# Some applications of the anisotropic diffraction in biaxial crystals

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## ABSTRACT

We analyze the propagation of pulses in noncentrosymmetric crystals by applying high-frequency expansions techniques for Maxwell equations. As a first application we give a closed-form expression for the anisotropic diffraction operator. Given this expression we identify a critical configuration in biaxial crystals where the diffraction reduces to a one-dimensional second-order operator for the ordinary wave instead of the standard transverse Laplacian. The beam propagation in such a configuration involves the generation of spatial solitons because of this anomalous one-dimensional diffraction. As a second application we present closed-form formulas for the interference patterns from biaxial crystal plates between two polarizers. These formulas agree with experimental patterns.

**Keywords:** Anisotropic media, diffraction, soliton.

## 1. INTRODUCTION

We consider the propagation of electromagnetic fields in anisotropic media so as to derive evolution equations for the slowly varying envelopes. Such results have already been obtained using more or less ad hoc methods.[1] In particular a modern method of solving optical problems is based on the integral formulation of the field equation and the determination of the Green's function.[2] The method requires an explicit representation of the Green's function which is obtained by the use of a Fourier transform. Then applying stationary phase method one gets the asymptotic form of the Green's function. This method is efficient for linear media, but it is not well-adapted for addressing nonlinear problems since the use of Fourier transform and then the derivation of an explicit form of the solution are then prohibited. We use a technique based on high-frequency expansions of the fields which has already been successfully applied to systems of linear, semilinear and quasilinear hyperbolic partial differential equations.[3] This technique is more robust than the Green's function approach in that the approximate solutions are not derived by an asymptotic analysis of an explicit solution, but through a direct asymptotic analysis of Maxwell equations. As a consequence this approach can handle nonlinearities.

The framework for high-frequency expansions of the solutions of Maxwell equations follows from the appearance of the small parameter $\varepsilon$ which has the order of magnitude of the carrier wavelength of light $\lambda$ divided by the next smallest characteristic length present in the problem. If a phase $\phi = kz - \omega t$ satisfies the characteristic equation which reads as the dispersion relation $k = k(\omega)$, then we look for solutions which can be expanded as a series of slowly varying functions of the type:

$$\mathcal{E} = \frac{1}{2}\varepsilon^{\alpha}\left(\sum_j \varepsilon^j \mathbf{E}_j(\varepsilon t, \varepsilon x, \varepsilon y, \varepsilon z)e^{i(kz-\omega t)} + cc\right),$$

where $cc$ is a shorthand for complex conjugate, while $\varepsilon^{\alpha}$ is the dimensionless parameter that characterizes the amplitude of the incoming wave. In case of linear media or for evanescent sources $\alpha \gg 1$ this amplitude has no importance, but it governs the strength of interaction in case of nonlinear media. The evolution equation for the slowly varying envelope $\mathbf{E}_0$ reads as the compatibility condition for the existence of the expansion. The strategy consists in two steps. We first consider the Maxwell equations in the scales of geometric optics, that is to say for propagation distance of the same order as the radius of the beam $\sim \lambda\varepsilon^{-1}$. In this framework the propagation equations read as transport equations along the rays of geometric optics. Second we revisit the Maxwell equations in the moving frame indicated by the above-derived rays. In the scales of geometric optics, the propagation equations are then trivial, which allows one to consider larger propagation distances, of the order of the Rayleigh distance $\sim \lambda\varepsilon^{-2}$.

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In this framework the propagation equations read as Schrödinger-like equations which give the evolutions of the slowly varying envelope according to the law of diffractive optics. The above results hold true in great generality in linear media, and they can be generalized to nonlinear propagation regimes. We may deal with three main nonlinear effects.

1) The first nonlinear effect we may encounter in anisotropic media is the sum frequency generation. A nonlinear $\chi^2$-type function applied to expressions of the form $\sum_f \mathbf{E}_f^\varepsilon(\varepsilon t, \varepsilon \mathbf{x}) \exp i(k^f z - \omega_f t)$ will produce harmonics, that is to say expression with rapid phase $(k^{f_1} + k^{f_2})z - (\omega_{f_1} + \omega_{f_2})t$. If the couples $(\omega_f, k^f)$ satisfy the dispersion relation, then the natural harmonic phases generally do not, due to the dispersive property of the material. The set of harmonics which satisfy the dispersion relation is generally very small (sometimes empty), because they need to fulfill a very drastic phase matching condition. If this set is not empty, then we must also take care that the strength of interaction and therefore the scale for interaction depends on the amplitude of the wave. If the amplitude of the wave is $\varepsilon^\alpha$, then an effective $p$-wave interaction process will be noticeable for propagation length of order $\lambda\varepsilon^{-(p-1)\alpha}$. As a consequence a second order nonlinear effect will appear for propagation length of the order of $\lambda\varepsilon^{-\alpha}$. Accordingly $\alpha = 1$ will correspond to nonlinear geometric optics and $\alpha = 2$ to nonlinear diffractive optics.

