Quantum Optics and Laser Science Group
Blackett Laboratory, Imperial College London

HHG Basic Physics: High Harmonic Generation & Matter in Strong Fields

1. Free Electrons in Strong Fields
2. Laser Ionisation
3. High harmonic generation
4. High harmonics from molecules

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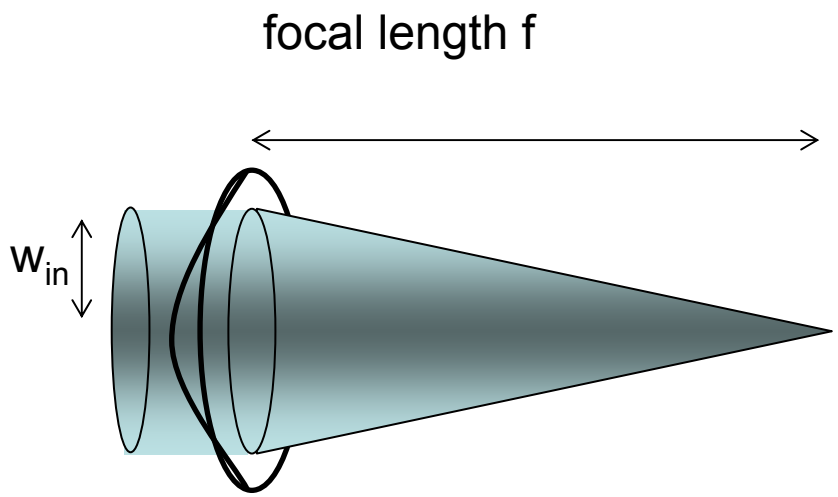
High intensity can be obtained at the focus of a lens mirror



Spot waist size (radius) for a Gaussian beam:

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

CW 10W 530nm laser focused by 20 cm lens (1cm beam diameter)
Intensity ~ 7×10^6 Wcm^{-2}

Pulsed (10ns) 1J mode-locked SHG Nd:YAG laser focused by 20 cm lens
Intensity ~ 7×10^{13} Wcm^{-2}

Pulsed (20 fs) 10mJ mode-locked/CPA amplified Ti:S laser focused by 20cm lens
Intensity ~ $5 \times 10^{17} Wcm^{-2}$

Amplified mode-locked lasers can very easily generate intensities > $10^{14} Wcm^{-2}$ when moderately focused

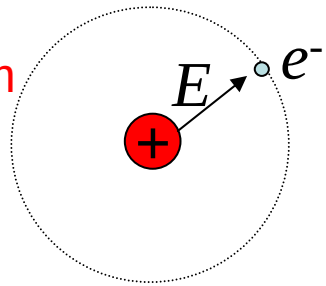
Electric Fields

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)

Hydrogen atom



$$E = \frac{e}{4\pi\varepsilon_0 a_0^2} = 5.1 \times 10^{11} Vm^{-1}$$

$$a_0 = \text{Bohr Radius} = 5.3 \times 10^{-11} m$$

For the laser field to reach the magnitude of the atomic field need $I \approx 3.5 \times 10^{16}$ Wcm-2

Basically same effects are seen in atoms or molecules when exposed to intense fields

Motion of a free electron in a laser field

- Free electron motion:

Field: $E(t)=E_0 \cos \omega t$

Charge: -e

$$\ddot{x} = \frac{e}{m} E \cos \omega t$$

$$\dot{x} = \frac{e}{m\,\omega} E \sin \omega t$$

$$x = -\frac{e}{m\,\omega^2} E \cos \omega t$$

- Amplitude of oscillation $x_0 = eE/m\omega$

e.g. for a 1μm wavelength field at 10^{16}Wcm^{-2}
amplitude ~10^{-8}m (~100 atomic radii)

- Cycle average kinetic energy (Ponderomotive energy)

$$U_P = \frac{e^2 E^2}{4\,m\,\omega^2}$$

e.g 1μm/10^{16}Wcm^{-2} U_P = 1keV

HIGH INTENSITY LASER-ELECTRON INTERACTIONS

$$U_P = \frac{e^2 E^2}{4 m \omega^2}$$

Cycle averaged kinetic energy of electron oscillating in field, Ponderomotive energy.

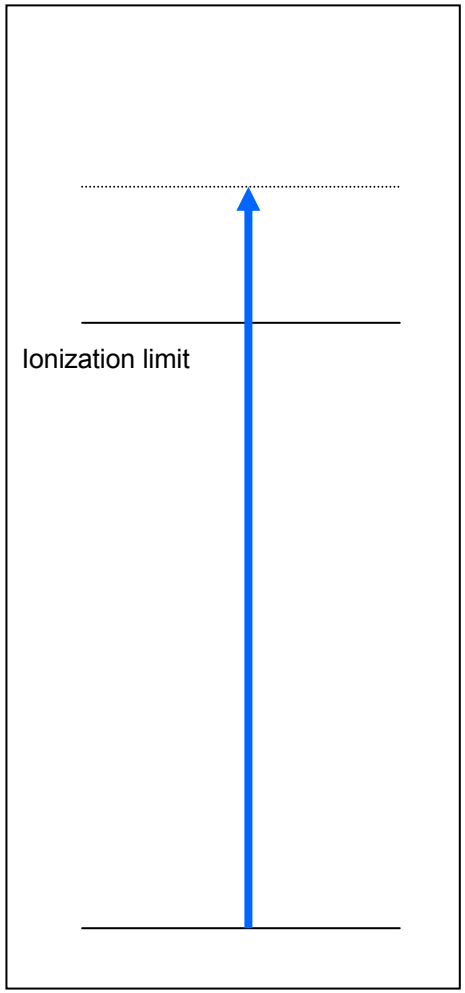
Proportional to $I \lambda^2$

at $1\mu m/10^{14} Wcm^{-2}$ this has a value of 10eV.
at $10^{16} Wcm^{-2}$ this is 1keV

Continuum state is a laser dressed state (Volkov state).
Property is that the continuum wave-packet position expectation <r> is oscillatory, so possibility for continuum part to return and re-interact with ion at a slightly later time (determined by the optical cycle) and this has big role in control of light emission and other strong field processes.

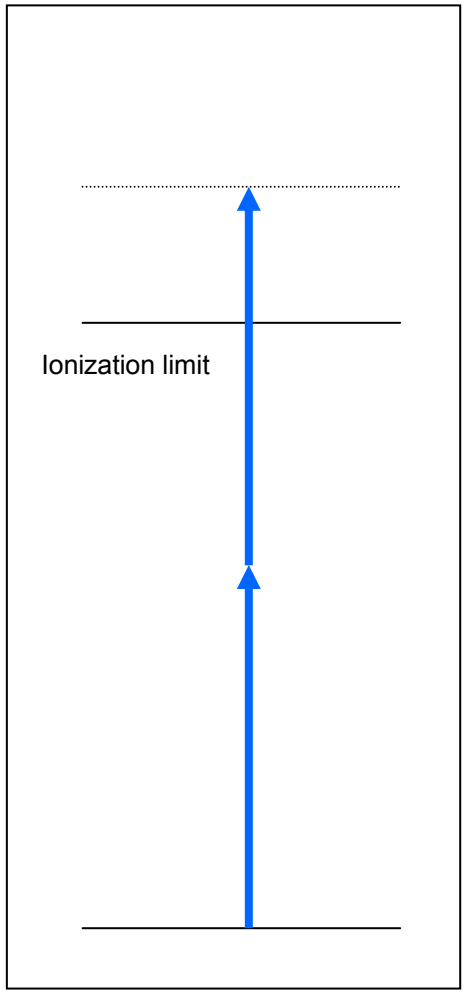
Single and multi-photon ionization

Single deep UV photon



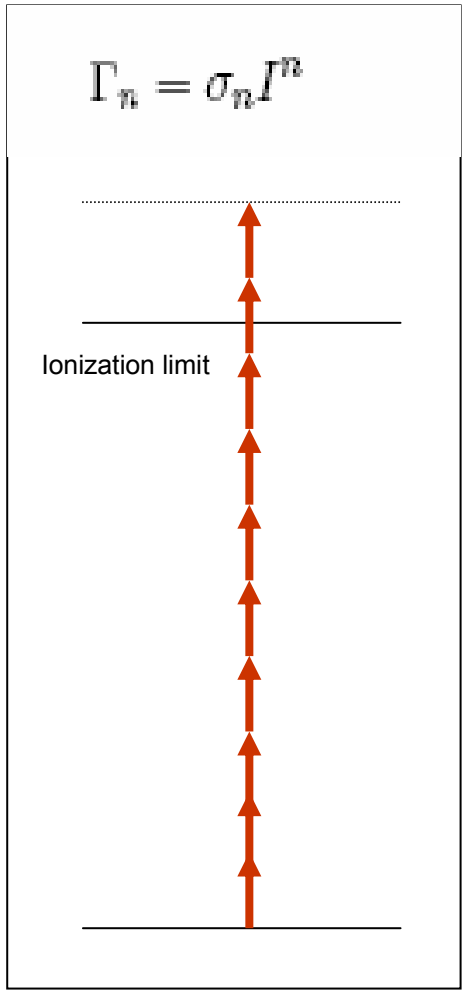
Possible at any intensity- Photoionization e.g. by Sun, UV lamp, Synchrotron

Two-photon ionization by UV photons



Requires laser (otherwise weak For intensity < 10^{9}Wcm^{-2})

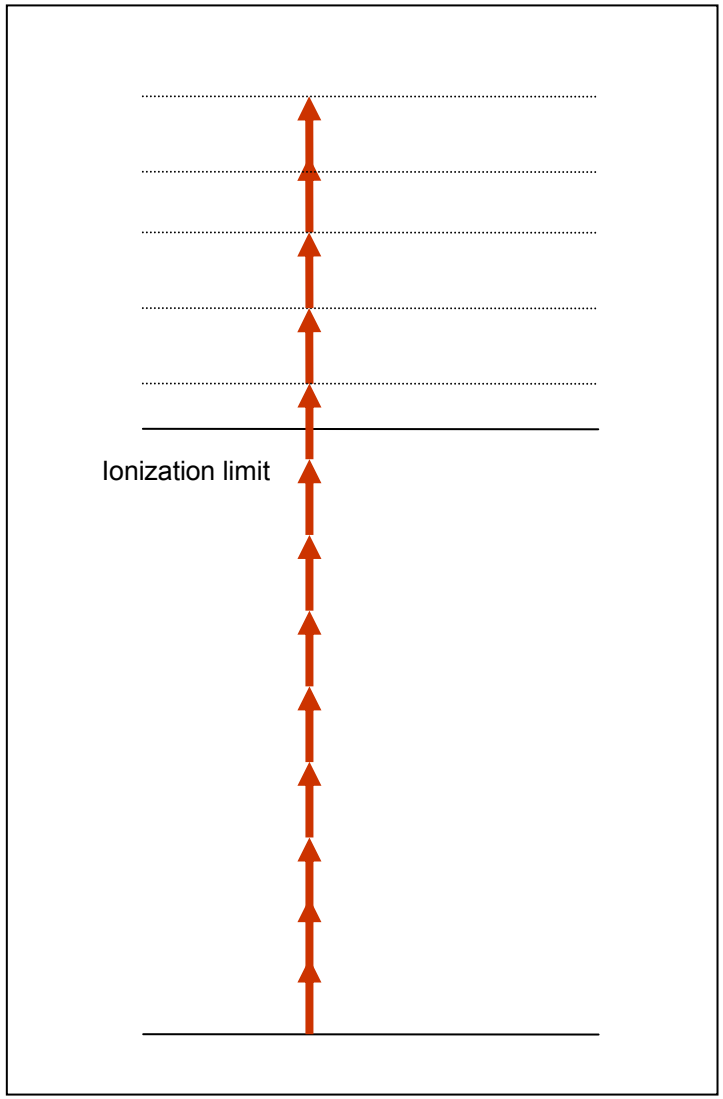
Multi-photon ionization By visible IR/photons



