Pure quadratic or higher-order optical effects in anisotropic crystals induced by external dc fields and probed by a single low-intensity plane electromagnetic wave

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The determination of a clear theoretical demarcation between a true or a false quadratic or higher-order low-intensity optical effect induced by an externally applied static or quasistatic (dc) vector field in anisotropic crystals is the scope of the present work. A complete set of necessary and sufficient conditions required for the practical possibility of direct detection, measurement, and tabulation of only those pure optical contributions is finally obtained. The dc electro-optic effect stands out as the most representative of all of these low-power dc optical effects. However, although the dc Kerr effect remains the main topic of application of the analytical treatment developed in this work, the current theoretical formalism is extended to include other dc conventional crystal optics effects, such as electrogyration, electroabsorption, and externally induced ray or energy propagation. Even more, the theoretical conditions are further generalized to apply to any pure higher-order crystal optics effect induced by external dc fields. These can be electric, magnetic, force, and even temperature or concentration gradient fields. The current treatment does not extend to multiple-beam high-intensity nonlinear optics effects induced by optical (ac) fields. Compared to previously published expressions, a more general Fresnel equation is also provided here together with the most general Jones vectors describing the eigenpolarizations of the single probing beam of light. All the generalizations and extensions mentioned in this article are valid as long as the field-dependent coefficients of the particular optical effect under consideration satisfy the equation of a positive-definite complex Hermitian form.

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I. INTRODUCTION

When an external vector field interacts with an optical medium the properties of that medium change. If the applied field is not strong enough to damage the material, then the physical parameters describing those changed properties can be expanded in a power series in the strength of the field. For optical media that have a center of symmetry, the odd-order terms in the field are null so the quadratic terms become the dominant ones by default. To determine those dominant quadratic coefficients in centrosymmetric crystals becomes an easy task. However, in noncentrosymmetric media, the first-order terms in the field are generally the dominant ones, with the effects of the quadratic or higher-order ones being partially eclipsed. The tacitly accepted idea in the crystal optics field was that one should not bother trying to detect the quadratic or higher-order terms in crystals lacking inversion symmetry because these terms are generally masked by the first-order ones. This is probably one of the main reasons why there are relatively very few, if any, of these quadratic or higher-order coefficients measured or tabulated. Yet, as was later reported by the current authors, in these acentric anisotropic crystals, there are many experimental configurations for which one can practically bypass the first-order or lower-order effects and get direct information about the second-order or higher-order effects, respectively. The convention throughout this work is that these coefficients are considered pure or true as long as they do not contain any contributions from the first-order or lower-order coefficients, respectively. If those quadratic or higher-order terms happen to contain, in any way, at least one contribution from the first-order or lower-order terms, respectively, then they are considered impure or false. Finding these particular experimental configurations in noncentrosymmetric crystals together with the determination, measurement, and tabulation of their corresponding quadratic or higher-order coefficients constitutes an experimentally demanding topic of research in the old field of conventional crystal optics. To the best knowledge of the current authors, the above research project was initiated in 2004 with the dc quadratic electro-optic effect [1]. As of now, this project proves to still be in its infancy [1–5], especially in regards to its experimental (data) side of research.

The dc electro-optic effect occurs when the application of a dc electric field across a crystal induces direct changes in the refractive indexes of that material. The electrical impermeabilities of that medium can be expressed as a perturbation series in the strengths of the components of that field, with the linear (first-order) terms being called Pockels terms and the quadratic (second-order) ones named Kerr terms. The zero-order terms are just the zero-field impermeabilities. It has been previously reported and tabulated that in almost 90% of the noncentrosymmetric crystals for which the Pockels effect is generally dominant, it is possible to bypass it, in principle, and obtain direct information only about the quadratic or higher-order electro-optic terms [2,3]. In that work, however, no clear demarcation between the configurations involving the true and the false quadratic terms was provided; it was tacitly and wrongly assumed that the false quadratic terms were insignificant by at least one order of magnitude relative to the true ones. The first interesting attempt at deriving an analytical way of categorizing the configurations for which

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quadratic electro-optic contributions could be isolated from the Pockels ones was published shortly afterwards for the case of one beam of low-intensity light propagating in an arbitrary direction through a uniaxial crystal [4]. Those results, however, proved to be only necessary, but not sufficient, for a complete distinction between the pure and the false quadratic influences. They were not sufficient in the sense that they did not allow for the complete elimination of all the influences from the Pockels coefficients into the expressions for the quadratic terms. From the current authors' perspective, the most important aspect of that work [4] was asserting the idea that in certain media and optical configurations, the false quadratic terms, made of products of two Pockels terms, could have the same order of magnitude as the Kerr ones. That idea immediately raised the important issue of figuring out the optical configurations where only the true second-order terms were made manifest in noncentrosymmetric crystals. The next step involved determining all those crystal configurations where only combinations of pure quadratic terms could be directly detected in materials where the Pockels effect is usually the dominant effect [5]. In that work, however, all those specific configurations were obtained by methodically considering all the crystal classes and all configurations, and selecting only the ones solely involving combinations of Kerr terms; no consistent analytical method of selection was provided. Aside from determining and tabulating all those special configurations, another positive aspect of that work was devising a technique for a three-in-one experimental setup to be used in detecting those specific experimental configurations [5].

This present article provides a continuation and completion of the above-mentioned work by the current authors [5]. It also contributes up to five important ideas to the previous efforts. First of all, it provides the complete set of necessary and sufficient conditions for direct detection of dc field-induced true quadratic optical effects in the case of arbitrary propagation of a single low-power probing beam of light in crystals lacking inversion symmetry. Secondly, it extends those conditions for the general case involving direct detection of pure terms of any order of magnitude higher or equal to 2; in that way those general expressions could be put to use in the cases of both noncentrosymmetric and centrosymmetric media. The general set of necessary and sufficient conditions derived in this work depends on the optical experimental configuration and the point group symmetry of the crystal under investigation. Thirdly, the conditions obtained here pertain to all anisotropic crystals including the ones belonging to the category with the lowest optical symmetry, the biaxial class; it is then shown how the restrictions of those conditions could then be relaxed further so as to apply to the other classes having higher optical symmetry—the uniaxial and anaxial. Fourthly, the real and symmetric impermeability tensor that is traditionally used in electro-optics will be extended to its more general form as a second-rank complex Hermitian tensor. In this way, the electrically induced linear birefringence (the electro-optic effect) [1–5,8–24] may be treated together with the electrically induced circular birefringence (the electrogyration effect) [6–24], if necessary. By analogy with these two nondissipative effects, the theory is further expanded to allow for the inclusion of the two corresponding dissipative effects of electrically induced linear absorption and circular dichroism [14–17,20,23,24]. As compared to a previous work by a different group of authors [4], a more general Fresnel equation is obtained.

