Polarization in EM

 $\mathbf{E}(r,t) = \mathbf{E}_{\mathbf{0}} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}$ Plane wave state, arbitrary direction: In vacuum, $\mathbf{D} = \varepsilon_0 \mathbf{E}$ $\vec{\nabla} \cdot \mathbf{E} = \mathbf{0} \rightarrow \begin{pmatrix} \partial_x & \partial_y & \partial_z \end{pmatrix} \cdot \mathbf{E}_{\mathbf{0}} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$ $\vec{\nabla} \cdot \mathbf{E} = 0 \longrightarrow \left(\begin{array}{ccc} \partial_x & \partial_y & \partial_z \end{array} \right) \bullet \left(\begin{array}{ccc} E_{x0} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} & E_{y0} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \end{array} \right)$ $\vec{\nabla} \cdot \mathbf{E} = \partial_x E_{x0} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + \partial_y E_{y0} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} + \partial_z E_{z0} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$ $\partial_{u} E_{u0} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} = E_{u0} \partial_{u} e^{i(k_{x}x+k_{y}y+k_{z}z-\omega t)} = ik_{u} E_{u0} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}$ $\vec{\nabla} \cdot \mathbf{E} = i \left(k_x E_{x0} + k_y E_{y0} + k_z E_{z0} \right) e^{i \left(\mathbf{k} \cdot \mathbf{r} - \omega t \right)} = i \mathbf{k} \cdot \mathbf{E}_0 e^{i \left(\mathbf{k} \cdot \mathbf{r} - \omega t \right)} = 0$ From this we can say that $\mathbf{k} \cdot \mathbf{E}_{\mathbf{0}} = 0$ and $\mathbf{k} \perp \mathbf{E}$ Therefore, the electric field lies in a plane perpendicular to k The polarization direction can take on any linear combination of horizontal and vertical states (this includes circular polarization).

Other vector relations

Similarly,

$$\vec{\nabla} \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \rightarrow i \mathbf{k} \times \mathbf{E} = +i \omega \mathbf{B}$$
 SO $\mathbf{B} \perp \mathbf{k}, \mathbf{E}$

Energy flow is given by the Poynting vector:

$$\mathbf{S} = \mathbf{E} \times \mathbf{H} = \frac{1}{\mu_0} \mathbf{E} \times \mathbf{B}$$
 so $\mathbf{S} \perp \mathbf{E}, \mathbf{B}$
and $\mathbf{S} \parallel \mathbf{k}$

These relations hold in any isotropic medium. But if the medium is anisotropic, the vector relations must be modified.

$$\vec{\nabla} \cdot \mathbf{E} = 0 \qquad \vec{\nabla} \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$
$$\vec{\nabla} \cdot \mathbf{B} = 0 \qquad \vec{\nabla} \times \mathbf{B} = \mu_0 \frac{\partial \mathbf{D}}{\partial t}$$

Maxwell's Equations: linear anisotropic medium

• The induced polarization, **P**, contains the effect of the medium:

$$\vec{\nabla} \cdot \mathbf{D} = 0 \qquad \vec{\nabla} \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$
Define the displacement vector
$$\vec{\nabla} \cdot \mathbf{B} = 0 \qquad \vec{\nabla} \times \mathbf{B} = \mu_0 \frac{\partial \mathbf{D}}{\partial t}$$

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$$

• In an anisotropic medium:

$$\mathbf{P}(\mathbf{E}) = \varepsilon_0 \vec{\chi} \cdot \mathbf{E}, \quad \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_0 (1 + \vec{\chi}) \cdot \mathbf{E} = \varepsilon_0 \vec{\varepsilon} \cdot \mathbf{E}$$

So now **D** and **E** are not necessarily parallel.

$$\mathbf{D} = \boldsymbol{\varepsilon}_{0} \boldsymbol{\vec{\varepsilon}} \cdot \mathbf{E} \rightarrow \begin{pmatrix} D_{x} \\ D_{y} \\ D_{z} \end{pmatrix} = \boldsymbol{\varepsilon}_{0} \begin{pmatrix} \boldsymbol{\varepsilon}_{xx} & \boldsymbol{\varepsilon}_{xy} & \boldsymbol{\varepsilon}_{xz} \\ \boldsymbol{\varepsilon}_{yx} & \boldsymbol{\varepsilon}_{yy} & \boldsymbol{\varepsilon}_{yz} \\ \boldsymbol{\varepsilon}_{zx} & \boldsymbol{\varepsilon}_{zy} & \boldsymbol{\varepsilon}_{zz} \end{pmatrix} \begin{pmatrix} E_{x} \\ E_{y} \\ E_{z} \end{pmatrix}$$

