



# Second Harmonic Generation and related second order Nonlinear Optics

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November 12, 2012



## Useful reading. . .

[Y84, DGN91, LKW99]

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

Springer Verlag, Heidelberg, Germany, 1991.

W. Lauterborn, T. Kurz, and M. Wiesenfeldt.  
*Coherent Optics: Fundamentals and Applications*.

Springer-Verlag, New York, 1999.

A. Yariv and P. Yeh.

*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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# A classical Maxwell framework

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

## Maxwell Model

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

## Matter equations

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

## 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

## Three wave interaction assumptions

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

# Three wave interaction solution ansatz

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Sum of three waves

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

How good is this ansatz ?

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

Separate investigation at each frequency

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

# Wave propagation for frequency $\omega_\nu$ $\nu \in \{1, 2, 3\}$

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## 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  $\omega_\nu$  part  $\Leftrightarrow$  the plane wave part

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



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

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Reminder

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

temporal harmonic framework

for  $\omega_1$

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

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

Wave propagation equation

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



# The Slow Varying Approximation

Closely related to the paraxial approximation

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## 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  $y$

put it in maths...

$$\begin{aligned} \bullet \frac{\partial^2 E^{(\omega_1)}(z,t)}{\partial z^2} &= \frac{\partial^2}{\partial z^2} \mathcal{R}e \left( E^{(\omega_1)}(z) \exp(i(k_1 z - \omega_1 t)) \right) \\ \bullet \dots &= \mathcal{R}e \left( \left[ 2ik_1 \frac{\partial E^{(\omega_1)}(z)}{\partial z} - k_1^2 E^{(\omega_1)}(z) \right] e^{i(k_1 z - \omega_1 t)} \right) \end{aligned}$$



# Wave Propagation Equation under SVA approximation

Obtaining an envelope equation, which is simpler

• 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

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



# The three waves

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## 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 $i \rightarrow j \rightarrow k$

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

- $\left[ \frac{\partial E(\omega_1)}{\partial z} \right]_i = + \frac{i\omega_1}{2} \eta_1 [d]_{ijk} [E(\omega_3)]_j \overline{[E(\omega_2)]_k} \exp(-i\Delta kz)$
- $\overline{\left[ \frac{\partial E(\omega_2)}{\partial z} \right]_k} = - \frac{i\omega_2}{2} \eta_2 [d]_{kij} [E(\omega_1)]_i \overline{[E(\omega_3)]_j} \exp(-i\Delta kz)$
- $\left[ \frac{\partial E(\omega_3)}{\partial z} \right]_j = + \frac{i\omega_3}{2} \eta_3 [d]_{jki} [E(\omega_2)]_k [E(\omega_1)]_i \exp(i\Delta kz)$



## 6 equations for various quadratic phenomena

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

6x equations

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

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

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

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

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
- ...

# Example : sum frequency generation

Input beams assumed constant

Undepleted pump approximation

## Assumptions

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

$$\bullet \frac{\partial [E^{(\omega_1)}]_i}{\partial z} = \overline{\frac{\partial [E^{(\omega_2)}]_k}{\partial z}} = 0$$

## One equation remains

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



## Solving the SVA wave propagation equation

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Equation to solve

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

 $\Delta k \neq 0$ 

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

Wave solution

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

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

Intensity

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

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

 $\Delta k = 0$ 

$$y' = a \Rightarrow y = ax$$

Wave solution

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

$$\frac{i\omega_3}{2} \eta_3 [d]_{jki} [E(\omega_2)]_k [E(\omega_1)]_i z$$

Intensity

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

$$\omega_3^2 \eta_3^2 [d^2]_{jki} |E(\omega_2)|_k^2 |E(\omega_1)|_i^2 z^2$$

# Phase match or not phase match

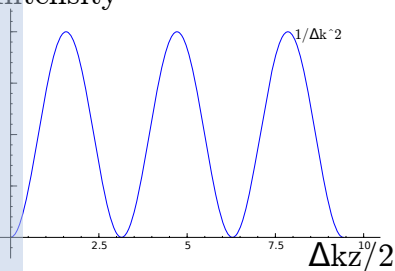
Phase matching is a key issue to sum frequency generation

