

Generation of ultrashort XUV femtosecond to attosecond pulses

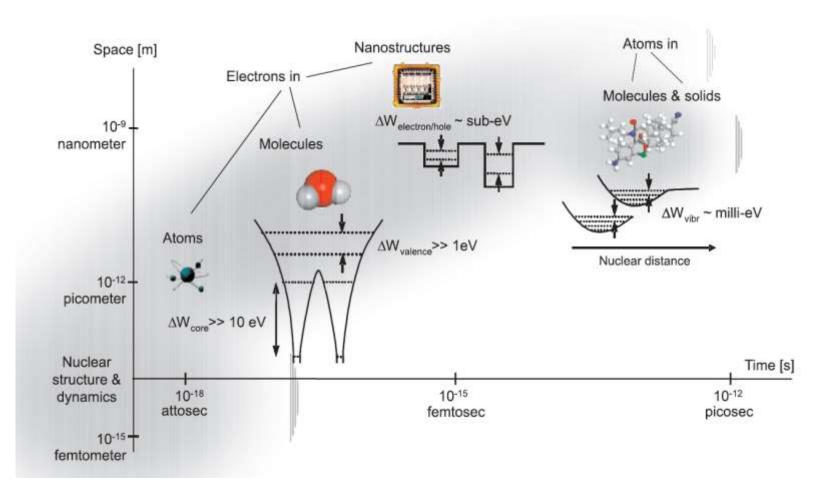
Katalin Varjú ELI-ALPS

2nd MOLIM Training School 6 - 10 March, 2017 Paris-Saclay



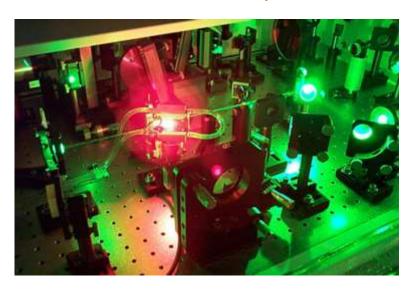






Fs light sources

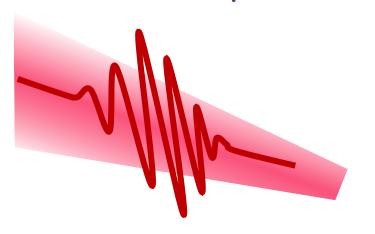
LASER mechanism \Rightarrow temporal and spatial coherence oscillator, phase locking, broad bandwidth gain medium \Rightarrow ultrashort pulse duration



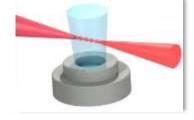
Can we build an XUV laser to transfer these properties to the XUV domain?

Mechanisms leading to fs-scale XUV generation

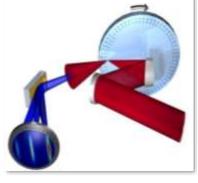
Intense laser pulse + nonlinear phenomenon



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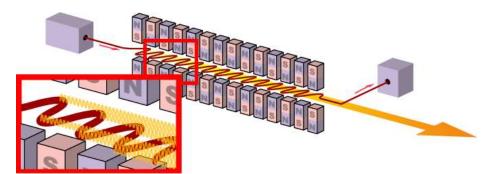


gas HHG



surface plasma HHG

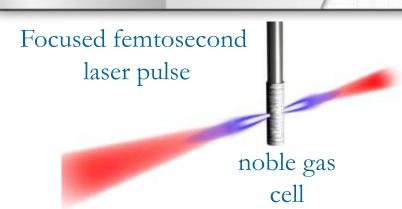
Accelerated e- based schemes



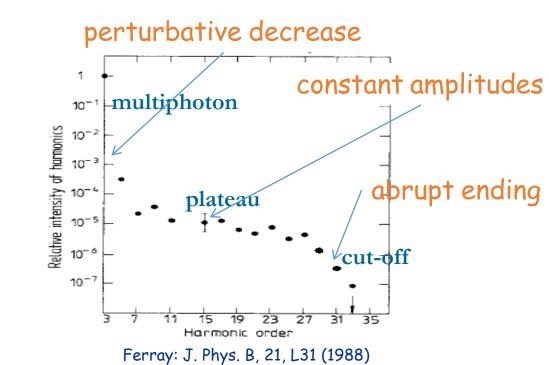
synchrotron, FEL, seeded FEL

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

Experimental observation of HHG



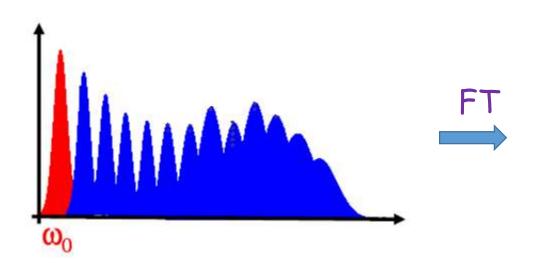
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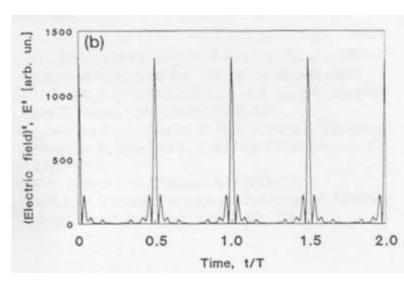


"Generating high order harmonics is experimentally simple."

Anne L'Huillier

The "birth" of attosecond science





Farkas, Phys. Lett. A (1992)

Atoms in a strong laser field

atomic electron:
$$E(r) = -\frac{1}{4\pi\varepsilon_0} \frac{e}{r^2}$$

$$r \approx 10^{-10} \mathrm{m}$$

$$E \approx 10^{11} \frac{\text{V}}{\text{m}}$$

intensity = |Poynting vektor|

$$I = 2 \cdot \frac{5 \text{ mJ}}{\pi \cdot (100 \mu\text{m})^2 \cdot 20 \text{ fs}} = 1.6 \times 10^{19} \frac{\text{W}}{m^2} = 1.6 \times 10^{15} \frac{\text{W}}{cm^2}$$

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

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

$$E_{\text{max}} = \sqrt{\frac{2 \cdot I}{\varepsilon_0 c}} = \sqrt{\frac{2 \cdot 1.6 \times 10^{19} \frac{\text{W}}{\text{m}^2}}{8.8 \times 10^{-12} \frac{\text{As}}{\text{Vm}} \cdot 3 \times 10^8 \frac{\text{m}}{\text{s}}}} \approx 1.1 \times 10^{11} \frac{\text{V}}{\text{m}}$$

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

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

III Electron captured by parent ion, photon emitted

Ser E_{kin}+I_p
E_{kin}

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

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Classical description:

Free electron in an oscillating E-field

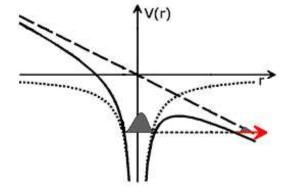
$$E(t) = E_0 \sin(\omega t)$$
 monochromatic field

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

$$x_i = 0$$
 and $v_i = 0$ at t_i

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

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

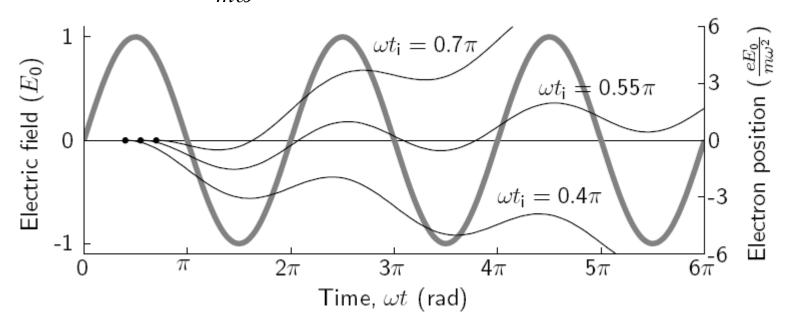


P. B. Corkum, Phys Rev Lett 71, 1994 (1993) K. Varjú, Am. J. Phys. 77, 389 (2009) analytic solution

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

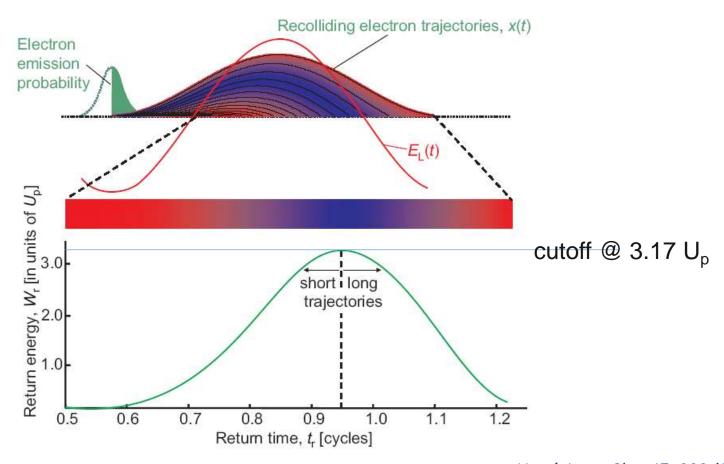
$$x(t) = \frac{eE_0}{m\omega^2} \left[\sin(\omega t) - \sin(\omega t_i) - \omega(t - t_i) \cos(\omega t_i) \right]$$



- 1, the electron may 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

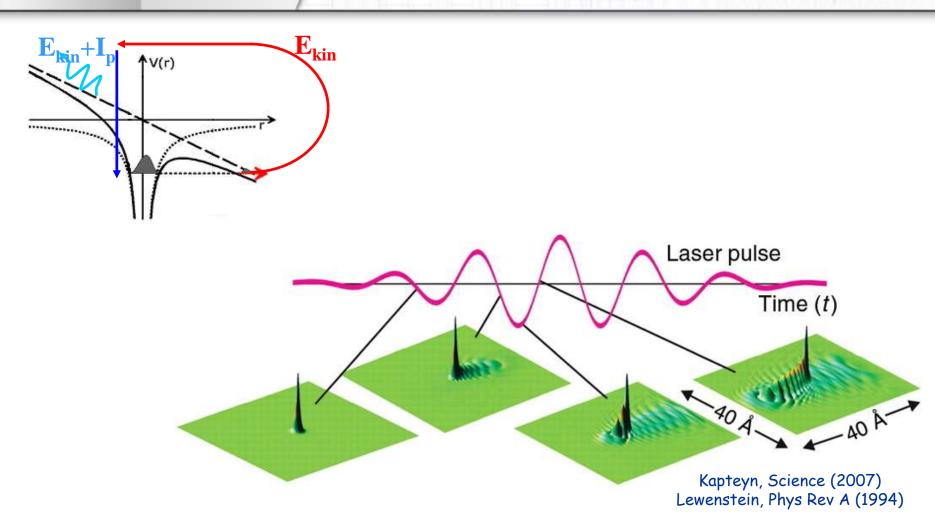
Chirp of attosecond pulses

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



HHG in the quantum picture





low efficiency!!!

