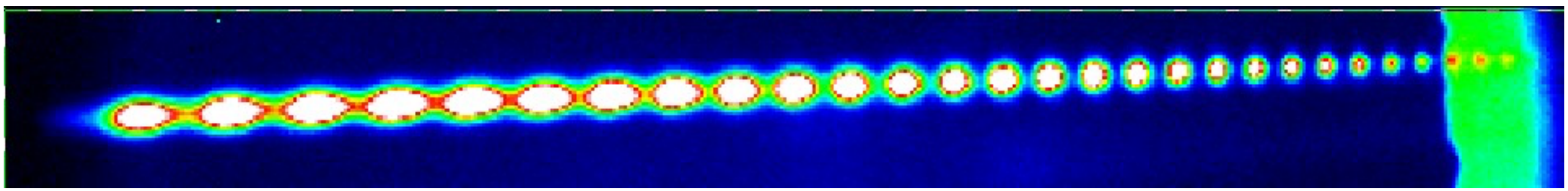
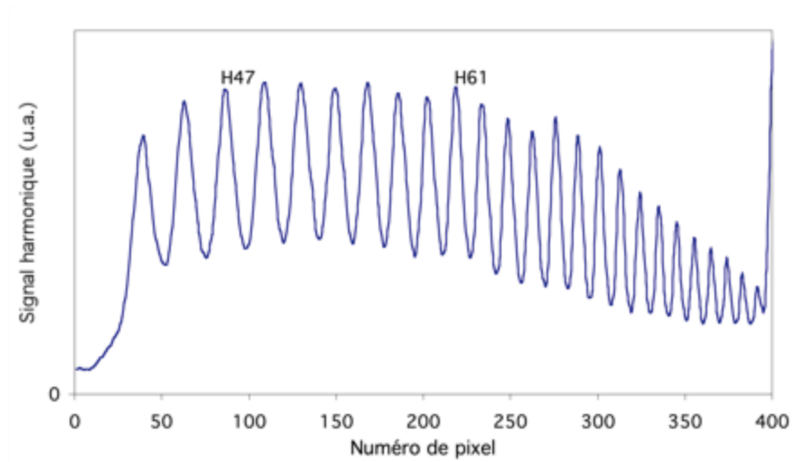
High order harmonic generation: From concept to experiment



Typical harmonic spectrum in Neon



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Plan of the lecture:

- The most characteristic features of HHG, historical aspects
- How it looks like experimentally?
- The physical origin of the non linear polarization of atoms
- The problem of phase matching
- An ultrashort story of the attosecond structure
- Modern trends (not exhaustive and subjective)

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The most characteristic features of HHG, historical aspects:

- In 1985: the famous article by Strickland and Mourou opens the way to high power lasers with ultrashort pulse duration
- End 80's: studies about photo-ionization of atoms by intense lasers, electron/ion spectrometers, ATI spectra, first harmonic spectra

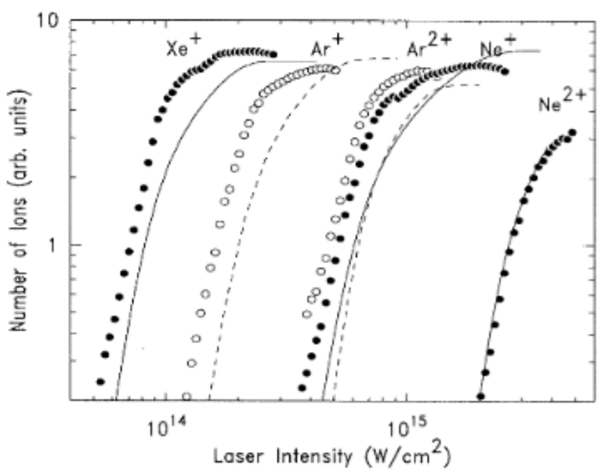


FIG. 3. Xe^{+} (full circles), Ar^{+}, Ar^{2+} (open circles), Ne^{+}, and Ne^{2+} (full circles) ions as a function of the laser intensity. The lines (dashed for the Ar ions and solid for the Xe and Ne ions) are the predictions from tunnel ionization.

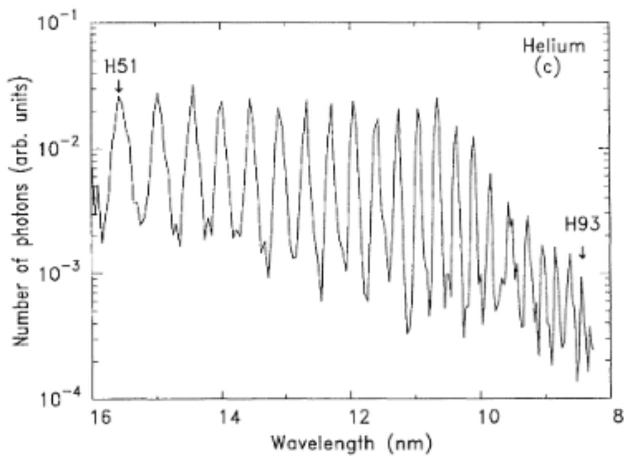


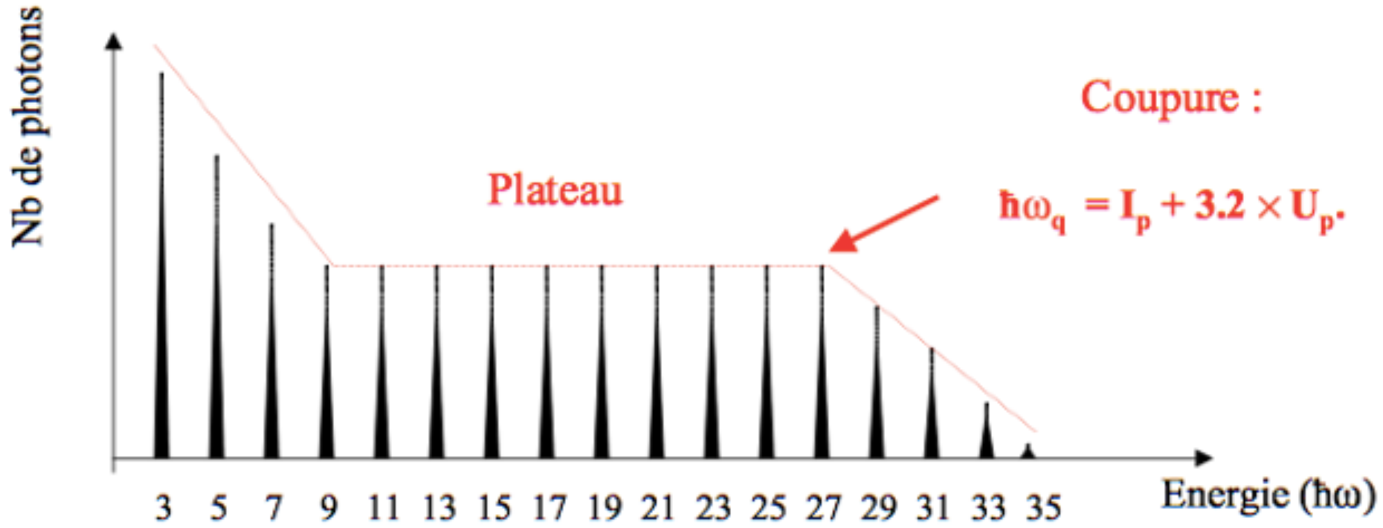
FIG. 4. Harmonic spectra in (a) Xe, (b) Ar, and (c) He. The laser intensity is approximately 10^{15} W/cm^2.

From C.G. Wahlström et al, PRA (1993)

The most characteristic features of HHG, historical aspects:

- 1993: the three step model is proposed to explain the physical origin of HHG, the cut-off law, still qualitative, semi-classical
- 1994/1995: the Lewenstein model, still quite "simple" but allows a more quantitative and quantic approach, explains the main features
- 1995: First prediction of the possibility for attosecond structure
- End 1990's: The macroscopic aspects of HHG is studied both theoretically and experimentally
- 1999: The definition of the absorption limit for HHG
- Beginning 2000: first attosecond characterizations with the RABBIT method, attoscience is born
- Afterwards: The HHG source is used as a tool for applications in atomic physics, molecular physics, solid state physics, etc.
- Now: It can be a compact commercial source at high rep rate and people use it as a turn key black box

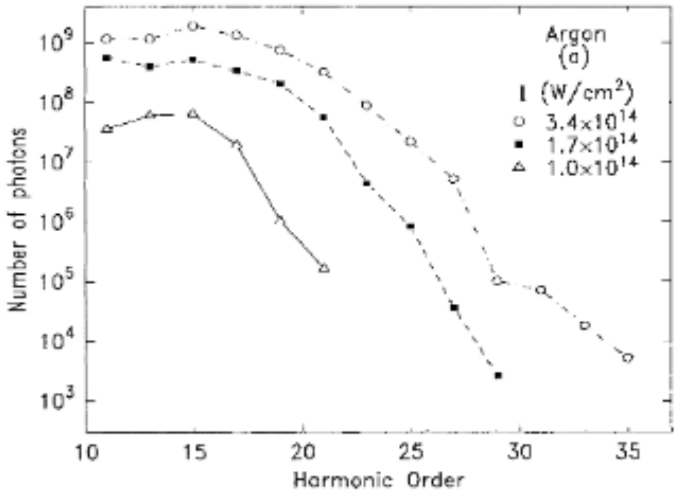
The most characteristic features of HHG:



$E_q = q\hbar\omega = \hbar\omega_{\mathrm{q}}$

q is odd, there are 3 spectral regions

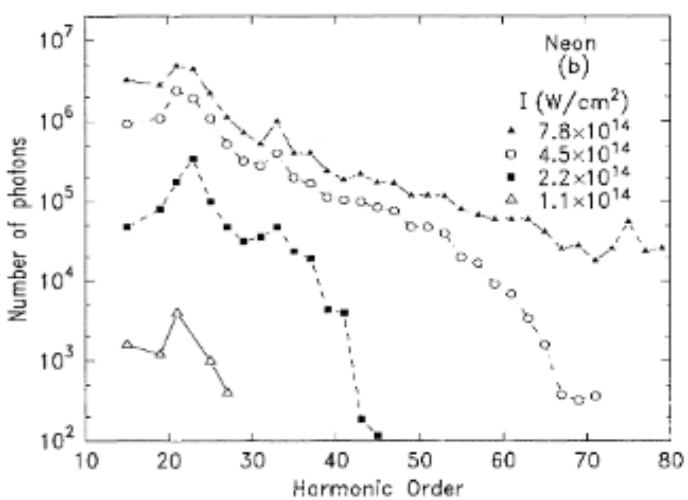
The most characteristic features of HHG: The cutoff law

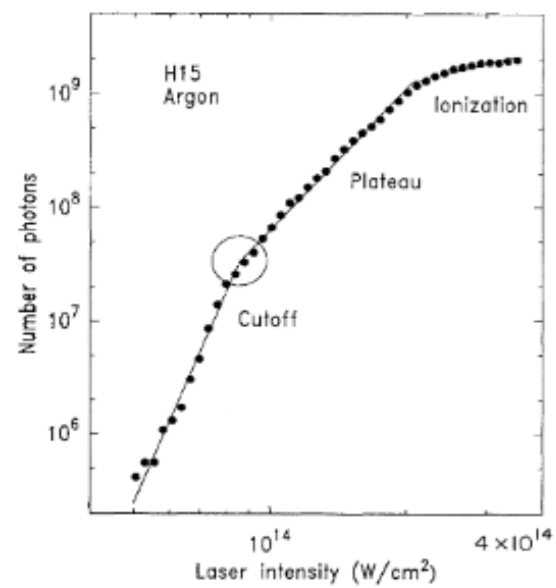


- $q_{max}\hbar\omega = I_p + 3{,}17\ U_p$

$$U_P = \frac{e^2}{8m\varepsilon_0 c^3 \pi^2}\lambda^2 I$$

Gaz considéré	Ip (eV)	I_{BSI} (W/cm^2)
Néon	21,56	8,20E+14
Argon	15,76	2,34E+14
Xénon	12,13	8,23E+13





