HH Spectroscopy of Polyatomic Molecules:

What do we learn that we do not know?

Theory

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Experiment

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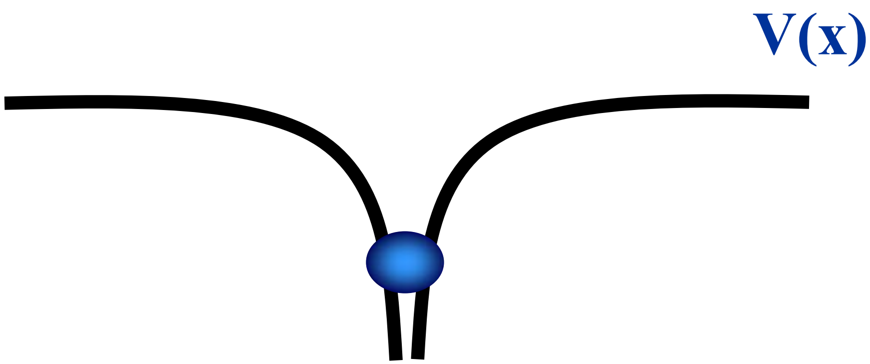
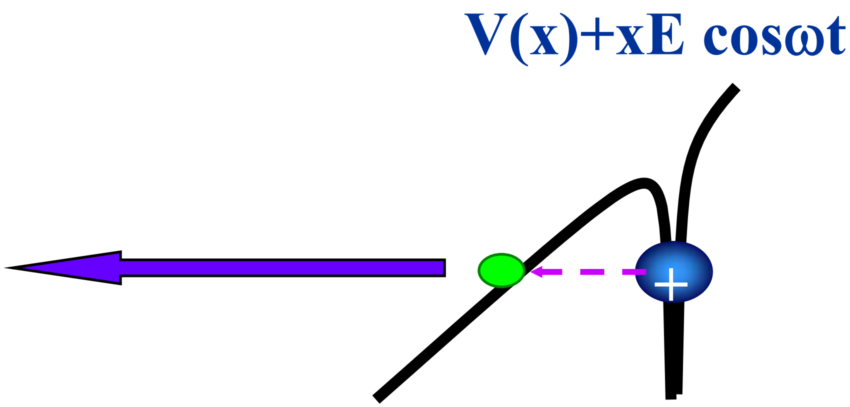
Strong Field Ionization: Optical Tunneling

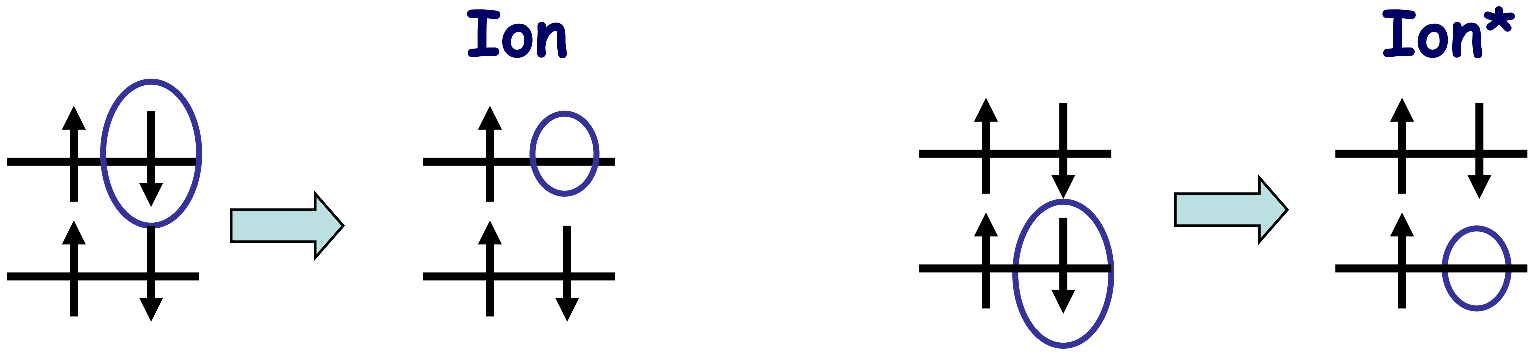
Field free
atom/molecule



Tunnel ionization in
strong laser fields



Many electrons and multiple orbitals



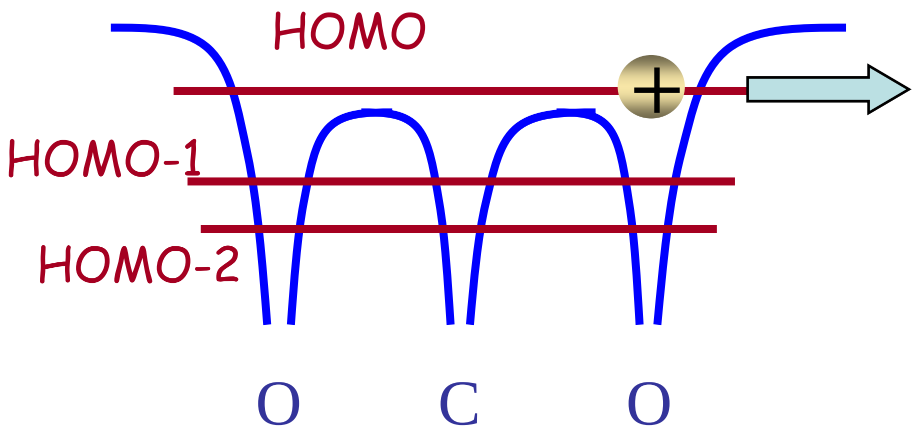
- We can remove an electron from different orbitals
- Ion can be left in excited states
- Multiple ionization channels are important in strong field ionization in molecules

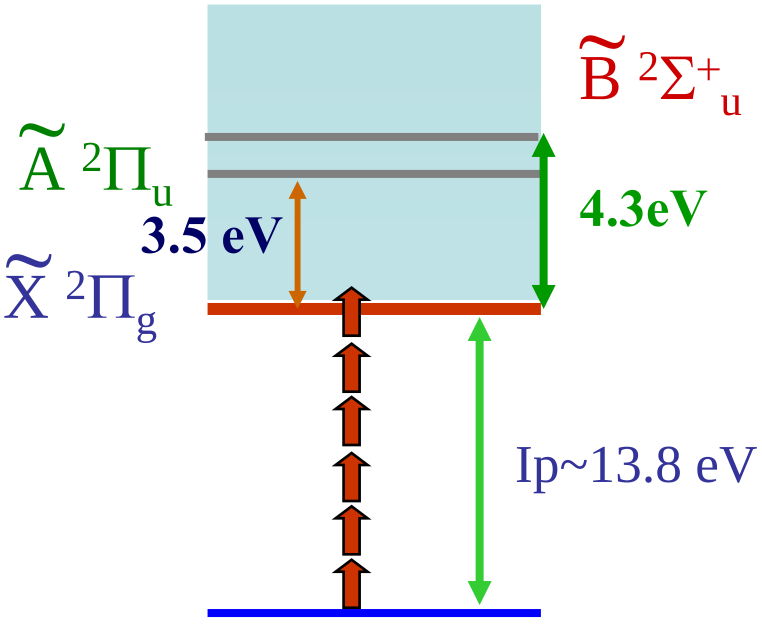
Key reasons: Proximity of electronic states in the ion

Geometry of Dyson orbitals

Consider CO_2

Ionization from molecules





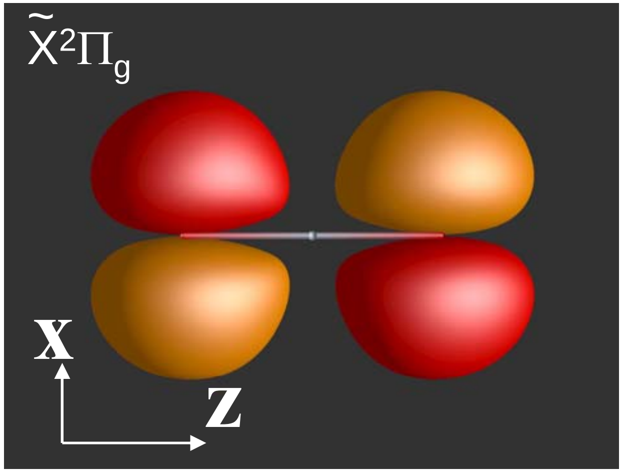
Tunnel ionization is exponentially sensitive to Ip

$$W \sim \exp\left[-\frac{2(2I_p)^{3/2}}{3E}\right]$$

There is more to ionization than I_p

Ionization from different orbitals: leaving the ion in excited states

Interference suppression of ionization: A. Becker et al, 2000, C.D. Lin et al 2002



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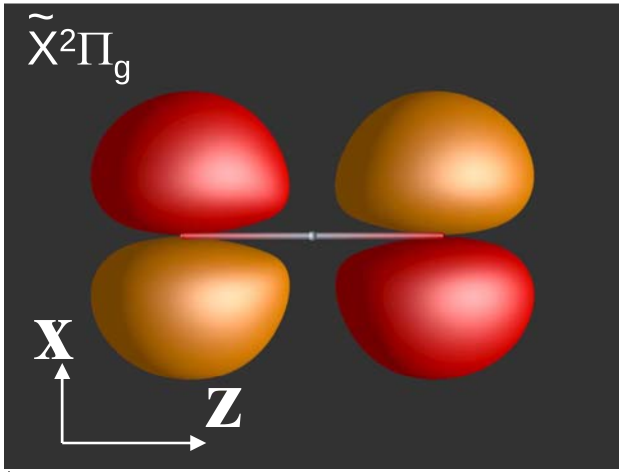

