

# Sources de lumière attoseconde : principes et applications

Journée Académique de l'Union des Professeurs de  
Physique et Chimie,  
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2. *Institut Universitaire de France, 1 rue Descartes, 75231 Paris, France*

- Prix Nobel de Physique
- Qu'est-ce qu'une attoseconde
- Etude de la dynamique des phénomènes
- Comment générer des impulsions attosecondes ?
- Comment mesurer des impulsions attosecondes ?
- Que fait-on avec des impulsions attosecondes ?

# Introduction



Pierre Agostini



Ferenc Krausz

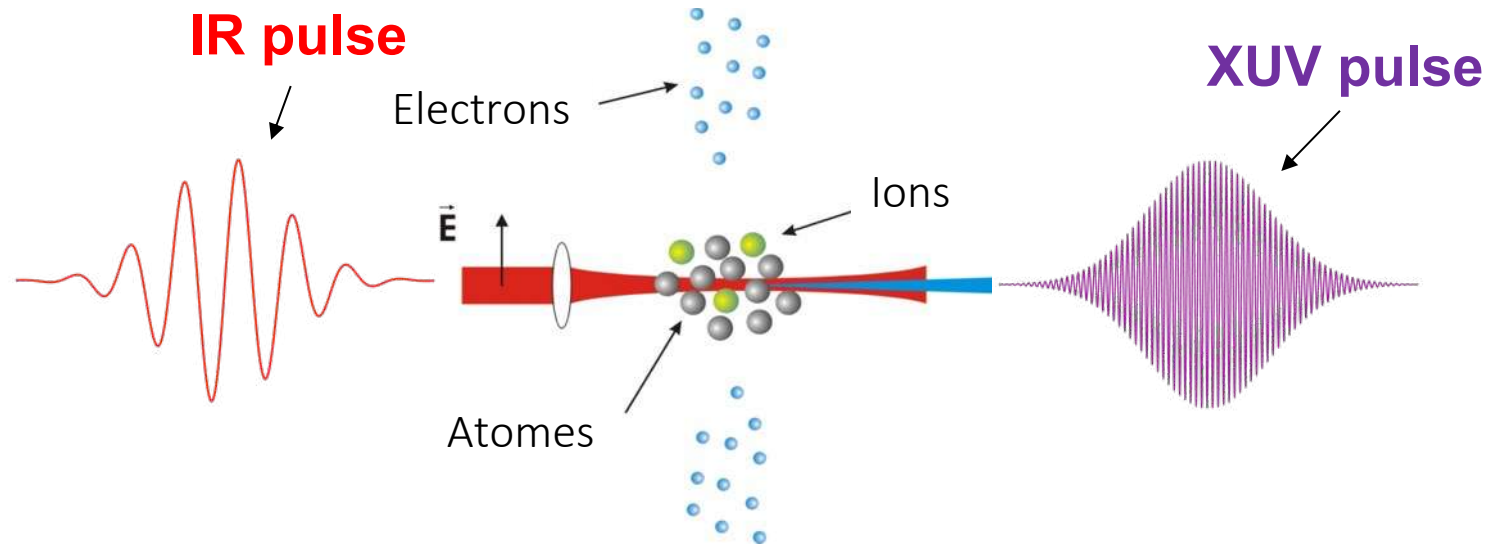


Anne L'Huillier

"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"



## Découverte de la génération d'harmoniques d'ordre élevé (1987)

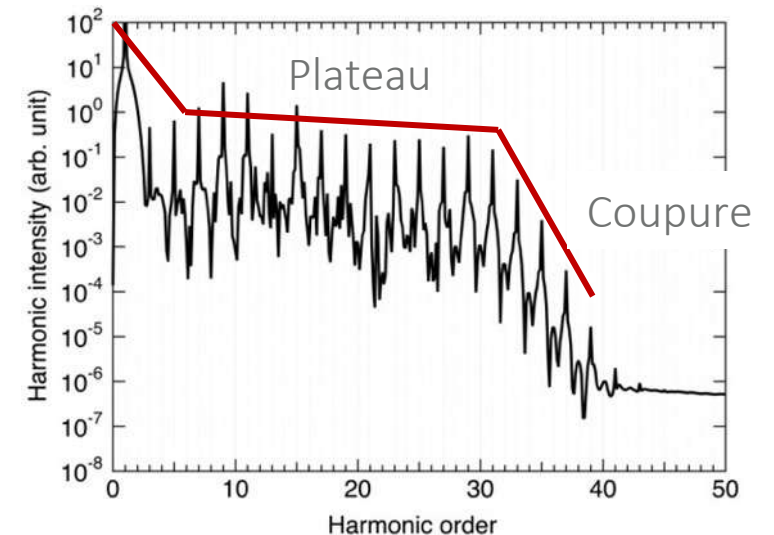
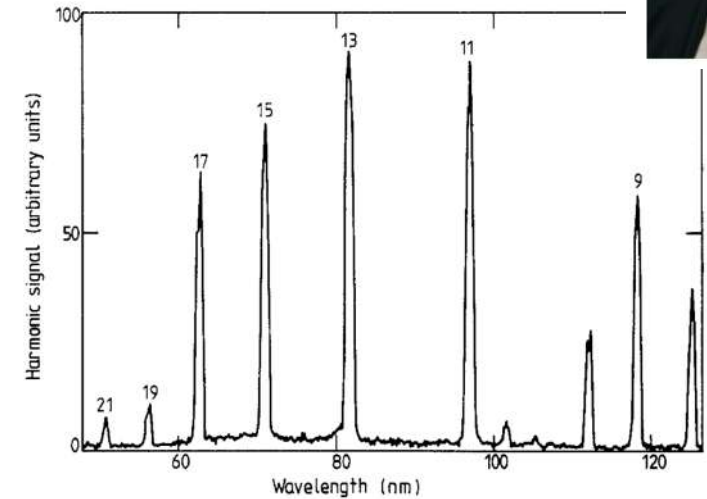


Harmonique  $n$  :

$$\lambda_n = \frac{\lambda_{laser}}{n}$$

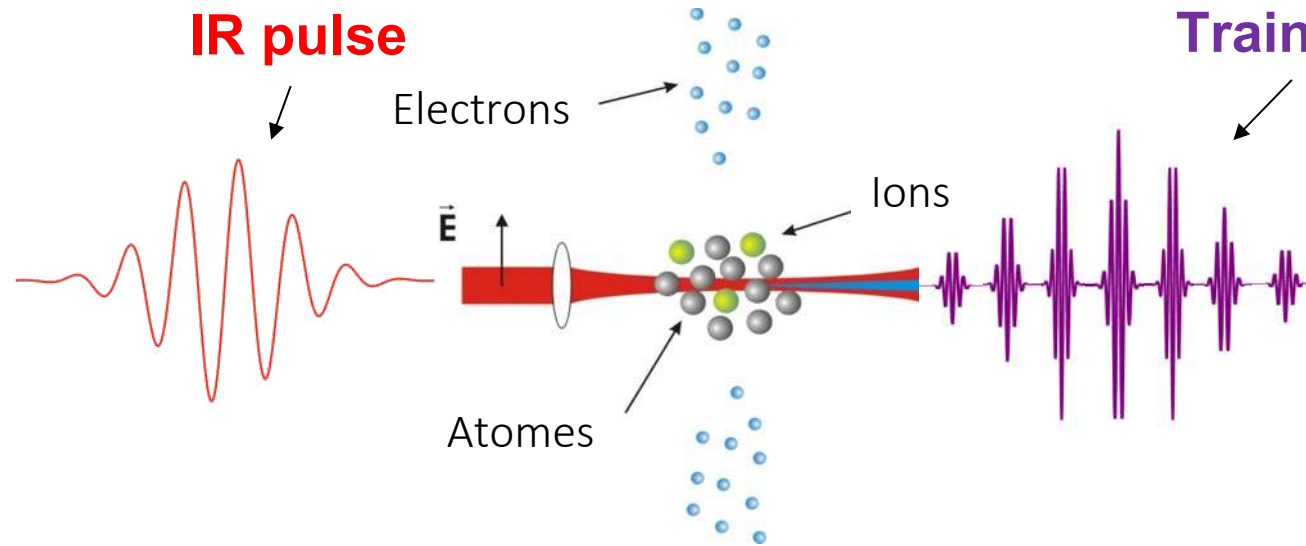
$$E_n = nE_{laser}$$

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

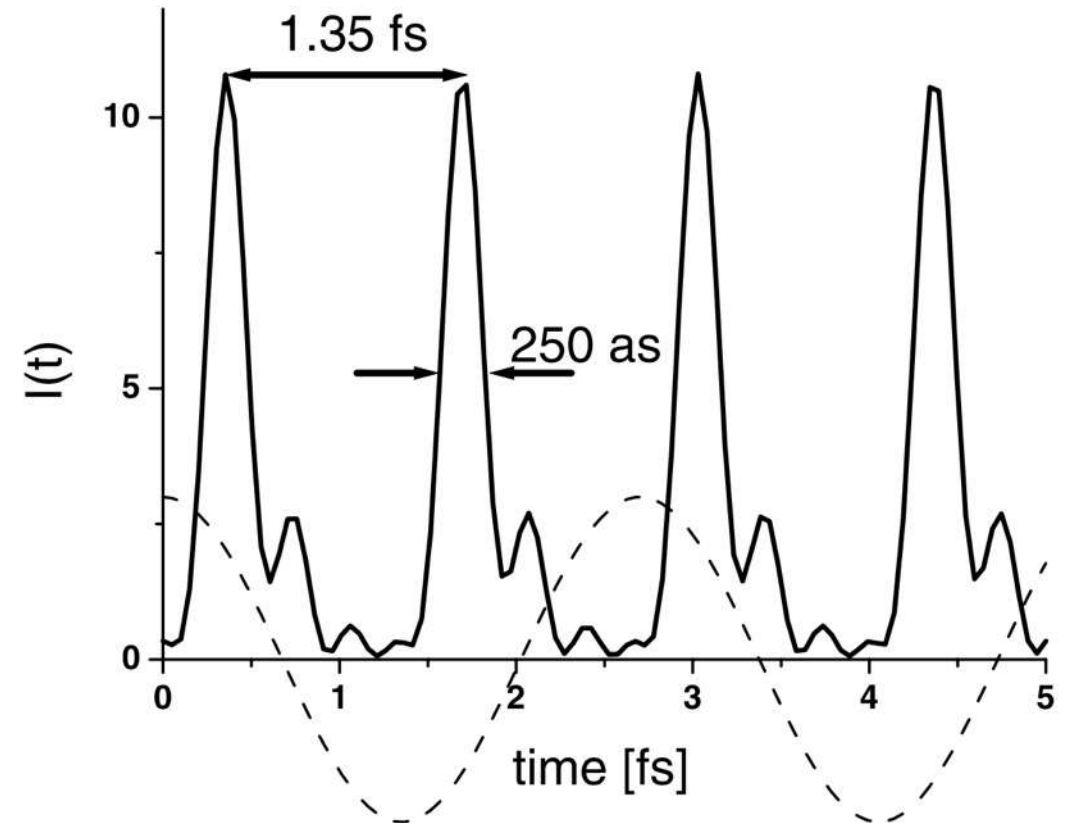




## Production d'une série d'impulsions attosecondes (2001)



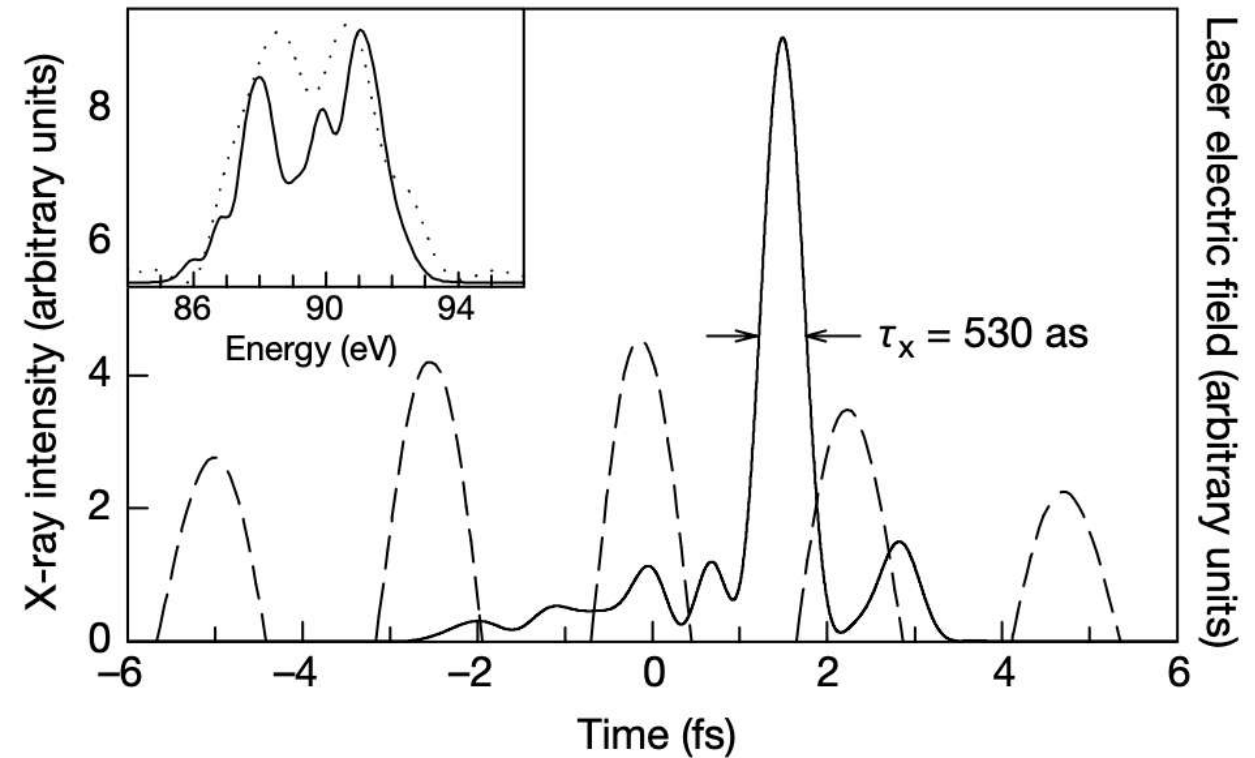
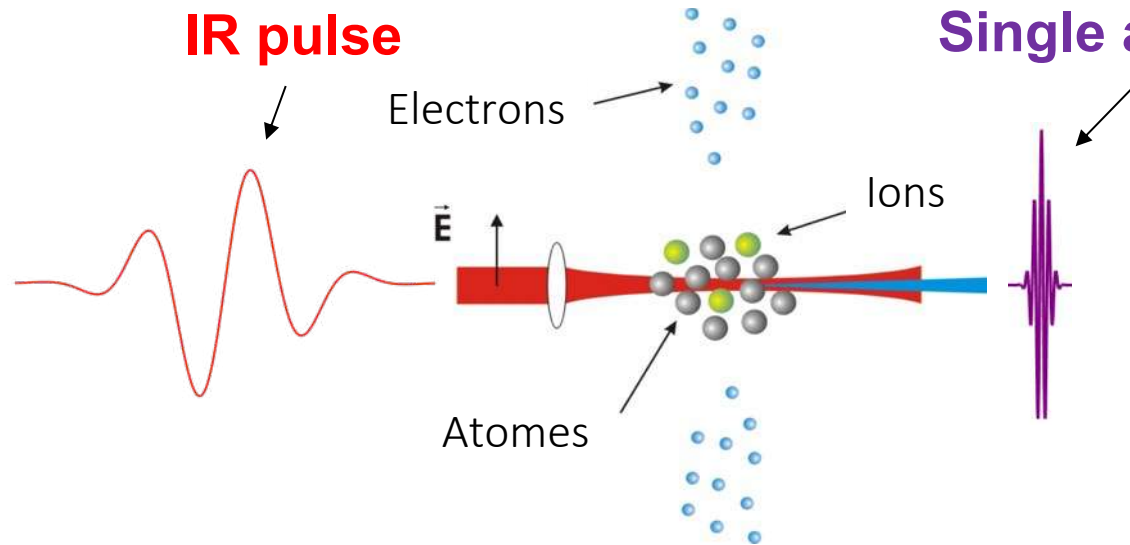
$$\tau = 250 \cdot 10^{-18} = 250 \text{ as}$$



