Extreme Nonlinear Optics

"Extreme":

1) Treating the polarization in terms of a perturbative expansion in powers of the electric field fails to converge
2) The electric field of the pulse must be considered, not just its intensity envelope

When these conditions are met:

Electric field strength of an optical pulse is comparable to that of an atom

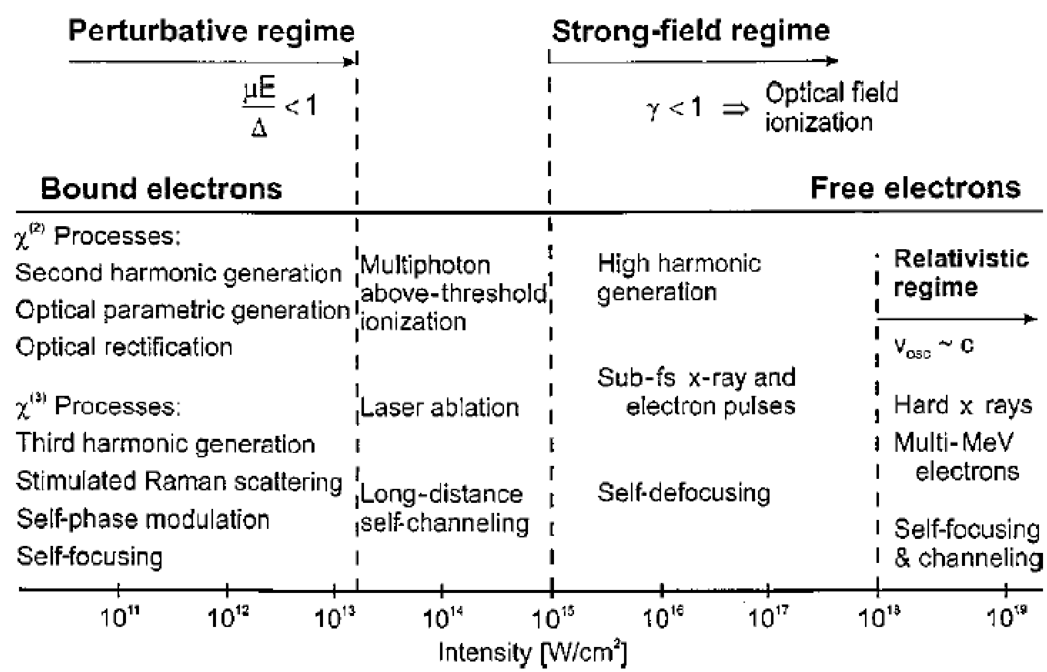
Interesting phenomena in this regime:

Tunneling and above-barrier ionization

High harmonic (EUV – soft x-ray) generation

Attosecond pulses

Regimes of Nonlinear Optics



Breakdown of perturbative nonlinear optics

The standard approach to nonlinear optics:

$$P \propto \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \chi^{(4)}E^4 + \cdots$$

In order for this to converge, require

$$\frac{\chi^{(k+1)}E^{k+1}}{\chi^{(k)}E^k} < 1$$

Consider a transition between two **bound** states

Perturbative expansion of the density matrix gives that successive orders of χ are related by a factor $\mu_{12}/\hbar\Delta$

