



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

November 4, 2011

Useful reading . . .

[YY84, DGN91, LKW99]



V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan.

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Wiley series in pure and applied optics. Wiley-Interscience, Stanford University, 1984.

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- 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

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

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

- Transversal E : has x and y components
- Wave equation : $\frac{\partial^2 E}{\partial z^2} - \mu_0 \epsilon \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}(z)}{\partial t^2}$

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 : ω_1 , ω_2 and ω_3
- Non linear interaction of two waves : sum, difference, doubling, rectification...
- We consider only those for which $\omega_1 + \omega_2 = \omega_3$

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Three wave interaction solution ansatz

Sum of three waves

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

How good is this ansatz ?

- We have assumed $\omega_1 + \omega_2 = \omega_3$
- Why would ω_3 not be a source as well ?
- OK if wave 3 is small enough

Separate investigation at each frequency

- At ω_{ν} , consider only the part of P_{NL} oscillating at frequency ω_{ν} : $P_{NL}^{\omega_{\nu}}$
- Three separate equations

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Wave propagation for frequency ω_ν

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

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 ω_ν part \Leftrightarrow the plane wave part
- $\frac{\partial^2 E^{(\omega_\nu)}(z,t)}{\partial z^2} + \mu_0 \epsilon_{\omega_\nu} \omega_\nu^2 E^{(\omega_\nu)}(z,t) = -\mu_0 \omega_\nu^2 P_{NL}^{\omega_\nu}(z,t)$

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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 ω_1

- Multiply complex fields
- Include Conjugates to take Real Part
- Select only the ω_1 component

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

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...

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

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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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Wave Propagation Equation under SVA approximation

Obtaining an envelope equation, which is simpler

Non SVA wave propagation equation

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

SVA equation

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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 : $k_1^2 = \mu_0 \epsilon_{\omega_1} \omega_1$
- Wave impedance : $\eta_\nu = \sqrt{\frac{\mu_0}{\epsilon_{\omega_\nu}}}$

Three wave propagation, rotating $i \rightarrow j \rightarrow k$ $(i, j, k) \in \{x, y\}^3$

- $\left[\frac{\partial E^{(\omega_1)}}{\partial z} \right]_i = +\frac{i\omega_1}{2} \eta_1 [d]_{ijk} [E^{(\omega_3)}]_j \overline{[E^{(\omega_2)}]}_k \exp(-i\Delta kz)$
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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
- The one that is the smallest and those which are large
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1 Three-wave interaction

- Assumptions framework
- Three Wave propagation equation
- **Sum frequency generation**
- Scalar approximation

2 Non Linear Optics Application

- Second Harmonic Generation
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3 Phase matching

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- Phase matching in uni-axial crystals
- Quasi-phase matching

Example : sum frequency generation

Input beams assumed constant

Undepleted pump approximation

Assumptions

- $\omega_1 + \omega_2 = \omega_3$
- Generated beam null at $z = 0$: $[E^{(\omega_3)}(z = 0)]_j = 0$
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One equation remains

$$\left[\frac{\partial E^{(\omega_3)}}{\partial z} \right]_j = +\frac{i\omega_3}{2} \eta_3 [d]_{jki} [E^{(\omega_2)}]_k [E^{(\omega_1)}]_i \exp(-i\Delta kz)$$

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Solving the SVA wave propagation equation

Equation to solve

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$$\Delta k \neq 0$$

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

Wave solution

$$[E^{(\omega_3)}]_j$$

$$\frac{i\omega_3}{2} \eta_3 [d]_{jki} [E^{(\omega_2)}]_k [E^{(\omega_1)}]_i \frac{e^{(i\Delta kz)} - 1}{i\Delta k}$$

Intensity

$$\propto [E^{(\omega_3)}]_j \overline{[E^{(\omega_3)}]_j}$$

$$\omega_3^2 \eta_3^2 [d^2]_{jki} |E^{(\omega_2)}|_k^2 |E^{(\omega_1)}|_i^2 \frac{2 \sin^2\left(\frac{\Delta kz}{2}\right)}{\Delta k^2}$$



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Phase match or not phase match

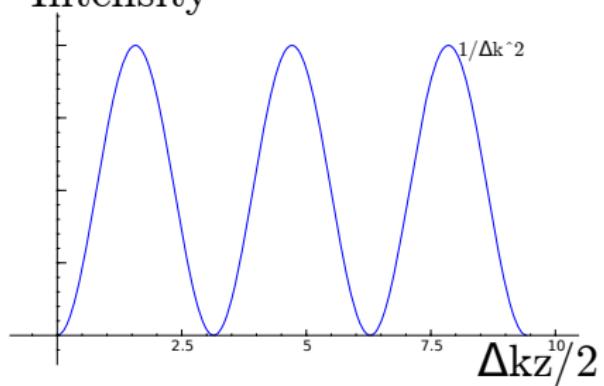
Phase matching is a key issue to sum frequency generation