Linear anisotropic response

For anisotropic linear response: $D_i = \varepsilon_0 \sum_i \varepsilon_{ij} E_j = \varepsilon_0 \varepsilon_{ij} E_j$

In a basis aligned with the crystal axes:

Contracted notation Repeated indices are summed

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \begin{bmatrix} \varepsilon_{XX} & 0 & 0 \\ 0 & \varepsilon_{YY} & 0 \\ 0 & 0 & \varepsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix} = \begin{bmatrix} \varepsilon_o & 0 & 0 \\ 0 & \varepsilon_o & 0 \\ 0 & 0 & \varepsilon_e \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix}$$

Uniaxial case: o = "ordinary"

Even here, D is not necessarily parallel to E

e = "extraordinary"

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \begin{bmatrix} \varepsilon_o & 0 & 0 \\ 0 & \varepsilon_o & 0 \\ 0 & 0 & \varepsilon_e \end{bmatrix} \begin{bmatrix} 0 \\ E_Y \\ E_Z \end{bmatrix} = \begin{bmatrix} 0 \\ \varepsilon_o E_Y \\ \varepsilon_e E_Z \end{bmatrix}$$

Linear tensor $\chi^{(1)}$

TABLE 1.5.1 Form of the linear susceptibility tensor $\chi^{()}$ as determined by the symmetry properties of the optical medium, for each of the seven crystal classes and for isotropic materials. Each nonvanishing element is denoted by its cartesian indices

Triclinic	xxxyxzyxyyyzzxzyzz	
Monoclinic	$\begin{bmatrix} xx & 0 & xz \\ 0 & yy & 0 \\ zx & 0 & zz \end{bmatrix}$	
Orthorhombic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$	biaxial
Tetragonal Trigonal Hexagonal	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$	uniaxial
Cubic Isotropic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & xx \end{bmatrix}$	isotropic

The dielectric tensor ε_{ij} is *symmetric* in a nonabsorbing medium.

 $\frac{\partial U}{\partial t} = -\nabla \cdot \mathbf{S}$ Continuity equation: Rate of change of er

 ∂t Rate of change of energy density = - div of power flow

$$\nabla \cdot \mathbf{S} = \nabla \cdot (\mathbf{E} \times \mathbf{H}) = \mathbf{H} \cdot (\nabla \times \mathbf{E}) - \mathbf{E} \cdot (\nabla \times \mathbf{H})$$
$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}$$

 $\Rightarrow -\nabla \cdot \mathbf{S} = \mathbf{E} \cdot \dot{\mathbf{D}} + \mathbf{H} \cdot \dot{\mathbf{B}} = \dot{U}_E + \dot{U}_H \Rightarrow \dot{U}_E = \mathbf{E} \cdot \dot{\mathbf{D}} = E_i \varepsilon_{ij} \dot{E}_j$

In the last step we're identifying the E and M components of the energy density. But we also know:

$$U_E = \frac{1}{2} \mathbf{E} \cdot \mathbf{D} \Longrightarrow \dot{U}_E = \frac{1}{2} \varepsilon_{ij} (\dot{E}_i E_j + E_i \dot{E}_j) = \frac{1}{2} (\varepsilon_{ji} + \varepsilon_{ij}) E_i \dot{E}_j$$

Therefore, the dielectric tensor is symmetric $\varepsilon_{ii} = \varepsilon_{ij}$

This is an example of an intrinsic symmetry. It comes from the nature of the thermodynamic requirement that U_E is a state function of **E**, which takes all E_i as independent variables. This does not require the symmetry of the crystal, or the linearity of the response.

