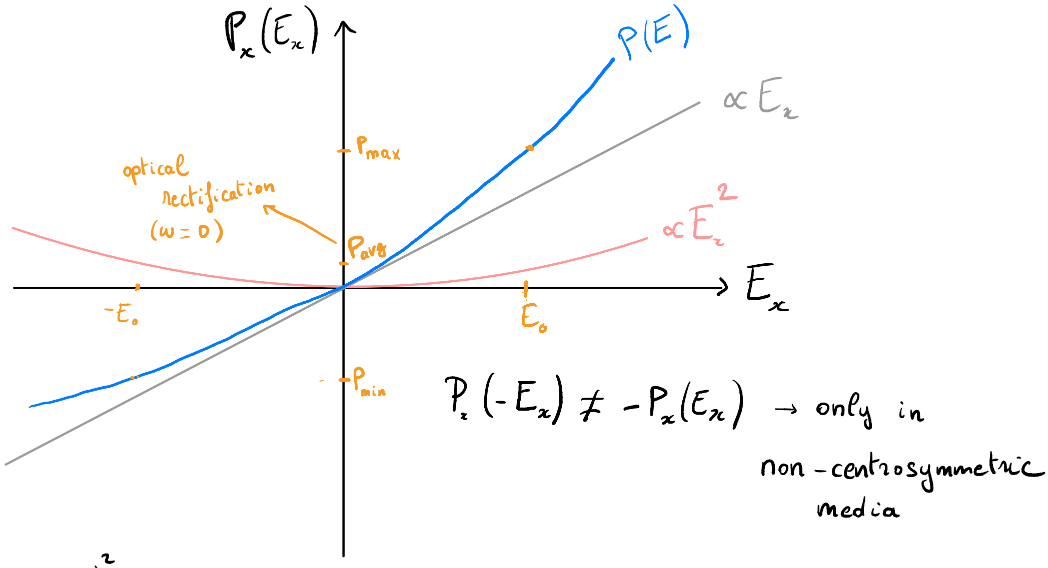
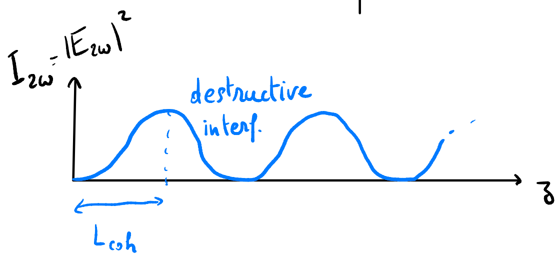
I. Motivation: Second Harmonic Generation (SHG)

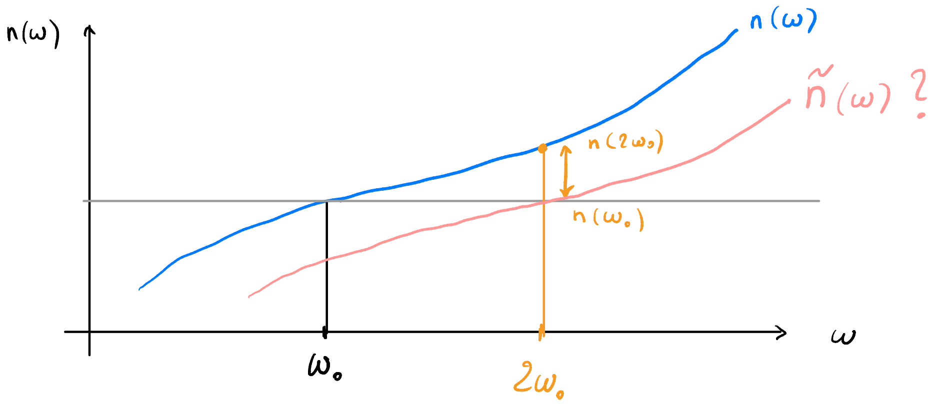
In Ex. 1 this week we have seen that a **quadratic** term in the response function $\vec{P}(\vec{E})$ induces a polarization density in the medium oscillating at frequency 2ω, under an external drive at frequency ω.





