

# HHG Sub-cycle dynamics

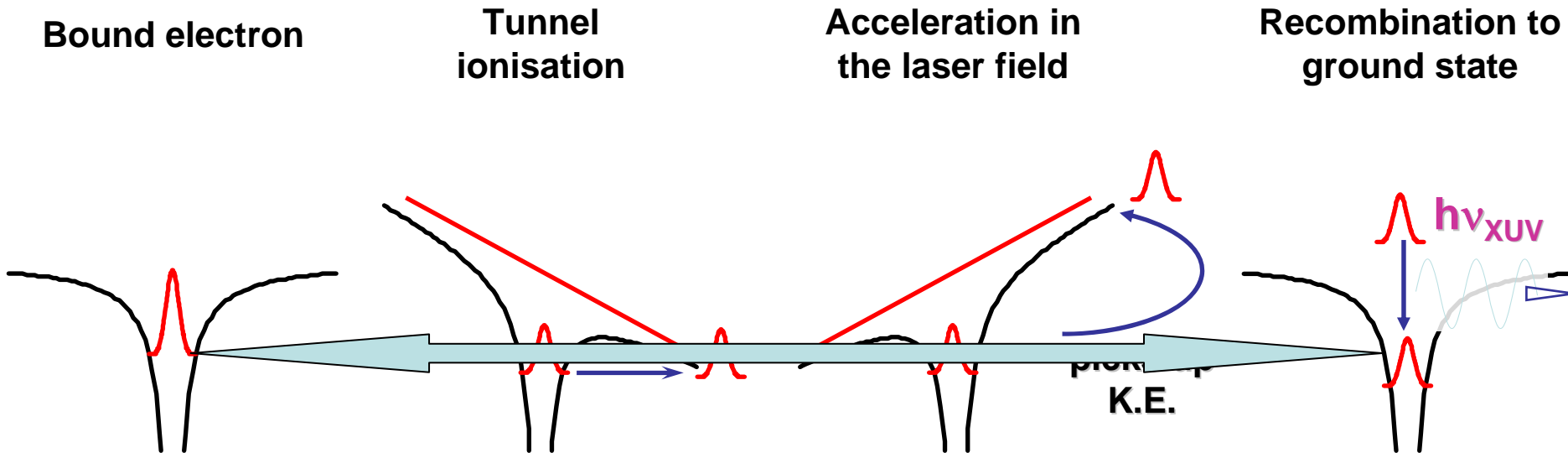
1. Chirp of electron recollision
2. Measuring ultra-fast intramolecular proton motion
3. Controlling recollisions via CEP
4. Controlling recollisions via multi-colour fields†

† including some recent research highlights from Imperial College node

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# High harmonic generation occurs on a sub-optical cycle timescale

- Recollisional model of HHG: e.g. Corkum PRL (1993), Lewenstein et al (1994)

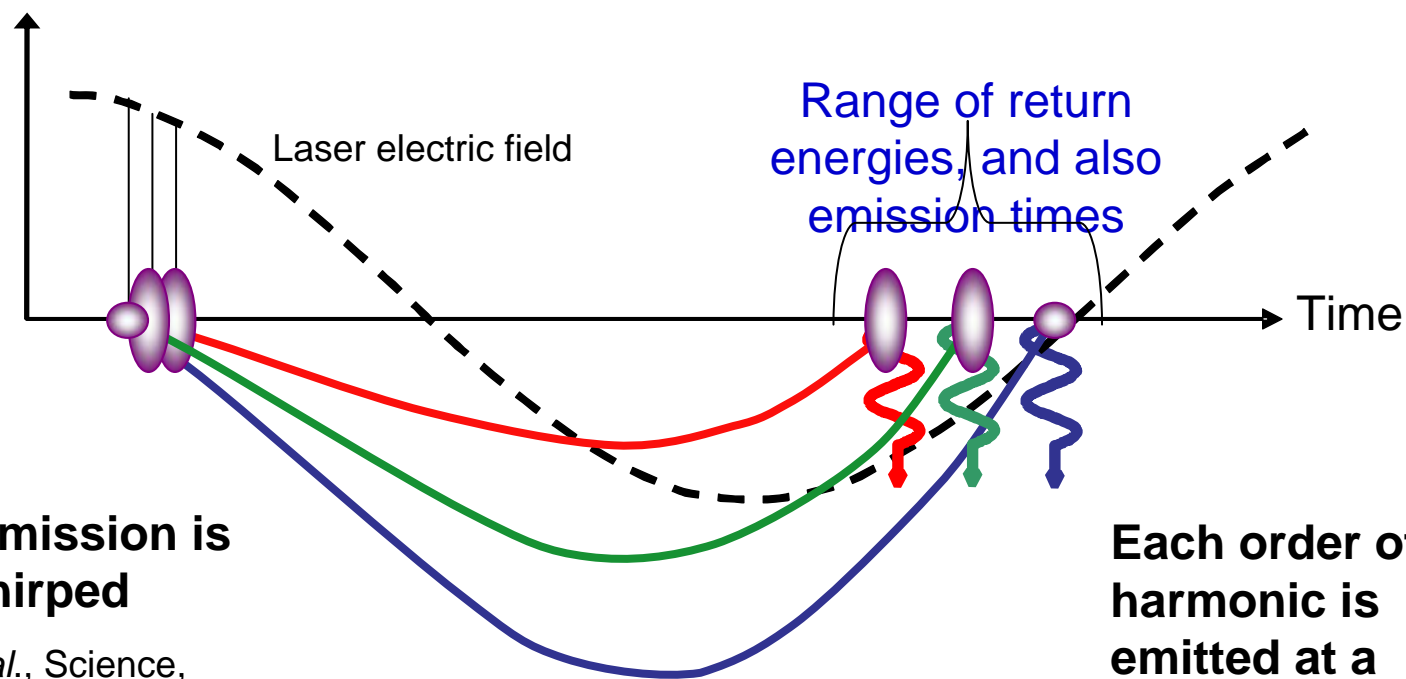


**HHG signal arises from coherent addition of contributions from atoms/molecules in the sample. For these contributions to add constructively initial and final states must be the same, giving the process a unique specificity.**

# 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**

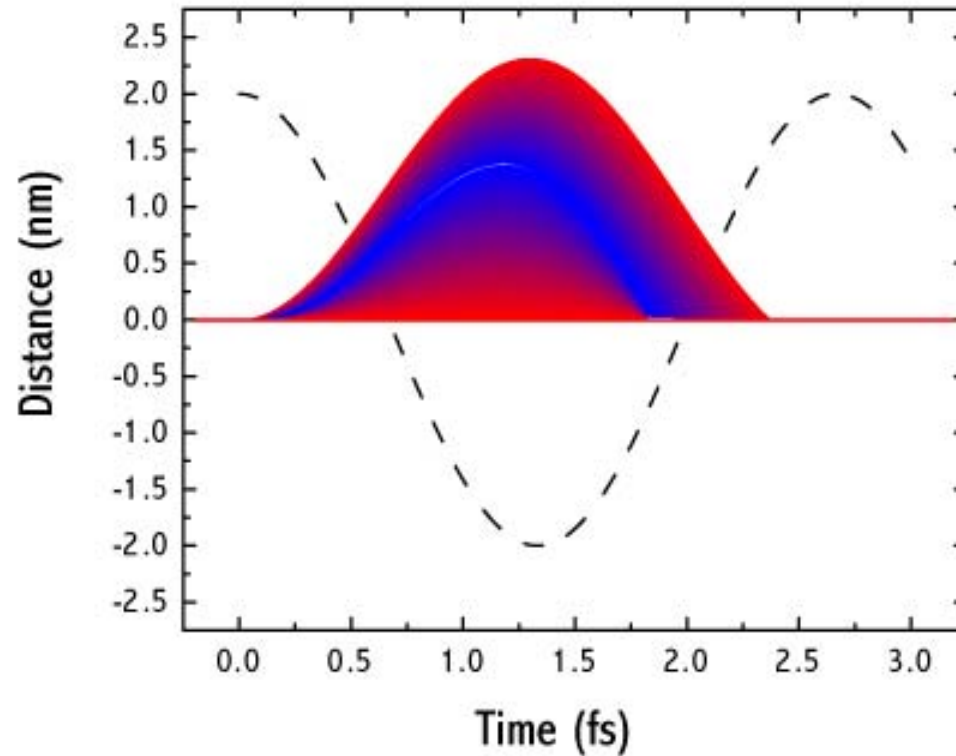
Y. Mairesse *et al.*, *Science*,  
**302**, 1540 (2003).

**Each order of harmonic is emitted at a different time**

**We use this to measure intra-molecular rearrangement following ionisation**

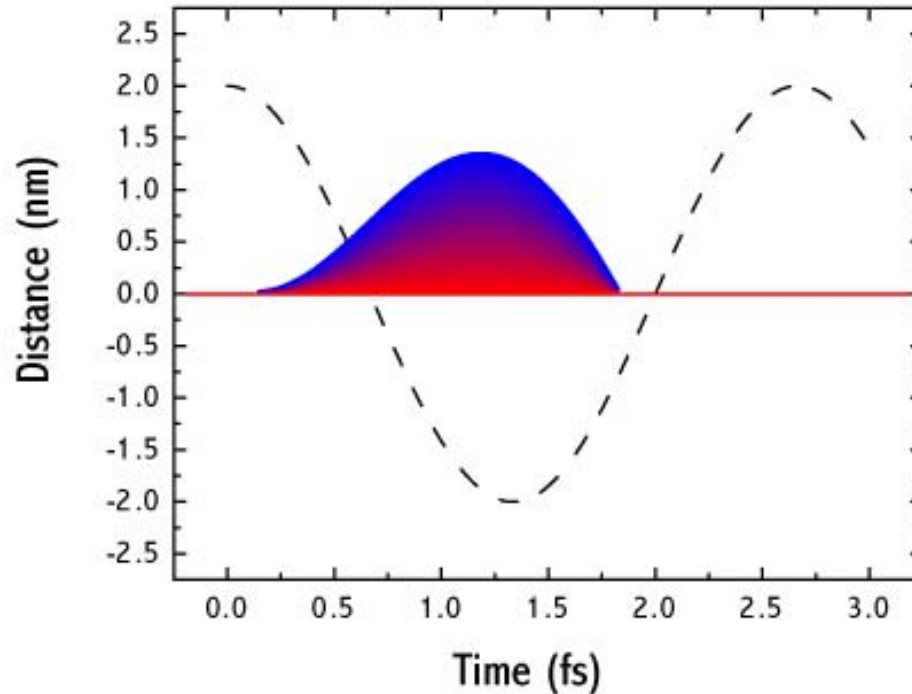
## Temporal chirp of electron wavepacket

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



# Temporal chirp of electron wavepacket

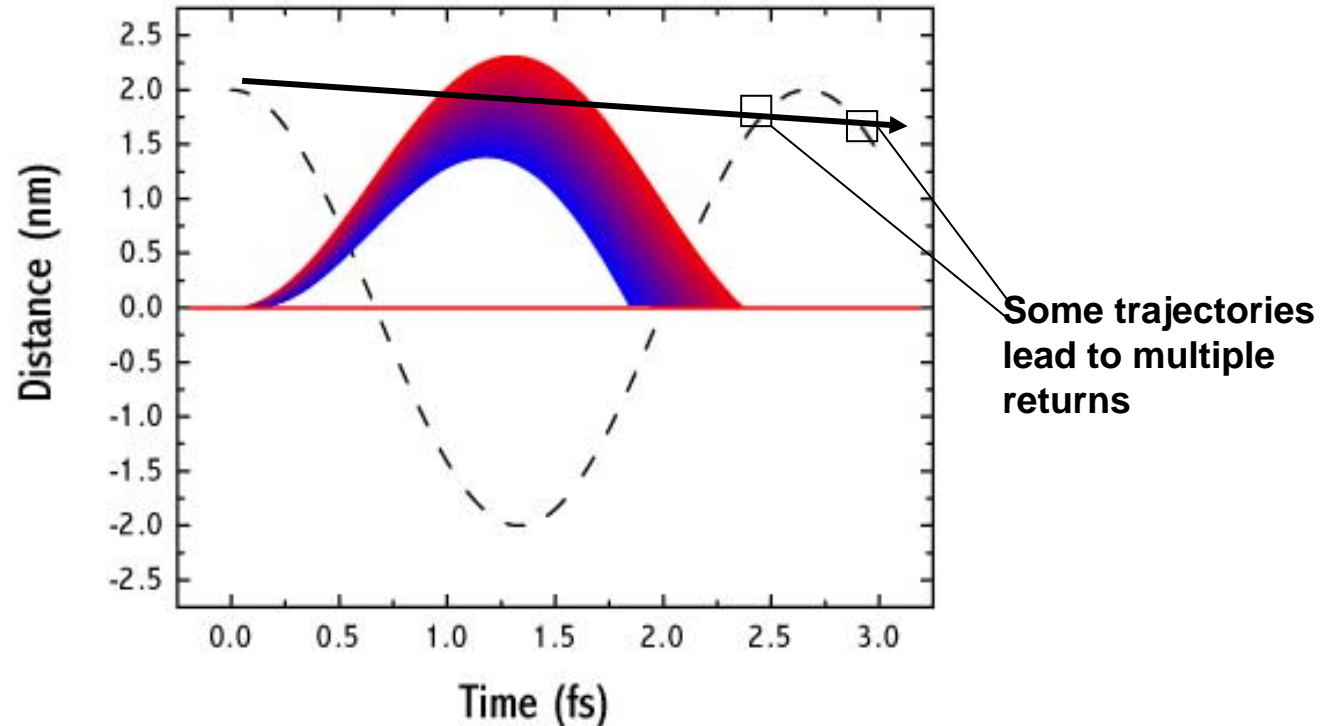
Temporal spread  $\sim 1$  fs for a 800 nm driving pulse (depends on optical cycle)



