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# Second Harmonic Generation and related second order Nonlinear Optics

Nicolas Fressengeas

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## Second Harmonic Generation and related second order Nonlinear Optics

N. Fressengeas

Laboratoire Matériaux Optiques, Photonique et Systèmes  
Unité de Recherche commune à l'Université Paul Verlaine Metz et à Supélec

October 5, 2010



## Usefull reading. . .

[YY84, DGN91, LKW99]



V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan.  
*Handbook of Nonlinear Optical Crystals*, volume 64 of *Springer Series in Optical Sciences*.  
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*Optical waves in crystals. Propagation and control of laser radiation*.  
Wiley series in pure and applied optics. Wiley-Interscience, Stanford University, 1984.

# Contents

- 1 Three-wave interaction
  - Assumptions framework
  - Three Wave propagation equation
  - Sum frequency generation
  - Scalar approximation
- 2 Non Linear Optics Application
  - Second Harmonic Generation
  - Optical Parametric Amplifier
  - Optical Parametric Oscillator
- 3 Phase matching
  - Phase matching conditions
  - Phase matching in uni-axial crystals
  - Quasi-phase matching

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# A classical Maxwell framework

With standard assumptions: no charge, no current, no magnet and no conductivity

## Maxwell Model

- $\text{div}(D) = 0$
- $\text{div}(B) = 0$
- $\text{curl}(E) = -\frac{\partial B}{\partial t}$
- $\text{curl}(H) = \frac{\partial D}{\partial t}$

## Matter equations

- $D = \epsilon_0 E + P = \epsilon E$
- $B = \mu_0 H$
- $P = \epsilon_0 \chi_L E + P_{NL}$

Wave equation in isotropic medium

$$\Delta E - \mu_0 \epsilon \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$$

Solutions to be found only in a specific framework:

We look here for :

- Quadratic non-linearity
- Three wave interaction

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# Three wave interaction assumptions

All waves are transverse plane waves propagating in the  $z$  direction

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- Wave equation :  $\frac{\partial^2 E}{\partial z^2} - \mu_0 \varepsilon \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}(z)}{\partial t^2}$

Quadratic non-linearity

- $P_{NL}$  is transversal
- $[P_{NL}]_i = \sum_{\{j,k\} \in \{x,y\}^2} [d]_{ijk} [E]_j [E]_k = [d]_{ijk} [E]_j [E]_k$

Three wave interaction only

- Three waves only are present :  $\omega_1$ ,  $\omega_2$  and  $\omega_3$
- Non linear interaction of two waves : sum, difference, doubling, rectification. . .
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# Three wave interaction solution ansatz

## Sum of three waves

- $[E]_{x,y}(z, t) = \mathcal{R}e \left( \sum_{\nu=1}^3 \exp(ik_{\nu}z - i\omega_{\nu}t) \right)$
- Dispersion law :  $k_{\nu}^2 = \mu_0 \epsilon_{\nu} \omega_{\nu}^2$
- $\epsilon$  dispersion :  $\epsilon_{\nu} = \epsilon(\omega_{\nu})$

## How good is this ansatz ?

- We have assumed  $\omega_1 + \omega_2 = \omega_3$
- Why would  $\omega_3$  not be a source as well ?
- OK if wave 3 is small enough

## Separate investigation at each frequency

- At  $\omega_{\nu}$ , consider only the part of  $P_{NL}$  oscillating at frequency  $\omega_{\nu}$  :  $P_{NL}^{\omega_{\nu}}$
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Wave propagation for frequency  $\omega_\nu$ 

$\nu \in \{1, 2, 3\}$

## 3 Perturbed wave equations

$$\frac{\partial^2 E^{(\omega_\nu)}(z,t)}{\partial z^2} - \mu_0 \epsilon_{\omega_\nu} \frac{\partial^2 E^{(\omega_\nu)}(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}^{\omega_\nu}(z,t)}{\partial t^2}$$

## Temporal harmonic notation

- $E^{(\omega_\nu)}$  is a plane wave
- Non linear polarization is a plane wave
- Considering only... the  $\omega_\nu$  part  $\Leftrightarrow$  the plane wave part

$$\bullet \frac{\partial^2 E^{(\omega_\nu)}(z,t)}{\partial z^2} + \mu_0 \epsilon_{\omega_\nu} \omega_\nu^2 E^{(\omega_\nu)}(z,t) = -\mu_0 \omega_\nu^2 P_{NL}^{\omega_\nu}(z,t)$$

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# Non Linear Polarisation $P_{NL}$ in harmonic framework

