Nonlinear optics in the short pulse regime: basics and practice

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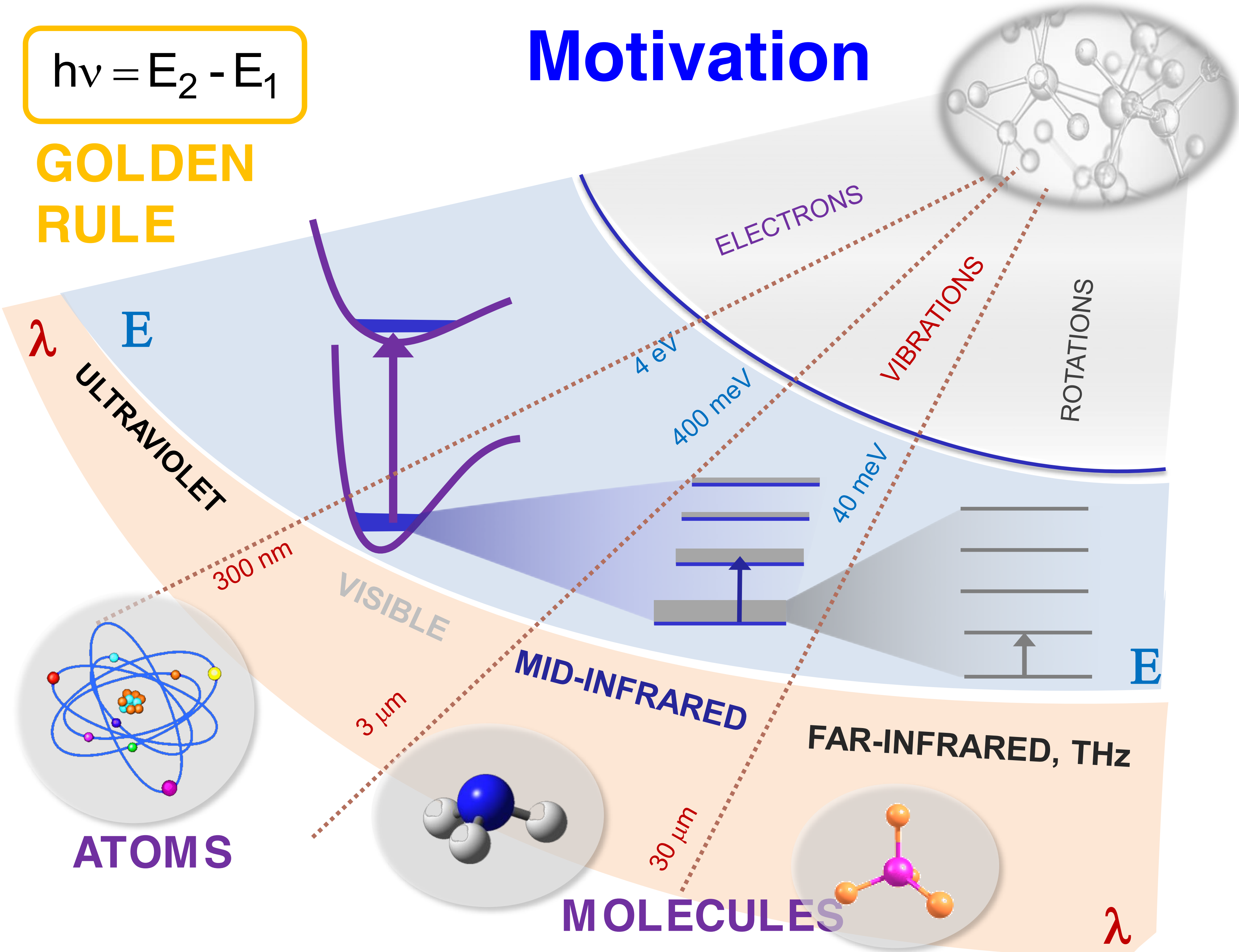
POLITECNICO MILANO

CNR IFN Istituto di Fotonica e Nanotecnologie



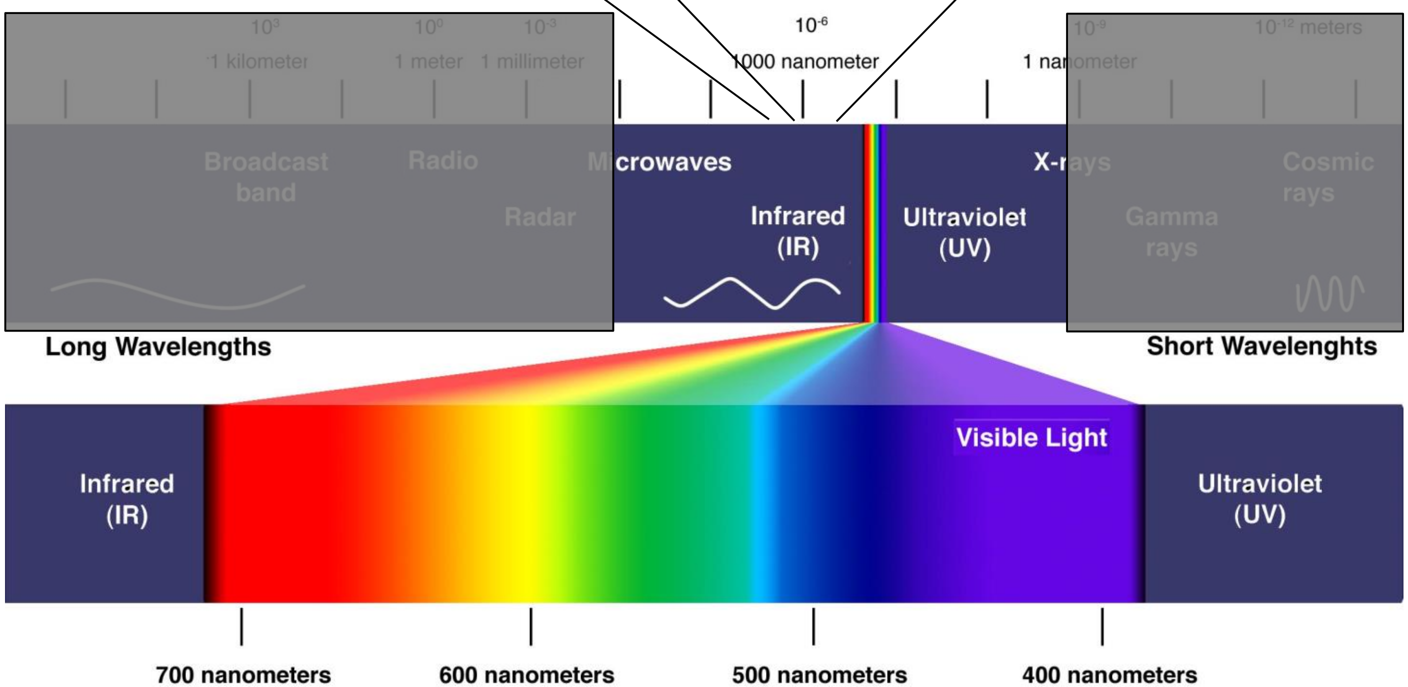
$h\nu = E_2 - E_1$
GOLDEN RULE
Motivation
E
λ
ULTRAVIOLET
ELECTRONS
VIBRATIONS
ROTATIONS
4 eV
400 meV
40 meV
300 nm
VISIBLE
3 μm
MID-INFRARED
30 μm
FAR-INFRARED, THz
E
ATOMS
MOLECULES
λ

Optical frequency comb synthesizers

Er: fibre
$\lambda_0 = 1.55\ \mu m$
$\lambda_{SC} = 0.8\text{-}2.2\ \mu m$
$f_{rep} = 0.1\text{-}0.25$ GHz
$P < 1$ W

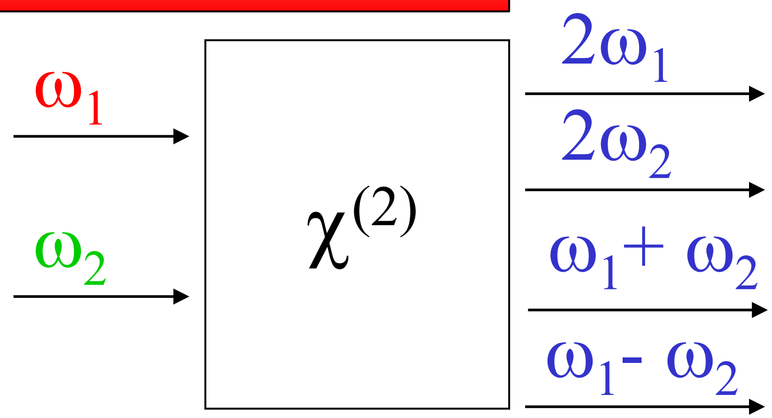
Yb: fibre
$\lambda_0 = 1.05\ \mu m$
$\lambda_{SC} = 0.6\text{-}1.6\ \mu m$
$f_{rep} = 0.1\text{-}1$ GHz
$P < 80$ W

Ti: Sapphire
$\lambda_0 = 0.8\ \mu m$
$\lambda_{SC} = 0.4\text{-}1.2\ \mu m$
$f_{rep} = 0.07\text{-}10$ GHz
$P < 3$ W



How to change spectral range ?

SECOND ORDER NONLINEAR OPTICS !!



$$E(t) = A_1 \exp(-i\omega_1 t) + A_2 \exp(-i\omega_2 t) + cc$$

Optical rectification

Second harmonic generation (SHG)

$$P^{(2)}(t) = \chi^{(2)} E^2(t) = 2\chi^{(2)}\left(A_1 A_1^* + A_2 A_2^*\right) + \chi^{(2)}[A_1^2 \exp(-2i\omega_1 t) +$$
$$A_2^2 \exp(-2i\omega_2 t) + +2A_1 A_2 \exp[-i(\omega_1 + \omega_2)t] + 2A_1 A_2^* \exp[-i(\omega_1 - \omega_2)t] + cc]$$

Difference frequency generation (DFG)

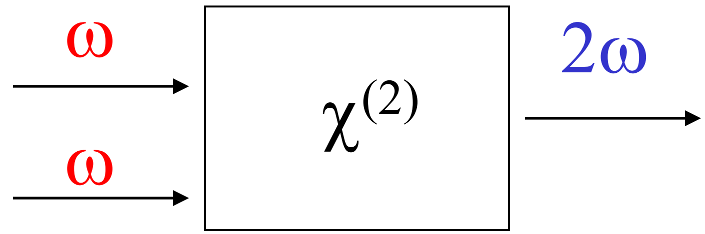
Sum frequency generation (SFG)

OUTLINE

- Equations governing a cw second order parametric process
- The problem of phase matching
- The equations of linear pulse propagation
- Parametric processes in the femtosecond pulse regime
- Examples: analytical and numerical discussion

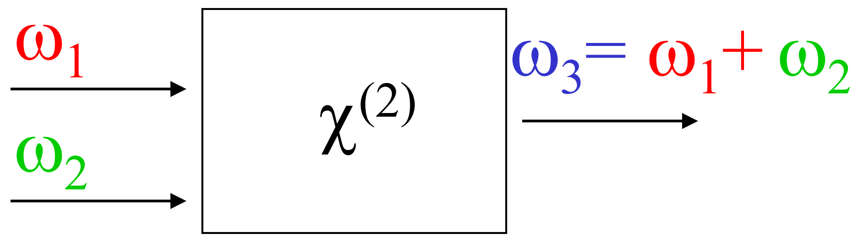
The photons picture

SHG



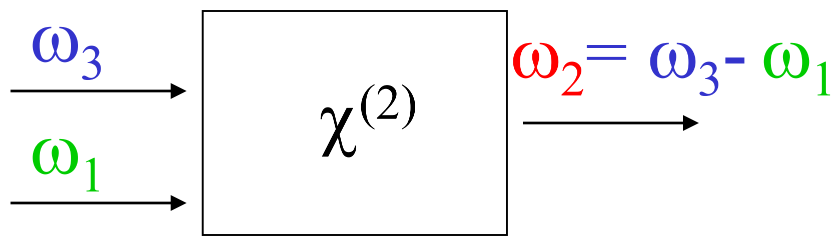
$$\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$$

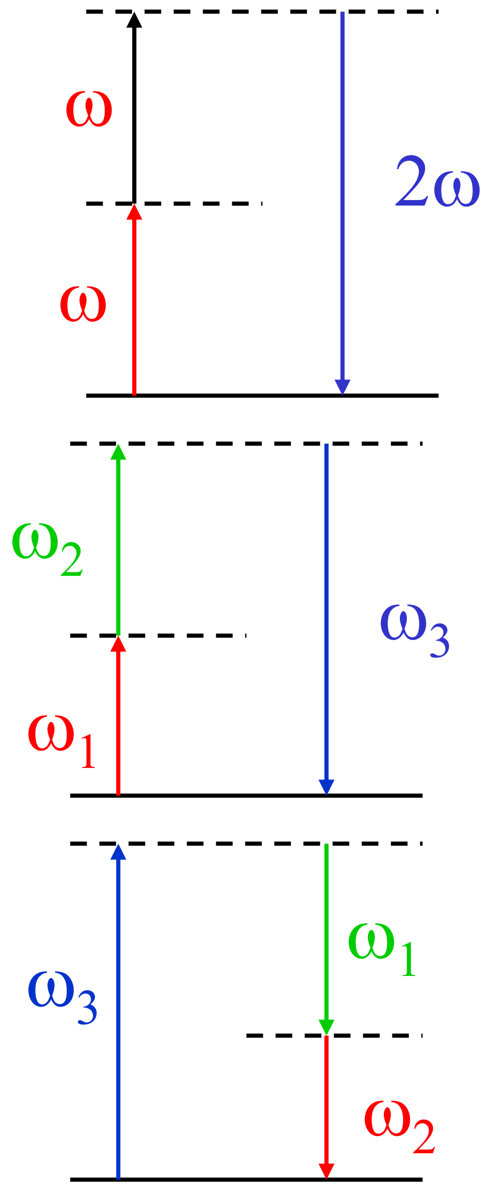
SFG



$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$$

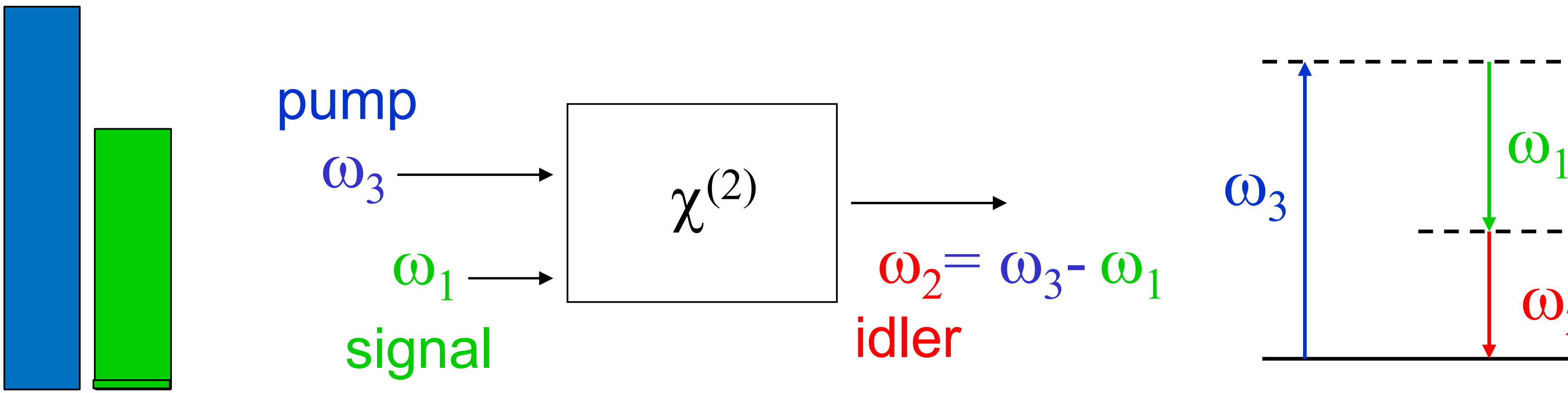
DFG





Optical parametric amplification (OPA) & optical parametric generation (OPG): what are they ?

