Lecture 15

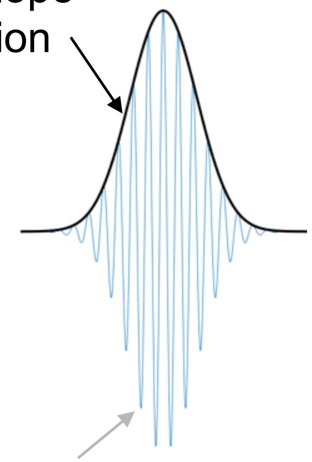
Frequency conversion using ultrashort optical pulses.

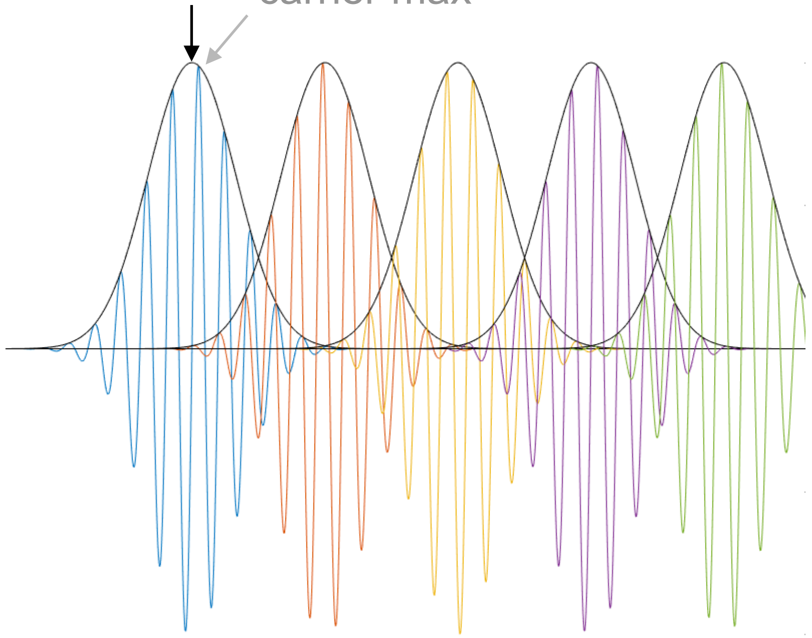
Problem solving practices

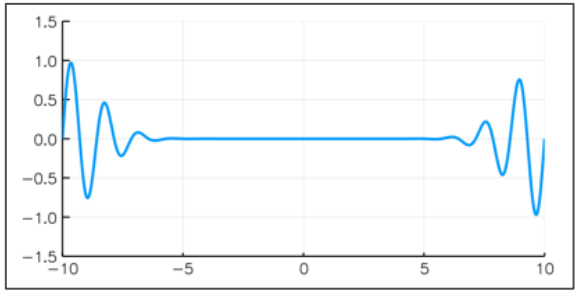
Group vs phase velocity

If you do not neglect dispersion:

The pulse propagation velocity (also known as the **group** velocity $v_g = c/n_g$) differs from the propagation velocity of the carrier (also known as the **phase** velocity $v_p = c/n$). Energy is transported through the medium at <u>group velocity</u>.







$$E(t,z) = Re\,\{\, a(z,t)\; e^{i(\omega_0 t - k_0 z)} \,\}$$

Let us show that $a(z,t)$ propagates with the **group velocity**

Time-domain formulation of nonlinear optics

In the time-domain formulation, we express the field in terms of slowly varying **envelope** multiplied by a **carrier**:

$$E(t,z) = Re\,\{\,a(z,t)\,e^{i(\omega_0 t - k_0 z)}\,\} \qquad (15.1)$$

envelope — carrier

The Fourier transform of the **envelope function** is centered near ZERO frequency

At z=0

$$E(t) = a(t)\,e^{i\omega_0 t}$$

$$A_{\tilde\omega} = \int_{-\infty}^{\infty} a(t)\,e^{-i\tilde\omega t}\,dt$$

The Fourier transform of the envelope function

$$a(t) = \int_{-\Delta\omega}^{\Delta\omega} A_{\tilde\omega}\,e^{i\tilde\omega t}\,\frac{d\tilde\omega}{2\pi}$$

