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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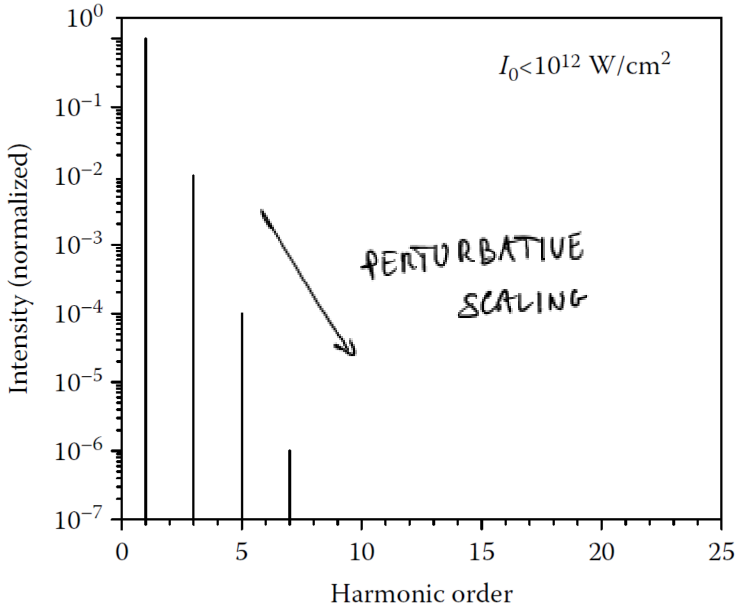
Perturbative nonlinear optics:

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

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:



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

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!

$$\frac{\mathcal{P}^n}{\mathcal{P}^{n-1}} \sim \frac{\chi^{(n)}}{\chi^{(n-1)}} \mathcal{E} \sim \frac{\mathcal{E}}{\mathcal{E}_{at}} \ll 1$$

$$E_{\rm at} = \frac{e}{a_0^2} = \frac{e}{(\hbar^2/me^2)^2}$$

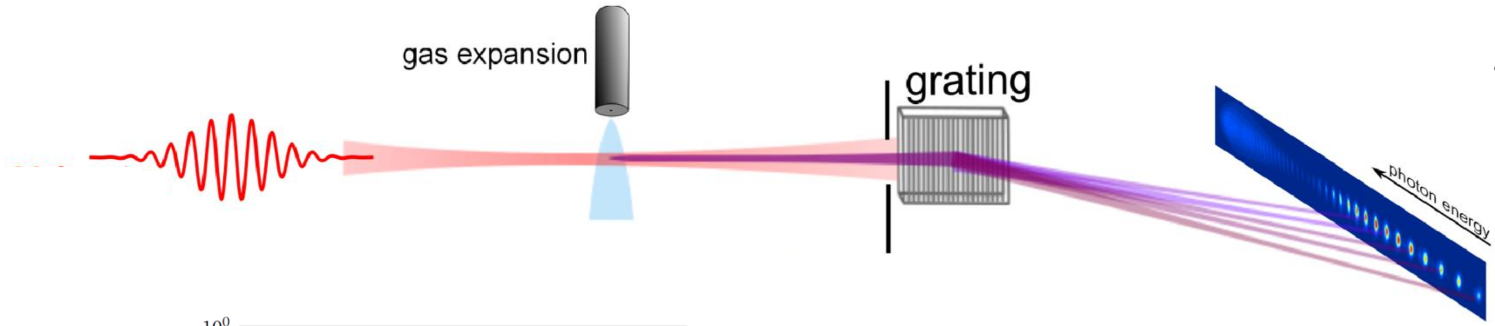
$$I_{\rm at} = \frac{c}{8\pi} E_{\rm at}^2 = 4 \times 10^{16}\ \mathrm{W/cm^2}$$

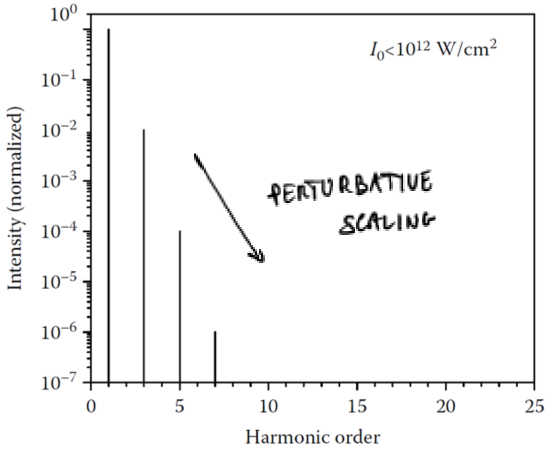
FOR 10^{13} W/cm^2

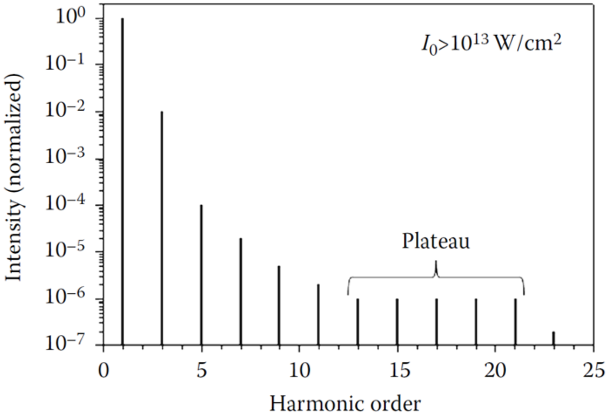
$\mathcal{E}/\mathcal{E}_{at} \sim 10^{-2}$

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

Nonperturbative effects: Harmonic generation in gases



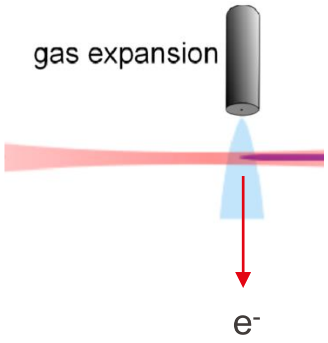


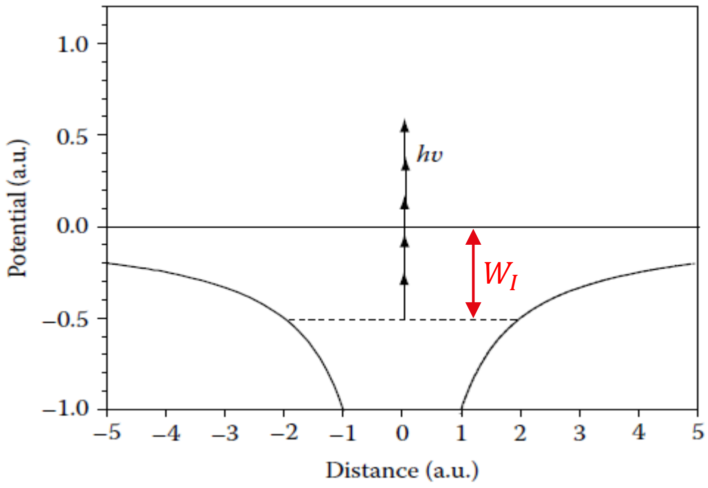


- At intensities approaching ≈10^{13} W/cm^2 the perturbative description fails.

