CN

Chapter 4

CT

Supplement: Anisotropic Media

In Section 2.4 of “CBGL,” we discuss how crystal birefringence can be used to achieve phasematching. Readers familiar with the behavior of anisotropic media will have understood the crystallographic and optical properties underlying that discussion. For those readers who might be less familiar with some of these concepts, this chapter of the online supplement will provide some of the needed background.

In Section 2.3 of “CBGL,” we stated that the linear change in electric displacement $\mathbf{D}$ due to an applied electric field $\mathbf{E}$ was given by $\mathbf{D} = \epsilon_o \mathbf{E} + \mathbf{P}_L$, where the linear polarization was given by $\mathbf{P}_L = \epsilon_o \chi^{(1)} \mathbf{E}$, so that we could also write $\mathbf{D} = \epsilon_o \epsilon^{(1)} \mathbf{E}$, where $\epsilon^{(1)} = 1 + \chi^{(1)}$. In general, the vector representing the polarization $\mathbf{P}_L$ may have an arbitrary direction and magnitude with respect to the applied electric field $\mathbf{E}$, so that the relationship between $\mathbf{P}_L$ and $\mathbf{E}$ must be written in some arbitrary $x - y - z$ coordinate system as:

$$\begin{bmatrix} P_{L,x} \\ P_{L,y} \\ P_{L,z} \end{bmatrix} = \epsilon_o \begin{bmatrix} \chi^{(1)}_{xx} & \chi^{(1)}_{xy} & \chi^{(1)}_{xz} \\ \chi^{(1)}_{yx} & \chi^{(1)}_{yy} & \chi^{(1)}_{yz} \\ \chi^{(1)}_{zx} & \chi^{(1)}_{zy} & \chi^{(1)}_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \tag{4.1}$$

However, it is possible to find a coordinate system in which this χ tensor, and the corresponding permittivity tensor $\epsilon^{(1)}$ become diagonal. In that system, called the “principal axis” system, we can write:

$$\epsilon^{(1)} = \begin{bmatrix} \epsilon^{(1)}_{xx} & 0 & 0 \\ 0 & \epsilon^{(1)}_{yy} & 0 \\ 0 & 0 & \epsilon^{(1)}_{zz} \end{bmatrix} \tag{4.2}$$

We will often express this tensor in a form that uses refractive indices instead of permittivity:

$$\epsilon^{(1)} = \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix} \tag{4.3}$$

C The Principal Axes and Crystal Symmetry

This permittivity tensor contains within it information about how the phase velocity of light waves in our nonlinear material varies with the direction of propagation and the polarization of the wave. We will see in the section below how to extract this information in order to find phasematching solutions for nonlinear interactions. However, at this point, it is worth taking a slight detour to look beyond the mathematical process of finding those phasematching solutions and consider what we will do with those solutions once we find them. In particular, we might anticipate that our phasematching solution will specify some direction in this principal axis coordinate system in which light must propagate for a phasematched nonlinear interaction, and ask ourselves the question "What do these principal axes have to do with a crystal in the real world? Given a chunk of nonlinear material sitting on my desk, how do I locate the propagation direction I have just determined mathematically?" Considering this question at this point also allows us to introduce the topic of crystal symmetry, which influences the form of the $\epsilon^{(1)}$ tensor given above.

In crystalline materials, the constituent atoms are arranged in a regular, repeating configuration. If we look at a tiled floor, we see that one individual tile is the smallest unit that can be replicated repeatedly in two dimensions to generate the pattern of the floor. In the same way, by examining the structure of the three-dimensional array of atoms in a crystal, we can determine the smallest possible building block that, by repeated replication in three dimensions, can generate the structure of the crystal. This "unit cell" may have one of seven basic shapes, as shown in Figure S-4-1. Inherent in each of these basic categories are certain symmetry properties that are exhibited by the unit cell. These symmetry properties tell how the cell can be reflected, rotated, and inverted to produce the same spatial arrangement of atoms. For example, rotating the trigonal unit cell by 120° about the axis indicated in Figure S-4-1 brings all the atoms in the cell to equivalent positions.

