Journal of the Optical Society of America B
OPTICAL PHYSICS

Light propagation in anisotropic materials and electro-optical effects: tutorial on the use of eigenvalue problems, tensors, and symmetries

PATRICK STEGLICH[1,2,*] AND ACHIM KEHREIN[3,4]

[1]Technische Hochschule Wildau, Hochschulring 1, 15745 Wildau, Germany
[2]IHP-Leibniz-Institut für innovative Mikroelektronik, Im Technologiepark 25, D-15236 Frankfurt (Oder), Germany
[3]Hochschule Rhein-Waal, Fakultät Technologie und Bionik, Marie-Curie-Str. 1, D-47533 Kleve, Germany
[4]achim.kehrein@hochschule-rhein-waal.de
*steglich@ihp-microelectronics.com

Received 19 March 2024; revised 12 August 2024; accepted 13 August 2024; posted 13 August 2024; published 29 August 2024

The properties of anisotropic materials are used in many optical components such as waveplates or polarizing beamsplitters. In particular, anisotropic materials that possess electro-optical properties allow the realization of actively controllable optical components like optical switches, phase shifters, or modulators. Hence, understanding and computation of light propagation in anisotropic materials with electro-optical effects are crucial in optical science and technology. On the one hand this tutorial stresses the use of eigenvalue problems to explain qualitatively and to compute quantitatively important properties such as polarization. On the other hand it discusses the mathematical model of both electro-optical effects, namely, the Pockels and the DC Kerr effect. This tutorial describes the basic concepts in a consistent tensor language, shows how the tensors are conveniently summarized in matrices, and points out that these matrices do not transform like tensors. The tensor approach clarifies how symmetry arguments affect tensor components. Further, this paper derives the more accurate nonlinear relationship between the refractive index and the externally applied electric field.

https://doi.org/10.1364/JOSAB.524213

1. INTRODUCTION

The propagation of light through an anisotropic optical material depends on the dielectric tensor of the material. This tensor determines the reduced speed of light inside the material and restricts the polarization direction of the light. For some materials, the dielectric tensor and hence the optical properties will change, if an external electric field is applied. The change of the tensor components is approximated by the first terms of a Taylor series with respect to the external electric field with center $\mathbf{E} = \mathbf{0}$. The effect of the linear term of the Taylor series is called the Pockels effect and the effect of the quadratic term is known as the DC Kerr effect.

Electro-optical effects feature several advantages for technological applications. First, the refractive index change is ultra-fast and is currently limited by the electronic driver circuit but not by the electro-optical material [1]. Second, the electric field modifies only the real part (refraction) of the complex refractive index but does not alter the imaginary part (absorption) [2]. For example, this is important for coherent optical modulators [3,4]. Third, electro-optical materials are highly transparent in the visible wavelength range [5] and also in the optical O-band and C-band [6], which are important for telecommunication applications. Electro-optical effects are used for high-speed modulators [7–9], for the generation of ultra-short laser pulses [10–12], in photoacoustics [5,13], and for electric field sensing [14–16]. Important materials are polymer systems [17–20], barium titanate (BTO) [21,22], lithium niobate [23,24], and lead zirconate titanate (PZT) [25,26].

Electro-optical effects are described in several textbooks. The classic book by Yariv and Yeh [27] provides an excellent introduction to electro-optical effects in crystals, but is out of print. Unfortunately, many texts take the decades-old model assumptions for granted, skip mathematical details, often avoid a consistent tensor language, and do not aim at a deeper understanding of the foundations. The present tutorial provides mathematical details in a general context. In particular, tensors and eigenvalue problems are stressed.

The first part of this tutorial starts in Section 2 by briefly reviewing the polarization effects of an electric field on matter and defines the impermeability tensor, which is the central object of interest. Then Section 3 derives the eigenvalue problem of the impermeability tensor from Maxwell's equations and establishes that this eigenvalue problem describes electromagnetic wave propagation through optical anisotropic materials. Section 4 shows that the eigenvalues determine the speed of light through the medium. The speed of light ratios relates to

the semi-axes of the index ellipsoid, which visualizes the impermeability tensor. This visualization is presented in Section 5. Then, Section 6 explains how eigenvectors correspond to optical polarizations. The mathematical details of the eigenvalue problem that connects a specific direction of the propagation of light with the impermeability tensor are presented. Section 7 closes the general part of the paper by visualizing the specific eigenvalue problem as one of the ellipses that is the intersection of the index ellipsoid with the plane normal to the propagation of light.

The second part of the paper starts with Section 8. It introduces the tensors of the linear and quadratic electro-optical effects, i.e., the Pockels effect and the DC Kerr effect, respectively. These tensors possess 27 and 81 components, respectively, of which many are repeating due to symmetries, so that their independent components are conveniently summarized in matrices even though these matrices are not tensors. On top of the mathematical symmetries the symmetries of the chosen anisotropic material produce some zero matrix entries and some repeating ones. The underlying tensor theory of symmetry is elaborated in Section 10. The following Section 11 is a case study of the symmetries of the electro-optical material barium titanate (BTO) for the Pockels effect. We point out that a commonly used linearized formula does not provide sufficient precision for a material like BTO with an ultra-large linear electro-optical effect. This is relevant for a more accurate determination of electro-optical material parameters from experiments. Finally, Section 12 studies how the physical symmetries of BTO are inherited by the quadratic electro-optical tensor.

2. EFFECT OF AN ELECTRIC FIELD ON MATTER–ELECTRIC DISPLACEMENT, DIELECTRIC TENSOR, AND IMPERMEABILITY TENSOR

When an external electric field acts on matter, it induces **dielectric polarization** effects. The electric field creates dipole moments in formerly unpolarized atoms or aligns already existing dipole moments of molecules. This paper considers a linear dielectric medium, i.e., the magnitude of the polarization density field $\mathbf{P}$ depends linearly on the magnitude of the electric field $\mathbf{E}$, but their directions need not be collinear. The dependence of the polarization density on the electric field is described by the electric susceptibility tensor χ also known as polarization tensor:

$$\begin{pmatrix} P_1 \\ P_2 \\ P_3 \end{pmatrix} = \epsilon_0 \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}, \tag{1}$$

where ϵ_0 is the permittivity of free space.

Actually, Eq. (1) is a coordinate representation of the polarization tensor. There are two interpretation styles of the coordinate invariance of tensor equations. Some scientists consider such a coordinate form already as an invariant equation, as there are fixed rules for transforming the equation into any other allowed coordinate system. For example, Einstein wrote the equations of general relativity in this coordinate form and considered them invariant. Other scientists, in particular those familiar with the modern presentation of differential geometry, prefer an extra level of abstraction. For them, the only acceptable invariant form of Eq. (1) looks like

$$\mathbf{P} = \epsilon_0 \chi \mathbf{E}. \tag{2}$$

We consider it advantageous to consider a concrete coordinate representation as a valid description of the invariant quantity. Moreover, when the coordinate system is fixed, some tensor equations can be rewritten using matrix products. We often do so, to show the connections with elementary results from matrix theory.

In this tutorial the allowed coordinate systems are Cartesian, i.e., the axes are orthogonal and there is a common unit for all axes. Consistently, the coordinate transformations of vector components correspond to orthogonal matrices, which describe rotations or reflections. A reason for this restriction is rather technical. (The important tensor η below transforms according to $S\eta S^T$ under a coordinate change S, while its eigenvalues are invariant under the transformation $S\eta S^{-1}$. To consider eigenvalues as invariant quantities, we need $S^T = S^{-1}$, which is the defining condition for an orthogonal transformation.)

This polarization tensor is symmetric, $\chi_{ij} = \chi_{ji}$. Feynman uses α_{xy} instead of χ_{ij} and points out that this property is a physical one, not a mathematical one:

> "(This [symmetry] is a *physical* property of the real crystal and not necessary for all tensors.) You can prove for yourself that this must be true by computing the change in energy of a crystal through the following cycle: (1) Turn on a field in the x-direction; (2) turn on a field in the y-direction; (3) turn *off* the x-field; (4) turn off the y-field. The crystal is now back where it started, and the net work done on the polarization must be back to zero. You can show, however, that for this to be true, α_{xy} must be equal to α_{yx}. The same kind of argument can, of course, be given for α_{xz}, etc. So the polarization tensor is symmetric" ([28], Volume II, Chapter 31 Tensors).

Appendix A provides more mathematical details by translating these steps in line integrals. The above argument is independent of the chosen coordinate system. It also follows from the transformation rules for tensors below that, mathematically, if tensor components are symmetric with respect to one coordinate system, they are symmetric in every coordinate system.

In this tutorial we work with a symmetric dielectric tensor, which assumes materials with negligible optical chirality [29]. The polarization density $\mathbf{P}$ is an electrical field inside the material that partially counteracts the external electrical field $\mathbf{E}$. The superposition of the external electric field and its induced polarization density is the **electrical displacement** $\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$:

$$\begin{pmatrix} D_1 \\ D_2 \\ D_3 \end{pmatrix} = \epsilon_0 \underbrace{\begin{pmatrix} 1+\chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & 1+\chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & 1+\chi_{33} \end{pmatrix}}_{\text{dielectric tensor } \epsilon_{ij}} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}. \tag{3}$$

The **dielectric tensor** $\boldsymbol{\epsilon} = \mathbf{1} + \boldsymbol{\chi}$ is symmetric due to its symmetric summands. Moreover, since every material responds to a

nonzero electric field with a nonzero electric displacement, the dielectric tensor is invertible. The inverse of the dielectric tensor is the **impermeability tensor** $\boldsymbol{\eta} = \boldsymbol{\epsilon}^{-1}$ and is the key tensor of interest in the discussion below.

For a symmetric tensor the Principal Axes Theorem from Linear Algebra guarantees that there is a Cartesian coordinate system (an orthonormal basis), with respect to which the tensor is diagonal. For example, diagonality of the dielectric tensor means $\epsilon_{ij} = 0$ for $i \neq j$. Then the diagonal entries $\epsilon_1 = \epsilon_{11}$, $\epsilon_2 = \epsilon_{22}$, and $\epsilon_3 = \epsilon_{33}$ are called the principal values or eigenvalues of the tensor. Because of the relationships between susceptibility tensor, dielectric tensor, and impermeability tensor, they are diagonal with respect to the same coordinate system.

If the optical medium is isotropic then the polarization density will always be directed along the external electric field. Hence, the susceptibility tensor, the dielectric tensor, and the impermeability tensor are diagonal with constant diagonal entries independent of the coordinate system. In the isotropic case, these tensors simplify to scalar quantities.

If the optical medium is anisotropic then, at least sometimes, the polarization density will be not collinear with the electric field. The eigenvalues are not all equal anymore, which leads to two subcases. In the **uniaxial** case two of the three eigenvalues are still equal, while in the **biaxial** case the three eigenvalues are pairwise different.

3. LIGHT PROPAGATION IN MATTER AS AN EIGENVALUE PROBLEM

Eigenvalues and eigenvectors determine the index of refraction and light polarization, respectively. For a deeper understanding we shall start with Maxwell's equations in matter:

1. Gauss's law for electric fields

$$\nabla \cdot \mathbf{D} = \rho_{\text{free}}, \tag{4}$$

where ρ_{free} is the density of free electric charges;

2. Gauss's law for magnetic fields

$$\nabla \cdot \mathbf{B} = 0; \tag{5}$$

3. Faraday's law

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}; \tag{6}$$

and the Maxwell-Ampère law

$$\nabla \times \mathbf{H} = \mathbf{J}_{\text{free}} + \frac{\partial \mathbf{D}}{\partial t}, \tag{7}$$

where $\mathbf{J}_{\text{free}}$ is the free electric current density.

The fields in the above equations are averaged over the volume of a unit cell. The averaging volume is large compared to the scale of the charge density modulation, which is the nonuniform distribution of charges. However, it is small compared to the wavelength of a light wave.

Moreover, we have the constitutive relations, which connect the fields with the material properties:

$$\begin{pmatrix} D_1 \\ D_2 \\ D_3 \end{pmatrix} = \epsilon_0 \begin{pmatrix} \epsilon_{11} & \epsilon_{12} & \epsilon_{13} \\ \epsilon_{21} & \epsilon_{22} & \epsilon_{23} \\ \epsilon_{31} & \epsilon_{32} & \epsilon_{33} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}, \quad \text{or} \quad \mathbf{D} = \epsilon_0 \boldsymbol{\epsilon} \mathbf{E}, \tag{8}$$

and

$$\mathbf{H} = \frac{1}{\mu_0}\mathbf{B} - \mathbf{M}, \tag{9}$$

where $\mathbf{M}$ describes the magnetization effect of the magnetic field on the medium similarly to the dielectric polarization effect of an electric field.

Our model assumes that there are neither free charges, $\rho_{\text{free}} = 0$, nor free currents, $\mathbf{J}_{\text{free}} = 0$. Besides that, we assume that the medium does not respond to magnetic fields ($\mathbf{M} = 0$) because light waves have very high frequencies (in the terahertz range), and magnetic dipole moments cannot effectively follow these rapid oscillations. The response of electrons to the electric field of the light wave is much stronger than to the magnetic field of the same wave.

Now we derive the relationships for light propagation in the medium. We substitute $\mu_0 \mathbf{H} = \mathbf{B}$, i.e., Eq. (9) with $\mathbf{M} = 0$, in Faraday's law, Eq. (6),

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t}, \tag{10}$$

apply the curl operation,

$$\nabla \times (\nabla \times \mathbf{E}) = -\mu_0 \nabla \times \frac{\partial \mathbf{H}}{\partial t}, \tag{11}$$

and plug in Maxwell-Ampere's law, Eq. (7) with $\mathbf{J}_{\text{free}} = 0$, to obtain

$$\nabla \times (\nabla \times \mathbf{E}) = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}. \tag{12}$$

The inverse constitutive equation $\frac{1}{\epsilon_0}\boldsymbol{\eta}\mathbf{D} = \mathbf{E}$ leads to the wave equation

$$\nabla \times (\nabla \times \boldsymbol{\eta}\mathbf{D}) = -\mu_0 \epsilon_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}, \tag{13}$$

where $\boldsymbol{\eta}$ is the impermeability tensor. We use the inverse constitutive relation instead of $\mathbf{D} = \epsilon_0 \boldsymbol{\epsilon} \mathbf{E}$ to obtain an eigenvalue problem $T\mathbf{D} = \alpha \mathbf{D}$ instead of a generalized eigenvalue problem $S\mathbf{E} = \alpha \boldsymbol{\epsilon} \mathbf{E}$.

There is also a physical reason to prefer $\mathbf{D}$ over $\mathbf{E}$ in our setting. The optical polarization of light waves traveling through free space is given by the direction of the electric field $\mathbf{E}$. But we are considering light waves traveling through an optical medium, so the **optical polarization** depends on the direction of the electrical displacement $\mathbf{D}$.

A light wave can be written as a superposition of plane waves of the form

$$\mathbf{D}(\mathbf{r}, t) = \mathbf{D}_0 \cdot \sin(\mathbf{k} \cdot \mathbf{r} - \omega t + \varphi) \quad \text{with} \quad \mathbf{r} = (x, y, z)^T, \tag{14}$$

where $\mathbf{D}_0 \neq \mathbf{0}$ is a constant vector, $|\mathbf{k}| = 2\pi/\lambda$ is the wave number with wavelength λ, frequency $\omega = 2\pi/T$ with period T, and φ is a constant phase shift. The plane wave travels in the direction of $\mathbf{k}$.

