# Advanced Radiation Sources - PHSY761

Lecture 07

12 November 2024

## Summary

Breakdown of perturbative nonlinear optics
- Harmonic generation in gases
- Multiphoton ionization
- Strong field effects
  - Above threshold ionization
  - High Harmonic Generation
  - Non-sequential double ionization

3 step model of High Harmonic Generation
1. Tunneling
2. Acceleration
3. Recombination
4. Cutoff and plateau harmonics

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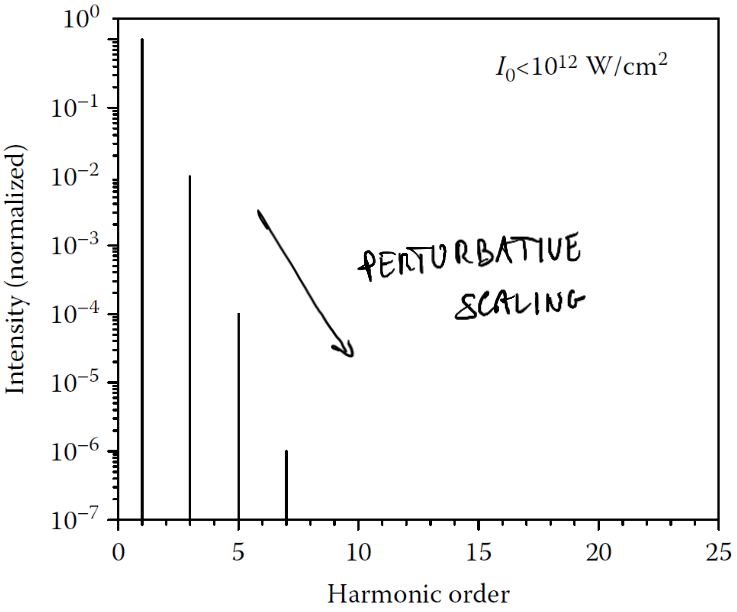
## Perturbative nonlinear optics:

$$\mathcal{P} = \varepsilon_0 \left[ \chi^{(1)} \mathcal{E} + \chi^{(2)} \mathcal{E}^2 + \chi^{(3)} \mathcal{E}^3 + \cdots \right]$$

Can we use these perturbative phenomena to generate short-wavelenght pulses?

- Coherent sources where no lasing transitions are available
- Spectroscopy studies of matter with short pulses
- Table-top sources vs large facilities

# Expected scaling law in the perturbative regime:




$$\mathscr{P} = \varepsilon_0 \left[ \chi^{(1)} \mathcal{E} + \chi^{(2)} \mathcal{E}^2 + \chi^{(3)} \mathcal{E}^3 + \cdots \right]$$

Intensity scaling:

$$I_{2\omega}(t) \propto I_\omega^2(t)$$

$$\downarrow$$

$$I_{q\omega} \propto I^q(t)$$

Pulse duration scaling:

$$\tau_{2\omega} = \frac{\tau_\omega}{\sqrt{2}}$$

$$\downarrow$$

$$\tau_{q\omega} = \frac{\tau_\omega}{\sqrt{q}}$$

- Possible to generate short pulses!

To observe a high-order process the intensity has to be increased!

$$I_{at} = \frac{c}{8\pi} E_{at}^2 = 4 \times 10^{16}\ \mathrm{W/cm^2}$$

$$\frac{P^n}{P^{n-1}} \sim \frac{\chi^{(n)}}{\chi^{(n-1)}} \mathcal{E} \sim \frac{\mathcal{E}}{E_{at}} \ll 1 \qquad E_{at} = \frac{e}{a_0^2} = \frac{e}{(\hbar^2/me^2)^2}$$

FOR $10^{13}$ W/cm$^2$
$\mathcal{E}/E_{at} \sim 10^{-2}$

- Damage threshold of bulk optical materials ≈ 100 GW/cm²: only **gases can withstand higher intensities.**

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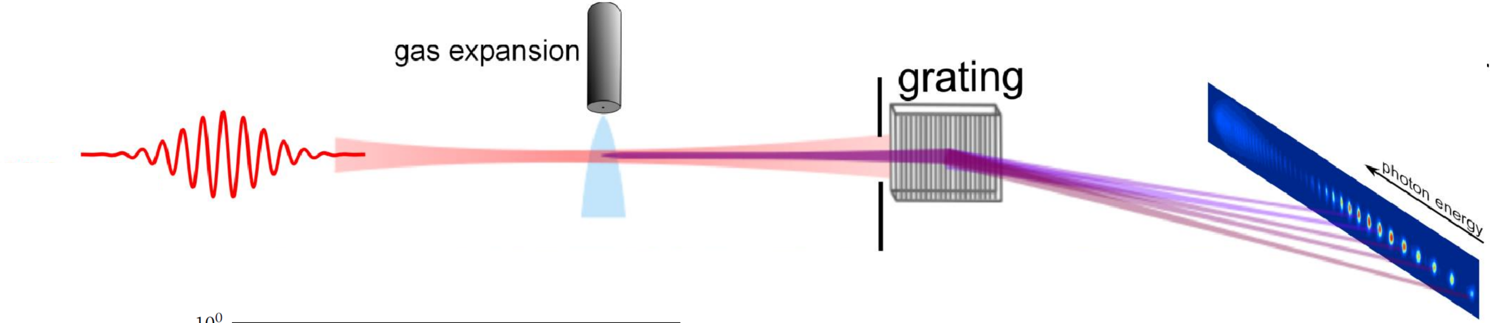
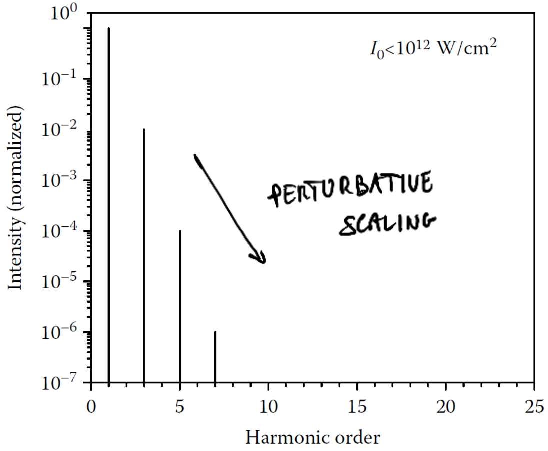
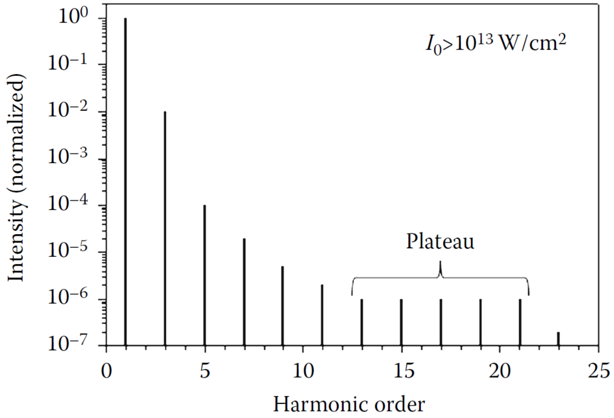
# Nonperturbative effects: Harmonic generation in gases





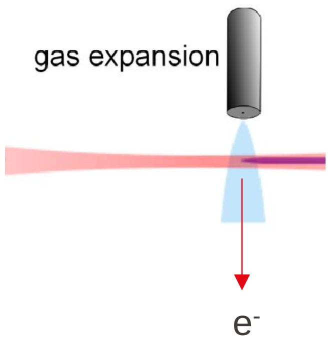



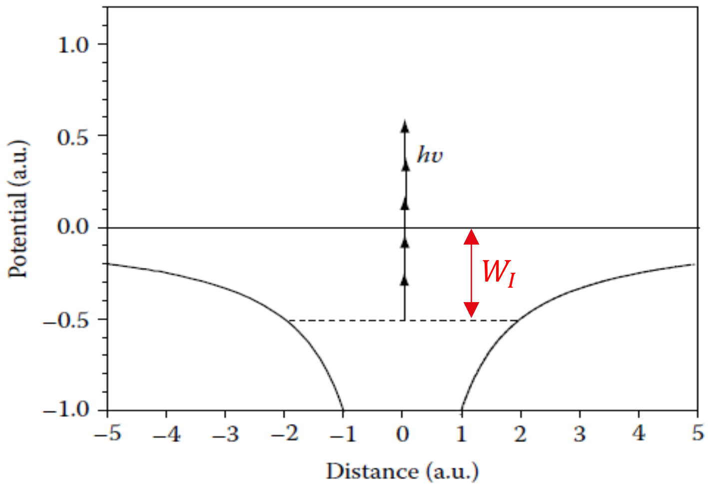




- At intensities approaching ≈$10^{13}$ W/cm$^2$ the perturbative description fails.