Throughout this work it will be assumed that the probing electromagnetic wave is a single, plane, monochromatic, and low-intensity wave. In other words, no multiple-beam, high-power nonlinear optics effects induced by optical (ac) fields will be covered here. All the theoretical treatment will be phenomenological in nature; it will start at a more abstract level and it will be gradually reduced to the particular case of the quadratic electro-optic effect—the main topic of application of this work. Other analogous low-power optical effects induced by static or quasistatic (dc) fields, such as electrogyration, electroabsorption, and externally induced ray (energy) propagation, will be mentioned afterwards. They will not be treated in too much detail. It will simply be shown how these similar dc optical effects can be described as special cases of the general method provided in this article. Everything from vector fields to physical terms and coefficients will be represented in a fixed laboratory-based Cartesian system of coordinates of unit vectors $\hat{x}_1$, $\hat{x}_2$, and $\hat{x}_3$.

II. WORK

In this section, the formalism of the general treatment will be provided together with the derivation of a generalized Fresnel equation and the set of necessary and sufficient conditions for direct detection of only true optical effects of a certain order without any involvement of the false or lower-order ones in anisotropic media.

A. Formalism of general treatment

When an external vector field $\vec{F} = F_1\hat{x}_1 + F_2\hat{x}_2 + F_3\hat{x}_3$ is applied to an optical medium, certain properties of that physical system change. If the perturbing field is not very strong, so as to irreversibly damage the medium, any physical parameter, generically labeled $\tilde{N}_{pq}(\vec{F})$, associated with a certain changed property of the optical system in question, can be expressed mathematically as a perturbation series in the field given by

$$\tilde{N}_{pq}(\vec{F}) = \tilde{N}^{(0)}_{pq}(\vec{F}) + \tilde{N}^{(1)}_{pq}(\vec{F}) + \tilde{N}^{(2)}_{pq}(\vec{F}) + \cdots . \quad (1)$$

$\tilde{N}^{(0)}_{pq}(\vec{F}) = \tilde{N}_{pq}(\vec{F}=0) = \tilde{c}^{(0)}_{pq}$, $\tilde{N}^{(1)}_{pq}(\vec{F}) = \sum_{j=1}^{3} \tilde{c}^{(1)}_{pqj} F_j$, and $\tilde{N}^{(2)}_{pq}(\vec{F}) = \sum_{j,k=1}^{3} \tilde{c}^{(2)}_{pqjk} F_j F_k$ are the zero-order, first-order (linear), and second-order (quadratic) power terms in the components of the field, respectively. The $\tilde{c}^{(0)}_{pq}$, $\tilde{c}^{(1)}_{pqj}$, and $\tilde{c}^{(2)}_{pqjk}$ are the corresponding coefficients associated with them; the tilde symbol "∼" on top of symbols implies that they can represent complex quantities. Throughout this work $p,q \in \{1,2,3\}$. For the sake of simplicity $\tilde{N}_{pq}(\vec{F})$ will be used as $\tilde{N}_{pq}$ from now on and the field dependence will be assumed in all of the first- or higher-order perturbation terms. The general treatment of this work is valid for all externally induced optical effects for which the $\tilde{N}_{pq}$ terms satisfy the equation of a positively definite complex Hermitian form given by

$$\sum_{p,q=1}^{3} \tilde{N}_{pq} x_p x_q = 1. \quad (2)$$

The geometrical representation of this equation is a closed surface in a three-dimensional space called a complex Hermitian triaxial (scalene) ellipsoid. The terms $\tilde{N}_{pq} = L_{pq} + iC_{pq}$ have $L_{pq} = L_{qp}$ and $C_{pq} = -C_{qp}$ with $i = \sqrt{-1}$. In matrix format

$$[\tilde{N}_{pq}]_{3\times 3} = [L_{pq}]_{3\times 3} + i[C_{pq}]_{3\times 3} = \begin{pmatrix} L_{11} & L_{12} + iC_{12} & L_{13} + iC_{13} \\ L_{12} - iC_{12} & L_{22} & L_{23} + iC_{23} \\ L_{13} - iC_{13} & L_{23} - iC_{23} & L_{33} \end{pmatrix}, \quad (3)$$

with $[L_{pq}]_{3\times 3}$ being a real symmetric matrix and $[C_{pq}]_{3\times 3}$ a real antisymmetric matrix quantifying the field-induced optical effects associated with the linear and circular polarizations of the probing electromagnetic wave, respectively. The matrix provided above in Eq. (3) may be used to model theoretically the optical effects associated with elliptical polarizations.

B. Generalized Fresnel equation

The generalized Fresnel equation will be obtained using the method of (undetermined) Lagrange multipliers, where the Lagrange function $\Lambda(x_1,x_2,x_3;l_1,l_2,l_3)$, containing the Lagrange multipliers l_1, l_2, and l_3, is here defined as

$$\Lambda(x_1,x_2,x_3;l_1,l_2,l_3) = \sum_{p,q=1}^{3} \delta_{pq} x_p x_q + l_1 \left(\sum_{p,q=1}^{3} \tilde{N}_{pq} x_p x_q - 1 \right) + l_2 \sum_{p,q=1}^{3} \delta_{pq} s_p x_q + l_3 \left(\sum_{p,q=1}^{3} \delta_{pq} s_p s_q - 1 \right). \quad (4)$$

The following system of equations is obtained:

$$\frac{\partial \Lambda}{\partial x_1} = 2x_1 + 2l_1 \left(\sum_{q=1}^{3} \tilde{N}_{1q} x_q \right) + l_2 s_1 = 0$$

$$\frac{\partial \Lambda}{\partial x_2} = 2x_2 + 2l_1 \left(\sum_{q=1}^{3} \tilde{N}_{2q} x_q \right) + l_2 s_2 = 0$$

$$\frac{\partial \Lambda}{\partial x_3} = 2x_3 + 2l_1 \left(\sum_{q=1}^{3} \tilde{N}_{3q} x_q \right) + l_2 s_3 = 0$$

$$\frac{\partial \Lambda}{\partial l_1} = \sum_{p,q=1}^{3} \tilde{N}_{pq} x_p x_q - 1 = 0$$

$$\frac{\partial \Lambda}{\partial l_2} = \sum_{p,q=1}^{3} \delta_{pq} s_p x_q = 0$$

$$\frac{\partial \Lambda}{\partial l_3} = \sum_{p,q=1}^{3} \delta_{pq} s_p s_q - 1 = 0. \quad (5)$$

The first constraint limits the range of values taken from the center of the complex Hermitian triaxial ellipsoid anywhere to its surface. The second constraint restricts even further those extrema values by imposing the condition that they must also belong only to the complex Hermitian biaxial ellipse in the plane that cuts the ellipsoid through its center and is perpendicular to the direction of the wave normal or phase propagation direction $\hat{s} = \sum_{p,q=1}^{3} \delta_{pq} s_p \hat{x}_q$. The third constrain guarantees that the $\hat{s}$ vector is actually a unit vector. δ_{pq} is the Kronecker delta symbol.

