Lecture 7

Anisotropic linear media

Linear susceptibility is a tensor

\[ \mathbf{P} = \varepsilon_0 \chi \mathbf{E}. \]

vector \quad tensor \quad vector

\[ P_i = \varepsilon_0 \sum_{j \in \{x,y,z\}} \chi_{ij} E_j \]

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 polarization 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 diagonalize 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>$n_0$</th>
<th>$n_e$</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$_3$)$_6$</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>1.960</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>1.967</td>
<td>+0.047</td>
</tr>
</tbody>
</table>

### Biaxial crystals

<table>
<thead>
<tr>
<th>Material</th>
<th>Crystal system</th>
<th>$n_0$</th>
<th>$n_\alpha$</th>
<th>$n_\beta$</th>
<th>$n_\gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>borax Na$_2$(B$_4$O$_6$)(OH)$_4$·8H$_2$O</td>
<td>Monoclinic</td>
<td>1.447</td>
<td>1.469</td>
<td>1.472</td>
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</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>
<td></td>
</tr>
<tr>
<td>mica, biotite K(Mg,Fe)$_3$(AlSi$_3$O$_10$)(F,OH)$_2$</td>
<td>Monoclinic</td>
<td>1.595</td>
<td>1.640</td>
<td>1.640</td>
<td></td>
</tr>
<tr>
<td>mica, muscovite KAl$_2$(AlSi$_3$O$_10$)(F,OH)$_2$</td>
<td>Monoclinic</td>
<td>1.563</td>
<td>1.596</td>
<td>1.601</td>
<td></td>
</tr>
<tr>
<td>olivine (Mg,Fe)$_2$SiO$_4$</td>
<td>Orthorhombic</td>
<td>1.640</td>
<td>1.660</td>
<td>1.680</td>
<td></td>
</tr>
<tr>
<td>perovskite CaTiO$_3$</td>
<td>Orthorhombic</td>
<td>2.300</td>
<td>2.340</td>
<td>2.380</td>
<td></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>
<td></td>
</tr>
<tr>
<td>ulexite NaCaB$_5$O$_6$(OH)$_6$·5H$_2$O</td>
<td>Triclinic</td>
<td>1.490</td>
<td>1.510</td>
<td>1.520</td>
<td></td>
</tr>
</tbody>
</table>
Nonlinear Susceptibility Tensor

Nonlinear susceptibility is a 3-rd rank tensor

\[ \chi^{(2)}_{ijk} \quad (3 \times 3 \times 3 \text{ tensor}) \]

\[ P_i = \varepsilon_0 \sum_{j,k} \chi_{ijk} E_j E_k \] (7.1)

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:

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

Kleinman’s full permutation symmetry condition

\[ P_i = \varepsilon_0 \sum_{j,k} \chi_{ijk} E_j E_k \]

... and does not actually depend of frequencies that participate, so instead of writing \( \chi^{(2)}_{ijk}(\omega) = \omega_3 \) 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

\[
E = \begin{bmatrix}
E_x \\
E_y \\
E_z
\end{bmatrix} = \begin{bmatrix}
E_1 \\
E_2 \\
E_3
\end{bmatrix} = \begin{bmatrix}
E_{1,\omega_1} \cos(\omega_1 t) + E_{1,\omega_2} \cos(\omega_2 t) + E_{1,\omega_3} \cos(\omega_3 t) \\
E_{2,\omega_1} \cos(\omega_1 t) + E_{2,\omega_2} \cos(\omega_2 t) + E_{2,\omega_3} \cos(\omega_3 t) \\
E_{3,\omega_1} \cos(\omega_1 t) + E_{3,\omega_2} \cos(\omega_2 t) + E_{3,\omega_3} \cos(\omega_3 t)
\end{bmatrix}
\]

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:

\[
x \quad P_1 = \varepsilon_0 \sum_{j,k} \chi_{1jk} E_j E_k = \varepsilon_0 \left\{ \chi_{111} E_1 E_1 + \chi_{112} E_1 E_2 + \chi_{113} E_1 E_3 + \right. \\
+ \chi_{121} E_2 E_1 + \chi_{122} E_2 E_2 + \chi_{123} E_2 E_3 + \right. \\
+ \chi_{131} E_3 E_1 + \chi_{132} E_3 E_2 + \chi_{133} E_3 E_3 \left\}
\]

\[
y \quad P_2 = \varepsilon_0 \sum_{j,k} \chi_{2jk} E_j E_k = ...
\]

\[
z \quad P_3 = \varepsilon_0 \sum_{j,k} \chi_{3jk} E_j E_k = ...
\]
Nonlinear Susceptibility Tensor

