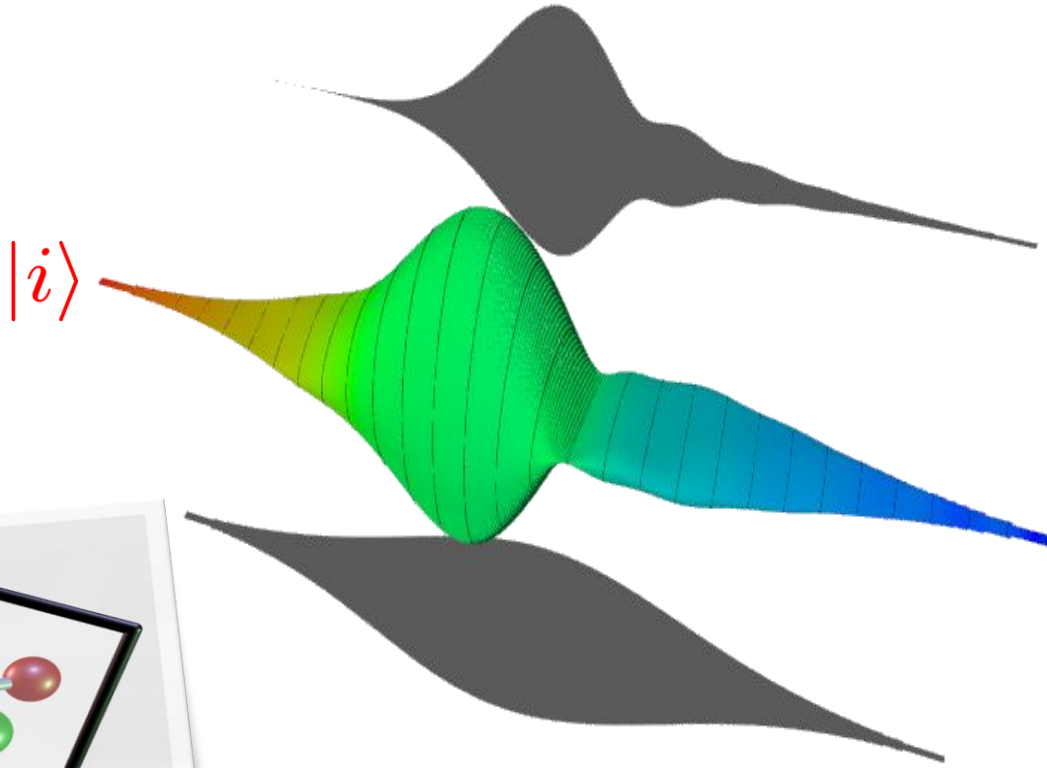
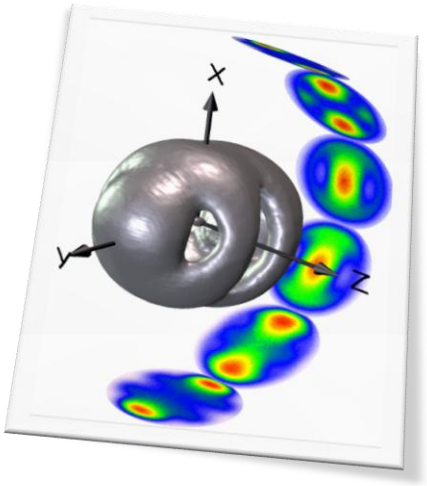
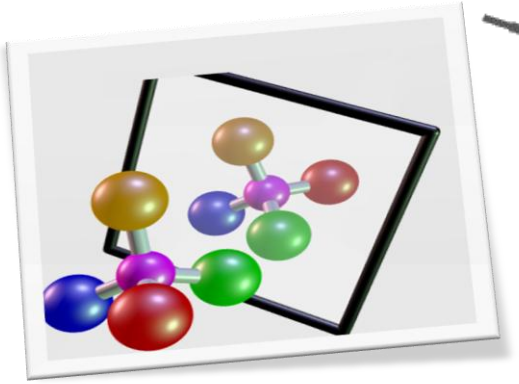
UNIKASSEL VERSITÄT

Control of sub-cycle electron dynamics in atoms and molecules

Matthias Wollenhaupt



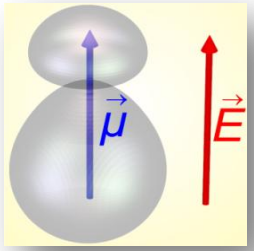




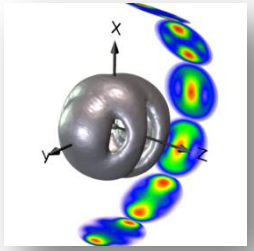
Universität Kassel

Institut für Physik

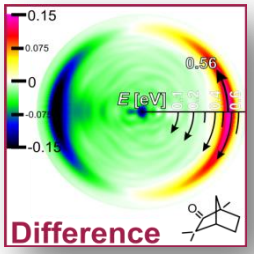
Laser control of ultrafast dynamics



- Strong-field control of *bound electron wave packets*

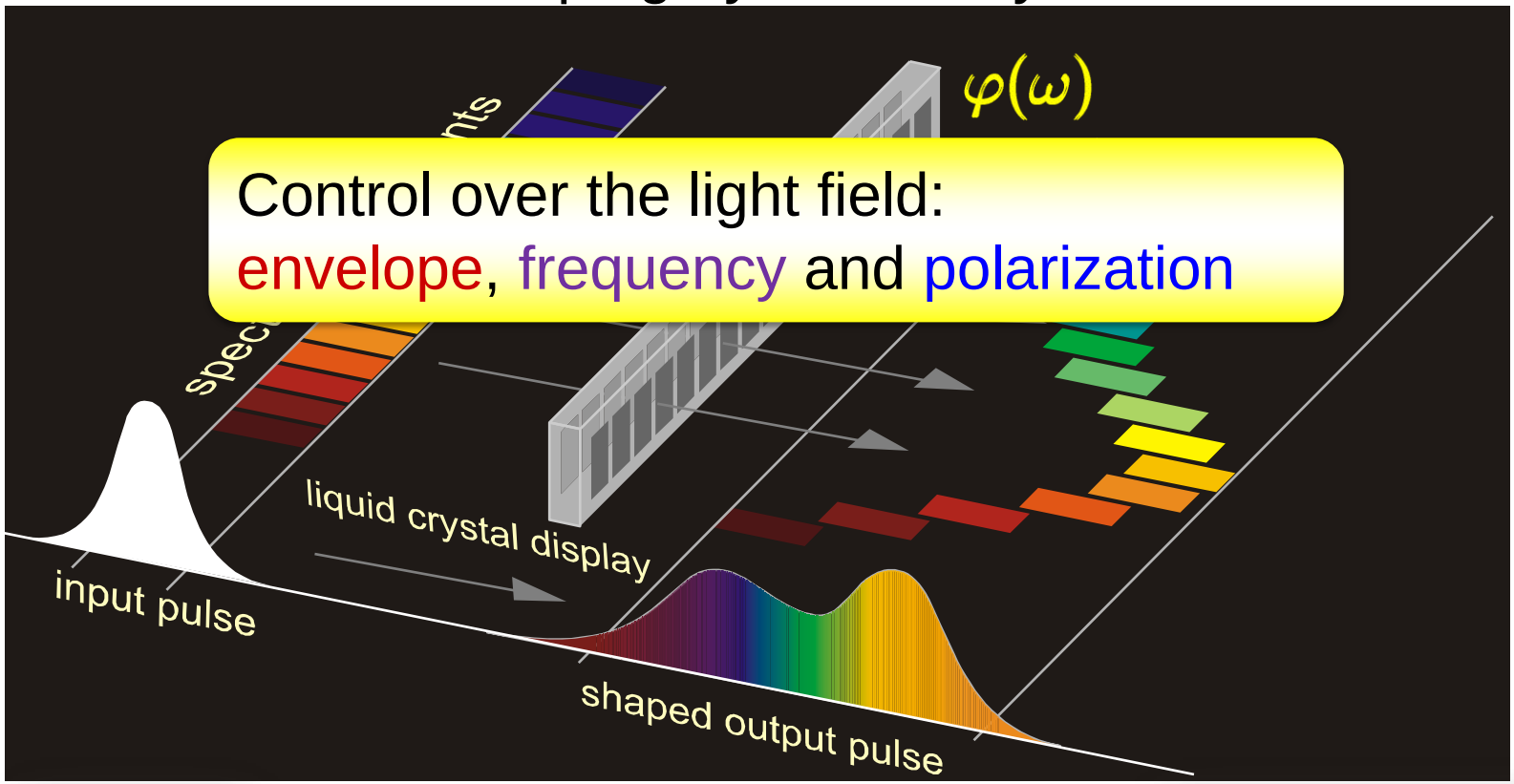


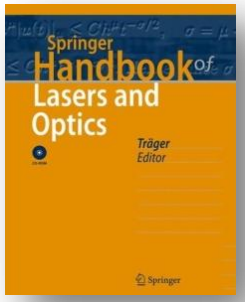
- 3d designer *free electron wave packets*



- Asymmetry in PAD *of chiral molecules*

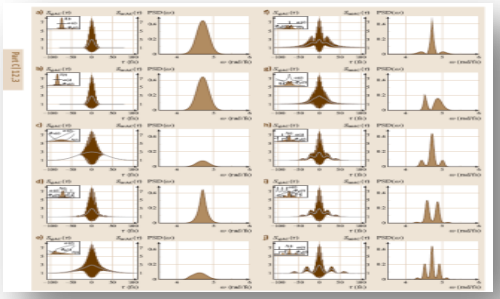
Pulse shaping by Fourier synthesis





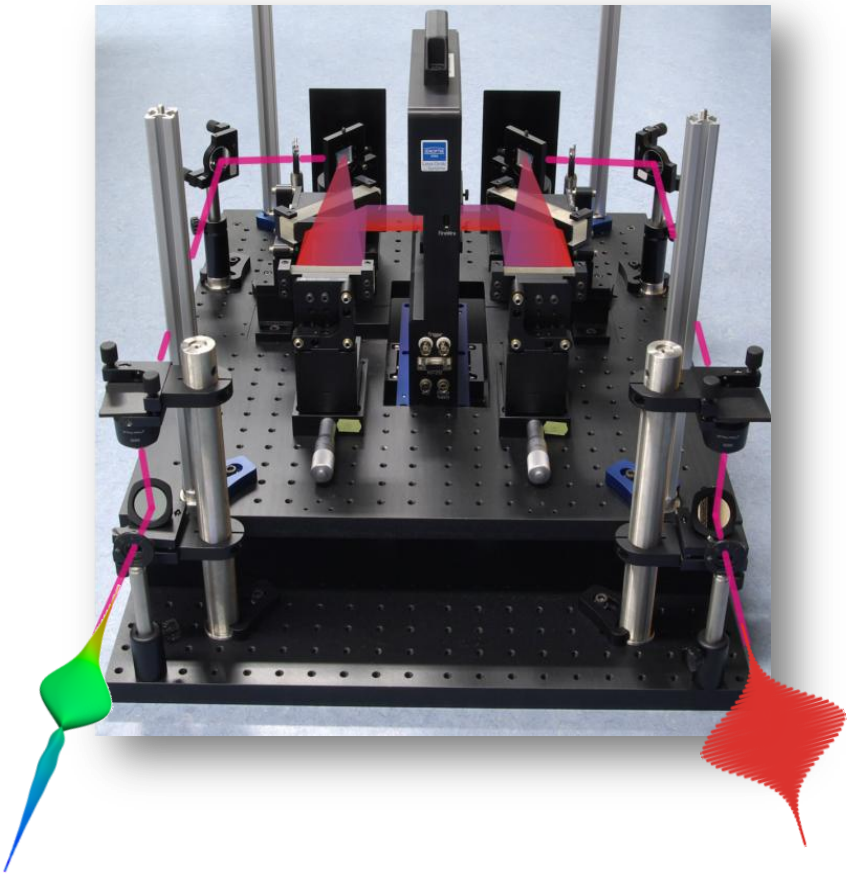
Review: Weiner: Rev. Sci. Instrum. ***71****, 1929 (2000)*

Springer Handbook of Laser and Optics, Chap. 12
Femtosecond Laser Pulses: Linear Properties, Manipulation, Generation and Measurement, (2007)

