

ULTRAFAST OPTICS

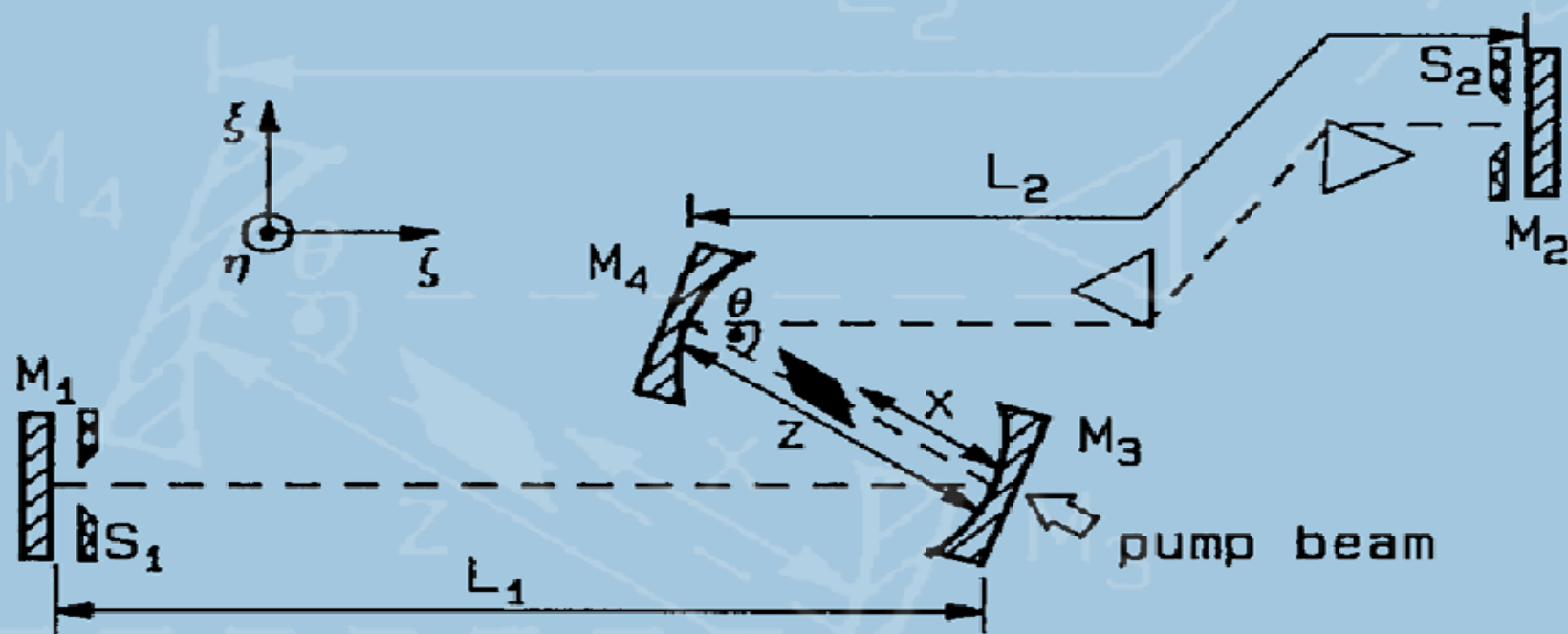


image from G. Cerullo et al., Opt. Lett. 19, 807 (1994), © CSA

by PIOTR WASYLCHYK

Nonlinear Optics

Why do nonlinear-optical effects occur?

Maxwell's equations in a medium

Nonlinear-optical media

Second-harmonic generation

Sum- and difference-frequency generation

Higher-order nonlinear optics

The Slowly Varying Envelope Approximation

Phase-matching and Conservation laws for photons

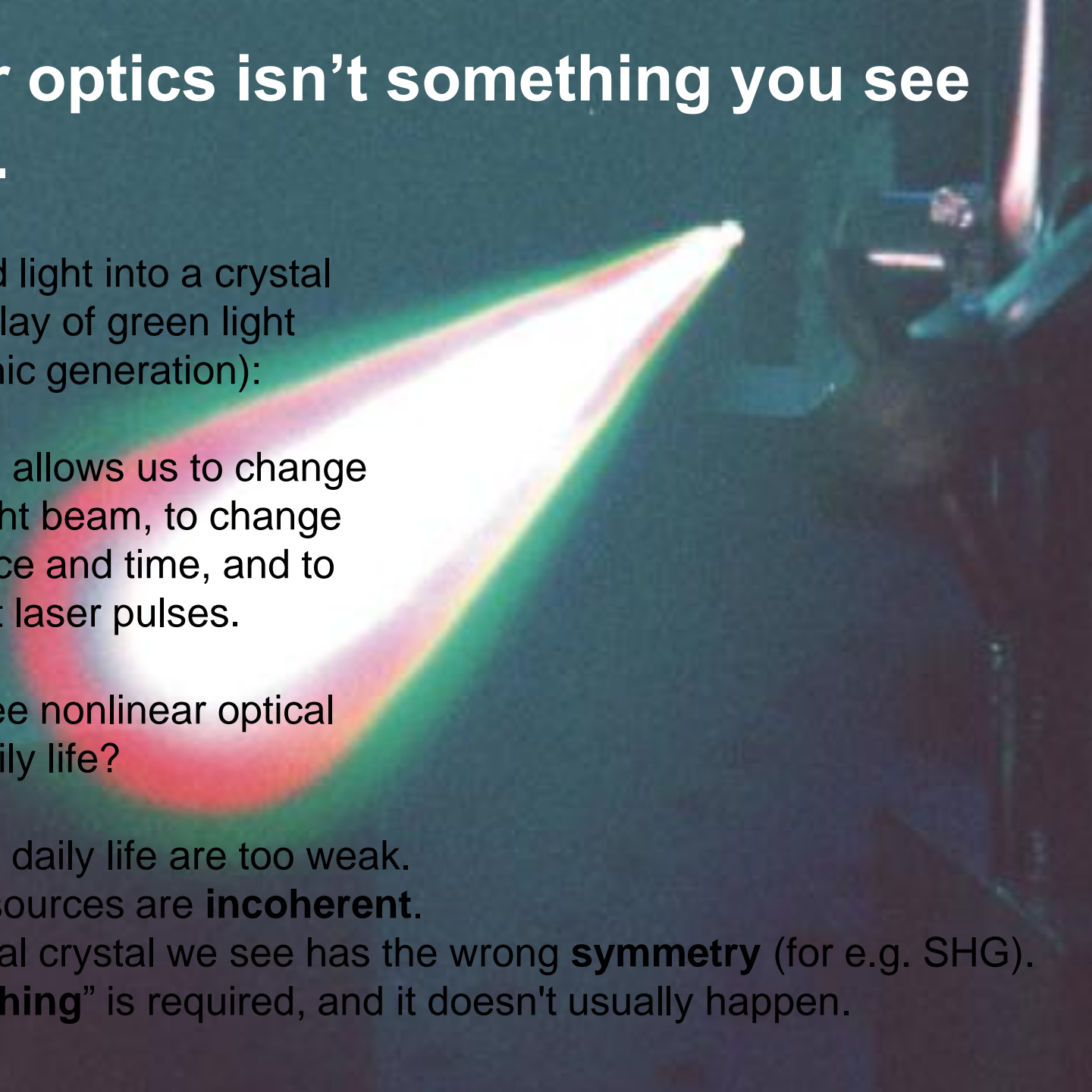
Nonlinear optics isn't something you see everyday.

Sending infrared light into a crystal yielded this display of green light (second-harmonic generation):

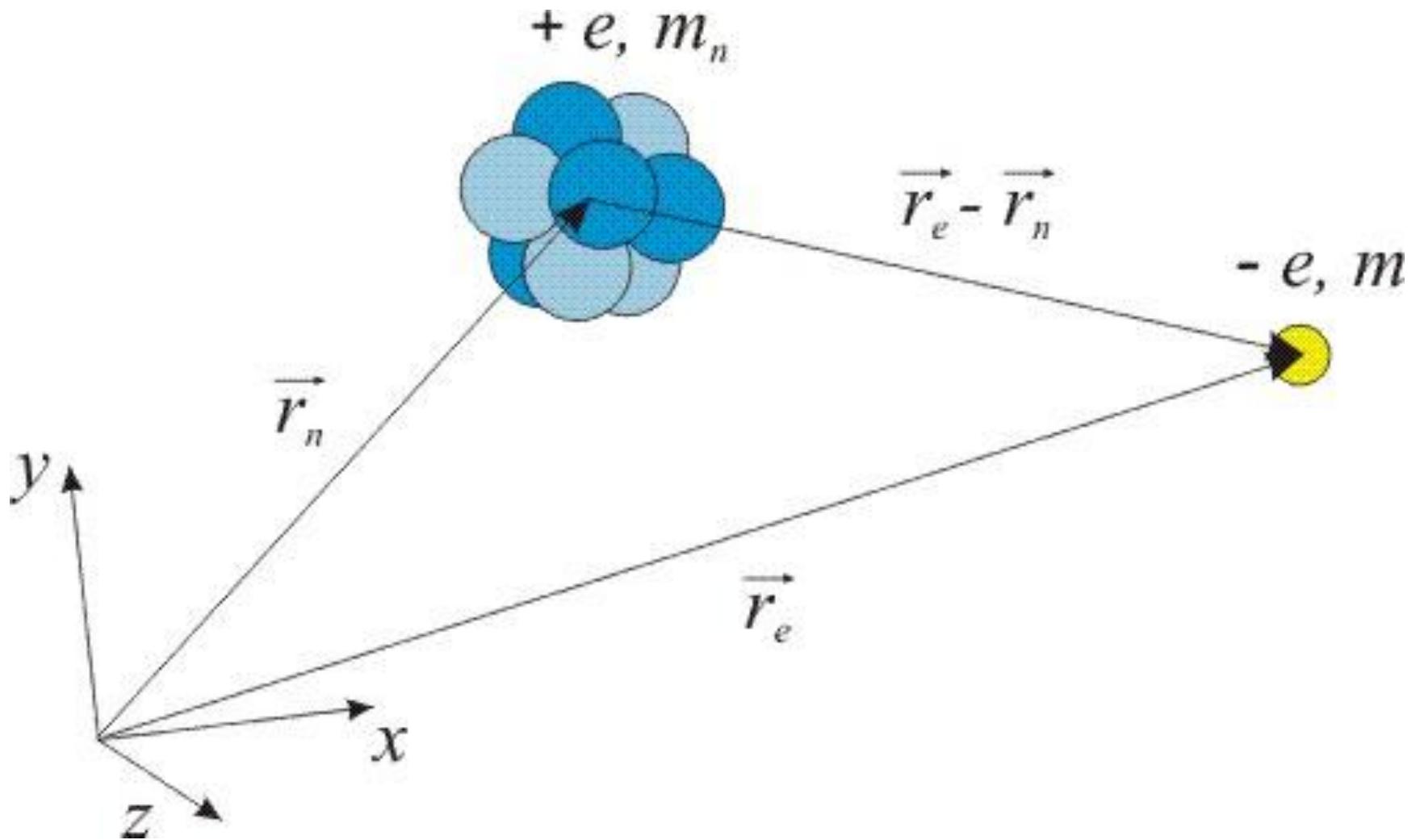
Nonlinear optics allows us to change the color of a light beam, to change its shape in space and time, and to create ultrashort laser pulses.

Why don't we see nonlinear optical effects in our daily life?

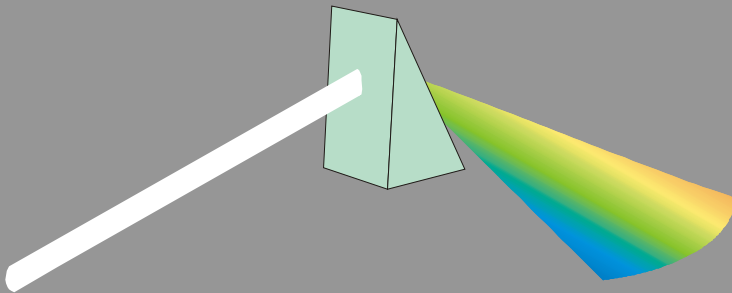
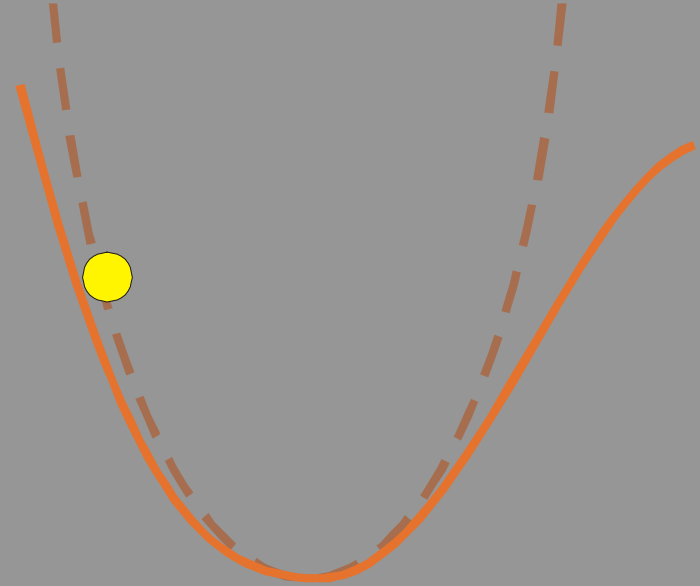
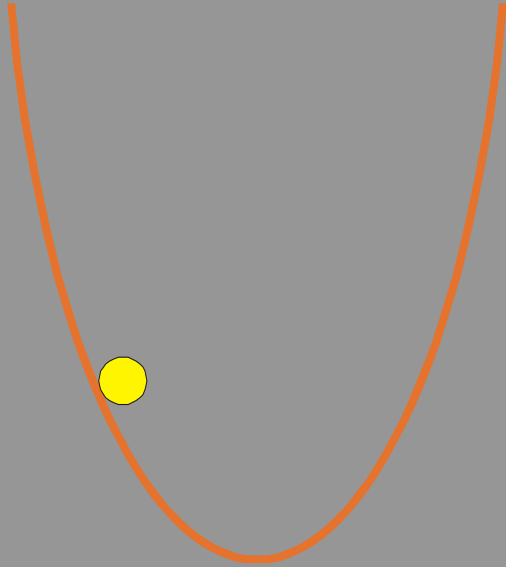
1. **Intensities** of daily life are too weak.
2. Normal light sources are **incoherent**.
3. The occasional crystal we see has the wrong **symmetry** (for e.g. SHG).
4. "**Phase-matching**" is required, and it doesn't usually happen.



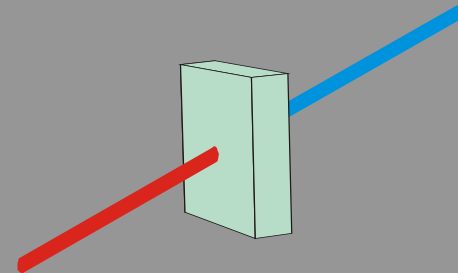
Electrons in a transparent dielectric (e.g. glass in the visible range)



Linear vs. nonlinear optics

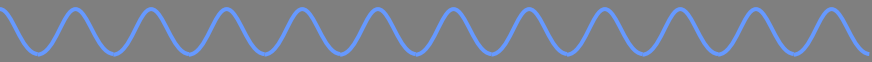


H. A. Lorentz (1909)



P. A. Franken (1961)

Nonlinearity



$$\mathbf{P} = \chi \varepsilon_0 \mathbf{E} + \chi^{(2)} \mathbf{E} \mathbf{E} + \dots$$



Nonlinear optics – the first „wave mixing” experiment

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FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.

ing. In this Letter we present a brief discussion of the requisite analysis and a description of experiments in which we have observed the second harmonic (at ~3472 Å) produced upon projection

$$x (E_x = 0)$$

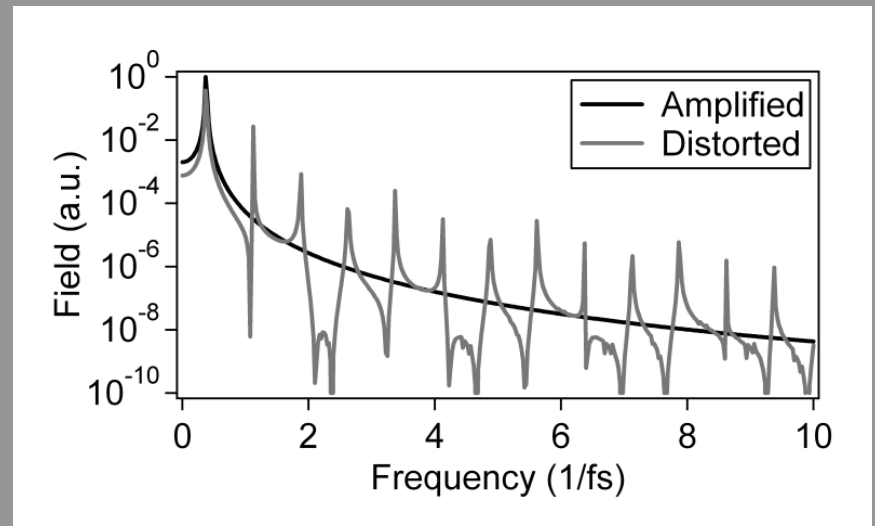
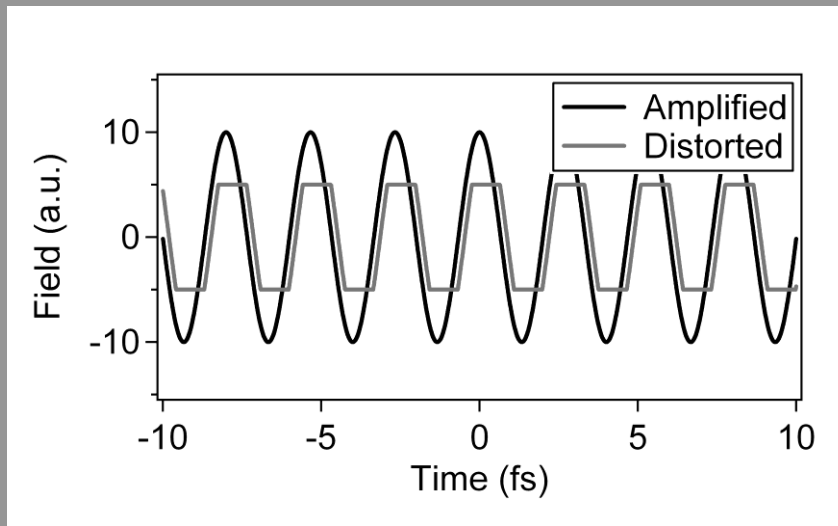
$$y (E_y = 0)$$

$$p_y^2 + p_z^2 = 0$$

$$p_z^2 + p_x^2 = \alpha^2 E_x^4$$

Nonlinear optics is analogous to nonlinear electronics, which we can observe easily.

