Lecture 8

Phase matching. Phase-matching directions in uniaxial birefringent crystals, 90º phase-matching, non-collinear phase-matching.

Phase matching

The rediced coupling equations (5.4) of Lecture 5 ignore the position dependence of the electrical fields, specifically their phases.
(only good for short interacting lengths).

$$\frac{\partial E(z)}{\partial z} = -\frac{i\omega}{2nc\varepsilon_0} P_{NL}$$

The electrical fields are traveling waves described by: $E(z,t) = \frac{1}{2}(Ee^{i(\omega t - kz)} + c.c.)$

For sum-frequency generation (SFG):

The second-order polarization at angular frequency $\omega_3 = \omega_1 + \omega_2$ travels as

$$P^{(2)}(z,t) \sim E_1 e^{i(\omega_1 t - k_1 z)} E_2 e^{i(\omega_2 t - k_2 z)} \sim E_1 E_2 e^{i[(\omega_1 +)t - (k_1 + k_2)z]} + c.c.$$

The electrical field at angular frequency $\omega_3 = \omega_1 + \omega_2$ travels as

$$E_3(z,t) = e^{i(\omega_3 t - k_3 z)}$$

$$phase\ velocity = \frac{\omega_3}{k_3} \neq \frac{\omega_1 + \omega_2}{k_1 + k_2}$$

Constructive interference, and therefore a high-intensity ω_3 field, will occur only if

$$k_3 = k_1 + k_2$$

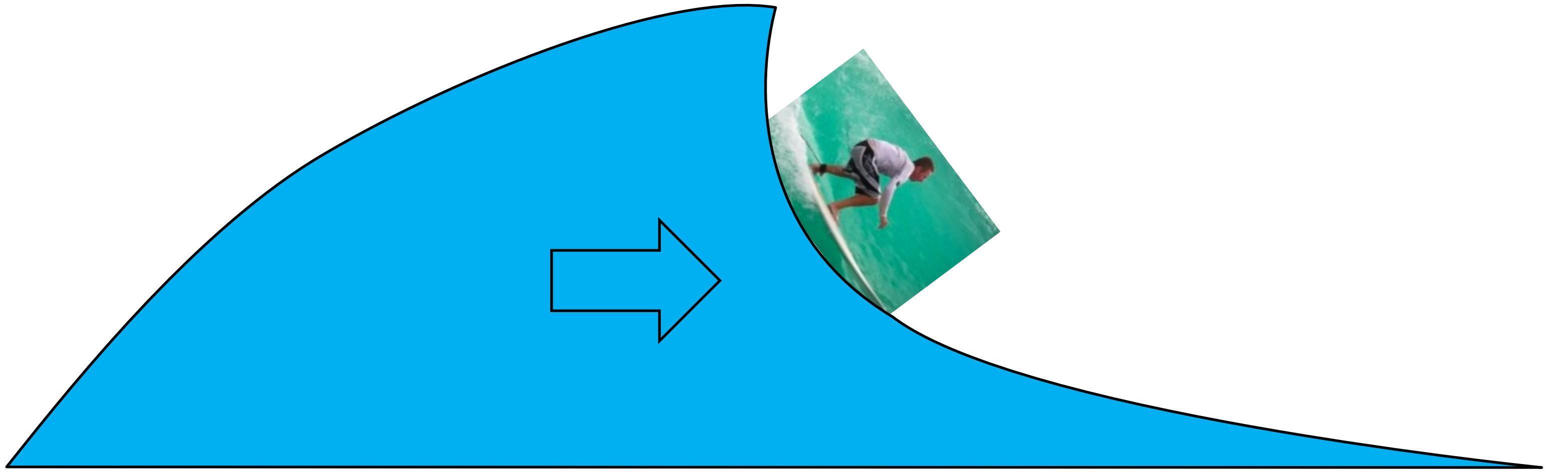
Strictly speaking this is a vector equation:

$$\boldsymbol{k_3} = \boldsymbol{k_1} + \boldsymbol{k_2}$$

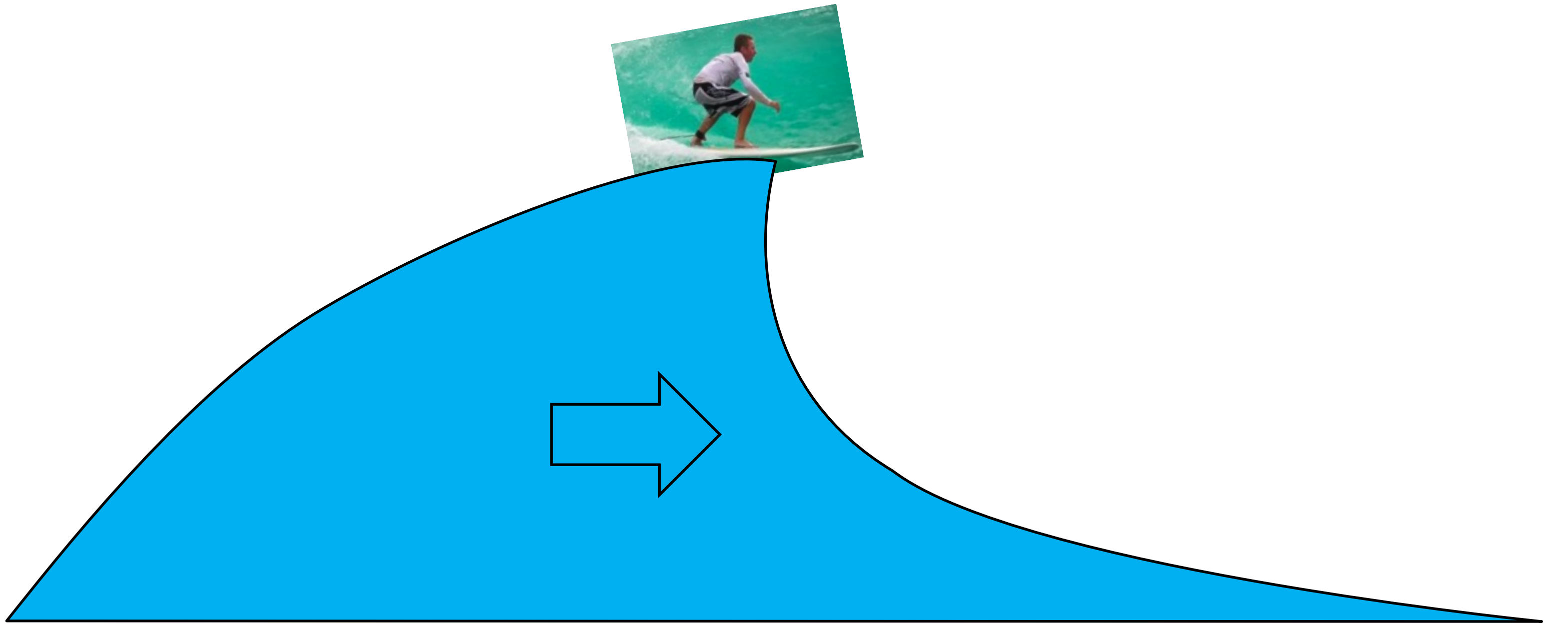
In more general terms (since $\boldsymbol{p} = \hbar \boldsymbol{k}$), it is momentum conservation:

$$\boldsymbol{p_3} = \boldsymbol{p_1} + \boldsymbol{p_2}$$

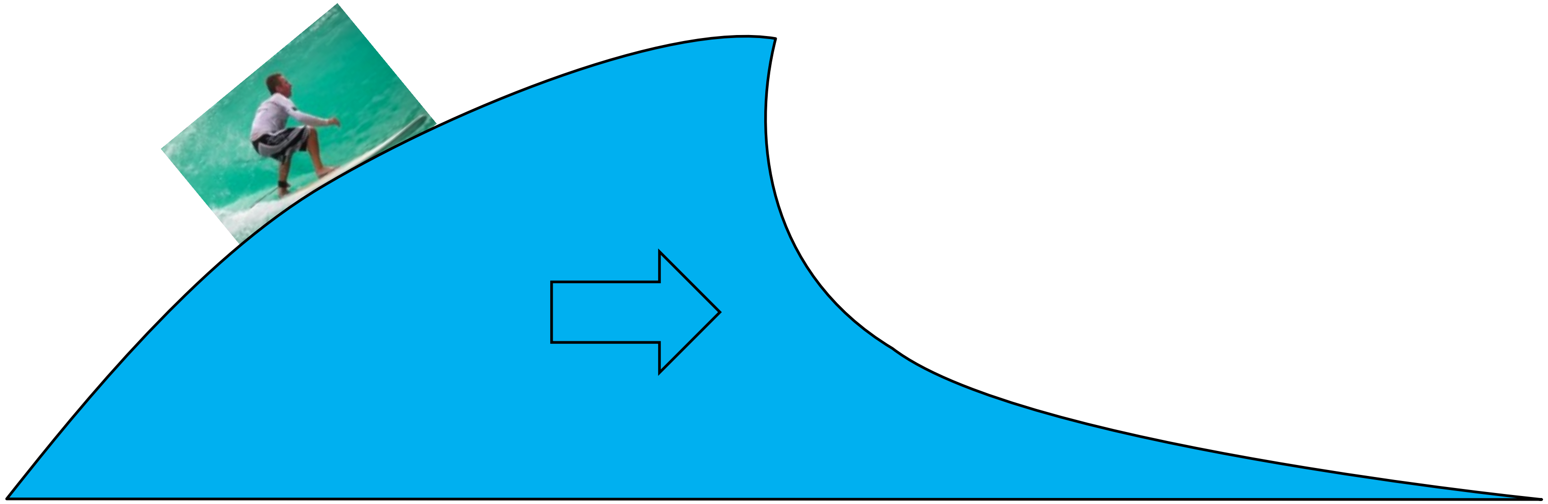
Phase matching



Phase matching



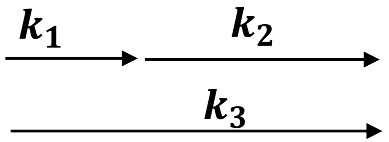
Phase matching



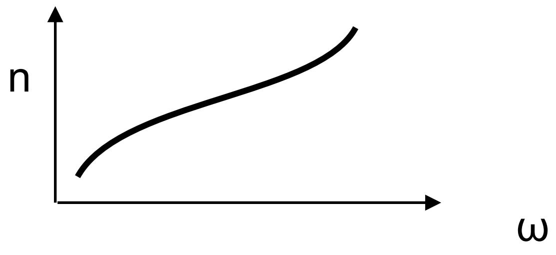
Phase matching

Constructive interference, and therefore a high-intensity ω_3 field, will occur only if

$$k_3 = k_1 + k_2$$



Transparent materials have normal dispersion: the index of refraction increases monotonically as a function of frequency (decreases with wavelength).