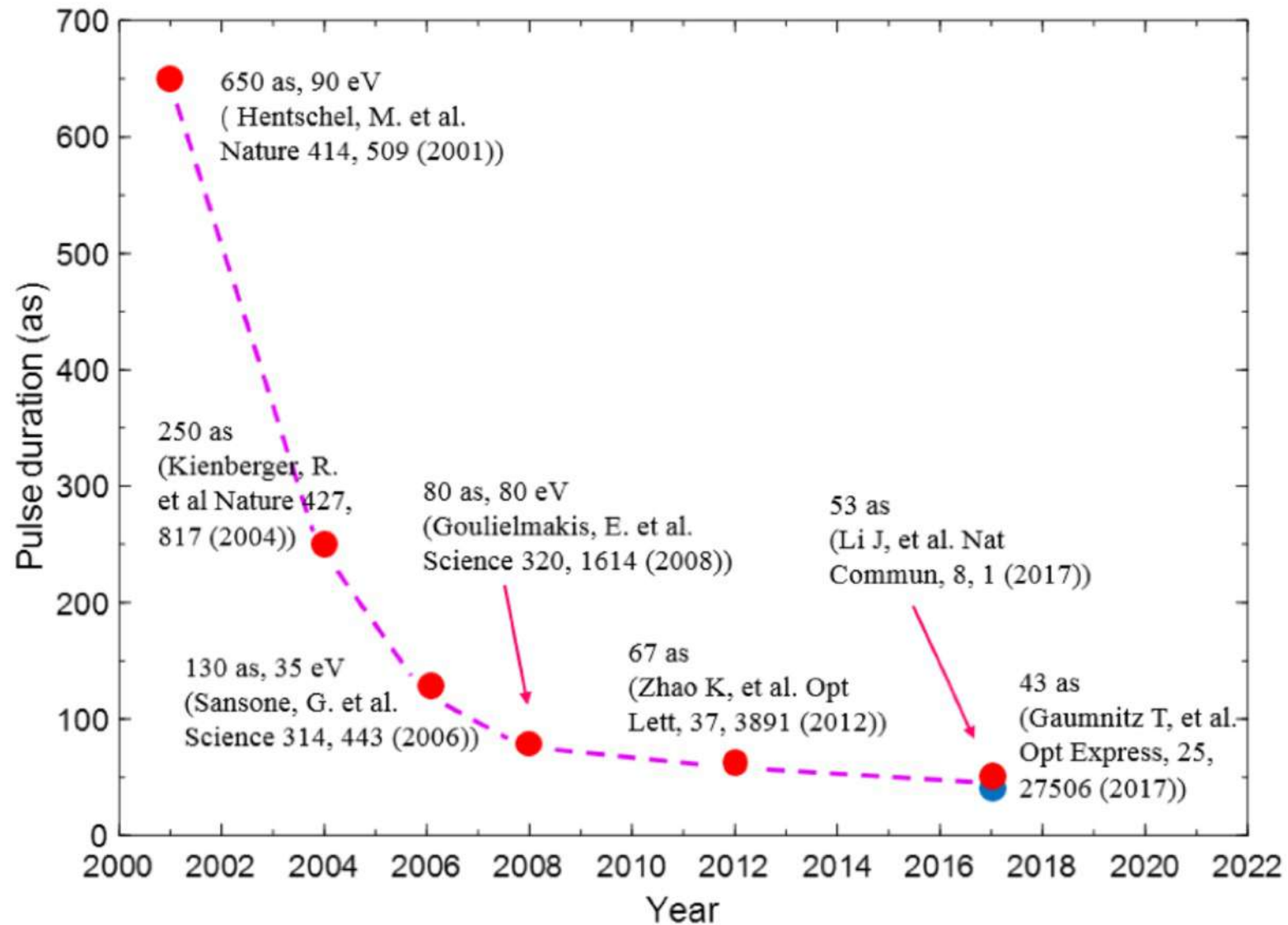
*P. M. Paul et al., Science 292 (2001)*



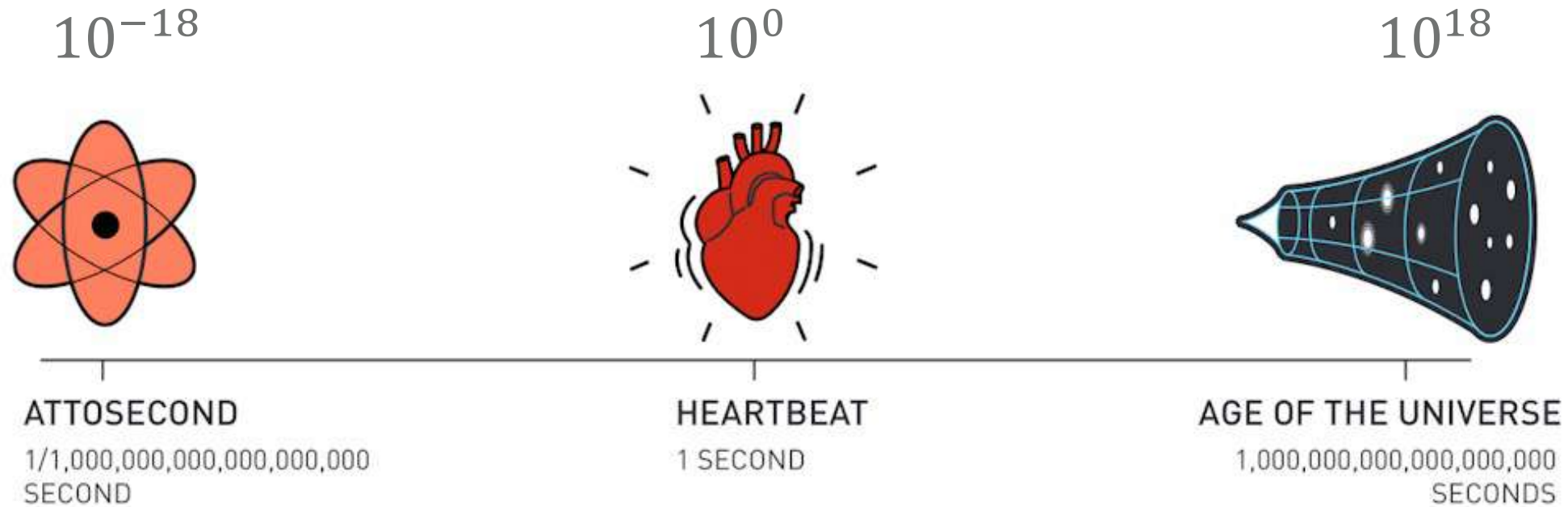
## Production d'une impulsion attoseconde isolée (2001)



*M. Hentschel et al. Nature 414 (2001).*





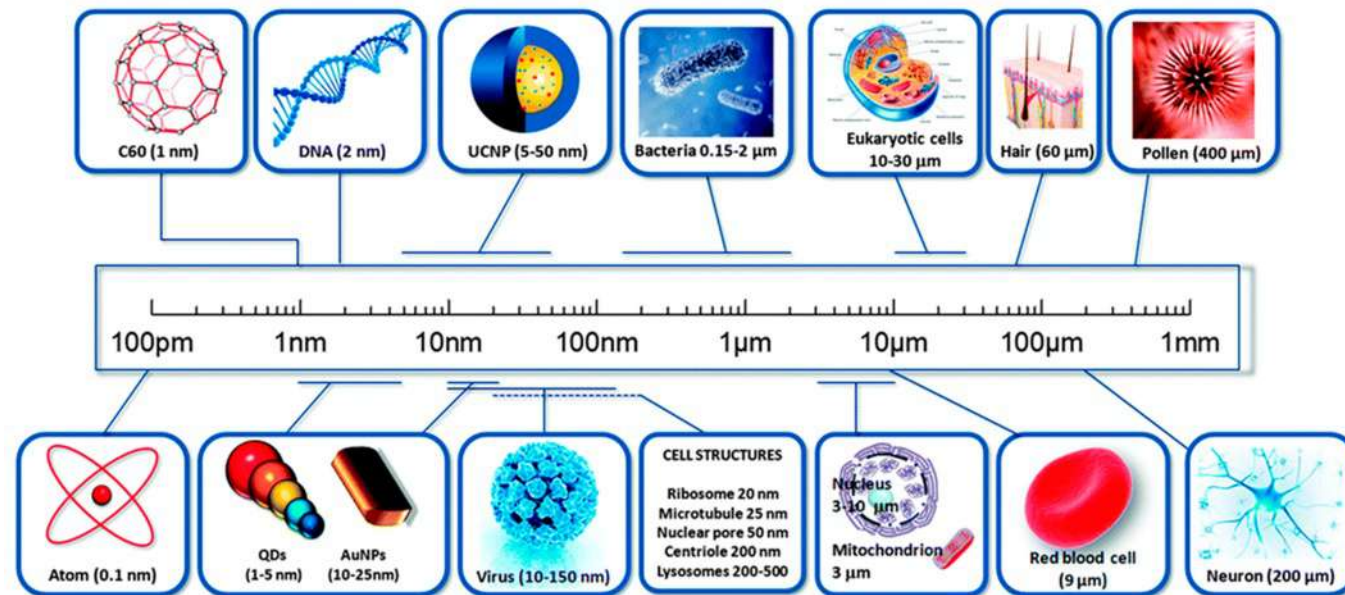


Electrons' movements in atoms and molecules are so rapid that they are measured in attoseconds. An attosecond is to one second as one second is to the age of the universe.

©Johan Jarnestad/The Royal Swedish Academy of Sciences

Distance parcourue en 1 as à la vitesse de la lumière:

- pour 1 as :  $d = c.t = 3.10^8 \times 1.10^{-18} = 3.10^{-10} = 0.3 \text{ nm}$
- pour 10 as :  $d = c.t = 3.10^8 \times 10.10^{-18} = 3.10^{-9} = 3 \text{ nm}$



Bayda, S et al. *Molecules*, 25, 112 (2020)

En 10 as, la lumière parcourt l'épaisseur d'une séquence ADN

Plus précisément, à l'échelle atomique:

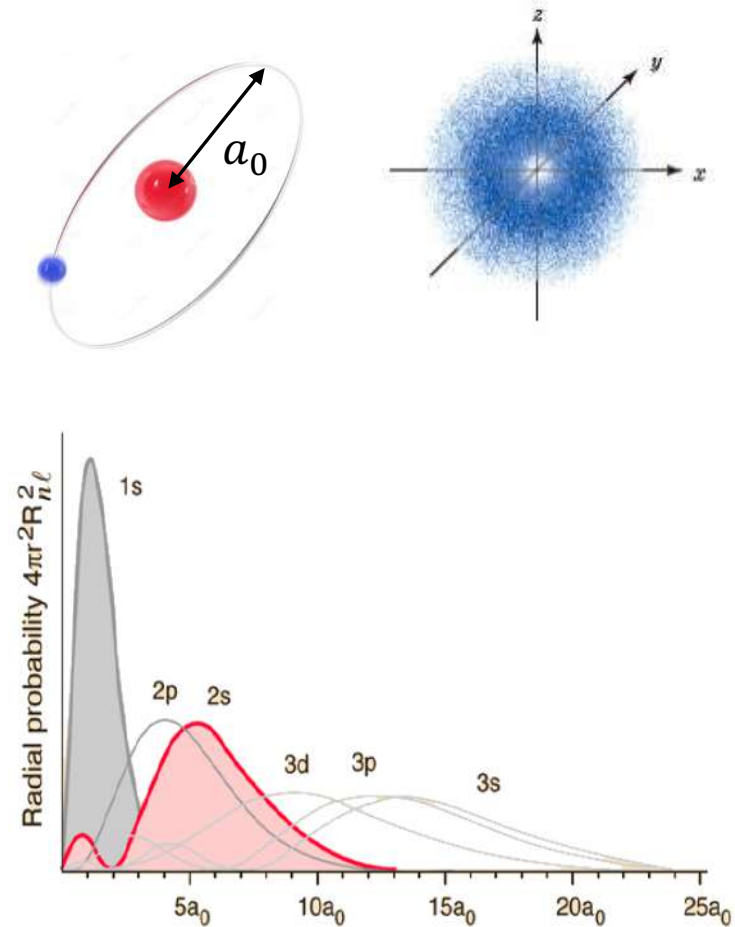
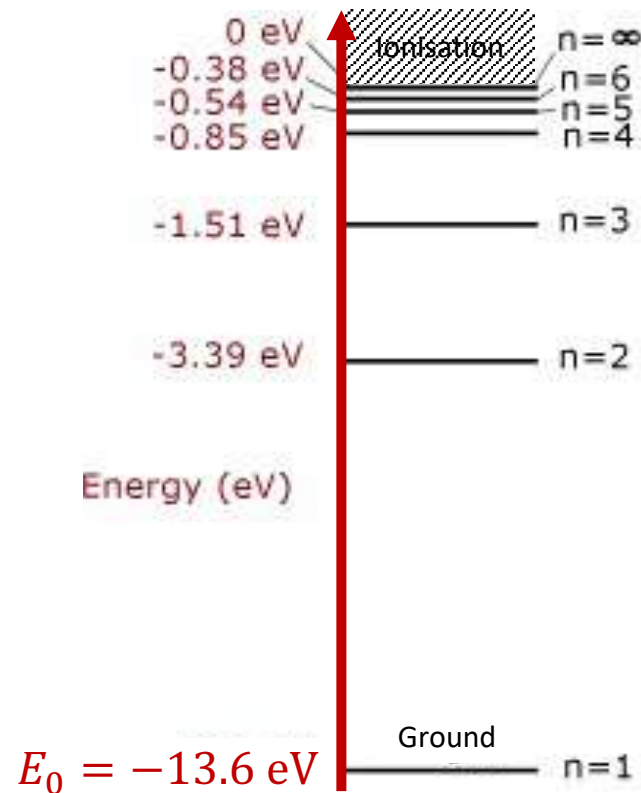
Atome d'hydrogène

$$a_0 = 52.9 \times 10^{-12} = 52.9 \text{ pm}$$

$$E_0 = -13.6 \text{ eV}$$

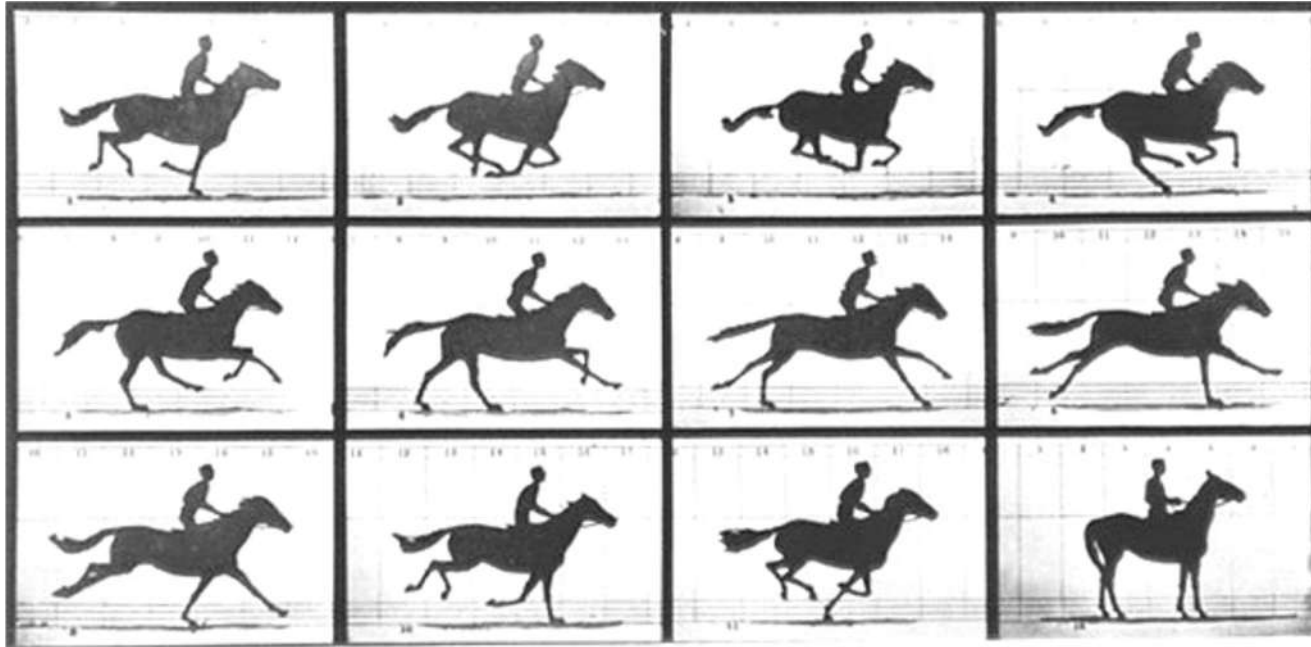
$$v_0 = \frac{c}{137} = 2.2 \times 10^6 \text{ m.s}^{-1}$$

$$t = \frac{2 \cdot a_0}{v_0} = 2 \frac{52.9 \times 10^{-12}}{2.2 \times 10^6} \\ = 48 \times 10^{-18} = \mathbf{48 \text{ as}}$$



Il faut 48 as à l'électron pour traverser l'atome d'H

## Eadweard Muybridge (1878), *Galloping Horse, Motion Study*



Etude de la dynamique d'un cheval au galop

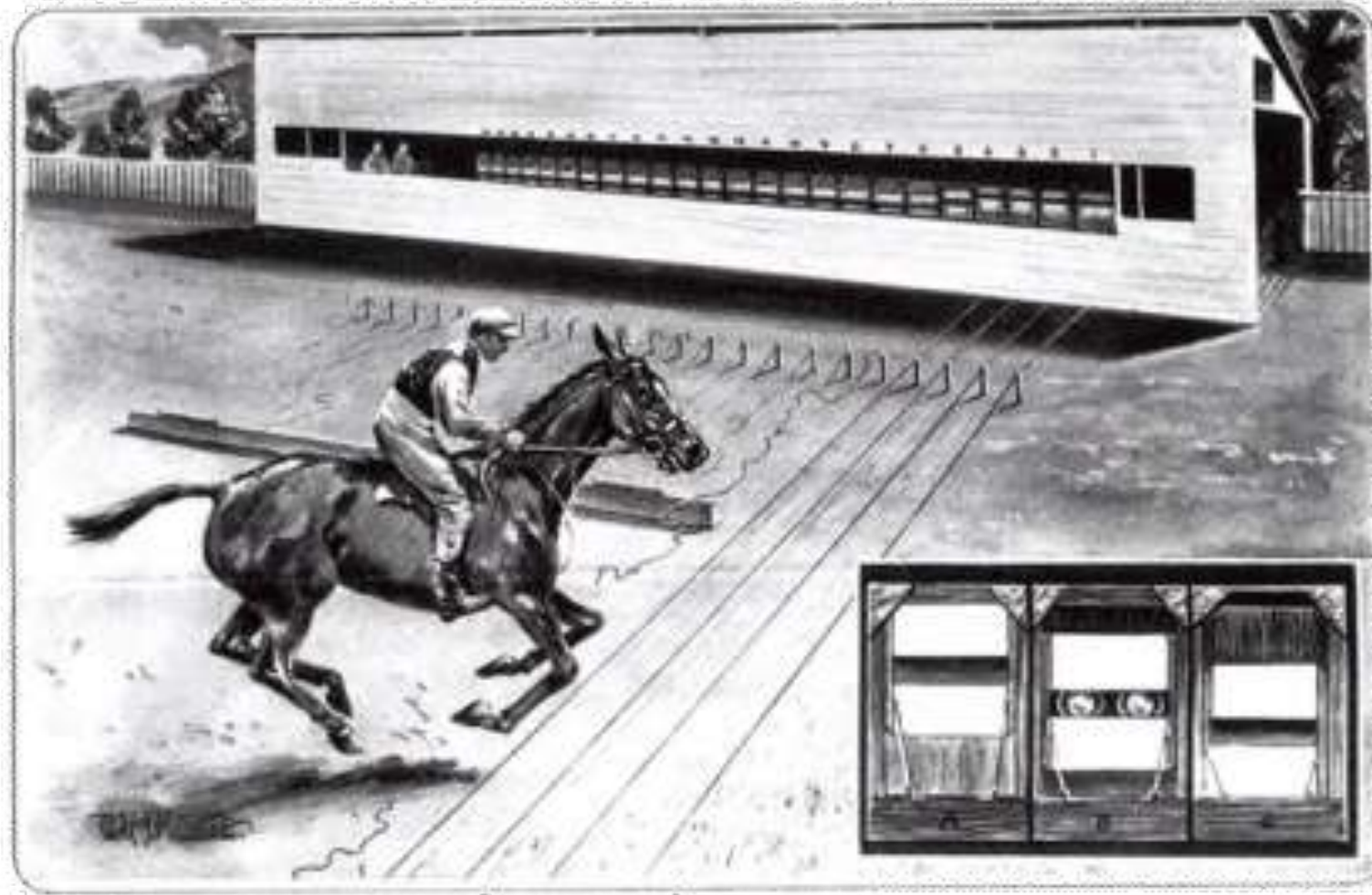
Conditions à vérifier:

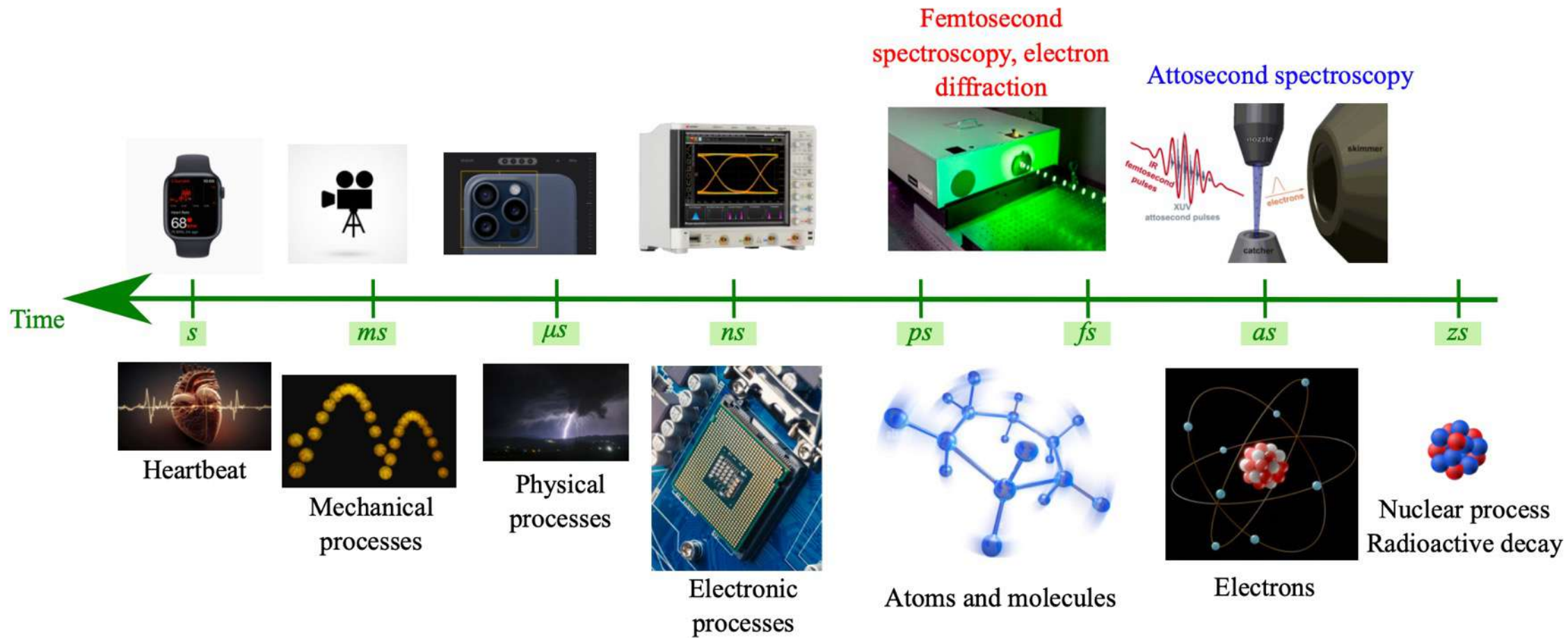
Vitesse =  $965 \text{ m/mn} = 58 \text{ km/h} = 1.142 \text{ m/s}$

Photo nette si déplacement  $< 1 \text{ cm}$

Durée d'acquisition  $< 1.4 \text{ ms}$

La Nature (n°291, 28 décembre 1878).





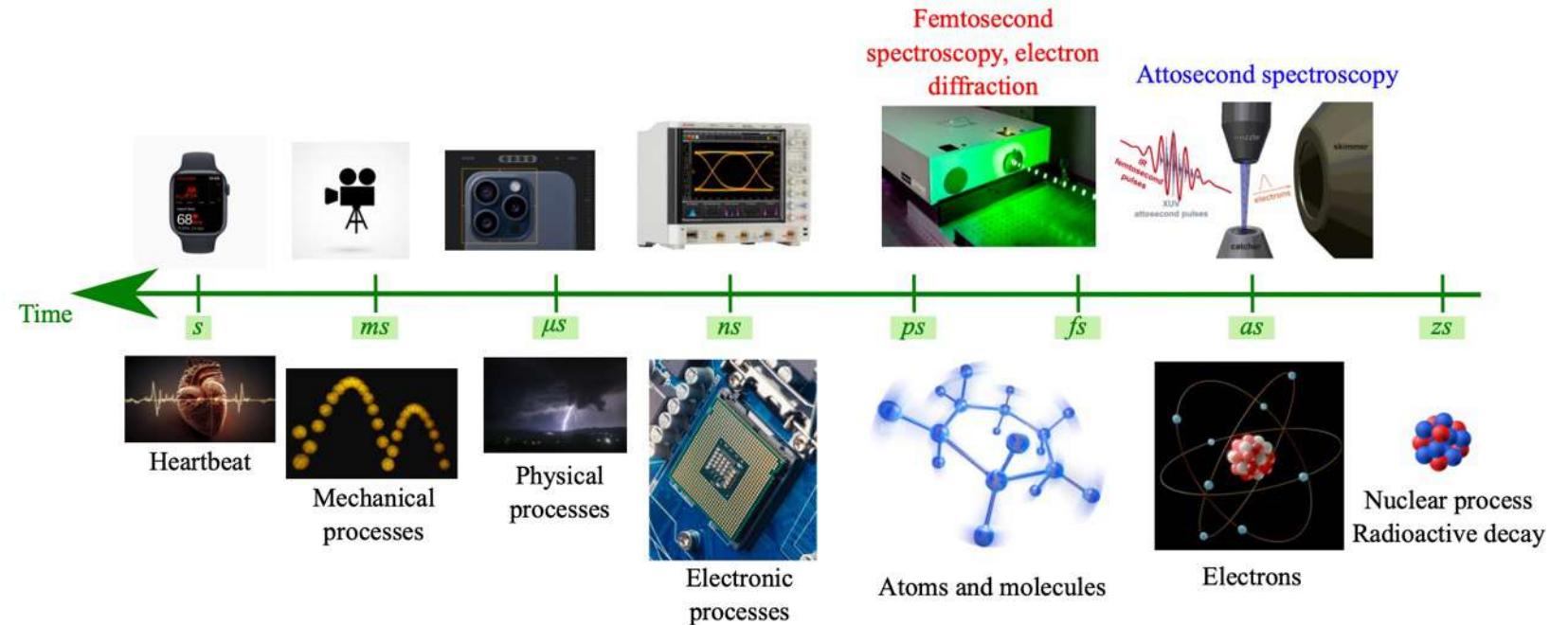


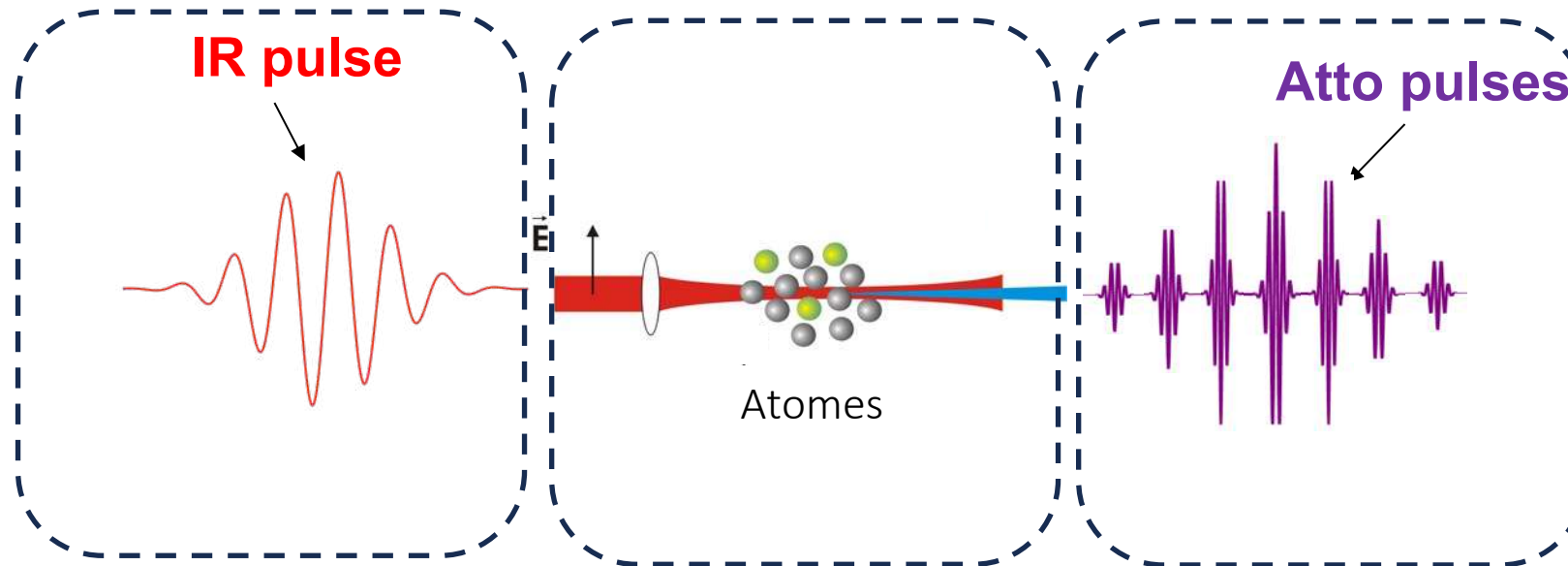
Ahmed Zewail

"for his studies of the transition states of chemical reactions using femtosecond spectroscopy"



1999



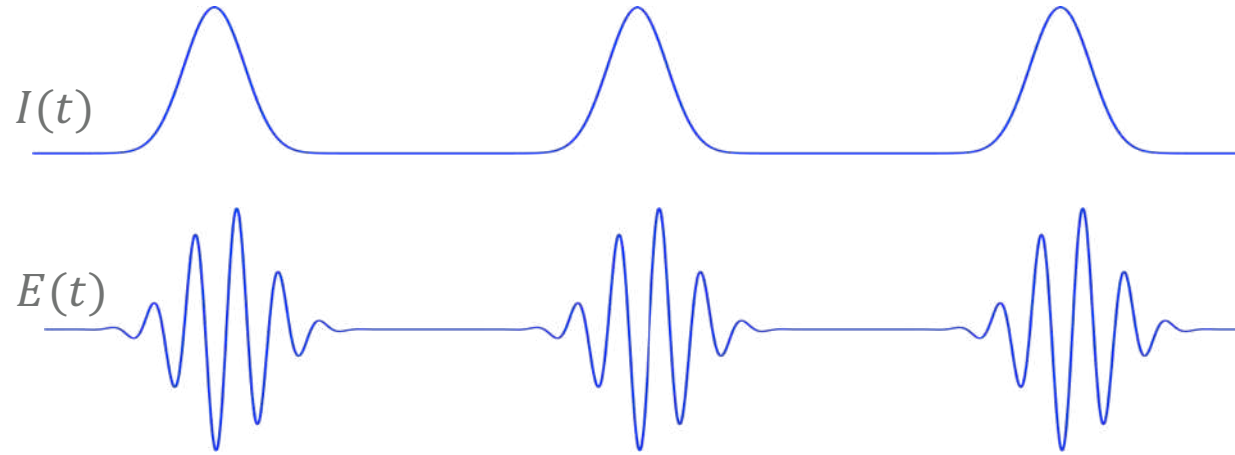


Laser d'excitation

Mécanisme de  
génération

Propriétés du  
rayonnement  
attoseconde

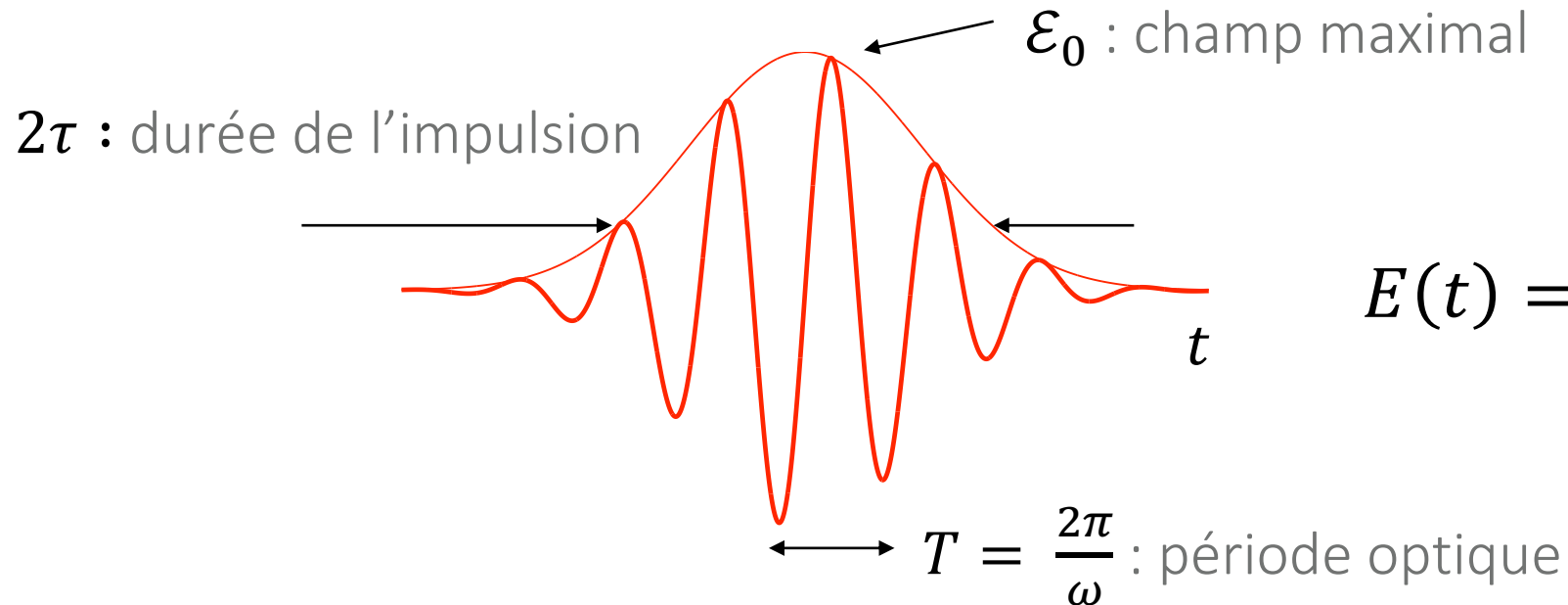




$$\mathcal{E}(t) = \varepsilon_0 e^{-\left(\frac{t}{\tau}\right)^2} e^{-i(\omega t + \varphi)}$$

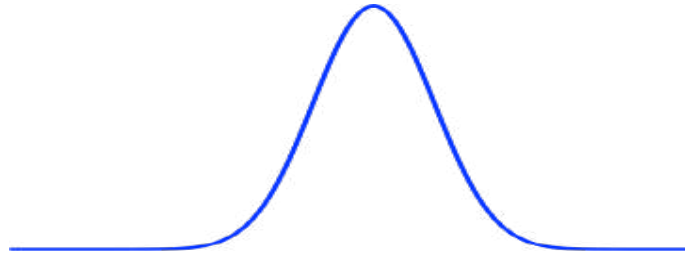
$$I(t) = |\mathcal{E}(t)|^2$$

$$E(t) = \text{Re}(\mathcal{E}(t))$$



$$E(t) = \varepsilon_0 e^{-\left(\frac{t}{\tau}\right)^2} \cos(\omega t + \varphi)$$

Longueur d'onde:  $\lambda = \frac{2\pi c}{\omega}$



Photon:



Energie:

$$E_{\text{photon}} = \hbar\omega = h\nu = \frac{hc}{\lambda}$$

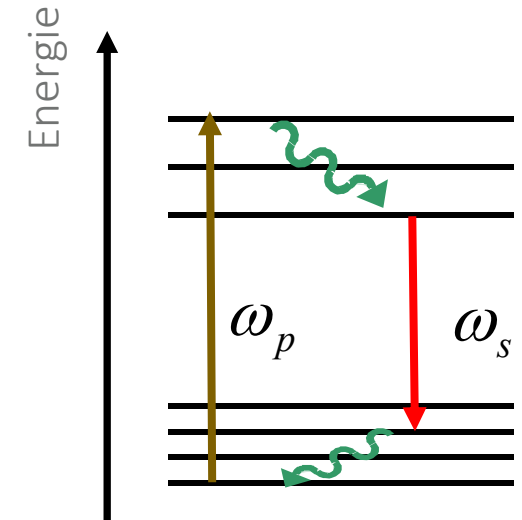
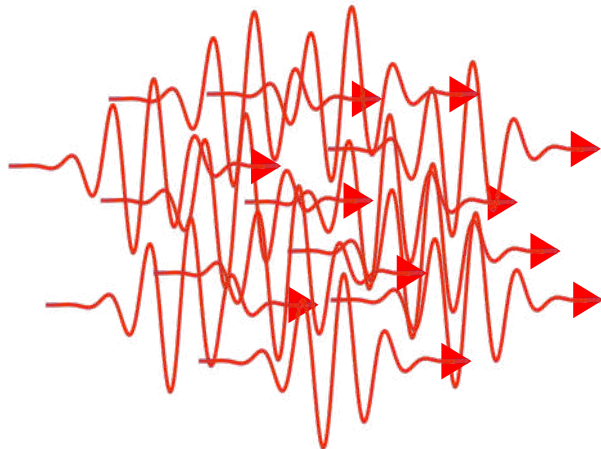
Dans un pulse:

$$\lambda = 800 \text{ nm}$$

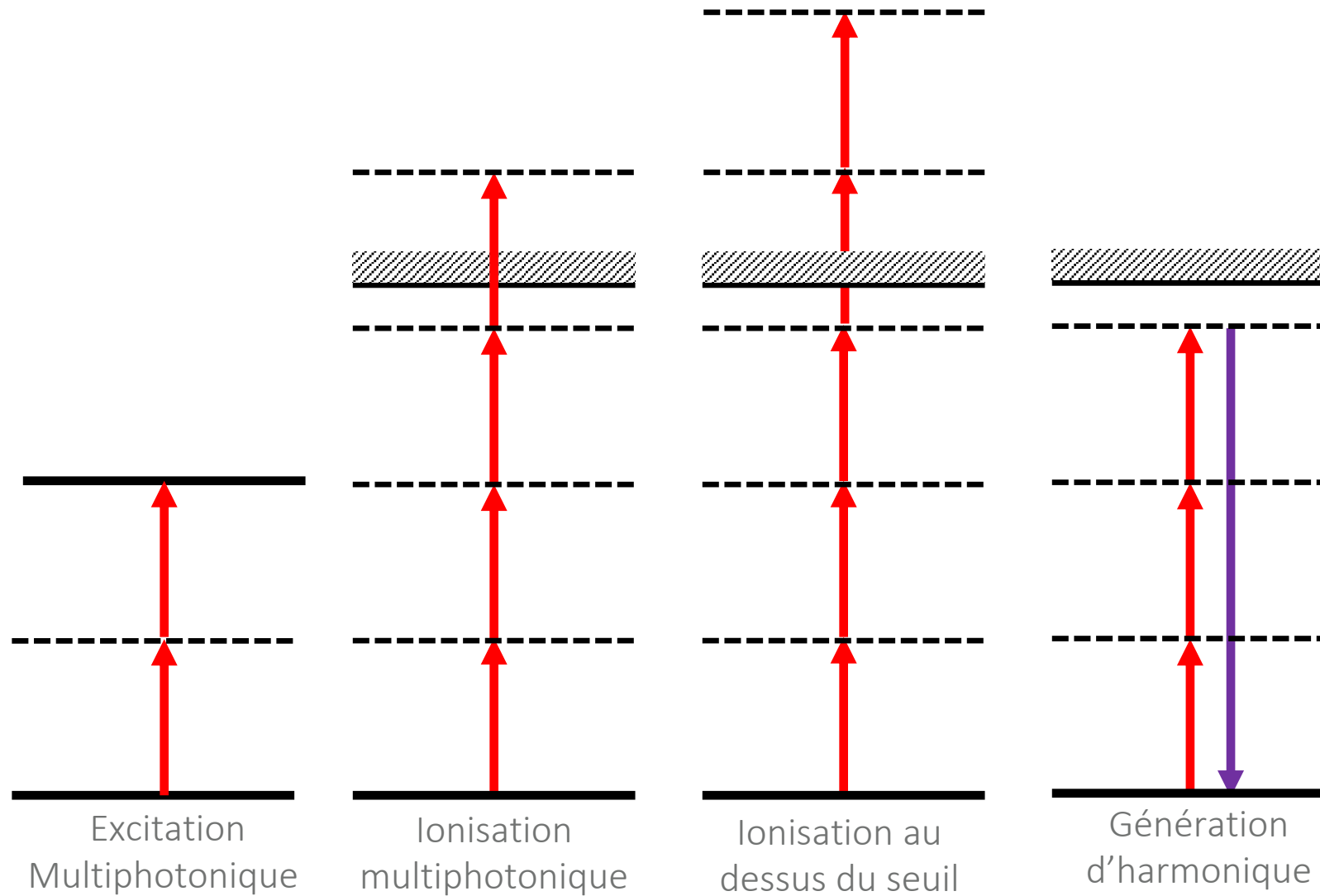
$$E_{\text{photon}} = 2.5 \cdot 10^{-19} \text{ J}$$

$$E_{\text{pulse}} = 1 \text{ mJ}$$

$$N = 4 \cdot 10^{15} \text{ photons}$$

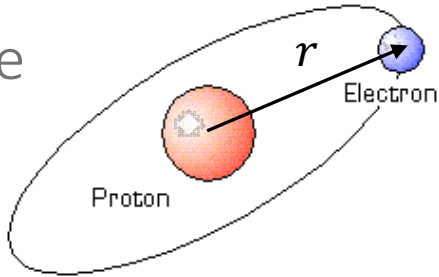


Transition atomique:

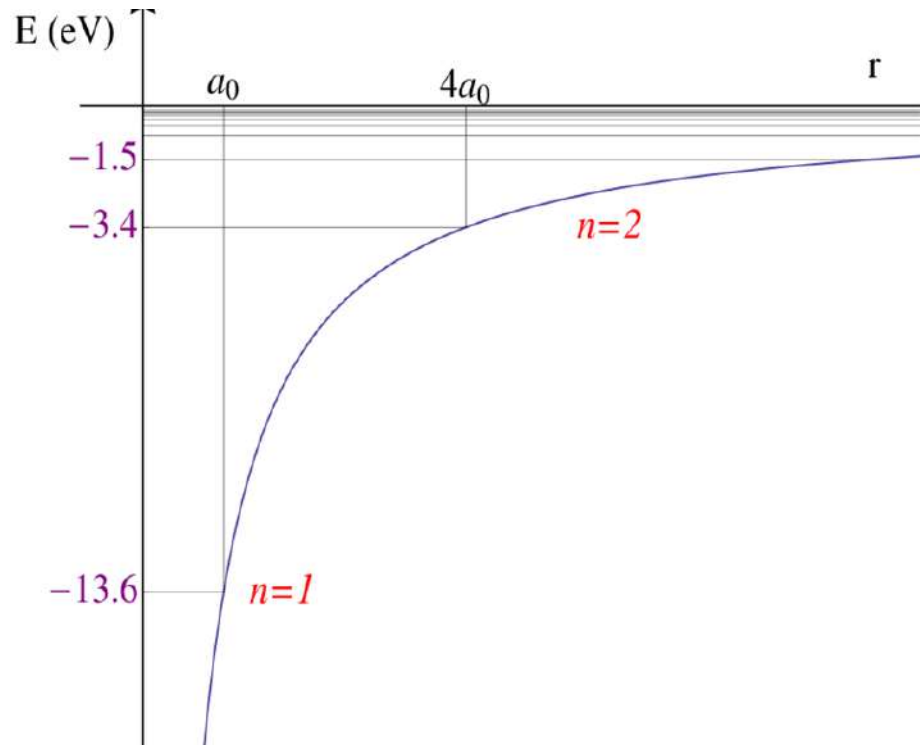


$$P \propto \alpha I^N$$

Hydrogène



$$V(r) = -\frac{Z}{r}$$



Unités atomiques:

- Longueur:

$$a_0 = 52.9 \times 10^{-12} = 52.9 \text{ pm}$$

- Champ:

$$E_a = \frac{E_h}{ea_0} = 5.1 \times 10^{11} \text{ Vm}^{-1}$$

- Intensité:

$$I_a = 3.5 \times 10^{16} \text{ Wcm}^{-2}$$

Champs forts : intensité laser  $I \approx I_a$

$10^{-1} \text{ W/cm}^2$  : Sun intensity on ground

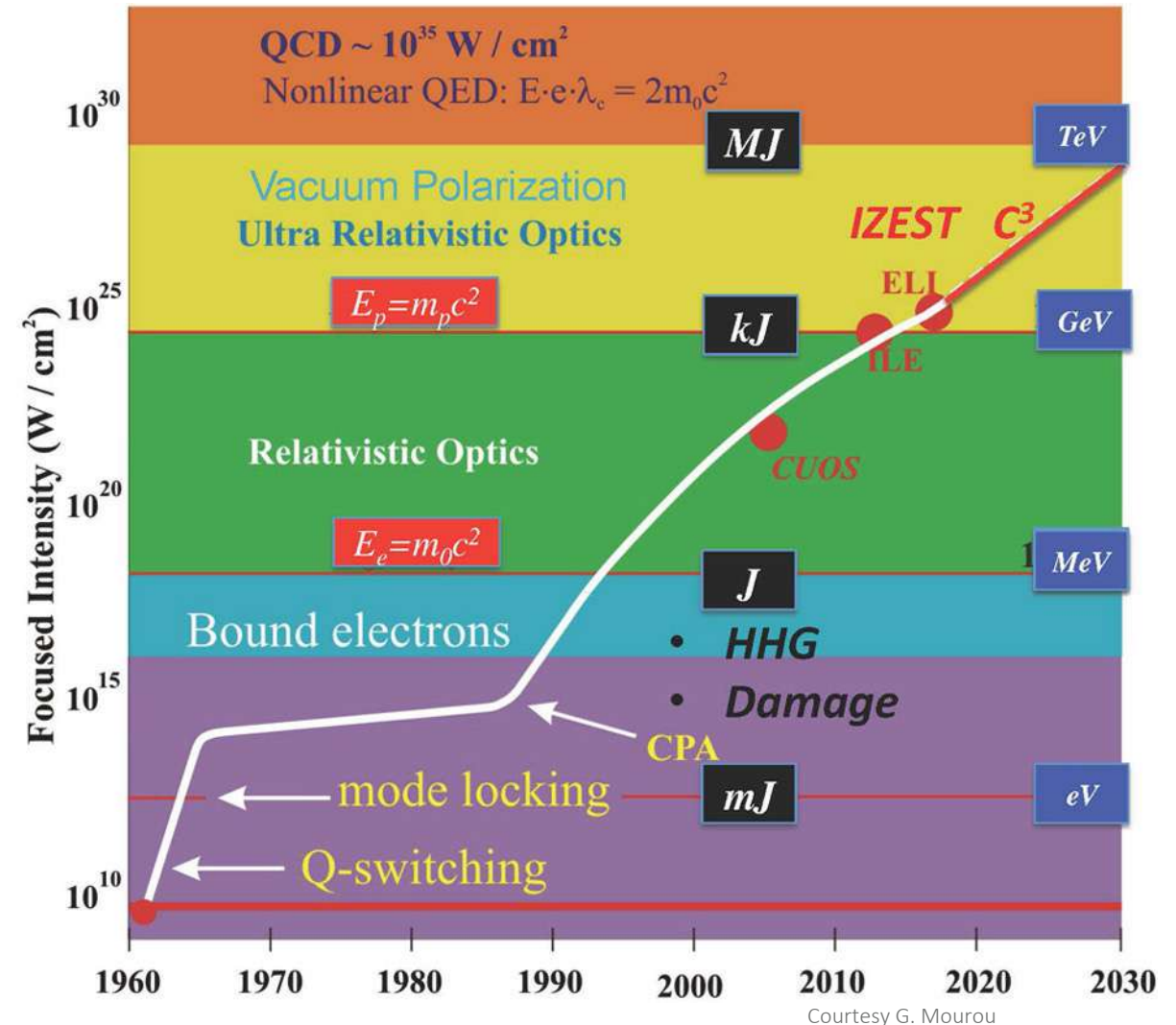
$10^{12} \text{ W/cm}^2$  : Ionization

$10^{16} \text{ W/cm}^2$  : Ionization saturation

$10^{18} \text{ W/cm}^2$  : Relativistic electrons

$10^{25} \text{ W/cm}^2$  :  $\text{U}^{92+}$

$10^{29} \text{ W/cm}^2$  : Pair creation





Gérard  
Mourou



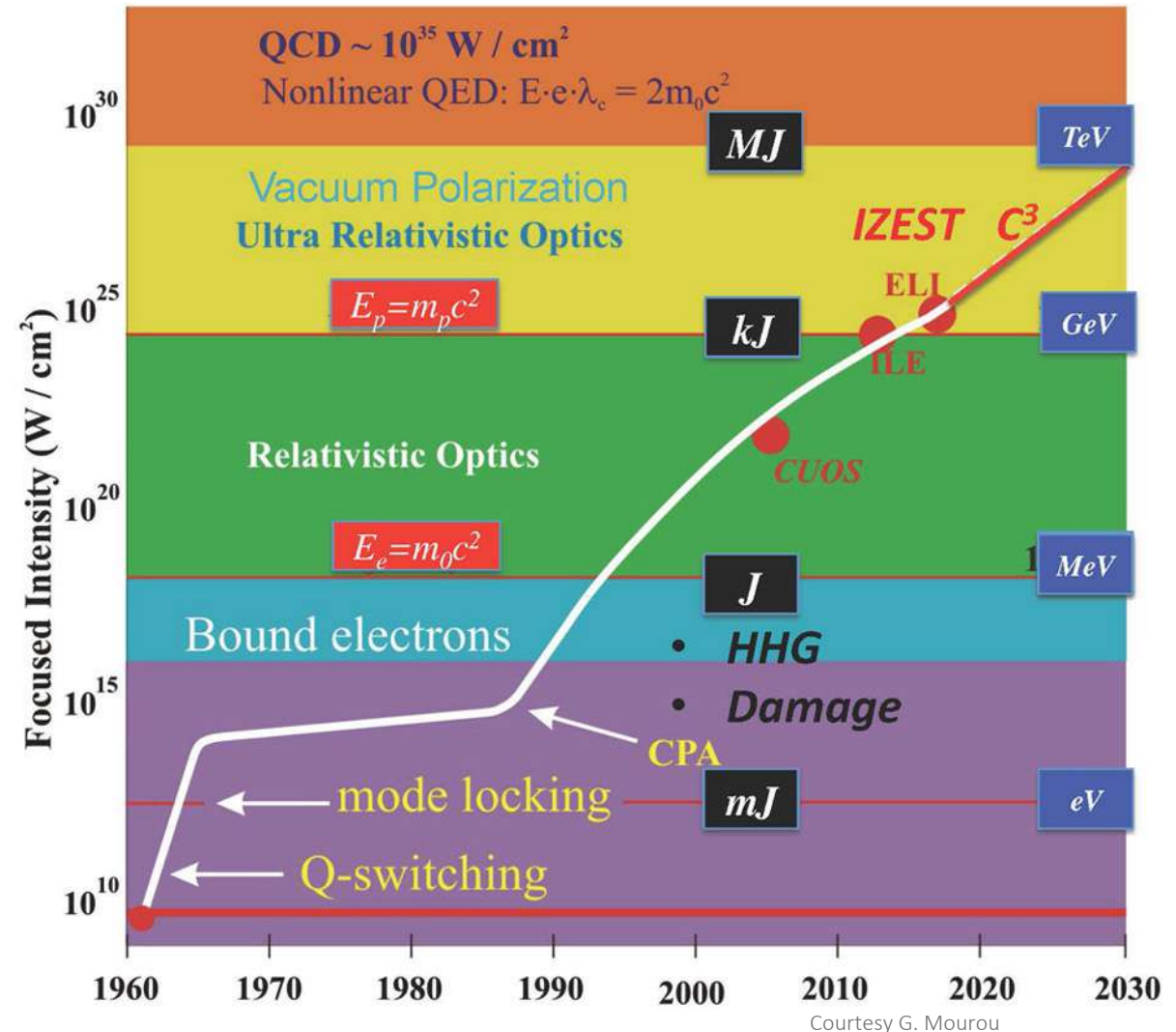
Donna  
Strickland



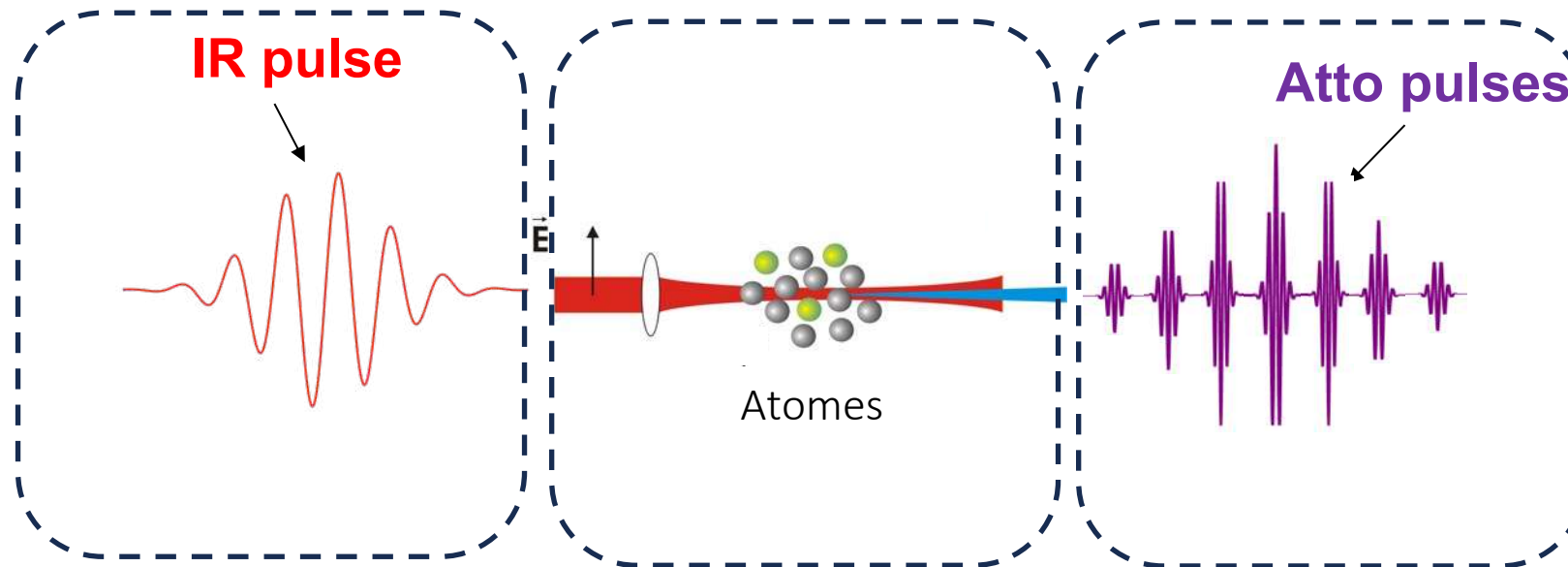
2018

“for their method of generating high-intensity, ultra-short optical pulses”