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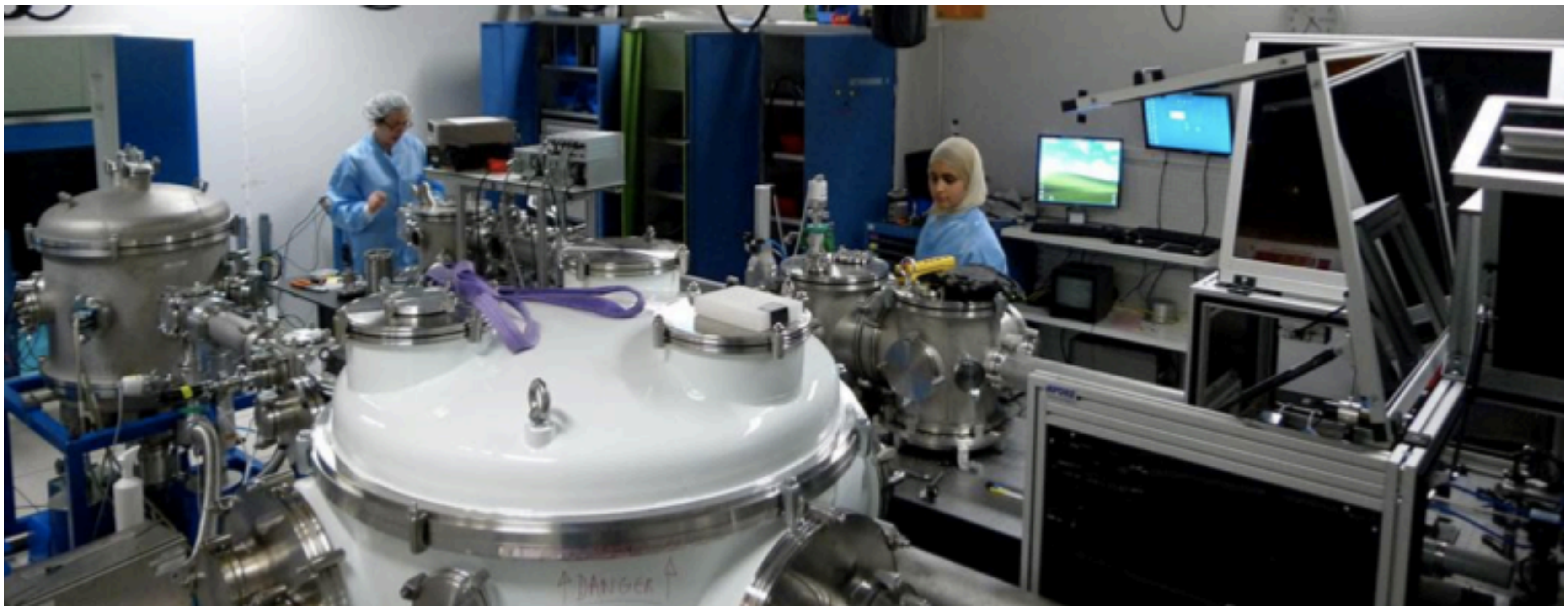
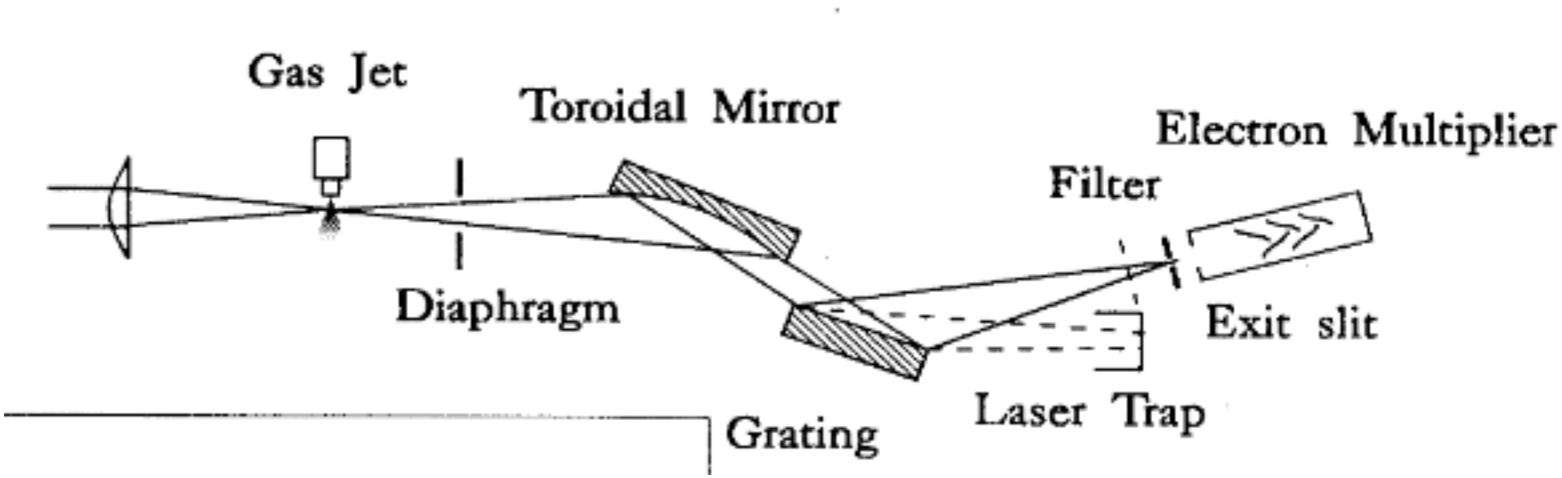
How it looks like experimentally?

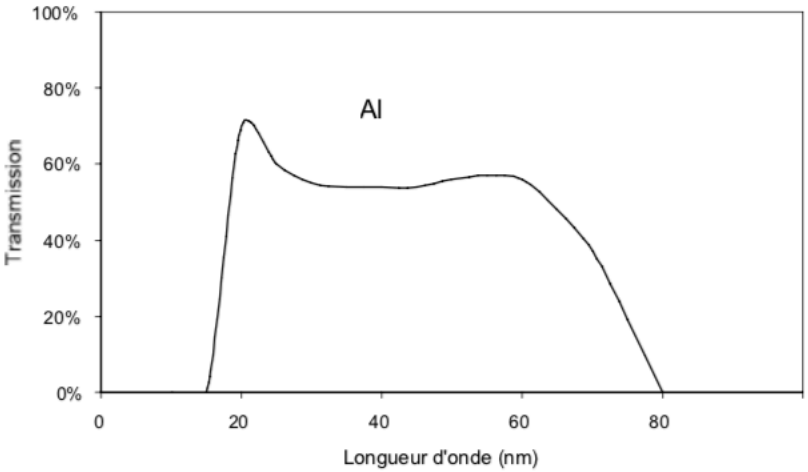


How it looks like experimentally?



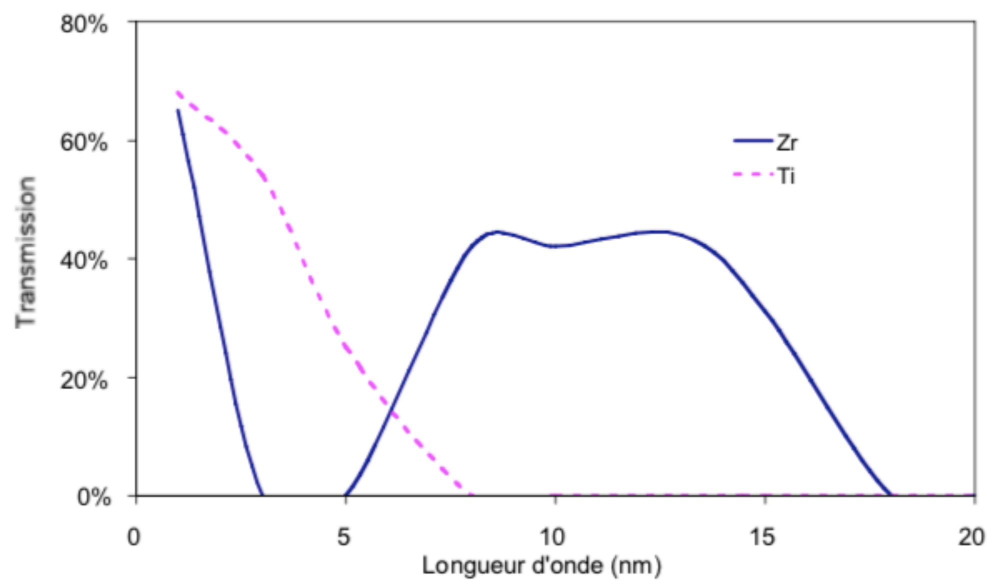
- The gas is a rare gas, it can be a jet, a cell, pulsed or not
- The experiment is fully in vacuum
- The emission is on axis->filter is required
- The laser is focused and apertured (the size of the experiment depends on laser energy)
- Laser intensity is in the range 10^{14} W/cm^2, polarization is linear
- The best way to detect is a EUV spectrometer, microchannel plates, photodiode, CCD
- Efficiency is low: 10^{-4} is the maximum, goes down to 10^{-7} for short wavelengths
- Optical quality is good: low divergence, coherent beams, linear polarization

The most characteristic features of HHG: Filter transmission for 250 nm width



An optical element in the EUV is generally:

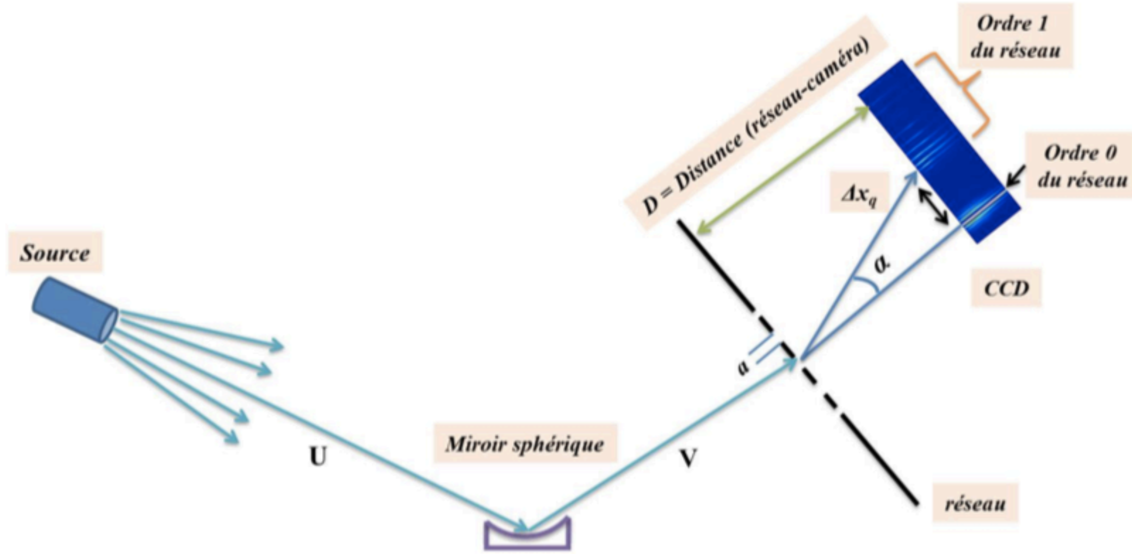
- Expensive,
- Fragile,
- Strongly absorbing

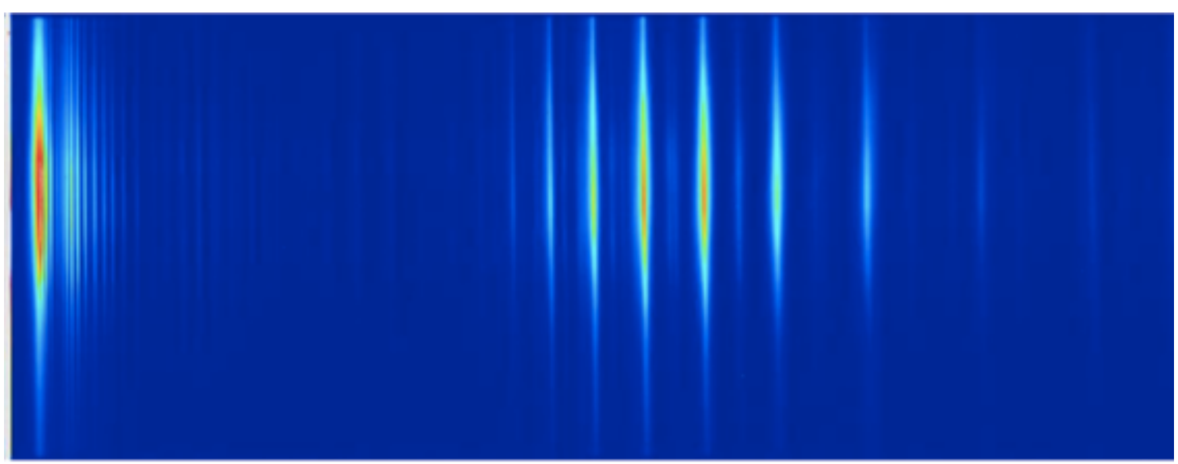


Depending on the target spectrum:

- Multilayer mirrors
- Grazing incidence mirrors

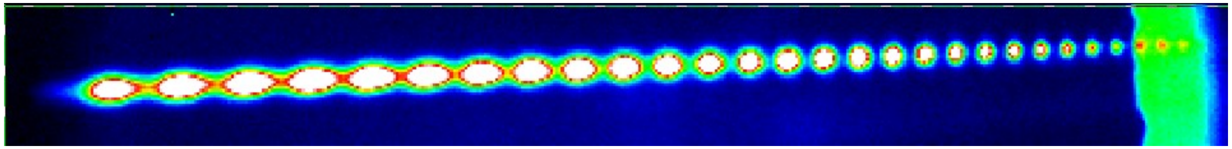
The most characteristic features of HHG: Typical experimental data



