Imperial College London

Experimental approaches to HHG spectroscopy

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Imperial College

EPSRC
Pioneering research and skills

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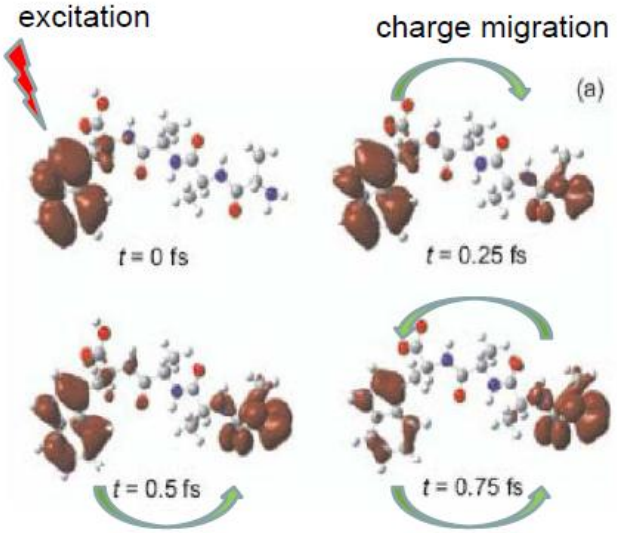
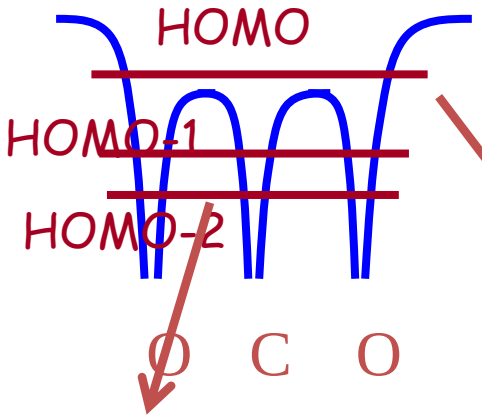
erc

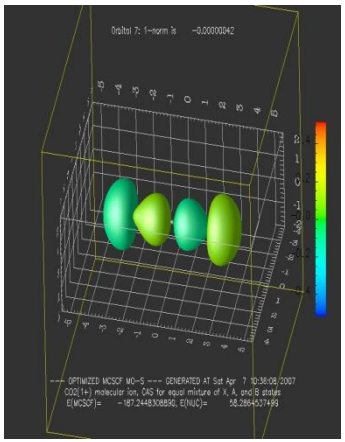
The Goal

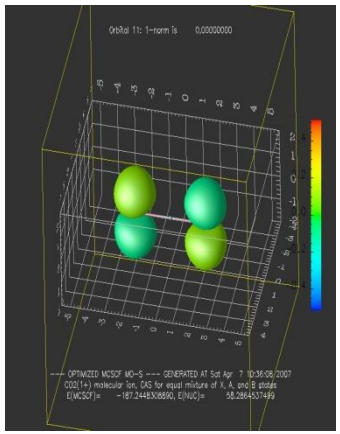


We expect complex electron dynamics in a cation following sudden ionization. How can we measure this sort of attosecond dynamics?



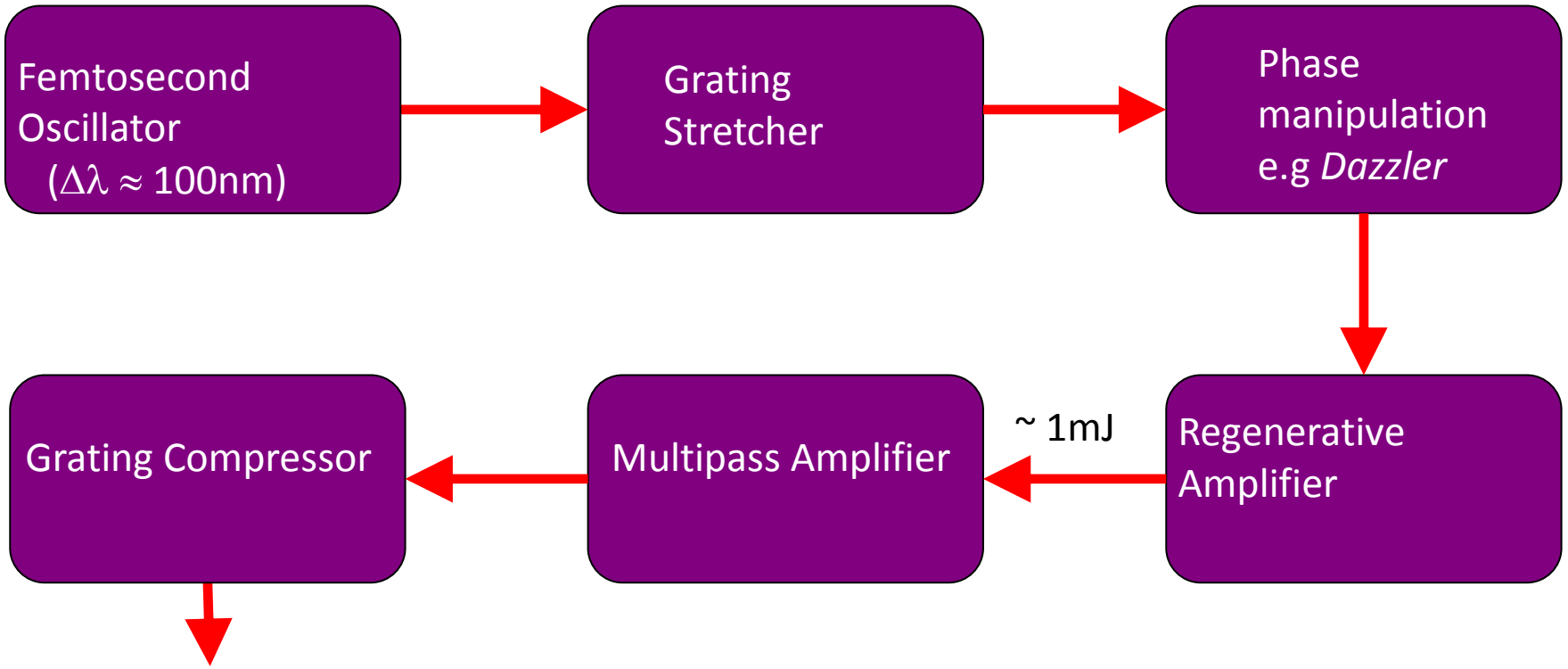
Tunnel ionization from different orbitals in HHG: A hole or superposition of holes can form. This results in charge motion on a sub-fs timescale.





How can we observe and measure this through HHG spectroscopy?

Tools 1:
High power short pulse lasers



800nm output, ~ 30 fs
(bandwidth ~80nm)

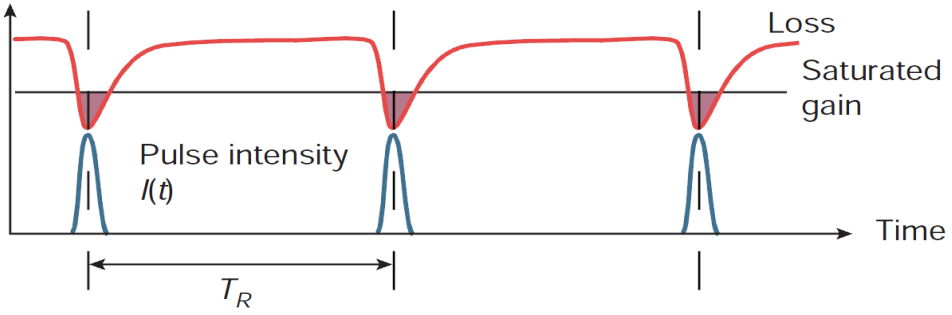
Ti:S = Titanium Sapphire

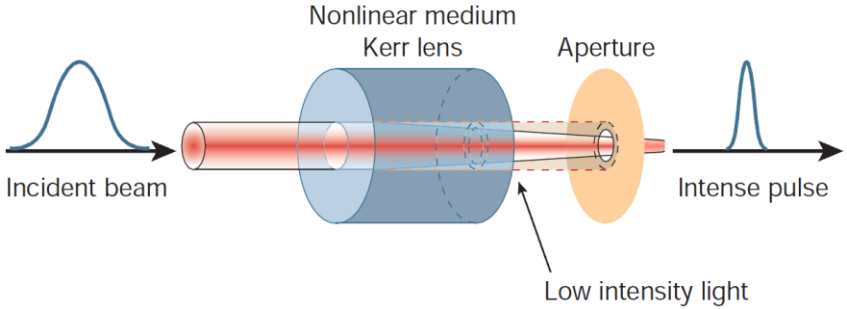
CPA = Chirped Pulse Amplified

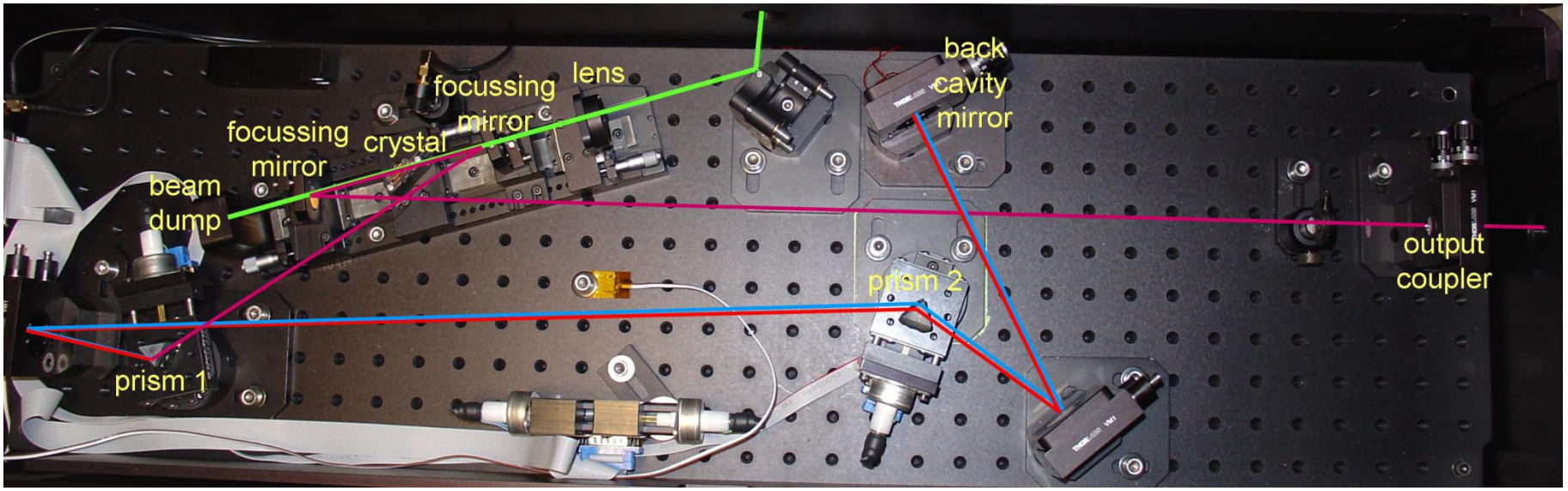
Kerr Lens Modelocking: Operation of Titanium Sapphire Oscillator

A 5fs pulse requires a bandwidth of ~10^{14} Hz to support it, which requires the phase locking of many oscillator modes to achieve this (~ 10^{6} given a typical 100MHz mode spacing)

Self-focusing caused by the intensity dependent refractive index can be used to ensure only the mode-locked (highest intensity) operation sees sufficient gain to lase in the oscillator



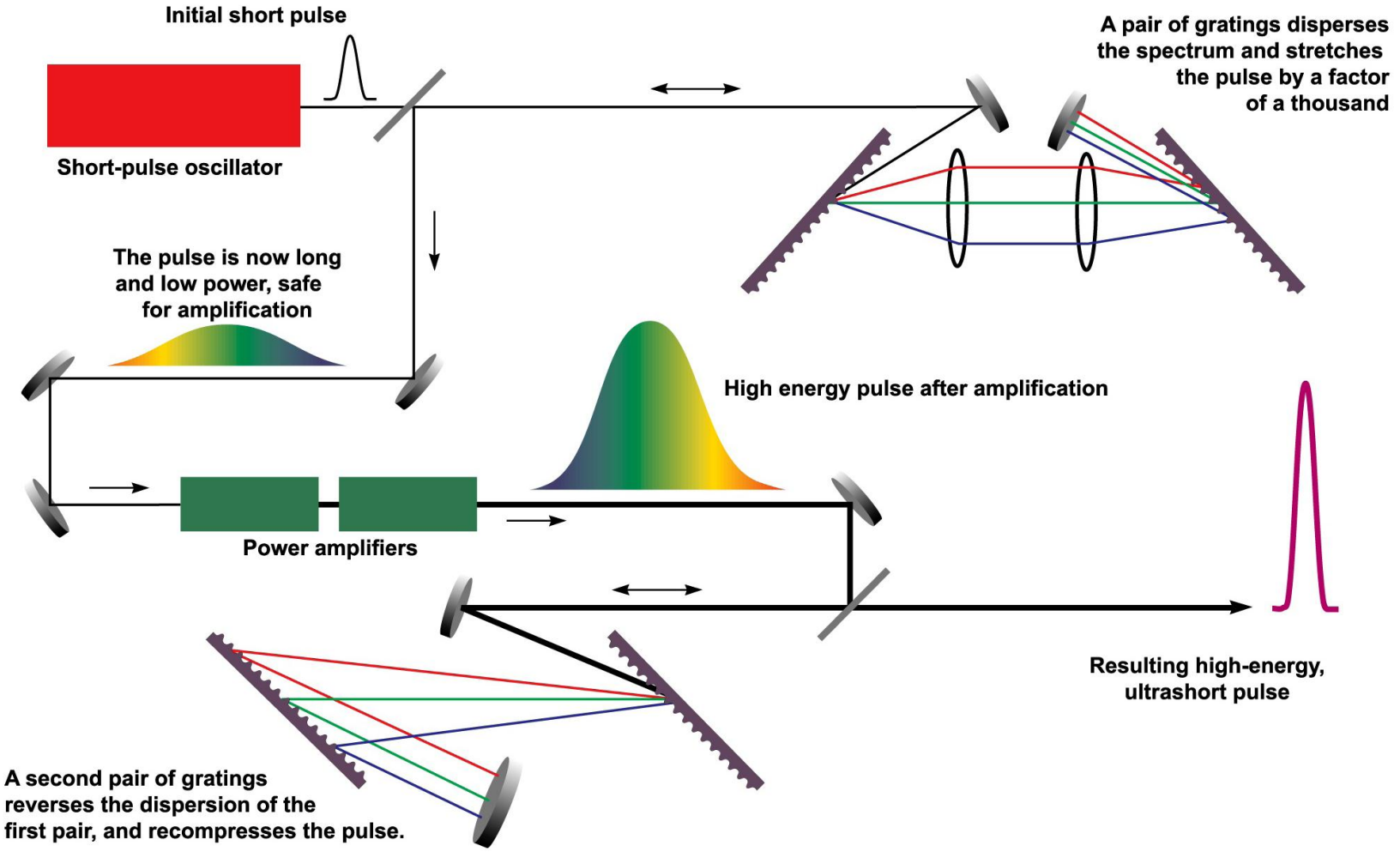




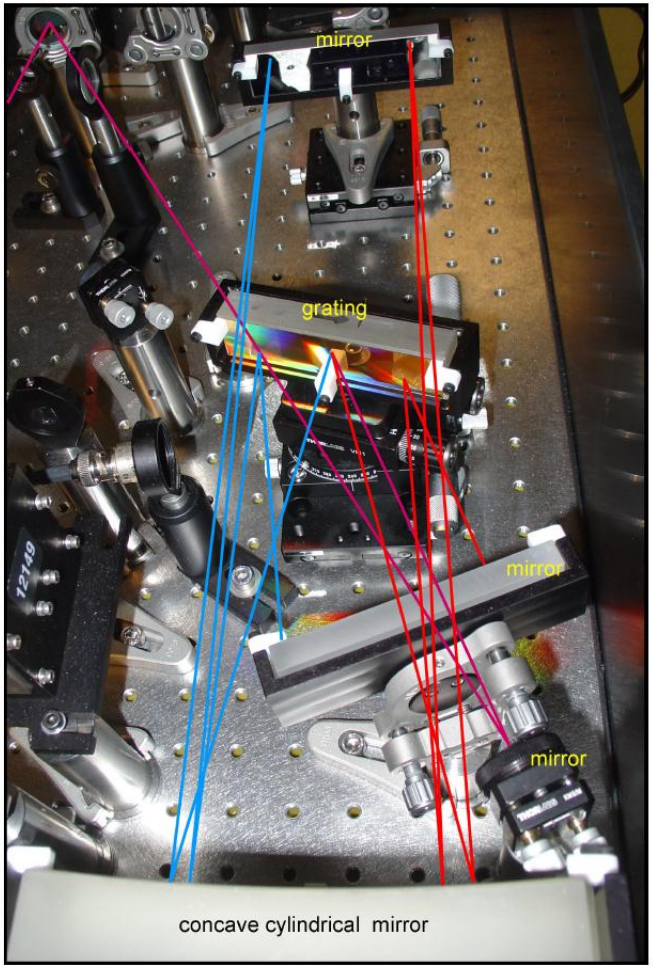
Opt.Lett. 16, 42 (1991)

Chirped Pulse Amplification

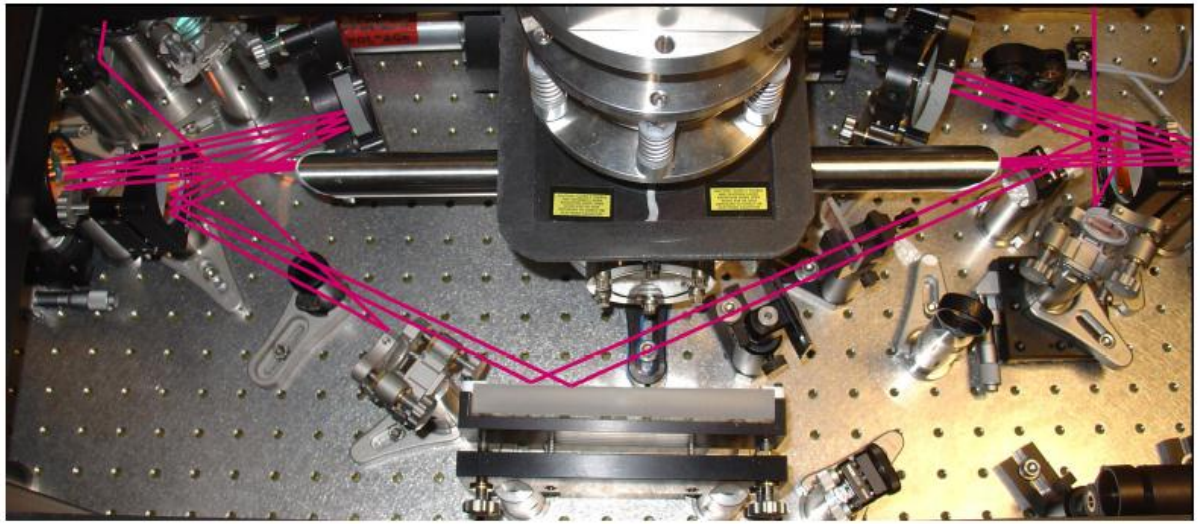
Invented by Gerard Mourou and Donna Strickland the CPA architecture enables the pulse to be amplified to high power whilst it is chirped and stretched **Opt.Comm. 56, 219 (1985)**
Temporal stretch from ~30fs to >300ps (factor 10^4)



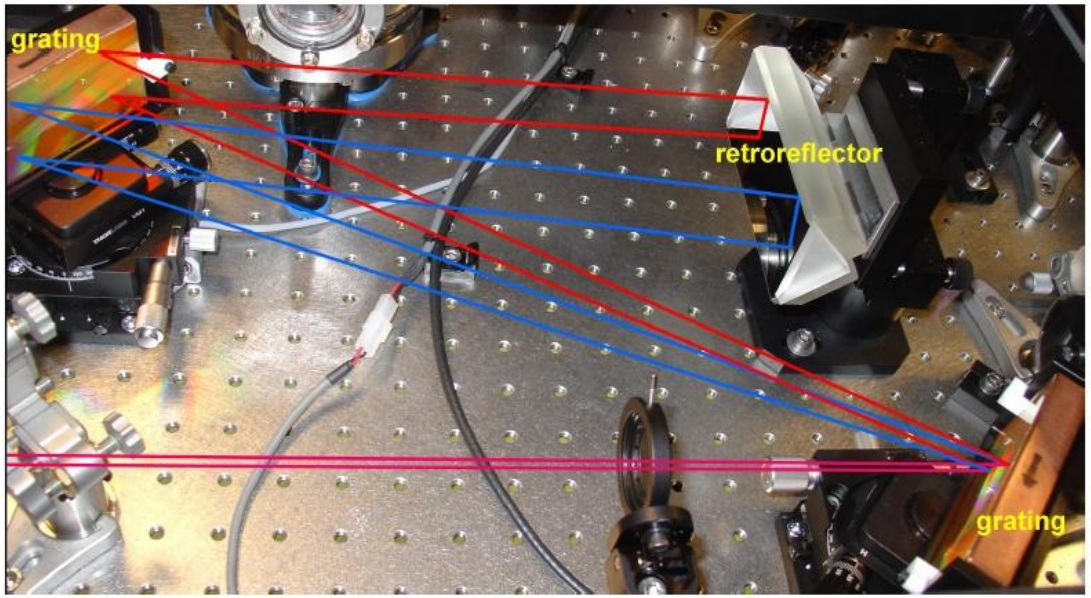
Optical Layout of Components in a CPA Ti:S System



Stretcher

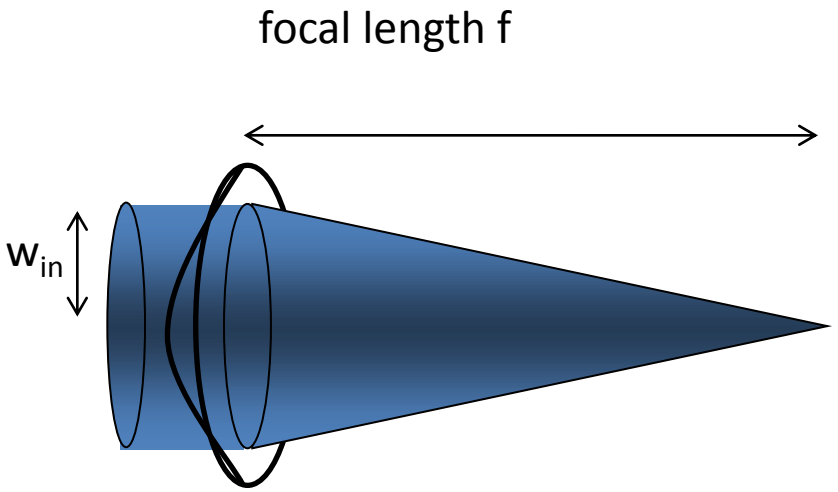


Amplifier



Compressor

High intensity can be obtained at the focus of a lens or spherical mirror



Spot waist size (radius) for a Gaussian beam:

$$w_0 = f \lambda / \pi w_{in}$$

Pulsed (20 fs) 10mJ mode-locked/CPA amplified Ti:S laser focused by 20cm lens

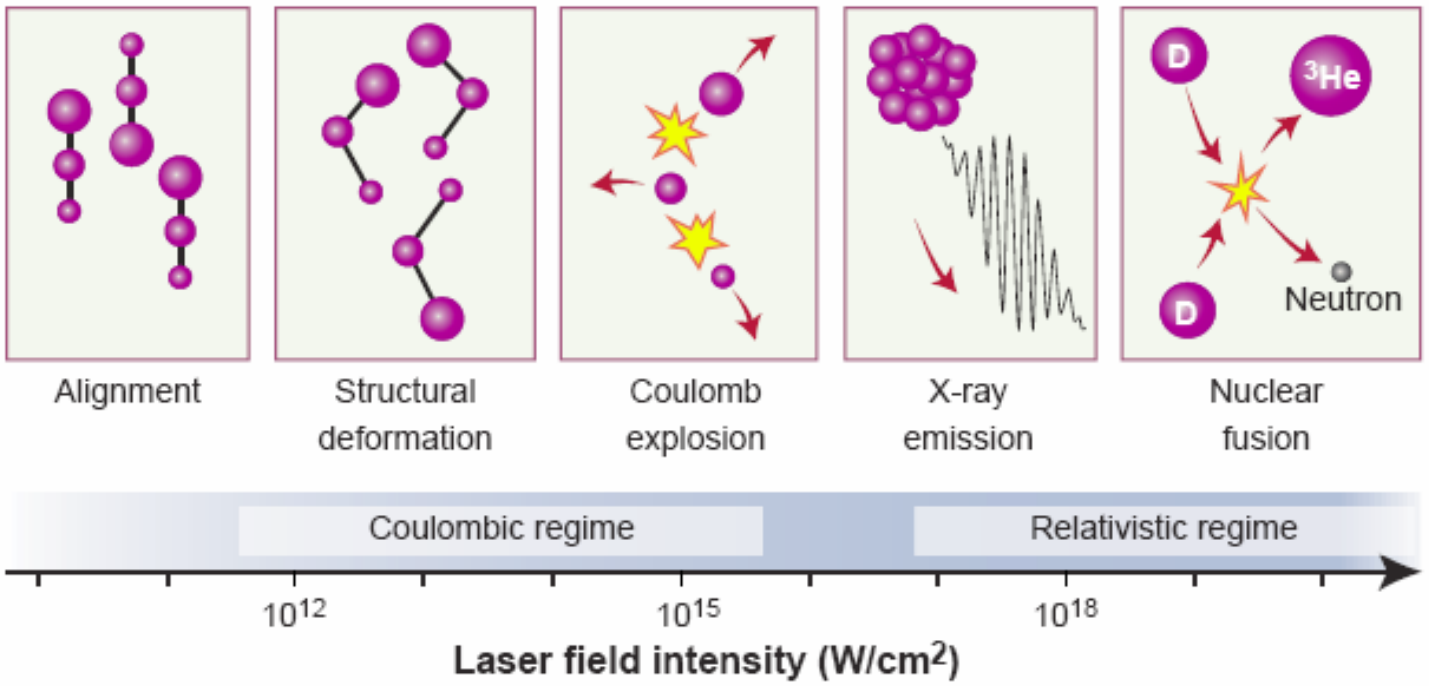
Intensity ~ 5 x 10^{17} Wcm^{-2}

Peak electric field in Vm^{-1}:

$E_0 = 2.745 \times 10^3 \, (I \, (Wcm^{-2}))^{1/2}$

e.g I = 10^{15} Wcm^{-2} $E_0 = 8.68 \times 10^{10}$ Vm^{-1} (8.68 V per Å)

Across a small molecule ~ 3 Å we can easily have a potential difference of 5 – 10 eV in the laser field



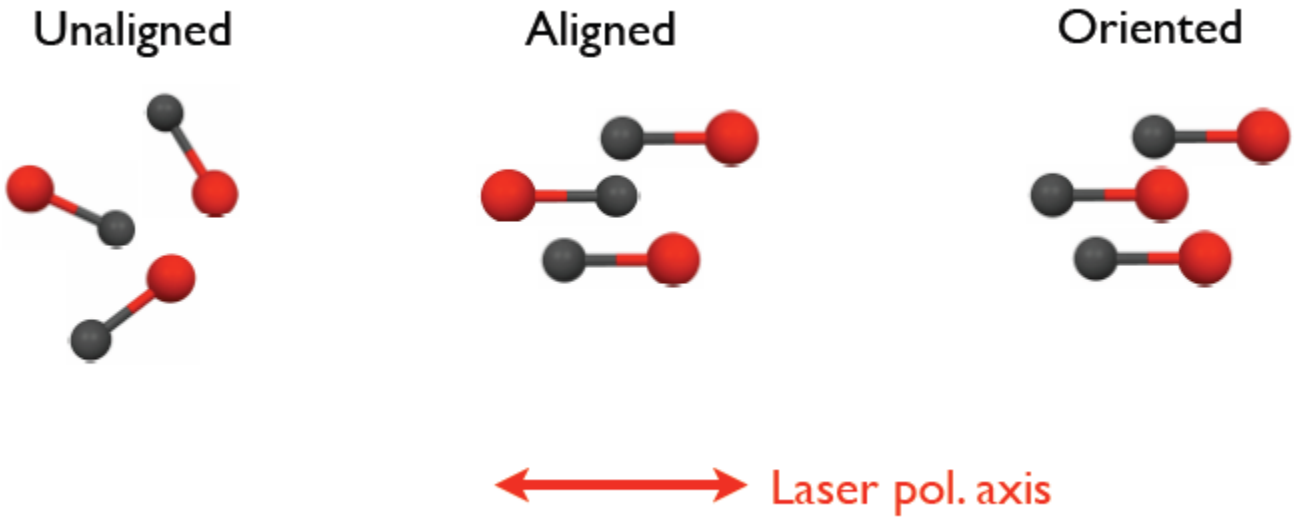
Exotic behavior of molecules and clusters in intense laser fields.