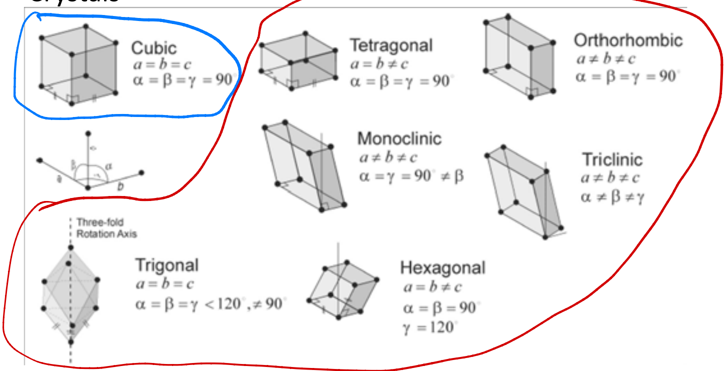
I. The problem of "phase matching"

- The polarization wave $\vec{P}_{2\omega}(z)$ at 2ω is phase-locked to the excitation field $\vec{E}_{\omega}(z)$ with phase velocity $\frac{c}{n(\omega)}$
- But the radiation $\vec{E}_{2\omega}(z)$ generated by $\vec{P}_{2\omega}$ propagated with phase velocity $\frac{c}{n(2\omega)}$
- In general, $n(2\omega) > n(\omega)$: the radiation $\vec{E}_{2\omega}$ generated at the beginning of the medium interferes destructively with one generated by $\vec{P}_{2\omega}$ some distance further
- No build up of power at 2ω !

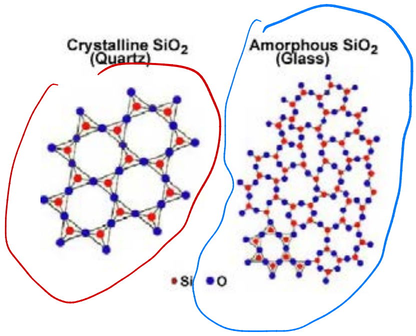


II. Isotropic vs. anisotropic media

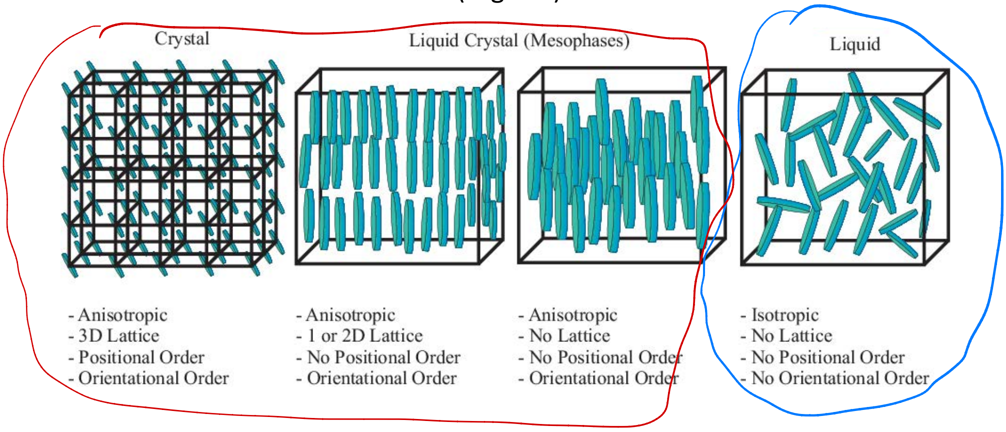
Crystals



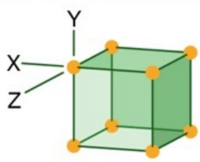
Amorphous materials (glasses)



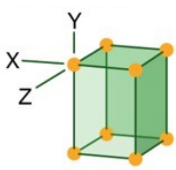
Molecular (organic) materials



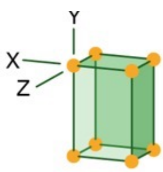
II. The seven primitive crystal systems



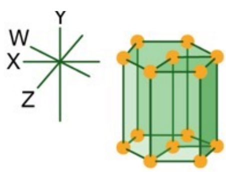
Isometric (or cubic)
All three axes are equal in length, and all are perpendicular to one another.



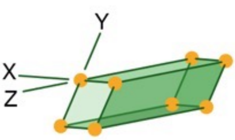
Tetragonal
Two of the three axes are equal in length, and all three axes are perpendicular to one another.



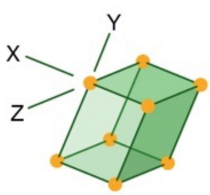
Orthorhombic
All three axes are unequal in length, and all are perpendicular to one another.



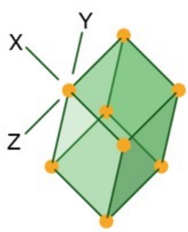
Hexagonal
Of four axes, three are of equal length, are separated by equal angles, and lie in the same plane. The fourth axis is perpendicular to the plane of the other three axes. Hexagonal cells have lattice points in each of the two six-sided faces.



Triclinic
All three axes are unequal in length, and none is perpendicular to another.



Monoclinic
All three axes are unequal in length, and two axes are perpendicular to each other.

