Second harmonic generation

Use a single input field:

$$E_1(z) = E_2(z)$$

Then:

$$\frac{d}{dz}E_3(z) = -\frac{\sigma}{2}\sqrt{\frac{\mu}{\varepsilon_3}}E_3(z) - \frac{i\omega_3}{2}\sqrt{\frac{\mu}{\varepsilon_3}}dE_1^2(z)e^{-i(2k_1-k_3)z}$$

Assume now:

- There is a nonlinearity d (only for certain symmetry)
- No absorption in the medium, so σ=0
- Only little production of wave ω_3, so no back-conversion
- Wave vector mismatch is

$$\Delta k = k^{(2\omega)} - 2k^{(\omega)}$$

The coupled wave equation can be integrated:

$$E^{(2\omega)}(z) = -i\omega\sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}}dE^2(\omega)\int e^{i\Delta kz}dz$$

Conditions

1) Integration for 0 to L (length of medium)
2) And boundary $E^{(2\omega)}(0) = 0$

Result of integration:

$$E^{(2\omega)}(L) = -\omega\sqrt{\frac{\mu}{\varepsilon^{(2\omega)}}}dE^2(\omega)\frac{e^{i\Delta kL}-1}{\Delta k}$$

Output of second harmonic is:

$$E^{(2\omega)}(L)E^{(2\omega)}(L)^* =$$

$$\frac{\omega^2\mu}{n^2\varepsilon_0}d^2|E(\omega)|^4 L^2\frac{\sin^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2}$$

Power at second harmonic:

$$P^{(2\omega)} \propto \omega^2 d^2 L^2\frac{\sin^2\left(\frac{\Delta kL}{2}\right)}{\left(\frac{\Delta kL}{2}\right)^2}\frac{P^{(\omega)2}}{A}$$

Second harmonic power; conditions

Conversion efficiency:

$$\eta_{SHG} = \frac{P^{(2\omega)}}{P^{(\omega)}} \propto \omega^2 d^2 L^2 \frac{\sin^2\left(\frac{\Delta k L}{2}\right)}{\left(\frac{\Delta k L}{2}\right)^2} \frac{P^{(\omega)}}{A}$$

1) Second harmonic produced is proportional to

$$P^{(2\omega)} \propto P^{(\omega)2}$$

nonlinear power production

2) Efficiency is proportional to d^2 or

$$\left|\chi^{(2)}\right|^2$$

3) Efficiency is proportional to L^2 and a sinc function

$$\eta_{SHG} \propto L^2 \operatorname{sinc}\left(\frac{\Delta k L}{2}\right)$$

4) Efficiency is optimal if

$$\Delta k = 0$$

This is the "phase-matching condition" cannot be met, because:

$$k^{(2\omega)} \neq 2k^{(\omega)}$$

Use: $k = \frac{n\omega}{c}$

$$k^{(2\omega)} = \frac{2n^{(2\omega)}\omega}{c} \qquad 2k^{(\omega)} = \frac{2n^{(\omega)}\omega}{c}$$

And dispersion in the medium:

$$n^{(2\omega)} > n^{(\omega)}$$

So always $\Delta k \neq 0$

Physics: two waves with

$$E_\omega(z,t) = E_\omega \exp\left[i\omega t - ik^{(\omega)}z\right]$$

$$E_{2\omega}(z,t) = E_{2\omega} \exp\left[2i\omega t - ik^{(2\omega)}z\right]$$

will run *out of phase*

Coherence length and Maker fringes

After a distance the waves will run out of phase

$$\Delta k l = \pi$$

Then the amplitude is at maximum.
The wave will die out in:

$$L_c = 2l$$

The coherence length:

$$L_c = \frac{2\pi}{\Delta k} = \frac{2\pi}{k^{(2\omega)} - 2k^{(\omega)}} = \frac{\pi c}{2\omega\left(n^{(2\omega)} - n^{(\omega)}\right)} = \frac{\lambda}{4\left(n^{(2\omega)} - n^{(\omega)}\right)}$$