Sending a high-volume sine-wave (“pure frequency”) signal into cheap speakers yields a truncated output signal, more of a square wave than a sine. This square wave has higher frequencies.

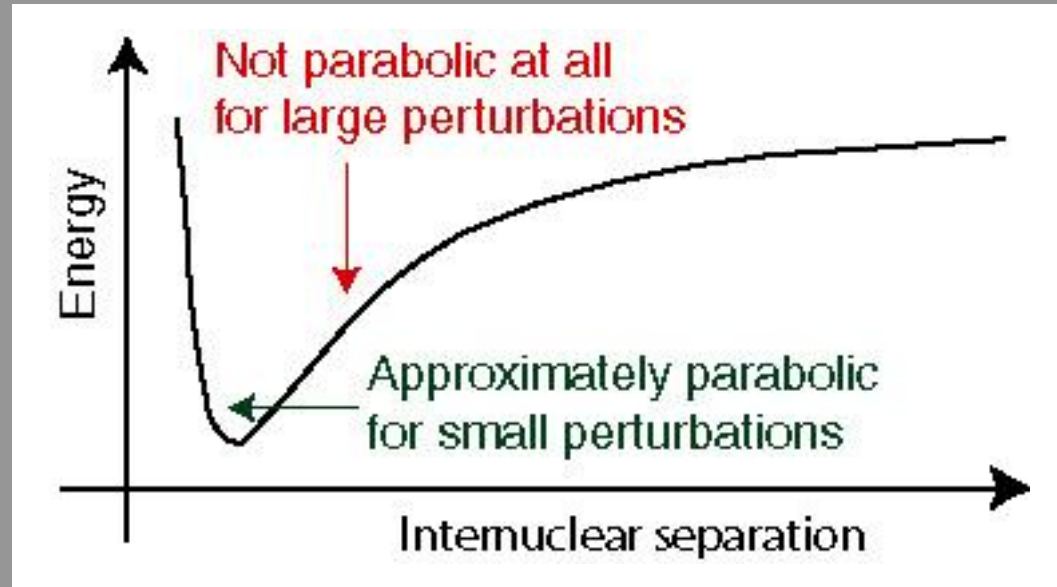


We hear this as distortion.

Nonlinear optics and anharmonic oscillators

Another way to look at nonlinear optics is that the potential of the electron or nucleus (in a molecule) is not a simple harmonic potential.

Example: vibrational motion:

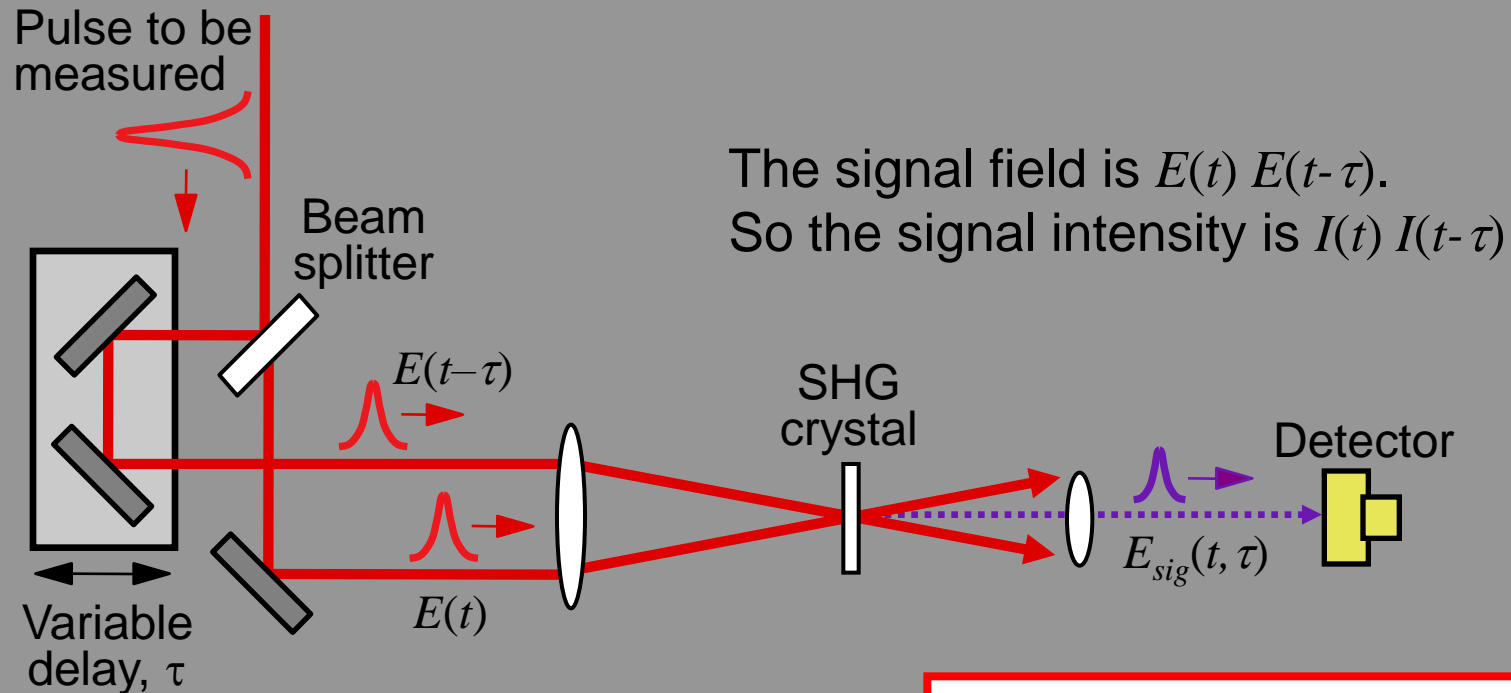


For weak fields, motion is harmonic, and linear optics prevails.
For strong fields (i.e., lasers), anharmonic motion occurs, and higher harmonics occur, both in the motion and the light emission.

Generating new frequencies



Pulse Measurement – only possible with nonlinear effects of some kind

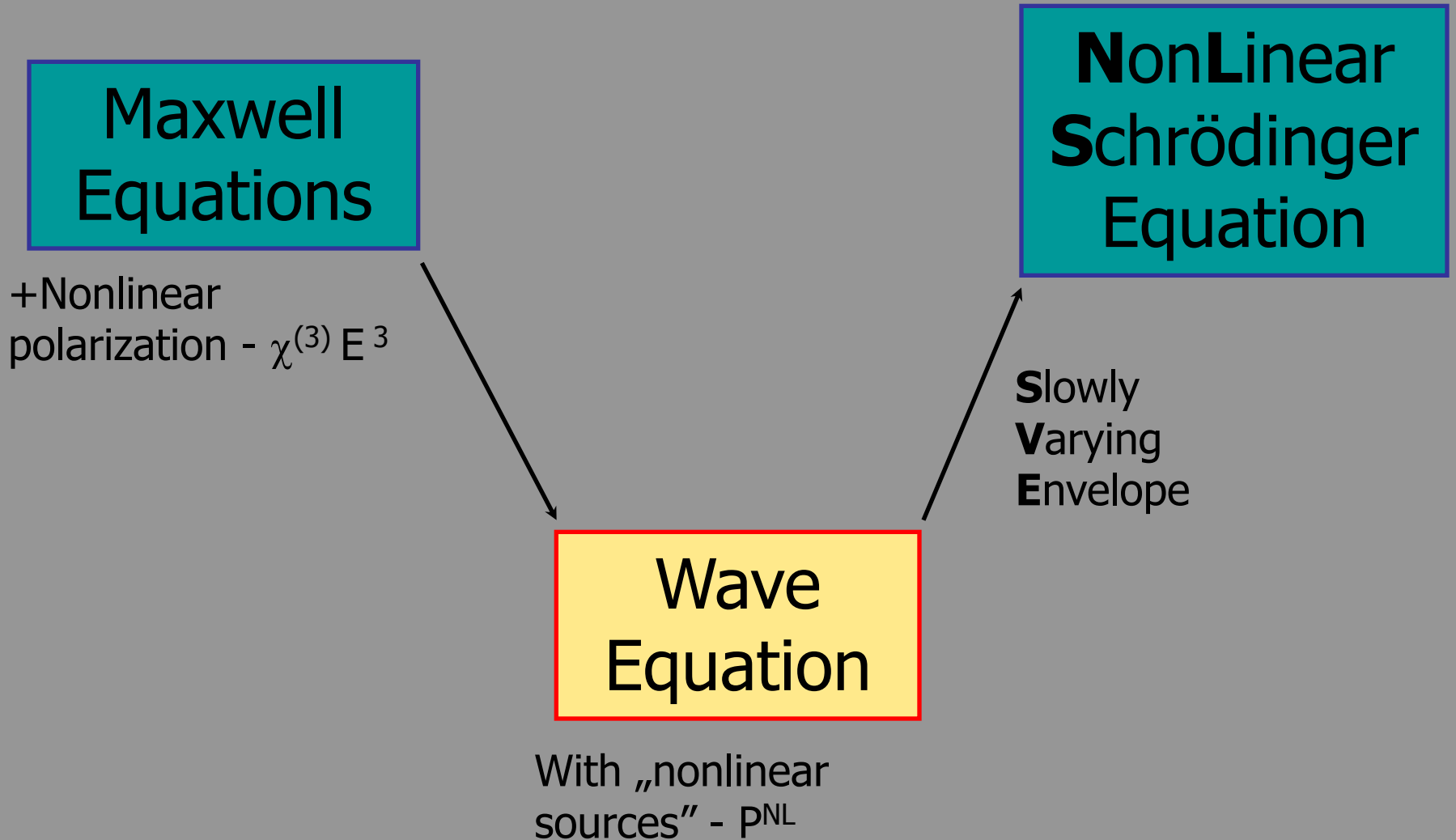


The signal field is $E(t) E(t-\tau)$.
So the signal intensity is $I(t) I(t-\tau)$

The Intensity
Autocorrelation:

$$A^{(2)}(\tau) \equiv \int_{-\infty}^{\infty} I(t) I(t-\tau) dt$$

From Maxwell Equations to NLS



Maxwell's Equations in a Medium

The induced polarization, \vec{P} , contains the effect of the medium:

$$\begin{aligned}\vec{\nabla} \cdot \vec{E} &= 0 & \vec{\nabla} \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t} \\ \vec{\nabla} \cdot \vec{B} &= 0 & \vec{\nabla} \times \vec{B} &= \frac{1}{c_0^2} \frac{\partial \vec{E}}{\partial t} + \mu_0 \frac{\partial \vec{P}}{\partial t}\end{aligned}$$

These equations reduce to the (scalar) wave equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$

“Inhomogeneous
Wave Equation”

Sine waves of all frequencies are solutions to the wave equation; it's the polarization that tells which frequencies will occur.

The polarization is the driving term for the solution to this equation.

Solving the wave equation in the presence of linear induced polarization

For low irradiances, the polarization is proportional to the incident field:

$$P = \epsilon_0 \chi E$$

In this simple (and most common) case, the wave equation becomes:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \frac{1}{c_0^2} \chi \frac{\partial^2 E}{\partial t^2} \quad \text{Using the fact that: } \epsilon_0 \mu_0 = 1/c_0^2$$

Simplifying:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1 + \chi}{c_0^2} \frac{\partial^2 E}{\partial t^2} = 0$$

This equation has the solution: $E(z, t) \propto E(0) \cos(\omega t - k z)$

where $\omega = c k$ and $c = c_0/n$ and $n = (1 + \chi)^{1/2}$

The induced polarization only changes the refractive index. Dull.
If only the polarization contained other frequencies...

Maxwell's Equations in a *Nonlinear* Medium

Nonlinear optics is what happens when the polarization is the result of higher-order (nonlinear!) terms in the field:

$$P = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots \right]$$

What are the effects of such nonlinear terms? Consider the second-order term:

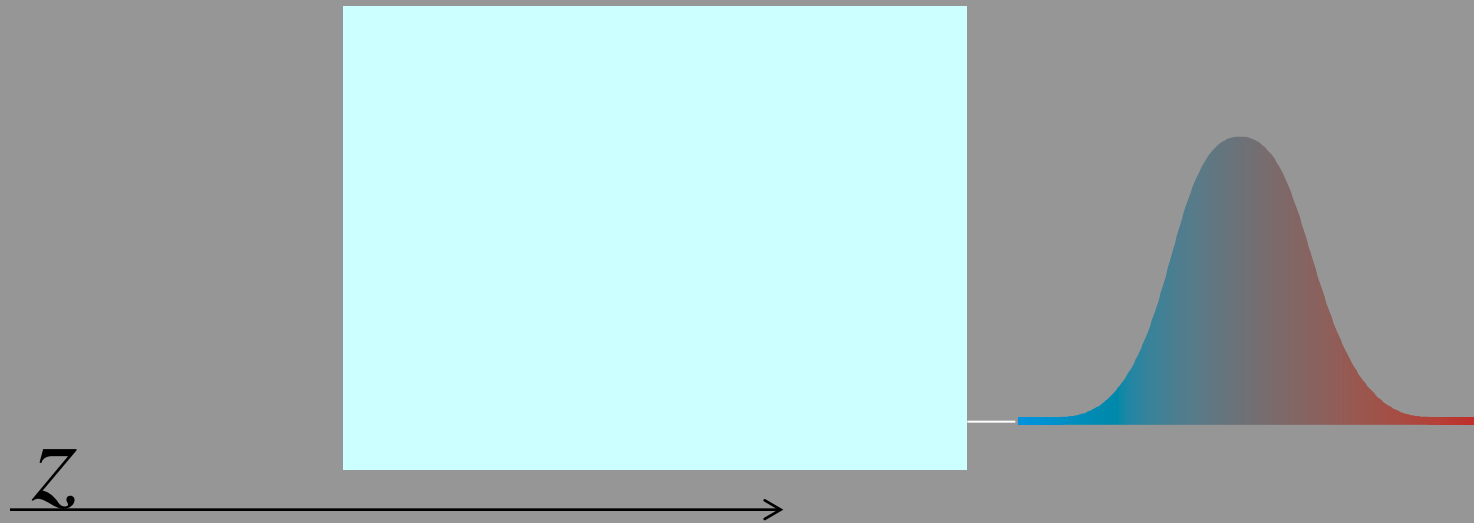
Since $E(t) \propto E \exp(i\omega t) + E^* \exp(-i\omega t)$,

$$E(t)^2 \propto E^2 \exp(2i\omega t) + 2|E|^2 + E^{*2} \exp(-2i\omega t)$$

$2\omega = 2\text{nd harmonic!}$

Harmonic generation is one of many exotic effects that can arise!