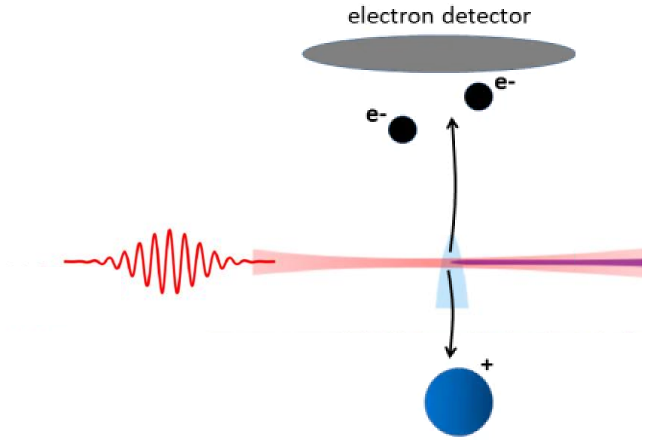
# Electron detection : perturbative multiphoton ionization (MPI)

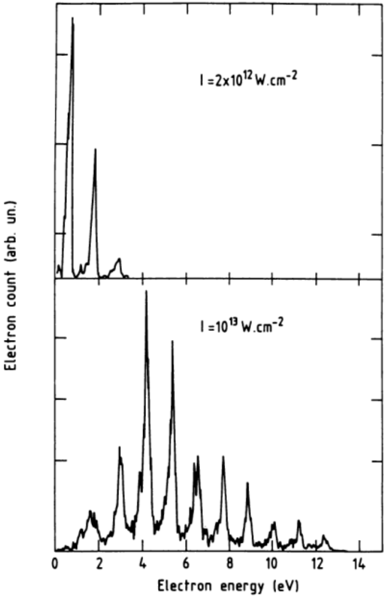
- Atoms can be ionized by a multiphoton process of high order!
- Photoelectrons and positive ions are created for $\hbar\omega \sim 1\ eV \ll W_I \sim 10\ eV$

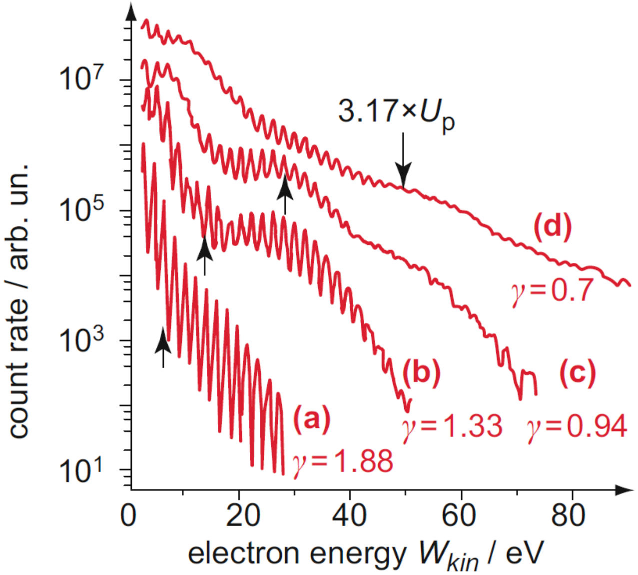


# Nonperturbative effects:  Above-threshold ionization (ATI)




- Photoelectrons and photoions emission

$$\text{Keldysh } \textit{parameter} \quad \gamma = \sqrt{\frac{W_I}{2U_p}}$$

- Keldysh parameter
  - γ >1 perturbative MPI
  - γ <1 non-perturbative ATI

# $U_p$ ponderomotive energy:



Classical electron in a periodic E field:

$$m_e \frac{dv}{dt} = eE_0 \cos \omega t$$

$$\begin{cases} x(0) = 0 \\ v(0) = 0 \end{cases}$$

$$v(t) = \frac{eE_0}{m_e \omega} \sin \omega t \quad \Rightarrow \quad \frac{1}{2} m_e v^2 = \frac{e^2 E_0^2}{2 m_e \omega^2} \sin^2 \omega t$$

CYCLE AVERAGE

$$\frac{1}{T} \int_0^T f(t)\,dt$$

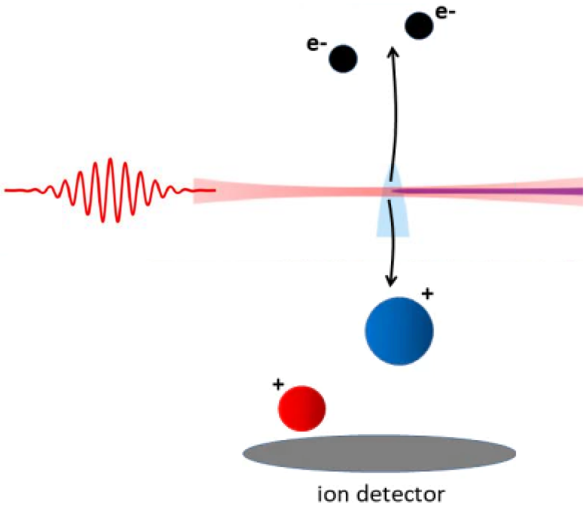
$$U_p = \overline{\frac{1}{2} m_e v^2} = \frac{e^2 E_0^2}{4 m_e \omega^2}$$

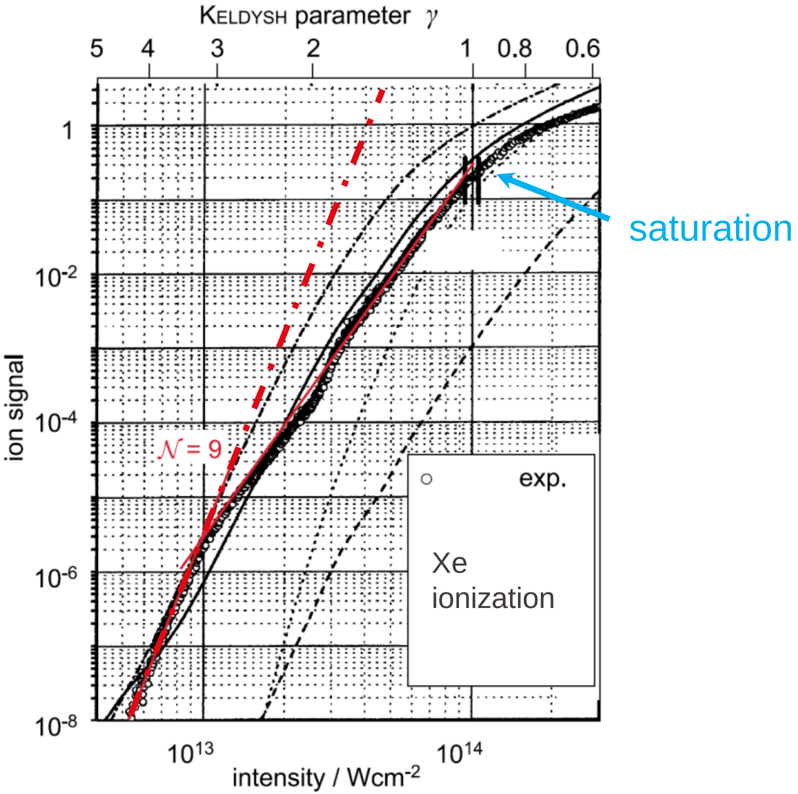
KELDYSH *parameter* $\gamma = \sqrt{\frac{W_I}{2U_p}}$

