The phase in molecular high-harmonic generation

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VERSITÄT

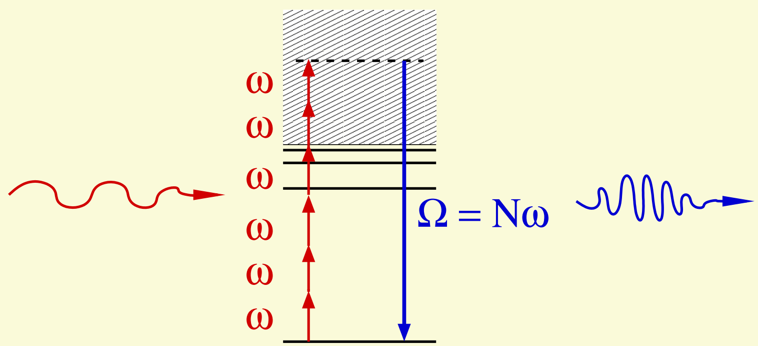
Coworkers: Elmar van der Zwan, Marcelo Ciappina, Ciprian Chirilă

Outline

- High-harmonic generation (HHG) in molecules
- Orbital tomography
- Orientation dependence of harmonic phase

High-harmonic generation

HHG process

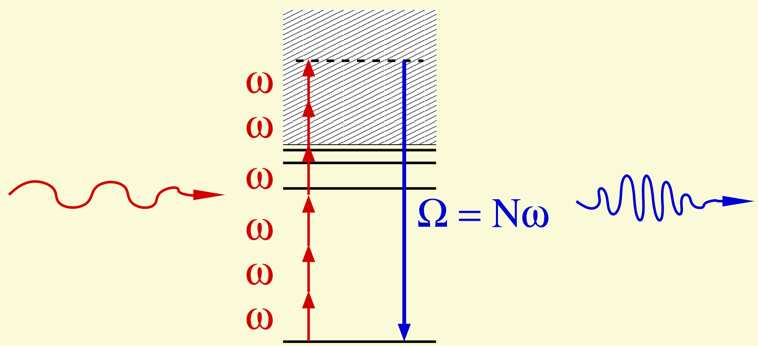


N photons of frequency ω

$\longrightarrow$ 1 photon of frequency $N\omega$.

High-harmonic generation

HHG process



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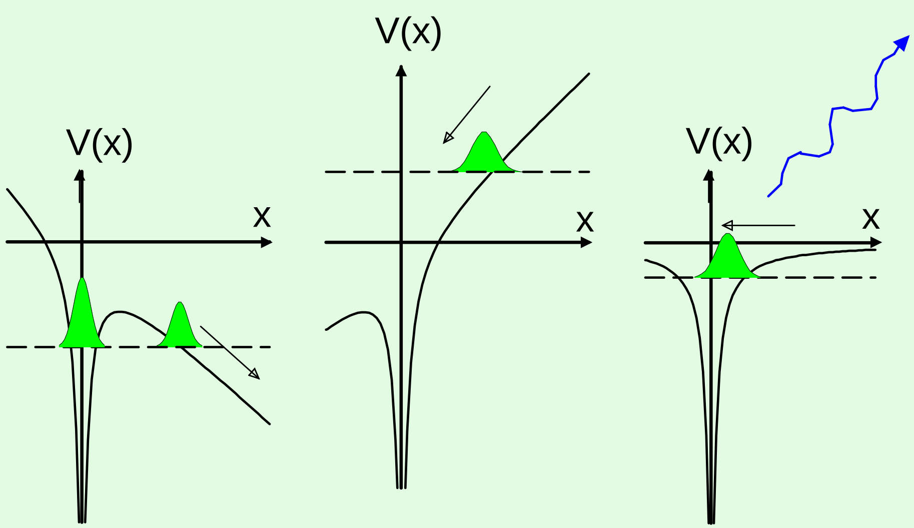
Recollision picture

Ionization ***Free acceleration*** ***Recombination***

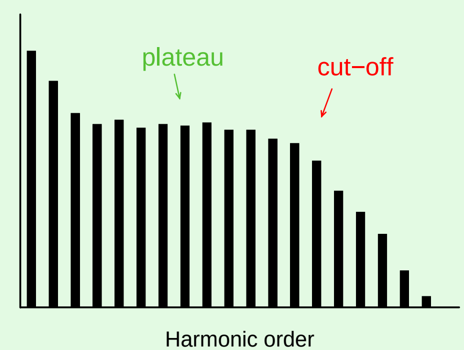


Maximum return energy:

$$E_{\rm max} = 3.17 U_{\rm p}$$

$\hookrightarrow$ Cut-off at $\boxed{\hbar\omega = 3.17 U_{\rm p} + I_{\rm p}}$

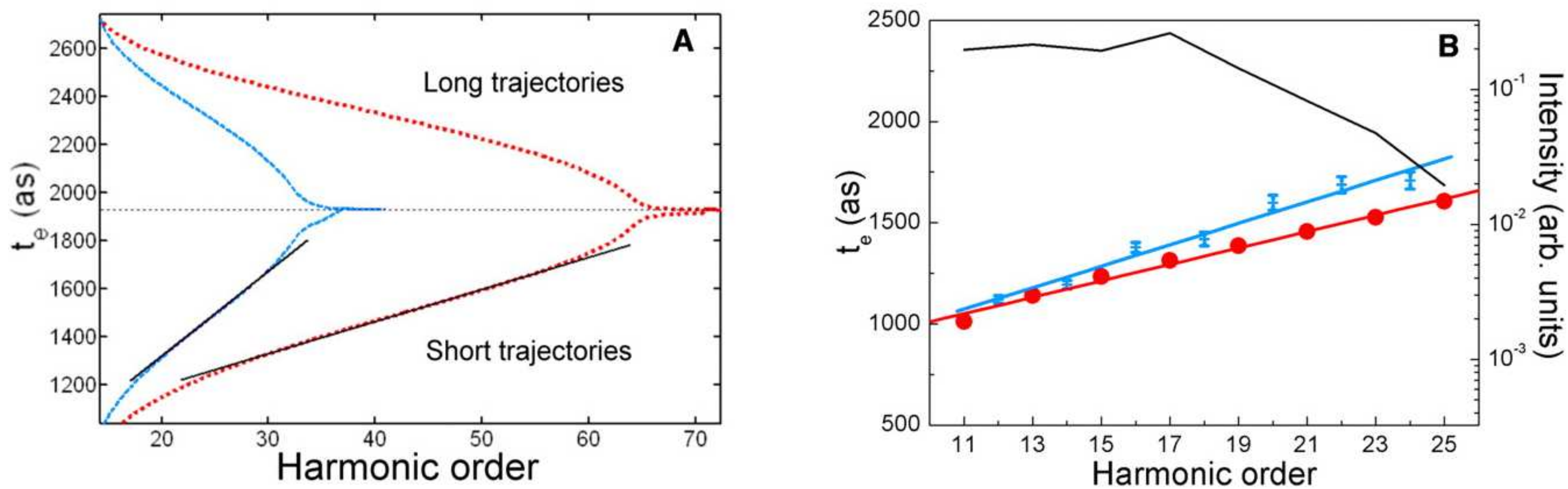
[Corkum, Schafer,. . . 1993]