Phase mismatch

$$\Delta k \neq 0$$

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

Intensity



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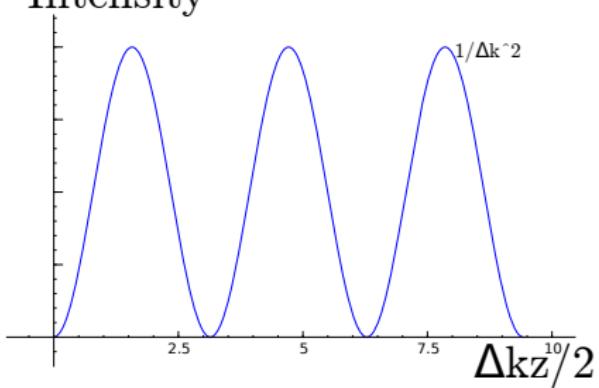
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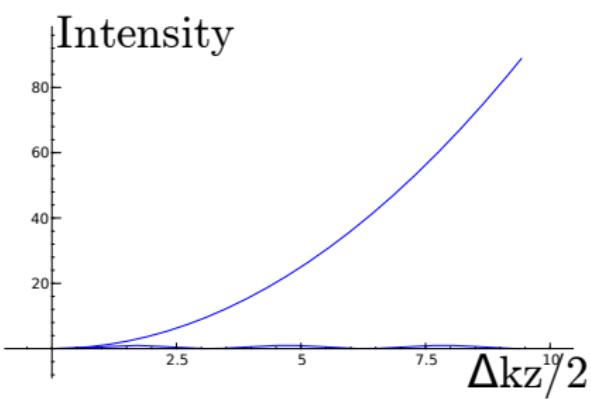


Phase match

$$\Delta k = 0$$

- 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 $\epsilon_\nu = n_\nu^2 \epsilon_0$
- Abbreviate $C = \sqrt{\frac{\mu_0}{\epsilon_0}} \sqrt{\frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}$
- For lossless media, d is isotropic.
- Assume d is frequency independent
- Let $K = dC/2$

Scalar three wave interaction

- $\frac{\partial A_1}{\partial z} = +iK\bar{A}_2 A_3 \exp(-i\Delta kz)$
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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

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

$$\Delta k = 0$$

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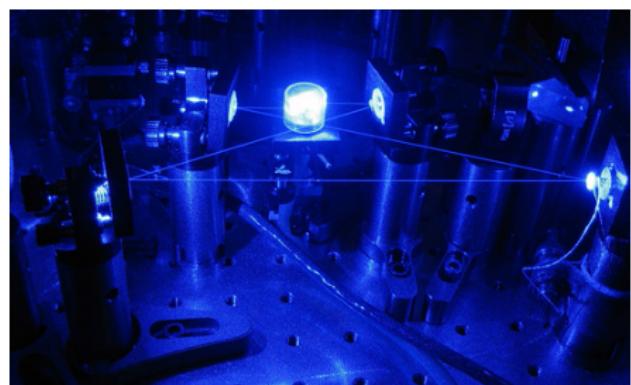


Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. ([source flickr](#))

Second Harmonic Generation

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

- $\frac{\partial A_1}{\partial z} = +iK\bar{A}_1 A_3 \exp(-i\Delta kz)$
- $\frac{\partial A_3}{\partial z} = +iKA_1^2 \exp(+i\Delta kz)$

Phase matching

$$\Delta k = 0$$

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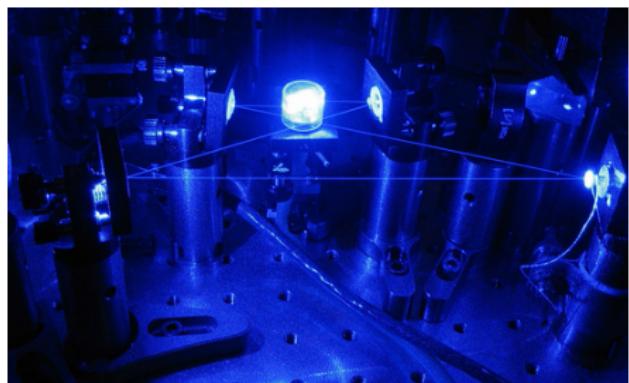


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Remember...

$$\Delta k = 0$$

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

- $\frac{\partial A_1}{\partial z} = -K\overline{A_1}\tilde{A}_3$
- $\frac{\partial \tilde{A}_3}{\partial z} = KA_1^2$

Multiply by A_1 and \tilde{A}_3

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

• 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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$$\frac{\partial \tilde{A}_3}{\partial z} = KA_1^2 = K(A_1^2(0) - \tilde{A}_3^2(z))$$

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I_3 expression

$$I_3(z) = I_1(0) \tanh^2(KA_1(0)z)$$

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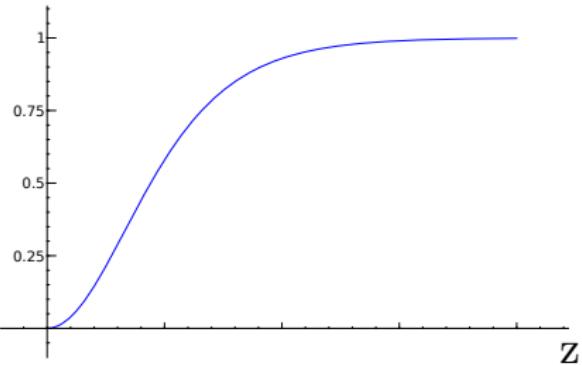
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$$I_3(z)/I_1(0) \quad I_1(0) = \text{constant}$$