## Reminder

$$[P_{NL}]_i = [d]_{ijk}[E]_j[E]_k$$

Temporal harmonic framework

for  $\omega_1$

- Multiply complex fields
- Include Conjugates to take Real Part
- Select only the  $\omega_1$  component

$$[P_{NL}^{\omega_1}(z, t)]_i = \mathcal{R}e \left( [d]_{ijk} [E^{(\omega_3)}(z)]_j [\overline{E^{(\omega_2)}(z)}]_k e^{i(k_3 - k_2)z - i(\omega_3 - \omega_2)t} \right)$$

Wave propagation equation

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# The Slow Varying Approximation

Closely related to the paraxial approximation

## The Slow Varying Approximation

- Beam envelope is assumed to vary slowly in the longitudinal direction
- Equivalent as assuming a narrow beam
- Second derivative with respect  $z$  neglected compared to
  - the first one with respect to  $z$
  - the others with respect to  $x$  and  $y$

To put it in maths...

- $\frac{\partial^2 E^{(\omega_1)}(z,t)}{\partial z^2} = \frac{\partial^2}{\partial z^2} \text{Re} (E^{(\omega_1)}(z) \exp(i(k_1 z - \omega_1 t)))$
- $\dots = \text{Re} \left( \left[ \frac{\partial^2 E^{(\omega_1)}(z)}{\partial z^2} + 2ik_1 \frac{\partial E^{(\omega_1)}(z)}{\partial z} - k_1^2 E^{(\omega_1)}(z) \right] e^{i(k_1 z - \omega_1 t)} \right)$

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# Wave Propagation Equation under SVA approximation

Obtaining an envelope equation, which is simpler

## Non SVA wave propagation equation

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## SVA equation

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- $+ \text{Re} \left( \mu_0 \varepsilon_{\omega_1} \omega_1^2 E^{(\omega_1)}(z) e^{i(k_1 z - \omega_1 t)} \right)$
- $= \text{Re} \left( -\mu_0 \omega_1^2 [d]_{ijk} [E^{(\omega_3)}(z)]_j [E^{(\omega_2)}(z)]_k e^{i(k_3 - k_2)z - i(\omega_3 - \omega_2)t} \right)$

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$$\frac{\partial^2 E^{(\omega_1)}(z, t)}{\partial z^2} + \mu_0 \varepsilon_{\omega_1} \omega_1^2 E^{(\omega_1)}(z, t) = -\mu_0 \omega_1^2 P_{NL}^{\omega_1}(z, t)$$

## SVA equation

- $\mathcal{R}e \left( \left[ 2ik_1 \frac{\partial E^{(\omega_1)}(z)}{\partial z} - k_1^2 E^{(\omega_1)}(z) \right] e^{i(k_1 z - \omega_1 t)} \right)$
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# The three waves

## Phase mismatch and dispersion relationship

- Phase mismatch :  $\Delta k = k_1 + k_2 - k_3$
- Recall the dispersion relationship :  $k_1^2 = \mu_0 \varepsilon_{\omega_1} \omega_1$
- Wave impedance :  $\eta_\nu = \sqrt{\frac{\mu_0}{\varepsilon_{\omega_\nu}}}$

## Three wave propagation, rotating $i \rightarrow j \rightarrow k$

$$(i, j, k) \in \{x, y\}^3$$

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## 6 equations for various quadratic phenomena

All in one for : frequency sum and difference, second harmonic generation and optical rectification, parametric amplifier...

### Six equations

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Why all those names ?

They differ by :

- The input frequencies and the generated ones
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# Example : sum frequency generation

Input beams assumed constant

Undepleted pump approximation

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$$y' = ae^{(ibx)} \Rightarrow y = \frac{ia}{b} (1 - e^{(ibx)})$$

Wave solution  $[E(\omega_3)]_j$

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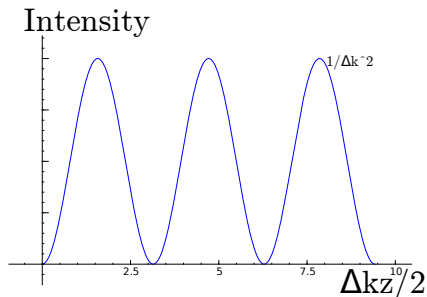
# Phase match or not phase match