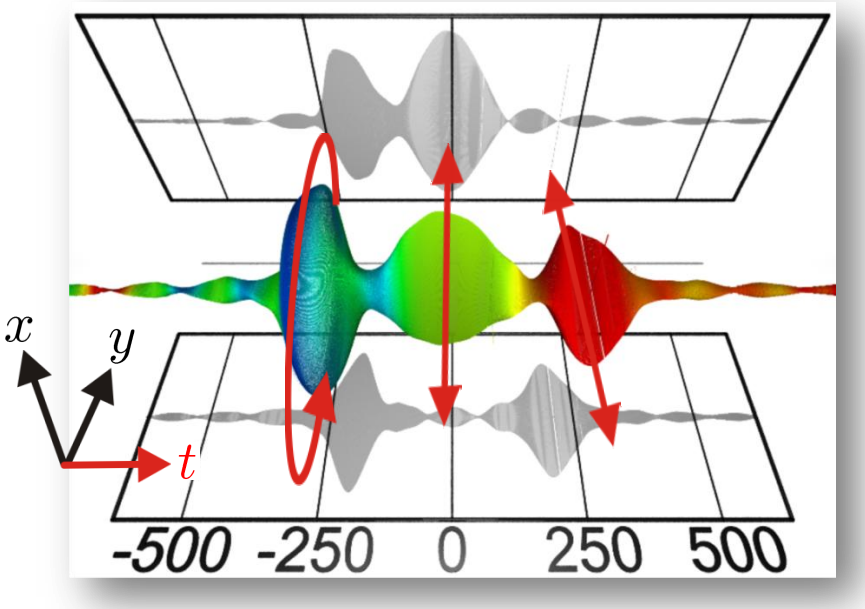


Pulse shaping capabilities

Experimental setup



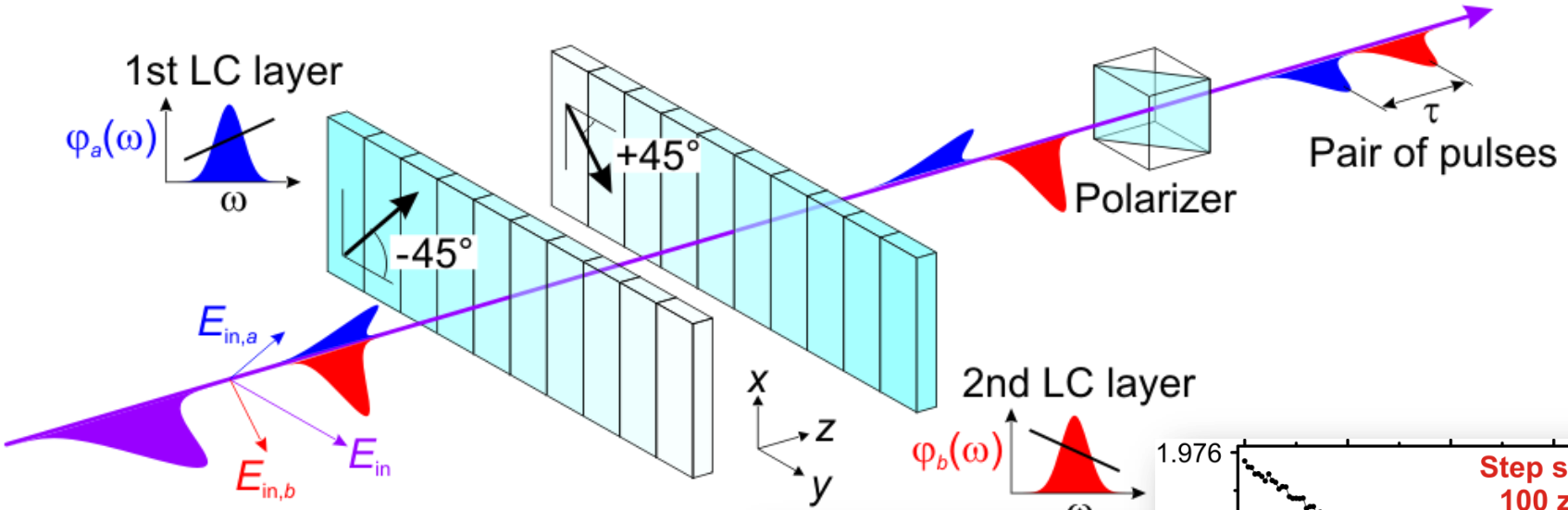
Polarization shaping
Designed and characterized pulse in interaction region



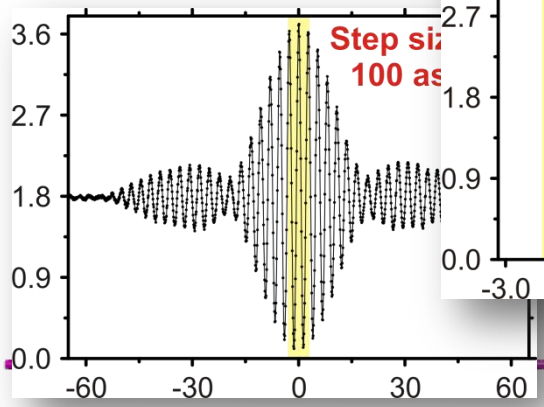
Time [fs]

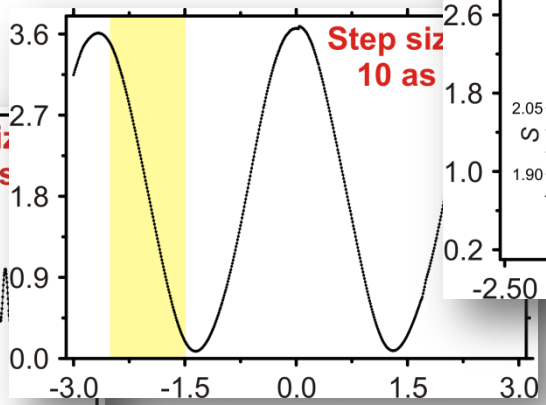
Phys. Rev. Lett, ***92***, *208301* (2004)

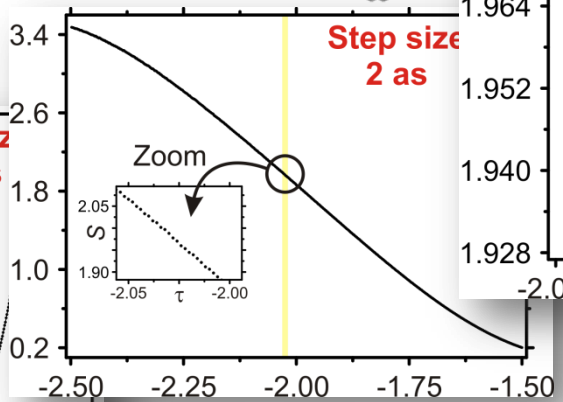
Zeptosecond precision pulse shaping

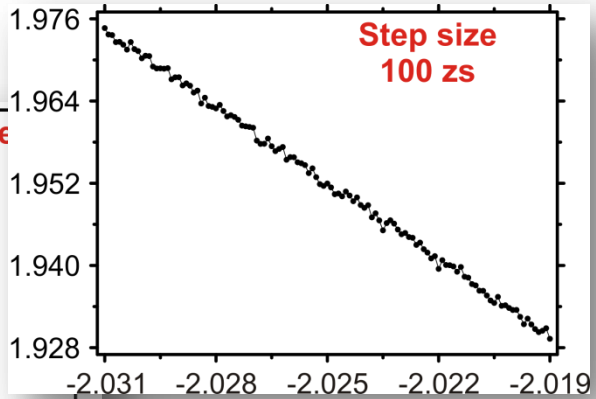


Autocorrelation







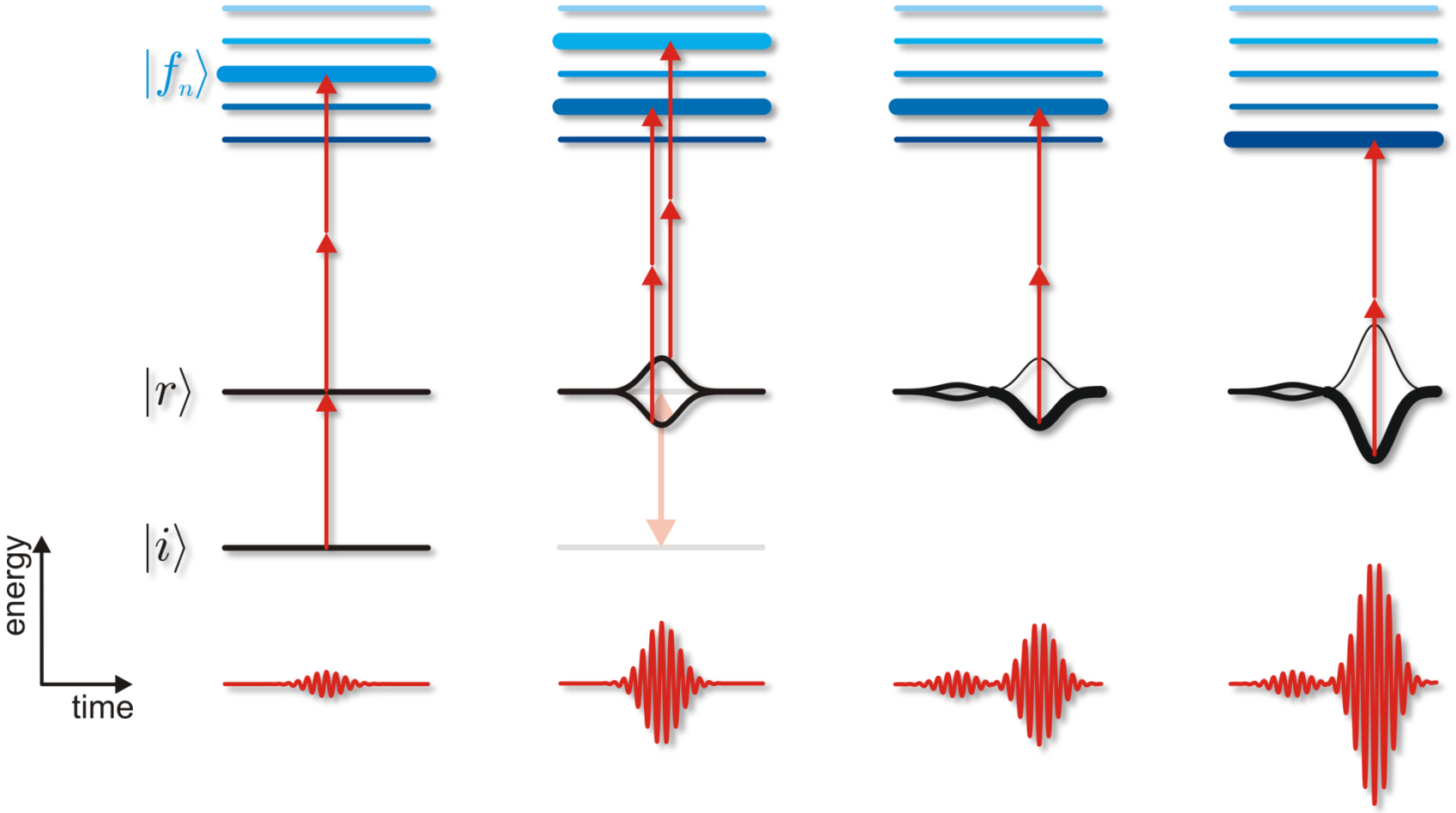


$$1\,\mathrm{zs} = \frac{1}{1\,000}\,\mathrm{as} = \frac{1}{1\,000\,000}\,\mathrm{fs}$$

Optics Express, ***19***, *11638* (*2011*)

Nature Photonics, ***5***, *513* (*2011*)