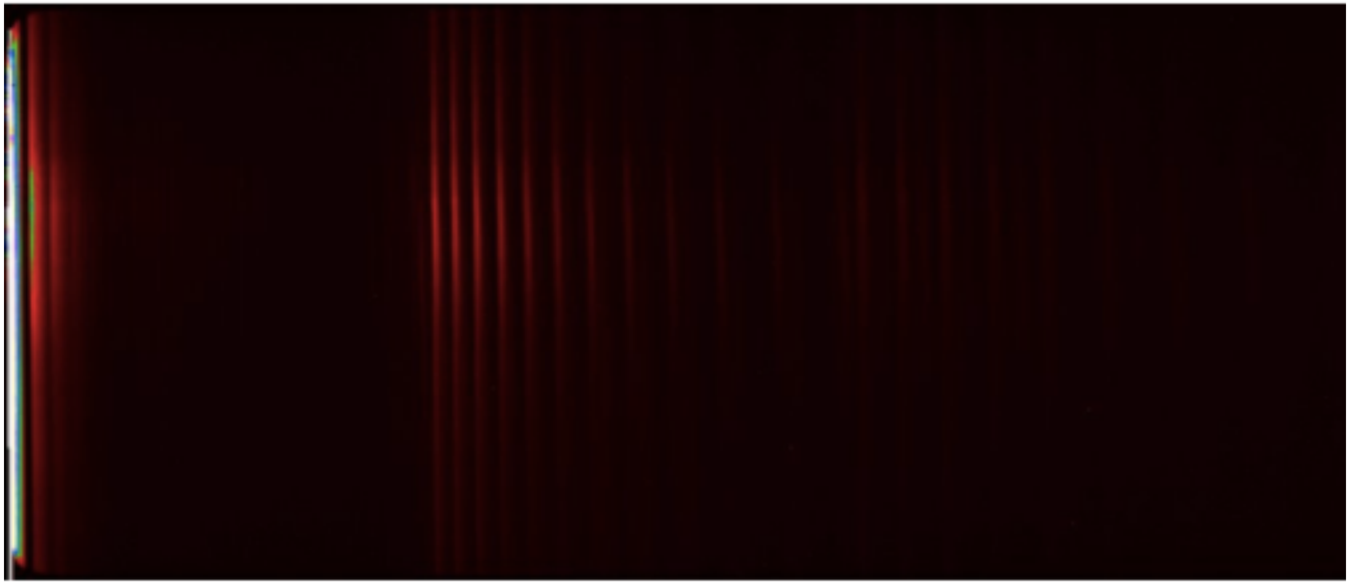


HHG spectrum in argon with a transmission spectrometer

The most characteristic features of HHG: Typical experimental data

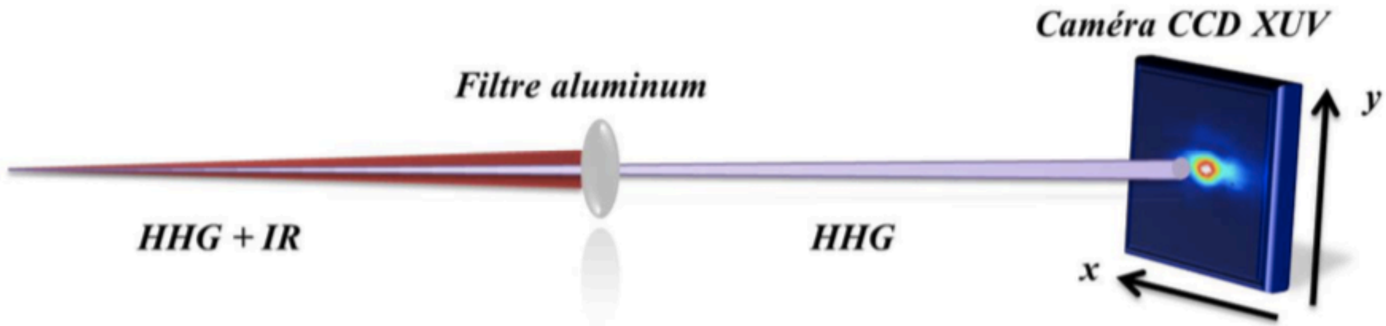


HHG spectrum in neon with an imaging spectrometer

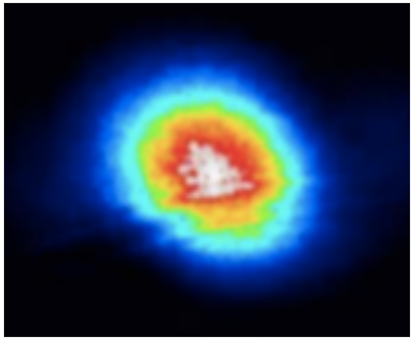


HHG spectrum in neon with a transmission spectrometer

The most characteristic features of HHG: Typical experimental data



HHG footprint after
Few meter
propagation



Gaz	Efficacité de conversion	q_{max}
Krypton	2-5 10^{-5}	21
Xénon	2-5 10^{-5}	21
Argon	10^{-5}	35
Néon	10^{-7}	81
Hélium	10^{-8}	301

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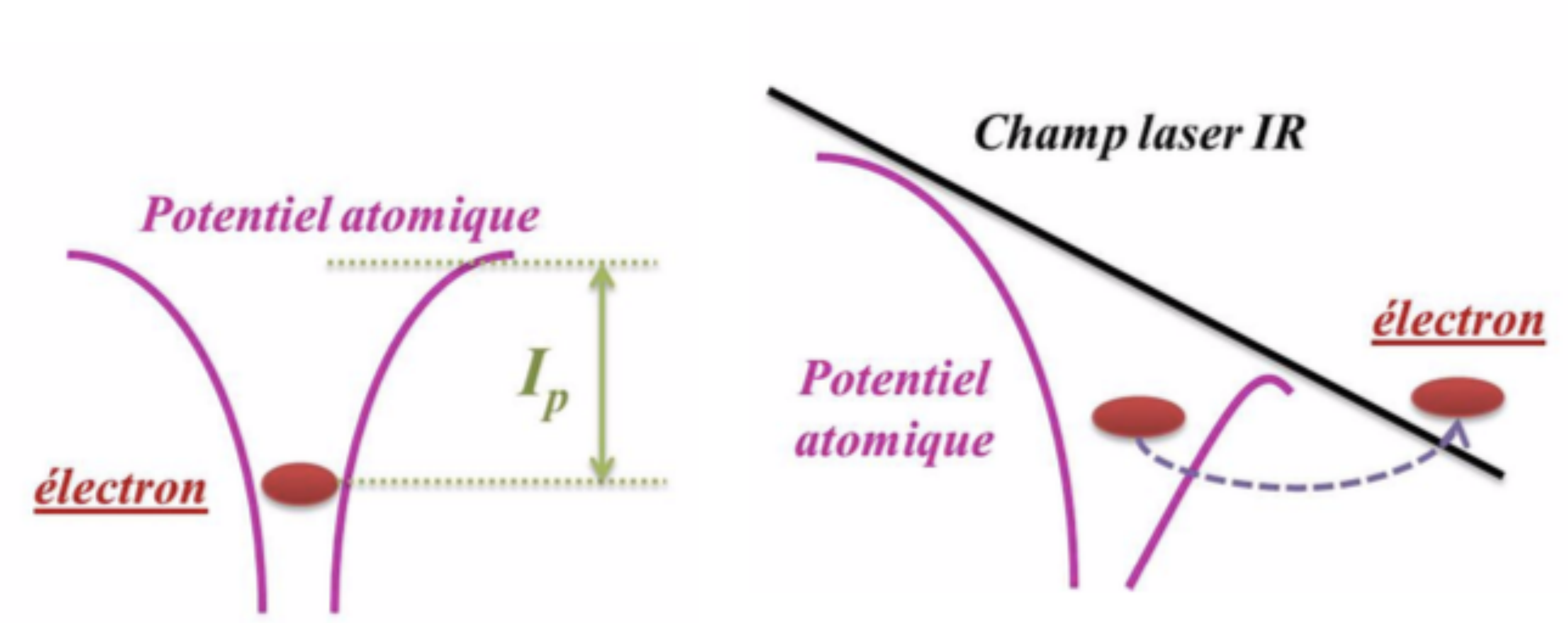
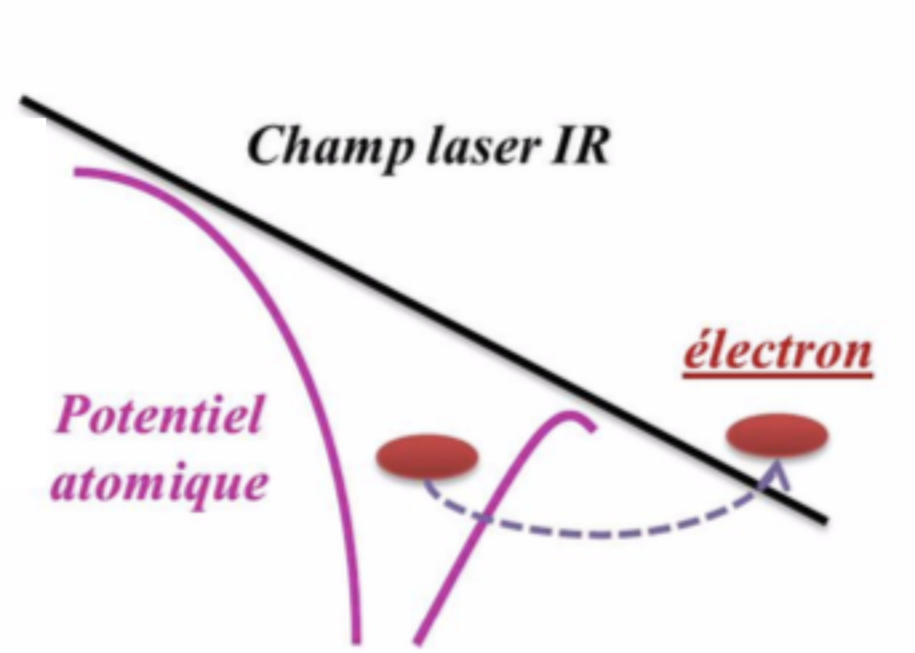
The physical origin of the non linear polarization Of atoms in strong laser fields



- In the bond state the electron energy is $-I_p$
- The atomic potential is $-Ze^2/4\pi\varepsilon_0 r$
- I_p is large as compared to 1 single laser photon energy (it should be multiphoton)
- U_p is large as compared to I_p
- The laser potential is -Er$\cos\omega t$, r being the coordinate in the direction of propagation
- I must stay below the Intensity for barrier suppression (I_{BSI})

The physical origin of the non linear polarization Of atoms in strong laser fields

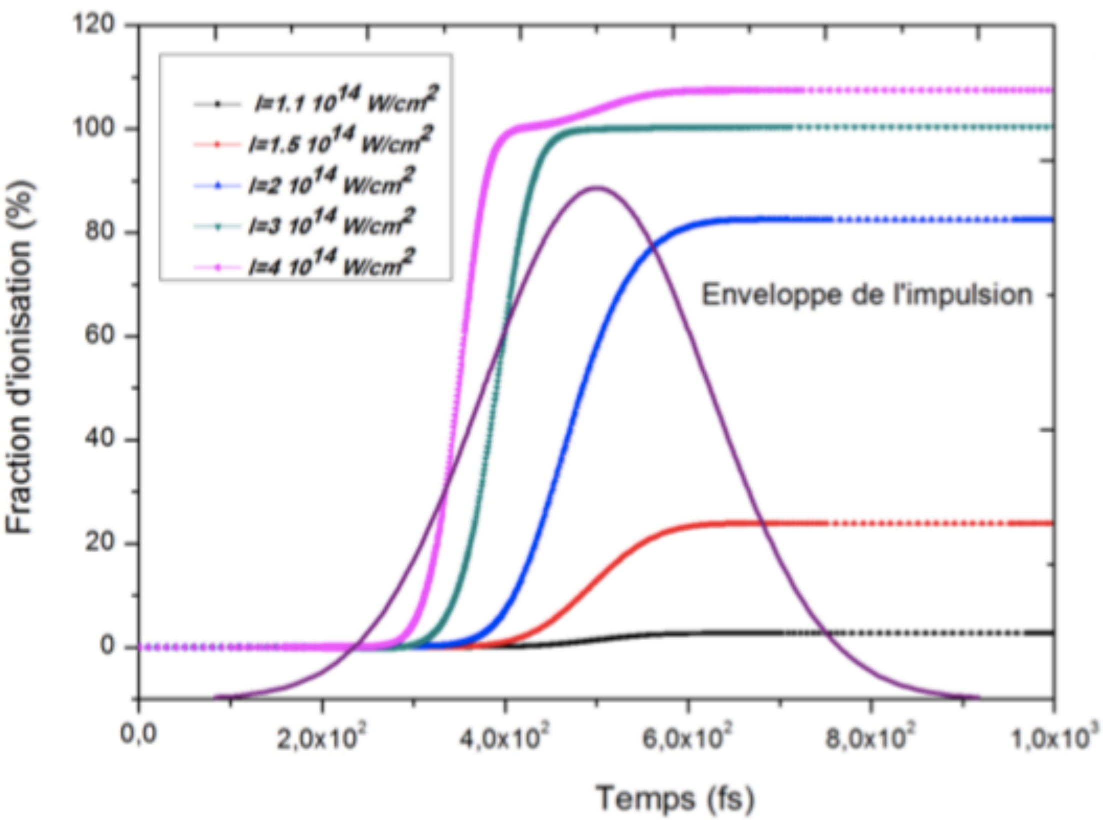
Gaz	I_P (eV)	I_{BSI} (W/cm^2)
Néon	21.56	$8.20\ 10^{14}$
Argon	15.75	$2.34\ 10^{14}$
Xénon	12.13	$8.23\ 10^{13}$