The edges of the unit cell define a set of directions, which we denote as a, b, and c. In some cases, such as crystals with an orthorhombic unit cell, these directions will be mutually orthogonal (as illustrated in Figure S-4-2 for the case of orthorhombic potassium niobate) and can be used to define a right-handed Cartesian coordinate system for the solution of Maxwell's equations. However, in other cases, such as the trigonal crystal lithium niobate, the edge directions are not mutually orthogonal. For these cases, there is a convention which establishes the correct orientation of a trio of

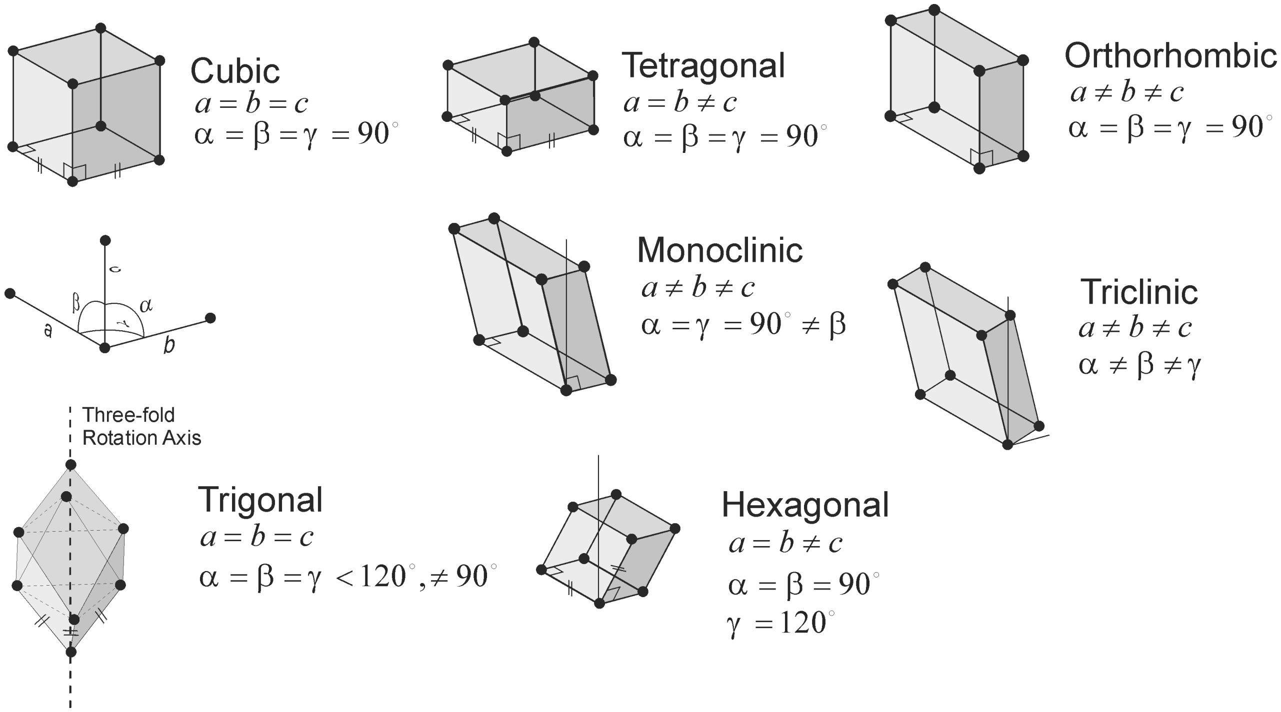

Figure 4.1: Figure S-4-1 Representations of the seven systems of unit cell types. The orientation of the trigonal cell is altered somewhat to highlight

Cartesian axes X, Y, and Z with respect to the unit cell geometry. For example, in trigonal crystals, two of the Cartesian axes are taken along orthogonal edges of the unit cell and the third is generated by requiring it to form a right-handed coordinate system with the other two.

This Cartesian axis system ($X-Y-Z$) is not necessarily the same as the principal axis system (x, y, and z) in which the permittivity tensor is diagonal. In most of the crystals commonly used for blue-green generation, the directions of the $x-y-z$ axes *will* coincide with those of the $X-Y-Z$ axes; however, for monoclinic and triclinic crystals, the $x-y-z$ and $X-Y-Z$ axes are not generally coincident—in fact, the orientation of the $x-y-z$ can be wavelength-dependent. Even in classes where the axes of the two systems lie along the same directions, the exact correspondence between the individual $a-b-c$, $X-Y-Z$ and $x-y-z$ axes has often been inconsistently stated in the literature, and this has led to confusion over the signs and designations of the nonlinear coefficients. This problem is particularly common for crystals with orthorhombic symmetry, a class that includes materials like potassium niobate (KN) and potassium titanyl phosphate (KTP) which are important for blue-green generation [Dimitriev and Nikogosyan (1993)]. For example, in KTP the usual correspondence is $x \rightarrow a$, $y \rightarrow b$, and $z \rightarrow c$, where a, b, and c can be uniquely distinguished from