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

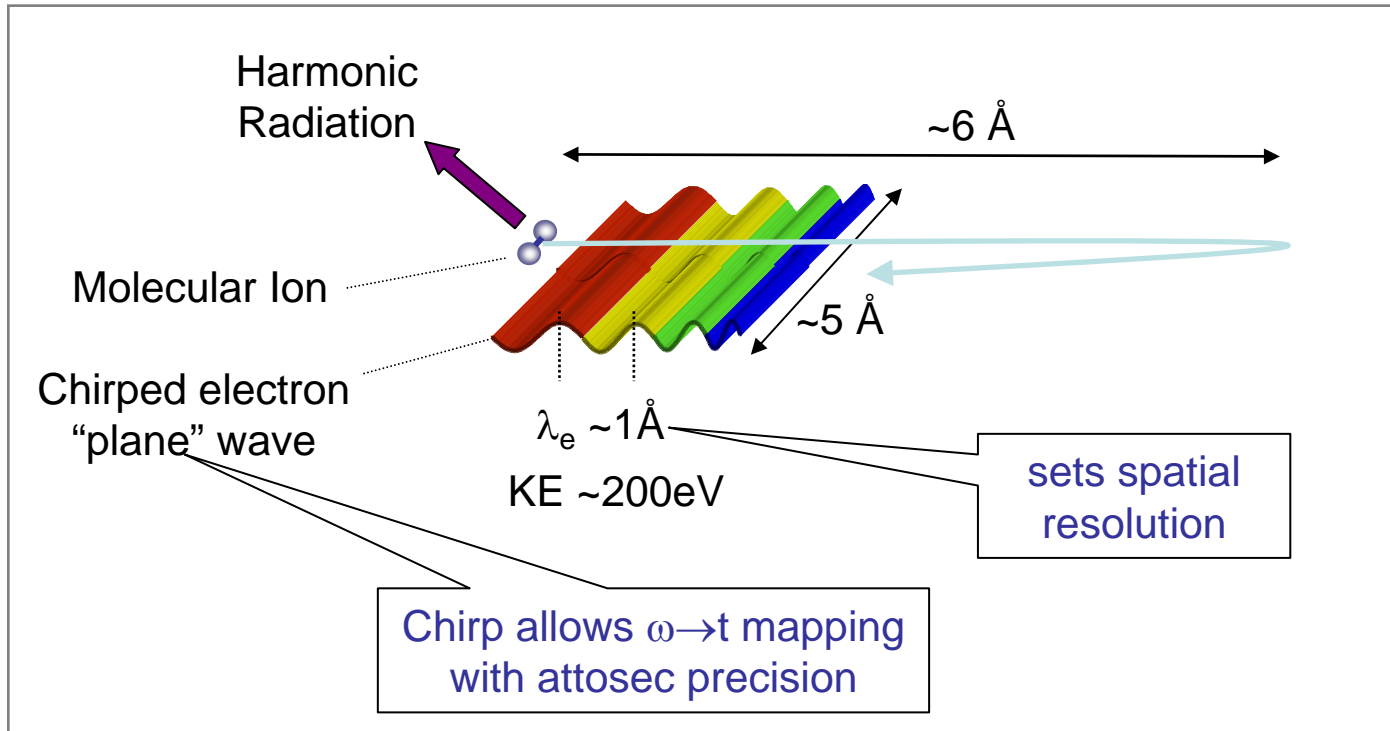
# Temporal chirp of electron wavepacket

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



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

# HHG as an ultra-fast structural probe



## Laser Parameters

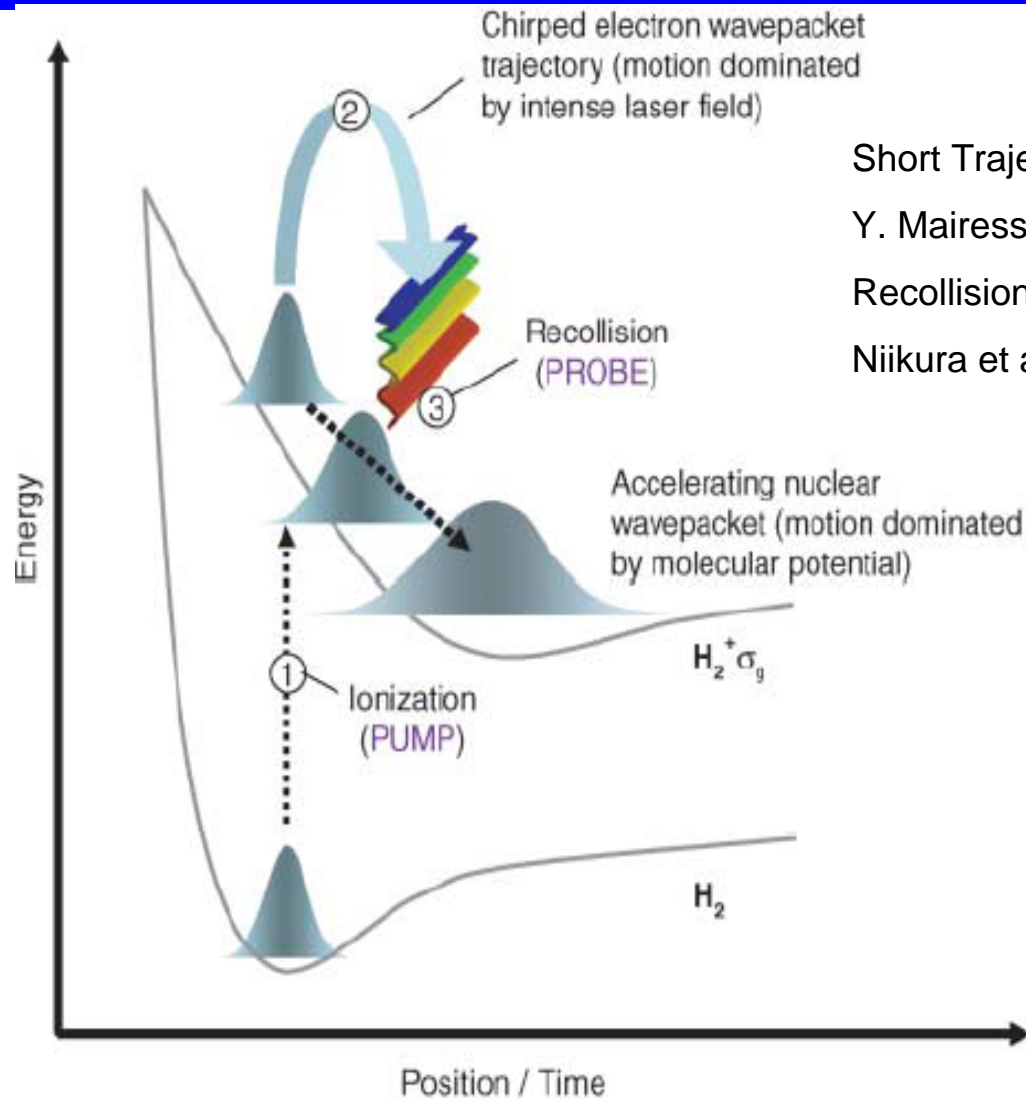
$$\lambda = 800 \text{ nm}$$

$$I = 10^{15} \text{ Wcm}^{-2}$$

Linearly Polarised

- Electron "beam" high current/ de Broglie wavelength  $\sim 10^{-10} \text{ m}$  for  $\sim 150 \text{ eV}$
- Recollision is highly coherent: produced from single electron orbital and manipulated with coherent laser light.
- Harmonic radiation encodes dynamical structural information from molecule...

# Part 2: HHG from molecules can be used to see fast nuclear dynamics in light molecular ions



Short Trajectories Selected:

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

Recollision probing concept

Niikura *et al.*, *Nature*, **417**, 917 (2002)



# Using HHG to probe ultrafast nuclear dynamics

Harmonic emission is sensitive to changes in the nuclear wavefunction, and will therefore be a probe of the subsequent nuclear dynamics .

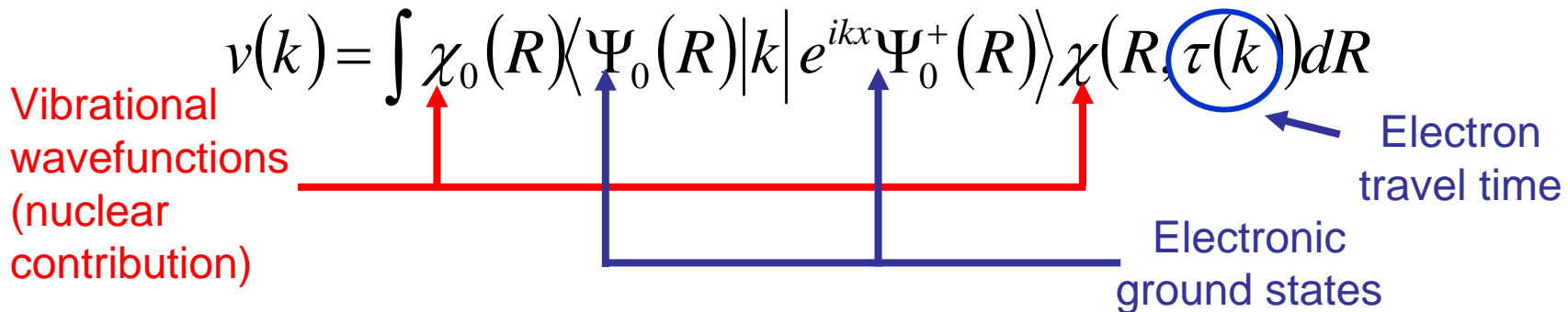
Ionisation also launches a nuclear wavepacket on the ionic PES.

The overlap decreases in time as nuclear wavepacket evolves. [Science, 312, 424 (2000).]



The chirp of harmonic emission allows us to encode time in frequency

A single harmonic spectrum has within it information about nuclear dynamics occurring on ionic PES during electron travel time.



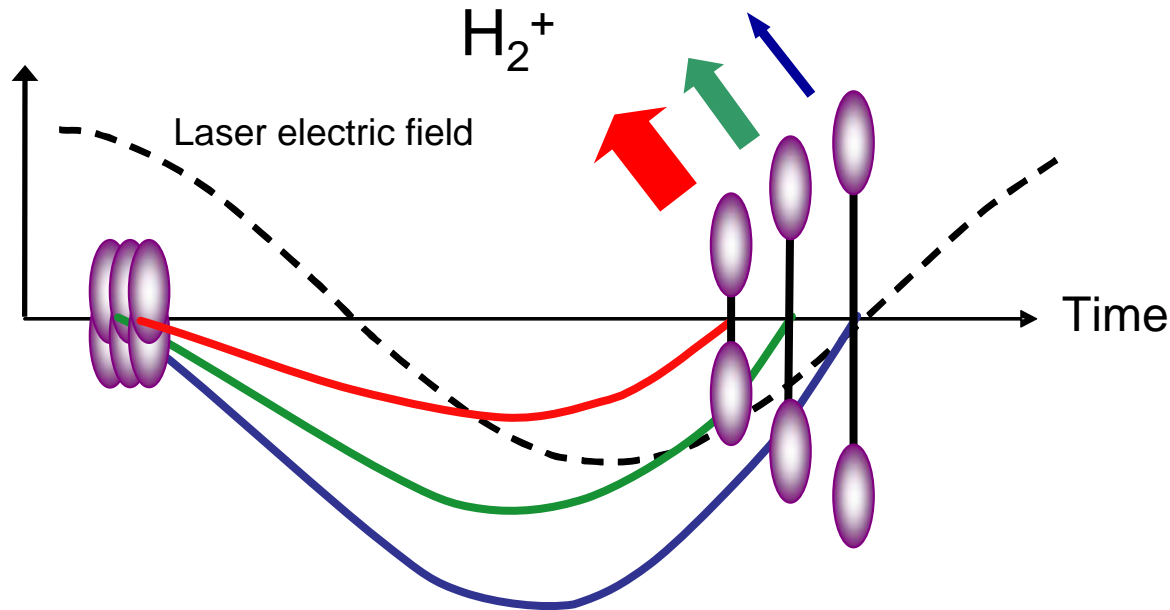
# ■ PACER:

## Probing attosecond dynamics by chirp encoded recollision

Intensity of each spectral component also depends on momentum distribution of returning electron wavepacket:

$$\psi_c = \int a(k)e^{ikx} dk$$

And on the momentum dependence of dipole moment....

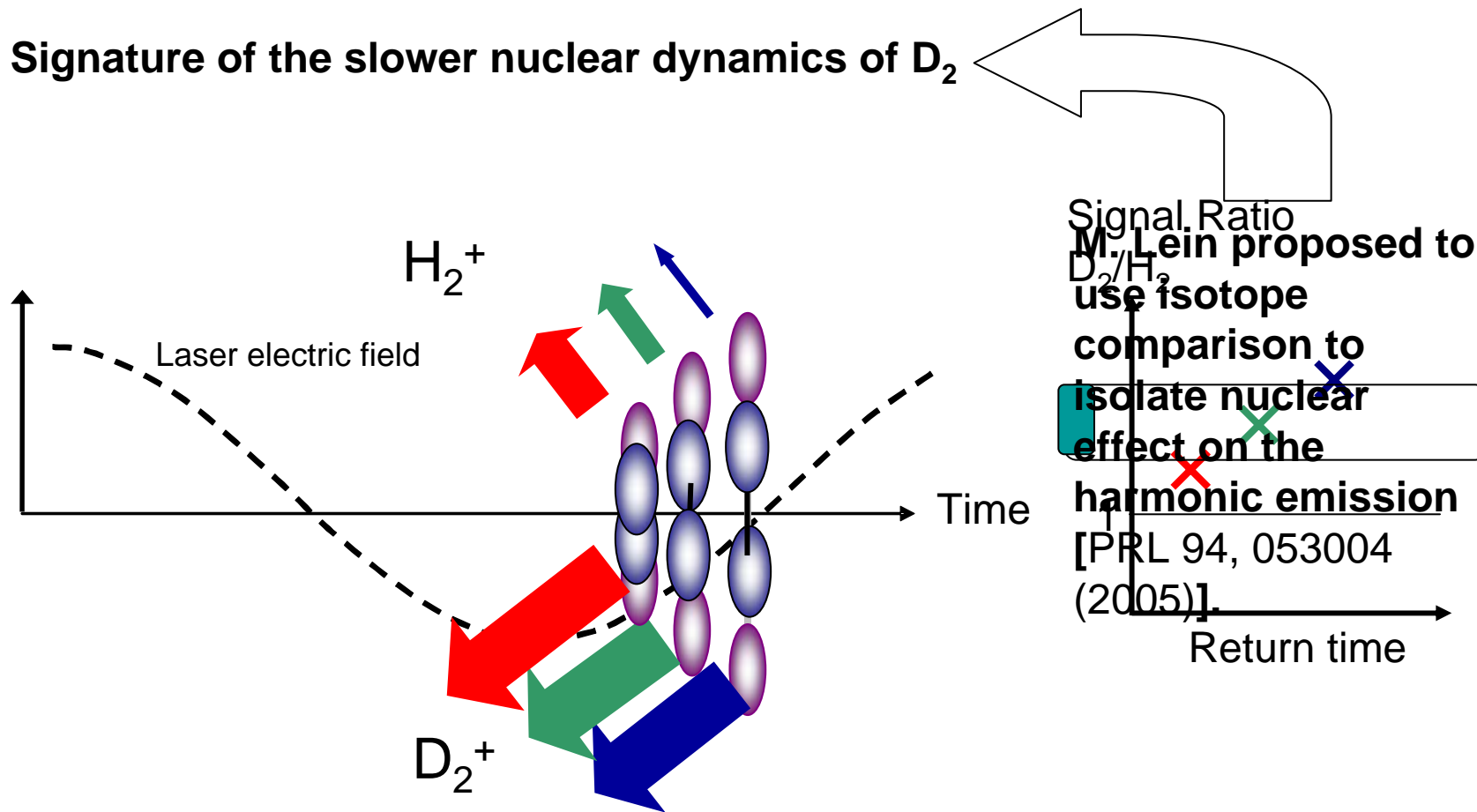


**M. Lein proposed to use isotope comparison to isolate nuclear effect on the harmonic emission [PRL 94, 053004 (2005)].**

