# Advanced Radiation Sources - PHSY761

Lecture 07

## 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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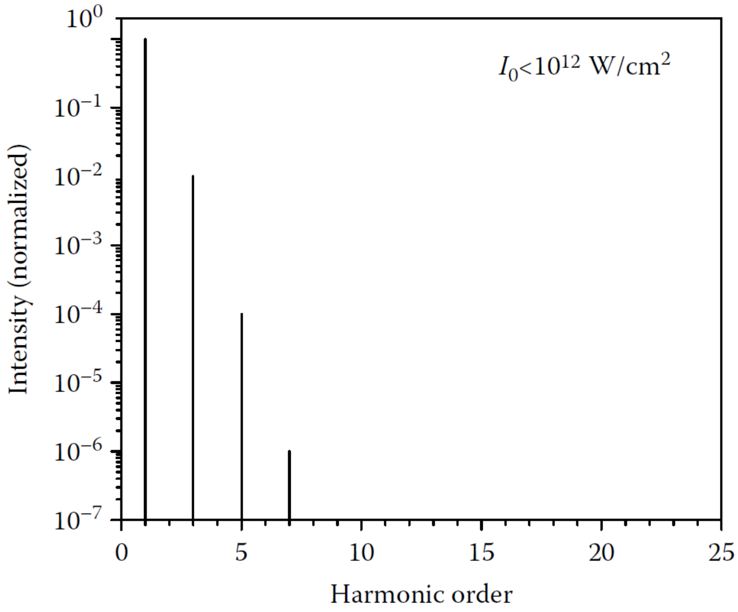
Perturbative nonlinear optics:

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

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

- Coherent sources where no lasing transitions are available
- Valuable for spectroscopic studies of matter
- Table-top sources vs large facilities

# Expected scaling laws 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:

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

Pulse duration scaling:

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

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

Possible to generate short pulses!

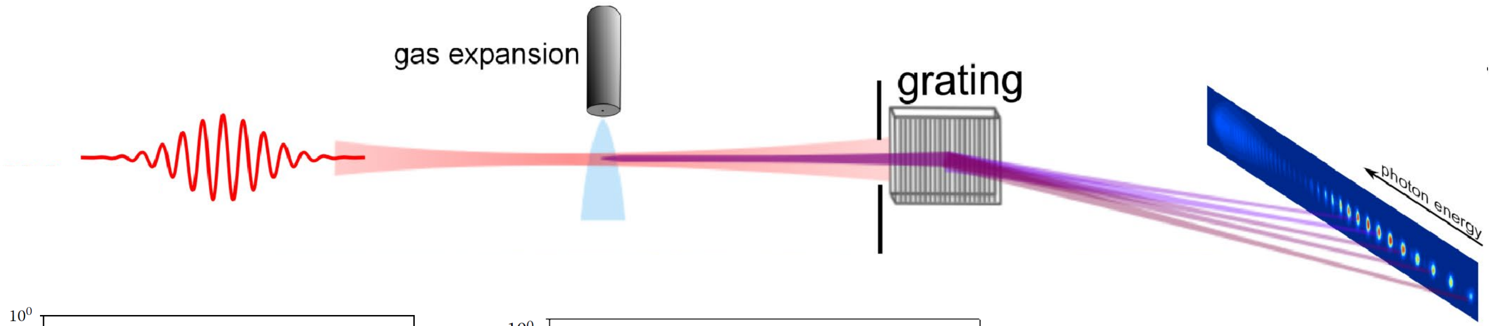
To observe a high-order process the intensity has to be increased!

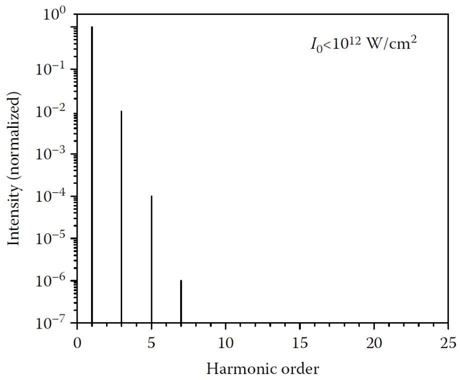
$$I_{\mathrm{at}} = \frac{c}{8\pi} E_{\mathrm{at}}^2 = 4 \times 10^{16}\ \mathrm{W/cm}^2$$

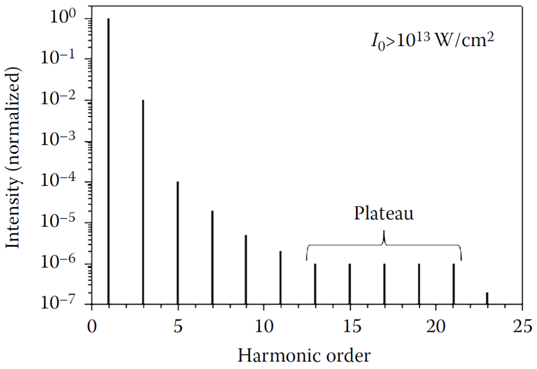
$$E_{\mathrm{at}} = \frac{e}{a_0^2} = \frac{e}{(\hbar^2/me^2)^2}$$

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

# Harmonic generation in gases



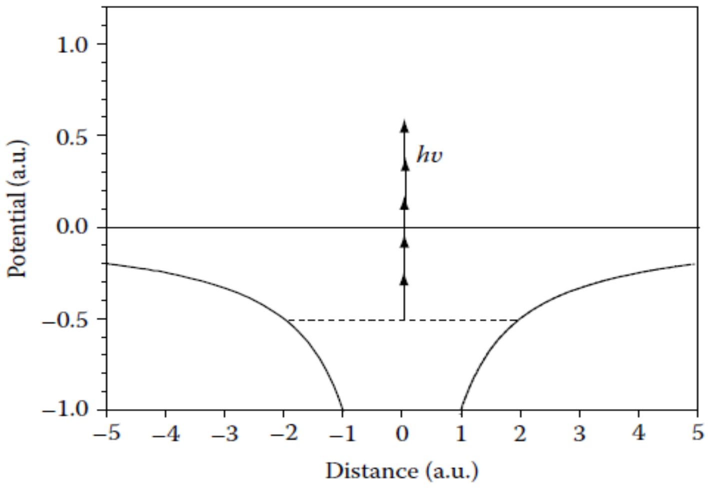




- At intensities approaching $\approx 10^{13}$ W/cm$^2$ the perturbative description fails.