Phase matching is a key issue to sum frequency generation

Phase mismatch

$$\Delta k \neq 0$$

- Oscillating intensity
- Max intensity  $\propto \frac{1}{\Delta k^2}$



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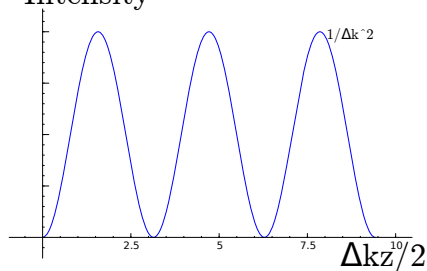
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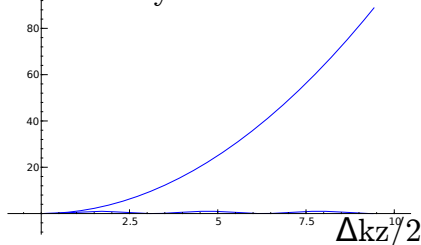


## Phase match

$$\Delta k = 0$$

- Intensity quadratic increase
- Approximations do not hold long

Intensity





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# A Scalar Three Wave Interaction model

Further approximations to remove vectors

## Simplifying notations

- Set indexes equal for polarization and frequency:  $A_\nu = [E^{(\omega_\nu)}]_\nu$
- Consider  $\varepsilon_\nu = n_\nu^2 \varepsilon_0$
- Abbreviate  $C = \sqrt{\frac{\mu_0}{\varepsilon_0}} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}$
- For lossless media,  $d$  is isotropic.
- Assume  $d$  is frequency independent
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- Assume  $d$  is frequency independent
- Let  $K = dC/2$

## Scalar three wave interaction

- $\frac{\partial A_1}{\partial z} = +iK \overline{A_2} A_3 \exp(-i\Delta kz)$
- $\frac{\partial A_2}{\partial z} = -iK A_1 \overline{A_3} \exp(+i\Delta kz)$
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# A Scalar Three Wave Interaction model

Further approximations to remove vectors

## Simplifying notations

- Set indexes equal for polarization and frequency:  $A_\nu = [E^{(\omega_\nu)}]_\nu$
- Consider  $\varepsilon_\nu = n_\nu^2 \varepsilon_0$
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  - Phase matching conditions
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# Second Harmonic Generation

# SHG

Sum frequency generation of two equal frequencies from the same source

One input beam counts for two

- $\omega_1 = \omega_2, A_1 = A_2, k_1 = k_2,$   
 $\omega_3 = 2\omega_1$
- $\Delta k = 2k_1 - k_3$

2 remaining equations

- $\frac{\partial A_1}{\partial z} = +iK \bar{A}_1 A_3 \exp(-i\Delta kz)$
- $\frac{\partial A_3}{\partial z} = +iK A_1^2 \exp(+i\Delta kz)$

Phase matching

$$\Delta k = 0$$

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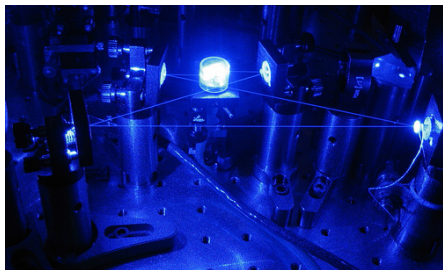


Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source flickr)

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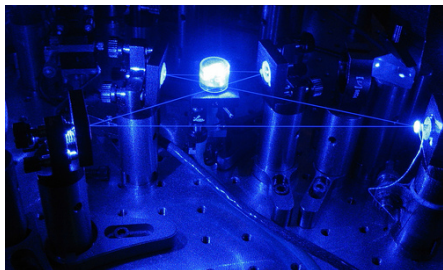


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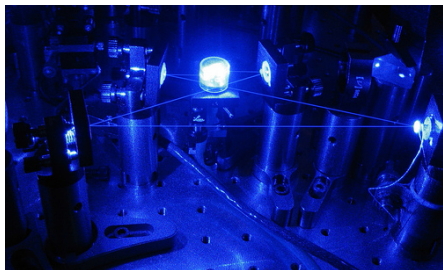


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# Second Harmonic Beam Generation

Remember...

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- $\frac{\partial A_1}{\partial z} = +iK \overline{A_1} A_3$
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$A_1 \in \mathbb{R}$

$A_3 \in i\mathbb{R}$

- $A_3 = i\tilde{A}_3 \Rightarrow \tilde{A}_3 \in \mathbb{R}$
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Real equations

- $\frac{\partial A_1}{\partial z} = -K \overline{A_1} \tilde{A}_3$
- $\frac{\partial \tilde{A}_3}{\partial z} = K A_1^2$

Multiply by  $A_1$  and  $\tilde{A}_3$

Sum

- $\frac{\partial (A_1^2 + \tilde{A}_3^2)}{\partial z} = 0$
- This is Energy Conservation