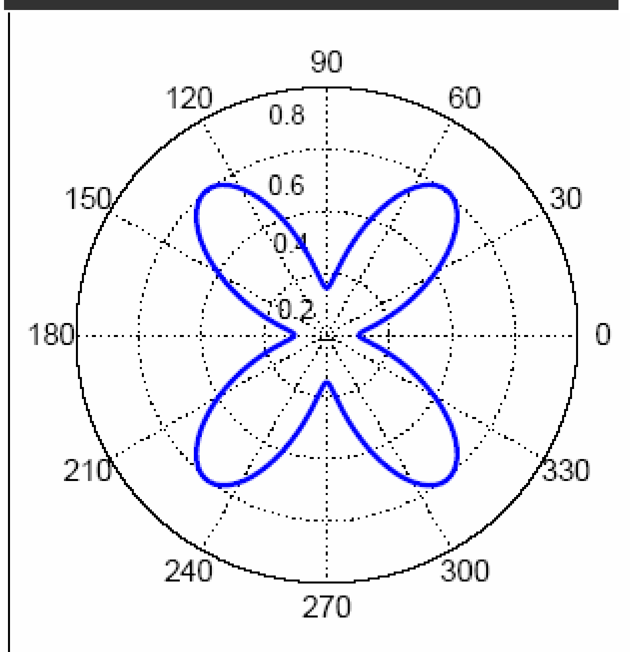

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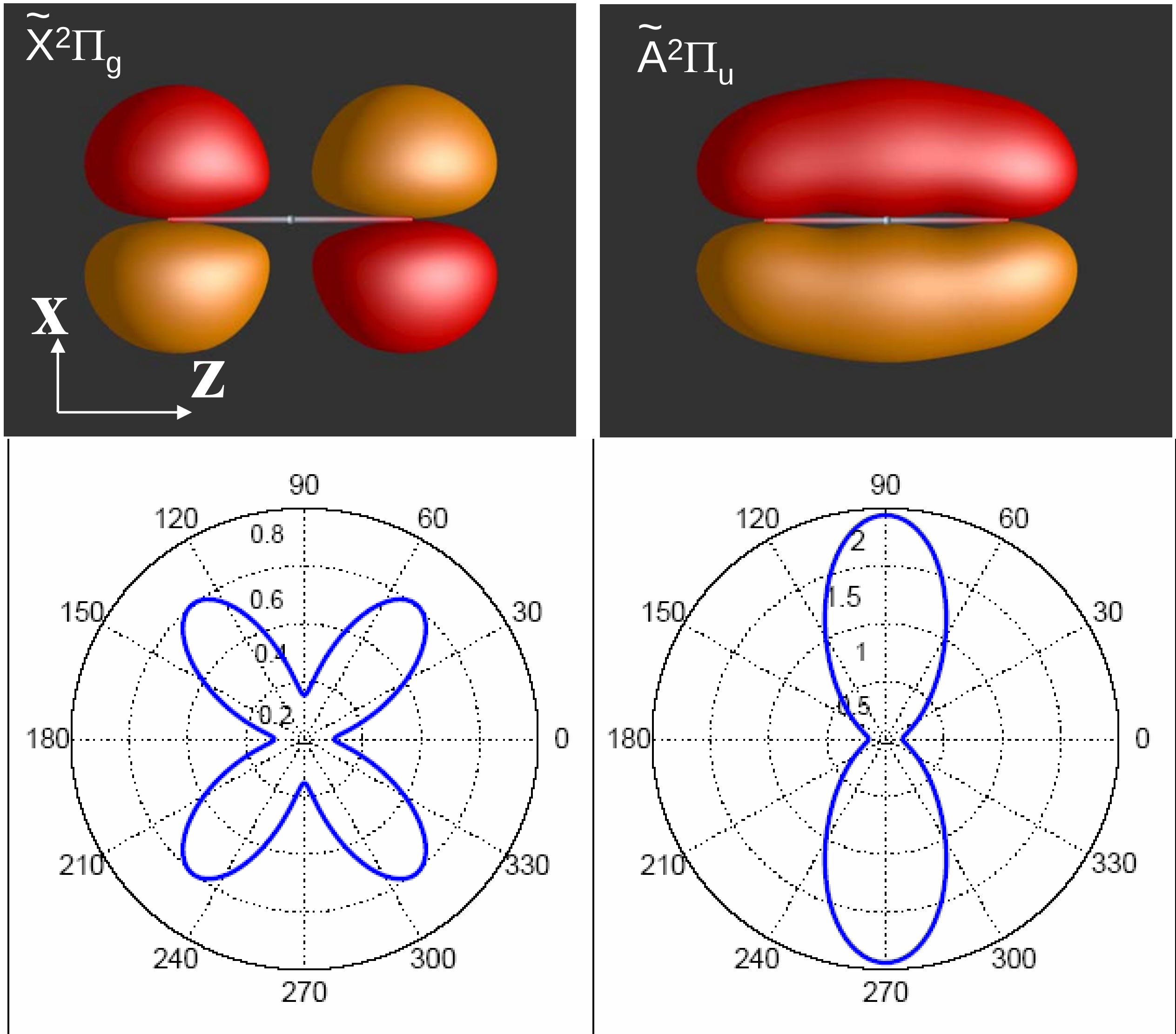

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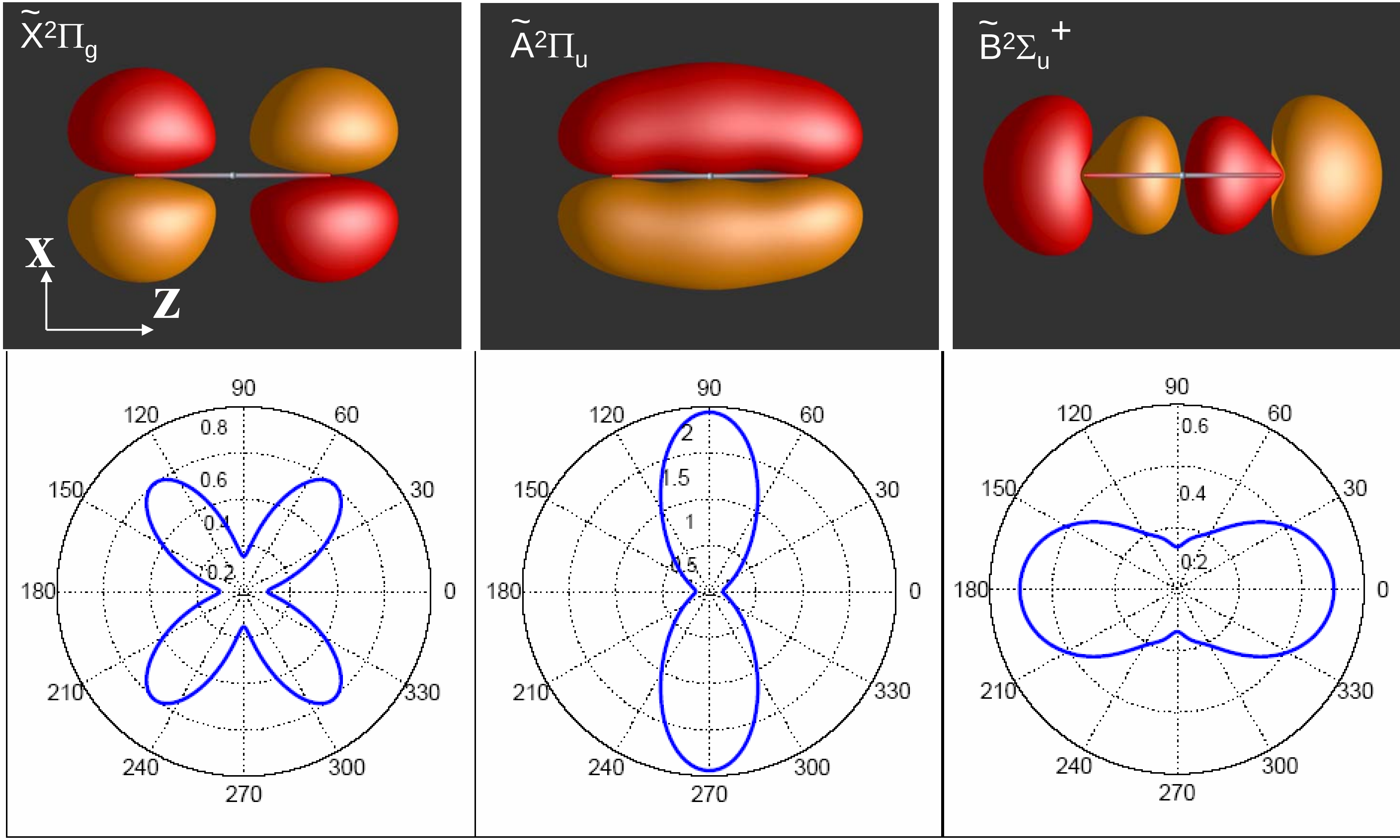

Ionization: intermediate conclusions

- Strong-field ionization almost always creates dynamics in the ion (Not specific for CO_2)
- This dynamics is different for different orientations of the molecule with respect to the laser field