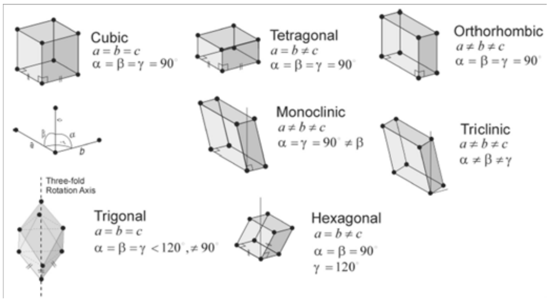


Rhombohedral (or trigonal)*
All three axes are of equal length, and none of the axes is perpendicular to another, but the crystal faces all have the same size and shape.

*Some sources do not separate the hexagonal and rhombohedral (trigonal) systems.

Optical Symmetry	Crystal System	Point Groups	Dielectric Tensor
Isotropic	Cubic	$\bar{4}3m$, 432, m3, 23, m3m	$\varepsilon = \varepsilon_0 \begin{pmatrix} n^2 & 0 & 0 \\ 0 & n^2 & 0 \\ 0 & 0 & n^2 \end{pmatrix}$
Uniaxial	Tetragonal	4, $\bar{4}$, 4/m, 422, 4mm, $\bar{4}2m$, 4/mmm	$\varepsilon = \varepsilon_0 \begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix}$
	Hexagonal	6, $\bar{6}$, 6/m, 622, 6mm, $\bar{6}m2$, 6/mmm	
	Trigonal	3, $\bar{3}$, 32, 3m, $\bar{3}m$	
Biaxial	Triclinic	1, $\bar{1}$	$\varepsilon = \varepsilon_0 \begin{pmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{pmatrix}$
	Monoclinic	2, m, 2/m	
	Orthorhombic	222, 2mm, mmm	

A. Yariv, P. Yeh (p 84)



uniaxial crystals possess a single optic axis, which is usually taken to be the *z* axis.

- *Ordinary* directions: *x* and *y*
- *Extraordinary* direction: *z*

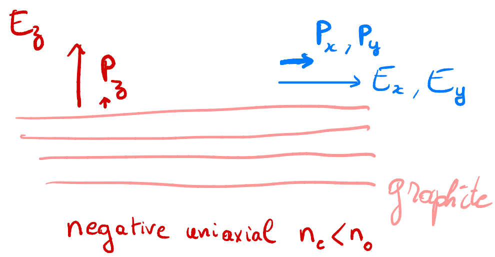


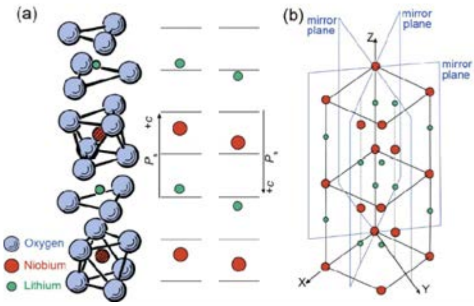
Table 2.1 Refractive indices of some common uniaxial crystals at 589.3 nm. Data from Driscoll & Vaughan (1978).

Crystal	Chemical structure	Symmetry class	Type	n_o	n_e
Ice	H_2O	trigonal	positive	1.309	1.313
Quartz	SiO_2	trigonal	positive	1.544	1.553
Beryl	$Be_3Al_2(SiO_3)_6$	hexagonal	negative	1.581	1.575
Sodium nitrate	$NaNO_3$	trigonal	negative	1.584	1.336
Calcite	$CaCO_3$	trigonal	negative	1.658	1.486
Tourmaline	complex silicate	trigonal	negative	1.669	1.638
Sapphire	Al_2O_3	trigonal	negative	1.768	1.760
Zircon	$ZrSiO_4$	tetragonal	positive	1.923	1.968
Rutile	TiO_2	tetragonal	positive	2.616	2.903

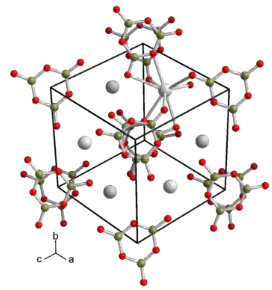
From *Mark Fox - Optical Properties of Solids* (2010)

II. Examples of uniaxial nonlinear crystals

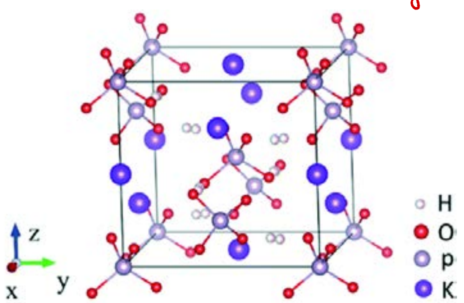
Lithium niobate ($LiNbO_3$, LN)
➢ Rhombohedral/trigonal *negative*



