Second Harmonic Generation and related second order Nonlinear Optics

Nicolas Fressengeas

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Usefull reading. . .

[YY84, DGN91, LKW99]

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Contents

A classical Maxwell framework

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

Maxwell Model

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

Matter equations

- $D = \varepsilon_0 E + P = \varepsilon E$
- $B = \mu_0 H$
- $P = \varepsilon_0 \chi_L E + P_{NL}$

Wave equation in isotropic medium

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

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

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 : 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

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

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

3 Perturbed wave equations

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

Temporal harmonic notation

- $E^{(\omega_\nu)}$ is a plane wave
- Non linear polarisation is a plane wave
- Considering only… the ω_ν part $\Leftrightarrow$ the plane wave part
- $\frac{\partial^2 E^{(\omega_\nu)}(z,t)}{\partial z^2} + \mu_0 \varepsilon_{\omega_\nu} \omega_\nu^2 E^{(\omega_\nu)}(z,t) = -\mu_0 \omega_\nu^2 P_{NL}^{\omega_\nu}(z,t)$

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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

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

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The Slow Varying Approximation

Closely related to the paraxial approximation

The Slow Varying Approximation

- Beam enveloppe is assumed ta 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...

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

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Wave Propagation Equation under SVA approximation

Obtaining an envoloppe equation, which is simpler

Non SVA wave propagation equation

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SVA equation

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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

- $\mathcal{R}e\left(\left[2ik_1 \frac{\partial E^{(\omega_1)}(z)}{\partial z}\right] e^{(i(k_1 z - \omega_1 t))}\right)$
- $= \mathcal{R}e\left(-\mu_0 \omega_1^2 [d]_{ijk} \left[E^{(\omega_3)}(z)\right]_j \overline{\left[E^{(\omega_2)}(z)\right]_k} e^{(i(k_3-k_2)z - i(\omega_3-\omega_2)t)}\right)$

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

- $\left[\frac{\partial E^{(\omega_1)}}{\partial z}\right]_i = +\frac{i\omega_1}{2}\eta_1 [d]_{ijk} \left[E^{(\omega_3)}\right]_j \overline{\left[E^{(\omega_2)}\right]_k} \exp\left(-i\Delta k z\right)$
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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 wich are large
- ...

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Example : sum frequency generation

Input beams assumed constant — Undepleted pump approximation

Assumptions

- $\omega_1 + \omega_2 = \omega_3$
- Generated beam null at $z = 0$: $\left[E^{(\omega_3)}(z=0)\right]_j = 0$
- $\dfrac{\partial\left[E^{(\omega_1)}\right]_i}{\partial z} = \overline{\dfrac{\partial\left[E^{(\omega_2)}\right]_k}{\partial z}} = 0$

One equation remains

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

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Solving the SVA wave propagation equation

Equation to solve

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$\Delta k \neq 0$

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

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

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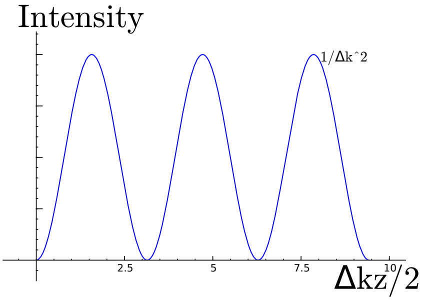
Phase match or not phase match

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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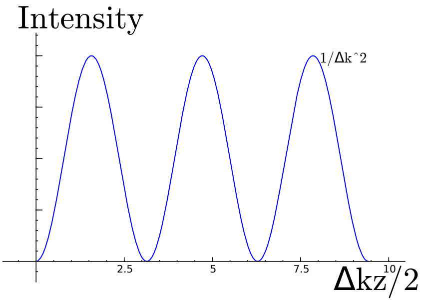
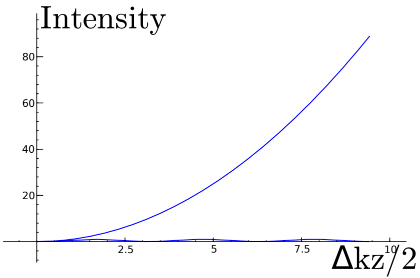


