Ultrafast Laser Physics

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Chapter 4b: $\chi^{(2)}$-nonlinearities with ultrashort pulses

Contents

- Second order nonlinear susceptibility and susceptibility tensor
- Overview of $\chi^{(2)}$ processes
- Crystal optics, tensors and effective nonlinearities
- Phase matching – with ultrashort pulses and realistic beams
- Quasi-phase matching
- Second harmonic generation and sum frequency mixing
- Difference frequency mixing and parametric amplification
- Ultrabroadband phase matching schemes
- Optical parametric chirped-pulse amplification
- Optical parametric oscillators
- Cascading of $\chi^{(2)}$ -nonlinearities

Wave equation in nonlinear optics

- Wave equation for homogenous, dielectric material:

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

- Linear optics:

$$P_i = \epsilon_0 \chi_{ij} E_j$$

- Nonlinear optics:

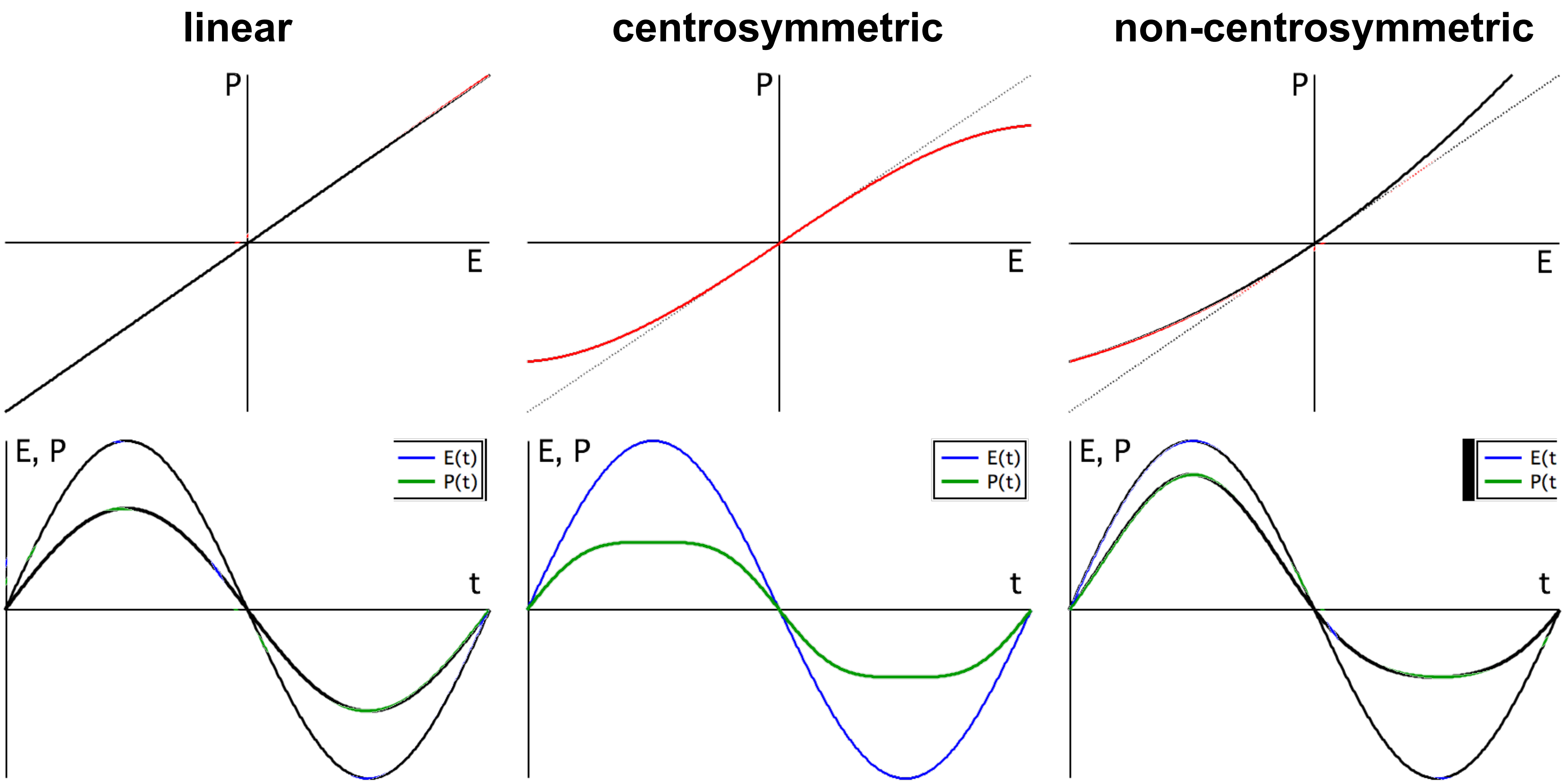
$$P_i = \epsilon_0 \chi_{ij}^{(1)} E_j + \epsilon_0 \chi_{ijk}^{(2)} E_j E_k + \epsilon_0 \chi_{ijkl}^{(3)} E_j E_k E_l + ...$$

2nd order nonlinearities

3rd order nonlinearities

- Examples of 2nd order nonlinear processes:
  - Second harmonic generation (SHG), sum-frequency generation (SFG)
  - Difference-frequency generation (DFG), optical rectification
- Examples of 3rd order nonlinear processes:
  - Third-harmonic generation (THG)
  - Optical Kerr effect, self-phase modulation, self-focusing

Why are third-order effects more common?



- Third-order effects can occur in all materials, even isotropic systems (e.g., glass, air)
- Second-order nonlinearities occur only in non-centrosymmetric materials (ferroelectric materials)

A 1-dimensional example

- Interaction of two monochromatic waves

$$P_{NL} = \epsilon_0 \chi^{(2)} E^2 \qquad\qquad E = E_1 + E_2$$

$$E_1 = E_{10} \cos\left(\omega_1 t - k_1 z\right)$$

$$E_2 = E_{20} \cos\left(\omega_2 t - k_2 z\right)$$

- Interaction at fixed position (z = 0)

$$P_{NL} = \epsilon_0 \chi^{(2)} \left(E_{10} \cos\left(\omega_1 t\right) + E_{20} \cos\left(\omega_2 t\right)\right)^2$$

$P_{NL} =$ **Opt. rectification**

$+$ **Second harmonic 1** $+$ **Second harmonic 2**

$+$ **Sum frequency generation**

$+$ **Difference frequency generation**

Nonlinear optical susceptibility tensor

- Nonlinear optical polarization in 3D materials (crystals):

$$P_i^{NL} = \epsilon_0 d_{ijk} E_j E_k$$

- Product of E-vectors is symmetric in *j* and *k*, therefore

$$d_{ijk} = d_{ikj}$$

- This allows reduction of 27-element tensor to 18-element matrix:

$$d_{ijk} \rightarrow d_{im} \quad (i = 1, 2, 3 \quad m = 1...6)$$

j	k	m
1	1	1
2	2	2
3	3	3
2 3	3 2	4
1 3	3 1	5
1 2	2 1	6

$$\begin{pmatrix} P_x^{NL} \\ P_y^{NL} \\ P_z^{NL} \end{pmatrix} = \epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \cdot \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_zE_x \\ 2E_xE_y \end{pmatrix}$$

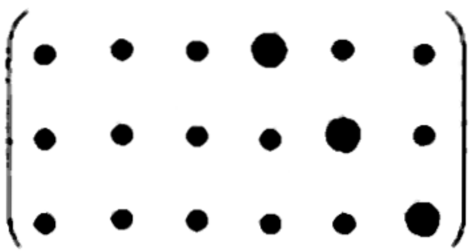
Further reduction through crystal symmetries

- Material tensors have to be invariant against the symmetry operations of the point group of a crystal
- This allows to further reduce the number of independent tensor elements

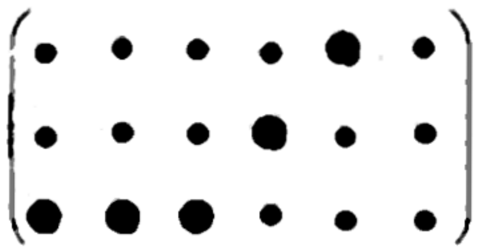
zero modulus
non-zero modulus
equal moduli
moduli numerically equal, but opposite in sign
a modulus equal to minus 2 times the heavy dot modulus to which it is joined

Orthorhombic

Class 222



Class mm2

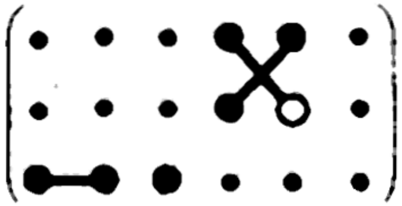


- For orthorhombic point groups: only 3 and 5 non-zero independent matrix elements

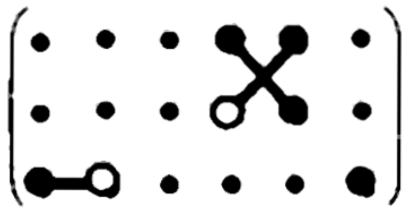
One more example: tetragonal point groups

- zero modulus
- non-zero modulus
- equal moduli
- moduli numerically equal, but opposite in sign
- a modulus equal to minus 2 times the heavy dot modulus to which it is joined

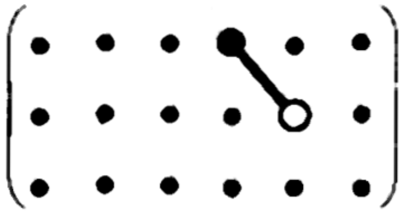
Class 4



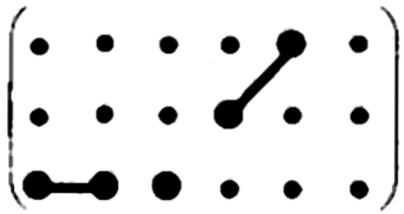
Class $\bar{4}$



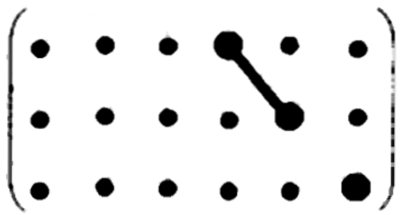
Class 422



Class 4mm



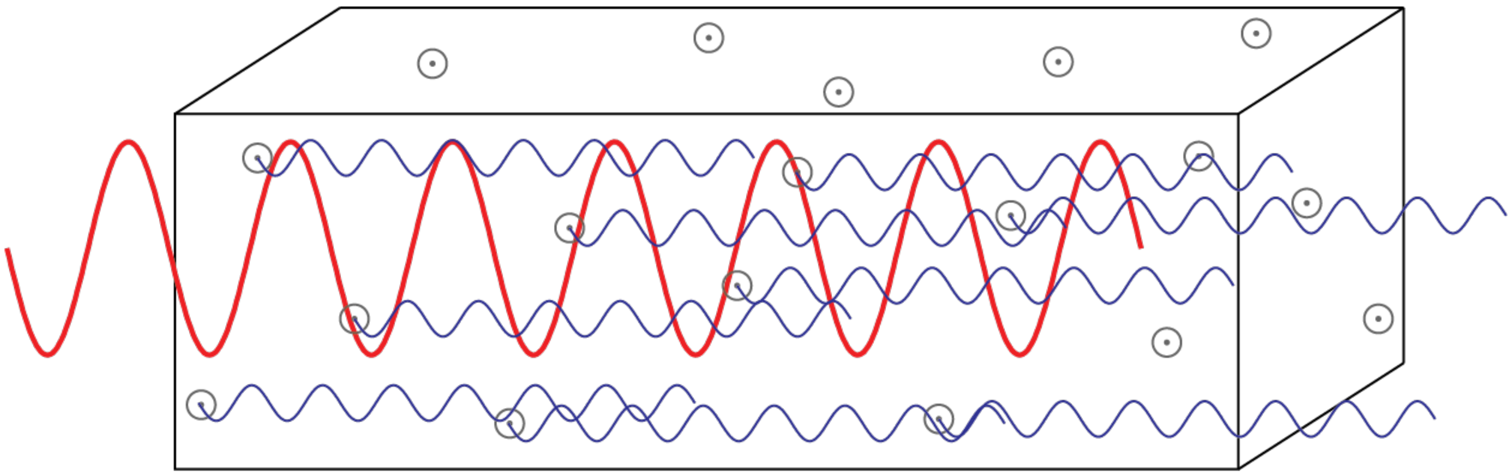
Class $\bar{4}2m$ ($2 \| x_1$)



Mixing processes to be observed macroscopically

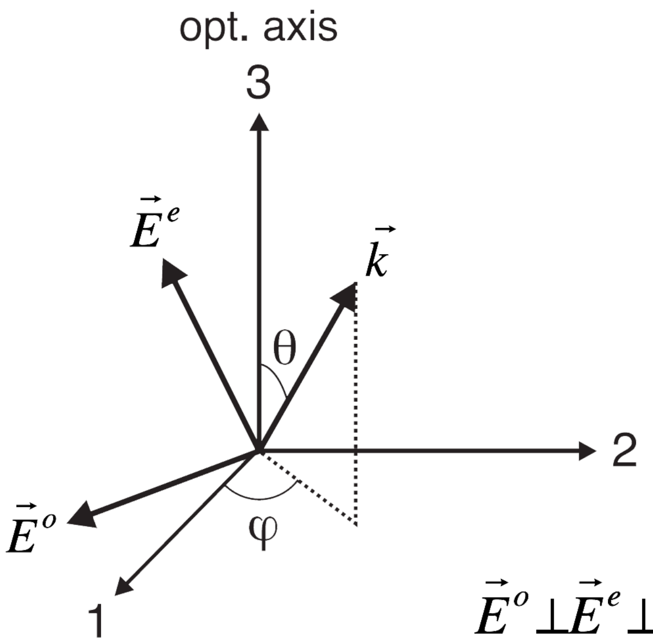
$$\begin{pmatrix} P_x^{NL} \\ P_y^{NL} \\ P_z^{NL} \end{pmatrix} = \epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \cdot \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_zE_x \\ 2E_xE_y \end{pmatrix}$$

- Microscopically, all wave-mixing processes mediated by non-zero tensor elements occur
- Macroscopically, mixing products can only build up significant power if the corresponding process is phase-matched (i.e., microscopic contributions add up constructively)



Back to 1D-problem

- For collinear beams, with one interaction selected (e.g., through phase-matching) and beam geometry fixed (polarization and propagation direction), the **nonlinear susceptibility can be projected down to a scalar effective nonlinearity d_{eff}**
- Assumptions: no dispersion, no absorption, uniaxial crystal

