Lasers

lecture 13

Czesław Radzewicz

nonlinear optics – a difficult start

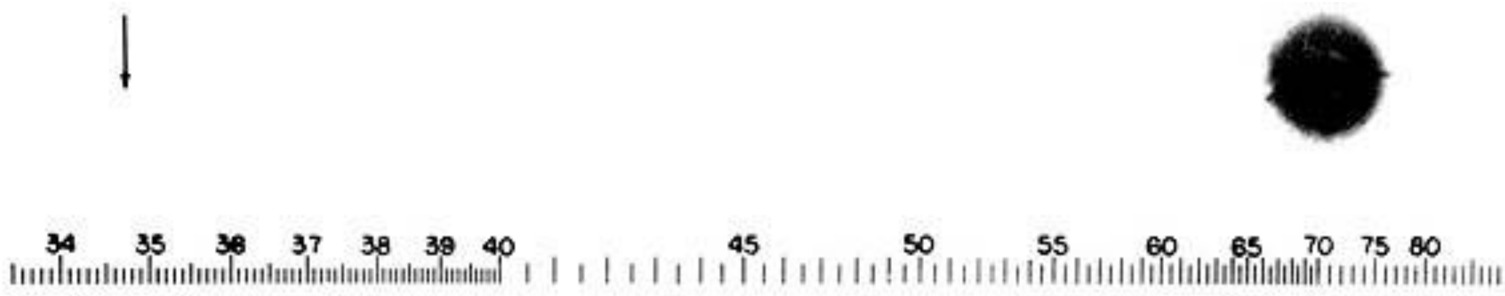


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

‘ruby optical maser’, 3 J, 1 ms
crystalline quartz
‘unambiguous indication of the second harmonic’

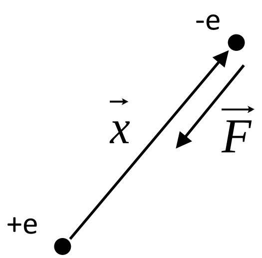
P. A. Franken et al., Phys. Rev. Lett. 7, 118 (1961)

optical nonlinearities

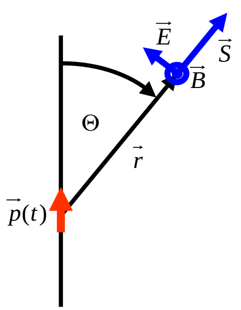
In most cases we assume that the response of a medium to an external em field is linear $\vec{P} = \alpha\varepsilon_0\vec{E}$

examples:

- Lorentz model for refractive index in gases
- Drude model for refractive index in metals

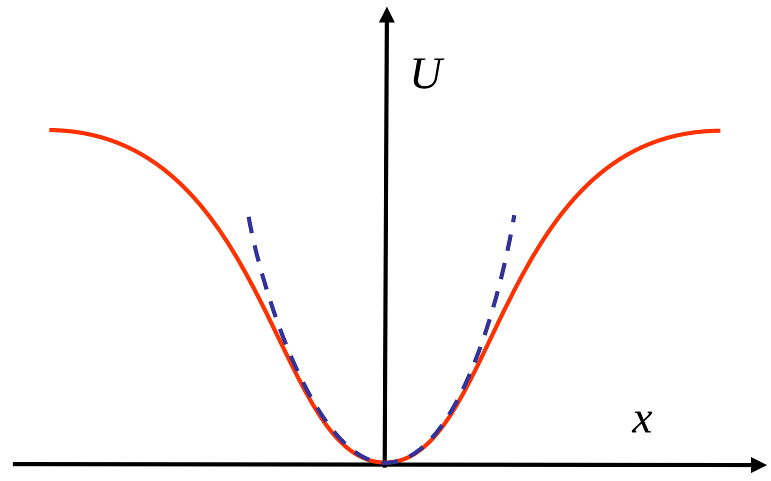


$$F = -kx = -m{\omega_0}^2 x$$
$$F = -\nabla U \rightarrow U = \frac{1}{2}kx^2$$



as long as the motion of the electron is small we can assume: $p(t) = \alpha\varepsilon_0 E(t)$

small amplitude motion breaks up with strong em field such as lasers beams



for large amplitude of electron motion we have

$$p(t) = \alpha\varepsilon_0 E(t) + \beta\varepsilon_0 E^2(t) + \cdots$$

this results in nonlinear polarization of the medium

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

the electric dipole of an atom/molecule oscillates with frequencies different than the driving field frequency

when should we account for nonlinear medium response?

the nonlinear polarization is important when

$$\frac{P^{(2)}}{P^{(1)}} \approx \frac{E_0}{E_a}$$

field amplitude

Coulomb field amplitude

$$I = \frac{c\epsilon_0}{2} {E_0}^2 \Longrightarrow E_0 = \sqrt{\frac{2I}{c\epsilon_0}} \cong 27.4\sqrt{I}$$

$$E_0 \left[\frac{\mathrm{V}}{\mathrm{m}}\right], \qquad I\left[\frac{\mathrm{W}}{\mathrm{m}^2}\right]$$

example: less than average fs CPA Ti:Sap amplifier: 1 mJ, 100 fs, focused down to 10 μm

$$I = \frac{1 \times 10^{-3}}{100 \times 10^{-15}(10^{-5})^2} = 10^{20}\mathrm{W/m^2}$$

$$E_0 \cong 2.7 \times 10^{11}\mathrm{V/m}$$

hydrogen atom

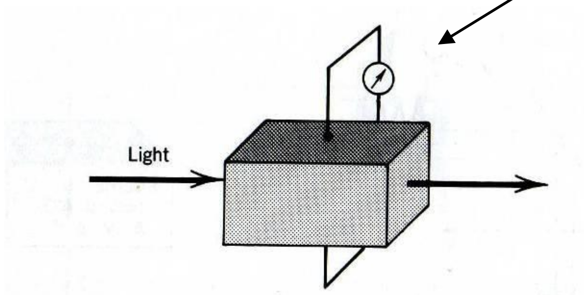
$$E_a \cong 5 \times 10^{11}\mathrm{V/m}$$

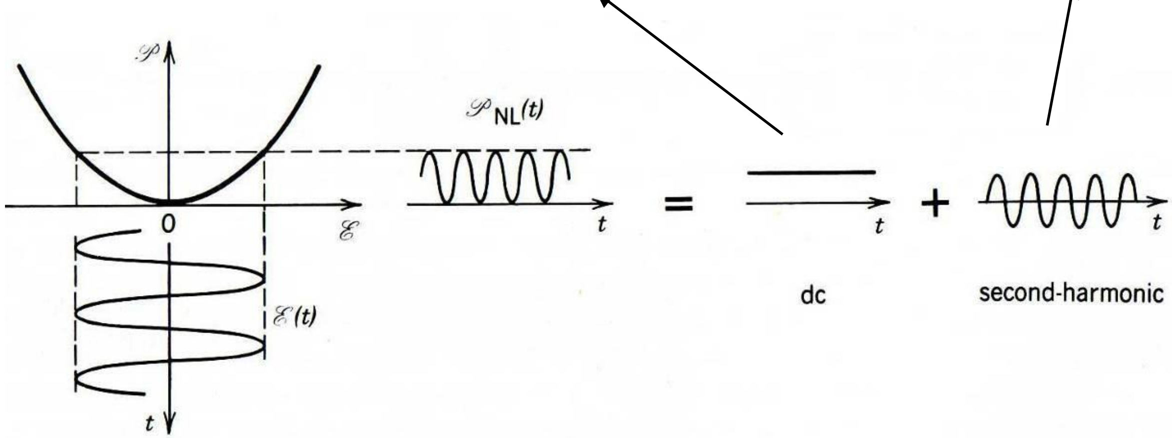
effects of nonlinearity, 1

example 1: a single monochromatic wave $E(t) = E_0 \cos(\omega t)$
+ 2-order nonlinearity $\chi^{(2)}$

$$P^{NL}(t) = P^{(2)}(t) = \chi^{(2)} {E_0}^2 \cos^2(\omega t) = \frac{1}{2} \chi^{(2)} {E_0}^2 [1 + \cos(2\omega t)]$$

optical rectification
second harmonic generation (SHG)





effects of nonlinearity, 2

example2: two monochromatic waves: $E(t) = E_1 \cos(\omega_1 t) + E_2 \cos(\omega_2 t)$
+ 2-order nonlinearity $\chi^{(2)}$:

$$
\begin{aligned}
P^{NL}(t) = P^{(2)}(t) = \chi^{(2)} E^2(t) = \chi^{(2)} [\mathrm{E_1}^2 \cos^2(\omega_1 t) + 2E_1 E_2 \cos(\omega_1 t)\cos(\omega_2 t) + {E_2}^2 \cos^2(\omega_2 t)] \\
= \frac{1}{2}\chi^{(2)} {E_1}^2 [1 + \cos(2\omega_1 t)] + \\
+ \frac{1}{2}\chi^{(2)} {E_2}^2 [1 + \cos(2\omega_2 t)] + \\
+ \chi^{(2)} E_1 E_2 \cos[(\omega_1 + \omega_2)t] + \chi^{(2)} E_1 E_2 \cos[(\omega_1 - \omega_2)t]
\end{aligned}
$$

optical rectification	$\omega_1, \omega_2 \rightarrow DC$
SHG	$\omega_1 \rightarrow 2\omega_1$
SHG	$\omega_2 \rightarrow 2\omega_2$
sum frequency generation (SFG)	$\omega_1 + \omega_2$
difference frequency generation (DFG)	$\omega_1 - \omega_2$

3 wave mixing (2 input + 1 output)

effects of nonlinearity, 3

example 3: a single monochromatic wave $E(t) = E_0 \cos(\omega t)$
+ 3-order nonlinearity $\chi^{(3)}$

$$P^{NL}(t) = P^{(3)}(t) = \chi^{(3)} {E_0}^3 \cos^3(\omega t) = \frac{1}{4} \chi^{(3)} {E_0}^3 [\cos(3\omega t) + 3\cos(\omega t)]$$

third harmonics

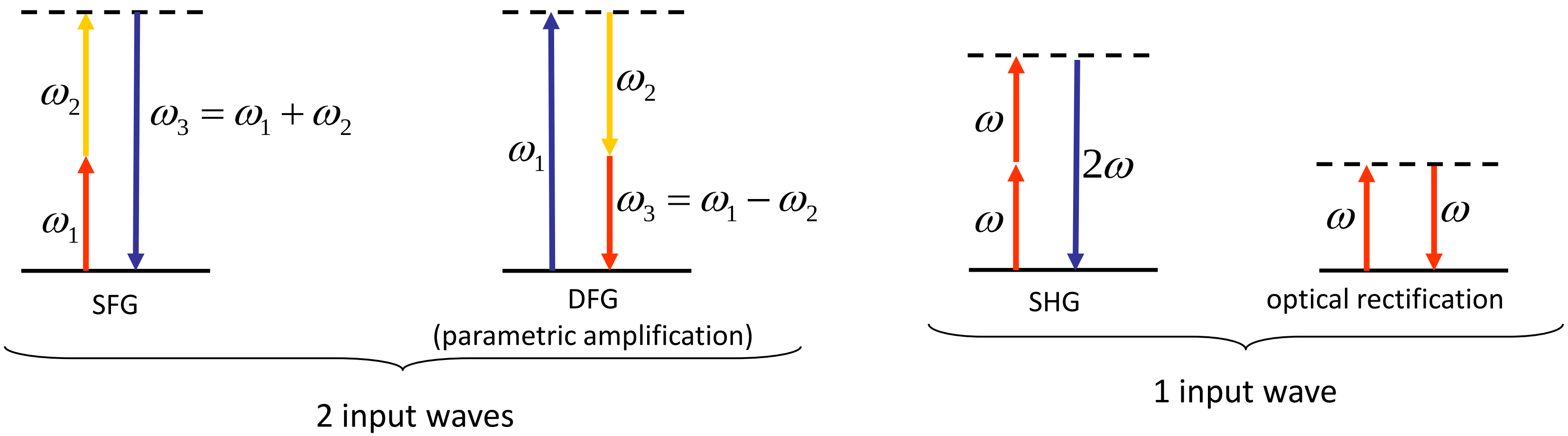
extra polarization at frequency ω
nonlinear index of refraction,
optical Kerr effect

