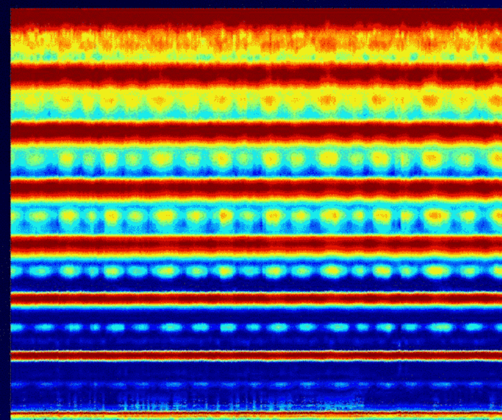
Measurement of attosecond pulses from aligned molecules



Hamed Merdji
SPAM
CEA Saclay

TUIXS

XTRA Network

Attosecond Science: Status and prospects
Aug. 01 - Aug. 04 2006

Our group:

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Collaborations

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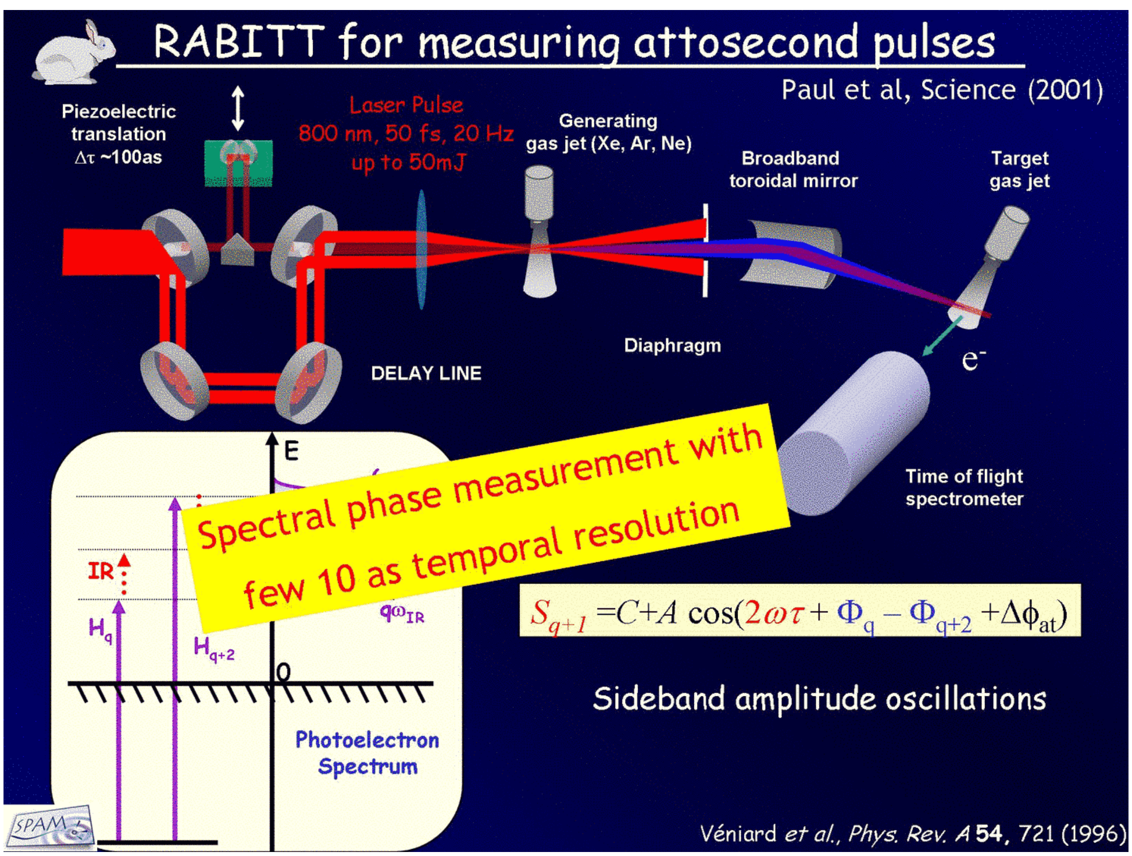
Outline

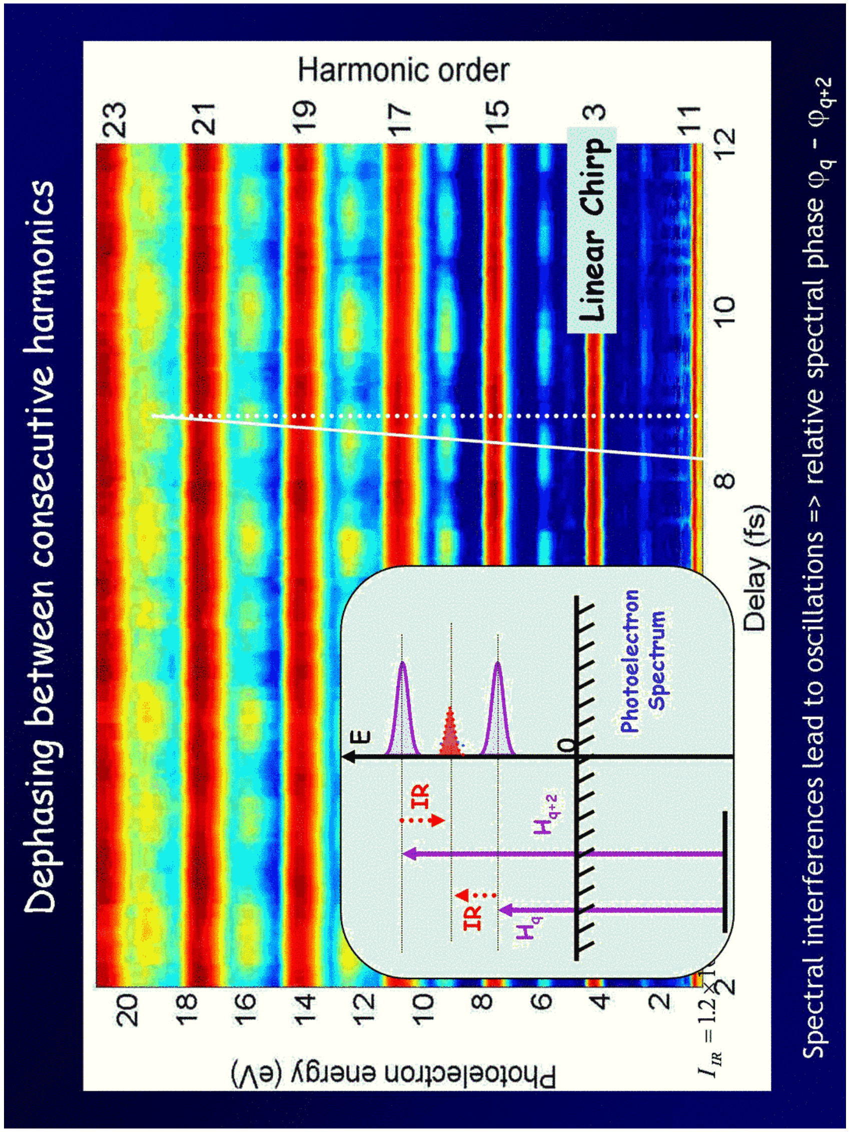
1 - Optimization of attosecond pulses in atoms

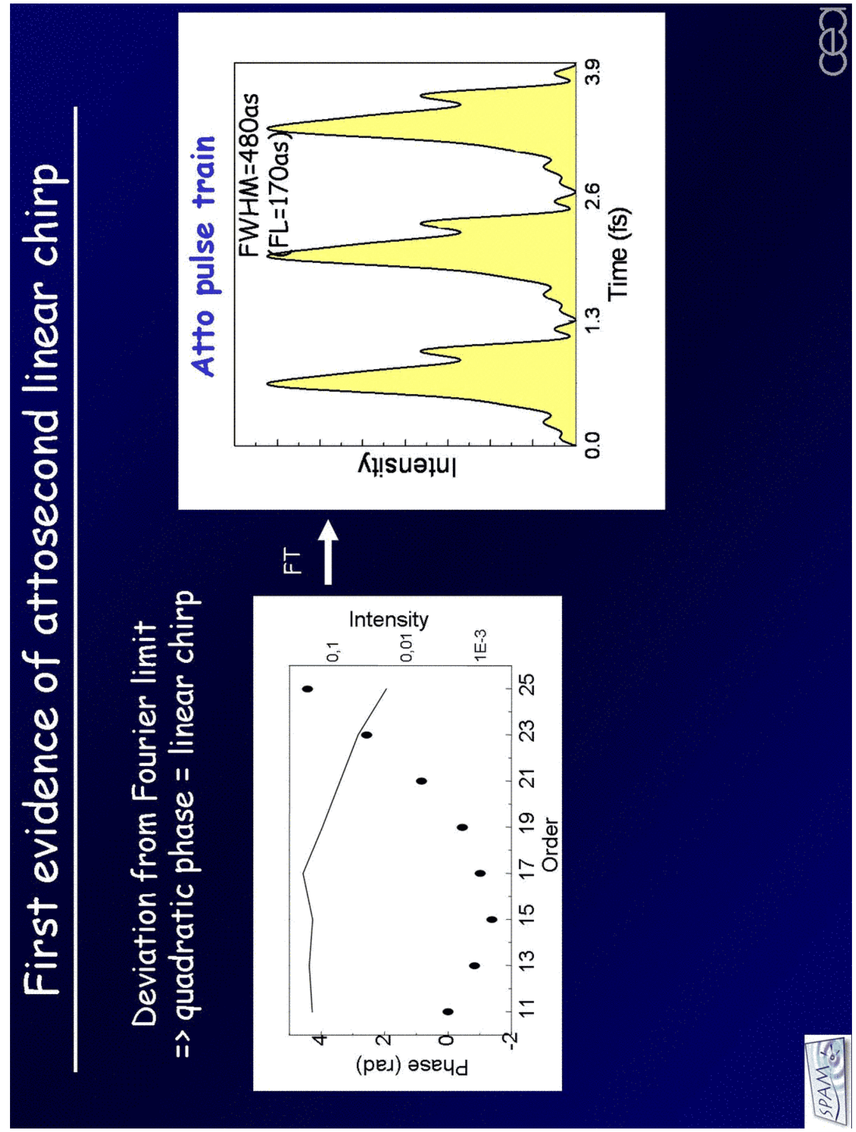
=> Broadband RABITT technique : amplitude and phase measurement of the harmonic emission