Phase and emission time in HHG

Emission of high-harmonics is *chirped*:

Theory / Measurement



Y. Mairesse et al., Science 302, 1540 (2002)

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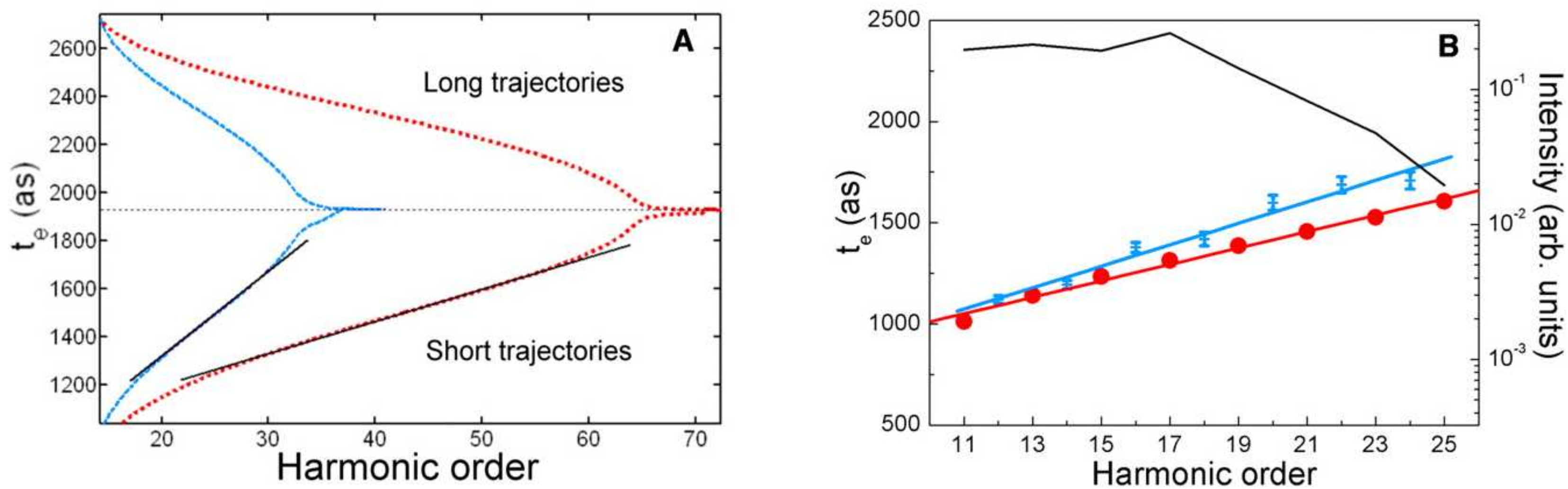


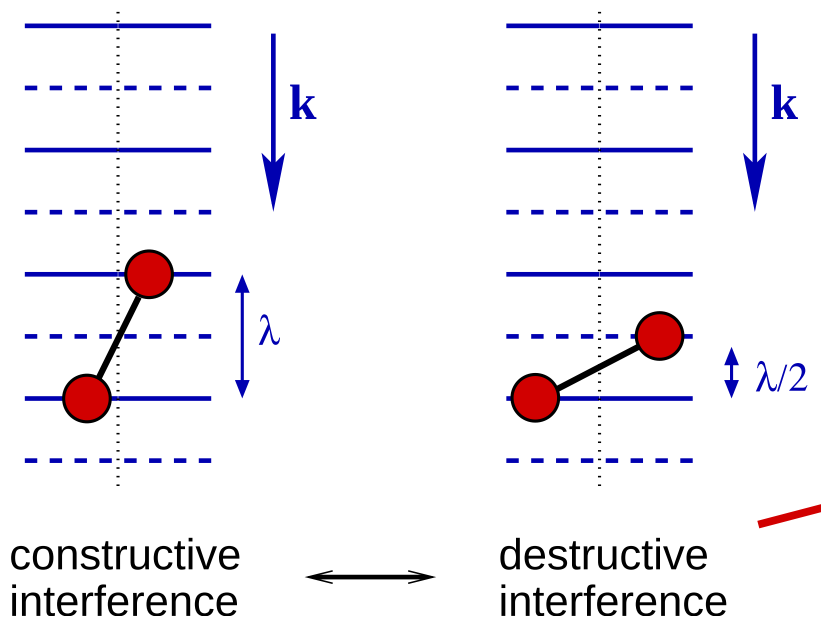
Y. Mairesse et al., Science 302, 1540 (2002)

Emission time is directly related to phase: $t_e = \frac{d\varphi}{d\omega}$

$\rightarrow$ Harmonic phase is essential for attosecond pulse shapes!

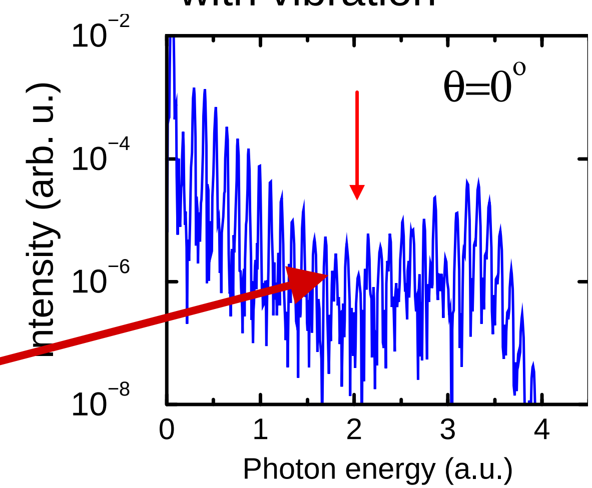
Two-center interference in molecules

Recolliding electron with wave vector $\mathbf{k}$



TDSE for H_2 in 2D

with vibration



M.L., N. Hay, R. Velotta, J.P. Marangos, P.L. Knight, PRL **88**, 183903 (2002),

Experimental confirmation for CO_2:

T. Kanai et al., Nature 435, 470 (2005)

C. Vozzi et al., PRL 95, 153902 (2005)

Theory of high-harmonic generation

Strong-field approximation (SFA, Lewenstein model)

$$\mathbf{D}(t) = i\int_0^t dt' E(t') \int d^3p \, \langle \mathbf{p}+\mathbf{A}(t')|x|\Psi\rangle \langle\Psi|\mathbf{r}|\mathbf{p}+\mathbf{A}(t)\rangle \exp[-iS(\mathbf{p},t,t')] \; + \text{c.c.}$$

where $S(\mathbf{p},t,t') = \int_{t'}^{t} dt'' \left[\frac{(\mathbf{p}+\mathbf{A}(t''))^2}{2} + I_\mathrm{p}\right]$

[Lewenstein et al., Phys. Rev. A **49**, 2117 (1994)]

Acceleration: $\mathbf{a}(t) = d^2\mathbf{D}(t)/dt^2$

Spectrum: $S(\omega) \sim |\mathbf{a}(\omega)|^2$

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Spectrum: $S(\omega) \sim |\mathbf{a}(\omega)|^2$

Choice of recombination operator:

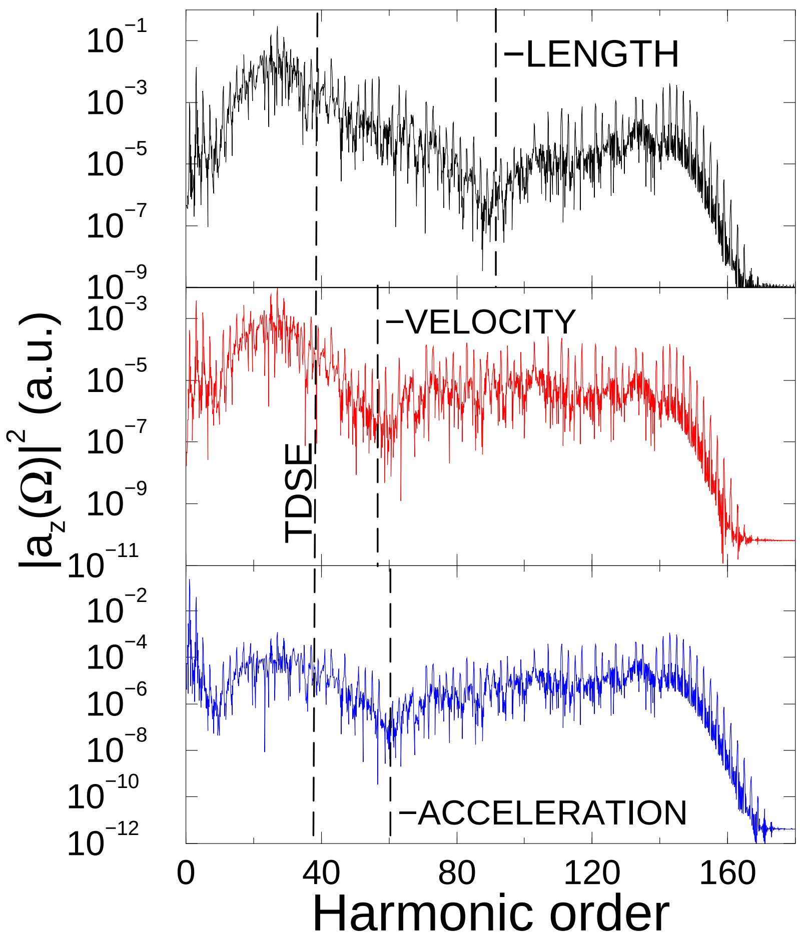
velocity or acceleration form preferable to length form

G.L. Kamta, A.D. Bandrauk, PRA 71, 053407 (2005)

A. Gordon, F.X. Kärtner, PRL 95, 223901 (2005)

Choice of recombination operator

Harmonics from H_2^+, $\theta = 40^o$, laser intensity 10^{15} W/cm^2, $\lambda = 800\,$nm



Dashed lines: two-center interference minimum

C.C. Chirilă and M. L., J. Mod. Opt. **54**, 1039 (2007).

Theory of high-harmonic generation

Lewenstein model in velocity form:

$$\mathbf{P}(t) = \int_0^t dt' E(t') \int d^3p \, \langle \mathbf{p}+\mathbf{A}(t')|x|\Psi\rangle \, \langle\Psi|\nabla|\mathbf{p}+\mathbf{A}(t)\rangle \, e^{-iS(\mathbf{p},t,t')} \, + \mathbf{c.c.}$$

Theory of high-harmonic generation

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Saddle-point approximation for momentum integral:

$$\mathbf{P}(t) = \int_0^t dt' E(t') \left[\frac{2\pi}{\epsilon+\mathrm{i}(t-t')}\right]^{\frac{3}{2}} \langle \mathbf{p}_\mathrm{s}+\mathbf{A}(t')|x|\Psi\rangle \, \langle \Psi|\nabla|\mathbf{p}_\mathrm{s}+\mathbf{A}(t)\rangle \, e^{-iS(t,t')} + \text{c.c.}$$

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Saddle-point approximation for recombination time:

$$\mathbf{a}(\omega) = \int_0^T dt' E(t') \sum_{t_\mathrm{s}} \sqrt{\frac{(2\pi)^4 \mathrm{i}\omega^2}{[\epsilon+\mathrm{i}(t-t')]^3 \left(\frac{\omega-I_\mathrm{p}}{t_\mathrm{s}-t'} + E(t_\mathrm{s}) k_\mathrm{ret}\right)}} \langle\Psi|\nabla|\mathbf{k}_\mathrm{ret}\rangle \langle \mathbf{p}_\mathrm{s}+\mathbf{A}(t')|x|\Psi\rangle e^{-iS(t')+\mathrm{i}\omega t_\mathrm{s}}$$

with the dispersion relation

$$\mathbf{k}_\mathrm{ret} = \pm \mathbf{e}_x \sqrt{2(\omega - I_\mathrm{p})}$$

Orbital tomography

Factorization into

$$\mathbf{a}(\omega) = \langle \Psi | \nabla | \mathbf{k}_{\rm ret} \rangle \times b(\omega)$$

is possible, if

- ***either***: orbital is symmetric/antisymmetric
  $\rightarrow \langle \Psi | \nabla | \mathbf{k}_{\rm ret} \rangle = \pm \langle \Psi | \nabla | -\mathbf{k}_{\rm ret} \rangle$
- ***or***: only one sign of $k_{\rm ret}$, e.g. by using *few-cycle laser pulses*

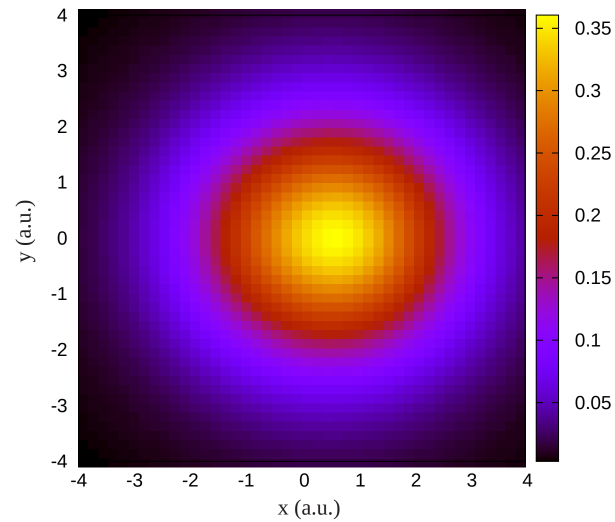
$\rightarrow$ In these cases, orbital tomography is possible.