The index ellipsoid

Energy density inside the medium: see Davis 18.3 for derivation

$$U_E = \frac{1}{2} \mathbf{D} \cdot \mathbf{E} = \frac{1}{2} \varepsilon_0 \sum_{ij} \varepsilon_{ij} E_i E_j$$

The index ellipsoid is a surface of constant energy density (in the crystal basis):

$$U_{E} = \frac{1}{2\varepsilon_{0}} \left(\frac{D_{X}^{2}}{\varepsilon_{XX}} + \frac{D_{Y}^{2}}{\varepsilon_{YY}} + \frac{D_{Z}^{2}}{\varepsilon_{ZZ}} \right) \rightarrow \frac{1}{2\varepsilon_{0}U} \left(\frac{D_{X}^{2}}{\varepsilon_{XX}} + \frac{D_{Y}^{2}}{\varepsilon_{YY}} + \frac{D_{Z}^{2}}{\varepsilon_{ZZ}} \right) = 1$$

Write this with new variables to make the ellipse equation more clear:

$$X = \left(\frac{1}{2\varepsilon_0 U_E}\right)^{1/2} D_X \quad \text{etc.} \qquad 1 = \frac{X^2}{\varepsilon_{XX}} + \frac{Y^2}{\varepsilon_{YY}} + \frac{Z^2}{\varepsilon_{ZZ}} = \frac{X^2}{n_o^2} + \frac{Y^2}{n_o^2} + \frac{Z^2}{n_e^2}$$

In an arbitrary basis, the ellipse equation looks like:

$$\left(\frac{1}{n^2}\right)_1 x^2 + \left(\frac{1}{n^2}\right)_2 y^2 + \left(\frac{1}{n^2}\right)_3 z^2 + 2\left(\frac{1}{n^2}\right)_4 yz + 2\left(\frac{1}{n^2}\right)_5 xz + 2\left(\frac{1}{n^2}\right)_6 xy = 1$$

The indices 1-6 are like the contracted notation we will use for second-order NLO

Wave propagation in birefringent crystals

Inside the medium, $\vec{\nabla} \cdot \mathbf{D} = 0$ So $\vec{\nabla} \cdot \mathbf{D} = i\mathbf{k} \cdot \mathbf{D} = 0$ and $\mathbf{k} \perp \mathbf{D}$ The wave is described by the D-field inside the medium.

If a wave is linearly polarized, and the D-field is oriented along one of the crystal axes, the wave sees only the refractive index corresponding to the direction of D..

$$\mathbf{D}(r,t) = \mathbf{D}_{\mathbf{0}} e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \rightarrow \hat{\mathbf{z}} D_{z} e^{i(k_{x}x-\omega t)}, \quad k_{x} = \frac{\omega}{c} n_{e}$$

If k is parallel to one of the axes, but D is not, the input polarization can be resolved along o- and e- axes:

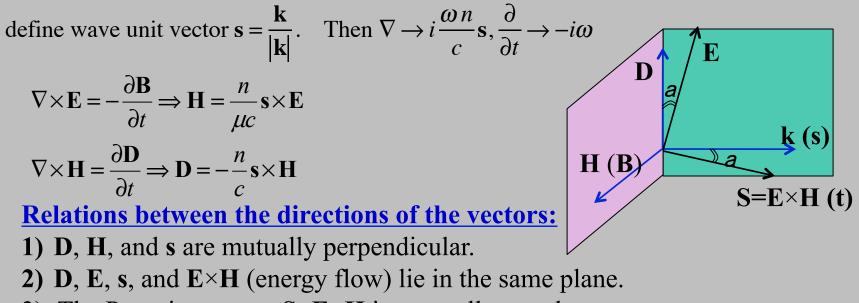
$$\mathbf{D}(r,t) = \mathbf{D}_{0}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \rightarrow \hat{\mathbf{z}} D_{z}e^{i\left(\frac{\omega}{c}n_{e}x-\omega t\right)} + \hat{\mathbf{y}} D_{y}e^{i\left(\frac{\omega}{c}n_{o}x-\omega t\right)}$$

In Jones vector notation,
$$\mathbf{D}(r,t) = D_{0}\begin{pmatrix}a\\b\end{pmatrix} \rightarrow D_{0}\begin{pmatrix}ae^{i\frac{\omega}{c}n_{o}x}\\be^{i\frac{\omega}{c}n_{e}x}\end{pmatrix} \qquad \text{Tr}$$
pic sh

The vector components pick up relative phase shift $\Delta \phi = \frac{\omega}{c} (n_0 - n_e) x$

Plane wave propagation: general direction

- In an anisotropic medium, the phase velocity of light depends on its polarization state and its propagation direction.
- For a given propagation direction, there exist in general two waves, each having its own refractive index (or equivalently phase velocity) and polarization.
- All light traveling in that direction can be decomposed onto the two eigenwaves.



