2.3 Optics of Uniaxial Crystals

In *uniaxial* crystals a special direction exists called the *optic axis* (Z axis). The plane containing the Z axis and the wave vector k of the light wave is termed the *principal plane*. The light beam whose polarization (i.e., the direction of the vector E oscillations) is normal to the principal plane is called an *ordinary beam* or an *o-beam* (Fig. 2.2). The beam polarized in the principal plane is known as the *extraordinary beam* or *e-beam* (Fig. 2.3). The refractive index of the o-beam does not depend on the propagation direction, whereas for the e-beam it does. Thus, the refractive index in anisotropic crystals generally depends both on light polarization and propagation direction.
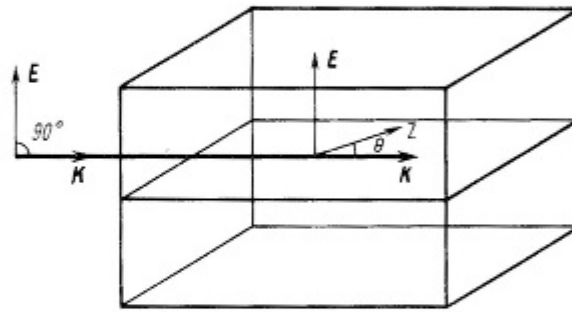

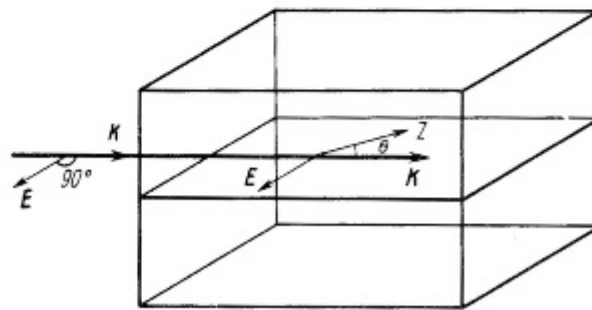**Fig. 2.2.** Principal plane of the crystal (kZ) and ordinary beam

Fig. 2.3. Principal plane of the crystal (kZ) and extraordinary beam

The difference between the refractive indices of the ordinary and extraordinary beams is known as *birefringence* Δn. The value of Δn is zero along the optic axis Z and maximum in the direction normal to this axis. The refractive indices of the ordinary and extraordinary beams in the plane normal to the Z axis are termed the *principal values* and are denoted by n_o and n_e, respectively. The refractive index of the extraordinary wave is, in general, a function of the polar angle θ between the Z axis and the vector k (Fig. 2.4). It is determined by the equation (index e in this case is written as a superscript):

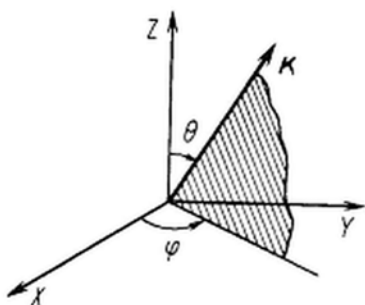$$n^e(\theta) = n_o [(1 + \tan^2 \theta)/(1 + (n_o/n_e)^2 \tan^2 \theta)]^{1/2} . \tag{2.14}$$

Fig. 2.4. Polar coordinate system for description of refractive properties of uniaxial crystal (k is the light propagation direction, Z is the optic axis, θ and φ are the coordinate angles)

The following equations are evident:

$$n^o(\theta) \equiv n_o , \tag{2.15}$$

$$n^e(0°) = n_o , \tag{2.16}$$

$$n^e(90°) = n_e , \tag{2.17}$$

$$\Delta n(0°) = 0 , \tag{2.18}$$

$$\Delta n(90°) = n_o - n_e , \tag{2.19}$$

$$\Delta n(\theta) = n_o - n^e(\theta) . \tag{2.20}$$

If $n_o > n_e$, the crystal is *negative*; if $n_o < n_e$, it is *positive*. The quantity n^e does not depend on the azimuthal angle φ (the angle between the projection of k onto the XY plane perpendicular to the Z axis and the X axis – see Fig. 2.4. The indicatrix of the refractive indices is a sphere with radius n_o for an ordinary beam and an ellipsoid of rotation with semiaxes n_o and n_e for an extraordinary beam (the axis of the ellipsoid of rotation is the Z axis). In the Z-axis direction the sphere and ellipsoid are in contact with each other. In a negative crystal the ellipsoid is inscribed in the sphere (Fig. 2.5a), whereas in a positive crystal the sphere is inscribed in the ellipsoid (Fig. 2.5b).

