3rd order nonlinearities

When are 3rd order effects important?

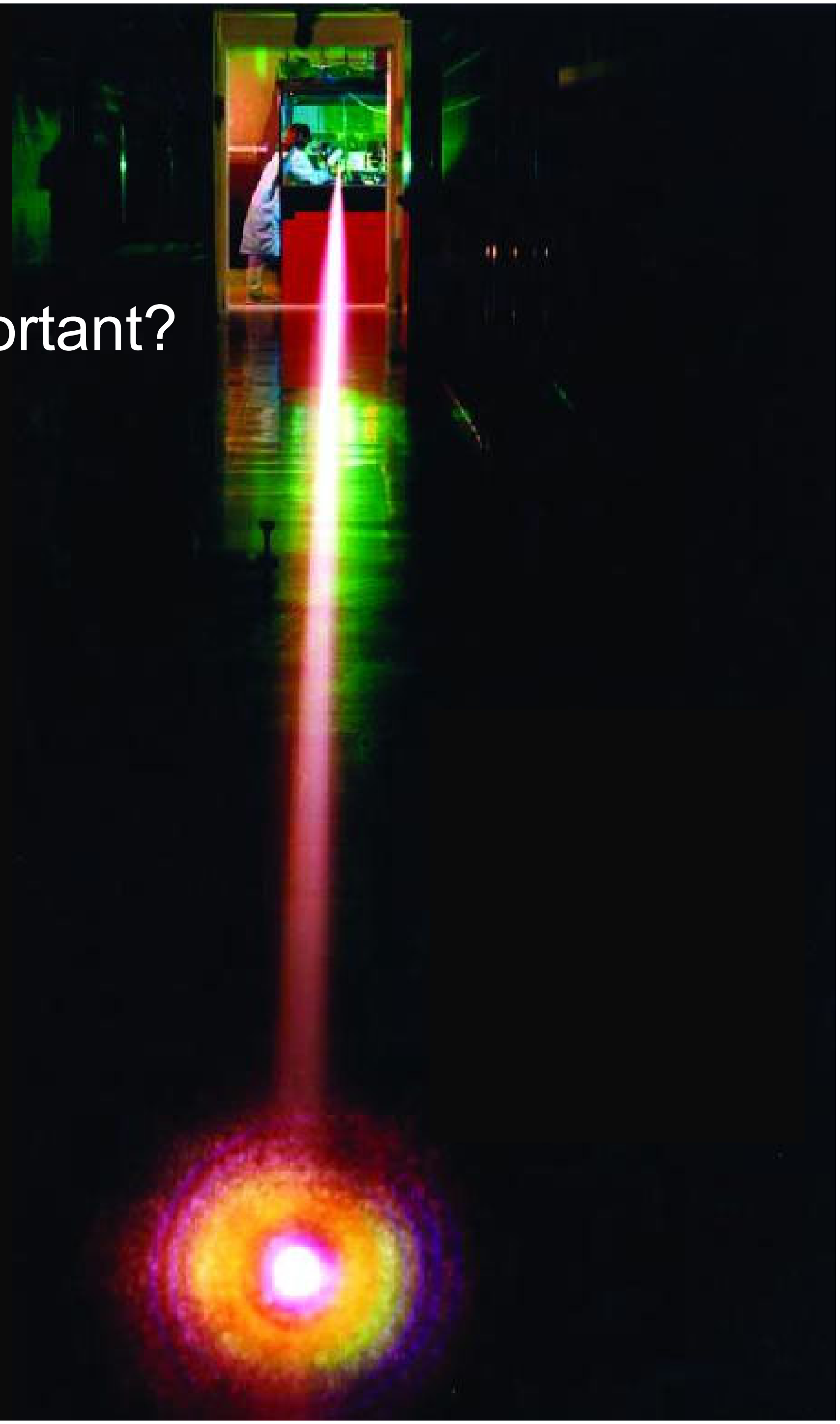
Characterizing 3rd order effects

The nonlinear refractive index

Self-lensing

Self-phase modulation

Non-perturbative effects



$\chi^{(3)}$ effects

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

Last lecture, we discussed $\chi^{(2)}$ non-linear optical effects:

- second harmonic generation
- sum frequency generation
- difference frequency generation
- optical rectification

If the power series is to converge, then $| \chi^{(3)} | << | \chi^{(2)} |$

So when are $\chi^{(3)}$ effects important?

→ when $\chi^{(2)}$ vanishes identically due to symmetry

Symmetry considerations

$$P^{(2)} = \chi^{(2)} E^2$$

Consider a medium which exhibits inversion symmetry. For example:

- many crystalline materials including all cubic crystals
- any amorphous material (glassy solid, liquid, gas)

In a material like that, reversing the sign of E must produce an induced polarization that is the same, but with opposite sign:

$$-P^{(2)} = \chi^{(2)} [-E]^2$$

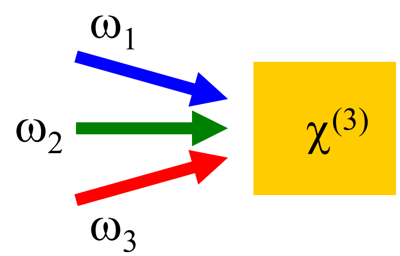
This requires that $P^{(2)} = -P^{(2)}$, which implies that $\chi^{(2)} = 0$

So: $\chi^{(2)}$ is zero most of the time. When is it not zero?

- in a material without inversion symmetry
  (e.g., KDP, BBO, $LiNbO_3$, liquid crystals, etc.)
- near any surface or interface

Third-order nonlinear effects

As with second order phenomena, we expect to find new frequency components at sum and difference frequencies:

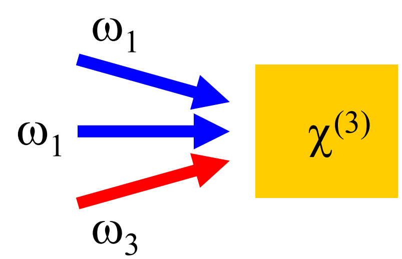


$$P^{(3)} = \varepsilon_0 \chi^{(3)} \left[E_1(t) + E_2(t) + E_3(t) \right]^3$$

$$= \varepsilon_0 \chi^{(3)} \left(E_1 e^{-i\omega_1 t} + E_1^* e^{i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_2^* e^{i\omega_2 t} + E_3 e^{-i\omega_3 t} + E_3^* e^{i\omega_3 t} \right)^3$$

Consider the case where two of the components are equal: $\omega_1 = \omega_2$

$$P^{(3)} = \varepsilon_0 \chi^{(3)} \left(E_1 e^{-i\omega_1 t} + E_1^* e^{i\omega_1 t} + E_1 e^{-i\omega_1 t} + E_1^* e^{i\omega_1 t} + E_3 e^{-i\omega_3 t} + E_3^* e^{i\omega_3 t} \right)^3$$



We can group the terms according to the frequency at which they oscillate:

$$P^{(3)} = P^{(3)}(2\omega_1 + \omega_3) + P^{(3)}(2\omega_1 - \omega_3) + P^{(3)}(\omega_3)$$

two components at shifted frequencies

one component at an **unshifted** frequency

Third-order nonlinear effects (cont.)

What if **all three of the components are equal**: $\omega_1 = \omega_2 = \omega_3$
(e.g., all from a single laser beam at frequency ω)

$$\omega \longrightarrow \boxed{\chi^{(3)}}$$

$$P^{(3)} = \varepsilon_0 \chi^{(3)} \left(E e^{-i\omega t} + E^* e^{i\omega t} \right)^3$$

$$= \varepsilon_0 \chi^{(3)} \left[E^3 e^{-3i\omega t} + \left(E^*\right)^3 e^{3i\omega t} + 3E\left|E\right|^2 e^{-i\omega t} + 3E^*\left|E\right|^2 e^{i\omega t} \right]$$

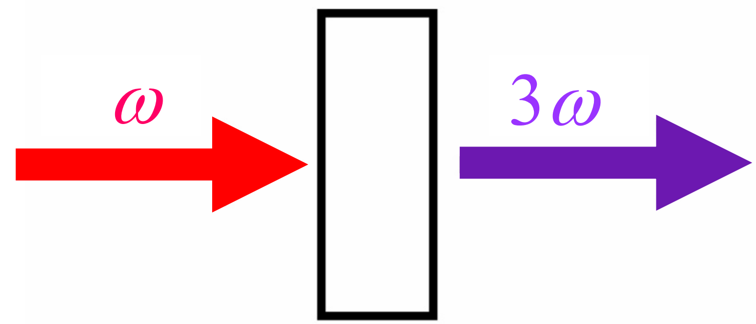
Again, we can group the terms according to frequency:

$$P^{(3)} = P^{(3)}\left(3\omega\right) + P^{(3)}\left(\omega\right)$$

again: there is a piece
at the same frequency
as the input beam

First, let's consider only the 3ω term. This is the simplest (though not the most useful) example of **four-wave mixing** (4WM).