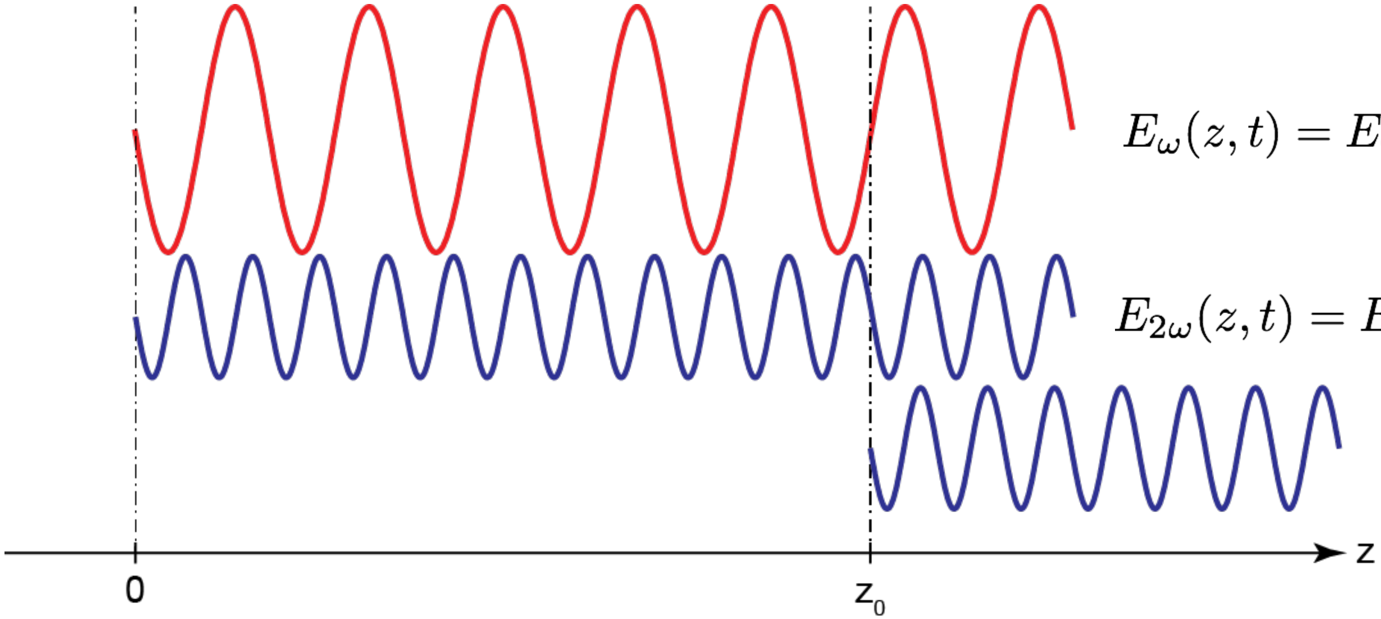


$\vec{E}^o \perp \vec{E}^e \perp \vec{k}$

Crystal symm.	2 e-rays and 1 o-ray	2 o-rays and 1 e-ray
6, 4	0	$d_{15} \sin\theta$
622, 422	0	0
6mm, 4mm	0	$d_{15} \sin\theta$
$\bar{6}$m2	$d_{22} \cos^2\theta \cos 3\varphi$	$-d_{22} \cos\theta \sin 3\varphi$
3m	$d_{22} \cos^2\theta \cos 3\varphi$	$d_{15} \sin\theta - d_{22} \cos\theta \sin 3\varphi$
$\bar{6}$	$\cos^2\theta\,(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi)$	$\cos\theta\,(d_{11} \cos 3\varphi - d_{22} \sin 3\varphi)$
3	$\cos^2\theta\,(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi)$	$d_{15} \sin\theta + \cos\theta\,(d_{11} \cos 3\varphi - d_{22} \sin 3\varphi)$
32	$d_{11} \cos^2\theta \sin 3\varphi$	$d_{11} \cos\theta \cos 3\varphi$
$\bar{4}$	$\sin 2\theta\,(d_{14} \cos 2\varphi - d_{15} \sin 2\varphi)$	$-\sin\theta\,(d_{14} \sin 2\varphi + d_{15} \cos 2\varphi)$
$\bar{4}$2m	$d_{14}\,(\sin 2\theta \cos 2\varphi)$	$-d_{14} \sin\theta \sin 2\varphi$

Phase matching

- Example: second-harmonic generation of collinear plane waves



- Second harmonic has to interfere constructively for all z_0 in crystal
- For fixed t = 0:

  Second harmonic light generated at z = 0, propagated to z_0:

$$E_{2\omega}(z_0) = E_{2\omega,0} e^{ik_{2\omega} z_0}$$

  Second harmonic light generated at z = z_0:

$$E'_{2\omega}(z_0) \propto E_\omega(z_0)^2$$

- For constructive interference for all z_0, phases of existing and newly generated light have to match

Phase matching

- Second harmonic has to interfere constructively for all z_0 in crystal
- For fixed t = 0:

  Second harmonic light generated at z = 0, propagated to z_0:

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  Second harmonic light generated at z = z_0:

  $$E'_{2\omega}(z_0) \propto E_{\omega}(z_0)^2$$

- For constructive interference for all z_0, phases of existing and newly generated light have to match

$$\varphi_{2\omega}(z_0) \stackrel{!}{=} \varphi'_{2\omega}(z_0)$$

$$k_{2\omega} z_0 \stackrel{!}{=} 2 k_{\omega} z_0$$

$$\frac{2\pi}{\lambda/2} n_{2\omega} \stackrel{!}{=} 2 \frac{2\pi}{\lambda} n_{\omega}$$

$$\boxed{n_{2\omega} \stackrel{!}{=} n_{\omega}}$$

- This is in general not possible because of dispersion!

Birefringent phase matching

- This condition can be met at some wavelengths in suitable birefringent materials

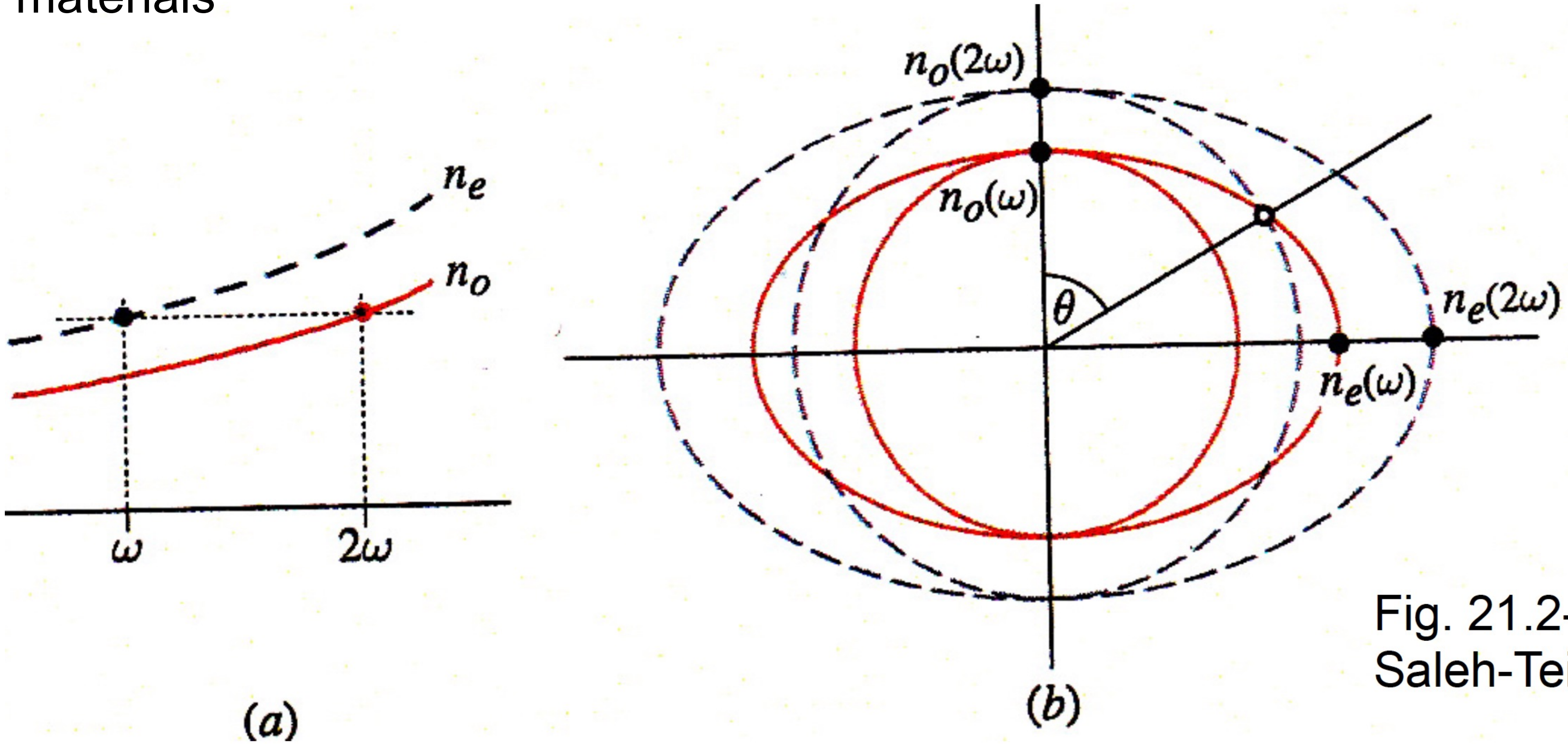


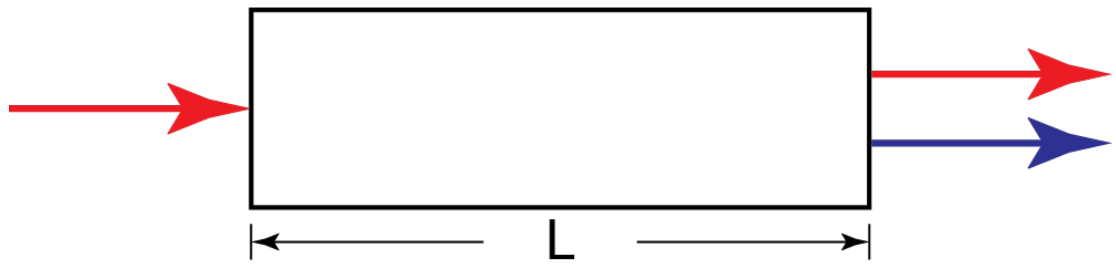
Fig. 21.2-10
Saleh-Teich

- In this example, two fundamental photons polarized along extraordinary axis generate one second-harmonic photon along ordinary axis
- However, this condition is typically met exactly at only one set of interacting wavelengths

Phase mismatch and conversion efficiency (SHG)

- Coupled-wave equations (1D, collinear case, non-absorbing medium)

$$\frac{\partial E_\omega}{\partial z} = -i\omega \frac{1}{cn_\omega} d_{\text{eff}} E_{2\omega} E_\omega^* e^{-i\Delta kz}$$
$$\frac{\partial E_{2\omega}}{\partial z} = -i\omega \frac{1}{cn_{2\omega}} d_{\text{eff}} E_\omega^2 e^{i\Delta kz}$$



- Phase mismatch:

$$\Delta k := k_3 - k_1 - k_2 = k_{2\omega} - 2k_\omega$$

- Efficiency without depletion of fundamental, $I_{2\omega}(z=0) = 0,\ I_\omega(z) = \text{const}$

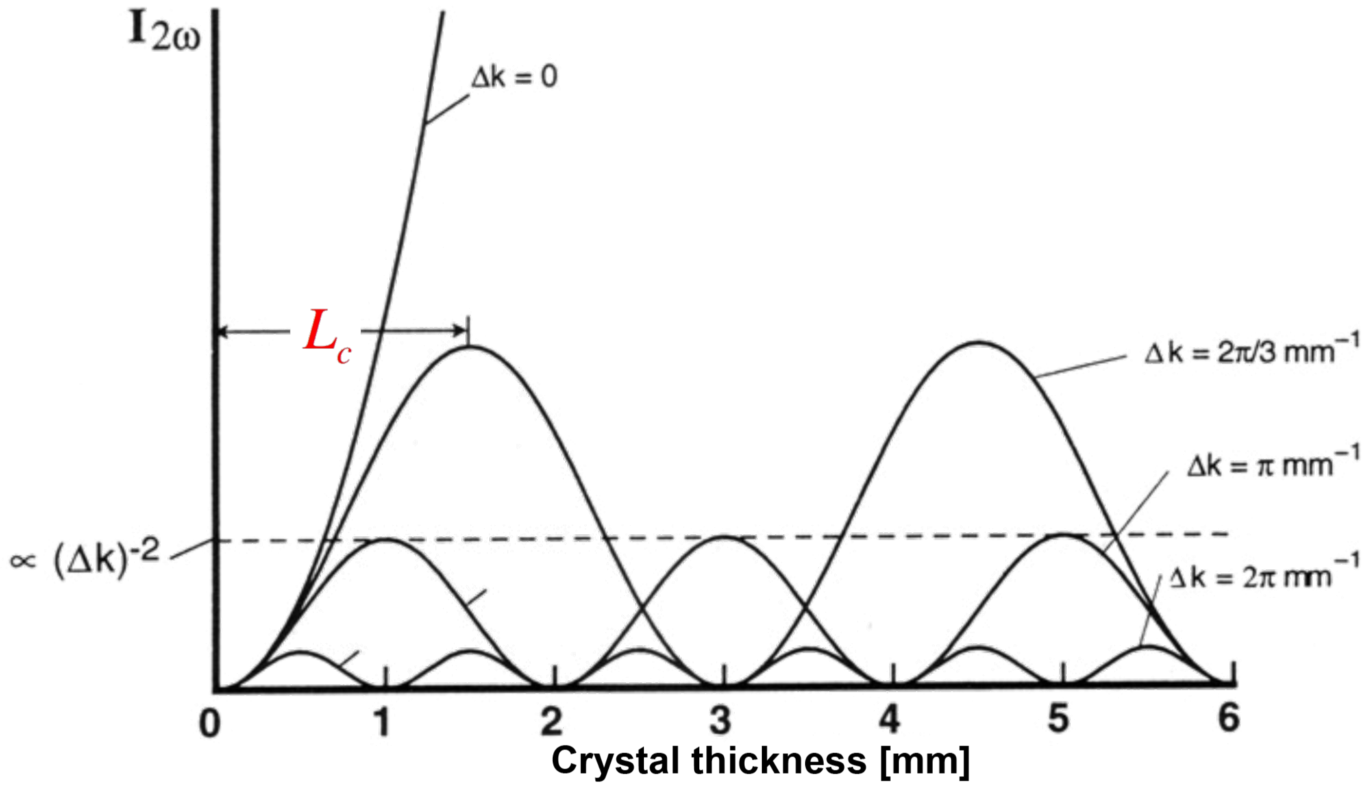
⇒ Integrate coupled wave equation for second harmonic along crystal length L

$$E_{2\omega}(L) = -i\omega \frac{1}{cn_{2\omega}} d_{\text{eff}} E_\omega^2 \frac{e^{i\Delta kL} - 1}{i\Delta kL} = -i\omega \frac{1}{cn_{2\omega}} d_{\text{eff}} E_\omega^2 \frac{\sin\left(\frac{\Delta kL}{2}\right)}{\frac{\Delta kL}{2}}$$

