# Lecture Series Buenos Aires

**18-3-2024 until 22-3-2024**

## Lecture M3 – High Harmonic Generation

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# Interactions of atoms with strong laser fields

# Distortion of the Coulomb potential

Over-the-barrier ionization:

$$I_{laser,OTB} = \frac{IP^4}{16}$$

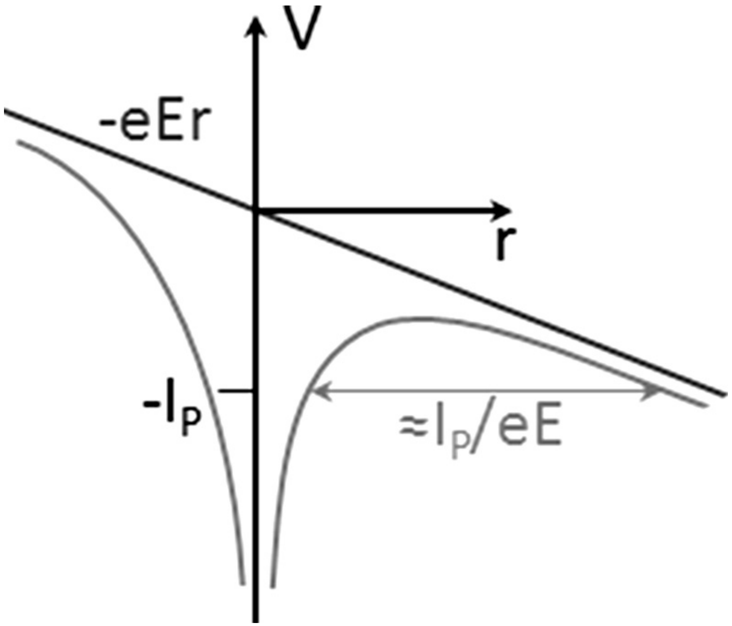
**1 a.u. = 3.51x10$^{16}$ W/cm$^2$**

Hydrogen atom (IP=0.5 a.u.) : $I_{laser} = 0.0039$ a.u. = 1.4x10$^{14}$ W/cm$^2$

Below $I_{laser,OTB}$ the electron can escape the atom by tunneling through the Coulomb + laser electric field potential, provided that the potential is sufficiently quasi-static

This condition is expressed by the Keldysh parameter $\gamma$

$$\gamma = \sqrt{\frac{IP}{2U_p}} \ll 1$$



# Tunneling formulas

Provided suitable approximations are made, the rate of tunnel ionization can be described by simple formulas

**Strong field approximation:**
Assume that after the ionization process the interaction of the electron with the core is negligible, and that the electron only interacts with the laser electric field

**Adiabatic approximation:**
Assume that in the presence of the laser field the atom remains in the lowest available state, and that no population is transferred to excited states

**Single active electron approximation:**
Assume only the most weakly bound electron is ionized

# After ionization: Propagation assuming the strong-field approximation (SFA)

Assume that the electron does not feel the ion anymore as soon as it has tunneled out

Assume, moreover, that the Coulomb-free motion starts with v=0 at r=0, and that the laser amplitude is constant

$a(t) = E_0 cos\omega t$ (a.u.)

$v(t) = v_0 sin\omega t + v_{0z}$ $\qquad$ $v_0 = E_0/\omega$ (a.u.)

$z(t) = z_0(-cos\omega t) + v_{0z}t + z_{0z}$ $\qquad$ $z_0 = E_0/\omega^2$(a.u.)

N.B. $\mathrm{E}(t) = -dA/dt, i.e. v_0 = A_0$

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

# Canonical momentum

$$E(t) = E_0 \cos(\omega t) = -\frac{dA(t)}{dt} \qquad A(t) = -A_0 \sin(\omega t) + constant \qquad A_0 = \frac{E_0}{\omega}$$

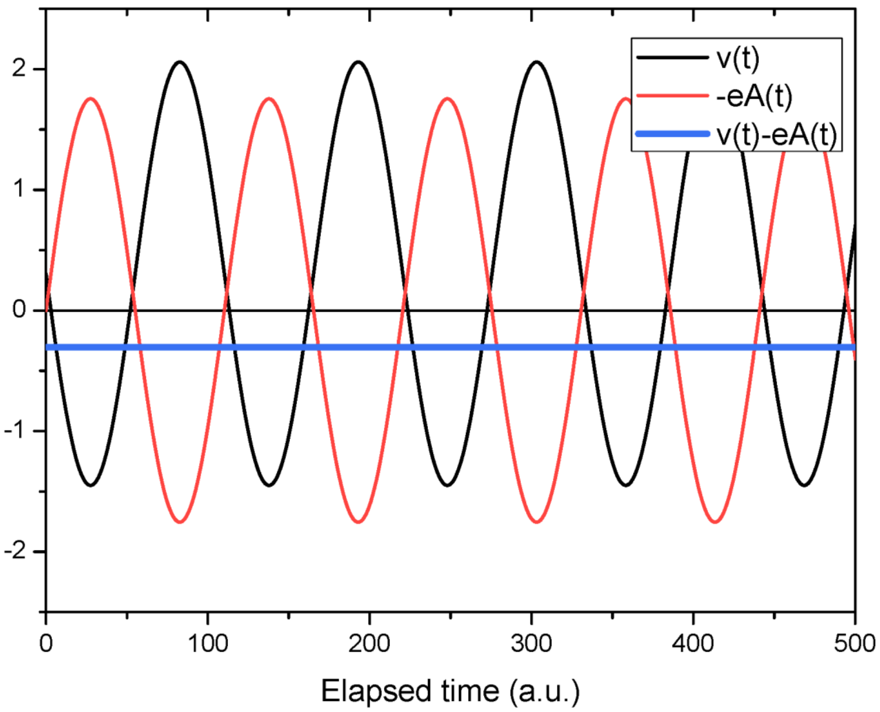
Vector potential

$$v(t) = v_0 + \int_{t_0}^{t} a(t)dt = v_0 + \int_{t_0}^{t} \frac{-eE(t)}{m} dt = v_0 + \frac{e}{m}(A(t) - A(t_0))$$

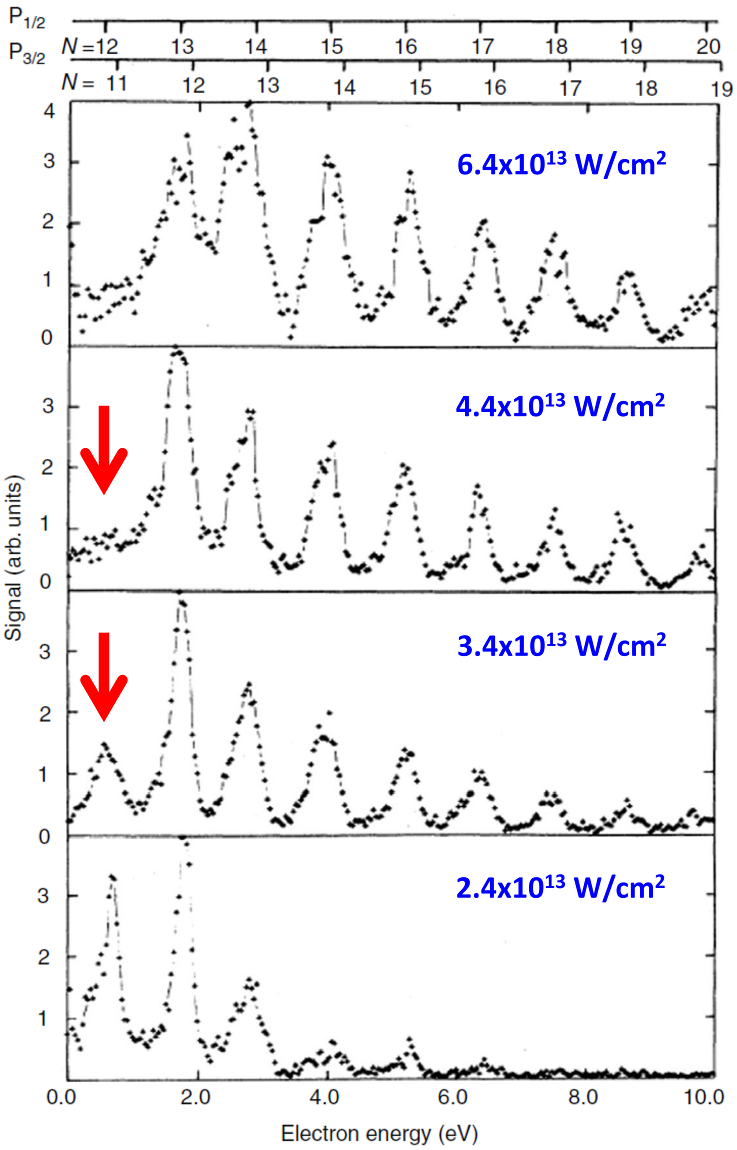
$$v(t) - \frac{e}{m}A(t) = v_0 - \frac{e}{m}A(t_0)$$

In a strong laser field the conserved quantity is the canonical momentum:

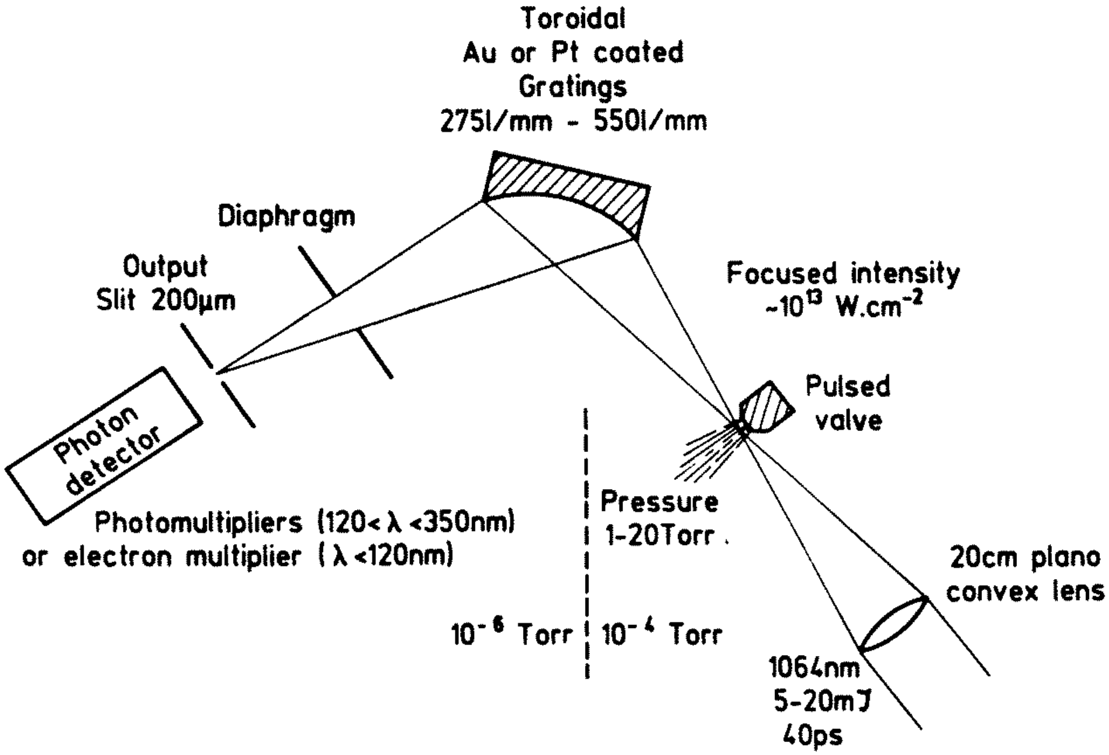
**p** = m**v**(t) – eA(t)



# Mid-1980´s: studies of ATI



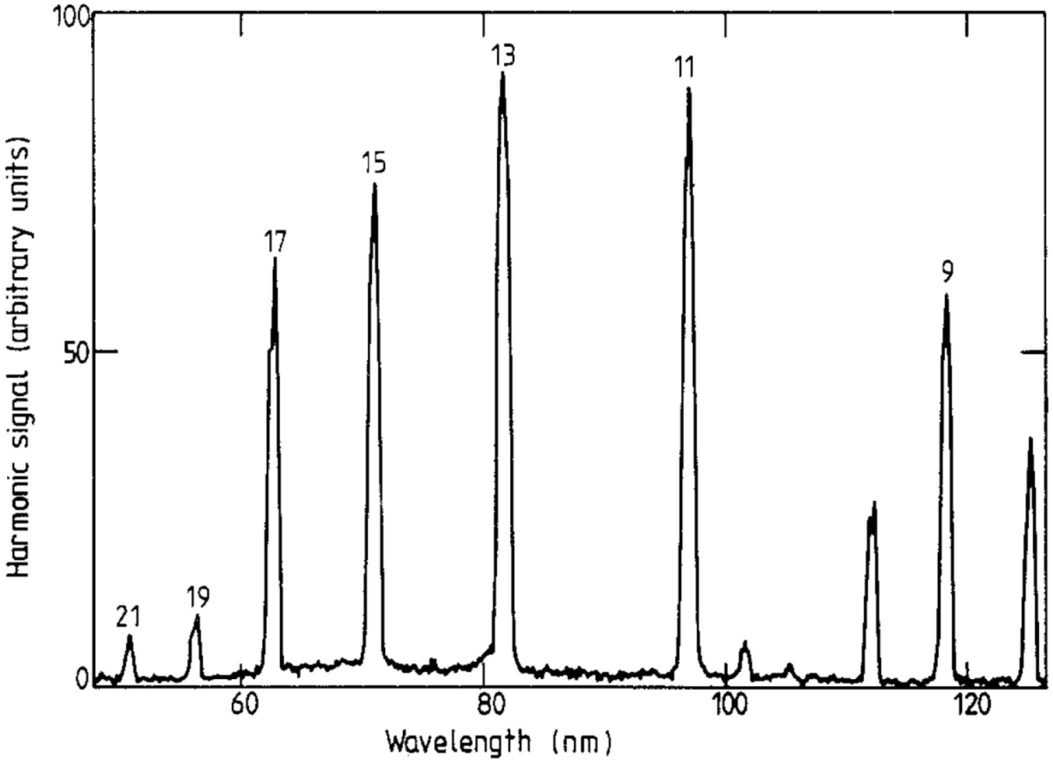
**P. Kruit et al., Phys. Rev. A 28, 248 (1983)**



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

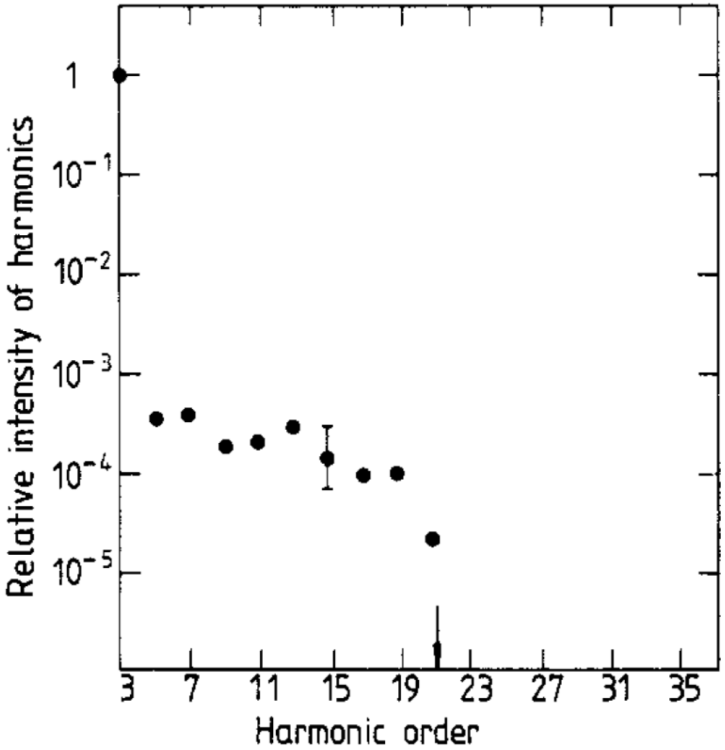
**A. L´Huillier et al., in ‘Atoms in Intense Laser Fields ’, edited by Gavrila and Muller, (Academic Press, 1992)**

# Discovery of High-Harmonic Generation (HHG)



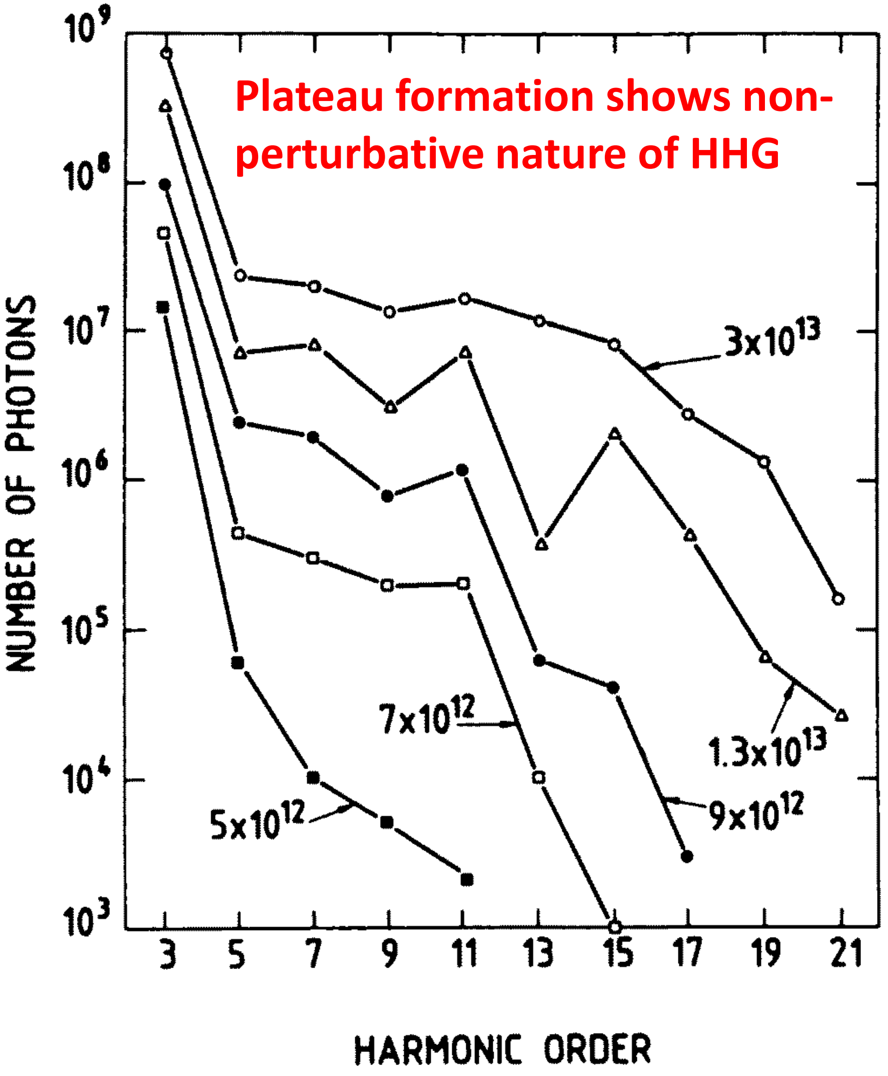
High-harmonic generation in Xe using a 30 ps, 1064 nm laser focused to ca. $10^{13}$ W/cm$^2$

