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ATTOSECOND PHYSICS

UNIT VIII HIGH HARMONIC GENERATION

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Ist Semester 2024, Buenos Aires, Argentina

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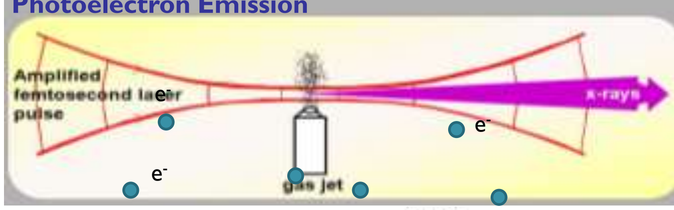
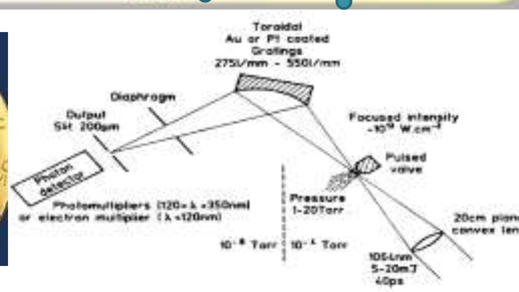
HIGH HARMONIC GENERATION

High-Harmonic Generation: Anne L'Huillier (1987)

Photoelectron Emission



Paris-Saclay



Experimental setup at Saclay to measure the light emitted during above-threshold ionization experiments

A. L'Huillier et al., in 'Atoms in Intense Laser Fields', edited by Gavrila and Muller, (Academic Press, 1992)

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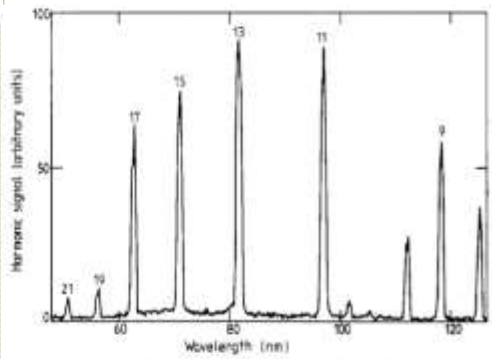

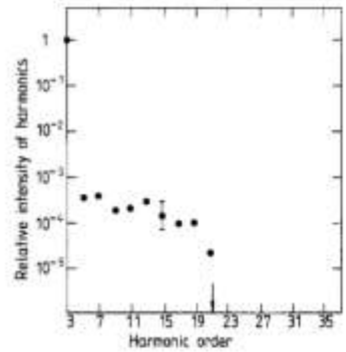

High-harmonic generation in Xe using a 30 ps, 1064 nm laser focused to ca. 10^{13} W/cm^2

M. Ferray et al., J. Phys. B 21 L31 (1988)

+ similar observations around the time in the Rhodes-group using 248 nm driver lasers

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HIGH HARMONIC GENERATION

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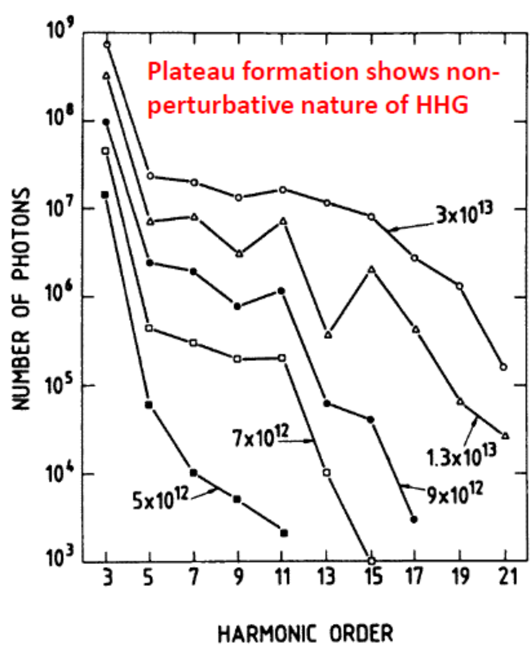
A. L´Huillier et al.,
in ‘Atoms in Intense Laser Fields’,
edited by Gavrila and Muller,
(AcademicPress, 1992)