Now introduce the tensor:

$$d_{ijk} = \frac{1}{2} \chi^{(2)}_{ijk}$$

(historical convention !)

and write:

\[ P_1 = 2\varepsilon_0 \left( d_{111}E_1E_1 + d_{122}E_2E_2 + d_{133}E_3E_3 + 2d_{123}E_2E_3 + 2d_{113}E_1E_3 + 2d_{112}E_1E_2 \right) \]

\[ P_2 = 2\varepsilon_0 \left( d_{211}E_1E_1 + d_{222}E_2E_2 + d_{233}E_3E_3 + 2d_{223}E_2E_3 + 2d_{213}E_1E_3 + 2d_{212}E_1E_2 \right) \]

\[ P_3 = 2\varepsilon_0 \left( d_{311}E_1E_1 + d_{322}E_2E_2 + d_{333}E_3E_3 + 2d_{323}E_2E_3 + 2d_{313}E_1E_3 + 2d_{312}E_1E_2 \right) \]

Now reduced to only 18 components

Reduce 3D tensor to 2D matrix

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

Second-order susceptibility described using contracted notation

\[ jk \rightarrow l \]

\[ jk = \begin{array}{cccc} 11 & 22 & 33 & 23,32 \\ 31,13 & 31,13 & 5 & 12,21 \end{array} \]

\[ l = \begin{array}{cccc} 1 & 2 & 3 & 4 \\ 5 & 6 \end{array} \]
Nonlinear Susceptibility Tensor

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

\[
\begin{bmatrix}
Px \\
Py \\
Pz
\end{bmatrix} = 2\epsilon_0 \begin{bmatrix}
d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{bmatrix} \begin{bmatrix}
E_x^2 \\
E_y^2 \\
E_z^2 \\
2E_yE_z \\
2E_xE_z \\
2E_xE_y
\end{bmatrix}
\]  \hspace{1cm} (7.3)

Imagine we have only x-components for \( P \) and \( E \) – get familiar (see 5.1 with \( \chi^{(2)} = 2\,d_{NL} \))

\[
P(t) = 2\epsilon_0 d_{NL} E^2(t)
\]  \hspace{1cm} (7.4)
By further applying Kleinman symmetry, we find that $d_{il}$ matrix has **only 10 independent elements**

$$d_{il} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix}.$$
Nonlinear Susceptibility Tensor

Only 10 independent elements

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

\[
\begin{align*}
&d_{11} \quad d_{12} \quad d_{13} \quad d_{14} \quad d_{15} \quad d_{16} \\
&d_{16} \quad d_{22} \quad d_{23} \quad d_{24} \quad d_{14} \quad d_{12} \\
&d_{15} \quad d_{24} \quad d_{33} \quad d_{23} \quad d_{13} \quad d_{14}.
\end{align*}
\]
Spatial symmetries of crystals further reduce the amount of independent tensor elements
Nonlinear Susceptibility Tensor

‘Cubic’ crystals e.g. GaAs, GaP, InAs, ZnS, ZnSe – are optically isotropic but nonlinear!

KDP

GaAs

LiNbO₃

GaSe

KTP

GaSe

Zero nonlinearity

TABLE 16.1. The Form of the Nonlinear Optical Tensor \( d_{ij} \) as Defined by (16.1-1)

Key to Notation

- Zero modulus
- Nonzero modulus
- Equal moduli numerically equal, but opposite in sign

Centrosymmetric Classes (all moduli vanish)

Noncentrosymmetrical Classes

Triclinic

Class 1

Monoclinic

Class 2

Class m

Orthorhombic

Class 222

Class mm2

Class 4

Class 4₁

Class 4₂

Class 4mm

Tetragonal

Class 4

Class 4₁

Class 4₂

Class 4mm

Hexagonal

Class 6

Class 6mm

Class 6m

Class 6₂2

Class 6₃2

Class 6m2

Class 6m

Source: Reference 2.
Nonlinear Susceptibility Tensor

Uniaxial crystal classes (Continued)

classes 6 and 4

classes 6mm and 4mm

classes 622 and 422

class 4

class 32

class $\overline{4}2m$

Isotropic crystal classes

GaAs
GaP
ZnSe
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

KTP, KNbO$_3$
BaNaNb$_5$O$_{15}$
LiB$_3$O$_3$ (LBO)
Nonlinear Susceptibility Tensor