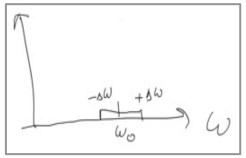
Inverse Fourier transform

$$E(t) = e^{i\omega_0 t}a(t) = e^{i\omega_0 t}\int_{-\Delta\omega}^{\Delta\omega} A_{\tilde\omega}\,e^{i\tilde\omega t}\,\frac{d\tilde\omega}{2\pi} = \int_{-\Delta\omega}^{\Delta\omega} A_{\tilde\omega}\,e^{i(\omega_0+\tilde\omega)t}\,\frac{d\tilde\omega}{2\pi} = \int_{\omega_0-\Delta\omega}^{\omega_0+\Delta\omega} A_{\tilde\omega}\,e^{i\omega t}\,\frac{d\omega}{2\pi} \qquad (15.2)$$

spectral components around 0

spectral components around ω_0

$$\omega = \omega_0 + \tilde\omega$$
$$\tilde\omega = \omega - \omega_0$$
$$d\omega = d\tilde\omega$$



Time-domain formulation of nonlinear optics

z≠0

Each spectral component propagates with its own phase velocity

$$e^{i\omega t} \rightarrow e^{i\omega t - ikz} \qquad k = k(\omega) \quad \text{-dispersion}$$

$$A_{\tilde{\omega}} \rightarrow A_{\tilde{\omega}}\, e^{-ikz}$$

$$E(t,z) = \int_{\omega_0-\Delta\omega}^{\omega_0+\Delta\omega} A_{\tilde{\omega}}\, e^{i(\omega t - k(\omega)z)} d\omega = \int_{-\Delta\omega}^{\Delta\omega} A_{\tilde{\omega}}\, e^{i(\omega_0+\tilde{\omega})t - i(k_0+\Delta k(\omega))z}\, d\tilde{\omega} =$$

$\tilde{\omega} = \omega - \omega_0$

Taylor expansion: $k = k(\omega) = k_0 + \Delta k(\omega) = k_0 + \frac{dk}{d\omega}\tilde{\omega} = k_0 + \frac{\tilde{\omega}}{v_g};$ $\quad v_g = \frac{d\omega}{dk}$

$\tilde{\omega} = \omega - \omega_0$

$$= e^{i(\omega_0 t - k_0 z)} \int_{-\Delta\omega}^{\Delta\omega} A_{\tilde{\omega}}\, e^{i(\tilde{\omega} t - \Delta k z)}\, d\tilde{\omega} = e^{i(\omega_0 t - k_0 z)} \int_{-\Delta\omega}^{\Delta\omega} A_{\tilde{\omega}}\, e^{i(\tilde{\omega} t - \frac{\tilde{\omega}}{v_g} z)}\, d\tilde{\omega} =$$

$$= e^{i(\omega_0 t - k_0 z)} \underbrace{\int_{-\Delta\omega}^{\Delta\omega} A_{\tilde{\omega}}\, e^{i\tilde{\omega}(t-\frac{z}{v_g})}\, d\tilde{\omega}}_{a(t - z/v_g)} = a\left(t - \frac{z}{v_g}\right) e^{i(\omega_0 t - k_0 z)} \qquad (15.3)$$

envelope (amplitude) — carrier (phase)

$$v_g = \frac{d\omega}{dk} = \left(\frac{dk}{d\omega}\right)^{-1} = \frac{c}{n_g}$$

$$n_g = c\left(\frac{dk}{d\omega}\right) = c\,\frac{d(\omega n/c)}{d\omega} = n + \omega\frac{dn}{d\omega} = n - \lambda\frac{dn}{d\lambda} > n \quad \rightarrow \quad v_g < v_p \quad \text{(typically)}$$

Time-domain formulation of nonlinear optics

from Lecture 2

$\nabla \times$

$$\nabla \cdot \boldsymbol{D} = \boldsymbol{0}$$

$$\nabla \cdot \boldsymbol{B} = 0$$

$$\nabla \times \boldsymbol{E} = -\frac{d\boldsymbol{B}}{dt}$$

$$\nabla \times \boldsymbol{H} = \frac{d\boldsymbol{D}}{dt}$$

Wave equation

$$\nabla^2 \boldsymbol{E} - \mu_0 \frac{d^2 \boldsymbol{D}}{dt^2} = 0$$

$$\frac{\partial^2 E}{\partial z^2} - \mu_0 \frac{d^2 D}{dt^2} = 0$$

for plane wave (scalar eq.)