Plateau formation shows non-perturbative nature of HHG



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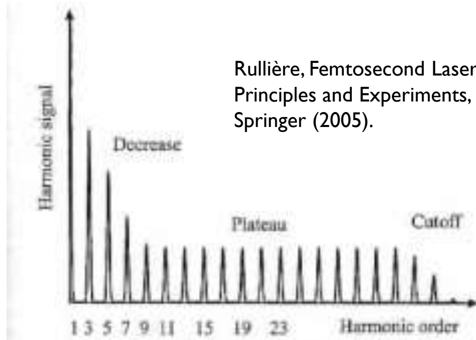
HIGH HARMONIC GENERATION

The emission of radiation occurs when a linearly polarized ultra-short electric field is focalized onto an atomic or molecular gas target.

The maximum kinetic energy of the photon (ii) is: $\hbar\omega_c \simeq 3U_p + I_p$



Rullière, Femtosecond Laser Pulses: Principles and Experiments, Chap. XIII, Springer (2005).

• The radiation spectrum consists of lines separated by 2ω and only the odd orders are present $\omega_q = q\omega = (2n + 1)\omega$.
• The efficiency decays fast for low orders (perturbative), followed by a plateau (non-perturbative) and then decays also very fast from a cutoff frequency ω_c .

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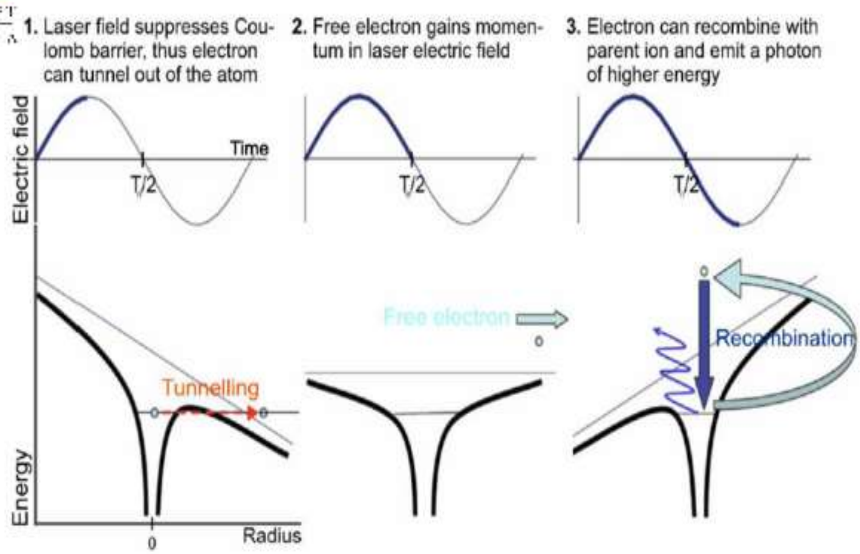
THE THREE-STEP MODEL

P. B. Corkum, *Phys. Rev. Lett.* **71**, 1994 (1993).



THE THREE-STEP MODEL (cont.)

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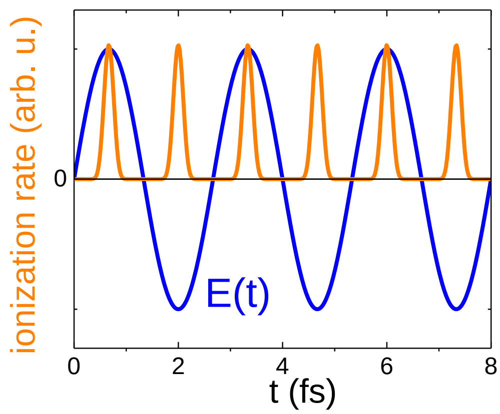
Semiclassical Model: Paul Corkum 1994

1. Tunneling ionization at time t_0: The ionization probability was calculated by Ammossov, Delone and Krainov.

$$w_{dc} \propto \exp\left[\frac{-2(2I_p)^{3/2}}{3E(t_0)}\right]$$



Ionization takes place during a fraction of a femtosecond: **hundreds of attoseconds**

THE THREE-STEP MODEL (cont.)

