UNI KASSEL VERSITÄT

# ULTRAFAST LASER CONTROL Of IONIZATION

## Fundamentals And Applications

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Institut fuer Physik
der Universitaet Kassel,
GERMANY

H. Baumann: first permanent Laser Sculpture / since Documenta 6 1977 / Kassel

# Ultrafast Laser Control

**General Idea: steer photophysical system from initial state to final state with high selectivity and efficiency by adapting (tailoring) light field to primary photophysical processes**


## Origine: coherent control of chemical reactions

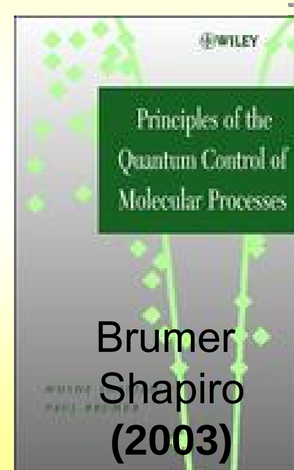
### Our experimental Reviews

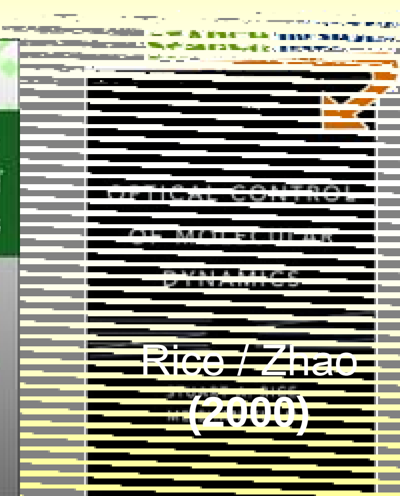
TB, Helbing, Gerber, in "Advances: Chemical Reactions and their Control on the Femtosecond Time Scale" John Wiley & Sons, Inc. New York; **(1997)** 47-77.

Wollenhaupt, Engel, TB, Annu. Rev. Phys. Chem. **56 (2005)** 25-56.

Brixner, Pfeifer, Gerber, Wollenhaupt, TB in "Femtosecond Laser Spectroscopy" Springer **(2005)** 225-266.

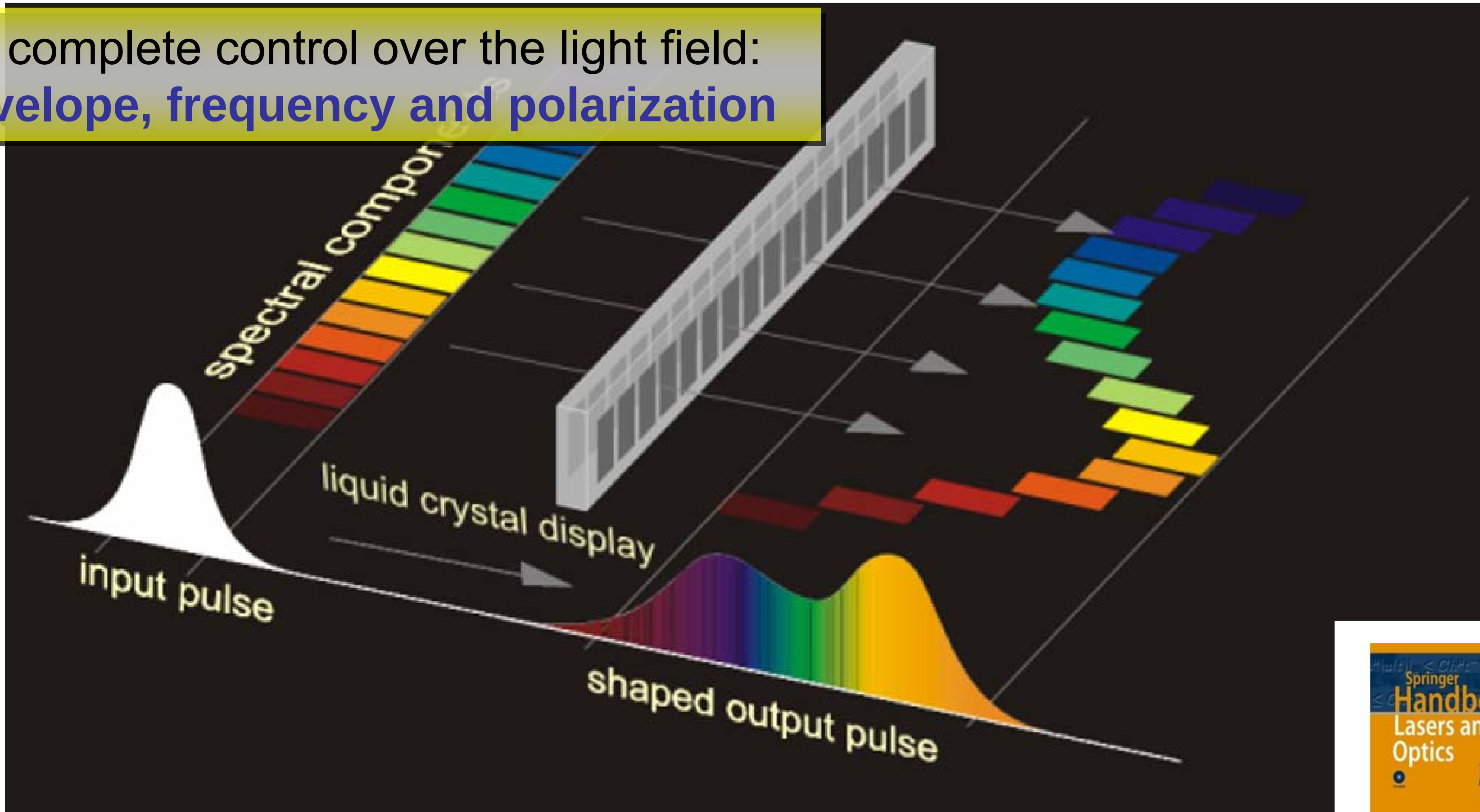
### Textbooks

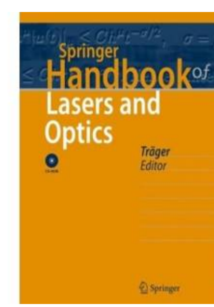


# Key Technology: Tailoring Light Via Spectral fs Pulse Shaping

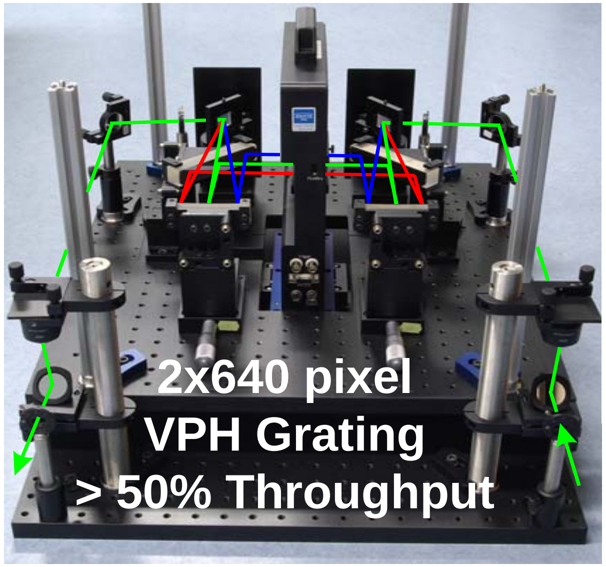
⇒ complete control over the light field:
**envelope, frequency and polarization**

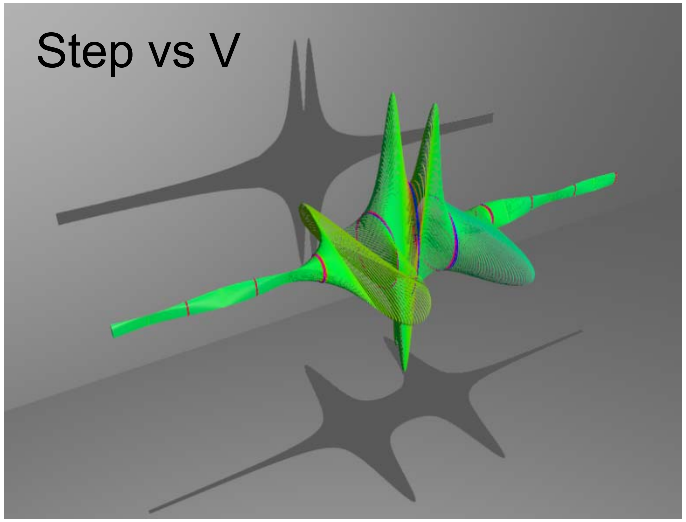


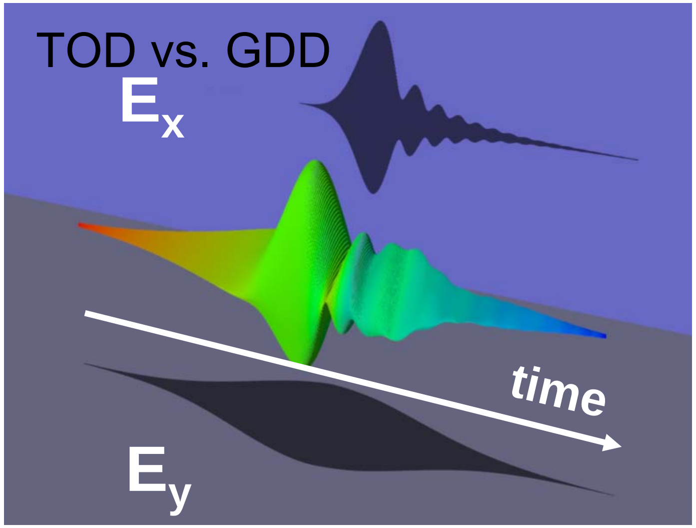



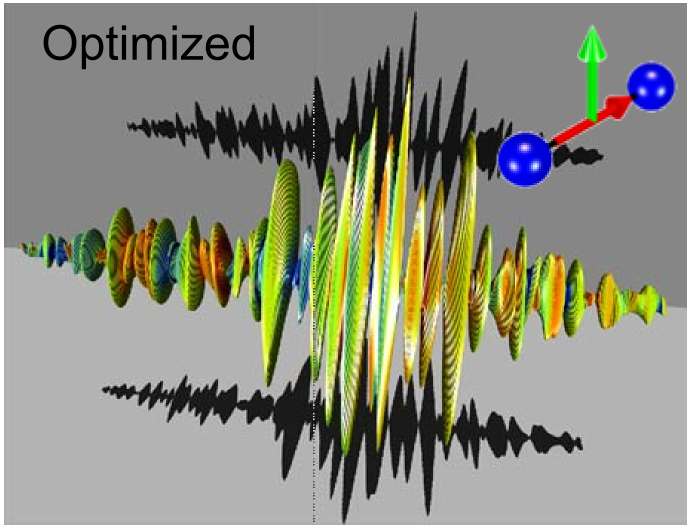

Review: Weiner: Rev. Sci. Instrum. 71(5), 1929-1960 (2000)
Wollenhaupt, Assion, TB in “Springer Handbook of Lasers and Optics“, ed. F. Träger, Springer (2007)

# Examples of Polarization Shaping*



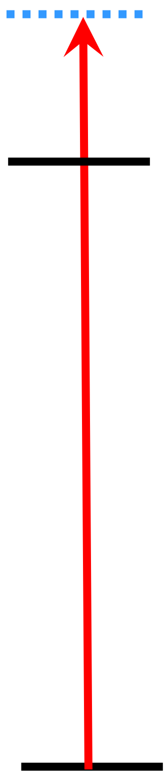




*Brixner/Gerber 2001

Phys. Rev. Lett. **92** (2004) 208301 in coop. T. Brixner and G. Gerber

# Ultrafast Laser Control of Ionization



ANNALEN
DER
PHYSIK.

BEGRÜNDET UND FORTGEFÜHRT DURCH
F. A. C. GREN, L. W. GILBERT, J. C. POGGENDORFF, G. UND E. WIEDEMANN.

VIERTE FOLGE.

BAND 17.

DER GANZEN REIHE 322. BAND.

KURATORIUM:
F. KOHLRAUSCH, M. PLANCK, G. QUINCKE,
W. C. RÖNTGEN, E. WARBURG.

UNTER MITWIRKUNG
DER DEUTSCHEN PHYSIKALISCHEN GESELLSCHAFT
UND INSBESONDERE VON
M. PLANCK

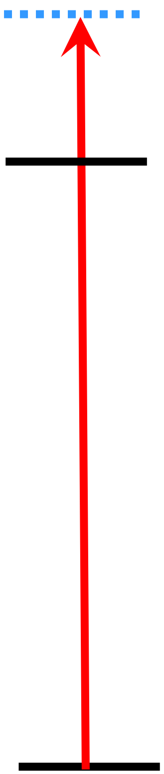
HERAUSGEGEBEN VON
PAUL DRUDE.

MIT FÜNF FIGURENTAFELN.

LEIPZIG, 1905.

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

# Ultrafast Laser Control of Ionization



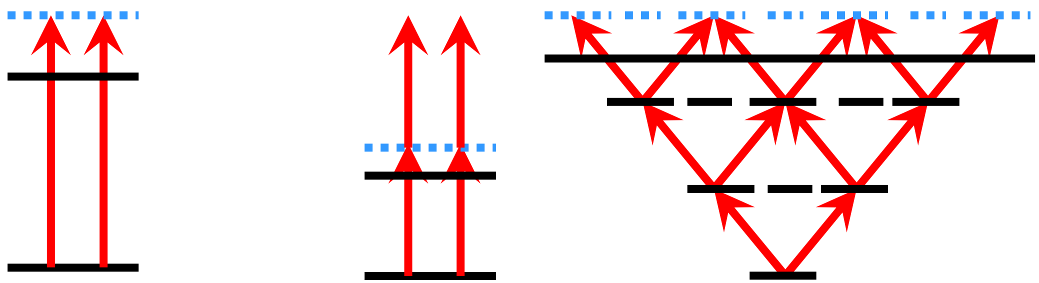
*Erzeugung und Verwandlung des Lichtes.* 145

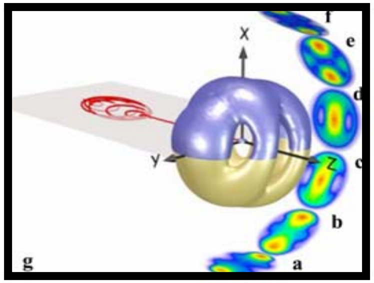
Abweichungen von der Stokesschen Regel sind nach der dargelegten Auffassung der Phänomene in folgenden Fällen denkbar:

1. wenn die Anzahl der gleichzeitig in Umwandlung begriffenen Energiequanten pro Volumeneinheit so groß ist, daß ein Energiequant des erzeugten Lichtes seine Energie von mehreren erzeugenden Energiequanten erhalten kann;

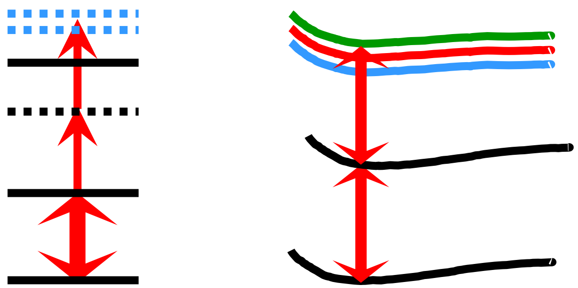
**Deviations if more than one photon is involved !**

