Non-Linear Optics

Chapter 12

Physics 208, Electro-optics

Peter Beyersdorf

Non-Linear Optics

We've seen that an externally applied electric field can alter the index of refraction of a material. At sufficiently high intensities, the electric field associated with a propagating wave can have the same effect. This is the premise behind the field of non-linear optics.

Non-Linear Response

Consider the response (material polarization) of a material to an applied electric field (i.e. a propagating wave)

$$P_i = \epsilon_0 X_{ij} E_j + 2 d_{ijk} E_j E_k + 4 X_{ijkl} E_j E_k E_l + \ldots$$

When the applied electric field is small compared to the internal binding fields of the material, the response is primarily linear

$$P_i \approx \epsilon_0 X_{ij} E_j$$

At higher intensities the higher order terms in the material polarization come into play. Like the electro-optic effect, centro-symmetric crystals do not have a 2^{nd} order nonlinearity (i.e. $d_{ijk}=0$)

Non-Linear d_{ijk} Coefficients

In a non-linear material ($d_{ijk} \neq 0$) the material polarization will have a component at twice the optical frequency. From:

$$P_i(t) = 2d_{ijk}E_j(t)E_k(t)$$

the fields can be written in complex form

$$P_i(t) = 2d_{ijk}\frac{\left(\widetilde{E}_j e^{i\omega_1 t} + \widetilde{E}_j^* e^{-i\omega_1 t}\right)}{2}\frac{\left(\widetilde{E}_k e^{i\omega_2 t} + \widetilde{E}_k^* e^{-i\omega_2 t}\right)}{2}$$

leading to

$$P_i(t) = \frac{1}{2}d_{ijk}\left(\widetilde{E}_j\widetilde{E}_k^* e^{i(\omega_1-\omega_2)t} + \widetilde{E}_j^*\widetilde{E}_k e^{-i(\omega_1-\omega_2)t} + \widetilde{E}_j\widetilde{E}_k e^{i(\omega_1+\omega_2)t} + \widetilde{E}_j^*\widetilde{E}_k^* e^{-i(\omega_1+\omega_2)t}\right)$$

giving

$$\widetilde{P}_i(\omega_1+\omega_2) = d_{ijk}\widetilde{E}_j(\omega_1)\widetilde{E}_k(\omega_2)$$

Non-Linear d_{ijk} Coefficients

The d_{ijk} tensor can be written in contracted form so that

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_zE_y \\ 2E_zE_x \\ 2E_xE_y \end{pmatrix}$$

and has the same form constraints due to crystal symmetry groups as the electrooptic tensor r_{ijk}
More specifically:

$$d_{ijk} = -\frac{\epsilon_{ii}\epsilon_{jj}}{4\epsilon_0} r_{ijk}$$

Wave Equation in a Non-Linear Medium

Starting with the usual Maxwell's equations

$$\vec{\nabla} \times \vec{H} = \vec{J} + \frac{\partial \vec{D}}{\partial t} = \vec{J} + \frac{\partial}{\partial t}\left(\epsilon_0 \vec{E} + \vec{P}\right)$$

$$\vec{\nabla} \times \vec{E} = -\frac{\partial}{\partial t}\left(\mu_0 \vec{H}\right)$$

and writing the first of these equations in terms of the linear and non-linear polarization components

$$\vec{P} = \epsilon_0 \chi_L \vec{E} + \vec{P}_{NL}$$

where

$$\left(\vec{P}_{NL}\right)_i = 2d'_{ijk} E_j E_k$$

we have

$$\vec{\nabla} \times \vec{H} = \sigma \vec{E} + \frac{\partial}{\partial t}\left(\epsilon \vec{E}\right) + \frac{\partial \vec{P}_{NL}}{\partial t}$$

and

$$\vec{\nabla} \times \vec{E} = -\frac{\partial}{\partial t}\left(\mu_0 \vec{H}\right)$$