Harmonic radiation

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

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

- 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 propagation in the laser field $\vec{x}(t) = i \int_0^t dt' \int d^3\vec{p} \ \vec{d}*(\vec{p} - \vec{A}(t))$ of electron $\times \exp[-iS(\vec{p},t,t')] \ \vec{E}(t') \cdot \vec{d}(\vec{p} - \vec{A}(t')) + \mathrm{c.c.},$

Lewenstein integral

ionization transition

Harmonic spectrum

Harmonics are emitted as a result of the dipole oscillations

Fourier transform of the dipole moment:

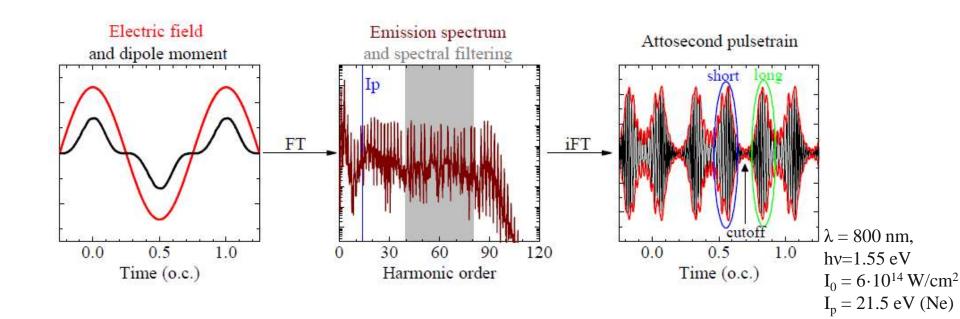
 $x(\omega) = \int_{-\infty}^{+\infty} dt \ x(t) \exp(i\omega t)$

can be decomposed as

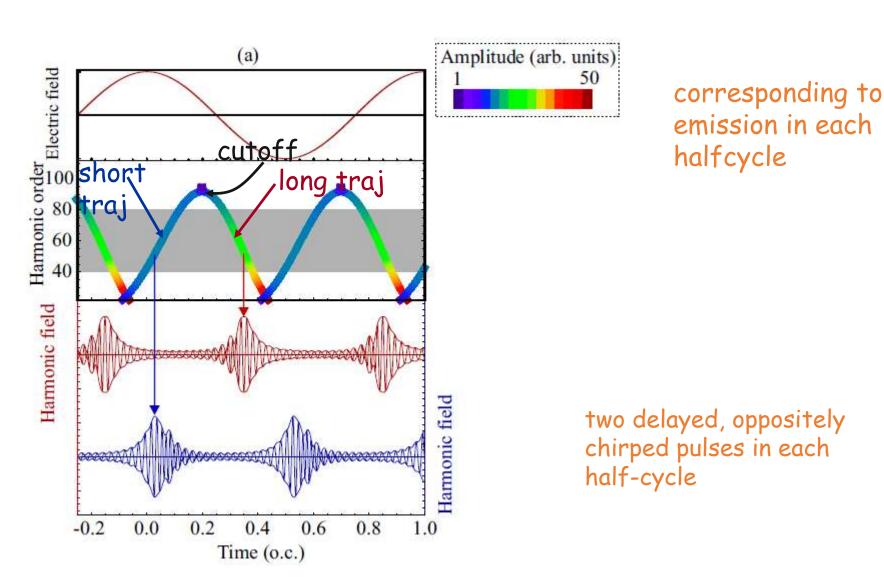
$$x(\omega) = \sum_{s} |x_{s}(\omega)| \exp[i\Phi_{s}(\omega)]$$

harmonic emission rate

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

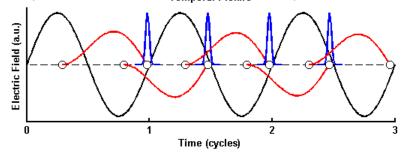


Half-cycle periodicity separation of short and long traj components

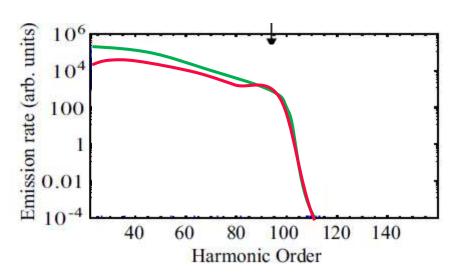


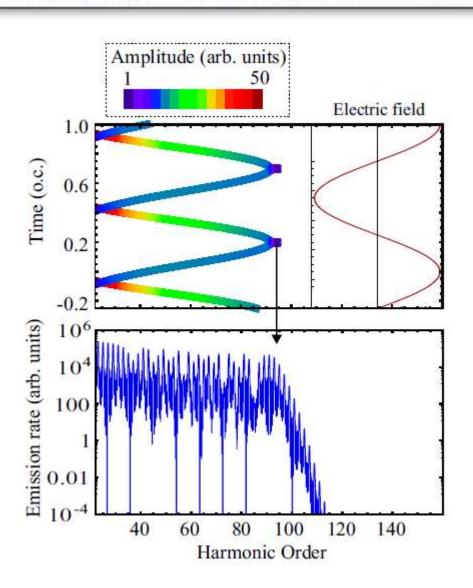
Periodicity

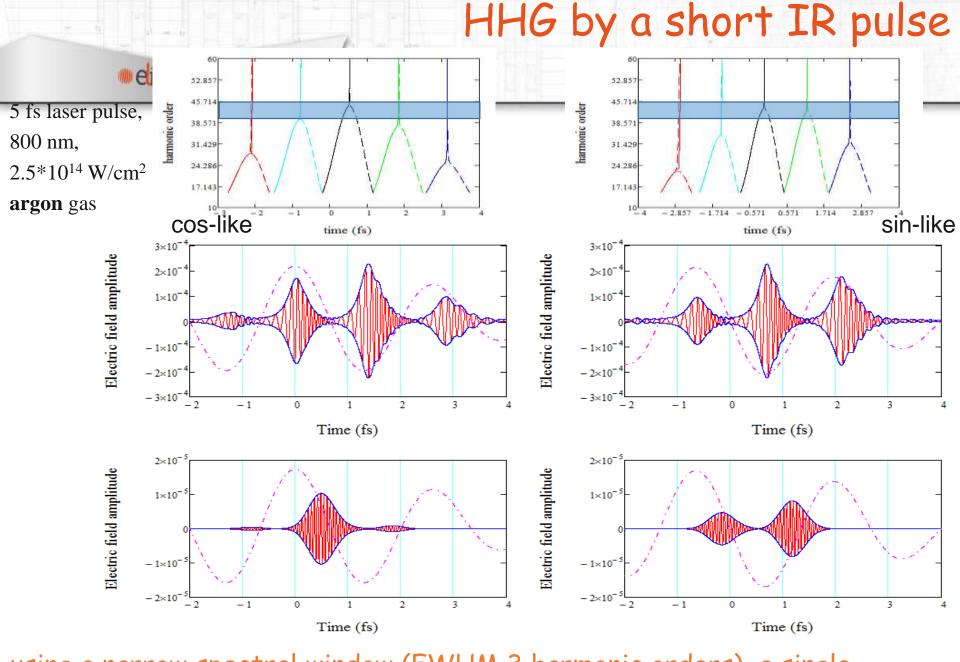
harmonic emission process repeated in each half cycle



radiation emitted in each half cycle



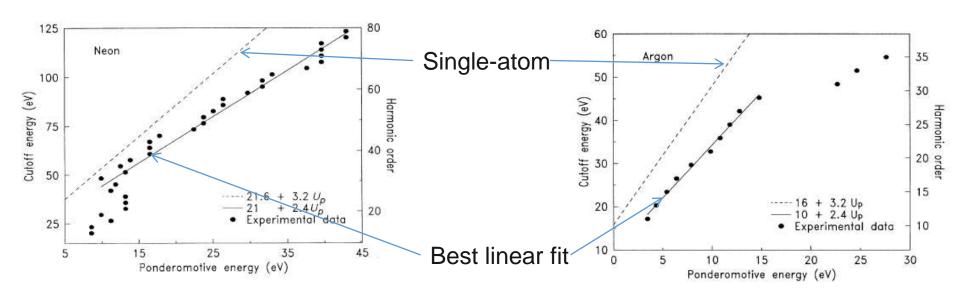




using a narrow spectral window (FWHM 3 harmonic orders), a single attosecond pulse can be selected - only short trajectories are considered!

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



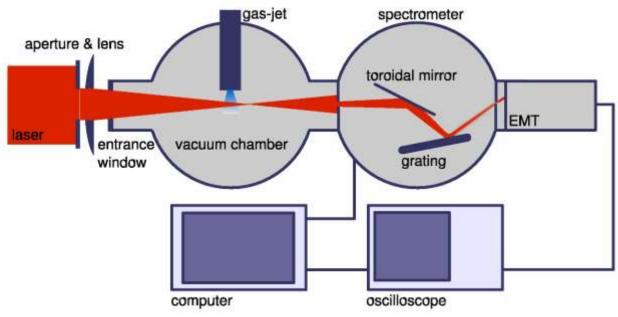
Macroscopic effects play an important role!!

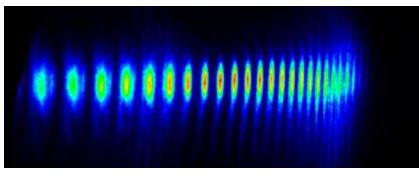
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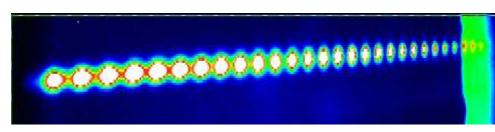
- · High order harmonic generation in gaseous media
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Photon detection







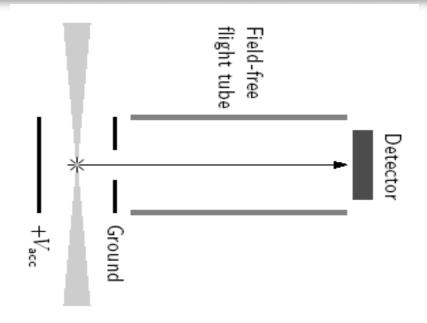
Kazamias et al.