Requires high power pulsed Laser >10^{12}Wcm^{-2}

Above Threshold Ionization (ATI) can occur if laser field sufficiently intense

Extra peaks in Photoelectron spectrum occur spaced by $h\nu$

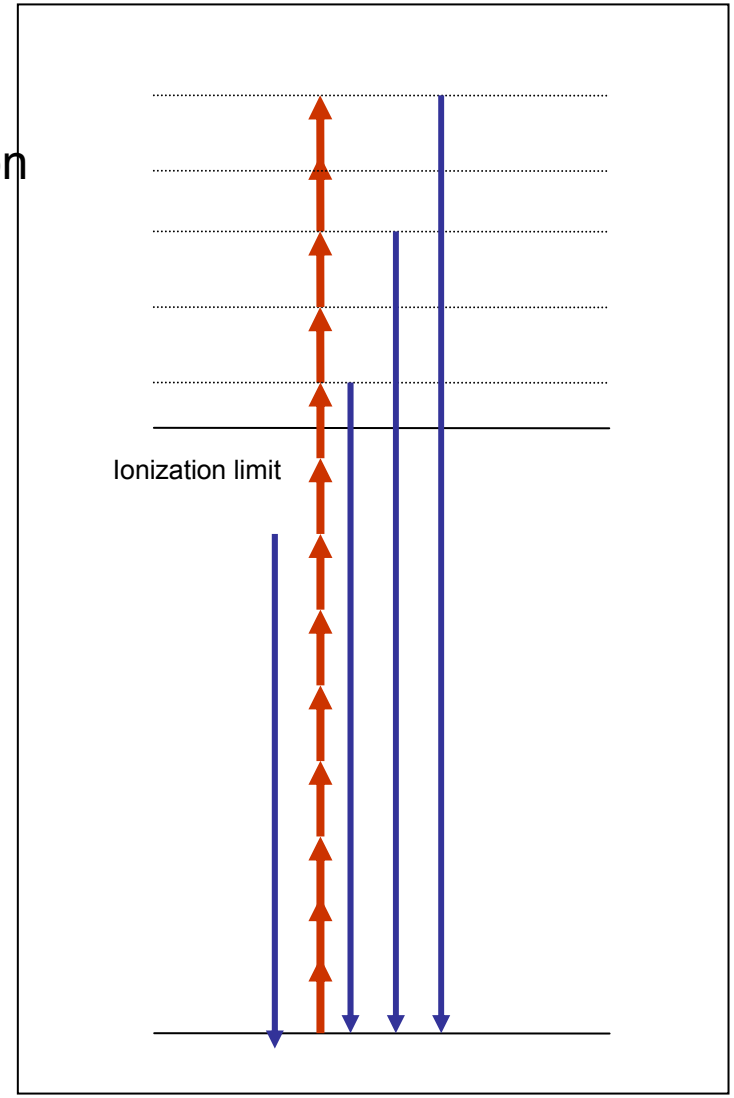


Electron energy peaks at:

$$E_{electron} = n\hbar\omega_0 - I$$

The same multi-photon picture can be used to explain high order harmonic emission

Emission of odd harmonics of field even above ionization energy.

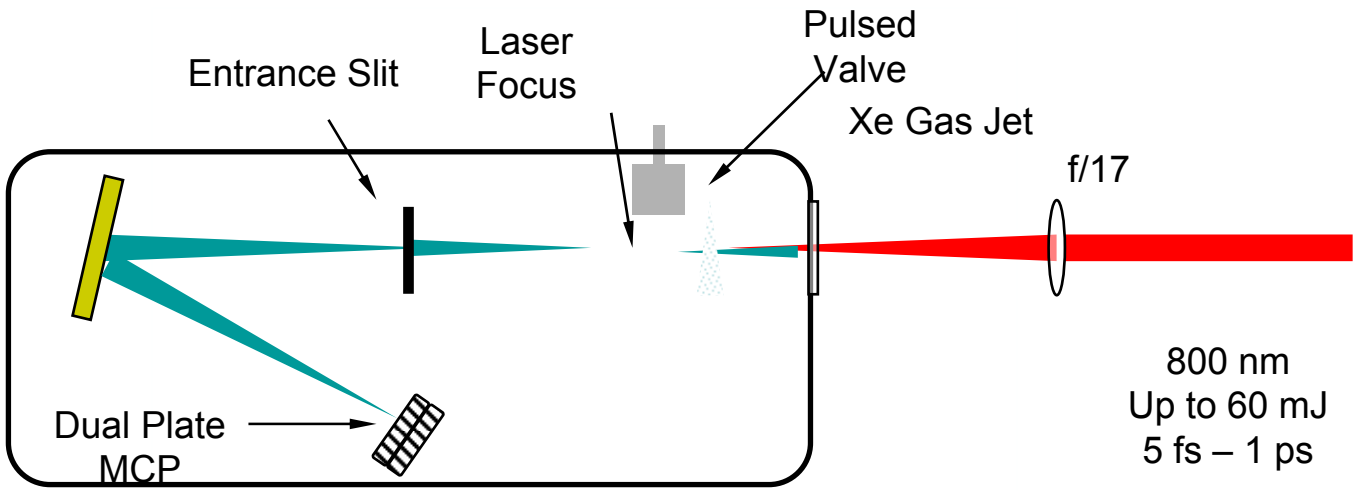


Extreme ultraviolet harmonics (XUV) result

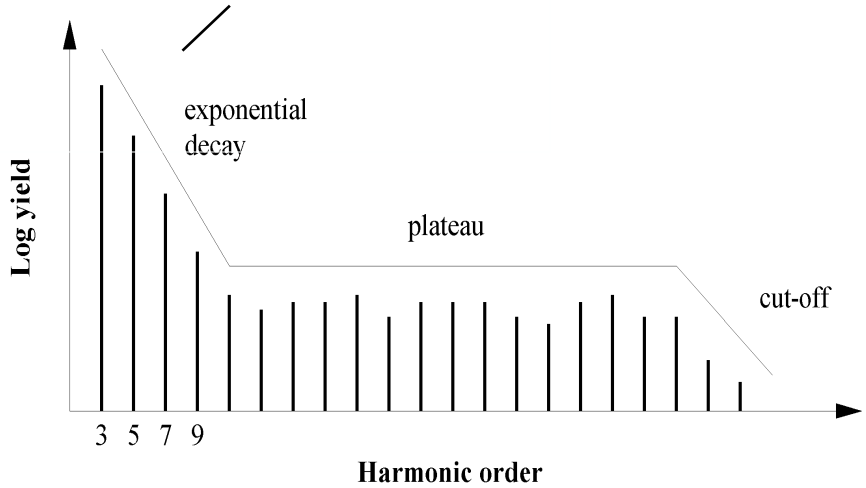
$$\hbar\omega_{xuv} = q\hbar\omega_0$$

q odd – for symmetry reasons (parity constraints for parametric process)

When an intense laser interacts with atoms or molecules very high order radiation harmonics are observed.



Laser intensities 10^{13}-10^{15}Wcm^{-2} required to strongly drive electrons.



This high harmonic generation results in coherent light pulses of high brightness into the X-ray region of the spectrum.

Strong Field Limit

For stronger fields the quantum nature of the field (photons) becomes less important and a classical field treatment can suffice.

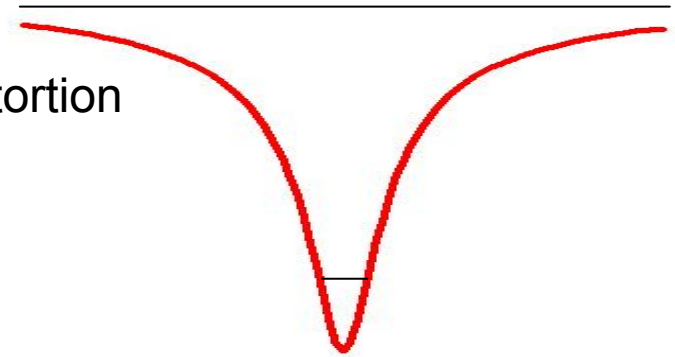
Combined Coulomb and Laser field (x polarised) potential:

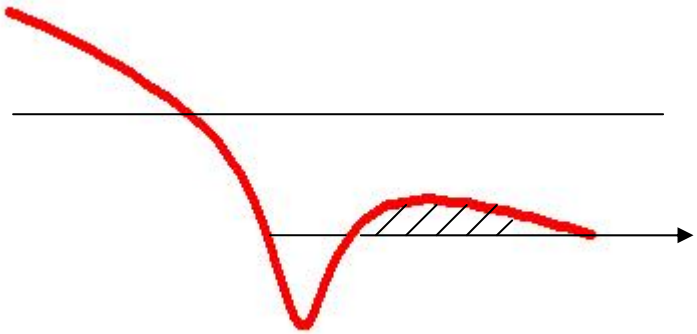
$$V = \frac{-Ze^2}{4\pi\varepsilon_0}\frac{1}{r} + (-e)E(t)x$$

Potential is most distorted near the times of the laser field extrema.
We can understand the effect of the potential upon the binding of a bound electron by considering a cut along the x-axis (as this is the direction where distortion is largest).

$$V = \frac{-Ze^2}{4\pi\varepsilon_0}\frac{1}{x} + (-e)E_0 x$$

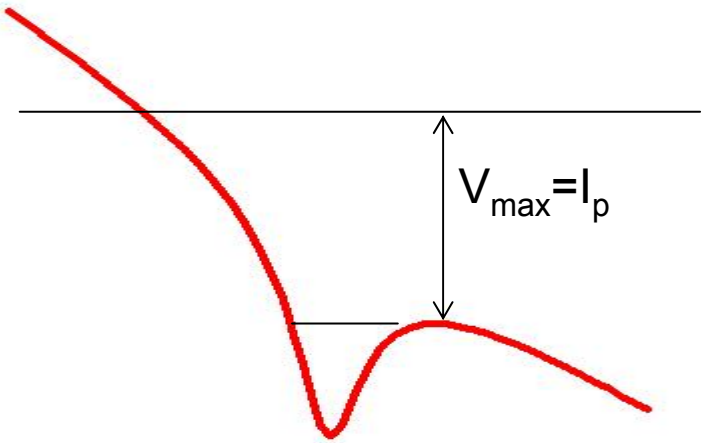
No distortion





Electron can quantum tunnel through barrier.

Ionisation prob. exponentially sensitive to area under barrier



Strongly distorted potential –
bound electron can escape directly over the reduced barrier
Over The Barrier Ionization (OTB)

Distorted Potential

$$V(x) = V_{atom} + V_{laser} = \frac{-Ze^2}{4\pi\epsilon_0 x} - eE_0 \cos\omega t \; x$$

At peak of laser field:

$$V(x) = \frac{-Ze^2}{4\pi\epsilon_0 x} - eE_0 x$$

Max (local max) of potential given by the condition:

$$\frac{\partial V(x_{max})}{\partial x} = \frac{Ze^2}{4\pi\epsilon_0 x^2} - eE_0 = 0$$

Which is located at:

$$x_{max} = \sqrt{\frac{Ze}{4\pi\epsilon_0 E_0}}$$

Typically this is located a few Angstroms from the centre of the potential.