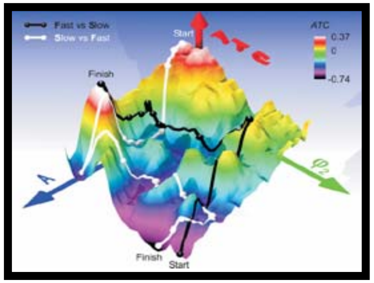
# I. Weak Field (perturbative) Coherent Control (K)



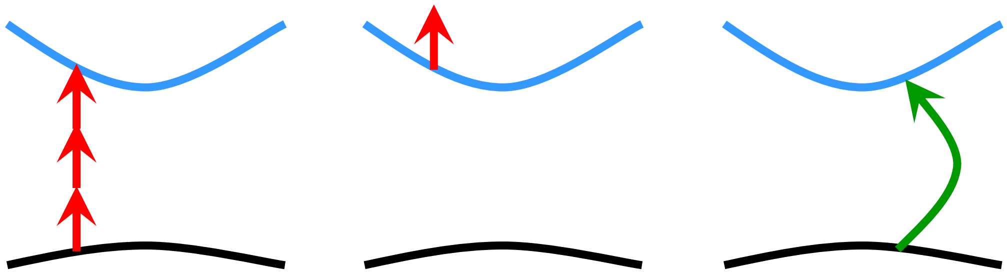


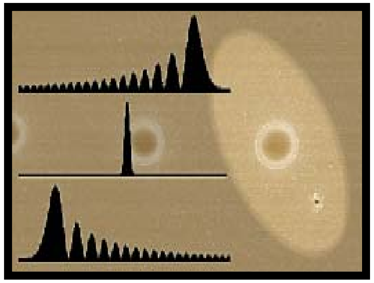
# II. Strong Field (non perturbative) Coherent Control (K, $K_2$, Na)



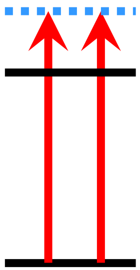
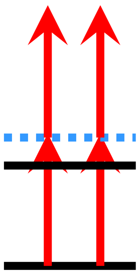
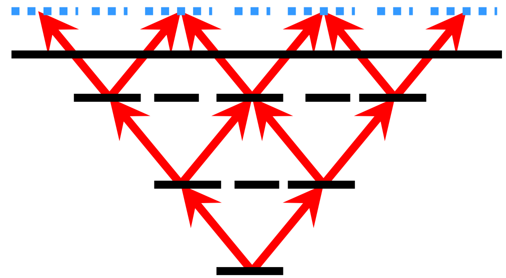
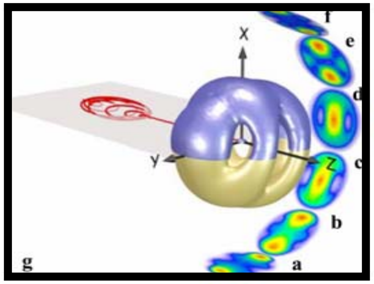


# III. Strong Field Incoherent Control (Dielectrics)





# I. Weak Field (perturbative) Coherent Control (K)

**Main Focus on Free Electron Interference**

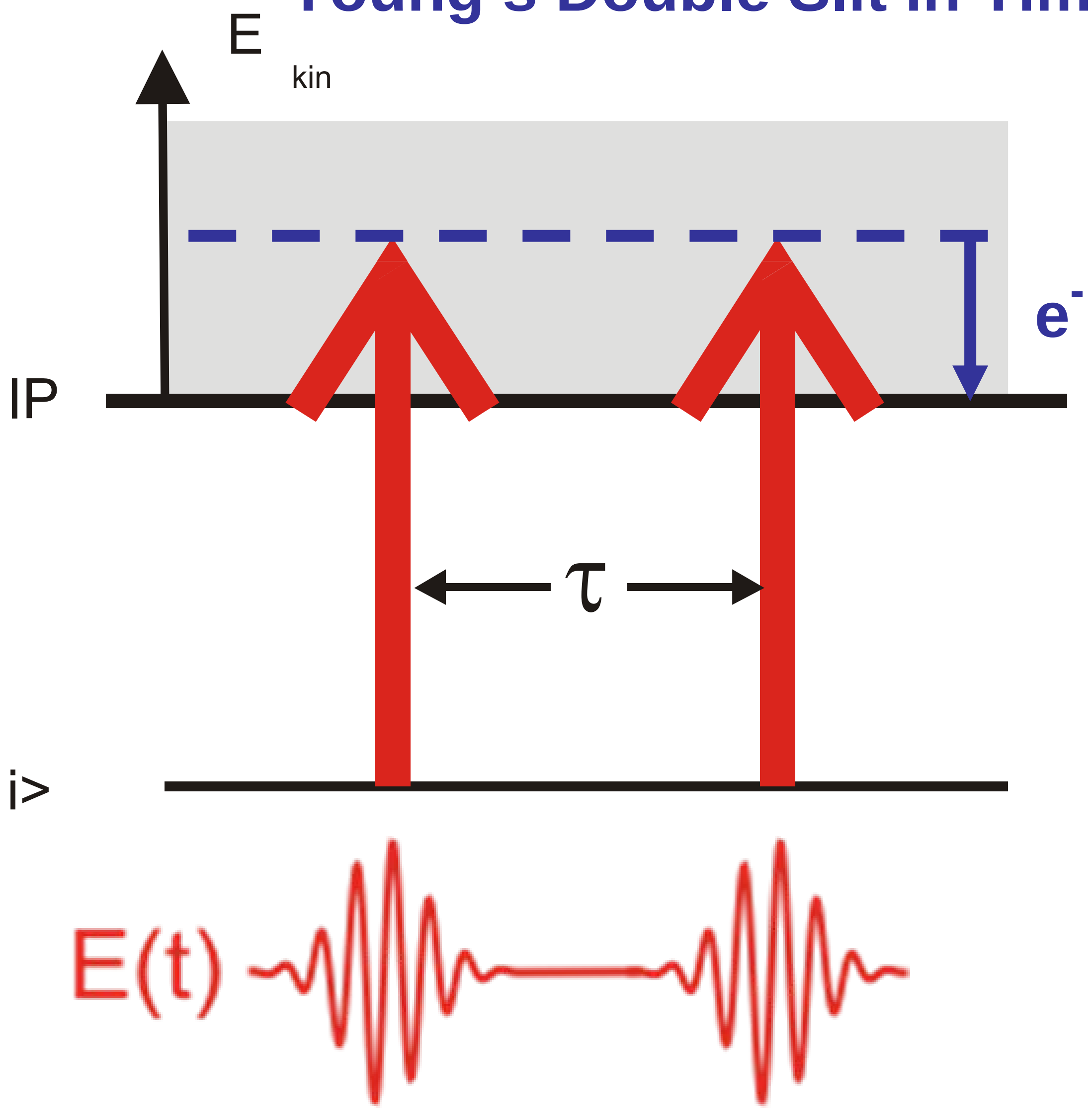
**Young's Double Slit in Time Domaine**

**Pulse Characterization on ATI**

**Possible Route for Molecular Identification**

**3D Wave Packet Sculpturing and Tomography**

# Free Electron Interference: Young's Double Slit in Time Domaine

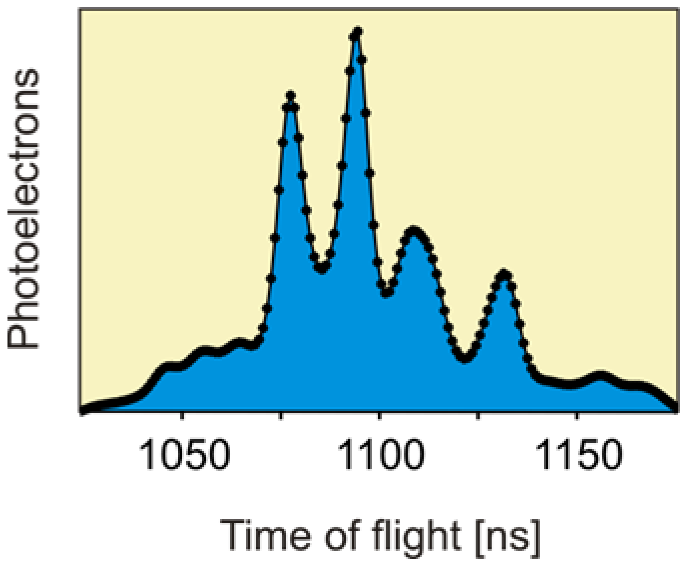


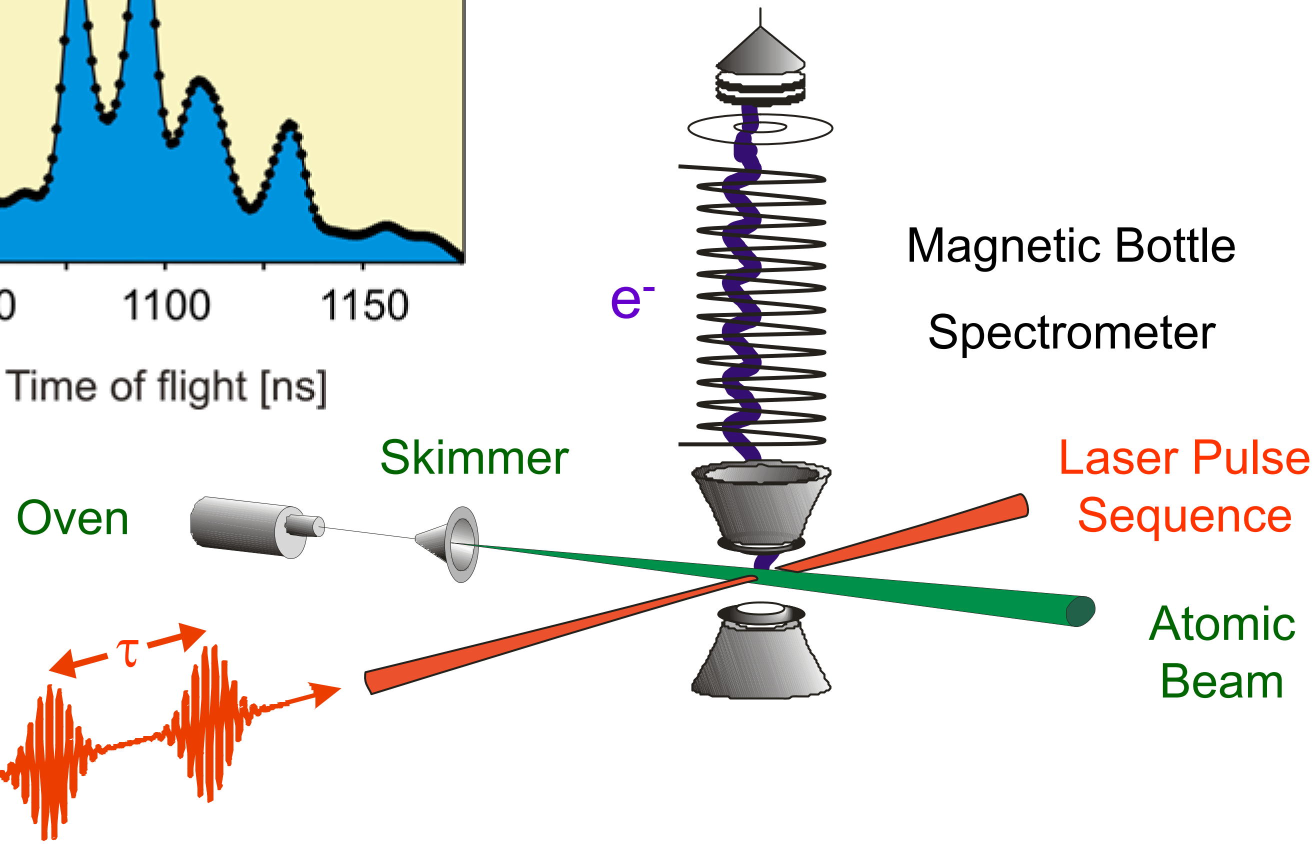

λ = 800 nm,

FWHM = 25 fs

i> = K(5p) via 405 nm laser preparation

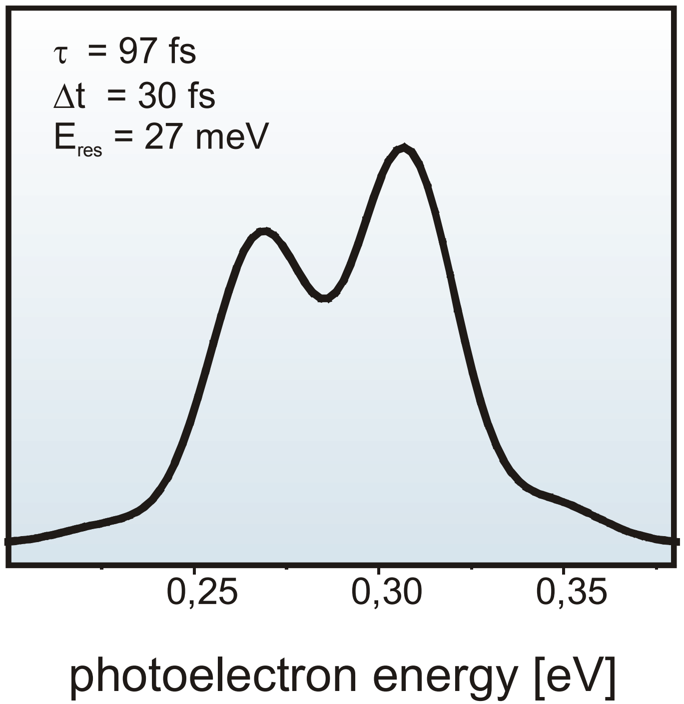
# Free Electron Interference: Experimental Setup



