6.7 Propagation of Light in Crystals

- The polarization produced in a crystal by an electric field is NOT just a simple scalar constant times the field, but varies in a manner that depends on the direction of the applied field in relation to the crystal lattice (or axis).

- The speed of the propagation of the light wave in a crystal is a function of the propagation direction and the polarization of the light.

* In general, two possible values of the phase velocity for given direction of propagation, associated with mutually orthogonal polarizations of the light waves.

→ Except for crystals with cubic symmetry, others are *doubly refracting* or *birefringent*.

- The dependence of $P$ on $E$; tensor relation

$$
\begin{pmatrix}
P_x \\
P_y \\
P_z
\end{pmatrix} = \varepsilon_0 \begin{pmatrix}
x_{11} & x_{12} & x_{13} \\
x_{21} & x_{22} & x_{23} \\
x_{31} & x_{32} & x_{33}
\end{pmatrix}
\begin{pmatrix}
E_x \\
E_y \\
E_z
\end{pmatrix}; \quad P = \varepsilon_0 \chi E \text{ with the susceptibility tensor } \chi = \begin{pmatrix}
x_{11} & x_{12} & x_{13} \\
x_{21} & x_{22} & x_{23} \\
x_{31} & x_{32} & x_{33}
\end{pmatrix}
$$

The corresponding displacement $D = \varepsilon_0 (1 + \chi) E = \varepsilon E$ where $\varepsilon = \begin{pmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{pmatrix}$

and $\varepsilon$ = the dielectric tensor.
- For ordinary non-absorbing crystals, $\chi$ is symmetric so there always exists a set of coordinate axes, called *principal axes*, such that $\chi$ assumes the diagonal form

$$\chi = \begin{pmatrix}
\chi_{11} & 0 & 0 \\
0 & \chi_{22} & 0 \\
0 & 0 & \chi_{33}
\end{pmatrix}$$

and the principal dielectric constants $K_{11} = 1 + \chi_{11} \ldots \ldots$, etc.

- The general wave equation:

$$\nabla \times (\nabla \times E) + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = - \frac{1}{c^2} \chi \frac{\partial^2 E}{\partial t^2}$$

Using the usual form of $e^{i(k \cdot r - \omega t)}$, $k \times (k \times E) + \frac{\omega^2}{c^2} E = - \frac{\omega^2}{c^2} \chi E$ or

$$\begin{align*}
-k_y^2 - k_z^2 + \frac{\omega^2}{c^2} E_x + k_x k_y E_y + k_x k_z E_z &= -\frac{\omega^2}{c^2} \chi_{11} E_x \\
k_y k_x E_x + \left( -k_x^2 - k_z^2 + \frac{\omega^2}{c^2} \right) E_y + k_y k_z E_z &= -\frac{\omega^2}{c^2} \chi_{22} E_y \\
k_z k_x E_x + k_z k_y E_y + \left( -k_x^2 - k_y^2 + \frac{\omega^2}{c^2} \right) E_z &= -\frac{\omega^2}{c^2} \chi_{33} E_z
\end{align*}$$
- Suppose that we have a particular case of a wave propagating in the direction of one of principal axes, say the $x$-axis. In the case $k_x = k, k_y = k_z = 0$, then the three equations are

\[
\frac{\omega^2}{c^2} E_x = -\frac{\omega^2}{c^2} \chi_{11} E_x, \quad \left( -k^2 + \frac{\omega^2}{c^2} \right) E_y = -\frac{\omega^2}{c^2} \chi_{22} E_y, \quad \left( -k^2 + \frac{\omega^2}{c^2} \right) E_z = -\frac{\omega^2}{c^2} \chi_{33} E_z
\]

$\rightarrow E_x = 0$ because neither $\omega$ nor $\chi_{11}$ is zero; The $E$ field is transverse to the $x$-axis which is the direction of propagation.

If $E_y \neq 0$, then $k = \frac{\omega}{c} \sqrt{1 + \chi_{22}} = \frac{\omega}{c} \sqrt{K_{22}}$.

