

Second Harmonic Generation and related second order Nonlinear Optics

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Second Harmonic Generation and related second order Nonlinear Optics

N. Fressengeas

Laboratoire Matériaux Optiques, Photonique et Systèmes
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Useful reading...

[YY84, DGN91, LKW99]

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V. G. Dmitriev, G. G. Gurzadyan, and D. N. Nikogosyan.
Handbook of Nonlinear Optical Crystals, volume 64 of *Springer Series in Optical Sciences*.
Springer Verlag, Heidelberg, Germany, 1991.



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Springer-Verlag, New York, 1999.



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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.



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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



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



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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

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

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...

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



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Wave Propagation Equation under SVA approximation

Obtaining an 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

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

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

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

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

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Example : sum frequency generation

Input beams assumed constant

Undepleted pump approximation

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

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

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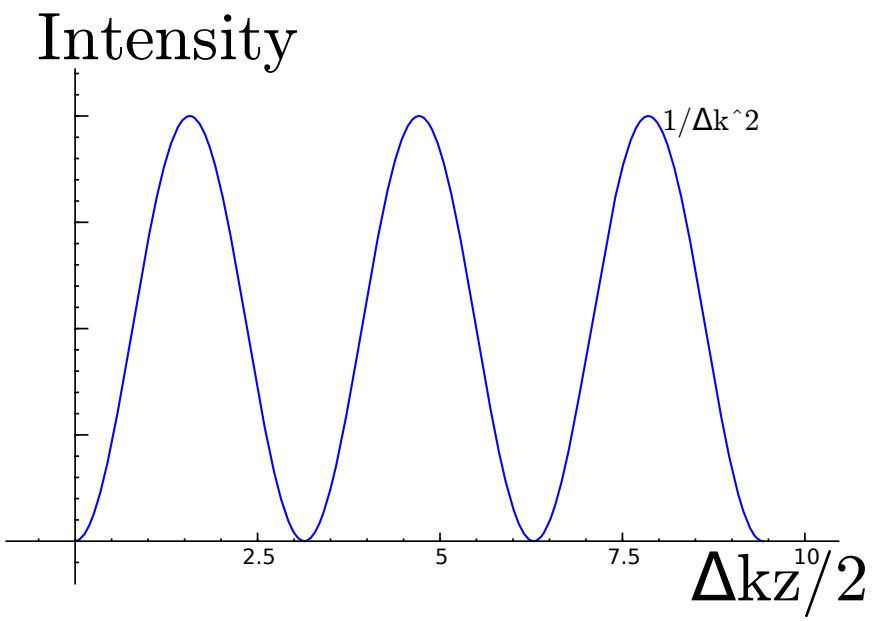


Phase match or not phase match

Phase matching is a key issue to sum frequency generation

Phase mismatch $\Delta k \neq 0$

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



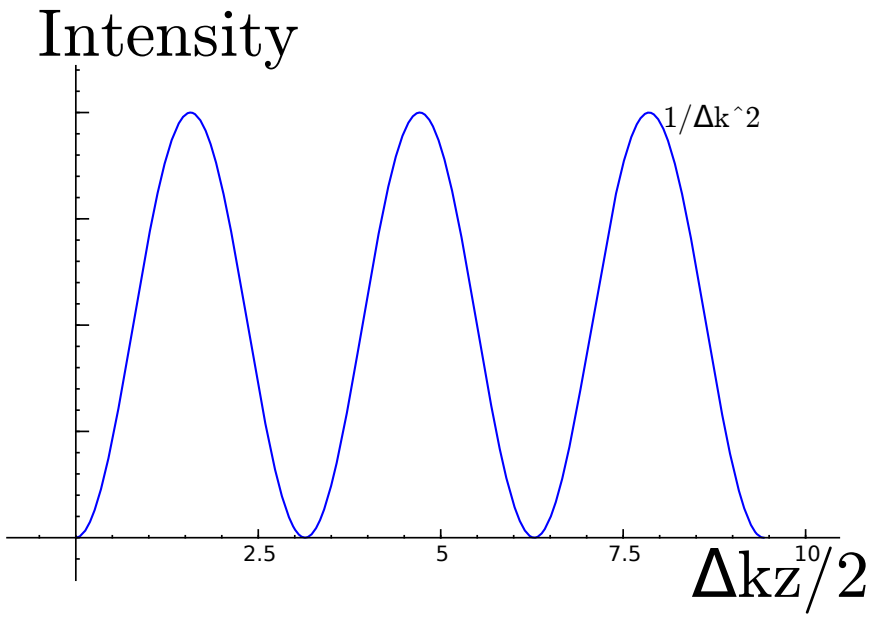


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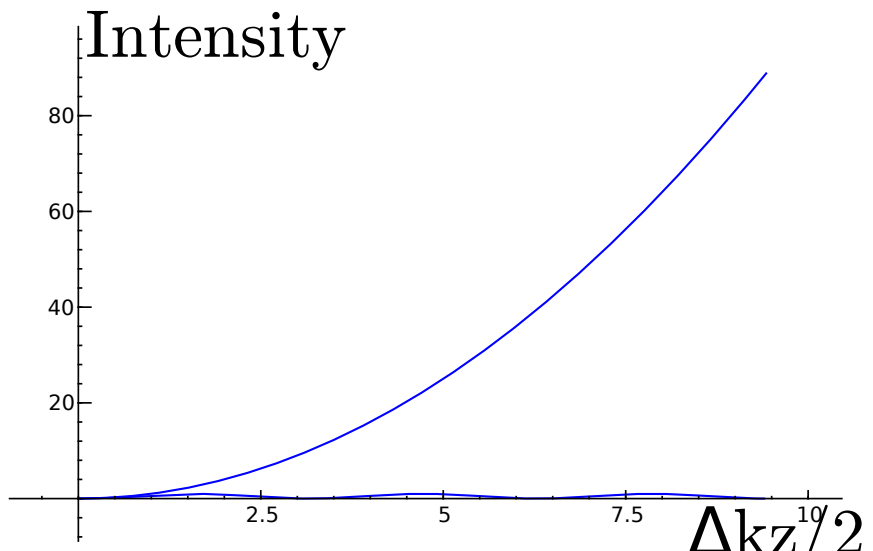
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Phase match $\Delta k = 0$

- Intensity quadratic increase
- Approximations do not hold long



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A Scalar Three Wave Interaction model

Further approximations to remove vectors

Simplifying notations

- Set indexes equal for polarization and frequency: $A_\nu = \left[E^{(\omega_\nu)}\right]_\nu$
- 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

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

Further approximations to remove vectors

Simplifying notations

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

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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

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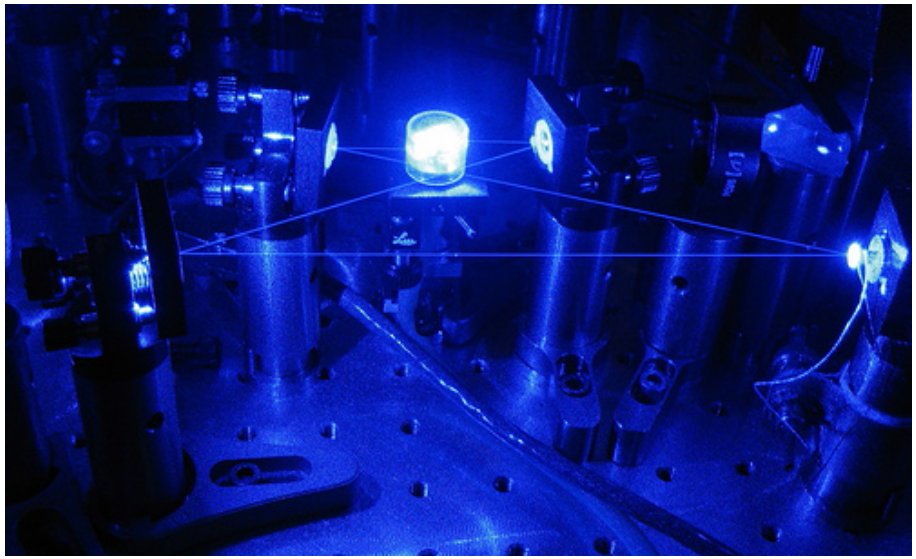


Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source `flickr`)