Third harmonic generation



We must now **cube** the input field:

$$\mathscr{E}(\vec{r},t) \propto E\exp[i(\omega t - kz)] + E^* \exp[-i(\omega t - kz)]$$

$$\mathscr{E}(\vec{r},t)^3 \propto E^3 \exp[i(3\omega t - 3kz)] + E^{*3} \exp[-i(3\omega t - 3kz)]$$
$$+ \text{other terms}$$

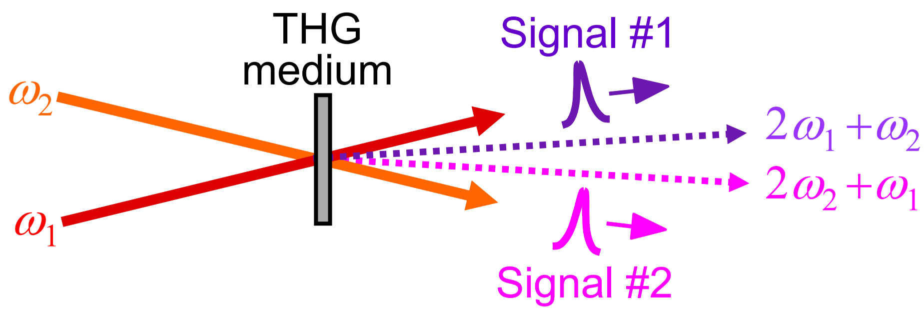
Third-harmonic generation is weaker than second-harmonic and sum-frequency generation, so the third harmonic is usually generated using SHG followed by SFG, rather than by direct THG.

Non-collinear third harmonic generation

We can also allow two different input beams, whose frequencies can be different.

So in addition to generating the third harmonic of each input beam, the medium will generate interesting sum frequencies. Note that the directions are not the same as those of either of the input beams!

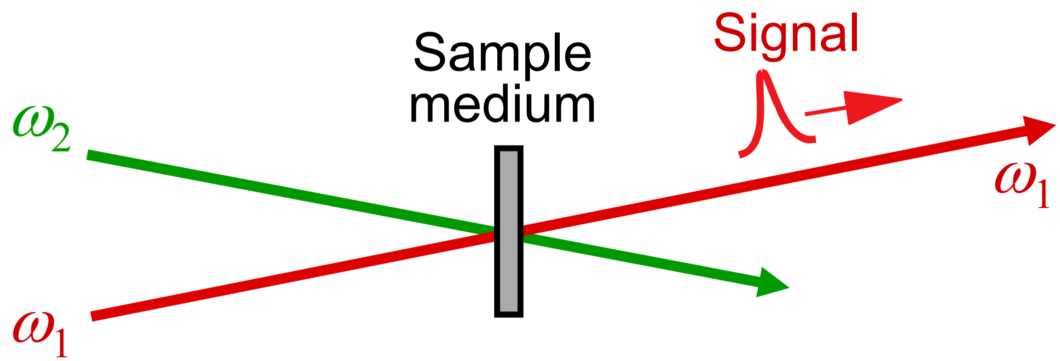
$$\mathscr{E}(\vec{r},t) \propto E_1 \exp[i(\omega_1 t - k_1 z)] + E_1{}^* \exp[-i(\omega_1 t - k_1 z)]$$
$$+ E_2 \exp[i(\omega_2 t - k_2 z)] + E_2{}^* \exp[-i(\omega_2 t - k_2 z)]$$



$$\mathscr{E}(\vec{r},t)^3 \propto E_1^2 E_2 \exp\{i[(2\omega_1 + \omega_2)t - (2\vec{k}_1 + \vec{k}_2)\cdot\vec{r})]\}$$
$$+ E_2^2 E_1 \exp\{i[(2\omega_2 + \omega_1)t - (2\vec{k}_2 + \vec{k}_1)\cdot\vec{r})]\}$$
$$+ \text{other terms}$$

The excite-probe geometry

One field can contribute two factors, one E and the other E^*. This involves both adding and subtracting the frequency and its k-vector in the phase-matching expressions.

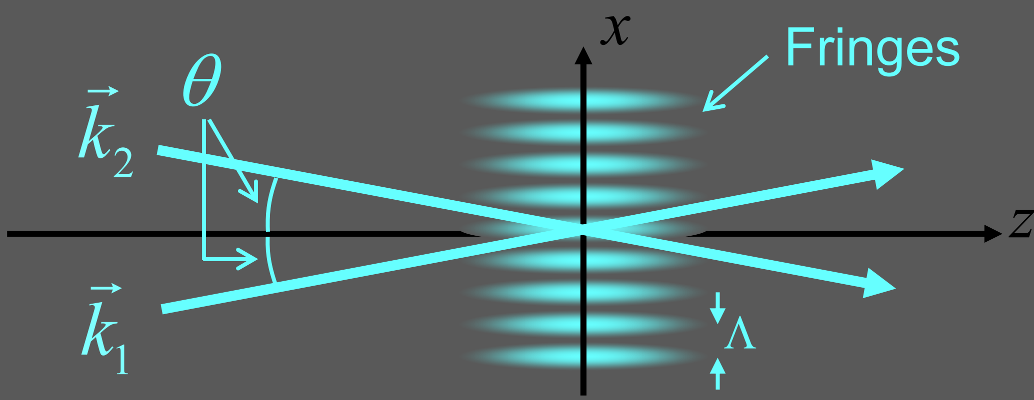


$$\mathscr{E}(\vec{r},t)^3 \propto E_1 \left|E_2\right|^2 \exp[i(\omega_1 t - \vec{k}_1 \cdot \vec{r})]$$
$$+ \text{other terms}$$

This effect is automatically phase-matched!

$$\omega_{sig} = \omega_1 - \omega_2 + \omega_2$$
$$\vec{k}_{sig} = \vec{k}_{pol} = \vec{k}_1 - \vec{k}_2 + \vec{k}_2$$

4WM as diffraction from an induced grating



$$\vec{k}_1 = k\cos\theta\,\hat{z} + k\sin\theta\,\hat{x}$$

$$\vec{k}_2 = k\cos\theta\,\hat{z} - k\sin\theta\,\hat{x}$$

Assume the beams have the same amplitudes E_0.

$$E_1 + E_2 = E_0\left\{\exp\left[i(\omega t - kz\cos\theta - kx\sin\theta)\right] + \exp\left[i(\omega t - kz\cos\theta + kx\sin\theta)\right]\right\}$$

$$= E_0\exp\left[i(\omega t - kz\cos\theta)\right]\left[\exp(-ikx\sin\theta) + \exp(+ikx\sin\theta)\right]$$

$$\boxed{\Rightarrow\quad E(x,z,t) = 2E_0\exp\left[i(\omega t - kz\cos\theta)\right]\cos(kx\sin\theta)}$$

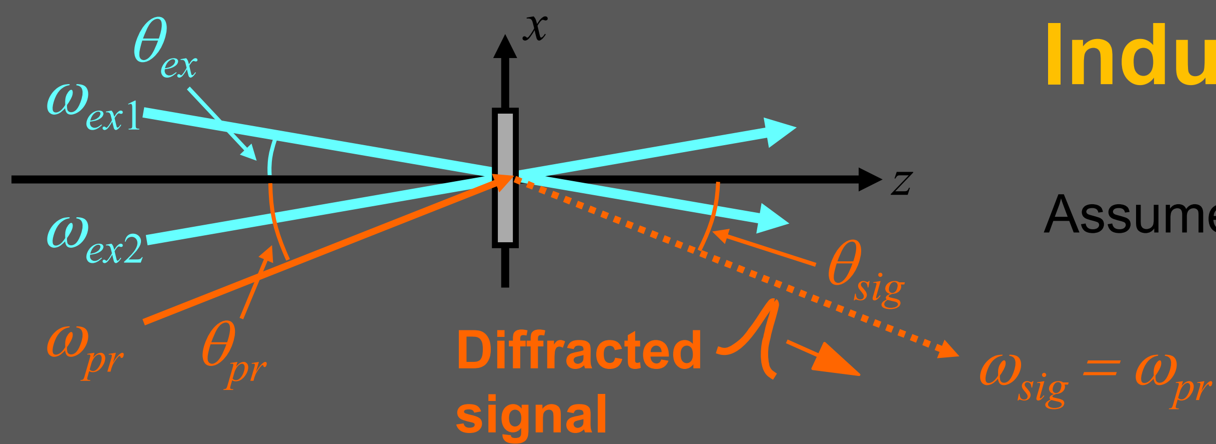
$$\boxed{\Rightarrow\quad I(x,t) = 4I_0\cos^2(kx\sin\theta) = 2I_0\left[1 + \cos(2kx\sin\theta)\right]}$$

Fringe spacing: $\Lambda = 2\pi/(2k\sin\theta)$ or $\boxed{\Lambda = \lambda/(2\sin\theta)}$

Two pulses crossing at an angle

Because the medium absorbs light at the peaks and not the troughs, its α and n will develop sinusoidal modulations: **diffraction gratings**!

Induced gratings



Assume: $\omega_{ex1} = \omega_{ex2} \equiv \omega_{ex}$

$E_{ex1} = E_{ex2} \equiv E_{ex}$

but $\vec{k}_{ex1} \neq \vec{k}_{ex2}$

$$\mathcal{E}(\vec{r},t)^3 \propto E_{pr} \left|E_{ex}\right|^2 \exp\{i[\omega_{sig} t - (\vec{k}_{ex1} - \vec{k}_{ex2} + \vec{k}_{pr}) \cdot \vec{r}]\} + \ldots$$

Phase-matching conditions: $\vec{k}_{sig} = \vec{k}_{ex1} - \vec{k}_{ex2} + \vec{k}_{pr}$

