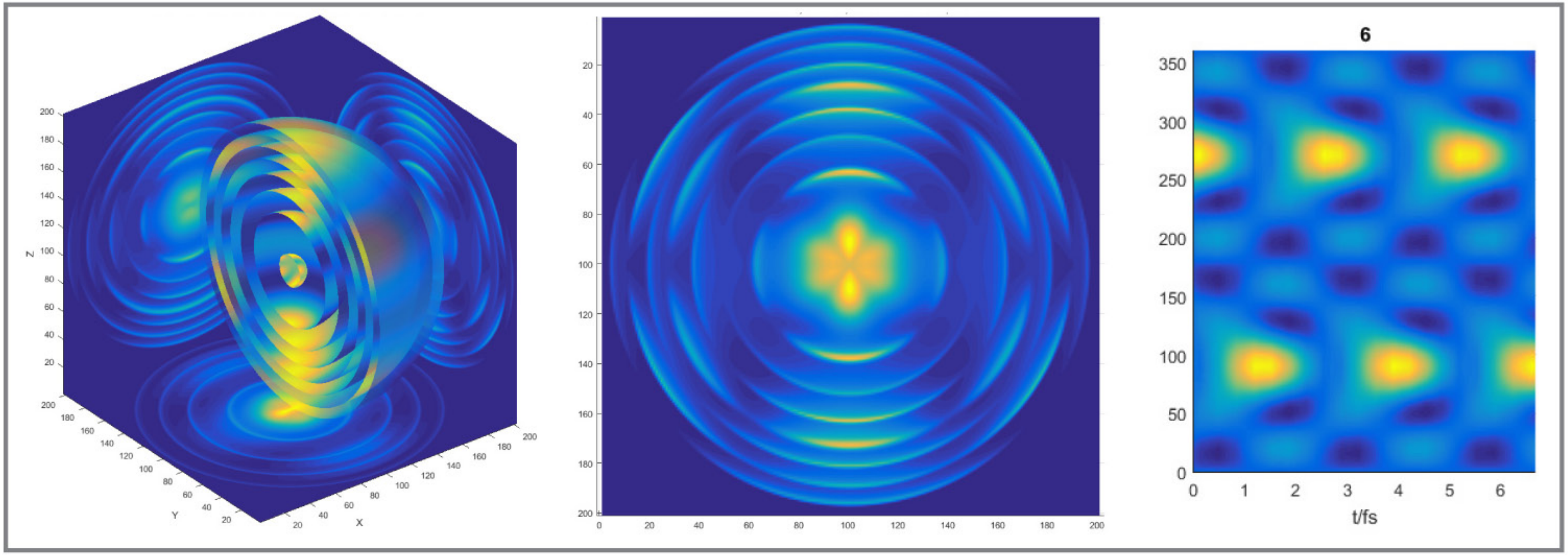
Phase-sensitive Photoelectron Metrology



Paul Hockett

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National Research Council of Canada, Ottawa

Available via Figshare, DOI: 10.6084/m9.figshare.5049142

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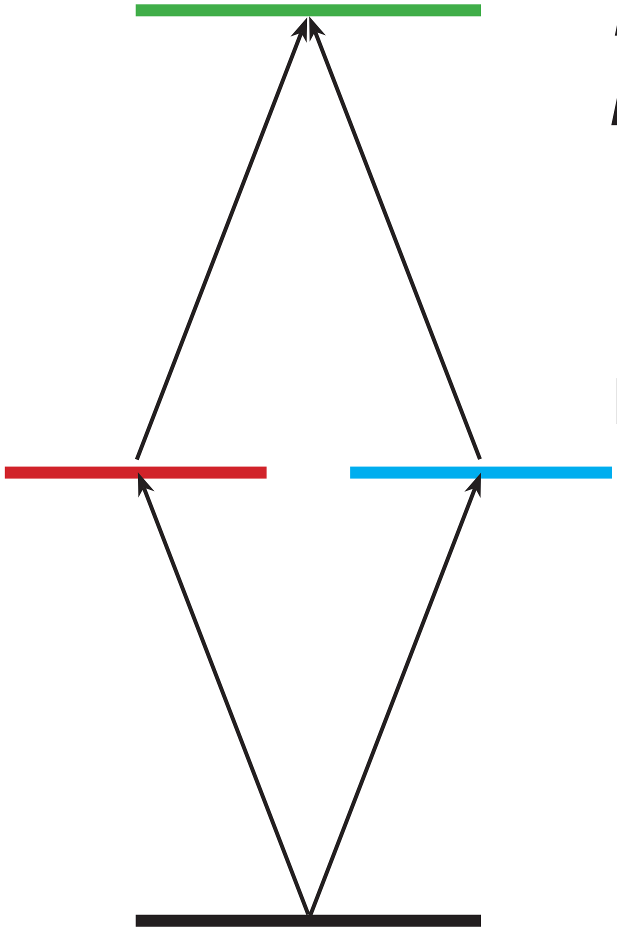
Interferometers...

Final state
Depends on amplitudes and relative phases of path 1 & 2

Path 1

Path 2



matter wave interferometry

Young's double slit with electrons... two-path quantum interferometry.

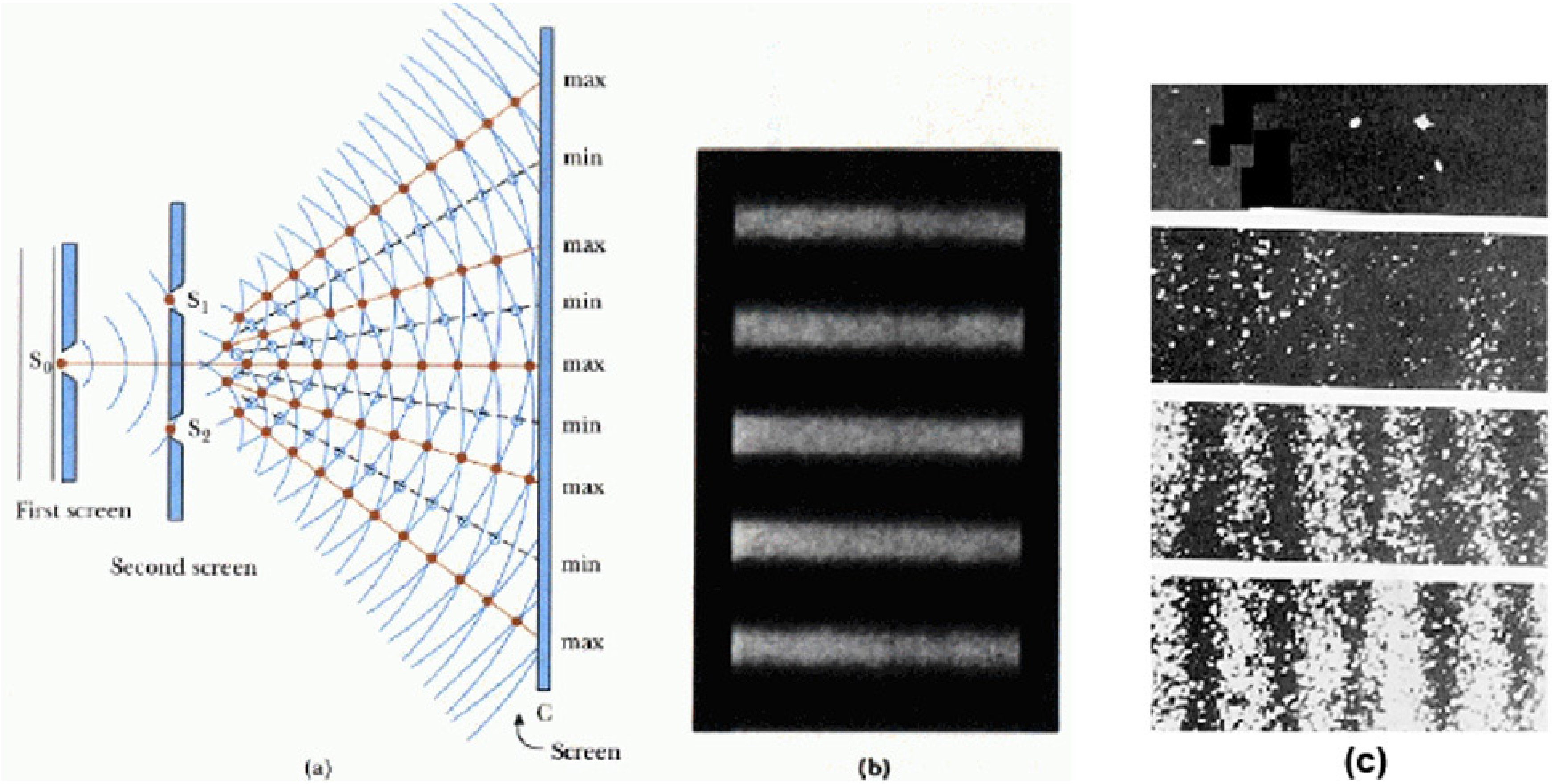


Figure credit: Simon Connell. http://psi.phys.wits.ac.za/teaching/connell/phys284/2005/lecture-02

general quantum interferometry

... pretty much any quantum mechanical process where *multiple paths* play a role.

For example, the phase of a bound-state wavefunction:

VOLUME 85, NUMBER 10 PHYSICAL REVIEW LETTERS 4 SEPTEMBER 2000

Direct Observation of a Breit-Wigner Phase of a Wave Function

Jeanette A. Fiss, Ani Khachatrian, Kaspars Truhins, Langchi Zhu, and Robert J. Gordon
Department of Chemistry (m/c 111), University of Illinois at Chicago, 845 West Taylor Street, Chicago, Illinois 60607-7061

Tamar Seideman
Steacie Institute for Molecular Sciences, National Research Council of Canada, Ottawa, Canada K1A-0R6
(Received 16 February 2000)

The Breit-Wigner phase of a wave function was obtained by measuring the interference between two independent ionization paths of a molecule. The state of interest was present in only one of the paths, thereby producing a phase shift in the observed signal. An analytical theory was used to determine the phase of the wave function from the observable.

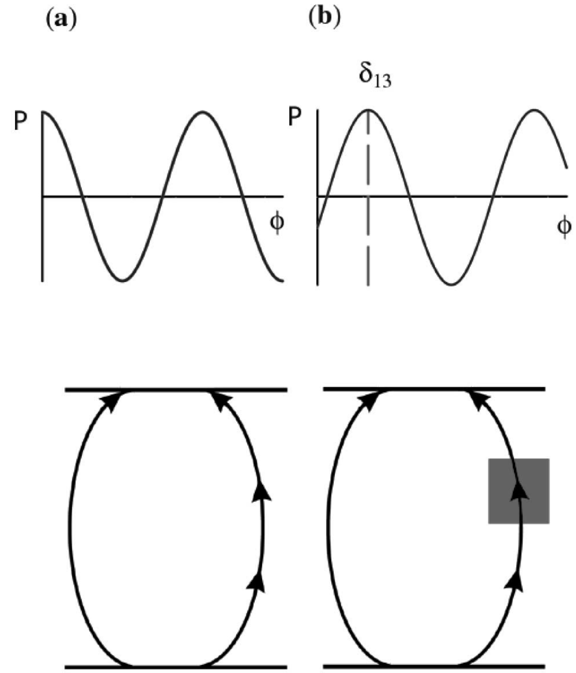


FIG. 1. Illustration of a molecular interferometer. Panel (a) shows that two competing quantum mechanical paths connecting the same initial and final states produce a sinusoidal variation of the product signal that depends on the relative phase of the two paths. In panel (b) an additional phase source is introduced at an intermediate (two-photon) level of the three-photon path. This source could be, for example, a predissociating resonance. The effect of this source is to produce a phase shift of δ_{13} in the signal.

general quantum interferometry

... pretty much any quantum mechanical process where *multiple paths* play a role.

For example, the phase of the free electron wavefunction(s) via multiple ionization paths:

VOLUME 69, NUMBER 16 PHYSICAL REVIEW LETTERS 19 OCTOBER 1992

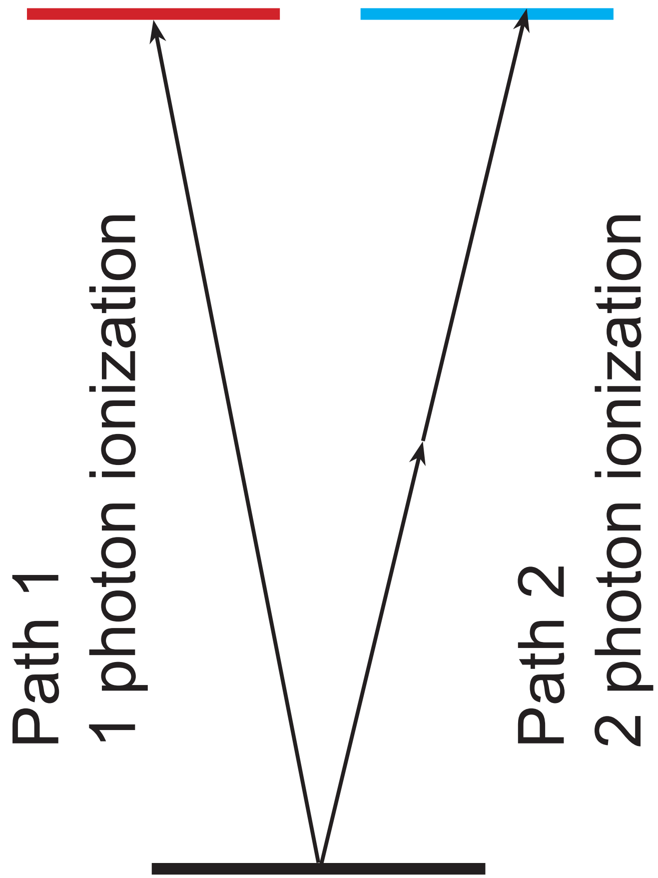
Asymmetric Photoelectron Angular Distributions from Interfering Photoionization Processes

Yi-Yian Yin, Ce Chen, and D. S. Elliott
School of Electrical Engineering, Purdue University, West Lafayette, Indiana 47907-1285

A. V. Smith
Sandia National Laboratories, Albuquerque, New Mexico 87185
(Received 14 May 1992)

We have measured asymmetric photoelectron angular distributions for atomic rubidium. Ionization is induced by a one-photon interaction with 280 nm light and by a two-photon interaction with 560 nm light. Interference between the even- and odd-parity free-electron wave functions allows us to control the direction of maximum electron flux by varying the relative phase of the two laser fields.

PACS numbers: 32.80.Fb, 32.80.Rm



... pretty much any quantum mechanical process where *multiple paths* play a role.

For example, the phase of free electron wavefunction(s).

VOLUME 69, NUMBER 16 PHYSICAL REVIEW LETTERS 19 OCTOBER 1992

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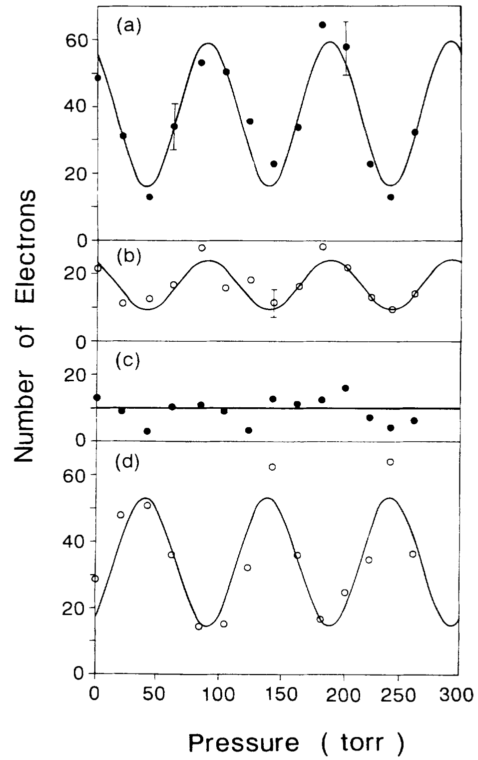

FIG. 3. Experimental data. The total electron count as a function of pressure of N_2 gas in the phase delay cell for the four detectors positioned at (a) 0°, (b) 45°, (c) 90°, and (d) 180°. The solid line is the result of a least-squares fit of a sinusoidally varying curve to the data.