Phase match $\Delta k = 0$

- Intensity quadratic increase
- Approximations do not hold long



A Scalar Three Wave Interaction model

Further approximations to remove vectors

Simplifying notations

- Set indices equal for polarization and frequency: $A_\nu = \left[E^{(\omega_\nu)}\right]_\nu$
- 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
- Let $K = dC/2$

Scalar three wave interaction

- $\frac{\partial A_1}{\partial z} = +iK\overline{A_2}A_3 \exp\left(-i\Delta k z\right)$
- $\frac{\partial A_2}{\partial z} = -iKA_1\overline{A_3} \exp\left(+i\Delta k z\right)$
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Second Harmonic Generation SHG

Sum frequency generation of two equal frequencies from the same source

One intput 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

- $\frac{\partial A_1}{\partial z} = +iK\overline{A_1}A_3 \exp(-i\Delta kz)$
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Phase matching $\Delta k = 0$

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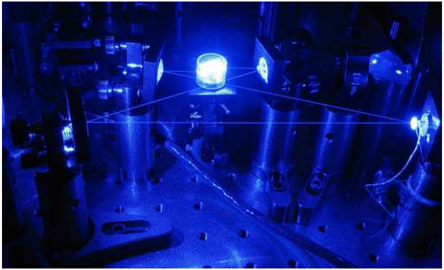

Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source flickr)

Second Harmonic Generation SHG

Sum frequency generation of two equal frequencies from the same source

One intput 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

- $\frac{\partial A_1}{\partial z} = +iK\overline{A_1}A_3 \exp\left(-i\Delta kz\right)$
- $\frac{\partial A_3}{\partial z} = +iKA_1{}^2 \exp\left(+i\Delta kz\right)$

Phase matching $\Delta k = 0$

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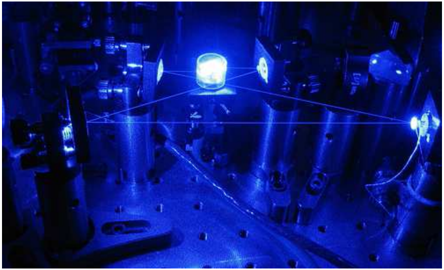

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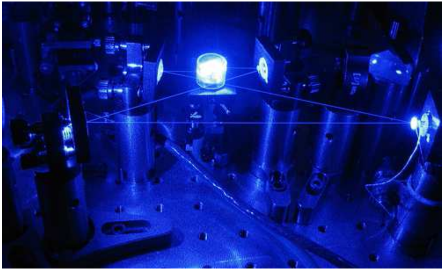

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- $A_3 = i\tilde{A}_3 \Rightarrow \tilde{A}_3 \in \mathbb{R}$
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Real equations

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

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

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

Start with no harmonic

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

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

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$\tilde{A}_3$ expression

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I_3 expression

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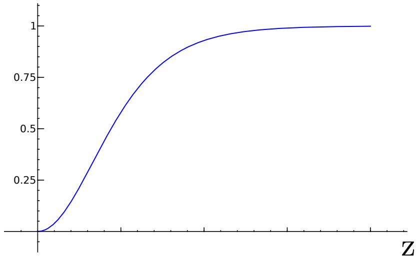

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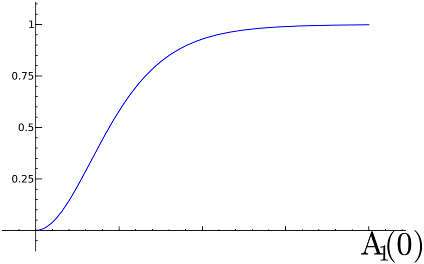
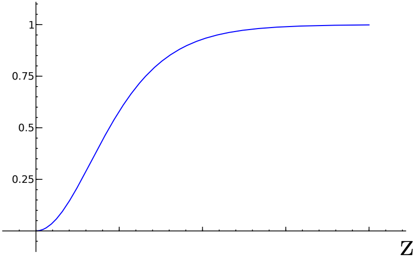
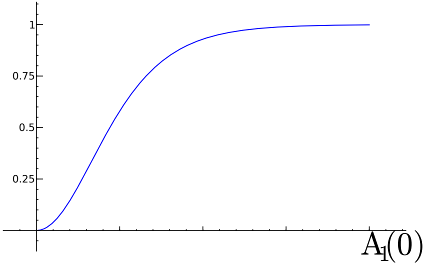