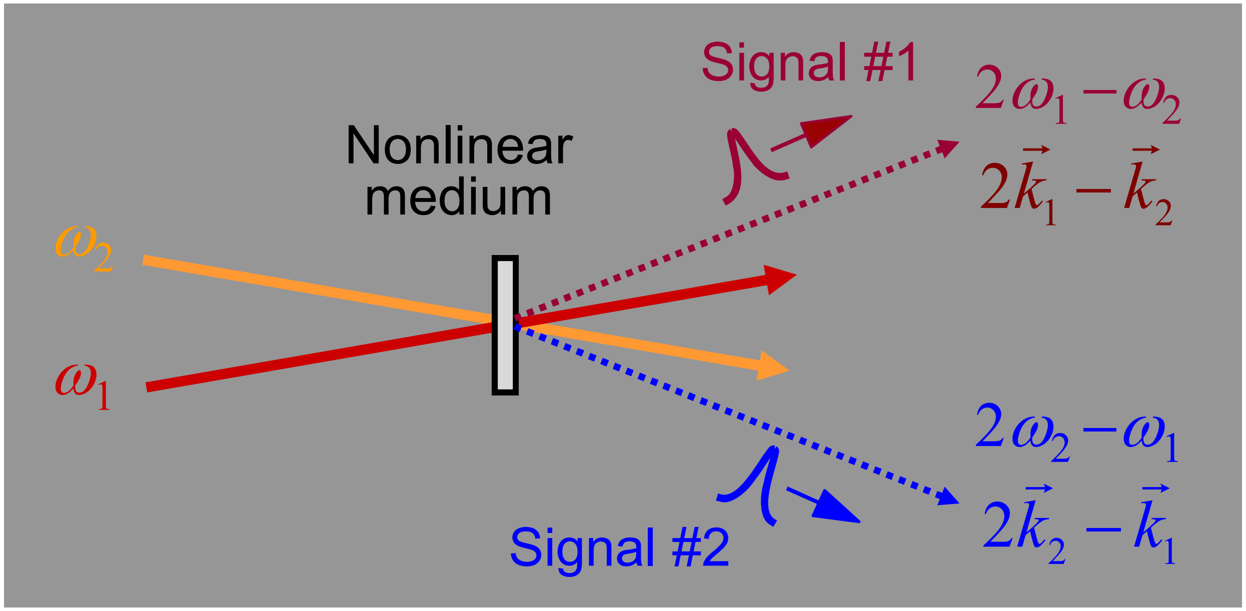
$$\omega_{sig} = \omega_{ex1} - \omega_{ex2} + \omega_{pr} = \omega_{pr} \quad \Rightarrow \quad \left|k_{sig}\right| = \left|k_{pr}\right|$$

It is not hard to show that the angle of the diffracted signal obeys:

$$k_{pr} \sin\theta_{sig} = k_{ex} \sin\theta_{ex}$$

Third-order difference-frequency generation: Self-diffraction

In self-diffraction, a beam that creates the grating also probes it:



Self-diffraction is not phase-matched.

So a very thin medium is necessary.

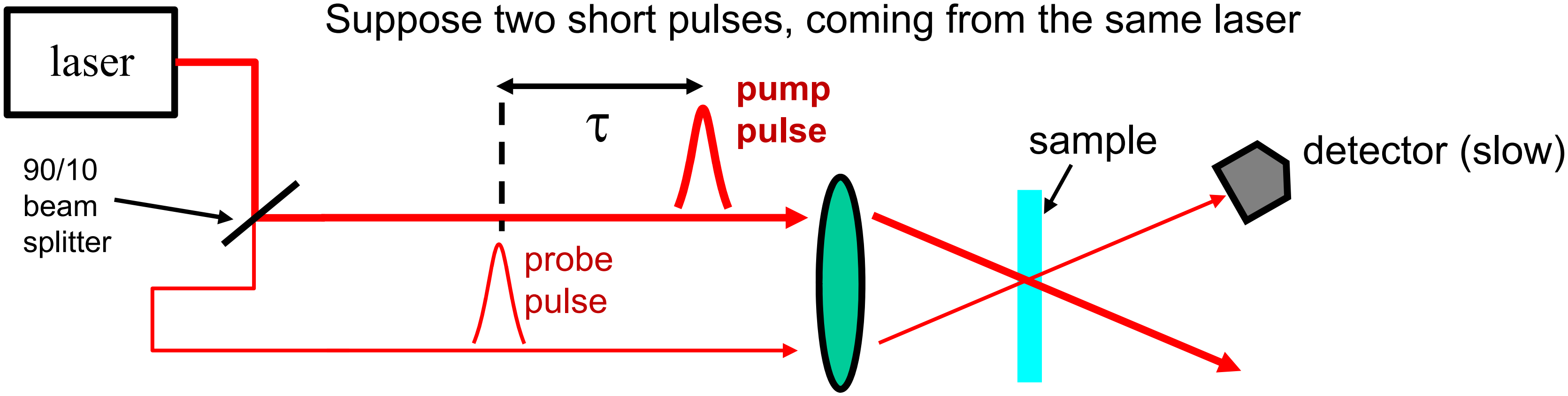
$$\mathcal{E}(\vec{r},t)^3 \propto E_1^2 E_2^* \exp\{i[(2\omega_1 - \omega_2)t - (2\vec{k}_1 - \vec{k}_2)\cdot\vec{r})]\} +$$

$$+ E_2^2 E_1^* \exp\{i[(2\omega_2 - \omega_1)t - (2\vec{k}_2 - \vec{k}_1)\cdot\vec{r})]\} +$$

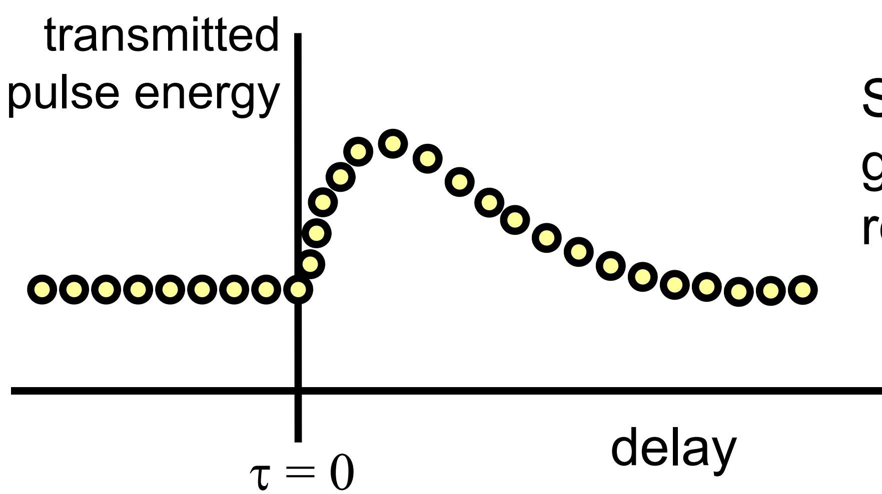
$$+ \text{other terms}$$

The pump-probe measurement

Suppose two short pulses, coming from the same laser



- A strong pump pulse perturbs the sample at t = 0.
- A time τ later, a weak probe pulse passes through the sample.
- Measure the transmission of the probe pulse
- Repeat for many different values of the delay τ.



Signal is the convolution of the gate function $I_{probe}(t)$ with the response of the medium $g(t)$:

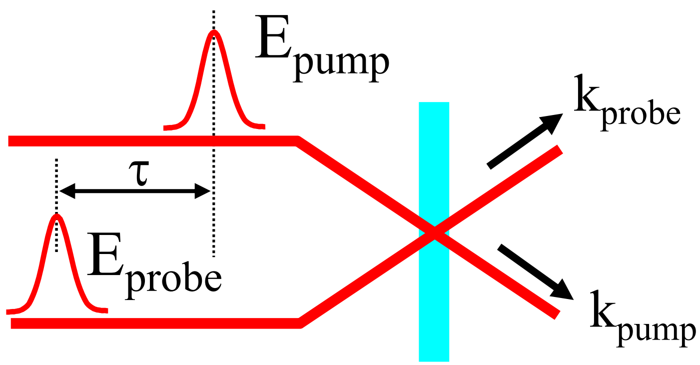
$$S(\tau) = \int_{-\infty}^{\infty} I_{probe}(t-\tau) \cdot g(t)\, dt$$

How do we know this is a 3rd order process?

As with the induced grating: consider the k dependence of the fields.

→ phase matching again, but focusing on momentum rather than energy

Suppose the two pulses are very short: $E(t) = \underbrace{E_{pump}\ \delta(t)\ e^{ik_{pump}r}}_{\text{pump pulse}} + \underbrace{E_{probe}\ \delta(t-\tau)\ e^{ik_{probe}r}}_{\text{probe pulse}} + \text{c.c.}$



- The product $E(t)^2$ has 16 terms, but none of them have the wave vector k_{probe}. So they could not contribute to the signal.
- The product $E(t)^3$ has 64 terms, containing wave vectors with all possible sums of three k vectors from k_{pump} and k_{probe}.

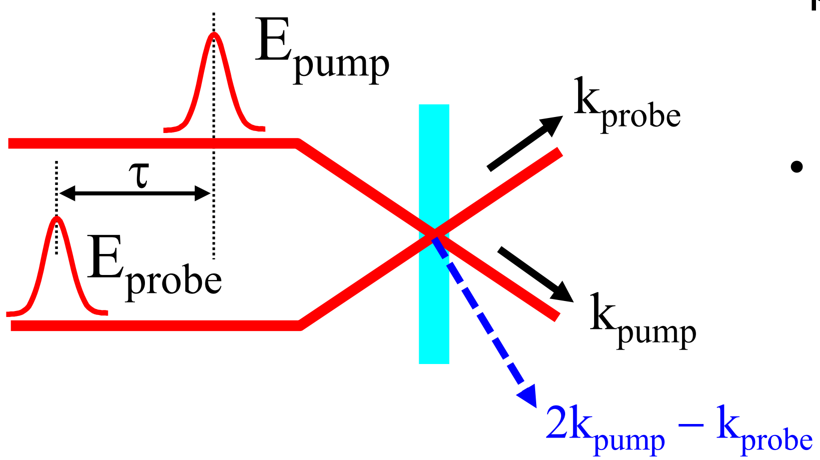
This includes the factor: $(k_{pump} + k_{probe} - k_{pump})$ which is equal to k_{probe}.