From Maxwell Equations to NLS



$$\frac{\partial}{\partial z} A = -\frac{i}{2} \beta_2 \frac{\partial^2}{\partial t^2} A - \frac{i}{2} \gamma_{xx} \Delta_{\perp} A + i\kappa |A|^2 A$$

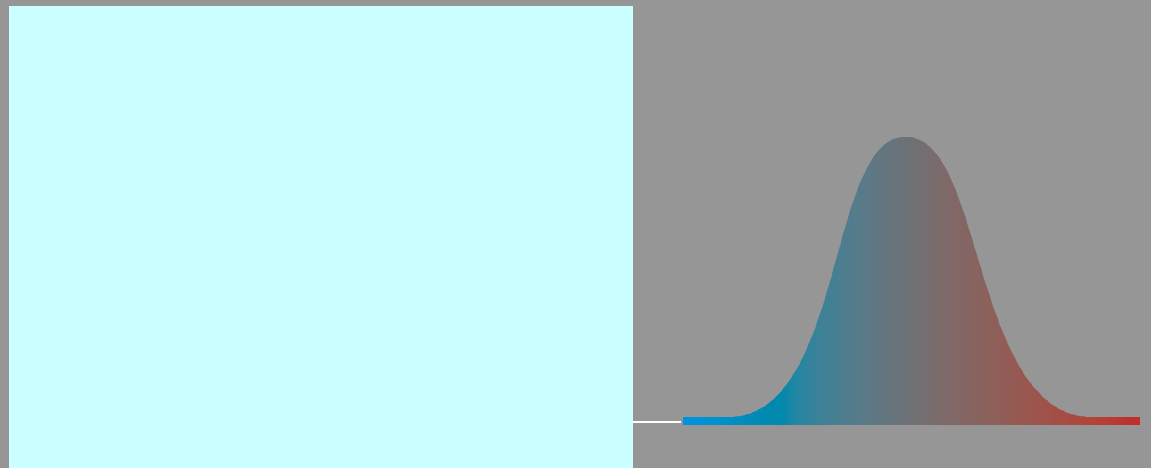
dispersion

diffraction

nonlinearity

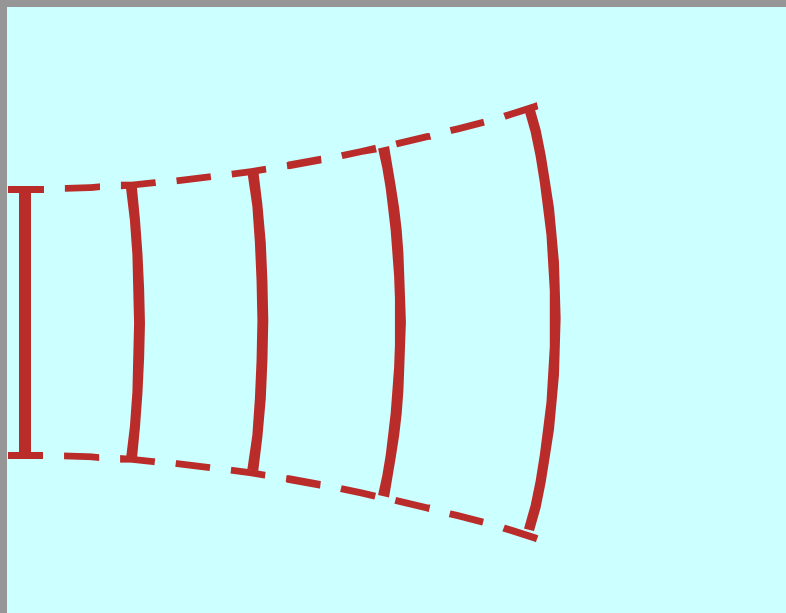
dispersion

$$-\frac{i}{2}\beta_2\frac{\partial^2}{\partial t^2}A$$



diffraction

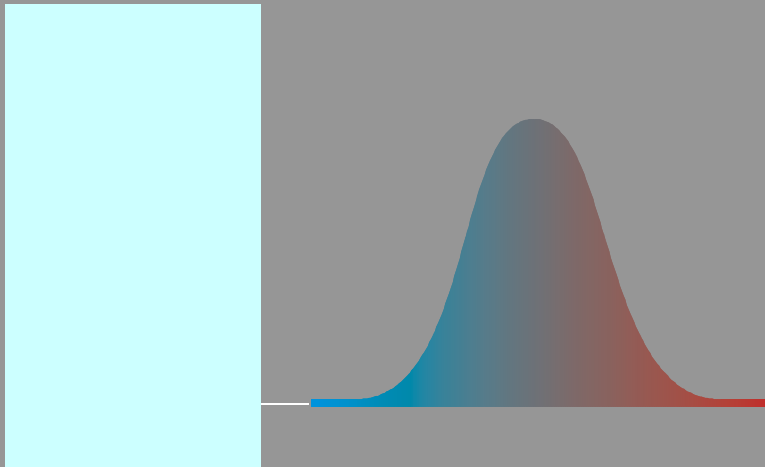
$$-\frac{i}{2}\gamma_{xx}\Delta_{\perp} A$$



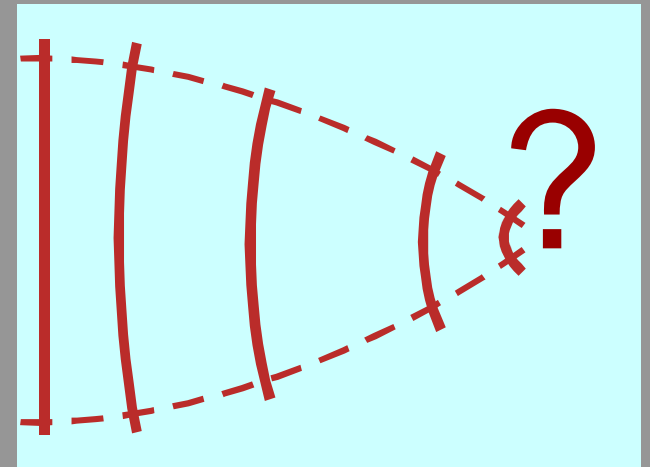
nonlinearity (Kerr-type)

$$i\kappa |A|^2 A$$

in time



in space



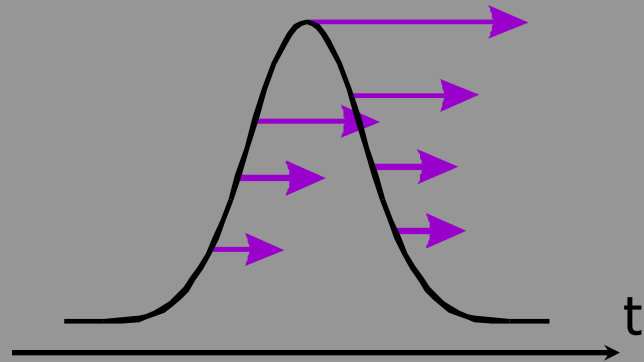
More accurate propagation equation

$$\begin{aligned}
 L_{\text{DF}}^{-1} \frac{\partial A}{\partial z} = & -\beta_1 \omega_0 \eta \frac{\partial A}{\partial t} - \frac{i\beta_2 \omega_0^2 \eta^2}{2} \frac{\partial^2 A}{\partial t^2} + \frac{\beta_3 \omega_0^3 \eta^3}{6} \frac{\partial^3 A}{\partial t^3} + \frac{i\beta_4 \omega_0^4 \eta^4}{24} \frac{\partial^4 A}{\partial t^4} \\
 & + i \frac{\gamma_{xx} k_0^2 \epsilon^2}{2} \left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} \right) + \frac{\gamma_{txx} \omega_0 k_0^2 \epsilon^2 \eta}{2} \left(\frac{\partial^3 A}{\partial x^2 \partial t} + \frac{\partial^3 A}{\partial y^2 \partial t} \right) \\
 & + i \frac{\gamma_{xxxx} k_0^4 \epsilon^4}{8} \left(\frac{\partial^4 A}{\partial x^4} + 2 \frac{\partial^4 A}{\partial x^2 \partial y^2} + \frac{\partial^4 A}{\partial y^4} \right) + \dots + \frac{2\pi \chi^{(3)} \omega_0}{nc} \\
 & \times \left\{ i |A|^2 A + \eta \left[\left(\frac{c\beta_1}{n} - 2 \right) \frac{\partial (|A|^2 A)}{\partial t} \right] - i \epsilon^2 \frac{1}{n^2} \left[A \frac{\partial A^*}{\partial x} \frac{\partial A}{\partial x} + A \frac{\partial A^*}{\partial y} \frac{\partial A}{\partial y} \right. \right. \\
 & \left. \left. + \frac{1}{2} A^* \left(\frac{\partial A}{\partial x} \right)^2 + \frac{1}{2} A^* \left(\frac{\partial A}{\partial y} \right)^2 + |A|^2 \frac{\partial^2 A}{\partial x^2} + |A|^2 \frac{\partial^2 A}{\partial y^2} \right] \right\}
 \end{aligned}$$

Higher nonlinear effects – an example

Self steepening

$$+\eta \left(\frac{\beta_1 c}{n_0} - 2 \right) \partial_t (|A|^2 A)$$



Split-step method

medium

Input pulse

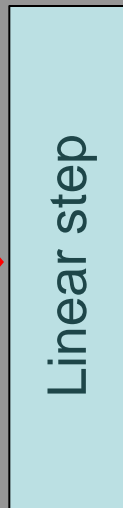


$$\begin{aligned}
 L_{\text{DF}}^{-1} \frac{\partial A}{\partial z} = & -\beta_1 \omega_0 \eta \frac{\partial A}{\partial t} - \frac{i\beta_2 \omega_0^2 \eta^2}{2} \frac{\partial^2 A}{\partial t^2} + \frac{\beta_3 \omega_0^3 \eta^3}{6} \frac{\partial^3 A}{\partial t^3} + \frac{i\beta_4 \omega_0^4 \eta^4}{24} \frac{\partial^4 A}{\partial t^4} \\
 & + i \frac{\gamma_{xx} k_0^2 \epsilon^2}{2} \left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} \right) + \frac{\gamma_{txx} \omega_0 k_0^2 \epsilon^2 \eta}{2} \left(\frac{\partial^3 A}{\partial x^2 \partial t} + \frac{\partial^3 A}{\partial y^2 \partial t} \right) \\
 & + i \frac{\gamma_{xxxx} k_0^4 \epsilon^4}{8} \left(\frac{\partial^4 A}{\partial x^4} + 2 \frac{\partial^4 A}{\partial x^2 \partial y^2} + \frac{\partial^4 A}{\partial y^4} \right) + \dots + \frac{2\pi \chi^{(3)} \omega_0}{nc} \\
 & \times \left\{ i |A|^2 A + \eta \left[\left(\frac{c\beta_1}{n} - 2 \right) \frac{\partial (|A|^2 A)}{\partial t} \right] - i \epsilon^2 \frac{1}{n^2} \left[A \frac{\partial A^*}{\partial x} \frac{\partial A}{\partial x} + A \frac{\partial A^*}{\partial y} \frac{\partial A}{\partial y} \right. \right. \\
 & \left. \left. + \frac{1}{2} A^* \left(\frac{\partial A}{\partial x} \right)^2 + \frac{1}{2} A^* \left(\frac{\partial A}{\partial y} \right)^2 + |A|^2 \frac{\partial^2 A}{\partial x^2} + |A|^2 \frac{\partial^2 A}{\partial y^2} \right] \right\}. \quad (12)
 \end{aligned}$$

Output pulse



Input pulse



Output pulse



More on solving the wave equation in nonlinear optics at the end of the lecture notes

Sum- and difference-frequency generation

Suppose there are two different-color beams present:

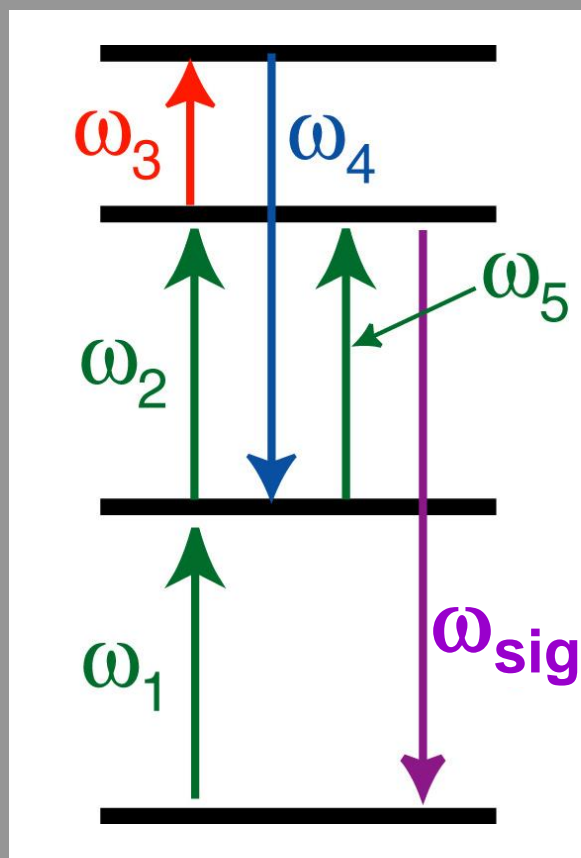
$$E(t) \propto E_1 \exp(i\omega_1 t) + E_1^* \exp(-i\omega_1 t) + E_2 \exp(i\omega_2 t) + E_2^* \exp(-i\omega_2 t)$$

So:

$$\begin{aligned} E(t)^2 \propto & E_1^2 \exp(2i\omega_1 t) + E_1^{*2} \exp(-2i\omega_1 t) && \text{2nd-harmonic gen} \\ & + E_2^2 \exp(2i\omega_2 t) + E_2^{*2} \exp(-2i\omega_2 t) && \text{2nd-harmonic gen} \\ & + 2E_1 E_2 \exp[i(\omega_1 + \omega_2)t] + 2E_1^* E_2^* \exp[-i(\omega_1 + \omega_2)t] && \text{Sum-freq gen} \\ & + 2E_1 E_2^* \exp[i(\omega_1 - \omega_2)t] + 2E_1^* E_2 \exp[-i(\omega_1 - \omega_2)t] && \text{Diff-freq gen} \\ & + 2|E_1|^2 + 2|E_2|^2 && \text{dc rectification} \end{aligned}$$

Note also that, when ω_i is negative inside the exp, the E in front has a $*$.

Complicated nonlinear-optical effects can occur.



Nonlinear-optical processes are often referred to as:

"N-wave-mixing processes"

where N is the number of photons involved (including the emitted one). This is a six-wave-mixing process.

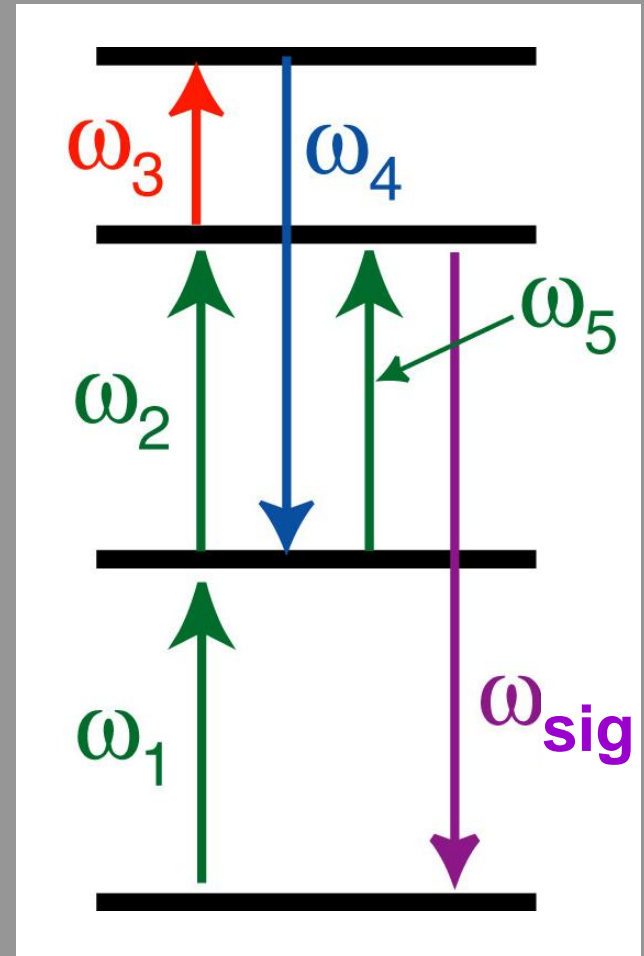
Emitted-light frequency

The more photons (i.e., the higher the order) the weaker the effect, however. Very-high-order effects can be seen, but they require very high irradiance. Also, if the photon energies coincide with the medium's energy levels as above, the effect will be stronger.