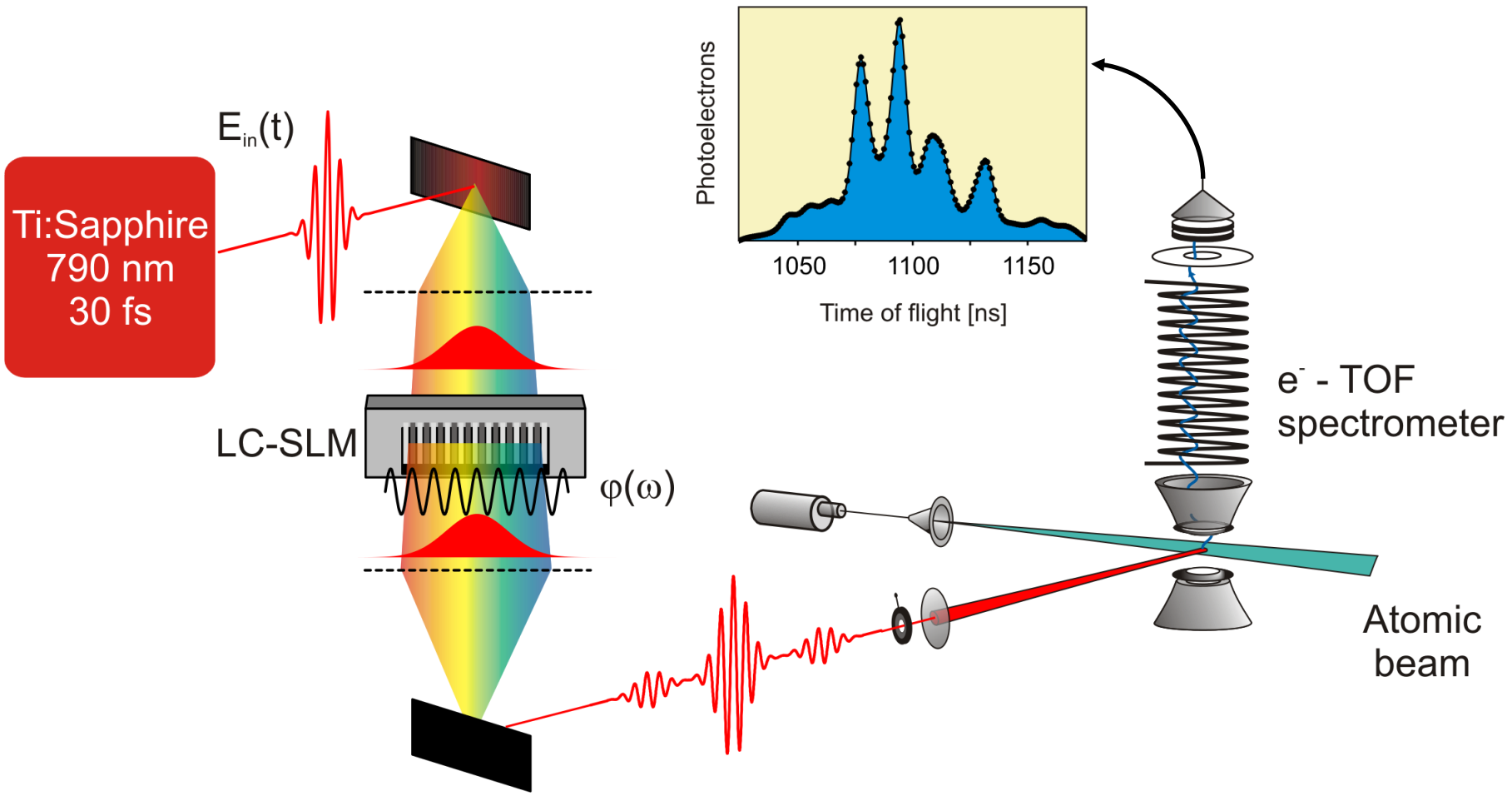
Strong-field control of quantum systems: selective and tunable



weak-field strong-field selectivity tunability

SELECTIVE POPULATION OF DRESSES STATES (SPODS)

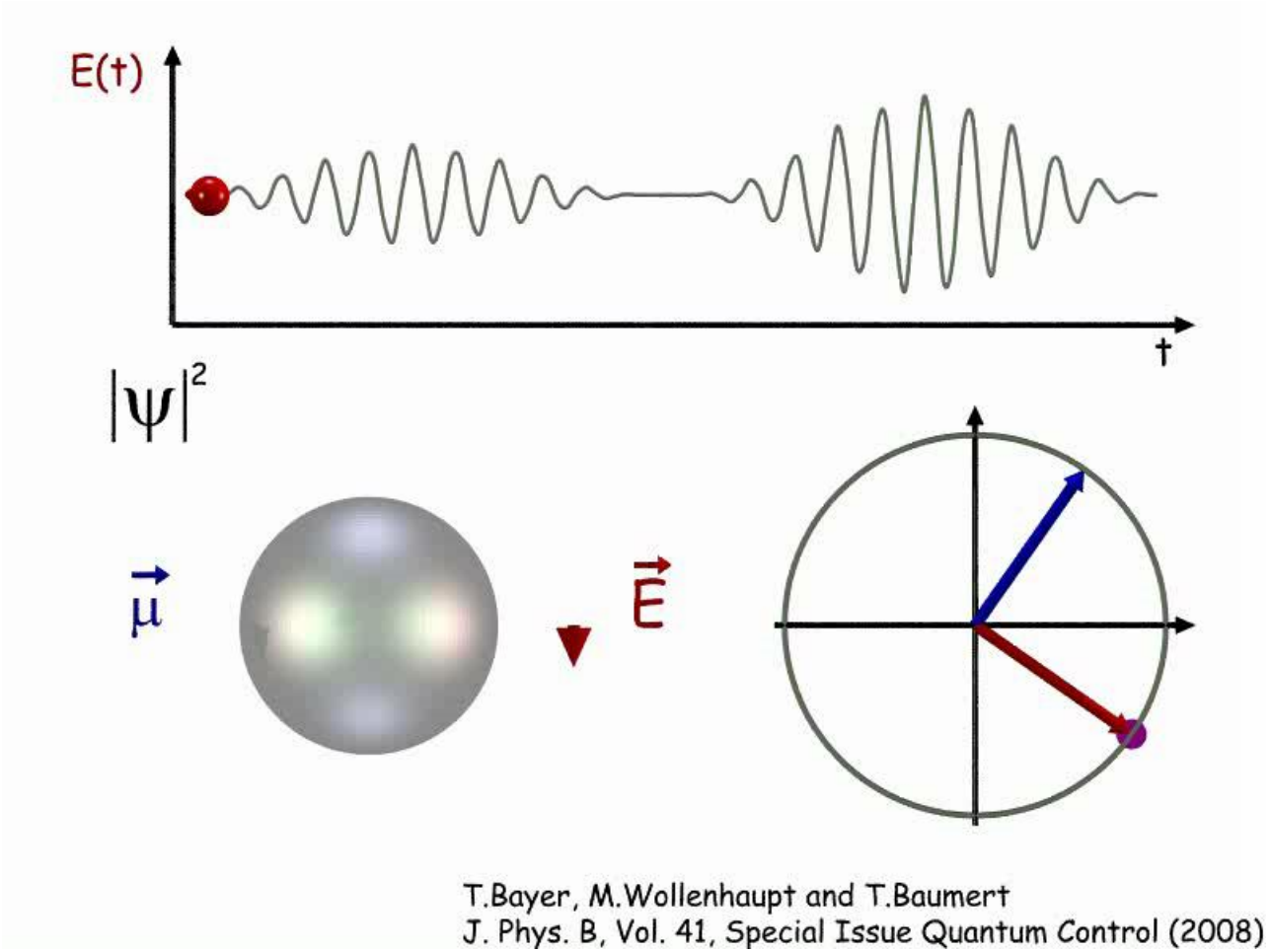
Probing coherent electron dynamics via photoelectron spectroscopy



Photoelectrons map *dressed* states!

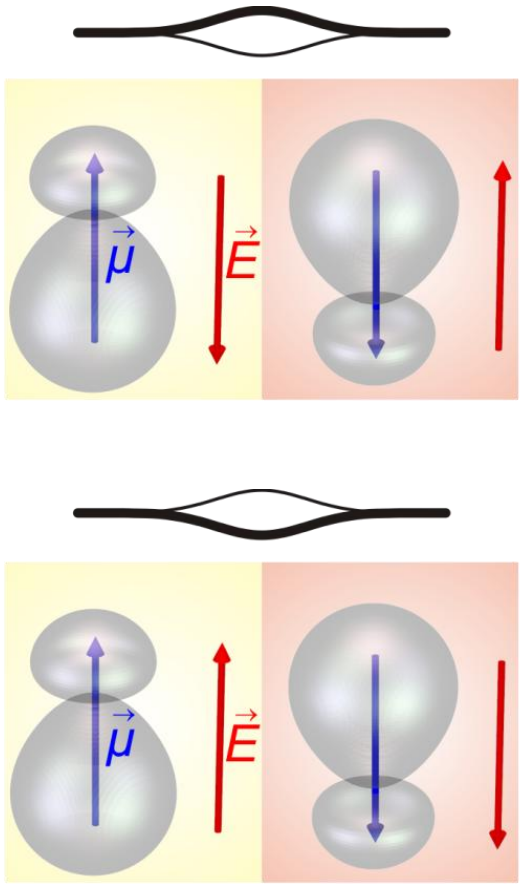
Phys. Rev. Lett, ***89***, *173001* (2002)

Control of *electronic* superposition states by phase tailoring

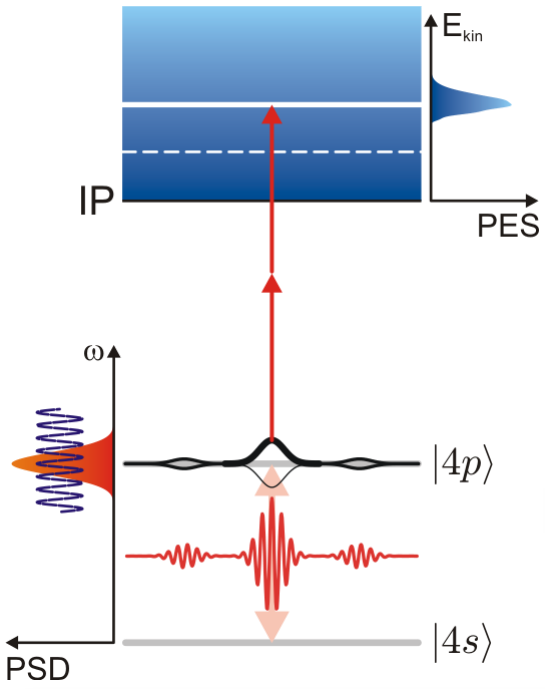


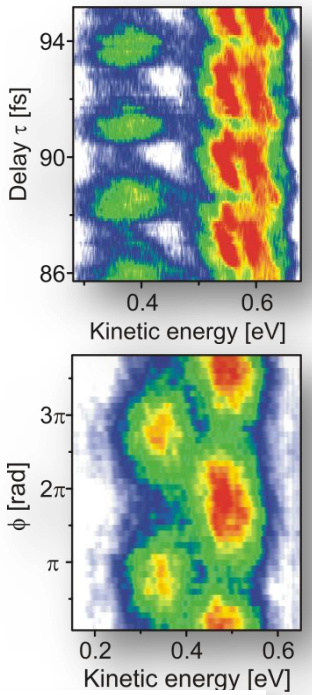
T.Bayer, M.Wollenhaupt and T.Baumert
J. Phys. B, Vol. 41, Special Issue Quantum Control (2008)

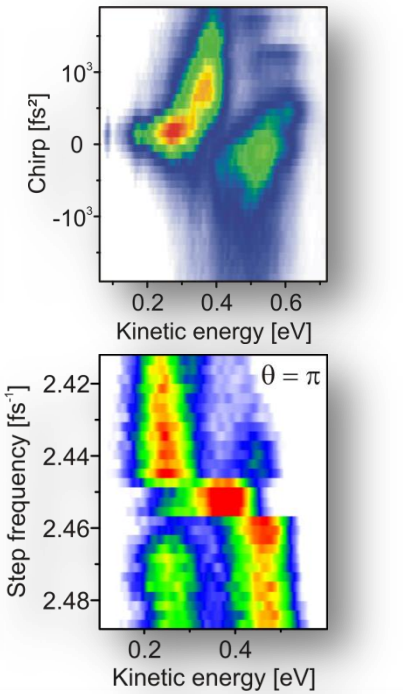
$$\hbar\Omega = -\vec{\mu} \cdot \vec{E}$$