From Yamanouchi Science (2002)

Technical Issues – High Power Lasers

- Commercially available – but expensive
- Require high grade diagnostics and a masters level training (minimum) to operate at spec.
- Great care must be taken with pulse duration and beam spatial profile to avoid optical damage and permit high quality measurements to be made
- Maintenance difficult and expensive
- Controlled environment – vibration, temperature & humidity (laser tables, air conditioning, chilled water supply)
- Intensity in laser beam focus (even for a Gaussian) is not spatially uniform – this must always be remembered when interpreting experimental results

Tools 2:
Fixing molecules in space



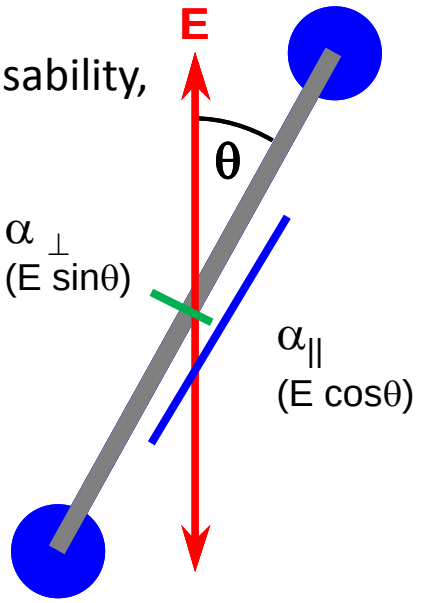
Laser Field Induced Electronic Polarisation Forces Alignment

Interaction Hamiltonian with a molecule due to the dynamic polarisability, the molecular polarisability anisotropy is:

$$(\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp})$$

$$\vec{E}(t) = \hat{\varepsilon} E_0 f(t) \cos\,\omega t,$$

The laser field is at a high frequency ω with a slowly varying envelope f(t).



$$Torque = -{}^{dV_{\text{int}}}\!/_{d\theta} = -E(t)^2 \Delta\alpha \cos\theta \sin\theta$$

The interaction depends upon the magnitudes of the parallel and perpendicular polarizability, the field strength and the direction of polarization

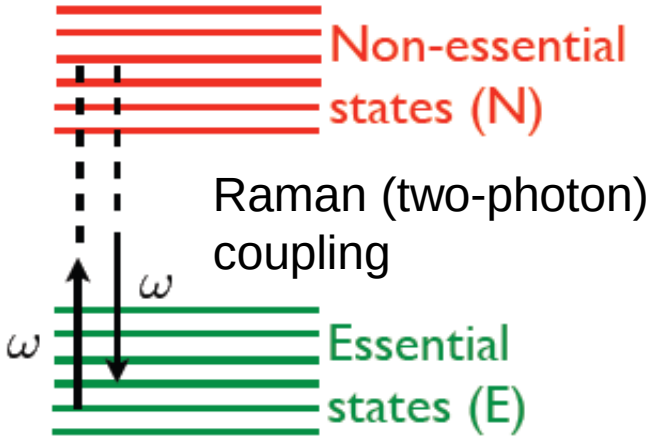
$$V_{\text{int}} = -\frac{1}{2} E(t)^2 (\Delta\alpha \cos^2\theta + \alpha_{\perp}),$$

See: **Torres et al Phys.Rev. A 72, 023420 (2005)**
Stapelfeldt & Seideman Rev.Mod Phys vol 75 543 (2003)

Laser induced alignment

The non-resonant laser creates a broad superposition of (rotational) states in E through all the states in N:

$$\Psi(t) = \sum_E c_E(t)\psi_E, \quad H_0\psi_E = \epsilon_E\psi_E$$



The coupling of states in E is well-described by an effective Hamiltonian:

$$i\frac{\partial\Psi(t)}{\partial t} = H_{\text{eff}}\Psi(t)$$

$$H_{\text{eff}} = H_0 - \frac{1}{4}\sum_{ij}\alpha_{ij}E_j(t)E_i(t)$$

Intensity profile

Dynamic polarizability

The temporal evolution of alignment depends upon the relative duration of the pulse and the rotation period.

impulsive

adiabatic

increasing pulse duration

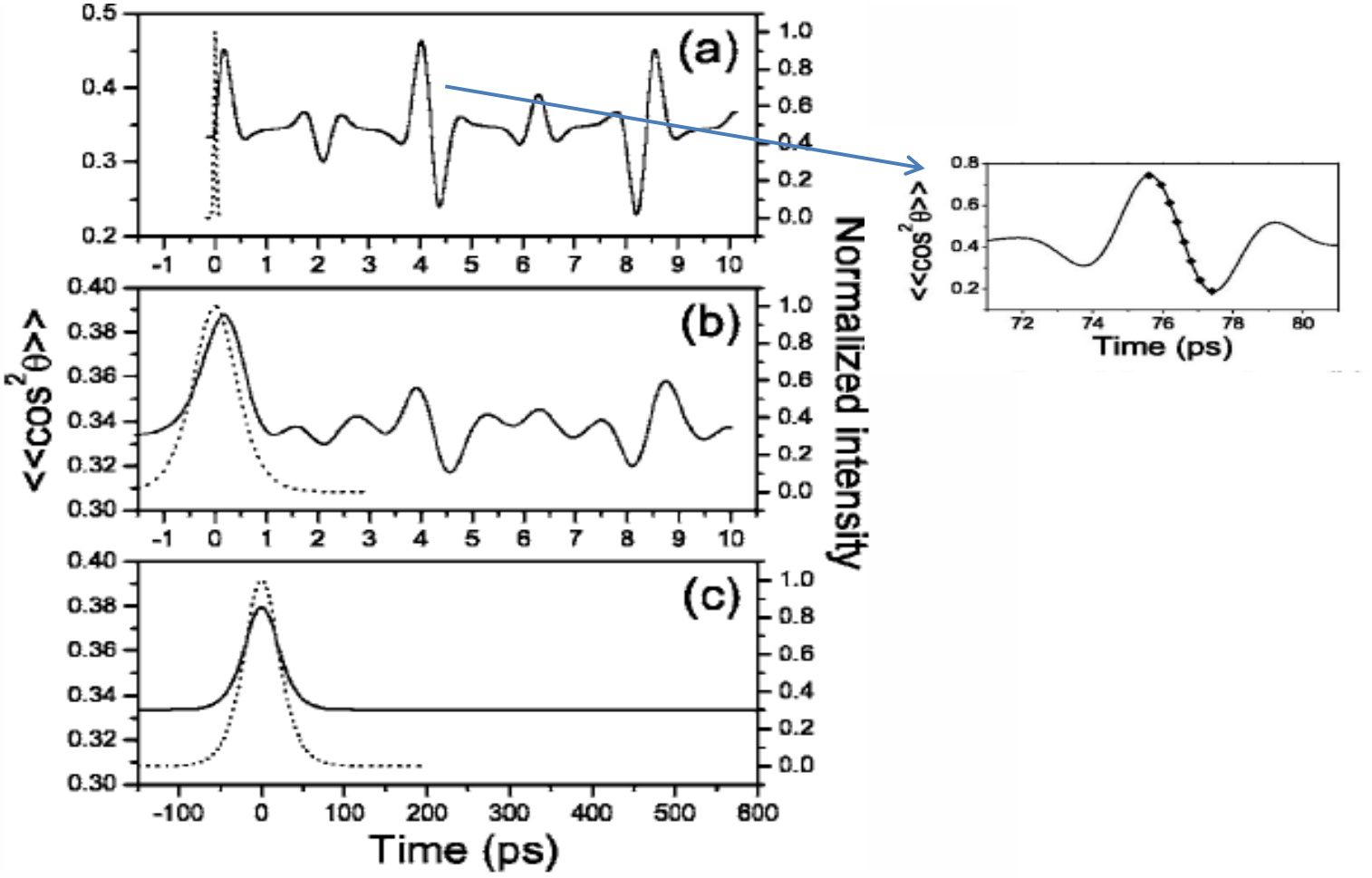
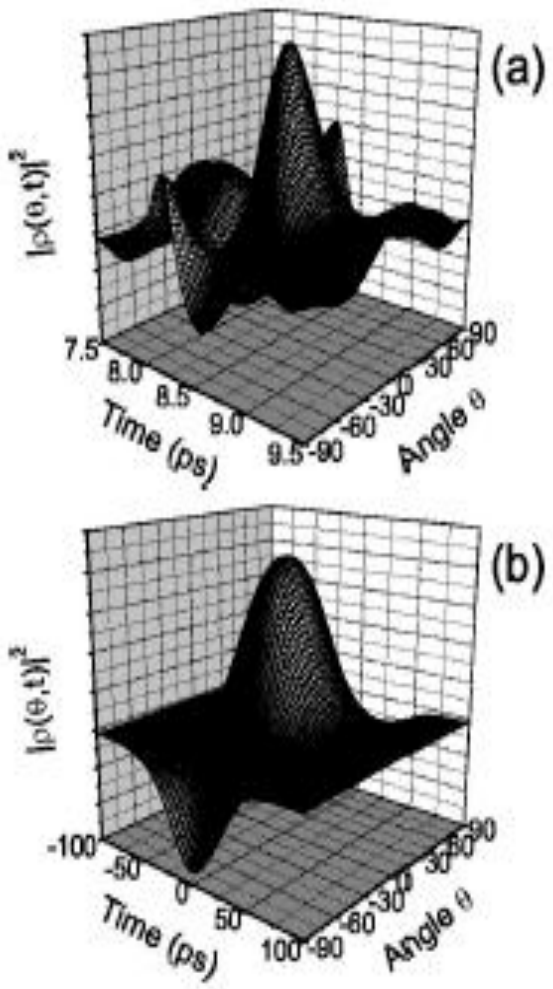


FIG. 1. Time evolution of the degree of alignment in N_2 at 50 K with different pulse durations and peak intensities; (a) τ=50 fs, I_0 =2.5×10^{13} W/cm^2, (b) τ=1 ps, I_0=2.5×10^{12} W/cm^2, (c) τ =50 ps, I_0=2.5×10^{12} W/cm^2. The pulse profiles are represented as dotted curves for reference.

Temporal evolution around a field free revival in the impulsive limit



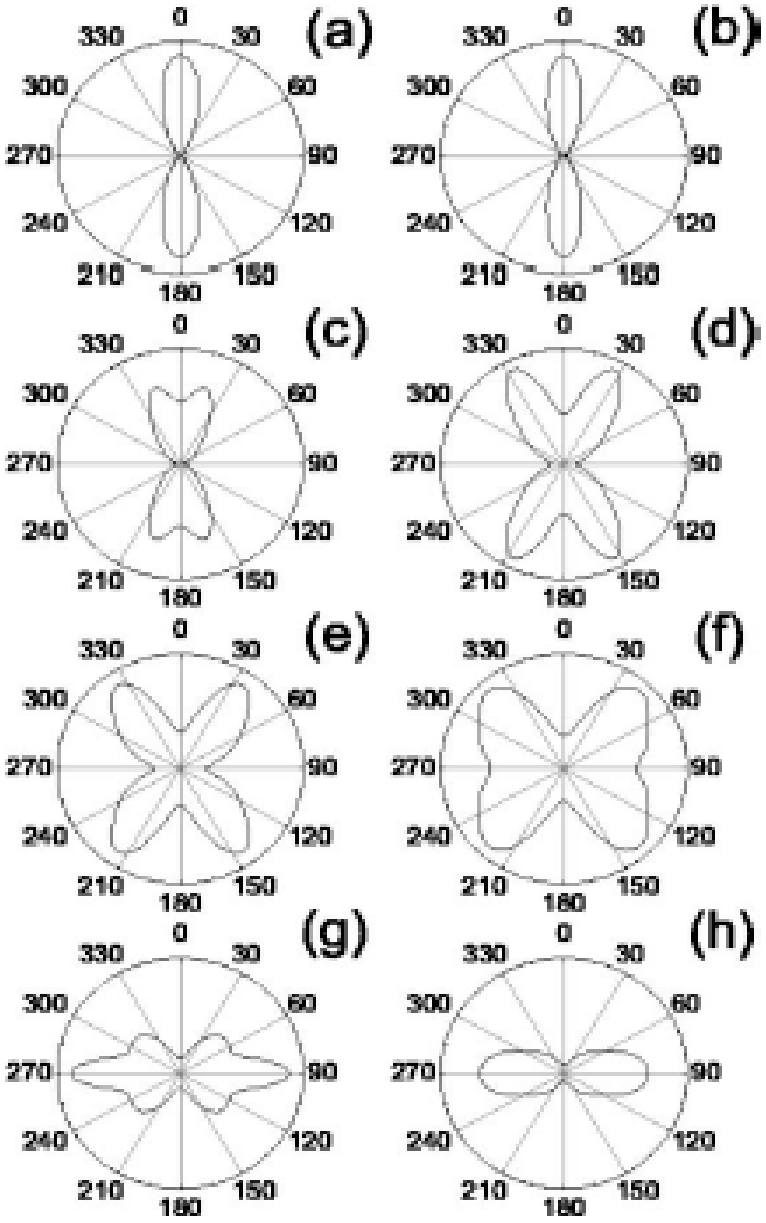
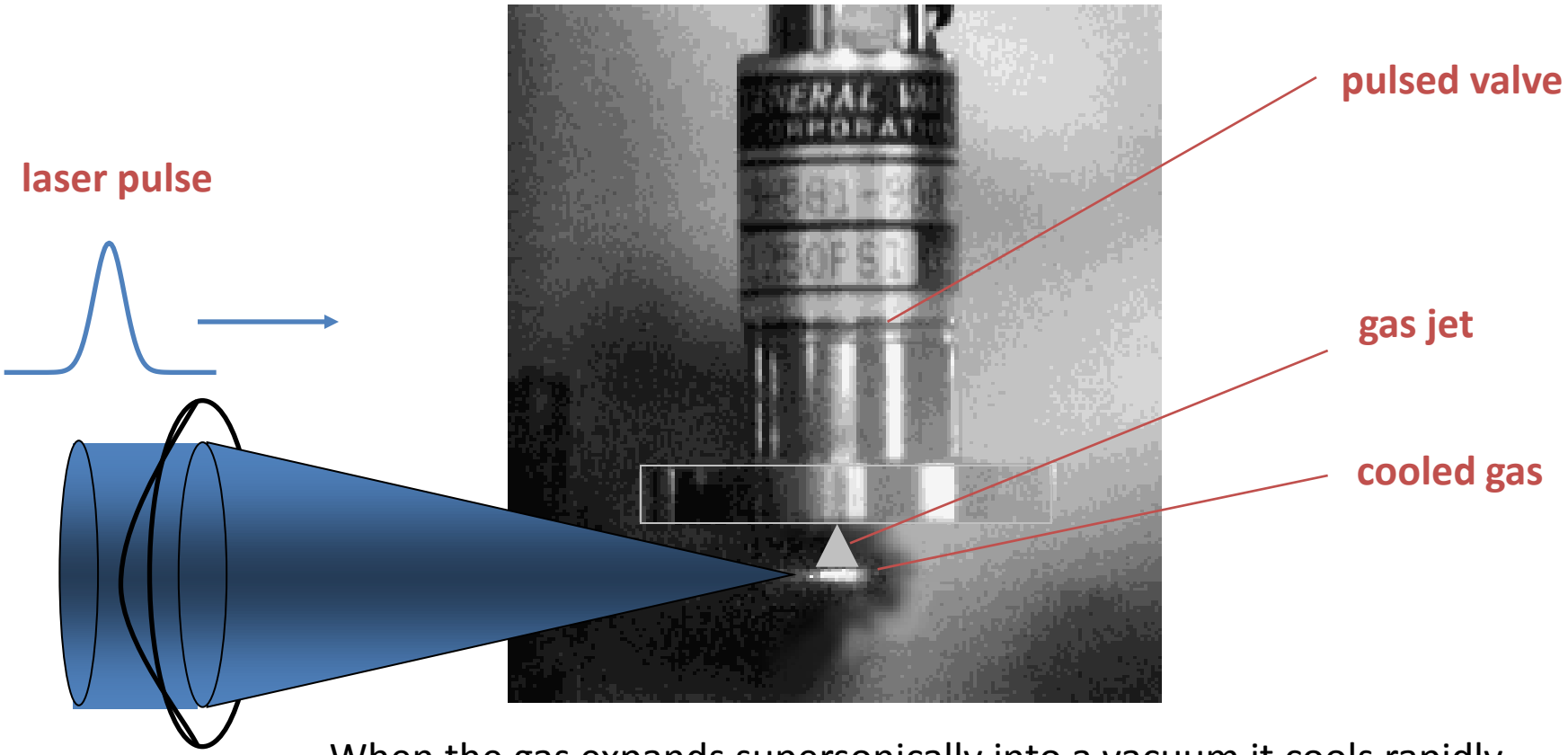


FIG. 5. Angular distribution of an ensemble of N_2 at 50 K. (a) Around the full revival time after the interaction with a laser pulse of 50 fs duration and $I_0=2.5\times10^{13}$ W/cm^2 peak intensity. (b) During the interaction with a 50 ps and $I_0=2.5\times10^{12}$ W/cm^2 laser pulse.

Require rotational cooling of molecules



When the gas expands supersonically into a vacuum it cools rapidly with collisions causing the translational cooling to couple to rotational cooling.

Nozzle diameter d is typically 100 – 500 microns and experiments are performed at a scaled distance x/d: x/d = 5 - 100

Technical Issues – Molecular Alignment

- Laser intensities ~ $10^{13}Wcm^{-2}$ for impulsive, but ionisation must be avoided, pulse duration must be optimised
- Adiabatic requires ~$10^{12}Wcm^{-2}$ and a transform limited pulse (injection seeding)
- Cooling often limited by cluster formation (molecules sticky)
- Only linear and symmetric top molecules will give alignment revivals – in general only partial alignment possible for the densities needed in HHG as cooling limited
- Many molecules too symmetric or too floppy or sticky to align at all

Tools 3:
Even shorter pulses

Self-phase modulation (SPM) arises from the third order non-linear response

Non-linear refractive index n_2

$$n \approx n_0 + \frac{\chi_3 I}{2n_0} = n_0 + n_2 I$$

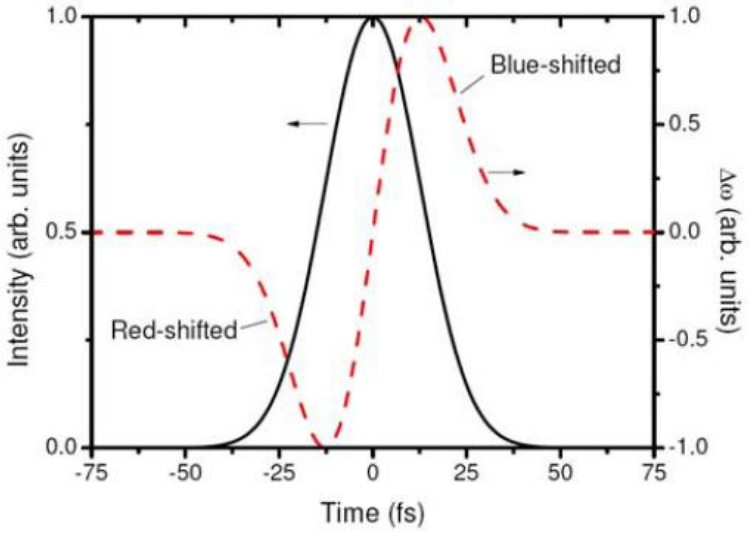
The resulting time dependence of the phase:

$$\phi(t) = \omega_0 t - \frac{2\pi n}{\lambda} L = \omega_0 t - \frac{2\pi \left(n_0 + n_2 I(t)\right) L}{\lambda}$$

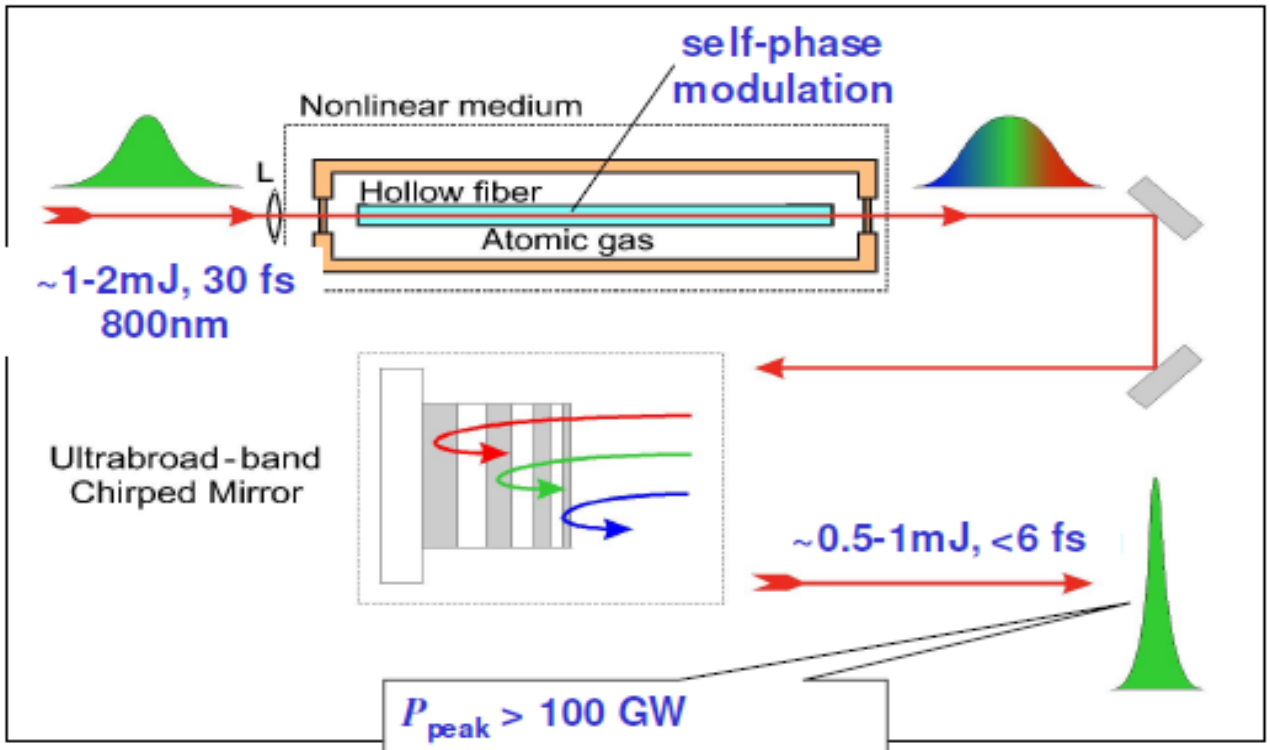
Leads to a time dependent frequency,

$$\omega(t) = \frac{d\varphi(t)}{dt} = \omega_0 - \frac{2\pi n_2 L}{\lambda} \frac{dI}{dt}$$

That may give rise to appreciable additional bandwidth.



