ELI-ALPS Research Institute
TOWARDS THE SHARP END OF ATTOSCIENCE

High-order harmonics and attosecond pulse generation

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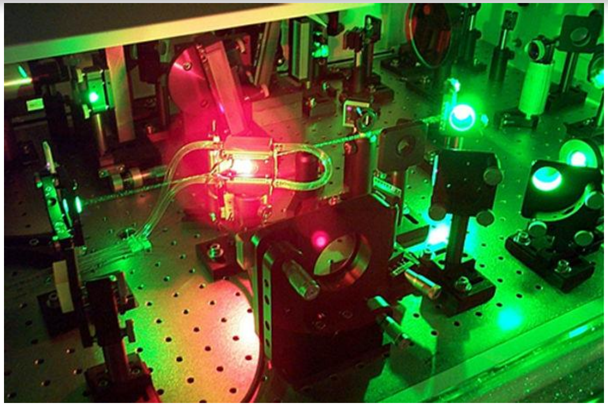
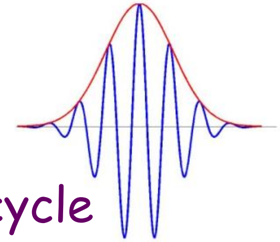
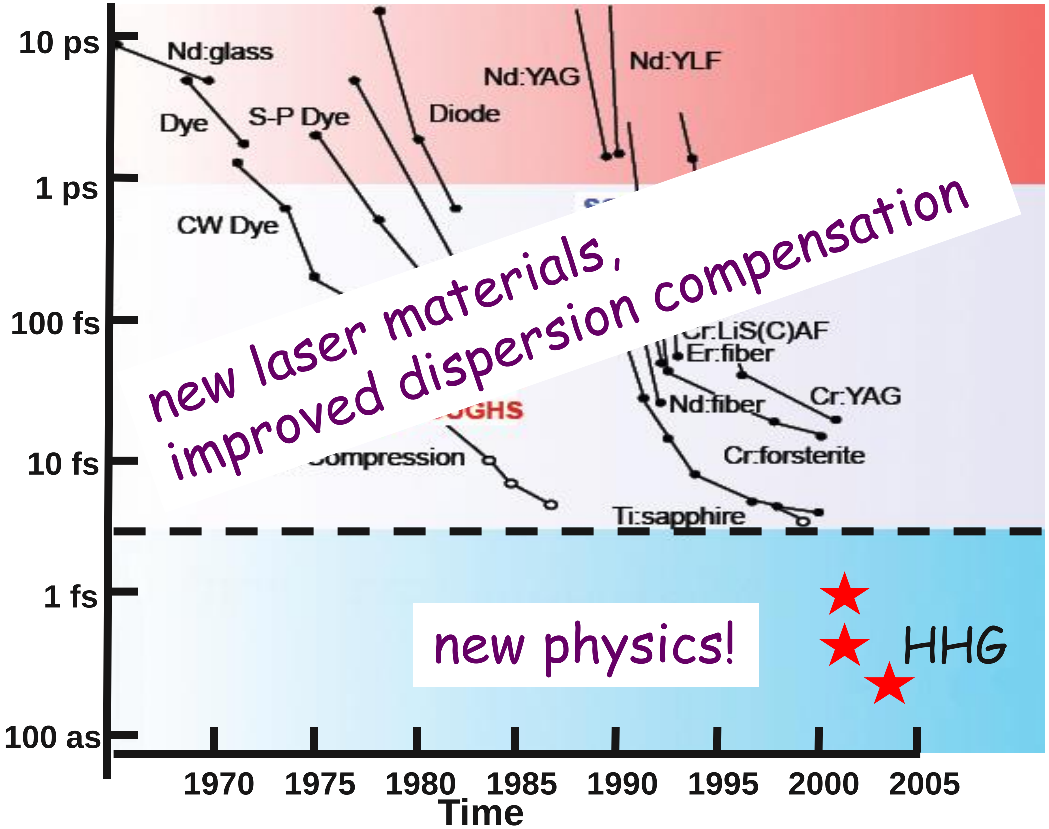
Fs laser sources

LASER mechanism
⇒ temporal and spatial coherence
⇒ oscillator, phase locking, broad gain medium
⇒ ultrashort pulse duration





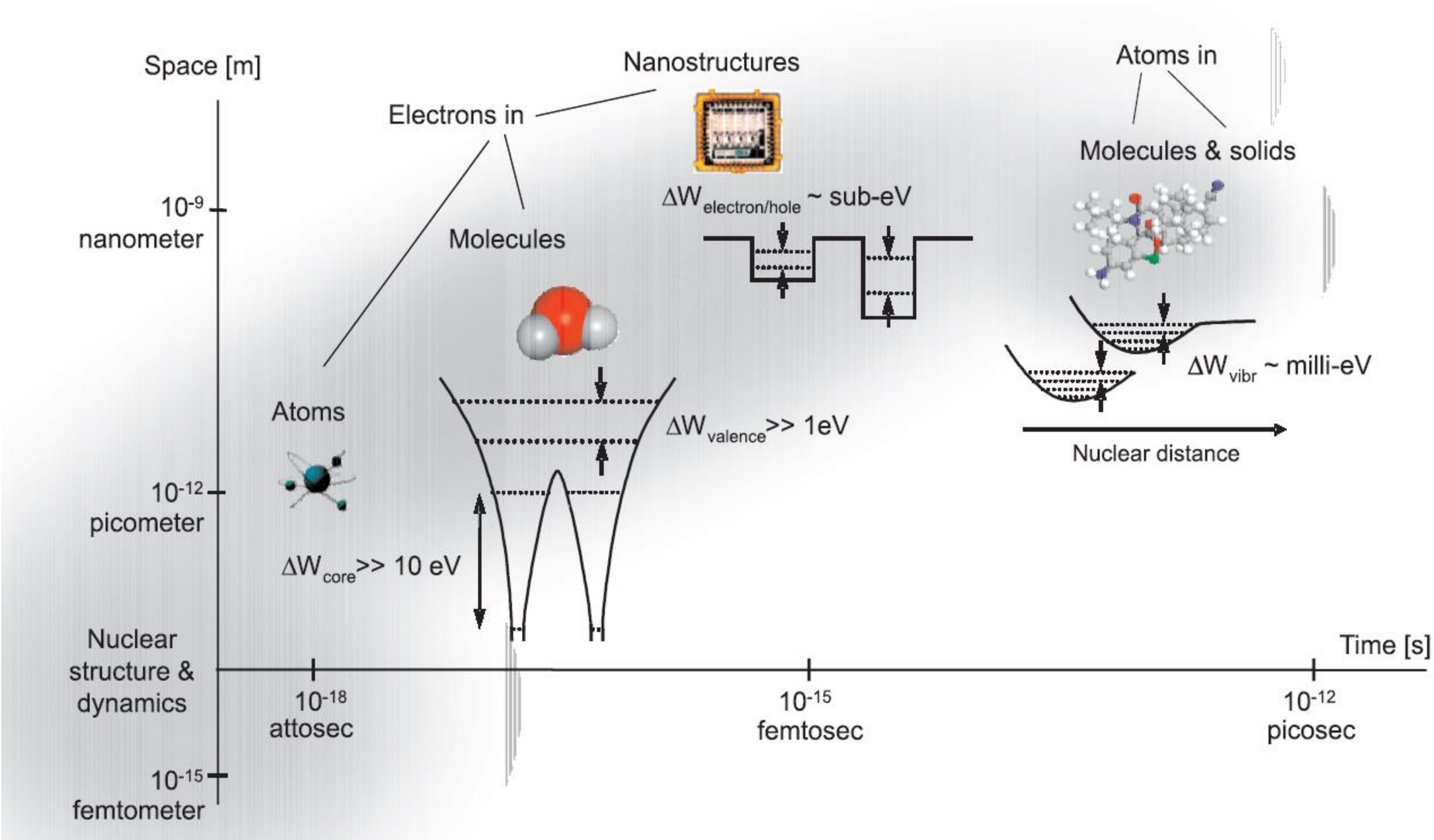
optical pulse:
800nm; 3,8 fs; 1,5 cycle



"Breaking the femtosecond barrier"

Corkum: Opt. Phot. News 6, 18 (1995)

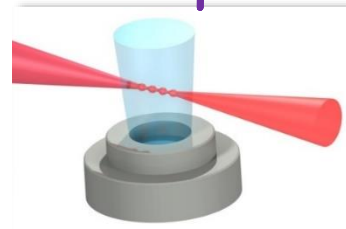
Why do we care about attosecond pulses?
Characteristic times



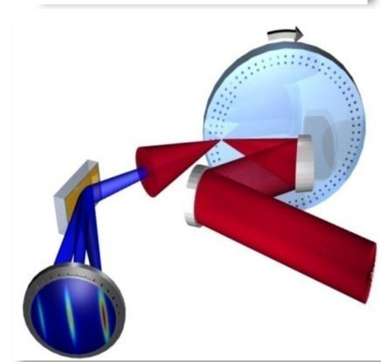
Krausz: RevModPhys 81, 163 (2009)

Mechanisms leading to femtosecond/attosecond XUV generation

Intense laser pulse + nonlinear phenomenon

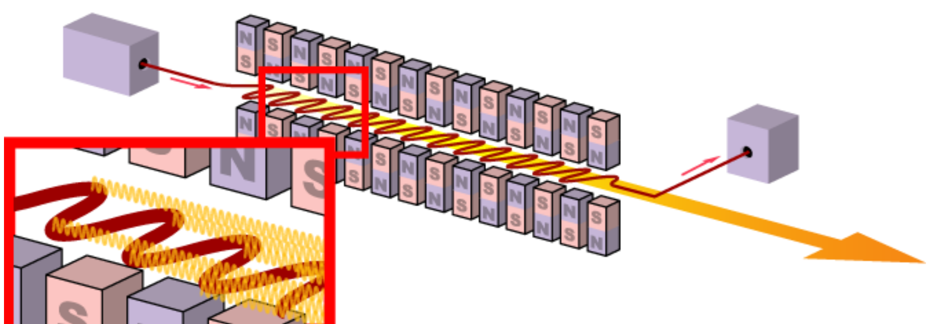


gas HHG



surface plasma HHG

Accelerated e- based schemes



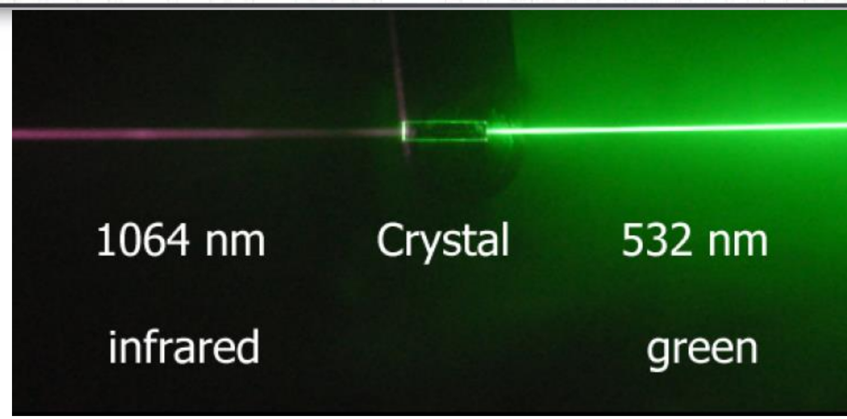
synchrotron, FEL, seeded FEL

Contents

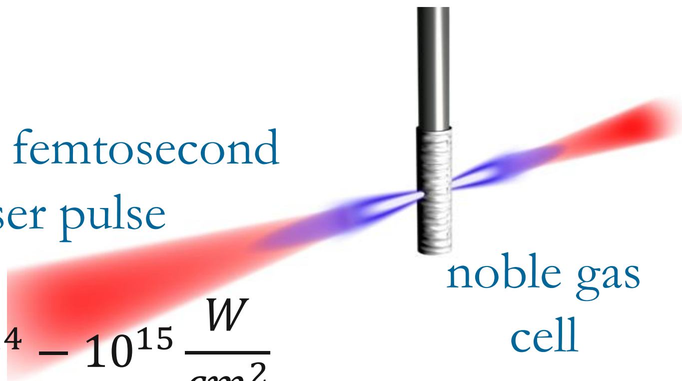
- High order harmonic generation in gaseous media
- Description of the generated radiation
- „Measuring" the radiation
- Phasematching in HHG
- Optimizing HHG

Experimental observation of HHG

High intensity laser light



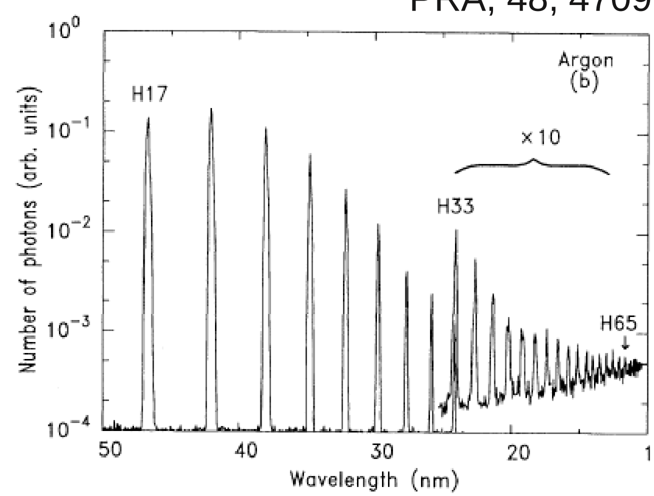
Focused femtosecond laser pulse



noble gas cell

$$I \approx 10^{14} - 10^{15}\,\frac{W}{cm^2}$$

Wahlström, PRA, 48, 4709



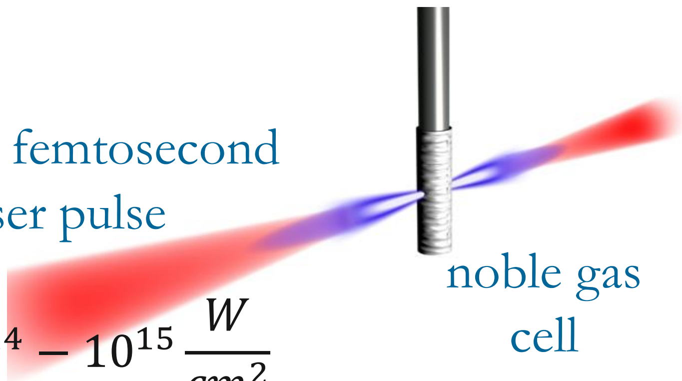
„Generating high order harmonics is experimentally simple."

Anne L'Huillier

Experimental observation of HHG

Focused femtosecond laser pulse

$$I \approx 10^{14} - 10^{15}\,\frac{W}{cm^2}$$

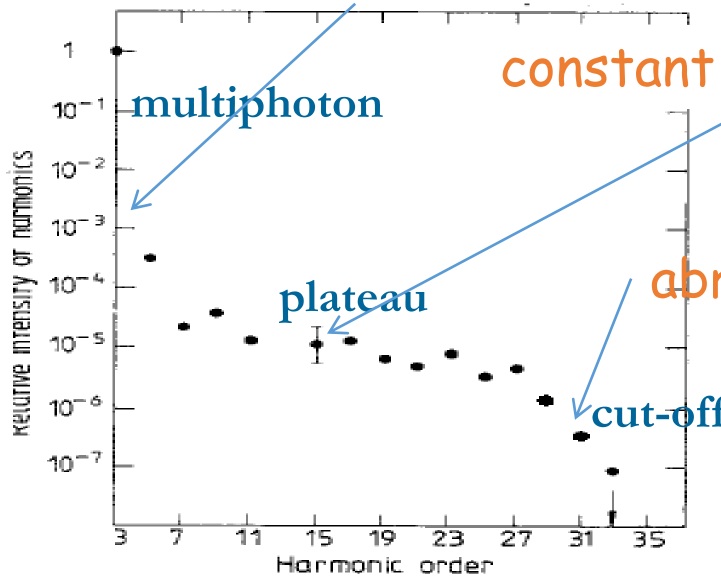


noble gas cell

perturbative decrease

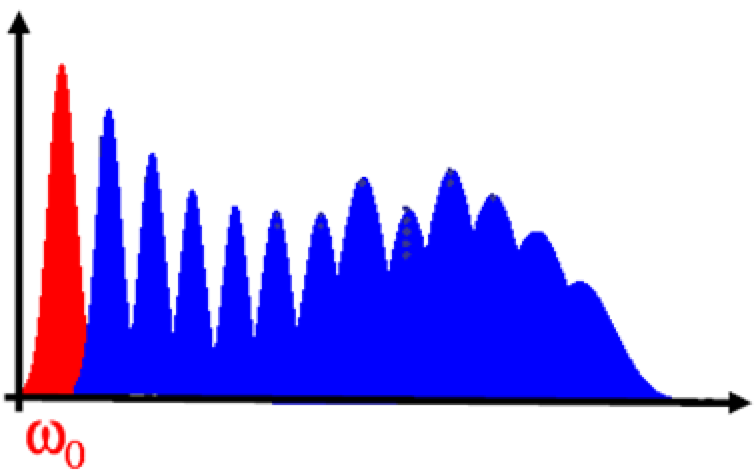
constant amplitudes

abrupt ending

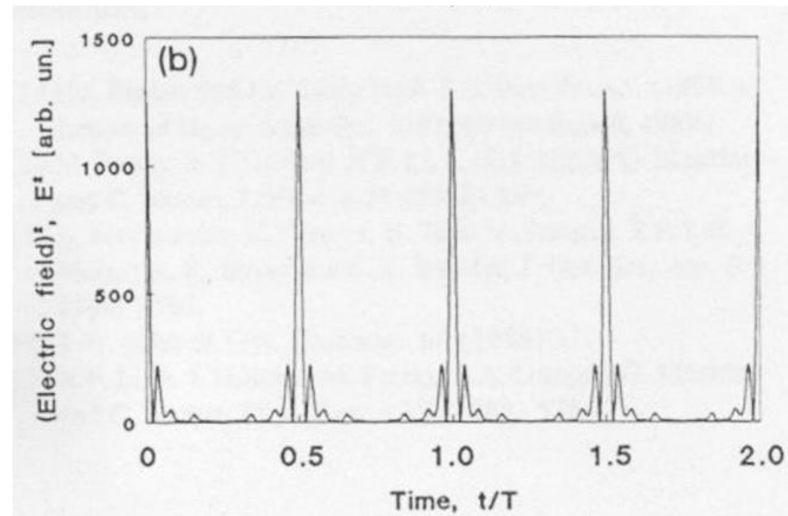


Ferray: J. Phys. B, 21, L31 (1988)

The „birth" of attosecond science



FT



Farkas, Phys. Lett. A (1992)

Atoms in a strong laser field

atomic electron: $E(r) = -\frac{1}{4\pi\varepsilon_0}\frac{e}{r^2}$ $\qquad r \approx 10^{-10}\,\mathrm{m}$ $\qquad E \approx 10^{11}\,\frac{\mathrm{V}}{\mathrm{m}}$

intensity = |Poynting vektor|

$$I = 2\cdot\frac{5\,\mathrm{mJ}}{\pi\cdot(100\,\mu\mathrm{m})^2\cdot 20\,\mathrm{fs}} = 1.6\times10^{19}\,\frac{W}{m^2} = 1.6\times10^{15}\,\frac{W}{cm^2}$$

$$I_0 \approx \frac{E}{\pi\cdot w_0^2\cdot\tau}\cdot 2$$

$$I = S = \frac{1}{2\mu_0}E_{\max}B_{\max} = \frac{1}{2}\varepsilon_0 c E_{\max}{}^2$$

$$E_{\max} = \sqrt{\frac{2\cdot I}{\varepsilon_0 c}} = \sqrt{\frac{2\cdot 1.6\times10^{19}\,\frac{\mathrm{W}}{\mathrm{m}^2}}{8.8\times10^{-12}\,\frac{\mathrm{As}}{\mathrm{Vm}}\cdot 3\times10^{8}\,\frac{\mathrm{m}}{\mathrm{s}}}} \approx 1.1\times10^{11}\,\frac{\mathrm{V}}{\mathrm{m}}$$

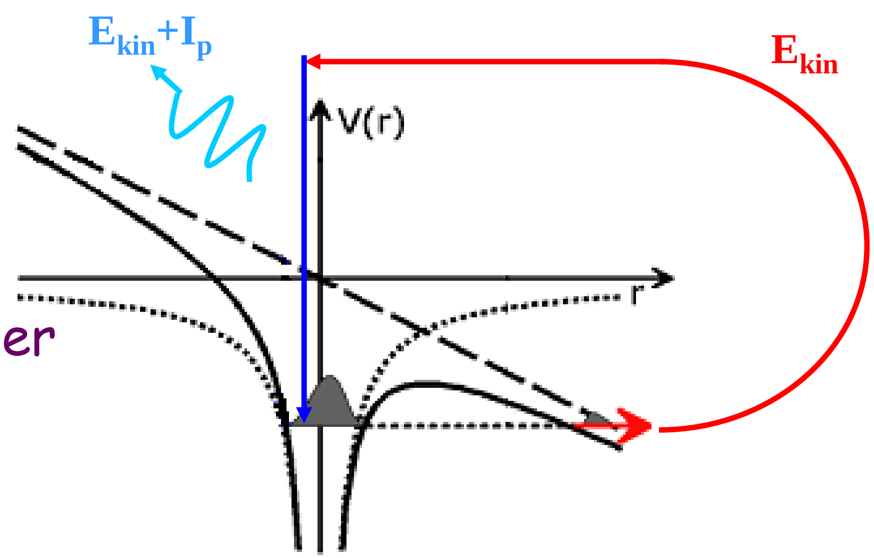
Field intensities $\sim 10^{14}\,\frac{W}{cm^2}$ correspond to the border between perturbative nonlinear optics and extreme NLO (where HHG occurs).