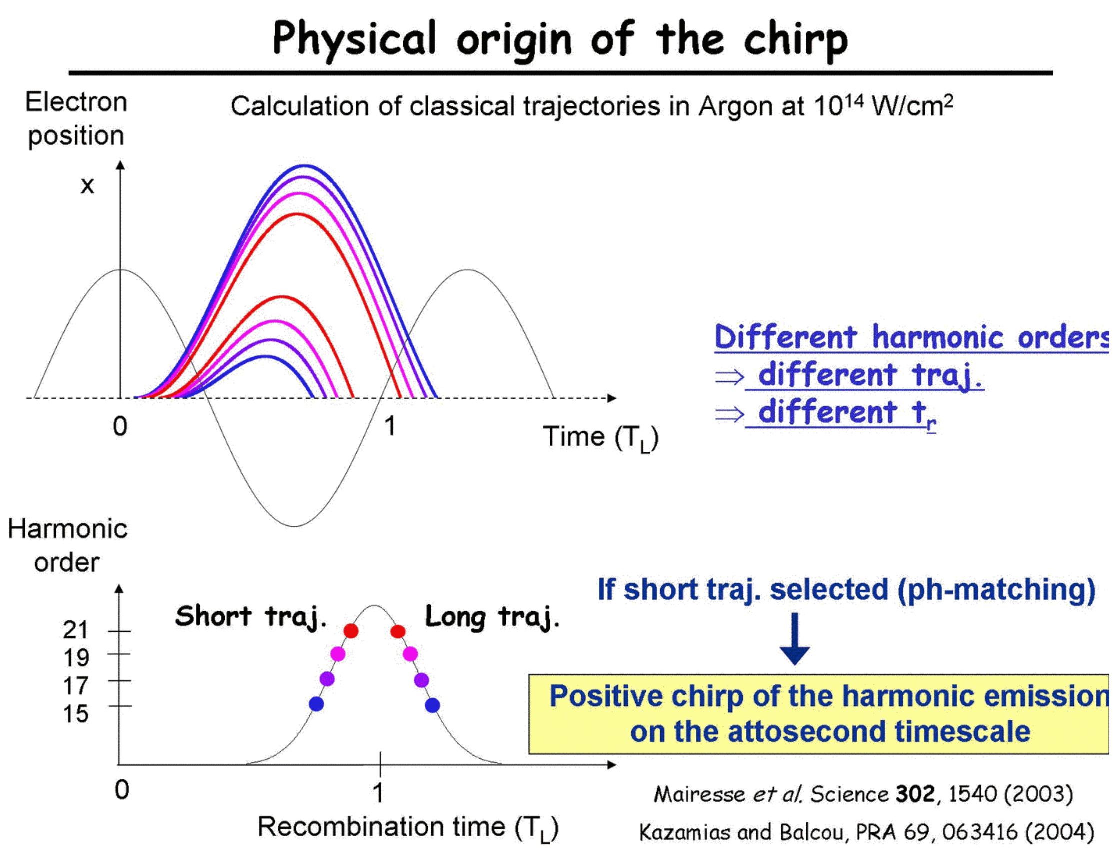
2 - RABITT measurements in molecules

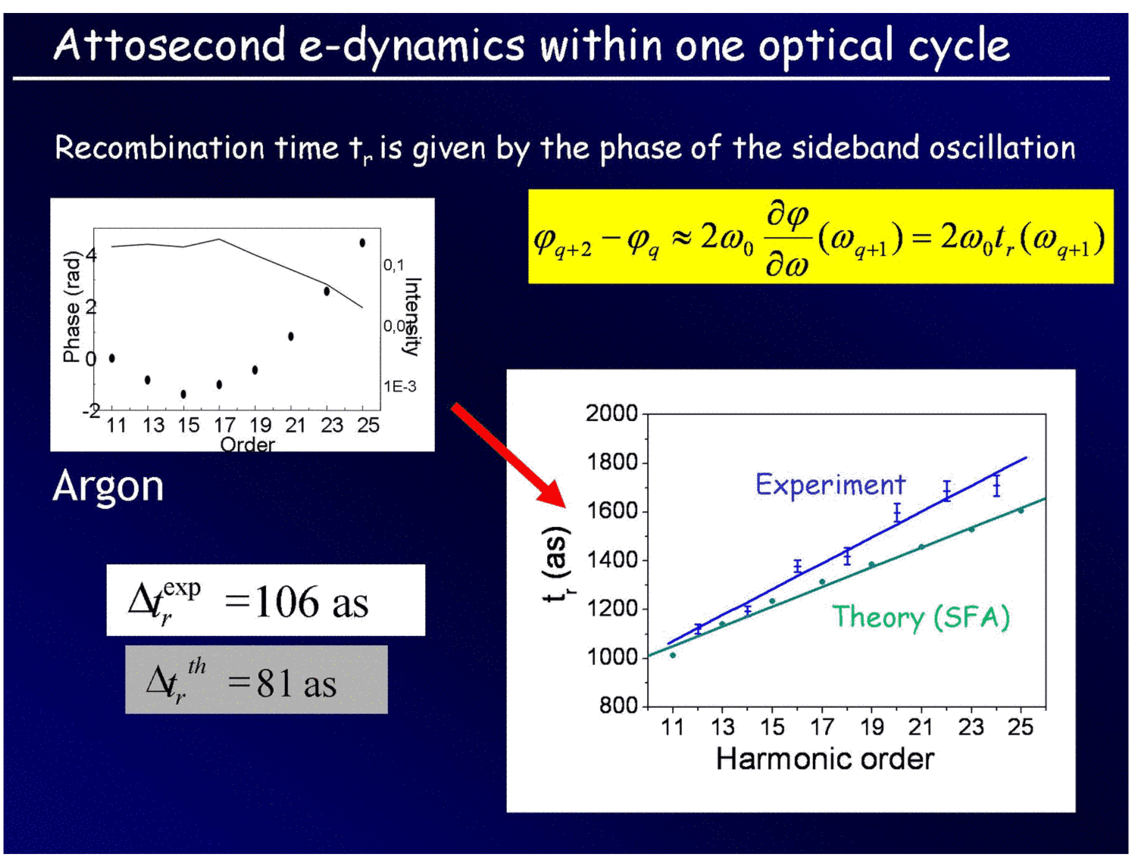
=> surprising features not predicted by models !

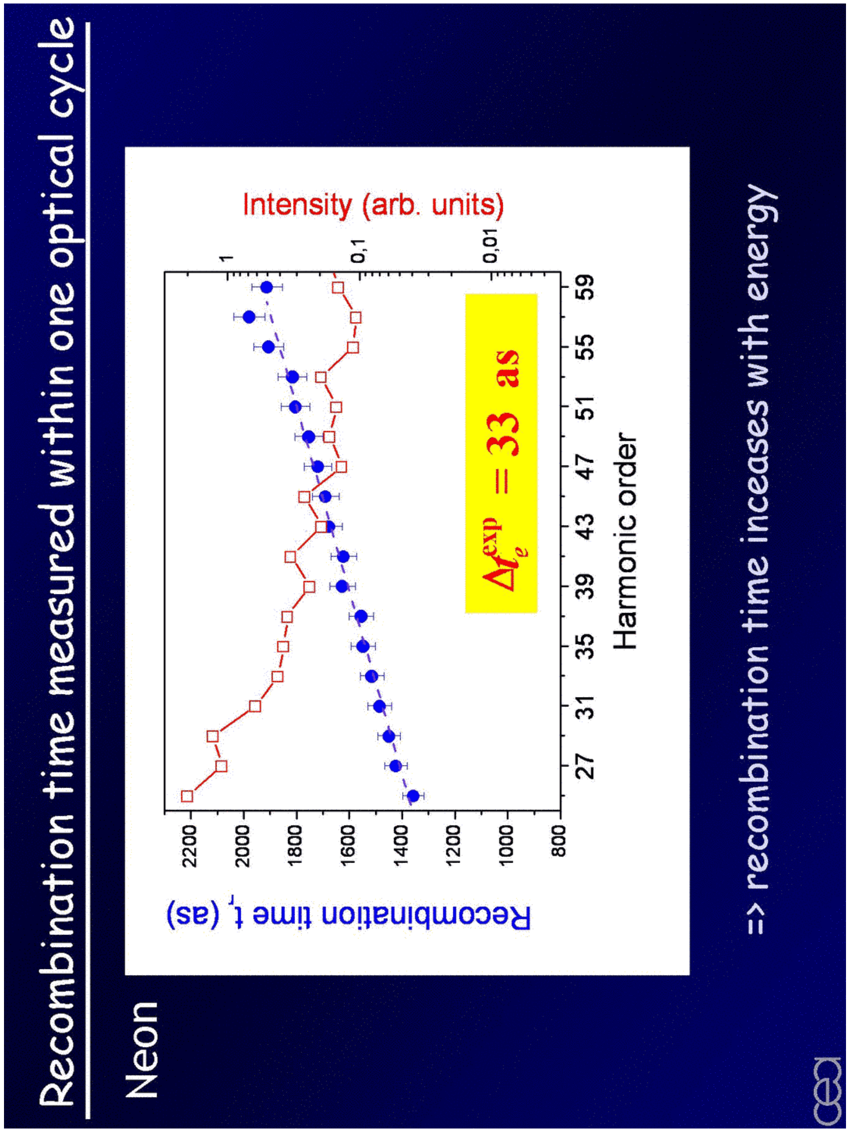


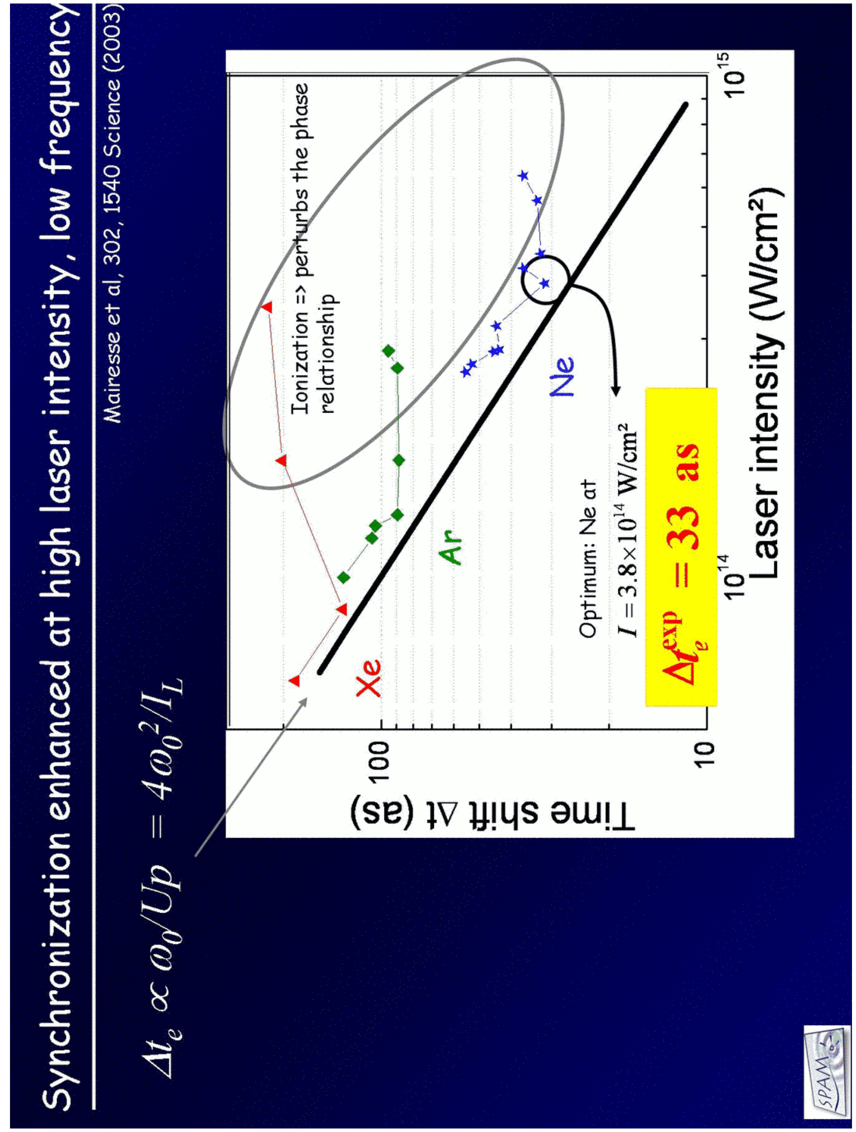

Dephasing between consecutive harmonics
Harmonic order
23
21
19
17
15
13
11
Linear Chirp
Photoelectron energy (eV)
20
18
16
14
12
10
8
6
4
2
Delay (fs)
8
10
12
E
0
IR
IR
H_q
H_{q+2}
Photoelectron Spectrum
Spectral interferences lead to oscillations => relative spectral phase $\varphi_q - \varphi_{q+2}$

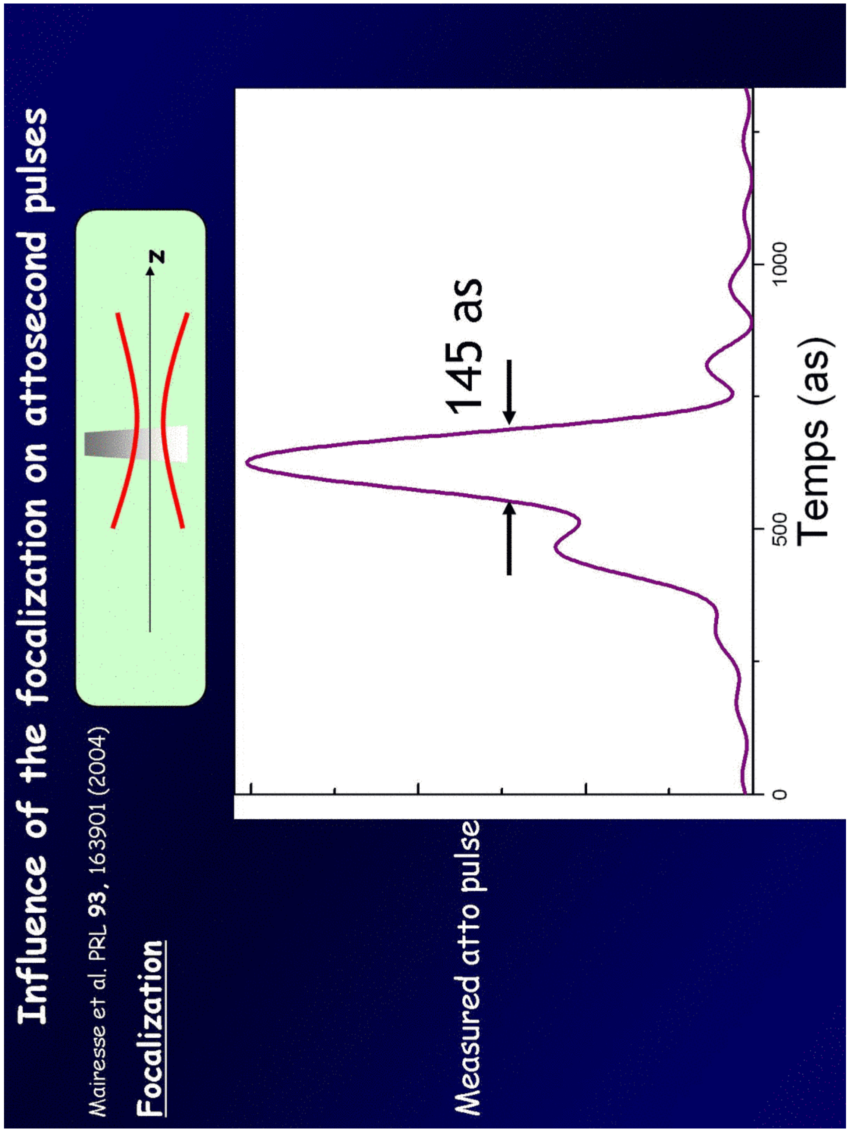

First evidence of attosecond linear chirp
Deviation from Fourier limit
=> quadratic phase = linear chirp
FT
Atto pulse train
FWHM=480as
(FL=170as)
Intensity
Time (fs)
0.0
1.3
2.6
3.9
Phase (rad)
Intensity
0,1
0,01
1E-3
Order
11
13
15
17
19
21
23
25

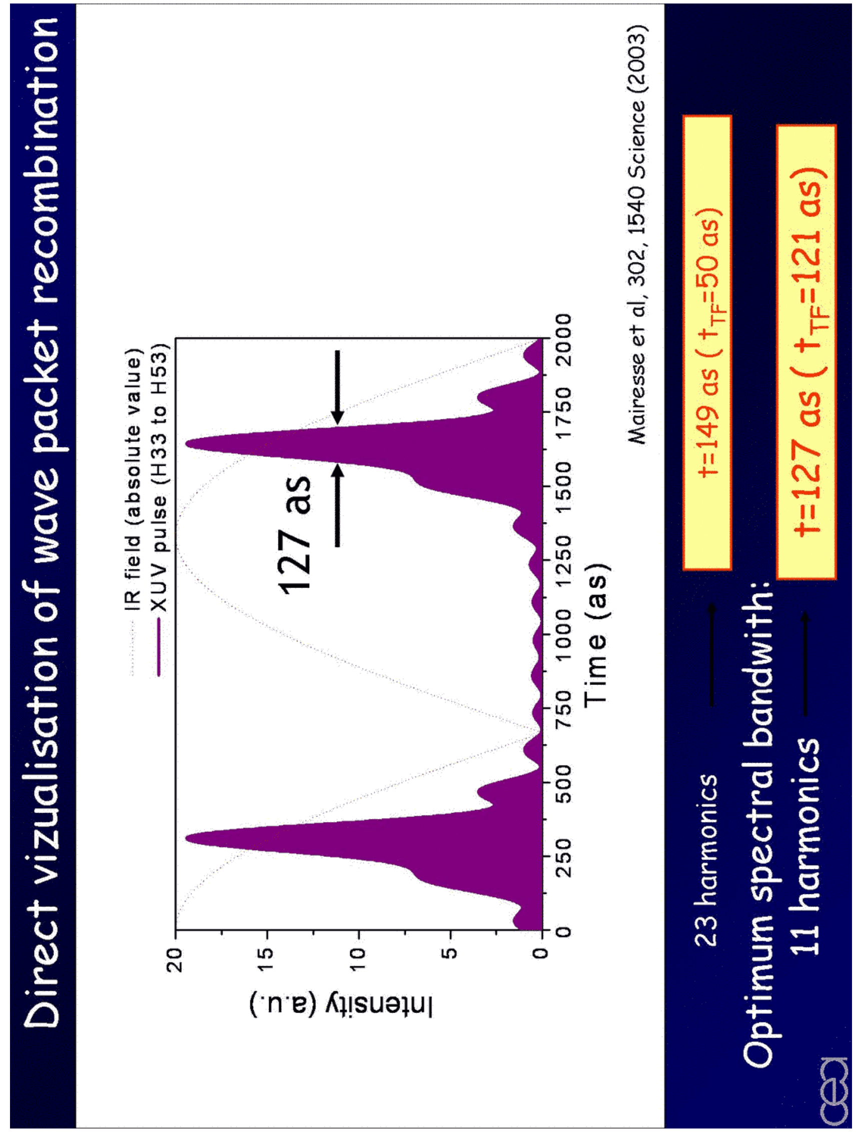

Physical origin of the chirp
Calculation of classical trajectories in Argon at 10^{14} W/cm^2
Electron position
x
0
1
Time (T_L)
Different harmonic orders
⇒ different traj.
⇒ different t_r
Harmonic order
21
19
17
15
Short traj.
Long traj.
0
1
Recombination time (T_L)
If short traj. selected (ph-matching)
Positive chirp of the harmonic emission on the attosecond timescale
Mairesse *et al.* Science **302**, 1540 (2003)
Kazamias and Balcou, PRA 69, 063416 (2004)