In a general case od the third order nonlinearity we have 3 different input fields
$P^{NL}(t) = P^{(3)}(t) = \chi^{(3)}[E_1(t) + E_2(t) + E_3(t)]^3 = \cdots$ (27 terms) + different polarizations …

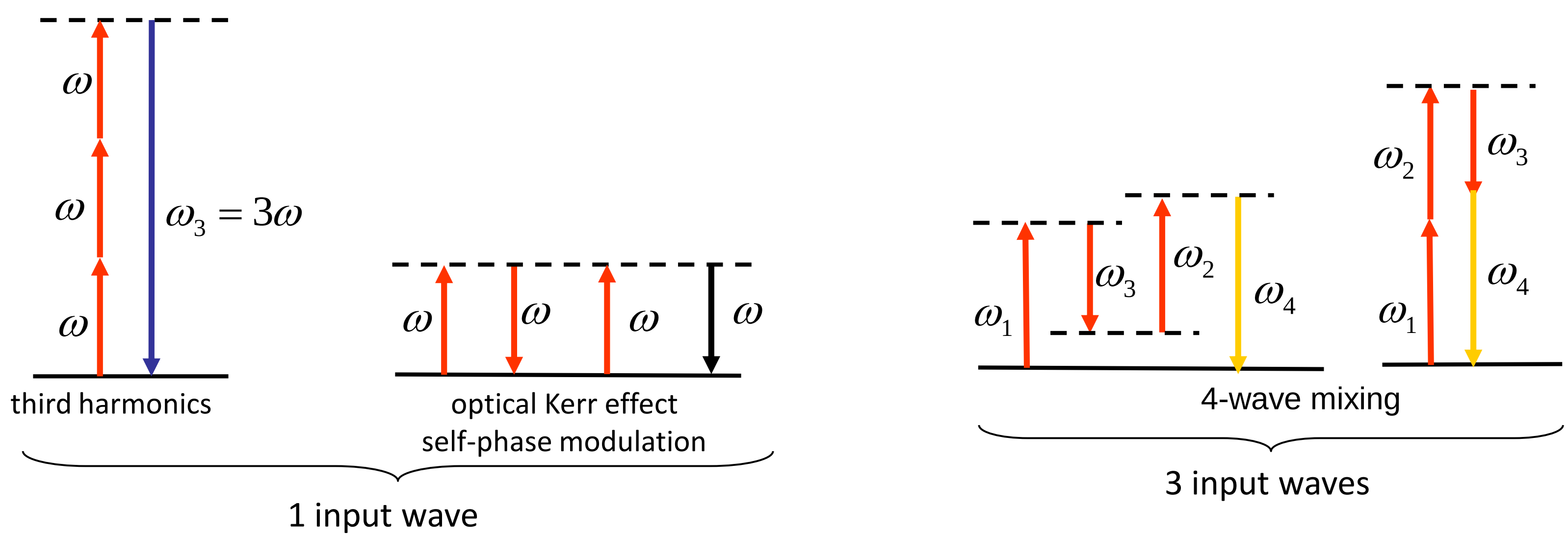
4 wave mixing (3 input + 1 output)

effects of nonlinearity, 4

$\chi^{(2)}$ processes (3-wave mixing) in pictograms



$\chi^{(3)}$ processes (4-wave mixing) in pictograms



some algebra and more pictograms

it is convenient to use complex notation

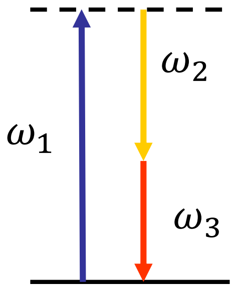
$$E(t) = \mathrm{Re}\left[E(\omega)e^{i\omega t}\right] = \frac{1}{2}\left[E(\omega)e^{i\omega t} + E(-\omega)e^{-i\omega t}\right]$$

with $E(-\omega) = E^*(\omega)$ (this notation will be used throughout this lecture)

for 3-wave mixing processes we have

$$E(t) = \frac{1}{2}\sum_{q=\pm 1,\pm 2} E\left(\omega_q\right)e^{i\omega_q t}$$

$$P^{(2)}(t) = \frac{\chi^{(2)}}{4}\sum_{q,r=\pm 1,\pm 2} E\left(\omega_q\right)E(\omega_r)e^{i(\omega_q+\omega_r)t}$$



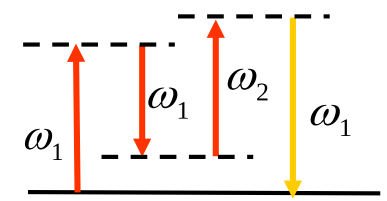
$$\omega_3 = \pm\omega_1 \pm \omega_2$$

$$P^{(2)}(\omega_1 - \omega_2) = \chi^{(2)}E(\omega_1)E^*(\omega_2)$$

4-wave mixing:

$$E(t) = \frac{1}{2}\sum_{q=\pm 1,\pm 2,\pm 3} E\left(\omega_q\right)e^{i\omega_q t}$$

$$P^{(3)}(t) = \frac{\chi^{(2)}}{8}\sum_{q,r,s=\pm 1,\pm 2,\pm 3} E\left(\omega_q\right)E(\omega_r)E(\omega_s)e^{i(\omega_q+\omega_r+\omega_s)t}$$



$$\omega_4 = \pm\omega_1 \pm \omega_2 \pm \omega_3$$

$$P^{(3)}(\omega_1 + \omega_2 - \omega_3) = \chi^{(2)}E(\omega_1)E(\omega_2)E^*(\omega_3)$$

symmetries, 1

Both, electrical field and nonlinear polarization are vectors. Thus $\chi^{(2)}$ i $\chi^{(3)}$ are tensors. Let's inspect $\chi^{(2)}$:

$$P_i^{(2)}(\omega_n+\omega_m)=\frac{1}{4}\sum_{jk}\sum_{mn}\chi^{(2)}{}_{ijk}E_j(\omega_n)\,E_k(\omega_m)=\sum_{jk}\sum_{mn}d_{ijk}E_j(\omega_n)\,E_k(\omega_m)$$

with $d_{ijk}=\frac{\chi^{(2)}{}_{ijk}}{4}$

Tensor d is a cube with 3 nodes on each edge (27 numbers) which is hard to draw in 2D. It is a common practice to use a simplified notation based on Kleiman's symmetry ($\chi^{(2)}{}_{ijk}=\chi^{(2)}{}_{ikj}$). Substituting

ij:	11	22	33	23, 32	31, 13	12, 21
m:	1	2	3	4	5	6

leads to tensor d which can be represented as a 2D matrix

$$d_{mn}=\begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16}\\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26}\\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}\end{bmatrix}$$

and polarization $P^{(2)}$ (for $\omega_3=\omega_1+\omega_2$)

$$\begin{bmatrix} P^{(2)}{}_x\\ P^{(2)}{}_y\\ P^{(2)}{}_z\end{bmatrix}=\begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16}\\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26}\\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}\end{bmatrix}\begin{bmatrix} E_x(\omega_1)E_x(\omega_2)\\ E_y(\omega_1)E_y(\omega_2)\\ E_z(\omega_1)E_z(\omega_2)\\ E_y(\omega_1)E_z(\omega_2)+E_z(\omega_1)E_y(\omega_2)\\ E_x(\omega_1)E_z(\omega_2)+E_z(\omega_1)E_x(\omega_2)\\ E_x(\omega_1)E_y(\omega_2)+E_y(\omega_1)E_x(\omega_2)\end{bmatrix}$$

symmetries, 2

explicit form of d tensor depends on the symmetry of the medium.

example 1: a crystal with $3m$ symmetry (e.g. BBO) can be described using 3 numbers

$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

example 2: a medium with inversion symmetry (e.g. liquid, gas, glass)

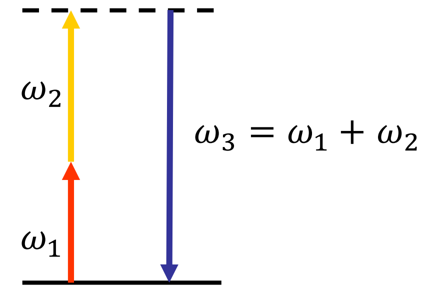
$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix}$$

for $\chi^{(3)}$ (81 numbers) there is no representation as a 2D matrix

wave equation with sources, 1

$$\Delta E - \left(\frac{n}{c}\right)^2 \frac{\partial^2 E}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2 P^{NL}}{\partial t^2} \qquad (1)$$

$$E_n(z,t) = \frac{1}{2}\left[A_n(z)e^{i(\omega_n t - k_n z)} + c.c.\right]$$



$$P^{(2)}(\omega_1+\omega_2) = d^{(2)}E(\omega_1)E(\omega_2)$$
$$P^{(2)}(\omega_3-\omega_2) = d^{(2)}E(\omega_3)E^*(\omega_2)$$
$$P^{(2)}(\omega_3-\omega_1) = d^{(2)}E(\omega_3)E^*(\omega_1)$$

consider the case $\omega_3 = \omega_1 + \omega_2$ and plane wave

$$\Delta E_3 - \left(\frac{n}{c}\right)^2 \frac{\partial^2 E_3}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2 P^{(2)}(\omega_3)}{\partial t^2}$$

$$\Delta E_3 = \frac{\partial^2 E_3}{\partial z^2} = \left(\frac{\partial^2 A_3}{\partial z^2} - 2ik_3\frac{\partial A_3}{\partial z} - {k_3}^2 A_3\right)e^{i(\omega_3 t - k_3 z)} \approx -\left(2ik_3\frac{\partial A_3}{\partial z} + {k_3}^2 A_3\right)e^{i(\omega_3 t - k_3 z)}$$

assume slowly varying envelope and neglect $\frac{\partial^2 A_3}{\partial z^2}$

let's consider $\omega_3 > \omega_1, \omega_2$

$$P^{(2)}(\omega_1+\omega_2) = d^{(2)}A_1(z)A_2(z)e^{i[\omega_3 t - (k_1+k_1)z]}$$

$$\frac{\partial^2 P^{(2)}}{\partial t^2} = -{\omega_3}^2 d^{(2)}A_1(z)A_2(z)e^{i[\omega_3 t - (k_1+k_1)z]}$$

wave equation with sources, 2

put the second spatial derivative of polarization into equation (1)

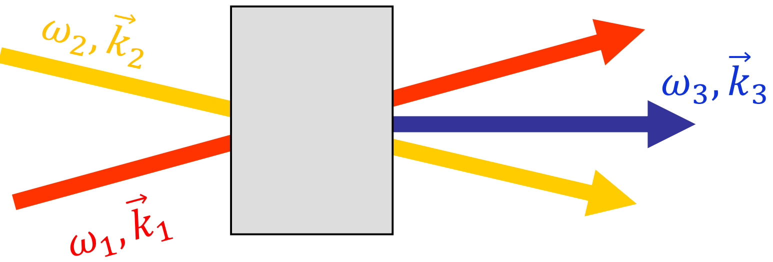
$$-2ik_3\frac{dA_3}{dz}e^{i(\omega_3 t-k_3 z)}=-\frac{4\pi}{c^2}d^{(2)}{\omega_3}^2A_1(z)A_2(z)e^{i[\omega_3 t-(k_1+k_1)z]}$$
$$\frac{dA_3}{dz}=i\frac{2\pi d^{(2)}{\omega_3}^2}{c^2k_3}A_1(z)A_2(z)e^{i(k_1+k_1-k_3)z}=i\frac{2\pi d^{(2)}\omega_3}{n_3c}A_1(z)A_2(z)e^{i\Delta kz}$$