The second partial derivatives of the plane wave are

$$\frac{\partial^2 \mathbf{D}}{\partial x_i \partial x_j} = -k_i k_j \mathbf{D}, \quad \text{and} \quad \frac{\partial^2 \mathbf{D}}{\partial t^2} = -\omega^2 \mathbf{D}. \tag{15}$$

Applying the partial derivative formulas to Eq. (13) yields

$$-\mathbf{k} \times (\mathbf{k} \times \boldsymbol{\eta})\mathbf{D} = \omega^2 \mu_0 \epsilon_0 \mathbf{D}. \tag{16}$$

Then, let $\mathbf{k} = k \cdot \hat{\mathbf{k}}$ with unit vector $\hat{\mathbf{k}}$ and divide by the squared magnitude k^2:

$$-\hat{\mathbf{k}} \times (\hat{\mathbf{k}} \times \boldsymbol{\eta})\mathbf{D} = \frac{\omega^2 \mu_0 \epsilon_0}{k^2} \mathbf{D}. \tag{17}$$

Since $c_0 = 1/\sqrt{\epsilon_0 \mu_0}$, the scalar on the right-hand side equals

$$\frac{\omega^2 \mu_0 \epsilon_0}{k^2} = \frac{(2\pi)^2}{T^2} \frac{\lambda^2}{(2\pi)^2} \frac{1}{c_0^2} = \left(\frac{v}{c_0}\right)^2, \tag{18}$$

where v is the speed of light inside the medium. Finally, we factor out the scalar function $\sin(\mathbf{k} \cdot \mathbf{r} - \omega t + \varphi)$ and obtain the eigenvalue problem for the polarization $\mathbf{D}_0$:

$$-\hat{\mathbf{k}} \times (\hat{\mathbf{k}} \times \boldsymbol{\eta})\mathbf{D}_0 = \left(\frac{v}{c_0}\right)^2 \mathbf{D}_0. \tag{19}$$

An eigenvalue problem has only specific solutions. The eigenvectors specify the allowed polarization directions of the light propagating along $\mathbf{k}$ through the medium. The square root of the eigenvalue determines the fraction by which the speed of light is reduced along $\mathbf{k}$ in the medium. The ratio $n = c_0/v$ is the refractive index of the medium in the direction of $\mathbf{k}$.

To learn more about the eigenvalue problem we rewrite the cross product of $\hat{\mathbf{k}}$ and $\mathbf{v}$,

$$\begin{pmatrix} \hat{k}_1 \\ \hat{k}_2 \\ \hat{k}_3 \end{pmatrix} \times \begin{pmatrix} v_1 \\ v_2 \\ v_3 \end{pmatrix}, \tag{20}$$

as the tensor-vector product

$$\underbrace{\begin{pmatrix} 0 & -\hat{k}_3 & \hat{k}_2 \\ \hat{k}_3 & 0 & -\hat{k}_1 \\ -\hat{k}_2 & \hat{k}_1 & 0 \end{pmatrix}}_{=R_{\mathbf{k}}} \begin{pmatrix} v_1 \\ v_2 \\ v_3 \end{pmatrix}. \tag{21}$$

The tensor $R_{\mathbf{k}}$ is skew-symmetric, $R_{\mathbf{k}}^T = -R_{\mathbf{k}}$. It has only one real-number eigenvalue, namely, zero in the direction of $\mathbf{k}$. Hence this tensor is non-invertible. Vectors in the plane perpendicular to $\mathbf{k}$ are rotated by 90° about $\mathbf{k}$, which corresponds to the pair of complex conjugate eigenvalues i and $-i$.

Applying the cross product twice, $\hat{\mathbf{k}} \times (\hat{\mathbf{k}} \times \mathbf{v})$, leads to the tensor product

$$\underbrace{\begin{pmatrix} -\hat{k}_2^2 - \hat{k}_3^2 & \hat{k}_1 \hat{k}_2 & \hat{k}_1 \hat{k}_3 \\ \hat{k}_2 \hat{k}_1 & -\hat{k}_1^2 - \hat{k}_3^2 & \hat{k}_2 \hat{k}_3 \\ \hat{k}_3 \hat{k}_1 & \hat{k}_3 \hat{k}_2 & -\hat{k}_1^2 - \hat{k}_2^2 \end{pmatrix}}_{=K} \begin{pmatrix} v_1 \\ v_2 \\ v_3 \end{pmatrix}, \tag{22}$$

with the symmetric tensor K, which still has the eigenvalue zero in the direction of $\hat{\mathbf{k}}$. Vectors in the plane perpendicular to $\hat{\mathbf{k}}$ are rotated by 180°, so they are eigenvectors with eigenvalue -1. Finally, we incorporate the negative sign from Eq. (17). With respect to an orthonormal eigenvector basis with third vector $\hat{\mathbf{k}}$, we have

$$P_{\mathbf{k}} = -K = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{23}$$

which is the orthogonal projection along $\hat{\mathbf{k}}$ onto its perpendicular plane.

Therefore, the propagation of light along $\mathbf{k}$ through an anisotropic medium is governed by the eigenvalue problem

$$(P_{\mathbf{k}} \boldsymbol{\eta})\mathbf{D}_0 = \left(\frac{v}{c_0}\right)^2 \mathbf{D}_0. \tag{24}$$

Of course, the eigenvalues, which determine the speed of light inside the medium, depend on the impermeability tensor of the medium and the direction of light propagation.

The speed of light inside the medium is $v = \omega/k = \lambda/T$. Light traveling through the medium consists of photons of energy $E = \hbar\omega$, where $\hbar$ is Planck's constant divided by 2π. The photons do not change their energy, so the angular frequency ω and, equivalently, the period T of the electromagnetic wave do not change. Therefore, the decreased velocity inside the medium corresponds to a decreased wavelength λ and, equivalently, to an increased wave number k. Here we assume a transparent material, i.e., a material in which the absorption is zero and other effects due to scattering (e.g., the Compton effect) can be neglected.

The following sections discuss the details of this eigenvalue problem. Section 4 establishes the connection with the index of refraction. Then Section 6 solves the eigenvalue problem, which leads to the ellipsoid visualization in Section 5, and Section 7 incorporates the eigenvalue problem in the ellipsoid visualization.

4. RELATIONSHIP BETWEEN EIGENVALUES OF THE IMPERMEABILITY TENSOR AND THE INDEX OF REFRACTION

Light travels through vacuum with constant velocity c_0. When it travels through an optical medium instead, the electromagnetic light wave induces dielectric polarization effects in the atoms or molecules of the medium. These dielectric polarization effects impede the light propagation and lead to a reduced velocity of $v = c_0/n$ with $n > 1$. The constant n is the **index of refraction**. It depends on the medium and may depend on the propagation direction and the polarization of the electromagnetic light wave relative to the medium. Unfortunately, the term polarization appears in two different meanings. On the one hand it describes the effect on the atoms or molecules that creates a partially offsetting electric field. This is **dielectric polarization**. On the other hand, **optical polarization** refers to the direction of the electric displacement $\mathbf{D}$ perpendicular to the propagation of light through the optical medium. However, while both meanings occur in this paragraph, only the latter meaning occurs in the remainder of this paper.

Amorphous media such as glasses, gases, or crystalline materials with cubic lattice symmetry are isotropic. Their index of refraction is a scalar quantity, which is independent of the propagation direction and polarization. In contrast, crystalline materials with non-cubic lattice symmetry are anisotropic. Their index of refraction depends on the propagation direction and on polarization. Some cubic point group symmetries do not have to be isotropic with respect to their other properties.

To find out more about the dependence of the index of refraction on propagation direction and polarization, we study special cases of the eigenvalue problem in Eq. (24). The impermeability tensor $\boldsymbol{\eta}$ is symmetric and, hence, can be diagonalized with respect to its orthogonal principal axes, say, $\mathbf{p}_1$, $\mathbf{p}_2$, and $\mathbf{p}_3$ with respective eigenvalues η_1, η_2, and η_3. (Please note, the real and imaginary parts separately can be diagonalized, but for low-symmetry crystals, i.e., monoclinic and triclinic, the principal axes for dispersion and absorption are different [30].)

First we consider the special case of a planar light wave propagating along one of the principal axes, say, $\mathbf{k} = \mathbf{p}_3$. See Fig. 1.

Then $\mathbf{p}_1$ and $\mathbf{p}_2$ are in the normal plane $\mathbf{k}^\perp$. Hence the principal axes of $\boldsymbol{\eta}$ are also eigenvectors of the projection $P_\mathbf{k}$. With respect to this basis we have the coordinate representation

$$P_\mathbf{k}\boldsymbol{\eta} = \begin{pmatrix} 1&0&0\\ 0&1&0\\ 0&0&0 \end{pmatrix} \begin{pmatrix} \eta_1&0&0\\ 0&\eta_2&0\\ 0&0&\eta_3 \end{pmatrix} = \begin{pmatrix} \eta_1&0&0\\ 0&\eta_2&0\\ 0&0&0 \end{pmatrix}, \tag{25}$$

and the eigenvalue problem attains the diagonal form

$$\underbrace{\begin{pmatrix} \eta_1&0&0\\ 0&\eta_2&0\\ 0&0&0 \end{pmatrix}}_{\text{if } \mathbf{k} \text{ along axis of } \eta_1} \begin{pmatrix} D_{0,1}\\ D_{0,2}\\ D_{0,3} \end{pmatrix} = \left(\frac{v}{c_0}\right)^2 \underbrace{\begin{pmatrix} D_{0,1}\\ D_{0,2}\\ D_{0,3} \end{pmatrix}}_{\text{polarization}}. \tag{26}$$

Similarly, light propagation along $\mathbf{k} = \mathbf{p}_2$ leads to eigenvalues $\eta_1, 0$, and η_3. We obtain the following important relationship.

The eigenvalues η_i of the impermeability tensor are related to the indices of refraction n_i along the eigenvector directions by

$$\eta_i = (v_i/c_0)^2 = 1/n_i^2 \quad \text{for } i = 1, 2, 3. \tag{27}$$

The second equation uses the definition of the refractive index, $n = c_0/v$, as the quotient of speed of light inside the medium and the speed of light in empty space.

This result shows another important mathematical property: the impermeability tensor is **positive-definite**, since all eigenvalues are positive. A positive-definite tensor possesses a useful visualization as an ellipsoid, which is discussed in Section 5.

Recall that the index of refraction depends not only on the direction of light propagation, which determines the matrix product on the left-hand side of Eq. (26), but also on the polarization given by the eigenvector. The simple diagonal form of this equation is due to the light propagation along one of the principal axes of the impermeability tensor. Section 6 treats the general case of an arbitrary direction of light propagation.

5. VISUALIZATION: THE INDEX ELLIPSOID OR OPTICAL INDICATRIX

This section introduces the visualization of a positive-definite tensor as an ellipsoid, which is particularly useful for the impermeability tensor.

We want to study how the impermeability tensor acts in different directions. Therefore, we have to remove its dependence on the magnitude of its argument $\mathbf{D}$. One way to do so, is to restrict our attention to arguments $\|\mathbf{D}\|_2 = 1$ with normalized Euclidean norm. However, equivalently, and more commonly used, is the normalization of the value $\mathbf{D}^T\boldsymbol{\eta}\mathbf{D} = 1$.

Due to the symmetry of the impermeability tensor, there is an eigenvector basis with respect to which the tensor is diagonal, with its eigenvalues on the diagonal. We consider all vectors $\mathbf{D}$ such that

$$\begin{aligned} &\begin{pmatrix} D_1&D_2&D_3 \end{pmatrix} \begin{pmatrix} \eta_1&0&0\\ 0&\eta_2&0\\ 0&0&\eta_3 \end{pmatrix} \begin{pmatrix} D_1\\ D_2\\ D_3 \end{pmatrix} \\ &\quad = \eta_1 D_1^2 + \eta_2 D_2^2 + \eta_3 D_3^2 = 1. \end{aligned} \tag{28}$$

Sections 3 and 4 have shown that the eigenvalues equal the reciprocal of the squared refractive index in the respective direction. So, the equation becomes

$$\frac{D_1^2}{n_1^2} + \frac{D_2^2}{n_2^2} + \frac{D_3^2}{n_3^2} = 1, \tag{29}$$

which is the equation of an ellipsoid with semi-axes of lengths n_1, n_2, and n_3 in the directions of the corresponding eigenvectors (see Fig. 1). This ellipsoid is called the **index ellipsoid** or **optical indicatrix** and is illustrated in Fig. 2.

The Principal Axes Theorem from Linear Algebra states that any symmetric tensor can be diagonalized using an orthogonal coordinate transformation, which in the three-dimensional case can even be chosen as a rotation. Thus, the equation

$$\begin{pmatrix} D_1&D_2&D_3 \end{pmatrix} \begin{pmatrix} \eta_{11}&\eta_{12}&\eta_{13}\\ \eta_{21}&\eta_{22}&\eta_{23}\\ \eta_{31}&\eta_{32}&\eta_{33} \end{pmatrix} \begin{pmatrix} D_1\\ D_2\\ D_3 \end{pmatrix} = 1 \tag{30}$$

with the positive-definite impermeability tensor describes a rotated ellipsoid.

Several mathematical concepts below are visualized using this index ellipsoid.

6. EIGENVALUE PROBLEM FOR A PRODUCT OF A PROJECTION AND A SYMMETRIC MATRIX

We will now consider the eigenvalue problem

$$(P_\mathbf{k}\boldsymbol{\eta})\mathbf{D}_0 = \alpha\mathbf{D}_0 \tag{31}$$

in three dimensions, $\mathbf{D}_0 \in \mathbb{R}^3$, for the orthogonal projection $P_\mathbf{k}$ along $\mathbf{k} \neq 0$ and the symmetric and invertible matrix $\boldsymbol{\eta}$ with inverse $\boldsymbol{\epsilon} = \boldsymbol{\eta}^{-1}$. In the setting of Section 4 the eigenvalue has the physical meaning $\alpha = (v_i/c_0)^2$.

Section 4 studied specific directions of $\mathbf{k}$ so that $P_\mathbf{k}$ and $\boldsymbol{\eta}$ have a common basis of eigenvectors. Now we allow an arbitrary direction $\mathbf{k} \neq \mathbf{0}$ as illustrated in Fig. 3 and compute the

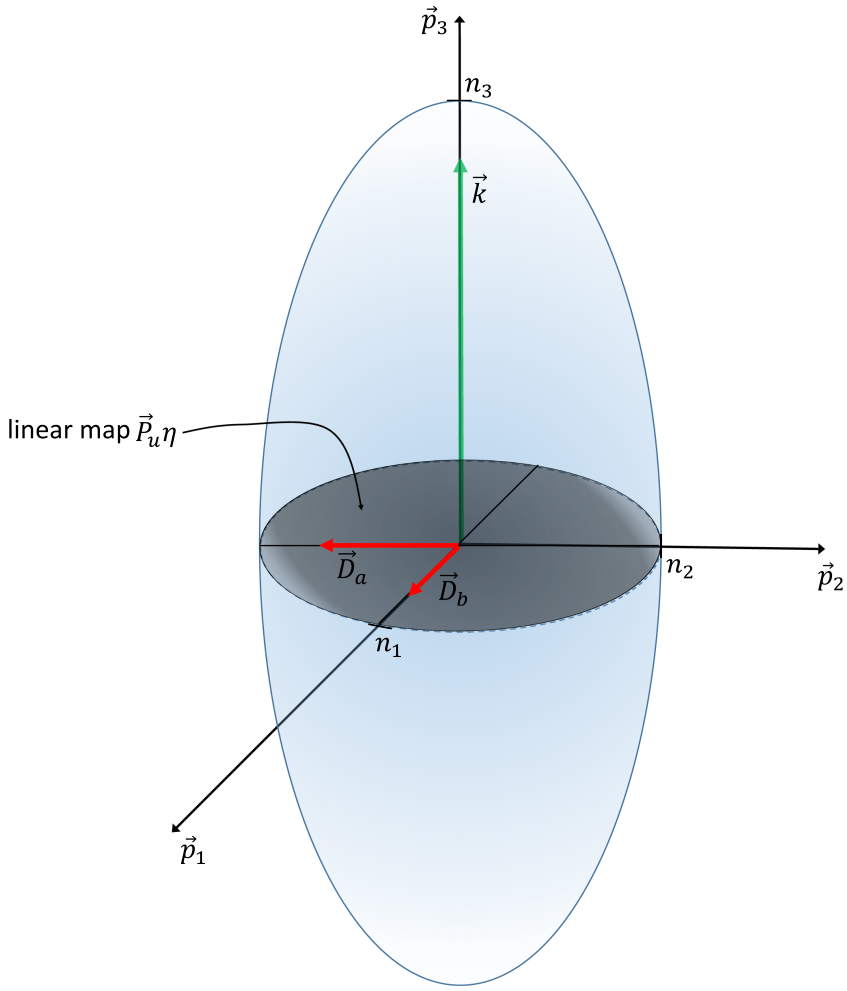

Fig. 1. Special case of a planar light wave propagating along one of the principal axes where $\mathbf{k} = \mathbf{p}_3$.

eigenvalues and the eigenvector basis of the product $P_{\mathbf{k}}\boldsymbol{\eta}$. We compute with respect to an orthonormal basis $(\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3)$ of eigenvectors of the projection $P_{\mathbf{k}}$, which has eigenvalues 1, 1, and 0.

