

Nonlinear Dielectric **d**-Tensor

For second-order nonlinear susceptibilities in the Cartesian coordinate system:

$$\mathbf{P}^{NL}(\mathbf{E}^2) = \varepsilon_0 \chi^{(2)} \mathbf{E}^2$$

$$P_i^{NL} = \sum_{j,k} \varepsilon_0 \chi_{i,j,k}^{(2)} E_j E_k$$

$\chi_{i,j,k}^{(2)}$ in general has 27 independent coefficients (3x3x3=27 for a 3 dimensional tensor). Taking into account the permutation symmetry condition, the order of E_j and E_k in equation $P_i^{NL} = \sum_{j,k} \varepsilon_0 \chi_{i,j,k}^{(2)} E_j E_k$ is not important (i.e. $\chi_{i,j,k}^{(2)} = \chi_{i,k,j}^{(2)}$); the number of independent coefficients will be reduced to 18. For convenience, we can define a two dimensional (6x3) nonlinear dielectric tensor, commonly known as Kleinman **d**-tensor:

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

One obvious advantage of **d**-tensor format is that the full tensor can be written in a two-dimensional matrix; otherwise it would be difficult to express a three-dimensional tensor on a paper. With **d**-tensor, one can derive the strength of nonlinear interacting process in a dielectric medium under electrical field (**E**).

In practical situation, depending on the crystal class, many tensor components actually can be reduced to zero for crystal structure symmetry reason. For example, the **d**-tensor of LiNbO₃ (which is triangular 3m crystal class) can be expressed as

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_y E_z \\ 2E_x E_z \\ 2E_x E_y \end{pmatrix}$$

Where the nonlinear d coefficient is $d_{22} = 2.59 \text{ pm/V}$, $d_{31} = 4.85 \text{ pm/V}$, and $d_{33} = 25.3 \text{ pm/V}$. Thus, in the case of LiNbO_3 , when input fundamental wave $E_z(\omega)$ interacts with nonlinear dielectric medium along the selected z-axis through largest nonlinearity d_{33} , the induced polarization P_z can be expressed as

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ E_z^2 \\ 0 \\ 0 \\ 0 \end{pmatrix} = \varepsilon_0 \begin{pmatrix} 0 \\ 0 \\ d_{33} E_z^2 \\ 0 \\ 0 \end{pmatrix}$$

, or simply as $P_z(2\omega) = \varepsilon_0 d_{33} E_z(\omega)^2$.

In QPM wave mixing process, the effective d_{eff} will have a QPM reduction factor of $2/\pi$ in the d-tensor along the selected crystal orientation, i.e. $d_{eff} = \frac{2}{\pi} d$.