# Ionization saturation



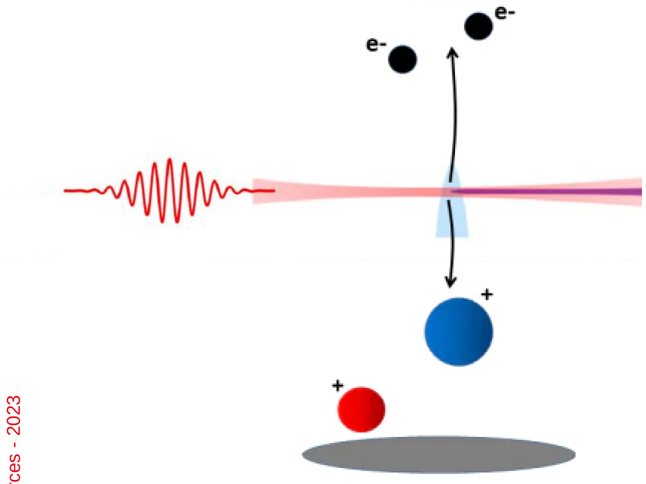


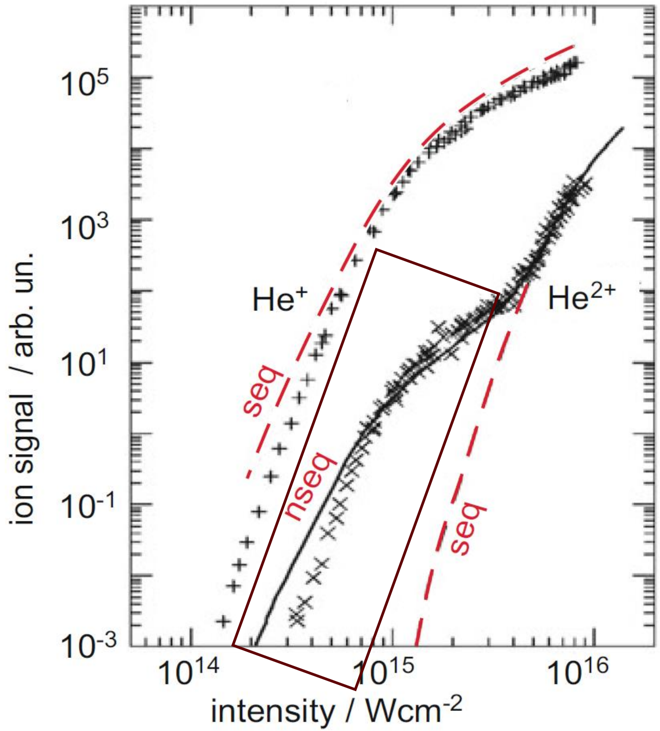



- deviation from perturbative scaling
- saturation due to ground state depletion

# Evidence of electron “recollisions”




- Non-sequential double ionization (He)
- Evidence for “electron” re-scattering with to the parent ions



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# Field ionization:

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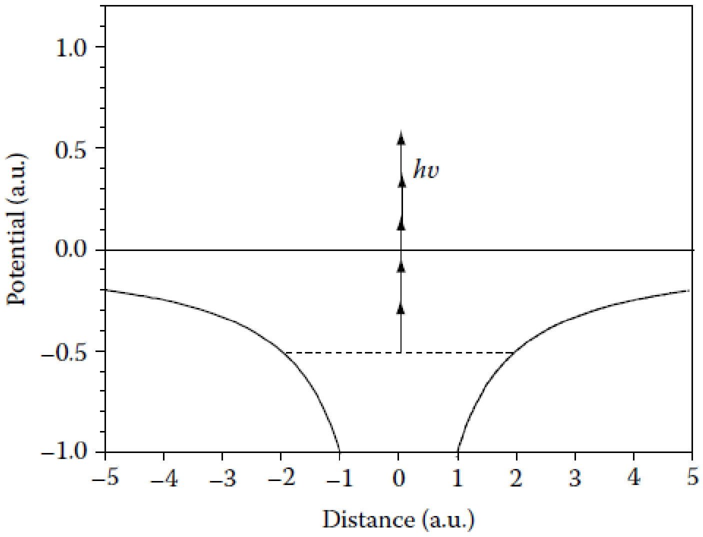
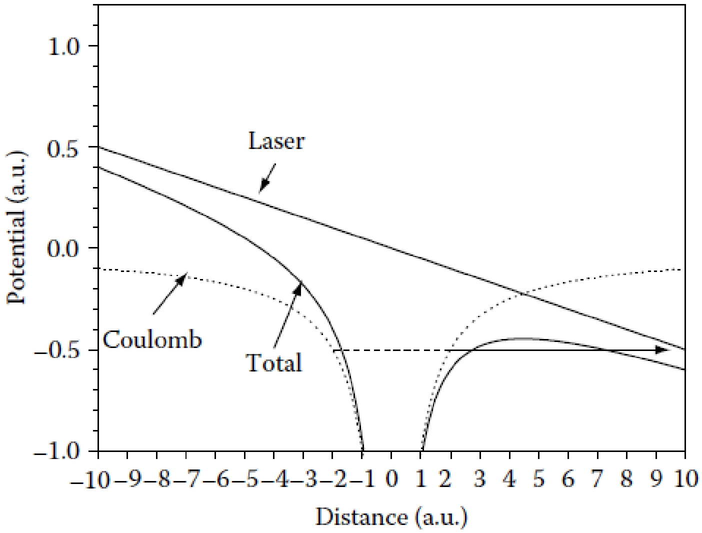
- At high intensities the electric field is comparable to the atomic Coulomb field



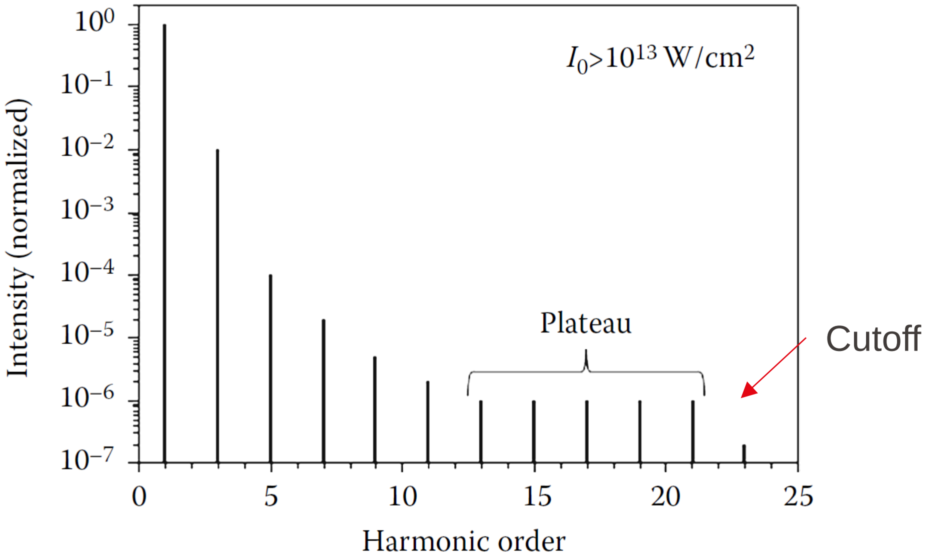

Atomic Coulomb field




Laser field + atomic Coulomb field

- Other ionization channels appears: the electron has a non-zero probability of tunneling outside the barrier : “field ionization”
- Many *strong-field* phenomena can be qualitatively understood in term of **semi-classical models**