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

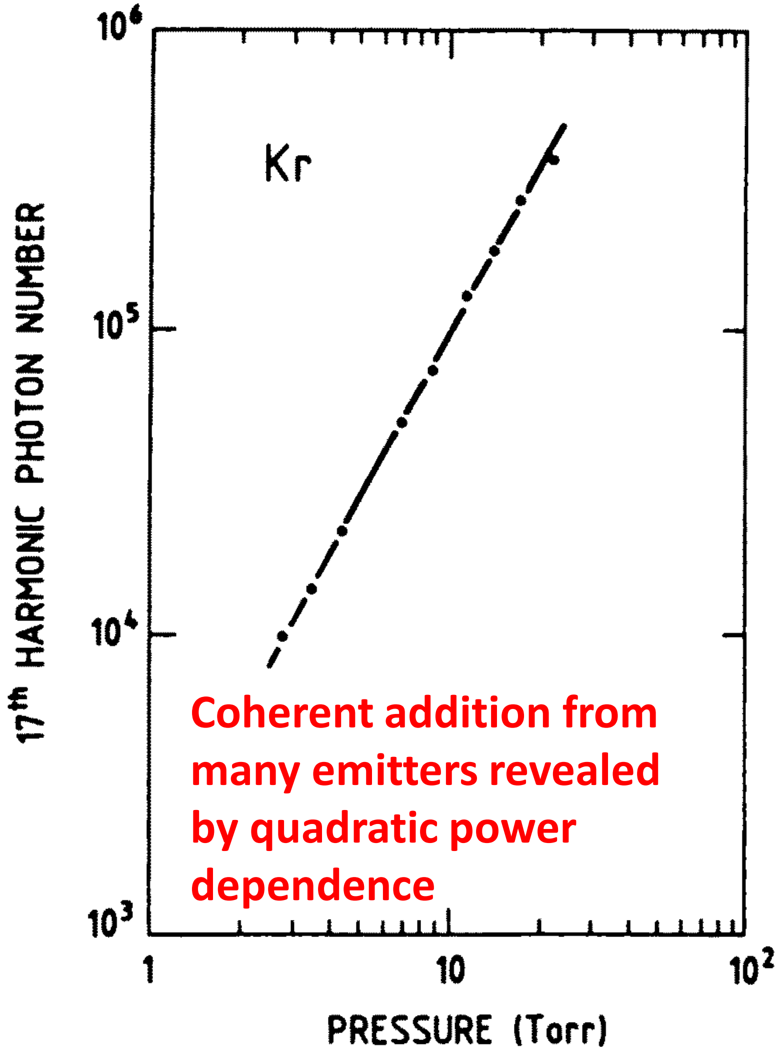


+ similar observations around the time in the Rhodes-group using 248 nm driver lasers

# Intensity and pressure dependence



Plateau formation shows non-perturbative nature of HHG



Coherent addition from many emitters revealed by quadratic power dependence

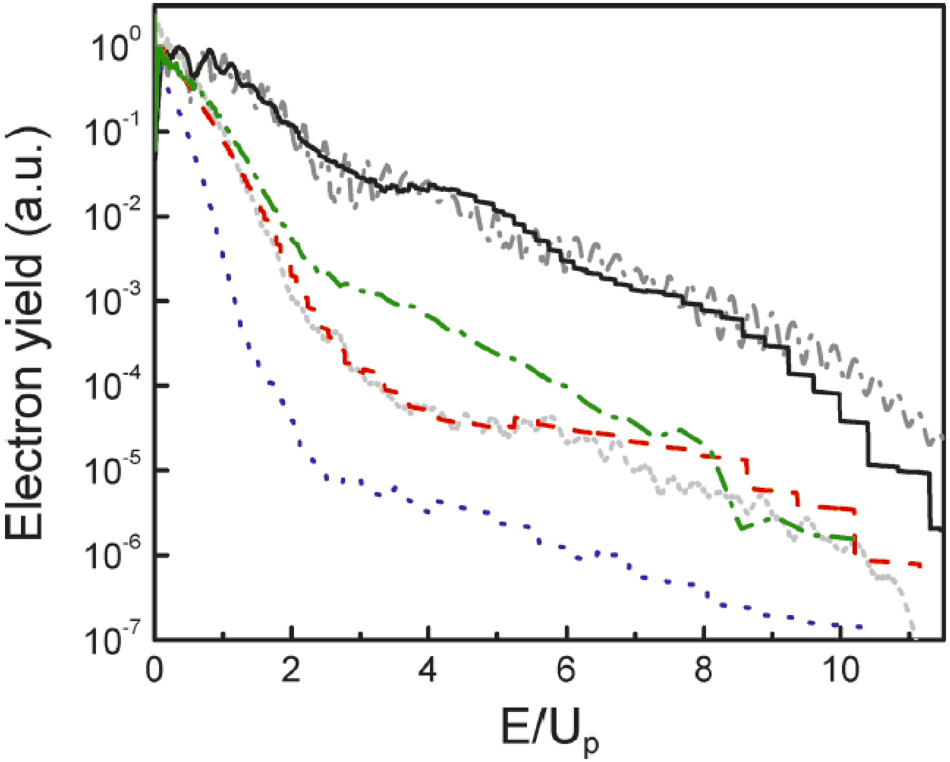
A. L´Huillier et al., in ‘Atoms in Intense Laser Fields ’, edited by Gavrila and Muller, (Academic Press, 1992)

**Remember lecture M2**

# Recollision trajectories

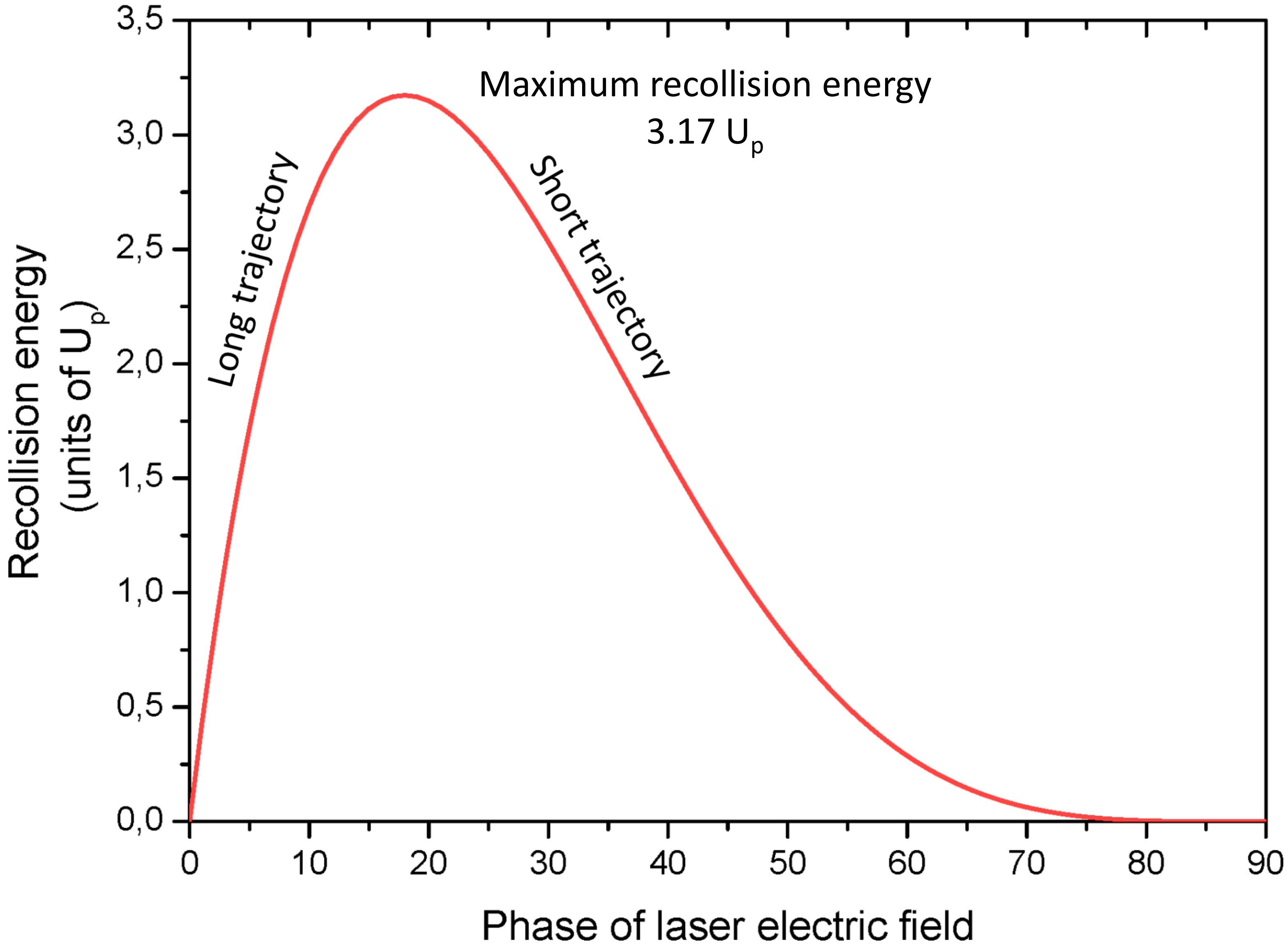
The highest possible return energy is 3.17 $U_p$ = 0.7925 $A_0^2$, corresponding to a velocity of 1.259 $A_0$. This recollision occurs near a zero crossing of the field.

When the recollision flips the sign of the velocity, the field increases the velocity to appr. -2.259 $A_{0,}$ corresponding to an energy of 10.2 $U_p$