This makes phase matching impossible in most frequency-mixing processes.

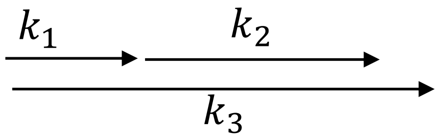
$$k = \frac{\omega n}{c}$$

$$\omega_3 = \omega_1 + \omega_2$$

$$k_3 = k_1 + k_2$$

$$\omega_3 n_3 = \omega_1 n_1 + \omega_2 n_2$$

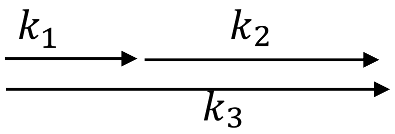
$n_3 > n_1, n_2$



However, **birefringent** materials avoid this problem by having two indices of refraction at once.

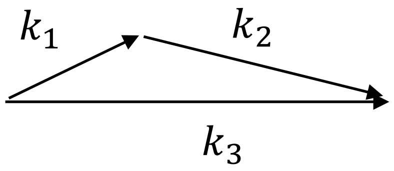
Phase matching

$$k_3 = k_1 + k_2$$



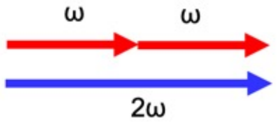
vector equation:

$$\boldsymbol{k_3} = \boldsymbol{k_1} + \boldsymbol{k_2}$$



non-collinear

Second harmonic generation (SHG), collinear case



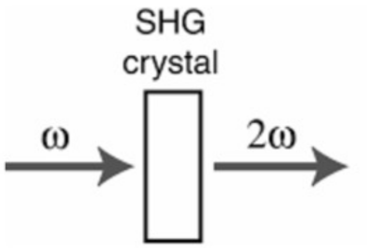
$$2\omega = \omega + \omega$$

$$k_{2\omega} = k_\omega + k_\omega$$

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

$$2\omega n_{2\omega} = \omega n_\omega + \omega n_\omega = 2\omega n_\omega$$

$$n_{2\omega} = n_\omega$$



Anisotropic linear media

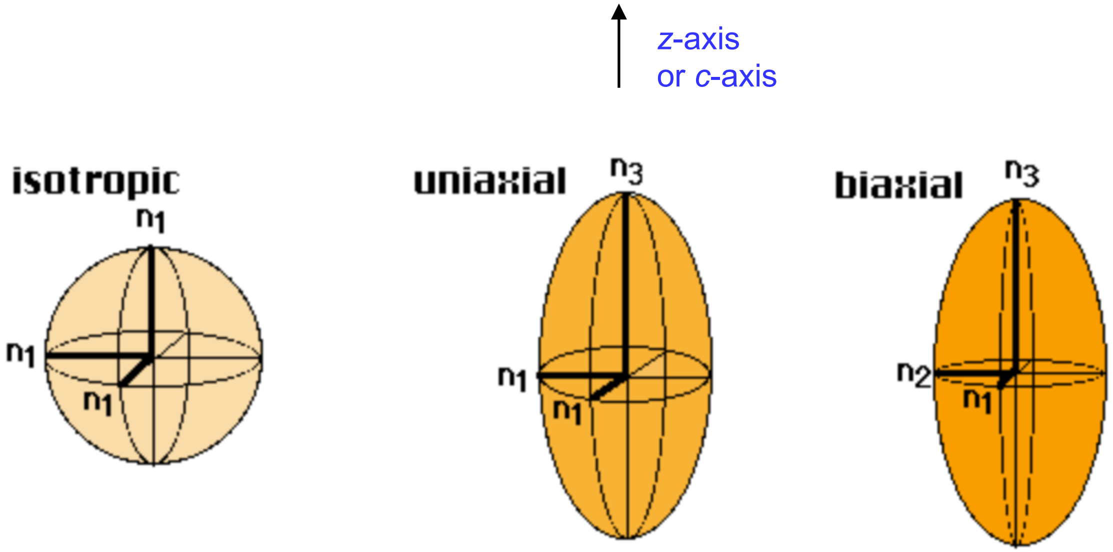
$$\varepsilon = \quad \varepsilon_0 \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix}$$

Waves with different polarizations will see different refractive indices and travel at different speeds.

This phenomenon is known as **birefringence** and occurs in some crystals such as calcite and quartz.

If $n_x = n_y \neq n_z$, the crystal is known as **uniaxial**.

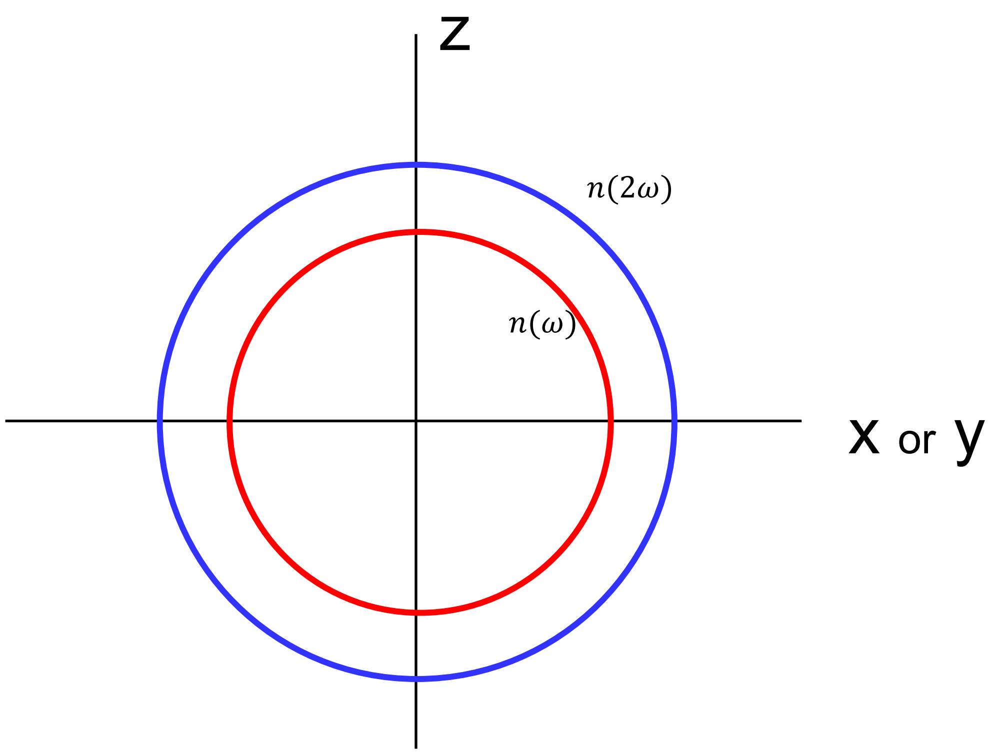
If $n_x \neq n_y \neq n_z$ the crystal is called **biaxial**.