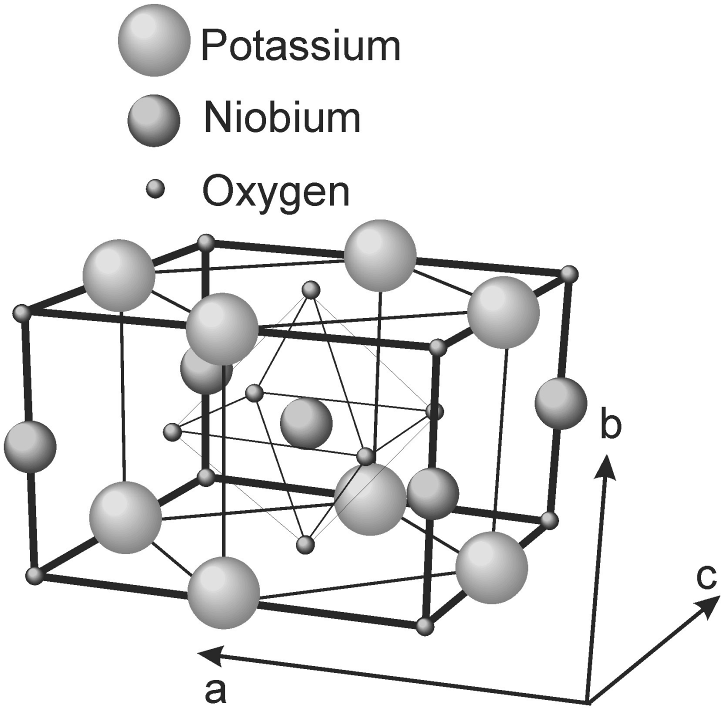

Figure 4.2: Figure S-4-2. Unit cell structure of orthorhombic potassium niobate

the geometry of the orthorhombic unit cell (a_o=12.814 Å, b_o=6.404 Å, c_o=10.616 Å). However, for KN the correspondence is $x \rightarrow b$, $y \rightarrow a$, and $z \rightarrow c$ (where the dimensions of the unit cell are a_o=5.6896 Å, b_o=3.9692 Å, c_o=5.7256 Å). Not only is the relationship between $x - y - z$ and $a - b - c$ different in these two cases, but the assignment of $a - b - c$ in terms of short, medium, and long unit cell dimensions differs.

This confusion over axis identification has arisen because the historical development of nonlinear optics has involved a confluence of several different disciplines: crystallography, mineralogy, electromagnetic theory, and classical optics, for example. Each of these disciplines has brought into the study of nonlinear optics certain conventions and traditions of that field. Roberts [Roberts (1992)] has recently reviewed this confusion and the attempts made to rectify it based on modifications of the longstanding "IEEE Standard 176 on Piezoelectricity." He advocates the adoption of some nomenclature for orthorhombic crystals that departs from the Standard but has become well-established in the literature. Thus, the reader should be aware that the literature contains a mixture of conventions and nomenclature (for example, n_a, n_b, and n_c are commonly used, rather than n_x, n_y, and n_z), and care must be taken as a result. In this book, we will tend to use the mostly designations most commonly found in literature related to blue-green generation, even though these may depart from the standard.