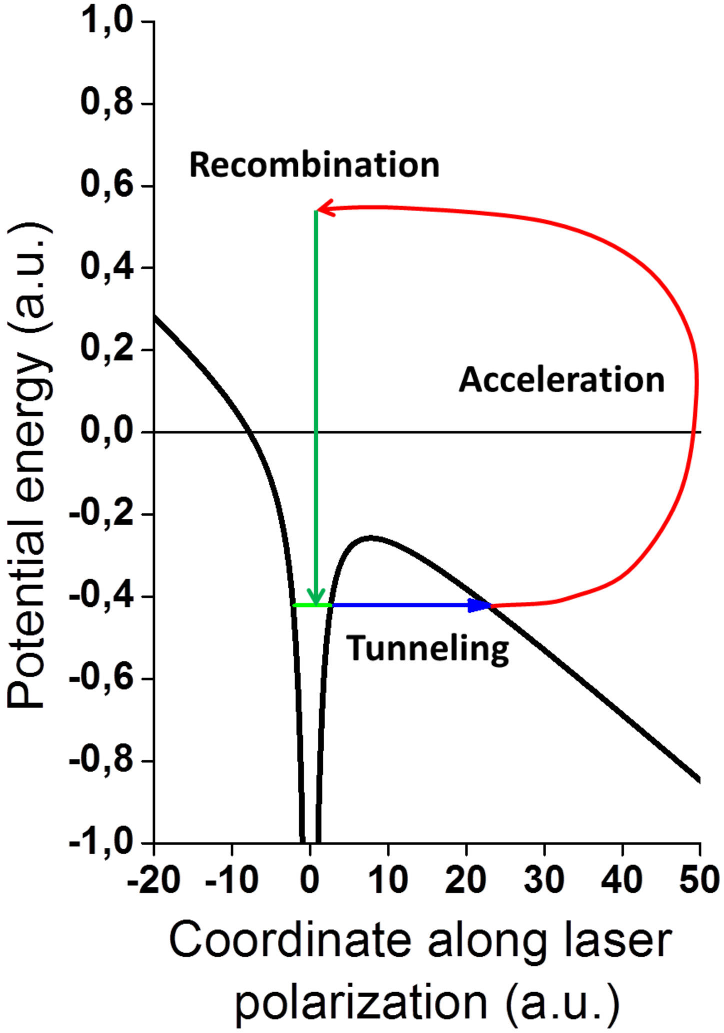


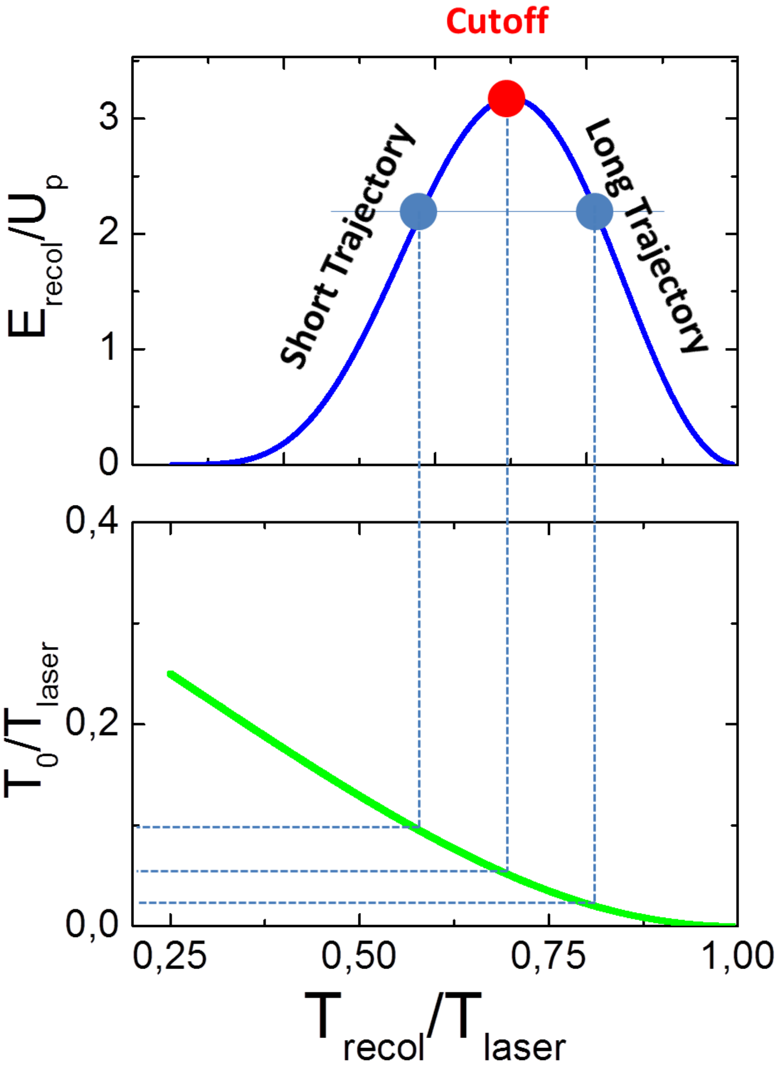
*ATI of Argon at 0.8 µm (black line), 1.3µm (green line), 2 µm (red line), and 3.6µm (blue line) at an intensity of 0.08 PW/cm2.*

**Colosimo, P. et al., Nat. Phys., 4, 386 (2008)**


Maximum recollision energy
3.17 $U_p$
Long trajectory
Short trajectory
Recollision energy (units of $U_p$)
3,5
3,0
2,5
2,0
1,5
1,0
0,5
0,0
0
10
20
30
40
50
60
70
80
90
Phase of laser electric field

# Cut-off law: $E_{cutoff} = IP + 3.17\ U_p$





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

**The main triumph of the three-step model was that it explained the cut-off law**

VOLUME 71, NU … PTEMBER 1993

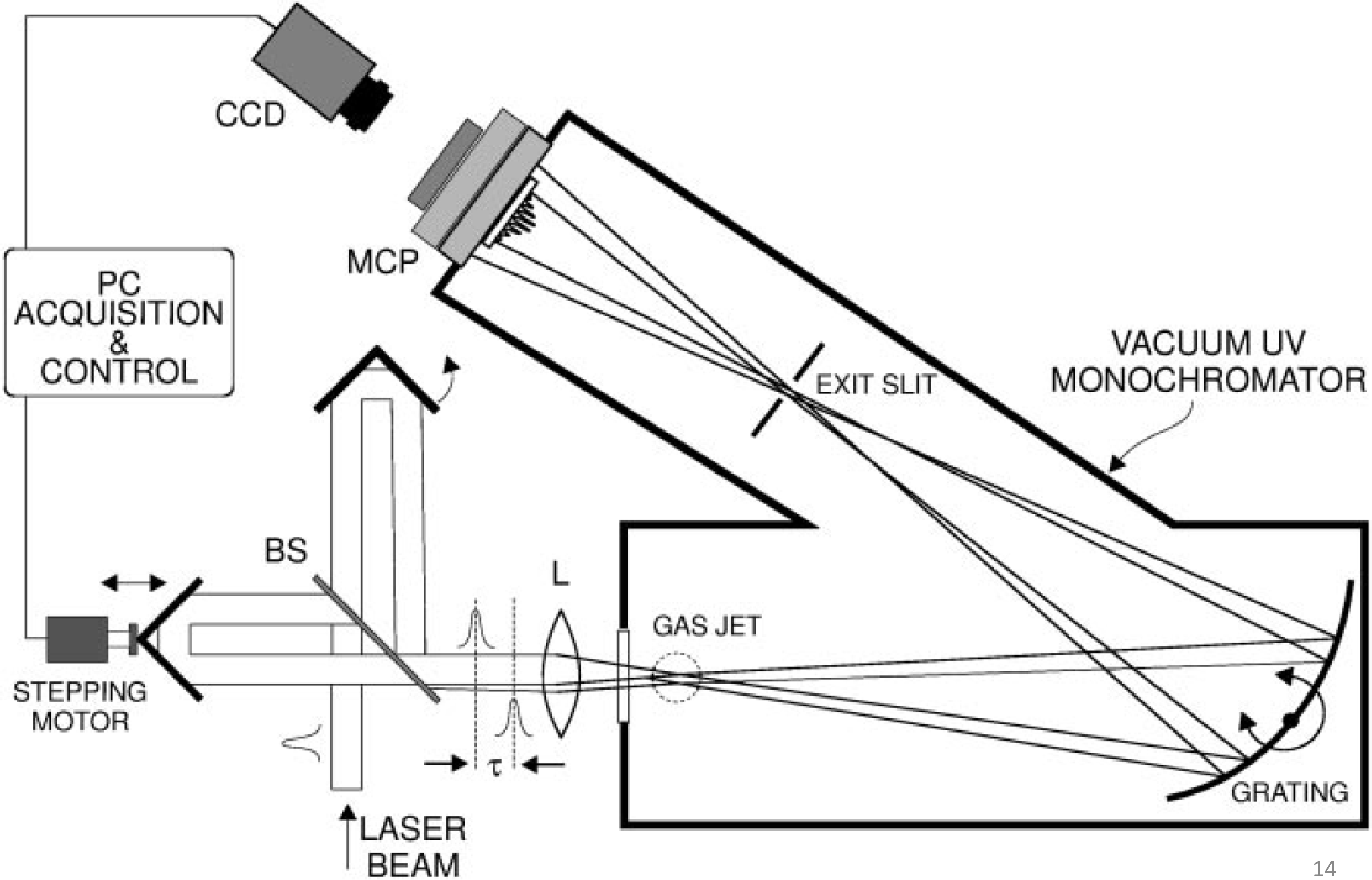
VOLUME 70, NUM … MARCH 1993

depending upon the phase of the electric field when the free electron is born. For electrons that are born with zero initial kinetic energy, the maximum drift velocity corresponds to measured energies of $3U_p$ in the long pulse limit. Additional drift velocity, resulting in energies substantially higher than $3U_p$, can be gained if the electron is born with some initial kinetic energy, or if it has a collision with the nucleus. This gives a likely source of the high energy electrons observed. OHG occurs only for those orbits which have at least one additional collision with the nucleus. This is borne out by numerical calculations which show that high-order harmonic production is completely accounted for by considering only transitions that end in the ground state [1]. Therefore, the maximum energy that the emitted photon can have must be the energy that the electron has *at the time it revisits the vicinity of the nucleus.* We find that for electrons that are born near the nucleus, regardless of their initial energy distribution, the maximum energy at the return time is 3.17 $U_p$ plus the field free ionization potential. This predicts the OHG cutoff remarkably well. The distinction between averaged and instantaneous energies is the key difference between ATI and OHG.

