

1. TENSORIAL ASPECTS OF PHYSICAL 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  $\omega$  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  $\omega = 2\pi c/\lambda$ ,  $\lambda$  is the wavelength and  $c$  is the velocity of light in a vacuum;  $\mu_0$  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)	$xxx, xyy = yxy = yyx, xzz = zxx = zzx,$ $xyz = xzy = yxz = yzx = zxy = zyx,$ $xxz = xzx = zxx, xxy = xyx = yxx, yyy,$ $yyz = zyz = zzy, yxz = yzx = zyx, zzz$
Monoclinic $C_2$ (2) (twofold axis parallel to $z$ ) $C_s$ ( $m$ ) (mirror perpendicular to $z$ )	$xyz = xzy = yxz = yzx = zxy = zyx,$ $xxz = xzx = zxx, yyz = yzy = zyy, zzz$ $xxx, xyy = yxy = yyx, xzz = zxx = zzx,$ $xyx = yxx = yxx, yyy, yxz = yzx = zyx = zzy$
Orthorhombic $C_{2v}$ ( $mm2$ ) (twofold axis parallel to $z$ ) $D_2$ ( $222$ )	$xzx = xxz = zxx, yyz = yzy = zyy, zzz$ $xyz = xzy = yxz = yzx = zxy = zyx$
Tetragonal $C_4$ (4) $S_4$ (4) $D_4$ (422) $C_{4v}$ ( $4mm$ ) $D_{2d}$ ( $42m$ )	$xzx = xxz = zxx = yzy = yyz = zyy, zzz$ $xyz = xzy = yxz = yzx = zxy = zyx, xzx =$ $xxz = zxx = -yzy = -yzy = -zyy$ All elements are nil $xzx = xxz = zxx = yyz = yzy = zyy, zzz$ $xyz = xzy = yxz = yzx = zxy = zyx$
Hexagonal $C_6$ (6) $C_{3h}$ (6) $D_6$ (622) $C_{6v}$ ( $6mm$ ) $D_{3h}$ ( $62m$ ) (mirror perpendicular to $x$ )	$xzx = xxz = zxx = yyz = yzy = zyy, zzz$ $xxx = -xyy = -yxy = -yyx, yyy = -yxx = -xyx = -xxy$ All elements are nil $xzx = xxz = zxx = yyz = yzy = zyy, zzz$ $yyy = -yxx = -xxy = -xyx$
Trigonal $C_3$ (3) $D_3$ (32) $C_{3v}$ ( $3m$ ) (mirror perpendicular to $x$ )	$xxx = -xyy = -yyx = -yxy, xzx = xxz = zxx = yyz = yzy = zyy, yyy = -yxx = -xxy = -xyx, zzz$ $xxx = -xyy = -yyx = -yxy$ $yyy = -yxx = -xxy = -xyx, xzx = xxz = zxx = yyz = yzy = zyy, zzz$
Cubic $T$ (23), $T_d$ ( $\bar{4}3m$ ) $O$ (432)	$xyz = xzy = yxz = yzx = zxy = zyx$ All elements are nil

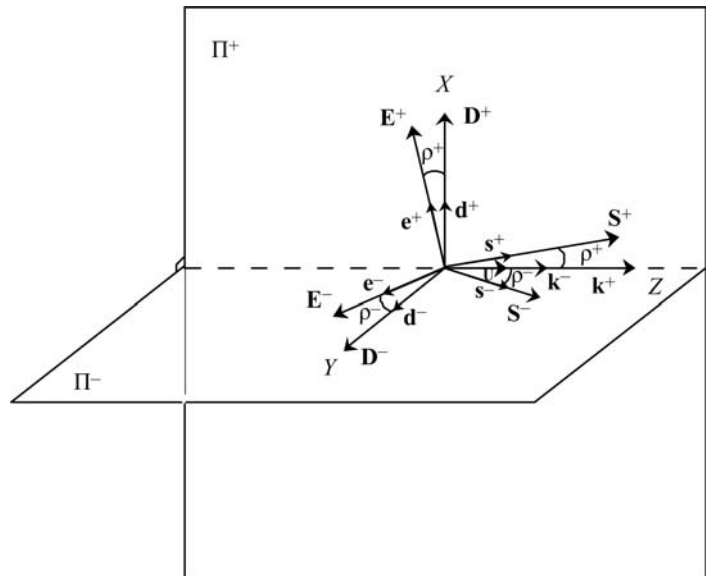


Fig. 1.7.3.1. Field vectors of a plane wave propagating in an anisotropic medium.  $(X, Y, Z)$  is the wave frame.  $Z$  is along the direction of propagation,  $X$  and  $Y$  are contained in  $\Pi^+$  and  $\Pi^-$  respectively, by an arbitrary convention.