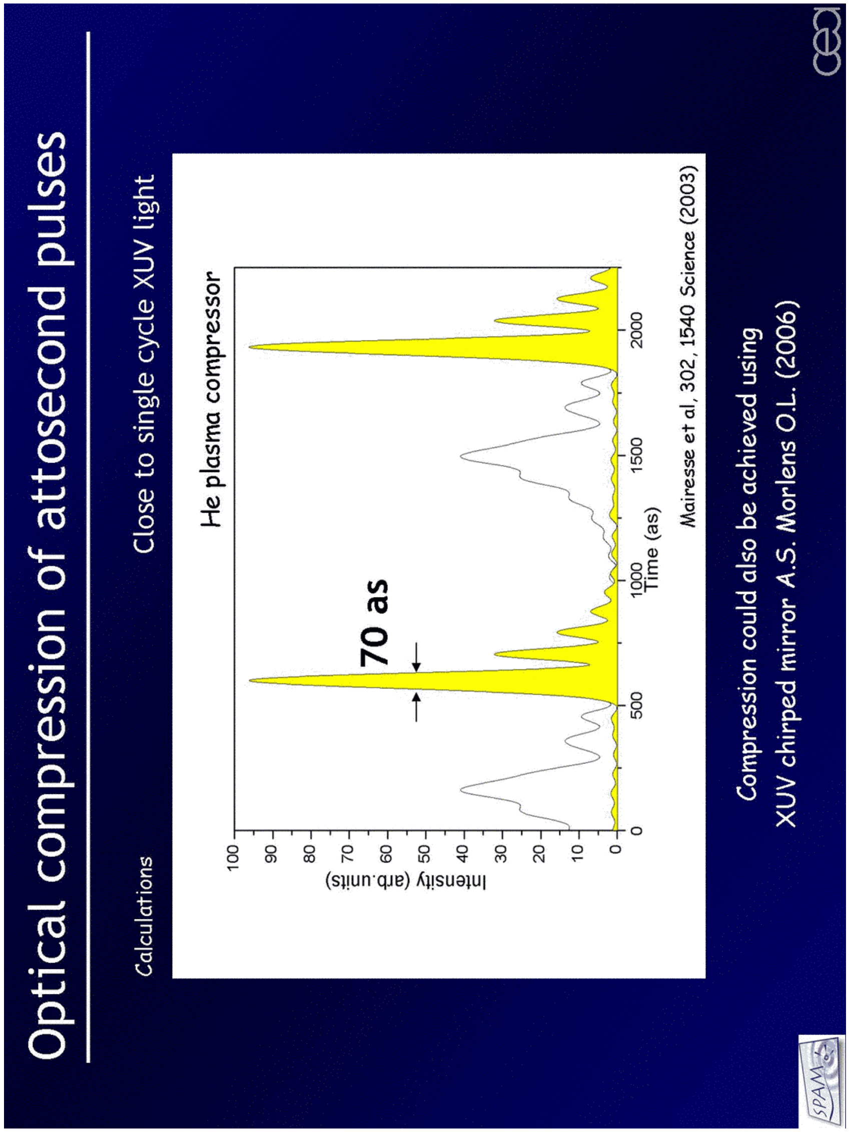

Attosecond e-dynamics within one optical cycle
Recombination time t_r is given by the phase of the sideband oscillation
$\varphi_{q+2} - \varphi_q \approx 2\omega_0 \frac{\partial \varphi}{\partial \omega}(\omega_{q+1}) = 2\omega_0 t_r(\omega_{q+1})$
Phase (rad)
Intensity
Order
Argon
$\Delta t_r^{exp} = 106$ as
$\Delta t_r^{th} = 81$ as
t_r (as)
Experiment
Theory (SFA)
Harmonic order

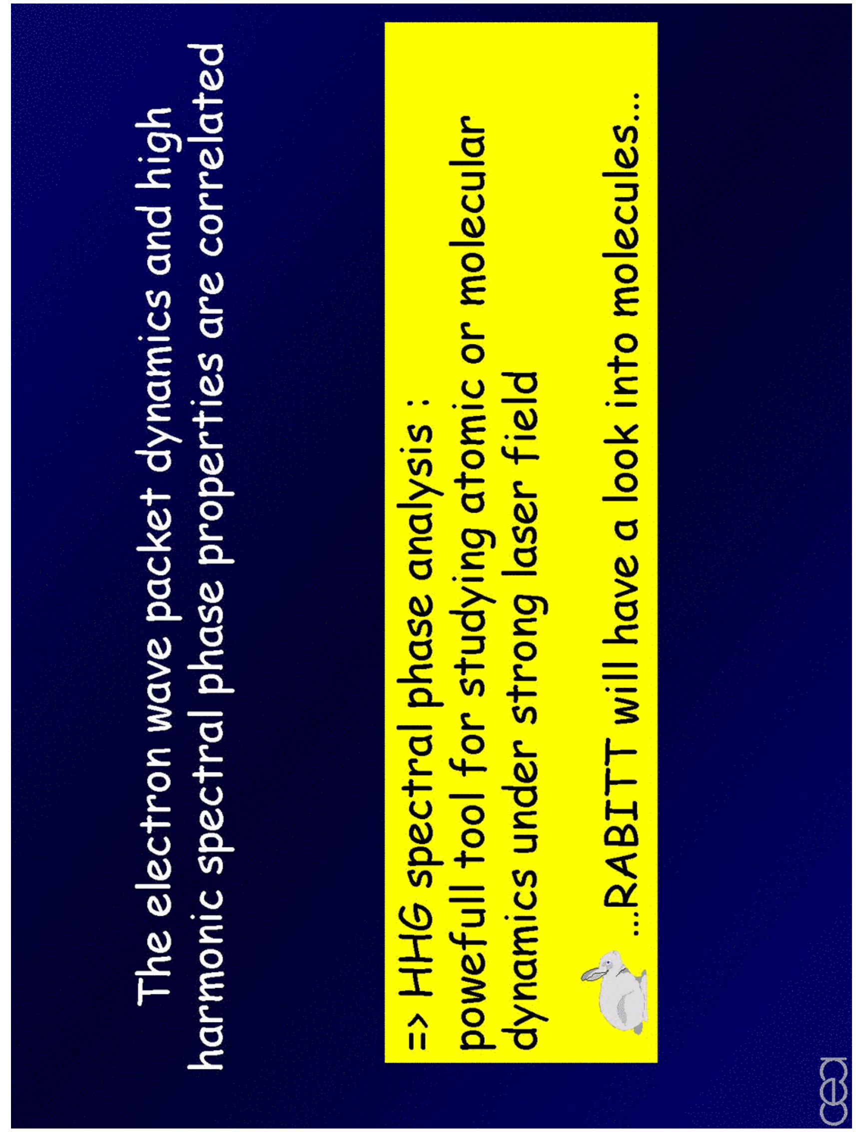

Recombination time measured within one optical cycle
Neon
Recombination time t_r (as)
Intensity (arb. units)
Harmonic order
$\Delta t_e^{exp} = 33$ as
=> recombination time inceases with energy

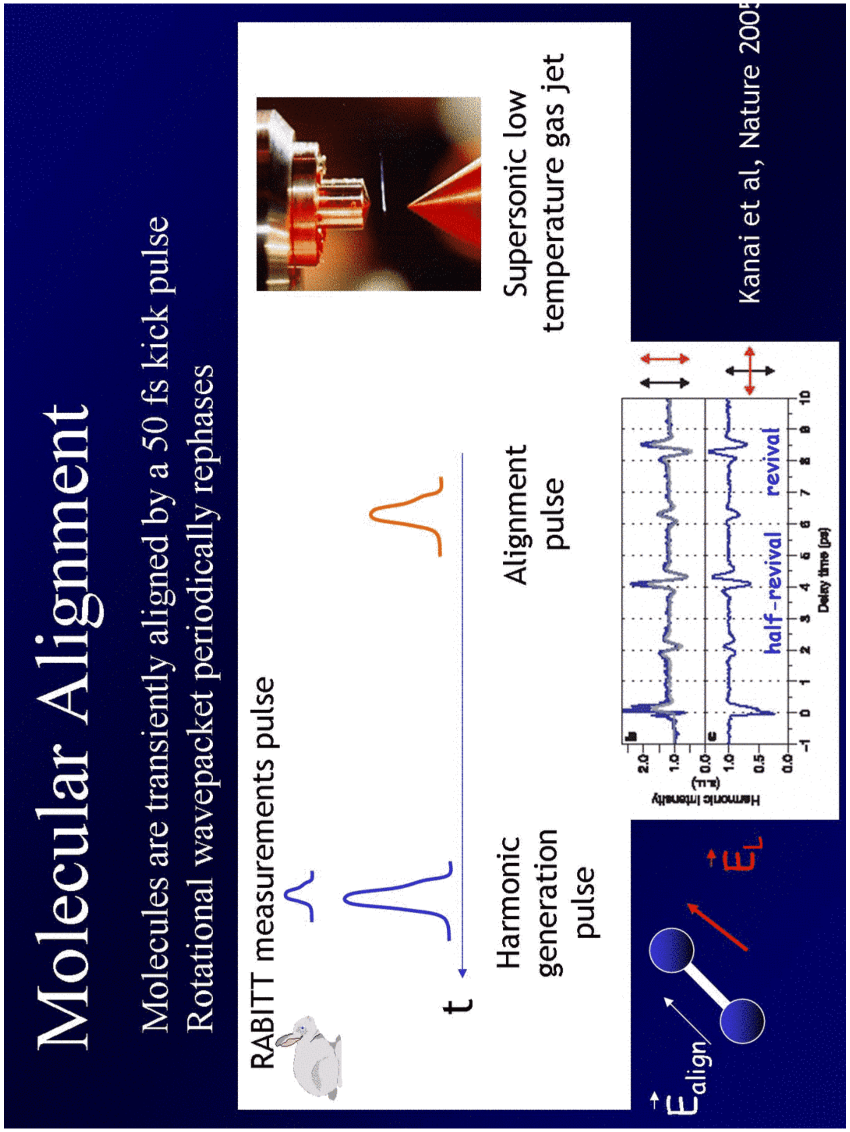

Synchronization enhanced at high laser intensity, low frequency
Mairesse et al, 302, 1540 Science (2003)
$\Delta t_e \propto \omega_0/Up = 4\omega_0^2/I_L$
Time shift Δt (as)
Laser intensity (W/cm²)
Ionization => perturbs the phase relationship
Xe
Ar
Ne
Optimum: Ne at
$I = 3.8\times10^{14}$ W/cm²
$\Delta t_e^{exp} = 33$ as

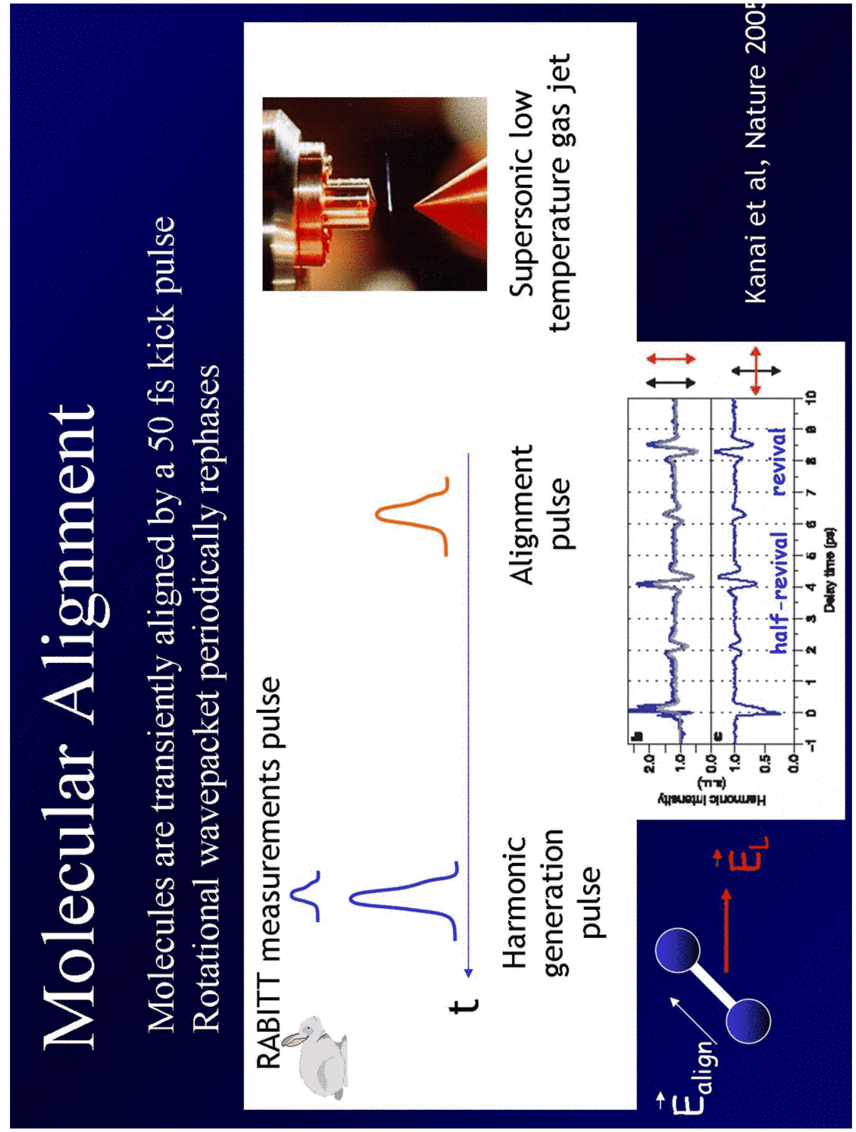

Influence of the focalization on attosecond pulses
Mairesse et al. PRL 93, 163901 (2004)
Focalization
z
Measured atto pulse
145 as
0
500
1000
Temps (as)