Start with no harmonic

$$(A_1^2(z) + \tilde{A}_3^2(z)) = A_1^2(0)$$

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$\tilde{A}_3$  equation

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$\tilde{A}_3$  expression

$$\tilde{A}_3(z) = A_1(0) \tanh(KA_1(0)z)$$

$I_3$  expression

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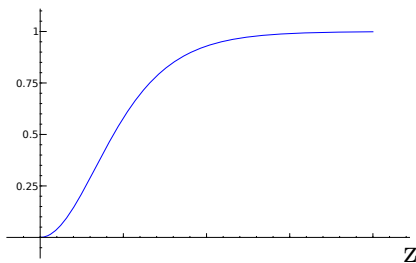
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$I_3(z) / I_1(0)$

$I_1(0) = \text{constant}$



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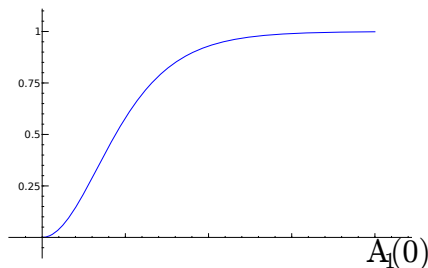
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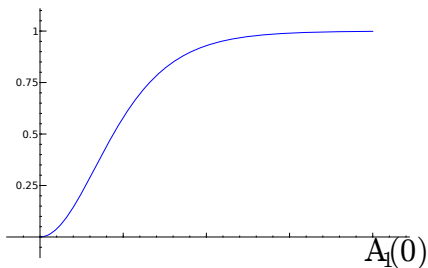
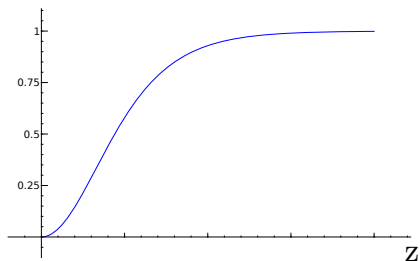
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# SHG conclusion

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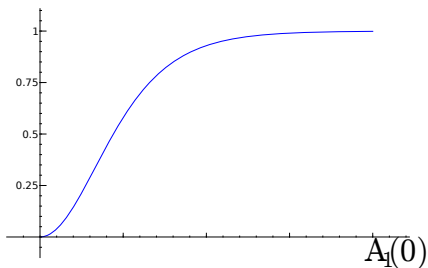
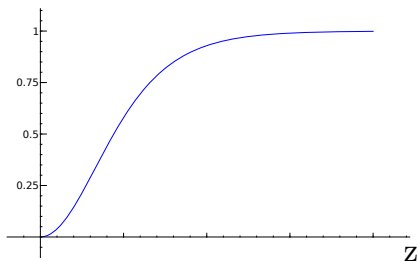


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- It is possible to convert 100% of a beam, with large interaction length or intensity
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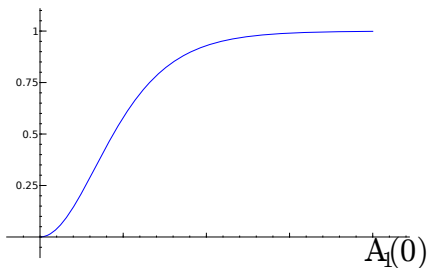
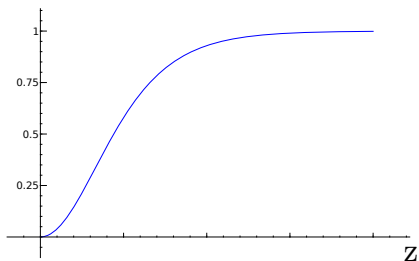


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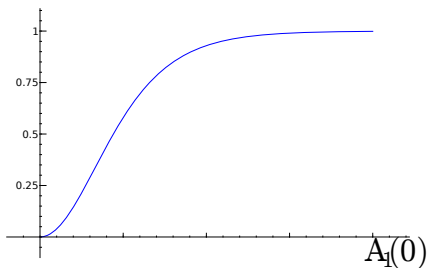
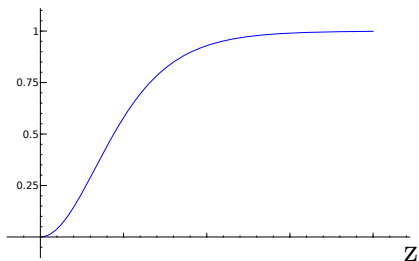


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# Optical Parametric Amplifier

# OPA

Optical Amplification of a weak signal beam thanks to a powerful pump beam

## Signal beam amplification

- $\omega_1$  : weak signal to be amplified
- $\omega_3$  : intense pump beam
- $\omega_2 = \omega_3 - \omega_1$  : difference frequency generation (idler)