# HHG spectrum

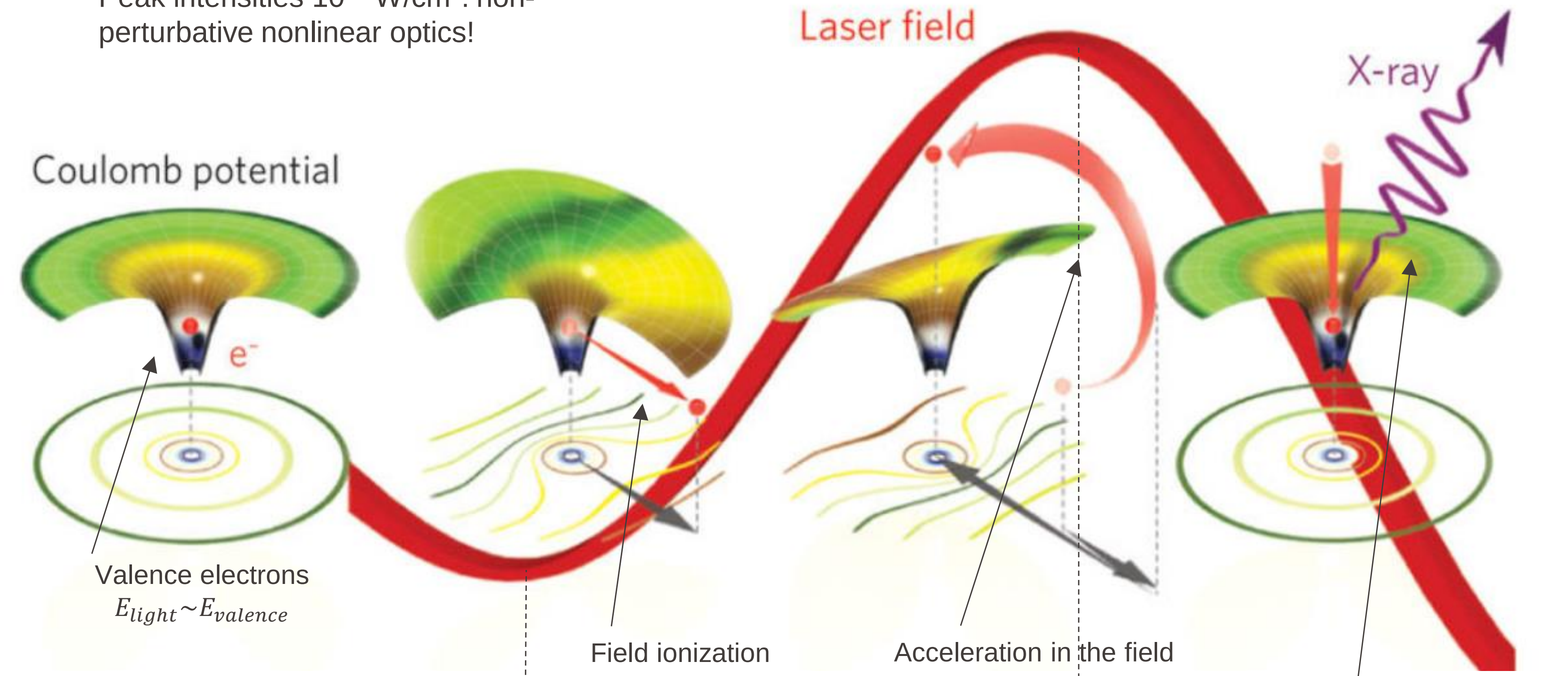



- How can we understand the harmonic generation process in terms of tunneling?
- Why there is “plateau” in the efficiency?
- $2\omega$ spacing?
- What determines the cut-off?
- What is the time structure of the harmonics?

# High-harmonic generation: three-step model



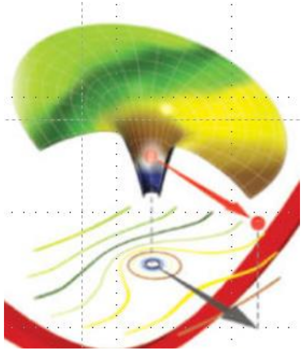
Peak intensities $10^{14}$ W/cm$^2$: non-perturbative nonlinear optics!

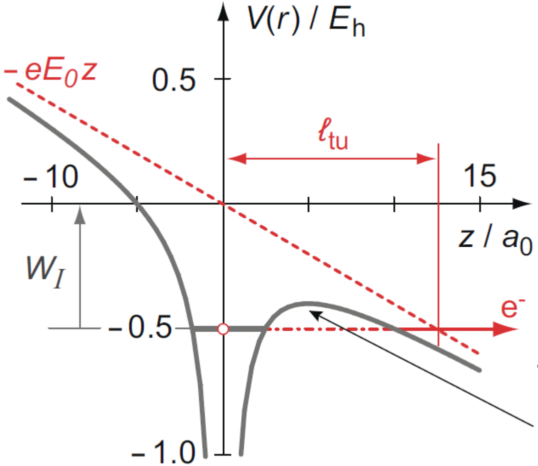



Recombination: emission of a high energy photon

Half-light cycle -> 2.7 fs/ 2 @ 800 nm

# STEP 1 : Ionization in the strong field






Tunneling "time":

- Assume that electrons have a kinetic energy given by the binding energy ($W_I$ )
- Assume it has to travel a (field-dependent ) tunneling lenght

$$\ell_{\mathrm{tu}} = W_I/(eE_0)$$

$$t_{\mathrm{tu}} = \frac{\ell_{\mathrm{tu}}}{v} = \frac{\sqrt{m_e W_I}}{\sqrt{2} e E_0}$$

$$\sqrt{2W_I/m_e}$$

# Keldysh parameter

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$$U_p = \frac{e^2 E_0^2}{4 m_e \omega^2}$$

TUNNELING TIME

$$2\pi \frac{t_{Tu}}{t_{1/2}} = t_{Tu}\, 2\omega$$

HALF-CYCLE DURATION

$$= \sqrt{\frac{m_e W_I}{2 e^2 E_0^2}}\, 2\omega = \sqrt{\frac{W_I\, 4\omega^2 m_e}{2\, e^2 E_0^2}}$$

$$= \sqrt{\frac{W_I}{2 U_p}} = \gamma$$

In the limit of small γ, we can treat tunneling through the barrier as a quasi-static process

# Crude approximation: rectangular energy barrier, stationary solution

$$\ell_{tu} = \frac{W_I}{eE(t)}$$

$$\frac{\Gamma_{ion}(t)}{\Gamma^0_{ion}} = \exp\left[-\frac{1}{\hbar}\sqrt{2 m_e W_I}\,\ell_{tu}(t)\right] \longrightarrow \exp\left(-\frac{\frac{1}{\hbar e}\sqrt{2m_e}\,W_I^{3/2}}{E(t)}\right)$$

Electric field appear in an exponential function!

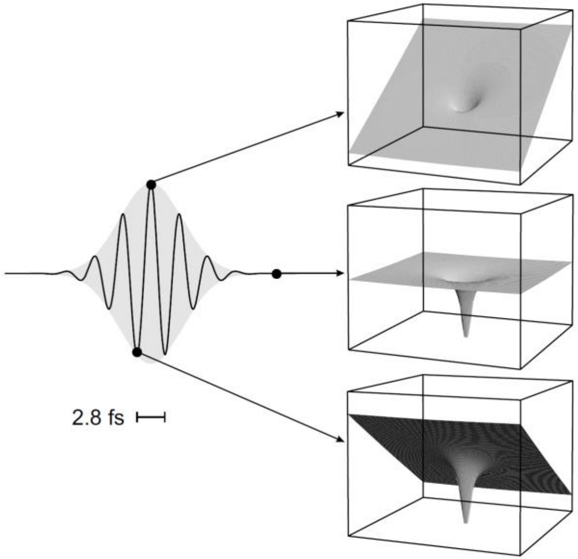
## More accurate: ADK ionization rate (Ammosov Delone Krainov 1986)