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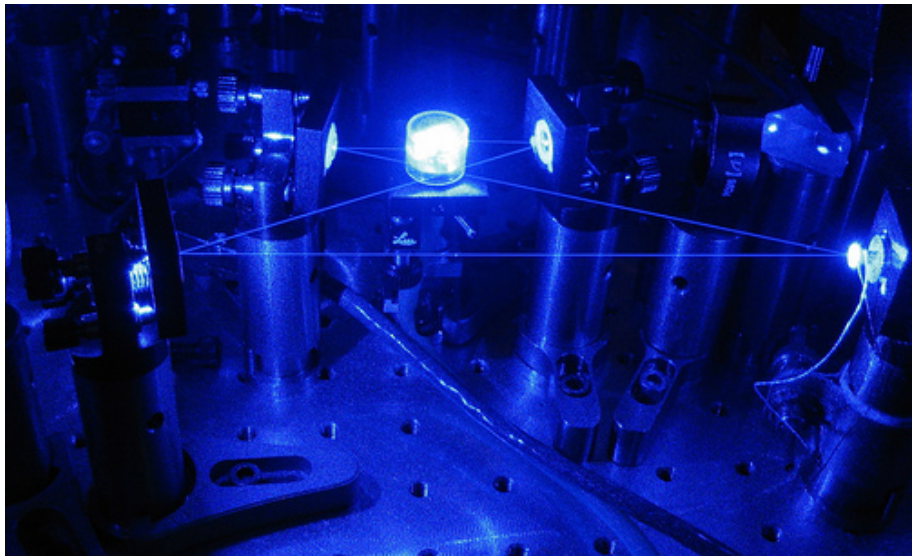


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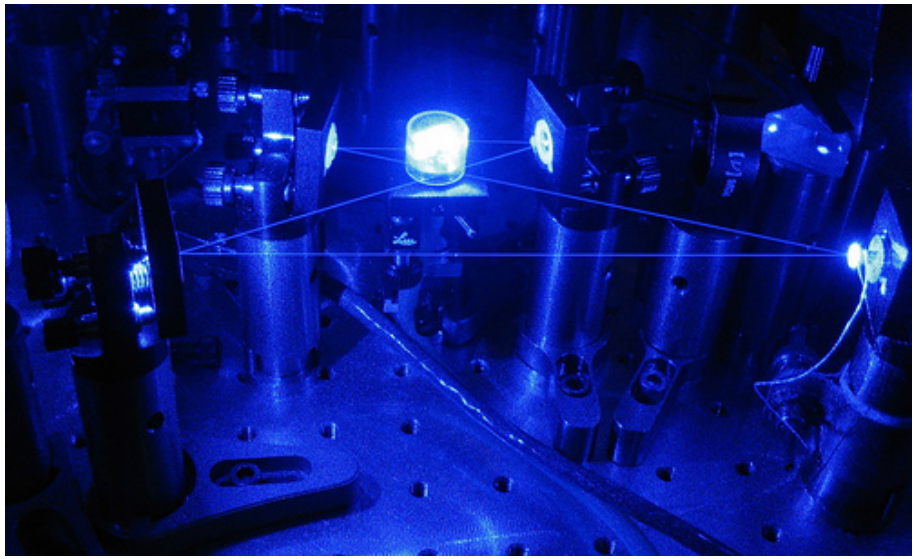


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Real equations

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

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

$\tilde{A}_3$ equation

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



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

$\tilde{A}_3$ expression

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

I_3 expression

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

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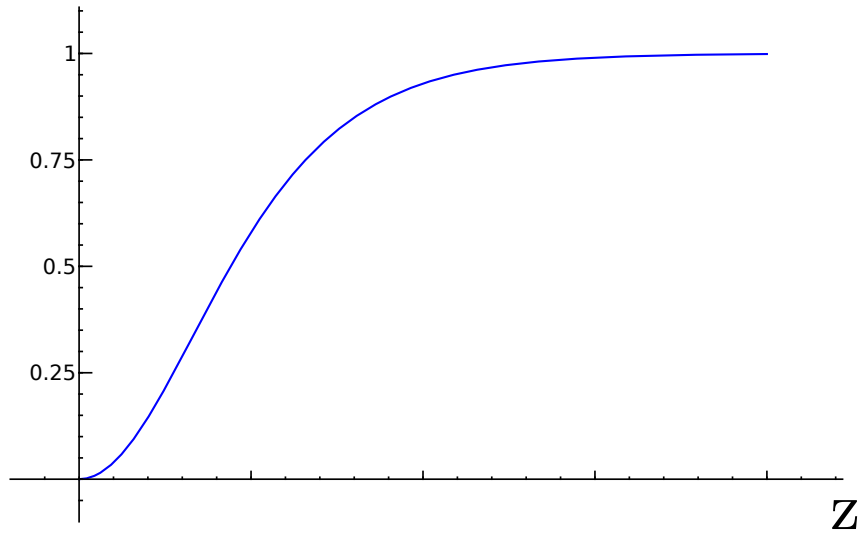
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$I_3(z)/I_1(0)$ $\qquad$ **$I_1(0) = \text{constant}$**





Second Harmonic Beam Evolution

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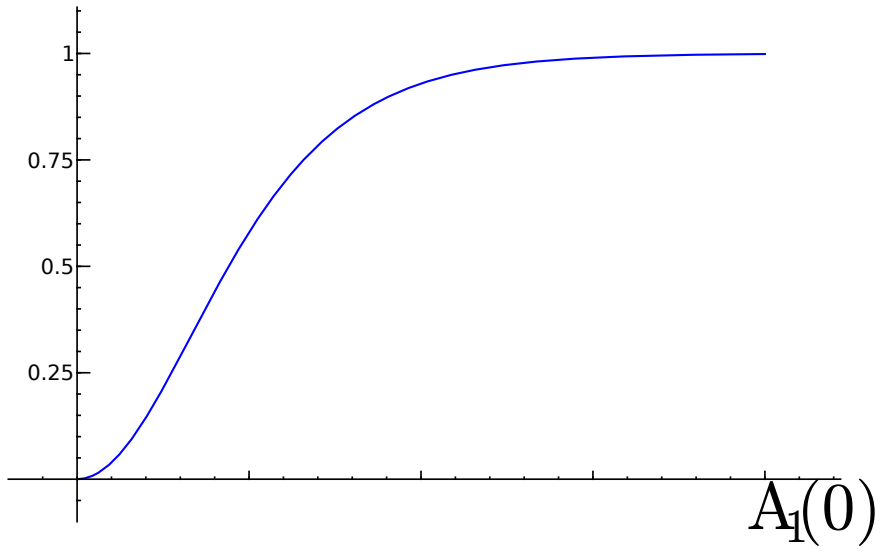
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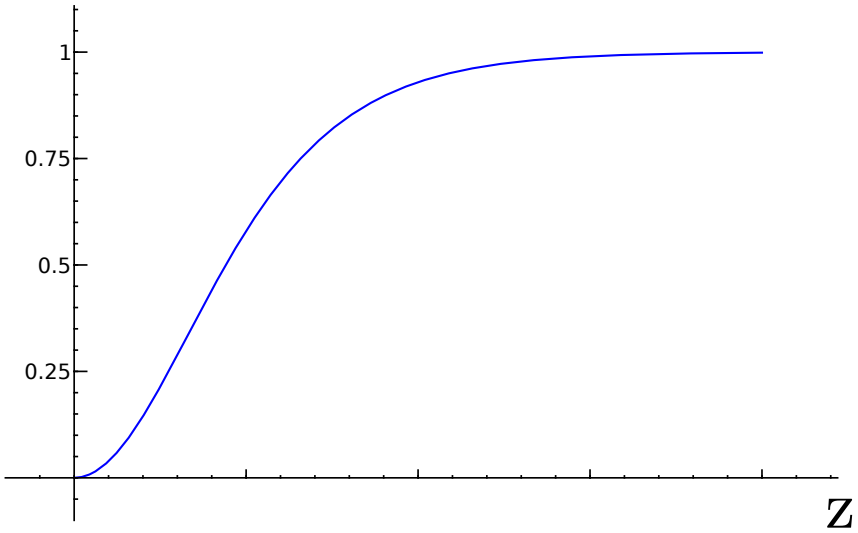
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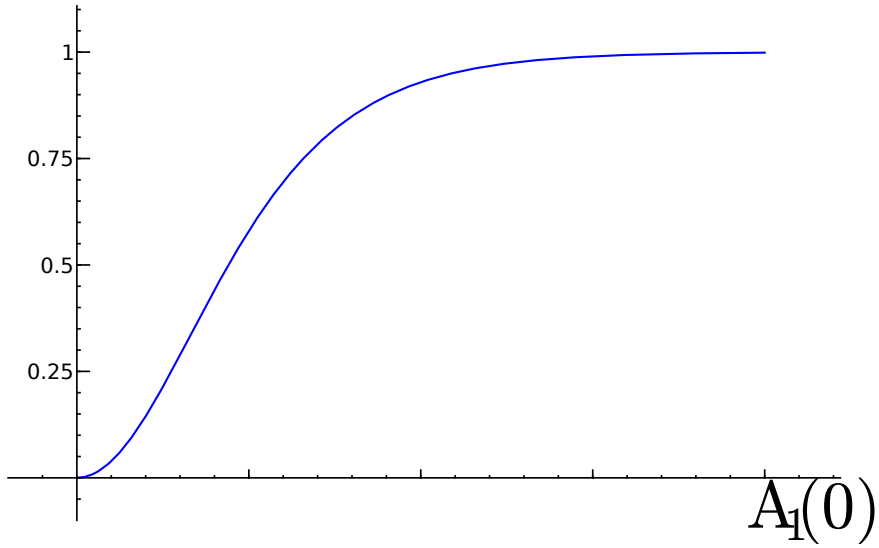