## Undepleted pump approximation

$$A_3(z) = A_3(0) = K_p/K$$

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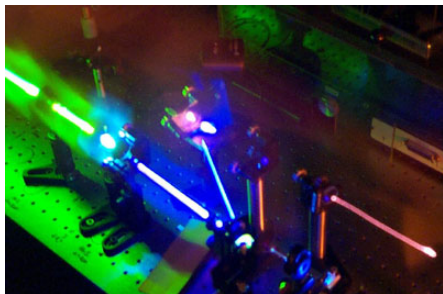


Figure: White light continuum seeded optical parametric amplifier (OPA) able to generate extremely short pulses. (source Freie Universität Berlin)

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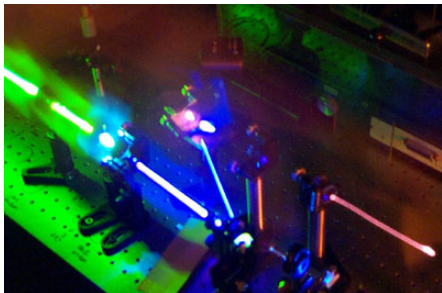


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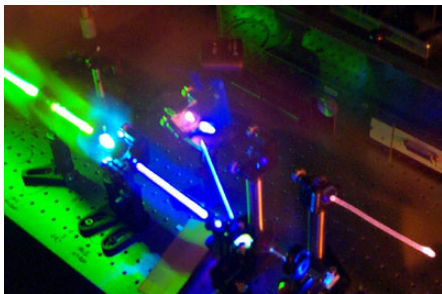


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## Initial conditions

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## Amplitude solution

- Amplified signal :  
 $A_1(z) = A_1(0) \cosh(K_p z)$
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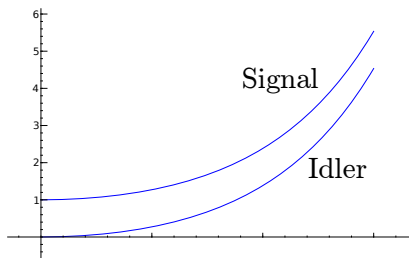
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# Optical Parametric Oscillator

# OPO

Use Optical Parametric Amplification to make a tunable laser

OPA pumped with  $\omega_3$

- Amplifier for  $\omega_1$  and  $\omega_2$
- With  $\omega_1 + \omega_2 = \omega_3$
- Phase matching:  $k_1 + k_2 = k_3$
- $\omega_1$  and  $\omega_2$  initiated from noise

Frequency tunable laser

- Get Non Linear Medium
- Adjust Cavity for  $\omega_1$  and  $\omega_2$
- Pump with  $\omega_3$
- You got it !

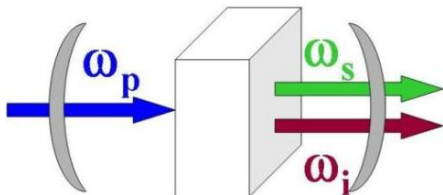


Figure: Optical Parametric Oscillator  
(source Cristal Laser)

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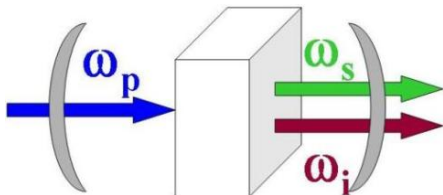


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# Colinear (scalar) phase matching

## Phase matching for co-propagation waves

- $k_1 + k_2 = k_3 \Rightarrow \omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3$
- for SHG :  $2k_1 = k_3 \Rightarrow n_1 = n_3$
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One and only solution

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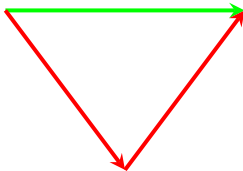
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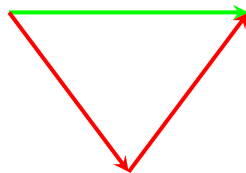
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# Non colinear phase matching

Use clever geometries  
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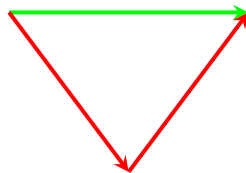


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Or even more clever. . .