Tunnel Ionization

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

Depends exponentially on the magnitude of barrier – but atomic prefactors due to the shape of the wave function in the region of the barrier determine absolute value. Quantitative theory for hydrogenic and non-hydrogenic atoms see:

M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].

In a non-hydrogenic atom the tunnel rate from ADK is:

$$w(t) = C_{n^*}^2 I_p \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!}\left(\frac{3E(t)}{\pi F_0}\right)^{1/2}\left(\frac{2F_0}{E(t)}\right)^{2n^*-|m|-1} \times e^{-2F_0/3E(t)} \quad (1$$

Where:

$$C_{n^*} = \left(\frac{2e}{n^*}\right)^{n^*}\frac{1}{\sqrt{2\pi n^*}}, \quad n^* = \frac{Z}{\sqrt{2I_P}},$$

Tunnel or Multi-Photon?

Determined by the per cycle tunnel ionization probability; if this is high then tunnel ionization dominates.

Keldysh parameter defined as the ratio of tunneling time through barrier to the cycle period :

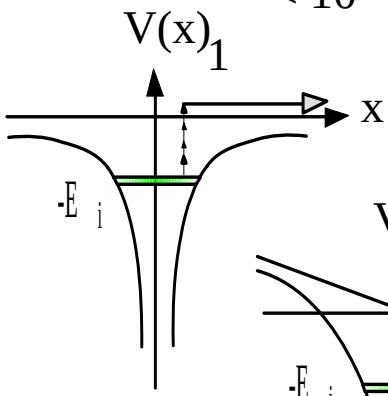
$$\gamma = \sqrt{\frac{I_P}{2U_P}}$$ or $$\frac{1}{\gamma} = \frac{eEa_0}{\hbar\omega}$$

γ < 1 equivalent to a large tunnel ionization rate whilst γ>>1 MPI dominates

HIGH INTENSITY LASER-MATTER INTERACTIONS

Multi-Photon

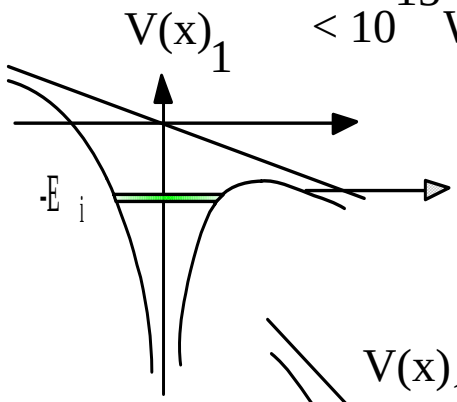
$< 10^{14}$ Wcm^{-2}



Some distortion of potential but multi-photon coupling to bound/continuum states dominates. Photoelectrons driven in laser field, continuum-continuum transitions probable.

Tunneling

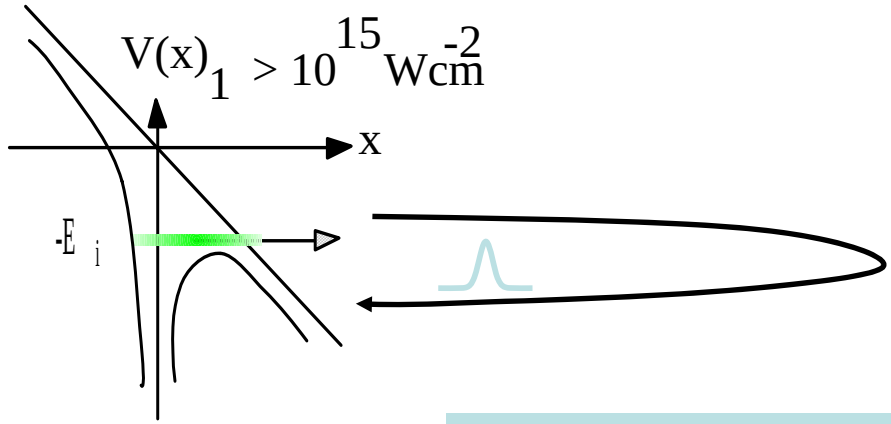
$< 10^{15}$ Wcm^{-2}



Strong distortion to potential, bound states strongly modified (excited states disappear due to over the barrier (OTB) ionisation). Photoelectrons strongly driven in laser field

Over the Barrier

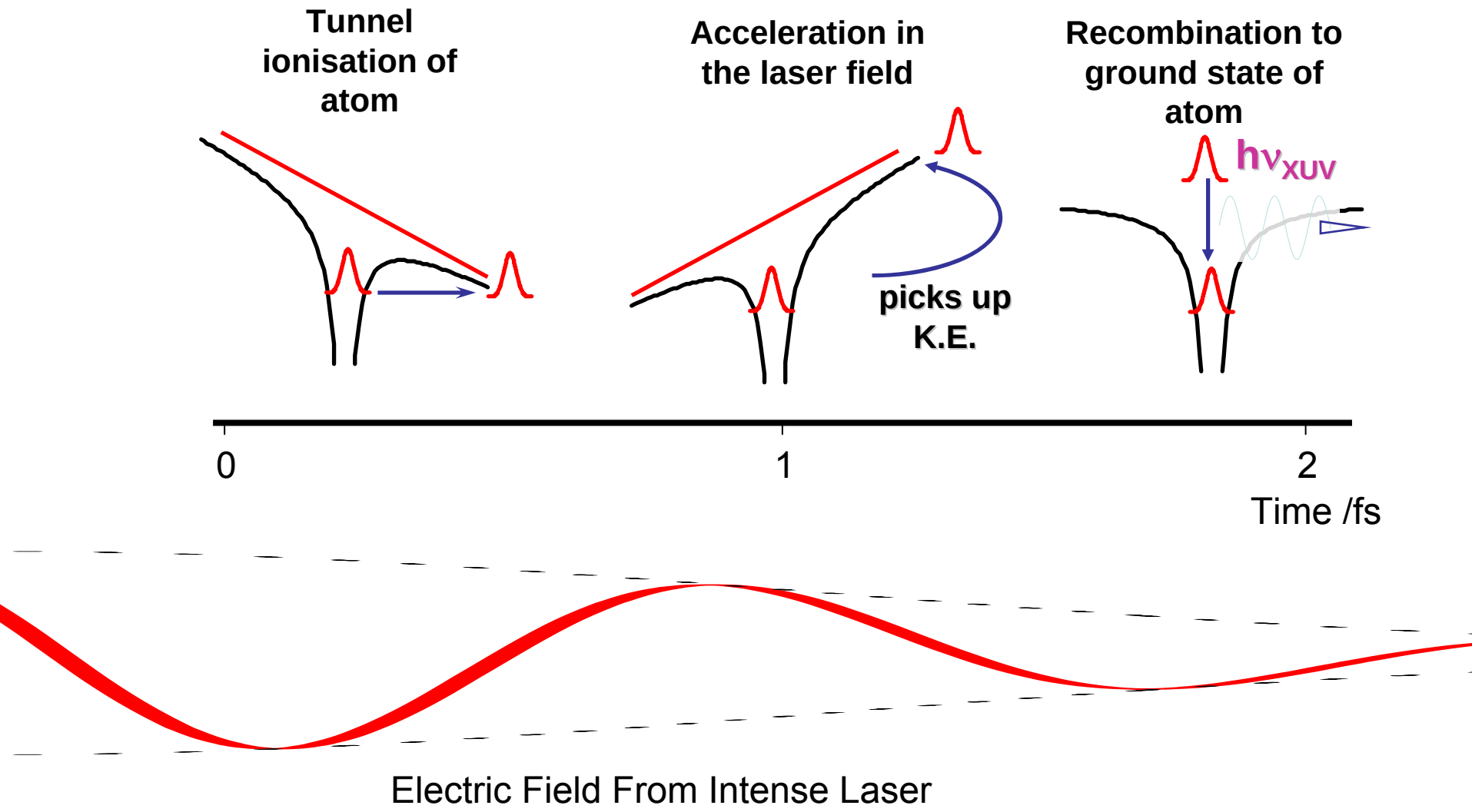
$V(x)_1 > 10^{15}$ Wcm^{-2}



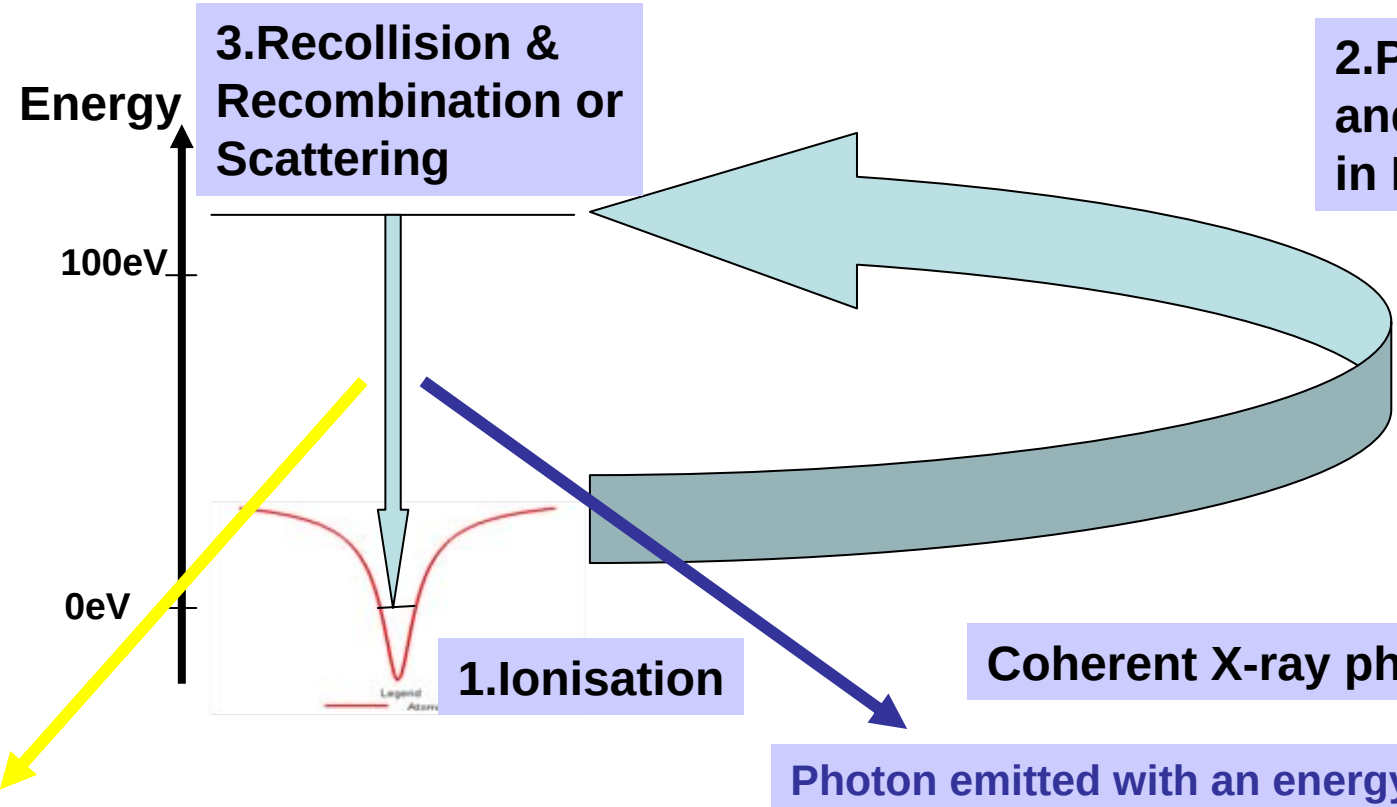
Leads to:
High harmonic generation (HHG).
Above threshold ionisation (ATI).