SHG conclusion

Second Harmonic Beam Evolution



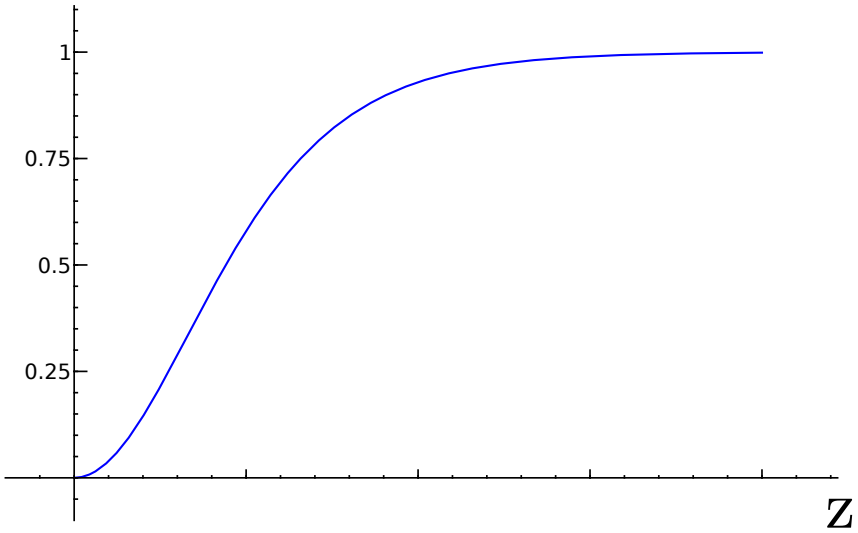


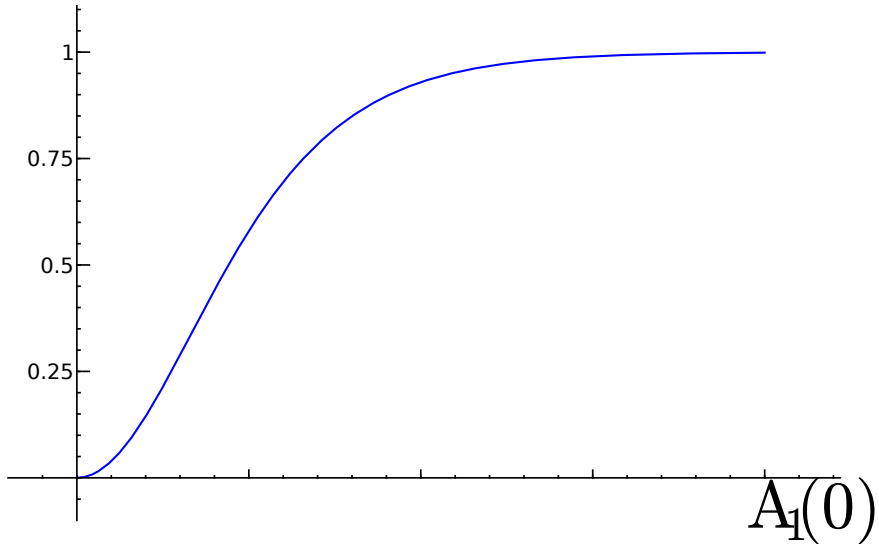
Suprinsingly. . .

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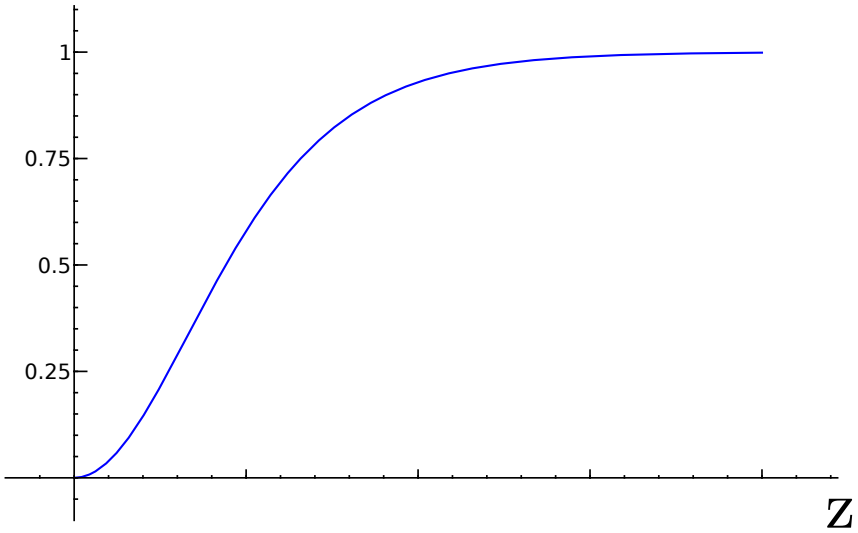


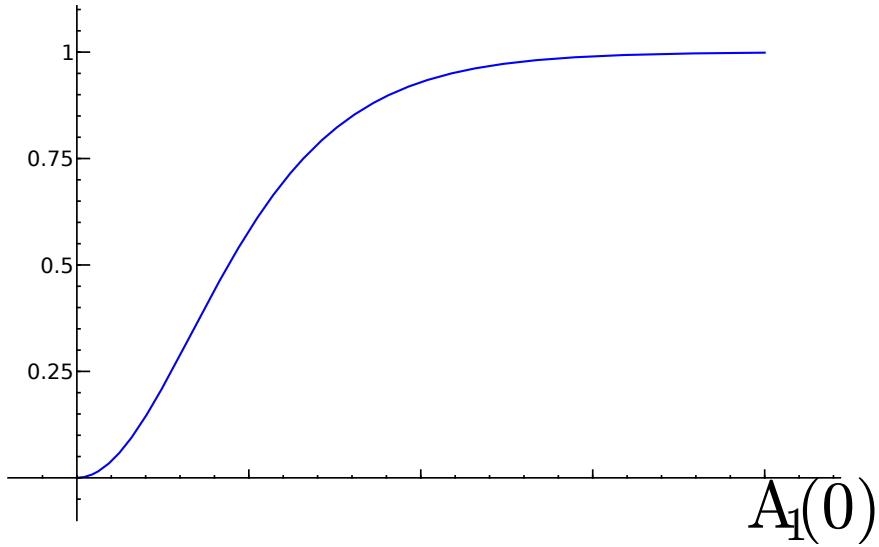
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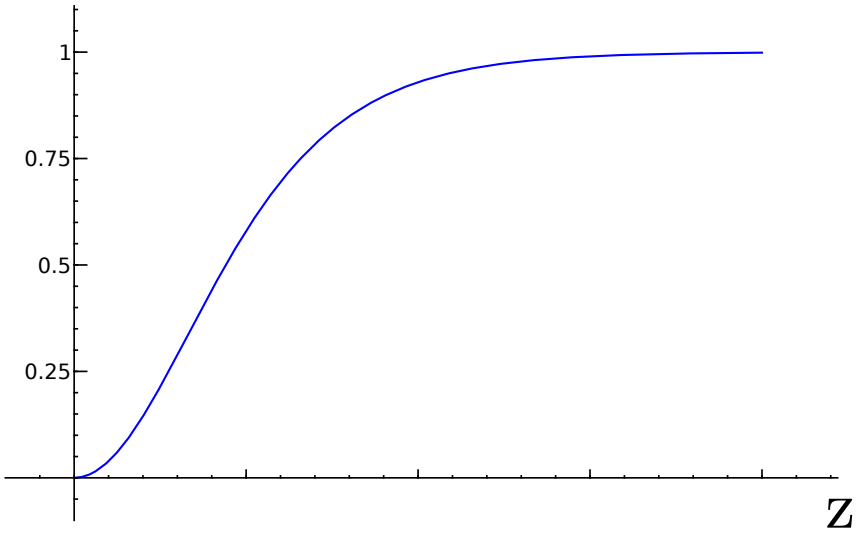