For a given $\mathbf{k}$, we concretely compute such a coordinate system in the following way. An eigenvector of $P_{\mathbf{k}}$ with eigenvalue 0 is the vector $\mathbf{b}_3 = \mathbf{k}/\|\mathbf{k}\|_2$ normalized with respect to the Euclidean norm $\|\cdot\|_2$. In the case that $\mathbf{k} = (k_1, k_2, k_3)^T$ does not have a zero component; let $\mathbf{b}_1 = 1/(k_1^2 + k_2^2) \cdot (k_2, -k_1, 0)^T$. In the case that $\mathbf{k}$ has a zero component, say, $k_i = 0$ is the first zero component, let $\mathbf{b}_1 = (\delta_{1i}, \delta_{2i}, \delta_{3i})^T$, i.e., the vector with a 1 in the ith component and 0s in the other components. Generally, the so-called Kronecker delta is defined by $\delta_{ii} = 1$ and $\delta_{ij} = 0$ for $i \neq j$. In either case of $\mathbf{k}$, the vector $\mathbf{b}_1$ is orthogonal to $\mathbf{b}_3$ and we let $\mathbf{b}_2 = \mathbf{b}_3 \times \mathbf{b}_1$. Since $\mathbf{b}_1$ and $\mathbf{b}_2$ are orthogonal to $\mathbf{k}$, they are eigenvectors of $P_{\mathbf{k}}$ with eigenvalue 1.

With respect to this basis, the eigenvalue problem Eq. (31) has the coordinate representation

$$\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} = \alpha \begin{pmatrix} x \\ y \\ z \end{pmatrix}. \tag{32}$$

The matrix (η_{ij}) inherits the symmetry from the tensor $\boldsymbol{\eta}$ and the symmetric coordinate change:

$$\begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{pmatrix} = \begin{pmatrix} - & \mathbf{b}_1^T & - \\ - & \mathbf{b}_2^T & - \\ - & \mathbf{b}_3^T & - \end{pmatrix} \boldsymbol{\eta} \begin{pmatrix} | & | & | \\ \mathbf{b}_1 & \mathbf{b}_2 & \mathbf{b}_3 \\ | & | & | \end{pmatrix}. \tag{33}$$

To determine the eigenvalues, we multiply the matrices on the left-hand side of Eq. (32):

$$\begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} = \alpha \begin{pmatrix} x \\ y \\ z \end{pmatrix}. \tag{34}$$

The characteristic equation is

$$\begin{aligned} p(\alpha) &= \alpha[(\alpha - \eta_{11})(\alpha - \eta_{22}) - \eta_{12}\eta_{21}] \\ &= \alpha[\alpha^2 - (\eta_{11} + \eta_{22})\alpha + \underbrace{\eta_{11}\eta_{22} - \eta_{12}\eta_{21}}_{>0}], \end{aligned} \tag{35}$$

in which the constant coefficient in brackets is positive because it is a leading principal minor of a positive-definite matrix (Sylvester's Criterion). Hence, 0 is a single eigenvalue.

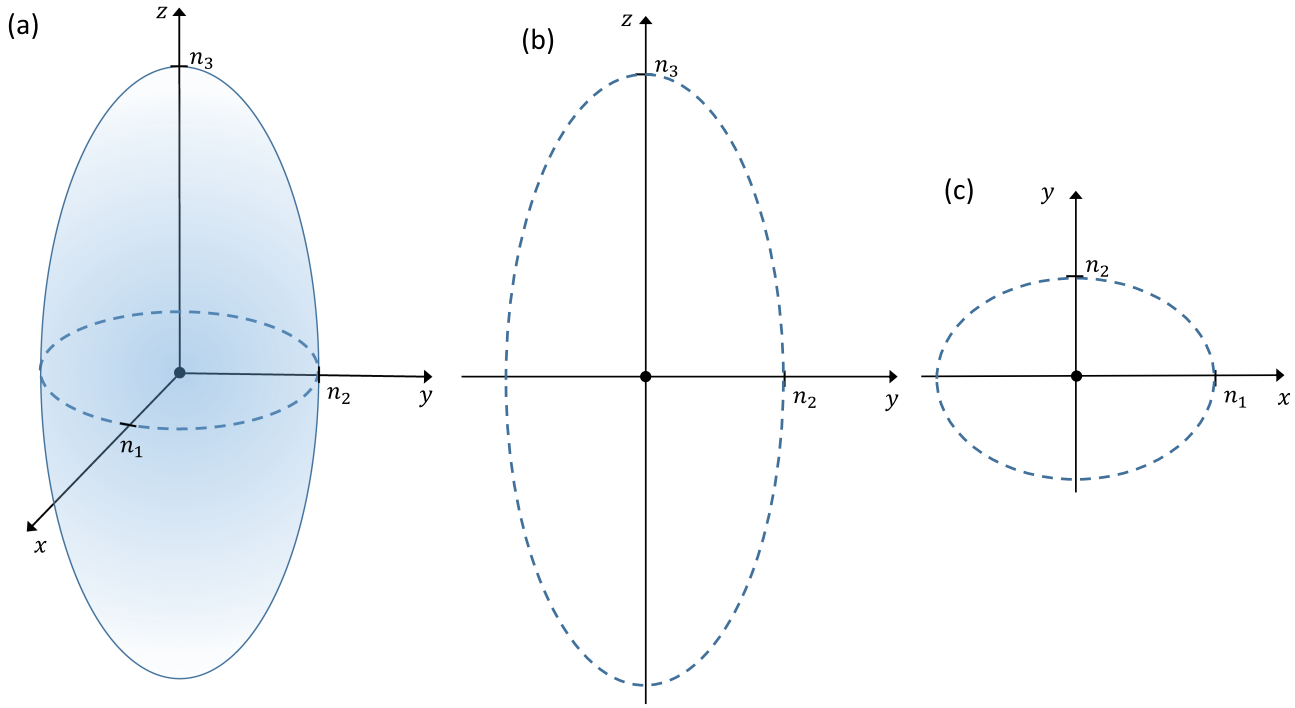

Fig. 2. (a) The tensor $\boldsymbol{\eta}$ is visualized as the index ellipsoid defined by $\eta_1 x^2 + \eta_2 y^2 + \eta_3 z^2 = 1$. Due to the relations $\eta_i = (1/n_i)^2$, $i = 1, 2, 3$, the equation also has the form $x^2/n_1^2 + y^2/n_2^2 + z^2/n_3^2 = 1$; therefore, the semi-axes indicated in the cross-sections of the ellipsoid, (b) and (c), are the indices of refraction.

The eigenvector $(\eta_{ij})^{-1}(0, 0, k)^T$ belongs to the eigenvalue 0. Its coordinate-free description is $\boldsymbol{\eta}^{-1}\mathbf{k} = \boldsymbol{\epsilon}\mathbf{k}$, i.e., the image of the wave propagation vector under the dielectric tensor.

Because of the zero row on the left-hand side of Eq. (34), eigenvectors for nonzero eigenvalues must have a coordinate representation $(x, y, 0)^T$. They lie in the plane onto which we project, which is normal to $\mathbf{k}$. The zero z-component reduces the problem to the two-dimensional symmetric eigenvalue problem

$$\begin{pmatrix} \eta_{11} & \eta_{12} \\ \eta_{21} & \eta_{22} \end{pmatrix} \begin{pmatrix} x \\ y \end{pmatrix} = \alpha \begin{pmatrix} x \\ y \end{pmatrix}, \quad \eta_{12} = \eta_{21}. \tag{36}$$

Therefore there are two real eigenvalues with orthogonal eigenvectors. The eigenvalues are the zeros of the characteristic polynomial

$$\begin{aligned} p(\alpha) &= (\eta_{11} - \alpha)(\eta_{22} - \alpha) - \eta_{12}^2 \\ &= \eta_{11}\eta_{22} - (\eta_{11} + \eta_{22})\alpha + \alpha^2 - \eta_{12}^2, \end{aligned} \tag{37}$$

that is,

$$\begin{aligned} \alpha_{1,2} &= \frac{\eta_{11} + \eta_{22}}{2} \pm \sqrt{\frac{(\eta_{11} + \eta_{22})^2}{4} - \eta_{11}\eta_{22} + \eta_{12}^2} \\ &= \frac{\eta_{11} + \eta_{22}}{2} \pm \sqrt{\frac{(\eta_{11} - \eta_{22})^2}{4} + \eta_{12}^2}. \end{aligned} \tag{38}$$

In the case $\eta_{12} = 0$, the eigenvalues are $\alpha_1 = \eta_{11}$ and $\alpha_2 = \eta_{22}$. In the case $\eta_{12} \neq 0$, the eigenvalues lie outside the interval with endpoints η_{11} and η_{22}. The further η_{12} is from zero, the further the eigenvalues are from the diagonal entries.

For the eigenvectors we have to solve the system

$$\begin{array}{cc|c} x & y & \\ \hline \eta_{11} - \alpha & 0 & 0 \\ 0 & \eta_{22} - \alpha & 0 \end{array} \quad \text{or} \quad \begin{array}{cc|c} x & y & \\ \hline \eta_{11} - \alpha & \eta_{12} & 0 \\ \eta_{12} & \eta_{22} - \alpha & 0, \end{array} \tag{39}$$

with $\eta_{12} = 0$ or $\eta_{12} \neq 0$, respectively, for $\alpha = \alpha_1$ and $\alpha = \alpha_2$.

In the case $\eta_{12} = 0$ the eigenvalues are $\alpha_1 = \eta_{11}$ and $\alpha_2 = \eta_{22}$. In the subcase $\eta_{11} \neq \eta_{22}$, the eigenvectors are $(1, 0)^T$ and $(0, 1)^T$. The coordinate representations of the eigenvectors of the original three-dimensional problem are $\mathbf{D}_0^{(1)} = (1, 0, 0)^T$ and $\mathbf{D}_0^{(2)} = (0, 1, 0)^T$. Coordinate-free, they are $\mathbf{D}_0^{(1)} = \mathbf{b}_1$ and $\mathbf{D}_0^{(2)} = \mathbf{b}_2$. In the subcase $\eta_{11} = \eta_{22}$, all nonzero vectors in the plane spanned by $\mathbf{b}_1$ and $\mathbf{b}_2$ are eigenvectors. In either case, the third eigenvector is $\mathbf{D}_0^{(3)} = \boldsymbol{\epsilon}\mathbf{k}$. Note that this vector need not be orthogonal to the others as the matrix in Eq. (34) is not necessarily symmetric.

In the case $\eta_{12} \neq 0$, the eigenvalues are different from the diagonal elements and the system reduces to

$$\begin{array}{cc|c} x & y & \\ \hline \eta_{11} - \alpha & \eta_{12} & 0 \\ 0 & \underbrace{(\eta_{22} - \alpha) - \dfrac{\eta_{12}^2}{\eta_{11} - \alpha}}_{=0} & 0. \end{array} \tag{40}$$

The lower right entry is zero for $\alpha = \alpha_1$ and $\alpha = \alpha_2$, since it corresponds to the characteristic polynomial.

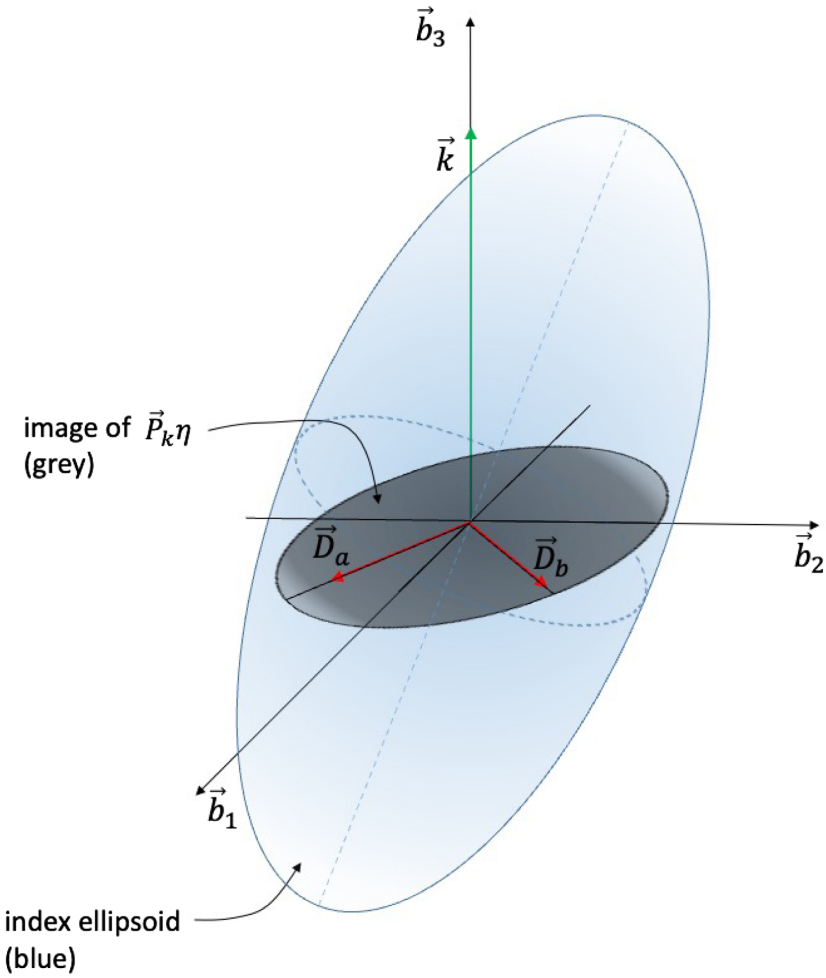

Fig. 3. Image of $P_{\mathbf{k}}\boldsymbol{\eta}$ is visualized while $\mathbf{k} \neq \mathbf{0}$ has an arbitrary direction.