Up and IBSI can be calculated analytically

The physical origin of the non linear polarization Of atoms in strong laser fields

The ADK rates: it is a probability of tunnel barrier transmission per unit of time, analytical formula for the ionization rate as a function
Of E_{laser} (the envelope) for a specific gas species



$$w_{ADK} = \sqrt{\frac{3n^* F}{\pi Z^3}} \frac{F D^2}{8\pi Z} \exp\left(-\frac{2Z^3}{3n^{*} F}\right)$$

avec

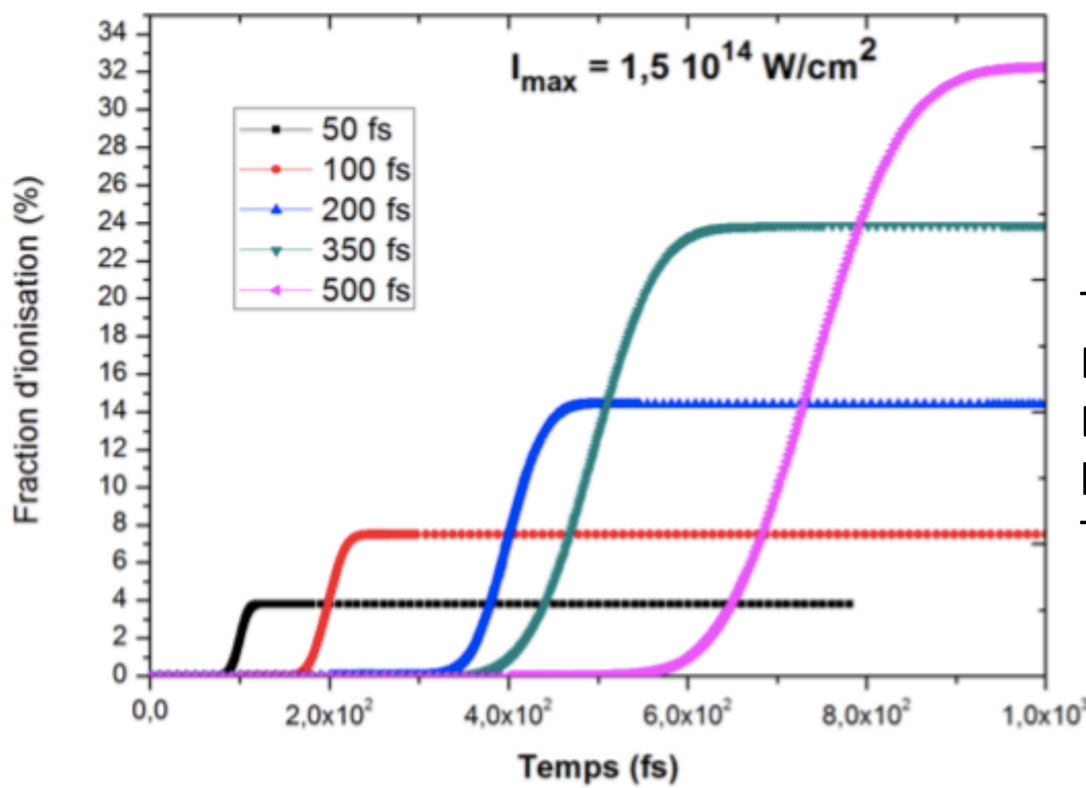
$$n^* = \frac{Z}{\sqrt{2I_p}}$$

$$D = \left(\frac{4eZ^3}{F n^{*4}}\right)^{n^*}$$

Argon, 350 fs

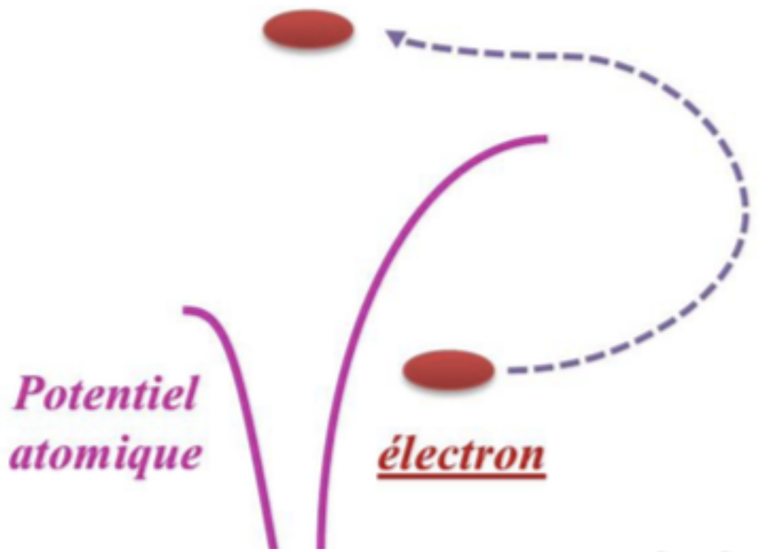
The physical origin of the non linear polarization Of atoms in strong laser fields

The ADK rates: it is a probability of tunnel barrier transmission per unit of time, analytical formula for the ionization rate as a function Of E_{laser} for a specific gas species



TDSE is closer to Reality:
Less approximations but more computation Time…

The physical origin of the non linear polarization Of atoms in strong laser fields



$$m_e a(t) = -eE_0 cos(\omega t)$$

$$E_c = \frac{e^2 E_0^2}{2 m_e \omega^2}(sin(\phi) - sin(\phi_i))^2$$

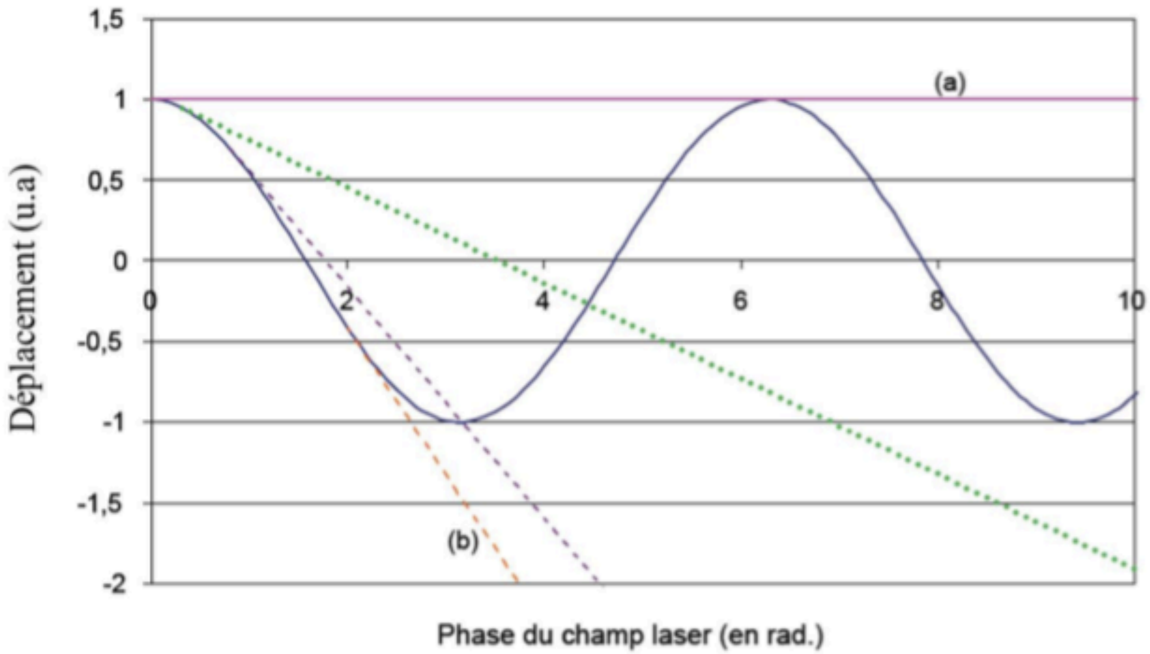
$$V(t, t_i) = -\frac{eE_0}{m_e \omega}(sin(\phi) - sin(\phi_i))$$

$$\frac{E_c}{U_P} = 2(sin(\phi) - sin(\phi_i))^2$$

$$x(t, t_i) = \frac{-eE_0}{m_e \omega^2}(cos(\phi_i) - cos(\phi) + sin(\phi_i)(\phi_i - \phi))$$

The physical origin of the non linear polarization Of atoms in strong laser fields

$$x(t,t_i) = \frac{-eE_0}{m_e\omega^2}(cos(\phi_i) - cos(\phi) + sin(\phi_i)(\phi_i - \phi))$$

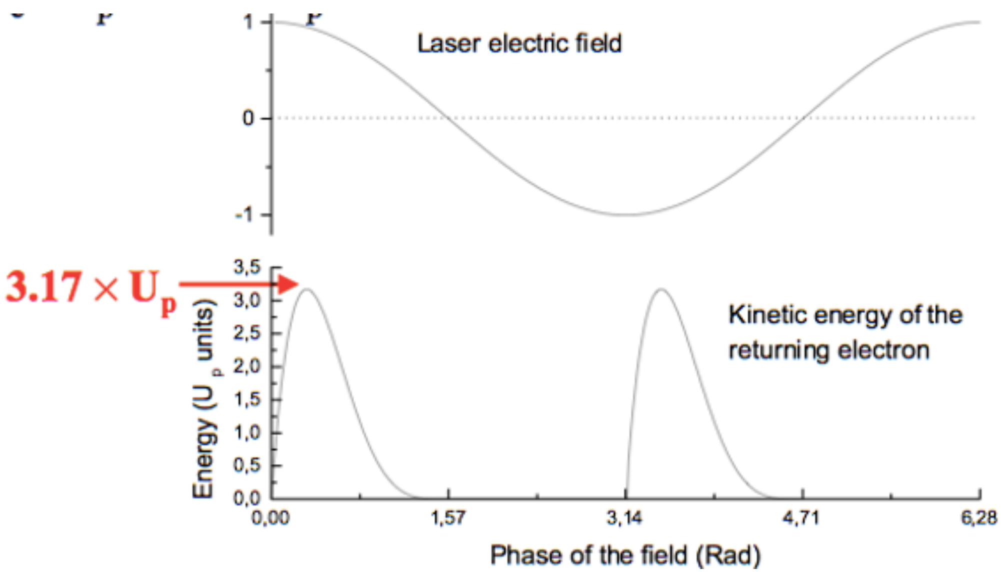


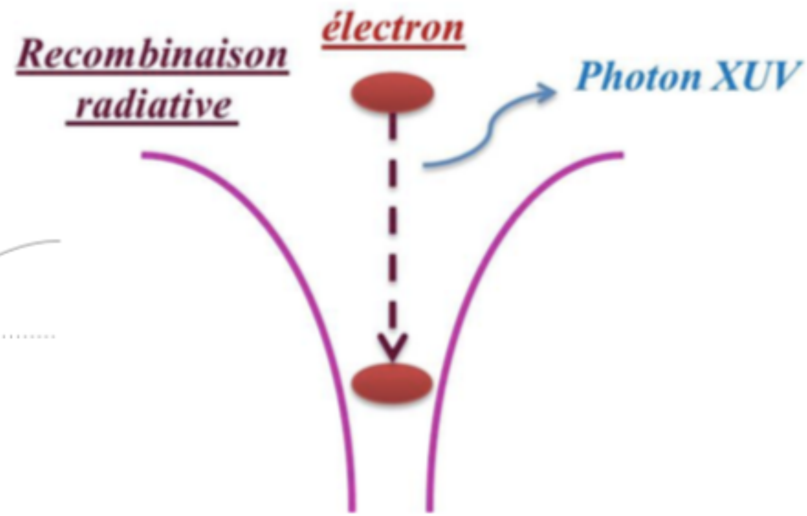
$$E_c = \frac{e^2E_0^2}{2m_e\omega^2}(sin(\phi) - sin(\phi_i))^2$$

$$\frac{E_c}{U_P} = 2(sin(\phi) - sin(\phi_i))^2$$

Two main trajectories for the same Kinetic energy gain :
The long one is emitted earlier and recombine later

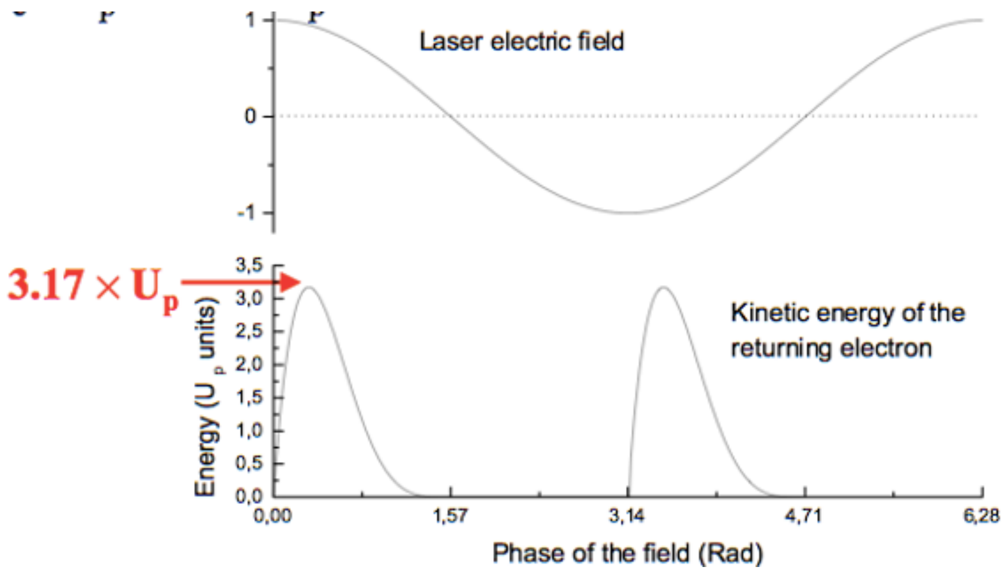
The physical origin of the non linear polarization Of atoms in strong laser fields





The three step model explains the cut-off law
The two trajectories are clearly visible, they converge in the cutoff region

Why is it odd harmonic?



