

1.7. NONLINEAR OPTICAL PROPERTIES

lographical frame by the standard conventions given in Chapter 1.6.

1.7.3. Propagation phenomena

1.7.3.1. Crystalline linear optical properties

We summarize here the main linear optical properties that govern the nonlinear propagation phenomena. The reader may refer to Chapter 1.6 for the basic equations.

1.7.3.1.1. Index surface and electric field vectors

The relations between the different field vectors relative to a propagating electromagnetic wave are obtained from the constitutive relations of the medium and from Maxwell equations.

In the case of a non-magnetic and non-conducting medium, Maxwell equations lead to the following wave propagation equation for the Fourier component at the circular frequency ω defined by (1.7.2.15) and (1.7.2.16) (Butcher & Cotter, 1990):

$$\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{E}(\omega) = (\omega^2/c^2)\mathbf{E}(\omega) + \omega^2\mu_0\mathbf{P}(\omega), \quad (1.7.3.1)$$

where $\omega = 2\pi c/\lambda$, λ is the wavelength and c is the velocity of light in a vacuum; μ_0 is the free-space permeability, $\mathbf{E}(\omega)$ is the electric field vector and $\mathbf{P}(\omega)$ is the polarization vector.

Table 1.7.2.3. Nonzero $\chi^{(2)}$ coefficients and equalities between them under the Kleinman symmetry assumption

Symmetry class	Independent nonzero $\chi^{(2)}$ elements under Kleinman symmetry
Triclinic C_1 (1)	$xxx, xyy = yxy = yyx, xzz = zxz = zzx,$ $xyz = xzy = yxz = yzx = zxy = zyx,$ $xxz = xzx = zxx, xxy = yxy = yxx, yyy,$ $yyz = zyz = zzy, yxz = yzx = zyx, zzz$
Monoclinic C_2 (2) (twofold axis parallel to z)	$xyz = xzy = yxz = yzx = zxy = zyx,$ $xxz = xzx = zxx, yyz = yzy = zyy, zzz$
C_s (m) (mirror perpendicular to z)	$xxx, xyy = yxy = yyx, xzz = zxz = zzx,$ $xyx = yxx = yxx, yyy, yxz = yzx = zyx$
Orthorhombic C_{2v} ($mm2$) (twofold axis parallel to z)	$xzx = xxz = zxx, yyz = yzy = zyy, zzz$
D_2 (222)	$xyz = xzy = yxz = yzx = zxy = zyx$
Tetragonal C_4 (4)	$xzx = xxz = zxx = yzy = yyz = zyy, zzz$
S_4 (4)	$xyz = xzy = yxz = yzx = zxy = zyx, xzx = xxz = zxx = -yzy = -yyz = -zyy$
D_4 (422)	All elements are nil
C_{4v} ($4mm$)	$xzx = xxz = zxx = yyz = yzy = zyy, zzz$
D_{2d} ($42m$)	$xyz = xzy = yxz = yzx = zxy = zyx$
Hexagonal C_6 (6)	$xzx = xxz = zxx = yyz = yzy = zyy, zzz$
C_{3h} (6)	$xxx = -xyy = -yyx = -yyx, yyy = -yxx = -xyx = -xxy$
D_6 (622)	All elements are nil
C_{6v} ($6mm$)	$xzx = xxz = zxx = yyz = yzy = zyy, zzz$
D_{3h} ($62m$) (mirror perpendicular to x)	$yyy = -yxx = -xxy = -xyx$
Trigonal C_3 (3)	$xxx = -xyy = -yyx = -yxy, xzx = xxz = zxx = yyz = yzy = zyy, yyy = -yxx = -xxy = -xyx, zzz$
D_3 (32)	$xxx = -xyy = -yyx = -yxy$
C_{3v} ($3m$) (mirror perpendicular to x)	$yyy = -yxx = -xxy = -xyx, xzx = xxz = zxx = yyz = yzy = zyy, zzz$
Cubic T (23), T_d ($\bar{4}3m$)	$xyz = xzy = yxz = yzx = zxy = zyx$
O (432)	All elements are nil