Three-step model

I Optical ionization through the distorted potential barrier

II Free electron propagating in the laser field, return to parent ion

III Electron captured by parent ion, photon emitted



Schafer: PRL, 70, 1599 (1993)
Corkum: PRL, 71, 1994 (1993)

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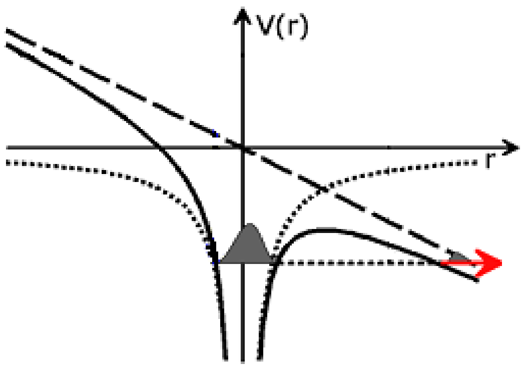
Classical description: Free electron in an oscillating E-field

$$E(t) = E_0 \sin(\omega t)$$ monochromatic field

$$F = -eE = m\ddot{x}$$ Newton's law of motion



$$\mathrm{x_i} = 0 \ \text{and} \ \mathrm{v_i} = 0 \quad \text{at} \quad t_i$$

P. B. Corkum, Phys Rev Lett 71, 1994 (1993)
K. Varjú, Am. J. Phys. 77, 389 (2009)

$$v(t) = -\frac{eE_0}{m\omega}\left[\cos(\omega t) - \cos(\omega t_i)\right]$$

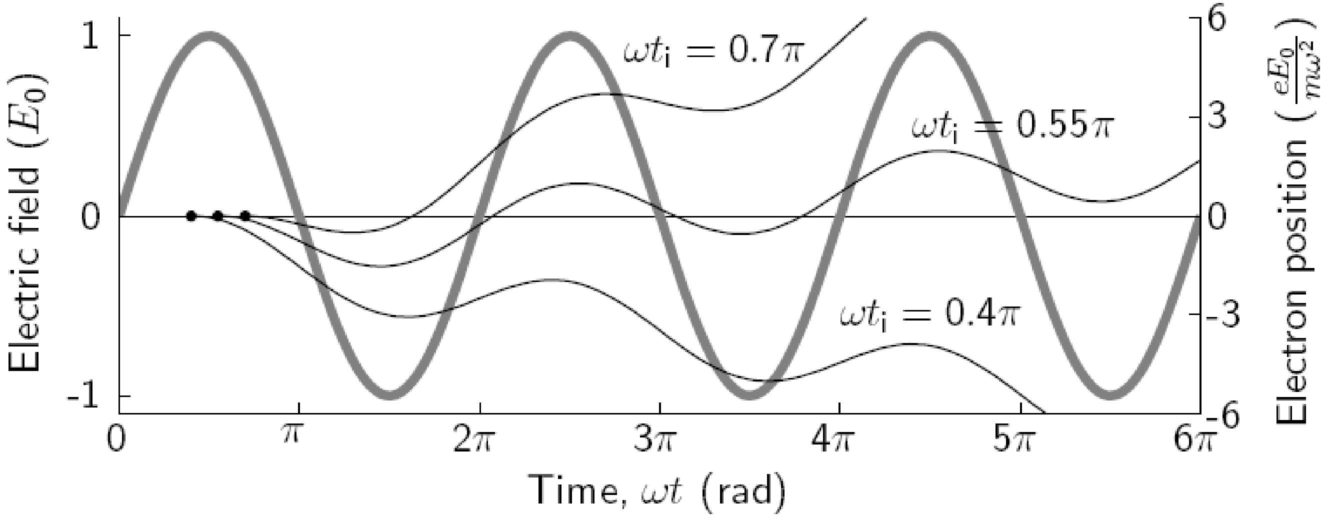
$$x(t) = \frac{eE_0}{m\omega^2}\left[\sin(\omega t) - \sin(\omega t_i) - \omega(t - t_i)\cos(\omega t_i)\right]$$

analytic solution

Assumptions:
- 1-dim case
- the electron is ionized into the vicinity of the ion with zero velocity, and recombines if its path returns to the same position (no quantum effects!)
- while in the laser field, the effect of the Coulomb field is neglected
- if the electron recombines, a photon is emitted with energy Ekin+ Ip

Closed electron trajectories

$$x(t) = \frac{eE_0}{m\omega^2}\left[\sin(\omega t) - \sin(\omega t_i) - \omega(t - t_i)\cos(\omega t_i)\right]$$



$$x_0 = \frac{2eE_0}{m\omega^2}$$

typical parameters:
$5 \times 10^{14}\,\mathrm{W/cm^2}$,
800 nm, $x_0 = 1.95$ nm

1, the electron may (or may not) return

2, return of the electron depends on ionization time

3, energy gained in the laser field (~ velocity squared ~ slope of trajectory) depends on ionization time

Producing photons

If the electron returns to the ion, it may recombine and a photon is emitted with energy:

$$\hbar\omega = I_p + \frac{1}{2}mv^2 = I_p + 2U_p\big(cos(\omega t) - cos(\omega t_i)\big)^2$$

where I_p is the inoization potential and $U_p = \frac{e^2 {E_0}^2}{4m\omega^2}$ is the ponderomotive potential.

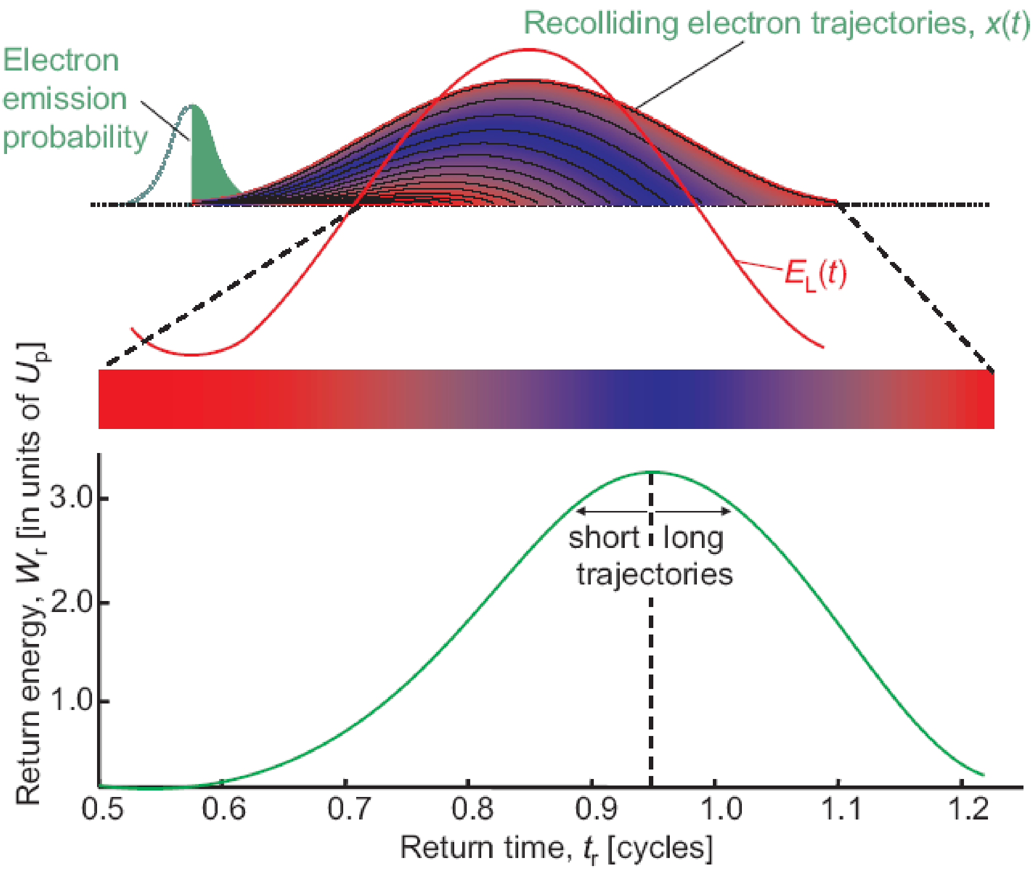
$U_p\,[eV] = 9.33 \times 10^{-14} I_0 \lambda^2$ where I_0 is expressed in W/cm^2 and λ in µm.

Spectrum of the emitted radiation

Cutoff @ 3.17 U_p

$$U_p\,[eV] = 9.33 \times 10^{-14} I_0 \lambda^2$$

Two electron trajectories contribute to the emission of the same photon energy

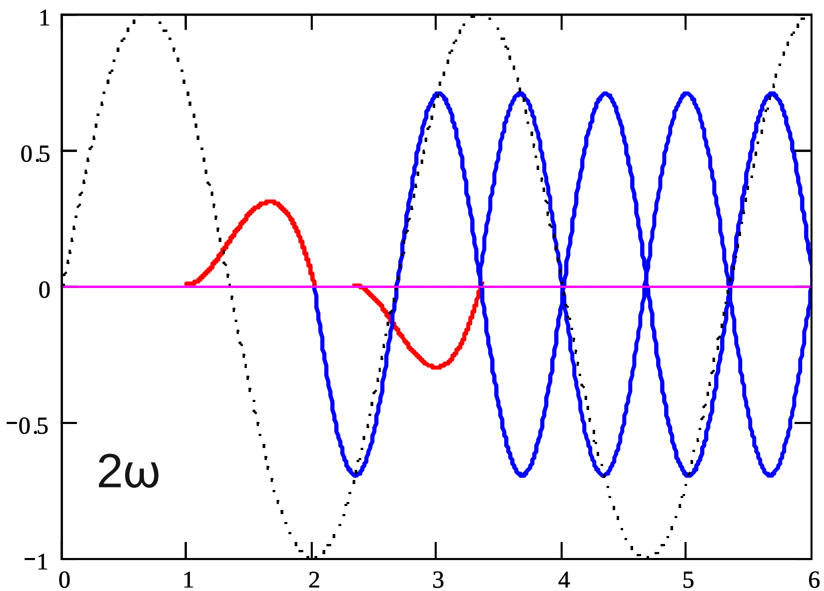


Trajectory	Tunneling time (T)	Recombination time (T)
Electrons that do not recombine	$0 < t_i < 1/4$	-
Long trajectory	$1/4 < t_i < 1/3$	$0.95 < t_r < 5/4$
Short trajectory	$1/3 < t_i < 1/2$	$1/2 < t_r < 0.95$

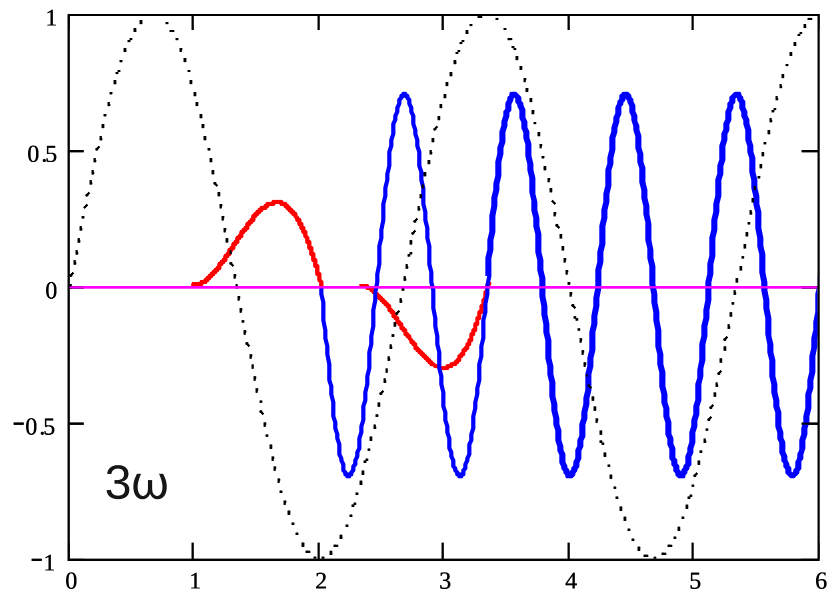
Varjú, Laser Phys 15, 888 (2005)
Krausz, Ivanov, Rev Mod Phys 81, 163 (2009)

Why (only) odd harmonics?

Due to the symmetry of the system the photon emission is periodic with T/2, so we expect spectral periodicity of 2ω. Due to the π phase-shift of the driving field between consecutive events the even harmonics destructively interfere:



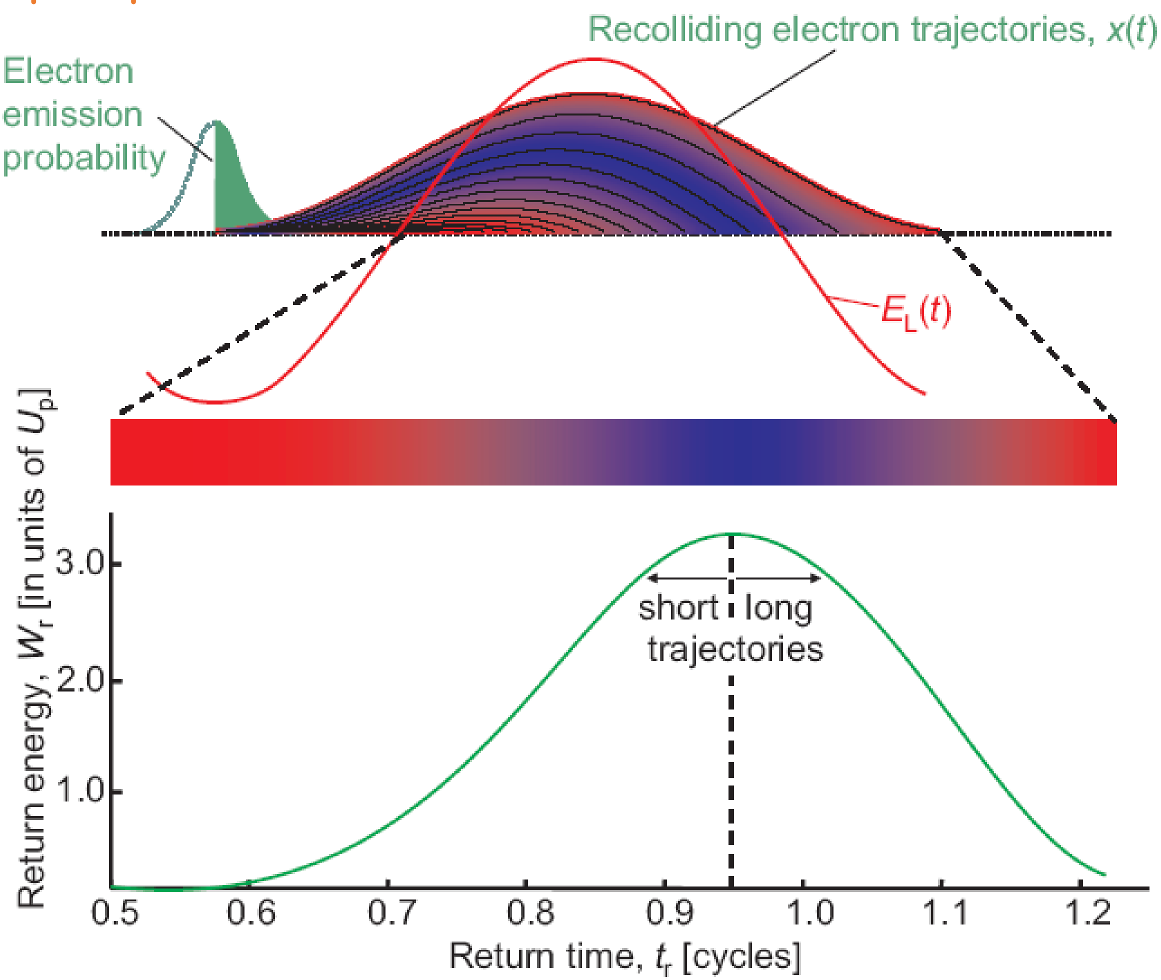
destructive interf.



constructive interf.

Time-frequency characteristics

E_{kin} depends on return time → photon energy / frequency will vary with time → chirped pulses



Varjú, Laser Phys 15, 888 (2005)
Krausz, Ivanov, Rev Mod Phys 81, 163 (2009)

Short vs long trajectory:

delayed in time
opposite chirp
phase has different intensity d

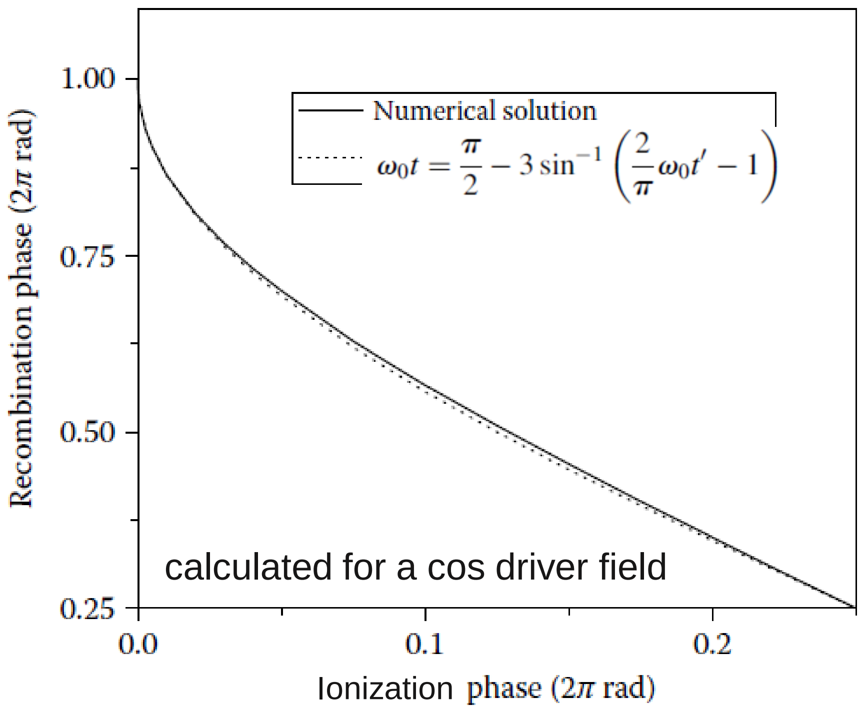
Numerical solutions vs fitting functions

$$x(t_r) = \frac{eE_0}{m\omega^2}\left[\sin(\omega t) - \sin(\omega t_i) - \omega(t - t_i)\cos(\omega t_i)\right] = 0$$

1) find the return time as a function of ionization time
2) calculate the return energy (to obtain photon energy) at return time

$$\{t_i, t_r, E_{kin}\}$$

Solutions can be approximated by

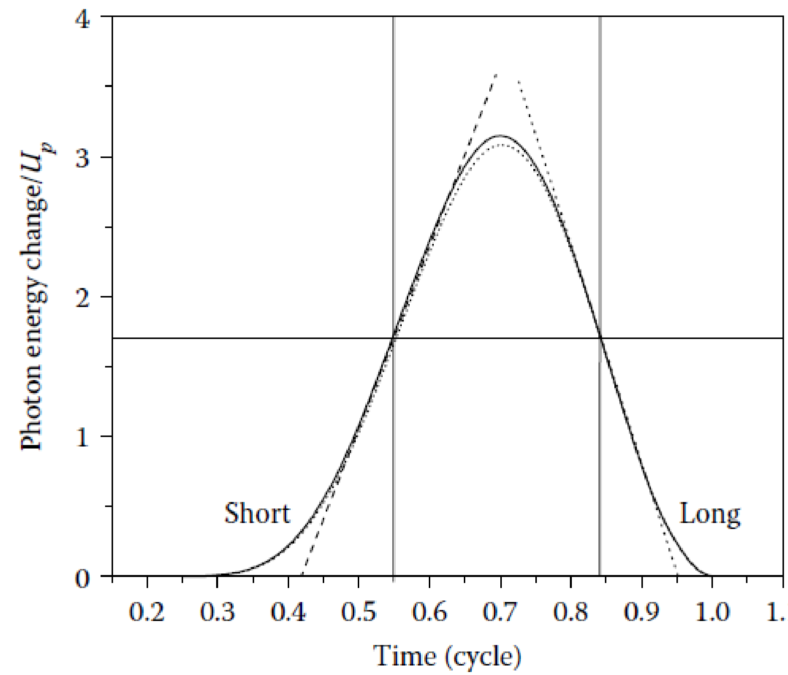


Note: return times span over 0.75 cycle, and the process is repeated every half cycle, the generated radiation don't necessarily have attosecond duration!

