Lecture 7

Second-order $X^{(2)}$ nonlinear optical materials. Crystal classes and their symmetries. Nonlinear optical tensors. Electrooptic effect.

Anisotropic linear media

Linear susceptibility is a **tensor**

$$\underbrace{\mathbf{P}}_{\text{vector}} = \varepsilon_0 \underbrace{\boldsymbol{\chi}}_{\text{tensor}} \underbrace{\mathbf{E}}_{\text{vector}}.$$

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

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, $\boxed{\chi_{ij} = \chi_{ji}}$, i.e. the χ tensor is real and symmetric.

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

$$\boxed{\begin{aligned} P_x &= \varepsilon_0 \chi_{xx} E_x \\ P_y &= \varepsilon_0 \chi_{yy} E_y \\ P_z &= \varepsilon_0 \chi_{zz} E_z \end{aligned}}$$

Anisotropic linear media

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

hence

$$n_{xx} = \sqrt{1+\chi_{xx}}$$
$$n_{yy} = \sqrt{1+\chi_{yy}}$$
$$n_{zz} = \sqrt{1+\chi_{zz}}$$

or

$$\varepsilon = \varepsilon_0 \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix}$$

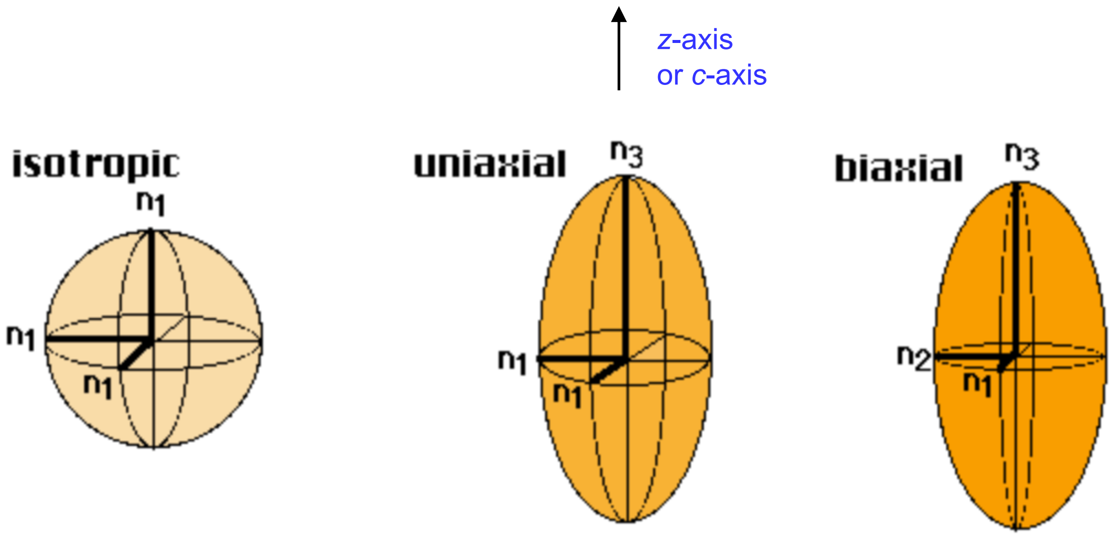
Waves with different polarizations will see different refractive indices and travel at different speeds.

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 $n_1 = n_2$ n_3

Material	Crystal system	n_o	n_e	Δn
barium borate BaB_2O_4	Trigonal	1.6776	1.5534	−0.1242
beryl $Be_3Al_2(SiO_3)_6$	Hexagonal	1.602	1.557	−0.045
calcite $CaCO_3$	Trigonal	1.658	1.486	−0.172
ice H_2O	Hexagonal	1.309	1.313	+0.004
lithium niobate $LiNbO_3$	Trigonal	2.272	2.187	−0.085
magnesium fluoride MgF_2	Tetragonal	1.380	1.385	+0.006
quartz SiO_2	Trigonal	1.544	1.553	+0.009
ruby Al_2O_3	Trigonal	1.770	1.762	−0.008
rutile TiO_2	Tetragonal	2.616	2.903	+0.287
sapphire Al_2O_3	Trigonal	1.768	1.760	−0.008
silicon carbide SiC	Hexagonal	2.647	2.693	+0.046
tourmaline (complex silicate)	Trigonal	1.669	1.638	−0.031
zircon, high $ZrSiO_4$	Tetragonal	1.960	2.015	+0.055
zircon, low $ZrSiO_4$	Tetragonal	1.920	1.967	+0.047

Biaxial crystals n_1 n_2 n_3

Material	Crystal system	n_α	n_β	n_γ
borax $Na_2(B_4O_5)(OH)_4 \cdot 8H_2O$	Monoclinic	1.447	1.469	1.472
epsom salt $MgSO_4 \cdot 7H_2O$	Monoclinic	1.433	1.455	1.461
mica, biotite $K(Mg,Fe)_3(AlSi_3O_{10})(F,OH)_2$	Monoclinic	1.595	1.640	1.640
mica, muscovite $KAl_2(AlSi_3O_{10})(F,OH)_2$	Monoclinic	1.563	1.596	1.601
olivine $(Mg,Fe)_2SiO_4$	Orthorhombic	1.640	1.660	1.680
perovskite $CaTiO_3$	Orthorhombic	2.300	2.340	2.380
topaz $Al_2SiO_4(F,OH)_2$	Orthorhombic	1.618	1.620	1.627
ulexite $NaCaB_5O_6(OH)_6 \cdot 5H_2O$	Triclinic	1.490	1.510	1.520

Nonlinear Susceptibility Tensor

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

$$\boxed{P_i = \epsilon_0 \sum_{j,k} \chi_{ijk}\, E_j E_k} \qquad (7.1)$$

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

... 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 !

Nonlinear Susceptibility Tensor

amplitudes

Consider mutual interaction of three waves at $\omega_1, \ \omega_2, \ \text{and} \ \ \omega_3 = \omega_1 + \omega_2$

Assume ***E*** is the total field <u>vector</u>

$$\boxed{\boldsymbol{E} = \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} = \begin{bmatrix} E_1 \\ E_2 \\ E_3 \end{bmatrix} = \begin{bmatrix} E_{1,\omega_1}\cos(\omega_1 t) + E_{1,\omega_2}\cos(\omega_2 t) + E_{1,\omega_3}\cos(\omega_3 t) \\ E_{2,\omega_1}\cos(\omega_2 t) + E_{2,\omega_2}\cos(\omega_2 t) + E_{2,\omega_3}\cos(\omega_3 t) \\ E_{3,\omega_1}\cos(\omega_3 t) + E_{2,\omega_2}\cos(\omega_2 t) + E_{3,\omega_3}\cos(\omega_3 t) \end{bmatrix}} \qquad (7.2)$$

then $$P_i = \epsilon_0 \sum_{j,k} \chi_{ijk}\, E_j E_k \qquad i,j,k = 1,2,3 \ \ (= x,y,z)$$

For example, x –component of the nonlinear polarization ***P*** is:

x $$P_1 = \epsilon_0 \sum_{j,k} \chi_{1jk}\, E_j E_k = \epsilon_0 \,\{ \chi_{111}E_1E_1 + \chi_{\underline{112}}\underline{E_1E_2} + \chi_{1\underline{13}}\underline{E_1E_3} + $$
$$+ \chi_{\underline{121}}\underline{E_2}E_1 + \chi_{122}E_2E_2 + \chi_{1\underline{23}}\underline{E_2E_3} + $$
$$+ \chi_{\underline{131}}\underline{E_3}E_1 + \chi_{\underline{132}}\underline{E_3}E_2 + \chi_{133}E_3E_3 \ \}$$

note that the terms underlined by the same color are equal

y $$P_2 = \epsilon_0 \sum_{j,k} \chi_{2jk}\, E_j E_k = \ \ldots$$

z $$P_3 = \epsilon_0 \sum_{j,k} \chi_{3jk}\, E_j E_k = \ \ldots$$

Nonlinear Susceptibility Tensor

Now introduce the tensor:

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

(historical convention !)

and write:

$d_{112}+d_{121}= 2d_{112}$

$$P_1 = 2\epsilon_0 \left(d_{111}E_1E_1 + d_{122}E_2E_2 + d_{133}E_3E_3 + 2d_{123}E_2E_3 + 2d_{113}E_1E_3 + 2d_{112}E_1E_2 \right)$$

$$P_2 = 2\epsilon_0 \left(d_{211}E_1E_1 + d_{222}E_2E_2 + d_{233}E_3E_3 + 2d_{223}E_2E_3 + 2d_{213}E_1E_3 + 2d_{212}E_1E_2 \right)$$

$$P_3 = 2\epsilon_0 \left(d_{311}E_1E_1 + d_{322}E_2E_2 + d_{333}E_3E_3 + 2d_{323}E_2E_3 + 2d_{313}E_1E_3 + 2d_{312}E_1E_2 \right)$$

now reduced to only 18 components

Matrix with 6x3 components

Reduce 3D tensor to 2D matrix

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

$$jk \rightarrow l$$

$jk =$	11	22	33	23,32	31,13	12,21
$l =$	1	2	3	4	5	6

Second-order susceptibility described using contracted notation

Nonlinear Susceptibility Tensor

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

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix} \qquad (7.3)$$

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

$$P(t) = 2\varepsilon_0 d_{NL} E^2(t) \qquad (7.4)$$

Nonlinear Susceptibility Tensor

By further applying Kleinman symmetry, we find that d_{il} matrix has **only 10 independent elements**

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

$$d_{il} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{bmatrix}.$$

for example: d_{14} is d_{123}

but d_{25} is d_{213} $=d_{123}$

and d_{36} is d_{312} $=d_{123}$

→ $d_{14}=d_{25}=d_{36}$ etc.

