Attosecond multielectron dynamics in tunnel ionization

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Theory

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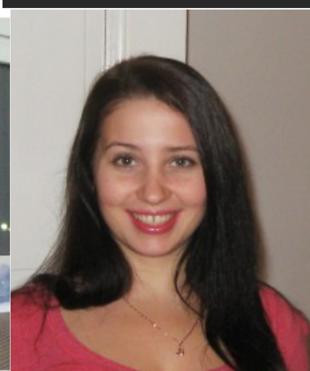


Valeria Serbinenko MBI Berlin

+ Misha Ivanov, MBI & Imperial College

Experiment

Yann Mairesse , CELIA, Bordeaux

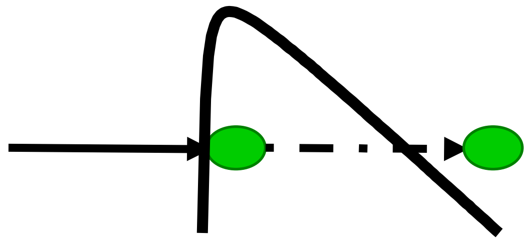
Nirit Dudovich , Weizmann Institute,

Dror Shafir , Weizmann Institute,

Hadas Soifer, Weizmann Institute,

MBI-Theory

Time-resolving tunneling dynamics



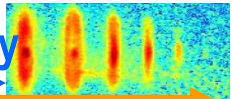
Time delays in tunneling: old and controversial
(recent experiments: U. Keller group, Science, 322, 1525, 2008)

Tunneling dynamics can be affected/probed via coupling to other degrees of freedom – how do they respond?

Examples:

Josephson junction:	energy loss during tunneling due to friction
Metal-insulator tunneling:	build-up of image charge

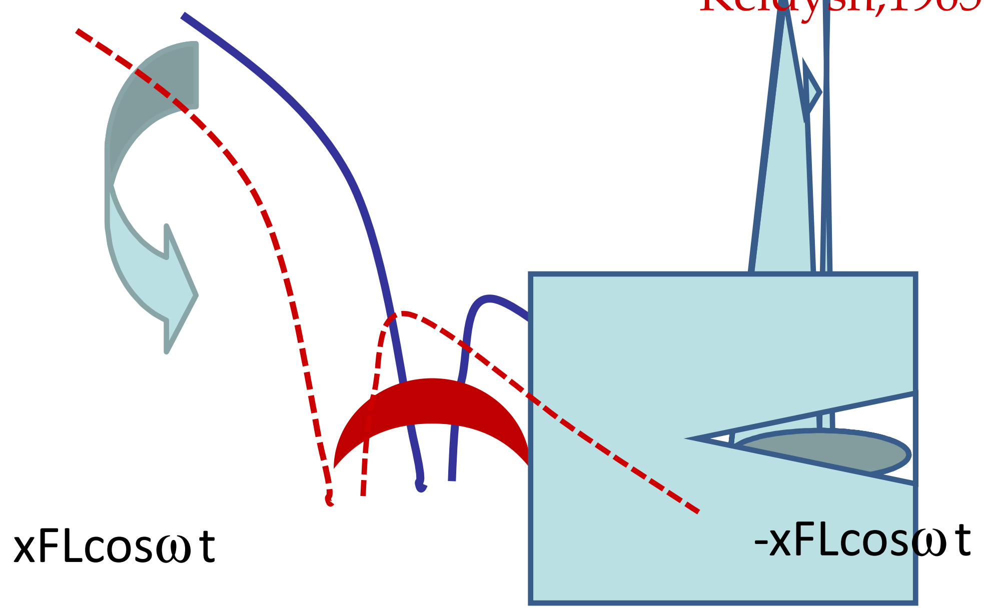
Strong-field ionization of molecules: electron-hole interaction in tunneling



Strong-field ionization in IR fields

ω/Ip << 1 – adiabatic tunnelling perspective on ionization.

Keldysh,1965

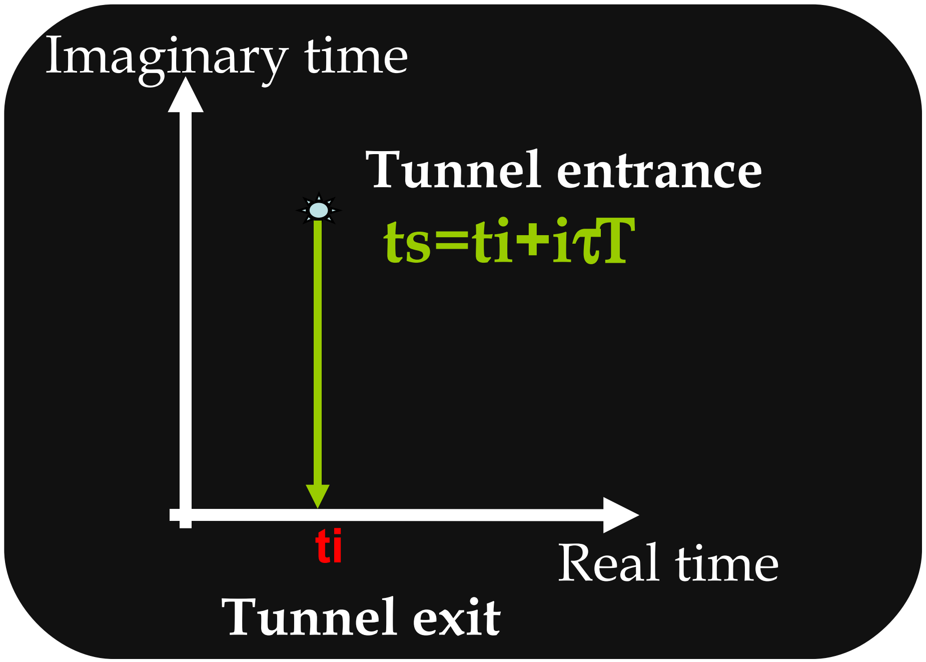


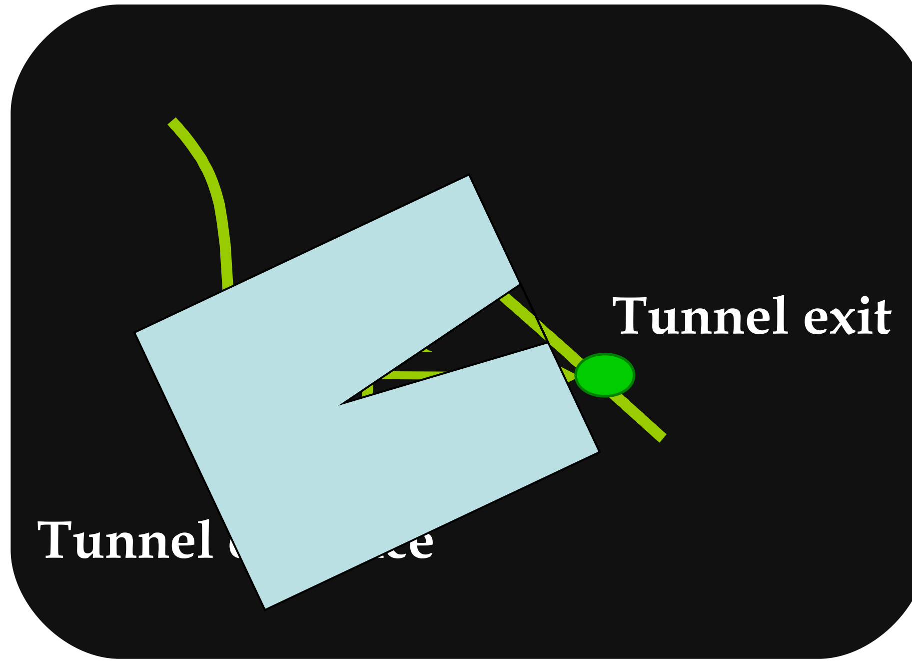
aT~exp[–(2Ip)3/2/3FL]

Optical tunneling in the oscillating laser field is the main ionization mechanism for ω<<Ip

Semiclassical perspective on tunneling

Electron trajectory during tunneling: imaginary velocity & imaginary time





ts -time of entering the barrier, ti - time of exiting the barrier;

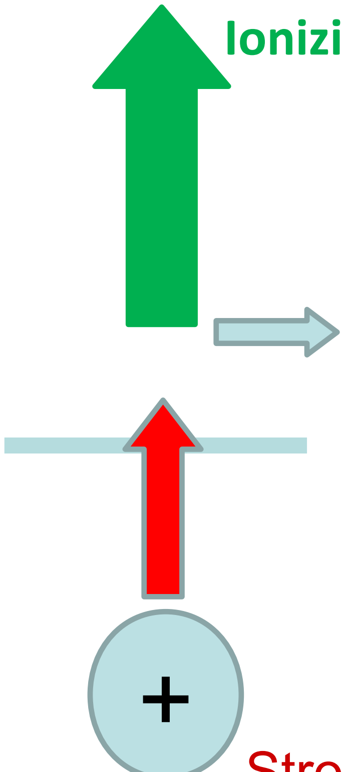
τT – time spent under barrier: imaginary