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Semiclassical Model: Paul Corkum 1994

2. After ionization, electron motion is subject to the external electric field. The core potential is neglected. When the electric field changes sign, the electron eventually can go back near the core.

$$\dot{z}(t) = k + A(t) = A(t) - A(t_0)$$

$$z(t) = \int_{t_0}^{t} A(t')dt' - A(t_0)(t - t_0)$$

$$z(t) = \alpha(t) - \alpha(t_0) - A(t_0)(t - t_0)$$

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THE THREE-STEP MODEL (cont.)

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3. When the electron collides with the parent ion, it can (i) rescatters elastically or (ii) recombine. In (i) there is emission of electrons, whereas in (ii) emission of radiation.

The recombination condition is $z(t_r) = z(t_0) = 0$

$$\Rightarrow \alpha(t_r) = \alpha(t_0) + A(t_0)\left(t_r - t_0\right)$$

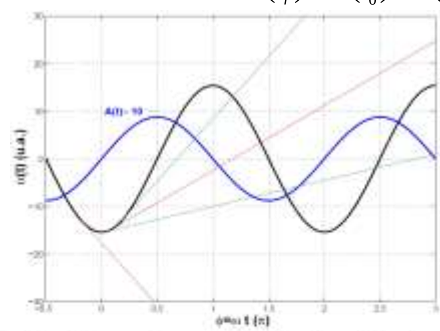

Fig. 2.2: Schématisation graphique pour déterminer les conditions de phase et de moment canonique des trajectoires directes et rediffusées. Le potentiel vecteur est représenté en bleu et $\alpha(t)$ en noir (le champ électrique étant directement proportionné à $\alpha(t)$). Les lettres D et I indiquent les domaines respectifs des trajectoires directes et indirectes sur une demi-période laser.

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THE THREE-STEP MODEL

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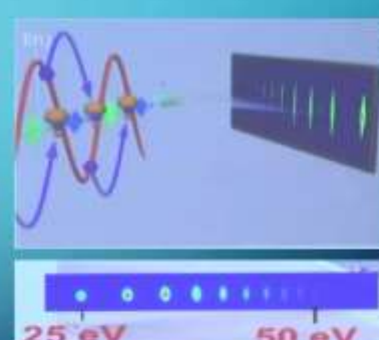
HIGH HARMONICS AND ATTOSECOND TRAIN PULSES

Attosecond pulses repeated each half period create a series of femtosecond duration harmonics. They can extend to more than 1kev if needed



ARGON ATOM AT 800nm

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THE THREE-STEP MODEL (Cont.)

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We can find the kinetic energy at recolission time as $T_{kin}(t_0)$ and $T_{kin}(t_r)$:

$$T_{KIN} = \frac{\dot{z}^2(t_r(t_0))}{2}$$

de Bohan, Ph.D. thesis (2005).

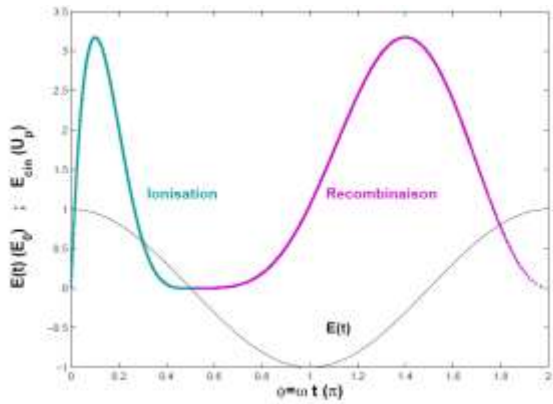

FIG. 2.4: Energie cinétique des électrons revenant vers le noyau reproduite en fonction de la phase d'éjection $\phi_i = \omega t_i$ en vert et dans la phase de "recombinaison" (de retour) en violet $\phi_r = \omega t_r$. L'énergie est représentée en unité de U_p. L'amplitude du champ est également représentée en noir normalisée en fonction de sa valeur maximale E_0.