Nisoli et al., 2002

Spectral amplitude - no information about temporal features

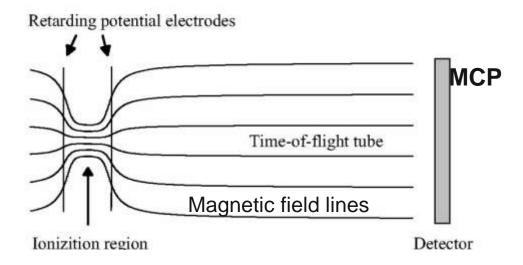
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Electron / ion detection



Time-of-flight spectrometer

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

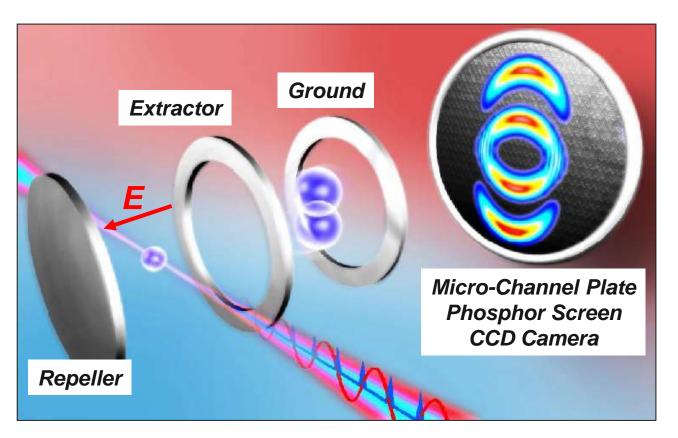


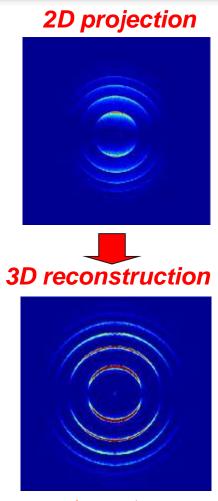
Magnetic Bottle Electron Spectrometer photoionisation in a strong magnetic field (1T), reduced gradually towards the end of flight tube enables 2π collection of electrons

1

Velocity map imaging (VMI)

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



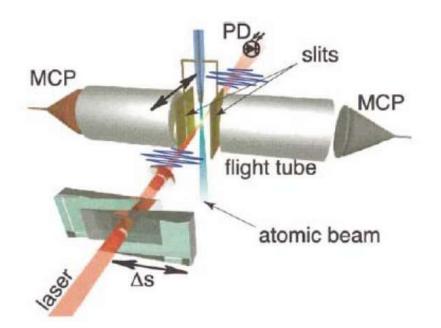


Detector captures the 2D projection of electron momentum distribution + assume symmetry around the E-field Abel inverson → reconstruction of the full distribution

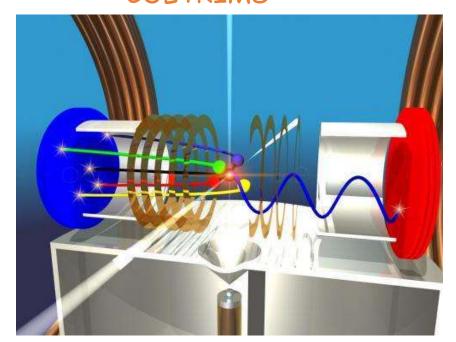


Stereo-machines

Coincidence measurements



Reaction Microscope / COLTRIMS



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ei
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ultrashort laser pulses:
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autocorrelations (second/third order)
SPIDER (spectral shear)
FROG (second/third order)

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"

I_p

in most cases we measure the electron/ion replicas



Temporal characterization Attosecond metrology

Temporal characterisation schemes

Autocorrelation

2nd order intensity volume autocorrelation (IVAC)

XUV SPIDER

Cross-correlation

X-FROG

RABITT

Asec streak camera

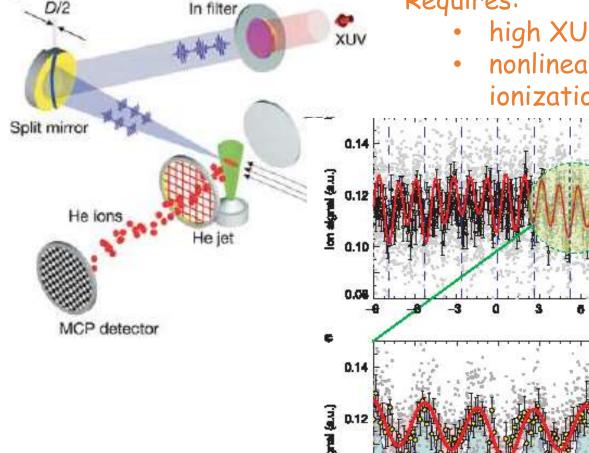
FROG - CRAB



2nd order autocorrelation

Direct measurement of pulse duration Requires:

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



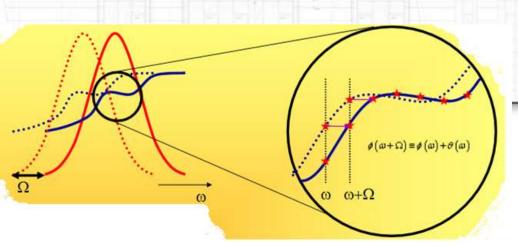
6 0.10

Delay, At (fa)

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

Status:

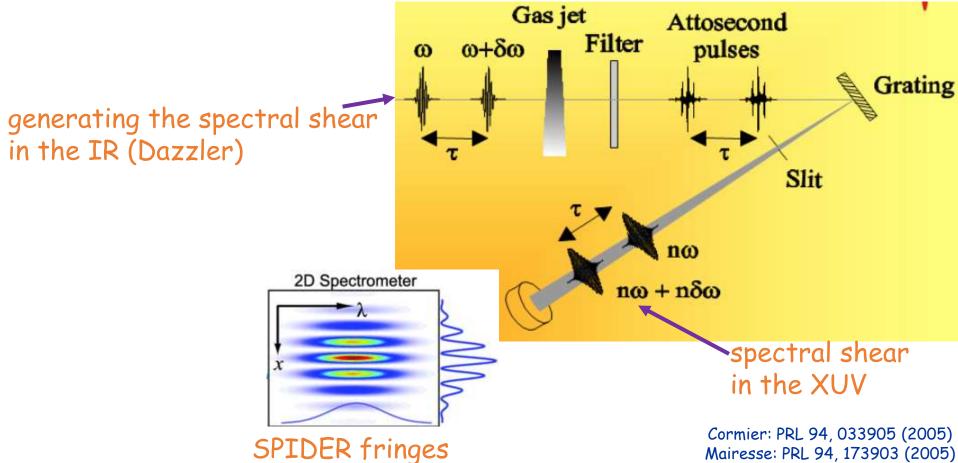
- spectral range: up till 30 eV
- pulse duration:320 as



XUV (SEA) SPIDER

Mairesse: PRL 94, 173903 (2005)

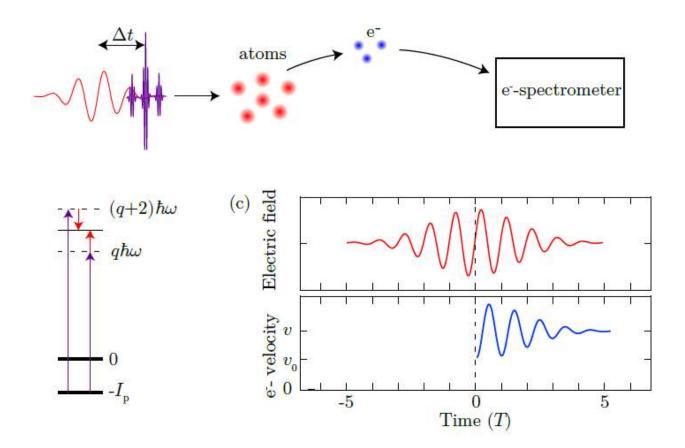
principle: spectral shear + delay interference



Cross-correlation

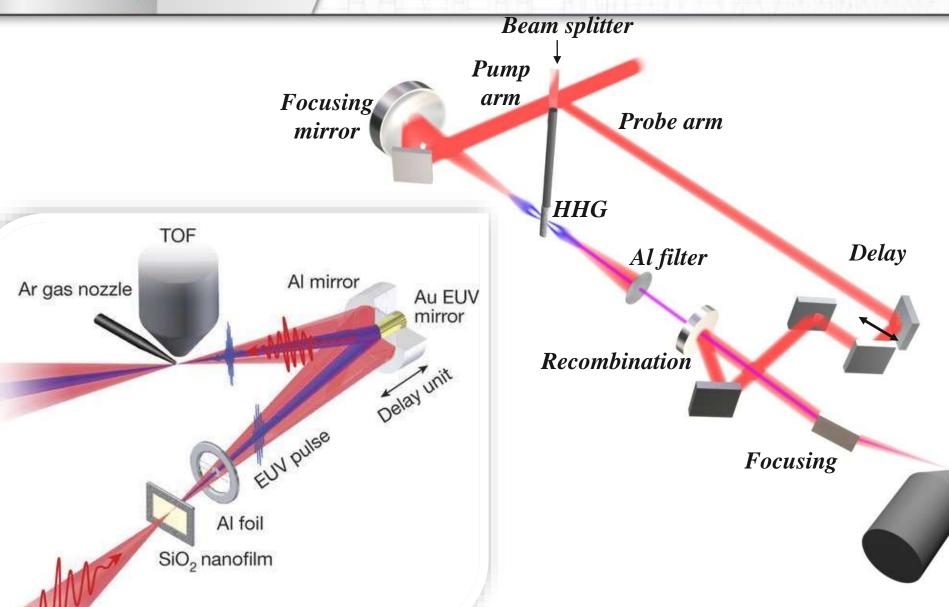
Low intensity of the high-harmonic radiation makes auto-correlation techniques less practical.