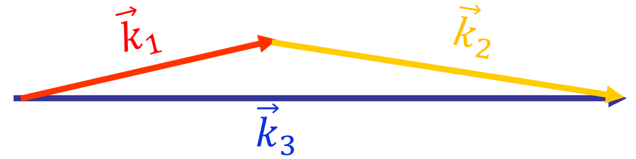
with $\Delta k\equiv-(k_3-k_1-k_2)$ being the phase mismatch

similar reasoning for the two remaining fields leads to:

$$\frac{dA_1}{dz}=i\frac{2\pi d^{(2)}\omega_1}{n_1c}A_1(z){A_2}^*(z)e^{i\Delta kz}$$
$$\frac{dA_2}{dz}=i\frac{2\pi d^{(2)}\omega_2}{n_2c}{A_1}^*(z)A_2(z)e^{i\Delta kz}$$
$$\frac{dA_3}{dz}=i\frac{2\pi d^{(2)}\omega_3}{n_3c}A_1(z)A_2(z)e^{i\Delta kz}$$

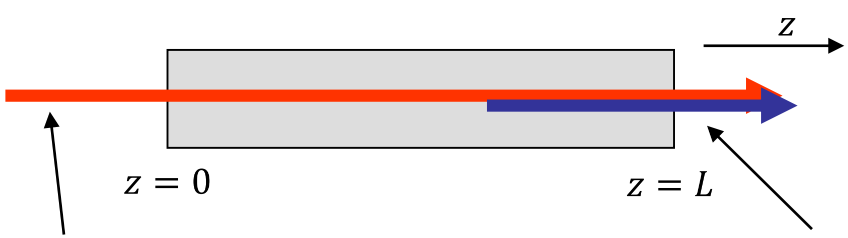
phase matching, 1





perfect phase match $\vec{k}_1 + \vec{k}_2 = \vec{k}_3$ results in high efficiency

an example: SHG



$E_\omega(z,t) = E_0 e^{i(k_\omega z - \omega t)}$

$E_{2\omega}(L,t) = ?$

nonlinear polarization phase

propagation phase

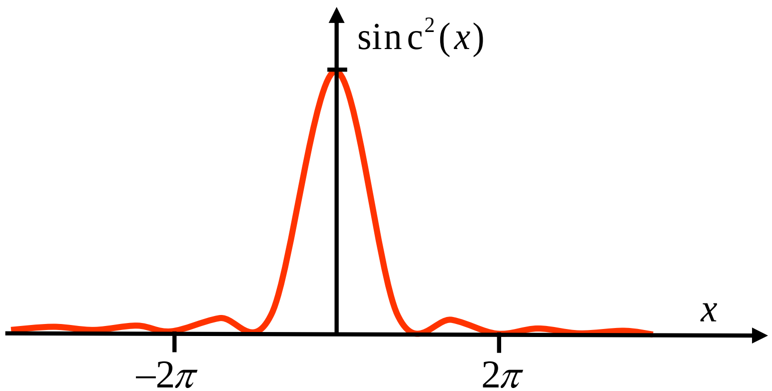
$$E_{2\omega}(L,t) \propto d^{(2)} {E_0}^2 \int_0^L e^{i2(k_\omega z - \omega t)} e^{i[k_{2\omega}(L-z)]} dz$$

$$E_{2\omega}(L,t) \propto d^{(2)} {E_0}^2 e^{i(k_{2\omega} L - 2\omega t)} \int_0^L e^{i(2k_\omega - k_{2\omega})z} dz$$

$$= \frac{d^{(2)} {E_0}^2 e^{i(k_{2\omega} L - 2\omega t)} e^{i\frac{\Delta k L}{2}}}{2i} \operatorname{sinc}\left(\frac{\Delta k L}{2}\right)$$

with phase mismatch $\Delta k = 2k_\omega - k_{2\omega} = \frac{2\omega}{c}[n(\omega) - n(2\omega)]$

phase matching, 2



$$I_{2\omega} \propto \left|d^{(2)}\right|^2 {I_\omega}^2 L^2 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

good efficiency for $|\Delta kL| < \pi$

numbers for SF10 i $\lambda = 1\mu\mathrm{m}$

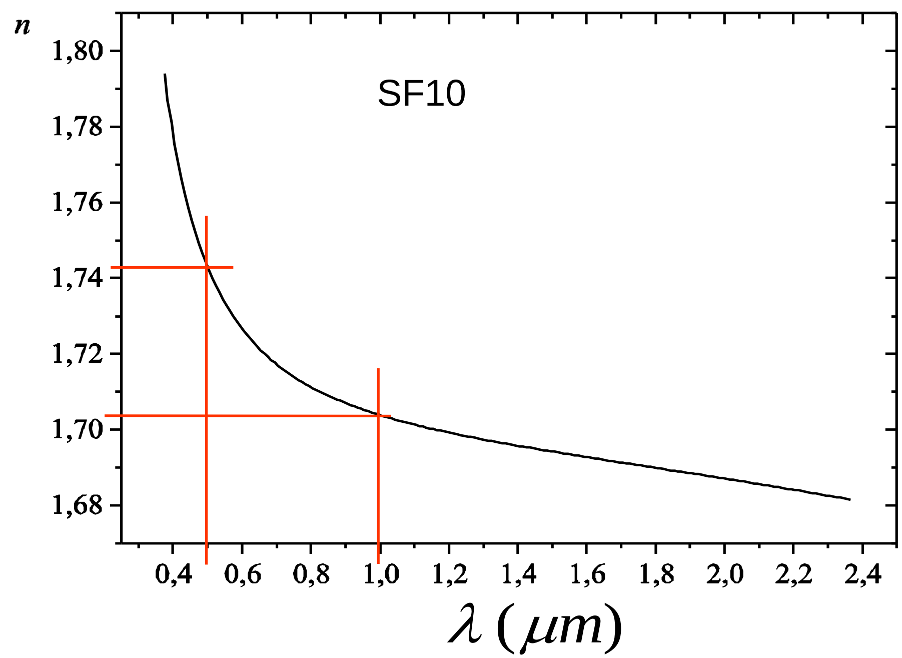
$$\Delta k = \frac{2\omega}{c}[n(\omega) - n(2\omega)]$$

$$= \frac{4\pi}{\lambda}[n(\lambda) - n(\lambda/2)] \cong 0.5\mu\mathrm{m}^{-1}$$

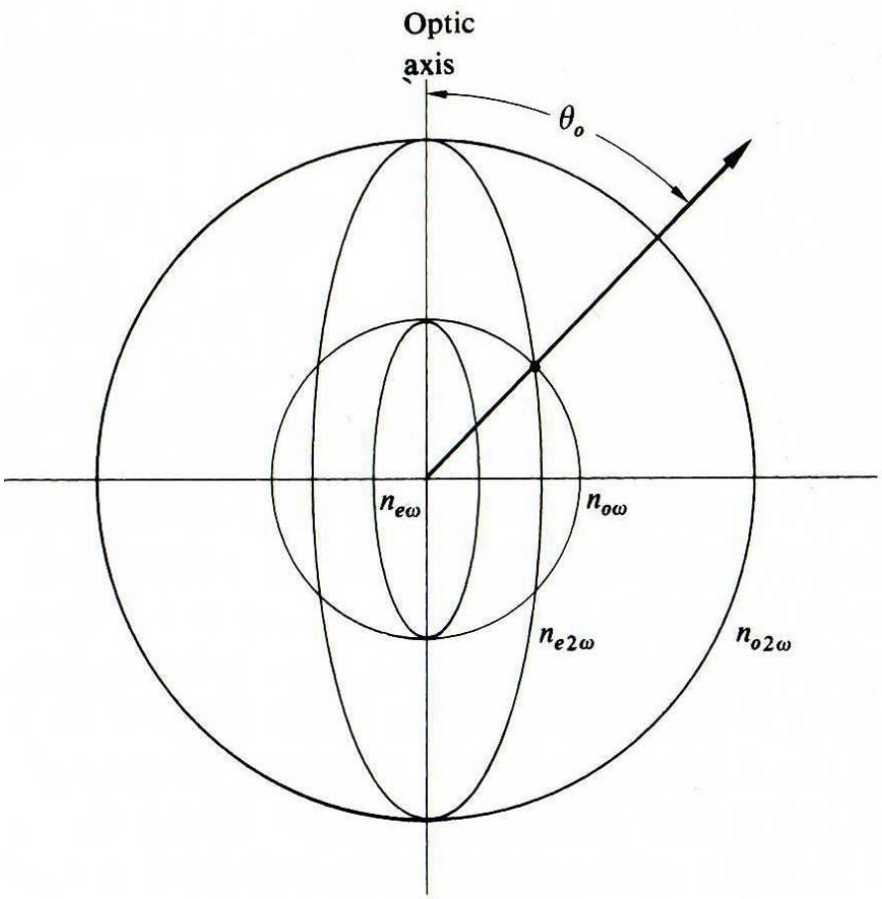
this limits the gain thickness to approx. 12μm

but very thin material = low efficiency

note: SF10 is a glass for which $\chi^{(2)} = 0$.

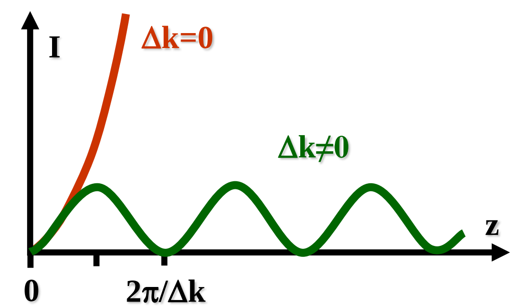


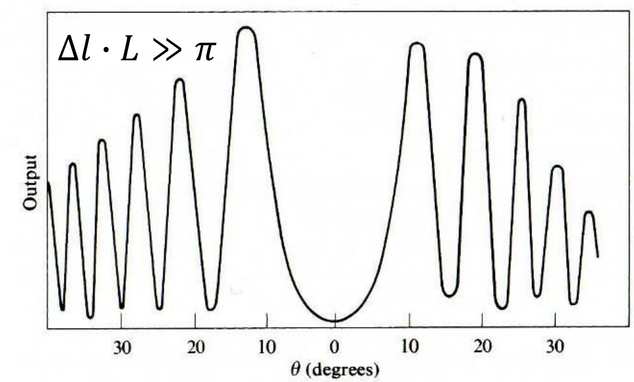
phase matching in birefringent crystals; example: SHG,
uniaxial negative crystal $n_e < n_o$



$$\Delta k = 0 \Leftrightarrow n(\omega) = n(2\omega)$$

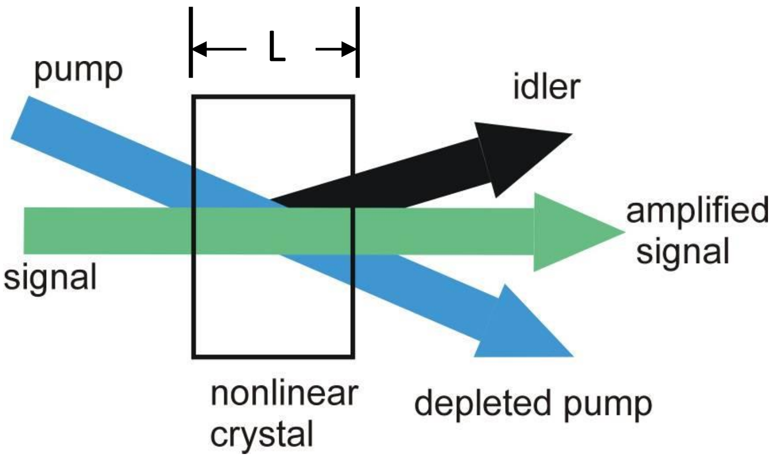
ordinary wave extraordinary wave

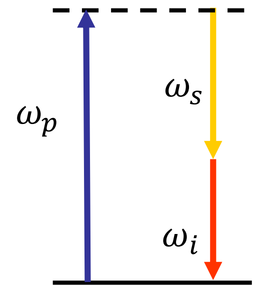




SHG, fused silica, d=0.78 mm

optical parametric amplifier





- strong pump ω_p
- weak signal ω_s
- additional beam is created – idler with frequency