Procedure:

1. Measure $\mathbf{a}(\omega)$. (Here: calculate exactly by solving TDSE.)
2. Use reference atom (with known orbital) to obtain factor $b(\omega)$.
3. Fourier transform to obtain molecular orbital.

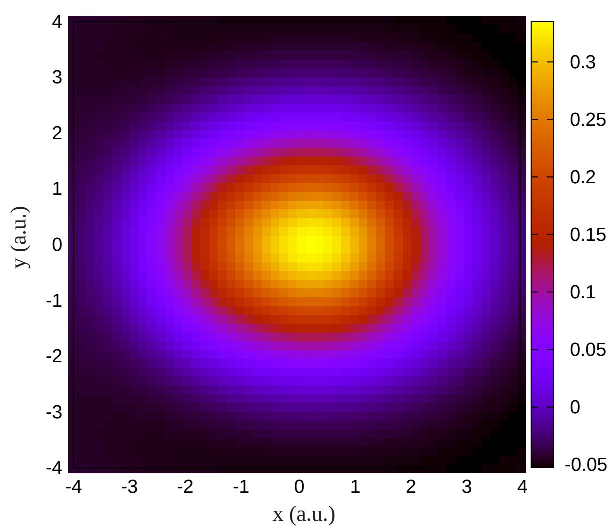
Simulation of orbital tomography

ground state for 2D 1-electron diatomic ion with $Z_1 = 1, Z_2 = 2$

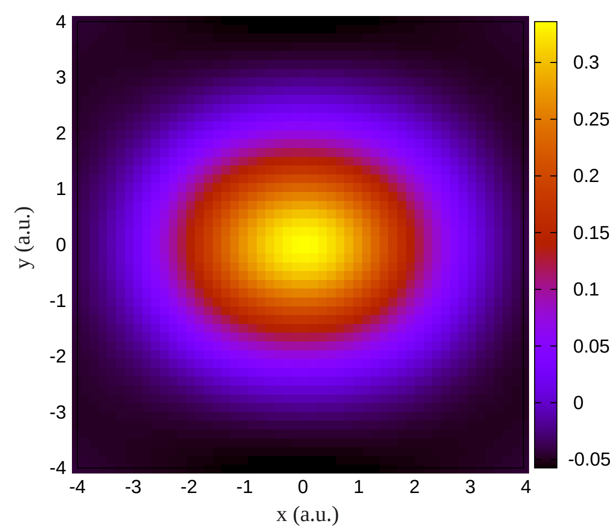


Reconstruction of the orbital using harmonic spectra/phases from TDSE:

velocity-form rec. with 3-cycle pulse

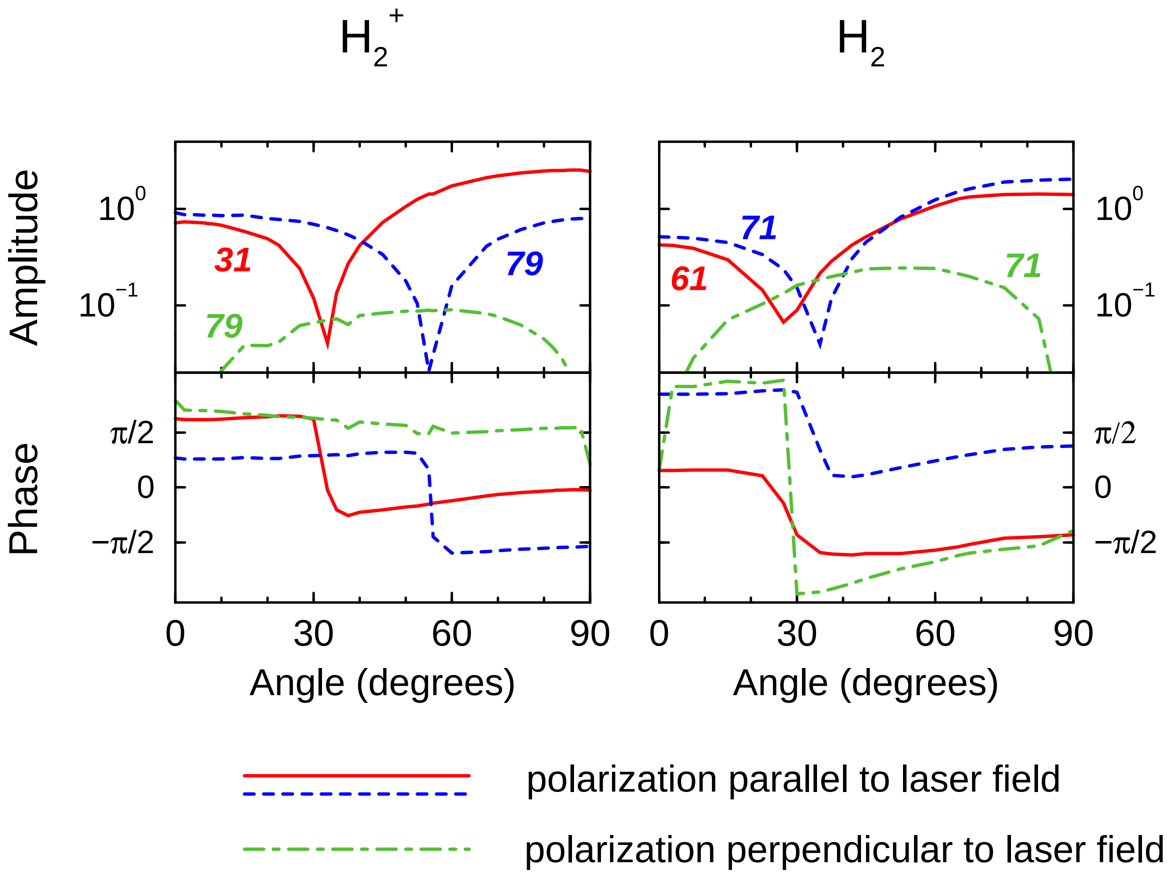


velocity-form rec. with long pulse



Phase jump in molecular HHG

Amplitude/phase dependence on molecular orientation for 2D molecules:



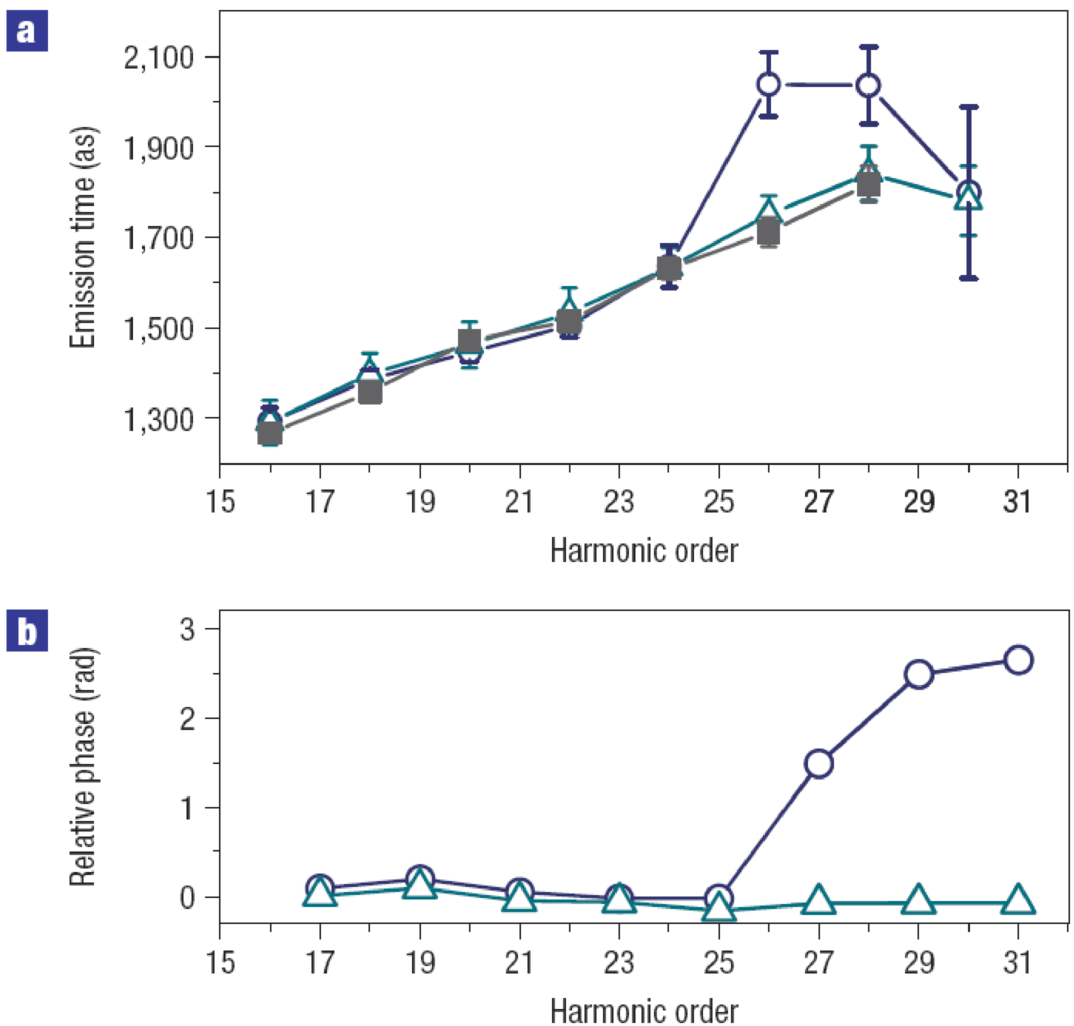
M.L., N. Hay, R. Velotta,
J.P. Marangos, P.L. Knight,
PRL **88**, 183903 (2002).

Destructive two-center interference goes along with a phase jump.

Size of the jump: $\Delta\varphi \lesssim \pi$.

Phase jump in molecular HHG

Measurement of emission time/phase of harmonics from CO_2:



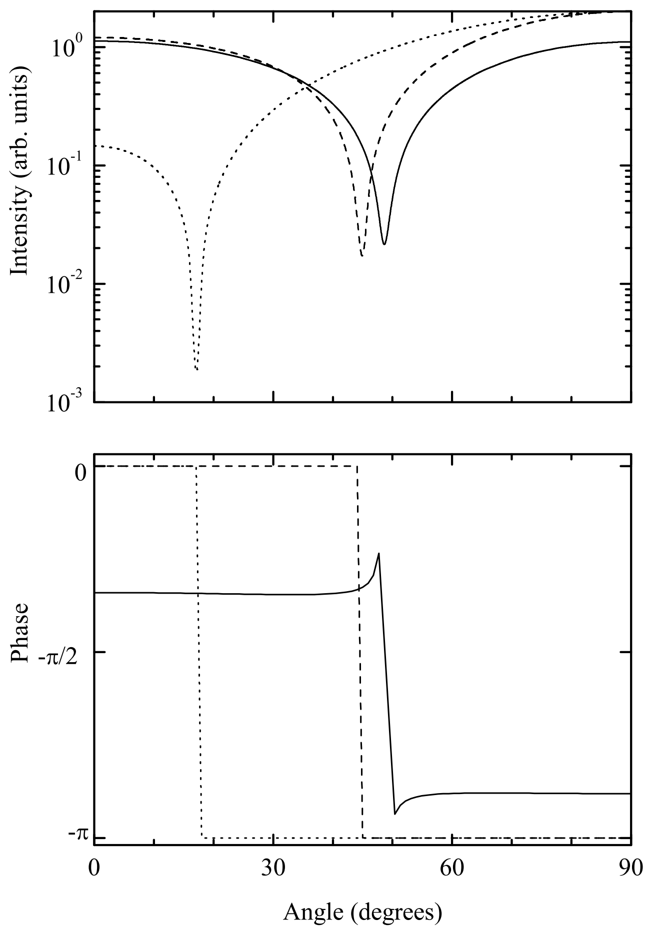
Phase jump for $CO_2||$ laser field:

- less than π
- smeared out

Boutu et al., Nature Physics (May 2008)

Effect of Coulomb corrections

Plane-wave vs Coulomb-corrected recombination amplitudes



43rd harmonic from H_2^+ in 780nm laser field

1) plane waves with $k^2/2+I_\mathrm{p}=\omega$ (dotted)

or $k^2/2=\omega$ (dashed)

2) approx. two-center Coulomb functions (solid)

$\Psi(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}C(\mathbf{r}+\mathbf{R}/2)C(\mathbf{r}-\mathbf{R}/2)$

with $C(\mathbf{r}) = N(k)\,{}_1F_1(i/k, 1, i(kr-\mathbf{k}\cdot\mathbf{r}))$

$\rightarrow$ Reduced and smoothed phase jump

M.F. Ciappina, C.C. Chirilă, M.L.,
PRA **75**, 043405 (2007)