The coordinate eigenvectors with respect to the basis $(\mathbf{b}_1, \mathbf{b}_2, \mathbf{b}_3)$ of the three-dimensional problem are

$$\mathbf{D}_0^{(1)} = \begin{pmatrix} \eta_{12} \\ \alpha_1 - \eta_{11} \\ 0 \end{pmatrix} \quad \text{and} \quad \mathbf{D}_0^{(2)} = \begin{pmatrix} \eta_{12} \\ \alpha_2 - \eta_{11} \\ 0 \end{pmatrix}. \tag{41}$$

Thus the eigenvectors are

$$\mathbf{D}_0^{(1)} = \eta_{12}\mathbf{b}_1 + \left(\frac{\eta_{22} - \eta_{11}}{2} - \sqrt{\frac{(\eta_{11} - \eta_{22})^2}{4} + \eta_{12}^2} \right) \mathbf{b}_2, \tag{42}$$

$$\mathbf{D}_0^{(2)} = \eta_{12}\mathbf{b}_1 + \left(\frac{\eta_{22} - \eta_{11}}{2} + \sqrt{\frac{(\eta_{11} - \eta_{22})^2}{4} + \eta_{12}^2} \right) \mathbf{b}_2, \tag{43}$$

$$\mathbf{D}_0^{(3)} = \epsilon\mathbf{k}. \tag{44}$$

7. VISUALIZATION: INTERSECTION OF INDEX ELLIPSOID AND PLANE

The eigenvalue problem $(P_{\mathbf{k}}\boldsymbol{\eta})\mathbf{D}_0 = \alpha\mathbf{D}_0$ of the product of the orthogonal projection along $\mathbf{k}$ and the impermeability tensor $\boldsymbol{\eta}$ has a nice geometric interpretation. It corresponds to finding the semi-axes of the ellipse, which is obtained as the intersection of the index ellipsoid and the normal plane $\mathbf{k}^{\perp}$, as illustrated in Figs. 4(a) and 4(b).

The points $\mathbf{r} = (x, y, z)^T$ in the normal plane $\mathbf{k}^{\perp}$ satisfy

$$0 = \mathbf{r} \cdot \mathbf{k} = x \cdot k_1 + y \cdot k_2 + z \cdot k_3. \tag{45}$$

As in Section 6 we rotate the coordinate system so that $\mathbf{k} = (0, 0, k)^T$. Then the points in $\mathbf{k}^{\perp}$ are characterized by $z = 0$. See Figs. 4(c) and 4(d).

We restrict the index ellipsoid equation

$$\begin{pmatrix} x & y & z \end{pmatrix} \begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} = 1 \tag{46}$$

to points in the xy-plane, $(x, y, 0)^T$, and obtain

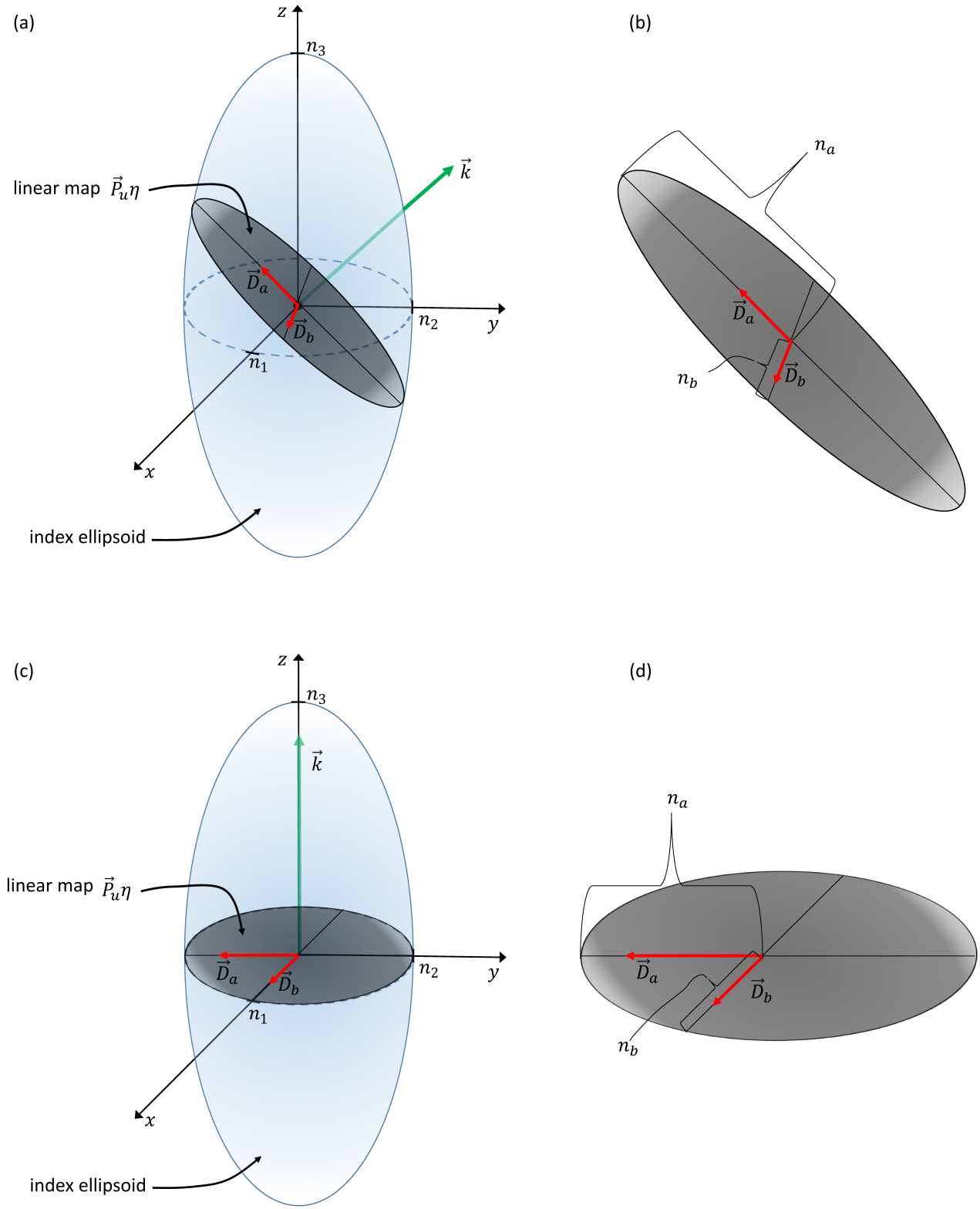

Fig. 4. (a) The linear map $P_{\mathbf{k}}\boldsymbol{\eta}$ is a projection onto a plane perpendicular to the light propagation along $\mathbf{k}$. The intersection of the index ellipsoid with this plane is called the index ellipse. (b) The eigenvalue problem $P_k\boldsymbol{\eta}\mathbf{v} = \alpha\mathbf{v}$ has the eigenvalue 0 in the direction $\mathbf{v} = \boldsymbol{\eta}^{-1}\mathbf{k}$ and the eigenvalues $1/n_a$ and $1/n_b$ related to the axes of the ellipse with eigenvectors $\vec{D}_a$ and $\vec{D}_b$ that are perpendicular to $\mathbf{k}$ and to each other. (c), (d) Rotated coordinate system so that $\mathbf{k} = (0, 0, k)^T$. Then the points in $\mathbf{k}^\perp$ are characterized by $z = 0$.

$$\begin{pmatrix} x & y \end{pmatrix} \begin{pmatrix} \eta_{11} & \eta_{12} \\ \eta_{21} & \eta_{22} \end{pmatrix} \begin{pmatrix} x \\ y \end{pmatrix} = 1. \tag{47}$$

This submatrix is symmetric and, by the leading principal minors criterion, inherits positive definiteness from $\boldsymbol{\eta}$. Hence this equation describes an ellipse. Its semi-axes are in the direction of the eigenvectors and the eigenvalues are $1/n_a^2$ and $1/n_b^2$, where n_a and n_b are the lengths of the semi-axes of the ellipse.

8. LINEAR AND QUADRATIC ELECTRO-OPTIC EFFECT

Now we apply an external electric field, which affects the impermeability tensor of the optical medium, and study the effects of these changes on light traveling through the optical medium.

We use the principal axes coordinate system of the impermeability tensor before the external field is applied. With respect to this coordinate system the impermeability tensor is diagonal. Then we apply the external electric field $\mathbf{E}$. The field affects the components of the impermeability tensor so that, usually, it is not diagonal anymore:

$$\boldsymbol{\eta}(0) = \begin{pmatrix} \eta_1 & 0 & 0 \\ 0 & \eta_2 & 0 \\ 0 & 0 & \eta_3 \end{pmatrix} \quad \overset{\mathbf{E}}{\rightarrow} \quad \boldsymbol{\eta}(\mathbf{E}) = \begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{pmatrix}. \tag{48}$$

The perturbed impermeability tensor $\boldsymbol{\eta}(\mathbf{E})$ is still symmetric and positive definite (see Appendix A). Then the corresponding equation

$$\eta_{11}x^2 + \eta_{22}y^2 + \eta_{33}z^2 + 2\eta_{23}yz + 2\eta_{31}zx + 2\eta_{12}xy = 1 \tag{49}$$

describes a rotated ellipsoid. But it need not be and usually is not a rotated version of $\boldsymbol{\eta}(\mathbf{0})$; the lengths of the semi-axes may have changed. Physically this means that the indices of refraction and the feasible polarization directions may have changed due to the externally applied electric field.

We write the dependence of the impermeability tensor on the external electric field as

$$\boldsymbol{\eta}(\mathbf{E}) = \boldsymbol{\eta}(\mathbf{0}) + \Delta\boldsymbol{\eta}(\mathbf{E}). \tag{50}$$

Its coordinate representation with respect to the principal axes of $\boldsymbol{\eta}(\mathbf{0})$ is

$$\boldsymbol{\eta}(\mathbf{E}) = \underbrace{\begin{pmatrix} 1/n_1^2 & 0 & 0 \\ 0 & 1/n_2^2 & 0 \\ 0 & 0 & 1/n_3^2 \end{pmatrix}}_{\boldsymbol{\eta}(\mathbf{0})} + \underbrace{\begin{pmatrix} \eta_{11} - 1/n_1^2 & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} - 1/n_2^2 & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} - 1/n_3^2 \end{pmatrix}}_{\Delta\boldsymbol{\eta}(\mathbf{E})}. \tag{51}$$

The components η_{ij} of $\boldsymbol{\eta}(\mathbf{E})$ depend in an unknown way on the applied external electric field. To model the unknown dependence approximately we use the multivariate Taylor polynomial up to quadratic degree for each tensor component about the center $\mathbf{E} = \mathbf{0}$:

$$\begin{aligned} \eta_{ij}(\mathbf{E}) &\approx \eta_{ij}(\mathbf{0}) + \sum_k \eta_{ij,k} E_k + \frac{1}{2} \sum_{k,l} \eta_{ij,kl} E_k E_l \\ &\approx \eta_{ij}(\mathbf{0}) + \begin{pmatrix} \eta_{ij,1} & \eta_{ij,2} & \eta_{ij,3} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix} \\ &\quad + \frac{1}{2} \begin{pmatrix} E_1 & E_2 & E_3 \end{pmatrix} \begin{pmatrix} \eta_{ij,11} & \eta_{ij,12} & \eta_{ij,13} \\ \eta_{ij,21} & \eta_{ij,22} & \eta_{ij,23} \\ \eta_{ij,31} & \eta_{ij,32} & \eta_{ij,33} \end{pmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix}, \end{aligned} \tag{52}$$

where $\eta_{ij}(\mathbf{0}) = \delta_{ij}/n_i^2$ with the Kronecker delta $\delta_{ii} = 1$ and $\delta_{ij} = 0$ for $i \neq j$. Moreover, the gradient and Hessian matrix with respective components

$$\eta_{ij,k} = \frac{\partial \eta_{ij}}{\partial E_k}(\mathbf{0}) \quad \text{and} \quad \eta_{ij,kl} = \frac{\partial^2 \eta_{ij}}{\partial E_k \partial E_l}(\mathbf{0}) \tag{53}$$

are the **linear electro-optical third-rank tensor** and the **quadratic electro-optical fourth-rank tensor** with constant components. The linear and the quadratic electro-optical tensors correspond to the **Pockels effect** and **DC Kerr effect**, respectively. The comma in the tensor indices indicates that the indices after the comma result from taking partial derivatives.

Since the impermeability tensor is symmetric, the linear and quadratic electro-optic tensors are symmetric in its first two indices, $\eta_{ij,k} = \eta_{ji,k}$ and $\eta_{ij,kl} = \eta_{ji,kl}$. Moreover, the quadratic electro-optical tensor is symmetric in the last two coefficients, $\eta_{ij,kl} = \eta_{ij,lk}$, because it is safe to assume that the dependence is sufficiently smooth and therefore that the order of the partial derivatives does not matter.

Because of the symmetries, only $6 \cdot 3 = 18$ of the $3^3 = 27$ components of the linear electro-optical tensor are independent: six independent component pairs $i \leq j$ times three independent partial derivatives. For the quadratic tensor $6 \cdot 6 = 36$ of $3^4 = 81$ components are independent: six independent component pairs $i \leq j$ times six independent second partial derivatives $k \leq l$.

It is customary and convenient to use the Voigt notation

$$\begin{aligned} \begin{bmatrix} \Delta\eta_1 \\ \Delta\eta_2 \\ \Delta\eta_3 \\ \Delta\eta_4 \\ \Delta\eta_5 \\ \Delta\eta_6 \end{bmatrix} &= \begin{bmatrix} \eta_{11} - \frac{1}{n_1^2} \\ \eta_{22} - \frac{1}{n_2^2} \\ \eta_{33} - \frac{1}{n_3^2} \\ \eta_{23} \\ \eta_{13} \\ \eta_{12} \end{bmatrix} = \begin{bmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \\ r_{41} & r_{42} & r_{43} \\ r_{51} & r_{52} & r_{53} \\ r_{61} & r_{62} & r_{63} \end{bmatrix} \begin{pmatrix} E_1 \\ E_2 \\ E_3 \end{pmatrix} \\ &\quad + \begin{bmatrix} s_{11} & s_{12} & s_{13} & s_{14} & s_{15} & s_{16} \\ s_{21} & s_{22} & s_{23} & s_{24} & s_{25} & s_{26} \\ s_{31} & s_{32} & s_{33} & s_{34} & s_{35} & s_{36} \\ s_{41} & s_{42} & s_{43} & s_{44} & s_{45} & s_{46} \\ s_{51} & s_{52} & s_{53} & s_{54} & s_{55} & s_{56} \\ s_{61} & s_{62} & s_{63} & s_{64} & s_{65} & s_{66} \end{bmatrix} \begin{bmatrix} E_1^2 \\ E_2^2 \\ E_3^2 \\ E_2E_3 \\ E_3E_1 \\ E_1E_2 \end{bmatrix}, \end{aligned} \tag{54}$$

in which the 18 coefficients r_{ij} correspond to the 18 independent tensor components of the linear electro-optical tensor:

$$r_{ij} = \eta_{ii,j}, \quad r_{4j} = \eta_{23,j} = \eta_{32,j}, \quad r_{5j} = \eta_{13,j} = \eta_{31,j},$$
$$r_{6j} = \eta_{12,j} = \eta_{21,j} \quad \text{for} \quad 1 \le i,\, j \le 3, \tag{55}$$

The 36 coefficients $s_{k\ell}$ are linear combinations of the 36 independent tensor components of the quadratic electro-optical tensor:

$$s_{ij} = \eta_{ii,jj} \quad \text{for} \quad 1 \le i,\, j \le 3, \tag{56}$$

$$s_{4j} = \eta_{12,jj}, \quad s_{5j} = \eta_{13,jj}, \quad s_{6j} = \eta_{23,jj} \quad \text{for} \quad 1 \le j \le 3 \tag{57}$$

$$s_{i4} = \eta_{ii,12} + \eta_{ii,21} = 2\eta_{ii,12},$$
$$s_{i5} = \eta_{ii,13} + \eta_{ii,31} = 2\eta_{ii,13},$$
$$s_{i6} = \eta_{ii,23} + \eta_{ii,32} = 2\eta_{ii,23} \quad \text{for} \quad 1 \le i \le 3, \tag{58}$$

$$s_{44} = 2\eta_{12,12}, \quad s_{45} = 2\eta_{12,13}, \quad s_{46} = 2\eta_{12,23},$$
$$s_{54} = 2\eta_{13,12}, \quad s_{55} = 2\eta_{13,13}, \quad s_{56} = 2\eta_{13,23},$$
$$s_{64} = 2\eta_{23,12}, \quad s_{65} = 2\eta_{23,13}, \quad s_{66} = 2\eta_{23,23}. \tag{59}$$

Due to the symmetries, both linear systems are invertible. Therefore, Voigt coefficients determined by measurements can be converted back into tensor components.