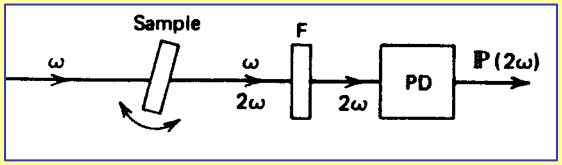
Typical values

$$\lambda = 1 \mu m$$

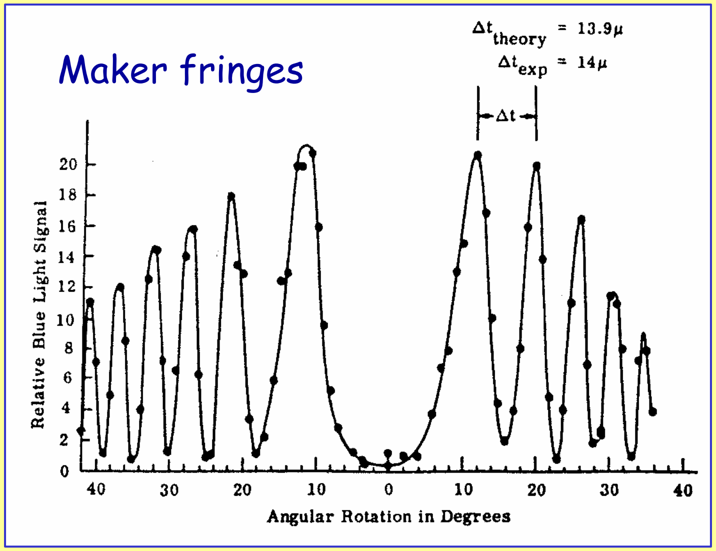
$$n^{(2\omega)} - n^{(\omega)} \approx 10^{-2}$$

$$L_c = 25 \mu m$$

Experiment:



P.D. Maker, R.W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Lett. **8**, 19 (1962).



Only effective length of L_c can be used
(Note: non-sinusoidal behavior due to "non-critical phase matching")

Maxwell equations for anisotropic media

Induced polarization in a medium:

$$\vec{P} = \varepsilon_0 \chi \vec{E}$$

Susceptibility is tensor of rank 2, causing the P and E vectors to have different directions

$$P_1 = \varepsilon_0 \left(\chi_{11} E_1 + \chi_{12} E_2 + \chi_{13} E_3 \right)$$
$$P_2 = \varepsilon_0 \left(\chi_{21} E_1 + \chi_{22} E_2 + \chi_{23} E_3 \right)$$
$$P_3 = \varepsilon_0 \left(\chi_{31} E_1 + \chi_{32} E_2 + \chi_{33} E_3 \right)$$

Elements of tensor depend on coordinate frame;

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 \left(1 + \chi_{ij} \right) \vec{E} = \varepsilon_{ij} \vec{E}$$

With permitivity tensor $\vec{\vec{\varepsilon}}_{ij}$

Monochromatic plane wave with perpendicular:

$$\vec{E} \exp[i\omega t - i\vec{k} \cdot \vec{r}]$$

$$\vec{H} \exp[i\omega t - i\vec{k} \cdot \vec{r}]$$

Wavefront vector

$$\vec{k} = \frac{n\omega}{c} \vec{s}$$

Maxwell's equations (non-magnetic media)

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \qquad \vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t}$$

Derivatives:

$$\vec{\nabla} \rightarrow -i\vec{k} = -i \frac{n\omega}{c} \vec{s} \qquad \frac{\partial}{\partial t} \rightarrow i\omega$$

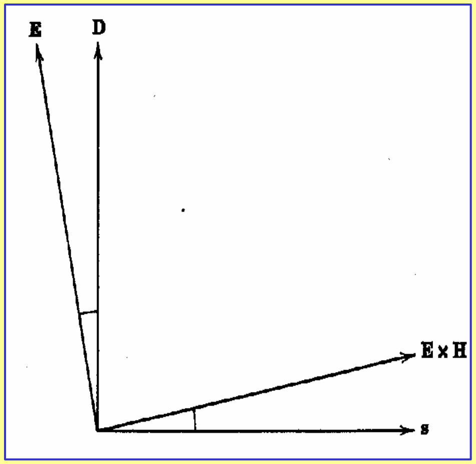
For the plane waves:

$$\vec{k} \times \vec{E} = \mu_0 \omega \vec{H} \qquad \vec{k} \times \vec{H} = -\omega \vec{D}$$

Two vectors orthogonal to k

$$\vec{k} \perp \vec{H} \qquad \vec{k} \perp \vec{D}$$

Group and Phase velocity



H and D perpendicular to wave vector
Verify:

$$\vec{E} \perp \vec{H}$$

Further

$$\vec{D} = \vec{\vec{\varepsilon}}\vec{E}$$

If ε is a scalar then D and E parallel,
but this is not the case in general

Poynting vector: $\vec{S} = \vec{E} \times \vec{H}$

Is not along k-vector

→

Group Velocity is not equal to Phase Velocity
- in magnitude
- in direction

Fresnel equations

Verify: $-\vec{k}\times\vec{k}\times\vec{E} = \vec{\nabla}\times\vec{\nabla}\times\vec{E} = \omega^2\mu\vec{D}$

Use: $\vec{k}\times\vec{k}\times\vec{E} = \vec{k}\left(\vec{k}\cdot\vec{E}\right) - \vec{E}\left(\vec{k}\cdot\vec{k}\right)$

$$\longrightarrow \quad \vec{D} = n^2\varepsilon_0\left[\vec{E} - \vec{s}\left(\vec{s}\cdot\vec{E}\right)\right]$$

Choose coordinate frame (x,y,z) along principal dielectric axes

$$\begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \begin{pmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix}\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

Permittivities ε_i differ along axes

$$D_i = n^2\varepsilon_0\left[\frac{D_i}{\varepsilon_i} - \left(\vec{s}\cdot\vec{E}\right)\right]$$