# PACER:

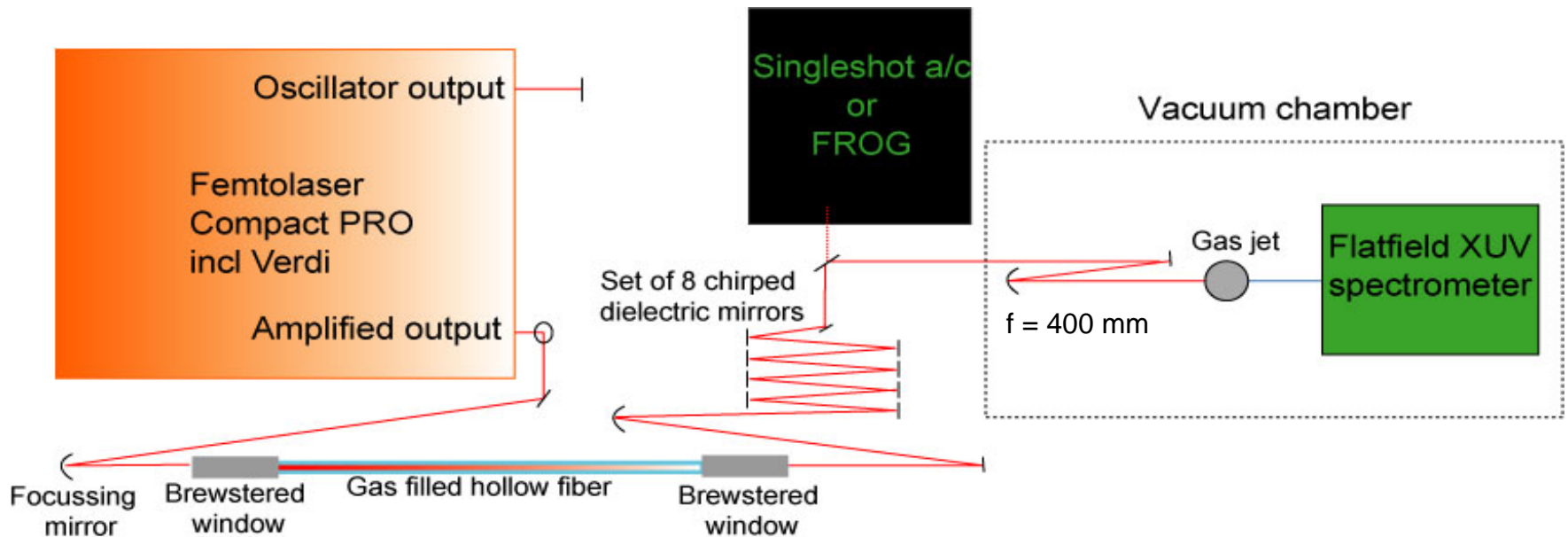
## Probing attosecond dynamics by chirp encoded recollision

Signature of the slower nuclear dynamics of  $D_2$



# Initial experiments

Used short (8fs) pulses to avoid “disturbance” of molecule prior to ionisation, e.g laser induced realignment or nuclear motion.



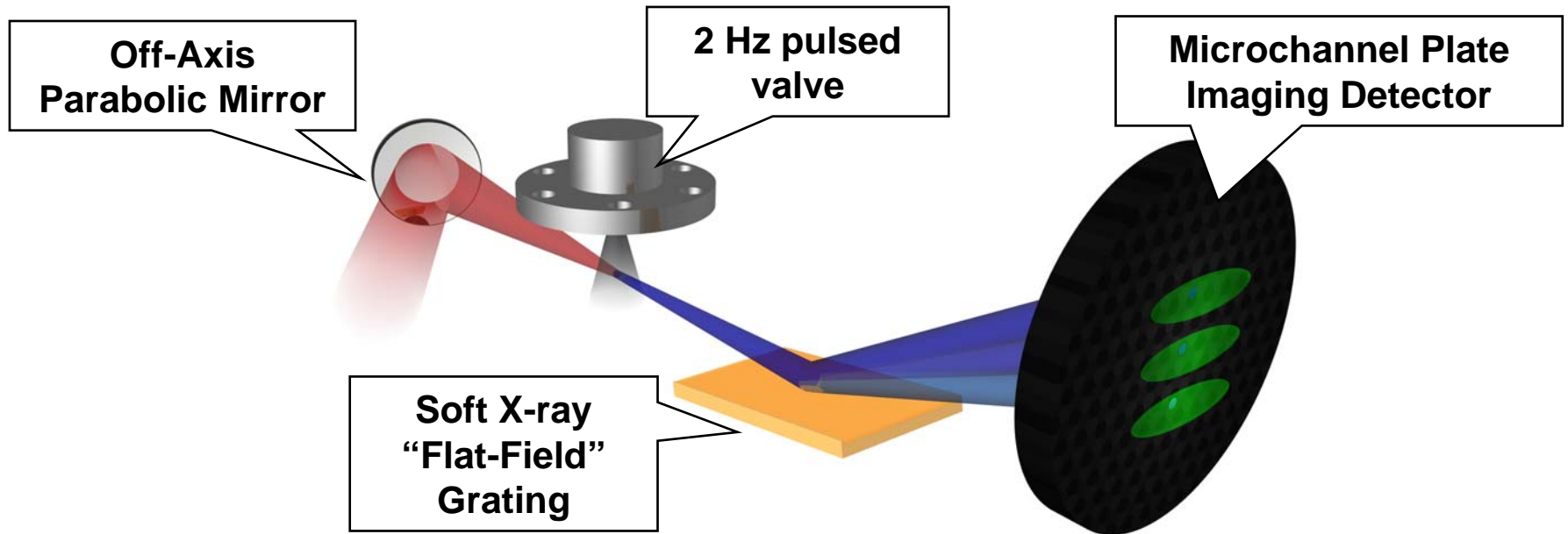
Robinson et al., App. Phys. B, **85**,4 (2006).

# Experimental approach part I

Focus 9 mm before jet to isolate short trajectories.

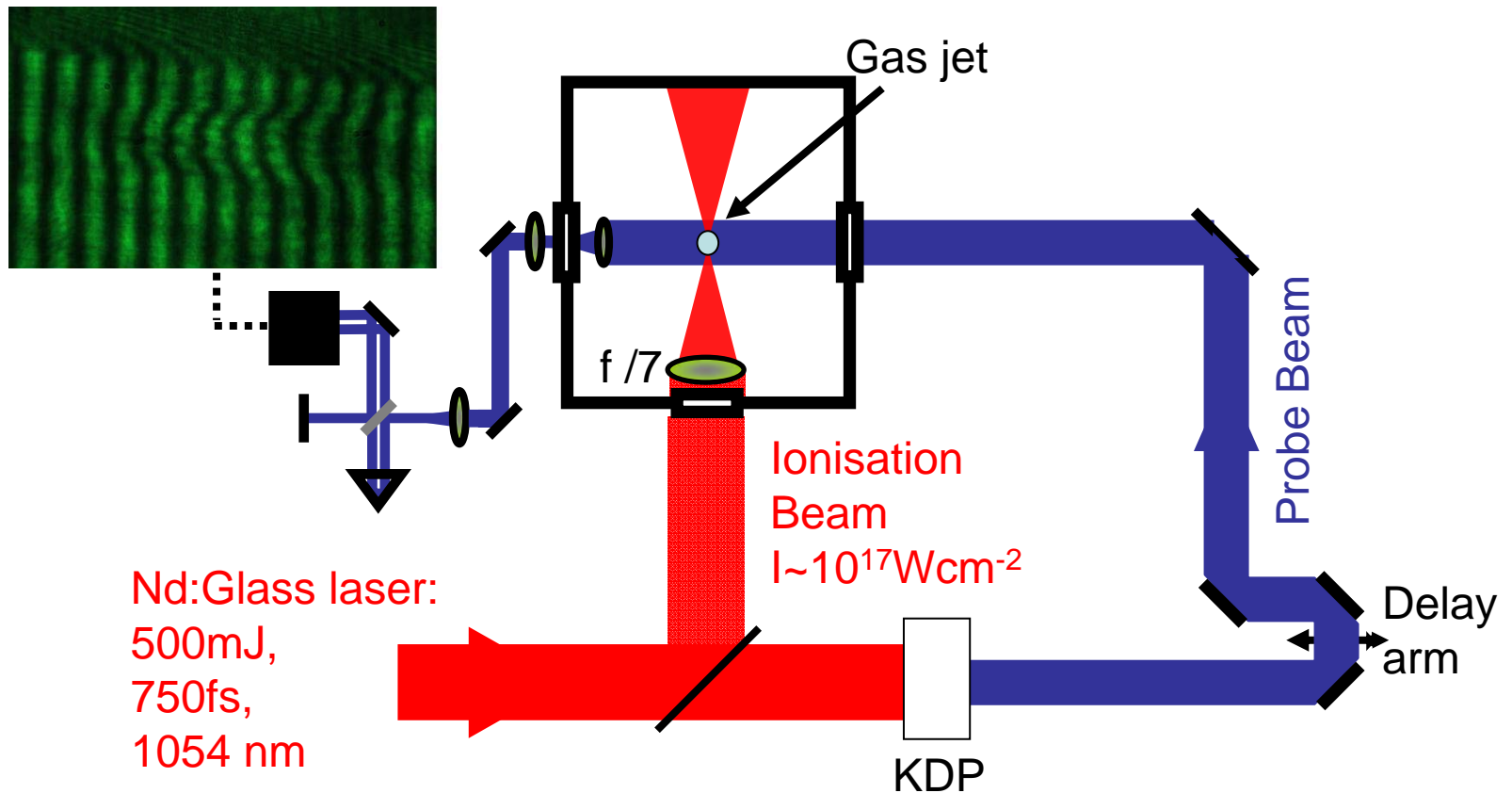
Intensity delivered at interaction region:  $2 \times 10^{14} \text{ Wcm}^{-2}$ : shot-to-shot fluctuation <3%, **monitored between data runs.**

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

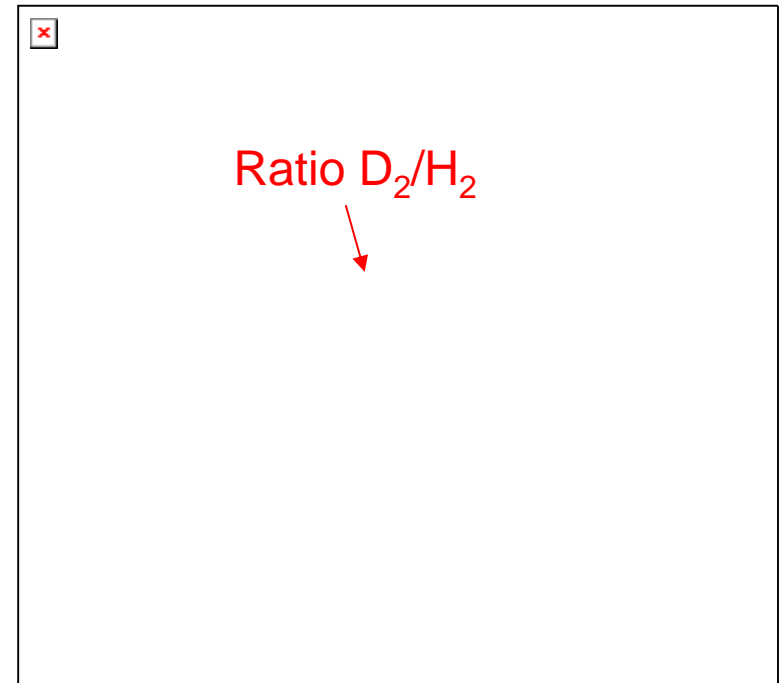
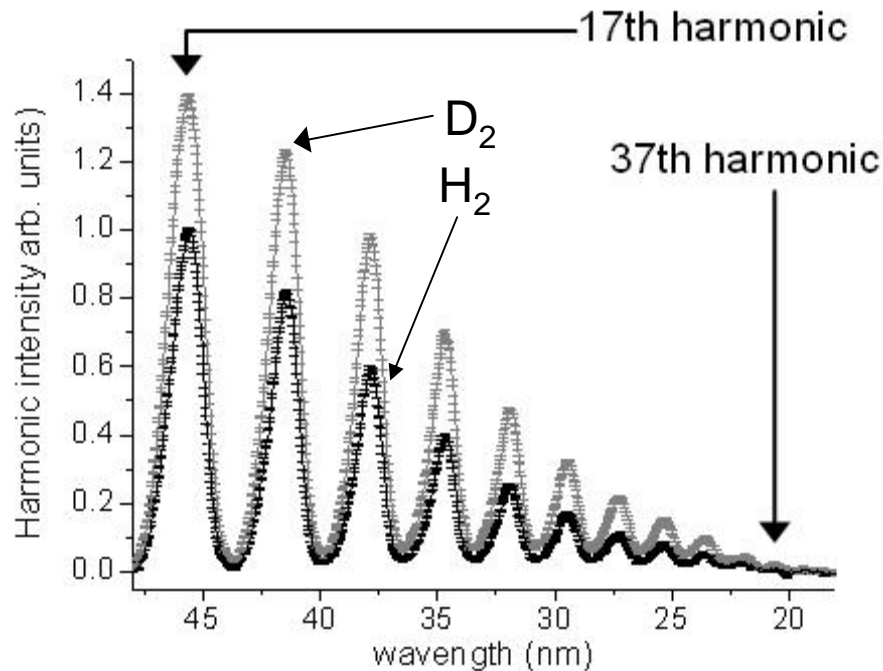


## Experimental approach part II

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



# Ratio of harmonic signals increases with electron return time

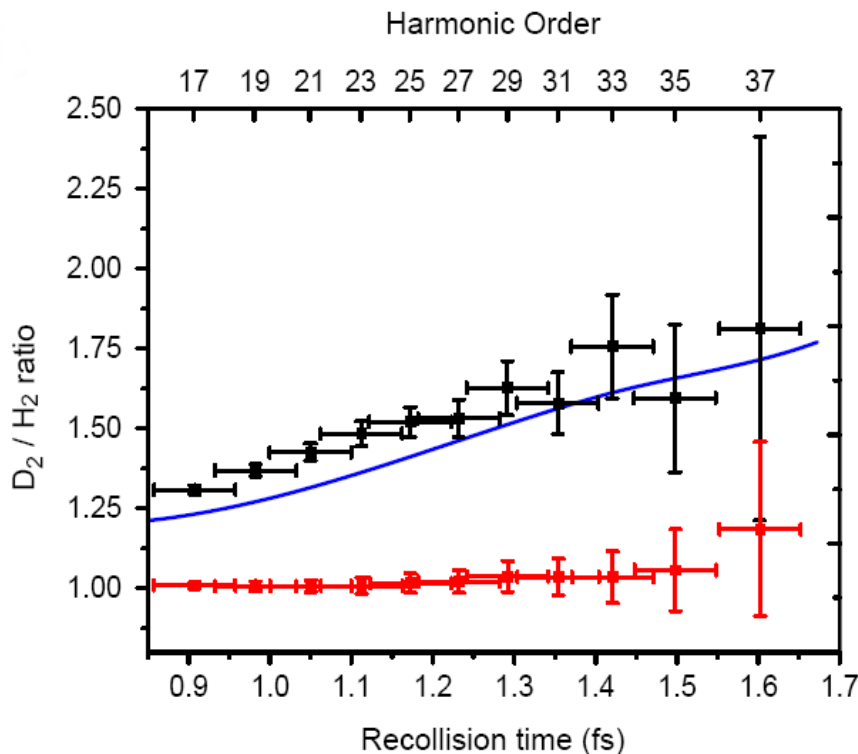


Effect robust to variations in gas jet backing pressure.

