High Harmonic Generation

Physics 208A Presentation

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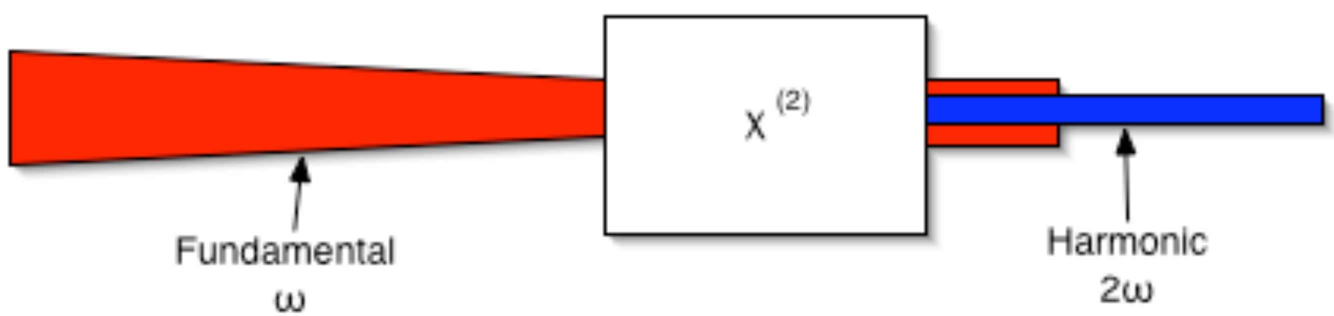
Perturbative Nonlinear Optics

$$P = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + ...$$

•Accurately treated by treating the polarization as a power series in E.

•With sufficiently intense laser fields, the higher order terms give rise to Fourier components of the polarization at harmonics of the laser frequency, creating radiation at harmonics of the laser frequency.



- First demonstration of second harmonic generation by P.A. Franken *et al.* (1961).
- Laser intensity of ~1×10^{7} W/cm^2
- E ~10^{5} V/cm = 10^{-3} V/Angstrom.
- For reference, the atomic field is

$$E_{at} \approx \frac{e}{a_0^2} \approx 50 \text{ V/Angstrom}$$

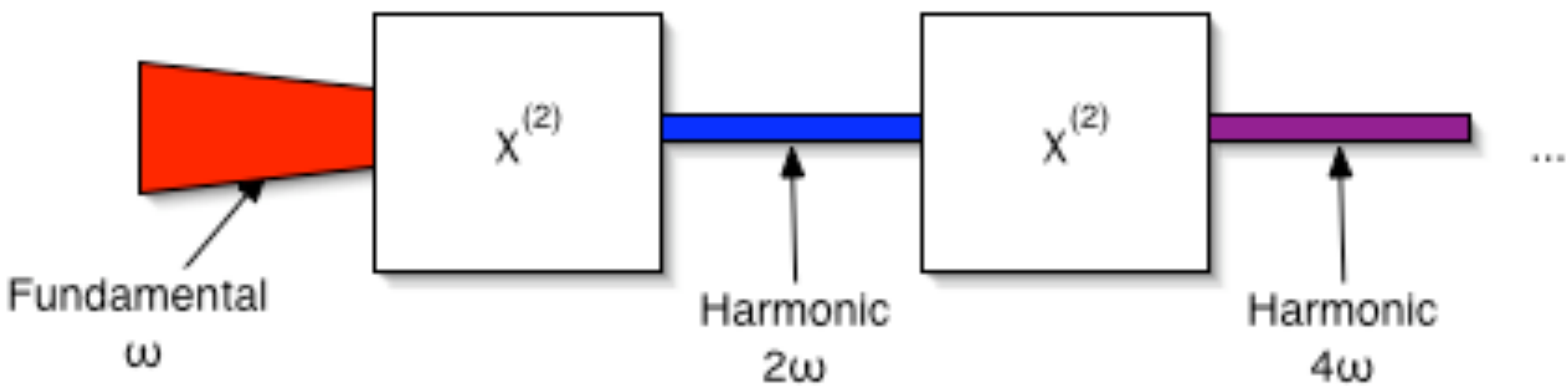
- With careful phase matching, generation of low order harmonics can be very efficient!

XUV or X-rays?

- ALS costs ~$100,000/day in operating costs.
- Large facility shared by many users $\Rightarrow$ beamtime is scarce.
- X-ray pulse durations typically ~100 picoseconds - too slow to resolve ultrafast dynamics.
- Can we use ~100th harmonic generation as a table-top, high brightness, ultrafast x-ray source?

How can we extend this process to higher harmonics?

- Most obvious first try would be to successively double:



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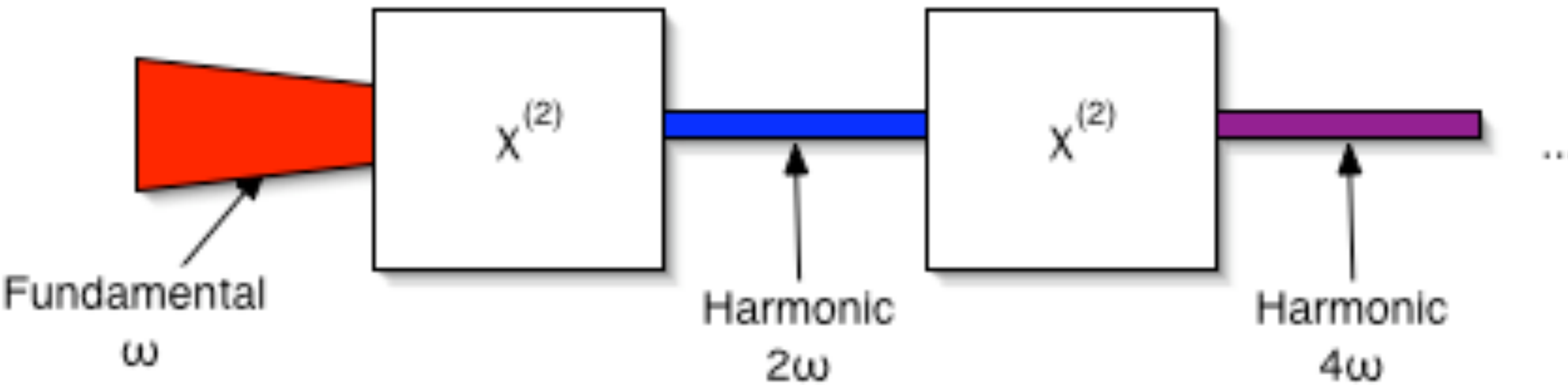
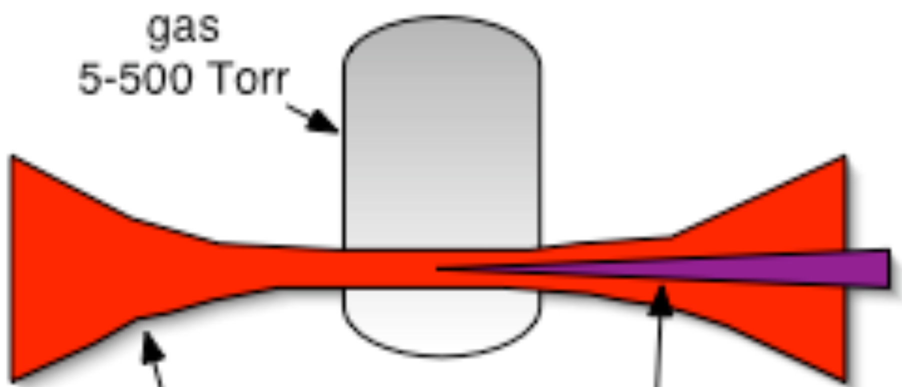


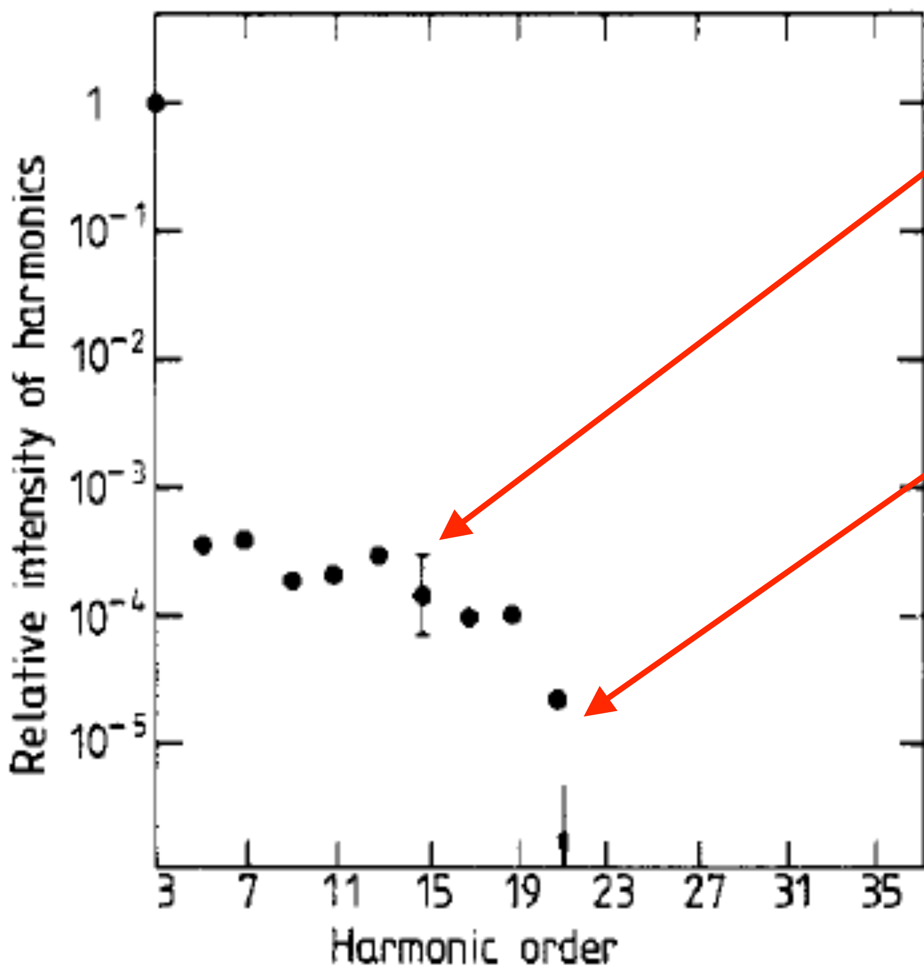
UV absorption in doubling crystals?!
Phase matching in region of strong dispersion?!