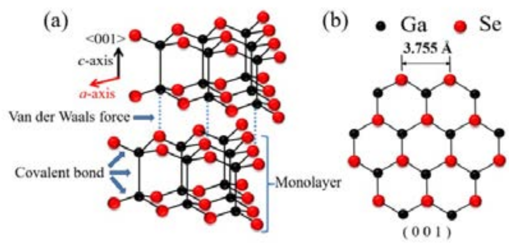
β-Barium borate (BBO)
➢ Rhombohedral/trigonal *negative*



Potassium dihydrogen phosphate (KDP)
➢ Tetragonal *negative*

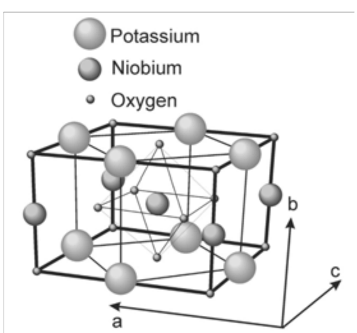


Gallium(II) selenide (GaSe)
➢ Hexagonal *negative*

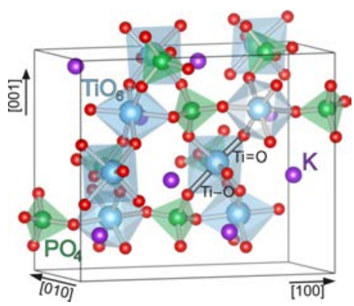


II. Examples of biaxial nonlinear crystals

Potassium Niobate ($KNbO_3$)
➢ Orthorhombic



Potassium Titanyl Phosphate (KTP)
➢ Orthorhombic



II. Anisotropic materials - definitions

- Crystals in which the three principal refractive indices are different are termed **biaxial**.
- For crystals with certain symmetries, namely *a single axis of threefold, fourfold, or sixfold symmetry*, two of the refractive indices are equal ($n_x = n_y$) and the crystal is called **uniaxial**.
  - In this case, the indices are usually denoted $n_x = n_y = n_o$ and $n_z = n_e$, which are known as the **ordinary** and **extraordinary** indices.
  - The crystal is said to be positive uniaxial if $n_e > n_o$, and negative uniaxial if $n_e < n_o$.
  - The z axis of a uniaxial crystal is called the **optic axis**.

- In crystals with greater symmetry (those with **cubic unit cells**), all three indices are equal and the medium is **optically isotropic**.

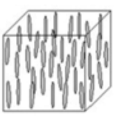
II. The index ellipsoid

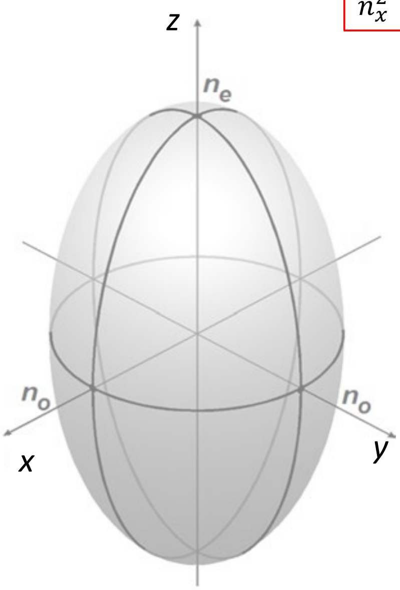
The index ellipsoid is a geometrical representation of the relative permittivity tensor, defined by the equation:

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

$$\frac{x_1^2}{n_1^2} + \frac{x_2^2}{n_2^2} + \frac{x_3^2}{n_3^2} = 1$$

Other notation for coordinates



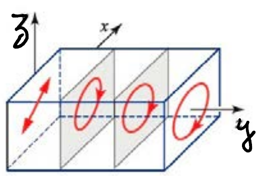


It is the quadratic representation of the electric impermeability tensor $\underline{\underline{\eta}} = \underline{\underline{\varepsilon_r}}^{-1}$:

$$\sum_{i,j} \eta_{ij}\, x_i\, x_j = 1$$

Along the principal axes:

$\eta_{xx} = \frac{1}{\varepsilon_{xx}} = \frac{1}{n_x^2}$ etc.

