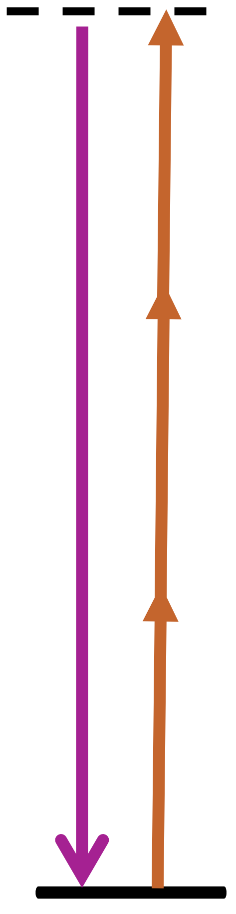
High-order harmonic generation and attosecond pulse production

Anne L'Huillier
Lund University

An historical perspective

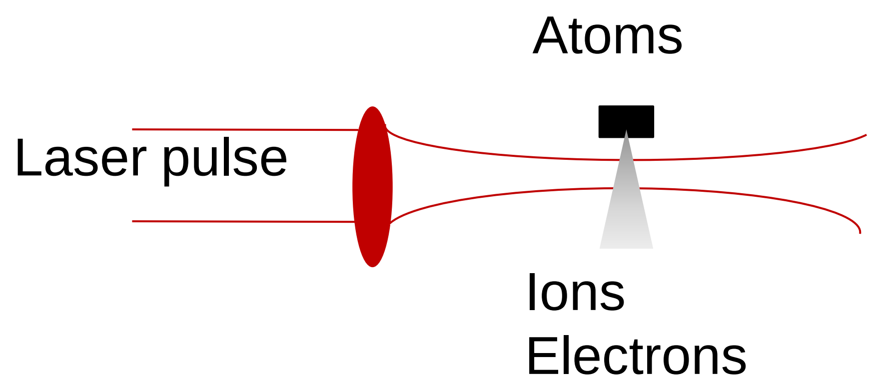
Nonlinear Optics



To get shorter wavelengths, one should start with short wavelengths (UV).

In order to use low-order processes for the frequency upconversion.

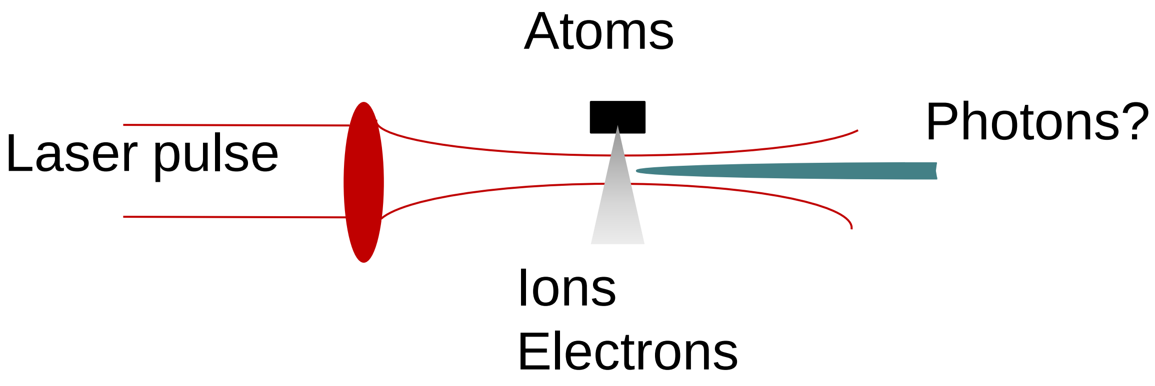
Strong-Field Atomic Physics



- **Picosecond pulses**
- **High intensity**
  10^{14} W/cm^2

Ionization of atoms in strong laser fields
Above-threshold-ionization, multiple ionization

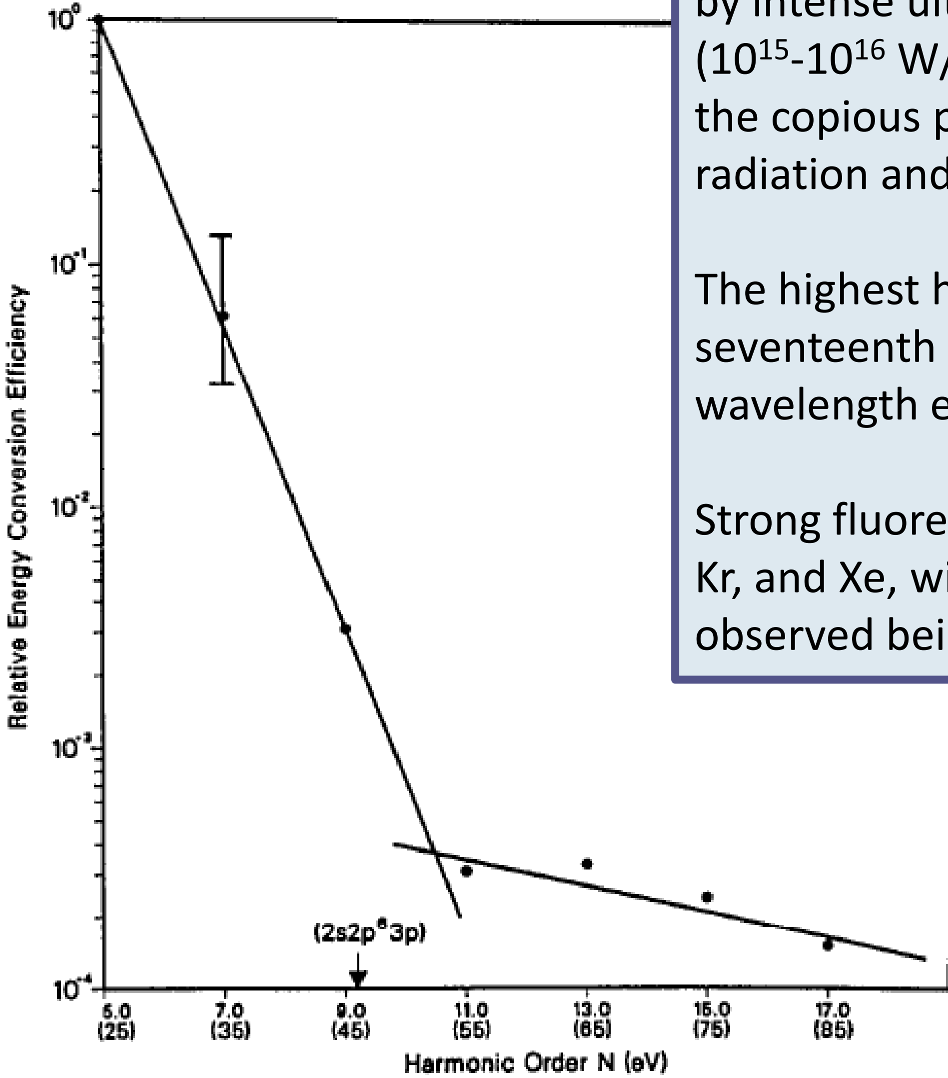
Strong-Field Atomic Physics



The beginning. . .

1987-1988

Fluorescence and high harmonics



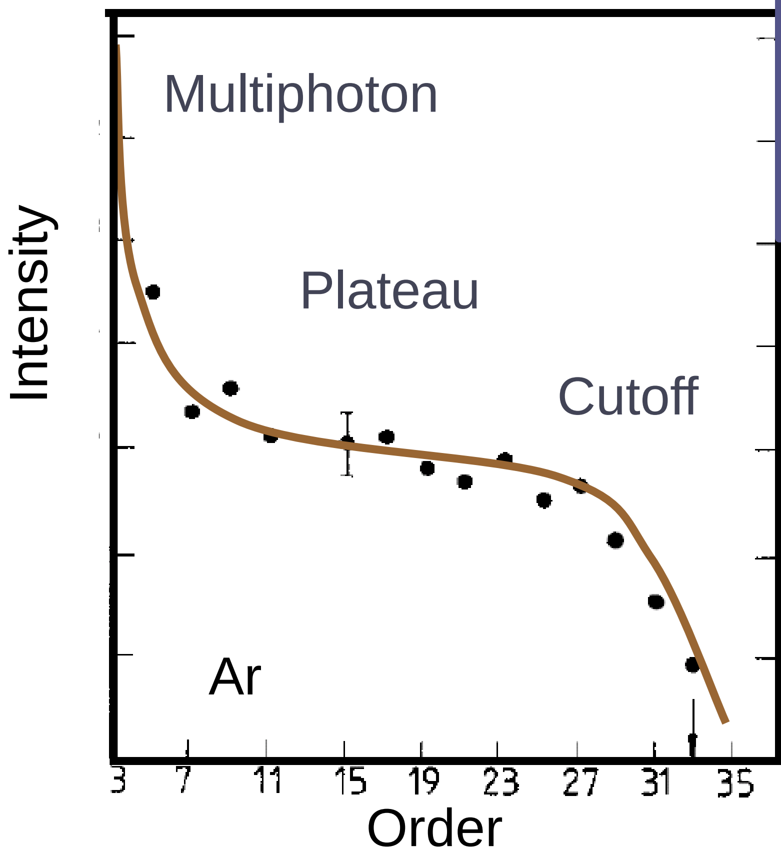
Measurements of (<80-nm) radiation produced by intense ultraviolet (248-nm) irradiation (10^{15}-10^{16} W/cm^2) of rare gases have revealed the copious presence of both harmonic radiation and fluorescence from excited levels.

The highest harmonic observed was the seventeenth (14.6 nm) in Ne, the shortest wavelength ever produced by that means.

Strong fluorescence was seen from ions of Ar, Kr, and Xe, with the shortest wavelengths observed being below 12 nm

McPherson, JOSA B 1987

High harmonic plateau



We report the observation of very-high-order odd harmonics of Nd: YAG laser radiation in rare gases at an intensity of about 10^{13} W cm^{-2}.

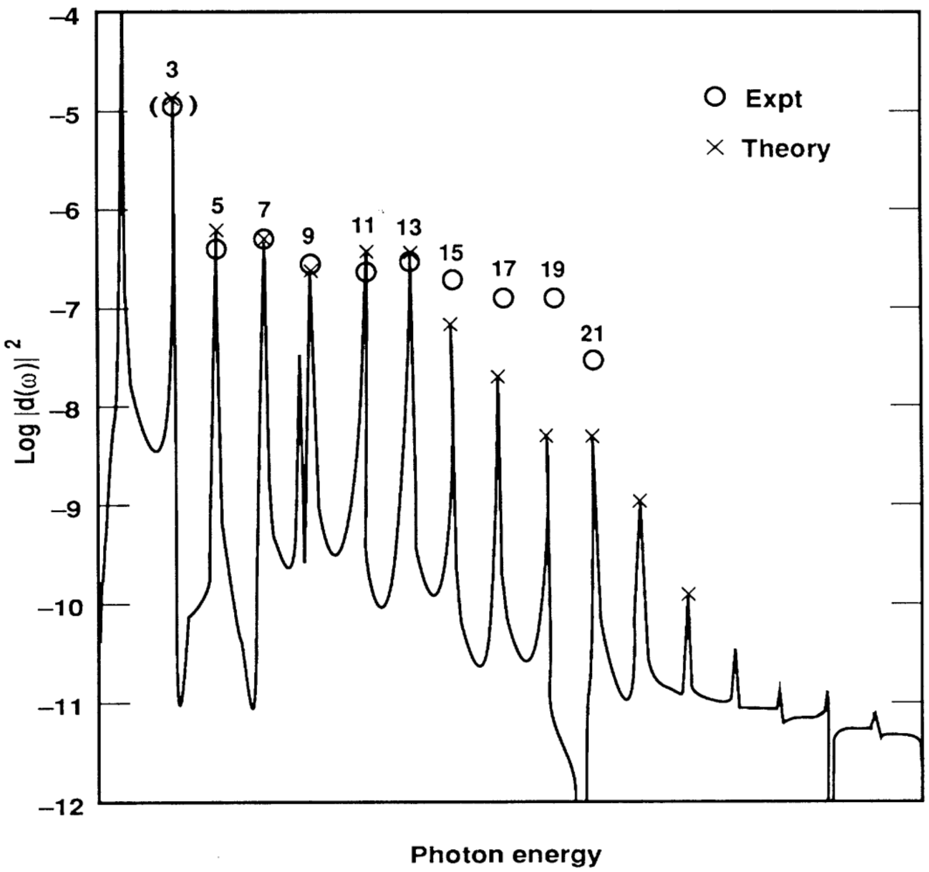
The key point is that the harmonic intensity falls slowly beyond the fifth harmonic as the order increases.

Nd-YAG 1 μm

30 ps, 10 Hz

Ferray, J. Phys B 1988

First numerical simulations



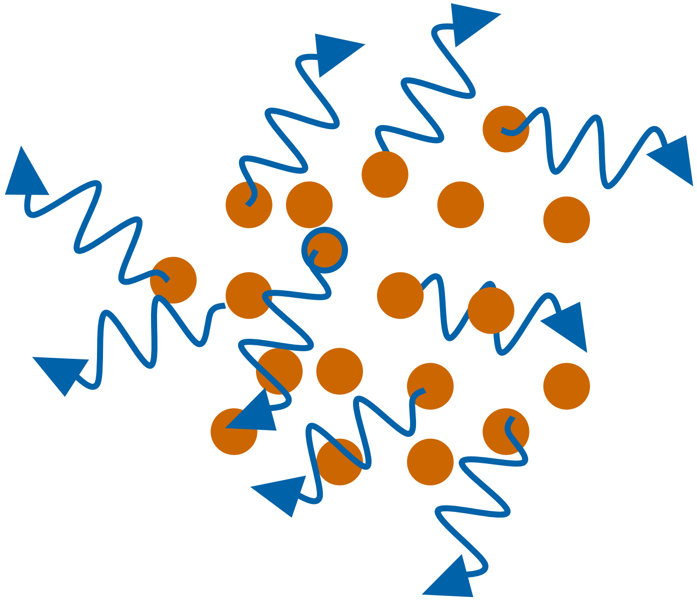
$$i\hbar \frac{\partial \Psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla^2 \Psi + \left[V(r) + e\vec{E}(t).\vec{r}\right]\Psi$$

$$d(t) = < \Psi(t) | ex | \Psi(t) >$$

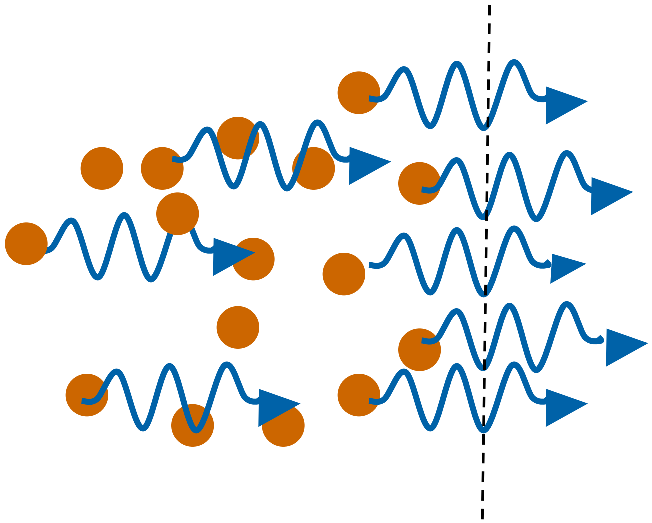
Time-dependent Schrödinger equation

K. C. Kulander and B. W. Shore, PRL, 1989

Phase matching ?