- They are the same process as DFG, but differ in the initial conditions

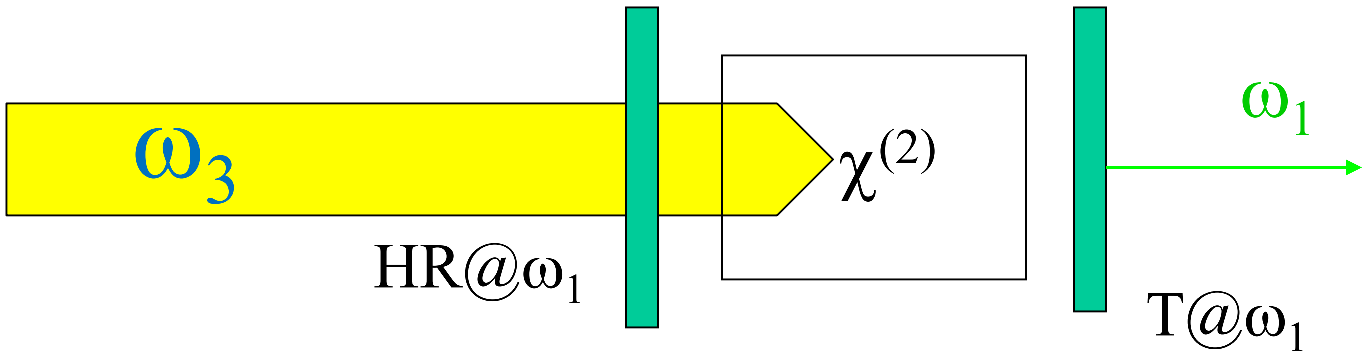


- In DFG, ω_3 and ω_1 have comparable energies and you look for an intense ω_2

- In OPA, ω_1 has an energy 100-10000 times lower than ω_3 and you look for a strong amplification of ω_1 (ω_1 acts as a seed)

- In OPG, ω_1 photons come from vacuum noise and you are looking for extreme parametric gains (10 nJ $\rightarrow$ > 10^{11} photons !!)

If the gain is not enough…
and/or you lack seed pulses…..



- You may enclose your crystal in an optical cavity
- …shine your powerful pump on the crystal
- …eventually get oscillation, like in a laser, if the parametric gain exceeds losses

OPTICAL PARAMETRIC OSCILLATOR

Femtosecond OPOs vs. OPAs:

Femtosecond OPOs

- are pumped by simple laser oscillators
- provide high repetition rates (100 MHz)
- have low output energy (nJ level)
- require matching of the OPO cavity length to pump laser
- large yet not huge oscillation bandwidth

FREQUENCY COMBS

Femtosecond OPAs

- require pumping by amplified laser systems
- provide low repetition rates (1-100 kHz)
- have high output energy (μJ-mJ level)
- are easy to operate (no length stabilization)
- ultrabroad bandwidth, up to the few-cycles regime

The wave equations for second order parametric processes

The wave equation for nonlinear optical media

- Starting from Maxwell's equations for an insulating medium without free charges and currents, we get the wave equation

$$\nabla^2 \boldsymbol{E} - \frac{1}{c_0{}^2}\frac{\partial^2 \boldsymbol{E}}{\partial t^2} = \frac{1}{\varepsilon_0\, c_0{}^2}\frac{\partial^2 \boldsymbol{P}}{\partial t^2}$$

- The polarization of the medium is made of a linear and a nonlinear contribution

$$\boldsymbol{P} = \boldsymbol{P}_L + \boldsymbol{P}_{NL}$$

- For a continuous wave, the linear polarization is $\boldsymbol{P}_L = \varepsilon_0\,(\varepsilon_r - 1)\,\boldsymbol{E}$

- Making the scalar approximation and considering a plane wave, the propagation equation becomes

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{\varepsilon_0\, c_0{}^2}\frac{\partial^2 P_{NL}}{\partial t^2}$$

The slowly varying envelope approximation

- Starting from the scalar propagation equation

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$$

we look for a solution $E(z,t) = A(z)\exp[i(kz - \omega t)]$

with $P_{NL}(z,t) = P(z)\exp[i(k_P z - \omega t)]$

- By substitution, we get the equation

$$\frac{d^2 A}{dz^2} + 2ik\frac{dA}{dz} - k^2 A + \frac{\omega^2}{c^2} A = -\mu_0 \omega^2 P \exp[i(k_p - k)z]$$

- Assuming $\frac{d^2 A}{dz^2} << 2ik\frac{dA}{dz}$ (slowly varying envelope approximation, SVEA) we get the equation

$$\frac{dA}{dz} = i\mu_0 \frac{\omega^2}{2k} P \exp[i(k_p - k)z]$$

The nonlinear polarization in second-order parametric interactions

- Consider the superposition of three waves at frequencies ω_1, ω_2 and ω_3 with $\omega_1 + \omega_2 = \omega_3$

$$E(z,t) = A_1(z)\exp[i(k_1 z - \omega_1 t)] + A_2(z)\exp[i(k_2 z - \omega_2 t)] + A_3(z)\exp[i(k_3 z - \omega_3 t)]$$

- By second order nonlinear effect, the following polarizations are generated at the three frequencies

$$P_{1NL}(z,t) = \varepsilon_0 2 d_{eff}\, A_2^*(z)\, A_3(z) \exp[i((k_3 - k_2)z - \omega_1 t)]$$

$$P_{2NL}(z,t) = \varepsilon_0 2 d_{eff}\, A_1^*(z)\, A_3(z) \exp[i((k_3 - k_1)z - \omega_2 t)]$$

$$P_{3NL}(z,t) = \varepsilon_0 2 d_{eff}\, A_1(z)\, A_2(z) \exp[i((k_1 + k_2)z - \omega_3 t)]$$

where d_{eff} is an effective second order nonlinear coefficient

Three-frequency interaction in a second order nonlinear medium

- Consider three waves at ω_3 (pump), ω_1 (signal) and ω_2 (idler), with $\omega_1 + \omega_2 = \omega_3$. We obtain the following equations

$$\frac{\partial A_1}{\partial z} = i \frac{\omega_1 d_{eff}}{n_1 c} A_2^* A_3 \exp[i \Delta k z]$$

$$\frac{\partial A_2}{\partial z} = i \frac{\omega_2 d_{eff}}{n_2 c} A_1^* A_3 \exp[i \Delta k z]$$

$$\frac{\partial A_3}{\partial z} = i \frac{\omega_3 d_{eff}}{n_3 c} A_1 A_2 \exp[-i \Delta k z]$$

where $\Delta k = k_3 - k_2 - k_1$ is the wave vector mismatch between the three waves

Setting $\Delta k = 0$ is crucial to get highly efficient energy transfer between the interacting waves

OPA/DFG solution for small pump depletion

- By **neglecting pump depletion (A_3 = cost.)** and assuming an input beam at the signal frequency ω_1 and no input at the idler frequency ω_2 ($A_2(0) = 0$) the coupled differential equations admit the solution:

$$I_1(L) = I_1(0)\left[1 + \frac{\gamma^2}{g^2}\right] sinh^2(gL)$$

$$I_2(L) = I_1(0)\frac{\omega_2}{\omega_1}\frac{\gamma^2}{g^2} sinh^2(gL)$$

with g and γ given by:

$$g = \sqrt{\gamma^2 - \left(\frac{\Delta k}{2}\right)^2} \qquad \gamma = \sqrt{\frac{\omega_1 \omega_2 d_{eff}}{2 n_1 n_2 n_3 \varepsilon_0 c^3} I_3}$$

the latter representing a figure of merit for the parametric gain.
The presence of a phase-mismatch clearly affects such gain.

Parametric gain

- In the high gain approximation ($\gamma L>>1$) and under phase-matching ($\Delta k = 0$): one has:

$$I_1(L) = \frac{I_1(0)}{4}\exp[2\gamma L] \qquad I_2(L) = \frac{I_1(0)}{4}\frac{\omega_2}{\omega_1}\exp[2\gamma L]$$

- This allows us to define a parametric gain:

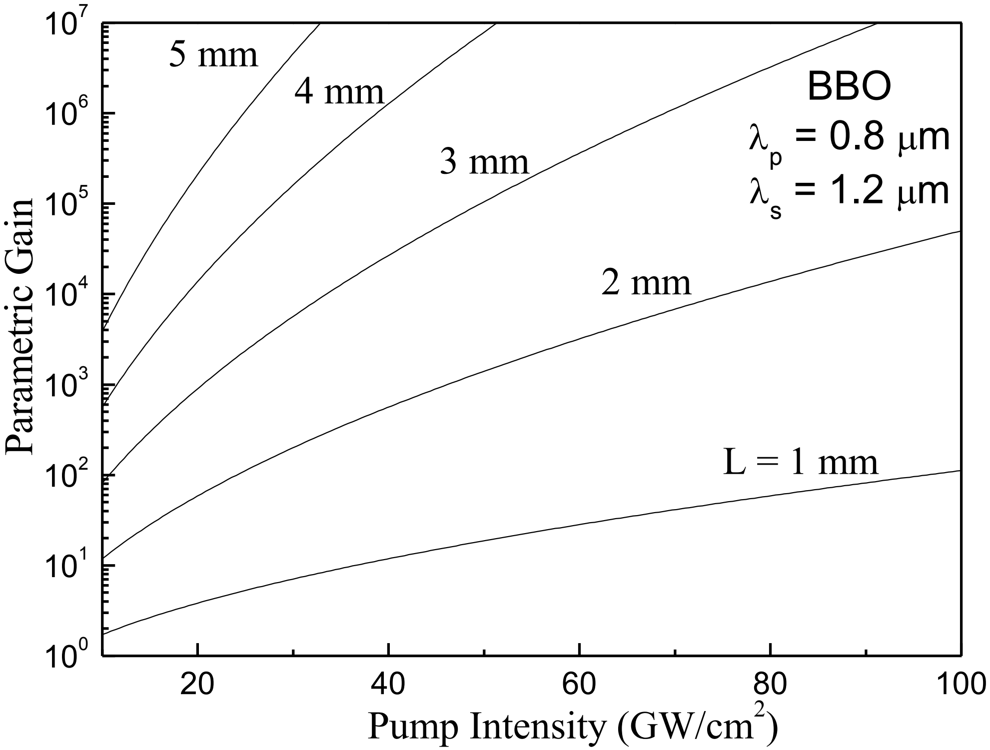
$$G = \frac{I_1(L)}{I_1(0)} = \frac{1}{4}exp[2\gamma L] = \boxed{\frac{1}{4}exp\left[2\gamma\sqrt{\frac{\omega_1\omega_2 d_{eff}}{2n_1n_2n_3\varepsilon_0c^3}I_3}\,L\right]}$$

For high gain we need high pump intensity (ultrashort pulses are good!), large nonlinear coefficient d_{eff} and high signal and idler frequencies

The gain is exponential since the presence of a seed photon at the signal wavelength stimulates the generation of an additional signal photon and of a photon at the idler wavelength. Due to the symmetry of signal and idler, the amplification of an idler photon stimulates in turn the generation of a signal photon. Therefore, the generation of the signal field reinforces the generation of the idler field and viceversa, giving rise to a positive feedback

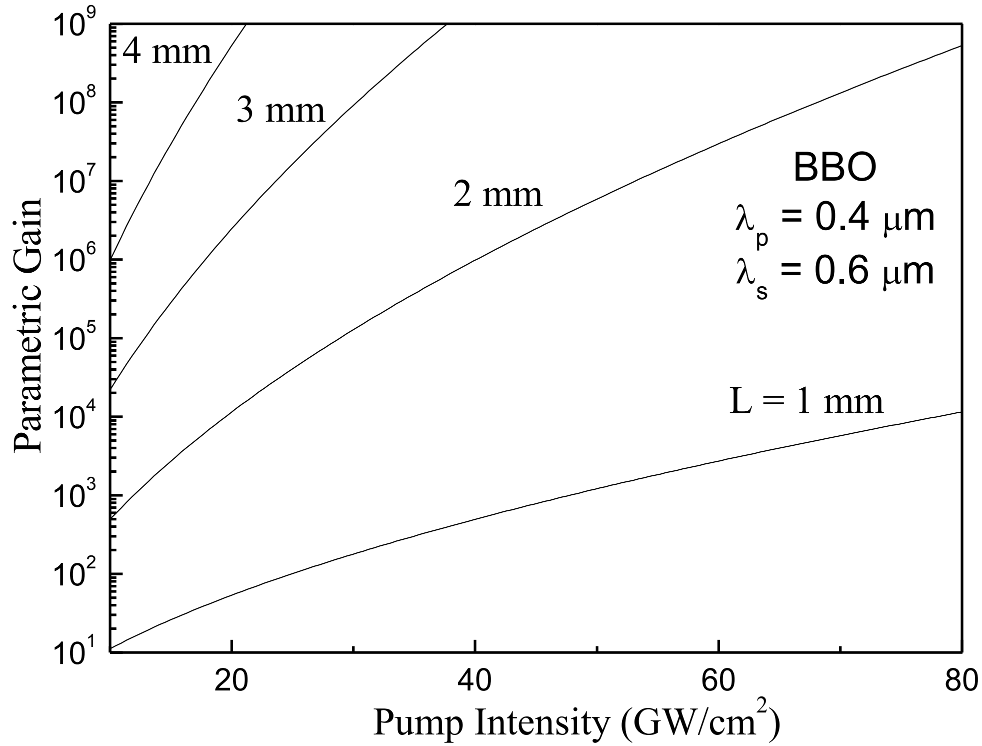
Parametric gain: examples with BBO

Red-pumped BBO crystal



Blue-pumped BBO crystal: higher gain because

$$\gamma \propto \sqrt{\omega_1 \omega_2}$$



G. Cerullo and S. De Silvestri, Rev. Sci. Instrum. **74**, 1 (2003).

Are those gains achievable with frequency combs ?

$$\gamma = \sqrt{\frac{\omega_s \omega_i d_{eff}}{2 n_p n_s n_i \varepsilon_0 c^3} I_p}$$

$$\lambda_p = 0.8\mu m; \quad \lambda_s = 1.2\mu m$$

$$\frac{1}{\lambda_i} = \frac{1}{\lambda_p} - \frac{1}{\lambda_s} \rightarrow \lambda_i = 2.4\mu m$$

$$BBO : n_{p,s,i} \approx 1.6; d_{eff} = 2\, pm/V$$

HIGH

$$I_p = \frac{U}{\tau A_{eff}}; \quad U = 10nJ; \quad \tau = 60fs$$

$$A_{eff} = \frac{1}{2}\pi w_0^2 = 15700\mu m^2$$

$$G = \frac{1}{4} exp[2\gamma L]$$

$L = 2mm$ It complies with bandwidth and temporal walk-off issues (see next slides)

$w_0 = 100\mu m$ It complies with spatial walk-off issues

I_p = 1 GW/cm^2

G = 0.89 !!!

Energy conservation in parametric interaction

- By manipulation of the previous equations, it is easy to show that

$$\frac{dI_1}{dz} + \frac{dI_2}{dz} + \frac{dI_3}{dz} = 0$$

i.e. the sum of the energies of the three waves is conserved (assuming a lossless medium)

- In addition, the following relationship (Manley-Rowe) can be proven

$$\frac{1}{\omega_1}\frac{dI_1}{dz} = \frac{1}{\omega_2}\frac{dI_2}{dz} = -\frac{1}{\omega_3}\frac{dI_3}{dz}$$

stating photon conservation: one photon at ω_3 is annihilated and two photons at ω_1 and ω_2 are simultaneously created

The problem of phase matching

SHG process

- Let us consider for simplicity second harmonic generation (SHG) ($\omega_1 = \omega_2 = \omega$, $\omega_3 = 2\omega$, $A_1 = A_2 = A_\omega$)
- Neglecting pump depletion ($A_\omega \approx$ cost)

$$\frac{dA_{2\omega}}{dz} = i\frac{2\omega\, d_{eff}}{n_{2\omega} c} A_\omega^2 \exp[-i\Delta k\, z]$$