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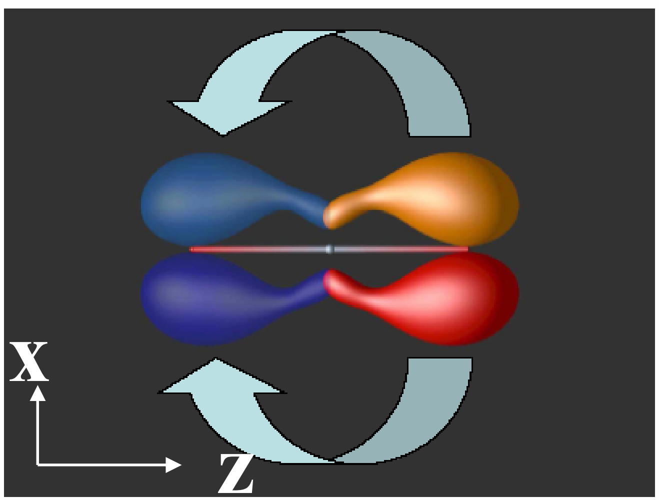
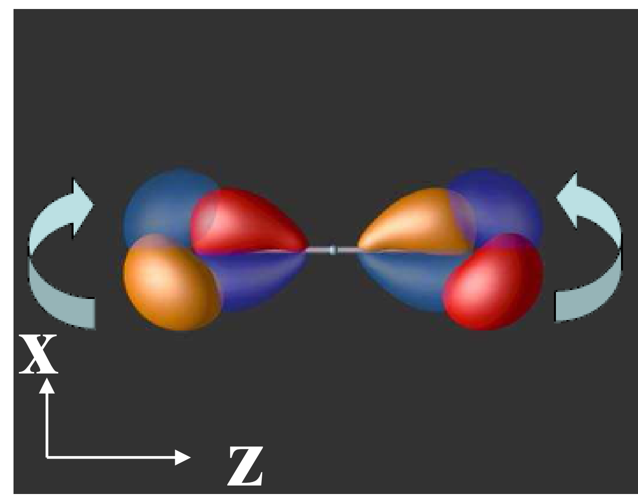
Visualization of hole dynamics

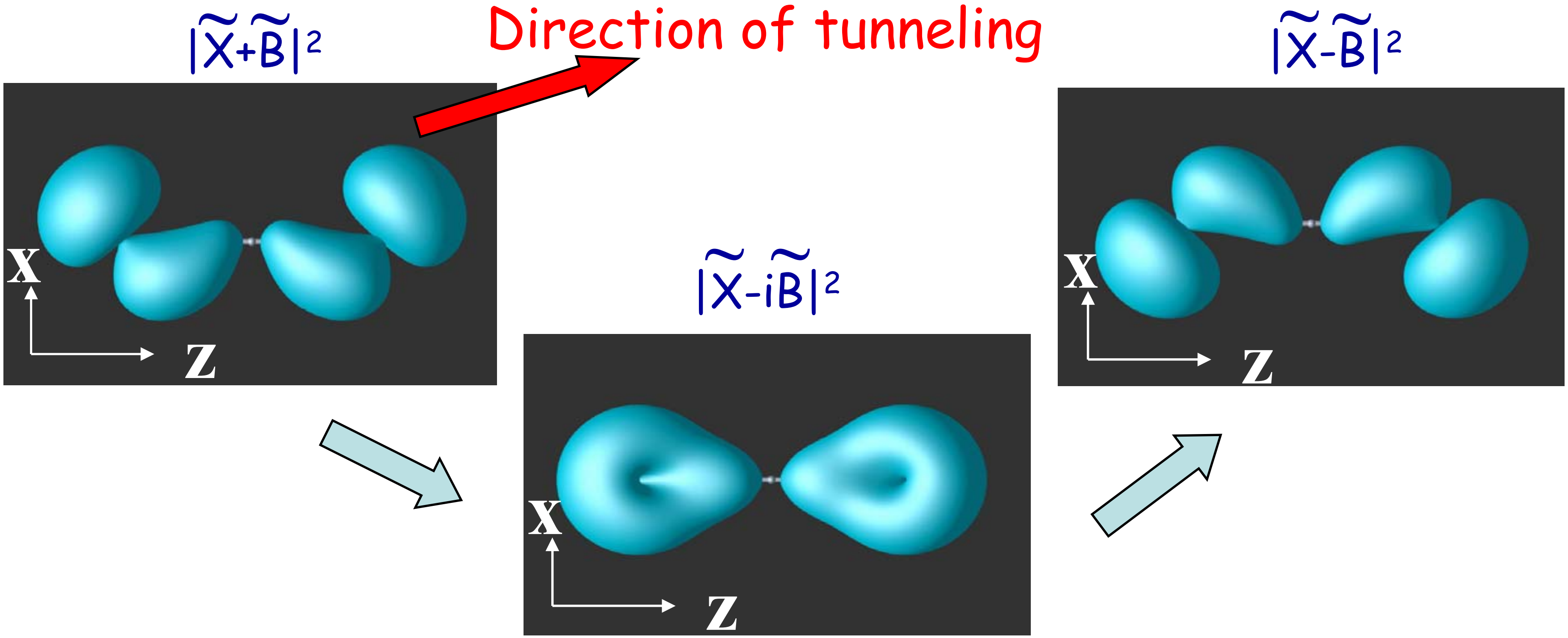
$\tilde{X},\tilde{A}$ channels



$\tilde{X},\tilde{B}$ channels



Visualization of hole dynamics for $\tilde{X},\tilde{B}$ channels



Where does the hole begin its motion after tunnel ionization, and how does it move?

Theory needs to address several problems…

New questions to theory

1. Channel coupling during ionization (laser-induced)
2. Interaction between the departing electron and hole

Main message

1. Channel coupling during ionization (laser-induced)
2. Interaction between the departing electron and hole

↓

1. Affects ionization rates into each channel
2. Affects ***phase*** between channels

Reflected in the shape and location of the hole

Main message

1. Channel coupling during ionization (laser-induced)
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⇓

1. Affects ionization rates into each channel
2. Introduces a new parameter: ***phase*** between channels

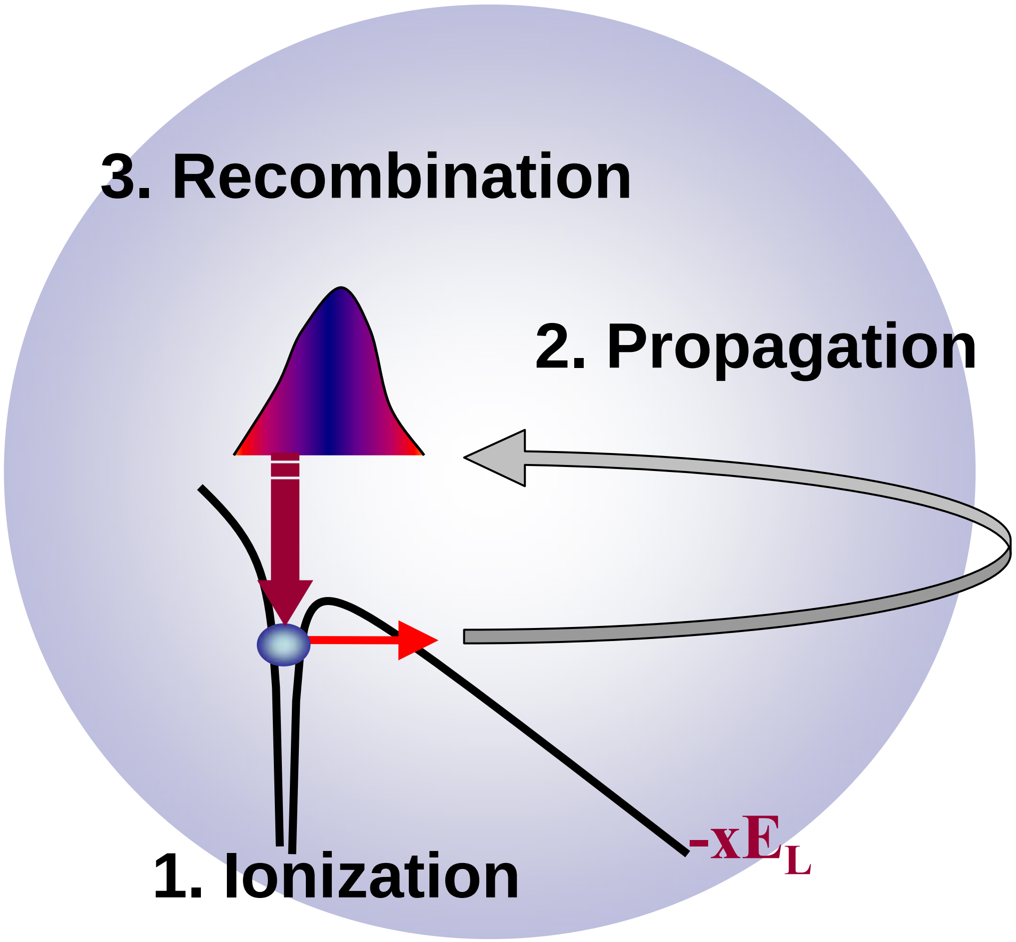
Reflected in the shape and location of the hole

How can we image the hole dynamics during tunneling?

Can we measure the phase of ionization?