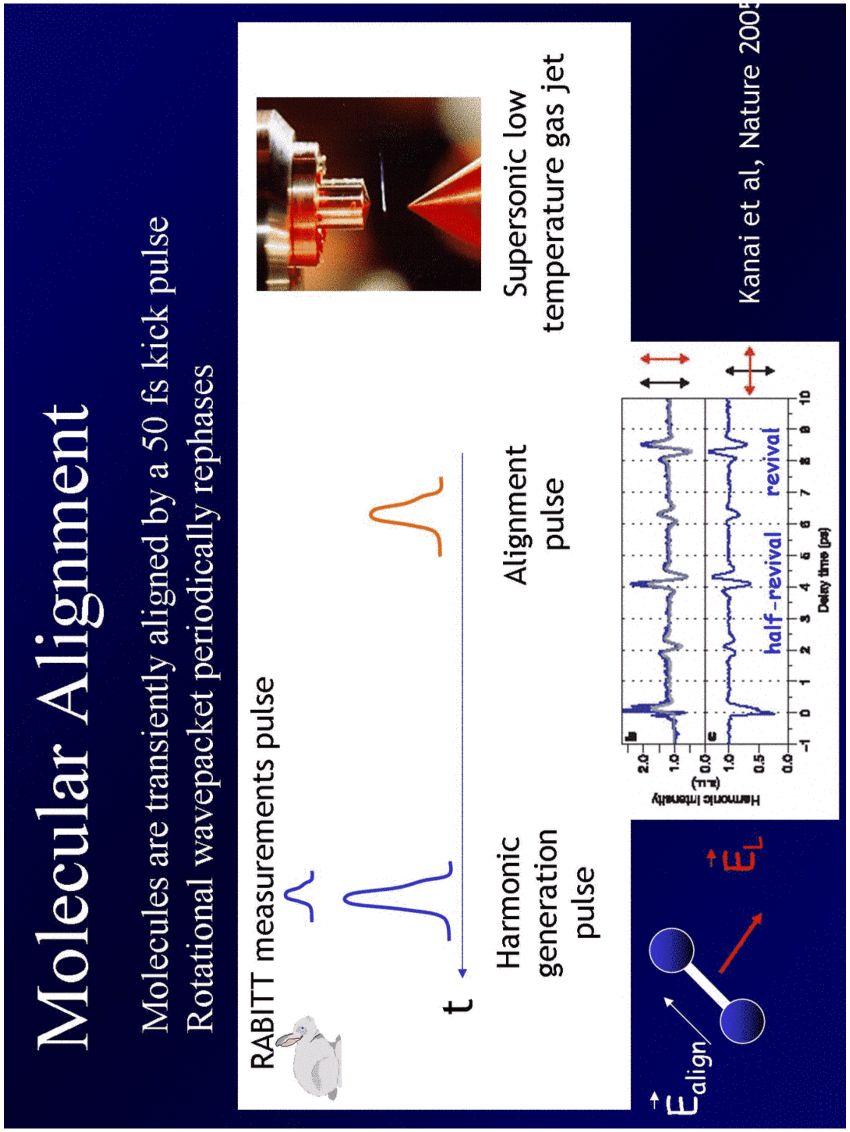

Direct vizualisation of wave packet recombination
IR field (absolute value)
XUV pulse (H33 to H53)
127 as
Intensity (a.u.)
0
5
10
15
20
0
250
500
750
1000
1250
1500
1750
2000
Time (as)
Mairesse et al, 302, 1540 Science (2003)
23 harmonics
t=149 as (t_{TF}=50 as)
Optimum spectral bandwith:
11 harmonics
t=127 as (t_{TF}=121 as)

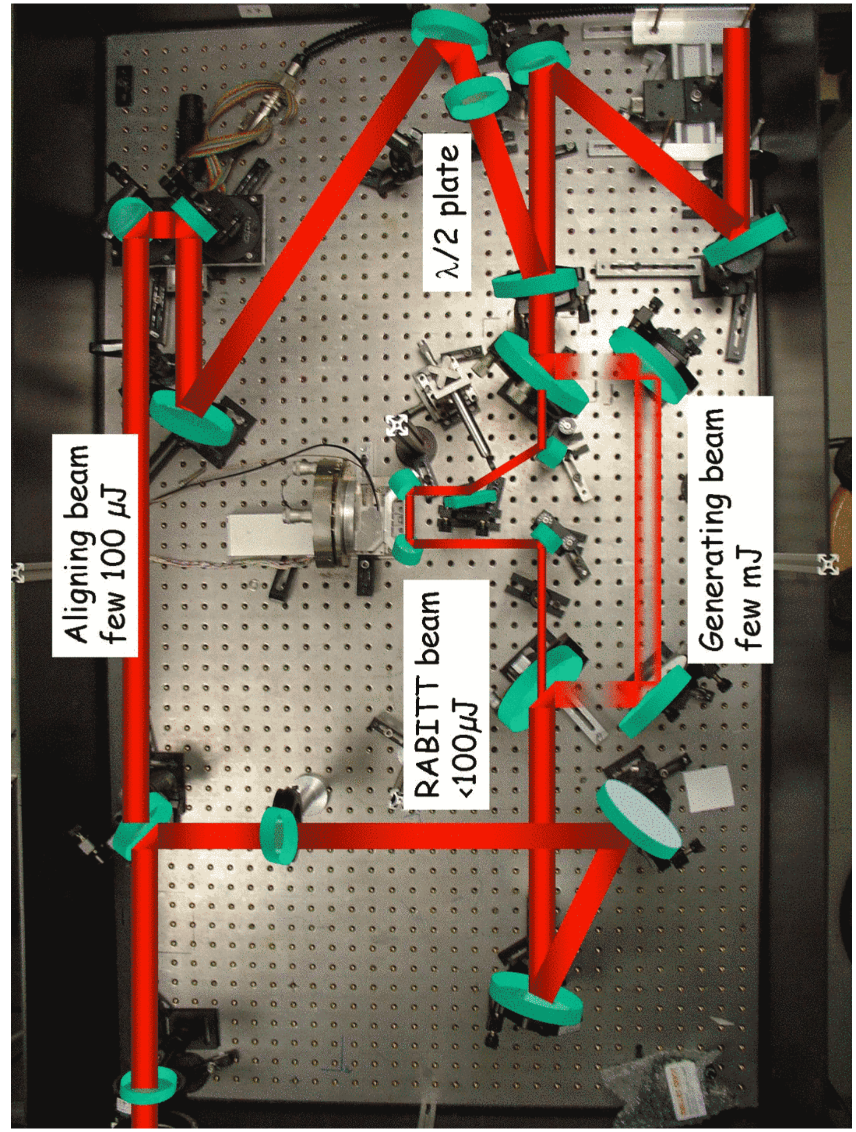

Optical compression of attosecond pulses
Calculations
Close to single cycle XUV light
He plasma compressor
70 as
Intensity (arb.units)
0
10
20
30
40
50
60
70
80
90
100
0
500
1000
1500
2000
Time (as)
Mairesse et al, 302, 1540 Science (2003)
Compression could also be achieved using
XUV chirped mirror A.S. Morlens O.L. (2006)
SPAM

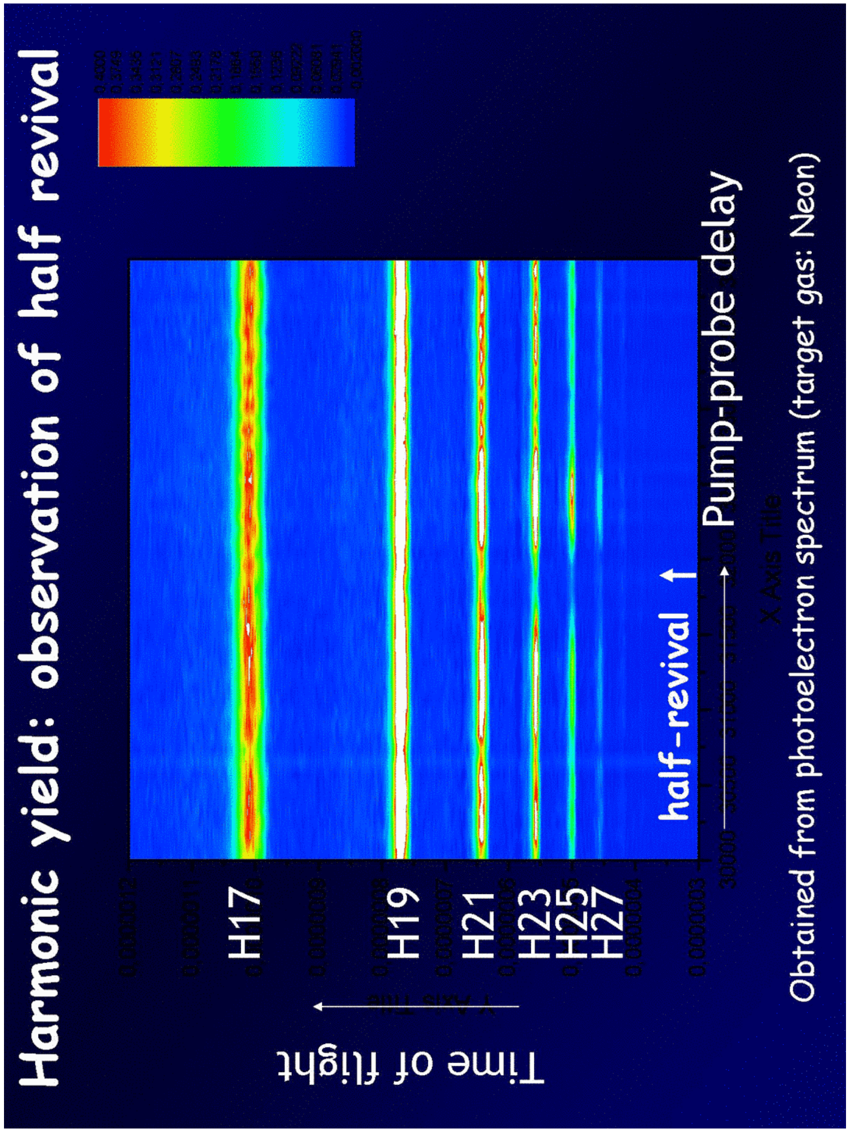

The electron wave packet dynamics and high harmonic spectral phase properties are correlated
=> HHG spectral phase analysis :
powefull tool for studying atomic or molecular dynamics under strong laser field
...RABITT will have a look into molecules...

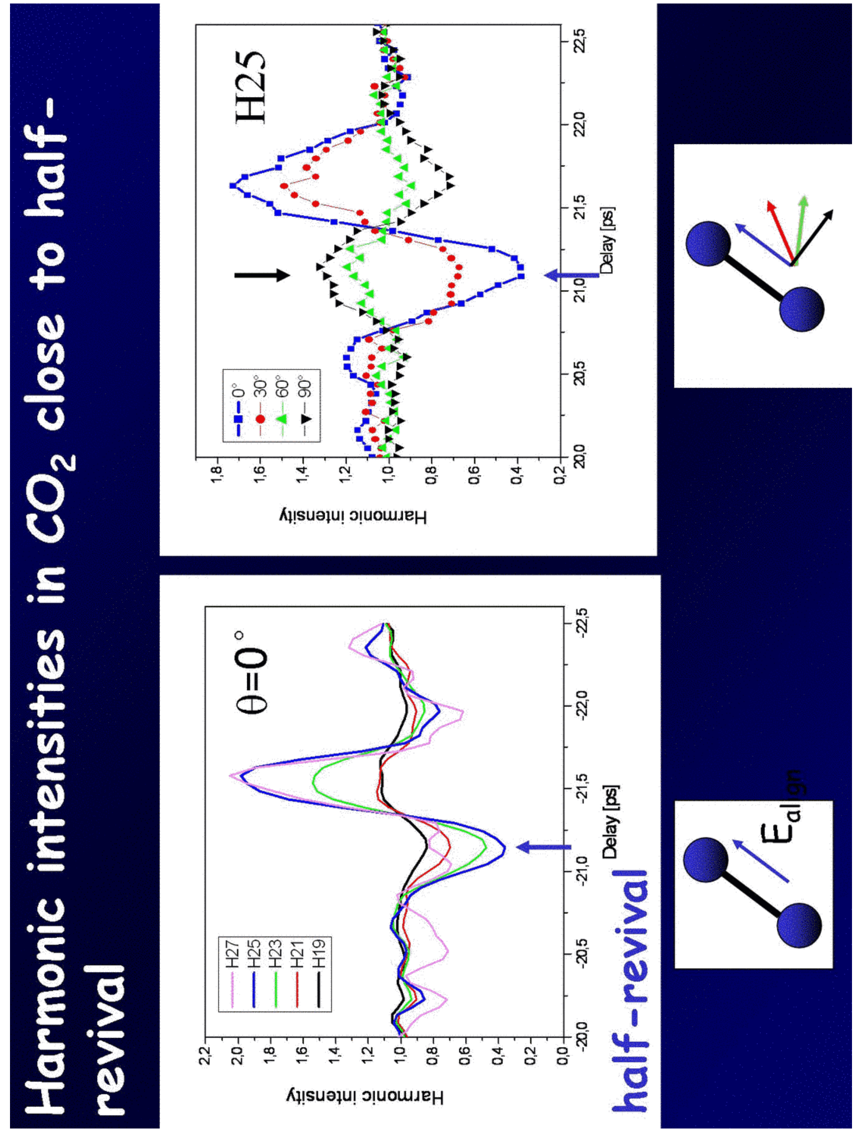

Molecular Alignment
Molecules are transiently aligned by a 50 fs kick pulse
Rotational wavepacket periodically rephases
RABITT measurements pulse
t
Harmonic generation pulse
Alignment pulse
Supersonic low temperature gas jet
Kanai et al, Nature 2005
half-revival
revival
Harmonic intensity (a.u.)
Delay time (ps)
$\vec{E}_{align}$
$\vec{E}_L$