Does this theory fit experiment? Need to:

- Tag each electron trajectory as it exits the barrier at ti
- Check if distribution of ti fits the theory picture above

Tagging trajectories as they are launched

Nirit Dudovich ,
Weizmann Institute,



Ionizing strong Fω cosω t

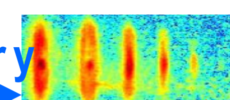
Weak probe
F2ω cos(2ω t+ϕ)

$$v\ (t,t_i) \qquad {}_{t_i}^{t_r} F_2\ (t')dt' \qquad A_2\ (t_r) \qquad A_2\ (t_i)$$

$$r\ (t,t_i) \qquad {}_{t_i}^{t_r} v\ (t',t_i)dt'$$

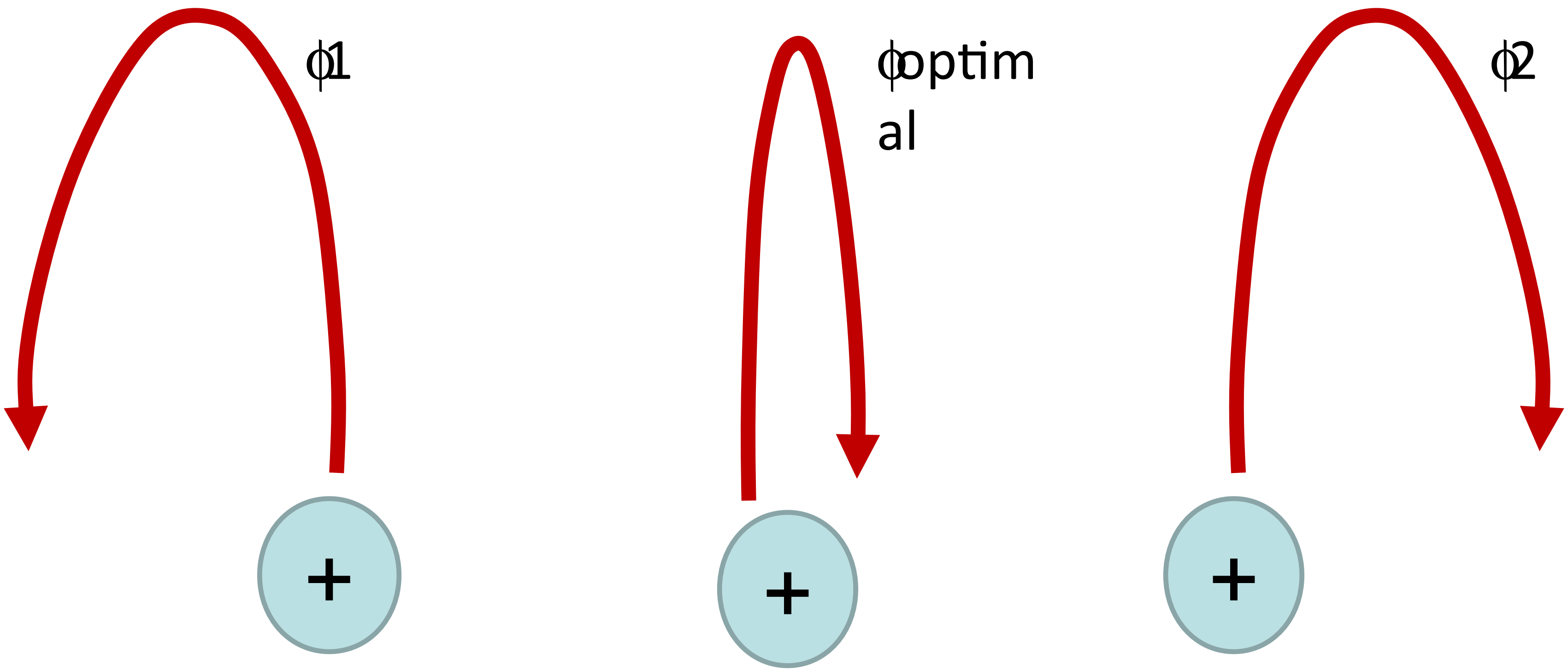
Strong ω field drives tunnelling
Weak 2ω tags the electron
Dependence of Δv and Δr on ϕ encodes ionization times

Δv and Δr are recorded in harmonic spectra



Measuring position shifts: High harmonic signal

Oscillating field brings the electron back – harmonic emission
Parent ion is a perfect measurement device

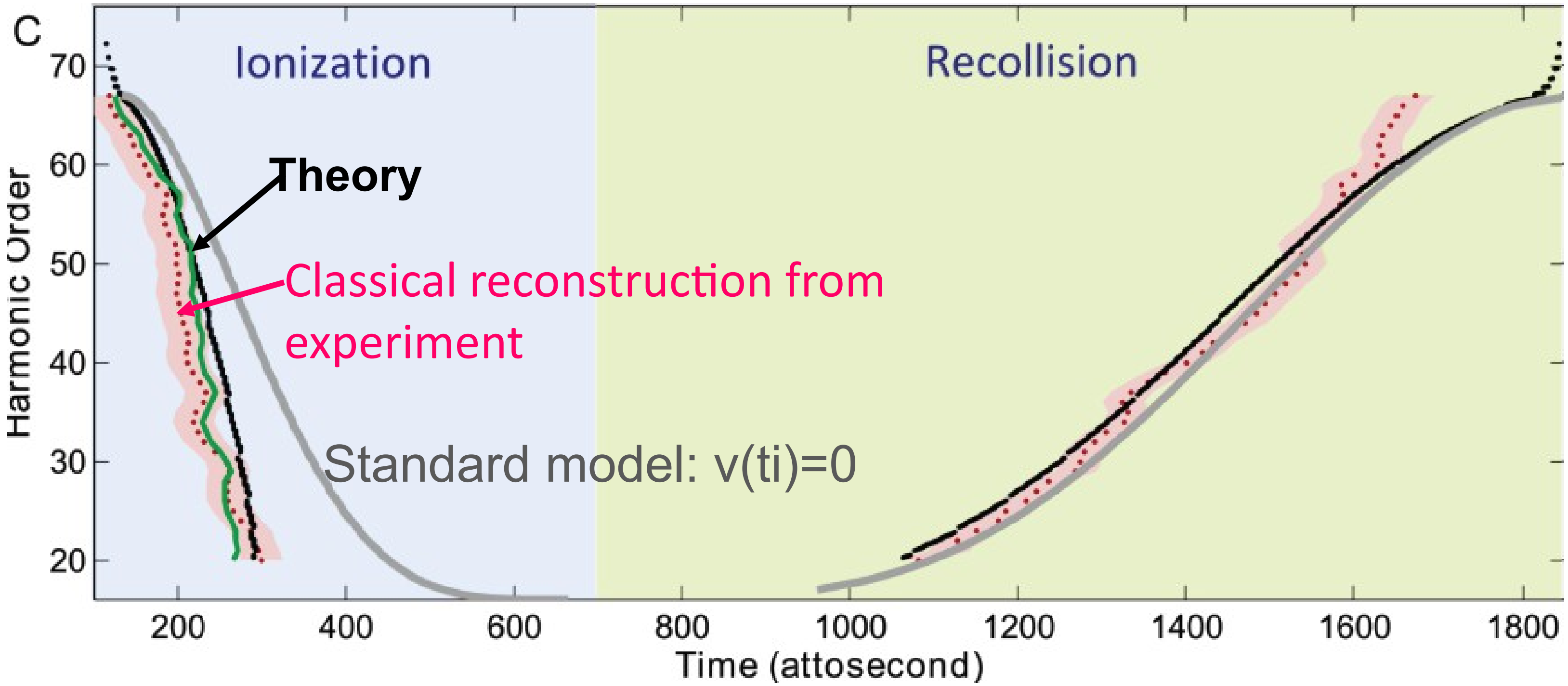


Odd harmonic intensities maximize for minimal Δr (φ)
Odd harmonics measure Δr (φ)

Reconstructed times in Helium

Helium, 40 fsec pulse, 800 nm at 4 1014 W/cm2, 400 nm at 1-2% intensity level

D. Shafir et al, Nature 2012

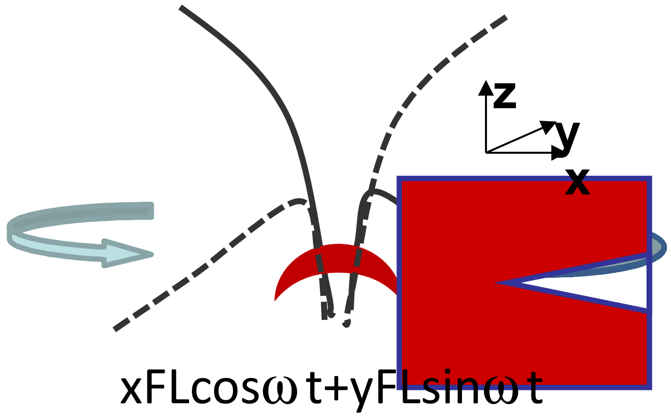


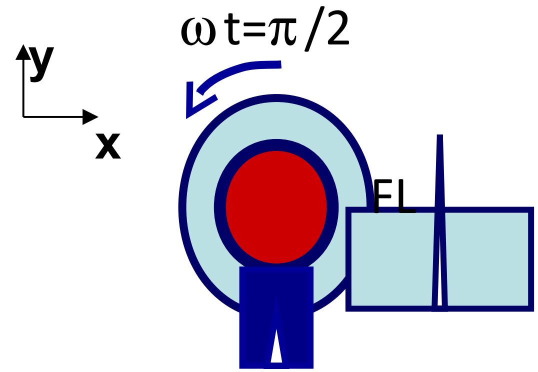
Green: Full quantum, including effect of 2ω on ionization

Results are consistent with the theory: purely imaginary ‘delay’

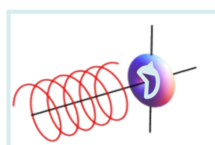
Are there any kinematic consequences of “dynamics in imaginary time”?

