Second Harmonic Generation and related second order Nonlinear Optics

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Useful 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 : $\varepsilon_\nu = \varepsilon\left(\omega_\nu\right)$

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

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

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

SVA equation

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

Wave Propagation Equation under SVA approximation

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

- $\left[\frac{\partial E^{(\omega_1)}}{\partial z}\right]_i = +\frac{i\omega_1}{2}\eta_1 [d]_{ijk} \left[E^{(\omega_3)}\right]_j \overline{\left[E^{(\omega_2)}\right]_k} \exp\left(-i\Delta k z\right)$
- $\overline{\left[\frac{\partial E^{(\omega_2)}}{\partial z}\right]_k} = -\frac{i\omega_2}{2}\eta_2 [d]_{kij} \left[E^{(\omega_1)}\right]_i \overline{\left[E^{(\omega_3)}\right]_j} \exp\left(-i\Delta k z\right)$
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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

Assumptions

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

One equation remains

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Solving the SVA wave propagation equation

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$\Delta k \neq 0$

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

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

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

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

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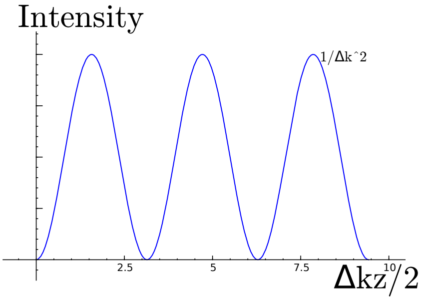
Phase match or not phase match

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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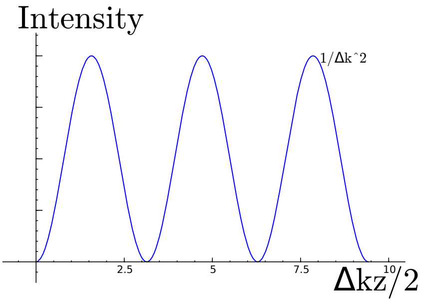
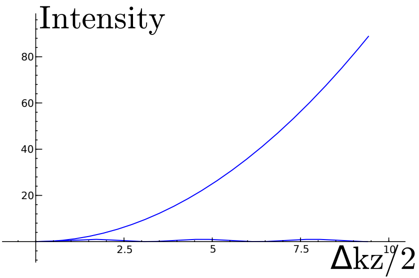


Phase match $\Delta k = 0$

- Intensity quadratic increase
- Approximations do not hold long



A Scalar Three Wave Interaction model

Further approximations to remove vectors

Simplifying notations

- Set 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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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

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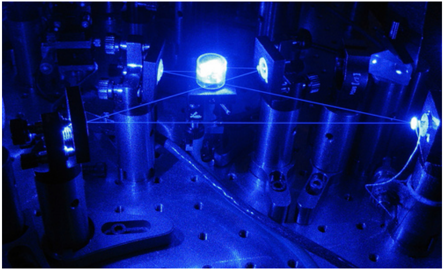

Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source flickr)

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

Phase matching $\Delta k = 0$

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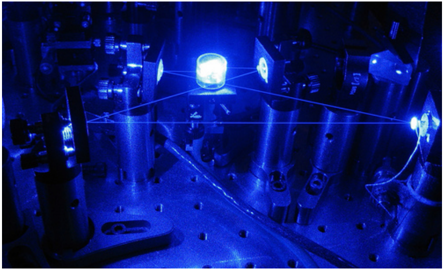

Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source `flickr`)

Second Harmonic Generation SHG

Sum frequency generation of two equal frequencies from the same source

One input beam counts for two

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

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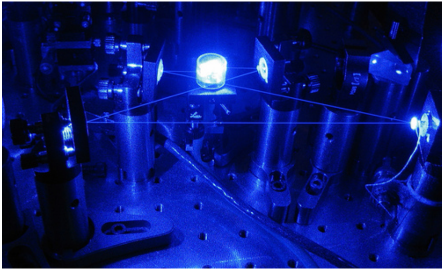

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- $\frac{\partial A_1}{\partial z} = +iK\overline{A_1}A_3$
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$A_1 \in \mathbb{R}$ $A_3 \in i\mathbb{R}$

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

- $\frac{\partial\left(A_1^2+\tilde{A}_3^2\right)}{\partial z} = 0$
- 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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Second Harmonic Beam Evolution

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

$\tilde{A}_3$ expression

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

I_3 expression

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

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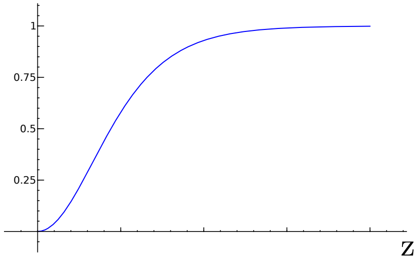
$I_3(z)/I_1(0)$ — $I_1(0) = \mathrm{constant}$