Δ is detuning between field and resonance

Giving

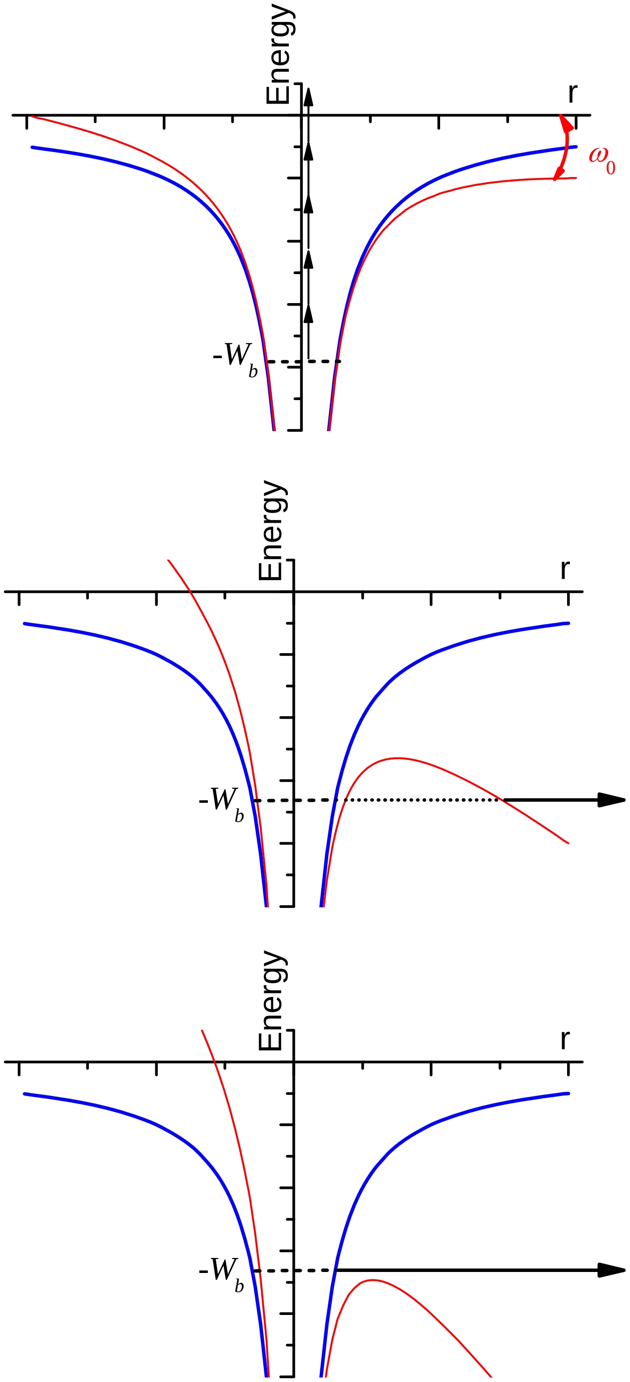
$$\frac{\mu_{12}E}{\hbar\Delta} < 1$$

For an atom, estimate $\mu_{12} \approx e\,a_0$ where a_0 is the Bohr radius

$$\frac{e\,a_0 E}{\hbar\Delta} < 1$$

But what about ionization (bound-free transitions)?

Ionization Mechanisms



Regime where photon energy << binding energy (W_b)

There are three ionization paths:

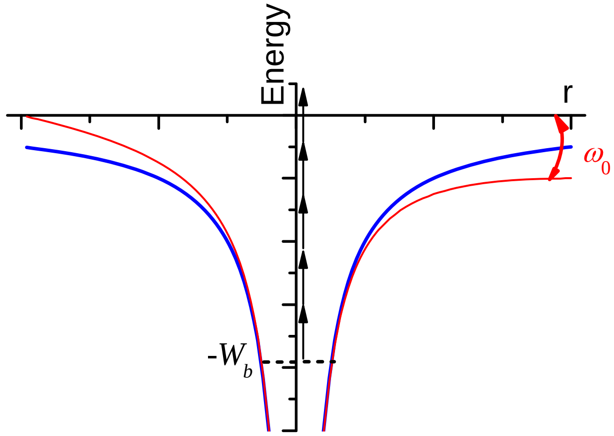
1) Multiphoton ionization (perturbative)
2) Tunneling ionization
3) Above-barrier ionization

Multiphoton regime

Perturbative

Potential due to E weak compared to atomic Coulomb potential

Ionization rate proportional to I^N where N is $W_b/\hbar\omega$



Classical picture:

Oscillating E heats electron

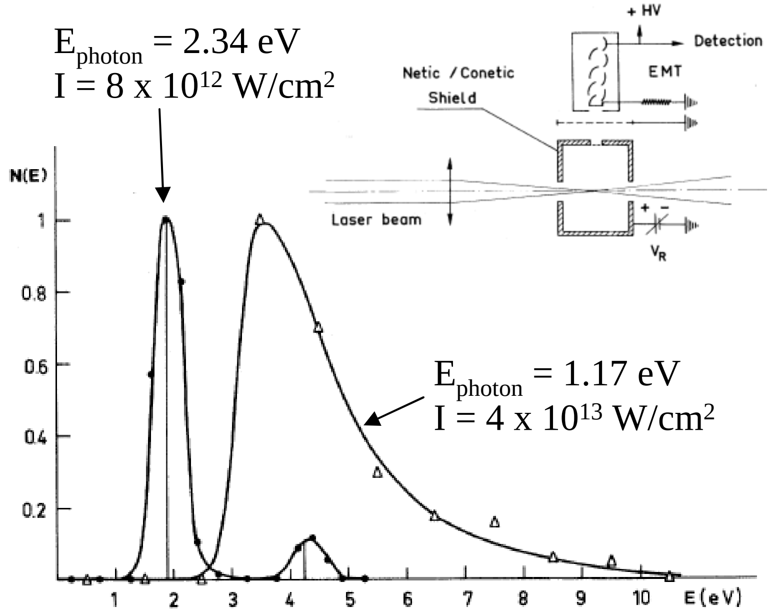
Quantum picture

Project eigenstates of Coulomb potential onto eigenstates of Coulomb + E potential

Repeat each half cycle of E

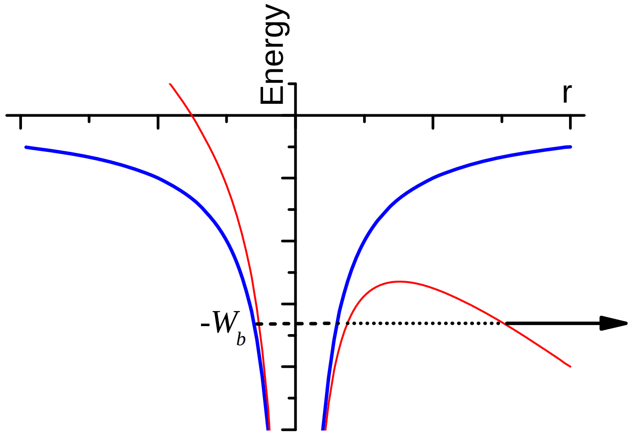
Coherently builds up over many oscillations periods

Acceleration of free-state by E is unimportant



Agostini, Fabre, Mainfray, Petite, Rahman, Phys. Rev. Lett. **42**, 1127 (1979)

Tunneling regime



Quasi-static:

Tunneling mainly occurs at peak field → ignore time variation

Does electron have time to tunnel?