The expressions for the first two Lagrange multipliers are found to be $l_1 = \sum_{p,q=1}^{3} \delta_{pq} x_p x_q = -\frac{1}{N}$ and $l_2 = \frac{2}{N} \sum_{p,q=1}^{3} \tilde{N}_{pq} s_p x_q$, with $N = \sum_{p,q=1}^{3} \delta_{pq} x_p x_q$ a real parameter. The third multiplier l_3 does not need to be determined. Substituting the two multipliers in Eq. (5), the following homogeneous matrix equation is obtained:

$$\begin{pmatrix} \tilde{\mathcal{N}}_{11} - N & \tilde{\mathcal{N}}_{12} & \tilde{\mathcal{N}}_{13} \\ \tilde{\mathcal{N}}_{21} & \tilde{\mathcal{N}}_{22} - N & \tilde{\mathcal{N}}_{23} \\ \tilde{\mathcal{N}}_{31} & \tilde{\mathcal{N}}_{32} & \tilde{\mathcal{N}}_{33} - N \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} = 0. \quad (6)$$

The matrix $[\tilde{\mathcal{N}}_{pq}]_{3\times 3}$ is given by

$$[\tilde{\mathcal{N}}_{pq}]_{3\times 3} = \{\boldsymbol{I}_{3\times 3} - [s_p s_q]_{3\times 3}\}[\tilde{N}_{pq}]_{3\times 3}, \quad (7)$$

where $[\tilde{N}_{pq}]_{3\times 3}$ is provided in Eq. (3), $\boldsymbol{I}_{3\times 3}$ is the unity matrix and

$$[s_p s_q]_{3\times 3} = \begin{pmatrix} s_1^2 & s_1 s_2 & s_1 s_3 \\ s_2 s_1 & s_2^2 & s_2 s_3 \\ s_3 s_1 & s_3 s_2 & s_3^2 \end{pmatrix} \quad (8)$$

is a real, symmetric matrix. Since $\mathrm{Det}\{\boldsymbol{I}_{3\times 3} - [s_p s_q]_{3\times 3}\} = 0$, the characteristic equation above [Eq. (6)] reduces to a second degree polynomial equation in $N = N_{L,C,\hat{s}} = N(L_{pq},C_{pq},s_p s_q)$ given by

$$N_{L,C,\hat{s}}^2 - U_{L,\hat{s}} N_{L,C,\hat{s}} + V_{L,\hat{s}} - W_{C,\hat{s}} = 0. \quad (9)$$

The three coefficients $U_{L,\hat{s}}$, $V_{L,\hat{s}}$, and $W_{C,\hat{s}}$ are all real and given by

$$U_{L,\hat{s}} = U(L_{pq},s_p s_q) = (L_{22} + L_{33}) s_1^2 + (L_{11} + L_{33}) s_2^2 + (L_{11} + L_{22}) s_3^2 - 2L_{12} s_1 s_2 - 2L_{13} s_1 s_3 - 2L_{23} s_2 s_3, \quad (10)$$

$$V_{L,\hat{s}} = V(L_{pq},s_p s_q) = (L_{22}L_{33} - L_{23}^2) s_1^2 + (L_{11}L_{33} - L_{13}^2) s_2^2 + (L_{11}L_{22} - L_{12}^2) s_3^2 + 2(L_{13}L_{23} - L_{12}L_{33}) s_1 s_2 + 2(L_{12}L_{23} - L_{13}L_{22}) s_1 s_3 + 2(L_{12}L_{13} - L_{23}L_{11}) s_2 s_3, \quad (11)$$

and

$$W_{C,\hat{s}} = W(C_{pq},s_p s_q) = C_{23}^2 s_1^2 + C_{13}^2 s_2^2 + C_{12}^2 s_3^2 - 2C_{13}C_{23} s_1 s_2 + 2C_{12}C_{23} s_1 s_3 - 2C_{12}C_{13} s_2 s_3. \quad (12)$$

This third term, $W_{C,\hat{s}}$, contains only coefficients of the type C_{pq} which are associated with optical effects involving the circular polarization of the probing wave. On the other hand, the first two terms, $U_{L,\hat{s}}$ and $V_{L,\hat{s}}$, contain only coefficients of the type L_{pq} that are associated with effects involving the linear polarization. Similar expressions for the first two terms

[Eq. (10) and (11)] were reported previously [4], but not for the third one [Eq. (12)].

To the authors' best knowledge, Eq. (9) gives the most general expression of a Fresnel's wave vectors equation for a plane electromagnetic wave propagating in an arbitrary direction $\hat{s}$ through an anisotropic crystal. A general Fresnel wave vector equation for the electro-optic effect has been previously obtained by a different group of authors [4]. The current authors, however, express their reservation as regarding the full correctness of those authors' equation as published. This issue will be discussed quantitatively in Sec. I.

The eigenvalues of Eq. (9) are real and of the form

$$N^{\pm}_{L,C,\hat{s}} = N^{\pm}(L_{pq},C_{pq},s_p s_q) = \frac{1}{2}U_{L,\hat{s}} \pm \sqrt{\frac{1}{4}U^2_{L,\hat{s}} - V_{L,\hat{s}} + W_{C,\hat{s}}}. \tag{13}$$

Throughout this work, the "±" superscript refers only to the sign before the square root. In no way does the ± symbol imply that one (eigen) value is positive and the other negative.

For the same probing electromagnetic wave, the eigenpolarizations associated with those eigenvalues are mutually perpendicular and elliptical in nature. These eigenpolarization ellipses are located in a plane normal to $\hat{s}$ and, when expressed in a Jones-vector format, have the most general form,

$$J^{\pm}_{L,C,\hat{s}} = J^{\pm}(L_{pq},C_{pq},s_p s_q) = \begin{pmatrix} \sqrt{\frac{1}{4}U^2_{L,\hat{s}} - V_{L,\hat{s}}} \pm \sqrt{\frac{1}{4}U^2_{L,\hat{s}} - V_{L,\hat{s}} + W_{C,\hat{s}}} \\ -i\sqrt{W_{C,\hat{s}}} \end{pmatrix}, \tag{14}$$

with corresponding generalized ellipticities,

$$e^{\pm}_{L,C,\hat{s}} = e^{\pm}(L_{pq},C_{pq},s_p s_q) = \frac{-\sqrt{W_{C,\hat{s}}}}{\sqrt{\frac{1}{4}U^2_{L,\hat{s}} - V_{C,\hat{s}}} \pm \sqrt{\frac{1}{4}U^2_{L,\hat{s}} - V_{L,\hat{s}} + W_{C,\hat{s}}}}. \tag{15}$$

C. Necessary and sufficient conditions

Since, according to Eq. (1), the $\tilde{N}_{pq}$'s may all be written as a perturbation series in the strengths of the components of the vector field, then the three coefficients above may also be expressed as the perturbation series $U_{L,\hat{s}} = \sum_\zeta U^{(\zeta)}_{L,\hat{s}}$, $V_{L,\hat{s}} = \sum_\zeta V^{(\zeta)}_{L,\hat{s}}$, and $W_{C,\hat{s}} = \sum_\zeta W^{(\zeta)}_{C,\hat{s}}$, where

$$U^{(\zeta)}_{L,\hat{s}} = \left[L^{(\zeta)}_{22} + L^{(\zeta)}_{33}\right]s_1^2 + \left[L^{(\zeta)}_{11} + L^{(\zeta)}_{33}\right]s_2^2 + \left[L^{(\zeta)}_{11} + L^{(\zeta)}_{22}\right]s_3^2 - 2L^{(\zeta)}_{12}s_1s_2 - 2L^{(\zeta)}_{13}s_1s_3 - 2L^{(\zeta)}_{23}s_2s_3, \tag{16}$$