SHG conclusion

Second Harmonic Beam Evolution





Suprinsingly. . .

- It is possible to convert 100% of a beam, with large interaction length or intensity
- The process has no threshold and does not need noise to start
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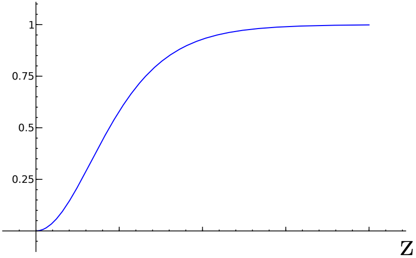

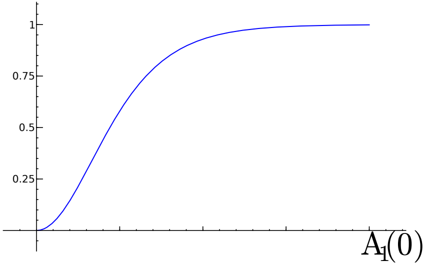

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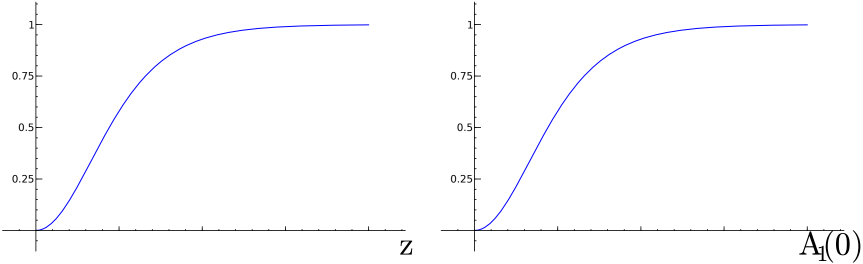

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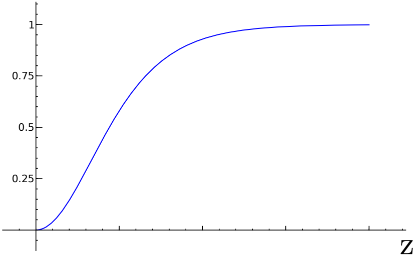

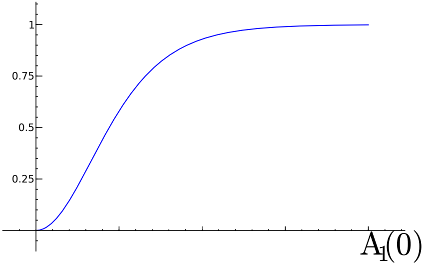

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Optical Parametric Amplifier OPA

Optical Amplification of a weak signal beam thanks to a powerfull 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

- $\frac{\partial A_1}{\partial z} = +iK_p\overline{A_2}$
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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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Initial conditions

- A weak signal : $A_1(0) \neq 0$
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Amplitude solution

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

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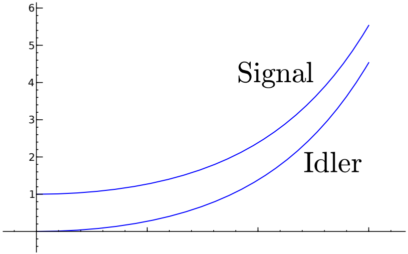


Optical Parametric Oscillator OPO

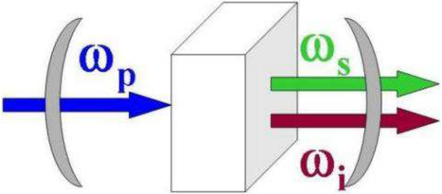
Use Optical Parametric Amplification to make a tunable laser

OPA pumped with ω_3

- Amplifier for ω_1 and ω_2
- With $\omega_1 + \omega_2 = \omega_3$
- Phase matching: $k_1 + k_2 = k_3$
- ω_1 and ω_2 initiated from noise

Frequency tunable laser

- Get Non Linear Medium
- Adjust Cavity for ω_1 and ω_2
- Pump with ω_3
- You got it !