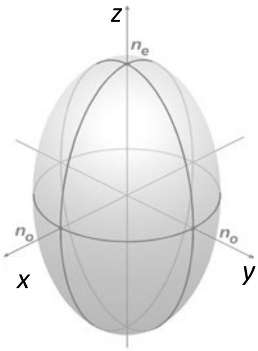


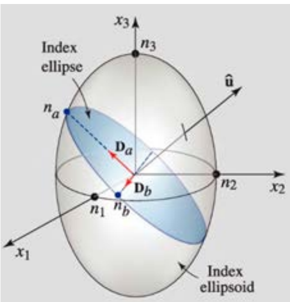
Born & Wolf, Ch. XV; Saleh & Teich, Ch. 6.3

II. The index ellipsoid

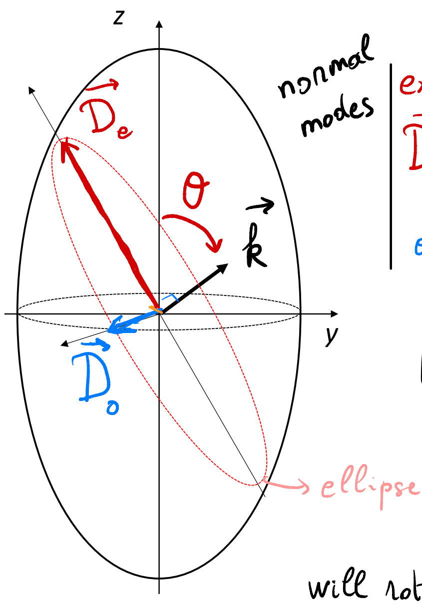
Propagation along an arbitrary direction

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





Saleh & Teich, Ch. 6.3



normal modes

- extraordinary wave $\vec{D}_e$
- ordinary wave $\vec{D}_o$ $\in (x, y)$

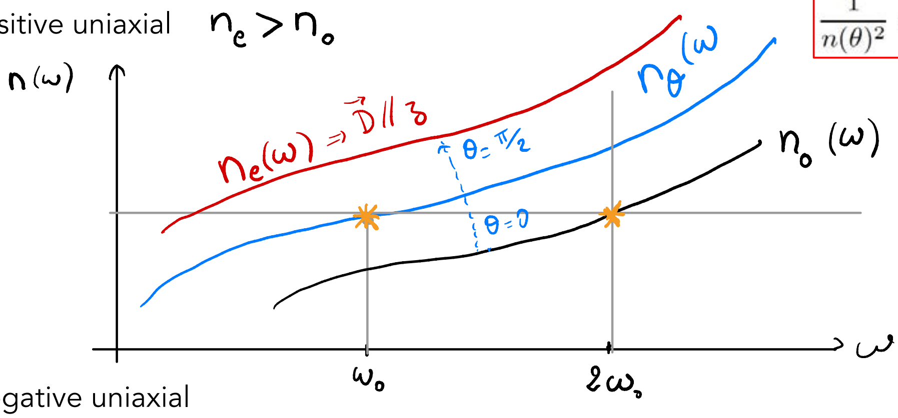
↳ propagate "unchanged" (linearly polarized)

All other polarizations will rotate upon propagation (linear → elliptical)

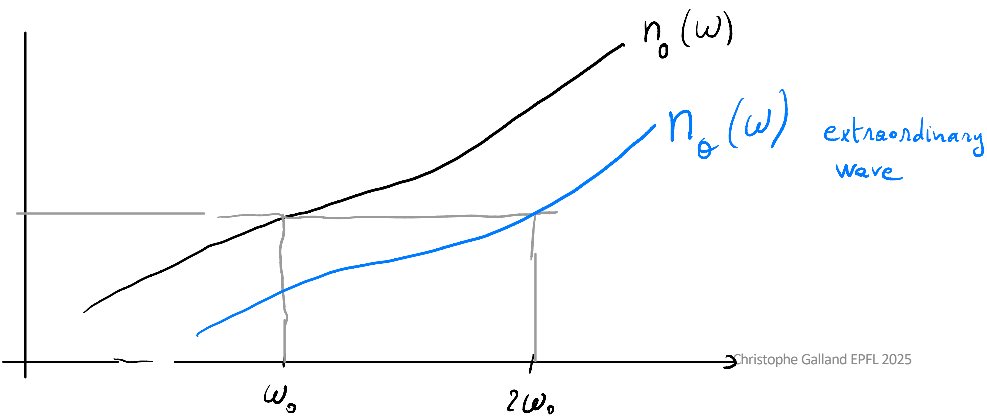
II. Phase matching of SHG in a uniaxial crystal