- The harmonic dipole changes Sign every half period:
- This is due to the spherical symetry of the atom and the fact That the dipole is related to The algebric distance to the Nucleus along the polarization
- If one single harmonic burst: no Harmonic structure

$$\tilde{S}(\omega) = \left| \sum_{\tau} E(\omega) \exp i(\omega\tau + \varphi) \right|^2$$

$$\tilde{S}(\omega) = |E(\omega)|^2 |1 - \exp i\omega\tau|^2$$

$$\tau = \frac{\pi}{\omega_{IR}}\ ; q = \frac{\omega_{XUV}}{\omega_{IR}}\ ; \tilde{S}(\omega) = |E(\omega)|^2 (2 - 2\cos q\pi)$$

"Theory of high harmonic generation by low-frequency laser fields", Phys Rev A vol 49, num 3, page 2117, (1994).

$$i\hbar\frac{\partial \mid \Psi(t,x)\rangle}{\partial t} = (-\frac{1}{2}\nabla^2 + V(x) - Ecos(t).x(t)) \mid \Psi(t,x)\rangle$$

Kinetic energy

Atomic potential

Laser energy term

If we assume:

$$\mid \Psi(t,x)\rangle = e^{i\frac{I_p t}{\hbar}}(a(t) \mid 0\rangle + \int d^3 v b(v,t) \mid v\rangle)$$

Inject, calculate and do the scalar product with the state v gives:

$$\frac{\partial b}{\partial t} = -i(\frac{v^2}{2} + I_p)b(v,t) - Ecost\frac{\partial b}{\partial v_x} + iEa(t)costd_x(v)$$

Scalar product with the state 0 gives:

$$\dot{a} = iEcost\int d^3 v d_x(v) b(v,t)$$

$$d_x(v) = \langle v \mid x \mid 0\rangle$$

"Theory of high harmonic generation by low-frequency laser fields", Phys Rev A vol 49, num 3, page 2117, (1994).

$$b(\vec{v},t) = i\int_0^t dt' E cos t' d_x(\vec{v}+\vec{A}(t)-\vec{A}(t'))exp(-i\int_{t'}^t dt''[I_p + \frac{(\vec{v}+\vec{A}(t)-\vec{A}(t''))^2}{2}])$$

We are looking for: $x(t) = \langle \Psi(t) \mid x \mid \Psi(t) \rangle$.

$$(a^*(t)\langle 0 \mid + \int d^3 v b^*(v,t)\langle v \mid) x (a(t) \mid 0\rangle + \int d^3 v b(v,t) \mid v\rangle)$$

After some maths, t' is ionization time, t is recombination time, v=p-A:

$$x(t) = i\int_0^t dt' \int d^3 p E cos t' d_x(p-A(t'))d_x^*(p-A(t))exp(-iS(p,t,t'))$$

$$S(p,t,t') = \int_{t'}^t dt''(I_p + \frac{(P-A(t''))^2}{2})$$

"Theory of high harmonic generation by low-frequency laser fields", Phys Rev A vol 49, num 3, page 2117, (1994).

$$x(t) = i\int_0^\infty d\tau\left(\frac{\pi}{\epsilon + i\frac{\tau}{2}}\right)^{\frac{3}{2}} d_x^*(p_{st}(t,\tau)-A(t))d_x(p_{st}(t,\tau)-A(t-\tau))Ecos(t-\tau)exp(-iS_{st}(t,\tau))+c.c.$$

Integration is done over all times spent in the continuum
With:

$$p_{st}(t,\tau) = \frac{E}{\tau}[cos(t) - cos(t-\tau)]$$

$$S_{st}(t,\tau) = \int_{t-\tau}^{t} dt" \, \frac{(p_{st} - A(t"))^2}{2} + I_p.$$

$$S_{st}(t,\tau) = (I_p + U_p)\tau - \frac{2U_p}{\tau}(1 - cos\tau) - U_p C(\tau) cos(2t - \tau)$$

$$C(\tau) = sin(\tau) - 4\frac{sin^2(\frac{\tau}{2})}{\tau}$$

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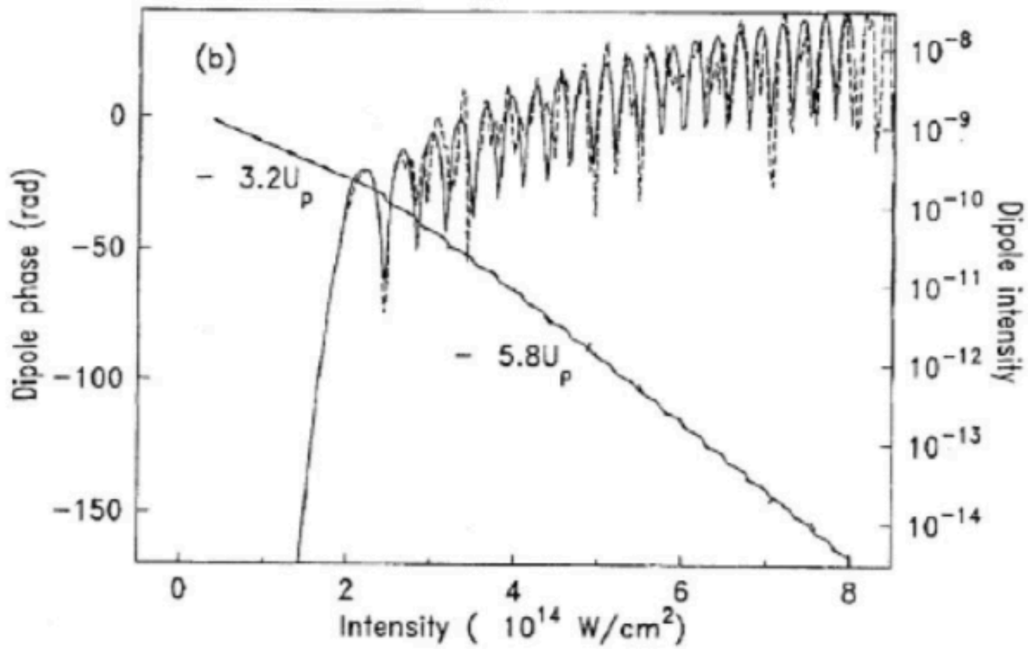
$$x(t) = i\int_0^\infty d\tau\left(\frac{\pi}{\epsilon + i\frac{\tau}{2}}\right)^{\frac{3}{2}} d_x^*(p_{st}(t,\tau) - A(t))\, d_x(p_{st}(t,\tau) - A(t-\tau))\, E cos(t-\tau) exp(-iS_{st}(t,\tau)) + c.c.$$

The Fourier transform of x(t) gives the harmonic spectrum in amplitude and phase



H45 in neon

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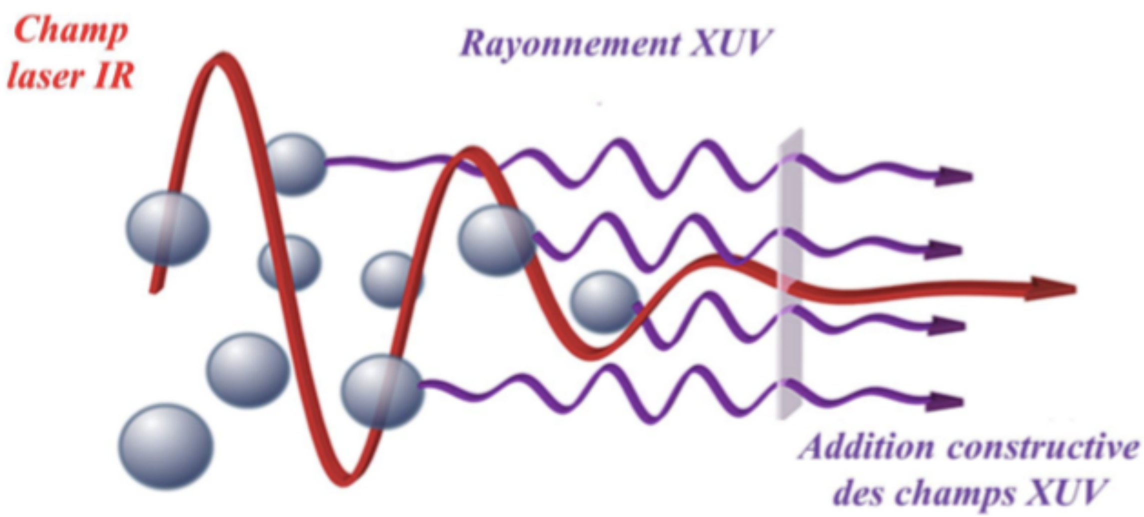
Macroscopic aspect of HHG:



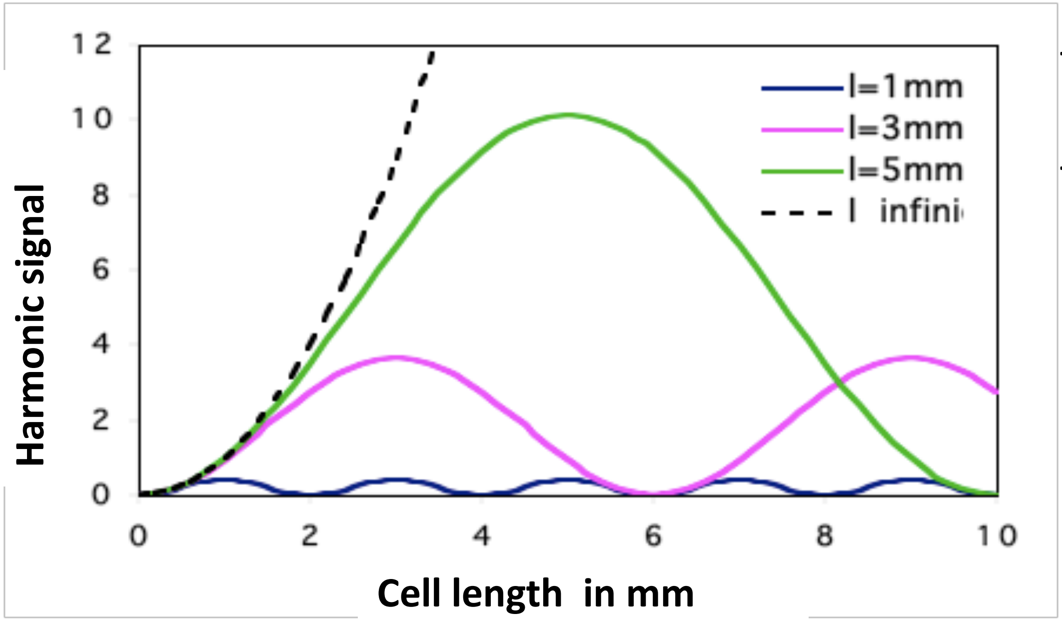
Each individual harmonic dipole is driven by the laser with its own amplitude and phase
It then propagates until the end of the medium with some delay
The total number of photons produced is given by the coherent sum of all dipoles
->there are constructive or destructive interferences following the phases between the dipoles

$$\nabla^2 E(\omega) + n^2(\omega)\frac{\omega^2}{c^2}E(\omega) = \frac{-\omega^2}{\varepsilon c^2}P^{NL}(\omega)$$

$$E_q \propto \int_0^{l_{med}} \rho |d_q(z)| e^{i\phi(z)} dz$$ with $$\phi(z) = (k_q - qk_{IR})z - \phi_{at,k}$$

Macroscopic aspect of HHG:

If $\varphi(z)$ *is linear* $= \delta k.z$
We introduce $l_{coh} = \pi/\delta k$



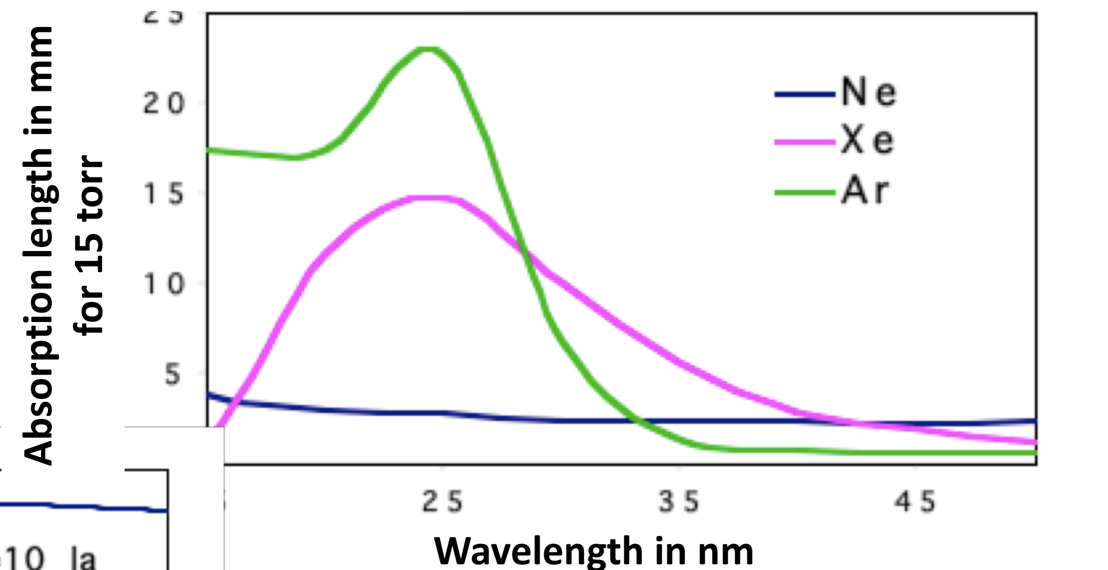
Phase matching means:
L_{coh} infinite

The signal growth is quadratic with
The number of emitters (Pressure)

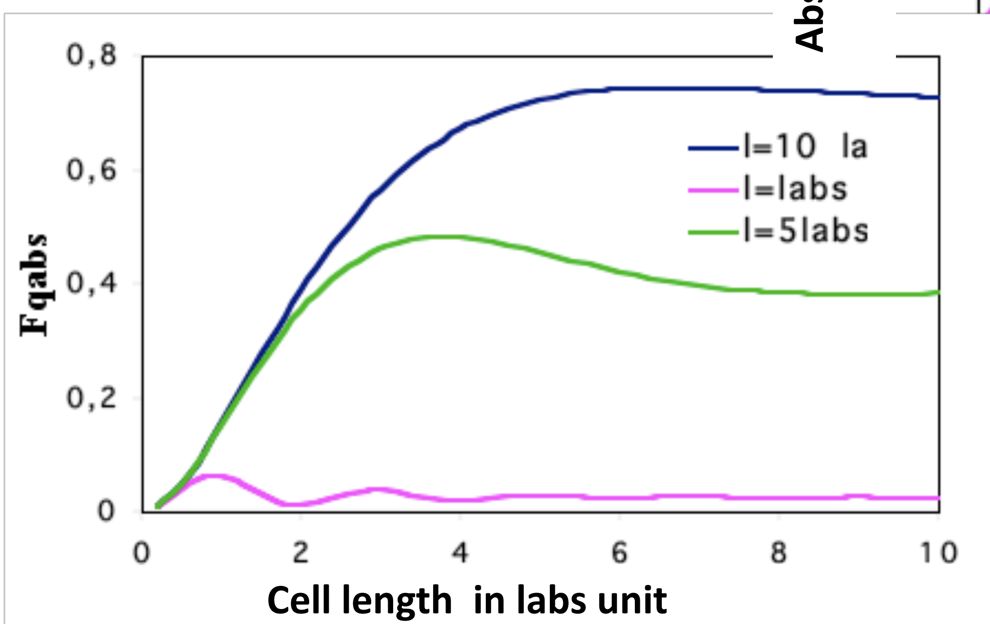
This is the sign of coherent
effect

The absorption limit:

$$N_{out}(t) \propto \left| \int_0^{l_{med}} |dq(z)| \exp\left(\frac{z - l_{med}}{2l_{abs}}\right) \exp(i\,\varphi(t,z))\,dz \right|^2$$



L_{abs} varies as 1/P
T=$e^{-l/l_{abs}}$



Saturation of the signal is obtained
When: $l_{med}>3l_{abs}$ and $l_{coh}>5l_{abs}$

The only way to increase further is transverse

Constant et al., PRL 82,1668 (1999)

General formula with absorption:

$$N_{photons} \propto \frac{l_{abs}^2}{1 + 4\delta k^2 l_{abs}^2}\left(1 + e^{\frac{-l_{med}}{l_{abs}}} - 2cos(\delta k l_{med}) e^{\frac{-l_{med}}{2l_{abs}}}\right)$$

What is the origin of dephasing?

If the propagation phase can be considered linear: $\delta k = k_q - qk_1$-K_{at}

k=$\frac{n\omega}{c}$, n=$\sqrt{1 - \frac{n_e}{n_c}}$, K_{at} is the gradient of atomic phase

If the beam can be considered gaussian:

$$(n_c = \frac{\omega^2 m_e}{\mu_0 e^2 c^2})$$

$$\delta k = \frac{q\omega}{c}\left(\frac{n_{elec}}{2n_c} - \delta n_{at}\right) + \frac{\frac{q}{z_0}}{1 + (\frac{z}{z_0})^2} - \left(\frac{2\alpha I_0 z/z_0^2}{1 + (z/z_0^2)^2}\right)$$

What is the origin of dephasing?

- Role of the Gouy phase term, balance with

Atomic dispersion if ionization is negligible

$$l_{\text{coh}}(\eta) = \frac{\pi/q}{\frac{1}{z_0} - 10^{-3} P\,(1.66 - 7366\eta)}$$