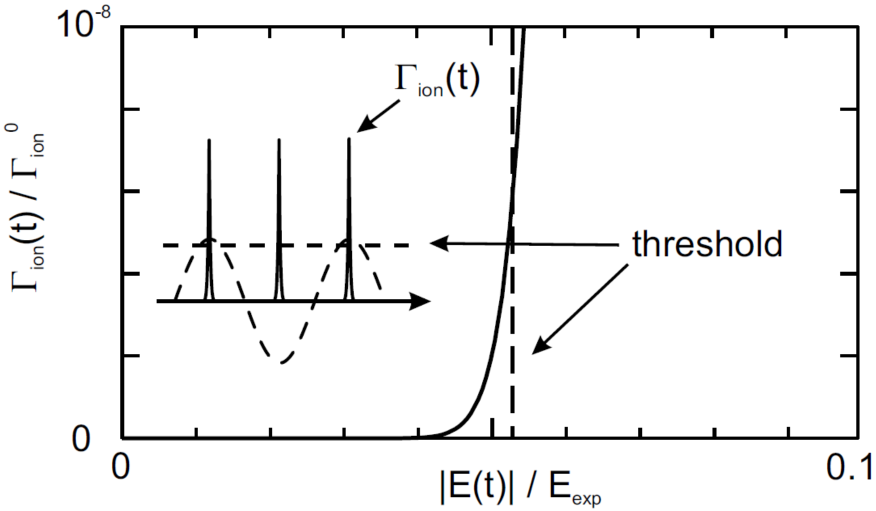
**TABLE 4.2** The ADK Parameters

| | $F_0$ (a.u.) | $n^*$ | $l^*$ | $l$ | $m$ | $\lvert C_{n^*l^*}\rvert^2$ | $G_{lm}$ |
|---|---|---|---|---|---|---|---|
| He | 2.42946 | 0.74387 | −0.25613 | 0 | 0 | 4.25575 | 1 |
| Ne | 1.99547 | 0.7943 | −0.2057 | 1 | 0 | 4.24355 | 3 |
| Ar | 1.24665 | 0.92915 | −0.07085 | 1 | 0 | 4.11564 | 3 |
| Kr | 1.04375 | 0.98583 | −0.01417 | 1 | 0 | 4.02548 | 3 |
| Xe | 0.84187 | 1.05906 | 0.05906 | 1 | 0 | 3.88241 | 3 |

$$w_{ADK} = \left|C_{n^*l^*}\right|^2 G_{lm} I_p \left(\frac{2F_0}{F}\right)^{2n^*-|m|-1} e^{-\frac{2F_0}{3F}},$$

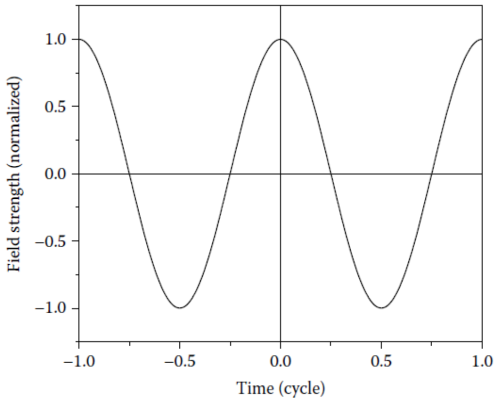
$$F[\text{a.u.}] = \sqrt{\frac{I\left[\text{w/cm}^2\right]}{3.55\times 10^{16}}}.$$



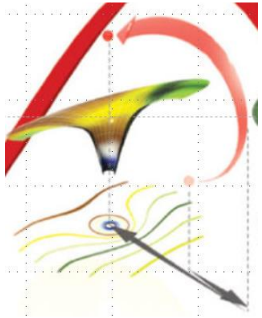




- Threshold-behaviour due to the tunneling probability
- Tunneling act as a fast (< half-cycle) shutter.
- Tunneling possible only for high field amplitude near the maximum

# STEP 2 – acceleration in the field




- A classical description give good physical insights: the coulomb potential is completely neglected compared to the laser field
- A more rigorous QM theory (Strong-field approximation) center of mass motion of the electron wavepackets follow classical trajectories



$$\frac{d^2x}{dt^2} = -\frac{e}{m_e}\varepsilon_L(t) = -\frac{e}{m_e}E_L\cos(\omega_0 t),$$

$v(t')=0 \quad x(t')=0$

$t'$ : IONIZATION TIME

$$x(t) = \frac{eE_L}{m_e\omega_0^2}\{[\cos(\omega_0 t) - \cos(\omega_0 t')] + \omega_0\sin(\omega_0 t')(t-t')\},$$

$= X_0$

nm DISPLACEMENT

$$\frac{x(t)}{X_0} = 0$$

$\downarrow$

RETURN TIME?



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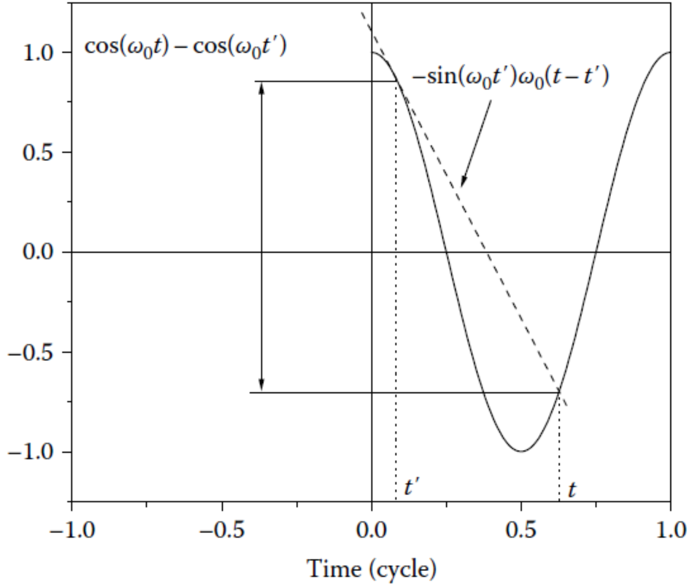
- The recollision time as a function of t' can be determined by the equation x(t)=0

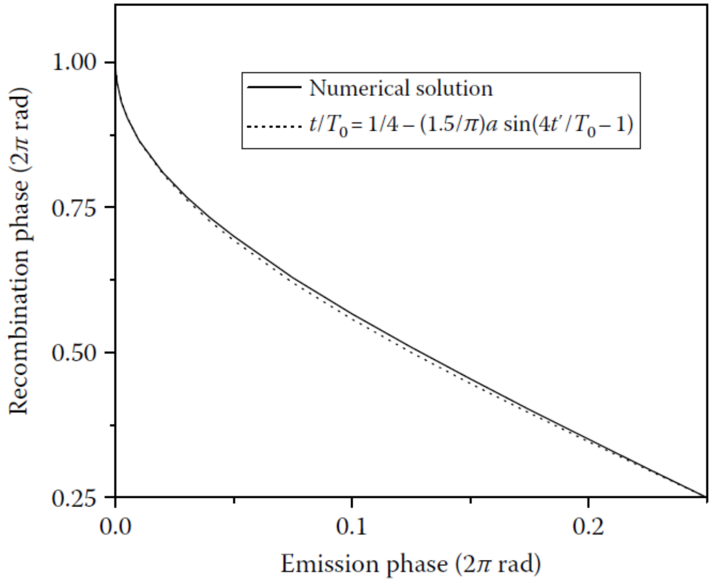
Equivalent form of x(t)=0

$$\cos(\omega_0 t) - \cos(\omega_0 t') = \frac{d}{d(\omega_0 t)} \cos(\omega_0 t)\Big|_{t'} (\omega_0 t - \omega_0 t').$$

Graphical solution:

(units of $x_0$)

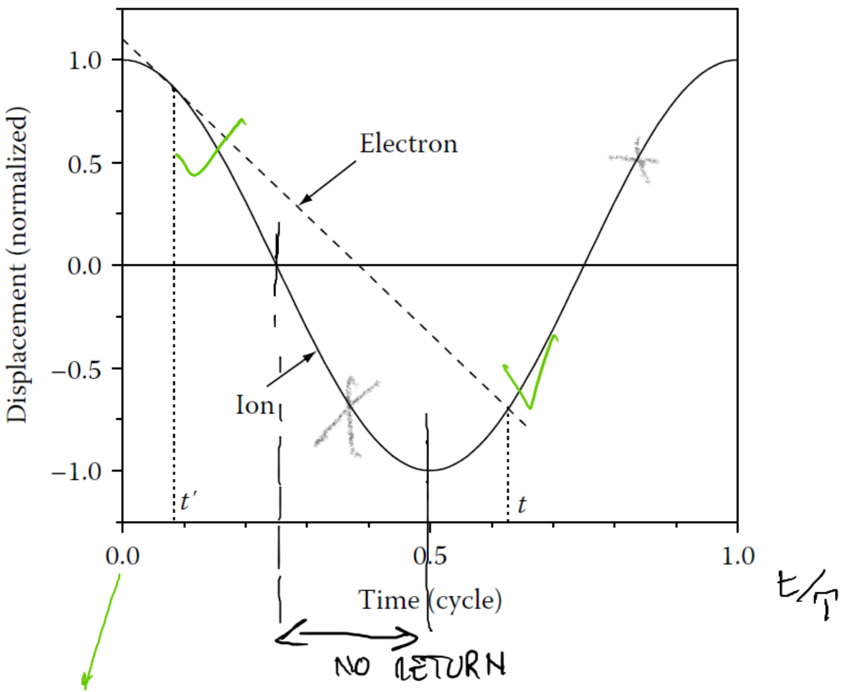


# Kramer-Hennerberger frame of reference



- Physical interpretation of the graphical solution:
  - ion moving in an oscillatory frame ($x'=x-x_0\sin(\omega_0 t)$ )
  - Electron performs a linear trajectory