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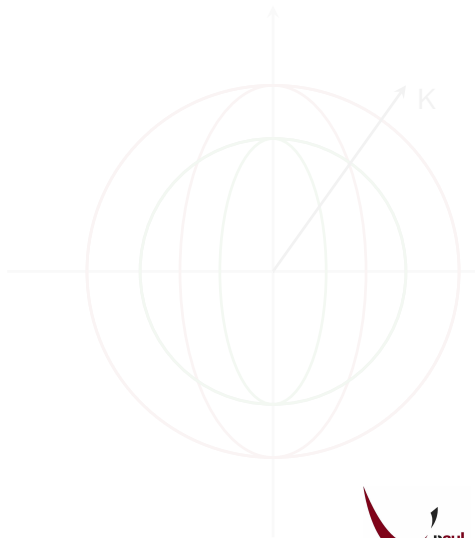
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## Waves polarization

- 1 incident wave counts for 2
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## Type I phase matching

- One refraction index for Fundamental
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- They must be equal
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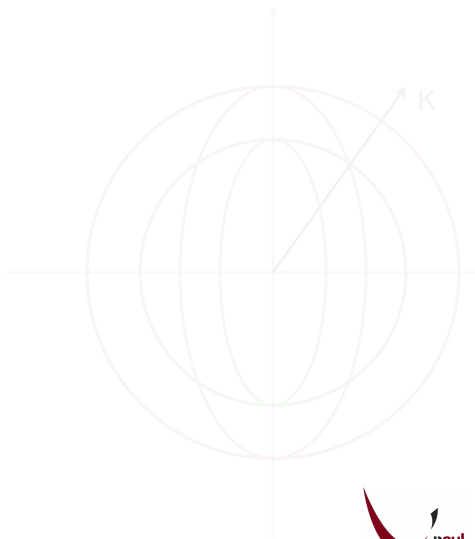
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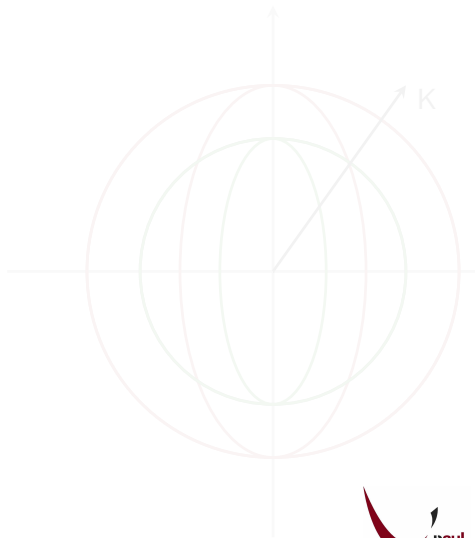
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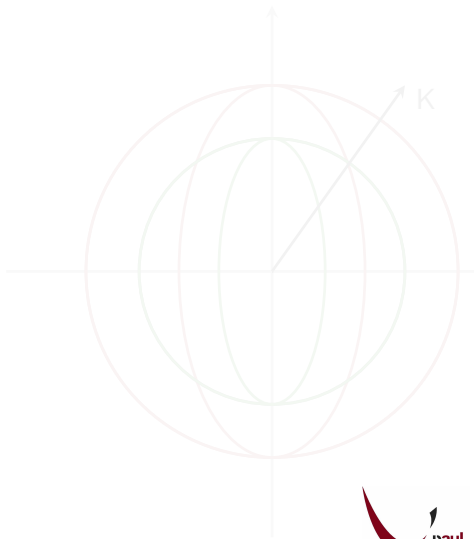