Compare oscillation frequency to tunneling time

$$\tau = \frac{\sqrt{2mW_B}}{eE}$$

(there are multiple definitions of tunneling times, and they are all fraught with conceptual potholes)

The “Keldysh parameter” is

$$\gamma = \omega_0 \tau = \frac{\sqrt{2mW_B}}{eE}\omega_0$$

For $\gamma << 1$, tunneling is strong

For $\gamma \sim 1$ both tunneling and multiphoton absorption must be simultaneously taken into account.

Compare bound-bound to bound-free transitions

Which type of transition, bound-bound or bound-free, violates perturbative limit?

Compare

$$\frac{e\,a_0 E}{\hbar\Delta}$$ (characterizes bound-bound transitions)

to

$$\frac{\sqrt{2mW_B}}{eE}\omega_0$$ (characterizes bound-free transitions)

rewrite the latter using the generalized Bohr radius $a_0 = \hbar \big/ \sqrt{2mW_B}$

$$\frac{\sqrt{2mW_B}}{eE}\omega_0 = \frac{eEa_0}{\hbar\omega_0}$$

thus the ratio is just Δ/ω_0

$\Delta > \omega_0$ – free-bound transitions (ionization) more important

above threshold ionization, high harmonics

$\Delta < \omega_0$ – bound-bound transitions more important

Rabi flopping → carrier-wave Rabi flopping

Relativistic regime

For a free electron, the amplitude of its wiggling in a e-m field is

$$a_w = \frac{eE}{m\omega_0^2}$$

The corresponding cycle averaged kinetic energy is

$$U_p = \frac{e^2 E^2}{4m\omega_0^2}$$

which is known as the "pondermotive potential"

(U_p = 93 eV, a_w = 12.4 nm for $I = 10^{15}$ W/cm^2 and λ = 1 μm)

Note:

Keldysh parameter is

$\gamma^2 = W_b/2U_p$

In the relativistic regime, U_p ~ rest mass of electron

$$\sqrt{\frac{4U_p}{mc^2}} \geq 1$$

This occurs around 10^{18} W/cm^2

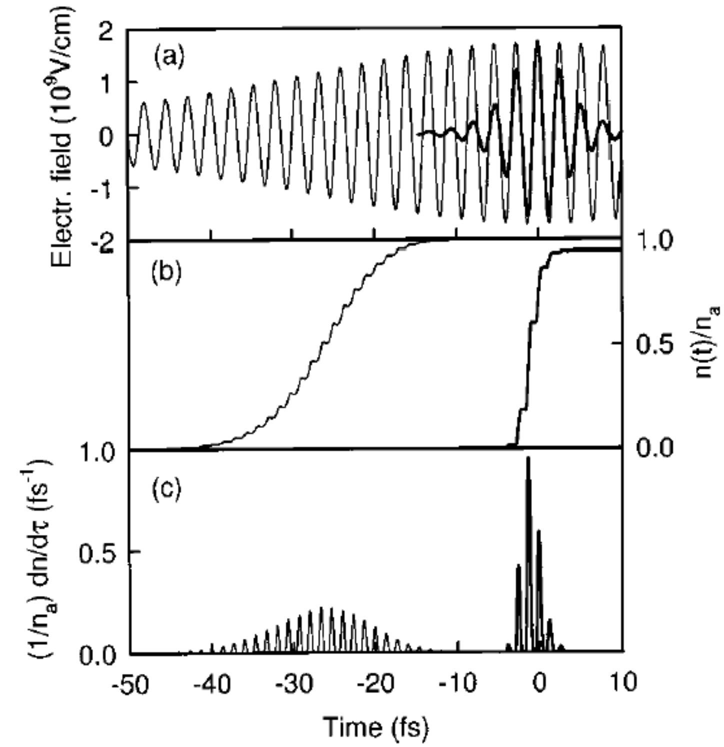
In this regime the electron wiggle amplitude is comparable to the wavelength of light and the polarization is no longer local.

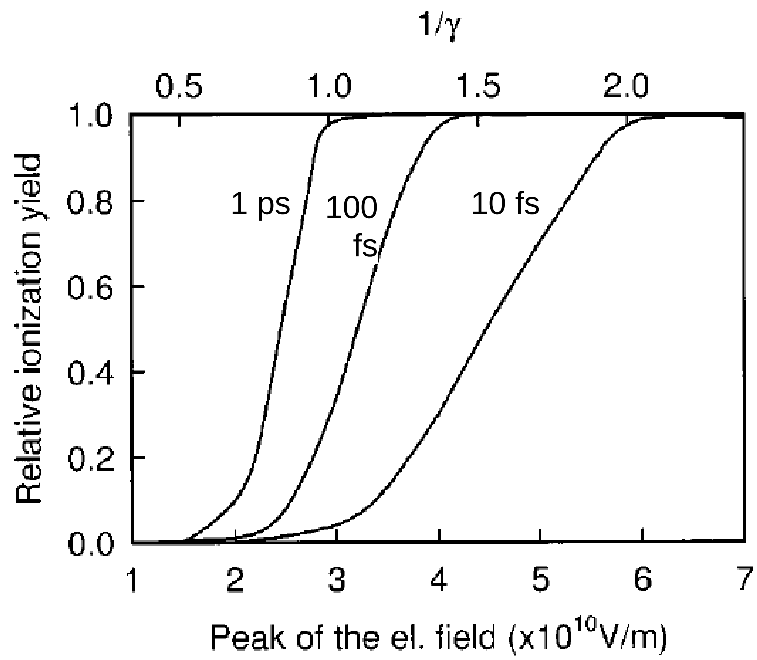
Pulsewidth

Does the pulsewidth make any difference in achieving the tunneling regime?