$$V^{(\zeta)}_{L,\hat{s}} = \sum_{\varsigma=0}^{\zeta} \left\{ \begin{array}{l} \left[L^{(\varsigma)}_{22}L^{(\zeta-\varsigma)}_{33} - L^{(\varsigma)}_{23}L^{(\zeta-\varsigma)}_{23}\right]s_1^2 + \left[L^{(\varsigma)}_{11}L^{(\zeta-\varsigma)}_{33} - L^{(\varsigma)}_{13}L^{(\zeta-\varsigma)}_{13}\right]s_2^2 \\ +\left[L^{(\varsigma)}_{11}L^{(\zeta-\varsigma)}_{22} - L^{(\varsigma)}_{12}L^{(\zeta-\varsigma)}_{12}\right]s_3^2 + 2\left[L^{(\varsigma)}_{13}L^{(\zeta-\varsigma)}_{23} - L^{(\varsigma)}_{12}L^{(\zeta-\varsigma)}_{33}\right]s_1s_2 \\ +2\left[L^{(\varsigma)}_{12}L^{(\zeta-\varsigma)}_{23} - L^{(\varsigma)}_{13}L^{(\zeta-\varsigma)}_{22}\right]s_1s_3 + 2\left[L^{(\varsigma)}_{12}L^{(\zeta-\varsigma)}_{13} - L^{(\varsigma)}_{23}L^{(\zeta-\varsigma)}_{11}\right]s_2s_3 \end{array} \right\}, \tag{17}$$

and

$$W^{(\zeta)}_{C,\hat{s}} = \sum_{\varsigma=0}^{\zeta} \left[\begin{array}{l} C^{(\varsigma)}_{23}C^{(\zeta-\varsigma)}_{23}s_1^2 + C^{(\varsigma)}_{13}C^{(\zeta-\varsigma)}_{13}s_2^2 + C^{(\varsigma)}_{12}C^{(\zeta-\varsigma)}_{12}s_3^2 \\ -2C^{(\varsigma)}_{13}C^{(\zeta-\varsigma)}_{23}s_1s_2 + 2C^{(\varsigma)}_{12}C^{(\zeta-\varsigma)}_{23}s_1s_3 - 2C^{(\varsigma)}_{12}C^{(\zeta-\varsigma)}_{13}s_2s_3 \end{array} \right], \tag{18}$$

with the extra clarification that $L^{(\varsigma)}_{pq}L^{(\zeta-\varsigma)}_{pq}$ and $C^{(\varsigma)}_{pq}C^{(\zeta-\varsigma)}_{pq}$ are of the same order of magnitude as the terms $L^{(\zeta)}_{pq}$ and $C^{(\zeta)}_{pq}$, respectively, but are not the same as them.

The general system of conditions necessary and sufficient to have the two (eigen) solutions $N^{\pm}_{L,C,\hat{s}}$ above dependent on the $L^{(\xi)}_{pq}$ and $C^{(\xi)}_{pq}$ terms and not at all dependent on $L^{(\zeta)}_{pq}$, $L^{(\varsigma)}_{pq}L^{(\zeta-\varsigma)}_{pq}$, $C^{(\zeta)}_{pq}$, or $C^{(\varsigma)}_{pq}C^{(\zeta-\varsigma)}_{pq}$ terms of lower order ζ ($0 \leqslant \varsigma < \zeta < \xi$) is

$$\begin{gathered} \sum_{\zeta=1}^{\xi-1} U^{(\zeta)}_{L,\hat{s}} = 0 \\ \sum_{\zeta=1}^{\xi-1} \left[V^{(\zeta)}_{L,\hat{s}} - W^{(\zeta)}_{C,\hat{s}}\right] = 0 \\ \sum_{\zeta=1}^{\xi-1} \left\{ \begin{array}{l} \left[L^{(\zeta)}_{22}L^{(\xi-\zeta)}_{33} - L^{(\zeta)}_{23}L^{(\xi-\zeta)}_{23} - C^{(\zeta)}_{23}C^{(\xi-\zeta)}_{23}\right]s_1^2 + \left[L^{(\zeta)}_{11}L^{(\xi-\zeta)}_{33} - L^{(\zeta)}_{13}L^{(\xi-\zeta)}_{13} - C^{(\zeta)}_{13}C^{(\xi-\zeta)}_{13}\right]s_2^2 \\ +\left[L^{(\zeta)}_{11}L^{(\xi-\zeta)}_{22} - L^{(\zeta)}_{12}L^{(\xi-\zeta)}_{12} - C^{(\zeta)}_{12}C^{(\xi-\zeta)}_{12}\right]s_3^2 + 2\left[\mathrm{L}^{(\zeta)}_{13}\mathrm{L}^{(\xi-\zeta)}_{23} - \mathrm{L}^{(\zeta)}_{12}\mathrm{L}^{(\xi-\zeta)}_{33} + \mathrm{C}^{(\zeta)}_{13}\mathrm{C}^{(\xi-\zeta)}_{23}\right]s_1s_2 \\ +2\left[L^{(\zeta)}_{12}L^{(\xi-\zeta)}_{23} - L^{(\zeta)}_{13}L^{(\xi-\zeta)}_{22} - C^{(\zeta)}_{12}C^{(\xi-\zeta)}_{23}\right]s_1s_3 + 2\left[L^{(\zeta)}_{12}L^{(\xi-\zeta)}_{13} - L^{(\zeta)}_{23}L^{(\xi-\zeta)}_{11} + C^{(\zeta)}_{12}C^{(\xi-\zeta)}_{13}\right]s_2s_3 \end{array} \right\} = 0. \end{gathered} \tag{19}$$

III. dc KERR ELECTRO-OPTIC EFFECT

When the physical system happens to be a nonabsorbing, nongyrotropic, and nonmagnetic optical crystal with the externally applied dc field on it being only electric in nature ($\vec{F} \to \vec{E}$), then the electro-optic effect (electrically induced linear birefringence) manifests itself. The physical parameters quantifying this phenomenon become the real symmetric impermeabilities of the crystal. In other words, $\hat{s} \to \hat{s}$, $L_{pq} \to \eta_{pq} = n_{pq}^{-2}$, $C_{pq} \to 0$, $L \to \eta$, $C \to \eta' = 0$, $N_{L,C,\hat{s}} \to \eta_{\eta,0,\hat{s}} = n_{\eta,0,\hat{s}}^{-2}$, and $N_{L,C,\hat{s}}^{\pm} \to \eta_{\eta,0,\hat{s}}^{\pm} = (n_{\eta,0,\hat{s}}^{\pm})^{-2}$. The symmetric real terms $\eta_{pq} = \eta_{qp}$ may be written as a perturbation series in the components of the electric field: $\eta_{pq} = \eta_{pq}^{(0)} + \eta_{pq}^{(1)} + \eta_{pq}^{(2)} + \ldots$, with $\eta_{pq}^{(0)} = 0$ for $p \neq q$ and $\eta_{pq}^{(0)} > 0$ for $p = q$, $\eta_{pq}^{(1)} = \sum_{j=1}^{3} r_{pqj}^{(1)} E_j$, and $\eta_{pq}^{(2)} = \sum_{j,k=1}^{3} r_{pqjk}^{(2)} E_j E_k$. The real quantities $r_{pqj}^{(1)}$ and $r_{pqjk}^{(2)}$ are the Pockels and Kerr coefficients, respectively. In this work they are considered effective electro-optic coefficients—in the sense that the coupled effects of inverse piezoelectricity, electrostriction, or similarly indirect effects like those will be assumed as already absorbed theoretically in their values.