Shortest High Power Laser Pulses Generated by SPM in Hollow Fiber & Pulse Compression: Few Cycle and CEP Stable

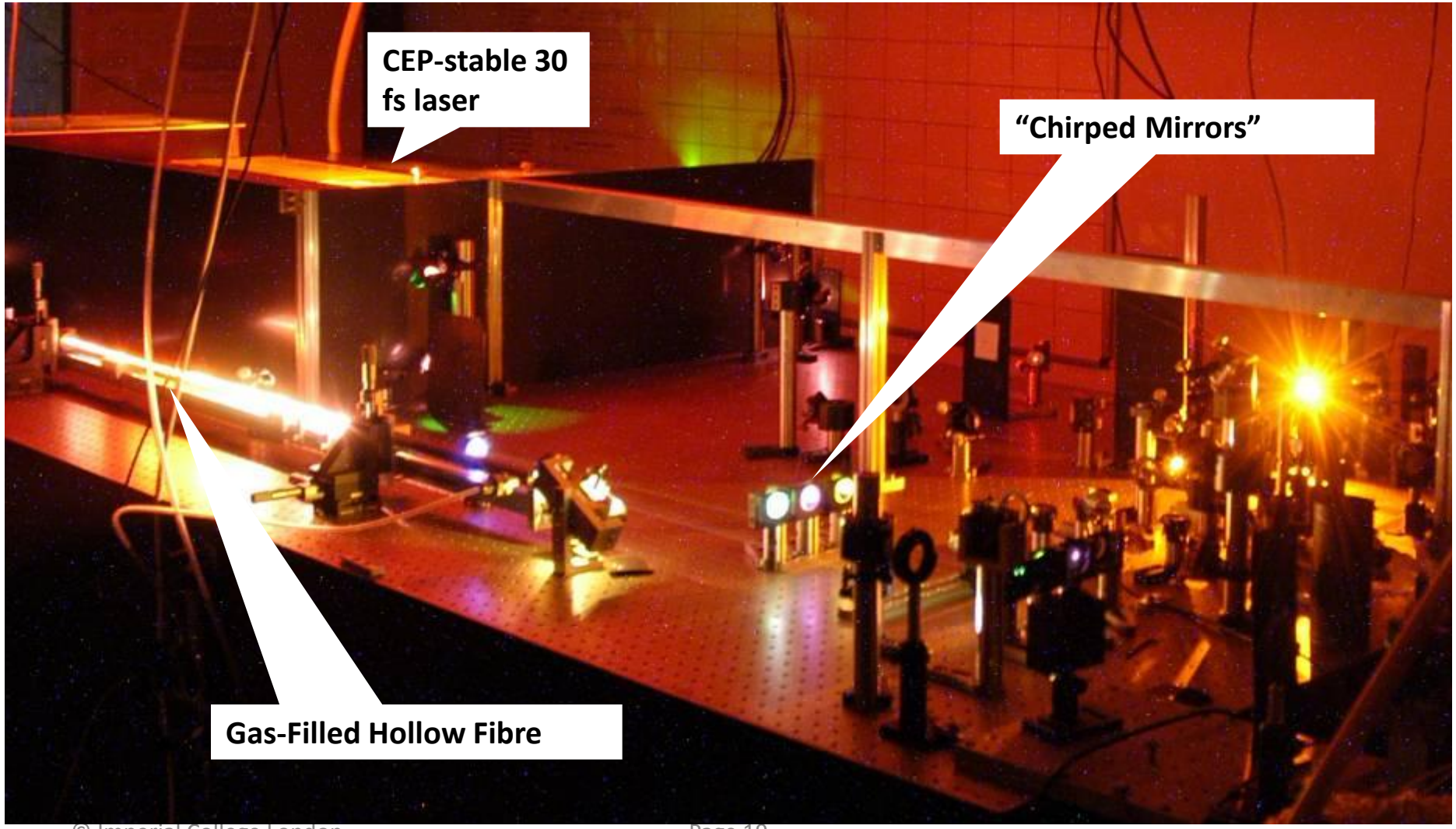


M. Nisoli et al. APL 68, 2793 (1996)

Filamentation provides another route C.P. Hauri et al. APB 79 673 (2004)

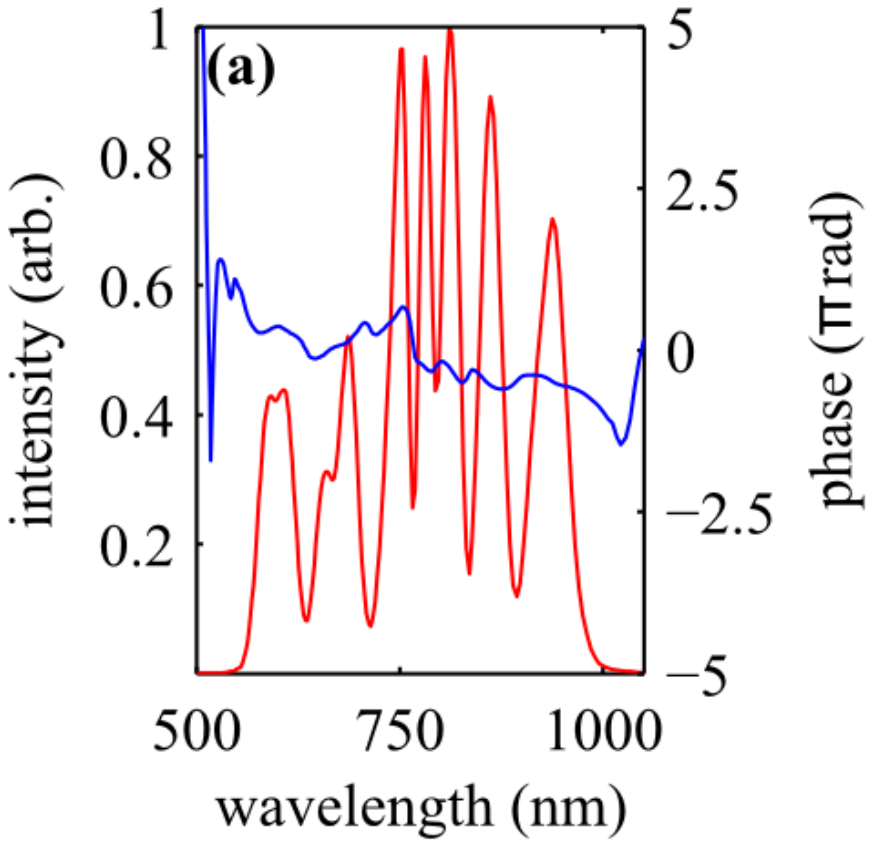
<4 fs at 800nm have been achieved (< 2 optical cycles)

Imperial College few-cycle source 0.4mJ/3.5fs min. CEP-stable

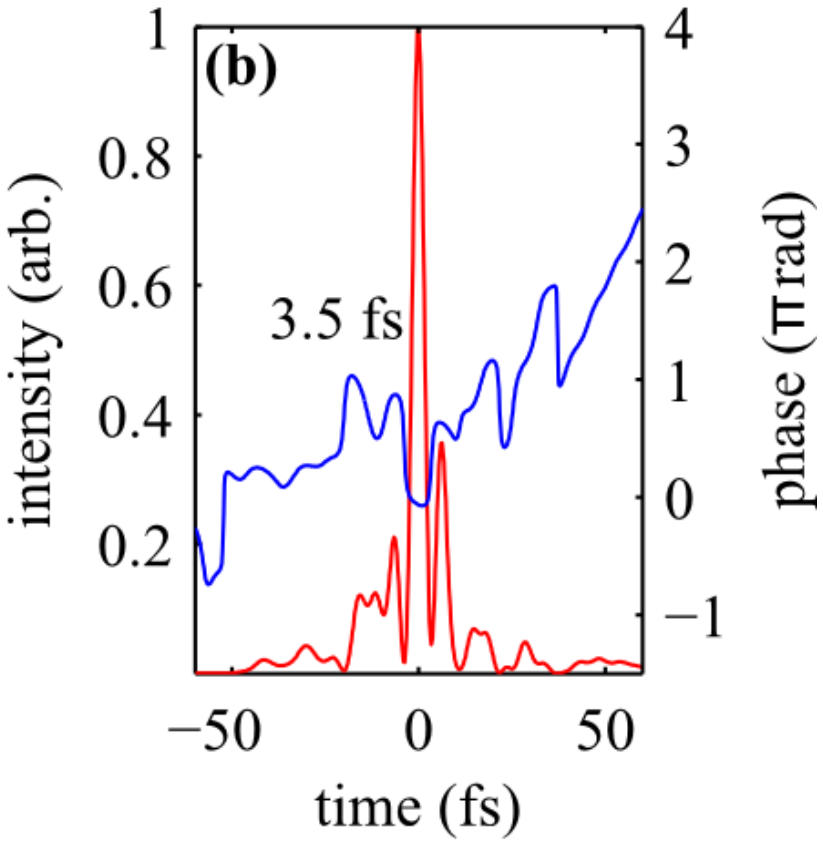


Sub 4 fs, 100 GW pulses at Imperial College

Spectrum & spectral phase



Pulse & temporal phase



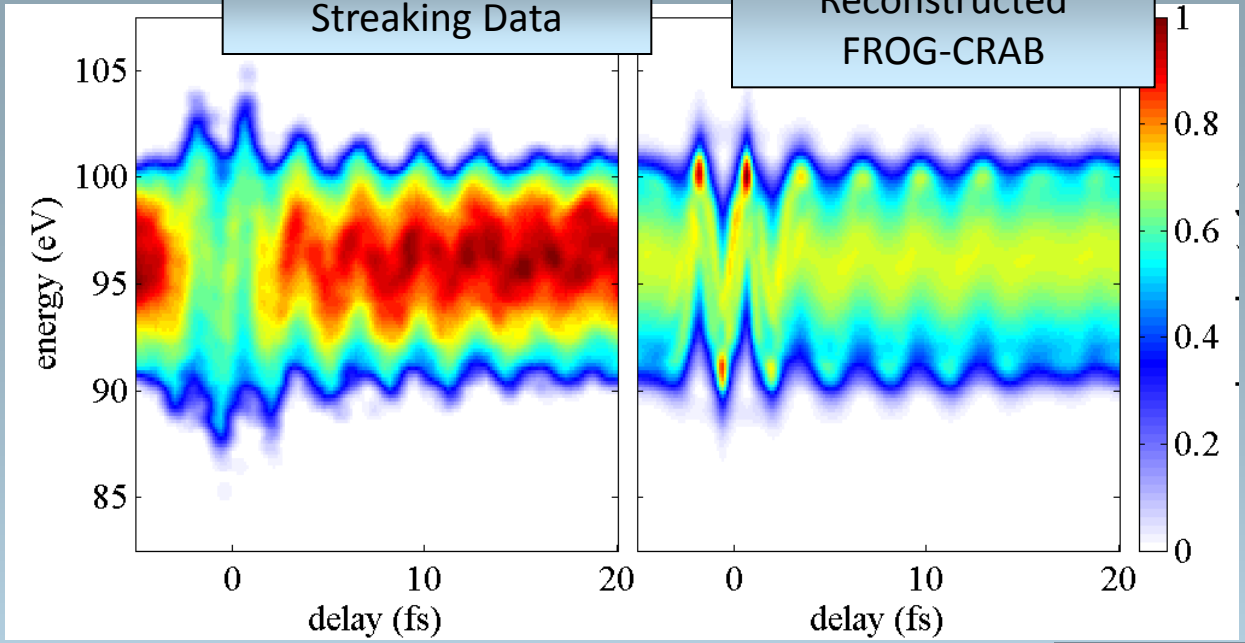
Shortest, fully-characterised pulse in the world!

T. Witting et al. Opt. Lett. 361680 (2011)

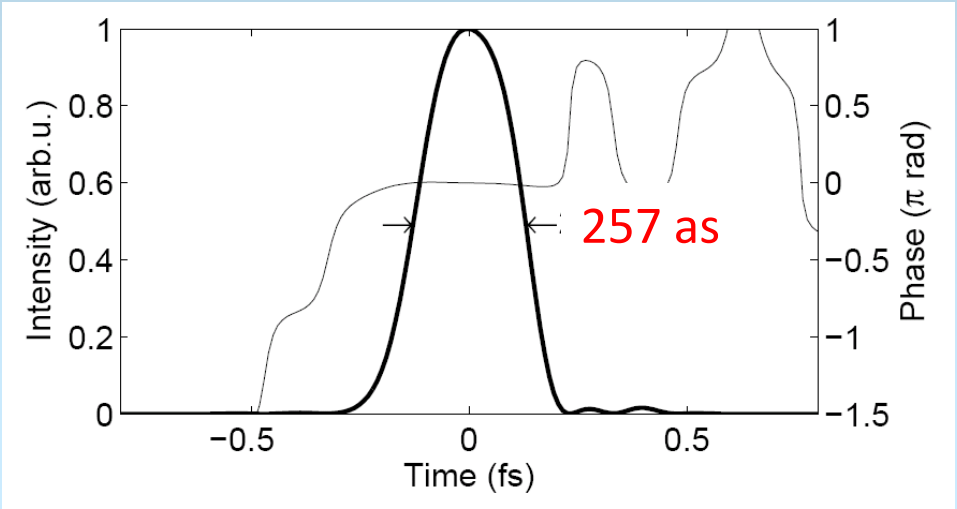
Attosecond Streaking Results

Streaking Data

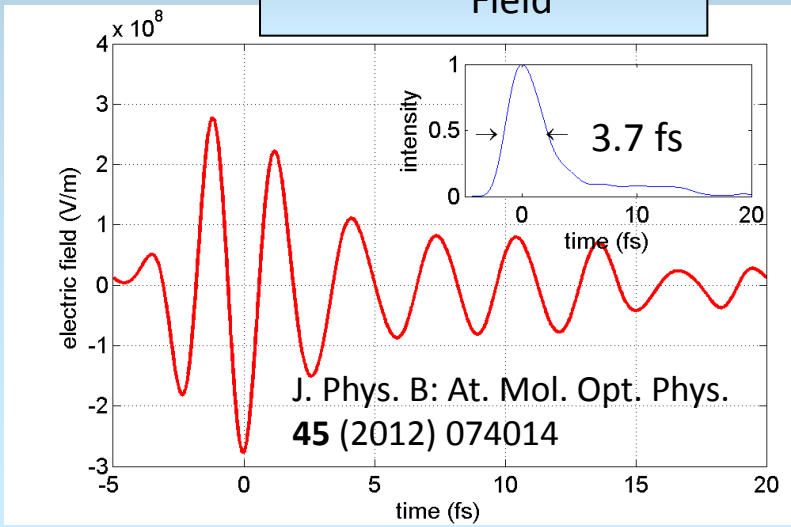
Reconstructed FROG-CRAB



Retrieved Attosecond Pulse



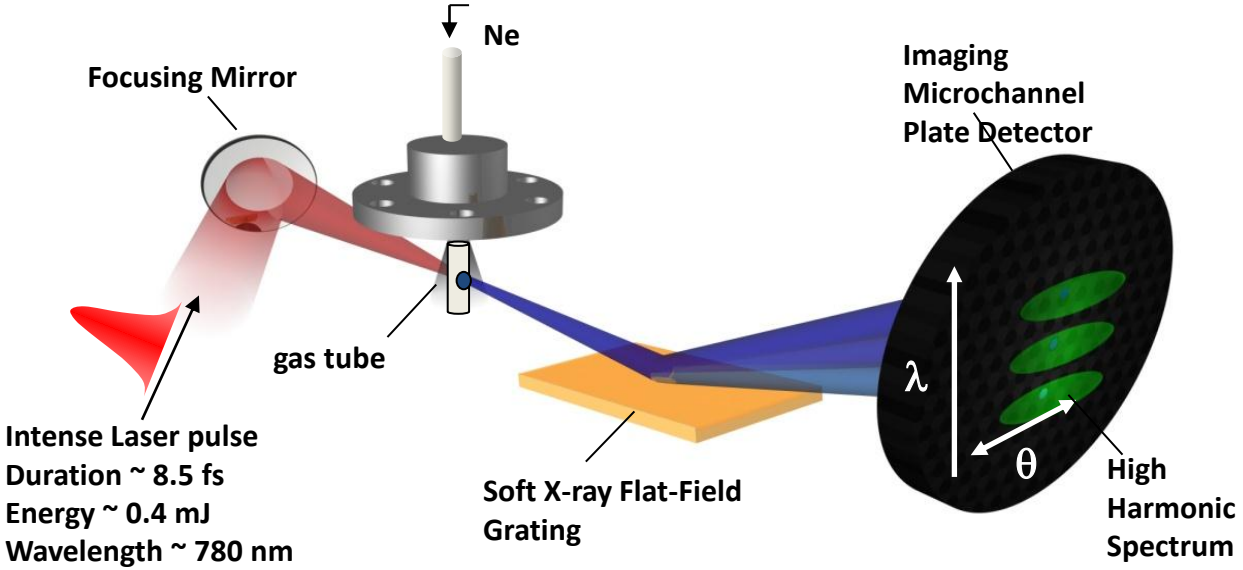
Reconstruction of Laser Field

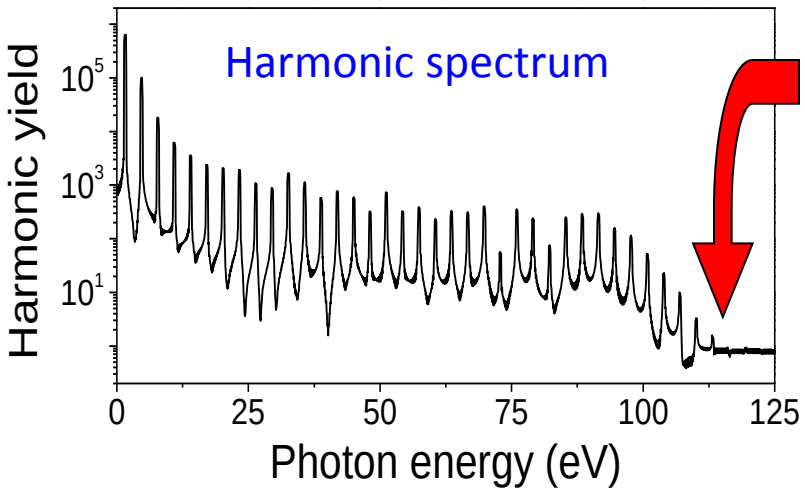


Technical Issues – Even Shorter Pulses

- Dispersion control of large bandwidths (several 100nm) very challenging
- Require diagnostics to give full amplitude/phase information (SPIDER or FROG)
- CEP stabilisation/control required for few cycle lasers
- Controlled environment – extreme vibration sensitivity
- Few-cycle pulses very fragile – duration doubled if it passes through ~1mm of glass, all optical components must be compensated

Tools 4:
High harmonic generation





Cutoff

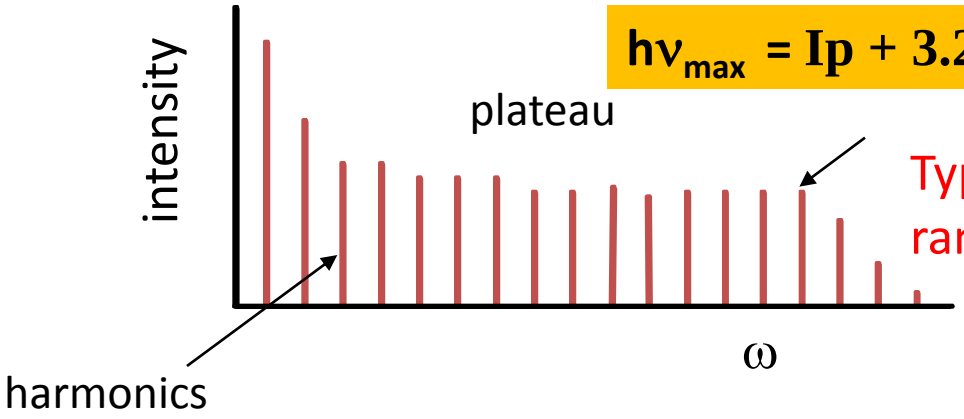
$$E_{photon} = I_P + 3.17\ U_P$$

$$U_P = \frac{1}{m}\left(\frac{eE_0}{2\omega}\right)^2$$

4 x 10^{14} Wcm^{-2} @ 800nm
U_p ~ 25 eV

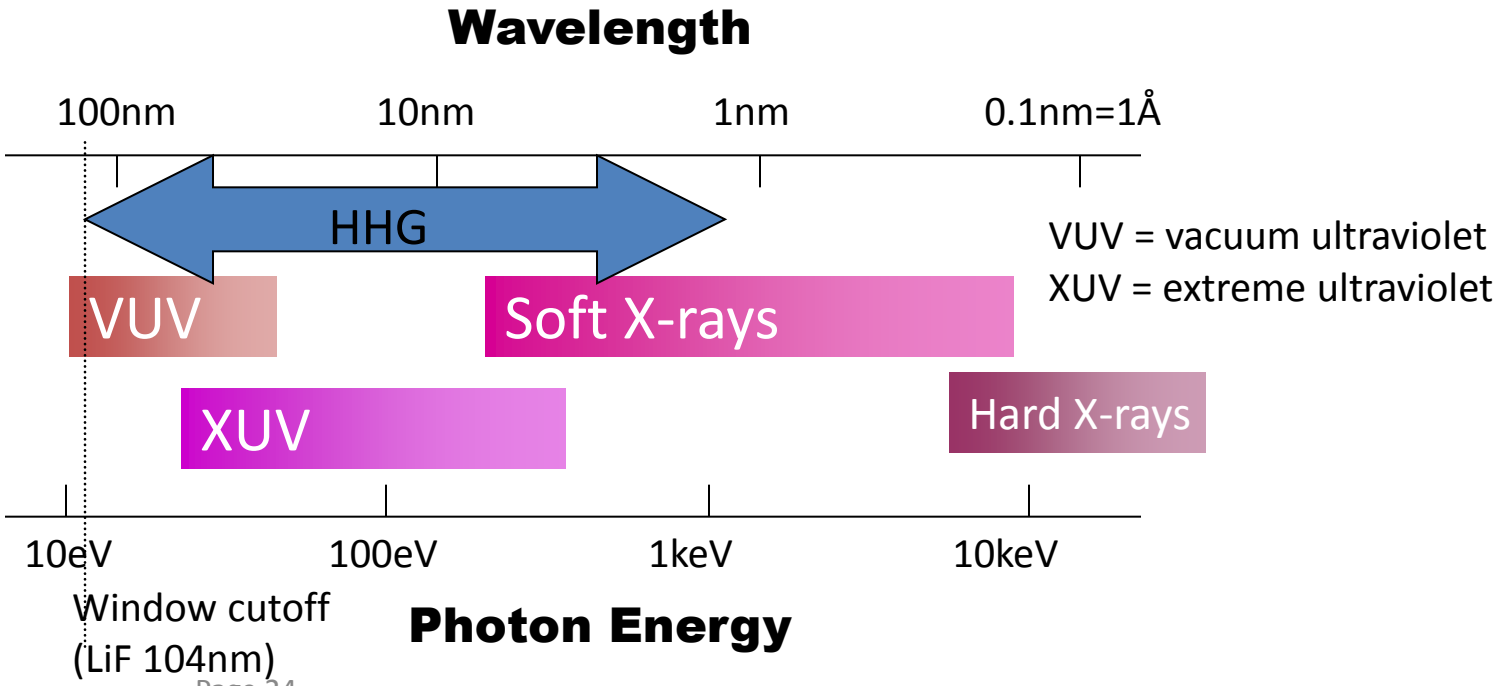
~100eV bandwidth coherent spectrum !

Wavelength range accessible with HHG



$h\nu_{max} = Ip + 3.2\ U_P$

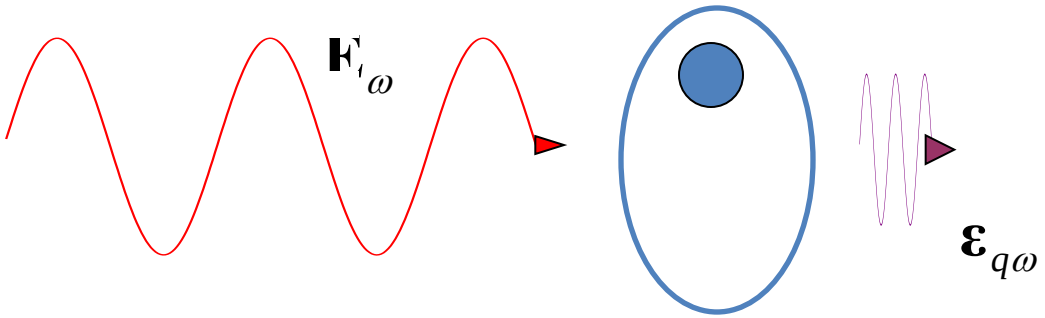
Typically 1-10nJ per pulse in 10-30nm range at 1-3 kHz rep-rates



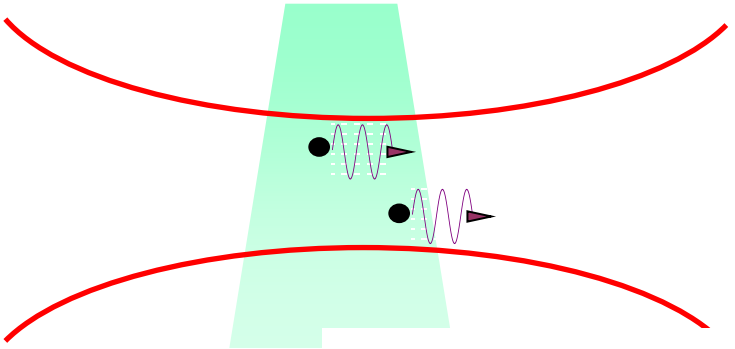
VUV = vacuum ultraviolet
XUV = extreme ultraviolet

Micro/Macroscopic aspects of HHG emission

Microscopic response: atom in strong laser field



Macroscopic response: coherent superposition of individual fields



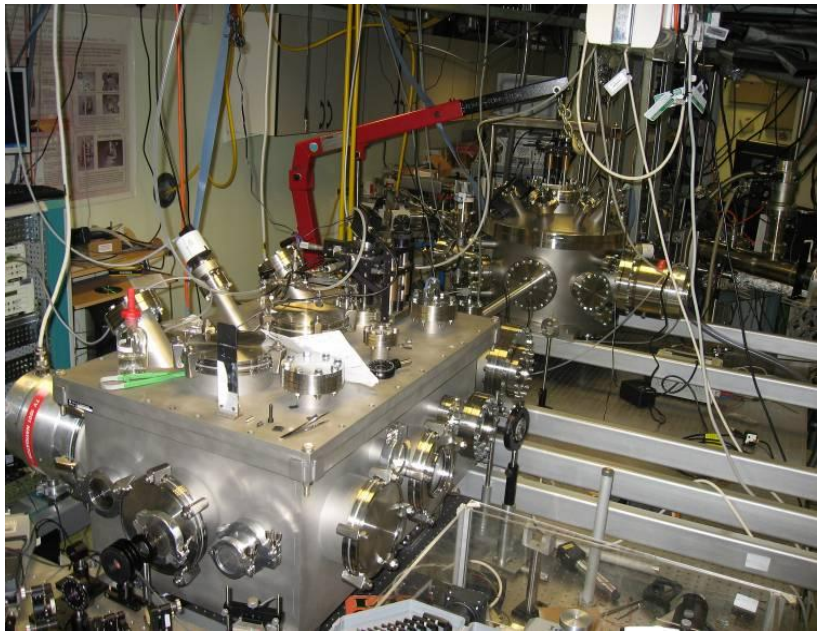
$$\mathbf{E}_{q\omega} = \sum_{atoms} \boldsymbol{\varepsilon}_{q\omega}$$

phase-matching

HHG has both longitudinal and transverse coherence

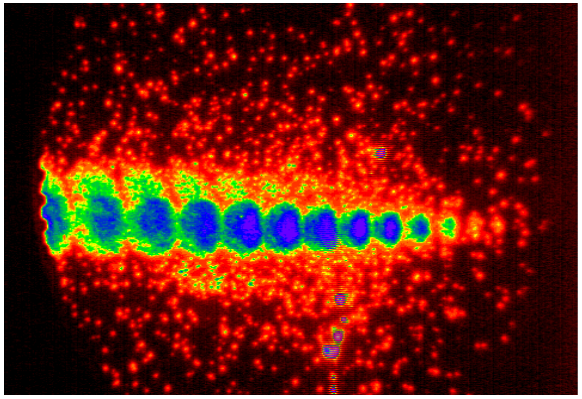
A compact, coherent, ultrafast XUV source.