# Multiphoton ionization

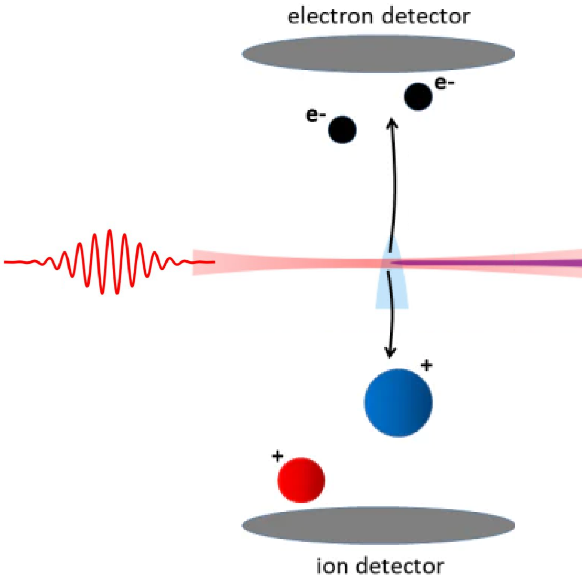
- Atoms can be ionized by a multiphoton process of high order!

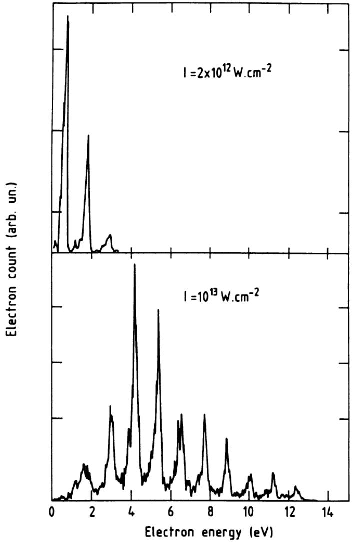


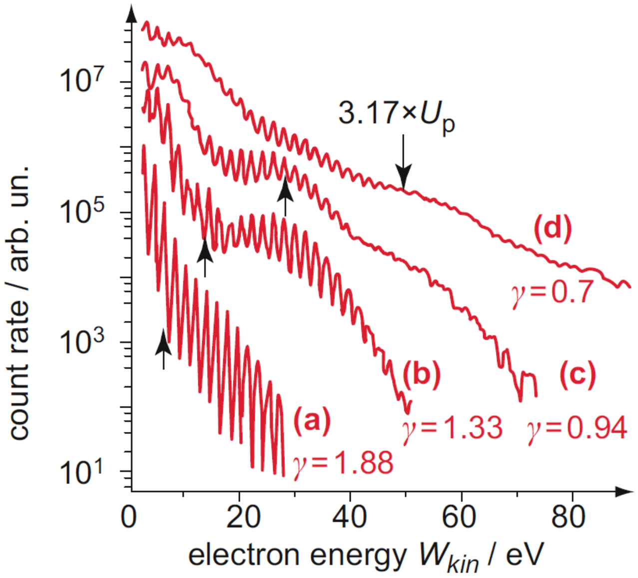

- Photoelectrons and positive ions are created for $\hbar\omega \sim 1\ eV \ll I_P \sim 10\ eV$