Tunneling in circularly-polarized IR fields

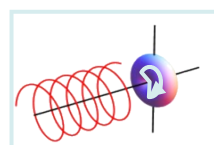




***p*+ orbital**



***p*–orbital**

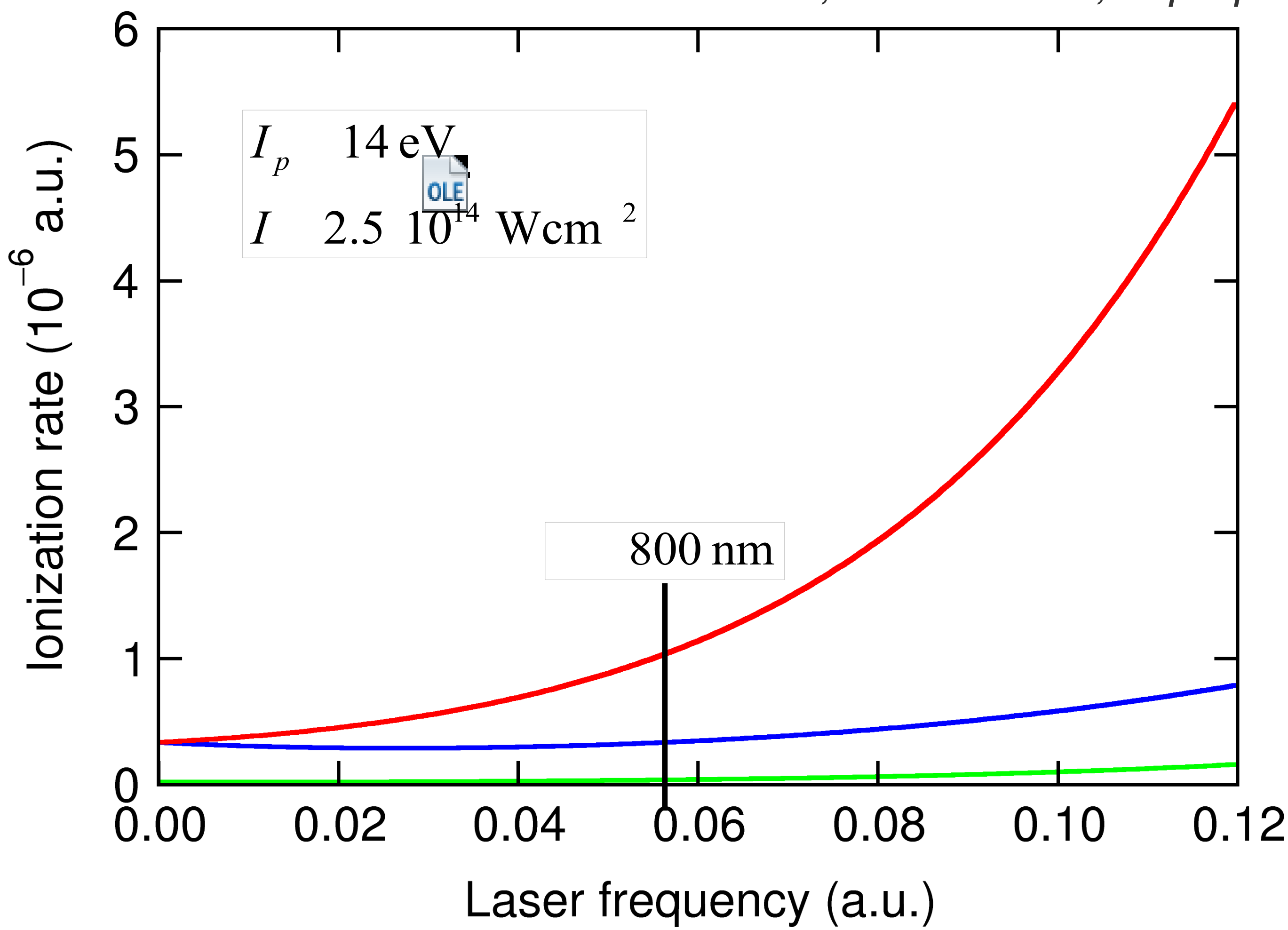


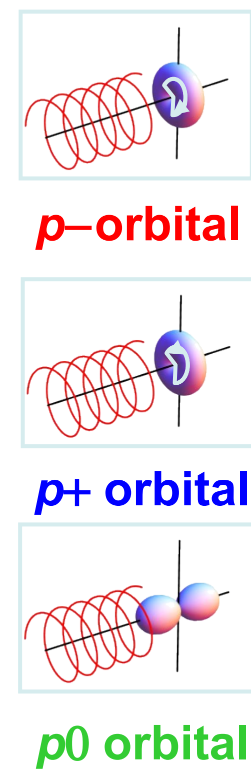
No real delays: laser field does not rotate during tunneling
Co-rotating and counter-rotating electron would tunnel with the same rate

Real delays: laser field rotates during tunneling
Co-rotating electron tunnels easier

Result for Kr: Counter-rotating electron tunnels easier!

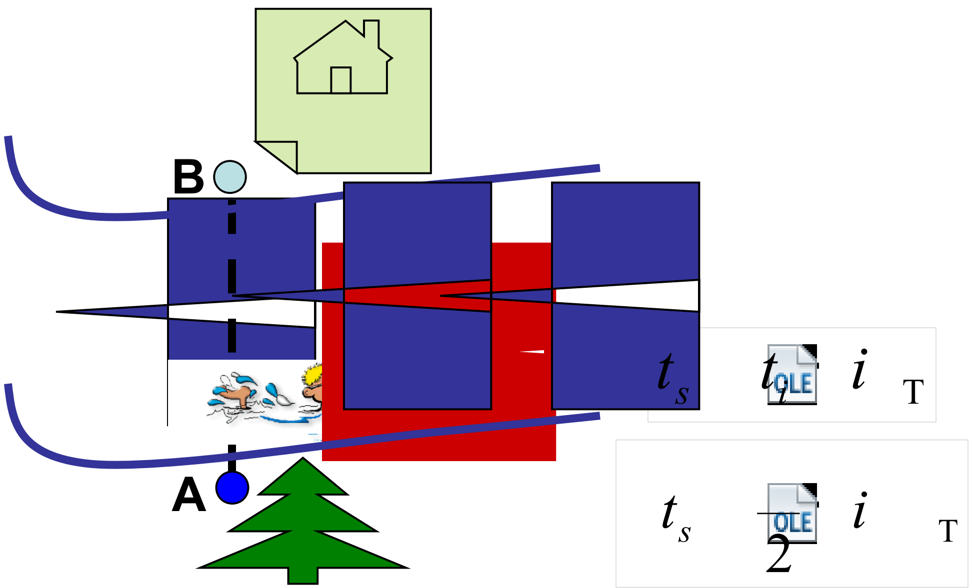
*I. Barth, O. Smirnova, Phys. Rev. A **84**, 063415 (2011)*
J. Kaushal, O. Smirnova, in preparation

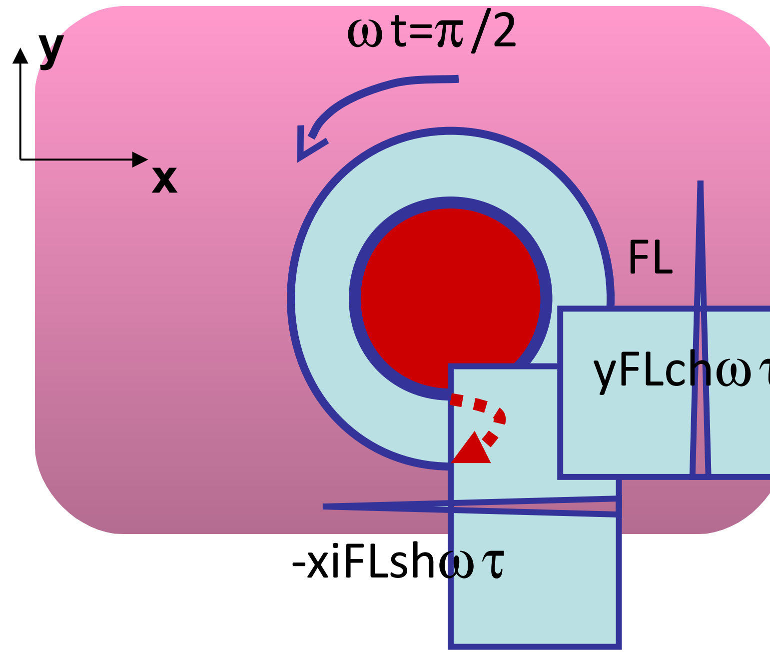




Why does the counter-rotating electron tunnel easier?

