ULTRAFAST LASER CONTROL Of IONIZATION

Fundamentals And Applications

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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


Textbooks

Brumer Shapiro (2003)

Rice / Zhao (2000)
Key Technology: Tailoring Light Via Spectral fs Pulse Shaping

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

Examples of Polarization Shaping*

2x640 pixel VPH Grating > 50% Throughput

TOD vs. GDD

*Brixner/Gerber 2001

Ultrafast Laser Control of Ionization

6. Über einen
die Erzeugung und Verwandlung des Lichtes
betreffenden heuristischen Gesichtspunkt;
von A. Einstein.
Ultrafast Laser Control of Ionization

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

1. wenn die Anzahl der gleichzeitig in Umwandlung be- griffenen 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, K₂, 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

- $\lambda = 800 \text{ nm}$,
- FWHM = 25 fs
- $|i> = K(5p) \text{ via } 405 \text{ nm}$
- laser preparation
Free Electron Interference: Experimental Setup

- Photoelectrons
- Time of flight [ns]
- Oven
- Skimmer
- Magnetic Bottle Spectrometer
- Laser Pulse Sequence
- Atomic Beam

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Free Electron Interference: Photoelectron Spectra

$\tau = 97$ fs
$\Delta t = 30$ fs
$E_{\text{res}} = 27$ meV

More than Spectral Interference: a Wigner Description

$|\Psi(p)|^2$

$W(x,p)$

$|\Psi(x)|^2$

$E_{kin}$

$e^-$

$I_P$

$i>$

$\tau$

time $= 30$ fs
Pulse Characterization on ATI (implementable in X-UV)

\[ E_{\text{kin}} [\text{eV}] \]

\[ \tau = 100 \text{ fs}, \lambda = 405 \text{ nm}, E = 0.25 \mu\text{J}, T = 4 \text{ ns} \]

\[ = 30 \text{ fs}, \lambda = 790 \text{ nm}, E = 1 \mu\text{J}, I < 10^{12} \text{ W/cm}^2 \]

\[ \tau = 4 \pm 2 \text{ fs} \]

\[ M^2 = 326 \text{ fs}^{-2} \]

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

$\varphi_\nu(\omega) = \tau \cdot |\omega - \omega_\nu|$ $280 \text{ nJ}$

$\varphi_\nu(\omega) = \tau \cdot |\omega - \omega_\nu| + \tau \cdot |\omega - \omega_\nu|$ $8 \mu\text{J}$
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, K₂, 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

\[ \Delta E = \hbar \Omega(t) = \mu E(t) \]

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

Selective Population Of Dressed States (SPODS)
Strong Field Control via SPODS Requires Temporal Phase Variations*

\[ \varphi(\omega) = \varphi_2(\omega - \omega_0)^2 \]

**Continuous**

**Discontinuous**

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

Rapid Adiabatic Passage

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)

\[ A = 0.2; \quad T = 170 \text{ fs}; \quad E = 0.5 \mu\text{J} \]

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Ultrafast Control of Coherent Electronic Excitation in Molecules: $K_2$
SPODS On $K_2$ Molecules: Experimental Setup

Ti:Sapphire
790 nm
30 fs

LC-SLM
$\varphi(\omega)$

E_{in}(t)

oscilloscope

Phoélectrons

5'$\Sigma_g^+$

1100
1150

TOF [ns]

Nd:YAG
532 nm
6 ns

Telescope

ns probe pulse

molecular beam

Energy [eV]

$5'\Sigma_g$

$2'\Pi_g$

$4'\Sigma_g$

$A'\Sigma_g^+$

$X'\Sigma_g^+$

R [Å]

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

Robust Photon Locking via Generalized $\theta$-Step (K)

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

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

- 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, K₂, 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

Nanostructures


fs-LIBS


Plasma dynamics


spatial

NA = 0.5; 1/e² diameter: 1.4 µm; 1/e² length: 9.1 µm
Basic Ionization Processes in Dielectrics to Reach Critical Electron Energy / Density for Ablation

Multiphoton Ionization

Free Carrier Absorption - Heating -

Impact Ionization

NEEDS INTENSITY

NEEDS SEED ELECTRONS & TIME
Control of Basic Ionization Processes via Temporally Asymmetric Femtosecond Pulses

\[ \phi (\omega) = \frac{\phi_3}{3t} \cdot (\omega - \omega_0)^3 \]

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, \( \phi_3 = 600\,000\,\text{fs}^3 \)

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Reduction In Structure Size Via Pulseshaping

SAME FOCUS CONDITIONS
SAME FLUENCE
SAME SPECTRUM

order of magnitude below diffraction limit!
Structure Size as Function of Fluence

Nanostructure size robust to variations in laser fluence

Simulation of Basic Ionization Processes

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, K², Na)

III. Strong Field Incoherent Control (Dielectrics)

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