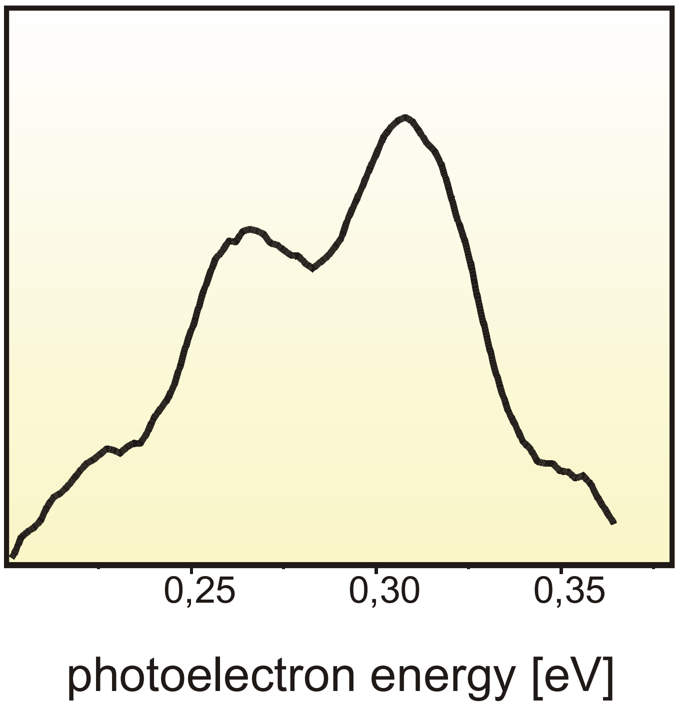



# Free Electron Interference: Photoelectron Spectra

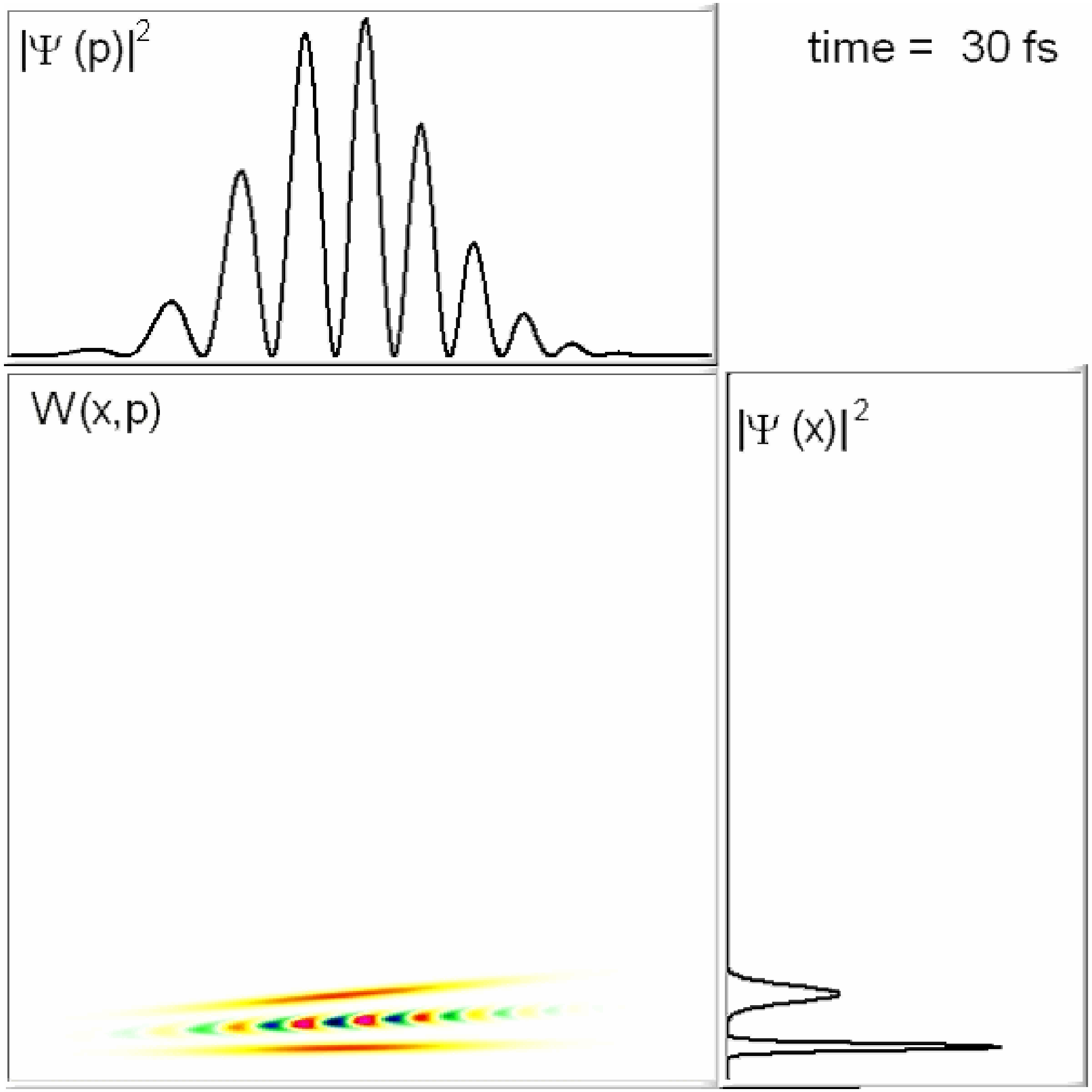
calculated

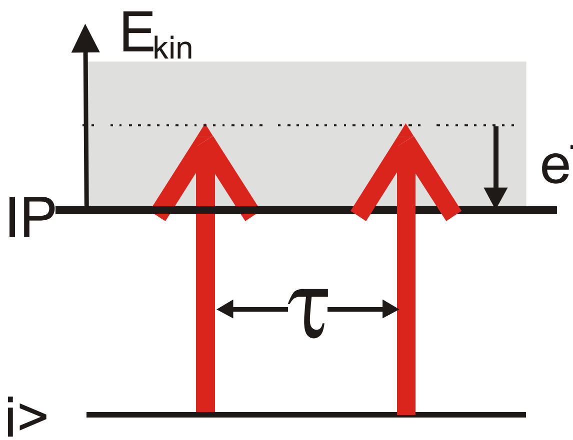



measured



# More than Spectral Interference: a Wigner Description

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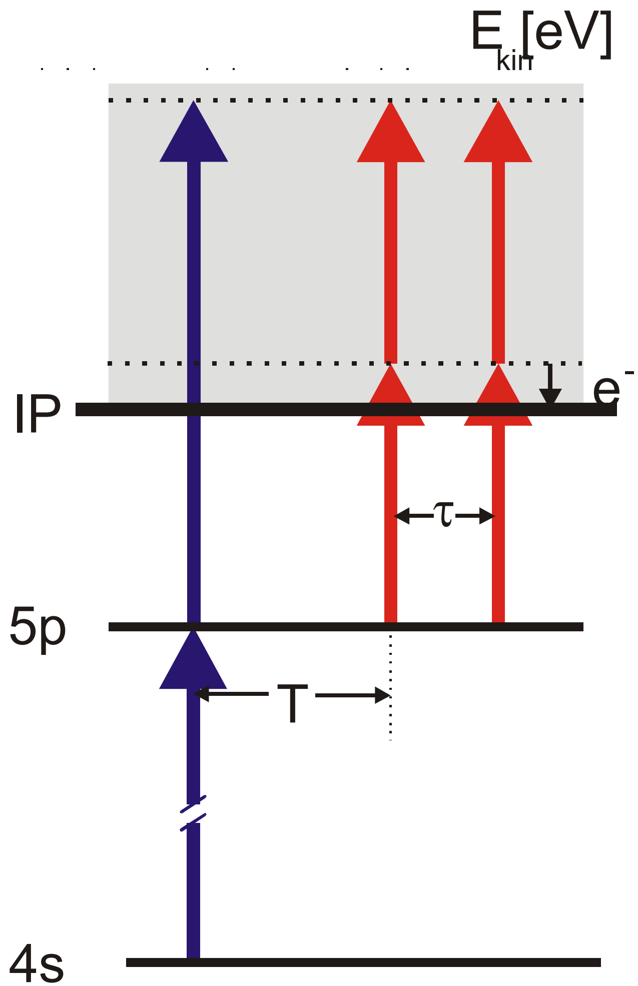
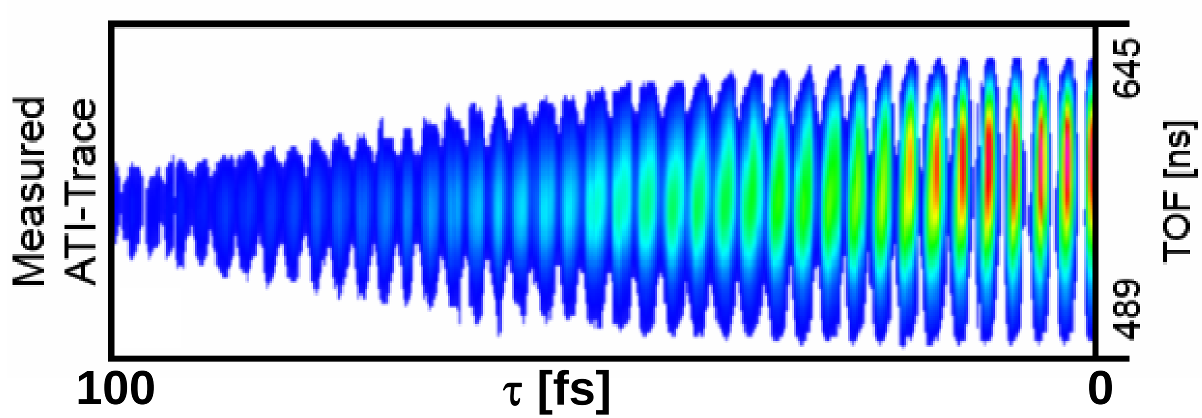
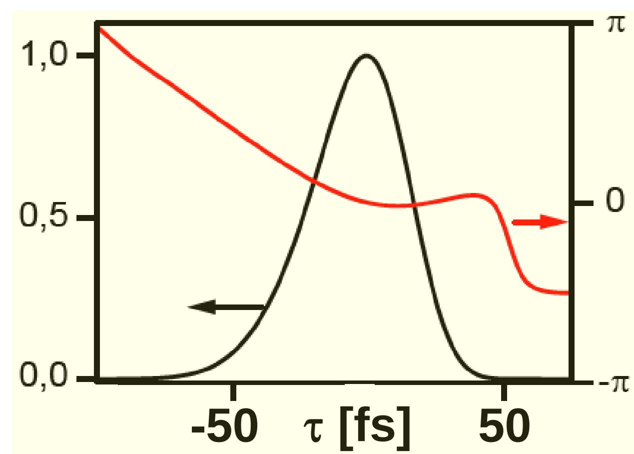
# Pulse Characterization on ATI (implementable in X-UV)




♦ = 42+/- 3 fs $\phi_2$= 326 fs$^{-2}$

τ =100 fs, λ = 405 nm, E = 0,25 µJ, T = 4 ns

♦ =30 fs, λ = 790 nm, E = 1 µJ, I < $10^{12}$ W/cm$^2$

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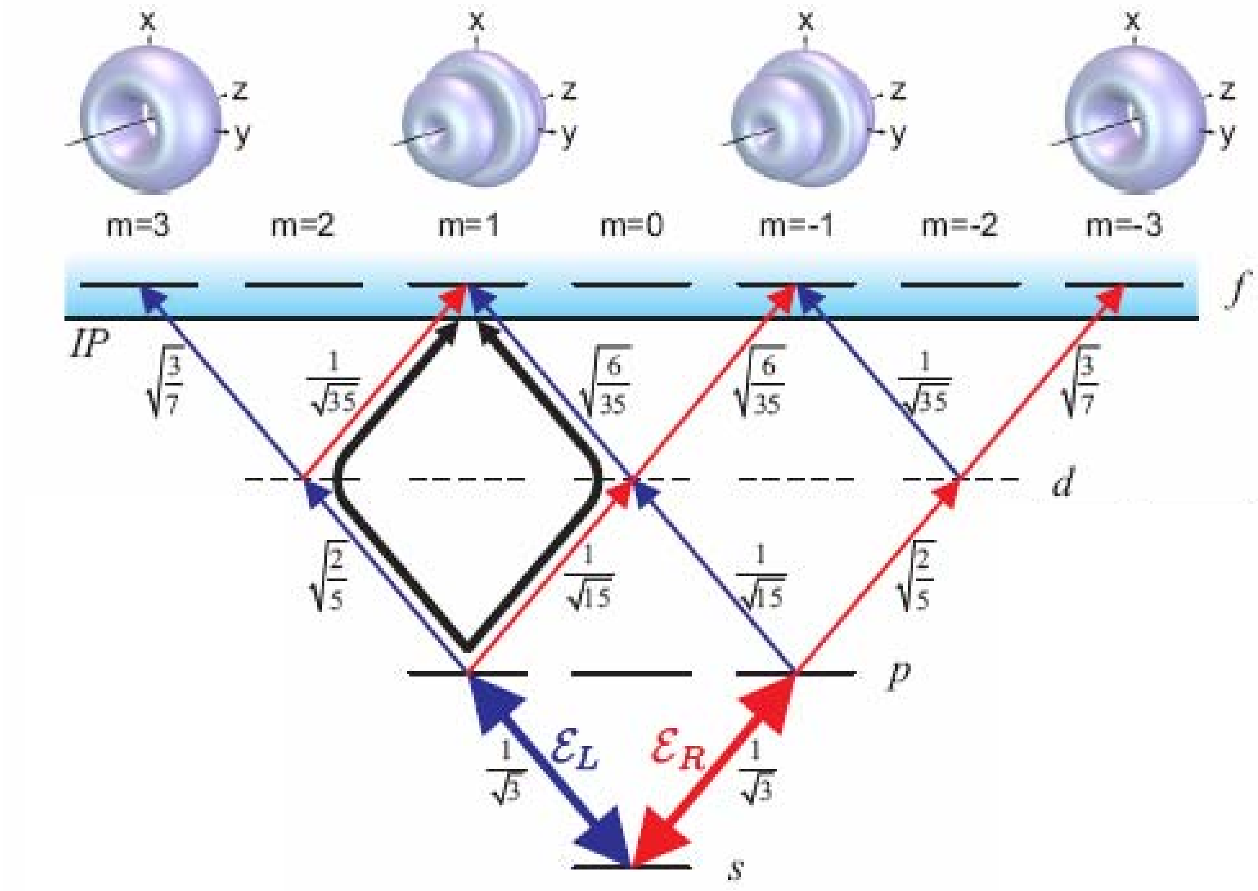
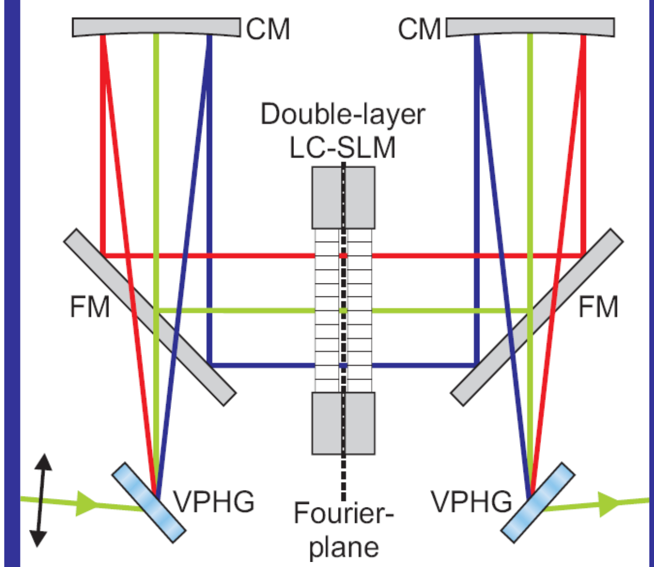
# Polarization Shaped Laser Pulses, Multi Photon Transitions and Photoelectron Angular Distributions (K)