Initial electron velocity prior to ionization is opposite to the rotation of the field





Laser field=-xiFLshωτ+yFLchωτ

Ion is created in polarized state- probe by transient absorption
Tunneling dynamics is recorded in the response of the ion

Hole dynamics in space and time

Time:

- When is the hole created?
  Is there a delay in hole formation?
- Does the hole move or remains static?

Space:

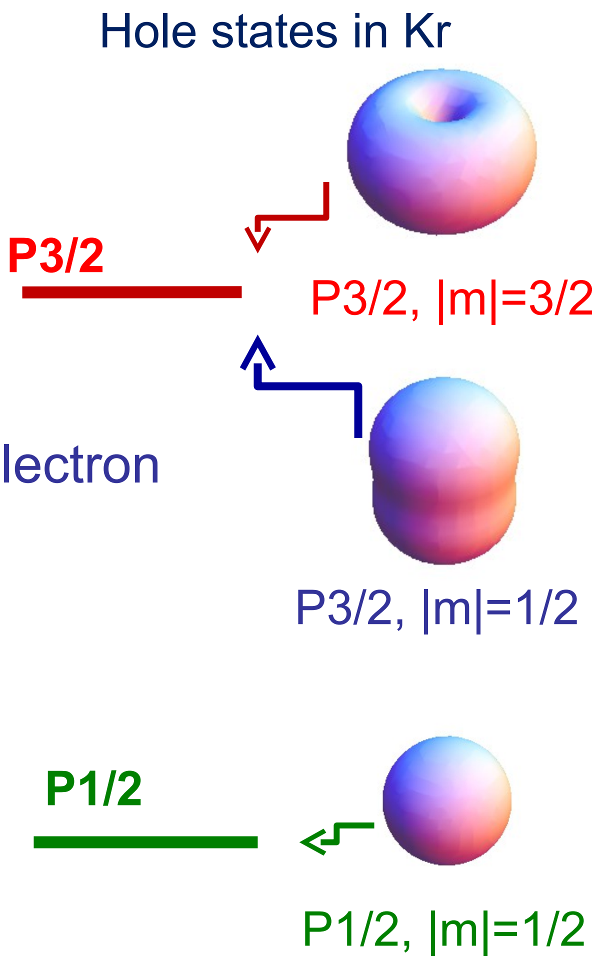
- Is the hole aligned along the direction of electron emission?

Initial kick:

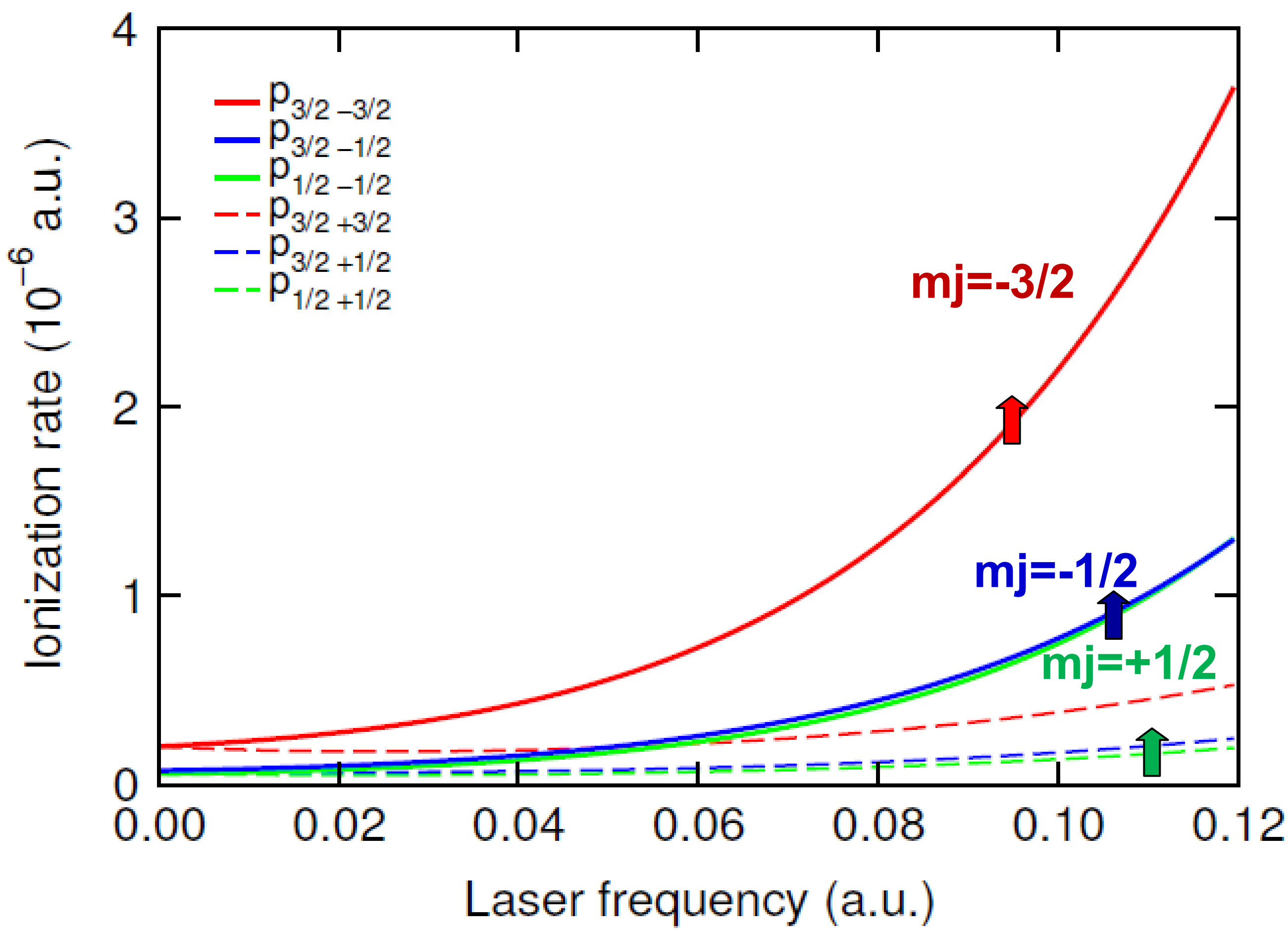
- Is there “electron-hole” recoil?

Need to find:

- Initial populations of hole states
- Relative phases between them
- Must take spin into account