d_{11} d_{22} d_{33} d_{12} d_{13} d_{14} d_{15} d_{16} d_{23} d_{24}

Nonlinear Susceptibility Tensor

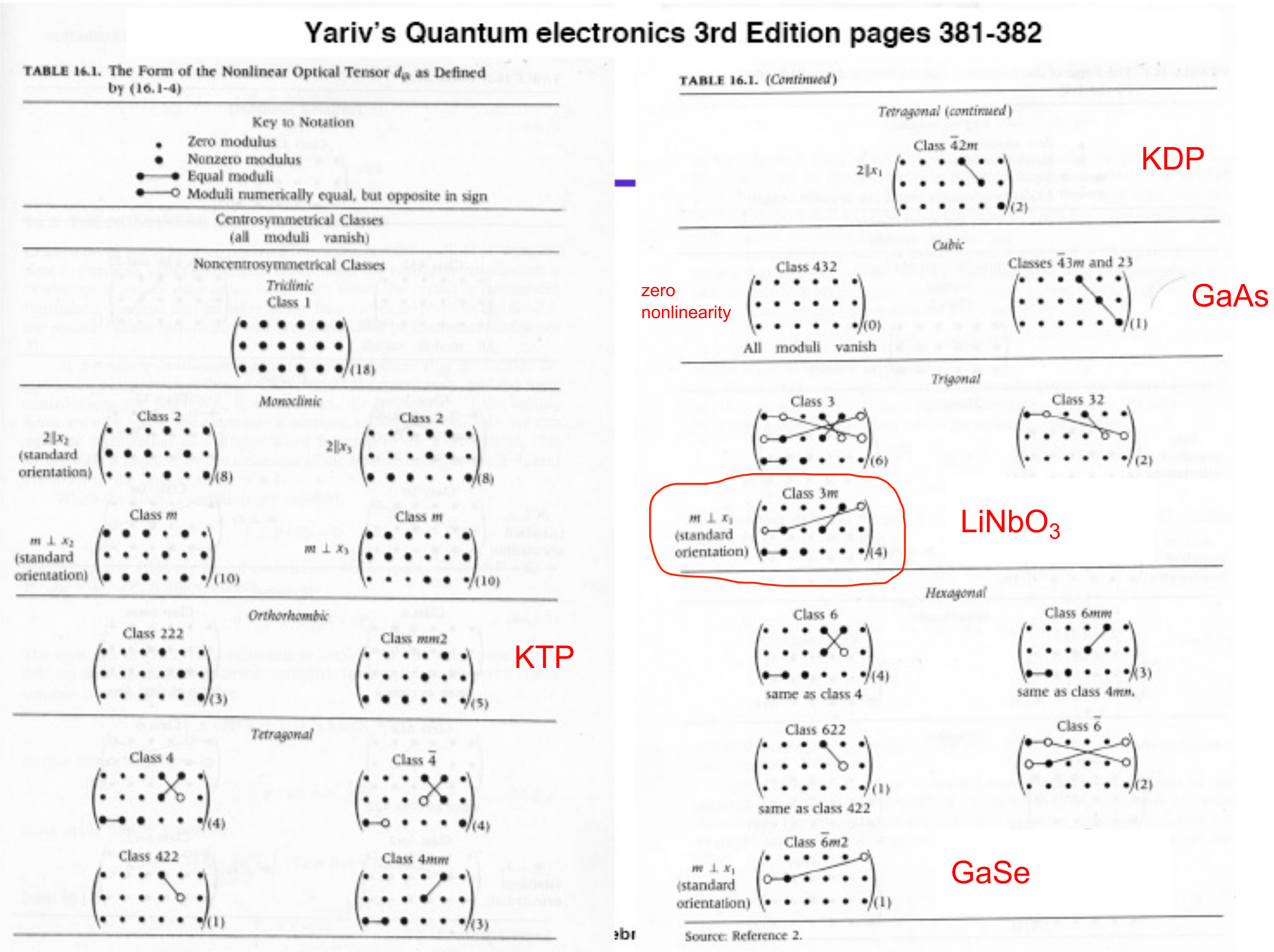
Only 10 independent elements

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

Nonlinear Susceptibility Tensor

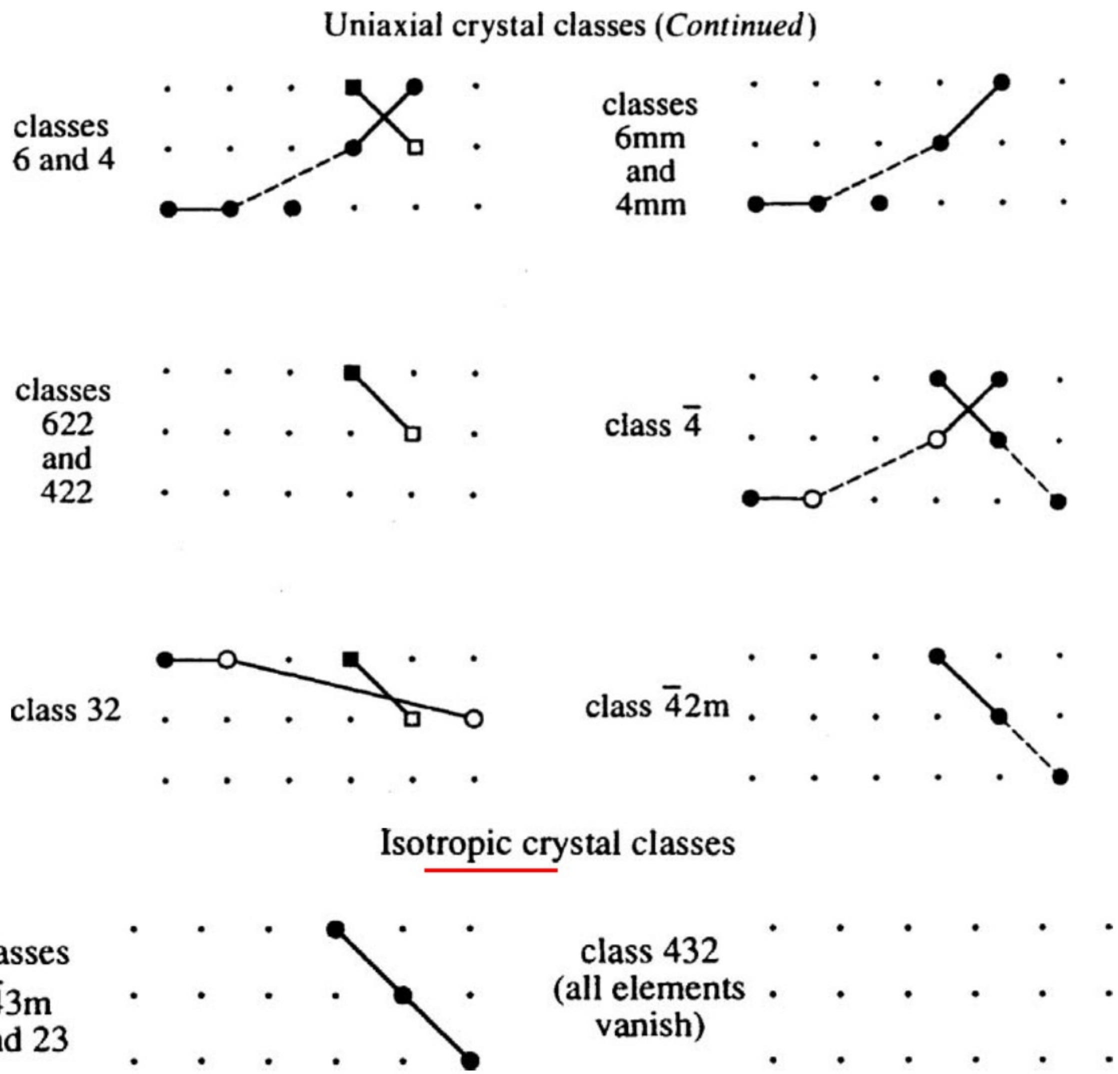
Spatial symmetries of crystals further reduce the amount of independent tensor elements

Nonlinear Susceptibility Tensor



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

Nonlinear Susceptibility Tensor



GaAs
GaP
ZnSe

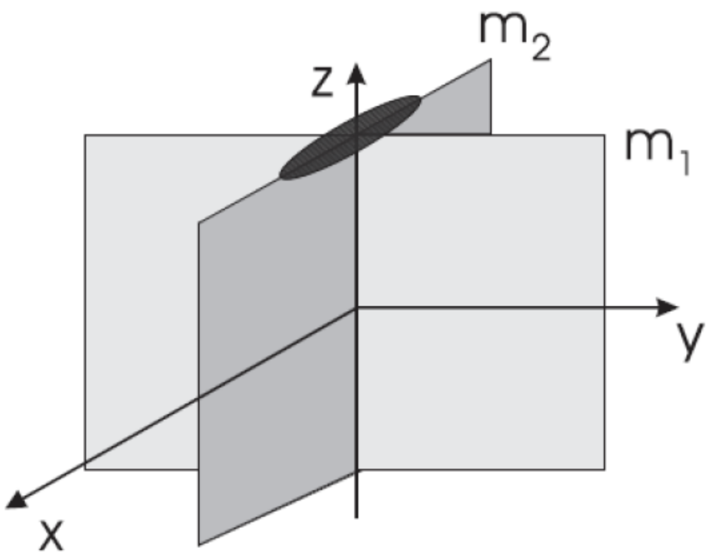
Nonlinear Susceptibility Tensor

point group mm2 KTP ($KTiO_2PO_4$) 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

$$\begin{aligned} m_1 &: (x, y, z) \longrightarrow (-x, y, z), \\ m_2 &: (x, y, z) \longrightarrow (x, -y, z), \\ 2 &: (x, y, z) \longrightarrow (-x, -y, z) \end{aligned}$$



Nonlinear Susceptibility Tensor

KTP ($KTiO_2PO_4$) crystal

$$\begin{matrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14}. \end{matrix}$$

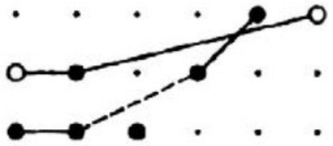
ortho-rhombic	$C_{2v} \parallel mm2$	$\begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix}$	KTP, $KNbO_3$ $BaNaNb_5O_{15}$ LiB_3O_3 (LBO)

Nonlinear Susceptibility Tensor

Crystal of class 3m (e.g. Lithium Niobate, **$LiNbO_3$**)

$$d_{il} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & \underline{\underline{d_{33}}} & 0 & 0 & 0 \end{bmatrix}$$