We need to record relative phase between different channels

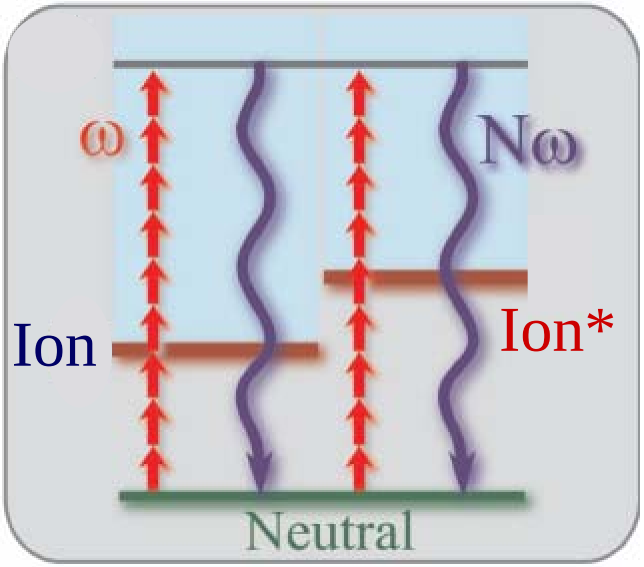
High harmonic spectroscopy



Same initial and final states: ground state of the neutral

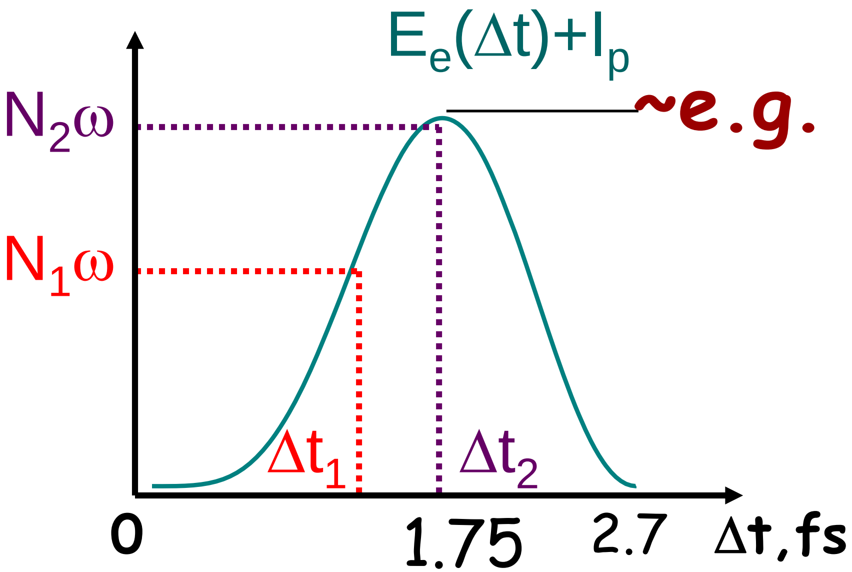
Multiple channels in HHG

Different ionization channels-
different HHG channels



Interference records relative phase between the channels by mapping it into harmonic amplitude modulations!

High harmonics: temporal resolution



~e.g. 60 eV

2fs/20 odd harmonics~100 asec

Δt - time delay between ionization and recombination

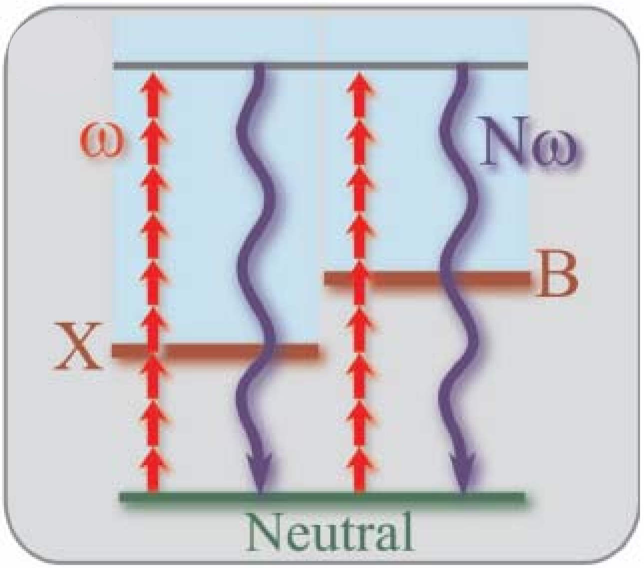
Each harmonic takes snapshot of the system at a particular time delay between ionization and recombination

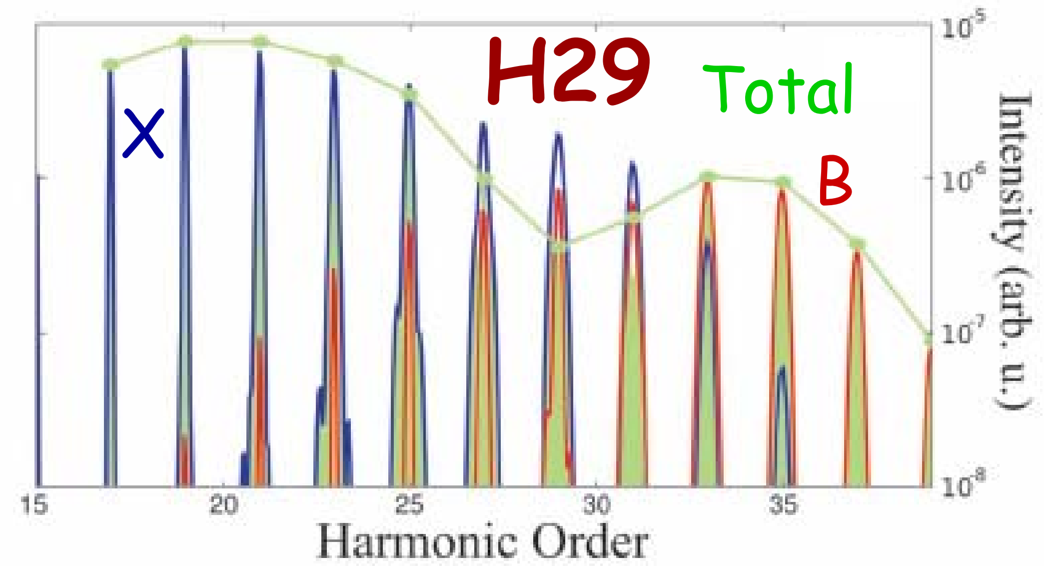
Atto-second temporal resolution

M. Lein,

S. Baker et al

Getting initial phase





H29 ➡ τ^*

Destructive interference at τ^*

$$\Delta\phi_{XB}(\tau^*)=(2n+1)\,\pi$$

$$\Delta\phi_{XB}=(E_X-E_B)\,\tau^*+\Delta\phi_{rec}+\Delta\phi_{ion}$$

$$\Delta\phi_{XB}(\tau=0)=\Delta\phi_{rec}+\Delta\phi_{ion}$$

Dynamical minimum is tied to τ^*, not harmonic number!

Modeling HHG: including different channels

$$\vec{D}(t) \propto < \Psi^{(N)}_{NEUT}(t) | \vec{d} | \Psi^{(N)}_{Ie}(t) >$$

$$\Psi^{(N)}_{Ie}(t) \propto \sum_j a^{ion}_j [t_b(t)] \; \hat{A}\Psi^{(N-1)}_{ION,j}(t)\Phi_{CONT,j}(t)$$