Yes, essential

- For a long pulse, multi-photon ionization during early part of pulse results in complete ionization before peak fields are achieved
- Only for ~ 10 fs pulses does significant ionization occur in tunneling regime





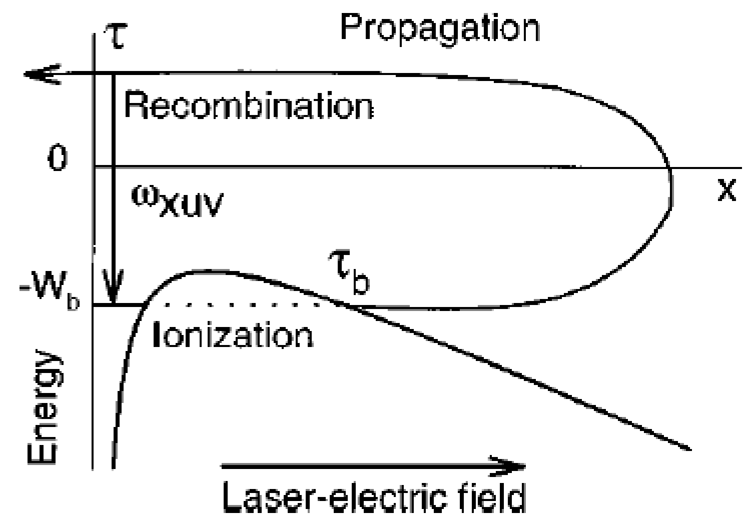
High Harmonic Generation

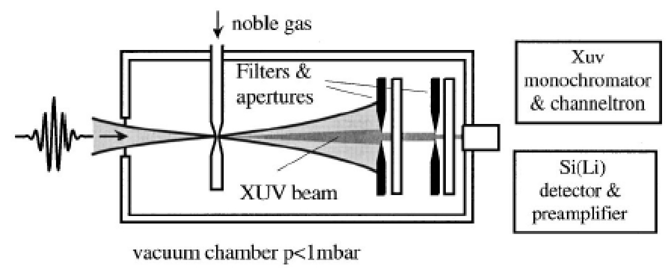
High harmonic generation occurs when the ionized electron is accelerated back to the ionic core by the laser field

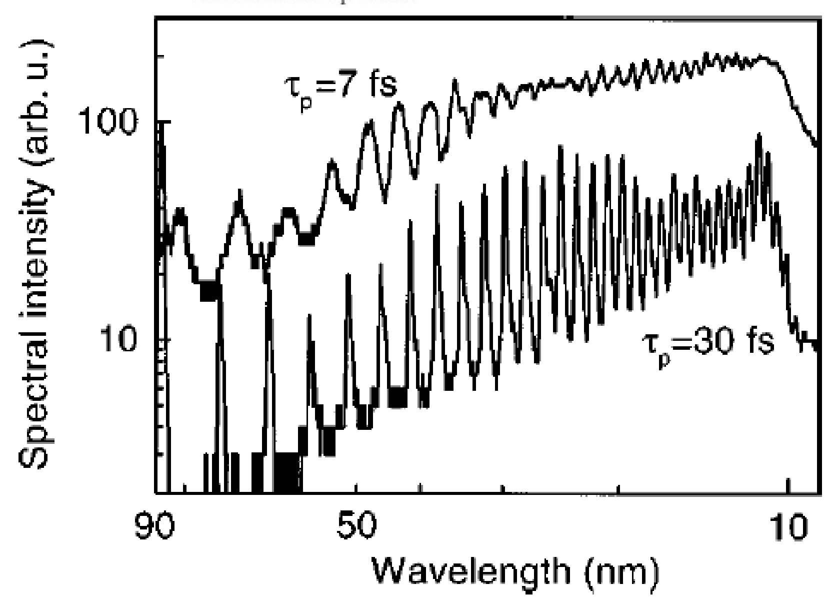
- Energy gained from the field is released as a high energy photon
- Only occurs for linearly polarized laser field

Spectrum is series of spikes at multiples of the frequency of the laser field

- →absorption of integer number of laser photons
- → Series of attosecond pulses
  - 1 per ½ cycle of laser field
  - train of pulses is comb of frequencies
  - consistent with broadened lines for shorter laser pulses







Strong field approximation

Theoretically, high harmonic generation is often treated in the strong field approximation (SFA):

- Divide time into 3:
  - Before the electron interacts with the laser, evolution only due to atomic potential
  - Absorption of photon, electron “promoted” from bound to free state
  - After absorption, electron moves only under influence of laser electric field (classical mechanics)
- Furthermore, atomic potential often treated as delta function in space

SFA allows analytic calculations, but misses significant physics:

- No multiphoton processes (needs finite potential)
- No interaction between ionized electron and atomic core (no HHG!)
- No gauge invariant (theoretical detail…)

But it actually works quite well for noble atoms

- Large binding energy for outer electron

Cutoff energy

The maximum energy of the emitted photon is related to the energy imparted by the laser field

maximum is “cutoff”