Molecular Alignment
Molecules are transiently aligned by a 50 fs kick pulse
Rotational wavepacket periodically rephases
RABITT measurements pulse
t
Harmonic generation pulse
Alignment pulse
Supersonic low temperature gas jet
Kanai et al, Nature 2005
half-revival
revival
Harmonic intensity (a.u.)
Delay time (ps)
$\vec{E}_{align}$
$\vec{E}_L$


Molecular Alignment
Molecules are transiently aligned by a 50 fs kick pulse
Rotational wavepacket periodically rephases
RABITT measurements pulse
t
Harmonic generation pulse
Alignment pulse
Supersonic low temperature gas jet
Kanai et al, Nature 2005
half-revival
revival
Harmonic intensity (a.u.)
Delay time (ps)
$\vec{E}_{align}$
$\vec{E}_L$


λ/2 plate
Aligning beam few 100 µJ
RABITT beam <100µJ
Generating beam few mJ


Harmonic yield: observation of half revival
Time of flight
H17
H19
H21
H23
H25
H27
half-revival
Pump-probe delay
Obtained from photoelectron spectrum (target gas: Neon)

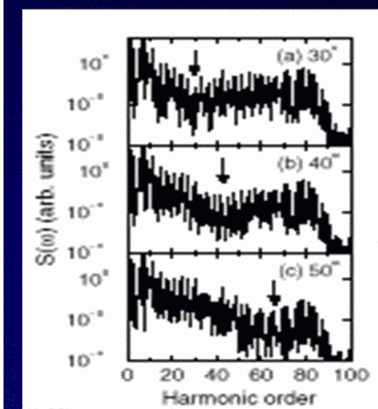

Harmonic intensities in CO_2 close to half-revival
θ=0°
H27
H25
H23
H21
H19
Harmonic intensity
Delay [ps]
half-revival
H25
0°
30°
60°
90°
E_{align}

What about the phase ?

=> crucial for attosecond physics in molecules and applications such as tomographic imaging of molecular orbitals

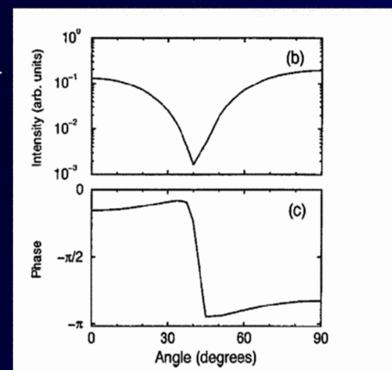
2D calculations of H_2^+

M. Lein et al. PRL 2002



Minimum of emission for a specific harmonic order due to interference effects

Phase jump of about π at critical angle



RABITT measurements in molecules

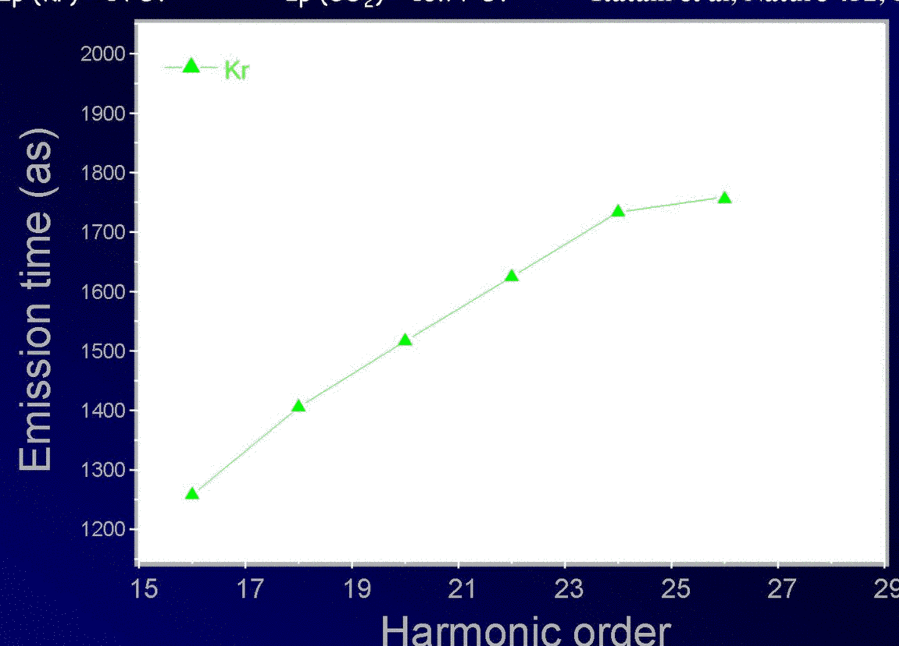
Harmonic emission times in CO_2

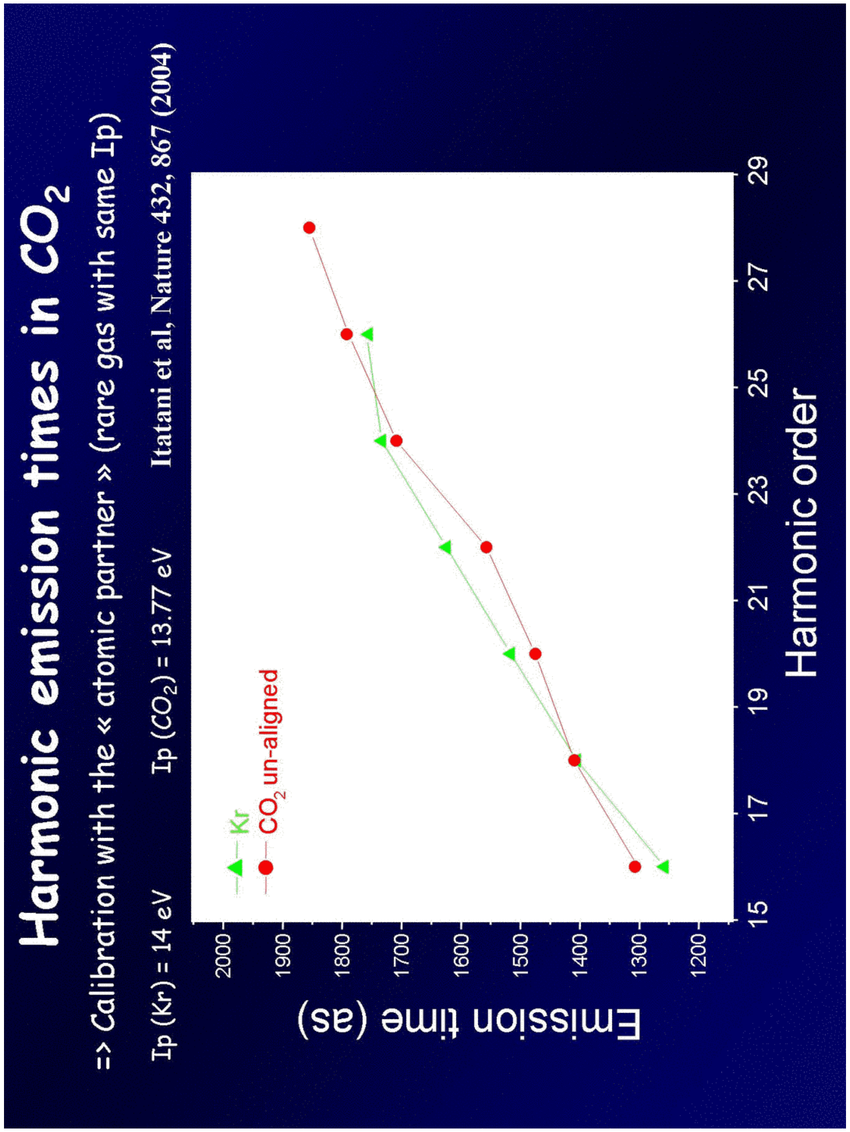
=> Calibration with the « atomic partner » (rare gas with same Ip)

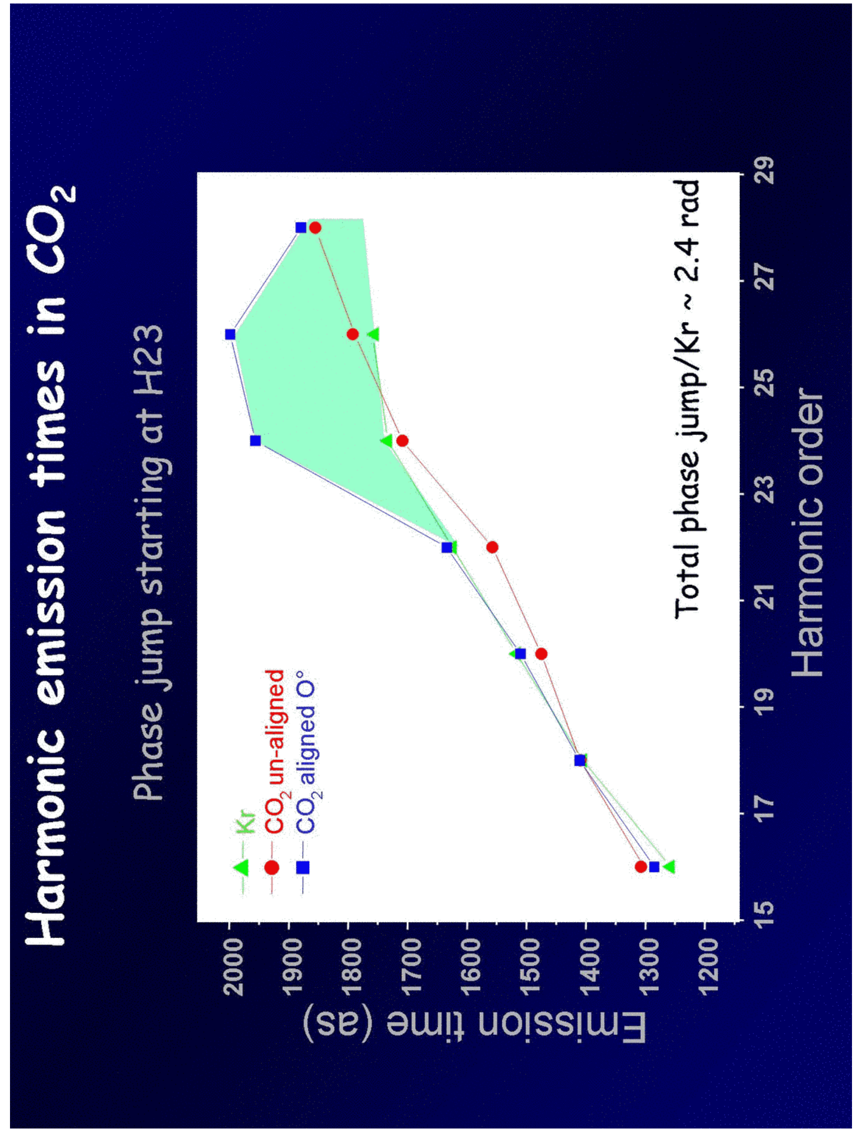
Ip (Kr) = 14 eV

Ip (CO_2) = 13.77 eV

Itatani et al, Nature 432, 867 (2004)




Harmonic emission times in CO_2
=> Calibration with the « atomic partner » (rare gas with same Ip)
Ip (Kr) = 14 eV
Ip (CO_2) = 13.77 eV
Itatani et al, Nature 432, 867 (2004)
Kr
CO_2 un-aligned
Emission time (as)
Harmonic order


Harmonic emission times in CO_2
Phase jump starting at H23
Kr
CO_2 un-aligned
CO_2 aligned O°
Total phase jump/Kr ~ 2.4 rad
Emission time (as)
Harmonic order

Results for CO2

Phase jump starting at H23
Amplitude minimum between H21 and H25
=>Good agreement with Kanai et al, Nature 2005

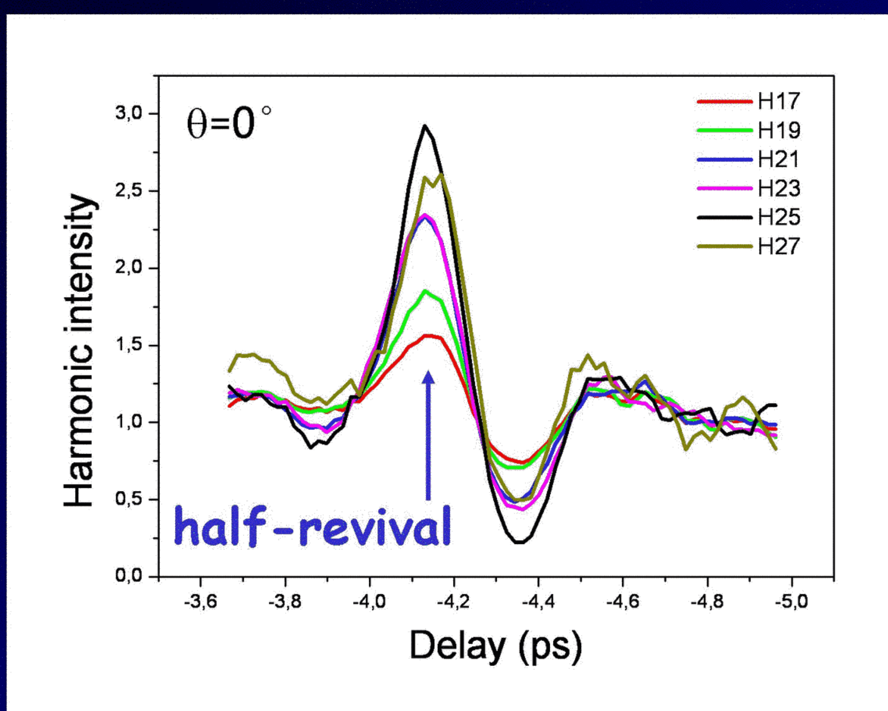
Phase jump and amplitude minimum vary with angle