Phase mismatch

$$\Delta k \neq 0$$

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

Intensity

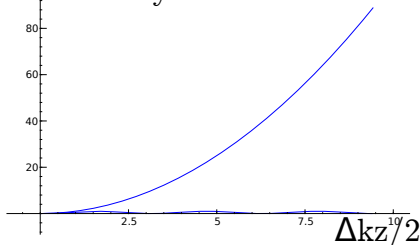


Phase match

$$\Delta k = 0$$

- Intensity quadratic increase
- Approximations do not hold long

Intensity



# A Scalar Three Wave Interaction model

Further approximations to remove vectors

## Simplifying notations

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

## Scalar three wave interaction

- $\frac{\partial A_1}{\partial z} = +iK \overline{A_2} A_3 \exp(-i\Delta kz)$
- $\frac{\partial A_2}{\partial z} = -iK A_1 \overline{A_3} \exp(+i\Delta kz)$
- $\frac{\partial A_3}{\partial z} = +iK A_2 A_1 \exp(+i\Delta kz)$

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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

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

Phase matching

$$\Delta k = 0$$

- $\frac{\partial A_1}{\partial z} = +iK\bar{A}_1 A_3$
- $\frac{\partial A_3}{\partial z} = +iK A_1^2$

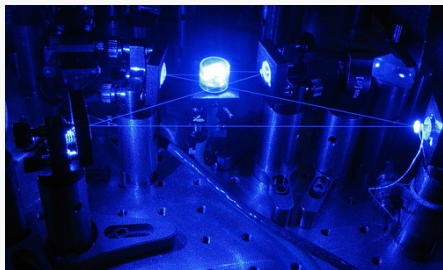


Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source flickr)



# Second Harmonic Beam Generation

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Remember...

$$\Delta k = 0$$

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

$$\frac{\partial A_3}{\partial z} = +iK A_1^2$$

 $A \in \mathbb{R}$ 
 $A_3 \in i\mathbb{R}$ 

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

$$A_1 = \overline{A_1}$$

Coupled 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

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

 $\bullet$  This is Energy Conservation

Start with no harmonic

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

 $\tilde{A}_3$  equation

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



# Second Harmonic Beam Evolution

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$A_3$  equation

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

$A_3$  expression

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

$I_3$  expression

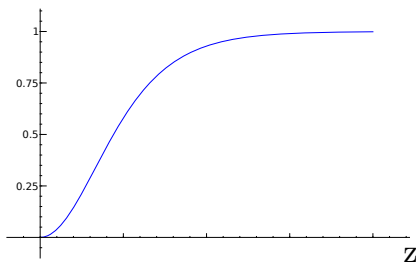
$$I_3(z) = I_1(0) \tanh^2(KA_1(0)z)$$

$$I_1(z) = I_1(0) - I_3(z)$$

$$I_1(z) = I_1(0) \operatorname{sech}^2(KA_1(0)z)$$

$I_3(z) / I_1(0)$

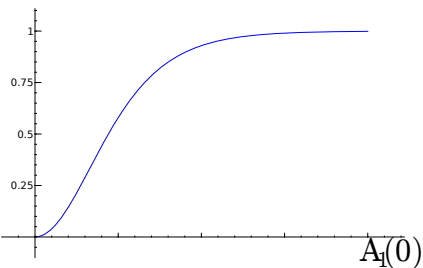
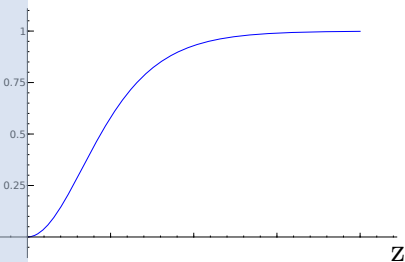
$I_1(0) = \text{constant}$



# SHG conclusion

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## Second Harmonic Beam Evolution



### Surprisingly...

- 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

# Optical Parametric Amplifier

## OPA

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

### Signal beam amplification

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

### Undepleted pump approximation

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

### Phase matched equations

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

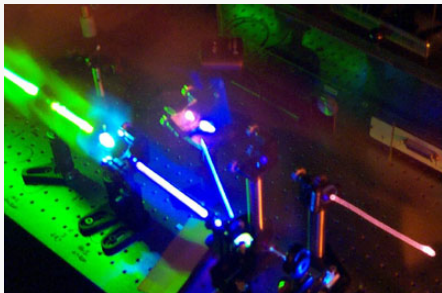


Figure: White light continuum seeded optical parametric amplifier (OPA) able to generate extremely short pulses. (source Freie Universität Berlin)