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

\[
d_{ll} = \begin{bmatrix}
0 & 0 & 0 & d_{31} & -d_{22} \\
-d_{22} & d_{22} & 0 & 0 & d_{31} \\
d_{31} & 0 & d_{31} & 0 & 0 \\
d_{31} & 0 & d_{33} & 0 & 0 \\
0 & 0 & 0 & 0 & 0
\end{bmatrix}
\]

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

Class: 3m → LiNbO$_3$, LiTaO$_2$, BaB$_2$O$_2$ (BBO)

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

As active electron bonds $E$-field applied in $xy$

GaAs crystal

GaAs

motion in $xy$

motion in $z$
Physical origin of off-diagonal elements in $d_{ijk}$ tensor

after Stegeman NLO book
Nonlinear Susceptibility Tensor

Crystal of class $\overline{4}3m$ (e.g. Gallium Arsenide, **GaAs**)

- **Ga** or **Zn**
- **As** or **S**

*only $d_{14}$*

<table>
<thead>
<tr>
<th>classes</th>
<th>$\overline{4}3m$</th>
<th>and 23</th>
</tr>
</thead>
<tbody>
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</tr>
</tbody>
</table>
Nonlinear Susceptibility Tensor

\[ \hat{P}^{(2)} = 2\epsilon_0 \begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix} \begin{pmatrix} \hat{E}_x \hat{E}_x \\ \hat{E}_y \hat{E}_y \\ \hat{E}_z \hat{E}_z \\ 2\hat{E}_y \hat{E}_z \\ 2\hat{E}_z \hat{E}_x \\ 2\hat{E}_x \hat{E}_y \end{pmatrix} \]

GaAs

(7.4)
Nonlinear Susceptibility Tensor

How to calculate effective nonlinearity?

$$\begin{bmatrix} P_x(t) \\ P_y(t) \\ P_z(t) \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x^2(t) \\ E_y^2(t) \\ E_z^2(t) \end{bmatrix}$$

$$\frac{E_x(t) E_y(t)}{E_z(t)}$$

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

The pump wave $E(t)$ is polarized along $z$-axis

$$\begin{bmatrix} P_x(t) \\ P_y(t) \\ P_z(t) \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & 0 & 0 & 0 & d_{31} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 0 \\ 0 \\ E_z^2(t) \\ 0 \\ 0 \\ 0 \end{bmatrix} = 2\epsilon_0 \ d_{33} \begin{bmatrix} 0 \\ 0 \\ E_z^2(t) \end{bmatrix}$$

$$P_z(t) = 2\epsilon_0 \ d_{33} \ E_z^2(t)$$

similar to scalar equation with $d_{eff}=d_{33}$
Nonlinear Susceptibility Tensor

input field \[ E_z(t) = \text{Re}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}) = \frac{1}{2} (E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + \text{c. c.}) \]

Need to find z-component of \[ P(\omega_3) = P(\omega_1 + \omega_2) = \text{Re}\{P(\omega_3)e^{i(\omega_1+\omega_2)t}\} = \frac{1}{2} \left( Pe^{i(\omega_1+\omega_2)t} + P^*e^{-i(\omega_1+\omega_2)t}\right) \]

\[ P_z^{(NL)}(t) = 2\varepsilon_0 d_{33} E_z^2 = 2\varepsilon_0 d_{33} \left( \frac{1}{4} (E_{1,z} e^{i\omega_1 t} + E_{2,z} e^{i\omega_2 t} + \text{c.c.})^2 \right) = \ldots \]

\[ = 2\varepsilon_0 d_{33} \frac{1}{4} (E_{1,z}^2 e^{2i\omega_1 t} + E_{2,z}^2 e^{2i\omega_2 t} + 2E_{1,z} E_{2,z} e^{i(\omega_1+\omega_2)t} + 2E_{1,z}^* E_{2,z}^* e^{i(\omega_1-\omega_2)t} + \text{c. c.}) + 2\varepsilon_0 d_{33} \frac{1}{2} (E_{1,z}^* E_{1,z} + E_{2,z}^* E_{2,z}) \]

