Lecture 7

Second-order $\chi^{(2)}$ nonlinear optical materials; crystal classes and their symmetries and nonlinear optical tensors; electrooptic effect;

Anisotropic linear media

Linear susceptibility is a tensor

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E}.$$ 

$$P_i = \varepsilon_0 \sum_{j=x,y,z} \chi_{ij} E_j$$

or

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

In an anisotropic medium, such as a crystal, the polarisation field $P$ is not necessarily aligned with the electric field of the light $E$. In a physical picture, this can be thought of as the dipoles induced in the medium by the electric field having certain preferred directions, related to the physical structure of the crystal.

In nonmagnetic and transparent materials, $\chi_{ij} = \chi_{ji}$ i.e. the $\chi$ tensor is real and symmetric.

It is possible to diagonalise the tensor by choosing the appropriate coordinate axes, leaving only $\chi_{xx}$, $\chi_{yy}$ and $\chi_{zz}$. This gives:

$$\begin{align*}
P_x &= \varepsilon_0 \chi_{xx} E_x \\
P_y &= \varepsilon_0 \chi_{yy} E_y \\
P_z &= \varepsilon_0 \chi_{zz} E_z
\end{align*}$$
Anisotropic linear media

The refractive index is: \[ n = \sqrt{1 + \chi} \]

hence
\[ n_{xx} = \sqrt{1 + \chi_{xx}} \]
\[ n_{yy} = \sqrt{1 + \chi_{yy}} \]
\[ n_{zz} = \sqrt{1 + \chi_{zz}} \]

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 \( \chi_{xx} = \chi_{yy} \neq \chi_{zz} \), the crystal is known as **uniaxial**.

If \( \chi_{xx} \neq \chi_{yy} \neq \chi_{zz} \) the crystal is called **biaxial**.

will come back to this in Lecture 8

### Anisotropic linear media

#### Uniaxial crystals

<table>
<thead>
<tr>
<th>Material</th>
<th>Crystal system</th>
<th>( n_\parallel )</th>
<th>( n_\perp )</th>
<th>( \Delta n )</th>
</tr>
</thead>
<tbody>
<tr>
<td>barium borate BaB(_2)O(_4)</td>
<td>Trigonal</td>
<td>1.6776</td>
<td>1.5534</td>
<td>-0.1242</td>
</tr>
<tr>
<td>beryl Be(_3)Al(_2)SiO(_6)</td>
<td>Hexagonal</td>
<td>1.602</td>
<td>1.557</td>
<td>-0.045</td>
</tr>
<tr>
<td>calcite CaCO(_3)</td>
<td>Trigonal</td>
<td>1.658</td>
<td>1.486</td>
<td>-0.172</td>
</tr>
<tr>
<td>ice H(_2)O</td>
<td>Hexagonal</td>
<td>1.309</td>
<td>1.313</td>
<td>+0.004</td>
</tr>
<tr>
<td>lithium niobate LiNbO(_3)</td>
<td>Trigonal</td>
<td>2.272</td>
<td>2.187</td>
<td>-0.085</td>
</tr>
<tr>
<td>magnesium fluoride MgF(_2)</td>
<td>Tetragonal</td>
<td>1.380</td>
<td>1.385</td>
<td>+0.006</td>
</tr>
<tr>
<td>quartz SiO(_2)</td>
<td>Trigonal</td>
<td>1.544</td>
<td>1.553</td>
<td>+0.009</td>
</tr>
<tr>
<td>ruby Al(_2)O(_3)</td>
<td>Trigonal</td>
<td>1.770</td>
<td>1.762</td>
<td>-0.008</td>
</tr>
<tr>
<td>rutile TiO(_2)</td>
<td>Tetragonal</td>
<td>2.616</td>
<td>2.903</td>
<td>+0.287</td>
</tr>
<tr>
<td>sapphire Al(_2)O(_3)</td>
<td>Trigonal</td>
<td>1.768</td>
<td>1.760</td>
<td>-0.008</td>
</tr>
<tr>
<td>silicon carbide SiC</td>
<td>Hexagonal</td>
<td>2.647</td>
<td>2.693</td>
<td>+0.046</td>
</tr>
<tr>
<td>tourmaline (complex silicate)</td>
<td>Trigonal</td>
<td>1.669</td>
<td>1.638</td>
<td>-0.031</td>
</tr>
<tr>
<td>zircon, high ZrSiO(_4)</td>
<td>Tetragonal</td>
<td>1.980</td>
<td>2.015</td>
<td>+0.055</td>
</tr>
<tr>
<td>zircon, low ZrSiO(_4)</td>
<td>Tetragonal</td>
<td>1.920</td>
<td>1.987</td>
<td>+0.047</td>
</tr>
</tbody>
</table>