If $E_z \neq 0$, then $k = \frac{\omega}{c} \sqrt{1 + \chi_{33}} = \frac{\omega}{c} \sqrt{K_{33}}$.

Now $\frac{\omega}{k}$ is the phase velocity of the wave, and thus we have two possible phase velocities, namely, $c/\sqrt{K_{22}}$ if the $E$ vector points in the $y$ direction and $c/\sqrt{K_{33}}$ if the $E$ vector in the $z$ direction.

- More generally, for any direction of the propagation vector $k$, there are two possible values of the magnitude $k$ and hence two possible values of the phase velocity.
Let three principal indices of refraction \( n_1, n_2, n_3 \) be

\[
n_1 = \sqrt{1 + \chi_{11}}, \quad n_2 = \sqrt{1 + \chi_{22}}, \quad n_3 = \sqrt{1 + \chi_{33}}
\]

- For a nontrivial solution for \( E_x, E_y, E_z \),

\[
\begin{vmatrix}
(n_1\omega/c)^2 - k_y^2 - k_z^2 & k_xk_y & k_xk_z \\
-k_yk_x & (n_2\omega/c)^2 - k_x^2 - k_z^2 & k_yk_z \\
k_zk_x & k_zk_y & (n_3\omega/c^2) - k_x^2 - k_y^2
\end{vmatrix} = 0
\]

This equation is represented by a 3-dimensional surface in \( k \) space (see Fig. 6.8).

- Consider any one of the coordinate planes, say the \( xy \) plane. In this plane \( k_z = 0 \) and the determinant reduces to the product of the two factors;

\[
\left( \frac{n_3\omega}{c} \right)^2 - k_x^2 - k_y^2 \right) \left( \left( \frac{n_1\omega}{c} \right)^2 - k_y^2 \right) \left( \left( \frac{n_2\omega}{c} \right)^2 - k_x^2 \right) - k_x^2k_y^2 = 0
\]

Then, \( k_x^2 + k_y^2 = \left( \frac{n_3\omega}{c} \right)^2 \), \( \frac{k_x^2}{(n_2\omega/c)^2} + \frac{k_y^2}{(n_1\omega/c)^2} = 1 \) (circle and ellipse)
The intercept of the $k$ surface with each coordinate plane; one circle and one ellipse.
- The complete $k$ surface is *double*; it consists of an inner sheet and an outer sheet.

  → For any given direction of $k$, there are two possible values for the magnitude $k$.

  The two phase velocities correspond to **two mutually orthogonal direction of polarization**.

- When unpolarized light, or light of arbitrary polarization propagates through a crystal, it can be considered to consists of two independent waves that are polarized orthogonally with respect to each other and travelling with different phase velocities.

  The point (like P in Fig. 6.8) at which the inner and outer sheets touch defines an **optic axis** of the crystal (uniaxial and biaxial).

  If all three indices are equal, then the $k$ space degenerates to a single sphere and the crystal is not doubly refracting at all but is **optically isotropic**.
- Ordinary index $n_o$ for $\chi_{11} = \chi_{22}$ and extraordinary index $n_e$ for $\chi_{33}$

<table>
<thead>
<tr>
<th>Isotropic</th>
<th>$\chi = \begin{bmatrix} a &amp; 0 &amp; 0 \ 0 &amp; a &amp; 0 \ 0 &amp; 0 &amp; a \end{bmatrix}$</th>
<th>$\chi_{11} = \chi_{22} = \chi_{33} = a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>cubic</td>
<td>$n = \sqrt{1 + a}$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Uniaxial</th>
<th>$\chi = \begin{bmatrix} a &amp; 0 &amp; 0 \ 0 &amp; a &amp; 0 \ 0 &amp; 0 &amp; b \end{bmatrix}$</th>
<th>$\chi_{11} = \chi_{22} = a$, $\chi_{33} = b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>trigonal</td>
<td>$n_0 = \sqrt{1 + a}$</td>
<td></td>
</tr>
<tr>
<td>tetragonal</td>
<td>$n_e = \sqrt{1 + b}$</td>
<td></td>
</tr>
<tr>
<td>hexagonal</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Biaxial</th>
<th>$\chi = \begin{bmatrix} a &amp; 0 &amp; 0 \ 0 &amp; b &amp; 0 \ 0 &amp; 0 &amp; c \end{bmatrix}$</th>
<th>$\chi_{11} = a$, $\chi_{22} = b$, $\chi_{33} = c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>triclinic</td>
<td>$n_1 = \sqrt{1 + a}$</td>
<td></td>
</tr>
<tr>
<td>monoclinic</td>
<td>$n_2 = \sqrt{1 + b}$</td>
<td></td>
</tr>
<tr>
<td>orthorhombic</td>
<td>$n_3 = \sqrt{1 + c}$</td>
<td></td>
</tr>
</tbody>
</table>