Note - control via E-field phases

NRC·CNRC

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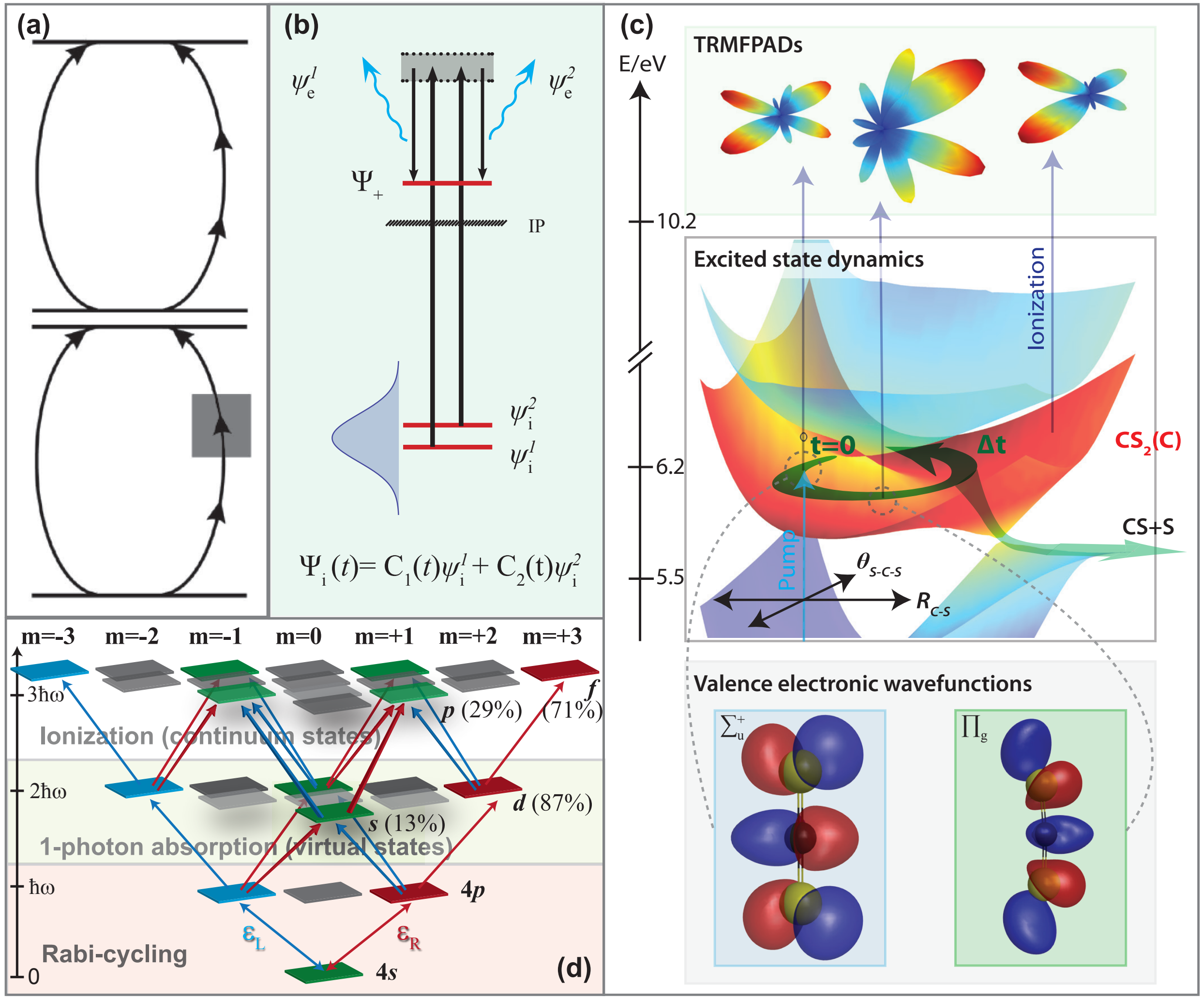
general quantum interferometry

... pretty much any quantum mechanical process where *multiple paths* play a role.



(a) from Fiss et. al., PRL 85 2096 2000

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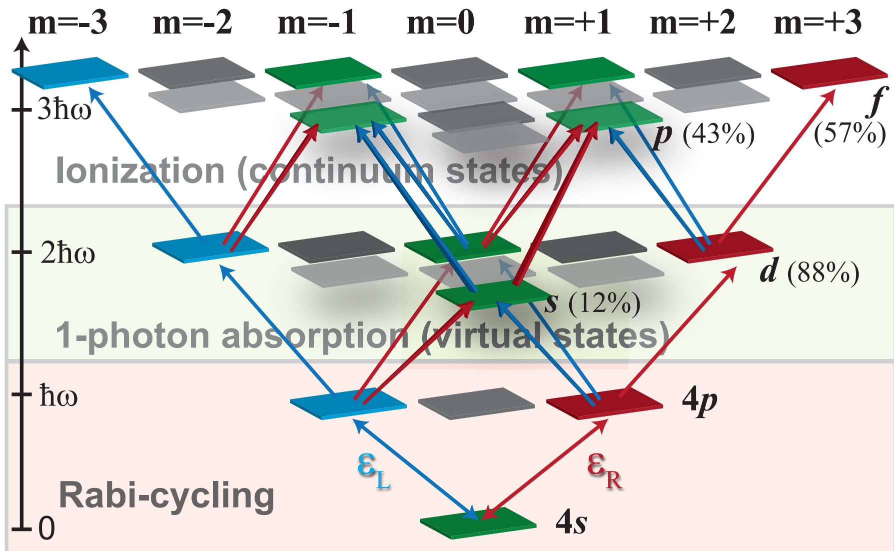
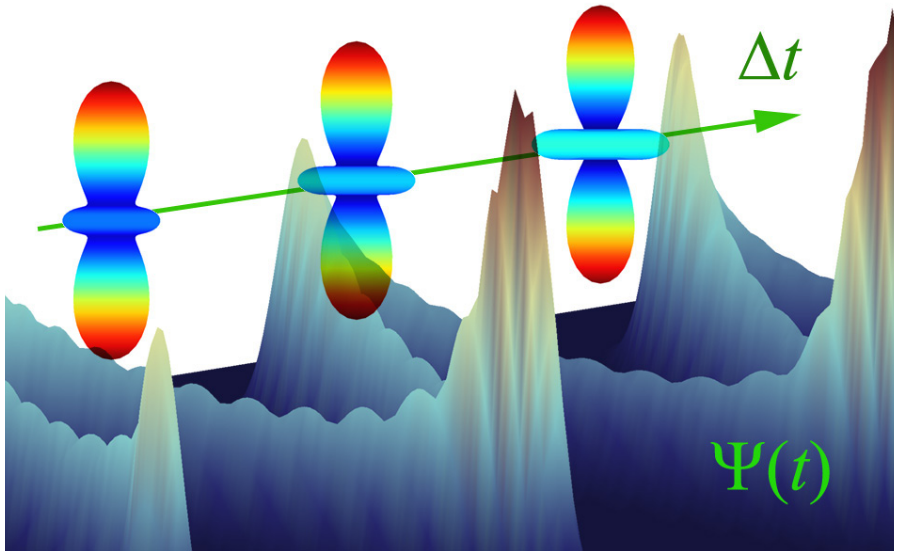
photoionization interferometry

Photoionization is an interferometric process, in which multiple paths can contribute to the final continuum photoelectron state.

(1) Intrinsic: interferences between final (continuum) states.



(2) Extrinsic/dynamic: additional pathways due to, e.g., prepared wavepacket, control fields, etc. etc.



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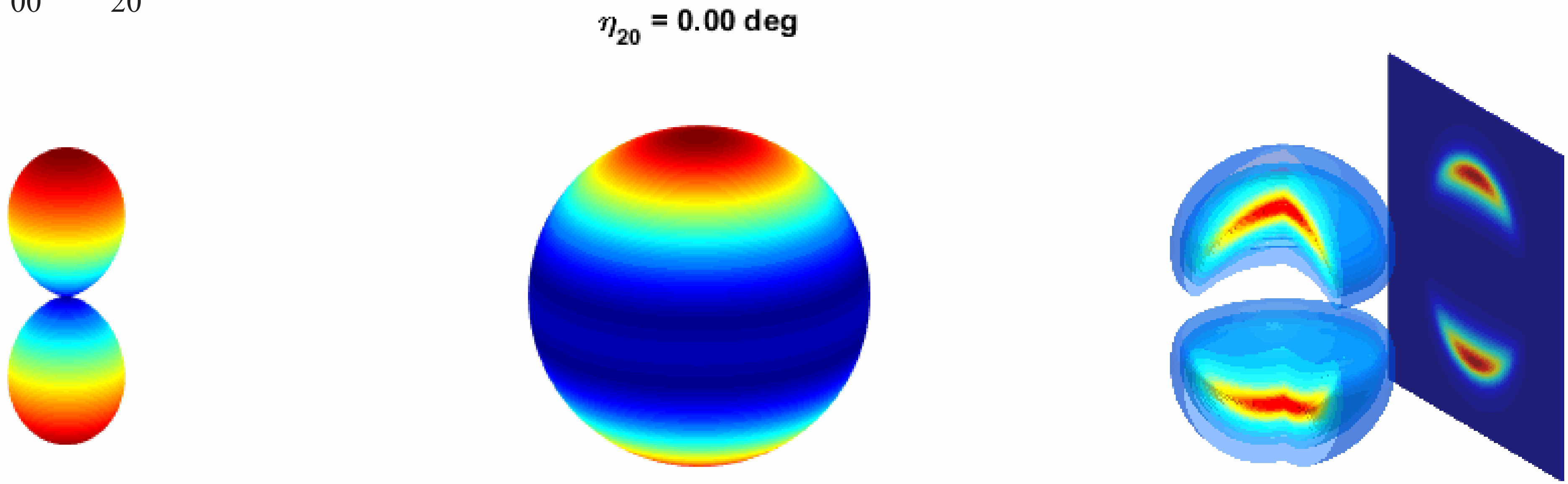
phase-sensitive photoelectron metrology

Need a phase-sensitive observable... photoelectron angular distributions (PADs) are angle-resolved photoelectron interferograms.

$$\psi_e = Y_{00} + Y_{20} e^{-i\eta}$$

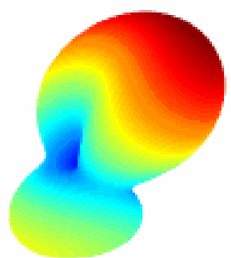


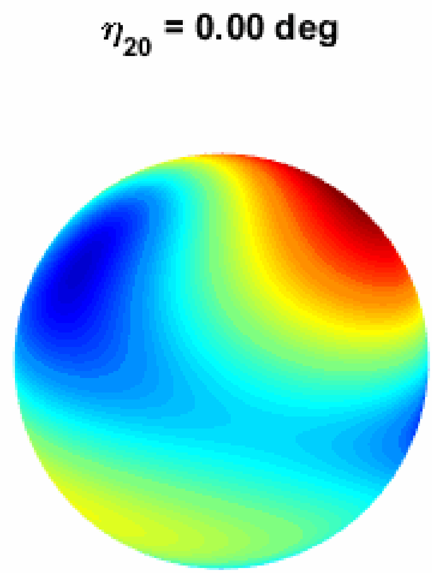
Although there are other factors, this illustrates why the PAD is so sensitive to the phase shifts - it is the interference due to these phase shifts which primarily determines the shape of the PAD.

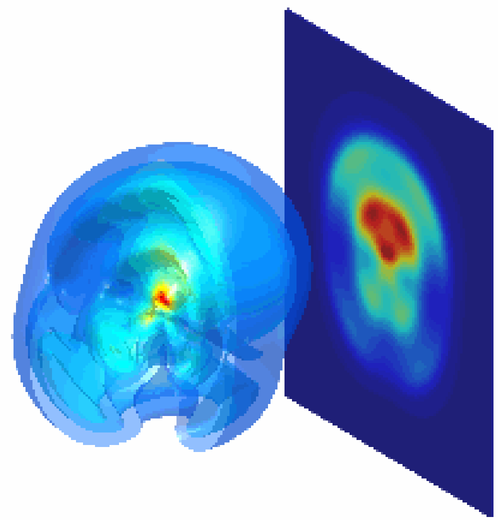
phase-sensitive photoelectron metrology

Need a phase-sensitive observable... photoelectron angular distributions (PADs) are angle-resolved photoelectron interferograms.

$$\psi_e = Y_{00} + Y_{10} e^{-i\eta} + Y_{21} e^{-3i\eta}$$







s+p(m=0)+d(m=1) waves, as a function of relative phase.

background - dipole matrix elements

Any measurement involving ionization projects the initial state wavefunction onto the ionization continuum - free electron + ion.

In the weak-field & dipole regime, this is described by the dipole matrix elements:

$$d = \langle \Psi_+;\ \psi_e | \hat{\mu}.\mathbf{E} | \Psi_i \rangle$$

Final state
Ion + electron

Dipole operator and incident field

Initial state

Observable - angle-resolved photoelectron flux:

$$I(\theta, \phi;\ E, t) = \langle \Psi_+;\ \psi_e | \hat{\mu}.\mathbf{E} | \Psi_i \rangle \langle \Psi_i | \hat{\mu}.\mathbf{E} | \Psi_+;\ \psi_e \rangle$$

$$= \sum_L \sum_M \beta_{LM}(E, t) Y_{LM}(\theta, \phi)$$

background - dipole matrix elements

By writing all the wavefunctions as products of radial & angular (geometric) parts, the cross-section can be written as:

$$I(\theta,\phi;E,t) = \sum_{l\lambda}\sum_{l'\lambda'} \gamma\, r_{l\lambda} r_{l'\lambda'} \cos(\eta_{l\lambda} - \eta_{l'\lambda'})\, Y_{l\lambda}(\theta,\phi)\, Y^{*}_{l'\lambda'}(\theta,\phi)$$

The summation shown here is over all angular momentum states of the free electron, hence the PAD is an ***(angular) interference pattern***.

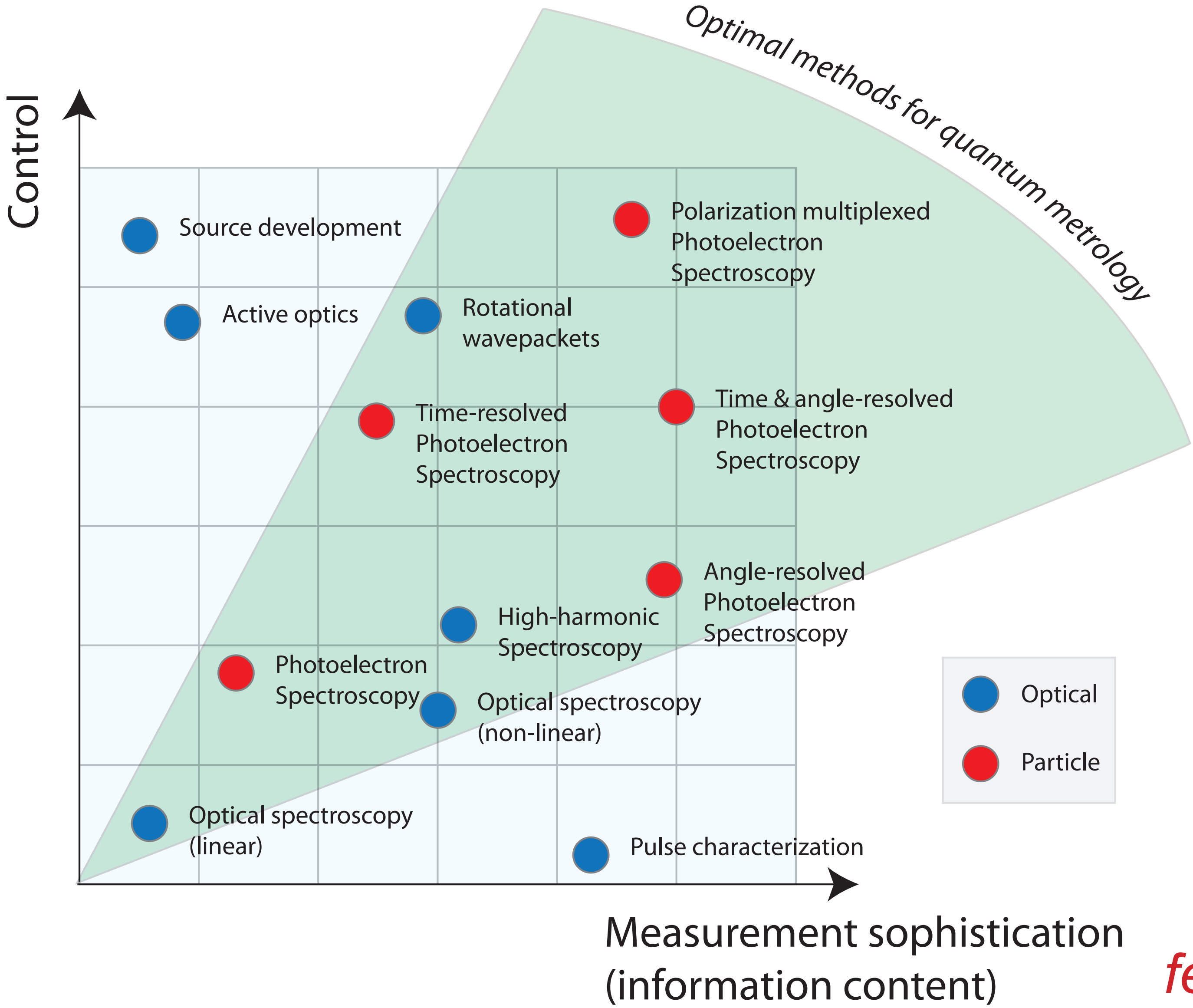
But... there may be many channels involved!

so…?