- With $I = \frac{1}{2}\epsilon_0 cnE^2$ it follows

$$\eta = \frac{I_{2\omega}}{I_\omega} = \frac{2\omega^2}{\epsilon_0 c^3}\left(\frac{d_{\text{eff}}^2}{n_{2\omega} n_\omega^2}\right) I_\omega L^2 \text{sinc}^2\left(\frac{\Delta kL}{2}\right) \propto L^2 \text{sinc}^2\left(\frac{\Delta kL}{2}\right)$$

Phase mismatch and coherence length



- Signal grows quadratically for vanishing phase mismatch
- For non-zero mismatch, signal oscillates periodically
- First local maximum reached after so-called coherence length L_c

$$L_c := \frac{\pi}{\Delta k}$$

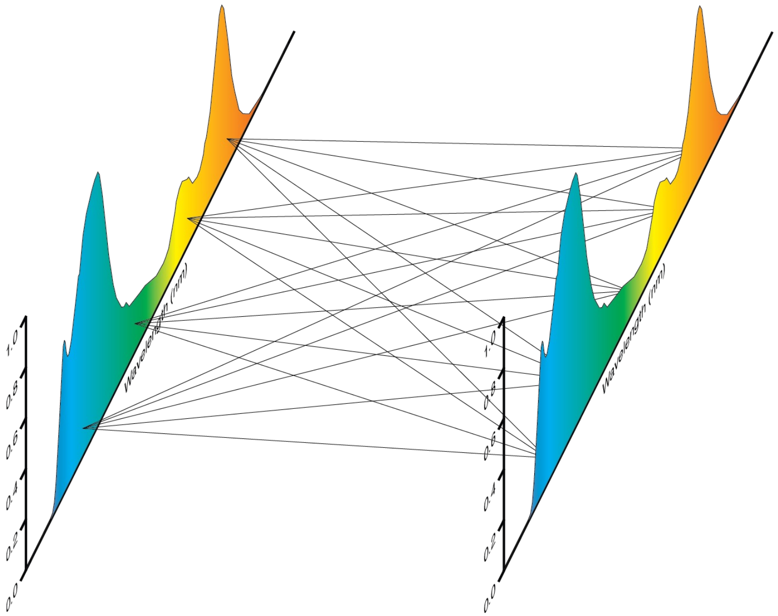
Second harmonic generation with pulses

- With pulses and perfect phase matching, the second harmonic spectrum corresponds to the autoconvolution of the fundamental pulse spectrum

$$P_{\mathrm{NL}}(t) \propto E_\omega(t)^2 \Rightarrow \tilde{P}_{\mathrm{NL}}(\omega) \propto \tilde{E}_\omega(\omega) * \tilde{E}_\omega(\omega)$$

$$\boxed{E_{2\omega}(t) \propto E_\omega(t)^2 \Rightarrow \tilde{E}_{2\omega}(\omega) \propto \tilde{E}_\omega(\omega) * \tilde{E}_\omega(\omega)}$$

- As a result, every frequency component of the fundamental is mixed with every frequency
- Potentially, this can broaden spectrum / shorten pulses



- How well is each of these sum-frequency mixing processes phase-matched?

Phase matching (general)

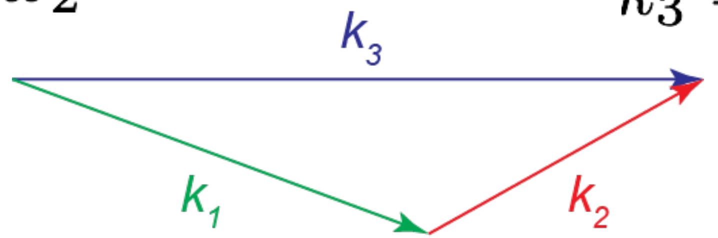
Sum frequency mixing ($\omega_3 > \omega_1, \omega_2$)

Energy conservation

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

Momentum conservation

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



$$\Delta\vec{k} = \vec{k}_3 - \vec{k}_1 - \vec{k}_2$$

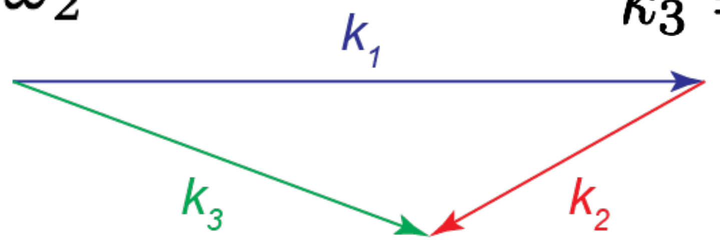
Difference frequency mixing ($\omega_1 > \omega_2, \omega_3$)

Energy conservation

$$\omega_3 = \omega_1 - \omega_2$$

Momentum conservation

$$\vec{k}_3 = \vec{k}_1 - \vec{k}_2$$



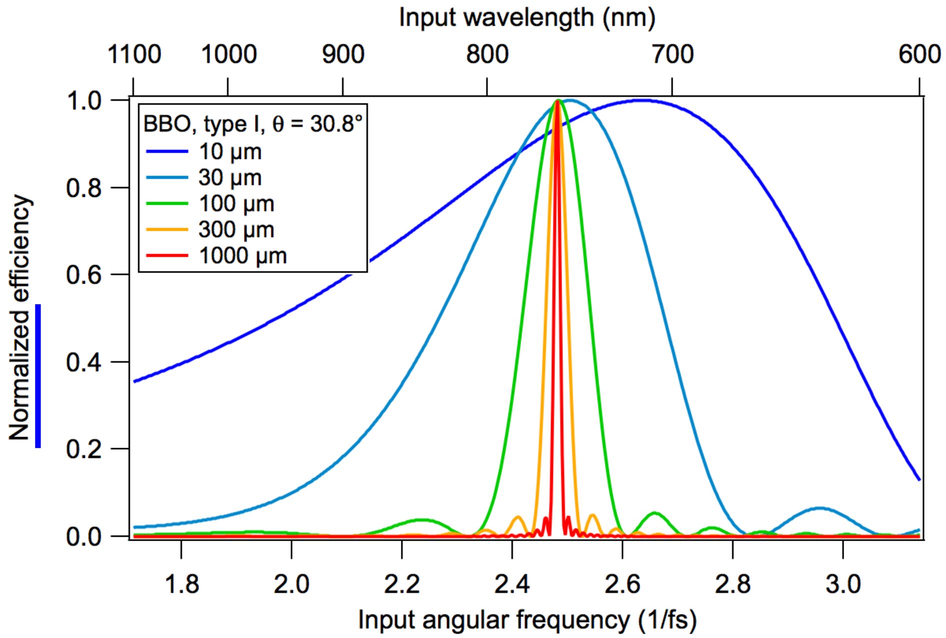
$$\Delta\vec{k} = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$$

Phase matching bandwidth

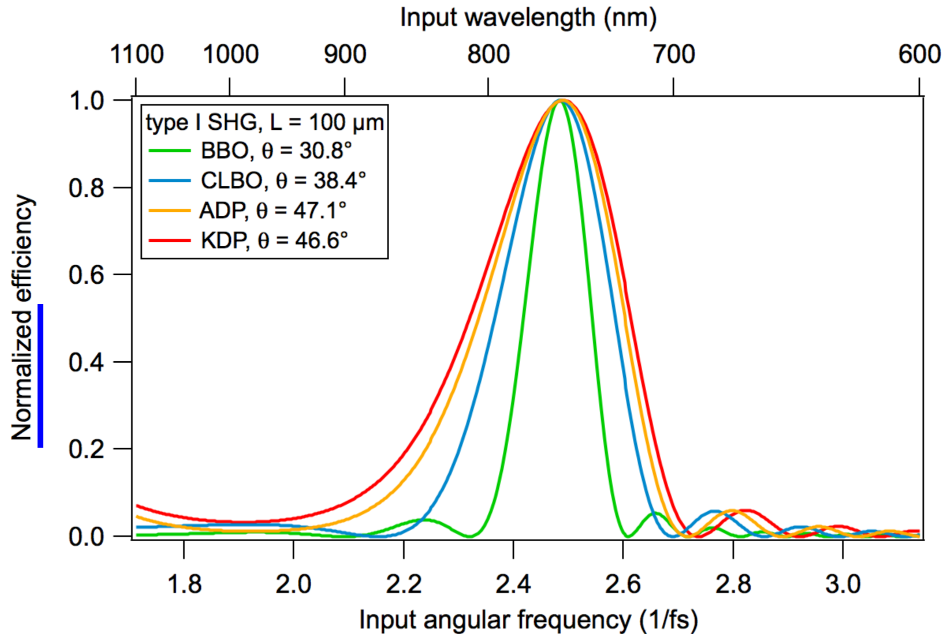
- Here, we estimate conversion bandwidth for a pulse by calculating pure SHG efficiency as a function of frequency (plane wave, no depletion, ignore all other sum-frequency mixing processes)

$$\eta = \frac{I_{2\omega}}{I_\omega} = \frac{2\omega^2}{\epsilon_0 c^3}\left(\frac{d_{\text{eff}}^2}{n_{2\omega} n_\omega^2}\right) I_\omega L^2 \text{sinc}^2\left(\frac{\Delta k L}{2}\right) \propto L^2 \text{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

Different crystal thicknesses



Different crystal materials



Group velocity matching

- Let's revisit the phase-mismatch (here for SHG – can be generalized):

$$k_{2\omega}(\omega) = k_\omega(2\omega)$$

$$\begin{aligned}
\Delta k(\omega) &= \Delta k(\omega_0) + \frac{\partial}{\partial\omega}\Delta k(\omega_0)\Big|_{\omega=\omega_0}(\omega-\omega_0) + \ldots \\
&= k_{2\omega}(\omega_0) - 2k_\omega(\omega_0) + \frac{\partial k_{2\omega}}{\partial\omega}\Big|_{\omega=\omega_0}(\omega-\omega_0) - 2\frac{\partial k_\omega}{\partial\omega}\Big|_{\omega=\omega_0}(\omega-\omega_0) + \ldots \\
&= k_{2\omega}(\omega_0) - 2k_\omega(\omega_0) + 2\frac{\partial k_\omega}{\partial\omega}\Big|_{\omega=2\omega_0}(\omega-\omega_0) - 2\frac{\partial k_\omega}{\partial\omega}\Big|_{\omega=\omega_0}(\omega-\omega_0) + \ldots \\
&= \frac{4\pi\omega_0}{c}\left(n_{2\omega_0} - n_{\omega_0}\right) + 2(\omega-\omega_0)\left(\frac{1}{v_g(2\omega_0)} - \frac{1}{v_g(\omega)}\right) + \ldots
\end{aligned}$$

Vanishes if group velocities match

Can be made to vanish at typically only one frequency (phase velocities matched)

- Δk varies slowly around phasematched frequency (= large bandwidth), if group velocities are matched as well

Considerations for ultrabroadband phase matching

- Ultrabroadband birefringent phase-matching implies very thin crystal
- Very thin crystal implies low conversion efficiency
- Crystal thickness might become comparable with coherence length of other non-phasematched (and possibly unwanted) conversion processes

Point group 3m (e.g. BBO)

$$\begin{pmatrix} P_x^{NL} \\ P_y^{NL} \\ P_z^{NL} \end{pmatrix} = \epsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & d_{16} \\ d_{21} & d_{22} & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \cdot \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_zE_x \\ 2E_xE_y \end{pmatrix}$$

Point group $\bar{4}$2m (e.g. KDP, ADP,..)

$$\begin{pmatrix} P_x^{NL} \\ P_y^{NL} \\ P_z^{NL} \end{pmatrix} = \epsilon_0 \begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{pmatrix} \cdot \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_zE_x \\ 2E_xE_y \end{pmatrix}$$

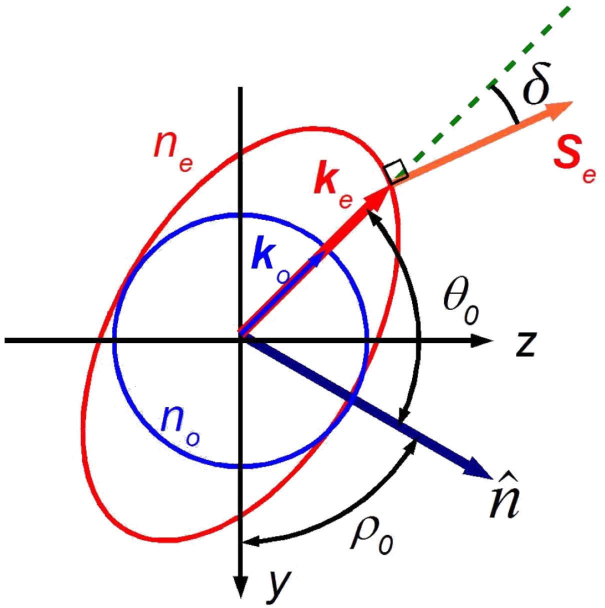
- May lead to signal background or interference effects
- Can sometimes be avoided by using crystal with higher symmetry (maybe in combination with polarizer to reject signals with ‘wrong’ polarization)

(L. Gallmann et al., *Opt. Lett.* **25** (4), 269 (2000))

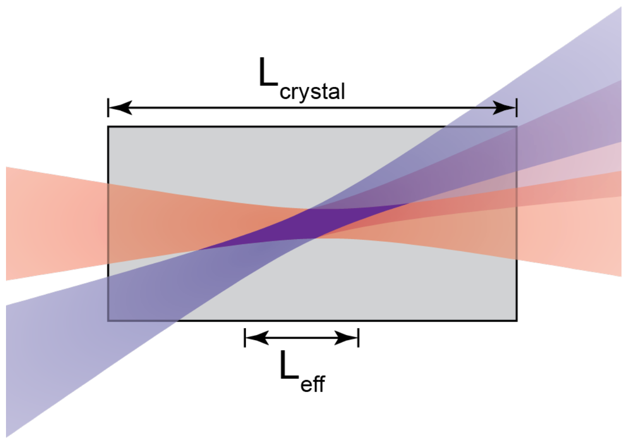
Walk-off effects with real beams and pulses

- Spatial and/or temporal overlap of finite beams and pulses can be limited by the following effects
- This affects conversion efficiency and bandwidth