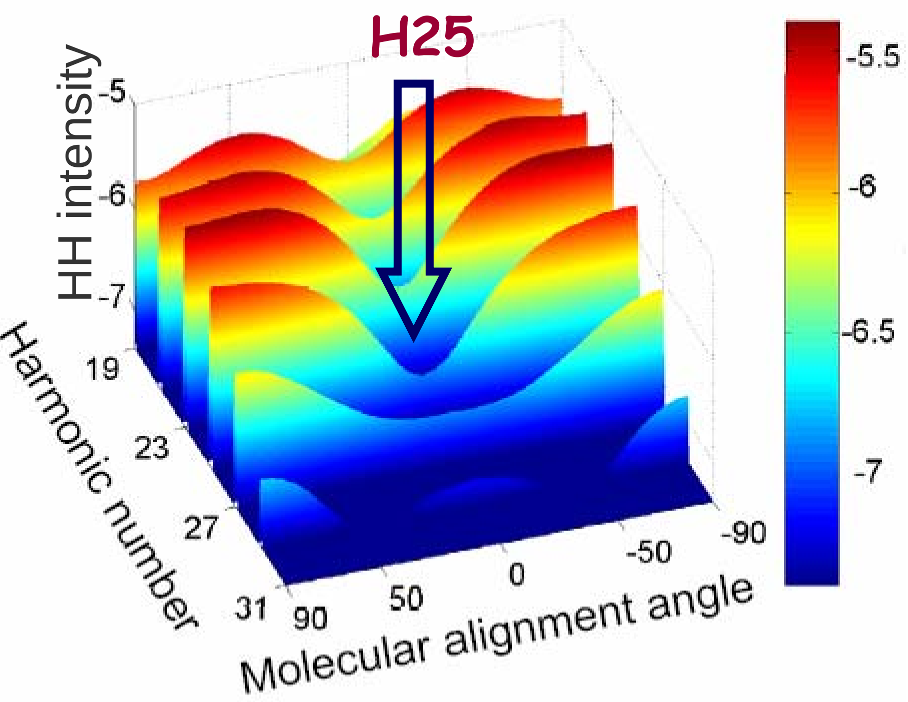
j – labels different states of the ion

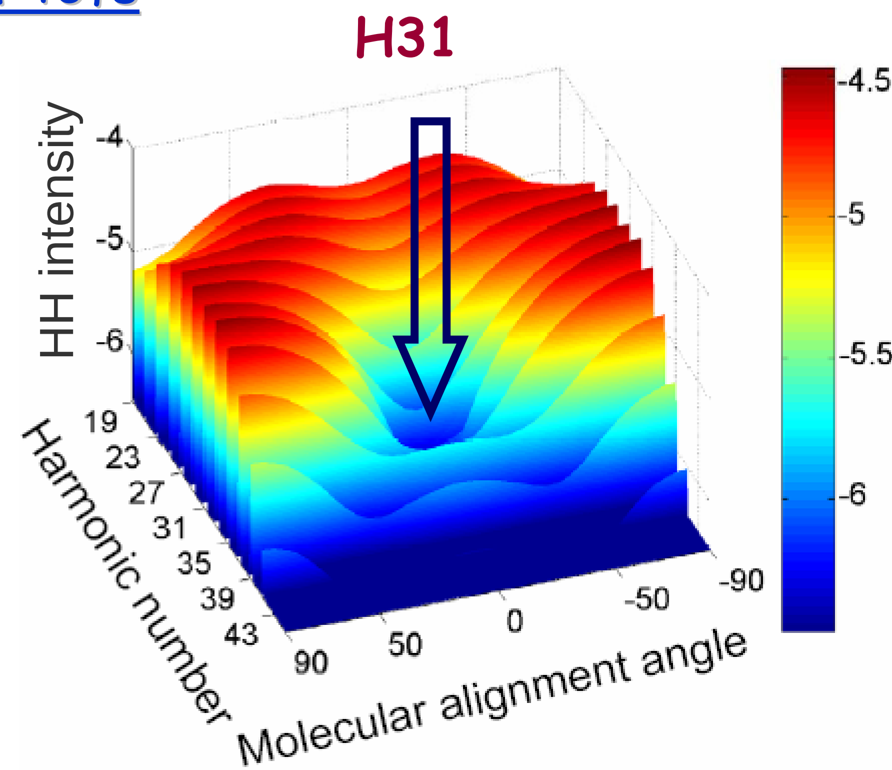
- Ψ_{NEUT} (t) – MC SCF, quasi-static field, for all relevant electrons (CAS)
- $\Psi_{ION,\,j}$ (t)
  - MC SCF for all (j=1..5) essential stationary ionic states, field-free
  - Calculate all dipole couplings between these states
  - Do TDSE dynamics in restricted basis of these ionic states
  - Channel-specific Hartree potential : **Hartree [$\Psi_{ION,J}$]**
- a_{ion} : syb-cycle YudinIvanov* orbital-dependent angular factor (CC-VGSFA)
- $\Phi_{CONT\,,\,j}$ (t) from SF EVA (*Smirnova, Spanner, Ivanov PRA (2008)*)