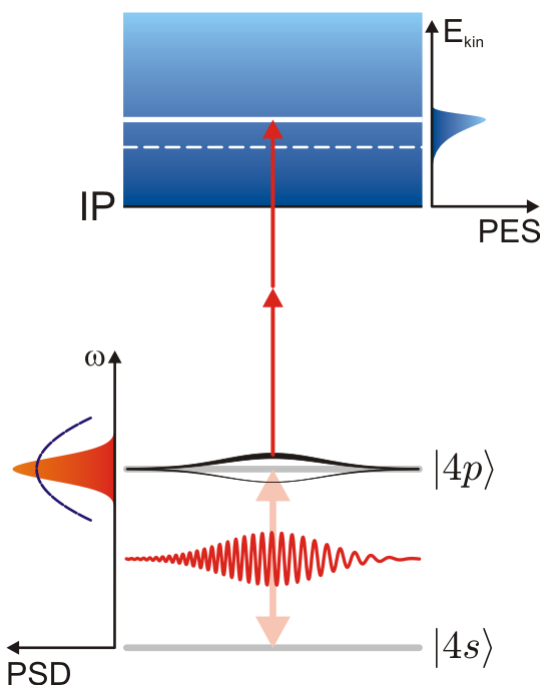


Observation of *SPODS*: results on *K* atoms









- SPODS by phase jumps

Phys. Rev. A, ***68***, *015401* (2003): Double pulses

Phys. Rev. A, ***73***, *063409* (2006): Pulse sequences

Chem. Phys. Lett., ***419***, *184* (2006): Theo: molecules

Optics Express, ***19***, *11638* (2011): Sub-atto precision

- SPODS by continually varying phase

Appl. Phys. B, ***82***, *183* (2006): Control by chirp

- SPODS by adaptive optimization

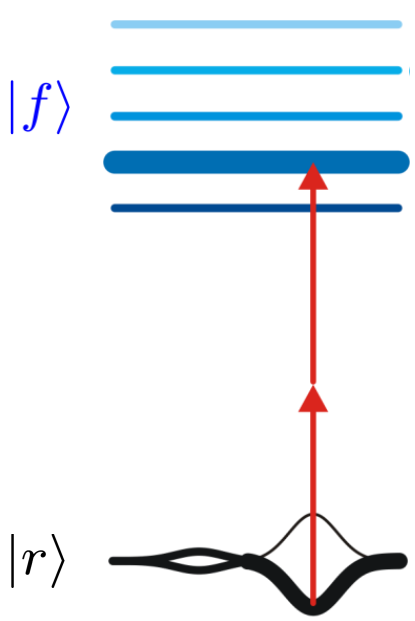
J. Opt. B, ***7***, *S 270* (2005): Evolutionary algorithm

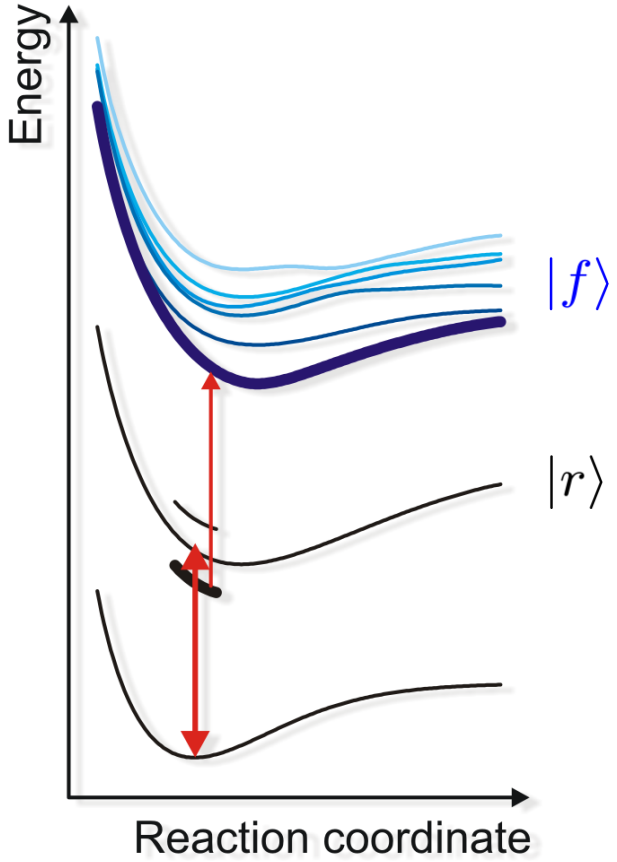
- Three states SPODS

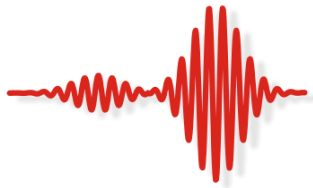
Phys. Rev. Lett. ***102***, *023004* (2009): Adiabatic Photon Locking

Phys. Rev. A, ***81***, *053422* (2010): Three state SPODS

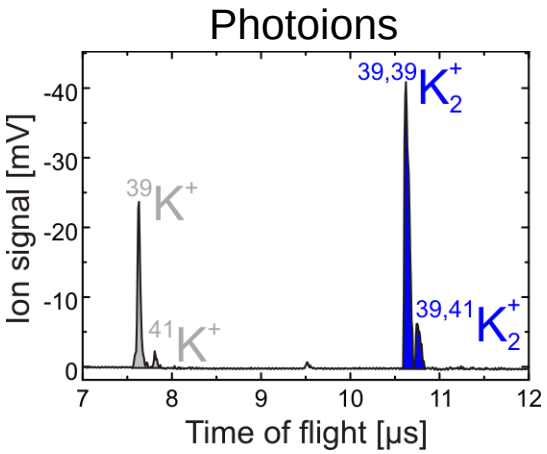
Controlling coherent electron dynamics in molecules