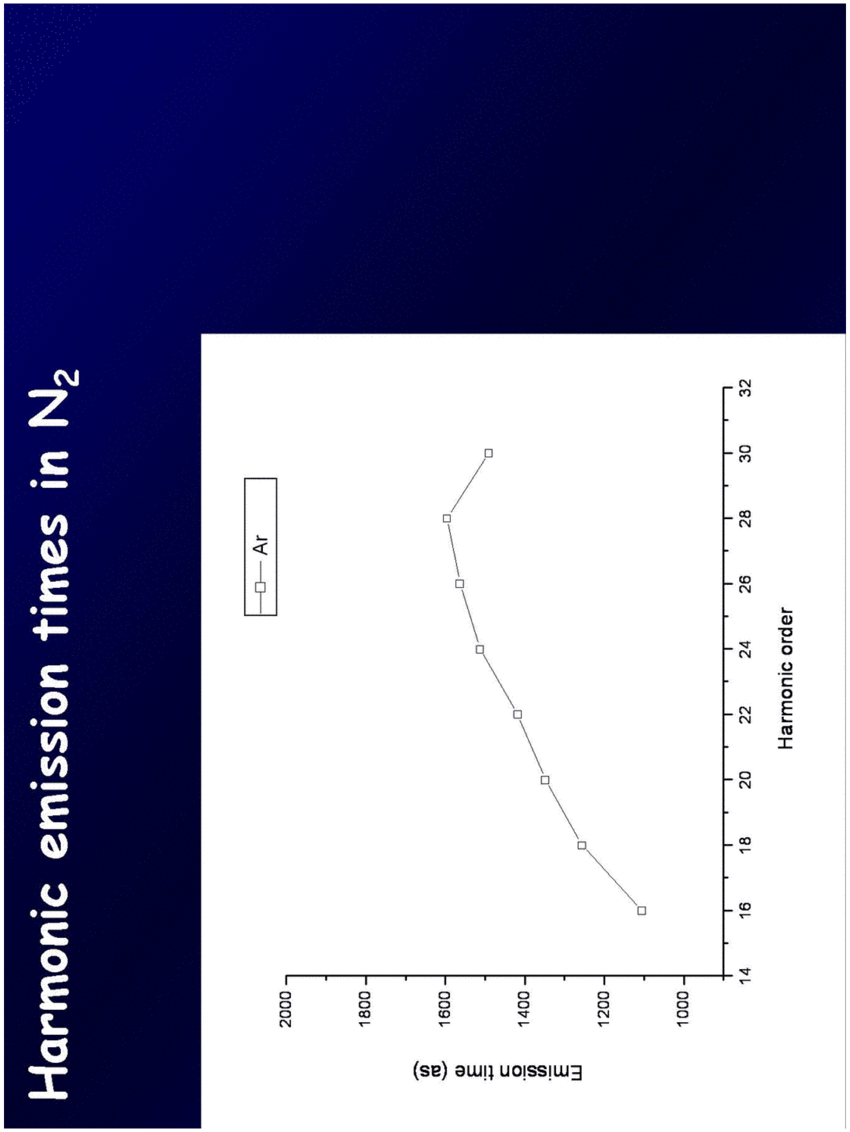
⇒two-point emitter model seems to apply

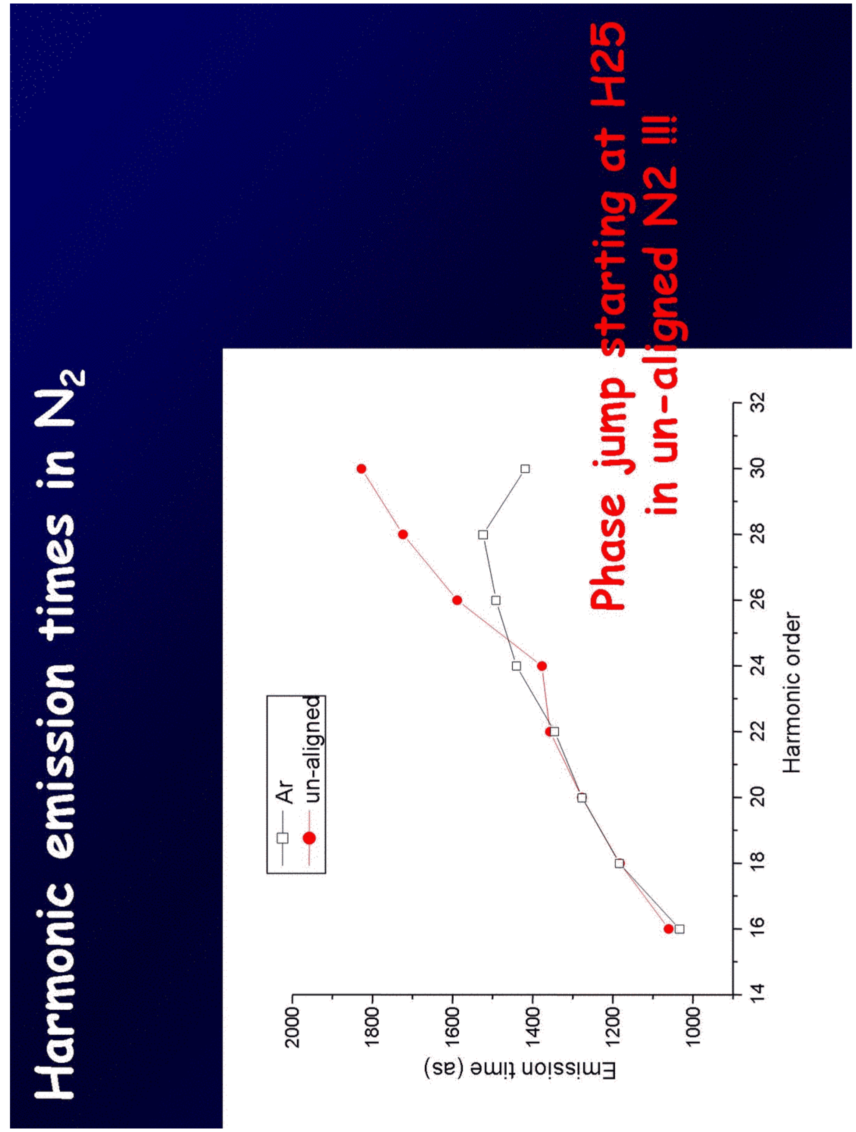
BUT not really a shift to higher order with angle: In fact: smaller phase jump and amplitude minimum
=> Contradiction with Lein et al PRL 2002 and Vozzi et al. PRL 2005 (minimum at H33)

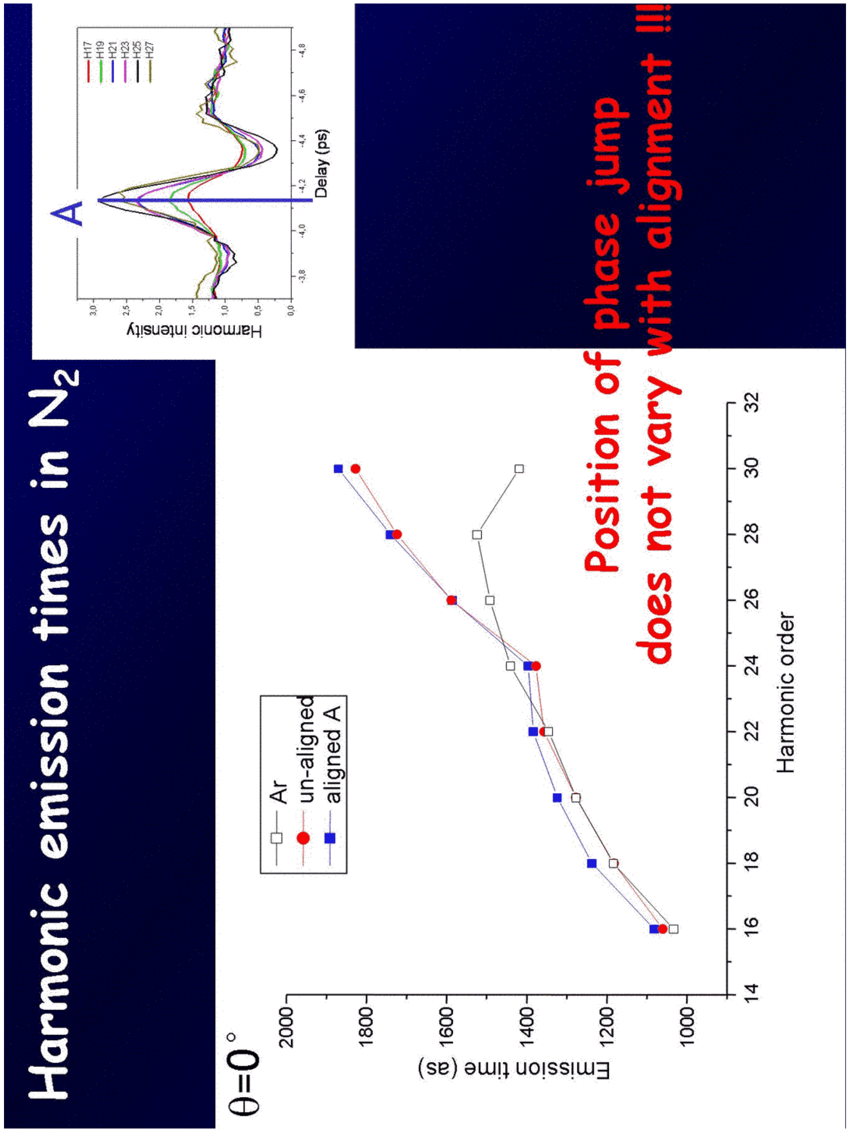
$$r_0 \cos(\theta) = n\lambda_e$$

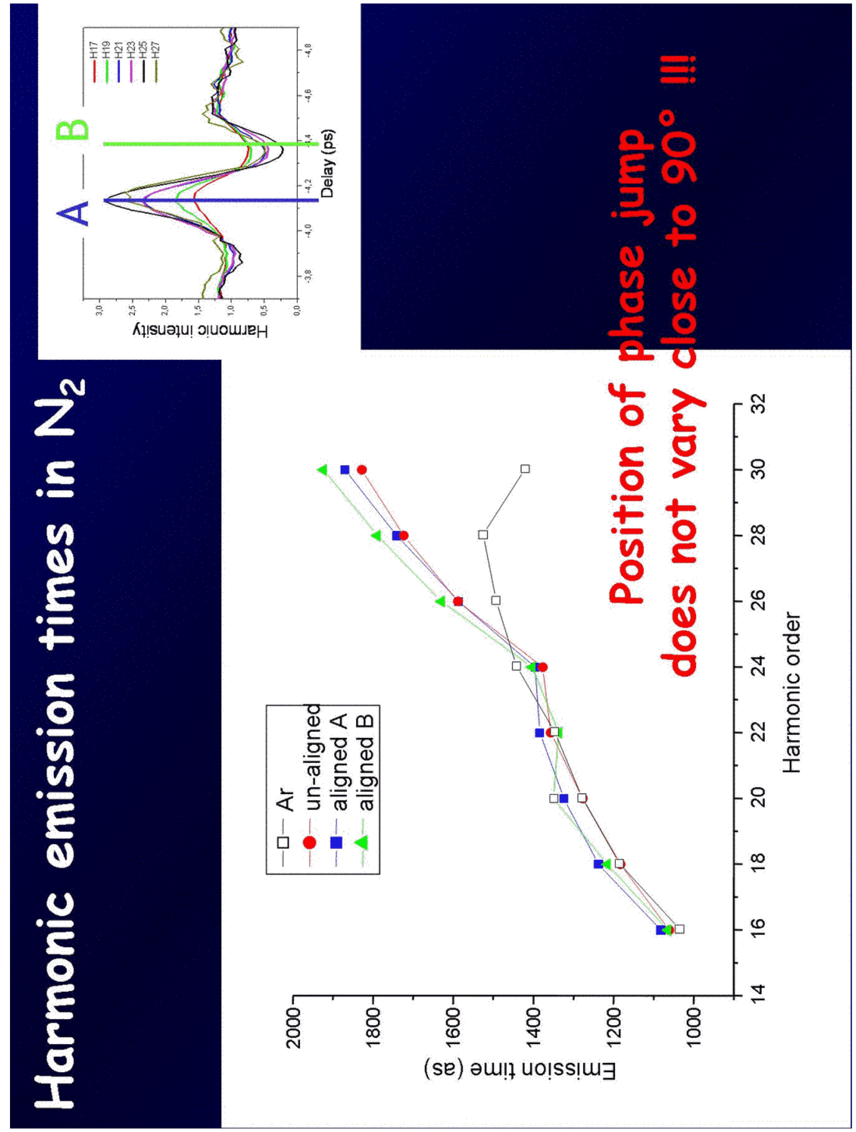
Observation of half revival in aligned N_2