SFA revisited

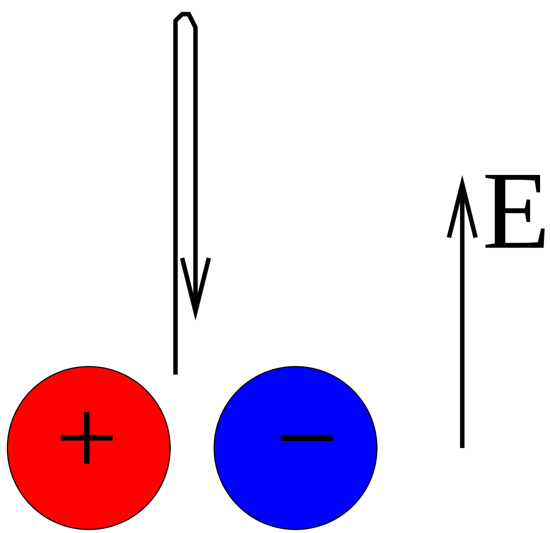
Different versions of SFA (all using plane waves)

- Full SFA:
  includes trajectories not parallel to driving field.
- SFA in saddle-point approximation for momentum (SP):
  only trajectories parallel to field,
  all ionization times contribute to one harmonic.
  ("conventional SFA")
- SFA in saddle point approximation for momentum + time:
  harmonic order determines ionization time and recollision momentum.

SFA revisited

Problems of conventional SFA

1) No harmonics when field $||$ nodal plane in antisymmetrical orbital



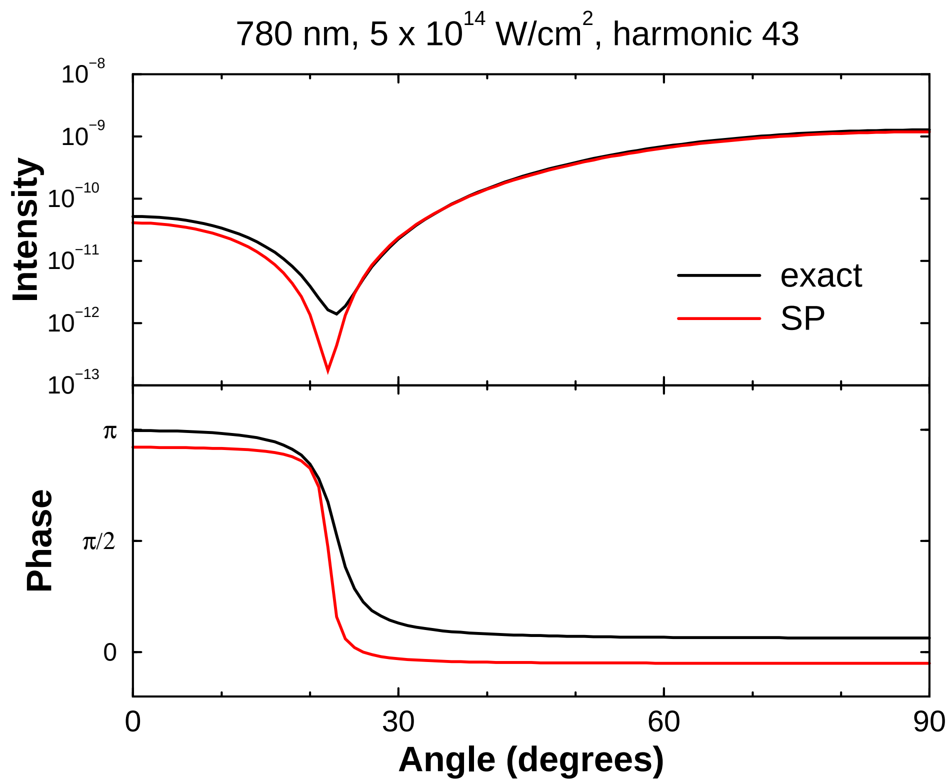
$$\langle\Psi|Ex|e^{ikx}\rangle = 0.$$

2) In velocity form no harmonics polarized $\perp$ laser field (for any orbital)