#### Biaxial crystals

<table>
<thead>
<tr>
<th>Material</th>
<th>Crystal system</th>
<th>( n_\parallel )</th>
<th>( n_\perp )</th>
<th>( n_\gamma )</th>
</tr>
</thead>
<tbody>
<tr>
<td>borax Na(_2)(H(_2)O)(_4)-8H(_2)O</td>
<td>Monoclinic</td>
<td>1.447</td>
<td>1.469</td>
<td>1.472</td>
</tr>
<tr>
<td>epsom salt MgSO(_4)-7H(_2)O</td>
<td>Monoclinic</td>
<td>1.433</td>
<td>1.455</td>
<td>1.461</td>
</tr>
<tr>
<td>mica, biotite K(Mg,Fe(_2))(_2)(Al(_2)SiO(_6))(_2)(F,OH(_2))</td>
<td>Monoclinic</td>
<td>1.595</td>
<td>1.640</td>
<td>1.640</td>
</tr>
<tr>
<td>mica, muscovite KAl(_2)(Al(_2)SiO(_6))(_2)(F,OH(_2))</td>
<td>Monoclinic</td>
<td>1.563</td>
<td>1.596</td>
<td>1.601</td>
</tr>
<tr>
<td>olivine (Mg,Fe(_2))(_2)SiO(_4)</td>
<td>Orthorhombic</td>
<td>1.640</td>
<td>1.660</td>
<td>1.680</td>
</tr>
<tr>
<td>perovskite CaTiO(_3)</td>
<td>Orthorhombic</td>
<td>2.300</td>
<td>2.340</td>
<td>2.380</td>
</tr>
<tr>
<td>topaz Al(_2)(SiO(_4))(F,OH(_2))</td>
<td>Orthorhombic</td>
<td>1.618</td>
<td>1.620</td>
<td>1.627</td>
</tr>
<tr>
<td>ulexite NaCa(_2)B(_2)O(_6)(OH(_2))(_4)-5H(_2)O</td>
<td>Triclinic</td>
<td>1.490</td>
<td>1.510</td>
<td>1.520</td>
</tr>
</tbody>
</table>
Nonlinear Susceptibility Tensor

Nonlinear susceptibility is a 3rd rank tensor \( \chi^{(2)}_{ijk} \)

When all of the optical frequencies are detuned from the resonance frequencies of the optical medium
the nonlinear susceptibility tensor \( \chi^{(2)}_{ijk} \) has full permutation symmetry:

\[
\chi_{ijk} = \chi_{kji} = \chi_{jki} = \chi_{ikj}
\]

... and does not actually depend of frequencies that participate, so instead of writing \( \chi^{(2)}_{ijk}(\omega_3 = \omega_1 + \omega_2, \omega_1, \omega_2) \) we write simply \( \chi^{(2)}_{ijk} \)

Full permutation symmetry can be deduced from a consideration of the field energy density within a nonlinear medium
(see Boyd or Stegeman books).

And also from quantum mechanics!

Consider mutual interaction of three waves at \( \omega_1, \omega_2, \) and \( \omega_3 = \omega_1 + \omega_2 \)

Assume \( E \) is the total field vector \( E = E_{\omega_1} + E_{\omega_2} + E_{\omega_3} \)

Or in vector components \( E_i = E_{i,\omega_1} + E_{i,\omega_2} + E_{i,\omega_3} \quad i = 1,2,3 \quad (= x, y, z) \)

then

\[
P_i = \varepsilon_0 \sum_{j,k} \chi_{ijk} E_j E_k \quad i,j,k = 1,2,3 \quad (= x, y, z)
\]

