# Guoqing (Noah) Chang, October 13, 2015 Nonlinear optics: a back-to-basics primer Lecture 3: phase matching

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## **Coupled wave equation**



## **Coupled wave equation**

$$\frac{d^{2}A_{n}}{dz^{2}} - 2jk_{n}\frac{dA_{n}}{dz} = -\frac{\omega_{n}^{2}}{\varepsilon_{0}c_{0}^{2}}P_{NL}(\omega_{n})e^{jk_{n}z} \qquad \mathbf{E}(\omega_{n}) = A_{n}(z)e^{-jk_{n}z} \qquad \lambda_{n} = \frac{2\pi}{k_{n}}$$

$$\boxed{\begin{array}{l} \text{Slowly varying} \\ \text{amplitude} \\ \text{approximation} \end{array}} \quad \left|\frac{d^{2}A_{n}}{dz^{2}}\right| << \left|k_{n}\frac{dA_{n}}{dz}\right| \Leftrightarrow \left(\left|\frac{d^{2}A_{n}}{dz^{2}}\right|\lambda_{n}\right)/\left|\frac{dA_{n}}{dz}\right| << 2\pi$$

$$\boxed{\begin{array}{l} \frac{dA_{n}}{dz} = -j\frac{\omega_{n}^{2}}{2\varepsilon_{0}c_{0}^{2}k_{n}}P_{NL}(\omega_{n})e^{jk_{n}z} \quad k_{n}^{2} = \frac{\omega_{n}^{2}n^{2}(\omega_{n})}{c_{0}^{2}} \end{array}} \qquad \boxed{\begin{array}{l} \frac{dA_{n}}{dz} = -j\frac{\omega_{n}}{2\varepsilon_{0}n(\omega_{n})c_{0}}P_{NL}(\omega_{n})e^{jk_{n}z} \end{array}}$$

Example: three wave mixing at three different frequencies,  $\omega_1, \omega_2, \omega_3$  and  $\omega_3 = \omega_1 + \omega_2$ 

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## **Monley-Rowe relations**

Example: three wave mixing at three different frequencies,  $\omega_1, \omega_2, \omega_3$  and  $\omega_3 = \omega_1 + \omega_2$ 

If we define wave vector mismatch  $\Delta k = k_1 + k_2 - k_3$ , we can rewrite the coupled wave equations:

$$\frac{dA_3}{dz} = -j\frac{2\omega_3 d_{eff}}{n(\omega_3)c_0}A_1A_2e^{-j\Delta kz} \qquad \frac{dA_1}{dz} = -j\frac{2\omega_1 d_{eff}}{n(\omega_1)c_0}A_3A_2^*e^{j\Delta kz} \quad \frac{dA_2}{dz} = -j\frac{2\omega_2 d_{eff}}{n(\omega_2)c_0}A_3A_1^*e^{j\Delta kz}$$

Intensity is a more convenient physical quantity, which is related to electric field as

 $I = 2n\varepsilon_0 c_0 |A|^2 = 2n\varepsilon_0 c_0 AA^* \quad \text{Intensity variation is described as:} \qquad \frac{dI}{dz} = 2n\varepsilon_0 c_0 (A\frac{dA^*}{dz} + A^*\frac{dA}{dz})$ 

Using coupled wave equations, we can derive the following intensity variation equations:

$$\frac{dI_1}{dz} = -8\varepsilon_0 d_{eff} \omega_1 \operatorname{Im}(A_3 A_1^* A_2^* e^{j\Delta kz}) \qquad \frac{dI_2}{dz} = -8\varepsilon_0 d_{eff} \omega_2 \operatorname{Im}(A_3 A_1^* A_2^* e^{j\Delta kz}) \qquad \frac{dI_3}{dz} = 8\varepsilon_0 d_{eff} \omega_3 \operatorname{Im}(A_3 A_1^* A_2^* e^{j\Delta kz})$$

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

$$\operatorname{Energy \ conservation \ in a \ lossless \ system} \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_1 \ photons \ are \ created = The \ rate \ at \ which \ } \omega_2 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ at \ which \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ rate \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ } \omega_3 \ photons \ are \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ } \omega_3 \ photons \ destroyed \qquad \operatorname{Ime \ } \omega_3 \ photons \ are \ } \omega$$

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## **Solution with no depletion**

Example: three wave mixing at three different frequencies,  $\omega_1, \omega_2, \omega_3$  and  $\omega_3 = \omega_1 + \omega_2$ Let's consider the special case that  $\omega_1$  field and  $\omega_2$  field are not depleted; that is,  $A_1$  and  $A_2$  are constant.