NOT: Incoherent radiation
from a collection of atoms

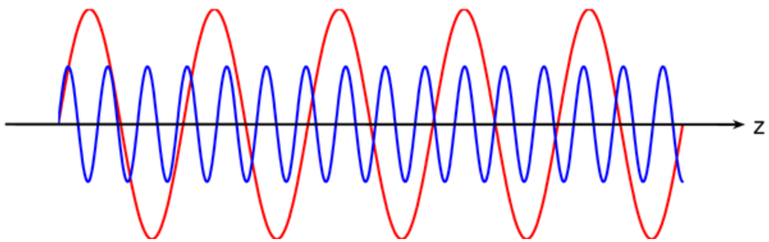


A nonlinear optical phenomenon

$$P(\vec{r},t) = N(\vec{r},t)d(\vec{r},t)$$

$$S = \left| \int_V P(\vec{r},t) e^{-i\Delta\vec{k}.\vec{r}} d\vec{r} \right|^2$$

$$\Delta k = k_q - qk_1$$



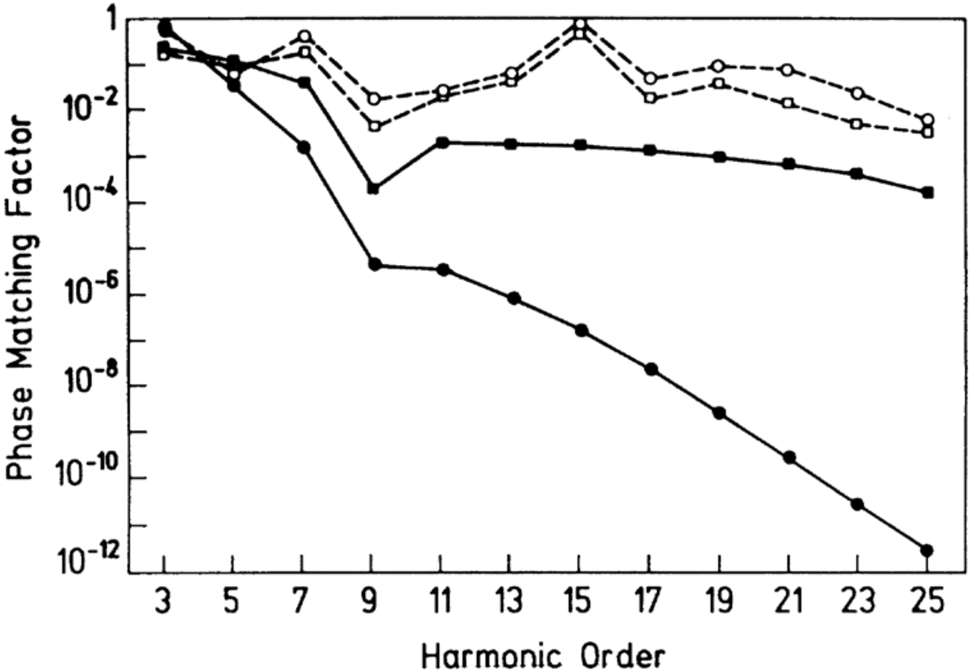
- o Dispersion
- o Gouy phase due to focusing

First propagation calculations

Role of phase matching ?

L'Huillier, Kulander and Schafer, PRL 1991

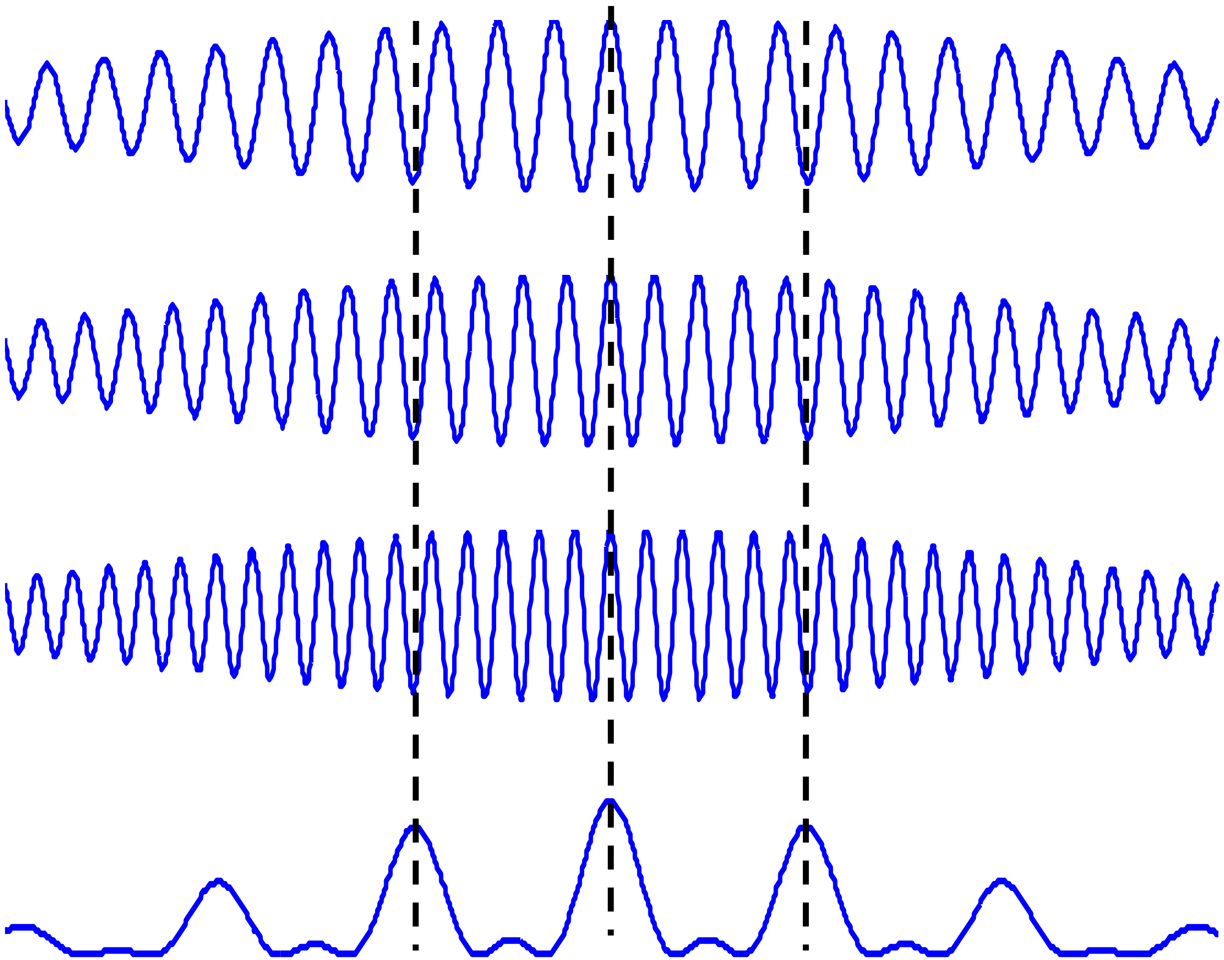
$$S = \left| \int_V P(\vec{r},t) e^{-i\Delta\vec{k}.\vec{r}} d\vec{r} \right|^2$$

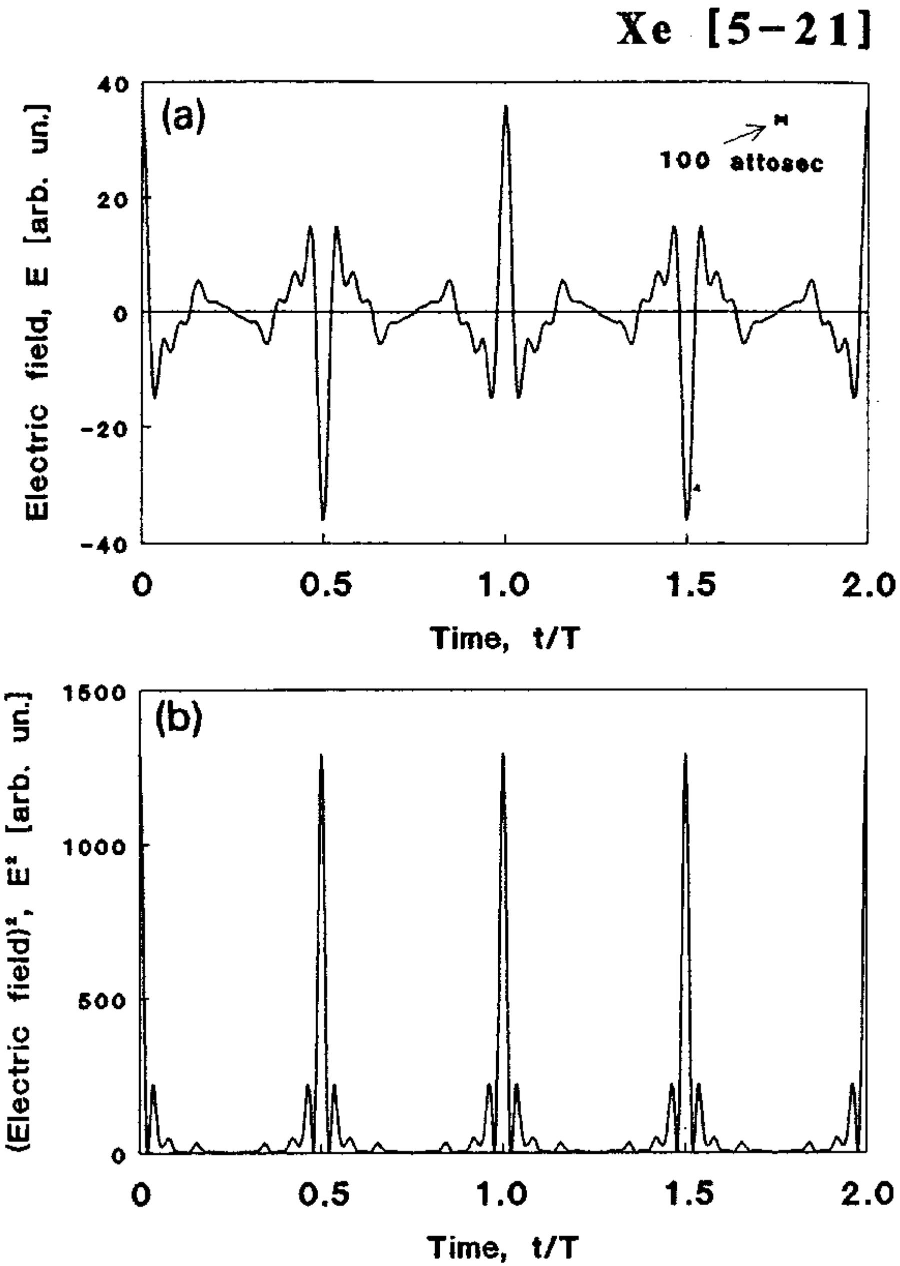


$$I_q = | d(q\omega) |^2 \ F_q$$

Attoseconds?

Attosecond pulses = Sum of phase-locked harmonics



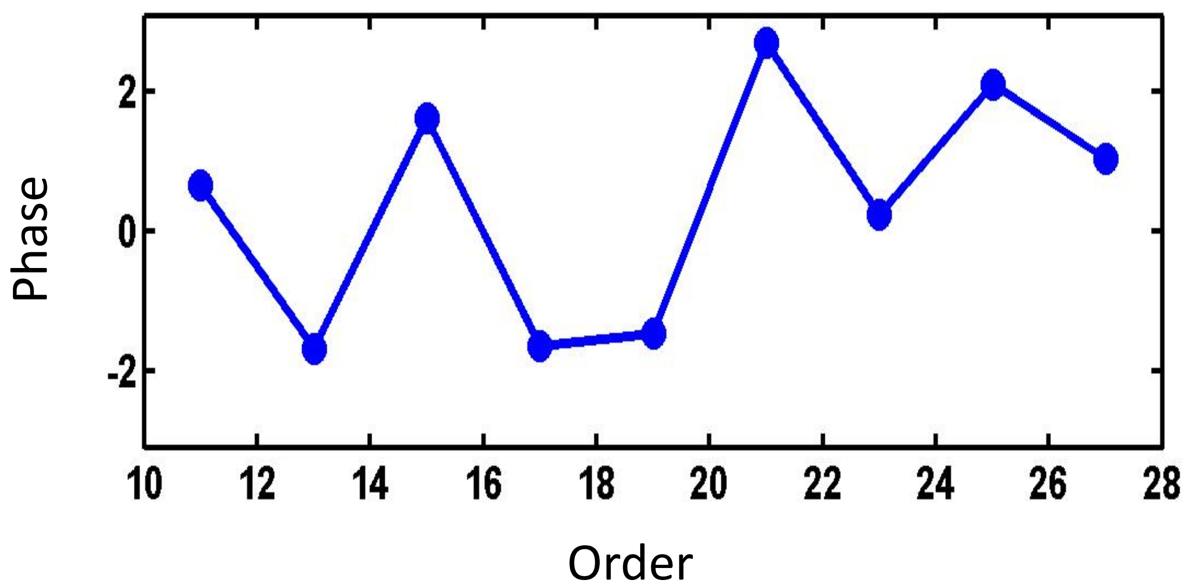


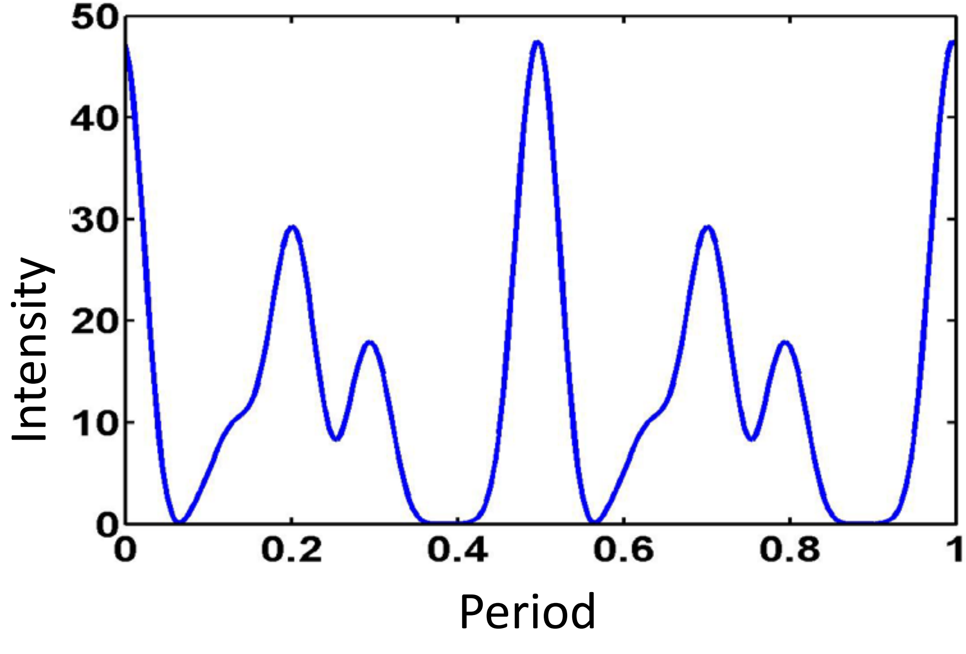
"If the harmonics are appropriately phased, this bandwidth corresponds to temporal pulses on the order of 5×10^{-17} s, and thereby motivates a search for a new regime of short-pulse generation."