For example, \( x \) – component of the nonlinear polarization \( P \) is:

\[
x \quad P_x = \varepsilon_0 \sum_{j,k} \chi_{1jk} E_j E_k = \varepsilon_0 \left\{ \chi_{111}E_1^2 + \chi_{112}E_1E_2 + \chi_{113}E_1E_3 + \chi_{121}E_2^2 + \chi_{122}E_2E_3 + \chi_{131}E_3^2 + \chi_{123}E_2E_3 + \chi_{132}E_3E_2 + \chi_{133}E_3E_3 \right\}
\]

\[
y \quad P_y = \varepsilon_0 \sum_{j,k} \chi_{2jk} E_j E_k = ...
\]

\[
z \quad P_z = \varepsilon_0 \sum_{j,k} \chi_{3jk} E_j E_k = ...
\]
Nonlinear Susceptibility Tensor

Now introduce the tensor:

\[ d_{ijl} = \frac{1}{2} \chi^{(2)}_{ijkl} \]

(historical convention !)

and write:

\[ P_1 = 2 \varepsilon_0 \left( d_{111} E_1 E_1 + d_{122} E_2 E_2 + d_{133} E_3 E_3 + 2d_{123} E_2 E_3 + 2d_{133} E_1 E_3 + 2d_{112} E_1 E_2 \right) \]

\[ P_2 = 2 \varepsilon_0 \left( d_{211} E_1 E_1 + d_{222} E_2 E_2 + d_{233} E_3 E_3 + 2d_{223} E_2 E_3 + 2d_{233} E_1 E_3 + 2d_{212} E_1 E_2 \right) \]

\[ P_3 = 2 \varepsilon_0 \left( d_{311} E_1 E_1 + d_{322} E_2 E_2 + d_{333} E_3 E_3 + 2d_{323} E_2 E_3 + 2d_{333} E_1 E_3 + 2d_{312} E_1 E_2 \right) \]

now reduced to only 18 components

Reduce 3D tensor to 2D matrix

Matrix with 6x3 components

\[ d_{jl} = \begin{bmatrix}
  d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
  d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
  d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \\
\end{bmatrix} \]

Second-order susceptibility described using contracted notation

the way to find nonlinear polarizations in 3D case using contracted notation

\[
\begin{bmatrix}
  P_x \\
  P_y \\
  P_z \\
\end{bmatrix} = 2 \varepsilon_0 \begin{bmatrix}
  d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
  d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
  d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \\
\end{bmatrix} \begin{bmatrix}
  E_x^2 \\
  E_y^2 \\
  E_z^2 \\
  \end{bmatrix} + \begin{bmatrix}
  2E_y E_x \\
  2E_x E_y \\
  2E_x E_z \\
  \end{bmatrix}
\]
Nonlinear Susceptibility Tensor

By further applying Kleinman symmetry, we find that $d_{ii}$ matrix has only 10 independent elements:

$$d_{ii} = \begin{bmatrix}
  d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
  d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
  d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{bmatrix}.$$
Spatial symmetries of crystals further reduce the amount of independent tensor elements.

'Cubic' crystals e.g. GaAs, GaP, InAs, ZnS, ZnSe - are optically isotropic but nonlinear!
Nonlinear Susceptibility Tensor

point group mm2  KTP (KTiO$_2$PO$_4$) crystal

This crystal class is invariant under 180° rotations around z-axis and mirror images on the planes m1 and m2, that contain the rotation axis.

tensor elements transform just like the coordinates

\[
\begin{align*}
m_1 &: (x, y, z) \rightarrow (-x, y, z), \\
m_2 &: (x, y, z) \rightarrow (x, -y, z), \\
2 &: (x, y, z) \rightarrow (-x, -y, z)
\end{align*}
\]
Nonlinear Susceptibility Tensor

KTP (KTiO$_2$PO$_4$) crystal

\[
\begin{pmatrix}
d_{13} & d_{12} & d_{11} & 0 & d_{15} & 0 \\
d_{16} & d_{20} & d_{24} & d_{14} & 0 & d_{12} \\
d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14}
\end{pmatrix}
\]