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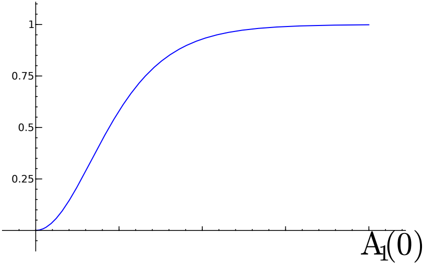
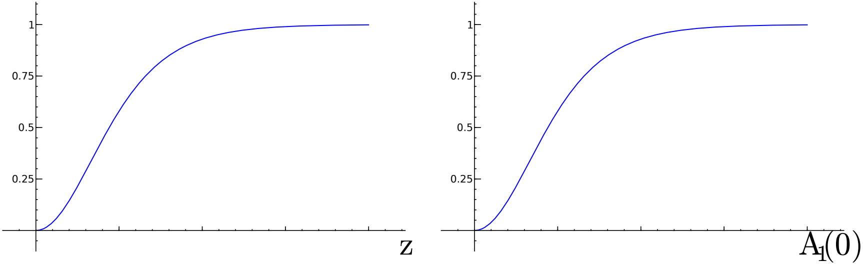


SHG conclusion

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
- We have retrieved Energy Conservation in spite of drastic approximations

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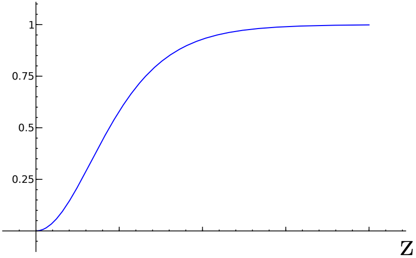

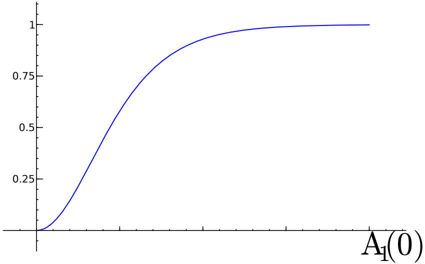

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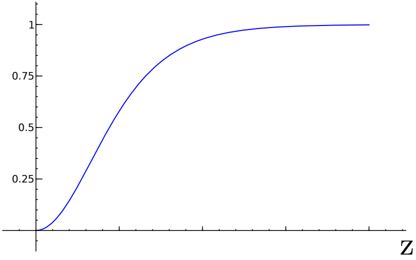

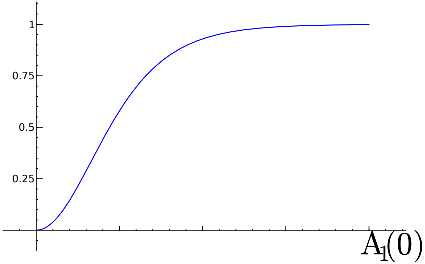

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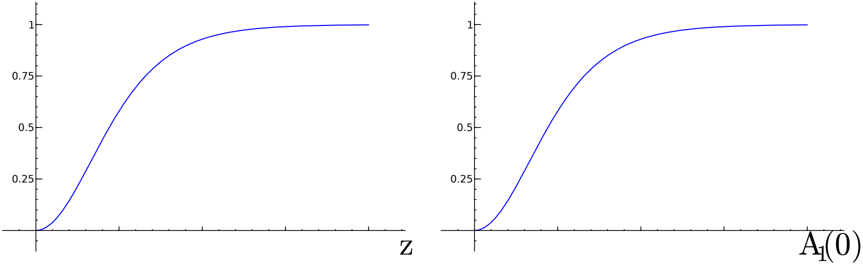

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Optical Parametric Amplifier OPA

Optical Amplification of a weak signal beam thanks to a 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

- $\frac{\partial A_1}{\partial z} = +iK_p\overline{A_2}$
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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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Solving the OPA equations

Phase matched equations

- $\frac{\partial A_1}{\partial z} = +iK_p\overline{A_2}$
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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 : $\overline{A_2(z)} = -iA_1(0)\sinh(K_pz)$

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Intensities

- Amplified signal :
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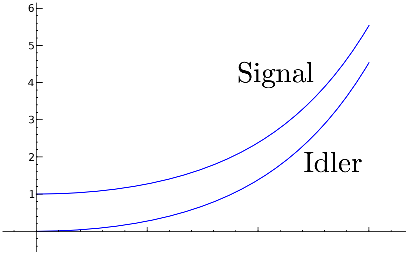
Amplification



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 !