Induced polarization for nonlinear optical effects

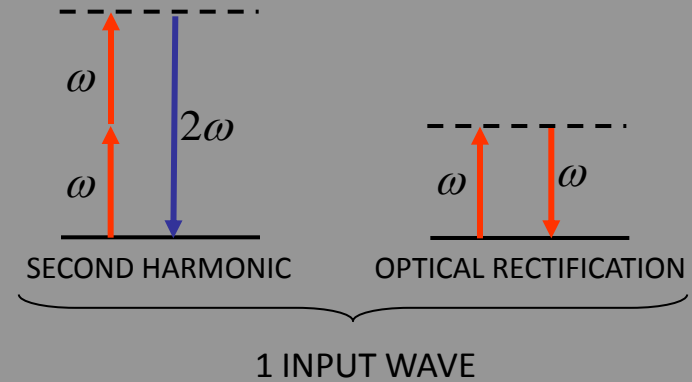
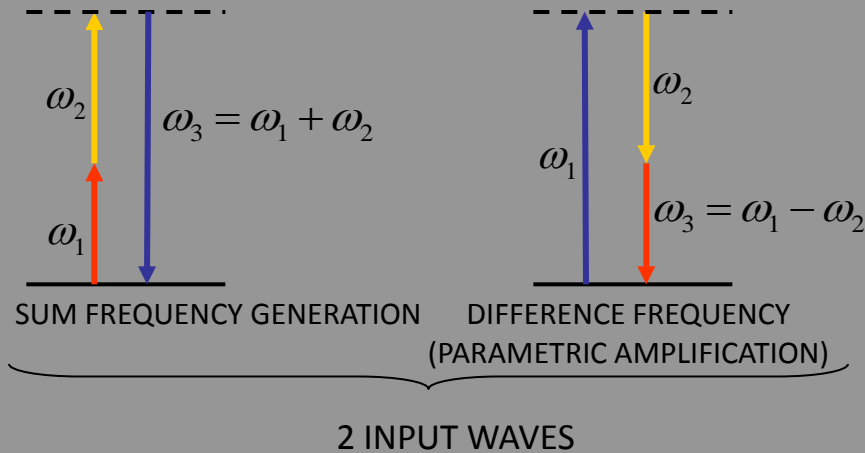
Arrows pointing upward correspond to absorbed photons and contribute a factor of their field, E_i ; arrows pointing downward correspond to emitted photons and contribute a factor the complex conjugate of their field:

$$P = \epsilon_0 \chi^{(5)} E_1 E_2 E_3 E_4^* E_5$$

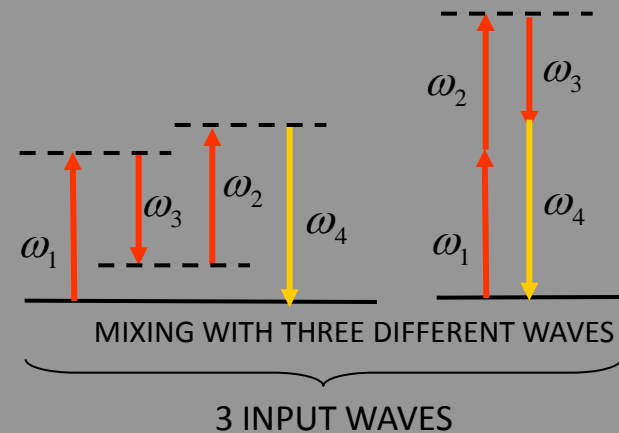
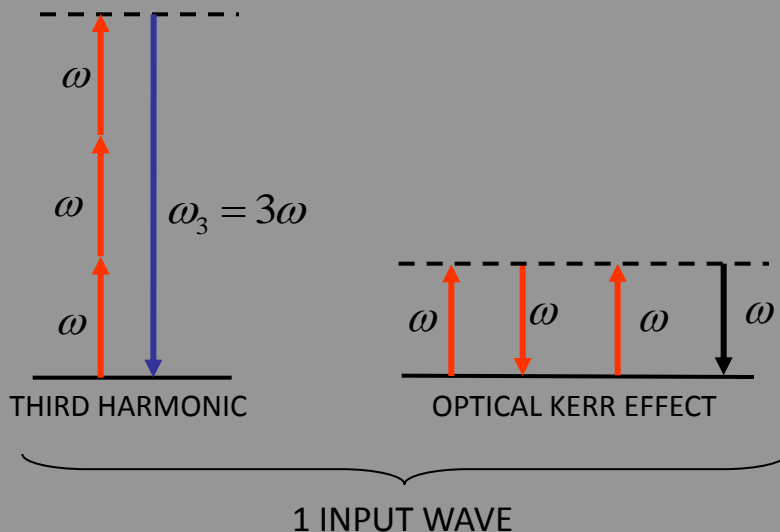


More nonlinear processes

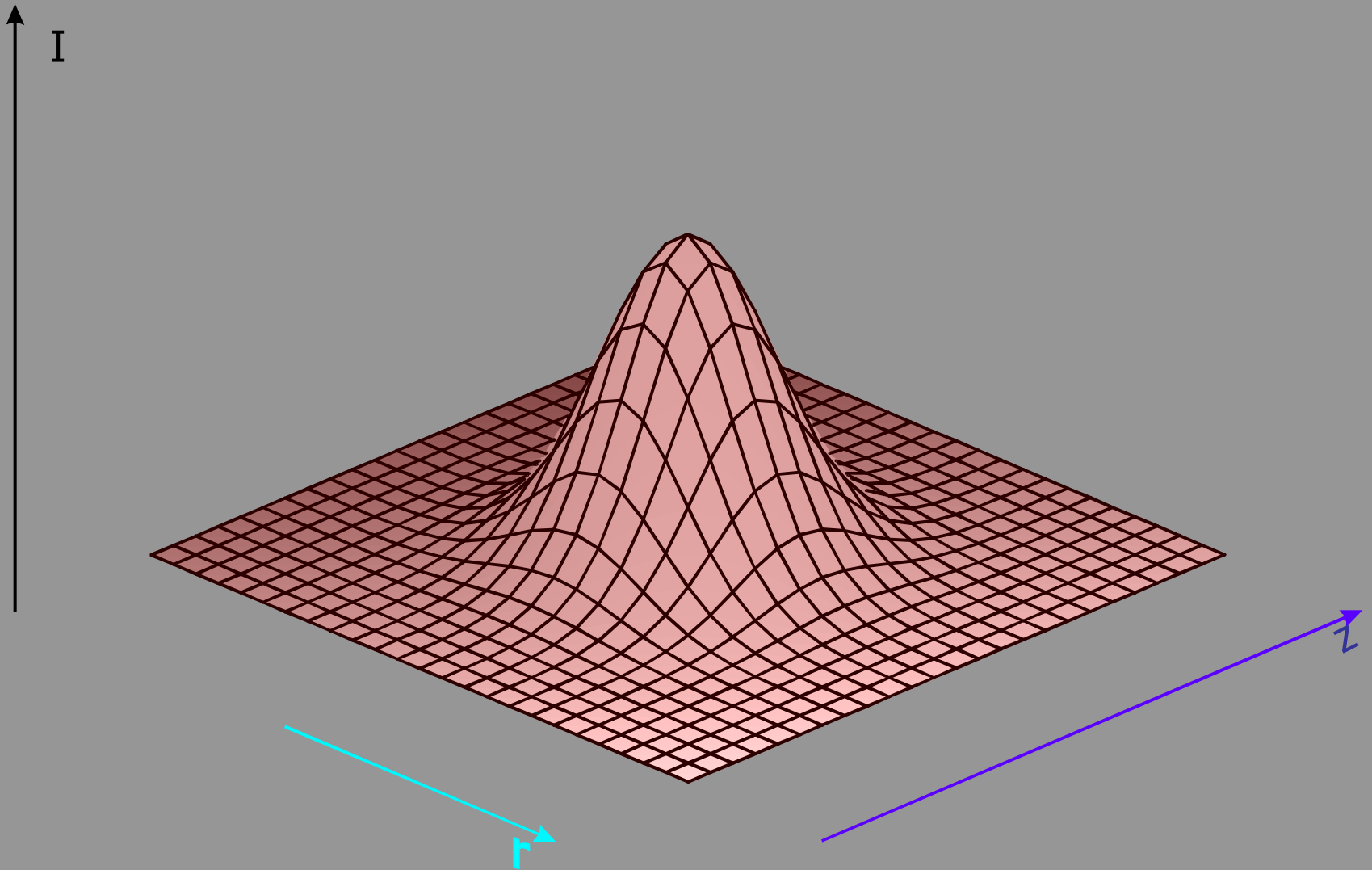
$\chi(2)$ processes (three wave mixing)



$\chi(3)$ processes (four wave mixing)



Pulse



The simplest nonlinear propagation



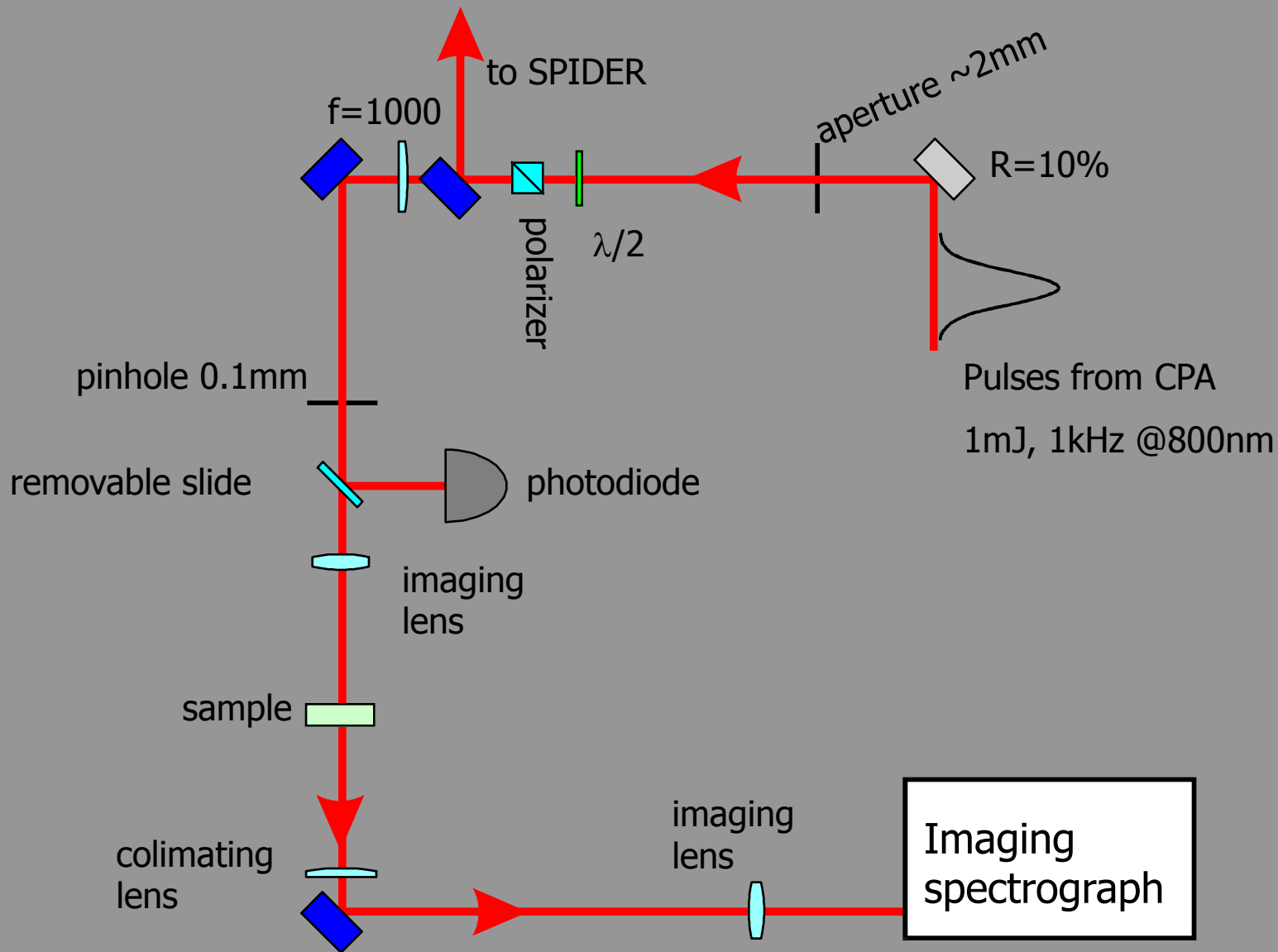
Dielectric

$n(\omega)$, $\chi^{(3)}$, isotropic, far from resonance

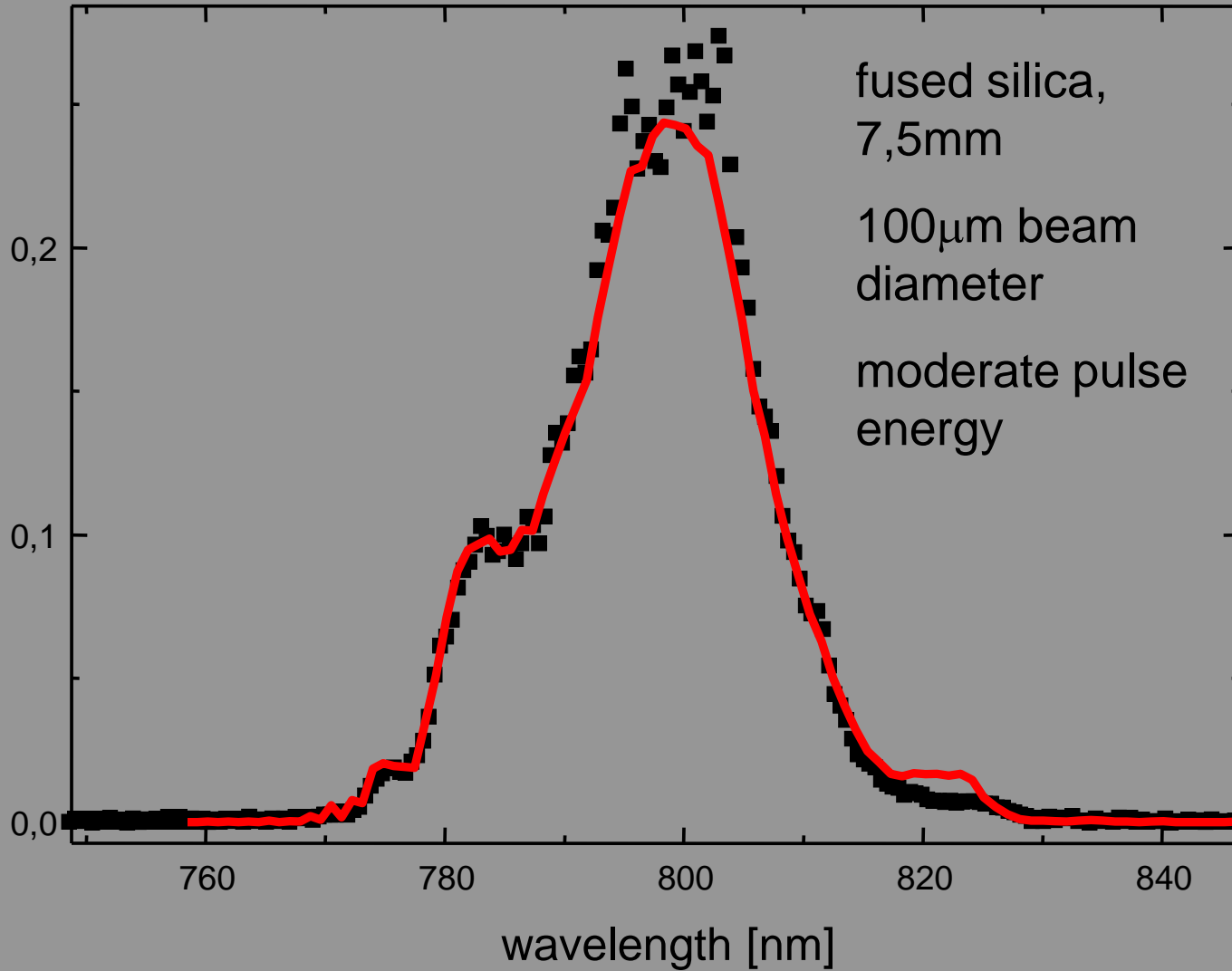
Model - assumptions

1. The pulse is longer than one optical cycle ($>3\text{fs}$ at $\lambda_0=800\text{nm}$)
2. The focus is wider than $\lambda_0/2n$ (about 300nm)
3. The medium is isotropic
4. Polarization consists of linear and Kerr ($\propto E^3$) parts
6. The pulse remains linearly polarized