Crystal of class 3m (e.g., Lithium Niobate, LiNbO$_3$)

\[
d_{ij} = \begin{pmatrix}
0 & 0 & 0 & 0 & d_{31} & -d_{22} \\
-d_{22} & d_{22} & 0 & (d_{31}) & 0 & 0 \\
(d_{31}) & d_{31} & 0 & 0 & 0 & 0 \\
(d_{33}) & d_{33} & 0 & 0 & 0 & 0
\end{pmatrix}
\]

Only $d_{22}$, $d_{31}$, and $d_{33}$

Class: 3m

\[
\rightarrow \begin{array}{c}
\text{LiNbO}_3, \text{LiTaO}_2 \\
\text{BaB}_2\text{O}_2 (\text{BBO})
\end{array}
\]

Other crystals of this class
Physical origin of off-diagonal elements in $d_{ijk}$ tensor

after Stegeman NLO book

$E$-field applied in $xy$

active electron

electron motion

GaAs crystal

As

Ga

GaAs crystal

As

As

As

As

motion in $xy$

motion in $z$

Physical origin of off-diagonal elements in $d_{ijk}$ tensor
Nonlinear Susceptibility Tensor

GaAs

Crystal of class 4\textit{3}m (e.g. Gallium Arsenide, \textbf{GaAs})

only \textit{d}_{14}

\[
\mathbf{\hat{P}}^{(2)} = 2\varepsilon_0 \begin{pmatrix}
0 & 0 & 0 & \textit{d}_{14} & 0 & 0 \\
0 & 0 & 0 & 0 & \textit{d}_{14} & 0 \\
0 & 0 & 0 & 0 & 0 & \textit{d}_{14}
\end{pmatrix}
\begin{pmatrix}
\hat{E}_x \hat{E}_x \\
\hat{E}_y \hat{E}_y \\
\hat{E}_z \hat{E}_z \\
2\hat{E}_y \hat{E}_z \\
2\hat{E}_z \hat{E}_x \\
2\hat{E}_x \hat{E}_y
\end{pmatrix}
\]
## Nonlinear Susceptibility Tensor

How to calculate effective nonlinearity?

\[
\begin{bmatrix}
P_x \\
P_y \\
P_z
\end{bmatrix}
= 2\varepsilon_0
\begin{bmatrix}
d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{bmatrix}
\begin{bmatrix}
E_x^2 \\
E_y^2 \\
E_z^2 \\
E_x E_y \\
E_y E_z \\
E_z E_x
\end{bmatrix}
\]

1) LiNbO\(_3\) \(d_{33} \neq 0\)

Simple case: SFG \(\omega_3 = \omega_1 + \omega_2\)
all waves \((\omega_1, \omega_2, \omega_3)\) are polarized along \(z\)-axis
input fields
\(E_x(t) = E_{1,x} e^{i\omega_1 t} + E_{2,x} e^{i\omega_2 t}\)

Need to find \(z\)-component of \(P_x^{(NL)} = P(\omega_3)e^{i\omega_3 t}\)

\(P_x^{(NL)}(t) = \frac{\varepsilon_0 d_{33} E_x^2}{4} (E_{1,x} e^{i\omega_1 t} + E_{2,x} e^{i\omega_2 t} + c.c.)^2 = \ ...
\)

\[2\varepsilon_0 d_{eff} E_x^2 \]
\(d_{eff} = d_{33}\)

---

## Nonlinear Susceptibility Tensor

For SFG term: \(P(\omega_3) = P(\omega_1 + \omega_2)\) — pick only components with \(\pm(\omega_1 + \omega_2)\)

At \(\omega_3\)

\(P_x^{(NL)}(t) = 2\varepsilon_0 d_{33} \frac{1}{2} (E_{1,x} e^{i(\omega_1 + \omega_2) t} + E_{2,x} e^{i(\omega_1 + \omega_2) t} + c.c.) = \frac{1}{2} (P_x(\omega_3) + c.c.) = \varepsilon_0 d_{33} (E_{1,x} E_{2,x} e^{i(\omega_1 + \omega_2) t} + c.c.)\)