When a plane light wave propagates in a uniaxial crystal, the direction of propagation of the wave phase (vector k) generally does not coincide with that
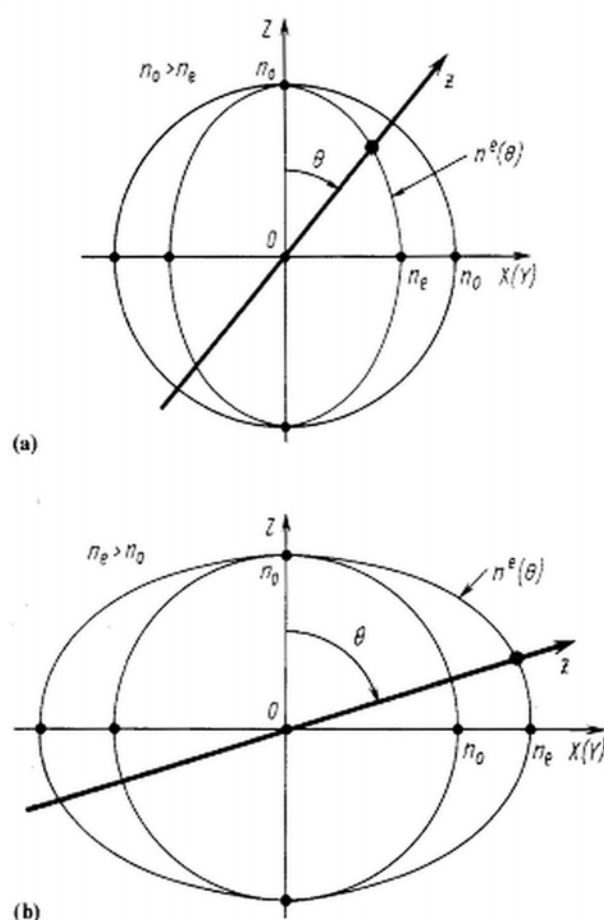
Fig. 2.5. Indicatrices of the refractive indices for ordinary and extraordinary waves in negative (a) and positive (b) uniaxial crystals

of the wave energy (vector s). The direction of s can be defined as the normal to the tangent drawn at the point of intersection of vector k with the $n(\theta)$ curve. For an ordinary wave the $n(\theta)$ dependence is a sphere with radius n_o. Therefore, the normal to the tangent coincides with the wave vector k. For an extraordinary wave the normal to the tangent (with the exception of the cases $\theta = 0°$ and $\theta = 90°$) does not coincide with the wave vector k but is rotated from it by the *birefringence angle* (Fig. 2.6):

$$\rho(\theta) = \pm \arctan[(n_o/n_e)^2 \cdot \tan \theta] \mp \theta , \qquad (2.21)$$

where the upper signs refer to a negative crystal and the lower signs to a positive one.
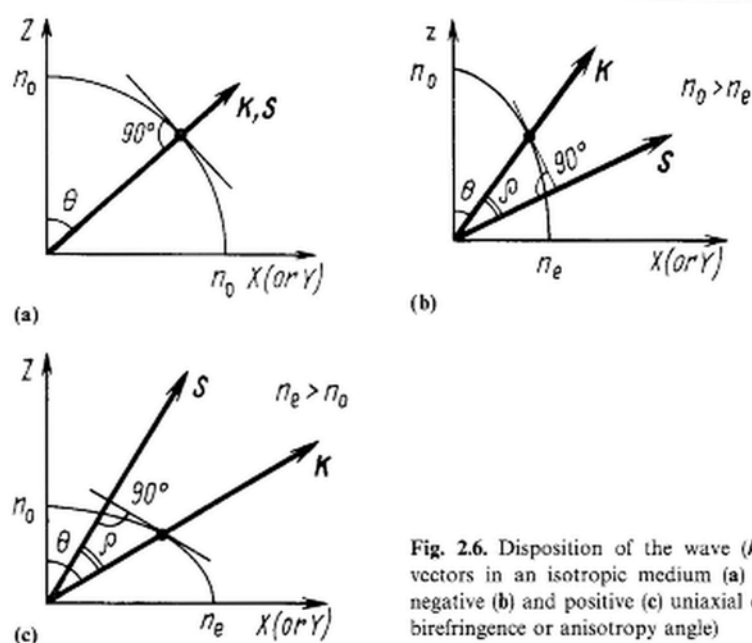
Fig. 2.6. Disposition of the wave (k) and beam (s) vectors in an isotropic medium (a) and anisotropic negative (b) and positive (c) uniaxial crystals (ρ is the birefringence or anisotropy angle)