Hence: $$D_i = \frac{\varepsilon_0\left(\vec{s}\cdot\vec{E}\right)}{\dfrac{1}{n^2} - \dfrac{\varepsilon_0}{\varepsilon_i}}$$

Form the scalar product $\vec{s}\cdot\vec{D} = 0$

$\longrightarrow$ Fresnel's equation

$$\frac{s_x^2}{\dfrac{1}{n^2} - \dfrac{\varepsilon_0}{\varepsilon_x}} + \frac{s_y^2}{\dfrac{1}{n^2} - \dfrac{\varepsilon_0}{\varepsilon_y}} + \frac{s_z^2}{\dfrac{1}{n^2} - \dfrac{\varepsilon_0}{\varepsilon_z}} = 0$$

Equation is quadratic in n and will have two solutions *n'* and *n"*

Two waves *D'(n')* and *D"(n")* obey the equation

$$\mathbf{D}'\cdot\mathbf{D}'' = \varepsilon_0^{\ 2}\left(\mathbf{s}\cdot\mathbf{E}\right)^2\left\langle \sum_{x,y,z}\frac{s_\alpha^2}{\left(\dfrac{1}{n'^2} - \dfrac{\varepsilon_0}{\varepsilon_\alpha}\right)\left(\dfrac{1}{n''^2} - \dfrac{\varepsilon_0}{\varepsilon_\alpha}\right)}\right\rangle$$

$$= \varepsilon_0^{\ 2}\left(\mathbf{s}\cdot\mathbf{E}\right)^2\frac{\left(n'n''\right)^2}{\left(n'^2 - n''^2\right)}\left\langle \sum_{x,y,z}\left[\frac{s_\alpha^2}{\left(\dfrac{1}{n'^2} - \dfrac{\varepsilon_0}{\varepsilon_\alpha}\right)} + \frac{s_\alpha^2}{\left(\dfrac{1}{n''^2} - \dfrac{\varepsilon_0}{\varepsilon_\alpha}\right)}\right]\right\rangle$$

Summation α is over x,y,z

$$\longrightarrow \quad \vec{D}'\cdot\vec{D}'' = 0$$

Anisotropic crystal can transmit two waves with perpendicular parallel polarizations (and any linear combination of these two)

Refraction at boundary of anisotropic crystal

Incident beam is always decomposed into two eigenmodes of the anisotropic crystal

$$\vec{D}'(n') \qquad\qquad \vec{D}''(n'')$$

These modes are orthogonal to each other.
Each of the two modes undergoes refraction with its index n' or n''

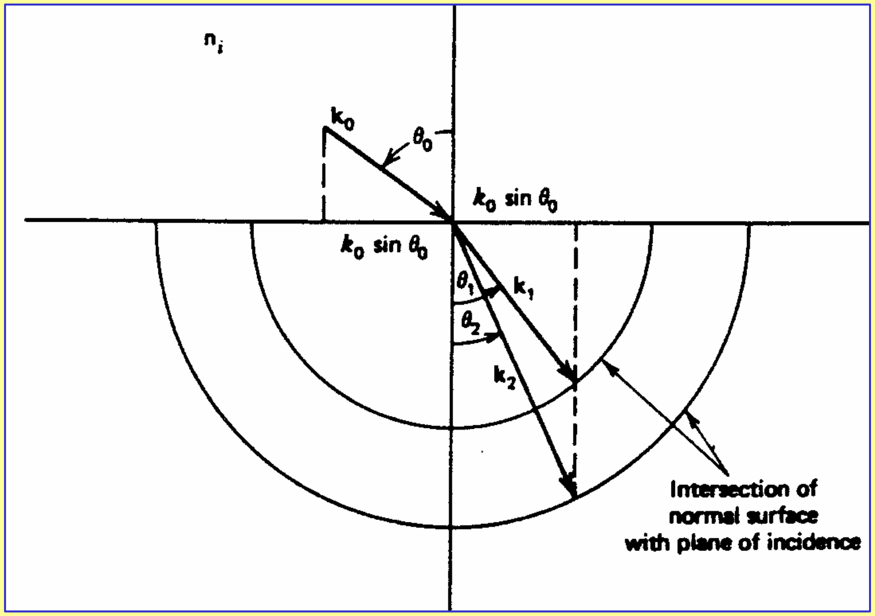
Hence:

$$k_0 \sin\theta_0 = k_1 \sin\theta_1 = k_2 \sin\theta_2$$

This is:

Double refraction

Birefringence



The index ellipsoid

Energy stored in an electric field in a medium:

$$U_e = \frac{1}{2}(\mathbf{E} \cdot \mathbf{D})$$

With: $D_i = \varepsilon_i E_i$

$$\frac{D_x^2}{\varepsilon_x} + \frac{D_y^2}{\varepsilon_y} + \frac{D_z^2}{\varepsilon_z} = 2U_e$$