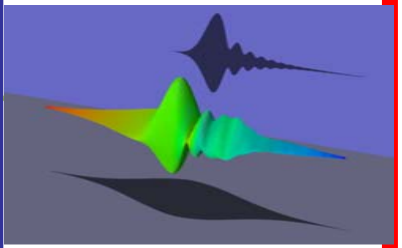



## …Because of Many Interfering Pathways in REMPI

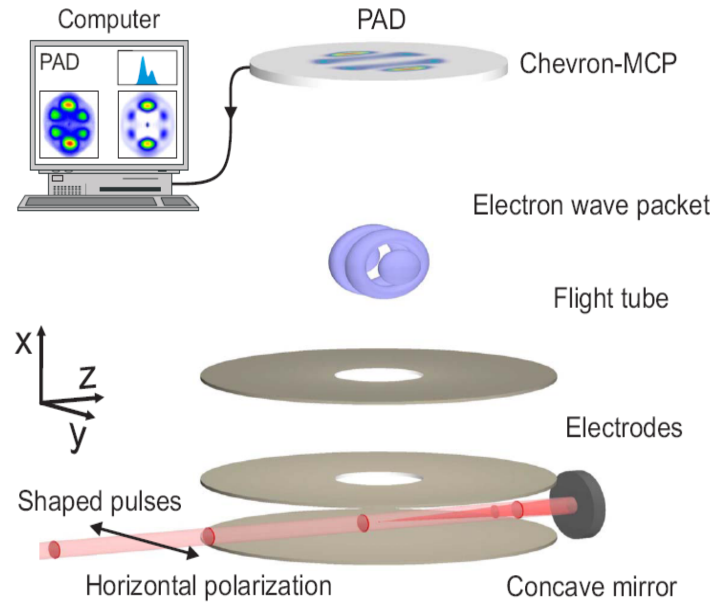
# Experimental Setup

## *Polarization Shaping*



## *Photoelectron Angular Distribution Imaging*

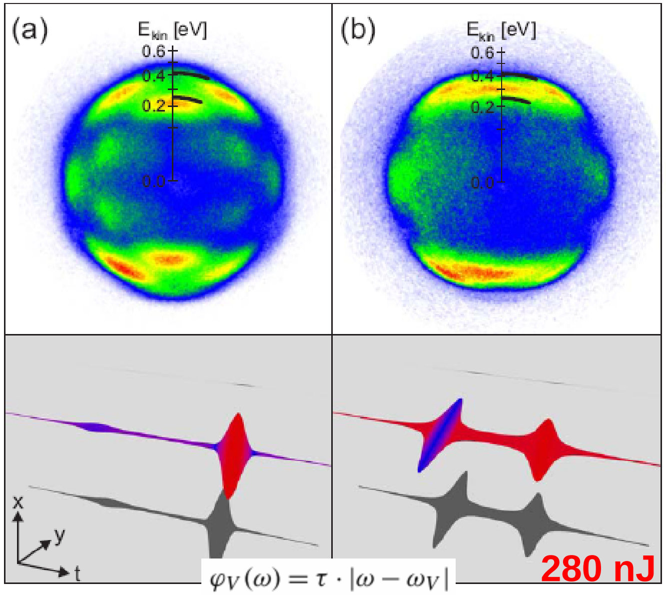


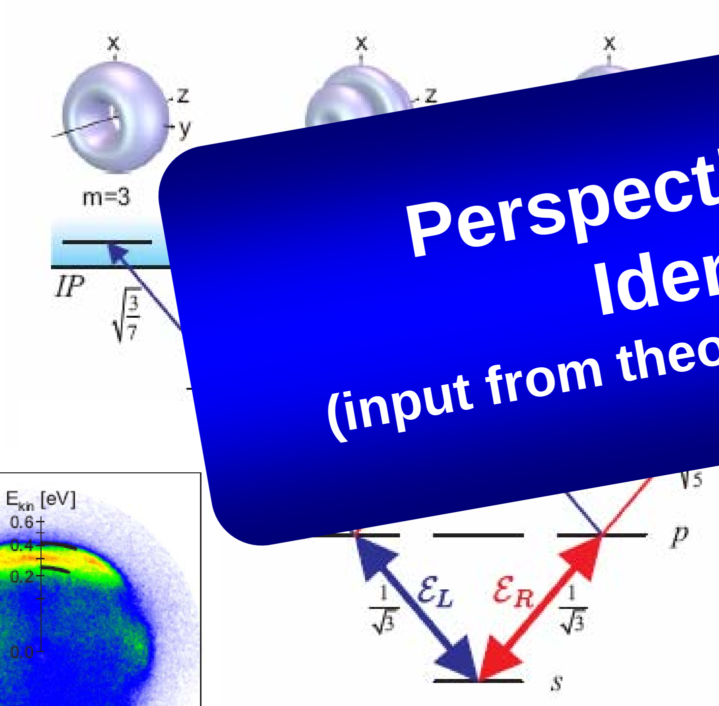

Velocity Map Imaging: Chandler, Houston, Bordas, Helm, Eppink, Parker, Vrakking..

# …Because of Many Interfering Pathways in REMPI

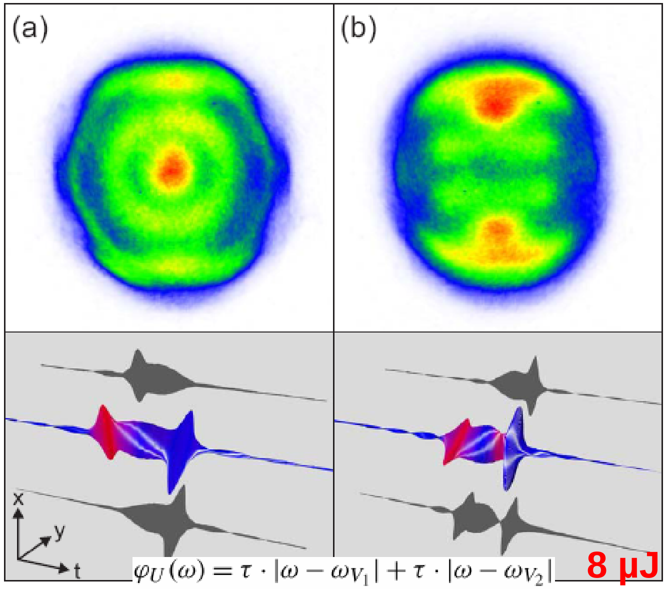
**Perspective: Molecular Identification**

**(input from theory would be appreciated)**



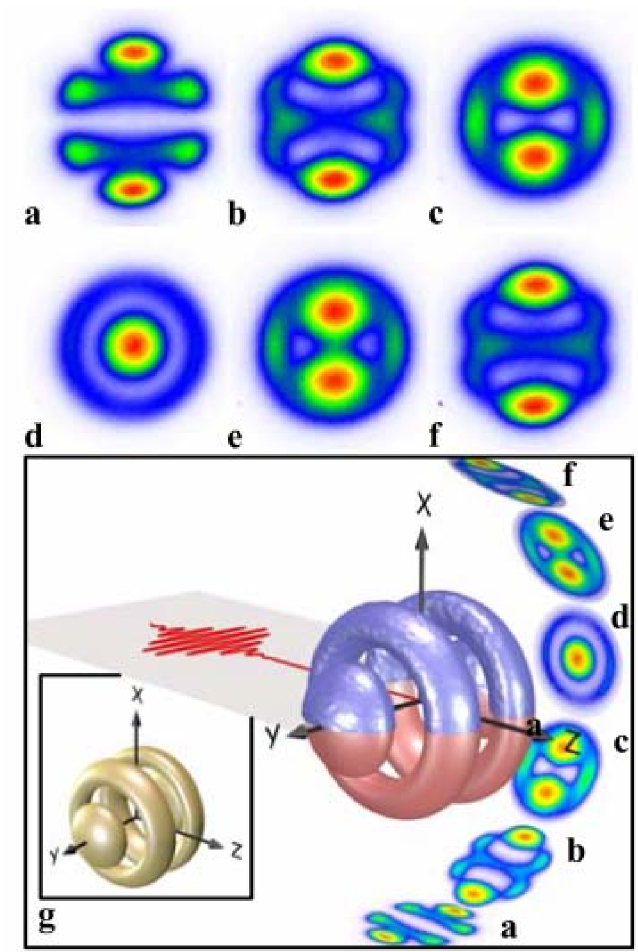

**Potassium Atoms**

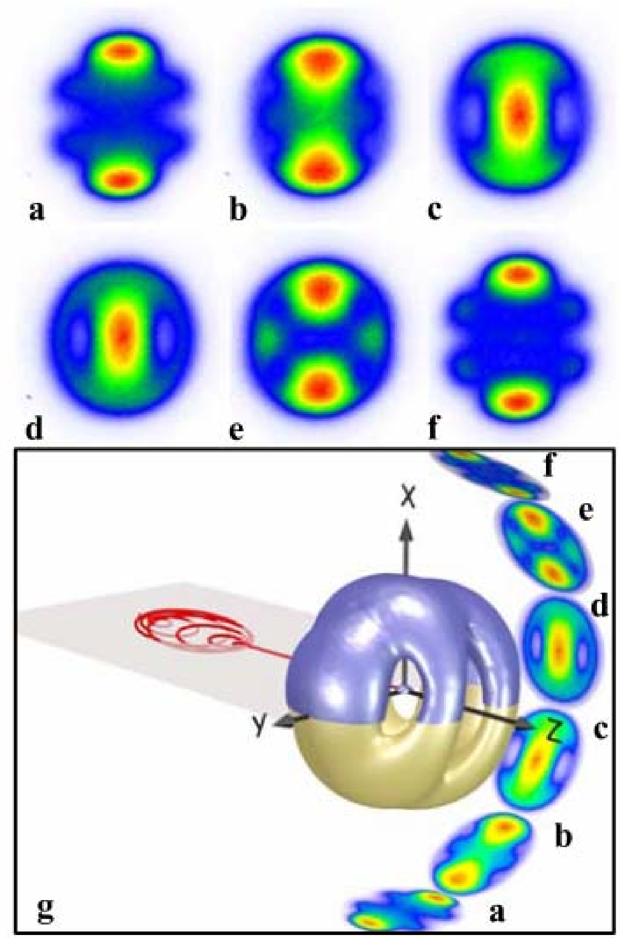



Same linear spectrum

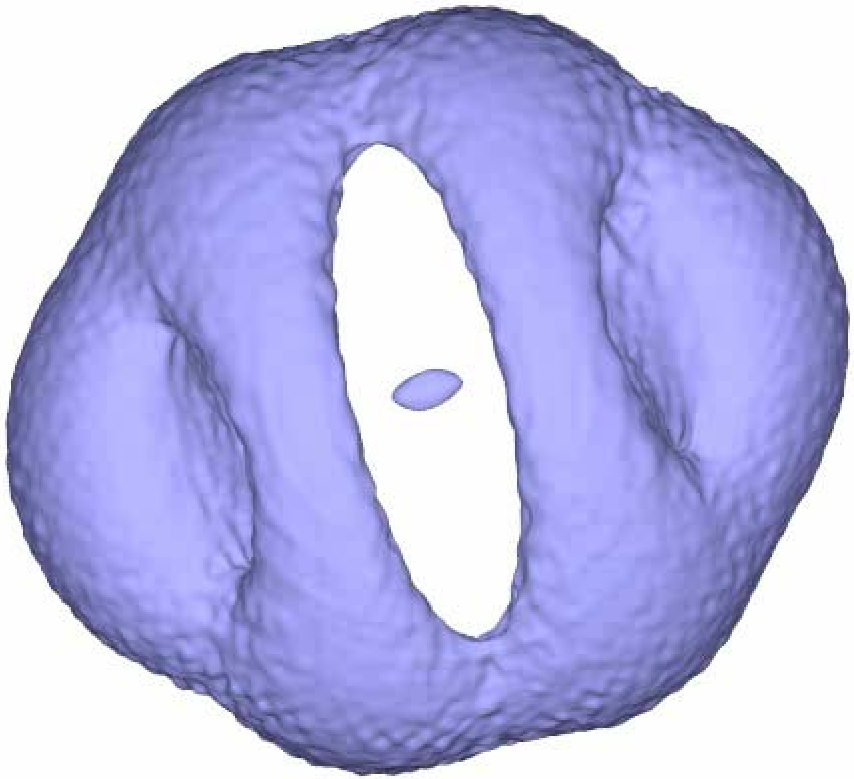
APB **95** (2009) 245

# No Abel Inversion for Polarization Shaped Interaction: Tomographic Reconstruction of Sculptured 3D Electron Distributions



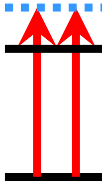



# Tomographic Reconstruction of Sculptured 3D Electron Distributions (here: elliptically polarized light)

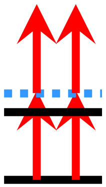


# SUMMARY

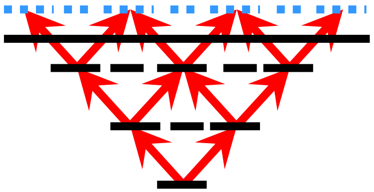
## I. Weak Field (perturbative) Coherent Control (K)



**Young's Double Slit in Time Domaine**

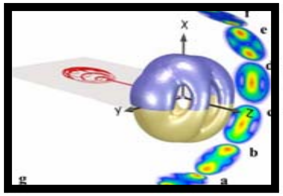


**Pulse Characterization on ATI**



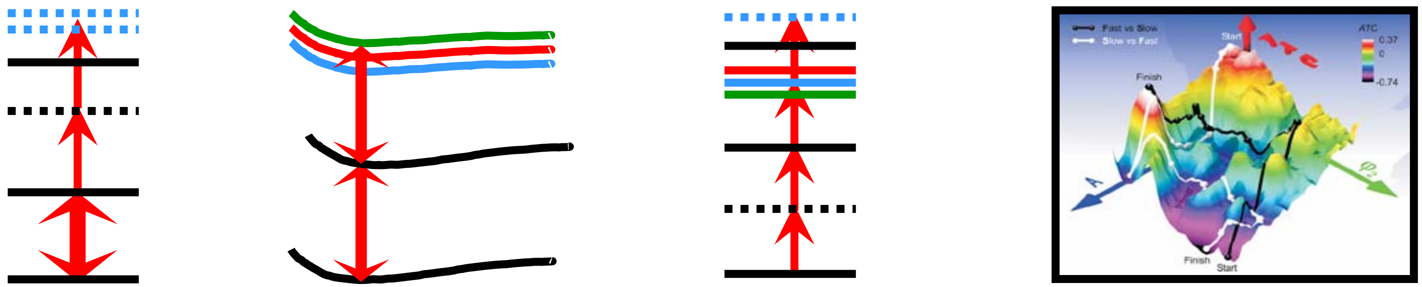
**Polarization Shaping + PADs:**

**(i) Possible Route for Molecular Identification**



**(ii) 3D Wave Packet Sculpturing and Tomography**

# II. Strong Field (non perturbative) Coherent Control (K, $K_2$, Na)



**Main Focus on Neutral States Dynamics**

**Unravel Physical Mechanisms Driving Strong Field Control with Shaped Laser Pulses**

**Resonant Processes Dominate Control Scenarios for (ultra) Broad Spectra**

**Strong Field Schemes are Efficient**

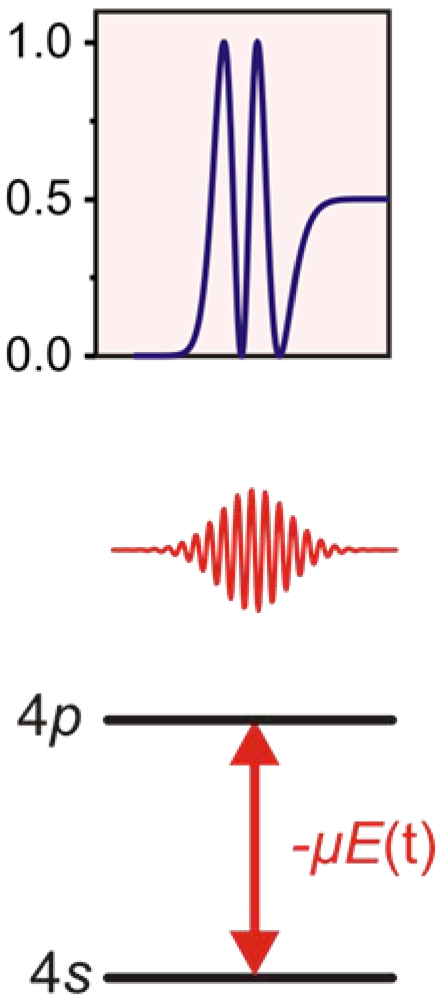
**Ultrafast Control of Coherent Electronic Excitation in Atoms and Molecules**