The correlation between ρ and θ may serve as the basis for a simple way to orient uniaxial single crystals [2.5]. Let a laser beam with an arbitrary linear polarization fall on the input face of a crystal of thickness L. After passing through the crystal, the beam is divided into two orthogonally polarized beams that, at the output face of the crystal, are separated by (Fig. 2.7)

$$\delta = L \tan\rho . \tag{2.22}$$

The *crystal cut angle* θ_c, the angle between the optic axis Z of the crystal and the

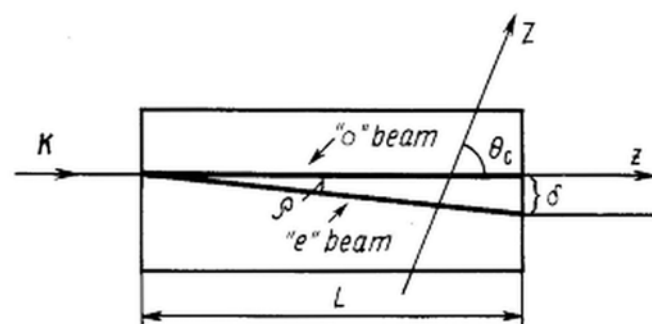Fig. 2.7. Calculation of the cut angle θ_c in a uniaxial crystal

normal to the crystal surface, corresponds to one of two values

$$\theta_c = \arctan \left\{ \frac{|(n_o/n_e)^2 - 1| \cdot L}{2\delta(n_o/n_e)^2} \pm \sqrt{\frac{[(n_o/n_e)^2 - 1]^2 \cdot L^2}{4\delta^2(n_o/n_e)^4} - (n_o/n_e)^2} \right\}. \quad (2.23)$$

A more rigorous consideration of crystal optics of anisotropic media is given in [2.6].

2.6 Reflection and Refraction of Light Waves at the Surfaces of Uniaxial Crystals

Reflection and refraction of light waves at the vacuum–dielectric interface must be taken into account. Therefore, we shall give the equations for the refraction angles and for the reflection coefficients for different incidence angles and polarizations of the light wave incident on the plane surface of a uniaxial nonlinear crystal. In all cases the reflection angles are equal to the incidence angles.

1) *The E vector is perpendicular to the principal plane, α is the incidence angle, ψ_o is the refraction angle (Fig. 2.11a): ψ_o can be found from*

$$\sin \alpha = n_o \sin \psi_o \, ; \tag{2.32}$$

the reflection coefficient is

$$R^o = \frac{\sin^2 (\alpha - \psi_o)}{\sin^2 (\alpha + \psi_o)} \, . \tag{2.33}$$