2) In isotropic media or in some particular classes of anisotropic media the second-order $\chi^2$ tensor is vanishing. However the $\chi^3$ tensor is never zero. A nonlinear $\chi^3$-type function applied to expressions of the form $\sum_f \mathbf{E}_f^\varepsilon(\varepsilon t, \varepsilon \mathbf{x}) \exp i(k^f z - \omega_f t)$ will produce phase-matched terms with phase $(k^f - k^f + k^f)z - (\omega_f - \omega_f + \omega_f)t = k^f z - \omega_f t$. Note that the phase matching condition is readily fulfilled for this combination, but this nonlinear term does not correspond to harmonic generation since the result of this sum has exactly the same carrier wavenumber and frequency as the fundamental mode. As pointed out in the above paragraph the scale for interaction depends on the amplitude of the wave. A third order nonlinear effect will appear for propagation length of the order of $\lambda\varepsilon^{-2\alpha}$. As a consequence, in case of vanishing second order nonlinearity, the third order nonlinearity prevails and $\alpha = 1/2$ will correspond to nonlinear geometric optics while $\alpha = 1$ will correspond to nonlinear diffractive optics.

3) It often happens when one considers anisotropic media that the second-order $\chi^2$ tensor is non-vanishing but the set of harmonics which satisfy the dispersion relation is empty. In such configurations cascaded $\chi^2$ terms are of the same order as $\chi^3$ terms, which gives rise to complicated effective nonlinear terms. Once again, the scale for interaction will depend on the amplitude of the input wave. $\alpha = 1/2$ will correspond to nonlinear geometric optics while $\alpha = 1$ will correspond to nonlinear diffractive optics.

Note that the general results stated in Section 4 are derived in Ref. 4 which is a paper devoted to the rigorous high-frequency expansions of solutions of nonlinear Maxwell equations. That is why we skip here the complete proofs, but only give the principle of the approach in Section 3. In this paper, we focus our attention to three applications of these results presented in Sections 5-7.

## 2. FORMULATION AND SCALING

We consider an incident beam onto a non-magnetic nonlinear crystal that occupies the domain $\mathbb{R}^3_+ := \{(x, y, z) \in \mathbb{R}^3, z > 0\}$ (see Fig. 1a). The propagation axis is perpendicular to the boundary surface $\Sigma := \{(x, y, z) \in \mathbb{R}^3, z = 0\}$ and is collinear to the $z$-axis. The evolution of the electric field $\mathcal{E}$ is governed by the Maxwell equation:

$$\textbf{rot rot } \mathcal{E} = -\mu_0 \partial_{tt} \mathcal{D}, \tag{1}$$

where the electric induction divides into the sum $\mathcal{D} = \mathcal{D}_l + \mathcal{P}_{nl}$ of a linear and a nonlinear part:

$$\begin{aligned} \mathcal{D}_l &= \varepsilon_0 \mathcal{E} + \varepsilon_0 \chi^{(1)} * \mathcal{E}, & (2)\\ \mathcal{P}_{nl} &= \varepsilon_0 \chi^{(2)} * (\mathcal{E}, \mathcal{E}) + \varepsilon_0 \chi^{(3)} * (\mathcal{E}, \mathcal{E}, \mathcal{E}) + ..., & (3)\\ \chi^{(j)} * (\mathcal{E}, ..., \mathcal{E}) &= \int_{-\infty}^t dt_1 ... \int_{-\infty}^t dt_j \chi^{(j)}(t - t_1, ..., t - t_j) : \mathcal{E}(t_1)...\mathcal{E}(t_j). & (4) \end{aligned}$$

$\varepsilon_0$ and $\mu_0$ are respectively the dielectric constant and magnetic permeability of vacuum. The electromagnetic wave is assumed to be far enough from all absorption lines of the medium so that we can neglect absorption and the tensors $\chi^{(j)}$ are real.

The boundary condition at the surface $\Sigma$ is imposed by the continuity of the tangential components of the magnetic and electric fields. The source $\mathcal{S}$ corresponding to the electric field of the incoming pulse at the interface

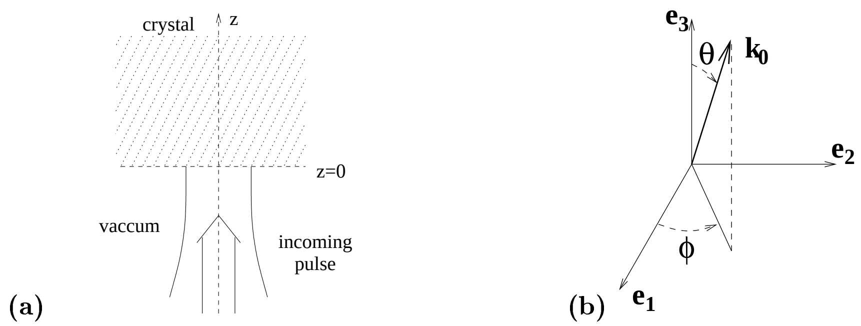



**Figure 1.** Picture (a): Boundary conditions. Picture (b): Description of the wavevector $\mathbf{k}_0$ collinear to $z$ in the crystallographic reference frame $(\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3)$.