class 3m

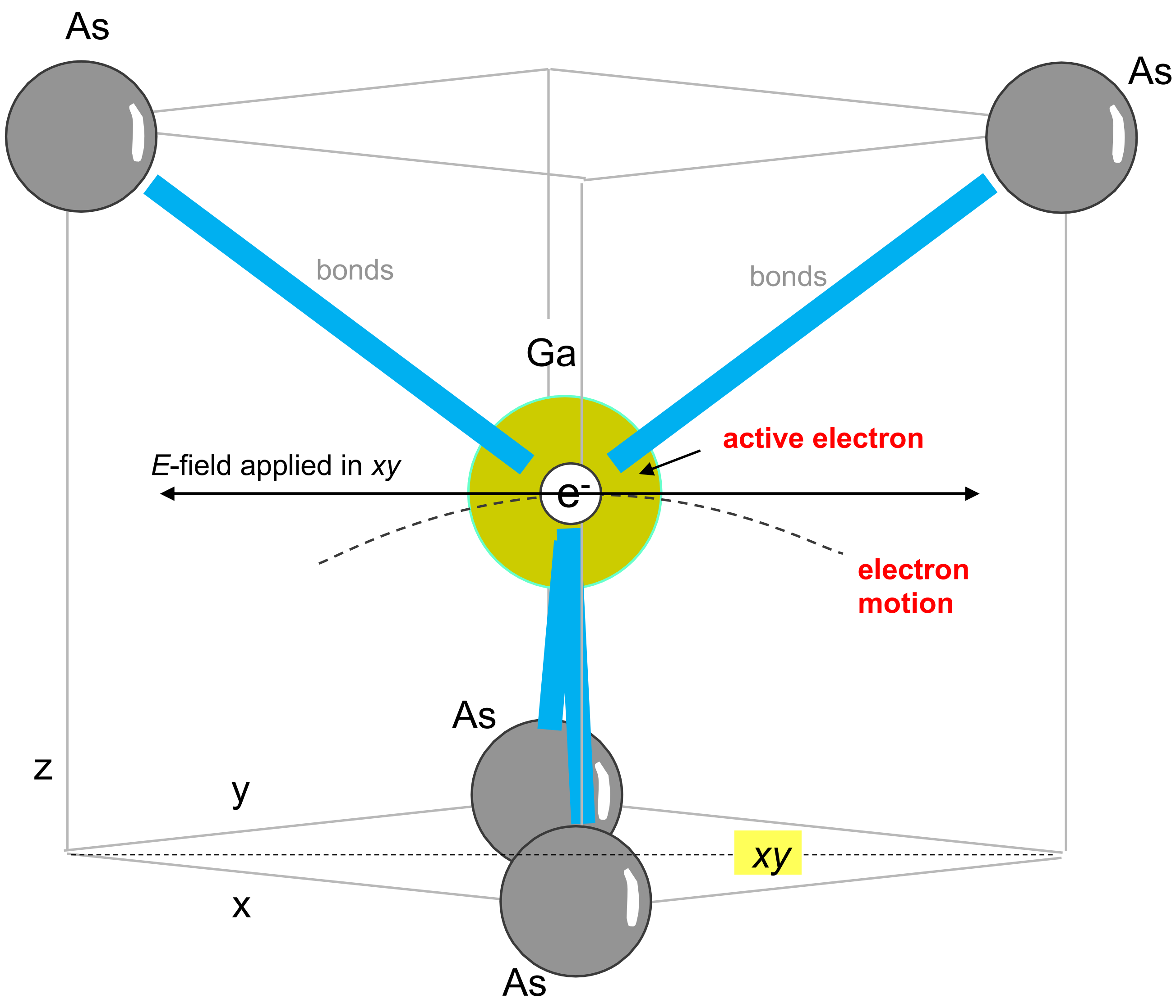


only d_{22} , d_{31} and d_{33}

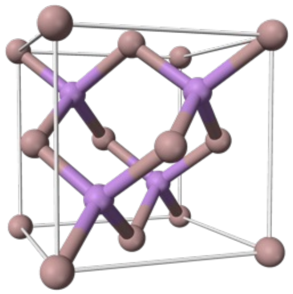
Class: 3m → $LiNbO_3$, $LiTaO_2$
BaB_2O_2 (BBO)

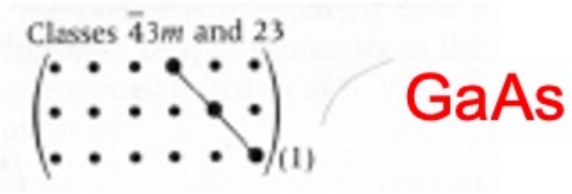
other crystals
of this class

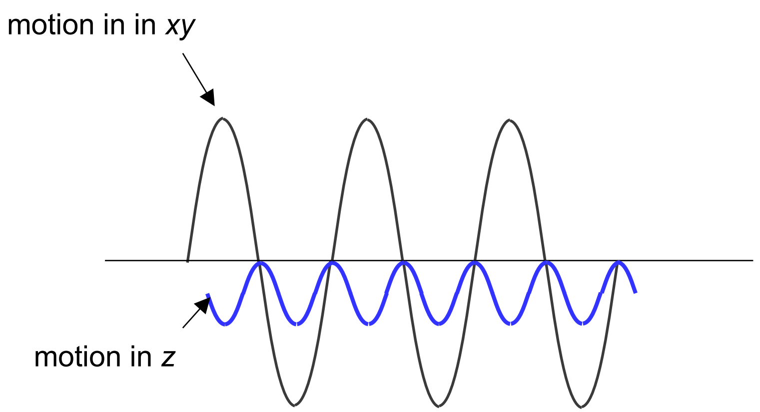
Physical origin of off-diagonal elements in d_{ijk} tensor