The Fresnel's wave vectors equation, as a function of the index of refraction $n_{\eta,0,\hat{s}} = \eta_{\eta,0,\hat{s}}^{-0.5}$, becomes

$$\begin{aligned}
&\left[\left(\eta_{22}\eta_{33} - \eta_{23}^2\right)n_{\eta,0,\hat{s}}^4 - (\eta_{22} + \eta_{33})n_{\eta,0,\hat{s}}^2 + 1\right]s_1^2 + \left[\left(\eta_{11}\eta_{33} - \eta_{13}^2\right)n_{\eta,0,\hat{s}}^4 - (\eta_{11} + \eta_{33})n_{\eta,0,\hat{s}}^2 + 1\right]s_2^2 \\
&\quad + \left[\left(\eta_{11}\eta_{22} - \eta_{12}^2\right)n_{\eta,0,\hat{s}}^4 - (\eta_{11} + \eta_{22})n_{\eta,0,\hat{s}}^2 + 1\right]s_3^2 + 2\left[(\eta_{12}\eta_{13} - \eta_{11}\eta_{23})n_{\eta,0,\hat{s}}^4 + \eta_{23}n_{\eta,0,\hat{s}}^2\right]s_2 s_3 \\
&\quad + 2\left[(\eta_{12}\eta_{23} - \eta_{22}\eta_{13})n_{\eta,0,\hat{s}}^4 + \eta_{13}n_{\eta,0,\hat{s}}^2\right]s_1 s_3 + 2\left[(\eta_{13}\eta_{23} - \eta_{33}\eta_{12})n_{\eta,0,\hat{s}}^4 + \eta_{12}n_{\eta,0,\hat{s}}^2\right]s_1 s_2 = 0. \qquad (20)
\end{aligned}$$

This expression is slightly different from a previously published one by a different group of authors [4]. Here the $n_{\eta,0,\hat{s}}$'s which multiply the terms $(\eta_{11}\eta_{33} - \eta_{13}^2)$ and $(\eta_{13}\eta_{23} - \eta_{33}\eta_{12})$ above are at the fourth power, not the second. Even if these discrepancies might turn out to be due to typographical errors alone, they still need to be properly addressed. This is especially important for new expressions.

The general system of conditions [Eq. (19)], when applied to the particular case of a quadratic electro-optic effect ($0 \leqslant \varsigma \leqslant 1$, $\zeta = 1$, and $\xi = 2$), reduces to

$$\begin{aligned}
&\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2 + \eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3 + \eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3 = 0 \\
&\left[\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2\right]\eta_{33}^{(0)} + \left[\eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3\right]\eta_{22}^{(0)} + \left[\eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3\right]\eta_{11}^{(0)} = 0 \\
&\left[\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2\right]\eta_{33}^{(1)} + \left[\eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3\right]\eta_{22}^{(1)} + \left[\eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3\right]\eta_{11}^{(1)} \\
&\quad - \left[\eta_{22}^{(1)}\eta_{33}^{(1)} + \eta_{23}^{(1)}\eta_{23}^{(1)}\right]s_1^2 - \left[\eta_{11}^{(1)}\eta_{33}^{(1)} + \eta_{13}^{(1)}\eta_{13}^{(1)}\right]s_2^2 - \left[\eta_{11}^{(1)}\eta_{22}^{(1)} + \eta_{12}^{(1)}\eta_{12}^{(1)}\right]s_3^2 + 2\eta_{13}^{(1)}\eta_{23}^{(1)}s_1s_2 + 2\eta_{12}^{(1)}\eta_{23}^{(1)}s_1s_3 + 2\eta_{12}^{(1)}\eta_{13}^{(1)}s_2s_3 = 0.
\end{aligned} \qquad (21)$$

Not considering electro-optic terms higher than quadratic, the system above provides the necessary and sufficient set of conditions that would allow for the eigenindexes of refraction to depend only on Kerr terms without any Pockels ones in optically anisotropic crystals. The third equation allows for the elimination of all false quadratic terms. In the case of biaxial crystals ($\eta_{11}^{(0)} \neq \eta_{22}^{(0)} \neq \eta_{33}^{(0)} \neq \eta_{11}^{(0)}$) the system above [Eq. (21)] becomes

$$\begin{aligned}
&\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2 = 0 \\
&\eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3 = 0 \\
&\eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3 = 0 \\
&\left[\eta_{22}^{(1)}\eta_{33}^{(1)} + \eta_{23}^{(1)}\eta_{23}^{(1)}\right]s_1^2 + \left[\eta_{11}^{(1)}\eta_{33}^{(1)} + \eta_{13}^{(1)}\eta_{13}^{(1)}\right]s_2^2 + \left[\eta_{11}^{(1)}\eta_{22}^{(1)} + \eta_{12}^{(1)}\eta_{12}^{(1)}\right]s_3^2 - 2\eta_{13}^{(1)}\eta_{23}^{(1)}s_1s_2 - 2\eta_{12}^{(1)}\eta_{23}^{(1)}s_1s_3 - 2\eta_{12}^{(1)}\eta_{13}^{(1)}s_2s_3 = 0. \quad (22)
\end{aligned}$$

For uniaxial materials ($\eta_{11}^{(0)} = \eta_{22}^{(0)} \neq \eta_{33}^{(0)} \neq \eta_{11}^{(0)}$), that same system of equations reduces to

$$\begin{aligned}
&\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2 = 0 \\
&\eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3 + \eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3 = 0 \\
&\left[\eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3\right]\left[\eta_{22}^{(1)} - \eta_{11}^{(1)}\right] + \left[\eta_{22}^{(1)}\eta_{33}^{(1)} + \eta_{23}^{(1)}\eta_{23}^{(1)}\right]s_1^2 + \left[\eta_{11}^{(1)}\eta_{33}^{(1)} + \eta_{13}^{(1)}\eta_{13}^{(1)}\right]s_2^2 \\
&\quad + \left[\eta_{11}^{(1)}\eta_{22}^{(1)} + \eta_{12}^{(1)}\eta_{12}^{(1)}\right]s_3^2 - 2\eta_{13}^{(1)}\eta_{23}^{(1)}s_1s_2 - 2\eta_{12}^{(1)}\eta_{23}^{(1)}s_1s_3 - 2\eta_{12}^{(1)}\eta_{13}^{(1)}s_2s_3 = 0. \qquad (23)
\end{aligned}$$