Harris, Hänsch, Maclin, OC, 1993

Toth and Farkas, PL, 1992

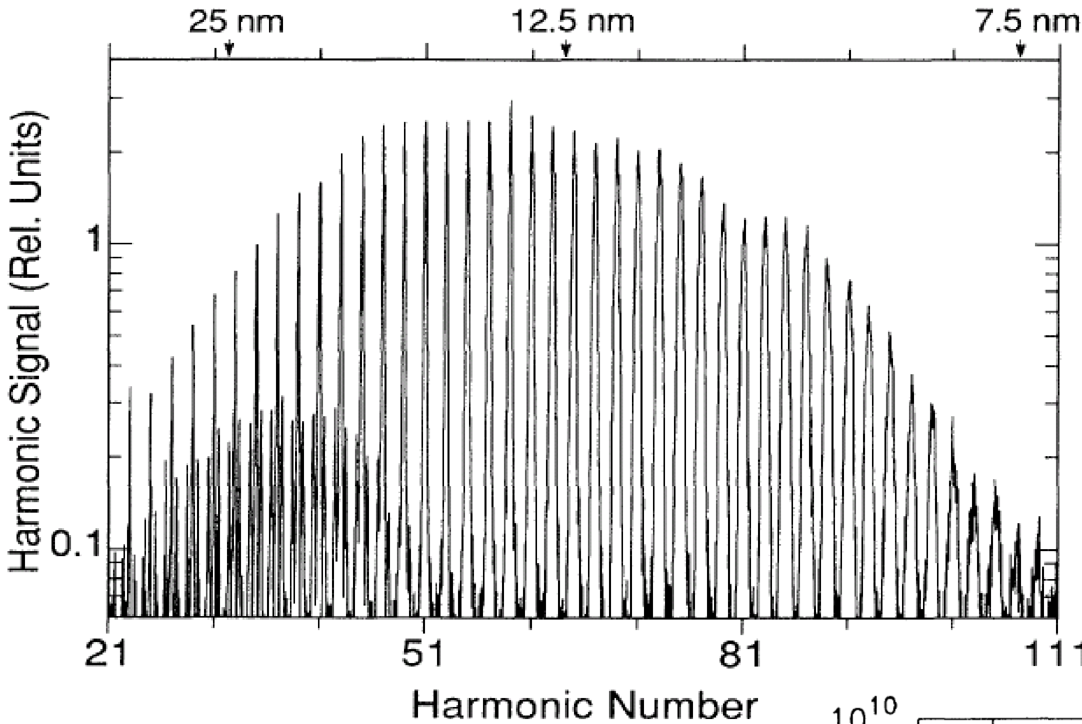
Attosecond pulses ?