Equation (54) is not a tensor equation because the components do not transform under coordinate changes the way tensors do. See Section 9 below. With the exception of the vector $(E_1, E_2, E_3)^T$ none of these quantities is a tensor; we use brackets instead of parentheses to emphasize this fact. Each index of a tensor in three-dimensional space must use exactly three values, corresponding to the number of dimensions. These vectors and matrices do not transform like tensors under coordinate changes. However, this non-tensor representation has the advantages that we neither have to write entries that repeat due to symmetry nor do we have to write tensors of rank three or even four. The disadvantage of using this coordinate-specific representation is negligible, since we do not change the coordinate system. We consistently use the principal axes of the dielectric tensor without an external electric field.

We are going to study how the index ellipsoid changes due to the linear electro-optical (Pockels) effect or the quadratic electro-optic (DC Kerr) effect. For centrosymmetric materials any change in the impermeability coefficients η_{ij} in one direction must be the same in the opposite direction. Hence the gradient $\eta_{ij,k}$ must be zero. Section 10 contains a more formal proof that the linear electro-optic tensor vanishes for centrosymmetric materials. While the linear electro-optical effect requires non-centrosymmetric material, the quadratic electro-optical effect occurs in centrosymmetric and non-centrosymmetric materials. Consequently, both effects need to be taken into account for non-centrosymmetric materials.

9. TENSOR QUANTITIES AND THEIR TRANSFORMATION BEHAVIOR

Many crystalline structures feature symmetries in the sense that they look the same when viewed from a specifically rotated perspective or in a mirror. These changes of perspective correspond to linear changes of coordinates: rotation about a certain axis by a certain angle, reflection about a certain plane, or inversion. A symmetry of the material means that the material is invariant under some coordinate change.

A key property of tensors is that their components, which depend on the chosen coordinate system, transform in a very specific linear or multilinear way, when the coordinate system is changed. The specific coordinate transformation depends on the rank of the tensor.

A scalar quantity such as temperature is a tensor of rank zero. A scalar is invariant under coordinate system changes. It has the same value in each coordinate system.

A position vector $(x_1, x_2, x_3)^T$ is an example of a vector or tensor of rank one, i.e., a tensor with one index. Its transformation is directly derived from the transformation of the coordinate system. Say we change from a coordinate system denoted by capital letters to a coordinate system denoted by small letters. The *small* coordinate-axis vectors $\hat{\mathbf{x}}_1, \hat{\mathbf{x}}_2, \hat{\mathbf{x}}_3$ have coordinates with respect to the *capital* vectors $\hat{\mathbf{X}}_1, \hat{\mathbf{X}}_2, \hat{\mathbf{X}}_3$, say,

$$\hat{\mathbf{x}}_1 = T_{11} \cdot \hat{\mathbf{X}}_1 + T_{21} \cdot \hat{\mathbf{X}}_2 + T_{31} \cdot \hat{\mathbf{X}}_3,$$
$$\hat{\mathbf{x}}_2 = T_{12} \cdot \hat{\mathbf{X}}_1 + T_{22} \cdot \hat{\mathbf{X}}_2 + T_{31} \cdot \hat{\mathbf{X}}_3,$$
$$\hat{\mathbf{x}}_3 = T_{13} \cdot \hat{\mathbf{X}}_1 + T_{23} \cdot \hat{\mathbf{X}}_2 + T_{33} \cdot \hat{\mathbf{X}}_3,$$

that is,

$$\hat{\mathbf{x}}_j = \sum_{i=1}^{3} T_{ij} \hat{\mathbf{X}}_i, \quad \text{for} \quad j = 1, 2, 3, \tag{60}$$

or in matrix form

$$\left(\hat{\mathbf{x}}_1\ \hat{\mathbf{x}}_2\ \hat{\mathbf{x}}_3\right) = \left(\hat{\mathbf{X}}_1\ \hat{\mathbf{X}}_2\ \hat{\mathbf{X}}_3\right) \begin{pmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{pmatrix}. \tag{61}$$

An arbitrary vector is a linear combination of the coordinate vectors and their coefficients in either coordinate system. Therefore,

$$\left(X_1\ X_2\ X_3\right) \begin{pmatrix} \hat{\mathbf{X}}_1 \\ \hat{\mathbf{X}}_2 \\ \hat{\mathbf{X}}_3 \end{pmatrix} = \left(x_1\ x_2\ x_3\right) \begin{pmatrix} \hat{\mathbf{x}}_1 \\ \hat{\mathbf{x}}_2 \\ \hat{\mathbf{x}}_3 \end{pmatrix}$$
$$= \left(x_1\ x_2\ x_3\right) \begin{pmatrix} T_{11} & T_{21} & T_{31} \\ T_{12} & T_{22} & T_{32} \\ T_{13} & T_{23} & T_{33} \end{pmatrix} \begin{pmatrix} \hat{\mathbf{X}}_1 \\ \hat{\mathbf{X}}_2 \\ \hat{\mathbf{X}}_3 \end{pmatrix}. \tag{62}$$

Comparing the coefficients of the capital basis vectors tells us that the coordinates transform according to $X_i = \sum_j T_{ij} x_j$ for $i = 1, 2, 3$, which is written in matrix form as

$$\left(X_1\ X_2\ X_3\right) = \left(x_1\ x_2\ x_3\right) \begin{pmatrix} T_{11} & T_{21} & T_{31} \\ T_{12} & T_{22} & T_{32} \\ T_{13} & T_{23} & T_{33} \end{pmatrix} \tag{63}$$

or—to put the matrix indices in the standard order—as the transposed matrix product

$$\begin{pmatrix} X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} T_{11} & T_{12} & T_{13} \\ T_{21} & T_{22} & T_{23} \\ T_{31} & T_{32} & T_{33} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix}. \tag{64}$$

On the one hand, triples of quantities that transform like the coordinates of a vector,

$$Q_j = \sum_{i=1}^{3} T_{ji} q_i, \quad \text{for} \quad j = 1, 2, 3, \tag{65}$$

form a contravariant tensor of rank one. On the other hand, any triple of quantities that transforms like the triple of coordinate vectors,

$$q_j = \sum_{i=1}^{3} T_{ij} Q_i, \quad \text{for} \quad j = 1, 2, 3, \tag{66}$$

is called a covariant vector or covariant tensor of rank one. These transformations differ because the matrix is transposed and the roles of the small and the capital quantities are exchanged, which corresponds to an additional inversion of the matrix.

The equality $\sum_{i=1}^{3} x_i \hat{\mathbf{x}}_i = \sum_{j=1}^{3} X_j \hat{\mathbf{X}}_j$ resulting from describing the same object in two coordinate systems is the prototype of the concept of duality in tensors. (Alas, there is no single definition of duality; see [31].) The product of a contravariant and a covariant vector is an invariant quantity under coordinate changes. For example, the electric field $\mathbf{E} = \sum_j E_j \hat{\mathbf{X}}_j = \sum_i e_i \hat{\mathbf{x}}_i$ is an instance of an invariant vector quantity. Its coordinates form a contravariant tensor of rank one.

The impermeability tensor is a covariant tensor of rank two because combining it with two contravariant vectors produces an invariant scalar. We transform the vectors in that product:

$$\begin{aligned} \sum_{k,\ell} \eta_{k\ell} x_k y_\ell &= \sum_{i,j} H_{ij} X_i Y_j \\ &= \sum_{i,j} H_{ij} \sum_k T_{ik} x_k \sum_\ell T_{j\ell} y_\ell \\ &= \sum_{k,\ell} \left(\sum_{i,j} H_{ij} T_{ik} T_{j\ell} \right) x_k y_\ell. \end{aligned} \tag{67}$$

Since this relationship has to hold for all contravariant vectors $(x_1, x_2, x_3)^T$ and $(y_1, y_2, y_3)^T$, we obtain the transformation rule for the components of the impermeability tensor:

$$\eta_{k\ell} = \sum_{ij} H_{ij} T_{ik} T_{j\ell}. \tag{68}$$

This can also be written as a matrix product involving the transposed transformation matrix on the left:

$$\begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{21} & \eta_{22} & \eta_{23} \\ \eta_{31} & \eta_{32} & \eta_{33} \end{pmatrix} = T^T \begin{pmatrix} H_{11} & H_{12} & H_{13} \\ H_{21} & H_{22} & H_{23} \\ H_{31} & H_{32} & H_{33} \end{pmatrix} T. \tag{69}$$

10. RELATIONS BETWEEN TENSOR COMPONENTS DUE TO SYMMETRIES OF THE MATERIAL

If a physical quantity of a material is described by a tensor, then the tensor components inherit relations from the symmetries of the material. If the material is invariant under a certain coordinate transformation, then the transformed tensor components must agree with the original ones.

More concretely, if the material that is described by the symmetric tensor $\boldsymbol{\eta}$ of rank two is invariant under the coordinate transformation, then the tensor components are invariant as well, that is, $H_{ij} = \eta_{ij}$. Hence each symmetry of the material under a coordinate transformation T induces relations

$$\eta_{k\ell} = \sum_{ij} \eta_{ij} T_{ik} T_{j\ell}. \tag{70}$$

For example, consider a rotation about the $\hat{\mathbf{X}}_3$-axis by 90°. It has the following effect on the coordinate-axis vectors:

$$\hat{\mathbf{x}}_1 = T(\hat{\mathbf{X}}_1) = \hat{\mathbf{X}}_2 = 0 \cdot \hat{\mathbf{X}}_1 + 1 \cdot \hat{\mathbf{X}}_2 + 0 \cdot \hat{\mathbf{X}}_3, \tag{71}$$

$$\hat{\mathbf{x}}_2 = T(\hat{\mathbf{X}}_2) = -\hat{\mathbf{X}}_1 = -1 \cdot \hat{\mathbf{X}}_1 + 0 \cdot \hat{\mathbf{X}}_2 + 0 \cdot \hat{\mathbf{X}}_3, \tag{72}$$

$$\hat{\mathbf{x}}_3 = T(\hat{\mathbf{X}}_3) = \hat{\mathbf{X}}_3 = 0 \cdot \hat{\mathbf{X}}_1 + 0 \cdot \hat{\mathbf{X}}_2 + 1 \cdot \hat{\mathbf{X}}_3, \tag{73}$$

and the transformation matrix is the transposed coefficient scheme

$$T = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{74}$$

If a material is symmetric under this rotation, the tensor component relations will be

$$\begin{aligned} \begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{12} & \eta_{22} & \eta_{23} \\ \eta_{13} & \eta_{23} & \eta_{33} \end{pmatrix} &= \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \eta_{11} & \eta_{12} & \eta_{13} \\ \eta_{12} & \eta_{22} & \eta_{23} \\ \eta_{13} & \eta_{23} & \eta_{33} \end{pmatrix} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} \\ &= \begin{pmatrix} \eta_{22} & -\eta_{12} & \eta_{23} \\ -\eta_{12} & \eta_{11} & -\eta_{13} \\ \eta_{23} & -\eta_{13} & \eta_{33} \end{pmatrix}, \end{aligned} \tag{75}$$

where we have already incorporated the tensor symmetry $\eta_{ij} = \eta_{ji}$, by using the representative η_{ij} with $i \le j$.

We read off $\eta_{11} = \eta_{22}$. Also, $\eta_{12} = -\eta_{12}$ and, hence, $\eta_{12} = 0$. Finally, $\eta_{13} = \eta_{23} = -\eta_{13} = -\eta_{23}$ and, thus, $\eta_{13} = \eta_{23} = 0$. A fourfold rotational symmetry about the x_3-axis forces that the impermeability tensor with respect to a Cartesian coordinate system with the third axis along the x_3-axis is of the form

$$\begin{pmatrix} \eta_o & 0 & 0 \\ 0 & \eta_o & 0 \\ 0 & 0 & \eta_e \end{pmatrix}, \tag{76}$$

where $\eta_{11} = \eta_{22} = \eta_o$ and $\eta_{33} = \eta_e$ are called **ordinary and extraordinary impermeability**, respectively. Similar to this

definition, an ordinary and extraordinary refractive index is used in optics, which is discussed in Section 11.

A fascinating aspect of this example is that fourfold rotational symmetry of a material about an axis implies full rotational symmetry of the impermeability tensor about that axis.

Next, let us consider tensors of higher rank. The linear electro-optical tensor is covariant of rank three, so it transforms according to

$$\eta_{ij,k} = \sum_{m,n,r} H_{mn,r}\, T_{mi}\, T_{nj}\, T_{rk}. \tag{77}$$

The coordinate transformation applies to each index, even the one belonging to partial differentiation. For the quadratic electro-optical tensor, which is a covariant of rank four, we get analogously

$$\eta_{ij,k\ell} = \sum_{m,n,r,s} H_{mn,rs}\, T_{mi}\, T_{jn}\, T_{rk}\, T_{s\ell}. \tag{78}$$

As a symmetry example, we now consider the inversion

$$\begin{pmatrix} X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix}, \quad \text{i.e.,} \quad T_{ij} = -\delta_{ij}. \tag{79}$$

The linear electro-optical tensor transforms according to

$$\eta_{ij,k} = \sum_{m,n,r} (-1)^3 H_{mn,r}\, \delta_{mi}\delta_{nj}\delta_{rk} = -H_{ij,k}. \tag{80}$$

If the crystalline structure is invariant under inversion, that is, if it is centrosymmetric, then we have

$$\eta_{ij,k} = -\eta_{ij,k}, \text{ i.e., } \quad \eta_{ij,k} = 0 \quad \text{for all} \quad i,\, j,\, k. \tag{81}$$

In the centrosymmetric case the linear electro-optical tensor is zero. There is no linear electro-optic effect.

For the quadratic electro-optic tensor the inversion transformation produces the relations

$$\eta_{ij,k\ell} = \sum_{m,n,r,s} (-1)^4 H_{mn,r}\, \delta_{mi}\delta_{nj}\delta_{rk}\delta_{s\ell} = H_{ij,k\ell}. \tag{82}$$

Invariance of the material under inversion only leads to the tautology

$$\eta_{ij,k\ell} = \eta_{ij,k\ell}. \tag{83}$$

Inversion symmetry does not restrict the quadratic electro-optic tensor.