For normal incidence ($\alpha = \psi_o = 0$)

$$R^o_{\alpha=0} = \frac{(n_o - 1)^2}{(n_o + 1)^2} \, . \tag{2.34}$$

2) *The E vector is in the main plane, α is the incident angle, ψ^e is the refraction angle (Fig. 2.11b,c): ψ^e can be found from*

$$\sin \alpha / \sin \psi^e = n^e (\theta) = n_o \sqrt{\frac{1 + \tan^2 (\theta_c \pm \psi^e)}{1 + (n_o/n_e)^2 \tan^2 (\theta_c \pm \psi^e)}} \, , \tag{2.35}$$

where θ_c is the cut angle of the crystal, and θ is the angle between the optic axis Z and vector k in the crystal. If the vector k and optic axis Z lie on different sides of the normal to the crystal surface (Fig. 2.11b), the plus sign is used in (2.23). When the vector k and optic axis Z are on the same side of the normal to the crystal surface (Fig. 2.11c), the minus sign is used.
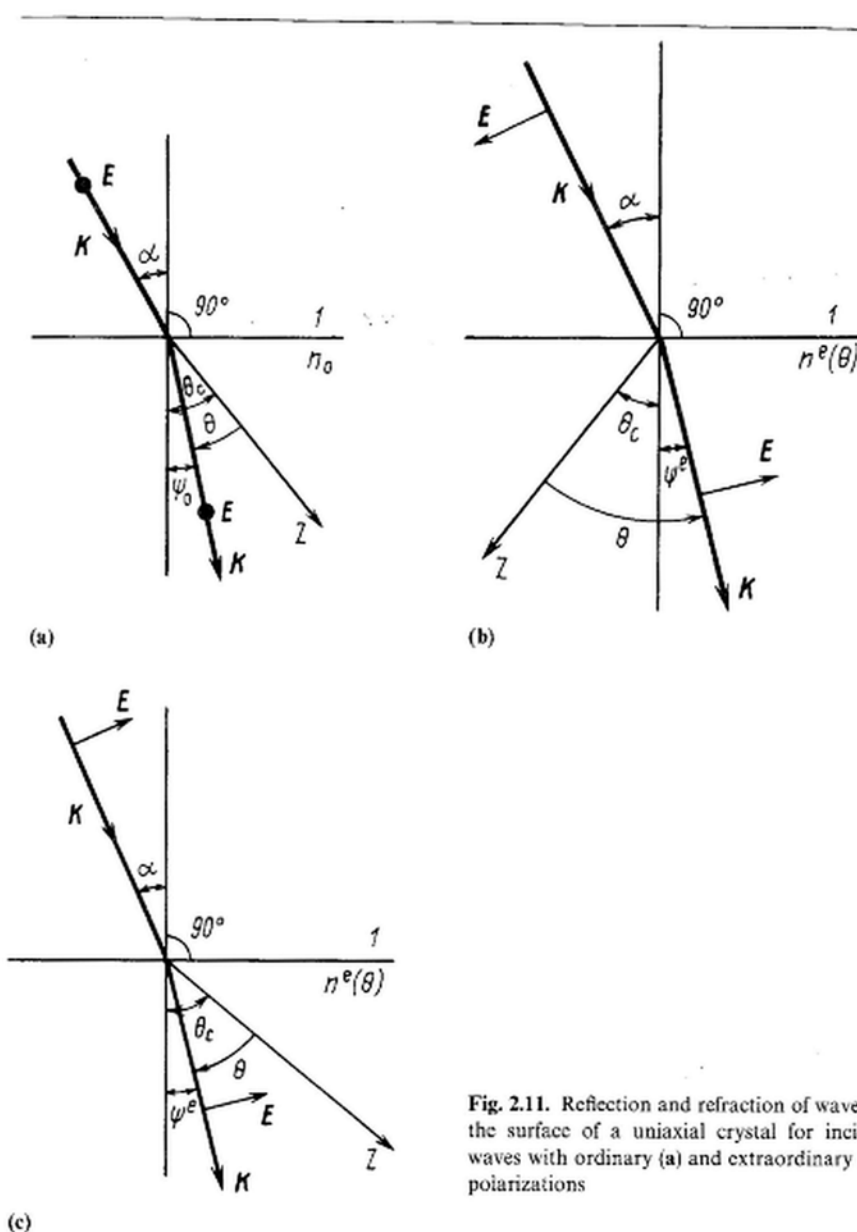
The reflection coefficient is

$$R^e = \frac{\tan^2 (\alpha - \psi^e)}{\tan^2 (\alpha + \psi^e)} \, . \tag{2.36}$$