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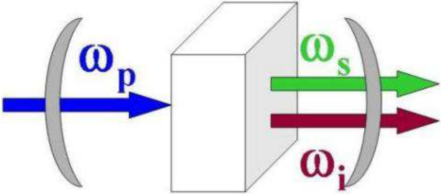

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1. Three-wave interaction
   - Assumptions framework
   - Three Wave propagation equation
   - Sum frequency generation
   - Scalar approximation
2. Non Linear Optics Application
   - Second Harmonic Generation
   - Optical Parametric Amplifier
  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

Colinear (scalar) phase matching

Phase matching for co-propagation waves

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

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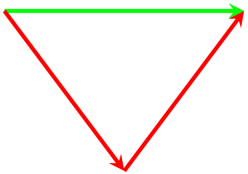
Use birefringent crystals and different polarizations

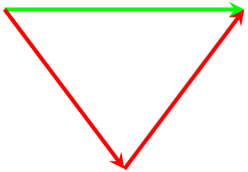
Non colinear phase matching



Non colinear phase matching

Use clever geometries

With reflections



Non colinear phase matching

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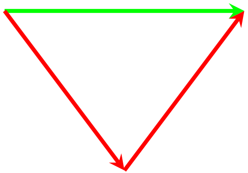
With reflections

Or even more clever...



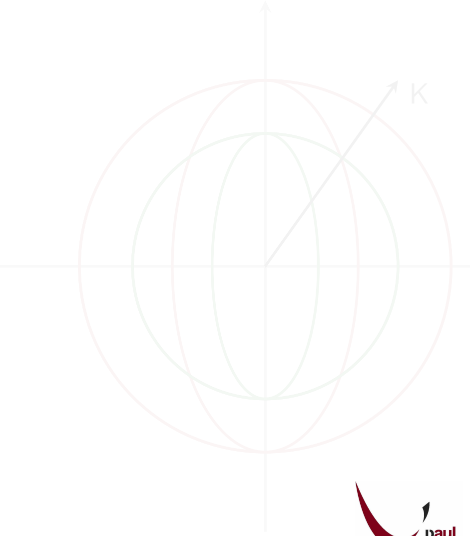
SHG Type I Phase Matching

Waves polarization

- 1 incident wave counts for 2
- They share the same polarisation
- 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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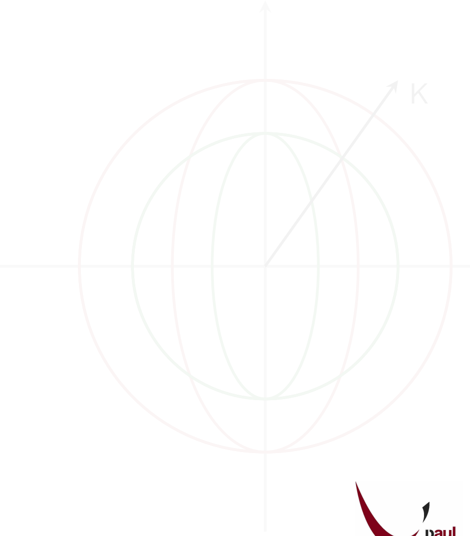