This is a surface (ellipsoid) of constant energy

Define a normalized polarization vector:

$$\vec{r} = \vec{D}\sqrt{2U_e}$$

Index ellipsoid:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1$$

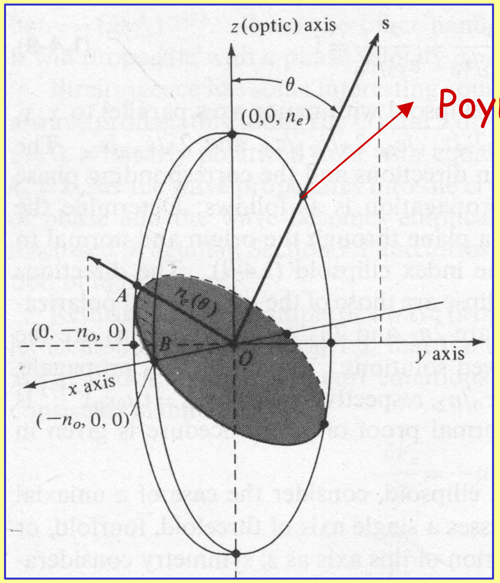
Three-dimensional body to find two indices of refraction for the two waves D

Uni-axial crystal:

$$n_0^2 = \frac{\varepsilon_x}{\varepsilon_0} = \frac{\varepsilon_y}{\varepsilon_0} \qquad n_e^2 = \frac{\varepsilon_z}{\varepsilon_0}$$

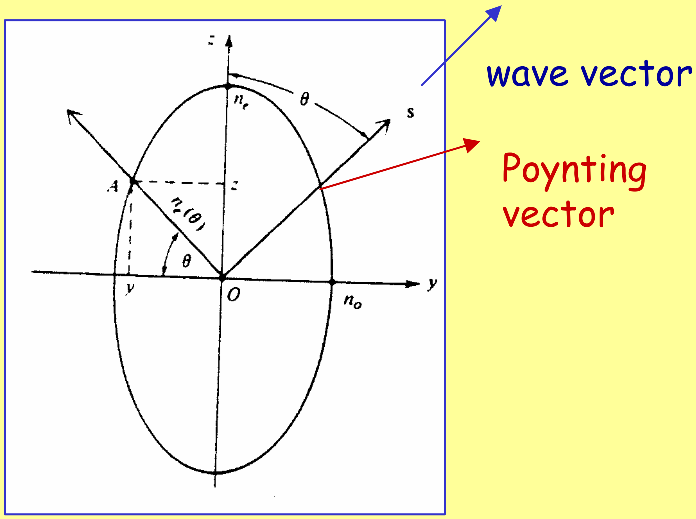
Index becomes:

$$\frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1$$



Poynting vector

Birefringent media



Two allowed polarization directions
-one polarized along the x-axis;
polarization vector perpendicular to the optic axis
ordinary wave; it transmits with index *no*.
-one polarized in the x-y plane but perpendicular to s;
polarization vector in the plane with
the optic axis is called the *extraordinary wave*.

For an arbitrary angle:

$$x = n_0 \qquad y = n_e(\theta)\cos\theta \qquad z = n_e(\theta)\sin\theta$$

Projection of the ellipsoid on x=0

$$\frac{y^2}{n_0^2} + \frac{z^2}{n_e^2} = 1$$

Insert:

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

So index depends on propagation
of wave vector (θ)

Birefringence	$n_e > n_0$	positive
	$n_e < n_0$	negative

Phase matching in Birefringent media

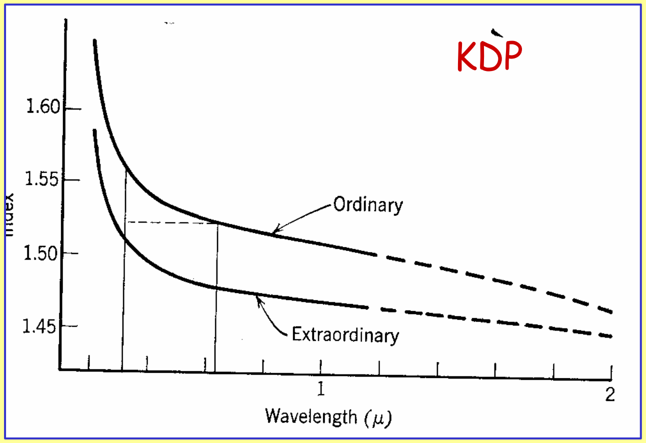
There exists an ordinary wave with

$$n_0$$

And an extra-ordinary wave with

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

Both undergo dispersion



Phase-matching, or Δk=0 can be reached now; required is

$$n^{\omega} = n^{2\omega}$$

In case of (for KDP) $n_e < n_0$

$$n_e^{2\omega}(\theta_m) = n_o^{\omega}$$

Equation to find the phase-matching angle:

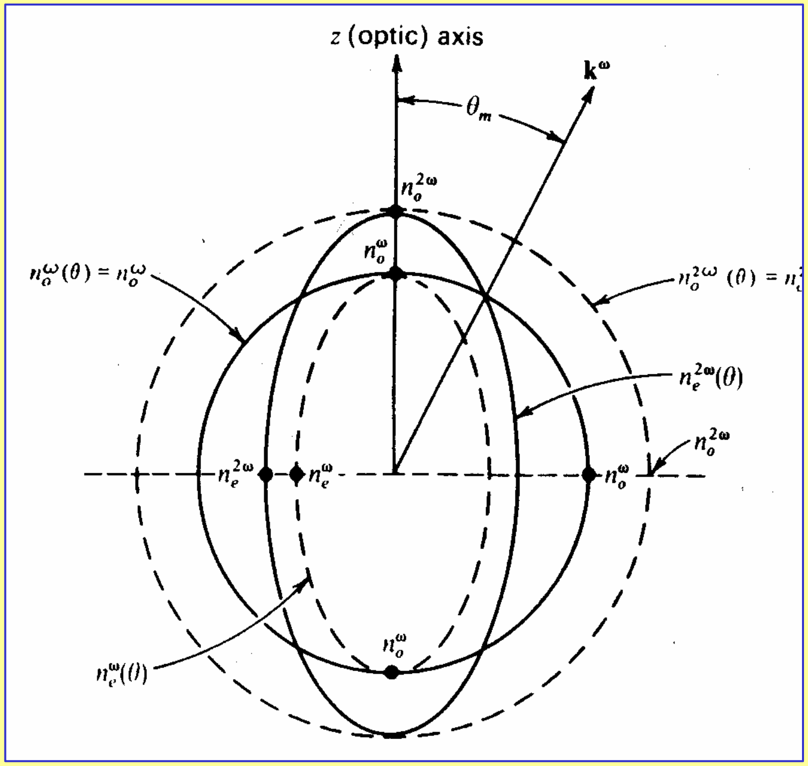
$$n_e^{2\omega}(\theta_m) = \frac{n_e^{2\omega} n_o^{2\omega}}{\sqrt{\left(n_o^{2\omega}\right)^2 \sin^2\theta_m + \left(n_e^{2\omega}\right)^2 \cos^2\theta_m}}$$

Solve for sinθ

$$\sin^2\theta_m = \frac{\left(n_o^{\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

Phase matching in Birefringent media

Graphical: index ellipsoid including dispersion



$$\sin^2\theta_m = \frac{\left(n_o^\omega\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

TYPE I phase matching

$E_o^\omega + E_o^\omega \rightarrow E_e^{2\omega}$	negative birefringence
$E_e^\omega + E_e^\omega \rightarrow E_o^{2\omega}$	positive birefringence

TYPE II phase matching

$E_o^\omega + E_e^\omega \rightarrow E_e^{2\omega}$	negative birefringence
$E_o^\omega + E_e^\omega \rightarrow E_o^{2\omega}$	positive birefringence

Type I → polarization of second harmonic is perpendicular to fundamental

Type II → can be understood as sumfrequency mixing

Phase matching and the "opening angle"

Consider Type I phase-matching and a negatively birefringent crystal.
Phase matching

$$\Delta k = \frac{2\omega}{c}\left[n_e^{2\omega}(\theta) - n_o^{\omega}\right] = 0$$

This works for a certain angle θ_m.
Near this angle a Taylor series

$$\frac{d\Delta k}{d\theta} = \frac{2\omega}{c}\frac{d}{d\theta}\left[n_e^{2\omega}(\theta) - n_o^{\omega}\right] =$$

$$\frac{2\omega}{c}\frac{d}{d\theta}\frac{n_e n_o}{\sqrt{n_o^2 \sin^2\theta + n_e^2 \cos^2\theta}} =$$

$$-\frac{\omega}{c}\frac{n_e n_o}{\left(n_o^2 \sin^2\theta + n_e^2 \cos^2\theta\right)^{3/2}}\left(n_o^2 - n_e^2\right)\sin 2\theta$$

$$= -\frac{\omega}{c}\frac{\left(n_e^{2\omega}(\theta)\right)^3}{n_o^2 n_e^2}\left(n_o^2 - n_e^2\right)\sin 2\theta$$

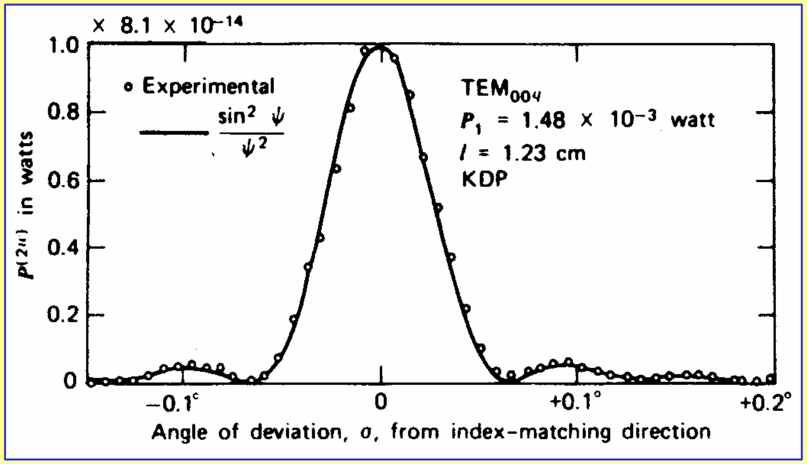
with: $n_e^{2\omega}(\theta) = n_o^{\omega}$