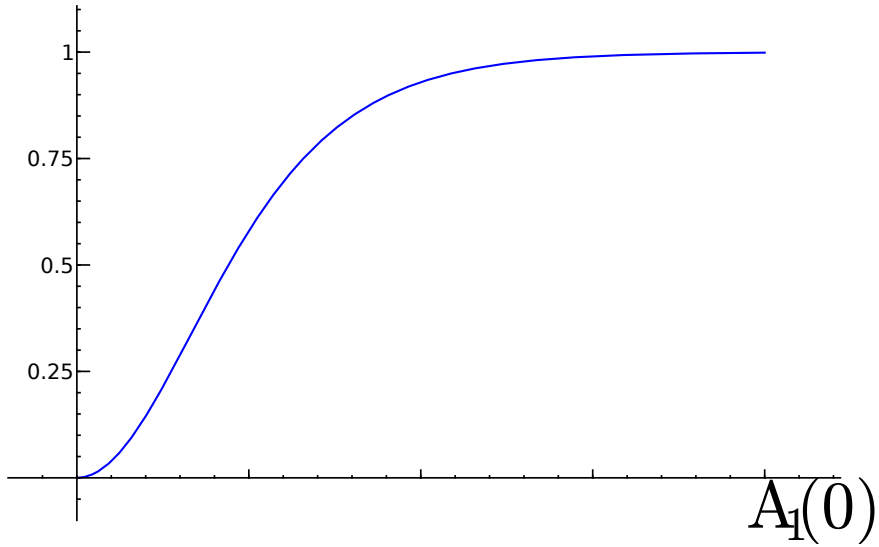
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Optical Parametric Amplifier OPA

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

Signal beam amplification

- 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

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

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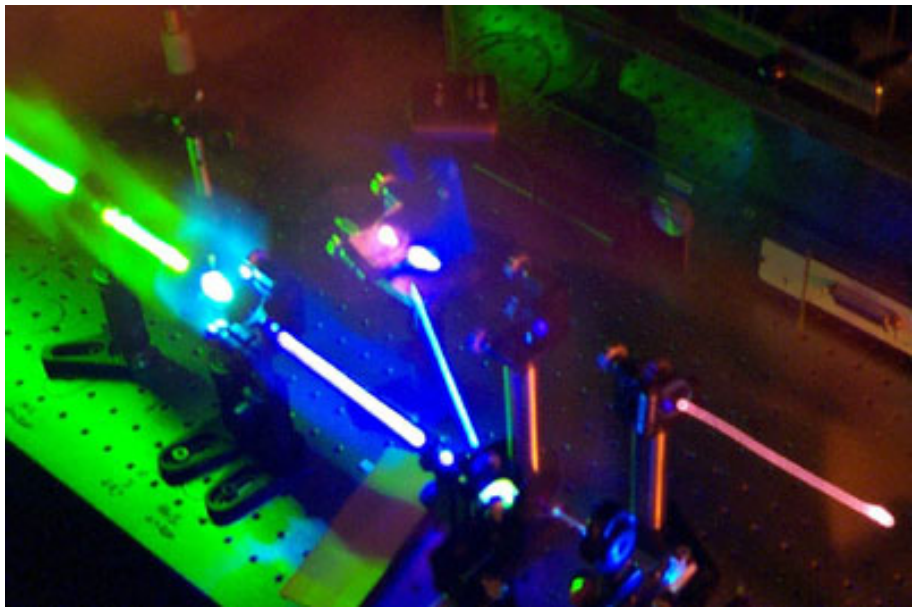


Figure: White light continuum seeded optical parametric amplifier (OPA) able to generate extremely short pulses. (source Freie Universität Berlin)

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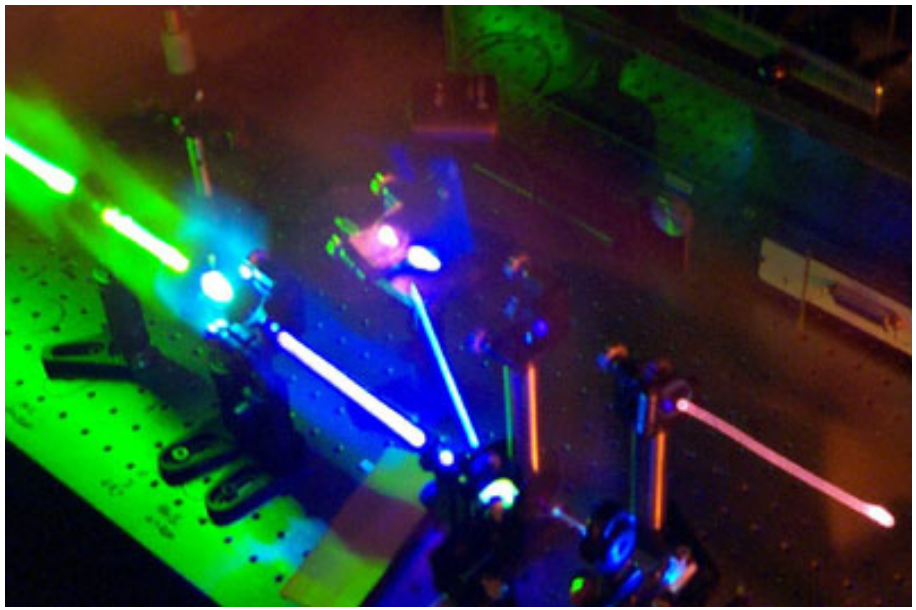


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Initial conditions

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- No idler : $A_2(0) = 0$

Amplitude solution

- Amplified signal : $A_1(z) = A_1(0)\cosh(K_pz)$
- Idler : $\overline{A_2(z)} = -iA_1(0)\sinh(K_pz)$



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- No idler : $A_2(0) = 0$

Amplitude solution

- Amplified signal :
  $A_1(z) = A_1(0)\cosh(K_pz)$
- Idler :
  $\overline{A_2(z)} = -iA_1(0)\sinh(K_pz)$



Solving the OPA equations

Phase matched equations

- $\frac{\partial A_1}{\partial z} = +iK_p\overline{A_2}$
- $\overline{\frac{\partial A_2}{\partial z}} = -iK_pA_1$

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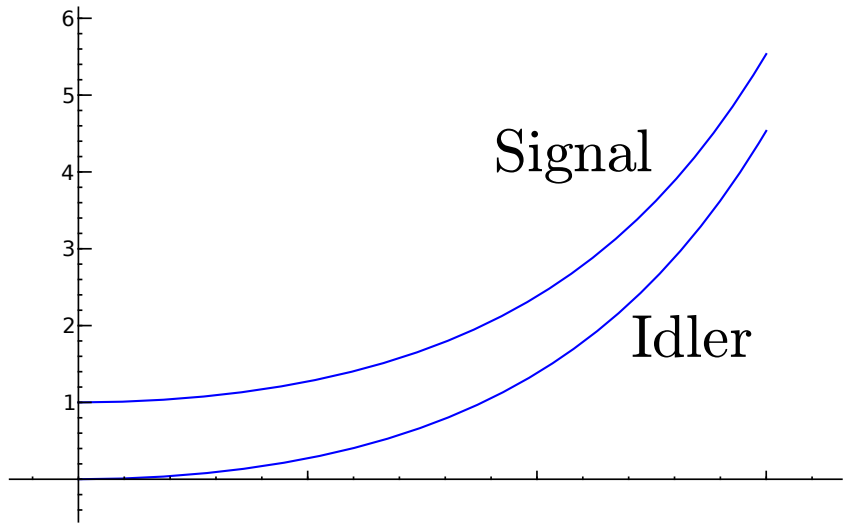
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Intensities

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  $I_1(z) = I_1(0)\cosh^2(K_pz)$
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Amplification



1. Three-wave interaction
   - Assumptions framework
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   - Second Harmonic Generation
   - Optical Parametric Amplifier
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   - Phase matching conditions
   - Phase matching in uni-axial crystals
   - Quasi-phase matching



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 !