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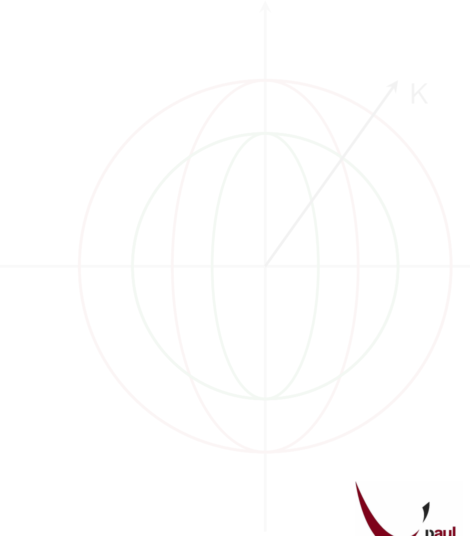

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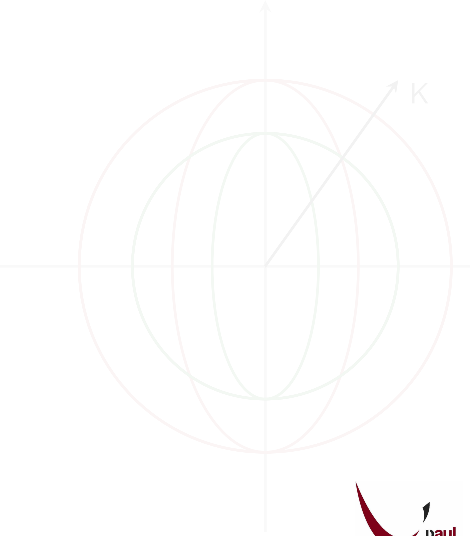

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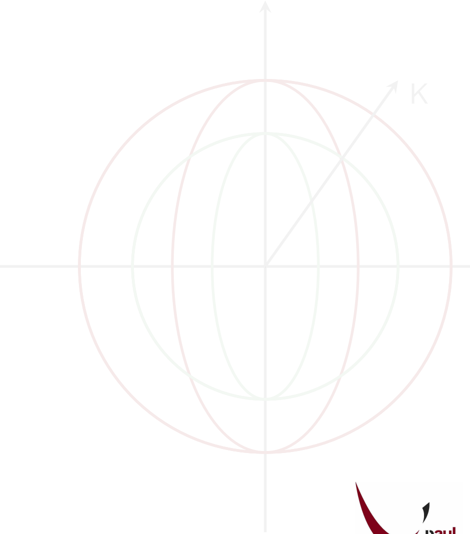

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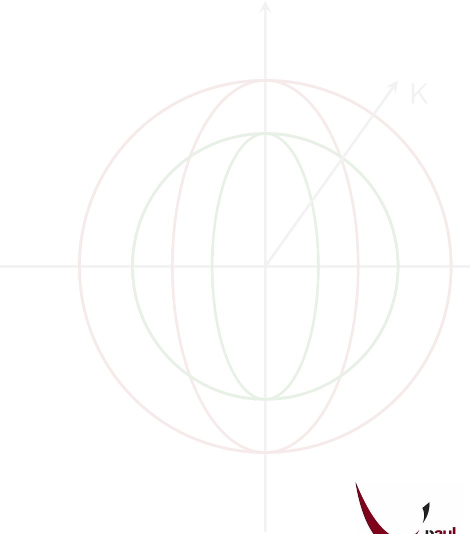

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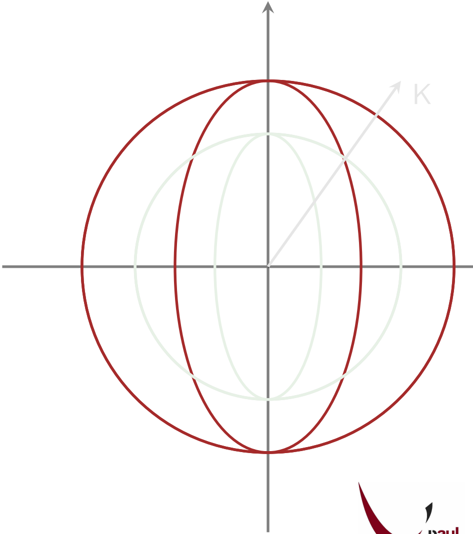