- Phase velocity surface; $k = \frac{\omega}{c}$; $k = v \frac{\omega}{v^2}$
- The Poynting Vector and the Ray Velocity

*Although the propagation vector $k$ defines the direction of planes of constant phase for light waves in a crystal, the actual direction of energy flow $E \times H$ is NOT in the same direction (wavefront $k$); In general, $E$ and $k$ are NOT mutually perpendicular in anisotropic media.

The magnetic field $H$ is perpendicular to both $E$ and $k$ because of $k \times E = \mu_0 \omega H$. The three vectors $E$, $k$, and $S = E \times H$ are all perpendicular to $H$.

Ray velocity $u = \frac{v}{\cos \theta}$; When the propagation direction is along one of the principal axes $(\theta = 0)$, $u = v$ ($S$ and $k$ have the same direction).

- The Ray Velocity Surface

In terms of the displacement vector $D = \epsilon_0 (1 + \chi) E$, for plane harmonic waves,

\[ k \times (k \times E) = -\frac{\omega^2}{c^2 \epsilon_0} D \quad ; \quad D \text{ is perpendicular to } k. \]

\[ k(k \cdot E) - k^2 E = -\frac{\omega^2}{c^2 \epsilon_0} D \quad \rightarrow \quad k^2 E \cdot D = \frac{\omega^2}{c^2 \epsilon_0} D \cdot D \quad \text{since } k \cdot D = 0, \]

In the $xy$ plane ($u_z = 0$), $u_x^2 + u_y^2 = c^2/n_3^2$, $n_x^2 u_x^2 + n_y^2 u_y^2 = c^2$; circle and ellipse.
6.8 Double Refraction at a Boundary

- The law of refraction; $k_0 \cdot r = k \cdot r$ (at boundary).
  Two possible propagation directions for given propagation in a crystal:

Figure 6.13. Wave vectors for double refraction at the boundary of a crystal.
For the two refracted waves, \( k_0 \sin \theta = k_1 \sin \phi_1, \quad k_0 \sin \theta = k_2 \sin \phi_2 \). 

In the case of uniaxial crystals, the \( k \) surface for the ordinary wave is a **sphere**. 
→ The corresponding \( k \) is constant for all directions of the wave and Snell's law is obeyed, \( \frac{\sin \theta}{\sin \phi} = n_0 \). 

The \( k \) space for the extraordinary wave is a **spheroid**, and the Snell's law is NOT valid.

- **Polarizing Prisms** (see Fig. 6.16)

Let a wave be incident on a plane boundary from the inside of a uniaxial crystal and consider the case in which the optic axis is perpendicular to the plane of incidence.

In this case, the Snell's law holds for both the ordinary wave and the extraordinary wave.

\[ n_0 \sin \phi_o = \sin \theta, \quad n_e \sin \phi_e = \sin \theta \quad \text{assuming that} \quad n_{air} = 1. \]

The \( E \) vector of the ordinary wave is perpendicular to the direction of the optic axis, and that of the extraordinary wave is parallel to the optic axis.