for SFG term: \[ P(\omega_3) = P(\omega_1 + \omega_2) - \text{pick only components with } \pm (\omega_1 + \omega_2) \]

at \( \omega_3 \)

\[ P_z^{(NL)}(t) = 2\varepsilon_0 d_{33} \frac{1}{4} (2E_{1,z} E_{2,z} e^{i(\omega_1+\omega_2)t} + \text{c. c.}) = \varepsilon_0 2d_{33} \frac{1}{2} E_{1,z} E_{2,z} e^{i(\omega_1+\omega_2)t} + \text{c. c.} \]

Amplitude (Fourier component) of polarization at \textbf{sum frequency}

\[ P_z(\omega_3) = \varepsilon_0 2d_{33} E_{1,z} E_{2,z} \]

same as \[ P(t) = P_z(\omega_3) \cos(\omega_3 t) \]
Nonlinear Susceptibility Tensor

once you know $d_{eff}$, you can treat fields as scalars

thus

$$P(\omega_1 + \omega_2) = 2\epsilon_0 d_{33}E_1E_2$$

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

- by analogy with the scalar case from previous lectures
2) Effective nonlinearity, GaAs

for input fields along XY direction

\[ E(t) = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} \]

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

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

\[
E_x = \text{Re}\left\{ \frac{1}{\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}) \right\} = \frac{1}{2\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c. c.)
\]
\[
E_y = \text{Re}\left\{ \frac{1}{\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}) \right\} = \frac{1}{2\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c. c.)
\]

\[
P_{NL}(t) = P_2(t) = 2\epsilon_0 d_{14}(2E_x E_y) = 4\epsilon_0 d_{14} \frac{1}{2} \frac{1}{4} (E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c. c.)^2
\]

\[ \rightarrow \text{pick terms with } \omega_1 + \omega_2 \]

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

for SFG \[ P(\omega_1 + \omega_2) = 2\epsilon_0 d_{14} E_1 E_2 \quad \rightarrow \quad d_{eff} = d_{14} \]

Can treat fields as scalars keeping in mind that \( E_1 \) and \( E_2 \) are in \( xy \) and \( E_3 \) is in \( z \)-direction.
Because of the off-diagonal tensor elements, we can generate SFG with the output polarization perpendicular to the input polarizations.
Electrooptic effect

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.

$$D_i = \varepsilon_0 \sum_j \varepsilon_{ij} E_j$$

Linear anisotropic medium:

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

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

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \varepsilon_{XX} & 0 & 0 \\ 0 & \varepsilon_{YY} & 0 \\ 0 & 0 & \varepsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix}.$$

the dielectric constant is a second-rank tensor

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

The index ellipsoid

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

\[ \frac{X^2}{\epsilon_{XX}} + \frac{Y^2}{\epsilon_{YY}} + \frac{Z^2}{\epsilon_{ZZ}} = 1. \]

two allowed directions of polarization with two distinct \( n \) coefficients

beam \( k \)-vector
First-order electrooptic effect in anisotropic crystals

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

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

two allowed directions of polarization with two distinct \( n \) coefficients

direction of propagation
beam \( k \)-vector

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:

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

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

Baseball shape
\( n_e > n_o \)

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

Uniaxial crystal: \( n_1 = n_2 = n_0; \quad n_3 = n_e \)

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:

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

\[
\Rightarrow n(\theta) = \frac{n_0 n_e}{(n_0^2 \sin^2 \theta + n_e^2 \cos^2 \theta)^{1/2}}
\]
First-order electrooptic effect in anisotropic crystals

the index ellipsoid

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

General case: (only 6 independent terms)

\[ \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{bmatrix} \]

General case for the index ellipsoid

\[ \left( \frac{1}{n_1^2} \right)_{1} x^2 + \left( \frac{1}{n_2^2} \right)_{2} y^2 + \left( \frac{1}{n_3^2} \right)_{3} z^2 + 2 \left( \frac{1}{n_2^2} \right)_{4} yz + 2 \left( \frac{1}{n_2^2} \right)_{5} xz + 2 \left( \frac{1}{n_2^2} \right)_{6} xy = 1. \]
The essence of linear electrooptic effect