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

Positive uniaxial $n_e > n_o$



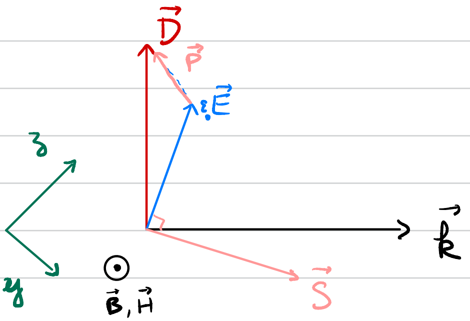
Negative uniaxial



III) Wave propagation

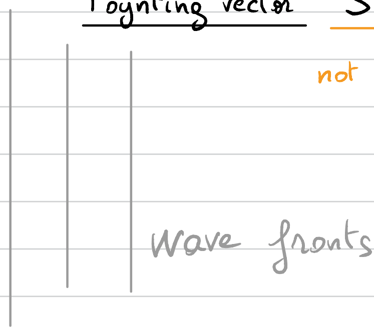
Reminder : $\vec{\nabla} \cdot \vec{D} = 0$ $(\rho_{ext} = 0)$ $\xrightarrow{FT}$ $\vec{k} \cdot \vec{D} = 0$ $\rightarrow$ $\vec{D} \perp \vec{k}$

$\vec{D}$ is transverse



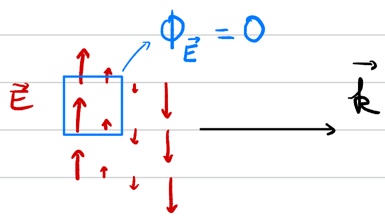
Poynting vector $\vec{S} = \vec{E} \times \vec{H}$

not // $\vec{k}$

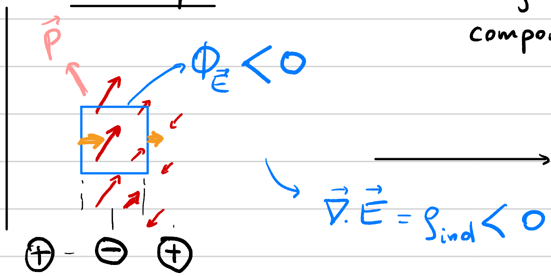


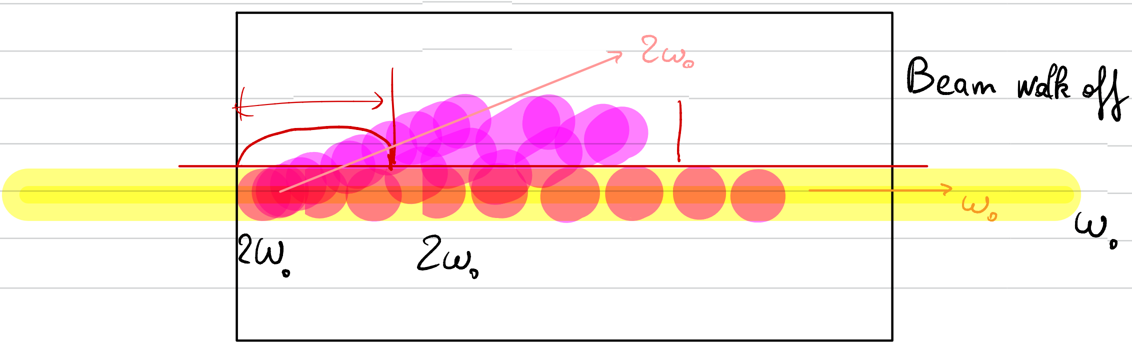
Consider a plane wave : what is the divergence of $\vec{E}$?

isotropic : $\vec{k} \perp \vec{E}$



anisotropic $\vec{k} \cdot \vec{E} \neq 0$ $\rightarrow$ Longitudinal component





Macroscopic Maxwell's equations

We remind the macroscopic Maxwell's equations in a non-magnetic medium without external charges, written in reciprocal space (after Fourier transform), with $\boldsymbol{B} = \mu_0 \boldsymbol{H}$ and $\boldsymbol{D} = \underline{\underline{\varepsilon}} \boldsymbol{E}$:

$$\begin{aligned}
\boldsymbol{k} \cdot \boldsymbol{B} &= 0 & \Longrightarrow & \quad \boldsymbol{B}, \boldsymbol{H} \perp \boldsymbol{k} & (1) \\
\boldsymbol{k} \times \boldsymbol{E} - \omega \boldsymbol{B} &= \boldsymbol{0} & \Longrightarrow & \quad \boldsymbol{B}, \boldsymbol{H} \perp \boldsymbol{E} & (2) \\
\boldsymbol{k} \times \boldsymbol{H} + \omega \boldsymbol{D} &= \boldsymbol{0} & \Longrightarrow & \quad \boldsymbol{D} \perp \boldsymbol{k}, \boldsymbol{H} & (3) \\
\boldsymbol{k} \cdot \boldsymbol{D} &= 0 & \Longrightarrow & \quad \boldsymbol{D} \perp \boldsymbol{k} & (4)
\end{aligned}$$

If $\underline{\underline{\varepsilon}} \neq \varepsilon I_3$, there is no reason to expect that $\boldsymbol{k} \cdot \boldsymbol{E} = 0$, and in general $\boldsymbol{E}$ is not a transverse field (unless the light is polarized exactly along one of the principal axes).
From eqs. (2) and (3) we can obtain an eigenvalue equation on $\boldsymbol{E}$:

$$(\boldsymbol{k} \cdot \boldsymbol{E})\boldsymbol{k} - k^2 \boldsymbol{E} + \omega^2 \mu_0 \underline{\underline{\varepsilon}} \boldsymbol{E} = \boldsymbol{0} \tag{5}$$

Dispersion relation in a uniaxial or biaxial media

Since the first term of eq. (5) does not cancel in general, the dispersion relation $\boldsymbol{k}(\omega)$ is not only a function of ω but also of the **polarization direction** of the field.