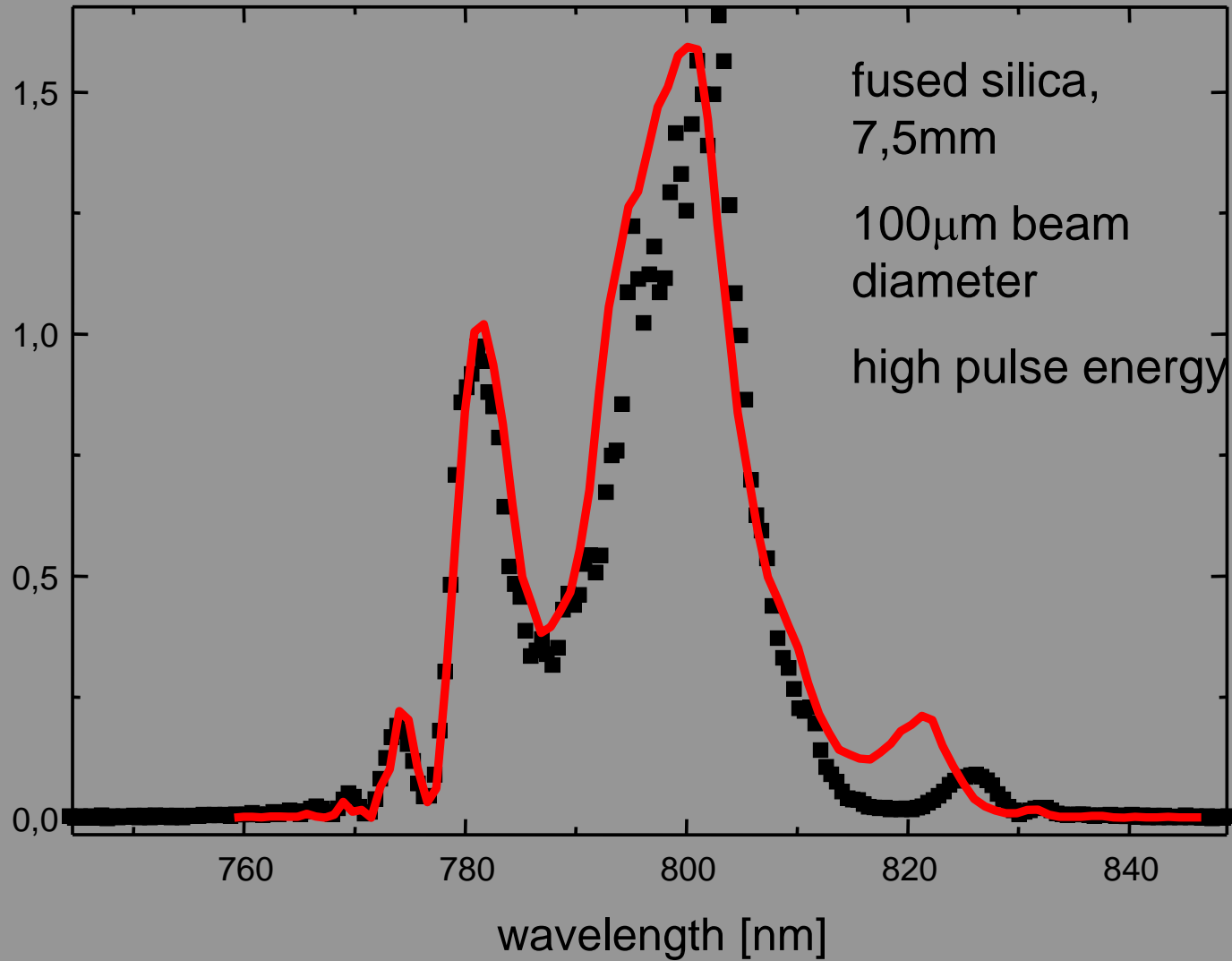
The basic nonlinear optics experiment



Does it fit ? (1)

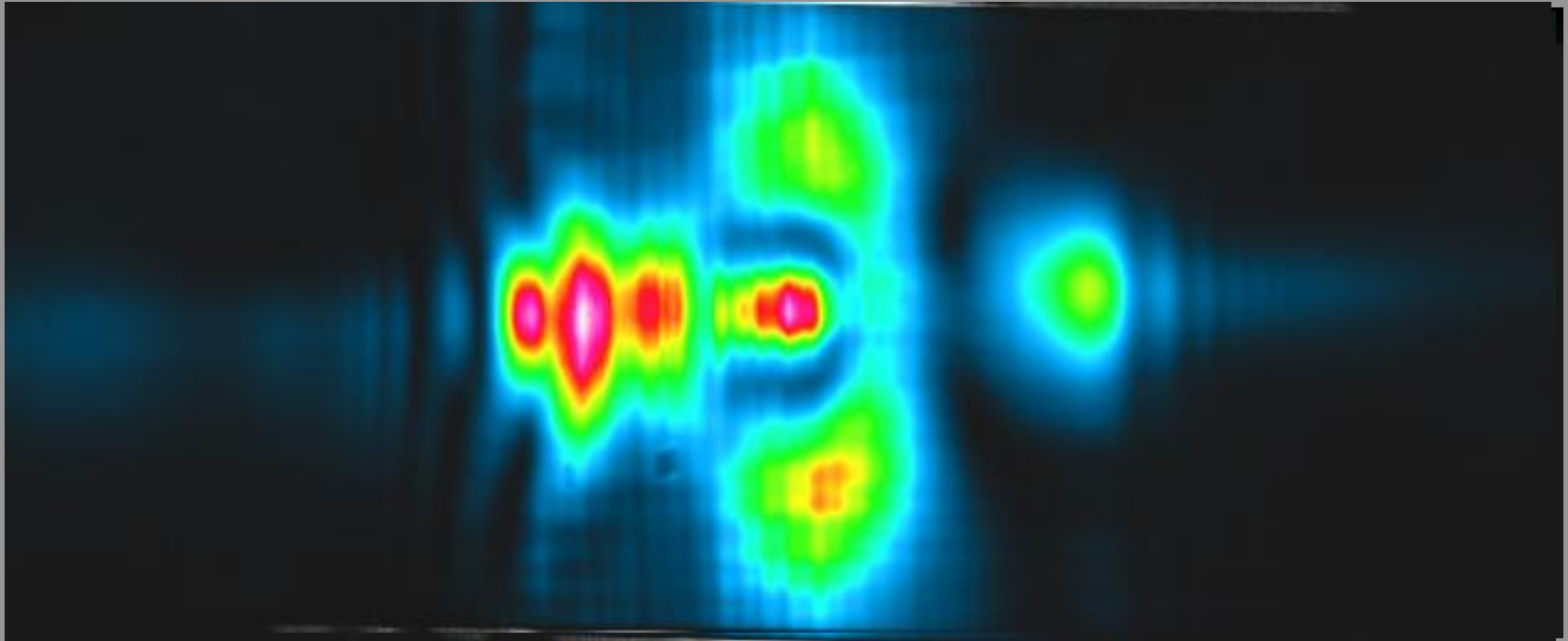


Does it fit ? (2)



Intensity vs. k_{\perp} and λ maps

k_{\perp}



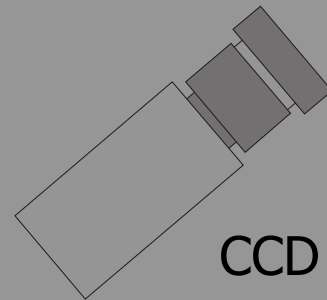
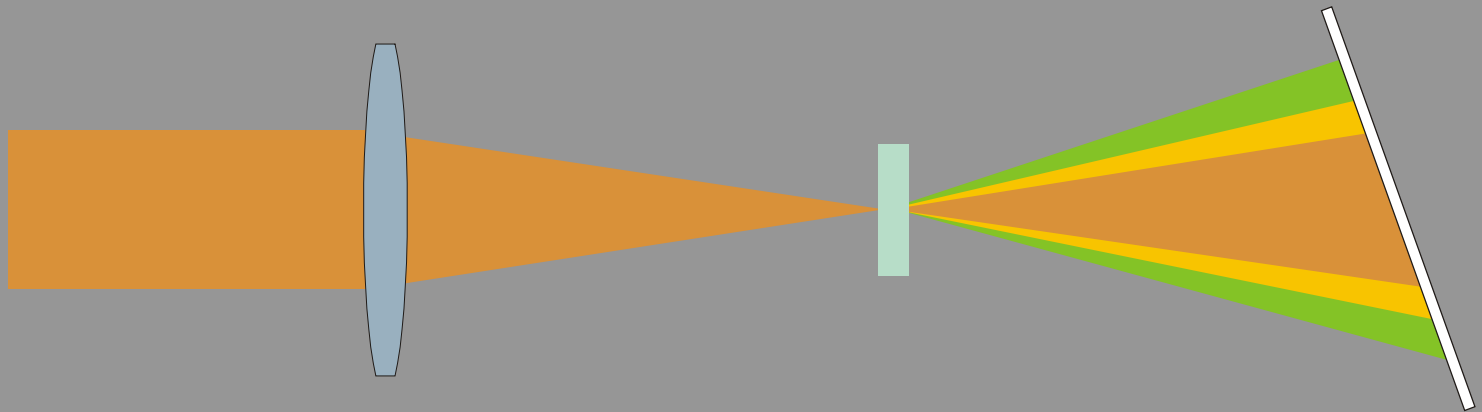
740

860nm

20 μ m input face at the focus

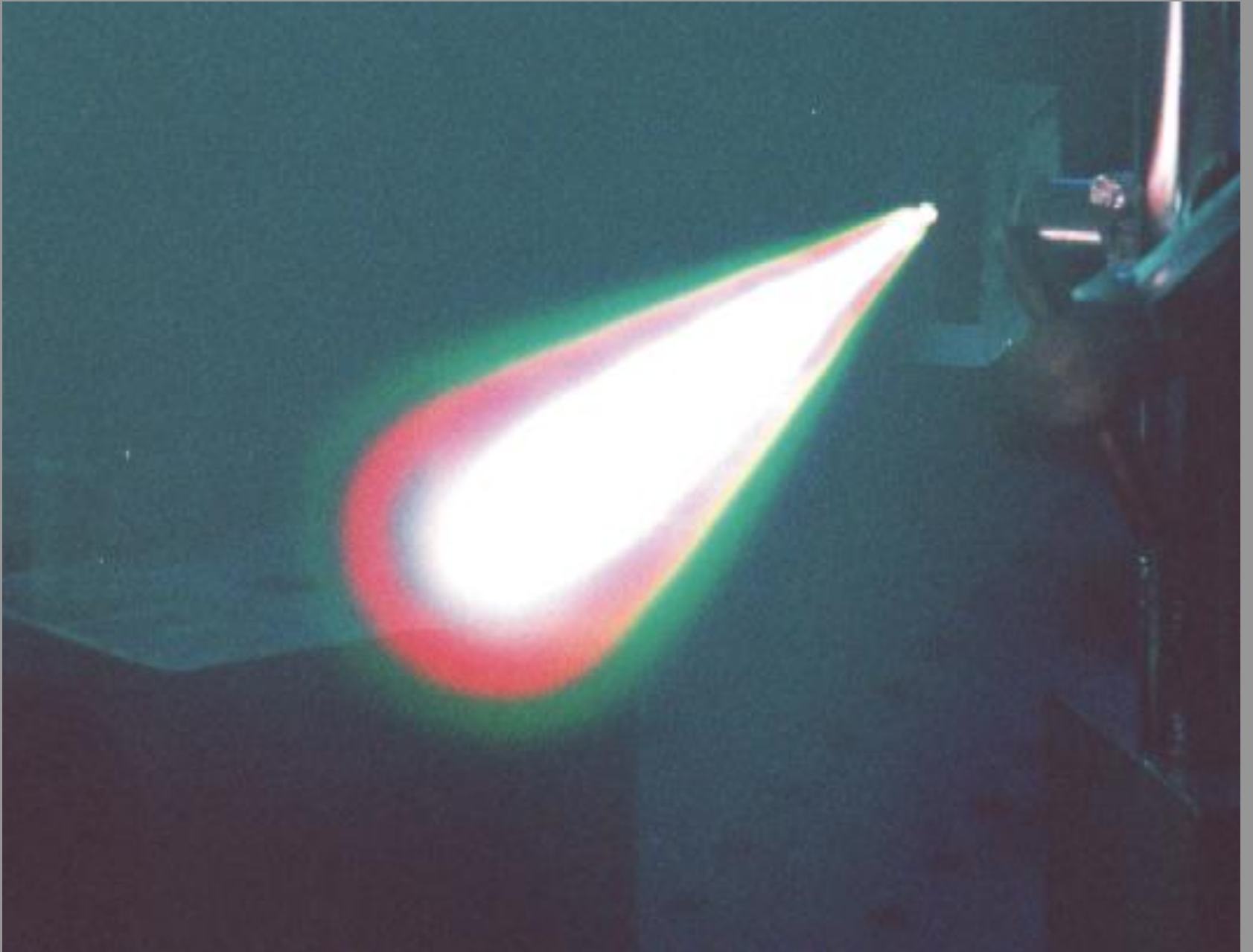
rings

80 fs
< 2 μ J
1 kHz

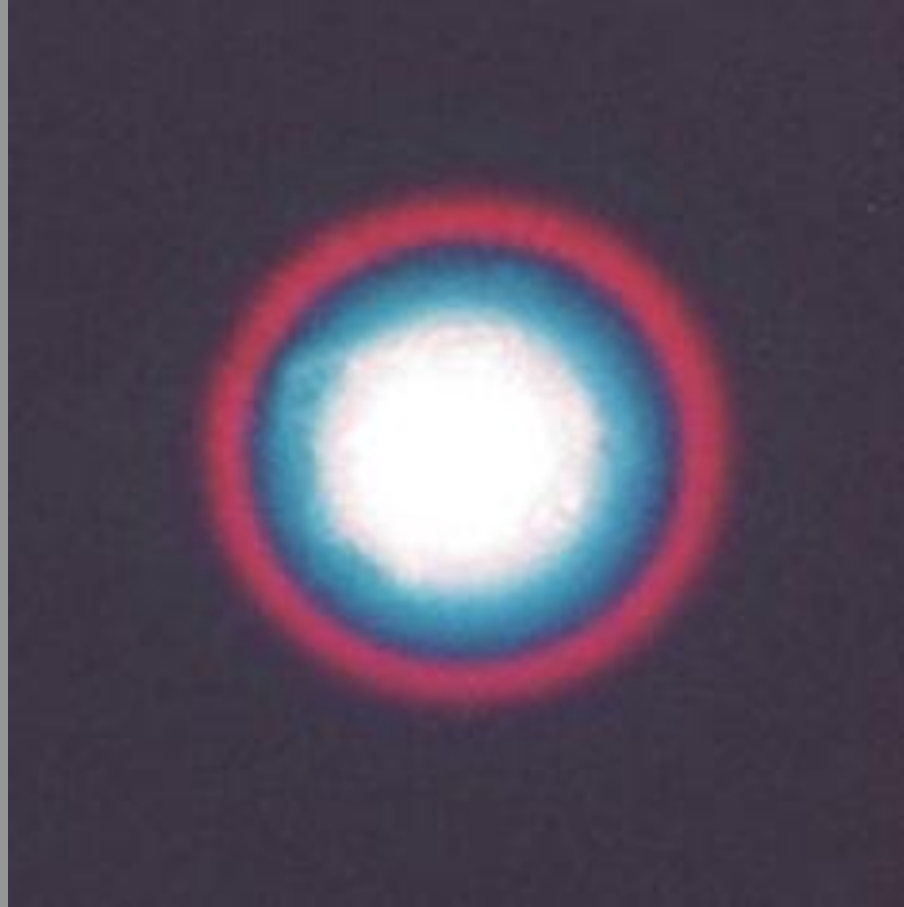


CCD camera

What do you see...



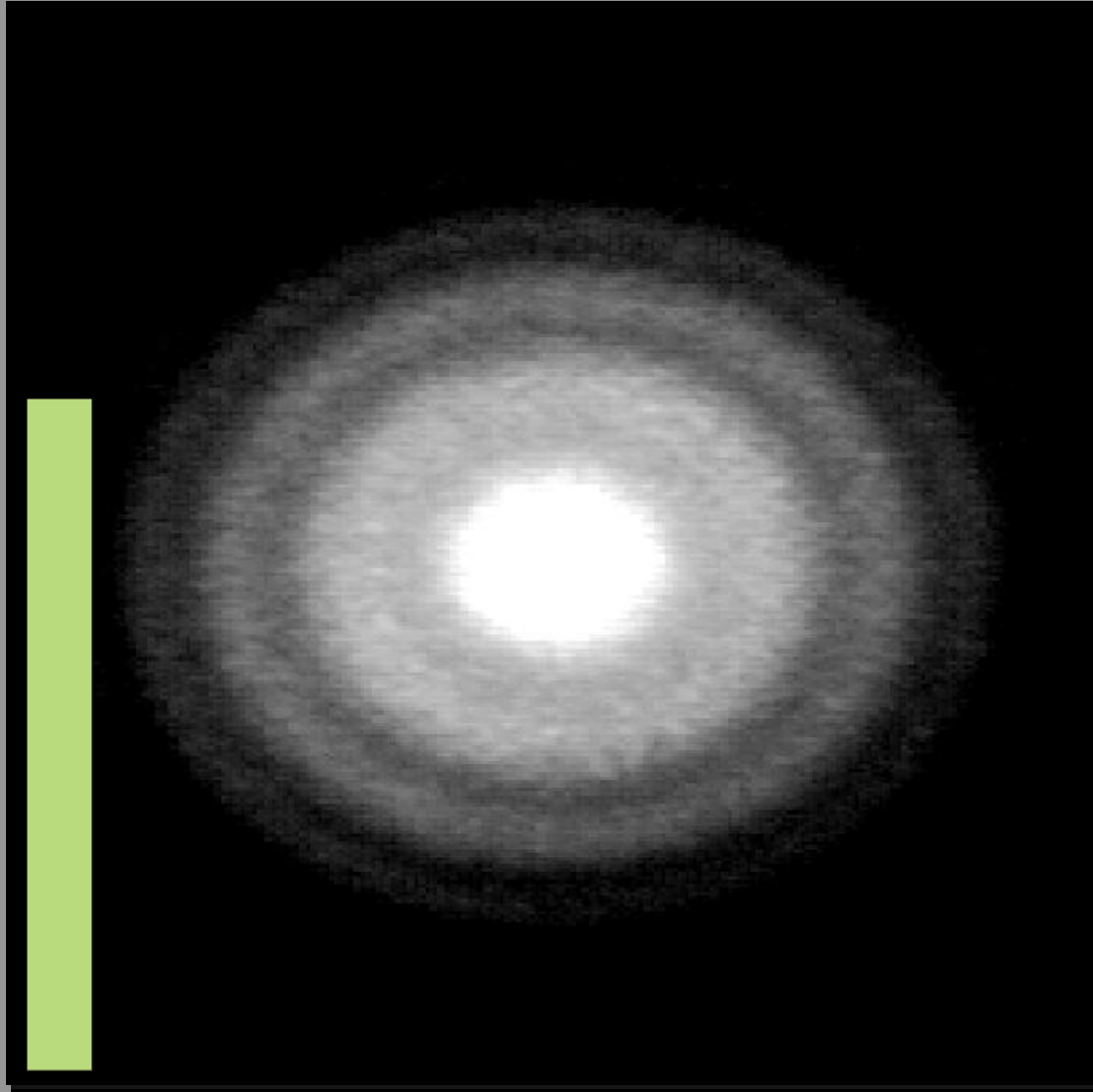
... with a naked eye...