\(P_x(\omega_3) = \varepsilon_0 2 d_{33} (E_{1,x} E_{2,x})\) 
real Fourier component \(P = P_x(\omega_3) \cos(\omega_3 t)\)
Nonlinear Susceptibility Tensor

\[ P(\omega_1 + \omega_2) = 2\epsilon_0 d_{33} E_1 E_2 \]
\[ P(2\omega_1) = \epsilon_0 d_{33} E_1^2 \]
\[ P(2\omega_2) = \epsilon_0 d_{33} E_2^2 \]

same as (5.2)–(5.3) with \( d = \frac{1}{2} \chi^{(2)} \)

once you know \( d_{\text{eff}} \), you can treat fields as scalars

Nonlinear Susceptibility Tensor

2) Effective nonlinearity, GaAs

SFG \( \omega_3 = \omega_1 + \omega_2 \)

Need to find \( P(\omega_3) \)

for input fields \( E(t) = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} \)

\[
E_1 = \frac{1}{\sqrt{2}}(E_{1x} + E_{1y}) e^{i\omega_1 t} \quad E_x = \frac{1}{\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}) \\
E_2 = \frac{1}{\sqrt{2}}(E_{2x} + E_{2y}) e^{i\omega_2 t} \quad E_y = E_x
\]

\[ P_{NL}(t) = P_\omega(t) = \epsilon_0 2d_{14} (2E_1 E_2) = 4\epsilon_0 d_{14} \frac{1}{2}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + \text{c.c.})^2 \]

\[ P_{NL}(t, \omega_3) = \epsilon_0 d_{14} \frac{1}{2}(2E_1 E_2 e^{i(\omega_1 + \omega_2)t} + \text{c.c.}) = \epsilon_0 d_{14} (E_1 E_2 e^{i(\omega_1 + \omega_2)t} + \text{c.c.}) = \epsilon_0 d_{14} \frac{1}{2}(E_1 E_2 e^{i(\omega_1 + \omega_2)t} + \text{c.c.}) \]

for SFG \( P(\omega_1 + \omega_2) = 2\epsilon_0 d_{14} E_1 E_2 \) \( \Rightarrow d_{\text{eff}} = d_{14} \)

Can treat fields as scalars keeping in mind that \( E_1, E_2 \) are in \( xy \) and \( E_3 \) is in \( z \)-direction.
Nonlinear Susceptibility Tensor

Because of the off-diagonal tensor elements, we can generate SFG with the output polarization perpendicular to the input polarizations.

First-order electrooptic effect in anisotropic crystals

The electrooptic effect (Pockels effect) is the change in refractive index of a material induced by the presence of a static (or low-frequency) electric field.

Linear anisotropic medium:

\[ D_i = \varepsilon_0 \sum_j \varepsilon_{ij} E_j \]

\[ \begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}. \]

The dielectric constant is a second-rank tensor.

Dielectric tensor is represented as a diagonal matrix (by a proper choice of the coordinate system).

\[ \begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \varepsilon_{XX} & 0 & 0 \\ 0 & \varepsilon_{YY} & 0 \\ 0 & 0 & \varepsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}. \]

in the principal dielectric axes.
First-order electrooptic effect in anisotropic crystals

The index ellipsoid

\[
\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} = 1
\]

two allowed directions of polarization with two distinct n coefficients

beam k-vector

Uniaxial crystal: \( n_1 = n_2 = n_o; \quad n_3 = n_e \)

Uniaxial crystals: The indicatrix is an ellipsoid of revolution.

For the direction of polarization perpendicular to the optic axis, known as the ordinary direction, the index is independent of the direction of propagation.

For the other direction of polarization, known as the extraordinary direction, the index changes between the value of the ordinary index \( n_o \), when the wave normal is parallel to the optic axis (z) and the extraordinary index \( n_e \), when the wave normal is perpendicular to the optic axis.

The two beams of light so produced are often referred to as o-rays and e-rays, respectively.

When the wave normal is in a direction \( \theta \) to the optic axis, the index is given by:

\[
\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} = 1
\]

\[
\left(\frac{\cos(\theta)}{n_0}\right)^2 + \left(\frac{\sin(\theta)}{n_e}\right)^2 = 1
\]

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

\[
\Rightarrow \quad n(\theta) = \sqrt{n_0^2 \cos^2(\theta) + n_e^2 \sin^2(\theta)}
\]

'positive' crystal
First-order electrooptic effect in anisotropic crystals

Uniaxial crystal: \( n_1 = n_2 = n_o \); \( n_3 = n_e \)

Uniaxial crystals: The indicatrix is an ellipsoid of revolution.