In the linear regime,  $\mathbf{P}(\omega) = \epsilon_0\chi^{(1)}(\omega)\mathbf{E}(\omega)$ , where  $\epsilon_0$  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  $\mathbf{u}$ :

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

$(\theta, \varphi)$  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

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

## 1.7. NONLINEAR OPTICAL PROPERTIES

The refractive indices  $n^\pm(\omega, \theta, \varphi) = [\varepsilon^\pm(\omega, \theta, \varphi)]^{1/2}$ , ( $n^+ > n^-$ ), real in the case of a lossless medium, are the two solutions of the Fresnel equation (Yao & Fahlen, 1984):

$$n^\pm = \left[ \frac{2}{-B \mp (B^2 - 4C)^{1/2}} \right]^{1/2}$$

$$B = -u_x^2(b+c) - u_y^2(a+c) - u_z^2(a+b)$$

$$C = u_x^2bc + u_y^2ac + u_z^2ab$$

$$a = n_x^{-2}(\omega), \quad b = n_y^{-2}(\omega), \quad c = n_z^{-2}(\omega).$$
(1.7.3.6)

$n_x(\omega)$ ,  $n_y(\omega)$  and  $n_z(\omega)$  are the principal refractive indices of the index ellipsoid at the circular frequency  $\omega$ .

Equation (1.7.3.6) describes a double-sheeted three-dimensional surface: for a direction of propagation  $\mathbf{u}$  the distances from the origin of the optical frame to the sheets (+) and (-) correspond to the roots  $n^+$  and  $n^-$ . This surface is called the index surface or the wavevector surface. The quantity  $(n^+ - n^-)$  or  $(n^- - n^+)$  is the birefringency. The waves (+) and (-) have the phase velocities  $c/n^+$  and  $c/n^-$ , respectively.

Equation (1.7.3.6) and its dispersion in frequency are often used in nonlinear optics, in particular for the calculation of the phase-matching directions which will be defined later. In the regions of transparency of the crystal, the frequency law is well described by a Sellmeier equation, which is the case for normal dispersion where the refractive indices increase with frequency (Hadni, 1967):

$$n^\pm(\omega_i) < n^\pm(\omega_j) \quad \text{for } \omega_i < \omega_j, \quad (1.7.3.7)$$

If  $\omega_i$  or  $\omega_j$  are near an absorption peak, even weak,  $n^\pm(\omega_i)$  can be greater than  $n^\pm(\omega_j)$ ; this is called abnormal dispersion.

The dielectric displacements  $\mathbf{D}^\pm$ , the electric fields  $\mathbf{E}^\pm$ , the energy flux given by the Poynting vector  $\mathbf{S}^\pm = \mathbf{E}^\pm \times \mathbf{H}^\pm$  and the collinear wavevectors  $\mathbf{k}^\pm$  are coplanar and define the orthogonal vibration planes  $\Pi^\pm$  (Shuvalov, 1981). Because of anisotropy,  $\mathbf{k}^\pm$  and  $\mathbf{S}^\pm$ , and hence  $\mathbf{D}^\pm$  and  $\mathbf{E}^\pm$ , are non-collinear in the general case as shown in Fig. 1.7.3.1: the walk-off angles, also termed double-refraction angles,  $\rho^\pm = \arccos(\mathbf{d}^\pm \cdot \mathbf{e}^\pm) = \arccos(\mathbf{u} \cdot \mathbf{s}^\pm)$  are different in the general case;  $\mathbf{d}^\pm$ ,  $\mathbf{e}^\pm$ ,  $\mathbf{u}$  and  $\mathbf{s}^\pm$  are the unit vectors associated with  $\mathbf{D}^\pm$ ,  $\mathbf{E}^\pm$ ,  $\mathbf{k}^\pm$  and  $\mathbf{S}^\pm$ , respectively. We shall see later that the efficiency of a nonlinear interaction is strongly conditioned by  $\mathbf{k}$ ,  $\mathbf{E}$  and  $\rho$ , which only depend on  $\chi^{(1)}(\omega)$ , that is to say on the linear optical properties.

The directions  $\mathbf{S}^+$  and  $\mathbf{S}^-$  are the directions normal to the sheets (+) and (-) of the index surface at the points  $n^+$  and  $n^-$ .

For a plane wave, the time-average Poynting vector is (Yariv & Yeh, 2002)

$$\|\mathbf{S}^\pm(\omega)\| = \left\| \frac{1}{2} \text{Re}[\mathbf{E}^\pm(\omega) \times \mathbf{H}^{\pm*}(\omega)] \right\|$$

$$= \frac{1}{2} \frac{\|\mathbf{k}^\pm(\omega)\|}{\mu_0 \omega} \|\mathbf{E}^\pm(\omega)\|^2 \cos^2 \rho^\pm(\omega).$$
(1.7.3.8)

$\|\mathbf{S}^\pm\|$  is the energy flow  $I = \hbar\omega N^\pm$ , which is a power per unit area *i.e.* the intensity, where  $\hbar\omega$  is the energy of the photon and  $N^\pm$  are the photons flows.  $\rho^\pm(\omega)$  is the angle between  $\mathbf{S}^\pm$  and  $\mathbf{u}$ ; it is detailed later on.