$$\boldsymbol{D} = \boldsymbol{D}_{(\boldsymbol{1})} + \boldsymbol{D}_{(\boldsymbol{2})} = \varepsilon \boldsymbol{E} \quad + \boldsymbol{P}_{NL}$$

$$\frac{\partial^2 E}{\partial z^2} - \mu_0 \varepsilon \frac{d^2 E}{dt^2} = \mu_0 \frac{\partial^2 P_{\boldsymbol{NL}}}{\partial t^2}$$

same as

$$\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{\boldsymbol{NL}}}{\partial t^2}$$

wave equation with an external driving force

$\varepsilon = \varepsilon_0\, n^2$

$c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$

- speed of light in vacuum

Frequency-domain form - looks much simpler

Monochromatic wave:

$\frac{d^2}{dt^2} \to -\omega^2$

$$\frac{\partial^2 E_\omega}{\partial z^2} + \left(\frac{n\omega}{c}\right)^2 E_\omega = \frac{\partial^2 E_\omega}{\partial z^2} + k(\omega)^2 E_\omega = \mu_0 \frac{\partial^2 P_{\boldsymbol{NL,\omega}}}{\partial t^2} \qquad (15.4)$$

Time-domain formulation of nonlinear optics

Our approach: Convert the time-domain field of the form (15.1) to the frequency domain, solve (15.4) and go back to the time domain

$$E(z,\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}E(z,t)\,e^{-i\omega t}d\omega=\frac{1}{2\pi}\int_{-\infty}^{\infty}\{a(z,t)\,e^{i(\omega_0 t-k_0 z)}\}\,e^{-i\omega t}d\omega$$

$$=e^{-ik_0 z}\frac{1}{2\pi}\int_{-\infty}^{\infty}a(z,t)\,e^{-i(\omega-\omega_0)t}d\tilde{\omega}=e^{-ik_0 z}\frac{1}{2\pi}\int_{-\Delta\omega}^{\Delta\omega}a(z,t)e^{-i\tilde{\omega}t}\,d\tilde{\omega}=A(z,\tilde{\omega})e^{-ik_0 z}$$

$\tilde{\omega}=\omega-\omega_0$

.. and now plug into (15.4)

$$\frac{\partial^2 E(z,\omega)}{\partial z^2}+k(\omega)^2 E(z,\omega)=\mu_0\frac{\partial^2 P_{\boldsymbol{NL,\omega}}}{\partial t^2}$$

$$k(\omega)^2\,A(z,\omega-\omega_0)e^{-ik_0 z}$$

$$\frac{\partial^2}{\partial z^2}[A(z,\tilde{\omega})e^{-ik_0 z}]=(\underbrace{A_{zz}}_{\approx 0}-2ik_0A_z-k_0^2A)e^{-ik_0 z}\approx(-2ik_0A_z-k_0^2A)e^{-ik_0 z}$$

$A_z=\frac{\partial A}{\partial z}$ etc.

slowly varying envelope approximation (SVEA) : $\left|\frac{\partial^2 A}{\partial z^2}\right|\ll\left|2k_0\frac{\partial A}{\partial z}\right|\ll\left|k_0^2 A\right|$

so we get $$\left(-2ik_0\frac{\partial A}{\partial z}+(k^2-k_0^2)A\right)e^{-ik_0 z}=\mu_0\frac{\partial^2 P_{\boldsymbol{NL,\omega}}}{\partial t^2}$$

$$(k^2-k_0^2)\approx 2k_0(k-k_0)\approx 2k_0[\frac{dk}{d\omega}(\omega-\omega_0)+\frac{1}{2}\frac{d^2k}{d\omega^2}(\omega-\omega_0)^2+\cdots]$$

$$\left(-2ik_0\frac{\partial A}{\partial z}+2k_0[\frac{dk}{d\omega}(\omega-\omega_0)+\frac{1}{2}\frac{d^2k}{d\omega^2}(\omega-\omega_0)^2+\cdots]A\right)e^{-ik_0 z}=\mu_0\frac{\partial^2 P_{\boldsymbol{NL,\omega}}}{\partial t^2}$$

$A=A(z,\omega-\omega_0)$

Time-domain formulation of nonlinear optics

Fourier Transform of the envelope function

$$\left(\frac{\partial}{\partial z} + i\frac{dk}{d\omega}(\omega-\omega_0) + \frac{1}{2}i\frac{d^2k}{d\omega^2}(\omega-\omega_0)^2 + \cdots\right)A(z,\omega-\omega_0)e^{-ik_0 z} = \frac{i}{2k_0}\mu_0\frac{\partial^2 P_{\boldsymbol{NL,\omega}}}{\partial t^2}$$