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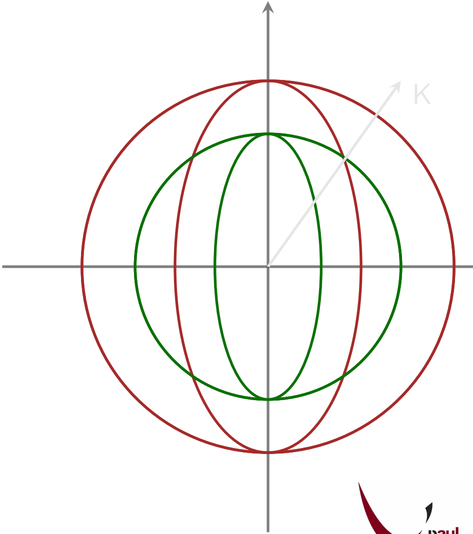

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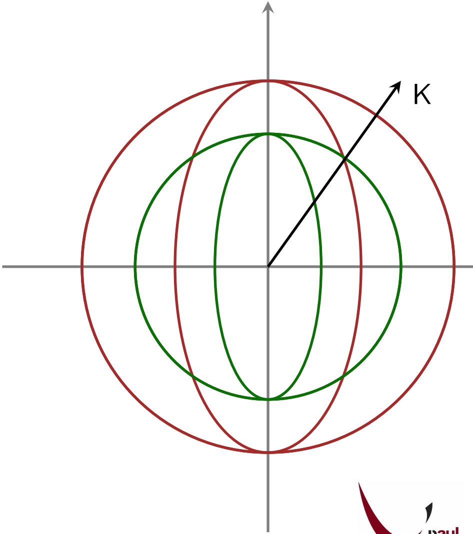
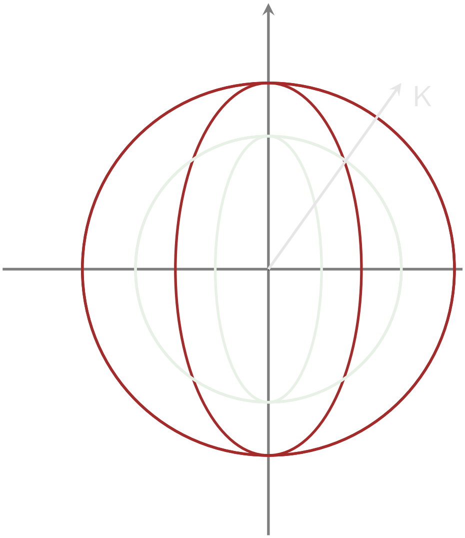


SHG Type I phase matching: a few numbers



Fundamental index ellipsoïd section

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

Harmonic index ellipsoïd section

$$\frac{1}{\tilde{n}_o^2(\theta)} = \frac{\cos^2(\theta)}{\tilde{n}_o^2} + \frac{\sin^2(\theta)}{\tilde{n}_e^2}$$

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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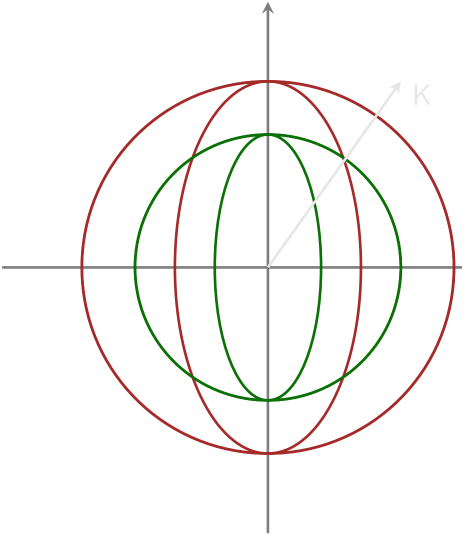