HHG with short pulses

1993

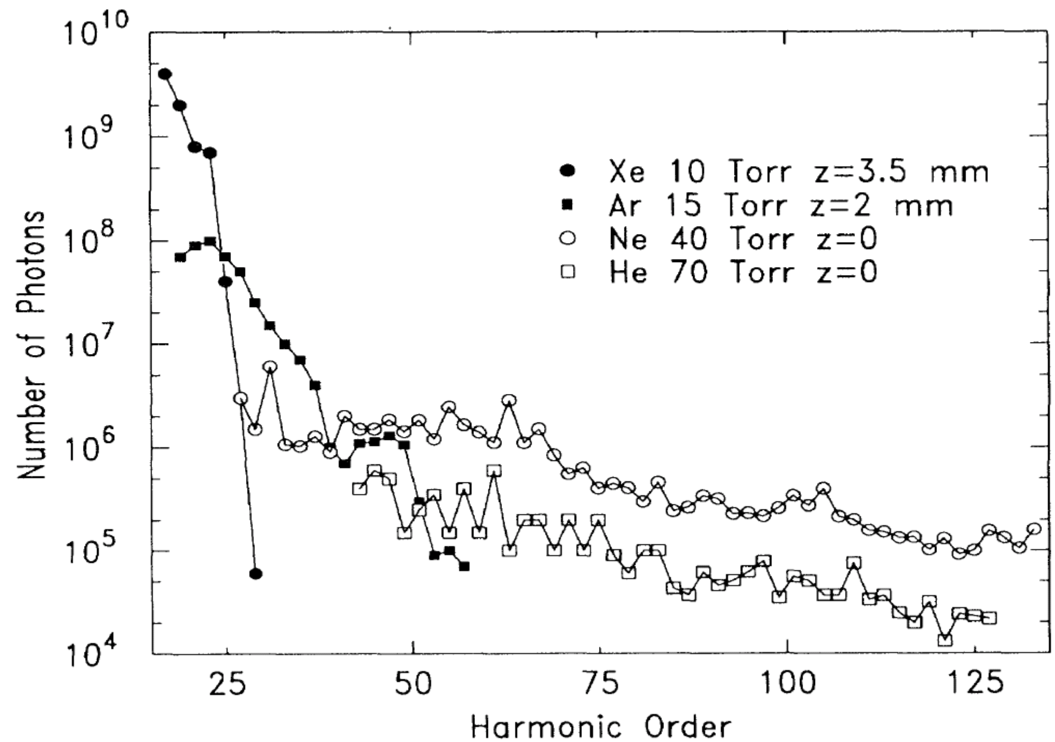


With short laser pulses Using the CPA technique

100 fs, Ti:Sa

Maclin, Harris PRL 1993

1 ps, Nd:Glass

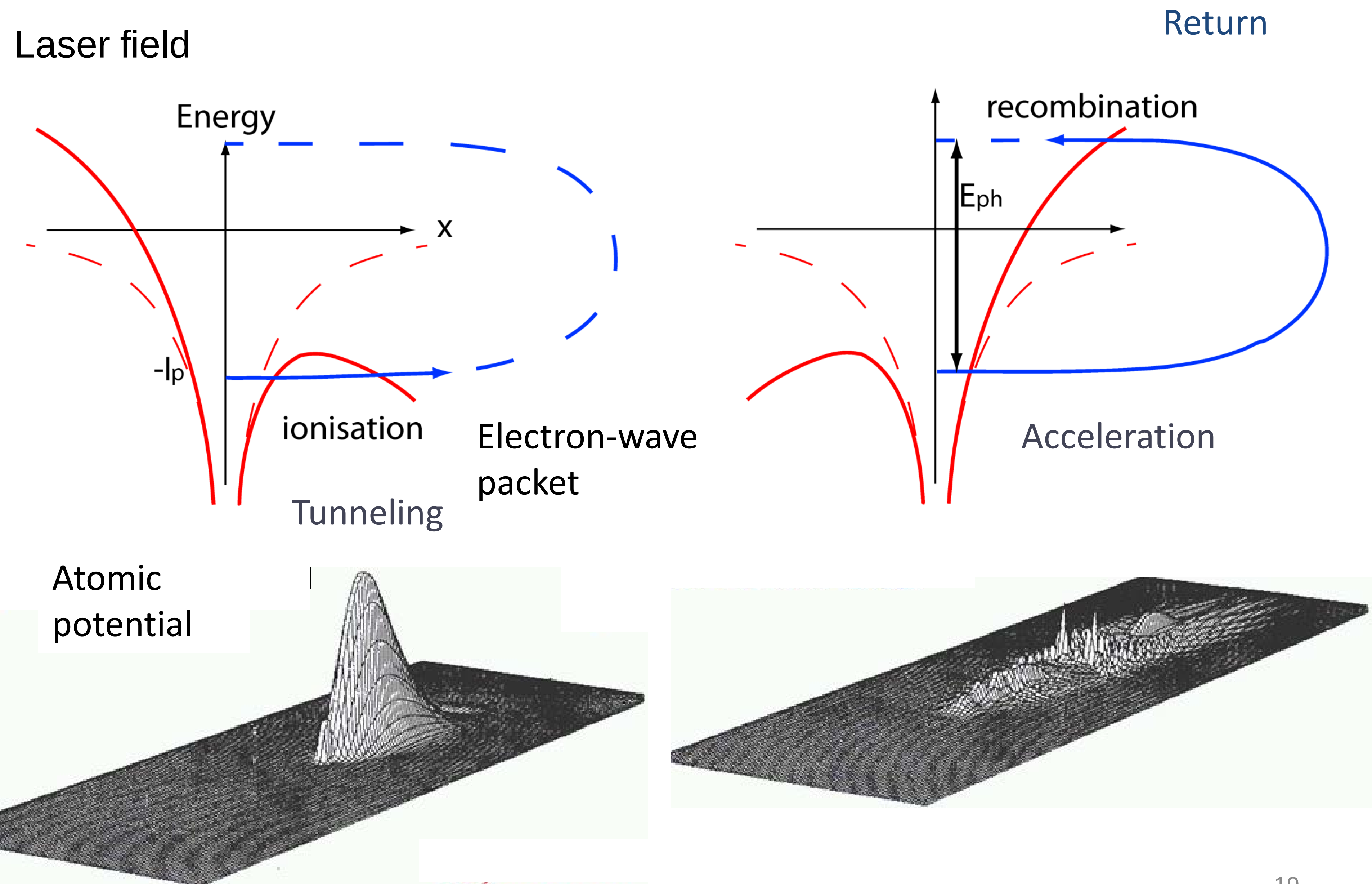


L'Huillier, Balcou PRL 1993

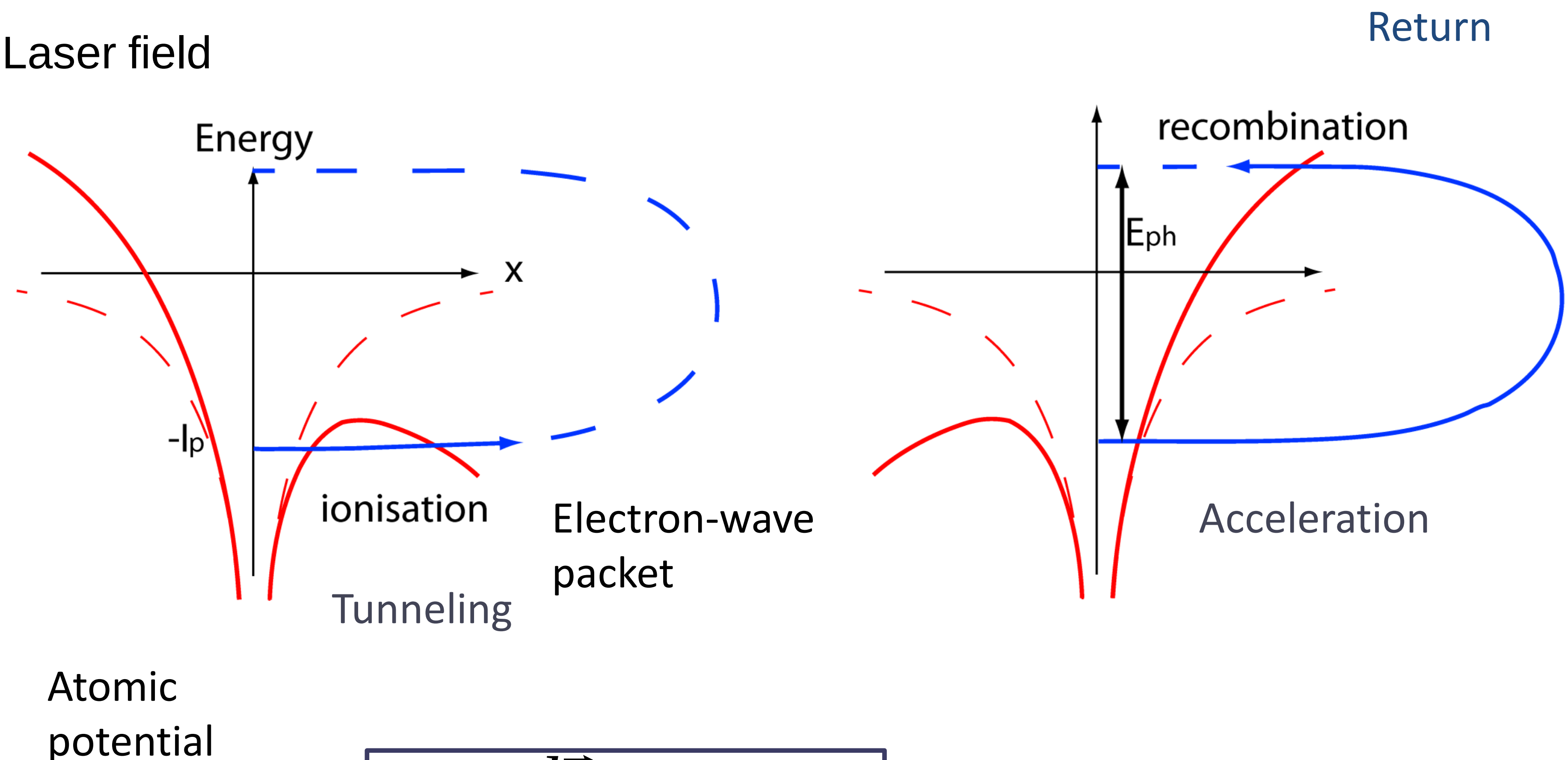
The breakthrough in understanding

1993

Quasi-classical interpretation



Quasi-classical interpretation



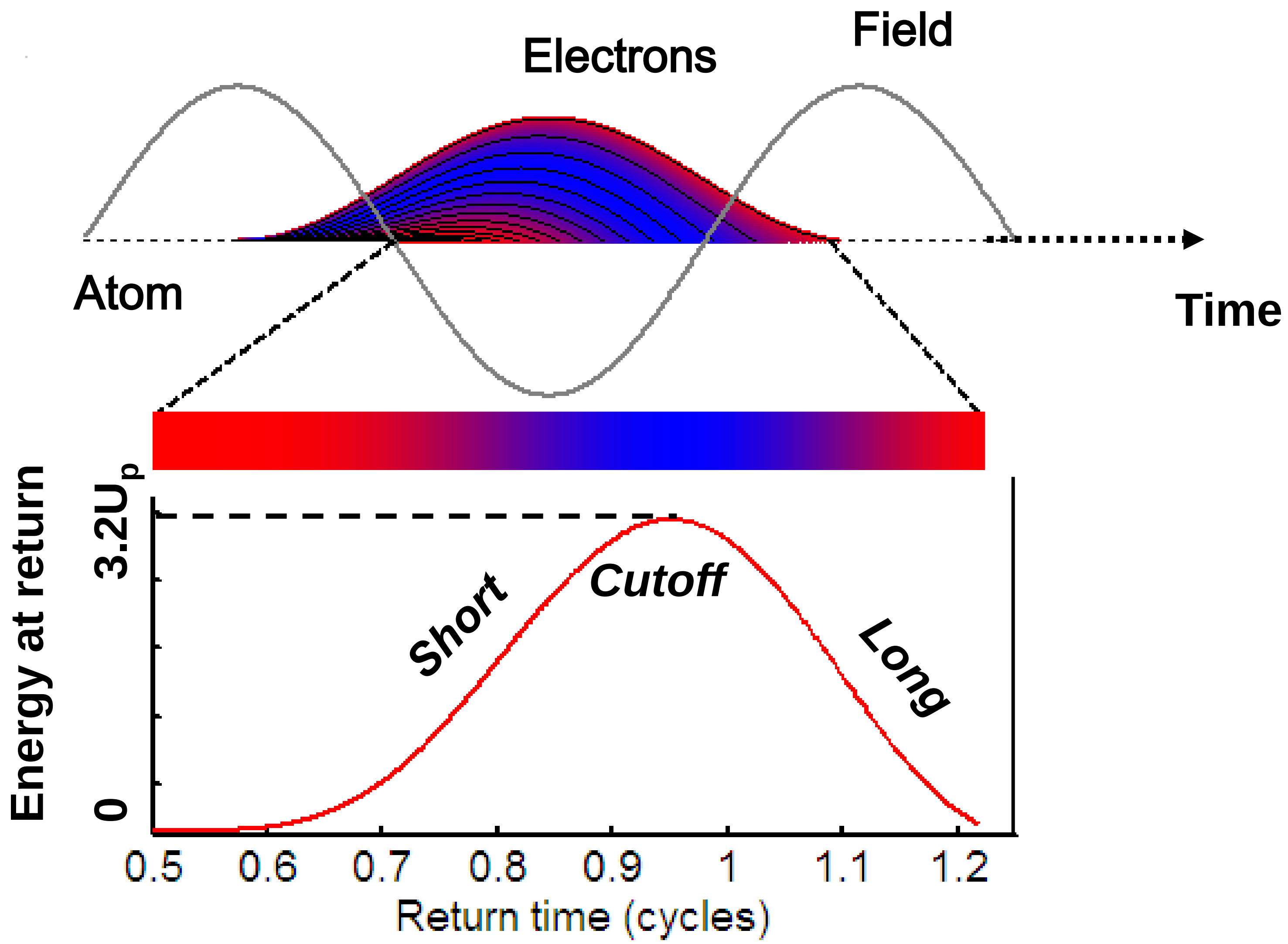
$$m\frac{d\vec{v}}{dt} = -e\vec{E}$$

$$E_{ph} = \tfrac{1}{2}mv^2 + I_p$$

P. Corkum, K. Kulander, presented at Han-sur-Lesse, Belgium, February 1993

P. Corkum, PRL, 1993

Electron trajectories



$$E_{ph} = \frac{1}{2} m v^2 + I_p$$

Strong field approximation

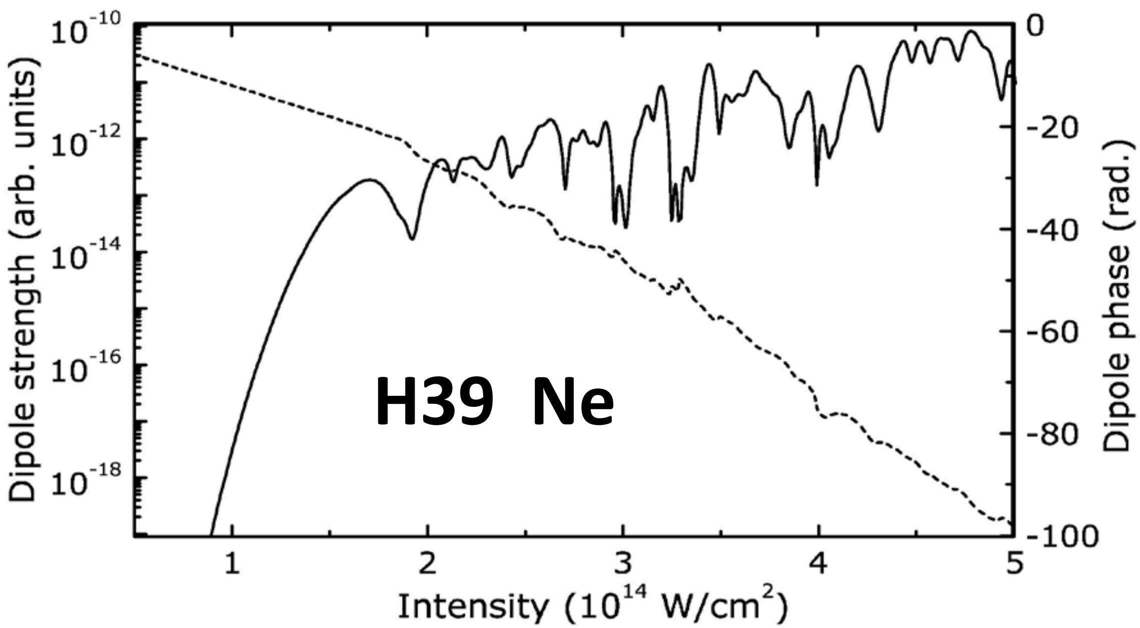
$$ex(t) = i\int_{-\infty}^{t} d\tau \int d\vec{p}\, d^*(\vec{p}+e\vec{A}(t))\, e^{-iS(\vec{p},t,\tau)/\hbar}\, E(t-\tau)\, d(\vec{p}+e\vec{A}(t-\tau))$$

Recombination

Acceleration in the continuum

Transition to the continuum

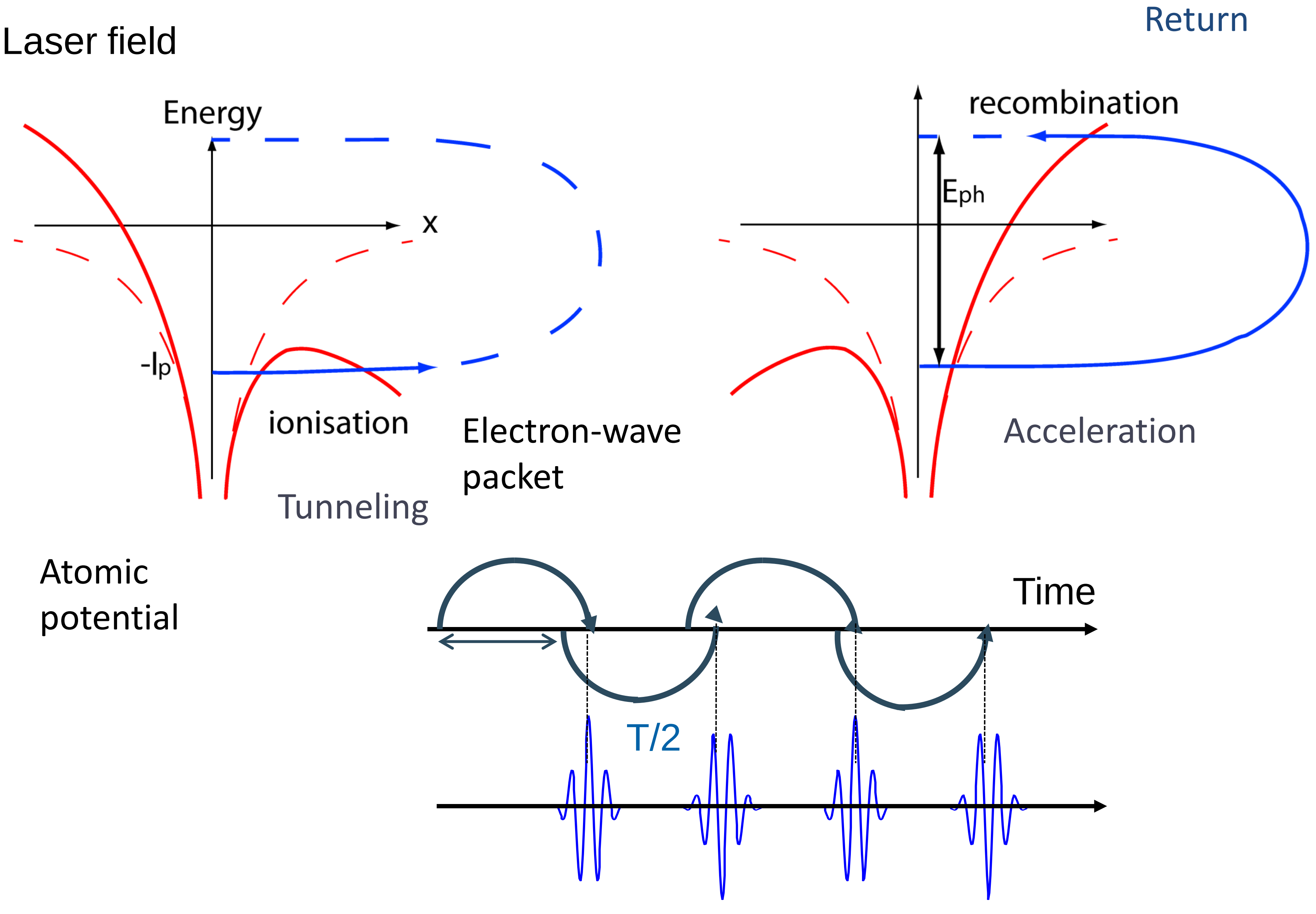
$$S(\vec{p},t,\tau) = \int_{t-\tau}^{t} dt' \frac{(\vec{p}+e\vec{A}(t'))^2}{2m} + I_p$$



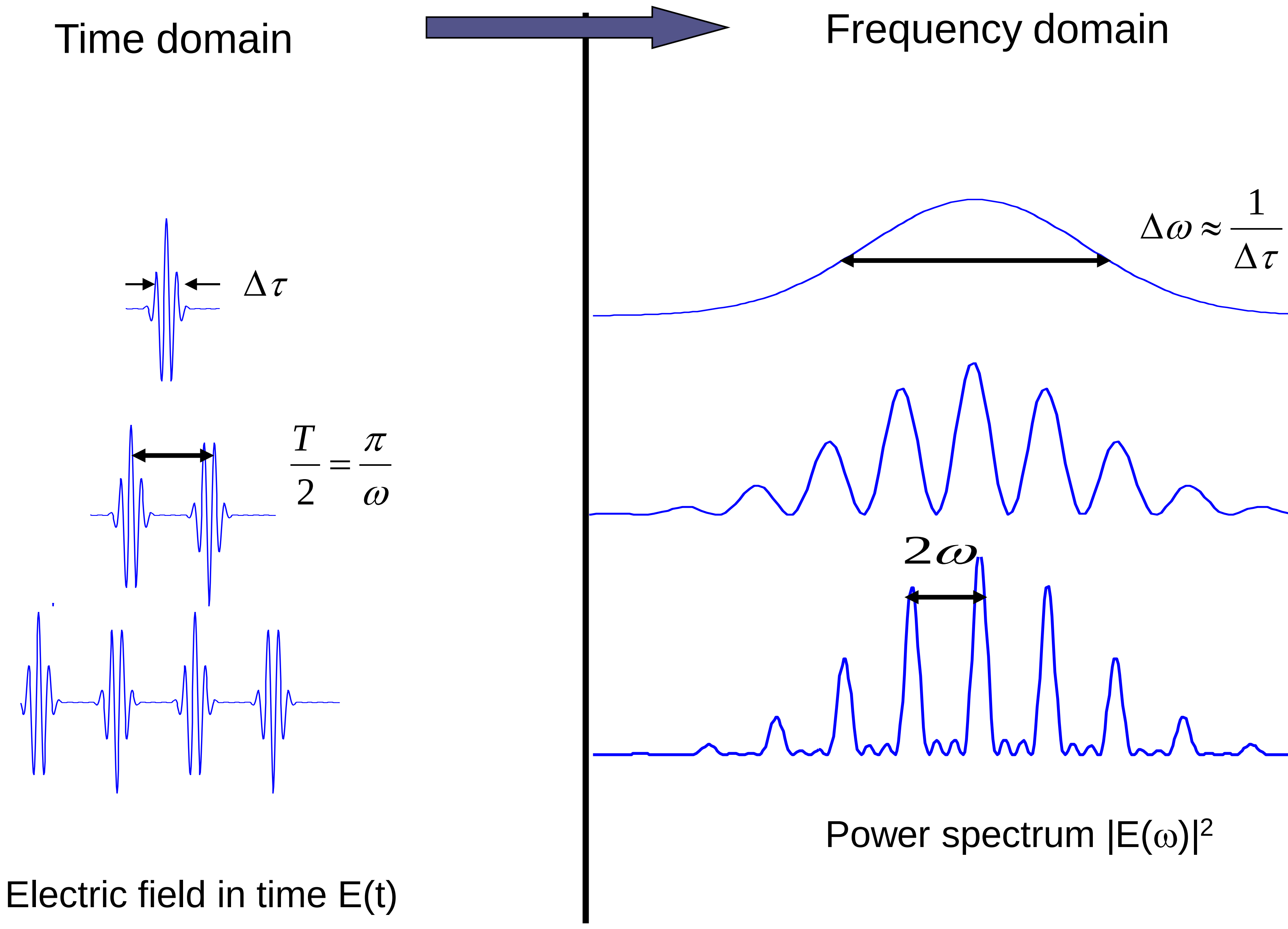
Lewenstein, PRA, 1994

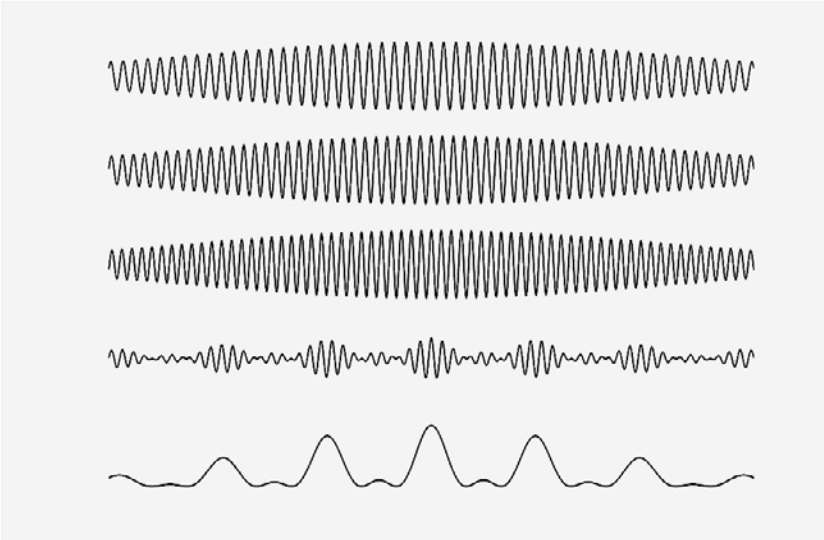
Attoseconds?

Quasi-classical interpretation



From the time to the frequency domain

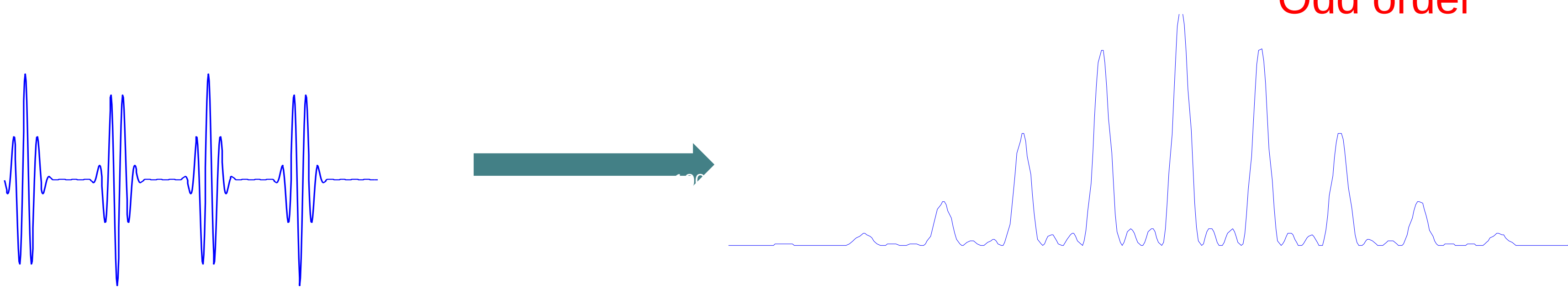




Attosecond pulses = Sum of phase-locked harmonics

Harmonics = Interferences of attosecond pulses

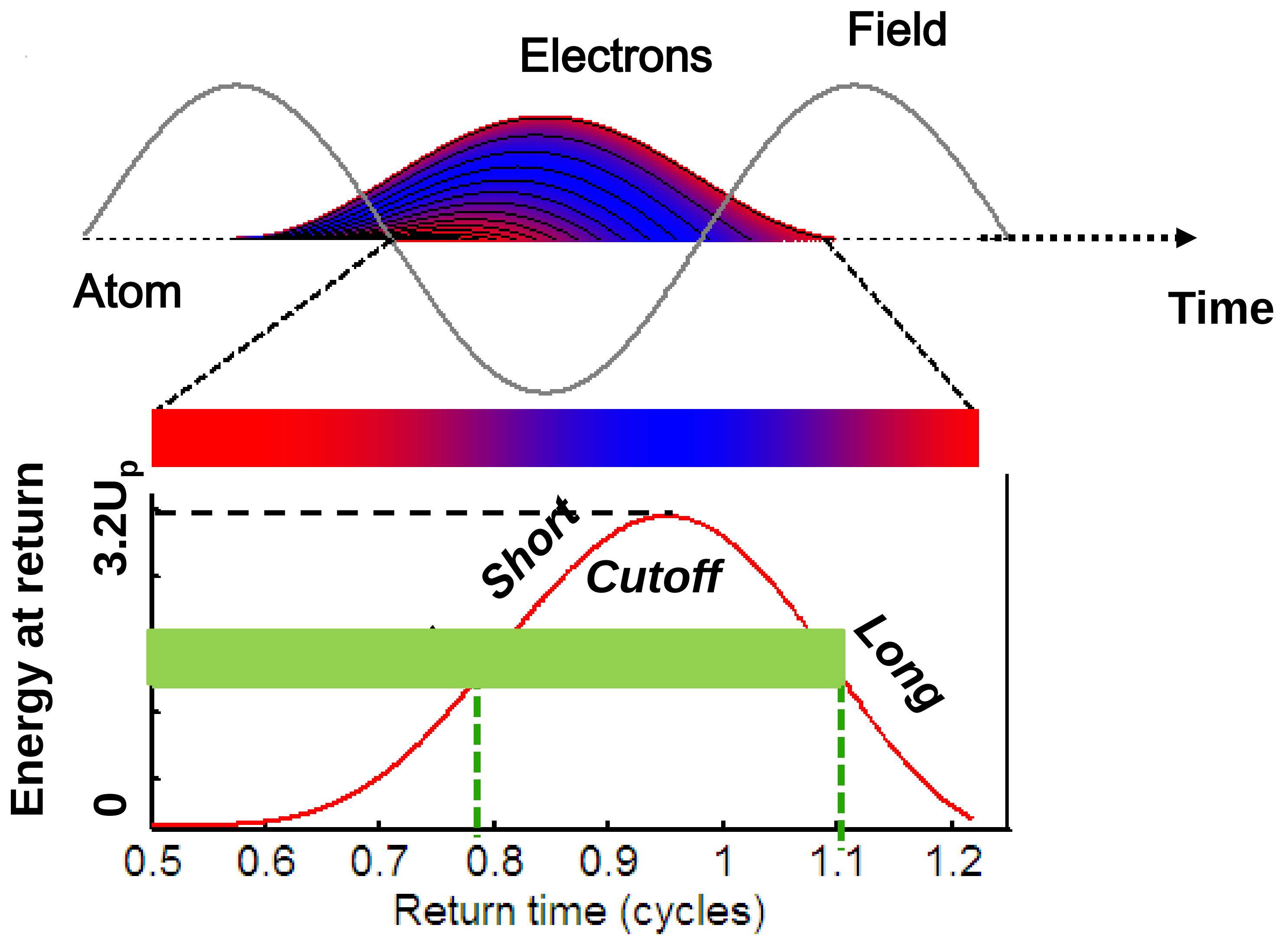
High harmonics
Odd order



Electric field in time E(t)