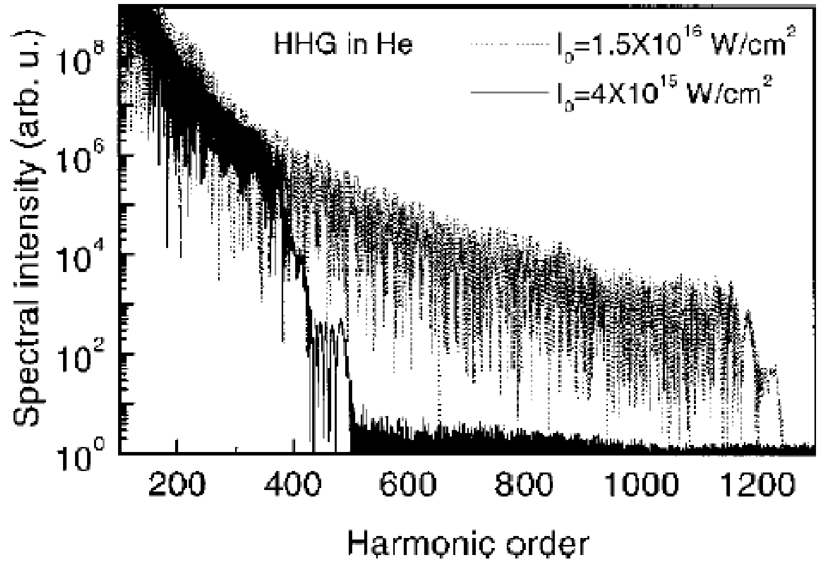
related to pondermotive potential:

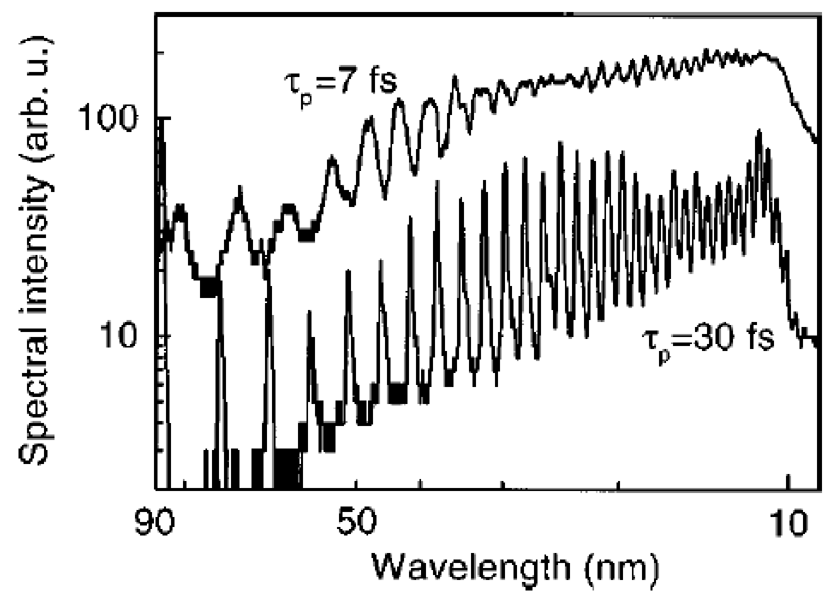
$N_c w_0 = W_b + 3.17\ U_p$

Predict:

Sharp drop at N_c

Independent of pulsewidth if peak intensity is same





Phase Matching

The conversion efficiency is typically limited “phase” matching

- really phase velocity of laser vs. group velocity of high harmonic

Overcome by using hollow fiber

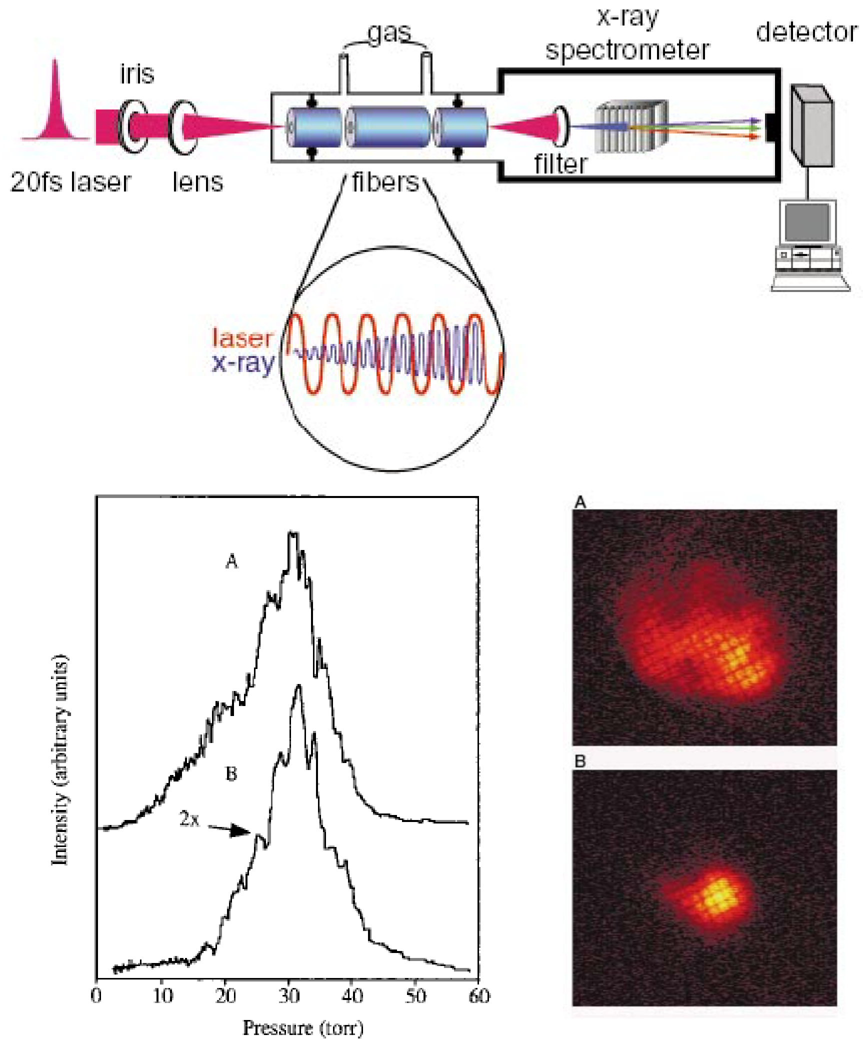
- Modifies phase velocity of laser
- Adjusted using pressure of gas
- high-harmonic not effected by fiber