GaAs crystal



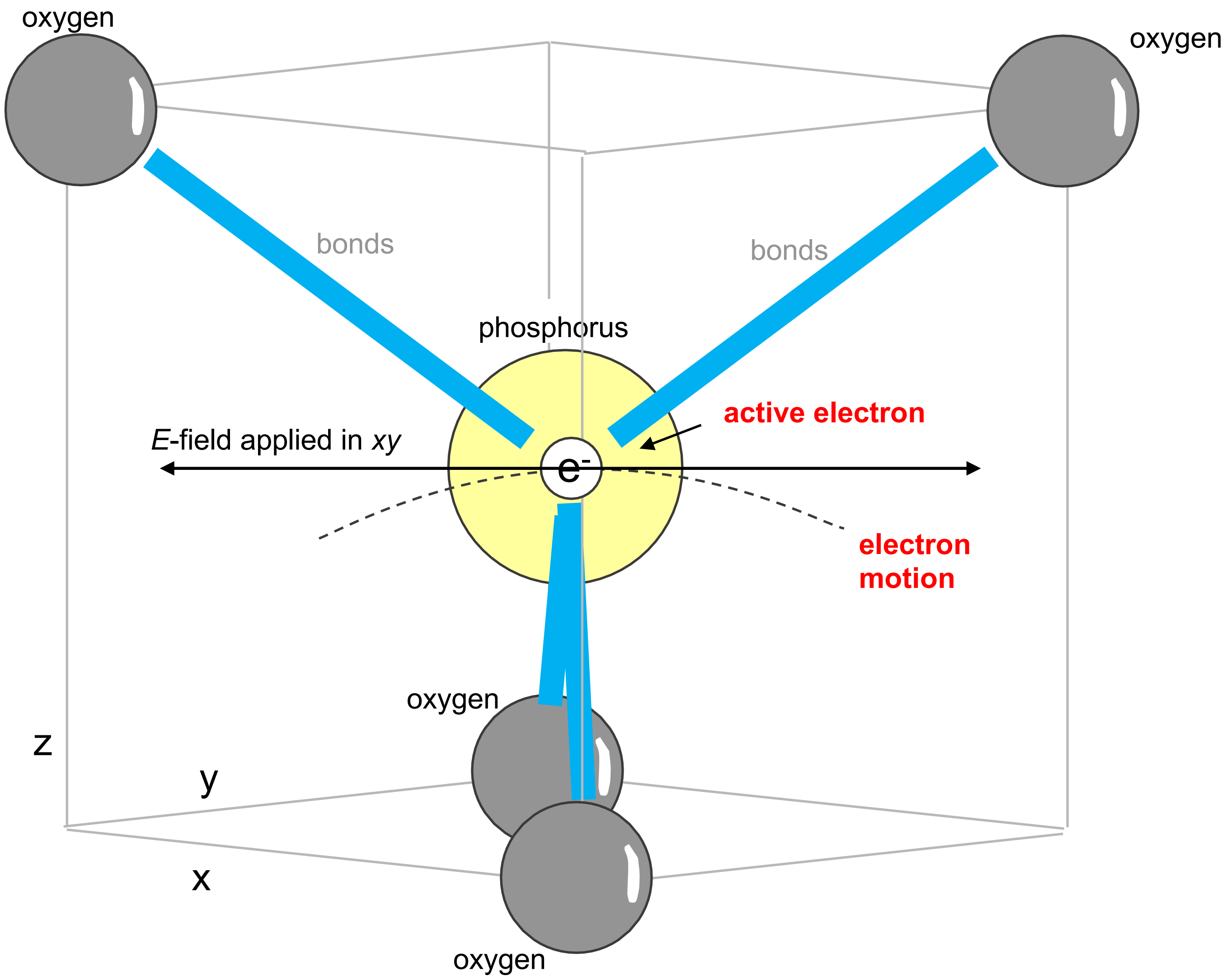


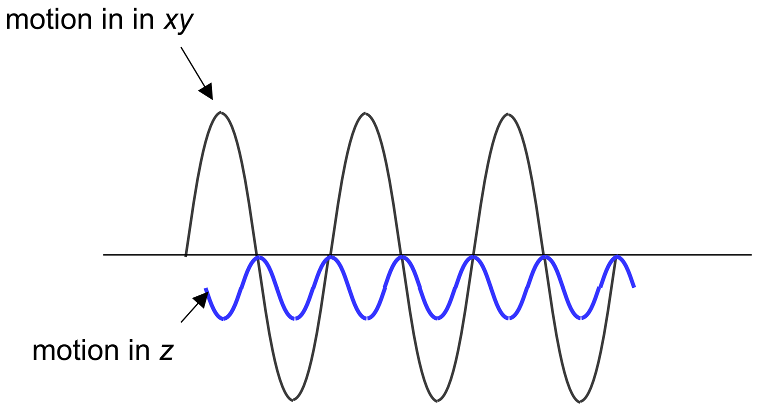


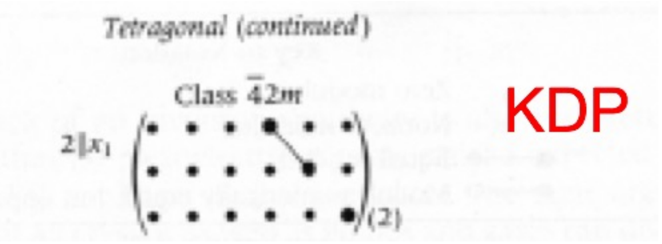
Physical origin of off-diagonal elements in d_{ijk} tensor

after Stegeman NLO book

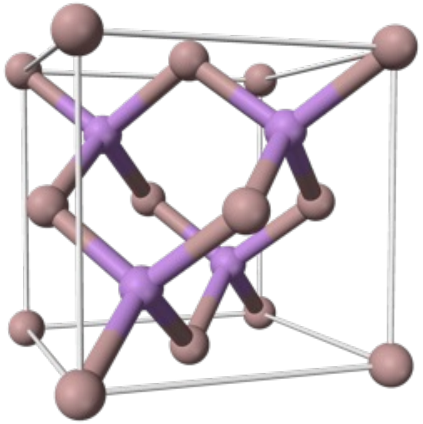
KDP crystal



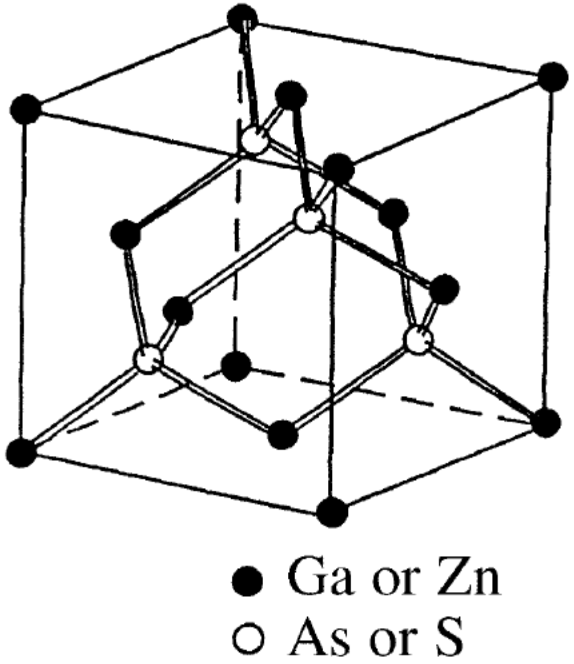




Nonlinear Susceptibility Tensor



GaAs



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

only d_{14}

Nonlinear Susceptibility Tensor

GaAs

$$\hat{P}^{(2)} = 2\epsilon_0 \begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix} \begin{pmatrix} \hat{E}_x\hat{E}_x \\ \hat{E}_y\hat{E}_y \\ \hat{E}_z\hat{E}_z \\ 2\hat{E}_y\hat{E}_z \\ 2\hat{E}_z\hat{E}_x \\ 2\hat{E}_x\hat{E}_y \end{pmatrix} \tag{7.4}$$

Nonlinear Susceptibility Tensor

How to calculate effective nonlinearity ?

NL polarization

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

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

$$d_{il} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & \underline{\underline{d_{33}}} & 0 & 0 & 0 \end{bmatrix}$$

1) $\mathrm{LiNbO_3}$ $d_{33} \neq 0$

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

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

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

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

Nonlinear Susceptibility Tensor

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

Need to find z-component of $P(\omega_3)$=$P(\omega_1+\omega_2) = \mathrm{Re}\{P(\omega_3)e^{i(\omega_1+\omega_2)t}\} = \frac{1}{2}\,(Pe^{i(\omega_1+\omega_2)t} + P^* e^{-i(\omega_1+\omega_2)t})$

$$P_z^{(NL)}(t) = 2\epsilon_0 d_{33} E_z^2 = 2\epsilon_0 d_{33}\frac{1}{4}(E_{1,z}e^{i\omega_1 t} + E_{2,z}e^{i\omega_2 t} + \text{c.c.})^2 = \cdots$$

$$= 2\epsilon_0 d_{33}\frac{1}{4}(E_{1,z}^2 e^{2i\omega_1 t} + E_{2,z}^2\, e^{2i\omega_2 t} + 2E_{1,z}E_{2,z}e^{i(\omega_1+\omega_2)t} + 2E_{1,z}E_{2,z}^* e^{i(\omega_1-\omega_2)t} + \mathrm{c.c.}) + 2\epsilon_0 d_{33}\frac{1}{2}(E_{1,z}E_{1,z}^* + E_{2,z}\, E_{2,z}^*)$$

for SFG term: $P(\omega_3)$=$P(\omega_1+\omega_2)$ – pick only components with $\pm(\omega_1+\omega_2)$

at ω_3 $$P_z^{(NL)}(t) = 2\epsilon_0 d_{33}\frac{1}{4}(2E_{1,z}E_{2,z}e^{i(\omega_1+\omega_2)t} + c.c.) = \epsilon_0 2d_{33}(\frac{1}{2}E_{1,z}E_{2,z}e^{i(\omega_1+\omega_2)t} + c.c.)$$

$\boxed{P_z(\omega_3) = \epsilon_0 2d_{33}E_{1,z}E_{2,z}}$ Amplitude (Fourier component) of polarization at **sum frequency**

same as $P(t) = P_z(\omega_3)\cos(\omega_3 \mathrm{t})$

Nonlinear Susceptibility Tensor

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

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

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

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

- by analogy with the scalar case from previous lectures

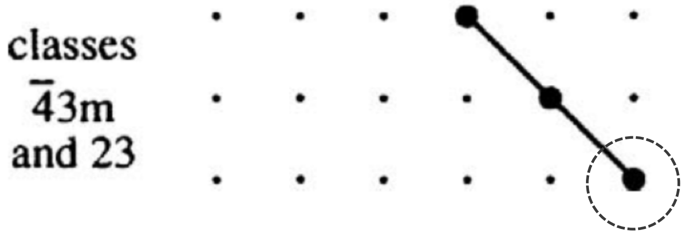
Nonlinear Susceptibility Tensor

2) Effective nonlinearity, GaAs

for input fields along XY direction

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

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



focus on this component:

GaAs

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

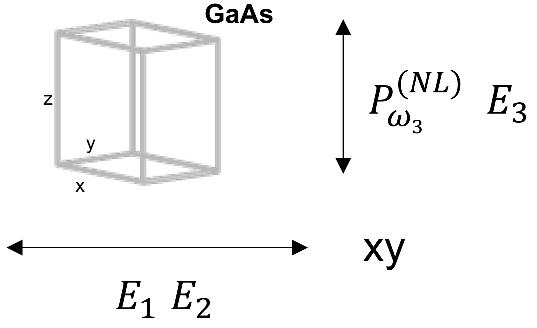
only d_{14}

SFG $\omega_3 = \omega_1 + \omega_2$

Need to find $\boldsymbol{P}(\omega_3)$

$$E_x = Re\{\frac{1}{\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t})\} = \frac{1}{2\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c)$$

$$E_y = Re\{\frac{1}{\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t})\} = \frac{1}{2\sqrt{2}}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c)$$



beam k-vector: goes into the page (so-called 110 direction)

$$\boldsymbol{P}_{NL}(t) = P_z(t) = 2\epsilon_0 d_{14}\left(2E_x E_y\right) = 4\epsilon_0 d_{14}\frac{1}{2}\frac{1}{4}(E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t} + c.c.)^2$$

→ pick terms with $\omega_1 + \omega_2$ $\quad P_{NL}(t,\omega_3) = \epsilon_0 d_{14}\frac{1}{2}(2E_1E_2 e^{i(\omega_1+\omega_2)t} + c.c.) = \epsilon_0 2d_{14}[\frac{1}{2}(E_1E_2 e^{i(\omega_1+\omega_2)t} + c.c.)]$

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

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

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.

Linear anisotropic medium:

$$D_i = \epsilon_0 \sum_j \epsilon_{ij} E_j$$

the dielectric constant is a second-rank tensor

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}.$$

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

$$\begin{bmatrix} D_X \\ D_Y \\ D_Z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \epsilon_{XX} & 0 & 0 \\ 0 & \epsilon_{YY} & 0 \\ 0 & 0 & \epsilon_{ZZ} \end{bmatrix} \begin{bmatrix} E_X \\ E_Y \\ E_Z \end{bmatrix}.$$

in the principal dielectric axes

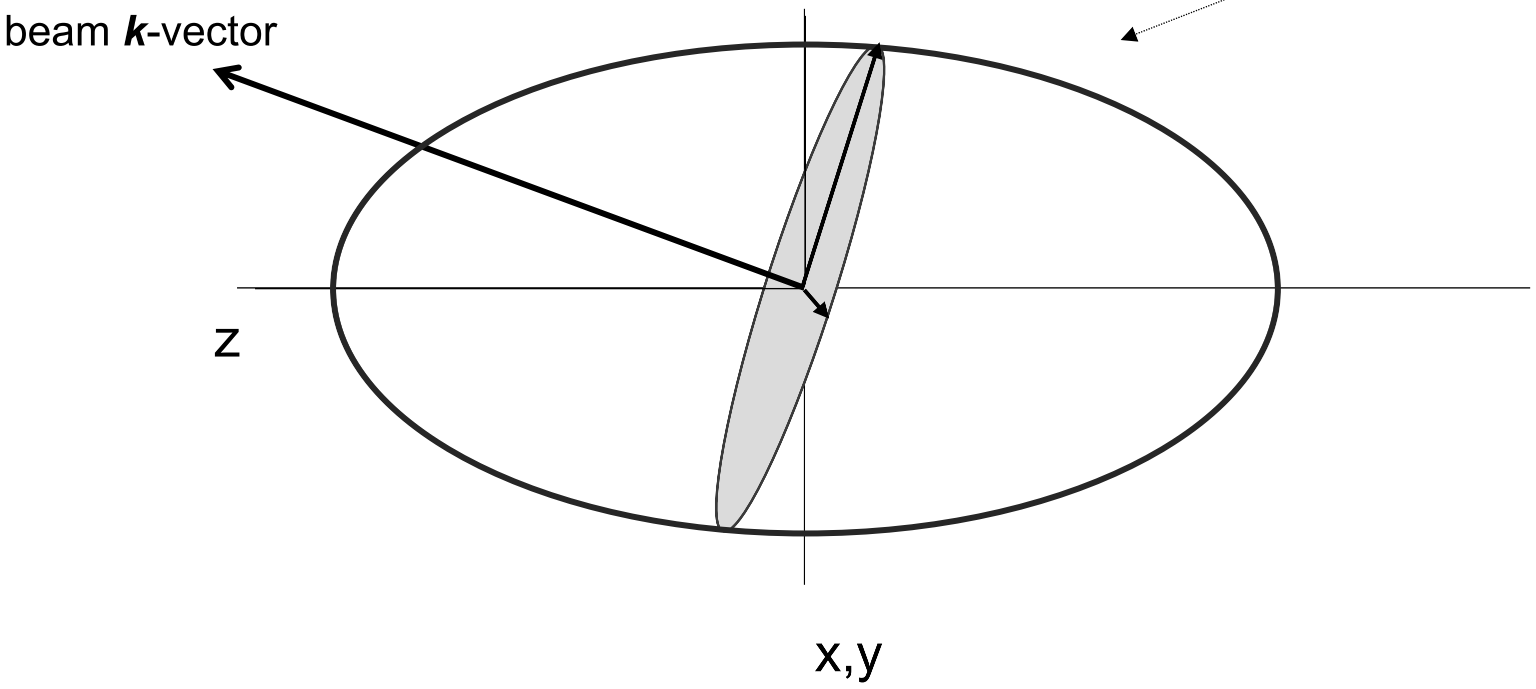
First-order electrooptic effect in anisotropic crystals