# Above-threshold ionization





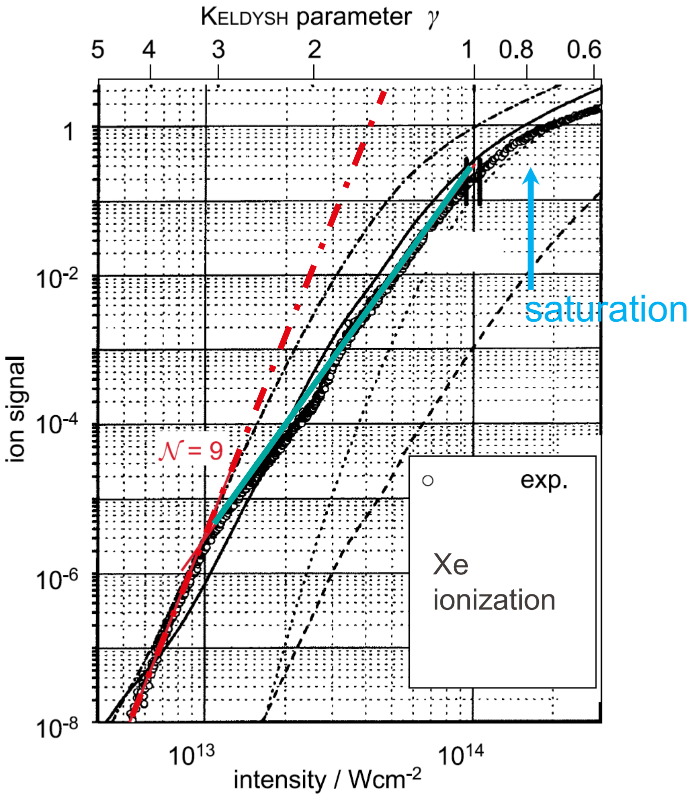




- Keldysh parameter
  - γ >1 perturbative MPI
  - γ <1 non-perturbative ATI

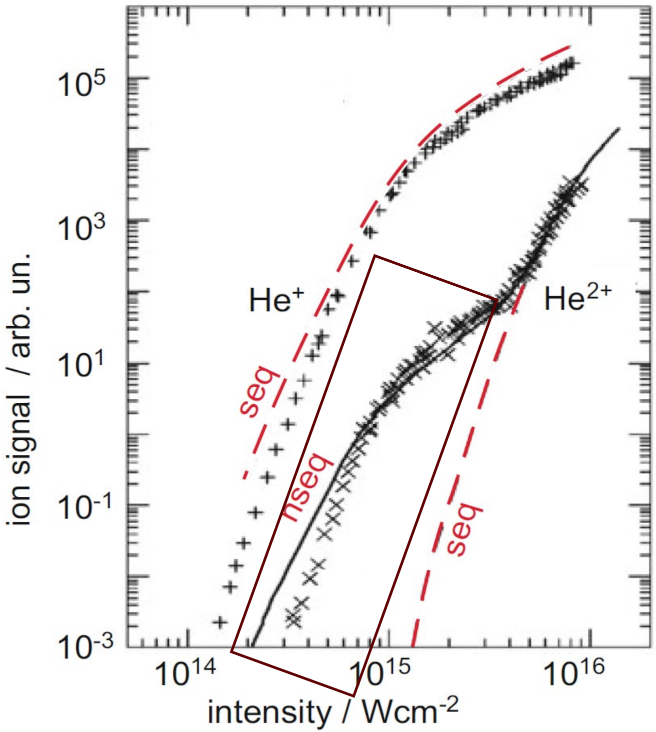
KELDYSH *parameter* $\gamma = \sqrt{\frac{W_I}{2U_p}}$

EPFL

# Ionization saturation and electron "recollisions"




- Non-sequential double ionization (Ar)




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

- Evidence for "electron" re-scattering with to the parent ions



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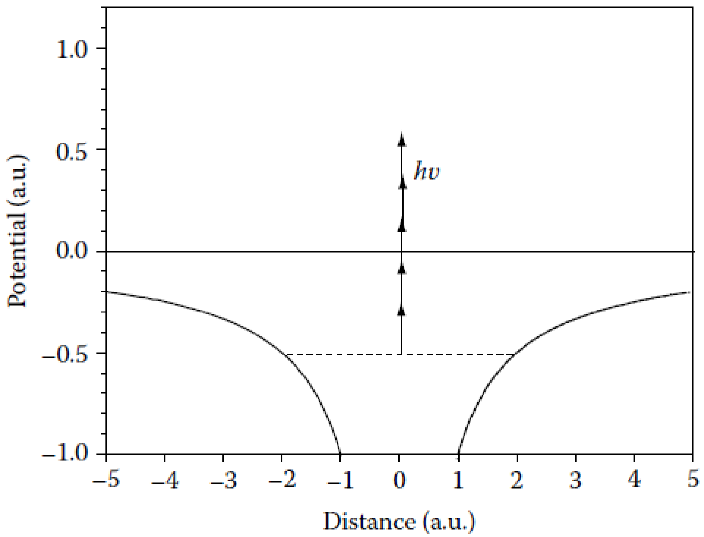
# Strong-field physics:

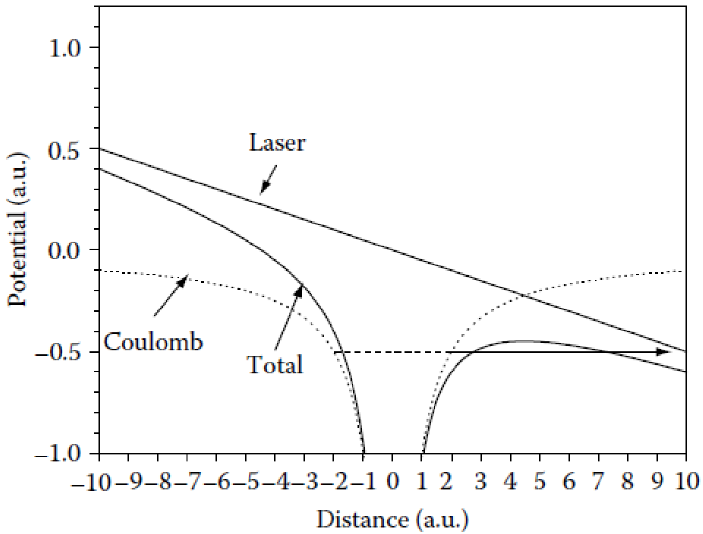




- At high intensities the electric field is of similar magnitude to the atomic Coulomb field




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”
- How can we understand in which regime we are?

# Ponderomotive energy

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Classical electron in a periodic E field:

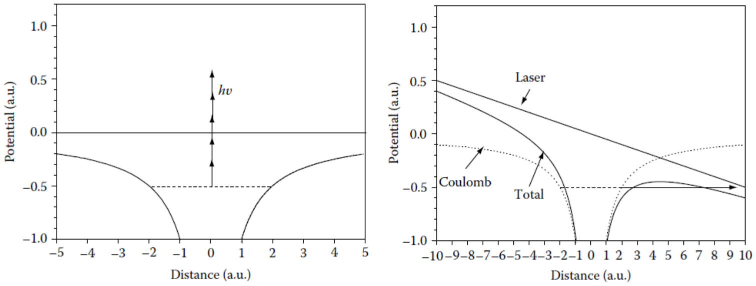
$$m_e \frac{dv}{dt} = eE_0 \cos \omega t$$

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

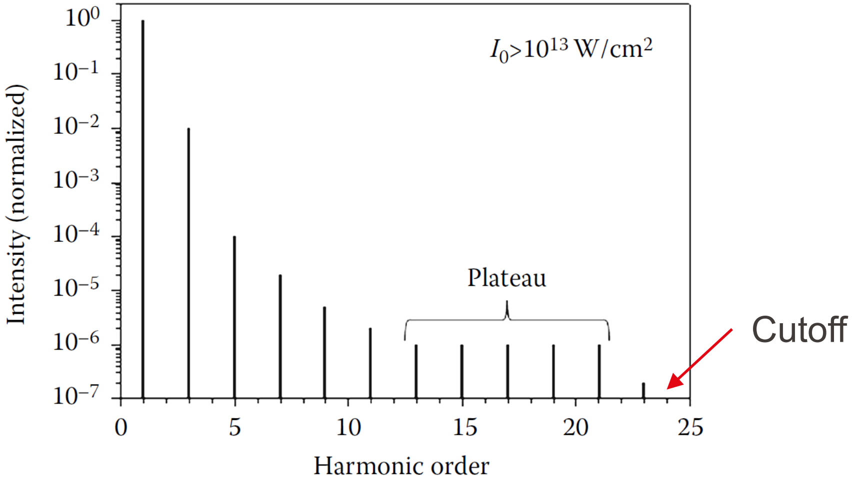
$$U_p = \overline{\frac{1}{2} m_e v^2} = \frac{e^2 E_0^2}{4 m_e \omega^2}$$