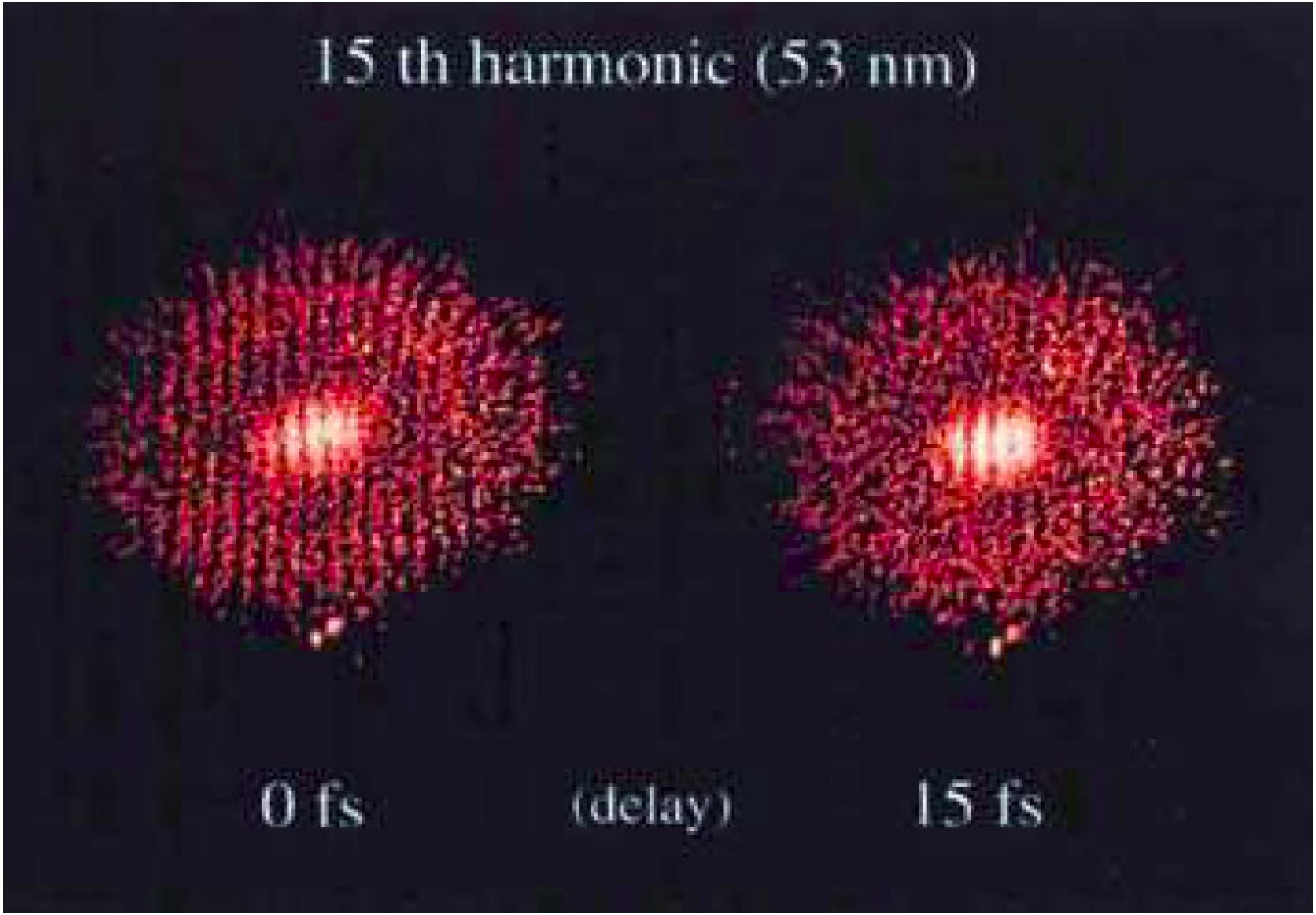
In summary, we have presented an experimental study

PACS numbers: 31.90.+s, 32.80.Fb, 32.80.Rm

# HHG via short and long trajectories



**M. Bellini et al., Phys. Rev. Lett. 81, 297 (1998)**



M. Bellini et al., Phys. Rev. Lett. 81, 297 (1998)

# Phase-matching

In the HHG medium, the driver laser and the generated harmonics move with a different phase velocity

$$v_{phase} = \square = \frac{\square}{2\pi} = \frac{\omega}{k}$$

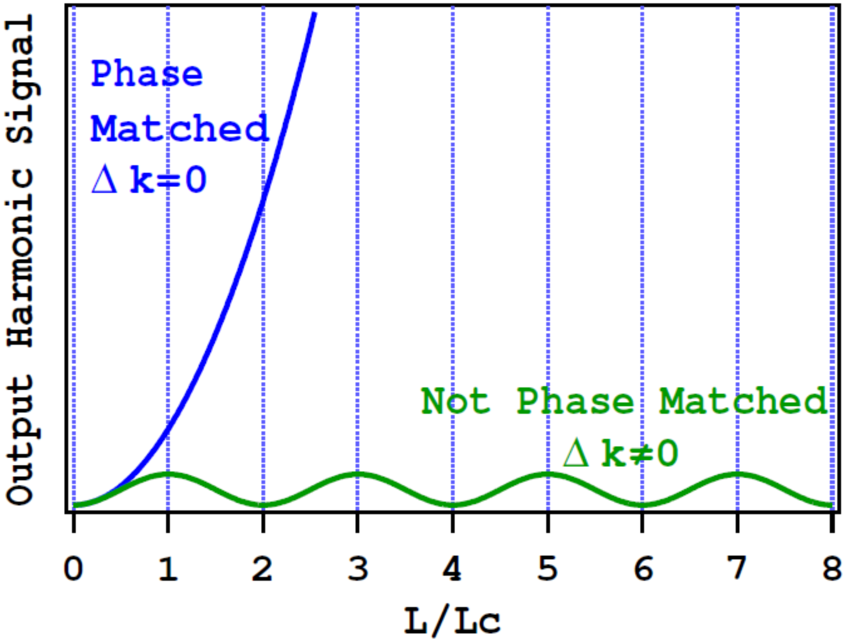
**laser frequency**
**laser wavevector**

**Lecture 1**

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

Phase matching: $\Delta k_q = k_q - qk_1 = 0$

Coherence length: $L_c = \frac{\pi}{\Delta k}$



**Thesis Amy Lytle, JILA 2008**

# Phase-matching

$$\Delta k = \overbrace{\underline{\Delta k_a + \Delta k_{fe}}}^{\sim\text{pressure}} + \Delta k_{foc} + \Delta k_{traj}$$

~intensity

In the HHG medium, phase matching is affected by

(i) The density of neutral gas $$\Delta k_a = \frac{q\omega}{2\varepsilon_0 c} N\left[\alpha_{pol}(q\omega) - \alpha_{pol}(\omega)\right]$$

(ii) The density of free electrons $$\Delta k_{fe} = \frac{qe^2}{2\varepsilon_0 c m \omega} N_e$$

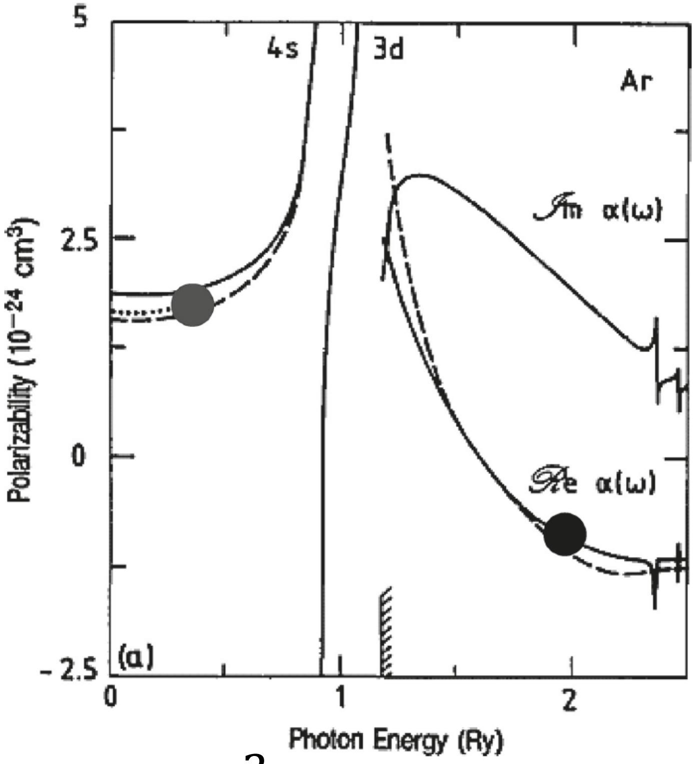
(iii) The laser focusing (no waveguiding assumed) $$\Delta k_{foc} \sim \frac{q}{z_0}$$

(iv) The electron trajectories $$\Delta k_{traj} = \alpha_{traj} \frac{\partial I}{\partial z}$$

# Serendipity in Phase-matching

$$\Delta k_a = \frac{q\omega}{2\varepsilon_0 c} N\left[\alpha_{pol}(q\omega) - \alpha_{pol}(\omega)\right]$$

**<0**



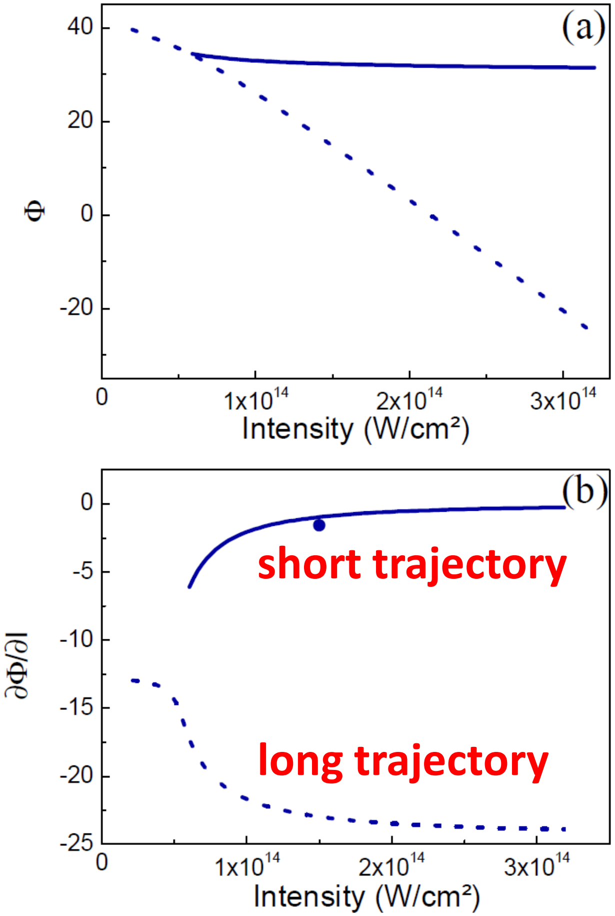
$$\Delta k_{fe} = \frac{qe^2}{2\varepsilon_0 c m\omega} N_e$$