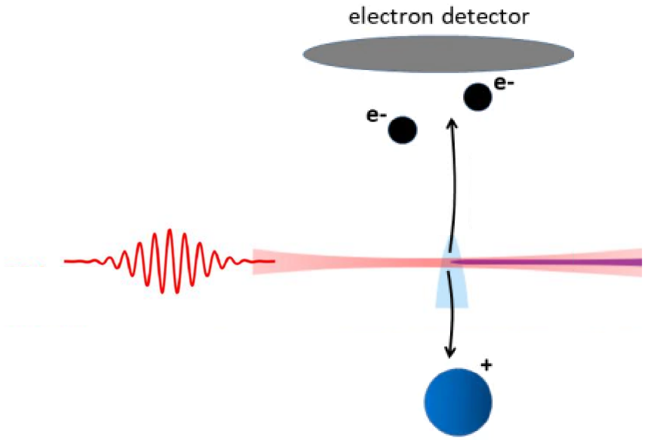
Electron detection : perturbative multiphoton ionization (MPI)

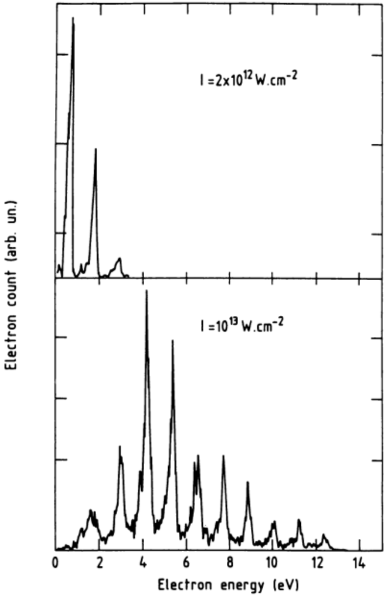
- Atoms can be ionized by a multiphoton process of high order!
- Photoelectrons and positive ions are created for $\hbar\omega \sim 1\ eV \ll W_I \sim 10\ eV$

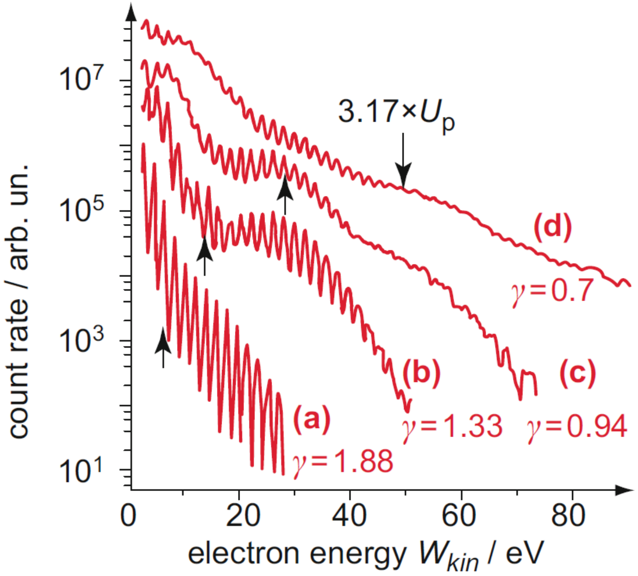




Nonperturbative effects: Above-threshold ionization (ATI)







- Photoelectrons and photoions emission

$$\text{K{\scriptsize ELDYSH}}\ \textit{parameter} \quad \gamma = \sqrt{\frac{W_I}{2U_\mathrm{p}}}$$

- Keldysh parameter
  - $\gamma > 1$ perturbative MPI
  - $\gamma < 1$ non-perturbative ATI

U_p ponderomotive energy:

Classical electron in a periodic E field:

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

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

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

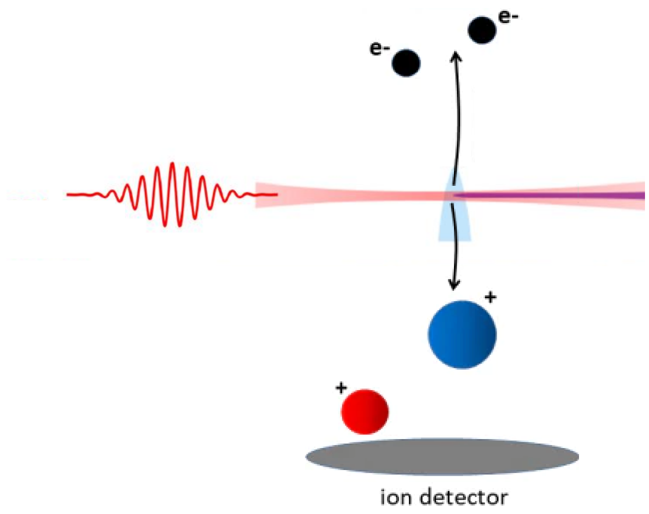
CYCLE AVERAGE

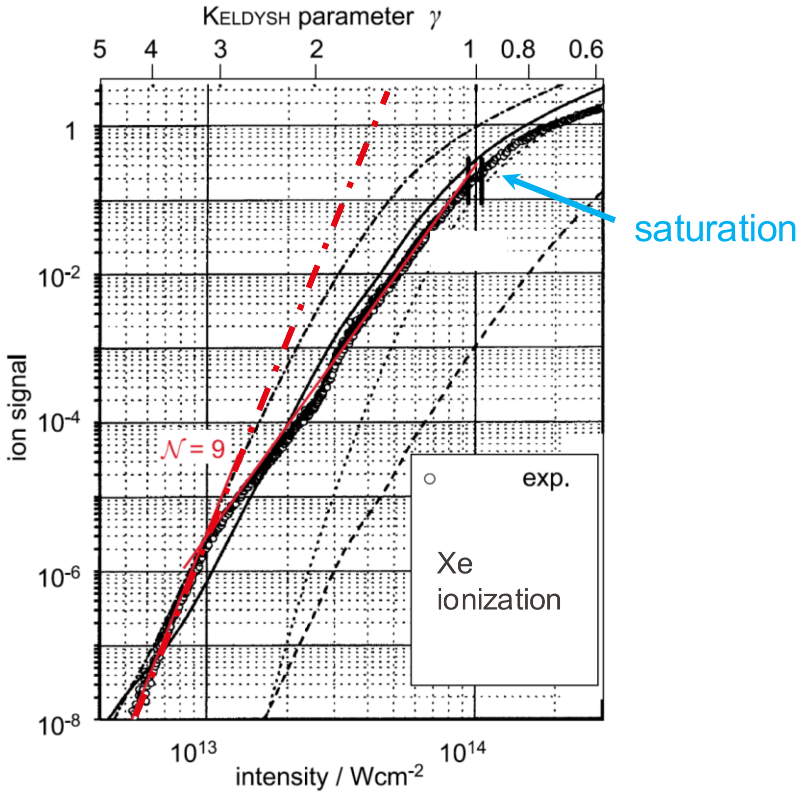
$$\frac{1}{T}\int_0^T f(t)\,dt$$

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

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

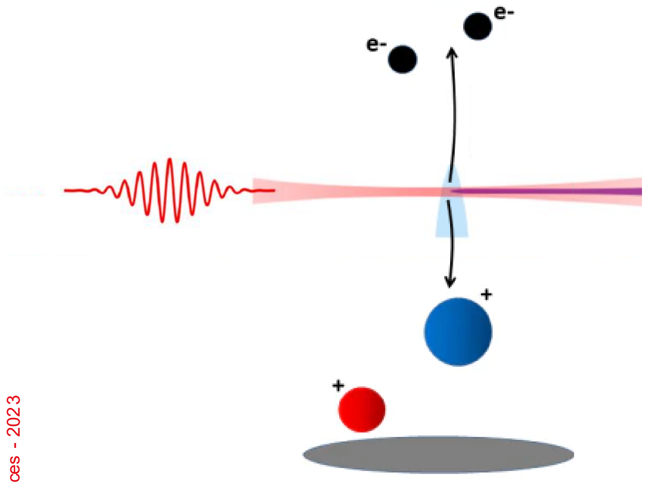
Ionization saturation

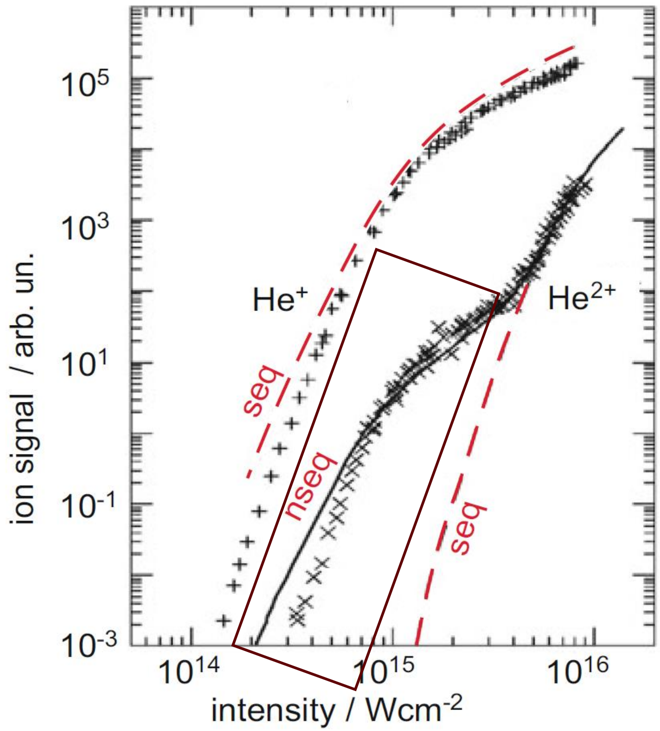




- deviation from perturbative scaling
- saturation due to ground state depletion

Evidence of electron "recollisions"





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

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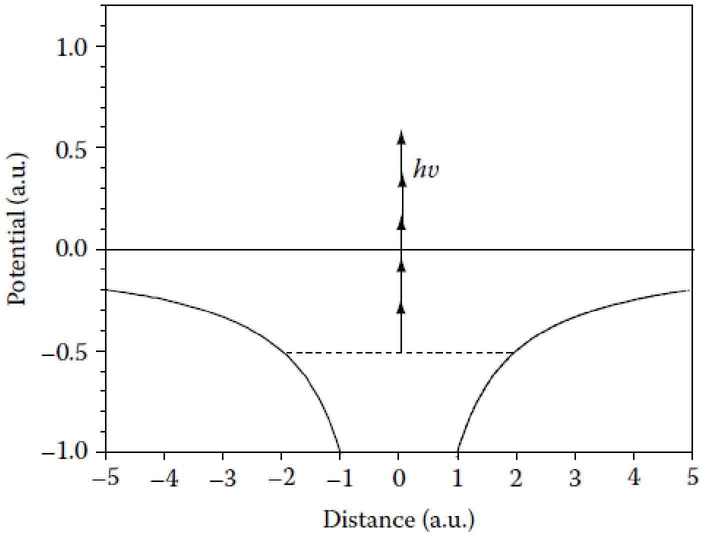
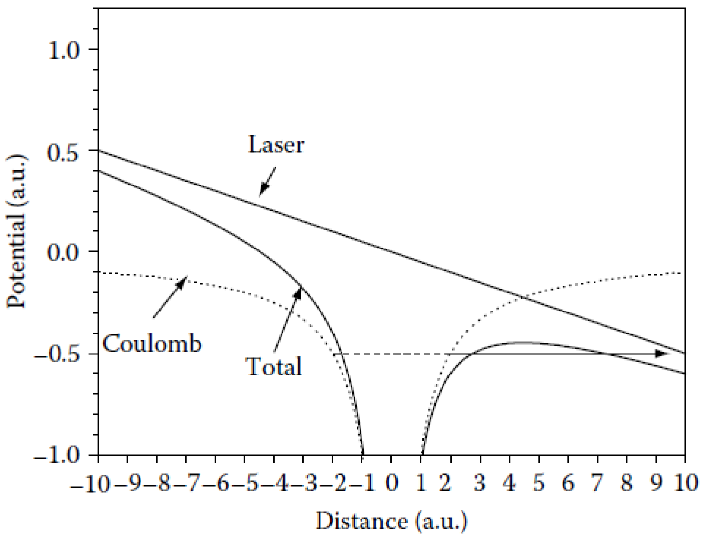
Field ionization:

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



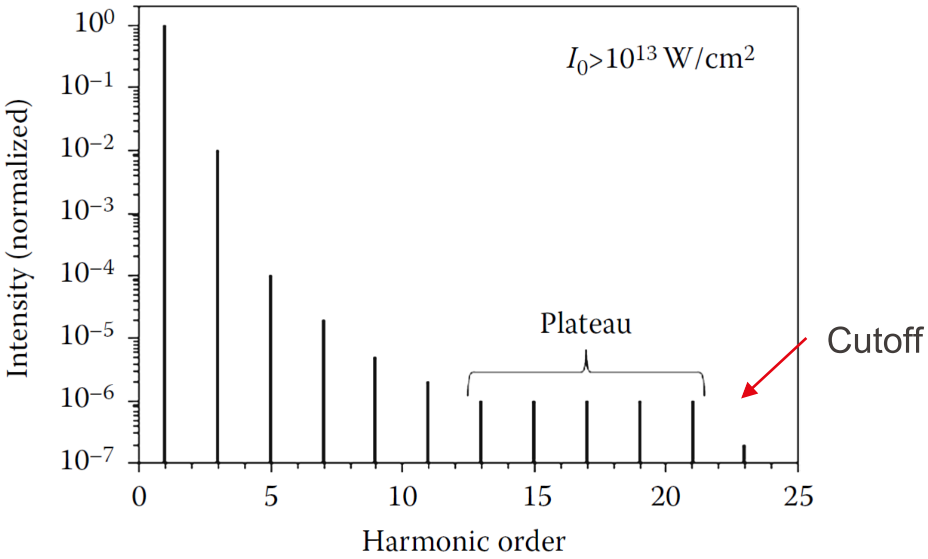
Atomic Coulomb field



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**

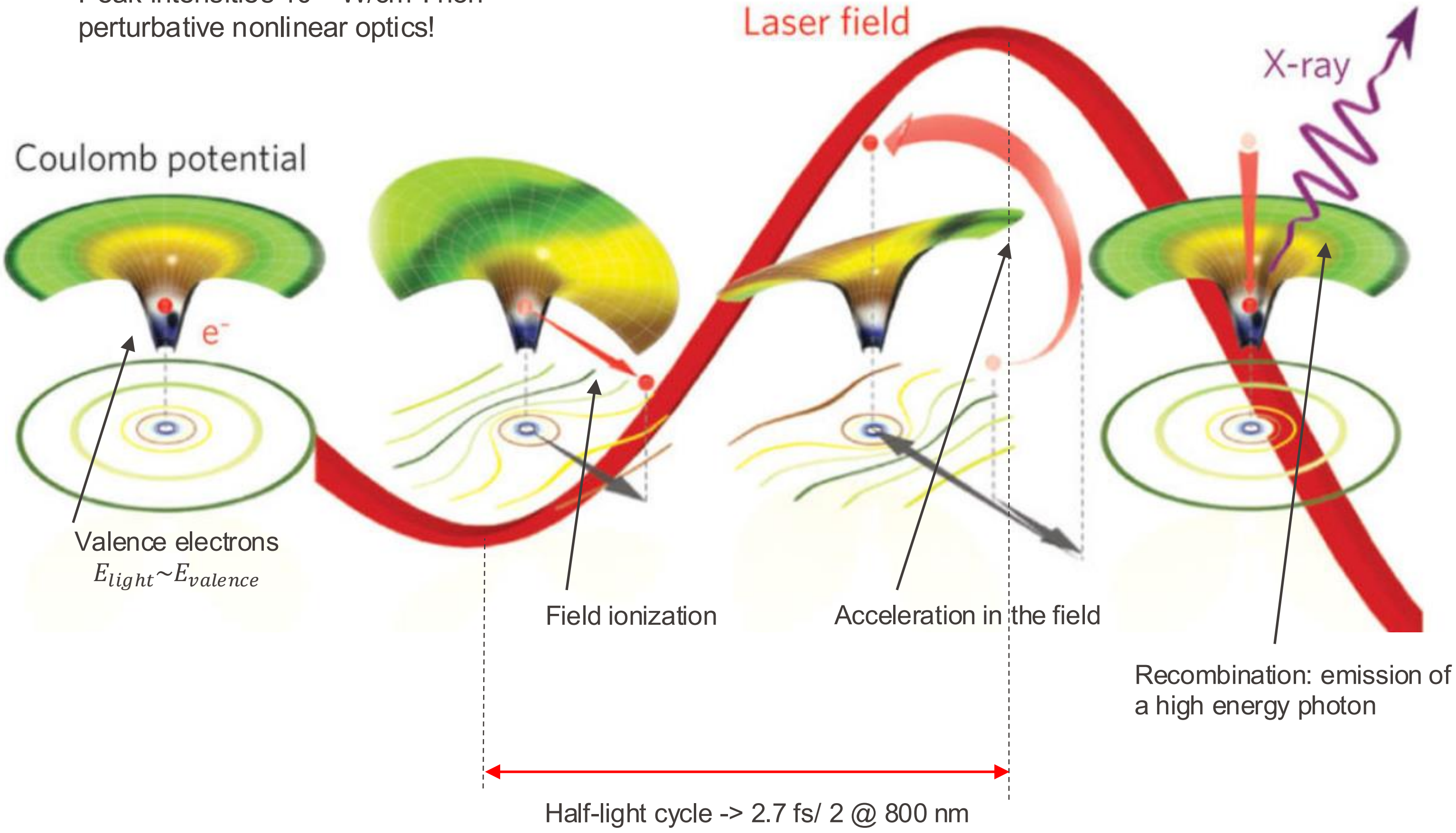
HHG spectrum



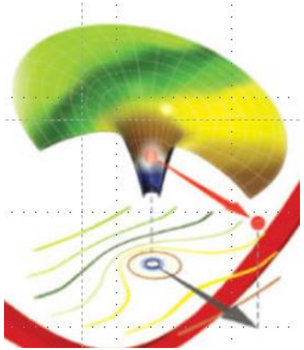
- How can we understand the harmonic generation process in terms of tunneling?
- Why there is “plateau” in the efficiency?
- 2ω spacing?
- What determines the cut-off?
- What is the time structure of the harmonics?

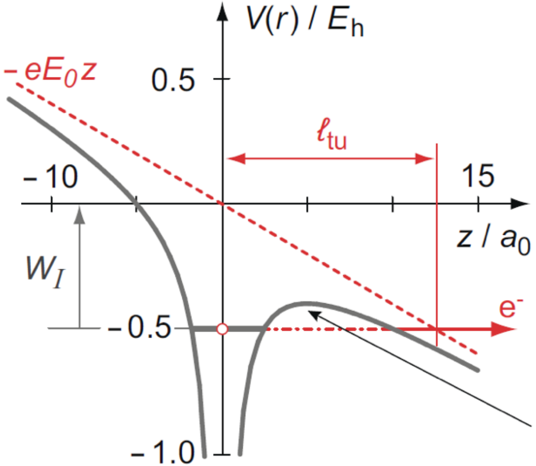
High-harmonic generation: three-step model

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



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

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

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

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

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Keldysh parameter

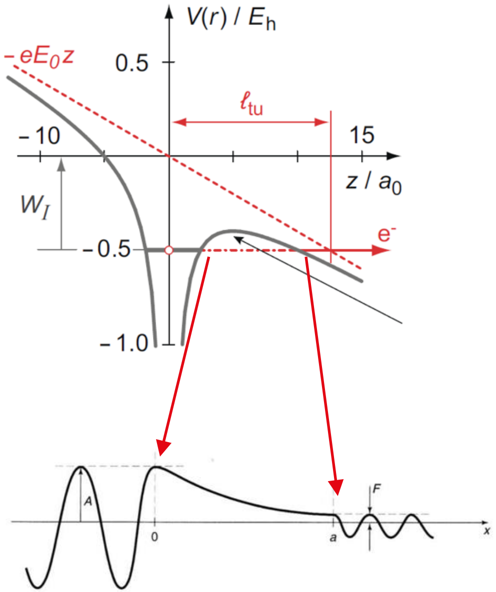
$$U_p = \frac{e^2 E_0^2}{4 m_e \omega^2}$$

TUNNELING TIME

HALF-CYCLE DURATION

$$2\pi \frac{t_{Tu}}{t_{1/2}} = t_{Tu}\, 2\omega = \sqrt{\frac{m_e W_1}{2 e^2 E_0^2}}\, 2\omega = \sqrt{\frac{W_1\, 4\omega^2 m_e}{2\, e^2 E_0^2}} = \sqrt{\frac{W_1}{2 U_p}} = \gamma$$

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



At a certain time t:

- 1d static tunneling problem   barrier length = l(t) (depends on E(t))
- Tunneling probability -> amplitude of the exponentially damped wavefunction

$$\psi(x) \propto \exp(-|k_x(x)|\, x)$$

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Crude approximation: rectangular energy barrier, stationary solution

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

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

Electric field appear in an exponential function!

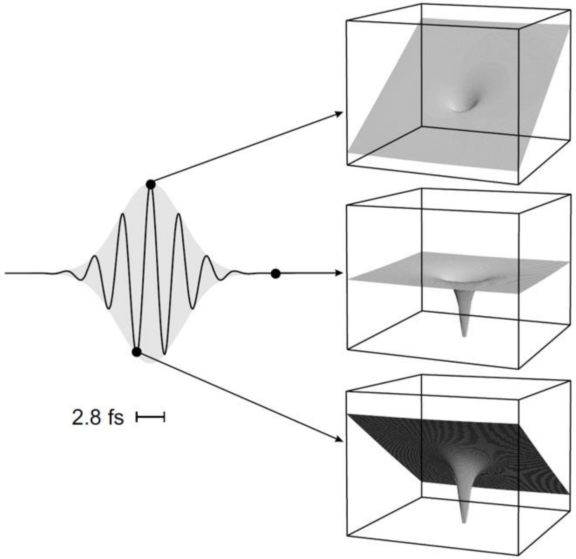
More accurate: ADK ionization rate (Ammosov Delone Krainov 1986)