$$\langle\Psi|\partial_y|e^{ikx}\rangle = 0$$

Effect of non-saddle point dynamics

Comparison of full and saddle-point SFA

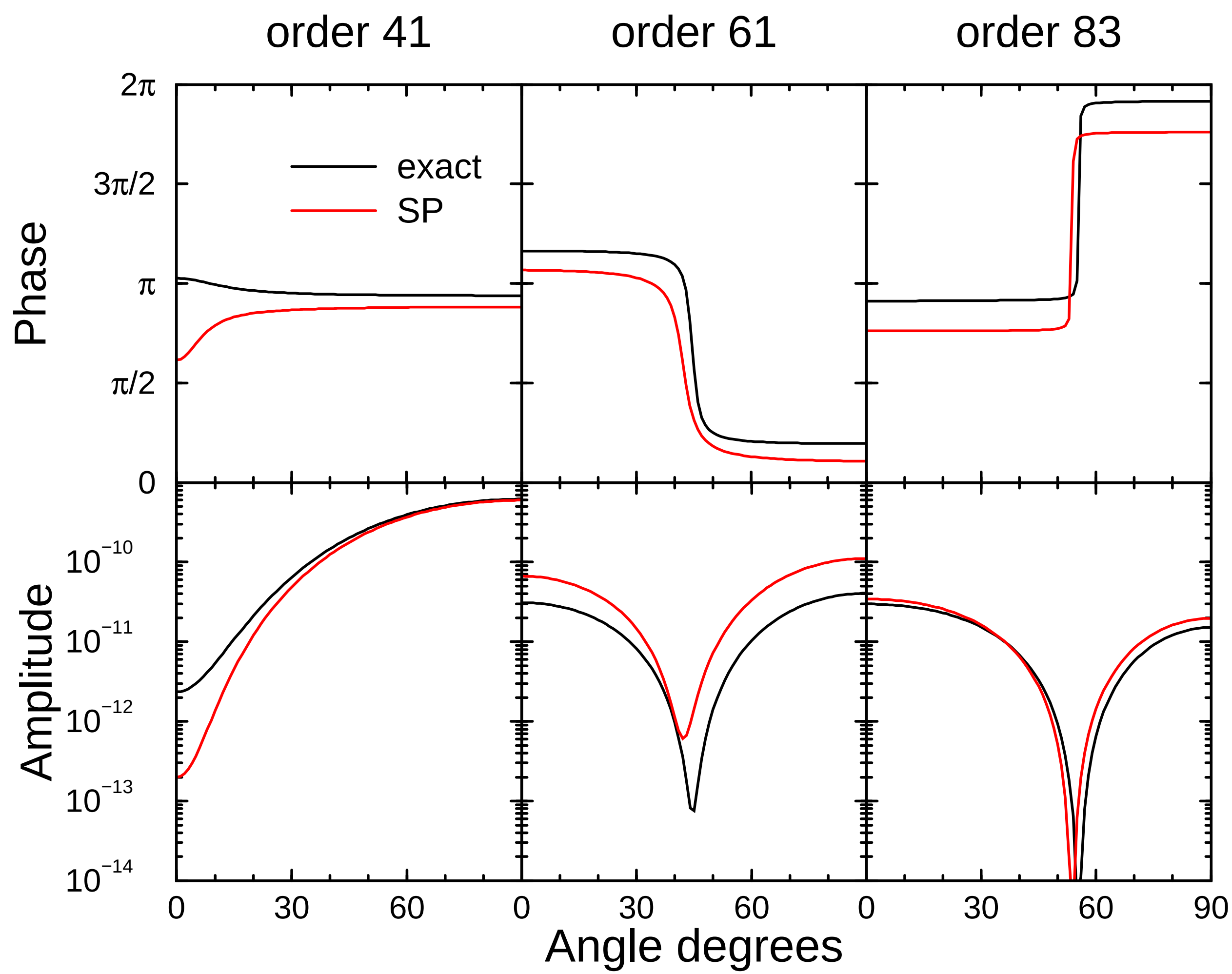


Phase jump is

- smooth in both calculations
- smoother in the full SFA (including nonparallel trajectories).

Effect of non-saddle-point dynamics

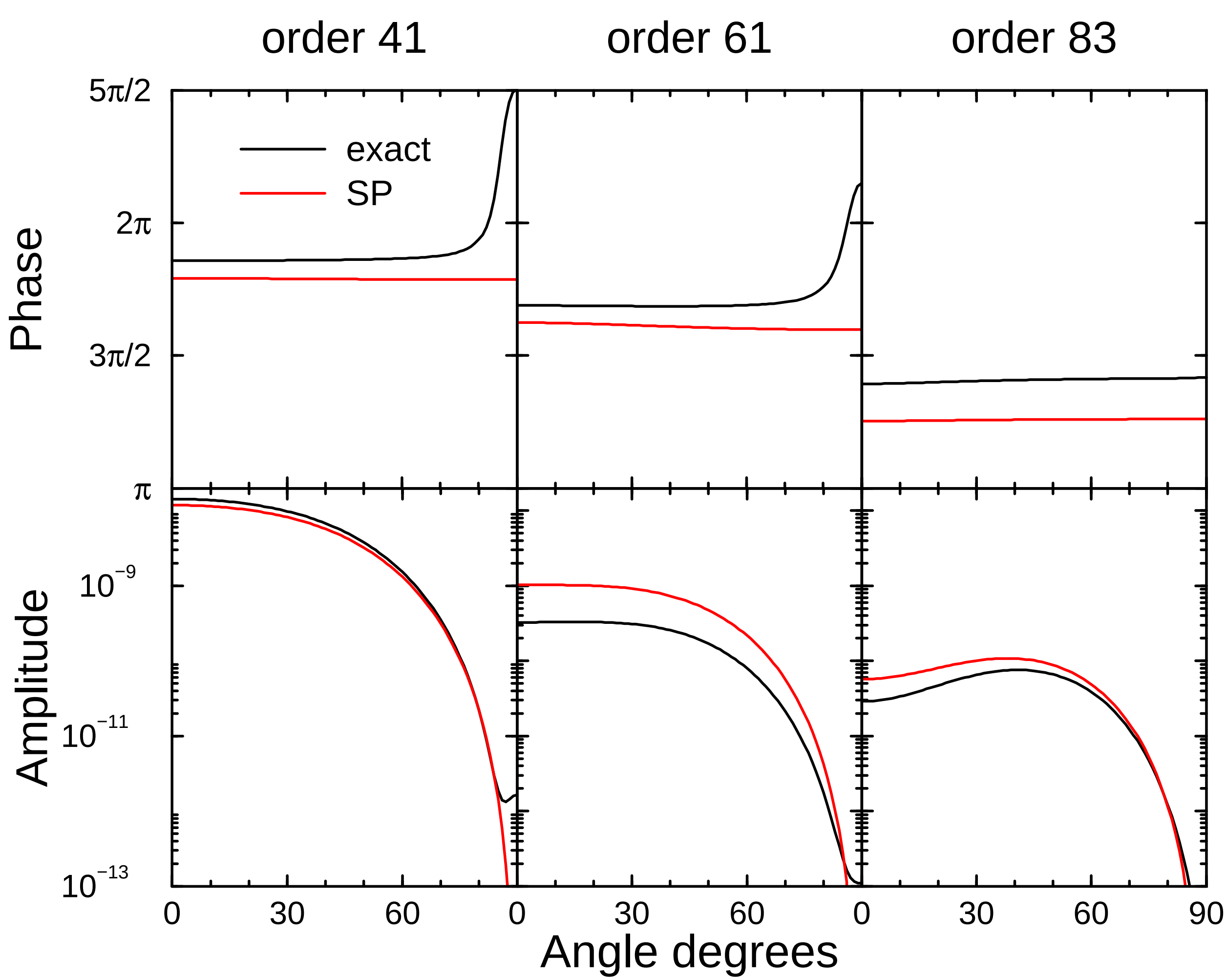
Orientation dependence for H_2^+ gerade ground state