Freed electrons driven to and fro In the strong laser field

High harmonic generation occurs on a sub-optical cycle timescale



The strong field interaction with an atom or molecule can be viewed as a 3-step process.

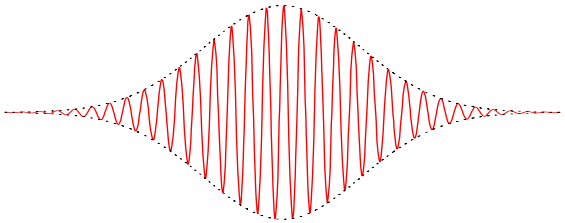


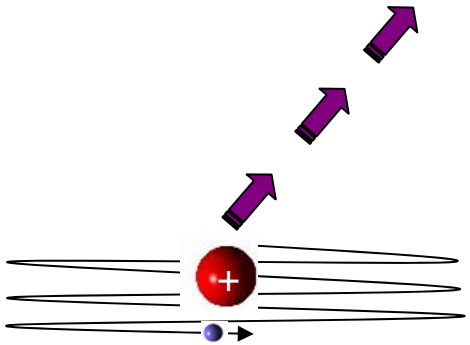
Corkum, PRL 71, 1995 (1993), Krause et al PRL, 68, 3535 (1992)

High harmonic generation with multi-cycle and few-cycle laser pulses.

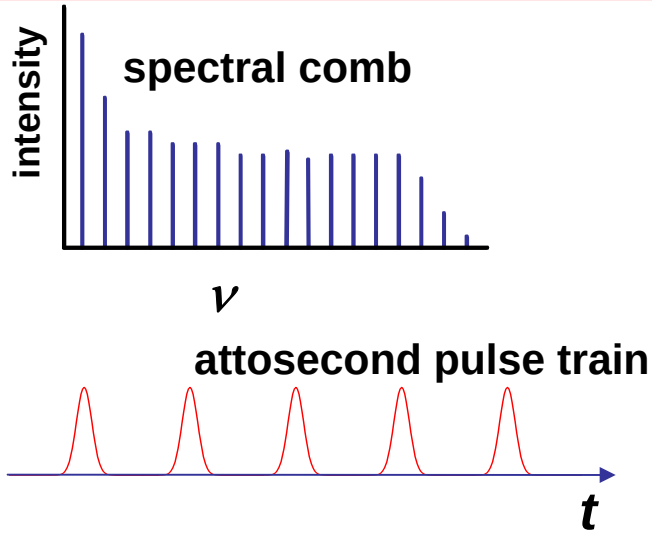
HHG with few-cycle laser pulses present a route to single attosecond pulses.