Power spectrum $|E(\omega)|^2$

Electron trajectories



Multiple pulses per half cycle ?
Chirp

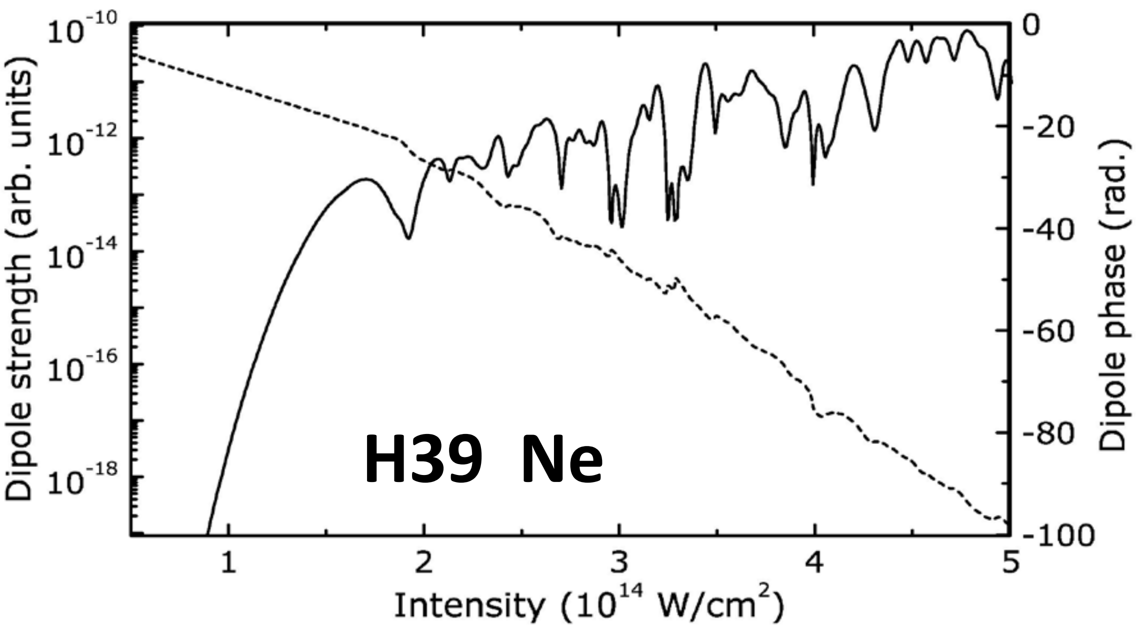
The trajectories

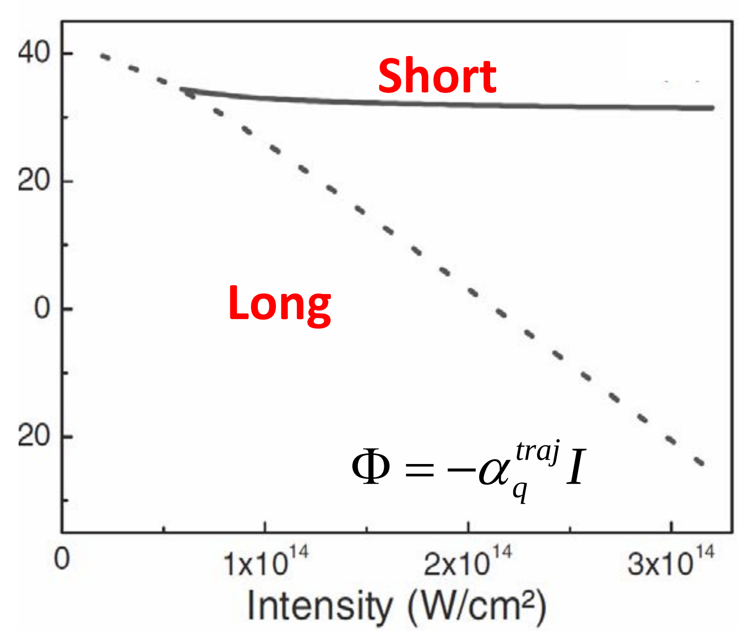
1995-2000

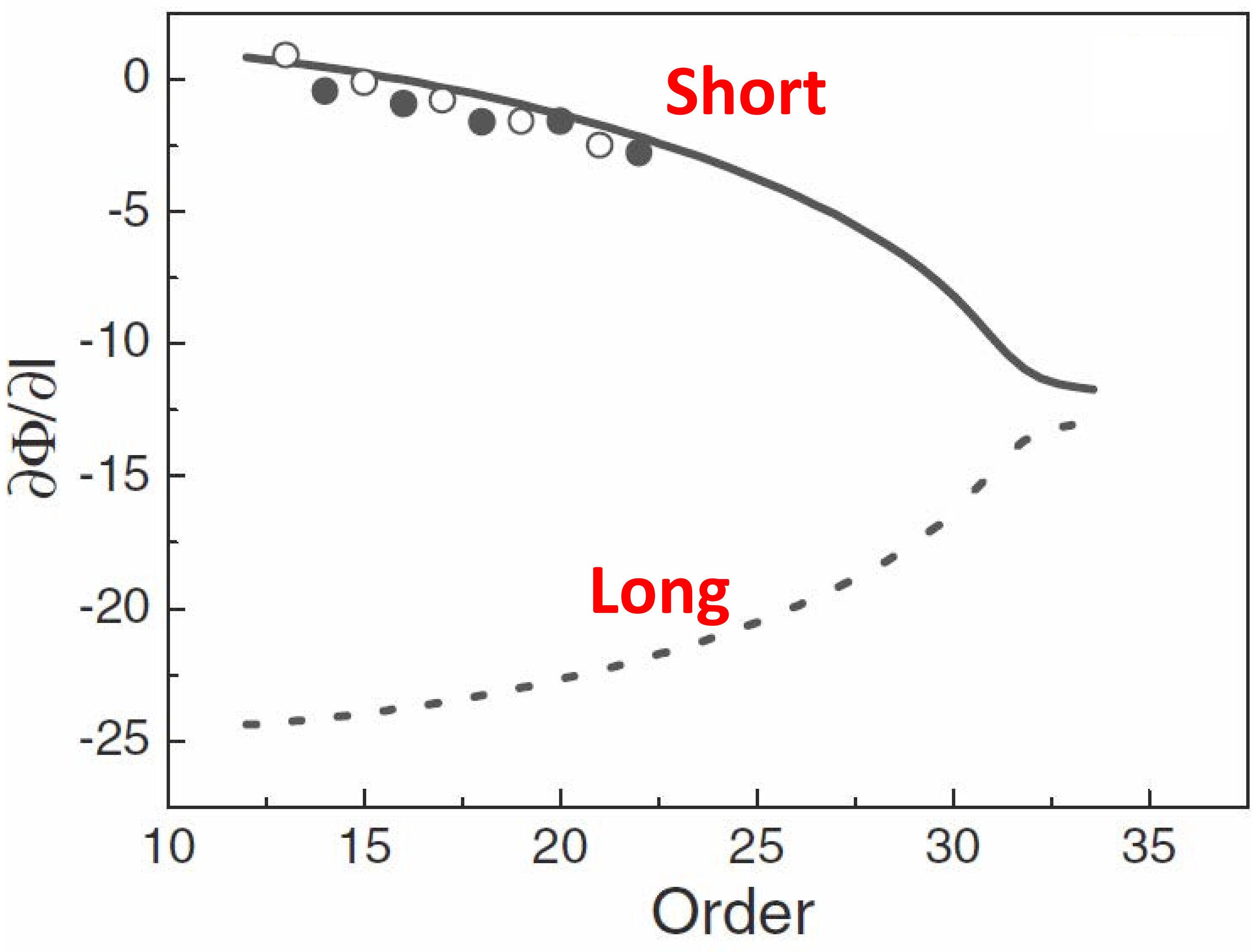
Strong field approximation

$$ex(t) = i\int_{-\infty}^{t} d\tau \int d\vec{p}\, d^{*}(\vec{p}+e\vec{A}(t))e^{-iS(\vec{p},t,\tau)/\hbar}E(t-\tau)d(\vec{p}+e\vec{A}(t-\tau))$$

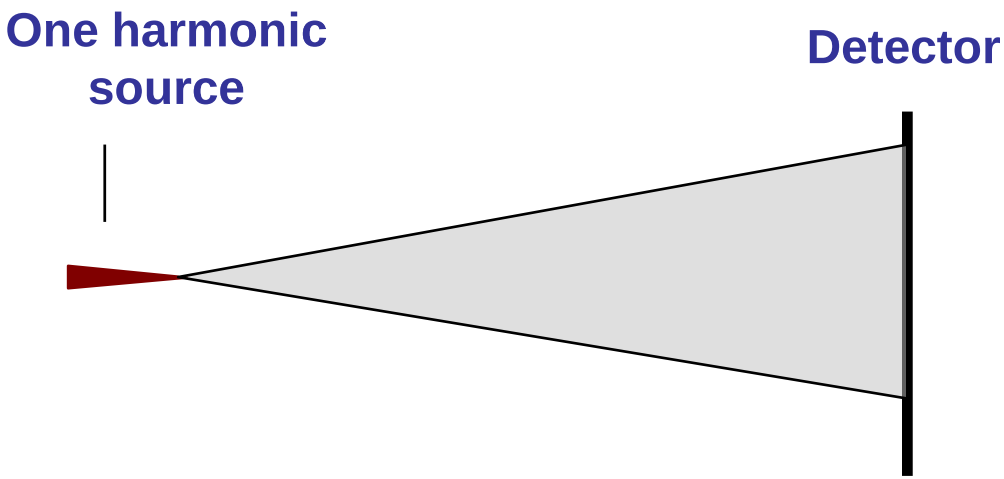
Saddle point equations → Disentangle the trajectories






Short
Long
∂Φ/∂I
0
-5
-10
-15
-20
-25
10
15
20
25
30
35
Order

Electron trajectories



Focus
r-dependent phase-
Wavefront R>0

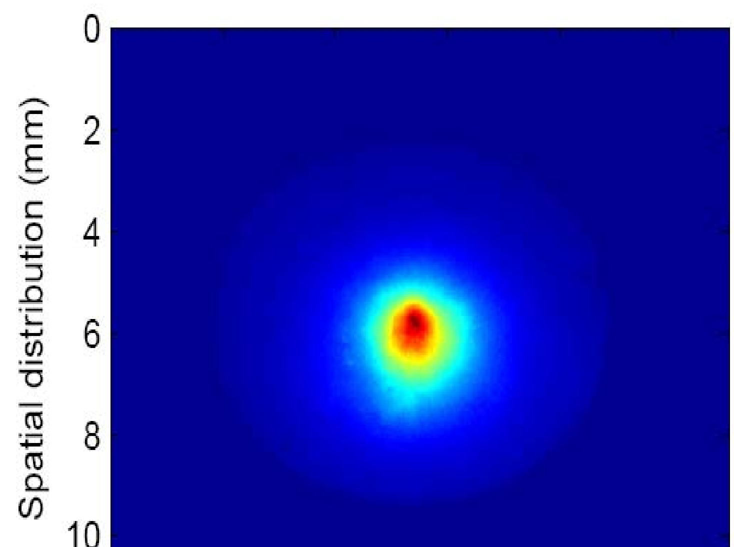
→

Far field
Increased divergence

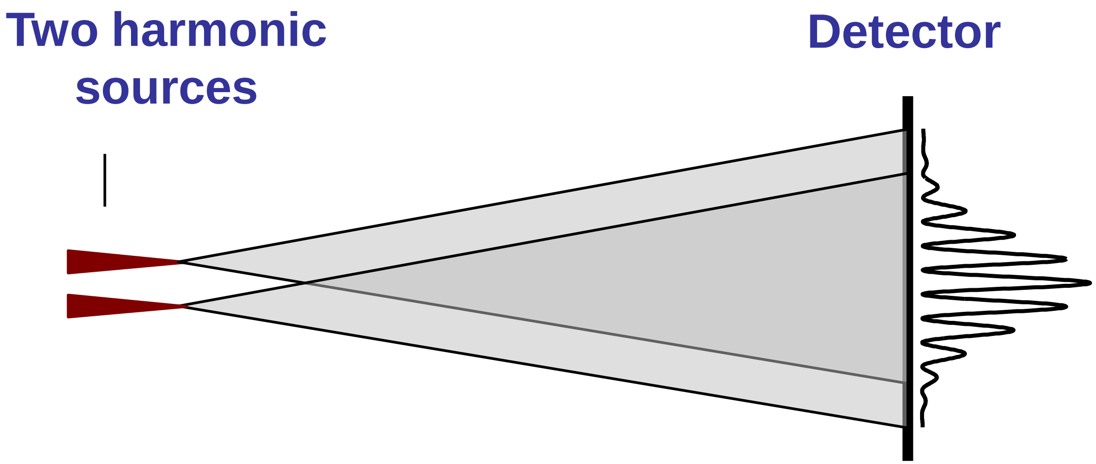
$$\theta = \frac{\lambda_q}{\pi w_q}\sqrt{1 + 4\alpha_q^2 I_0^2 \frac{w_q^4}{w_0^4}}$$

$$E_s = A_s e^{-i\alpha_s I(r)}$$

$$E_\ell = A_\ell e^{-i\alpha_\ell I(r)}$$



Electron trajectories



Long trajectory:

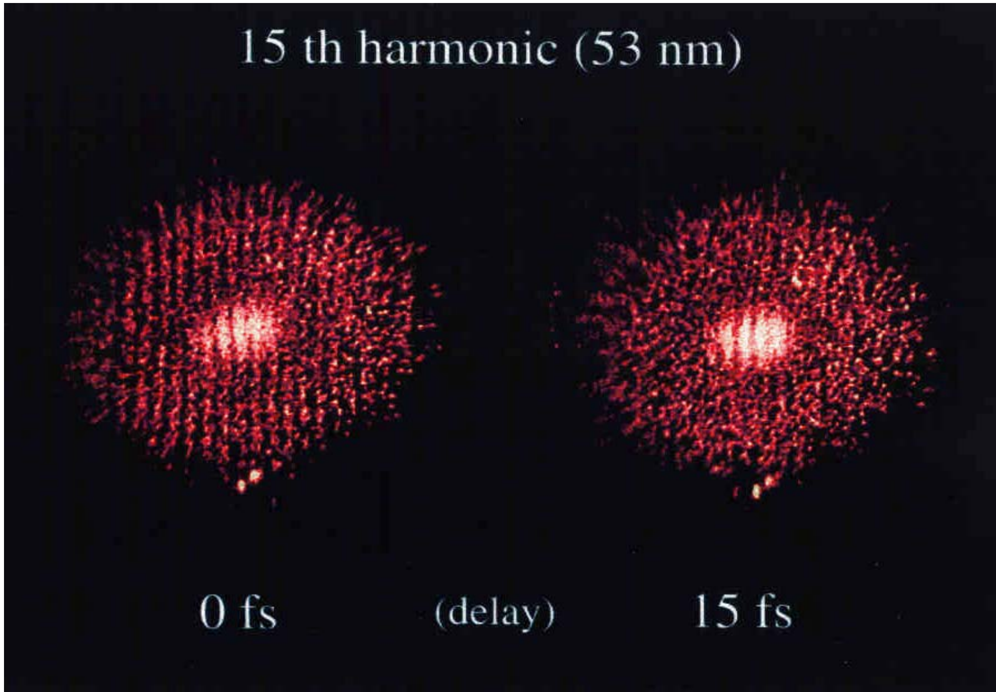
Increased spectral bandwidth
Reduced coherence time
Divergent