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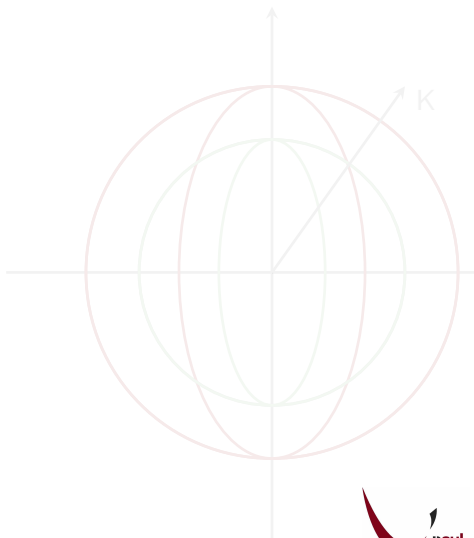
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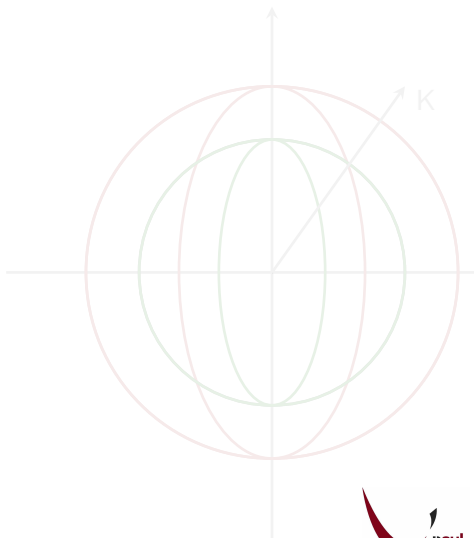
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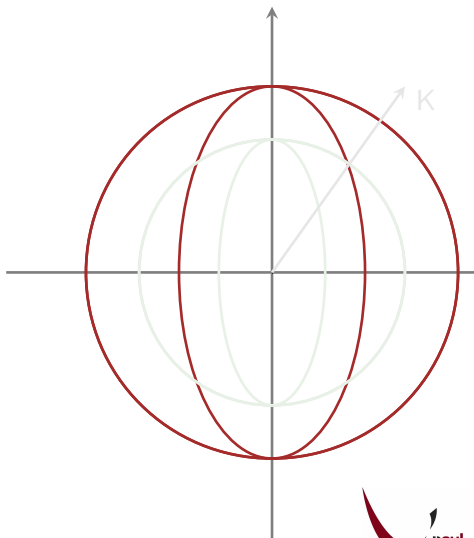
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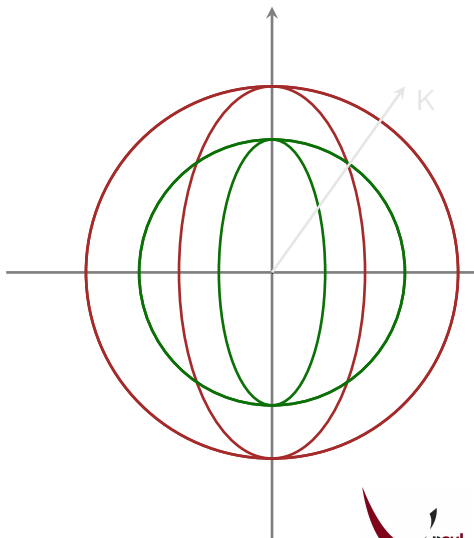
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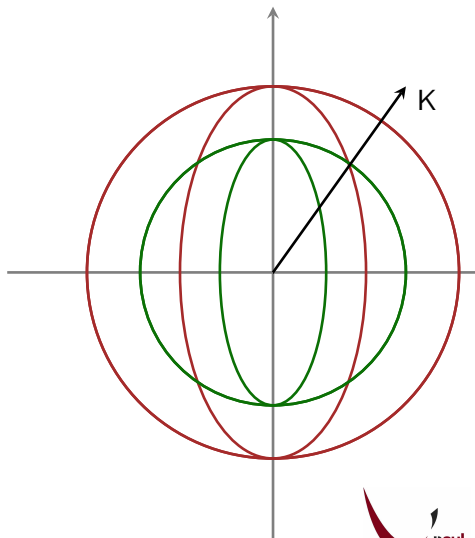
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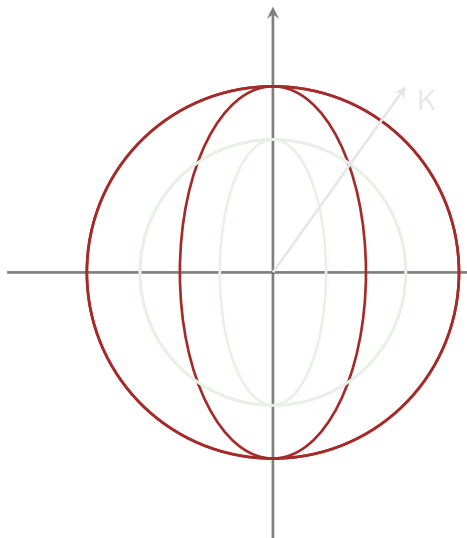
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# SHG Type I phase matching: a few numbers