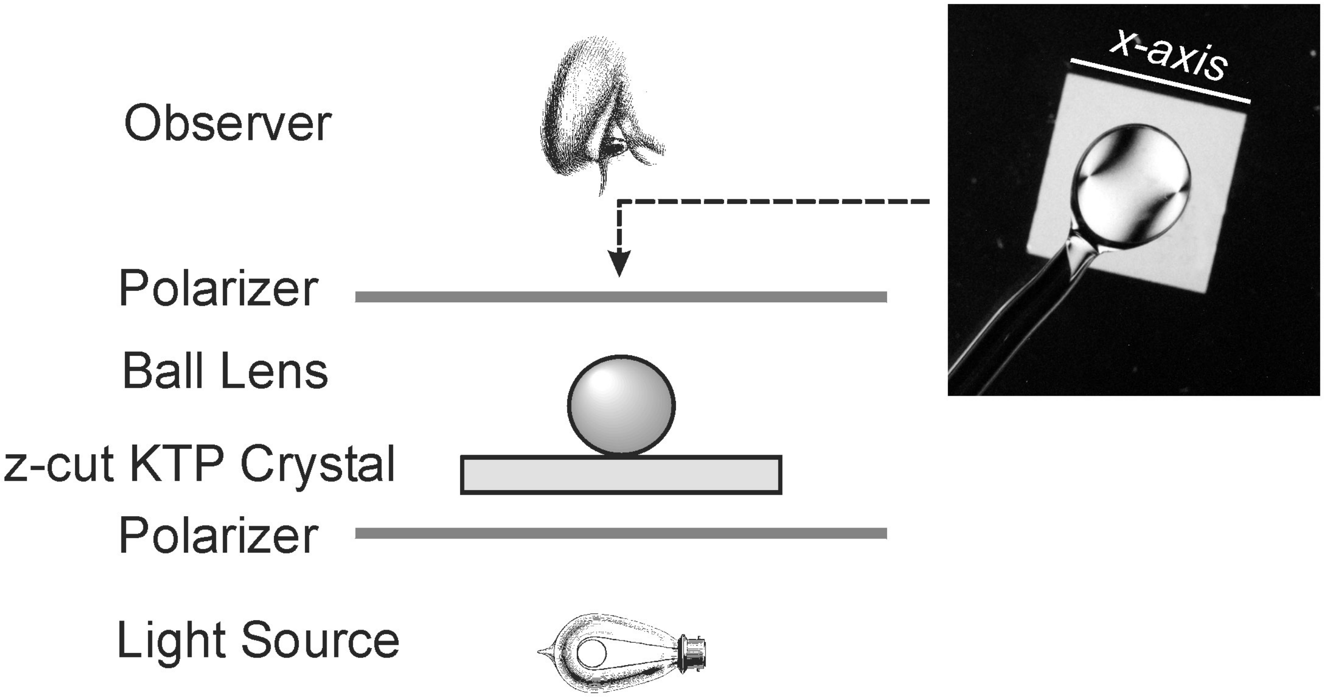

Figure 4.3: Figure S-4-3. Use of conoscopy to distinguish the x- and y- axis in a KTP plate cut normal

To return to our original question, once we have determined the correct direction of propagation in the $x-y-z$ principal axis sytem, and once we have clarified any confusion about the relationship between these axes and the $a-b-c$ crystallographic axes, we can use techniques like x-ray diffraction that are sensitive to the crystallographic structure to establish the correct direction for propagation. In addition, we can use optical techniques such as conoscopy that are sensitive to the alteration of the polarization state of light by the crystal to discover the orientation of the principal axes (Figure S-4-3). A detailed description of how techniques like these are used in beyond the scope of this book, but the interested reader can consult works like that of Wahlstrom [Wahlstrom 1969].

C Eigenpolarizations

When we solve Maxwell's equations using the full tensor permittivity of Eq. 4.2, we discover that for any direction of propagation in the medium, there exist two linearly polarized waves that can propagate through the material and retain their original linear polarization. We will refer to these two particular waves as "eigenpolarizations", since they can be expressed as the solutions of an eigenvector problem. These two waves travel at different speeds; that is, they are characterized by different refractive indices. (The proof of this statement is beyond the scope of this text, but the interested reader can find it in several discussions of crystal optics, for example, [Yariv and Yeh 1984]). The refractive indices for these two linear eigenpolarizations depend upon the direction of propagation. Therefore, in order to use birefringence for phasematching, we need to know how to calculate the orientations of these eigenpolarizations and their refractive indices as a function of propagation direction.

In Chapter 2 of "CBGL," we introduced the "index normal surface" for solving certain phasematching problems. In this supplemental chapter, we present an additional tool that is extremely useful for understanding the propagation of waves in anisotropic media, the "index ellipsoid."

C The Index Ellipsoid

The energy density associated with an electromagnetic wave is given by

$$U = \frac{1}{2}\mathbf{E}\cdot\mathbf{D} \tag{4.4}$$

In the principal axis system, we have:

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \epsilon_o \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} \tag{4.5}$$

so that

$$U = \frac{1}{2}\left[\frac{D_x^2}{\epsilon_o n_x^2} + \frac{D_y^2}{\epsilon_o n_y^2} + \frac{D_z^2}{\epsilon_o n_z^2}\right] \tag{4.6}$$

This equation has the form of an ellipsoid in the coordinates D_x, D_y, and D_z. This ellipsoid tells us what the magnitude and direction of the **D** vector must be in order for a wave propagating in a crystal characterized by n_x, n_y, n_z to have an energy density U.