... and in the infrared (800nm)

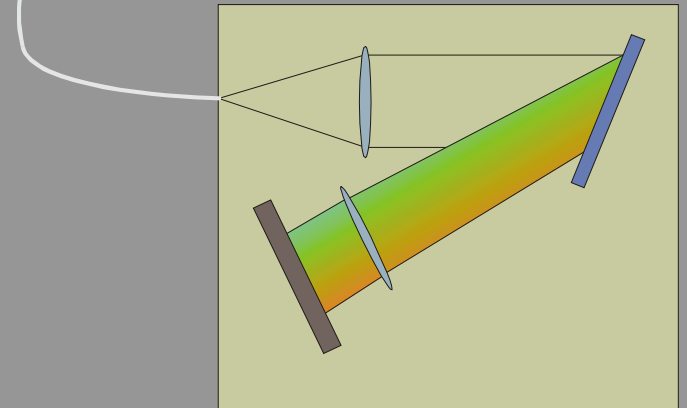
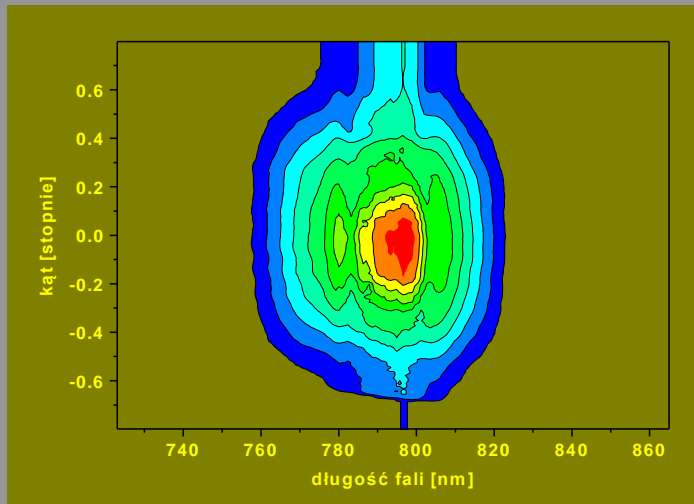
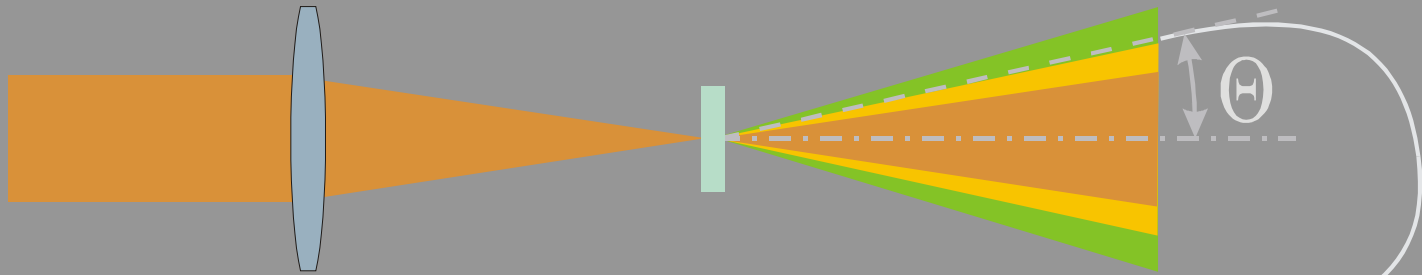
fused silica,
7,5mm

20 μ m

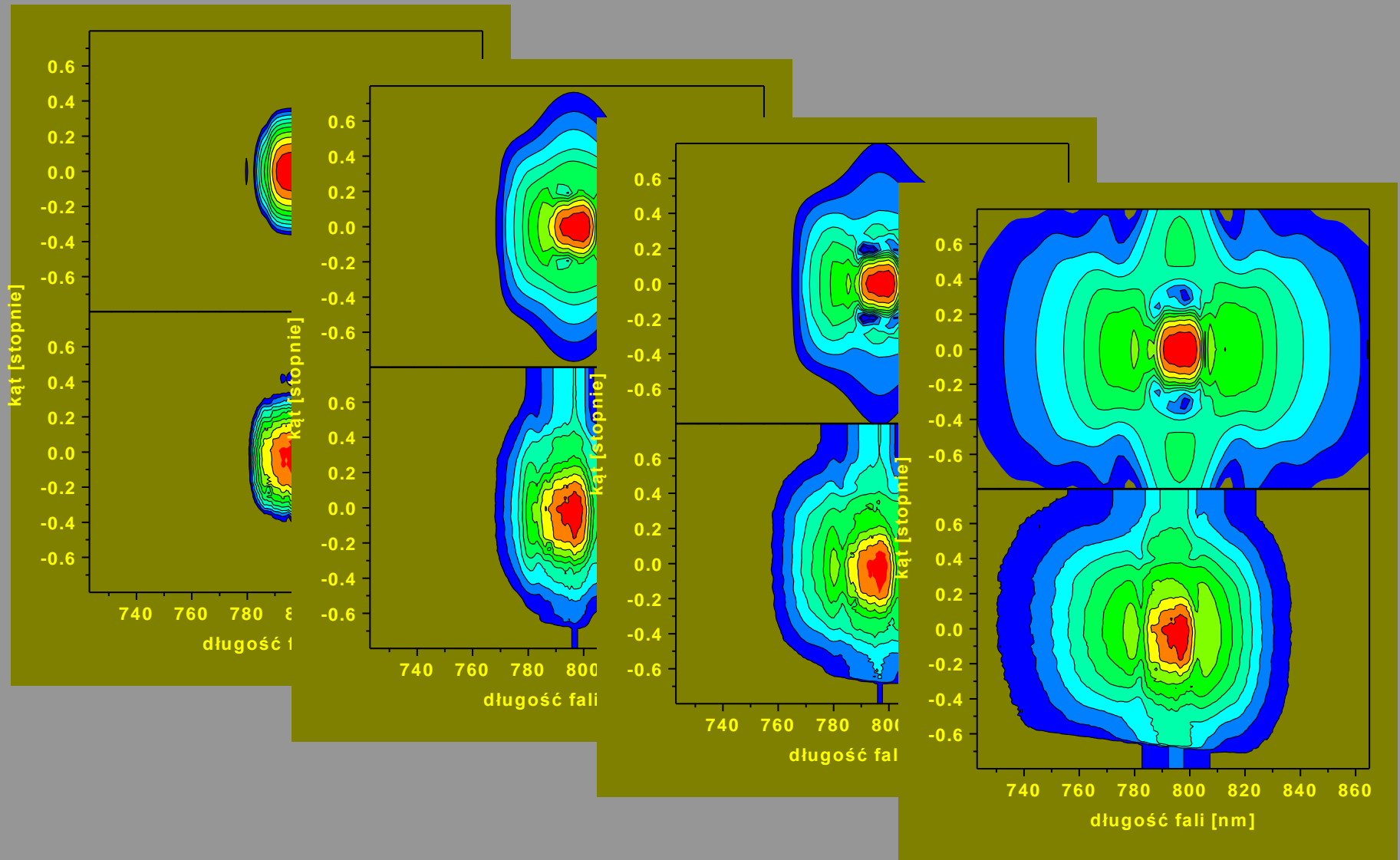


Off-axis spectra

80 fs
< 2 μJ
1 kHz



Off-axis spectra – experiment vs. theory



At high enough intensity, any medium has a nonlinear optical response.

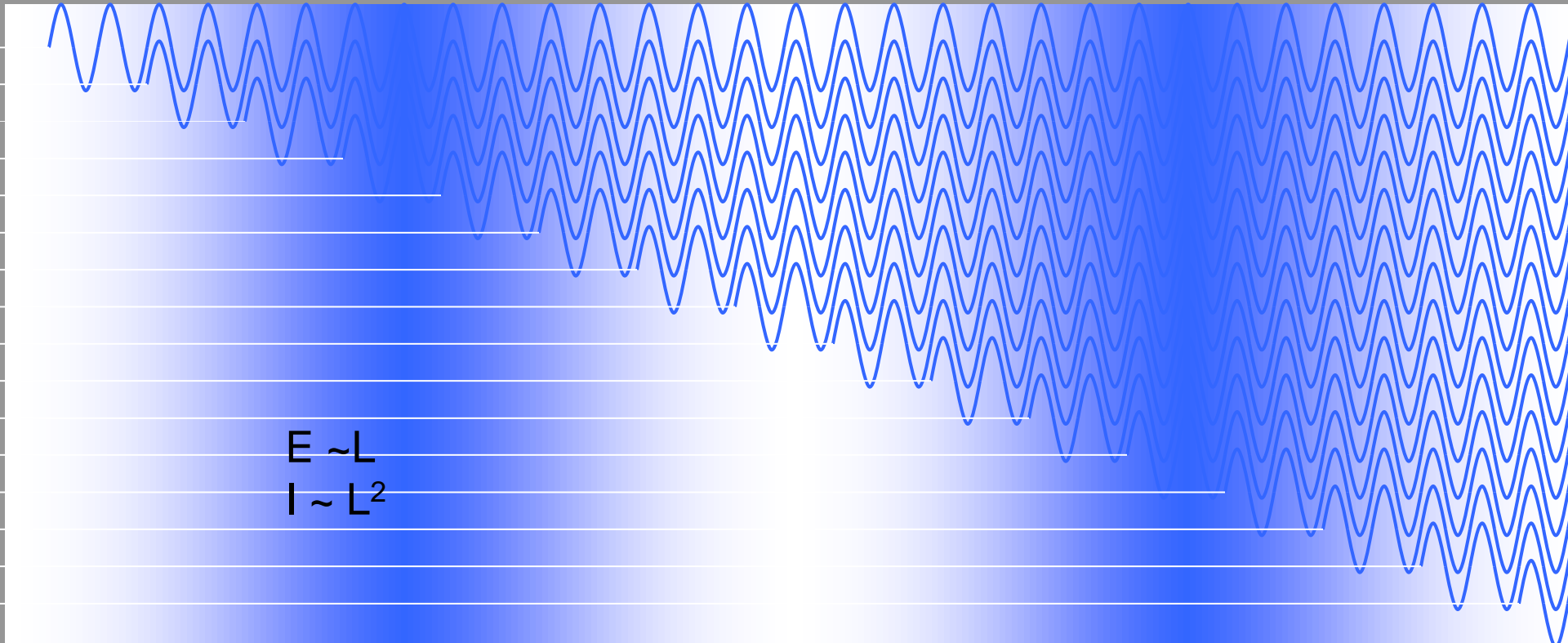
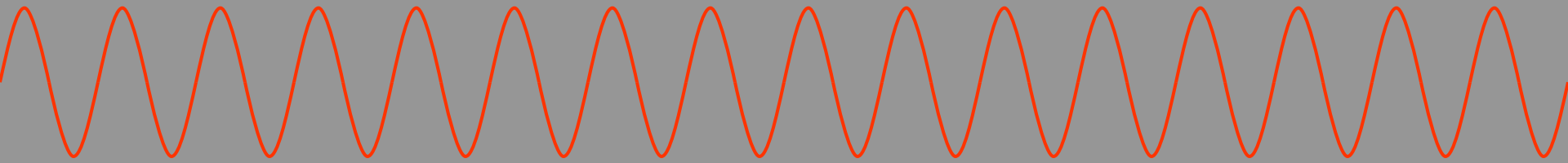
„If you try hard, you can generate second harmonic in dirt”

(prof. W. Gadomski)

But for practical application (i.e. for reasonable conversion efficiency), one more condition must be met...

Phase matching

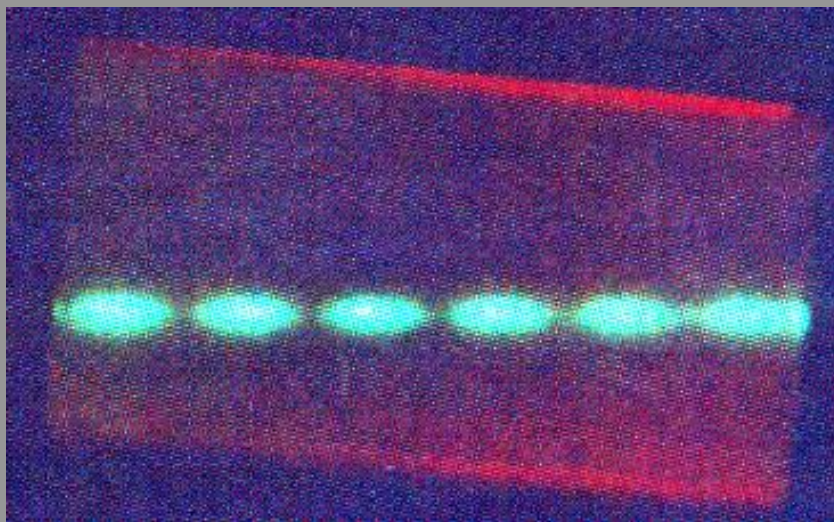
Mismatch



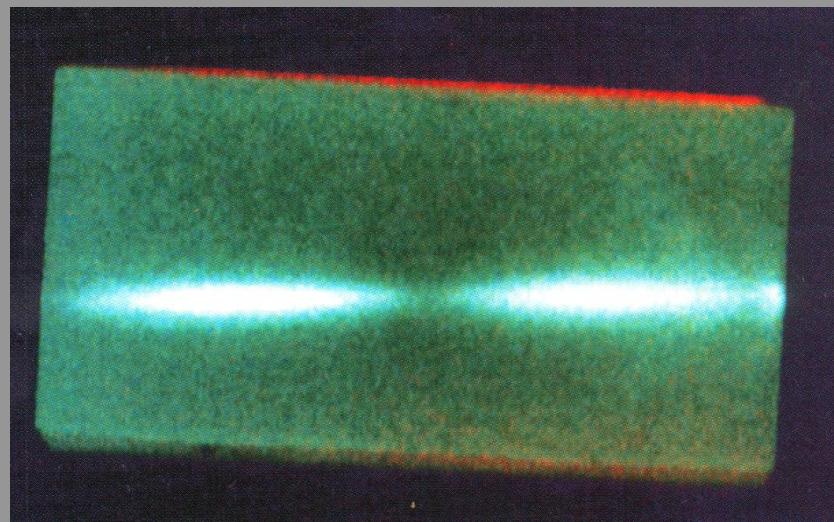
$$E \sim L$$
$$I \sim L^2$$

Sinusoidal dependence of SHG intensity on length

Large Δk

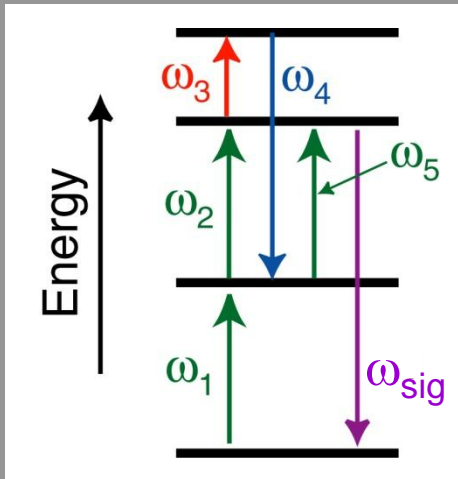


Small Δk



Notice how the intensity is created as the beam passes through the crystal, but, if Δk isn't zero, newly created light is out of phase with previously created light, causing cancellation.

But the signal E-field and polarization k-vectors aren't necessarily equal.

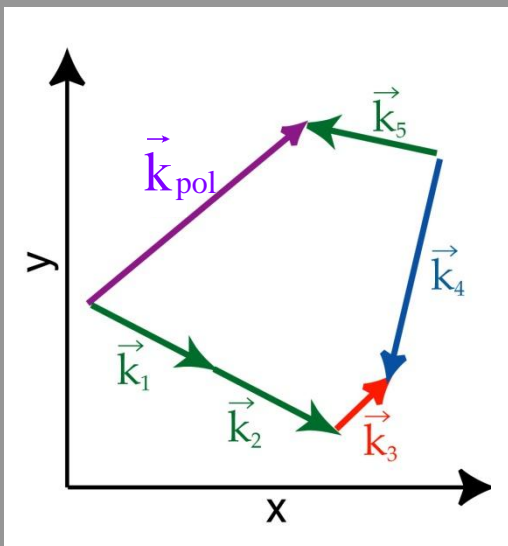


We choose ω_{sig} to be the sum of the input ω 's:

$$\omega_{sig} = \omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5$$

The k-vector mag of light at this frequency is:

$$k_{sig} = \omega_{sig} / c = \omega_{sig} n(\omega_{sig}) / c_0$$



But the k-vector of the polarization is:

$$\vec{k}_{pol} = \vec{k}_1 + \vec{k}_2 + \vec{k}_3 - \vec{k}_4 + \vec{k}_5$$

So k_{pol} may not be the same as k_{sig} !