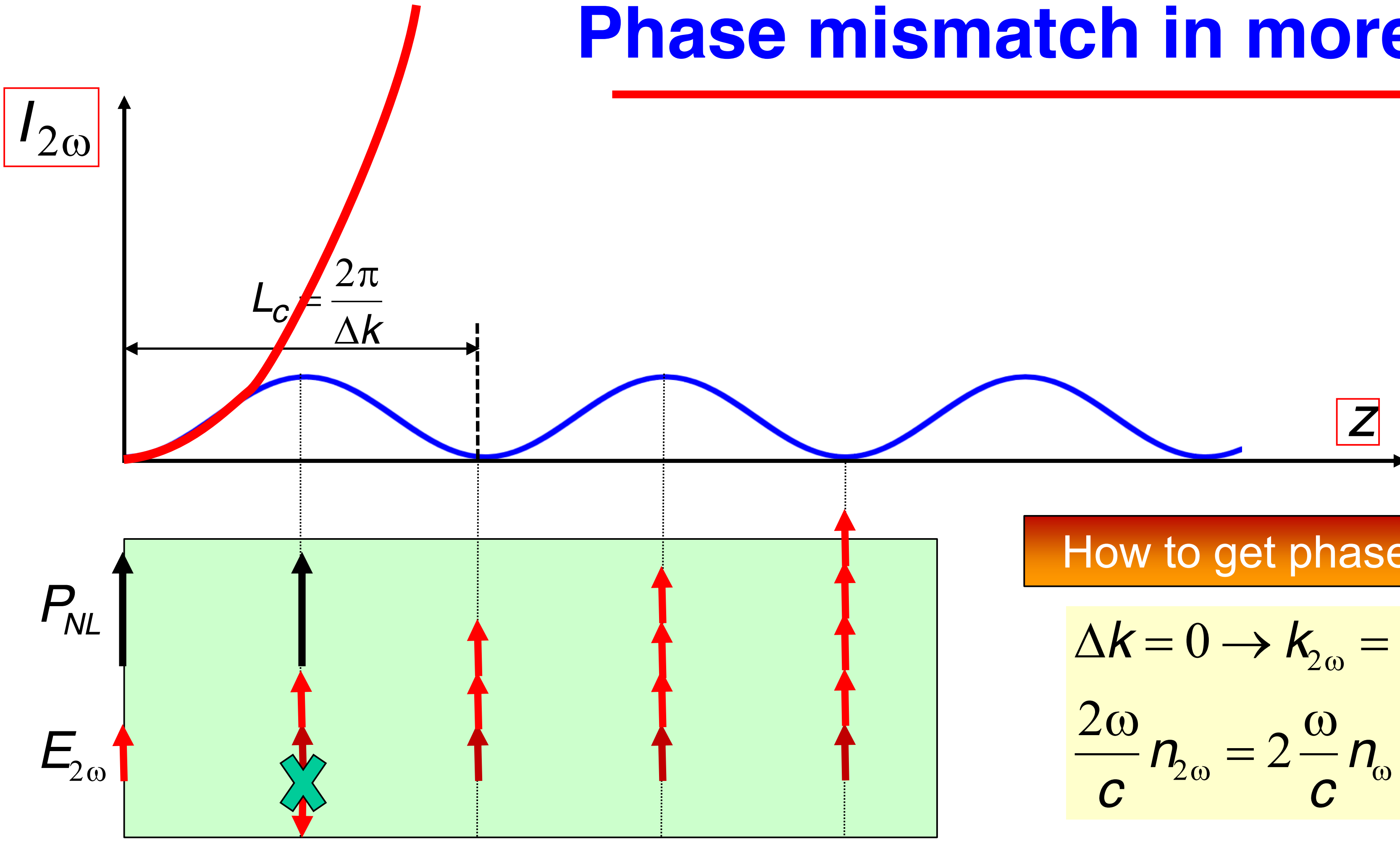
- After a length L of nonlinear medium

$$I_{2\omega}(L) = \gamma^2 I_\omega L^2 \, sinc^2\left(\frac{\Delta kL}{2}\right) = \frac{4\gamma\, I_\omega}{\Delta k^2} sin^2\left(\frac{\Delta kL}{2}\right)$$

$$I_{2\omega}(L) \propto I_\omega^2$$
$$I_{2\omega}(L) \propto d_{eff}^2$$

$$I_{2\omega}(L) \propto L^2 \qquad \Delta k = 0$$
$$I_{2\omega}(L) \propto sin^2\left(\frac{\Delta kL}{2}\right) \qquad \Delta k \neq 0$$

Phase mismatch in more detail



How to get phase matching ?

$$\Delta k = 0 \rightarrow k_{2\omega} = 2k_{\omega}$$

$$\frac{2\omega}{c} n_{2\omega} = 2\frac{\omega}{c} n_{\omega} \rightarrow n_{2\omega} = n_{\omega}$$

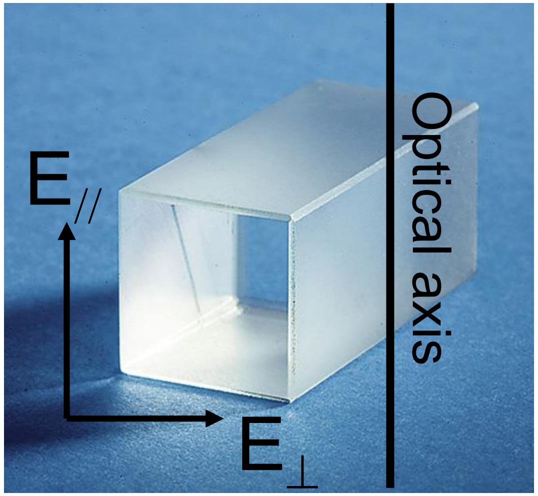
?

Driving wave $\quad P_{NL} \propto E_{\omega}^2 \propto A_{\omega}^2 \, \exp\left[i\left(2k_{\omega} z - 2\omega t\right)\right]$

Generated wave $\quad E_{2\omega} \propto \exp\left[i\left(k_{2\omega} z - 2\omega t\right)\right]$

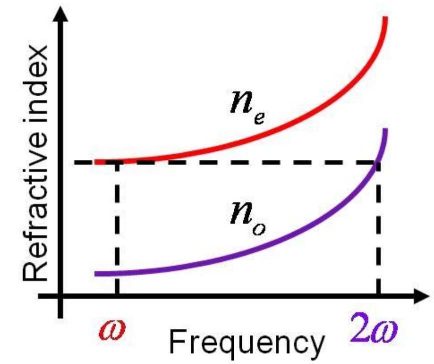
Phase shift at $L_c/2$ $\quad \varphi\left(P_{NL}\right) - \varphi\left(E_{2\omega}\right) \propto \left(2k_{\omega} - k_{2\omega}\right)\cdot\frac{L_c}{2} = \Delta k \cdot \frac{\pi}{\Delta k} = \pi$

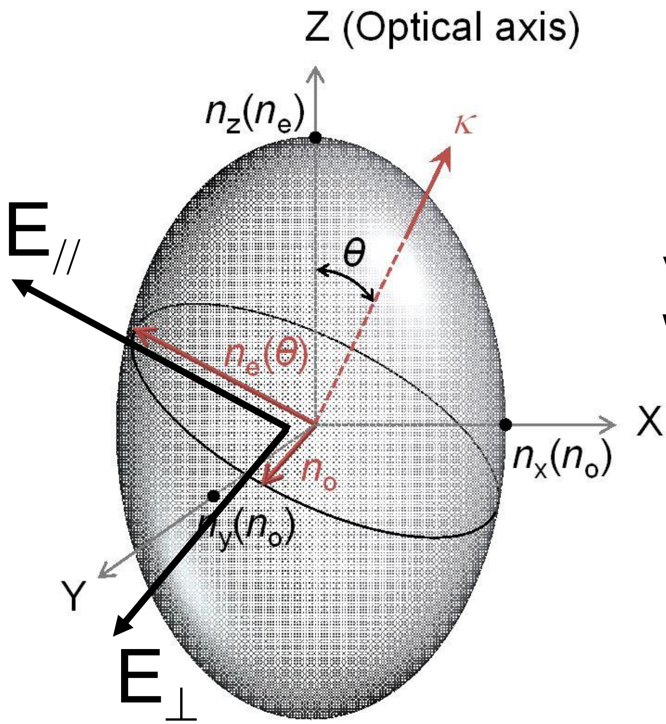
Propagation in birefringent media



In the simpler case of uniaxial crystals, propagation may be described recurring to a pair of refractive indices, n_e and n_o (extraordinary and ordinary index, respectively, each one with its own dispersion), and to an index-ellipsoid model:

$$\frac{X^2}{n_o^2}+\frac{Y^2}{n_o^2}+\frac{Z^2}{n_e^2}=1$$





Each propagation direction, which is given by the wave-vector ***k***, defines in the plane perpendicular to ***k*** an ellipse whose axes correspond to two polarization eigenstates:

$$E_{\perp} \rightarrow n_o$$

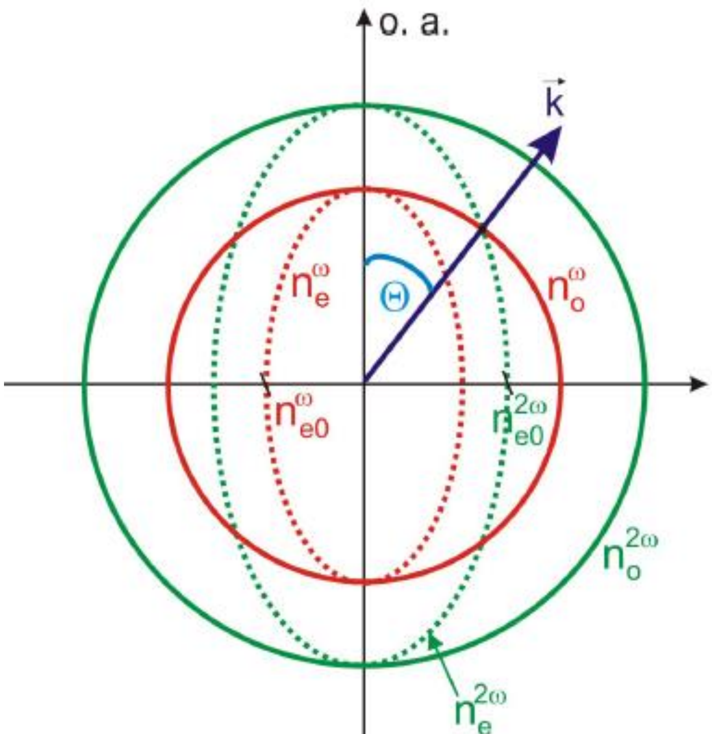
Ordinary wave

$$E_{//} \rightarrow n_e^2(\theta)=\frac{n_o^2\, n_e^2}{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta}$$

Extraordinary wave

Birifringence phase matching

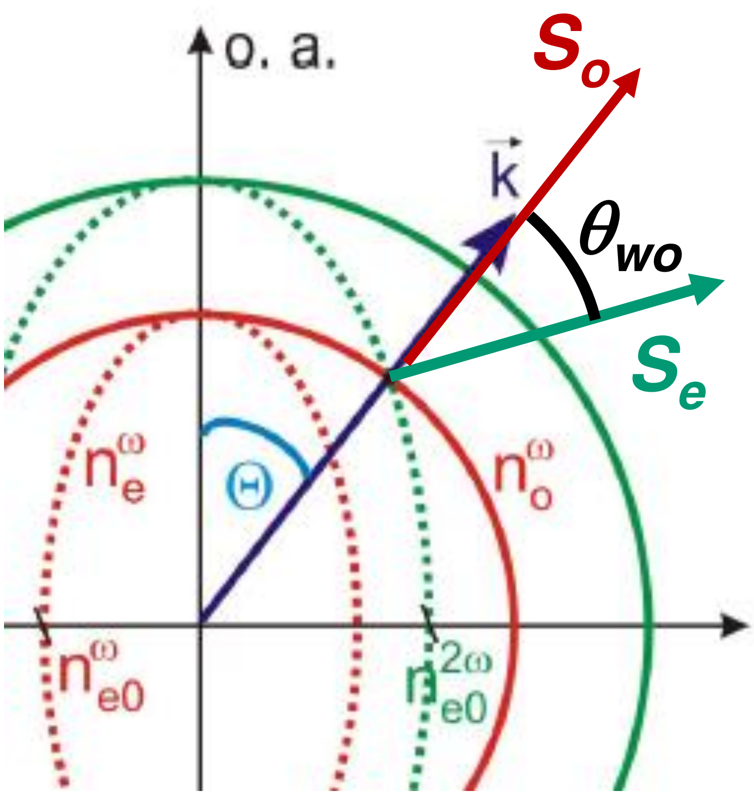
Negative uniaxial crystals: $n_e < n_o$	Positive uniaxial crystals: $n_e > n_o$
$n_e(2\omega,\theta) = n_o(\omega)$	$n_e(\omega,\theta) = n_o(2\omega)$



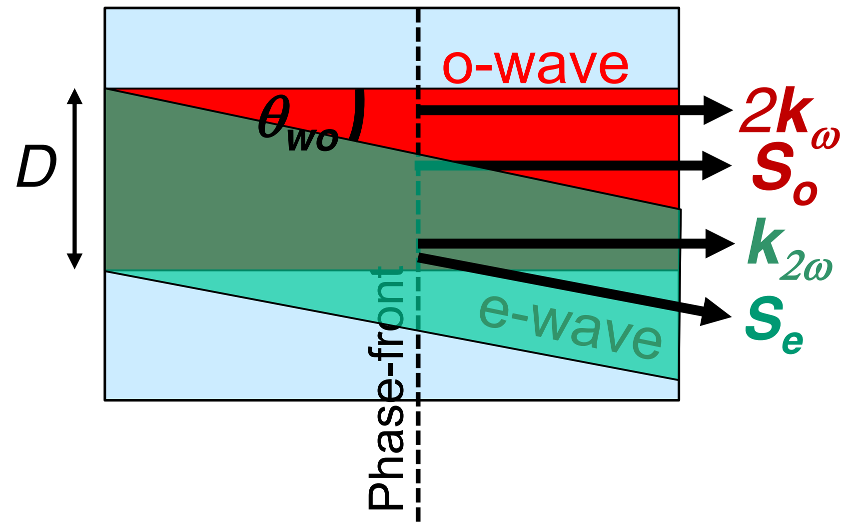
Polar diagram showing the refractive index dependence as a function of the angle θ between ***k*** and the optical axis, at the two frequencies

- NOTE: the refractive indexes n_e and n_o at each frequency are obtained by Sellmeier equations
- Birifringence phase-matching involves coupling between **orthogonally polarized fields** - **non diagonal terms** of the second-order nonlinear-susceptibility χ^2 tensor

The spatial walk-off probelm



- In birefringent crystals the pointing vector of the extraordinary wave $S_e = E \times H$, **which gives the energy propagation direction,** suffers from an angular offset from the ***k*** vector. This is referred to as **the walk-off angle** θ_{wo}.
- **It seriously limits the interaction length *L* for a given input field diameter *D*:**



The Pointying vector of the extraordinary wave S_e my be shown to be perpendicular to the extraordinary normal index surface at its crossing point with ***k***. This does not happen for the ordinary wave, with S_o // ***k*.**

- Length limitation approximately given by:

$$L = D\tan\theta_{wo}$$

Birifringence phase matching: examples

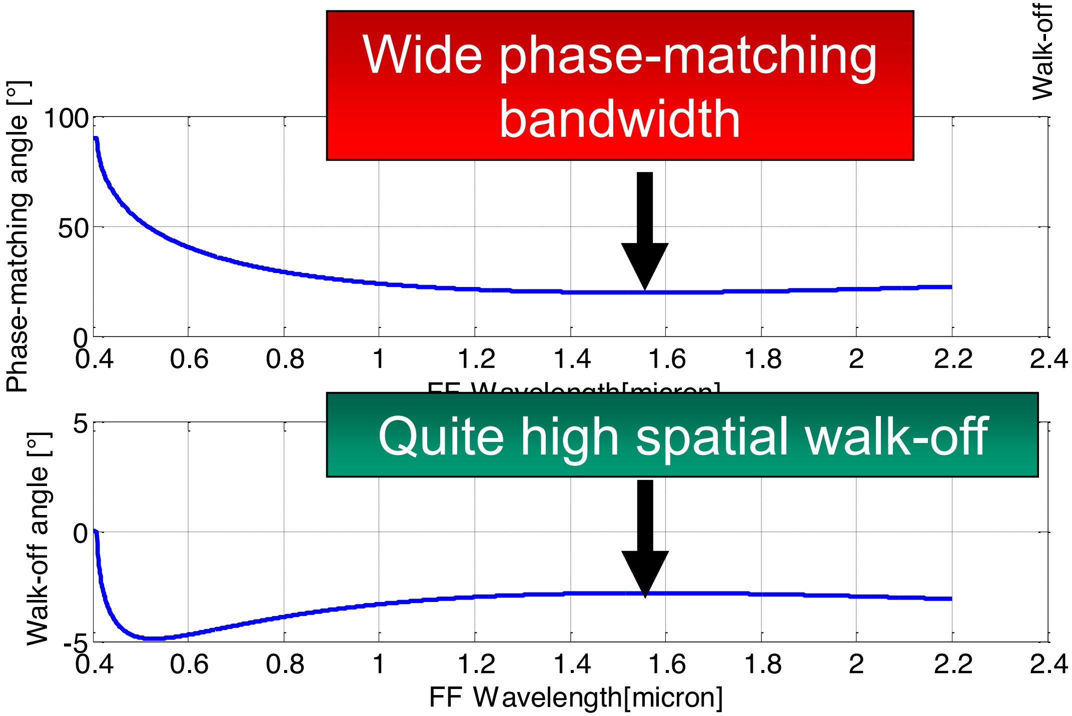
BBO

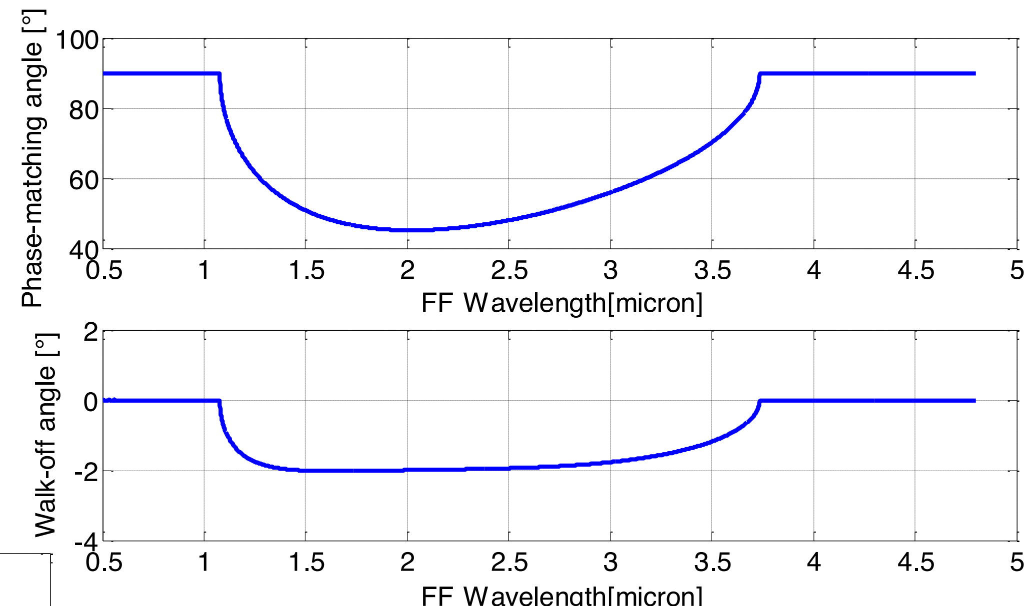
- negative uniaxial crystal ($n_e<n_o$)
- high-birifringence:

n_o = 1.672 @ 633 nm → FF ordinary

n_e = 1.549 @ 633 nm → SH extraord

d_{NL} ~2.3 pm/V rather LOW





$LiNbO_3$

- negative uniaxial crystal ($n_e<n_o$)
- small-birifringence:

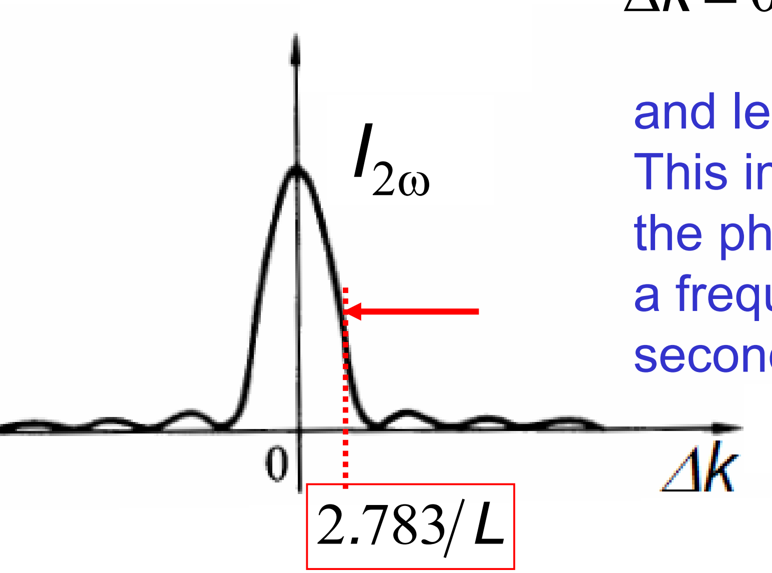
n_o = 2.283 @ 633 nm → FF ordinary

n_e = 2.203 @ 633 nm → SH extraord

d_{NL} ~4 pm/V LOW-MEDIUM

Phase matching bandwidth: calculation

$$I_{2\omega}(L) \propto sinc^2\left(\frac{\Delta k L}{2}\right)$$



- Let us assume phase-matching satisfied at a given fundamental frequency (FF) ω_0:

$$\Delta k = 0 \quad \rightarrow \quad k(2\omega_0) - 2k(\omega_0) = 0 \quad \rightarrow \quad n(2\omega_0) = n(\omega_0)$$

and let us determine the FWHM spectral width of the $I_{2\omega}$ curve. This implies evaluating Δk for a given frequency shift $\Delta\omega$ from the phase-matching frequency ω_0, while taking into account that a frequency shift at the fundamental frequency is doubled at the second harmonic:

$$\Delta k(\Delta\omega) = \left.\frac{dk}{d\omega}\right|_{2\omega_0} 2\Delta\omega - 2\left.\frac{dk}{d\omega}\right|_{\omega_0} \Delta\omega$$

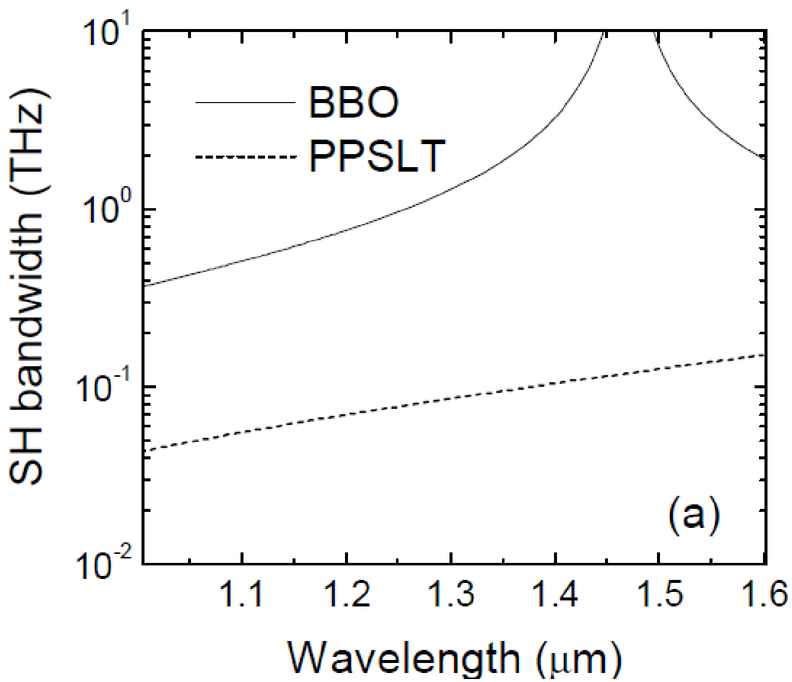
- The FWHM bandwidth at the second harmonic, $\Delta\omega_{SH} = 2\Delta\omega_{FF}$, becomes (see figure):

$$[k'(2\omega_0) - k'(\omega_0)]\frac{\Delta\omega_{SH}}{2} = \frac{2.783}{L} \quad \Rightarrow \quad \Delta\nu_{SH} = \frac{\Delta\omega_{SH}}{2\pi} = \frac{0{,}886}{[k'(2\omega_0) - k'(\omega_0)]L}$$

Phase matching bandwidth & dispersion

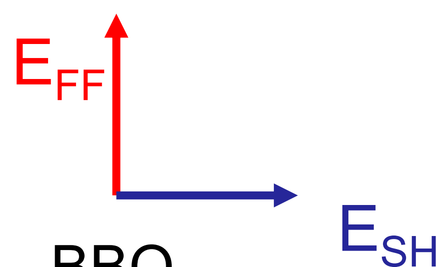
- Recalling that: $k(\omega) = \frac{\omega}{c} n(\omega);\qquad \frac{dn}{d\omega} = \frac{dn}{d\lambda}\frac{d\lambda}{d\omega};\qquad \frac{d\lambda}{d\omega} = -\frac{\lambda^2}{2\pi c}$
- one may easily refer the SH bandwidth to the crystal dispersion at FF & SH:

$$\Delta\nu_{SH} = \frac{0.886 \cdot c}{\left|1/2\, n'(\lambda_0/2) - n'(\lambda_0)\right| L\lambda_0}$$

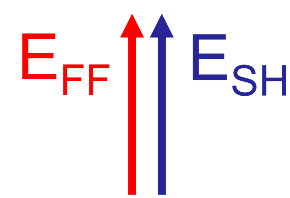


SHG in the visible range

L= 2.5 cm



BBO
Birifringence phase-matching



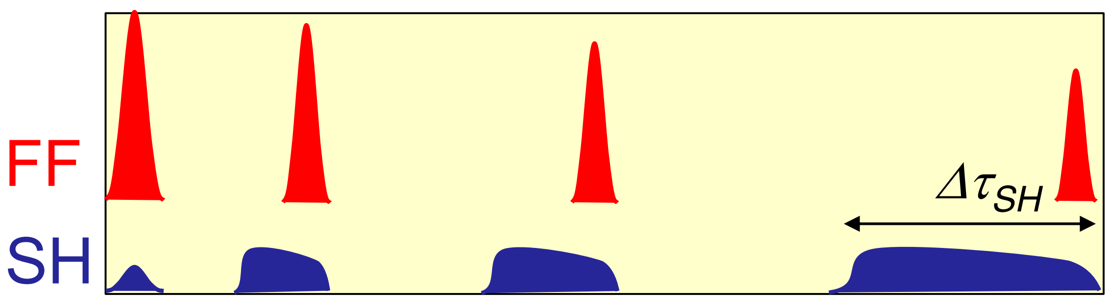
PPSLT
Quasi-phase-matching

Phase matching bandwidth: an insight

- Anticipating a result of the short-pulse regime, i.e. the fact that FF and SH pulses propagate at different “group” velocities given by:

$$v_{g,FF} = \frac{1}{k'(\nu_0)} \qquad v_{g,SH} = \frac{1}{k'(2\nu_0)}$$

- we may describe the SHG process between these two pulses as follows:



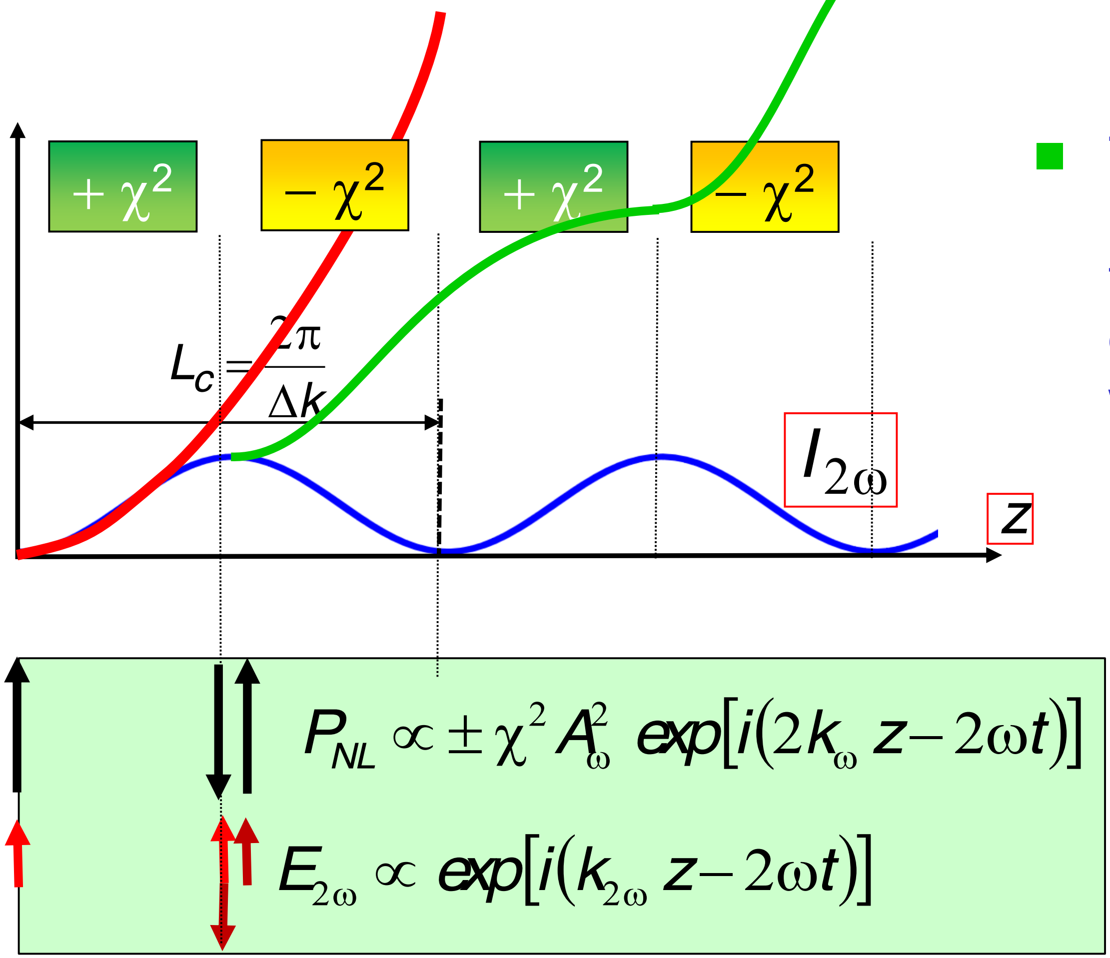
$$\Delta\tau_{SH} = \tau_{g,FF} - \tau_{g,SH} = GDM$$

GROUP DELAY MISMATCH

- According to Fourier theory we could figure out that:

$$\Delta\nu_{SH} \approx \frac{1}{\tau_{SH}} = \frac{1}{GDM} = \frac{1}{\left|\tau_{g,FF} - \tau_{g,SH}\right|} = \frac{1}{\left|\frac{L}{v_{g,FF}} - \frac{L}{v_{g,FF}}\right|} = \frac{1}{L\left|k'(\nu_0) - k'(2\nu_0)\right|}$$

Quasi-phase matching (QPM)



- It occurs in special crystals that exhibit a **periodic change of the sign of χ^2**, with a period:

$$\Lambda = mL_c$$

- This period allows a periodic re-phasing of the driving field (P_{NL}) with the generated SH field, resulting in a **quadratic dependence of $I_{2\omega}$ with L** with an effective nonlinear χ_2:

$$\chi_{2,eff} = \frac{2}{\pi m}\chi_2$$

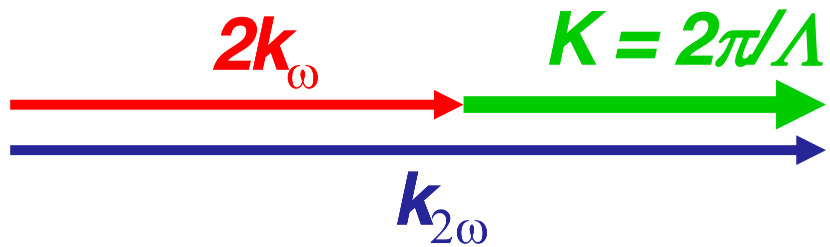
- The **quasi-phase-matching condition** is thus:

$$\frac{2\pi}{\Lambda} = k(2\omega_0) - 2k(\omega_0)$$

QPM: pros and cons

PROS

- Just need to change the poling period to adjust phase matching (the grating provides the momentum you need to get phase matching
- You may phase match fields with parallel polarization direction and exploit extremely high nonlinear coefficients
- Absence of any spatial walk-off because interacting fields may be set parallel to the crystal optical axis.



CONS

- Few crystals lend themselves to QPM since you need ferroelectric crystals (e.g. $LiNbO_3$, KTP, $LiTaO_3$) or semiconductors (GaAs)
- The fabrication procedure is rather complex for ferroelectrics – periodic poling needed – and very complex for semiconductors – orientation patterning
- Pretty hard to get phase-matching at short wavelengths due to the technological barrier of μm-level poling periods
- Optical damage at high fluence, especially for $LiNbO_3$.