The unit electric field vectors  $\mathbf{e}^+$  and  $\mathbf{e}^-$  are calculated from the propagation equation projected on the three axes of the optical

Table 1.7.2.4. Nonzero  $\chi^{(3)}$  coefficients and equalities between them in the general case

Symmetry class	$\chi^{(3)}$ nonzero elements
Triclinic $C_1 (1), C_i (\bar{1})$	All 81 elements are independent and nonzero
Monoclinic $C_s (m), C_2 (2), C_{2h} (\frac{2}{m})$ (twofold axis parallel to $z$ )	$xxxx, xyyy, xyzz, xzyz, xzzy, xxzz, xzxx, xzxx, xxyy, xyxy, xyxx, xxxy, xxyx, yxxx, yyyy, yvzz, yzyz, yzzy, yxzz, yzxx, yzzx, yxyy, yyxy, yyyx, yxxy, yxyx, yyxx, zzzz, zyyz, zyzy, zzyy, zxxz, zxzx, zzxx, zxyz, zxzy, zyxz, zzxy, zzyx, zzyx$
Orthorhombic $C_{2v} (mm2), D_2 (222), D_{2h} (mmm)$ (twofold axis parallel to $z$ )	$xxxx, xxzz, xzxx, xzxx, xxyy, xyxy, xyxx, yyyy, yvzz, yzyz, yzzy, yxxy, yxyx, yxxx, zzzz, zyyz, zyzy, zzyy, zxxz, zxzx, zzxx$
Tetragonal $S_4 (\bar{4}), C_4 (4), C_{4h} (\frac{4}{m})$	$xxxx = yyyy, xyyy = -yxxx, xyzz = -yxzz, xzyz = -yzxz, xzzy = -yzzx, xxzz = yvzz, xzxx = yzyz, xzxx = yzyz, xxyy = yyxx, xyxy = yxyx, xyxx = yxxy, xxxy = -yyyx, xxyx = -yyxy, xyxx = -yxyy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx, zxyz = -zyxz, zxzy = -zyzx, zzyx = -zyyx$
$C_{4v} (4mm), D_{2d} (\bar{4}2m), D_4 (422), D_{4h} (\frac{4}{m}mm)$	$xxxx = yyyy, xxzz = yvzz, xzxx = yzyz, xzxx = yzyz, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx$
Hexagonal $C_{3h} (\bar{6}), C_6 (6), C_{6h} (\frac{6}{m})$	$xxxx = yyyy = xxyy + xyxy + xyxx, xyyy = xxyx + xxyx + xyxx = -yxxx, xyzz = -yxzz, xzyz = -yzxz, xzzy = -yzzx, xxzz = yvzz, xzxx = yzyz, xzxx = yzyz, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, xxxy = -yyyx, xxyx = -yyxy, xyxx = -yxyy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx, zxyz = -zyxz, zxzy = -zyzx, zzyx = -zyyx$
$C_{6v} (6mm), D_{3h} (\bar{6}2m), D_6 (622), D_{6h} (\frac{6}{m}mm)$	$xxxx = yyyy = xxyy + xyxy + xyxx, xxzz = yvzz, xzxx = yzyz, xzxx = yzyz, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx, zxyz = -zyxz, zxzy = -zyzx, zzyx = -zyyx$
Trigonal $C_3 (3), C_{3i} (\bar{3})$	$xxxx = yyyy = xxyy + xyxy + xyxx, xyyy = xxyx + xxyx + xyxx = -yxxx, xyzz = -yxzz, xzyz = -yzxz, xzzy = -yzzx, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, xyyx = yxxy, xxxy = -yyyx, xxyx = -yyxy, xyxx = -yxyy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx, zxyz = -zyxz, zxzy = -zyzx, zzyx = -zyyx$
$C_{3v} (3m), D_3 (32), D_{3d} (\bar{3}m)$ (mirror perpendicular to $x$ ) (twofold axis parallel to $x$ )	$xxxx = yyyy = xxyy + xyxy + xyxx, xxzz = yvzz, xzxx = yzyz, xzxx = yzyz, xxyy = yyxx, xyxy = yxyx, xyyx = yxxy, xxyx = yxxy, xxyx = yxxy, zzzz, zyyz = zxxz, zyzy = zxzx, zzyy = zzxx, zxyz = -zyxz, zxzy = -zyzx, zzyx = -zyyx$
Cubic $T (23), T_h (m\bar{3})$	$xxxx = yyyy = zzzz, xxzz = yyxx = zzyy, xzxx = yxyx = zyzy, xzxx = yxxy = zyyz, xxyy = yyzz = zzzx, xyxy = yzyz = zxxz, xyyx = yzzy = zxxx$
$T_d (\bar{4}3m), O (432), O_h (m\bar{3}m)$	$xxxx = yyyy = zzzz, xxzz = xxyy = yyzz = yyxx = zzyy = zzxx, xzxx = xyxy = yzyz = yxyx = zyzy = zxxz, xzxx = xyxx = yzzy = yxxx = zyyz = zxxx$