As another example we consider the reflection about the x_2x_3-plane:

$$\begin{pmatrix} X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix}. \tag{84}$$

The linear electro-optic tensor transforms according to

$$\eta_{ij,k} = \sum_{m,n,r} (-1)^{\ell} H_{mn,r}\, \delta_{mi}\delta_{nj}\delta_{rk} = (-1)^{\ell} H_{ij,k}, \tag{85}$$

where ℓ counts how often the index 1 occurs in i, j, and k. If the crystalline structure is invariant under this reflection, then we have $\eta_{ij,k} = -\eta_{ij,k}$, whenever the index 1 occurs an odd number of times. There is no restriction if the index 1 occurs an even number of times. Thus,

$$\eta_{ij,k} = 0, \quad \text{if index 1 occurs an odd number of times.} \tag{86}$$

As the final symmetry example in this context, we consider the coordinate transformation that exchanges the x_1 and the x_2 variable:

$$\begin{pmatrix} X_1 \\ X_2 \\ X_3 \end{pmatrix} = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix}. \tag{87}$$

Geometrically, this transformation is the reflection about the plane spanned by the x_3-axis and the bisecting line between the x_1-axis and x_2-axis.

The effect of this transformation is an exchange of tensor components whose position indices differ by an exchange of the index value 1 and the index value 2. For instance,

$$\eta_{12,1} = H_{21,2} \quad \text{and} \quad \eta_{33,2} = H_{33,1}. \tag{88}$$

If the crystalline structure is symmetric about the plane spanned by the x_3-axis and the x_1x_2-bisector, then the exchanged components of the linear electro-optic tensor must agree, e.g.,

$$\eta_{12,1} = \eta_{21,2} \quad \text{and} \quad \eta_{33,2} = \eta_{33,1}. \tag{89}$$

Similarly, we obtain the quadratic electro-optic tensor relationships of the form

$$\eta_{12,13} = \eta_{21,23} \quad \text{and} \quad \eta_{33,22} = \eta_{33,11}, \tag{90}$$

in which the index va1ues 1 and 2 are exchanged. This includes indices belonging to partial differentiation.

As shown by the preceding examples, symmetries of the material provide valuable information about tensor quantities.

11. EXAMPLE OF SYMMETRY CONSTRAINTS: CHANGE OF REFRACTIVE INDEX OF BARIUM TITANATE DUE TO AN EXTERNAL ELECTRIC FIELD

Electro-optical effects in anisotropic materials play a major role in science and technology. For example, lithium niobate has been the key material for telecommunication and laser technologies for 50 years, due to its—for a long time unrivaled—large Pockels effect of an externally applied electric field on the refractive index of the material. The Pockels effect is described by the linear electro-optical tensor, whose largest component for lithium niobate is 32.8 pm/V. On the basis of this size, the theory and mathematical foundations for electro-optical effects have been developed and used for 50 years. However, progress in material engineering has led to materials with dramatically larger Pockels effects. The ceramic barium titanate (BTO), though already having appeared in the Feynman Lectures ([28], Volume II, 11-7 Ferroelectricity: $BaTiO_3$), has become prominent in applications rather recently [32–34]. It possesses an electro-optical tensor component of 1300 pm/V [35]. Therefore, the mathematical models and, in particular, the original linear approximations for electro-optical

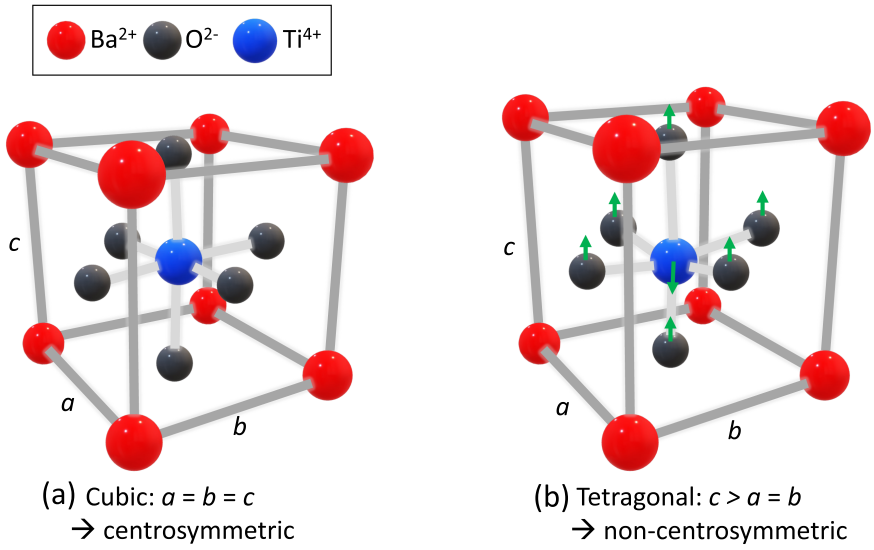

Fig. 5. Crystal structure of BTO (a) above Curie temperature and (b) below. The ionic radii relative to the size of the unit cell are substantially larger than indicated in the figures.

effects need to be updated. Linearization errors have become non-negligible.

First we study the crystallographic point group of BTO, a biaxial crystal, to understand the origin of its electro-optical effect. Then we derive how the index ellipsoid for BTO gets rotated by applying an external electric field. Finally, we compute the principal values (eigenvalues) of this rotated ellipsoid and determine the change of the refractive index due to the external electric field.

BTO has a Curie temperature of 120°C [36]. Above this temperature, BTO is paraelectric in the cubic phase: see Fig. 5; barium ions Ba^{2+} are located at the corners of a cube. A titanium ion Ti^{4+} is at the center of the cube, and oxygen ions O^{2-} are at the center of the faces of the cube. The oxygen ions form the vertices of an octahedron. In this phase BTO is centrosymmetric, and, therefore, does not feature a linear electro-optic effect. Its point group in the Hermann-Mauguin notation is $m3m$, that is, there is a mirror symmetry about each of the cubic faces ($m\cdot\cdot$), a threefold rotational symmetry about the space diagonals ($\cdot 3\cdot$), and a mirror symmetry about the planes through one edge and the bisector line of the other two edges ($\cdot\cdot m$).

Below the Curie temperature BTO is ferroelectric and its fundamental cell is a parallelepiped with a square base and a height that is only slightly longer than the base sides [37]. As in the cubic phase the barium ions are located at the corners and the oxygen ions at the center of the faces (Fig. 5). The titanium ion however is not located at the center, but slightly shifted in vertical direction. Because of the shift, this structure is non-centrosymmetric. In this phase BTO possesses tetragonal symmetry with the point group 4 mm: there is fourfold rotational symmetry about the longer edges ($4\cdot\cdot$), mirror symmetry about the vertical faces ($\cdot m\cdot$), and mirror symmetry about the planes through a longer edge and the bisector line of two shorter edges ($\cdot\cdot m$).

The tetragonal phase possesses a linear electro-optical (Pockels) effect. It originates from the shift of the Ti^{4+} ion from the center toward an oxygen ion (O^{2-}) at one of the face centers of the unit cell. An external electric field induces a redistribution of the bond charges and possibly a slight deformation of the ion lattice. As net result, the optical impermeability tensor is changed.

This section only considers the linear electro-optical effect of BTO in the tetragonal phase. We use a coordinate system with the x-axis and y-axis parallel to the edges along the square base of the fundamental cell. The z-axis is parallel to the elongated height.

With respect to the principal axes coordinate system (without an external electric field) we have

$$\boldsymbol{\eta}(\mathbf{E}) \approx \begin{pmatrix} \frac{1}{n_1^2} & 0 & 0 \\ 0 & \frac{1}{n_2^2} & 0 \\ 0 & 0 & \frac{1}{n_3^2} \end{pmatrix} + \underbrace{\begin{pmatrix} \sum \eta_{11,k}E_k & \sum \eta_{12,k}E_k & \sum \eta_{13,k}E_k \\ \sum \eta_{21,k}E_k & \sum \eta_{22,k}E_k & \sum \eta_{23,k}E_k \\ \sum \eta_{31,k}E_k & \sum \eta_{32,k}E_k & \sum \eta_{33,k}E_k \end{pmatrix}}_{=\Delta\boldsymbol{\eta}^{(1)}},$$

$$\text{with} \quad \eta_{ij,k} = \eta_{ji,k} \quad \text{and sums over} \quad k = 1, 2, 3. \tag{91}$$

Because of the fourfold rotational symmetry along the z-direction we have $n_1 = n_2$. The longer height indicates that n_3 differs from the other two principal indices of refraction. If two refractive indices are equal, the common value is called the **ordinary refractive index** n_o and the deviant refractive index is the **extraordinary refractive index** n_e. The **optical axis** of the crystal is always normal to the plane belonging to the ordinary refractive index. The optical axis of the crystal is a different concept from the optical axis of the light beam, which is parallel to k. See Fig. 6: the optical axis of the crystal is along the z-axis while the direction of light propagation **k** is different from the z-axis.

The Hermann-Mauguin notation of BTO—below the Curie temperature—is 4 mm, so the symmetries are: (1) fourfold

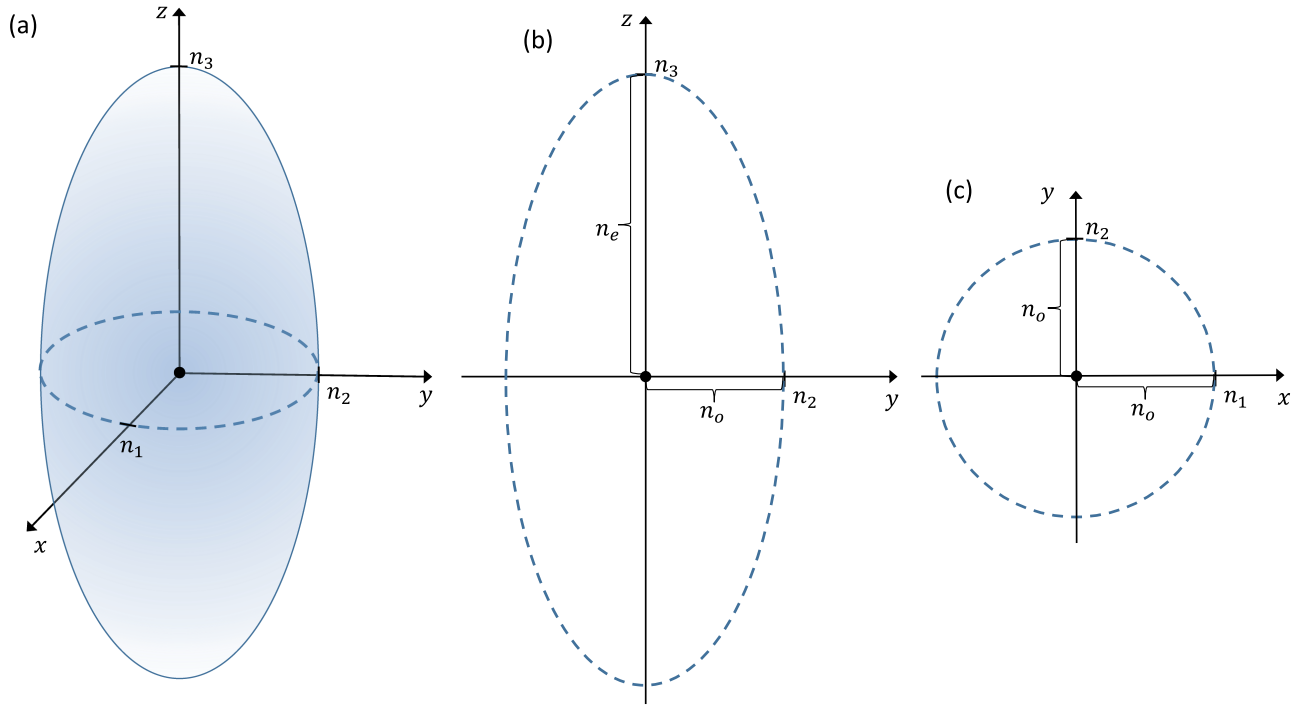

Fig. 6. (a) The tensor $\boldsymbol{\eta}$ is visualized by the so-called index ellipsoid $\eta_1 D_x^2 + \eta_2 D_y^2 + \eta_3 D_z^2 = 1$. Due to the relations $\eta_i = (1/n_i)^2$, $i = 1, 2, 3$, the semi-axes are the indices of refraction. This particular ellipsoid and its cross-sections in (b) and (c) show the special case $n_1 = n_2 \neq n_3$, which fits the case of barium titanate.

rotational symmetry about the z-axis, (2) reflections about the yz-plane and about the xz-plane, and (3) reflection about the plane spanned by the z-axis and the xy-bisector, that is, exchange of the x and y coordinates. The corresponding coordinate transformations are

$$\begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \text{and} \quad \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{92}$$

The use of symmetry arguments in this setting is subtle. Of course, if we apply an external electric field, then the ions in BTO will move and the symmetry will be broken. The above symmetries only hold in the absence of the external electric field. But the tensor components that we are going to study are actually the function values and derivatives with argument $\mathbf{E} = \mathbf{0}$.

As seen in Section 10, the fourfold rotational symmetry implies

$$\boldsymbol{\eta}(\mathbf{0}) = \begin{pmatrix} \eta_o & 0 & 0 \\ 0 & \eta_o & 0 \\ 0 & 0 & \eta_e \end{pmatrix} \tag{93}$$

for the tensor values without an external electric field. The other symmetries do not add any more constraints.

For the partial derivatives tensor $\eta_{ij,k}$ of rank three, we start with the other symmetries. The reflection symmetries about the yz-plane and the xz-plane lead to $\eta_{ij,k} = 0$ if an odd number of indices is 1 or if an odd number of indices is 2. Finally, the bisector symmetry stipulates that two tensor coefficients are equal when their index tuples transform into one another by exchanging the values 1 and 2. Overall, we obtain (we use u, v, and w to keep track of equal values due to bisector symmetry)

$$\begin{aligned} &\eta_{11,1} = 0 \quad \eta_{12,1} = 0 \quad \eta_{13,1} = v \\ &\eta_{11,2} = 0 \quad \eta_{12,2} = 0 \quad \eta_{13,2} = 0 \\ &\eta_{11,3} = u \quad \eta_{12,3} = 0 \quad \eta_{13,3} = 0 \\ \\ &\eta_{22,1} = 0 \quad \eta_{23,1} = 0 \\ &\eta_{22,2} = 0 \quad \eta_{23,2} = v \\ &\eta_{22,3} = u \quad \eta_{23,3} = 0 \\ \\ &\eta_{33,1} = 0 \\ &\eta_{33,2} = 0 \\ &\eta_{33,3} = w. \end{aligned} \tag{94}$$

The fourfold rotation does not add any more constraints since the rotation is obtained by first reflecting about the xy-bisector and z-axis plane and then reflecting about the yz-plane:

$$\begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} = \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{95}$$

We summarize the results efficiently in the Pockels matrix

$$\begin{bmatrix}\Delta\eta_1\\ \Delta\eta_2\\ \Delta\eta_3\\ \Delta\eta_4\\ \Delta\eta_5\\ \Delta\eta_6\end{bmatrix} = \begin{bmatrix}\eta_{11}(\mathbf{E})-\frac{1}{n_1^2}\\ \eta_{22}(\mathbf{E})-\frac{1}{n_2^2}\\ \eta_{33}(\mathbf{E})-\frac{1}{n_3^2}\\ \eta_{23}(\mathbf{E})\\ \eta_{31}(\mathbf{E})\\ \eta_{12}(\mathbf{E})\end{bmatrix} = \begin{bmatrix}0&0&u\\0&0&u\\0&0&w\\0&v&0\\v&0&0\\0&0&0\end{bmatrix}\begin{pmatrix}E_1\\E_2\\E_3\end{pmatrix}$$

$$= \begin{bmatrix}0&0&r_{13}\\0&0&r_{23}\\0&0&r_{33}\\0&r_{42}&0\\r_{51}&0&0\\0&0&0\end{bmatrix}\begin{pmatrix}E_1\\E_2\\E_3\end{pmatrix}. \tag{96}$$

Our goal is to find an expression for the refractive index change due to the external electric field. The refractive index change depends not only on the anisotropic material but also on the polarization of the light and the orientation of the external electric field. First, we read off the entries $\eta_{ij}(\mathbf{E})$ from Eq. (96) (special case: if $E_1 = E_2 = 0$, then the fourfold rotational symmetry still holds, and consistent with the symmetry, the tensor is diagonal with the first two entries being equal because $n_1 = n_2$ and $r_{13} = r_{23}$; for a suitable E_3—moving the titanium ion into the center—all three eigenvalues will agree):

$$\boldsymbol{\eta}(\mathbf{E}) = \begin{pmatrix}\frac{1}{n_1^2}+r_{13}E_3 & 0 & r_{51}E_1\\ 0 & \frac{1}{n_2^2}+r_{23}E_3 & r_{42}E_2\\ r_{51}E_1 & r_{42}E_2 & \frac{1}{n_3^2}+r_{33}E_3\end{pmatrix}. \tag{97}$$

This tensor is characterized by the rotated ellipsoid defined by

$$\left(\frac{1}{n_1^2}+r_{13}E_3\right)x^2+\left(\frac{1}{n_2^2}+r_{23}E_3\right)y^2+\left(\frac{1}{n_3^2}+r_{33}E_3\right)z^2 + 2r_{42}E_2yz+2r_{51}E_1zx=1. \tag{98}$$

We compute its semi-axes. Their lengths are the reciprocals of the square-roots of the eigenvalues and their directions are given by the respective eigenvectors.