Amplitude Minima in CO_2

$I=1.1*10^{14} W/cm^2$ $I=2.0*10^{14} W/cm^2$

800nm 40fs

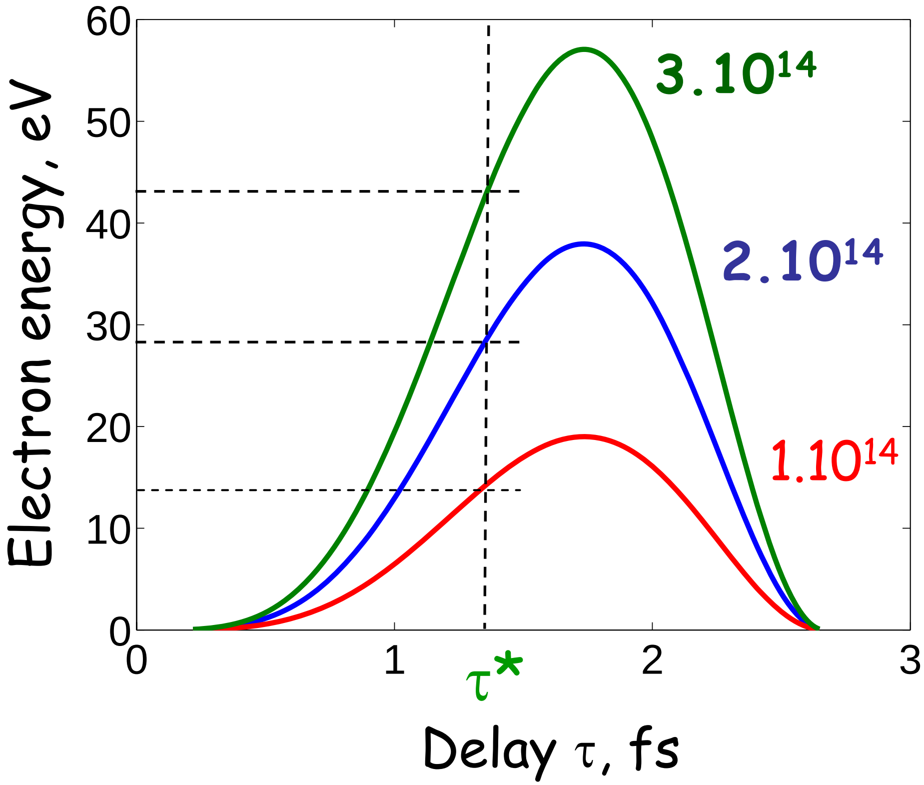


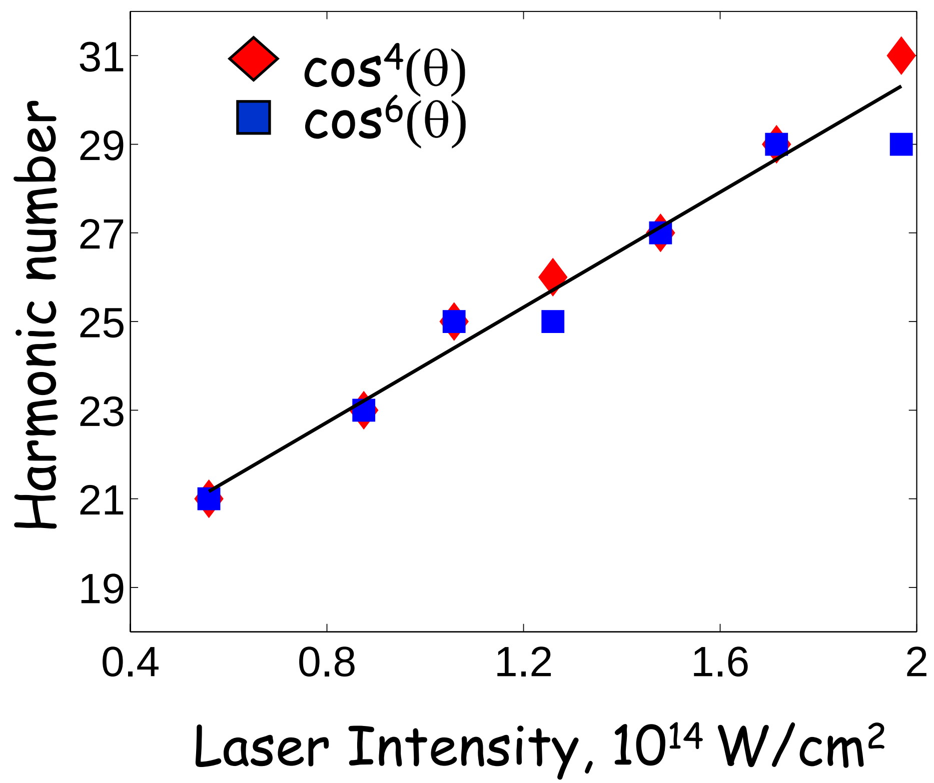


Minimum shifts with intensity

It is a signature of dynamical minimum and channel interference

Intensity dependence of the dynamical minimum



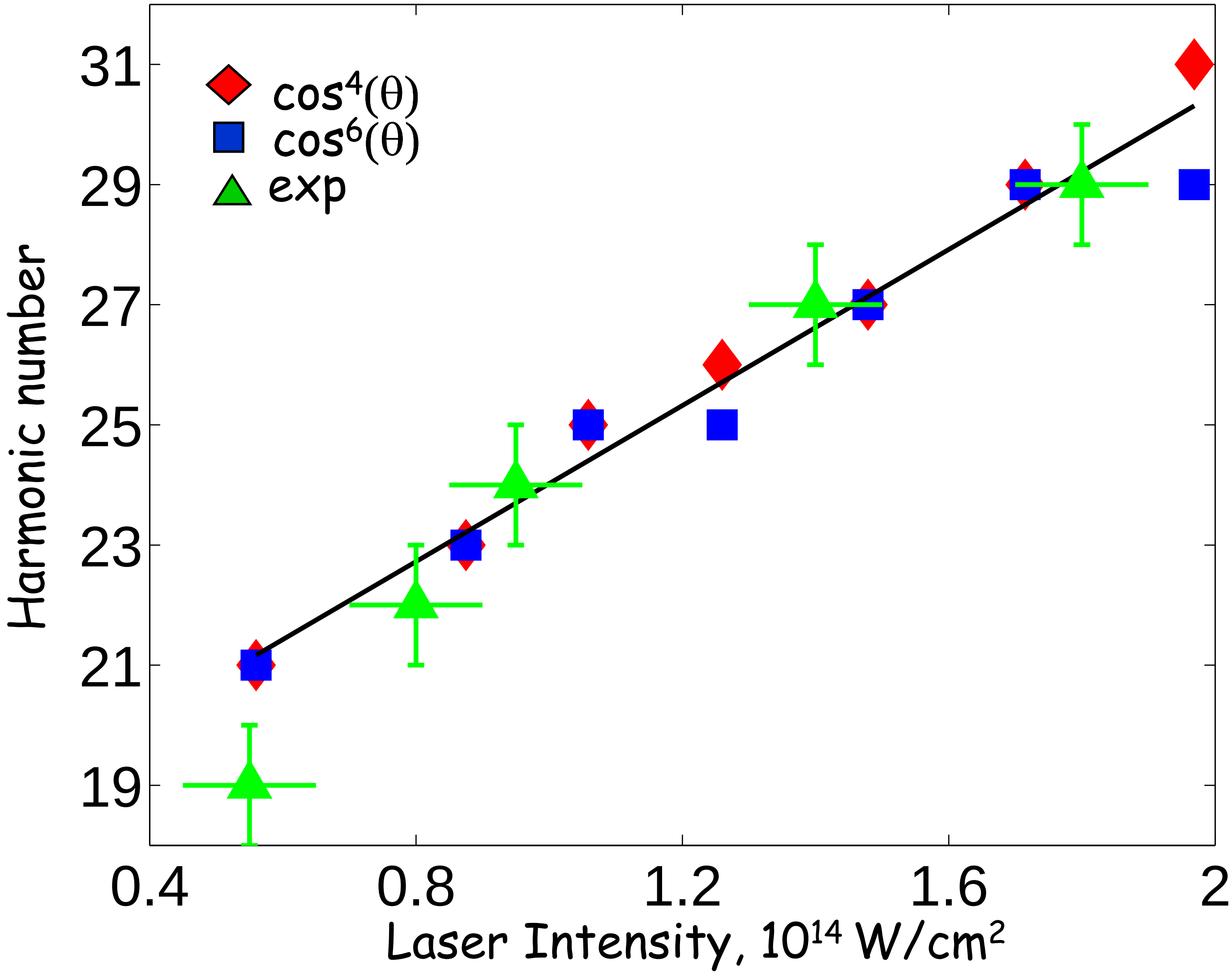


$$N^*\omega = E_e(\tau^*) + I_p$$

$$N^*\omega \propto (1.7 \pm 0.2) U_p \;\longrightarrow\; \tau^* = 1.17 \pm 0.1\,\text{fs}$$

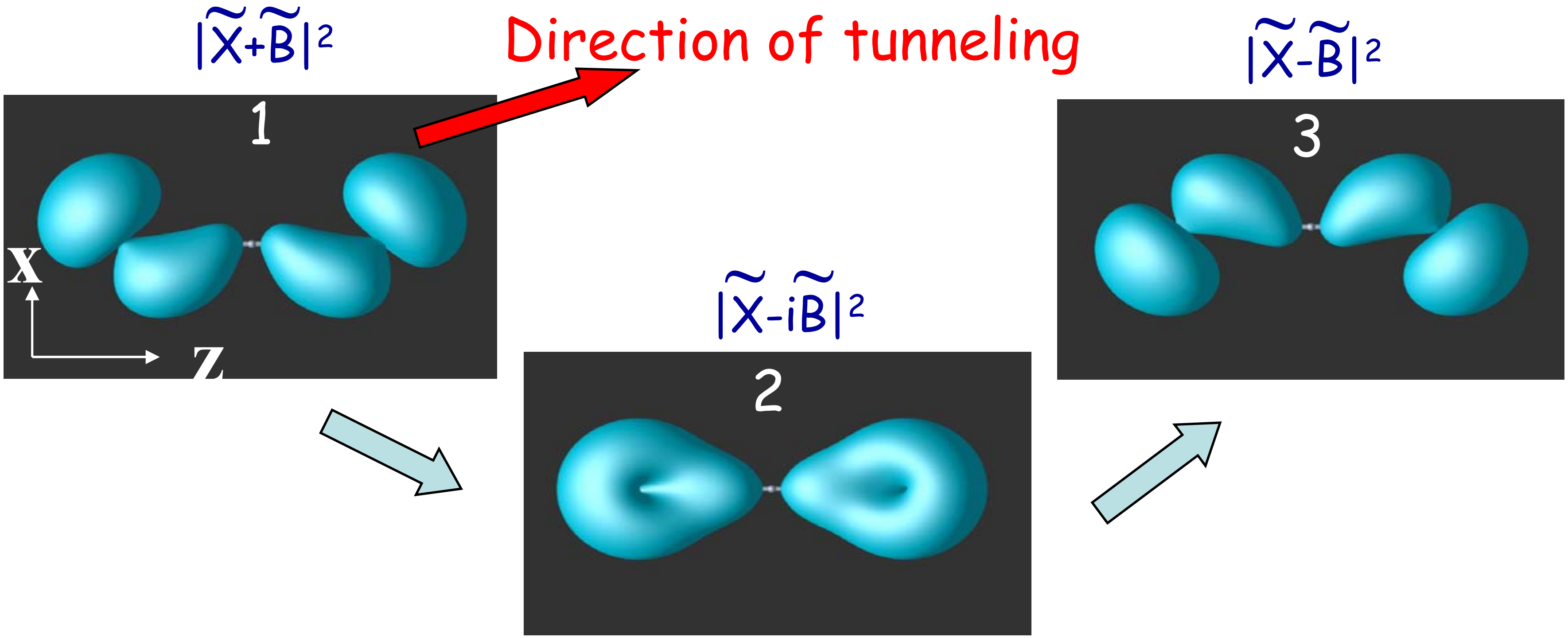
Theory vs experiment

Experiment: Yann Mairesse, Nirit Dudovich, David Villeneuve, Paul Corkum



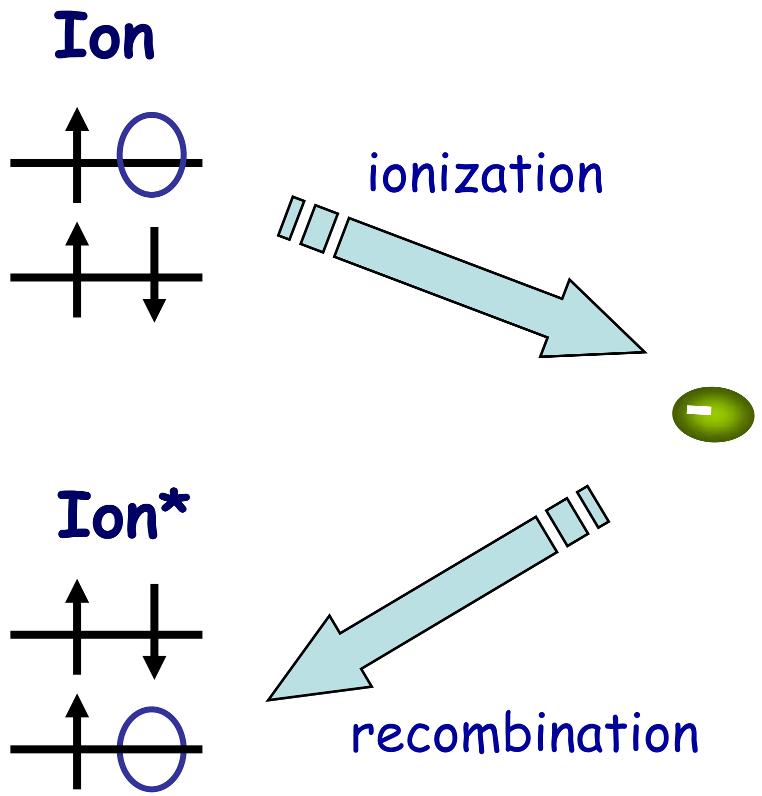
The harmonic movie decoded

- Visualization of hole dynamics for aligned CO_2: $\tilde{X},\tilde{B}$ channels



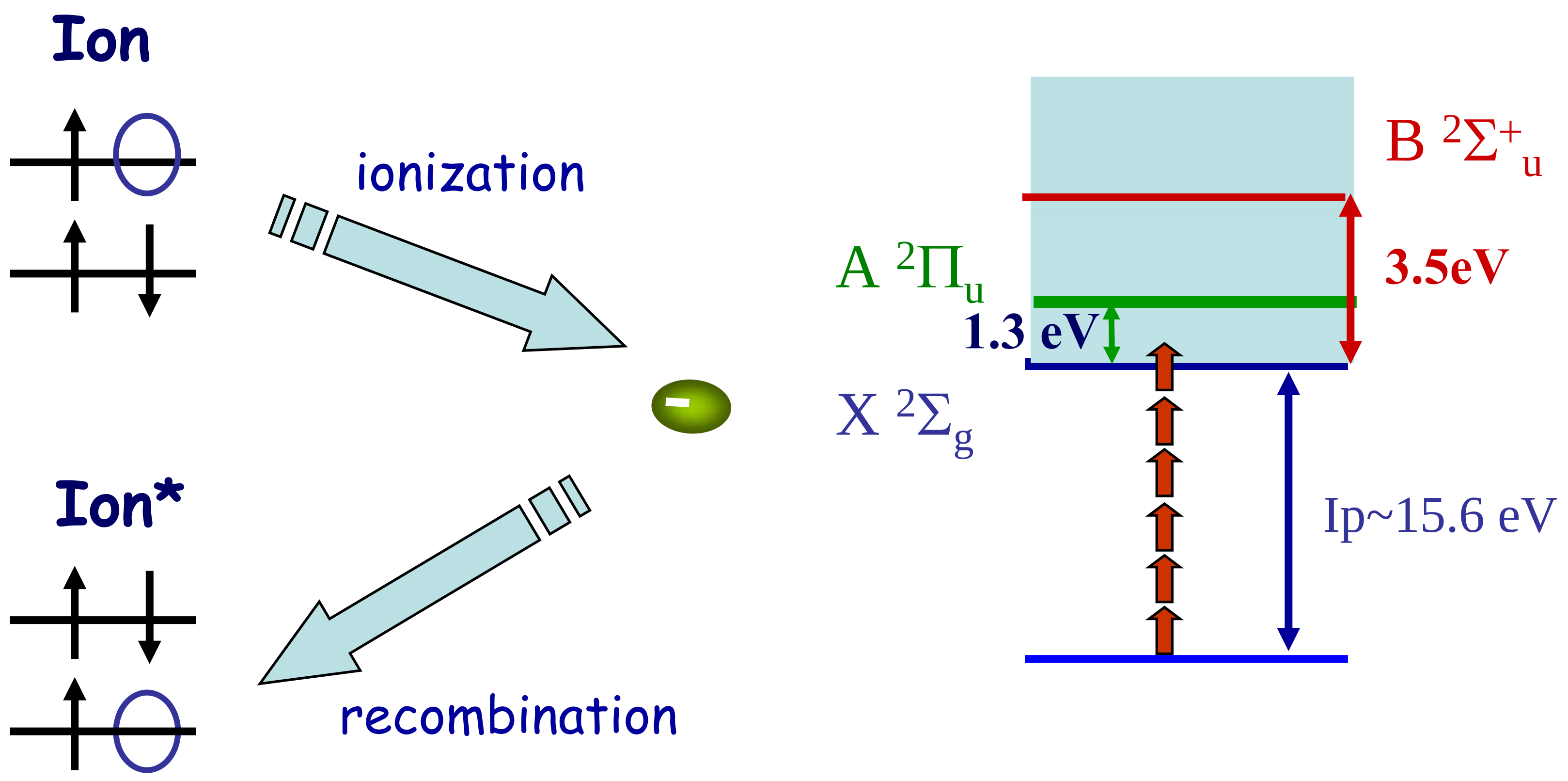
- In tunneling regime, the initial phase between $\tilde{X}$ and $\tilde{B}$ is zero, see frame 1: maximum extension in the direction of tunneling.