Chang: Fundamentals..

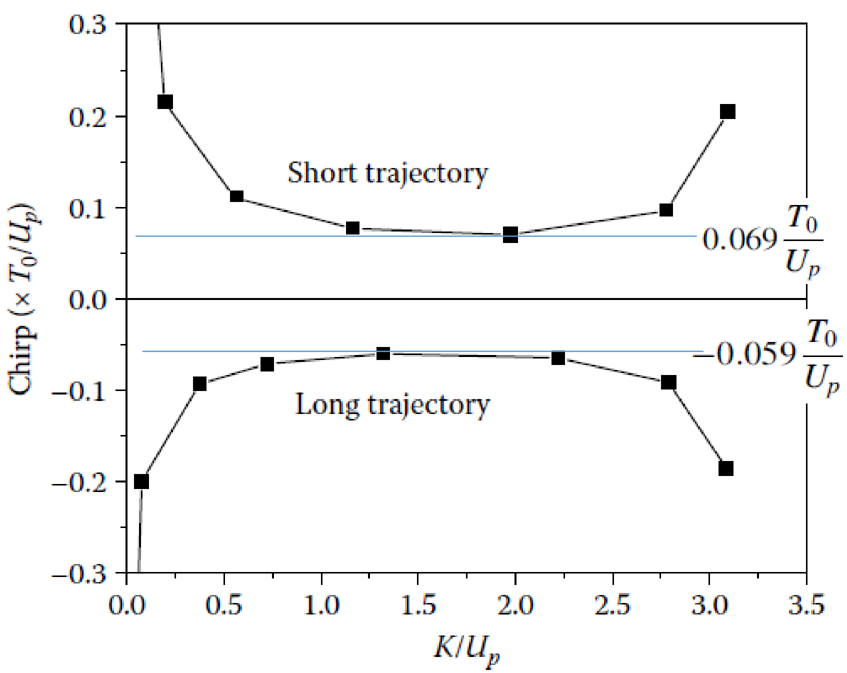
Chirp of the harmonic radiation

$$\frac{K(t)}{U_p} = 2\left\{\sin(\omega_0 t) - \cos\left[\frac{\pi}{2}\sin\left(\frac{1}{3}\omega_0 t - \frac{\pi}{6}\right)\right]\right\}^2$$



Observations:

1) the short trajectory is positively chirped, while the long trajectory is negatively chirped
2) the chirp is almost linear for most part of the spectral range



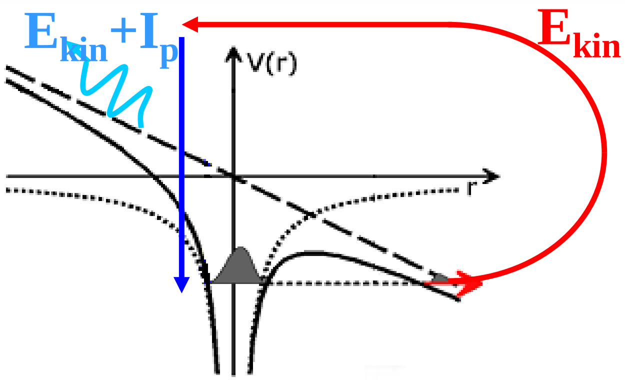
e.g. 2.67 fs,
GDD=10 as/eV = 6.6×10^3 as^2

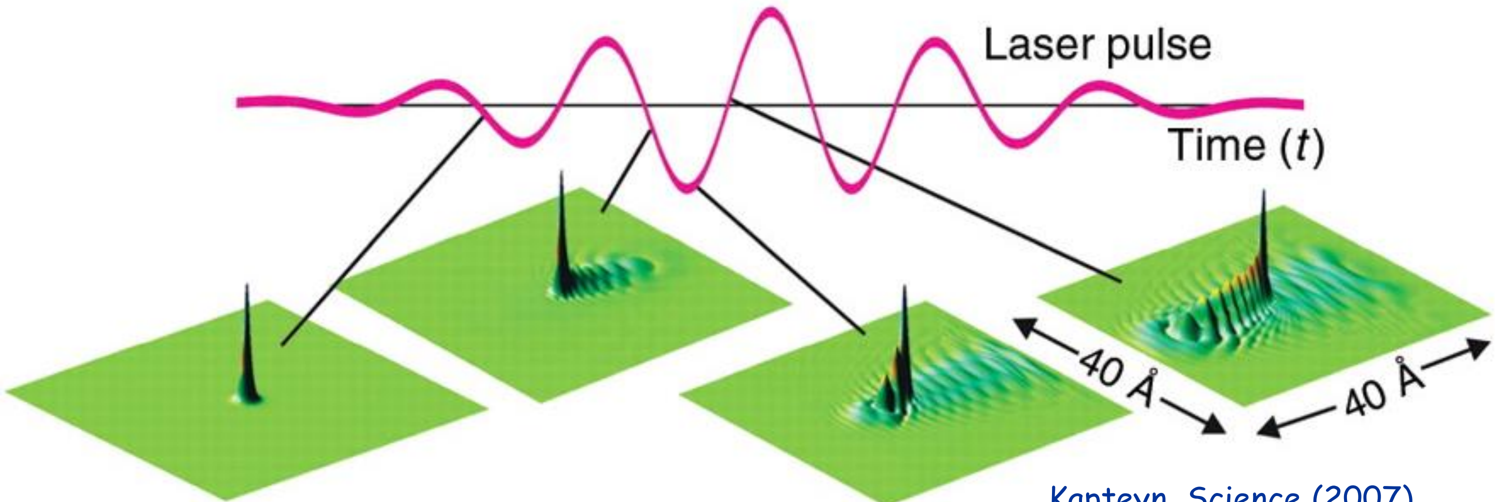
$$GDD[\mathrm{as}^2] = 16.3 \times 10^{17}\,\frac{1}{I_0\lambda_0}$$

inversely proportional to laser intensity and wavelength

Chang: Fundamentals…

HHG in the quantum picture





Kapteyn, Science (2007)
Lewenstein, Phys Rev A (1994)

low efficiency!!!

Harmonic radiation

is a result of oscillation of the quasi-bound electron.
Desciption: quantum mechanics

TDSE:

$$i\frac{\partial}{\partial t}|\Psi(\vec{x},t)\rangle = [-\tfrac{1}{2}\nabla^2 + V(\vec{x}) - \vec{E}(t)\cdot\vec{x}]|\Psi(\vec{x},t)\rangle.$$

- one-electron approximation (initially in the bound ground state)
- classical laser field (high photon density)
- dipole approximation (we neglect the magnetic field and the electric quadrupole)
- laser field is assumed to be linearly polarized

SOLUTION: numerical integration
long computational time

Strong field approximation (SFA)

- the ionized electron is under the influence of the laser field, only (Coulomb potential neglected)
- only a single bound state is considered
- neglect depletion of the bound state

Dipole moment:

$$\vec{x}(t) = \langle \Psi(t) | \vec{x} | \Psi(t) \rangle$$

capture
of electron

$$\vec{x}(t) = i \int_0^t dt' \int d^3\vec{p} \;\; \vec{d}^*(\vec{p} - \vec{A}(t))$$

electron propagation
in the laser field

$$\times \exp[-iS(\vec{p},t,t')] \;\; \vec{E}(t') \cdot \vec{d}(\vec{p} - \vec{A}(t')) + \text{c.c.},$$

Lewenstein integral

ionization transition

M. Lewenstein Phys.Rev.A 49, 2117 (1994)

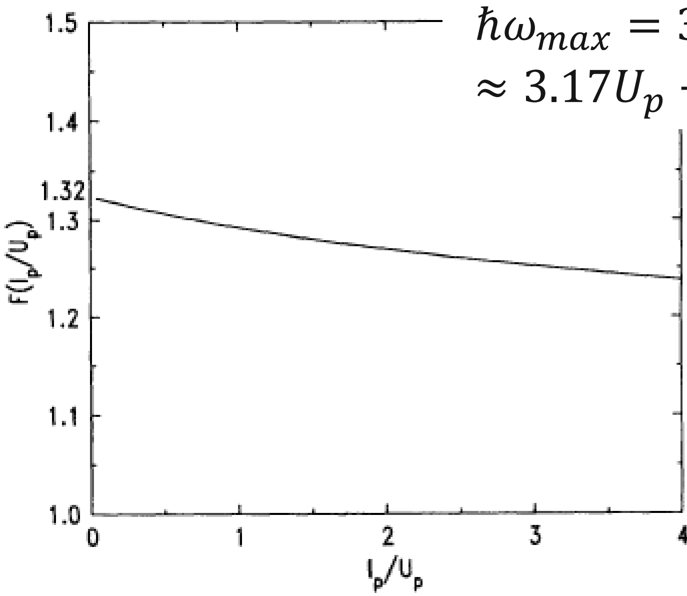
The cutoff law - QM

classical calculation: energy conservation $I_p + 3.17 \cdot U_p$

quantum description:

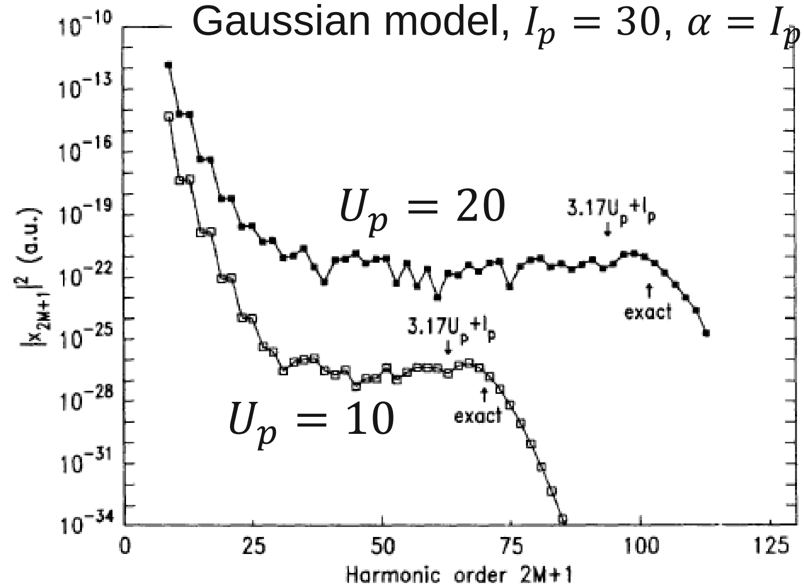
- tunneling; the electrons are not born at $x_0 = 0$, but at a position $I_p = x_0 \cdot E(t')$, thus the e can gain additional kinetic energy between x_0 and the origin

- diffusion; averages (and decreases) the additional kinetic energy effect



$$\hbar\omega_{max} = 3.17U_p + F \cdot I_p$$
$$\approx 3.17U_p + 1.32 \cdot I_p$$

Gaussian model, $I_p = 30$, $\alpha = I_p$

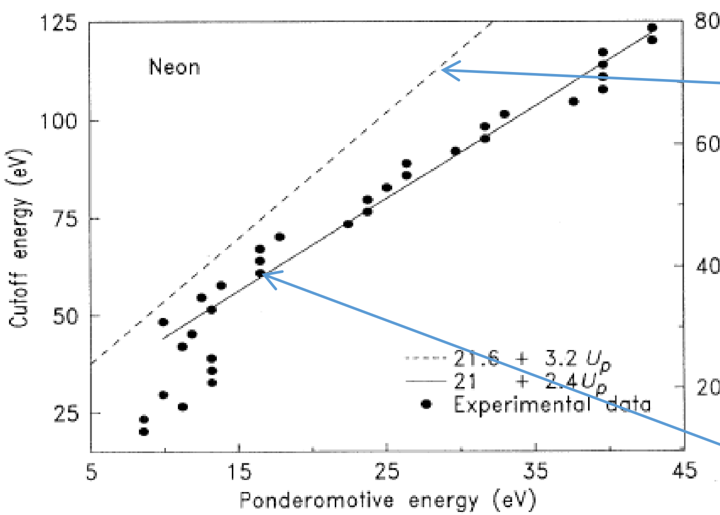


M. Lewenstein Phys.Rev.A 49, 2117 (1994)

The cutoff law – in reality

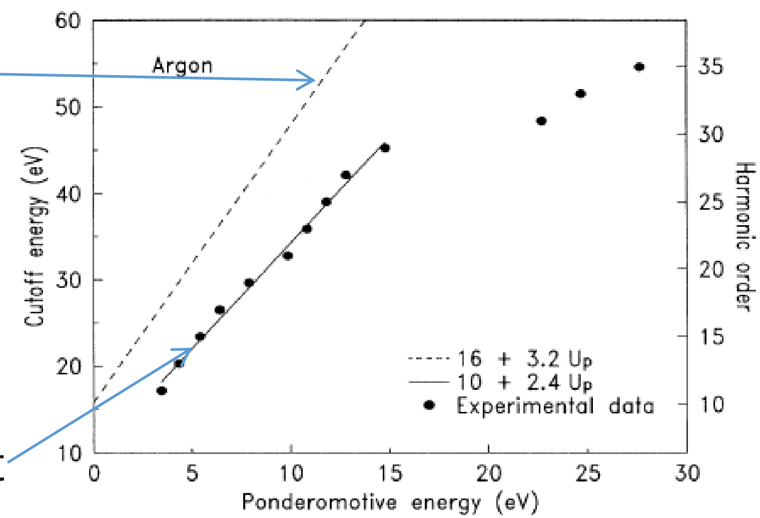
At high intensities saturation effects restrict the maximum photon energy to below cutoff, when the medium gets fully ionized before the peak (especially for long driving pulses)

- depletion of ground state
- prevents phasematching due to high concentration of electrons
- contributes to defocusing of the laser pulse



Single-atom

Best linear fit



Macroscopic effects play an important role!!

Harmonic spectrum

Harmonics are emitted as a result of the dipole oscillations

Fourier transform of the dipole moment:

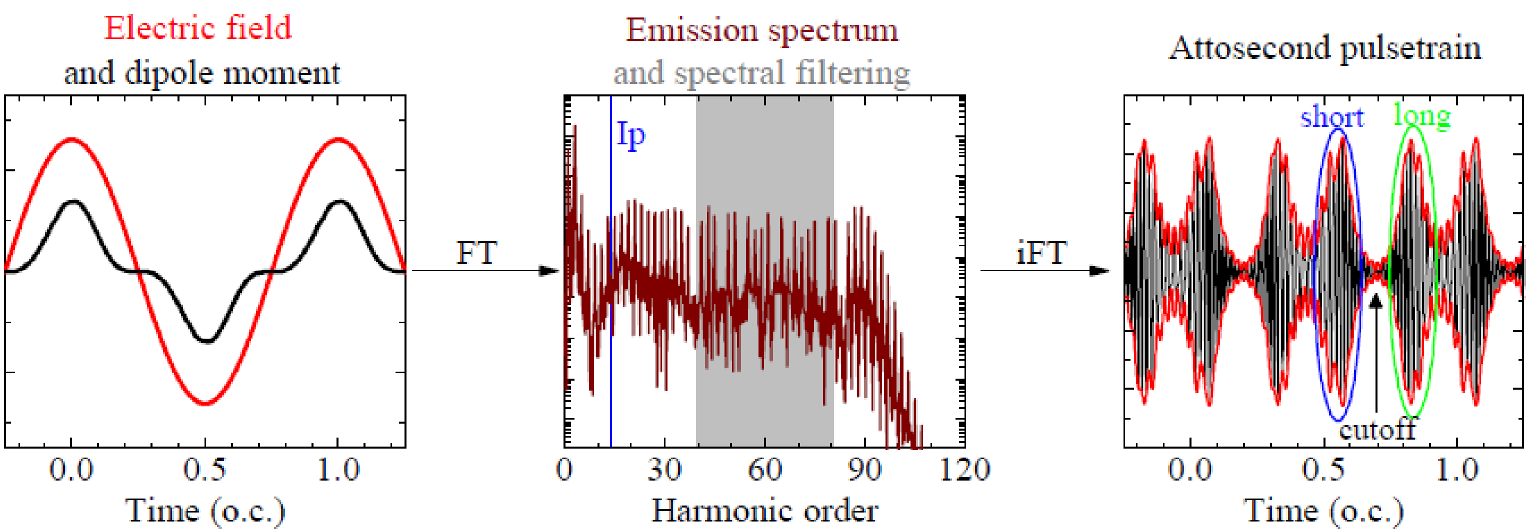
$$x(\omega) = \int_{-\infty}^{+\infty} dt\ x(t)\exp(i\omega t)$$

can be decomposed as

$$x(\omega) = \sum_s |x_s(\omega)| \exp[i\Phi_s(\omega)]$$

harmonic emission rate

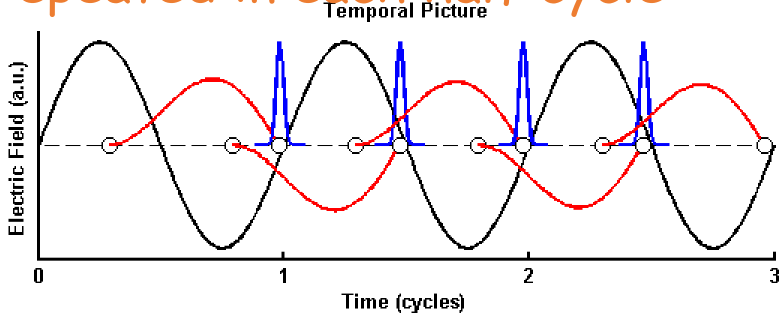
$$W(\omega) \propto \omega^3 |x(\omega)|^2$$



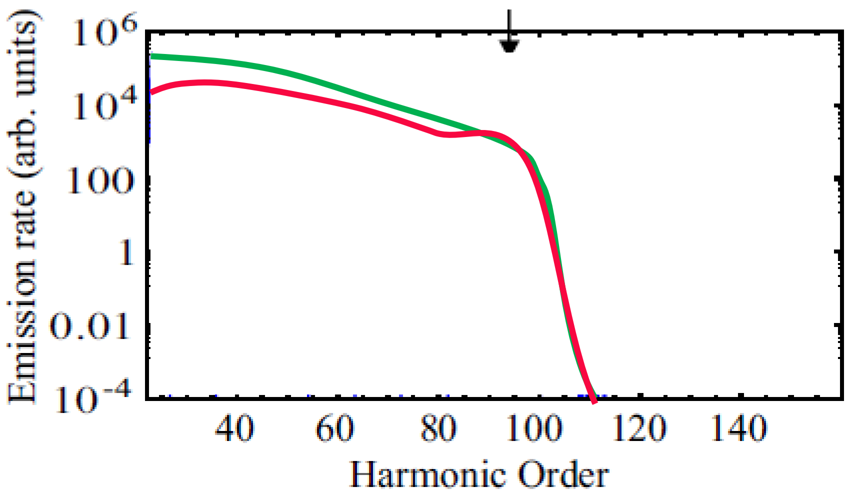
λ = 800 nm,
$h\nu$=1.55 eV
$I_0 = 6\cdot10^{14}$ W/cm^2
I_p = 21.5 eV (Ne)