Scheme: XUV photoionization in the presence of the IR field

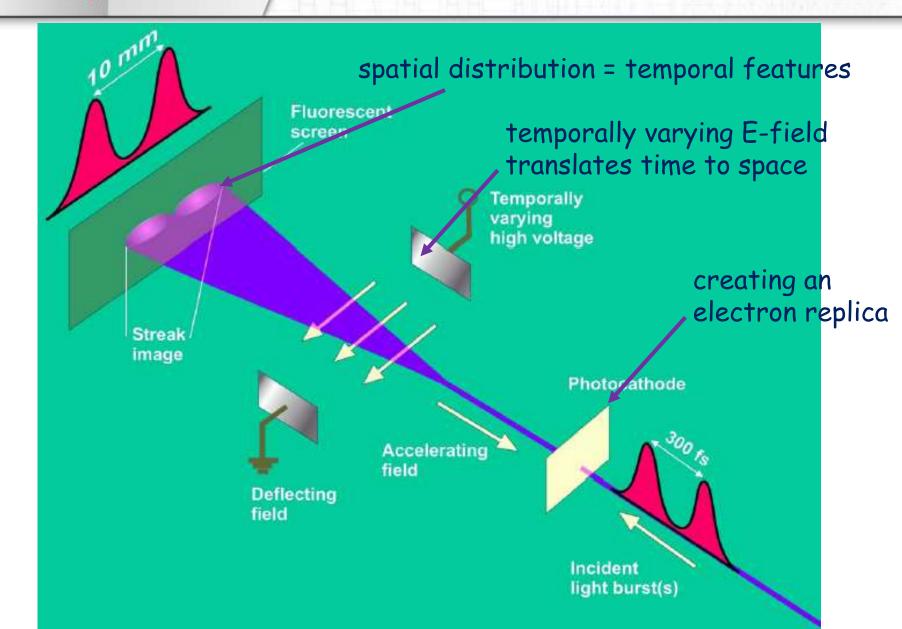


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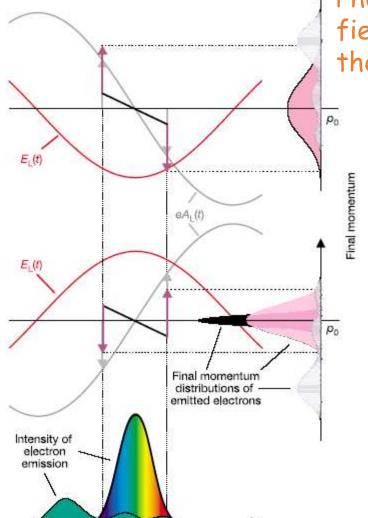
Experimental arrangement



Conventional streak camera (ps)



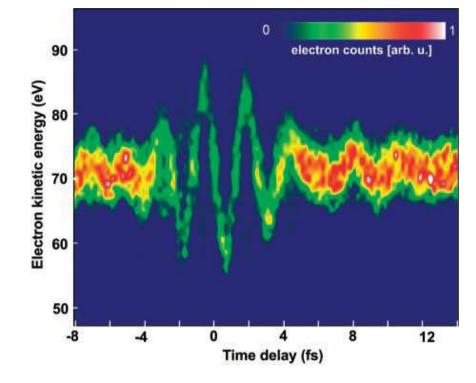
Attosecondstreak camera (strong IR)



Time

Photoionization in the presence of the IR field:

the IR E-field provides the fast streaking



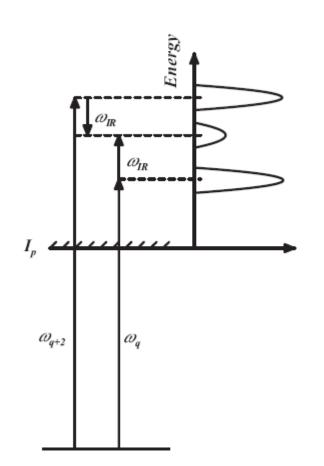
Drescher: Science 291, 1923 (2001) Itatani: PRL 88, 173903 (2002)

Kitzler: PRL 88, 173904 (2002)

Gouliemakis: Science 305, 1267 (2004)

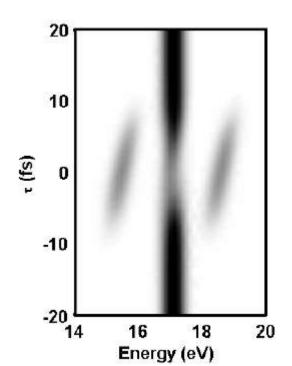
Creation of sidebands (weak IR)

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



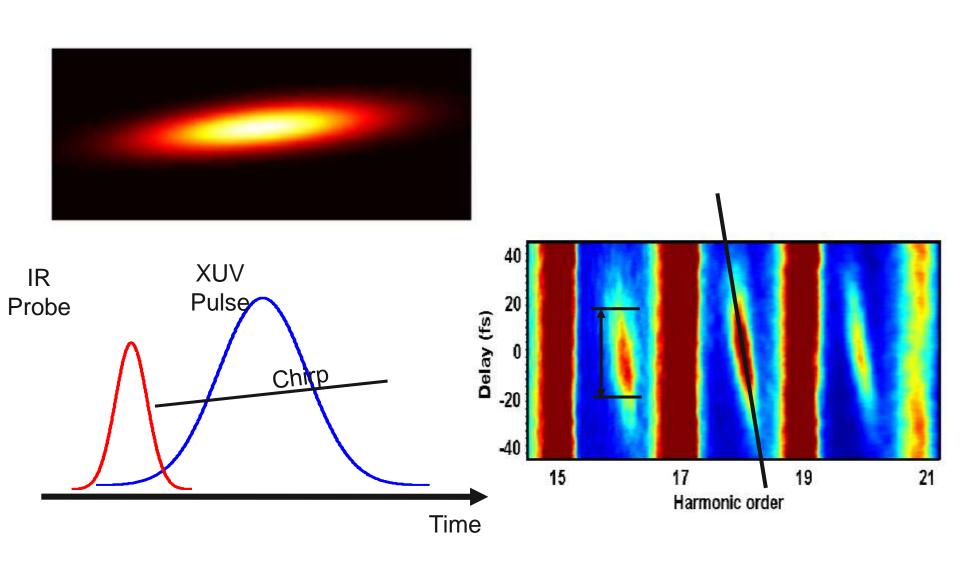
$$\begin{split} \tilde{\varepsilon}_{IR}(t) &= \tilde{\varepsilon}_{IR}^+(t) + \tilde{\varepsilon}_{IR}^-(t), & \text{absorption, emission} \\ \tilde{\varepsilon}_{IR}^+(t) &= \tilde{A}_{IR}(t) \, e^{-i\omega_0 t}, \\ \tilde{\varepsilon}_{IR}^-(t) &= \tilde{A}_{IR}^*(t) \, e^{+i\omega_0 t}. \end{split}$$

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



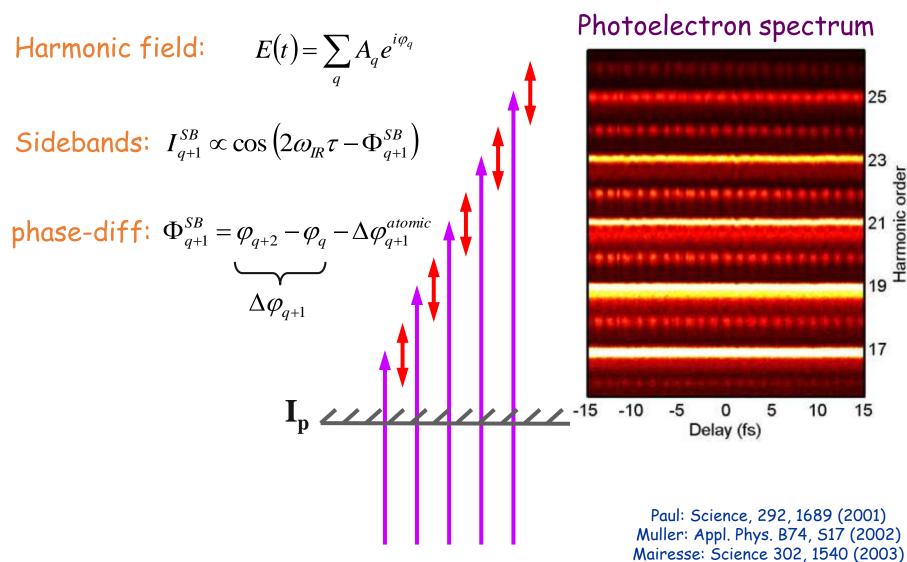
Femtosecond characteristics: XFROG

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



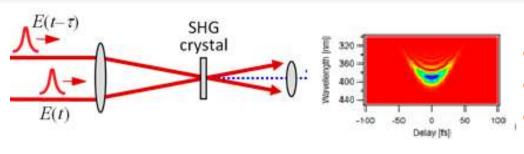
Attosecond characteristics: RABITT

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



FROG CRAB

Frequency Resolved Optical Gating for Complete Resolution of Attosecond Bursts

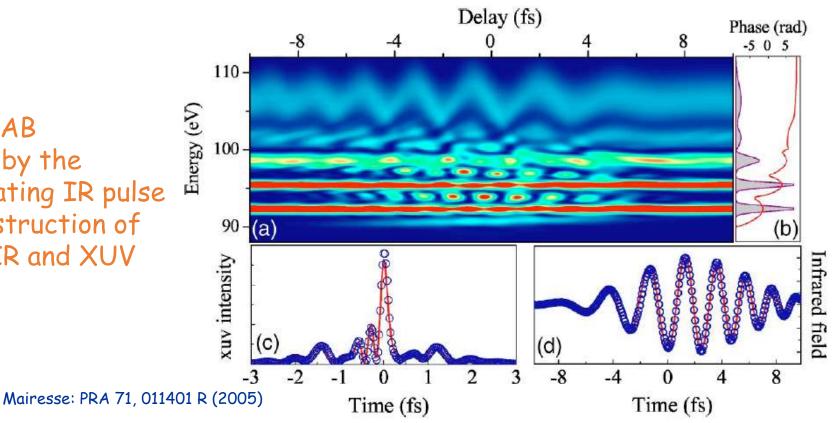


FROG:

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

FROG CRAB

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



FROG CRAB - examples

isolated attosecond burst

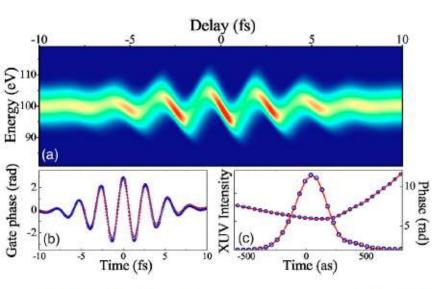


FIG. 1. (a) CRAB trace of a single 315 as pulse [full width at half maximum (FWHM) of intensity], having second- and third-order spectral phases (Fourier limit=250 as), gated by a Fourier-limited 6-fs 800 nm laser pulse, of 0.5 TW/cm² peak intensity. The electrons are collected around θ =0 with an acceptance angle of $\pm 30^{\circ}$. (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