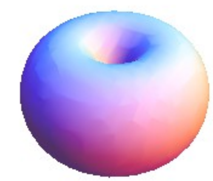


Populations of hole states in Kr



P1/2, |m|=1/2

P3/2, |m|=1/2

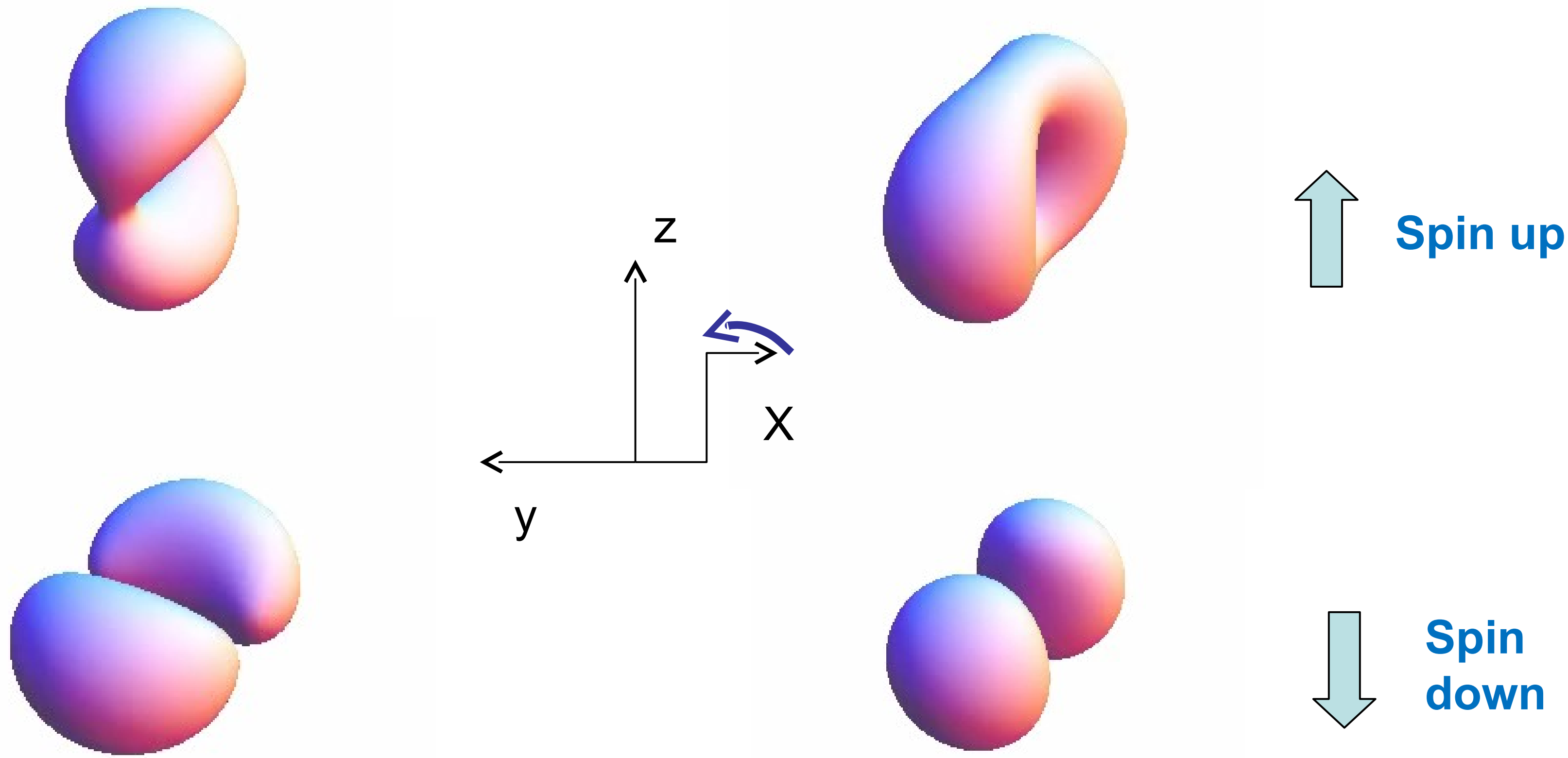


P3/2, |m|=3/2

More spin-down states then spin-up, for right circular field: consequence of higher ionization rate for m=-1

Hole dynamics in Kr: 800nm 2.5 1014 W/cm2

~Up electron- Rotating hole | **2Up+Ip electron- Swinging hole**

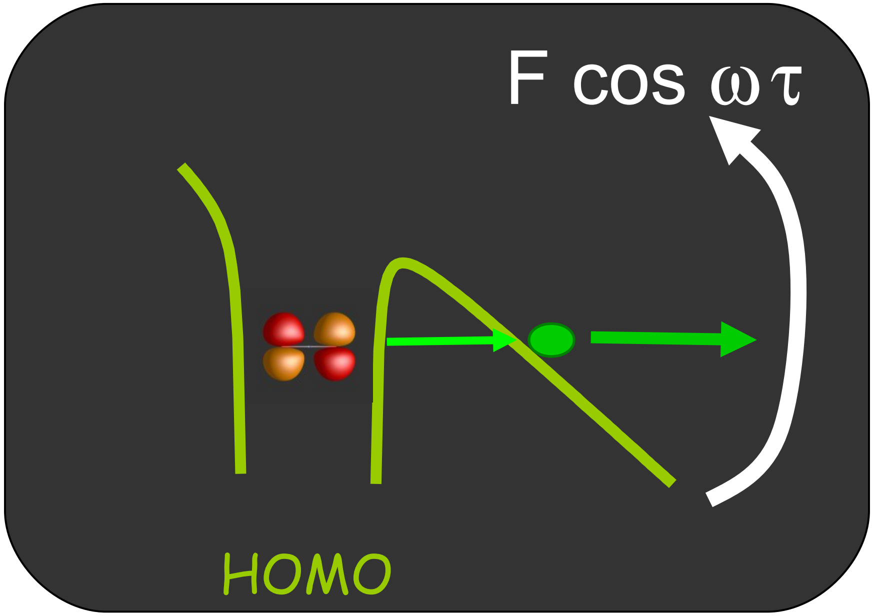


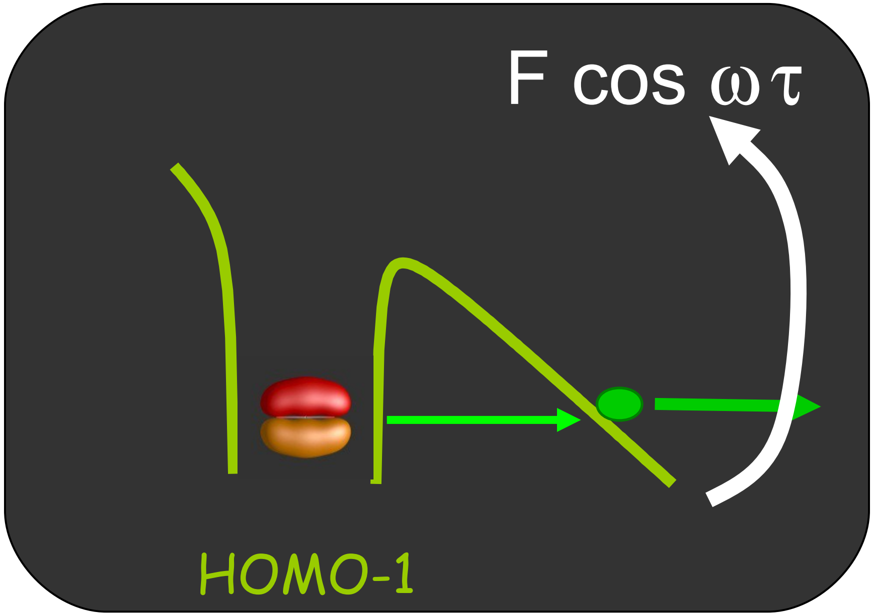
Theory shows:

- Electron-hole entanglement plays crucial role
- Hole is aligned along the instantaneous direction of electron emission
- No delays (in multielectron case with no channel interaction)

Tunneling for coupled channels

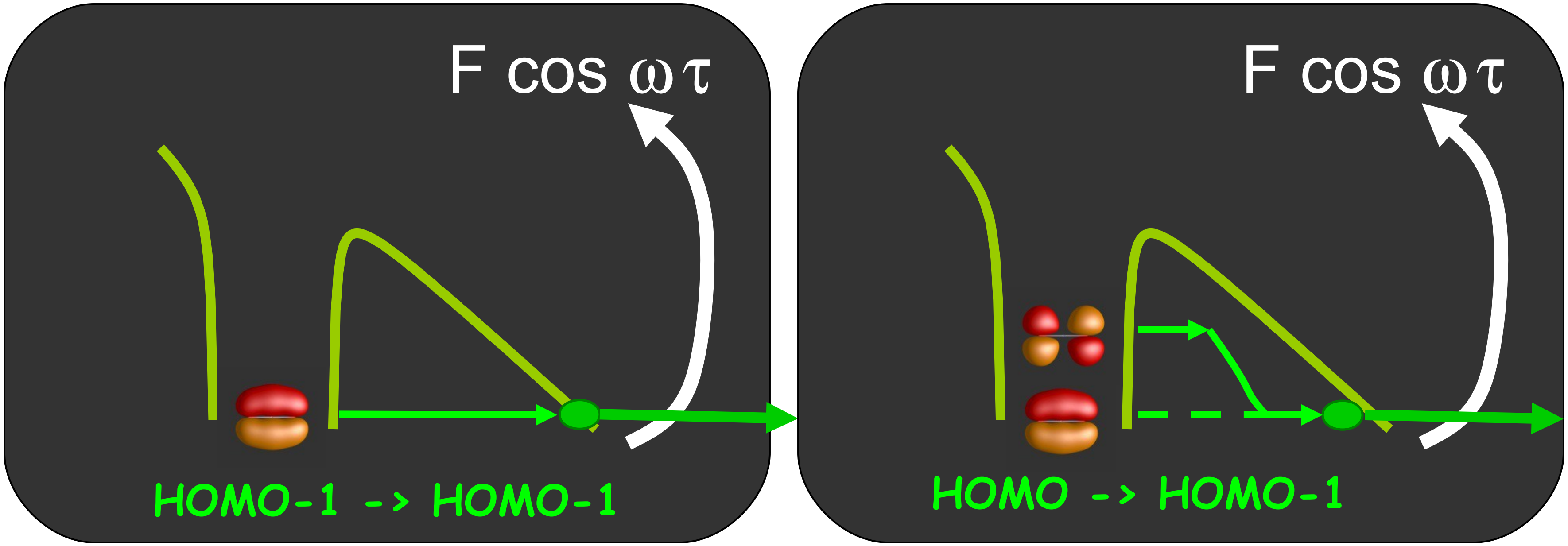
Tunneling from lower orbitals: usually expect exponential suppression for deeper orbitals





This talk: Role of electron correlation during tunneling – overcoming exponential suppression