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

or

\[
\begin{bmatrix}
\Delta(1/n^2)_1 \\
\Delta(1/n^2)_2 \\
\Delta(1/n^2)_3 \\
\Delta(1/n^2)_4 \\
\Delta(1/n^2)_5 \\
\Delta(1/n^2)_6 \\
\end{bmatrix} =
\begin{bmatrix}
r_{11} & r_{12} & r_{13} \\
r_{21} & r_{22} & r_{23} \\
r_{31} & r_{32} & r_{33} \\
r_{41} & r_{42} & r_{43} \\
r_{51} & r_{52} & r_{53} \\
r_{61} & r_{62} & r_{63} \\
\end{bmatrix}
\begin{bmatrix}
E_x \\
E_y \\
E_z \\
\end{bmatrix}
\]
First-order electrooptic effect in anisotropic crystals

\[ r_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \]

KDP

(for class \( \bar{4}2m \),

\[ r_{ji} \leftrightarrow d_{ij} \]

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

rotate \( xy \) plane by 45º and get:

\[
\frac{x^2}{n_{x_{r}}^2} + \frac{y^2}{n_{y_{r}}^2} = 1
\]

\[ n_{x_{r}} = n_0 - \frac{1}{2} n_0^3 r_{63} E_z, \]

\[ n_{y_{r}} = n_0 + \frac{1}{2} n_0^3 r_{63} E_z, \]

\[
\frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + 2xyr_{63}E_z = 1
\]
First-order electrooptic effect in anisotropic crystals

Electrooptic modulator, KDP

Evolution of the vertical polarization originally sent to the modulator

Gamma - phase difference

Half-wave voltage \( \sim \text{kV range} \)

\[
V_{\lambda/2} = \frac{\pi c}{\omega n_0^3 r_{63}},
\]
First-order electrooptic effect in anisotropic crystals

Electrooptic modulator, lithium niobate LiNbO$_3$

$$
\begin{bmatrix}
0 & -r_{22} & r_{13} \\
0 & r_{22} & r_{13} \\
0 & 0 & r_{33} \\
r_{42} & 0 & 0 \\
r_{42} & 0 & 0
\end{bmatrix}
$$

Apply field along x-axis (transverse effect)

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

Half-wave voltage in lithium niobate is 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.

\[ \omega_3 = \omega_1 + \omega_2 \]

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

**EO**

\[ \omega = 0 + \omega \]

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

\[ E(t) = E_0 + E_\omega \cos(\omega t) \]

(we take one-dimensional scalar form)

1) From NLO point of view

\[ P(t) = \varepsilon_0 \chi^{(2)} E^2(t) = \varepsilon_0 2d_{NL} E^2(t) \Rightarrow \varepsilon_0 2d_{NL} (E_0 + E_\omega \cos(\omega t))^2 \]

\[ = \varepsilon_0 2d_{NL} (E_0^2 + 2E_0 E_\omega \cos(\omega t) + [E_\omega \cos(\omega t)]^2) \]

\[ P_\omega(t) = \varepsilon_0 4d_{NL} E_0 E_\omega \cos(\omega t); \quad \text{but from} \quad P = \varepsilon_0 \chi E \quad \rightarrow \quad P_\omega(t) = \varepsilon_0 \Delta \chi E_\omega \cos(\omega t); \quad \rightarrow \quad \Delta \chi = 4d_{NL} E_0 \]

also \[ \chi = n^2 - 1 \quad \rightarrow \quad \Delta \chi = 2n \Delta n \]

thus\[ 4d_{NL} E_0 = 2n \Delta n \]

\[ \Delta n = \frac{2d_{NL} E_0}{n} \]

2) From EO point of view

\[ \Delta \left( \frac{1}{n^2} \right) = r_{EO} E_0 \quad \rightarrow \quad \left( -\frac{2}{n^3} \right) \Delta n = r_{EO} E_0 \]

\[ \Delta n = -\frac{n^3}{2} r_{EO} E_0 \]

\[ d_{NL} = -\frac{n^4}{4} r_{EO} \]

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

Electrooptic effect : - see Yariv 'Opt Waves in Cryst' (p 561) - for rijk vs dijk relations