Finally, for the anaxial media ($\eta_{11}^{(0)} = \eta_{22}^{(0)} = \eta_{33}^{(0)}$), we have

$$\begin{aligned}
&\eta_{11}^{(1)}s_2^2 + \eta_{22}^{(1)}s_1^2 - 2\eta_{12}^{(1)}s_1s_2 + \eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3 + \eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3 = 0 \\
&\left[\eta_{11}^{(1)}s_3^2 + \eta_{33}^{(1)}s_1^2 - 2\eta_{13}^{(1)}s_1s_3\right]\left[\eta_{33}^{(1)} - \eta_{22}^{(1)}\right] + \left[\eta_{22}^{(1)}s_3^2 + \eta_{33}^{(1)}s_2^2 - 2\eta_{23}^{(1)}s_2s_3\right]\left[\eta_{33}^{(1)} - \eta_{11}^{(1)}\right] \\
&\quad + \left[\eta_{22}^{(1)}\eta_{33}^{(1)} + \eta_{23}^{(1)}\eta_{23}^{(1)}\right]s_1^2 + \left[\eta_{11}^{(1)}\eta_{33}^{(1)} + \eta_{13}^{(1)}\eta_{13}^{(1)}\right]s_2^2 + \left[\eta_{11}^{(1)}\eta_{22}^{(1)} + \eta_{12}^{(1)}\eta_{12}^{(1)}\right]s_3^2 \\
&\quad - 2\eta_{13}^{(1)}\eta_{23}^{(1)}s_1s_2 - 2\eta_{12}^{(1)}\eta_{23}^{(1)}s_1s_3 - 2\eta_{12}^{(1)}\eta_{13}^{(1)}s_2s_3 = 0. \qquad (24)
\end{aligned}$$

The last equation in each one of the two systems above [Eqs. (23) and (24)] can be further simplified depending on the relationships between the $\eta_{11}^{(1)}$, $\eta_{22}^{(1)}$, and $\eta_{33}^{(1)}$ terms characteristic to each particular point group class of symmetry. The optical configurations satisfying the above system of conditions [Eq. (21)] have been already determined and tabulated in a previous work [5].

IV. DISCUSSION AND CONCLUSION

A. dc externally induced birefringence

The general treatment developed in this work may be adapted to other optical phenomena for which the property in Eq. (2) remains valid. For example, the treatment of the externally induced linear birefringence may be extended to include the circular birefringence (dc electrogyration effect) by making the following substitutions in the general treatment from Sec. I: $\hat{s} \to \hat{s}$, $\tilde{N}_{pq} \to \tilde{\eta}_{pq}$, $L_{pq} \to \eta_{pq} = n_{pq}^{-2}$, $C_{pq} \to \eta'_{pq} = V_{\eta,\hat{s}} G_{pq}$, $L \to \eta$, $C \to \eta'$, $N_{L,C,\hat{s}} \to \eta_{\eta,\eta',\hat{s}} = n_{\eta,\eta',\hat{s}}^{-2} = (\frac{u_{\eta,\eta',\hat{s}}}{c})^2$, and $N_{L,C,\hat{s}}^{\pm} \to \eta_{\eta,\eta',\hat{s}}^{\pm} = (n_{\eta,\eta',\hat{s}}^{\pm})^{-2} = (\frac{u_{\eta,\eta',\hat{s}}^{\pm}}{c})^2$. The $n_{\eta,\eta',\hat{s}}$ and $n_{\eta,\eta',\hat{s}}^{\pm}$'s are the index and eigenindexes of refraction, respectively. The $u_{\eta,\eta',\hat{s}}$ and $u_{\eta,\eta',\hat{s}}^{\pm}$'s are the propagation (phase) speed and eigenspeeds, respectively; c is the speed of light. G_{pq}'s are the elements of a real, antisymmetric gyration vector $\vec{G} = (G_1 s_1 + G_2 s_2 + G_3 s_3)\hat{s}$ and are related to the components g_{pq} of a second-rank axial gyration tensor through the relationship given by

$$\begin{pmatrix} G_1 \\ G_2 \\ G_3 \end{pmatrix} = \begin{pmatrix} G_{32} \\ -G_{31} \\ G_{21} \end{pmatrix} = \begin{pmatrix} -G_{23} \\ G_{13} \\ -G_{12} \end{pmatrix} = \begin{pmatrix} g_{11} & g_{12} & g_{13} \\ g_{12} & g_{22} & g_{23} \\ g_{13} & g_{23} & g_{33} \end{pmatrix} \begin{pmatrix} s_1 \\ s_2 \\ s_3 \end{pmatrix}. \tag{25}$$

The symmetric real terms g_{pq} can be written as a perturbation series in the components of the electric field: $g_{pq} = g_{pq}^{(0)} + g_{pq}^{(1)} + g_{pq}^{(2)} + \cdots$, with $g_{pq}^{(0)}$ describing the natural activity, $g_{pq}^{(1)} = \sum_{j=1}^{3} \gamma_{pqj}^{(1)} E_j$ and $g_{pq}^{(2)} = \sum_{j,k=1}^{3} \gamma_{pqjk}^{(2)} E_j E_k$. The quantities $\gamma_{pqj}^{(1)}$ and $\gamma_{pqjk}^{(2)}$ are the linear and quadratic electrogyration coefficients, respectively.

The general Fresnel equation and its (eigen) solutions, when both field-induced linear and circular birefringence are considered in Eqs. (9)–(13), are given by

$$n_{\eta,\eta',\hat{s}}^{-4} - U_{\eta,\hat{s}} n_{\eta,\eta',\hat{s}}^{-2} + V_{\eta,\hat{s}} - \left[V_{\eta,\hat{s}}\left(g_{11}s_1^2 + g_{22}s_2^2 + g_{33}s_3^2 + 2g_{12}s_1s_2 + 2g_{13}s_1s_3 + 2g_{23}s_2s_3\right)\right]^2 = 0, \tag{26}$$

and

$$(n_{\eta,\eta',\hat{s}}^{\pm})^{-2} = \frac{1}{2}U_{\eta,\hat{s}} \pm \sqrt{\frac{1}{4}U_{\eta,\hat{s}}^2 - V_{\eta,\hat{s}} + \left[V_{\eta,\hat{s}}\left(g_{11}s_1^2 + g_{22}s_2^2 + g_{33}s_3^2 + 2g_{12}s_1s_2 + 2g_{13}s_1s_3 + 2g_{23}s_2s_3\right)\right]^2}, \tag{27}$$

respectively. The term $V_{\eta,\hat{s}} = V_{\eta,\hat{s}}^{(0)} + V_{\eta,\hat{s}}^{(1)} + V_{\eta,\hat{s}}^{(2)} + \cdots$ is provided by Eq. (11) when replacing the L_{pq}'s with η_{pq}'s. In the more accurate expression above, $\eta'_{pq} = V_{\eta,\hat{s}} G_{pq}$, both $V_{\eta,\hat{s}}$ and G_{pq} are indirectly field dependent through the η_{pq} and g_{pq} terms that they contain, respectively. In practical applications, however, it has become customary to approximate $V_{\eta,\hat{s}}$ by its zeroth-order term $V_{\eta,\hat{s}}^{(0)}$, thus making it field independent. That way, all the measured changes in $\eta'_{pq} \simeq V_{\eta,\hat{s}}^{(0)} G_{pq}$ will be associated only with changes in the g_{pq}'s by default [7,12,13]. Taking into account this approximation, the two expressions above [Eqs. (26) and (27)] reduce to