3) The Poynting vector $S = E \times H$ is generally not along s.

Using the index ellipsoid: tuning the refractive index

The role of the index ellipsoid:

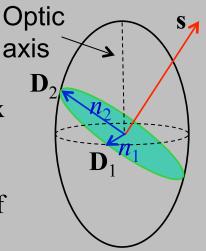
For a given arbitrary wave normal direction \mathbf{s} , the index ellipsoid can be used to

Find the indices of refraction of the two eigenwaves.
 Find the corresponding directions of the **D** vectors of the two eigen waves.

The **prescription** is as follows:

1)Draw a plane that is through the origin and is perpendicular to s. This plane intersects the index ellipsoid surface with a particular *intersection ellipse*. 2)The lengths of the two semiaxes of the intersection ellipse, n_1 and n_2 , are the two indices of refraction of the eigenwaves.

3)The two axes of the intersection ellipse are each parallel to the allowed **D** vectors of the eigenwaves.



Computation of the angle-dependent refractive index The index ellipsoid:

The equation of the index ellipsoid of a uniaxial crystal is

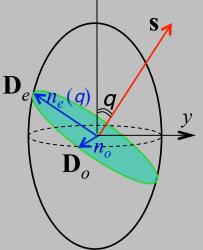
$$\frac{x^2 + y^2}{n_o^2} + \frac{z^2}{n_e^2} =$$

 $n_o = \sqrt{\varepsilon_x / \varepsilon_0} = \sqrt{\varepsilon_y / \varepsilon_0}$, ordinary refractive index

 $n_e = \sqrt{\varepsilon_z / \varepsilon_0}$, extraordinary refractive index

 $n_e > n_o$: positive uniaxial crystal \rightarrow prolate spheroid

 $n_e < n_o$: negative uniaxial crystal \rightarrow oblate spheroid



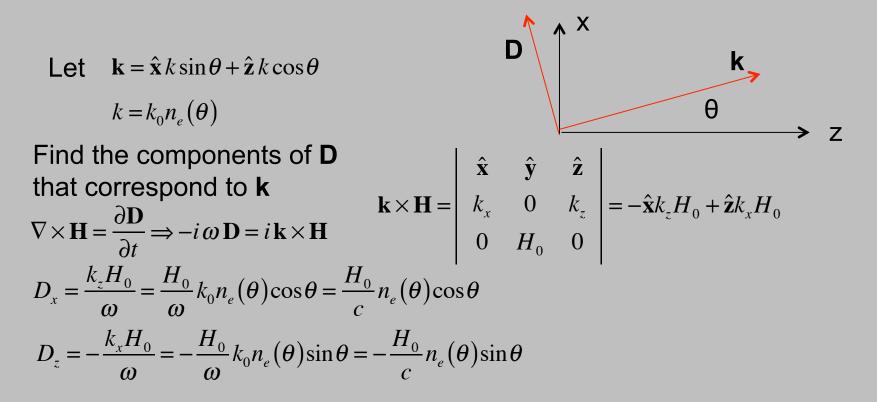
The uniaxial index ellipsoid is rotationally symmetric around the z-axis. Let **s** be in the y-z plane with a polar angle q. The two polarization directions of the **D** vectors are: \mathbf{D}_o is parallel to the x-axis, \mathbf{D}_e is in the y-z plane and is perpendicular to **s**.

The corresponding refractive indices are:

$$\begin{cases} n_o = n_o, \\ \frac{\left[n_e(\theta)\cos\theta\right]^2}{n_o^2} + \frac{\left[n_e(\theta)\sin\theta\right]^2}{n_e^2} = 1 \Rightarrow n_e(\theta) = \left(\frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}\right)^{-1/2} \end{cases}$$

When **s** is on the *z* direction, $n_e(0^\circ) = n_o$. Therefore the *z*-axis is the optic axis.