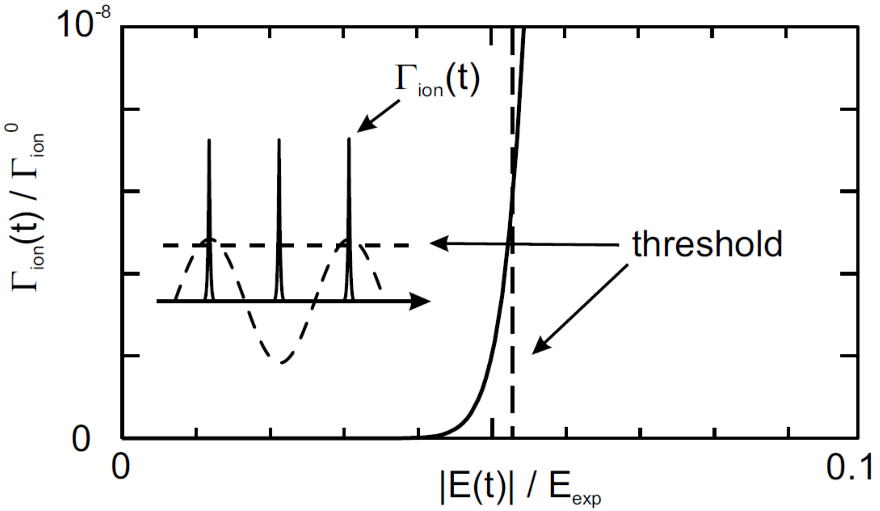
TABLE 4.2 The ADK Parameters

	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

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

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

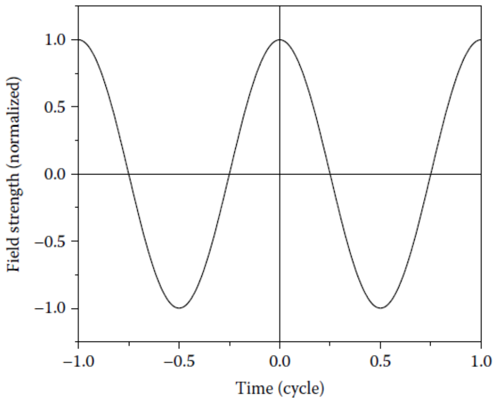




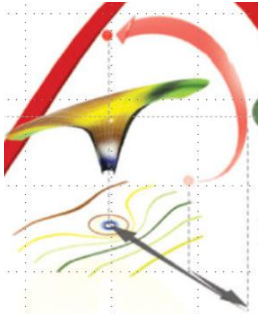
- Threshold-behaviour due to the tunneling probability
- Tunneling act as a fast (< half-cycle) shutter.
- Tunneling possible only for high field amplitude near the maximum

$$\exp\left(-\frac{\frac{1}{\hbar e}\sqrt{2m_e}\,W_1^{3/2}}{E(t)}\right) = e^{-\frac{E_{\mathrm{exp}}}{|E(t)|}}$$

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

MAX DISPLACEMENT

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

↓

RETURN TIME?

EPFL

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

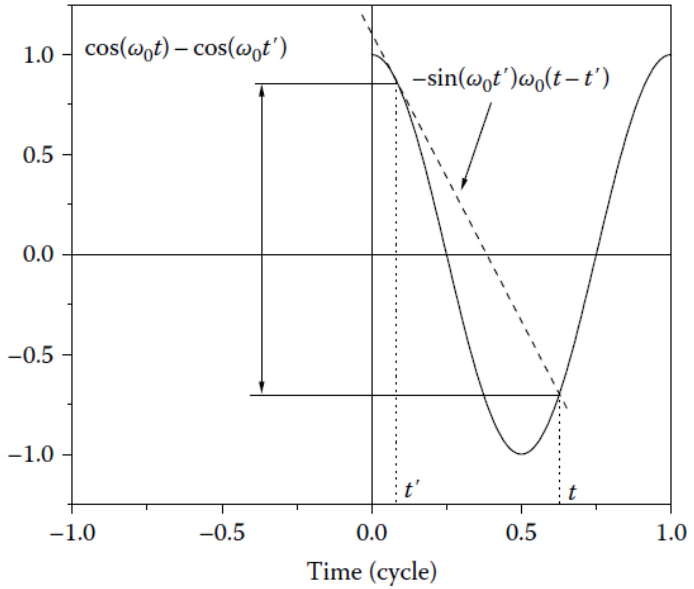
Equivalent form of x(t)=0

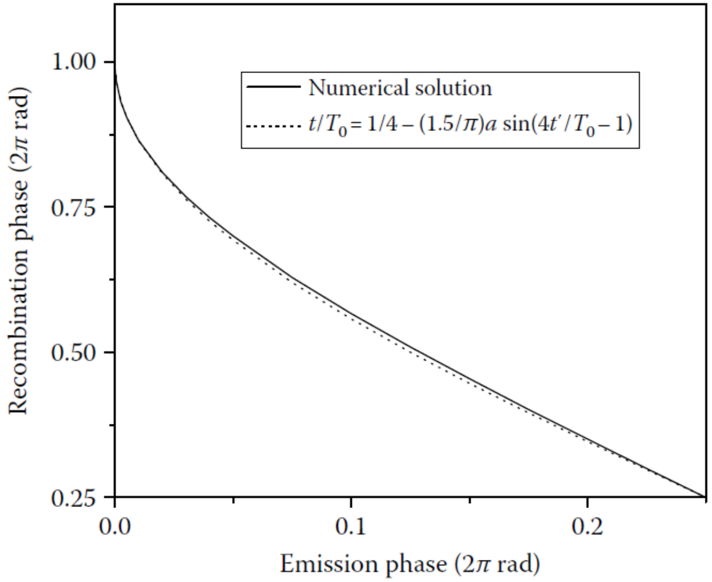
$$x(t) = \frac{eE_L}{m_e \omega_0^2} \{ [\cos(\omega_0 t) - \cos(\omega_0 t')] + \omega_0 \sin(\omega_0 t')(t - t') \},$$

Graphical solution:

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

(units of x_0)





Kramer-Hennerberger frame of reference

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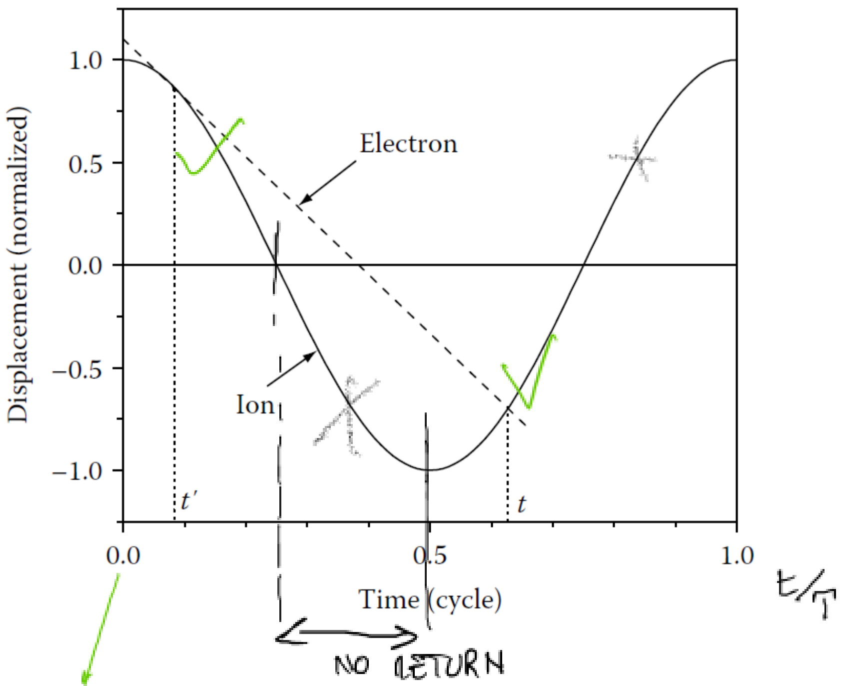
- Physical interpretation of the graphical solution:
  - ion moving in an oscillatory frame ($x' = x - x_0 \sin(\omega_0 t)$)
  - Electron performs a linear trajectory