Idea: new tunneling channels



Direct channels

Cross channel

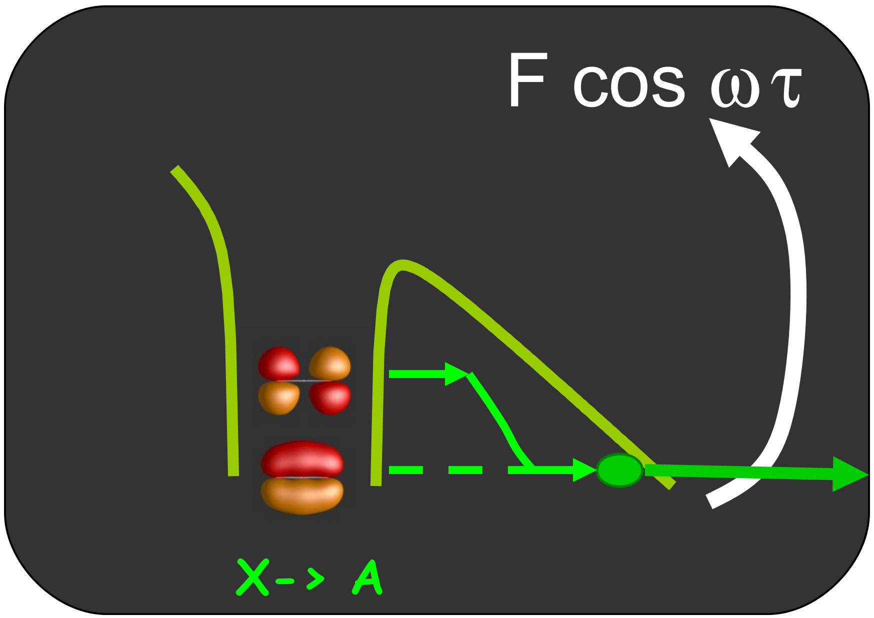
- Tunneling electron excites the core while moving through the barrier
- This channel is not subject to the full exponential suppression characteristic of the direct tunnel ionization from deeper orbitals.

Z. Walters, O. Smirnova J. Phys B 2010

L. Torlina, M. Ivanov, Z. Walters, O. Smirnova PRA, in press

Coupled electron- ion dynamics: the ion

Ion: few-level system driven by the laser field and correlation "pulse" (in complex time)



aX->A(p) =cXA aX(p)

$$c_{mn} \qquad {}^{0} \qquad dt\, V_{corr}^{mn}(t)\, e^{I_p t}$$

t is the time left before exiting the tunnel.

Correlation –induced excitation is an integral of Vcorr along the trajectory of the tunneling electron.

- Tunneling occurs in imaginary time
- Excitation amplitudes in the ion are the imprints of this dynamics

Comparison with numerical simulations

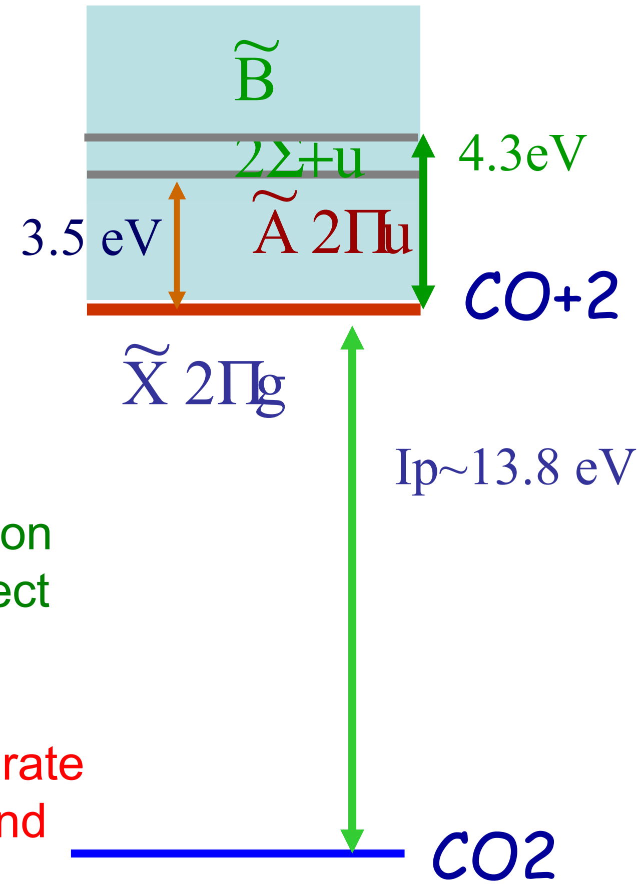
Serguei Patchkovskii, NRC, Ottawa

Michael Spanner, NRC, Ottawa

M. Spanner & S. Patchkovskii

Ab-initio close – coupling approach (**Phys Rev A, 2010)**

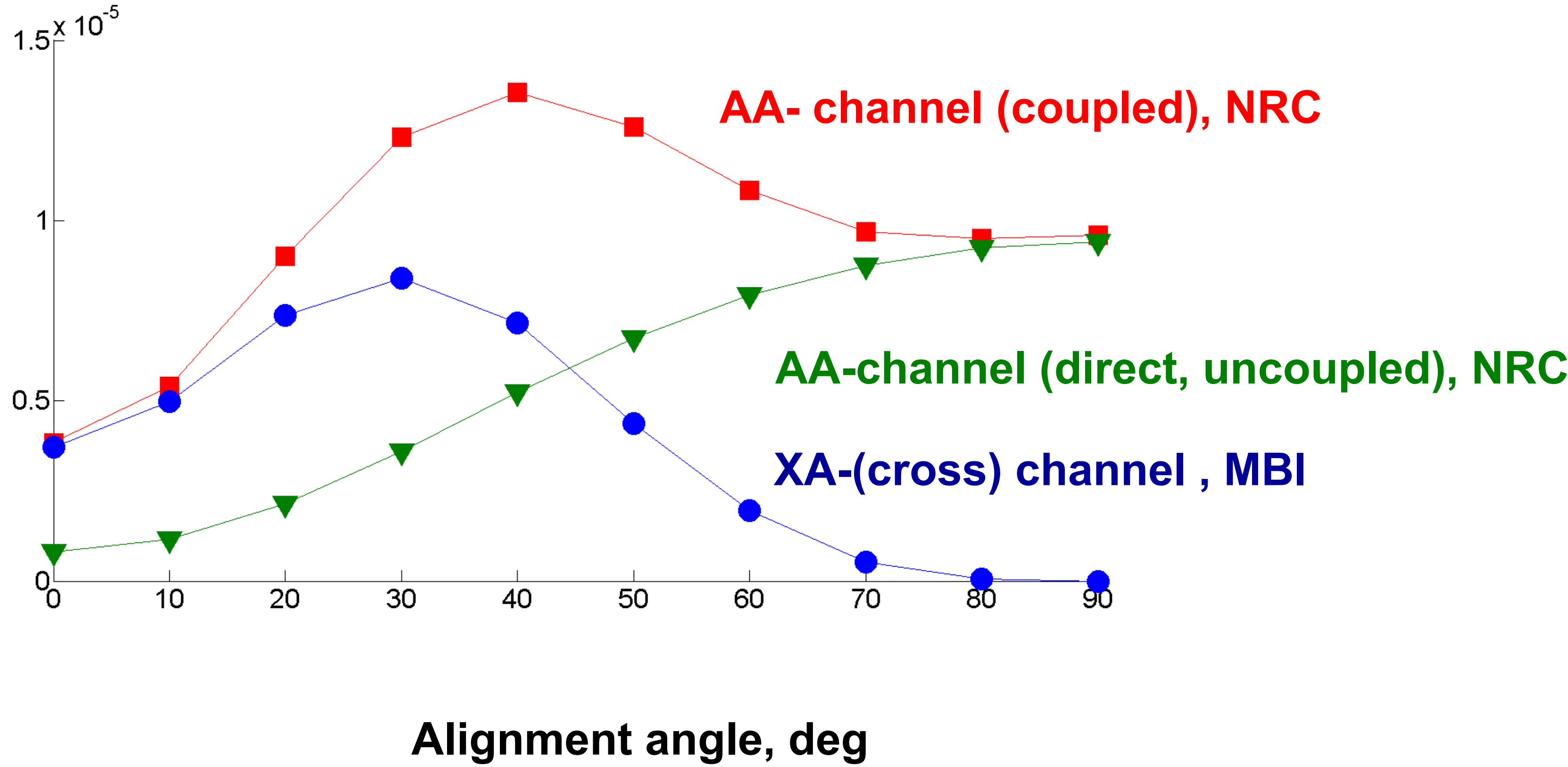
CO2 I=1.3 1014 W/cm2 ,
800 nm, half cycle pulse

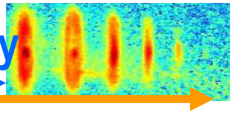


The NRC team made two simulations:

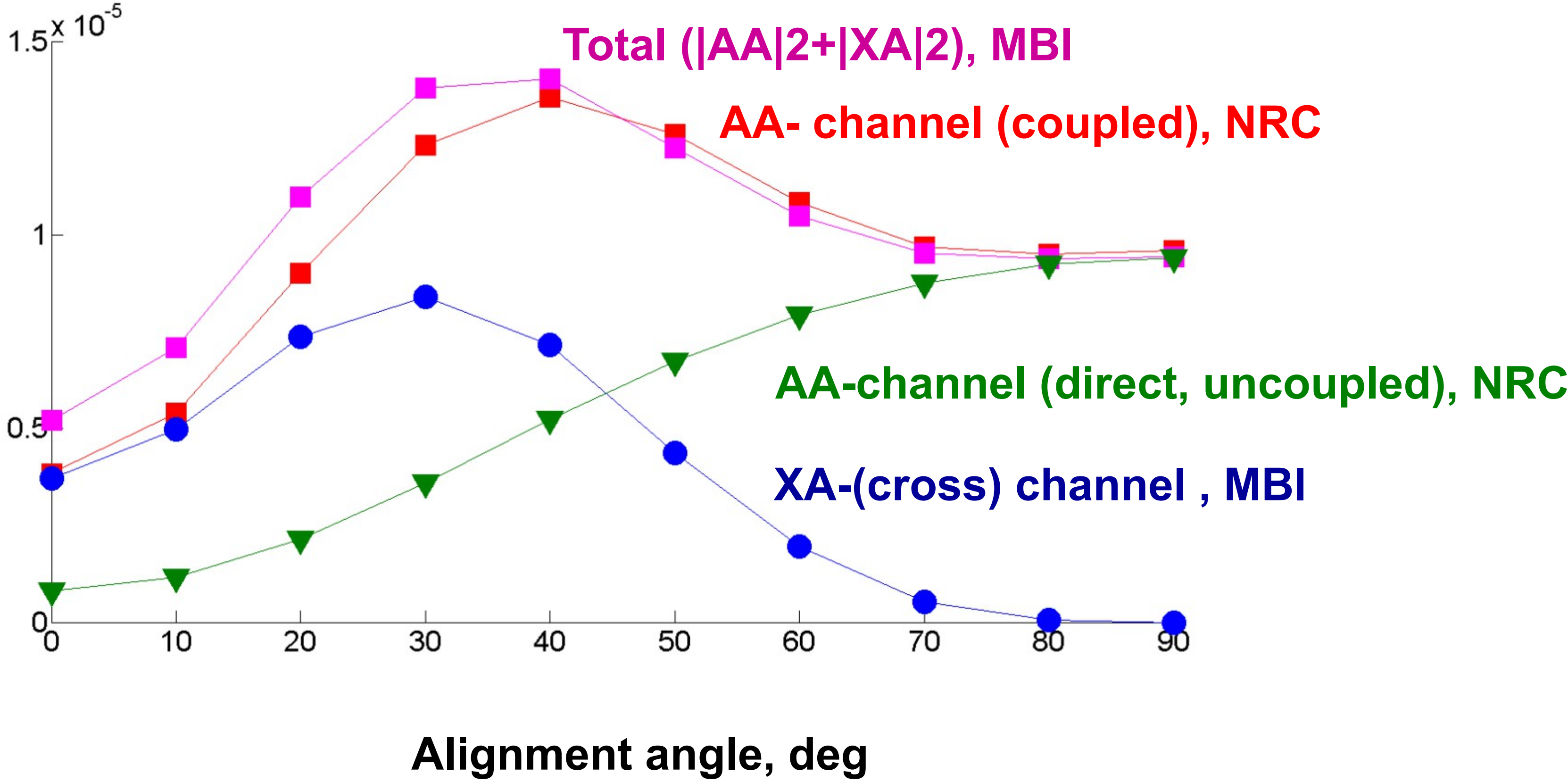
- Uncoupled channels: ionization rate in channel A includes direct channel only
- Coupled channels: ionization rate in channel A includes direct and cross-channels

Comparison numerical (NRC) vs analytical (MBI)



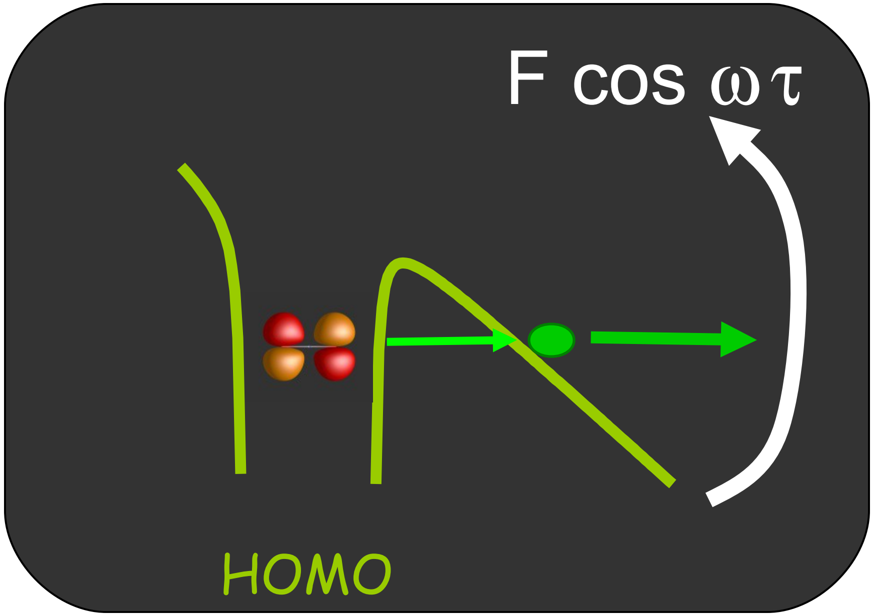


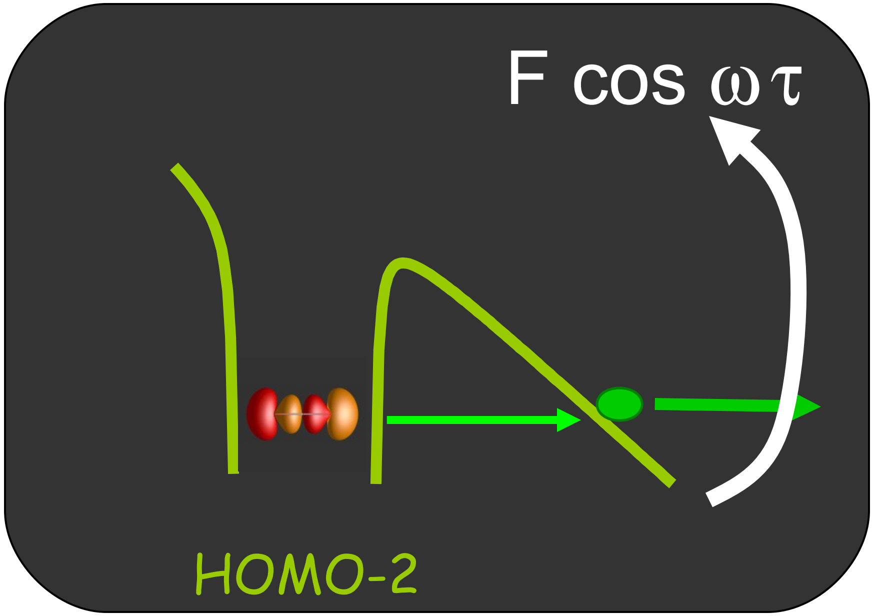
Comparison numerical (NRC) vs analytical (MBI)



Direct and cross channels should interfere: phase is close to $\pi/2$

Time-resolving hole dynamic

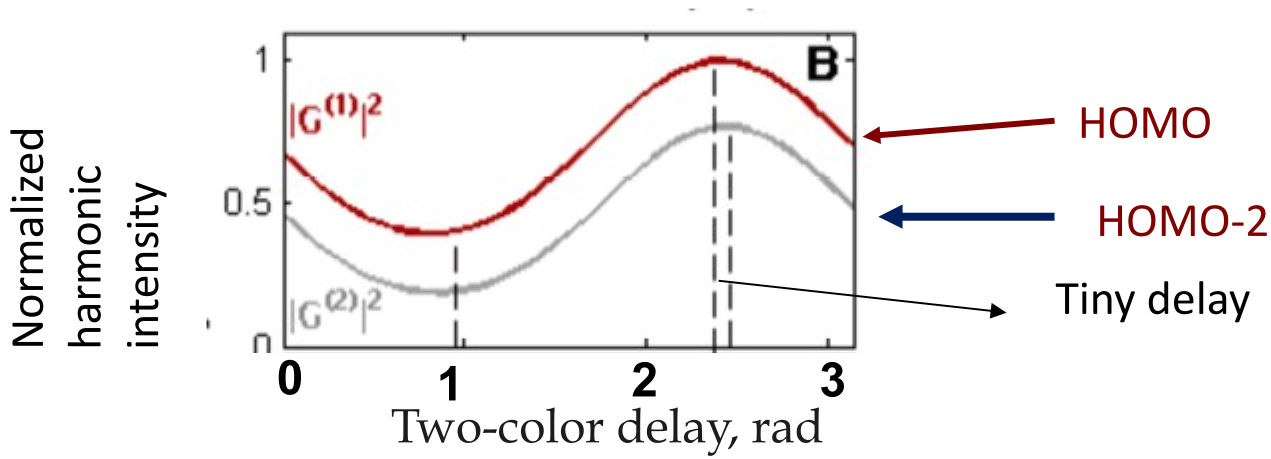




Can we detect tunneling from two orbitals?