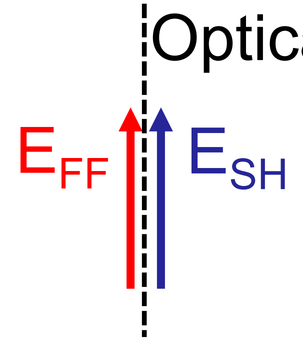
QPM: example

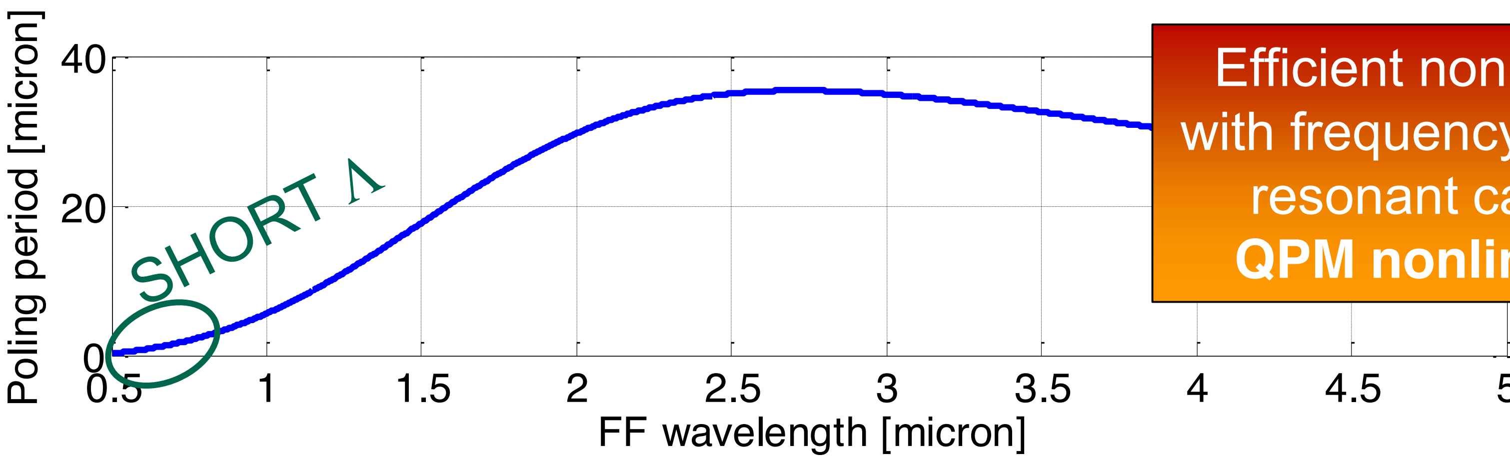
PPLN: periodically-poled lithium niobate

FF → extraordinary

SH → extraordinary

$d_{NL,eff}$ ~15-20 pm/V **HIGH**

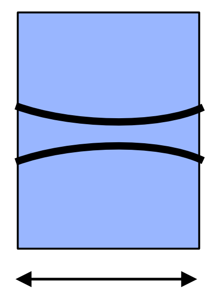




Efficient nonlinear processes with frequency combs & without resonant cavities need for **QPM nonlinear crystals !!**

Absence of spatial walk-off allows for confocal focusing: **interaction length only limited by diffraction !**

$$L = 2\frac{\pi w_0^2}{\lambda} n$$



L=2mm

PPLN

$w_{0,min}$ =15 μm

BBO

$w_{0,min}$ =120 μm

θ_{wo} = 3.5°

Phase matching in a parametric interaction

- $k_1 + k_2 = k_3$ or $\omega_1 n(\omega_1) + \omega_2 n(\omega_2) = \omega_3 n(\omega_3)$
- In a medium with normal dispersion $(dn/d\omega > 0)$

$$n(\omega_1) < n(\omega_2) < n(\omega_3) \quad \text{if} \quad \omega_1 < \omega_2 < \omega_3$$

- the phase matching condition can't be satisfied:

$$n(\omega_3) = \frac{n(\omega_1)\omega_1 + n(\omega_2)\omega_2}{\omega_3}$$

$$n(\omega_3) - n(\omega_2) = [n(\omega_1) - n(\omega_2)]\frac{\omega_1}{\omega_3}$$

- Types of possible birefringence phase matching:

	negative uniaxial $(n_e < n_o)$		positive uniaxial $(n_e > n_o)$	
TYPE I	$n_3^e\, \omega_3 = n_1^o\, \omega_1 + n_2^o\, \omega_2$	$(o+o \rightarrow e)$	$n_3^o\, \omega_3 = n_1^e\, \omega_1 + n_2^e\, \omega_2$	$(e+e \rightarrow o)$
TYPE II	$n_3^e\, \omega_3 = n_1^e\, \omega_1 + n_2^o\, \omega_2$	$(e+o \rightarrow e)$	$n_3^o\, \omega_3 = n_1^e\, \omega_1 + n_2^o\, \omega_2$	$(e+o \rightarrow e)$
	$n_3^e\, \omega_3 = n_1^o\, \omega_1 + n_2^e\, \omega_2$	$(o+e \rightarrow e)$	$n_3^o\, \omega_3 = n_1^o\, \omega_1 + n_2^e\, \omega_2$	$(o+e \rightarrow e)$

Example: type I phase matching

Birifringence phase-matching in a negative uniaxial crystal

- The phase matching condition is $n_{e3}(\theta_m)\,\omega_3 = n_{o1}\,\omega_1 + n_{o2}\,\omega_2$

giving $$n_{e3}(\theta_m) = \frac{n_{o1}\omega_1 + n_{o2}\omega_2}{\omega_3}$$

- In a uniaxial crystal, the extraordinary index for propagation along θ is

$$n_e^2(\theta) = \frac{n_o^2\, n_e^2}{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta}$$

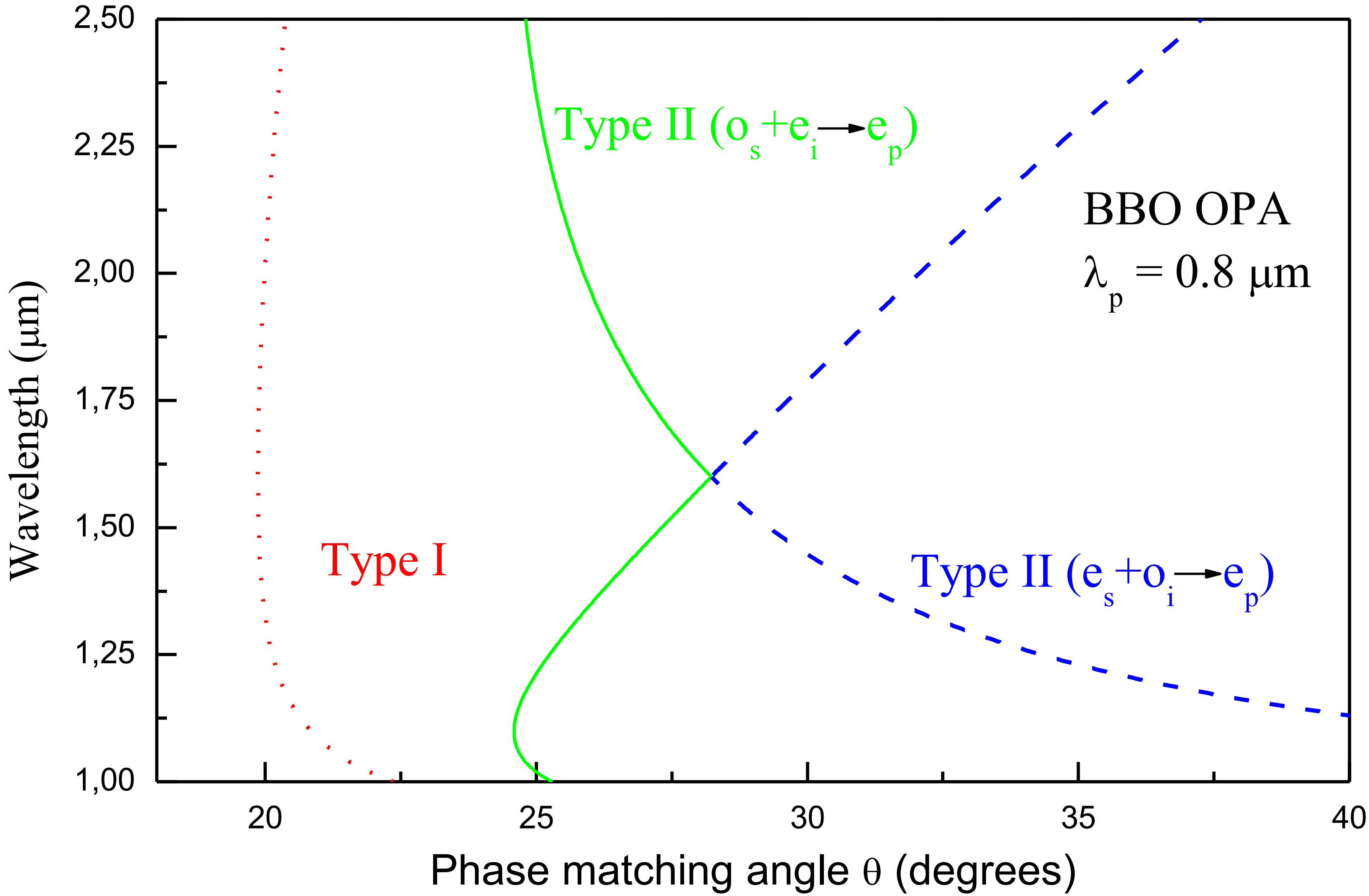
which gives $$\sin\theta_m = \frac{n_{e3}}{n_{e3}(\theta_m)} \sqrt{\frac{n_{o3}^2 - n_{e3}^2(\theta_m)}{n_{o3}^2 - n_{e3}^2}}$$

…the refractive indexes at each wavelength being obtained by Sellmeier equations

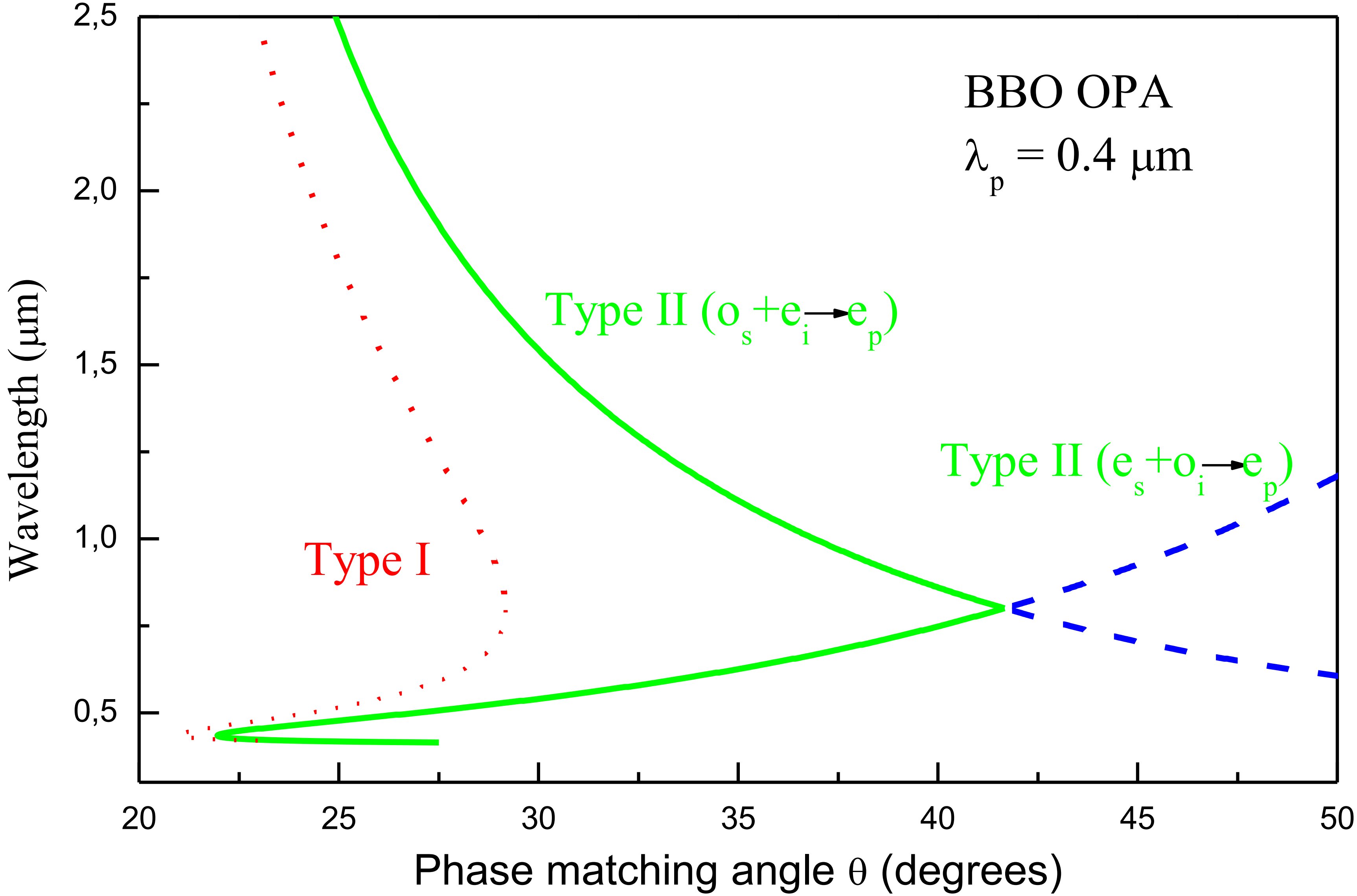
QPM in a periodically-poled crystal

$$k_1 + k_2 + \frac{2\pi}{\Lambda} = k_3 \quad \Rightarrow \quad \frac{\omega_1}{c} n_1 + \frac{\omega_2}{c} n_2 + \frac{2\pi}{\Lambda} = \frac{\omega_3}{c} n_3$$

Phase matching curves of a near-IR OPA



Phase matching curves of a visible OPA



The equations of linear pulse propagation

The polarization as a driving term

Starting from Maxwell's equations

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

the polarization on the r.h.s. acts as a driving term.
The electric field is a plane wave

$$E(z,t) = A(z,t)\exp[i(\omega_0 t - k_0 z)]$$

The polarization can be decomposed in linear and nonlinear parts:

$$P(z,t) = P_L(z,t) + P_{NL}(z,t)$$

we consider only the linear component:

$$P_L(z,t) = p_L(z,t)\exp[i(\omega_0 t - k_0 z)]$$

Switching to the Fourier domain

By introducing the Fourier transform

$$\tilde{E}(z,\omega) = \Im[E(z,t)] = \int_{-\infty}^{+\infty} E(z,t)\exp(-i\omega t)\,dt$$

we get:

$$\tilde{E}(z,\omega) = \tilde{A}(z,\omega-\omega_0)\exp(-ik_0 z)$$

$$\tilde{P}_L(z,\omega) = \tilde{p}_L(z,\omega-\omega_0)\exp(-ik_0 z)$$

Recalling the derivative rule for the Fourier transform:

$$\Im\left[\frac{d^n F(t)}{dt^n}\right] = (i\omega)^n \tilde{F}(\omega)$$

we obtain:

$$\frac{\partial^2 \tilde{E}}{\partial z^2} + \frac{\omega^2}{c_0^2}\tilde{E} = -\mu_0 \omega^2 \tilde{P}_L$$

The slowly varying envelope approximation

We express the second derivative as:

$$\frac{\partial^2 \widetilde{E}}{\partial z^2} = \left(\frac{\partial^2 \widetilde{A}}{\partial z^2} - 2ik_0 \frac{\partial \widetilde{A}}{\partial z} - k_0^2 \widetilde{A} \right) \exp(-ik_0 z)$$

We assume:

$$\frac{\partial^2 \widetilde{A}}{\partial z^2} << k_0 \frac{\partial \widetilde{A}}{\partial z}$$

The **Slowly Varying Envelope Approximation (SVEA)** neglects variations of the envelope over propagation of the order of wavelength.

With this assumption we obtain:

$$-2ik_0 \frac{\partial \widetilde{A}}{\partial z} - k_0^2 \widetilde{A} + \frac{\omega^2}{c_0^2} \widetilde{A} = -\mu_0 \omega^2 \widetilde{p}_L$$

The frequency-dependent polarization

For a monochromatic wave:

$$\tilde{P}_L(\omega) = \varepsilon_0 \chi^{(1)}(\omega) E(\omega)$$

recalling that:

$$n_L(\omega) = \sqrt{\left(1 + \chi^{(1)}(\omega)\right)}$$

We obtain:

$$-2ik_0 \frac{\partial \tilde{A}}{\partial z} - k_0^2 \tilde{A} + \frac{\omega^2}{c_0^2} \tilde{A} = -\frac{\omega^2}{c_0^2}\left[n_L^2(\omega) - 1\right]\tilde{A}$$

which simplifies to:

$$2ik_0 \frac{\partial \tilde{A}}{\partial z} = \left[k^2(\omega) - k_0^2\right]\tilde{A}$$

Propagation in a dispersive medium (I)

Starting from the propagation equation:

$$2ik_0 \frac{\partial \tilde{A}}{\partial z} = \left[k^2(\omega) - k_0^2\right]\tilde{A}$$

We expand k(ω) in a Taylor series around the carrier frequency ω_0 :

$$k(\omega) = k_0 + \left(\frac{dk}{d\omega}\right)_{\omega_0}(\omega - \omega_0) + \frac{1}{2}\left(\frac{d^2k}{d\omega^2}\right)_{\omega_0}(\omega - \omega_0)^2 + \frac{1}{6}\left(\frac{d^3k}{d\omega^3}\right)_{\omega_0}(\omega - \omega_0)^3 + \ldots$$

An expansion up to the third order (or to the second order for moderate pulse bandwidths) is sufficient. By approximating:

$$k^2(\omega) - k_0^2 = \left[k(\omega) - k_0\right]\left[k(\omega) + k_0\right] \cong 2k_0\left[k(\omega) - k_0\right]$$

we obtain:

$$i\frac{\partial \tilde{A}(\omega - \omega_0)}{\partial z} \cong k'_0(\omega - \omega_0)\tilde{A} + \frac{1}{2}k''_0(\omega - \omega_0)^2\tilde{A} + \frac{1}{6}k'''_0(\omega - \omega_0)^3\tilde{A}$$

Propagation in a dispersive medium (II)

$$i\frac{\partial \tilde{A}(\omega-\omega_0)}{\partial z} \cong k'_0(\omega-\omega_0)\tilde{A} + \frac{1}{2}k''_0(\omega-\omega_0)^2\tilde{A} + \frac{1}{6}k'''_0(\omega-\omega_0)^3\tilde{A}$$

$$k'_0 = \left(\frac{dk}{d\omega}\right)_{\omega_0} = \frac{1}{v_{g0}}$$

where v_{g0} is the **group velocity** of the carrier frequency

$$k''_0 = \left(\frac{d^2k}{d\omega^2}\right)_{\omega_0} = GVD$$

is known as **Group Velocity Dispersion (GVD)**

Propagation in a dispersive medium (III)

We now Fourier transform back to the time domain. Recalling the derivative rule:

$$\Im^{-1}\left[\omega^n \tilde{F}(\omega)\right] = (-i)^n \frac{d^n F(t)}{dt^n}$$

we obtain:

$$\frac{\partial A(z,t)}{\partial z} + \frac{1}{v_{g0}}\frac{\partial A}{\partial t} - \frac{i}{2}k''_0 \frac{\partial^2 A}{\partial t^2} + \frac{1}{6}k'''_0 \frac{\partial^3 A}{\partial t^3} = 0$$

Which, neglecting third order dispersion ($k'''_0 \cong 0$) becomes:

$$\frac{\partial A(z,t)}{\partial z} + \frac{1}{v_{g0}}\frac{\partial A}{\partial t} - \frac{i}{2}k''_0 \frac{\partial^2 A}{\partial t^2} = 0$$

The **parabolic equation** captures the main physics of linear propagation of ultrashort pulses in dispersive media.