(1) (2) (3)

This equation is now Fourier transformed back from the frequency domain – to the time domain:

$$f(t) = \mathcal{F}^{-1}\{\tilde{f}(\omega)\} = \frac{1}{2\pi}\int_{-\infty}^{\infty}\tilde{f}(\omega)\ e^{i\omega t}\,d\omega$$

Recall (2.3) from L2

$$\mathcal{F}^{-1}\{\boldsymbol{i\omega}\tilde{f}(\omega)\} = \frac{d}{dt}f(t)$$

$$\mathcal{F}^{-1}\{-\boldsymbol{\omega^2}\tilde{f}(\omega)\} = \frac{d^2}{dt^2}f(t)$$

(1)
$$\int_{-\infty}^{\infty}\frac{\partial}{\partial z}A(z,\omega-\omega_0)e^{-ik_0 z}\,e^{i\omega t}\frac{d\omega}{2\pi} = e^{-ik_0 z}\frac{\partial}{\partial z}\int_{-\infty}^{\infty}A(z,\omega-\omega_0)\ e^{i\omega t}\frac{d\omega}{2\pi} = e^{i\omega_0 t-ik_0 z}\frac{\partial}{\partial z}\int_{-\infty}^{\infty}A(z,\omega-\omega_0)\ e^{i(\omega-\omega_0)t}\frac{d\omega}{2\pi} =$$

$$= e^{i\omega_0 t-ik_0 z}\frac{\partial}{\partial z}\underbrace{\int_{-\infty}^{\infty}A(z,\tilde{\omega})\ e^{i\tilde{\omega}t}\frac{d\tilde{\omega}}{2\pi}}_{a(z,t)} = e^{i\omega_0 t-ik_0 z}\ \frac{\partial a(z,t)}{\partial z}$$

$\beta_1 = \frac{dk}{d\omega}$

$v_g = \frac{1}{\beta_1} = \frac{d\omega}{dk}$

(2)
$$\rightarrow\ e^{i\omega_0 t-ik_0 z}\left(\frac{dk}{d\omega}\right)\frac{\partial a(z,t)}{\partial t} = e^{i\omega_0 t-ik_0 z}\beta_1\frac{\partial a(z,t)}{\partial t}$$

(3)
$$\rightarrow\ e^{i\omega_0 t-ik_0 z}\left(-\frac{1}{2}i\right)\left(\frac{d^2k}{d\omega^2}\right)\frac{d^2a(z,t)}{dt^2} = e^{i\omega_0 t-ik_0 z}\left(-\frac{i}{2}\beta_2\right)\frac{d^2a(z,t)}{dt^2}$$

$\beta_2 = \frac{d^2k}{d\omega^2} = \frac{d}{d\omega}\left(\frac{1}{v_g}\right)$ group velocity dispersion (GVD)

Time-domain formulation of nonlinear optics

Finally we get:

$$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} - \frac{i}{2}\beta_2 \frac{\partial^2 a(z,t)}{\partial t^2} = e^{-i\omega_0 t + ik_0 z}\, \mathcal{F}^{-1}\left\{\frac{i}{2k_0}\mu_0 \frac{\partial^2 P_{\boldsymbol{NL},\boldsymbol{\omega}}}{\partial t^2}\right\} = e^{-i(\omega_0 t + k_0 z)} \frac{i}{2k_0}\mu_0 \frac{\partial^2 P_{\boldsymbol{NL}}}{\partial t^2} \qquad (15.5)$$

In the time-domain formulation, the nonlinear polarization is also expressed in terms of a slowly varying envelope multiplied by a carrier:

$$\boxed{P_{\boldsymbol{NL}}(t,z) = Re\left\{p_{NL}(z,t)e^{i(\omega_0 t - k_{NL} z)}\right\}} \qquad (15.6)$$

$$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} - \frac{i}{2}\beta_2 \frac{\partial^2 a(z,t)}{\partial t^2} = \frac{i}{2k_0}\mu_0(-\omega_0^2)p_{NL}(z,t)e^{-i\Delta k z}$$

$$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} - \frac{i}{2}\beta_2 \frac{\partial^2 a(z,t)}{\partial t^2} = -i\frac{\mu_0\omega_0 c}{2n} p_{NL}(z,t)e^{-i\Delta k z} \qquad (15.7)$$