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

# PACER: a new technique for probing ultrafast nuclear motion in molecules

We attribute the increasing ratio to the differing nuclear dynamics of  $D_2$  and  $H_2$ .



Calculations performed in strong-field approximation, using known molecular potentials, allowing for short trajectories only.

Include nuclear motion, and two-centre interference effects.

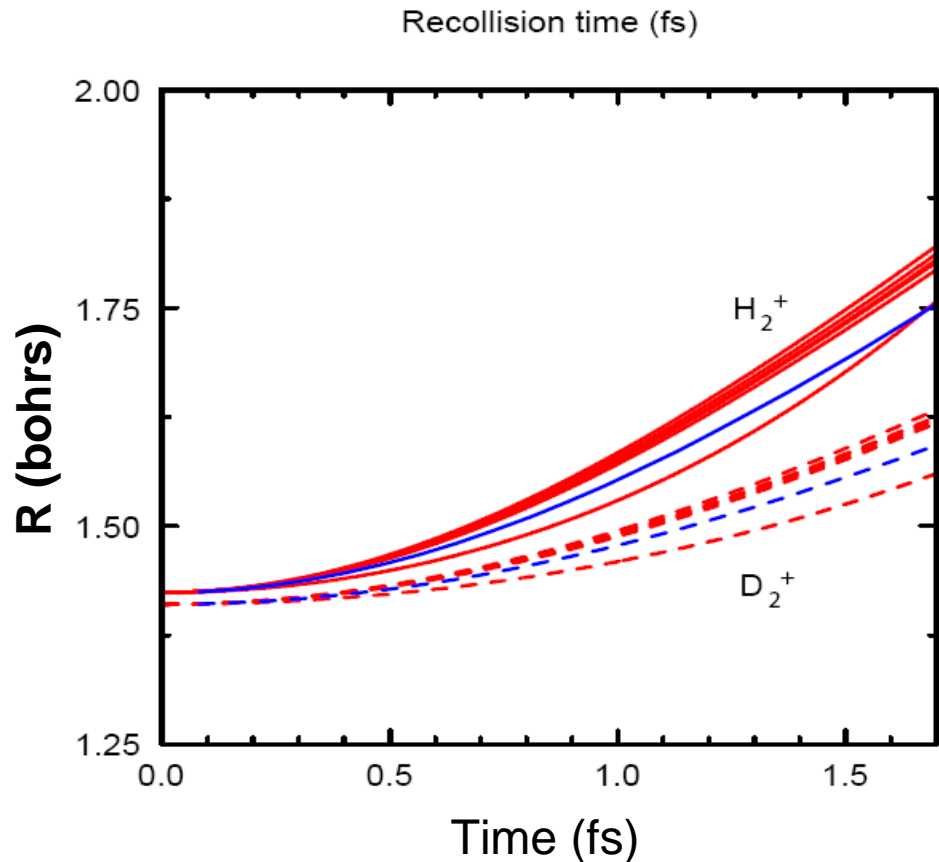
Blue: single molecule response, averaged over randomly aligned ensemble, including re-absorption of generated harmonics.

**Experiment and theory match very well!**



# PACER: a new technique for probing ultrafast nuclear motion in molecules

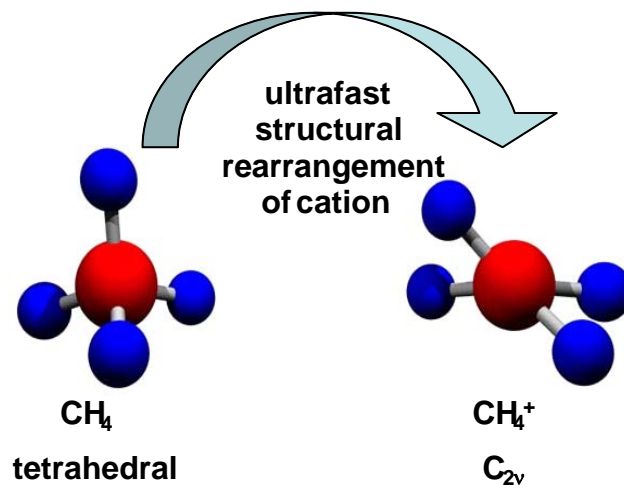
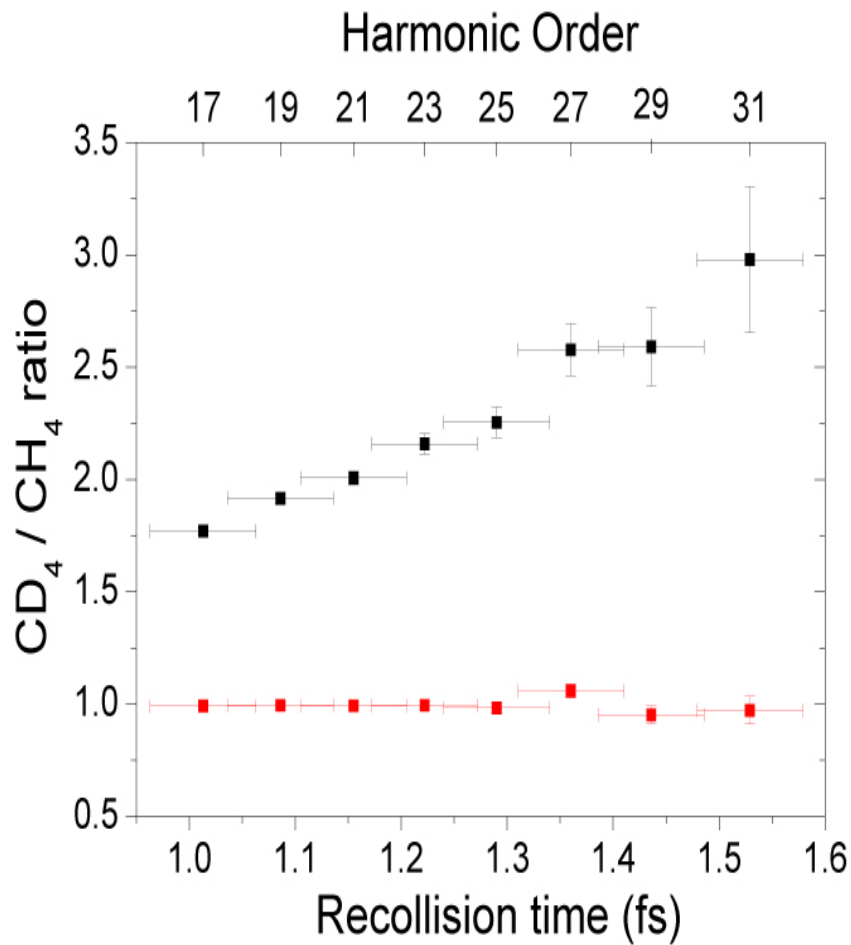
The increasing ratio can be used to gain information about the nuclear motion.



We have made a measurement with **~100 as time resolution**, using 8 fs pulses: launch of electron and nuclear wavepackets is synchronous by nature of the process.

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

# The nuclear correlation function ratio between $\text{CH}_4$ and $\text{CD}_4$ provides the first evidence of an ultra-fast rearrangement of methane upon ionisation



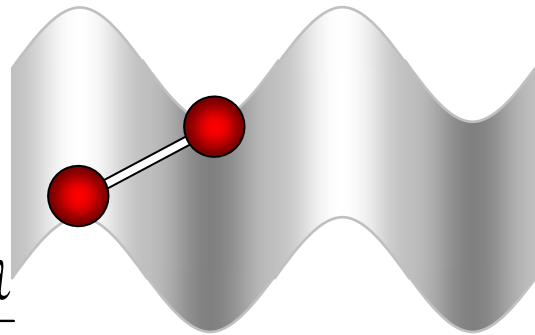
Baker *et al* Science **312**, 424 (2006)  
Marangos *et al* PCCP **10**, 35 (2008)

# Does two-centre interference play a role?

Interference between emission from different centres can enhance or suppress harmonic emission.

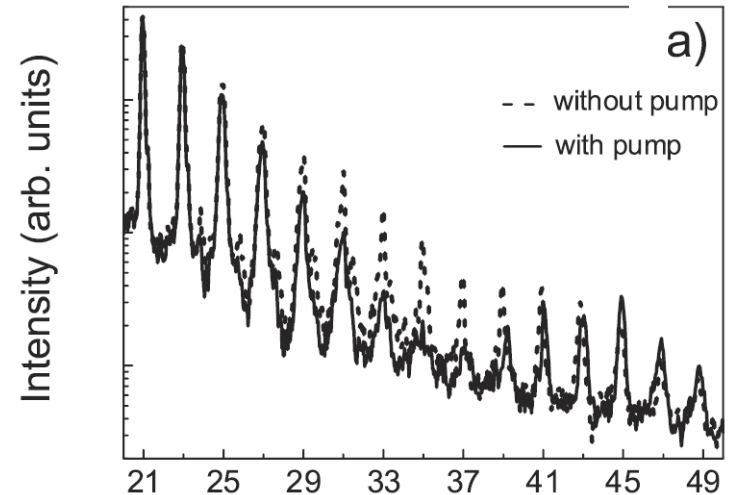
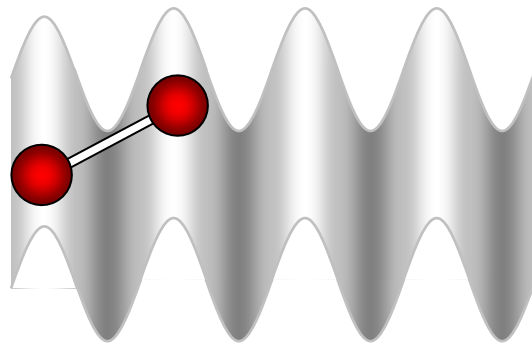
Destructive  
interference

$$R \cos(\mathcal{G}) = \frac{(n+1)\lambda}{2}$$



Constructive  
interference

$$R \cos(\theta) = n\lambda$$

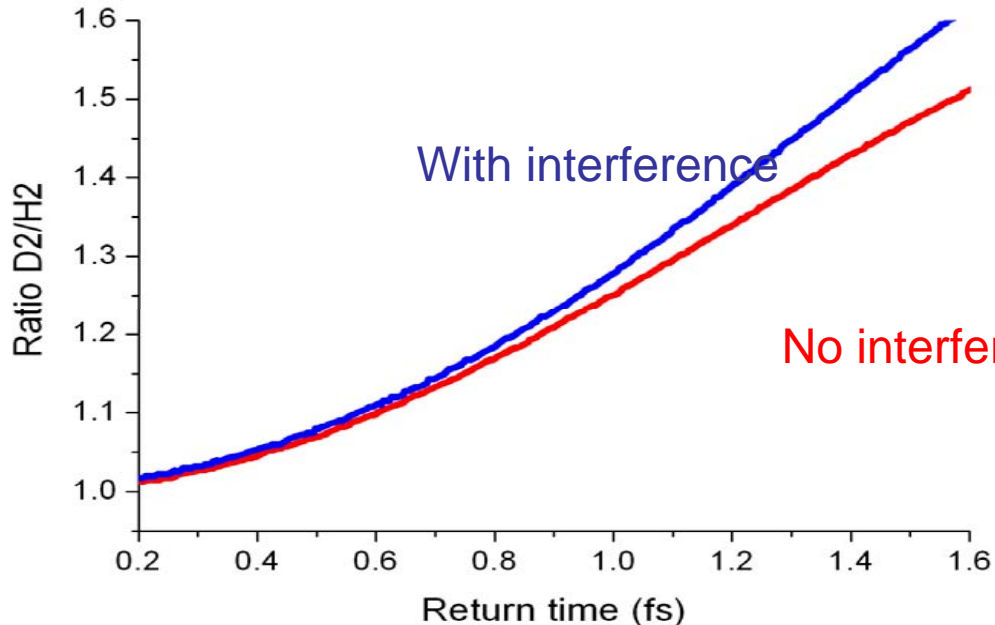


Vozzi et al.,

PRL 95, 153902 (2005)

# Does two-centre interference play a role?

For 8 fs pump pulses, sample alignment is negligible, therefore effect small, but discernable.