**Control of Multiple States by a Single Chirped Pulse**

**Parameterizations of Strong Field Control (Landscapes)**

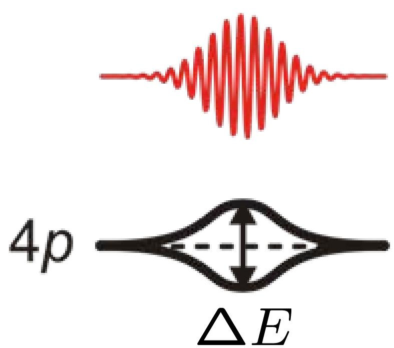
# Ultrafast Control of Coherent Electronic Excitation (K)

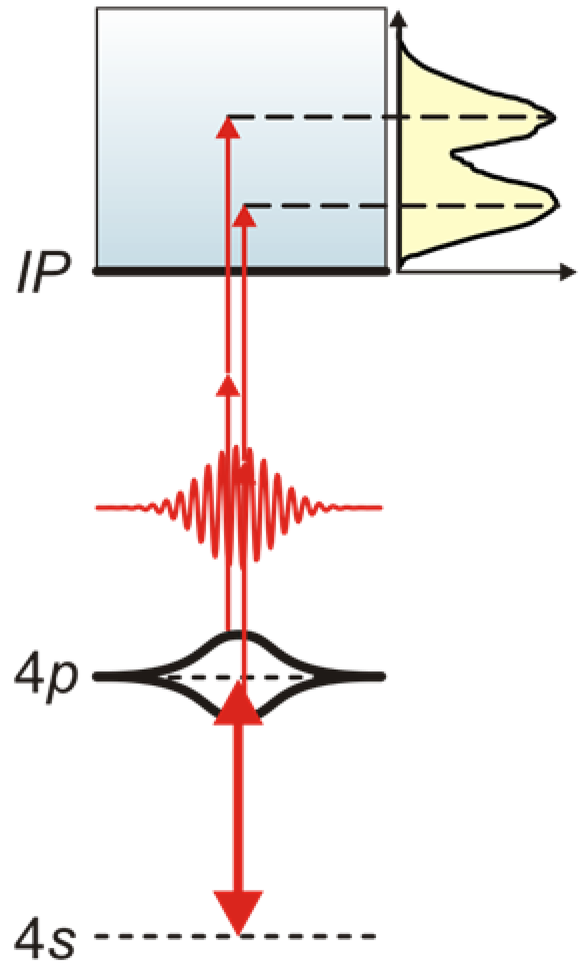
Bare States | Dressed States | Photoelectrons probe Dressed States




$$\Delta E = \hbar\Omega(t) = \mu E(t)$$

$$\Delta E[\text{eV}] = \frac{4.14}{T_R[\text{fs}]}$$



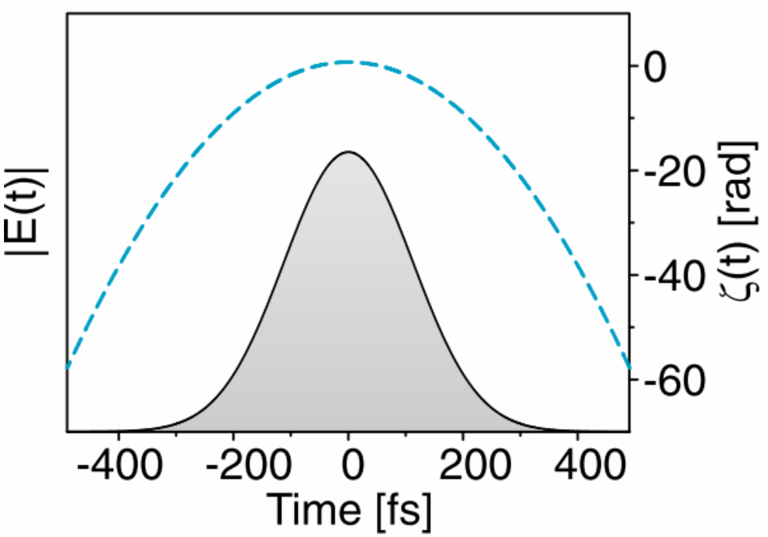

Annu. Rev. Phys. Chem **56** (2005) 25

**Selective Population Of Dressed States (SPODS)**

# Strong Field Control via SPODS Requires Temporal Phase Variations*

**continuous**

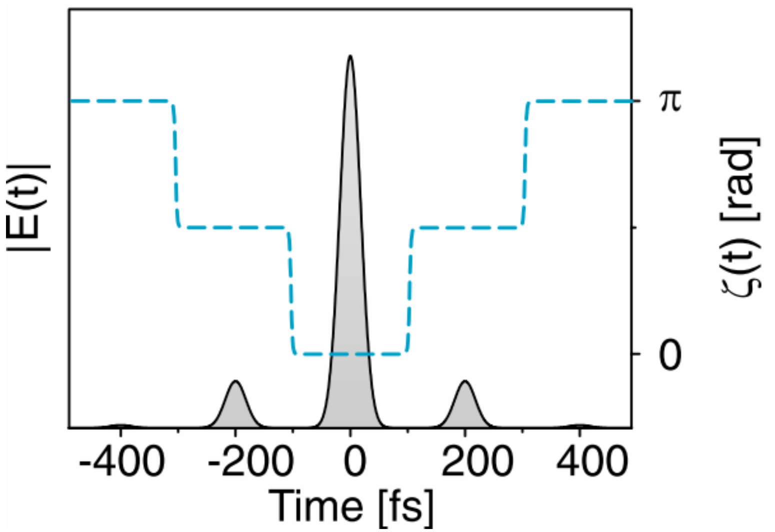
$\varphi(\omega)=\varphi_2(\omega-\omega_0)^2$




**Rapid Adiabatic Passage**

**discontinuous**

$\varphi(\omega)=A\sin[(\omega-\omega_0)T+\phi]$

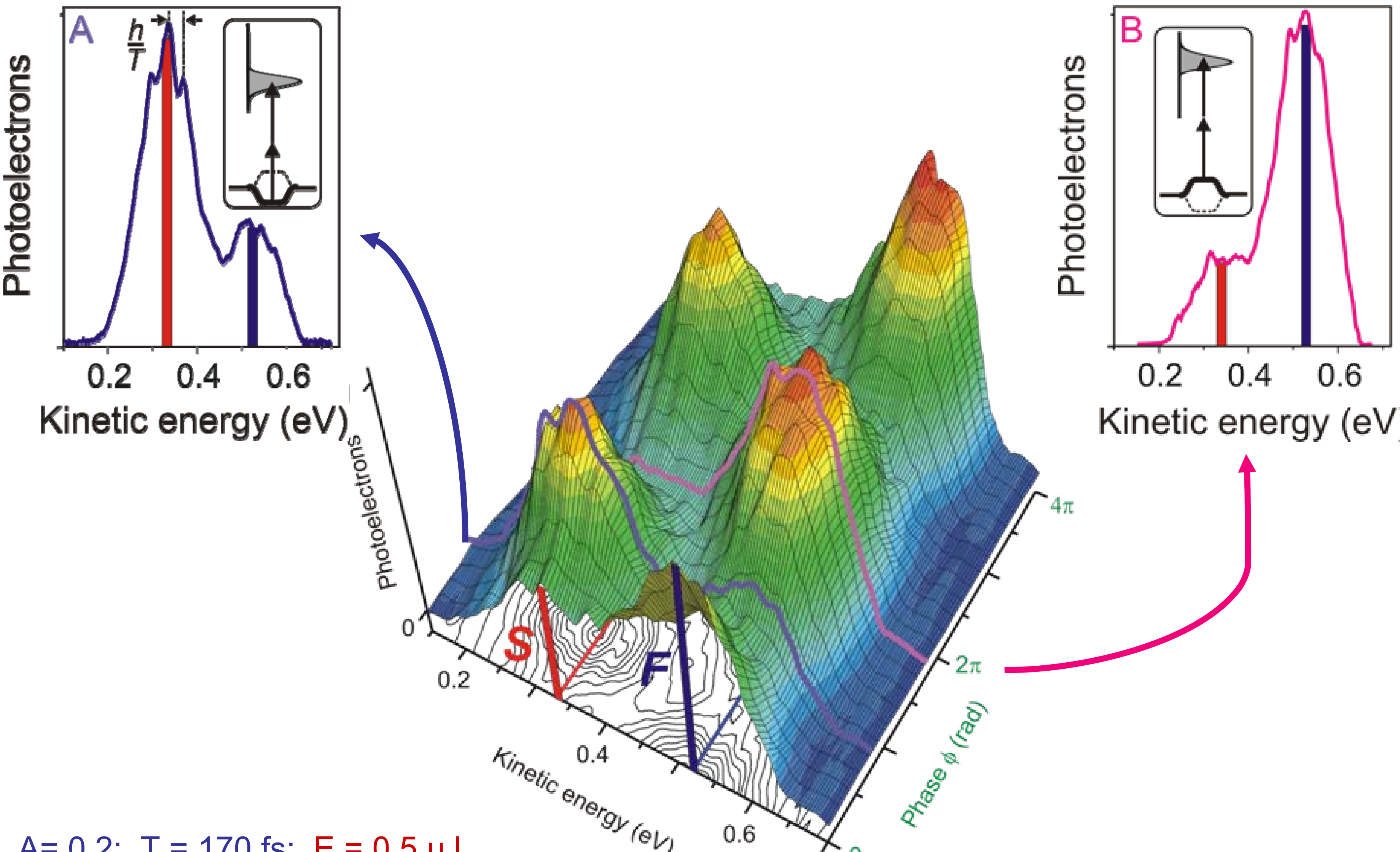



**Photon Locking**

**Relative temporal phase between subpulses is controlled with attosecond precision**

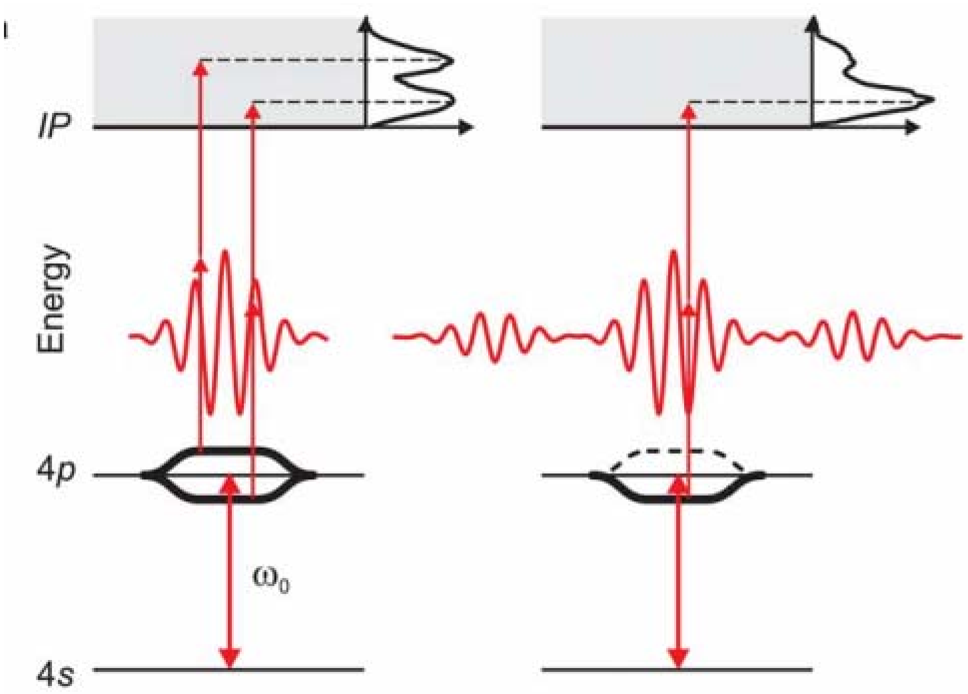
*“Real“ Electric Fields extend weak field control schemes (Silberberg PRL (2005))

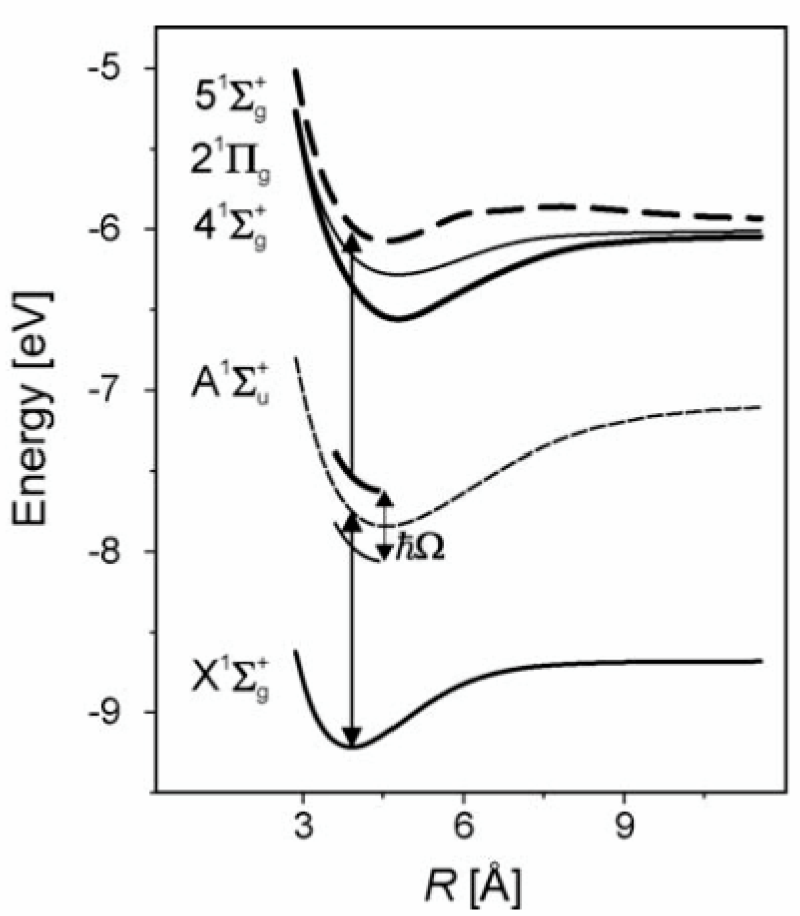
# Experimental Results (K)