High Order Harmonics

- In order to get harmonics of very high order, we need to go to extremely intense electric fields, and use gas density targets.



Harmonic Spectrum



Ferray *et al.* (1988)

Harmonic spectrum is flat!

Sharp cutoff!

Old odd harmonics appear!

Not well described
By expansion of the
Polarization!

Outline

1. Microscopics: basic physics of high Harmonic Generation
2. Macroscopics: optimizing harmonic generation.
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Normal Nonlinear Optics

$$\frac{\chi^{(k+1)}E^{k+1}}{\chi^{(k)}E^{k}} \approx \frac{ea_0}{\hbar\Delta}E$$

$$\frac{1}{\gamma} = \frac{ea_B}{\hbar\omega_0}E = \text{Keldysh parameter}$$

$$a_0 = \text{ "Generalized" Bohr radius}$$

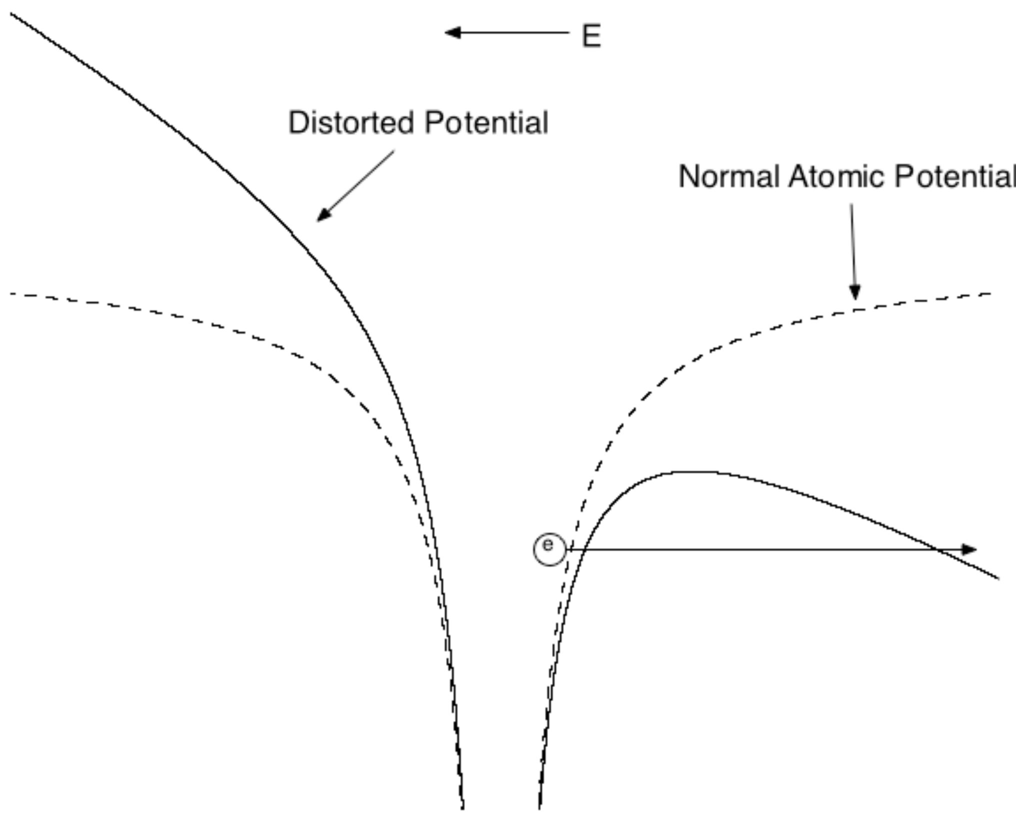
$$\omega_0 = \text{Laser frequency}$$

$$\Delta = \text{detuning} = \left|\omega_0 - \omega_{\text{tr}}\right|$$

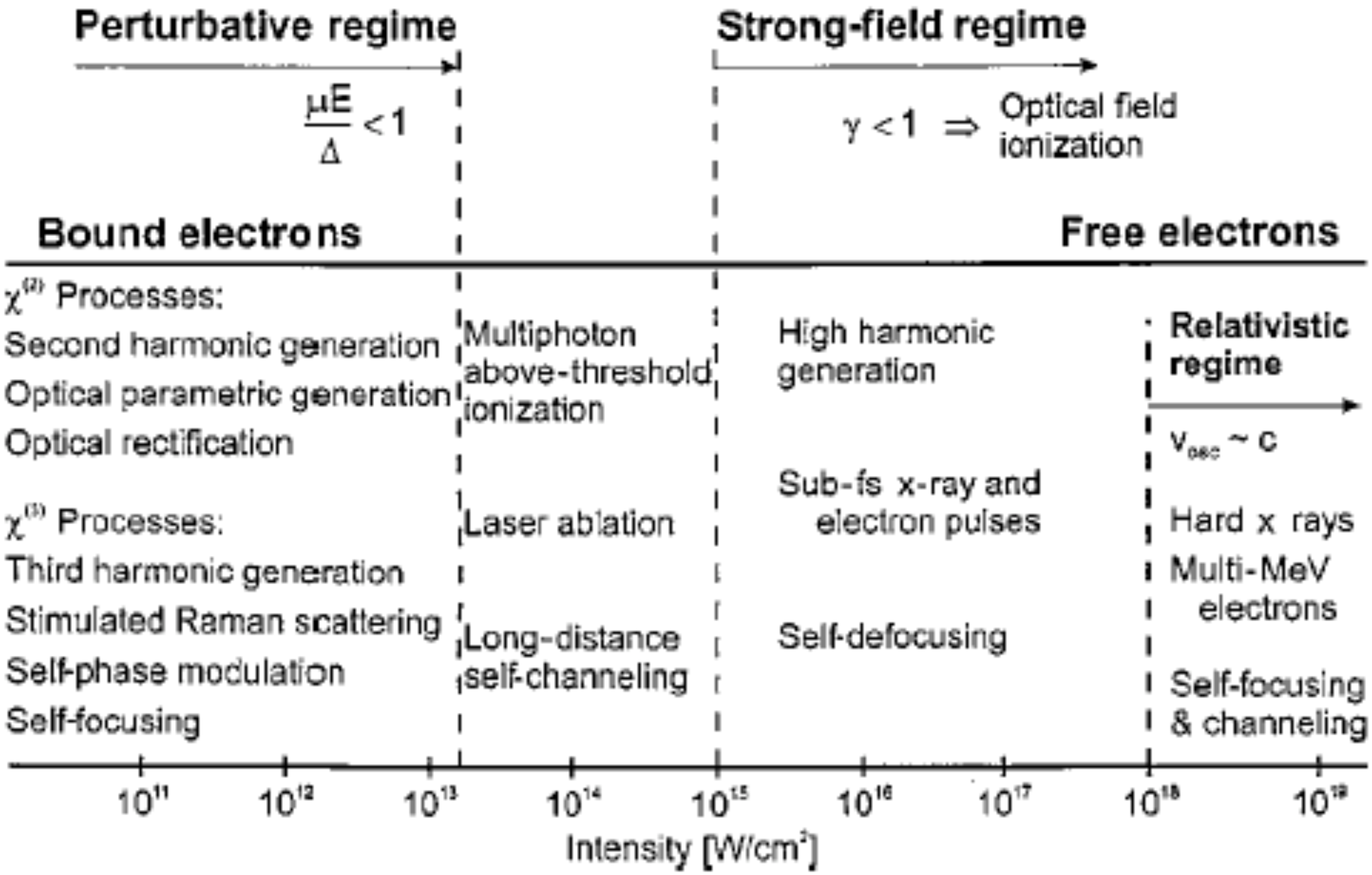
- Can treat problem adequately using perturbation theory in the regime of electric fields where these expressions are much less than one (I < ~10^{13} W/cm^2)

Strong Field Regime

- $1/\gamma$ greater than one ⇒ electric field of laser becomes comparable to atomic electric field felt by outer shell electrons.
- Atomic Potential is severely distorted ⇒ field ionization