$\omega t' = 0 \rightarrow$ RETURN AFTER 1 CYCLE $\omega t = 2\pi$

$\pi/2 < \omega t < \pi \rightarrow$ THE ELECTRON DON'T RETURN

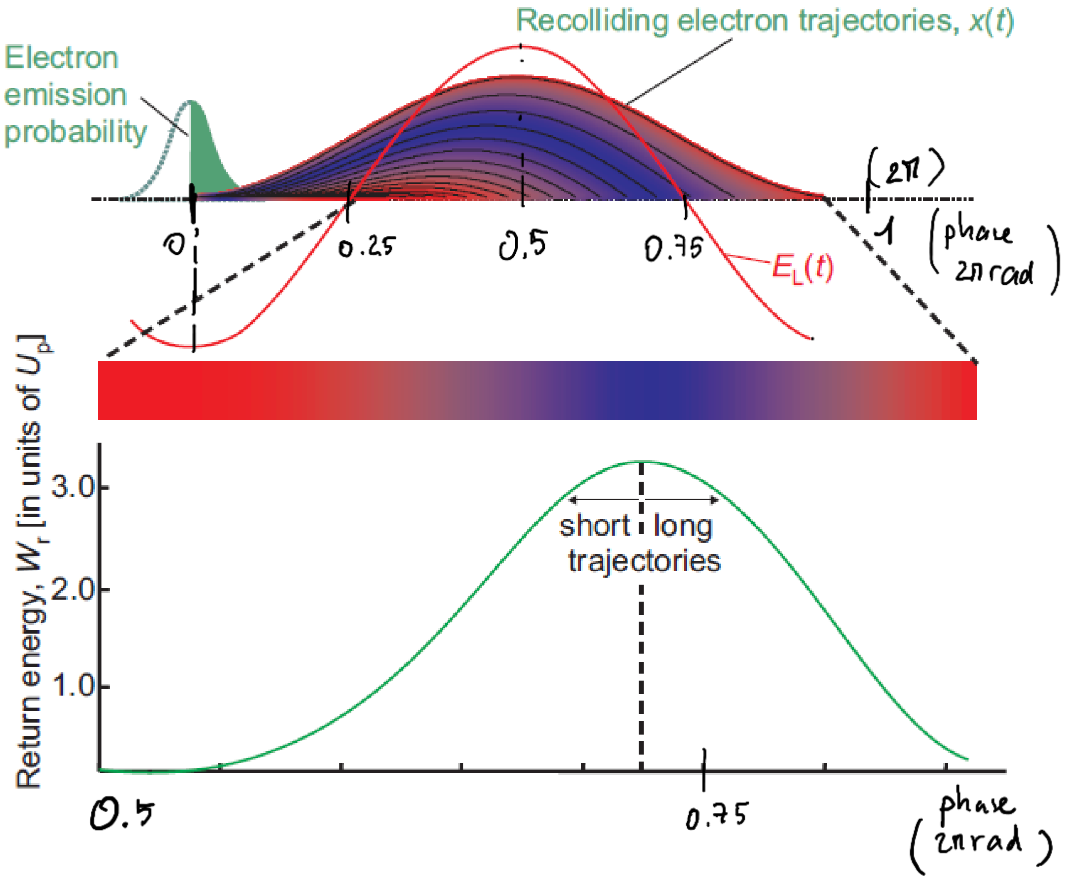
EPFL

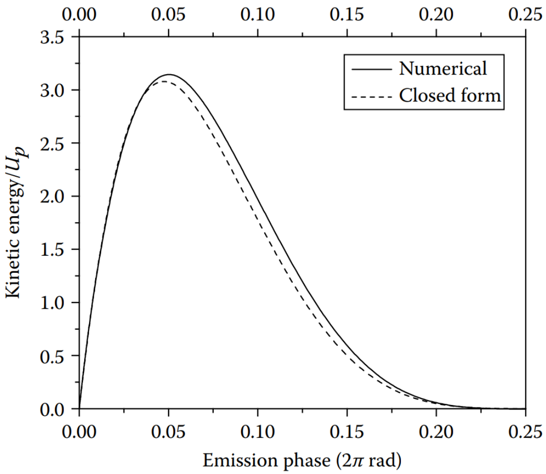
# Long and short trajectories

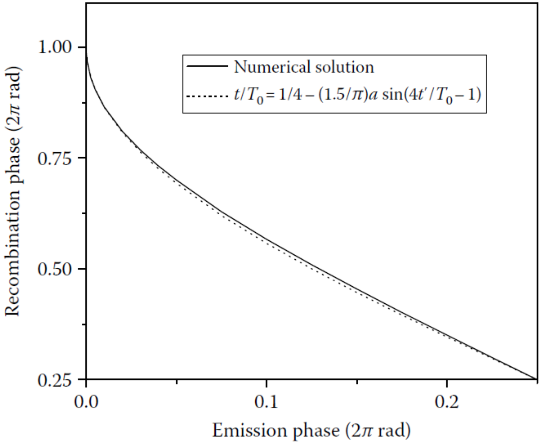
- Kinetic energy depend on the path in the field
- ≈0.5 cycles = 1.3 fs@ 800 nm (FWHM < 1 fs)
- "atto-chirp"





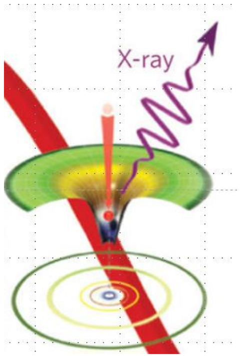


EPFL

# STEP 3: recombination

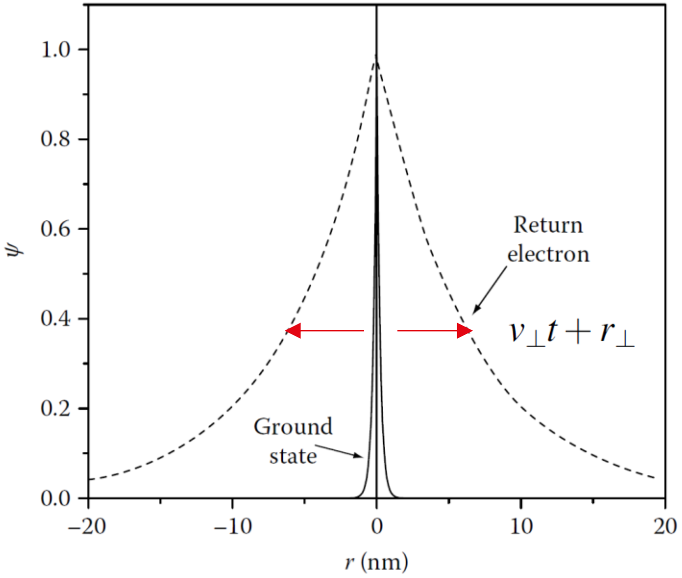



- This step determines the efficiency of the process.
- Quantum diffusion of the wavepacket

- Electron localized at ionization $r_\perp \approx$ atomic radius
- Transverse velocity spread from position-momentum uncertainty

$$v_\perp = \frac{h}{m_e r_\perp}$$

- Wavefunction width increases with time




- The probability depends on the atomic species: heavier noble gases have higher probability