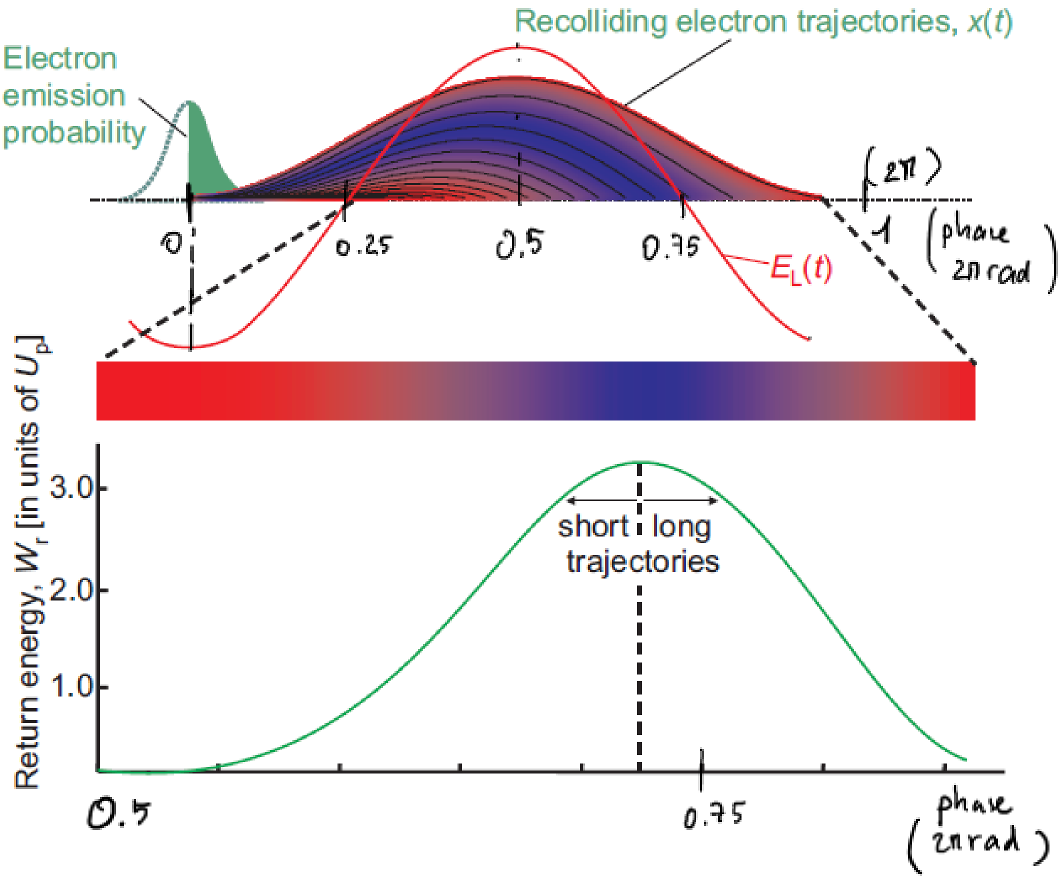
$\omega t' = 0$ → RETURN AFTER 1 CYCLE $\omega t = 2\pi$

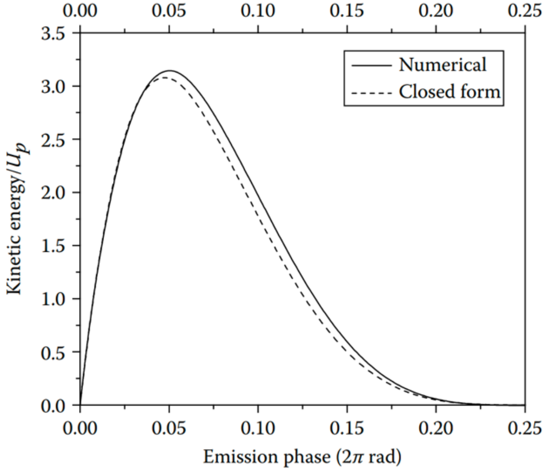
$\pi/2 < \omega t < \pi$ → THE ELECTRON DON'T RETURN

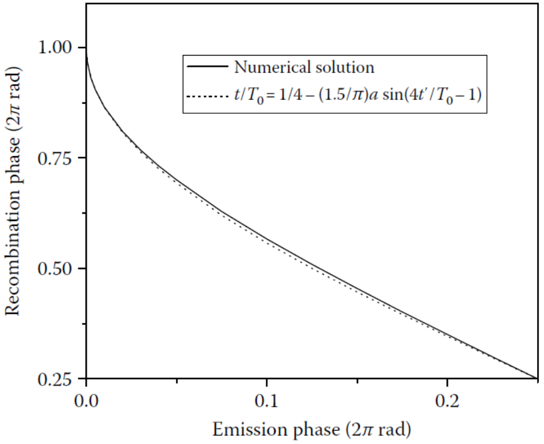
EPFL

Long and short trajectories

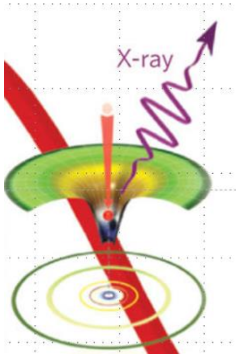
- Kinetic energy depend on the path in the field
- ≈0.5 cycles = 1.3 fs@ 800 nm (FWHM < 1 fs)
- “atto-chirp”







STEP 3: recombination

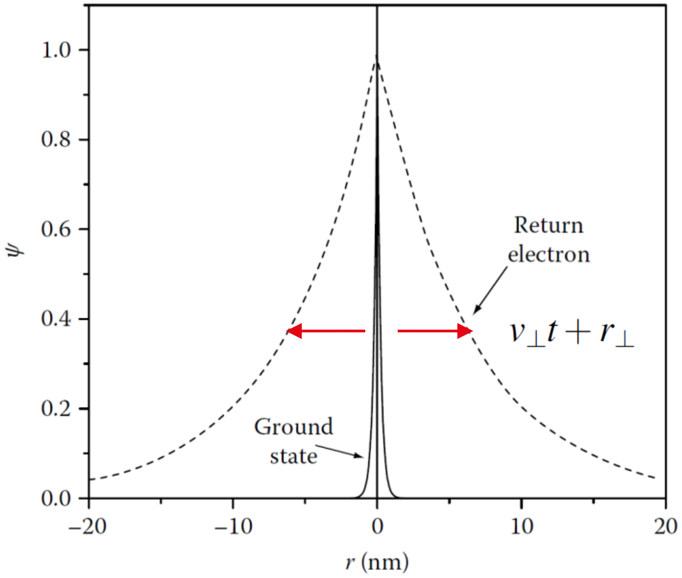


- This step determines the efficiency of the process.
- Quantum diffusion of the wavepacket

- Electron localized at ionization r⊥≈ atomic radius
- Transverse velocity spread from position-momentum uncertainty

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

- Wavefunction width increases with time



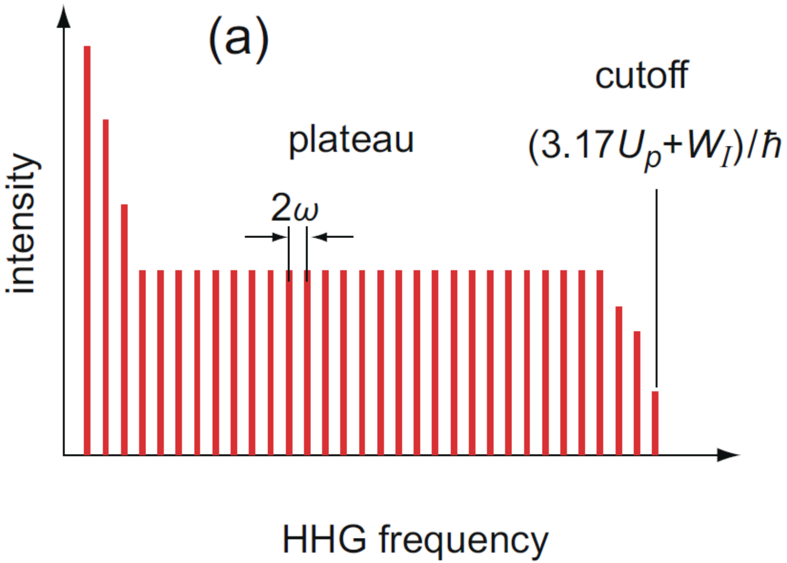
- The probability depends on the atomic species: heavier noble gases have higher probability