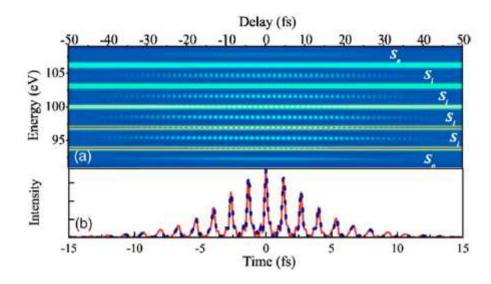
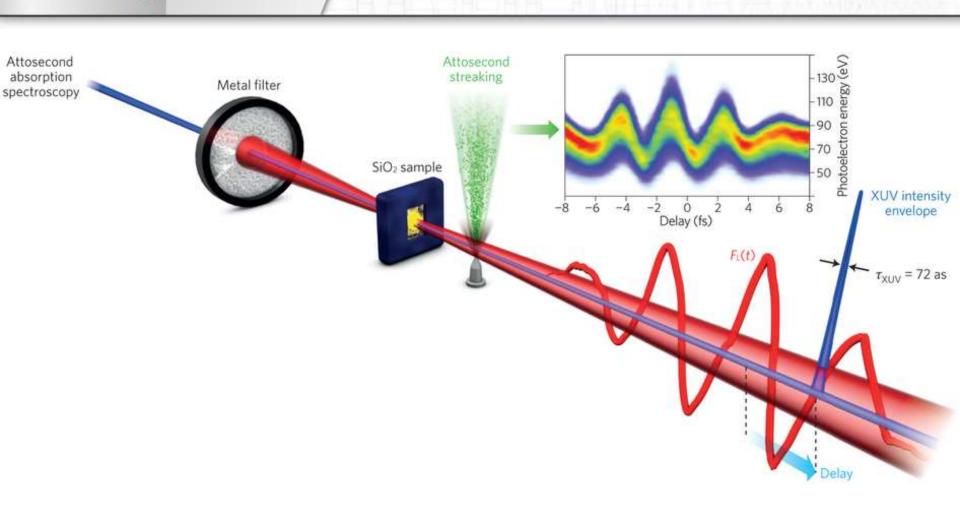


FIG. 2. (a) CRAB trace at θ =0 of a 12-fs-train of nonidentical as pulses, of period T/2=1.3 fs, gated by a 30-fs-800 nm- (T=2.6 fs) laser pulse, of 0.05 TW/cm² peak intensity, assuming a spectrometer resolution of 100 meV. The as pulses are shorter in the center of the train (\approx 250 as), than in 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)

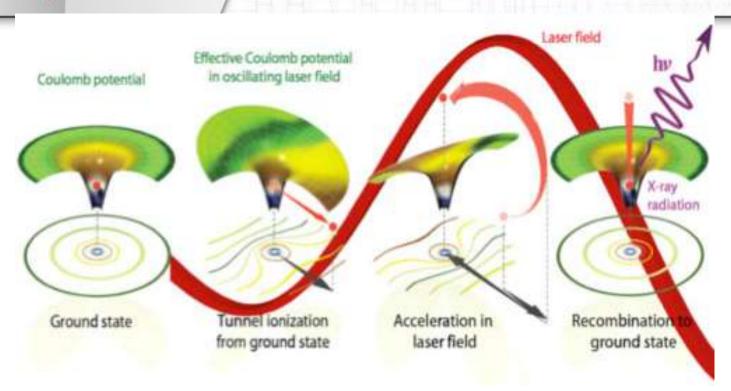
On-line characterisation



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"Delay" of the HHG process



HHG is a non-instantaneous process: electron travels in the continuum, ie the harmonics are shifted in phase relative to the fundamental: phase of the dipole moment.

Phase depends on harmonic order and intensity and trajectory type (short/long)!

Short andlong trajectory components can be distinguished based on spectral and spatial characteristics.

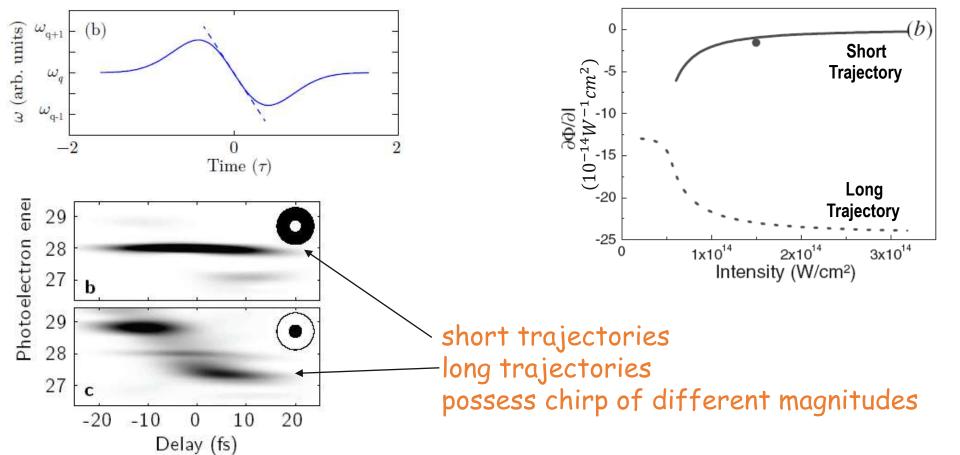
Multicycle phase modulations: harmonic chirp

The intensity of the driving pulse varies on the multicycle timescale

+ dipole phase depends on intensity \Rightarrow time-dependent phase

$$\omega_q(t) = q\omega_0 + \frac{\partial \Phi_j}{\partial t} = q\omega_0 + \alpha(q)\frac{\partial I}{\partial t} \approx q\omega_0 - \alpha(q)\frac{8\ln(2)I_0}{\tau^2}t$$

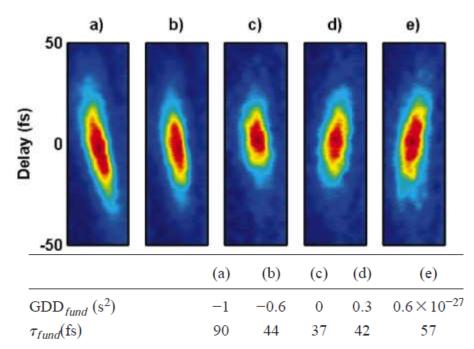
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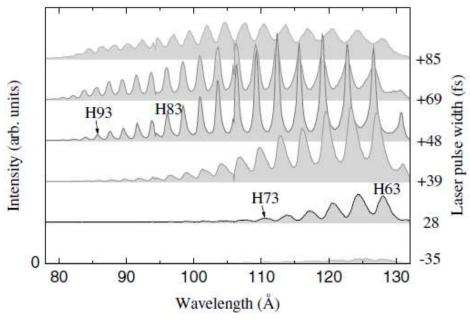
Generating harmonics with a chirped fundamental

varying the chirp of the fundamental

$$b_j^q = q b_{\text{fund}} + 8 \ln(2) \frac{I_0}{\tau_0^2} \frac{\partial \Phi_j^q}{\partial I}$$



adding a suitable positive chirp to the fundamental can compensate the chirp resulting from the intensity dependence of the atomic phase:



Subcycle phase modulations: attosecond structure

related to chirp

Determined by the frequency dependence of the harmonic phase

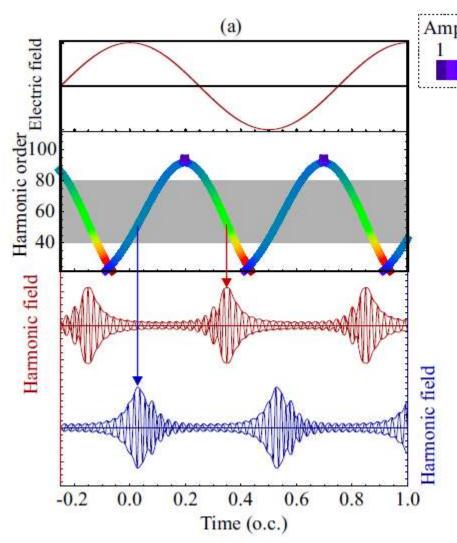
$$\Phi = \Phi_q$$

$$\Phi_{q} = \Phi_{q0} + \frac{\partial \Phi}{\partial q} (q - q0) + \frac{1}{2} \frac{\partial^{2} \Phi}{\partial q^{2}} (q - q0)^{2} + \dots$$
group delay
group delay
group delay dispersion

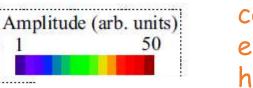
$$\frac{\partial}{\partial q} \rightarrow \frac{1}{\omega 0} \frac{\partial}{\partial q} = \frac{\partial}{\partial \Omega}$$

emission time or group delay
$$t_{\mathrm{e},\,j}^q = \frac{1}{\omega} \frac{\partial \Phi_j^q}{\partial q}$$

Attochirp for short and long trajectory components



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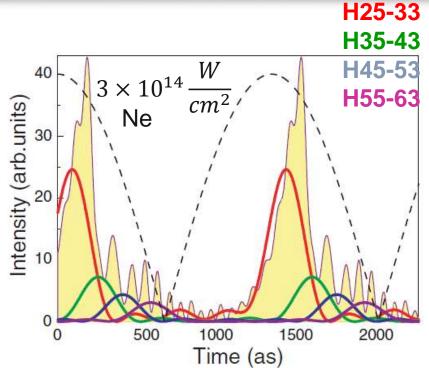


corresponding to emission in each halfcycle

Saddlepoint analysis enables separation of short and long trajectories.

Short and long trajectory components are delayed and oppositely chirped: attochirp.