HHG beam-line at Imperial College



Properties of High Harmonic Radiation

- high spatial coherence
- highly directional
- short wavelength (into 2-4nm “water window”)
- ultrafast (shorter than laser pulse – attosecond with few-cycle laser pulses)

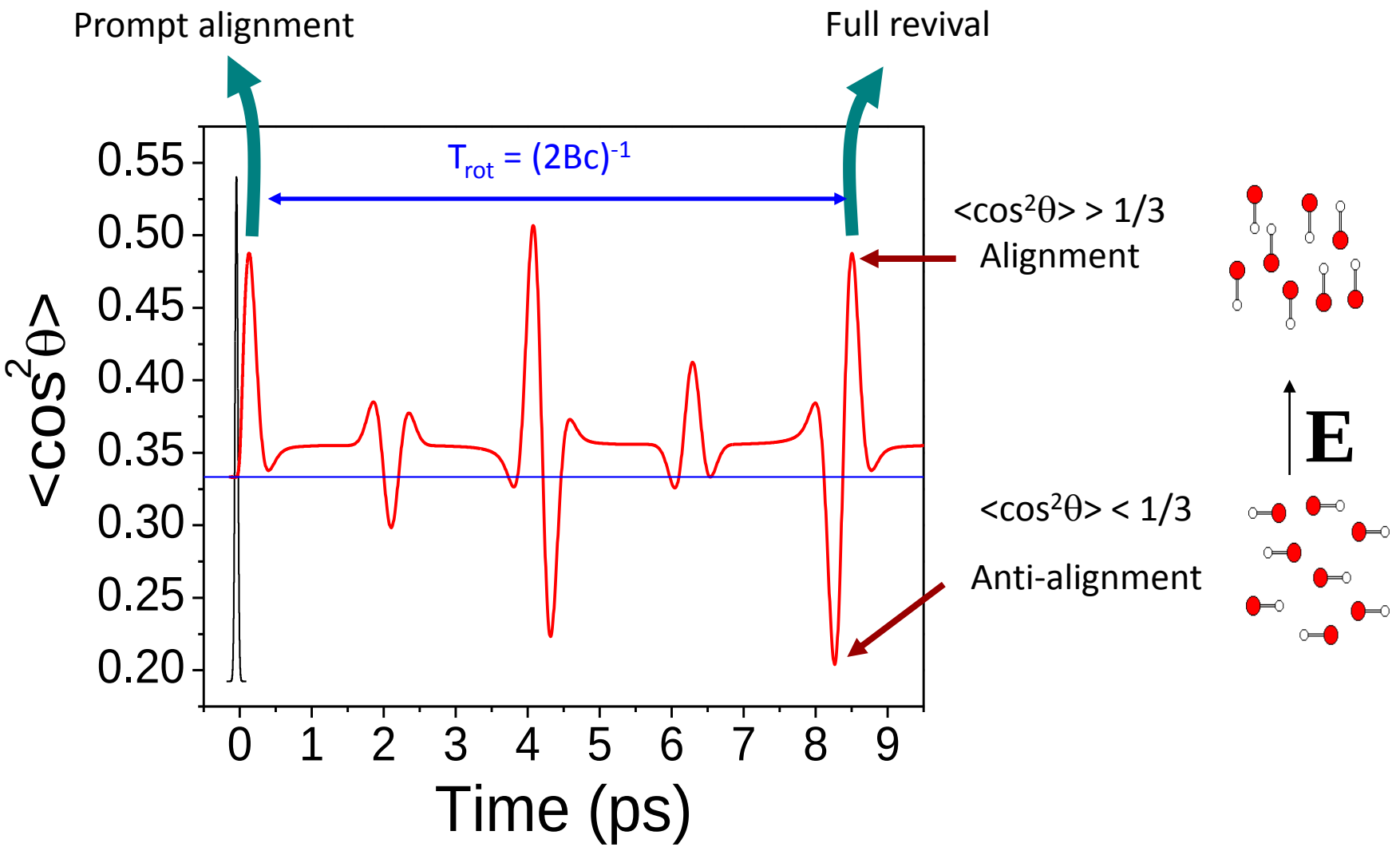


High Order Harmonics Spectrum

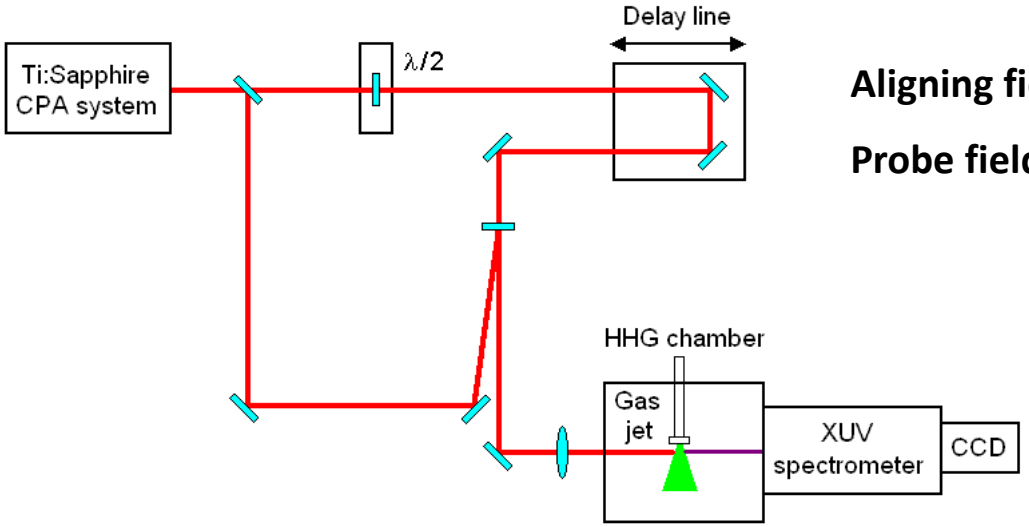
Technical Issues – HHG

- Soft X-rays need a vacuum!
- Beams invisible and hard to diagnose what's going on
- Detection requires specialist detectors (MCP's or X-ray CCDs) that are easy to damage, have limited dynamic range and degrade with time
- Difficult to maintain conditions for optimum generation (alignment, gas jet issues, burnt optics etc.)
- Process very inefficient – only nJ or pJ per pulse
- High order nonlinearity so laser fluctuations greatly increased in HHG signal
- Need spatially confined gas targets – solids and liquids must be vaporised by special methods (ablation plume, heated samples and delivery systems) into gas jet or differentially pumped tube

An Example Experiment: Molecular HHG sensitive to angular dependence of recombination matrix element

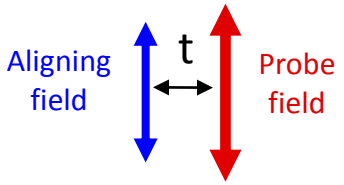


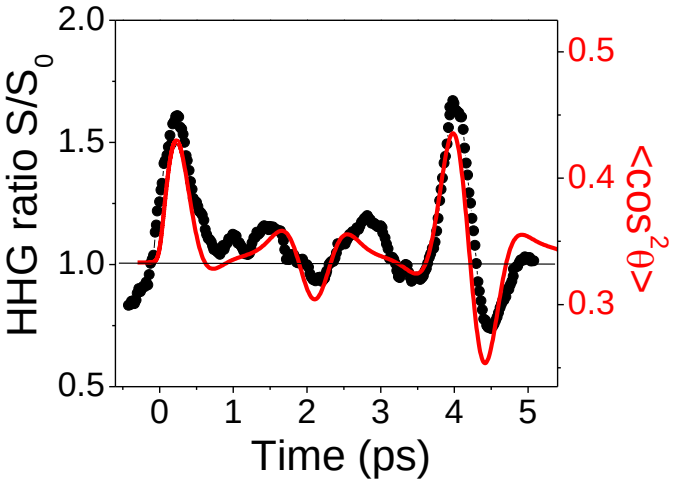
Basic experimental setup for HHG in aligned molecules

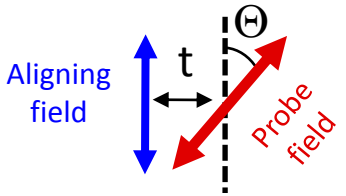


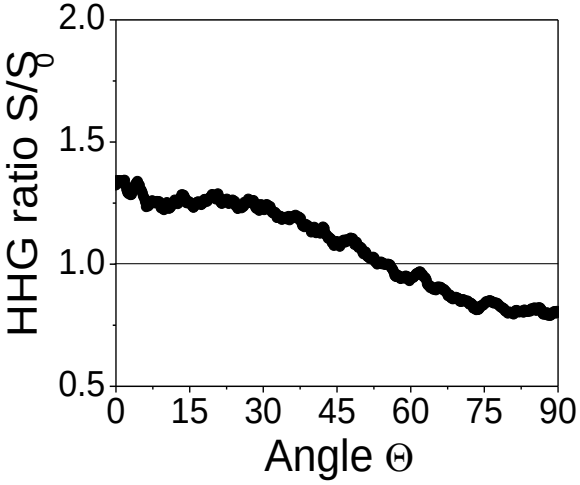
Aligning field: 800 nm, ~10^{13} W/cm^2, ~100 fs

Probe field: 800 nm, ~10^{14} W/cm^2, ~40 fs









Tomographic reconstruction of orbitals

HHG spectra from N_2 aligned at different angles

Tomographic reconstruction

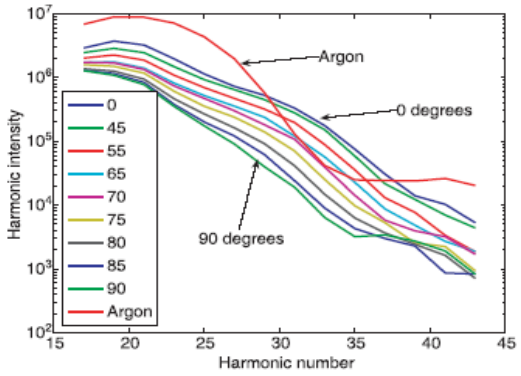
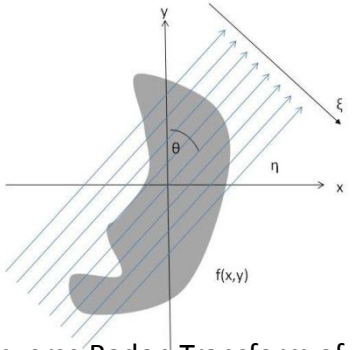
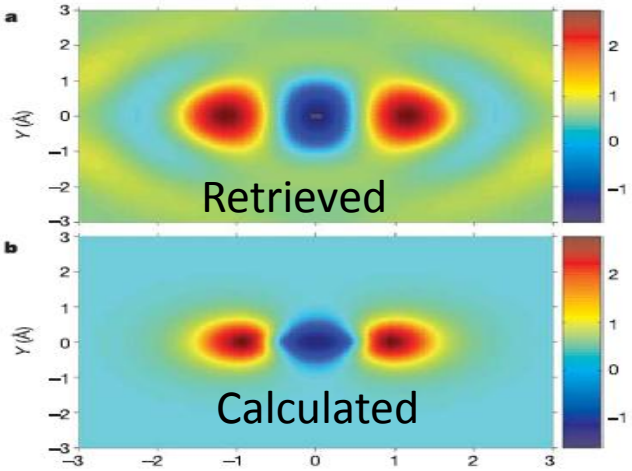


Figure 3 High harmonic spectra were recorded for N_2 molecules aligned at 19 different angles between 0 and 90° relative to the polarization axis of the laser. For clarity, only some of the angles have been plotted above. The high harmonic spectrum from argon is also shown; argon is used as the reference atom. Clearly the spectra depend on both the alignment angle and shape of the molecular orbital.



Inverse Radon Transform of extracted dipole amplitude and phase



- Reconstructed orbital is HOMO + contribution from lower lying orbitals.

J. Itatani *et al.*, Nature 432, 867 (2004)

S. Patchkovskii *et al*., PRL 97, 123003 (2006)

Tools 5:
Complete characterization of the harmonic electric field

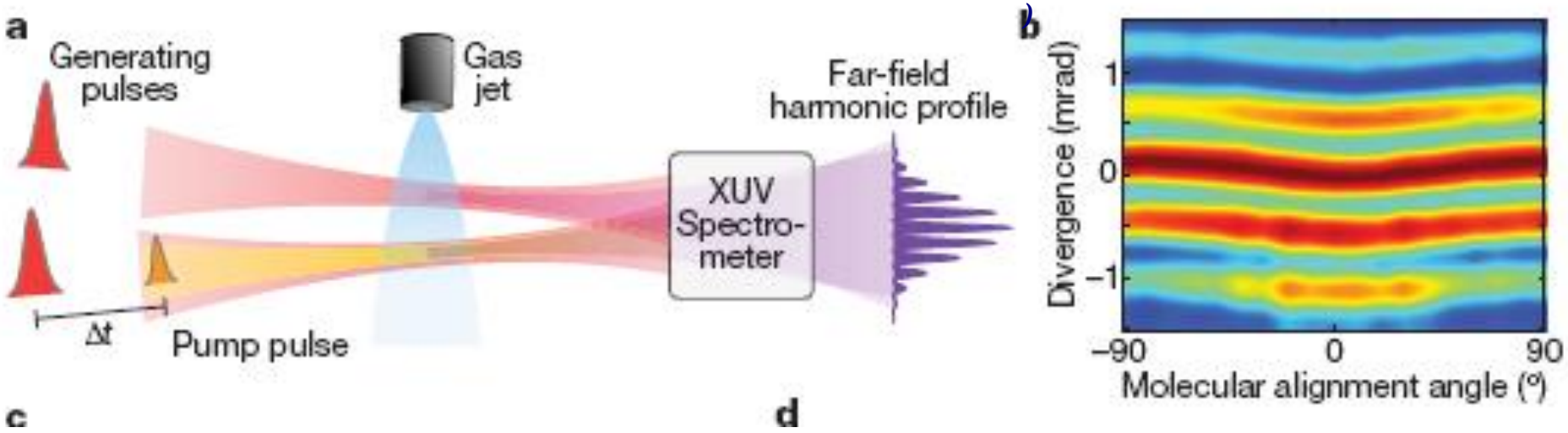
Spectrum

Phase

Polarization (angle and ellipticity)

Measuring HHG Phase: Example Interferometry

O. Smirnova et al.,Nature 460, 972 (2009



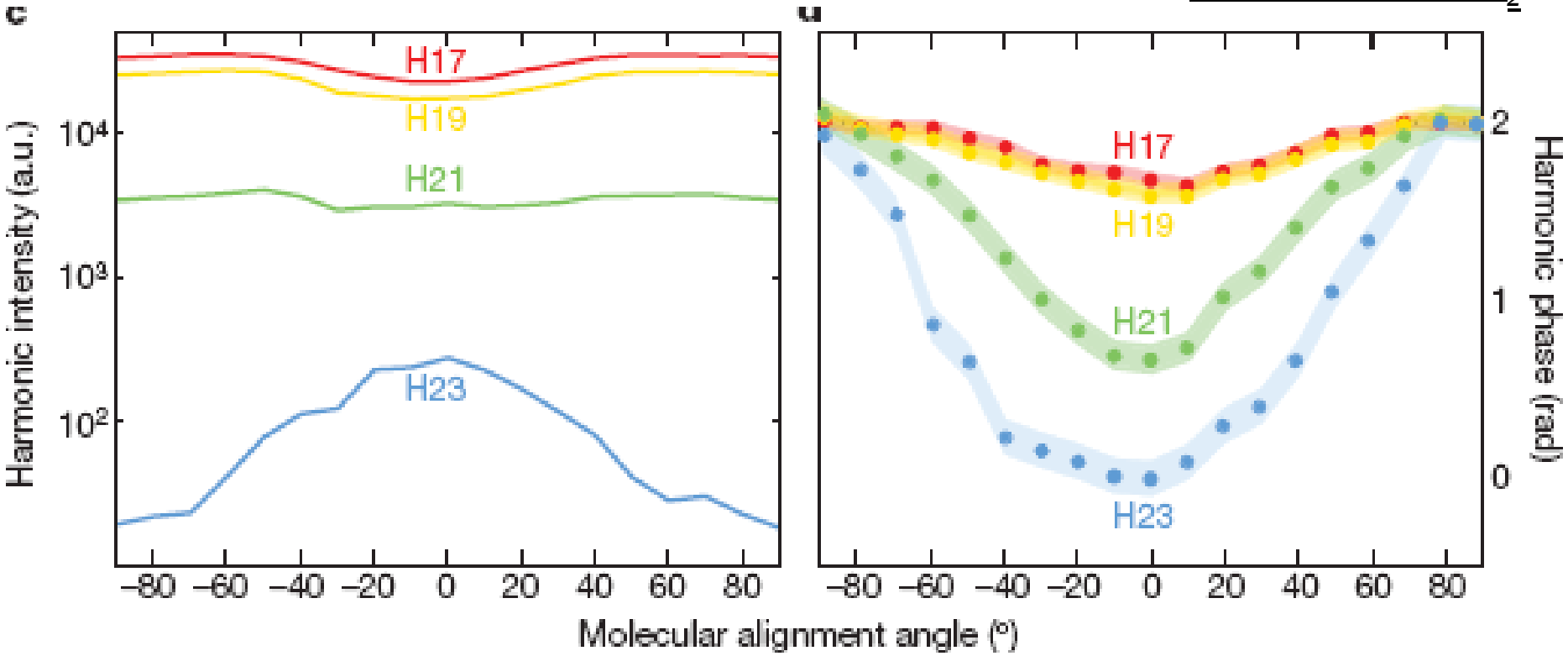
Technical difficulty: need good stability between the two arms of the interferometer

Harmonic q optical period: T_q=2.7fs/q (130 as for H21)

Courtesy Yann Mairesse

Harmonic amplitude and phase vs alignement

Mesurement in CO_2



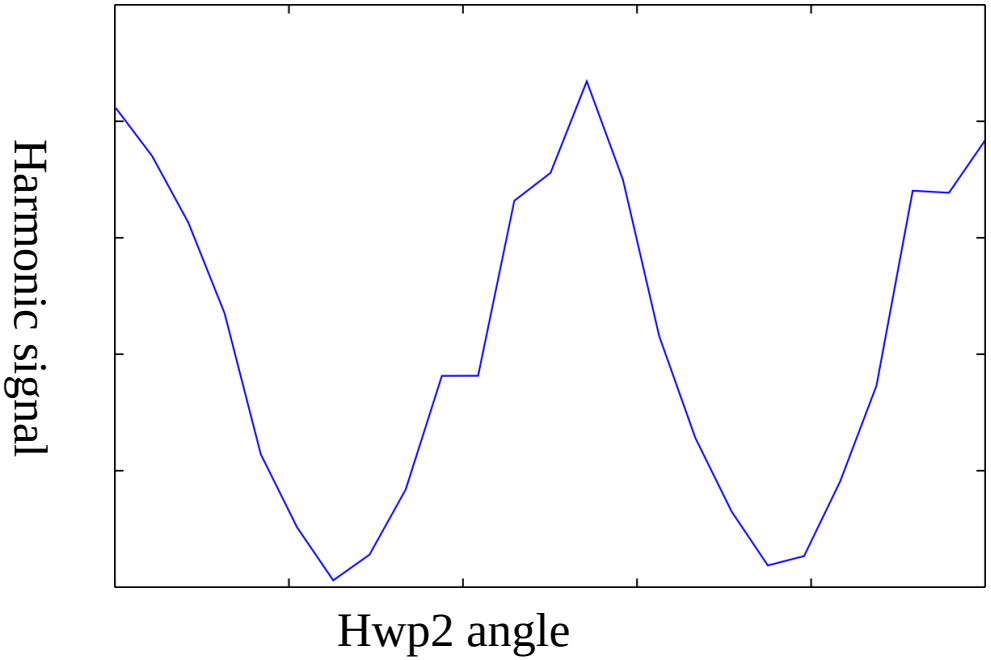
Courtesy Yann Mairesse

Polarimetry

Principle:

Send harmonic radiation to polarization analyzer
= rotating polarizer

J. Levesque et al., PRL 99, 243001 (2007)
Y. Mairesse et al., Nex J. Phys. 10, 025028 (2008)
X. Zhou et al., PRL 102, 073902 (2009)
Y. Mairesse et al., PRL 104, 213601 (2010)



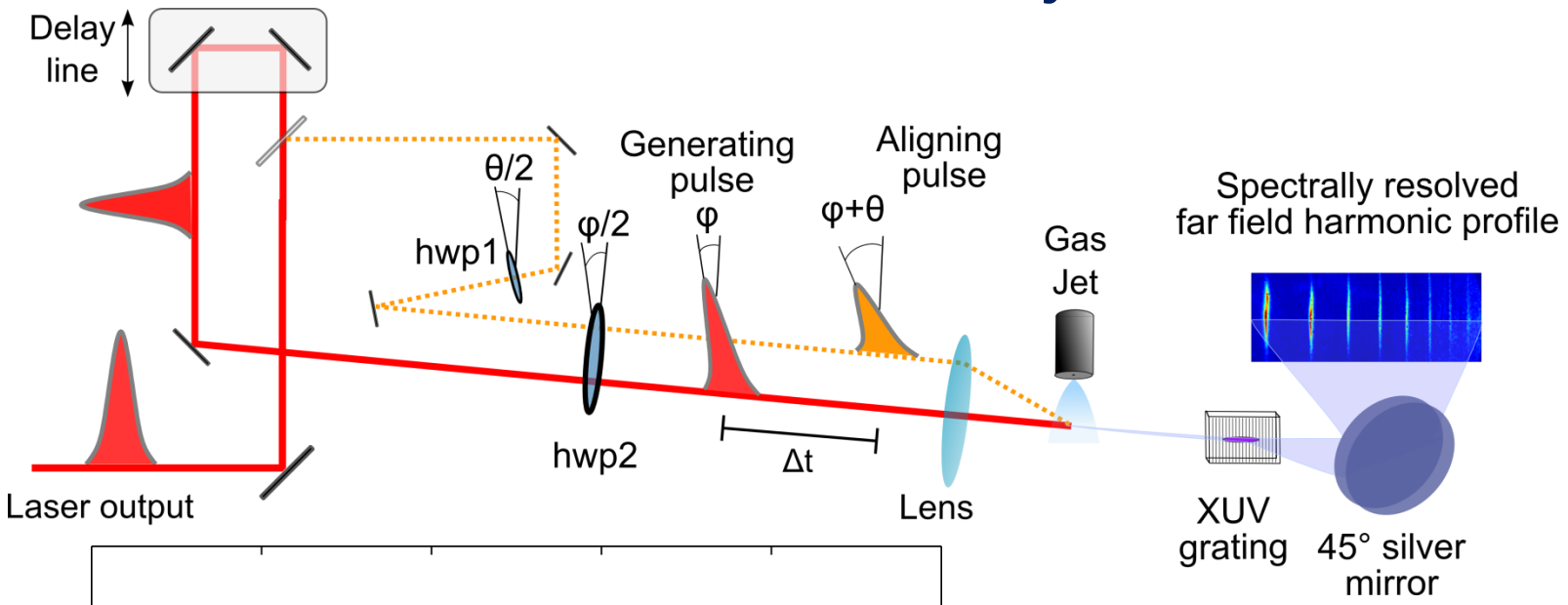
Malus law:

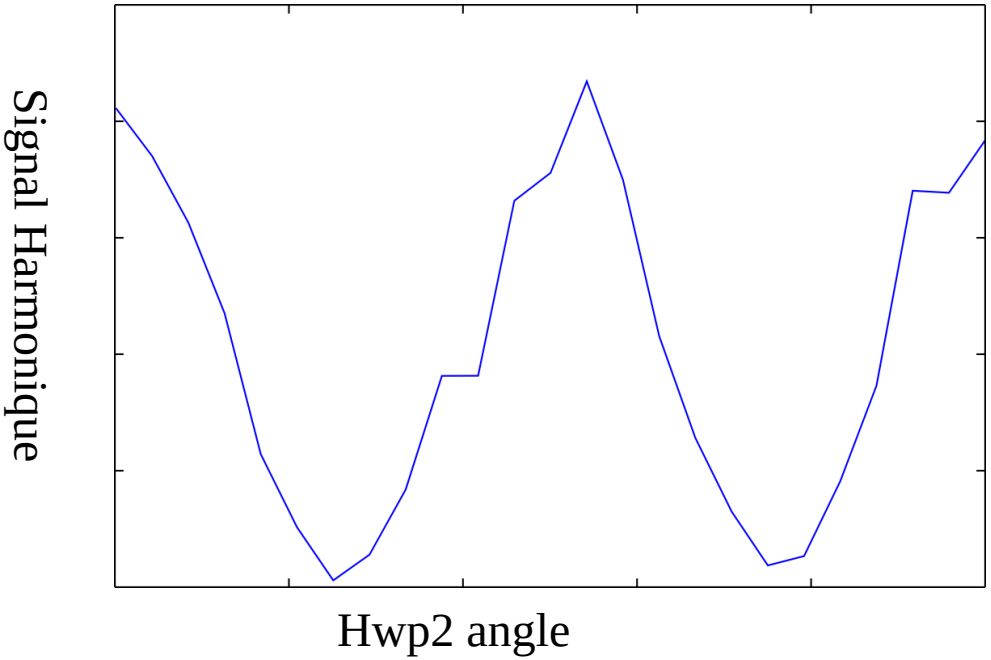
The phase of the oscillation gives the polarization direction

The contrast gives the ellipticity

Courtesy Yann Mairesse

Polarimetry





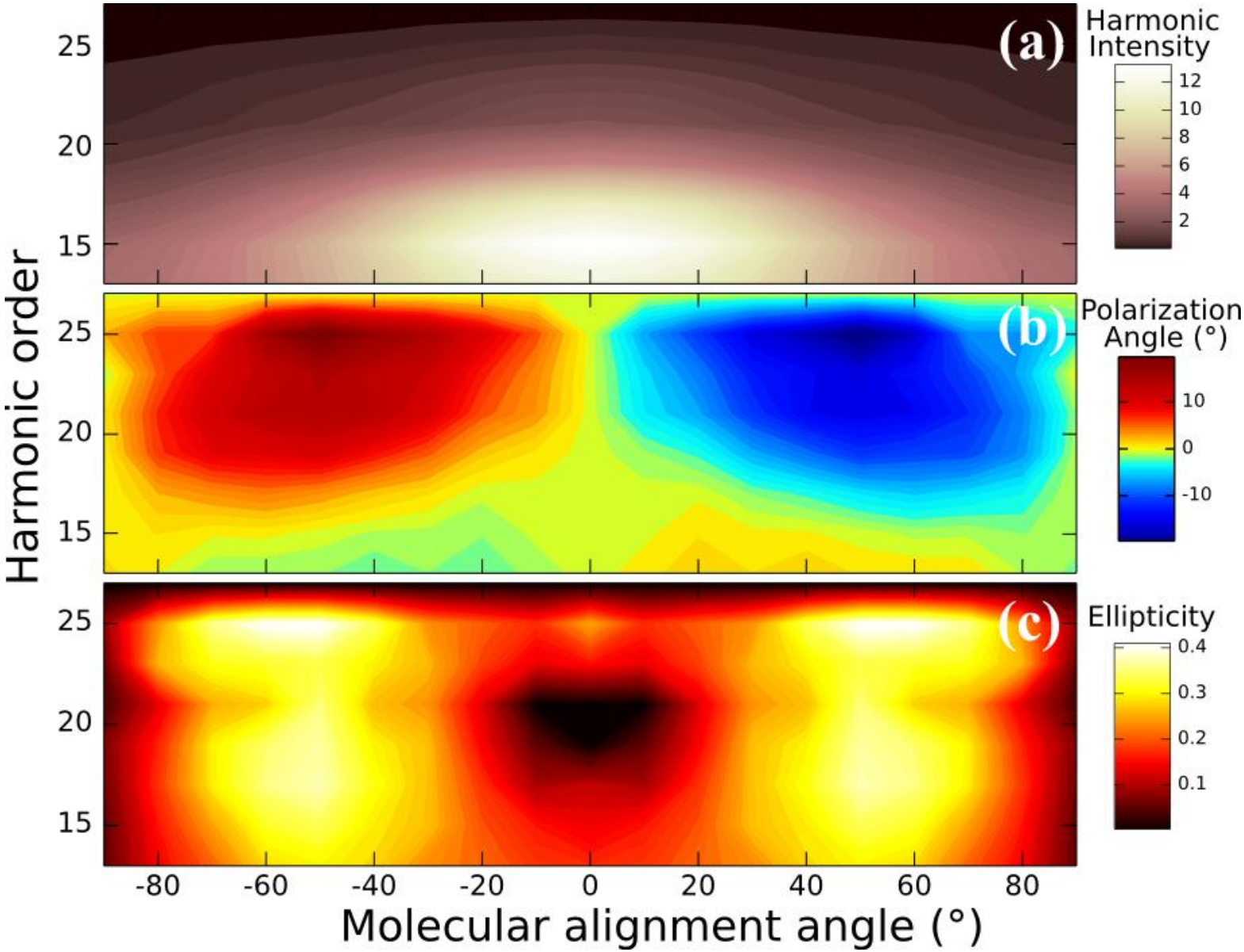
Malus law:

The phase of the oscillation gives the polarization direction

The contrast gives the ellipticity

Courtesy Yann Mairesse

Example: polarization map in aligned N2

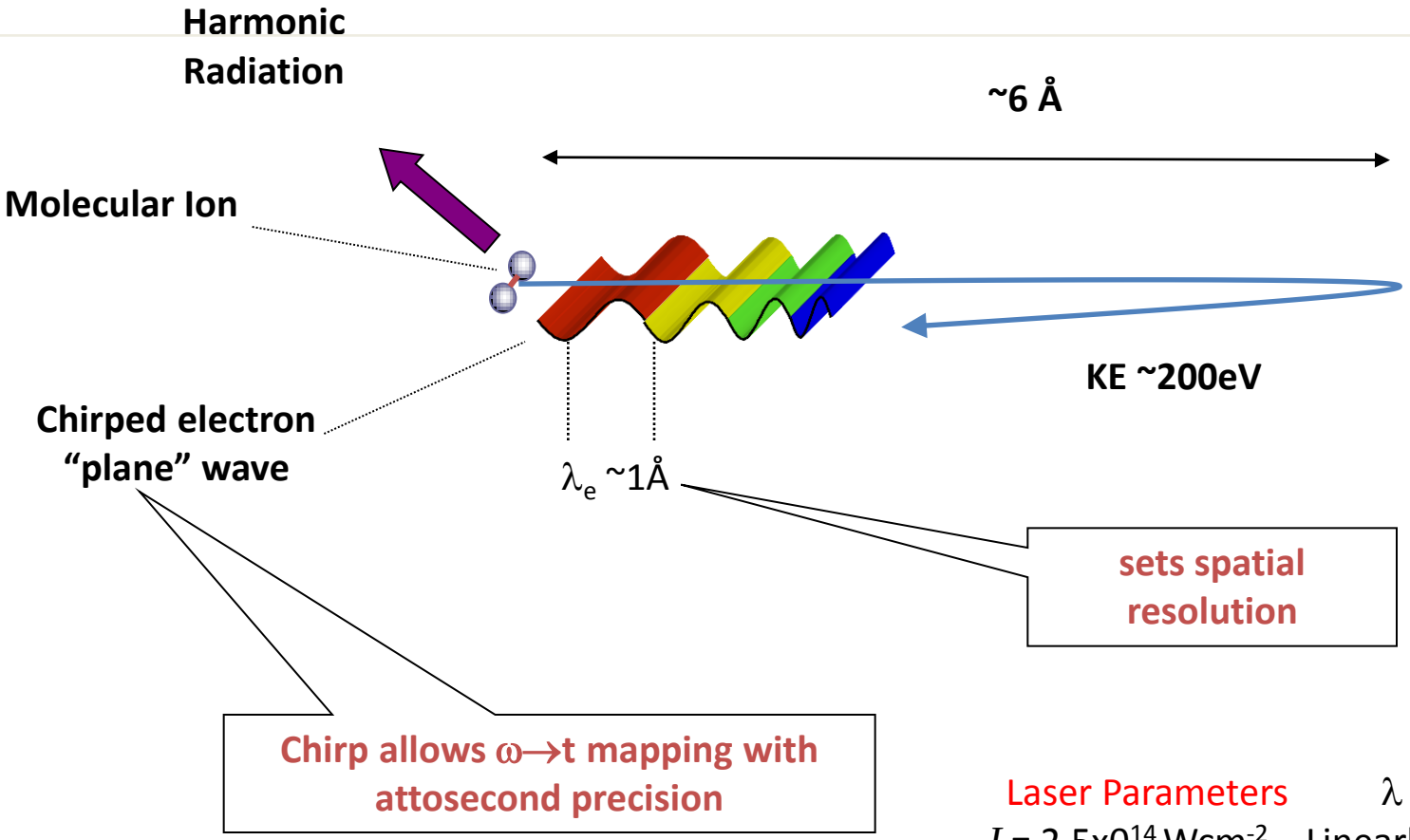


Y. Mairesse et al., PRL 104, 213601 (2010)

Technical Issues – Full Characterisation of HHG

- Lack of soft X-ray optics make these set ups tricky and require special precautions
- So far mostly only implemented for “standard” 800nm drive lasers
- Most phase methods invasive – split beams so loose power
- Hard to disentangle propagation phase from single molecule phase (often hard to account for all sources of phase shift) – in general phase matching can have a major effect upon the spectrum and phase (this is too often ignored)
- Measurements require highly stable set-up and photon hungry (to get good S/N)
- In practice a full characterisation is rarely done (so far only for a few molecules)

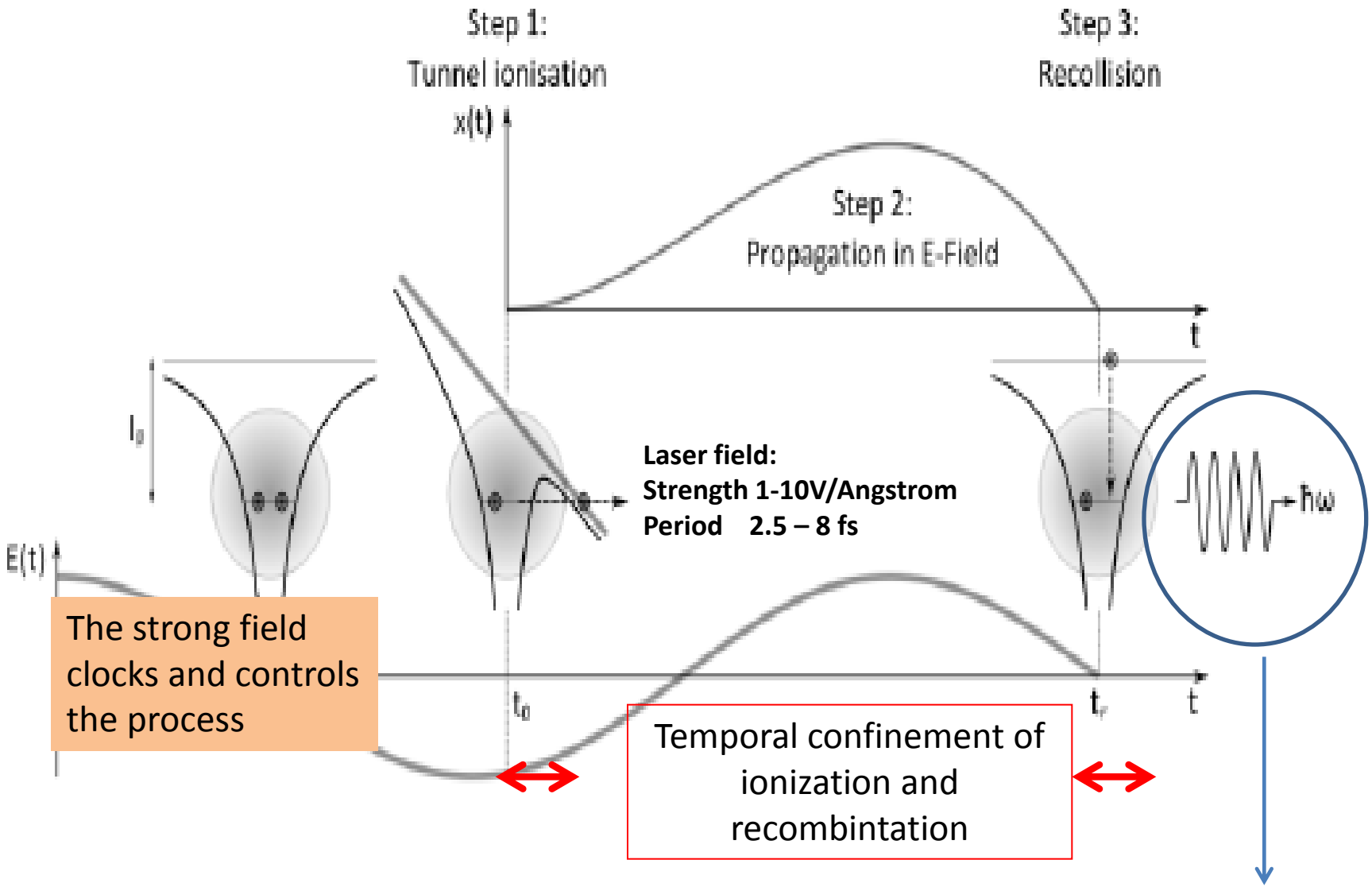
Tools 6:
Accessing Temporal Properties of HHG



Laser Parameters λ = 1600nm

I = 2.5x0^{14} Wcm^{-2} Linearly Polarised

High Harmonic Generation in the time domain: 3 step picture



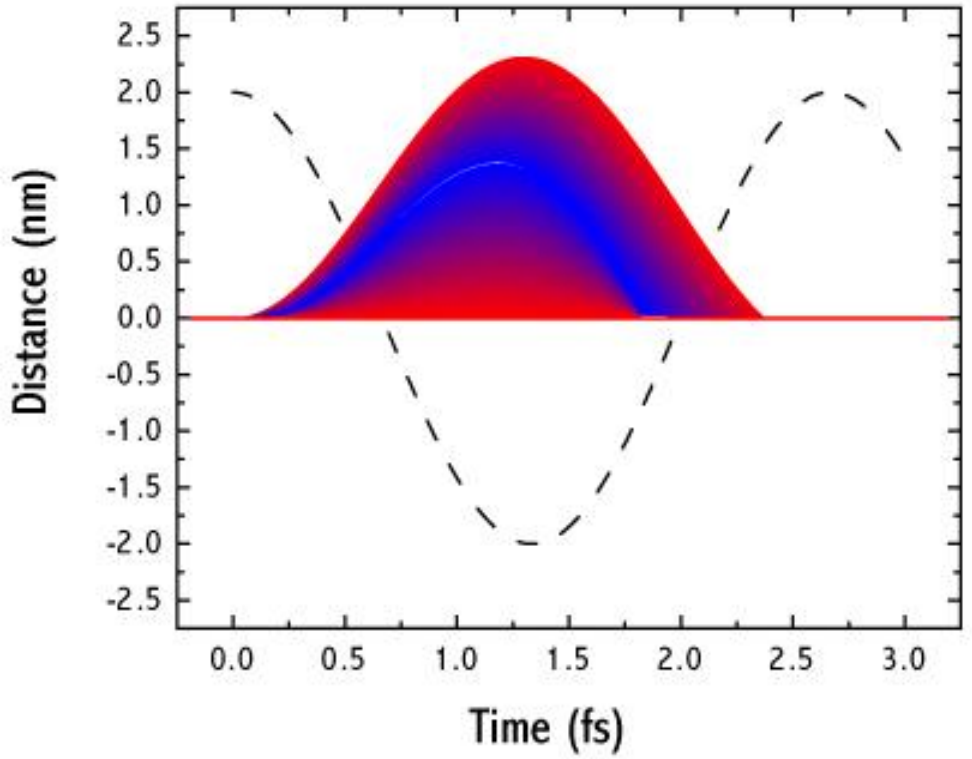
The strong field clocks and controls the process

Temporal confinement of ionization and recombintation

Temporal confinement of soft X-ray pulse emission provides tool for attosecond photoionization

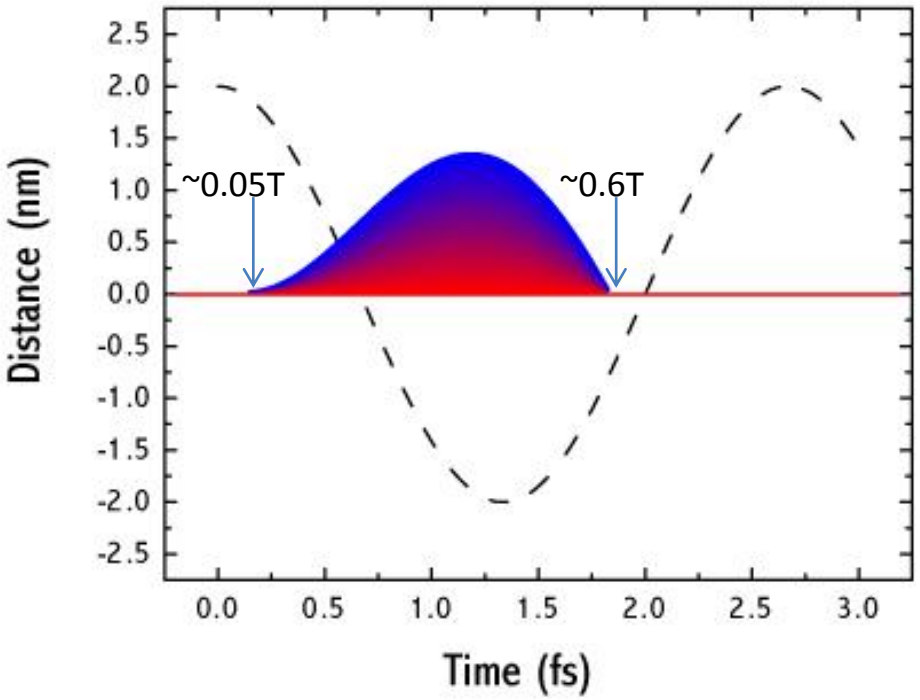
Temporal chirp of electron wavepacket

Temporal spread ~1 fs for a 800 nm driving pulse (depends on optical cycle)



Temporal chirp of electron wavepacket

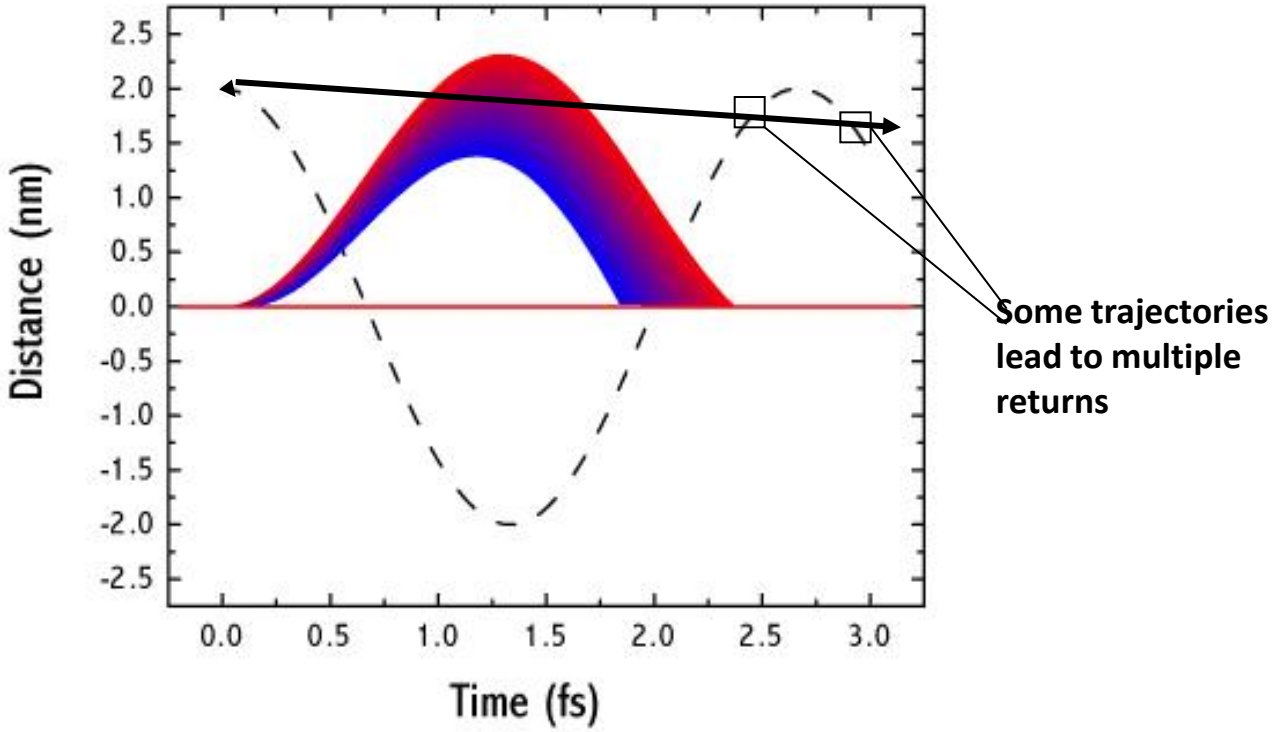
Temporal spread ~1 fs for a 800 nm driving pulse (depends on optical cycle)



For short trajectories (returning before 2nd zero of field within the cycle) the energy increases with return time.

Temporal chirp of electron wavepacket

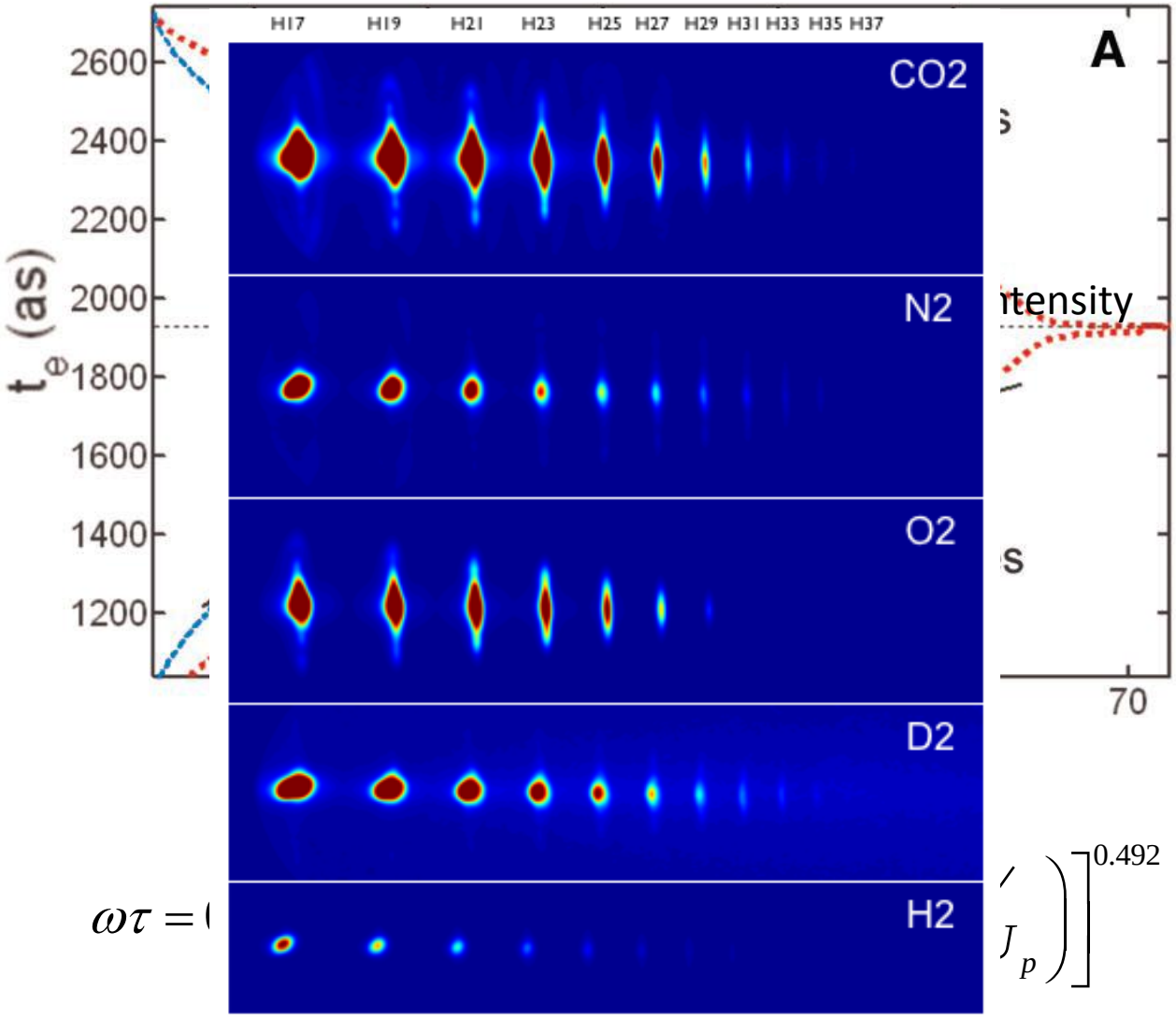
Temporal spread ~1 fs for a 800 nm driving pulse (depends on optical cycle)



For long trajectories (returning around and after 2nd zero of field within the cycle) the energy decreases with return time.

.

In SFA an approximate analytical form can be extracted for the energy-time mapping

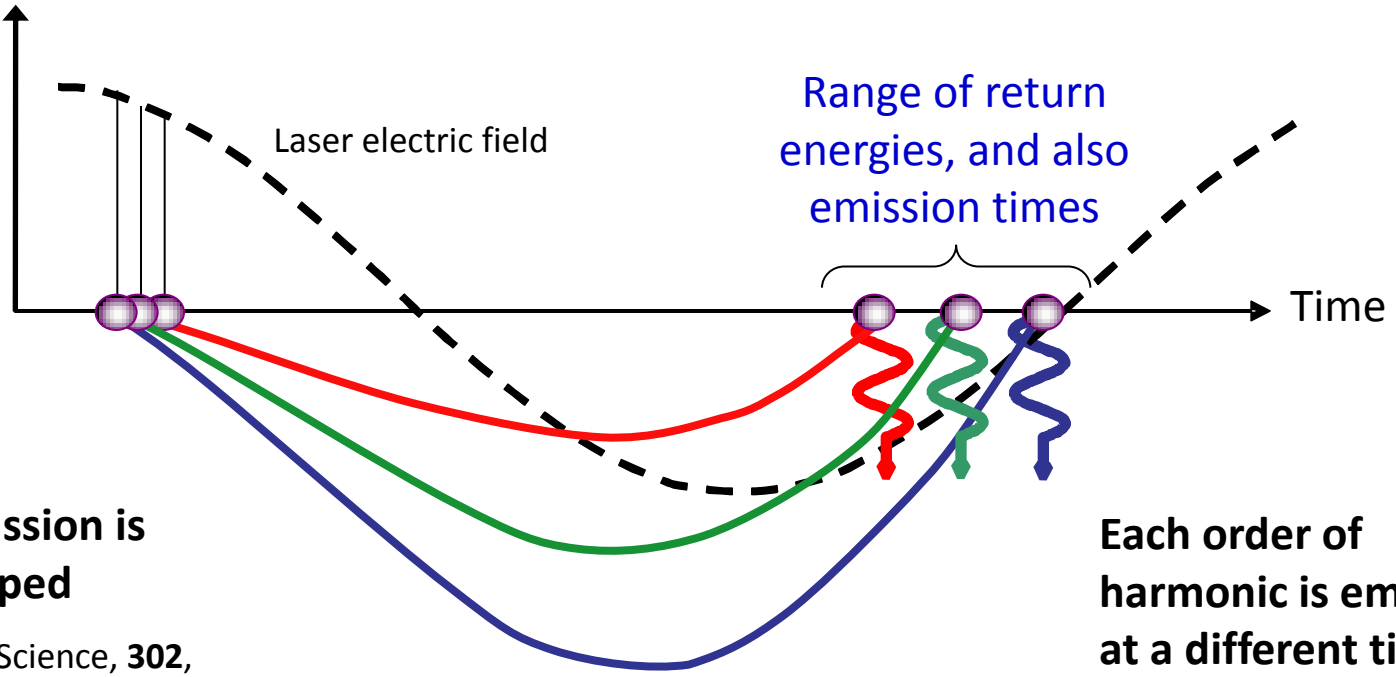


$$f(x) = \arccos\left(1 - x/1.5866\right)/\pi$$

Temporal chirp of harmonic spectrum

Ionisation can occur for a range of times around the peak of the electric field.

Electrons born at different times follow different trajectories, and gain varying amounts of energy from the field. The continuum electron wavepacket is chirped.