**>0**

L'Huillier et al., JOSA B 7, 529 (1990)

$$\Delta k_{traj} = \alpha_{traj} \frac{\partial I}{\partial z}$$

$$\alpha_{traj} = -\frac{\partial \Phi}{\partial I} > 0$$

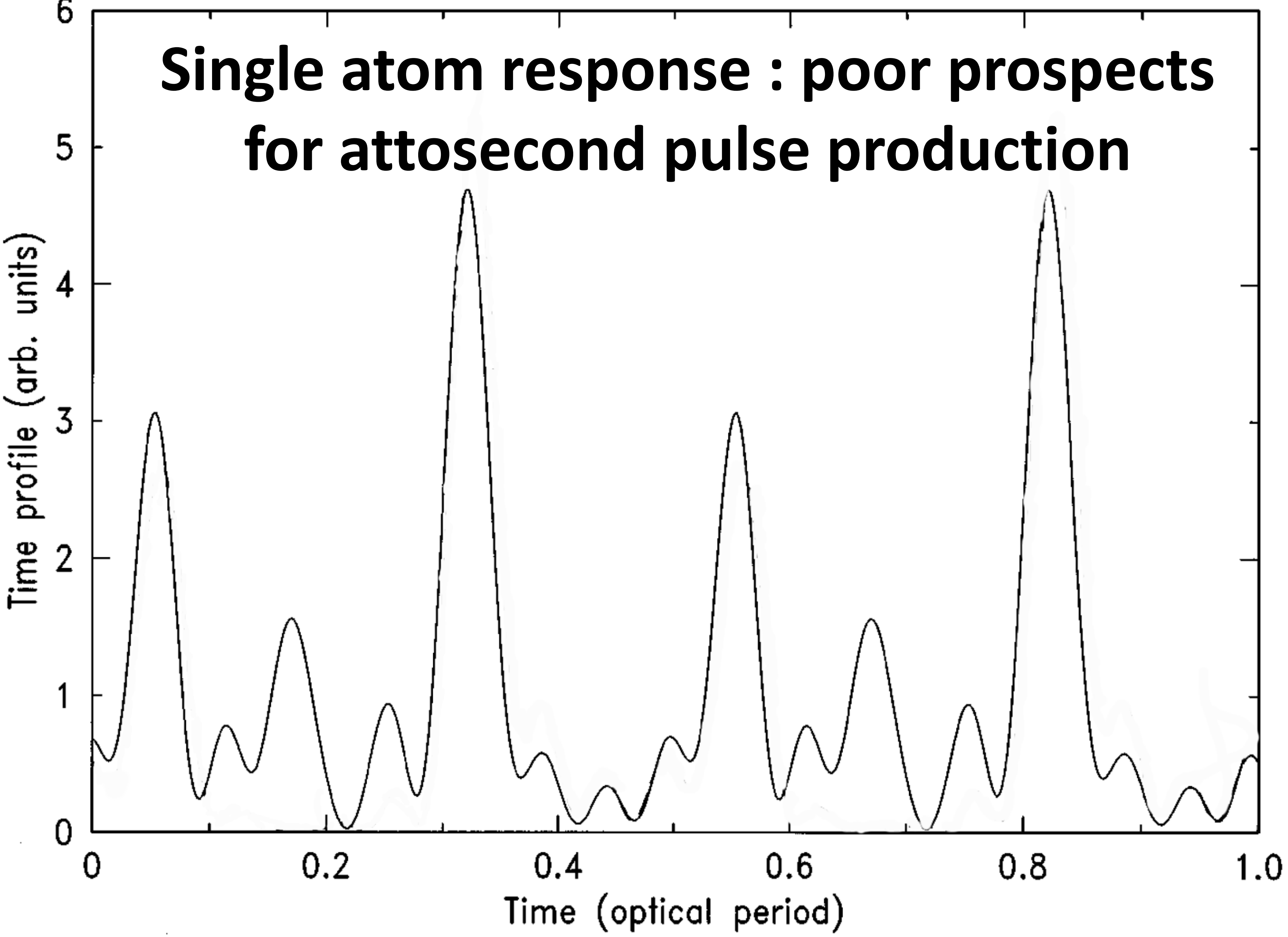


Short and long trajectory have different phase-matching conditions!

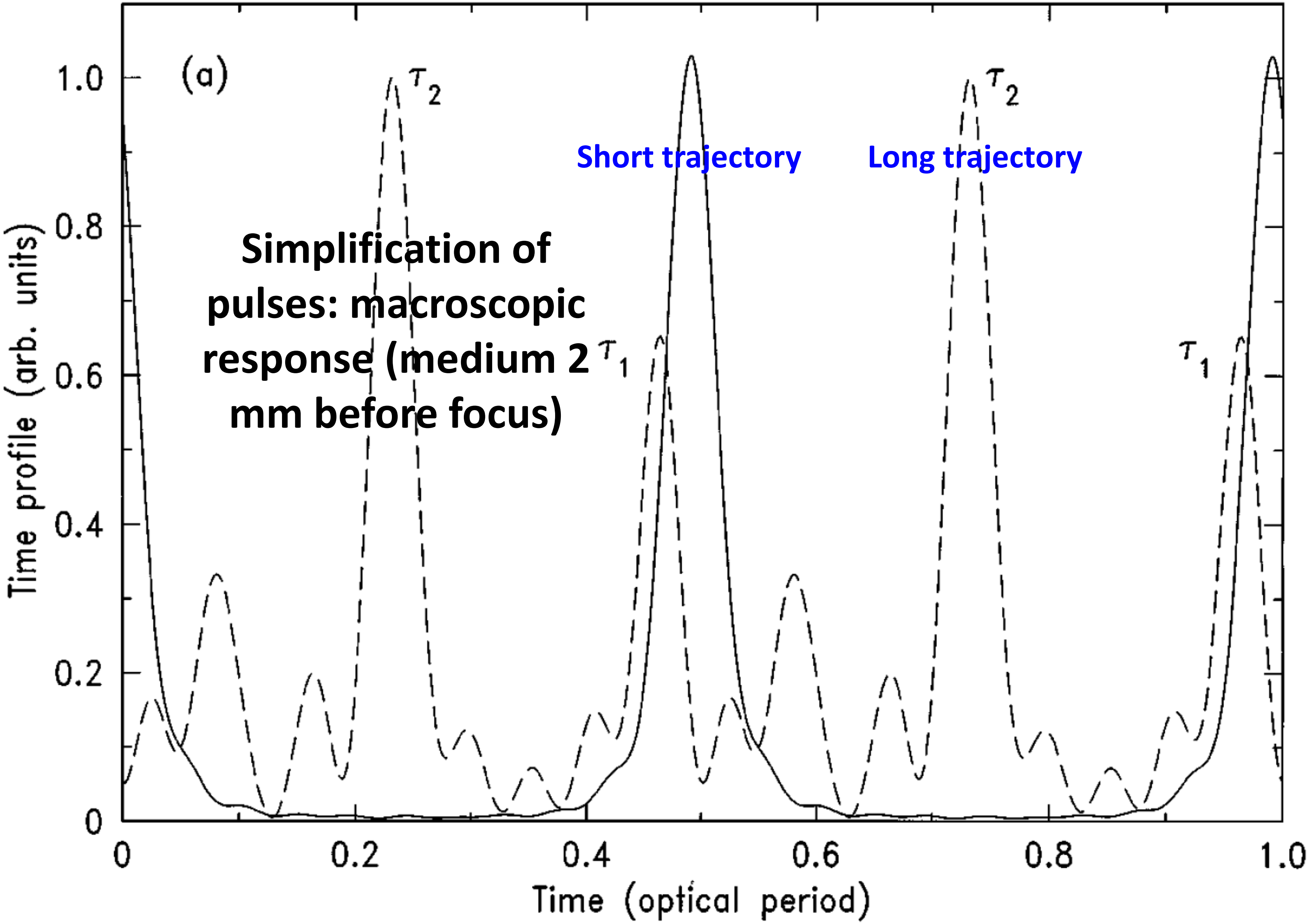
Varju et al., J. Mod. Opt. 2, 379 (2004)

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

# Single atom response : poor prospects for attosecond pulse production



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



Short trajectory

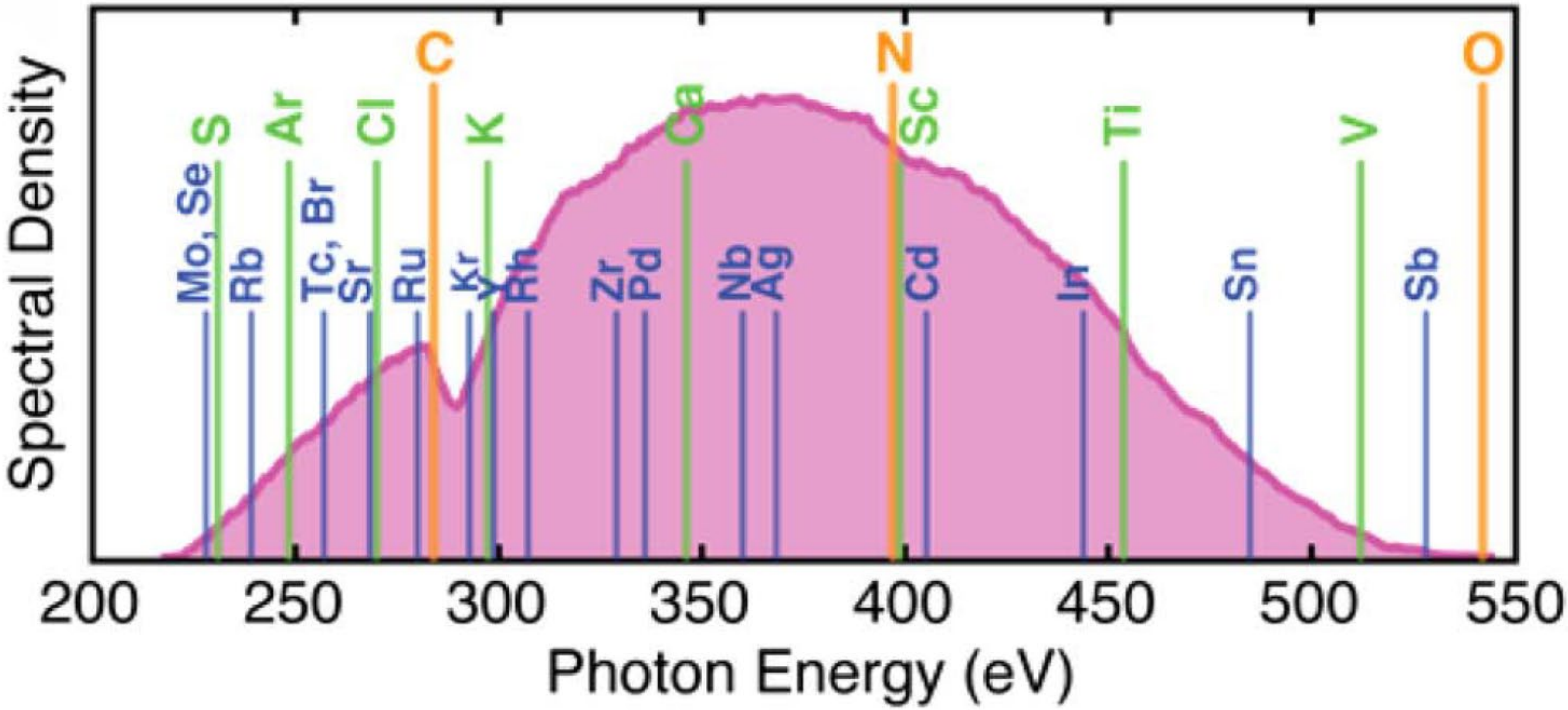
Long trajectory

**Simplification of pulses: macroscopic response (medium 2 mm before focus)**

# Generating high photon energies

- Introduction yesterday. HHG has two favorable aspects:
  - 1) it gives us a path towards the generation of attosecond pulses
  - 2) it generates radiation in the extreme ultra-violet (XUV) and soft X-ray range where element-specific core level transitions occur
- Main use: *core-level transient absorption*
  - Element-specific
  - Sensitive to electronic environment (oxidation state, coordinate number)
  - Sensitive to (molecular) structure around the element