Since the crystalline structure of BTO in the x-direction equals the structure in the y-direction, $n_1 = n_2$ and $r_{13} = r_{23}$, this matrix is of the form

$$\boldsymbol{\eta}(\mathbf{E}) = \begin{pmatrix}A&0&C\\0&A&D\\C&D&B\end{pmatrix}. \tag{99}$$

Its characteristic polynomial is

$$\begin{aligned}p(\lambda) &= (A-\lambda)^2(B-\lambda) - C^2(A-\lambda) - D^2(A-\lambda)\\ &= (A-\lambda)[(A-\lambda)(B-\lambda)-(C^2+D^2)]\\ &= (A-\lambda)[\lambda^2-(A+B)\lambda+AB-(C^2+D^2)],\end{aligned} \tag{100}$$

so that the eigenvalues are $\lambda_1 = A$ and

$$\begin{aligned}\lambda_{2,3} &= \frac{A+B}{2} \pm \sqrt{\frac{(A+B)^2}{4}-\frac{4AB}{4}+C^2+D^2}\\ &= \frac{A+B}{2} \pm \sqrt{\frac{(A-B)^2}{4}+C^2+D^2}.\end{aligned} \tag{101}$$

Plugging in the original expressions, we obtain the eigenvalues

$$\lambda_1 = \frac{1}{n_1^2}+r_{13}E_3, \tag{102}$$

$$\lambda_2 = \frac{1}{2}\left(\frac{1}{n_1^2}+\frac{1}{n_3^2}+(r_{13}+r_{33})E_3\right) + \sqrt{\frac{1}{4}\left(\frac{1}{n_1^2}-\frac{1}{n_3^2}+(r_{13}-r_{33})E_3\right)^2+(r_{51}E_1)^2+(r_{42}E_2)^2}, \tag{103}$$

$$\lambda_3 = \frac{1}{2}\left(\frac{1}{n_1^2}+\frac{1}{n_3^2}+(r_{13}+r_{33})E_3\right) - \sqrt{\frac{1}{4}\left(\frac{1}{n_1^2}-\frac{1}{n_3^2}+(r_{13}-r_{33})E_3\right)^2+(r_{51}E_1)^2+(r_{42}E_2)^2}. \tag{104}$$

To determine the changed indices of refraction N_i we solve the equation $1/N_i^2 = \lambda_i$ for N_i. We obtain for the first eigenvalue

$$\frac{1}{N_1^2} = \frac{1+n_1^2r_{13}E_3}{n_1^2} \tag{105}$$

or, equivalently,

$$N_1 = \frac{n_1}{\sqrt{1+n_1^2r_{13}E_3}}. \tag{106}$$

For $\Delta \ll 1$ we can use the approximation

$$N_1 = \frac{n_1}{\sqrt{1+\Delta}} \approx n_1(1-\Delta/2) = n_1 - \frac{1}{2}n_1^3r_{13}E_3. \tag{107}$$

To find the directions of the semi-axes, we compute the eigenvectors using again the abbreviated matrix entries. For an eigenvalue λ we have to solve the linear system

x	y	z	RHS
$A-\lambda$	0	C	0
0	$A-\lambda$	D	0
C	D	$B-\lambda$	0

with $A \neq B$. (108)

For the eigenvalue $\lambda = A$ we have to consider several cases. Case $C \neq 0$: one elimination step leads to

x	y	z	RHS
0	0	C	0
0	0	0	0
C	D	$B-A$	0,

(109)

and the eigenvectors are the scalar multiples of $(-D,\ C,\ 0)^T$.

Case $C = 0$ and $D \neq 0$: the system is already in reduced form

$$\begin{array}{cccc|c} x & y & z & & \text{RHS} \\ \hline 0 & 0 & 0 & & 0 \\ 0 & 0 & D & & 0 \\ 0 & D & B-A & & 0, \end{array} \tag{110}$$

and the eigenvectors are the scalar multiples of $(1, 0, 0)^T$.

Case $C = 0$ and $D = 0$: again the system is already in reduced form

$$\begin{array}{cccc|c} x & y & z & & \text{RHS} \\ \hline 0 & 0 & 0 & & 0 \\ 0 & 0 & 0 & & 0 \\ 0 & 0 & B-A & & 0. \end{array} \tag{111}$$

The eigenvalue $\lambda = A$ has multiplicity two and the eigenvectors are linear combinations of $(1, 0, 0)^T$ and $(0, 1, 0)^T$.

For an eigenvalue $\lambda \neq A$ there are no cases to distinguish. We reduce Eq. (108) to

$$\begin{array}{cccc|c} x & y & z & & \text{RHS} \\ \hline A-\lambda & 0 & C & & 0 \\ 0 & A-\lambda & D & & 0 \\ 0 & 0 & \underbrace{(B-\lambda) - \dfrac{C^2}{A-\lambda} - \dfrac{D^2}{A-\lambda}}_{=0} & & 0, \end{array} \tag{112}$$

where the (3, 3) entry is zero since this is the eigenvalue condition (zero of characteristic polynomial). The eigenvectors are scalar multiples of $(-C, -D, A-\lambda)^T$.

As an example, for the case that the electric field is in z-direction, $E_1 = E_2 = 0$, we have $C = D = 0$ and

$$\lambda_1 = \frac{1}{n_1^2} + r_{13}E_3, \tag{113}$$

$$\begin{aligned} \lambda_2 &= \frac{1}{2}\left(\frac{1}{n_1^2} + \frac{1}{n_3^2} + (r_{13}+r_{33})E_3\right) \\ &\quad + \sqrt{\frac{1}{4}\left(\frac{1}{n_1^2} - \frac{1}{n_3^2} + (r_{13}-r_{33})E_3\right)^2} \\ &= \frac{1}{n_1^2} + r_{13}E_3, \end{aligned} \tag{114}$$

$$\begin{aligned} \lambda_3 &= \frac{1}{2}\left(\frac{1}{n_1^2} + \frac{1}{n_3^2} + (r_{13}+r_{33})E_3\right) \\ &\quad - \sqrt{\frac{1}{4}\left(\frac{1}{n_1^2} - \frac{1}{n_3^2} + (r_{13}-r_{33})E_3\right)^2} \\ &= \frac{1}{n_3^2} + r_{33}E_3, \end{aligned} \tag{115}$$

and consequently we find

$$N_i = \frac{n_i}{\sqrt{1 + n_i^2 r_{i3} E_3}}. \tag{116}$$

For $\Delta \ll 1$ we can use again the approximation

Table 1. Measured Values of Pockels Matrix Elements for BTO [35]

Matrix Entry	Value
$r_{13} = r_{23}$	10.2 ± 0.6 pm/V
r_{33}	105 ± 10 pm/V
$r_{42} = r_{51}$	1300 ± 100 pm/V

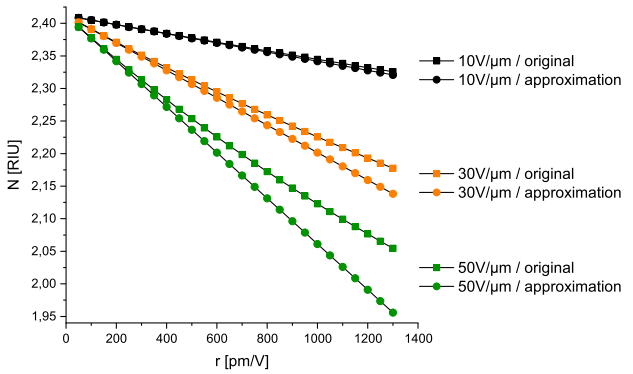

Fig. 7. Calculated refractive index as a function of the Pockels coefficient for different electric field strengths. The unit of the refractive index N is dimensionless but in optics it is common to call it refractive index unit (RIU). The approximated Formula (117) can differ significantly from the original Formula (116) at large field strengths.

$$N_i = \frac{n_i}{\sqrt{1+\Delta}} \approx n_i(1 - \Delta/2) = n_i - \frac{1}{2}n_i^3 r_{i3} E_3. \tag{117}$$

The eigenvectors do not change. The double eigenvalue possesses two eigenvectors that can be arbitrarily chosen to span the xy-plane while the third one is parallel to the z-axis.

Table 1 shows values for the matrix entries measured by [35].

In practice we usually have linearly polarized light, especially in the case of a chip-integrated electro-optical modulator. In this case the electric field vector of the electro-magnetic wave oscillates always in the same direction. For example, if the electro-magnetic wave propagates in the direction of k_x, it is reasonable to polarize the light in z-direction to obtain the largest refractive index change because r_{33} is about 10 times larger than r_{23} (see Table 1).

To optimize the performance of electro-optical modulators, the light polarization, which is the direction of the electric field vector of the electromagnetic wave, as well as the direction of the external electric field need to be considered. In practice, the approximation of the refractive index [see Eq. (117)] is typically used for device optimizations. At this point it is important to note that the approximation can introduce non-negligible error if, for example, the large Pockels coefficient r_{42} is employed. With larger electric field strengths, the error increases. Figure 7 shows calculated values for the refractive index N as a function of the Pockels coefficient and different external electric field strengths as a parameter. In this figure, we compare the calculated values using the approximated equation [Eq. (117)] and the original equation [Eq. (116)]. It is clearly shown that the linearized Formula (117) differs significantly from the nonlinear Formula (116). It is therefore recommended to use the original (nonlinear) formula at high external electric field strengths and large Pockels coefficients.

12. SYMMETRY CONSTRAINTS ON THE QUADRATIC ELECTRO-OPTICAL TENSOR FOR BARIUM TITANATE

Finally we give a brief introduction to the symmetry constraints on the quadratic electro-optical tensor. The symmetries of the non-centrosymmetric barium titanate are: (1) fourfold symmetry about the z-axis, (2) reflection about the yz-plane, (3) reflection about the xz-plane, and (4) reflection about the plane spanned by the z-axis and xy-bisector. The same arguments apply to the tensor of second-order partial derivatives as for the one of first-order. The symmetry (1) is a consequence of the others. Symmetries (2) and (3) lead to $\eta_{ij,kl} = 0$ if an odd number of indices is 1 or an odd number of indices is 2. Finally, (4) stipulates that two tensor components are equal when the indices 1 and 2 in one of them are flipped to 2 and 1, respectively. Moreover, there are the (tensor) symmetry of the impermeability tensor, so $\eta_{ij,kl} = \eta_{ji,kl}$ and the irrelevance of the order of the partial derivatives $\eta_{ij,kl} = \eta_{ij,lk}$.

We obtain (brackets are used because each matrix is only a part of the tensor with $3^4 = 81$ components)

$$\eta_{11,kl} = \begin{bmatrix} u & 0 & 0 \\ 0 & v & 0 \\ 0 & 0 & w \end{bmatrix} \quad \eta_{12,kl} = \begin{bmatrix} 0 & x & 0 \\ x & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad \eta_{13,kl} = \begin{bmatrix} 0 & 0 & y \\ 0 & 0 & 0 \\ y & 0 & 0 \end{bmatrix},$$

$$\eta_{22,kl} = \begin{bmatrix} v & 0 & 0 \\ 0 & u & 0 \\ 0 & 0 & w \end{bmatrix} \quad \eta_{23,kl} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & y \\ 0 & y & 0 \end{bmatrix},$$

$$\eta_{33,kl} = \begin{bmatrix} p & 0 & 0 \\ 0 & p & 0 \\ 0 & 0 & q \end{bmatrix}. \tag{118}$$

This leads to the second-order change

$$\begin{bmatrix} \Delta\eta_{11}^{(2)} \\ \Delta\eta_{22}^{(2)} \\ \Delta\eta_{33}^{(2)} \\ \Delta\eta_{23}^{(2)} \\ \Delta\eta_{31}^{(2)} \\ \Delta\eta_{12}^{(2)} \end{bmatrix} = \frac{1}{2} \begin{bmatrix} u & v & w & 0 & 0 & 0 \\ v & u & w & 0 & 0 & 0 \\ p & p & q & 0 & 0 & 0 \\ 0 & 0 & 0 & 2y & 0 & 0 \\ 0 & 0 & 0 & 0 & 2y & 0 \\ 0 & 0 & 0 & 0 & 0 & 2x \end{bmatrix} \begin{bmatrix} E_1^2 \\ E_2^2 \\ E_3^2 \\ E_2E_3 \\ E_3E_1 \\ E_1E_2 \end{bmatrix}. \tag{119}$$

Hence, the quadratic contribution for the change of $\boldsymbol{\eta}$ is

$$\Delta\boldsymbol{\eta}^{(2)}(\mathbf{E}) = \frac{1}{2} \cdot \begin{pmatrix} uE_1^2 + vE_2^2 + wE_3^2 & 2xE_1E_2 & 2yE_1E_3 \\ 2xE_2E_1 & vE_1^2 + uE_2^2 + wE_3^2 & 2yE_2E_3 \\ 2yE_3E_1 & 2yE_3E_2 & pE_1^2 + pE_2^2 + qE_3^2 \end{pmatrix}. \tag{120}$$

13. DISCUSSION AND CONCLUSION

The purpose of this tutorial is the presentation of the mathematical foundations of the classical—not quantum-theoretical—approach to electro-optics. The Pockels effect and the Kerr effect in anisotropic materials are accurately described by tensors, but due to symmetries are more efficiently summarized in matrices that do not transform like tensors. So, one important aspect of this tutorial is to explain the tensor transformation properties, the implementation of symmetry properties, and the transition between the tensors and the non-tensor matrices. In our experience, theses mathematical aspects are often left out from or at most treated cursorily in engineering curricula.

Another mathematical aspect is the use of eigenvalue problems. They explain why the polarization is limited to specific directions. The eigenvalues of a tensor are conveniently illustrated by ellipsoids. Then the influence of an external electric field transforms the ellipsoid and we can compute how its semi-axes change not only in direction but also in length. This corresponds to the change of the refraction indices of the material due to the external electric field.