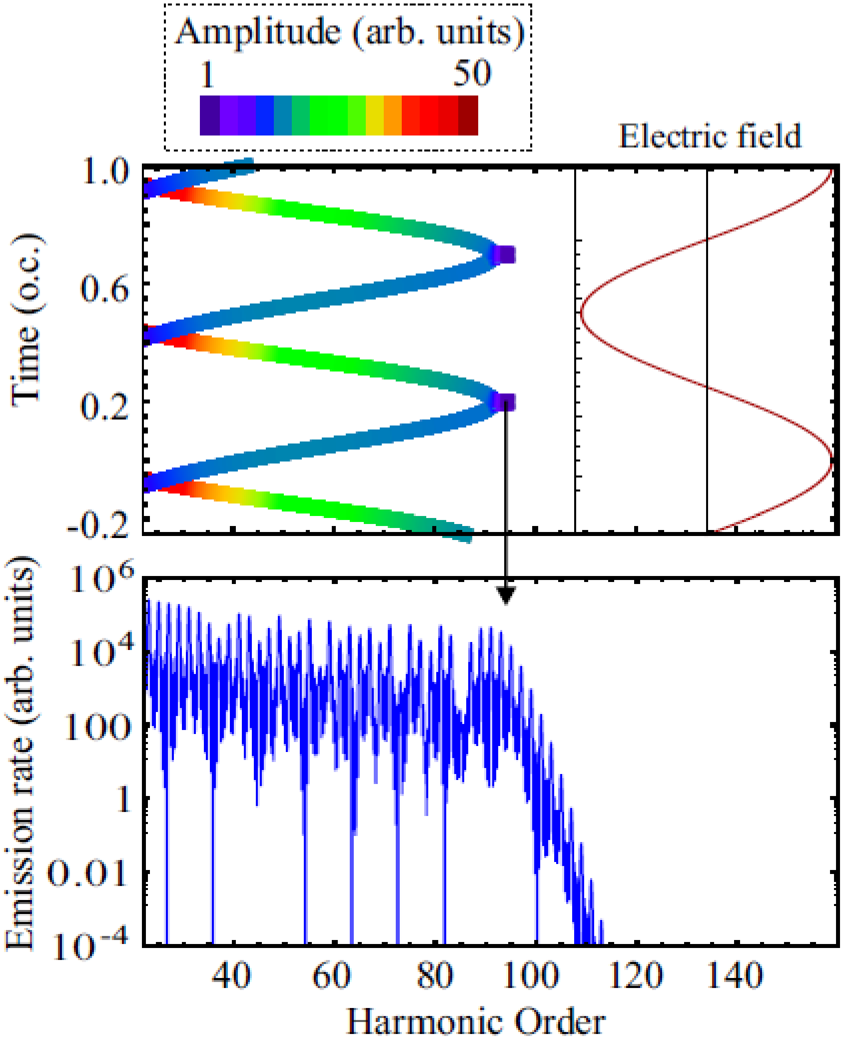
Periodicity

harmonic emission process
repeated in each half cycle



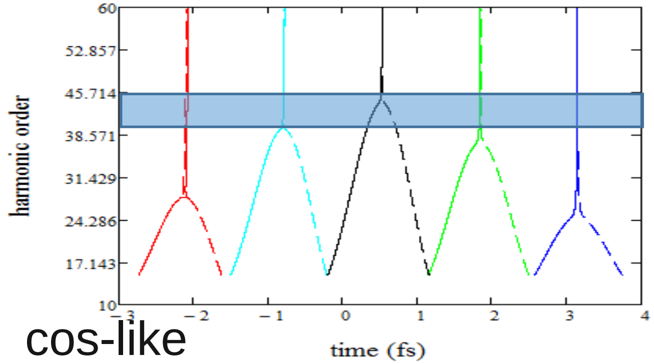
radiation emitted in each half cycle



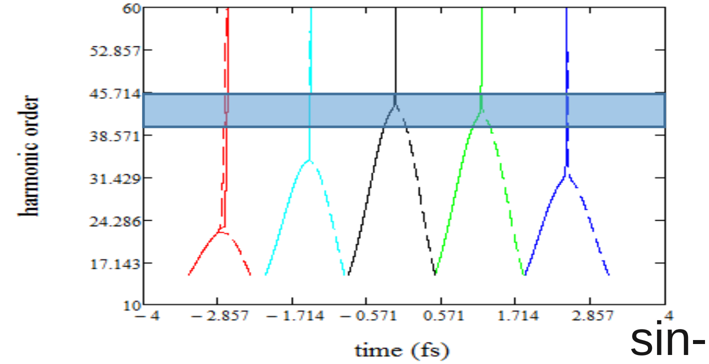


HHG by a short IR pulse

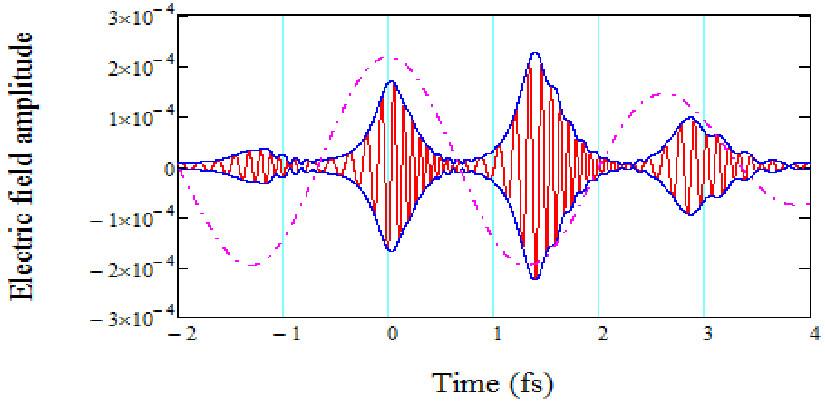
5 fs laser pulse,
800 nm,
$2.5*10^{14}$ W/cm^2
argon gas

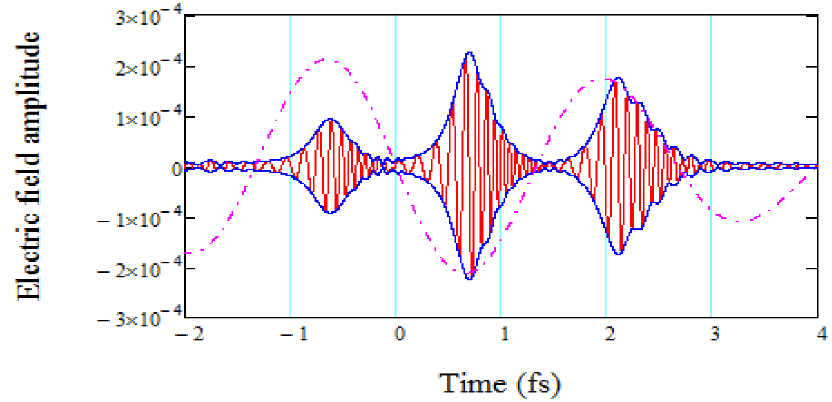


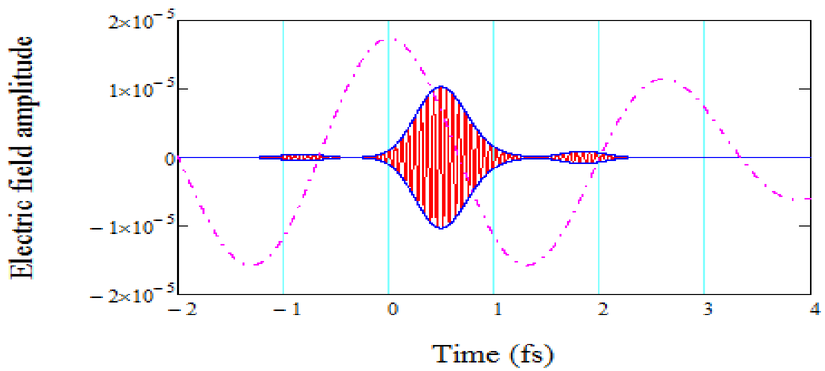
cos-like

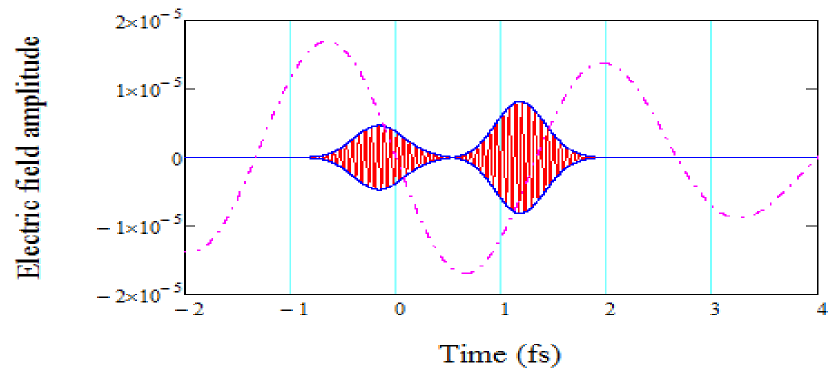


sin-like









using a narrow spectral window (FWHM 3 harmonic orders), a single attosecond pulse can be selected - only short trajectories are considered!

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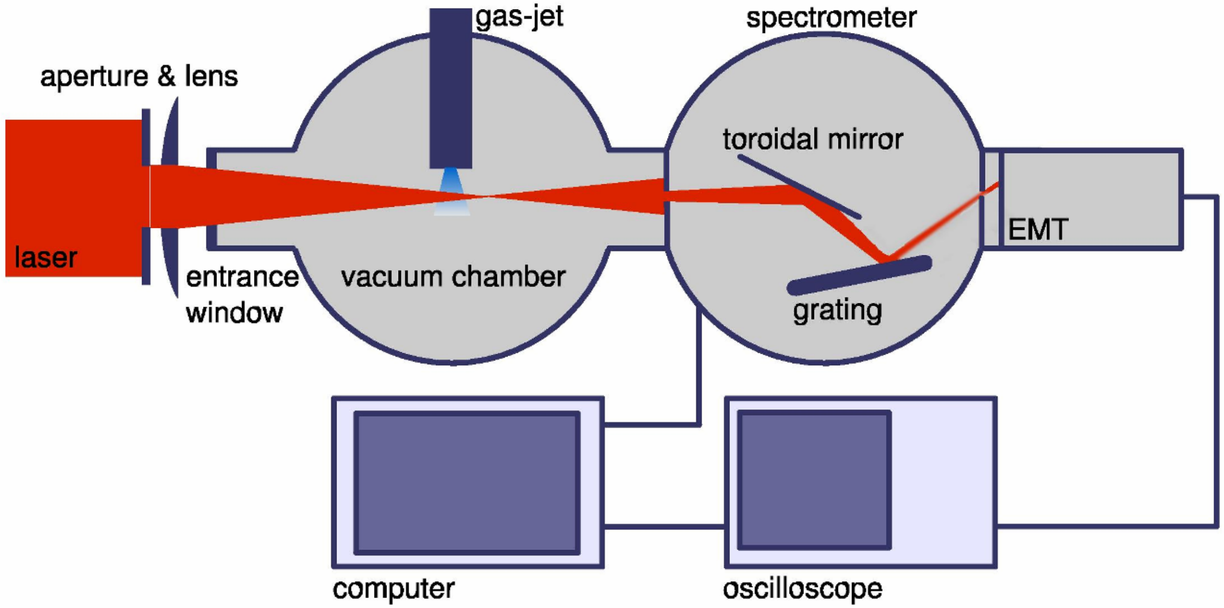
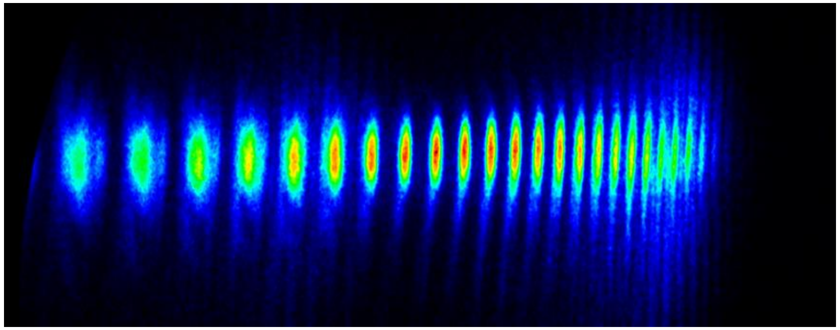
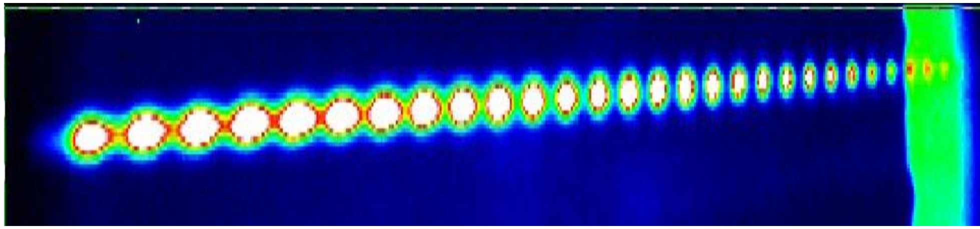
Photon detection





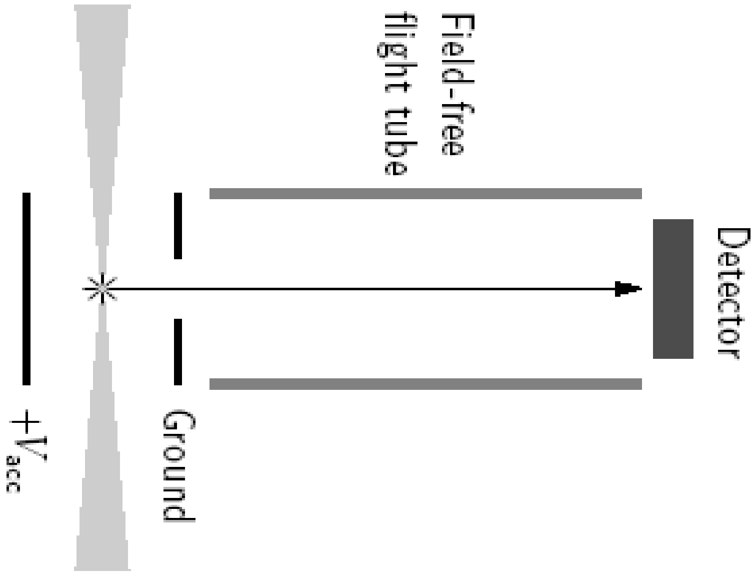
Nisoli et al., 2002



Kazamias et al.

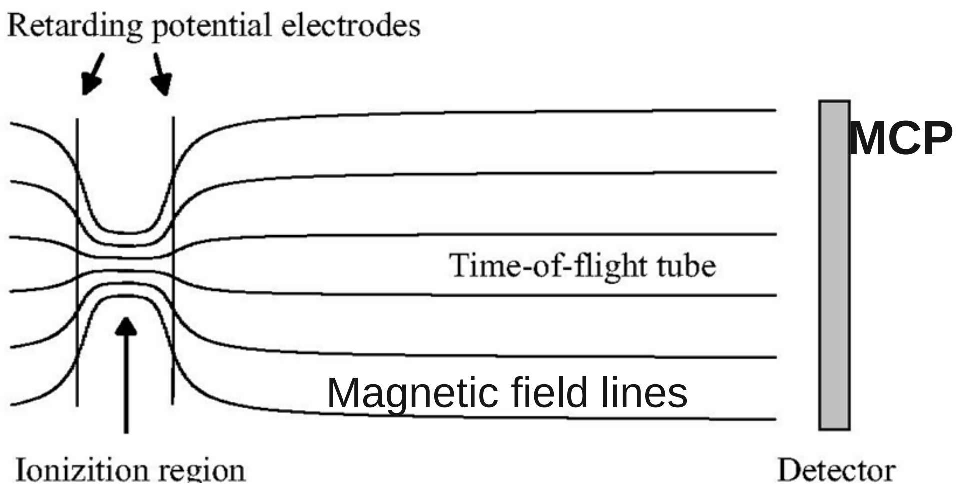
Spectral amplitude – no information about temporal features

Electron / ion detection produced in photo-ionization



Time-of-flight spectrometer

$$t \propto \frac{1}{v} \propto \sqrt{\frac{m}{q}}$$

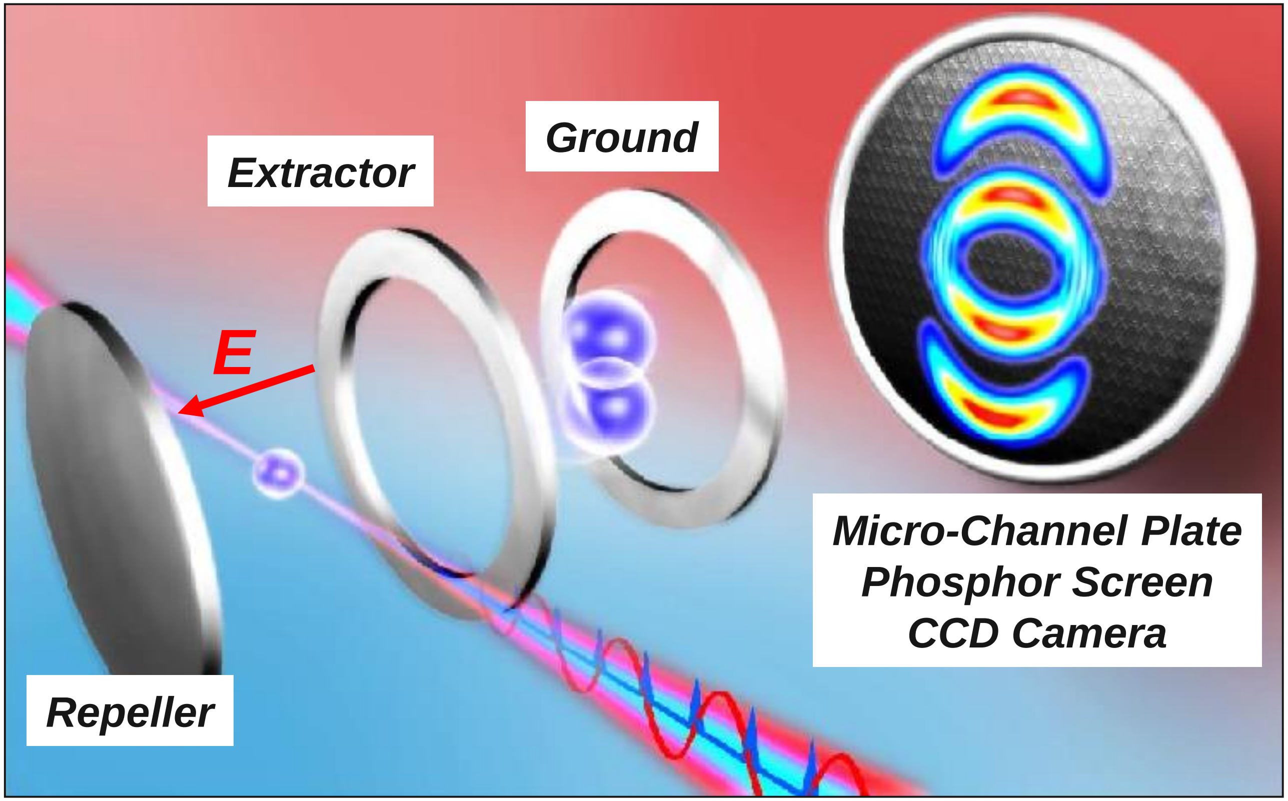


Magnetic Bottle Electron Spectrometer

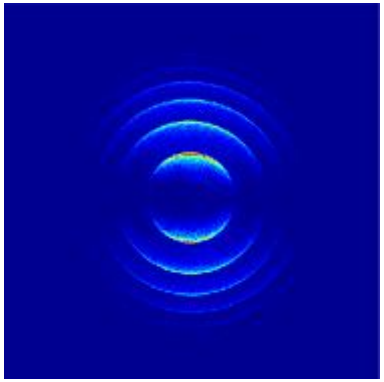
photoionisation in a strong magnetic field (1T), reduced gradually towards the end of flight tube enables 2π collection of electrons

Velocity map imaging (VMI)

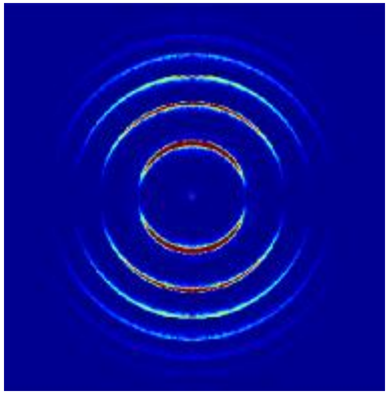
Eppink and Parker, Rev. Sci. Instr., 68, 3477 (1997)
Vrakking, Rev. Sci. Instrum., 72, 4084 (2001)



2D projection



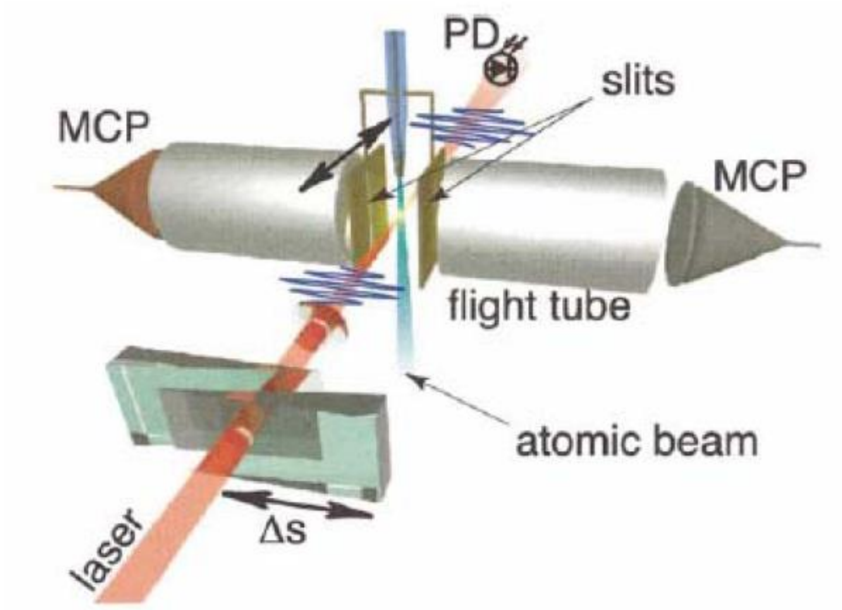
3D reconstruction



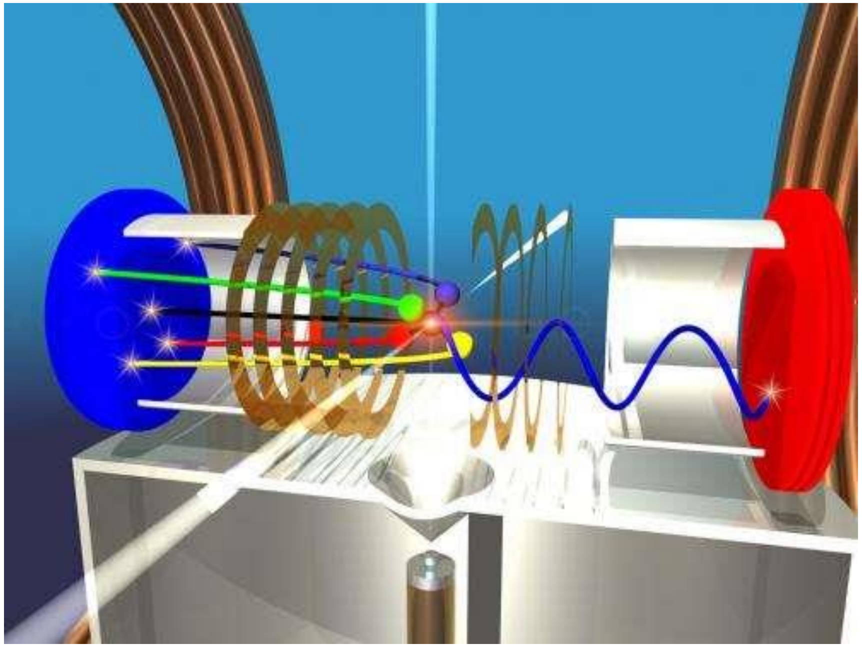
Detector captures the 2D projection of electron momentum distribution
+ assume symmetry around the E-field
Abel inverson → reconstruction of the full distribution