$$\omega_i = \omega_p - \omega_s$$

$$\frac{dA_p}{dz} = i\frac{2\pi d^{(2)}\omega_p}{n_p c}A_s(z)A_i(z)e^{i\Delta kz} \quad (1)$$

$$\frac{dA_i}{dz} = i\frac{2\pi d^{(2)}\omega_i}{n_i c}A_p(z){A_s}^*(z)e^{i\Delta kz} \quad (2)$$

$$\frac{dA_s}{dz} = i\frac{2\pi d^{(2)}\omega_s}{n_s c}{A_i}^*(z)A_p(z)e^{i\Delta kz} \quad (3)$$

these equations must be integrated numerically

for undepleted pump $A_p = \text{const}$

The equations can be solved analytically:

$$G = \frac{I_s(L)}{I_s(0)} = 1 + (\gamma L)^2\left(\frac{\sinh B}{B}\right)^2$$

parametric gain

with

$$B = \sqrt{(\gamma L)^2 - \left(\frac{\Delta k L}{2}\right)^2}$$

$$\gamma = 4\pi d_{eff}\sqrt{I_p \big/ (2\varepsilon_0 n_p n_s n_i c \lambda_s \lambda_i)}$$

without pump depletion

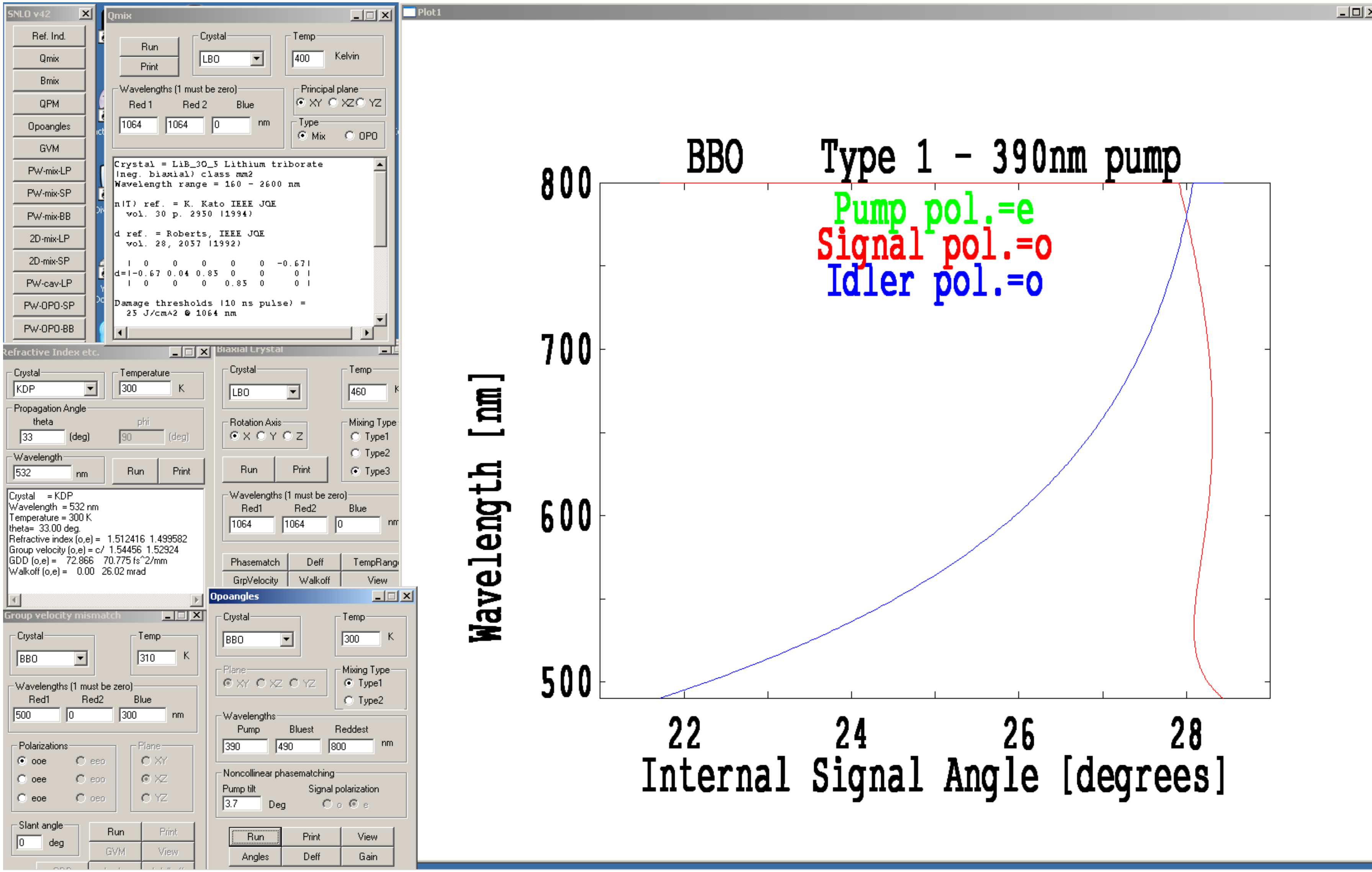
$$I_s^{\,out} = I_s^{\,in} e^{\xi\sqrt{I_p}L}$$

the same as in laser amplifier with

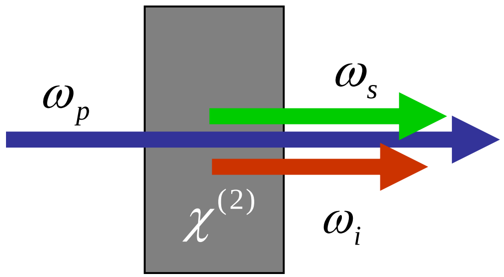
$$\gamma_0 = \xi\sqrt{I_p}$$

numerical codes for nonlinear optics

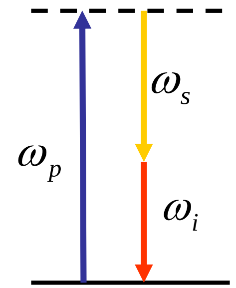
example: SNLO - http://www.as-photonics.com/SNLO/

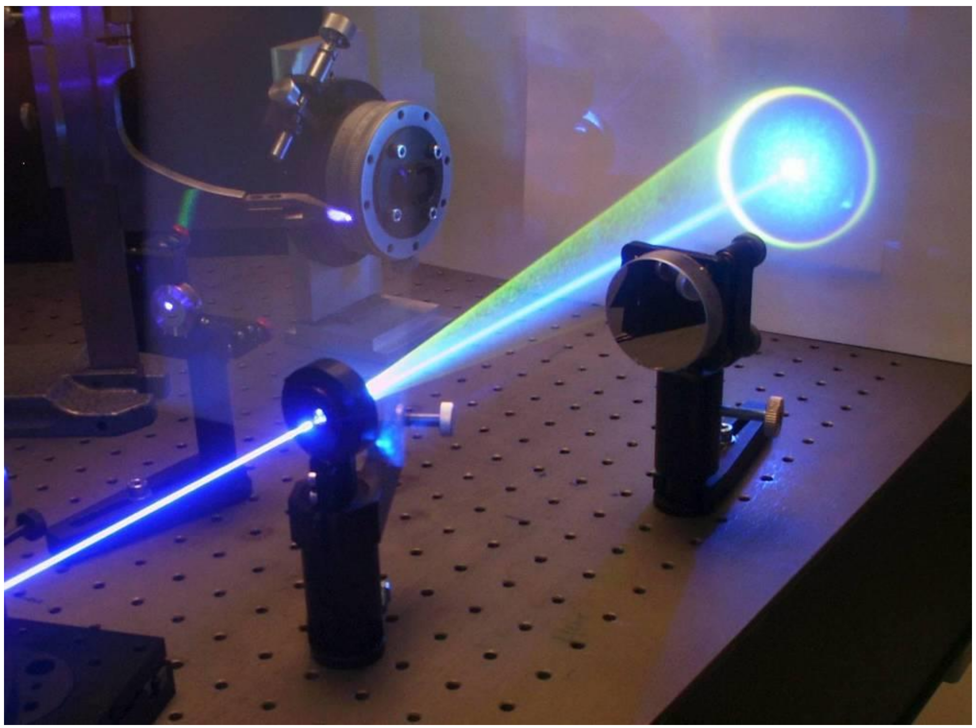


parametric fluorescence – a spontaneous process

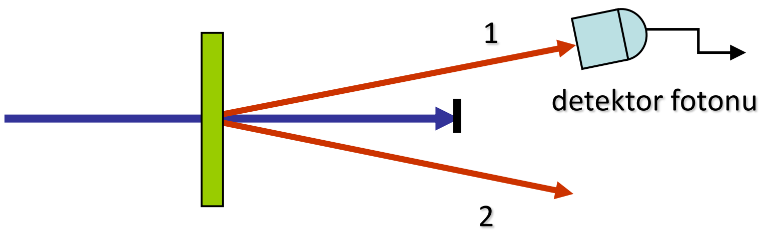


$$\omega_s + \omega_i = \omega_p$$
$$\vec{k}_s + \vec{k}_i = \vec{k}_p$$

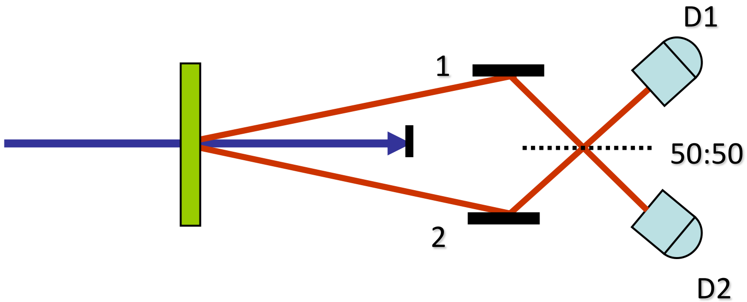




- ❑ single photon generation (heralded photon)