Uzbek melon

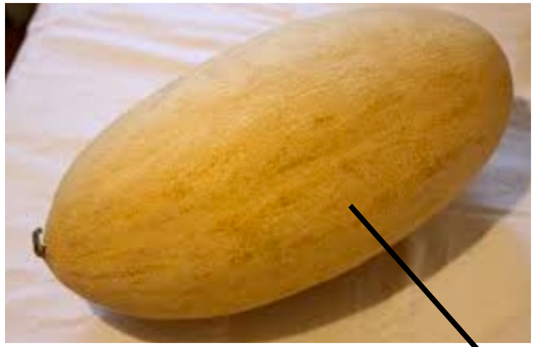
Phase matching

Isotropic crystal

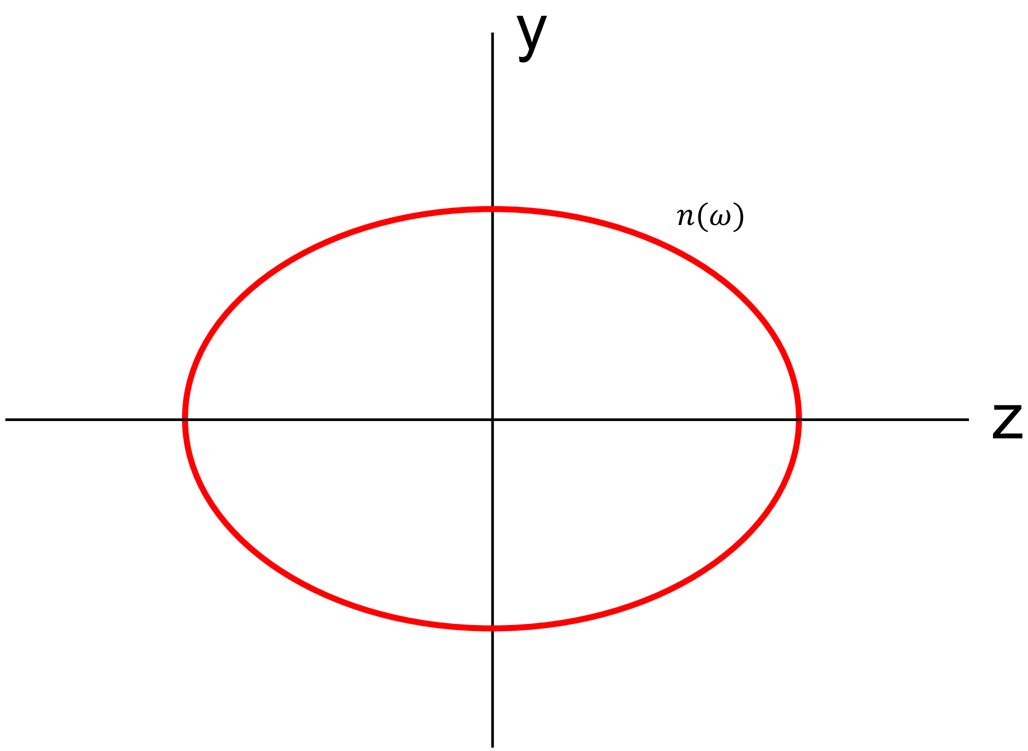


Phase matching

Un-isotropic crystal



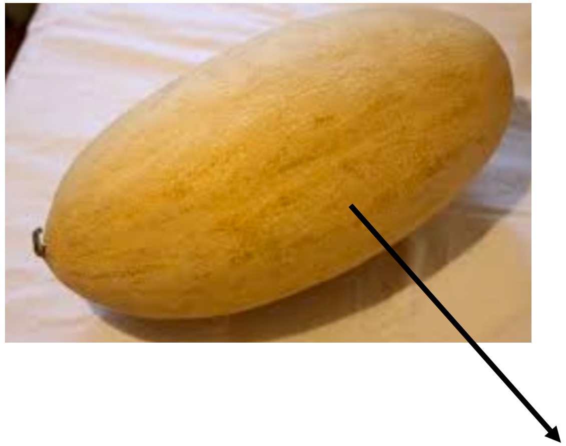
k-vector
along x



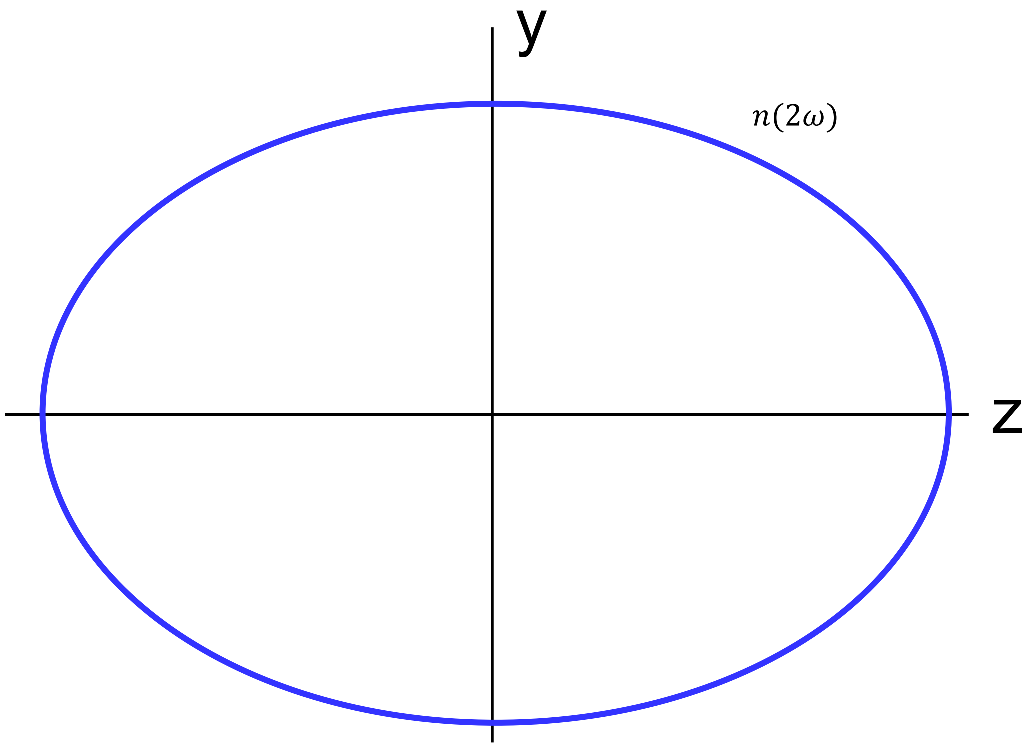
Phase matching

Un-isotropic crystal

$n_e > n_o$



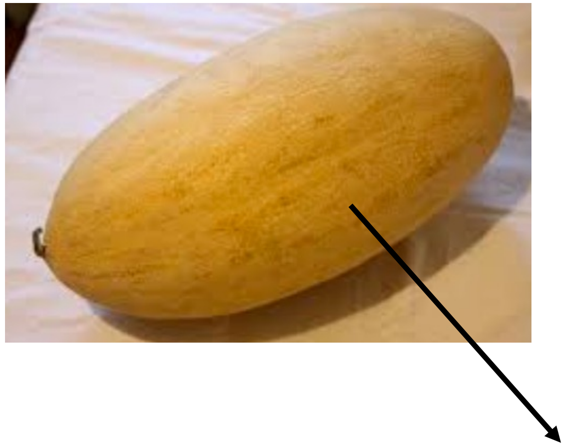
k-vector along x



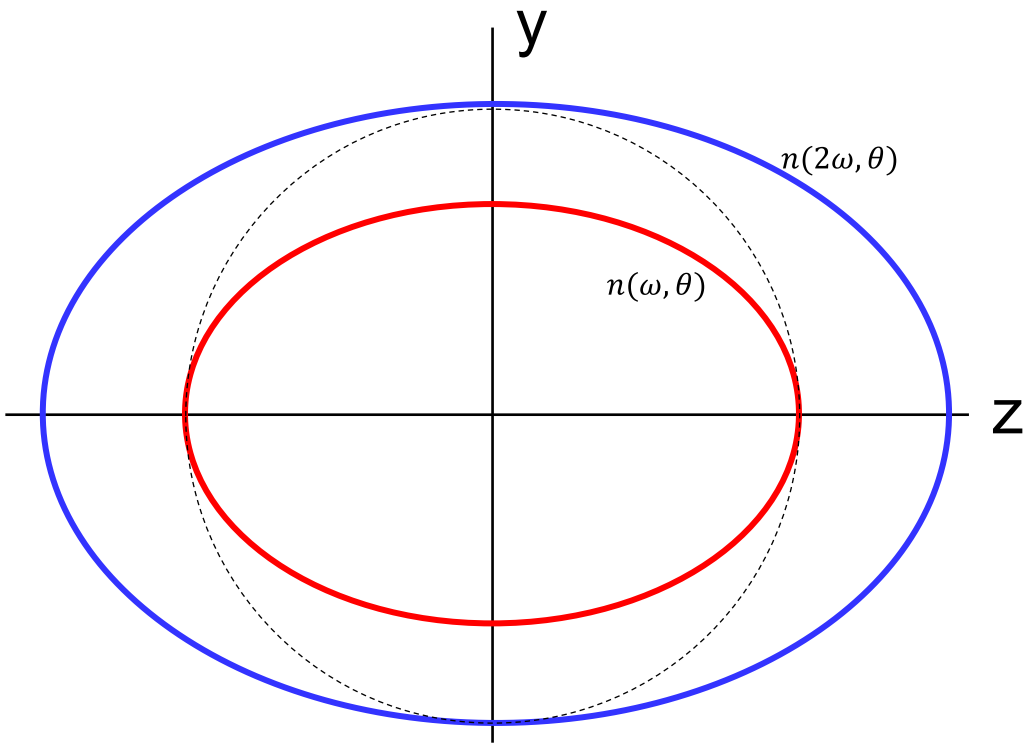
Phase matching

Un-isotropic crystal

$n_e > n_o$



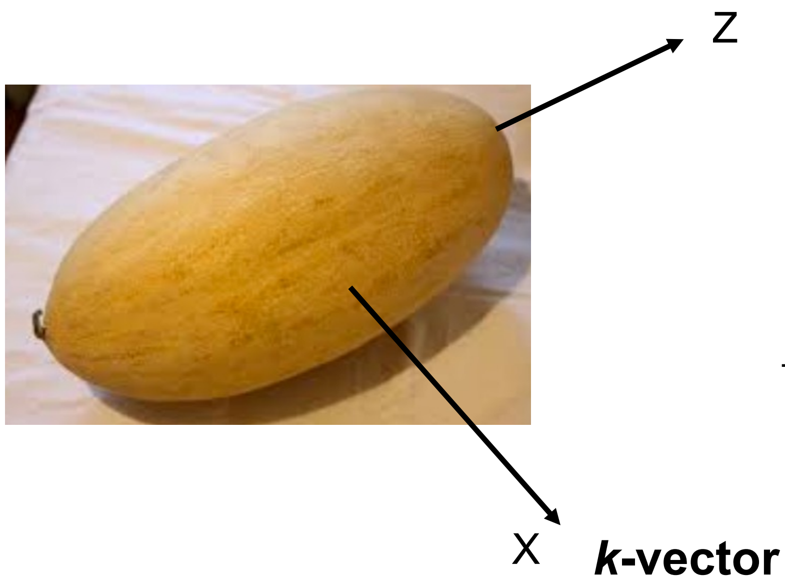