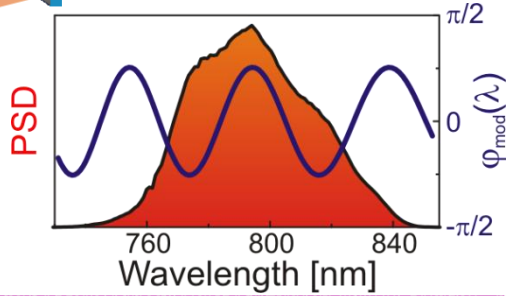






Experiment on K_2: two-color control and probe





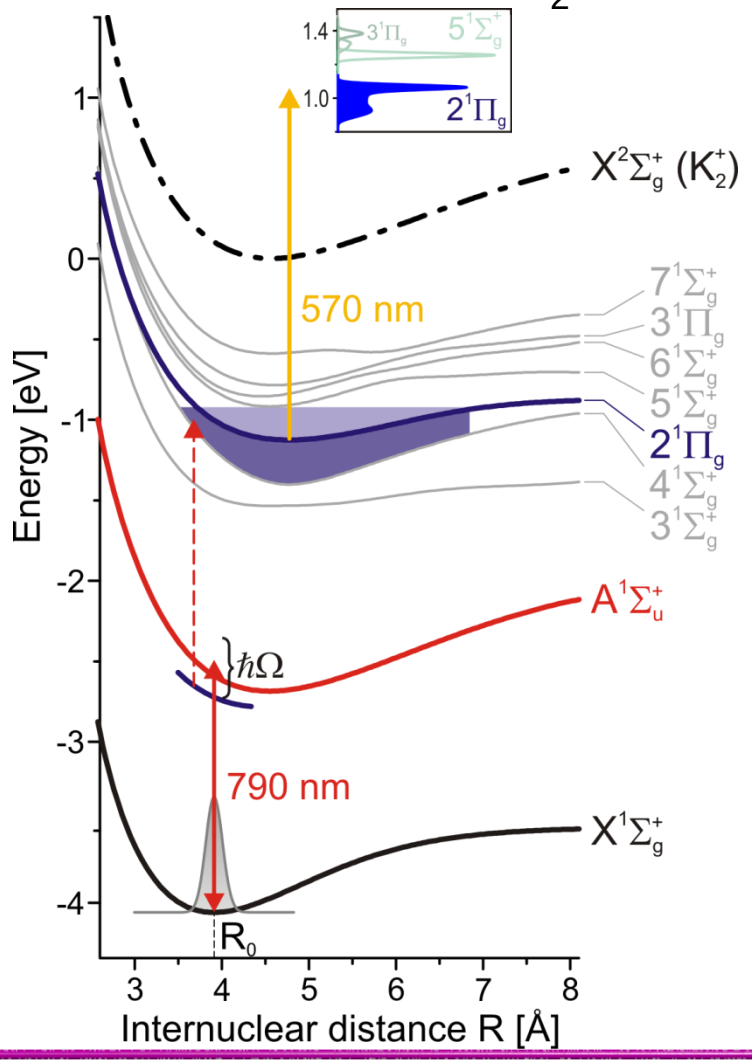
$\lambda_0 = 790$ nm

$\Delta t = 25$ fs

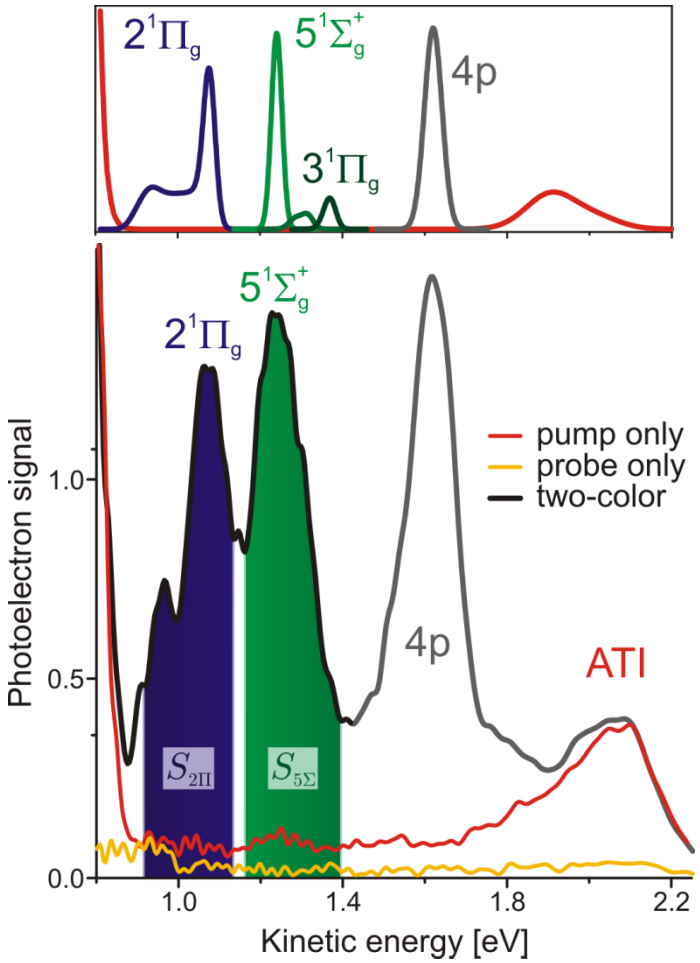
$I_0 \approx 8 \times 10^{11}$ W/cm^2

Control of molecular states via electron dynamics

Potentials of K_2

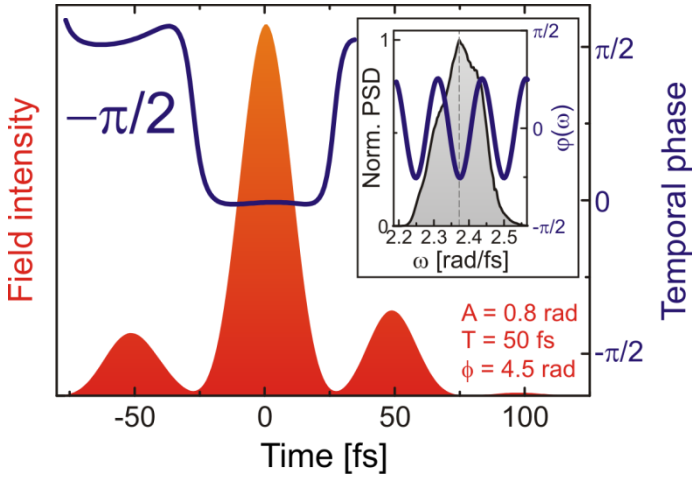


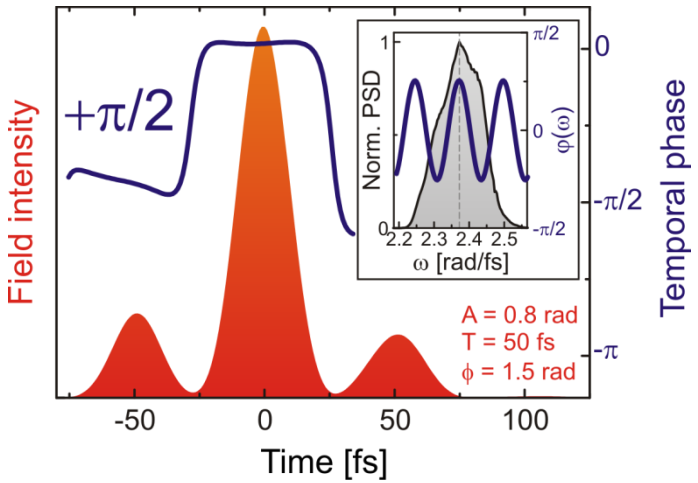
Photoelectron spectra



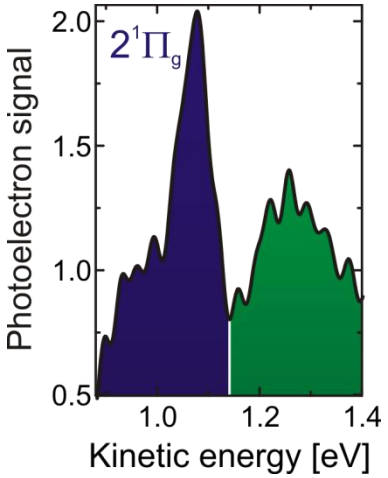
Switching population via electronic coherence by sub-cycle phase control

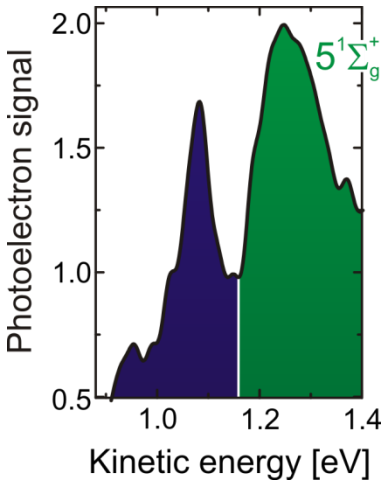
Temporal pulse shape





Photoelectrons

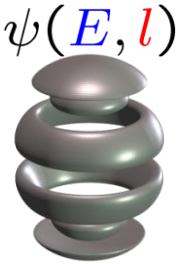


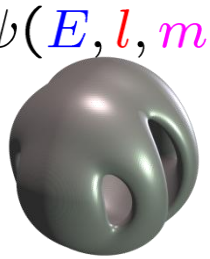


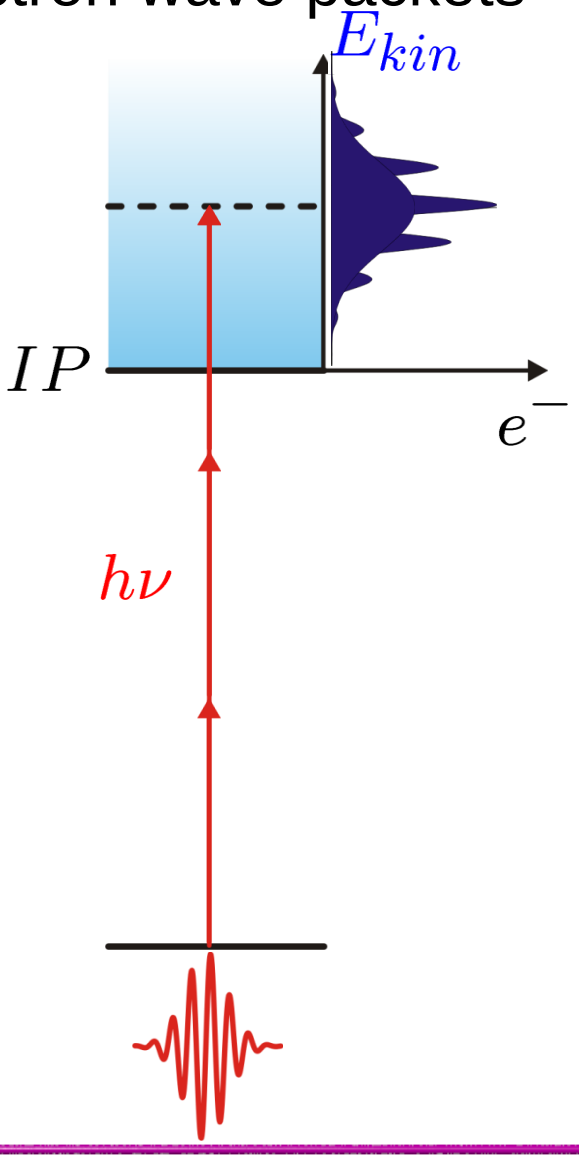
Control in the continuum: 3D designer electron wave packets

6. *Über einen die Erzeugung und Verwandlung des Lichtes betreffenden heuristischen Gesichtspunkt; von A. Einstein.*

$$\Pi \varepsilon = \frac{R}{N} \beta v - P$$



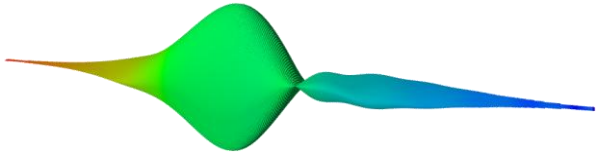




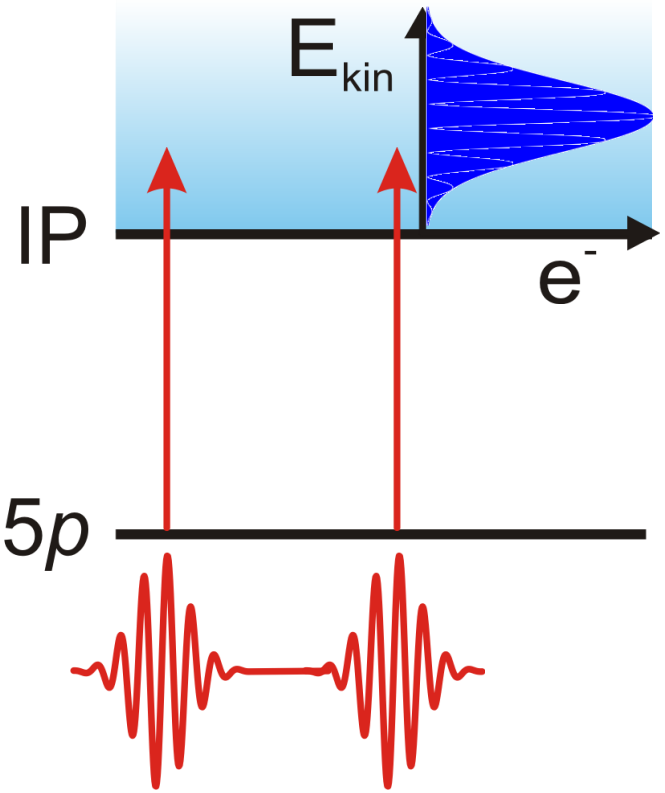
- Ultrashort laser pulses
- Shaped laser pulses
- Multi photon ionization