Isn't this just photoelectron spectroscopy?

Isn't this just photoelectron spectroscopy?

Absolutely... but with a focus on (quantitative) phase-sensitive metrology.



background - radial integrals

The geometric terms are analytic, so can be calculated directly, leaving only the radial integrals as unknowns. How can we treat these?

$$I(\theta,\phi;E,t) = \sum_{l\lambda}\sum_{l'\lambda'} \gamma\, r_{l\lambda} r_{l'\lambda'} \cos(\eta_{l\lambda} - \eta_{l'\lambda'})\, Y_{l\lambda}(\theta,\phi)\, Y^{*}_{l'\lambda'}(\theta,\phi)$$

Radial integrals ≡ scattering amplitudes & phases

Ab initio (numerical) scattering calculation

Extract from experimental data

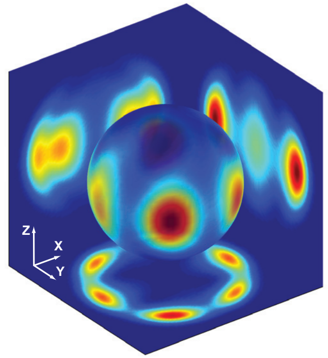
Quantitative methods

Symmetry based modelling

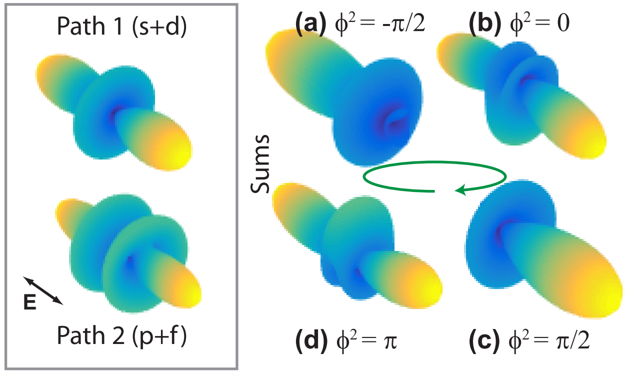
Qualitative methods

recent examples

Complete Photoionization Experiments via Ultrafast Coherent Control with Polarization Multiplexing



Hockett, P., Wollenhaupt, M., Lux, C., & Baumert, T.
Physical Review Letters, 112(22), 223001 (2014).
http://doi.org/10.1103/PhysRevLett.112.223001
arXiv 1403.3315 (https://arxiv.org/abs/1403.3315)



Angle-resolved RABBIT: theory and numerics

Paul Hockett
J. Phys. B (under review, 2017),
arXiv 1703.08586 (https://arxiv.org/abs/1703.08586).
Authorea https://dx.doi.org/10.22541/au.149037518.89916908

Bootstrapping to the Molecular Frame with Time-domain Photoionization Interferometry

Claude Marceau, Varun Makhija, Dominique Platzer, A. Yu. Naumov, P. B. Corkum, Albert Stolow, D. M. Villeneuve and Paul Hockett
Phys. Rev. Lett. (under review, 2017),
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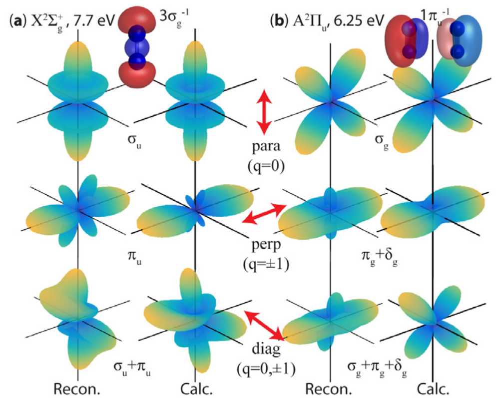

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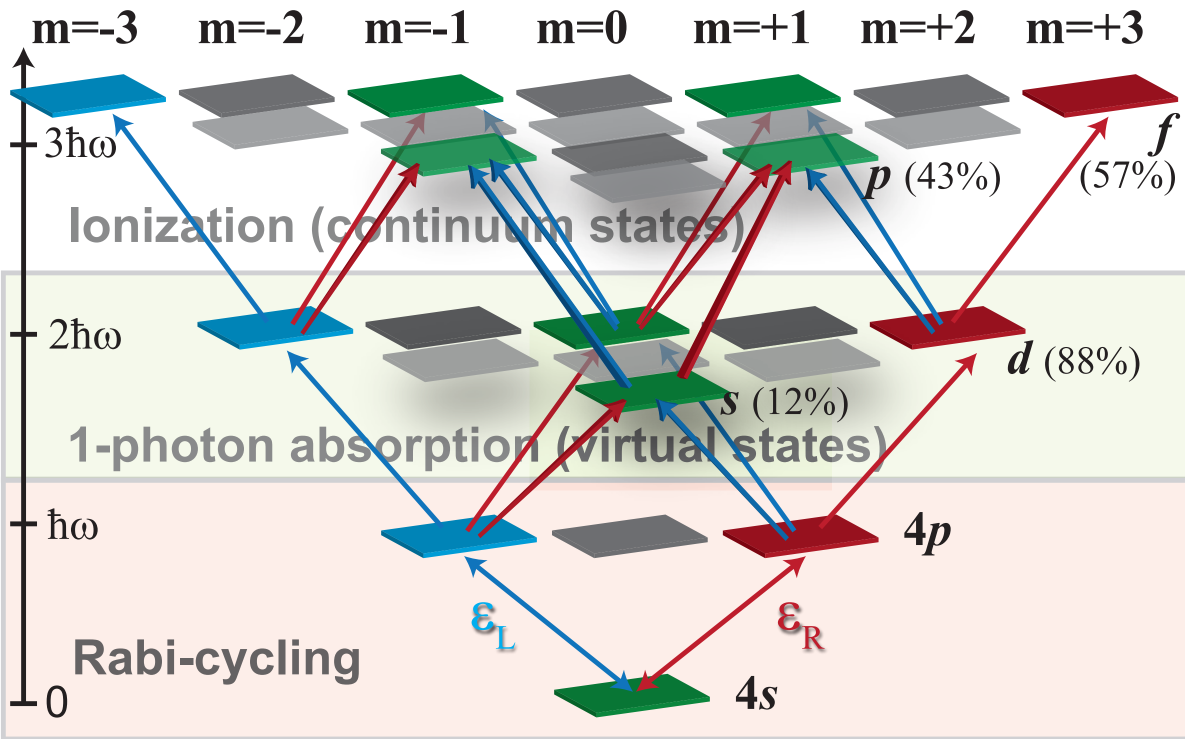
multi-path atomic interferometry

Modelling the dynamics & ionization for this 3-photon process.

Weak coupling
2-photon ionization

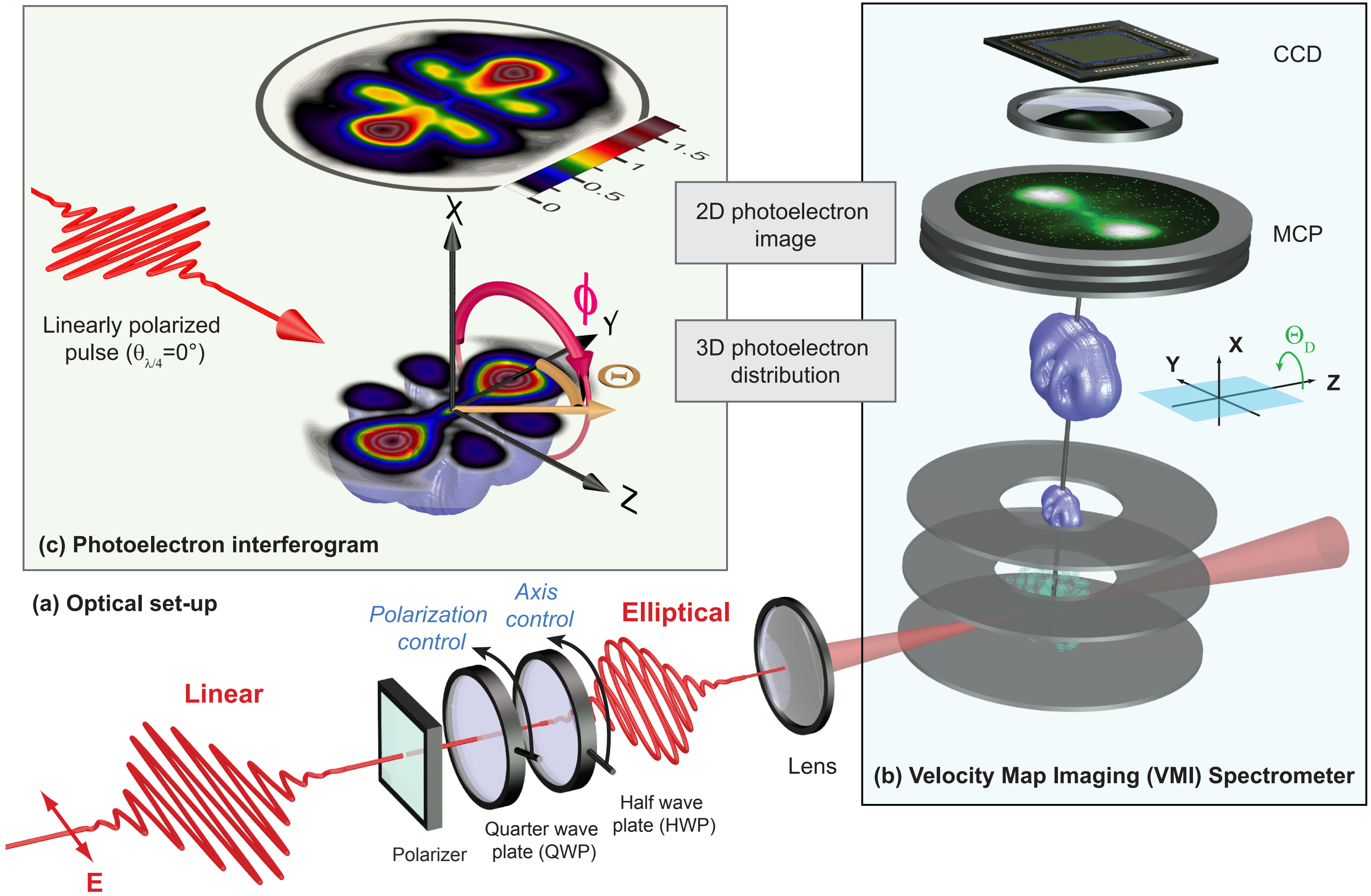
Strong-coupling
Intra-pulse dynamics



“Control” via interferences:
L vs R pathways to same final states.

multi-path atomic interferometry

Experiments (Christian Lux)



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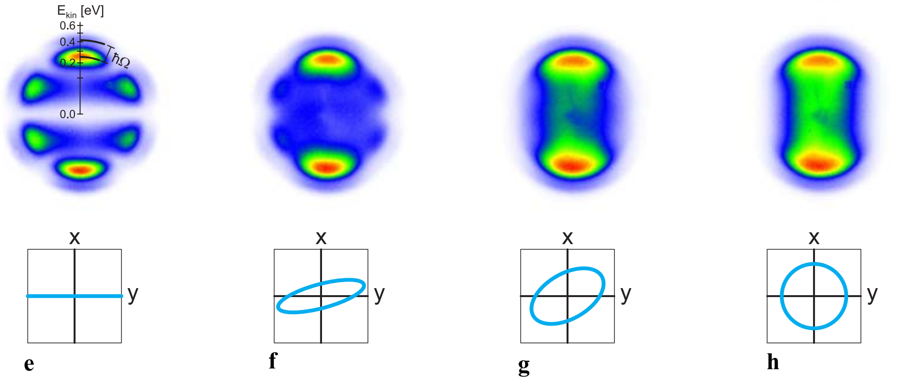
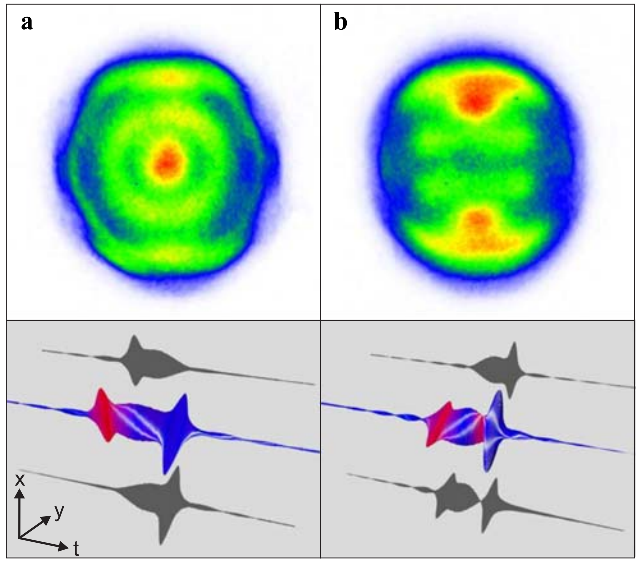
multi-path atomic interferometry

Experiments (Christian Lux)

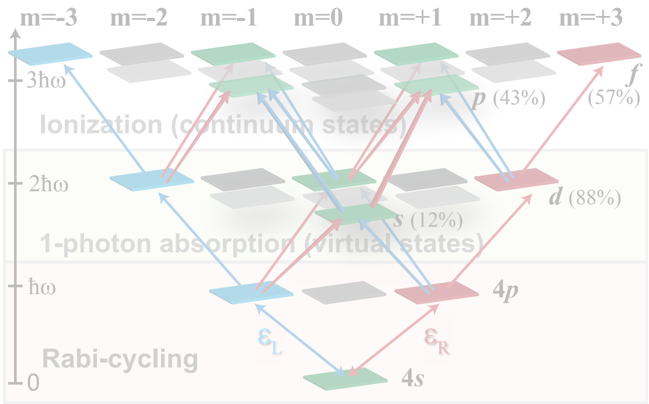
Ellipticity →



Control
Polarization-shaped pulses



multi-path atomic interferometry



2-photon ionization
Ionization dynamics

Rabi-cycling
Intra-pulse population dynamics

$$d_{l_f m_f} = \sum_{\substack{l_i, m_i \\ l_v, m_v \\ q, q'}} R_{l_v l_f}(k) \langle l_f m_f, 1q' | l_v m_v \rangle R_{l_i l_v}(k) \langle l_v m_v, 1q | l_i m_i \rangle \int E_{q'}(t) E_q(t) p_{m_i}(t) dt$$

Laser field
Polarization dynamics (control)

Radial ionization matrix elements

We can (try) fitting the data to determine these.
Complete experiment with intra-pulse dynamics.