$\Delta k = k_{NL} - k_0$

$k_0 = \frac{\omega_0 n}{c}$

but what are these terms?

this looks very similar to the eq. (2.11) of L2 (monochr. waves)

$$\boxed{\frac{\partial E(z)}{\partial z} = -i\frac{\mu_0 \omega c}{2n} P_{NL}}$$

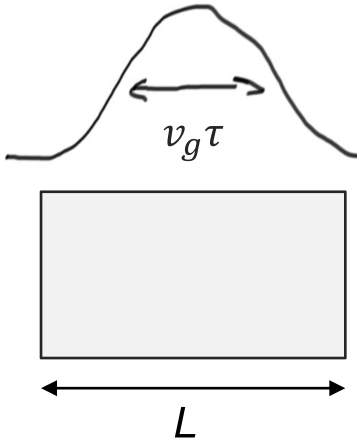
Time-domain formulation of NLO

Let us leave only $\frac{\partial}{\partial t}$ and ignore $\frac{\partial^2}{\partial t^2}$ term for now. What is the difference in adding the time derivative?

$$\frac{\partial a(z,t)}{\partial z} + \frac{1}{v_g}\frac{\partial a(z,t)}{\partial t} = -i\frac{\mu_0\omega_0 c}{2n} p_{NL}(z,t)$$

this looks very similar to the eq. (2.11) of L2 (monochr. waves)

$$\frac{\partial E(z)}{\partial z} = -i\frac{\mu_0\omega c}{2n} P_{NL}$$



$$\frac{\partial a}{\partial t} \sim \frac{a}{\tau}$$

$$\frac{\partial a}{\partial z} \sim \frac{a}{L}$$

When $\frac{1}{v_g}\frac{\partial a(z,t)}{\partial t}$ becomes comparable to $\frac{\partial a(z,t)}{\partial z}$?

when $\frac{a}{v_g\tau} \sim \frac{a}{L}$

-> pulse spread $v_g\tau$ ~ crystal length *L* (or less)

<u>Example</u>: *L*=1 cm, $v_g = \frac{c}{n_g}$; $n_g = 2$

$v_g\tau \approx \mathrm{L}$ -> $\tau = {}^{\mathrm{L}}/_{v_g} \approx 67ps$

For input pulses sufficiently long (>1ns), the time derivative may be neglected

Free pulse propagation

Once again:

$$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} - \frac{i}{2}\beta_2 \frac{\partial^2 a(z,t)}{\partial t^2} = -i\frac{\mu_0 \omega_0 c}{2n} p_{NL}(z,t) e^{-i\Delta k z} \quad (15.7a)$$

term for pulse broadening

nonlinear polarization

Let us ignore the 2nd order dispersion and assume there is no nonlinear polarization

$$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} = 0$$

Free pulse propagation: →

$$\boxed{\frac{\partial a(z,t)}{\partial z} + \frac{1}{v_g}\frac{\partial a(z,t)}{\partial t} = 0} \quad (15.8)$$

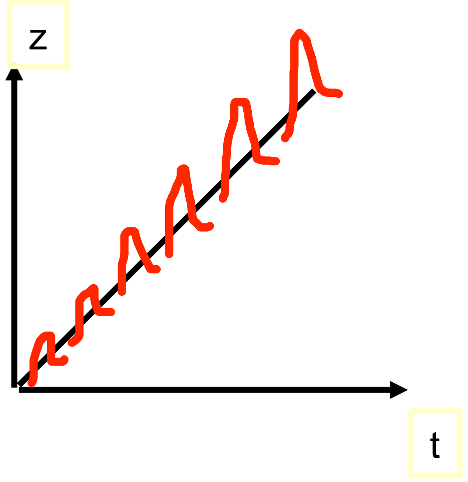
Moving frame: new coordinates:

$$z' = z; \qquad t' = t - \frac{z}{v_g}$$

$$\frac{\partial a}{\partial z} = \frac{\partial a}{\partial t'}\frac{\partial t'}{\partial z} + \frac{\partial a}{\partial z'}\frac{\partial z'}{\partial z} = \frac{\partial a}{\partial t'}\left(-\frac{1}{v_g}\right) + \frac{\partial a}{\partial z'}$$

$$\frac{\partial a}{\partial t} = \frac{\partial a}{\partial t'}\frac{\partial t'}{\partial t} + \frac{\partial a}{\partial z'}\frac{\partial z'}{\partial t} = \frac{\partial a}{\partial t'}$$

chain rule of differentiation



plug into (15.8)

$$-\frac{1}{v_g}\frac{\partial a}{\partial t'} + \frac{\partial a}{\partial z'} + \frac{1}{v_g}\frac{\partial a}{\partial t'} = 0$$

In the moving frame $\frac{\partial a(z',t')}{\partial z'} = 0$ (15.9)

In the absence of nonlinear polarization and high-order dispersion, the electric field envelope a(z, t) would propagate at the group velocity without any distortion or change.