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THE THREE-STEP MODEL (cont.)

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Semiclassical Model: Paul Corkum 1994

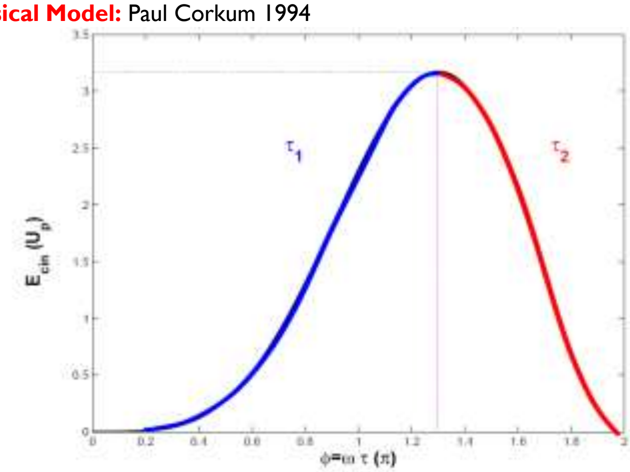
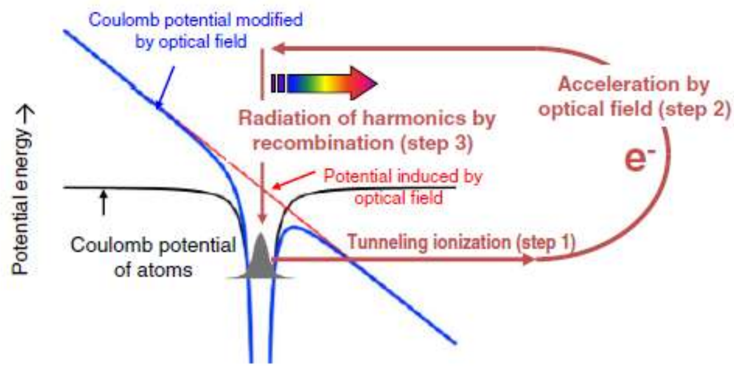


FIG. 2.5: Energie cinétique acquise par les électrons à leur retour au noyau en fonction de leur phase caractéristique correspondant au temps de retour des électrons, $\phi = \omega\tau = \omega \times (t_r - t_i)$. Une même énergie cinétique correspond à deux temps de retour sur une demi-période conventionnellement notés τ_1 (temps de retour le plus court) et τ_2 (temps de retour le plus long).

THE THREE-STEP MODEL (cont.)



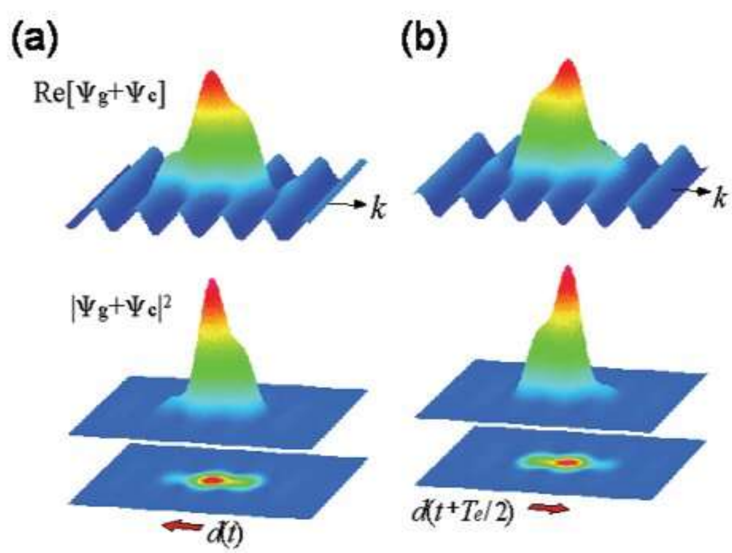
The emission of radiation occurs when a **linearly polarized** short electric field is focalized onto an atomic or molecular gas target.
An elliptical polarized laser produces neither HHG nor rescattering

$$\varepsilon = \hbar\omega_q = T_{\text{kin}} + I_p$$

The maximum kinetic energy of the photon (ii) is: $\hbar\omega_c = 3.17U_p + I_p$