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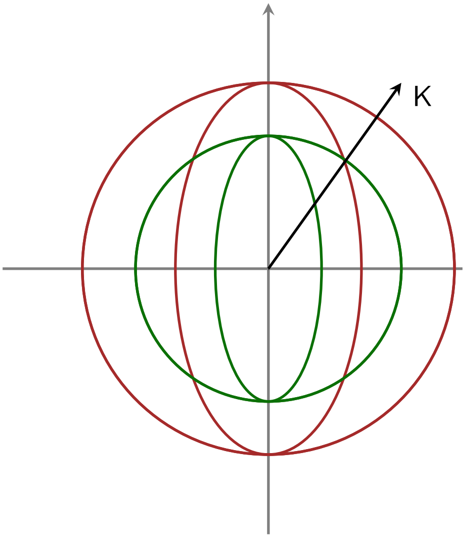

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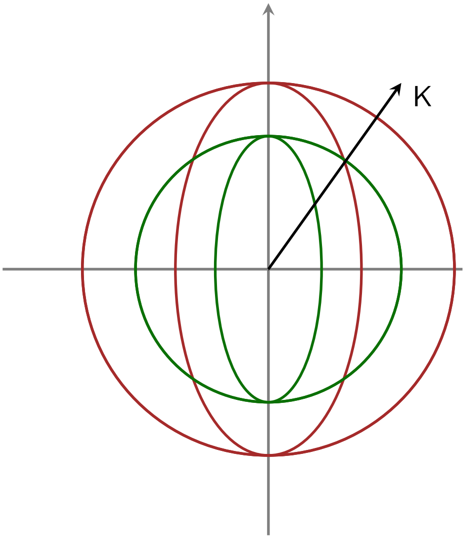

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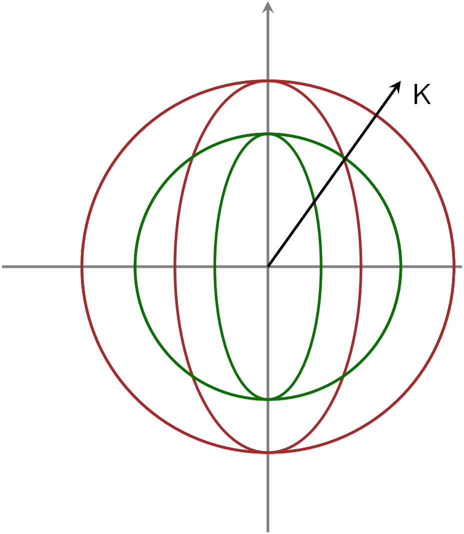

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Type II phase matching

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
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- How is the angle calculated ?

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Phase matching in bi-axial crystals

A hard task

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

Paper by *Bœuf* can help

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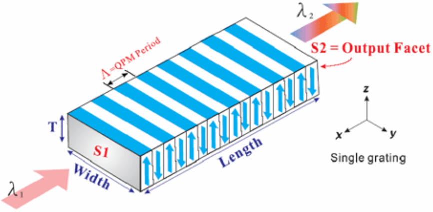
Quasi phase matching in layered media

Perodicaly Poled Lithium Niobate

- Periodic Domain Reversal
- d sign reversal



Wave solution

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

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

Intsensity Solution

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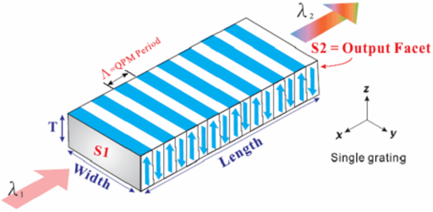

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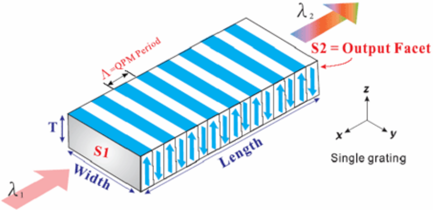

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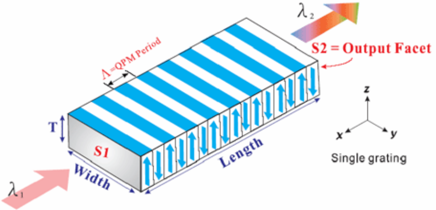

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