Short trajectory:

Spectrally narrow
Long coherence time
Collimated

Zerne et al., PRL 1997

Bellini et al, PRL 1998



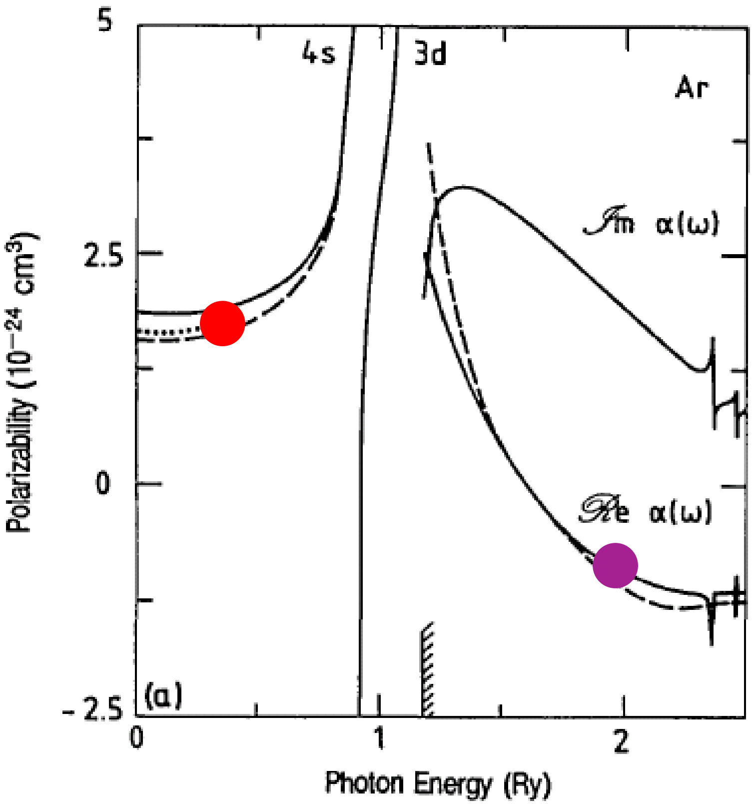
Attoseconds?

Phase matching

$$\Delta k = \underbrace{\Delta k_a}_{<0} + \underbrace{\Delta k_{fe}}_{>0} + \underbrace{\Delta k_{foc}}_{>0} + \underbrace{\Delta k_{dip}^{traj}}_{\text{-sign}(z)}$$

$\propto$ Pressure

$\propto I,\ f(I), t$



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

$$\Delta k_{fe} = \frac{qe^2}{2\varepsilon_0 cm\omega} N_e \qquad \Delta k_{foc} \approx \frac{q}{z_0}$$

$$\Delta k_{traj} = \alpha_{traj} \frac{\partial I}{\partial z} \qquad (\alpha_{traj} > 0)$$

Degree of ionization = a few %

Conventions

$$E(t) = \int d\omega \, A(\omega) \, e^{-i[\omega t - kz - \varphi(\omega)]} + cc \qquad \varphi(\omega) = -\alpha I$$

$$\Delta k = k_q - q k_1$$

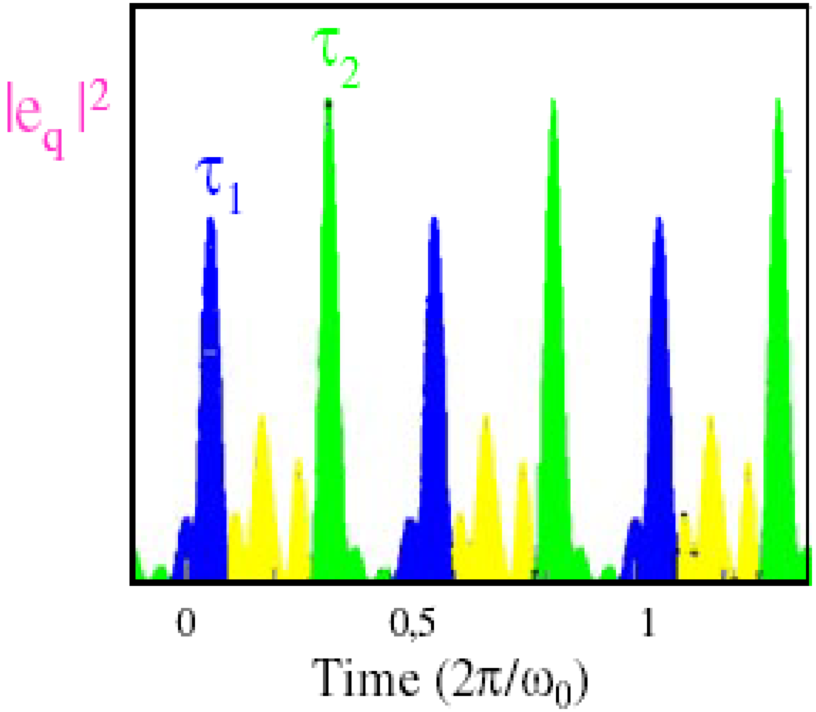
Generated field – Induced polarization @ $q\omega$

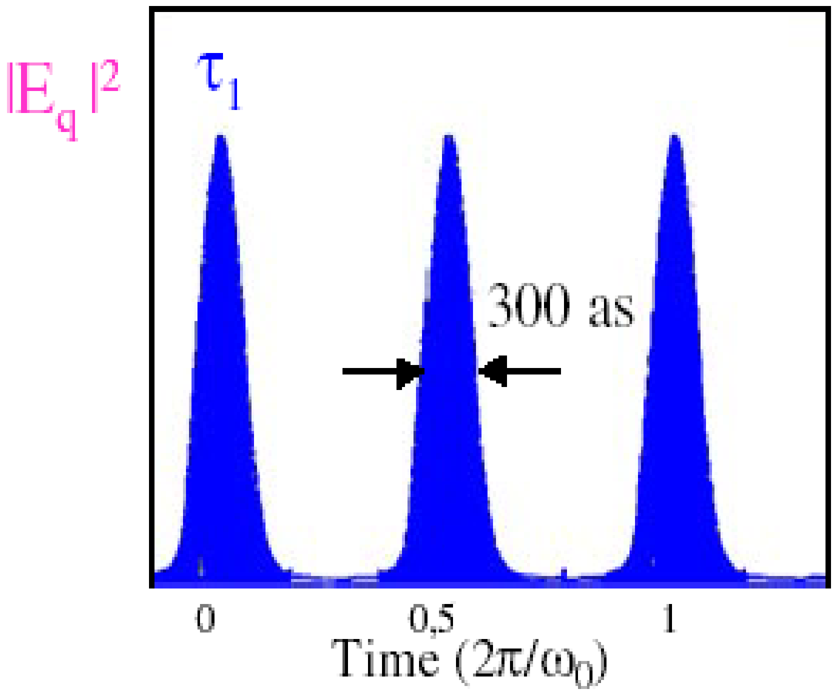
SI units!

Attosecond pulse trains

$$\Delta k = \underbrace{\Delta k_a}_{<0} + \underbrace{\Delta k_{fe}}_{>0} + \underbrace{\Delta k_{foc}}_{>0} + \underbrace{\Delta k_{dip}^{traj}}_{\text{-sign}(z)}$$

The two trajectories are phase matched differently!



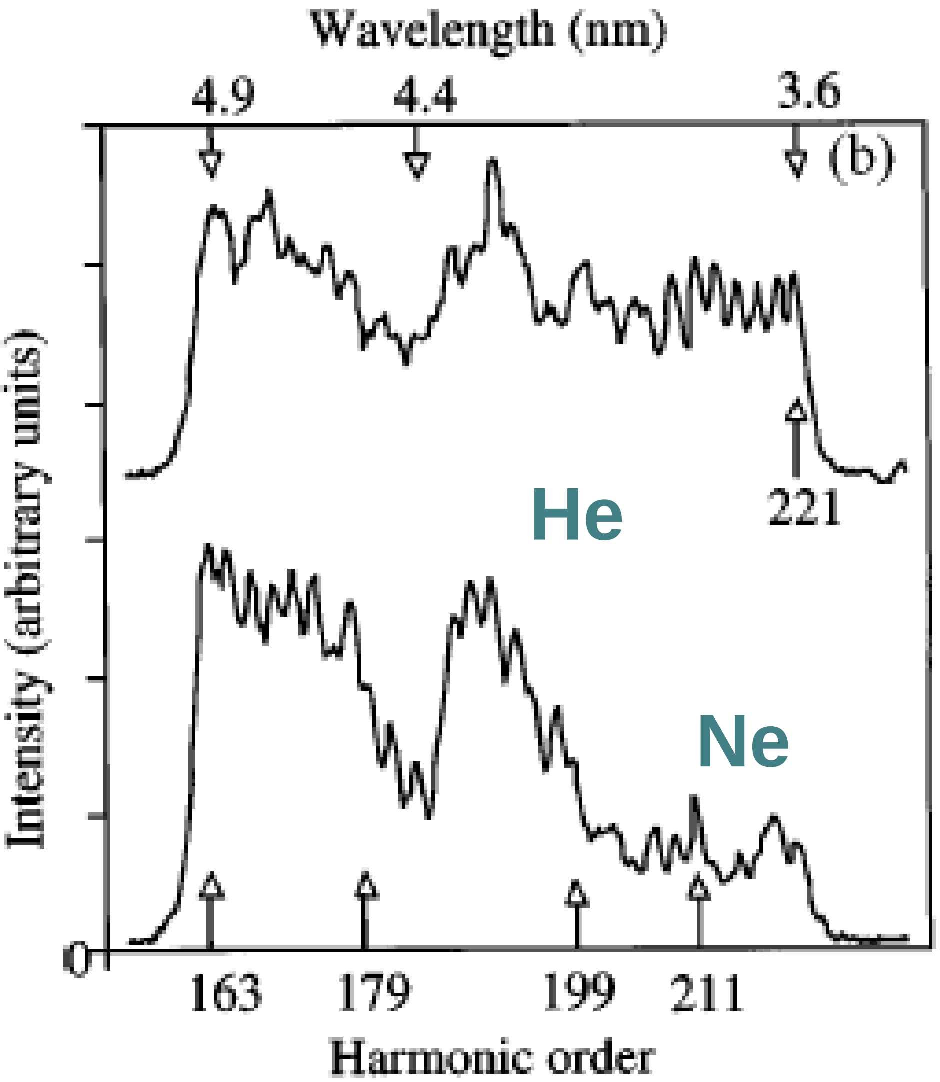


Antoine et al, PRL 1996

HHG with even shorter pulses "few-cycle"

1997

Harmonics in the water window



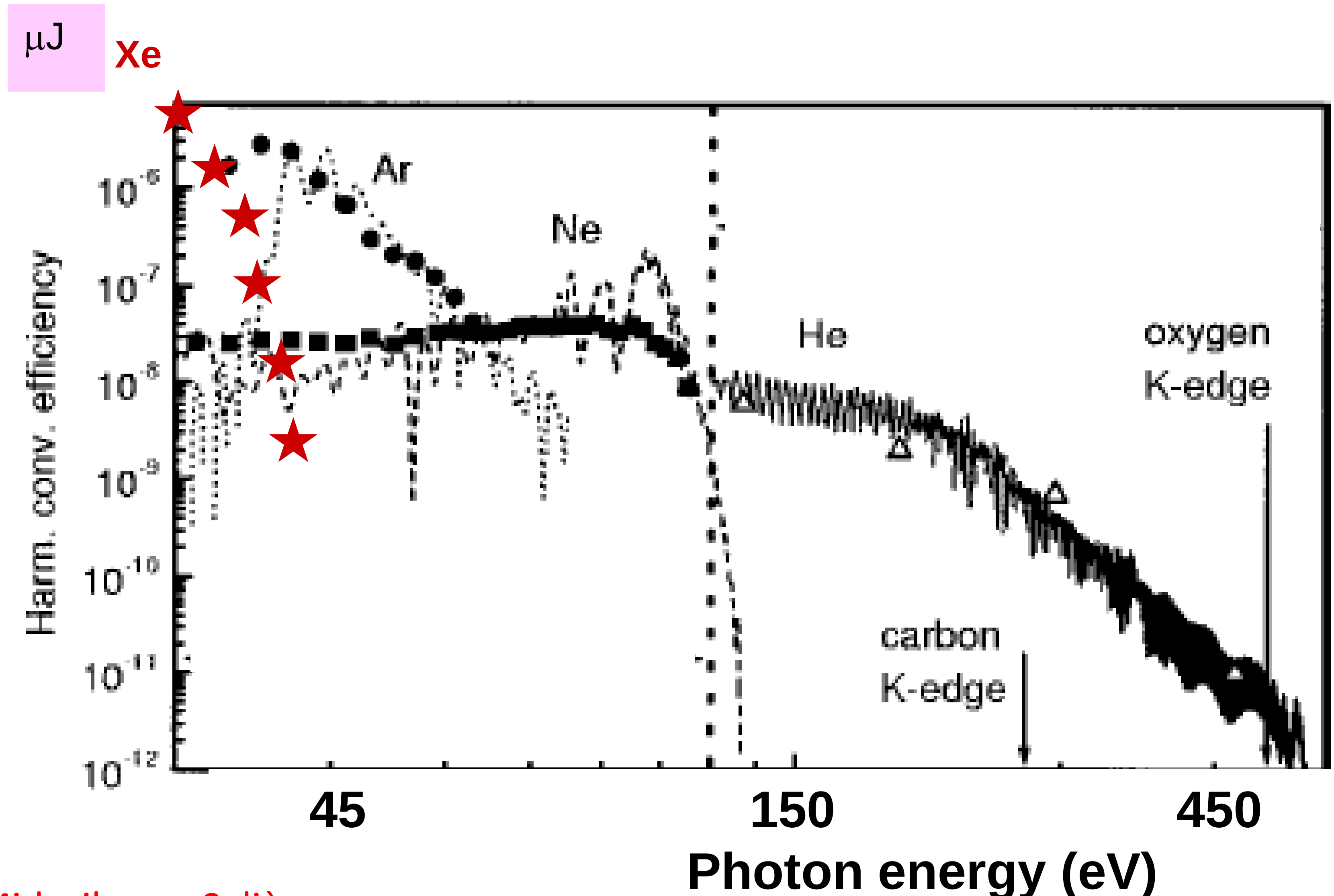
Short pulses < 20 fs

Spielman et al, 1997

Chang et al, 1997

Harmonic generation in capillaries

Harmonics in the water window



Midorikawa, Salières
Murnane, Kapteyn
Brabec and Krausz 2000

Multicolor / Low frequency

Attoseconds?

Yes but how ?