So the third-order polarization contains a term that gives rise to a signal propagating parallel to k_{probe}.

Self-diffraction with short pulses

Suppose there are two pulses:

$$E(t) = \underbrace{E_{pump}\,\delta(t)\,e^{ik_{pump}r}}_{\text{pump pulse}} + \underbrace{E_{probe}\,\delta(t-\tau)\,e^{ik_{probe}r}}_{\text{probe pulse}} + \text{c.c.}$$



- The product $E(t)^3$ has 64 terms, containing wave vectors with all possible sums of three k vectors from k_{pump} and k_{probe}.

Notice: another subset of the 64 terms is proportional to this wave vector:

$$(k_{pump} - k_{probe} + k_{pump})$$

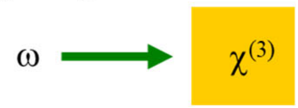
which is not parallel to either k_{pump} or k_{probe}. This term gives rise to radiation that emerges from the sample in a different direction!

Depending on the details, this is known as a “photon echo”, or a “free induction decay”, or a “transient grating”.

Third-order nonlinear effects (revisited)

Returning back to this:

What if **all three of the components are equal**: $\omega_1 = \omega_2 = \omega_3$
(e.g., all from a single laser beam at frequency ω)



$$P^{(3)} = \varepsilon_0 \chi^{(3)} \left(E e^{-i\omega t} + E^* e^{i\omega t} \right)^3$$

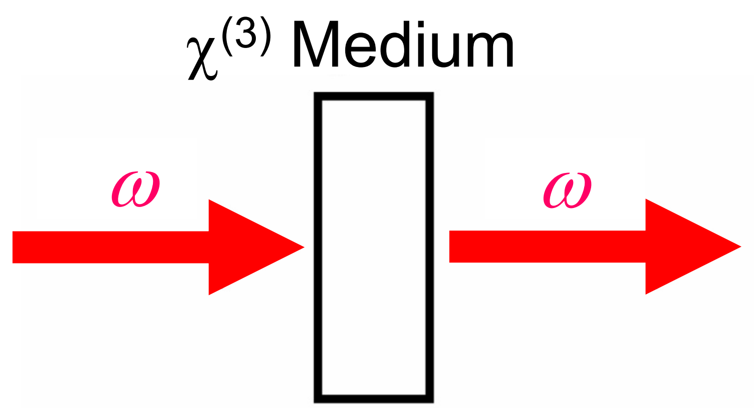
$$= \varepsilon_0 \chi^{(3)} \left[E^3 e^{-3i\omega t} + \left(E^*\right)^3 e^{3i\omega t} + 3E|E|^2 e^{-i\omega t} + 3E^* |E|^2 e^{i\omega t} \right]$$

Again, we can group the terms according to frequency:

$$P^{(3)} = P^{(3)}(3\omega) + P^{(3)}(\omega)$$

again: a piece at the same frequency as the input beam

Let's now focus on the term that varies at frequency ω.



Degenerate third-order nonlinear effects

Considering just the unshifted polarization component (and assuming that $\chi^{(2)} = 0$), the total polarization (up to 3rd order) is:

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

We can define an *effective* susceptibility χ_{eff}, such that $P^{TOT} = \varepsilon_0 \chi_{eff} E$

$$\chi_{eff} = \chi^{(1)} + 3\chi^{(3)} \left| E(\omega) \right|^2$$

Then, the *effective index* of the medium is: $n_{eff} = \sqrt{1 + \chi_{eff}}$

$$n_{eff} = \sqrt{1 + \chi^{(1)} + 3\chi^{(3)} \left| E(\omega) \right|^2} = \sqrt{\left(1 + \chi^{(1)}\right)\left(1 + \frac{3\chi^{(3)}}{1 + \chi^{(1)}} \left| E(\omega) \right|^2\right)}$$

But $n_0 = \sqrt{1 + \chi^{(1)}}$ is the usual low-intensity refractive index.

$$n_{eff} \approx n_0 + \frac{3\chi^{(3)}}{2n_0} \left| E(\omega) \right|^2$$ = n_0 + a small intensity-dependent correction

Refractive index depends on intensity

$$n \approx n_0 + \frac{3\chi^{(3)}}{2n_0}\left|E(\omega)\right|^2$$

Interpretation: the refractive index of a $\chi^{(3)}$ medium has an intensity-dependent term. This is usually written:

$$\boxed{n = n_0 + n_2 I} \qquad \text{where } \; n_2 \propto \chi^{(3)}$$

n_2 has units of inverse intensity, or m^2/ Watt. It is usually very small.

Typical numbers for n_2:

air	4×10^{-19} cm^2/W
glass	2.7×10^{-16} cm^2/W

If the incident radiation is very intense (i.e., approaching $1/n_2$), then the index of the medium changes in response to the light field.

This can lead to self-induced effects.

This is known as the "optical Kerr effect".

How realistic is it to get to these intensities?

For silica glass: $n_2 = 2.7\times10^{-20}$ m^2/W (at $\lambda = 1.5$ μm)

So the interesting intensity range is when I approaches $1/n_2 = 3.7\times10^{19}$ W/m^2.

Suppose our light is focused to a spot size of 10 microns.

Then: area = $\pi R^2 = 8\times10^{-11}$ m^2

necessary power = $I \times$ area = 3×10^9 W

Suppose our pulse duration is τ_P = 100 femtoseconds.

Then:

necessary pulse energy = power $\times\ \tau_P$ = 0.3 millijoules

Thus: focusing a pulse with an energy of 300 microjoules and a duration of 100 femtoseconds gives $n_2 I \sim 1$ in glass.

In that case, the refractive index is changed by ~100% at the peak of the pulse. If the pulse energy is only 3 microjoules (instead of 300), this still causes a ~1% change in the index (which is not a trivially small change!)

These pulse energies are readily achievable using femtosecond lasers.

Intensity-dependent index

Conclusion:
An intense light field changes the refractive index of the medium in which the light is propagating.

This modified refractive index can in turn change the characteristics of the light field that caused the change.

$\longrightarrow$ self-induced effects are possible

If the material response is slow, then the effect is not observable with very short light pulses. But if the response is fast, then it can have dramatic effects on short light pulses.

Mechanisms for intensity-dependent index

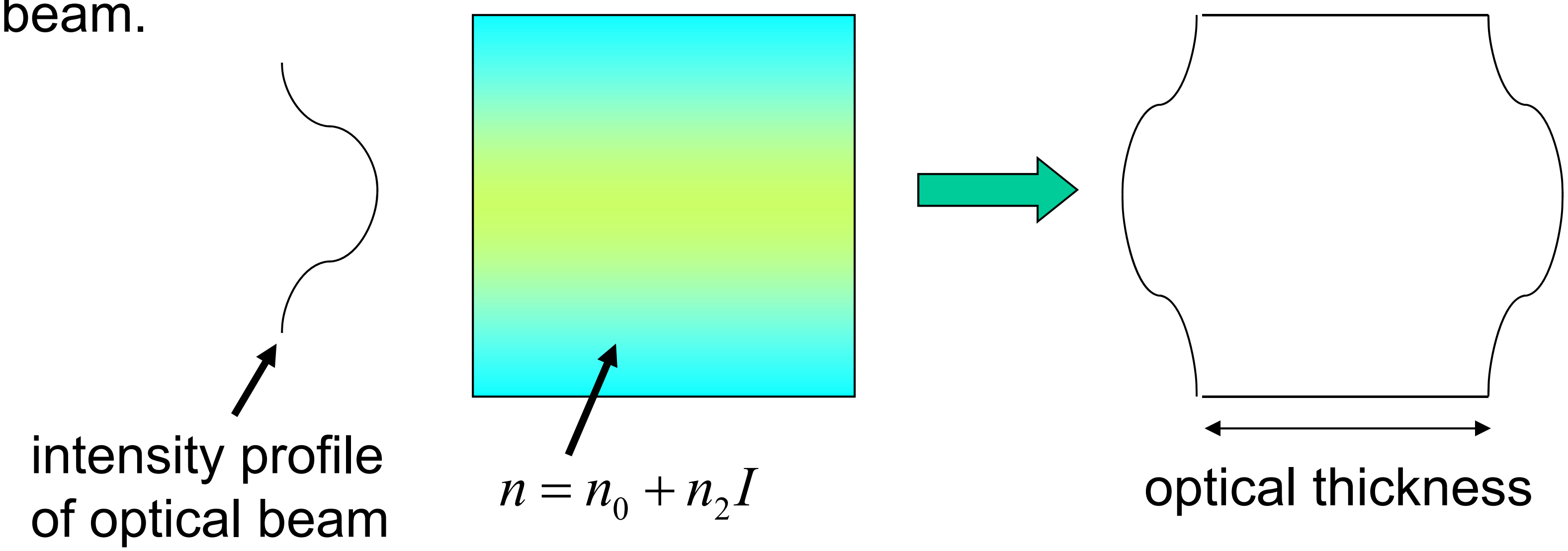
Different nonlinear mechanisms can manifest in very different regimes of intensity and time response.

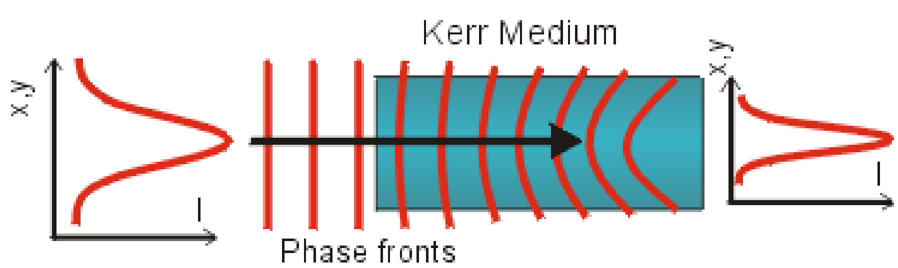
Mechanism	n_2 (cm^2/W)	$\chi^{(3)}$ (esu)	Response time (sec)
electronic hyperpolarization	10^{-16}	10^{-14}	10^{-15}
molecular orientation	10^{-14}	10^{-12}	10^{-12}
electrostriction	10^{-14}	10^{-12}	10^{-9}
saturated atomic absorption	10^{-10}	10^{-8}	10^{-8}
thermal effects	10^{-6}	10^{-4}	10^{-3}
photorefractive effect	(large)	(large)	(intensity-dependent)

Typically: the larger the effect, the slower the material response!