QUANTUM HHG

Simulation of wave function



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Lewenstein Model

- We calculate the mean value of the dipole moment.

$$\vec{d}(t) = \langle \Psi(t) | e\vec{r} | \Psi(t) \rangle$$

In the Lewenstein's model:

$$|\Psi(t)\rangle = e^{iI_p t}\left[|i\rangle + \int d\vec{v}\, b(\vec{v},t)|\vec{v}\rangle\right]$$

$$\Rightarrow \vec{d}(t) = \underbrace{\langle i|\vec{r}|i\rangle}_{\substack{0 \\ \text{(symmetry of} \\ \text{the ground state)}}} + \int d\vec{v}\, b(\vec{v},t)\langle i|\vec{r}|\vec{v}\rangle + c.c. + \underbrace{\int d\vec{v} \int d\vec{v}'\, b^*(\vec{v},t)\langle \vec{v}'|\vec{r}|\vec{v}\rangle b(\vec{v}',t)}_{\substack{0 \\ \text{no continuum-continuum transitions}}}$$

$$b(\vec{v},t) = -i\int_0^t dt' E(t') d^*(\vec{v}) e^{-iS(t,t')}$$

$$\vec{d}(t) = -i\int_0^t dt' \int d\vec{k}\; d(\vec{k}+\vec{A}(t)) E(t') e^{-iS(t',t)} d^*(\vec{k}+\vec{A}(t')) + c.c.$$

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Lewenstein Model (cont.)

$$-i\int_0^t dt' \int d\vec{k}\; d(\vec{k}+\vec{A}(t)) e^{-iS(t,t')} E(t') d^*(\vec{k}+\vec{A}(t'))$$

The integral must be read from right to left:

$d^*(\vec{k}+\vec{A}(t')) = \langle \vec{v}|\vec{r}|i\rangle$: atomic ionization

$e^{-iS(t,t')}$: evolution of the electron in the continuum

$d(\vec{k}+\vec{A}(t)) = \langle i|\vec{r}|\vec{v}\rangle$: recapture of the electron

$$S(t,t') = -\int_{t'}^{t} dt'' \left\{ \frac{\left[\vec{k}+\vec{A}(t'')\right]^2}{2} + I_p \right\}$$

Lewenstein Model (cont.)

change of variables: $\tau = t - t'$ (flight time)

$$-i\int_0^t d\tau \int d\vec{k}\; d(\vec{k}+\vec{A}(t))e^{-iS(t,t-\tau)}E(t-\tau)d^*(\vec{k}+\vec{A}(t-\tau))$$

We solve the integral within the saddle-point approximation.
If the phase S changes a lot, then the positive and negative values will cancel out. The only surviving part is:

$$\delta S(t',t) = 0 \quad \Rightarrow \quad \delta S(\vec{k},t,t-\tau) = 0$$ Principle of least action

Exercise 14: Prove that:

$$\nabla_{\vec{k}} S(\vec{k},t,t-\tau) = 0 \quad \Rightarrow \quad \vec{r}(t) - \vec{r}(t-\tau) = 0$$ (return condition)

$$\frac{\partial}{\partial \tau} S(\vec{k},t,t-\tau) = 0 \quad \Rightarrow \quad \tfrac{1}{2}\left(\vec{k}+\vec{A}(t-\tau)\right)^2 + I_p = 0$$ (ionization condition)

Lewenstein Model (cont.)