Attosecond laser-induced dynamics



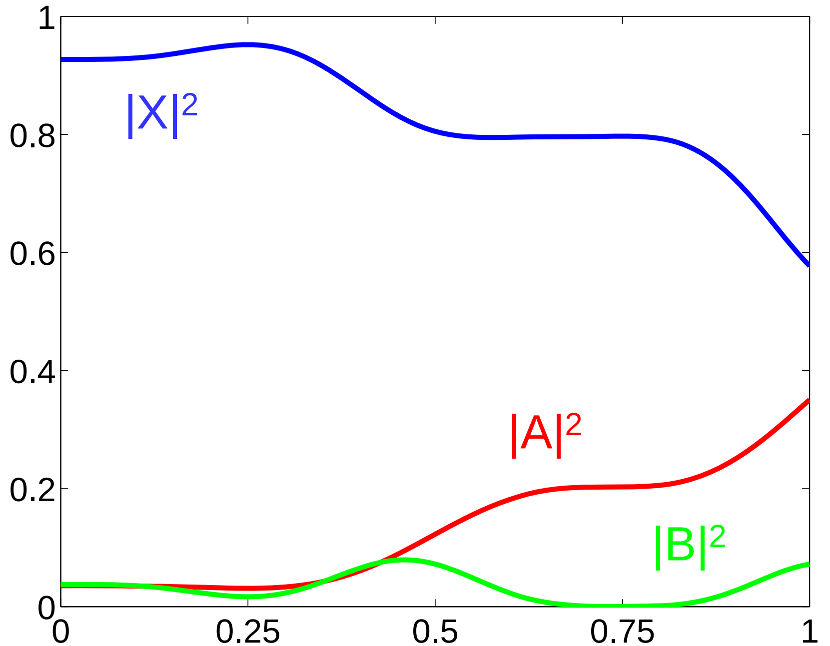
Consider N_2

Laser-induced dynamics in N_2

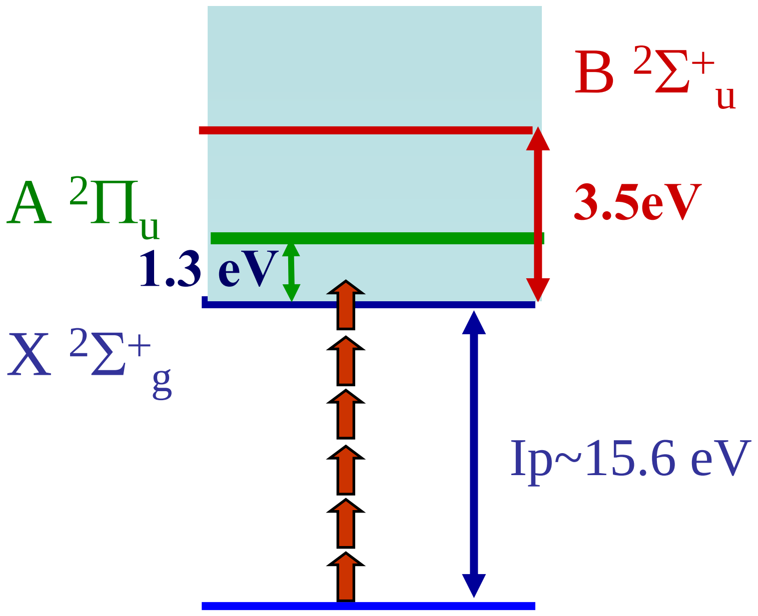


Laser-induced dynamics in N_2

Populations of ionic states



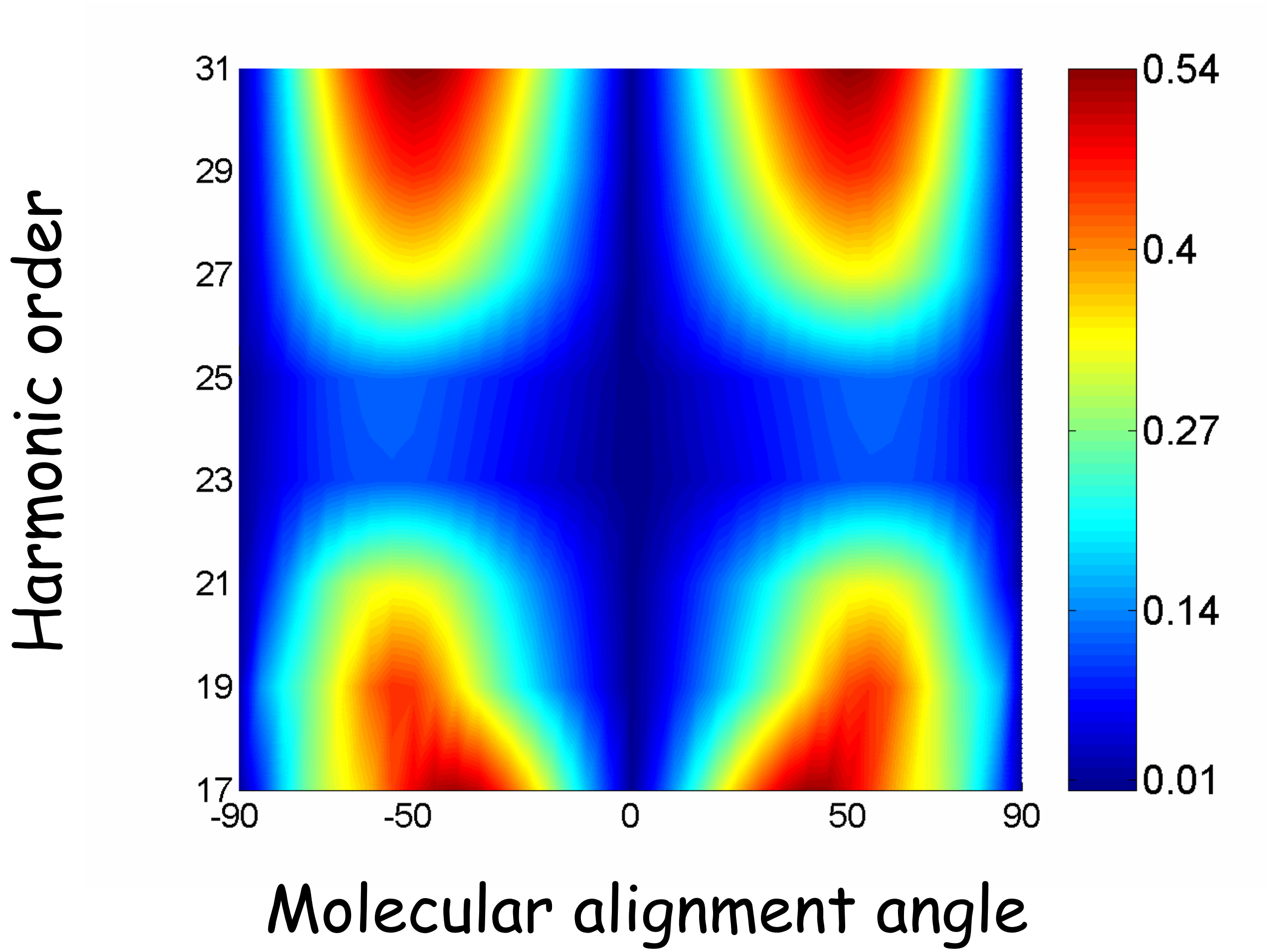
Time, laser cycle



Is there any special property of harmonic emission associated with this dynamics?