We can re-write this expression in the form

$$\begin{aligned} \frac{\left(\frac{D_x}{\sqrt{2\epsilon_o U}}\right)^2}{n_x^2} + \frac{\left(\frac{D_y}{\sqrt{2\epsilon_o U}}\right)^2}{n_y^2} + \frac{\left(\frac{D_z}{\sqrt{2\epsilon_o U}}\right)^2}{n_z^2} &= 1 && (4.7) \\ \frac{\alpha^2}{n_x^2} + \frac{\beta^2}{n_y^2} + \frac{\gamma^2}{n_z^2} &= 1 && (4.8) \end{aligned}$$

This equation defines an ellipsoid in the normalized coordinates $\alpha = \dfrac{D_x}{\sqrt{2\epsilon_o U}}$, $\beta = \dfrac{D_y}{\sqrt{2\epsilon_o U}}$, and $\gamma = \dfrac{D_y}{\sqrt{2\epsilon_o U}}$, the size and shape of which depends only on the refractive indices n_x, n_y, and n_z. This three-dimensional ellipsoid intersects the "index space" axes at $\alpha = \pm n_x$, $\beta = \pm n_y$, $\gamma = \pm n_z$ (Figure S-4-4), and is called the "index ellipsoid" or "optical indicatrix" or "ellipsoid of wave normals."

What is the relationship between the $x-y-z$ coordinates of physical space and the $\alpha-\beta-\gamma$ coordinates of index space? Suppose that we have a wave propagating along the $x-$axis in physical space. Since the **D** vector must be normal to the direction of propagation, even in anisotropic media, it must lie somewhere in the physical $y-z$ plane; that is, it must have only D_y and D_z components with $D_x = 0$. In index space, this condition means that we must confine our attention to the two-dimensional ellipse defined by $\alpha = 0$. Hence, propagation along the $x-$axis in physical space corresponds in this way to $\alpha = 0$ in index space; similarly, propagation along the $y-$axis corresponds to $\beta = 0$ and propagation along the $z-$axis to $\gamma = 0$.

This correspondence between physical-space coordinates and index-space coordinates lies behind the following recipe for determining the orientation and refractive index of the two linear eigenpolarizations for any direction of propagation. The recipe is this: First, draw a ray from the origin in the direction of propagation of the wave, according to the correspondence just described ($x, y, z \leftrightarrow \alpha, \beta, \gamma$). Second, construct a plane passing through the origin and normal to that ray. The intersection of this plane with the index ellipsoid is an ellipse (as we just saw to be the case for propagation along a physical principal axis). The directions of the **D** vectors of the two linear eigenpolarizations lie along the major and minor axes of this ellipse, and the corresponding refractive indices are given by the length of the semi-major and semi-minor axes (that is, the distance from the origin to the ellipse). This procedure is depicted in Figure S-4-5 for a propagation direction that lies in the $y-z$ plane. A proof that this recipe is mathematically sound can be found in [Yariv and Yeh 1984].

As a simple example, we can consider the case of propagation along the crystallographic a-axis in potassium niobate, as discussed earlier (Figure S-4-6) In potassium niobate, the crystallographic coordinate designations are often used to avoid the problem with conventions described earlier, and we will follow that practice here. The perpendicular plane is thus the $b-c$ plane The intersection between this plane and the indicatrix is an ellipse with minor axis along c and major axis along b. Thus, the directions of the linear eigenpolarization lie along the $b-$ and $c-$axes, and the corresponding refractive indices are n_b and n_c. This is a somewhat trivial example, but when the direction of propagation does not lie along a principle axis of the crystal, this method is very powerful for determining the appropriate refractive indices.