matching condition: 
$$\Delta k = k_1 + k_2 - k_3 = 0 \qquad I_3(z) = \frac{8\omega_3 u_{eff} I_1 I_2 z}{n(\omega_1)n(\omega_2)n(\omega_3)\varepsilon_0 c_0^2}$$

How to select a nonlinear crystal to maximize the nonlinear effect? We can define a figure of merit (FOM) to compare different crystals:  $d_{aff}^2$ 

$$FOM = \frac{d_{eff}^2}{n(\omega_1)n(\omega_2)n(\omega_3)}$$
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## **Phase matching**

$$I_3(z) = \frac{8\omega_3^2 d_{eff}^2 I_1 I_2 z^2}{n(\omega_1)n(\omega_2)n(\omega_3)\varepsilon_0 c_0^2} \sin c^2 (\frac{\Delta kz}{2}) \qquad L_{coh} \equiv \frac{\pi}{|\Delta k|}$$

Coherence length: the propagation distance at which the three waves accumulate a  $\pi$  phase difference.

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## A more intuitive picture: SHG

Z=0z z+dz7=I  $E_1 \exp[j(\omega_1 t - k_1 z)]$ Input (fundamental) field at z  $d_{eff} E_1^2 \exp[j(2\omega_1 t - 2k_1 z)]$ SH polarization of the medium at z  $d_{eff} E_1^2 \exp[j(2\omega_1 t - 2k_1 z)]dz$ SH field radiated by SH polarization within z and z+dz  $d_{eff} E_1^2 \exp[j(2\omega_1 t - 2k_1 z) - jk_2(l-z)]dz$ SH field at Z=L radiated  $d_{eff} E_1^2 \exp[j(2\omega_1 t - k_2 l) + j(k_2 - 2k_1)z] dz$ by SH polarization at z  $\omega_2 = 2\omega_1$  $k_2 = \omega_2 c / n(\omega_2)$ 

If the phase matching condition  $\Delta k \equiv k_2 - 2k_1 = 0$  is satisfied, the SH field arriving at Z=L is independent on the position z from where the SH field originates. In other words all SH field contribution, from 0 to L, add in phase at z=L, leading to the 7 highest SHG efficiency.

## Wavelength conversion using the 2<sup>nd</sup> order nonlinear optics

## Sellmeier equation to model refractive index

If the frequency is far away from the absorption resonance  $|\omega_0^2 - \omega^2| >> 2\omega\gamma$ 

$$\chi(\omega) = \frac{\omega_p^2}{(\omega_0^2 - \omega^2)} \qquad \omega_p^2 = Ne^2 / (m\varepsilon_0)$$

Normally there are multiple resonant frequencies for the electronic oscillators. It means in general the refractive index will have the form

$$n^{2}(\omega) = 1 + \chi(\omega) = 1 + \sum_{i} A_{i} \frac{\omega_{i}^{2}}{\omega_{0}^{2} - \omega^{2}} = 1 + \sum_{i} a_{i} \frac{\lambda^{2}}{\lambda^{2} - \lambda_{i}^{2}}$$

For the frequency (wavelength) far away from absorption resonance, refractive index increases with increasing frequency, which leads to

$$\omega_3 n(\omega_3) = (\omega_1 + \omega_2) n(\omega_3) > \omega_1 n(\omega_1) + \omega_2 n(\omega_2)$$

Therefore, dispersion prevents phase matching in an isotropic medium. How about an anisotropic medium?

## Example: SHG of o wave in BBO



We can project the polarization onto the direction of o wave and e wave which are normal to k:

$$P_o(2\omega) = -2\varepsilon_0 d_{16} E^2(\omega) \cos 3\phi$$

Case 1:  $\phi = 0, \theta = 0$  Fundamental o wave generates SH o wave, demanding phase matching condition of

$$n_o(2\omega) = n_o(\omega)$$

 $P_e(2\omega) = 2\varepsilon_0 (d_{31}\sin\theta + d_{16}\cos\theta\sin3\phi)E^2(\omega)$ Case 2: $\phi = 90^\circ, \theta = 0$  Fundamental o wave

generates SH e wave, demanding phase matching condition of

$$n_e(2\omega, \theta = 0) = n_o(\omega)$$
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## Example: SHG of o wave in BBO

We can project the polarization onto the direction of o wave and e wave which are normal to k:

 $P_o(2\omega) = -2\varepsilon_0 d_{16}E^2(\omega)\cos 3\phi$ Case 1:  $\phi = 0, \theta = 0$  Fundamental o wave generates SH o wave (o $\rightarrow$ o), demanding phase matching condition of

$$\begin{split} P_e(2\omega) &= 2\varepsilon_0 (d_{31}\sin\theta + d_{16}\cos\theta\sin3\phi)E^2(\omega) \\ \text{Case } 2:\phi &= 90^\circ, \theta = 0 \text{ Fundamental o wave} \\ \text{generates SH e wave}(o \rightarrow e), \text{ demanding} \\ \text{phase matching condition of} \end{split}$$

$$n_e(2\omega, \theta = 0) = n_o(\omega)$$





### Phase matching for $o \rightarrow e$ SHG in BBO





## **Angle tuning for phase-matching**



For second harmonic generation, the wave with lower frequency is called fundamental wave. From photon picture, two o wave photons generate one e wave SHG photon. So we note this type of SHG process as  $o + o \rightarrow e$ 

How about other possible combinations?

$$e + e \rightarrow o \quad o + e \rightarrow o \quad o + e \rightarrow e$$

Can they satisfy phase matching condition in BBO?



## **Example: SHG phase matching in BBO**

Phase matching condition:

$$e + e \rightarrow O \quad k_{e}(\omega) + k_{e}(\omega) - k_{o}(2\omega) = 0 \Leftrightarrow \frac{\omega c_{0}}{n_{e}(\omega)} + \frac{\omega c_{0}}{n_{o}(\omega,\theta)} - \frac{2\omega c_{0}}{n_{o}(2\omega)} = 0 \Leftrightarrow n_{e}(\omega,\theta) = n_{o}(2\omega)$$

$$o + e \rightarrow O \quad k_{o}(\omega) + k_{e}(\omega) - k_{o}(2\omega) = 0 \Leftrightarrow \frac{\omega c_{0}}{n_{o}(\omega)} + \frac{\omega c_{0}}{n_{e}(\omega,\theta)} - \frac{2\omega c_{0}}{n_{o}(2\omega)} = 0 \Leftrightarrow \frac{1}{n_{o}(\omega)} + \frac{1}{n_{e}(\omega,\theta)} = \frac{2}{n_{o}(2\omega)}$$

$$o + e \rightarrow e \quad k_{o}(\omega) + k_{e}(\omega) - k_{e}(2\omega) = 0 \Leftrightarrow \frac{\omega c_{0}}{n_{o}(\omega)} + \frac{\omega c_{0}}{n_{e}(\omega,\theta)} - \frac{2\omega c_{0}}{n_{e}(2\omega,\theta)} = 0 \Leftrightarrow \frac{1}{n_{o}(\omega)} + \frac{1}{n_{e}(\omega,\theta)} = \frac{2}{n_{e}(2\omega,\theta)}$$
BBO is negative uniaxial crystal.
$$n_{e}(\omega,\theta) < n_{o}(2\omega) \quad \frac{1}{n_{e}(\omega,\theta)} + \frac{1}{n_{e}(\omega,\theta)} < \frac{2}{n_{e}(2\omega,\theta)}$$
Therefore  $e + e \rightarrow 0$  or  $e + e \rightarrow 0$  are not allowed. Only  $o + o \rightarrow e$  and  $o + e \rightarrow e$  can take place.
For positive uniaxial crystal,
$$e + e \rightarrow 0 \quad o + e \rightarrow 0$$

$$e \rightarrow e \quad o + e \rightarrow e \quad are allowed \quad o + e \rightarrow e \quad are forbidden.$$

## Acceptance angle (angular phase-matching bandwidth)

Phase matching using birefringence requires to align the input optical beam at some angle with respect to the crystal's optical axis. How accurately the angle should be? Let  $\theta_{pm}$  be the phase matching angle.

$$\Delta k(\theta_{pm}) = 0$$
 sin  $c^2(\frac{\Delta kL}{2}) = 1$  *L* is the crystal length.

We define acceptance angle  $\Delta \theta_{pm}$  be the phase matching angle such that:

$$\Delta k(\theta_{pm} + \Delta \theta_{pm})L = \pi \qquad \sin c^2(\frac{\pi}{2}) \approx 0.4$$

The SHG power drops by 60% from its peak value achieved at phase matching angle.