A= 0.2; T = 170 fs; E = 0.5 μJ

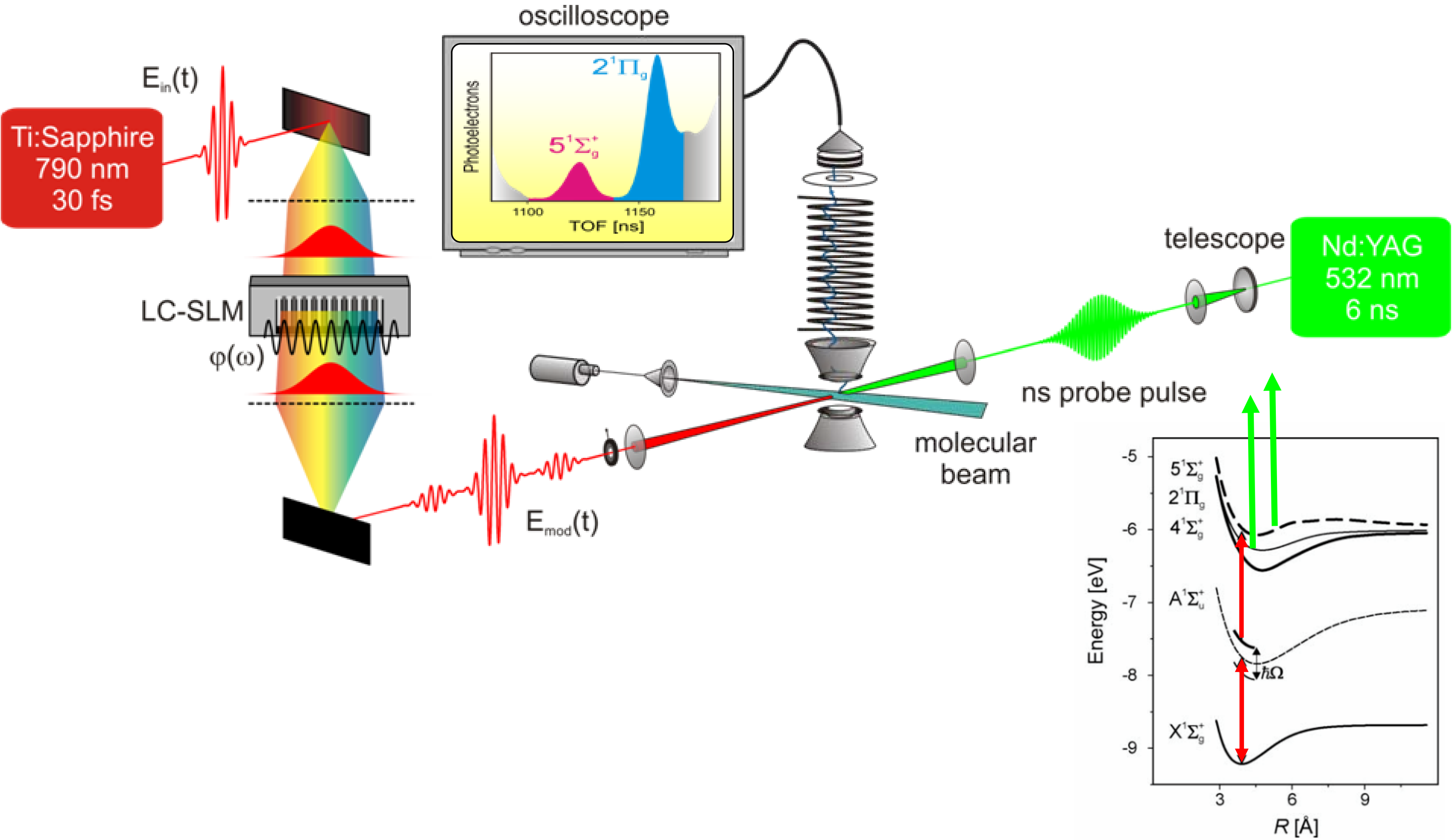
# Ultrafast Control of Coherent Electronic Excitation in Molecules: $K_2$



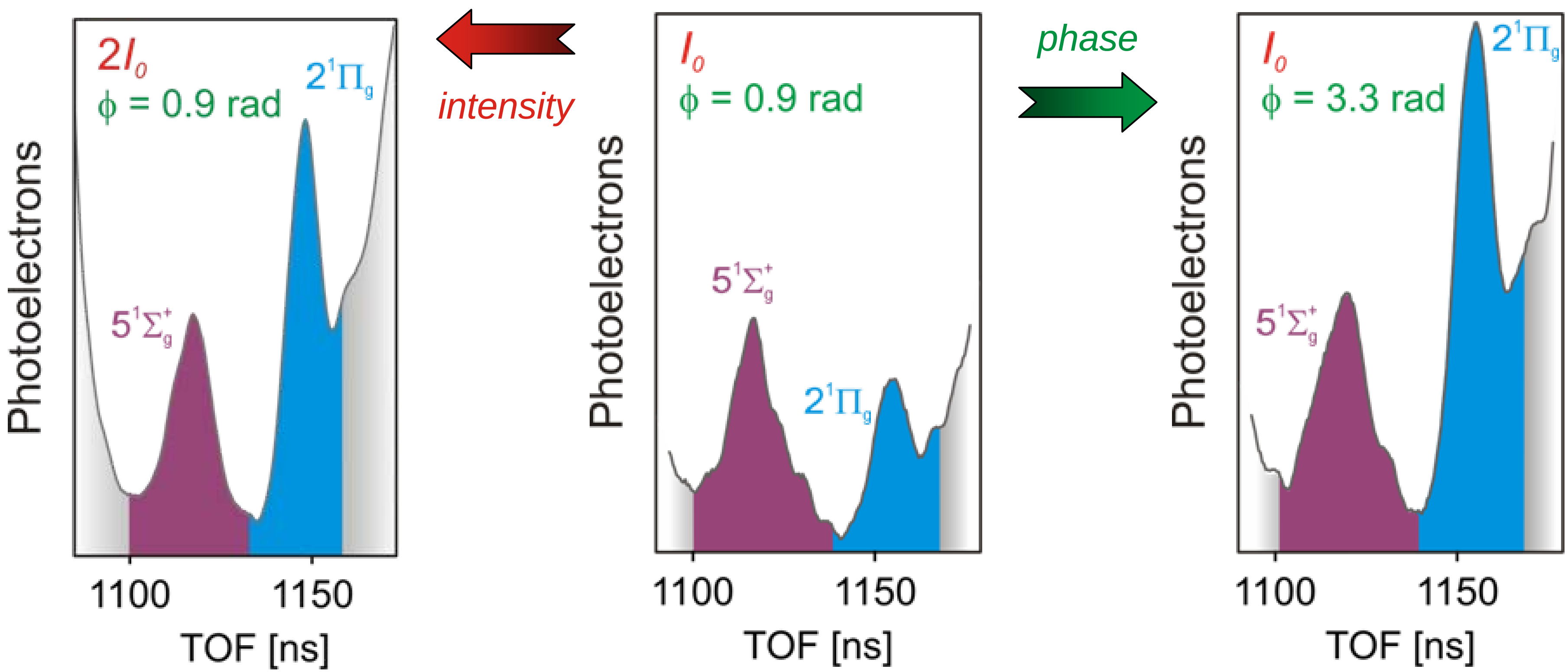




THEORY: CPL **419** (2006) 184 J. Photochem. Photobiol. A **180** (2006) 248 

# SPODS On $K_2$ Molecules: Experimental Setup



# Preliminary Experimental Result on $K_2$: Control Via *Phase* And *Intensity*

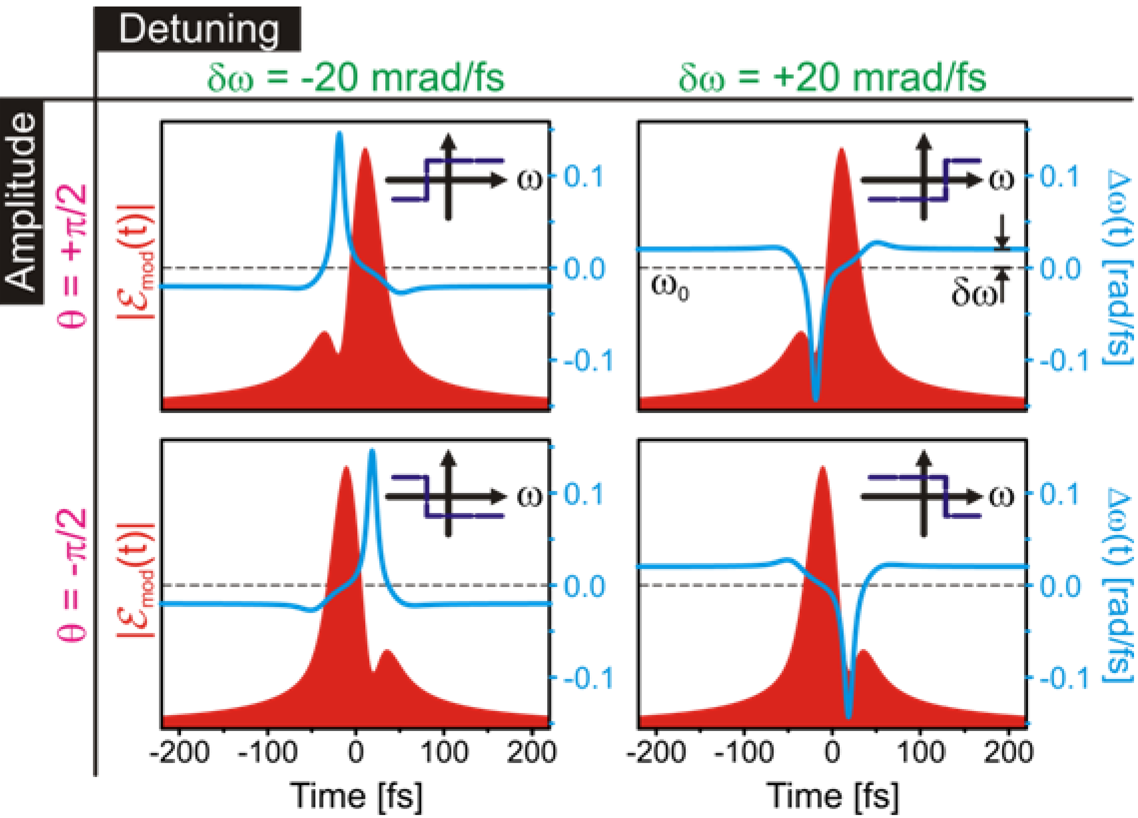


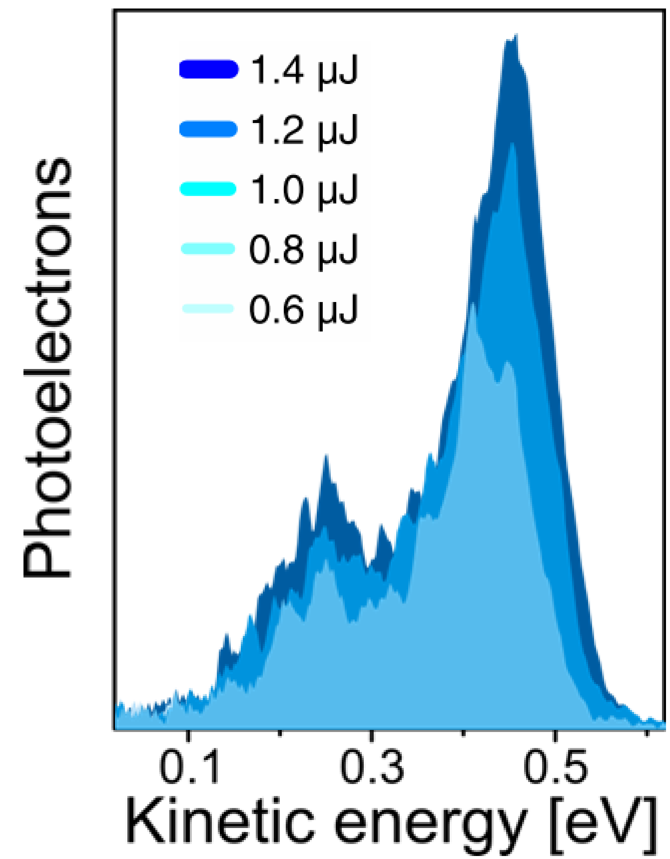

Signal times 0.5

Intensity and phase act in same manner: Phys. Rev. A **68** (2003)

# Robust Photon Locking via Generalized θ-Step (K)

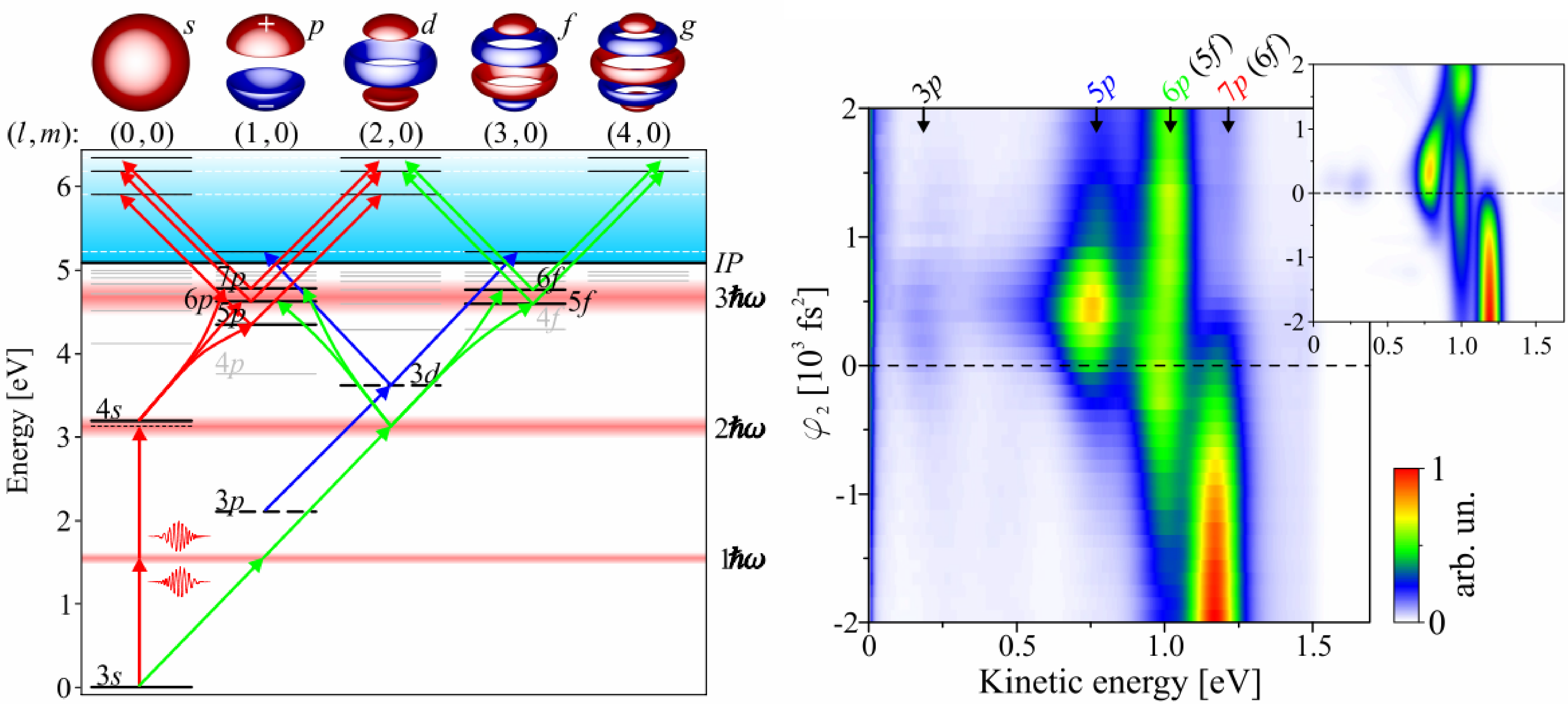
$$\tilde{\mathcal{E}}_{mod}(\omega) = \tilde{\mathcal{E}}(\omega) e^{-i\frac{\theta}{2}\sigma(\omega-\delta\omega)}$$