Long pulse

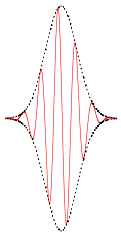




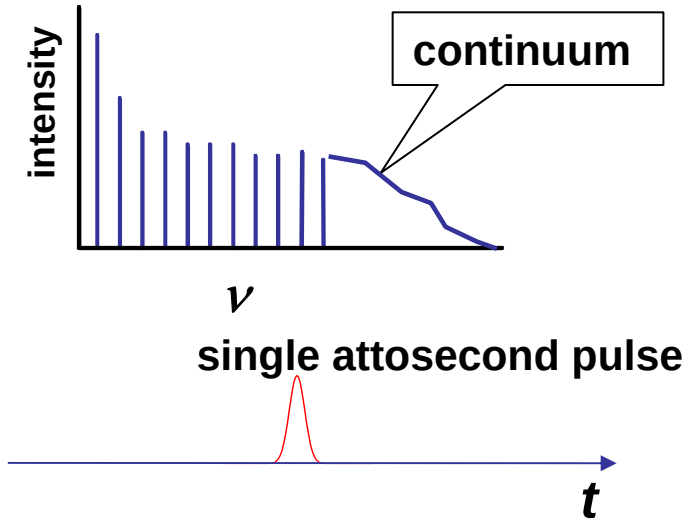
many recollisions



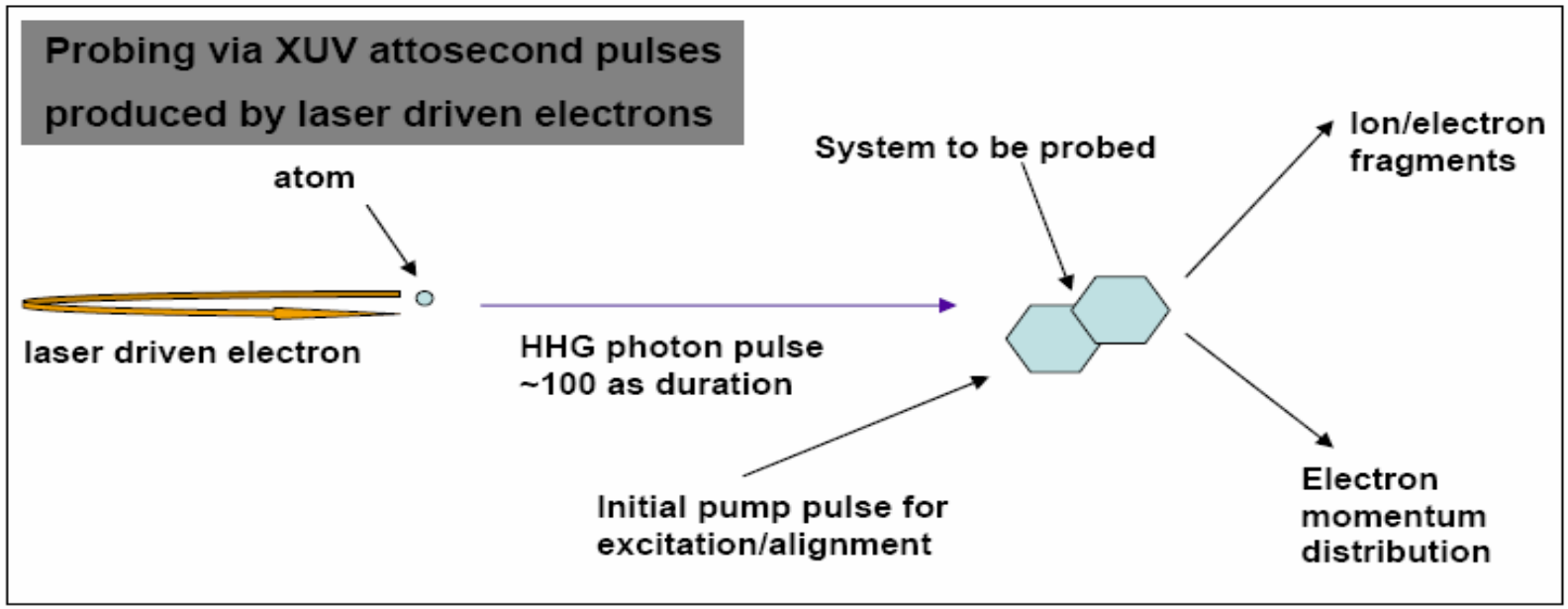
Few-cycle pulse

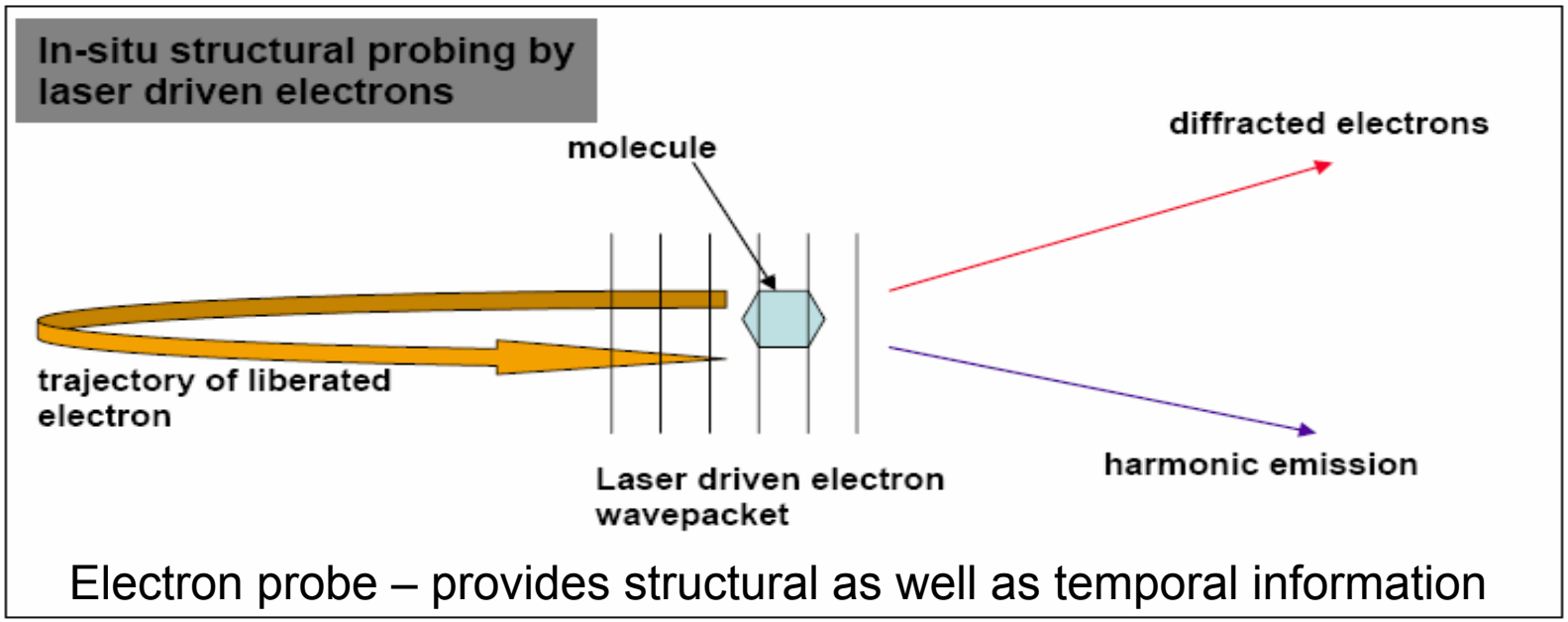


single recollision of maximum energy



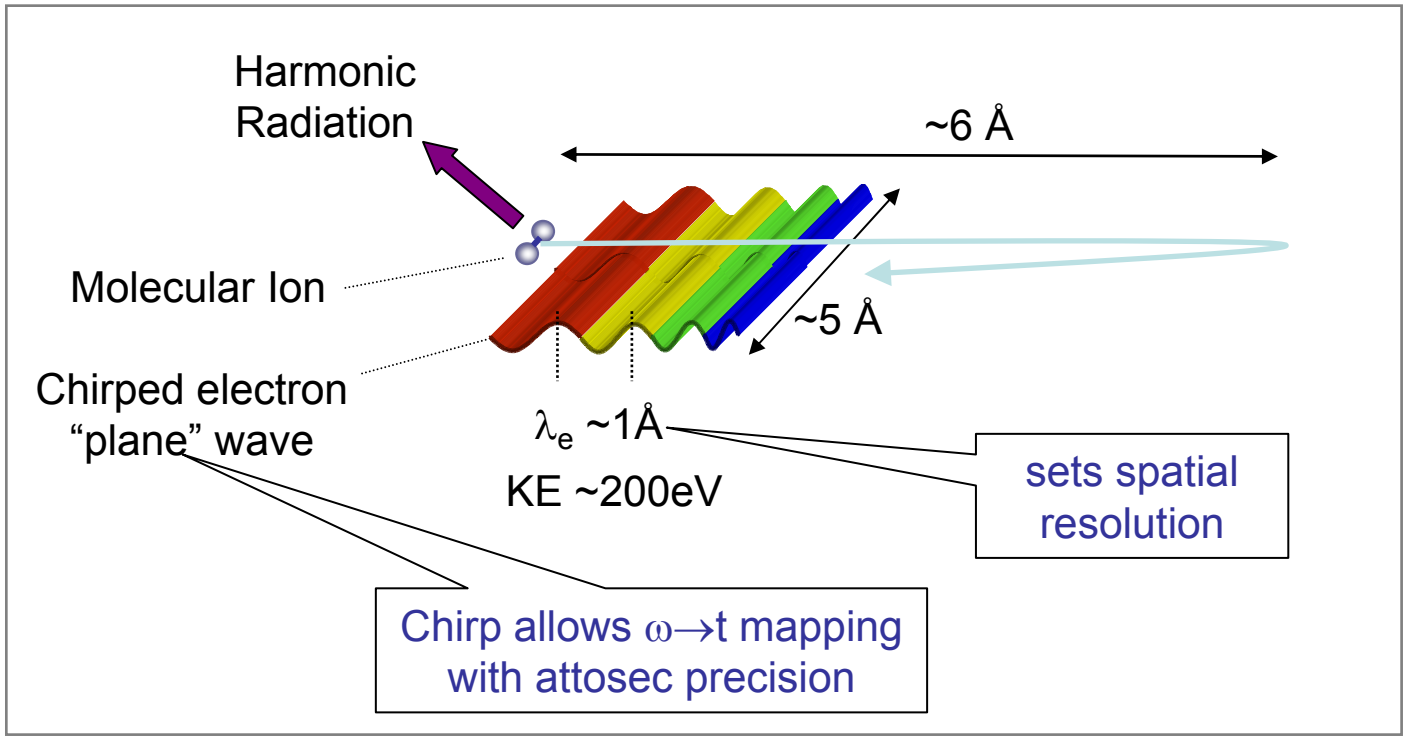
We are looking at two approaches to attosecond measurement





Electron probe – provides structural as well as temporal information

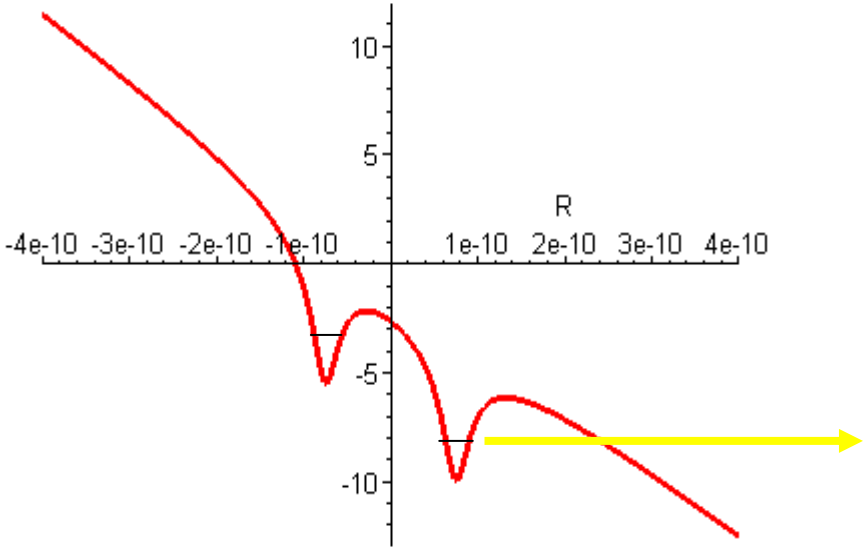
HHG is an ultra-fast structural probe



Laser Parameters
λ = 800nm
$I = 10^{15}$ Wcm^{-2}
Linearly Polarised

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

The strong oscillating optical field causes oscillatory distortion of the molecular potential that leads to tunnel ionisation as in atoms.

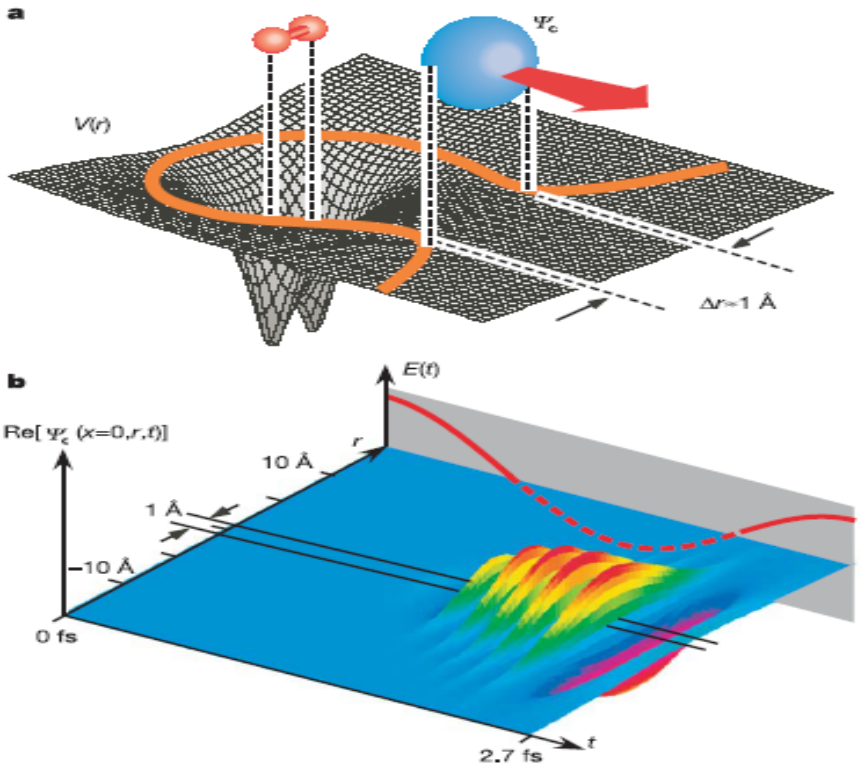


Electron can escape due to the quantum mechanical process of tunnelling through the barrier created by the distortion of the potential well.

Following escape (ionisation) the electron moves predominately under the influence of the strong laser field - which accelerates the electron to a high energy (of order $U_{p,}$ i.e. 65eV for an 800nm laser at $10^{15}Wcm^{-2}$).

The when the field reverses the electron may be driven back to recollide with the molecule.

According to the rules of quantum mechanics an electron wavepacket is formed after tunnel ionisation.



The electron wavepacket will be approximately Gaussian in form with an initial spatial width ~10^{-10}m (about the size of the molecule).

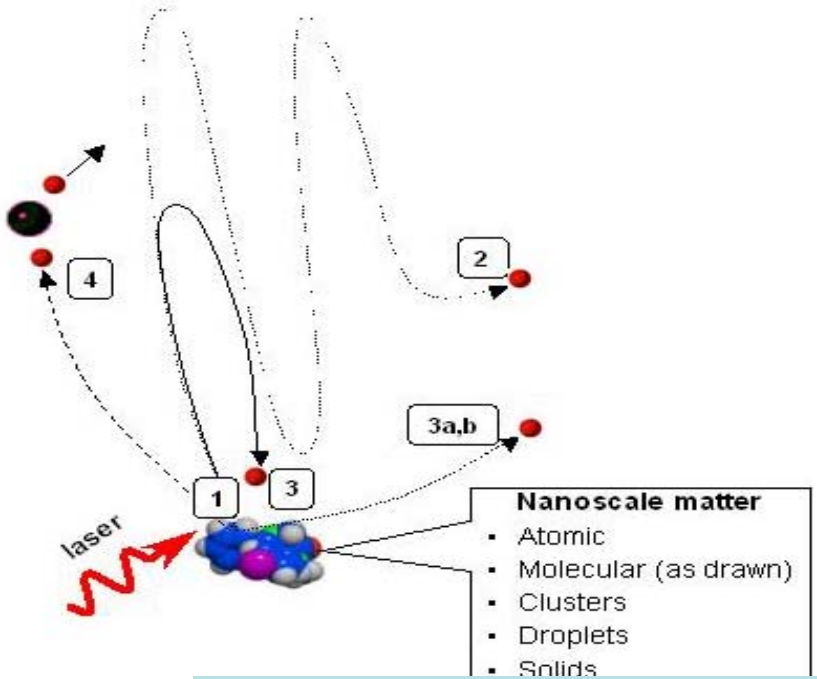
It then spreads in all directions as time proceeds. The rate of spreading is determined by the particle mass and initial size of the wavepacket

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

Despite the wave-packet spreading the recollision event still lasts less than the optical cycle time (<1fs), and although reduced the probability remains finite.

The electron momentum distribution will be significantly altered by the presence of the intense laser field.

MOLECULAR ELECTRON DYNAMICS IN THE STRONG FIELD LIMIT



1. Tunnel ionisation step.
2. This can lead directly to Ionisation without the electron ever returning to the molecule.
Or/
3. Electron may recollide with molecule leading to either;
(a) recombination (HHG)
(b) Elastic/inelastic scattering.