k-vector along x

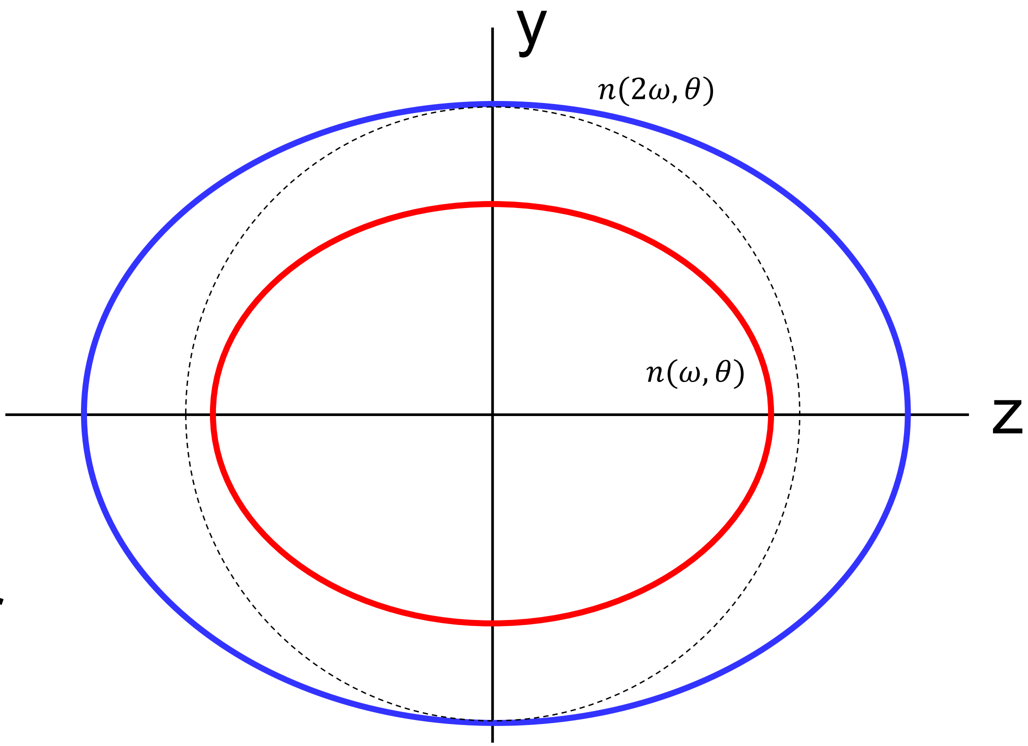


Phase matching

Un-isotropic crystal

$$n_e > n_o$$



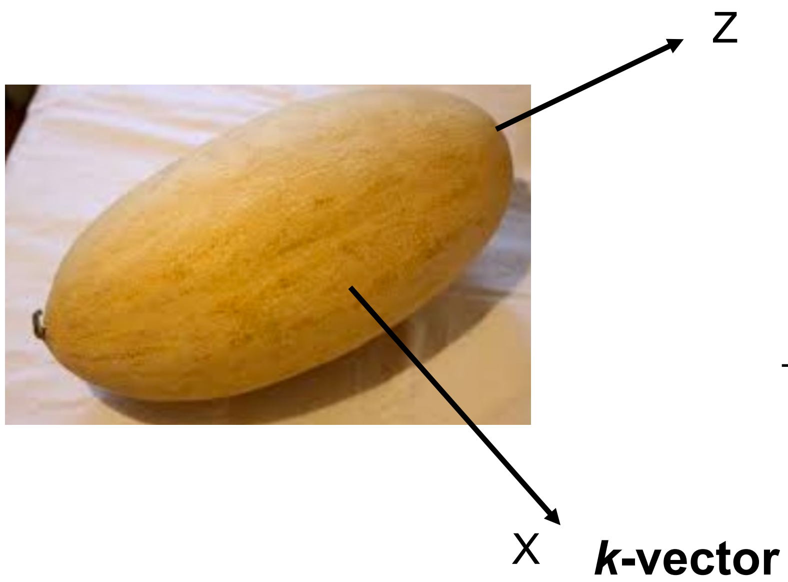


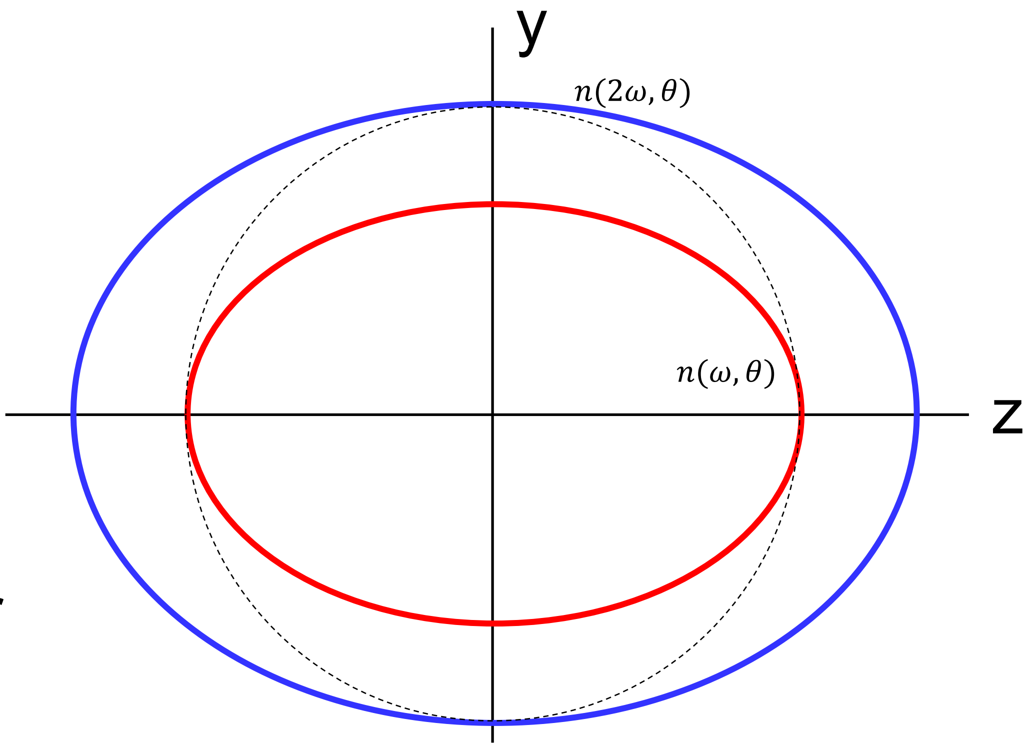
Here, there is **not enough** birefringence to compensate chromatic dispersion

Phase matching

Un-isotropic crystal

$$n_e > n_o$$



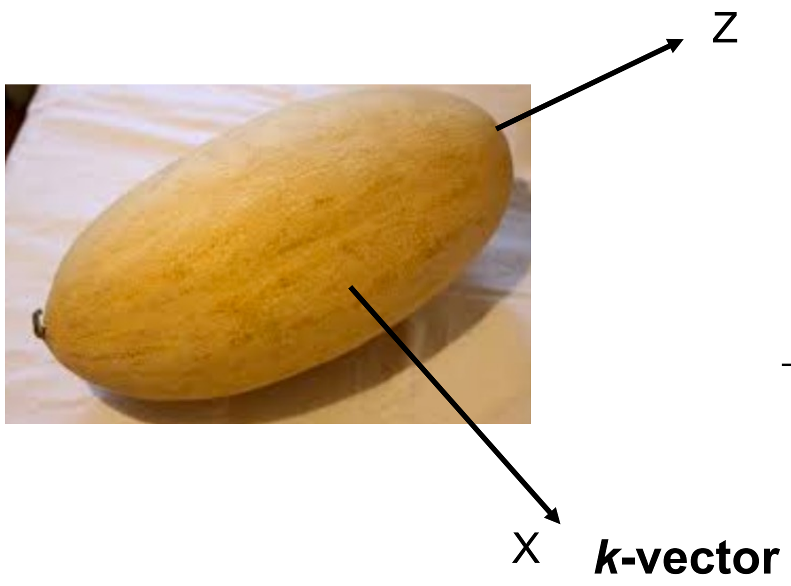


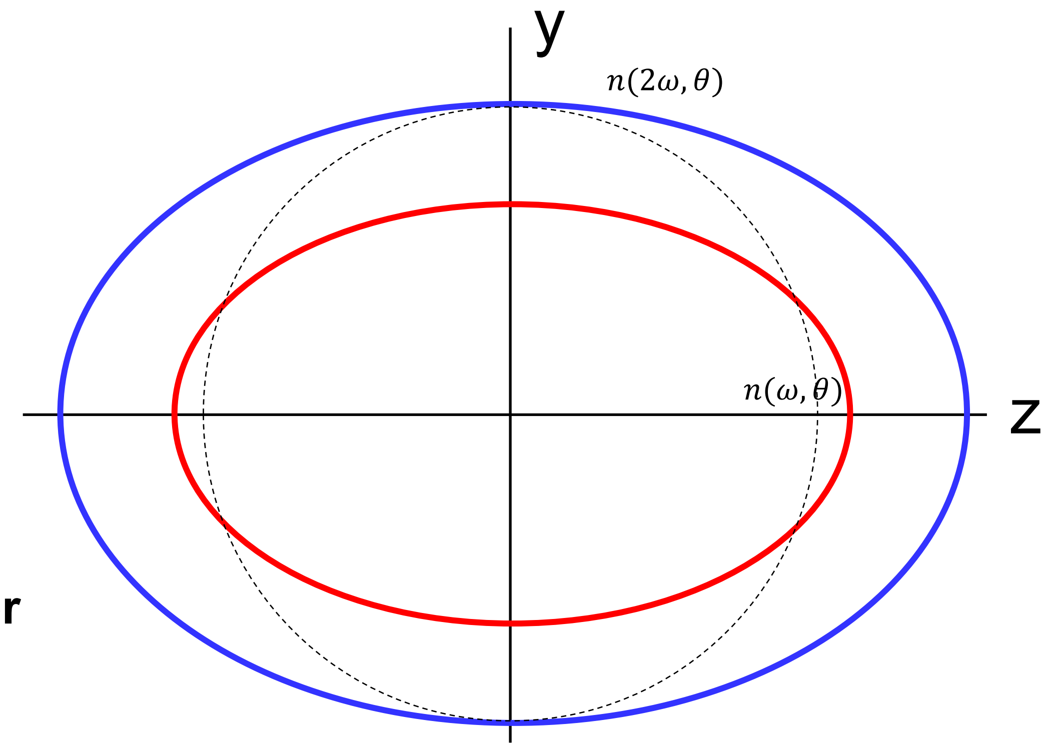
Now, there is **barely enough** birefringence to compensate chromatic dispersion