Wave Equation in a Non-Linear Medium

which can be combined to get

$$\nabla^2 \vec{E} = \mu_0 \sigma \frac{\partial \vec{E}}{\partial t} + \mu_0 \epsilon \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2}{\partial t^2} \vec{P}_{NL}$$

considering the one-dimensional case of propagation in the z-direction with fields of the form

$$E_i(\omega_1, z, t) = \frac{1}{2}\left[E_{1i}(z)\, e^{i(\omega_1 t - k_1 z)} + c.c.\right]$$

$$E_k(\omega_2, z, t) = \frac{1}{2}\left[E_{2k}(z)\, e^{i(\omega_2 t - k_2 z)} + c.c.\right]$$

$$E_j(\omega_3, z, t) = \frac{1}{2}\left[E_{3j}(z)\, e^{i(\omega_3 t - k_3 z)} + c.c.\right]$$

c.c. stands for "complex conjugate"

where i,j, and k can be either the x or y directions, and $\omega_3 = \omega_1 + \omega_2$.

Wave Equation in a Non-Linear Medium

computing the derivative with the plane-wave solutions gives at frequency ω_l

$$\nabla^2 E_i(\omega_l, z, t) = -\frac{1}{2}\left[k_l^2 E_{li}(z) + 2ik_l \frac{dE_{li}(z)}{dz}\right] e^{i(\omega_l t - k_l z)} + c.c.$$

for $l=1,2,3$ where $E_{li}(z)=E_i(\omega_l)$ and we have neglected terms of the form d^2E/dz^2 because we assume the field amplitudes are varying slowly compared to the optical period, i.e.

$$\frac{dE_{li}}{dz}k_l \gg \frac{d^2 E_{li}}{dz^2}$$

Wave Equation in a Non-Linear Medium

The wave equation can thus be written as

$$\left[\frac{k_1^2}{2}E_{1i} + ik_1\frac{dE_{1i}}{dz}\right]e^{i(\omega_1 t - k_1 z)} + c.c = \left[\left(-i\omega_1\mu_0\sigma + \omega_1^2\mu_0\epsilon\right)\frac{1}{2}E_{1i}e^{i(\omega_1 t - k_1 z)} + c.c\right] - \mu_0\frac{\partial^2}{\partial t^2}\left[P_{NL}(z,t)\right]_i$$

which can be written using $\left(\vec{P}_{NL}\right)_i = 2d'_{ijk}E_jE_k$,

$k_1^2=\omega_1^2\mu_0\epsilon$ and $\omega_1=\omega_3-\omega_2$ as

$$ik_1\frac{dE_{1i}}{dz}e^{-ik_1 z} = -\frac{i\omega_1\sigma\mu_0}{2}E_{1i}e^{-ik_1 z} + \mu_0\omega_1^2 d'_{ijk}E_{3j}E_{2k}^*e^{-i(k_3-k2)z}$$

giving components of $E(\omega_1)$, $E(\omega_2)$ and $E(\omega_3)$ of

$$\frac{dE_{1i}}{dz} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_0}{\epsilon_1}}E_{1i} - i\omega_1\sqrt{\frac{\mu_0}{\epsilon_1}}d'_{ijk}E_{3j}E_{2k}^*e^{-i(k_3-k_2-k_1)z}$$

$$\frac{dE_{2k}^*}{dz} = -\frac{\sigma_2}{2}\sqrt{\frac{\mu_0}{\epsilon_2}}E_{2k}^* + i\omega_2\sqrt{\frac{\mu_0}{\epsilon_2}}d'_{kij}E_{1i}E_{3j}^*e^{-i(k_1-k_3+k_2)z}$$

$$\frac{dE_{3j}}{dz} = -\frac{\sigma_3}{2}\sqrt{\frac{\mu_0}{\epsilon_3}}E_{3j} - i\omega_3\sqrt{\frac{\mu_0}{\epsilon_3}}d'_{jik}E_{1i}E_{2k}e^{-i(k_1+k_2-k_3)z}$$

Wave Equation in a Non-Linear Medium

$$\frac{dE_{1i}}{dz} = -\frac{\sigma_1}{2}\sqrt{\frac{\mu_0}{\epsilon_1}}E_{1i} - i\omega_1\sqrt{\frac{\mu_0}{\epsilon_1}}d'_{ijk}E_{3j}E^*_{2k}e^{-i(k_3-k_2-k_1)z}$$

Absorption, i.e. for solution $E=E_0e^{-\alpha z/2}$ we have

$$\alpha = \sigma\sqrt{\frac{\mu_0}{\epsilon}}$$

Conversion to/from other frequencies. Whether conversion is from or to this field depends on the phase of this field relative to E_2 and E_3. If Δkz , i.e. $(k_3-k_2-k_1)z$ changes by π the conversion switches signs.