Fundamental index ellipsoid section

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2(\theta)}{n_o^2} + \frac{\sin^2(\theta)}{n_e^2}$$

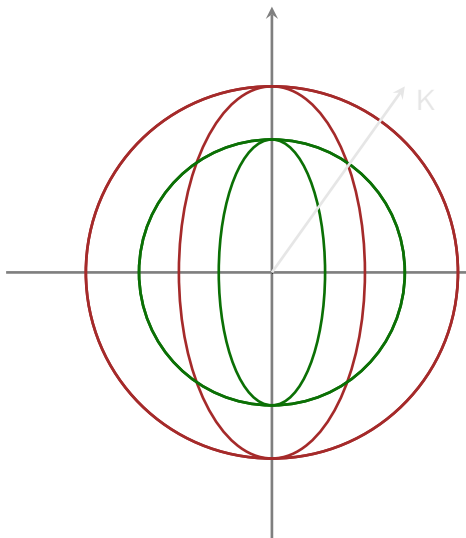
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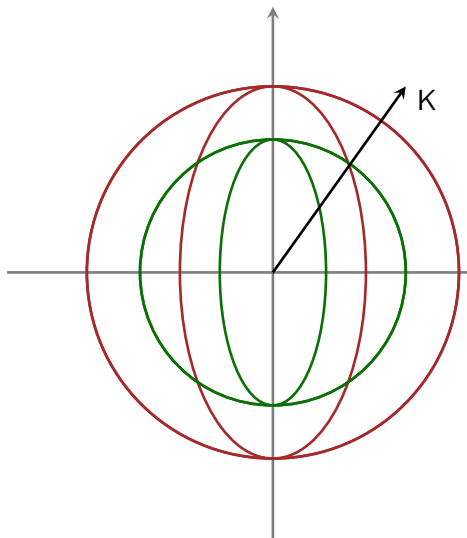
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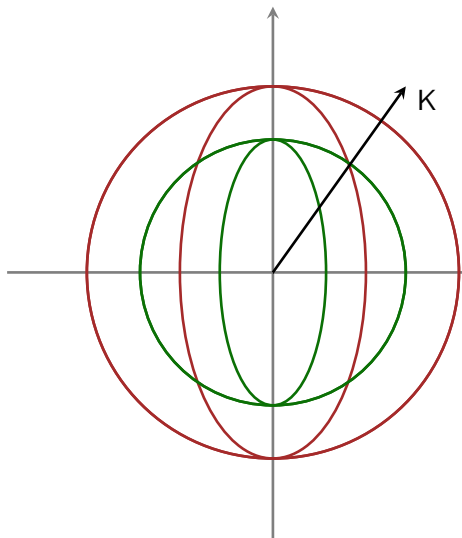
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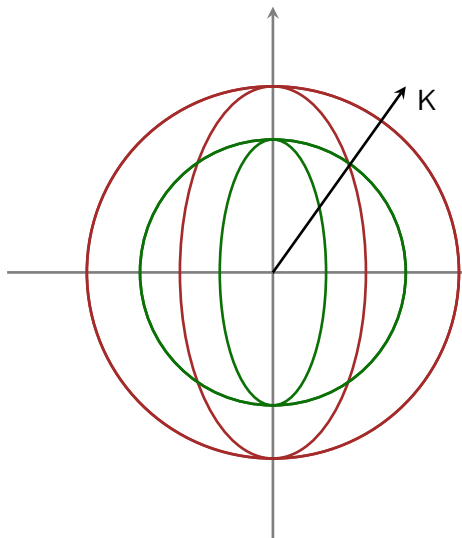
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In the three beam interaction, Type I was

- Both input beams  $\omega_1$  and  $\omega_2$  share the same polarization
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Another solution : Type II

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## A hard task

- Phase matching is seldom colinear
- Vector phase matching in a complex index ellipsoid
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Paper by *Bœuf* can help



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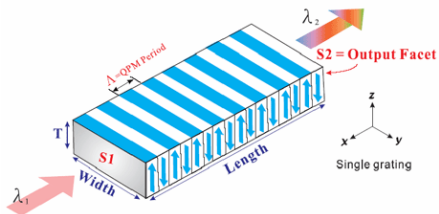
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# Quasi phase matching in layered media

## Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- $d$  sign reversal



## Wave solution

$$\left[ \frac{\partial E(\omega_3)}{\partial z} \right]_j$$

$$\frac{i\omega_3}{2} \eta_3 [d]_{jki} |E(\omega_1)|^2 \frac{e^{i\Delta k \Lambda} - 1}{\Delta k} \sum_{n=1}^N (-1)^n e^{i\Delta k \Lambda}$$

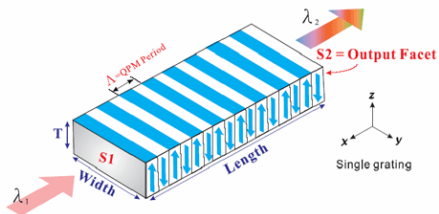
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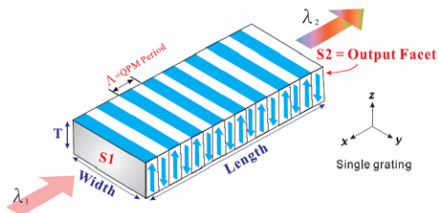
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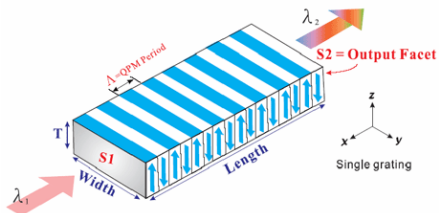
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