For normal incidence ($\alpha = \psi^e = 0$)

$$R^e_{\alpha=0} = \frac{(n^e (\theta) - 1)^2}{(n^e (\theta) + 1)^2} \, , \tag{2.37}$$

where

$$n^e (\theta) = n^e (\theta_c) = n_o \sqrt{\frac{1 + \tan^2 \theta_c}{1 + (n_o/n_e)^2 \tan^2 \theta_c}} \, . \tag{2.38}$$

(a)

(b)

(c)

Fig. 2.11. Reflection and refraction of waves on the surface of a uniaxial crystal for incident waves with ordinary (a) and extraordinary (b,c) polarizations

Note that the reflection coefficients on the input and output surfaces of the uniaxial crystal are identical, as with an isotropic dielectric.

The equations given here can be used for calculating the external rotation crystal angles necessary for angular tuning of the phase-matching conditions and for evaluating the reflection losses.

NONLINEAR OPTICS

2.10 Crystal Symmetry and Effective Nonlinearity: Uniaxial Crystals

For anisotropic media the dielectric susceptibility coefficients κ_0 and $\chi^{(2)}$ in (2.2), are in general case the tensors of the second and third ranks, respectively. Below we shall consider the uniaxial crystals. In dielectric reference frame X, Y, Z, where Z is the optic axis, the tensors κ_0 and ε_0 are diagonal. The following components:

$$\varepsilon_{0XX} = \varepsilon_{0YY} = n_0^2 \; ;$$
$$\varepsilon_{0ZZ} = n_e^2 \; ; \tag{2.55}$$

are nonzero components of the linear dielectric polarization tensor ε_0. In practice the tensor d_{ijk} is used instead of tensor χ_{ijk}, the two tensors being interrelated by the equation

$$\chi_{ijk} = 2d_{ijk} \; . \tag{2.56}$$

Unlike tensor ε_0, tensors χ and d can be given only in a three dimensional representation. Usually a "plane" representation of tensor d_{ijk} in the form d_{il} is used, where $i = 1$ corresponds to (X), $i = 2$ to (Y), $i = 3$ to (Z), and l takes the following values:

$$
\begin{array}{cccccc}
XX & YY & ZZ & YZ = ZY & XZ = ZX & XY = YX \\
l = 1 & 2 & 3 & 4 & 5 & 6
\end{array} \tag{2.57}
$$

The expression (2.2) can be rewritten in a reduced form (with respect to the components):

$$P_i = \kappa_{0ik}E_k + 2d_{il}E_l^2 + \ldots , \tag{2.58}$$

where E_j^2 is the six-dimensional vector of the field products (summation over the repeating indices is carried out). For SHG in matrix form we have:

$$
\left[\begin{pmatrix} P_X \\ P_Y \\ P_Z \end{pmatrix} \right] = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \cdot \begin{bmatrix} E_X^2 \\ E_Y^2 \\ E_Z^2 \\ 2E_Y E_Z \\ 2E_X E_Z \\ 2E_X E_Y \end{bmatrix} . \tag{2.59}
$$

The total number of the components of the square nonlinearity tensor d_{il} is 18. In centrosymmetrical crystals (where the center is a symmetry element) all the components of the square nonlinearity tensor d are equal to zero. The non-centrosymmetrical crystals comprising 21 crystallographic classes out of 32 usually have one or more symmetry elements (axes or planes of different orders), which considerably decrease the number of independent components of the tensor d_{il}.

Kleinman [2.15] has established additional *symmetry conditions* for the case of no dispersion of electron nonlinear polarizability. When the Kleinman symmetry conditions are valid (in the great majority of practical cases), the number of independent components of the tensor d_{il} decreases from 18 to 10, because

$$
\begin{aligned}
d_{21} &= d_{16}; & d_{24} &= d_{32}; & d_{31} &= d_{15}; \\
d_{13} &= d_{35}; & d_{12} &= d_{26}; & d_{32} &= d_{24}; \\
d_{14} &= d_{36} &= d_{25} \; .
\end{aligned} \tag{2.60}
$$