Second Harmonic Generation

Consider case of $\omega_1=\omega_2=\omega_3/2$ and define $\Delta k \equiv k_3-(k_1+k_2)$ where $\Delta k=0$ in the absence of dispersion (i.e. $dn/d\lambda=0$).

Assume the "pump" wave at ω_1 is not depleted ($dE_1/dz=0$) and the material is transparent at frequency ω_3 (i.e. $\sigma=0$) and solve for dE_3/dz to get

$$\frac{dE_{3j}}{dz} = -\frac{\sigma_3}{2}\sqrt{\frac{\mu_0}{\epsilon_3}}E_{3j} - i\omega_3\sqrt{\frac{\mu_0}{\epsilon_3}}d'_{jik}E_{1i}E_{2k}e^{-i(k_1+k_2-k_3)z}$$

$$\downarrow$$

$$\frac{dE_{3j}}{dz} = -i\omega\sqrt{\frac{\mu_0}{\epsilon}}d'_{jik}E_{1i}E_{1k}e^{i\Delta kz}$$

Second Harmonic Generation

$$\frac{dE_{3j}}{dz} = -\omega\sqrt{\frac{\mu_0}{\epsilon}} d'_{jik} E_{1i} E_{1k} e^{i\Delta kz}$$

giving at the end of a crystal of length L

$$E_{3j}(L) = -i\omega\sqrt{\frac{\mu_0}{\epsilon}} d'_{jik} E_{1i} E_{1k} \frac{e^{i\Delta kL} - 1}{i\Delta k}$$

or

$$E_{3j}(L) = -2ie^{i\Delta kL/2}\omega\sqrt{\frac{\mu_0}{\epsilon}} d'_{jik} E_{1i} E_{1k} \frac{\sin(\Delta kL/2)}{\Delta k}$$

or for the intensity of the sum frequency component

$$\langle I(\omega_3)\rangle = \frac{1}{2}\sqrt{\frac{\epsilon}{\mu_0}} E^*_{3j}(L) E_{3j}(L) = 2\sqrt{\frac{\mu_0}{\epsilon}} \omega^2 \left(d'_{ijk}\right)^2 E^2_{1i} E^2_{1k} L^2 \frac{\sin^2 \frac{1}{2}\Delta kL}{\left(\frac{1}{2}\Delta kL\right)^2}$$

$$\langle I_j(2\omega)\rangle = 8\left(\frac{\mu_0}{\epsilon}\right)^{\frac{3}{2}} \frac{\omega^2 d'^2_{ijk} L^2}{n^3} \langle I_i(\omega_1)\rangle \langle I_k(\omega_1)\rangle \frac{\sin^2 \frac{1}{2}\Delta kL}{\left(\frac{1}{2}\Delta kL\right)^2}$$