D. Strickland, G. Mourou, Optics Comm., 56, 219 (1985)



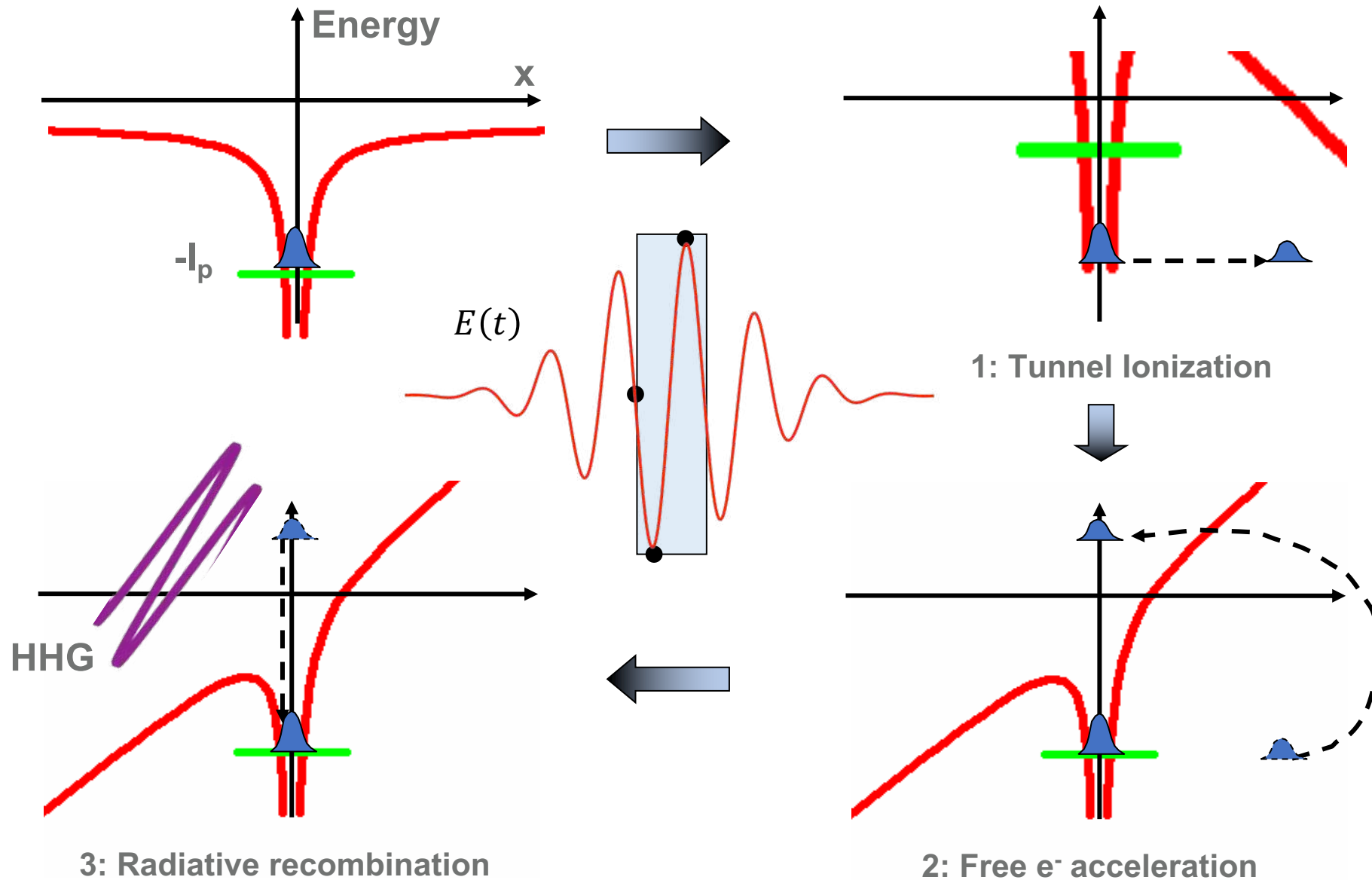
Courtesy G. Mourou



Laser d'excitation

Mécanisme de  
génération

Propriétés du  
rayonnement  
attoseconde

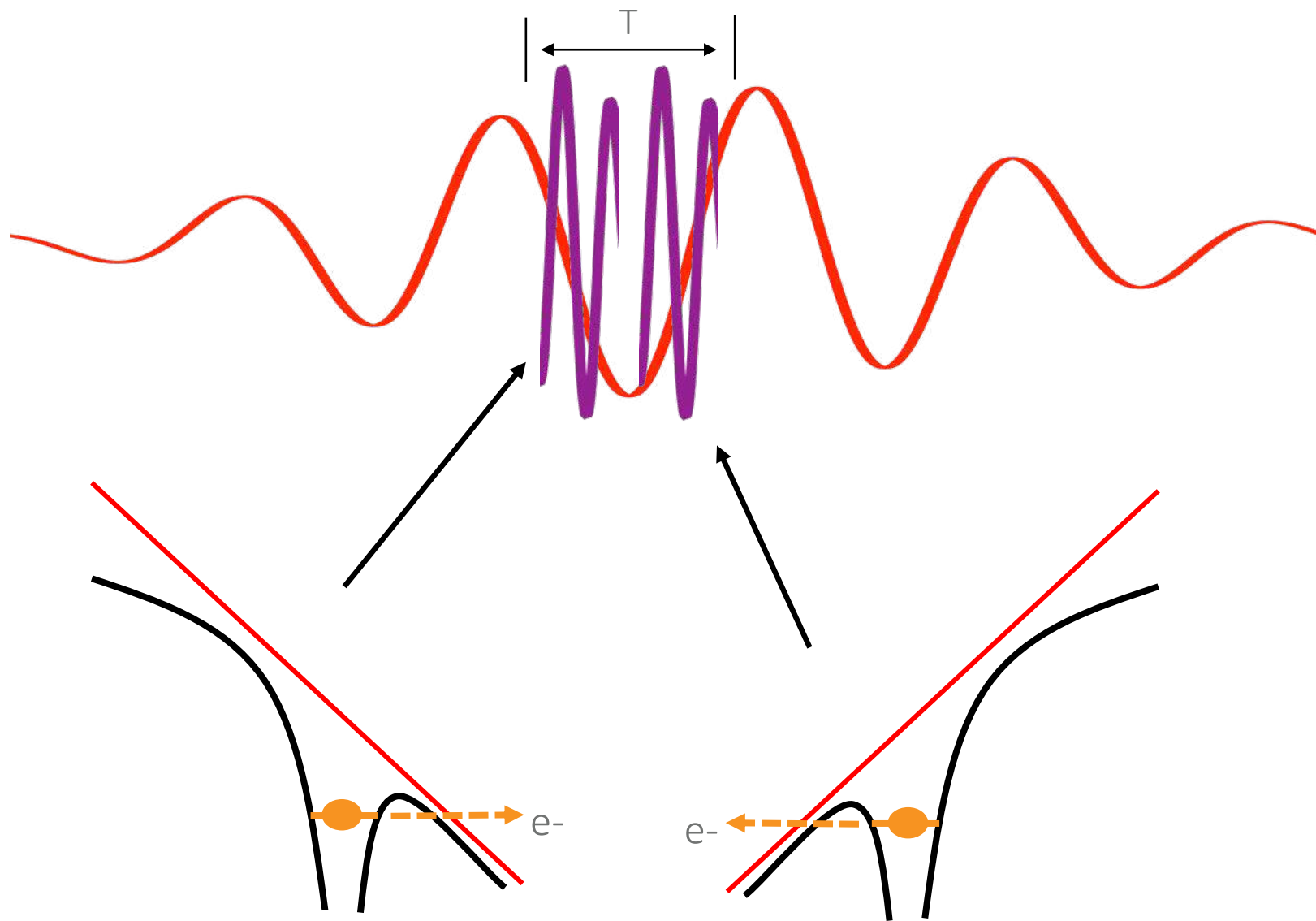


$$V(x,t) = -\frac{Z}{x} + E(t) \cdot x$$

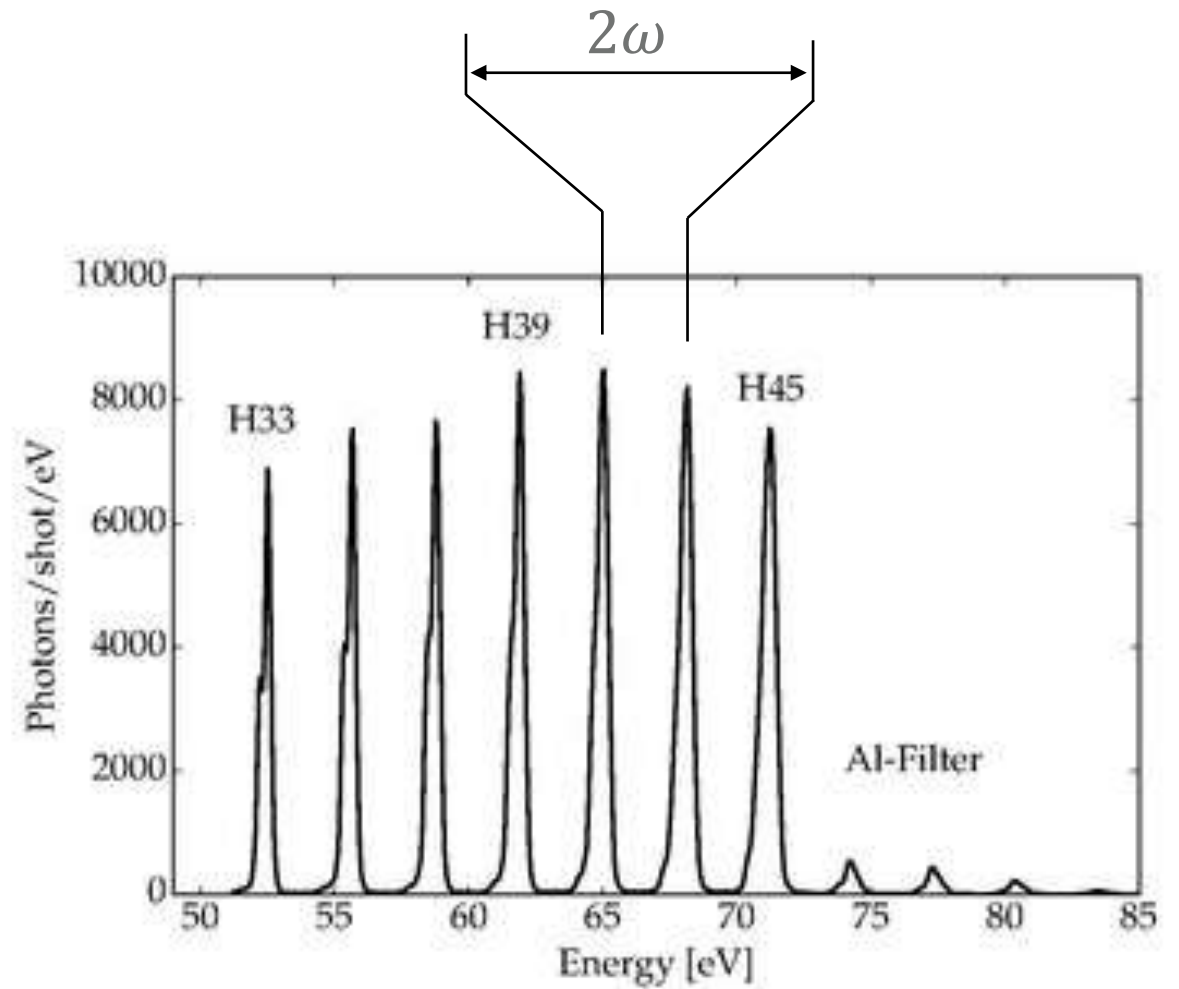
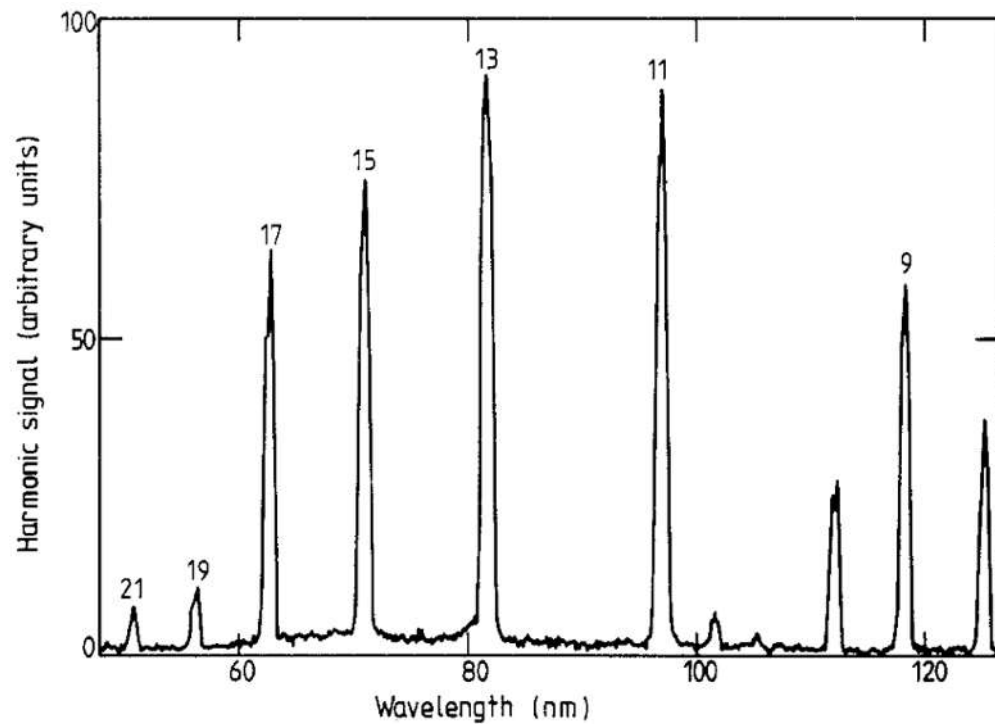
Potentiel  
Coulombien

Interaction  
dipolaire  
électrique





2 émissions par cycle optique

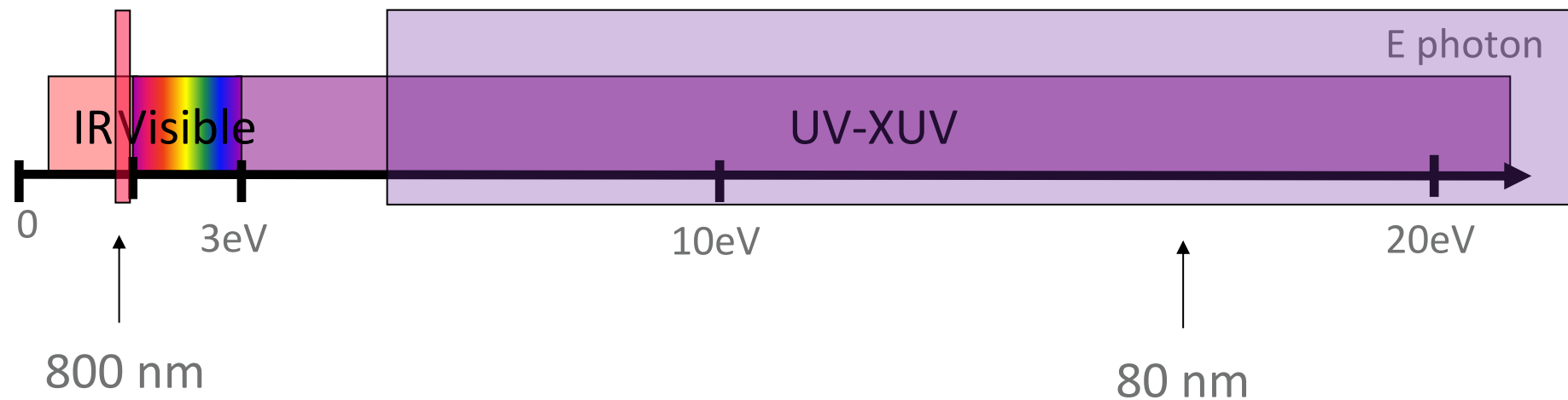


*M. Ferray et al., J. Phys. B 21 (1988)*

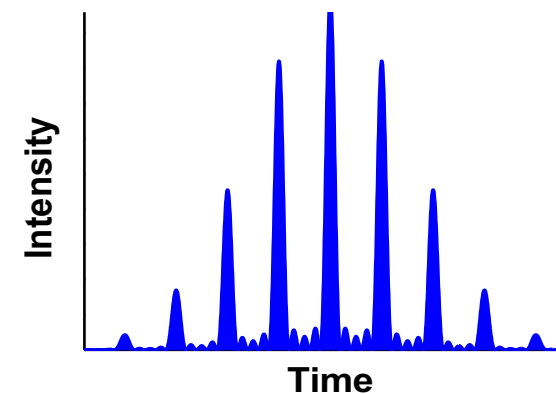
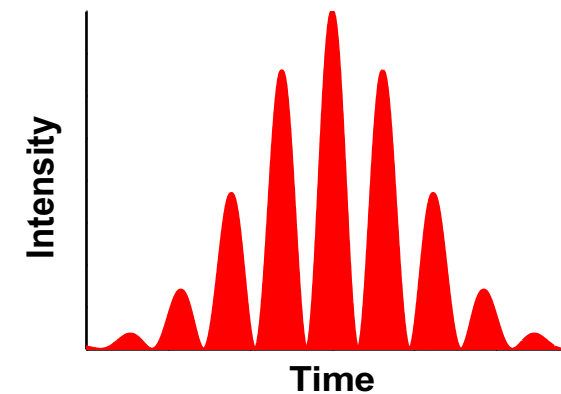
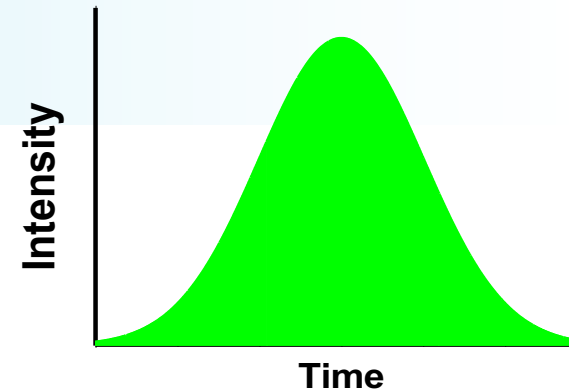
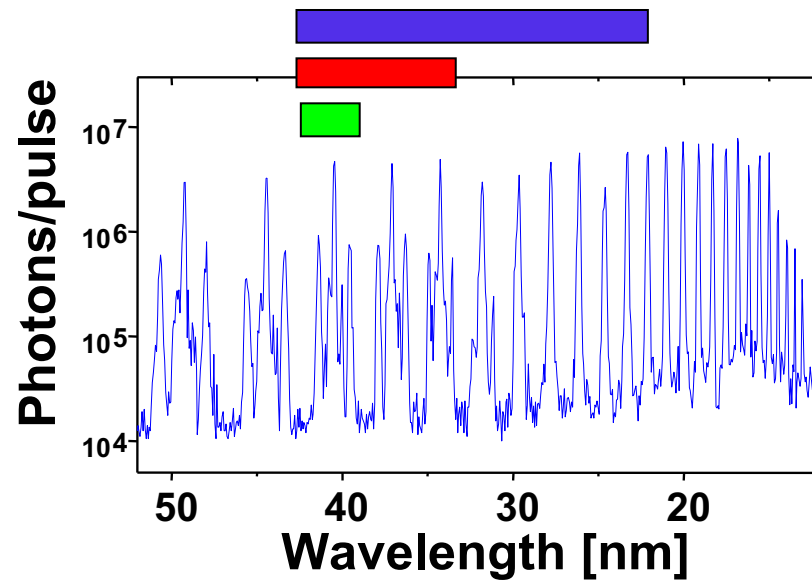
Principe d'incertitude:  $\Delta\tau \propto \frac{1}{\Delta\omega}$