Then we must Fourier transform:

$$d(q\omega) = \tfrac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dt\; d(t) e^{iq\omega t}$$

Exercise 15: Prove that:

$$\frac{\partial}{\partial t} S(\vec{k},t,t-\tau) = 0 \quad \Rightarrow \quad \tfrac{1}{2}\left(\vec{k}+\vec{A}(t)\right)^2 - \tfrac{1}{2}\left(\vec{k}+\vec{A}(t-\tau)\right)^2 + I_p = q\omega$$

(energy conservation for emission of a harmonic photon)

$$\varepsilon = \hbar q\omega = T_{\text{kin}} + I_p \quad \text{iff} \quad v(t-\tau) = 0 \quad \text{(SMM)}$$

$$\Rightarrow \varepsilon_{\max} = \hbar q_{\max}\omega = T_{\max} + I_p = 3.17U_p + I_p$$

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Quantum Considerations

- There is an XUV emission every laser semicycle *T*/2.
- The phase corresponding to each XUV is locked to the fundamental phase.
- The sign of the laser field changes every semicycle => XUV field changes.

In conclusion, the real periodicity of the harmonic emission is T with periodicity T/2 for the intensity

The total XUV pulse emitted with frequency ω_q is:

$$d(q\omega) = \int A_{q\omega} e^{-i(q\omega t + \varphi_q)} dt$$

Summing over a lot of semicycles:

$$d(q\omega) \propto \sum A_{q\omega} (-1)^n e^{-i(q\omega(t+nT/2)+\varphi_q)}$$

$$\propto A_{q\omega} e^{-i(q\omega t+\varphi_q)} \sum e^{-i(q\omega nT/2 - n\pi)}$$

- The first factor corresponds to an oscillating wave with frequency qω

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Quantum Considerations

- The second factor corresponds to an amplitude:

$$\sum e^{-i(q\omega T/2-\pi)n} = \frac{1-e^{-Ni(q\omega T/2-\pi)}}{1-e^{-i(q\omega T/2-\pi)}}$$

geometrical sum

$$= e^{-(N-1)i(q\omega T/2-\pi)} \frac{\sin\left[N\left(q\omega T/2-\pi\right)/2\right]}{\sin\left[\left(q\omega T/2-\pi\right)/2\right]}$$

The square modulus

$$\xrightarrow[N\to\infty]{} \delta\left(\frac{q\omega T/2-\pi}{2} - n\pi\right)$$

Conservation of energy

$$\Rightarrow \frac{q\omega T/2-\pi}{2} = n\pi$$

$$\Rightarrow q\omega = \frac{(2n+1)\pi}{T/2} = \underbrace{(2n+1)}_{q:\ \text{odd}} \underbrace{\frac{2\pi}{T}}_{\omega}$$

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Quantum Considerations

- To calculate the harmonic generation exactly the TDSE must be solved numerically and then calculate d(t) and Fourier Transform.
- The propagation in the media must also be considered.

Some people also use the acceleration of the expected value of the dipole:

$$\ddot{\vec{d}}(t) = \frac{d^2}{dt^2}\langle \Psi(t) | \vec{r} | \Psi(t) \rangle$$

HHG is useful for the generation of an attosecond pulse source.

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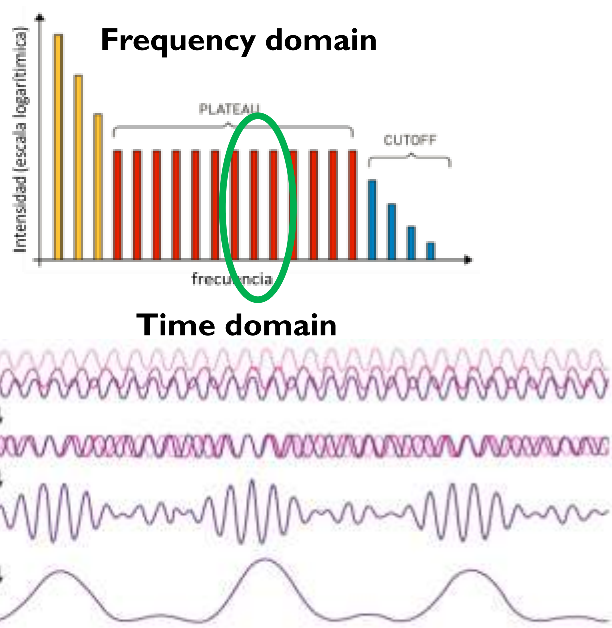
Quantum Considerations