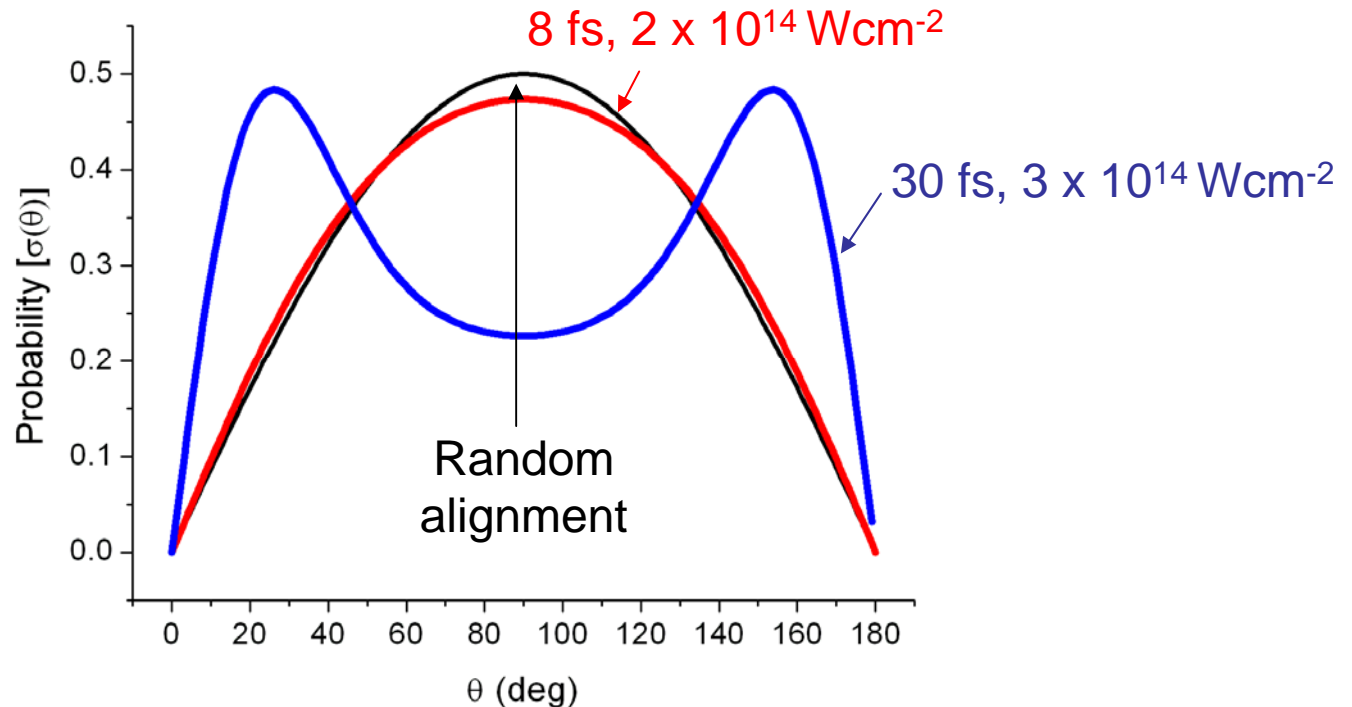
Effect small because:

- 1/ At equilibrium separation TCI will be at HHG orders beyond cut-off
- 2/ Sample isotropic so few molecules aligned along polarisation direction, most nearly perpendicular so no TCI

**Can we tailor conditions to observe strong two-centre interference effects?**

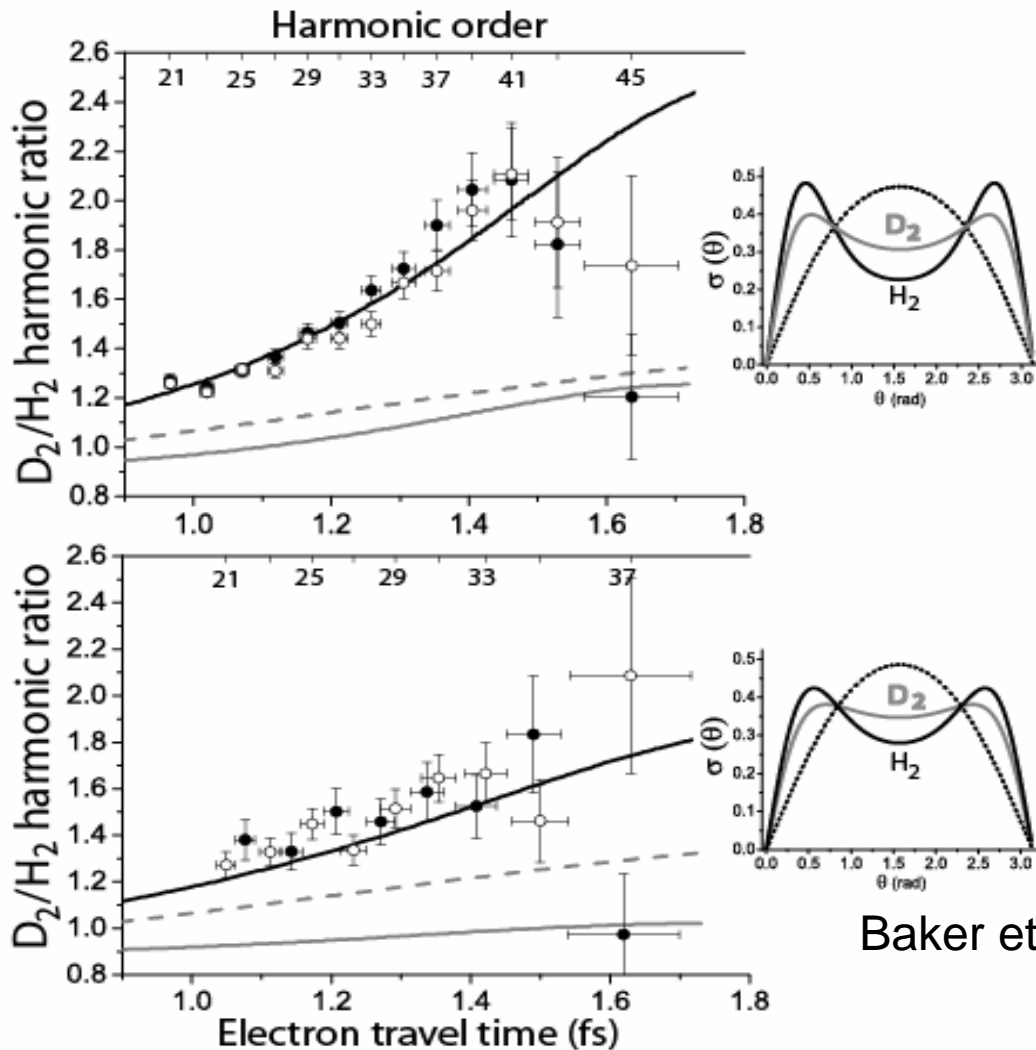
# Can we tailor conditions to observe strong two-centre interference effects?

**Stronger degree of alignment:** Repeat PACER measurement using 30 fs pulses at 800nm (conducted at Advanced Laser Light Source, Canada).



**Ensure short enough electron wavelengths:** Increase intensity compared to previous measurements.

# Experiment to observe strong two-centre interference effects show enhanced ratio at times predicted by theory

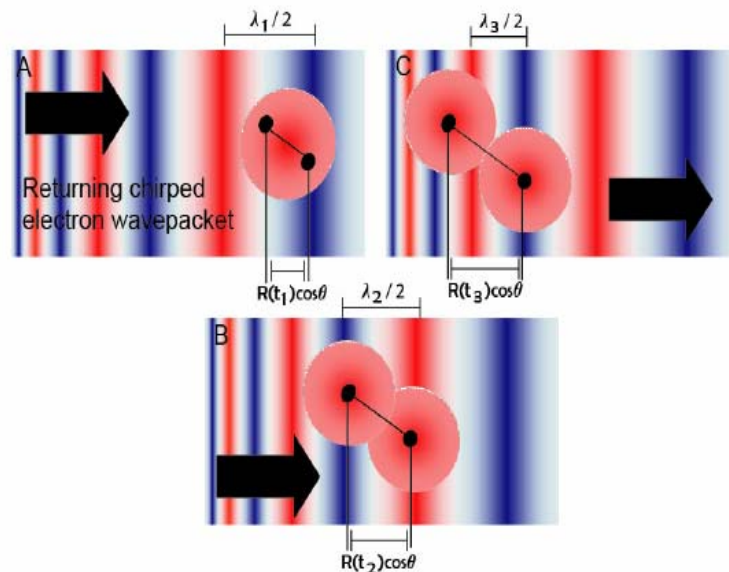
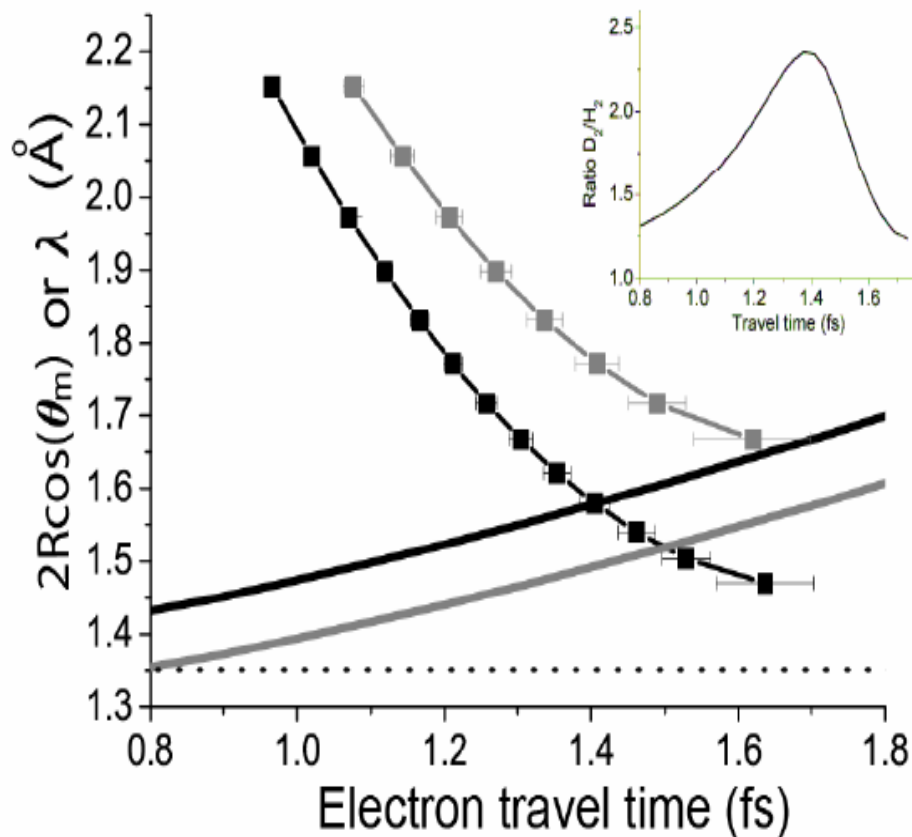


Ratio of harmonic signals in  $D_2$  and  $H_2$  at intensity  $3.0 \pm 0.1 \times 10^{14} \text{ Wcm}^{-2}$  (upper) and  $2 \times 10^{14} \text{ Wcm}^{-2}$  (lower). Black line prediction from SFA. Grey line shows SFA calculation in which the nuclear motion has been neglected. Dotted line shows SFA calculation in which two-centre interference is neglected.

Baker et al, PRL, **101**, 053901(2008)

Is it reasonable to expect two-centre interference for these conditions?

# A simple model accounting for the temporal variation of both inter-nuclear distance and electron de Broglie wavelength provides a qualitative explanation



Baker et al, PRL, **101**, 053901(2008)

This appears to be a *transient* two-centre interference, occurring only as the nuclear wavepacket evolves

# Summary

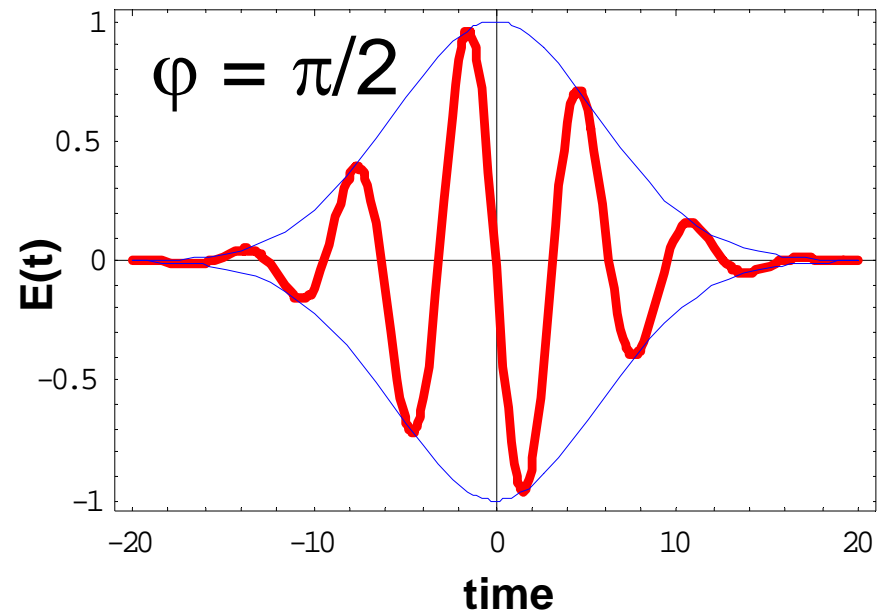
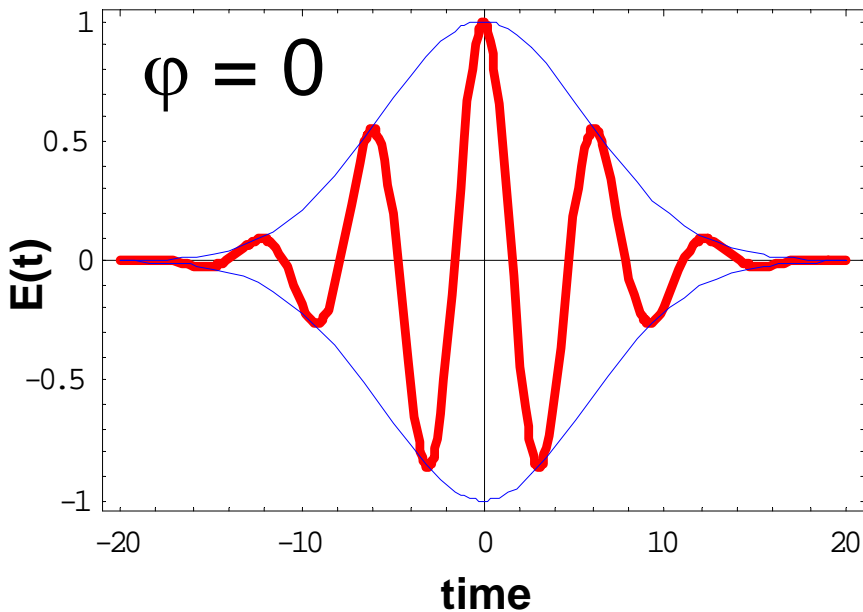
- Proton dynamics can be imaged using inherent chirp within the harmonic emission process
- Observations of transient 2-centre interference have recently been made
- This technique is applicable in principle to measurement of electronic dynamics also.