Parameters of re-colliding electron:

● Energy at recollision say around $2U_P$ (for 800nm at (1) 10^{15}Wcm^{-2}, T=119eV at (2) 10^{16}Wcm^{-2} ,T= 1.19keV) (max return energy $3U_p$)

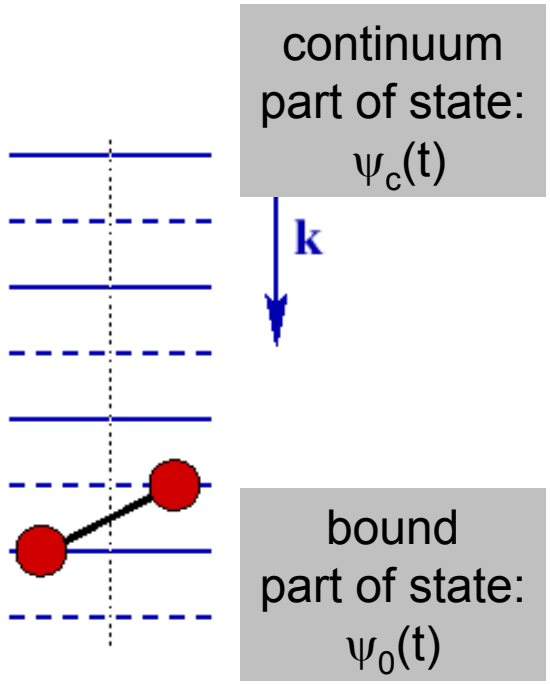
● Wiggle amplitude significantly larger than molecule (e.g. (1) 2.7nm, (2) 8.7nm)

● $\lambda_{\text{de Broglie}}$ comparable to inter-nuclear spacing (e.g. (1) 1.12 x 10^{-10} m, (2) 3.5 x 10^{-11}m)

The emission from the atom/molecule is determined by the dipole driven at the harmonic frequency

$\psi(t) = \psi_0(t) + \psi_c(t)$

Time dependent wavefunction is a superpositon of continuum and bound parts. Contains components oscillating at high frequencies – leading to emission at these frequencies



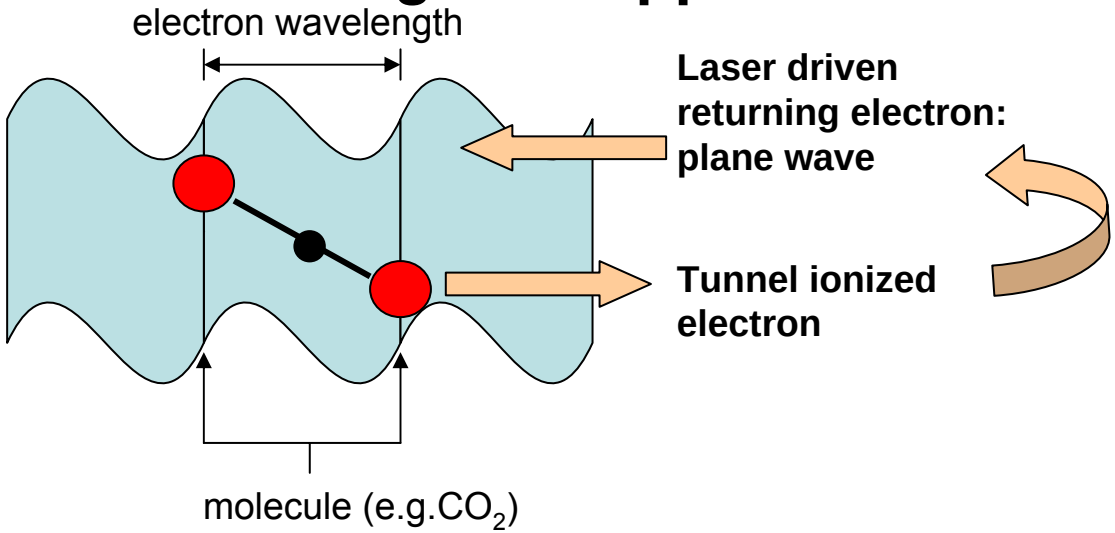
Single atom/molecule response can be treated either by numerical integration of the Schrödinger equation or analytically (in the SFA) (Lewenstein et al PRA, 49, 2117 (1994)).

Matrix element for harmonic generation at frequency ω:

$$A_{\vec{e}}(\omega) = \int \langle \psi_0(t) | \hat{e} \cdot \nabla V | \psi_c(t) \rangle \, e^{i\omega t} \, dt$$

[A full modelling of HHG requires also the propagation (phase-matching) to be included. This can be done but requires full account of all sources of phase-mismatch (dipole phase, material dispersion, free electron dispersion, geometrical phase slippage) and account of transverse (beam-profile) effects. Careful choice of experimental parameters minimizes these complications.]

The HHG process can be transparently formulated in the strong field approximation



Amplitude of HHG process determined by (c.f Lewenstein et al 1994) :

$$D_x(t) = 2i \int_0^t dt' E(t') C(t-t') \int \frac{d^3p}{(2\pi)^3} \bar{d}_x^*[\mathbf{p} + \mathbf{A}(t)]$$
$$\times \bar{d}_x(\mathbf{p} + \mathbf{A}(t')) e^{-i \int_{t'}^t dt'' \{[\mathbf{p}+\mathbf{A}(t'')]^2/2 - E_0\}} + \text{c.c.},$$

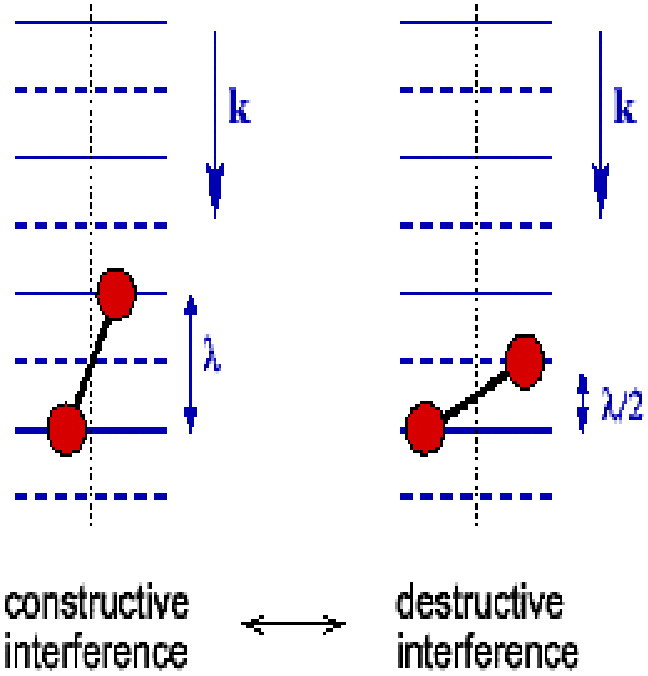
Tunnel ionization from the molecular bound-state;

Propagation in the laser field;

Recombination back into molecular bound-state.

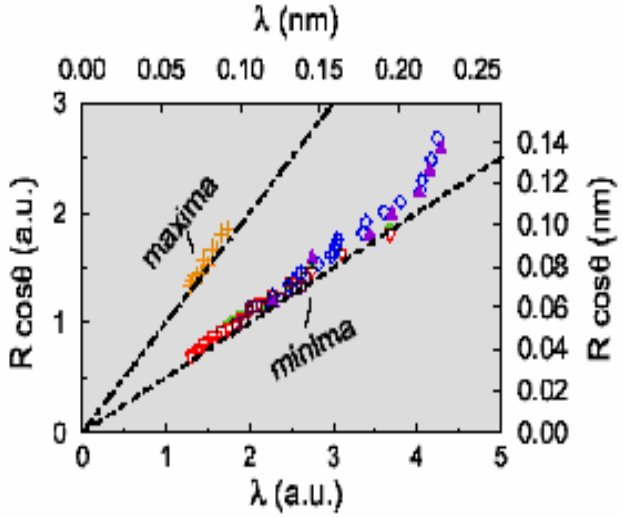
The last factor plays a critical role in new methods to use HHG to image molecular structure in aligned molecular samples e.g. Tomographic reconstruction (Itatani et al Nature (2004)) or recombination step interference signatures studied by our group (Lein et al PRL (2002), Vozzi et al PRL (2005)) and Kanai et al Nature (2005). See review: M.Lein, *J.Phys.B:At.Mol.Opt.Phys.* 40, R135-R173 (2007)

In diatomic molecules the dipole amplitude is determined by relative phase of the recolliding electron wave at the two centres.



Projected internuclear separation versus electron wavelength

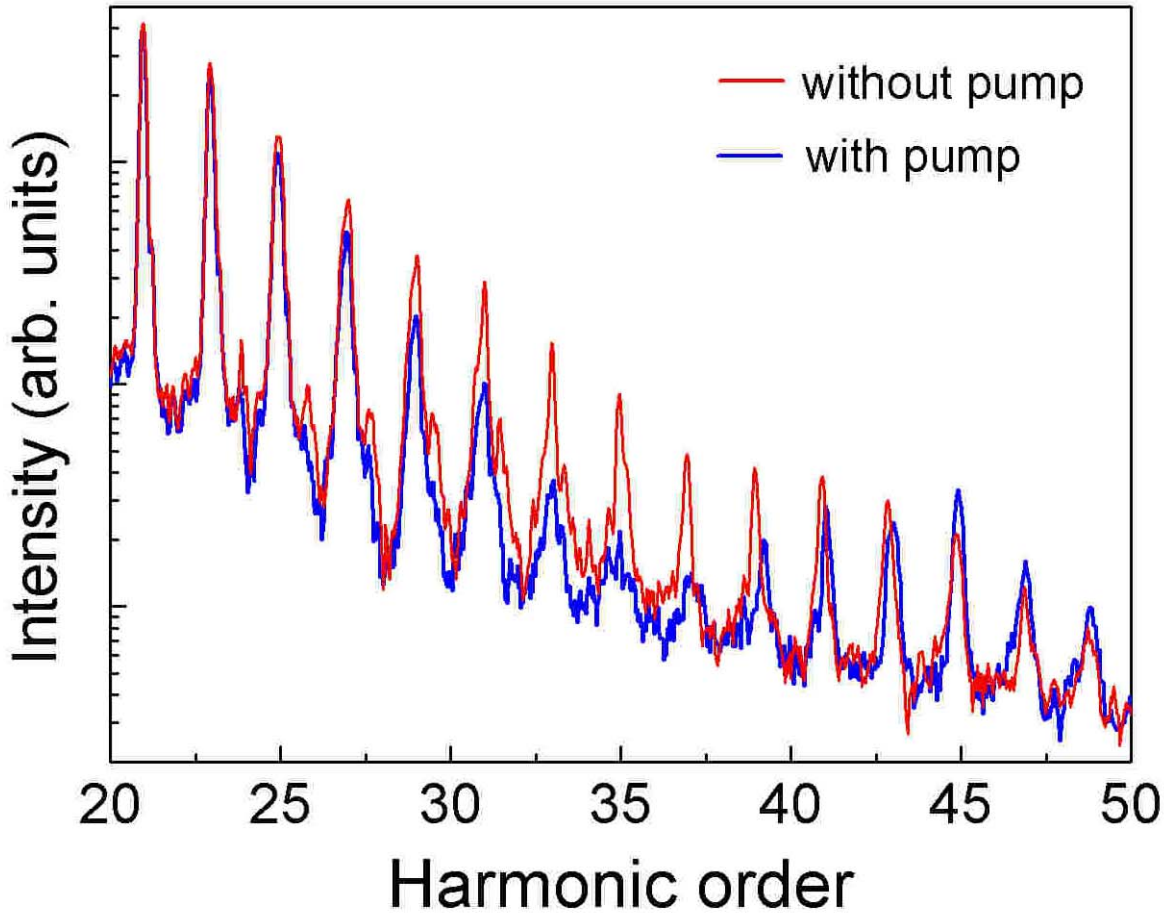
$$\lambda = 2\pi/k, \quad k^2/(2m) = \omega$$



Lein M, Hay N, Velotta R, Marangos J P and Knight P L 2002, *Phys. Rev. A* 66 023805.

This process has recently been observed in HHG from CO_2 in the group of Hirofumi Sakai (Tokyo) *Nature* **435,** 470 (2005) **(perhaps…)**

At the peak of the rotational revival a robust minimum is observed in the HHG due (perhaps) to the destructive interference between 2-centres



Vozzi et al. Physical Review Letters **95** 153902 (2005)

Recently phase shifts also measured c.f. Boutu et al Nature Physics 4, 545 (2008). In fact recent theory (Smirnova et al) finds hole dynamics within core may play dominant role – dynamical interference between ion core channels…

Tomographic reconstruction of N_2 HOMO

$$d(\omega) = \int \langle \psi_0 | z | a(k) e^{ikz} \rangle e^{i\omega t} dt$$

Amplitude has form of a Fourier transform of ψ_0. Amplitude measured as |k| scanned across harmonic spectrum, k direction scanned by molecular rotation: Sufficient information for orbital reconstruction.