- This is particularly

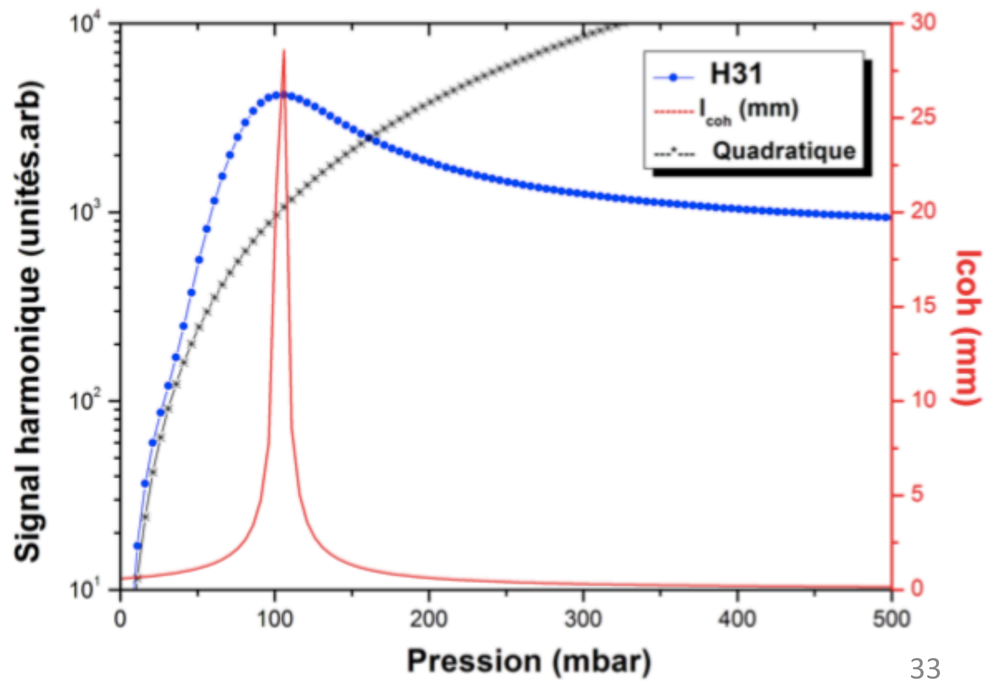
Relevant for high rep rate

Lasers: Strong focussing

- The approx of linear

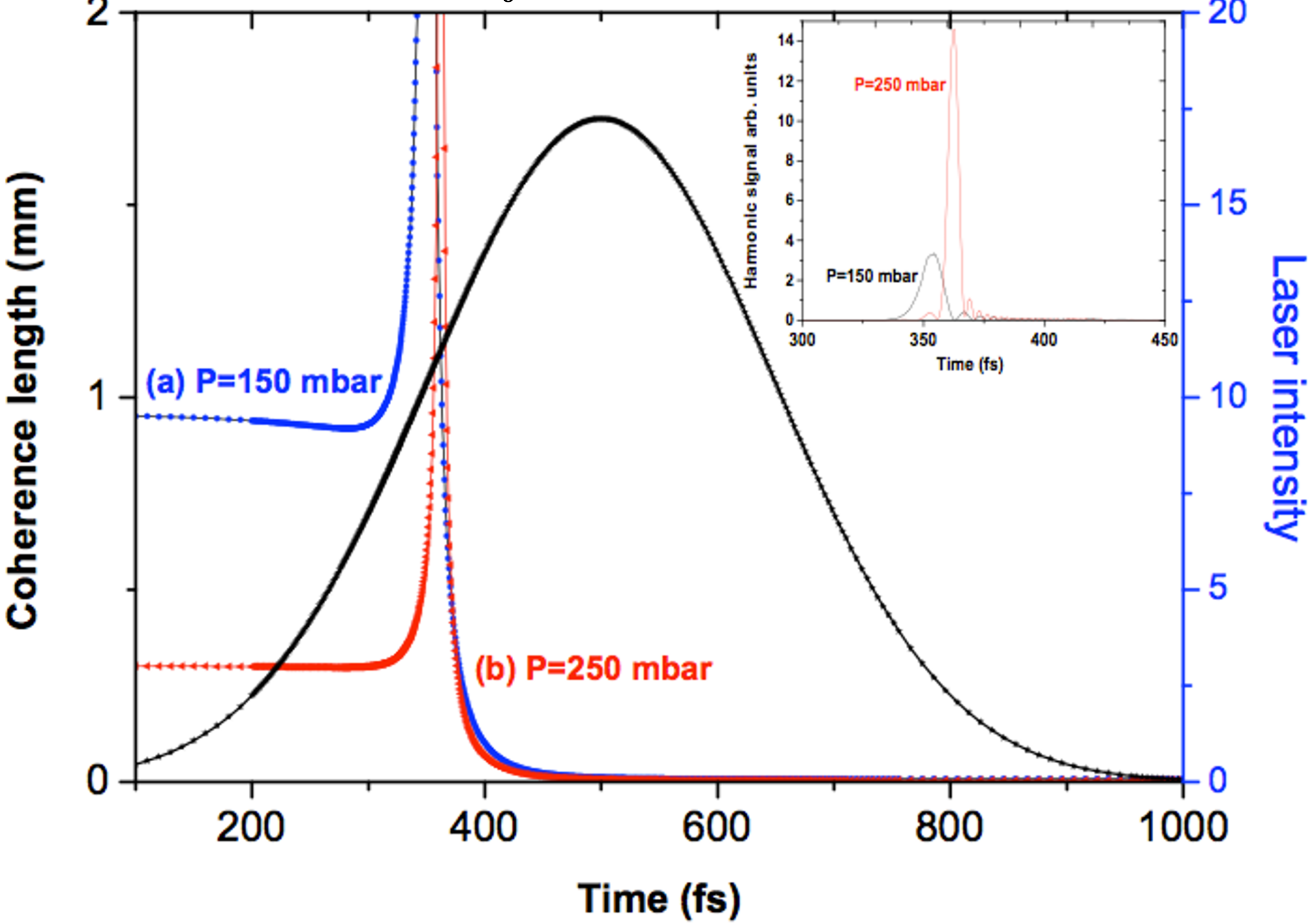
Phase might not be so

good

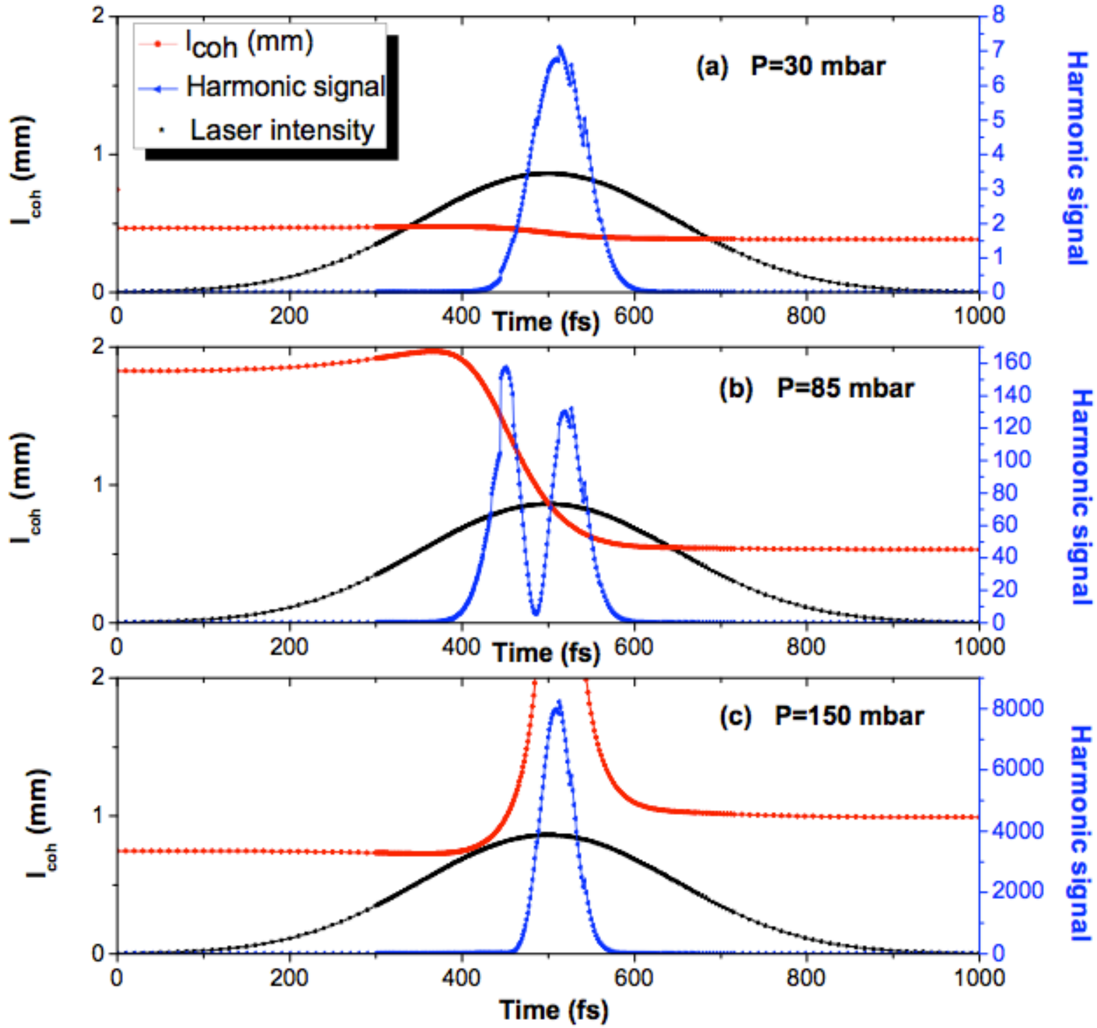


The coherence length is time dependant!

$$l_{\text{coh}}(\eta) = \frac{\pi/q}{\frac{1}{z_0} - 10^{-3}P\,(1.66 - 7366\eta)}$$



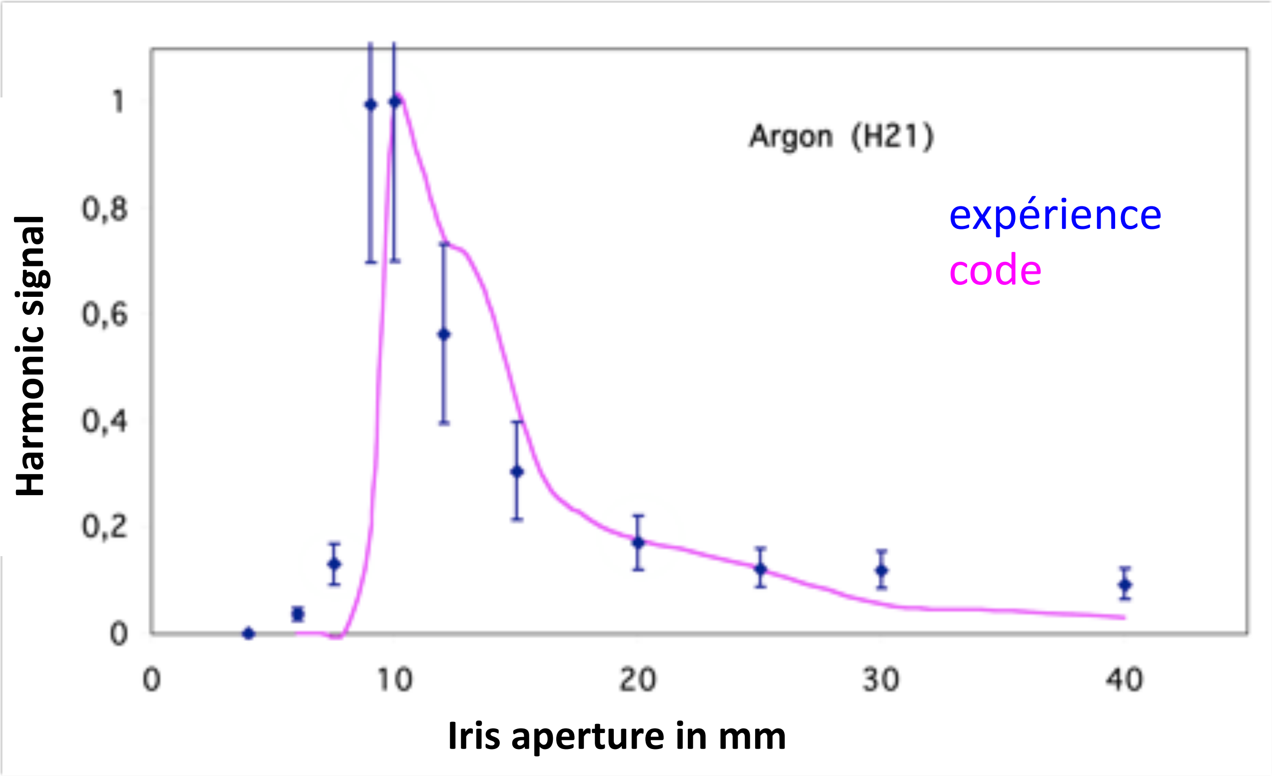
The coherence length is time dependant!



In theory,
the HHG signal reaches
Zero when the coherence
Length is exactely half
Of the medium length

The loose focussing geometry

When the laser energy is high enough: the best is to Increase the Rayleigh range as much as possible, this Increases the volume, the ionization rate for phase Matching, the cell length

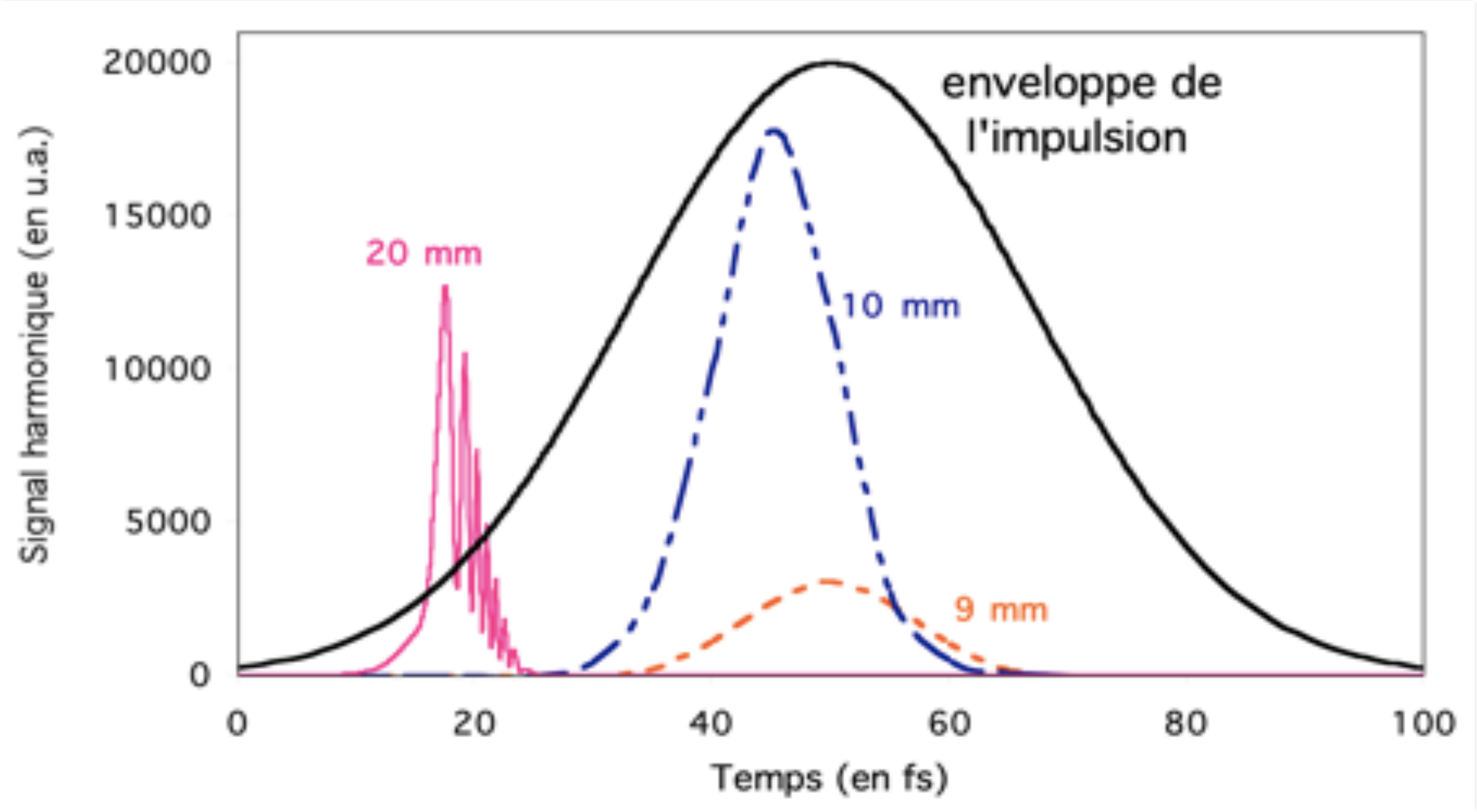


Optimum for z_0=17mm

E_{laser}=6 mJ, f=1m

(Cell length:2mm, pressure 10 torr, at focus)

What happens in time?



Each HHG source has to be designed following the pump laser characteristics

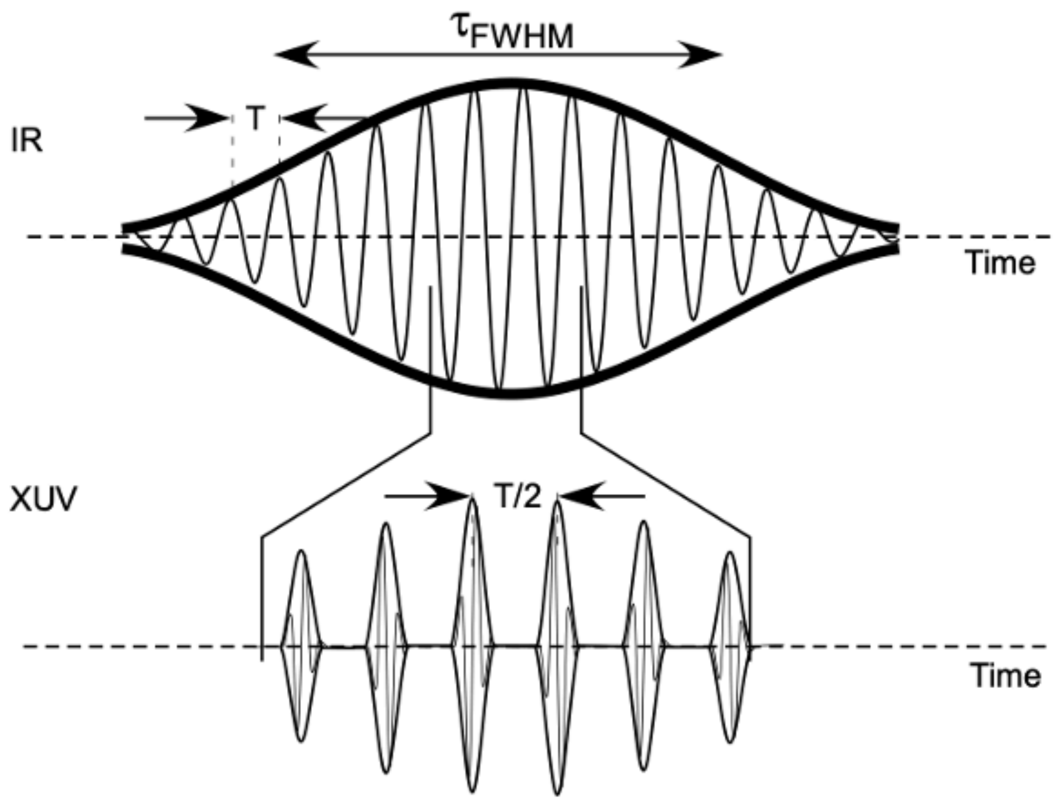
Depending on the harmonic you want:
->You choose the gas
->You calculate the focussed intensity you need
->You obtain the focussing geometry
->This gives you the medium length and optimum pressure
->You do the phase matching optimization to reach the typical Conversion efficiency (for each gas)