Spatial walk-off

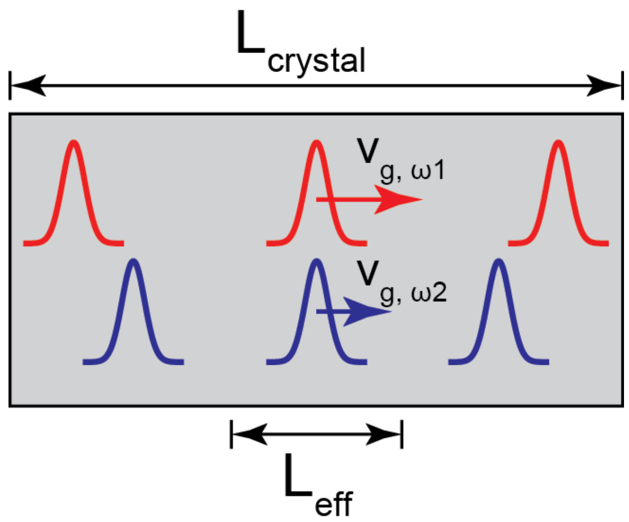


Noncollinear beams

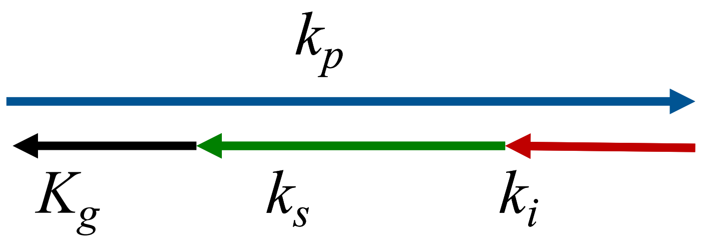


Temporal walk-off

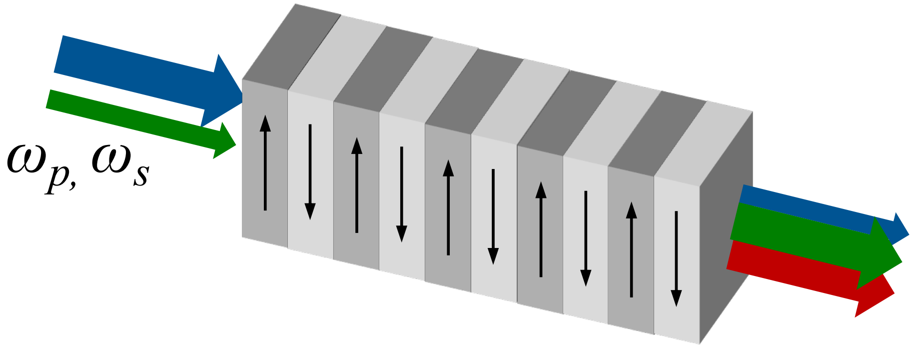
(Group velocity mismatch)



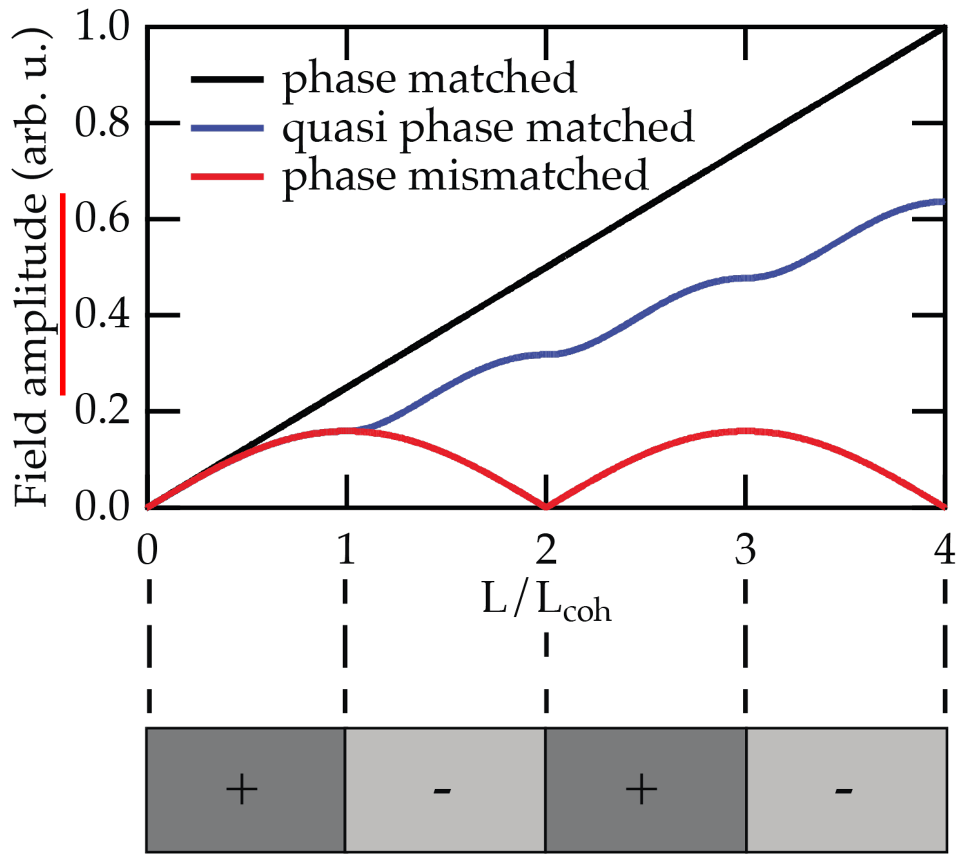
Quasi-phase-matching



$$L_c := \frac{\pi}{\Delta k}$$

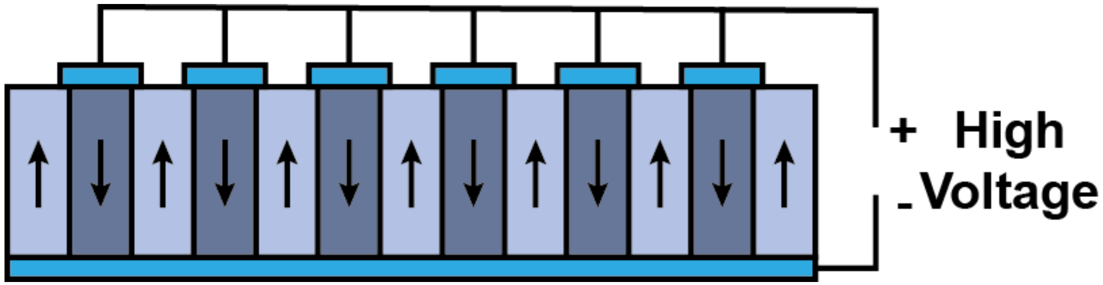


PP $LiNbO_3$ (PPLN)



- Orientation of ferroelectric domain is reversed every L_c
- This 'poling' changes sign of d_{eff}
- Turns destructive interference to constructive interference

Periodic poling: fabrication and benefits



- No birefringence needed for phasematching

  ⇒ Non-critical phasematching possible without spatial walk-off

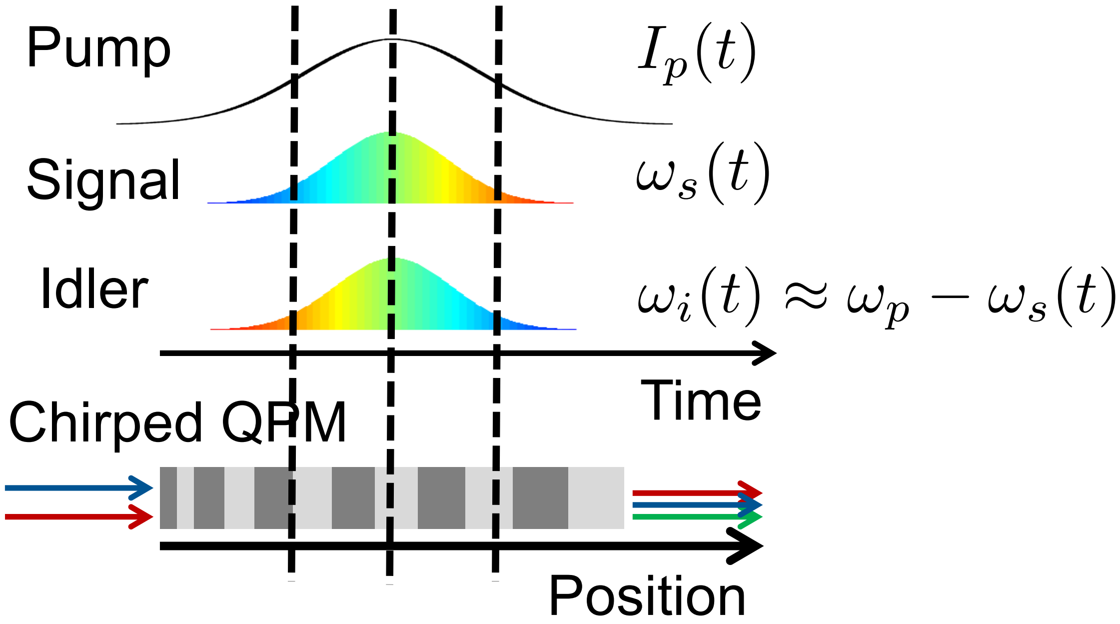
- Can phase-match diagonal elements of nonlinear optical susceptibility tensor

  ⇒ Are typically much larger than off-diagonal elements that are phasematched in birefringent phasematching

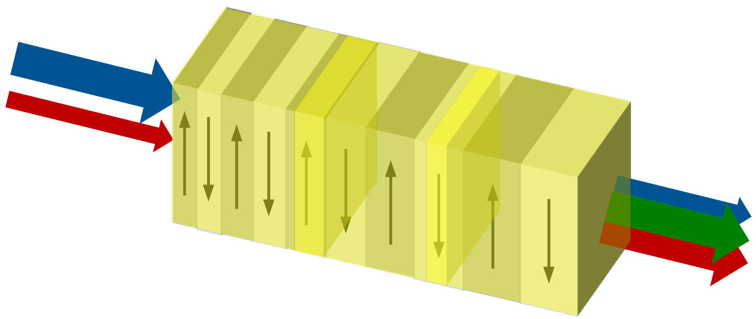
- Can phase-match materials that are not phase-matcheable otherwise (e.g., GaAs)

- Electrodes for poling are manufactured lithographically, domain lengths are therefore precisely engineerable

How to increase the bandwidth?

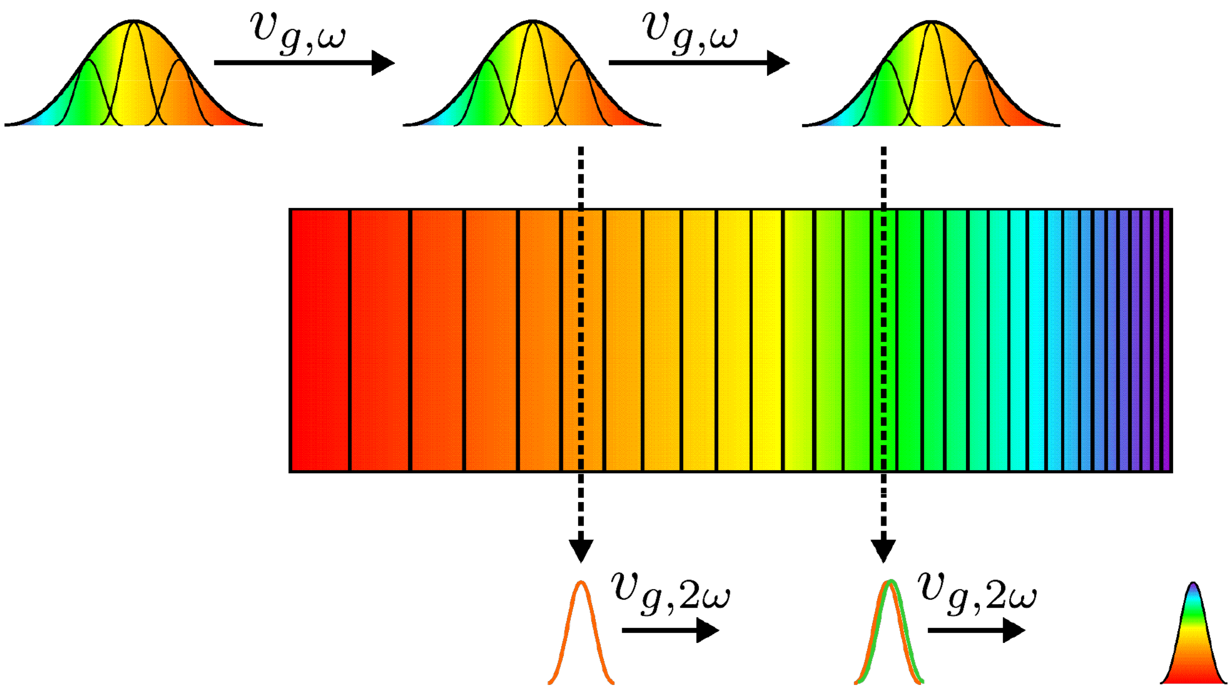


$$K_g(z) = \Delta k(\omega_s(t))$$



QPM-SHG pulse compression

Group velocity mismatch + spatial localization of conversion
⇓
frequency-dependent delay



Group velocity mismatch (GVM):

$v_{g,\omega} \neq v_{g,2\omega}$

Assumption: negligible group velocity dispersion (GVD)

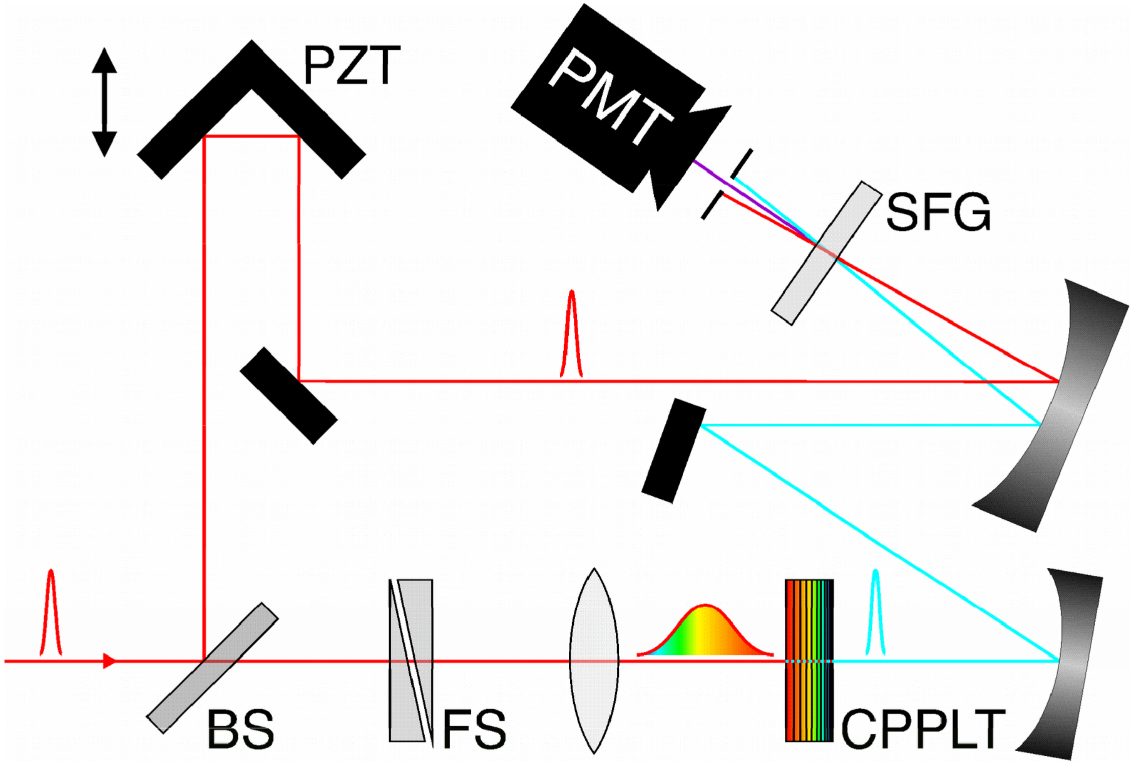
compressed SH if pump chirp and grating chirp are matched