# Absorption edges in the water window

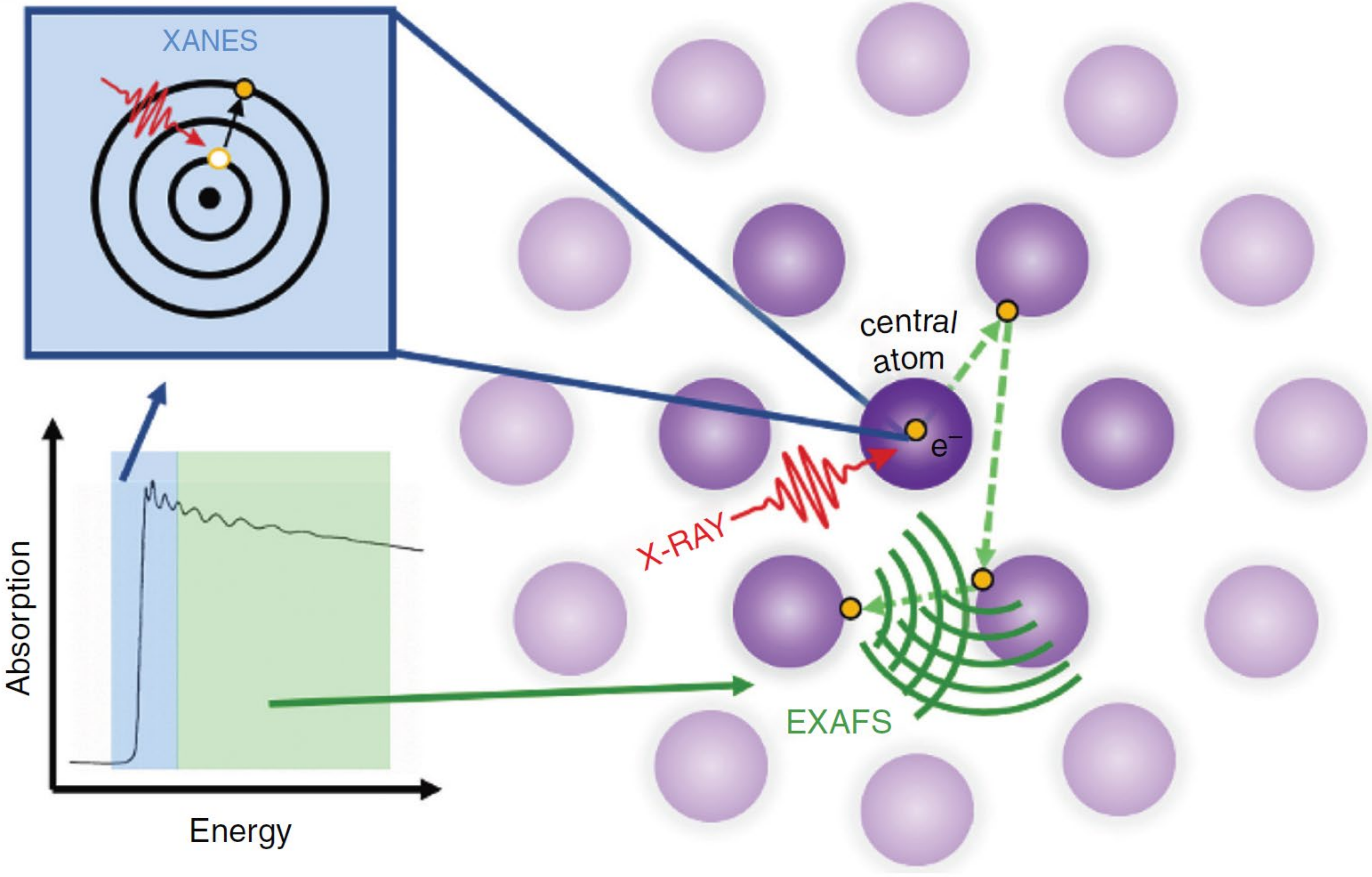


K-edges L-edges M-edges

When absorption edges from deeper-lying core levels are exploited, the theoretical description and – thus – interpretation of XAS experiments becomes easier → drive towards implementation of ATAS in the water window

B. Buades et al., Optica 5, 502 (2018)

# XANES and NEXAFS



XANES: information about electronic structure
EXAFS: structural information

B. Buades et al., Optica 5, 502 (2018)

# Extended Photon Energies

Today: Cut-off law: $E_{cutoff} = IP + 3.17\ U_p$

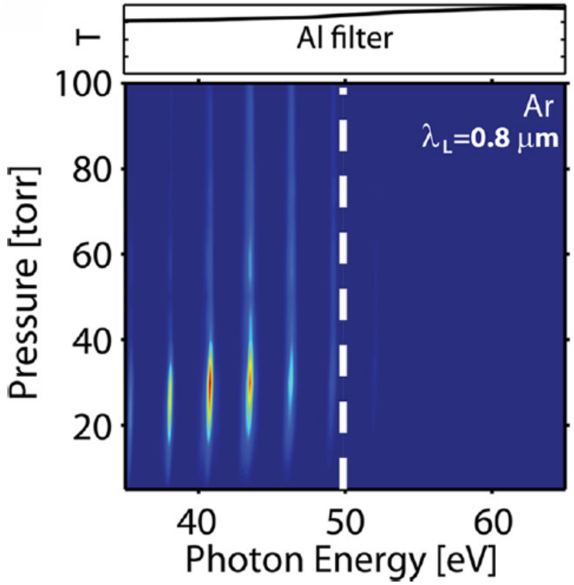
Yesterday: $U_p(eV) = 9.337\ I_{laser}\left(10^{14}\frac{W}{cm^2}\right)\ \lambda^2\ (\mu m)$

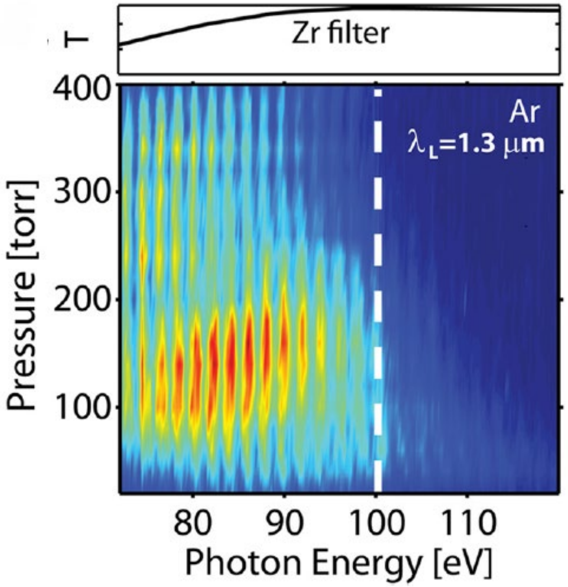
- Scaling of the cut-off energy by increasing the intensity? No (phase-matching)
- Scaling of the cut-off energy by increasing the wavelength? Yes

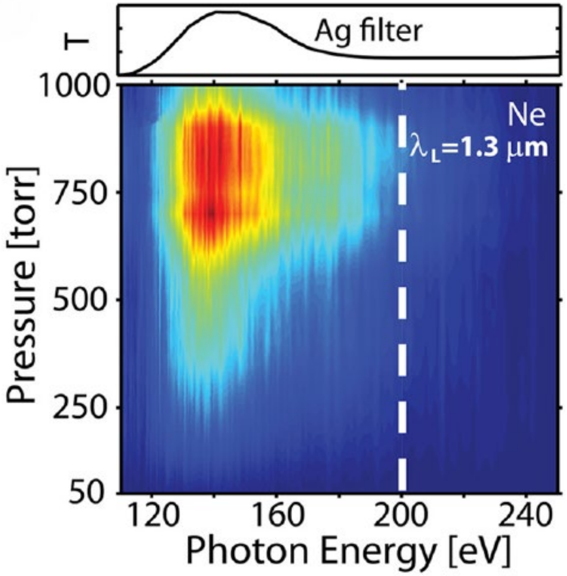
- Challenge: the single atom response scales very unfavorably with the driver laser wavelength ($\lambda^{-5}$)
- However, the re-absorption of the high-harmonics is much less severe at high photon energies + the phase-matching is optimized for higher pressures

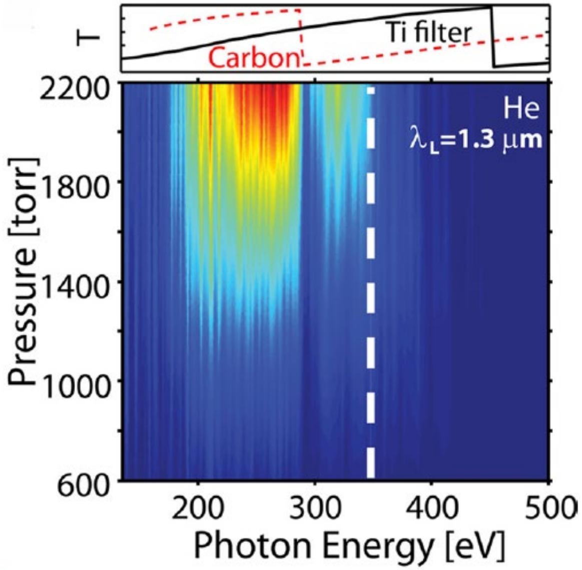
**Popmintchev et al., PNAS 106, 10516 (2009)**