# The emitted photon energy and the cut off law

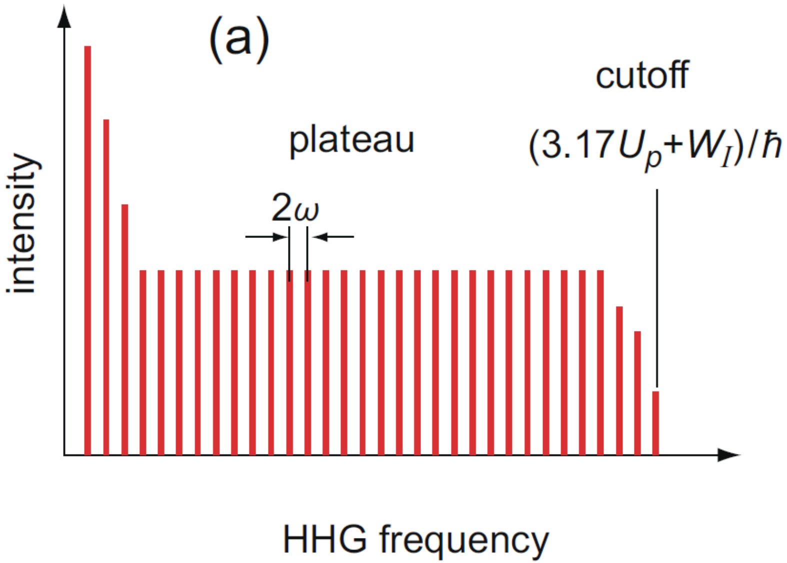


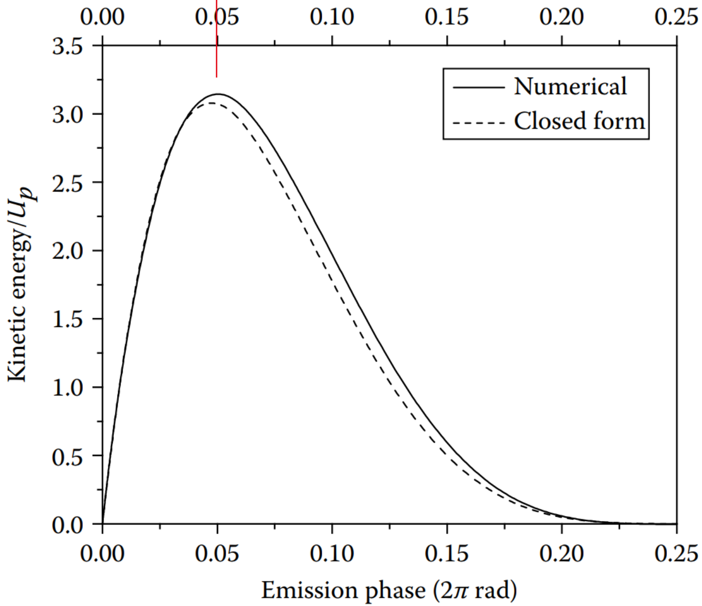


Emitted photon energy:

$$\hbar\omega_X(t) = I_p + \frac{1}{2}mv^2(t) = I_p + 2U_p[\sin(\omega_0 t) - \sin(\omega_0 t')]^2,$$

$$E_{cut} \approx I_p + 3.2U_p$$

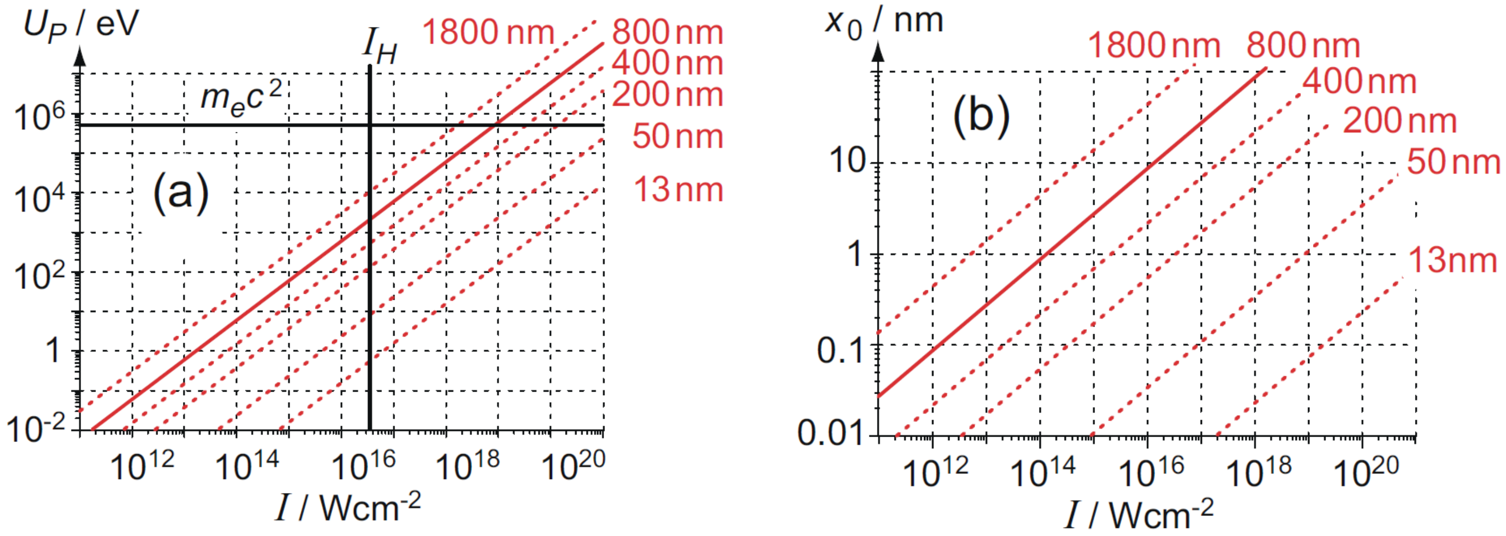


# Wavelenght scaling of HHG:



- Ponderomotive energy increase with the driving wavelength: higher cutoff

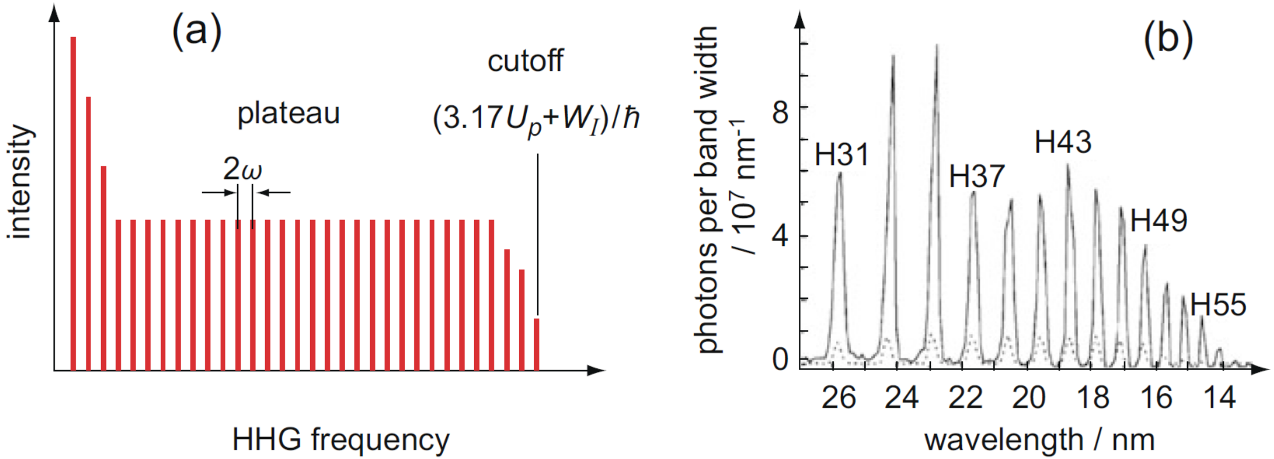



- The probability of recombination decreases with the driver wavelength: short wavelenght are more efficient

NIR $\quad \eta \propto \lambda^{-6.9 \pm 0.2} \quad 0.8\,\mu m < \lambda < 2.0\,\mu m$

VIS $\quad \eta \propto \lambda^{-4.7 \pm 1.0} \quad 0.5\,\mu m < \lambda < 0.8\,\mu m$

- Our model shows that for every  half light cycle there is a burst of short wavelength radiation, with a maximum energy detemined by the cutoff