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

the index ellipsoid

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

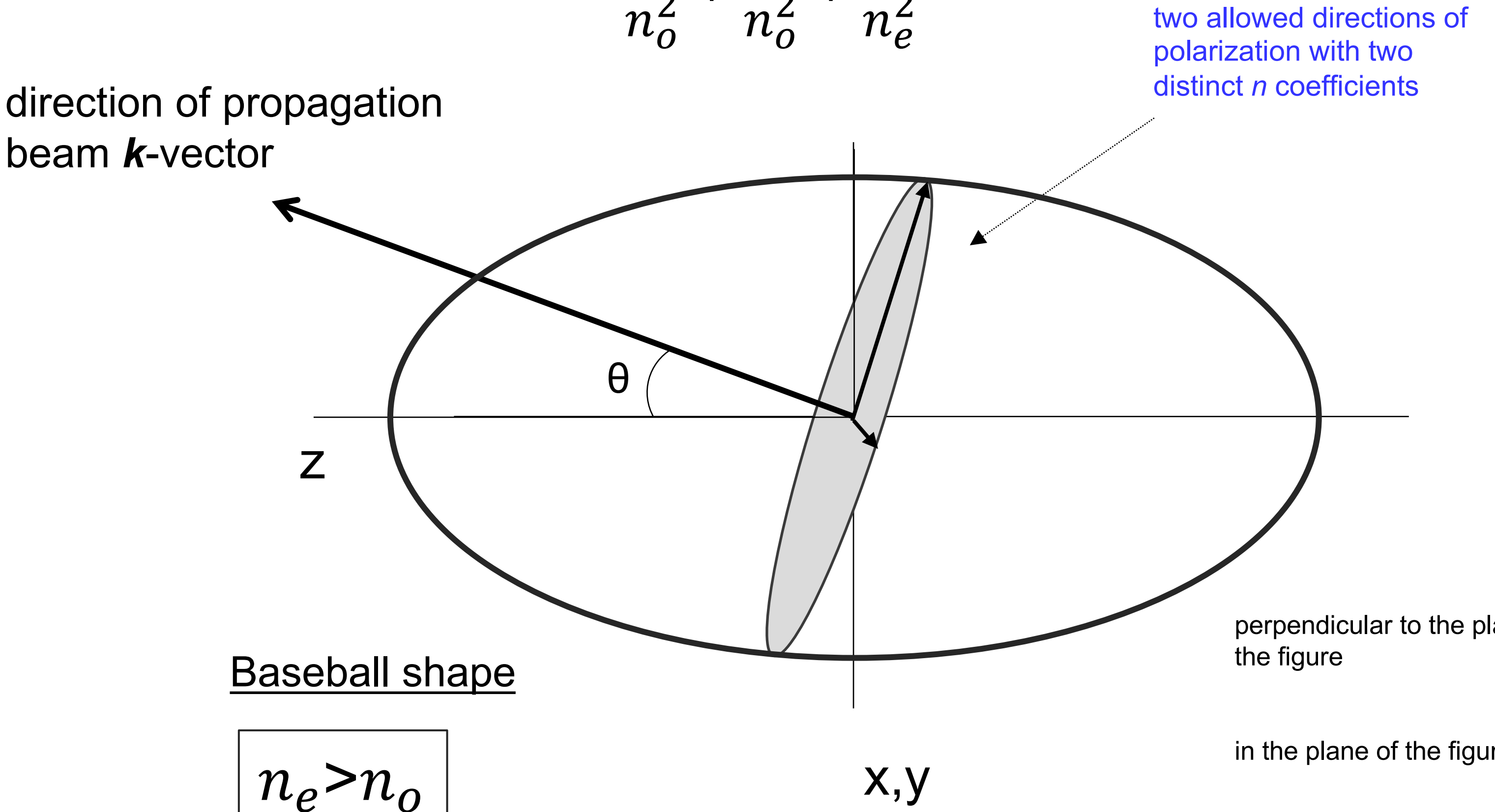
two allowed directions of polarization with two distinct *n* coefficients



First-order electrooptic effect in anisotropic crystals

Uniaxial crystal: $n_1=n_2=n_o;$ $n_3=n_e$

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



Baseball shape

$\boxed{n_e > n_o}$

'positive' crystal

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 θ to the optic axis, the index is given by:

perpendicular to the plane of the figure $\quad n = n_o$

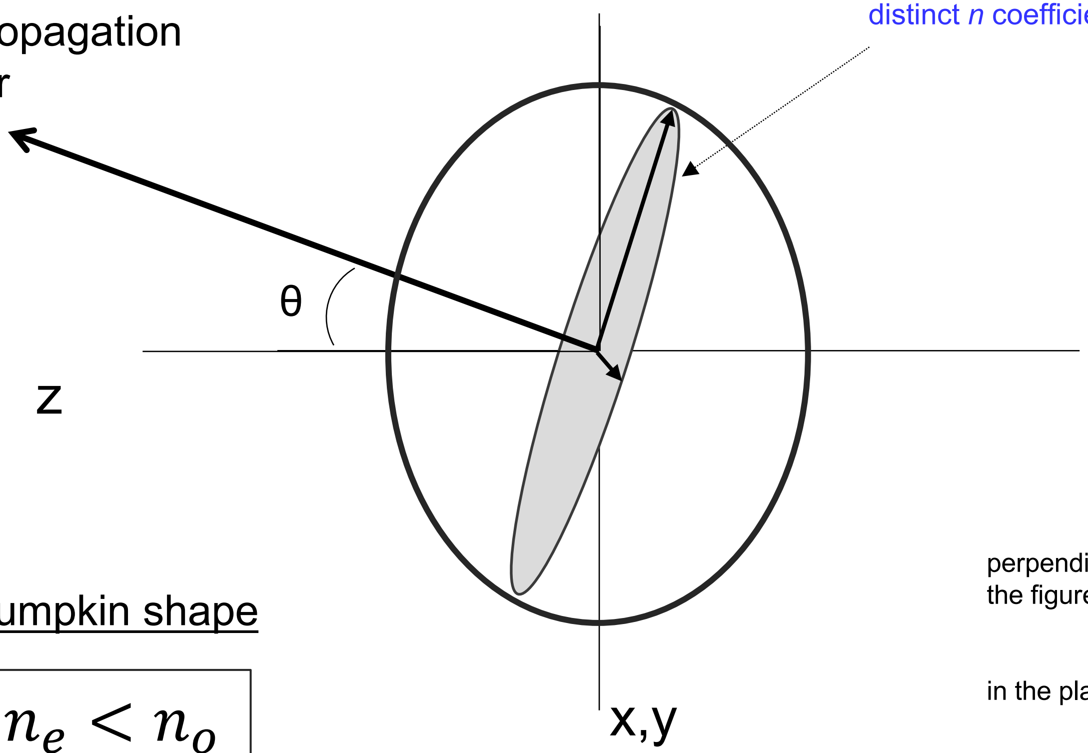
in the plane of the figure $\quad \frac{1}{n(\theta)^2}=\frac{cos(\theta)^2}{n_o^2}+\frac{\sin(\theta)^2}{n_e^2}$

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

First-order electrooptic effect in anisotropic crystals

Uniaxial crystal: $n_1=n_2=n_o$; $n_3=n_e$

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



Pumpkin shape

$$\boxed{n_e < n_o}$$

'negative' crystal

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 θ to the optic axis, the index is given by:

perpendicular to the plane of the figure $\quad n=n_o$

in the plane of the figure $\quad \dfrac{1}{n(\theta)^2}=\dfrac{cos(\theta)^2}{n_o^2}+\dfrac{\sin(\theta)^2}{n_e^2}$

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

First-order electrooptic effect in anisotropic crystals

the index ellipsoid

$$\frac{x^2}{n_1^2}+\frac{y^2}{n_2^2}+\frac{z^2}{n_3^2}=1 \quad \longleftrightarrow \quad \frac{X^2}{\epsilon_{XX}}+\frac{Y^2}{\epsilon_{YY}}+\frac{Z^2}{\epsilon_{ZZ}}=1.$$

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 the index ellipsoid

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

$$\begin{array}{ll} 1 & \text{for } ij=11, \\ 2 & \text{for } ij=22, \\ 3 & \text{for } ij=33, \\ 4 & \text{for } ij=23 \text{ or } 32, \\ 5 & \text{for } ij=13 \text{ or } 31, \\ 6 & \text{for } ij=12 \text{ or } 21. \end{array}$$

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

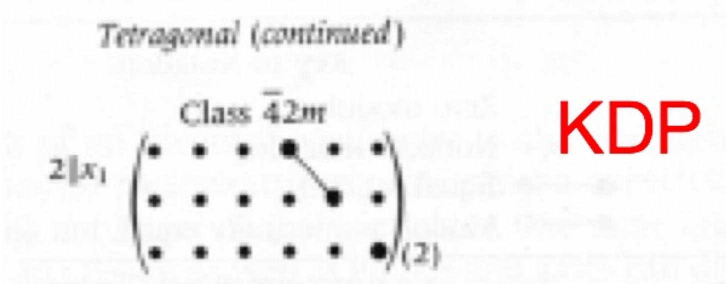
$$\begin{bmatrix} \Delta(1/n^2)_1 \\ \Delta(1/n^2)_2 \\ \Delta(1/n^2)_3 \\ \Delta(1/n^2)_4 \\ \Delta(1/n^2)_5 \\ \Delta(1/n^2)_6 \end{bmatrix} = \begin{bmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \\ r_{41} & r_{42} & r_{43} \\ r_{51} & r_{52} & r_{53} \\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