Stereo-machines

Coincidence measurements



Reaction Microscope / COLTRIMS



Temporal characterization

ultrashort laser pulses:

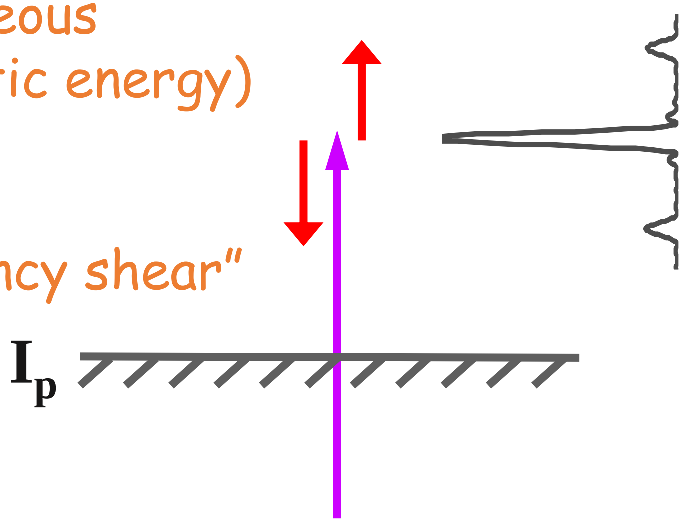
autocorrelations (second/third order)
SPIDER (spectral shear)
FROG (second/third order)

$\Bigg\}$ nonlinear effect

in the XUV regime???

photoionization can be considered an instantaneous process, conserving time-frequency (time-kinetic energy) properties

+ electron in the laser field undergoes „frequency shear"



in most cases we measure the electron/ion replicas

Temporal characterisation schemes

Autocorrelation

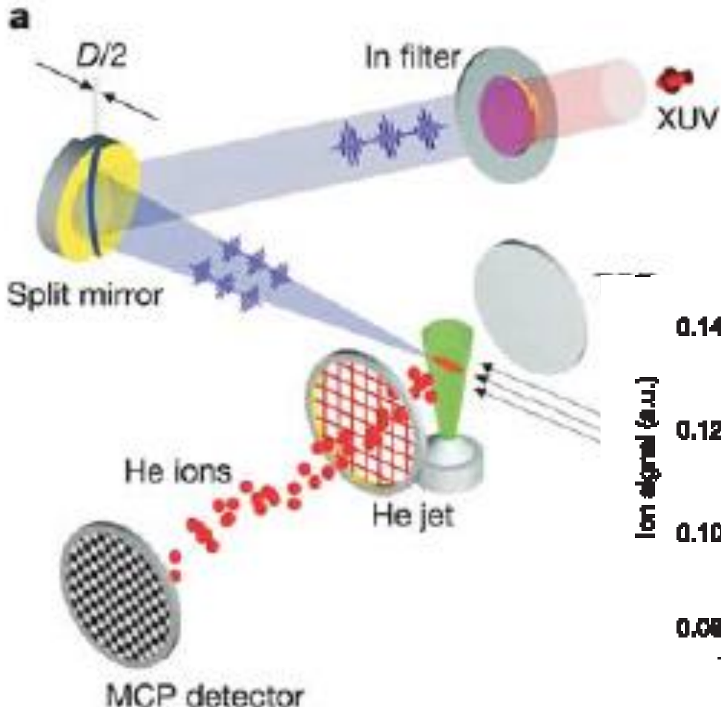
- 2nd order intensity volume autocorrelation (IVAC)

XUV SPIDER

Cross-correlation

- X-FROG
- RABITT
- Asec streak camera
- FROG - CRAB

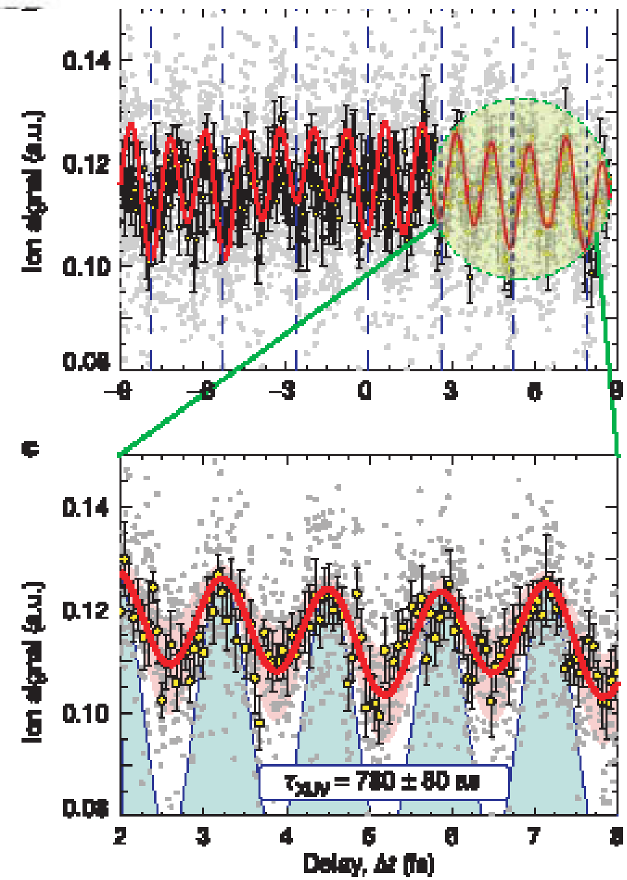
2nd order autocorrelation



Direct measurement of pulse duration

Requires:

- high XUV intensity
- nonlinear XUV detector (2-photon ionization of He)



Status:

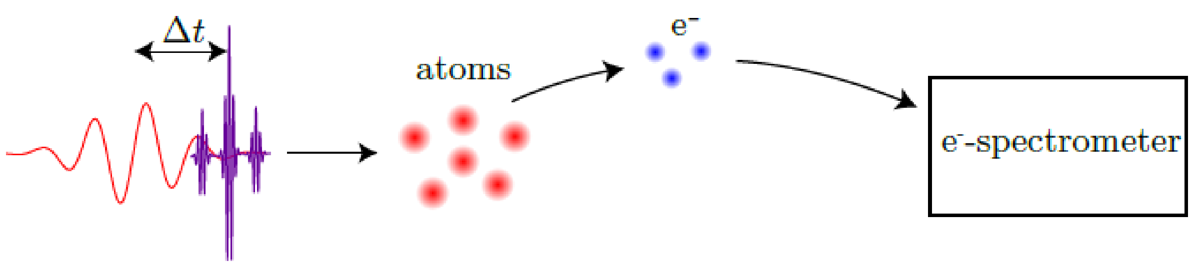
- spectral range: up till 30 eV
- pulse duration: 320 as

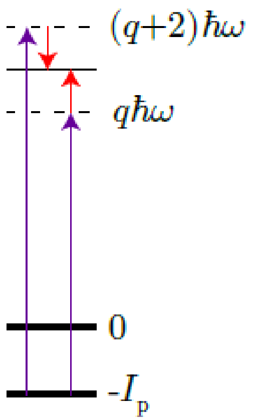
Tzallas: Nature, 426, 267 (2003)
Nabekawa: PRL 96, 083901 (2006)

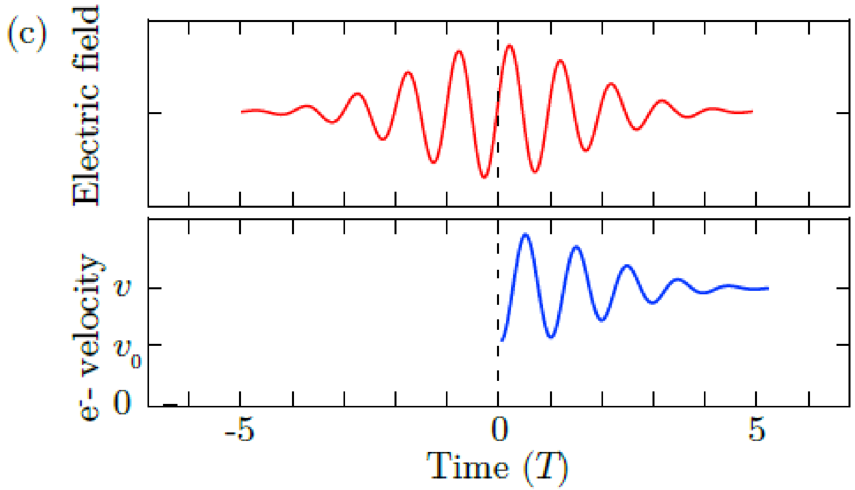
Cross-correlation

Low intensity of the high-harmonic radiation makes auto-correlation techniques less practical.

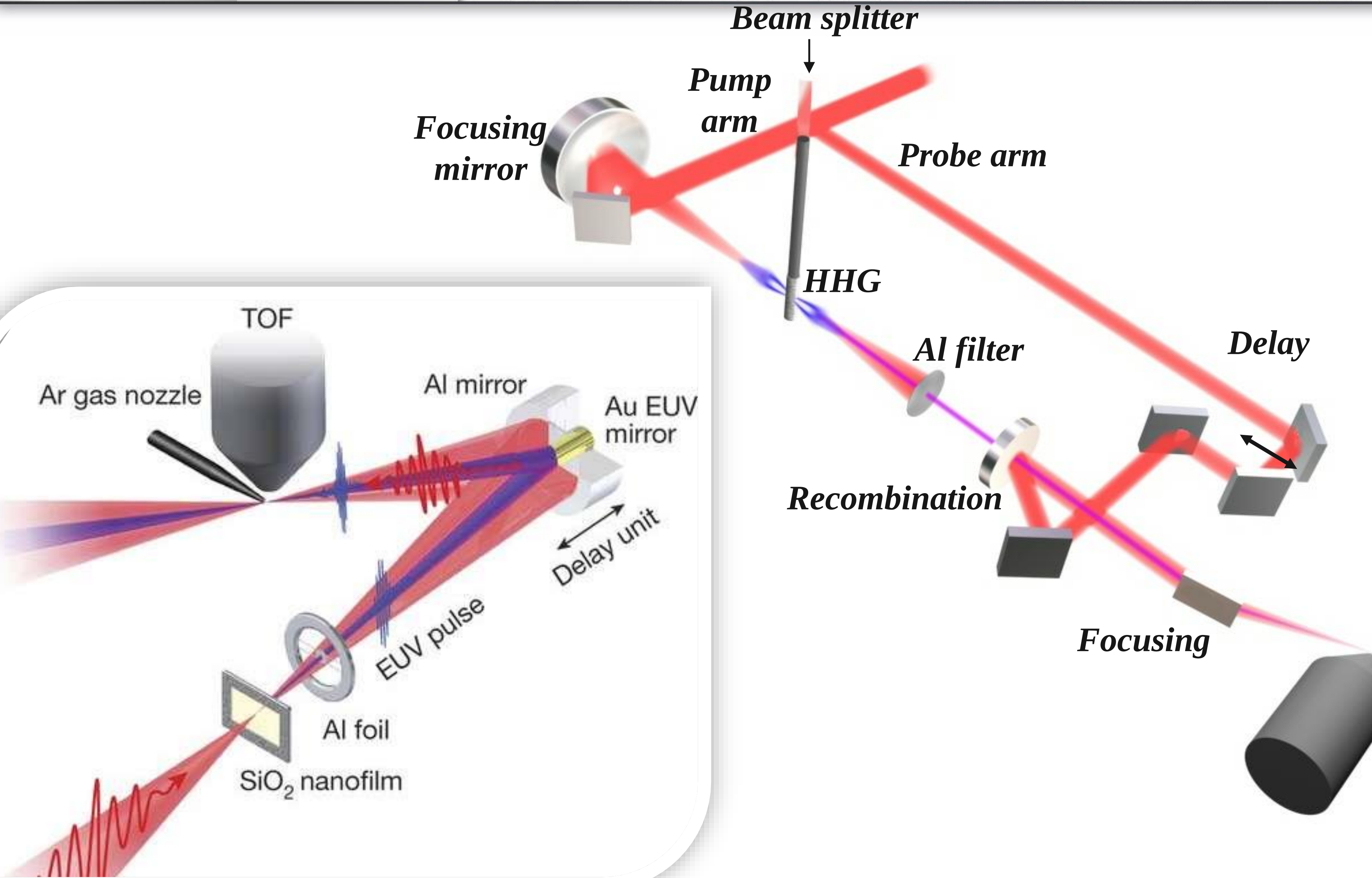
Scheme: XUV photoionization in the presence of the IR field



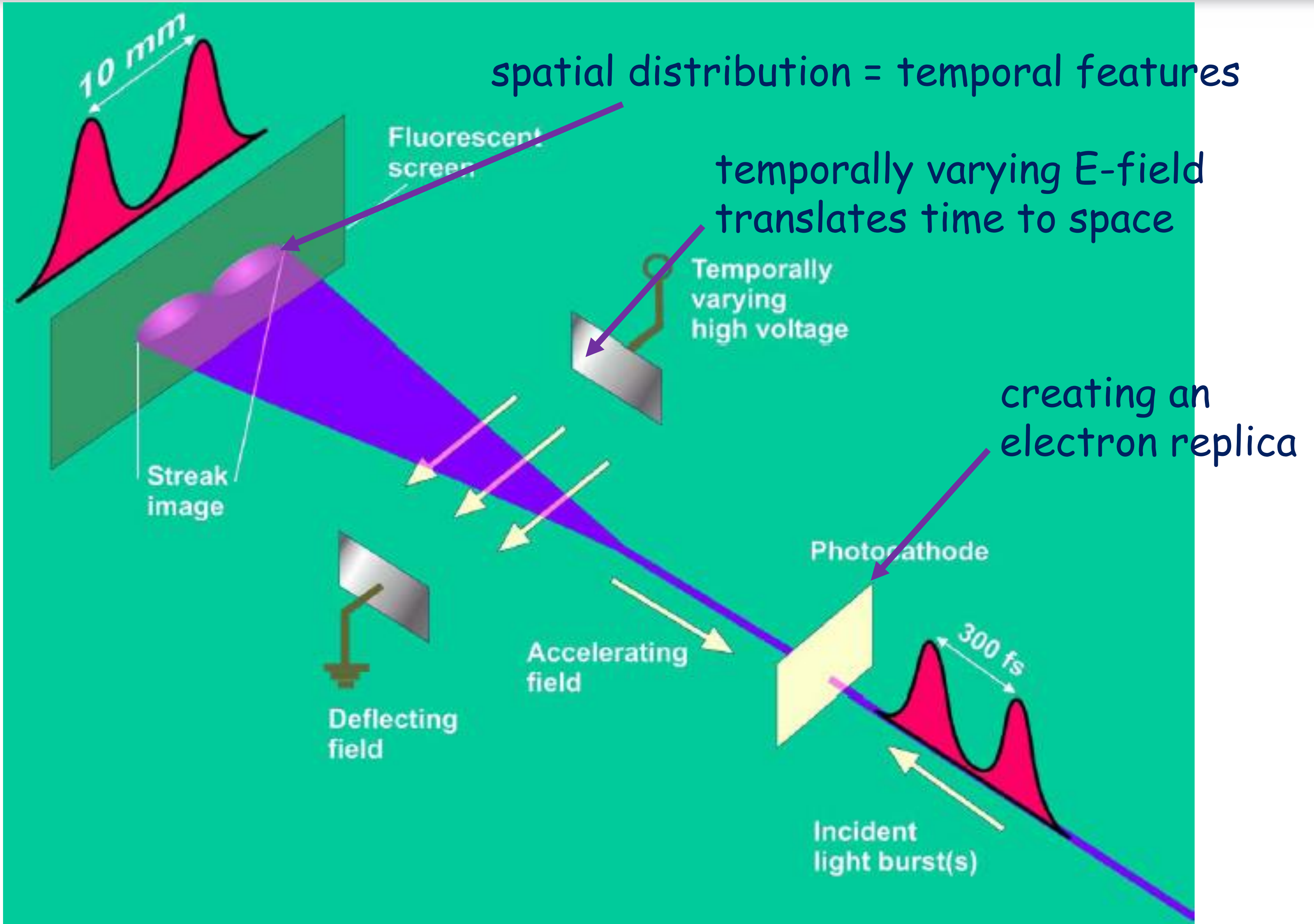




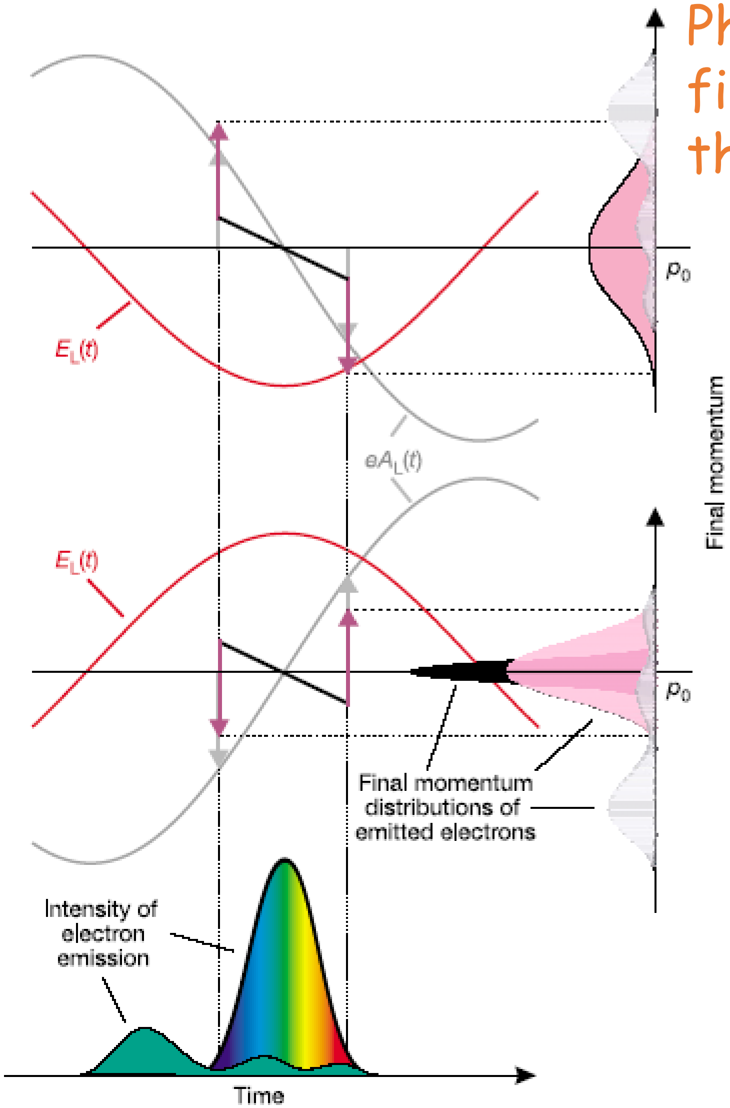
Experimental arrangement



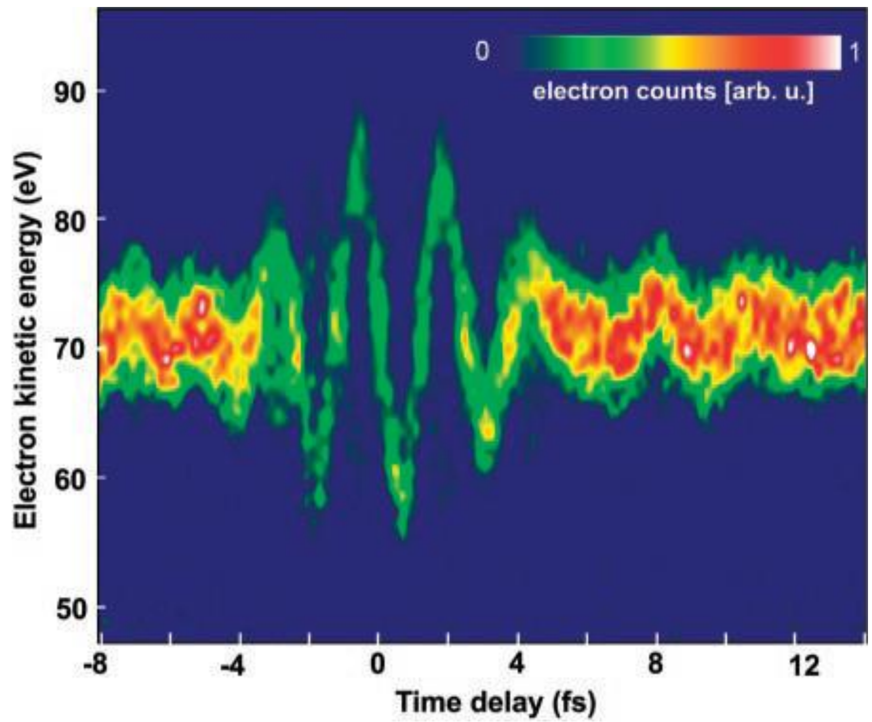
Conventional streak camera (ps)