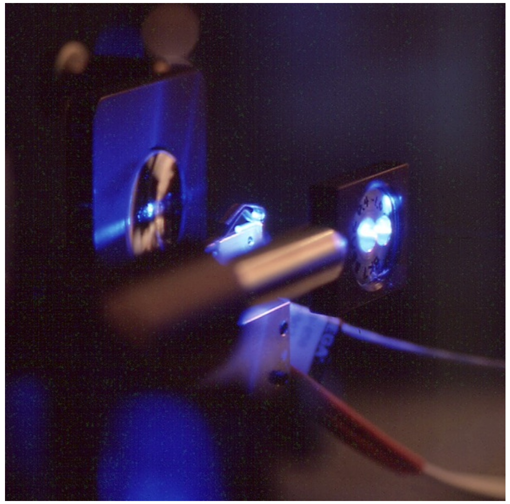
Arbore et al., *Opt. Lett.* **22**, 865 (1997)
Imeshev et al., *J. Opt. Soc. Am.* B **17**, 304 (2000)

- Fundamental and SH chirp engineerable

e.g.: positively chirped fundamental ⇨ negatively chirped SH

Experimental setup

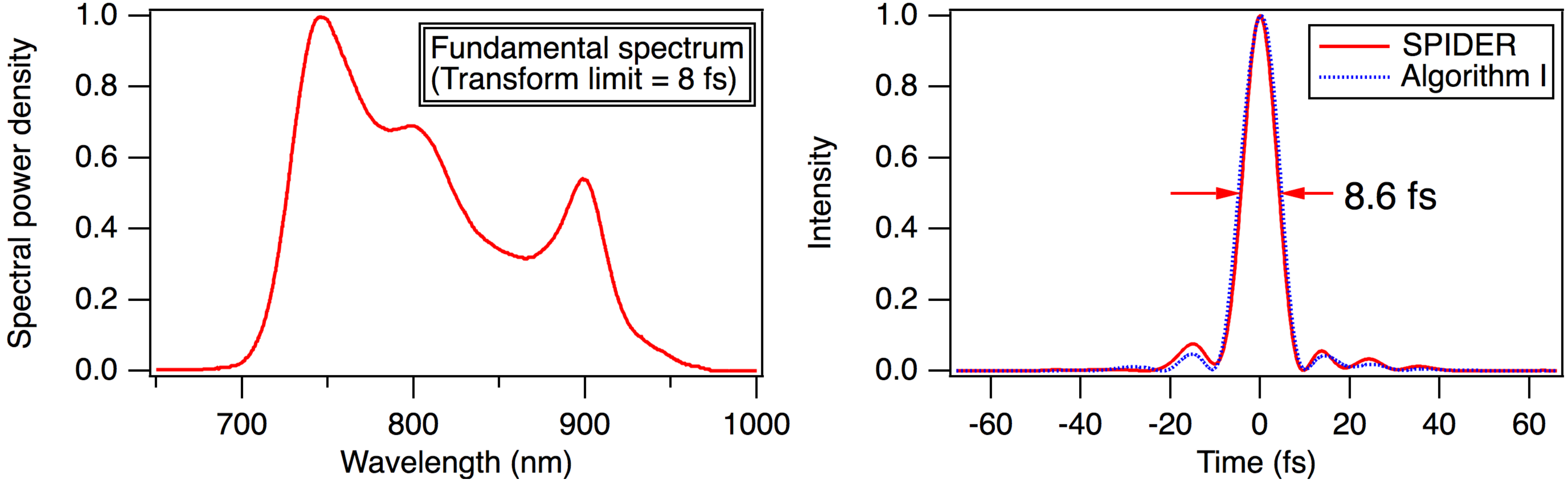




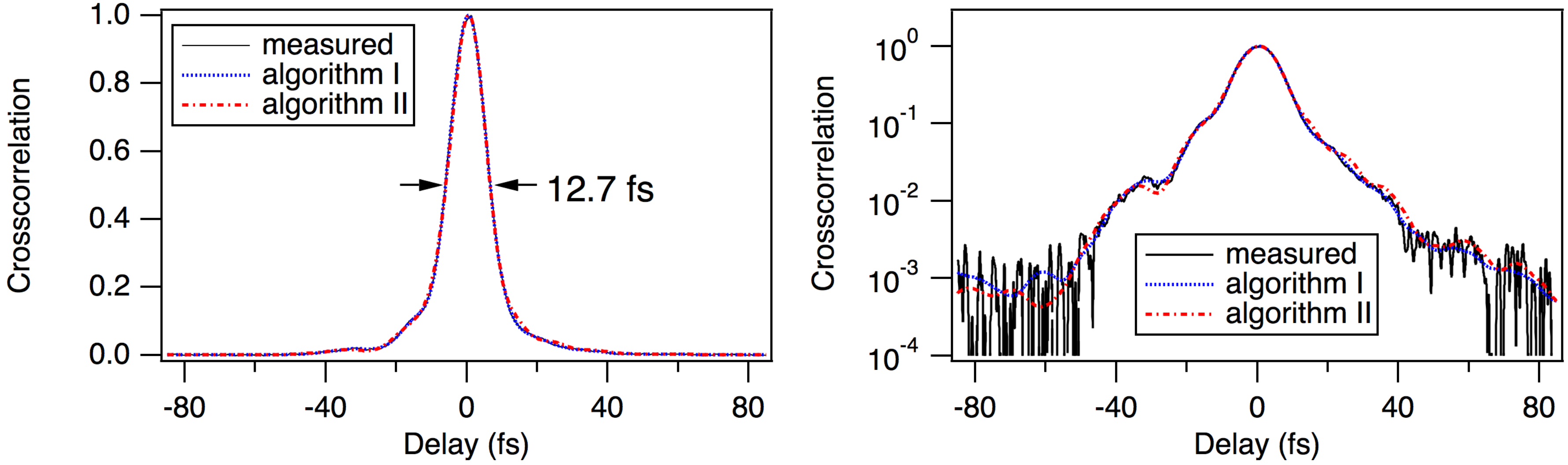
- < 10 μm thick free-standing KDP crystal for crosscorrelation
- Solar-blind PMT for detection

Input pulse & crosscorrelation

Fundamental pulse

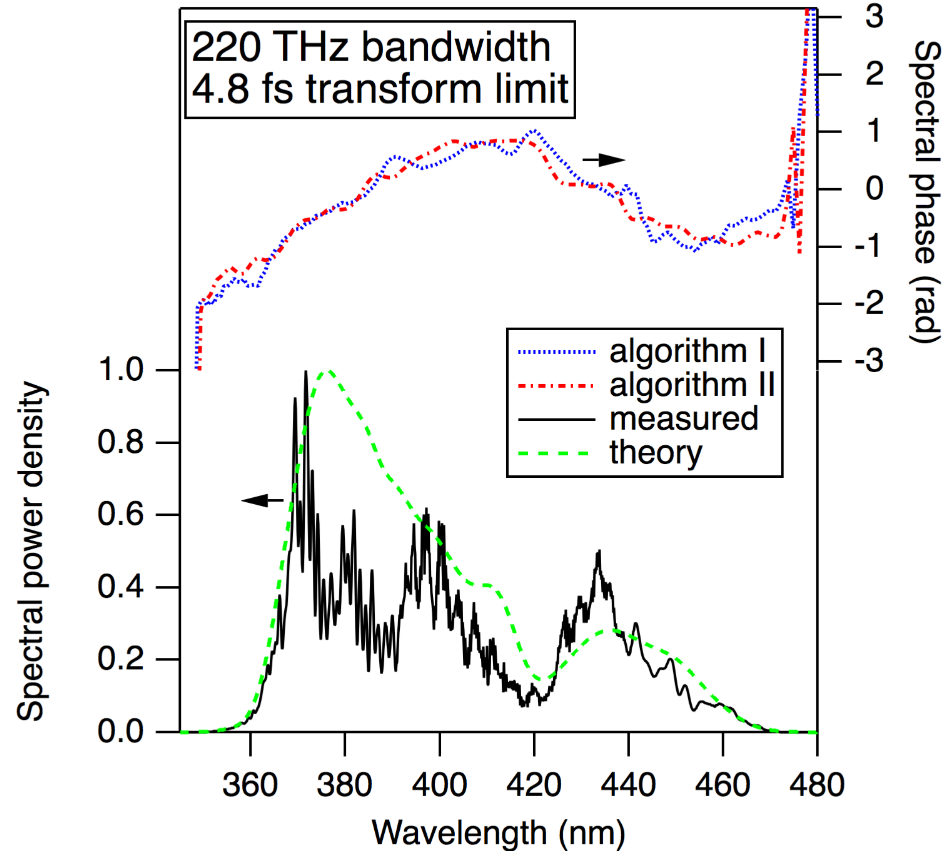


Crosscorrelation

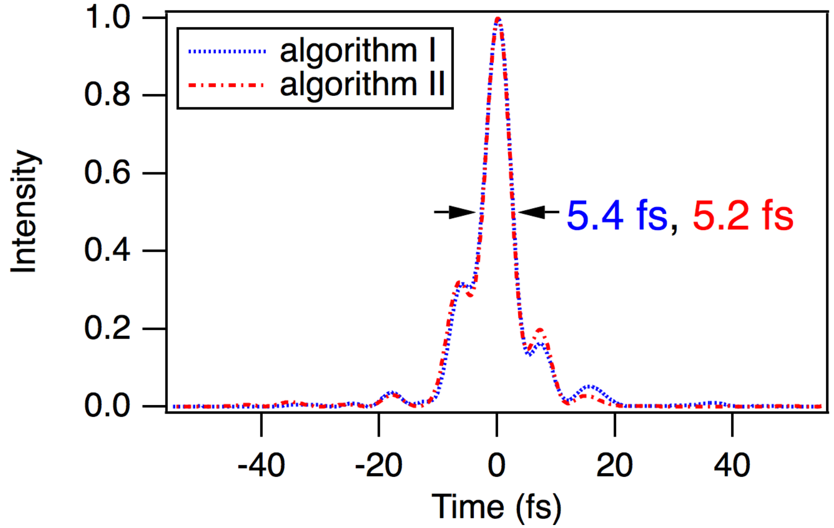


Output pulse

SH spectrum and phase



SH pulse shape



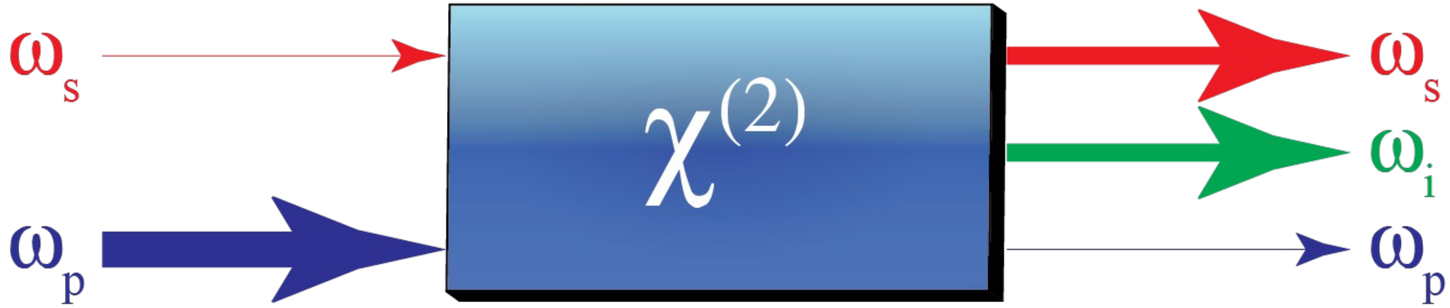
- 90 mW fundamental => 0.45 mW SH
- efficiency: 0.45 %/nJ (theory: 2 %/nJ)

L. Gallmann et al., *Opt. Lett.* **26**, 614 (2001)

Optical parametric amplification (OPA)

- Optical parametric amplification is a special case of difference-frequency generation:
  - Process starts from strong pump wave and weak seed wave
  - Generates strong signal and idler waves (transfers energy from pump)