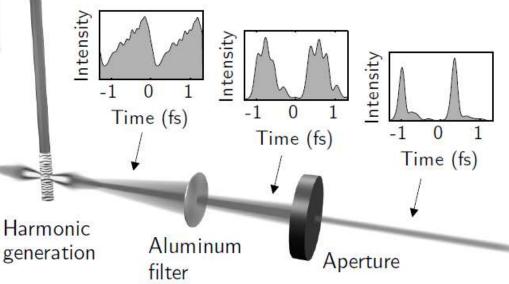
Attosecond pulse train synthetised from a broad sectrum



Mairesse, Science 302, 1540

m eli

postcompression is required for short pulse generation

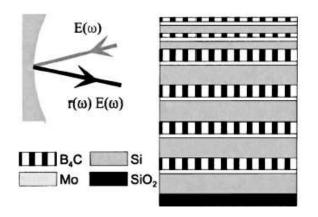


Postcompression of attosecond pulses

■ ei

XUV chirped mirror

aperiodic multilayer structure

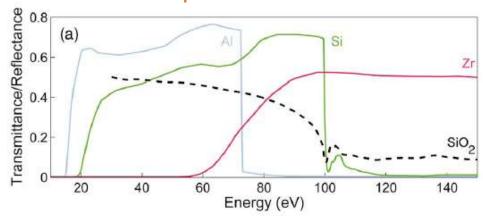


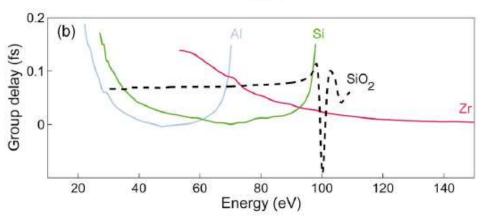
uncertainty in refractive indices very severe conditions on layer thickness precision

Morlens: Opt Lett 31, 1558 (2006)

Metallic films

anomalous dispersion just above the absorption band



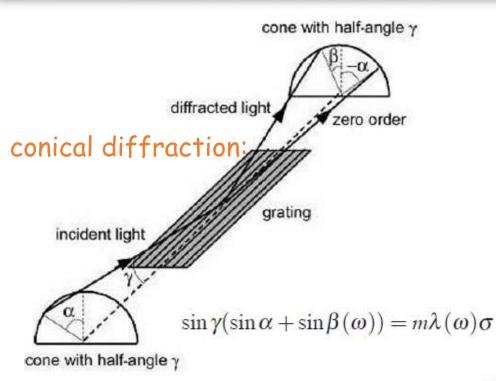


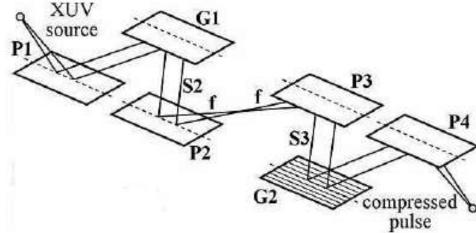
200 nm thick filters

Gustafsson: Opt Lett 32, 1353 (2007)

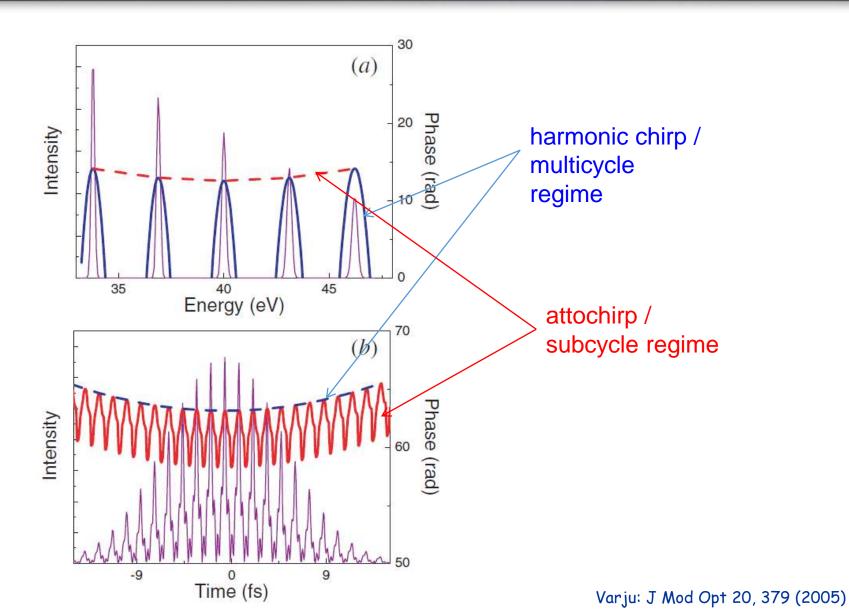


Grazing incidence grating compressor

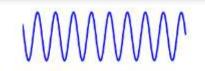




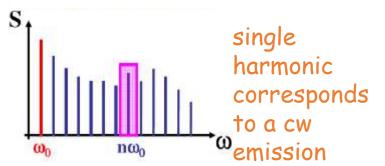
Spectral and temporal phase of the high harmonic radiation

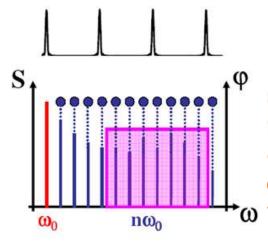


Complete characterization of the spectral phase

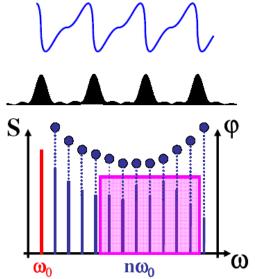


m eli

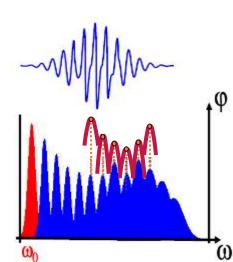




multiple harmonics phaselocked corresponds to an attosecond pulse 'w train

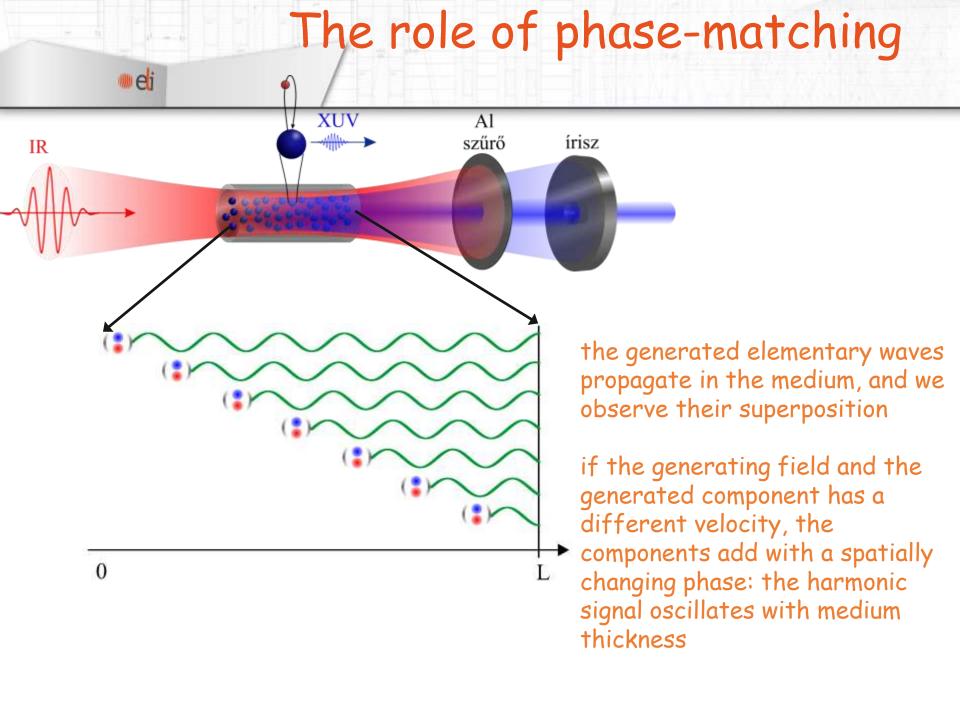


multiple harmonics with quadratic spectral phase corresponds to chirped attosecond pulses in the train

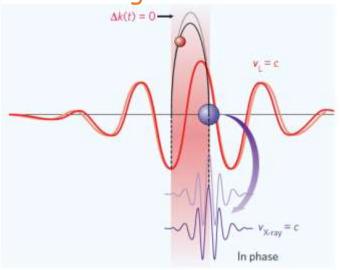


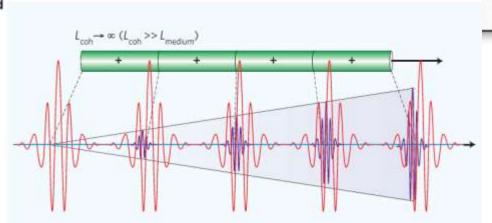
chirped individual harmonics correspond to broader harmonics and pulse-to-pulse variations in the attosecond pulse train

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



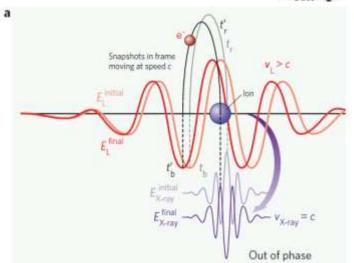
Phase-matched generation

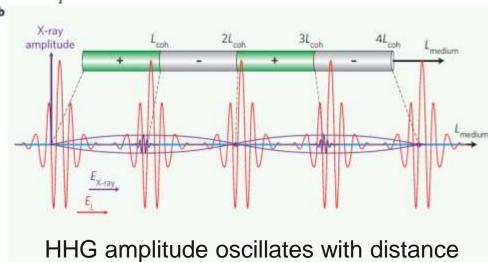




HHG amplitude grows linearly with distance

Non-phase-matched generation $L_{\rm coh}(q) = \pi/\Delta k_q$





Popmintchev: Nphot 4, 822 (2010)

Components of phase-mismatch

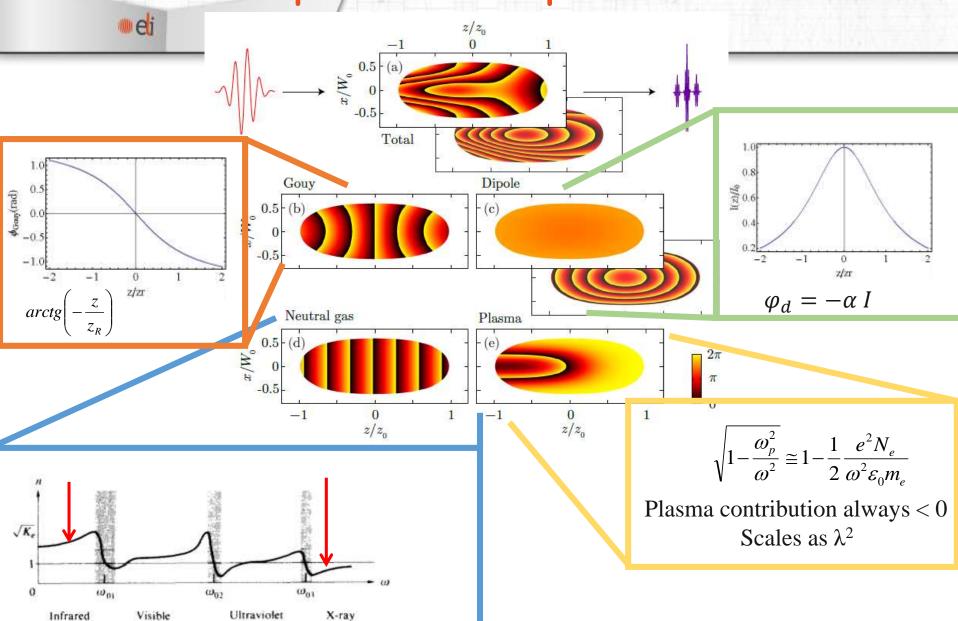
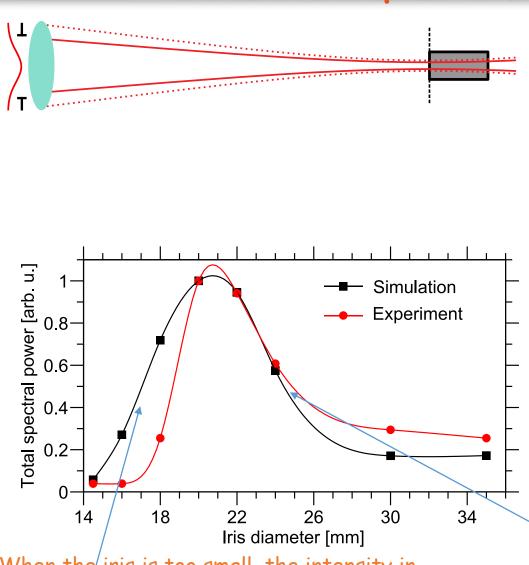