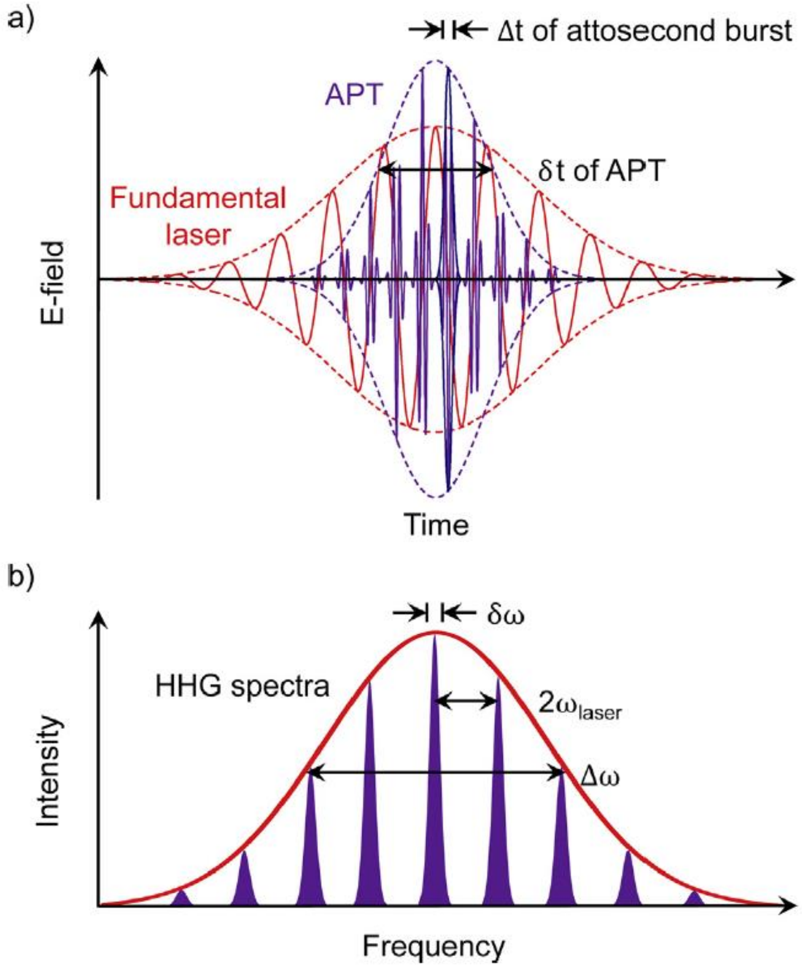



- Why do we observe harmonics in the frequency domain?

# High order harmonics, attosecond pulse train








Time domain structure: periodic train burst of radiation which last less than half-cycle

Broadband XUV pulses with T/2 periodicity (frequency 2ω )

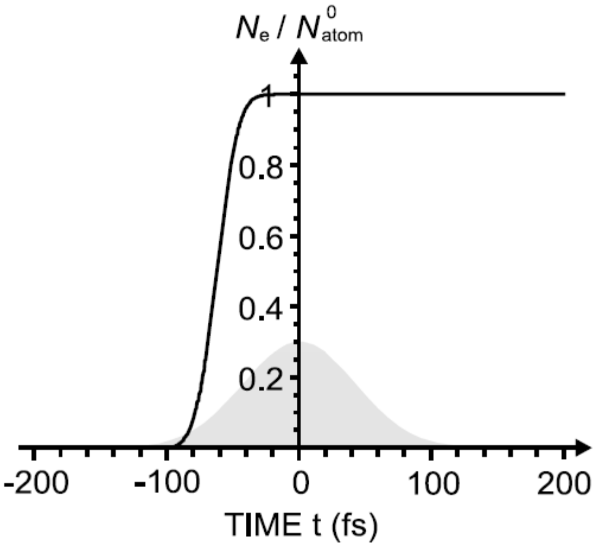
Destructive interference between the continua emitted every half cycle in the inversion symmetric medium

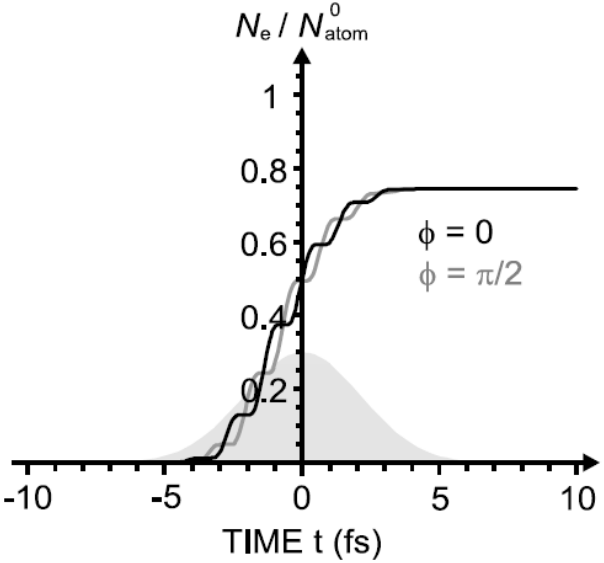
Frequency domain : Frequency comb of odd harmonics.

$$E_{cut} \approx I_p + 3.2U_p \qquad U_p = \frac{e^2\mathcal{E}_0^2}{4m\omega^2}$$

# Ground state depletion: Ionization fraction

$$N_e(t) = N_{\mathrm{atom}}^0 \left[ 1 - \exp\left( - \int_{-\infty}^{t} \Gamma_{\mathrm{ion}}(t')\, \mathrm{d}t' \right) \right]$$




- For high intensity pulses with many cycles the leading edge ionizes the medium fully
- Few-cycle pulses are necessary to extend the cutoff!

# CEP effects with few cycle pulses

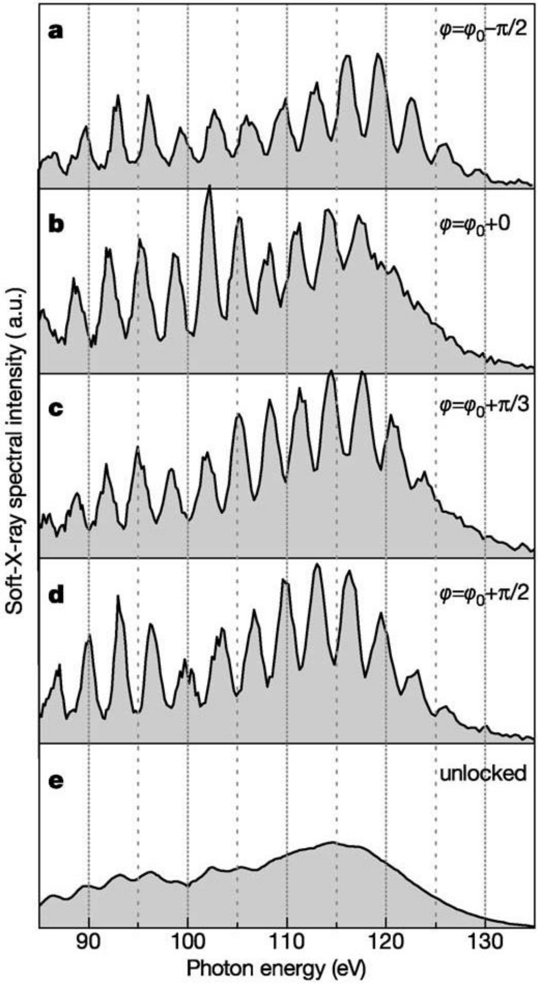
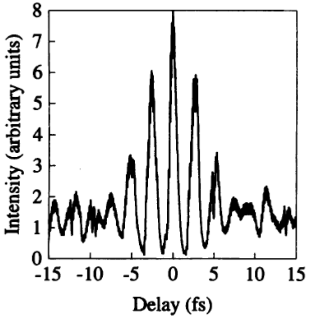


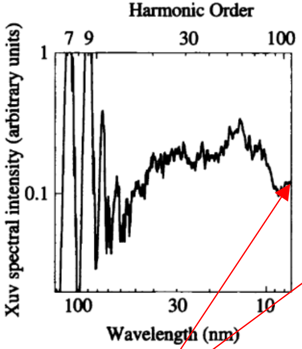

**Fig. 8.4.** Measured EUV spectra from neon at 16 000 Pa (160 mbar) pressure. Excitation is with 5-fs pulses around $\hbar\omega_0 = 1.5\,eV$ and at an estimated intensity of $I = 7 \times 10^{14}\ \mathrm{W/cm^2}$.

# Extending the HHG cutoff



- Increase the field by shortening the pulse




Coherent EUV in the «water window»

C k-edge (280 eV)