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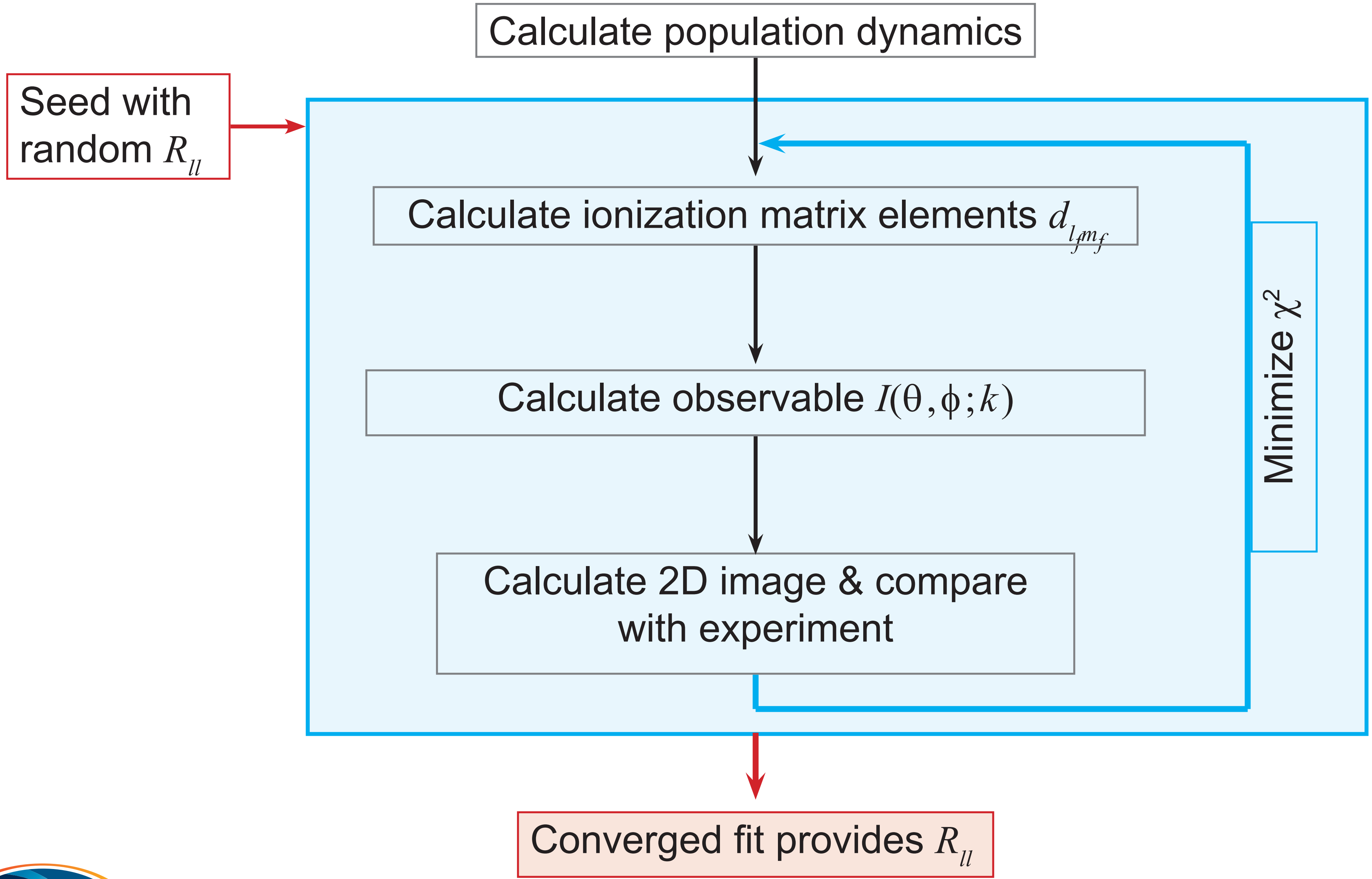
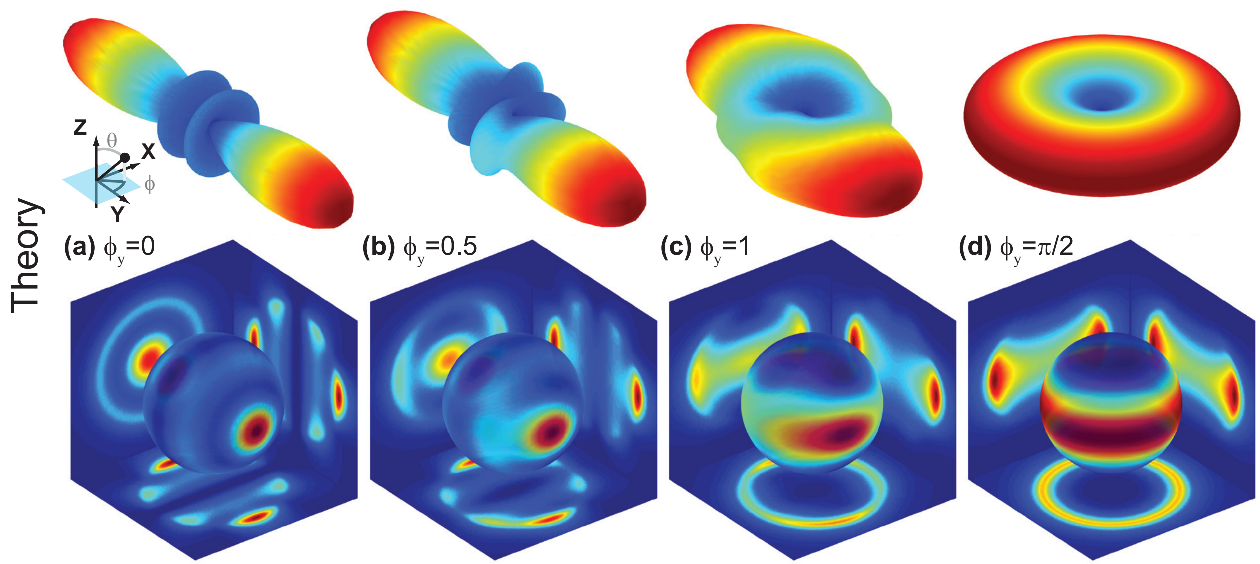
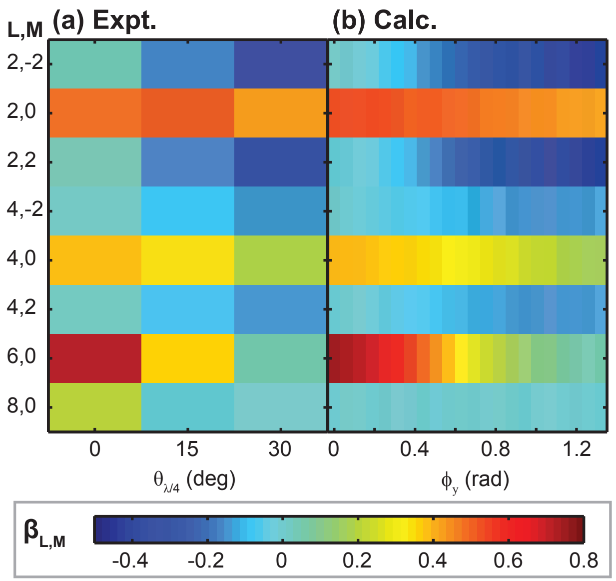
extracting the ionization dynamics

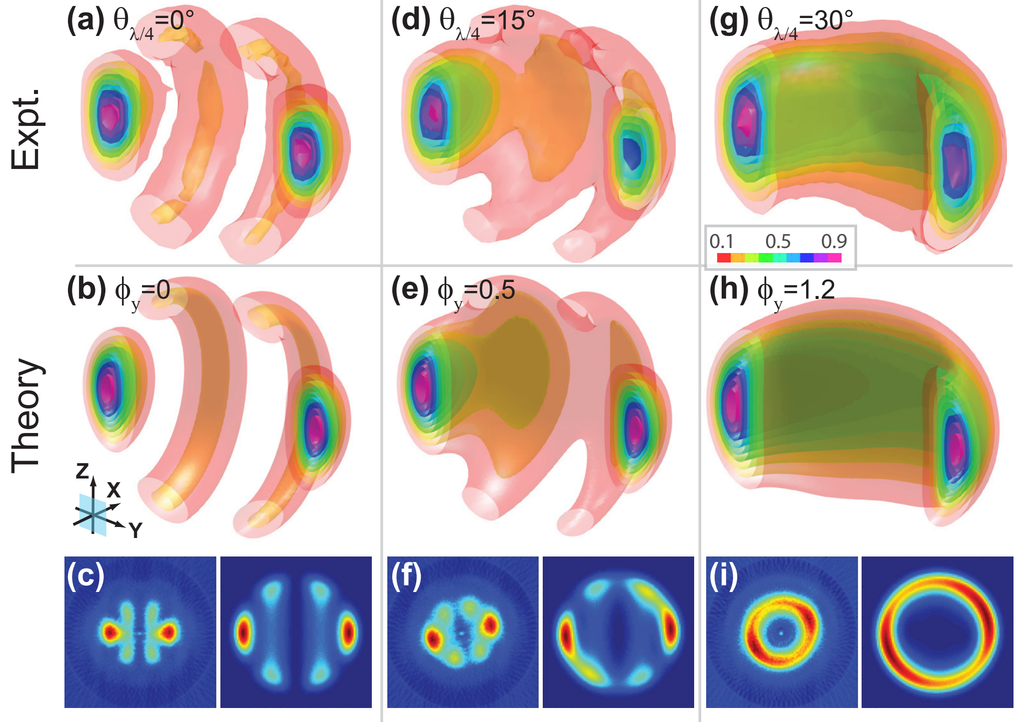


theory & experiment comparison



Theory - results based on the fitted matrix elements.
Expt. - tomographic (3D) experimental measurements.





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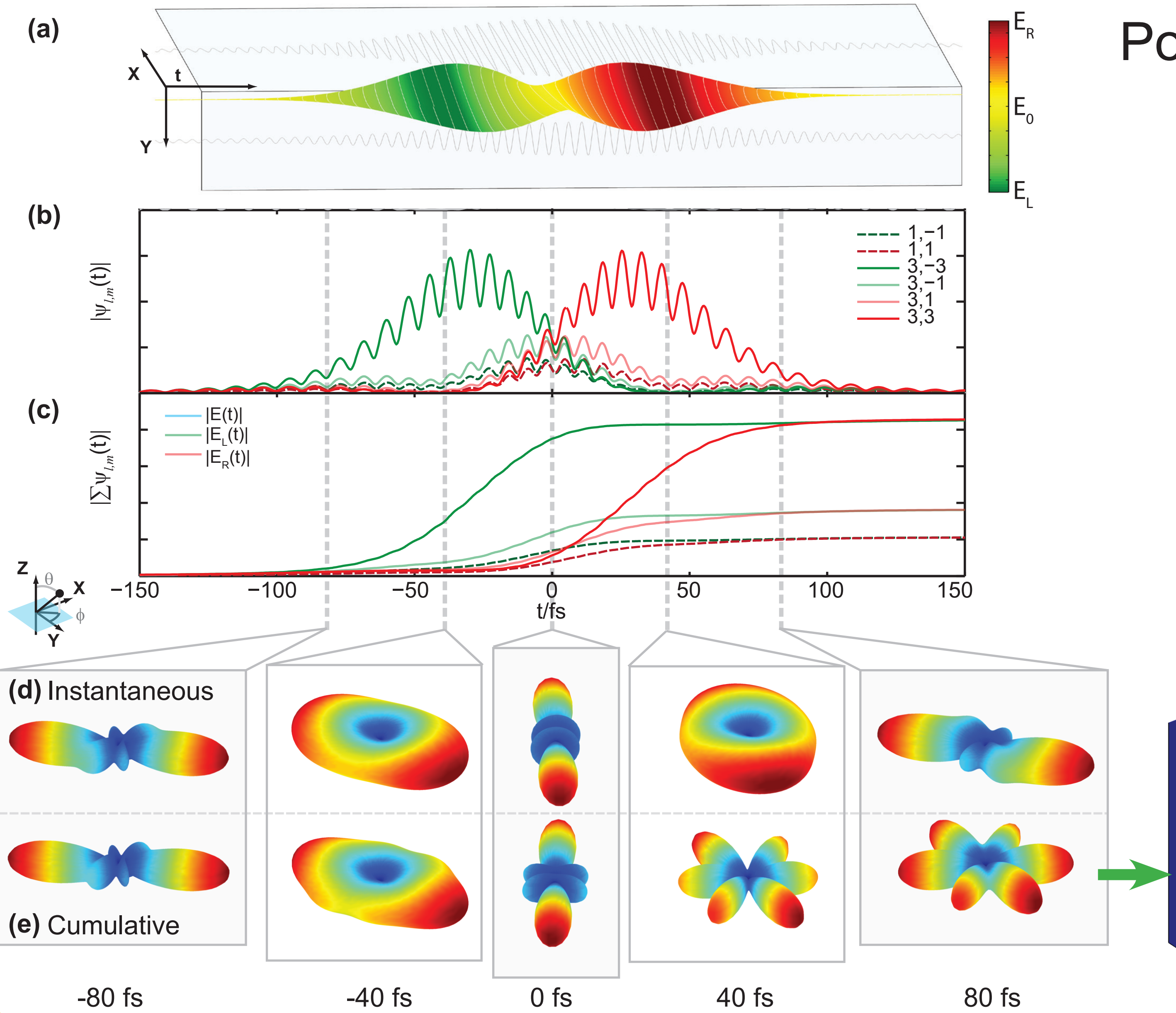
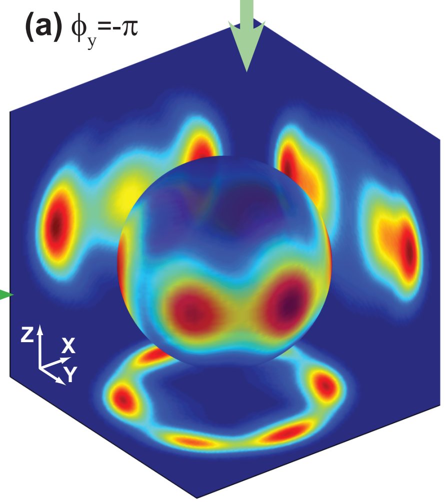
dynamics & control



Polarization-shaped laser pulse.

Intra-pulse dynamics.

Photoelectron interferograms.



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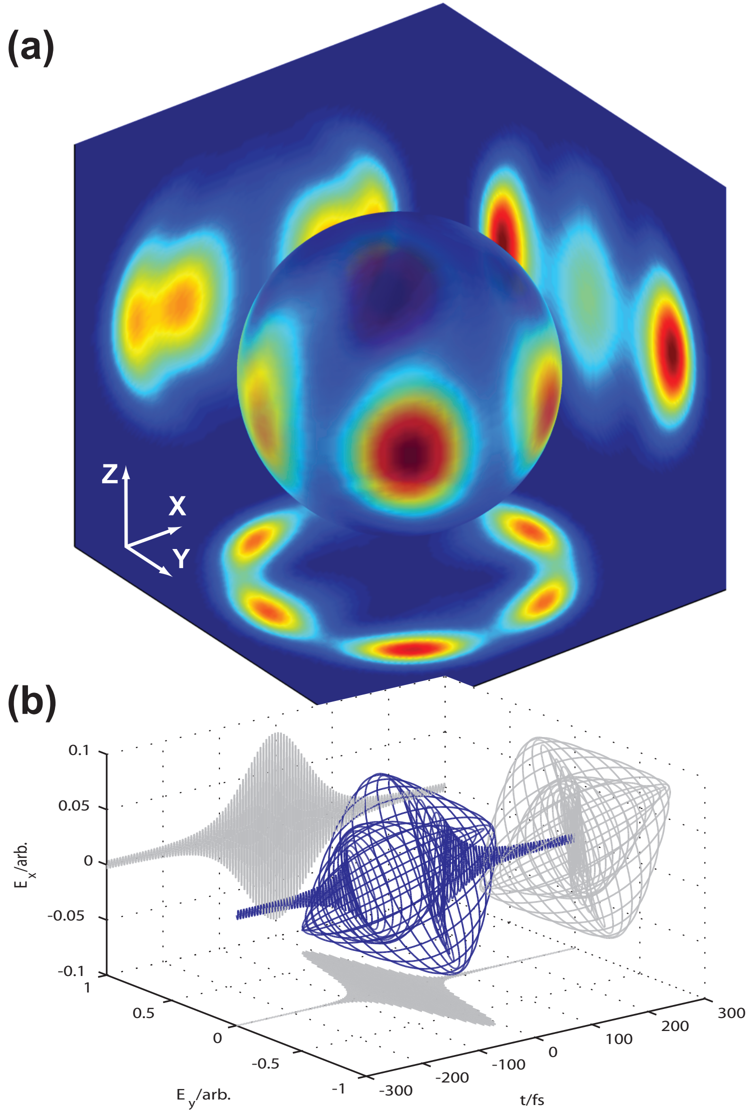
multi-path atomic interferometry

Conclusions & future work

- Modelling as 1-photon Rabi-cycling (*intra-pulse dynamics*), followed by 2-photon ionization, seems to be valid.
- 2D images provide a route to **extraction of the ionization matrix elements** ("complete photoionization experiment").
- For potassium we determine a dominant f-wave, with significant p-wave contribution.
- We can now model the **dynamics of polarization control**.
- *Fuller treatment of radial distributions to model, e.g., chirped laser pulse.*
- *3D data (tomographic measurements) should be more sensitive, and allow more robust fitting.*



for more...