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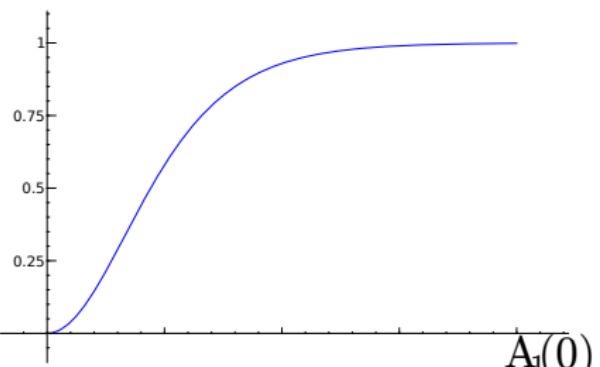
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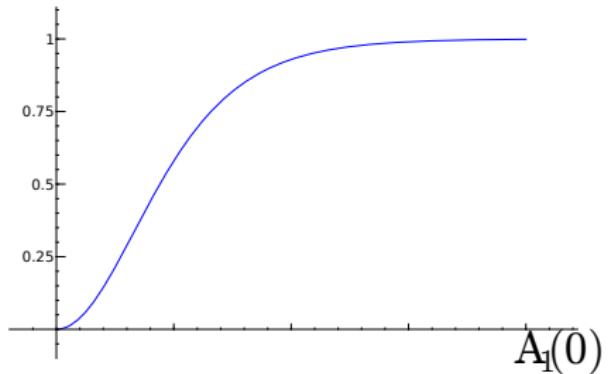
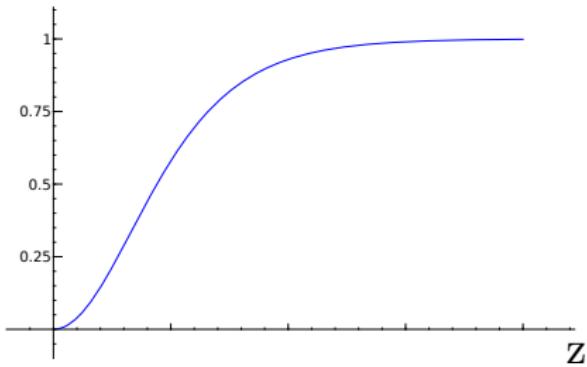
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SHG conclusion

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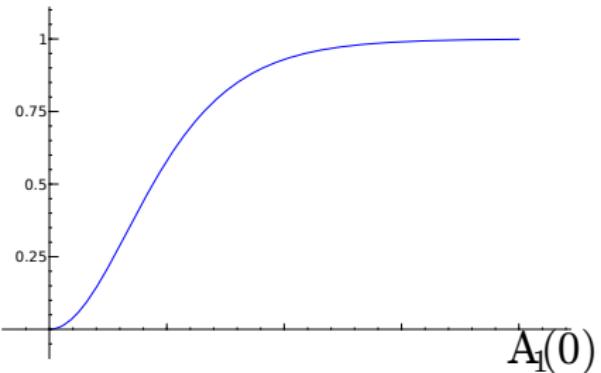
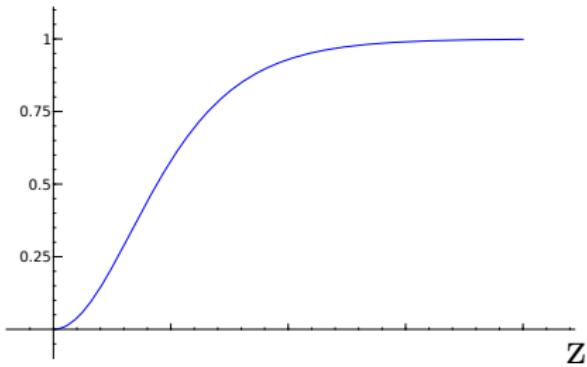


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- It is possible to convert 100% of a beam, with large interaction length or intensity
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- We have retrieved Energy Conservation in spite of drastic approximations

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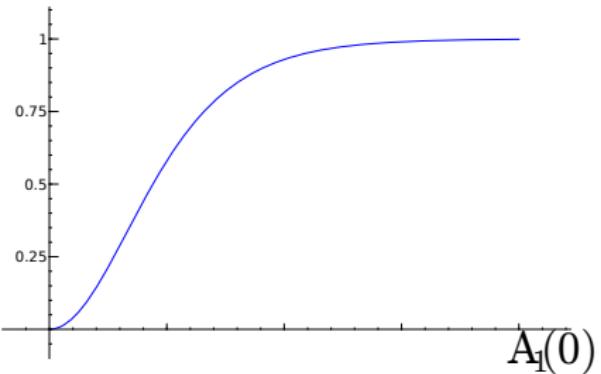
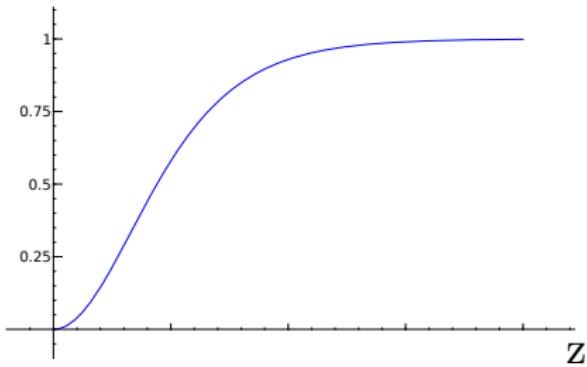