Phase matching

Un-isotropic crystal

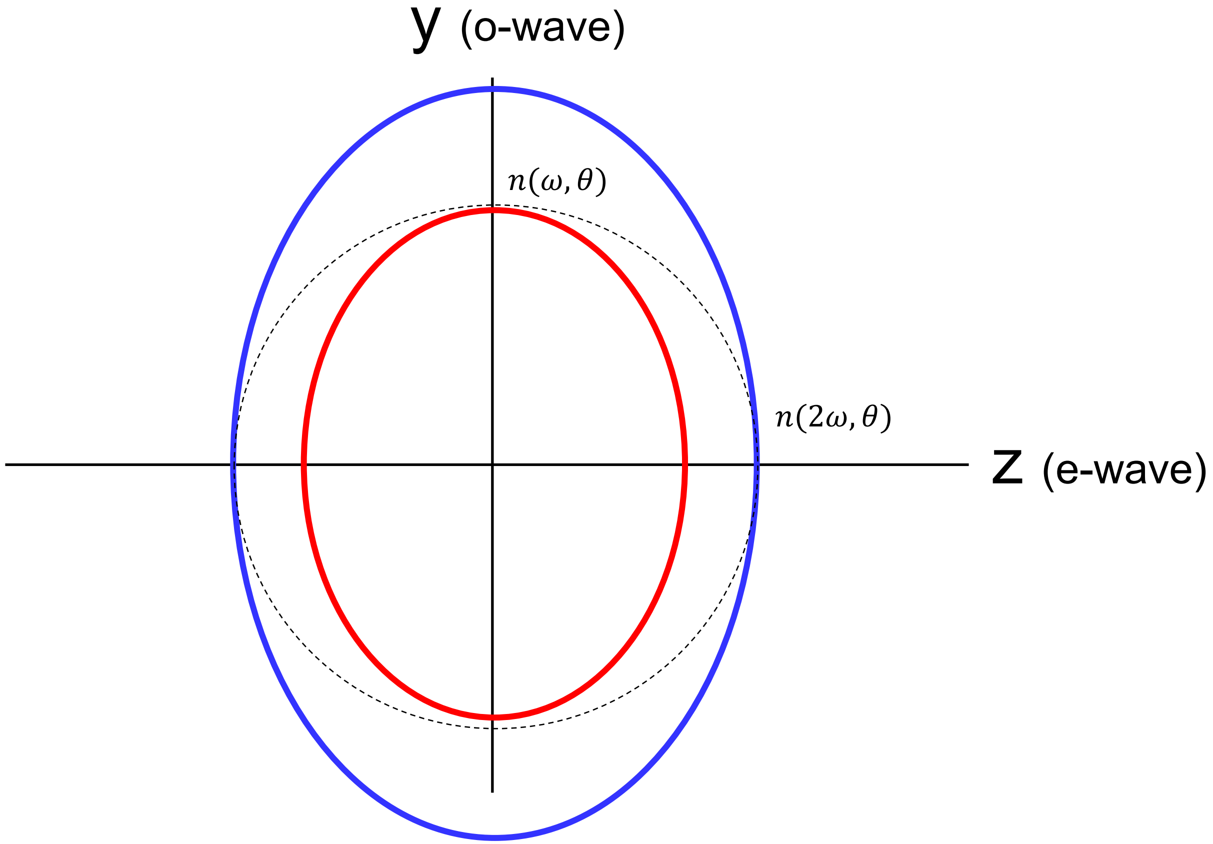
$$n_e > n_o$$





Now, there is **enough** birefringence to compensate chromatic dispersion

$$n_e < n_o$$



Second harmonic generation in **LBO crystal**

LBO crystal (mm2 symmetry)

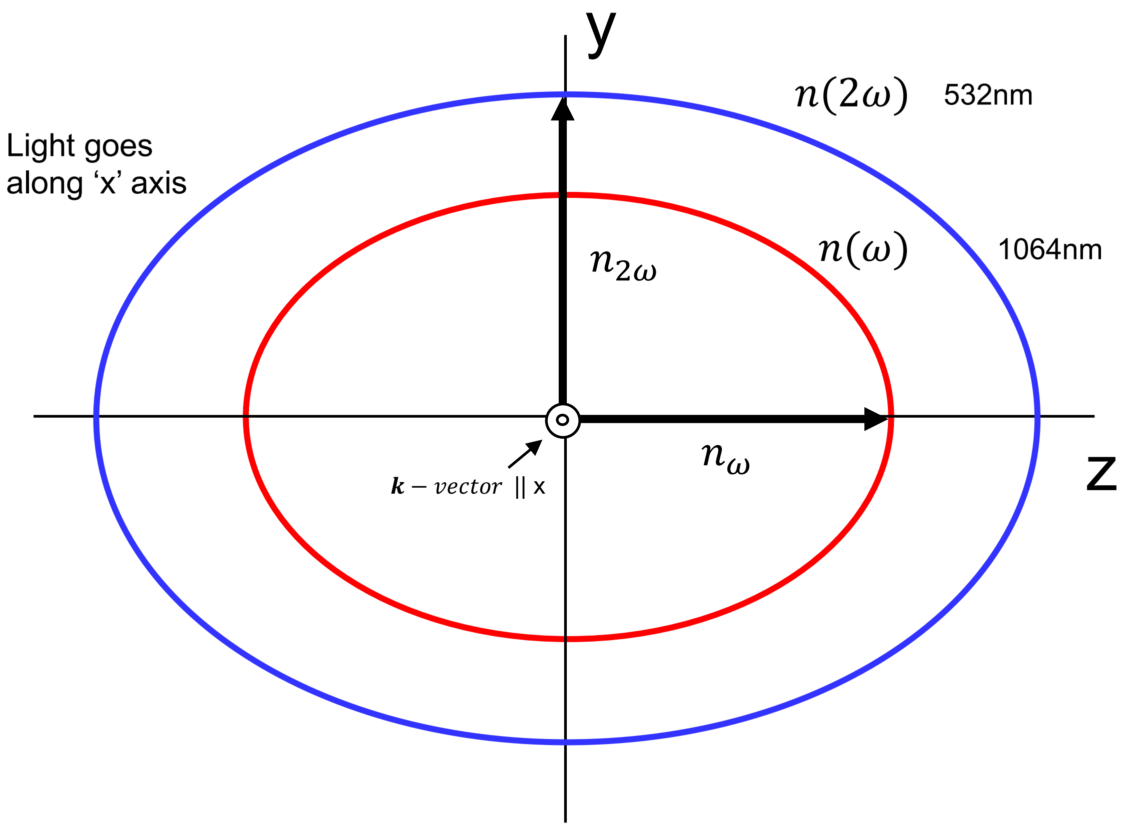
$$\boldsymbol{n_z > n_y > n_x}$$

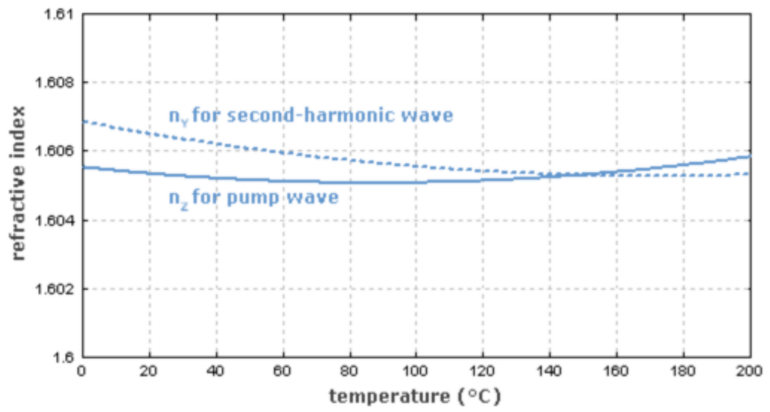
Birefringent noncritical phase matching of frequency doubling in LBO with a pump wavelength of 1064 nm.

The beams propagate in the X direction, the pump wave (ω) is polarized in the Z direction, and the second-harmonic wave in the Y direction.

At *t*=149 ºC, the **birefringence compensates** the effect of **chromatic dispersion**, so that the refractive indices for both waves are equal.

90º phase matching
= noncritical synchronism





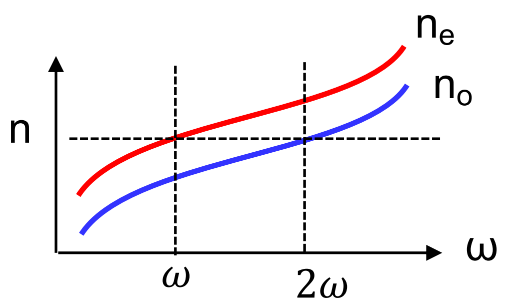
Phase matching

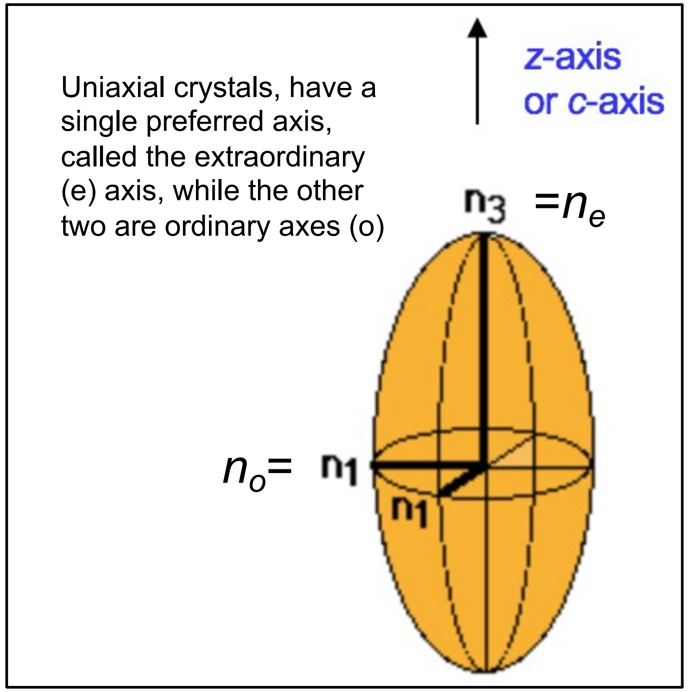
Example: SHG

Typically, three-wave mixing is done in a birefringent crystalline material, where the refractive index depends on the polarization and direction of the light that passes through.