Energy scales: ponderomotive energy and ionization potential of the atom

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



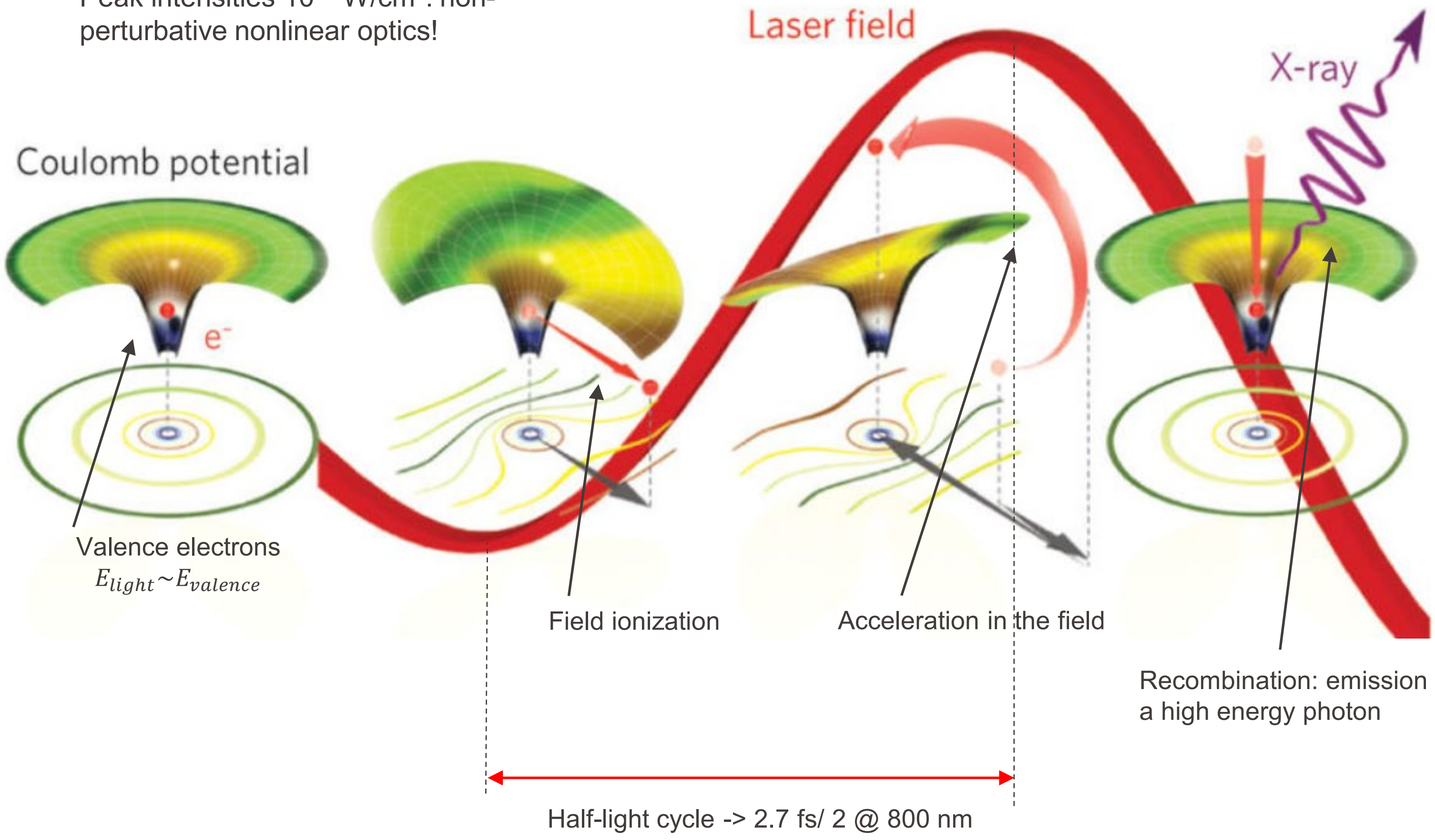
# HHG spectrum




- 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

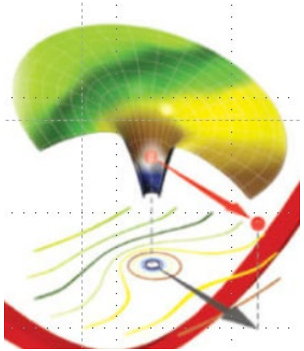
Peak intensities $10^{14}$ W/cm$^2$: non-perturbative nonlinear optics!