Since any linearly polarized wave in a uniaxial crystal can be represented as a superposition of two waves with "ordinary" and "extraordinary" polarizations, we provide the components of a unit polarization vector p given in polar coordinates θ and ϕ along the dielectric axes X, Y, Z, where Z is the optic axis and $|p| = 1$:

$$
\begin{aligned}
p_{oX} &= -\sin\phi, & p_X^e &= \cos\theta\cos\phi \\
p_{oY} &= \cos\phi, & p_Y^e &= \cos\theta\sin\phi \\
p_{oZ} &= 0, & p_Z^e &= -\sin\theta \; .
\end{aligned} \tag{2.61}
$$

The equations for calculating the conversion efficiency use the *effective nonlinearity* d_{eff}, which comprises all the summation operations along the polarization directions of the interacting waves:

$$
d_{\text{eff}} = p_1 d p_3 p_2 = p_2 d p_1 p_1 = p_3 d p_1 p_2 \tag{2.62}
$$

The quantity d_{eff} represents a scalar product of the first vector in (2.62) and a tensor-vector product of the $d\,pp$ type, which is also a vector. Depending on the type of interaction (ooe, oee, and so on), the vector components p_i are calculated by (2.61), and the product (2.62) is found by the known rules of vector

Table 2.3. Expressions for d_{eff} in uniaxial crystals of different point groups when Kleinman symmetry relations are valid

Point group	Type of interaction	
	ooe, oeo, eoo	eeo, eoe, oee
$\bar{4}2m(D_{2d})$	$d_{36}\sin\theta\sin2\phi$	$d_{36}\sin2\theta\cos2\phi$
$3m(C_{3v})$	$d_{31}\sin\theta - d_{22}\cos\theta\sin3\phi$	$d_{22}\cos^2\theta\cos3\phi$
$\left.\begin{array}{l}4(C_4)\\4mm(C_{4v})\\6(C_6)\\6mm(C_{6v})\end{array}\right\}$	$d_{31}\sin\theta$	0
$\bar{4}(S_4)$	$(d_{36}\sin2\phi + d_{31}\cos2\phi)\sin\theta$	$(d_{36}\cos2\phi - d_{31}\sin2\phi)\sin2\theta$
$3(C_3)$	$(d_{11}\cos3\phi - d_{22}\sin3\phi)\cos\theta + d_{31}\sin\theta$	$(d_{11}\sin3\phi + d_{22}\cos3\phi)\cos^2\theta$
$32(D_3)$	$d_{11}\cos\theta\cos3\phi$	$d_{11}\cos^2\theta\sin3\phi$
$\bar{6}(C_{3h})$	$(d_{11}\cos3\phi - d_{22}\sin3\phi)\cos\theta$	$(d_{11}\sin3\phi + d_{22}\cos3\phi)\cos^2\theta$
$\bar{6}m2(D_{3h})$	$d_{22}\cos\theta\sin3\phi$	$d_{22}\cos^2\theta\cos3\phi$
$422(D_4)$	0	0
$622(D_6)$	0	0

algebra. Table 2.3 illustrates the values of d_{eff} determined in this way for nonlinear uniaxial crystals of 13 point groups [2.3, 16].

The inclusion of the birefringence or "walk-off" angle (Fig. 2.6) leads to the change of the expressions for the *nonlinear coupling coefficients* (see below) and for the *effective nonlinearity*. Although the angle θ is defined as the angle between axis Z and light propagation direction z, the unit polarization vectors p_i are perpendicular to the direction of propagation of the wave energy s. Therefore, it is necessary to correct the expressions for components of the unit polarization vector p given by (2.61). The sign of *birefringence angle* ρ in these formulas will depend on "walk-off" direction, i.e., in the case of a uniaxial crystal, on the sign of the crystal (Fig. 2.6): for a negative crystal the angle ρ must be added to θ, for a positive crystal it must be subtracted from θ. So, in formulas (2.61) the angle θ must be changed for $(\theta + \rho)$ for the negative crystal and for $(\theta - \rho)$ for the positive one. Remember that the value ρ is the function of the angle θ for the uniaxial crystal (2.21). The dispersion of ρ should be also taken into account. Therefore, in (2.61), instead of θ, we have substitute $\theta \pm \rho(\omega, 2\omega)$, and the corresponding changes should be done also in expressions for d_{eff} (Table 2.3).