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Phase Matching

From the expression of the intensity of the sum frequency wave

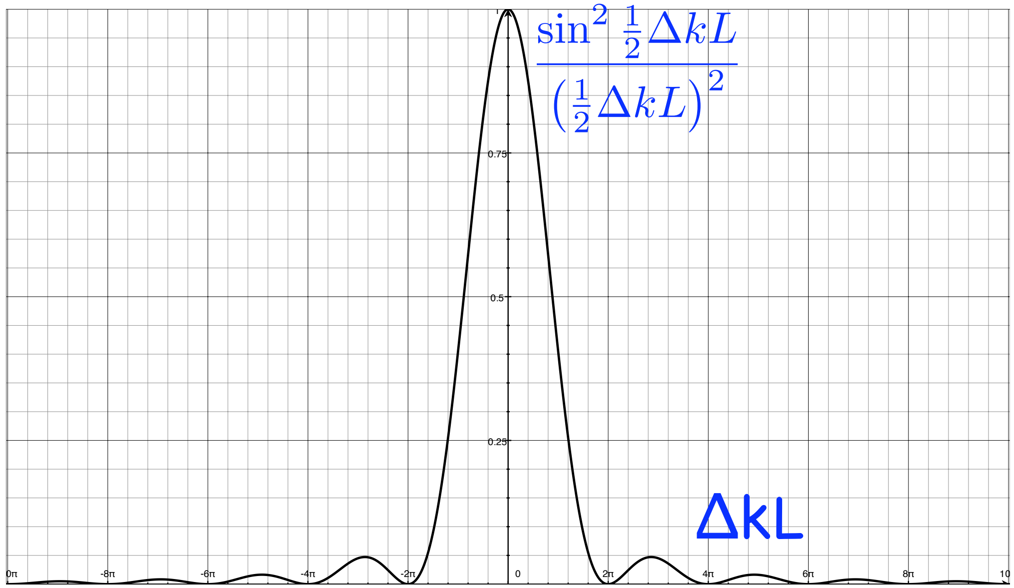
$$\langle I_j(2\omega)\rangle = 8\left(\frac{\mu_0}{\epsilon}\right)^{\frac{3}{2}} \frac{\omega^2 d'^2_{ijk} L^2}{n^3} \langle I_i(\omega_1)\rangle \langle I_k(\omega_1)\rangle \frac{\sin^2 \frac{1}{2}\Delta k L}{\left(\frac{1}{2}\Delta k L\right)^2}$$

we can see that a long crystal benefits frequency conversion efficiency, but the length is limited by the $\mathrm{sinc}^2(\Delta kL/2)$ factor

$$\frac{\sin^2 \frac{1}{2}\Delta k L}{\left(\frac{1}{2}\Delta k L\right)^2}$$

unless $\Delta k=0$.

$\text{Sinc}^2 \Delta kL/2$ term



Since $$< I(\omega_3) > \propto L^2 \frac{\sin^2 \frac{1}{2}\Delta kL}{\left(\frac{1}{2}\Delta kL\right)^2}$$

$$< I(\omega_3) >_{max} \propto \frac{1}{(\Delta k)^2}$$ at $$L_{max} = \frac{2\pi}{\Delta k}$$

Thus Δk should be minimized for maximum conversion efficiency

Phase Matching

Physical interpretation of the $\mathrm{Sinc}^2 \Delta kL/2$ term is that if $\Delta k \neq 0$ the pump wave at ω and the second harmonic wave at 2ω will drift out of phase over a distance $l_c/2 = \pi/\Delta k$ causing the conversion to cancel the second harmonic wave rather than augment it.

In normally dispersive materials ($dn/d\omega > 0$) Δk is not zero without special efforts to arrange it to be so.

In typical materials, $l_c \approx 100\ \mu m$

Angle Phase Matching

In crystals the index of refraction seen by one polarization can be tuned by adjusting the angle, for example in a uniaxial crystal

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2\theta}{n_0^2} + \frac{\sin^2\theta}{n_e^2}$$

allowing the value of Δk to be adjusted if the waves at ω_1, ω_2 and ω_3 do not all have the same polarization state. Depending on the relative polarization states we have different "types" of phase matching

Scheme	Polarizations		
$E(\omega_1)$	$E(\omega_1)$	$E(\omega_2)$	$E(\omega_3)$
I	o	o	e
II (IIA)	e	o	e
III (IIB)	o	e	e
IV	e	e	e
V	o	o	o
VI (IIB)	e	o	o
VII (IIA)	o	e	o
VIII (I)	e	e	o

For $\omega_1 \leq \omega_2 < \omega_3$

Angle Phase Matching

Most common non-linear crystal are negative uniaxial and normally dispersive ($dn/d\omega>0$) therefore requiring type I, II or III phase matching.

What types of phase matching would be useful in a positive uniaxial crystal with normal dispersion?