Figure 3.41 Refractive index versus frequency. Hecht, Optics (2002)

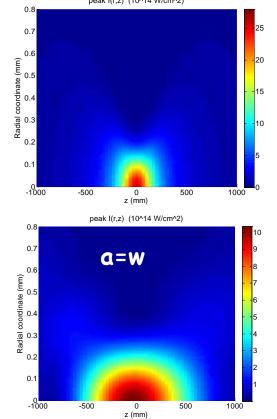
Ch Heyl, PhD dissertation

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



₩ ei

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



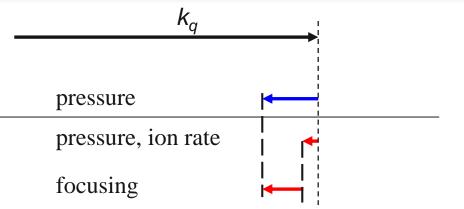
With 86% of the energy the peak intensity is only 40% of the untruncated value

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.

Phase velocity c for harmonic q

- neutral dispersion for the XUV
- neutral + plasma dispersion for the IR
- Gouy phase-shift

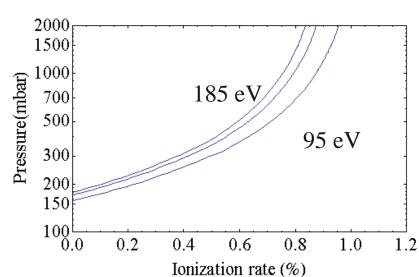


For a given focusing geometry and ionization rate, there is an optimal pressure:

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

$$\uparrow$$
Has to be positive!

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



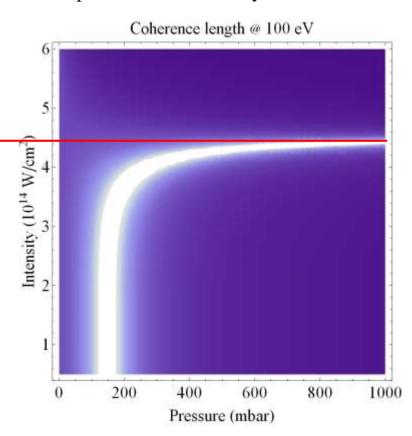
Balogh E, PhD dissertation

eli

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

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

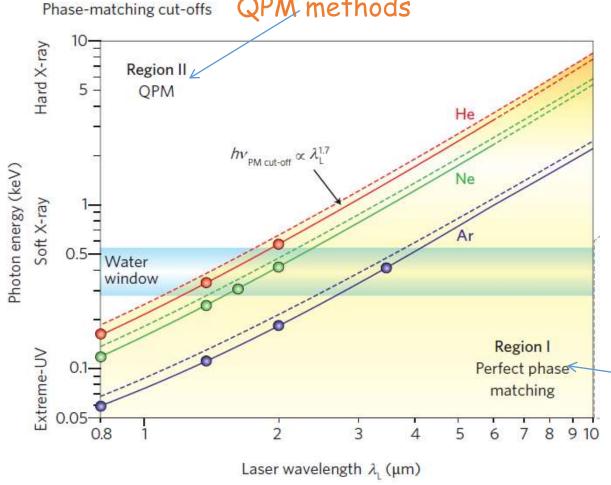
Coherence length as a function of pressure and intensity



₩ ei

Pressure-tuned phase-matching





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

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

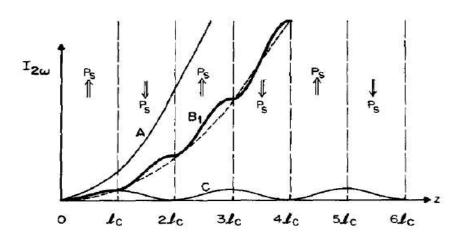
PHYSICAL REVIEW

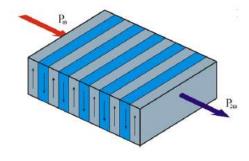
VOLUME 127, NUMBER 6

SEPTEMBER 15, 1962

Interactions between Light Waves in a Nonlinear Dielectric*

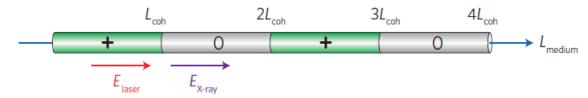
J. A. Armstrong, N. Bloembergen, J. Ducuing, And P. S. Pershan Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts (Received April 16, 1962)



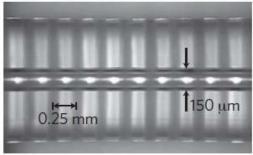


QPM techniques in HHG

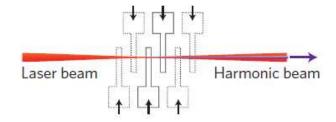
to periodically switch off HHG in destructive zones



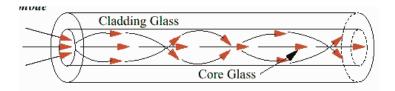
aPeriodically modulated hollow waveguide



Successive gas jets

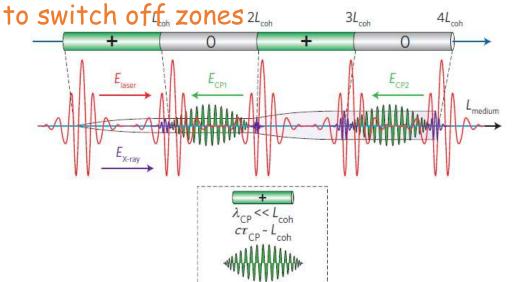


multimode waveguide



QPM techniques in HHG

counter-propagating pulses scramble the phase of high-harmonic emission

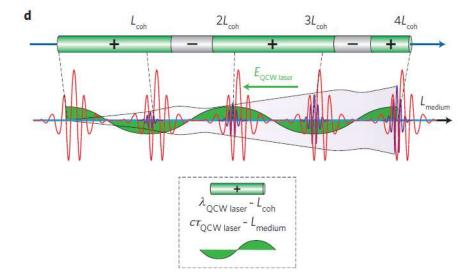


X-ray-generating
laser pulse
Argon-filled
waveguide

CP laser pulses

Pulse collision point in plasma

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



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

- **""** Cu
- 1, increasing the achievable photon energy ("water-window")
- 2, increasing the XUV photon flux (up-scaling)
- 3, producing a Single Attosecond Pulse (gating)

Spectral extension

$$\hbar\omega_{max} = I_p + 3.17 \ U_p$$
$$U_p \propto I \ \lambda^2$$

How to increase the cutoff?

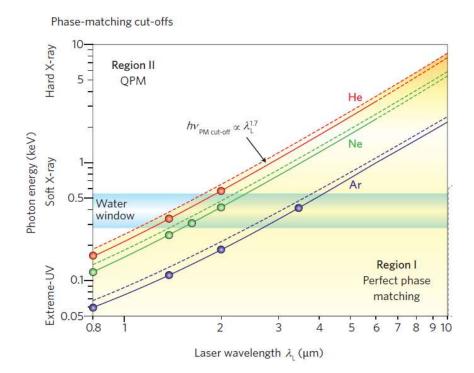
- increase laser intensity
 limit: ionization of the medium
 (phase matching, depletion)
 avoid: short pulses, QPM
- increase laser wavelength limit: laser technology
- increase ionization potential e.g. generate with ions limit: phase matching

typical values:

$$I_p = 10..24 \text{ eV}$$

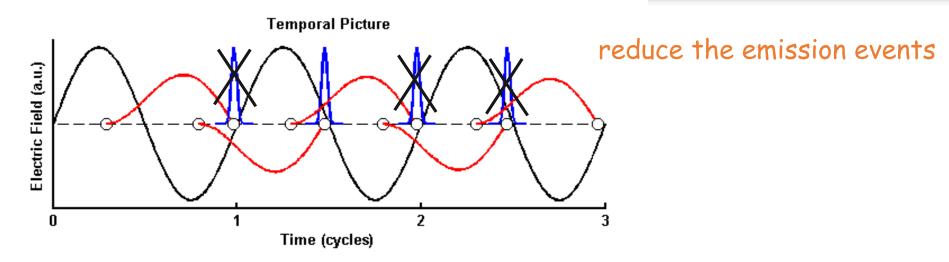
$$I = 10^{15} \text{ W/cm}^2 \ @ 800 \text{ nm gives } U_p = 60 \text{ eV}$$