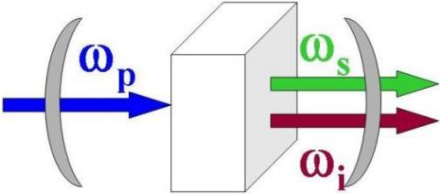


Figure: Optical Parametric Oscillator (source Cristal Laser)

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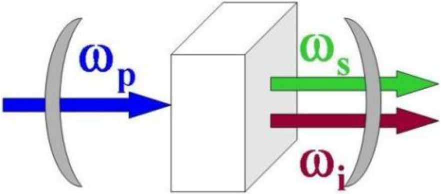

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1. Three-wave interaction
   - Assumptions framework
   - Three Wave propagation equation
   - Sum frequency generation
   - Scalar approximation
2. Non Linear Optics Application
   - Second Harmonic Generation
   - Optical Parametric Amplifier
   - 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

- $k_1 + k_2 = k_3 \Rightarrow \omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3$
- 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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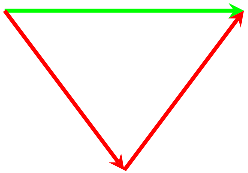
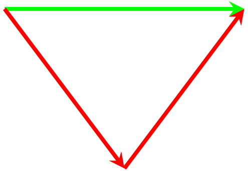
Non colinear phase matching



Non colinear phase matching

Use clever geometries

With reflections



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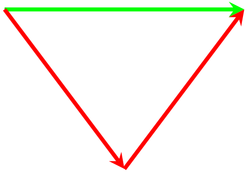
Or even more clever. . .



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

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

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Waves polarization

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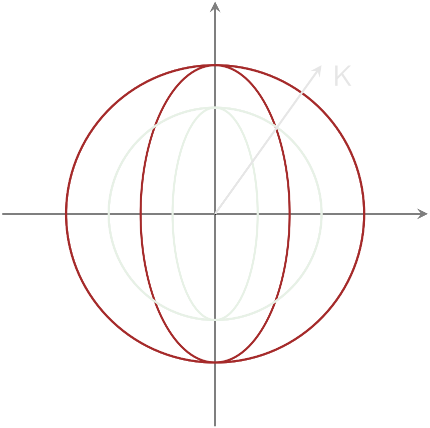

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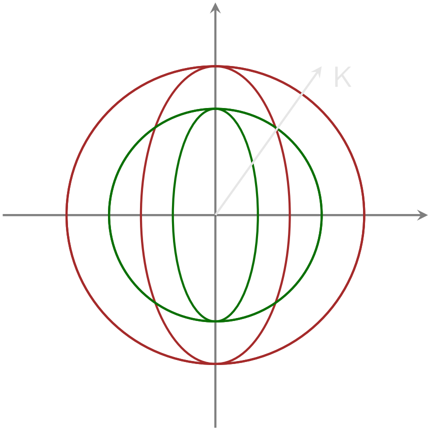

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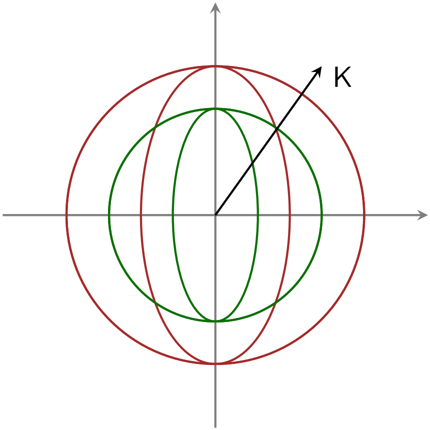
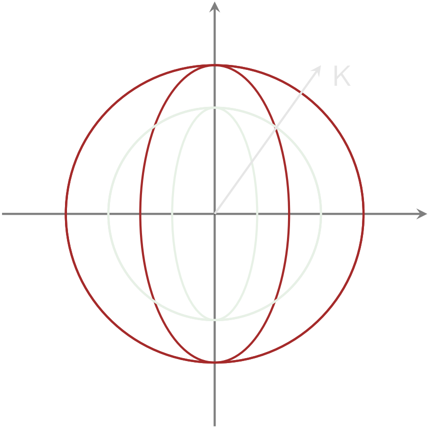