Choose 2 orthogonal polarizations for ω and 2ω

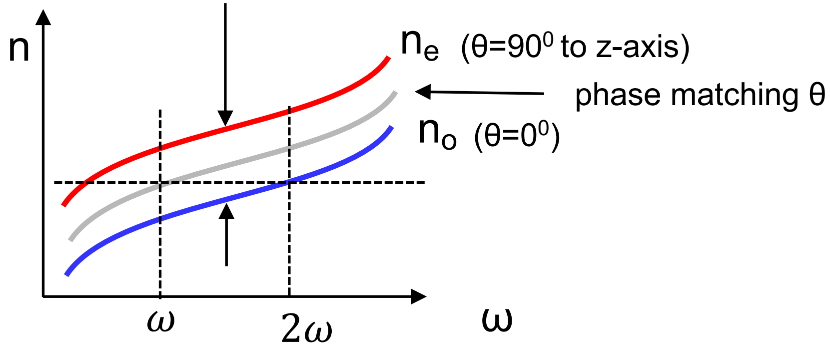
So called 90-deg phase matching (light goes perpendicular to z-axis)





More general case: angle tuning

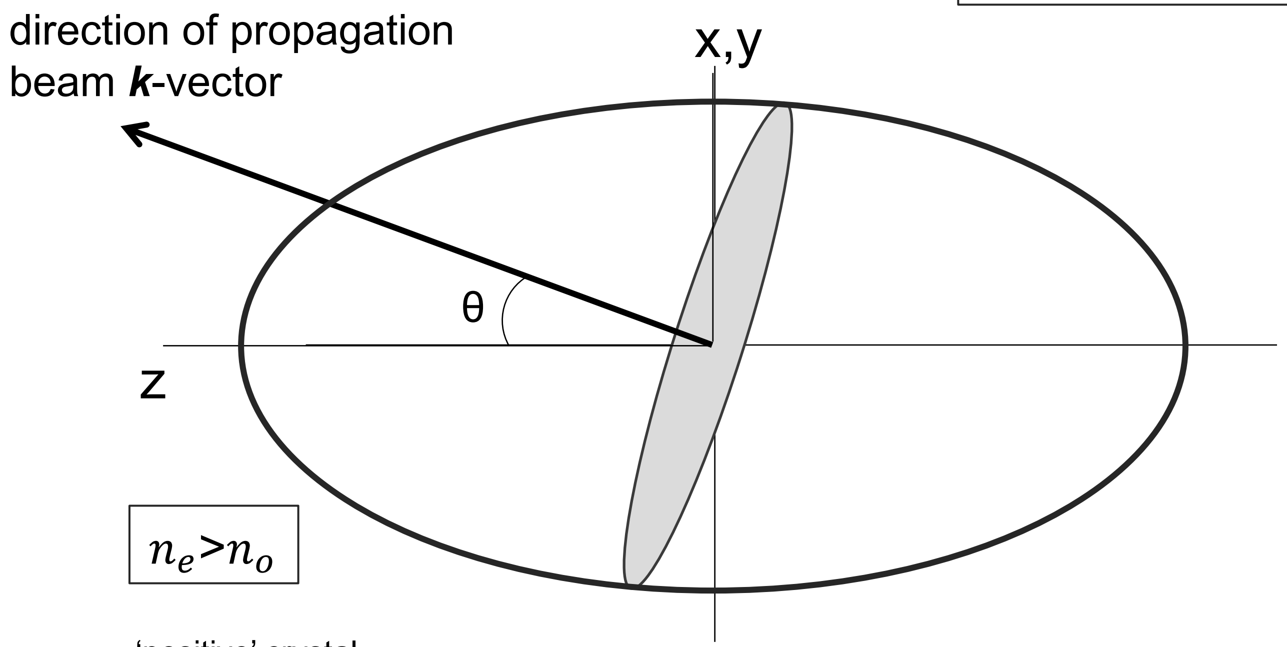
The polarizations of the fields and the orientation (angle) of the crystal are chosen such that the phase-matching condition is fulfilled.



Uniaxial crystals

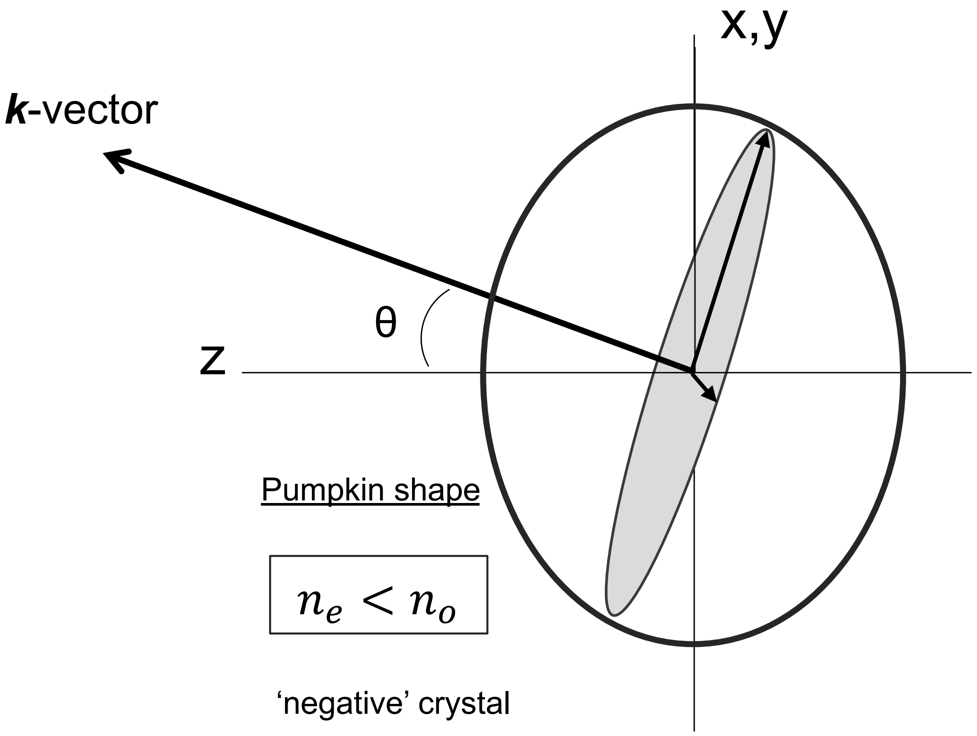
$n_1=n_2=n_o$; $n_3=n_e$

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



$n_e > n_o$

'positive' crystal



Pumpkin shape

$n_e < n_o$

'negative' crystal

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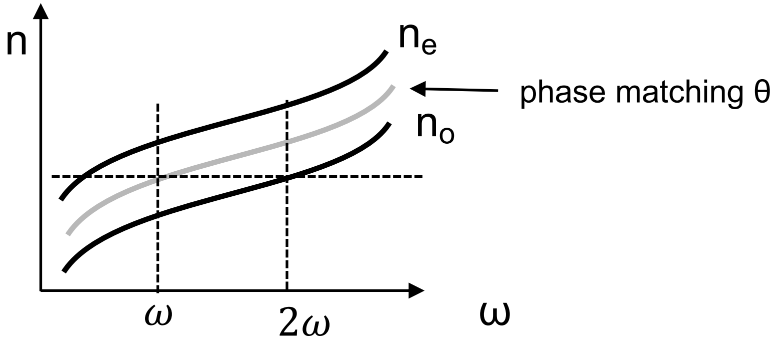
perpendicular to the plane of the figure $n = n_o$

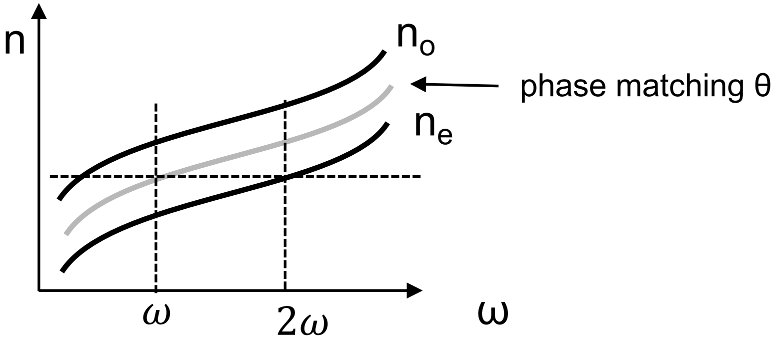
in the plane of the figure $$\frac{1}{n(\theta)^2}=\frac{cos(\theta)^2}{n_o^2}+\frac{\sin(\theta)^2}{n_e^2}$$ → $$n_e(\theta)=\left(\frac{cos(\theta)^2}{n_o^2}+\frac{\sin(\theta)^2}{n_e^2}\right)^{-\frac{1}{2}}$$

Phase matching

$n_e > n_o$



$n_e < n_o$



Refractive index data (Sellmeier equations)

$$n^2(\lambda) = 1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3},$$

two equations:
for o-wave and for e-wave

Lithium niobate

$LiNbO_3$ point group 3m   uniaxial crystal

$n_e < n_o$
'negative' crystal

from lecture 7:

$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

$$d_{22} = 2.1 \frac{pm}{V}$$
$$d_{31} = -4.35 \frac{pm}{V}$$
$$d_{33} = 27.2 \frac{pm}{V}$$

Dmitriev et al., Handbook of NLO crystals (1997), p.125

Two scenarios for SFG:

1) Type-I SFG $\omega_1 + \omega_2 = \omega_3$
$o + o = e$

2) Type-II SFG $\omega_1 + \omega_2 = \omega_3$
$e + o = e$

Lithium niobate, $LiNbO_3$

$LiNbO_3$ point group 3m   uniaxial crystal

the index ellipsoid

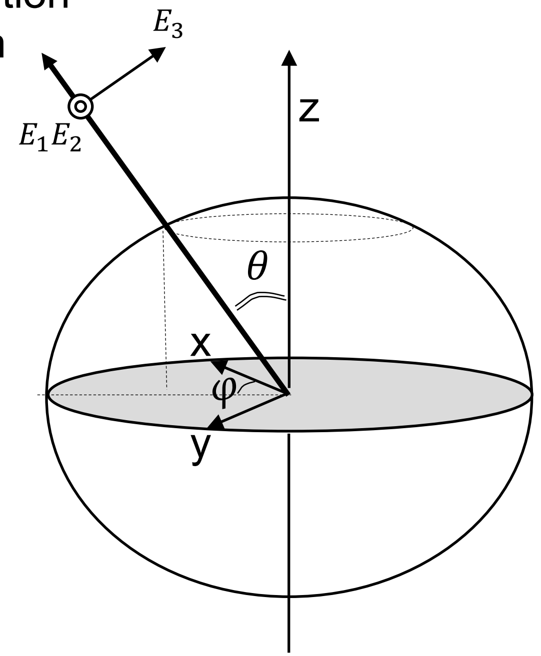
$n_e < n_o$

1) **Type-I SFG** $\omega_1 + \omega_2 = \omega_3$

$o + o = e$

input fields $E(t) = \boldsymbol{E_1} e^{i\omega_1 t} + \boldsymbol{E}_2 e^{i\omega_2 t}$ only *x,y* components

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix}$$



– pick only components at $\pm(\omega_1+\omega_2)$

$$E_x^2 = \frac{1}{4}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c.)^2 \sin\varphi^2 \to \frac{1}{2}(E_1 E_2 e^{i\omega_3 t} + c.c.)\sin\varphi^2$$

$$E_y^2 = \frac{1}{4}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c.)^2 \cos\varphi^2 \to \frac{1}{2}(E_1 E_2 e^{i\omega_3 t} + c.c.)\cos\varphi^2$$

$$2E_xE_y = 2\frac{1}{4}\left(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c.\right)^2 \sin\varphi\,(-\cos\varphi) \to -\frac{1}{2}\left(E_1 E_2 e^{i\omega_3 t} + c.c.\right) sin2\varphi$$

θ is the polar angle

φ is the azimuthal angle

Lithium niobate

$LiNbO_3$ point group 3m uniaxial crystal

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \left| \begin{matrix} d_{22}\,\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right)sin2\varphi \\ d_{22}\,\frac{1}{2}(E_1E_2e^{i\omega_3 t} + c.c.)(\cos\varphi^2\text{-}\sin\varphi^2) = d_{22}\,\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right)cos2\varphi \\ d_{31}\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right)(\cos\varphi^2 + \sin\varphi^2) = d_{31}\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right) \end{matrix} \right|$$

$$P_{NL}^{\omega_3}(t) = (P_x cos\varphi + P_y sin\varphi)cos\theta + P_z sin\theta = 2\epsilon_0\,\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right)\{d_{22}(sin2\varphi cos\varphi + sin\varphi cos2\varphi)cos\theta + d_{31}sin\theta\}$$

$$\frac{1}{2}\left(P(\omega_3)e^{i\omega_3 t} + c.c.\right) = 2\epsilon_0\,\frac{1}{2}\left(E_1E_2e^{i\omega_3 t} + c.c.\right)\underbrace{(d_{22}sin3\varphi cos\theta + d_{31}sin\theta)}$$

Finally, $$P(\omega_3) = 2\epsilon_0(d_{31}sin\theta + d_{22}sin3\varphi cos\theta)E_1E_2$$

$$\boxed{d_{eff} = d_{ooe} = d_{31}sin\theta + d_{22}sin3\varphi cos\theta \quad \rightarrow \quad |d_{31}|sin\theta + |d_{22}|cos\theta}$$

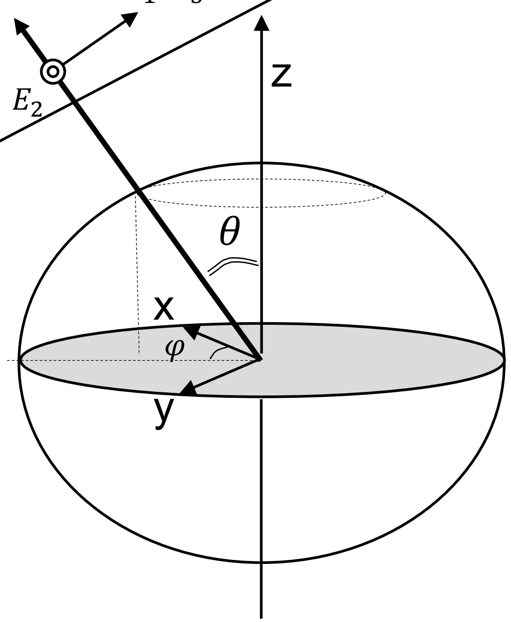
taking into account opposite signs of d_{31} and d_{22}, choose $sin3\varphi = -1$, i.e. φ=-90°

Lithium niobate

2) **Type-II SFG** $\omega_1 + \omega_2 = \omega_3$

$e + o = e$

the two lower frequency waves have orthogonal polarizations



input fields $E(t) = Re(\boldsymbol{E_1} e^{i\omega_1 t} + \boldsymbol{E}_2 e^{i\omega_2 t})$

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix}$$

this produces only $2\omega_2$ component

these two will cancel

– pick only components at $\pm(\omega_1+\omega_2)$

$$E_x^2 = \frac{1}{4}(E_1 \cos\theta(-\cos\varphi)\, e^{i\omega_1 t} + E_2 \sin\varphi\, e^{i\omega_2 t} \;+ c.c.)^2 \rightarrow \;\frac{1}{2}(-\cos\theta\cos\varphi\sin\varphi\, E_1E_2 e^{i\omega_3 t} \;+ c.c.)$$

$$E_y^2 = \frac{1}{4}(E_1 \cos\theta(-\sin\varphi)\, e^{i\omega_1 t} + E_2\,(-\cos\varphi)\, e^{i\omega_2 t} \;+ c.c.)^2 \rightarrow \frac{1}{2}(\cos\theta\cos\varphi\sin\varphi\, E_1E_2 e^{i\omega_3 t} \;+ c.c.)$$

$$2E_xE_y = 2\frac{1}{4}(E_1 \cos\theta(-\cos\varphi)\, e^{i\omega_1 t} + E_2 \sin\varphi\, e^{i\omega_2 t} \;+ c.c.)(E_1\cos\theta(-\sin\varphi)\, e^{i\omega_1 t} + E_2(-\cos\varphi)\, e^{i\omega_2 t} \;+ c.c.) =$$

$$\rightarrow \frac{1}{2}\left(E_1E_2\,(\cos\theta cos^2\varphi - cos\theta sin^2\varphi)\, e^{i\omega_3 t} \;+ c.c.\right) = \frac{1}{2}\left(E_1E_2 \cos\theta cos2\varphi\, e^{i\omega_3 t} \;+ c.c.\right)$$

Lithium niobate

$LiNbO_3$ point group 3m   uniaxial crystal

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \begin{vmatrix} -d_{22}\,\frac{1}{2}\left(E_1 E_2 \cos\theta cos2\varphi\, e^{i\omega_3 t} + c.c.\right) \\ 2d_{22}\,\frac{1}{2}\left(E_1 E_2 \cos\theta \cos\varphi \sin\varphi\, e^{i\omega_3 t} + c.c.\right) \\ 0 \end{vmatrix}$$

$$P_{NL}^{\omega_3}(t) = (P_x cos\varphi + P_y sin\varphi)cos\theta = 2\epsilon_0 d_{22}\,\frac{1}{2}\left(E_1 E_2 e^{i\omega_3 t} + c.c.\right)(-\cos\theta cos2\varphi cos\varphi cos\theta + \cos\theta \sin 2\varphi\, sin\varphi cos\theta) =$$

$$= -2\epsilon_0 d_{22}\,\frac{1}{2}\left(E_1 E_2 e^{i\omega_3 t} + c.c.\right)cos^2\theta(cos2\varphi cos\varphi - \sin 2\varphi\, sin\varphi) = -2\epsilon_0 d_{22}\,\frac{1}{2}\left(E_1 E_2 e^{i\omega_3 t} + c.c.\right)cos^2\theta cos3\varphi$$

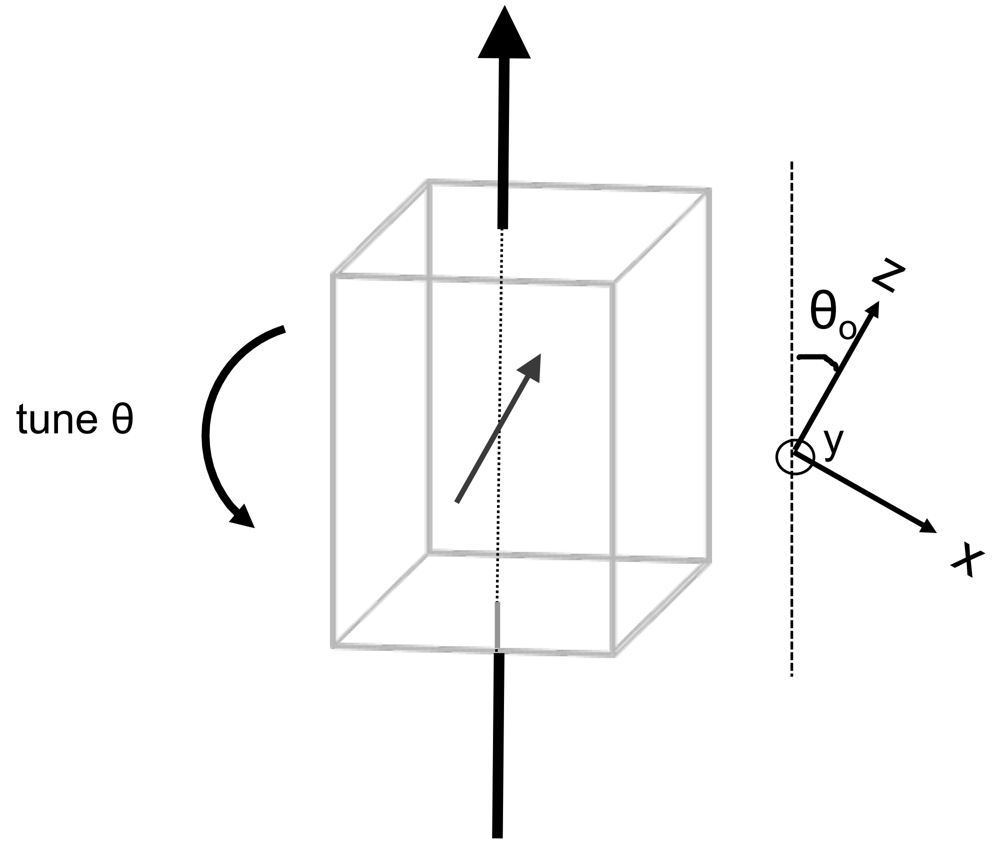
$$d_{eff} = d_{eoe} = -d_{22}cos^2\theta cos3\varphi \quad \rightarrow |d_{22}|cos^2\theta$$