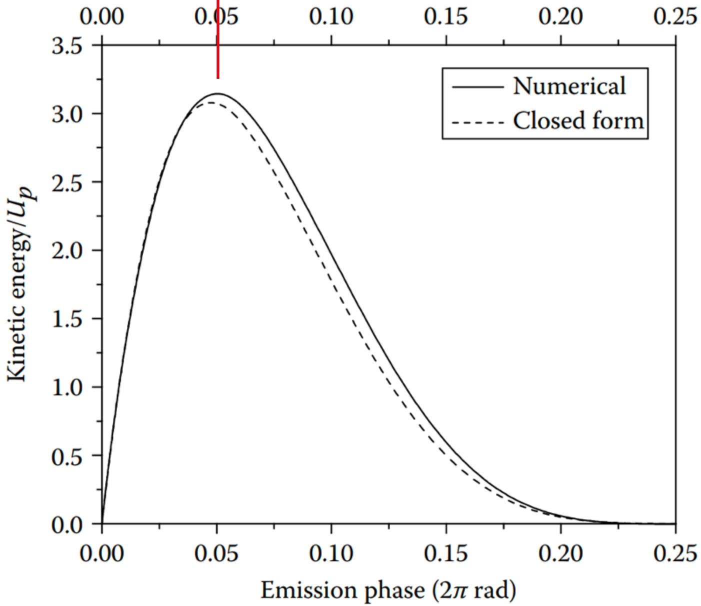
The emitted photon energy and the cut off law

Emitted photon energy:

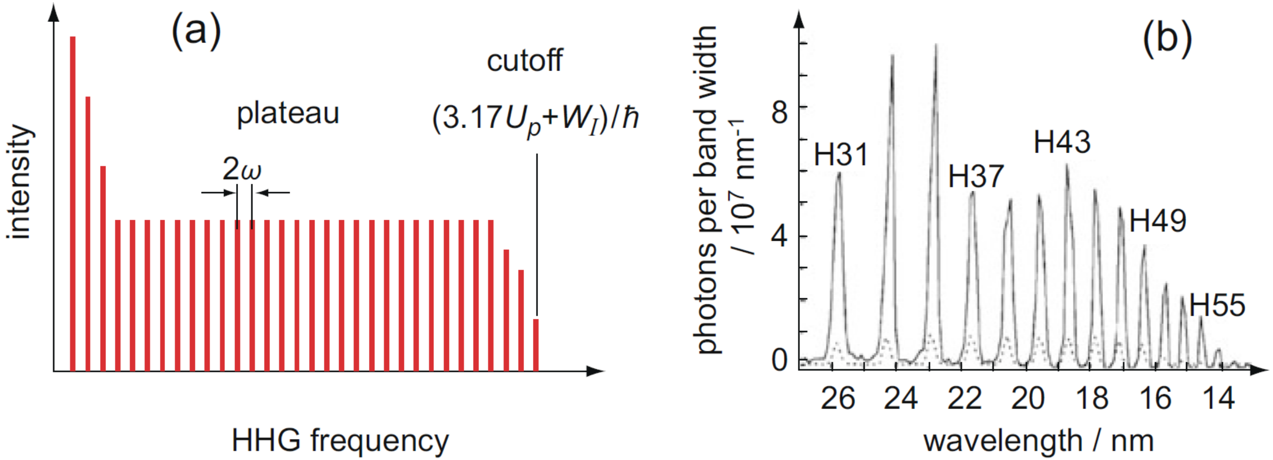
$$\hbar\omega_X(t) = I_p + \frac{1}{2}mv^2(t) = I_p + 2U_p[\sin(\omega_0 t) - \sin(\omega_0 t')]^2,$$

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



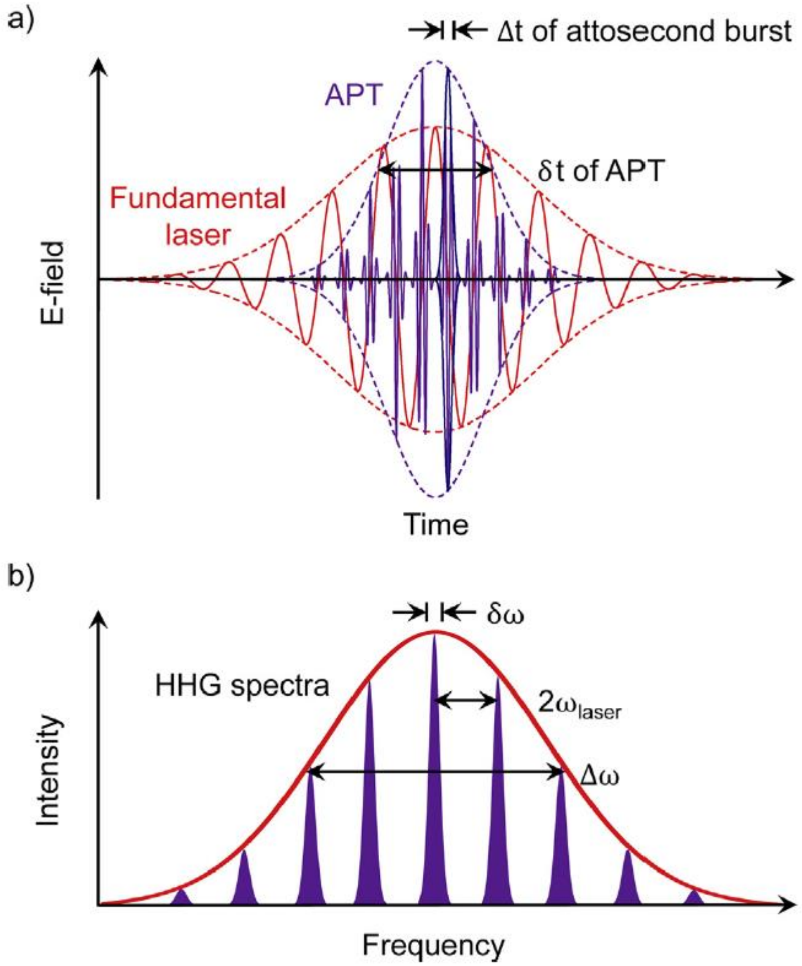


- Our model shows that for every half light cycle there is a burst of short wavelength radiation, with a maximum energy detemined by the cutoff



- 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)