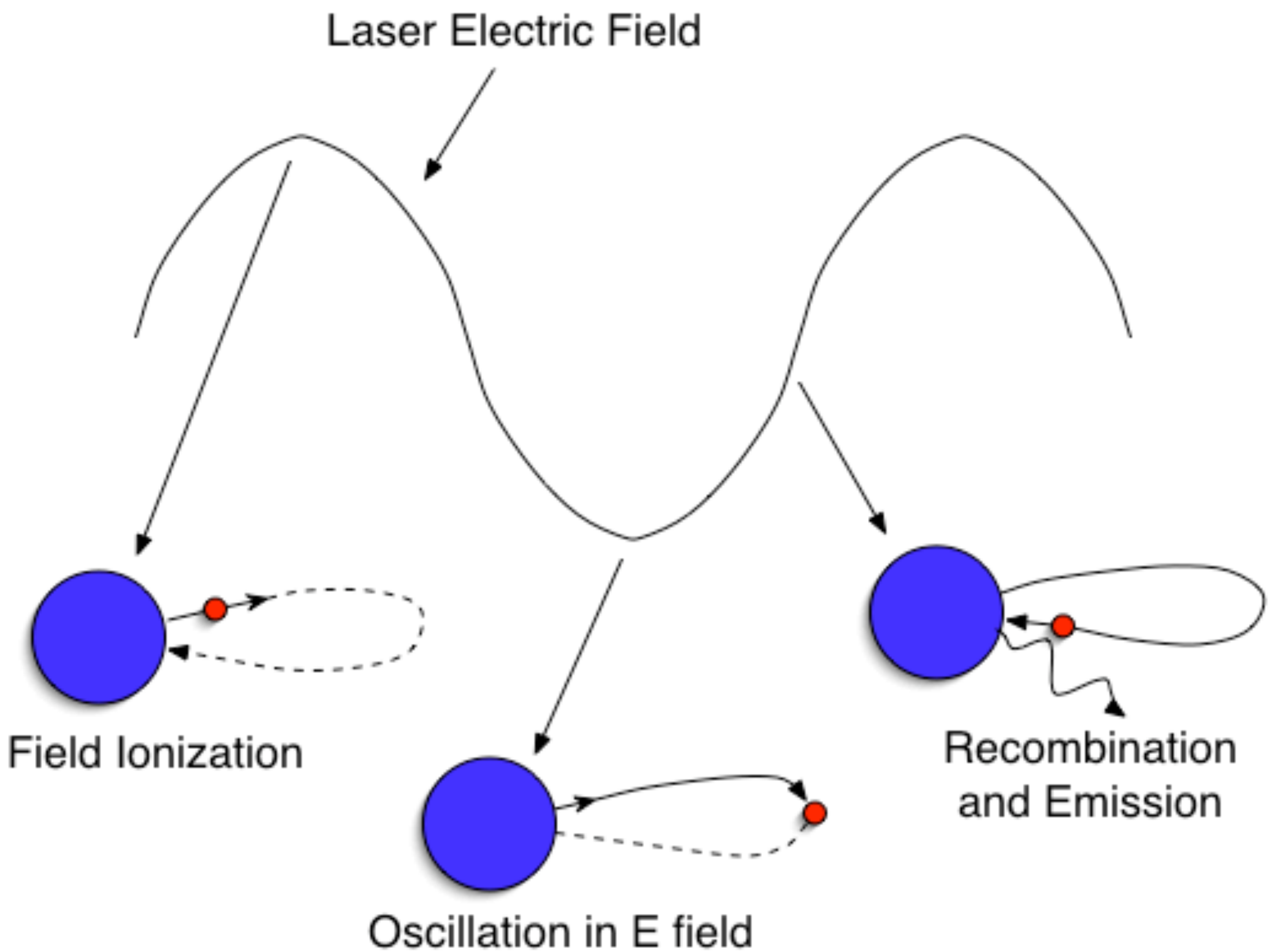


Regimes of Nonlinear Optics



T. Brabec and F. Krausz. (2000)

Corkum Model



Advantages of the Corkum Model

- Clear interpretation of the cutoff energy as the maximum energy that can be extracted from the oscillating electron in a collision

$$\hbar\omega_{c} = W_b + 3.17U_p$$

$$U_p = \frac{e^2E^2}{4m\omega^2} = \text{Pondermotive Potential}$$

$$W_b = \text{ Ionization Potential}$$

- Simplifies Calculations substantially from extremely High order quantum mechanical perturbation theory

Can rewrite the Keldysh Parameter:

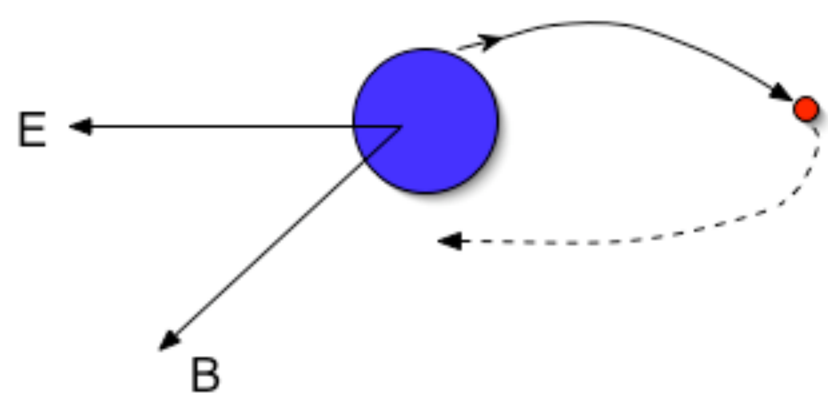
$$\frac{1}{\gamma} = \sqrt{\frac{U_p}{2W_b}}$$

So when the Pondermotive energy of a free electron exceeds the binding energy of the atom, we are in the strong field regime.

With the scaling law for the cutoff frequency, why not just go to higher laser intensity to produce an arbitrarily high pondermotive potential?

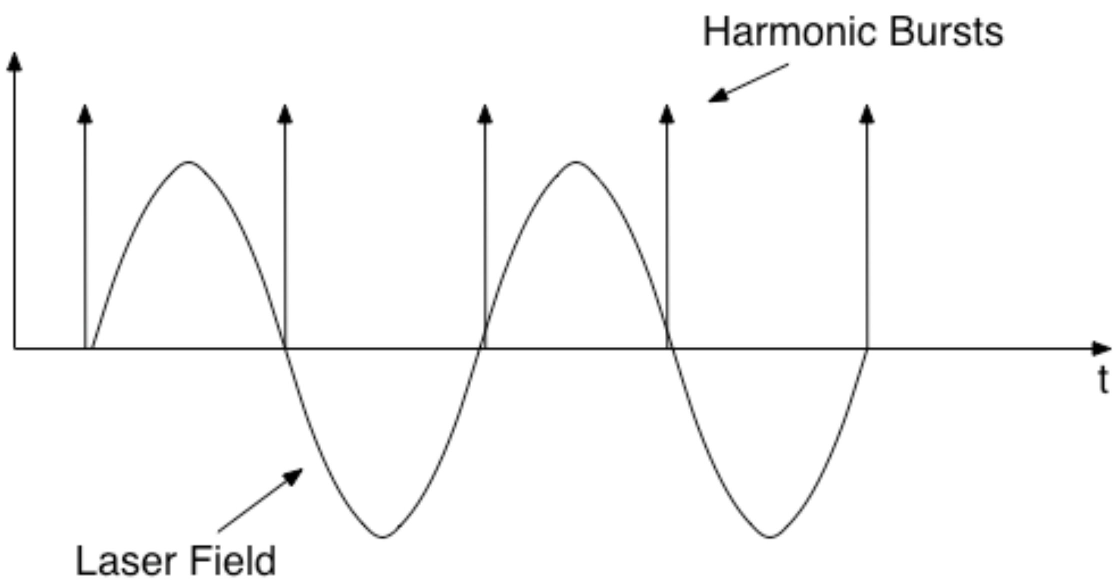
Reason: At intensities larger than ~ 10^{17} W/cm^2, the effect of the magnetic field is no longer negligible, and the electron can be diverted nm distances, missing a collision with its parent ion.

Most people use intensities in the range of 10^{14}-10^{15} W/cm^2



- Why are there only odd harmonics?

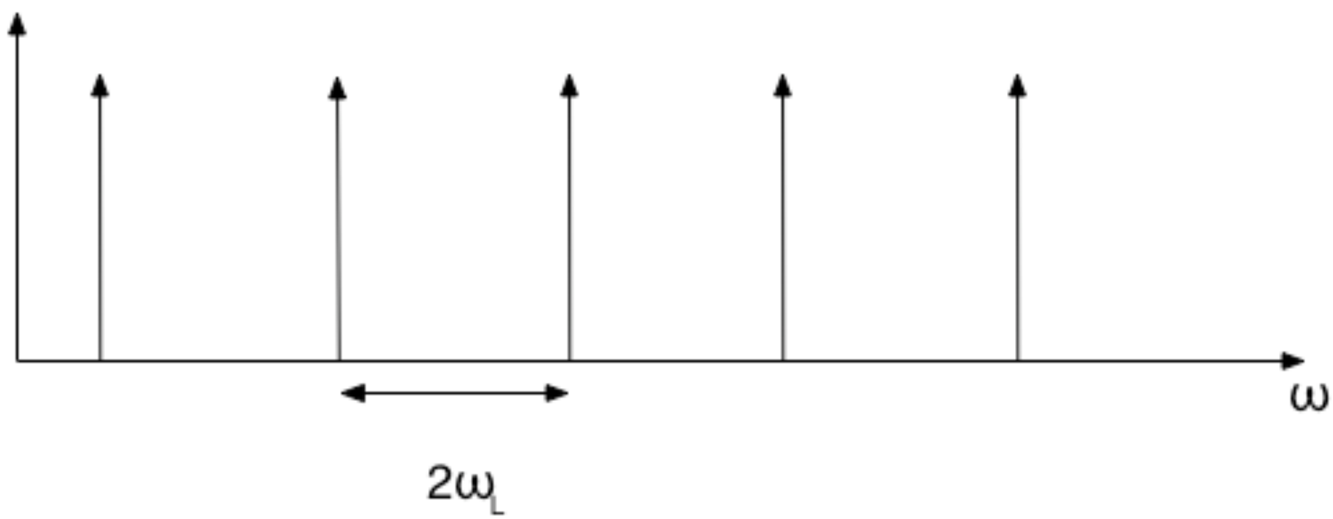
(1) Harmonics are produced by a series of sparks in time, seperated by half the laser period. We can model this in the time domain as the following train of delta-functions:



$$I_{harm}(t) = Ce^{+i\omega t} \sum_n \delta\left(t - n\frac{T_L}{2} \right)$$

Fourier Transform of “spark” train is given by a comb in freqency space:

$$I_{harm}(\omega) = 2\frac{\sqrt{2\pi}}{T_L} C \sum_n \delta\left(\omega - \left[(2n)\frac{2\pi}{T_L} + \omega_L \right] \right)$$



(2) Since a gas must possess inversion symmetry, Any induced polarization of the gas must be an odd function of the Electric field $\Rightarrow$Only odd harmonics!

Outline

1. Microscopics: basic physics of high Harmonic Generation
2. Macroscopics: optimizing harmonic generation.
3. Single attosecond pulse generation.
4. High harmonics and frontiers of physics.