One manifestation of the optical Kerr effect: self-lensing

The intensity-dependent refractive index means that the center of a Gaussian beam sees a different refractive index from the edges of the beam.





A planar material can act like a lens!

Self-lensing and the formation of filaments

Suppose the material in question is air.

Self-focusing leads to an ever-increasing intensity at the center of the laser beam. Eventually, the intensity is high enough to ionize atoms.

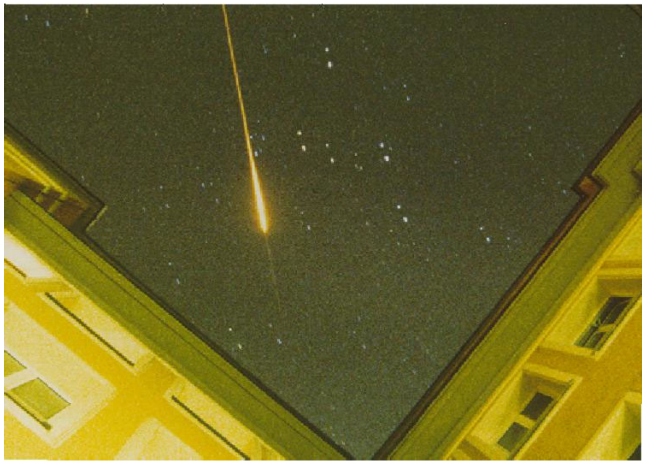
In air, ionizing atoms produces a plasma. This plasma then contributes to the refractive index. Plasmas have a refractive index which is less than that of air, so this reduces the index at the center of the beam, leading to de-focusing.

If these two contributions offset, then a stable filament is formed. This filament can propagate for many meters!

Optical filaments - the Teramobile

A stable filament in air acts as a conductive channel, which is essentially a lightning rod. This can be used as a mobile lightning protection system.

Teramobile



a self-guided filament induced in air by a high-power, infrared (800 nm) laser pulse

guided and unguided lightning

Another manifestation of the optical Kerr effect: self-phase modulation

As a light beam propagates a distance z in a medium, it acquires a phase:

$$\phi = \omega t - kz = \omega t - \frac{n\omega}{c} z$$

Optical Kerr effect: the refractive index depends on intensity.

$$\phi = \omega t - \left(n_0 + n_2 I\right)\frac{\omega}{c} z$$

Instantaneous frequency is equal to the time derivative of the temporal phase:

$$\omega_{inst} = \frac{d\phi}{dt} = \omega - \frac{n_2 \omega z}{c}\frac{dI(t)}{dt}$$

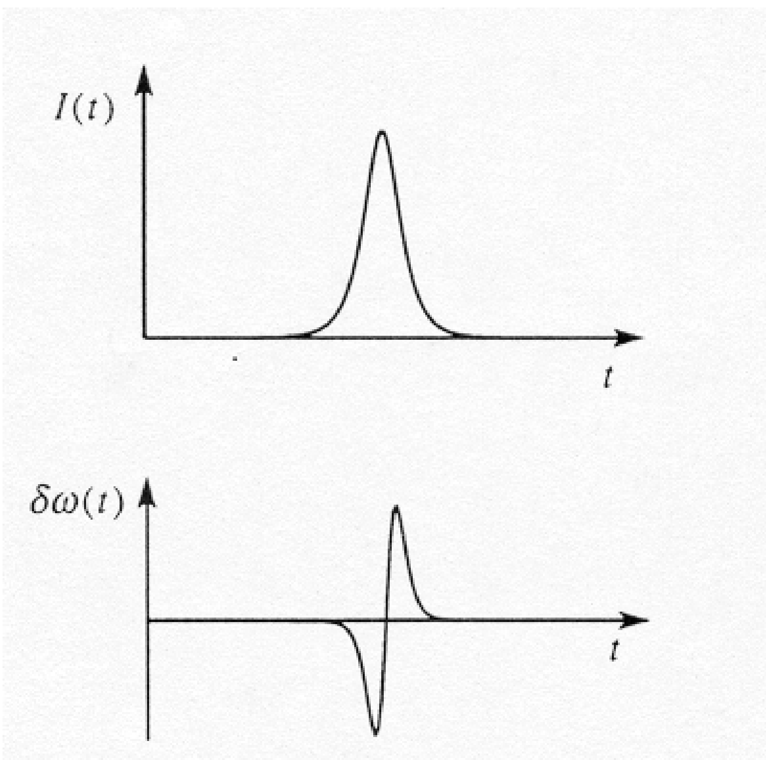
If intensity depends on time, then the pulse frequency changes with time!

"self-phase modulation"

Self-phase modulation

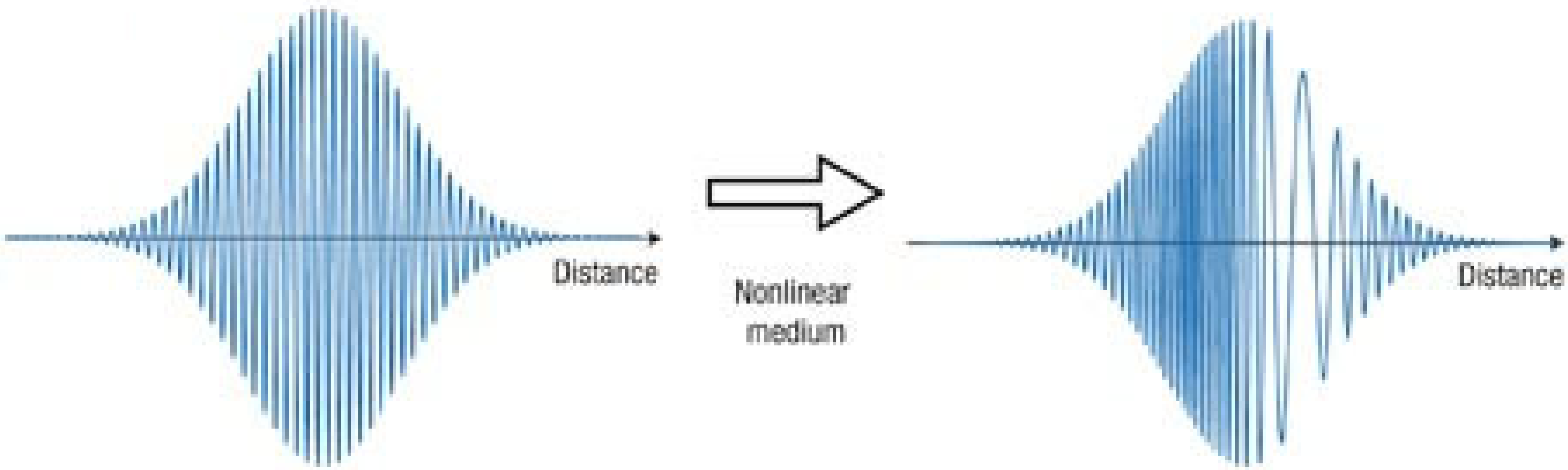
The nonlinear phase gives rise to an instantaneous frequency which depends on time: $\omega(t) = \omega_0 - \delta\omega(t)$

where: $$\delta\omega(t) = \frac{d}{dt}\phi_{NL} = n_2 \frac{\omega_0 l}{c} \frac{dI(t)}{dt}$$



If the light is a pulse, then the instantaneous frequency is first smaller than, and then larger than, the central frequency ω_0.

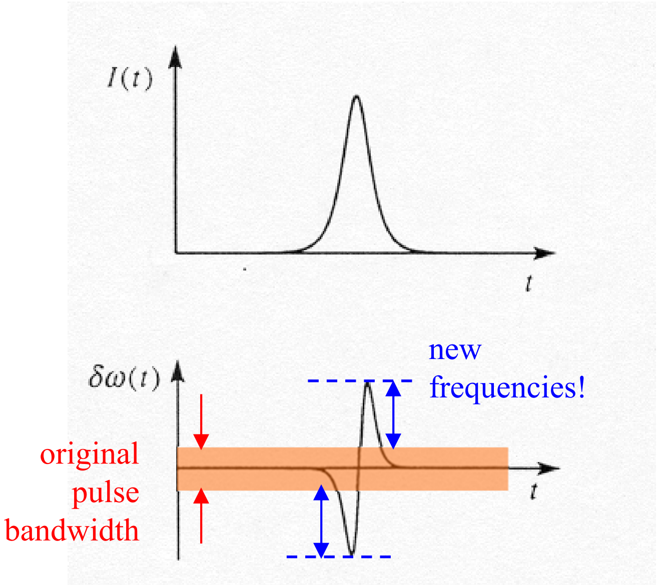
Self-phase modulation: ω depends on t



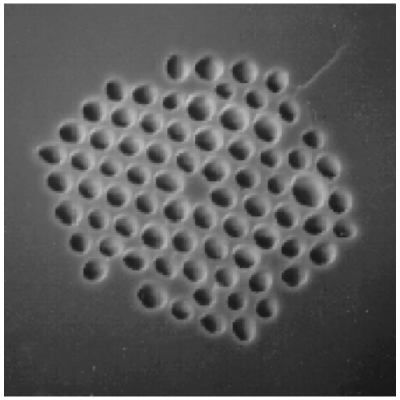
This can be extremely dramatic if the excursions of $\omega(t)$ away from its original value are large.