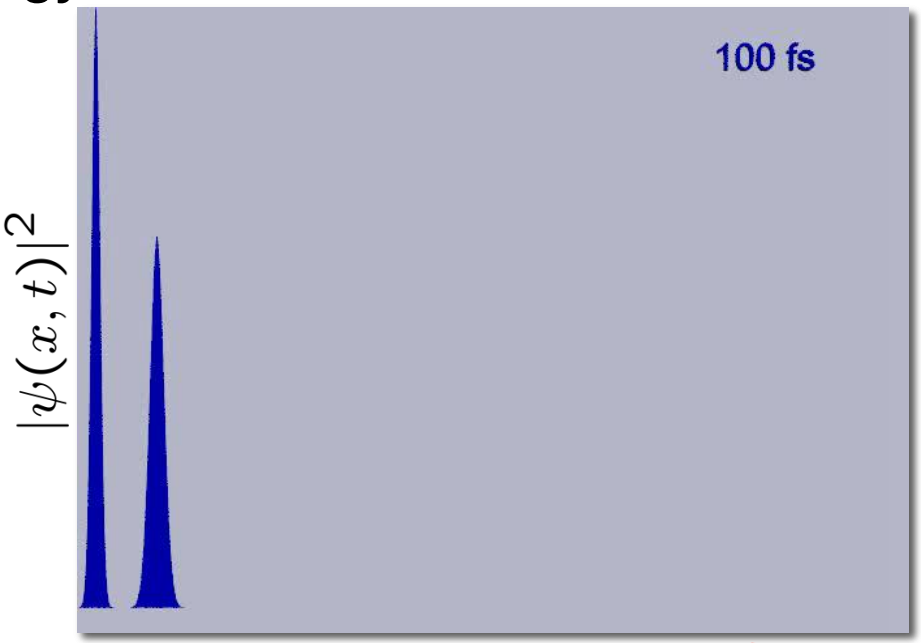
$$E_{kin} = n\,h\nu - IP$$

- Polarization shaping



Controlling the energy distribution: double slit in time

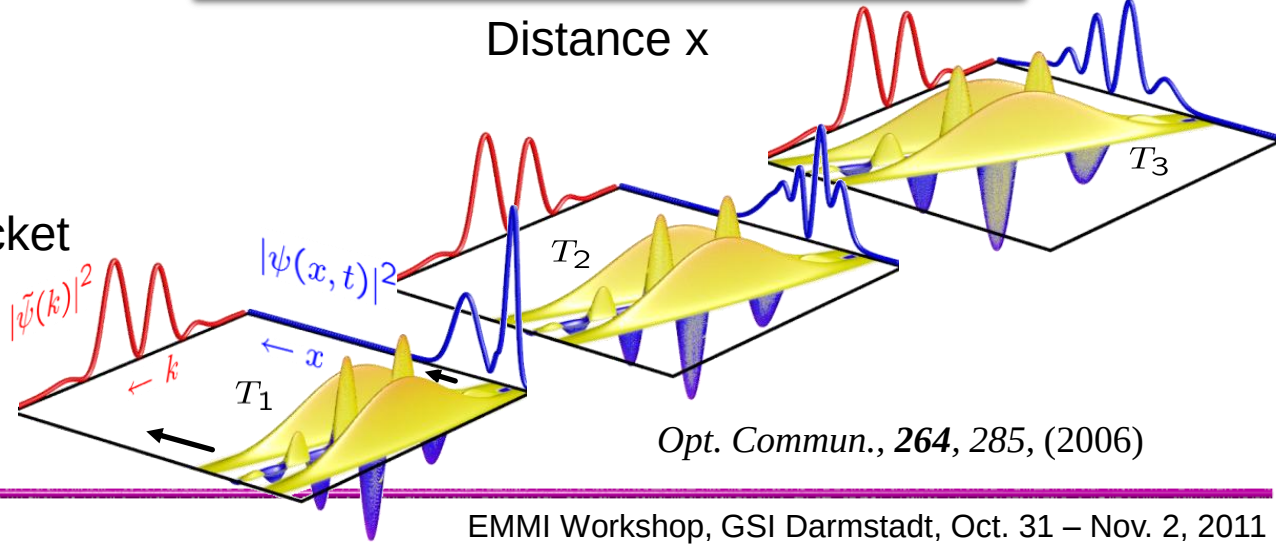




Distance x

Electron wave packet

$\psi_s(E)$

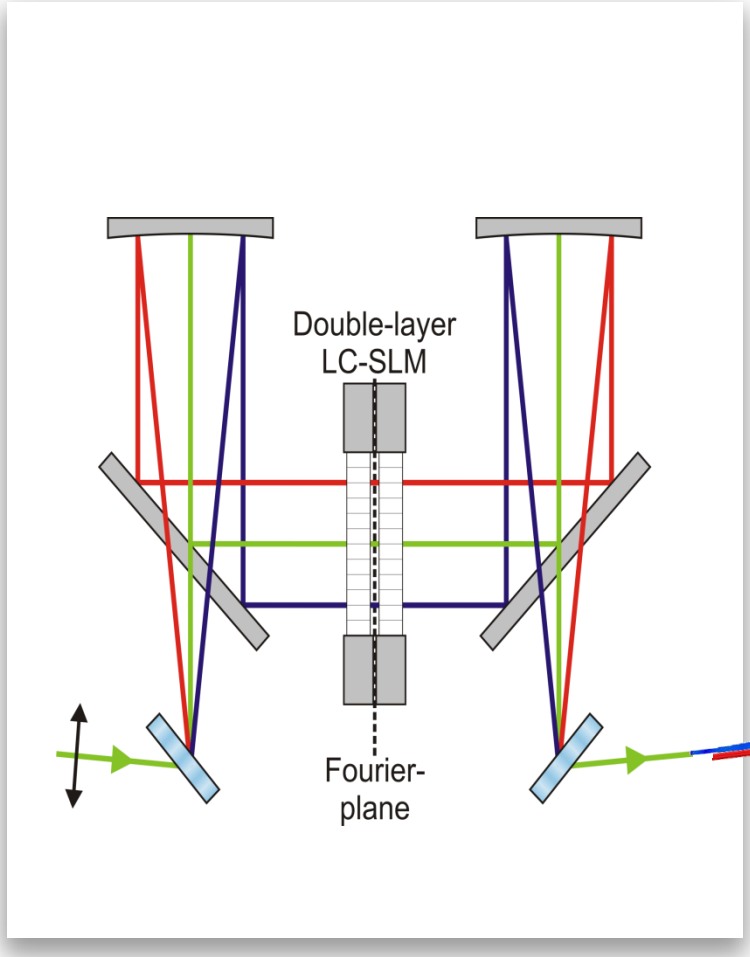


Opt. Commun., ***264***, *285*, (2006)

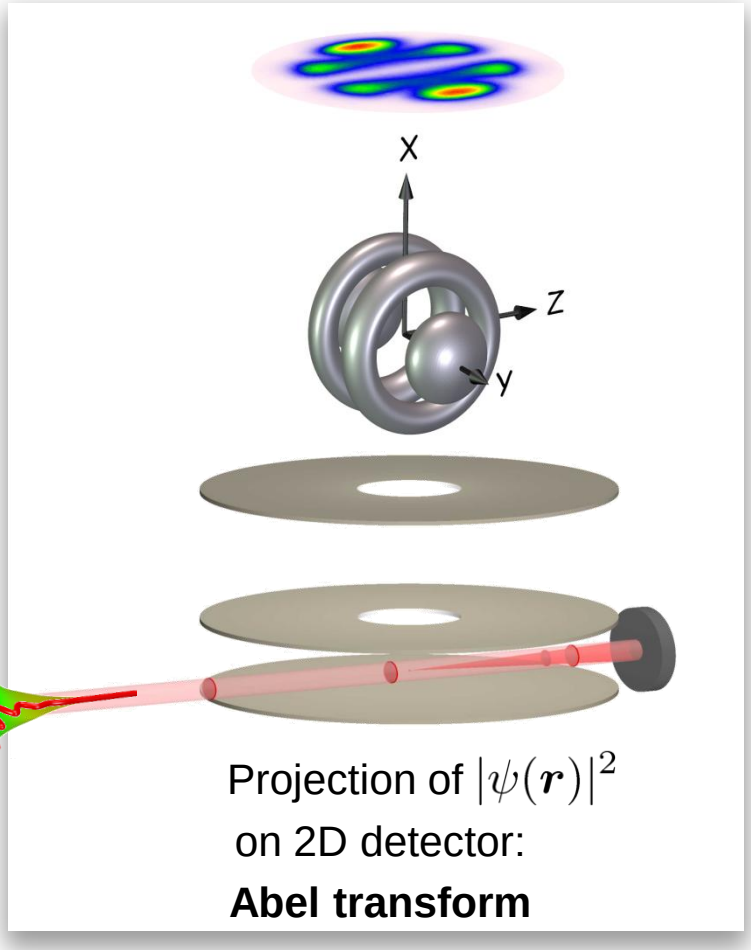
Projections: 3d information matters!