Three-wave interaction with ultrashort pulses

Now ignore the high-order dispersion $\frac{\partial^2}{\partial t^2}$ term and leave the nonlinear polarization driving term

(from 15.7) $$\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} - \frac{i}{2}\beta_2 \frac{\partial^2 a(z,t)}{\partial t^2} = -i\frac{\mu_0 \omega_0 c}{2n} p_{NL}(z,t) e^{-i\Delta k z}$$

$$\boxed{\frac{\partial a(z,t)}{\partial z} + \beta_1 \frac{\partial a(z,t)}{\partial t} = -i\frac{\mu_0 \omega_0 c}{2n} p_{NL}(z,t) e^{-i\Delta k z}} \quad (15.10)$$

For example, in the difference frequency generation case, $p_{NL}(z,t)$ is created as a product of two waves (E_3 and E_2) propagating at (group) velocities $v_{g3}$and v_{g2} so the nonlinear polarization envelope propagates as $a_3(t - \frac{z}{v_{g3}})\ a_2(t - \frac{z}{v_{g2}})$

see A. Weiner "Ultrafast Optics" Wiley (2009), p. 240

Difference frequency generation with ultrashort pulses and group velocity walk-off

The input waves at ω_3 and ω_2 and the difference frequency field (ω_1) have different group velocities in the crystal. Assume nondepleted pump approximation – the field at ω_1 remains weak compared to the input fields. With the assumption that Δk = 0, the equation (15.10) becomes:

$$\frac{\partial a_1}{\partial z} + \frac{1}{v_{g1}}\frac{\partial a_1}{\partial t} = -i\kappa_1 a_3\left(t - \frac{z}{v_{g3}}\right) a_2^*\left(t - \frac{z}{v_{g2}}\right) \qquad (15.11)$$

$$\kappa_1 = \frac{\omega_1 d}{n_1 c}$$

- NL coupling coeff. (d -nonlinear coeff)

New coordinates: $z' = z;\ \ t' = t - \frac{z}{v_{g1}}$ – ride with the wave a_1

This eliinates the 2nd term on the left side and simplifies (15.11) to:

$$\frac{\partial a_1(z,t')}{\partial z} = -i\kappa_1 a_3(t' - \eta_{31} z)\ a_2^*(t' - \eta_{21} z) \qquad (15.12)$$

$t' = 0 \rightarrow$ ride at peak of the intensity

where we have introduced: $\eta_{21} = \frac{1}{v_{g2}} - \frac{1}{v_{g1}} = \frac{1}{c}(n_{g2}\text{-}n_{g1})$ and $\eta_{31} = \frac{1}{v_{g3}} - \frac{1}{v_{g1}} = \frac{1}{c}(n_{g3}\text{-}n_{g1})$

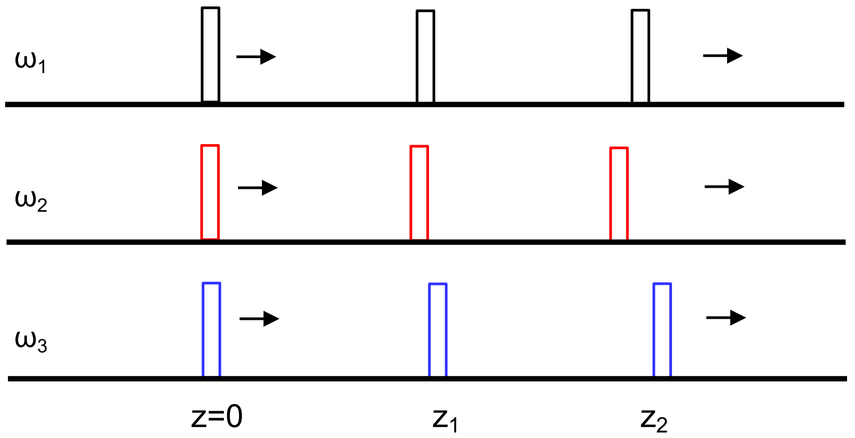
Group velocity walk-off – becomes significant for ps-fs pulses