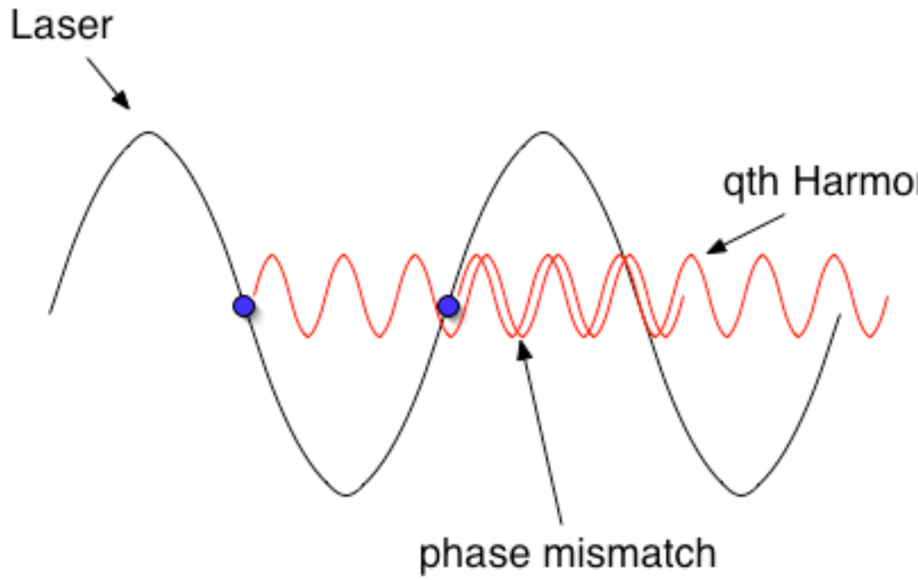
Optimization

- With an understanding of the basics of harmonic generation, how do we proceed to design high intensity, high flux soft x-ray sources
- Parameters
  - Phase Matching
  - Pulse Length
  - Gas Target (gas species? density? clusters?)

Phase Matching

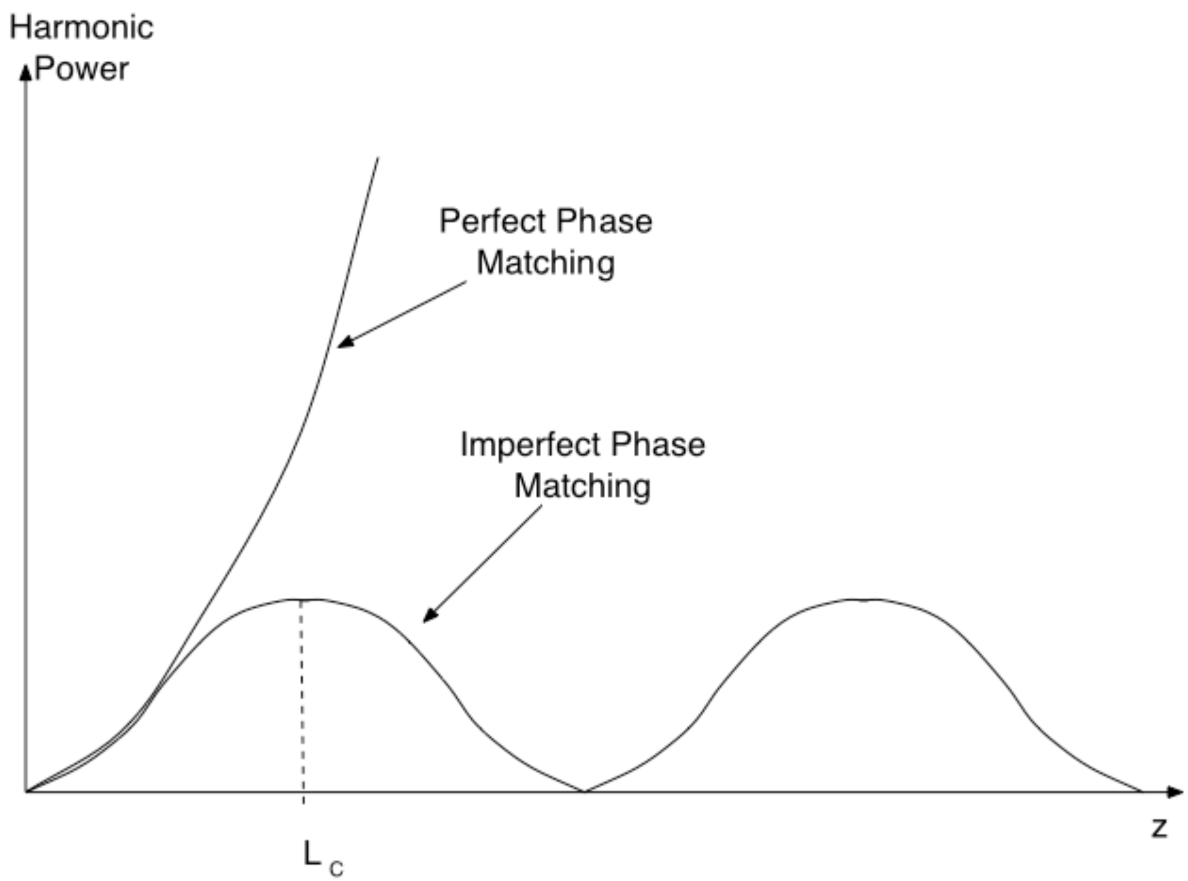
- Harmonics generated at different positions in the medium must interfere constructively in order to have the harmonic intensity build up
- Must satisfy the phase matching condition

$$\Delta k = qk_L - k_q = 0$$



Coherence Length

- If waves are not phase matched, only get coherent growth of harmonic over roughly one "coherence length" $L_c = \pi / \Delta k$



Sources of Phase Mismatch

- Dispersion. Becomes more of at higher harmonic order!

$$\Delta k = \frac{2\pi}{\lambda_0} q\left[n(\omega) - n(q\omega)\right]$$

How do I have dispersion in a dilute gas? The created plasma has a frequency dependant refractive index:

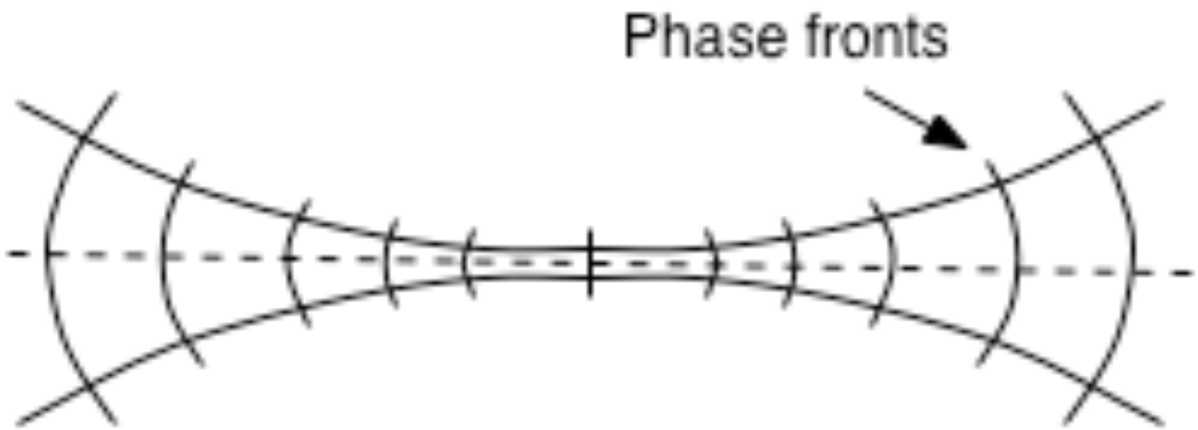
$$n(\omega) = \sqrt{1 - \frac{\omega_p^2}{\omega^2}} \approx 1 - \frac{1}{2}\frac{\omega_p^2}{\omega^2}$$

Plasma Dispersion

- For P ≈ 10 Torr, assuming a fully ionized gas, and an 800 nm laser field, the coherence length is only 44 μm for the 100th harmonics.
- This is not long enough for good conversion efficiency. We would like to be ultimately limited by re-absoprtion of x-rays by the gas. For example, for 10 Torr of Argon:

$$L_c \gg 1/\alpha_{harm} \approx 5 \text{ cm}$$

Geometrical Phase Shifts



$$\phi(z) = k_{pw} z - \tan^{-1}\left(\frac{\lambda z}{\pi w_0^2}\right) \approx \left(k_{pw} - \frac{\lambda}{\pi w_0^2}\right) z$$

$k_{pw} =$ wavevector of plane wave: $2\pi/\lambda$

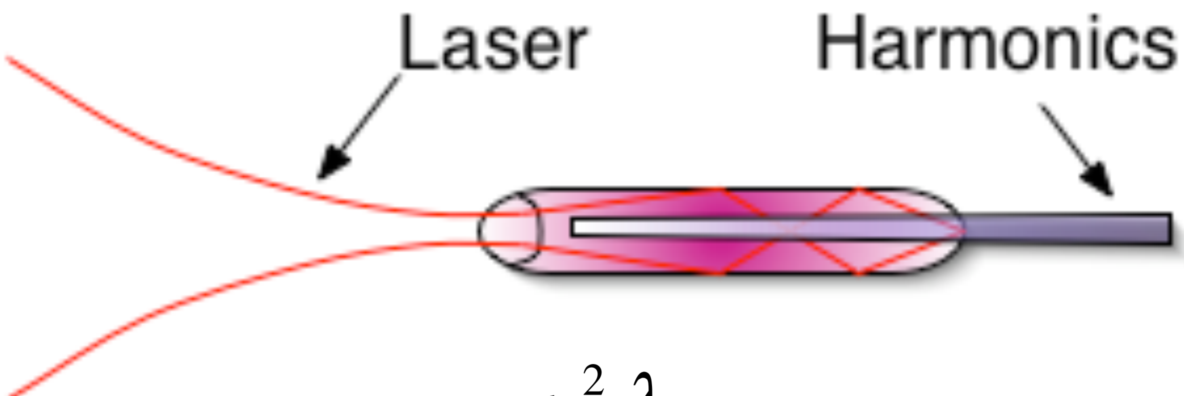
$\mathrm{w}_0 =$ Gaussian beam radius

- Can avoid this Guoy phase shift by going to larger spot size and large confocal parameter, but this requires more laser energy!