Attosecond streak camera (strong IR)



Photoionization in the presence of the IR field:
the IR E-field provides the fast streaking



Drescher: Science 291, 1923 (2001)
Itatani: PRL 88, 173903 (2002)
Kitzler: PRL 88, 173904 (2002)
Gouliemakis: Science 305, 1267 (2004)

Creation of sidebands (weak IR)

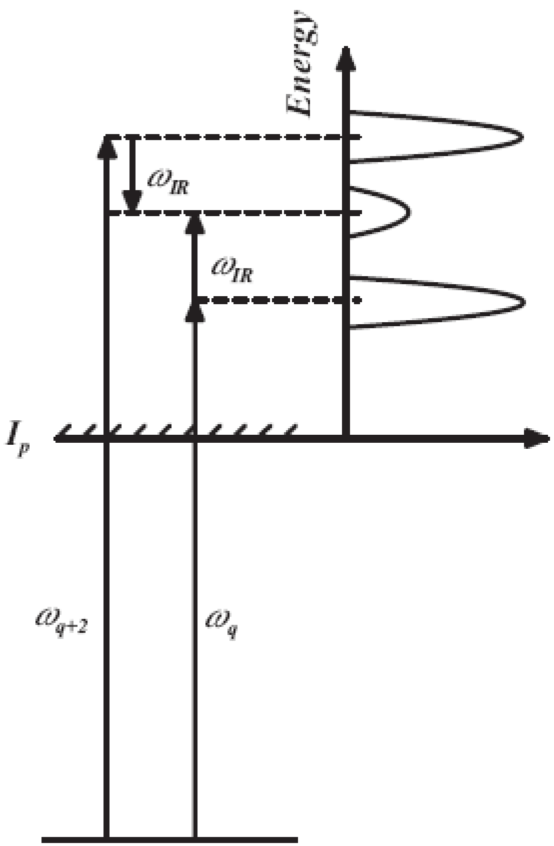
$$A_{SB}(\omega) = \int_{-\infty}^{\infty} dt\, e^{i(\omega - I_p/\hbar)t} \tilde{\varepsilon}_{XUV}(t)\, \varepsilon_{IR}(t-\Delta t) e^{i\Phi_{IR}(t-\Delta t)}$$

$$\tilde{\varepsilon}_{IR}(t) = \tilde{\varepsilon}_{IR}^{+}(t) + \tilde{\varepsilon}_{IR}^{-}(t),$$

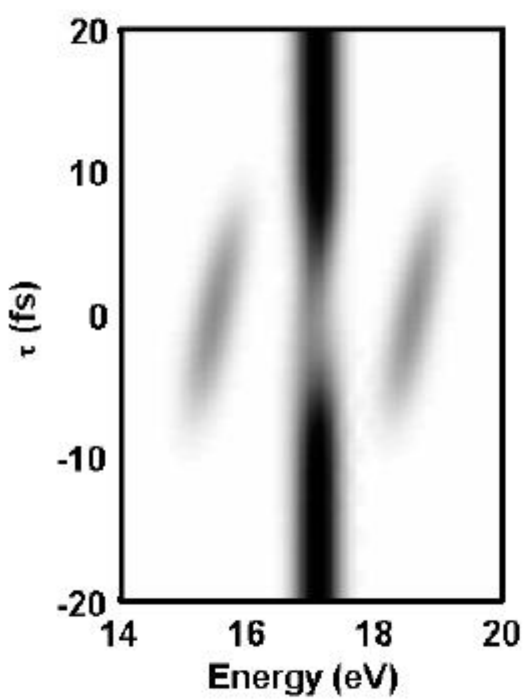
$$\tilde{\varepsilon}_{IR}^{+}(t) = \tilde{A}_{IR}(t)\, e^{-i\omega_0 t},$$

$$\tilde{\varepsilon}_{IR}^{-}(t) = \tilde{A}_{IR}^{*}(t)\, e^{+i\omega_0 t}.$$

absorption, emission

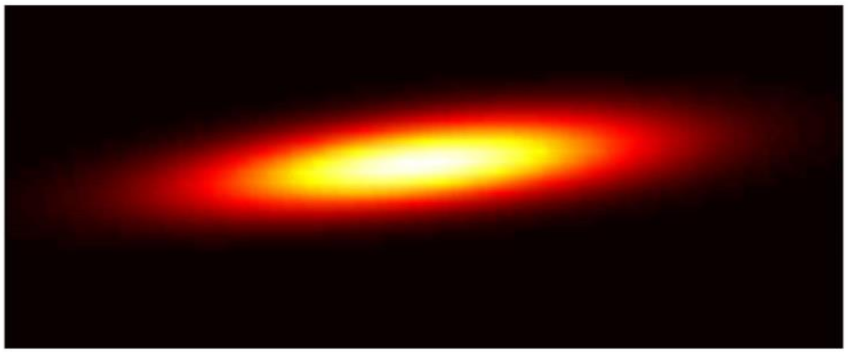


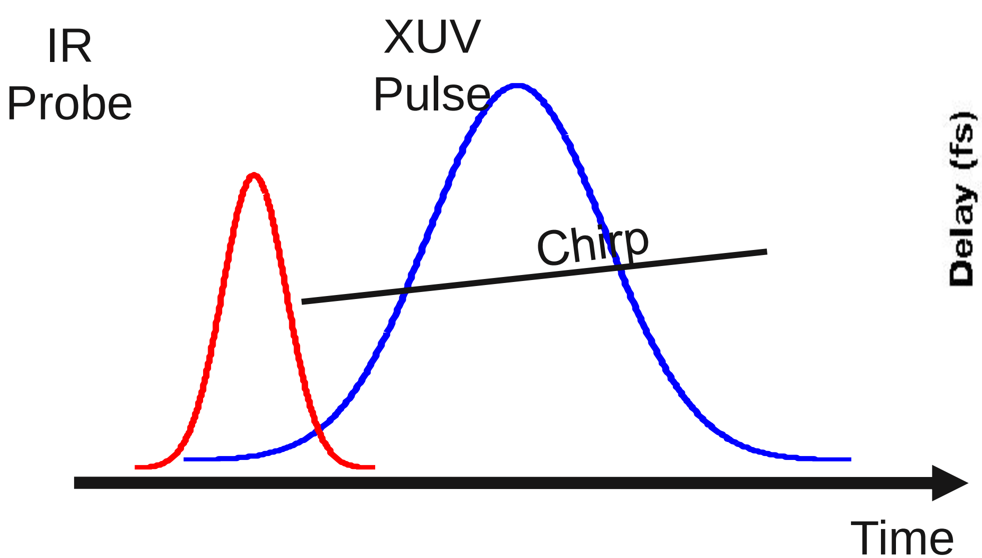
$$\tau_{XUV}^2 = \tau_{SB}^2 - \tau_{IR}^2$$

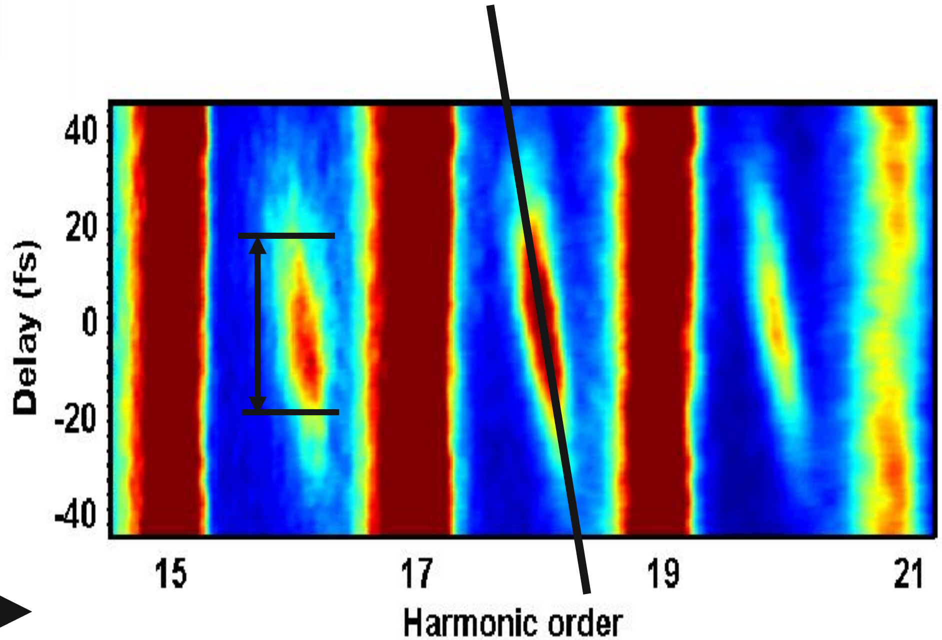


Femtosecond characteristics: XFROG

MAURITSSON: Phys. Rev. A. 70 021801R (2004)







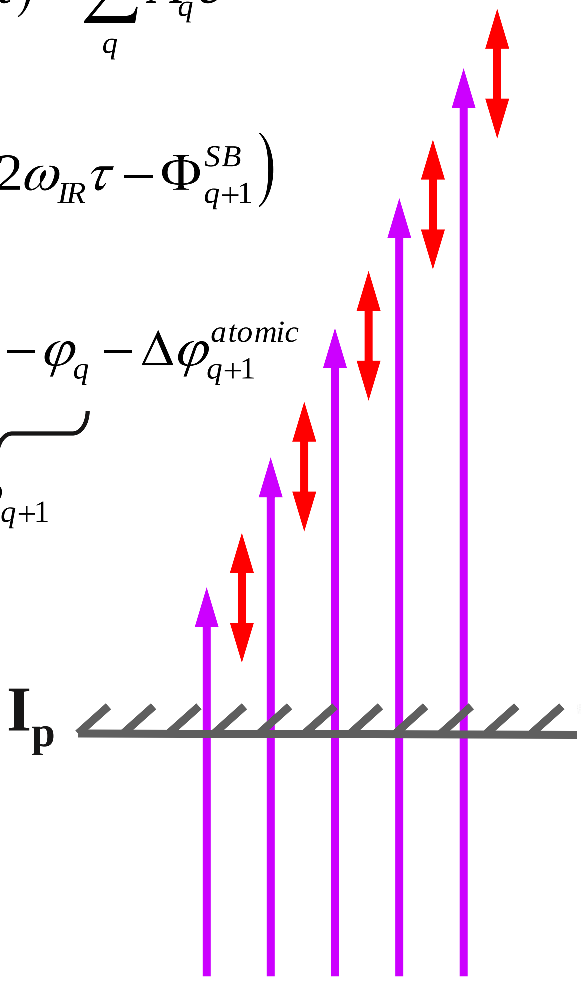
Attosecond characteristics: RABITT

Reconstruction of Attosecond Beating by Interference of Two-photon Transitions (RABITT)

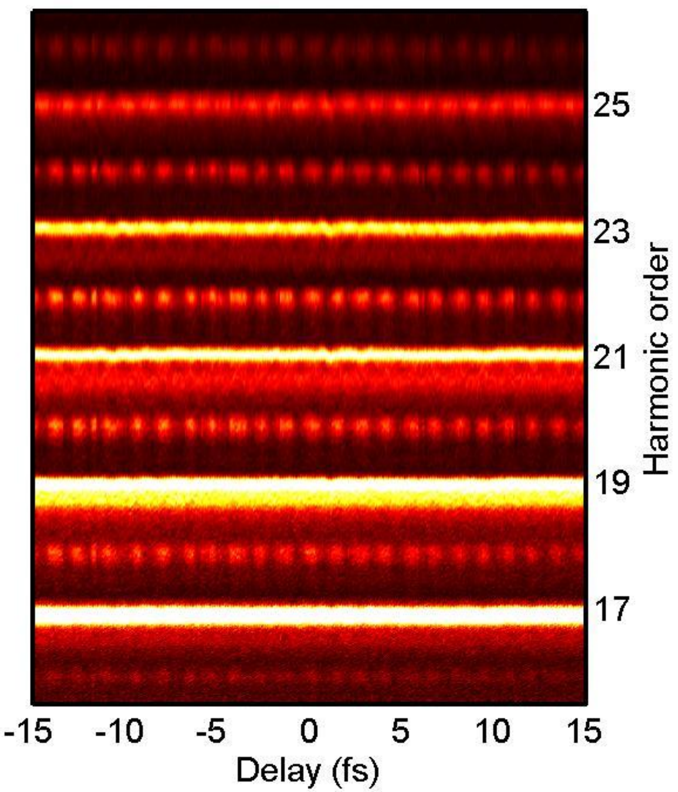
Harmonic field: $E(t) = \sum_q A_q e^{i\varphi_q}$

Sidebands: $I_{q+1}^{SB} \propto \cos\left(2\omega_{IR}\tau - \Phi_{q+1}^{SB}\right)$

phase-diff: $\Phi_{q+1}^{SB} = \underbrace{\varphi_{q+2} - \varphi_q}_{\Delta\varphi_{q+1}} - \Delta\varphi_{q+1}^{atomic}$



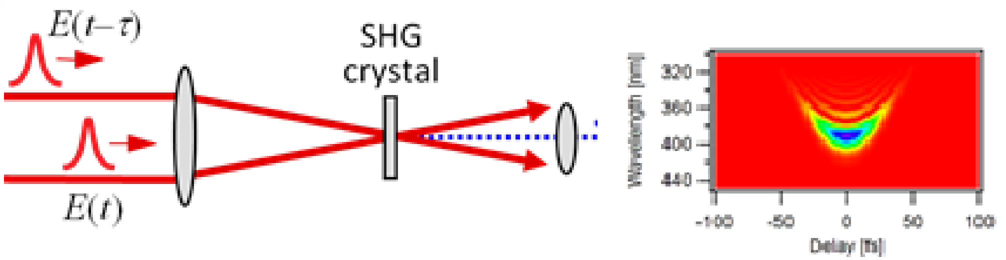
Photoelectron spectrum



Paul: Science, 292, 1689 (2001)
Muller: Appl. Phys. B74, S17 (2002)
Mairesse: Science 302, 1540 (2003)

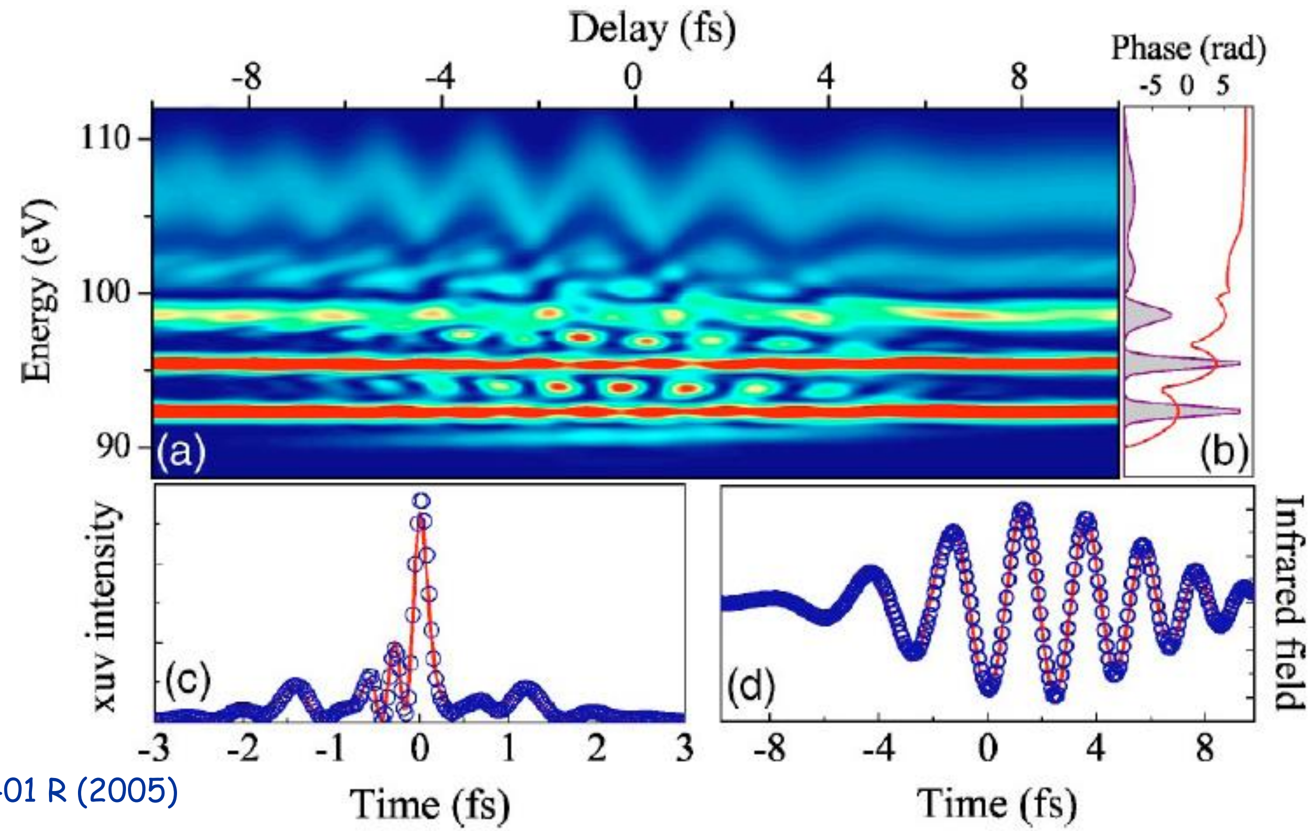
FROG CRAB

Frequency Resolved Optical Gating for Complete Resolution of Attosecond Bursts



FROG:
- gate pulse
- delay resolved spectrogram
- iterative reconstruction proc.