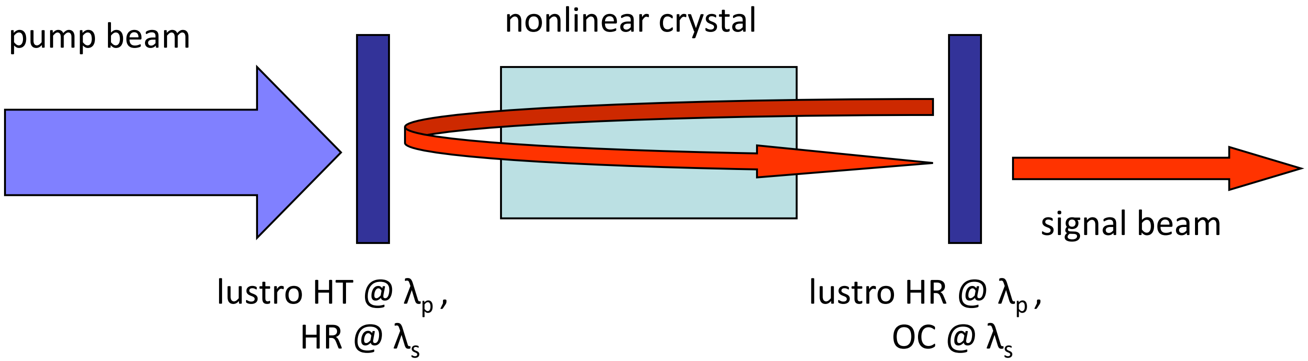


- ❑ photon pair generation



parametric oscillator

singly resonant design

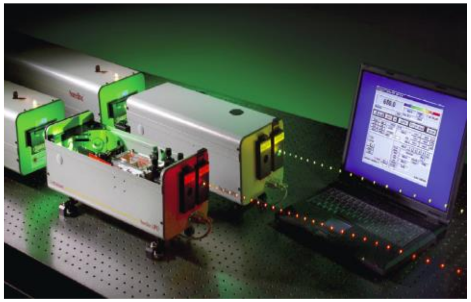


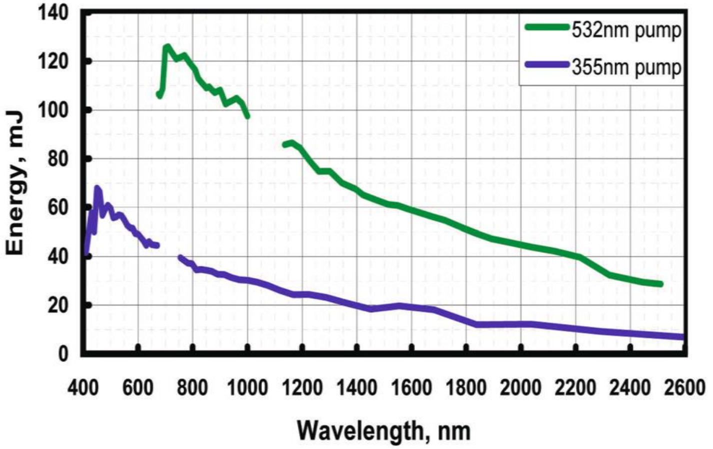
threshold condition: $e^{2\gamma L} = \alpha_s$

with α_s describing total cavity losses per round-trip

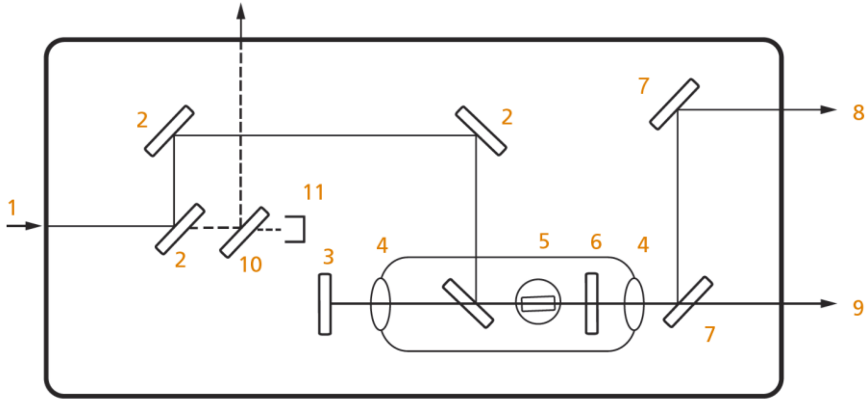
ns commercial OPG, 1

example: Continuum Lasers Surelite OPO – OPG with low spectra resolution 10-400cm^{-1}



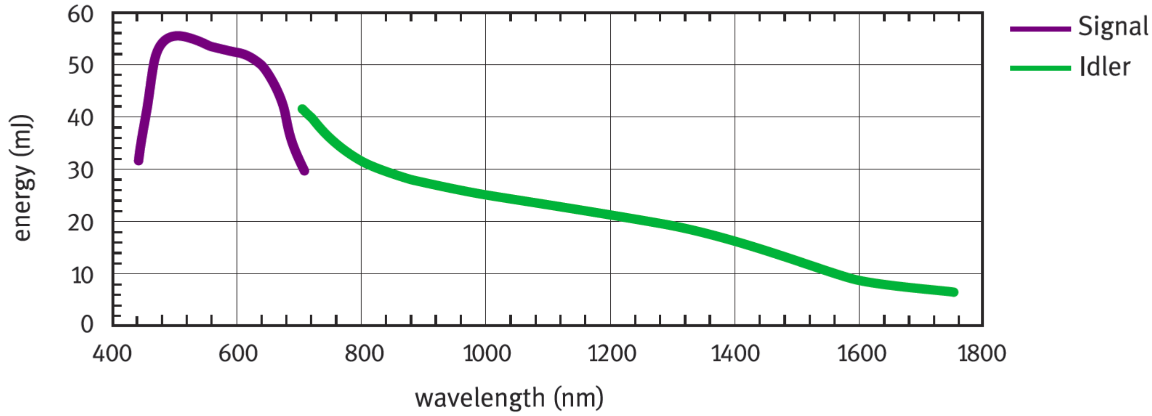


SURELITE™ OPO PLUS OPTICAL LAYOUT

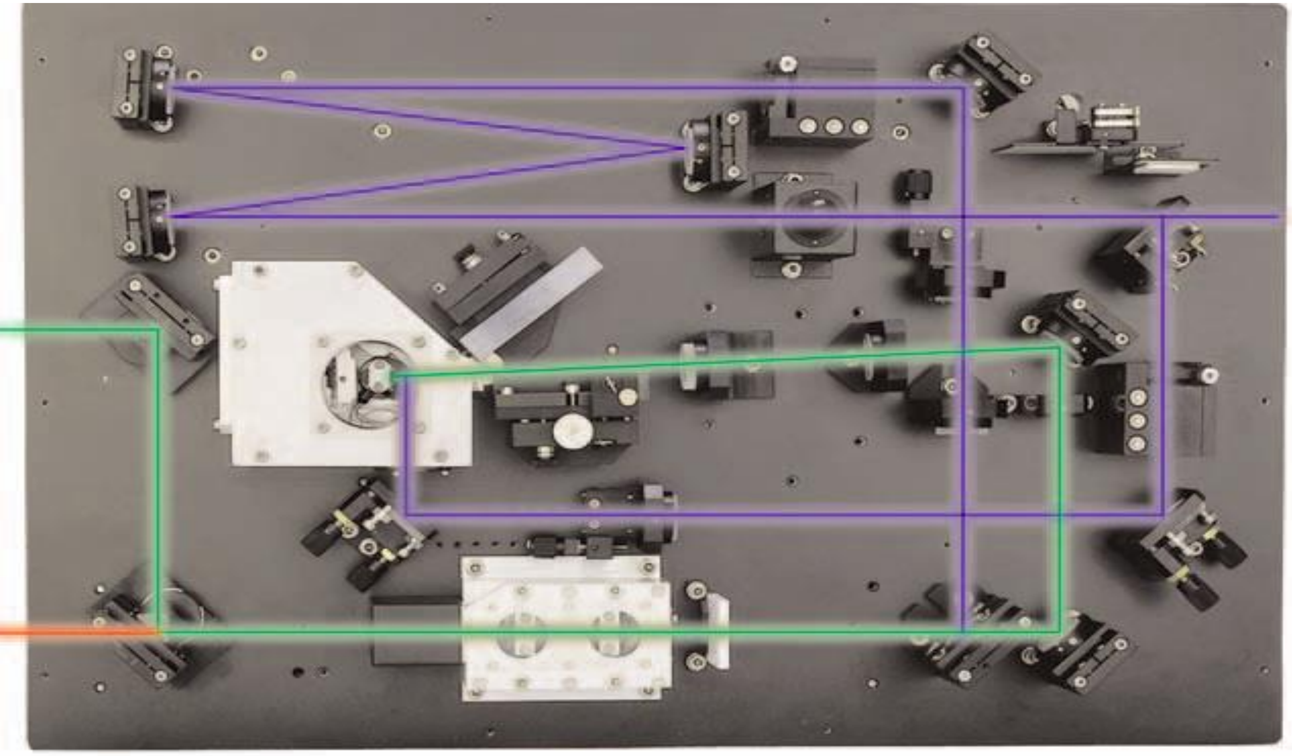


1. pump beam
2. steering mirrors
3. resonator mirror
4. fused silica windows
5. nonlinear crystal
6. output coupler
7. dichroic mirrors
8. signal beam
9. idler beam

ns commercial OPG, 2



example: Continuum lasers Sunlite OPO – OPG with high spectral resolution <0.1cm^{-1}

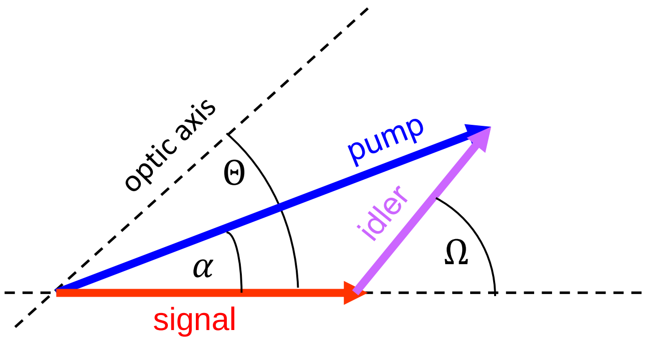


noncollinear optical parametric amplifier (NOPA) for fs pulses

major consideration – group velocity mismatch leads to:

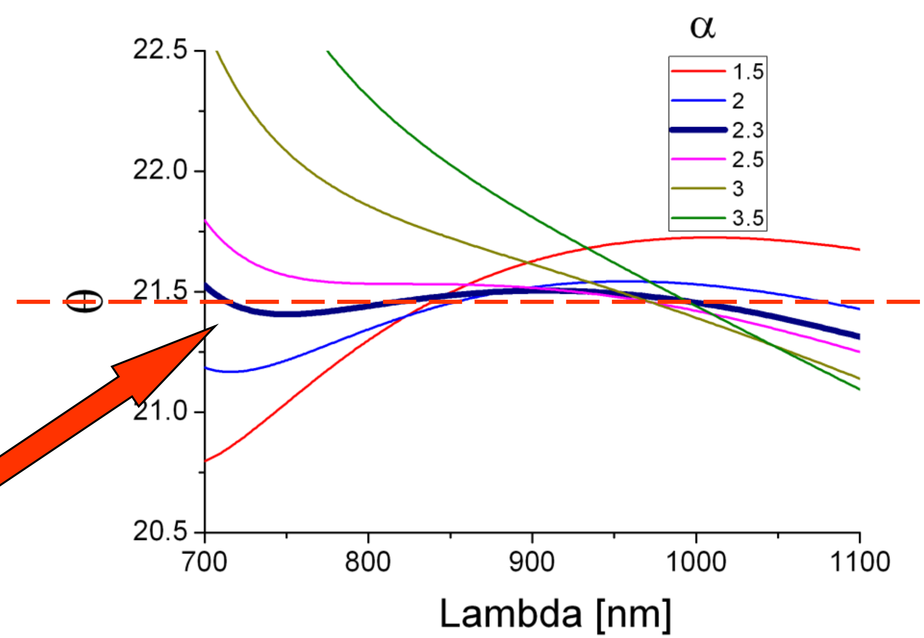
- limited gain (pulses overlap over limited distance only)
- narrow band – amplified pulses are longer than the seed

solution: Noncollinear Optical Parametric Amplifier (NOPA)



$$\Delta k_{\parallel} = k_p \cos\alpha - k_s - k_i \cos\Omega$$
$$\Delta k_{\perp} = k_p \sin\alpha - k_i \sin\Omega$$