Gas Filled Waveguides

• Allow long interaction length without enormous laser energy.

• Problem: They also have a frequency dependant *k*.



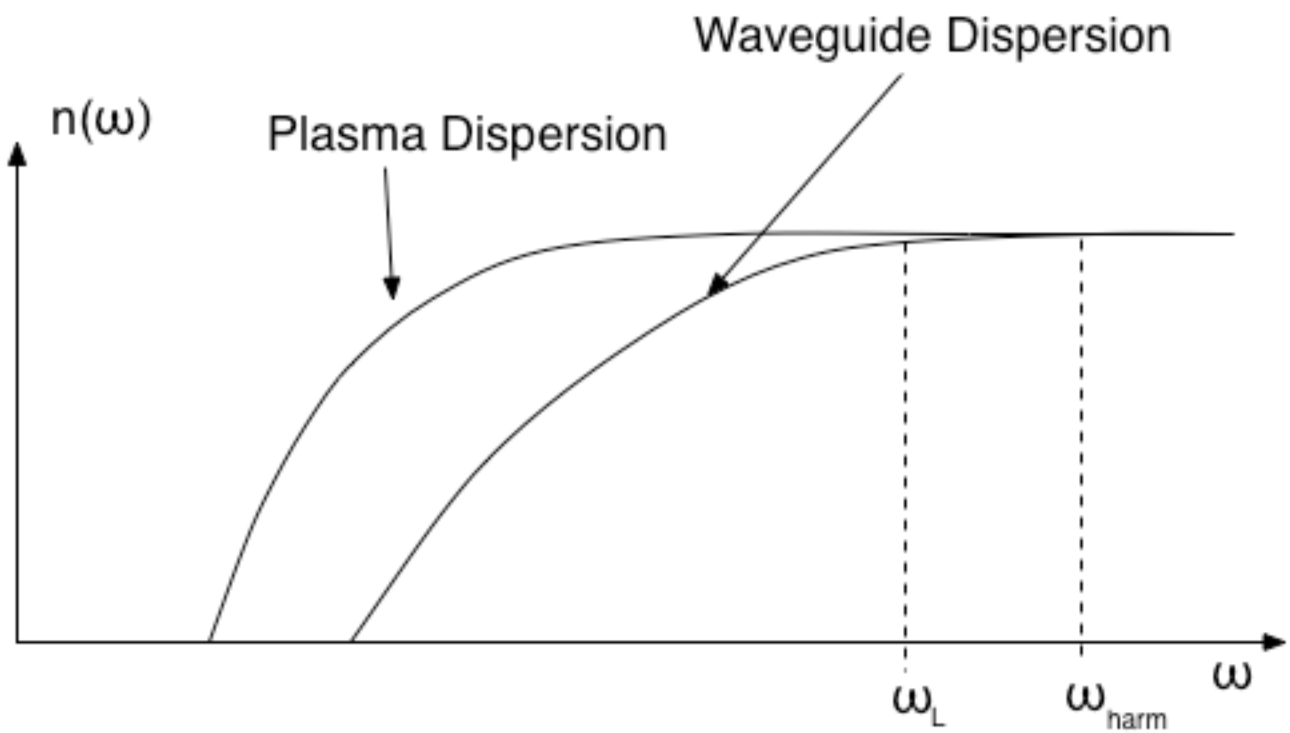
$$k_{wg} = k_{vac} - \frac{u_{11}^2 \lambda}{4\pi a^2}$$

$$u_{11} = 2.405 = \text{ First zero of } J_0$$

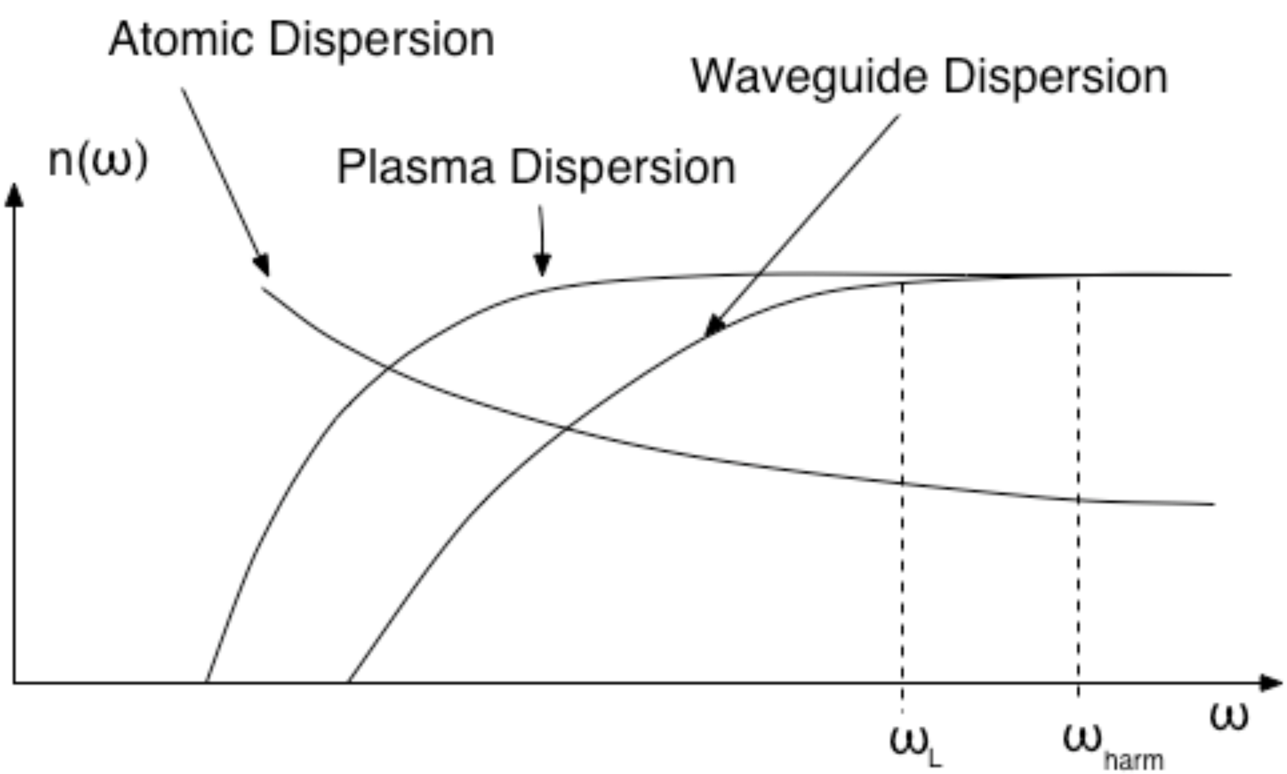
$$a = \text{ Waveguide bore radius}$$

Phase Matching

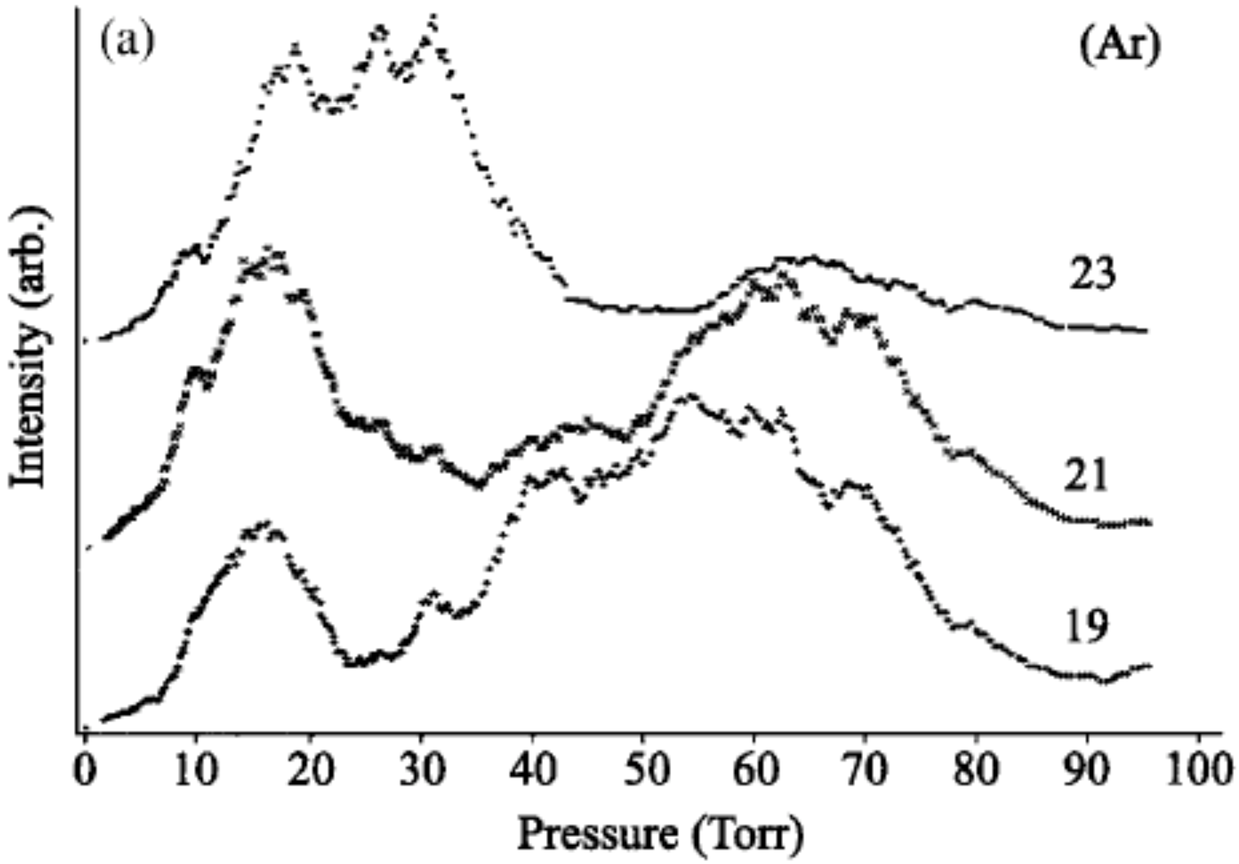
- All of the sources of phase mismatch listed above have the same sign! They are all forms of *normal* dispersion that give the laser a larger phase velocity than the harmonics



•At low intensities (~10^{14} W/cm^2), the ionization fraction of the gas is small (~1%), and the pressure in the waveguide can be tuned so that the normal dispersion of the waveguide and the plasma can be compensated for by the anamolous dispersion of the atoms.