## Part 3: Sculpting the electron recollision by control of the few-cycle field

$$E(t) = E_o(t) \cos(\omega t + \varphi)$$

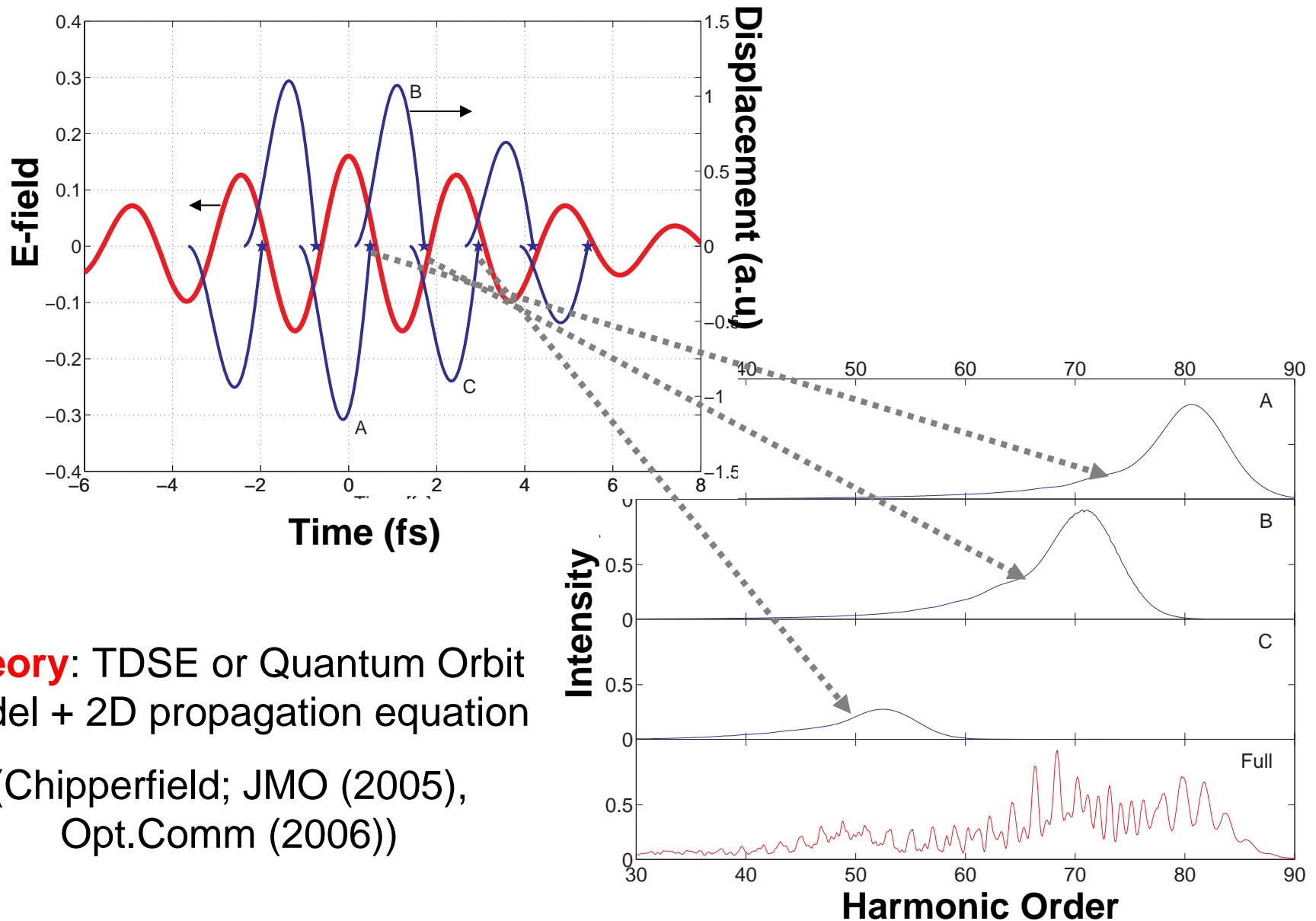
**CEP**



The CEP is an important parameter for many strong-field processes, e.g. attosecond pulse generation.

Determination of the value of CEP is a challenging problem

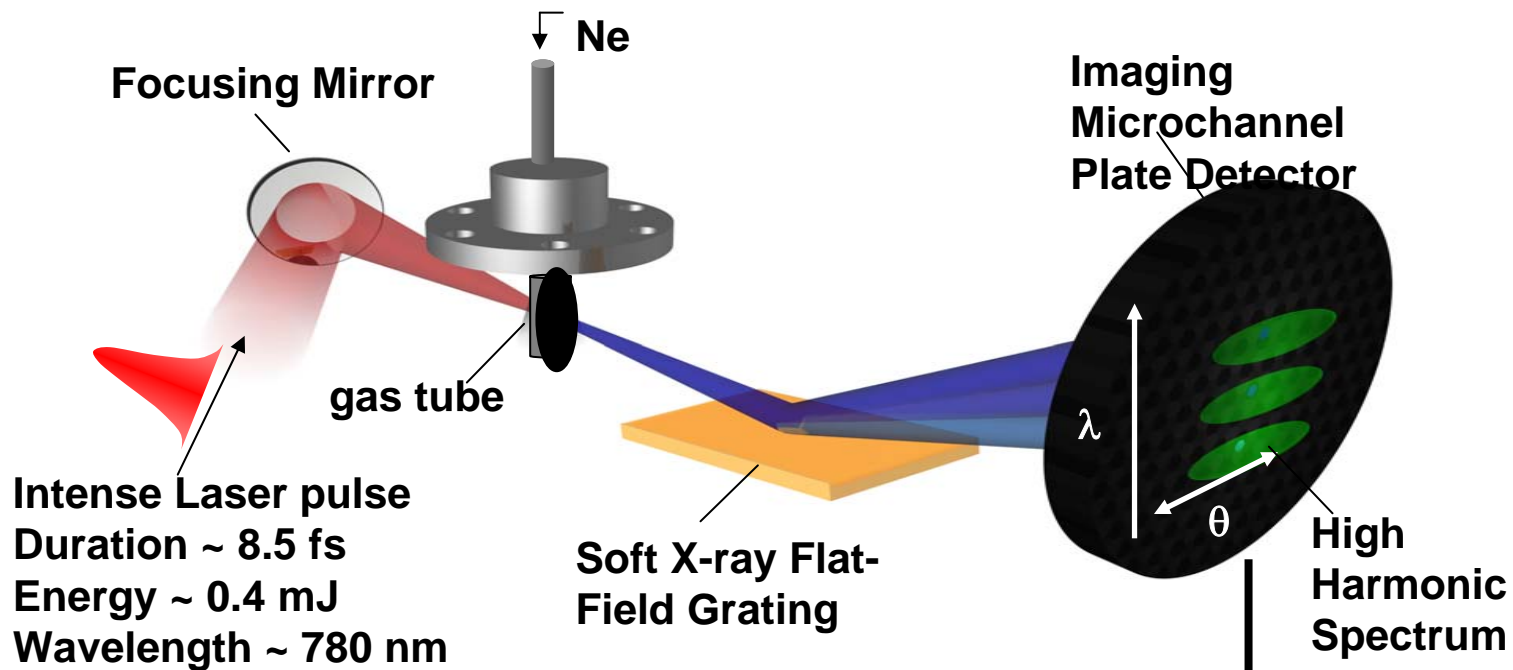
# Our modelling showed that “half-cycle cut-offs” (HCO’s) are determined by the CEP



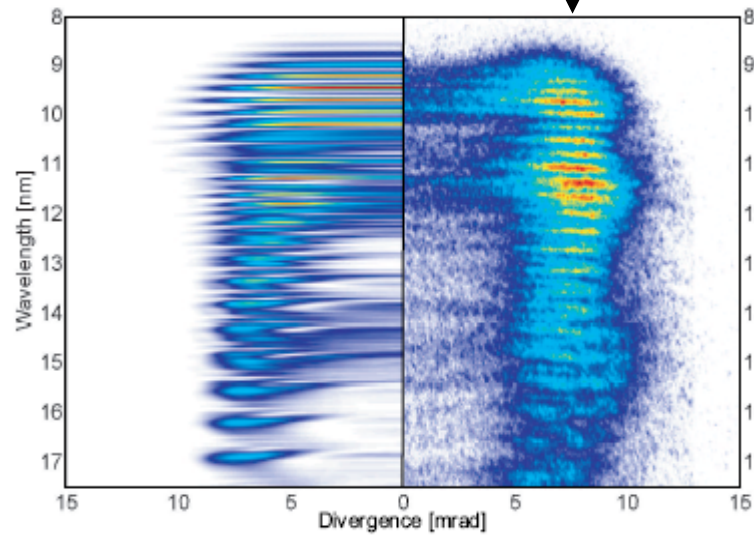
**Theory:** TDSE or Quantum Orbit model + 2D propagation equation

(Chipperfield; JMO (2005), Opt.Comm (2006))

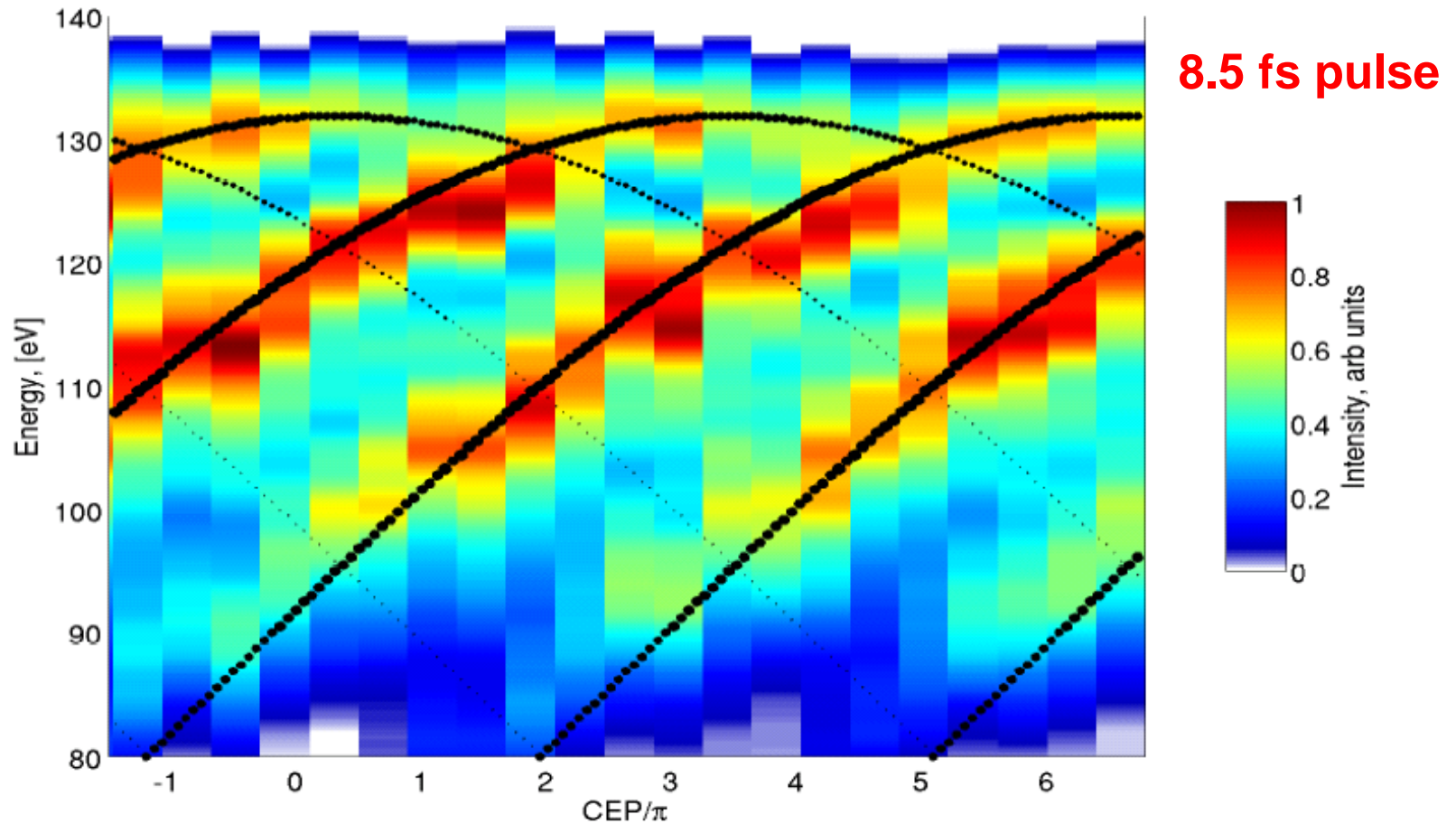
# CEP control of HHG spectra demonstrated



**Measured spatially resolved spectra were in excellent agreement with a SFA calculation including propagation.**



# Excellent match between theory and experiment for HCO shifts as function of CEP



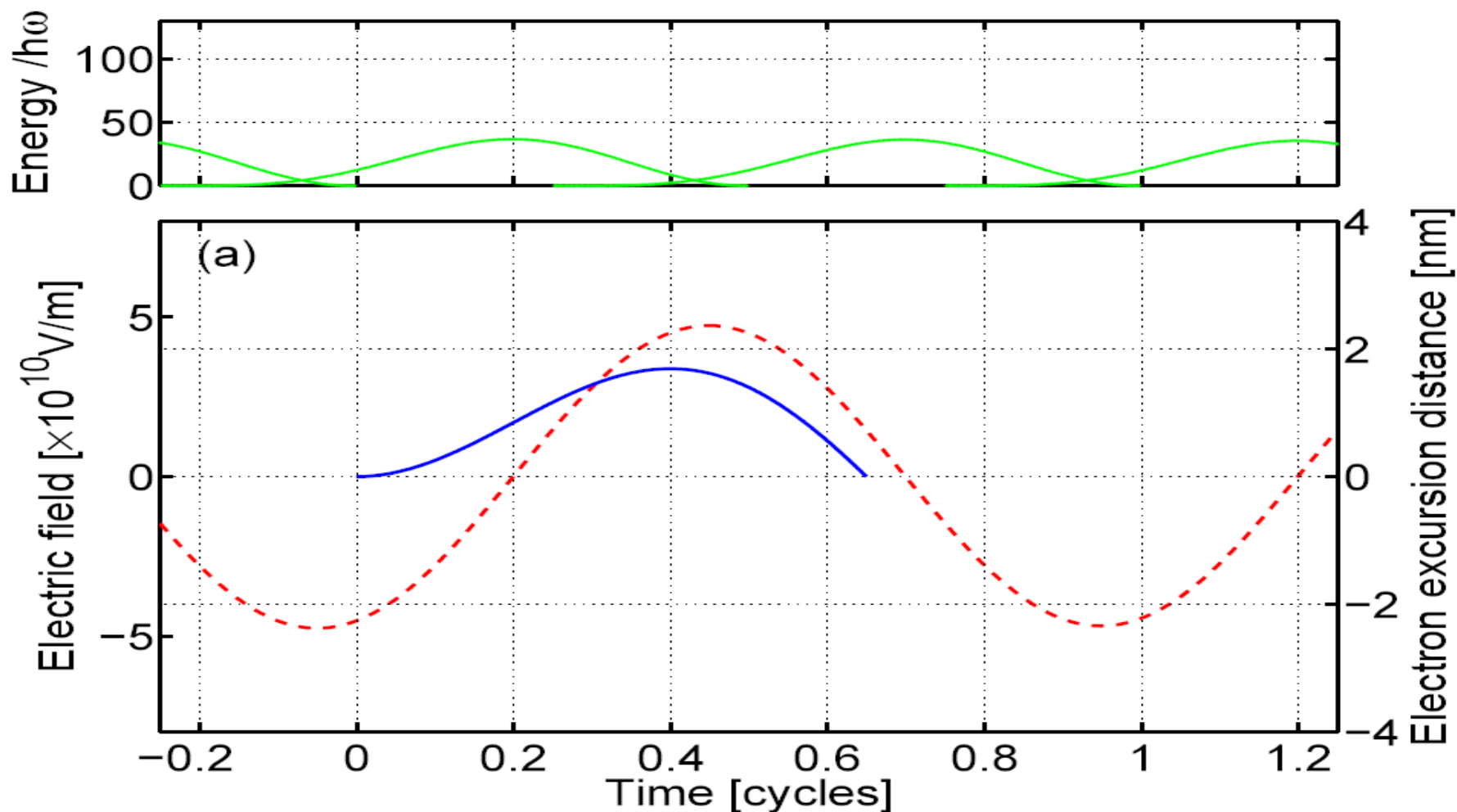
**This provides basis for new CEP measurement technique !**

In principle full electric field waveform can be retrieved from the measurement  
*Nature Physics*, **3**, 52 (2007)

# Part 4: Optimum waveform for the highest energy recollision in a strong field

- There are many instances when we would like to optimise the recollision cut-off energy whilst preserving a reasonably high recollision probability.
- What is the optimum electric field waveform that achieves this for a given total energy and periodicity?