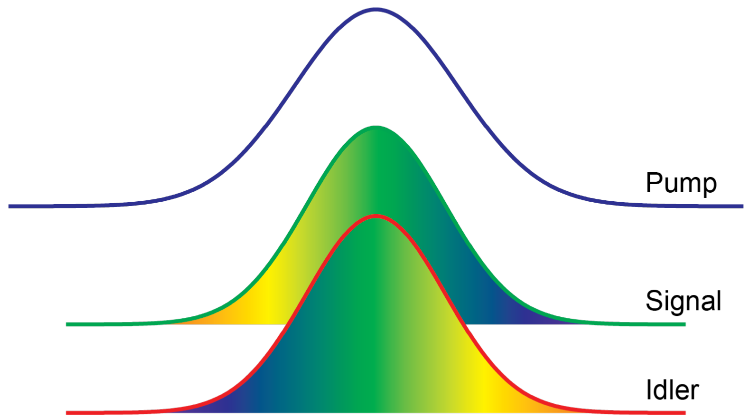
$$\omega_p = \omega_s + \omega_i$$



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

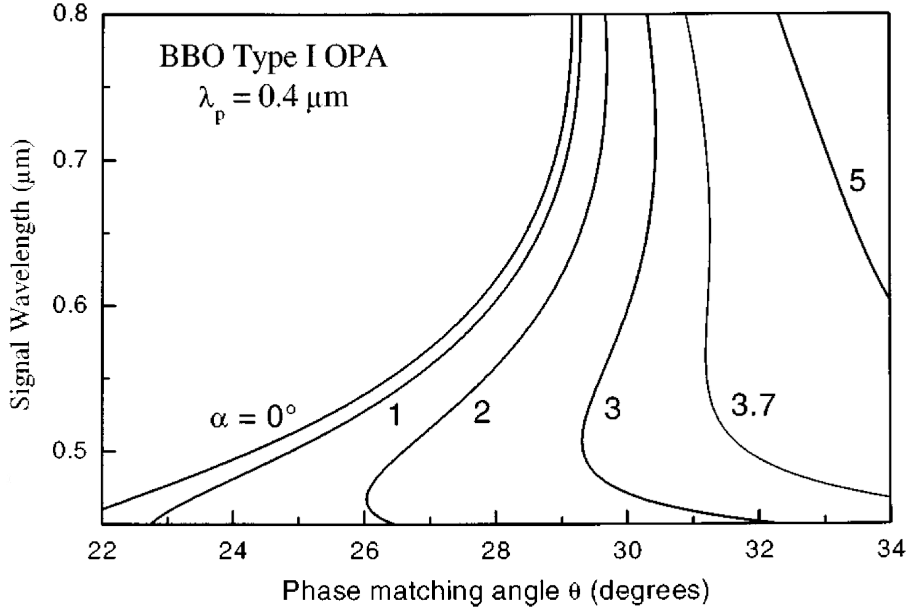
OPA vs. inversion-based amplification

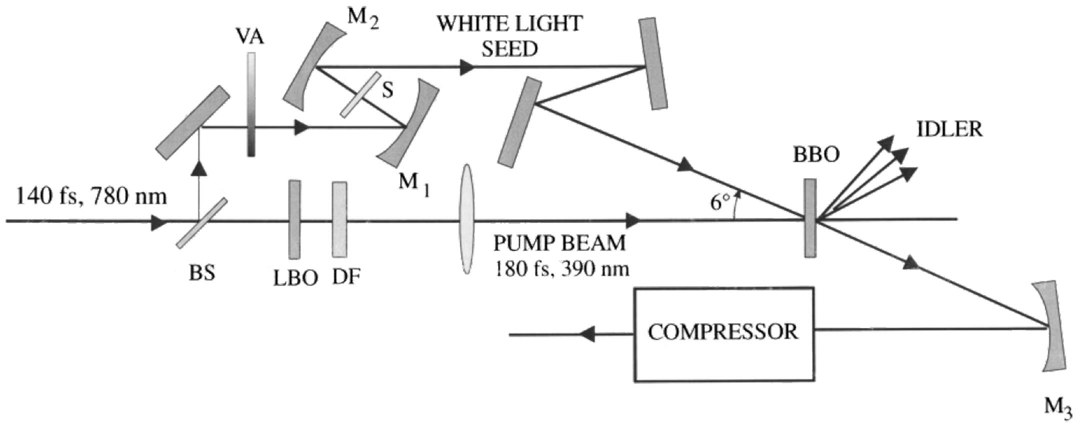
- OPA gain not limited to characteristic atomic or molecular transition (process only needs to be phase-matchable) – can thus provide **gain at "unusual wavelengths"**
- OPA can have **very large single-pass gain** (>50 dB easily possible)
- Amplification happens in OPA through instantaneous process, no energy is stored in crystal
  - ⇒ Pump pulse needs to **temporally coincide** with seed and have **similar pulse duration** (timing critical)
  - ⇒ **Low heat load** in crystal (there are no "non-radiative" transitions involved)
- **Back-conversion** of signal and idler photons into pump photons can happen through sum-frequency mixing (for strong saturation of the pump)
- Gain in OPA is proportional to **pump temporal intensity profile: may reduce bandwidth** of (chirped) amplified pulses

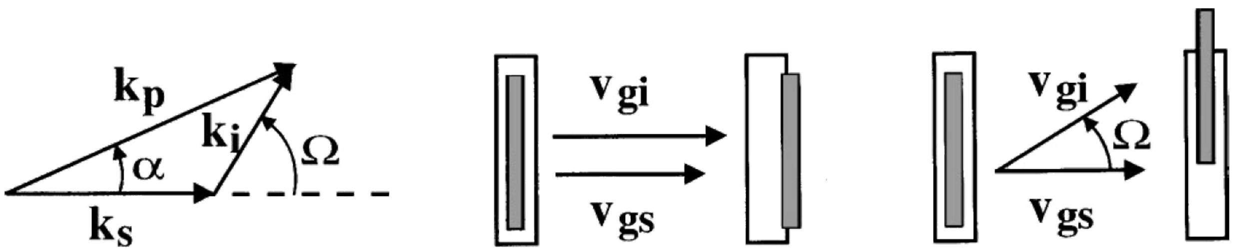


Ultrabroadband, noncollinear OPA

- Ultrabroadband amplification bandwidth can be achieved in a birefringently phasematched OPA by group-velocity matching through non-collinear geometry
- Famous example: BBO-based OPA pumped at 400 nm, emitting in visible

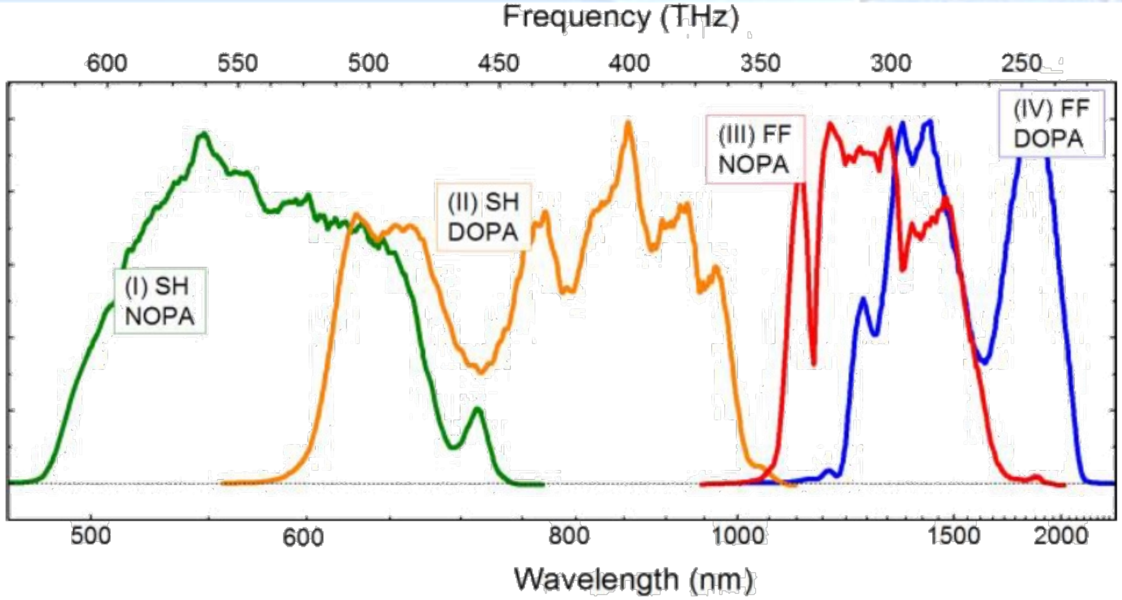


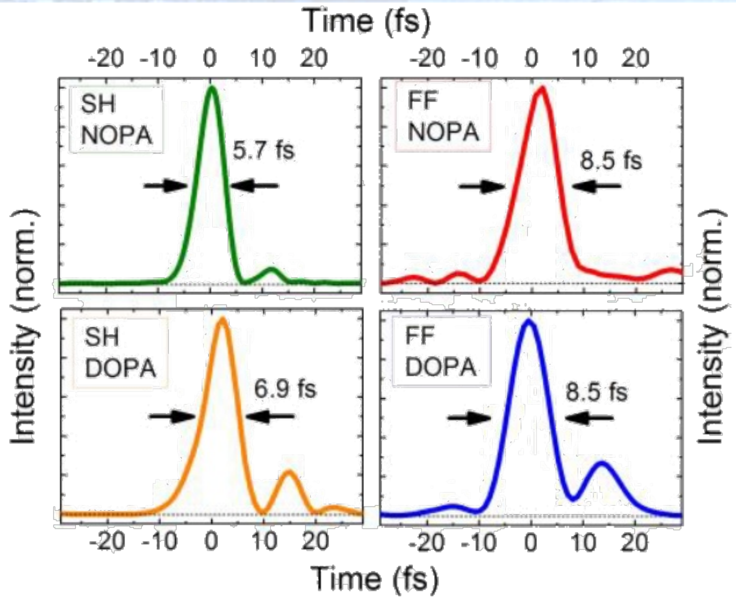




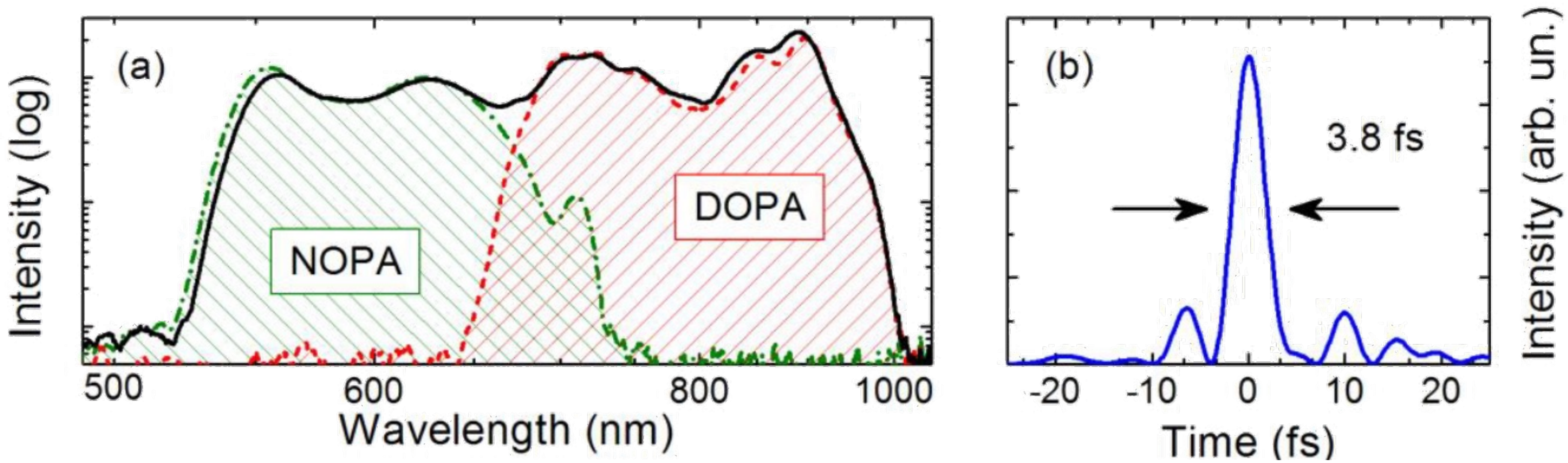
(G. Cerullo et al., *Rev. Sci. Instr.* **74**, 1 (2003))

Waveform synthesis with ultrabroadband OPA



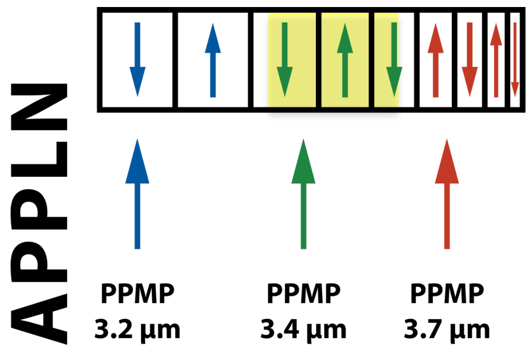


- Combination of two or more amplification channels can yield very short pulses
- Challenge: channels have to be phase stable with respect to each other

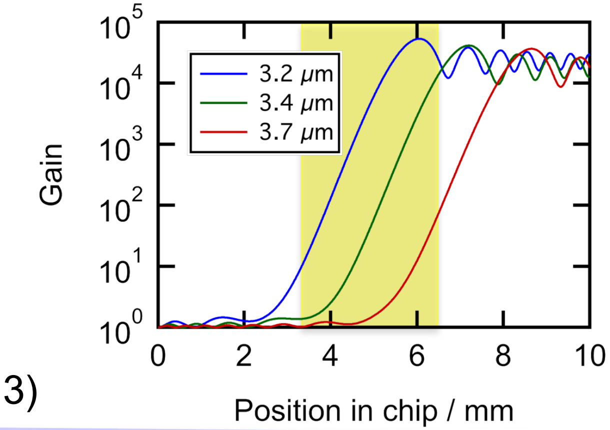


(D. Brida et al., *Nonlinear Optics 2013*, paper NW3A.1 (2013))

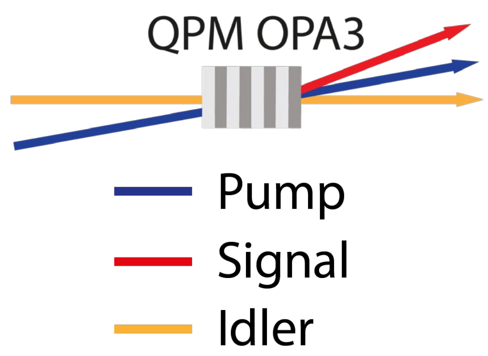
Ultrabroadband QPM amplification schemes



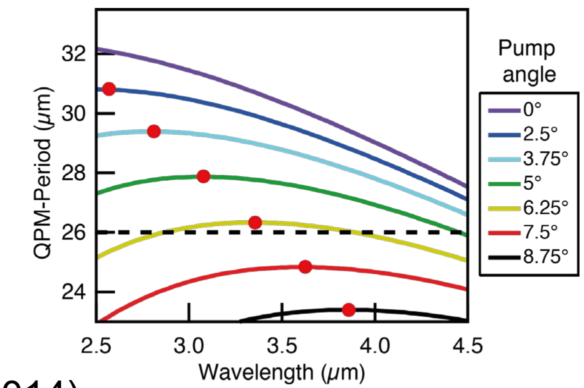
Aperiodic quasi-phase matching



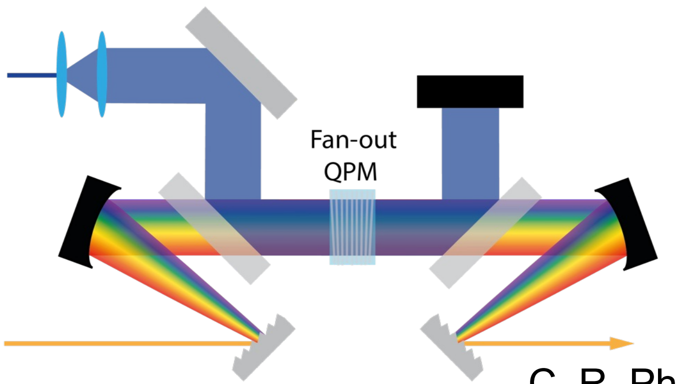
B. W. Mayer et al., *Opt. Lett.* **38**, 4625 (2013)