Coherent Control of Photoelectron Wavepacket Angular Interferograms

Hockett, P., Wollenhaupt, M., Lux, C. & Baumert, T.
J. Phys. B, 48, 214004 (2015)
http://doi.org/10.1088/0953-4075/48/21/214004
arXiv 1505.00035

Maximum Information Photoelectron Metrology

Hockett, P., Lux, C., Wollenhaupt, M. & Baumert, T.
Phys. Rev. A, 92, 013412 (2015)
http://doi.org/10.1103/PhysRevA.92.013412
arXiv 1503.08308

Complete Photoionization Experiments via Ultrafast Coherent Control with Polarization Multiplexing II: Numerics & Analysis Methodologies

Hockett, P., Wollenhaupt, M., Lux, C. & Baumert, T.
Phys. Rev. A, 92, 013411 (2015)
http://doi.org/10.1103/PhysRevA.92.013411
arXiv 1503.08247

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P. Hockett, M. Wollenhaupt, C. Lux, T. Baumert
Physical Review Letters 112, 223001 (2014).
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arXiv 1403.3315

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recent examples

Complete Photoionization Experiments via Ultrafast Coherent Control with Polarization Multiplexing

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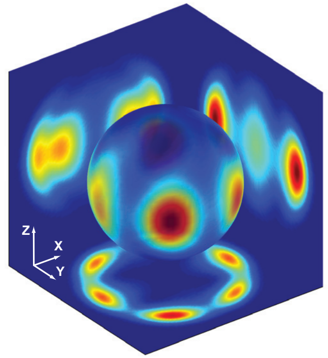

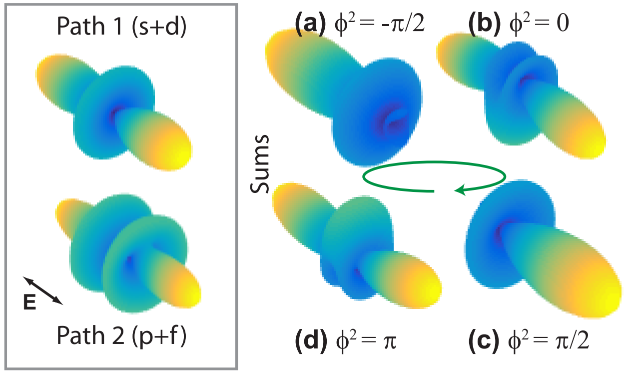

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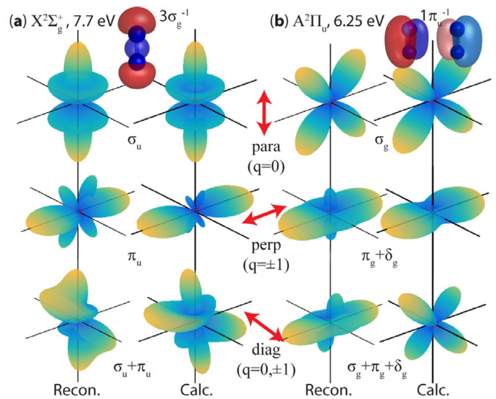

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RABBIT overview

Appl. Phys. B 74 [Suppl.], S17–S21 (2002)
DOI: 10.1007/s00340-002-0894-8

Applied Physics B
Lasers and Optics

H.G. MULLER

Reconstruction of attosecond harmonic beating by interference of two-photon transitions

FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

Received: 19 September 2001/
Revised version: 7 November 2001
Published online: 5 July 2002 •

ABSTRACT A method is proposed for detailed determination of the temporal structure of XUV pulses. The method is especially suited for diagnostics on attosecond pulses and pulse trains that originate from temporal beating of various harmonics of an ultrashort laser pulse. A recent experiment already showed the feasibility of this method when applied to long attosecond pulse trains, where it measured the average pulse characteristics. Here we argue that the same method is also suitable for determining differences between the individual attosecond pulses in a short train, or the properties of a single attosecond pulse.

PACS 32.80.Rn; 42.30.Rx

and leave the other pulse unchanged, since the light required for up-conversion by sum-frequency mixing would map in the far or mid infrared. It is much preferable to up-convert both pulses by mixing with an optical photon of slightly different, precisely defined frequency. A convenient way to obtain the narrow-band photons required for the up-conversion process is by selecting different portions of a strongly chirped version of the pulse under study. This pulse is guaranteed to have enough bandwidth to generate a suitable spectral shear Ω.

The major limitation of SPIDER is that to measure pulses that are far from their bandwidth limit, the spectral sampling has to be rather dense (i.e. small Ω). To make a frequency that is sufficiently constant over the (long) duration of the original pulse, one has to chirp out the up-converting pulse so far that it might not have enough intensity left. In cases

Muller, H. G. (2002). Reconstruction of attosecond harmonic beating by interference of two-photon transitions. Applied Physics B: Lasers and Optics, 74, 17–21. http://doi.org/10.1007/s00340-002-0894-8

RABBIT overview

J. Phys. B: At. Mol. Opt. Phys. **45** (2012) 183001

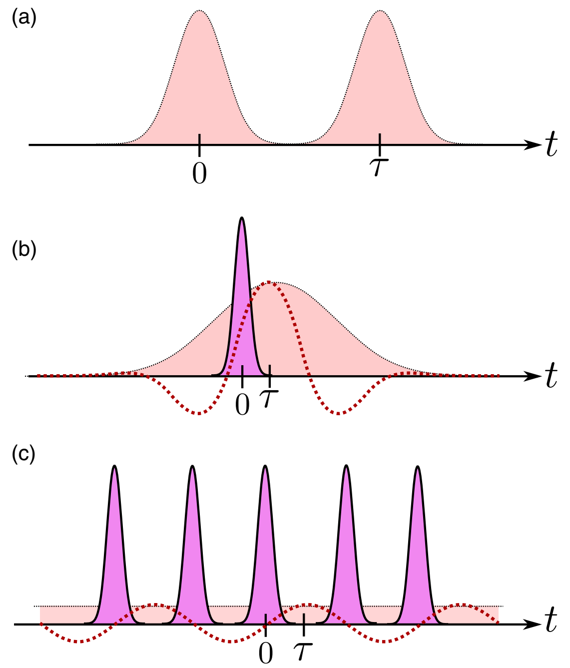


Figure 4. Pump–probe schemes. (a) Traditional pump–probe experiment with two pulses separated in time by τ. (b) Simultaneous pump–probe experiment between a SAP and a few-cycle IR field. (c) Simultaneous pump–probe experiment between an APT and a monochromatic IR field. The narrow purple area represents the attosecond XUV pulse envelope and the broader red area represents the one of the probing laser pulse, while the dotted red lines indicate the corresponding E-field.

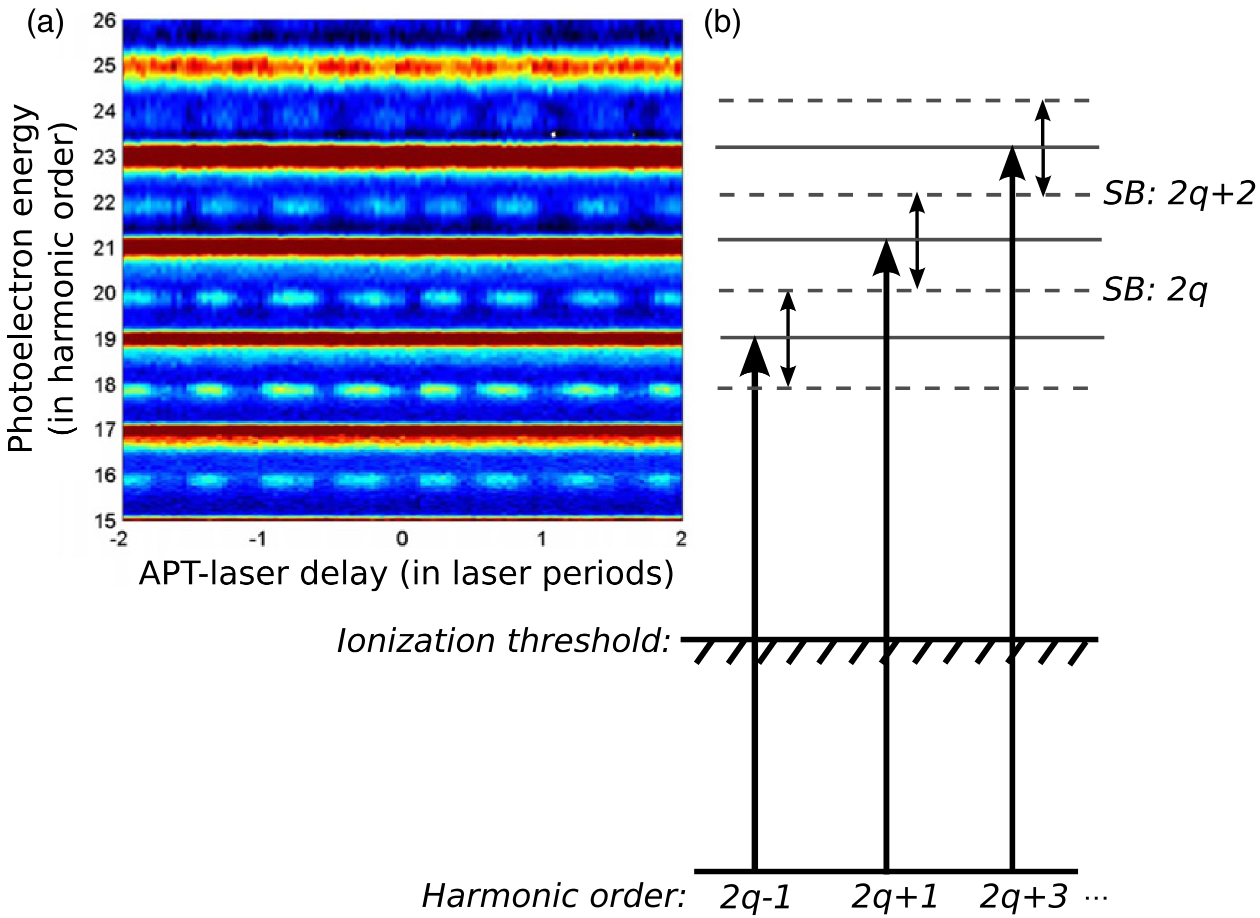
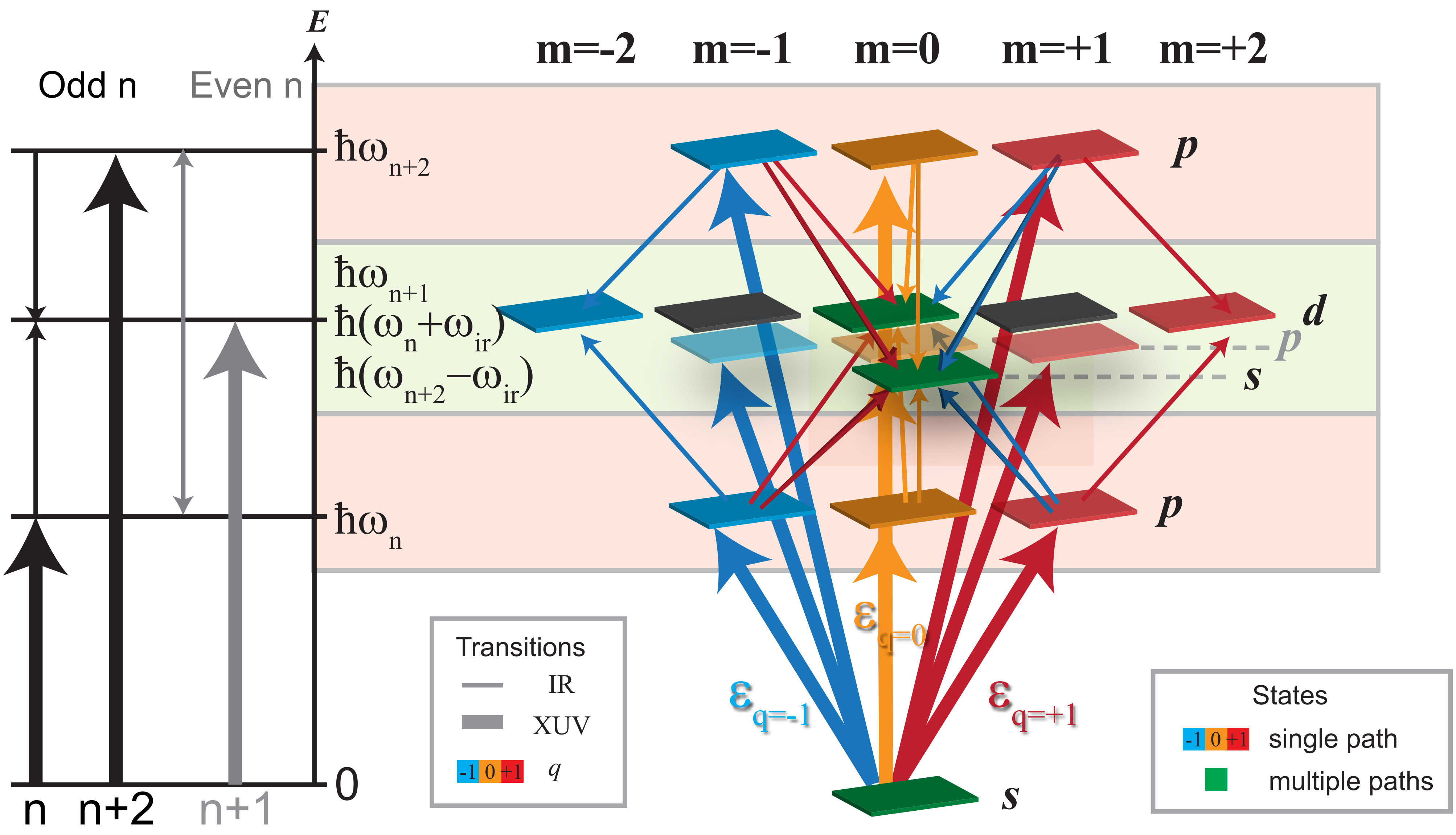


Figure 5. RABITT method. (a) Photoelectron spectrogram over photon energy and delay between the APT and the IR field. The offset in the modulation of the SBs contains information about the attosecond pulses and the ionization process. (b) Schematic energy diagram over the quantum paths leading to the same final energy in SB $2q$. The experimental data were gathered from [56].

Dahlström, J. M., L'Huillier, A., & Maquet, A. (2012). Introduction to attosecond delays in photoionization. Journal of Physics B: Atomic, Molecular and Optical Physics, 45(18), 183001. http://doi.org/10.1088/0953-4075/45/18/183001

RABBIT in detail



Each channel contains multiple paths and phases.

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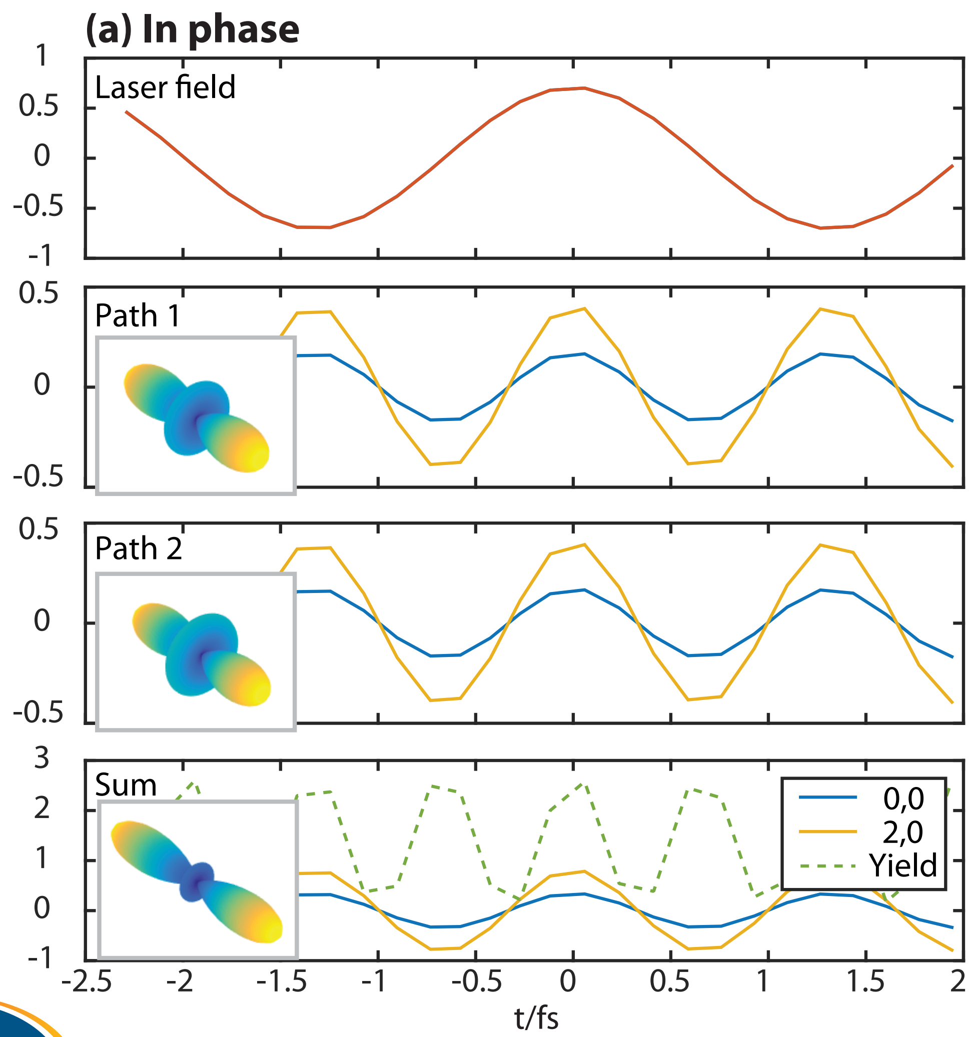
modelling AR-RABBIT

Channels: $s \xrightarrow{xuv} p \xrightarrow{ir} s{+}d.$

To obtain maximum information, angular resolution is preferable.