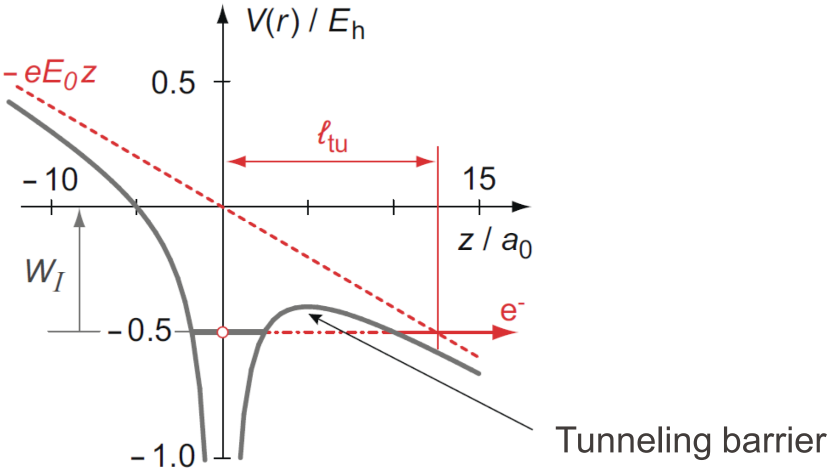


# STEP 1 : ionization





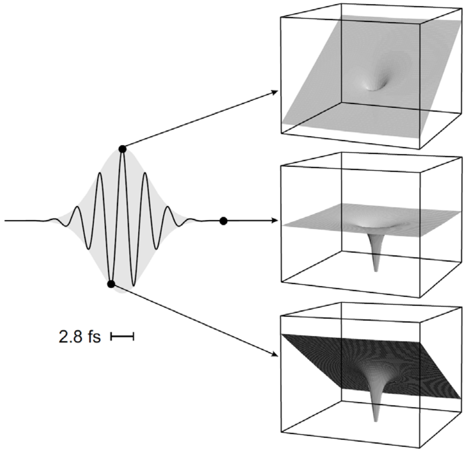


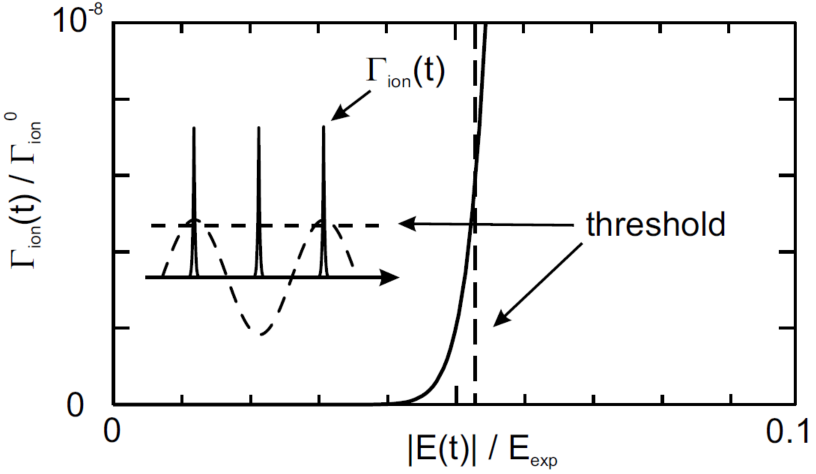



$$\ell_{tu} = W_I/(eE_0)$$

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

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

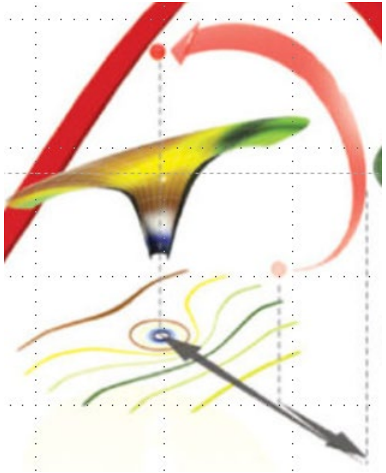



- Tunneling act as a fast (< half-cycle) shutter.
- Tunneling possible only for high field amplitude near the maximum

# STEP 2 – acceleration in the field

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



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

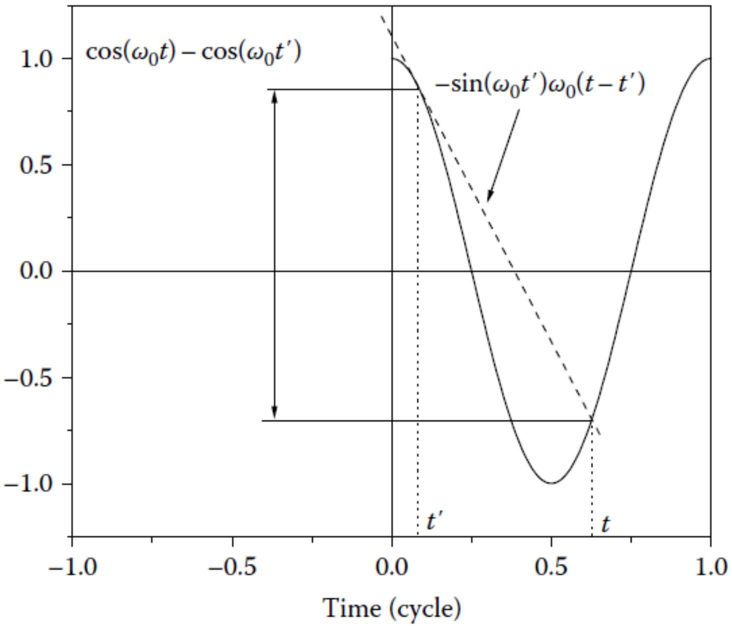
$$x(t) = \frac{eE_L}{m_e\omega_0^2}\{[\cos(\omega_0 t) - \cos(\omega_0 t')] + \omega_0\sin(\omega_0 t')(t - t')\},$$

- Ponderomotive energy = cycle averaged energy of a free electron in the field

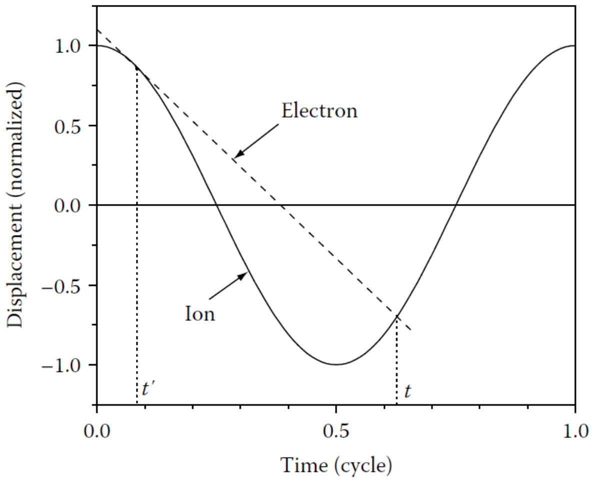
- The recollision time as a function of t' can be determined by the equation x(t)=0