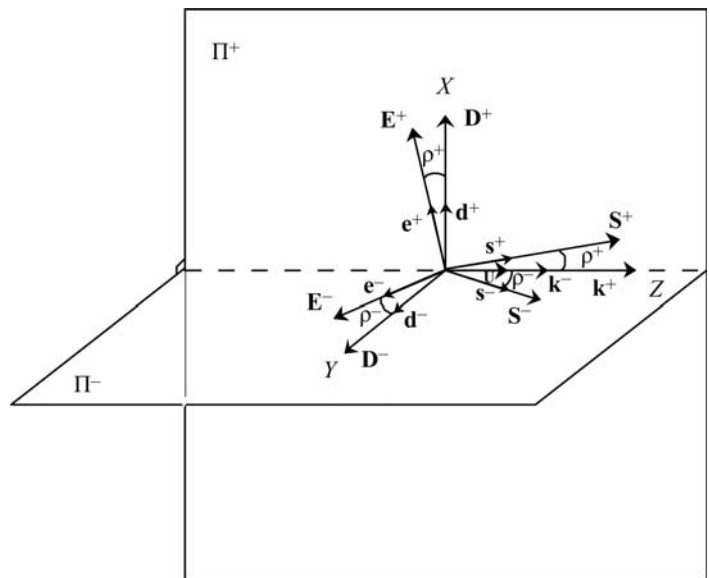


Fig. 1.7.3.1. Field vectors of a plane wave propagating in an anisotropic medium. (X, Y, Z) is the wave frame. Z is along the direction of propagation, X and Y are contained in Π^+ and Π^- respectively, by an arbitrary convention.

In the linear regime, $\mathbf{P}(\omega) = \epsilon_0\chi^{(1)}(\omega)\mathbf{E}(\omega)$, where ϵ_0 is the free-space permittivity and $\chi^{(1)}(\omega)$ is the first-order electric susceptibility tensor. Then (1.7.3.1) becomes

$$\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{E}(\omega) = (\omega^2/c^2)\epsilon(\omega)\mathbf{E}(\omega). \quad (1.7.3.2)$$

$\epsilon(\omega) = 1 + \chi^{(1)}(\omega)$ is the dielectric tensor. In the general case, $\chi^{(1)}(\omega)$ is a complex quantity *i.e.* $\chi^{(1)} = \chi^{(1)'} + i\chi^{(1)''}$. For the following, we consider a medium for which the losses are small ($\chi^{(1)'} \gg \chi^{(1)''}$); it is one of the necessary characteristics of an efficient nonlinear medium. In this case, the dielectric tensor is real: $\epsilon = 1 + \chi^{(1)'}$.

The plane wave is a solution of equation (1.7.3.2):

$$\mathbf{E}(\omega, X, Y, Z) = \mathbf{e}(\omega)\mathbf{E}(\omega, X, Y, Z) \exp[\pm ik(\omega)Z]. \quad (1.7.3.3)$$

(X, Y, Z) is the orthonormal frame linked to the wave, where Z is along the direction of propagation.

We consider a linearly polarized wave so that the unit vector \mathbf{e} of the electric field is real ($\mathbf{e} = \mathbf{e}^*$), contained in the XZ or YZ planes.

$\mathbf{E}(\omega, X, Y, Z) = A(\omega, X, Y, Z) \exp[i\Phi(\omega, Z)]$ is the scalar complex amplitude of the electric field where $\Phi(\omega, Z)$ is the phase, and $\mathbf{E}^*(-\omega, X, Y, Z) = \mathbf{E}(\omega, X, Y, Z)$. In the linear regime, the amplitude of the electric field varies with Z only if there is absorption.

k is the modulus of the wavevector, real in a lossless medium: $+kZ$ corresponds to forward propagation along Z , and $-kZ$ to backward propagation. We consider that the plane wave propagates in an anisotropic medium, so there are two possible wavevectors, \mathbf{k}^+ and \mathbf{k}^- , for a given direction of propagation of unit vector \mathbf{u} :

$$\mathbf{k}^{\pm}(\omega, \theta, \varphi) = (\omega/c)n^{\pm}(\omega, \theta, \varphi)\mathbf{u}(\theta, \varphi). \quad (1.7.3.4)$$

(θ, φ) are the spherical coordinates of the direction of the unit wavevector \mathbf{u} in the optical frame; (x, y, z) is the optical frame defined in Section 1.7.2.

The spherical coordinates are related to the Cartesian coordinates (u_x, u_y, u_z) by

$$u_x = \cos \varphi \sin \theta \quad u_y = \sin \varphi \sin \theta \quad u_z = \cos \theta. \quad (1.7.3.5)$$