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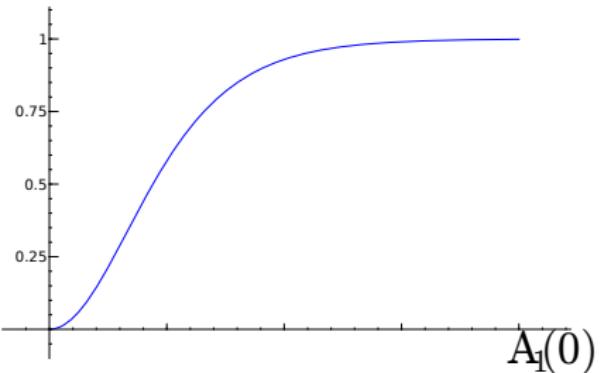
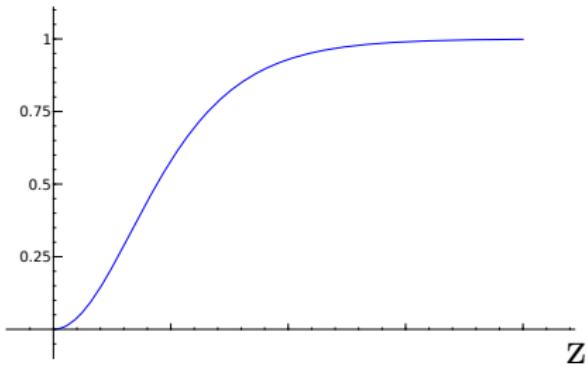


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1 Three-wave interaction

- Assumptions framework
- Three Wave propagation equation
- Sum frequency generation
- Scalar approximation

2 Non Linear Optics Application

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- **Optical Parametric Amplifier**
- Optical Parametric Oscillator

3 Phase matching

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Optical Parametric Amplifier

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

Signal beam amplification

- ω_1 : weak signal to be amplified
- ω_3 : intense pump beam
- $\omega_2 = \omega_3 - \omega_1$: difference frequency generation (idler)

Undepleted pump approximation

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

Phase matched equations

- $\frac{\partial A_1}{\partial z} = +iK_p \bar{A}_2$
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Figure: White light continuum seeded optical parametric amplifier (OPA) able to generate extremely short pulses.
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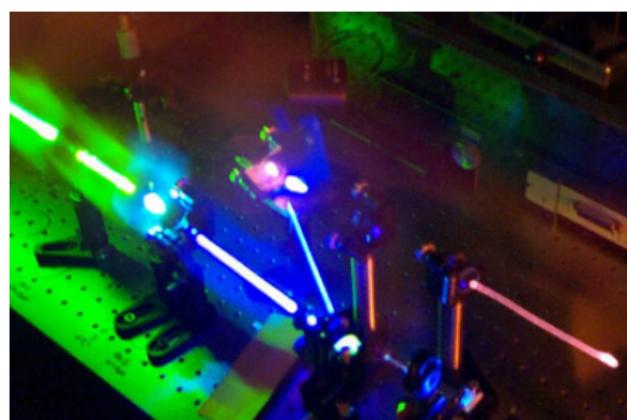


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Initial conditions

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- No idler : $\overline{A}_2(0) = 0$

Amplitude solution

- Amplified signal :
$$A_1(z) = A_1(0) \cosh(K_p z)$$
- Idler :
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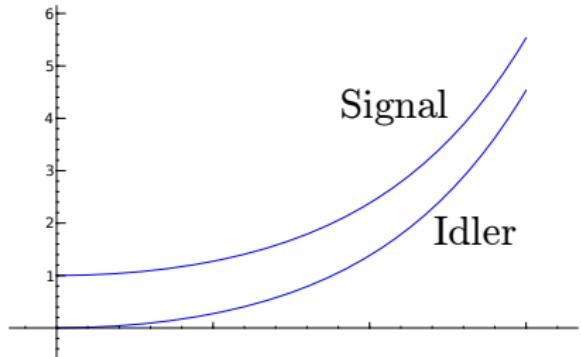
Intensities

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OPO

Use Optical Parametric Amplification to make a tunable laser

OPA pumped with ω_3

- Amplifier for ω_1 and ω_2
- With $\omega_1 + \omega_2 = \omega_3$
- Phase matching: $k_1 + k_2 = k_3$
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Frequency tunable laser

- Get Non Linear Medium
- Adjust Cavity for ω_1 and ω_2
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- You got it !

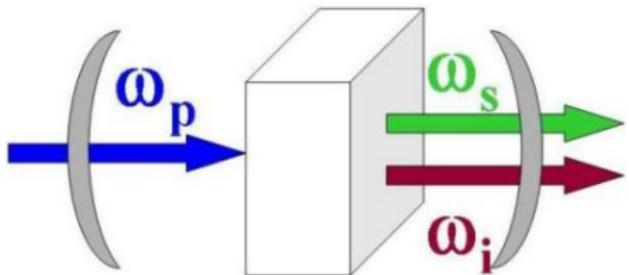


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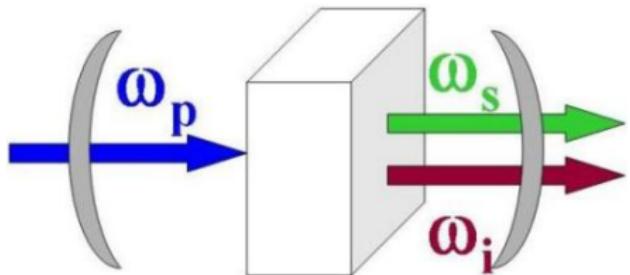


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Colinear (scalar) phase matching

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 : $2k_1 = k_3 \Rightarrow n_1 = n_3$
- The last is **never** achieved, due to normal dispersion: $n_1 < n_3$

