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Second Harmonic Generation and related second order Nonlinear Optics

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Useful reading. . .

[YY84, 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

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

Wave equation in isotropic medium

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

Solutions to be found only in a specific framework:

We look here for :

- Quadratic non-linearity
- Three wave interaction

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 $\omega_1 + \omega_2 = \omega_3$

Three wave interaction solution ansatz

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 : $k_{\nu}^2 = \mu_0 \varepsilon_{\nu} \omega_{\nu}^2$
- 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 $\omega_{\nu} : P_{NL}^{\omega_{\nu}}$
- Three separate equations

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

3 Perturbed wave equations

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

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

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

Non Linear Polarisation P_{NL} in harmonic framework

Reminder

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

Temporal harmonic framework

for ω_1

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

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

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

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...

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

Wave Propagation Equation under SVA approximation

Obtaining an envelope equation, which is simpler

Non SVA wave propagation equation

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

SVA equation

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

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 $i \rightarrow j \rightarrow k$

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

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

6 equations for various quadratic phenomena

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

Six 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 $z = 0$: $[E^{(\omega_3)}(z = 0)]_j = 0$
- $\frac{\partial [E^{(\omega_1)}]_i}{\partial z} = \overline{\frac{\partial [E^{(\omega_2)}]_k}{\partial z}} = 0$

One equation remains

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

Solving the SVA wave propagation equation

Equation to solve

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

$$\Delta k \neq 0$$

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

Wave solution

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

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

Intensity

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

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

$$\Delta k = 0$$

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

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

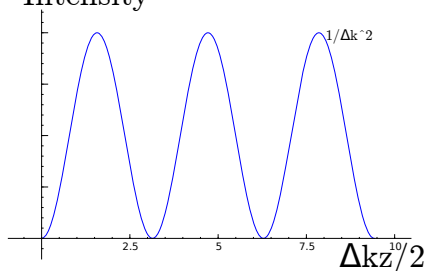
Phase matching is a key issue to sum frequency generation

Phase mismatch

$$\Delta k \neq 0$$

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

Intensity

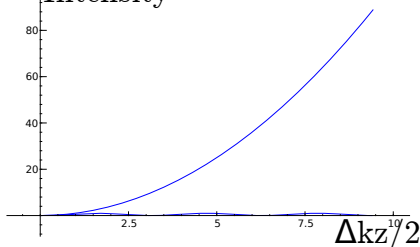


Phase match

$$\Delta k = 0$$

- 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

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

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

$$\Delta k = 0$$

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

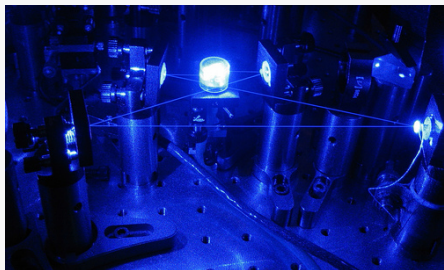


Figure: Closeup of a BBO crystal inside a resonant build-up ring cavity for frequency doubling 461 nm blue light into the ultraviolet. (source flickr)

Second Harmonic Beam Generation

Remember...

$$\Delta k = 0$$

- $\frac{\partial A_1}{\partial z} = +iK\overline{A_1}A_3$
- $\frac{\partial A_3}{\partial z} = +iKA_1^2$

$A_1 \in \mathbb{R}$

$A_3 \in i\mathbb{R}$

- $A_3 = i\tilde{A}_3 \Rightarrow \tilde{A}_3 \in \mathbb{R}$
- $A_1 = \overline{A_1}$

Real equations

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

Multiply by A_1 and \tilde{A}_3

Sum

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

Start with no harmonic

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

\tilde{A}_3 equation

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

Second Harmonic Beam Evolution

\tilde{A}_3 equation

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

\tilde{A}_3 expression

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

I_3 expression

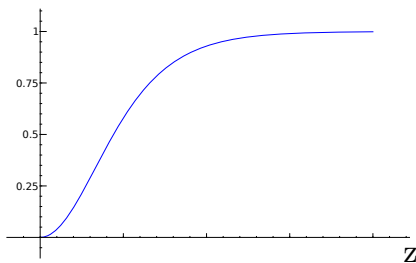
$$I_3(z) = I_1(0) \tanh^2(KA_1(0)z)$$

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

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

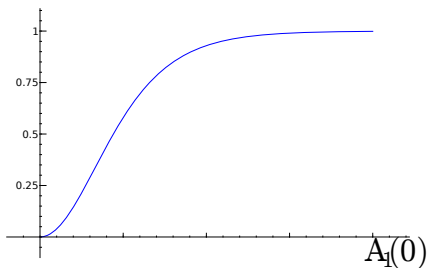
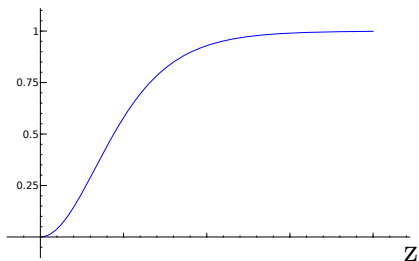
$I_3(z) / I_1(0)$