$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2\theta}{n_0^2} + \frac{\sin^2\theta}{n_e^2}$$

Scheme	Polarizations		
$E(\omega_1)$	$E(\omega_1)$	$E(\omega_2)$	$E(\omega_3)$
I	o	o	e
II (IIA)	e	o	e
III (IIB)	o	e	e
IV	e	e	e
V	o	o	o
VI (IIB)	e	o	o
VII (IIA)	o	e	o
VIII (I)	e	e	o

For $\omega_1 \leq \omega_2 < \omega_3$

Type I and II phase matching

The most common angle phase matching is type I and II:

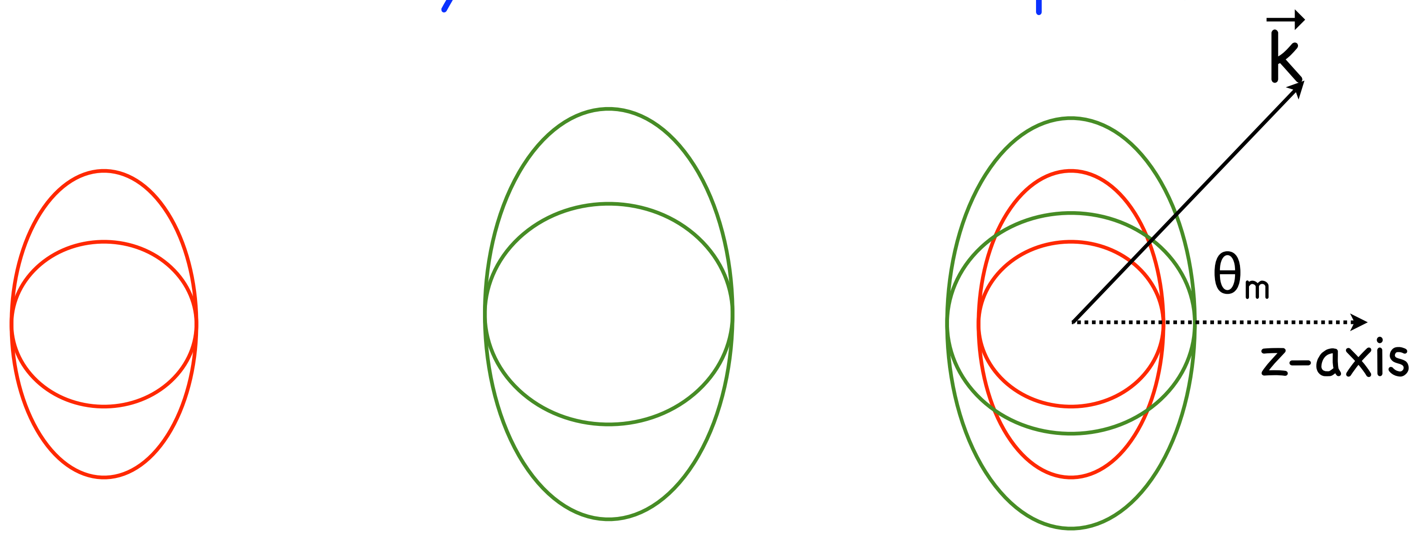
Type I phase-matching has the sum frequency wave $E(\omega_3)$ with a different polarization than the other two waves

Type II phase-matching has one of either $E(\omega_1)$ or $E(\omega_2)$ with a different polarization state than the other two waves.

Type I Phase Matching

Technically this is type VIII, but it is commonly referred to as type 1 since the sum frequency wave is orthogonally polarized to both pump waves

Normal shells can be used as a geometric tool to determine proper phase matching angle. In a positive uniaxial crystal with normal dispersion:



Normal shells for $\omega_1=\omega_2$

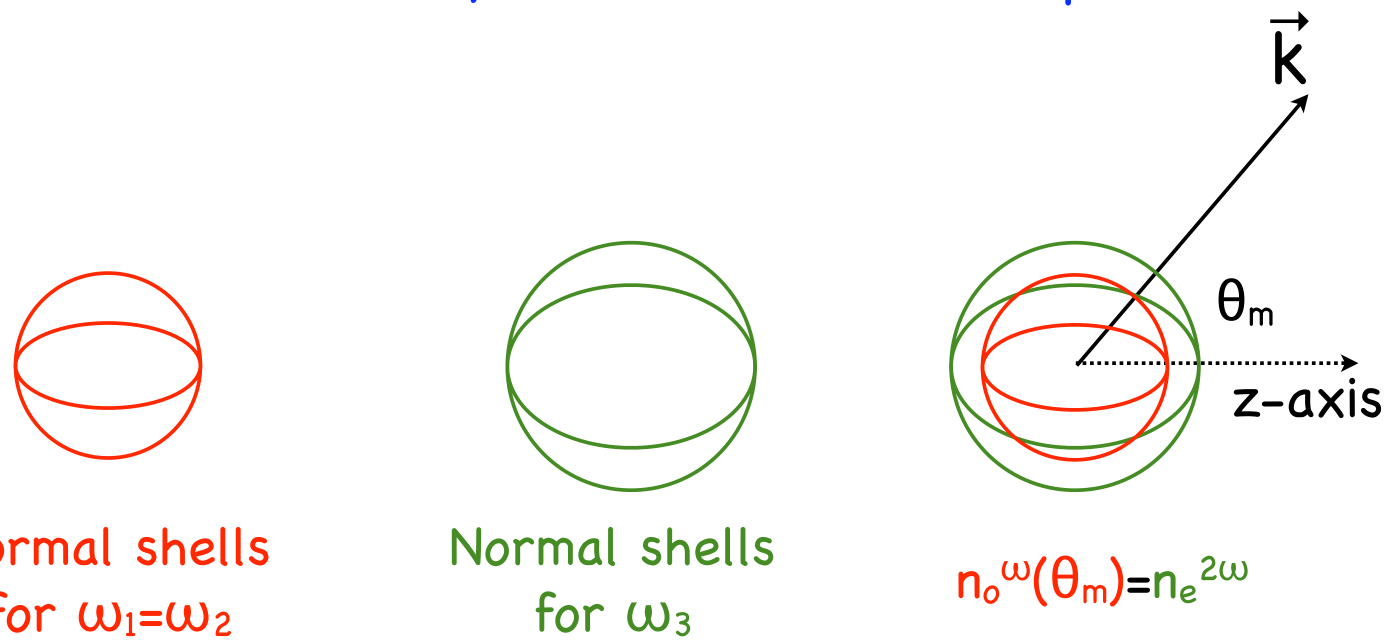
Normal shells for $\omega_3=2\omega_1$

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

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

Type I Phase Matching

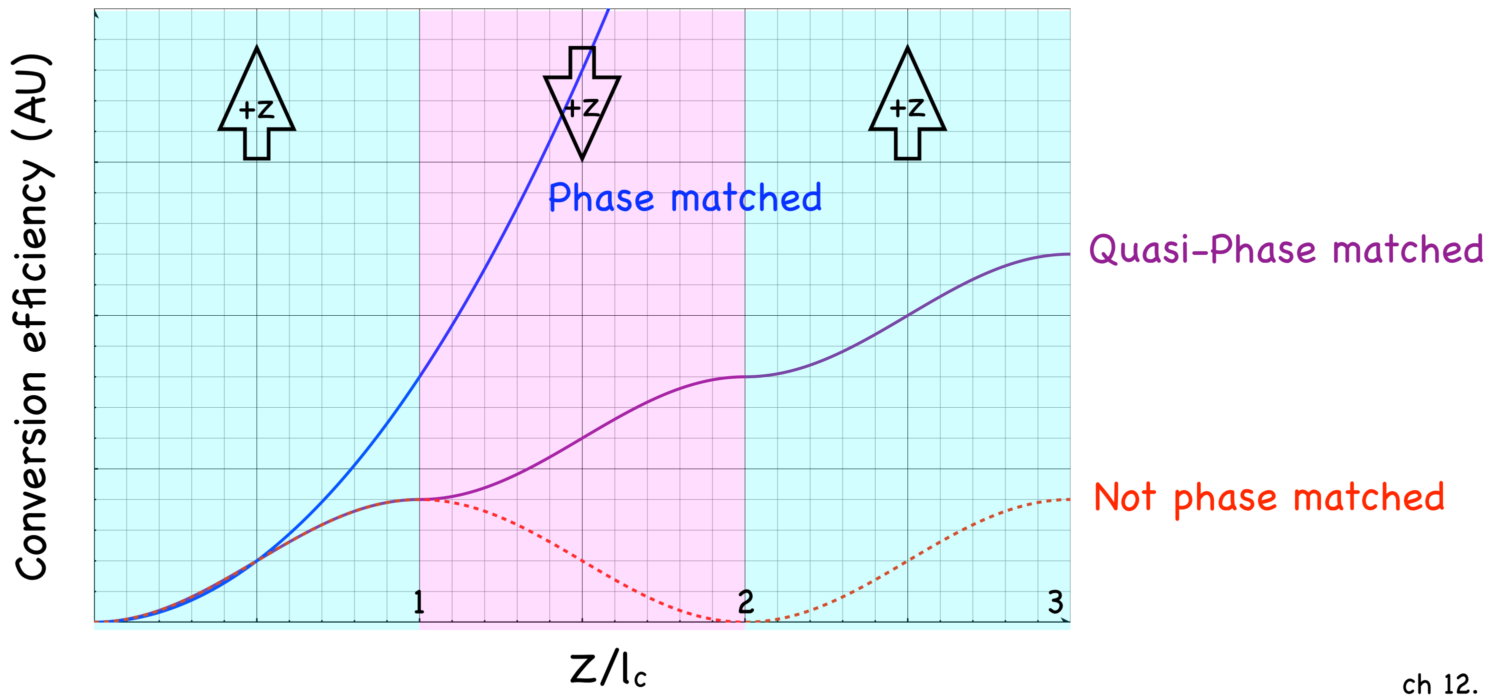
Normal shells can be used as a geometric tool to determine proper phase matching angle. In a negative uniaxial crystal with normal dispersion:



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

Quasi Phase Matching

If the crystal domain polarity can be engineered to flip signs every l_c the polarity of the sum frequency wave being generated can flip every time the sum frequency wave drifts π out-of-phase with the driving waves.