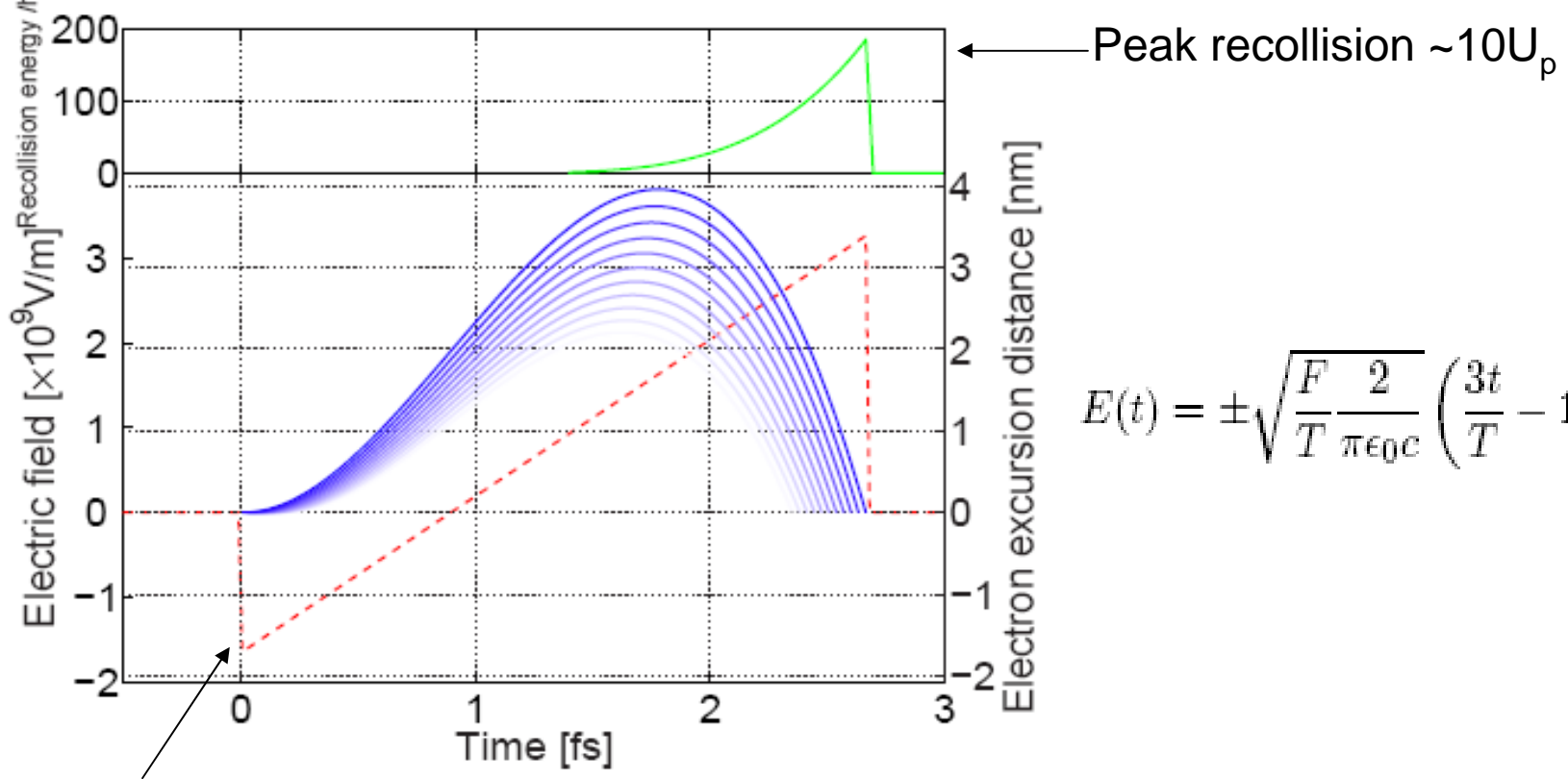
## A sine wave drives electron recollisions with a maximum energy $3U_p$



Classical simulations of the electron trajectories generated by a 10 cycle 800nm field with a peak intensity of  $3 \times 10^{14}$  Wcm<sup>-2</sup>,

# A sawtooth waveform with an DC offset is found to give a factor ~3 increase in recollision energy for a fixed periodicity and laser energy compared to a sinusoid

Fluence within cycle equivalent to the 800nm sine wave



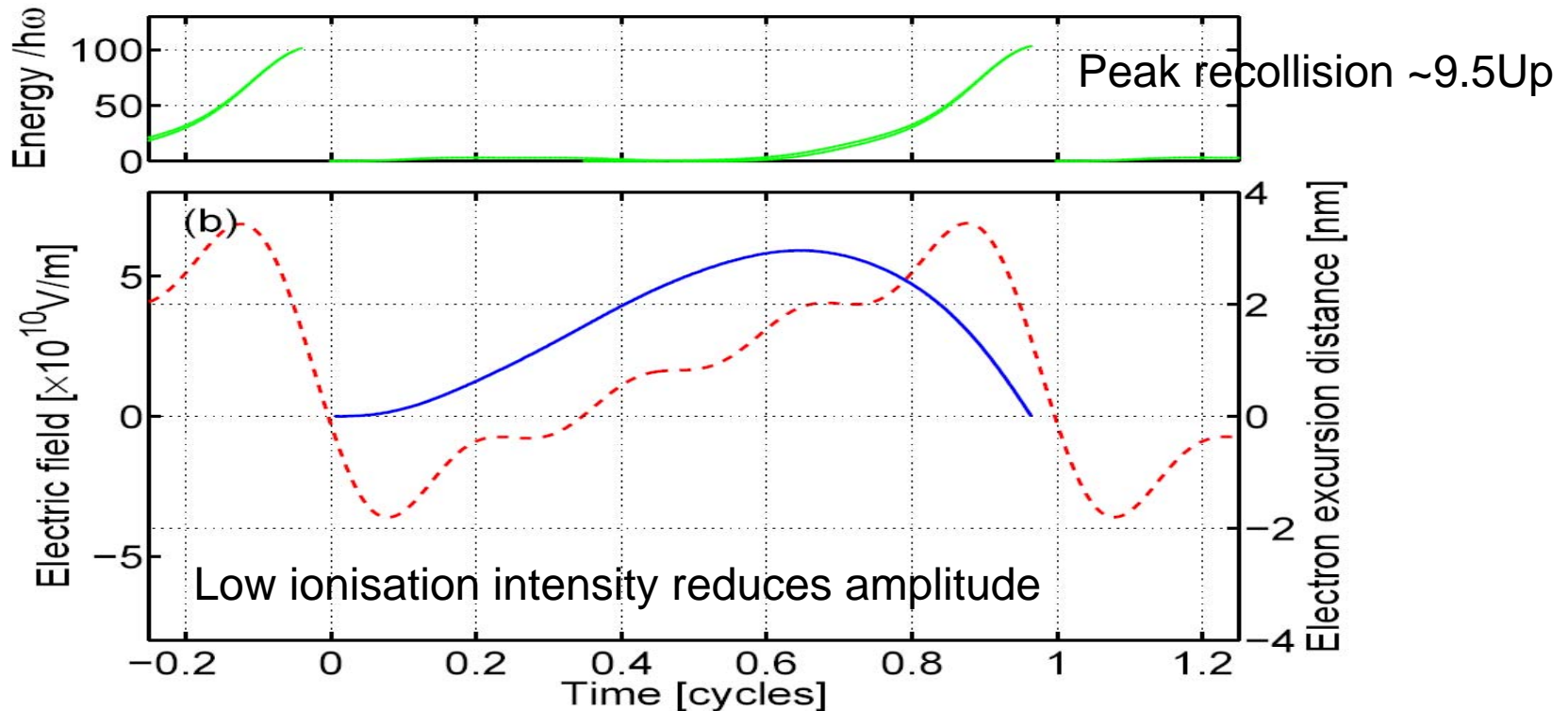
$$E(t) = \pm \sqrt{\frac{F}{T} \frac{2}{\pi \epsilon_0 c}} \left( \frac{3t}{T} - 1 \right)$$

DC Off -set  $\frac{1}{2}$  of amplitude

**Waveform maximizes the electron reacceleration energy prior to recollision**

# A waveform approximating to this optimum can be synthesized from a finite number of harmonics of the laser field

Waveform synthesized from 800nm + 400nm + 267nm + 200nm+DC

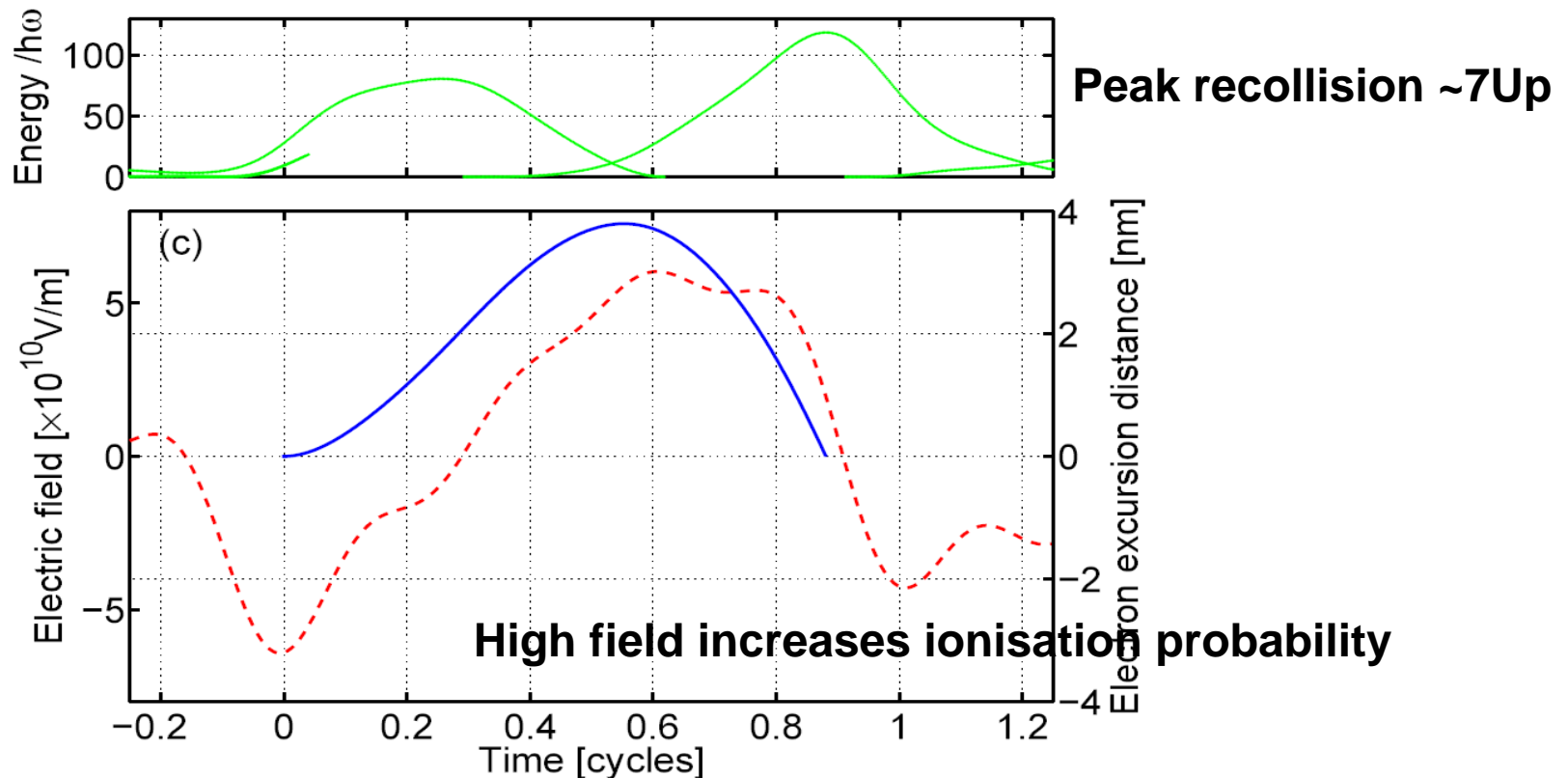


The optimum sawtooth waveform with all frequencies higher than the fourth harmonic removed (GA optimisation).



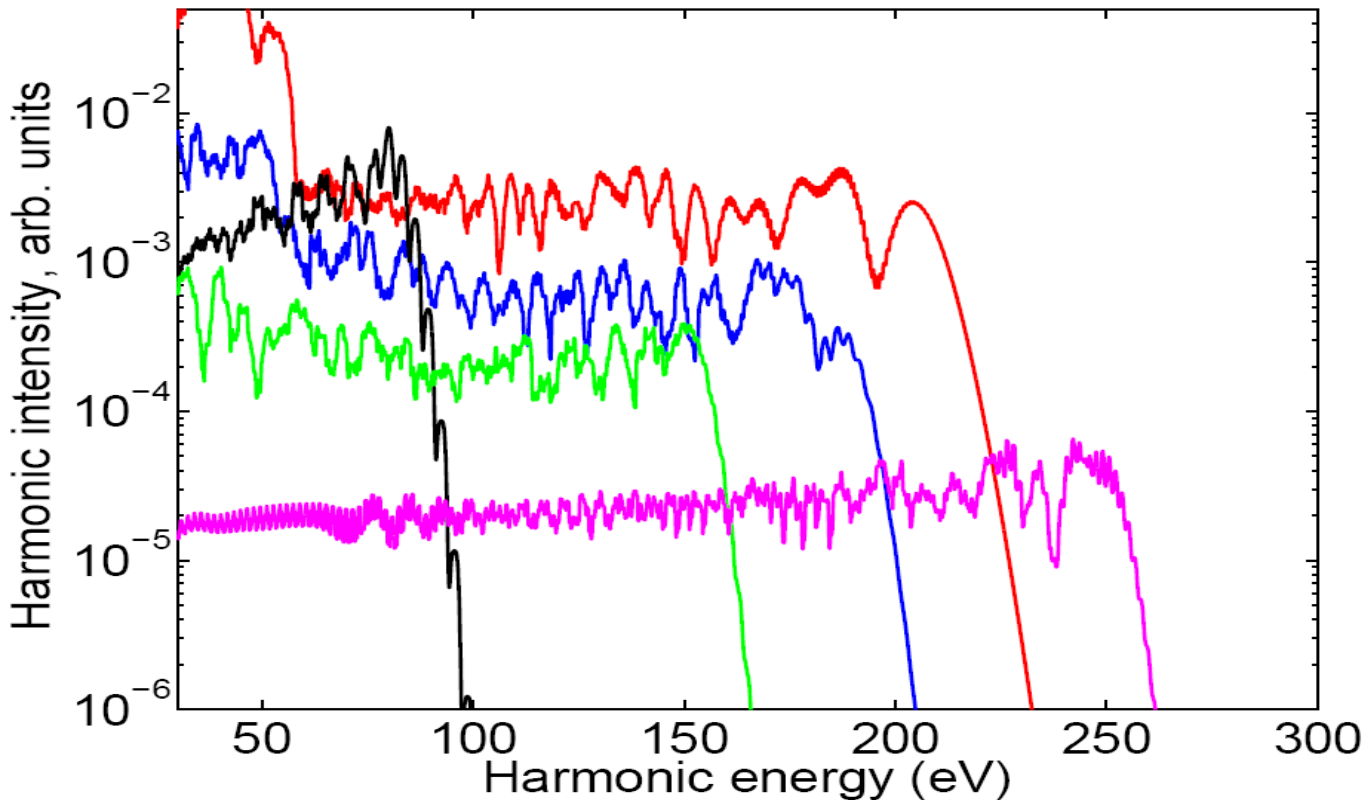
# A better waveform can be synthesized that has a large field intensity also at the ionisation time

Waveform synthesized from 800nm + 400nm + 267nm + 200nm+1600nm



An optimised waveform found using a genetic algorithm, composed of the 800nm fundamental field, a 1600nm field, with half the energy of the fundamental, and weak second, third and fourth harmonics.