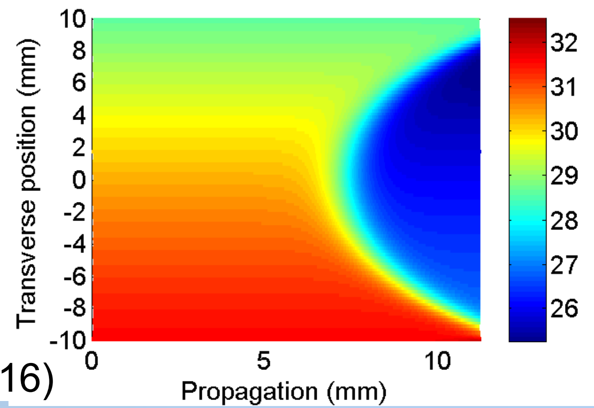
Non-collinear quasi-phase matching



B. W. Mayer et al., *Opt. Express* **22**, 20798 (2014)



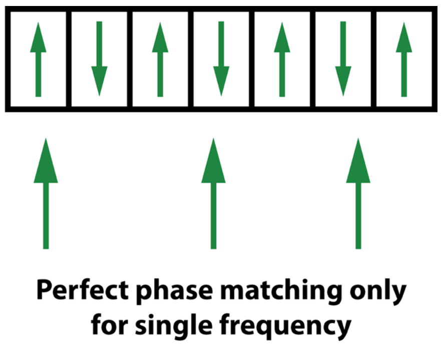
Fourier domain 2D QPM



C. R. Phillips et al., *Opt. Express* **24**, 15940 (2016)

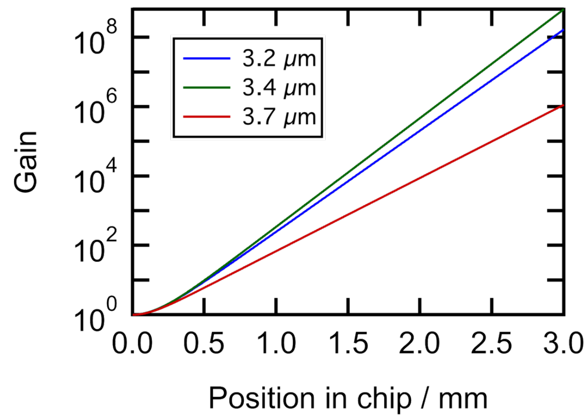
Concept of amplification in APPLN

PPLN

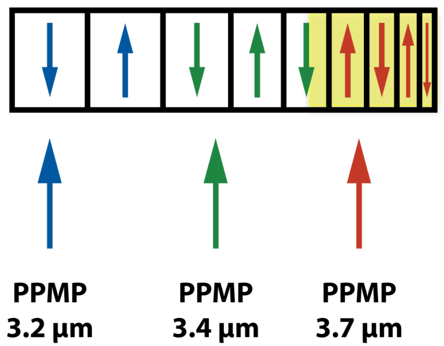


$$K_g = \text{const.}$$

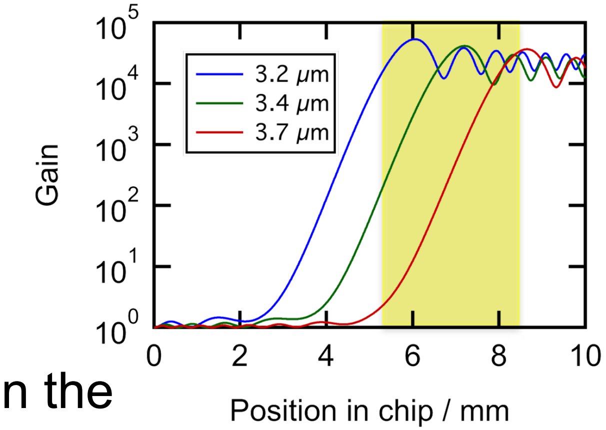
Phase matching for single wavelength



APPLN



$$K_g = K_g(z) = \underbrace{\kappa' \cdot z + \text{const.}}_{\text{linearly chirped grating}}$$



Each spectral component is amplified in a limited region in the grating around its phase matching point (PMP)

- Potential for very broadband phase matching bandwidths and customized gain profiles[1,2]
- Potential for high conversion efficiency[3]

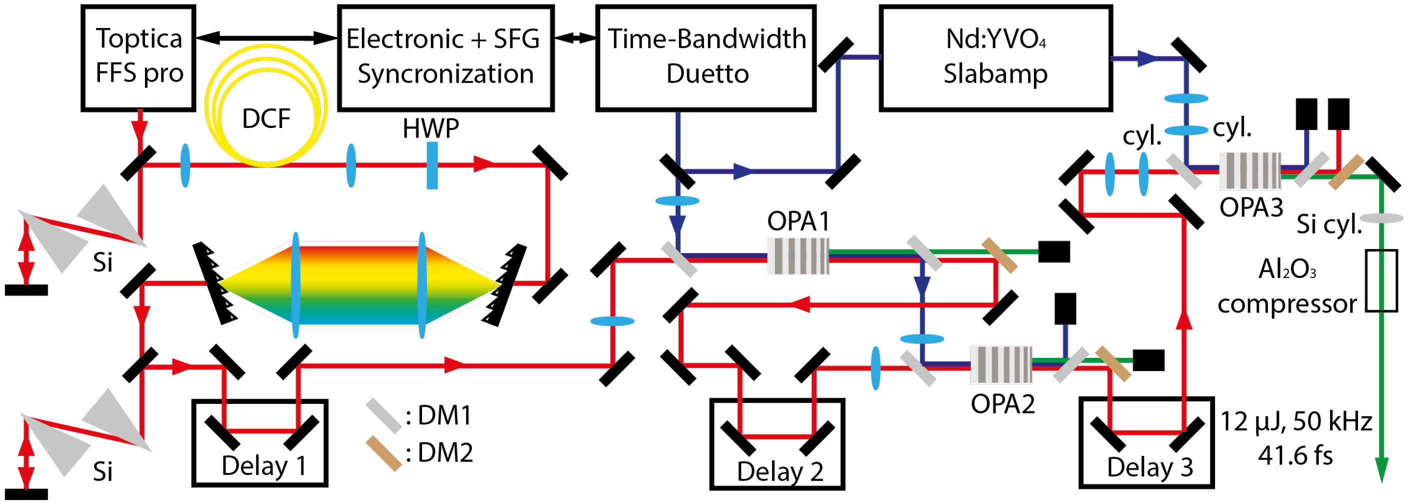
[1]M. Charbonneau-Lefort, B. Afeyan, M. M. Fejer, *J. Opt. Soc. Am. B* **25**, 1402 (2008)

[2]C. R. Phillips, L. Gallmann, M. M. Fejer, *Opt. Express* **21**, 10139 (2013)

[3]C. R. Phillips, C. Langrock, D. Chang, Y. W. Lin, L. Gallmann, M. M. Fejer, *J. Opt. Soc. Am. B* **30**, 1551 (2013)

Mid-infrared OPCPA setup

1.56-µm seeded femtosecond mid-infrared pulse generation

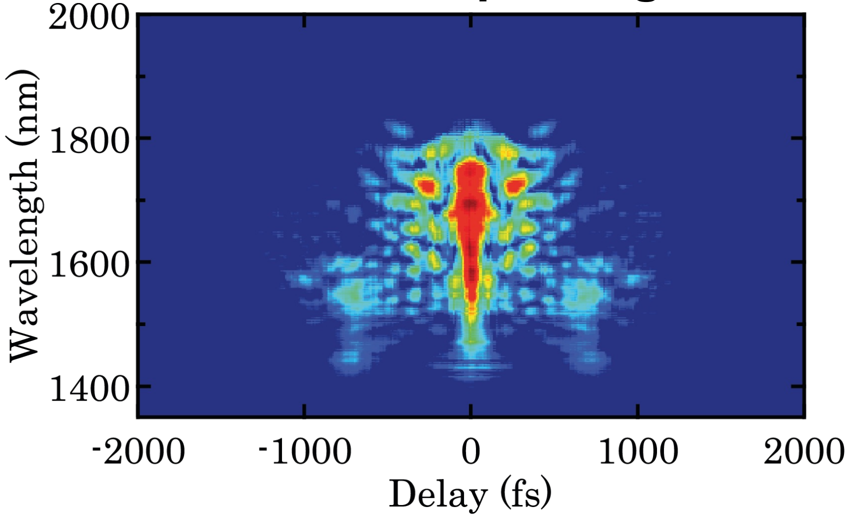


- Seed at 1.56 µm signal wavelength
- Dispersion compensation (stretcher/compressor) mainly on 1.5 µm – NIR seed side
- All-collinear setup
- Idler pulse compression with bulk material (sapphire)
- 3.4 µm idler extracted at last amplification stage

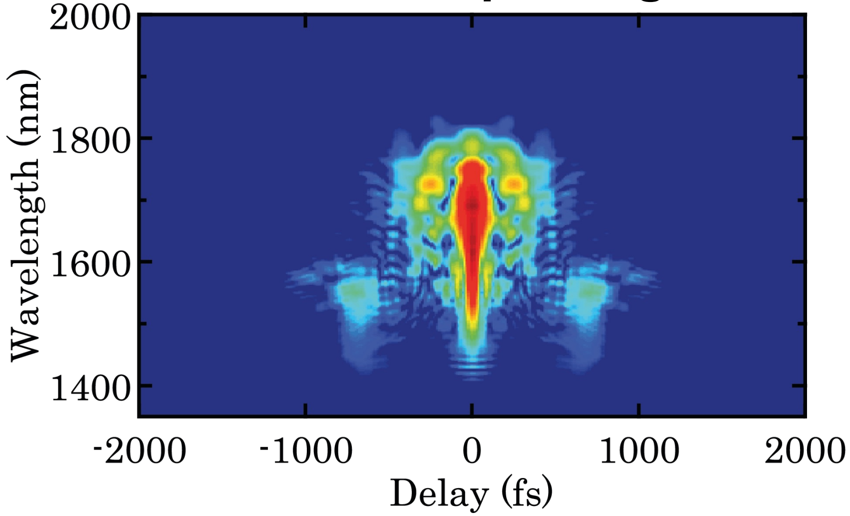
[1]B. W. Mayer, C. R. Phillips, L. Gallmann, M. M. Fejer, U. Keller, *Opt. Lett.* **38**, 4625 (2013)
[2]C. Heese, A. E. Oehler, L. Gallmann, U. Keller, *Appl. Phys. B* **103**, 5 (2011)
[3]C. R. Phillips, B. W. Mayer, L. Gallmann, M. M. Fejer, U. Keller, *Opt. Express* **22**, 9327 (2014)

Measured pulses with SHG FROG

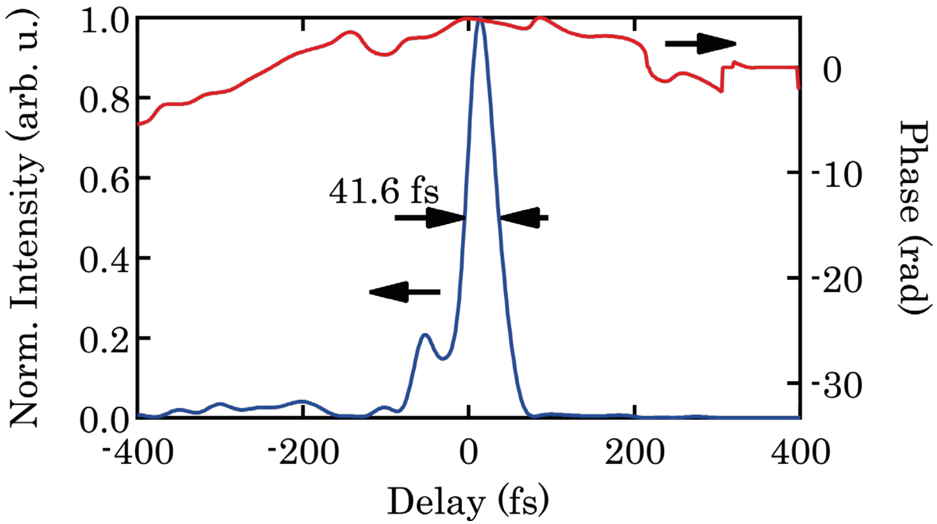
Measured spectrogram



Retrieved spectrogram



Retrieved pulse



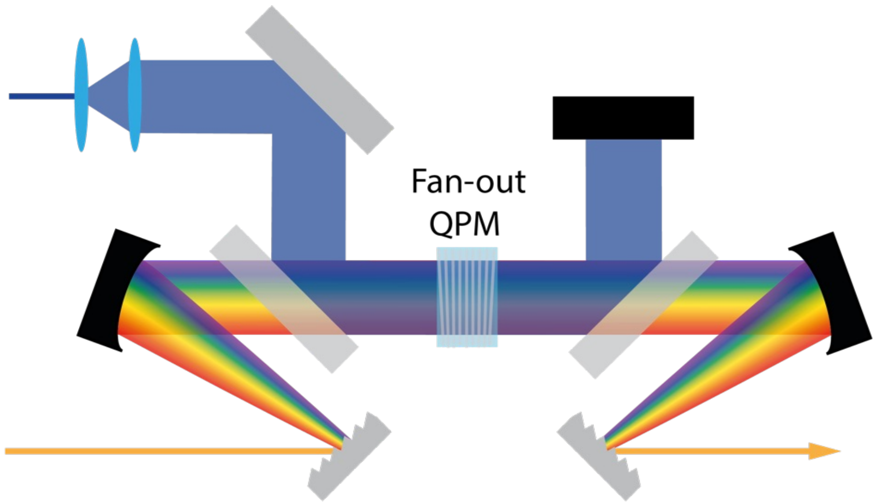
Output parameters

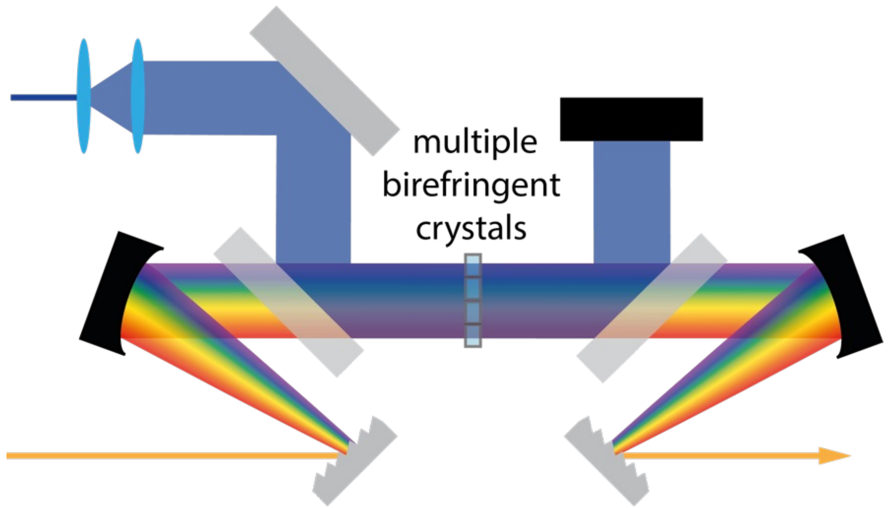
- Pulse energy: **12 μJ**
- Repetition rate: **50 kHz**
- Pulse duration: **41.6 fs**
- Center wavelength**: 3.4 μm**
- Energy inside of crystal: **19 μJ**
- Int. conversion efficiency: **24.5%**