A derivation of the angle-dependent refractive index



A derivation of the angle-dependent refractive index

Put these into equation for ellipsoid: $\frac{1}{2\varepsilon_0 U_E} \left(\frac{D_X^2}{\varepsilon_{XX}} + \frac{D_Y^2}{\varepsilon_{YY}} + \frac{D_Z^2}{\varepsilon_{ZZ}} \right) = 1 = \frac{1}{2\varepsilon_0 U_E} \left(\frac{D_X^2}{n_o^2} + \frac{D_Y^2}{n_o^2} + \frac{D_Z^2}{n_e^2} \right)$

$$\frac{1}{2\varepsilon_0 U_E} \frac{H_0^2}{c^2} n_e^2(\theta) \left(\frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}\right) = 1$$

Magnetic energy density $U_H = \frac{\mu_0 H_0^2}{2}$ Is equal to electric $U_H = U_E$ energy density

$$\frac{1}{2\varepsilon_0 U_E} \frac{H_0^2}{c^2} = \frac{1}{2\varepsilon_0 U_E} \frac{2U_H}{\mu_0 c^2} = \frac{U_H}{U_E} \frac{1}{\mu_0 \varepsilon_0 c^2} = 1$$

Finally:
$$n_e^2(\theta) = \left(\frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2}\right)^2$$

The refractive index can be angle-tuned anywhere between n_e and n_o .

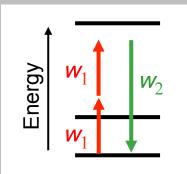
Phase matching: an important application for the the angle-dependent refractive index

Recall that for SHG:

$$\frac{\partial A_2}{\partial z}e^{i(k_2z-\omega_2t)} = i\frac{\omega_2^2d}{k_2c^2}A_1^2e^{i(2k_1z-2\omega_1t)}$$

Energy *must* be conserved:

$$\omega_1 + \omega_1 = \omega_2 \implies \omega_2 = 2\omega_1$$



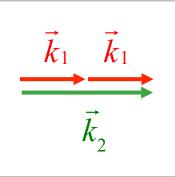
Momentum *may or may not* be conserved:

 $\frac{\partial A_2}{\partial z} = i \frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i\Delta kz} \quad \text{where} \quad \Delta k = 2k_1 - k_2$

Conversion will be most efficient if $\Delta k = 0$

$$\Rightarrow 2\frac{\omega_1}{c_0}n(\omega_1) = \frac{2\omega_1}{c_0}n(2\omega_1)$$

 $n(\omega_1) = n(2\omega_1)$



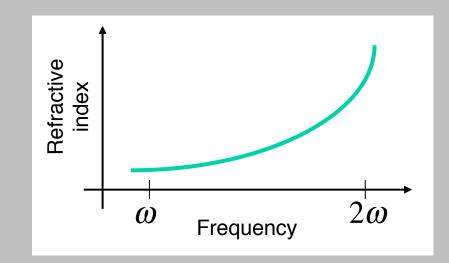
This is the *phase-matching condition* for SHG

Phase-matching Second-Harmonic Generation

The phase-matching condition for SHG:

 $n(\omega) = n(2\omega)$

Unfortunately, dispersion prevents this from ever happening!



First Demonstration of Second-Harmonic Generation

•P.A. Franken, et al, Physical Review Letters 7, p. 118 (1961)

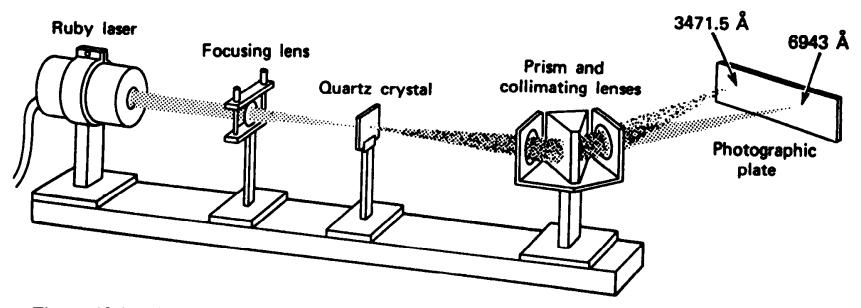
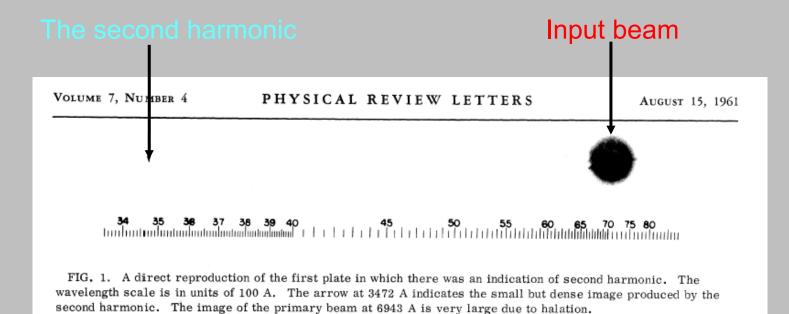