# This waveform also overcomes the efficiency reduction of using a longer wavelength field



HHG calculated via SFA, including the effects of wavepacket spreading and neutral depletion. [black] generated by an 800nm field, [green] 1200nm field [pink] a 1600nm field. [red] 800nm field plus a 1600nm field at half the energy of the 800nm and harmonics up to the fourth order [blue] as the red spectrum, but without the second, third and fourth harmonic components.

# HHG polarisation properties can be controlled by using two colour perpendicularly polarized fields

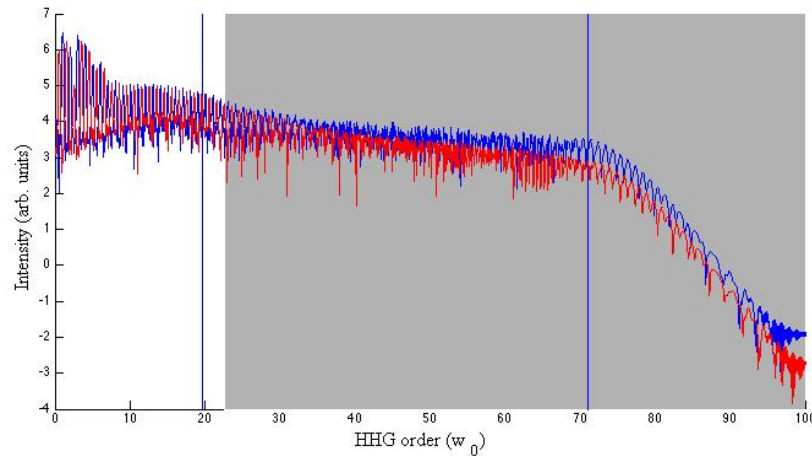
Faster frequency  $\omega_2 = 1.5 \omega_1$   
Moderate Intensity ( $I_1 = 1-3 \cdot 10^{14} \text{ W/cm}^2$ )  
Steers electron.



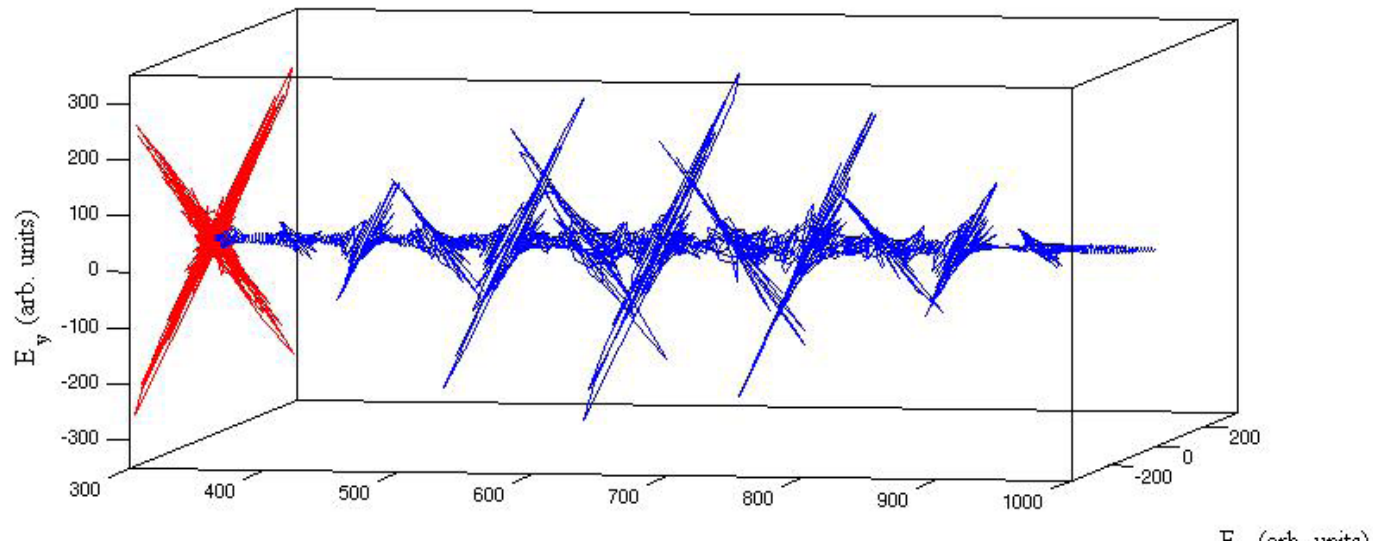
Fundamental laser field freq. ( $\omega_1$ ).  
High Intensity ( $I_1 = 2-6 \cdot 10^{14} \text{ W/cm}^2$ )  
Ionize & accelerate the electrons

Classical, SFA and TDSE calculations have been used to study the effect upon the amplitude, phase and polarisation of generated HHG of the laser field amplitudes, relative frequencies and phases

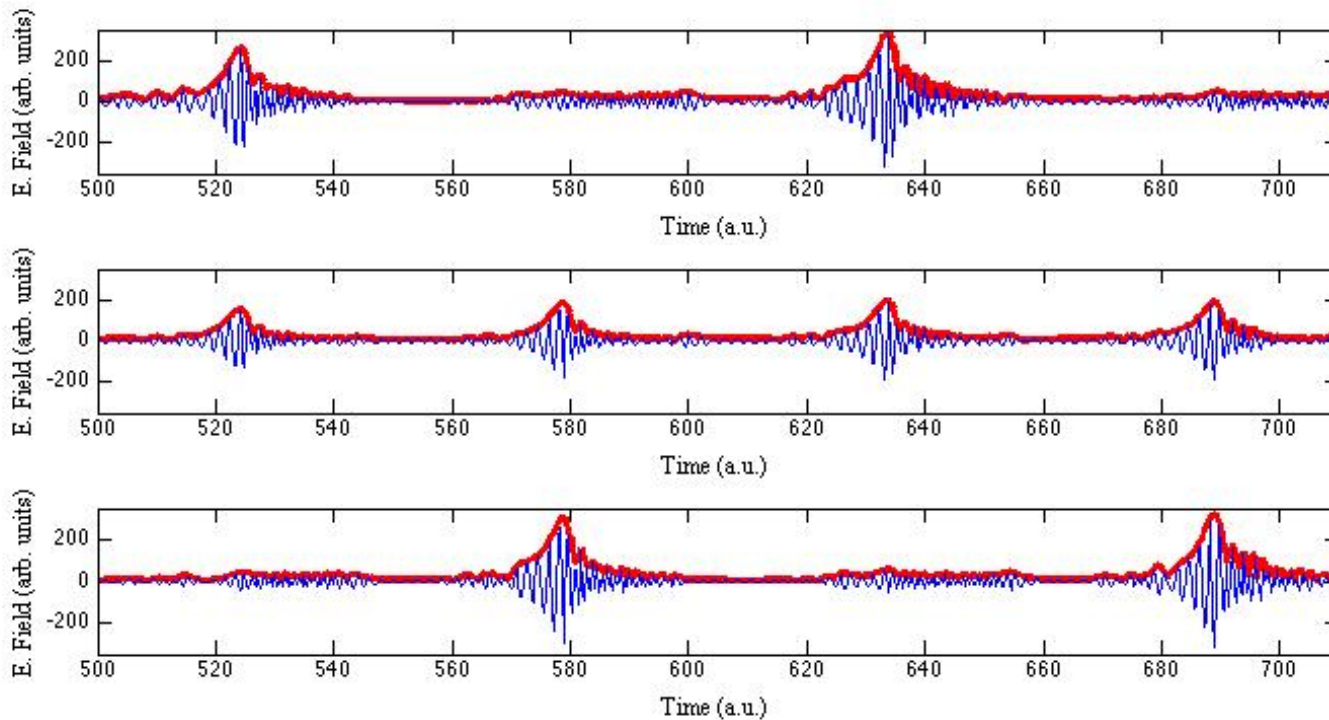
# Generation of an attosecond pulse train of alternating polarization



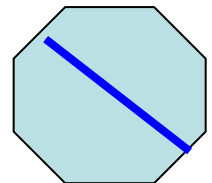
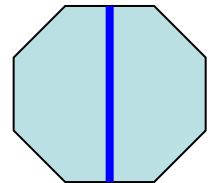
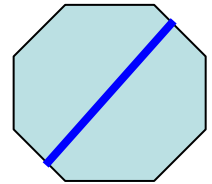
Driving fields at 800 nm and 533 nm,  $5 \times 10^{14}$  and  $2 \times 10^{14}$  W/cm<sup>2</sup>, and CEP  $\pi/2$  for both. Spectral filtering (grey) results in an attosecond pulse train with alternating polarization



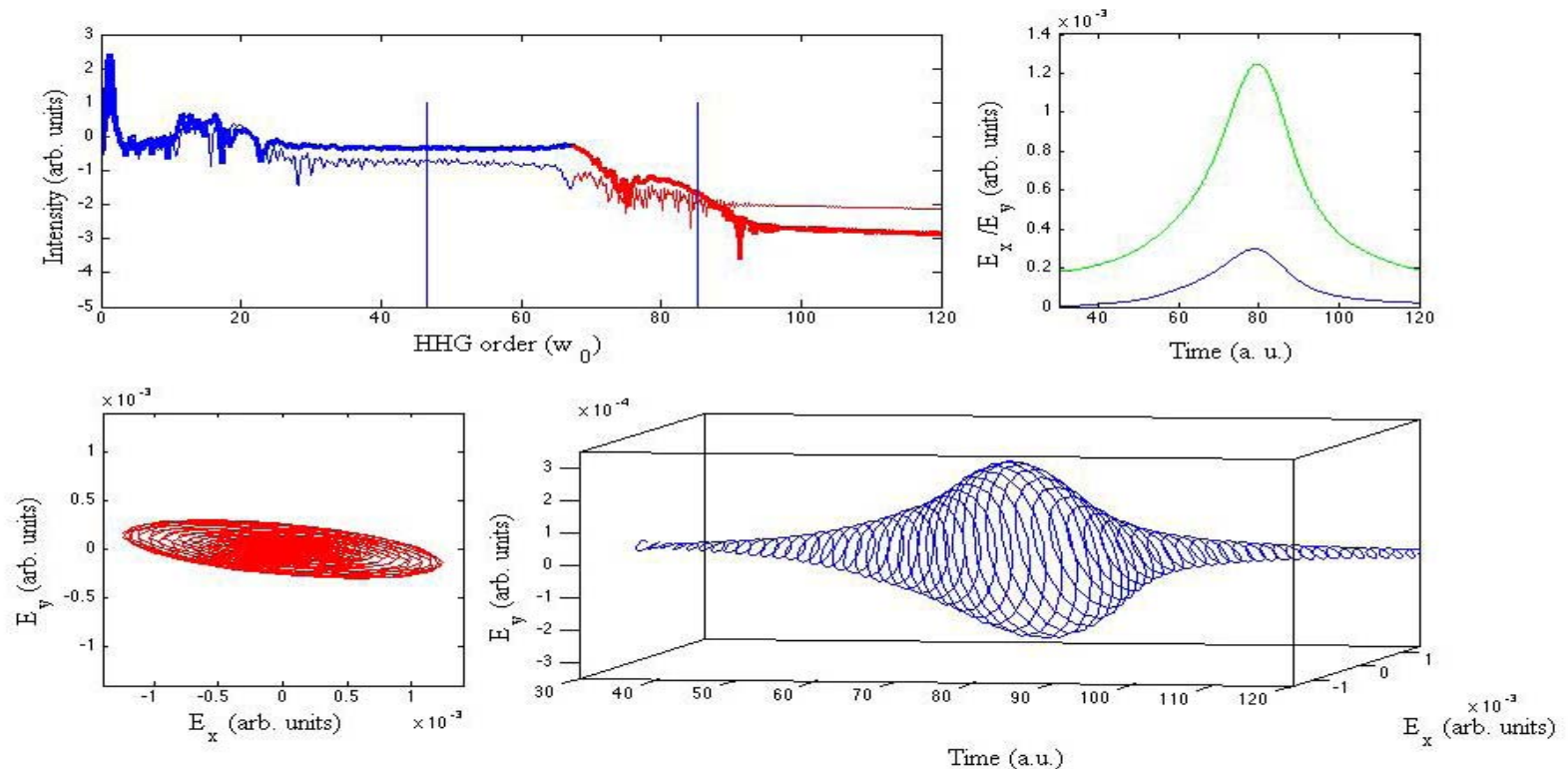
# Pulse train repetition can be changed from $T_1$ to $T_1/2$ through choice of polarisation



Polarizer UV



# Spectral filtering in cut-off region and correct choice of intensity and CEP leads to elliptically polarized harmonic radiation



Single attosecond pulse with elliptical polarization., Selecting the cut off region of the spectrum, we can obtain a single pulse with elliptical polarized light. TDSE on helium 800nm  $6 \times 10^{14} \text{Wcm}^{-2}$  and 533nm  $6 \times 10^{14} \text{Wcm}^{-2}$ , CEP=0 for both fields.

# Control of HHG emission can be achieved through choice of multi-colour laser field parameters

- CEP can control HCO in few-cycle fields (leading to a robust measurement of laser field waveform)
- Optimum electric field waveforms for HHG are identified that can be realised in the lab using existing technology
- Two-colour fields of perpendicular the polarisation can be used to control polarisation properties of an APT
- Many other control possibilities remain to be explored. For instance the direction and energy-time encoding of the returning electron can be altered leading to improved molecular structural imaging