Femto  
 $\Delta\tau = 10 \text{ fs}$   
 $\Delta\omega = 0.18 \text{ eV}$

Atto  
 $\Delta\tau = 100 \text{ as}$   
 $\Delta\omega = 18 \text{ eV}$

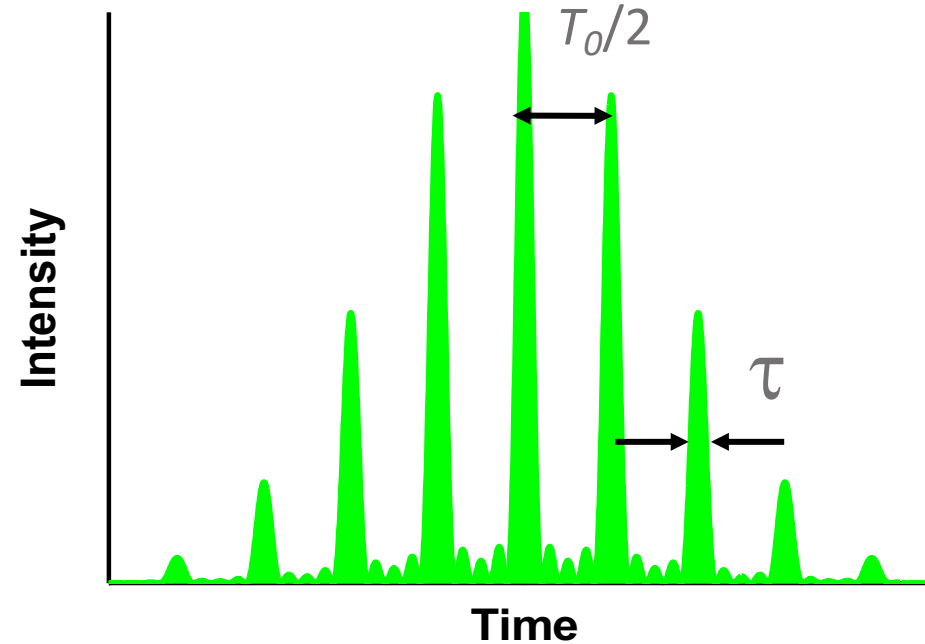
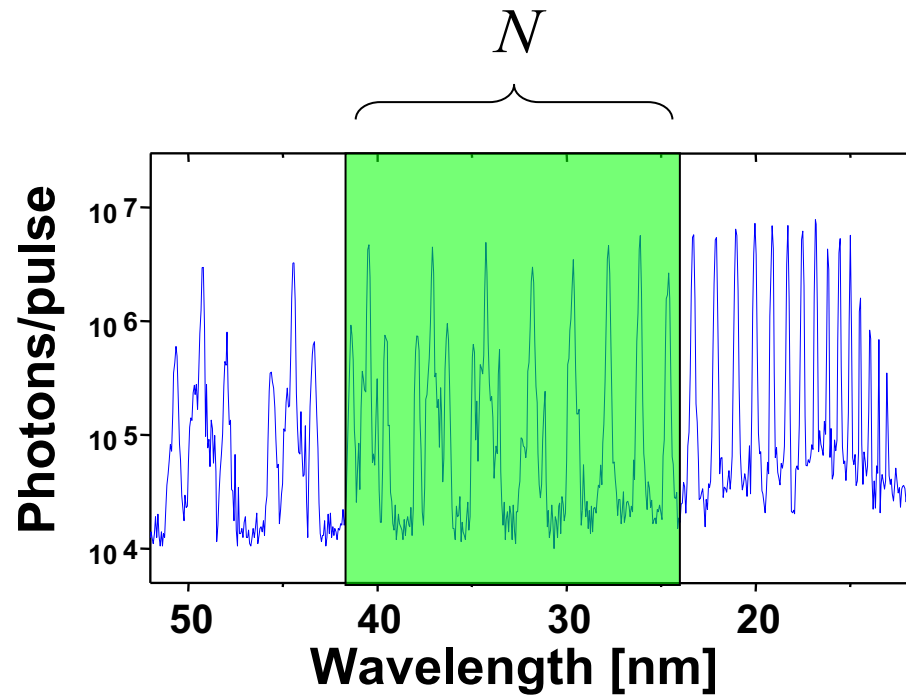


Spectres ultra-larges dans l'XUV



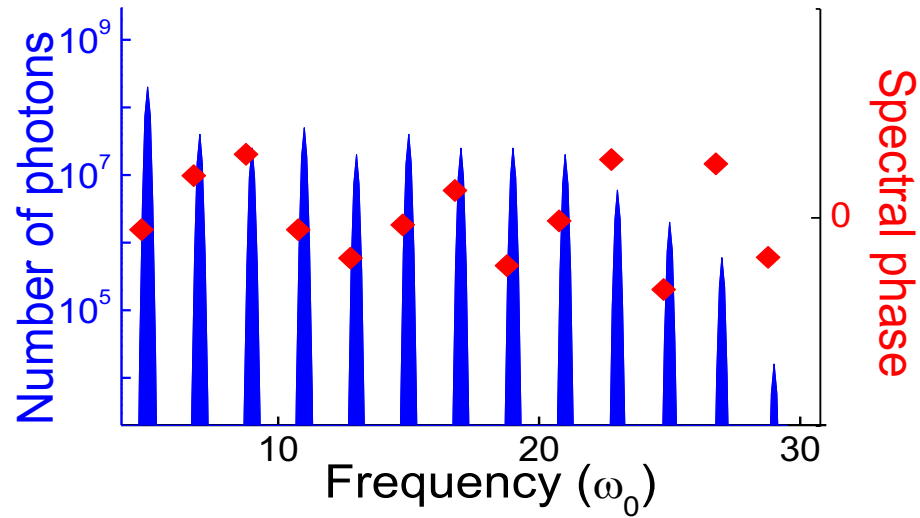
Superposition cohérente de:  $\omega_q$

$$I(t) = \left| \sum_N A_q(t) \exp(-i\omega_q t) \right|^2$$



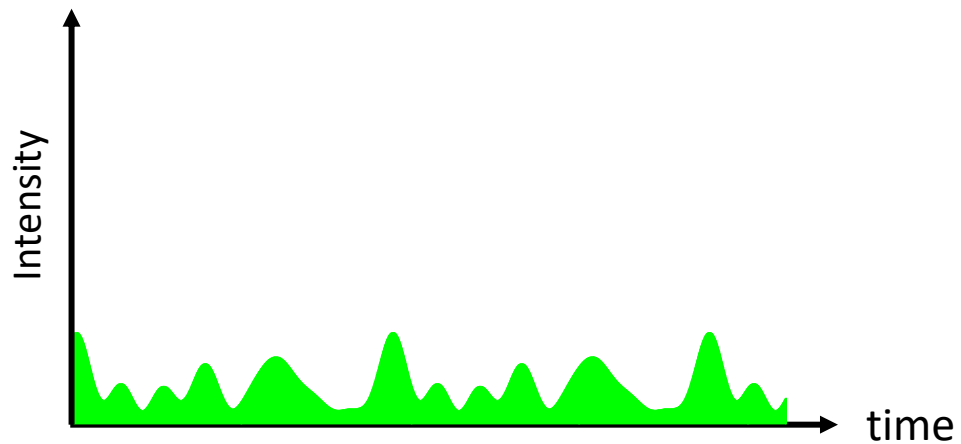
Durée des impulsions atto :  $\tau \approx \frac{T_0}{2N}$

Train d'impulsions attoseconde

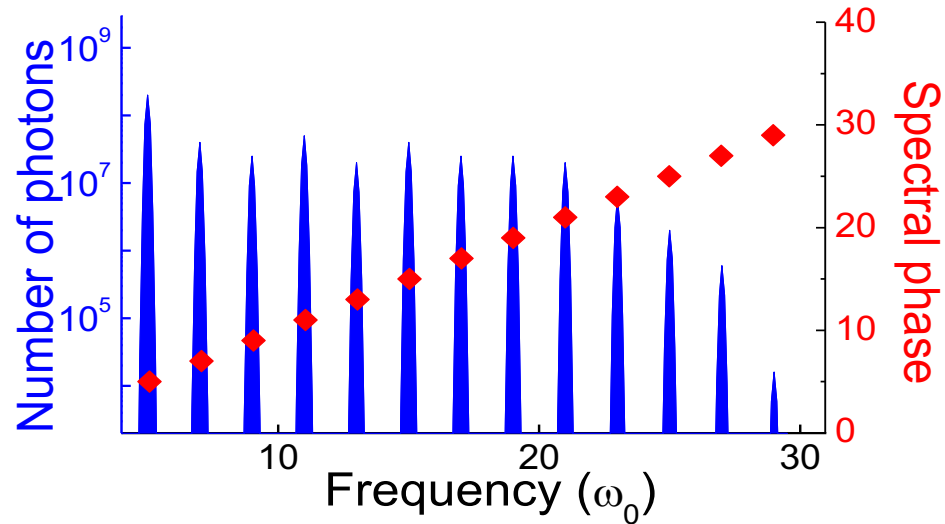


Phases aléatoires

$$I(t) = \left| \sum_N A_q(t) \exp(-i\omega_q t + i\varphi_q) \right|^2$$

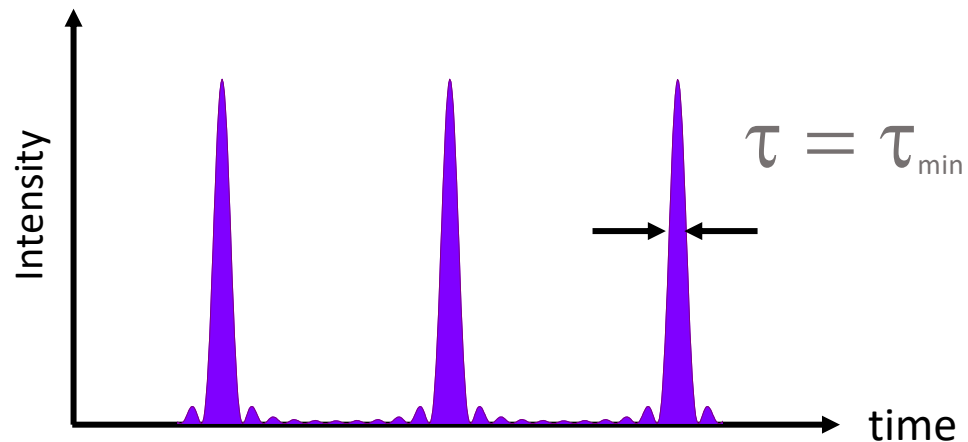


Emission continue chaotique



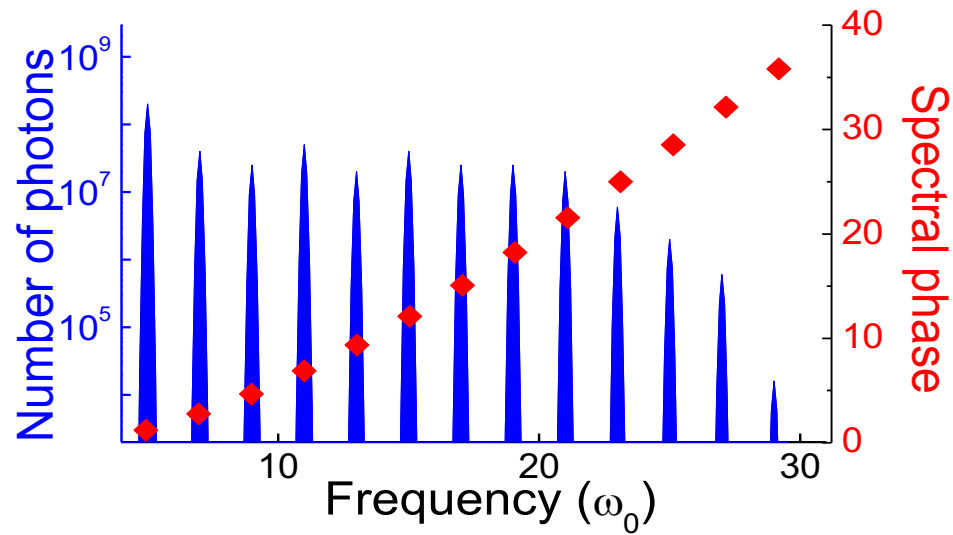
Dépendance linéaire:  $\varphi_q = \omega_q t_e$

$$I(t) = \left| \sum_N A_q(t) \exp(-i\omega_q t + i\varphi_q) \right|^2$$



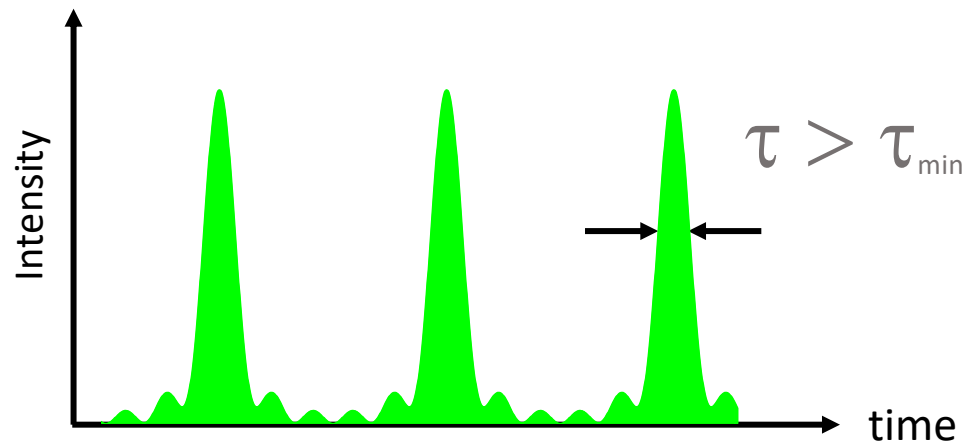
Emission attoseconde synchrone

(verrouillage de phase)



Dépendance quadratique:  $\varphi_q \approx \alpha \omega_q^2$

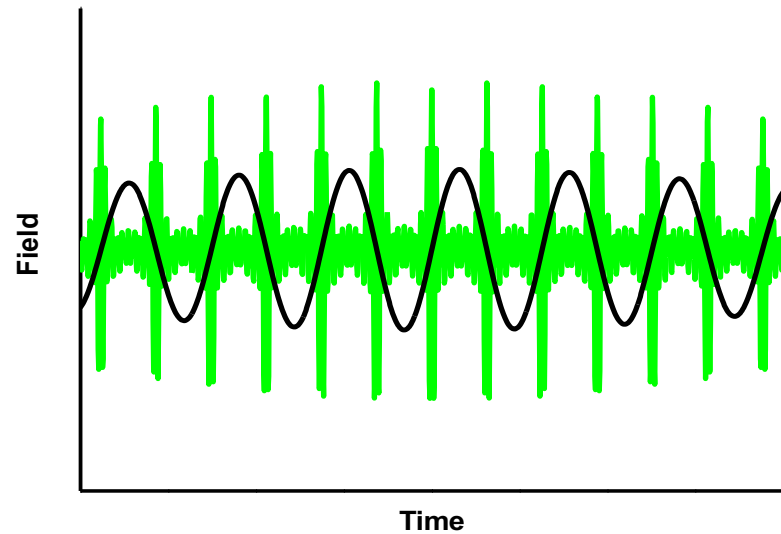
$$I(t) = \left| \sum_N A_q(t) \exp(-i\omega_q t + i\varphi_q) \right|^2$$



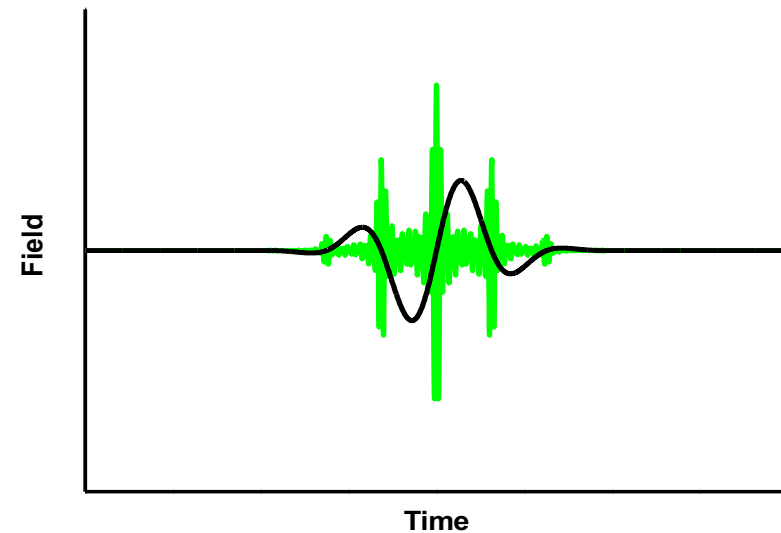
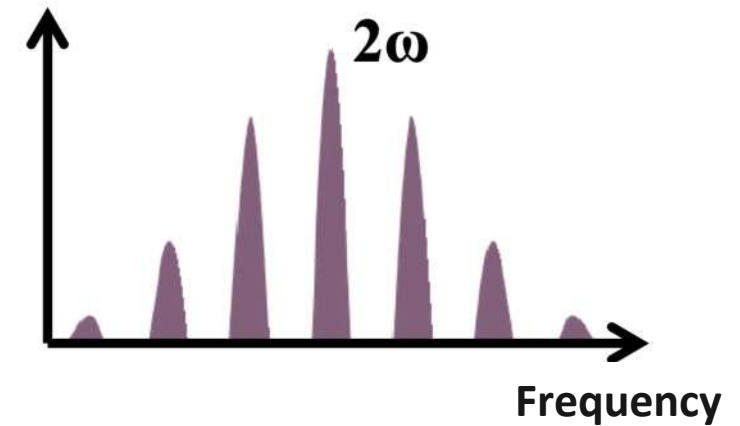
Emission attoseconde synchrone

(verrouillage de phase)