For the direction of polarization perpendicular to the optic axis, known as the ordinary direction, the index is independent of the direction of propagation.

For the other direction of polarization, known as the extraordinary direction, the index changes between the value of the ordinary index \( n_0 \), when the wave normal is parallel to the optic axis (z) and the extraordinary index \( n_e \), when the wave normal is perpendicular to the optic axis.

The two beams of light so produced are often referred to as o-rays and e-rays, respectively.

When the wave normal is in a direction \( \theta \) to the optic axis, the index is given by:

\[
\frac{1}{n(\theta)^2} = \frac{\cos(\theta)^2}{n_0^2} + \frac{\sin(\theta)^2}{n_e^2} \\
\Rightarrow n(\theta) = \frac{n_0 n_e}{\sqrt{n_0^2 \sin^2(\theta) + n_e^2 \cos^2(\theta)}}
\]

First-order electrooptic effect in anisotropic crystals

the index ellipsoid

General case:

\[
\frac{x^2}{n_1^2} + \frac{y^2}{n_2^2} + \frac{z^2}{n_3^2} = 1
\]

General case for the index ellipsoid

\[
\left( \frac{1}{n_1^2} \right) x^2 + \left( \frac{1}{n_2^2} \right) y^2 + \left( \frac{1}{n_3^2} \right) z^2 + 2 \left( \frac{1}{n_1 n_2} \right) x z + 2 \left( \frac{1}{n_1 n_3} \right) x y + 2 \left( \frac{1}{n_2 n_3} \right) y z = 1.
\]
First-order electrooptic effect in anisotropic crystals

The essence of linear electrooptic effect

\[ \Delta \left( \frac{1}{n^2} \right)_i = \sum_j r_{ij} E_j, \]

or

\[
\begin{pmatrix}
\Delta(1/n^2)_{11} \\
\Delta(1/n^2)_{22} \\
\Delta(1/n^2)_{33} \\
\Delta(1/n^2)_{12} \\
\Delta(1/n^2)_{13} \\
\Delta(1/n^2)_{23}
\end{pmatrix}
= 
\begin{pmatrix}
r_{11} & r_{12} & r_{13} \\
r_{21} & r_{22} & r_{23} \\
r_{31} & r_{32} & r_{33} \\
r_{41} & r_{42} & r_{43} \\
r_{51} & r_{52} & r_{53} \\
r_{61} & r_{62} & r_{63}
\end{pmatrix}
\begin{pmatrix}
E_x \\
E_y \\
E_z
\end{pmatrix}
\]

Electrooptic effect and NLO effects are present in the same classes of crystals

Electrooptic modulator, KDP

Apply field along \( z \)-axis

This causes (via \( r_{63} \)) induced index change in \( xy \) plane

\[
n_{yx} = n_0 - \frac{1}{2} r_{63} E_z,
\]

\[
n_{yx} = n_0 + \frac{1}{2} r_{63} E_z.
\]
First-order electrooptic effect in anisotropic crystals

Electrooptic modulator, KDP

Evolution of the vertical polarization originally sent to the modulator

Half-wave voltage ~ kV range

\[ V_{\lambda/2} = \frac{\pi c}{\omega n_0^3 r_{63}} \]

First-order electrooptic effect in anisotropic crystals

Electrooptic modulator, lithium niobate LiNbO₃

NLO tensor

Apply field along x-axis (transverse effect)

This causes (via \( r_{22} \)) induced index change in xy plane

Half-wave voltages are much lower than in KDP
Let us now have another look at this phenomenon. Before we saw that we can generate SFG with the output polarization perpendicular to the input polarizations.

\[ \omega_3 = \omega_1 + \omega_2 \quad \omega_1 \to 0, \quad \omega_3 \to \omega_2 \]

The electrooptic effect can be seen as a frequency-mixing interaction (SFG or DFG) between the incident radiation and an externally applied DC voltage.

General relation between Pockels and NLO coefficients:

\[ d_{ij} = -\frac{n^4}{4} r_{ji} \]