One and only solution

Use birefringent crystals and different polarizations

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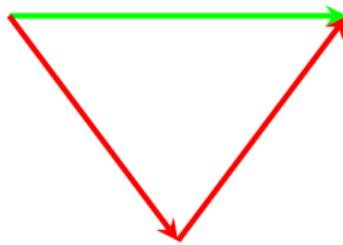
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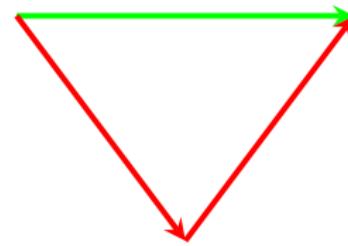
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Non colinear phase matching

Use clever geometries

With reflections

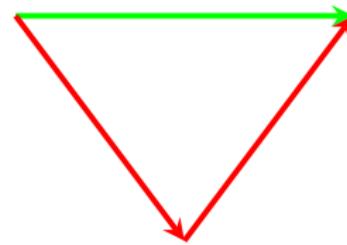


Non colinear phase matching

Use clever geometries

With reflections

Or even more clever...



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

SHG Type I Phase Matching

Waves polarization

- 1 incident wave counts for 2
- They share the same polarization
- Second Harmonic polarization is orthogonal

Type I phase matching

- One refraction index for Fundamental
- The other for Second Harmonic
- They must be equal
- Propagate in the right direction