Allows for increased interaction distance and generation of coherent beam

More recently, quasi-phase matching has been demonstrated (modulate inner diameter of fiber)

Phase-Matched Generation of Coherent Soft X-rays

Andy Rundquist, Charles G. Durfee III, Zenghu Chang, Catherine Herne, Sterling Backus, Margaret M. Murnane,* Henry C. Kapteyn



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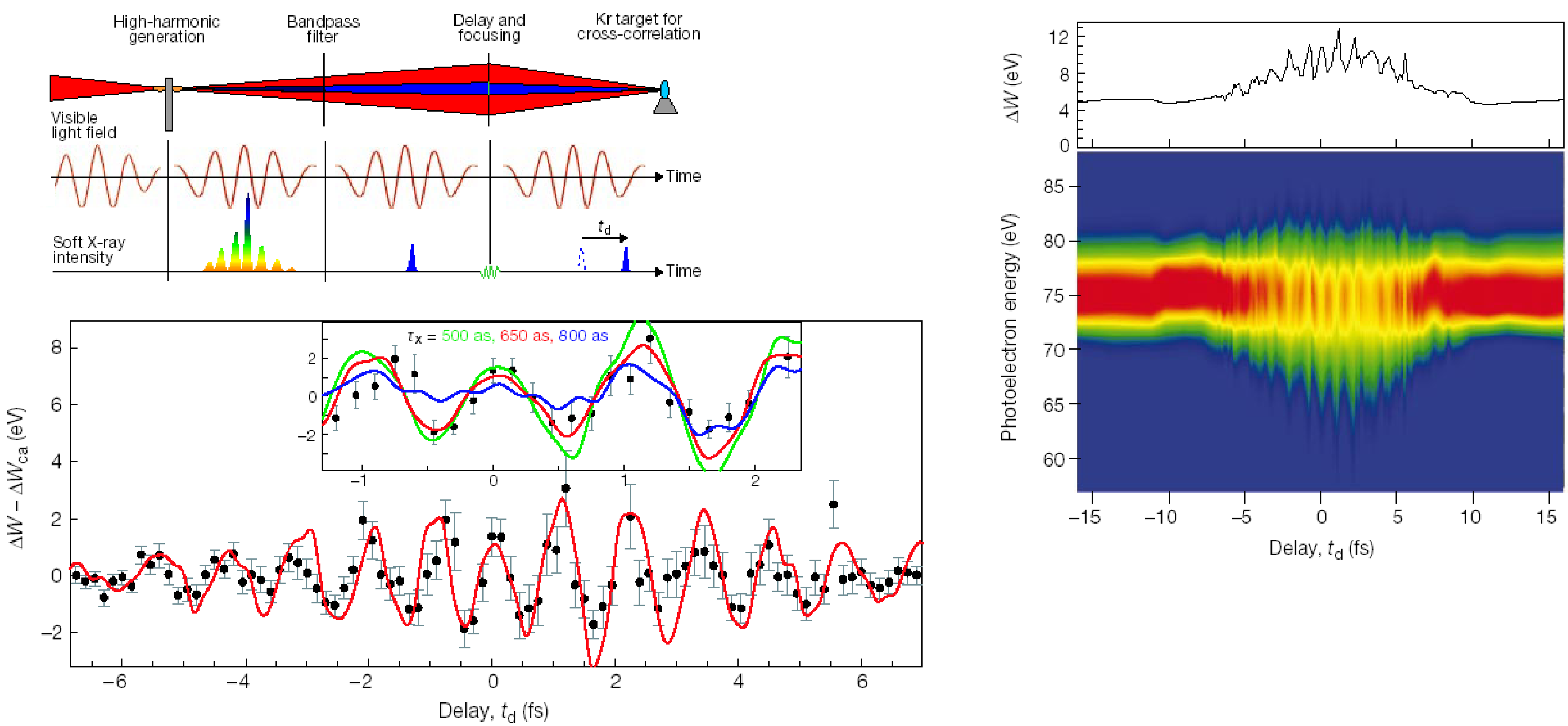
Attosecond pulses

The Fourier transform of the high harmonic spectrum corresponds to a train of attosecond pulses, but how to prove there are actually as pulses?