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Quantum Considerations

POLARIZATION GATING

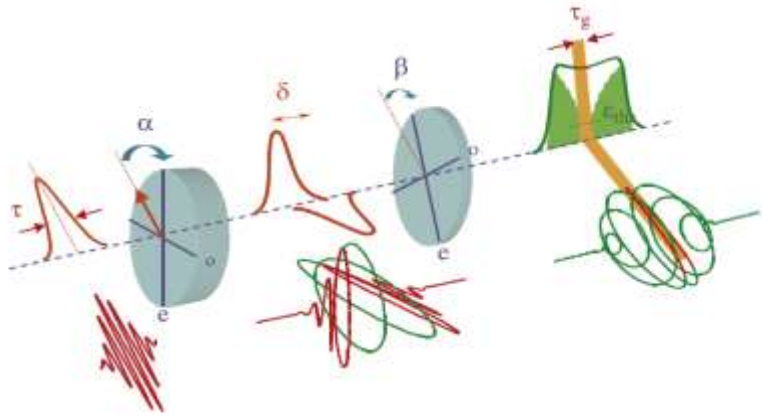

Fig. 1. Polarization gating based on two birefringent plates. α indicates the angle between the polarization axis of the linearly polarized incident pulse (FWHM = τ) and the axis of the first thick plate. The ordinary and extraordinary components of the pulse acquire a temporal delay δ. β is the angle between the initial polarization direction and the axis of the zero-order quarter waveplate. τ_g and ϵ_{thr} indicates the time gate window and the threshold ellipticity, respectively.

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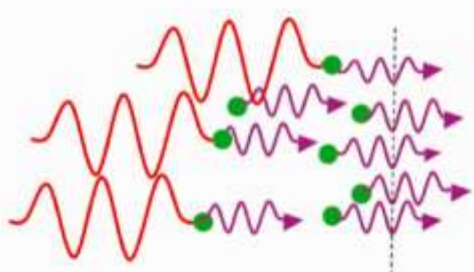
MACROSCOPIC ASPECTS

Phase matching

Many-atom response



Maxwell equations ➡ Wave equation

$$\nabla^2 \mathcal{E} - \frac{1}{c^2}\frac{\partial^2 \mathcal{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2 \mathcal{P}}{\partial t^2}$$

Generated field

Medium Polarization

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MACROSCOPIC ASPECTS

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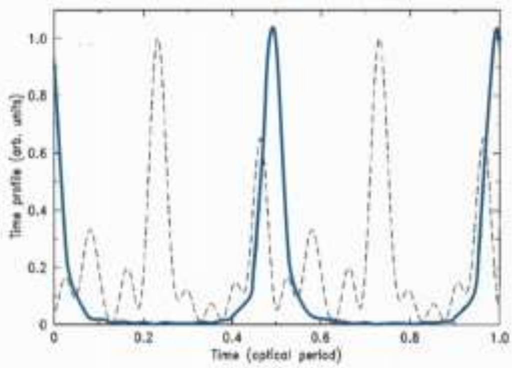
Phase matching $v_1 = \frac{\omega}{k_1}$ $v_q = \frac{q\omega}{k_q}$

Progress in understanding: Strong field nonlinear optics



$$\mathcal{P}_q \propto e^{iqk_1 z} \qquad \mathcal{E}_q \propto e^{ik_q z}$$

$$\Delta k = qk_1 - k_q = 0$$

$$\Delta k = \Delta k_{\text{disp}} + \Delta k_{\text{foc}} + \Delta k_i = 0$$

Dispersion, Laser focusing, Electron trajectory

- One attosecond pulse per laser half-cycle
- Phase-locked (synchronized) harmonics

Antoine et al. Phys. Rev. Lett. **77**, 1234 (1996)

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MEASUREMENTS OF ATTOSECOND PULSES

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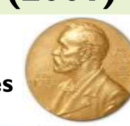
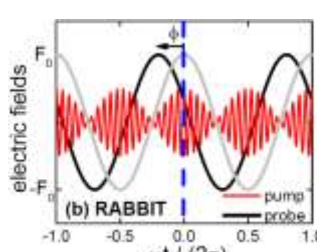
Attosecond pulses were demonstrated (2001)