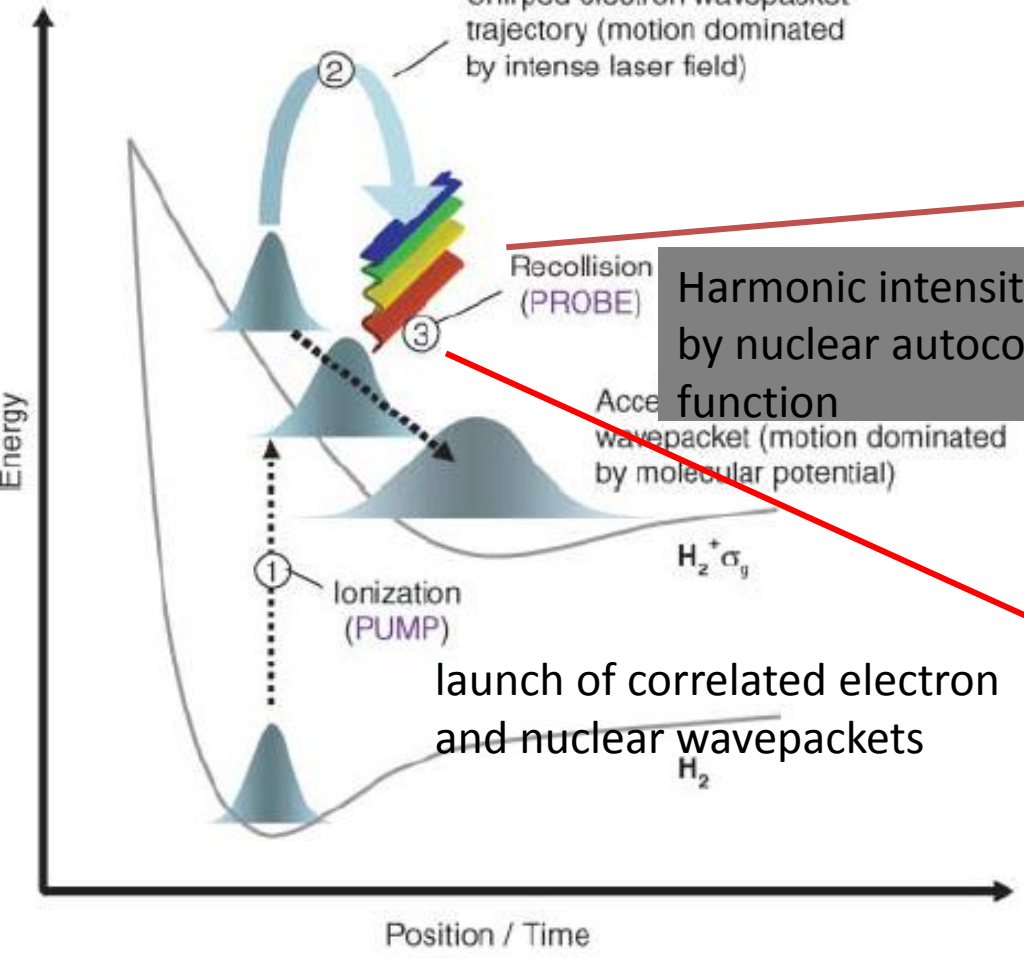
Harmonic emission is therefore chirped

Y. Mairesse *et al*., Science, **302**, 1540 (2003).

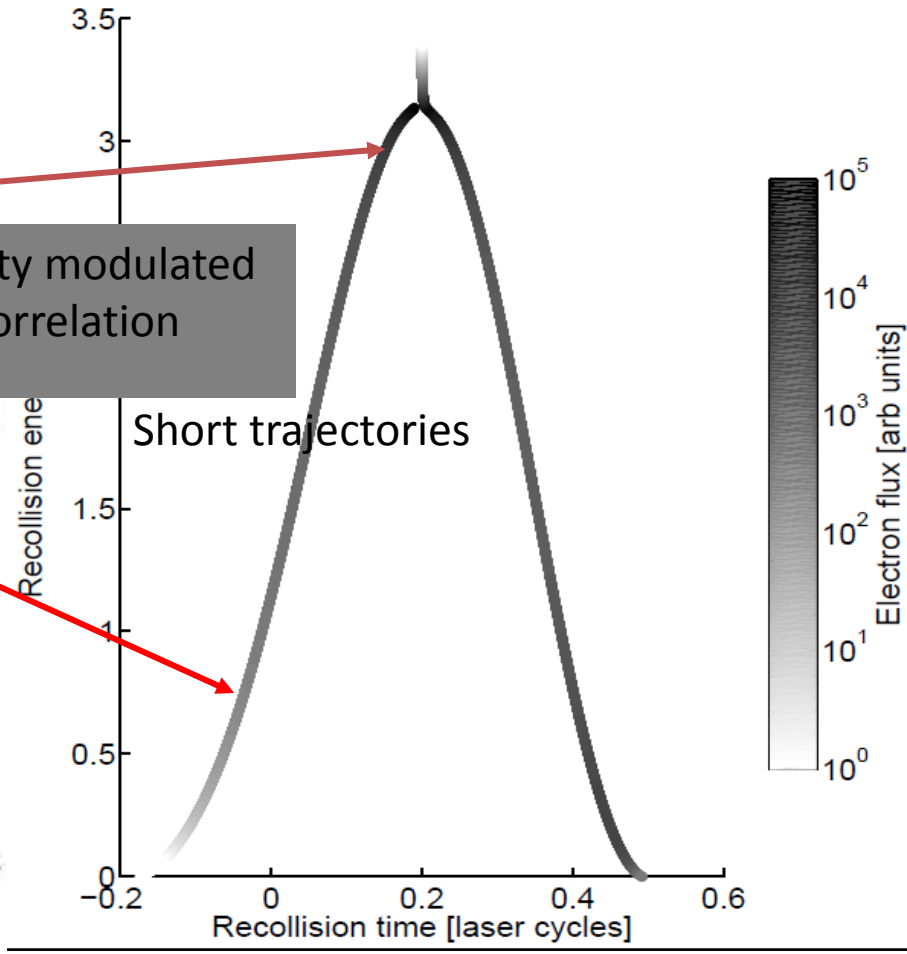
Each order of harmonic is emitted at a different time

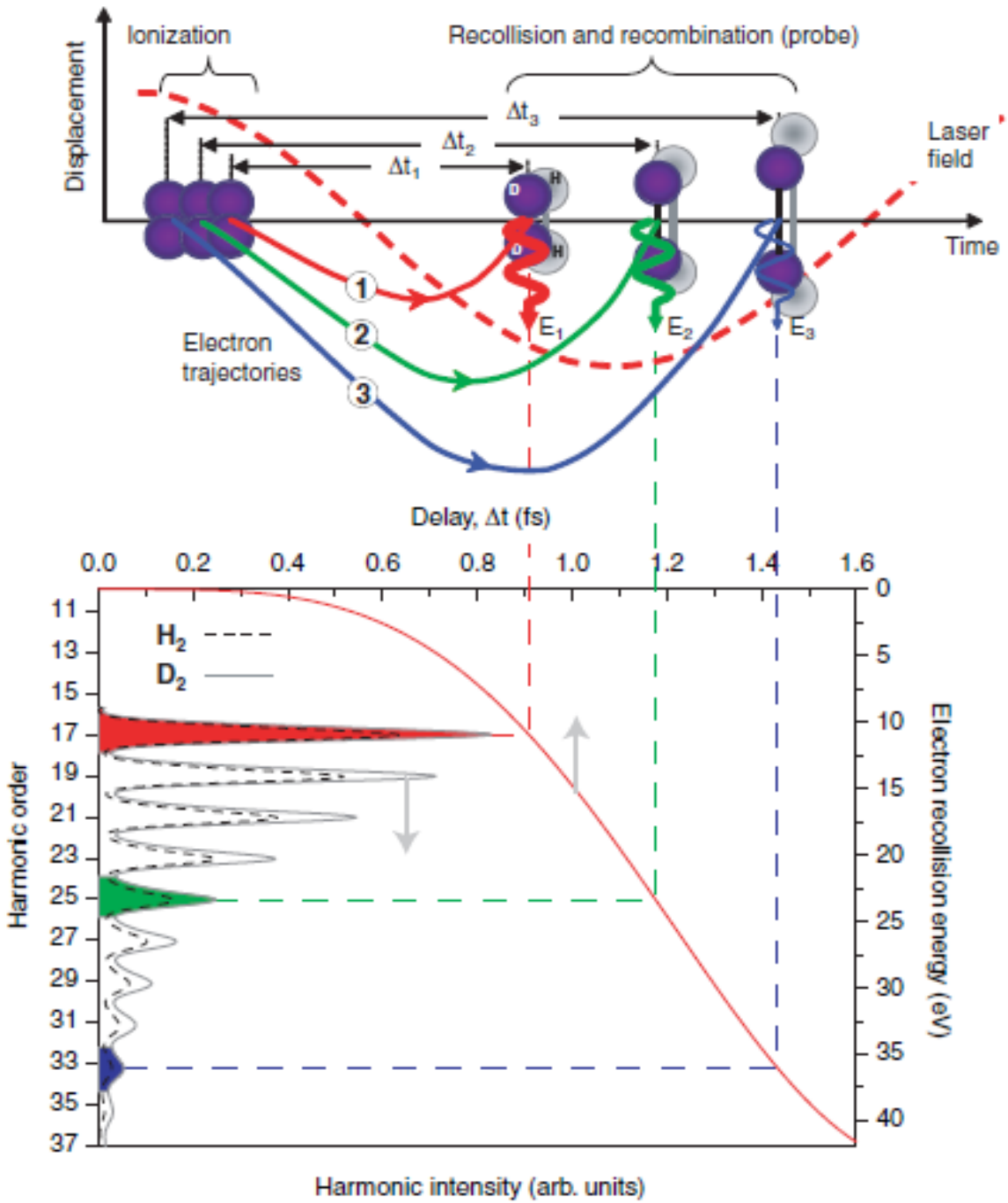
We use this to measure intra-molecular rearrangement following ionisation

HHG for probing attosecond dynamics:
Probing attosecond dynamics via chirp encoded recollision (PACER)



Harmonic intensity modulated by nuclear autocorrelation function



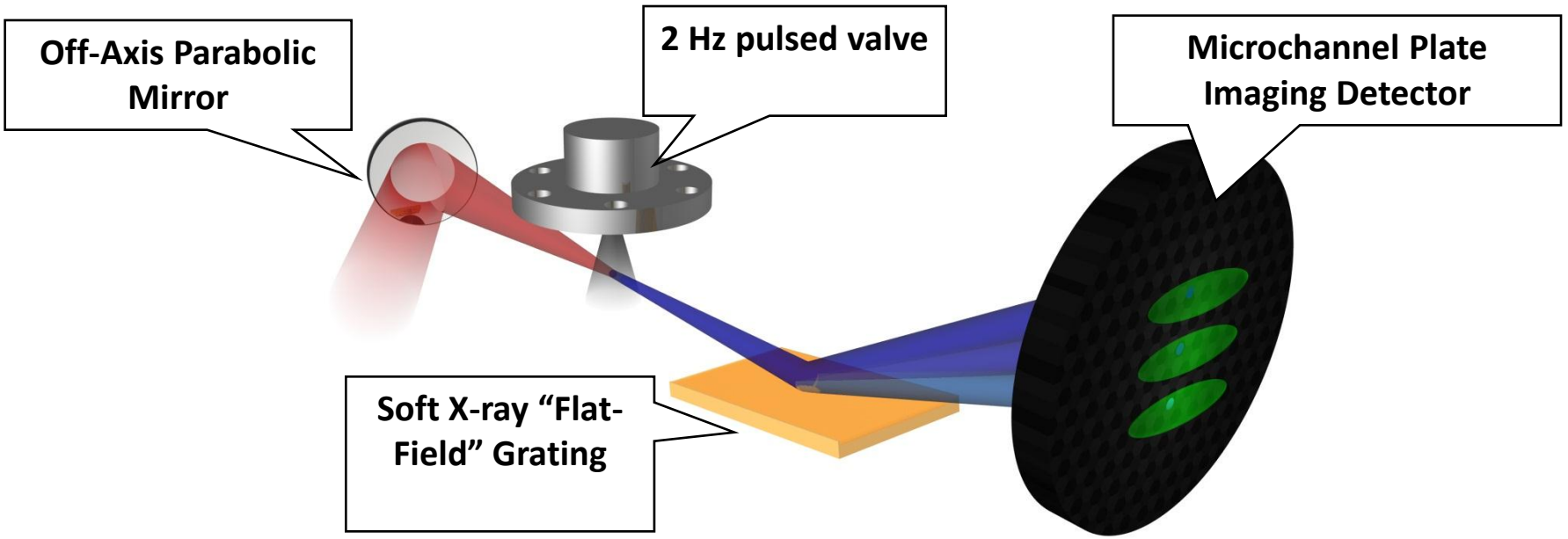

Ionization
Recollision and recombination (probe)
Displacement
Time
Laser field
Δt_1
Δt_2
Δt_3
1
2
3
E_1
E_2
E_3
Electron trajectories
Delay, Δt (fs)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
H_2
D_2
Harmonic order
11
13
15
17
19
21
23
25
27
29
31
33
35
37
Electron recollision energy (eV)
0
5
10
15
20
25
30
35
40
Harmonic intensity (arb. units)

Experimental approach part I

Focus 9 mm before jet to isolate short trajectories.

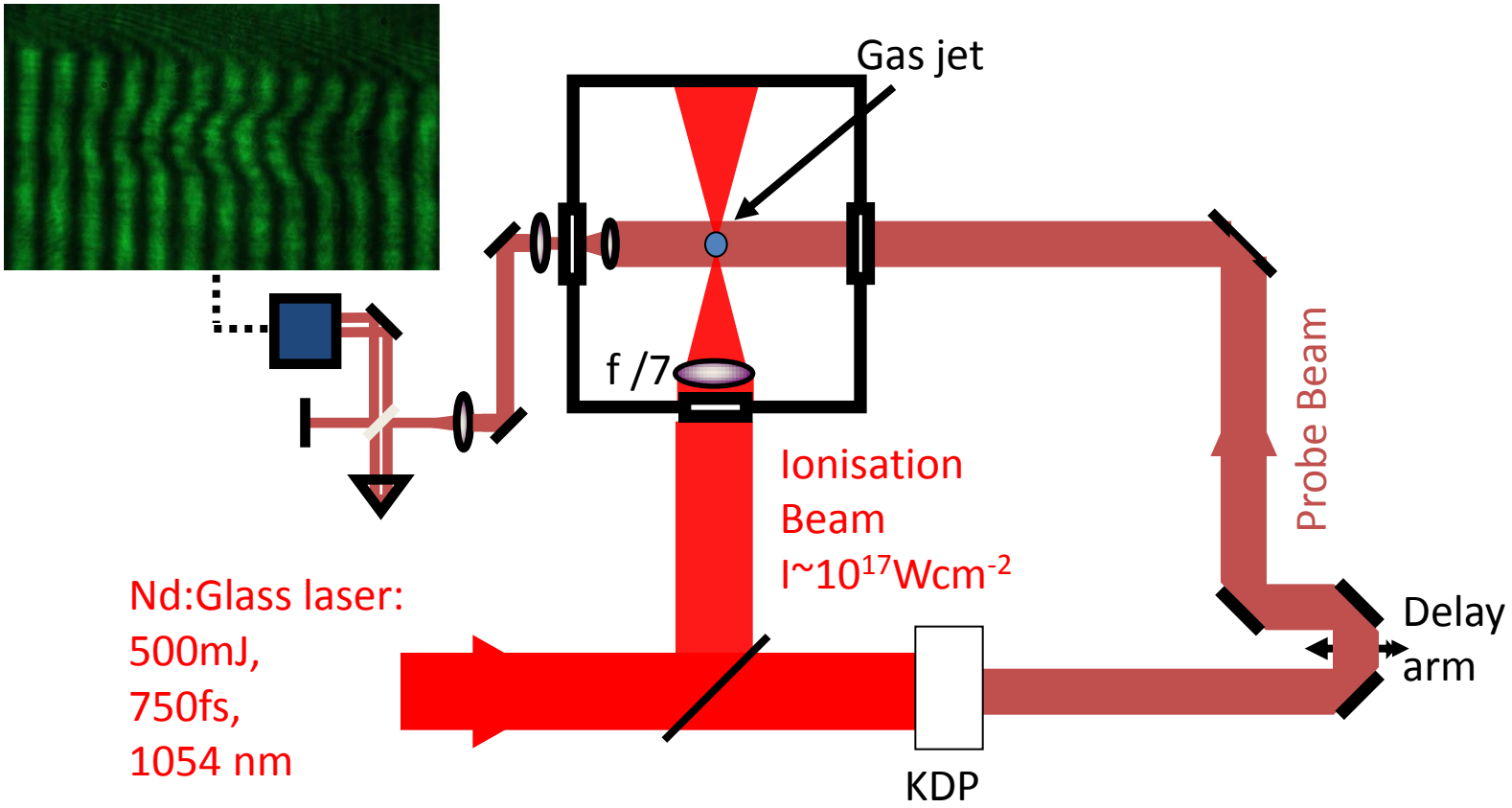
Intensity delivered at interaction region: 2 x 10^{14} $Wcm^{-2:}$ shot-to-shot fluctuation <3%, **monitored between data runs**.

Apply correct gas jet backing pressures **to ensure equal gas densities at the interaction region.**

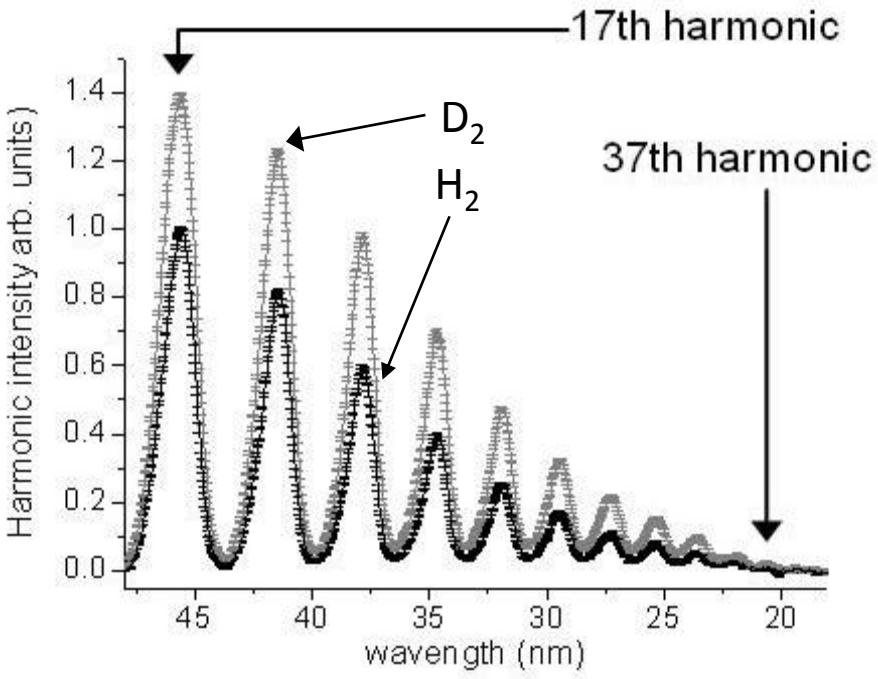


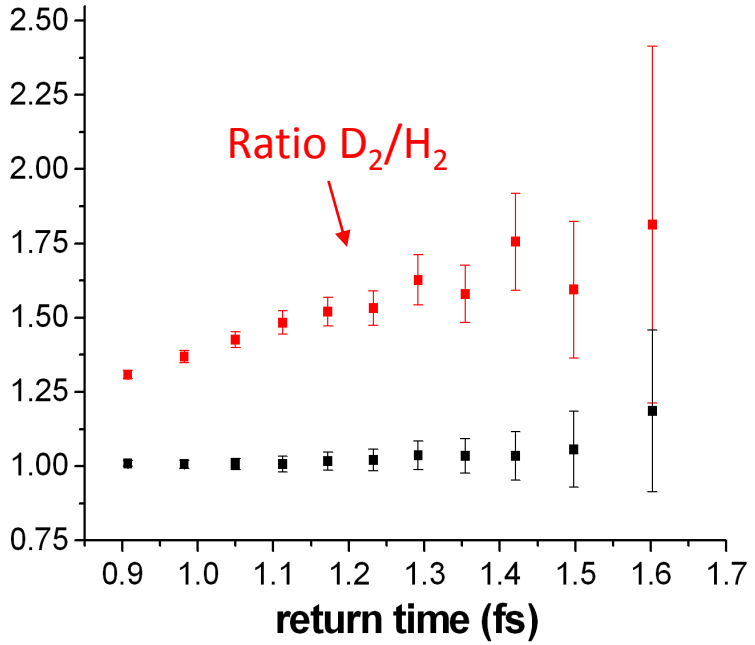
Experimental approach part II

Apply correct gas jet backing pressures **to ensure equal gas densities at the interaction region.**



Ratio of harmonic signals increases with electron return time

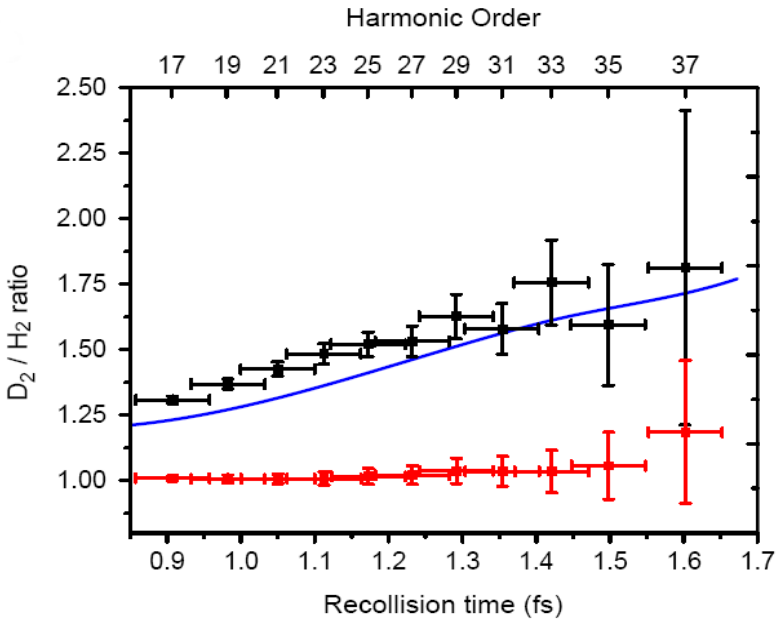




Effect robust to variations in gas jet backing pressure.

Re-absorption cannot account for increase observed, although makes small contribution.

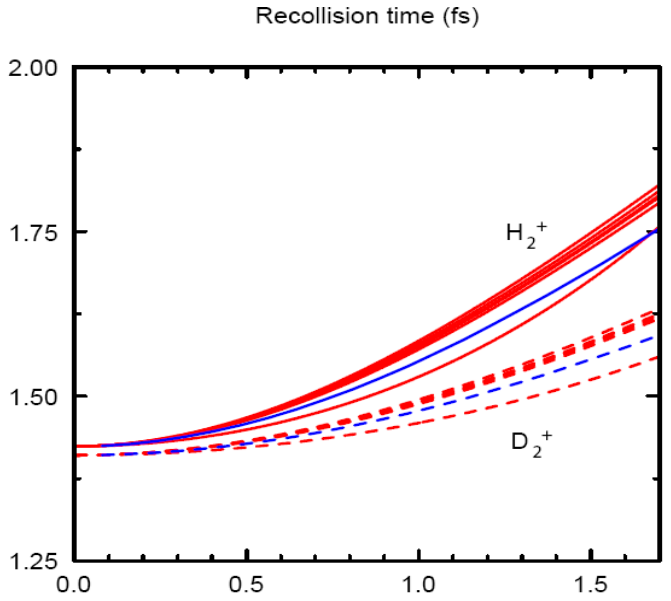
PACER (Probing Attosecond Dynamics with Chirp Encoded Recollisions): a technique for probing ultrafast nuclear motion in molecules



Iterative reconstruction of motion with ~100 as time resolution, using 8 fs pulses

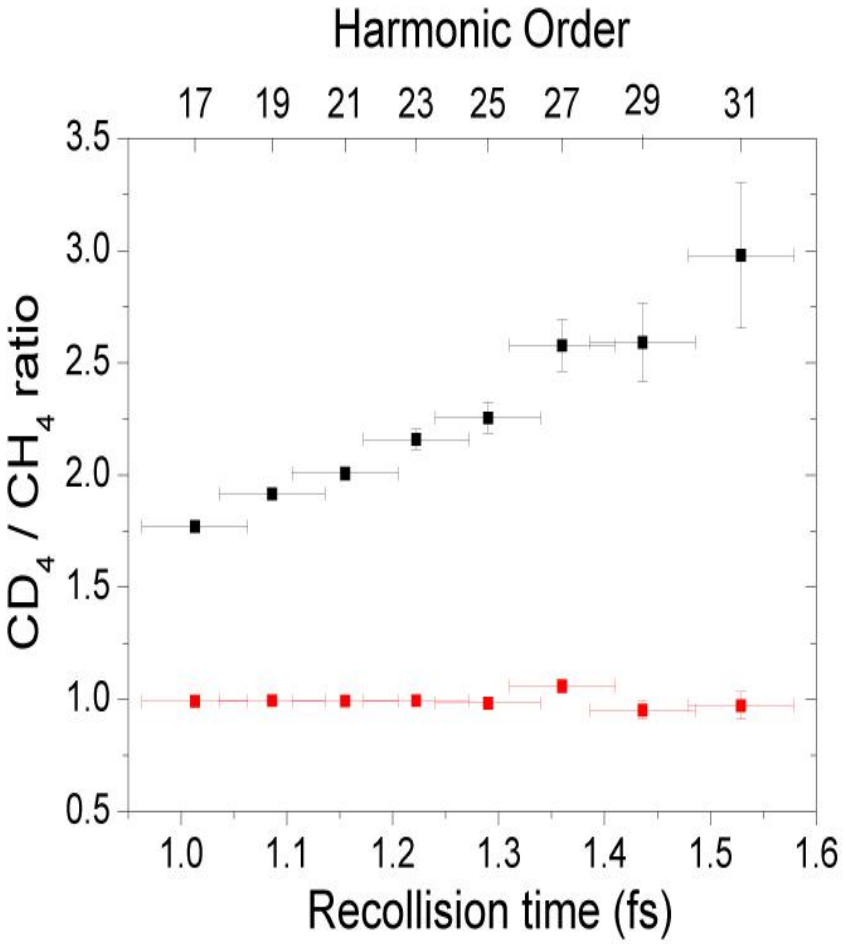
The fastest ever measurement of motion in a molecule:

Ratio D_2/H_2 emission as predicted by Lein accounting for time dependent nuclear correlation function [PRL 94, 053004 (2005)].

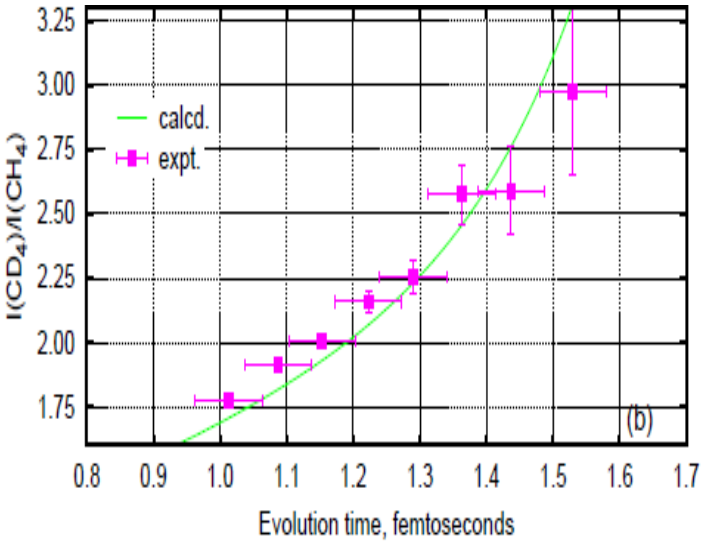


Baker *et al., Science* 312 , 424 (2006).