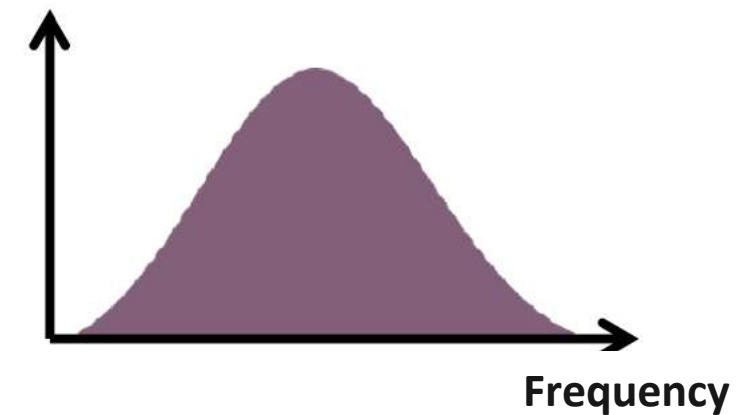




Durée du laser IR : 30 fs  
 → Train d'impulsions atto  
 → Peigne de fréquence

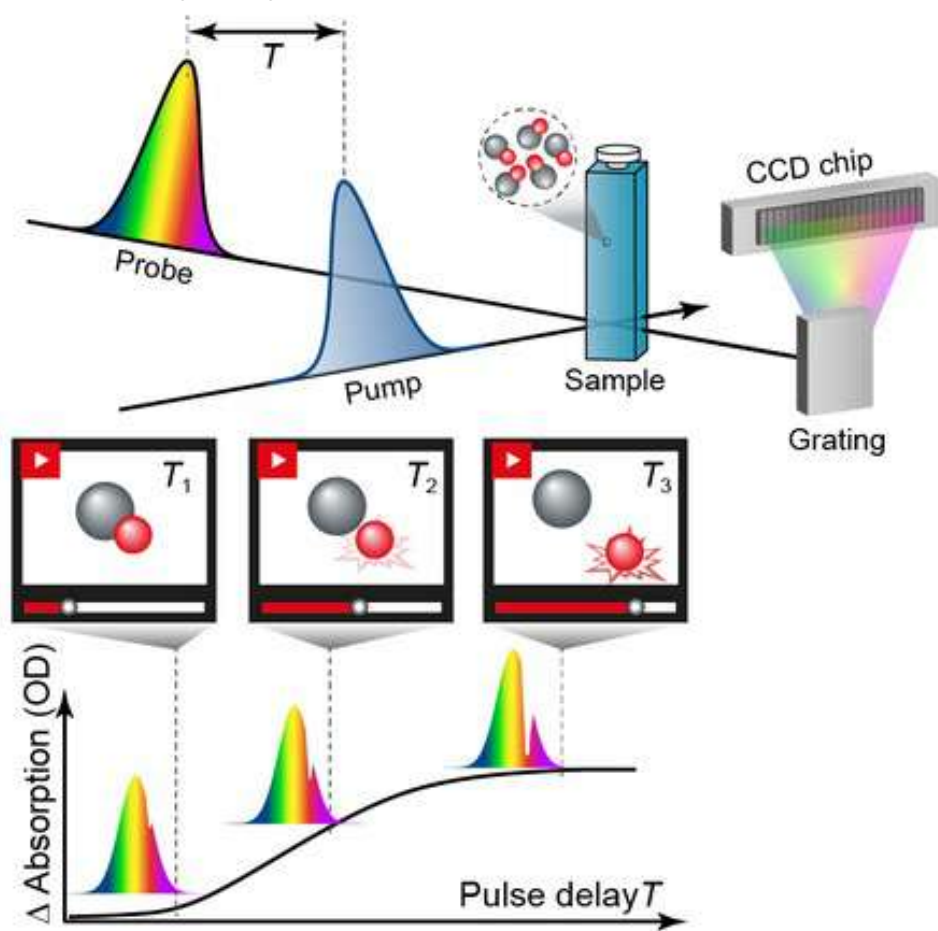


Durée du laser IR : 4 fs  
 → Impulsion atto unique  
 → Spectre continu



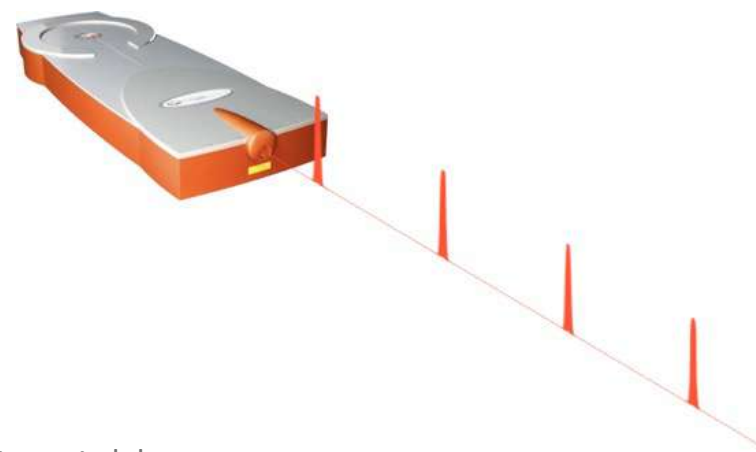
Besoin de laser IR de quelques cycles optiques

Schéma pompe-sonde

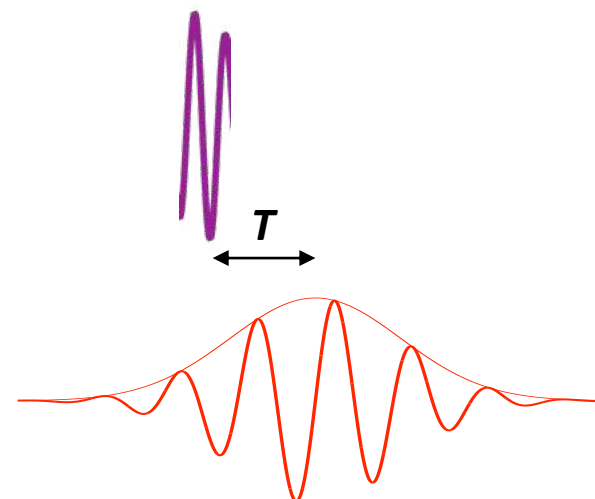


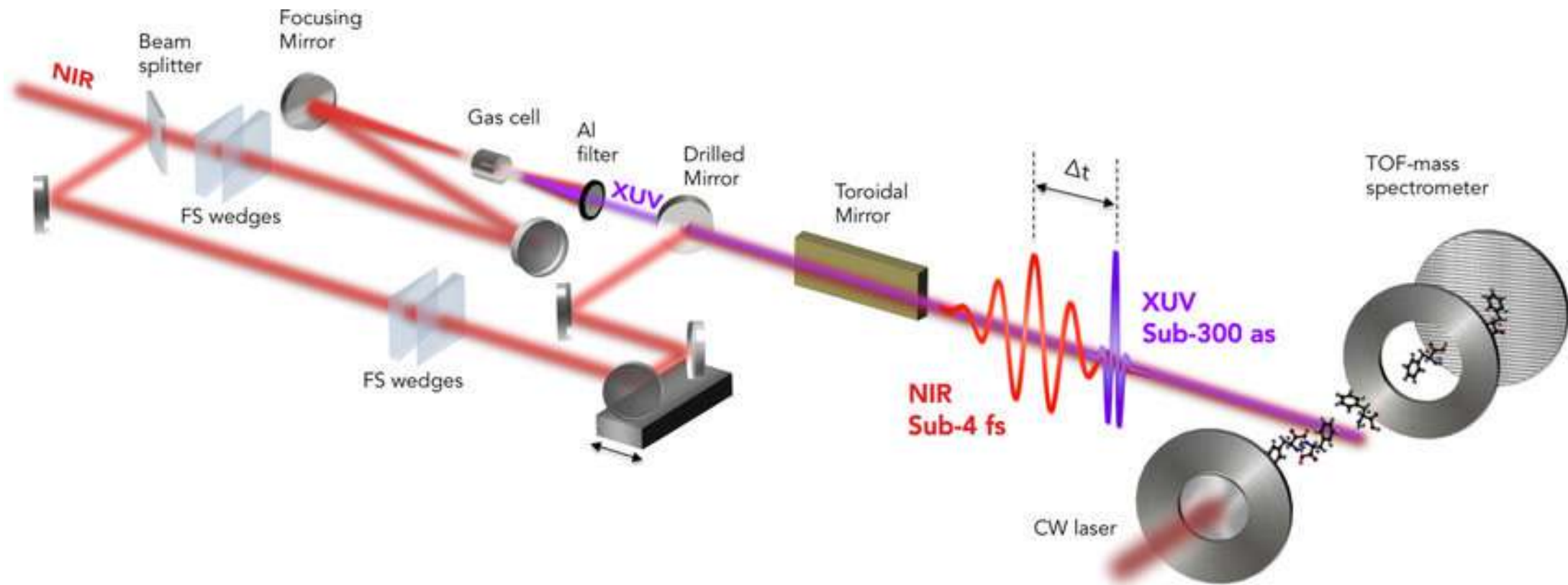
T. Brixner, Universität Würzburg

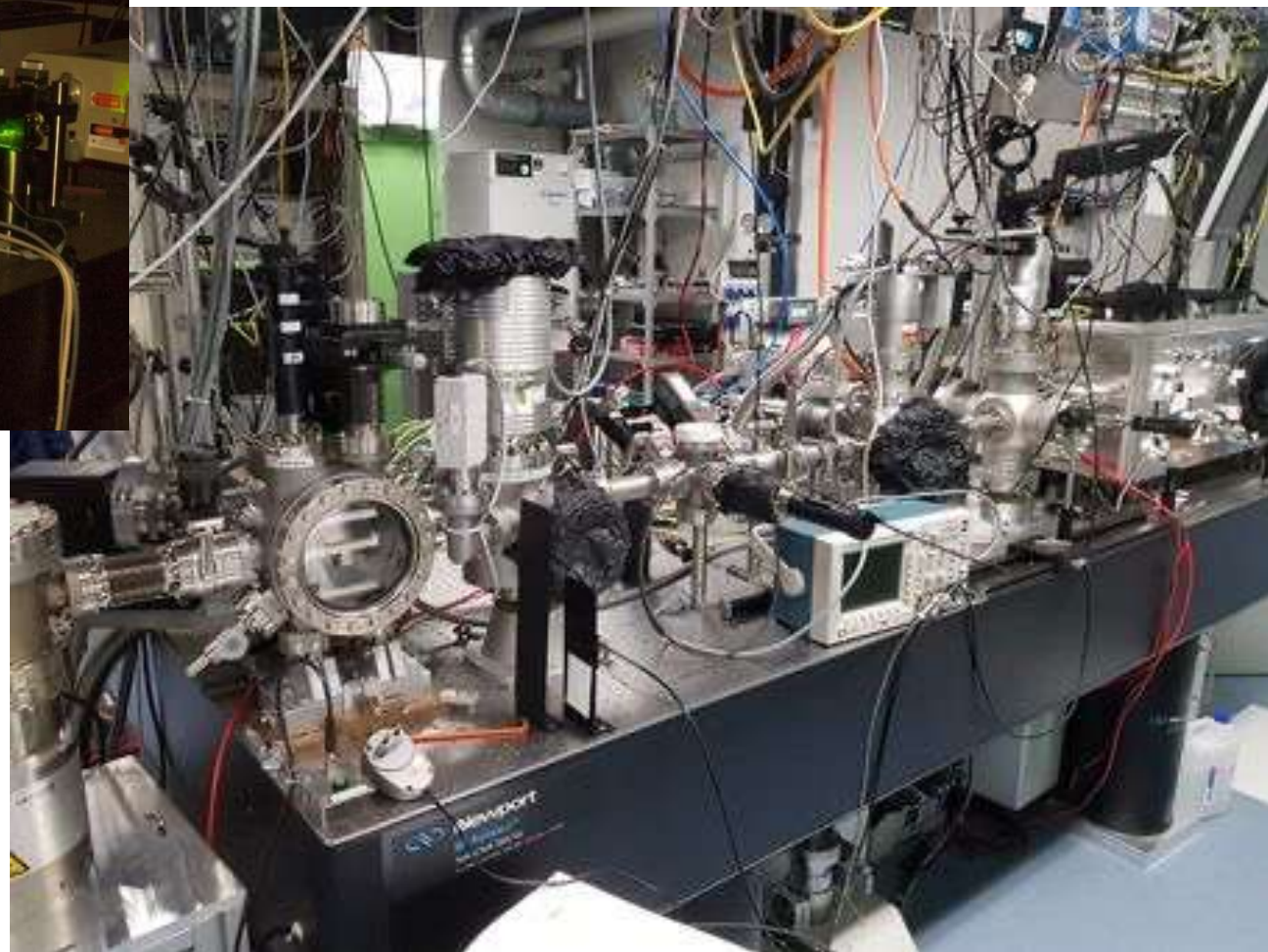
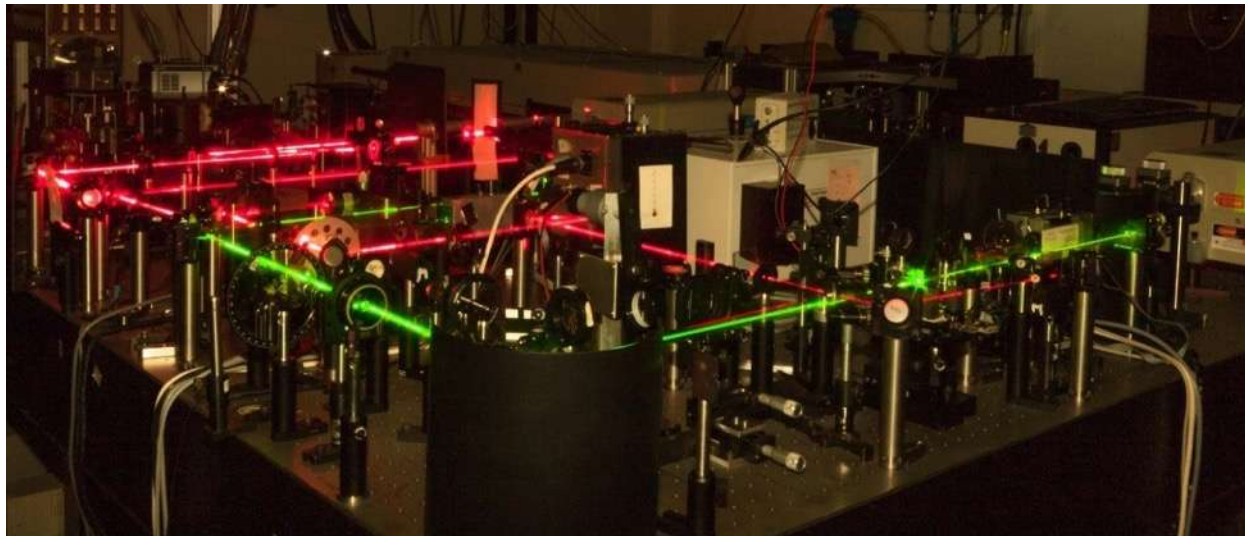
Laser pulsé haute cadence



Délai variable

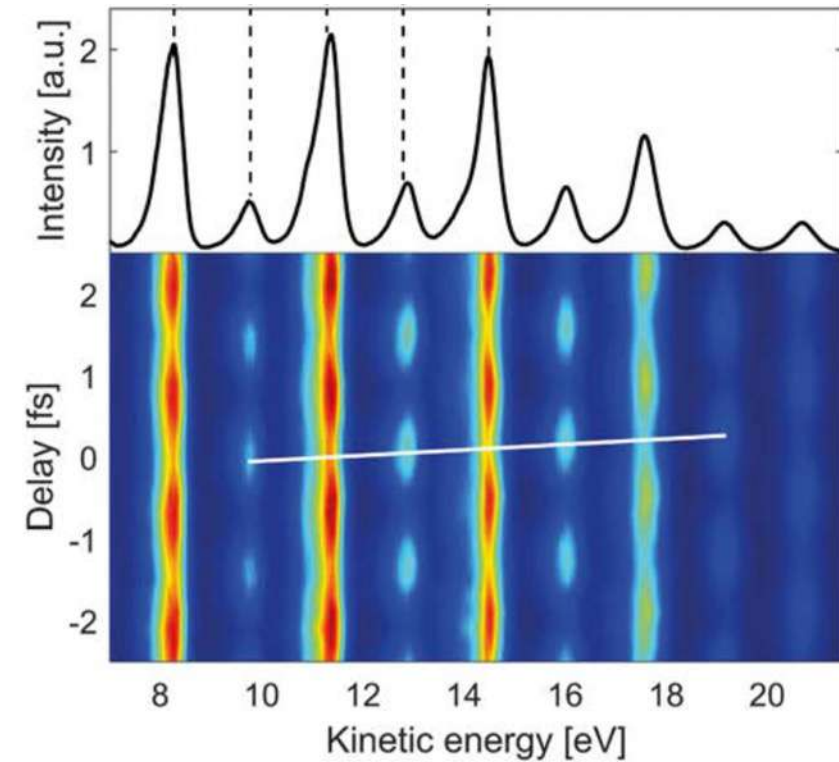
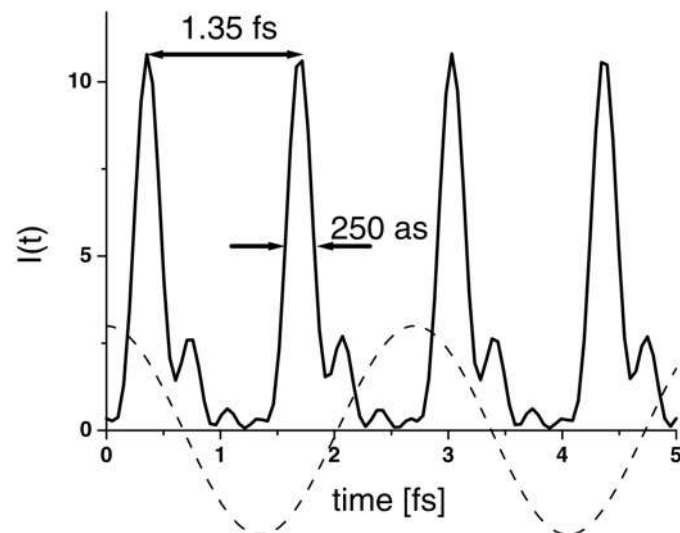
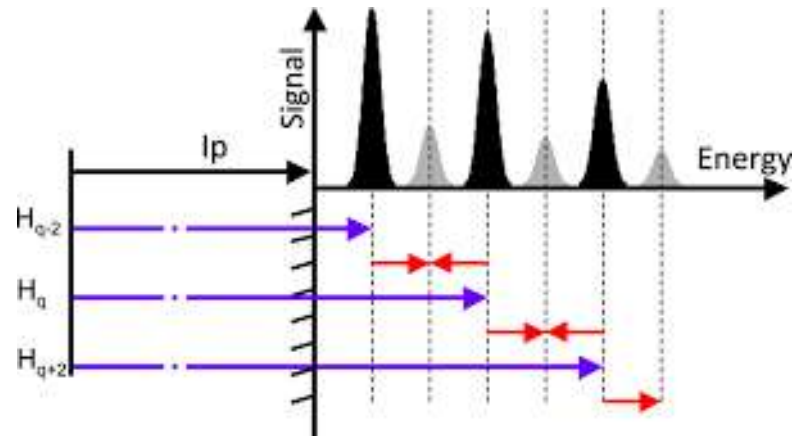