If we examine Figure S-4-5, we can imagine that there will be some direction of

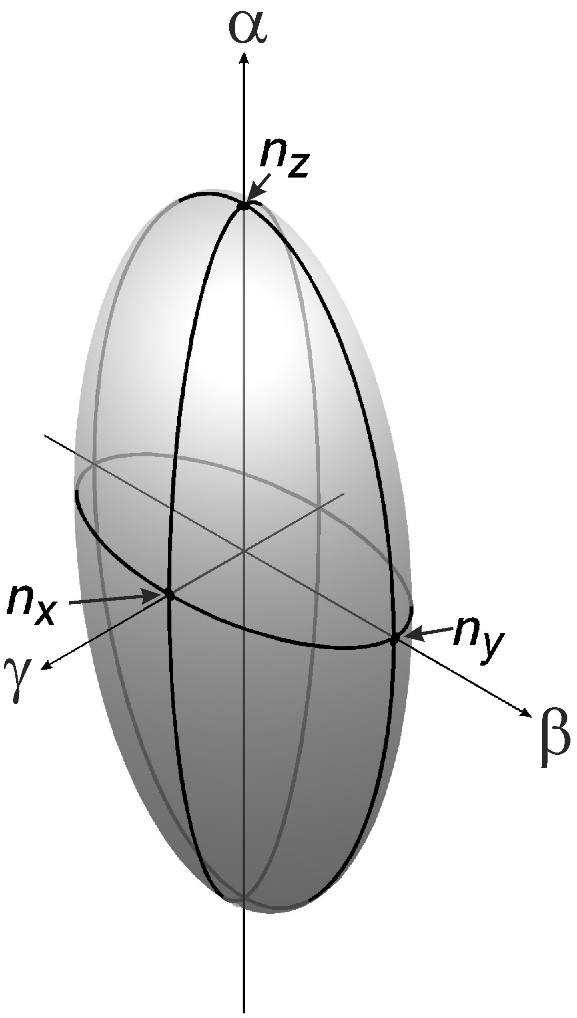

Figure 4.4: Figure S-4-4. The index ellipsoid, or optical indicatrix

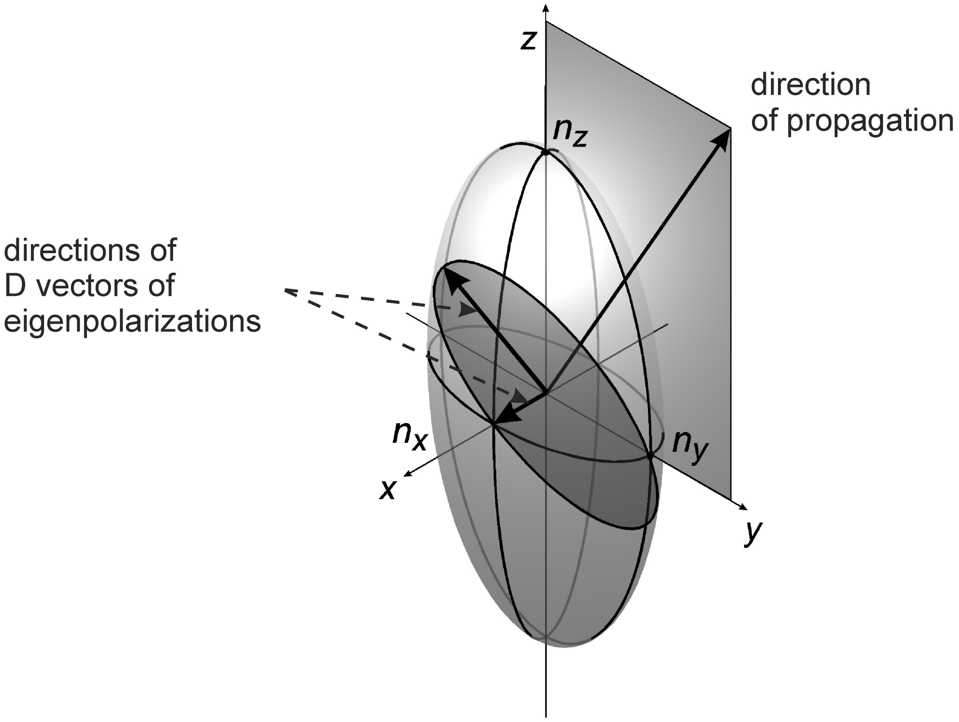

Figure 4.5: Figure S-4-5. Use of the index ellipsoid to find the directions of the eigenpolarizations for propagation in the $y-z$ plane.