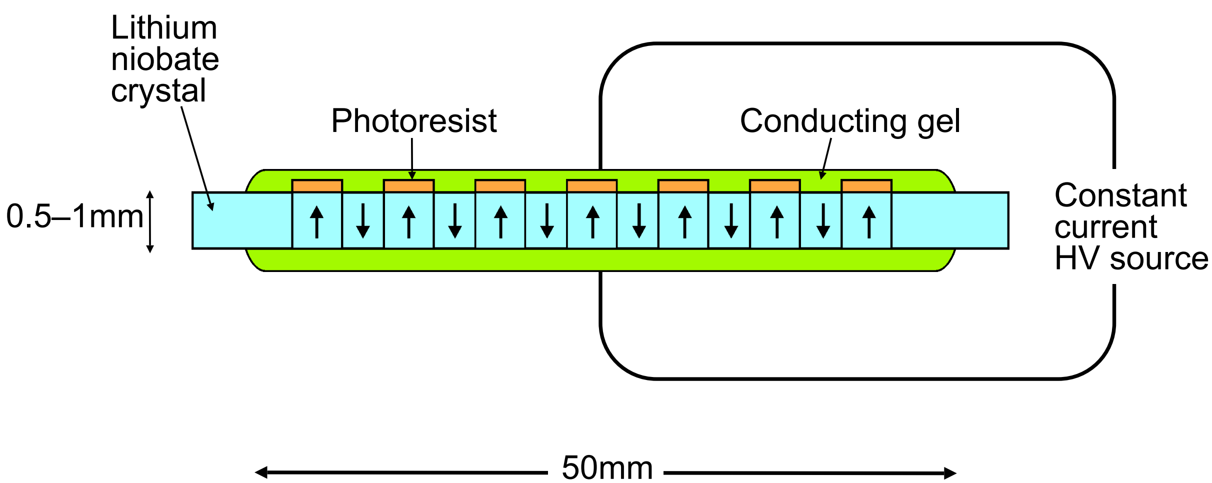


Engineered QPM materials

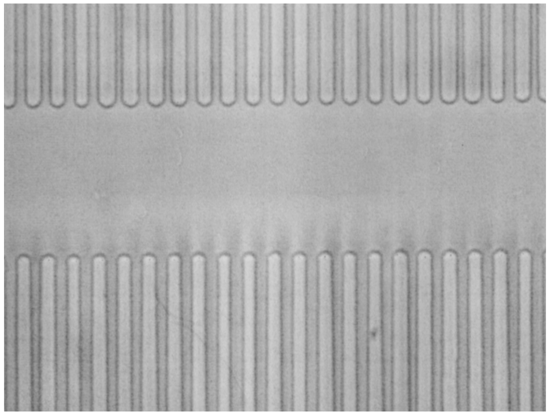
Periodically Poled Lithium Niobate (PPLN)

Periodically Poled Lithium Tantalate (PPLT)

Orientation Patterned Gallium Arsenide (OpGaAs)



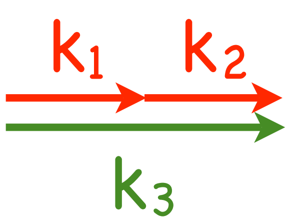
Poling method for PPLN



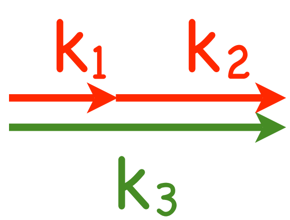
Phase contrast image of PPLN

Momentum Conservation

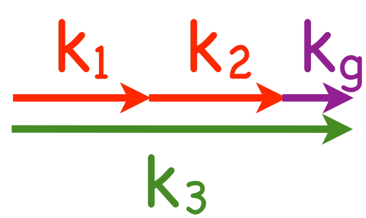
Phase matching is a form of momentum conservation ($k_1+k_2=k_3$) that must be satisfied along with energy conservation ($\omega_1+\omega_2=\omega_3$) in non-linear processes. If we don't require the beams be collinear then phase matching can be achieved by crossing beams



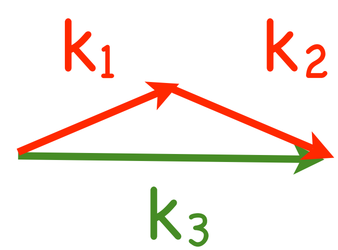
collinear type I phase matching



collinear type II phase matching



Quasi phase matching



non-collinear type IV phase matching

Example of SHG in KDP

Determine the type of phase matching to use for second harmonic generation in KDP with a fundamental wavelength of λ=694.3 nm, and determine the phase matching angle using

$n_e(\omega)=1.466$ $n_e(2\omega)=1.487$

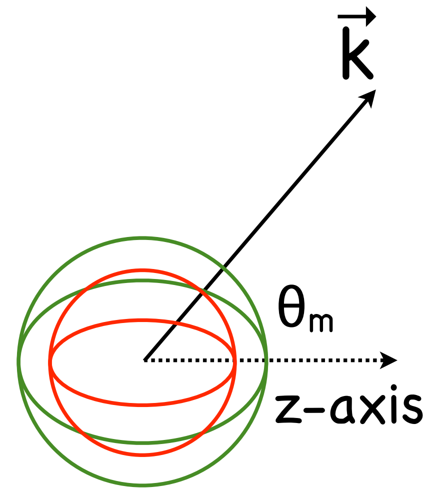
$n_o(\omega)=1.506$ $n_o(2\omega)=1.534$

Example of SHG in KDP

With $n_e(\omega)=1.466$ $n_e(2\omega)=1.487$

$n_o(\omega)=1.506$ $n_o(2\omega)=1.534$

this is a negative uniaxial crystal with normal dispersion. We can use type I phase matching and require



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

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

References

- Yariv & Yeh "Optical Waves in Crystals" chapter 12