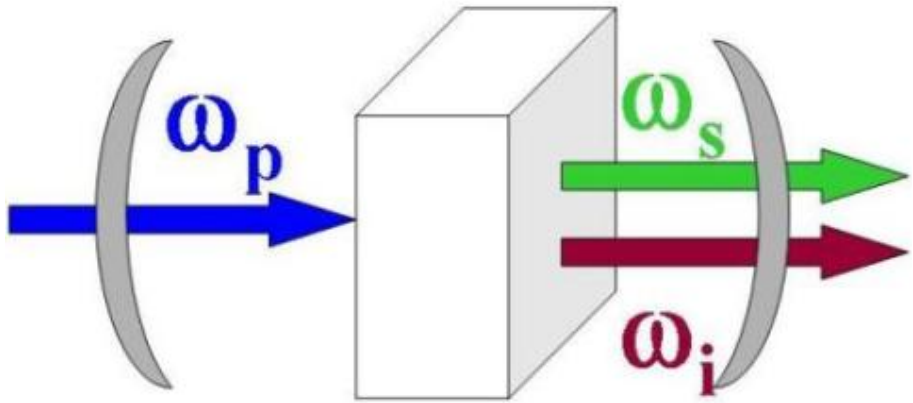




Figure: Optical Parametric Oscillator (source Cristal Laser)

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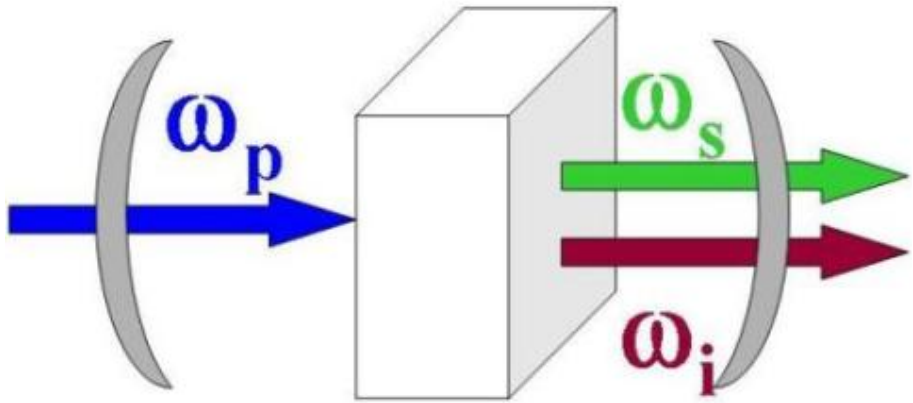




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Colinear (scalar) phase matching

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 : $2k_1 = k_3 \Rightarrow n_1 = n_3$
- 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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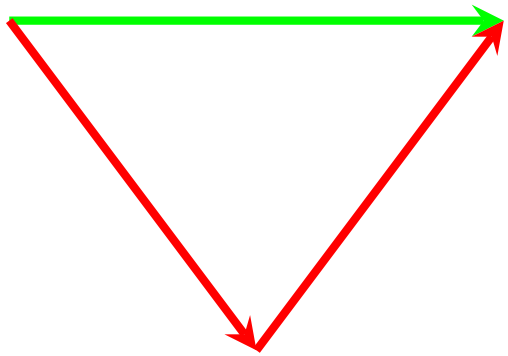
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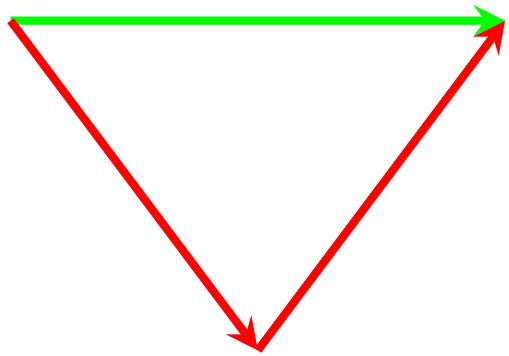




Non colinear phase matching

Use clever geometries

With reflections



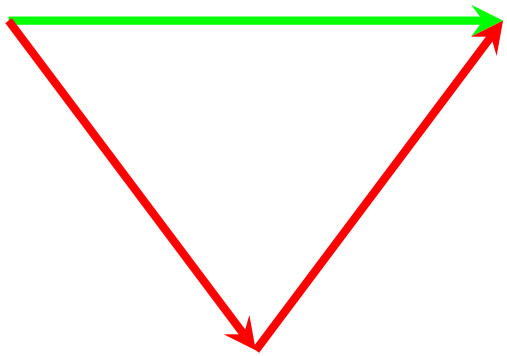


Non colinear phase matching

Use clever geometries

With reflections

Or even more clever...





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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
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n_0 and n_e function of propagation direction: index ellipsoid cross-section