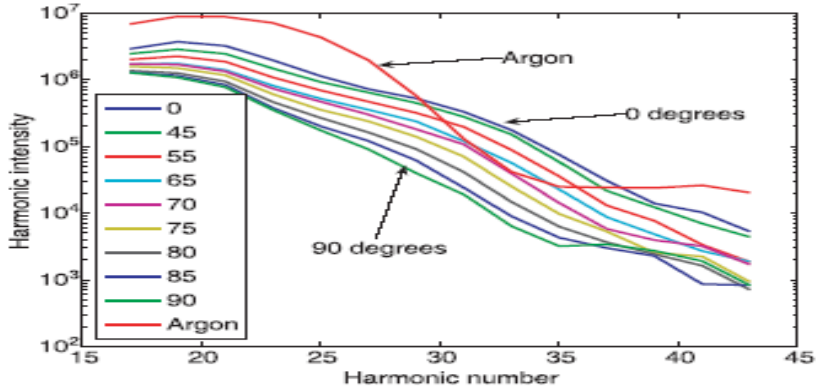
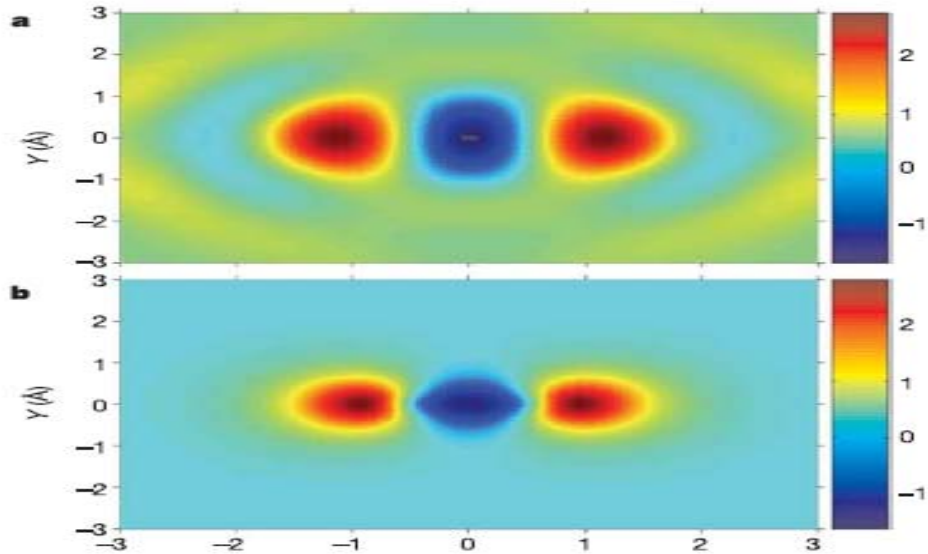


Figure 3 High harmonic spectra were recorded for N_2 molecules aligned at 19 different angles between 0 and 90° relative to the polarization axis of the laser. For clarity, only some of the angles have been plotted above. The high harmonic spectrum from argon is also shown; argon is used as the reference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

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



Tomographic reconstruction from HHG

Accurate calculation of wavefunction

Tomographic retrieval currently relies on accuracy of SFA

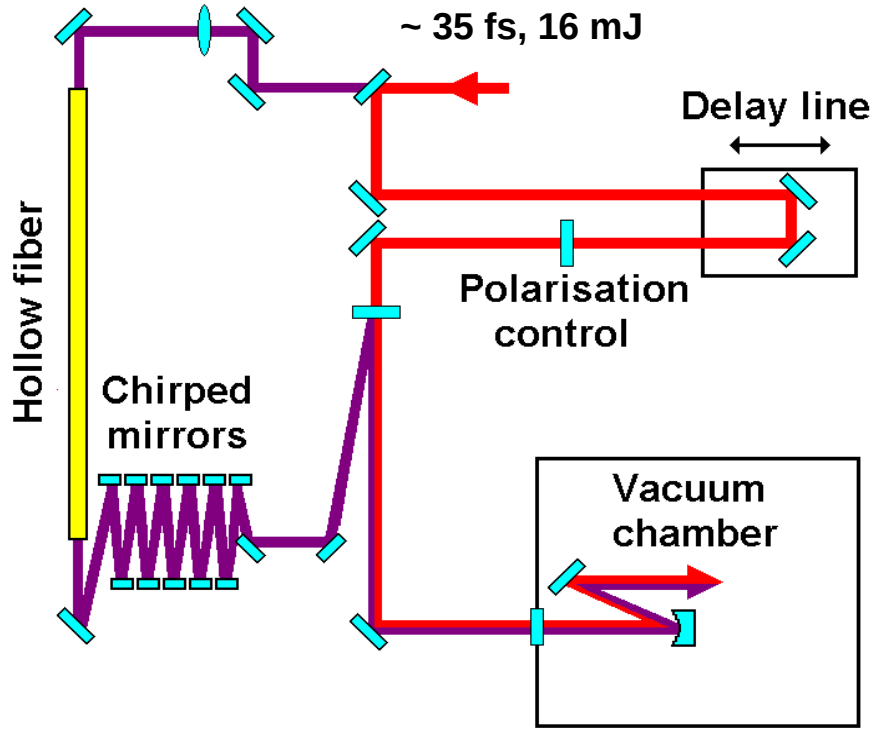
Essence of SFA Treatment

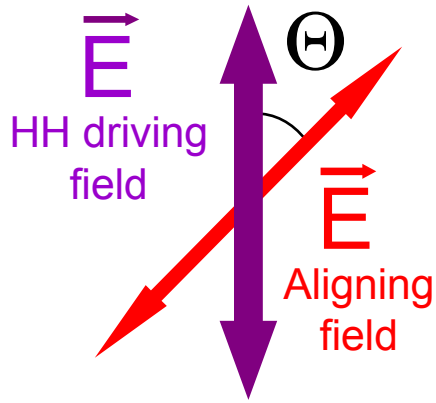
$$d(\omega) = \int \langle \psi_0 \,|\, z \,|\, a(k) e^{ikz} \rangle e^{i\omega t} dt$$

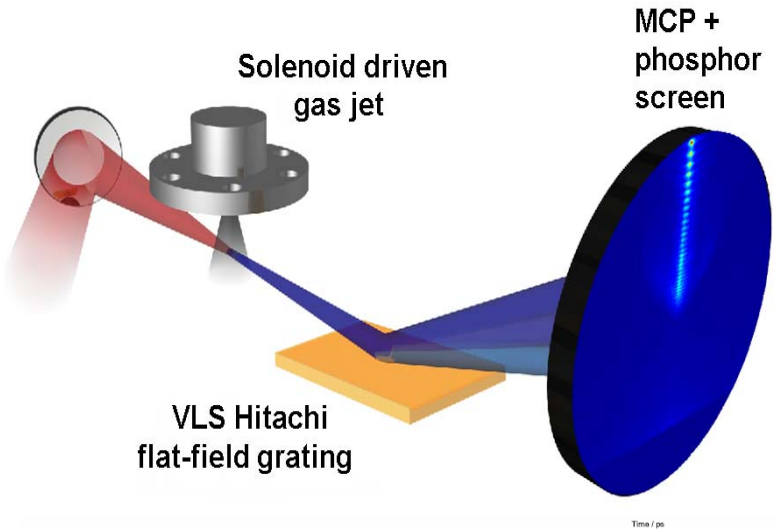
- Essential quantity is dipole matrix element.
- Assumes single-active electron and ignores multi-electron properties of molecular wavefunction (unless explicitly modified).
- Assumes continuum states are approximated as plane waves.
- Ignores influence of laser field (very strong) upon molecular bound state (assumes only a single bound state and no bound-bound or free-free transitions).

ALL THE SAME IT HAS THE HUGE ADVANTAGE OF TRACTABILITY WHEREAS “FULL” CALCULATIONS NOT CONCEIVABLE FOR ANYTHING OTHER THAN H_2^+

Can HHG from polyatomic molecules be mapped to electronic structure via SFA?





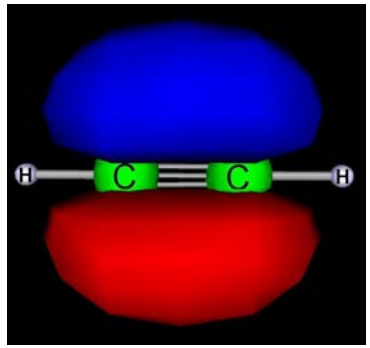


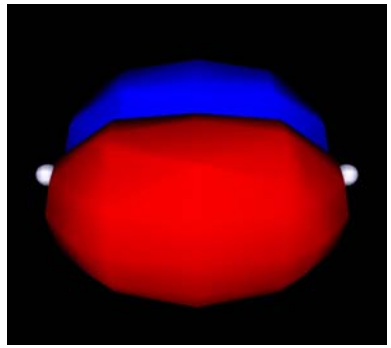
alignment in non-adiabatic regime

The molecules:

Acetylene (CH≡CH)

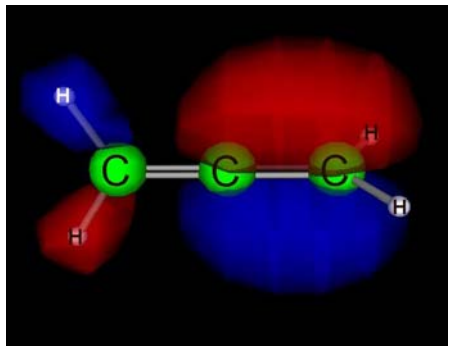
- Linear – Symmetric top
- Triple bond
- I_P = 11.4 eV

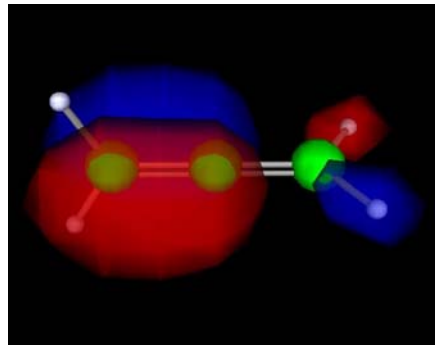




Allene (CH2=C=CH2)