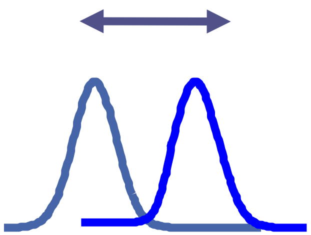
1999

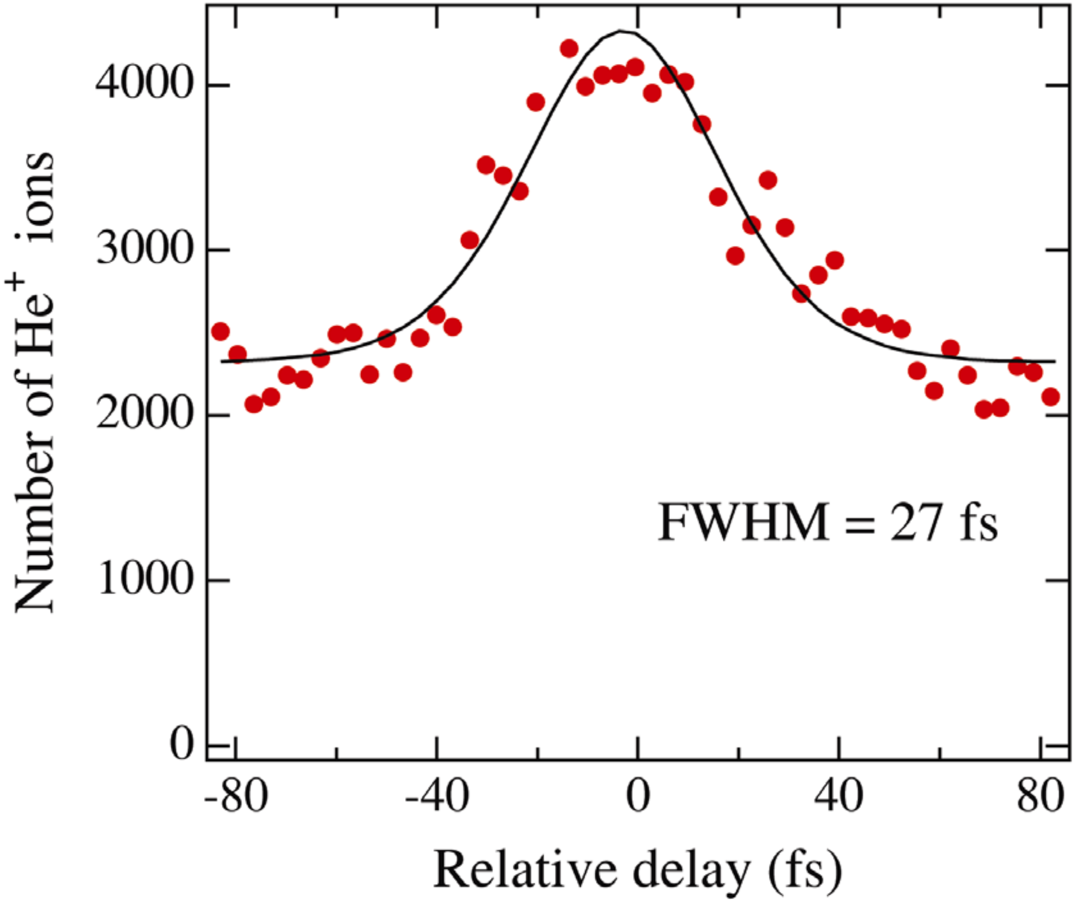
ATTO Network (XTRA, ATTOFEL)

Aim: ”bring attosecond physics into experimental reality”

How: autocorrelation, streaking with a low frequency field

Autocorrelation of harmonic pulses





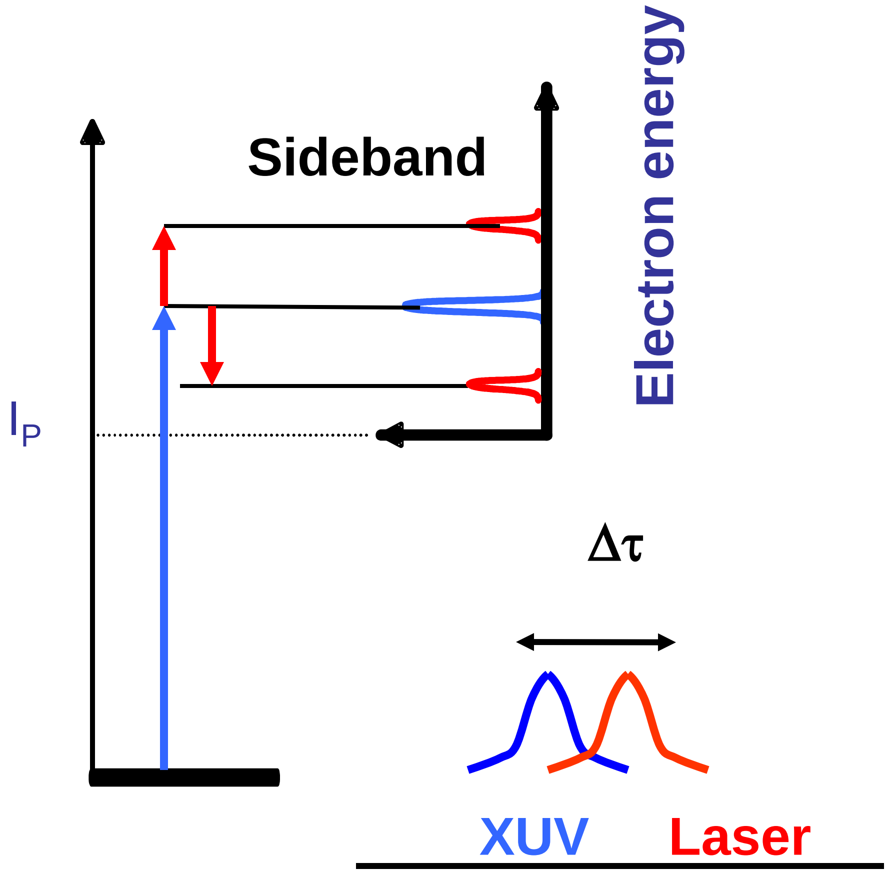
9th harmonic

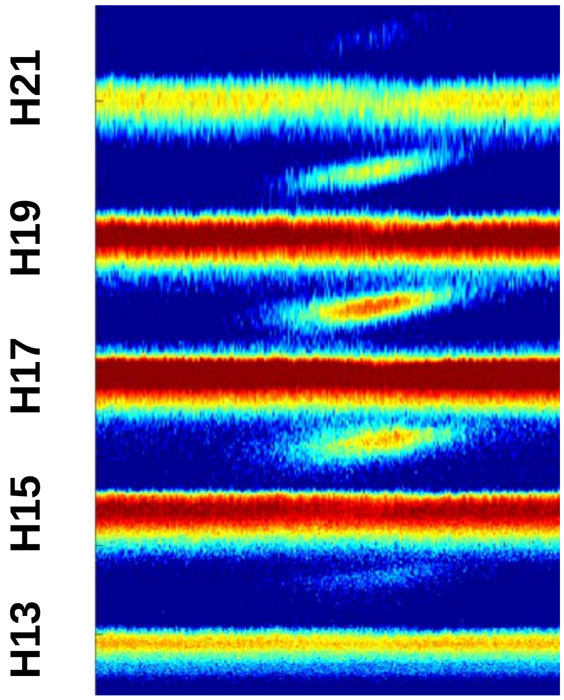
Kobayashi et al, 1998

Tzallas et al., 2003

→ **Autocorrelation of attosecond pulses ?**

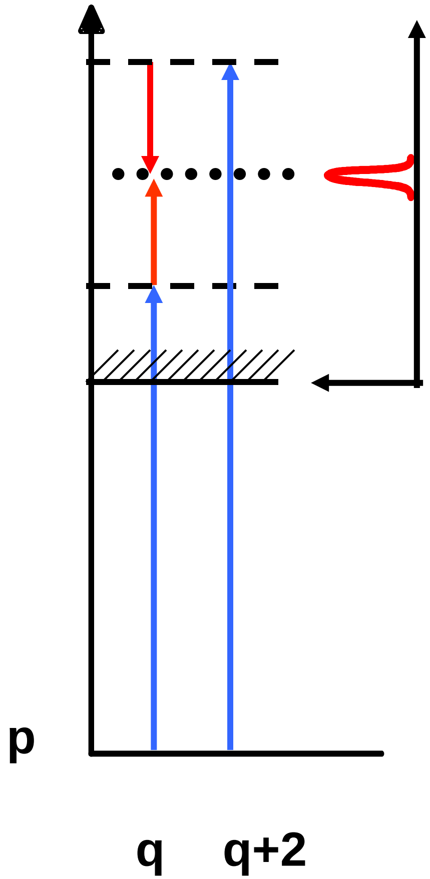
Cross-correlation of harmonic pulses

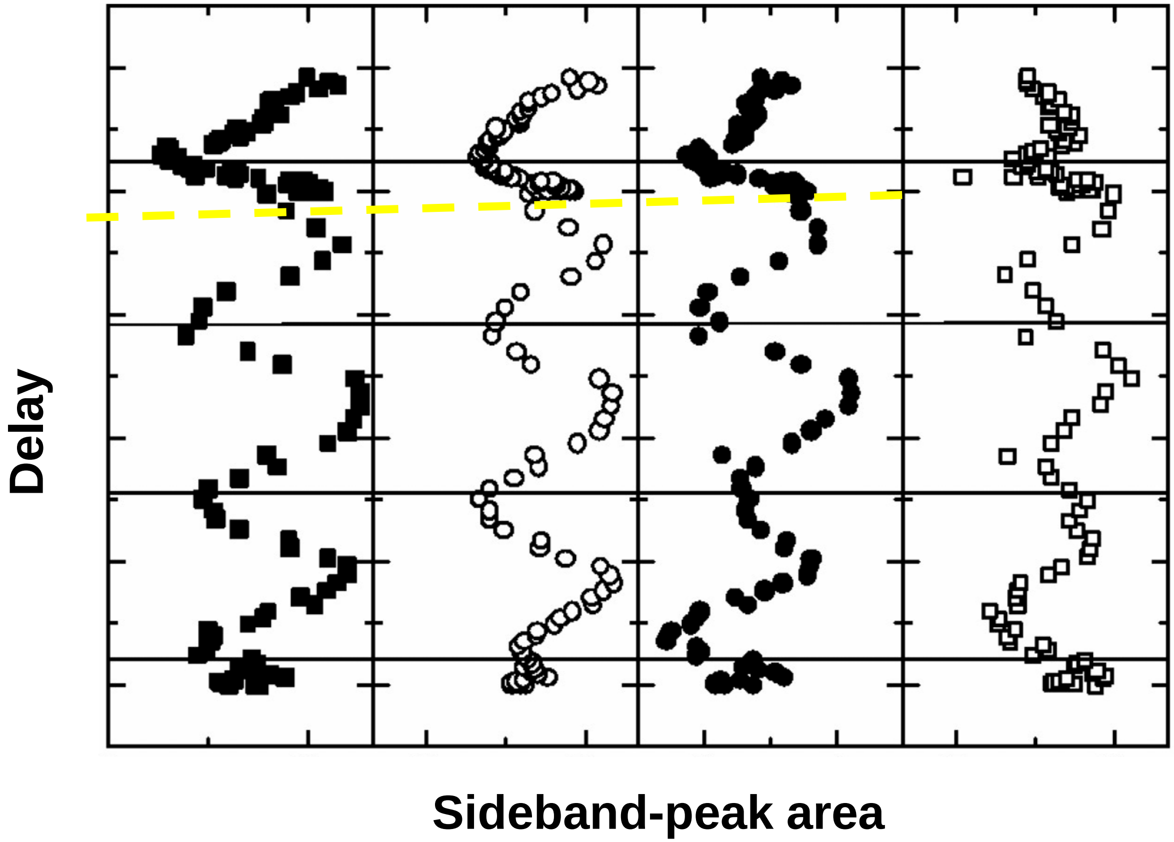




Glover et al., 1996
Schins et al, 1996
Mauritsson et al., 2003

Attosecond pulse trains

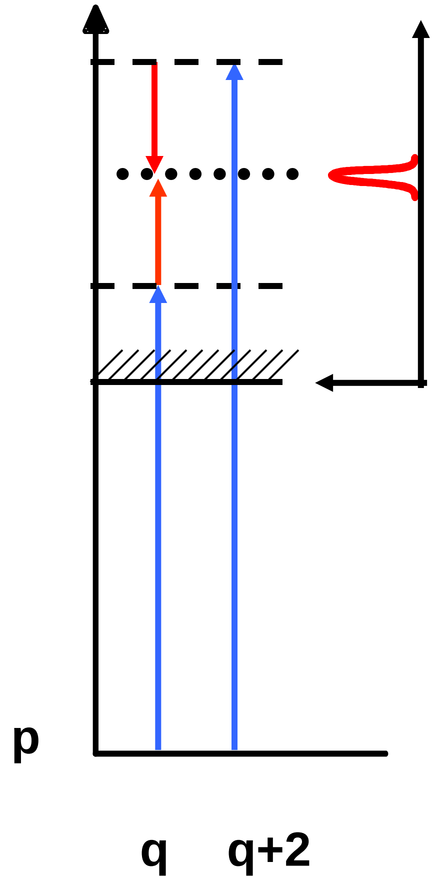


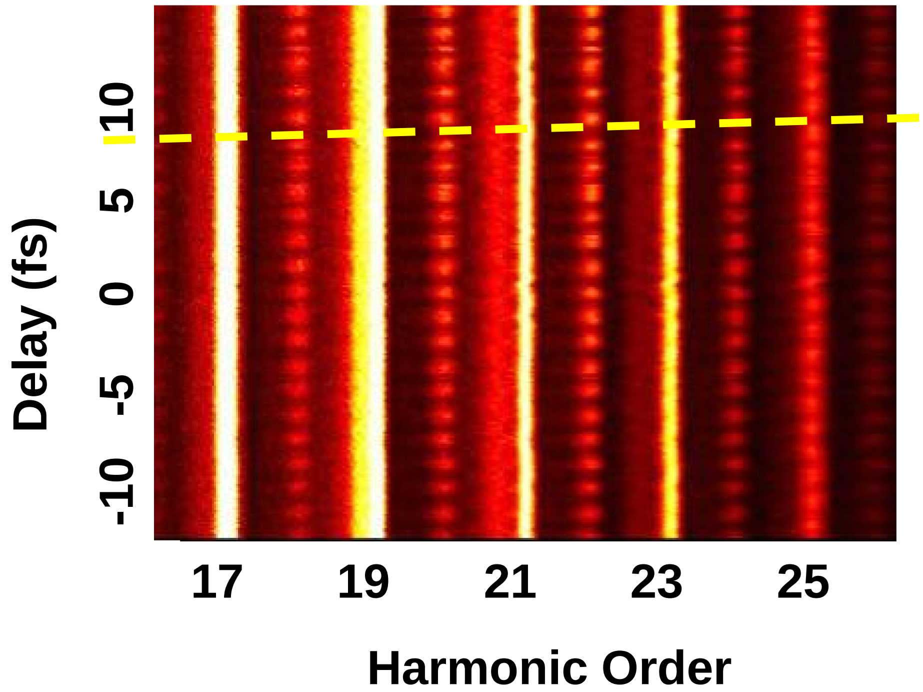


“RABBITT”

Paul et al, 2001
Mairesse et al. 2003

Attosecond pulse trains



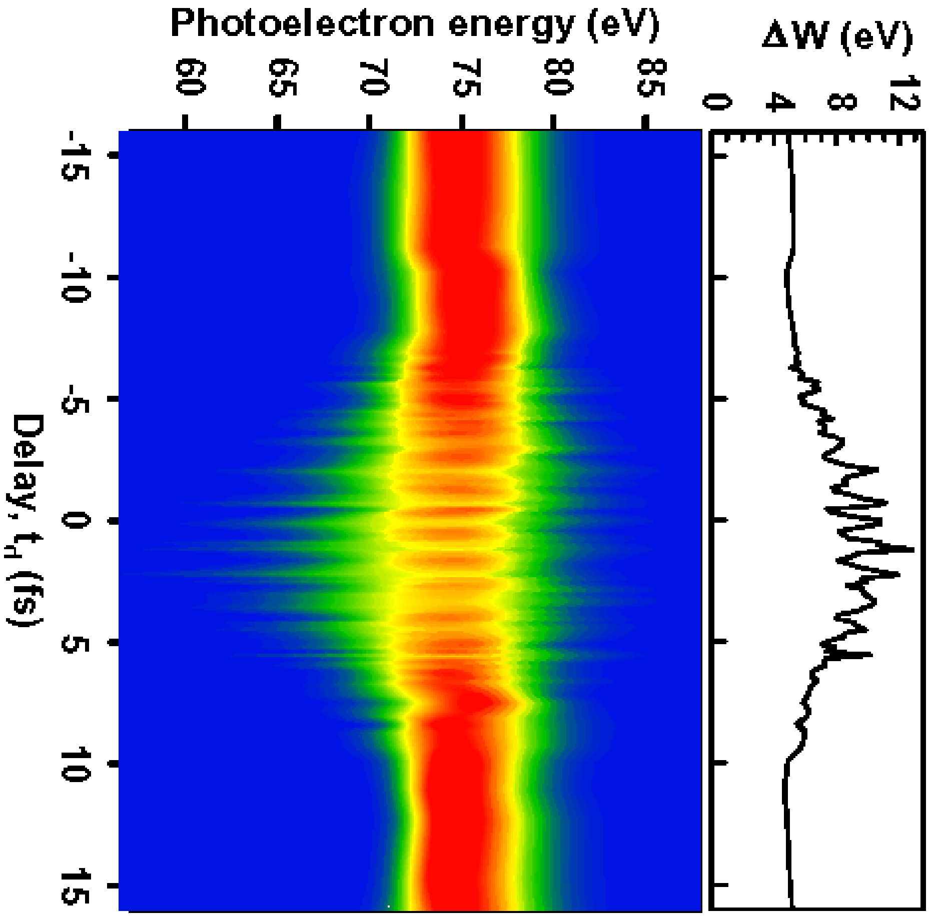


"RABBITT"