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694 \,\mu$ m is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347 \,\mu$ m. The two beams are then separated by a prism and detected on a photographic plate.

The second-harmonic beam was very weak because the process wasn't phase-matched.

First demonstration of SHG: The Data

The actual published result...



Note that the very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt.

SHG without phase-matching

Non-depleted pump approximation: treat A_1 as constant

$$\frac{\partial A_2}{\partial z} = i \frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i\Delta kz} \qquad \text{Integrate:} \quad A_2(L) = i \frac{\omega_2^2 d}{k_2 c^2} A_1^2 \int_0^L e^{i\Delta kz} dz$$
$$A_2(L) = i \frac{\omega_2^2 d}{k_2 c^2} A_1^2 L \frac{\left(e^{i\Delta kL} - 1\right)}{i\Delta kL}$$

Convert to intensity $I_2 = 2\varepsilon_0 n_2 c |A_2|^2$

$$\rightarrow \frac{1}{2\varepsilon_0 n_2 c} I_2(z) = \left(\frac{1}{2\varepsilon_0 n_1 c}\right)^2 I_1^2 \left(\frac{\omega_2 d}{n_2 c}\right)^2 L^2 \left(\frac{\sin(\Delta k L/2)}{\Delta k L/2}\right)^2$$

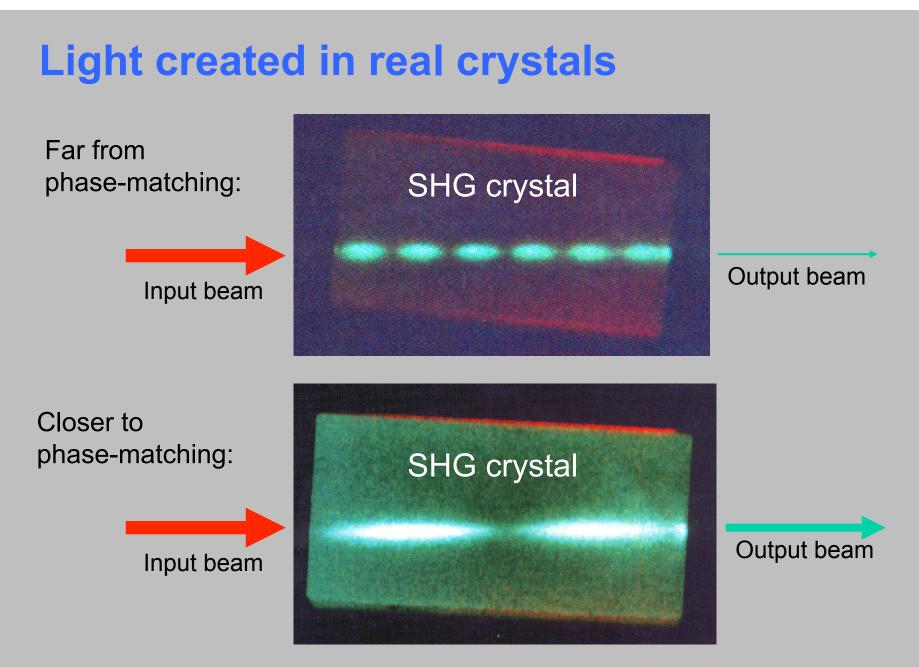
$$\rightarrow I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 L^2 \operatorname{sinc}^2 \left(\Delta k L / 2 \right)$$

As a function of L and fixed $|\Delta \mathbf{k}| > 0$: $I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 \frac{4}{\Delta k^2} \sin^2(\Delta k L/2)$

Yield oscillates:

 k_2c^2

- Period = "coherence length" $L_{coh} = 2\pi / \Delta k$
- Amplitude proportional to $\max(I_2) \propto 1/\Delta k^2$



Note that SH beam is brighter as phase-matching is achieved.