PRL **102** (2009) 023004

# Coherent Strong Field Control of Multiple States in Na by a Single Chirped Femtosecond Laser Pulse

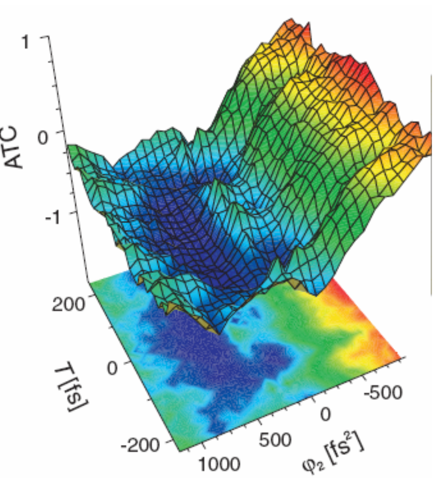


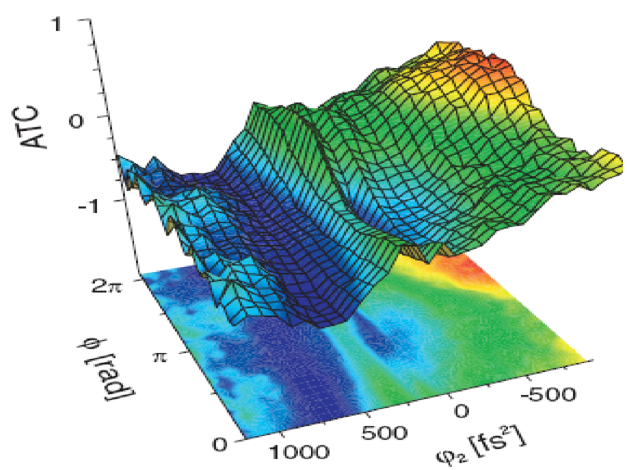
# Suggested Strong Field Parameterization for Adaptive Control Experiments

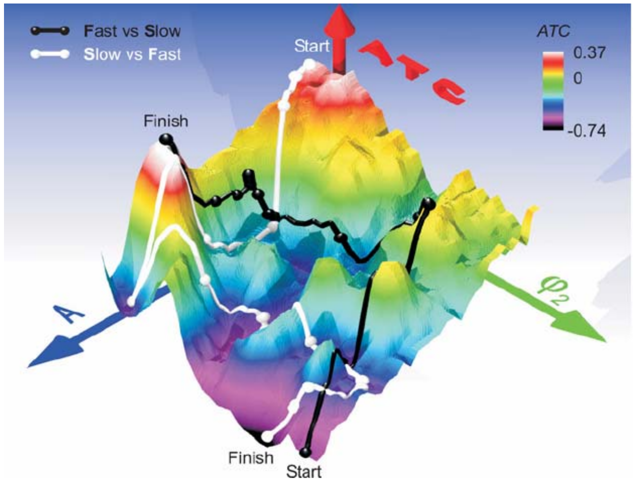
- **based on complementary physical mechanisms (RAP vs. PL):**

$$\varphi(\omega) = \varphi_2\ (\omega-\omega_0)^2\ +\ A\ \sin[T\ (\omega-\omega_0)+\phi]$$ and **Intensity**

- **both are two sides of same coin (SPODS)**
- **tested experimentally and theoretically on Strong Field Control Landscapes**

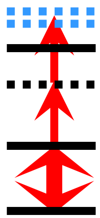


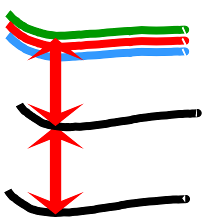



# SUMMARY

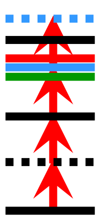
## II. Strong Field (non perturbative) Coherent Control (K, $K_2$, Na)



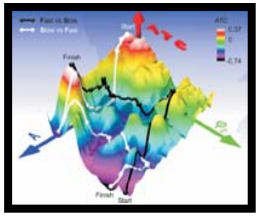
Ultrafast Control of Coherent Electronic Excitation with attosecond Precision in Atoms



and Molecules

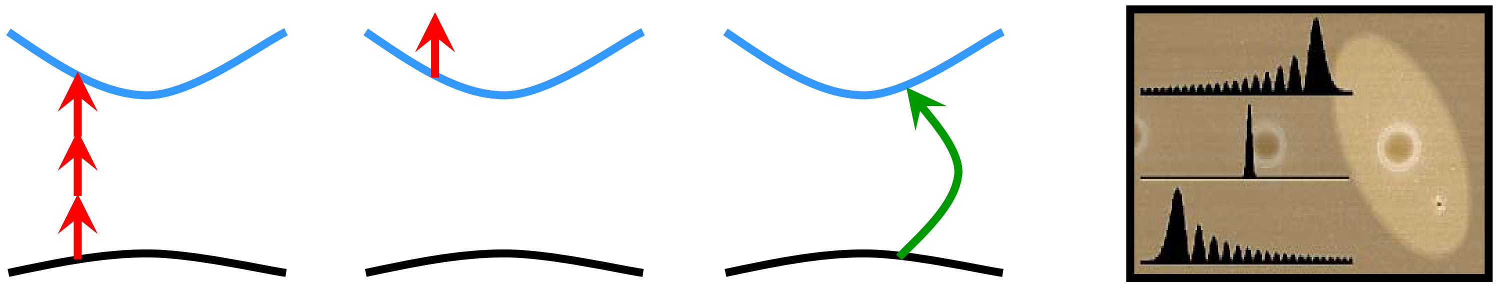


Control of Multiple States by a Single Chirped Pulse



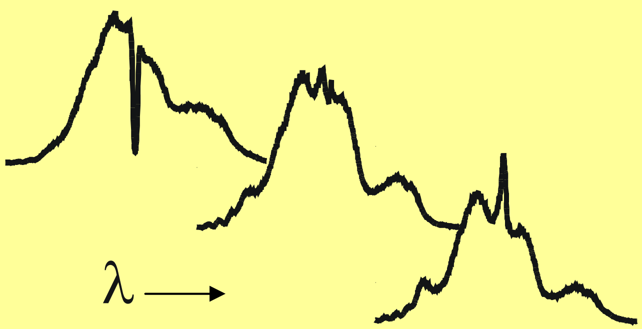
Physical Meaningful Parameterizations of Strong Field Control (Landscapes)

# III. Strong Field Incoherent Control (Dielectrics)



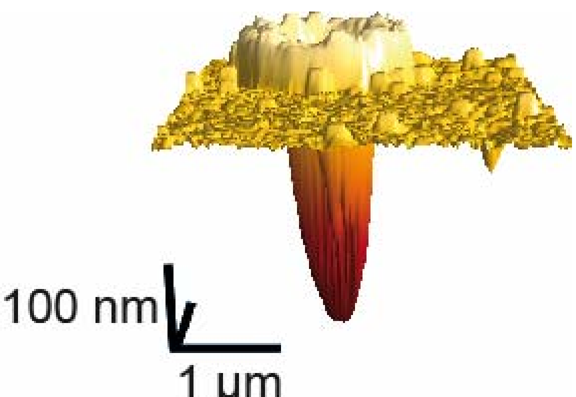
## Direct Nanoscale Laser Processing Of Dielectrics

## SPM Microscopy




Appl. Phys. Lett **78** (2005)

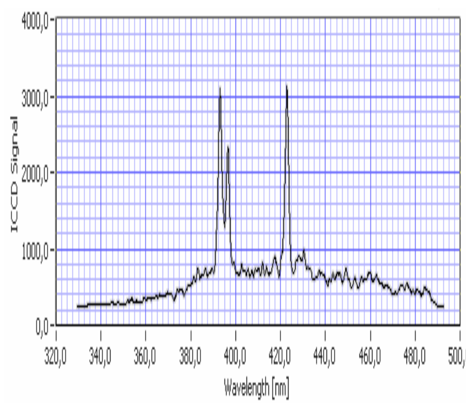

## Nanostructures




**spatial**

Opt. Express **15** (2007)
Appl. Phys. A **92** (2008)

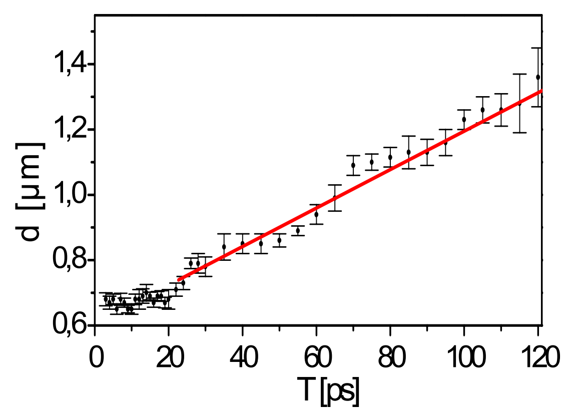
## fs-LIBS




**spectral**

Appl. Phys. B **77** (2003)

## Plasma dynamics



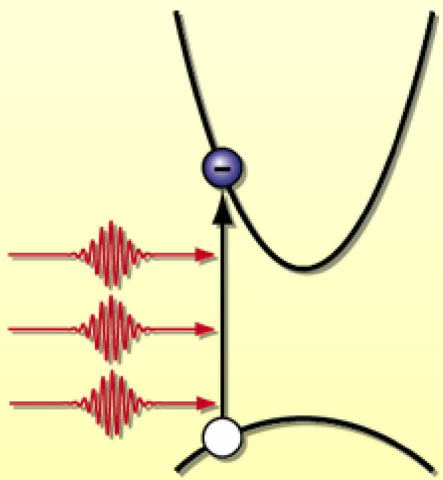

**time**

Appl. Phys. Lett **88** (2006)

NA = 0.5; $1/e^2$ diameter: 1.4 µm; $1/e^2$ length: 9.1 µm

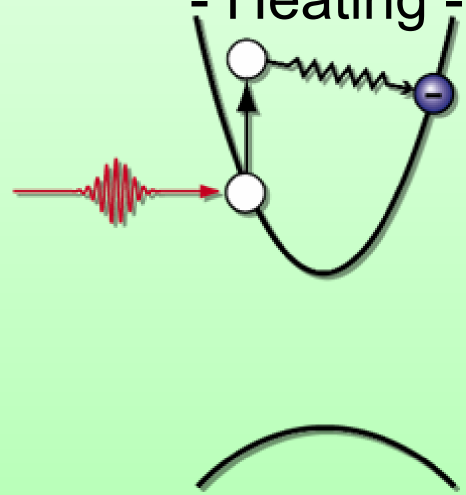
# Basic Ionization Processes in Dielectrics to Reach Critical Electron Energy / Density for Ablation

Multiphoton Ionization

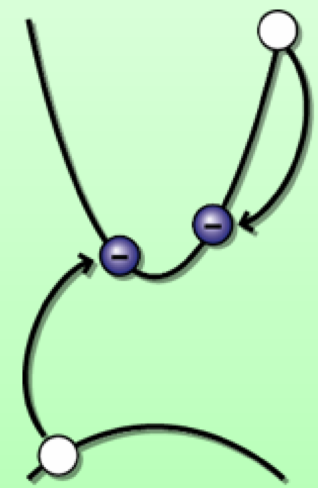


**NEEDS INTENSITY**

Free Carrier Absorption - Heating -



Impact Ionization

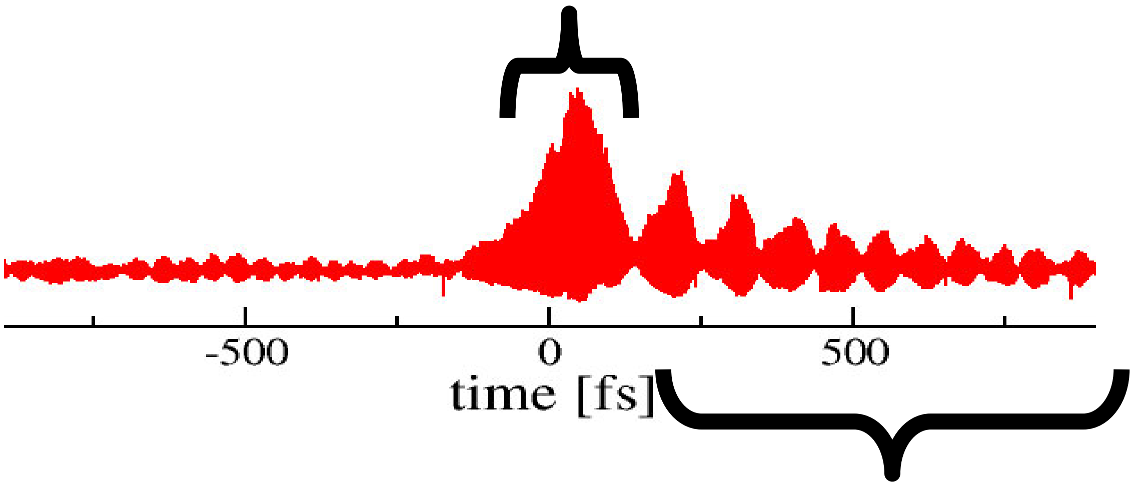


**NEEDS SEED ELECTRONS & TIME**

# Control of Basic Ionization Processes viaTemporally Asymmetric Femtosecond Pulses

$$\phi(\omega) = \frac{\phi_3}{3!} \cdot (\omega - \omega_0)^3$$

Generation of seed electrons
well **below damage threshold** for short pulse ablation
i.e. **strong spatial confinement**

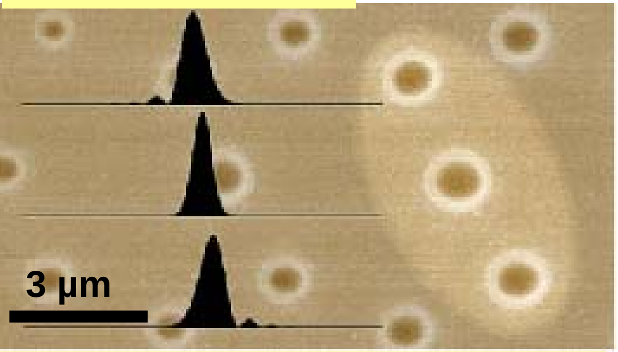



Heating and electron impact ionization
to **reach critical energy / density** for ablation

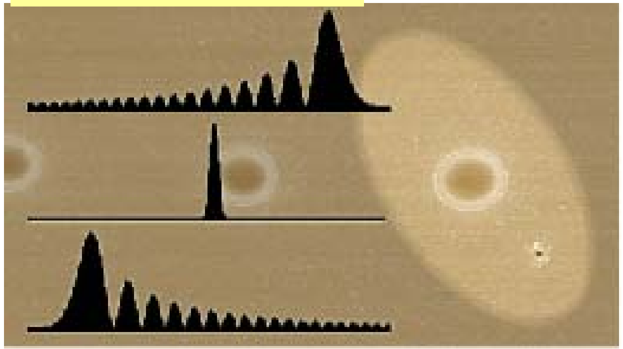
Crosscorrelation of tailored pulses on target, $\phi_3$ = 600 000 $fs^3$