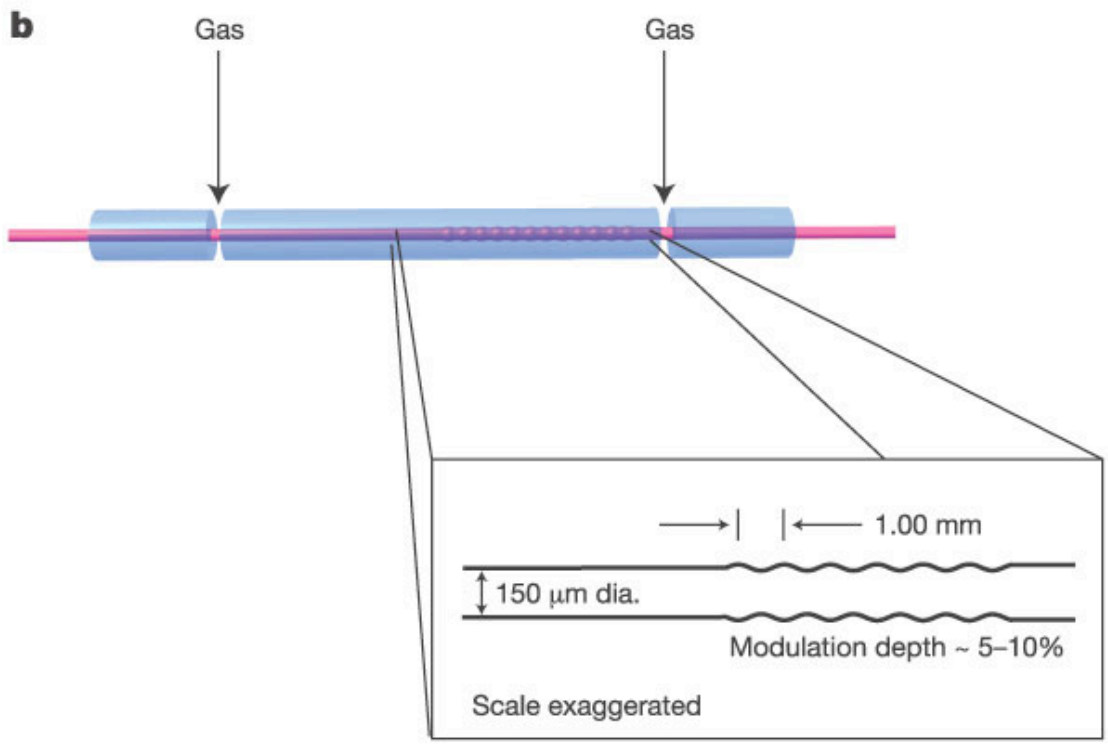


Pressure Tuning



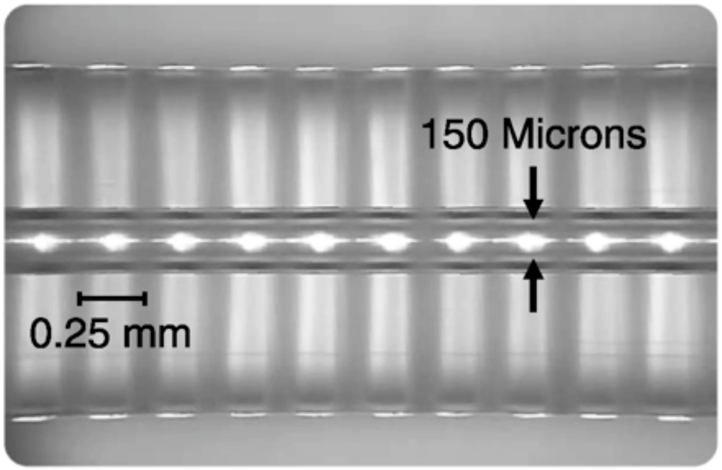
C. G. Durfee *et al.* (1999)

Quasi-Phase-Matching

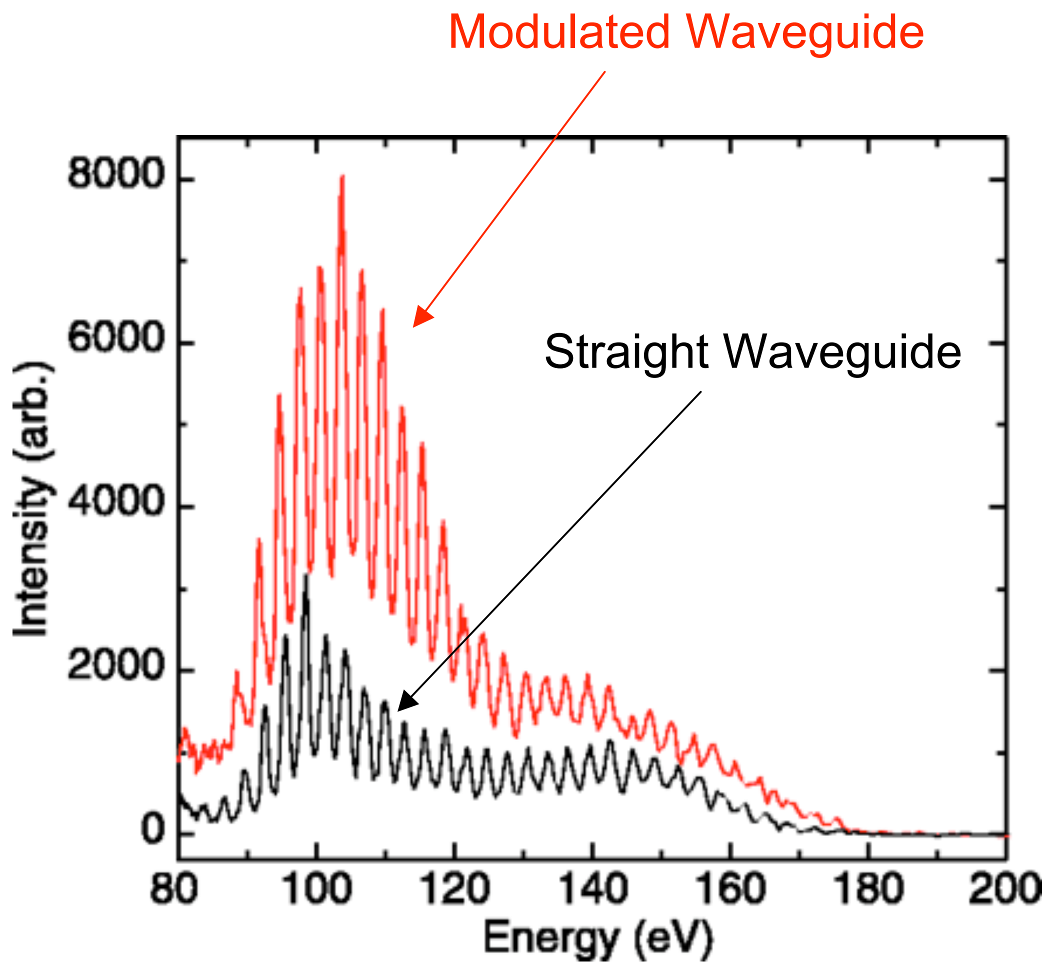


- By periodically modulating the diameter of a hollow-core fiber,The light intensity is periodically modulated.
- Since the ionized fraction of the gas depends on the light intensity, there is a periodic modulation of the index of refraction, leading to quasi-phase matching.

Quasi-Phase-Matching

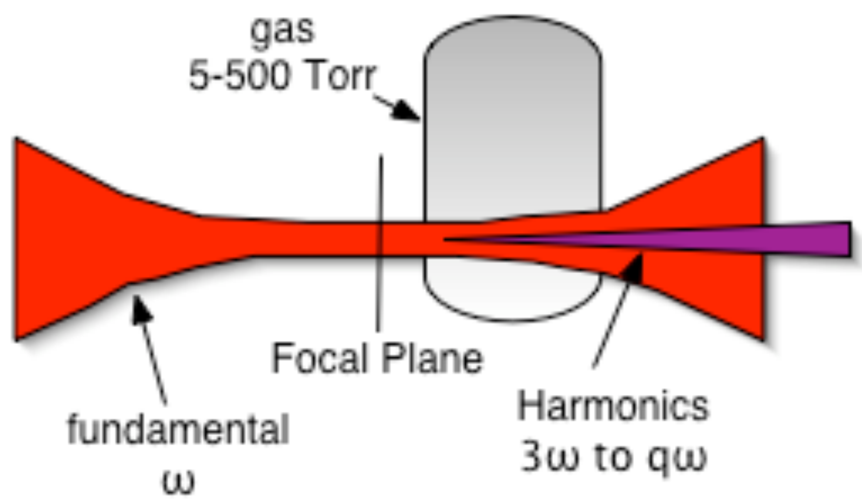


E. A. Gibson *et al. (2003)*



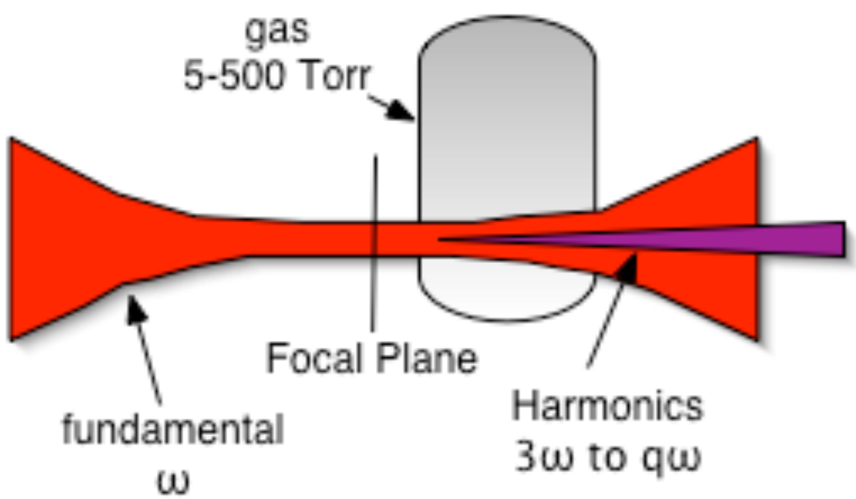
Loose Focusing Geometry

- The phase of the harmonics emitted in a collision depends sensitively on the instantaneous intensity of the field.
- By gently focusing the laser before the gas into the gas. The “atomic phase gradient” of the harmonics before the pulse can compensate for the wave-vector mismatch.
- No waveguide required!