$$\left|\frac{d\Delta k}{d\theta}\right|_{\theta_m} = -\frac{\omega}{c} n_0^3 \left(n_e^{-2} - n_o^{-2}\right)\sin 2\theta_m$$

Spread in k-values relates to spread in $\Delta\theta$

$$\Delta k = \frac{2\beta}{L}\Delta\theta \qquad \text{with} \qquad \beta \propto \sin 2\theta_m$$

$$P^{(2\omega)}(\theta) \propto \frac{\sin^2\left[\frac{\Delta kL}{2}\right]}{\left[\frac{\Delta kL}{2}\right]^2} \propto \frac{\sin^2\left[\beta(\theta - \theta_m)\right]}{\left[\beta(\theta - \theta_m)\right]^2}$$



Opening angle:
1) Interpret as angle – 0.1° – of collimated beam
2) As a divergence (convergence) of a laser beam
3) As a wavelength spread

$$\frac{\Delta k}{k} = -\frac{\Delta\lambda}{\lambda}$$

phase matching by angle tuning

For the example of $LiIO_3$

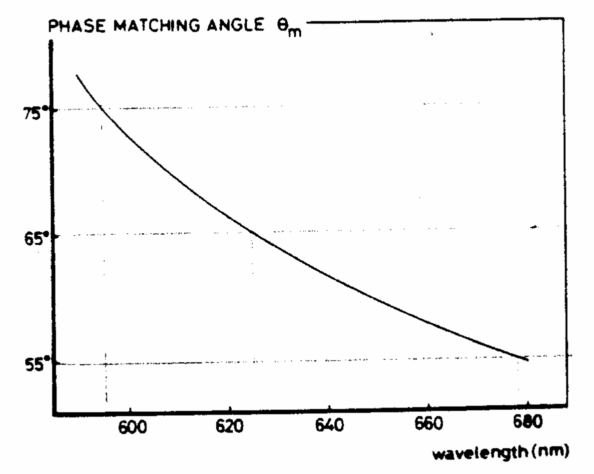
Dispersion:

A	n_o	n_e
4000	1.948	1.780
4360	1.931	1.766
5000	1.908	1.754
5300	1.901	1.750
5780	1.888	1.742
6900	1.875	1.731
8000	1.868	1.724
10600	1.860	1.719

Use dispersion and phase-matching relation:

$$\sin^2\theta_m = \frac{\left(n_o^{\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}$$

Calculate phase matching angle



Practical issue of limitation:

$LiIO_3$ starts absorbing at 295 nm

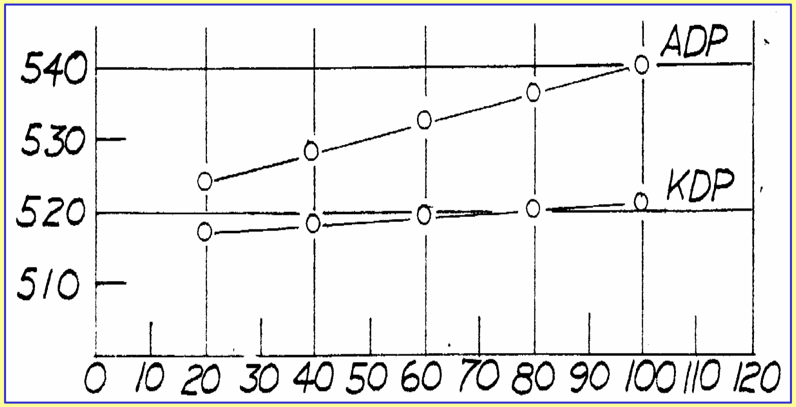
Non-critical phase matching and temperature tuning

Opening angle for wave vectors:

$$\Delta k = \frac{2\beta}{L}\Delta\theta \qquad \beta \propto \sin 2\theta_m$$

Best if $\theta_m = 90^o$

Calculation of Type I for temperatures



→ Temperature tuning

Advantages of 90° phase matching

1) Poynting vector coincides with phase vector so no "walk-off"

2) The first order derivative in Taylor expansion

$$\frac{d\Delta k}{d\theta} = -\frac{\omega}{c}\frac{\left(n_e^{2\omega}(\theta)\right)^3}{n_o^2 n_e^2}\left(n_o^2 - n_e^2\right)\sin 2\theta_m$$