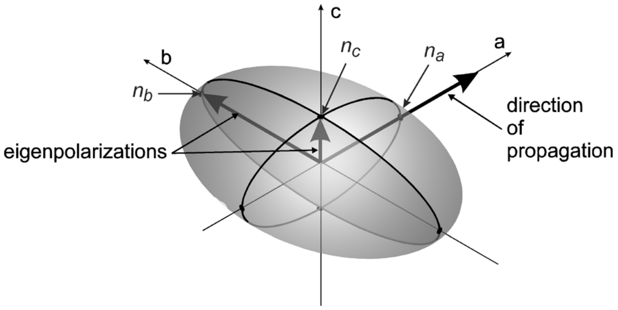

Figure 4.6: Figure S-4-6. Simple example of the use of the ellipsoid for analyzing propagation along the $a-$axis in potassium niobate.

propagation for which the ellipse found by intersection with the normal plane will be a circle—in fact, there will be *two* such directions at equal angles from the x axis in the $x-z$ plane. For these two directions of propagation, there is no preferred direction for the linear polarization, and any linear polarization will propagate with the same refractive index. Such a direction is referred to as an "optical axis" of the crystal. We can see that in *bi*axial crystal, in which $n_x \neq n_y \neq n_z$, there will be *two* such axes. In a *uni*axial crystal, in which, say, $n_x = n_y \neq n_z$, the ellipsoid will have a circular cross-section perpendicular to the $z-$ axis; thus, there will be only *one* optical axis, and it will lie along the $z-$ principal axis (Figure S-4-7). We designate the two equal components the *ordinary* index ($n_o = n_x = n_y$) and the other component the extraordinary index ($n_e = n_z$). If $n_e > n_o$, the crystal is said to be *positive uniaxial*; if $n_e < n_o$, it is said to be *negative uniaxial.* In an *an*axial crystal, in which $n_x = n_y = n_z$, the optical indicatrix is a sphere, and there is *no* unique optical axis; all directions of propagation are equivalent (Figure S-4-8). Such materials are also said to be *optically isotropic.*

Returning to the example of potassium niobate as shown in Figure S-4-6, suppose that instead of propagating along the $a-$axis, we propagate in the $a-b$ plane at some angle θ to the $a-$axis. What are the allowed eigenpolarizations, and the corresponding refractive indices in this case? From Figure S-4-5, we can see that one of the linear polarizations will still lie along the $c-$axis and will have refractive index n_c for any value of θ. The other polarization will lie in the $a-b$ plane, and the refractive index will depend on the angle, falling somewhere between n_b for $\theta = 0$ and n_a for $\theta = 90°$. We can determine the refractive index for arbitrary values of θ by setting $\gamma = 0$ in Eq. 4.8. We obtain:

$$\frac{\alpha^2}{n_a^2} + \frac{\beta^2}{n_b^2} = 1 \tag{4.9}$$

$$\frac{(n_\theta \sin\theta)^2}{n_a^2} + \frac{(n_\theta \cos\theta)^2}{n_b^2} = 1 \tag{4.10}$$

or

$$n_\theta = \left[\frac{\sin^2\theta}{n_a^2} + \frac{\cos^2\theta}{n_b^2}\right]^{-\frac{1}{2}} \tag{4.11}$$

When $\theta = 0$, $n_\theta = n_b$. When $\theta = 90°$, $n_\theta = n_a$. For intermediate angles, the value of n_θ lies somewhere between n_a and n_b.

We can see how this might be useful for phasematching. We said earlier that propagation along the $a-$axis in potassium niobate provided phasematching for SHG of a fundamental wavelength of $\lambda_1 = 858$ nm. At this wavelength, for this direction of propagation, dispersion exactly matches birefringence. For fundamental wavelengths longer than 858 nm, the dispersion is lower than birefringence. Therefore, if we want