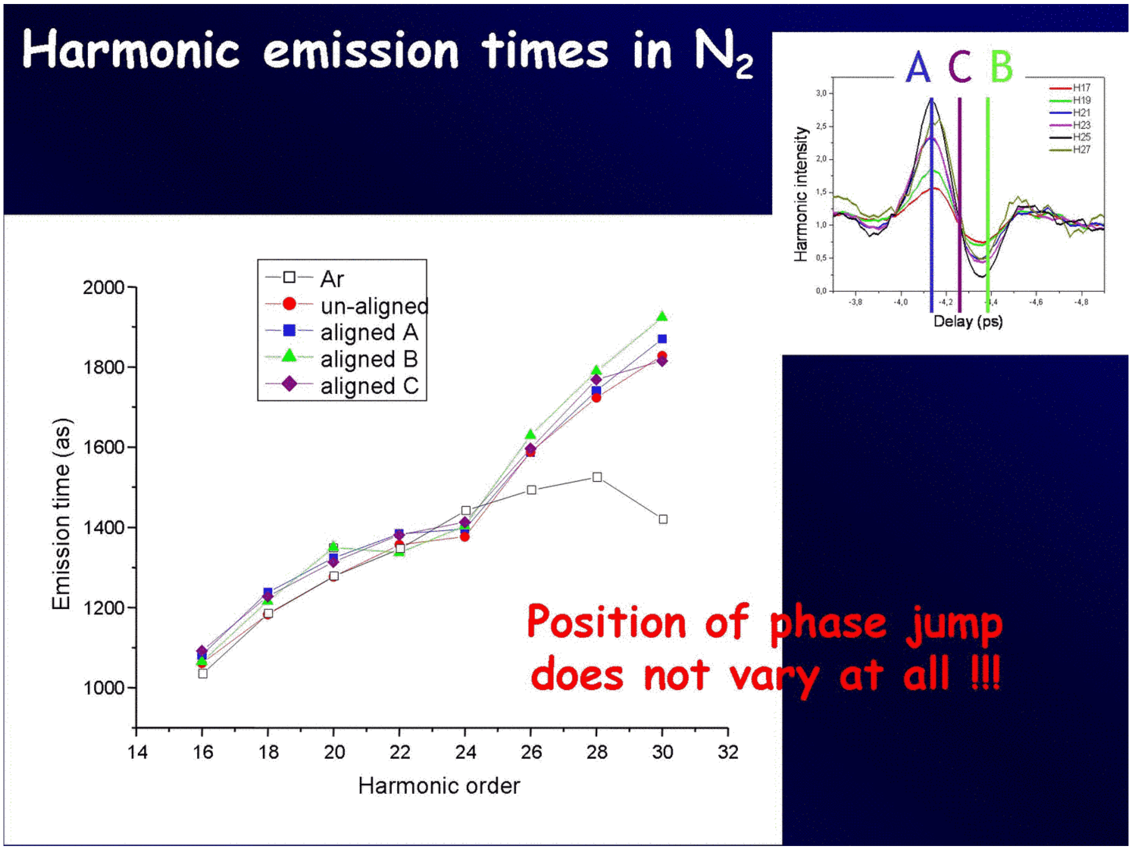


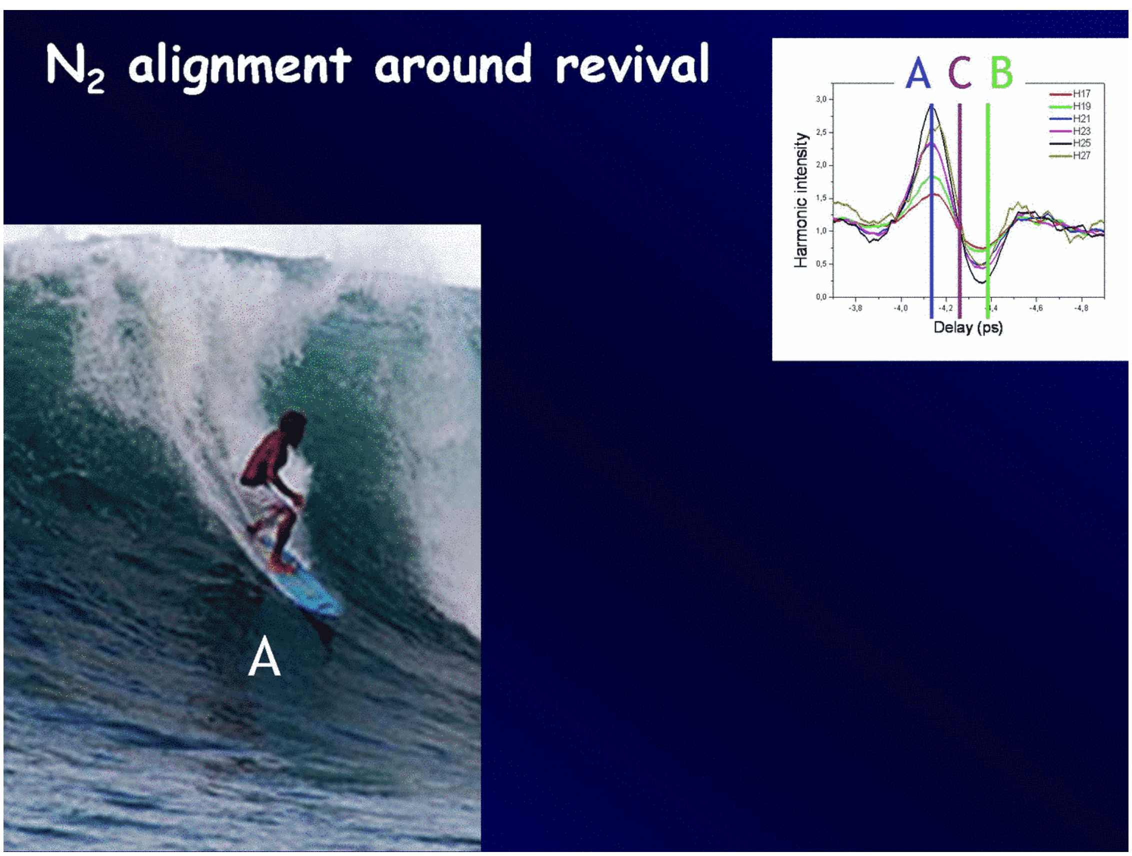

Harmonic emission times in N2
Ar
Emission time (as)
2000
1800
1600
1400
1200
1000
Harmonic order
14
16
18
20
22
24
26
28
30
32

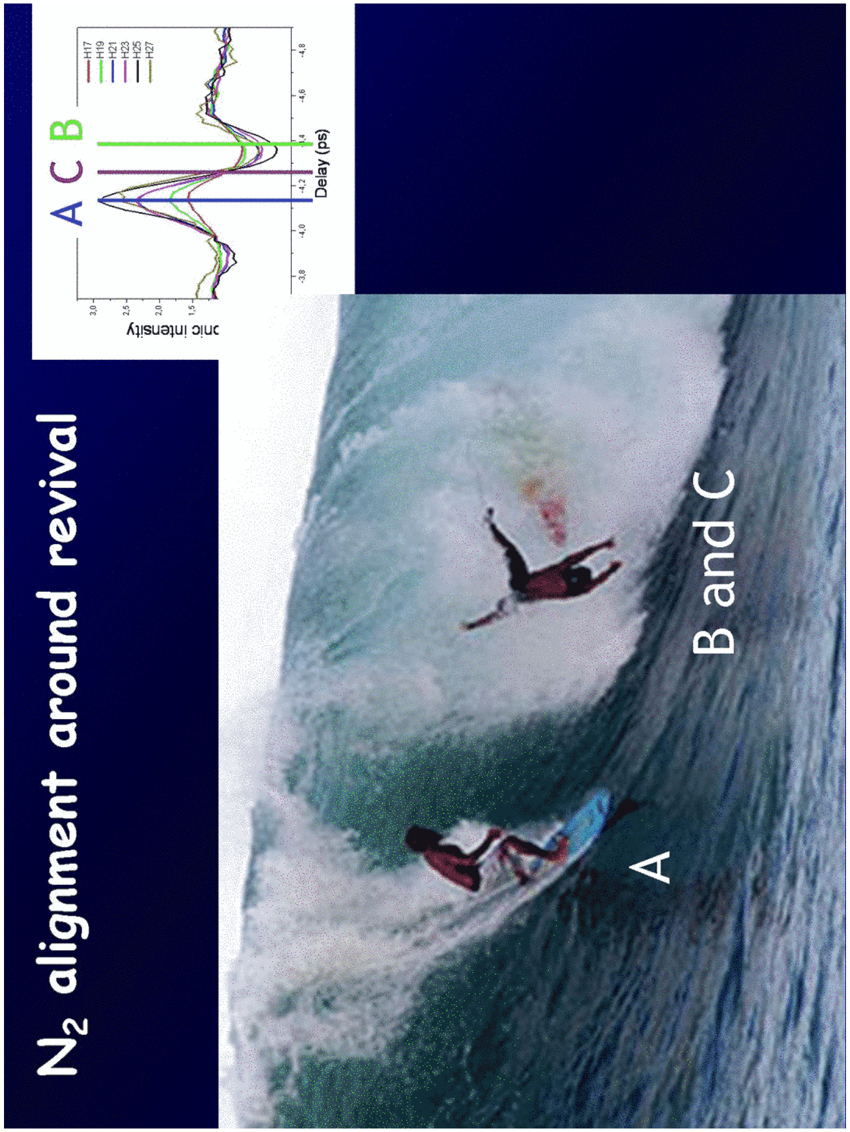

Harmonic emission times in N2
Phase jump starting at H25
in un-aligned N2 !!!
Ar
un-aligned
Emission time (as)
2000
1800
1600
1400
1200
1000
Harmonic order
14
16
18
20
22
24
26
28
30
32

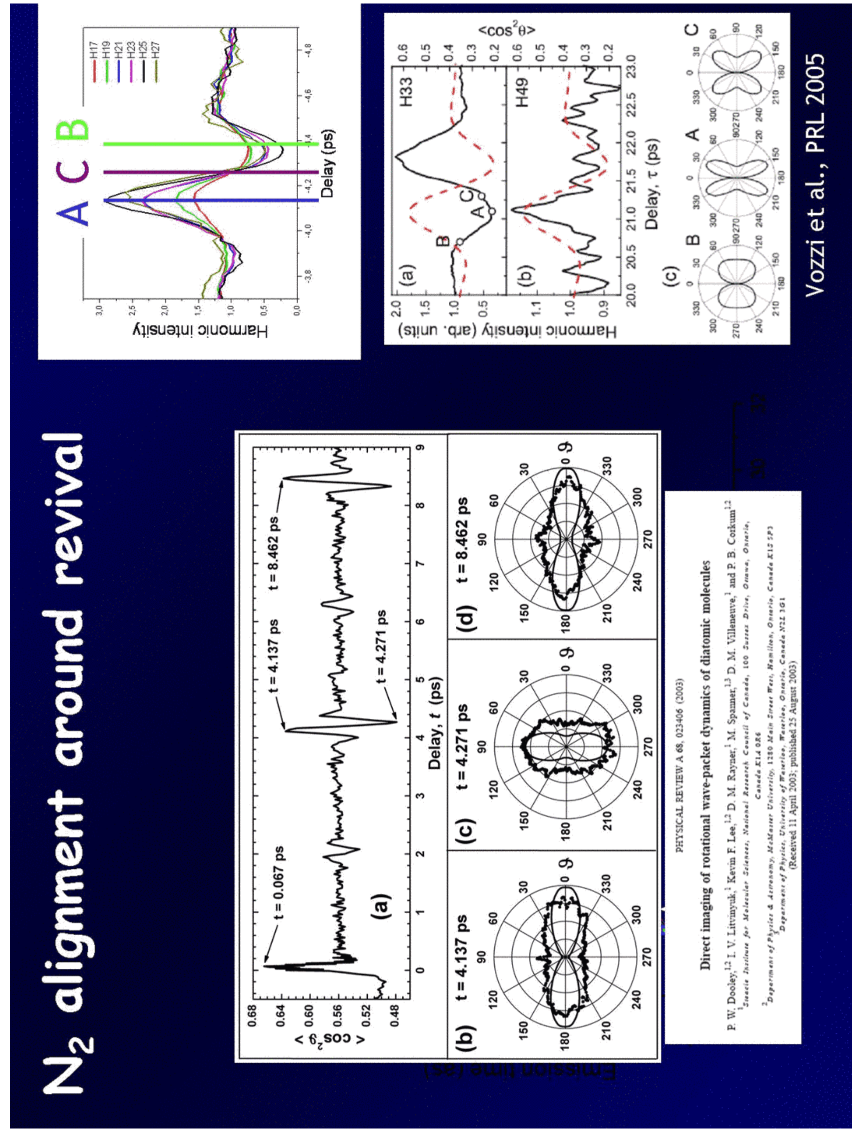

Harmonic emission times in N_2
θ=0°
Ar
un-aligned
aligned A
Emission time (as)
Harmonic order
Harmonic intensity
Delay (ps)
A
H17
H19
H21
H23
H25
H27
Position of phase jump
does not vary with alignment !!!


Harmonic emission times in N_2
Ar
un-aligned
aligned A
aligned B
Emission time (as)
Harmonic order
Harmonic intensity
Delay (ps)
A
B
H17
H19
H21
H23
H25
H27
Position of phase jump
does not vary close to 90° !!!

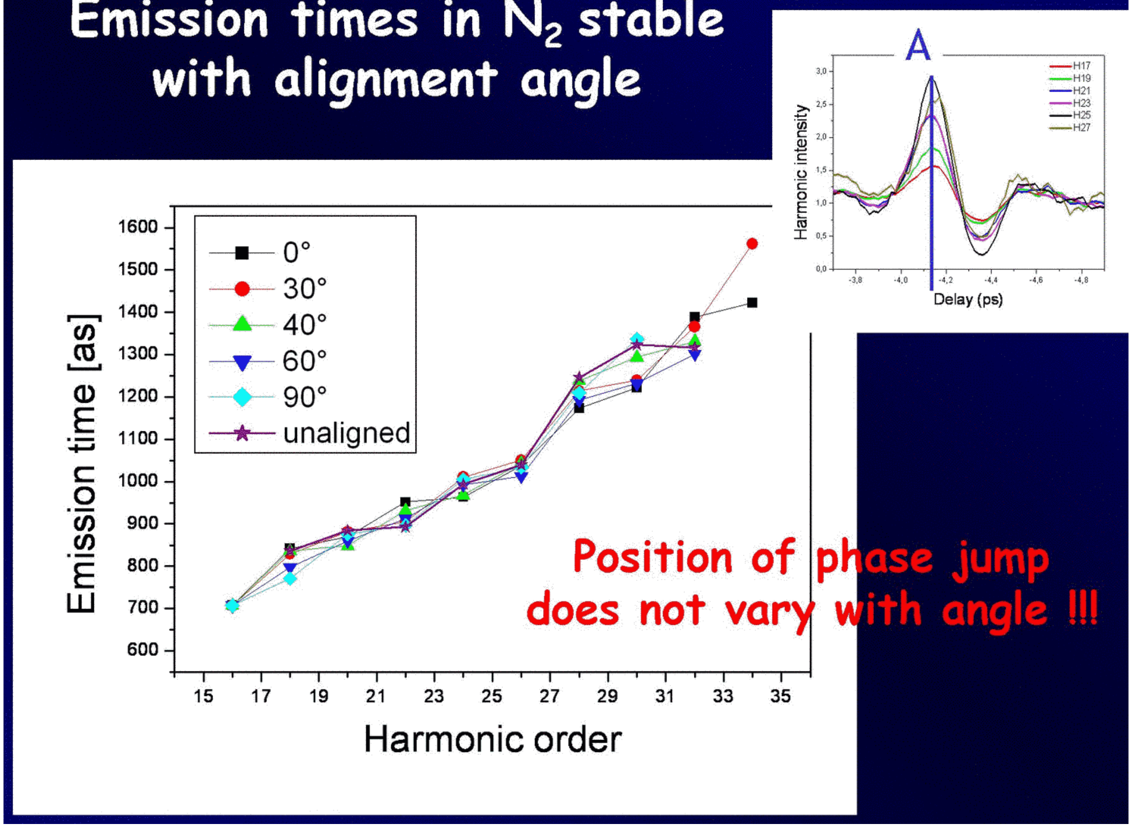

Harmonic emission times in N_2
A C B
H17
H19
H21
H23
H25
H27
Harmonic intensity
Delay (ps)
Ar
un-aligned
aligned A
aligned B
aligned C
Emission time (as)
Harmonic order
Position of phase jump
does not vary at all !!!