see A. Weiner "Ultrafast Optics" Wiley (2009), p. 240

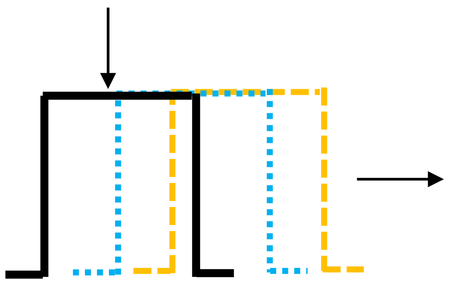
Difference frequency generation (DFG) with ultrashort pulses

The solution for the DFG wave (a_3, a_2 -constant) is:

$$a_1(L,t') = -i\kappa_1 \int_0^L a_3(t' - \eta_{31} z)\, a_2^*(t' - \eta_{21} z)\; dz \qquad (15.13)$$



So when either $\eta_{31} z$ or $\eta_{21} z$ reaches pulse duration ($\boldsymbol{\tau}$), the intercation between 3 waves stops.



Hence the maximum interaction length is

$$L_{max} \approx \frac{c\tau}{\Delta n_g}, \qquad (15.14)$$

where Δn_g =max $\{n_{g2}$-n_{g1}, n_{g3}-$n_{g1}\}$

see A. Weiner “Ultrafast Optics” Wiley (2009), p. 240

Difference frequency generation with ultrashort pulses

Numerical example: DFG in GaSe crystal with fs pulses

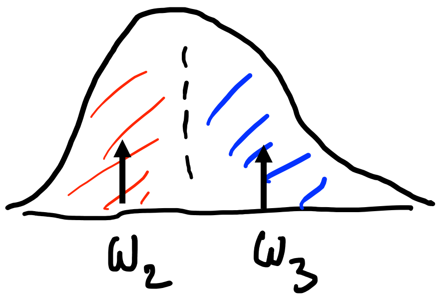
DFG: $\boldsymbol{\omega_1 = \omega_3 - \omega_2}$

Assume a rectangular pulse with the pulsewidth τ=20 fs

The interaction length L_{eff} corresponds to:
The lag time = $\eta_{21} L_{eff}$ equal to the pulse duration, that is

lag time: $\frac{1}{c}\Delta n_g L_{eff} = \tau \ \rightarrow \quad \boldsymbol{L_{eff} = \frac{c\tau}{\Delta n_g}}$

Laser spectrum

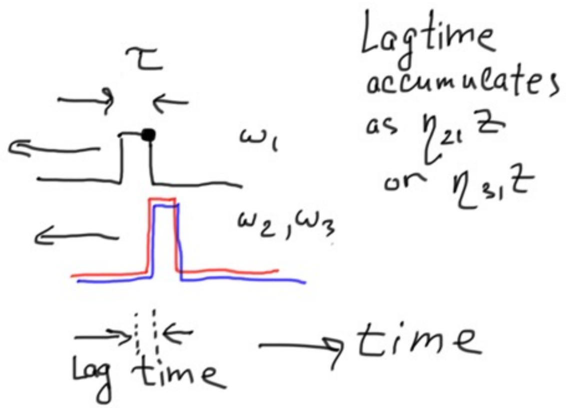


Scenario #1

λ_1 =10 μm
λ_2 = 0.83 μm
λ_3 = 0.77 μm
n_{g1} =2.761
n_{g2} =3.098
n_{g3} =3.171
$\rightarrow \Delta n_g$ =0.41

Ti: Sapphire laser

Scenario #1
L_{eff} = 15 μm



Scenario #2

λ_1 =10 μm
λ_2 = 2.66 μm
λ_3 = 2.1 μm
n_{g1} =2.78
n_{g2} =2.76
n_{g3} =2.78
$\rightarrow \Delta n_g$ =0.02

Cr: ZnS laser

Scenario #2
L_{eff} = 300 μm

DFG conversion efficiency (at the same focussing) scales as L_{eff}^2
→
400 times difference between the two scenarios

Time-domain formulation of NLO

As we discusses in L13, matching group velocities in **time domain** is the same as matching phase-matching bandwidths in **frequency domain**