- The most efficient lasers are as short as possible, with short λ
- The mid IR ones are good for high energy photons, are generally tunable but phase matching is harder to reach in the presence of ionization

Plan of the lecture:

- The most characteristic features of HHG, historical aspects
- How it looks like experimentally?
- The physical origin of the non linear polarization of atoms
- The problem of phase matching
- An ultrashort story of the attosecond structure
- Modern trends (not exhaustive and subjective)

Origin of the attosecond structure for HHG :



The wider the spectrum, the shorter the pulse envelope

Origin of the attosecond structure for HHG :

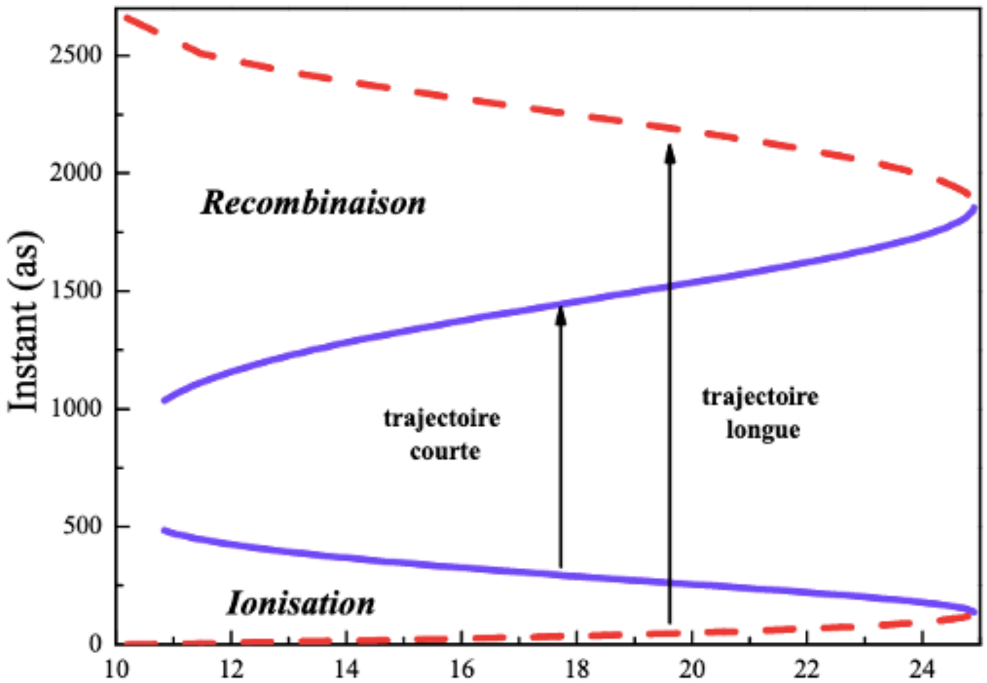


Figure 16: Ionization and recombination times as a function of the harmonic order for a generation in argon at 1.2 10^{14} W/cm^2, in blue the first quantum path, in red the second one, the cutoff is clearly visible (Ph.D. thesis of Yann Mairesse).

How to measure the pulse duration: The Rabbitt method

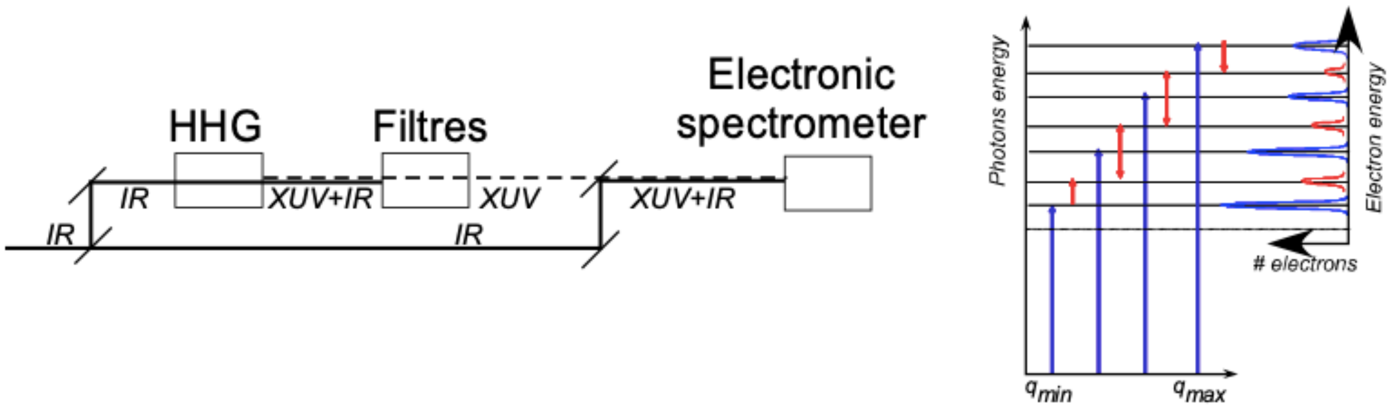


Figure 11: *Principle of the RABBITT technique. (left) The laser IR beam is split into two parts: one used for HHG, the other for dressing. Filters are used to remove the remaining IR after HHG. (Right) Cartoon of the spectra obtained. Sidebands show up right in between odd harmonics electrons coming from XUV photoionization by the attosecond pulse (taken from a review paper by Thierry Ruchon).*

$$2\omega\tau + \Psi_{q'+1} - \Psi_{q'-1}$$

How to measure the pulse duration: The Rabbitt method

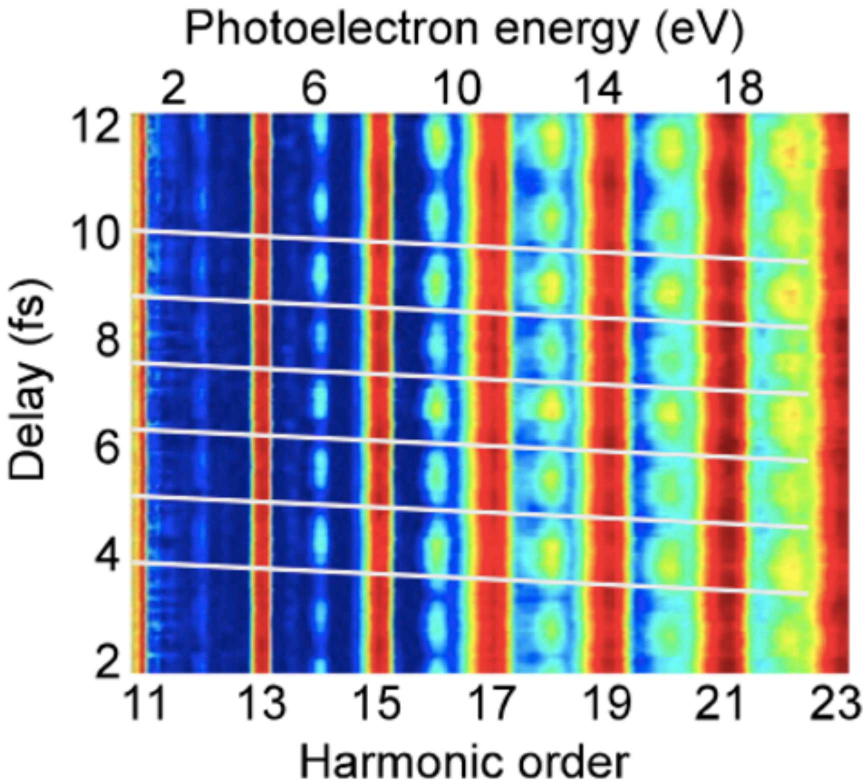
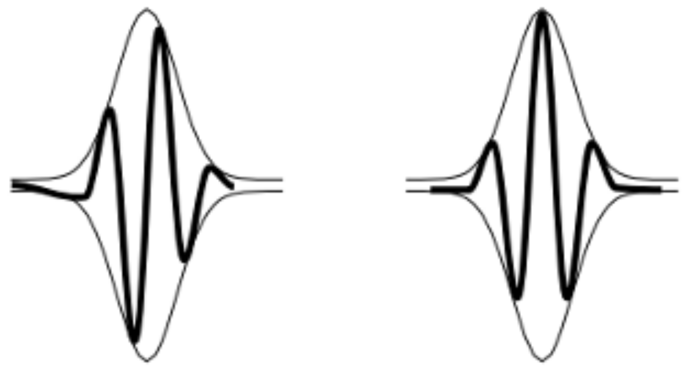


Figure 20: Example of a RABBITT trace taken in Ar for both the generating and detecting gas. The minima in the sidebands vs the delay drift from one sideband to the next signaling a lack of synchronization of the harmonics (superimposed white lines). This figure is taken from a review paper by Th. Ruchon

Single attosecond pulses



$e^{-t^2/\tau^2} \sin \omega_0 t$ $e^{-t^2/\tau^2} \cos \omega_0 t$

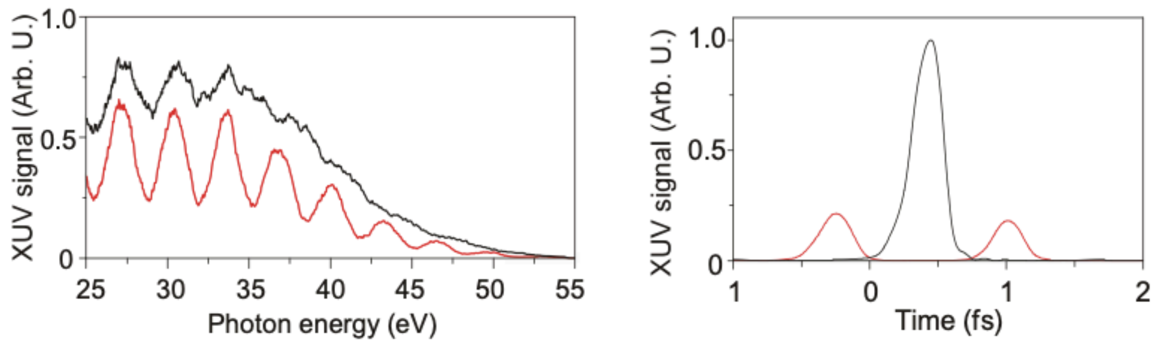


Figure 18: Effect of a change of the CEP on the spectrum (left panel) and corresponding temporal profile (right panel) for an attosecond source driven by a polarization gate field (from [Sola 2006]).

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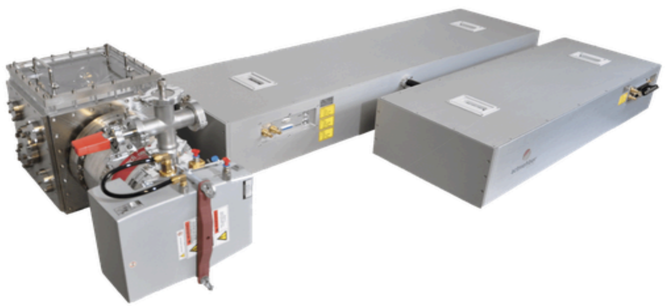
Commercial sources:

Generally high rep rate, quite compact:

Products **Applications** **Company** **Career** **Publications** **Contact**

High-power XUV beam lines



Sources of short-wavelength radiation, such as synchrotrons or free-electron lasers, have already enabled numerous applications and will facilitate more seminal studies. On the other hand, sources of coherent extreme ultraviolet to soft x-ray radiation via high-harmonic generation (HHG) of ultrashort-pulse lasers have gained significant attention in the last years due to their enormous potential to address a plethora of applications in a cost-effective and tabletop format. Therefore, they constitute a complementary source to large-scale facilities. The photon-flux values obtained by fiber-laser-driven HHG sources can be considered the highest of all laser systems for photon energies between 20 eV – 150 eV. Even higher photon energies up to the soft X-ray regime are feasible using Tm-based driving lasers.

AFS ultrafast fiber lasers are ideal high-harmonic drivers. These turnkey HHG beamlines can address several applications in the EUV to X-ray spectral region.

Applications

- Photoelectron spectroscopy
- Coherent diffractive imaging (CDI) nanoscope / XUV imaging
- Attosecond science
- Pump-probe experiments

Commercial sources:

Generally high rep rate, quite compact:

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The following specs show only our exemplary main platforms. We happily customize a system exactly to your needs.

	Examplary configurations		
Photon energy	21 eV	90 eV	150 eV
Wavelength	59 nm	13 nm	8.5 nm
Photon flux per harmonic	up to 10^{14} s^{-1}	up to 5×10^{10} s^{-1}	up to 10^{10} s^{-1}
Average power per harmonic	up to 330 μW	up to 0.7 μW	up to 0.4 μW
Repetition rate	flexible, up to 10 MHz		
Pulse duration	pulse duration < laser pulse duration i.e. < 30 fs (or shorter)		
Spectral bandwidth	can remain close to the transform limit with flexible bandwidths (down to < 10 meV)		
Beam profile	Gaussian		
Dimensions of HHG chamber	80 cm × 40 cm × 40 cm		

Specific original sources:

- Tunable
- Circularly polarized
- Crystal target HHG for single attosecond compact sources