2d imaging: photoelectron angular distributions

Polarization shaper

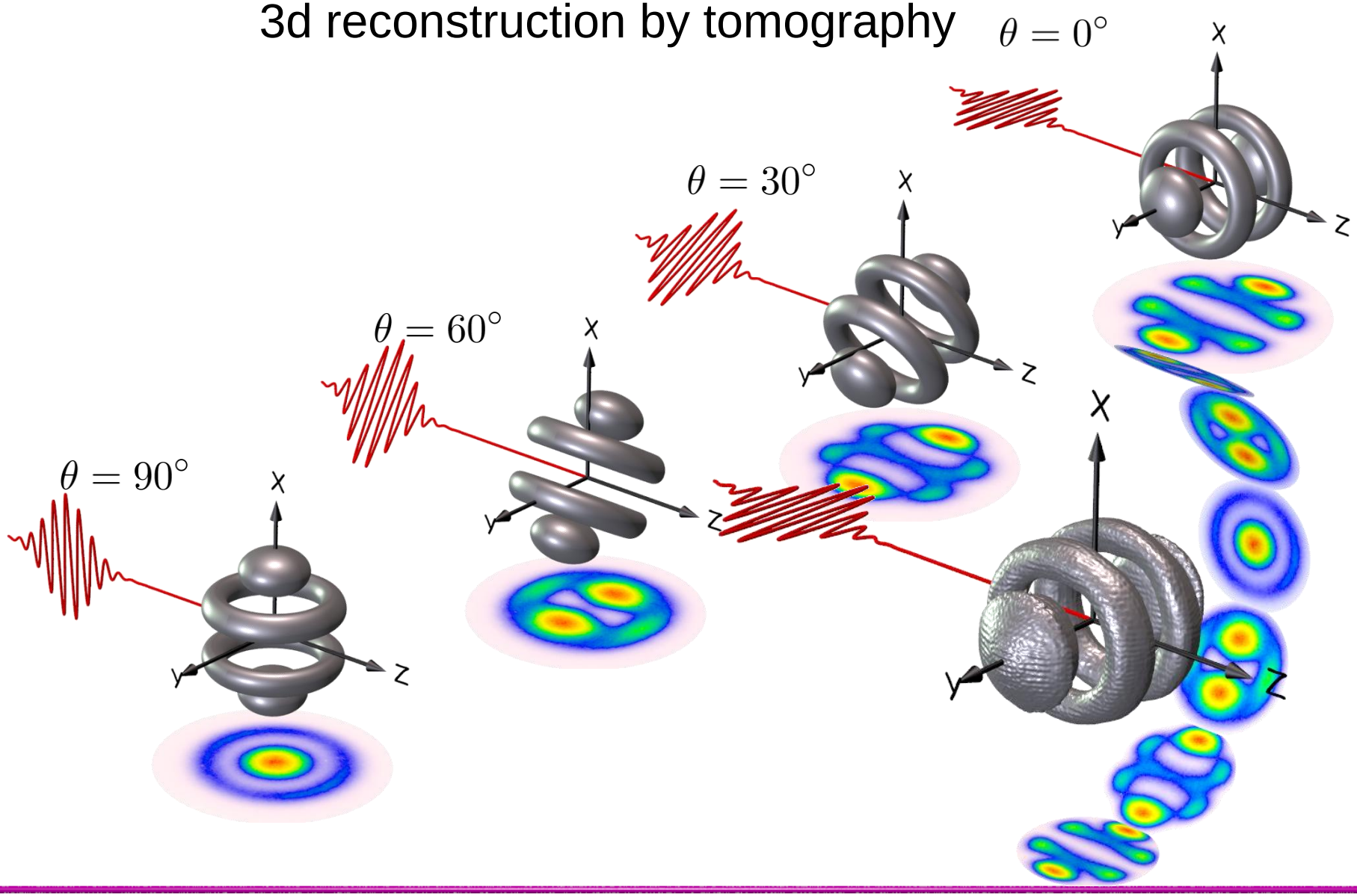


Imaging spectrometer

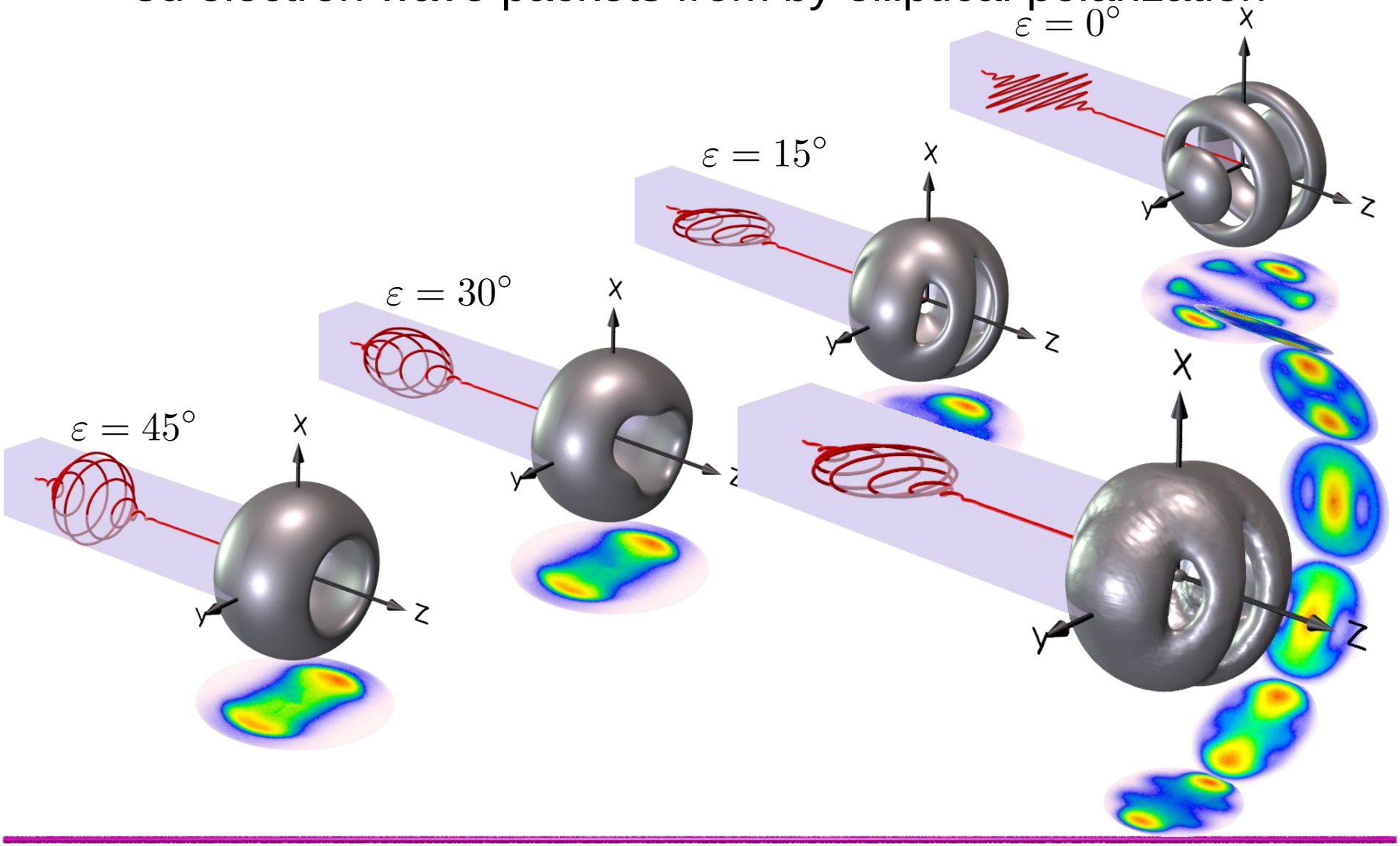


Projection of $|\psi(\boldsymbol{r})|^2$
on 2D detector:
Abel transform

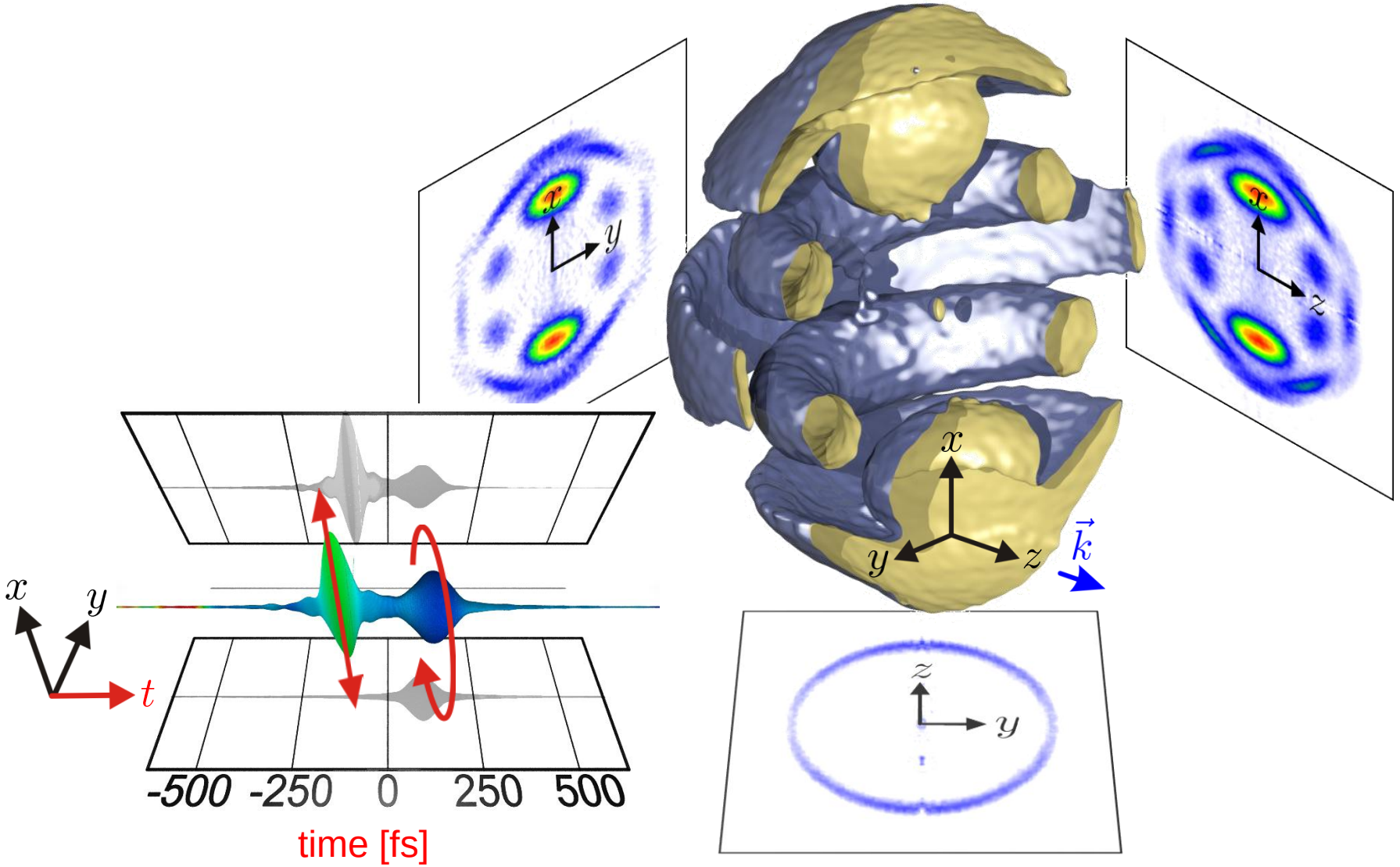
3d reconstruction by tomography



3d electron wave packets from by elliptical polarization

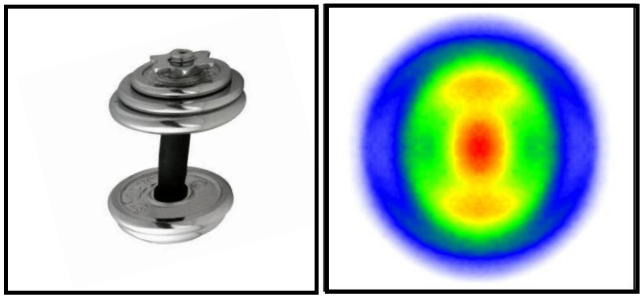


Creating designer wave packets by polarization shaping

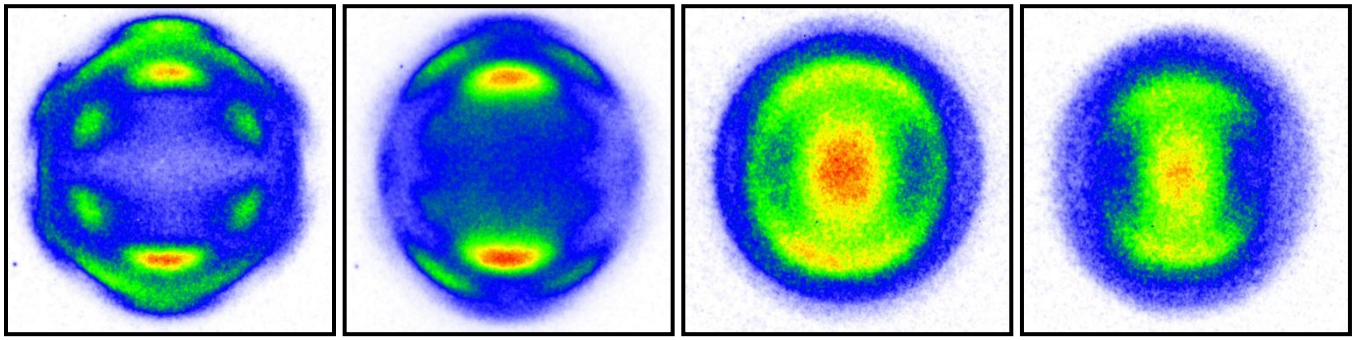


Adaptive optimization of photoelectron angular distributions

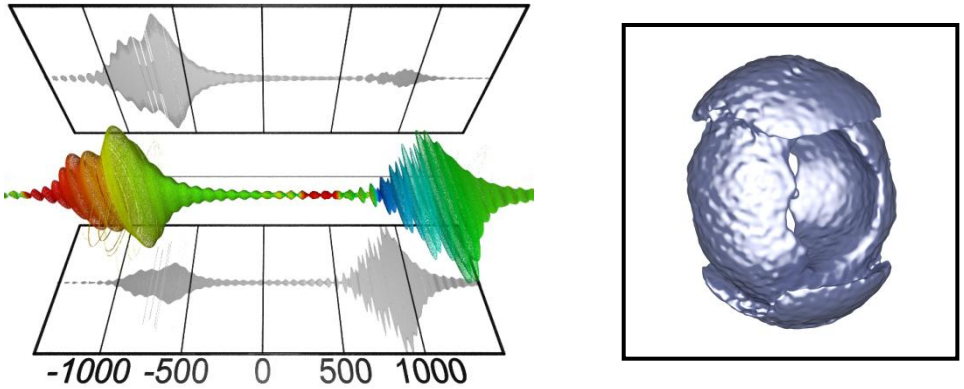
Target image in projection:



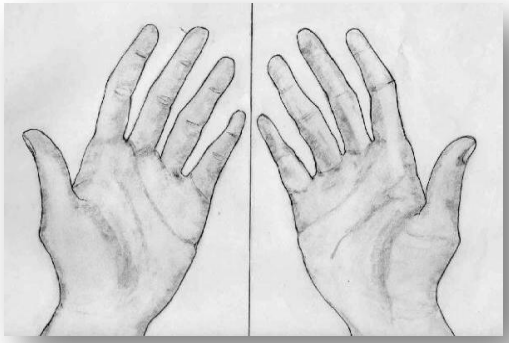
Evolution



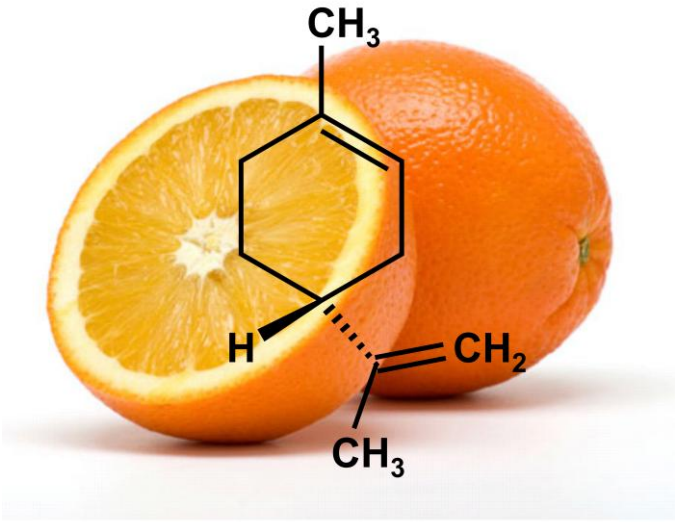
Reconstruction



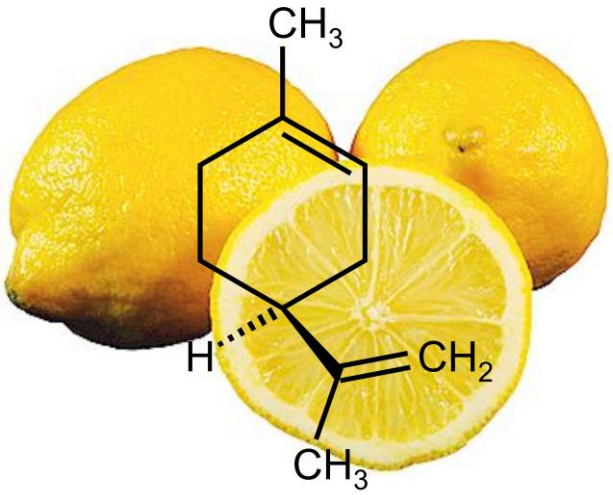
Chirality



- Physics: circular dichroism
- Chemistry: enantioselective reactions
- Biology: homochirality
- Medicine: chirality of drugs