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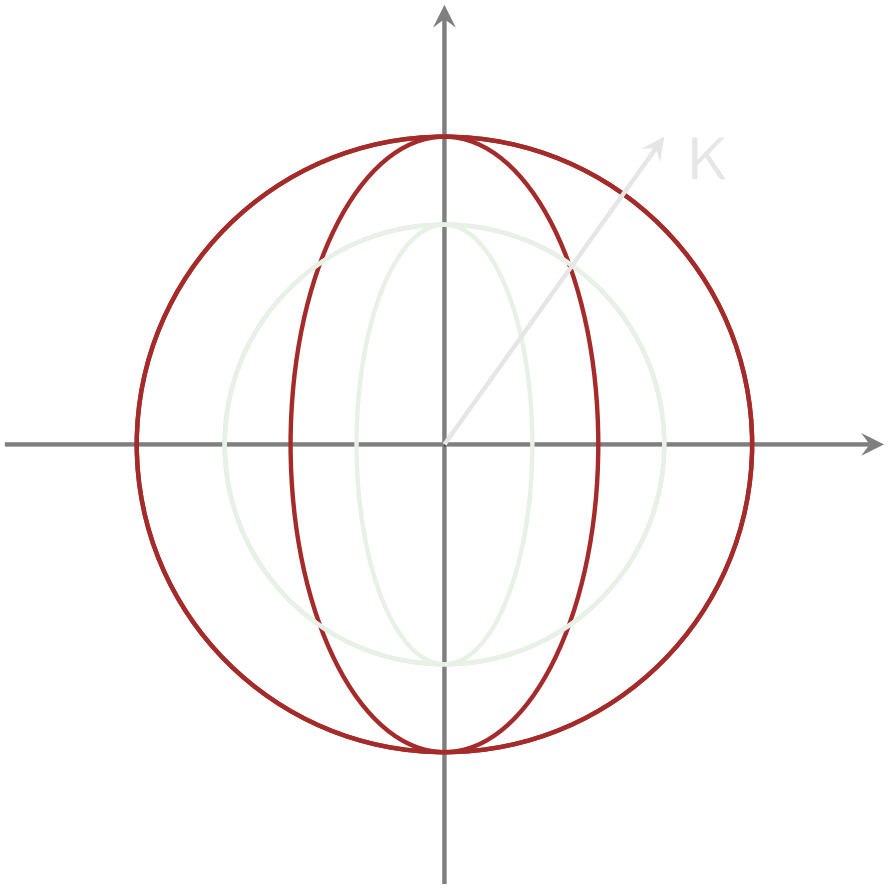
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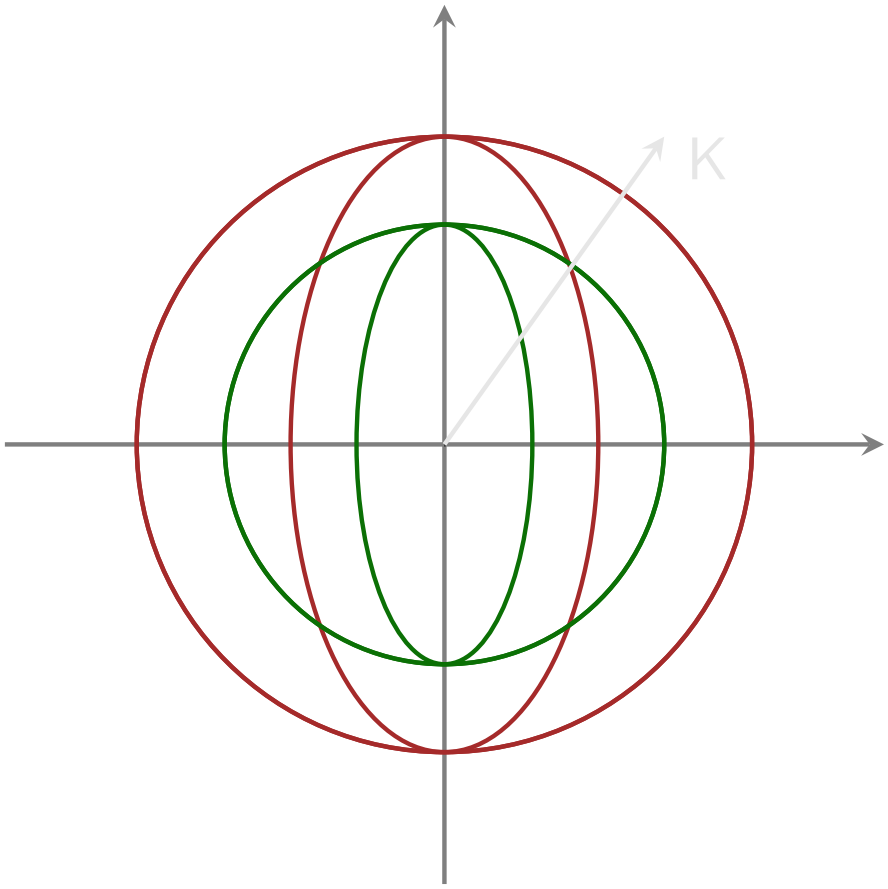
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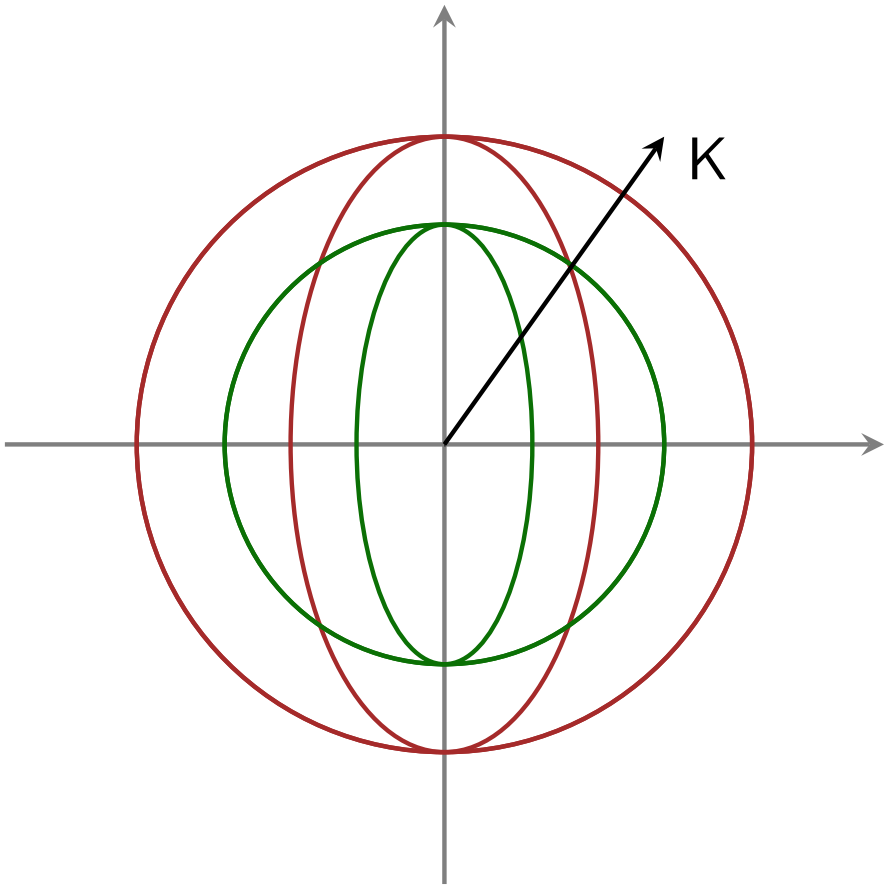
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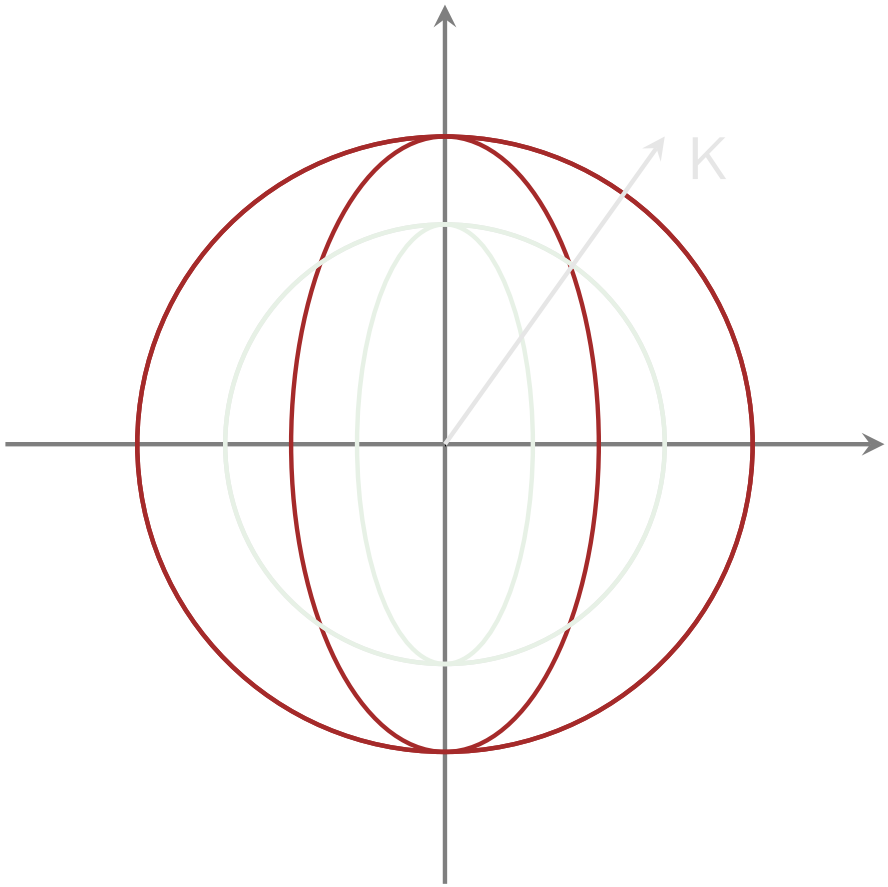
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SHG Type I phase matching: a few numbers