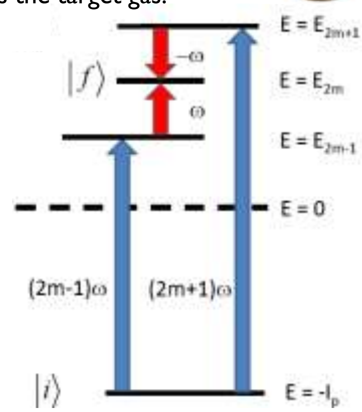
Pump-probe experiment: XUV+IR



In **Paris-Saclay**, the Agostini's group produced a **train of pulses with a duration of 250 as** using argon as the target gas.

RABBIT: Reconstruction of Attosecond harmonic Beating By Interference of Two-photon transitions





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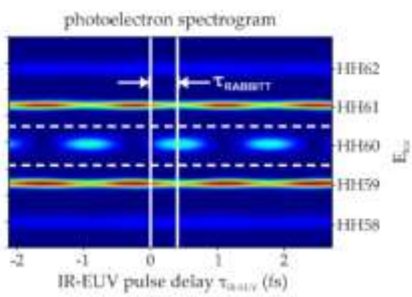
MEASUREMENTS OF ATTOSECOND PULSES

$$|T|^2 = A + B\cos(2\phi + \delta)$$

$$\delta = \underbrace{\phi_m - \phi_{m-1}}_{\text{HH}} + \underbrace{\arg\left[\tilde{I}^{(n_1)}_{m-1}\right] - \arg\left[\tilde{I}^{(n_2)}_{m}\right]}_{\text{atomic}}$$

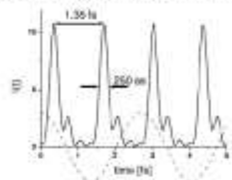


SCIENCE VOL 292 1 JUNE 2001

Observation of a Train of Attosecond Pulses from High Harmonic Generation

P. M. Paul,[1] E. S. Toma,[2] P. Breger,[1] G. Mullot,[3] F. Augé,[3] Ph. Balcou,[3] H. G. Muller,[2*] P. Agostini[1]

In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure each phase through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.



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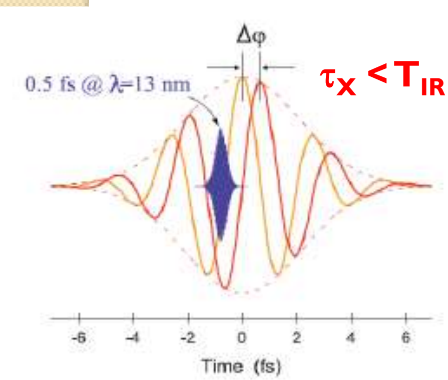
MEASUREMENTS OF ATTOSECOND PULSES

In **Vienna**, the Krausz's group produced isolated pulses of duration **650 as.**

Attosecond streaking



They used spectral filtering to select relevant harmonics with a multilayer XUV mirror. Then they measured the kinetic energy spectrum of 4*p* photoelectrons ejected from krypton atoms under simultaneous irradiation by 90 eV photons and the light pulses at 750 nm from the drive laser generating the harmonic radiation (streaking).

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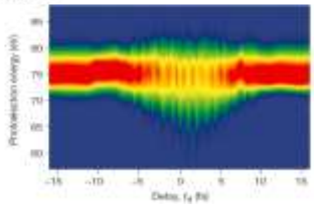
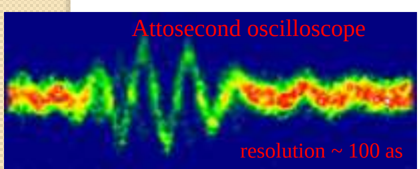
MEASUREMENTS OF ATTOSECOND PULSES




NATURE | VOL 414 | 29 NOVEMBER 2001 | www.nature.com

Attosecond metrology

SMM: $k + A(t_0) = 0$



E. Goulielmakis *et al*, Science **305**, 1267 (2004)