SHG Type I phase matching: a few numbers



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

$$\frac{1}{\tilde{n}_o^2(\theta)} = \frac{1}{\tilde{n}_o^2}$$

Solve the equation

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

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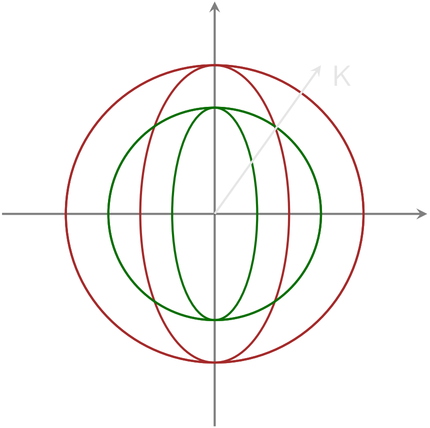

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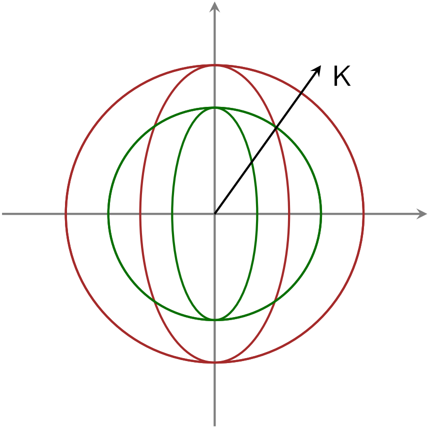

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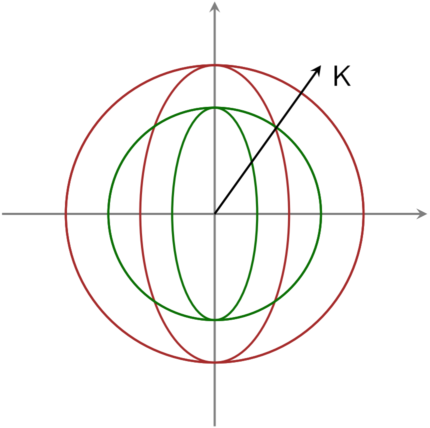

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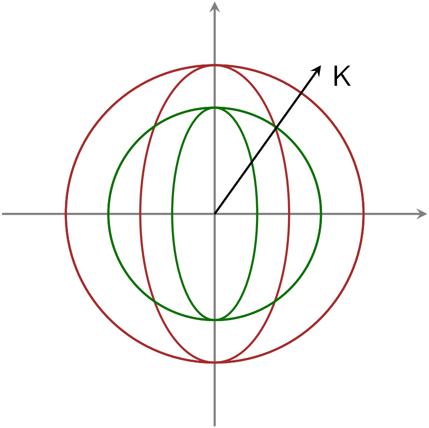

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In the three beam interaction, Type I was

- Both input beams ω_1 and ω_2 share the same polarization
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Another solution : Type II

- Input beams polarization are orthogonal
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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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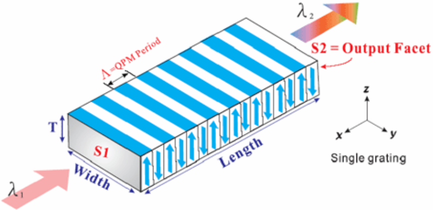
Quasi phase matching in layered media

Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- d sign reversal



Wave solution

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

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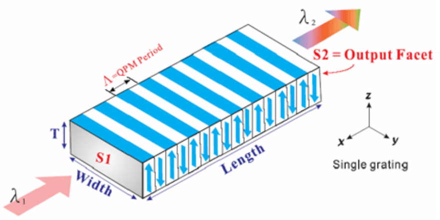

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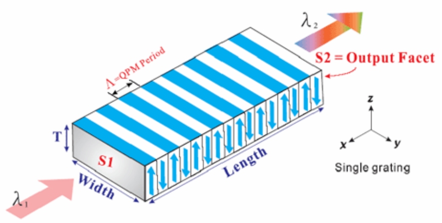

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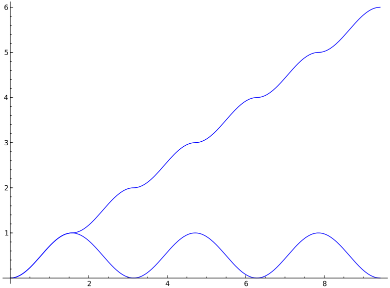

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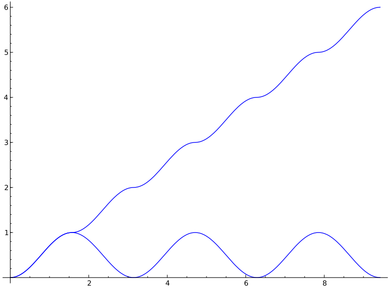

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Intsensity Solution

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