# Reduction In Structure Size Via Pulseshaping

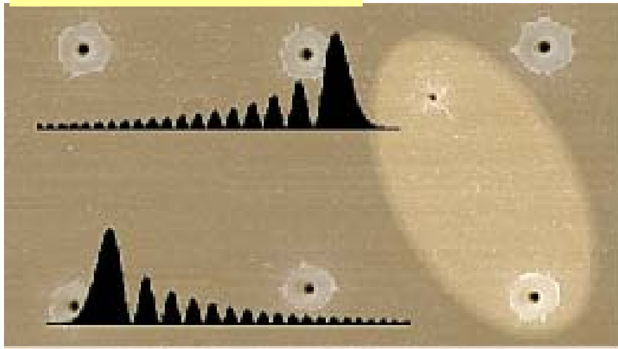
**77 nJ; 2σ = 50 fs**




**71 nJ; 2σ = 960 fs**

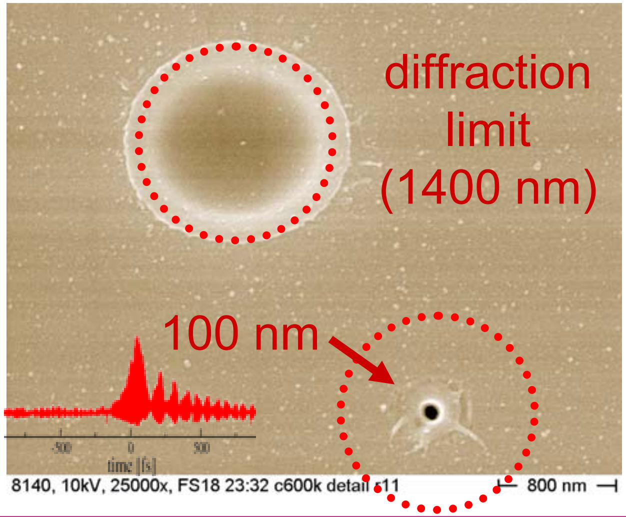


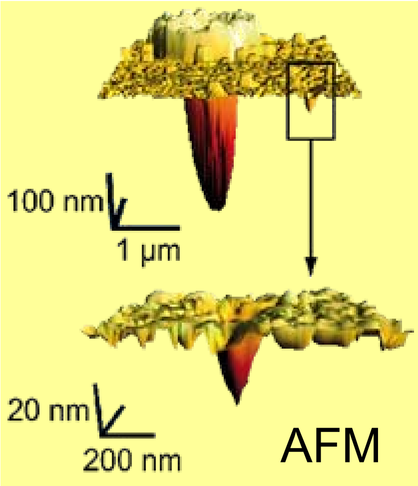
**110 nJ; 2σ = 960 fs**



**SAME FOCUS CONDITIONS**
**SAME FLUENCE**
**SAME SPECTRUM**

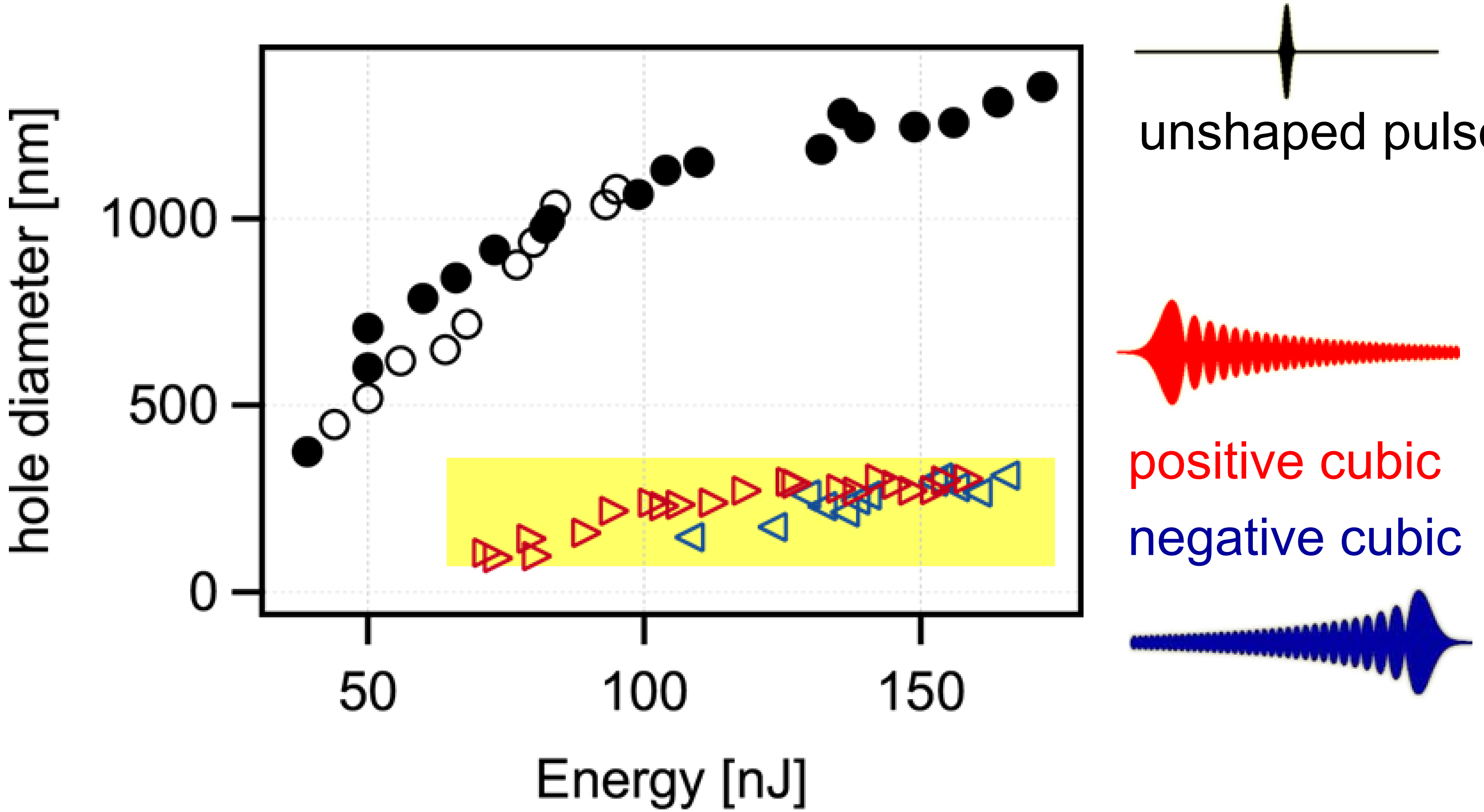
**order of magnitude below diffraction limit!**



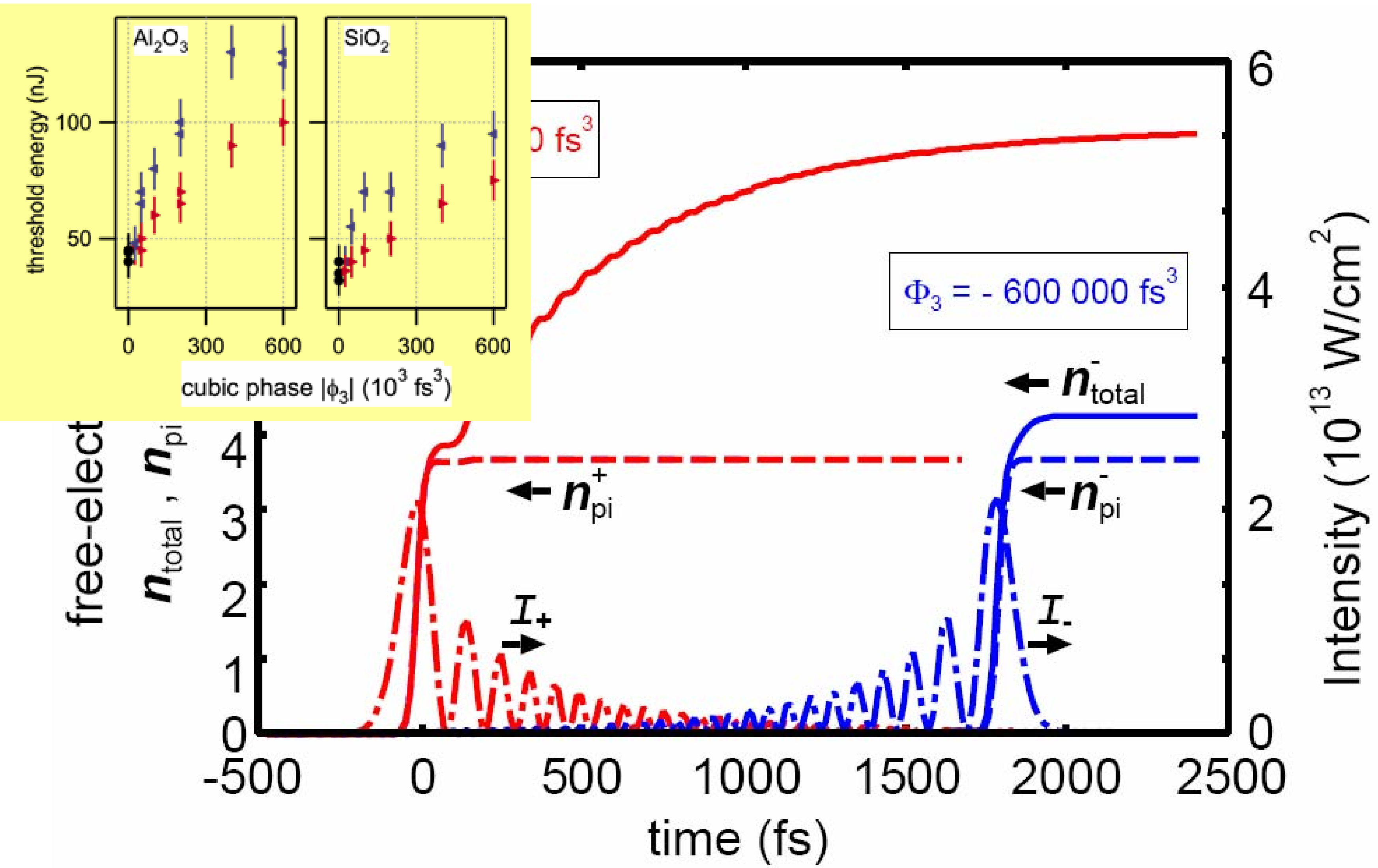

Opt. Express **15** (2007); Appl. Phys. A **92** (2008)

# Structure Size as Function of Fluence




**Nanostructure size robust to variations in laser fluence**

# Simulation of Basic Ionization Processes

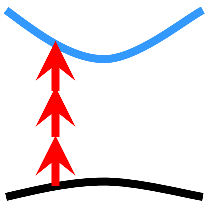



calculated with multiple rate equation: B. Rethfeld PRL 92, 187401 (2004).

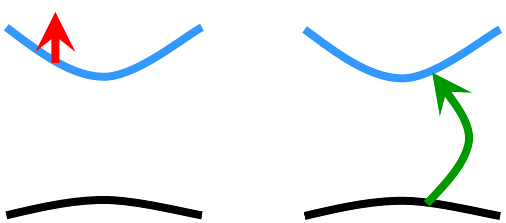
# SUMMARY

## III. Strong Field Incoherent Control (Dielectrics)

**Control of**

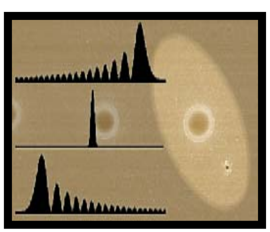


**MPI**



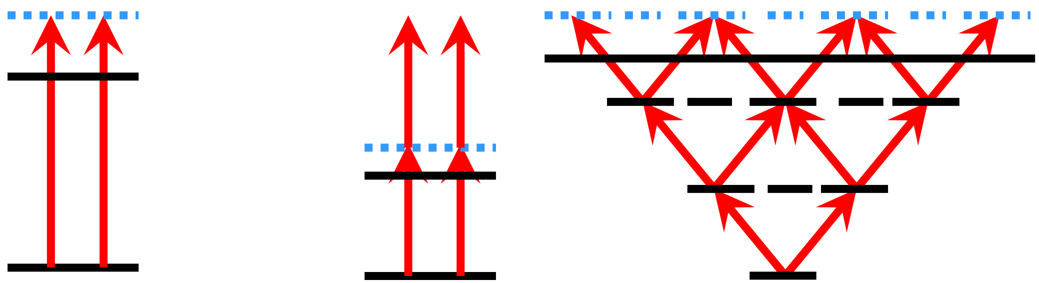
**vs. Heating and Avalanche**

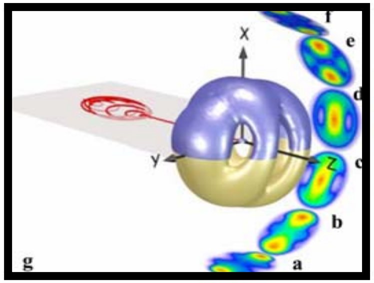
**(via temporally asymmetric pulse shapes)**



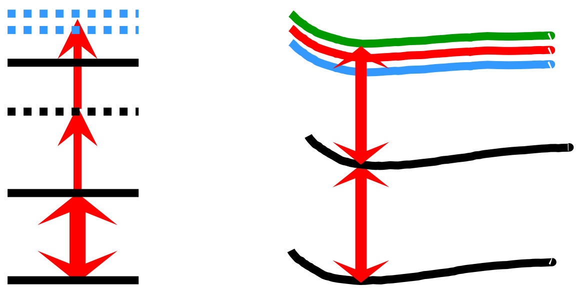
**Leads to Structures in fused silica**
**One Order of Magnitude Below Diffraction Limit**
**that are robust to variations in laser intensity**

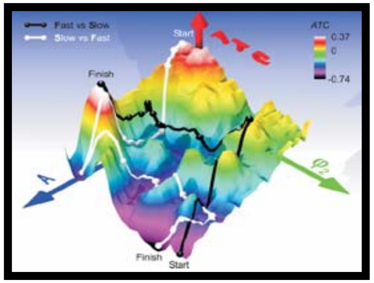
# I. Weak Field (perturbative) Coherent Control (K)



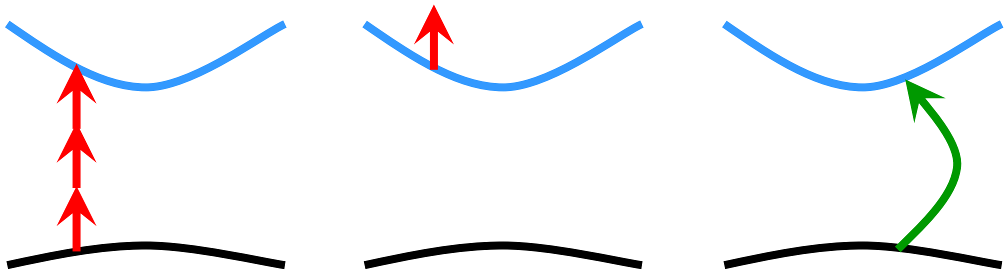


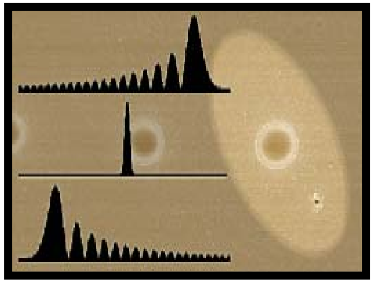
# II. Strong Field (non perturbative) Coherent Control (K, $K_2$, Na)





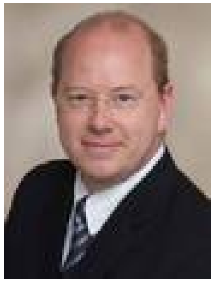
# III. Strong Field Incoherent Control (Dielectrics)





UNI KASSEL VERSITÄT

# THANKS to the group…



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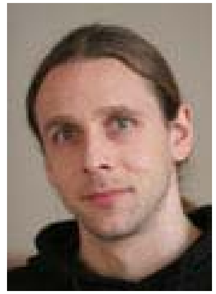

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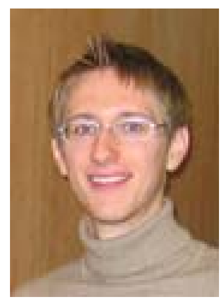

J. Schneider

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…and

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Forschungsgemeinschaft
DFG