FROG CRAB
- gated by the generating IR pulse
- reconstruction of both IR and XUV pulses



Mairesse: PRA 71, 011401 R (2005)

FROG CRAB - examples

isolated attosecond burst

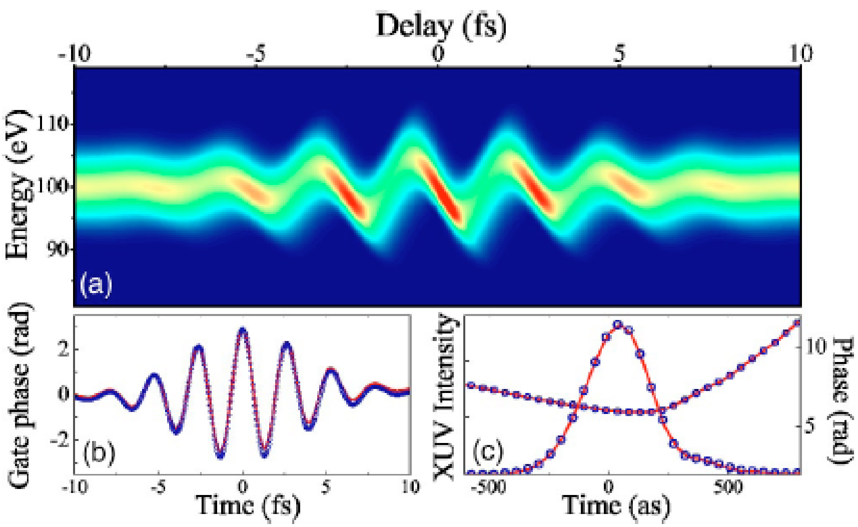


FIG. 1. (a) CRAB trace of a single 315 as pulse [full width at half maximum (FWHM) of intensity], having second- and third-order spectral phases (Fourier limit=250 as), gated by a Fourier-limited 6-fs 800 nm laser pulse, of 0.5 TW/cm^2 peak intensity. The electrons are collected around $\theta=0$ with an acceptance angle of $\pm30°$. (b), (c) A comparison of the exact as pulse and the laser-induced gate phase $\phi(t)$ (full line) with the corresponding reconstructions (dots) obtained from the CRAB trace after 100 iterations of the PCGPA algorithm [20]. The gate modulus $|G(t)|$ is constant and equals to 1.

attosecond pulse train

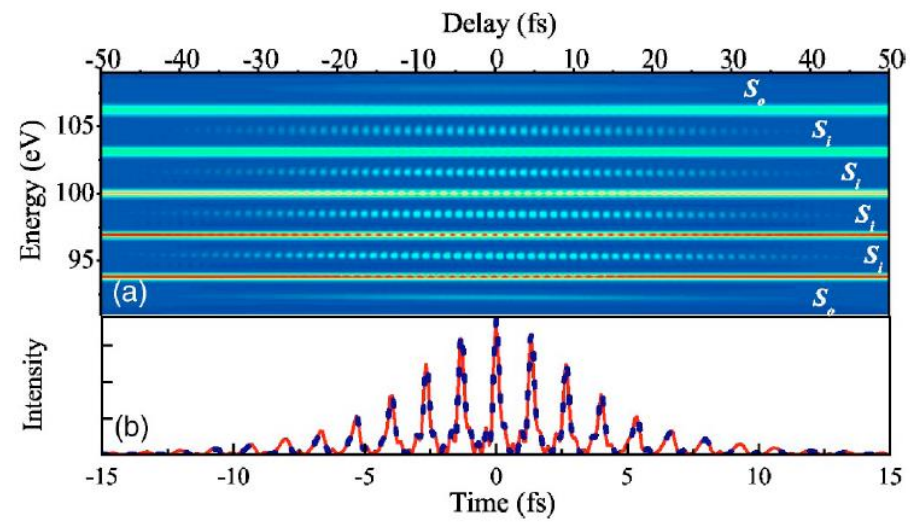
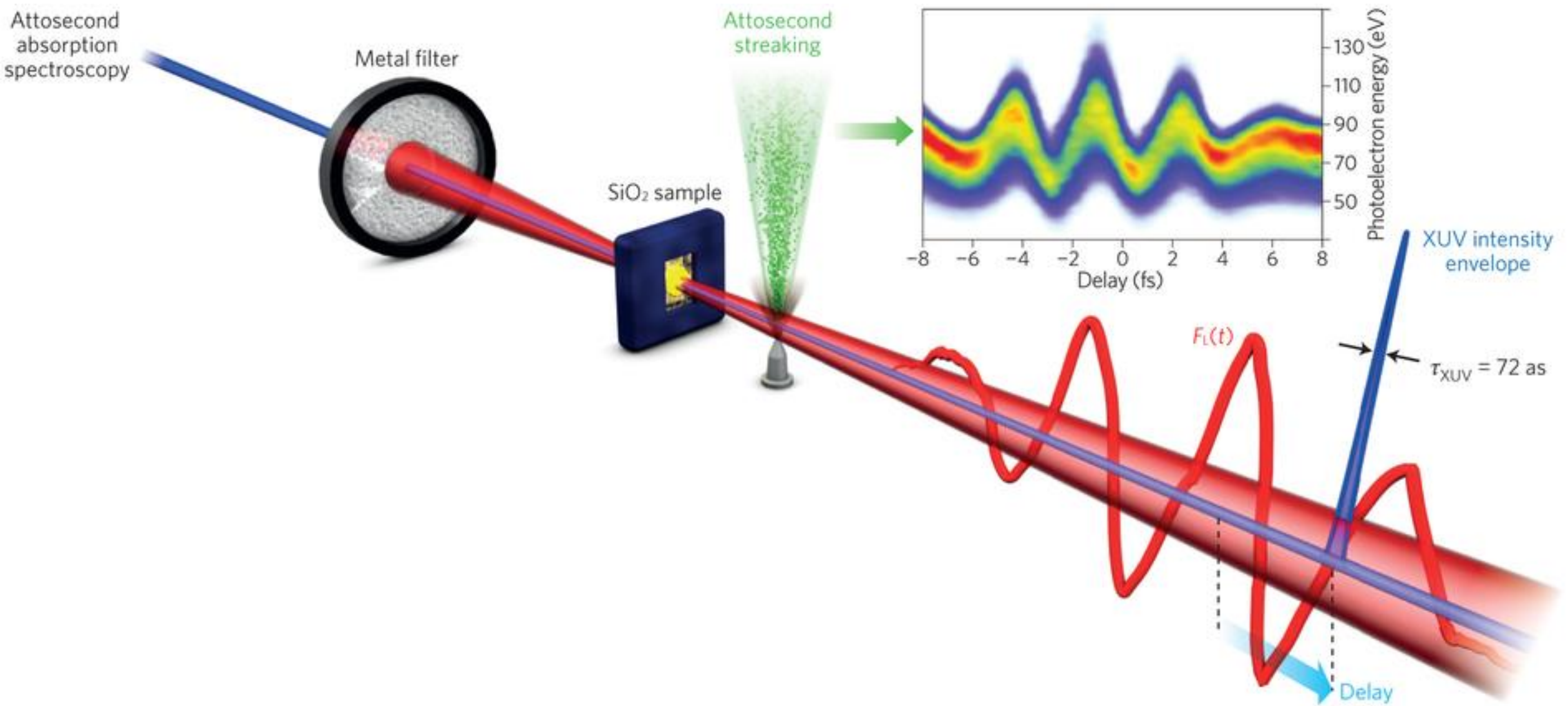


FIG. 2. (a) CRAB trace at $\theta=0$ of a 12-fs-train of nonidentical as pulses, of period $T/2=1.3$ fs, gated by a 30-fs-800 nm- ($T=2.6$ fs) laser pulse, of 0.05 TW/cm^2 peak intensity, assuming a spectrometer resolution of 100 meV. The as pulses are shorter in the center of the train ($\approx$250 as), than in the edges ($\approx$400 as). The outer and inner sidebands are respectively labelled S_o and S_i. (b) A comparison of the exact as train (red line) and the reconstruction (dotted blue line) obtained from the CRAB trace after 750 iterations of the PCGPA algorithm.

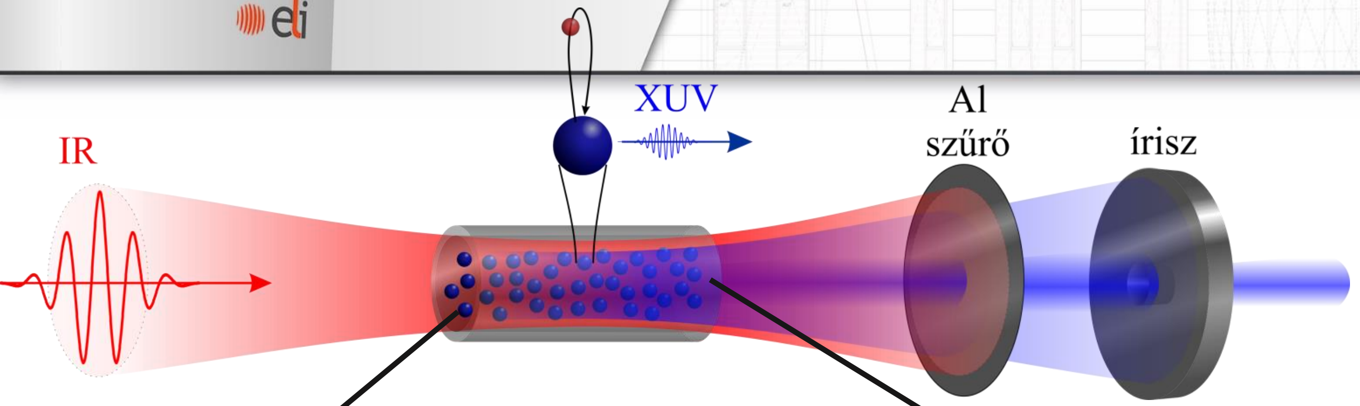
Mairesse: PRA 71, 011401 R (2005)

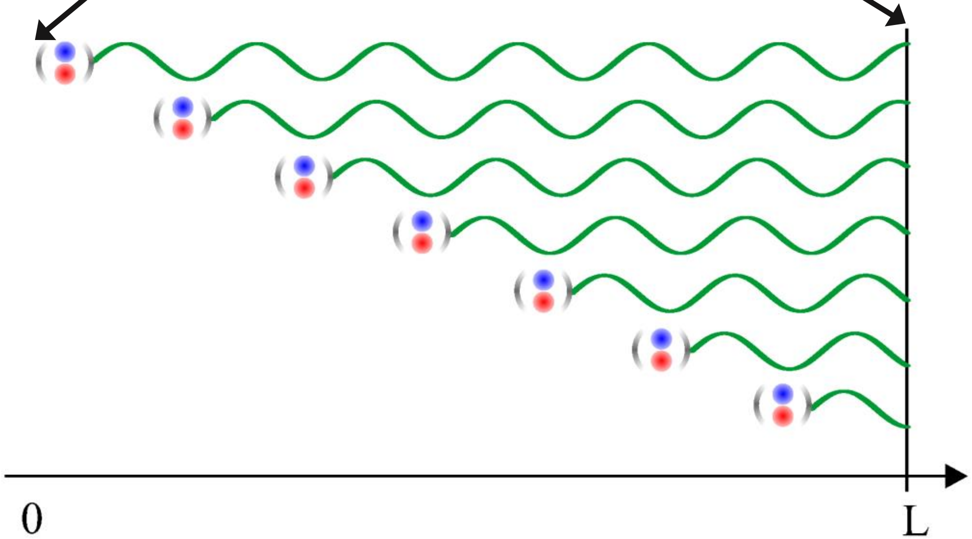
On-line characterisation



Contents

The role of phase-matching

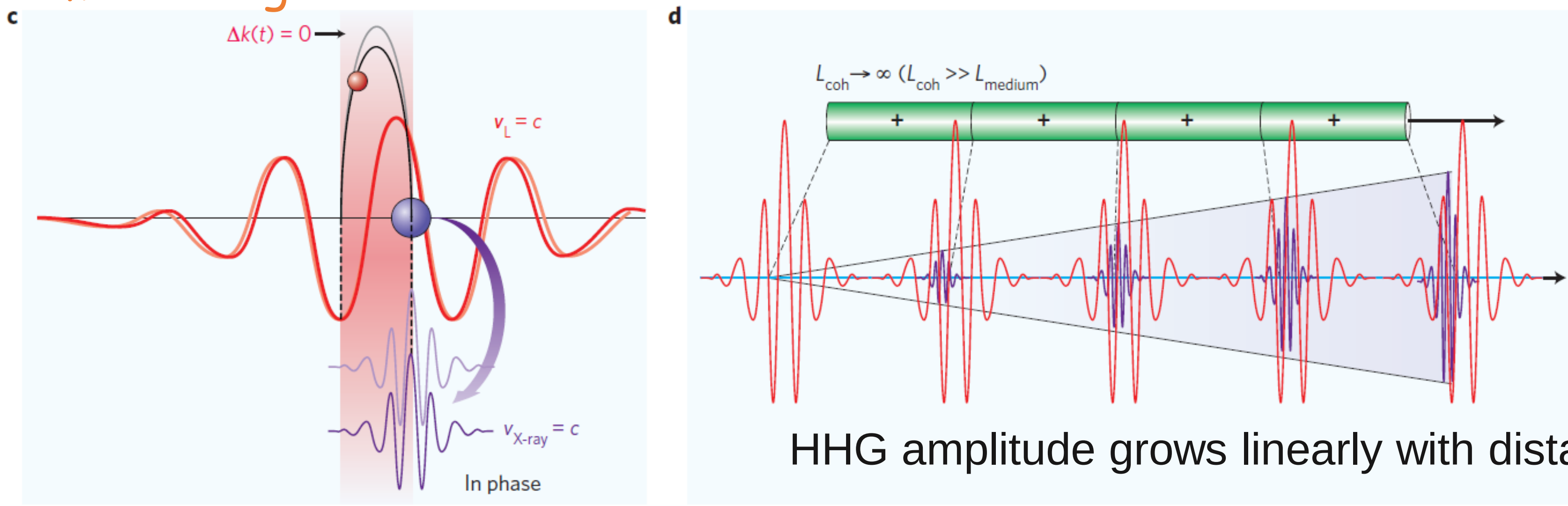




the generated elementary waves propagate in the medium, and we observe their superposition

if the generating field and the generated component has a different velocity, the components add with a spatially changing phase: the harmonic signal oscillates with medium thickness

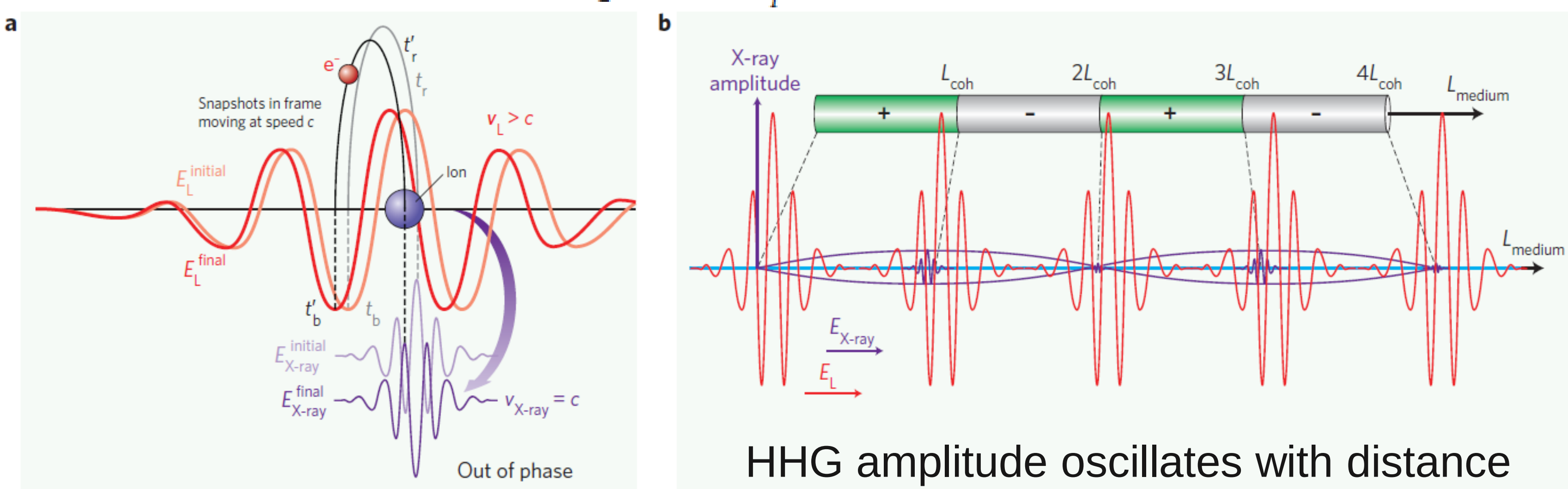
Phase-matched generation



HHG amplitude grows linearly with distance

Non-phase-matched generation

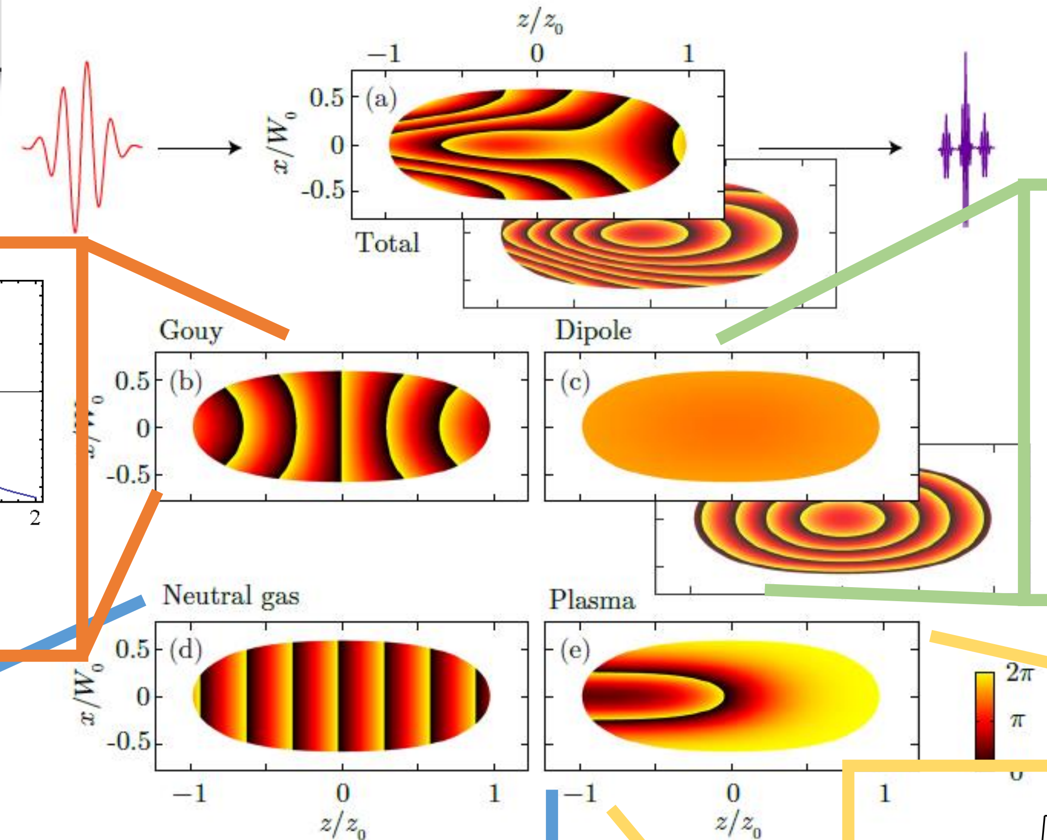
$$L_{coh}(q) = \pi/\Delta k_q$$

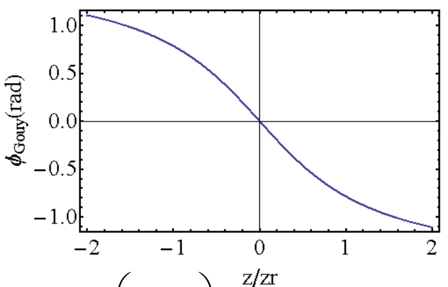


HHG amplitude oscillates with distance

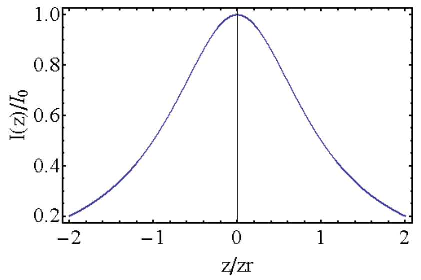
Popmintchev: Nphot 4, 822 (2010)

Components of phase-mismatch





$$arctg\left(-\frac{z}{z_R}\right)$$



$$\varphi_d = -\alpha\, I$$

$$\sqrt{1-\frac{\omega_p^2}{\omega^2}} \cong 1-\frac{1}{2}\frac{e^2 N_e}{\omega^2 \varepsilon_0 m_e}$$

Plasma contribution always < 0

Scales as λ^2

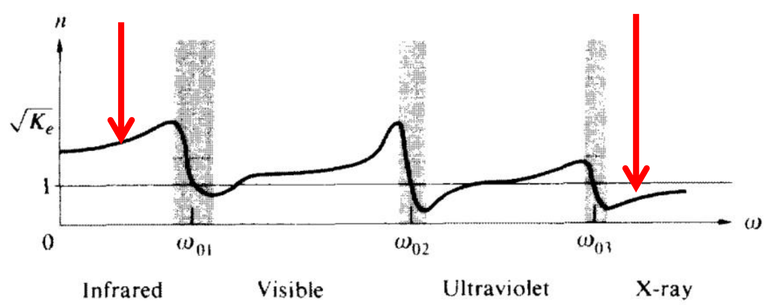
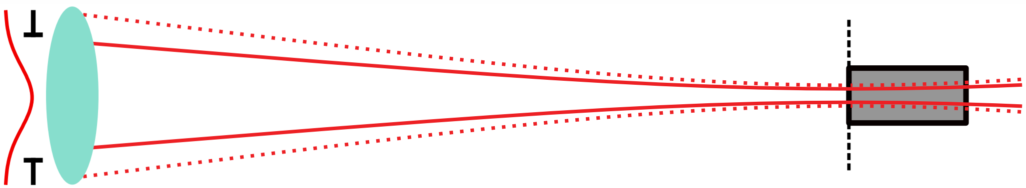


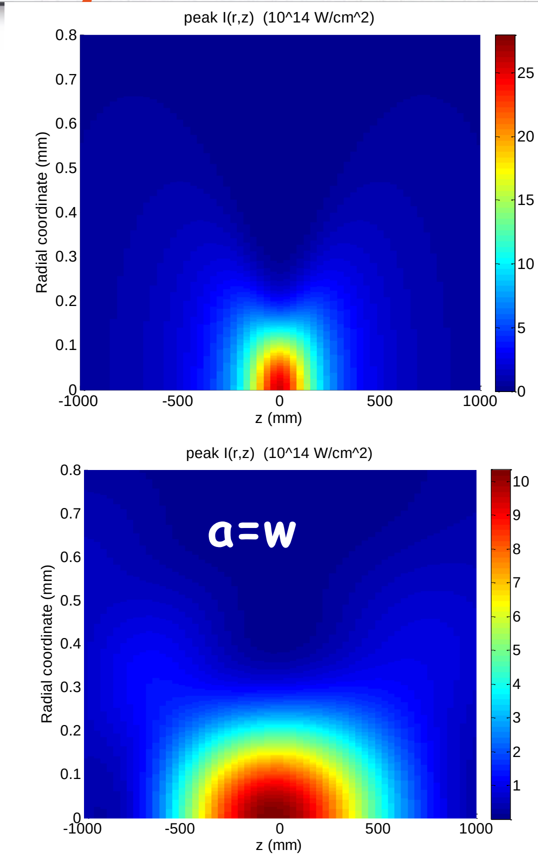
Figure 3.41 Refractive index versus frequency.