Self-phase modulation: spectral broadening

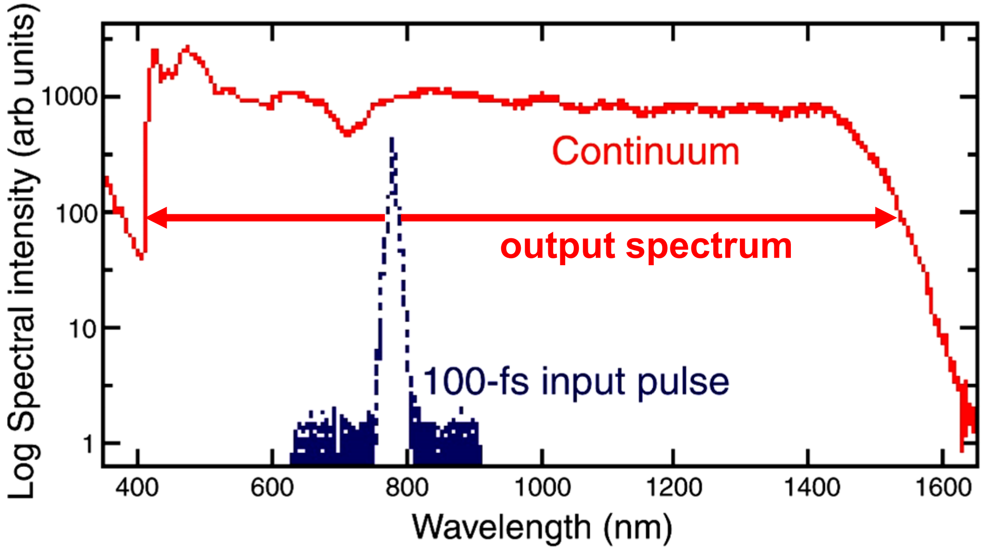
If $I(t)$ changes very rapidly (e.g., femtosecond pulse), then its derivative is large - so that the excursions of the frequency $\delta\omega$ could be larger than the initial bandwidth of the pulse! The spectrum of the light must be broadened!



microstructured optical fiber



broadened by a factor of 100!



Optics Letters, vol. 25, p. 25 (2000)

The world's shortest light pulse (1986 - 1997)

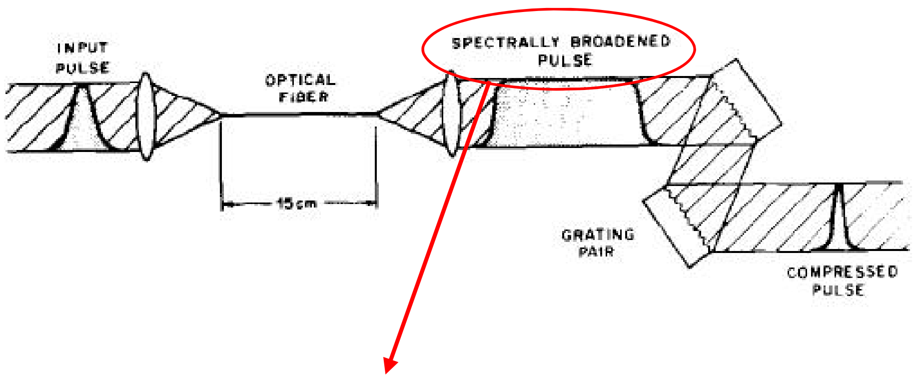
July 1987 / Vol. 12, No. 7 / OPTICS LETTERS 483

Compression of optical pulses to six femtoseconds by using cubic phase compensation

R. L. Fork, C. H. Brito Cruz, P. C. Becker, and C. V. Shank

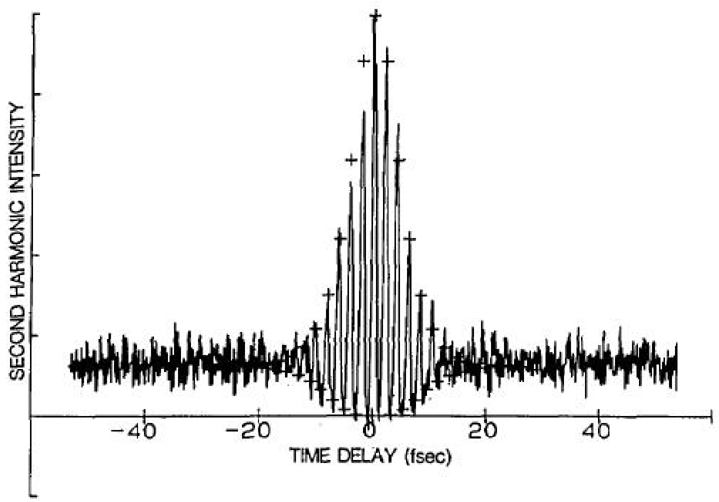
AT&T Bell Laboratories, Crawfords Corner Road, Holmdel, New Jersey 07733-1988

A diagram from an earlier paper (gratings only):



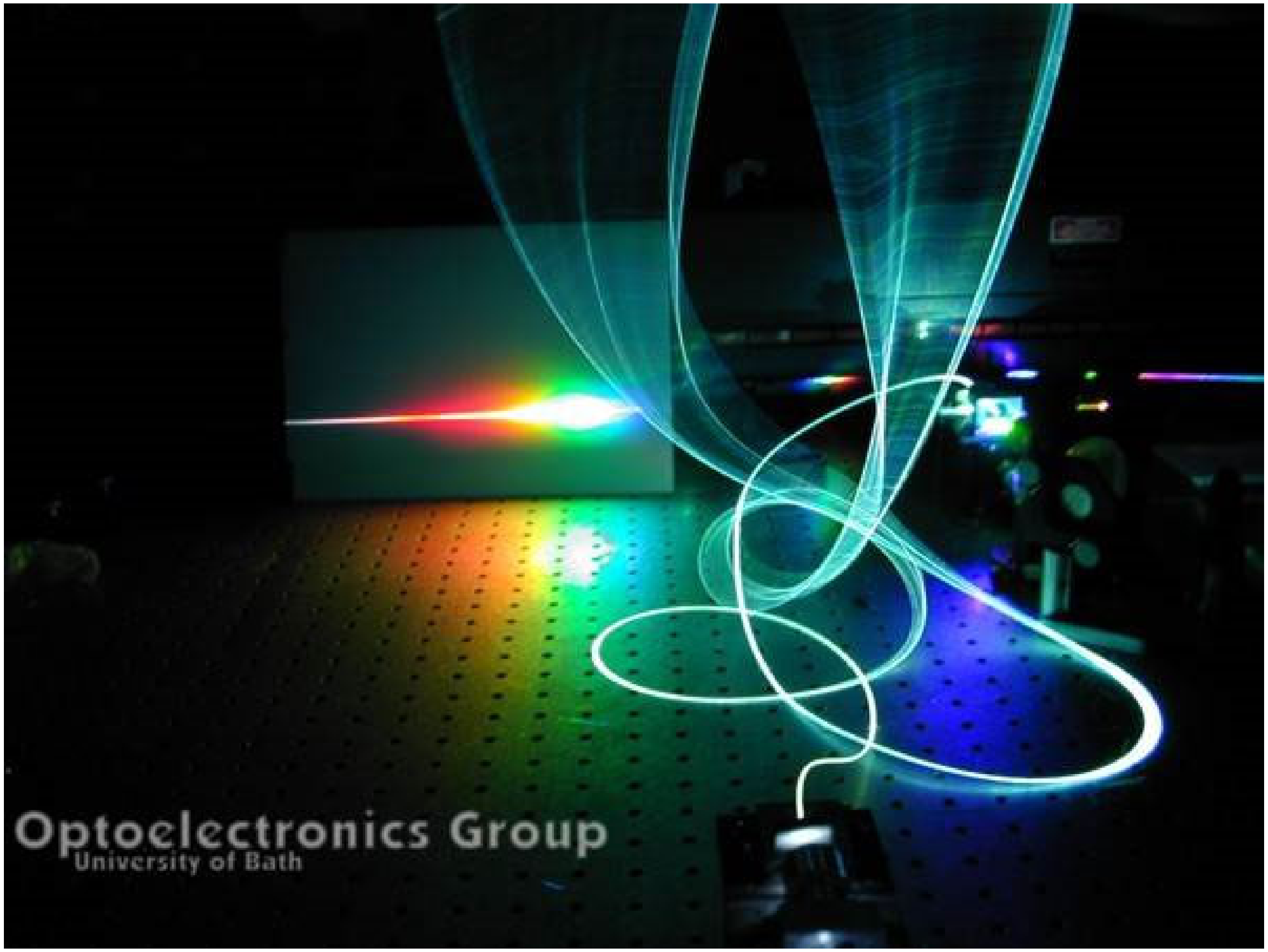
spectrally *and* temporally broadened!

the result with gratings & prisms:



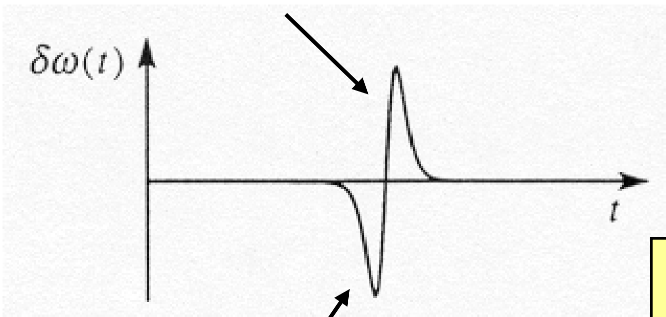
Supercontinuum generation

red light in…   white light out



Self-phase modulation + anomalous dispersion

new frequency components generated by the later (falling) edge of the pulse, at $\omega > \omega_0$



new frequency components generated by the early (rising) edge of the pulse, at $\omega < \omega_0$