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

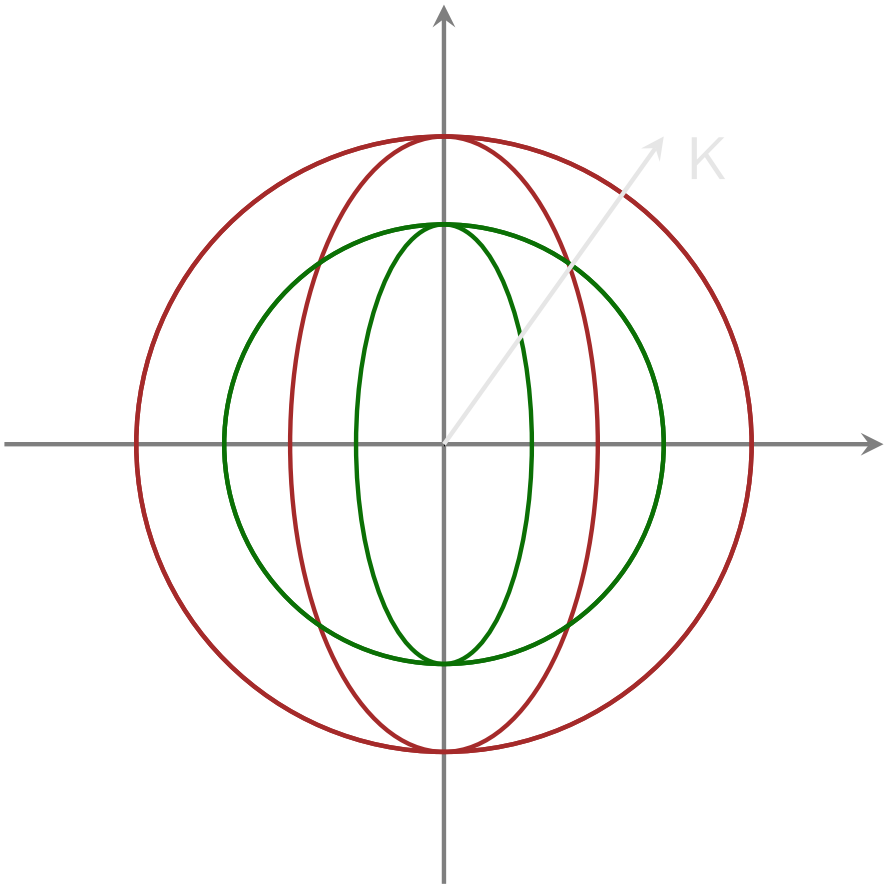
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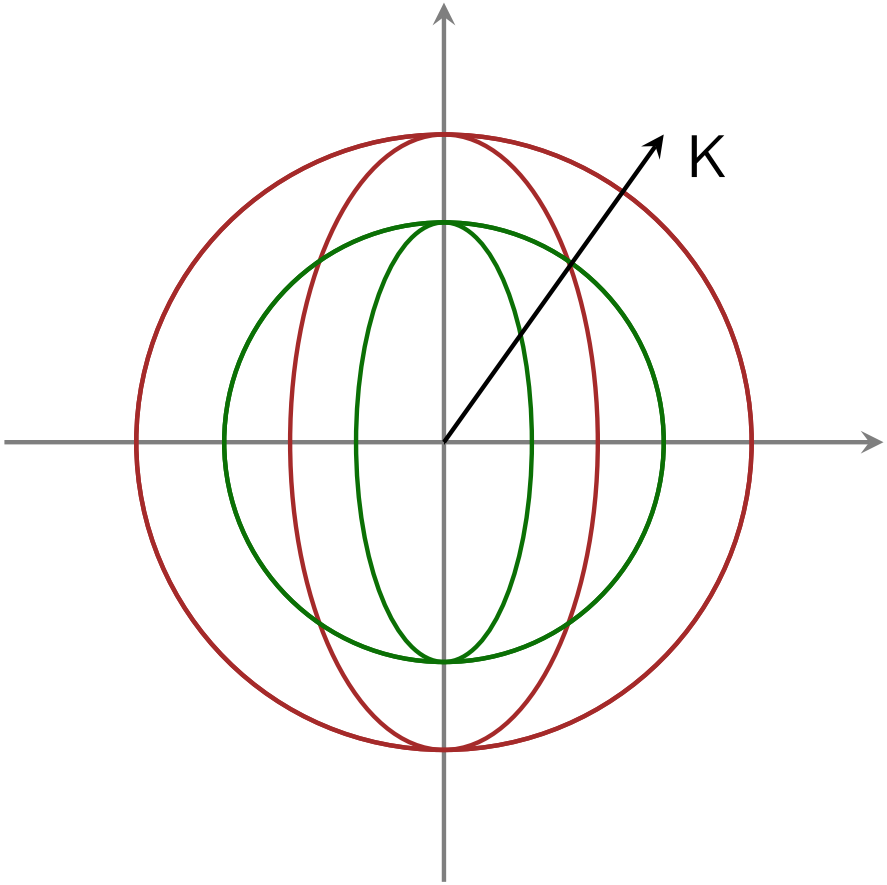
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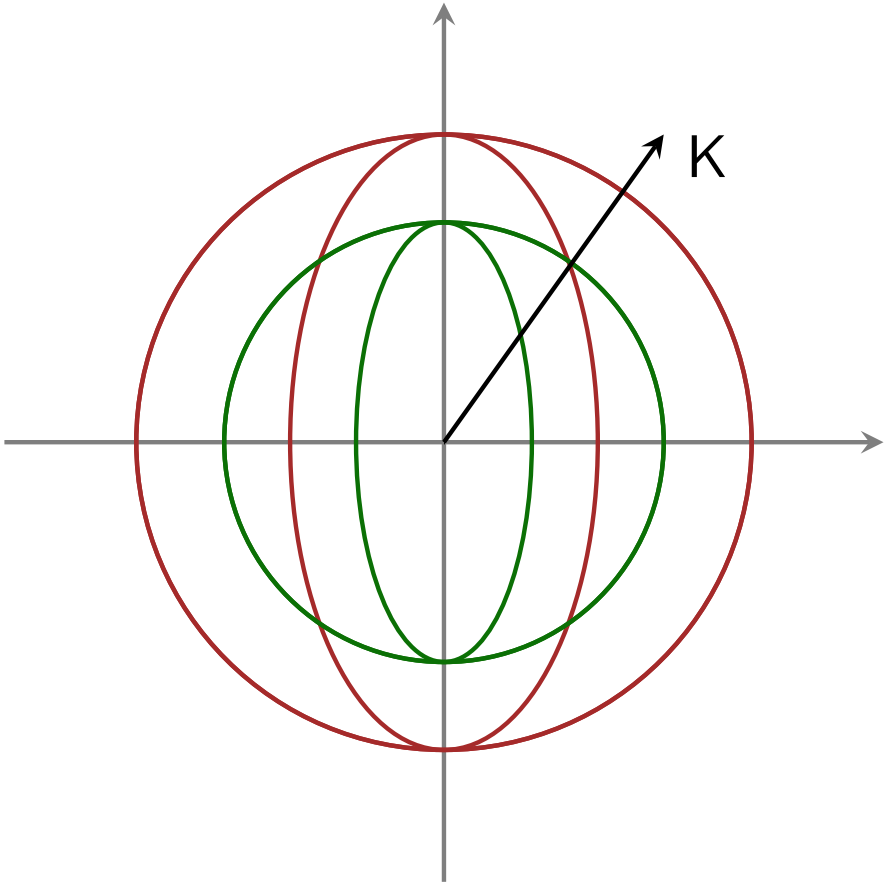
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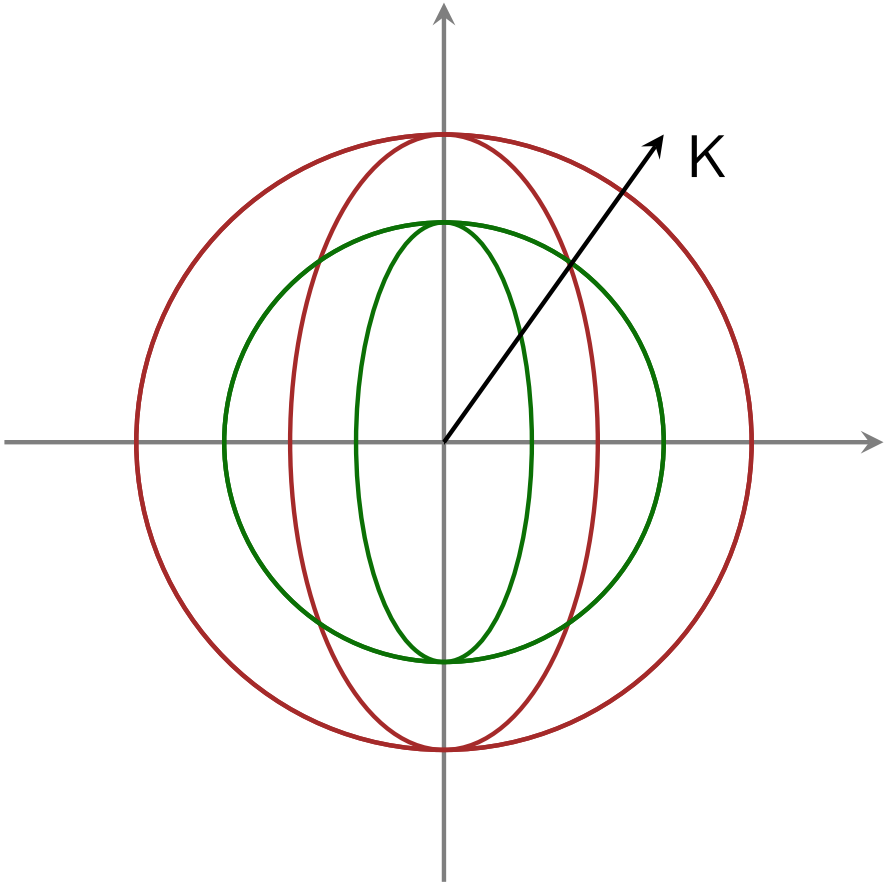
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Type II phase matching