And we may not be able to cancel the $\exp(-ikz)$'s...

Phase-matching

That k_{pol} may not be the same as k_{sig} is the all-important effect of **phase-matching**. It must be considered in **all** nonlinear-optical problems.

If the k 's don't match, the induced polarization and the generated electric field will drift in and out of phase.

$$E_{sig}(z,t) = E_{sig}(z,t) \exp[i(\omega_{sig} t - k_{sig} z)] \quad P_{sig}(z,t) = P_{sig}(z,t) \exp[i(\omega_{sig} t - k_{pol} z)]$$

The SVEA becomes:

$$\frac{\partial E_{sig}}{\partial z} = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \exp(i \Delta k z)$$



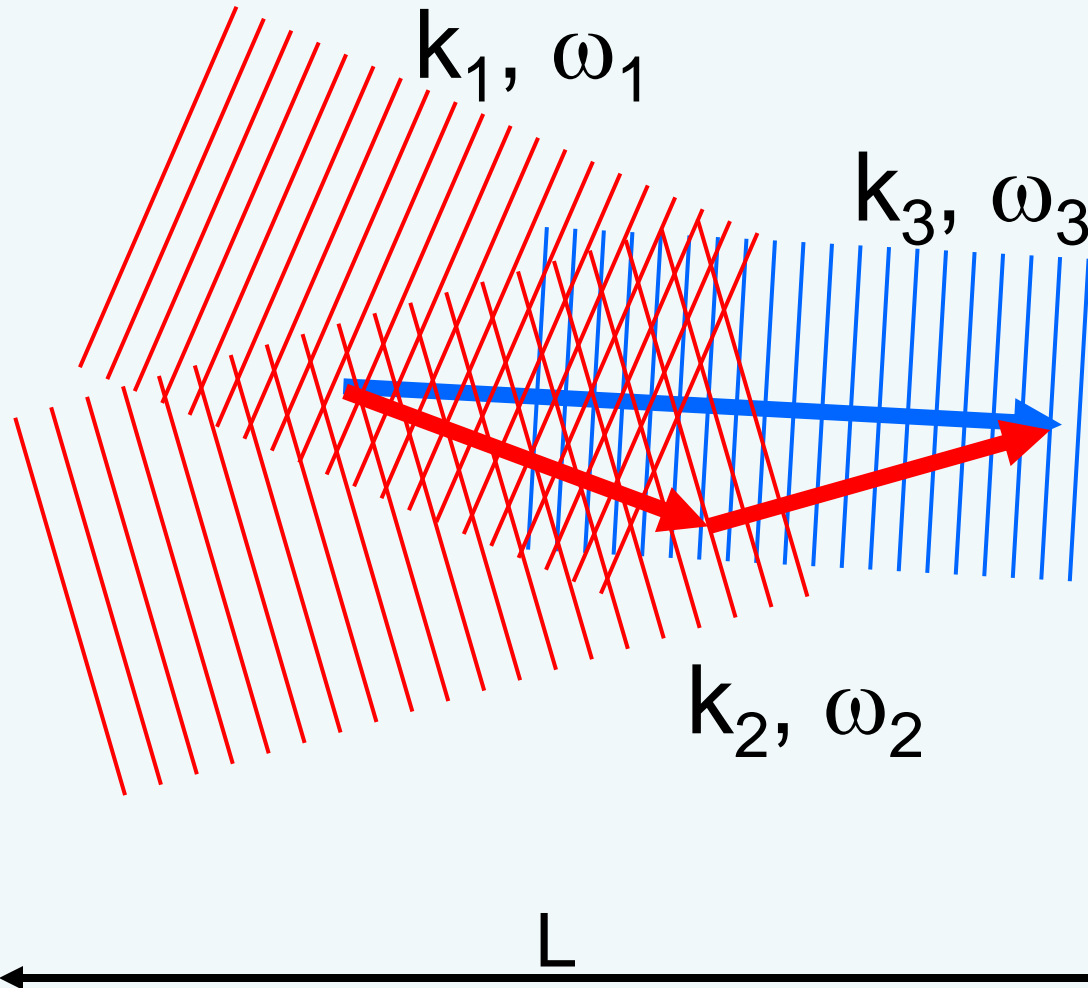
where:

$$\Delta k \equiv k_{pol} - k_{sig}$$

Integrating the SVEA in this case over the length of the medium (L) yields:

$$E_{sig}(L,t) = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \left[\frac{\exp(i \Delta k z)}{i \Delta k} \right]_0^L$$

3 wave mixing in the... wave picture



$$\omega_3 = \omega_1 + \omega_2$$

$$k_{3\perp} = k_{1\perp} + k_{2\perp}$$

Efficiency

$$\sim [\sin(\Delta k L/2)/\Delta k]^2$$

$$\Delta k = k_{3z} - k_{1z} - k_{2z}$$

Phase matching – first notions

VOLUME 8, NUMBER 1

PHYSICAL REVIEW LETTERS

JANUARY 1, 1962

EFFECTS OF DISPERSION AND FOCUSING ON THE PRODUCTION OF OPTICAL HARMONICS

VOLUME 8, NUMBER 1

PHYSICAL REVIEW LETTERS

JANUARY 1, 1962

MIXING OF LIGHT BEAMS IN CRYSTALS

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(Received November 29, 1961)

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This Letter reports the mixing of plane light waves having different directions of propagation and the attainment of coherence volumes of about 0.2 cm^3 in the production of second harmonic radiation. In these experiments a sizable intensity of second harmonic radiation is observed in potassium dihydrogen phosphate (KDP) without the use of focussed beams. This is one of a new class of optical experiments and techniques suggested by the work of Franken et al.¹ and made possible by maser sources of highly intense monochromatic light.

A plane light wave described by the form $\exp[i(\omega_1 t - \vec{k}_1 \cdot \vec{r})]$ launches in a suitable¹ nonlinear medium a travelling wave of polarization of the form $\exp[i(2\omega_1 t - 2\vec{k}_1 \cdot \vec{r})]$. This polarization can radiate a wave \vec{k}_2 at frequency $\omega_2 = 2\omega_1$ with maximum efficiency if the two remain in phase, that is, if $\vec{k}_2 = 2\vec{k}_1$. This condition implies that in the direction of \vec{k}_1 the phase velocity $v_1 = v_2$ or $n_1 = n_2$. Since almost all materials have normal dispersion

When Eq. (1)² is satisfied, the entire irradiated crystal volume can radiate coherently, electromagnetic momentum is conserved, and optimum radiation efficiency is achieved. Consider the particular case in a negative uniaxial crystal of an O wave \vec{k}_1 at angle ψ_1 with the optic axis \hat{z} mixing with a second O wave \vec{k}_1' to form the harmonic E wave \vec{k}_2 emitted at an angle θ to \vec{k}_1 . Equation (1) is satisfied if $\cos\theta = v_1/v_2(\theta)$. This transcendental equation has particularly simple solutions when $\psi_1 - \psi_0 = \Delta\psi$ is small, i.e., when the direction of \vec{k}_1 is close to the direction for which $v_1 = v_2$. Then Eq. (1) can be approximated by $\theta^2 = 2(v_2 - v_1)/v_2 = K(\Delta\psi - \theta \cos\alpha)$, where K denotes the constant $2[(v_2^e)^2 - (v_1^o)^2]^{1/2} / [2[(v_1^o)^2 - (v_2^o)^2]^{1/2} (v_1^o)^{-2}]$ and α is the angle between \vec{k}_1, \vec{k}_2 and \hat{z}, \vec{k}_1 planes. The solution of this quadratic equation in θ for a particular \vec{k}_1 represents a circular cone of wave vectors \vec{k}_2 of half-angle $\frac{1}{2}K(1 + 4\Delta\psi/K)^{1/2}$ centered about the direction in the \vec{k}_1, \hat{z} plane making an angle $\psi + \frac{1}{2}K$ with the optic axis. No completely in-

J. A. Armstrong, N. Bloembergen, J. Ducuing, P. S. Pershan, *Interactions between Light Waves in a Nonlinear Dielectric*, Phys. Rev. **127**, 1918–1939 (1962)

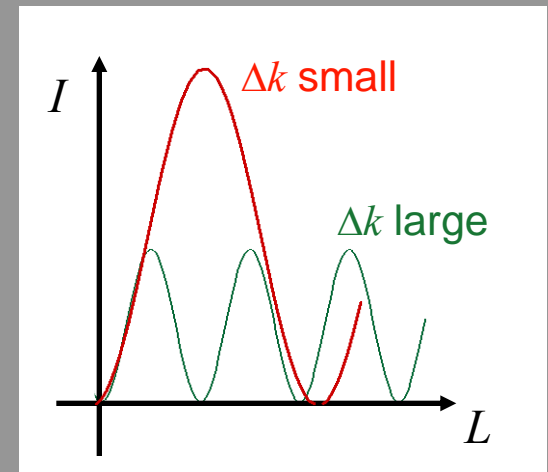
Phase-matching (continued)

$$\begin{aligned}
 E_{sig}(L,t) &= -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \left[\frac{\exp(i \Delta k z)}{i \Delta k} \right]_0^L \\
 &= -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \left[\frac{\exp(i \Delta k L) - 1}{i \Delta k} \right] \\
 &= -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \exp(i \Delta k L/2) \left[\frac{\exp(i \Delta k L/2) - \exp(-i \Delta k L/2)}{i \Delta k} \right] \\
 &= -i \frac{\mu_0 \omega_{sig}^2}{k_{sig}} P_{sig} \exp(i \Delta k L/2) \left[\frac{\exp(i \Delta k L/2) - \exp(-i \Delta k L/2)}{2i \Delta k} \right] \\
 &= -i \frac{\mu_0 \omega_{sig}^2}{k_{sig}} P_{sig} \exp(i \Delta k L/2) \left[\frac{\sin(\Delta k L/2)}{\Delta k} \right]
 \end{aligned}$$

So:

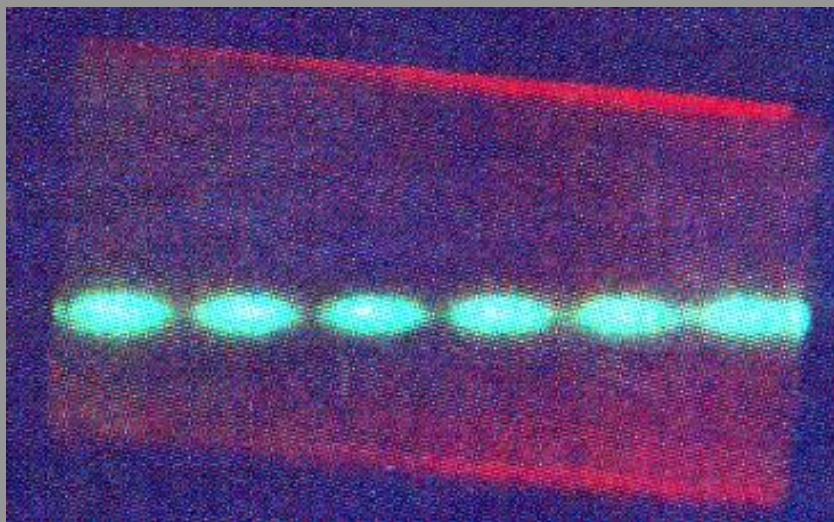
$$E_{sig}(L,t) \propto \sin(\Delta k L/2) / \Delta k$$

$$I_{sig}(L,t) \propto \sin^2(\Delta k L/2) / \Delta k^2 \longrightarrow$$

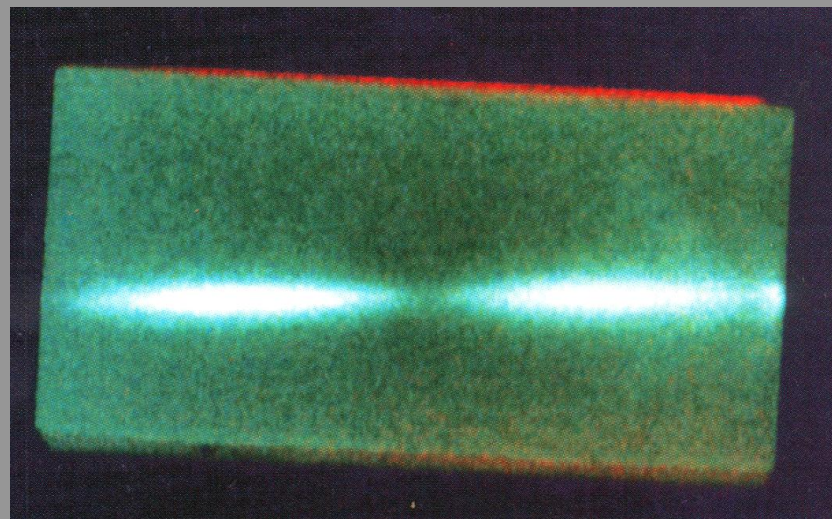


Sinusoidal dependence of SHG intensity on length

Large Δk



Small Δk



Notice how the intensity is created as the beam passes through the crystal, but, if Δk isn't zero, newly created light is out of phase with previously created light, causing cancellation.

The ubiquitous $\text{sinc}^2(\Delta k L / 2)$

Recall that:

$$E_{sig}(L, t) = -i \frac{\mu_0 \omega_{sig}^2}{k_{sig}} P_{sig} \exp(i \Delta k L / 2) \left[\frac{\sin(\Delta k L / 2)}{\Delta k} \right]$$

Multiplying and dividing by $L/2$:

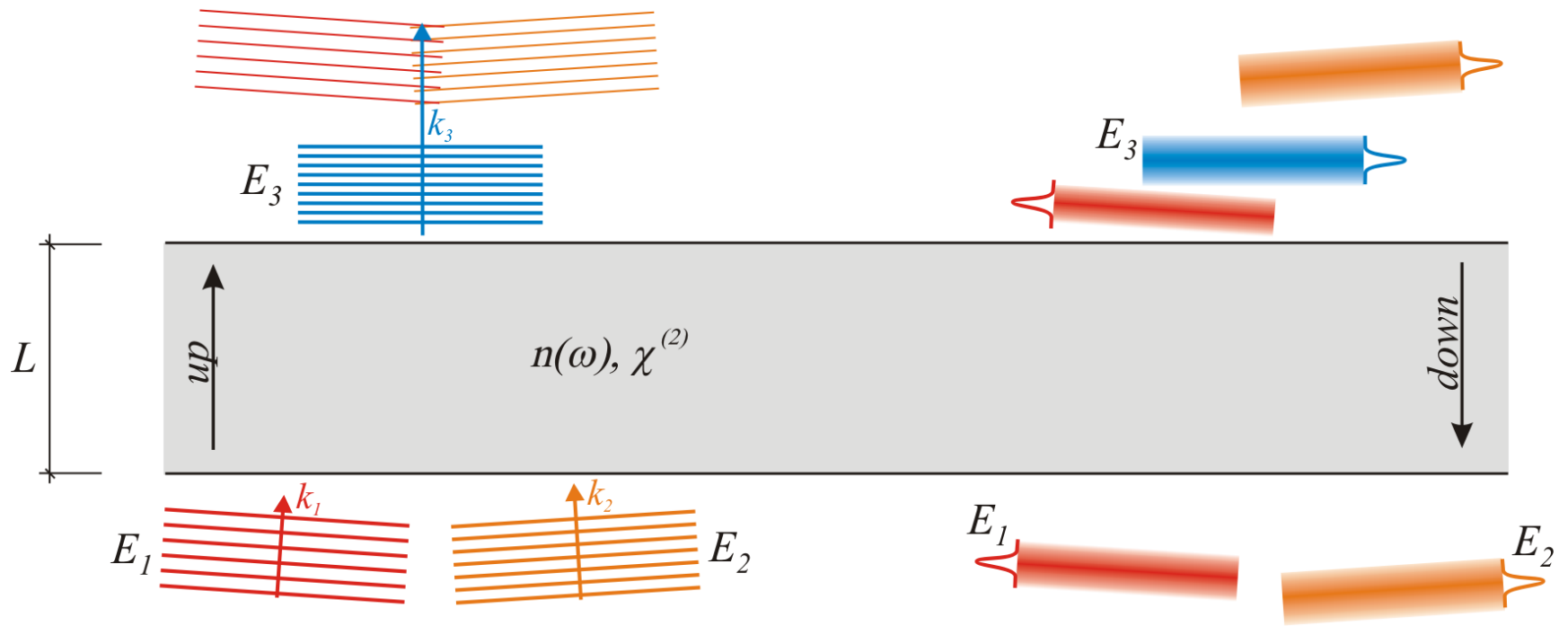
$$\begin{aligned} &= -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \exp(i \Delta k L / 2) L \left[\frac{\sin(\Delta k L / 2)}{\Delta k L / 2} \right] \\ &= -\frac{i}{2} c \mu_0 \omega_{sig} P_{sig} \exp(i \Delta k L / 2) L \text{sinc}(\Delta k L / 2) \end{aligned}$$