The nuclear correlation function ratio between CH_4 and CD_4 provides evidence of an ultra-fast rearrangement of methane upon ionisation



Baker *et al* Science 312, 424 (2006)



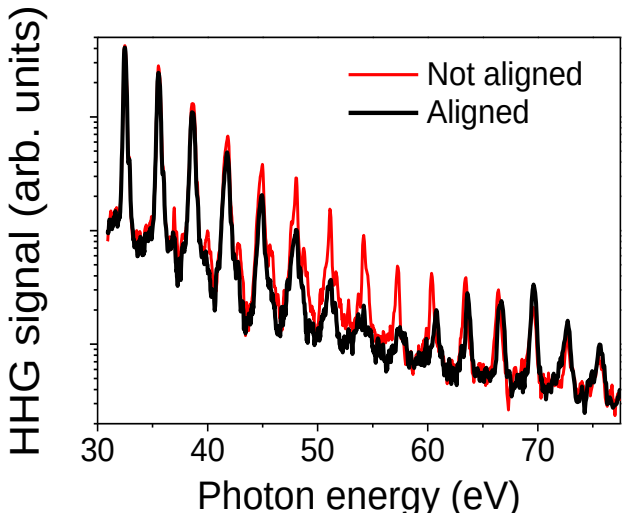
Courtesey of Serguei Patchovskii. (to be published)

Short time autocorrelation functions calculated using simple analytical theory.

Technical Issues – PACER

- Must select a single trajectory (use far field separation of short and long – some groups not equipped to do this)
- Multiple cycles (especially if at 2-4 cycles) mess up encoding, ideally use a long pulse (but this causes other problems or single cycle or polarisation gating)
- Reference not readily available to disentangle dynamical effects from the frequency dependence of the harmonic dipole (requires careful comparisons of wavelength/intensity)
- Limited time window – using long trajectories so far tricky to use

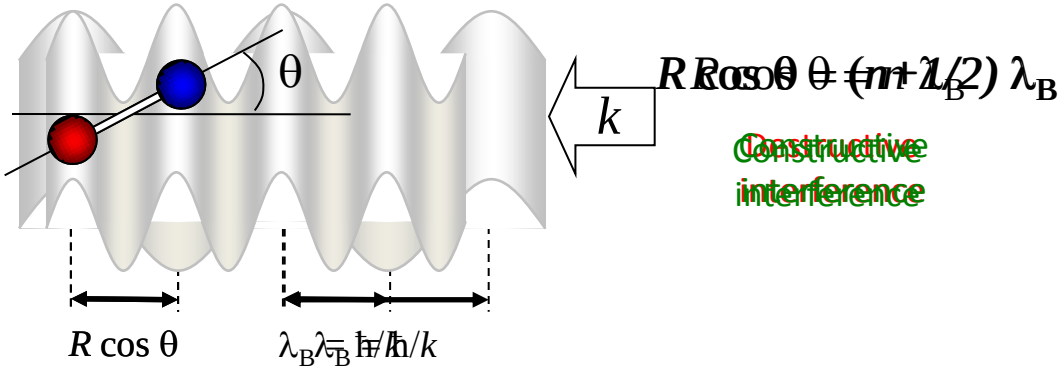
HHG in aligned CO_2: Strong dip observed with 800nm laser has it a structural or a dynamical cause?



C. Vozzi *et al. Phys. Rev. Lett.* 95, 153902 (2005)

Kanai et al, Nature 435, 2005

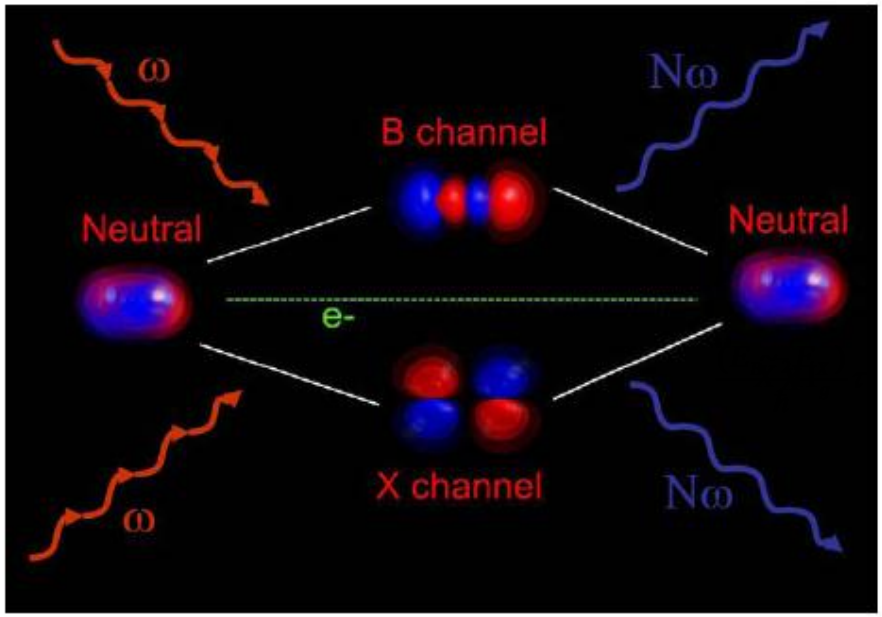
Two-center interference

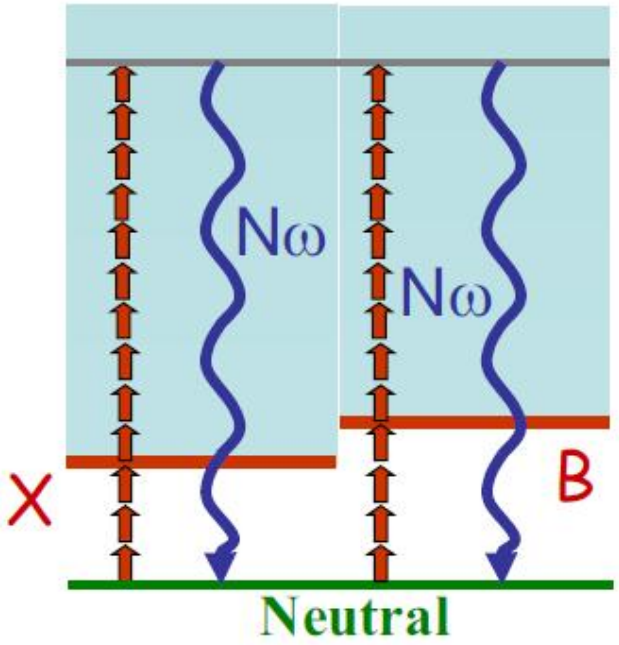


M. Lein *et al. Phys. Rev. Lett.* 88, 183903 (2002)

There is also evidence for a role of different ion states: intensity dependent shift of minimum position in CO_2 with 800nm

Smirnova et al Nature 2009
Using a PACER type measurement



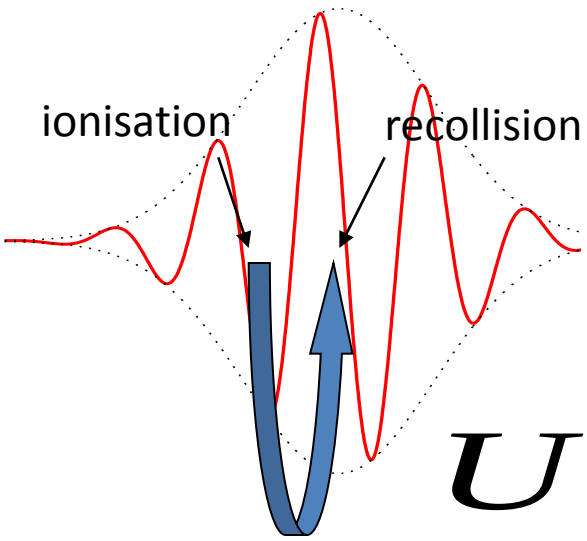


Ionisation forms a superposition of states of the molecular ion:
X (HOMO) and B (HOMO-2)

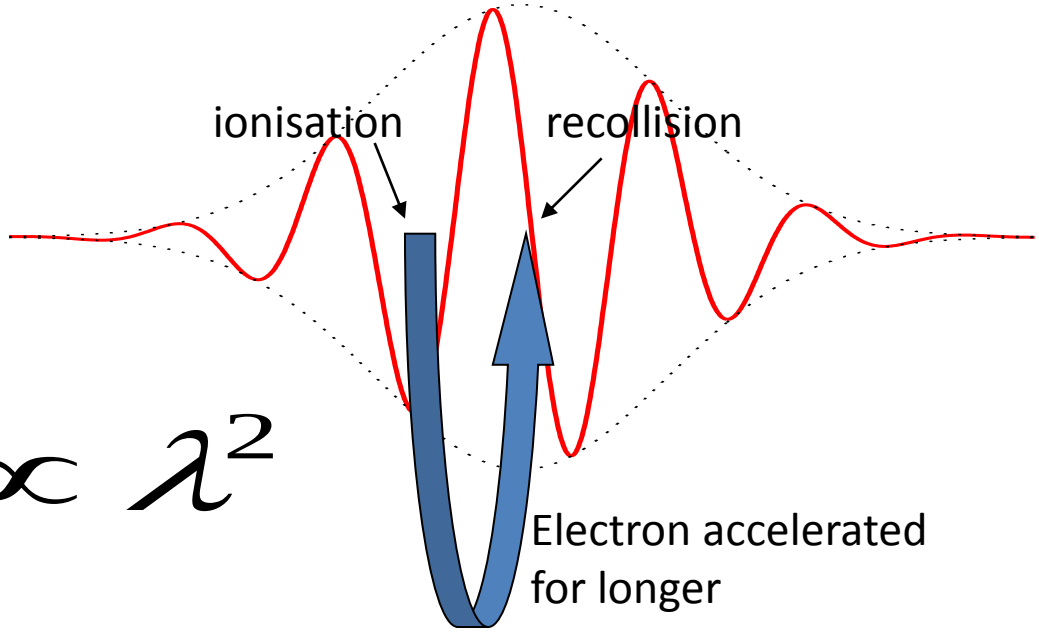
A dynamical interference between the complex amplitudes of the two channels (time for interference maxima depends on energy differences).

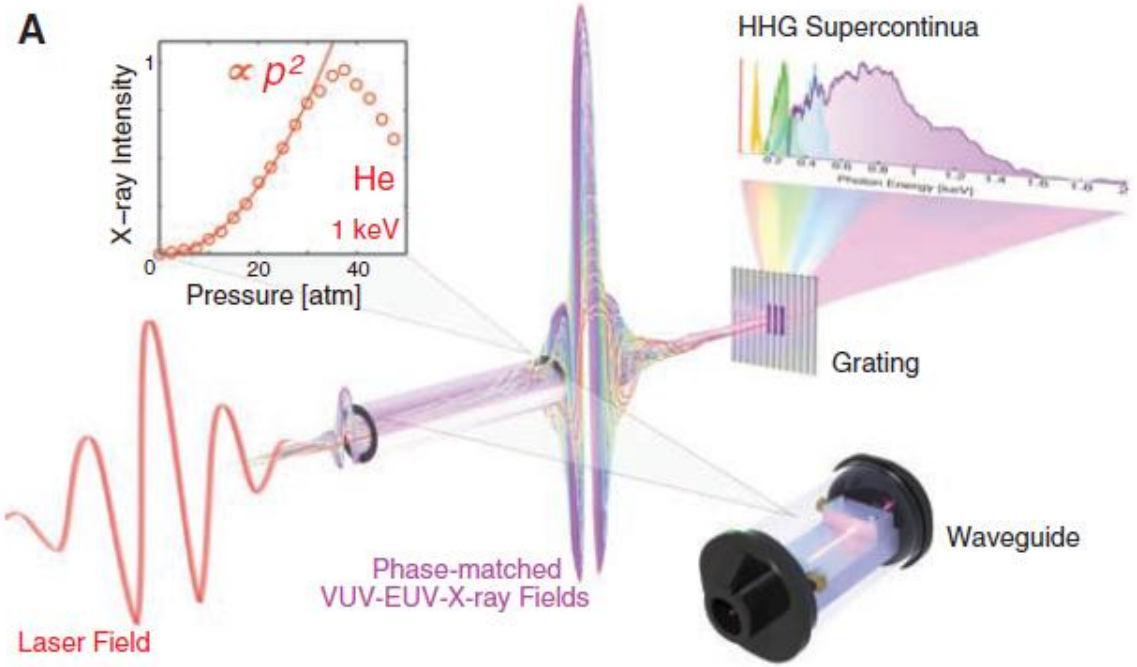
Relative Phase ~ $(E_B - E_x)\tau$ so position of destructive interference in HHG spectrum depends upon the emission time

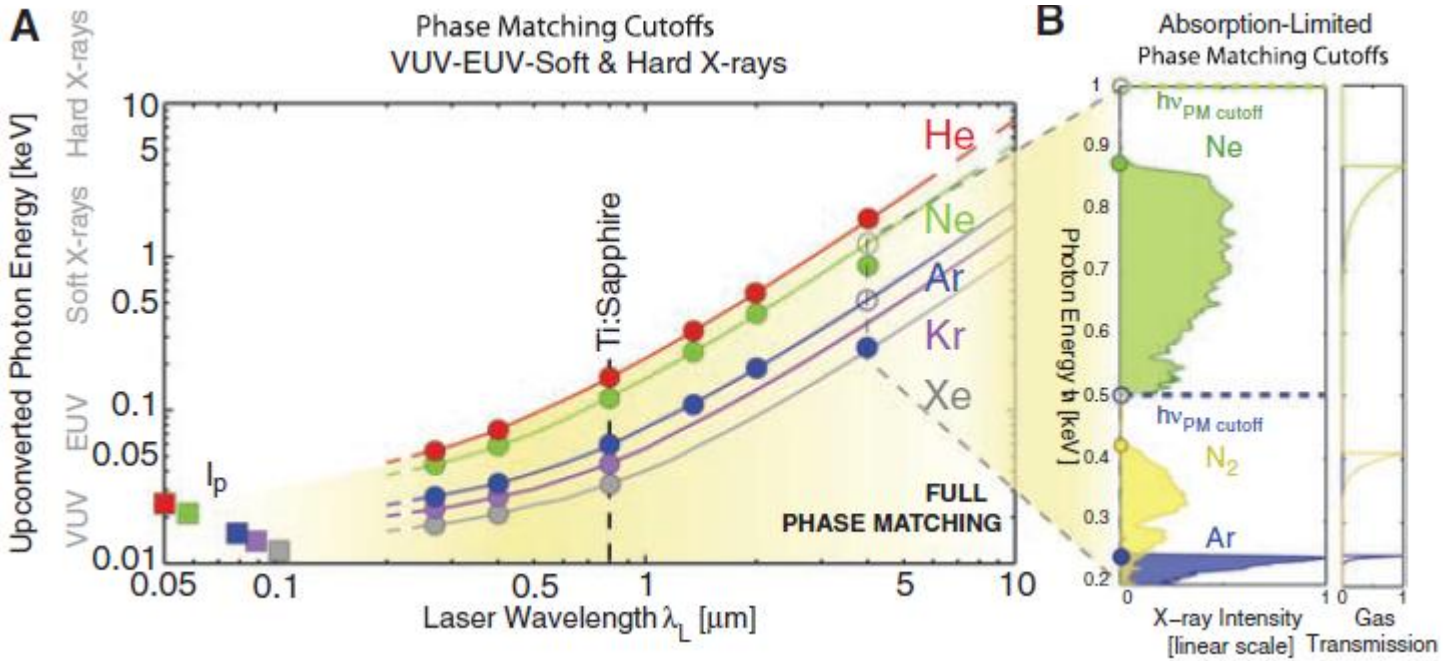
Tools 7:
Longer Wavelength Fields



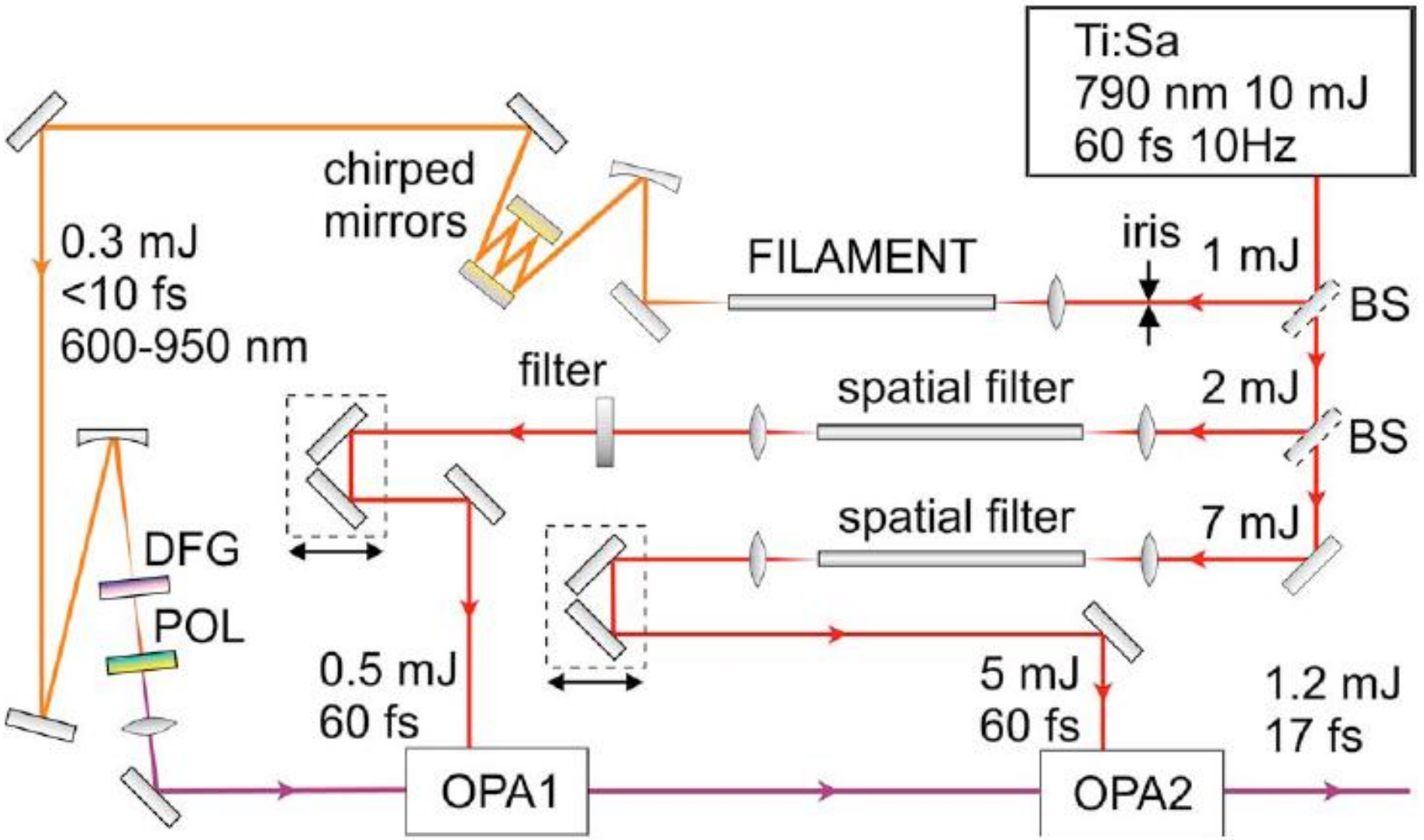
$$U_P \propto \lambda^2$$




A
X-ray Intensity
∝ p²
He
1 keV
Pressure [atm]
HHG Supercontinua
Photon Energy [keV]
Grating
Waveguide
Phase-matched
VUV-EUV-X-ray Fields
Laser Field


A
Phase Matching Cutoffs
VUV-EUV-Soft & Hard X-rays
Upconverted Photon Energy [keV]
Hard X-rays
Soft X-rays
EUV
VUV
He
Ne
Ar
Kr
Xe
Ti:Sapphire
I_p
FULL
PHASE MATCHING
Laser Wavelength λ_L [μm]
B
Absorption-Limited
Phase Matching Cutoffs
$h\nu_{PM\ cutoff}$
Ne
$h\nu_{PM\ cutoff}$
N_2
Ar
Photon Energy [keV]
X-ray Intensity
[linear scale]
Gas
Transmission

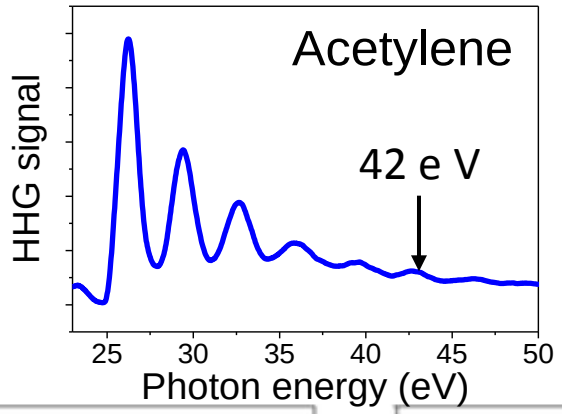
Few-Cycle Pulses at Longer Wavelengths



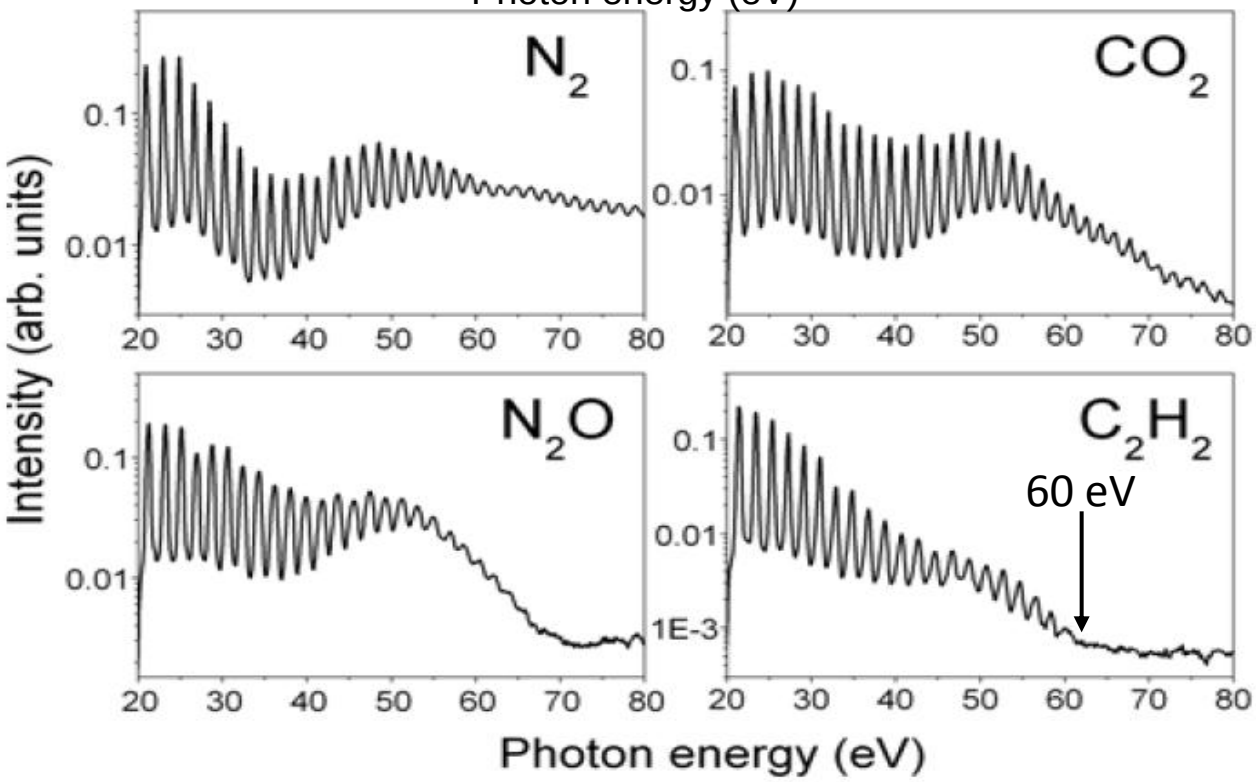
1450 – 1700 nm

DFG- difference frequency generation; OPGA – optical parametric amplification

Benefit of 1300nm compared to 800nm immediately seen in cut-off extension



10fs 800nm – cut-off fixed by ionisation saturation intensity, especially if Ip lower.



40 fs 1300nm
Un-normalized spectra

Longer Wavelengths for HHG in Organic and Bio-Molecules

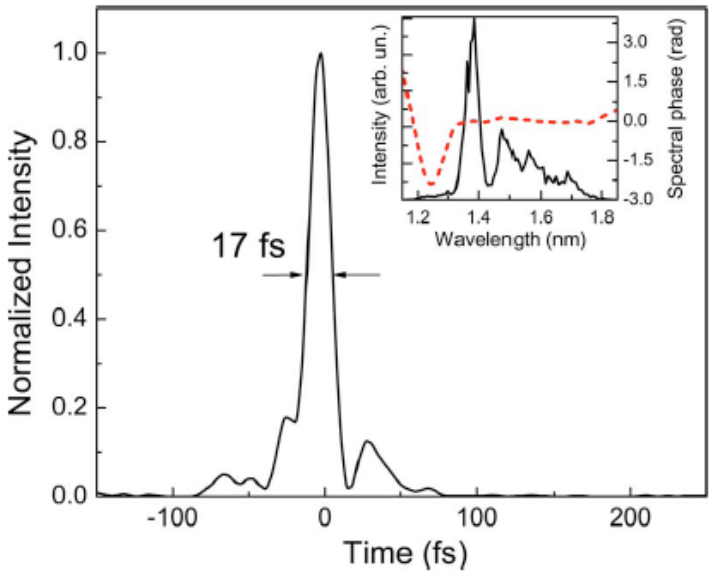
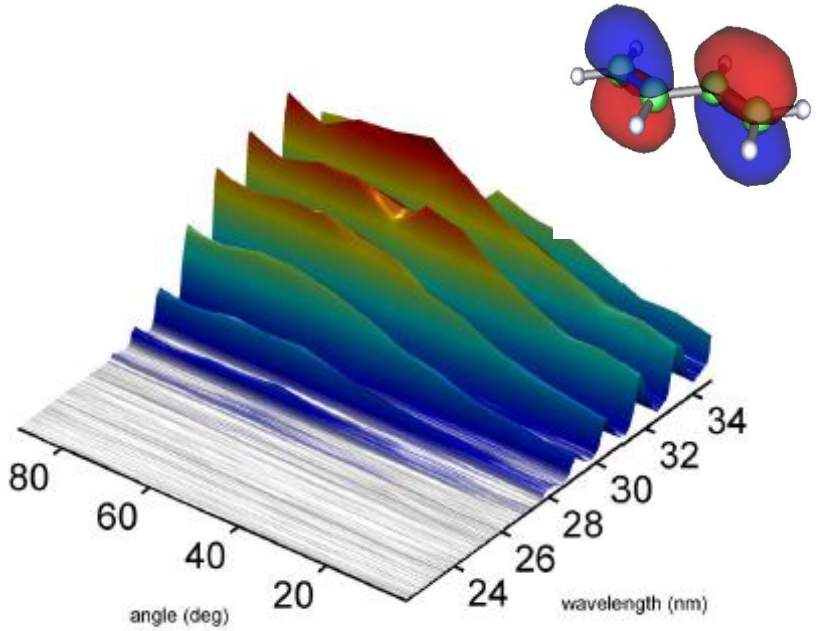


Fig. 3. (Color online) ZAP-SPIDER reconstructed temporal intensity profile corresponding to a pulse duration of 17 fs. The inset shows the amplified DF spectrum (solid curve) and the retrieved spectral phase (dashed curve).