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



SHG conclusion

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

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

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

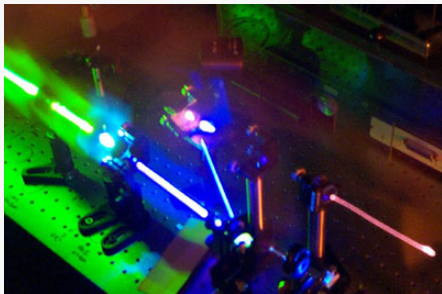


Figure: White light continuum seeded optical parametric amplifier (OPA) able to generate extremely short pulses.
(source Freie Universität Berlin)

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 : $A_2(0) = 0$

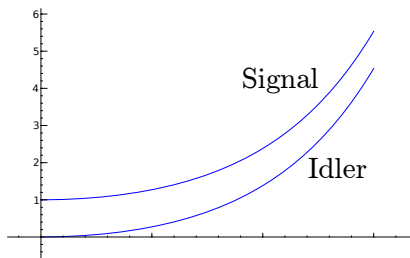
Amplitude solution

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

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 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 !

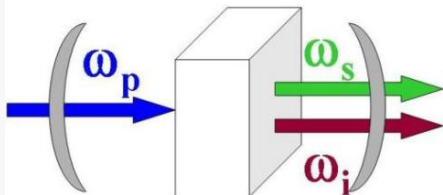


Figure: Optical Parametric Oscillator
(source Cristal Laser)

Colinear (scalar) phase matching

Phase matching for co-propagation waves

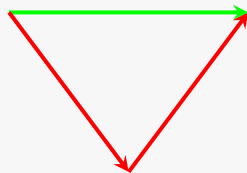
- $k_1 + k_2 = k_3 \Rightarrow \omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3$
- for SHG : $2k_1 = k_3 \Rightarrow n_1 = n_3$
- The last is **never** achieved, due to normal dispersion: $n_1 < n_3$

One and only solution

Use birefringent crystals and different polarizations

Non colinear phase matching

Use clever geometries
With reflections



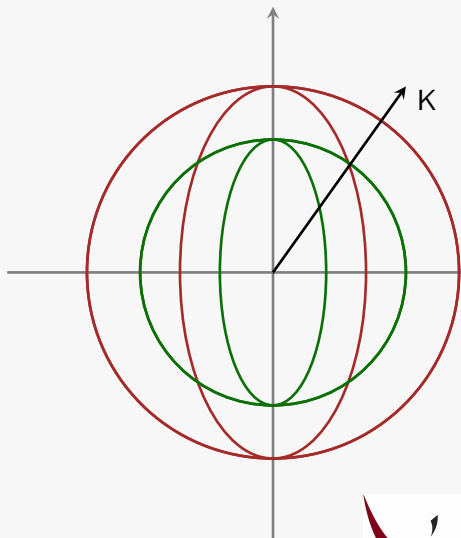
SHG Type I Phase Matching

Waves polarization

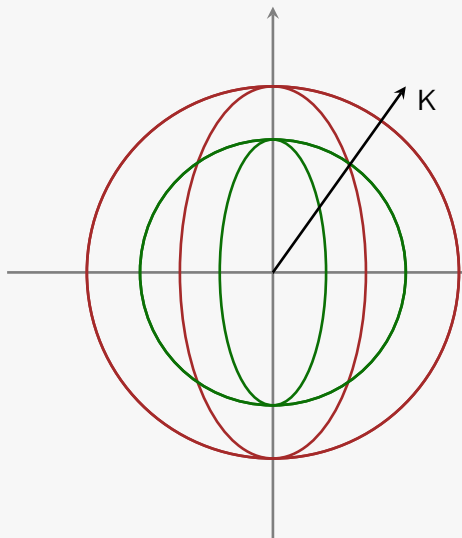
- 1 incident wave counts for 2
- They share the same polarization
- Second Harmonic polarization is orthogonal

Type I phase matching

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



SHG Type I phase matching: a few numbers



Fundamental index ellipsoid section

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

Harmonic index ellipsoid 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

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

Type II phase matching

In the three beam interaction, Type I was

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

Another solution : Type II

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

Phase matching in bi-axial crystals

A hard task

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

Paper by *Bœuf* can help



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

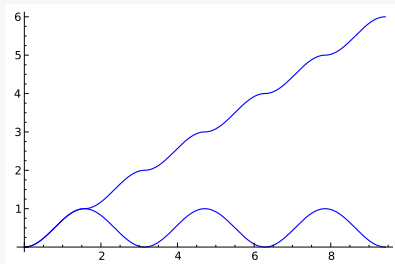
Calculating characteristics of noncolinear phase matching in uniaxial and biaxial crystals.

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

Quasi phase matching in layered media

Periodically Poled Lithium Niobate

- Periodic Domain Reversal
- d sign reversal



Wave solution

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

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

Intensity Solution

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

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