Equivalent form of x(t)=0

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

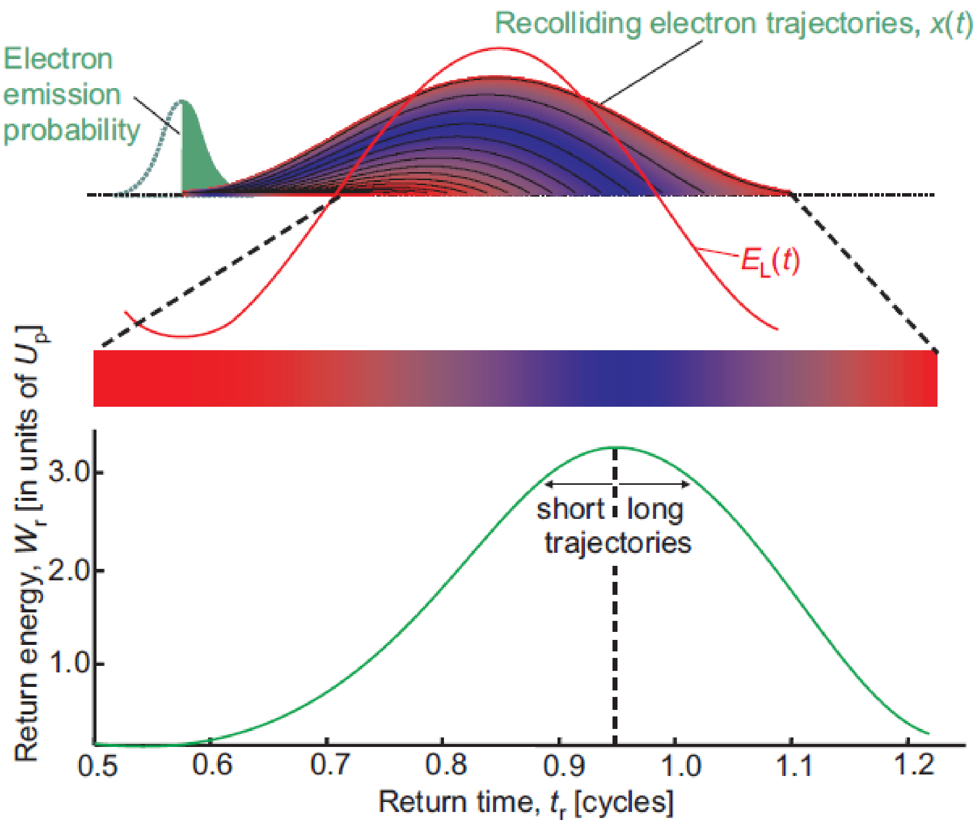



- No analitycal solutions: a graphical solution can be found in an oscillatory frame ($x'=x-x_0\sin(\omega_0 t)$ )



Emitted photon energy:

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




Many possible electron trajectories – a continuum is produced each half cycle

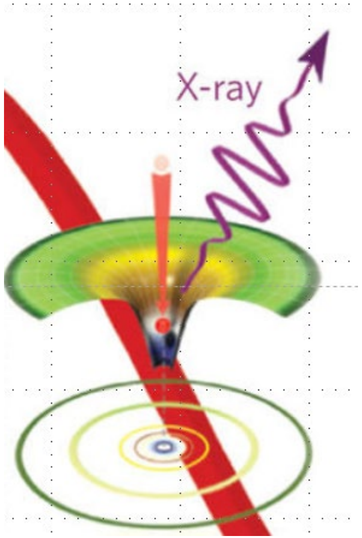
Long and short trajectories, depending on the total path in the field

They have different chirp! Experimentally one can selected the radiation coming form the short trajectories:

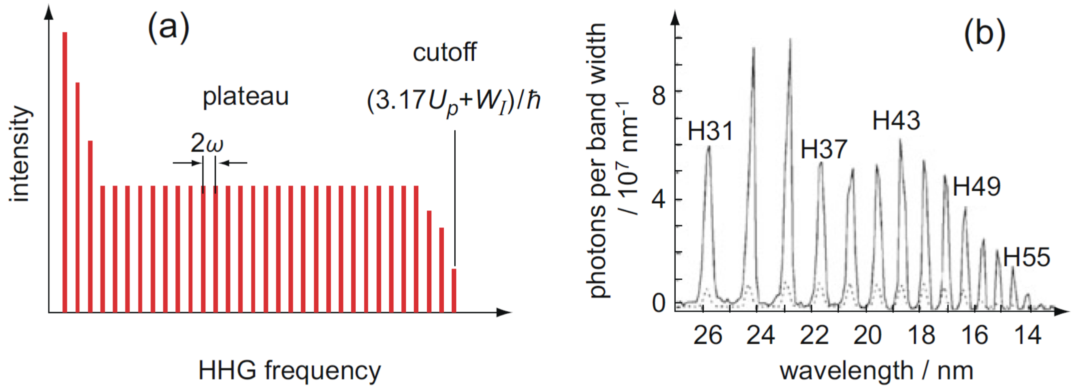
ST Return time = 0.5 to 0.95 Cycles ≈0.5 cycles = 1.3 fs@ 800 nm (FWHM < 1 fs)

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

# STEP 3: recombination




- This determines the efficiency of the process.
- Due to the diffusion of the wavepacket is very unlikely to recombine with the parent atom.
- The probability depends on the atomic species: heavier noble gases have higher probability