First-order electrooptic effect in anisotropic crystals

$$r_{ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \quad \text{(for class } \bar{4}2m\text{)},$$

KDP

$$\longleftrightarrow$$



KDP

$$\boxed{r_{ji} \longleftrightarrow d_{ij}}$$

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

Electrooptic modulator, KDP

Apply field along z-axis

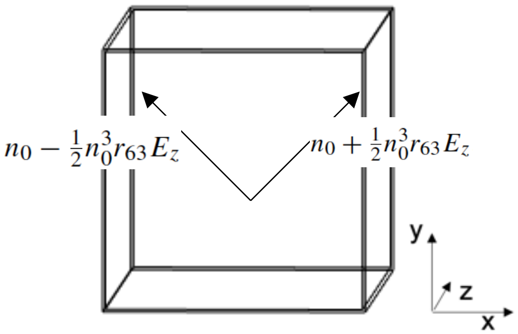
$$\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \longrightarrow \frac{x^2}{n_0^2} + \frac{y^2}{n_0^2} + 2xyr_{63}E_z = 1$$

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

rotate xy plane by 45º and get: $$\frac{x^2}{n_{x'}^2} + \frac{y^2}{n_{y'}^2} = 1$$

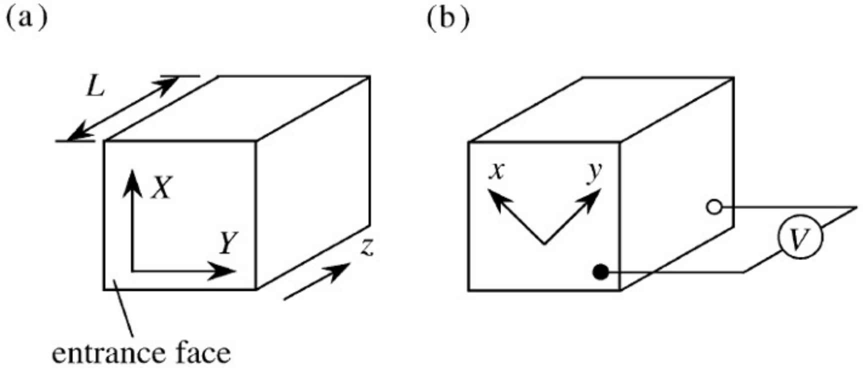
$$n_{x'} = \quad n_0 - \tfrac{1}{2} n_0^3 r_{63} E_z,$$

$$n_{y'} = \quad n_0 + \tfrac{1}{2} n_0^3 r_{63} E_z.$$

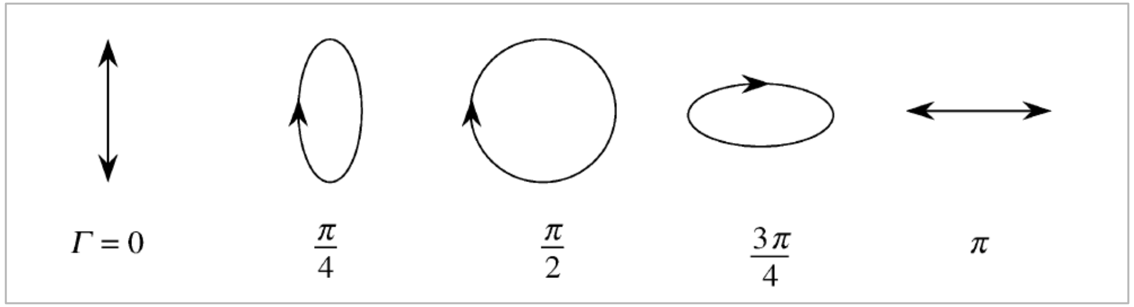


First-order electrooptic effect in anisotropic crystals

Electrooptic modulator, KDP



Evolution of the vertical polarization originally sent to the modulator



Γ - phase difference

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 $LiNbO_3$

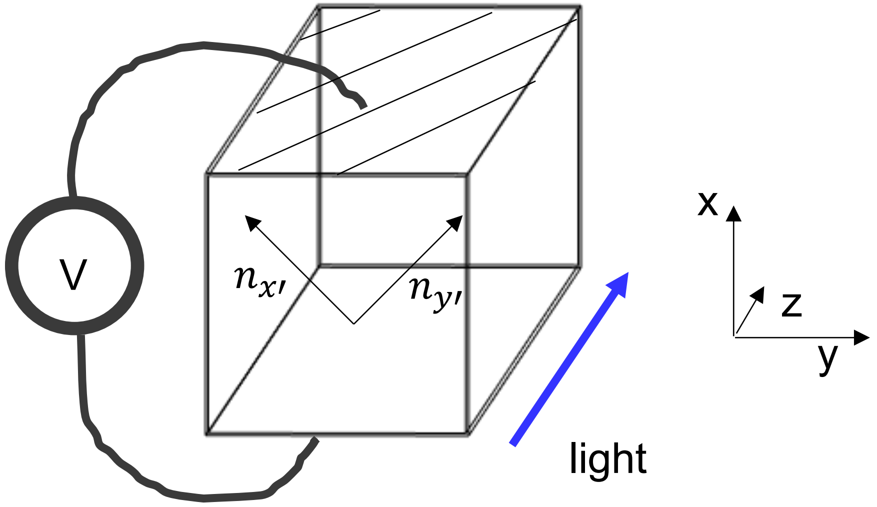
NLO tensor

$$d_{il} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

$$r_{ij} = \begin{bmatrix} 0 & -r_{22} & r_{13} \\ 0 & r_{22} & r_{13} \\ 0 & 0 & r_{33} \\ 0 & r_{42} & 0 \\ r_{42} & 0 & 0 \\ r_{22} & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

Apply field along <u>x-axis</u> (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

Electrooptic effect vs NLO effect

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.

SFG $\omega_3 = \omega_1 + \omega_2$

$\omega_1 \to 0, \qquad \omega_3 \to \omega_2$

EO $\omega \ = 0 + \omega$

may be orthogonal polarizations

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

$$E(t) = \underset{\text{DC}}{E_0} + \underset{\text{optical}}{E_\omega \cos(\omega t)}$$

(we take one-dimensional scalar form)

1) From NLO point of view

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

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

$P_\omega(t) = \varepsilon_0 4 d_{NL} E_0 E_\omega \cos(\omega t)$; but from $\boxed{P = \varepsilon_0 \chi E}$ $\rightarrow$ $P_\omega(t) = \varepsilon_0 \Delta\chi\, E_\omega \cos(\omega t)$; $\rightarrow$ $\Delta\chi = 4 d_{NL} E_0$

also $\chi = n^2 - 1$ $\longrightarrow$ $\Delta\chi = 2n\Delta n$

thus $4 d_{NL} E_0 = 2n\Delta n$ $\quad \Delta n = \dfrac{2 d_{NL} E_0}{n}$

2) From EO point of view

$$\Delta\left(\frac{1}{n^2}\right) = r_{EO} E_0 \longrightarrow \left(-\frac{2}{n^3}\right)\Delta n = r_{EO} E_0 \qquad \Delta n = -\frac{n^3}{2} r_{EO} E_0$$

$$\boxed{d_{NL} = -\frac{n^4}{4} r_{EO}} \xrightarrow[\text{tensor form}]{} \boxed{d_{ij} = -\frac{n^4}{4} r_{ji}}$$

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