What if this occurs in a regime of anomalous dispersion?

$$\frac{dn}{d\omega} < 0 \quad \rightarrow$$

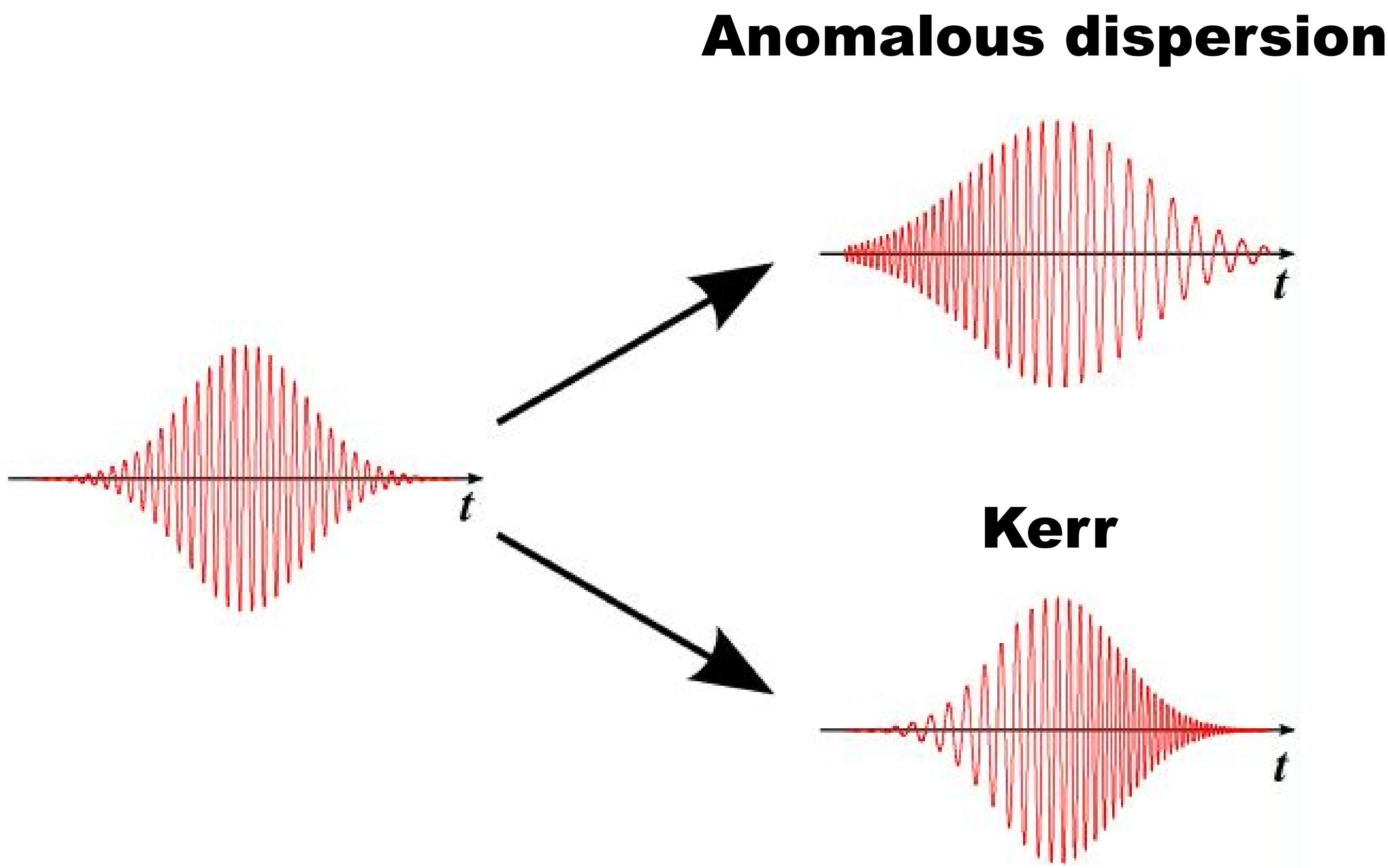
lower ω = lower velocity
higher ω = higher velocity

The new (lower) frequencies generated on the leading edge travel a bit slower, so the pulse catches up to them.

The new (higher) frequencies generated on the trailing edge travel a bit faster, so they catch up to the pulse.

Result: the pulse shape is stable! A "solitary wave", or "soliton"

Anomalous dispersion + Kerr effect = soliton



Solitons

Soliton: a localized traveling wave whose intensity profile is stablized by the interplay of (linear) anomalous dispersion and non-linearity, so that its shape doesn't change as the wave propagates.

Discovered in 1834: John Scott Russell observed "solitary waves" of water propagating for long distances along the Union canal in Scotland.

"…a well-defined heap of water which continued its course along the channel apparently without change of form or diminution of speed."

a recreation of his observation, on the John Scott Russell Acqueduct, 1995

Solitons are a solution to the nonlinear Schroedinger equation

$$\frac{\partial E}{\partial z} = iD\frac{\partial^2 E}{\partial t^2} - i\xi\left|E\right|^2 E$$

where $D \equiv \frac{1}{2}\frac{\partial^2 k}{\partial \omega^2}L$ and $\xi \equiv \frac{2\pi}{\lambda}n_2 L$

Group delay dispersion, GDD

Kerr nonlinearity

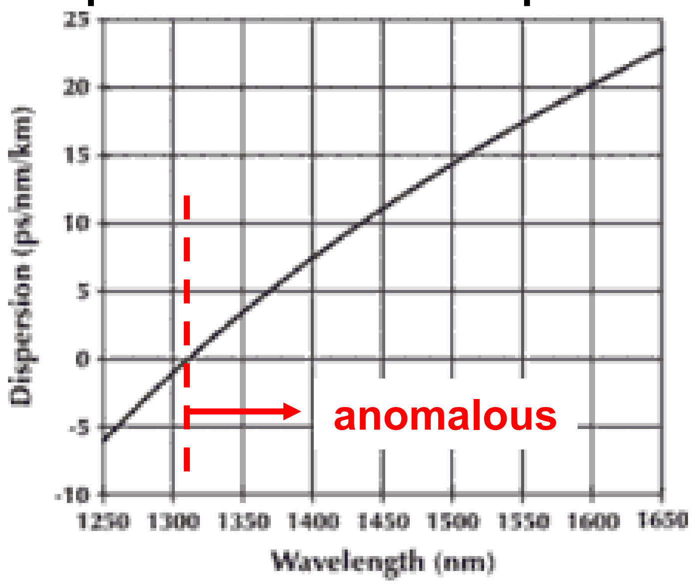
The envelope of the solution is a pulse: $\left|E(z,t)\right| = E_0 \operatorname{sech}\left(\frac{t}{\tau}\right)$ with duration: $\frac{1}{\tau^2} = -\frac{\xi\left|A_0\right|^2}{2D}$

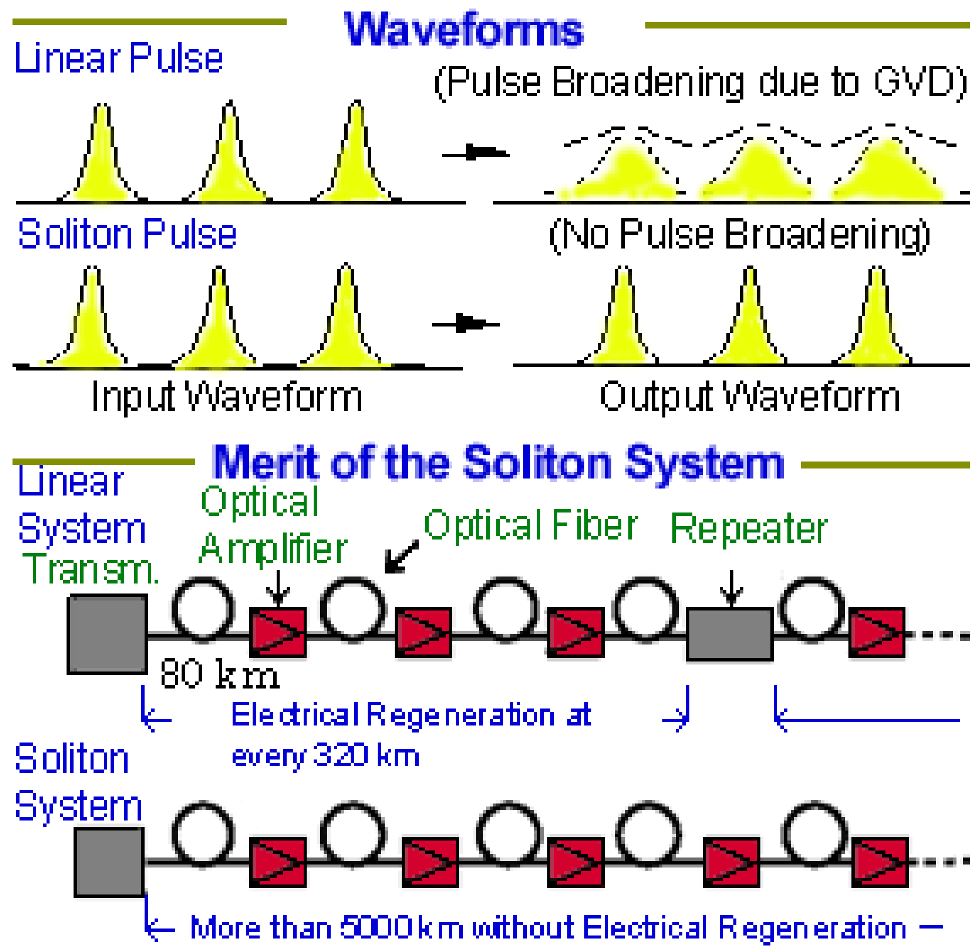
- solution only exists if $\xi/D < 0$ (requires anomalous dispersion)
- pulse duration is independent of propagation distance!