How to detect ionization from lower orbital?

Modulation of harmonic signal vs two-color delay



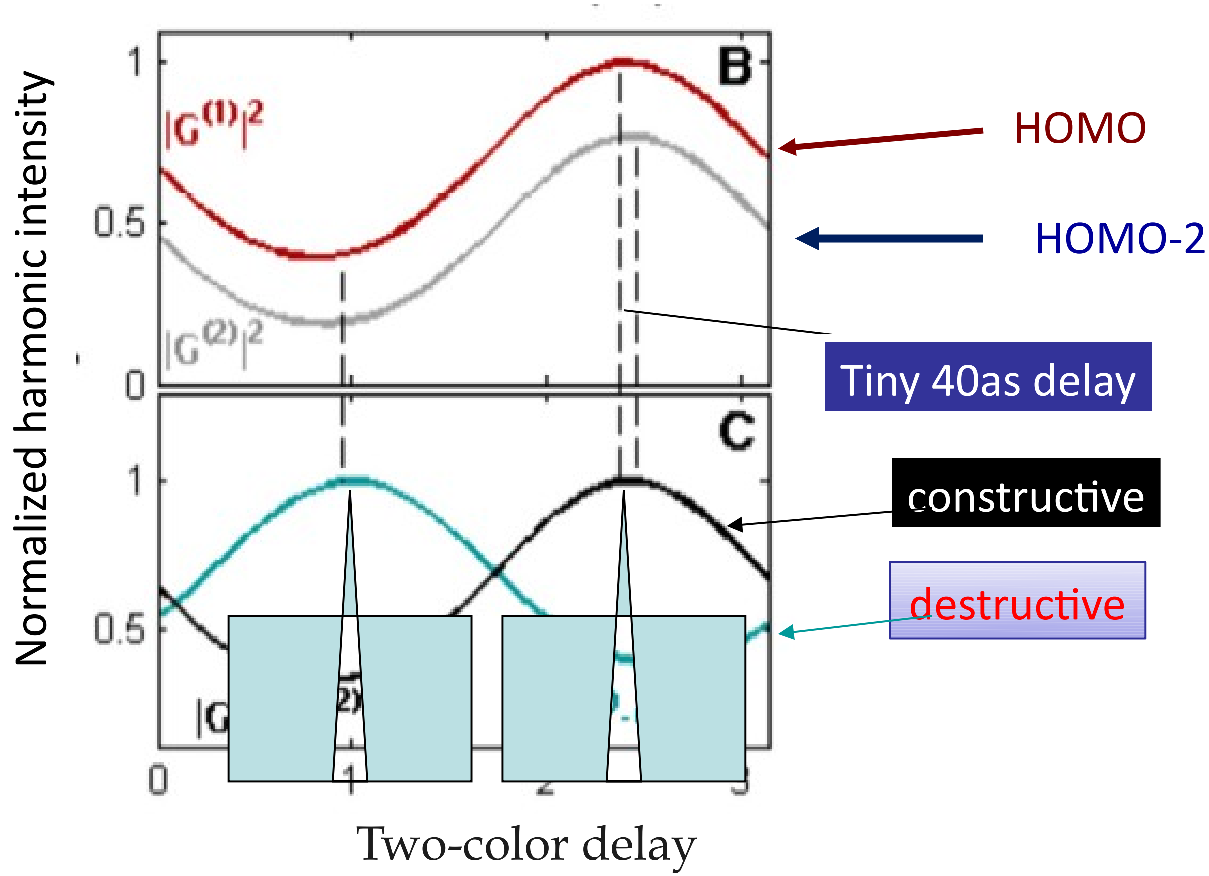
Strong Fω cosω t

Due to different ionization times, modulation for Homo and Homo-2 channels is slightly shifted

The shift is tiny. Can it be seen?

Weak probe
F2ω cos(2ω t+ϕ)

Detecting signal from two orbitals

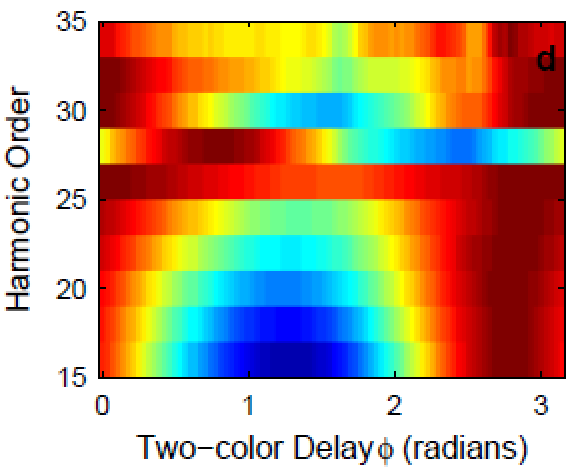


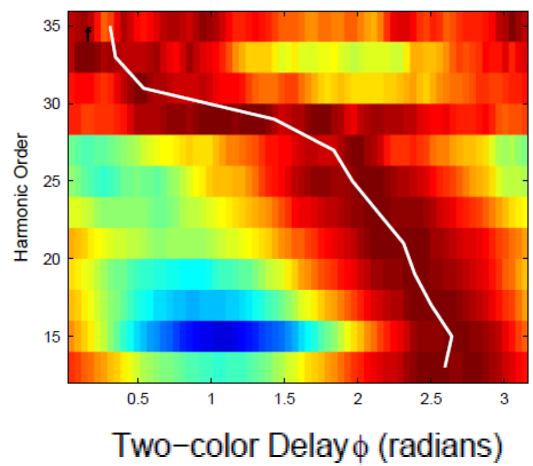
- Due to different ionization times, interference shifts optimal delay
- The shift is largest near destructive interference between the channels

Results

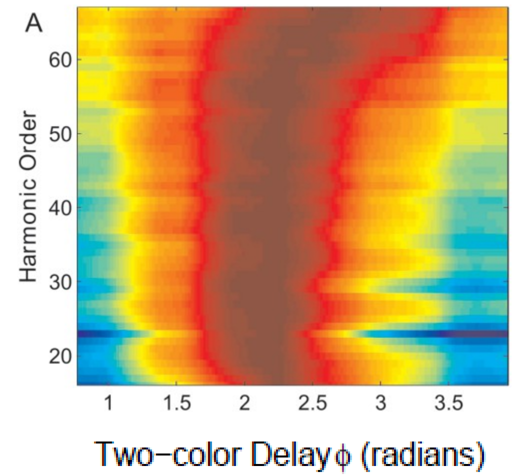
Normalized HHG signal
1.41014 W/cm2 800nm, 2% of orthogonally polarized 400nm

CO2, aligned at 0 deg





He atom



Theory **Experiment**

$\pi/2$ phase shift marks the positions of dynamical minima.

Position of the phase jump identifies the initial phase between the hole states

D. Shafir et al, Nature 2012

MBI-Theory

Conclusions

1. Tunneling delays:

- No delays in one-electron systems
- No delays in multielectron systems without coupling between different hole states
- Delays might be possible due to electron-hole interaction during tunneling

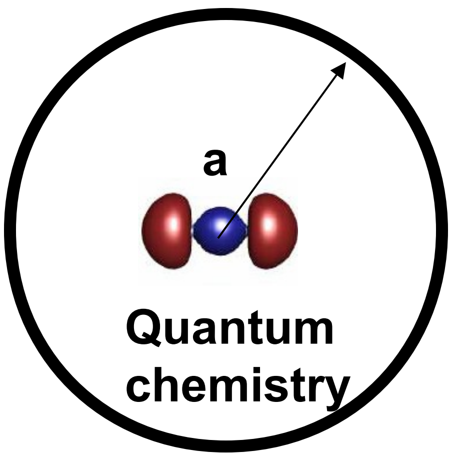
2. Electron-hole interaction during tunneling can lead to non-trivial tunneling dynamics

3. Using multidimensional high harmonic spectroscopy to resolve these dynamics is our next goal

Our Method

Analytical time-dependent R-Matrix (ARM):

- R-matrix-type partitioning of configuration space
- The eikonal-Volkov approach in the outer region
- Quantum chemistry in the inner region (for molecules)
- Matching the two using the Bloch operator approach
- The saddle point method



WKB

(Eikonal-Volkov states
Smirnova et al PRA, 2008)

Results: ARMed with ARM

One electron:

Analytical ionization amplitude for arbitrary potentials.

Benchmark:

- Same results as using the PPT method
  - for Hydrogen
  - for short range potentials
  - for linearly and circularly polarized fields

L. Torlina, O. Smirnova PRA, in press
J. Kaushal, O. Smirnova, in preparation

Advantages:

Arbitrary long-range potentials

Many electrons: Analytical ionization amplitudes including electron- electron correlation and core rearrangement during ionization

Gauge invariant, Technically much simpler than PPT

L. Torlina, M. Ivanov, Z. Walters, O. Smirnova, in press