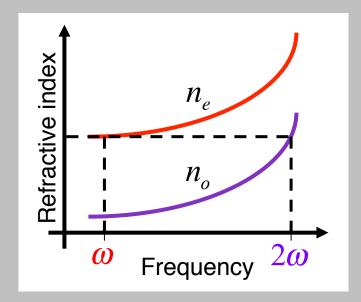
Phase-matching Second-Harmonic Generation using birefringence

Birefringent materials have different refractive indices for different polarizations. "Ordinary" and "Extraordinary" refractive indices can be different by up to 0.1 for SHG crystals.

We can now satisfy the phase-matching condition.

Put the highest frequency on the lowest index: for negative uniaxial use the extraordinary polarization for ω and the ordinary for 2ω :

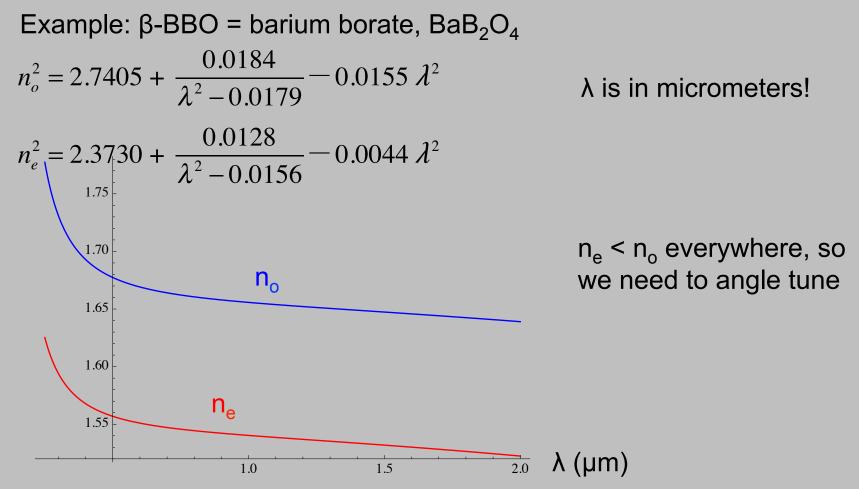
$$n_e(\omega,\theta) = n_o(2\omega)$$



 n_e depends on propagation angle, so we can tune for a given ω . Some crystals have $n_e < n_o$, so the opposite polarizations work.

Real crystal dispersion data

- Best resource: refractive index.info
- Others: crystal manufacturers, Handbook of Optics



Types of phase matching

- Type 1:
 - -2ω on low index (n_e)
 - ω on high (n_o)

$$\Delta k = 2 \frac{\omega_1}{c} n_o(\omega_1) - \frac{\omega_2}{c} n_e(\omega_2, \theta)$$
$$= 2 \frac{\omega_1}{c} (n_o(\omega_1) - n_e(\omega_2, \theta))$$

- Opposite polarizations ($\chi^{(2)}$ tensor allows this)
- Type 2: $\Delta k = \frac{\omega_1}{n} \left(\omega_1 \right) + \frac{\omega_1}{n} \left(\omega_2 \theta \right) - \frac{\omega_2}{n} \left(\omega_2 \theta \right)$
 - -2ω on low index (n_e)

$$k = \frac{\omega_1}{c} n_o(\omega_1) + \frac{\omega_1}{c} n_e(\omega_1, \theta) - \frac{\omega_2}{c} n_e(\omega_2, \theta)$$

- Project E_1 equally on both axes (n_o and n_e)
- Type 3:
 - "non-critical" or "90°" phase matching
 - Temperature-tuned $\Delta k = 2 \frac{\omega_1}{c} \left(n_o(\omega_1, T) n_e(\omega_2, 90, T) \right)$
 - Only for particular crystals and wavelengths

Practical issues

- Phase matching bandwidth
 - Type 1 has more BW, choose L of crystal
- Group velocity walk-off (for short pulses)
- Angular acceptance
- Birefringent beam walk-off
- Strength of nonlinearity
- Crystal damage threshold
- Thermal stability:
 - typically angle-tuned, temperature stabilized
- Available size of crystals, \$\$