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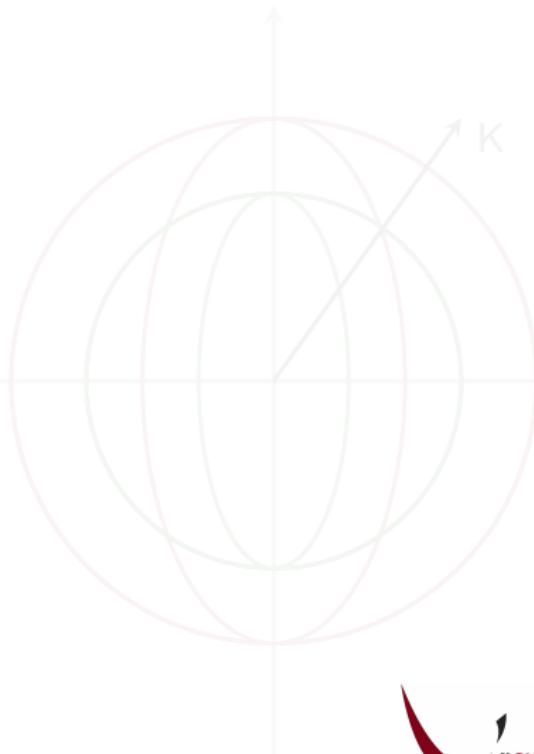
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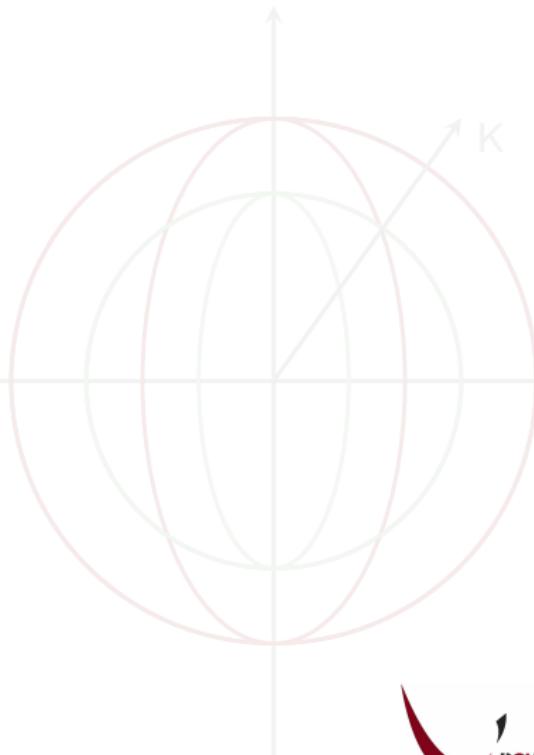
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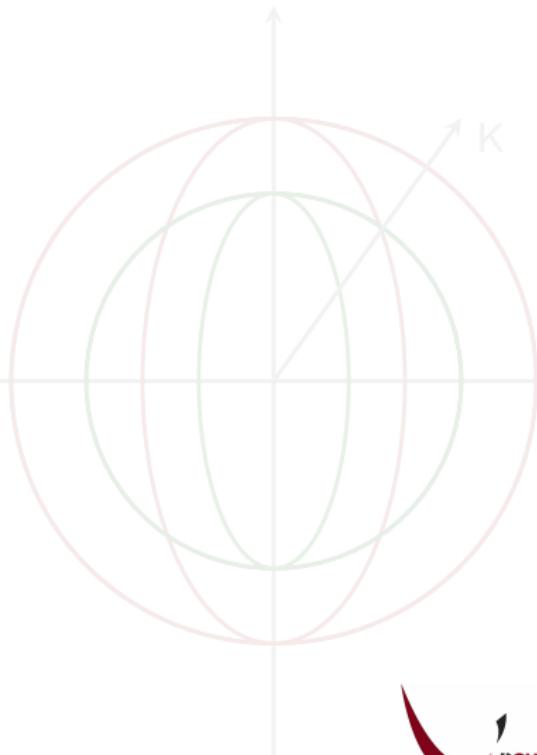
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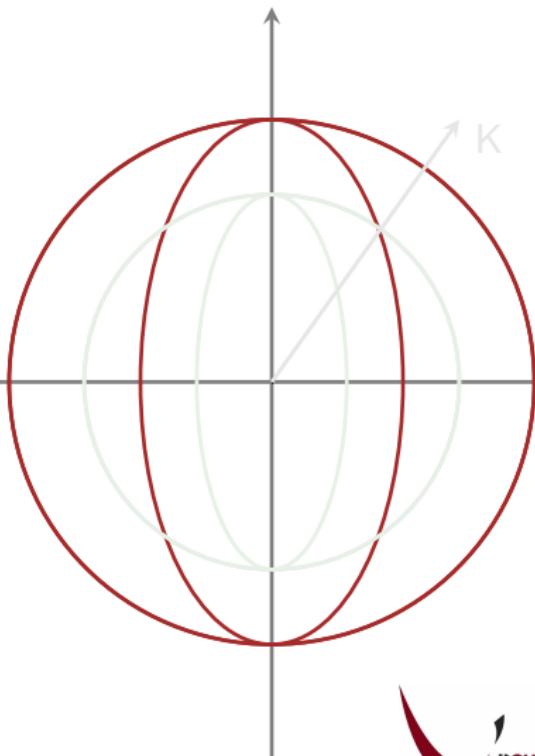
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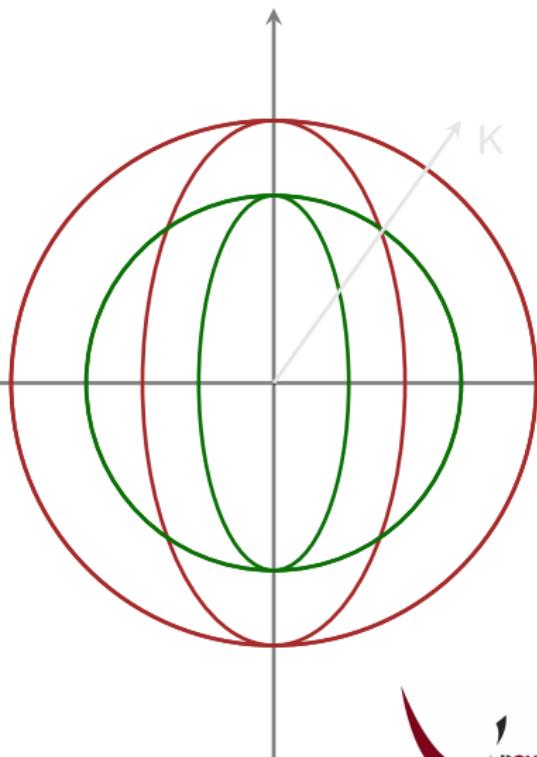
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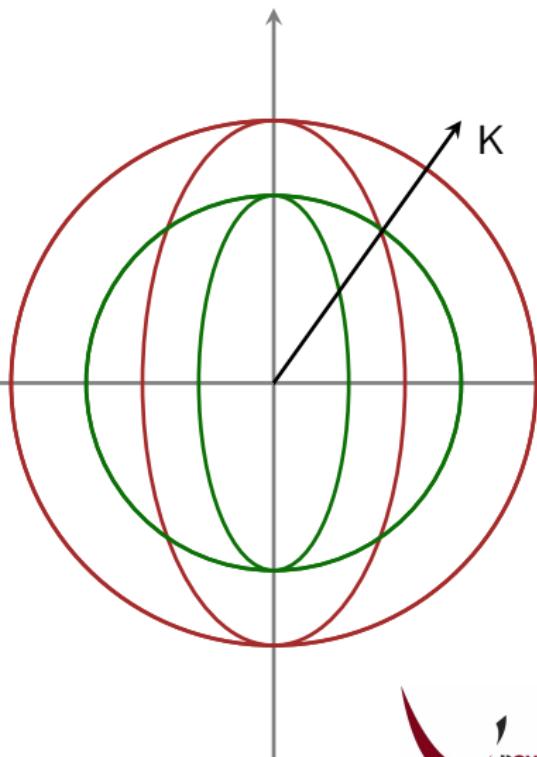
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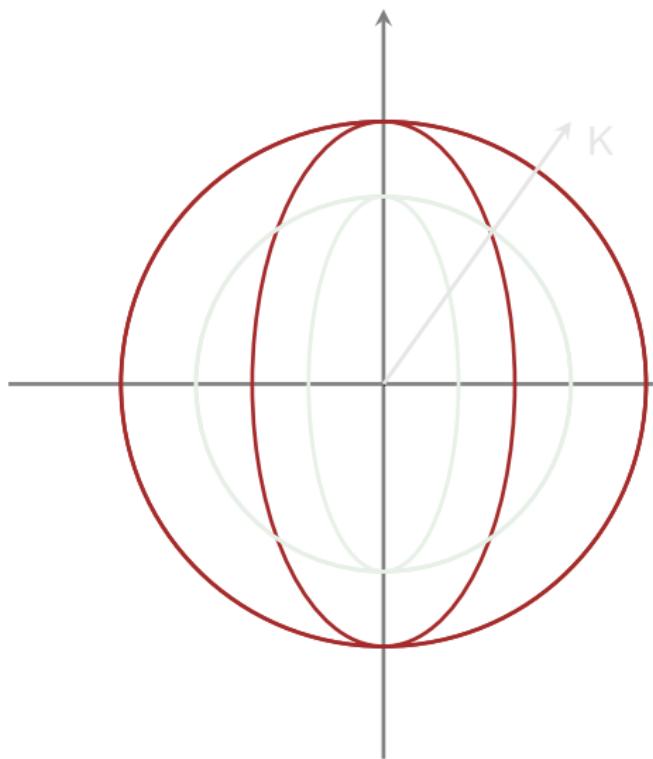
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SHG Type I phase matching: a few numbers