In the absence of dispersion

The original equation takes the form:

$$\frac{\partial A(z,t)}{\partial z}+\frac{1}{v_{g0}}\frac{\partial A(z,t)}{\partial t}=0$$

Let us set it in a new reference-frame moving at v_{g0}, with space/time variables:

$$z'=z; \qquad t'=t-\frac{z}{v_{g0}}$$

By transformation of derivatives in the new reference frame :

$$\frac{\partial A}{\partial z}=\frac{\partial A}{\partial z'}\frac{\partial z'}{\partial z}+\frac{\partial A}{\partial t'}\frac{\partial t'}{\partial z}=\frac{\partial A}{\partial z'}-\frac{1}{v_{g0}}\frac{\partial A}{\partial t'}$$

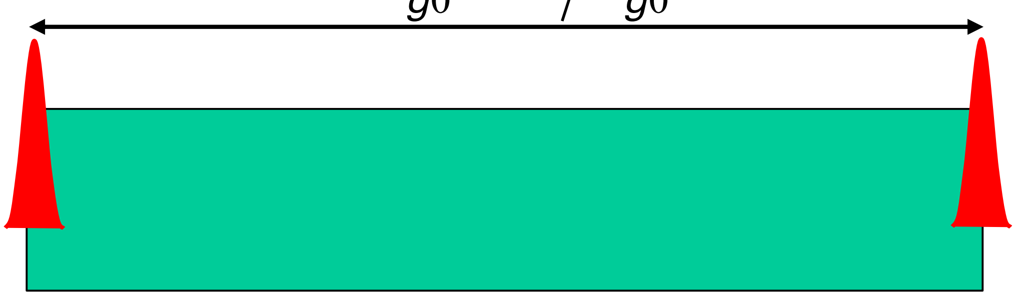
$$\frac{\partial A}{\partial t}=\frac{\partial A}{\partial t'}\frac{\partial t'}{\partial t}+\frac{\partial A}{\partial z'}\frac{\partial z'}{\partial t}=\frac{\partial A}{\partial t'}$$

one gets:

$$\frac{\partial A}{\partial z'}-\frac{1}{v_{g0}}\frac{\partial A}{\partial t'}+\frac{1}{v_{g0}}\frac{\partial A}{\partial t'}=0$$

$$\frac{\partial A(z',t')}{\partial z'}=0$$

$$\tau_{g0}=L/v_{g0}$$



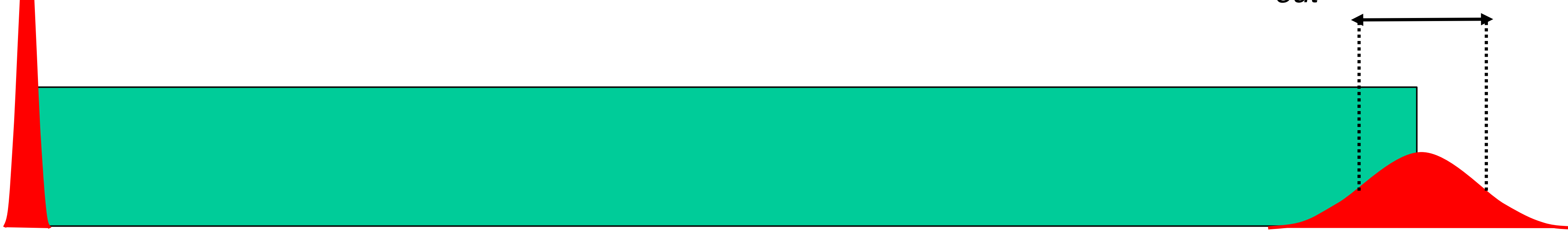
The pulse envelope propagates without distortion at a speed v_{g0} taking a time τ_{g0} to cross the crystal

In the presence of dispersion

The pulse gets more and more broadened while propagating, with a pulse broadening per unit bandwidth given by the GDD (group-delay-dispersion) parameter (expressed in fs^2) :

$$GDD = \frac{\partial \tau_{g0}}{\partial \omega} = \frac{\partial}{\partial \omega}\left(\frac{L}{v_{g0}}\right) = \frac{\partial}{\partial \omega}\left(Lk_0'\right) = Lk_0'' = L \cdot GVD$$

If the dispersion-induced pulse broadening is far in excess of the input pulse duration, at the crystal output one has: $\tau_{out} = GDD \cdot B$



where B is the angular-frequency bandwidth B

The equations of nonlinear pulse propagation

Propagation in a nonlinear medium (I)

We start from the equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_L}{\partial t^2} + \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$$

where:

$$P_{NL}(z,t) = p_{NL}(z,t)\exp\left[i\left(\omega_0 t - k_p z\right)\right]$$

We emphasize that the wavenumber $\mathbf{k_p}$ of the nonlinear polarization at ω_0 is **different** from that of the electric field $\mathbf{k_0}$. We express:

$$\frac{\partial^2 P_{NL}}{\partial t^2} = \left(\cancel{\frac{\partial^2 p_{NL}}{\partial t^2}} + \cancel{2i\omega_0 \frac{\partial p_{NL}}{\partial t}} - \omega_0^2 p_{NL} \right) \exp\left[i\left(\omega_0 t - k_p z\right)\right]$$

assuming that the envelope p_{NL} varies slowly over the timescale of an optical cycle:

$$\frac{\partial^2 p_{NL}}{\partial t^2}, \omega_0 \frac{\partial p_{NL}}{\partial t} << \omega_0^2 p_{NL}$$

Propagation in a nonlinear medium (II)

From the equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_L}{\partial t^2} - \mu_0 \omega_0^2 p_{NL} \exp\left[i\left(\omega_0 t - k_p z\right)\right]$$

By the same procedure applied to the linear propagation equation, we obtain:

$$-2ik_0 \frac{\partial A}{\partial z} - 2\frac{ik_0}{v_{g0}}\frac{\partial A}{\partial t} - k_0 k''_0 \frac{\partial^2 A}{\partial t^2} = -\mu_0 \omega_0^2 p_{NL} \exp\left[-i\Delta k z\right]$$

which can be rewritten as:

$$\frac{\partial A}{\partial z} + \frac{1}{v_{g0}}\frac{\partial A}{\partial t} - \frac{i}{2}k''_0 \frac{\partial^2 A}{\partial t^2} = -i\frac{\mu_0 \omega_0 c}{2n_0} p_{NL} \exp\left[-i\Delta k z\right]$$

where **$\Delta k = k_p - k_0$** is the "**wave-vector mismatch**" between the nonlinear polarization and the field

The nonlinear polarization in second-order parametric interaction (I)

Consider the superposition of three waves at frequencies ω_1, ω_2 and ω_3 with $\omega_1 + \omega_2 = \omega_3$

$$E(z,t) = \frac{1}{2}\left\{ \begin{array}{l} A_1(z,t)\exp[i(\omega_1 t - k_1 z)] + A_2(z,t)\exp[i(\omega_2 t - k_2 z)] + \\ A_3(z,t)\exp[i(\omega_3 t - k_3 z)] + c.c. \end{array} \right\}$$

impinging on a medium with a second order nonlinear response:

$$P_{NL}(z,t) = \varepsilon_0 \chi^{(2)} E^2(z,t)$$

The nonlinear polarization has components at several frequencies, such as $2\omega_1$, $2\omega_2$ etc. We assume that the **phase-matching condition** selects only the interaction between the three fields at ω_1, ω_2 and ω_3 to be efficient.

The nonlinear polarization in second-order parametric interaction (II)

We derive the following terms:

$$P_{1NL}(z,t)=\frac{\varepsilon_0\chi^{(2)}}{2}A_2^*A_3\exp\{i[(\omega_3-\omega_2)t-(k_3-k_2)z]+c.c.\}$$

$$P_{2NL}(z,t)=\frac{\varepsilon_0\chi^{(2)}}{2}A_1^*A_3\exp\{i[(\omega_3-\omega_1)t-(k_3-k_1)z]+c.c.\}$$

$$P_{3NL}(z,t)=\frac{\varepsilon_0\chi^{(2)}}{2}A_1A_2\exp\{i[(\omega_1+\omega_2)t-(k_1+k_2)z]+c.c.\}$$

Which we plug into the nonlinear propagation equations:

$$\frac{\partial A}{\partial z}+\frac{1}{v_{g0}}\frac{\partial A}{\partial t}-\frac{i}{2}k''_0\frac{\partial^2 A}{\partial t^2}=-i\frac{\mu_0\omega_0 c}{2n_0}p_{NL}\exp[-i\Delta kz]$$

The nonlinear coupled propagation equations (I)

thus deriving the three coupled equations:

$$\frac{\partial A_1}{\partial z} + \frac{1}{v_{g1}}\frac{\partial A_1}{\partial t} - \frac{i}{2}k''_1\frac{\partial^2 A_1}{\partial t^2} = -i\frac{\mu_0\varepsilon_0 c\omega_1}{2n_1} d_{eff} A_2^* A_3 \exp\left[-i\left(k_3 - k_2 - k_1\right)z\right]$$

$$\frac{\partial A_2}{\partial z} + \frac{1}{v_{g2}}\frac{\partial A_2}{\partial t} - \frac{i}{2}k''_2\frac{\partial^2 A_2}{\partial t^2} = -i\frac{\mu_0\varepsilon_0 c\omega_2}{2n_2} d_{eff} A_1^* A_3 \exp\left[-i\left(k_3 - k_1 - k_2\right)z\right]$$

$$\frac{\partial A_3}{\partial z} + \frac{1}{v_{g3}}\frac{\partial A_3}{\partial t} - \frac{i}{2}k''_3\frac{\partial^2 A_3}{\partial t^2} = -i\frac{\mu_0\varepsilon_0 c\omega_3}{2n_3} d_{eff} A_1 A_2 \exp\left[-i\left(k_1 + k_2 - k_3\right)z\right]$$

with $d_{eff} = \frac{\chi^{(2)}}{2}$ $\Delta k = k_3 - k_1 - k_2$

These are **coupled nonlinear partial differential equations** which are in general not amenable to an analytic solution and must be treated numerically.

The nonlinear coupled propagation equations (II)

As a first simplification we neglect the GVD terms. This is justified by considering that the three interacting pulses are propagating at very different group velocities v_{gi}. The effects of this group velocity mismatch are more relevant than those of GVD between the different frequency components of a single pulse.

$$\frac{\partial A_1}{\partial z} + \frac{1}{v_{g1}} \frac{\partial A_1}{\partial t} = -i\kappa_1 A_2^* A_3 \exp[-i\Delta k z]$$

$$\frac{\partial A_2}{\partial z} + \frac{1}{v_{g2}} \frac{\partial A_2}{\partial t} = -i\kappa_2 A_1^* A_3 \exp[-i\Delta k z]$$

$$\frac{\partial A_3}{\partial z} + \frac{1}{v_{g3}} \frac{\partial A_3}{\partial t} = -i\kappa_3 A_1 A_2 \exp[i\Delta k z]$$

where the nonlinear coupling constants are defined as: $\kappa_i = \frac{\omega_i d_{eff}}{2cn_i}$

The nonlinear coupled propagation equations (III)

By moving to a frame of reference translating with the group velocity of the pump pulse: $t' = t - \frac{z}{v_{g3}}$.

$$\frac{\partial A_1}{\partial z} + \delta_{13}\frac{\partial A_1}{\partial t} = -i\kappa_1 A_2^* A_3 \exp[-i\Delta k z]$$

$$\frac{\partial A_2}{\partial z} + \delta_{23}\frac{\partial A_2}{\partial t} = -i\kappa_2 A_1^* A_3 \exp[-i\Delta k z]$$

$$\frac{\partial A_3}{\partial z} = -i\kappa_3 A_1 A_2 \exp[i\Delta k z]$$

where

$$\delta_{i3} = \frac{1}{v_{gi}} - \frac{1}{v_{g3}} \qquad i = 1,2$$

is the Group Velocity Mismatch (**GVM**) between signal/idler and pump waves, typically expressed in **ps/mm**. It gives the **group delay accumulated by the two pulses per unit length**.

Phase matching bandwidth in OPA/DFG

It may be estimated from the results obtained in the cw regime under the high gain approximation:

$$G = \frac{1}{4}\exp(2gL) \qquad g = \sqrt{\gamma^2 - \left(\frac{\Delta k}{2}\right)^2}$$

$$\Delta k = k_p - k_s - k_i$$ with $\Delta k = 0$ for a given (ω_p ω_s ω_i) set

For a given fixed pump frequency ω_p, if the signal frequency ω_s increases to $\omega_s+\Delta\omega$, by energy conservation the idler frequency decreases to $\omega_i-\Delta\omega$. The wave vector may thus be written as:

$$\Delta k = -\frac{\partial k_s}{\partial \omega}\Delta\omega + \frac{\partial k_i}{\partial \omega}\Delta\omega = \left(\frac{1}{v_{gs}} - \frac{1}{v_{gi}}\right)\Delta\omega$$

Introducing Δk in the expression for the gain G and looking for a solution at 50% of the maximum gain, one gets a FWHM bandwidth:

$$\Delta\nu \cong \frac{2(\ln 2)^{1/2}}{\pi}\left(\frac{\gamma}{L}\right)^{1/2}\frac{1}{\left|\frac{1}{v_{gs}} - \frac{1}{v_{gi}}\right|} \propto \left(\frac{\gamma}{L}\right)^{1/2}\frac{1}{\delta_{si}}$$

High gain bandwidth demands for group-velocity matching between signal and idler

Few general rules for ultrashort-pulse interactions

OPA/DFG
REGIME

PUMP	(ω_p, ω_3)	$A_p(0) \neq 0$
SIGNAL	(ω_s, ω_1)	$A_s(0) \neq 0$
IDLER	(ω_i, ω_2)	$A_i(0) = 0$

- Input pump duration > input signal duration
- Interaction length limited by temporal walk-off
- Length of the crystal primarily chosen as a function of δ_{ps}
- Signal delayed from the pump
- Exponential gain only as long as the three pulses remain superimposed
- Pulse distortion without temporal overlap
- High gain for $v_{gi} < v_{gp} < v_{gs}$
- Low δ_{si} for broadband amplification

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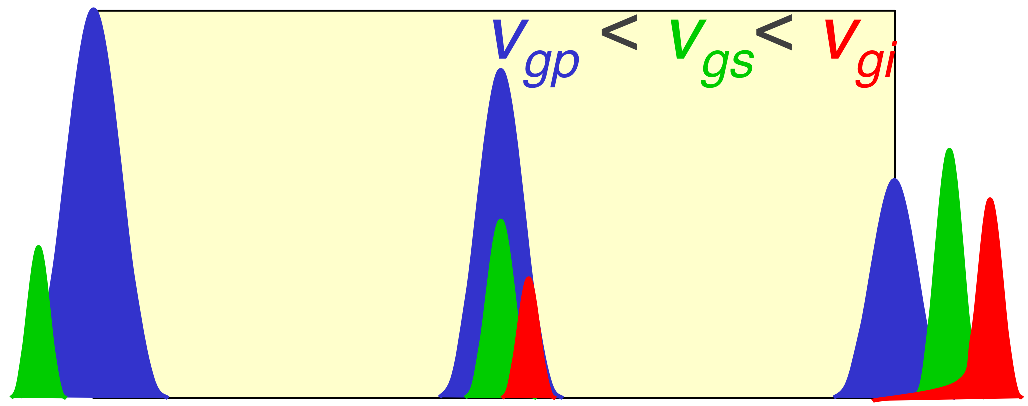
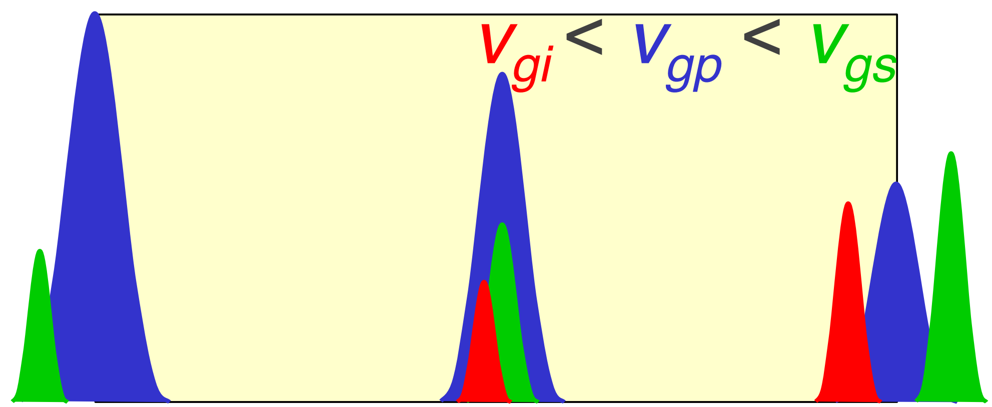