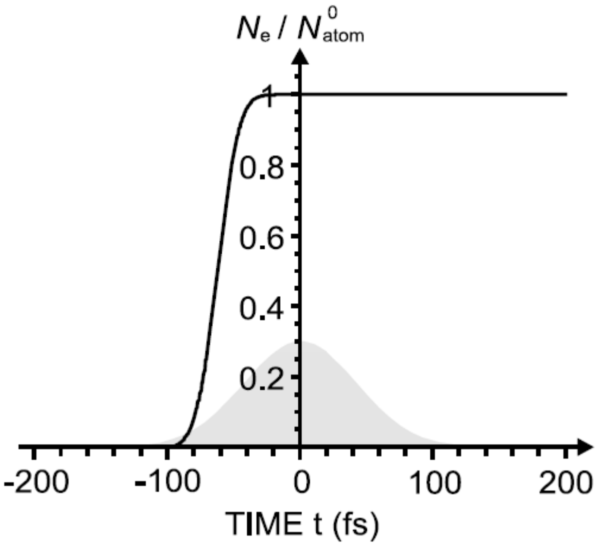
Destructive interference between the continua emitted every half cycle in the inversion symmetric medium

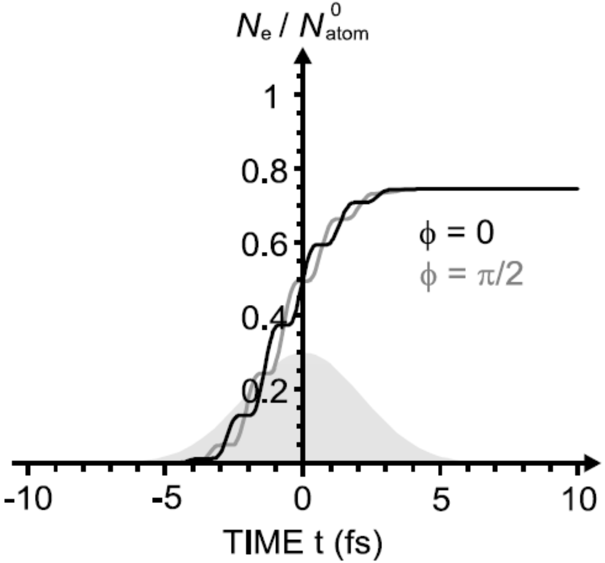
Frequency domain : Frequency comb of odd harmonics.

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

Ground state depletion: Ionization fraction

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

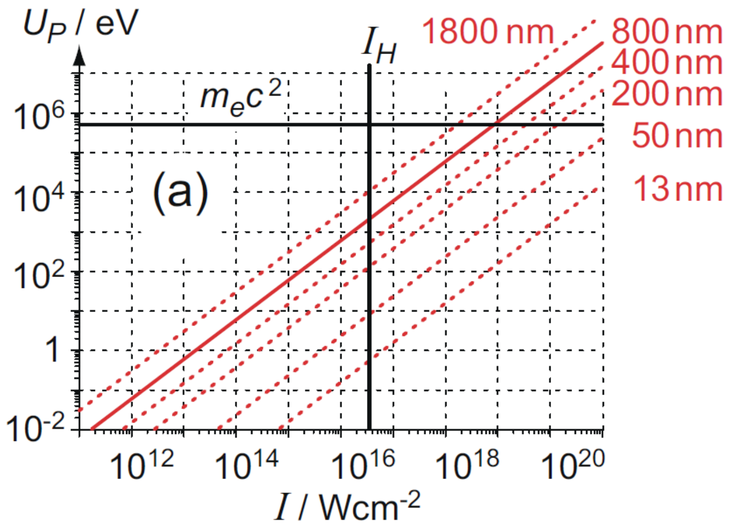


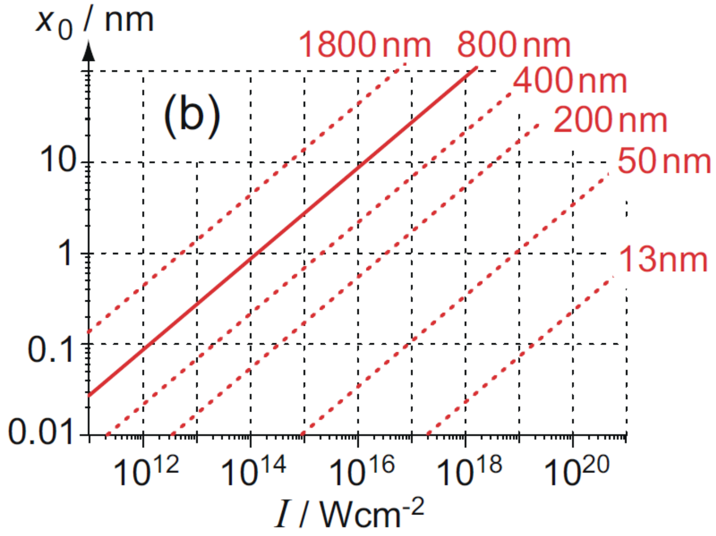


- For high intensity pulses with many cycles the leading edge ionizes the medium fully
- Few-cycle pulses are necessary to extend the cutoff!

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

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CEP effects with few cycle pulses

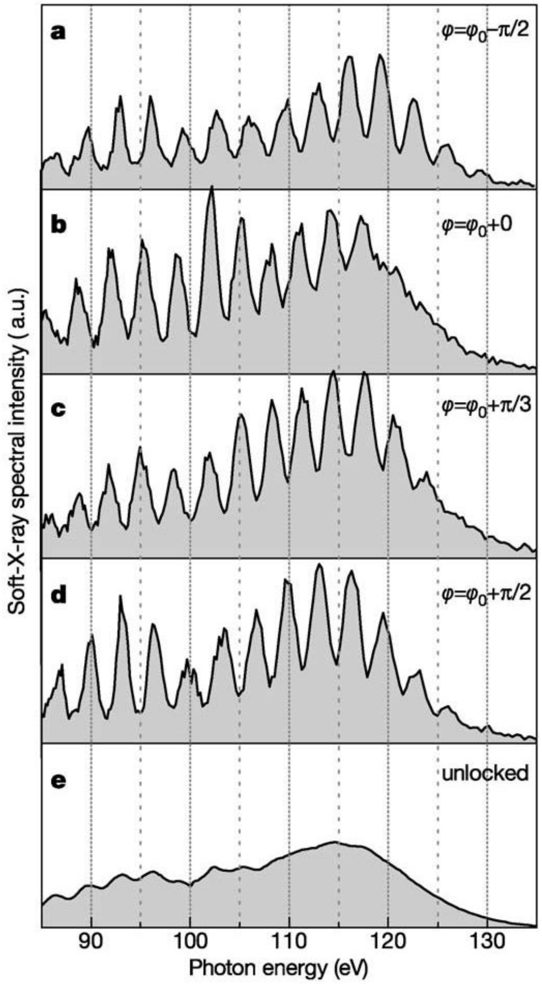

Fig. 8.4. Measured EUV spectra from neon at 16 000 Pa (160 mbar) pressure. Excitation is with 5-fs pulses around $\hbar\omega_0 = 1.5\,eV$ and at an estimated intensity of $I = 7 \times 10^{14}$ W/cm^2.

Extending the HHG cutoff

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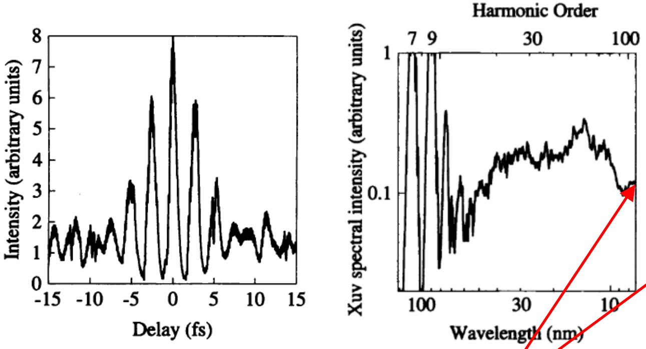
- Increase the field by shortening the pulse



Coherent EUV in the «water window»

C k-edge (280 eV)