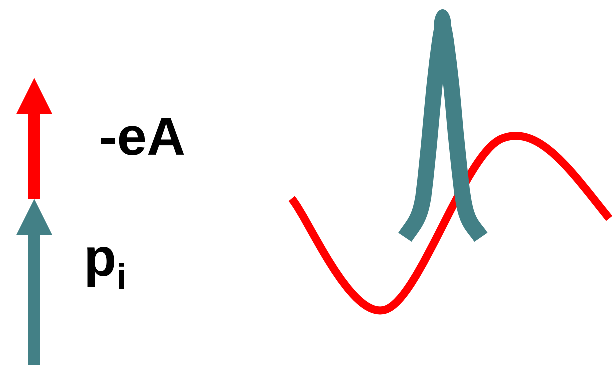
$$\Phi_{q+2} - \Phi_q \propto \frac{\partial \Phi}{\partial \omega} \qquad I(\omega), \frac{\partial \Phi}{\partial \omega} \Rightarrow E(\omega), E(t)$$

Paul et al, 2001
Mairesse et al. 2003

Single attosecond pulses



AC Streak camera

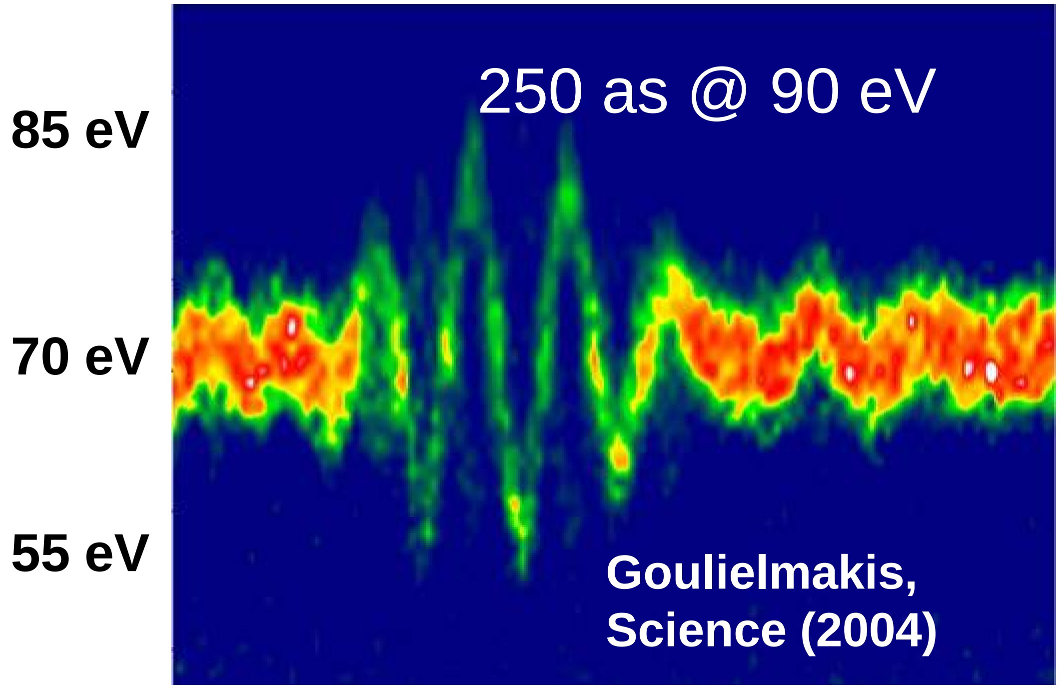


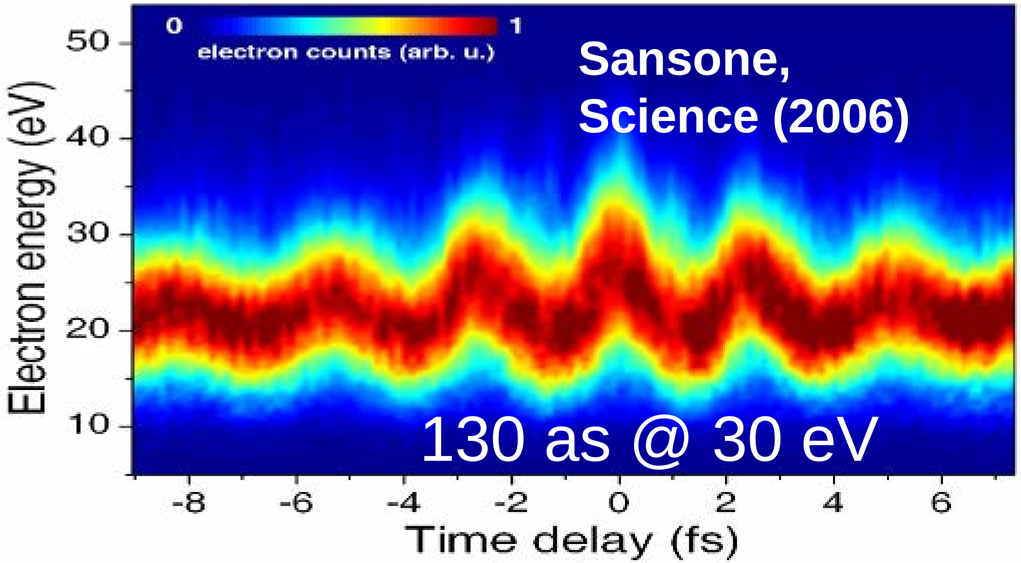
Hentschel et al, 2001

Kienberger et al, 2002

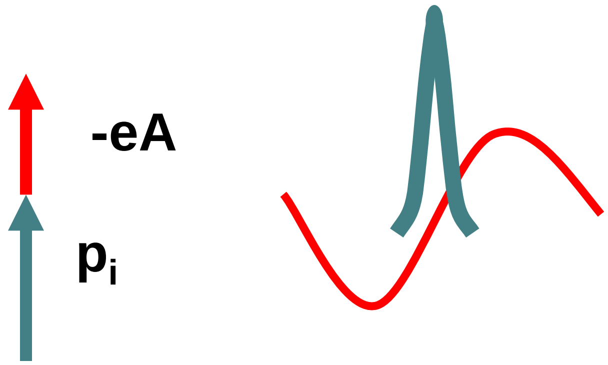
τ_x=650 (±150) as

Single attosecond pulses





AC Streak camera



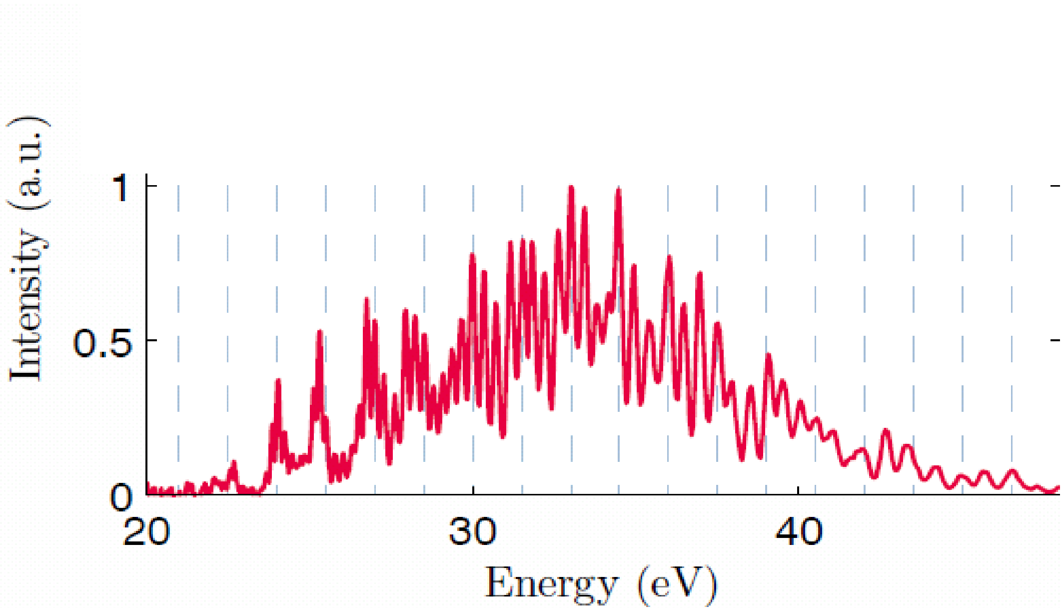
Where are we now?

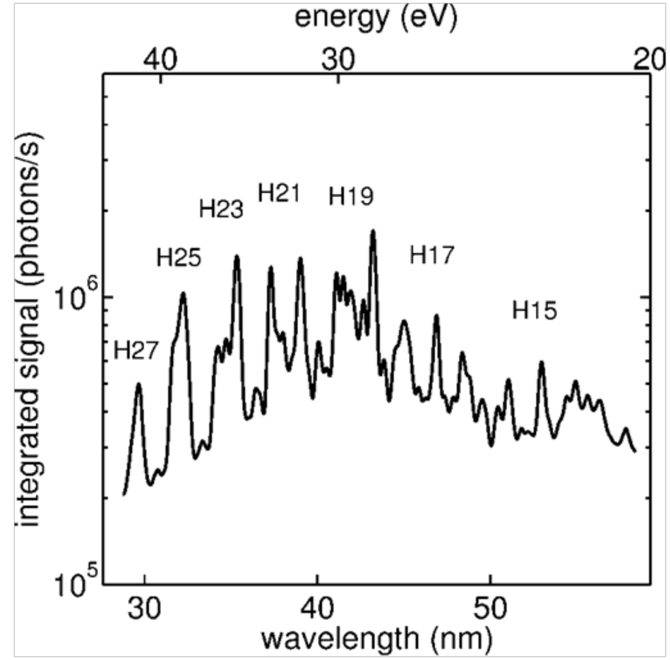
Applications

Optimisation

2011

Fun with harmonics !





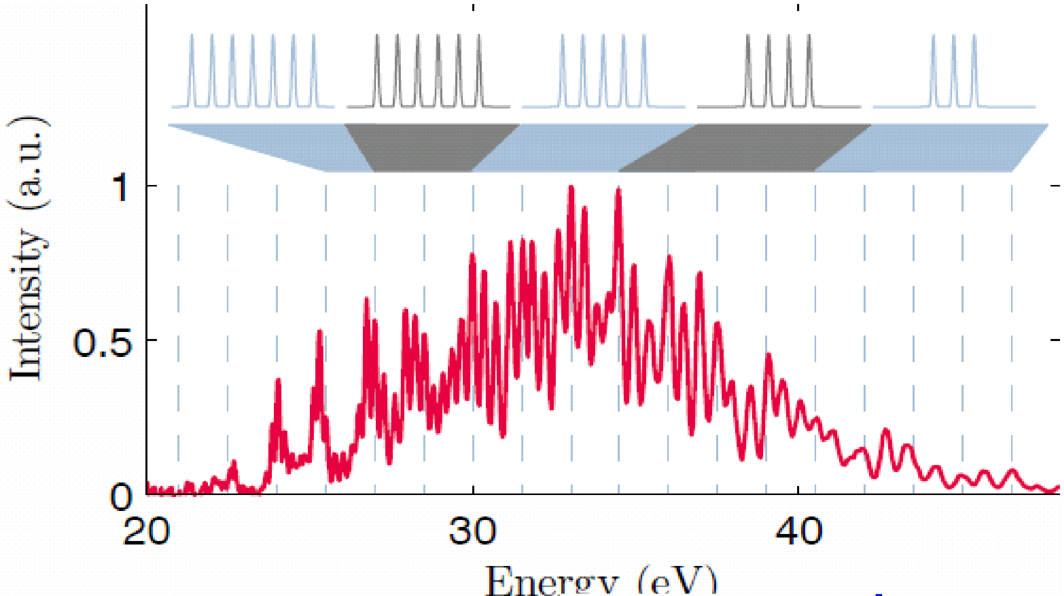
Few cycle laser pulse (15 fs)
CEP-stable - CEP-dependent effect
Two color field

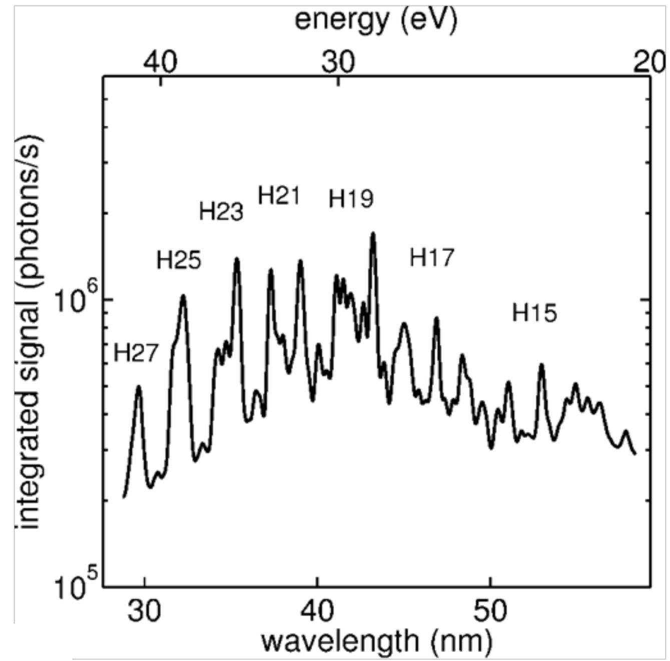
Mansten et al., PRL 2009

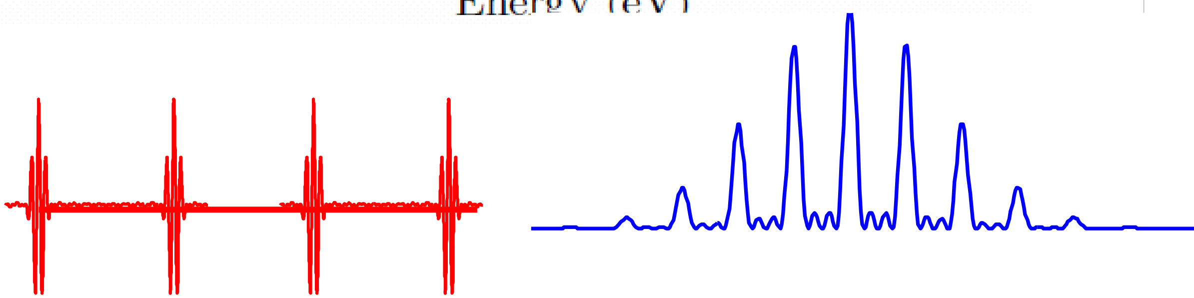
Long pulse (45 fs)
No CEP stability
One color
Tight focusing (100 kHz)

Heyl et al., submitted, see poster

Fun with harmonics !







”Secondary maxima”

Transient phase matching of the long trajectory!

Thank you and
have fun in
your research!

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