For type I phase matching:

$$\Delta k(\theta_{pm} + \Delta \theta_{pm}) = \frac{2\omega}{c_0} [n_o(\omega) - n_e(2\omega, \theta_{pm} + \Delta \theta_{pm})] = \frac{2\omega}{c_0} [n_o(\omega) - n_e(2\omega, \theta_{pm} + \Delta \theta_{pm}) - \frac{\partial n_e(2\omega, \theta)}{\partial \theta} \bigg|_{\theta_{pm}} \Delta \theta_{pm}]$$

After some mathematics, we have

$$\Delta \theta_{pm} \approx \frac{\omega}{4Lc_0} \frac{1}{[n_e(2\omega) - n_o(2\omega)]\sin 2\theta_{pm}} = \frac{\lambda_0}{4L} \frac{1}{[n_e(2\omega) - n_o(2\omega)]\sin 2\theta_{pm}}$$



### **Phase matching: critical Vs noncritical**

$$\Delta \theta_{pm} \approx \frac{\omega}{4Lc_0} \frac{1}{[n_e(2\omega) - n_o(2\omega)]\sin 2\theta_{pm}} = \frac{\lambda_0}{4L} \frac{1}{[n_e(2\omega) - n_o(2\omega)]\sin 2\theta_{pm}}$$
(1)

SHG from 800 nm light using 1 mm BBO crystal:  $\lambda_0 = 800 nm \quad \theta_{pm} = 29.2^{\circ} \quad n_o(400 nm) = 1.6903 \quad n_e(400 nm) = 1.5679$  $\Delta \theta_{pm} = 3.28 mrad \approx 0.2^{\circ}$ 

There is a trade-off between the acceptance angle and crystal length. For a fixed crystal length, acceptance angle limits how tight we can focus the incident beam onto the crystal—smaller focal spot leads to larger divergence angle.

One specific case:  $\theta_{pm} = 90^{\circ}$ , Eq. (1) diverges, and we need to use higher order term in the Taylor expansion.

After some mathematics, we have  $\Delta \theta_{pm} \approx \left\{ \frac{\lambda_0}{4L} \frac{1}{\left| n_e(2\omega) - n_o(2\omega) \right|} \right\}^{1/2}$  In this case, the acceptance angle

is normally one order of magnitude larger. Phase matching might be achieved by temperature tuning.

For phase matching with  $\theta_{pm} \neq 90^{\circ}$ , acceptance angle (or angular phase-matching bandwidth) is smaller. We call this type of birefringence-enabled phase matching as <u>critical phase matching</u>.

In contrast, phase matching with  $\theta_{pm} = 90^{\circ}$ , acceptance angle (or angular phase-matching bandwidth) is larger. We call this type of birefringence-enabled phase matching as <u>noncritical phase</u> <u>matching</u>.

## Phase matching: type I Vs. type II

In general, second-order nonlinear effects involve three waves with frequencies linked by the equation

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

Here  $\omega_3$  is the highest frequency of the three.

#### Type I phase matching:

 $\mathcal{O}_1$  wave and  $\mathcal{O}_2$  wave have the same polarization; that is, they are both ordinary waves or extraordinary waves:

 $o + o \rightarrow e$  or  $e + e \rightarrow o$ 

#### Type II phase matching:

 $\mathcal{O}_1$  wave and  $\mathcal{O}_2$  wave have different polarization:

$$o + e \rightarrow e \quad o + e \rightarrow o$$

 $e + o \rightarrow o \quad e + o \rightarrow e$ 



TABLE 2.3.2 Phase-matching methods for uniaxial crystals

	Positive uniaxial	Negative uniaxial	
	$(n_e > n_0)$	$(n_e < n_0)$	
Туре І	$n_3^o\omega_3 = n_1^e\omega_1 + n_2^e\omega_2$	$n_3^e \omega_3 = n_1^o \omega_1 + n_2^o \omega_2$	
Type II	$n_3^{\bar{o}}\omega_3 = n_1^{\bar{o}}\omega_1 + n_2^{\bar{e}}\omega_2$	$n_3^{\bar{e}}\omega_3 = n_1^{\bar{e}}\omega_1 + n_2^{\bar{o}}\omega_2$	

Robert Boyd, *Nonlinear optics*, chapter 2

## Type 0 phase matching to maximize nonlinearity

$$d_{np} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & d_{16} \\ d_{16} & -d_{16} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

For BBO crystal:

 $d_{16}(1.064um) = 2.2 \, pm/V$   $d_{15}(1.064um) = 0.03 \, pm/V$ 

 $d_{31}(1.064um) = 0.04 \, pm/V \quad d_{33}(1.064um) = 0.04 \, pm/V$ 

LiNbO<sub>3</sub> belongs to the same group:  $d_{16}(1.064um) = -2.1pm/V$   $d_{15}(1.064um) = -4.3pm/V$  $d_{31}(1.064um) = -4.3pm/V$   $d_{33}(1.064um) = -27.2pm/V$ 

To use  $d_{33}$  to maximize the nonlinearity, we need to align the E-field along z. Take SHG as an example:

$$P_x(2\omega) = 0$$
  $P_y(2\omega) = 0$   $P_z(2\omega) = 2\varepsilon_0 d_{33} E^2(\omega)$ 

The generated SHG wave is e wave, so we end up with

 $e + e \rightarrow e$  When all waves are in the same polarization, we can it Type 0 phase matching.

Perfect phase matching  $\Delta k = 0$  is impossible.



## Quasi phase matching (QPM) by periodic arrangement of nonlinearity



## Quasi phase matching (QPM) by periodic arrangement of nonlinearity

When the mismatched phase accumulates to  $\pi$ ,  $3\pi$ ,  $5\pi$ ..., energy starts to flow back from SH to the fundamental. <u>How about we shut down nonlinearity to avoid this back conversion?</u>



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## **QPM** by periodically poled LiNbO<sub>3</sub> (PPLN)



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## Homemade **QPM**

GaAs has a second-order nonlinear coefficient about 7 times larger than  $\text{LiNbO}_{3.}$  However it is an isotropic medium, and thus we cannot use birefringence enabled phase matching.  $\text{LiNbO}_{3}$  works up to 5 um and GaAs is transparent up to 17 um.

$$d_{np} = \begin{bmatrix} 0 & 0 & 0 & d_{36} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{36} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix}$$
$$d_{36}(1.064um) = 170 \, pm/V$$

Orientation patterned GaAs (OP-GaAs) or GaP (OP-GaP): material growing with opposite orientation.





## **Summary of phase matching conditions**



SHG intensity

#### Interactions between Light Waves in a Nonlinear Dielectric\*

J. A. ARMSTRONG, N. BLOEMBERGEN, J. DUCUING,<sup>†</sup> AND P. S. PERSHAN Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts (Received April 16, 1962)

The induced nonlinear electric dipole and higher moments in an atomic system, irradiated simultaneously by two or three light waves, are calculated by quantum-mechanical perturbation theory. Terms quadratic and cubic in the field amplitudes are included. An important permutation symmetry relation for the nonlinear polarizability is derived and its frequency dependence is discussed. The nonlinear microscopic properties are related to an effective macroscopic nonlinear polarization, which may be incorporated into Maxwell's equations for an infinite, homogeneous, anisotropic, nonlinear, dielectric medium. Energy and power relationships are derived for the nonlinear dielectric which correspond to the Manley-Rowe relations in the theory of parametric amplifiers. Explicit solutions are obtained for the coupled amplitude equations, which describe the interaction between a plane light wave and its second harmonic or the interaction between three plane electromagnetic waves, which satisfy the energy relationship  $\omega_3 = \omega_1 + \omega_2$ , and the approximate momentum relationship  $\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2 + \Delta \mathbf{k}$ . Third-harmonic generation and interaction between more waves is mentioned. Applications of the theory to the dc and microwave Kerr effect, light modulation, harmonic generation, and parametric conversion are discussed.

## The 22-page paper describes most of the basic principles of nonlinear optics as we know it today.

Then why the field is still growing?

New laser technology, novel optical materials, and emerging applications.

## **Take-home message**

- Coupled wave equations describe the wave-mixing process.
- Phase matching is critical in maximizing the power conversion efficiency in the wave-mixing process.
- Phase matching can be achieved using birefringence in an anisotropic medium.
- Quasi-phase matching allows type 0 phase matching to access to the largest tensor element.

## **Suggested reading**

## Coupled wave equation

-- Robert Boyd, *Nonlinear optics*, chapter 2

-- George Stegemann and Robert Stegemann, *Nonlinear* <u>optics</u>, chapter 2 (chapter 4 presents detailed analytical solution)

## Phase matching

-- Geoffrey New, Introduction to nonlinear optics, chapter 2

-- George Stegemann and Robert Stegemann, *Nonlinear* optics, chapter 3

-- Robert Boyd, *Nonlinear optics*, chapter 2