choose $cos3\varphi = \pm 1$, i.e. φ=0, 60, 120, or 180º

Phase matching

Finally, need to make a proper cut and polish the crystal faces

θ – phase matching angle

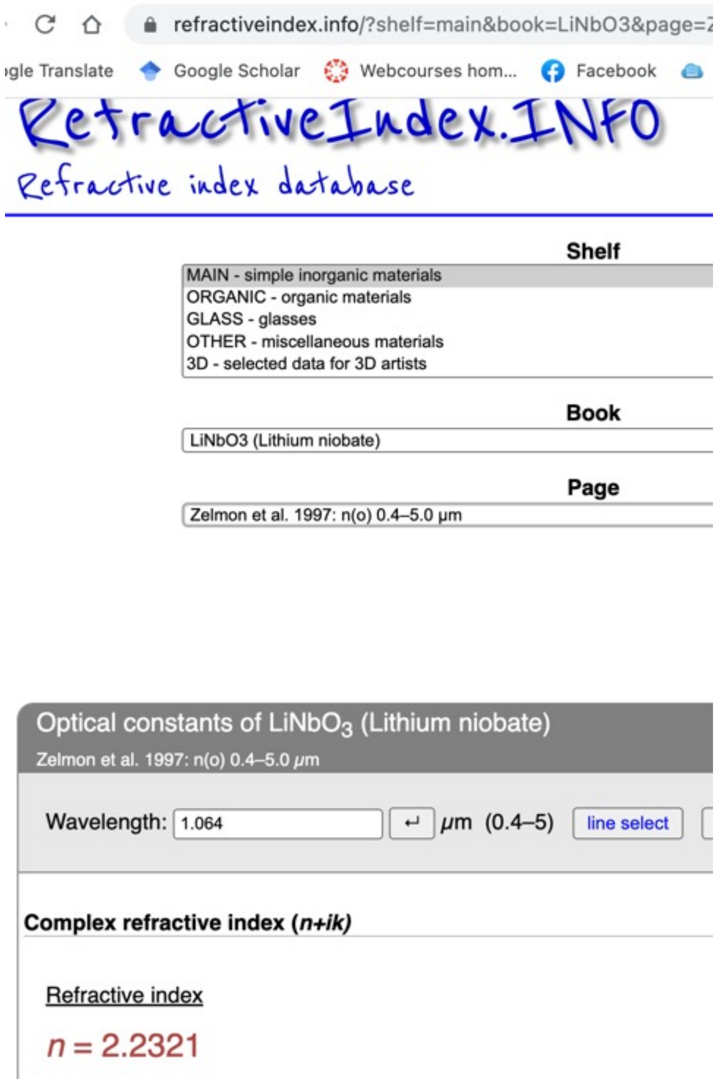


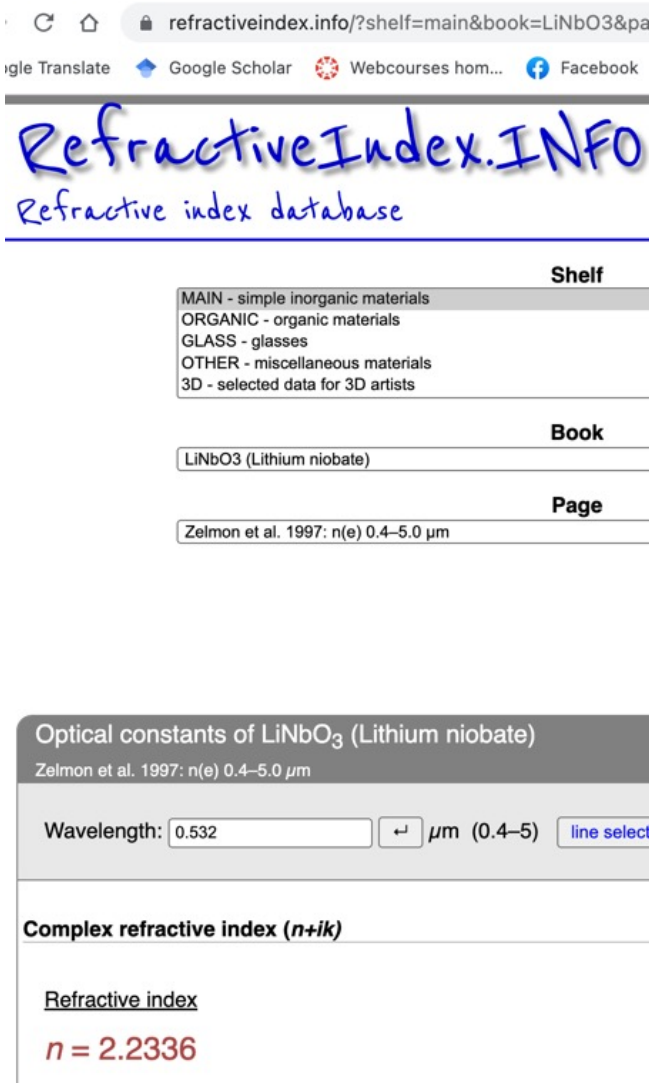
"θ_o - cut crystal"

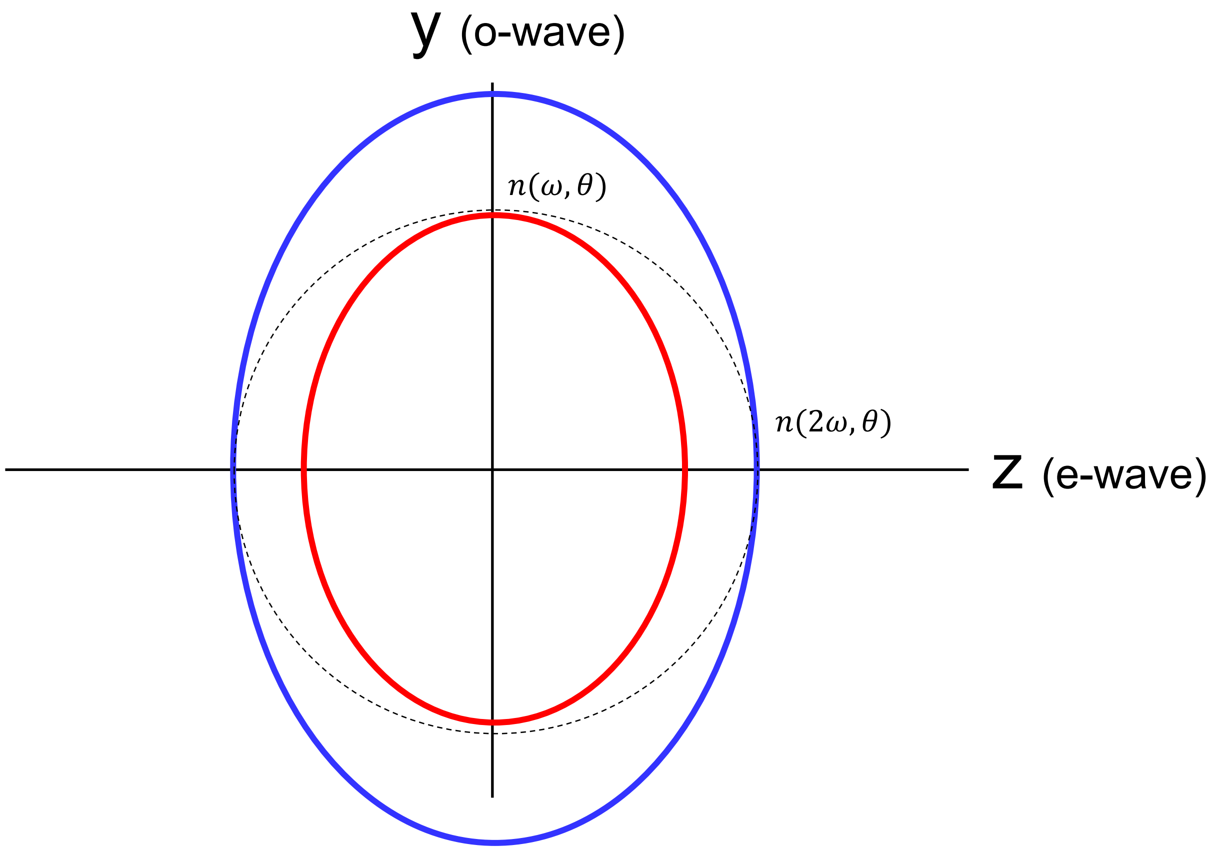
rotate crystal in the plain ZX

Phase matching, lithium niobate SHG from 1064 nm

$$n_e < n_o$$






y (o-wave)
$n(\omega, \theta)$
$n(2\omega, \theta)$
z (e-wave)

Collinear vs non-collinear phase matching

(a) (b)