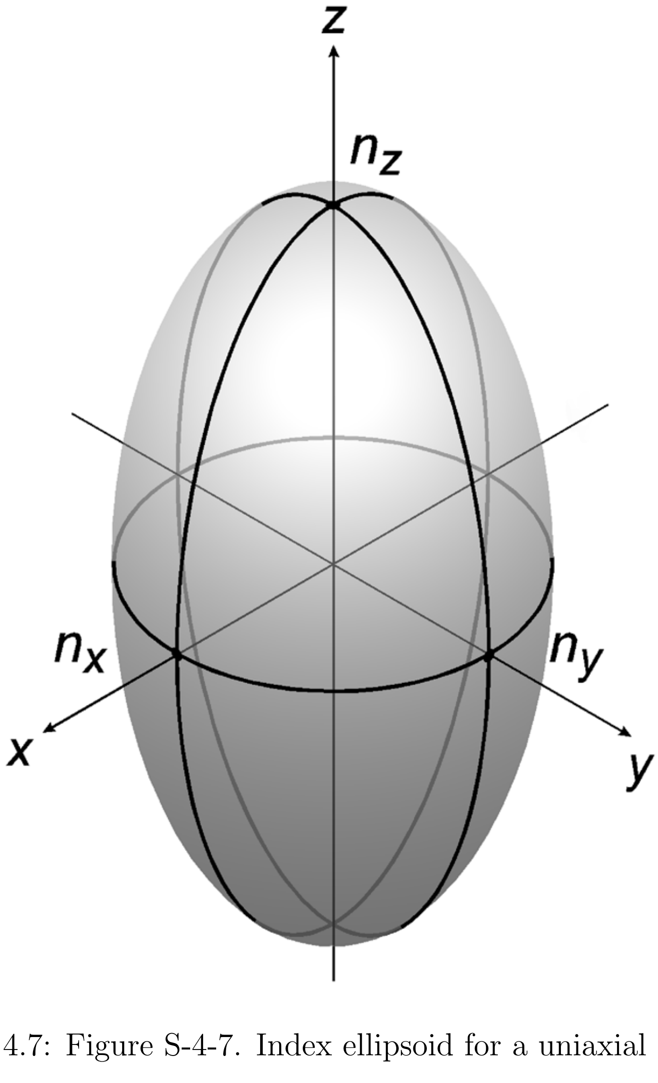

Figure 4.7: Figure S-4-7. Index ellipsoid for a uniaxial crystal

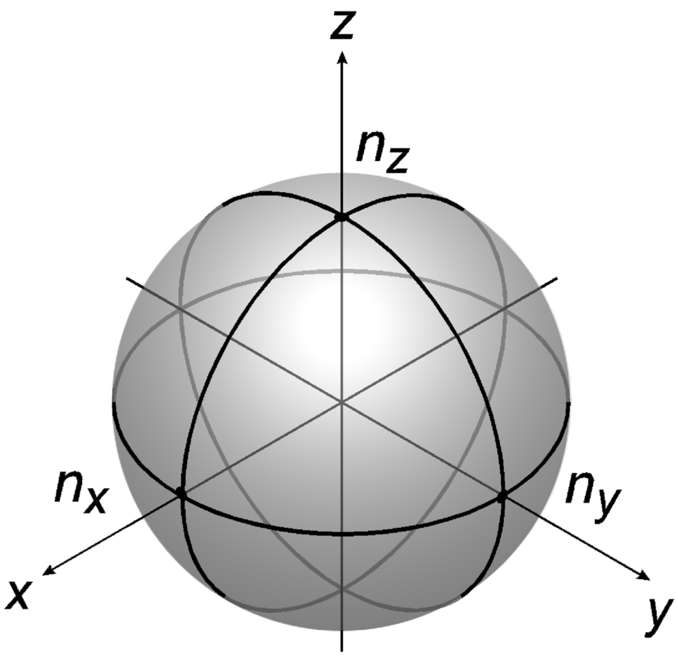

Figure 4.8: Figure S-4-8. Index ellipsoid for an anaxial crystal

to use, say, $\lambda_1 = 946$ nm (which is the wavelength of a particular kind of Nd:YAG laser), we need to reduce the birefringence provided by the crystal. Now we can see how to do this: simply change the direction of propagation in the $a - b$ plane so that the birefringence matches the dispersion.

CT

Bibliography

[Dimitriev and Nikogosyan (1993)] Dimitriev, V. G. and Nikogosyan, D. N. (1993) Effective nonlinearity coefficients for three-wave interactions in biaxial crystals of $mm2$ point group symmetry. *Opt. Comm.*, **95**, 173-182.

[Roberts (1992)] Roberts, D. A. (1992) Simplified Characterization of Uniaxial and Biaxial Nonlinear Optical Crystals: A Plea for Standardization of Nomenclature and Conventions. *IEEE J. Quantum Electron.*, **28**, 2057-2074.

[Yariv and Yeh 1984] Yariv, A.and Yeh, P. (1984) Optical Waves in Crystals (New York: Wiley and Sons)

[Wahlstrom 1969] Wahlstrom, E. E. (1969) Optical Crystallography, (New York: Wiley and Sons)