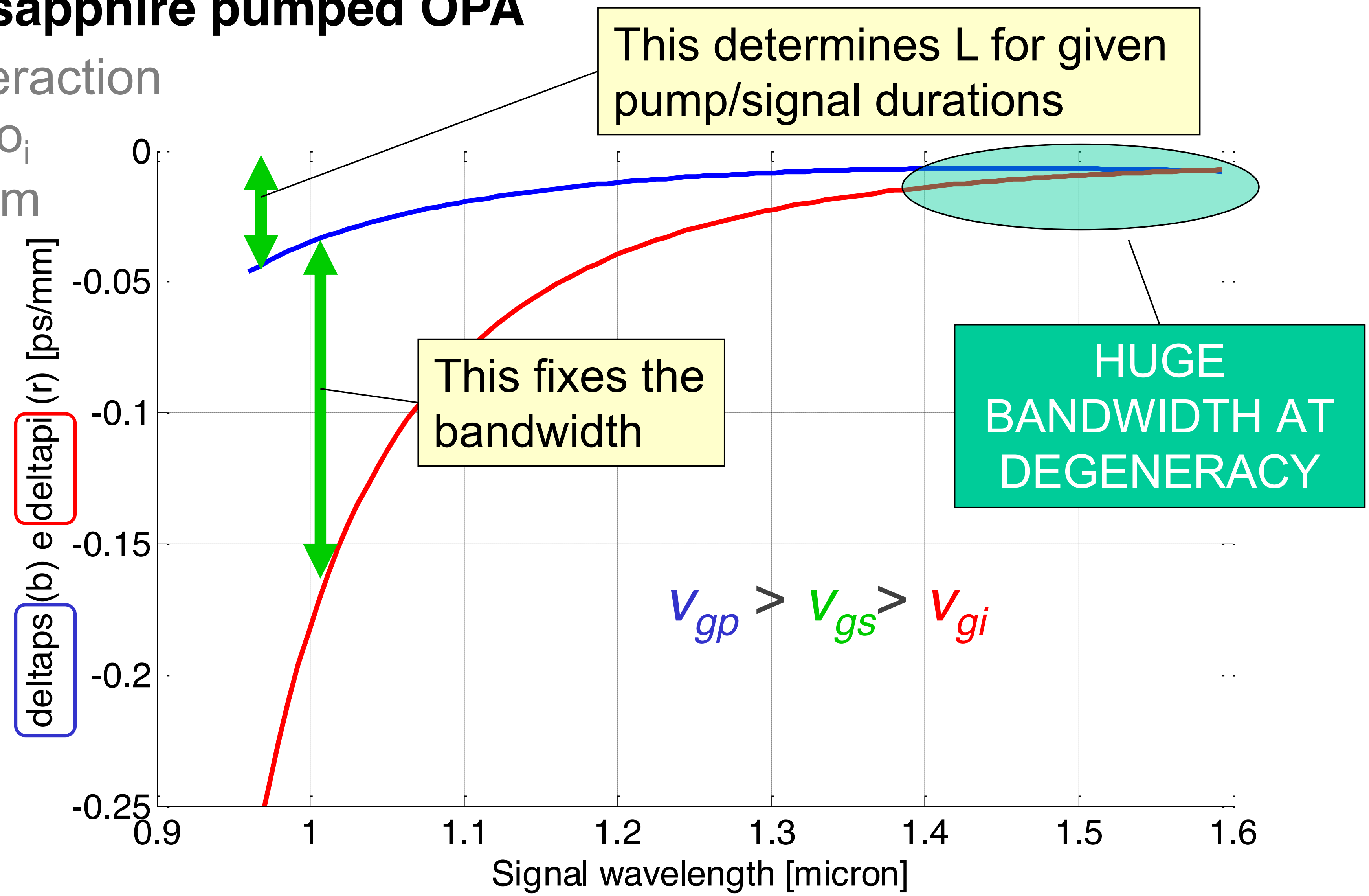
The starting point: the GVM curves (I)

BBO: Ti-sapphire pumped OPA

Type I interaction

$e_p \rightarrow o_s + o_i$

$\lambda_p = 0.8\ \mu m$



The starting point: the GVM curves (II)

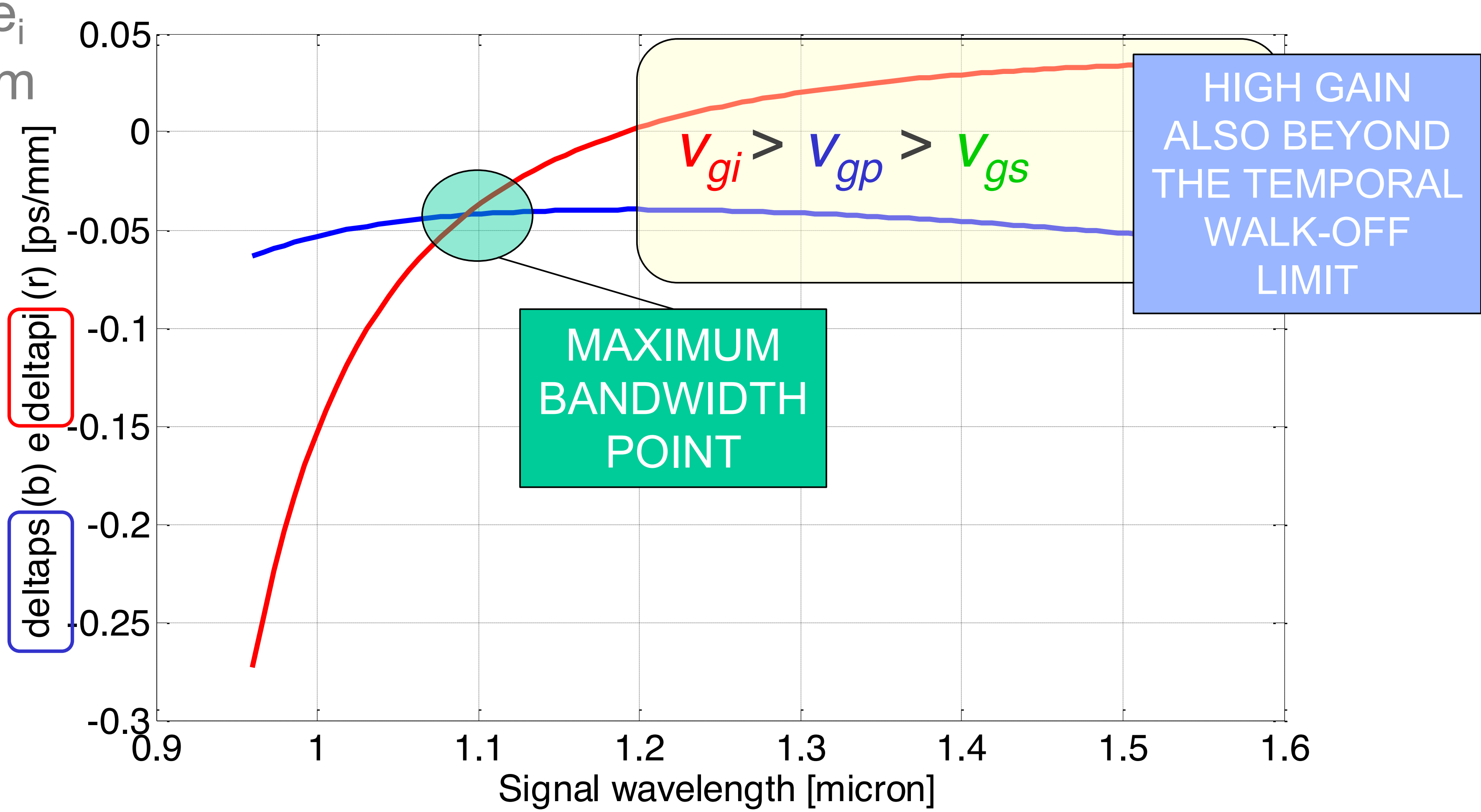
BBO: Ti-sapphire pumped OPA

Type II interaction

$e_p \rightarrow o_s + e_i$

$\lambda_p = 0.8\ \mu m$

δ_{ps} ~ 50 fs/mm over the whole tuning range



Generating a frequency comb above 5 μm (I)

GaSe: Er:fiber pumped DFG

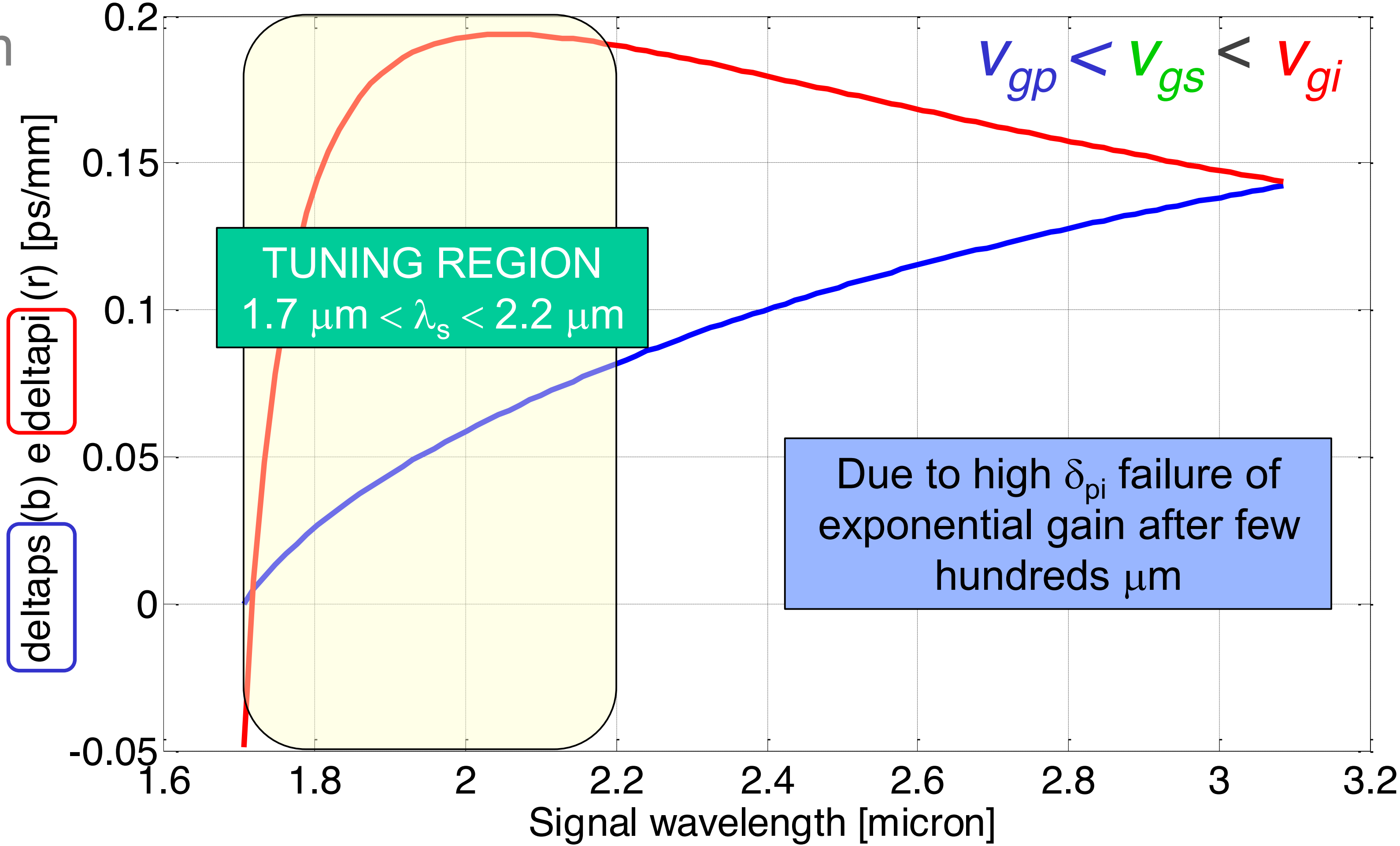
Type I interaction

$e_p \rightarrow o_s + o_i$

$\lambda_p = 1.55\ \mu m$

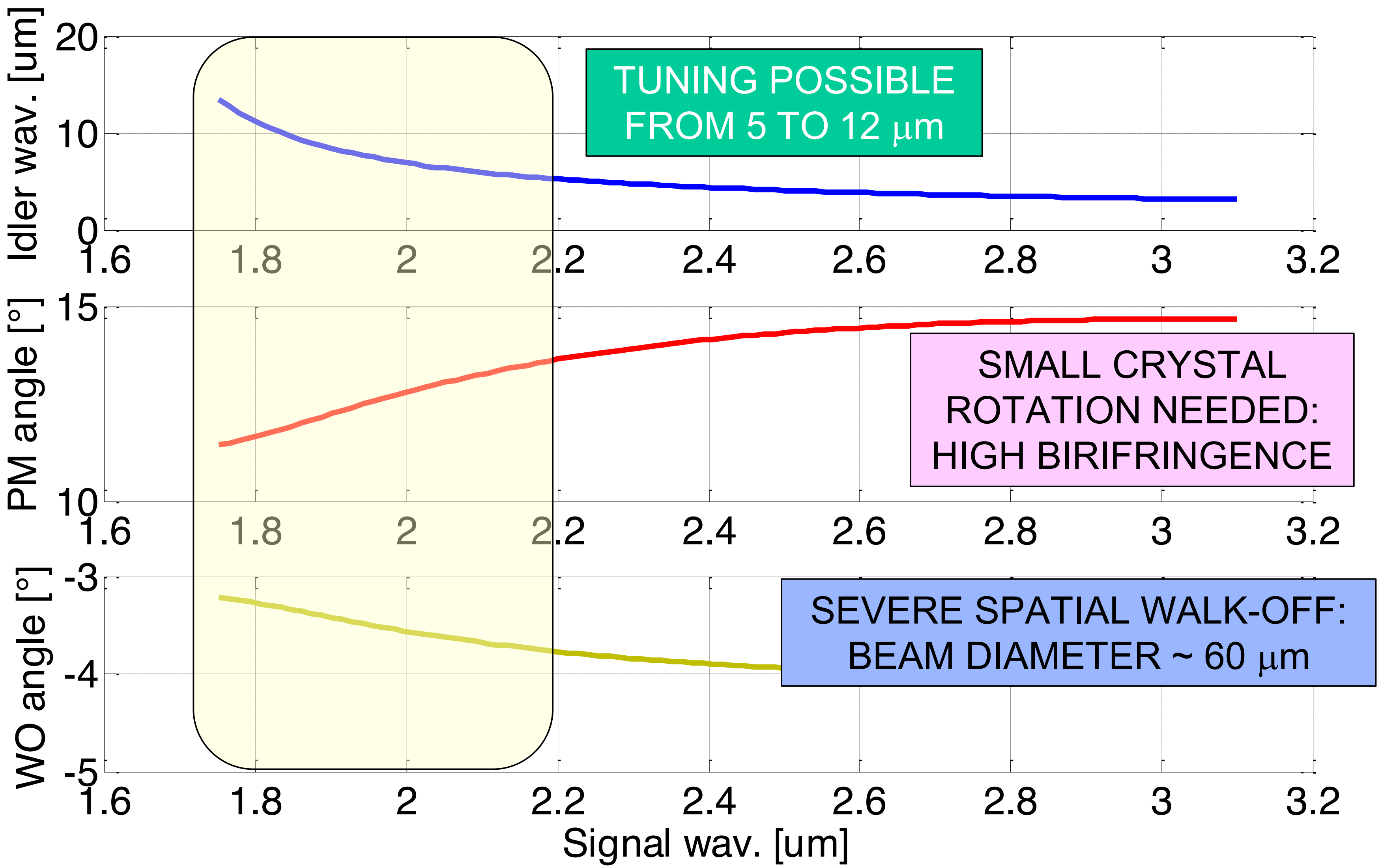
$\tau_p = 70\ fs$

$\delta_{ps} < 80$ fs/mm over the whole tuning range $\rightarrow L = 1$ mm



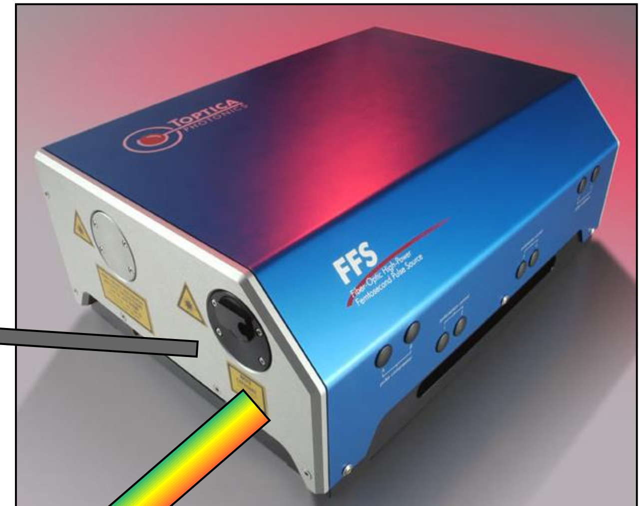
Generating a frequency comb above 5 μm (II)