For \( n_e < \frac{1}{\sin \theta} < n_0 \), **total internal reflection** for the o-wave but not for the e-wave.
Figure 6.15. (a) Separation of the extraordinary and ordinary rays at the boundary of a crystal in the case of internal refraction; (b) construction of the Glan polarizing prism; (c) the Nicol prism.
6.9 Optical Activity

- Optical Activity: the ability to rotate the plane of polarization of light.

  Sodium chlorate, cinnabar, sugar, crystalline quartz

*Specific rotary power: the amount of rotation per unit length of travel

*dextro*-rotatory (right-handed) and *levo*-rotatory (left-handed) in the propagation direction.
- Let $n_R$ and $n_L$ denote the indices of the medium for the right and left circularly polarized light;

\[ \begin{pmatrix} 1 \\ -i \end{pmatrix} e^{i(k_{R}z - \omega t)} \quad \text{and} \quad \begin{pmatrix} 1 \\ i \end{pmatrix} e^{i(k_{L}z - \omega t)} , \]

Consider a beam of linearly polarized light travels a distance $l$ through the medium (the initial polarization is along the $x$ axis).

The initial Jones vector can be separated into right and left circular components as

\[ \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 \\ -i \end{pmatrix} + \frac{1}{2} \begin{pmatrix} 1 \\ i \end{pmatrix} . \]

The complex amplitude of the wave after travelling a distance $l$ through the medium is

\[ \frac{1}{2} \begin{pmatrix} 1 \\ -i \end{pmatrix} e^{i k_{R}l} + \frac{1}{2} \begin{pmatrix} 1 \\ i \end{pmatrix} e^{i k_{L}l} = \frac{1}{2} e^{i (k_{R} + k_{L})l/2} \begin{pmatrix} 1 \\ -i \end{pmatrix} e^{i (k_{R} - k_{L})l/2} + \begin{pmatrix} 1 \\ i \end{pmatrix} e^{-i (k_{R} - k_{L})l/2} \]

With the quantities $\psi = (k_{R} + k_{L})l/2$, $\theta = (k_{R} - k_{L})l/2$,

\[ e^{i \psi} \left\{ \frac{1}{2} \begin{pmatrix} 1 \\ -i \end{pmatrix} e^{i \theta} + \frac{1}{2} \begin{pmatrix} 1 \\ i \end{pmatrix} e^{-i \theta} \right\} = e^{i \psi} \left\{ \frac{(e^{i \theta} + e^{-i \theta})}{2} / i (e^{i \theta} - e^{-i \theta}) / 2 \right\} = e^{i \psi} \begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix} \rightarrow \text{polarization rotation} . \]
Since $\theta = (n_R - n_L) \frac{\omega l}{2c} = (n_R - n_L) \frac{\pi l}{\lambda}$, the specific rotary power $\delta = (n_R - n_L) \frac{\pi}{\lambda}$.

- Susceptibility Tensor of an Optically Active Medium

$$\chi = \begin{pmatrix} \chi_{11} & i\chi_{12} & 0 \\ -i\chi_{12} & \chi_{11} & 0 \\ 0 & 0 & \chi_{33} \end{pmatrix}$$

[Proof]

In the case of a wave propagating in the $z$ direction ($E_z = 0$), solving Maxwell's equations to have nontrivial solutions, $k = \frac{\omega}{c} \sqrt{1 + \chi_{11} \mp \chi_{22}}$. Substituting this back into the one of the Maxwell eqn, $E_x = \pm i E_y$.

$$n_R = \sqrt{1 + \chi_{11} + \chi_{12}} \quad \text{and} \quad n_L = \sqrt{1 + \chi_{11} - \chi_{12}} \quad \rightarrow \quad n_r - n_L \approx \frac{\chi_{12}}{\sqrt{1 + \chi_{11}}} = \frac{\chi_{12}}{n_0}$$

Then, the specific rotary power $\delta = \frac{\chi_{12} \pi}{n_0 \lambda}$.
6.10 Faraday Rotation in Solids

- A rotation of the plane of polarization under a magnetic field; the presence of the magnetic field causes the dielectric to become optically active (Faraday Effect, 1845).