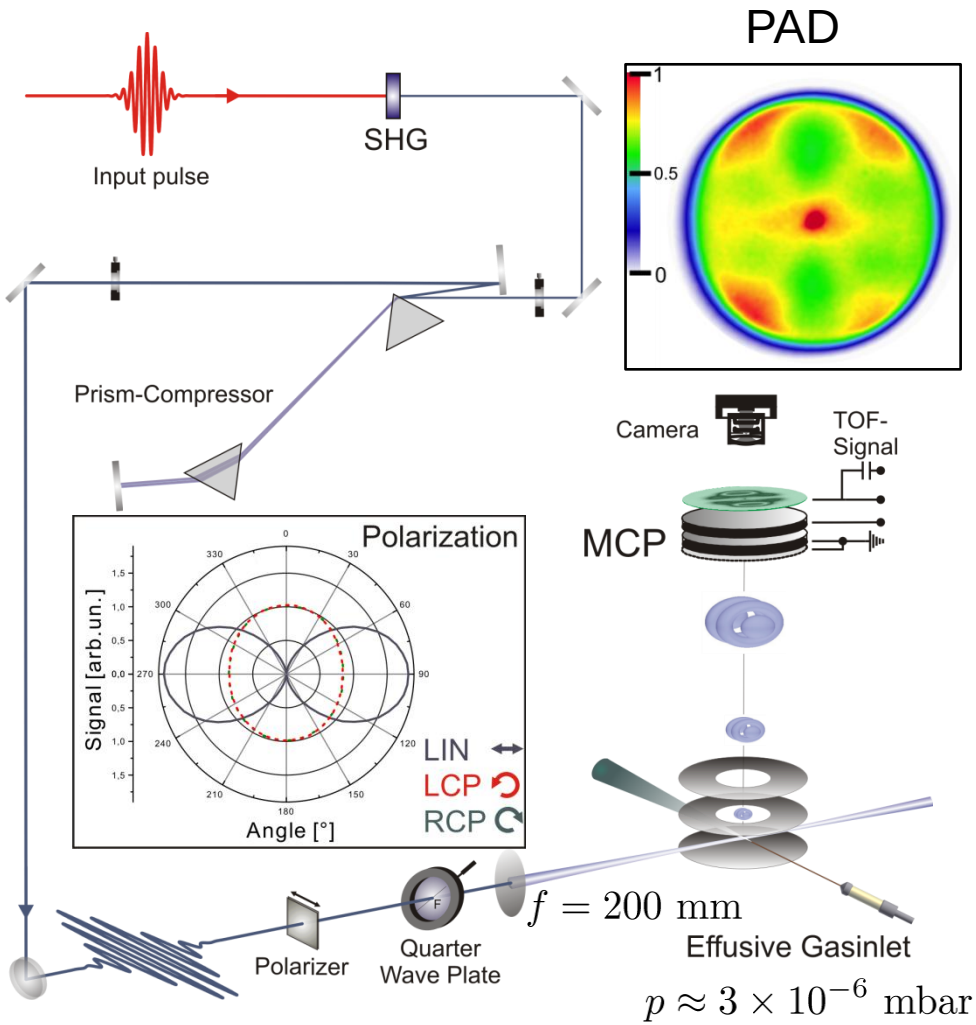


R-(+)-limonene



S-(-)-limonene

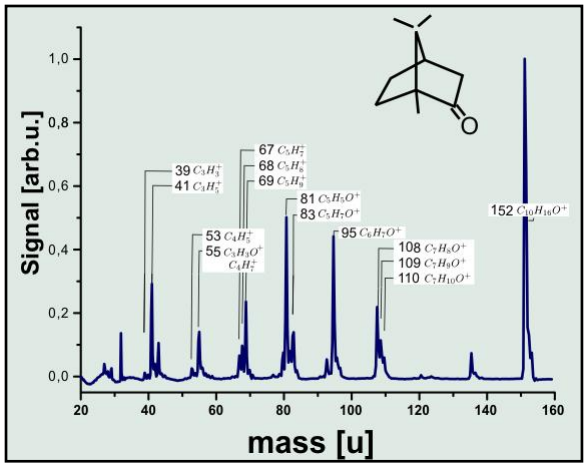
Photoelectron angular distributions of chiral molecules



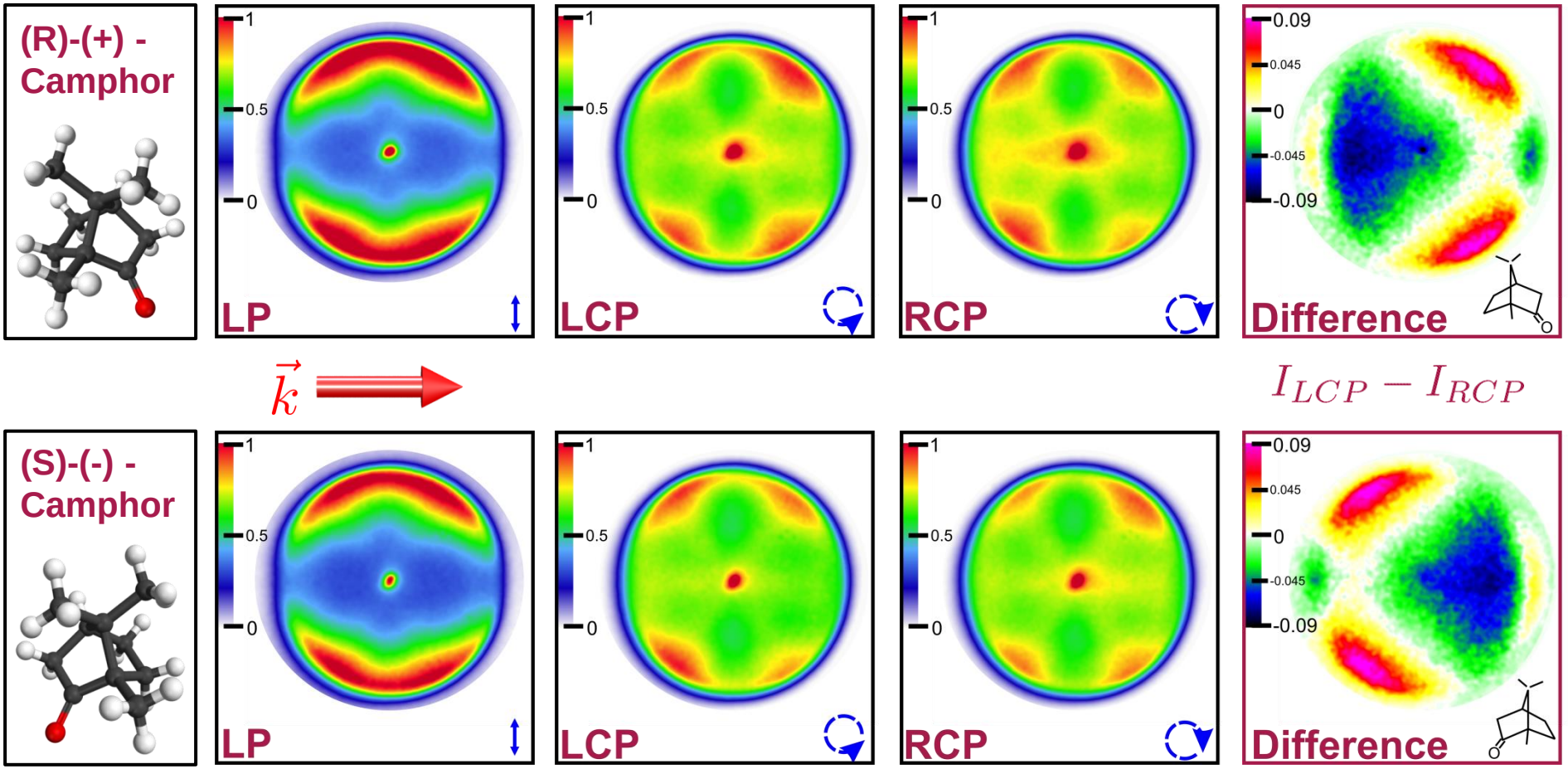
Our approach

- Multi-photon ionization (REMPI)
- Shaped femtosecond laser pulses
- Photoelectron Angular Distributions

Mass spectra



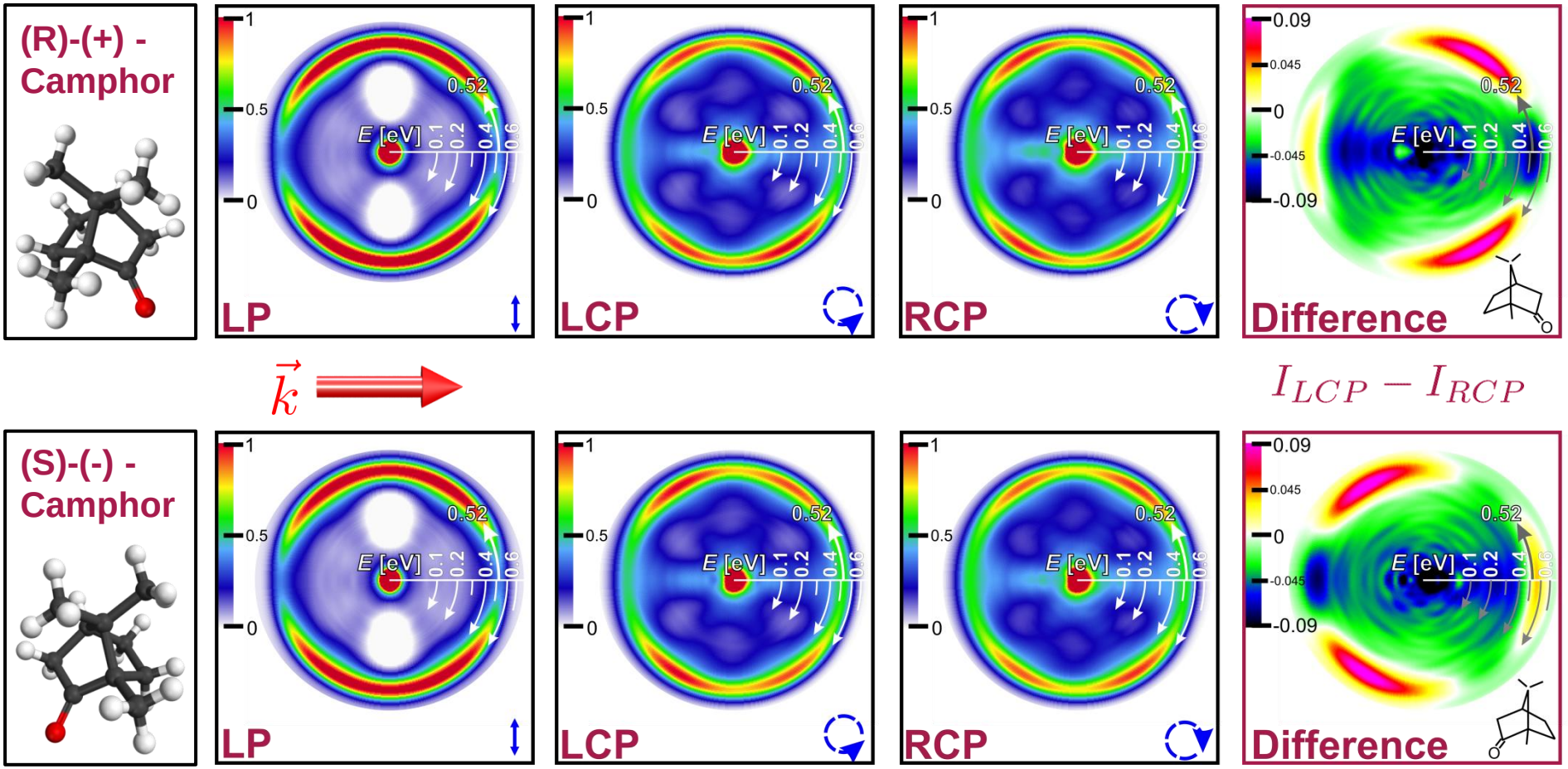
PAD of (randomly oriented) camphor molecules



$Laser : \lambda = 790\ \text{nm}, \Delta t = 25\ \text{fs}, 1\ \text{kHz}$

$SHG : \lambda = 395\ \text{nm}, \Delta t \approx 33\ \text{fs}, I_0 \approx 1.3 \times 10^{14}\ \text{W/cm}^2$

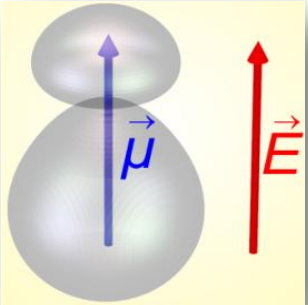
PAD of camphor molecules: Abel inversion



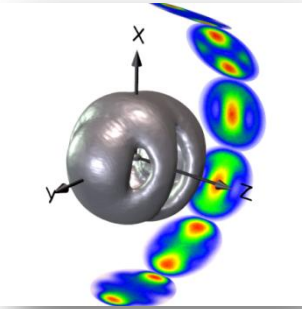
$Laser : \lambda = 790\ \text{nm}, \Delta t = 25\ \text{fs}, 1\ \text{kHz}$

$SHG : \lambda = 395\ \text{nm}, \Delta t \approx 33\ \text{fs}, I_0 \approx 1.3 \times 10^{14}\ \text{W/cm}^2$

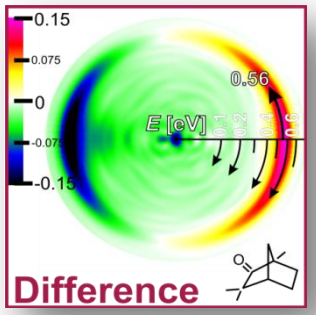
Summary



- Strong-field control of bound electron wave packets
- Selective population of dressed states on atoms
- Control of sub-cycle electron dynamics in molecules



- Designer free electron wave packets
- 3d tomographic reconstruction of electron wave packets
- 3d control with polarization shaped pulses
- Adaptive optimization of PAD



- Chiral light and chiral moleculess
- Examples on camphor and fenchone
- Asymmetry in PAD: mirror images of light and matter

Thanks to the group…


T. Bayer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…and thank for your attention !
FastQuast
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Otto-Braun Fonds
Photo: C. Sarpe