→ 0

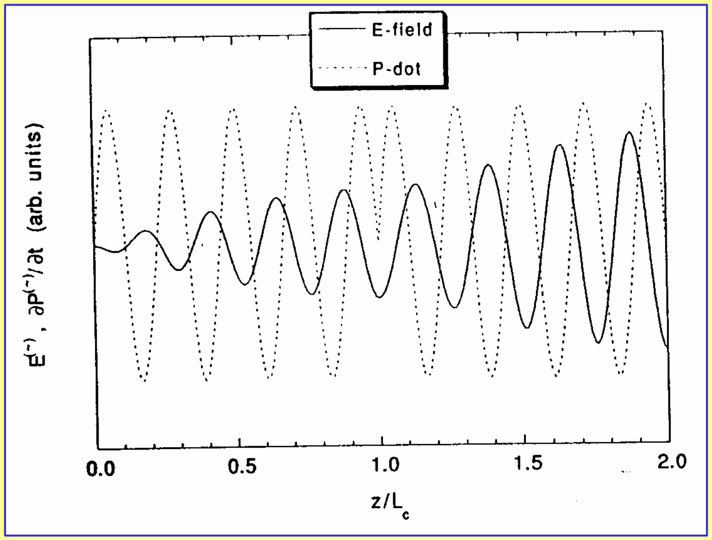
Hence non-critical phase matching:

$$\Delta k \propto (\Delta\theta)^2$$

3) In many cases d is larger at θ_m=90°

Quasi phase matching by periodic poling

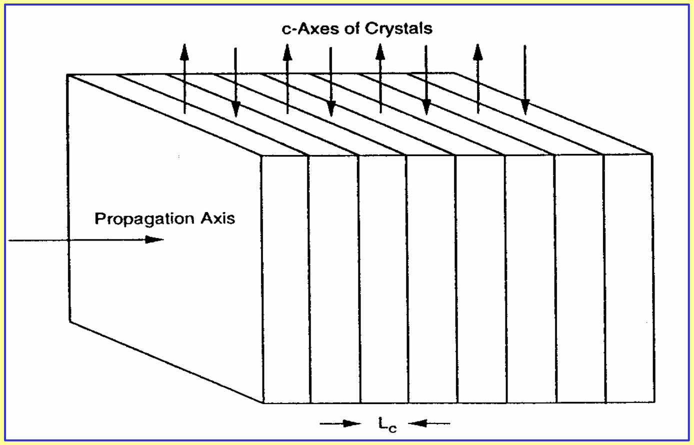
Fundamental and harmonic run out of phase
in conversion processes.
→ Coherence length is limited



Stick segments of material together with
opposite optical axes- crystal modulation.
Change of sign of polarization in each L_c
→ Coherence "runs back"

Periodic poling

Manufacturing of segments by external fields
During/after growth



Quasi phase matching: analysis

Coupled wave equation, with $\Gamma = i\omega E_1^2 / n_2 c$

$$\frac{d}{dz}E_2 = \Gamma d(z)\exp[-i\Delta k' z]$$

Integrate for second harmonic

$$E_2(L) = \Gamma\int_0^L d(z)\exp[-i\Delta k' z]dz$$

d(z) consists of domains with alternating signs

$$E_2 = \frac{i\Gamma d_{eff}}{\Delta k'}\sum_{k=1}^{N} g_k\left[\exp(-i\Delta k' z_k) - \exp(-i\Delta k' z_{k-1})\right]$$

Sign changes (should) occur at: $e^{-i\Delta k_0' z_{k,0}} = (-1)^k$

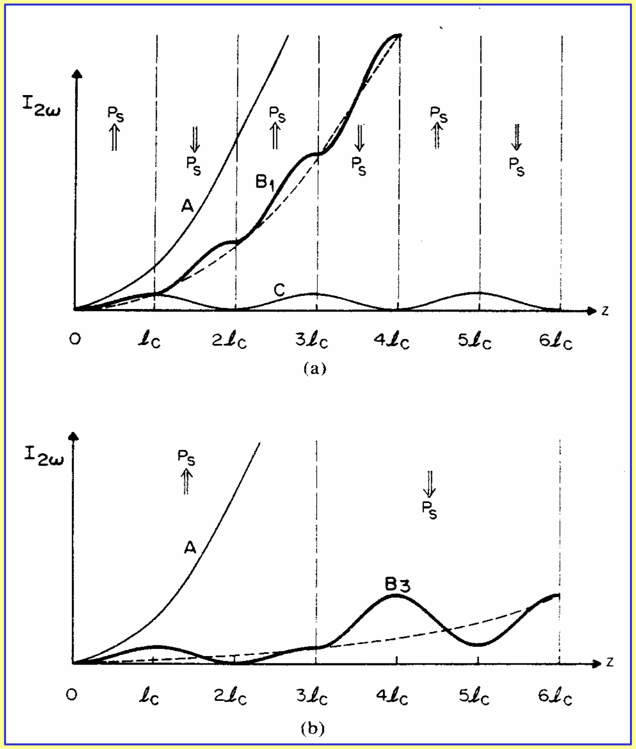
$\Delta k_0'$ wave vector mismatch at design wavelength