Solitons in optics

Solitons in fiber optics are the basis for many telecommunications transmission systems.

Dispersion of standard optical fiber

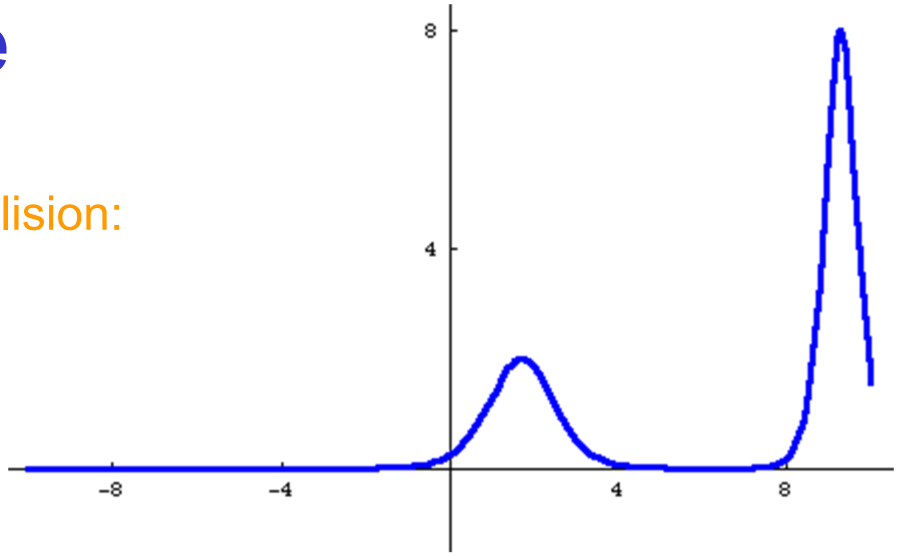




Solitons can exist in standard optical fibers for wavelengths $\lambda > 1310$ nm.

When solitons collide

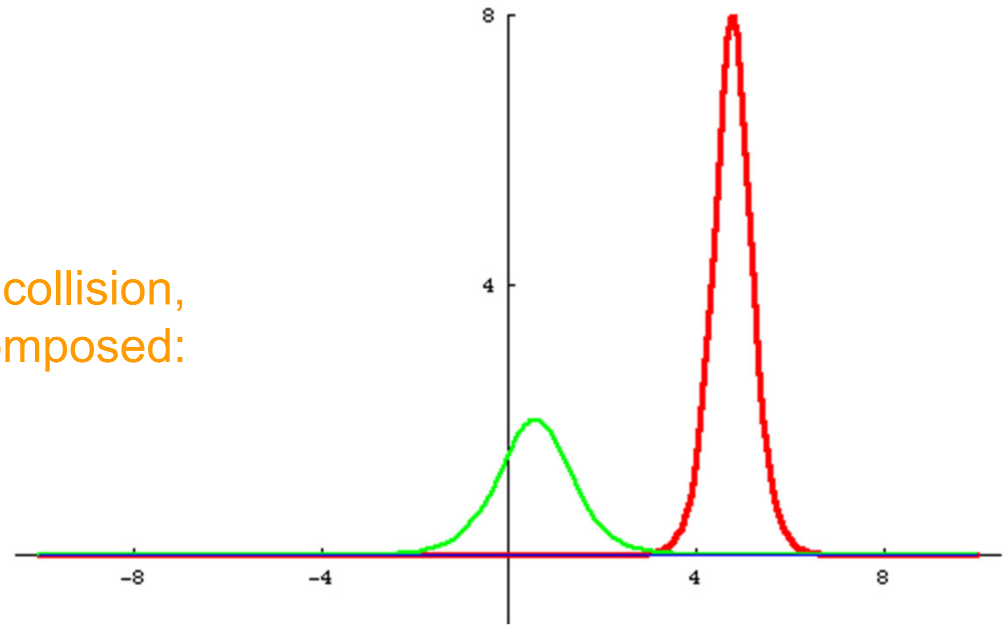
A soliton collision:



Because of the non-linear nature of the equation, superposition does not hold for solitons.

But they can still collide with each other, and when they do, they act like particles!

The same collision, decomposed:



A cool and easy-to-understand discussion about solitons:

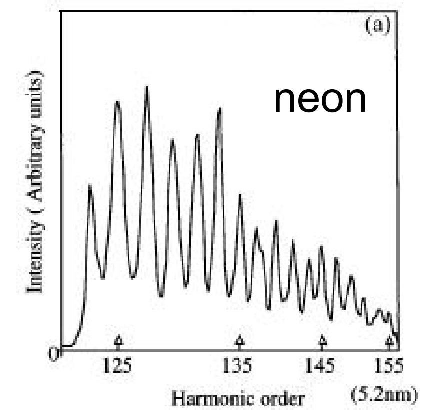
http://kasmana.people.cofc.edu/SOLITONPICS/

Limits to the perturbative approach

Consider propagation of an intense pulse in a gas.

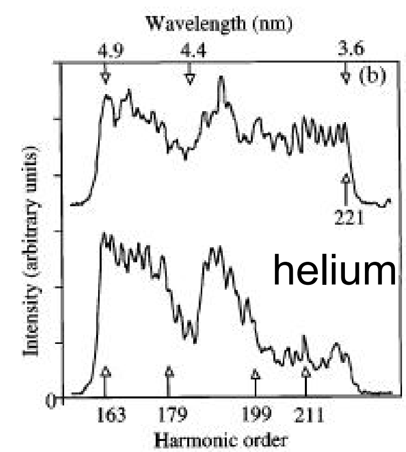
Symmetry: all even orders of χ vanish

$$P(t) = \chi^{(1)} E(t) + \chi^{(3)} E(t)^3 + \chi^{(5)} E(t)^5 + \ldots$$

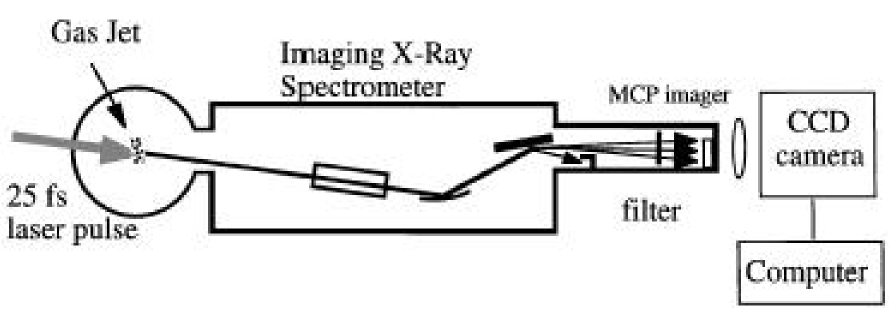


This treatment assumes $|\chi^{(3)}| >> |\chi^{(5)}| >> |\chi^{(7)}| \ldots$

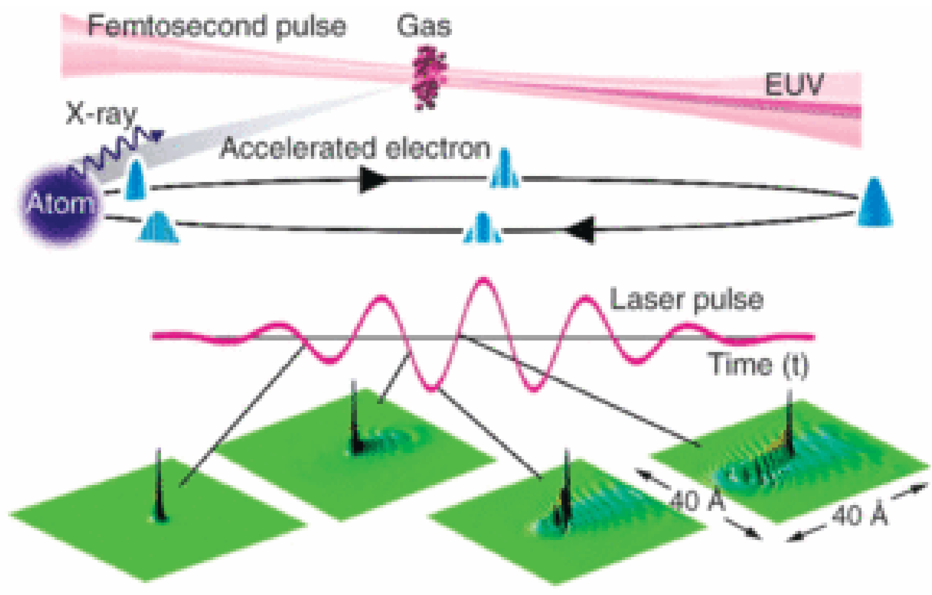
We would therefore expect each successive high harmonic to be weaker than the preceding one.



Counter-example:
Kapteyn and Murnane, *Phys. Rev. Lett.*, **79**, 2967 (1997)



How to explain high harmonic generation?



We cannot use a perturbative approach, i.e., $\chi^{(n)}$.

We must resort to an atomic picture of the dynamics:
1. ionization of an atom
2. acceleration of a free electron
3. impact with the parent ion

If all of the x-ray harmonics are in phase (and since there are a lot of them!), they could be used to generate attosecond pulses.