N_2 alignment around revival
A C B
H17
H19
H21
H23
H25
H27
Harmonic intensity
Delay (ps)
A


N_2 alignment around revival
A C B
Delay (ps)
A
B and C


N_2 alignment around revival
A C B
Harmonic intensity
Delay (ps)
Vozzi et al., PRL 2005
PHYSICAL REVIEW A 68, 023406 (2003)
Direct imaging of rotational wave-packet dynamics of diatomic molecules

Emission times in N_2 stable with alignment angle



Conclusion for N2

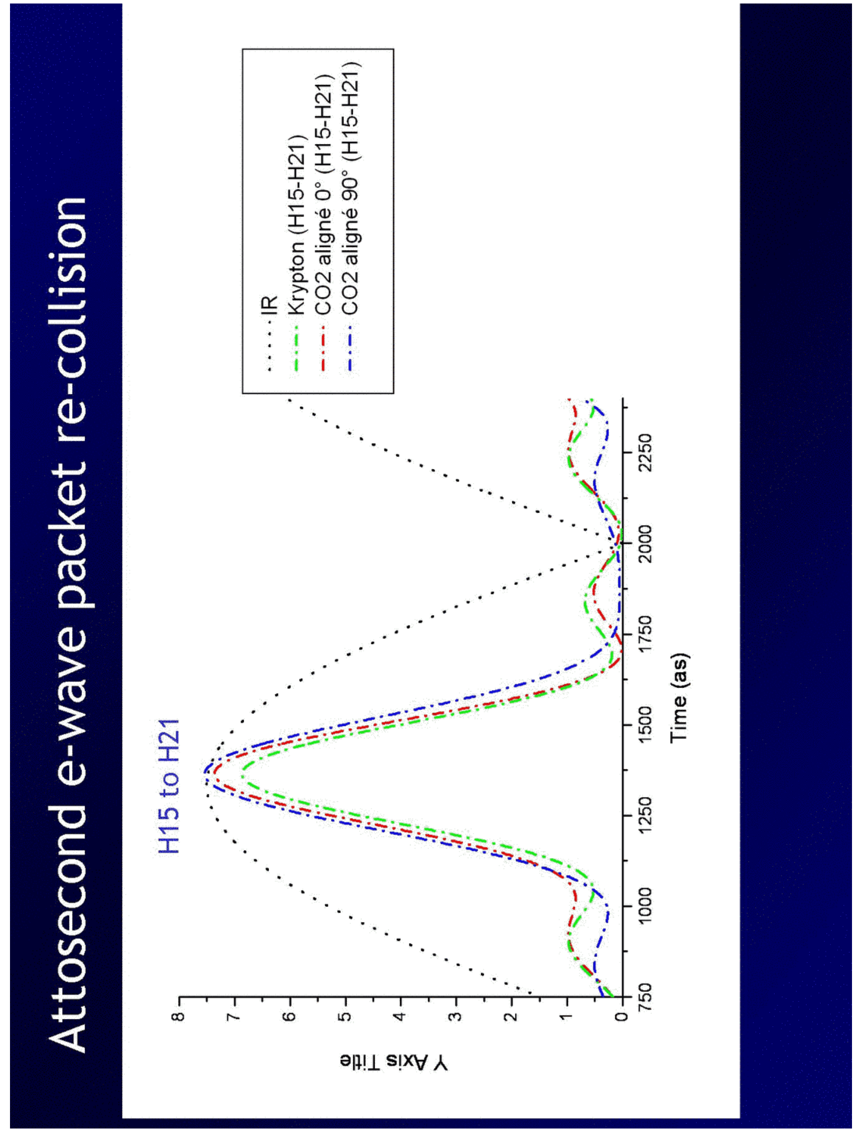
Phase jump starting at H25
Amplitude minimum at H25 → OK with
Itatani et al Nature 2004
Kanai et al Nature 2005

Position of phase jump and amplitude minimum do not vary with angle !!! → Never mesured before in N_2

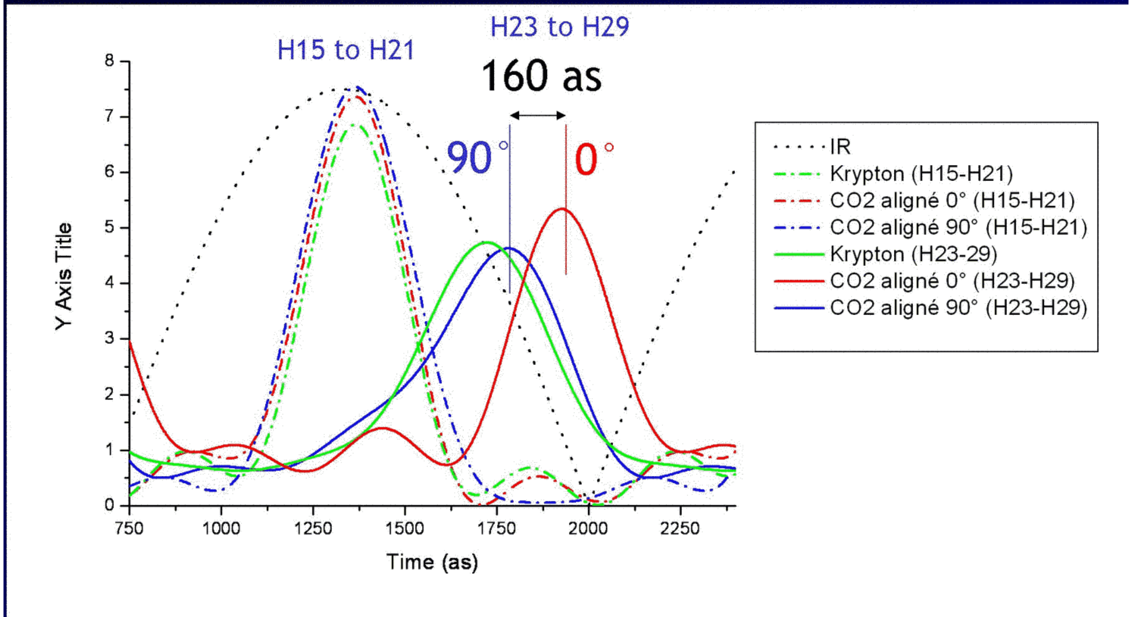
⇒ two-point emitter model does not apply

Theoretical input needed!!


Insight into
attosecond dynamics
Please be patient only two slides left...thanks


Attosecond e-wave packet re-collision
H15 to H21
Y Axis Title
Time (as)
IR
Krypton (H15-H21)
CO2 aligné 0° (H15-H21)
CO2 aligné 90° (H15-H21)
0
1
2
3
4
5
6
7
8
750
1000
1250
1500
1750
2000
2250

Attosecond e-wave packet re-collision



Conclusions

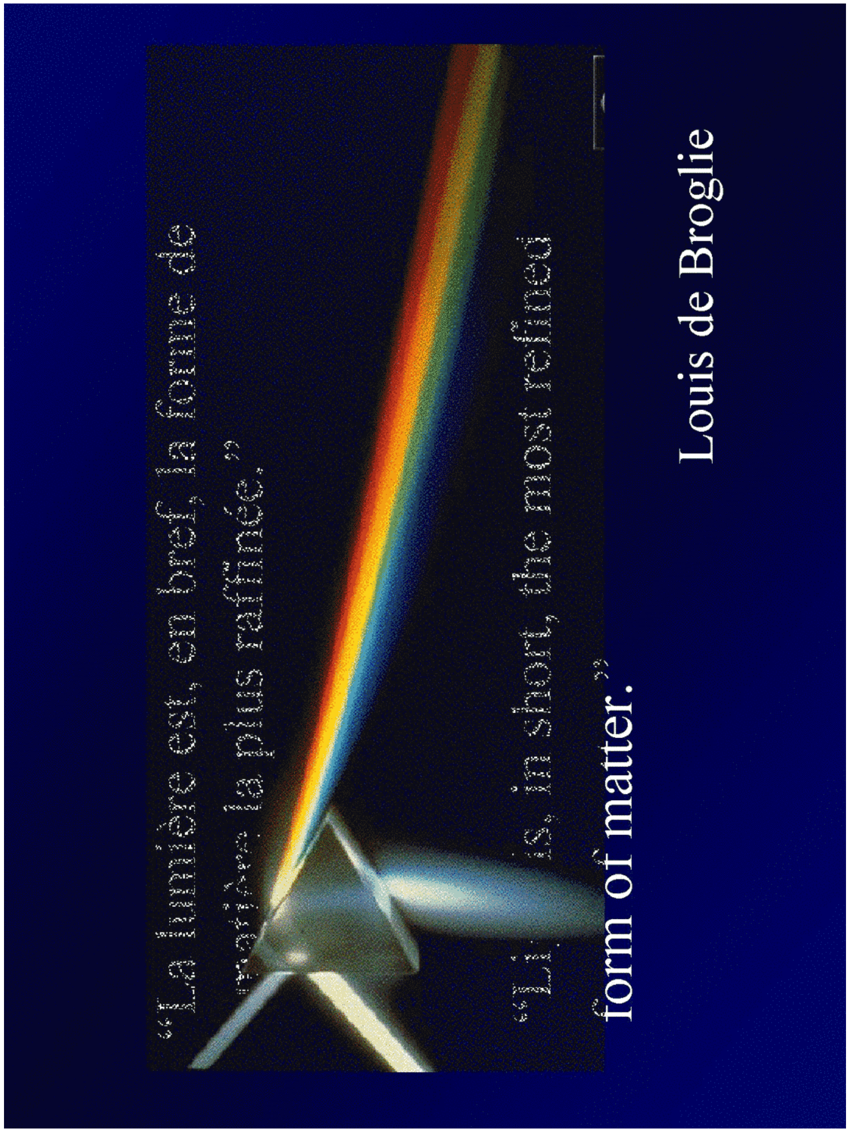
APT emitted by an atom are chirped
due to the wavepacket dynamics in the continuum

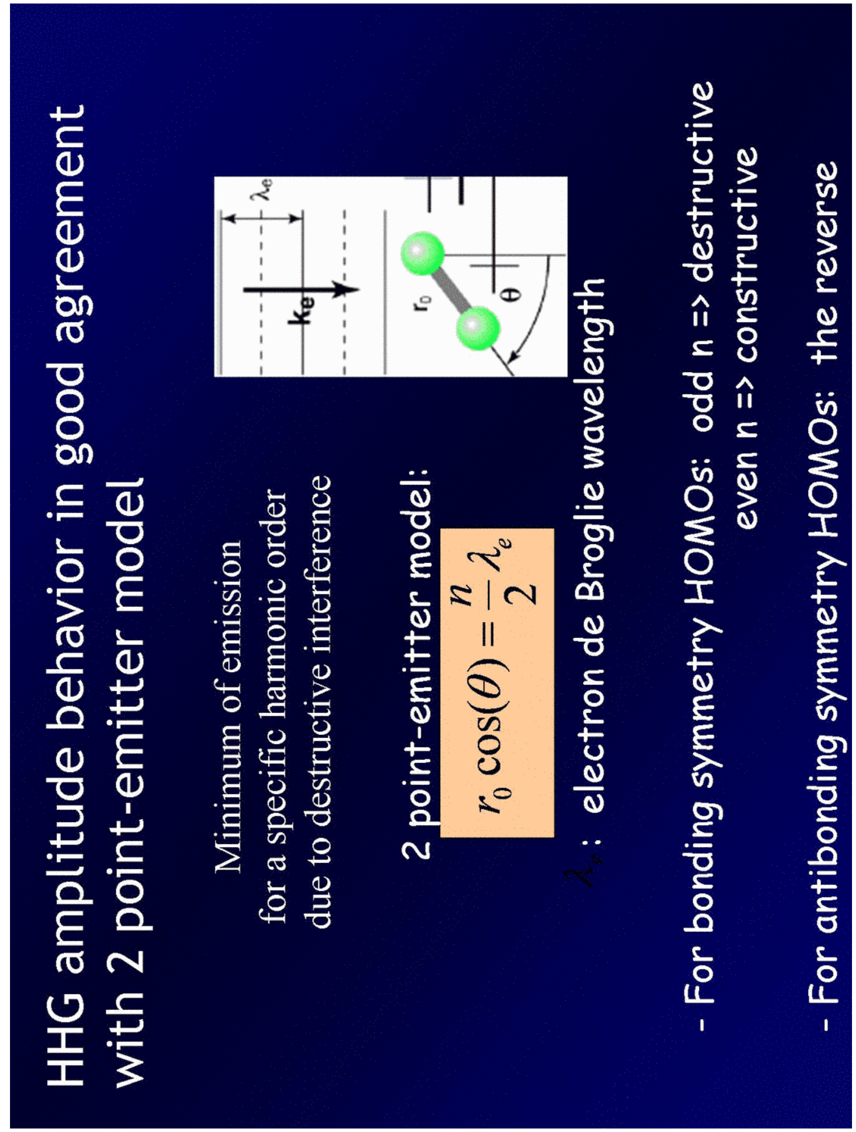
APT emitted by molecules : chirp + phase jump
due to interference in the recombination process

CO_2: jump partially follows two-point emitter model
N_2: jump independent of angle
=> two-point emitter model is not enough sophisticated

Perspectives

- Extensive experimental data for validating theory
- Accurate tomographic reconstruction of orbitals
- Attosecond dynamics in molecules (pump-probe)


“La lumière est, en bref, la forme de
la plus raffinée.”
is, in short, the most refined
form of matter.”
Louis de Broglie


HHG amplitude behavior in good agreement with 2 point-emitter model
Minimum of emission for a specific harmonic order due to destructive interference
λ_e
k_e
r_0
θ
2 point-emitter model:
$r_0 \cos(\theta) = \frac{n}{2}\lambda_e$
λ_e : electron de Broglie wavelength
- For bonding symmetry HOMOs: odd n => destructive
even n => constructive
- For antibonding symmetry HOMOs: the reverse