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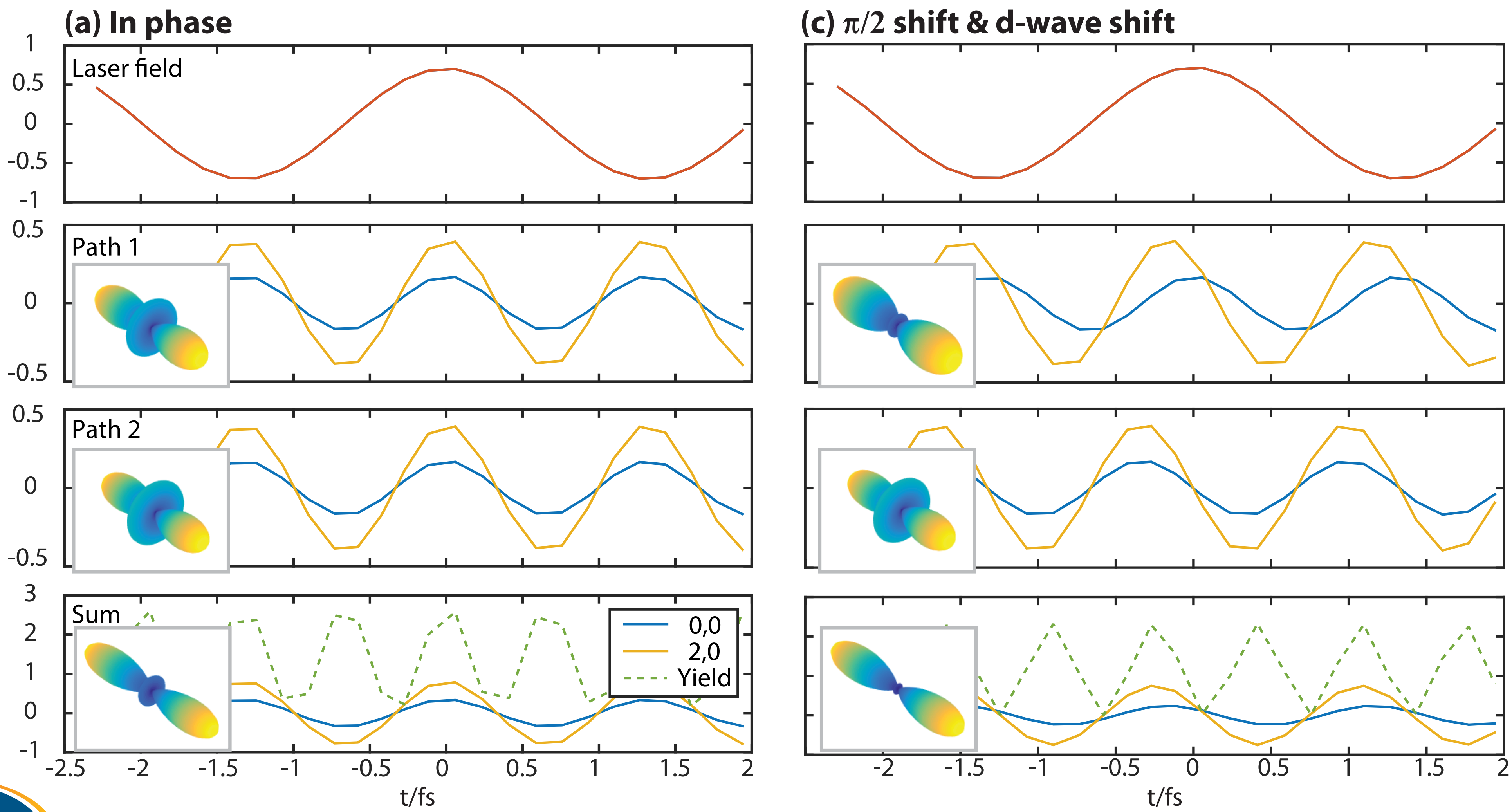
modelling AR-RABBIT

Channels: s $\xrightarrow{xuv}$ p $\xrightarrow{ir}$ s+d.

To obtain maximum information, angular resolution is preferable.



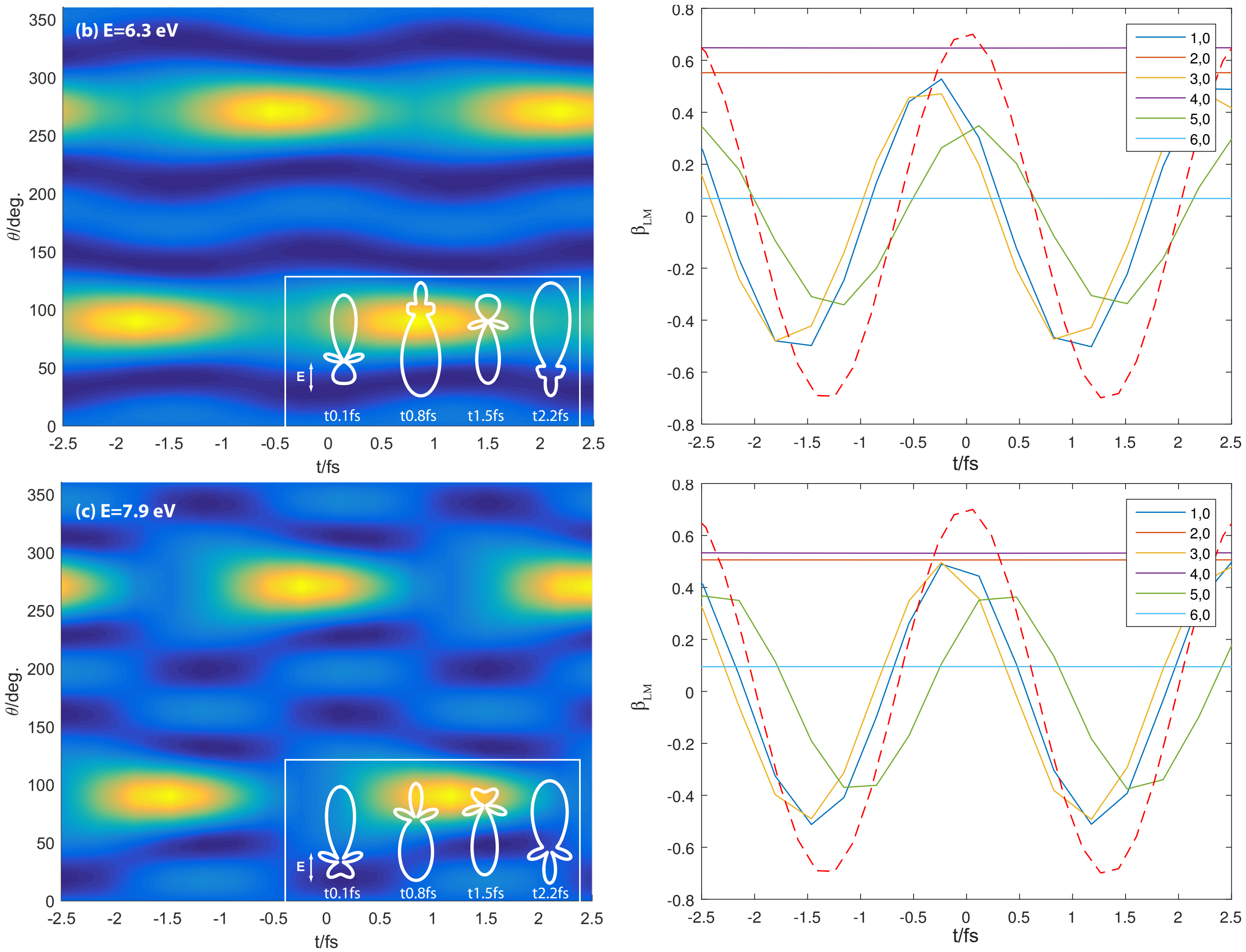
Realistic numerical model:

- Bound-free (xuv) matrix elements from ePolyScat. http://www.chem.tamu.edu/rgroup/lucchese/ePolyScat.E3.manual/manual.html
- Continuum-continuum (ir) matrix elements using Coulomb scattering solutions (cf. treatment by Dahlström et. al).

Dahlström, J. M., L'Huillier, A., & Maquet, A. (2012). Introduction to attosecond delays in photoionization. Journal of Physics B: Atomic, Molecular and Optical Physics, 45(18), 183001. http://doi.org/10.1088/0953-4075/45/18/183001

numerical results

AR-RABBIT bands (odd + even harmonics), I(θ,τ)



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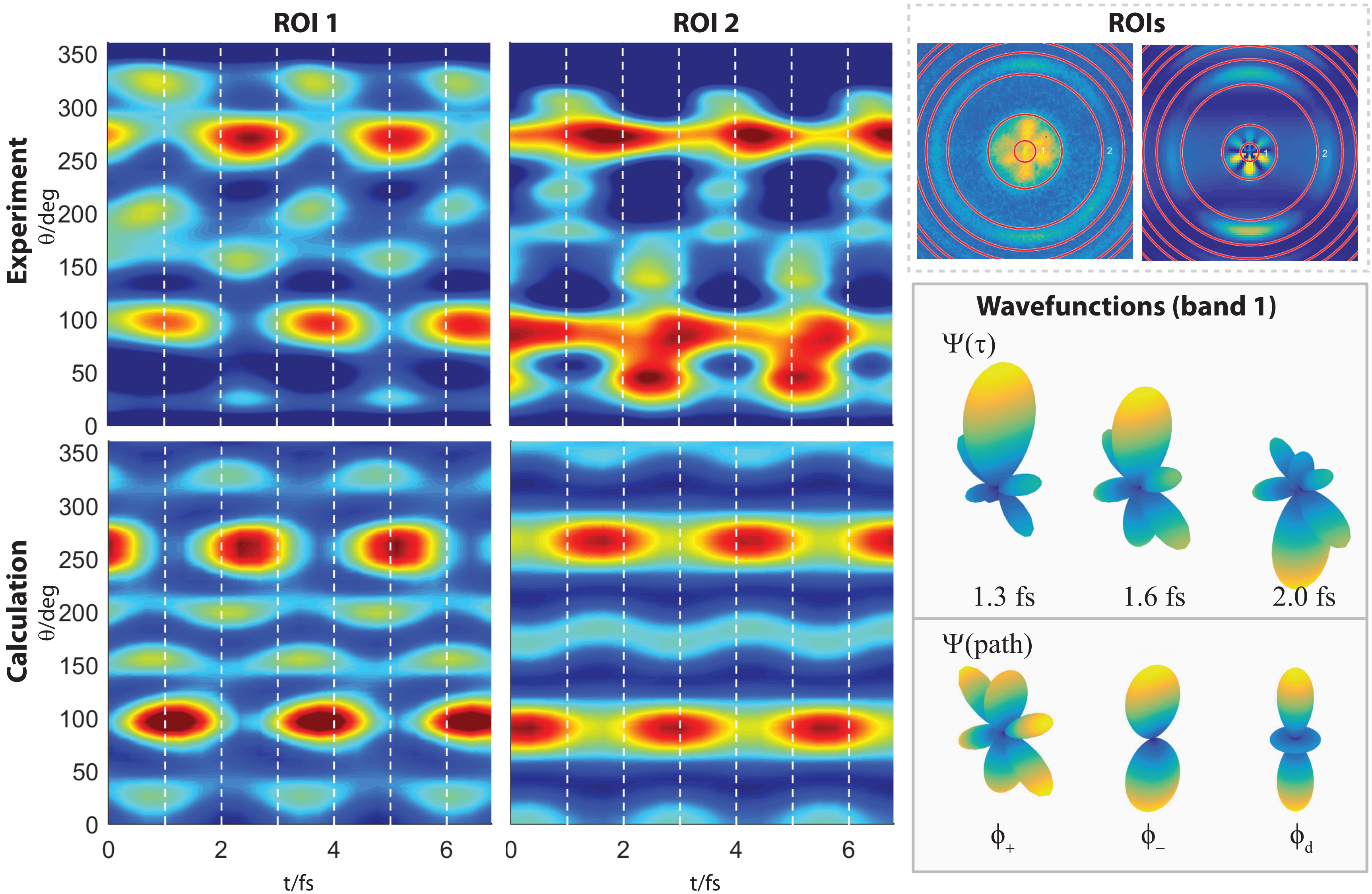
VMI simulation

AR-RABBIT VMI simulation, and comparison with experiments (2 bands).



Experimental results - Hiromichi Niikura (*Science*, in press, 2017)
Model results also include calculated XUV phases - David Villeneuve, SFA calculations

summary

AR-RABBIT is a little bit complex, but is an information rich measurement...

It is suitable for both pulse and photoelectron metrology, and control.

Numerical modelling using established photoionization techniques reproduces the expected phenomena, and yields detailed understanding.

The numerical modelling techniques are general (any atom or molecule).*

(Preliminary) comparisons with experimental results (Hiromichi Niikura**) look promising...

* although the method used here is expected to be poor at low (near threshold) photoelectron energies due to the form of the continuum-continuum functions. Strong field effects are neglected.

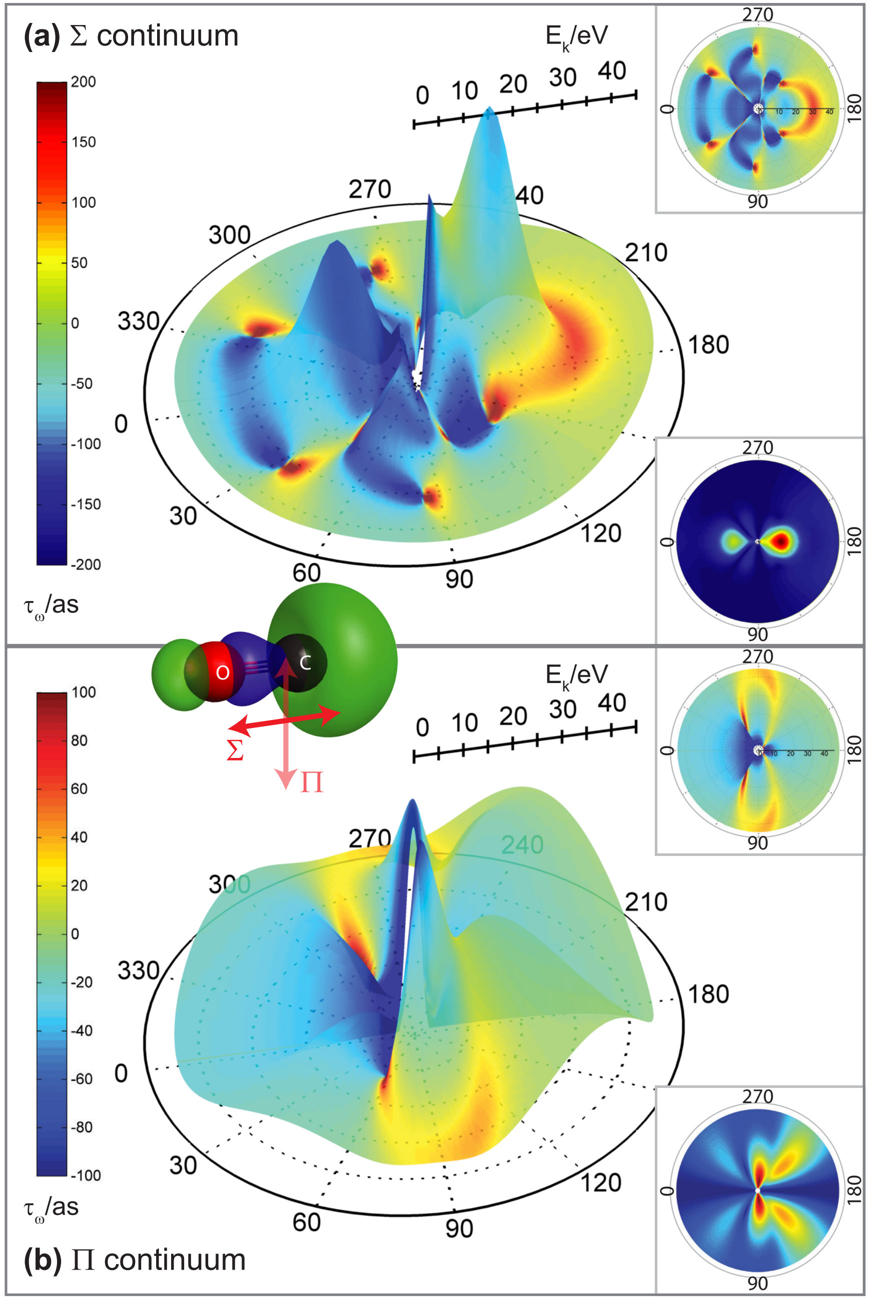
** *Coherent Imaging of an Attosecond Electron Wave Packet*, D M Villeneuve, P Hockett, M J J Vrakking and H Niikura, Science (in press, 2017)

AR-RABBIT for Wigner delays

For molecules the continuum phase, hence Wigner (photoionization) delay, is a complex function of energy and angle.