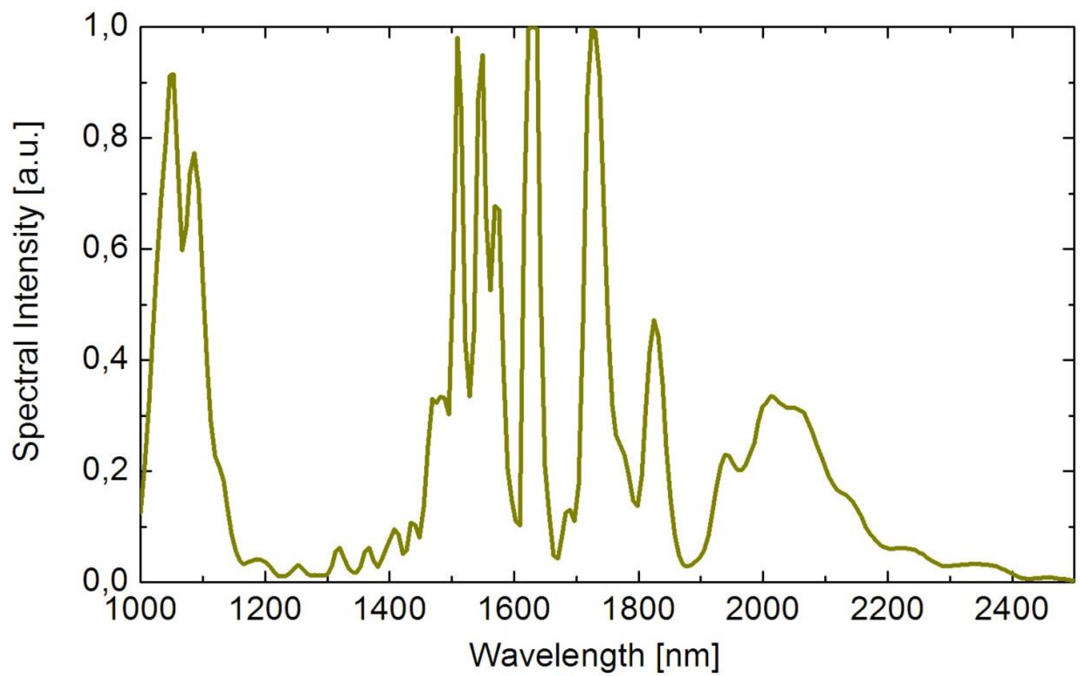
GaSe: Er:fiber pumped DFG



The first frequency comb above 5 μm (I)

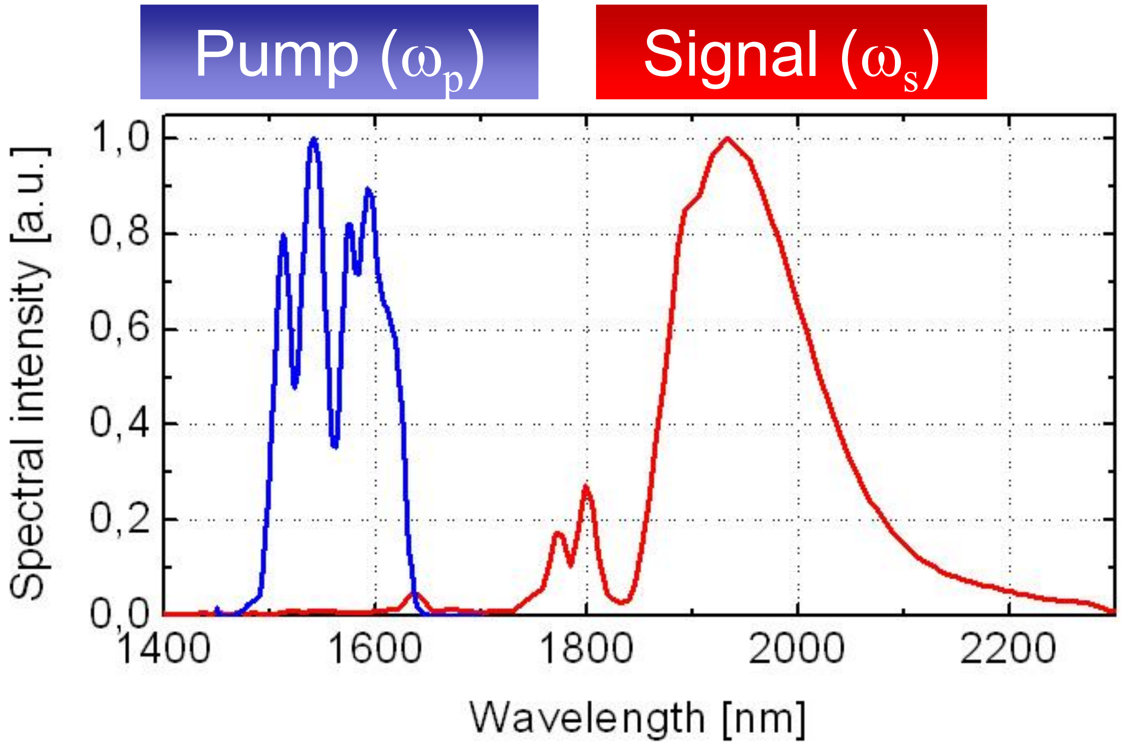
- $f_R \approx 100$ MHz
- $\lambda_p = 1.55$ μm
- $<P> = 250$ mW
- $\tau = 65$ fs





- $\lambda_s = 1\text{-}2.2$ μm
- $<P> = 160$ mW

The first frequency comb above 5 μm (II)

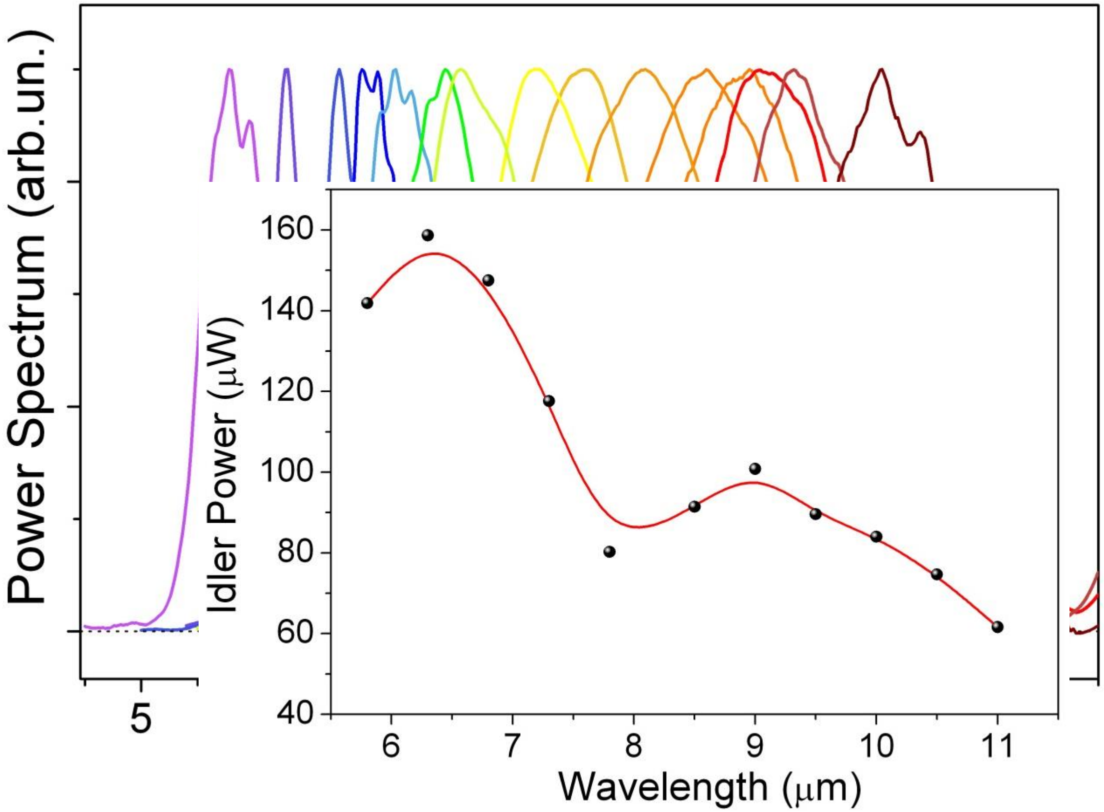


mid-IR radiation generated as idler beam

GaSe

- extremely broad tunability: 5 -16 μm
- f_{ceo}-free comb synthesis
- absence of 2-photons absorption

The first frequency comb above 5 μm (III)



A. Gambetta et al, Opt. Lett. ***33***, 2671 (2008)

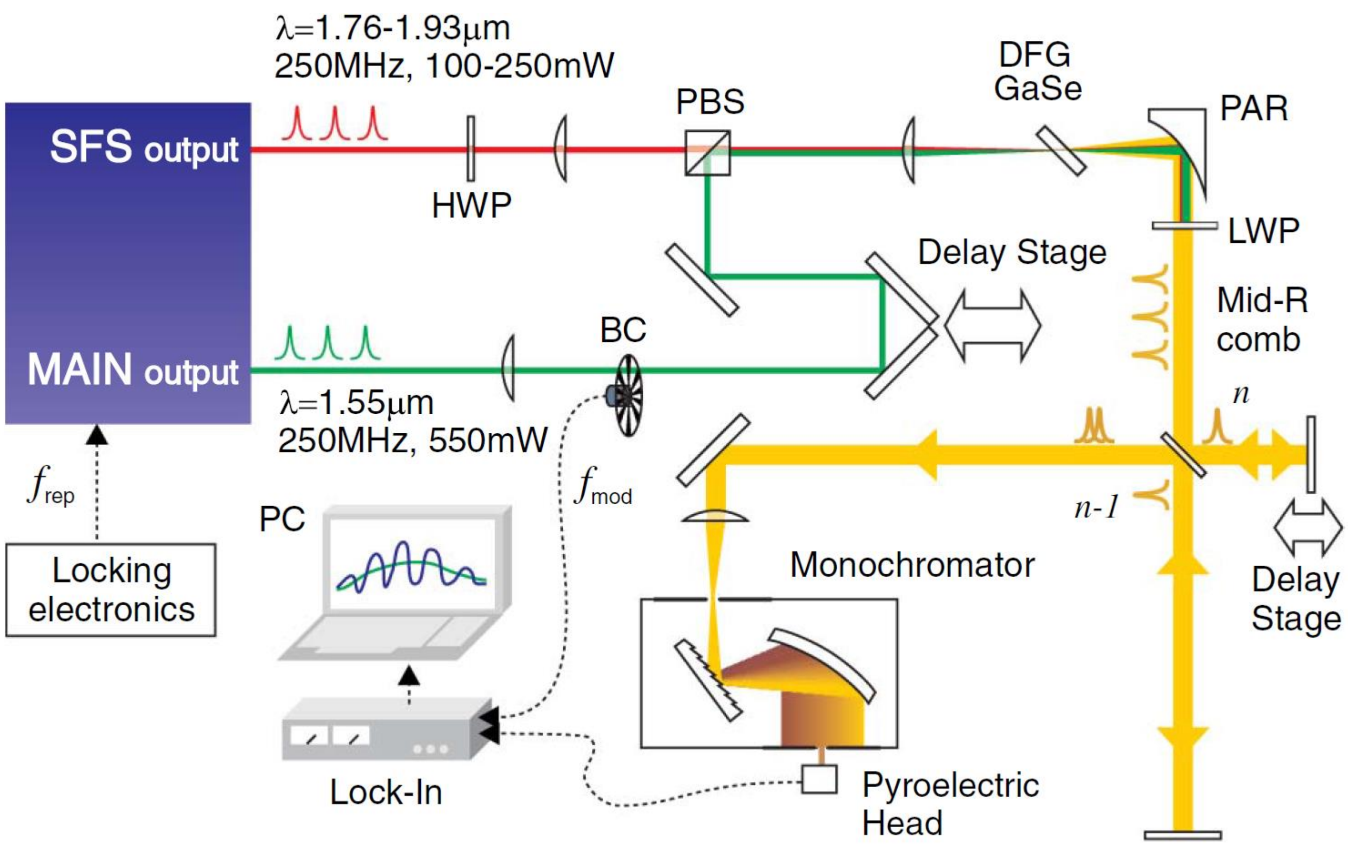
Tunability through:

- angle tuning
- chirp tuning

Comb mode power: ~ 1-2 nW

Spectrum limited to $\lambda > 5$ μm

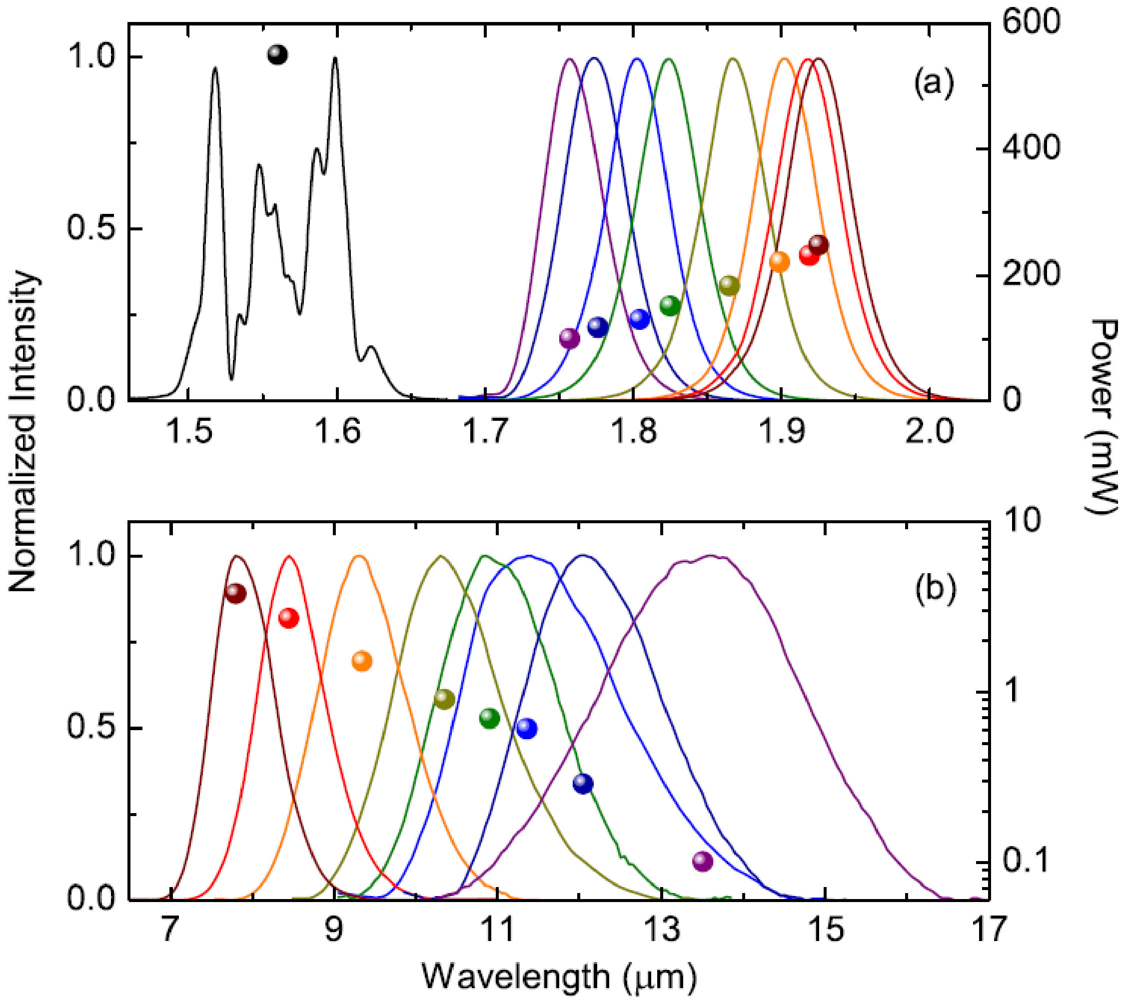
A more recent experiment with a more powerful Er:fiber oscillator



Menlo Systems @ 250 MHz

Raman fiber for signal pulse generation

Second experiment: results



GaSe

Tunability through:

- angle tuning
- power tuning

Spectrum limited to λ > 7 µm

Comb mode power: ~ 100-200 nW

A. Gambetta et al, Opt. Lett. 38 1155 (2013)