Loose Focusing Geometry

Intensity dependant phase does not scale with Harmonic order, but effects reducing phase matching do! Not effective for really high order harmonics.

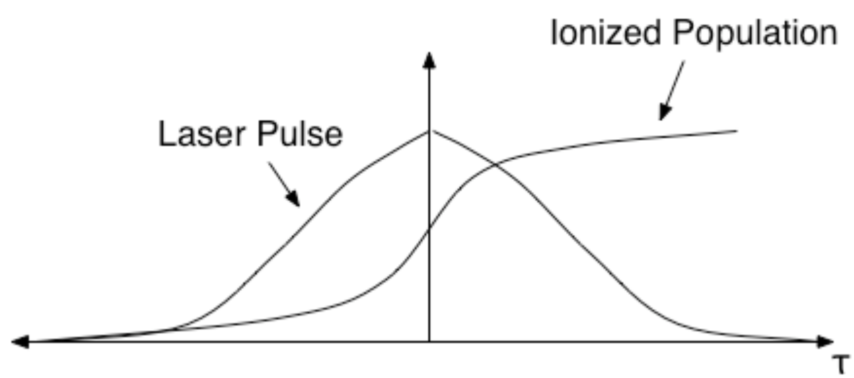


Transverse Coherence

- Sources operating with long interaction lengths produce soft x-rays with a high degree of spatial coherence, allowing focusing to high intensity.
- Intesities as high as 10^{14} W/cm^2 have been observed!
- At these intensities we can start to look for nonlinear optics in the x-ray regime.

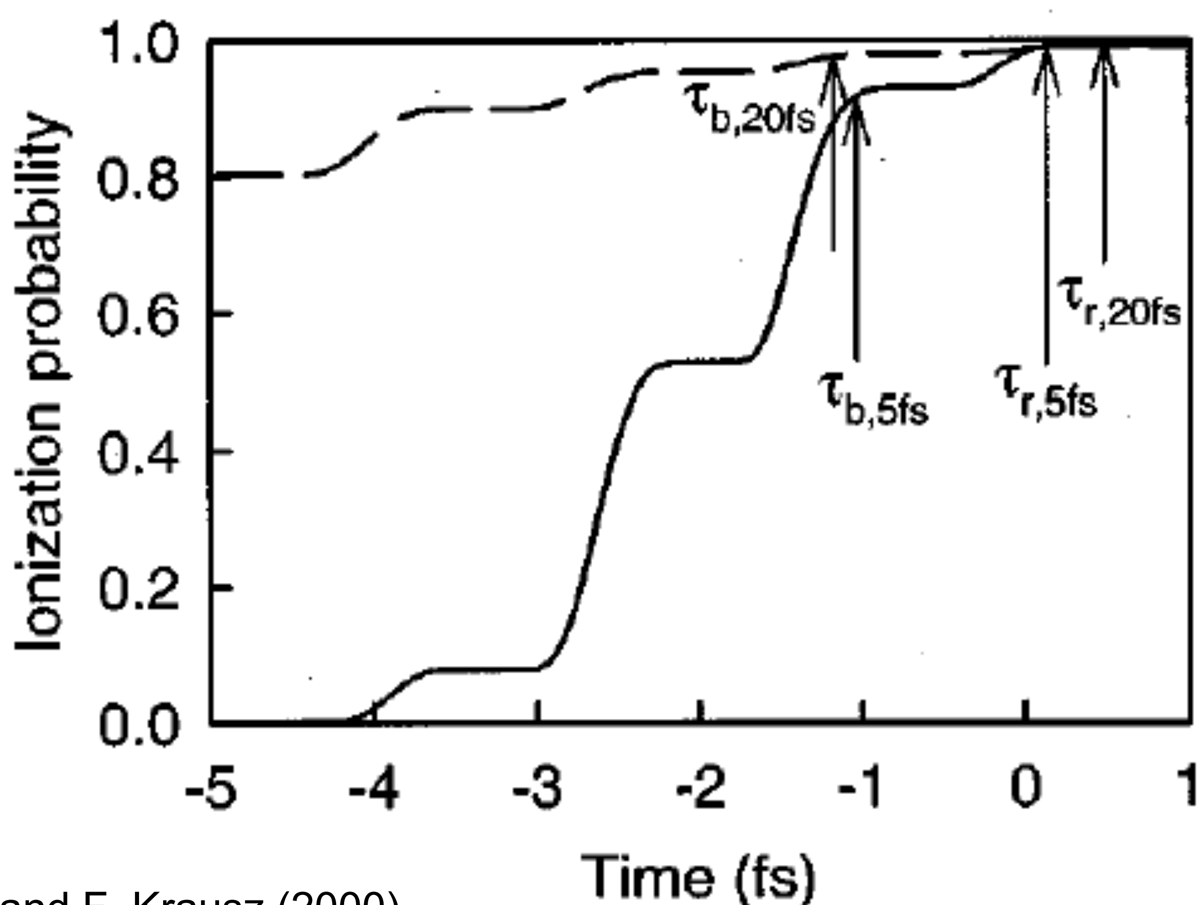
Effects of Pulse Duration

- All above phase matching schemes are effective when the fraction of the atoms ionized does not change much during one optical cycle!
- Pretty good for pulses longer than ~ 10 fs, where the ionization happens gradually over the length of the pulse.



“Non-Adiabatic” Phase Matching

For extremeley short pulses, the ionization happens rapidly in steps at the maxima of the electric field.



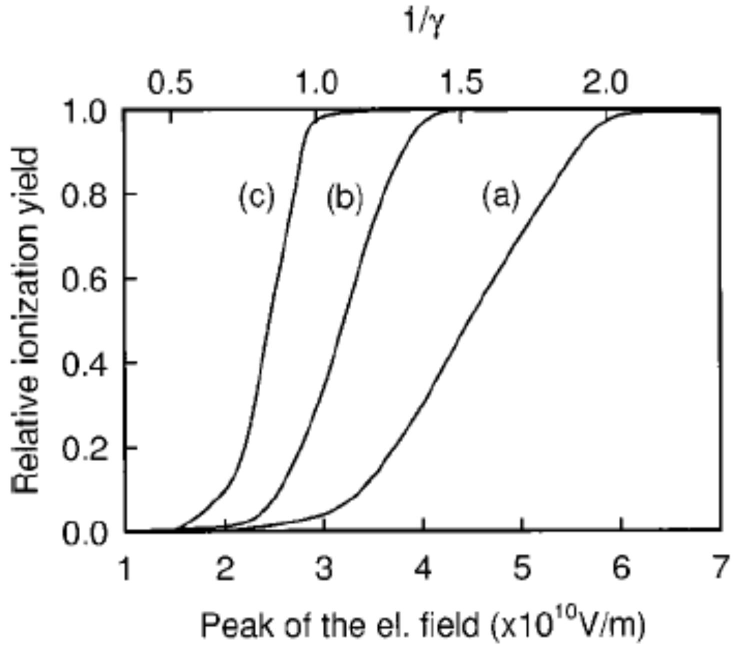
T. Brabec and F. Krausz (2000)

“Non-Adiabatic” Phase Matching

With ultrashort pulses, you can have less ionization for the same electric field.

⇒ Phase matching is much easier, because the peak of the pulse barely sees any plasma at all!

⇒ Can have absorption limited harmonic generation for very high order harmonics!