$$I_p + 3.17 U_p \approx 200 \text{ eV}$$

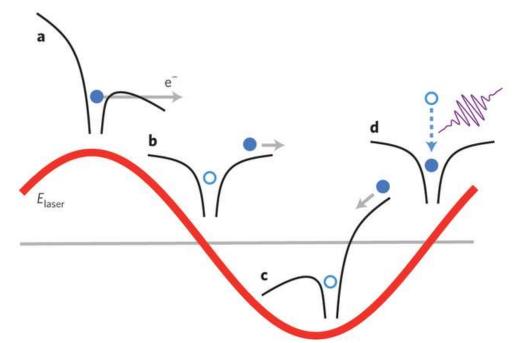




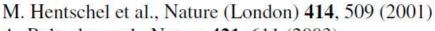
Temporal gating



by avoiding ionization, or recombination, or shortening the generating pulse

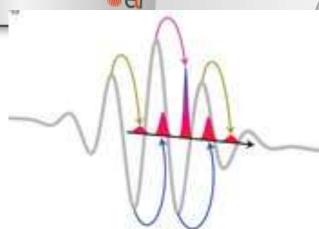


/Amplitude/intensity gating

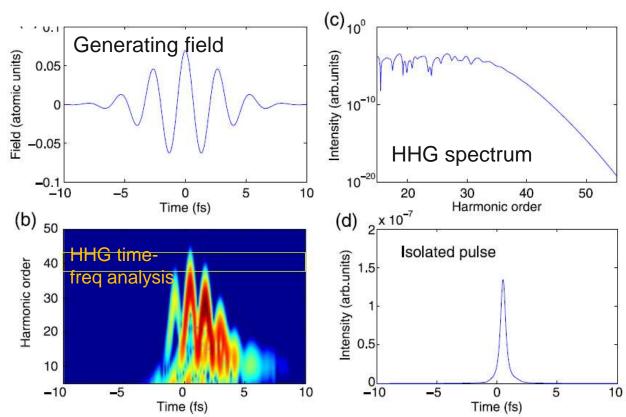


A. Baltuska et al., Nature 421, 611 (2003)

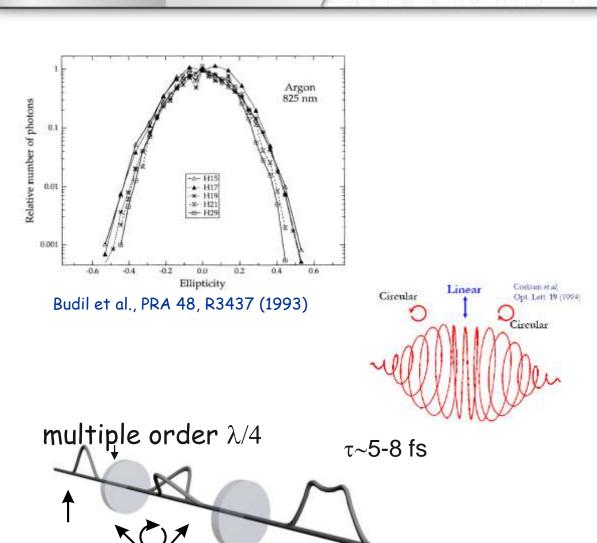
spectrally filtering the cutoff small intensity small bandwidth



 τ < 5 fs, CEP-stable driving laser

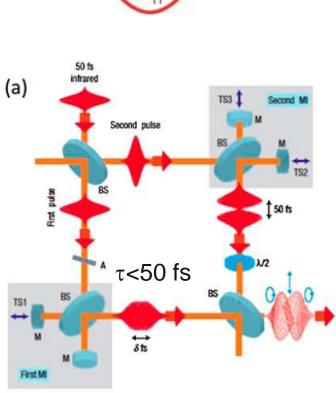


Ellipticity-gating



₩ ei

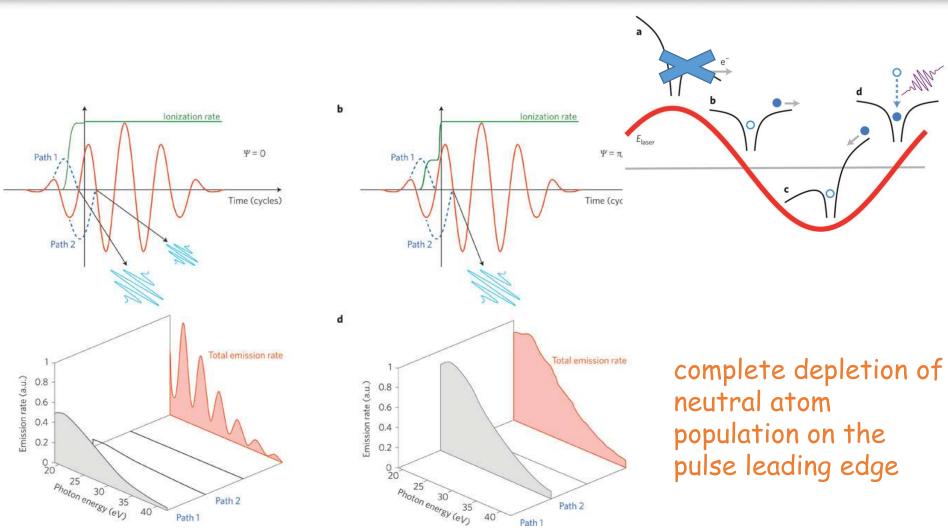
Sansone: Science 314 (2016)



Tzallas: Nature Physics 3, 846 - 850 (2007)

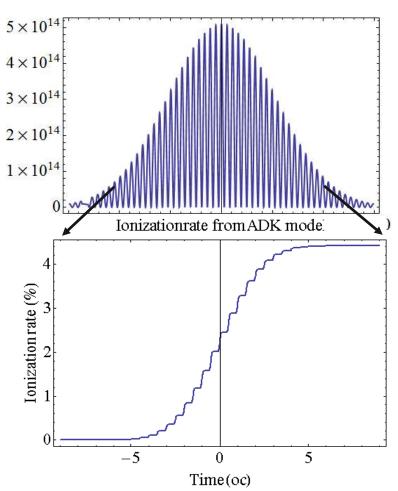


Ionisation gating I. single atom effect



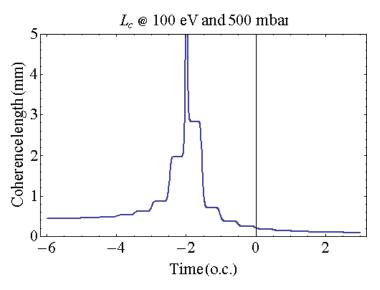
Ionisation gating II.

Macroscopic: time-dependent coherence length



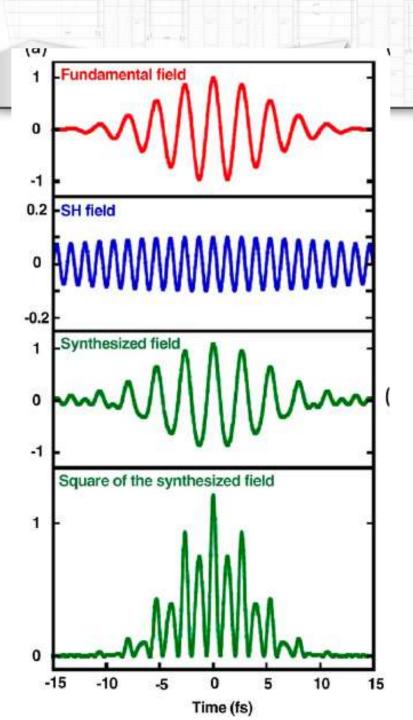
Lcoh > 1 mm for only 1 optical cycle

Temporal gating: isolation of a single attosecond pulse



Balogh E, PhD dissertation

for ex.: $5.1*10^{14}$ W/cm², 35 fs pulse

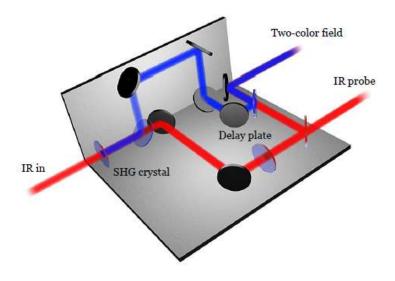


Two-color gating (with SH or MIR)

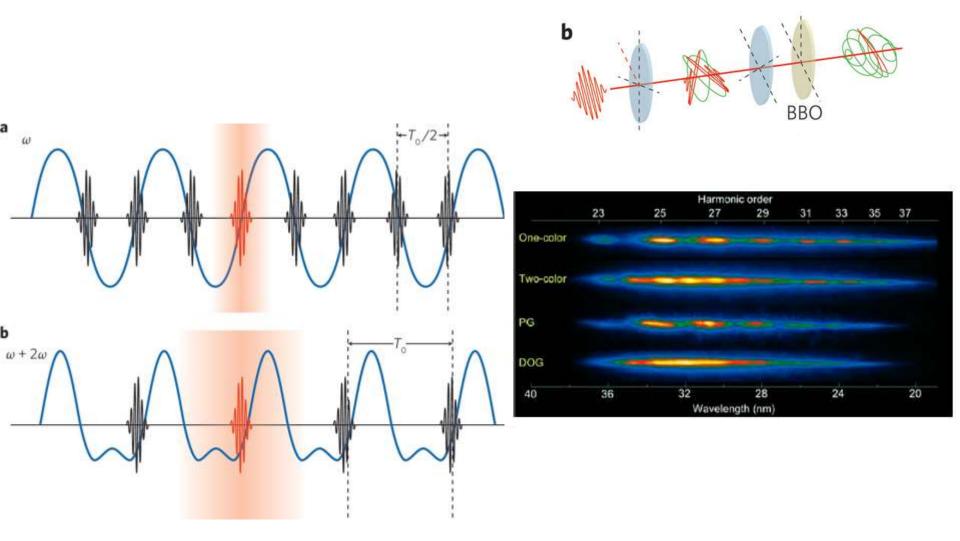
Tunable weak perturbing pulse (harmonic or longer wavelength)

Increases the period of the process (least common multiple)

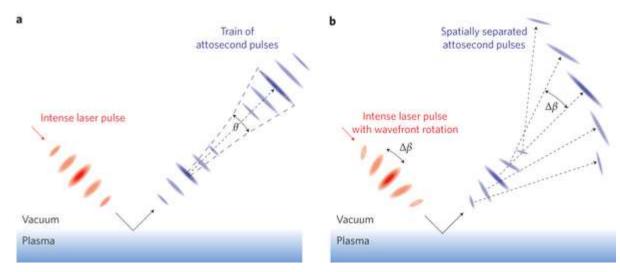
Can be combined with any other gating process



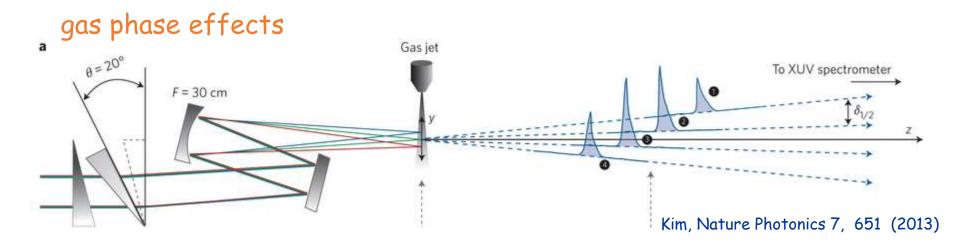
Polarization + two-color gating = Double Optical Gating (DOG)



surface plasma effect



Wheeler, Nat Phot 6, 829 (2012)



et /

Further reading

Boyd: Nonlinear Optics

Chang: Fundamentals of attosecond optics

Plaja (ed): Attosecond Physics

Vrakking (ed): Attosecond and XUV Physics





THANK YOU FOR YOUR ATTENTION!









INVESTING IN YOUR FUTURE