# Extended Photon Energies





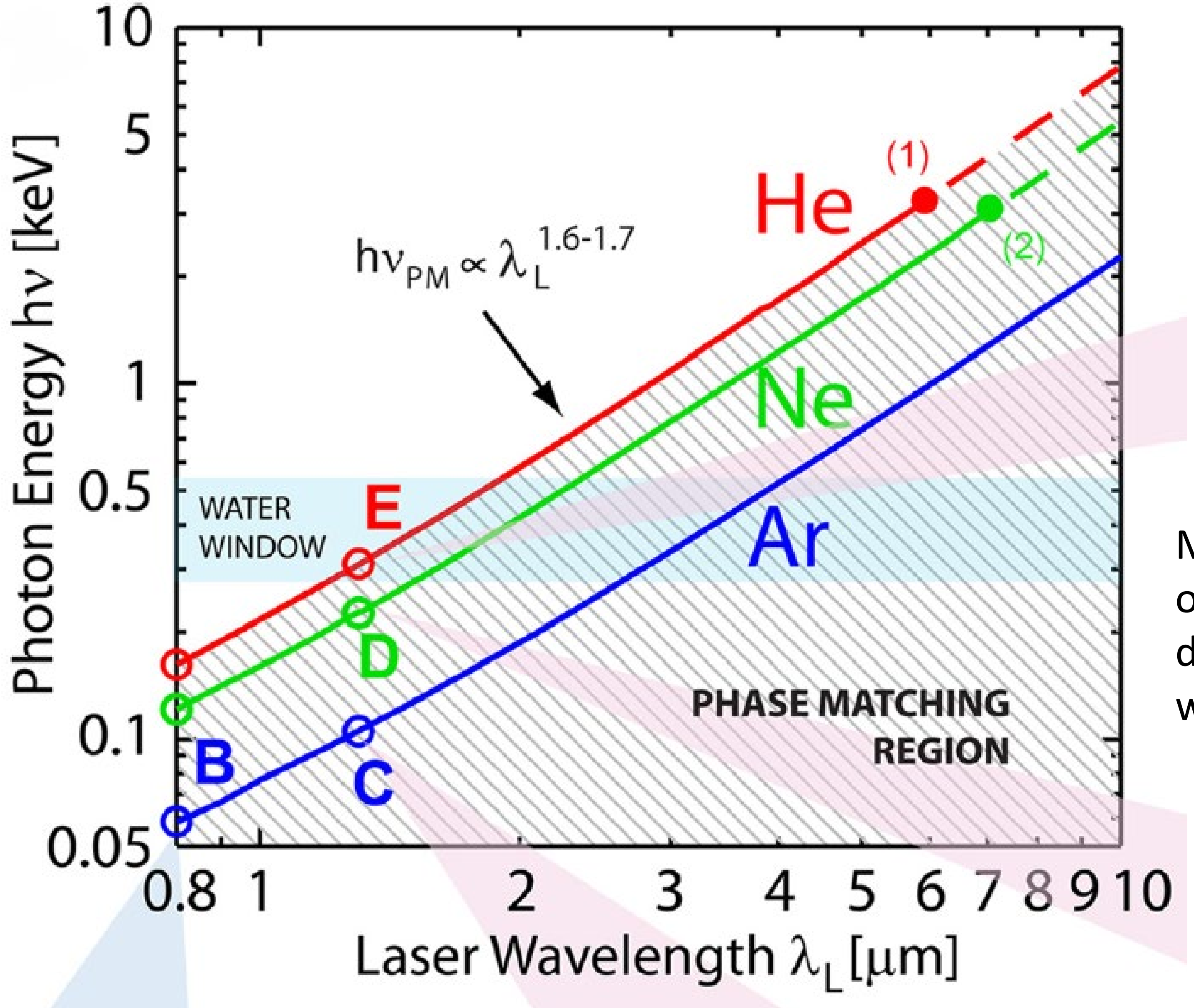




Significant extension of the cut-off by going to longer wavelength and by going to atom (He) with a higher ionization potential and lower re-absorption cross-section

**Popmintchev et al., PNAS 106, 10516 (2009)**

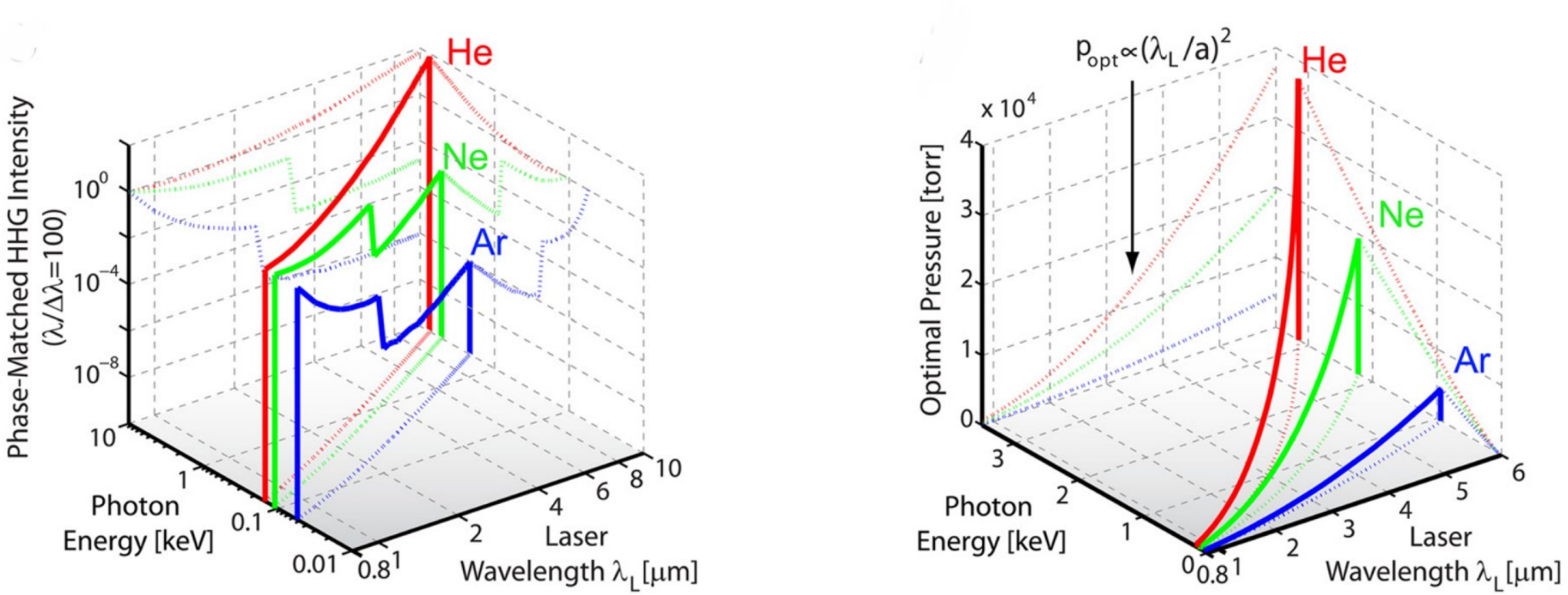
# Extended Photon Energies



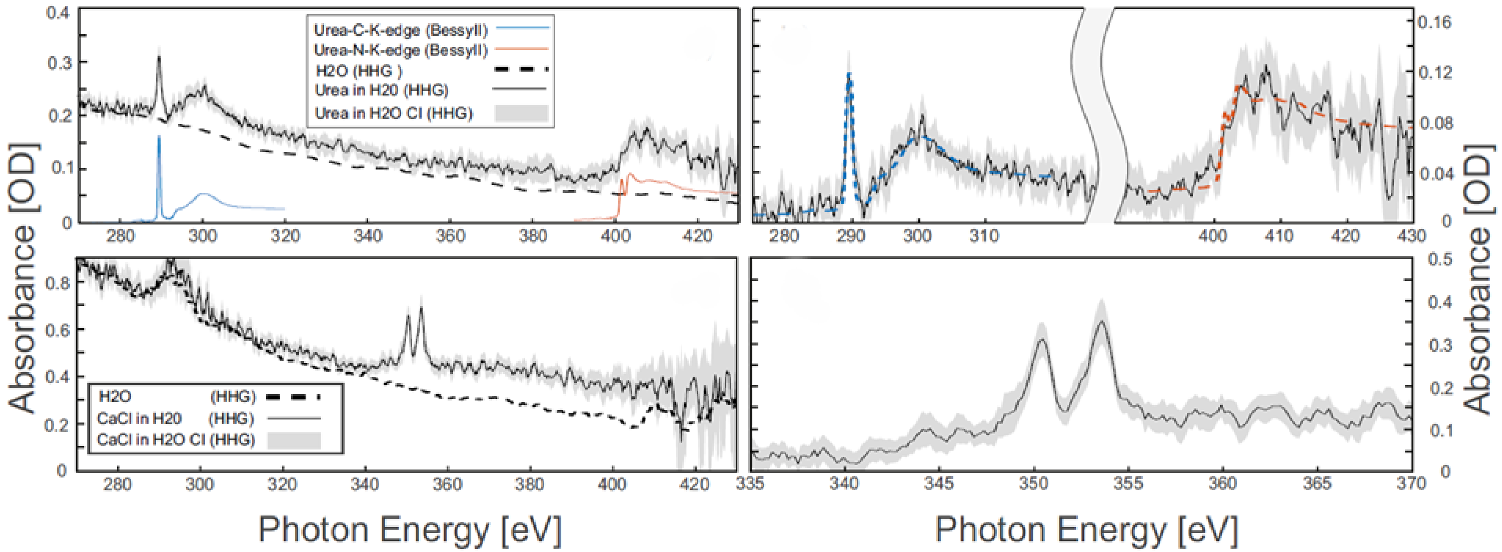
Motivation for ongoing laser development at wavelengths ≥5 μm

Popmintchev et al., PNAS 106, 10516 (2009)

# Extended Photon Energies



**Popmintchev et al., PNAS 106, 10516 (2009)**



HHG-based
XAS at MBI

He, 4 bar

# Useful materials for further reading (strong field ionization):

R. Weissenbilder et al., Nature Reviews Physics 4, 713 (2022)

T. Popmintchev et al, PNAS 106, 10516 (2009)

+ several chapters (DiMauro, Ivanov, Smirnova, L´Huillier) in upcoming book „Attosecond and XUV Physics“ (ed. by M.J.J. Vrakking and Th. Schultz, Wiley, december 2013)