The light that travels through the material can be described by planar waves due to the scale of the applications in mind. The wave vector $\mathbf{k}$ determines an orthogonal plane in three-dimensional space. Its intersection with the index ellipsoid yields an index ellipse whose semi-axes provide the relevant information about the possible polarization directions.

We also point out that a commonly used formula for the change of the refraction index involves linearization. However, with today's materials the electro-optical effects become so strong that we should include more accuracy and consider a quadratic approximation. We believe that experimentalists should make measurements to fit quadratic models to today's materials and find out if the quadratic effects are still negligible compared to the linear effects.

We also believe that a detailed understanding of this classical approach, as it is presented here, is only a first step towards incorporating quantum-theoretical models in the light-matter interactions. The presented classical approach provides meaningful insights into light propagation in anisotropic materials and electro-optical effects, which is important for both classical optics and modern photonics such as integrated photonic circuits.

APPENDIX A: SYMMETRY OF TENSORS WITHOUT OR WITH AN EXTERNAL ELECTRIC FIELD

This appendix provides more mathematical details for the symmetry and positive-definiteness of the polarization tensor χ, the dielectric tensor ϵ, and the impermeability tensor η.

It suffices to show the symmetry and positive-definiteness for the polarization tensor χ. The dielectric tensor inherits these properties due to its definition $\epsilon_{ij} = \delta_{ij} + \chi_{ij}$, which adds the symmetric identity tensor. Since positive definiteness is characterized by all eigenvalues being positive, the all positive eigenvalues of χ are shifted by $+1$ to become the all positive eigenvalues of ϵ. Finally, the inverse of a rank two symmetric, the positive definite tensor is also a symmetric, positive definite tensor: the symmetry can be seen from the determinant cofactor formula for the inverse. The positive-definiteness follows, since the eigenvalues of the inverse are the inverses of the original eigenvalues, i.e., they stay positive.

The argument for the polarization tensor uses the line integral

$$\int \mathbf{E}\cdot \mathrm{d}\mathbf{P} = \int \mathbf{E}\cdot \mathrm{d}(\chi \mathbf{E}), \tag{A1}$$

which expresses the potential energy created in the material due to the polarization ([28], Volume II, Chapter 31 Tensors).

In coordinates and with a parameterization $t \mapsto \mathbf{E}(t)$, for $a \le t \le b$, the line integral becomes

$$\int_a^b \begin{pmatrix} E_1(t) & E_2(t) & E_3(t) \end{pmatrix} \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{pmatrix} \begin{pmatrix} T_1(t) \\ T_2(t) \\ T_3(t) \end{pmatrix} \mathrm{d}t, \tag{A2}$$

where

$$\begin{pmatrix} T_1(t) \\ T_2(t) \\ T_3(t) \end{pmatrix} = \frac{1}{\|\mathbf{E}'(t)\|} \begin{pmatrix} E_1'(t) \\ E_2'(t) \\ E_3'(t) \end{pmatrix} \mathrm{d}t \tag{A3}$$

is the unit tangent vector of the parameterization.

The quoted text from Feynman in Section 2 translates into the parameterizations

$$(i) \quad t \mapsto \begin{pmatrix} t & 0 & 0 \end{pmatrix}^T \quad \text{for} \quad 0 \le t \le \Delta E_x, \tag{A4}$$

$$(ii) \quad t \mapsto \begin{pmatrix} \Delta E_x & t & 0 \end{pmatrix}^T \quad \text{for} \quad 0 \le t \le \Delta E_y, \tag{A5}$$

$$(iii) \quad t \mapsto \begin{pmatrix} \Delta E_x - t & \Delta E_y & 0 \end{pmatrix}^T \quad \text{for} \quad 0 \le t \le \Delta E_x, \tag{A6}$$

$$(iv) \quad t \mapsto \begin{pmatrix} 0 & \Delta E_y - t & 0 \end{pmatrix}^T \quad \text{for} \quad 0 \le t \le \Delta E_y. \tag{A7}$$

The line integrals become

$$(i) \quad \int_0^{\Delta E_x} \begin{pmatrix} t & 0 & 0 \end{pmatrix} \begin{pmatrix} \chi_{11} & \chi_{12} & \chi_{13} \\ \chi_{21} & \chi_{22} & \chi_{23} \\ \chi_{31} & \chi_{32} & \chi_{33} \end{pmatrix} \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \mathrm{d}t = \frac{1}{2}\chi_{11}(\Delta E_x)^2, \tag{A8}$$

and

$$(ii) \quad \chi_{12}\Delta E_x \Delta E_y + \frac{1}{2}\chi_{22}(\Delta E_y)^2, \tag{A9}$$

$$(iii) \quad -\chi_{21}\Delta E_x \Delta E_y - \frac{1}{2}\chi_{11}(\Delta E_x)^2, \tag{A10}$$

$$(iv) \quad -\frac{1}{2}\chi_{22}(\Delta E_y)^2. \tag{A11}$$

The sum of the line integrals over all four paths is

$$(\chi_{12} - \chi_{21})\Delta E_x \Delta E_y, \tag{A12}$$

which has to equal zero, since the closed path leads the material back to its original state. There is neither gain nor loss in potential energy. Since ΔE_x and ΔE_y are arbitrary, $\chi_{12} = \chi_{21}$.

Now, we consider the polarization tensor of a material subjected to an external electric field $\mathbf{E} = (E_x, E_y, E_z)^T$. We integrate along the boundary of the rectangle with vertices $(E_x \pm \Delta x/2,\ E_y \pm \Delta y/2,\ E_z)$. As before the integral has to be zero. We assume that Δx and Δy are so small that the values of the edge midpoints are a good approximation along the whole edge. For notational convenience we omit the dependence of the constant E_z in the following formulas. Then the partial results in Eq. (A8) to Eq. (A11) become

$$\frac{1}{2}\chi_{11}\left(E_x, E_y - \frac{\Delta y}{2}\right)\cdot \Delta x^2, \tag{A13}$$

$$\chi_{12}\left(E_x + \frac{\Delta x}{2}, E_y\right)\cdot \Delta x \Delta y + \frac{1}{2}\chi_{22}\left(E_x + \frac{\Delta x}{2}, E_y\right)\cdot \Delta y^2, \tag{A14}$$

$$-\chi_{21}\left(E_x, E_y + \frac{\Delta y}{2}\right)\cdot \Delta x \Delta y - \frac{1}{2}\chi_{11}\left(E_x, E_y + \frac{\Delta y}{2}\right)\cdot \Delta x^2, \tag{A15}$$

$$-\frac{1}{2}\chi_{22}\left(E_x - \frac{\Delta x}{2}, E_y\right)\cdot \Delta y^2. \tag{A16}$$

We add these up and—for normalization with respect to the rectangle size—divide by $\Delta x \cdot \Delta y$:

$$\begin{aligned} &\frac{\Delta x}{2}\cdot \frac{\chi_{11}\left(E_x, E_y - \frac{\Delta y}{2}\right) - \chi_{11}\left(E_x, E_y + \frac{\Delta y}{2}\right)}{\Delta y} \\ &+ \frac{\Delta y}{2}\cdot \frac{\chi_{22}\left(E_x + \frac{\Delta x}{2}, E_y\right) - \chi_2\left(E_x - \frac{\Delta x}{2}, E_y\right)}{\Delta x} \\ &+ \chi_{12}\left(E_x + \frac{\Delta x}{2}, E_y\right) - \chi_{21}\left(E_x, E_y + \frac{\Delta y}{2}\right). \end{aligned} \tag{A17}$$

With the practically safe assumption that the components of χ are continuously differentiable, we let Δx and Δy approach zero. The big fractions become partial derivatives. As they are still multiplied by a delta, they become zero in the limit and do not contribute to the integral. By continuity, the last two terms form the difference

$$\chi_{12}(E_x, E_y) - \chi_{21}(E_x, E_y), \tag{A18}$$

which has to equal zero because there is no potential energy change along the integral that leads us back to the starting value.

Funding. Europäischer Sozialfonds (86001106).

Disclosures. The authors declare no conflicts of interest.

Data availability. No data were generated or analyzed in the presented research.

REFERENCES

1. L. Alloatti, R. Palmer, S. Diebold, *et al.*, "100 GHz silicon–organic hybrid modulator," Light Sci. Appl. **3**, e173 (2014).
2. J. Leuthold, C. Koos, and W. Freude, "Nonlinear silicon photonics," Nat. Photonics **4**, 535–544 (2010).
3. M. Xu, M. He, H. Zhang, *et al.*, "High-performance coherent optical modulators based on thin-film lithium niobate platform," Nat. Commun. **11**, 1 (2020).
4. S.-A. Li, H. Huang, Z. Pan, *et al.*, "Enabling technology in high-baud-rate coherent optical communication systems," IEEE Access **8**, 111318 (2020).
5. C. Villringer, P. Steglich, S. Pulwer, *et al.*, "Electro-optical properties of doped polymers with high transparency in the visible wavelength range," Opt. Mater. Express **11**, 3801–3811 (2021).

6. P. Steglich, C. Villringer, B. Dietzel, *et al.*, "On-chip dispersion measurement of the quadratic electro-optic effect in nonlinear optical polymers using a photonic integrated circuit technology," IEEE Photonics J. **11**, 4900510 (2019).
7. M. Jin, J. Chen, Y. Sua, *et al.*, "Efficient electro-optical modulation on thin-film lithium niobate," Opt. Lett. **46**, 1884–1887 (2021).
8. P. Steglich, C. Mai, A. Peczek, *et al.*, "Quadratic electro-optical silicon-organic hybrid RF modulator in a photonic integrated circuit technology," in *IEEE International Electron Devices Meeting (IEDM)* (2018), pp. 23.3.1–23.3.4.
9. L. R. Dalton, J. Leuthold, B. H. Robinson, *et al.*, "Perspective: nanophotonic electro-optics enabling THz bandwidths, exceptional modulation and energy efficiencies, and compact device footprints," APL Mater. **11**, 050901 (2023).
10. T. Yu, S. Jiang, J. Fang, *et al.*, "Passive repetition-rate stabilization for a mode-locked fiber laser by electro-optic modulation," Opt. Lett. **47**, 1178–1181 (2022).
11. D. D. Hudson, K. W. Holman, R. J. Jones, *et al.*, "Mode-locked fiber laser frequency-controlled with an intracavity electro-optic modulator," Opt. Lett. **30**, 2948–2950 (2005).
12. V. Morozov, A. Olenin, V. Tunkin, *et al.*, "Picosecond pulsed-periodic high-peak power Nd:YAG laser operationally controlled by KTP-based Pockels cell," Crystals **12**, 368 (2022).
13. C. Villringer, T. S. Gilani, E. Zhang, *et al.*, "Development of tuneable Fabry-Pérot sensors for parallelised photoacoustic signal acquisition," Proc. SPIE **10878**, 28–33 (2019).
14. M. Passard, C. Barthod, M. Fortin, *et al.*, "Design and optimization of a low-frequency electric field sensor using Pockels effect," IEEE Trans. Instrum. Meas. **50**, 1053–1058 (2001).
15. Y. Li, L. Gao, J. Wan, *et al.*, "Optical dc electric field sensing based on the Pockels effect using bismuth germanate crystals," Appl. Opt. **59**, 6237–6244 (2020).
16. Q. Yang, R. Liu, Y. He, *et al.*, "Ac/dc hybrid electric field measurement method based on Pockels effect and electric field modulation," Rev. Sci. Instrum. **91**, 055004 (2020).
17. P. Steglich, C. Mai, C. Villringer, *et al.*, "Silicon-organic hybrid photonics: overview of recent advances, electro-optical effects and CMOS-integration concepts," J. Phys.: Photonics **3**, 022009 (2021).
18. B. H. Robinson, L. E. Johnson, D. L. Elder, *et al.*, "Optimization of plasmonic-organic hybrid electro-optics," J. Lightwave Technol. **36**, 5036–5047 (2018).
19. P. Steglich, C. Mai, C. Villringer, *et al.*, "Direct observation and simultaneous use of linear and quadratic electro-optical effects," J. Phys. D **53**, 125106 (2020).
20. P. Steglich, C. Villringer, B. Dietzel, *et al.*, "Electric field-induced linear electro-optic effect observed in silicon-organic hybrid ring resonator," IEEE Photonics Technol. Lett. **32**, 526–529 (2020).
21. S. Abel, F. Eltes, J. E. Ortmann, *et al.*, "Large Pockels effect in micro- and nanostructured barium titanate integrated on silicon," Nat. Mater. **18**, 42–47 (2019).
22. A. Karvounis, F. Timpu, V. V. Vogler-Neuling, *et al.*, "Barium titanate nanostructures and thin films for photonics," Adv. Opt. Mater. **8**, 2001249 (2020).
23. C. Wang, M. Zhang, X. Chen, *et al.*, "Integrated lithium niobate electro-optic modulators operating at CMOS-compatible voltages," Nature **562**, 101–104 (2018).
24. D. Zhu, L. Shao, M. Yu, *et al.*, "Integrated photonics on thin-film lithium niobate," Adv. Opt. Photonics **13**, 242–352 (2021).
25. K. Alexander, J. P. George, J. Verbist, *et al.*, "Nanophotonic Pockels modulators on a silicon nitride platform," Nat. Commun. **9**, 3444 (2018).
26. D. Ban, G. Liu, H. Yu, *et al.*, "Low driving voltage and low optical loss electro-optic modulators based on lead zirconate titanate thin film on silicon substrate," J. Lightwave Technol. **40**, 2939–2943 (2022).
27. A. Yariv and P. Yeh, *Optical Waves in Crystals* (Wiley New York, 1984), Vol. **5**.
28. R. P. Feynman, R. B. Leighton, and M. Sands, *The Feynman Lectures on Physics* (Addison-Wesley, 1965).
29. H. Arwin, S. Schoeche, J. Hilfiker, *et al.*, "Optical chirality determined from Mueller matrices," Appl. Sci. **11**, 6742 (2021).
30. Y. Petit, S. Joly, P. Segonds, *et al.*, "Recent advances in monoclinic crystal optics," Laser Photonics Rev. **7**, 920–937 (2013).
31. T. Gowers, ed., *The Princeton Companion to Mathematics* (Princeton University, 2008).
32. M. Kohli, D. Chelladurai, A. Messner, *et al.*, "Plasmonic ferroelectric modulator monolithically integrated on sin for 216 GBd data transmission," J. Lightwave Technol. **41**, 3825–3831 (2023).
33. D. Chelladurai, M. Kohli, Y. Horst, *et al.*, "Electro-optic frequency response of thin-film barium titanate (BTO) from 20 to 270 GHz," in *European Conference on Optical Communication (ECOC)* (2022), pp. 1–4.
34. P. Steglich and A. Mai, "Analysis of BTO-on-Si-waveguides for energy-efficient electro-optical modulators," Proc. SPIE **11775**, 58–63 (2021).
35. M. Zgonik, P. Bernasconi, M. Duelli, *et al.*, "Dielectric, elastic, piezoelectric, electro-optic, and elasto-optic tensors of batio 3 crystals," Phys. Rev. B **50**, 5941 (1994).
36. B. Jiang, J. Iocozzia, L. Zhao, *et al.*, "Barium titanate at the nanoscale: controlled synthesis and dielectric and ferroelectric properties," Chem. Soc. Rev. **48**, 1194–1228 (2019).
37. M. Harwood, P. Popper, and D. Rushman, "Curie point of barium titanate," Nature **160**, 58–59 (1947).