$$n_{\eta,\eta',\hat{s}}^{-4} - U_{\eta,\hat{s}} n_{\eta,\eta',\hat{s}}^{-2} + V_{\eta,\hat{s}} - \left[V_{\eta,\hat{s}}^{(0)}\left(g_{11}s_1^2 + g_{22}s_2^2 + g_{33}s_3^2 + 2g_{12}s_1s_2 + 2g_{13}s_1s_3 + 2g_{23}s_2s_3\right)\right]^2 \simeq 0, \tag{28}$$

and

$$(n_{\eta,\eta',\hat{s}}^{\pm})^{-2} \simeq \frac{1}{2}U_{\eta,\hat{s}} \pm \sqrt{\frac{1}{4}U_{\eta,\hat{s}}^2 - V_{\eta,\hat{s}} + \left[V_{\eta,\hat{s}}^{(0)}\left(g_{11}s_1^2 + g_{22}s_2^2 + g_{33}s_3^2 + 2g_{12}s_1s_2 + 2g_{13}s_1s_3 + 2g_{23}s_2s_3\right)\right]^2}, \tag{29}$$

respectively.

B. dc externally induced absorption

The dc electroabsorption effect may also be quantified using the same general treatment from Sec. I by making the following substitutions: $\hat{s} \to \hat{\sigma}$, $\tilde{N}_{pq} \to \tilde{\alpha}_{pq}$, $L_{pq} \to \alpha_{pq}$, $C_{pq} \to \alpha'_{pq}$, $L \to \alpha$, $C \to \alpha'$, $N_{L,C,\hat{s}} \to \alpha_{\alpha,\alpha',\hat{\sigma}}$, and $N_{L,C,\hat{s}}^{\pm} \to \alpha_{\alpha,\alpha',\hat{\sigma}}^{\pm}$. The real, symmetric matrix $[\alpha_{pq}]_{3\times3}$ describes the linear absorption while the real antisymmetric one $[\alpha'_{pq}]_{3\times3}$ is associated with the circular dichroism. The term $\alpha_{\alpha,\alpha',\hat{\sigma}}$ represents the absorption coefficient. The two terms $\alpha_{\alpha,\alpha',\hat{\sigma}}^{\pm}$ are the absorption eigenvalues in a constant-amplitude plane that is perpendicular to the unit vector $\hat{\sigma} = \sigma_1\hat{x}_1 + \sigma_2\hat{x}_2 + \sigma_3\hat{x}_3$ denoting the attenuation direction. Boundary conditions show that $\hat{\sigma}$ is also normal to the interface separating the medium of incidence from the medium of refraction which, in this case, constitutes the anisotropic crystal. In the most general case, at oblique incidence, the plane wave inside the material becomes inhomogeneous and $\hat{s}$ and $\hat{\sigma}_{\substack{\text{inhomogeneous}\\ \text{plane wave}}}$ are not parallel ($\hat{\sigma}_{\substack{\text{inhomogeneous}\\ \text{plane wave}}} \neq \hat{s}$) [19]. In most of the interferometric experimental configurations, however, the incoming wave is normally incident on the surface of the crystal and, therefore, may be modeled theoretically by a homogeneous plane wave having its $\hat{\sigma}_{\substack{\text{homogeneous}\\ \text{plane wave}}}$ collinear with $\hat{s}$ ($\hat{\sigma}_{\substack{\text{homogeneous}\\ \text{plane wave}}} = \hat{s}$). The complex Hermitian absorption ellipsoid $\sum_{p,q=1}^{3} \tilde{\alpha}_{pq} x_p x_q = 1$

and the complex Hermitian index ellipsoid $\sum_{p,q=1}^{3}\tilde{\eta}_{pq}x_p x_q = 1$ are always concentric but their principal axes are not necessarily coaxial, especially for the lower symmetry point groups [14,15,24]. The slowness bivector $\tilde{\vec{K}}_{\hat{s},\hat{\sigma}}$ is of the type $\tilde{\vec{K}}_{\hat{s},\hat{\sigma}} = \vec{k}_{\hat{s}} + i\vec{\kappa}_{\hat{\sigma}}$, with $\vec{k}_{\hat{s}}$ and $\vec{\kappa}_{\hat{\sigma}}$ in the directions of $\hat{s}$ and $\hat{\sigma}$, respectively [17,19,20,23–27]. Modeling field-induced linear and circular birefringence together with linear and circular absorption is still an ongoing topic of research in crystal optics. Many interesting approaches to deal with this theoretical challenge have been proposed over the years [23–35].

Taking into account the above substitutions in Eqs. (9)–(13), the general Fresnel equation and its corresponding eigenvalues, for field-induced absorption, are given by

$$\alpha^2_{\alpha,\alpha',\hat{\sigma}} - U_{\alpha,\hat{\sigma}}\alpha_{\alpha,\alpha',\hat{\sigma}} + V_{\alpha,\hat{\sigma}} - W_{\alpha',\hat{\sigma}} = 0, \tag{30}$$

and

$$\alpha^{\pm}_{\alpha,\alpha',\hat{\sigma}} = \frac{1}{2}U_{\alpha,\hat{\sigma}} \pm \sqrt{\frac{1}{4}U^2_{\alpha,\hat{\sigma}} - V_{\alpha,\hat{\sigma}} + W_{\alpha',\hat{\sigma}}}, \tag{31}$$

respectively. These expressions take into account the effects of both linear and circular dichroism.

C. dc externally induced ray or energy propagation

The propagation of electromagnetic energy inside a material may also be described with the general formalism in Sec. I. In this case the following substitutions are in order: $\hat{s} \to \hat{S}$, $\tilde{N}_{pq} \to \tilde{\varepsilon}_{pq}$, $L_{pq} \to \varepsilon_{pq}$, $C_{pq} \to \varepsilon'_{pq}$, $L \to \varepsilon$, $C \to \varepsilon'$, $N_{L,C,\hat{s}} \to \varepsilon_{\varepsilon,\varepsilon',\hat{S}} = (\frac{c}{v_{\varepsilon,\varepsilon',\hat{S}}})^2$, and $N^{\pm}_{L,C,\hat{s}} \to \varepsilon^{\pm}_{\varepsilon,\varepsilon',\hat{S}} = (\frac{c}{v^{\pm}_{\varepsilon,\varepsilon',\hat{S}}})^2$. The $v_{\varepsilon,\varepsilon',\hat{S}}$ and $v^{\pm}_{\varepsilon,\varepsilon',\hat{S}}$'s are the ray (group) speed and eigenspeeds, respectively. The $\hat{S}$ represents the unit vector associated with the Poynting vector. The angle between $\hat{S}$ and $\hat{s}$ is the same as the angle between the electric field vector and the electric displacement vector for the same type of polarization of the probing electromagnetic wave inside the anisotropic material. The complex Hermitian Fresnel (ray) ellipsoid $\sum_{p,q=1}^{3}\tilde{\varepsilon}_{pq}x_p x_q = 1$ is always concentric with both the indicatrix and absorption ellipsoid but its principal axes are collinear only with the principal axes of the index ellipsoid for a monochromatic probing wave [21]. If temporal dispersion is present, then the Fresnel ellipsoid is only concentric with the indicatrix.