- Non linear – Symmetric top
- Two double bonds
- I_P = 9.7 eV



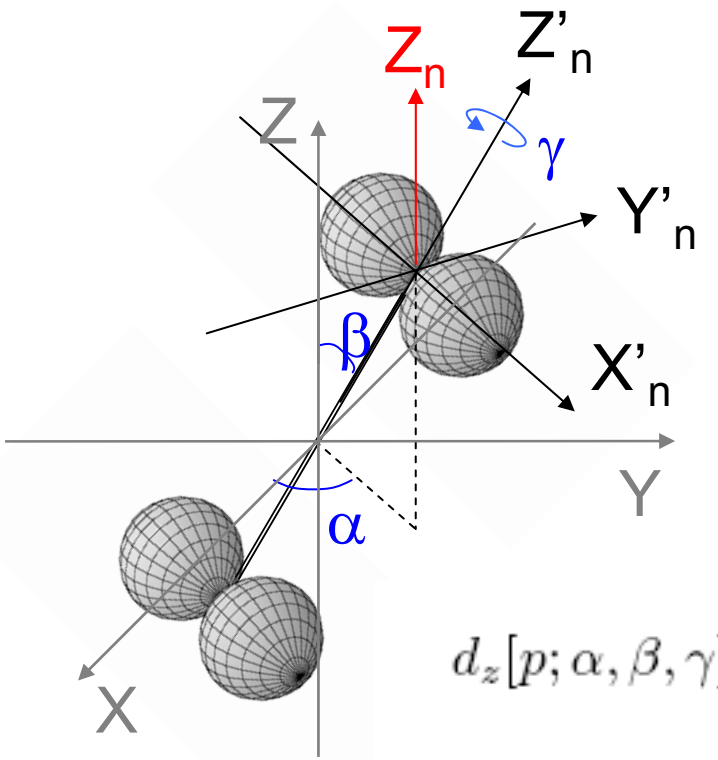


Calculations use the SFA (Lewenstein model):

$$z(t) = i\int_0^t dt' \int d^3p\, E_0 \cos(t')\, d_z^*[\mathbf{p}-\mathbf{A}(t')]\, d_z[\mathbf{p}-\mathbf{A}(t)]\, e^{-iS(p,t,t')} + c.c.$$

$$d_z[p] = \langle \psi_0 | z | e^{-ipz} \rangle$$

Molecular orbital is a LC of atomic orbitals of gaussian type:

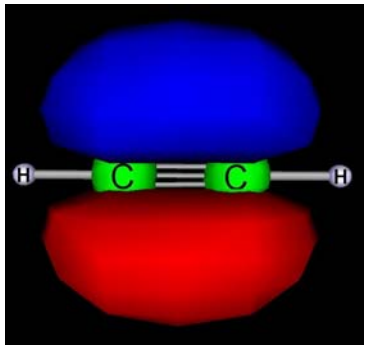


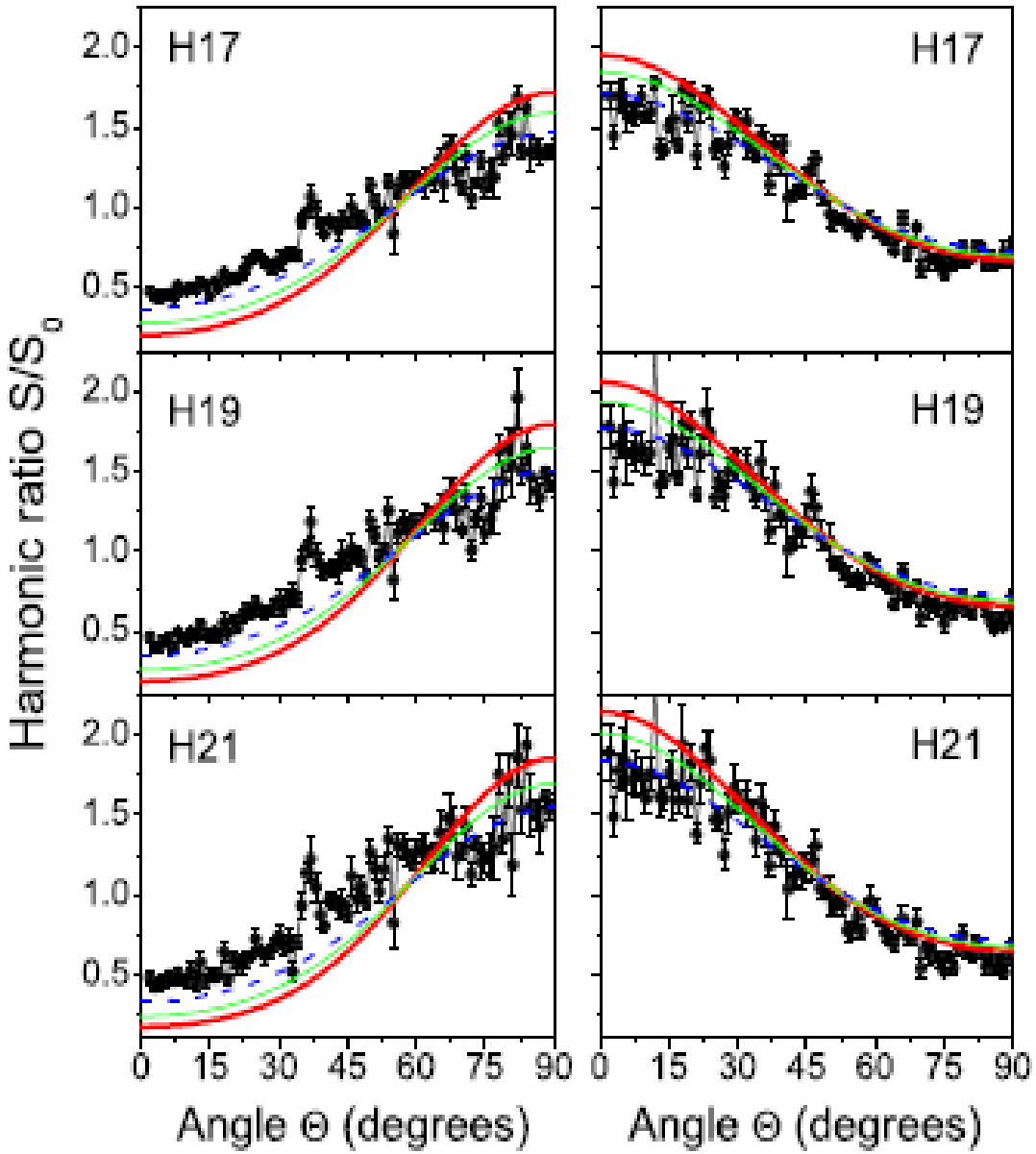
$$\psi_0(\mathbf{r}) = \sum_n \sum_i a_{n,i}\, G_i(\mathbf{r}-\mathbf{r}_n)$$

$$d_z[p] = \sum_n e^{-ipz_n} \sum_i a_{n,i}\, \langle G_i(\mathbf{r}) | z | e^{-ipz} \rangle$$

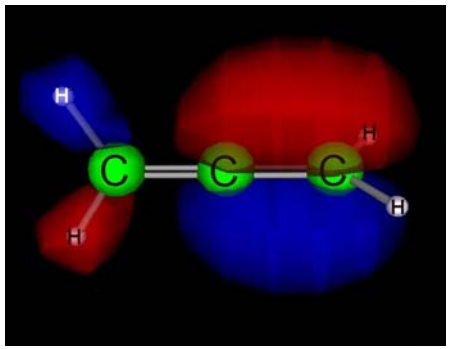
$$d_z[p;\alpha,\beta,\gamma] = \sum_n e^{-ipz_n} \sum_i a_{n,i}\, \langle G_i(\tilde{\mathbf{E}}_{\alpha\beta\gamma}\mathbf{r}) | z | e^{-ipz} \rangle$$

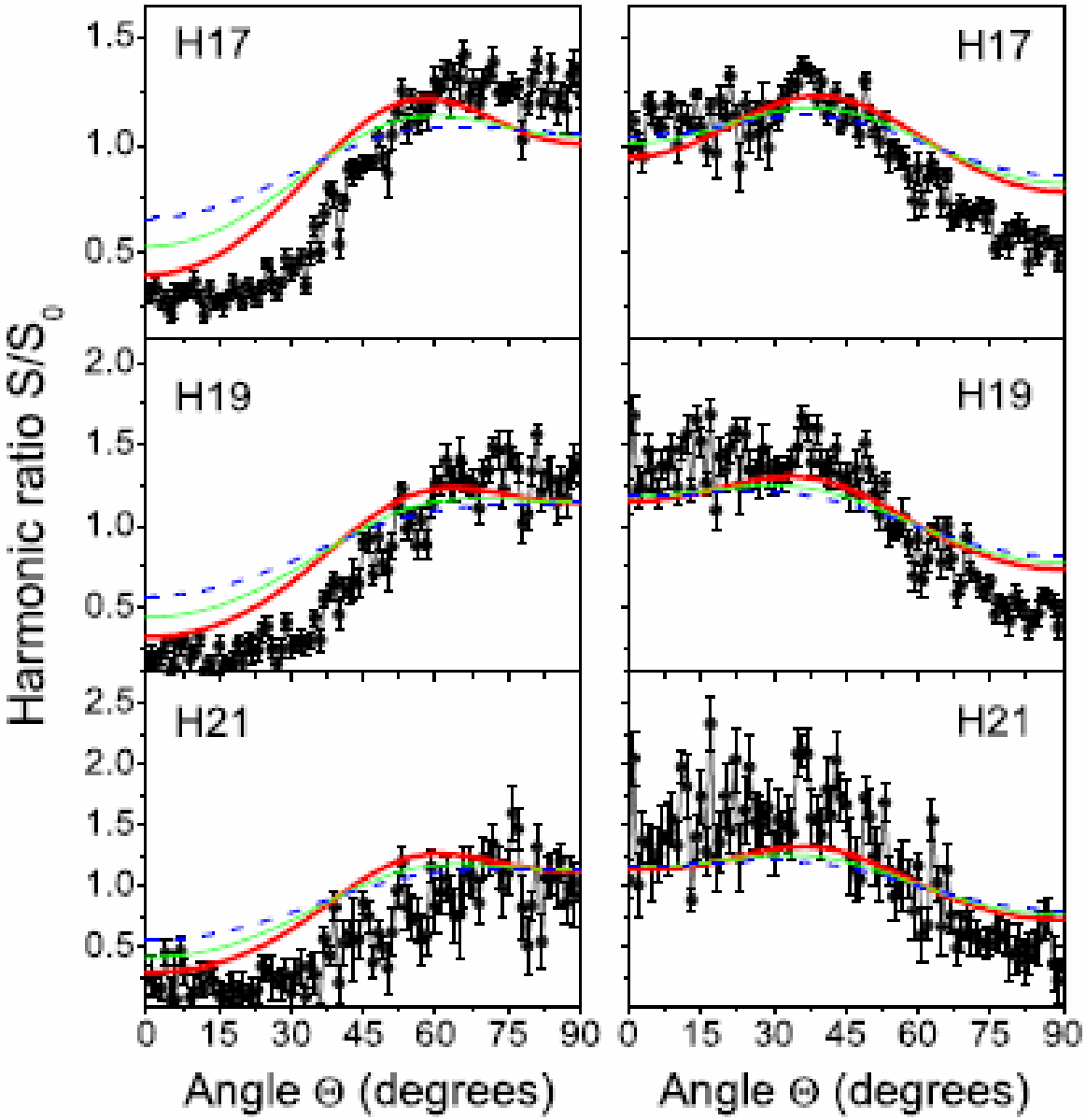
Acetylene– ratio of HHG signal in aligned to unaligned sample as a function of alignment direction θ





Allene – ratio of HHG signal in aligned to unaligned sample as a function of alignment direction θ





We conclude that SFA can work in some cases: (but unlikely to work always…)

-**The measurements of HH emission as a function of the alignment angle show a behaviour which is characteristic of the bonding π structure in the HOMO orbital of the molecules. Extra features are observed in Allene.**

- **A calculation using SFA reproduces the general features with fair quantitative agreement.**

-**The angular distribution of the aligned sample must be taken into account in order to compare the calculations quantitatively with the experimental results.**

- **At least for these molecules and harmonic orders SFA works, but further checks required**

Torres et al Physical Review Letters **98,** 203007 (2007)