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
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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
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Paper by *Bœuf* can help



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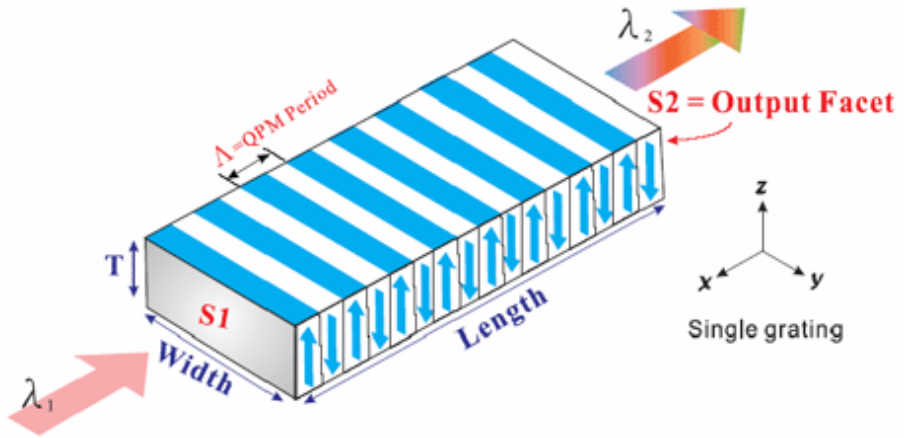
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Quasi phase matching in layered media

Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- d sign reversal



Wave solution

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

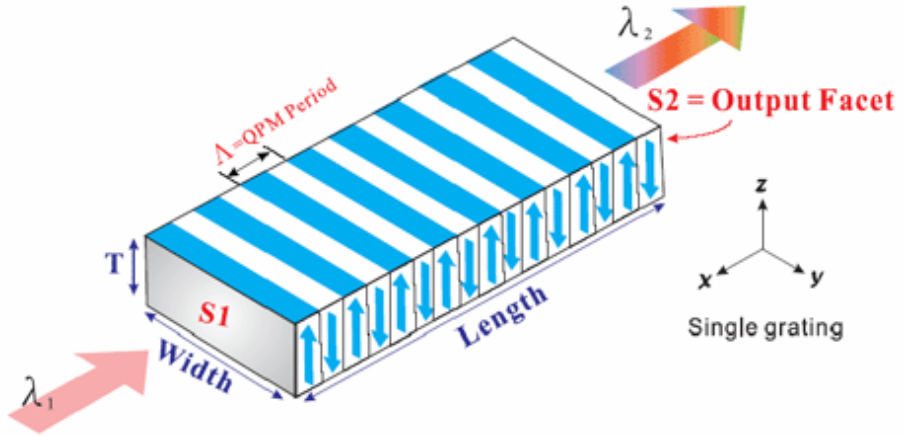
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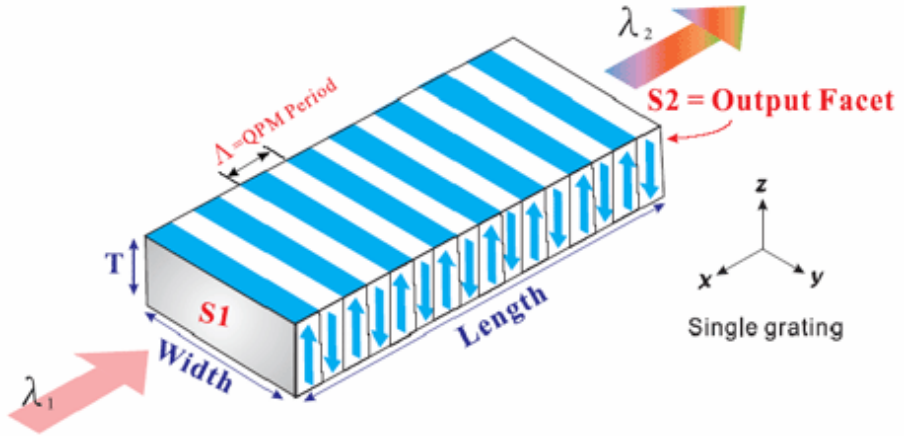
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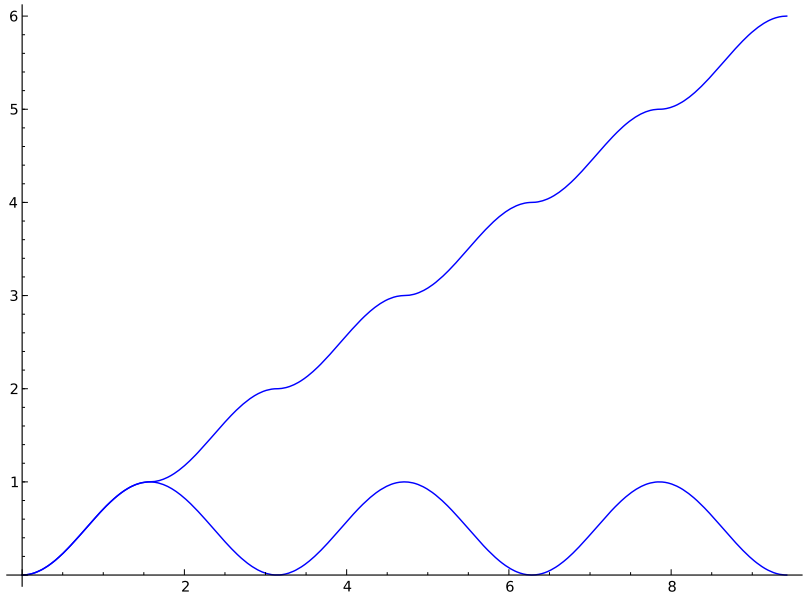
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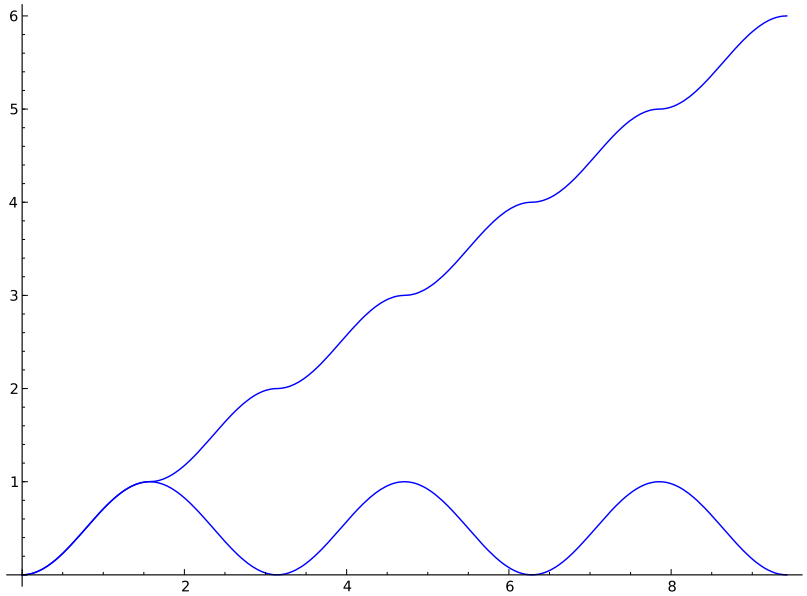
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Intsensity Solution

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