## Rabbit (Reconstruction of Attosecond Bursts by Interference of Two-photon Transitions)

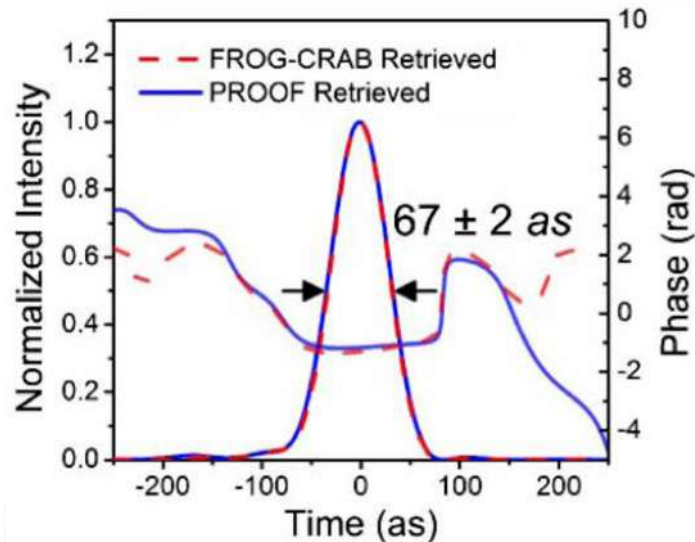
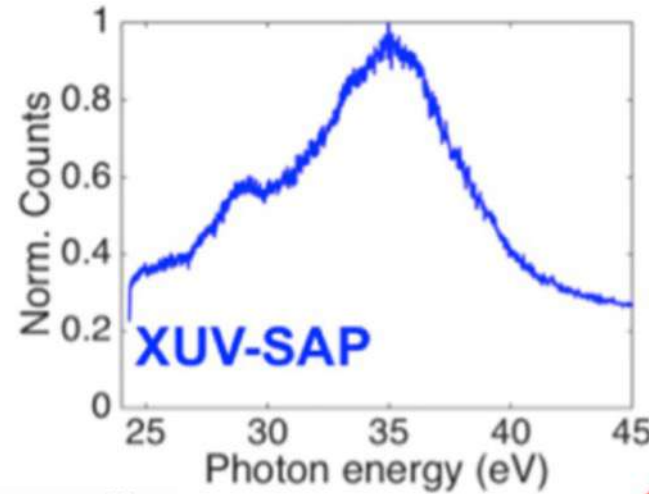
Interférométrie quantique



$$I(t) = \left| \sum_N A_q(t) \exp(-i\omega_q t + i\varphi_q) \right|^2$$

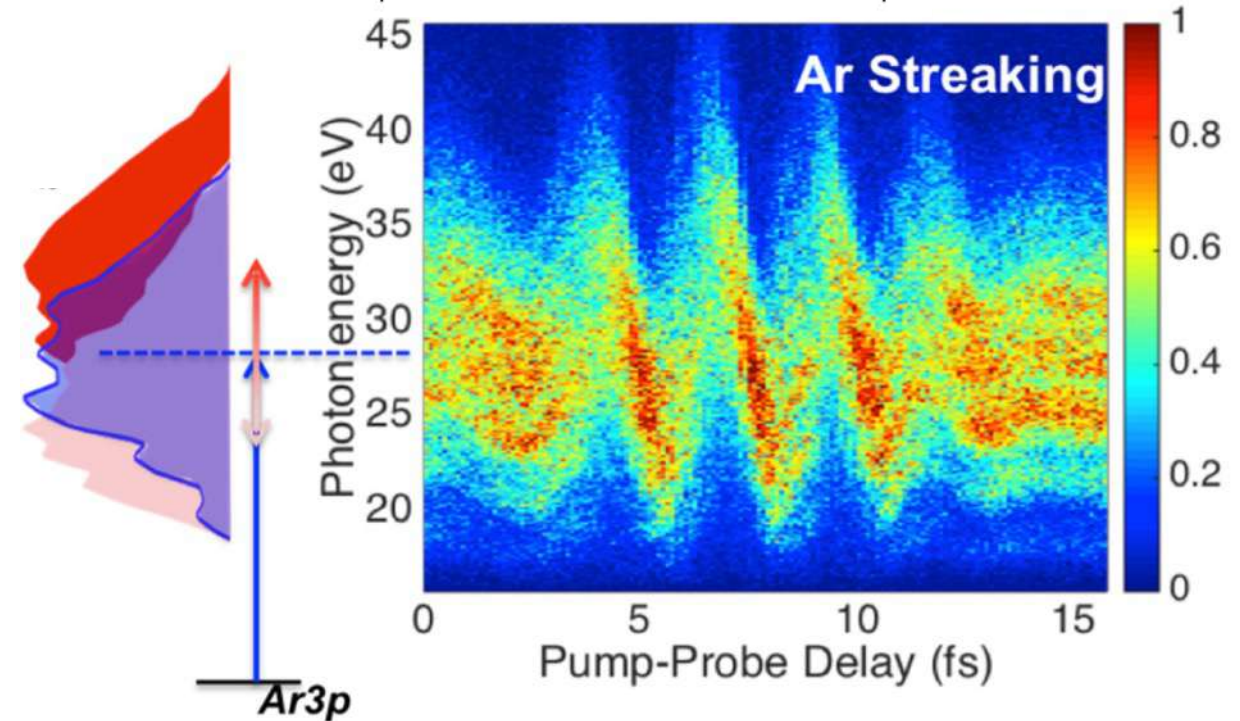
*P. M. Paul et al., Science 292 (2001)*

## Electron Energy Streaking and FROG-CRAB reconstruction



*K. Zhao et al.,  
Opt. Lett. 37,  
3891 (2012)*

$$S_{FROG}(\omega, t_d) = \left| \int_{-\infty}^{+\infty} P(t-t_d)G(t)e^{i\omega t} dt \right|^2$$



*L. Cattaneo et al., Opt. Exp.. 24, 29060 (2016)*

$$S_{FROG}(\omega, t_d) = \left| \int_{-\infty}^{+\infty} P(t - t_d) G(t) e^{i\omega t} dt \right|^2$$

## Mesure du champ électrique infrarouge

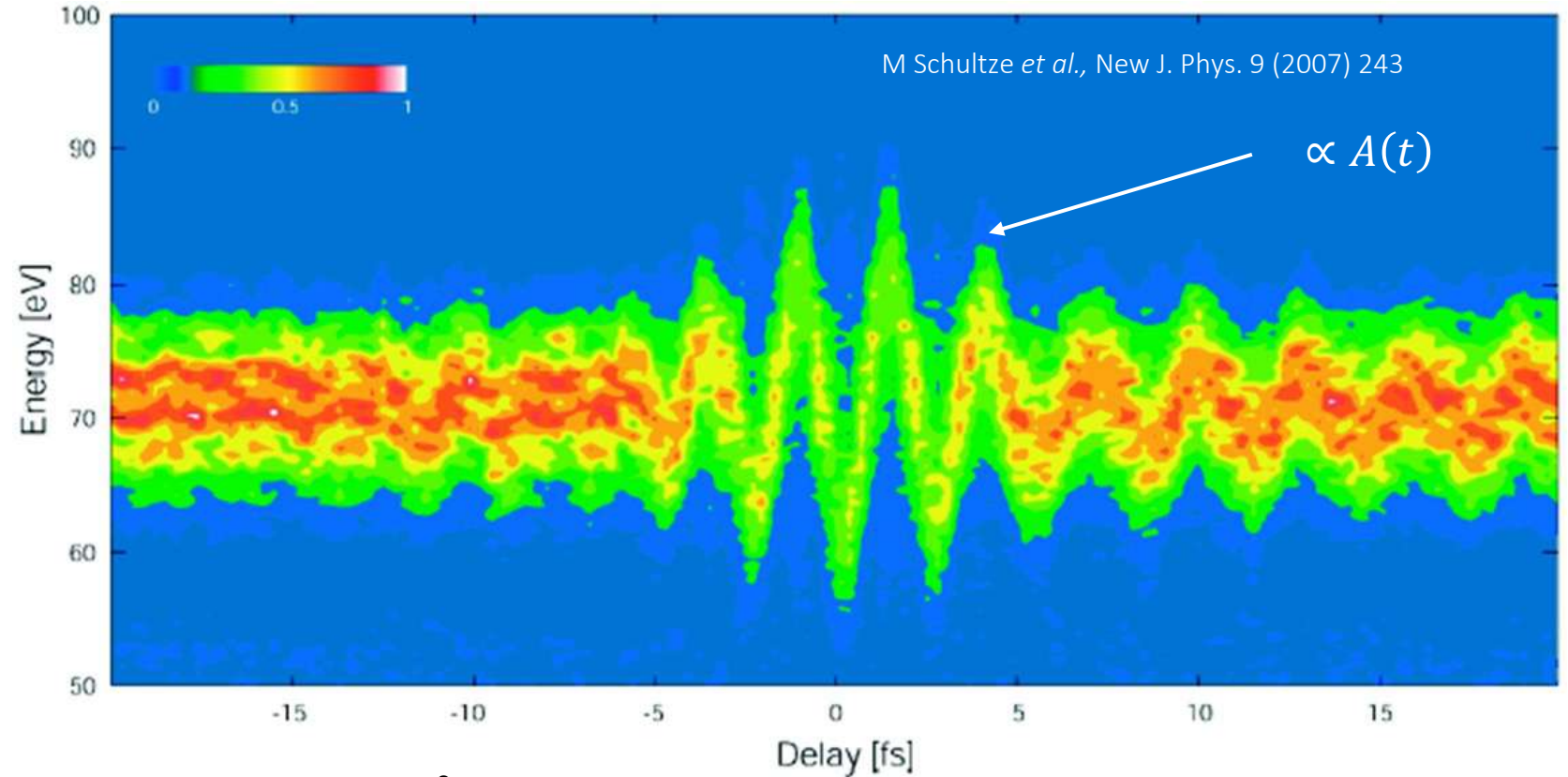
$$E(t) = -\frac{\partial A(t)}{\partial t}$$



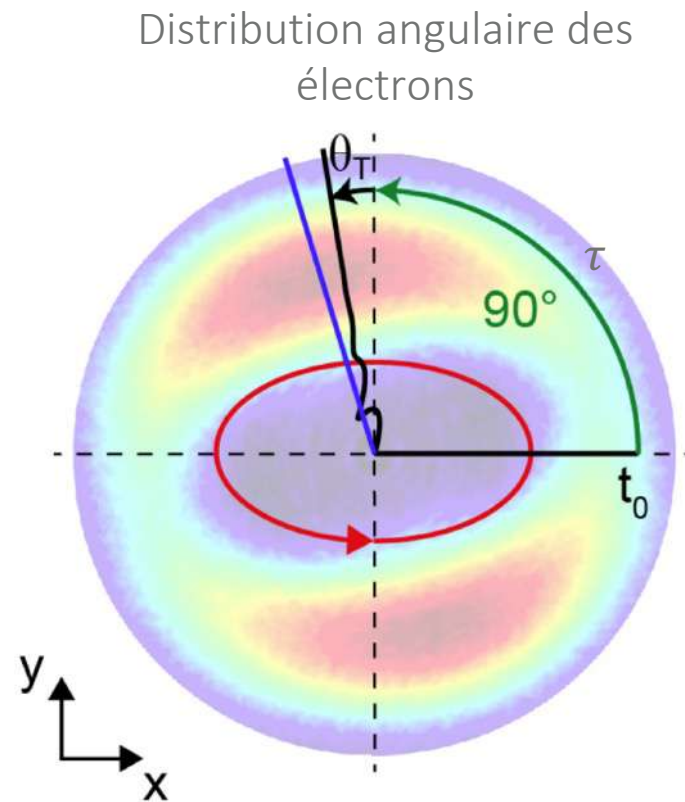
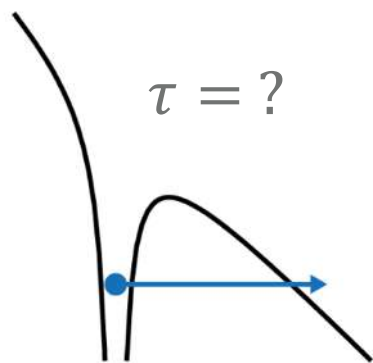
Time varying amplitude

$$E(t) \approx E_0 e^{-\left(\frac{t}{\tau}\right)^2} \cos(\omega t + \varphi)$$

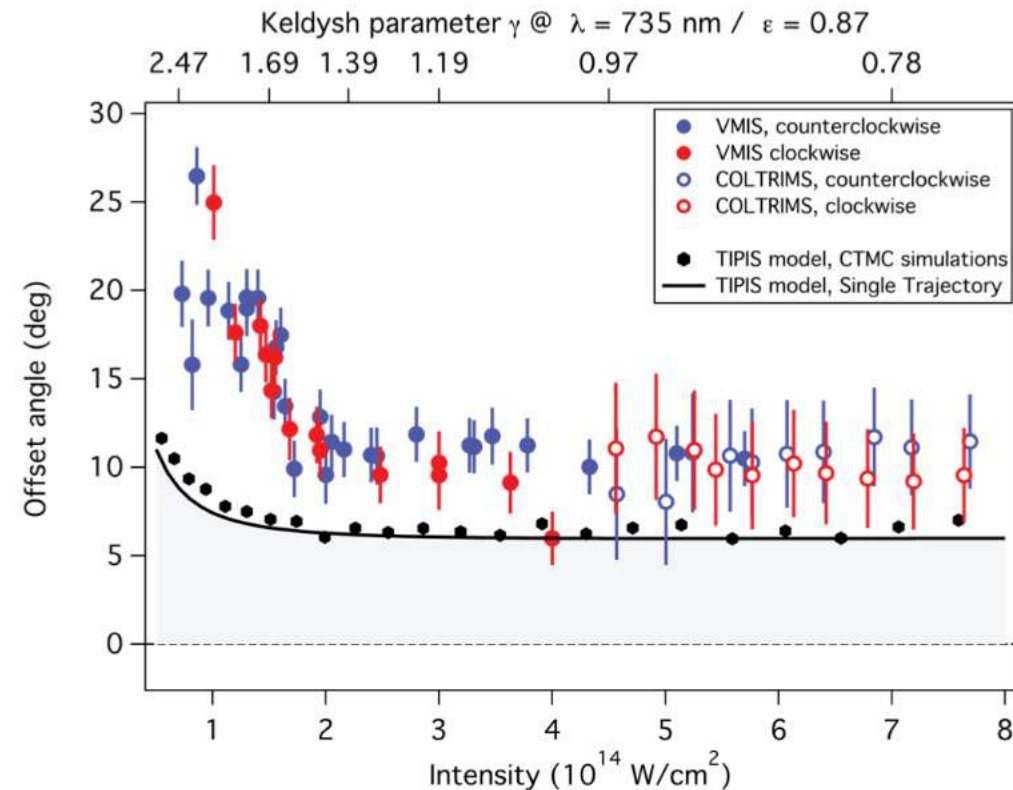
Time varying phase



## Dynamique de l'ionisation tunnel

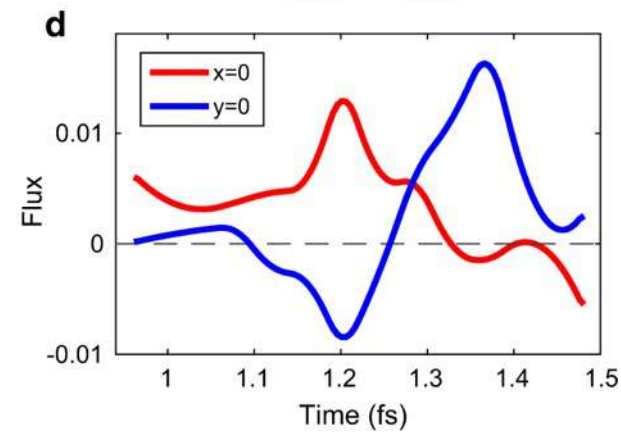
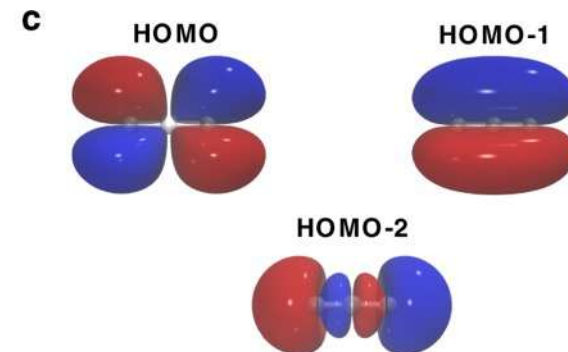
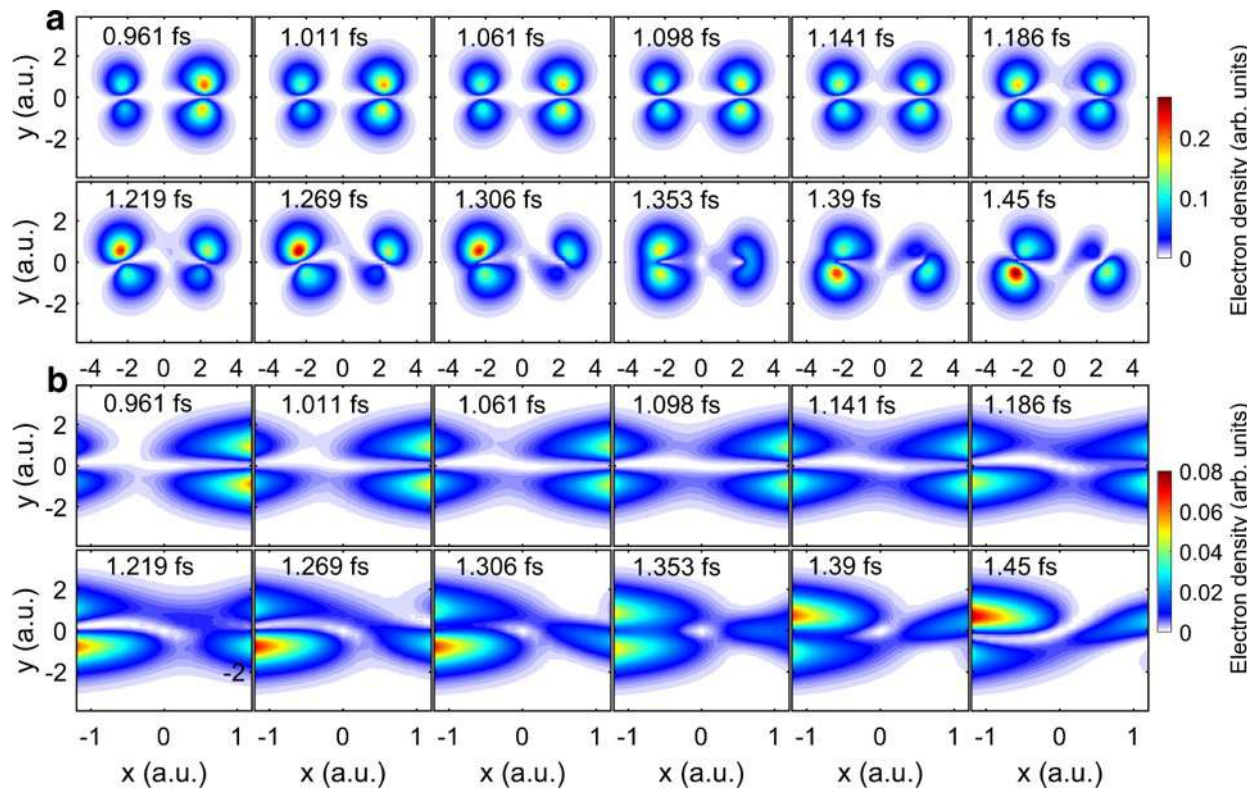
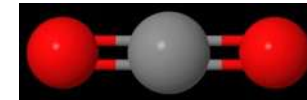


1° pour 7 as





## Dynamique de transfert de charge dans $\text{CO}_2^+$



He, L., *Nat Commun* **13**, 4595 (2022)

Merci

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