Effect of non-saddle-point dynamics

Orientation dependence for H_2^+ **ungerade** 1st excited state

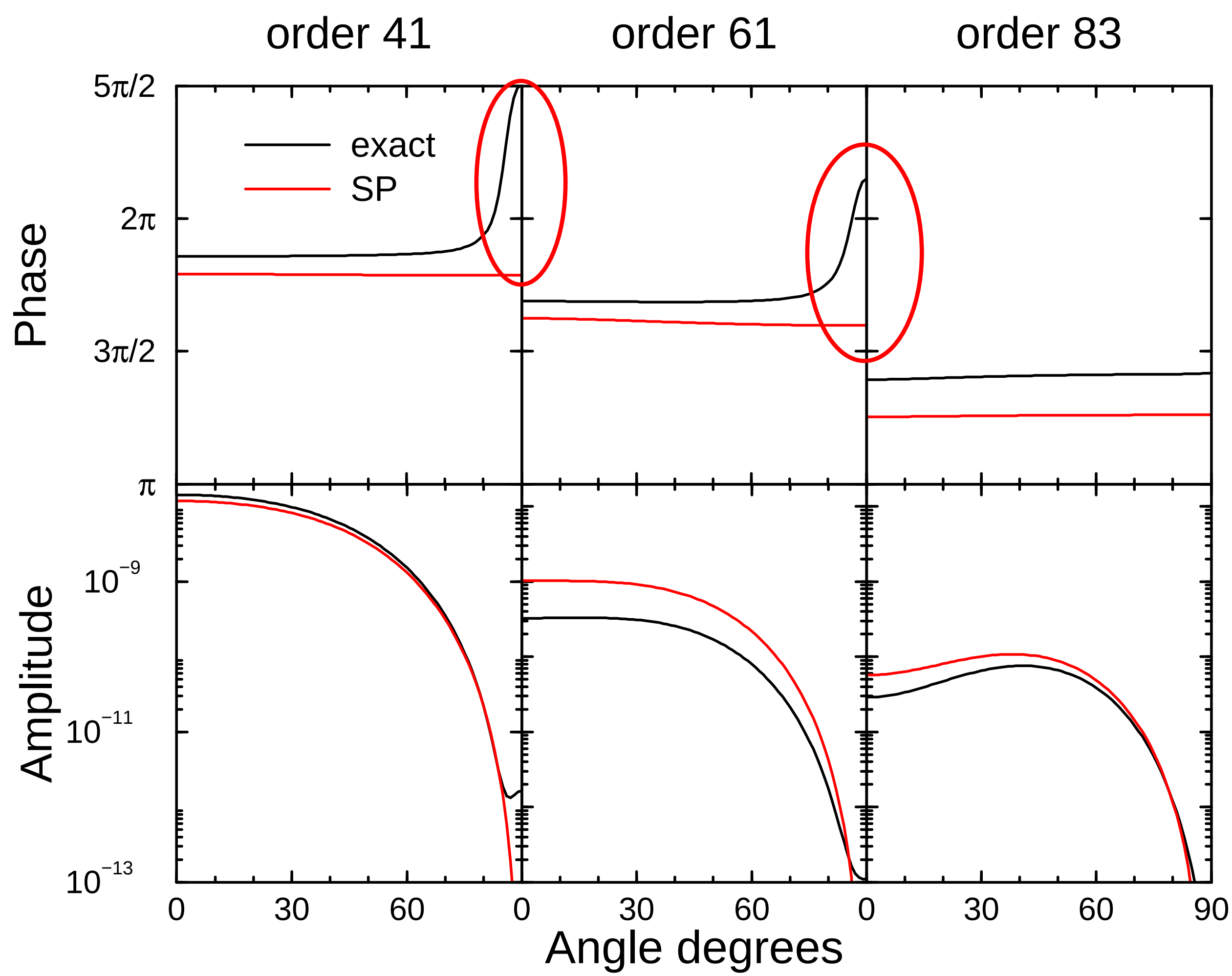
(adjusted to same $I_{\rm p}$)



Effect of non-saddle-point dynamics

Orientation dependence for H_2^+ **ungerade** 1st excited state

(adjusted to same I_p)

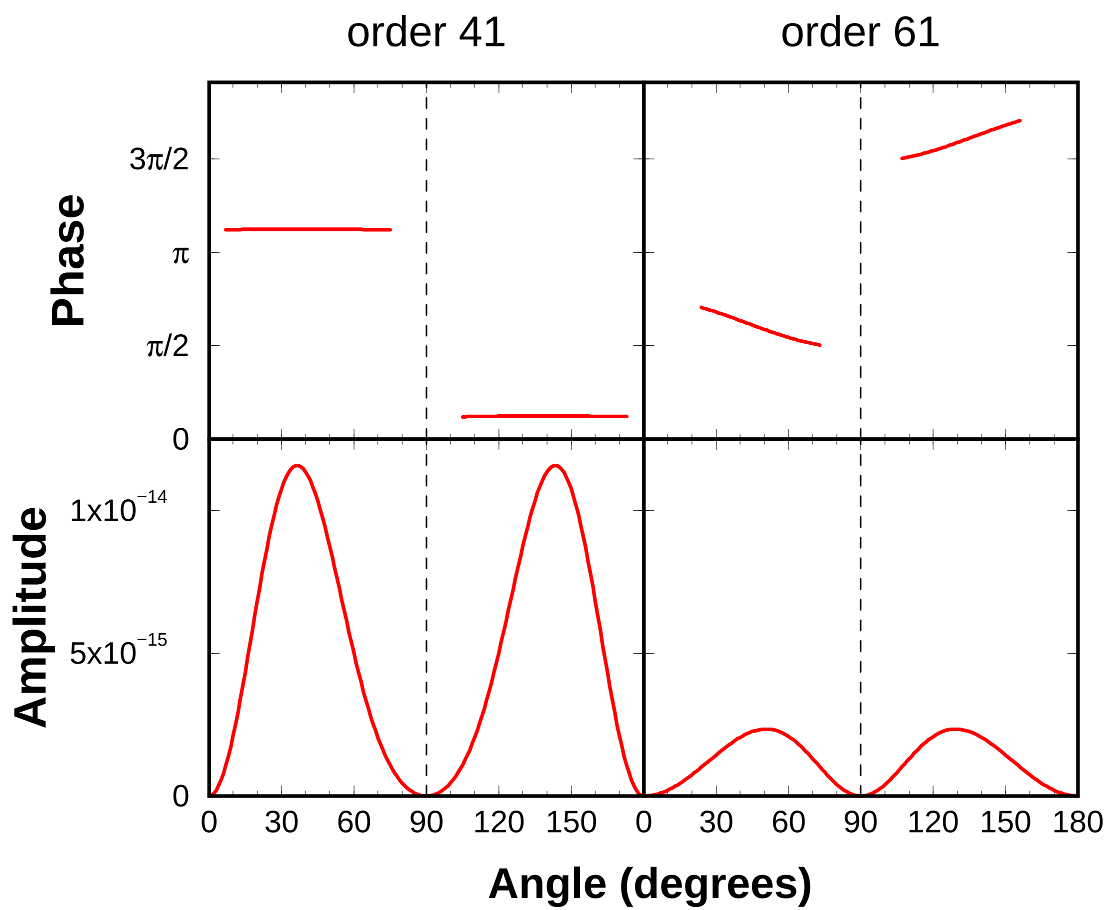


$\rightarrow$ Saddle-point SFA fails near 90^o.

Effect of non-saddle-point dynamics

Orientation dependence of **perpendicularly polarized harmonics**

for H_2^+ gerade ground state



- Small intensity
- π difference due to symmetry

Conclusions

- Orbital tomography of asymmetric molecules requires special pulses.
- Phase jumps in orientation dependence of HHG:
  - smoothness explained by non-saddle-point dynamics,
  - size of jump deviates from π due to Coulomb correction.