Attosecond metrology

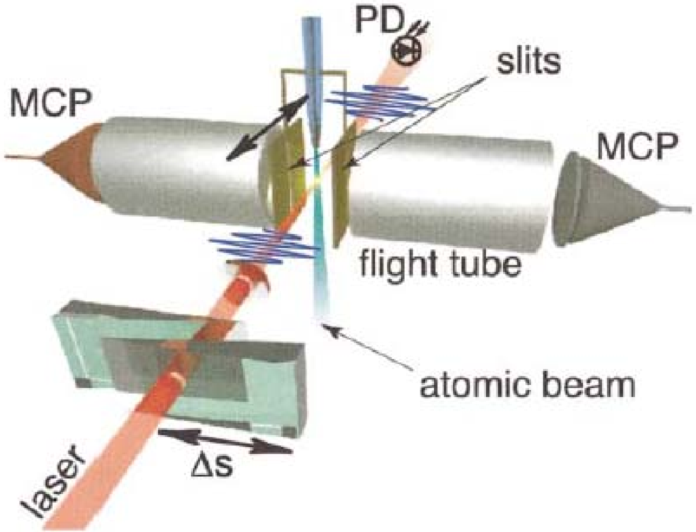
NATURE | VOL 414 | 29 NOVEMBER 2001 | www.nature.com 509

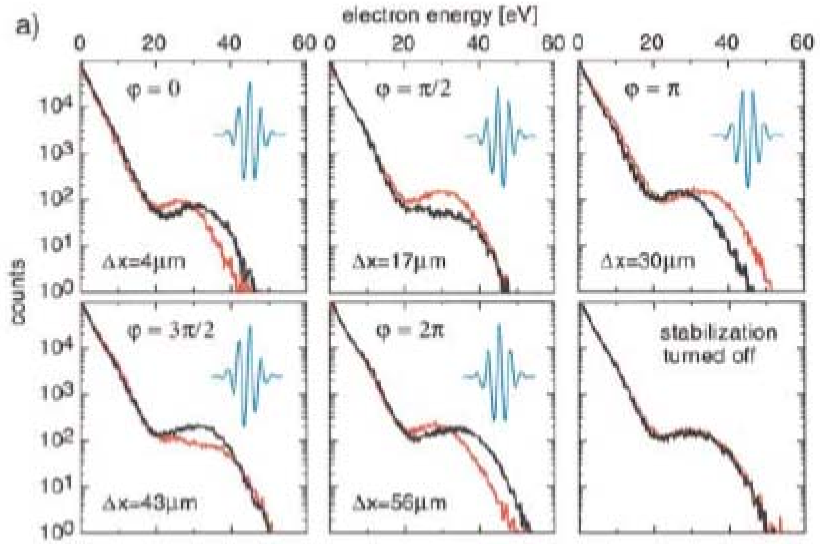
M. Hentschel*†, R. Kienberger*†, Ch. Spielmann*, G. A. Reider*, N. Milosevic*, T. Brabec*, P. Corkum‡, U. Heinzmann§, M. Drescher§ & F. Krausz*



Carrier-phase effects

But, hey, non perturbative is supposed to mean that the electric field not the intensity matters…doesn't that mean the carrier envelope phase matters?



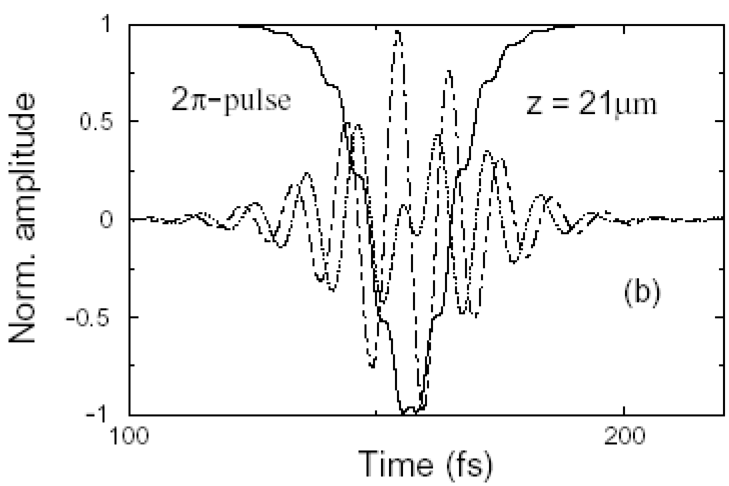


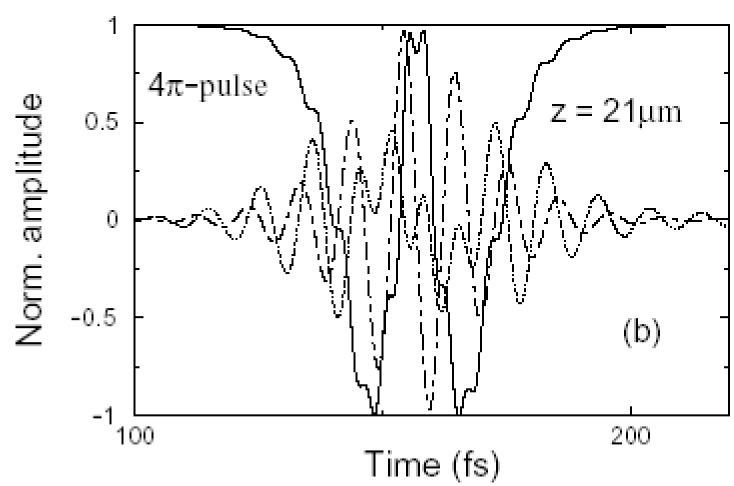
Paulus, Lindner, Walther, Baltus¡ka, Goulielmakis, Lezius, Krausz, Phys. Rev. Lett. **91**, 253004 (2003).