When the substitutions above are inserted into Eqs. (9)–(13), one obtains a general Fresnel equation and its corresponding (eigen) solutions associated with the speeds of the ray (energy) propagation in anisotropic crystals, respectively:

$$v^{-4}_{\varepsilon,\varepsilon',\hat{S}} - c^{-2}U_{\varepsilon,\hat{S}}v^{-2}_{\varepsilon,\varepsilon',\hat{S}} + c^{-4}(V_{\varepsilon,\hat{S}} - W_{\varepsilon',\hat{S}}) = 0, \tag{32}$$

and

$$(v^{\pm}_{\varepsilon,\varepsilon',\hat{S}})^{-2} = \frac{\frac{1}{2}U_{\varepsilon,\hat{S}} \pm \sqrt{\frac{1}{4}U^2_{\varepsilon,\hat{S}} - V_{\varepsilon,\hat{S}} + W_{\varepsilon',\hat{S}}}}{c^2}. \tag{33}$$

D. Conclusion

In the present work the authors have put forward a general theoretical treatment of low-power single-beam optical effects in anisotropic crystals induced by externally applied static or quasistatic (dc) external fields. This phenomenological formalism may also be applied to other similar optical effects induced by various kinds of dc vector fields such as magnetic, mechanical, thermal, or concentration gradient fields. The work does not involve any multiple-beam high-intensity nonlinear optics effects induced by optical (ac) fields. The general set of expressions and the system of necessary and sufficient conditions derived for direct detection of true higher-order effects without the interference of any false lower-order ones are used in detail for the particular case of the dc quadratic electro-optic effect which constitutes the main topic of application of this work. These conditions depend on the optical experimental configuration and the point group symmetry of the anisotropic crystal in question. Other similar effects, such as the dc electrogyration, dc linear electroabsorption, dc circular electrodichroism, and dc externally induced energy propagation are mentioned. The current treatment can be adapted to describe (separately) the optical effects of dc field-induced birefringence, absorption, and ray propagation in anisotropic crystals for configurations associated with both the linear and/or circular polarizations of a single yet arbitrarily propagating low-intensity plane electromagnetic wave. Finally, the current authors hope that the theoretical findings of their present work will be very helpful in guiding future experimentalists in their quest for the direct determination, measurement, and tabulation of pure quadratic or higher-order optical coefficients of anisotropic crystals under the effects of externally applied dc vector fields. The experimental values of these pure quadratic or higher-order coefficients are very scarce, if almost nonexistent, especially for noncentrosymmetric optical media.

[1] M. Melnichuk and L. T. Wood, J. Opt. Soc. Am. A **22**, 377 (2004).
[2] M. Melnichuk and L. T. Wood, J. Opt. Soc. Am. A **23**, 1236 (2006).
[3] M. Melnichuk and L. T. Wood, J. Opt. Soc. Am. A **24**, 2843 (2007).
[4] M. Izdebski and W. Kucharczyk, J. Opt. Soc. Am. B **25**, 149 (2008).
[5] M. Melnichuk and L. T. Wood, Phys. Rev. A **82**, 013821 (2010).
[6] O. G. Vlokh and R. O. Vlokh, Opt. Photonics News **20**, 34 (2009).
[7] A. T. Efremidis, N. C. Deliolanis, K. Vyrsokinos, C. Manolikas, S. Ves, and E. D. Vanidhis, J. Appl. Crystallogr. **44**, 1100 (2011).
[8] E. U. Condon, in *Handbook of Physics*, edited by E. U. Condon and H. Odishaw (McGraw-Hill, New York, 1958).
[9] I. P. Kaminow, *An Introduction to Electrooptic Devices* (Academic, Orlando, FL, 1974).
[10] F. Agullo-Lopez, J. M. Cabrera, and F. Agullo-Rueda, *Electrooptics: Phenomena, Materials, and Applications* (Academic, San Diego, 1994).
[11] F. Nye, *Physical Properties of Crystals: Their Representation by Tensors and Matrices* (Oxford University Press, Oxford, 1985).

[12] A. Yariv and P. Yeh, *Optical Waves in Crystals: Propagation and Control of Laser Radiation* (Wiley, Hoboken, NJ, 2003).
[13] S. Huard, *Polarization of Light* (Wiley, Chichester, UK, 1997).
[14] S. Haussühl, *Physical Properties of Crystals: An Introduction* (Wiley-VCH, Weinheim, 2007).
[15] H.-J. Weber, in *Crystals: Growth, Properties, and Applications*, edited by H. C. Freyhardt (Springer, Berlin, 1988).
[16] A. M. Glazer and K. G. Cox, in *International Tables for Crystallography: Physical Properties of Crystals*, edited by A. Authier (Kluwer Academic, Dordrecht, 2003).
[17] M. Born and E. Wolf, *Principles of Optics*, 7th ed. (expanded) (Cambridge University Press, Cambridge, 1999).
[18] B. N. Grechushnikov, in *Modern Crystallography IV: Physical Properties of Crystals*, edited by L. A. Shuvalov (Springer, Berlin, 1988).
[19] V. M. Agranovich and V. L. Ginzburg, *Crystal Optics with Spatial Dispersion and Excitons* (Springer, Berlin, 1984).
[20] L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media* (Elsevier Butterworth-Heinemann, Oxford, 2008).
[21] Y. I. Sirotin and M. P. Shaskolskaya, *Fundamentals of Crystal Physics* (Mir, Moscow, 1982).
[22] A. V. Shubnikov, *Principles of Optical Crystallography* (Springer, New York, 1960).
[23] G. Szivessy, in *Handbuch der Physik*, edited by L. Grebe (Springer, Berlin, 1928), Vol. 20.
[24] G. N. Ramachandran and S. Ramaseshan, in *Encyclopaedia of Physics*, edited by S. Flügge (Springer, Berlin, 1961), Vol. 25(I).
[25] F. I. Fedorov, *Theory of Elastic Waves in Crystals* (Springer, New York, 1968).
[26] G. Caviglia and A. Morro, *Inhomogeneous Waves in Solids and Fluids* (World Scientific, Singapore, 1992).
[27] Ph. Boulanger and M. Hayes, *Bivectors and Waves in Mechanics and Optics* (Chapman & Hall, London, 1993).
[28] M. Hayes, Arch. Ration. Mech. Anal. **97**, 221 (1987).
[29] S. Pancharatnam, Proc. Indian Acad. Sci., Sect. A **42**, 86 (1955).
[30] A. Yariv and J. F. Lotspeich, J. Opt. Soc. Am. **72**, 273 (1982).
[31] B. N. Grechushnikov and A. F. Konstantinova, Comput. Math. Appl. **16**, 637 (1988).
[32] G. Caviglia and A. Morro, Continuum Mech. Thermodyn. **7**, 231 (1995).
[33] H. Greiner, Optik **114**, 109 (2003).
[34] S. Alfonso, C. Alberdi, J. M. Diñeiro, M. Berrogui, B. Hernández, and C. Sáenz, J. Opt. Soc. Am. A **21**, 1776 (2004).
[35] Y. Wang, L. Liang, H. Xin, and L. Wu, J. Opt. Soc. Am. A **25**, 653 (2008).