The amount of polarization rotation $\theta = VBl$ with $V = \text{the Verdet constant}.$

- The equation of motion of the bound electrons under a static magnetic field $B$ and an oscillating electric field $E$ of the optical wave.

$$m\frac{d^2r}{dt^2} + Kr = -eE - e\left(\frac{dr}{dt}\right) \times B \rightarrow -m\omega^2r + Kr = -eE + i\omega r \times B \text{ for harmonic waves.}$$

Let $P = -Ne\omega r$; \(-m\omega^2 + K)P = Ne^2E + i\omega r P \times B$

In a form of $\chi = \begin{pmatrix} \chi_{11} & i\chi_{12} & 0 \\ -i\chi_{12} & \chi_{11} & 0 \\ 0 & 0 & \chi_{33} \end{pmatrix}$, $\chi_{11} = \frac{Ne^2}{m\epsilon_o} \left[ \frac{\omega^2 - \omega^2}{\left(\omega_0^2 - \omega^2\right)^2 - \omega^2 \omega_c^2} \right]$

$\chi_{33} = \frac{Ne^2}{m\epsilon_o} \left[ \frac{1}{\omega^2 - \omega^2} \right]$

$\chi_{12} = \frac{Ne^2}{m\epsilon_o} \left[ \frac{\omega\omega_c}{\left(\omega_0^2 - \omega^2\right)^2 - \omega^2 \omega_c^2} \right]$
Here, $\omega_0 = \sqrt{K/m}$ (resonance freq) and $\omega_c = \sqrt{eB/m}$ (cyclotron freq).

The specific rotary power $\delta = \frac{\pi Ne^2}{\lambda m\epsilon_0} \left[ \frac{\omega\omega_c}{(\omega_0^2 - \omega^2)^2} \right] = \frac{\pi Ne^3}{\lambda m^2\epsilon_0} \left[ \frac{\omega B}{(\omega_0^2 - \omega^2)^2} \right]

for $\omega\omega_c \ll |\omega_0^2 - \omega^2|$

6.11 Other Magneto-optic and Electro-optic Effects

- **The Kerr Electro-optic Effect**

  Double refraction of an optically isotropic substance under a strong electric field (Kerr, 1875)

  $$n_\parallel - n_\perp = KE^2\lambda_0$$

- **The Cotton-Mouton Effect**

  The magnetic analog of the Kerr effect ; $\Delta n \propto B^2$
- The Pockels Effect

Changes of indices of refraction of a birefringent crystal (ADP, KDP, etc) by an electric field; light shutters and modulators

Figure 6.21. Setup for using a Pockels cell light modulator. The quarter-wave plate is used to provide “optical bias.”
6.12 Nonlinear Optics

- When the radiation field of a light wave is comparable to the atomic fields \( \approx 10^8 \text{ V/cm} \) that bind the electrons the atoms (beyond a small perturbation on the outer or valence electrons), nonlinear optical effects such as optical harmonic generation, optical rectification, etc, are observed.

**Figure 6.24.** (a) Graphs of electric field and polarization as functions of time for the nonlinear case; (b) resolution of the polarization into the fundamental and the second harmonic. (There will also be a dc term that is not shown.)
- In an isotropic medium, the polarization $P = \varepsilon_0 (\chi E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \ldots)$.

Under the field of $E_0 e^{-i\omega t}$, $P = \varepsilon_0 (\chi E_0 e^{-i\omega t} + \chi^{(2)} E_0^2 e^{-2i\omega t} + \chi^{(3)} E_0^3 e^{-3i\omega t} + \ldots \ldots)$

- In the case of anisotropic crystals, $P$ and $E$ are NOT necessarily parallel.

\[ P = \varepsilon_0 (\chi E + \chi^{(2)} E E + \chi^{(3)} E E E + \ldots \ldots) \] where $\chi$ = a tensor.

The second harmonic polarization $P_i(2\omega) = \sum_j \sum_k \chi^{(2)}_{ijk} E_j E_k$. 