AR-RABBIT is one potential method for mapping this phase-dependence.

See:
Time delay in molecular photoionization
P Hockett, E Frumker, D M Villeneuve and P B Corkum, *J. Phys. B: At. Mol. Opt. Phys. 49, 095602, 2016.*
http://dx.doi.org/10.1088/0953-4075/49/9/095602
arXiv 1512.03788



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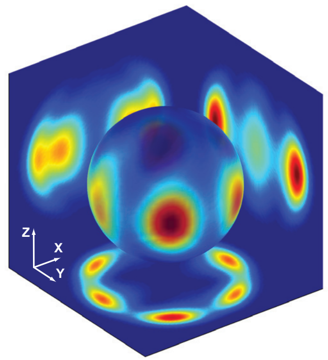
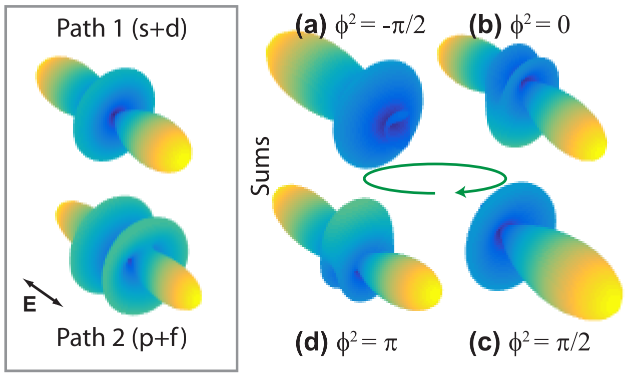
recent examples

Complete Photoionization Experiments via Ultrafast Coherent Control with Polarization Multiplexing

Hockett, P., Wollenhaupt, M., Lux, C., & Baumert, T.
Physical Review Letters, 112(22), 223001 (2014).
http://doi.org/10.1103/PhysRevLett.112.223001

arXiv 1403.3315 (https://arxiv.org/abs/1403.3315)





Angle-resolved RABBIT: theory and numerics

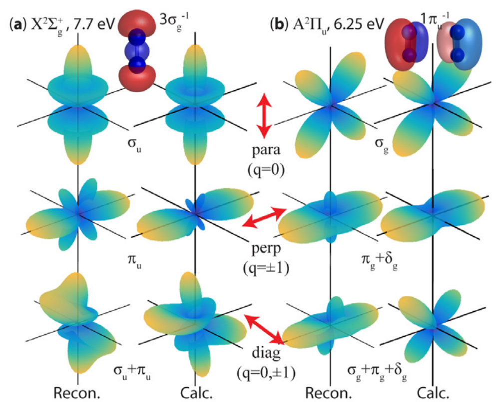
Paul Hockett
J. Phys. B (under review, 2017),
arXiv 1703.08586 (https://arxiv.org/abs/1703.08586).

Authorea https://dx.doi.org/10.22541/au.149037518.89916908

Bootstrapping to the Molecular Frame with Time-domain Photoionization Interferometry

Claude Marceau, Varun Makhija, Dominique Platzer, A. Yu. Naumov, P. B. Corkum, Albert Stolow, D. M. Villeneuve and Paul Hockett
Phys. Rev. Lett. (under review, 2017),

arXiv 1701.08432 (https://arxiv.org/abs/1701.08432).



towards quantitative imaging...

Experimental PADs combined with detailed analysis & theory offer the potential for a move beyond current phenomenological time-resolved imaging techniques by utilizing the photoelectron interferometer.

towards quantitative imaging...

Experimental PADs combined with detailed analysis & theory offer the potential for a move beyond current phenomenological time-resolved imaging techniques by utilizing the photoelectron interferometer.

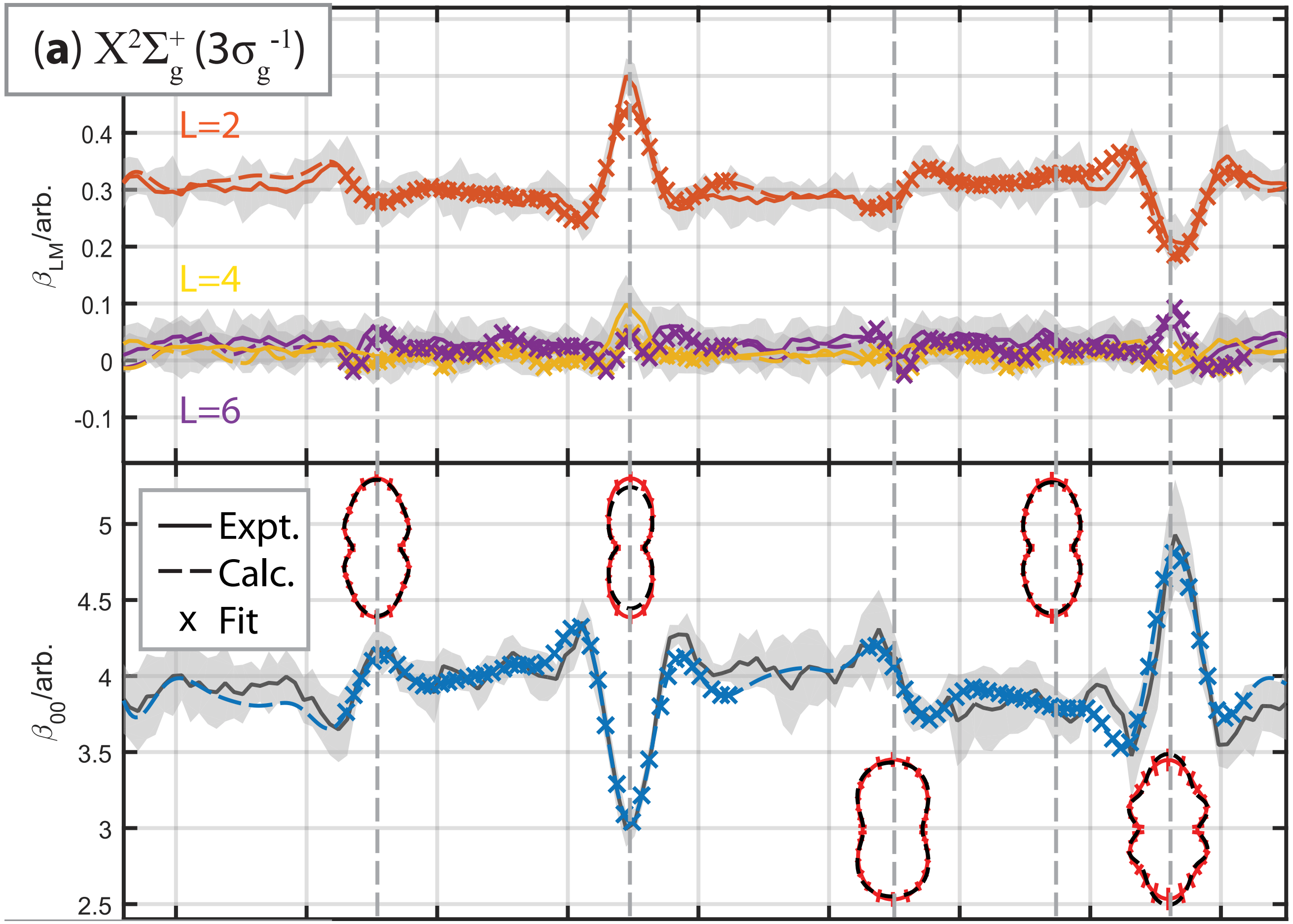
The examples discussed so far show some of this potential...

To proceed, we can consider "maximum information" experimental measurements, which allow for determination of the partial waves as a function of time.

towards quantitative imaging...

One example is the use of impulsive alignment techniques (rotational wavepackets).

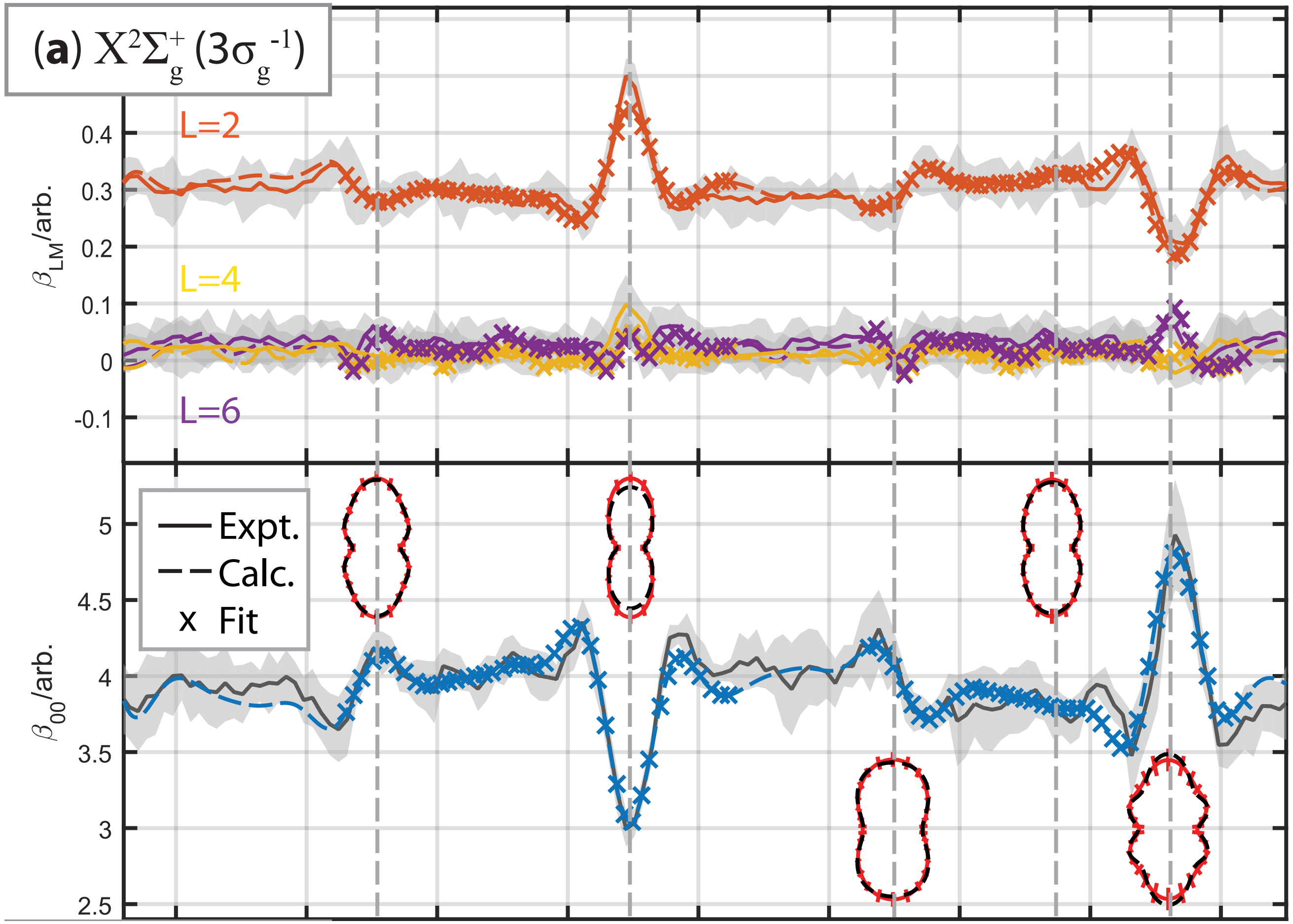
Align-probe angle-resolved measurements from N_2 ($h\nu$=23.3eV).



rotational wavepacket interferometry

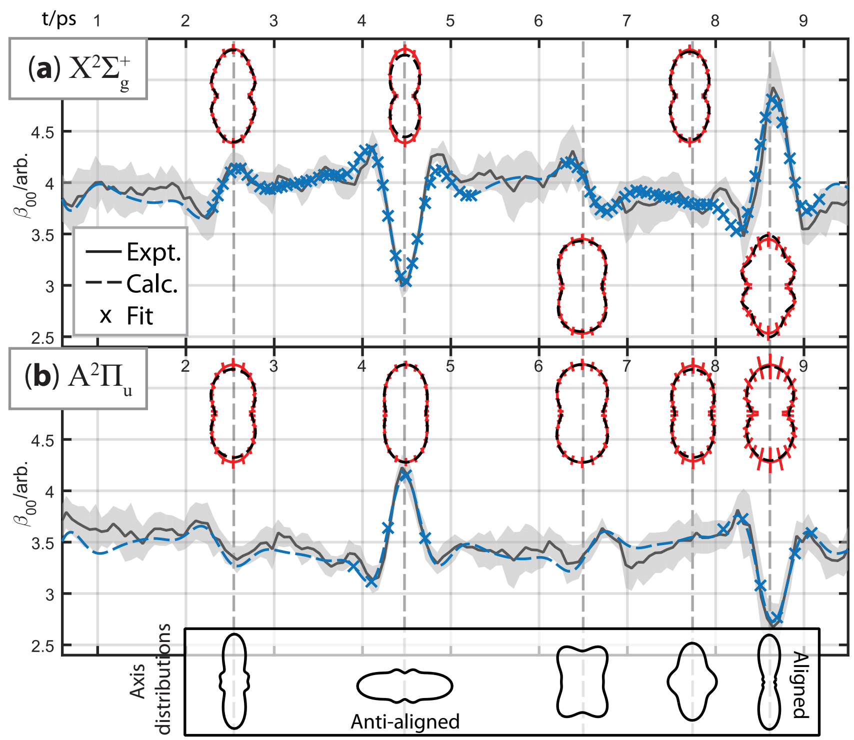
Fitting such data as a function of alignment can provide the ionization matrix elements and phases.

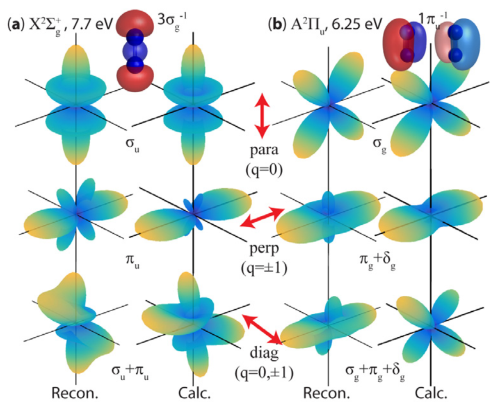
Essentially, the rotational wavepacket acts as a geometric contribution to the interferometer.



molecular frame reconstruction

For N_2, this has been demonstrated for matrix element retrieval for three different final ion states, and verified via molecular frame reconstruction & comparison with theory (ePolyScat).





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Phys. Rev. Lett. (under review, 2017),

arXiv 1701.08432 (https://arxiv.org/abs/1701.08432).