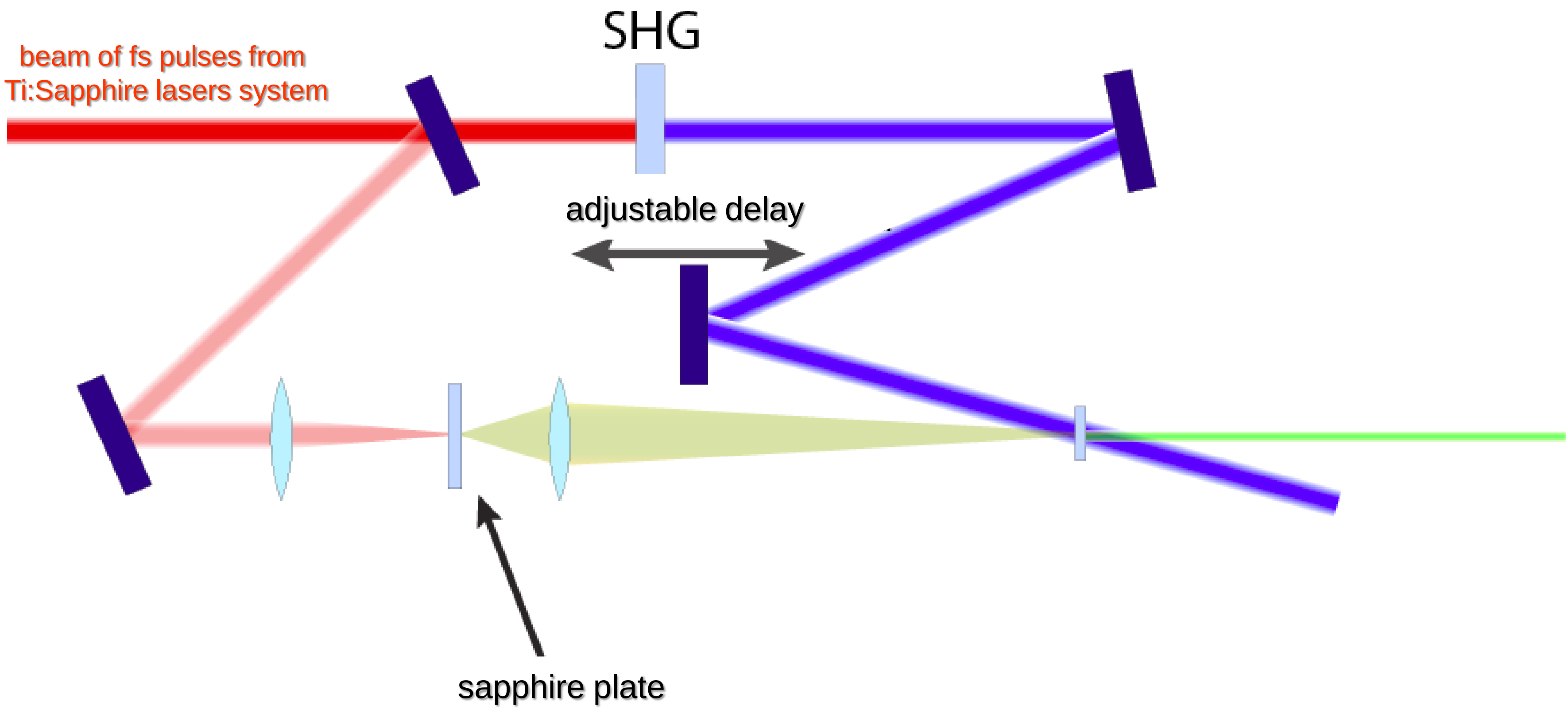
phase matching angle is almost constant over 700-1100nm range



$$\boldsymbol{\Delta k \approx 0}$$

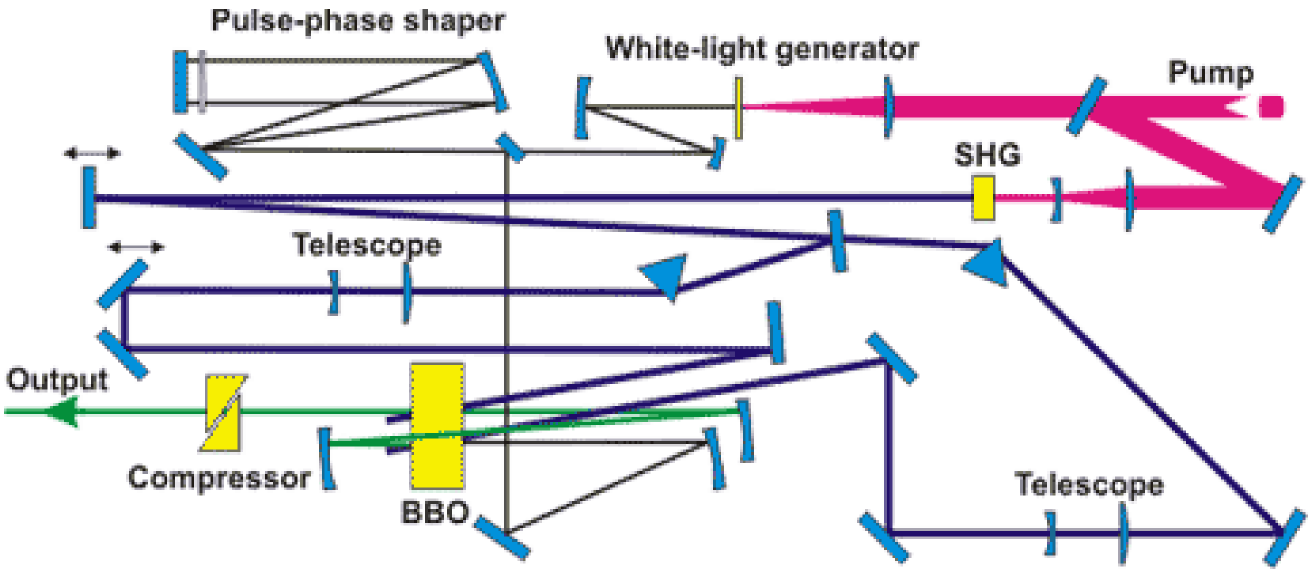
NOPA - construction

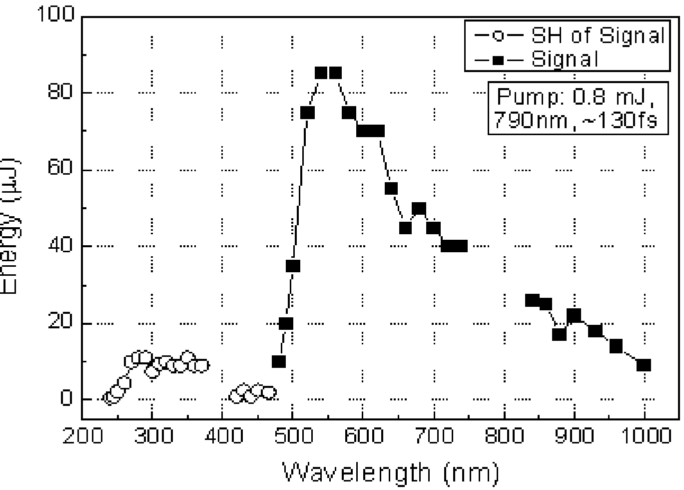
Non-collinear Optical Parametric Amplifier