- Why do we observe harmonics?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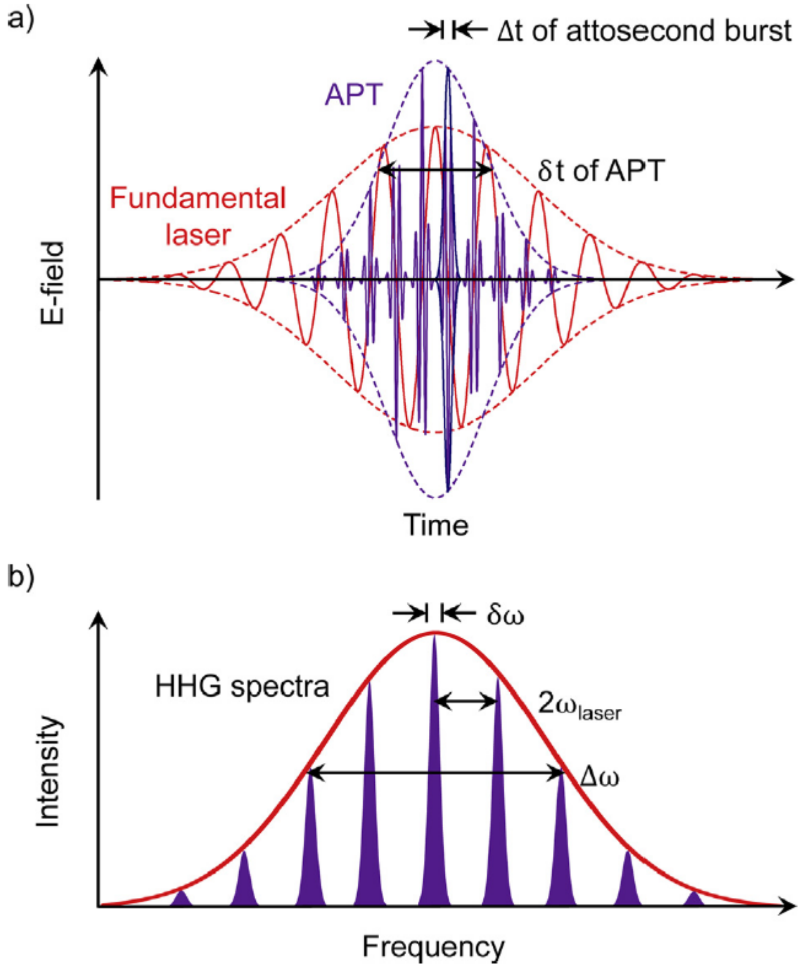
# High order harmonics, attosecond pulse train








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

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

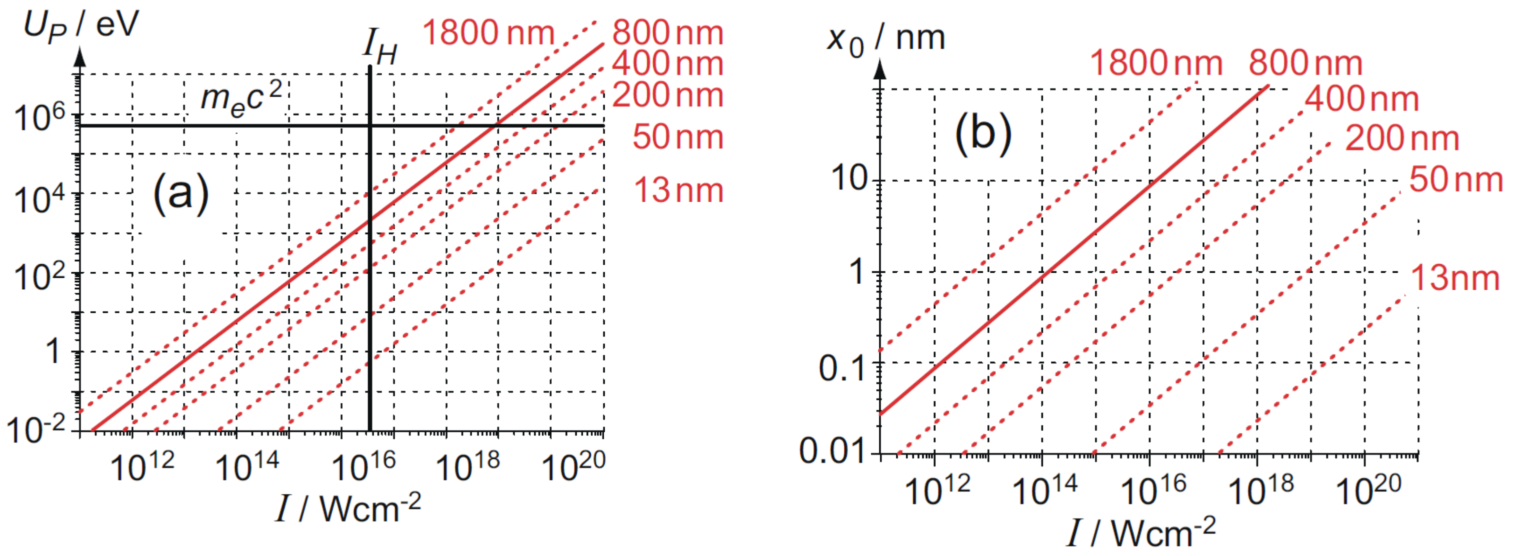
Destructive interference between the continua emitted every half cycle in the inversion symmetric medium

Frequency domain : Frequency comb of odd harmonics.

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

# 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



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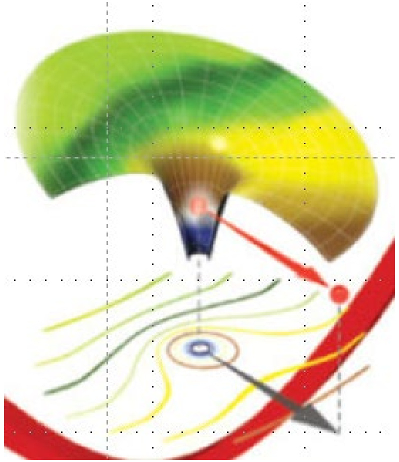


ADK ionization rate (Ammosov Delone Krainov 1986)

- The rate of ionization, as a function of the field strenght w(E) must be calculated with QM
- Several analytical approximations exists, depending on $\gamma_K$ (example ADK theory: $\gamma_K$<<1 Multi-photon processes negligible)