Hecht, Optics (2002)

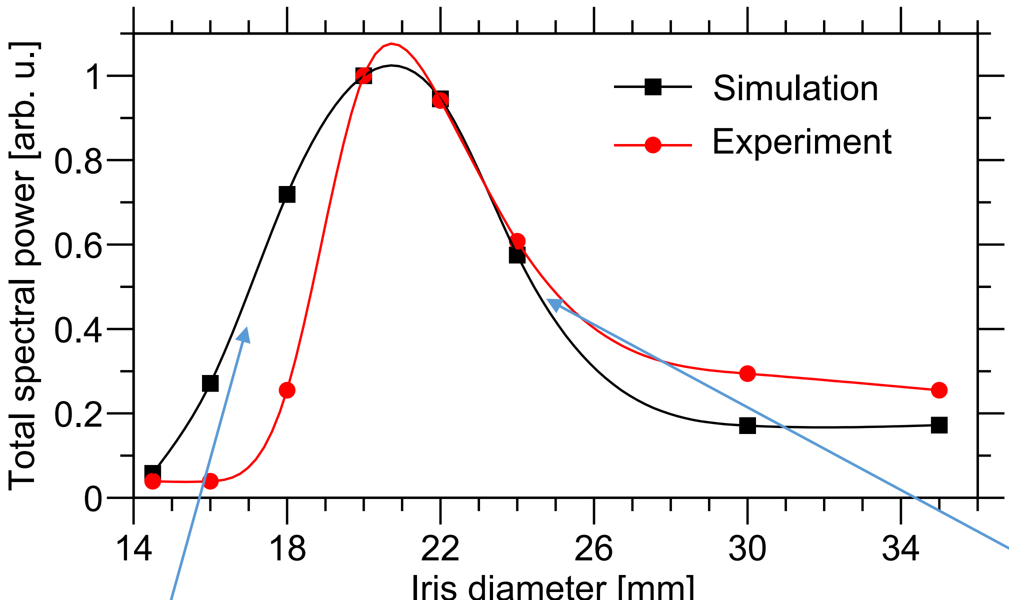
Ch Heyl, PhD dissertation

Phase-matching in the laboratory: via aperturing the laser beam





With 86% of the energy the peak intensity is only 40% of the untruncated value



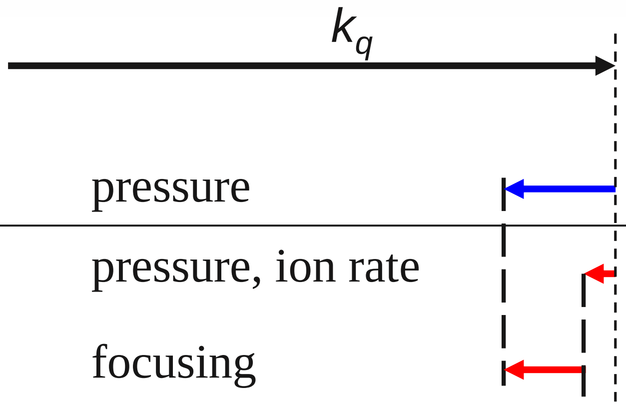
Closing the aperture the pulse energy is decreased and the Rayleigh range is increased, finely tuning phase-matching conditions.

+ focal spot size increases, ie higher number of photons contribute to HHG.

When the iris is too small, the intensity in the focus falls below the HHG threshold.

Pressure-tuned phase-matching

Phase velocity c for harmonic q



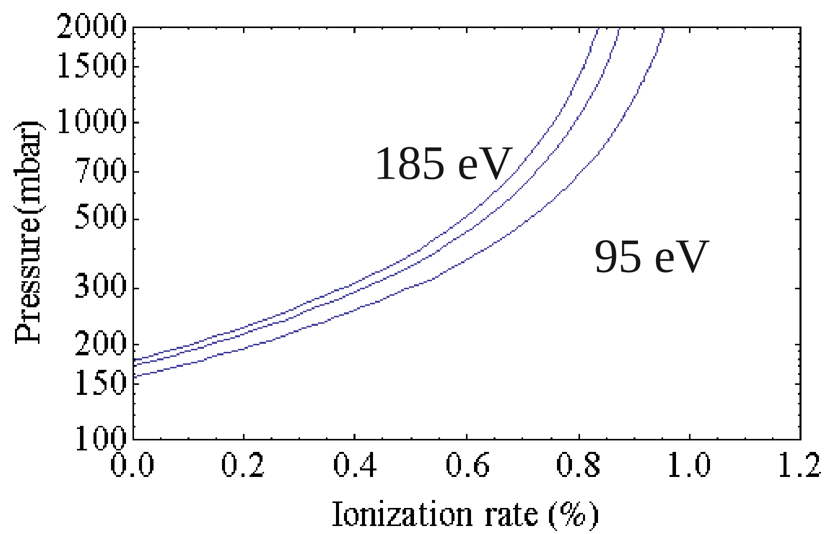
- neutral dispersion for the XUV — pressure
- neutral + plasma dispersion for the IR — pressure, ion rate
- Gouy phase-shift — focusing

For a given focusing geometry and ionization rate, there is an optimal pressure:

$$p_{match} = \frac{-\Delta k_G}{\boxed{\frac{\partial \Delta k_n}{\partial p} + \frac{\partial \Delta k_p}{\partial p}}}$$

Has to be positive!

Critical ionization rate: $\Delta k_n = \Delta k_p$



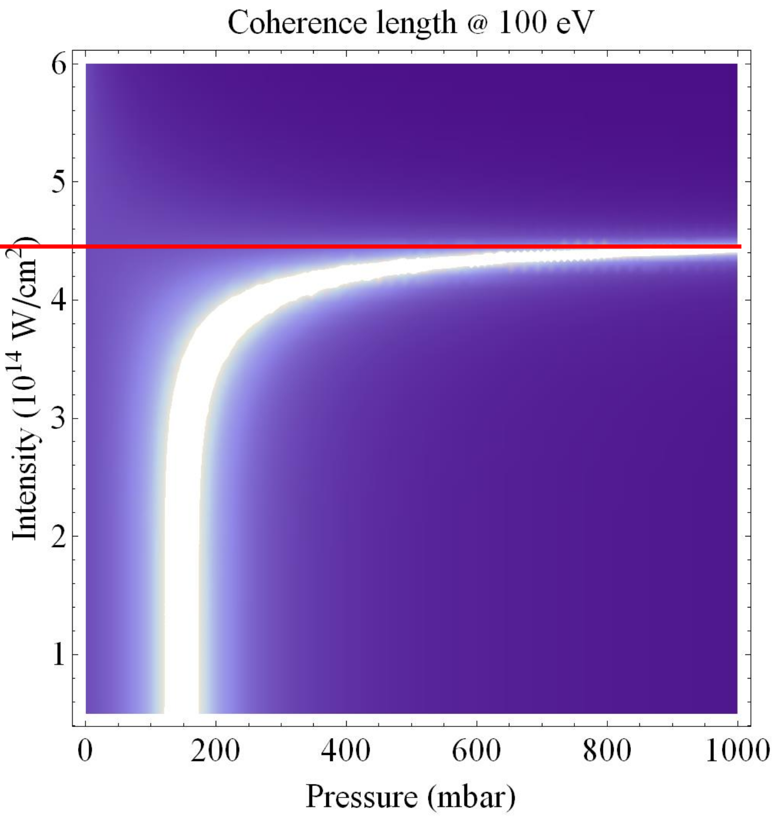
Balogh E, PhD dissertation

Pressure-tuned phase-matching

Using the ionization rate at the peak of the pulse (i.e. where cutoff harmonics are generated)

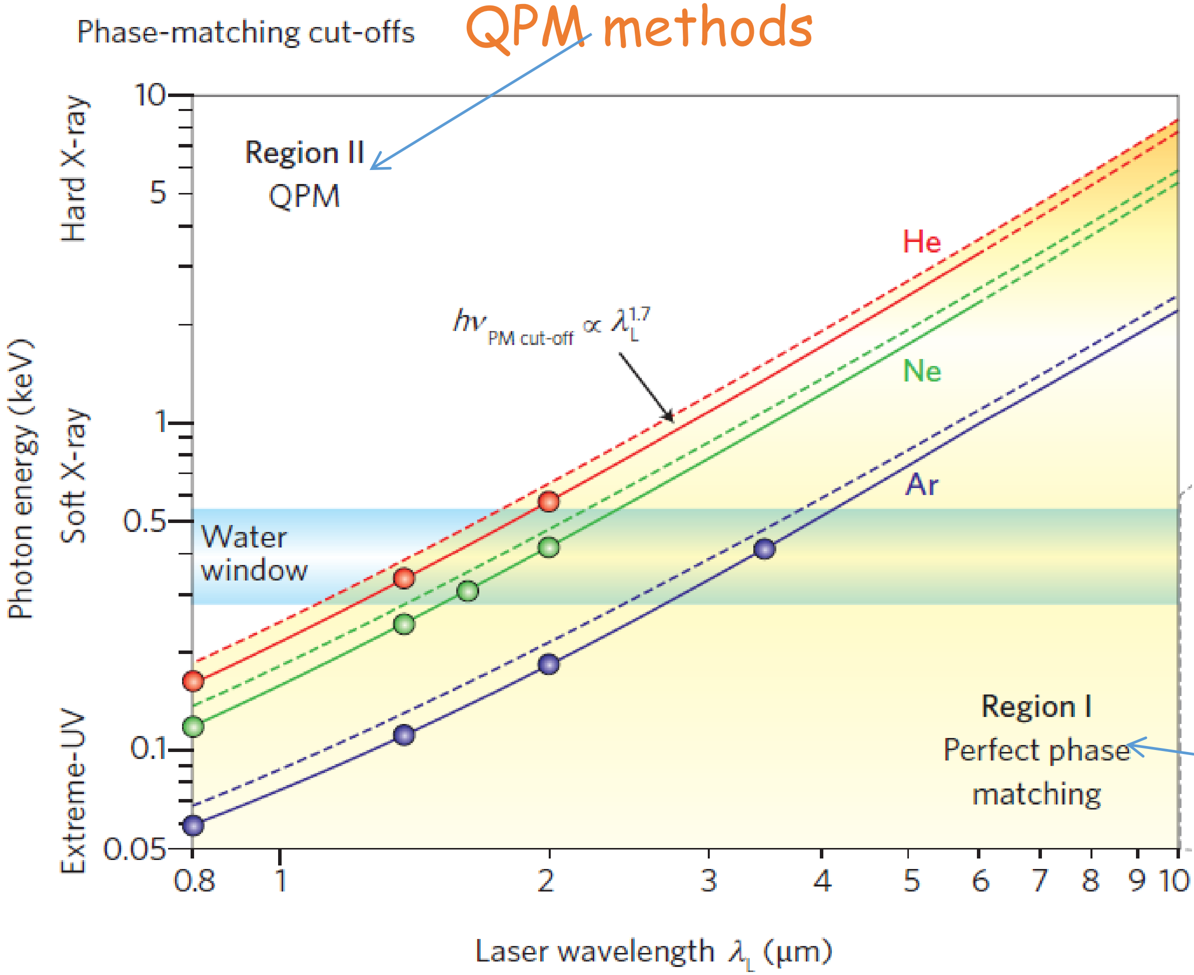
Coherence length as a function of pressure and intensity

At this intensity the cutoff is at 105 eV:
highest achievable photon energy is limited



Pressure-tuned phase-matching

phase-matching not possible:
QPM methods



phase-matching cutoffs for an 800 nm driver field (8 cycles):

in Ar 60 eV H39
in Ne 110 eV H71
in He 180 eV H117

phase-matching possible with pressure-tuning

Popmintchev: Nphot 4, 822 (2010)

Macroscopic effects

- Phasematching
  - selective for short or long trajectories
  - determines divergence of harmonics
  - determines spatial distribution of harmonics (collimated short trajectories, annular long trajectories)
  - limits optimal cell length and position
- Self-focusing and plasma defocusing:
  - balance between them may cause self-guiding (filamentation)
  - limits maximum pulse intensity (setting of "working intensity")
- Self-phase modulation:
  - broadens spectra of harmonics
  - affects the harmonic chirp

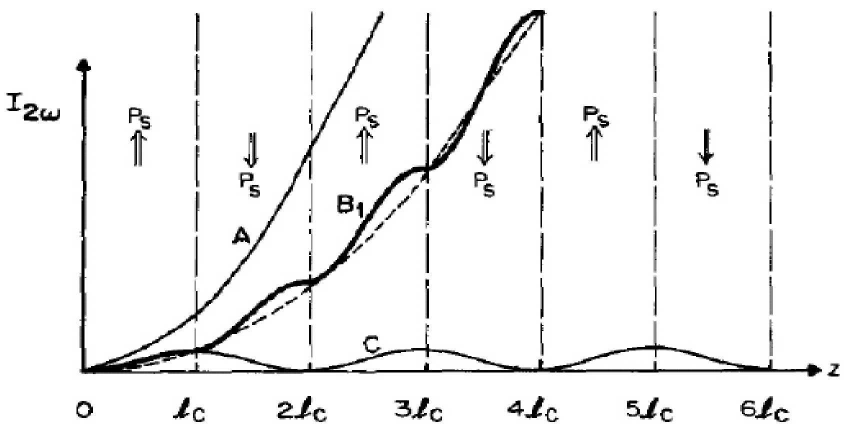
When phase-matching is not possible: Quasi phase-matching

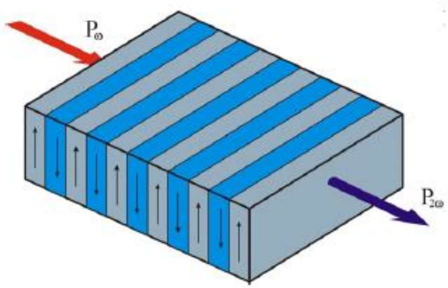
PHYSICAL REVIEW VOLUME 127, NUMBER 6 SEPTEMBER 15, 1962

Interactions between Light Waves in a Nonlinear Dielectric*

J. A. Armstrong, N. Bloembergen, J. Ducuing,† and P. S. Pershan
Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts

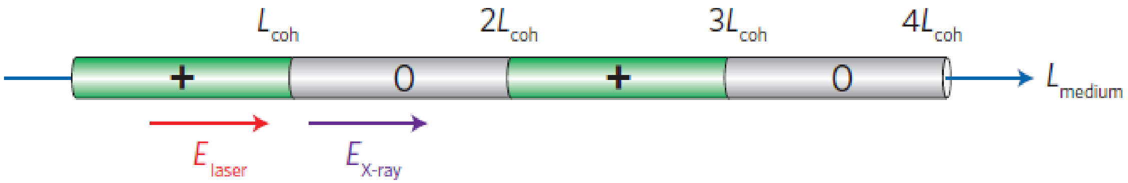
(Received April 16, 1962)





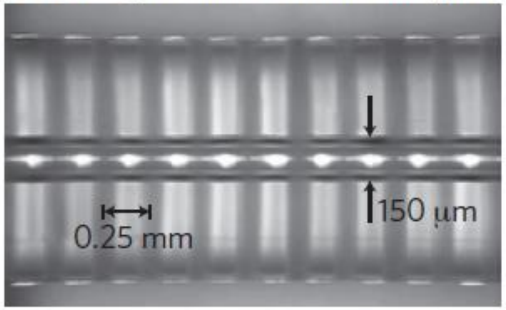
QPM techniques in HHG

to periodically switch off HHG in destructive zones

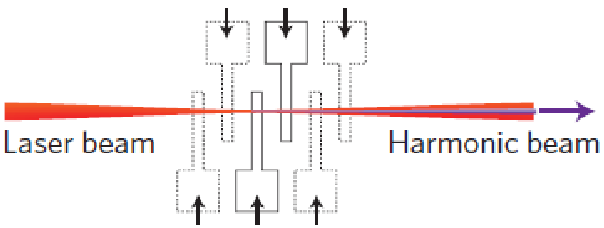


a

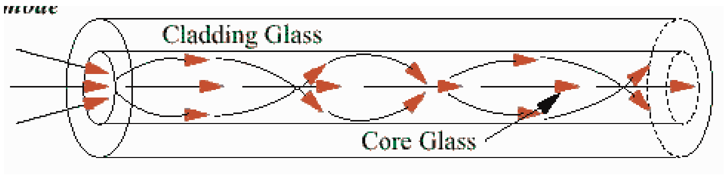
Periodically modulated hollow waveguide



Successive gas jets

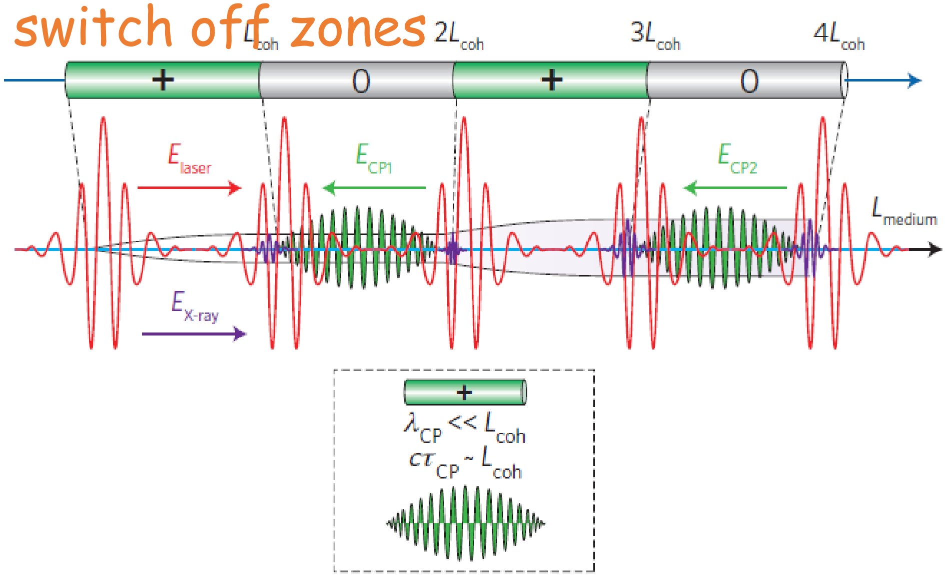


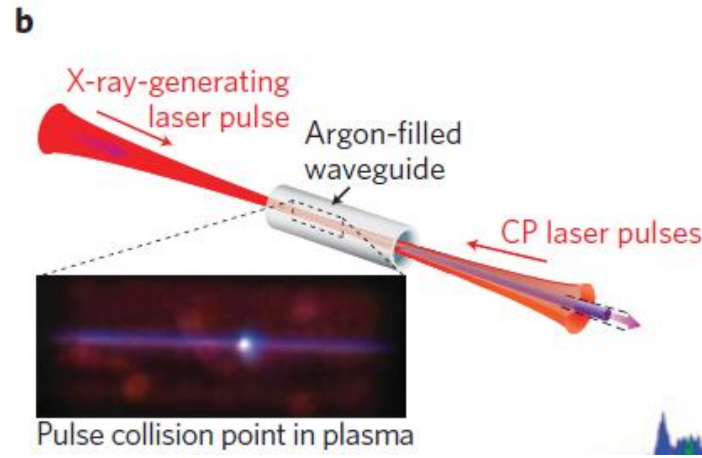
multimode waveguide



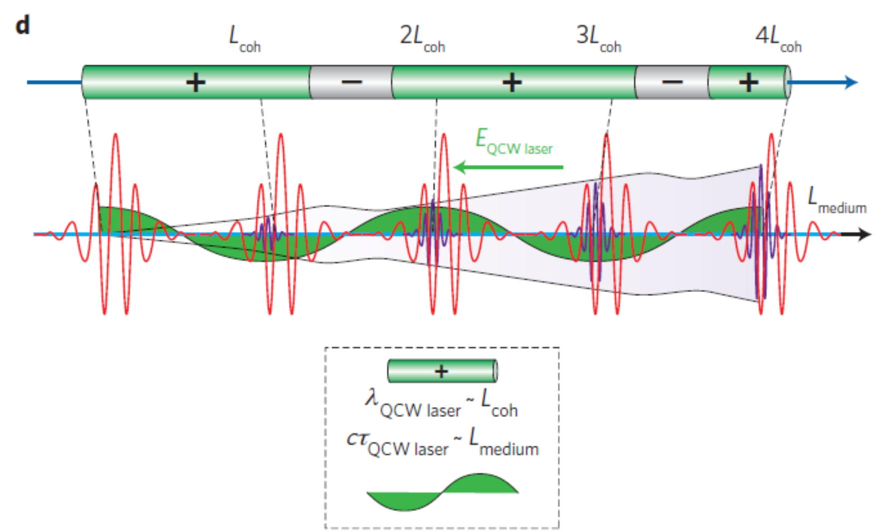
QPM techniques in HHG

counter-propagating pulses scramble the phase of high-harmonic emission to switch off zones





counter-propagating or perpendicularly propagating quasi-CW laser shifts the phase of the emission to increase constructive zones



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