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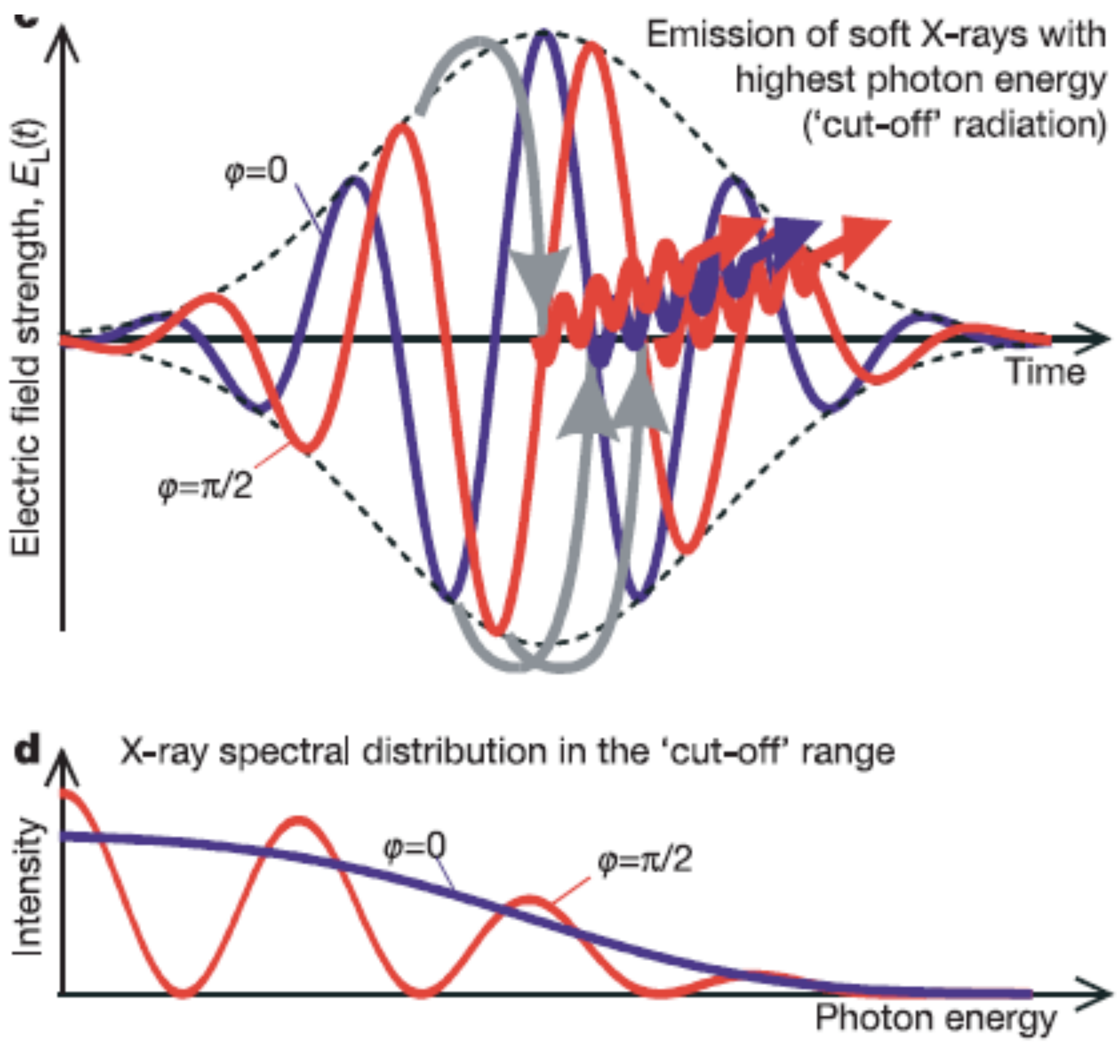
Attosecond Pulse Generation

- With pulses of ~ 5fs. The electric field makes only one or two zero crossings inside the FWHM of the pulse can provide the highest energy photons.

- Only one or two energetic collisions means the periodicty is lost in the time domain.

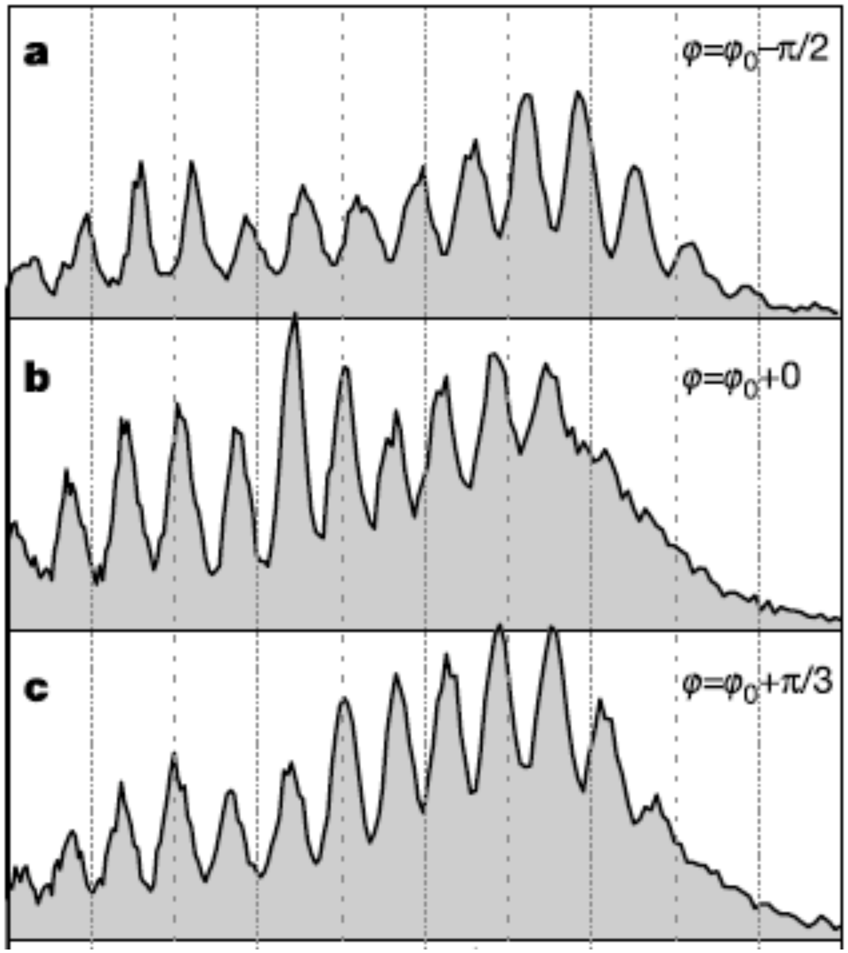
- $\Rightarrow$ Spectrum is flat! $\Rightarrow$ Attosecond Pulses!

Observation of Attosecond Pulses



A. Baltuska *et al.* Nature **421,** 611

Observation of Attosecond Pulses

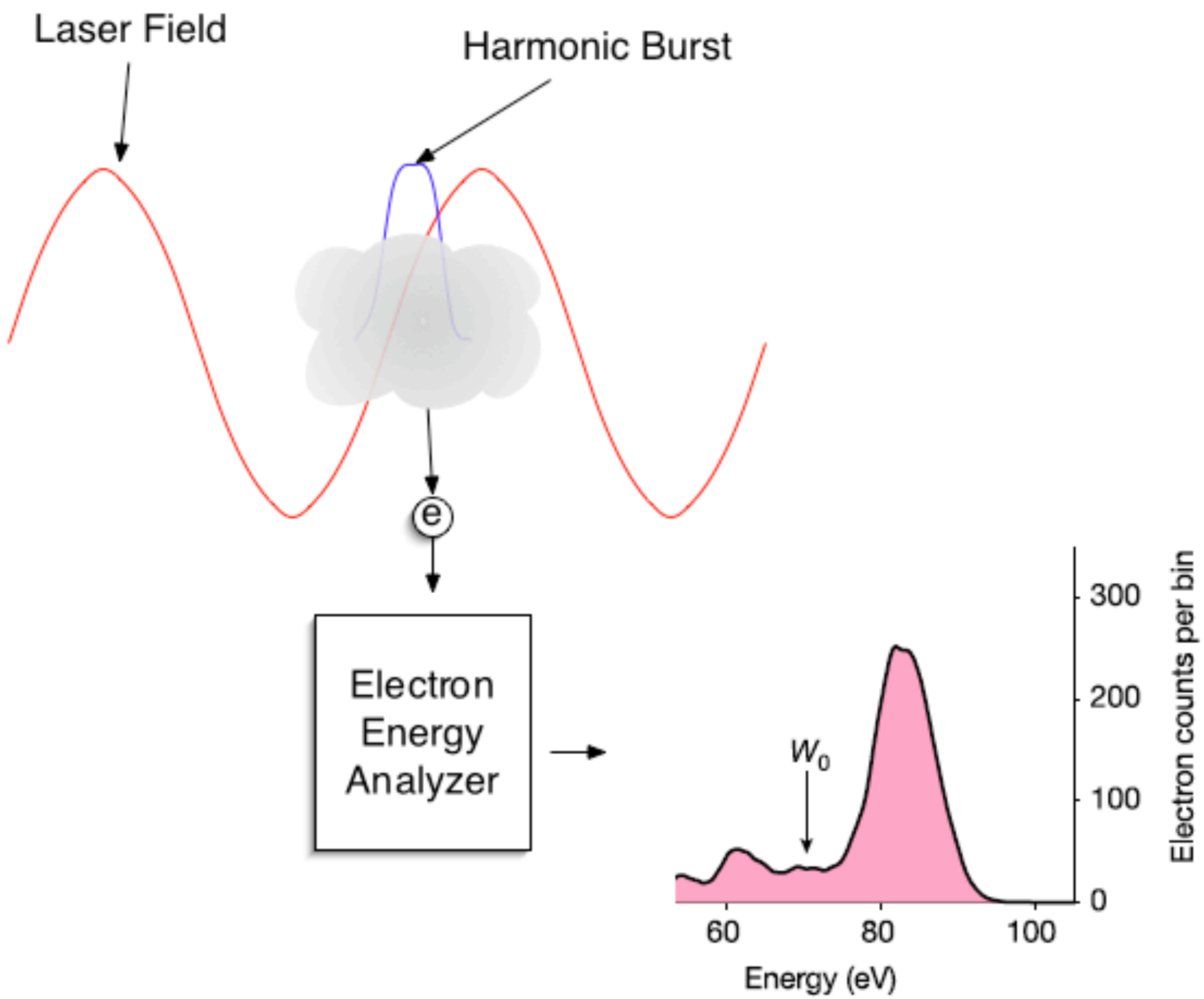


Two zero crossing, cutoff region is flat.

One zero crossing, cutoff region is flat

A. Baltuska *et al.* Nature **421,** 611

Observation of Attosecond Pulses



R. Keinberger *et al.* Nature, **427**, 817

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New Frontiers Of Physics

- Time resolved measurements in the attosecond regime.
- “Attosecond Metrology”
- Table Top High intensity coherent x-ray sources.
- X-ray nonlinear optics

Free Electron Lasers

- Free electron lasers currently being built will have intensities in the range 10^{12} - 10^{20} W/cm^2!
- Need to understand scattering and absorption phenomena of x-rays at high intensity, dynamics of Coloumb explosion, rapid ionization, etc.
- With Harmonic Sources, we can begin to explore this regime of x-ray matter interaction.

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