Bound state Extreme NLO: Carrier-wave Rabi Flopping

For bound states, the non-perturbative regime is easier to reach – just make detuning small

- Indeed Rabi flopping is non-perturbative
- But what happens when the pulse becomes really intense?
  - Rabi frequency approaches carrier frequency: Carrier-wave Rabi flopping
  - Slowly varying envelope approximation completely invalid
  - Area theorem invalid
  - Phase flip in polarization reminiscent of a "0π pulse"





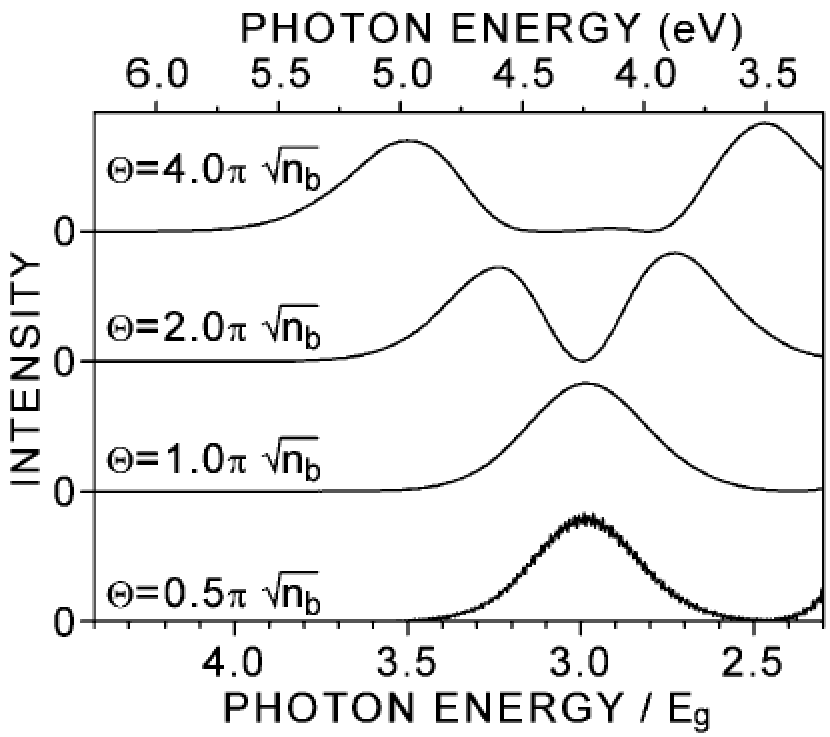
Hughes, Phys. Rev. Lett. **81**, 3363 (1998)

Experimental Carrier-wave Rabi Flopping evidence

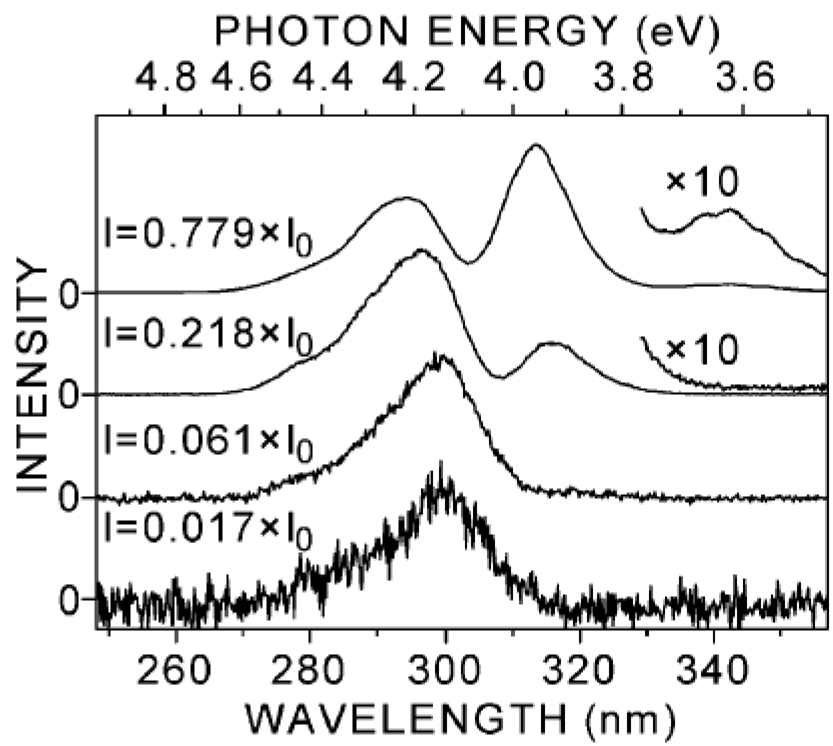
Observe 3rd harmonic (experimental reasons)

5 fs pulses hitting GaAs at band gap

Theory



Experiment



Muecke, Tritschler, Wegener, Morgner, Kaertner, Phys. Rev. Lett. **87**, 057401 (2001).