Fundamental index ellipsoid section

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

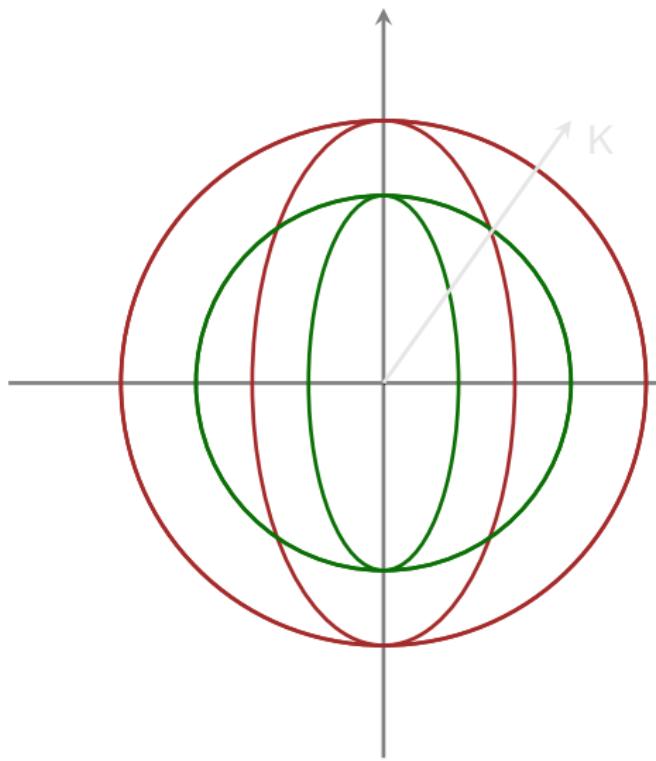
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Solve the equation

$$\sin^2(\theta) = \frac{n_o^{-2} - \tilde{n}_o^{-2}}{\tilde{n}_e^{-2} - \tilde{n}_o^{-2}}$$

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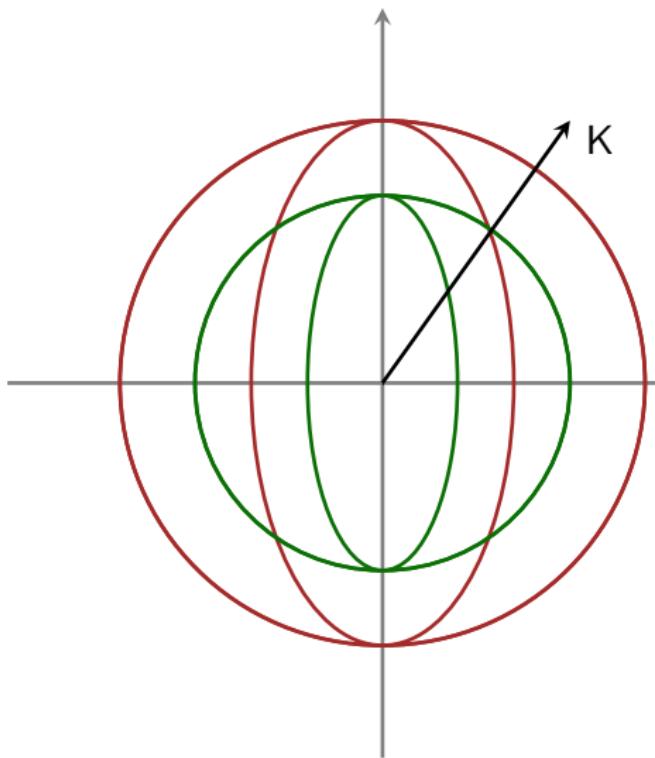
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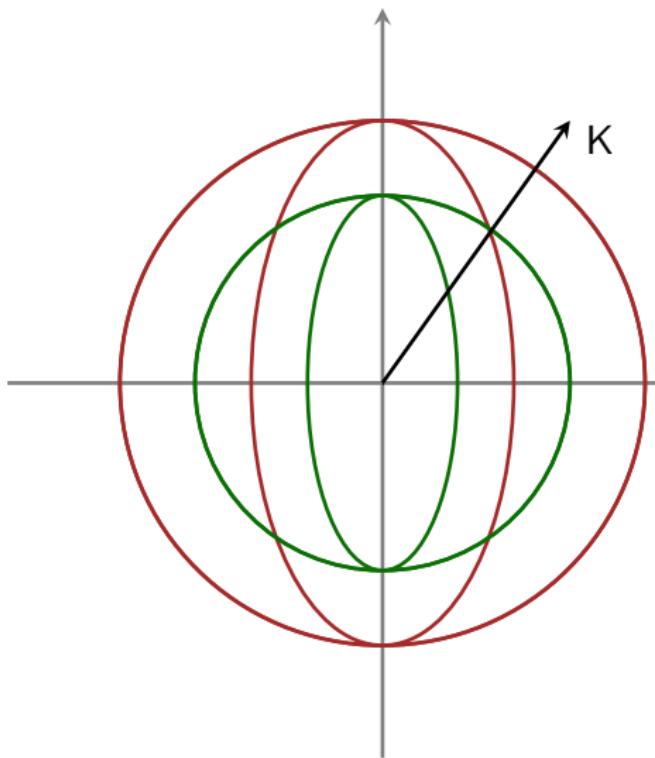
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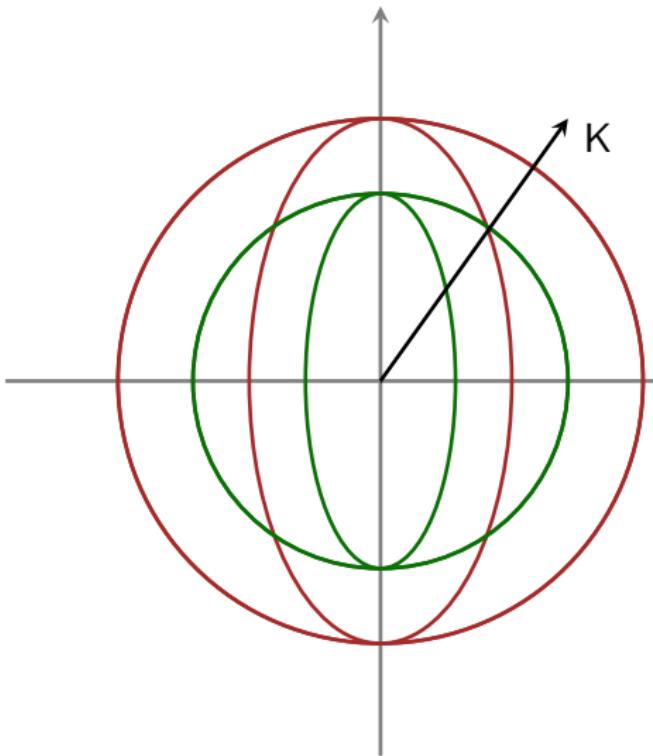
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Type II phase matching