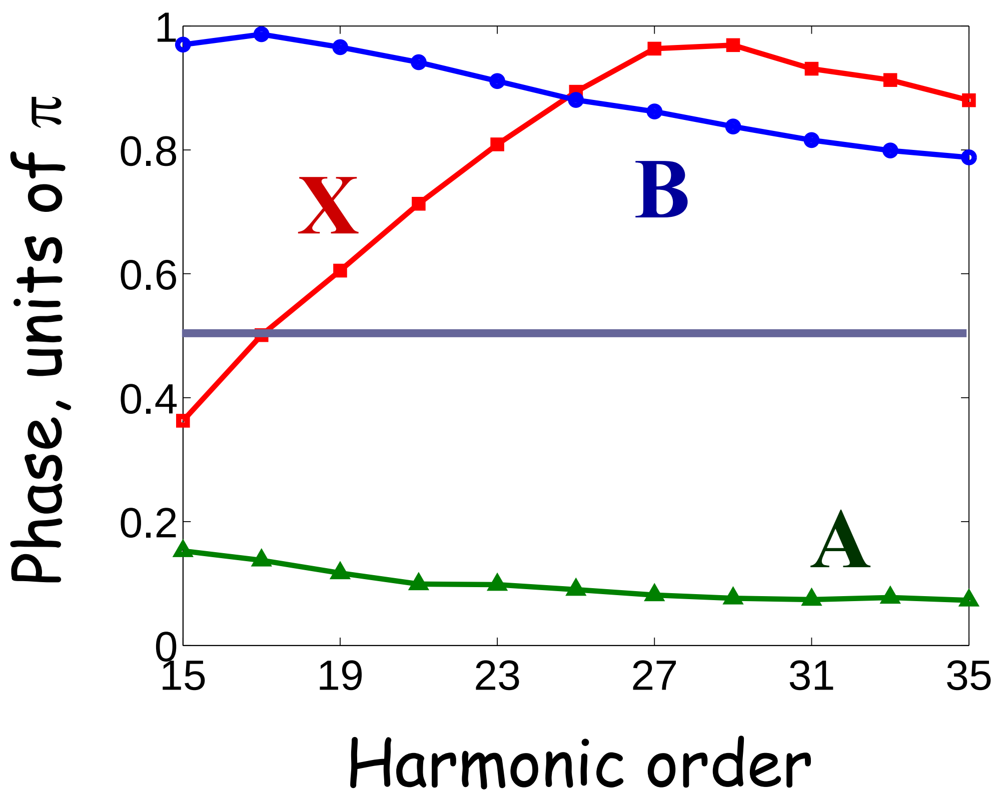
Strong ellipticity of harmonic light

I=1.3 10^{14}W/cm^2



Phase between parallel and perpendicular light

Θ=50 deg   I=1.3 10^{14}W/cm^2



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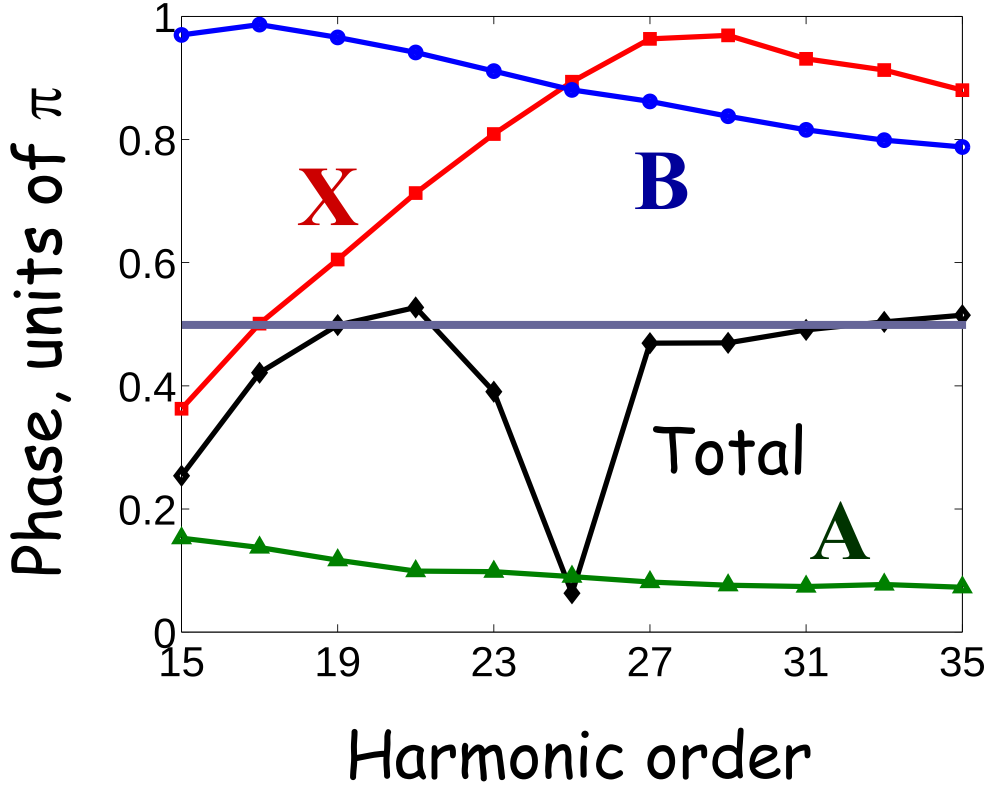
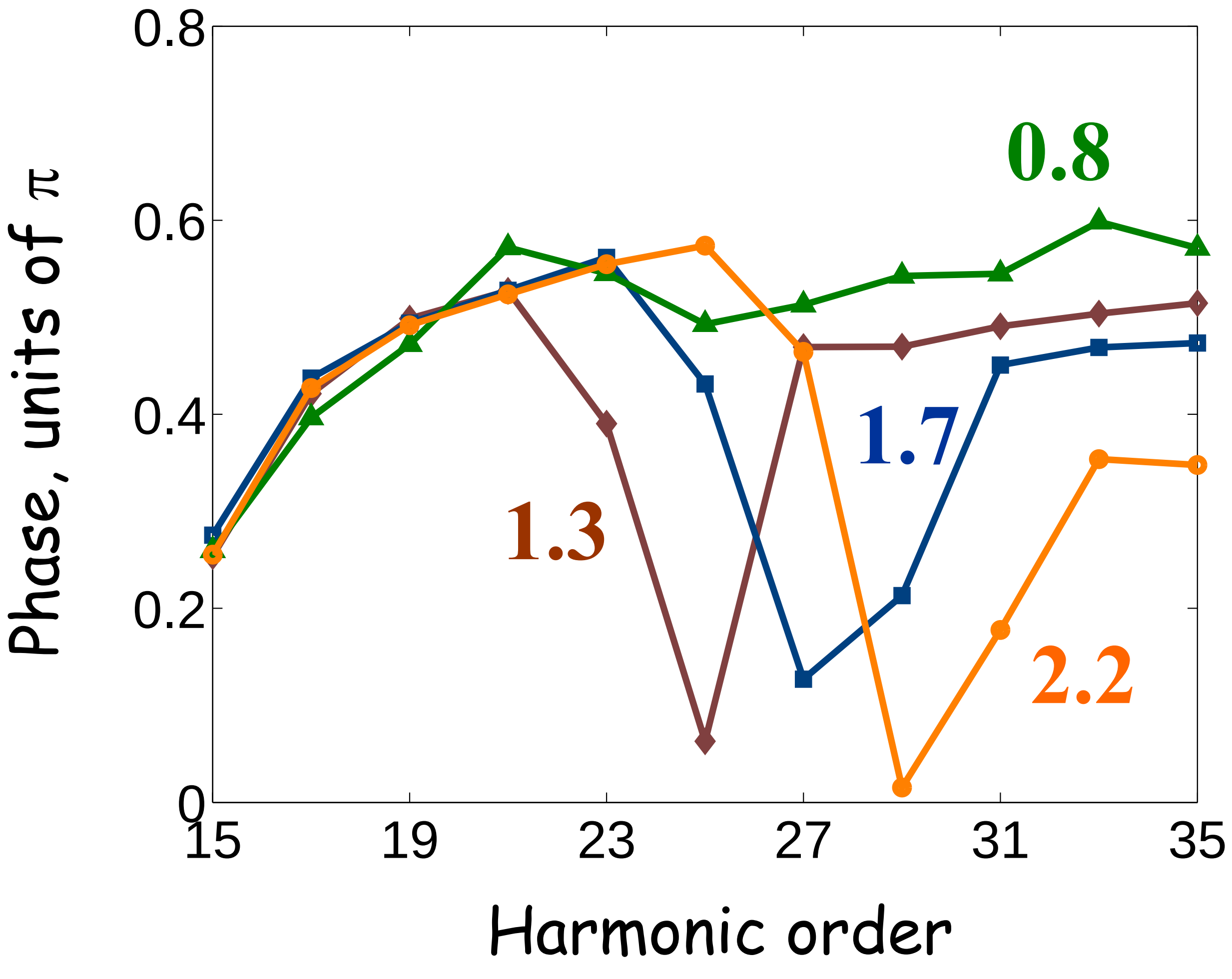


Dynamical min in N_2

Phase between parallel and perpendicular light vs Intensity



Small ellipticity indicates dynamical min in perpendicular light

HH Spectroscopy of Polyatomic Molecules:

What do we learn that we do not know?

Conclusions

New questions for theory and experiment:

- Channel coupling during strong-field ionization
- Relative phase of strong-field ionization between channels, in tunneling and multi-photon regimes.

N_2:

- Strong interaction between the channels is the reason of high ellipticity of harmonic light
- Small ellipticity indicates the position of dynamical minimum for N_2
- Controling laser-induced dynamics of ionic states one can shape the polarization of asec pulses.