We take x, y, z along the principal axes and write

$$\omega^2 \mu_0 \underline{\underline{\varepsilon}} = k_0^2 \begin{bmatrix} n_x & 0 & 0 \\ 0 & n_y & 0 \\ 0 & 0 & n_z \end{bmatrix} \qquad \text{where} \qquad k_0 = \omega\sqrt{\mu_0 \varepsilon_0} = \frac{\omega}{c} \tag{6}$$

from which eq. (5) can be recast in a matrix form as

$$\begin{bmatrix} (k_0^2 n_x^2 - k_y^2 - k_z^2) & k_x k_y & k_x k_z \\ k_x k_y & (k_0^2 n_y^2 - k_x^2 - k_z^2) & k_y k_z \\ k_x k_z & k_y k_z & (k_0^2 n_z^2 - k_x^2 - k_y^2) \end{bmatrix} \cdot \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} = 0 \tag{7}$$

Non-zero solutions for the $\boldsymbol{E}$ field exist only if the determinant of the matrix is zero.

Dispersion relation in a uniaxial media

We now restrict ourselves to uniaxial media where $n_x = n_y = n_o$ and $n_z = n_e$. Equaling to zero the determinant of the matrix in eq. (7) yields after simplification

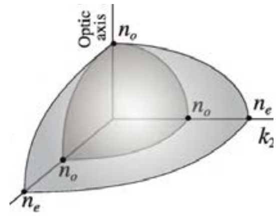
$$\left(k^2 - n_0^2 k_0^2 = 0\right) \left(\frac{k_x^2 + k_y^2}{n_e^2} + \frac{k_z^2}{n_0^2} - k_0^2 \right) = 0 \qquad (8)$$

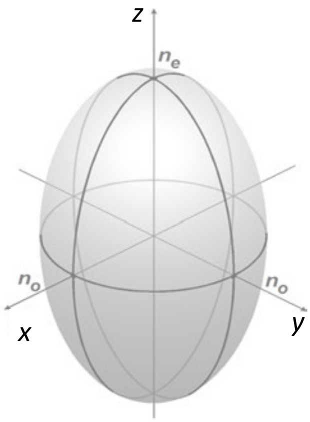
which has two solutions:

- $\frac{k^2}{n_0^2} = k_0^2 \quad \longrightarrow \quad \boldsymbol{k}(\omega)$ lies on a sphere of radius $n_0 k_0 = n_0 \frac{\omega}{c}$ for waves polarized in the x, y plane (ordinary waves)
- $\frac{k_x^2 + k_y^2}{n_e^2} + \frac{k_z^2}{n_0^2} = k_0^2 \quad \longrightarrow \quad \boldsymbol{k}(\omega)$ lies on an ellipsoid of principal semi-axes
  - n_0 along k_z (wave propagating exactly along z must be polarized in the x, y plane)
  - n_e along k_x and k_y (extraordinary waves propagating along x or y are polarized exactly along z).

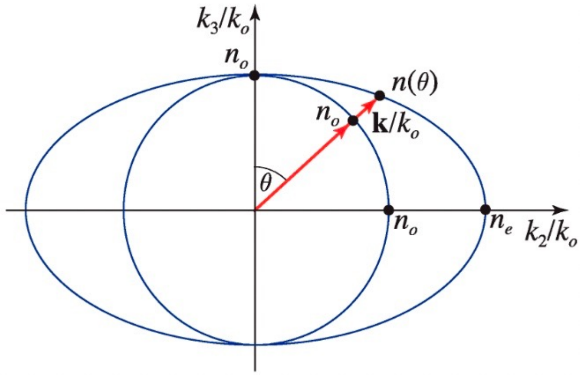
III. Dispersion relation in a uniaxial crystal

$$(k^2 - n_o^2 k_o^2)\left(\frac{k_1^2 + k_2^2}{n_e^2} + \frac{k_3^2}{n_o^2} - k_o^2\right) = 0$$