For m[th] order QPM: $z_{k,0} = mkl_c$

$$E_{2,ideal} \approx i\Gamma d_{eff}\frac{2}{m\pi}L$$

$E_2(L) = \Gamma d_{eff} L$ for perfect phase matching

Loss factor: $\frac{2}{m\pi}$



A: perfect phase matching
C: phase mismatch for non-poling
B_1: poling at L_c
B3: poling after $3\,L_c$

Pump depletion in SHG

In case of high conversion also revers processes play a role:

$$\omega_1 + \omega_2 \rightarrow \omega_3 \qquad \omega_3 - \omega_1 \rightarrow \omega_2$$

$$\omega_3 - \omega_2 \rightarrow \omega_1$$

Define amplitudes and assume no absorption

$$A_i = \frac{\sqrt{n_i}}{\omega_i} E_i \qquad \kappa = d \frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}$$

Then coupled wave equations turn to Coupled amplitude equations

$$\frac{d}{dz}A_1 = -i\kappa A_3 A_2^* e^{-i\Delta kz}$$

$$\frac{d}{dz}A_2 = +i\kappa A_1 A_3^* e^{i\Delta kz}$$

$$\frac{d}{dz}A_3 = -i\kappa A_1 A_2 e^{i\Delta kz}$$

Assume second harmonic generation $\Delta k=0$;
no field with A_2;
field A_1 is degenerate $\quad A_1 A_2 = \frac{1}{2}A_1^2$

Rewrite: $\quad A_3' = -iA_3$

Then:

$$\frac{d}{dz}A_1 = -\kappa A_3' A_1 \qquad \frac{d}{dz}A_3' = \frac{1}{2}\kappa A_1^2$$

Calculate:

$$\frac{d}{dz}\left[A_1^2 + 2\left(A_3'(z)\right)^2\right] = 2A_1\frac{d}{dz}A_1 + 4A_3'\frac{d}{dz}A_3' = 0$$

So in crystal: $\quad A_1^2 + 2\left(A_3'(z)\right)^2 = \text{constant} = A_1^2(0)$

Consider:

$$I_i = \frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}} n_i |E_i|^2 = \frac{1}{2}\sqrt{\frac{\mu_0}{\varepsilon_0}}\omega_i |A_i|^2 \qquad I_i \propto N_i \hbar\omega_i$$

Hence: #photons(ω_1) + 2#photons(ω_3)=constant

Energy and photon numbers are conserved

Pump depletion in SHG - 2

Solve amplitude equation

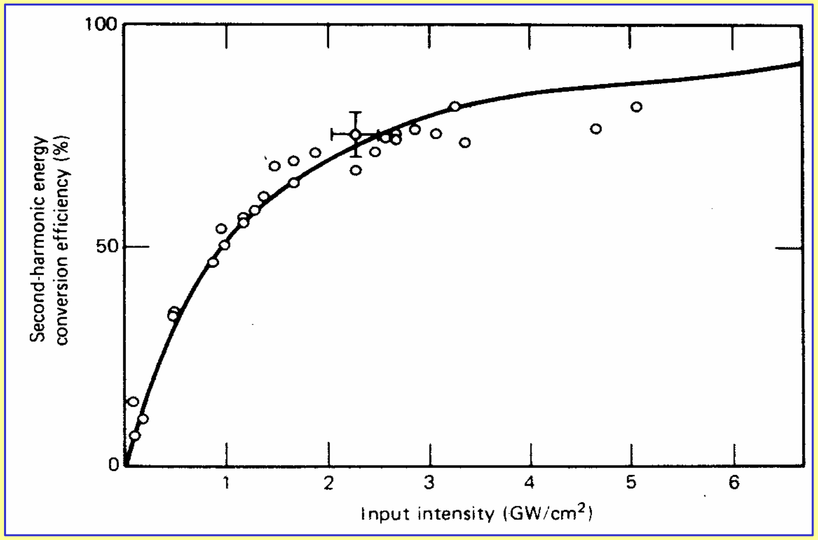
$$\frac{d}{dz}A_3' = -\frac{1}{2}\kappa\left[A_1^2(0) - 2(A_3')^2\right] = 0$$

For:

$$A_1(0)\kappa z \to \infty \qquad \left|A_3'(z)\right|^2 \to \frac{1}{2}\left|A_1(0)\right|^2$$

Solution:

$$A_3'(z) = \frac{A_1(0)}{\sqrt{1/2}}\tanh\left[\frac{A_1(0)\kappa z}{\sqrt{1/2}}\right]$$



Conversion efficiency

$$\eta_{SHG} = \frac{P^{(2\omega)}}{P^{(\omega)}} = \frac{\left|A_3(z)\right|^2}{\frac{1}{2}\left|A_1(0)\right|^2} = \tanh^2\left[\frac{A_1(0)\kappa z}{\sqrt{1/2}}\right]$$