$$E_{sig}(L, t) \propto P_{sig} L \text{sinc}(\Delta k L / 2)$$

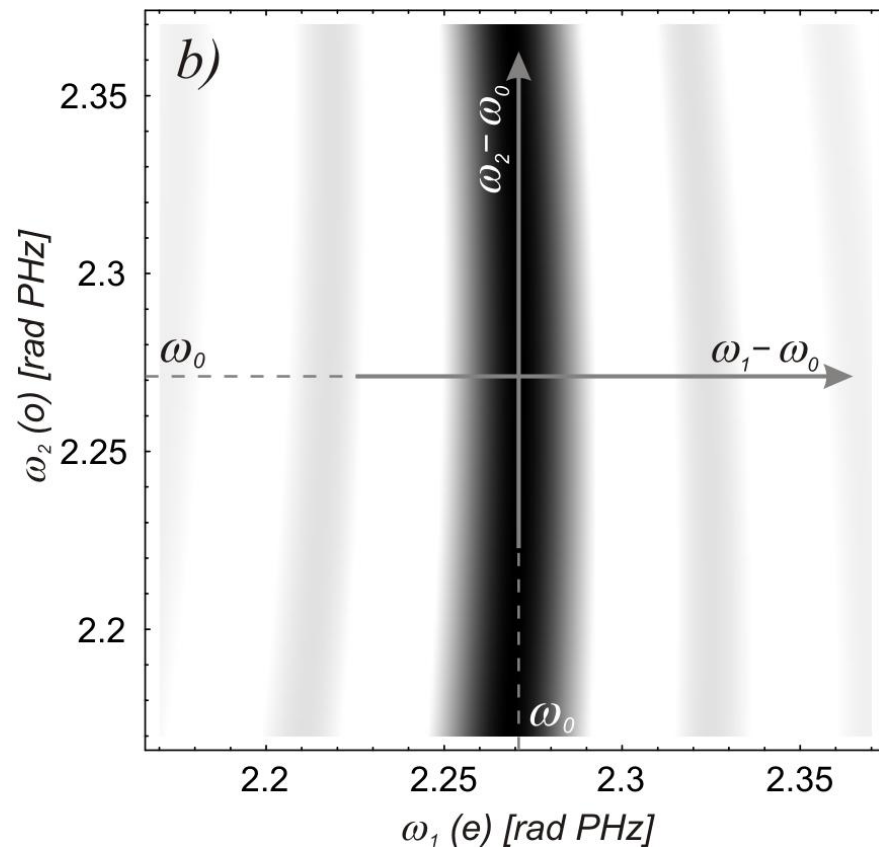
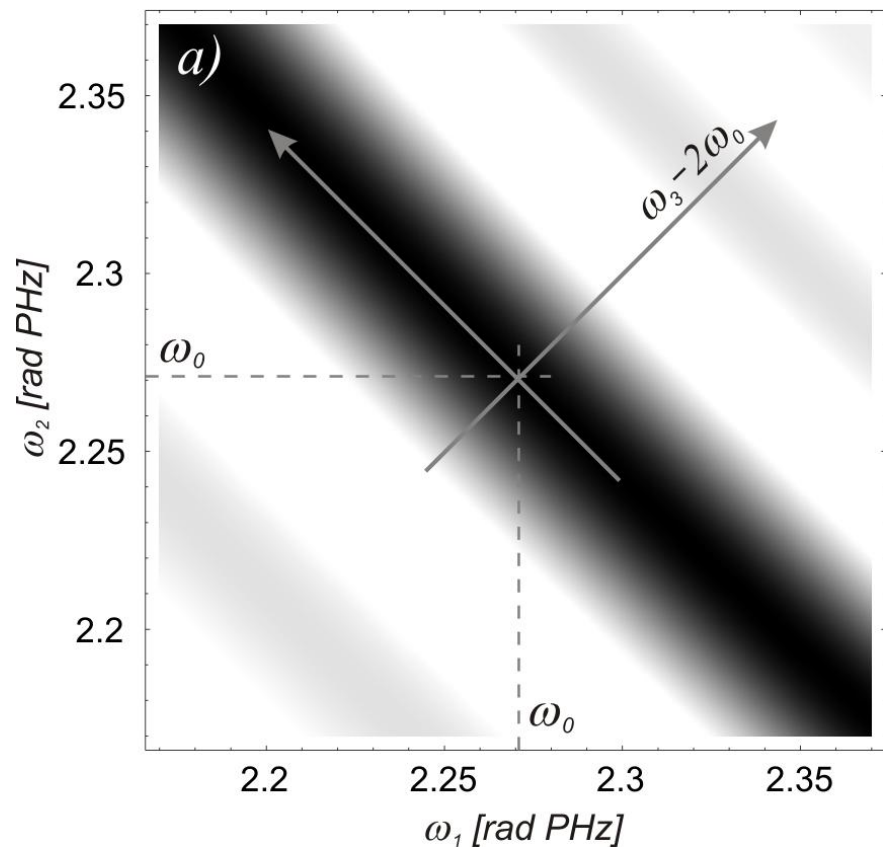
$$I_{sig}(L, t) \propto |P_{sig}|^2 L^2 \text{sinc}^2(\Delta k L / 2)$$

Phase Mismatch almost always yields a $\text{sinc}^2(\Delta k L / 2)$ dependence.

Wave mixing for CW and pulsed fields



type I and type II in KDP



Solving the wave equation in nonlinear optics

Recall the inhomogeneous wave equation:

$$\frac{\partial^2 \mathbf{E}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}$$

Take into account the linear polarization by replacing c_0 with c .

Because it's second-order in both space and time, and \mathbf{P} is a nonlinear function of \mathbf{E} , we can't easily solve this equation. Indeed, nonlinear differential equations are really hard.

We'll have to make approximations...

Separation-of-frequencies approximation

The total E-field will contain several nearly discrete frequencies, ω_1, ω_2 , etc.

So we'll write separate wave equations for each frequency, considering only the induced polarization at the given frequency:

$$\frac{\partial^2 E_1}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E_1}{\partial t^2} = \mu_0 \frac{\partial^2 P_1}{\partial t^2}$$

where E_1 and P_1 are the E-field and polarization at frequency ω_1 .

$$\frac{\partial^2 E_2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E_2}{\partial t^2} = \mu_0 \frac{\partial^2 P_2}{\partial t^2}$$

where E_2 and P_2 are the E-field and polarization at frequency ω_2 .

etc.

This will be a reasonable approximation even for relatively broadband ultrashort pulses

The non-depletion assumption

We'll also assume that the nonlinear-optical effect is weak, so we can assume that the fields at the input frequencies won't change much. This assumption is called **non-depletion**.

As a result, we need only consider the wave equation for the field and polarization oscillating at the new **signal** frequency, ω_{sig} .

$$\frac{\partial^2 E_{sig}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E_{sig}}{\partial t^2} = \mu_0 \frac{\partial^2 P_{sig}}{\partial t^2}$$

where E_{sig} and P_{sig} are the E-field and polarization at frequency ω_{sig} .

The Slowly Varying Envelope Approximation

We'll write the pulse E-field as a product of an envelope and complex exponential:

$$E_{sig}(z,t) = E_{sig}(z,t) \exp[i(\omega_{sig} t - k_{sig} z)]$$

We'll assume that the new pulse envelope won't change too rapidly. This is the Slowly Varying Envelope Approximation (SVEA).

If d is the length scale for variation of the envelope, SVEA says: $d \gg \lambda_{sig}$

$$\left| \frac{\partial E_{sig}}{\partial z} \right| \sim \left| \frac{E_{sig}}{d} \right| \ll \left| 2\pi \frac{E_{sig}}{\lambda_{sig}} \right| = \left| k_{sig} E_{sig} \right|$$

$$\left| \frac{\partial^2 E_{sig}}{\partial z^2} \right| \sim \left| \frac{\partial E_{sig} / \partial z}{d} \right| \ll \left| 2\pi \frac{\partial E_{sig} / \partial z}{\lambda_{sig}} \right| = \left| k_{sig} \frac{\partial E_{sig}}{\partial z} \right|$$

Comparing E_{sig} and its derivatives:

$$\left| \frac{\partial^2 E_{sig}}{\partial z^2} \right| \ll \left| k_{sig} \frac{\partial E_{sig}}{\partial z} \right| \ll \left| k_{sig}^2 E_{sig} \right|$$

The Slowly Varying Envelope Approximation (continued)

We'll do the same in time:

If τ is the time scale for variation of the envelope, SVEA says: $\tau \gg T_{sig}$ where T_{sig} is one optical period, $2\pi/\omega_{sig}$.

$$\left| \frac{\partial E_{sig}}{\partial t} \right| \sim \left| \frac{E_{sig}}{\tau} \right| \ll \left| 2\pi \frac{E_{sig}}{T_{sig}} \right| = \left| \omega_{sig} E_{sig} \right|$$

$$\left| \frac{\partial^2 E_{sig}}{\partial t^2} \right| \sim \left| \frac{\partial E_{sig} / \partial t}{\tau} \right| \ll \left| 2\pi \frac{\partial E_{sig} / \partial t}{T_{sig}} \right| = \left| \omega_{sig} \frac{\partial E_{sig}}{\partial t} \right|$$

Comparing E_{sig} and its time derivatives:

$$\left| \frac{\partial^2 E_{sig}}{\partial t^2} \right| \ll \left| \omega_{sig} \frac{\partial E_{sig}}{\partial t} \right| \ll \left| \omega_{sig}^2 E_{sig} \right|$$

The Slowly Varying Envelope Approximation (continued)

And we'll do the same for the polarization:

$$P_{sig}(z,t) = P_{sif}(z,t) \exp[i(\omega_{sig} t - k_{sig} z)]$$

If τ is the time scale for variation of the envelope, SVEA says: $\tau \gg T_{sig}$ where T_{sig} is one optical period, $2\pi/\omega_{sig}$.

$$\left| \frac{\partial P_{sig}}{\partial t} \right| \sim \left| \frac{P_{sig}}{\tau} \right| \ll \left| 2\pi \frac{P_{sig}}{T_{sig}} \right| = \left| \omega_{sig} P_{sig} \right|$$

$$\left| \frac{\partial^2 P_{sig}}{\partial t^2} \right| \sim \left| \frac{\partial P_{sig} / \partial t}{\tau} \right| \ll \left| 2\pi \frac{\partial P_{sig} / \partial t}{T_{sig}} \right| = \left| \omega_{sig} \frac{\partial P_{sig}}{\partial t} \right|$$

Comparing P_{sig} and its time derivatives:

$$\left| \frac{\partial^2 P_{sig}}{\partial t^2} \right| \ll \left| \omega_{sig} \frac{\partial P_{sig}}{\partial t} \right| \ll \left| \omega_{sig}^2 P_{sig} \right|$$

SVEA (continued)

$$E_{sig}(z,t) = E_{sig}(z,t) \exp[i(\omega_{sig} t - k_{sig} z)]$$

Computing the derivatives: $\frac{\partial E_{sig}}{\partial z} = \left[\frac{\partial E_{sig}}{\partial z} - ik_{sig} E_{sig} \right] \exp[i(\omega_{sig} t - k_{sig} z)]$

$$\frac{\partial^2 E_{sig}}{\partial z^2} = \left[\frac{\partial^2 E_{sig}}{\partial z^2} - 2ik_{sig} \frac{\partial E_{sig}}{\partial z} - k_{sig}^2 E_{sig} \right] \exp[i(\omega_{sig} t - k_{sig} z)]$$

Similarly, $\frac{\partial^2 E_{sig}}{\partial t^2} = \left[\frac{\partial^2 E_{sig}}{\partial t^2} + 2i\omega_{sig} \frac{\partial E_{sig}}{\partial t} - \omega_{sig}^2 E_{sig} \right] \exp[i(\omega_{sig} t - k_{sig} z)]$

$$\frac{\partial^2 P_{sig}}{\partial t^2} = \left[\frac{\partial^2 P_{sig}}{\partial t^2} + 2i\omega_{sig} \frac{\partial P_{sig}}{\partial t} - \omega_{sig}^2 P_{sig} \right] \exp[i(\omega_{sig} t - k_{sig} z)]$$

Neglect all 2nd derivatives of envelopes with respect to z and t .

Also, neglect the 1st derivative of the polarization envelope (it's small compared to the $\omega_{sig}^2 P_{sig}$ term). We must keep E_{sig} 's first derivatives, as we'll see in the next slide...

The Slowly Varying Envelope Approximation

Substituting the remaining derivatives into the inhomogeneous wave equation for the signal field at ω_0 :

$$\frac{\partial^2 E_{sig}}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E_{sig}}{\partial t^2} = \mu_0 \frac{\partial^2 P_{sig}}{\partial t^2}$$

$$\left[-2ik_0 \frac{\partial E_0}{\partial z} - \frac{2i\omega_{sig}}{c^2} \frac{\partial E_{sig}}{\partial t} - k_{sig}^2 E_{sig} + \frac{\omega_{sig}^2}{c^2} E_{sig} \right] \exp[i(\omega_{sig}t - k_{sig}z)] = -\mu_0 \omega_{sig}^2 P_{sig} \exp[i(\omega_{sig}t - k_{sig}z)]$$

Now, because $k_0 = \omega_0 / c$, the last two bracketed terms cancel. And we can cancel the complex exponentials, leaving:

$$2ik_{sig} \frac{\partial E_{sig}}{\partial z} + \frac{2i\omega_{sig}}{c^2} \frac{\partial E_{sig}}{\partial t} = \mu_0 \omega_{sig}^2 P_{sig}$$

Dividing by $2ik_0$:

$$\frac{\partial E_{sig}}{\partial z} + \frac{1}{c} \frac{\partial E_{sig}}{\partial t} = -i \frac{\mu_0 \omega_{sig}^2}{2k_0} P_{sig}$$

Slowly Varying
Envelope Approximation

Including dispersion in the SVEA

We can include dispersion by Fourier-transforming, expanding $k_{sig}(\omega)$ to first order in ω , and transforming back. This replaces c with v_g :

$$\frac{\partial E_{sig}}{\partial z} + \frac{1}{v_g} \frac{\partial E_{sig}}{\partial t} = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig}$$

We can include GVD also expanding to 2nd order, yielding:

$$\frac{\partial E_{sig}}{\partial z} + \frac{1}{v_g} \frac{\partial E_{sig}}{\partial t} - \frac{i}{2} \frac{d^2 k_{sig}}{d\omega_{sig}^2} \frac{\partial^2 E_{sig}}{\partial t^2} = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig}$$

We can understand most nonlinear-optical effects best by neglecting GVD, so we will, but this extra term can become important for very very short (i.e., very broadband) pulses.

Transforming to a moving co-ordinate system

Define a moving co-ordinate system:

$$z_v = z$$

$$t_v = t - z / v_g$$

Transforming the derivatives:

$$\frac{\partial E_{sig}}{\partial z} = \frac{\partial E_{sig}}{\partial z_v} \frac{\partial z_v}{\partial z} + \frac{\partial E_{sig}}{\partial t_v} \frac{\partial t_v}{\partial z}$$

$$\frac{\partial E_{sig}}{\partial t} = \frac{\partial E_{sig}}{\partial z_v} \frac{\partial z_v}{\partial t} + \frac{\partial E_{sig}}{\partial t_v} \frac{\partial t_v}{\partial t}$$

$$\frac{\partial E_{sig}}{\partial z} = \frac{\partial E_{sig}}{\partial z_v} + \frac{\partial E_{sig}}{\partial t_v} \left[-\frac{1}{v_g} \right]$$

$$\frac{\partial E_{sig}}{\partial t} = 0 + \frac{\partial E_{sig}}{\partial t_v}$$

The SVEA becomes:

$$\frac{\partial E_{sig}}{\partial z_v} + \frac{\partial E_{sig}}{\partial t_v} \left[-\frac{1}{v_g} \right] + \frac{1}{v_g} \left[\frac{\partial E_{sig}}{\partial t_v} \right] = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} \quad \text{The time derivatives cancel!}$$

Canceling terms, the SVEA becomes:

$$\frac{\partial E_{sig}}{\partial z_v} = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig}$$

We'll drop the subscript (v) to simplify our equations.

Integrating the SVEA

$$\frac{\partial E_{sig}}{\partial z} = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig}$$

Usually, $P_{sig} = P_{sig}(z, t)$, and even this simple equation can be difficult to solve (integrate). For now, we'll just assume that P_{sig} is a constant, and the integration becomes trivial:

$$E_{sig}(z, t) = -i \frac{\mu_0 \omega_{sig}^2}{2k_{sig}} P_{sig} z \quad \text{when } P_0 \text{ is constant}$$

And the field amplitude grows linearly with distance.

The irradiance (intensity) then grows quadratically with distance.