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

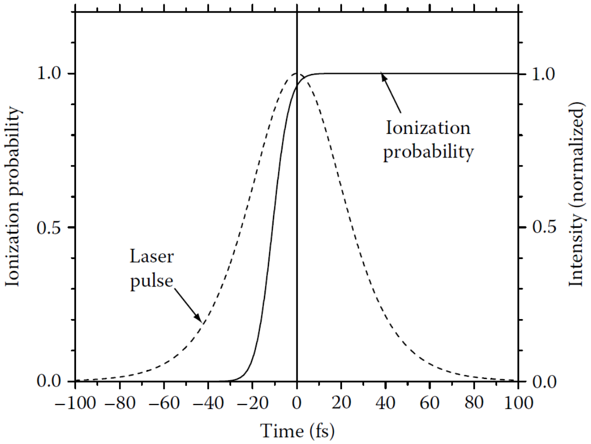
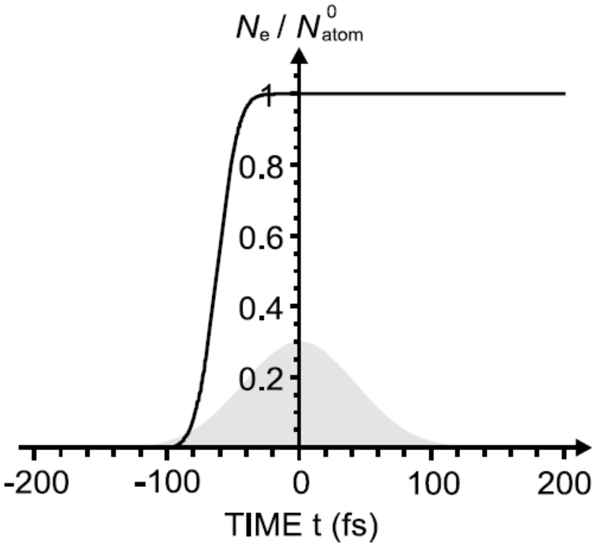


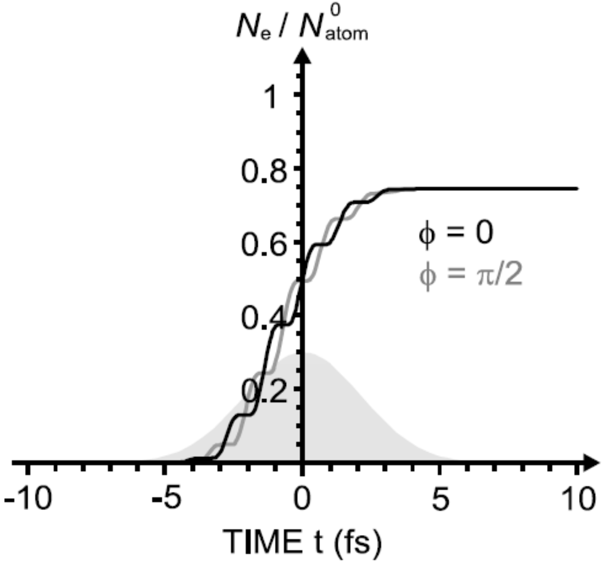

**TABLE 4.2** The ADK Parameters

| | $F_0$ (a.u.) | $n^*$ | $l^*$ | $l$ | $m$ | $\left\|C_{n^* l^*}\right\|^2$ | $G_{lm}$ |
|---|---|---|---|---|---|---|---|
| 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 |

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

# Ionization fraction:




- 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

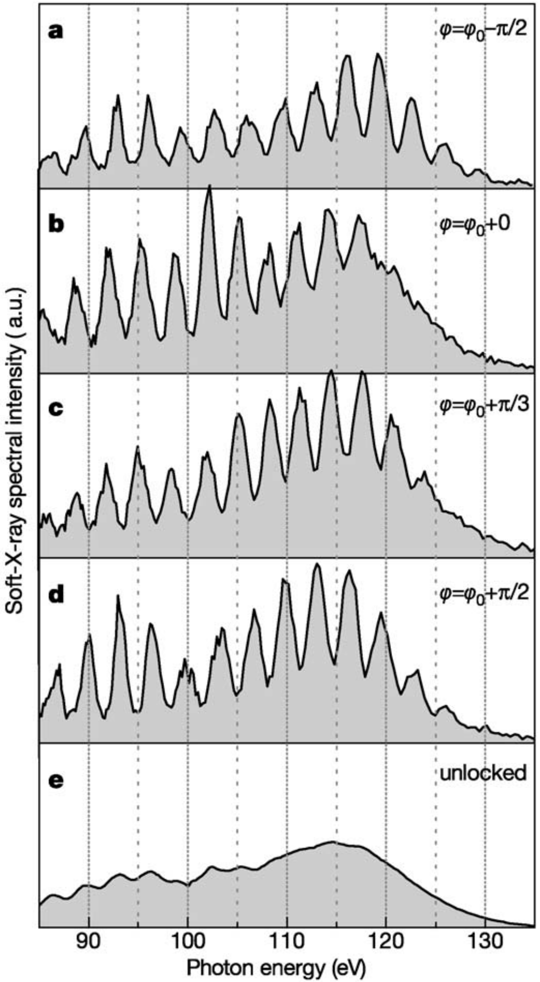
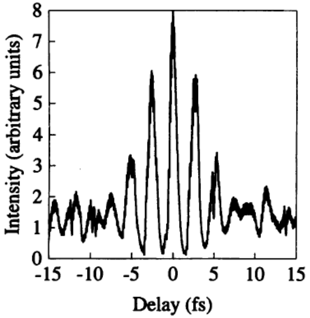


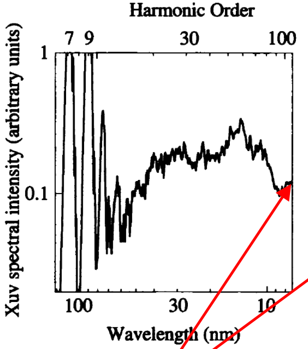

**Fig. 8.4.** Measured EUV spectra from neon at 16 000 Pa (160 mbar) pressure. Excitation is with 5-fs pulses around $\hbar\omega_0 = 1.5\,eV$ and at an estimated intensity of $I = 7 \times 10^{14}\ \mathrm{W/cm^2}$.

# Extending the HHG cutoff



- Increase the field by shortening the pulse




Coherent EUV in the «water window»

C k-edge (280 eV)