# Solving the OPA equations

## Phase matched equations

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

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

## Initial conditions

A weak signal :  $A_1(0) \neq 0$

No idler :  $A_2(0) = 0$

## Amplitude solution

Amplified signal :

$$A_1(z) = A_1(0) \cosh(K_p z)$$

Idler :

$$A_2(z) = -iA_1(0) \sinh(K_p z)$$

## Intensities

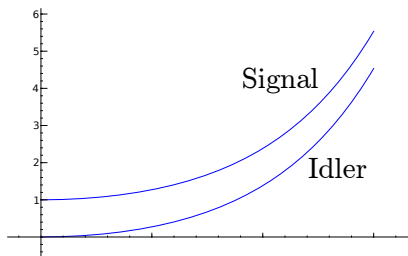
- Amplified signal :

$$I_1(z) = I_1(0) \cosh^2(K_p z)$$

- Idler :

$$I_2(z) = I_1(0) \sinh^2(K_p z)$$

## Amplification



# Optical Parametric Oscillator

## OPO

Use Optical Parametric Amplification to make a tunable laser

OPA pumped with  $\omega_3$

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

Frequency tunable laser

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

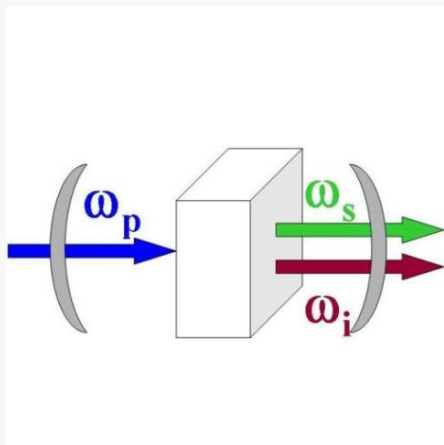


Figure: Optical Parametric Oscillator  
(source Cristal Laser)



## Colinear (scalar) phase matching

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### 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



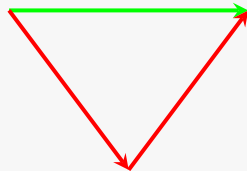


# Non colinear phase matching

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Use clever geometries

With reflections



# SHG Type I Phase Matching

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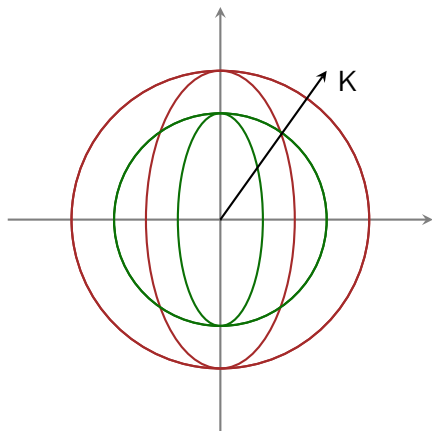
## Waves polarization

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

## Type I phase matching

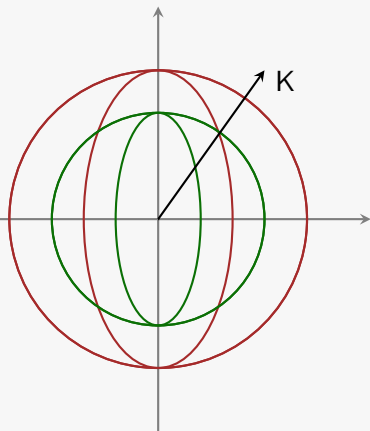
- One refraction index for Fundamental
- The other for Second Harmonic
- They must be equal
- Propagate in the right direction

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



# SHG Type I phase matching: a few numbers

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Fundamental index ellipsoid section

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

Harmonic index ellipsoid section

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

Solve the equation

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



## Type II phase matching

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the three beam interaction, Type I was

- Both input beams  $\omega_1$  and  $\omega_2$  share the same polarization
- The generated beam  $\omega_3$  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 ?



# Phase matching in bi-axial crystals

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A hard task

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

A paper by *Bœuf* can help

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

Calculating characteristics of noncolinear phase matching in uniaxial and biaxial crystals.

*Optical Engineering*, 39(4):1016–1024, 2000.

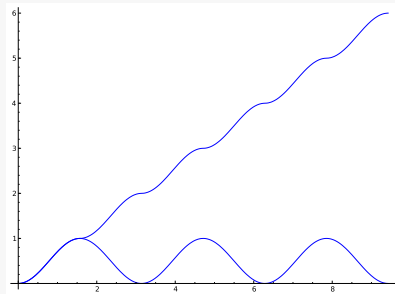


# Quasi phase matching in layered media

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## Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- $d$  sign reversal



Intensity Solution

$$\Delta k \Lambda = \pi$$

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