In the three beam interaction, Type I was

- Both input beams ω_1 and ω_2 share the same polarization
- The generated beam ω_3 polarization is orthogonal

Another solution : Type II

- Input beams polarization are orthogonal
- Generated beam share one of them
- Not possible for SHG
- How is the angle calculated ?

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Phase matching in bi-axial crystals

A hard task

- Phase matching is seldom colinear
- Vector phase matching in a complex index ellipsoïd
- I will let you think on it

Paper by *Bœuf* can help

 N. Boeuf, D. Branning, I. Chaperot, E. Dauler, S. Guerin, G. Jaeger, A. Muller, and A. Migdall.

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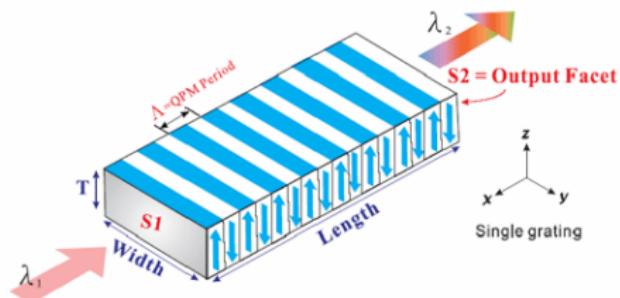
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Quasi phase matching in layered media

Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- d sign reversal



Wave solution

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

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

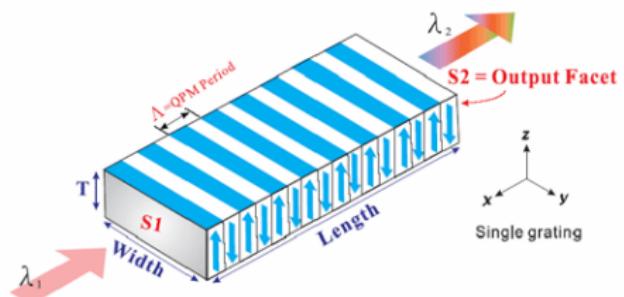
Intensity Solution

$$\left| \frac{i\omega_3}{2} \eta_3 [d]_{jki} |E^{(\omega_1)}|_i^2 \right|^2 \Lambda^2 \text{sinc}^2 (\Delta k \Lambda / 2) \frac{1 - (-1)^N \cos(\Delta k \Lambda N)}{1 + \cos(\Delta k \Lambda)}$$

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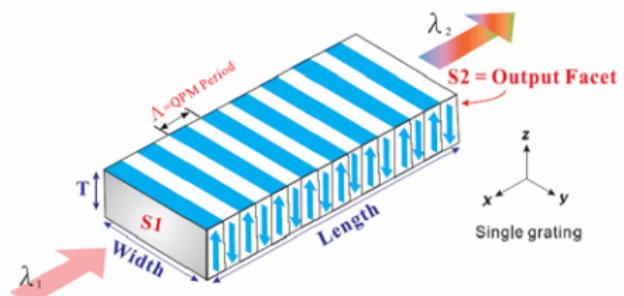
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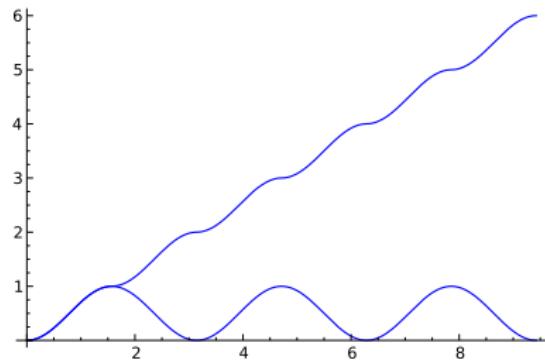
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Intensity Solution

$$\Delta k A = \pi$$

$$\left| \frac{i\omega_3}{2} \eta_3 [d]_{jki} |E^{(\omega_1)}|^2_i \right|^2 4 A^2$$