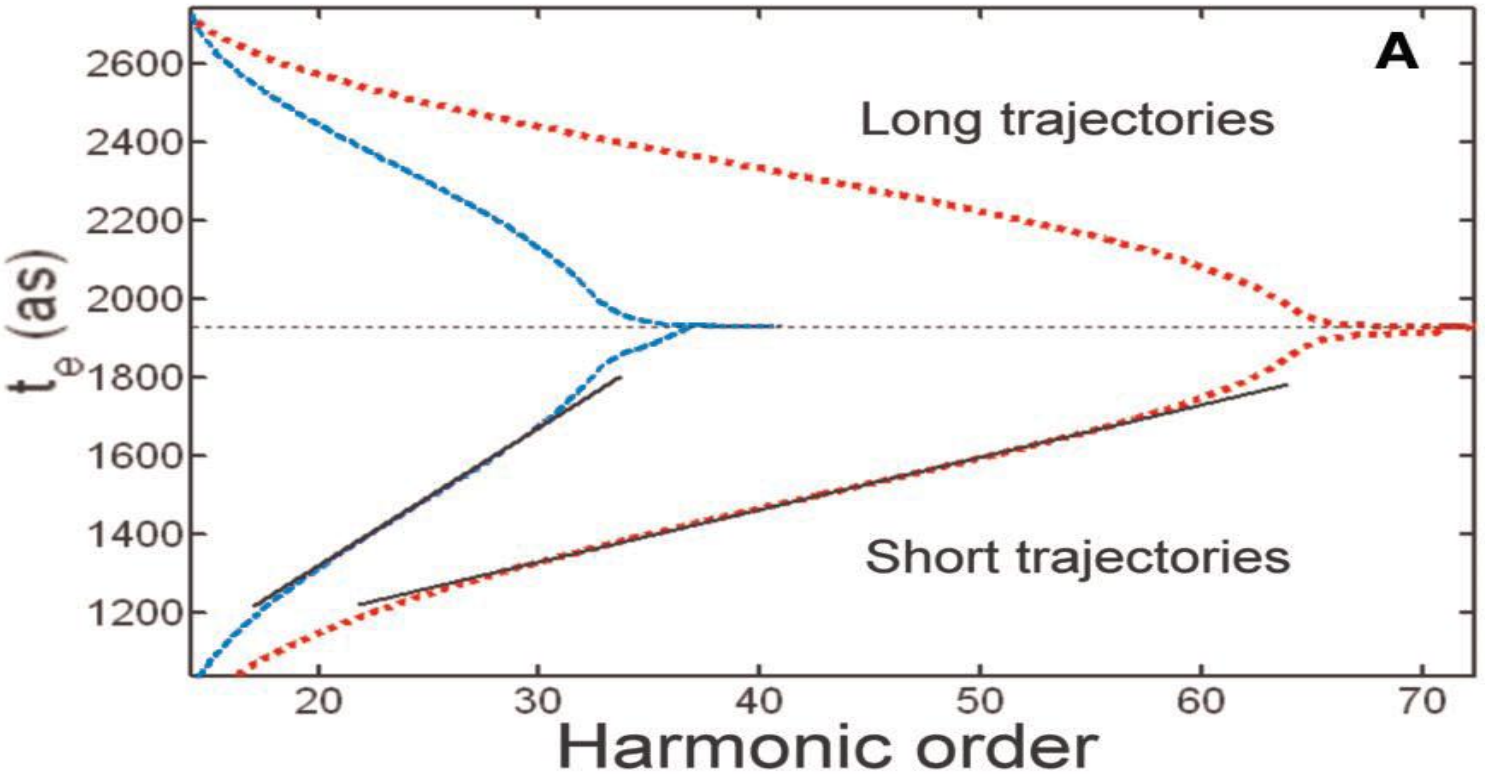
HHG from (1,3) Butadiene with 1450 nm pump, harmonic spectrum to > 45 eV showing clear angular dependence

The IP of butadiene is ~ 9 eV, which is in the same range as the IP of chromophores in biomolecules (e.g. peptides) in which non-trivial charge migration has been predicted. Therefore we believe that HHG spectroscopy performed with long wavelength fields has the real prospect to provide information on attosecond charge migration and structural features in a wide range of molecules.

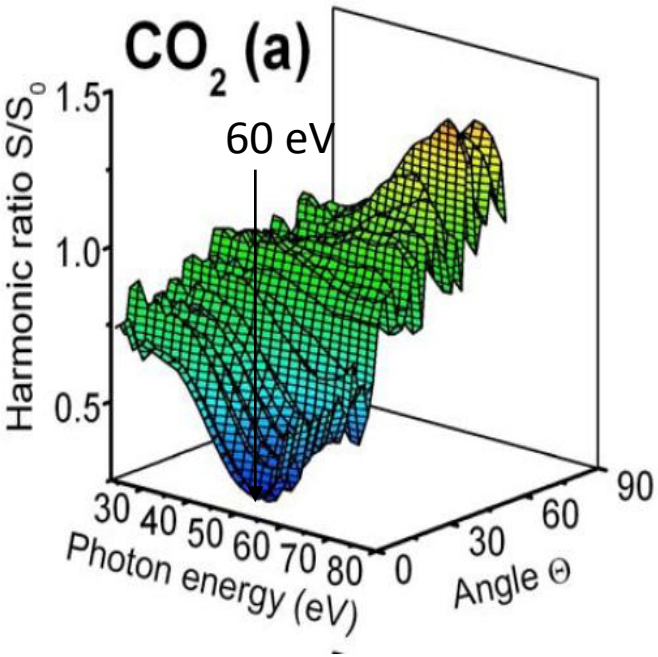
Technical Issues – Long Wavelengths

- OPG often give poor beam quality with hot-spots and larger energy fluctuations (spatial filter clean up may be required)
- Diagnostics need to be specially prepared for lasers in this wavelength range
- Lower efficiency HHG $\sim\lambda^{-6}$
- Few cycle pulses require new phase compensation methods (but nice window around 1750nm)
- Hollow fiber pulse compression has proved viable at these wavelengths
- Few suitable laser sources beyond 2000nm

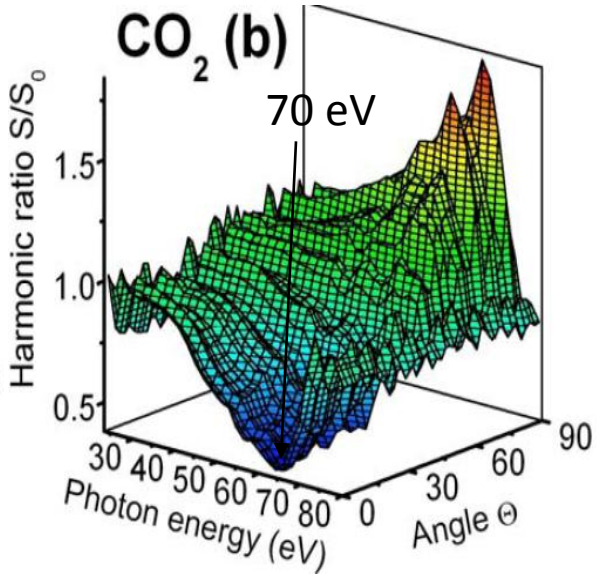
By using longer wavelength we can more readily control the time the electron spends in the continuum by changing intensity



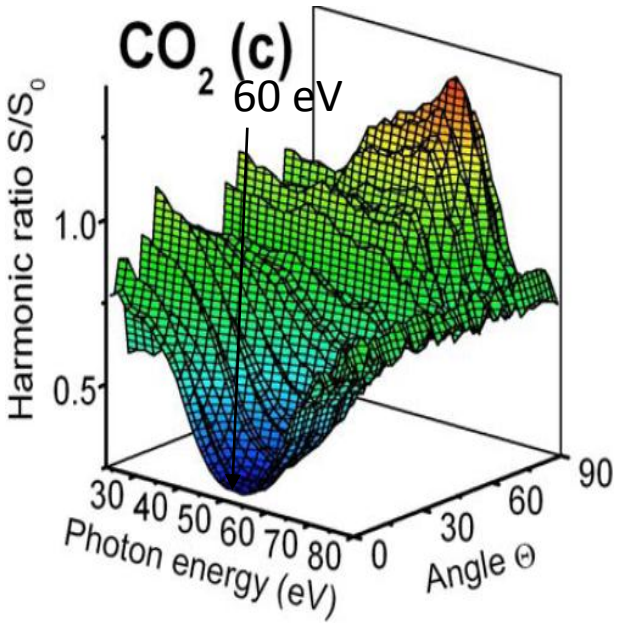
Minimum in CO2 is observed to be modulated by intensity of 1300nm field



~0.9 x 10^{14} Wcm^{-2}



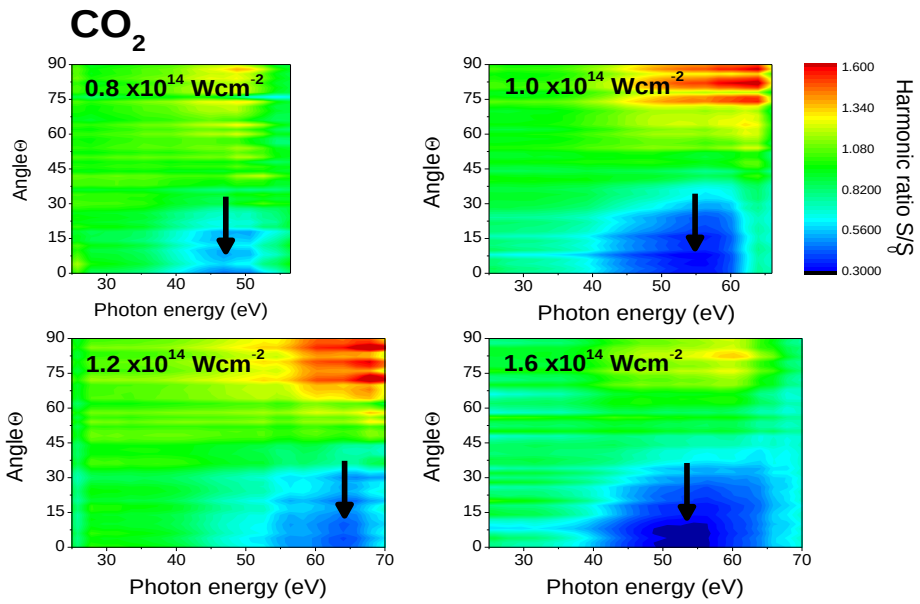
~1.2 x 10^{14} Wcm^{-2}

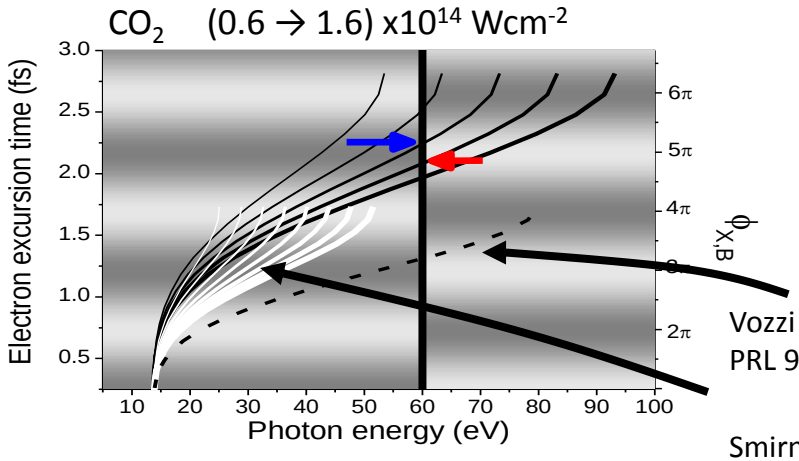


~1.6 x 10^{14} Wcm^{-2}

This requires an explanation that needs to invoke dynamical interference between **X** and **B** channels

Measurements with a 1300nm laser has been used to follow the dynamics associated with a superposition of hole states in CO_2





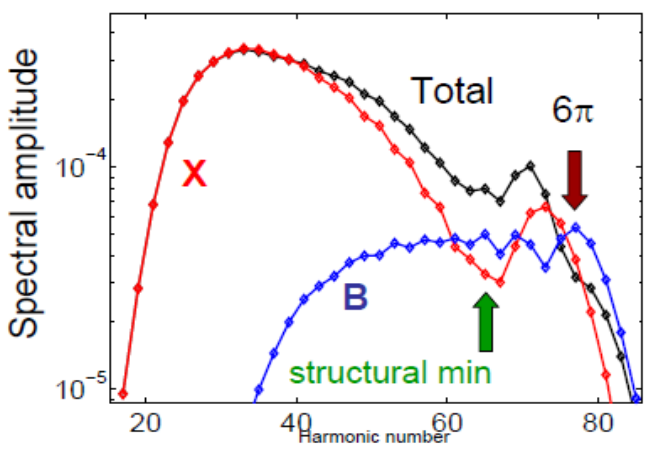
Vozzi *et al.*
PRL 95, 153902 (2005)

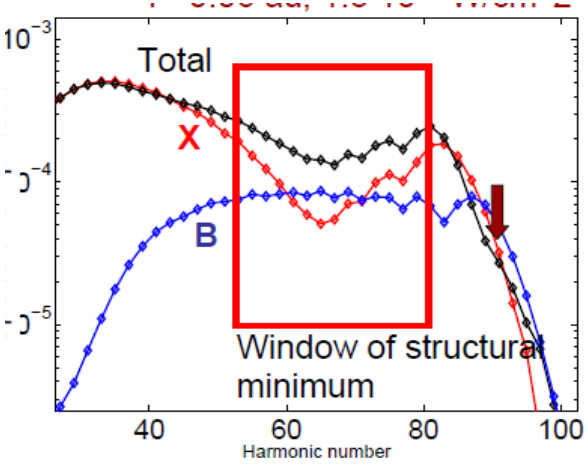
Smirnova *et al.*
Nature 460, 972 (2009)

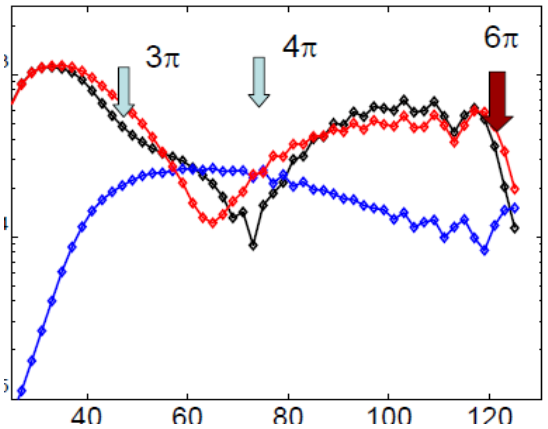
Torres *et al.* PRA 81, 051802(R) (2010)

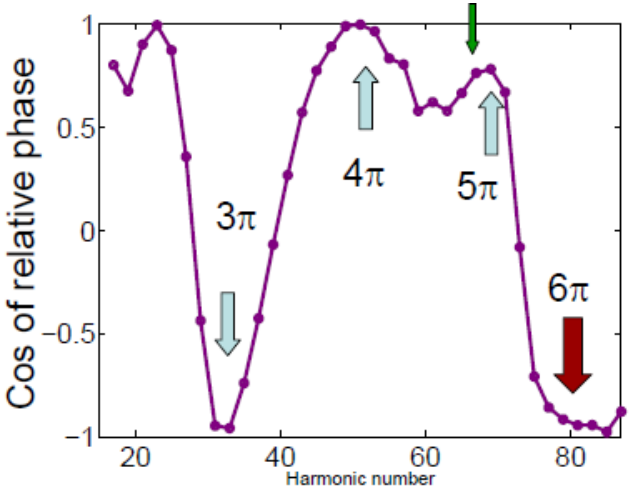
Calculations at 1300nm: Structural minimum in X remains fixed but the minimum in the total spectrum shifts with intensity due to change of relative phase of X and B channels

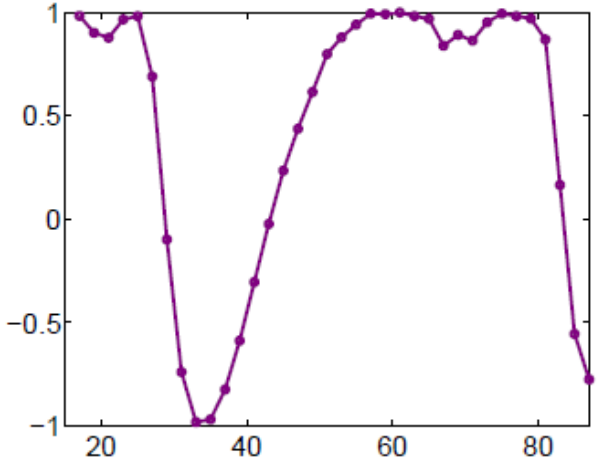
Calculation by Smirnova, Patchovskii and Ivanov

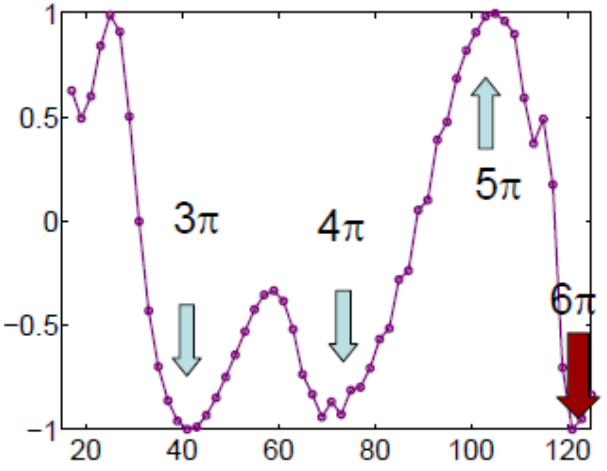










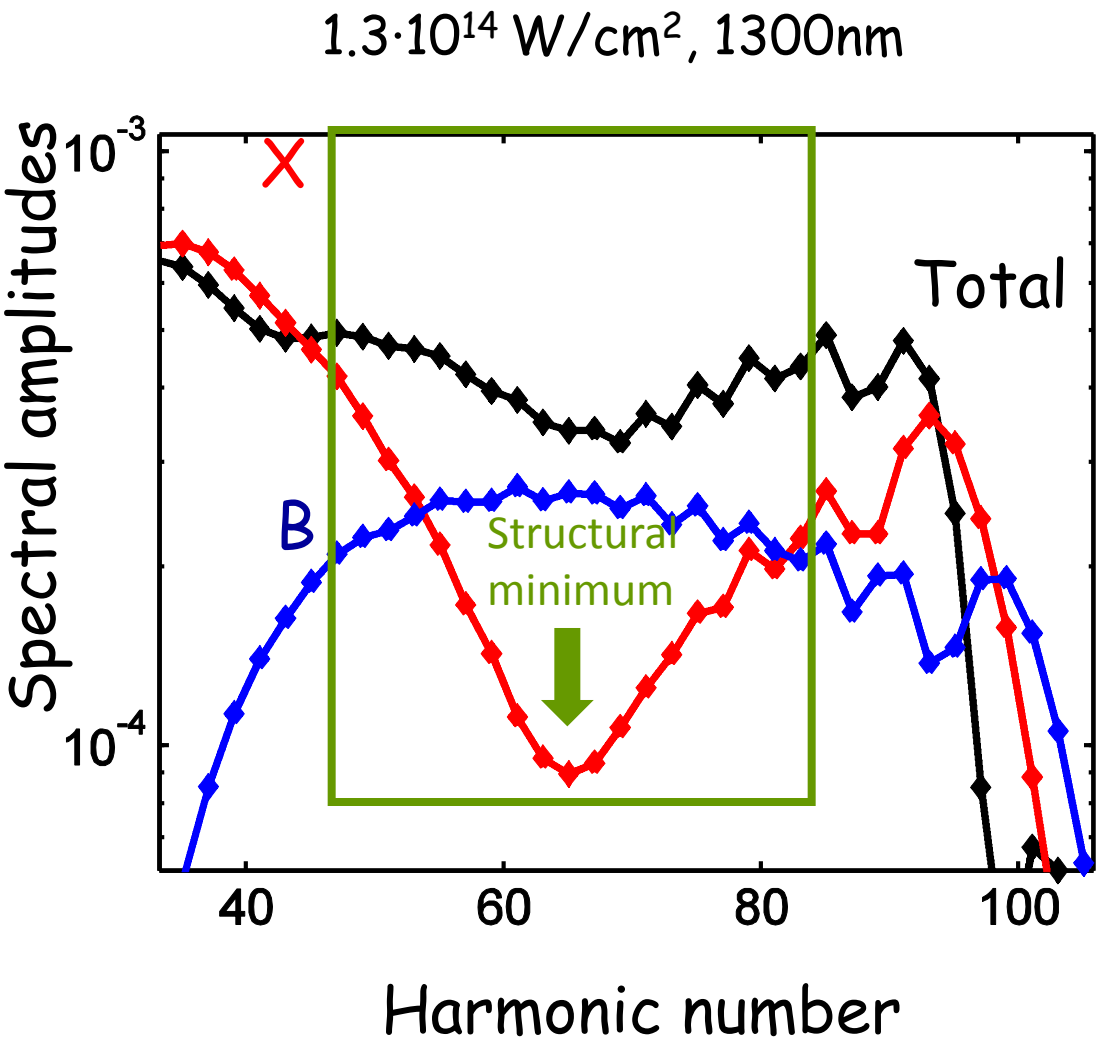


10^{14} Wcm^{-2}

1.3 x 10^{14} Wcm^{-2}

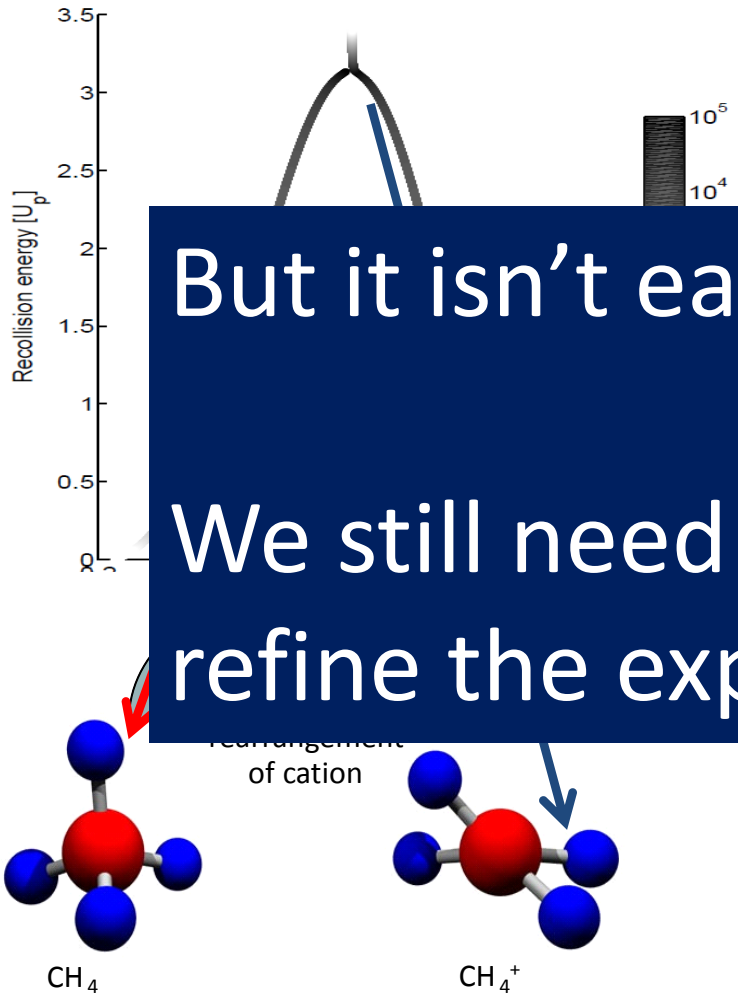
2 x 10^{14} Wcm^{-2}

Measurements of HHG in CO_2 at 1300nm varying intensity reveals dynamical interference between channels BUT requires structural interference to be seen



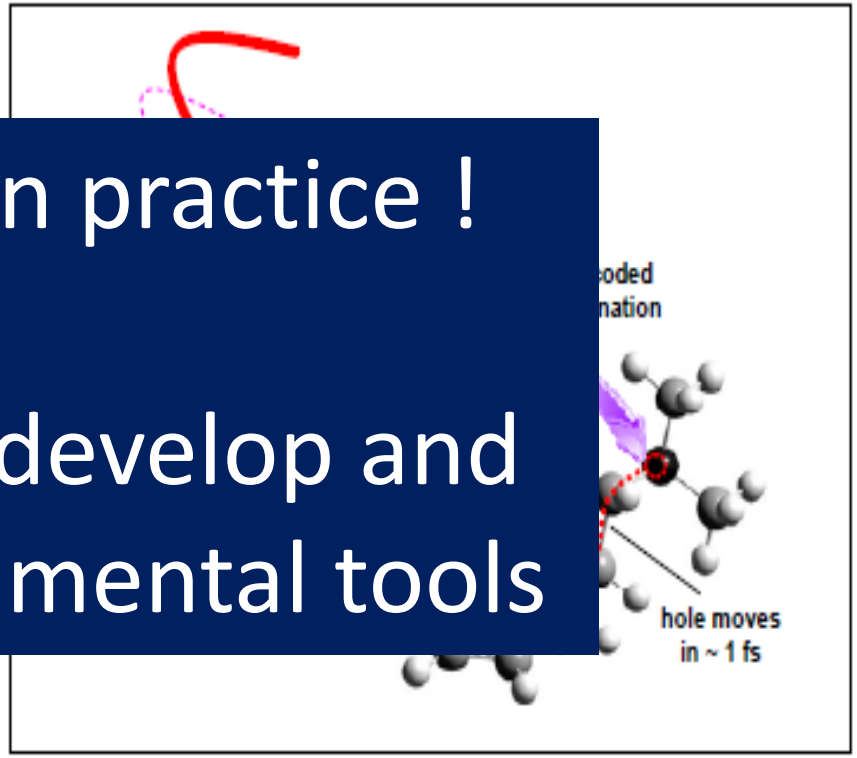
Torres *et al.* PRA 81, 051802(R) (2010)

Conclusion: HHG spectroscopy is a tool for attoscience



But it isn't easy in practice !

We still need to develop and refine the experimental tools



Prospects to measure hole dynamics in molecular cations .

Baker *et al.,* "Probing proton dynamics in molecules on an attosecond timescale" *Science* 312 , 424 (2006).

Torres *et al.* PRA 81, 051802(R) (2010)
Smirnova *et al.* Nature 460, 972 (2009)