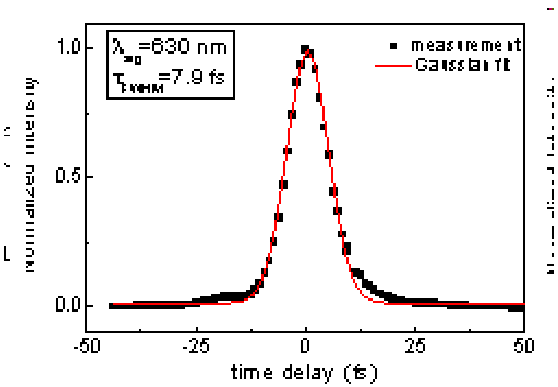


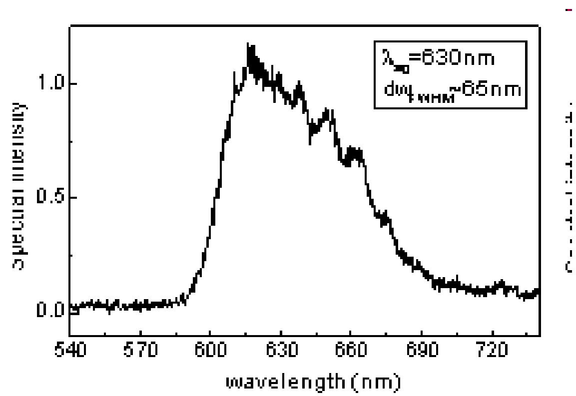
commercial NOPA

example: Light Conversion - NOPA White

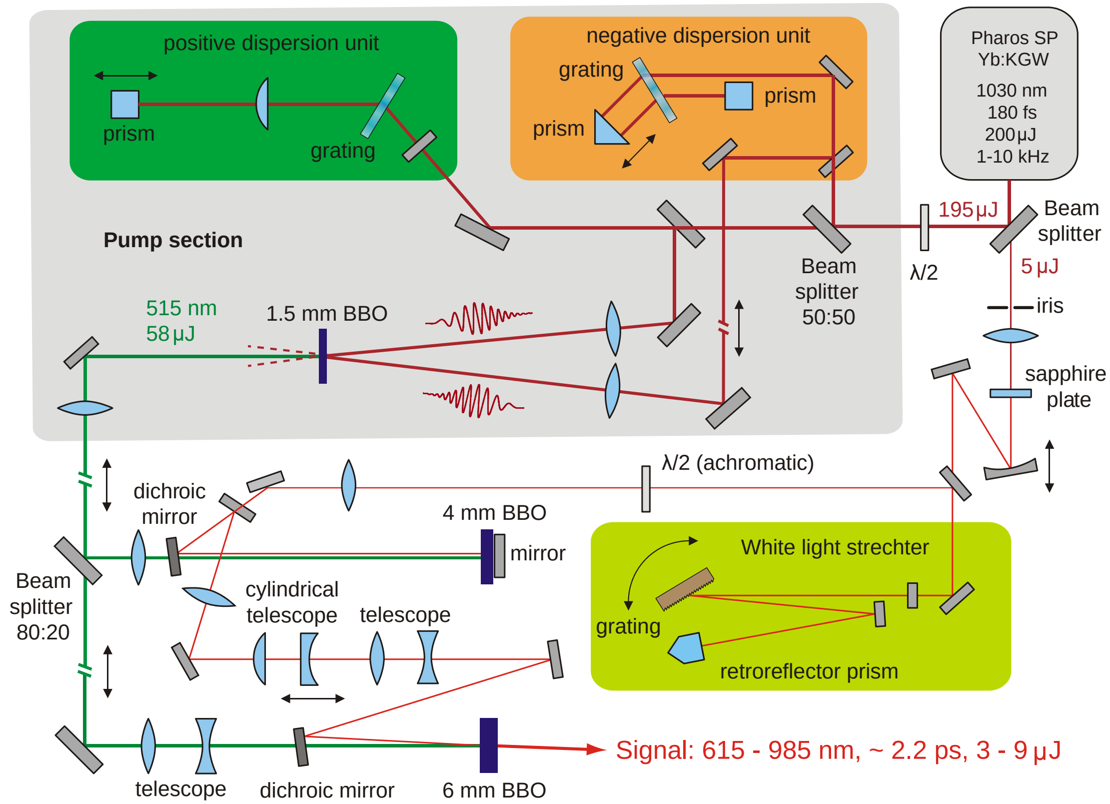


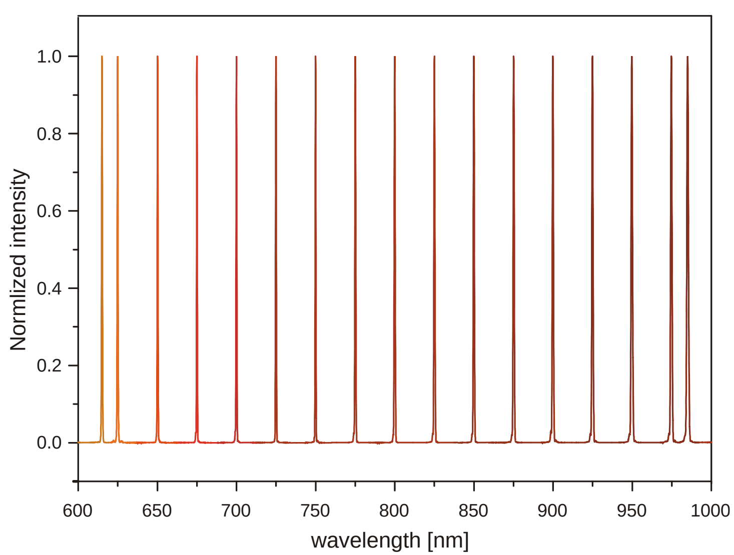


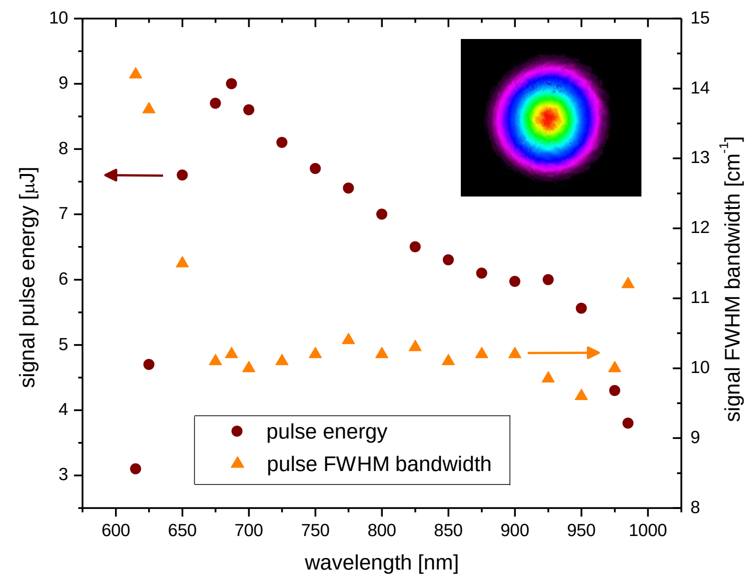




our ps NOPA

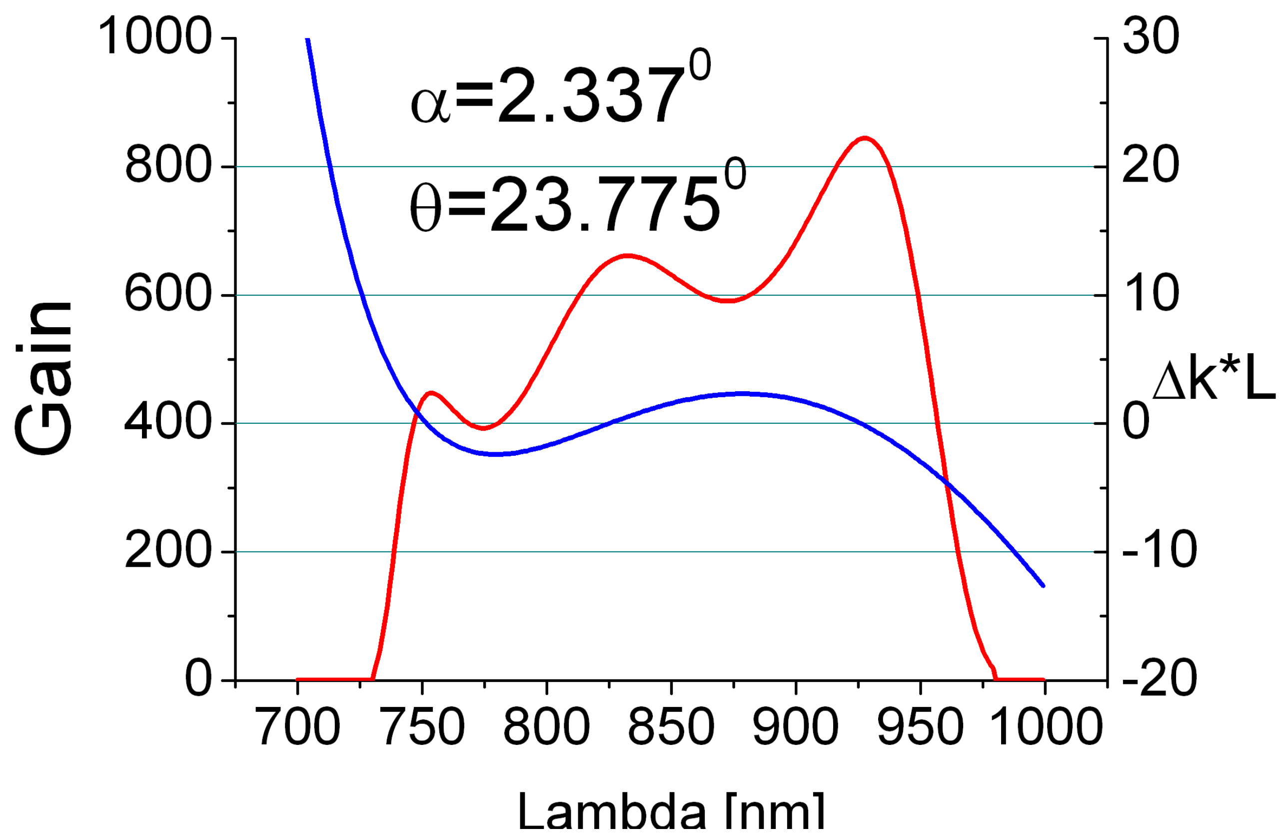






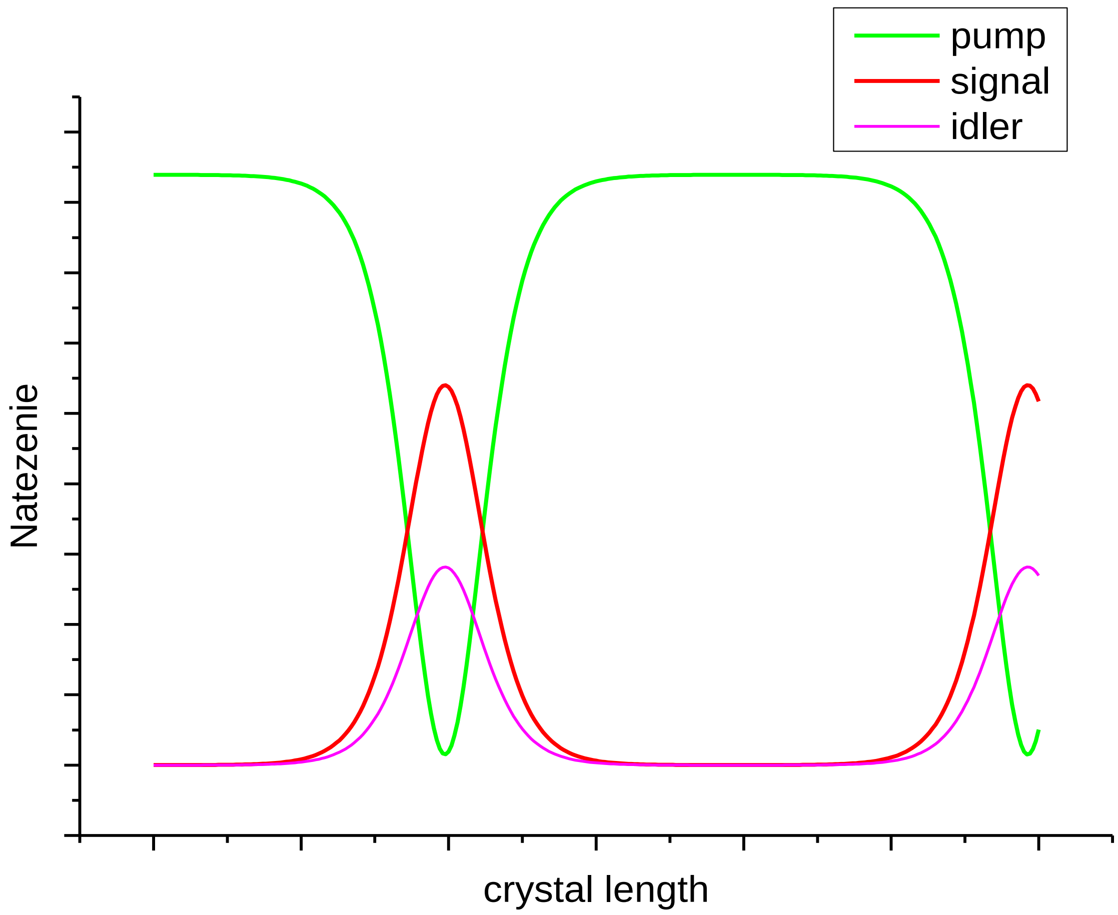
M. Nejbauer, C. Radzewicz, *Efficient spectral shift and compression of femtosecond pulses by parametric amplification of chirped light*, Optics Express **20**, 2136-2142 (2012)

optical chirped pulse parametric amplifier (OPCPA)



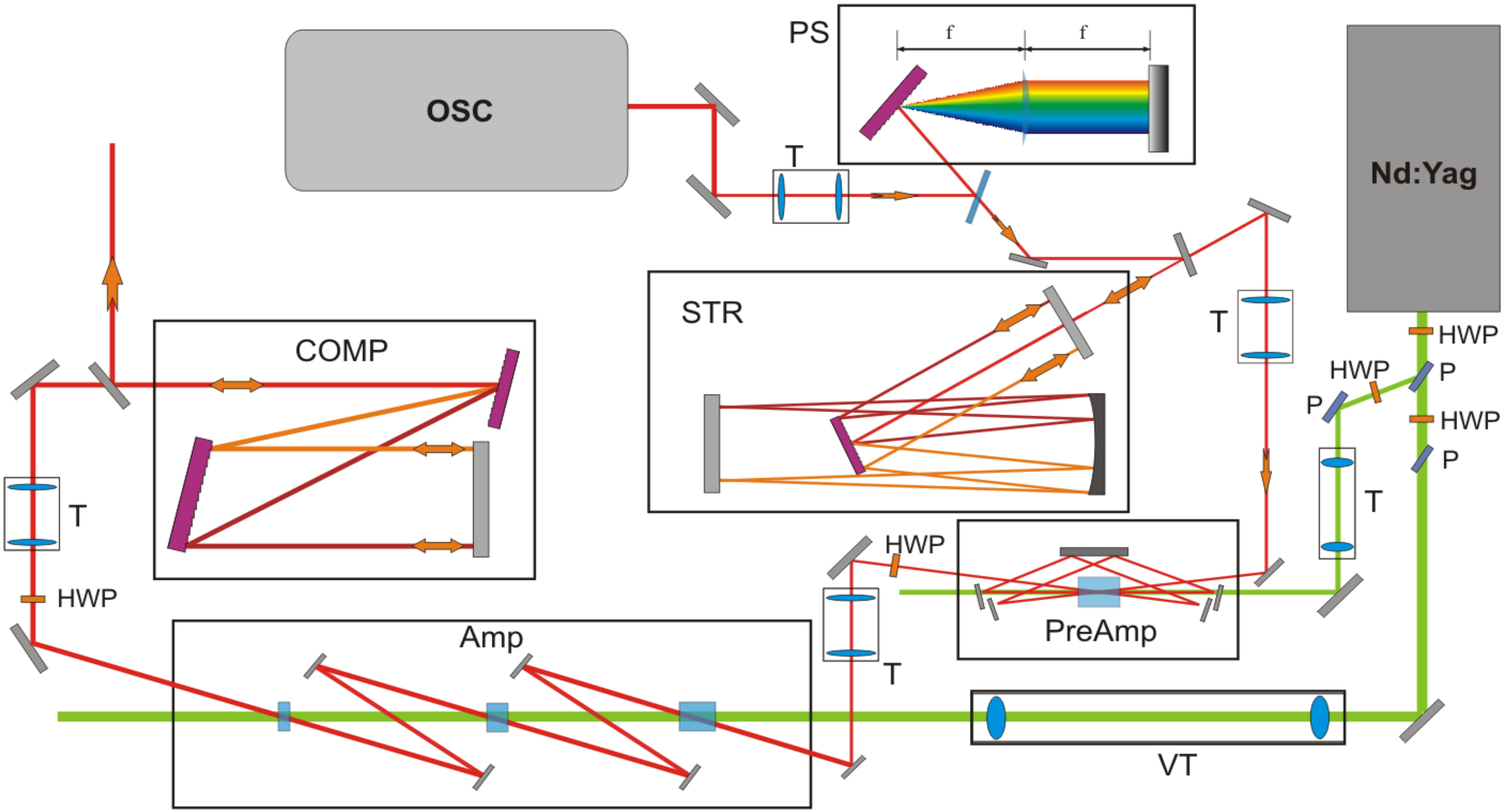
unsaturated gain in BBO crystal pumped by 532 nm pulses