- Single cycle corresponds to 11.3 fs at 3.4 μm center wavelength

B. W. Mayer, C. R. Phillips, L. Gallmann, M. M. Fejer, U. Keller, *Opt. Lett.* **38**, 4625 (2013)

Fourier-domain OPA





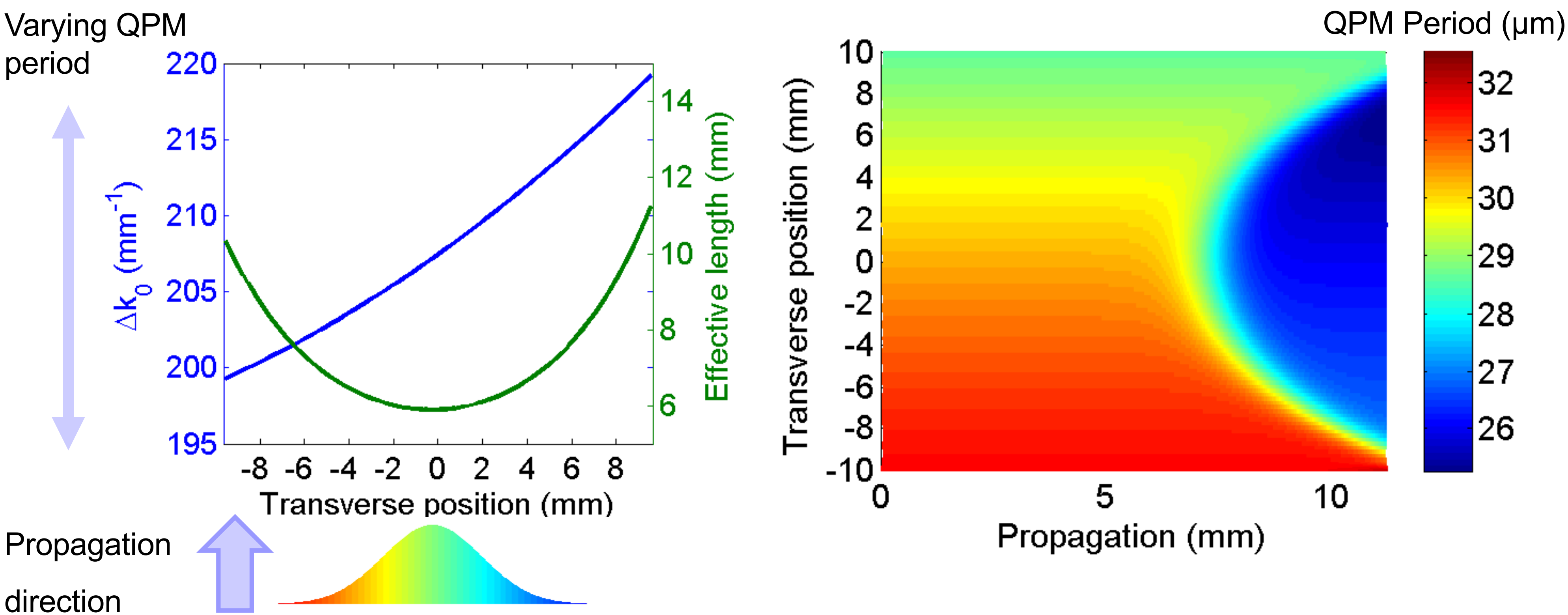
- Concept proposed with fan-out PPLN:
  - L. Chen, *et al.* "Ultrabroadband optical parametric chirped-pulse amplifier using a fan-out periodically poled crystal with spectral spatial dispersion" PRA **82**, 043843 (2010)

- Experiment with multiple birefringent phase-matching crystals:
  - B. E. Schmidt *et al.* "Frequency domain optical parametric amplification," Nat. Commun. **5** (2014)

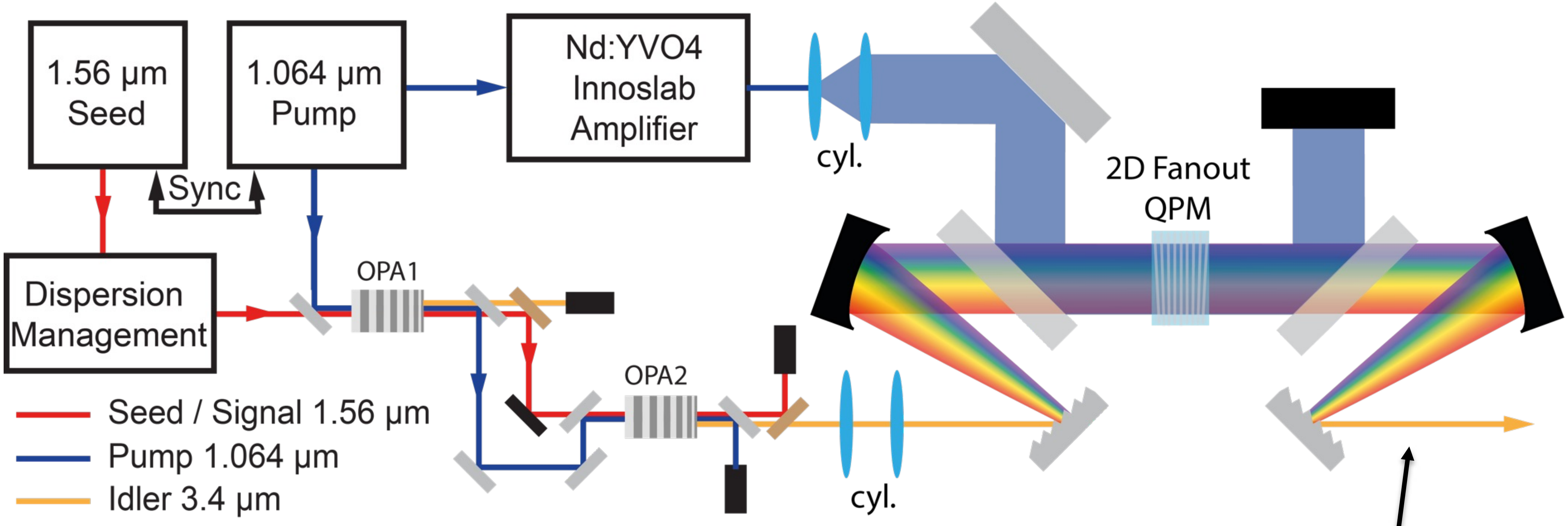
Advanced 2D-QPM functionalities

What can we do with a more general structure than conventional straight-domain fan-out?

- **Choose optimal QPM period trajectory vs. transverse position**
- **Vary interaction length for different frequencies (accounting for a gaussian transverse intensity pump beam profile)**
  - Still monolithic / plane-parallel / non-critical phase-matching crystal



Experimental results: power & spectrum



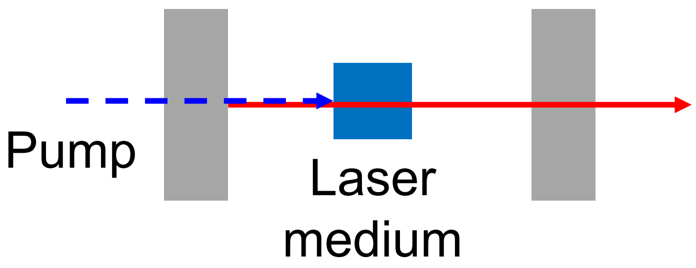
- Output power
  - **33 µJ (1.65 W @ 50 kHz)** after QPM grating
  - Corresponding to a **photon conversion efficiency of 32%**
  - **21 µJ (1.05 W @ 50 kHz)** including losses from the diffraction grating

C. R. Phillips et al., *Opt. Express* **24**, 15940 (2016)

Optical parametric oscillator (OPO)

Laser

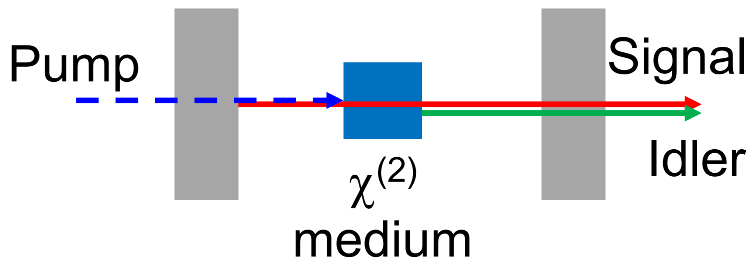
- Optical cavity
- Gain from stimulated emission
- "Pump" photons create a population inversion



$$\omega_{pump} > \omega_{laser}$$

OPO

- Optical cavity
- Gain from parametric amplification
- "Pump" photons converted directly into "signal" photons

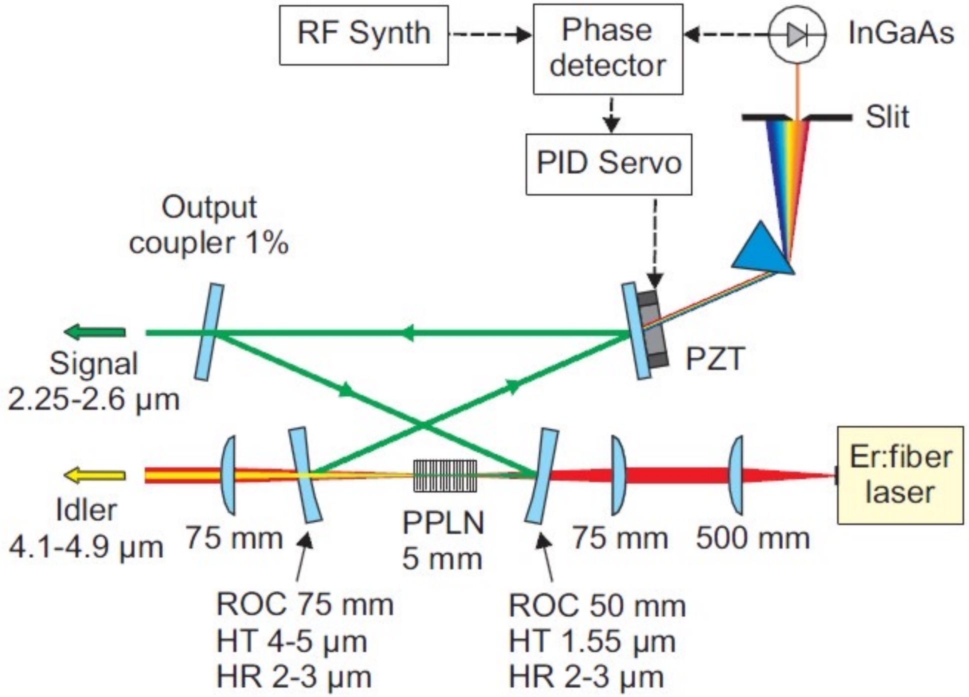


$$\omega_{pump} = \omega_{signal} + \omega_{idler}$$

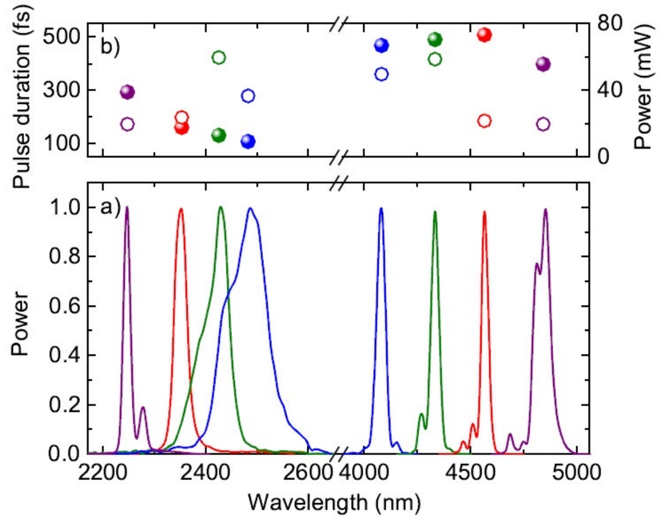
An OPO lets us generate light at wavelengths where we don't have a laser available

Ultrafast optical parametric oscillator (OPO)

- Parametric amplification can also be used inside oscillators (i.e., with feedback from cavity)
- Instantaneous gain process: pump pulse train needs to be synchronized with pulse oscillating in cavity
- Benefit: source is not limited to existing laser transitions ➔ OPOs are often widely tunable in wave-length
- OPOs are available from UV to far infrared



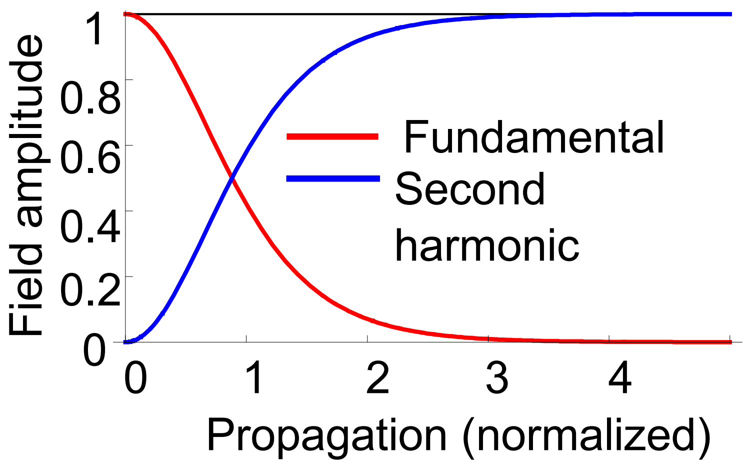
(image source: https://www.fisi.polimi.it/en/research/research_structures/laboratories/54057)



Cascaded quadratic nonlinearities

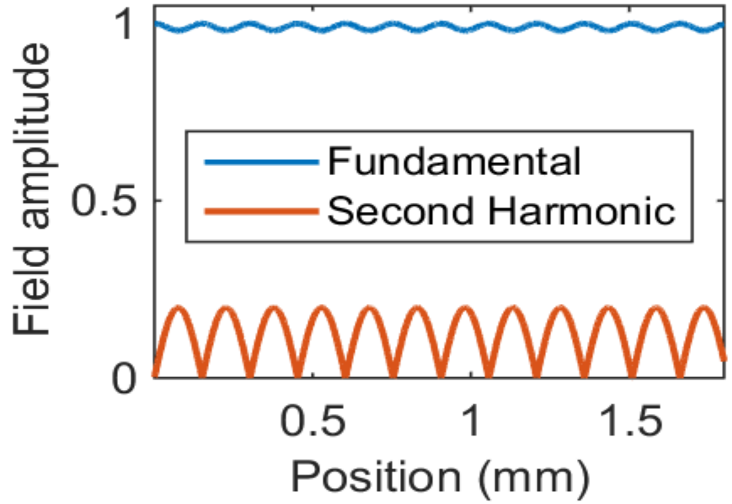
Normal cw SHG:

- Phase-matched, Δk=0
- Energy transfer from fundamental to SH



Cascaded quadratic nonlinearities:

- Large SHG phase-mismatch Δk
- Periodic transfer of energy



$$\frac{dE_{2\omega}}{dz} = -i\frac{\omega d_{eff}}{n_{2\omega}c}E_\omega^2 e^{i\Delta kz} \Rightarrow E_{2\omega} \sim -\frac{1}{\Delta k}\frac{\omega d_{eff}}{n_{2\omega}c}E_\omega^2 e^{i\Delta kz} \Rightarrow \frac{dE_\omega}{dz} = i\frac{1}{\Delta k}\frac{\omega^2 d_{eff}^2}{n_\omega n_{2\omega}c^2}\left|E_\omega\right|^2 E_\omega$$

$$\frac{dE_\omega}{dz} = -i\frac{\omega d_{eff}}{n_\omega c}E_{2\omega}E_\omega^* e^{-i\Delta kz}$$

Effective $\chi^{(3)}$ whose sign depends on Δk! ⇒ Solitons in the positive GDD regime