Index ellipsoid

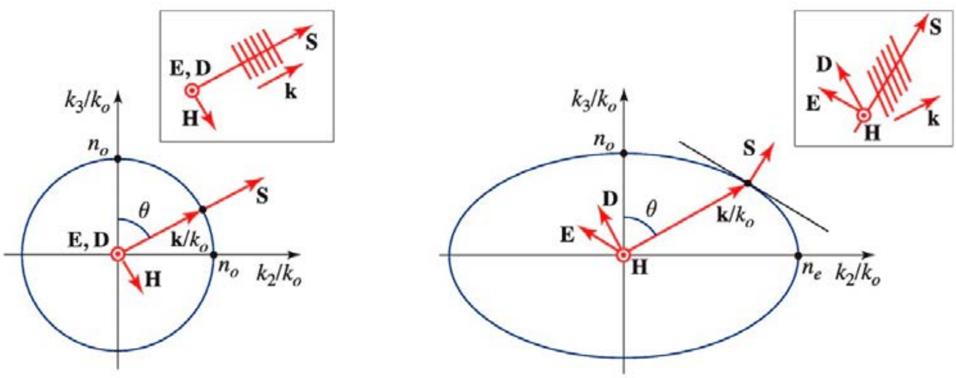


Dispersion ellipsoid

 Saleh & Teich, Ch. 6.3

III. Double refraction (birefringence)

It is possible to show that the Poynting vector and the Ray propagation direction are normal to the *k* surface for the corresponding normal mode polarization



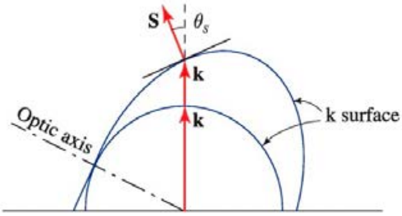
(a) Ordinary (b) Extraordinary

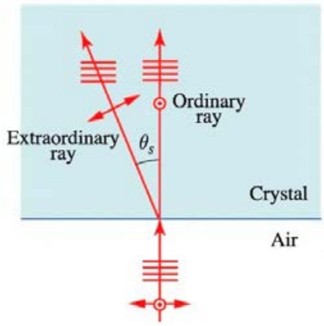
Saleh & Teich, Ch. 6.3

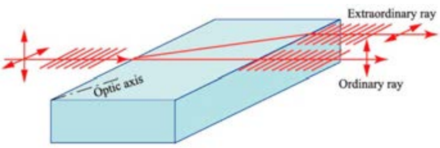
$\int \dots d\omega$
↑
- Temporal wave packets = pulse → group velocity $|\vec{v}| = \frac{\partial \omega}{\partial k}$
- Spatial wave packet = ray (beam) → ——— $\vec{v} = \vec{\nabla}_{\vec{k}}\, \omega(\vec{k})$ // $\vec{S}$

$\int \dots dk$

III. Birefringence and beam walk-off







Saleh & Teich, Ch. 6.3

For calcite the beam displacement can be up to 10% of the thickness

III. Applications of birefringent crystals

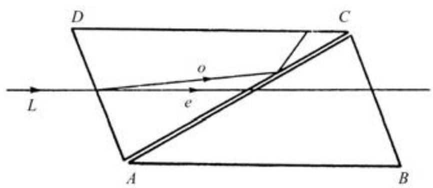
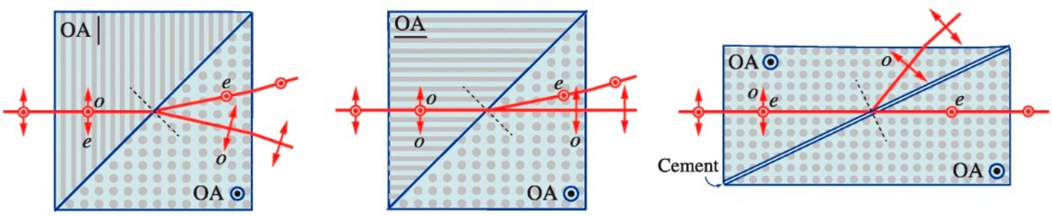


Fig. 15.17 The Nicol prism.



(*a*) Wollaston prism (*b*) Rochon prism (*c*) Glan-Thompson prism

Cemented uniaxial prism with propagation along o direction, with prism where propagation is along the o direction but with the other two axes rotated, so that both polarization components are refracted, almost symmetrically.

Cemented uniaxial prism with propagation along e direction, (essentially isotropic) with prism where propagation is along the o direction, so that one polarization component is refracted.

Cemented prism with propagation along the ordinary axis, based on total internal reflection (and a different refractive index for both polarizations). Air spaced prism gives a larger angular acceptance. Typical extinction ratios are 10^5 – 10^7

Born & Wolf, Ch. XV; Saleh & Teich, Ch. 6.3