numerical modeling

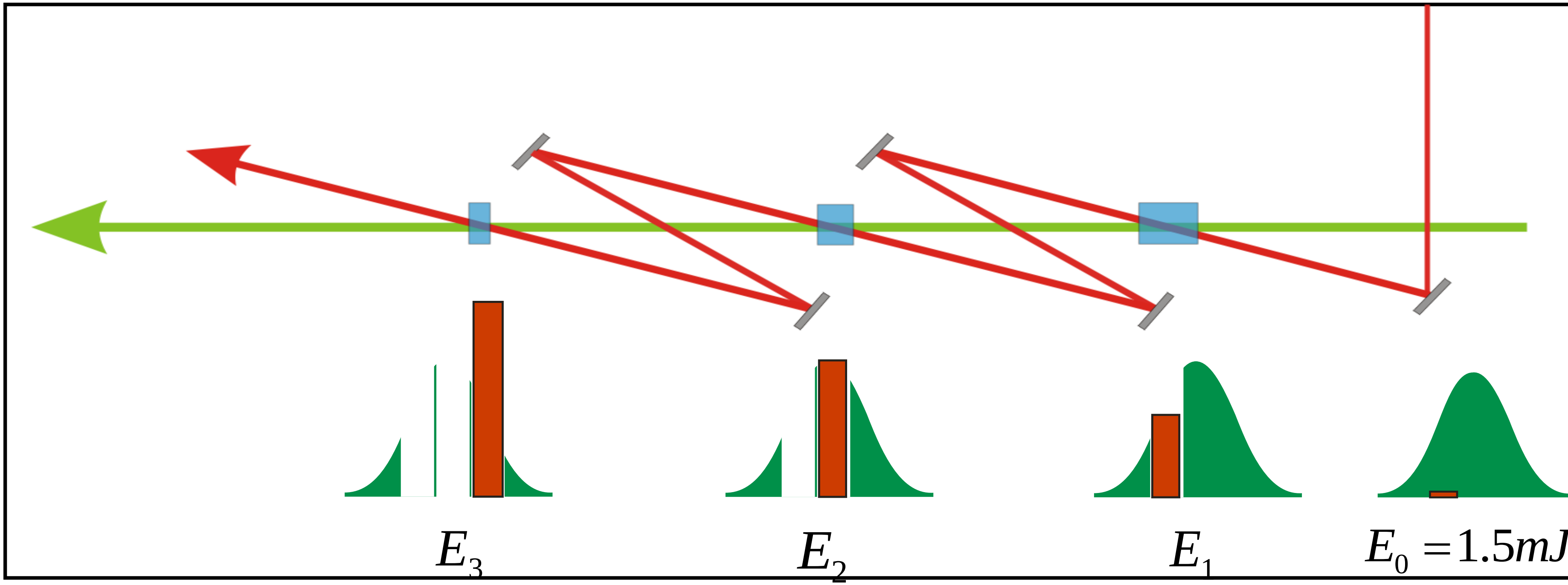


selection of proper parameters (intensities, crystal length, etc. is quite critical
(no problems of this type with lasers amplifiers)

our TW system, 1



our TW system, 2

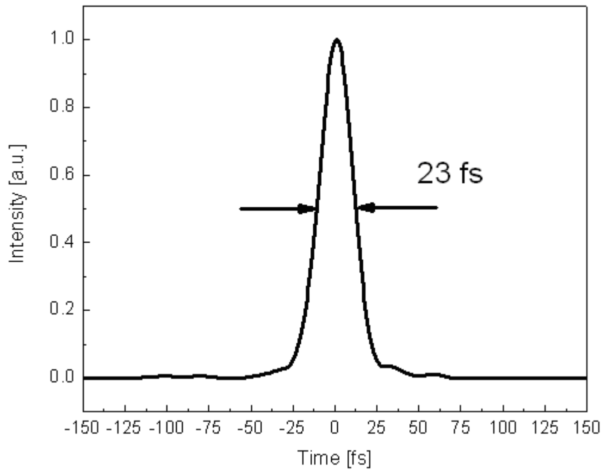


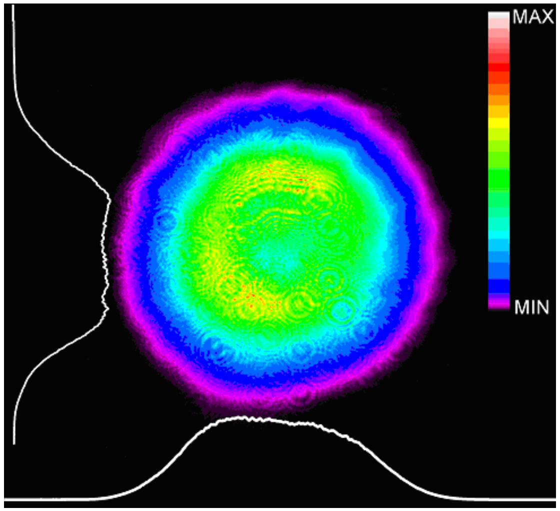
time shearing:

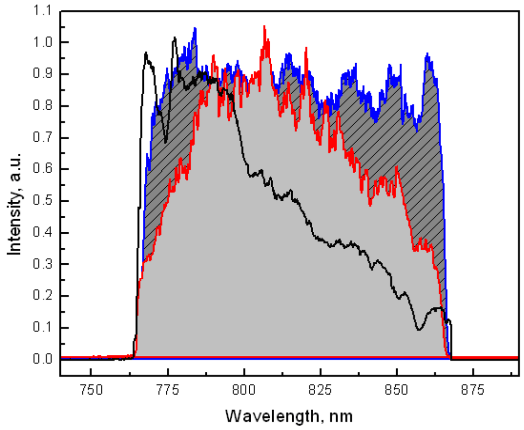
to efficiently extract Energy from a long pump pulse the amplified pulse is shifted in time so in the subsequent crystals it interacts with undepleted part of the pump pulse.

each crystal operates in saturation – stabilization of the output pulse energy

our TW system, 3







pulse energy: ~50 mJ → peak power– 2×10^{12} W

Y. Stepanenko and C. Radzewicz, *Multipass non-collinear optical parametric amplifier for femtosecond pulses*, Opt.Expr. **14**, 779-785 (2006)

P. Wnuk, Y. Stepanenko, and C. Radzewicz, *High gain broadband amplification of ultraviolet pulses in optical parametric chirped pulse amplifier*, Opt. Expr. **18**, 7911-7916 (2010)

P. Wnuk, Y. Stepanenko, and C. Radzewicz, *Multi-terawatt chirped pulse optical parametric amplifier with a time-shear power amplification stage*, OE **17** , 15264-15273 (2009)

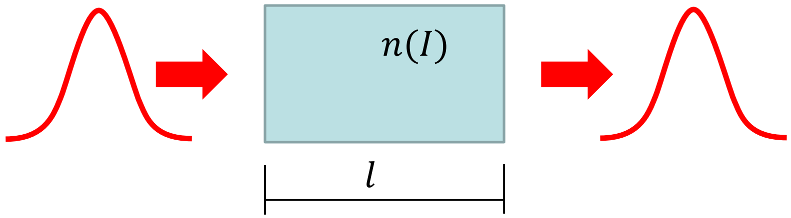
reminder: lecture 8

For high light intensities the index of refraction is not constant – it depends on light intensity. In most practical cases it suffices to include a term proportional to the light intensity :

$$n(I) = n_0 + n_2 I$$

The values of n_2 coefficient are small. For example, for quartz glass (SiO_2) $n_2 \cong 2 \cdot 10^{-16}\mathrm{cm}^2/\mathrm{W}$.

For very high light intensity $I = \frac{100\mathrm{GW}}{\mathrm{cm}^2} = 10^{11}\mathrm{W/cm}^2$ we have $\Delta n = n_2 I \cong 2 \cdot 10^{-5}$.



Nonlinear index of refraction influences: (1) pulse phase and (2) pulse wavefront.

1. Self-phase modulation

Consider 1-D case (plane wave, or fiber): $E_{in}(t) = E_0 e^{i\omega_0 t}$.

At the output $E_{out}(t, z) = E_0 e^{i(\omega_0 t - kl)}$ with

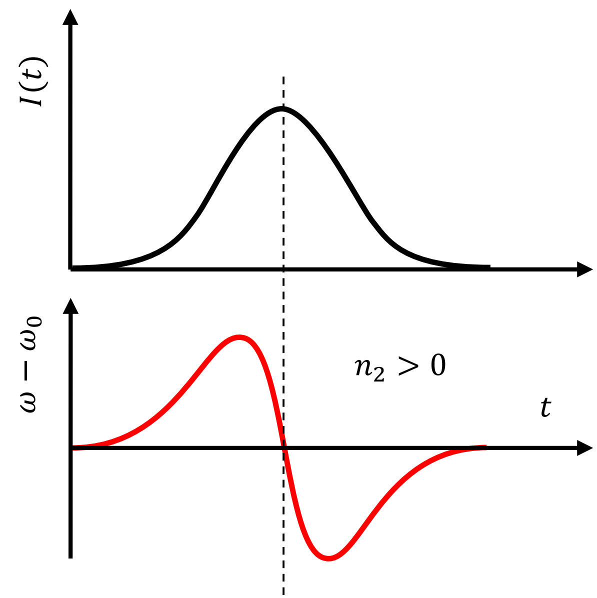
$$k = k(I) = \frac{n(I)\omega_0}{c} = \frac{n_0\omega}{c} + \frac{n_2 I \omega_0}{c} = k_0 + \frac{n_2\omega_0}{c} I$$

which leads to

$$E_{out}(t, z) = E_0 e^{i(\omega_0 t - k_0 z)} e^{i\frac{n_2\omega_0 l}{c} I(t)}$$

resulting in time-dependent phase and frequency

$$\varphi(t) = \omega_0 t + \frac{n_2\omega l}{c} I(t), \qquad \omega(t) = \omega_0 + \frac{n_2\omega l}{c}\frac{dI}{dt}$$



- pulse envelope remains the same
- spectrum is broadened

supercontinuum from photonic crystal fibers