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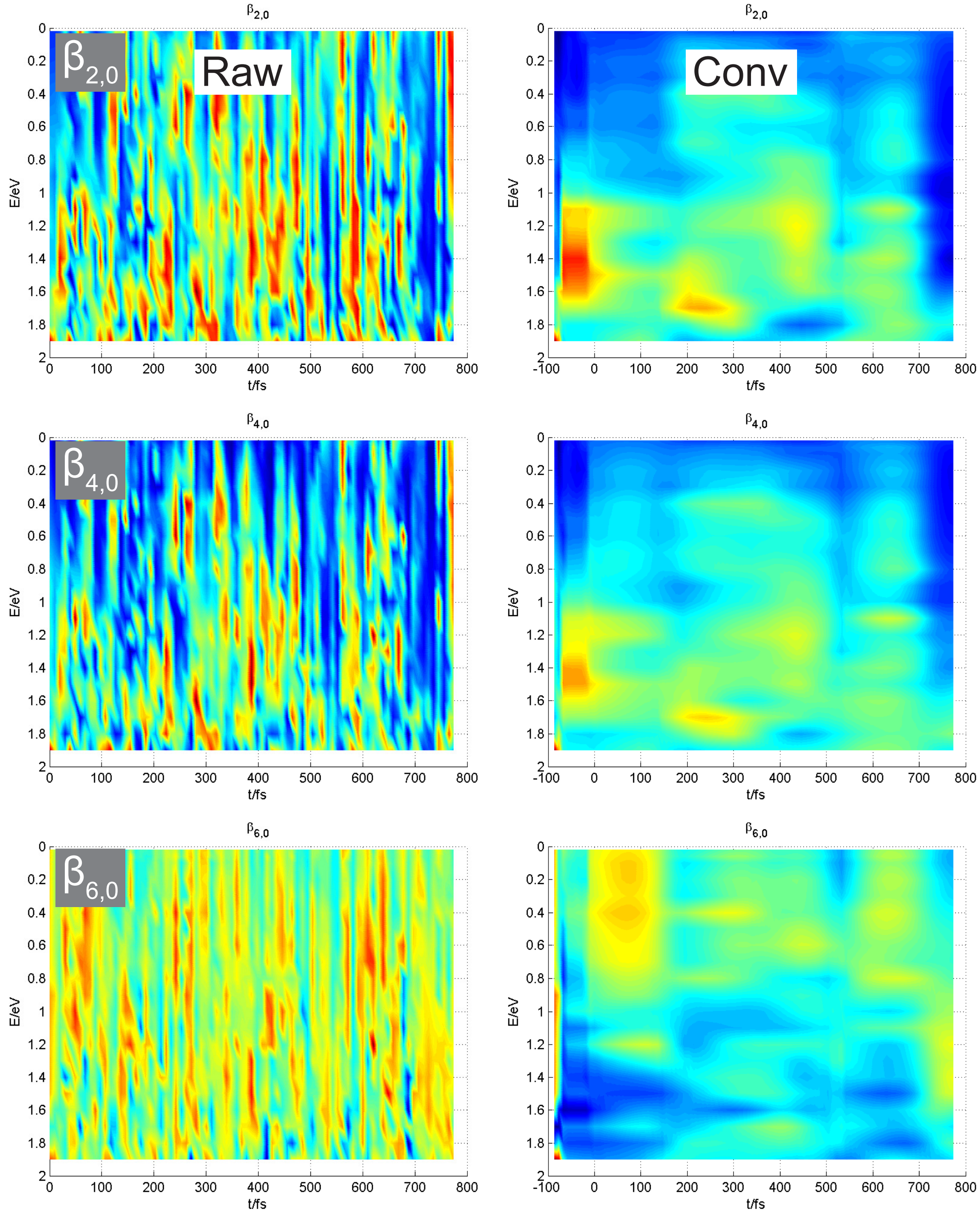
quantitative molecular dynamics

Most generally, we can look at the full $\beta_{LM}(E,t)$ spectra...

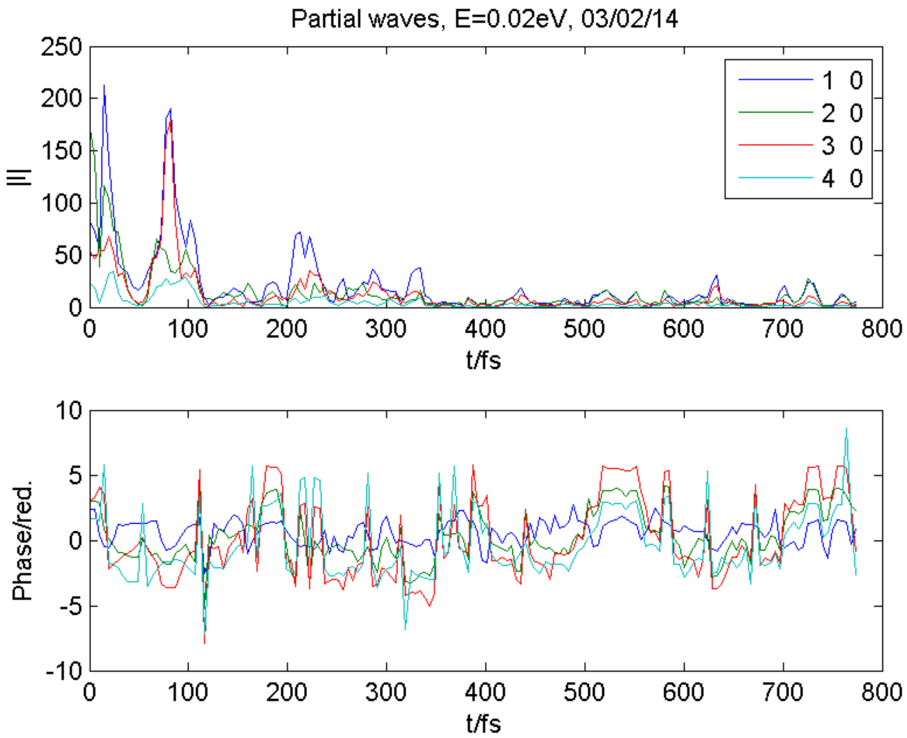


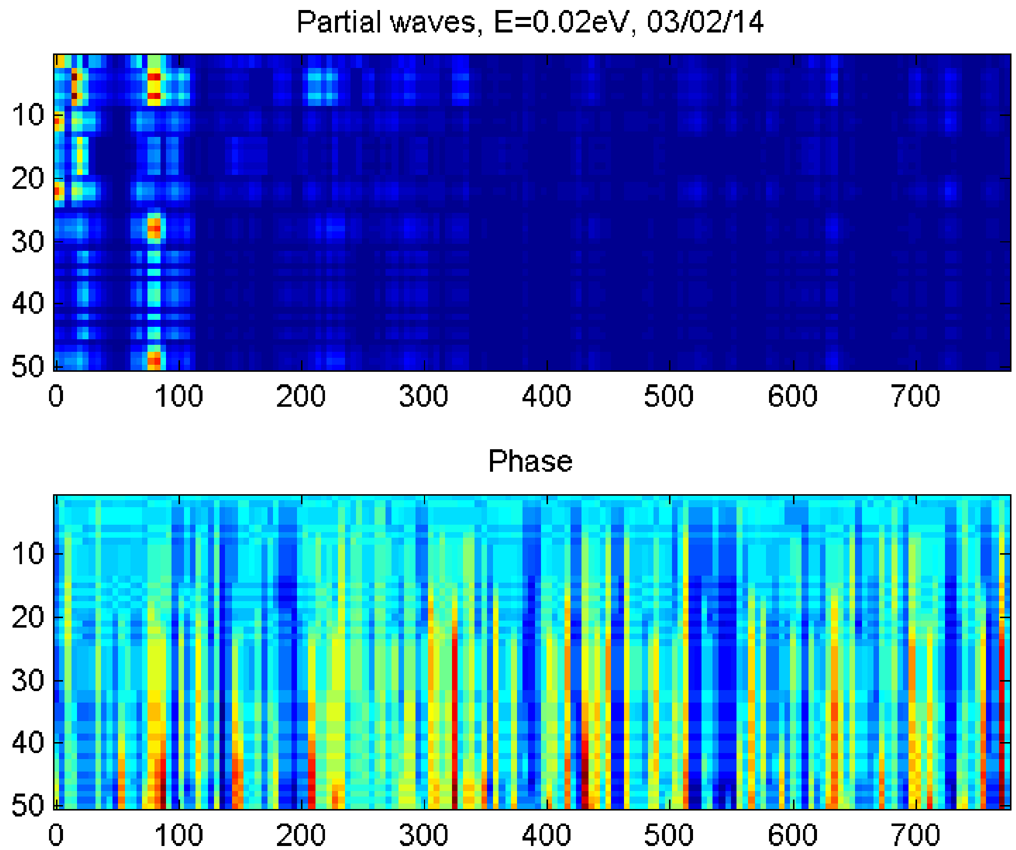
Full excited state molecular dynamics & observable calculations for CS_2.

quantitative molecular dynamics

Most generally, we can look at the full $\beta_{LM}(E,t)$ spectra...

... and the underlying partial wave amplitudes and phases.





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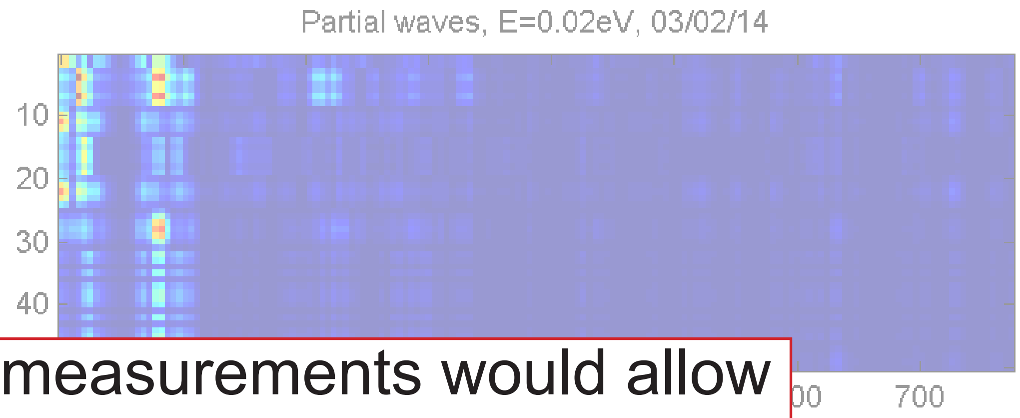
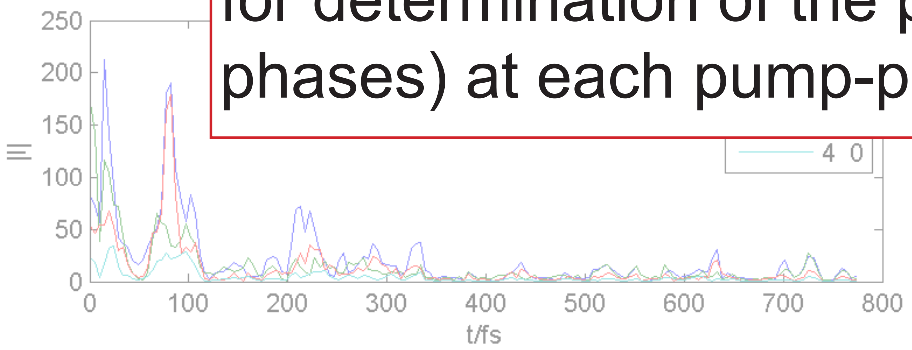
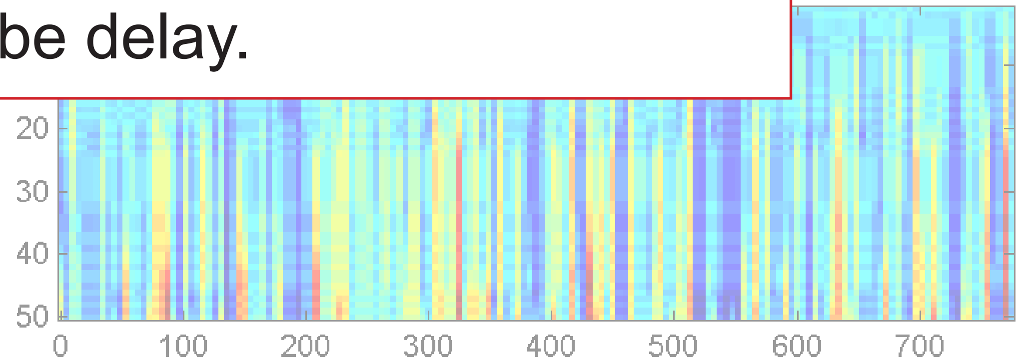
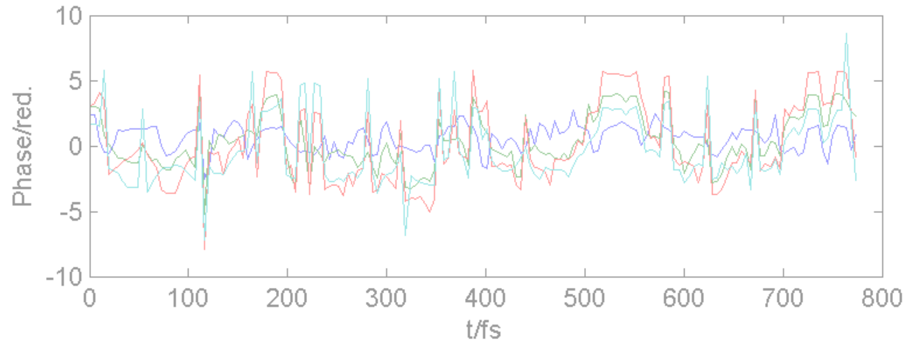


A set of align-pump-probe measurements would allow for determination of the partial waves (amplitudes & phases) at each pump-probe delay.







towards quantitative imaging...

Other examples of “maximum information” measurements include tomographic imaging, multi-path ionization schemes and complex light-matter interactions.

Quantum dynamical imaging

The tools are now in place, we just need to use them!

Quantum Dynamical Imaging via Time-resolved Photoelectron Interferometry:
Beyond a Phenomenological Imaging of Molecular Dynamics
P. Hockett (research proposal, 2013)
Available on Figshare, https://dx.doi.org/10.6084/m9.figshare.3580734

acknowledgements

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Kassel University, Germany
Christian Lux
Matthias Wollenhaupt
Thomas Baumert

University of Nottingham, UK
Katharine Reid

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where are we


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Ottawa, ON, Canada

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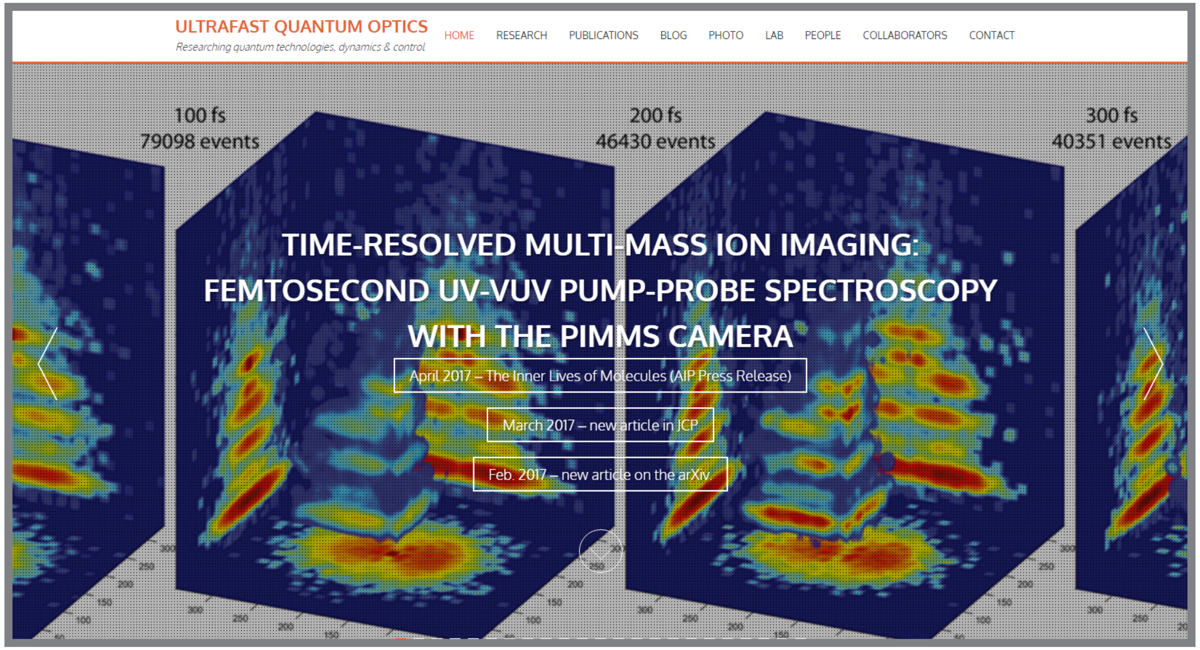
We're always interested in new collaborations and new directions...

If you have an idea, or work that could benefit from our expertise and facilities...

...please get in touch!



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for more information...

Slides available via Figshare, DOI: 10.6084/m9.figshare.5049142

arXiv: http://arxiv.org/a/hockett_p_1.html
Figshare: http://figshare.com/authors/Paul_Hockett/100955

Orcid: http://orcid.org/0000-0001-9561-8433
Scholar: https://scholar.google.ca/citations?user=e4FgTYMAAAAJ&hl=en

Web: www.femtolab.ca

Coming soon:
Quantum Metrology with Photoelectrons [working title]
New book for the IOP Concise series, due 2018

paul.hockett@nrc.ca

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