$\Sigma$ is assumed to be a modulation of a high-frequency signal whose carrier wavelength is $\lambda_0$, or the superposition of a finite number of such modes. From the characteristic spatial radius (resp. time duration) of the source we can also define a length scale $R_0$ (resp. a time scale $T_0$, associated with the length $L_0 := cT_0$). Our study will take place in the framework where the dimensionless parameter $\varepsilon := \min\{\lambda_0/R_0, \lambda_0/L_0\}$ is small. As pointed out in the introduction, the order of magnitude $\bar{\mathcal{S}}$ of the source also plays a crucial role in that it determines the strength of the nonlinear interaction. Let us denote by $\bar{\chi}_1$ (resp. $\bar{\chi}_j$) the typical value taken by the Fourier transforms of the components of the $\chi^{(1)}$-tensor (resp. $\chi^{(j)}$-tensor) evaluated at frequency $2\pi c/\lambda_0$. The characteristic nonlinear amplitude is defined by $\bar{E}_{nl} := \min\left\{\bar{\chi}_1/\bar{\chi}_2, \sqrt{\bar{\chi}_1/\bar{\chi}_3}, ..., \sqrt[j]{\bar{\chi}_1/\bar{\chi}_j}, ...\right\}$. Our study takes place in the framework of weakly nonlinear waves, which reads as $\bar{\mathcal{S}}/\bar{E}_{nl} \ll 1$. This ratio may be related to the small parameter $\varepsilon$ through a new parameter $\alpha > 0$ such that $\bar{\mathcal{S}}/\bar{E}_{nl} = \varepsilon^\alpha$. Setting $\tilde{x} = x/\lambda_0$, $\tilde{y} = y/\lambda_0$, $\tilde{z} = z/\lambda_0$, $\tilde{t} = ct/\lambda_0$, $\tilde{\mathcal{D}} = \mathcal{D}/(\varepsilon_0 \bar{E}_{nl})$, and $\tilde{\mathcal{E}} = \mathcal{E}/\bar{E}_{nl}$ the dimensionless Maxwell equation reads as:

$$\tilde{\mathbf{rot}}\ \tilde{\mathbf{rot}}\ \tilde{\mathcal{E}} = -\tilde{\mu_0}\partial_{\tilde{t}\tilde{t}}\tilde{\mathcal{D}},$$

where $\tilde{\mu_0} = \varepsilon_0\mu_0 c^2 = 1$. The source $\tilde{\mathcal{S}}$ has a high-frequency expansion of the form:

$$\tilde{\mathcal{S}}(\tilde{x},\tilde{y},\tilde{t}) = \frac{1}{2}\varepsilon^\alpha \sum_{\omega_f \in \Omega_{\mathcal{S}}} \begin{pmatrix} v_x^f(\varepsilon\tilde{t}, \varepsilon\tilde{x}, \varepsilon\tilde{y}) \\ v_y^f(\varepsilon\tilde{t}, \varepsilon\tilde{x}, \varepsilon\tilde{y}) \\ 0 \end{pmatrix} e^{-i\omega_f \tilde{t}} + cc, \tag{5}$$

where $\Omega_{\mathcal{S}}$ is the collection of the high carrier frequencies $\omega_f$. $\mathbf{v}^f$ is the slowly varying envelope of the mode with carrier frequency $\omega_f$. Here "slowly varying" means that the typical scale of the variations of the smooth function $(T, X, Y) \mapsto \mathbf{v}^f(T, X, Y)$ is of order 1. Note that a dimensionless propagation distance $\tilde{z}$ of the order of $\varepsilon^{-1}$ corresponds to a physical distance of the order of $R_0$, while a dimensionless distance $\tilde{z}$ of the order of $\varepsilon^{-2}$ corresponds to a physical distance of the order of $R_0^2/\lambda_0$ which is the so-called Rayleigh distance.

From now on we drop the tildes. We assume a priori that the electric field can be expanded in a power series of the small parameter $\varepsilon$ and in a series with respect to a set of rapid phases $k^f z - \omega_f t$:

$$\mathcal{E} = \frac{1}{2}\varepsilon^\alpha \sum_{(\omega_f, k^f) \in H} \left(\mathbf{E}^f(\varepsilon t, \varepsilon x, \varepsilon y, \varepsilon z) e^{i(k^f z - \omega_f t)} + cc\right), \tag{6}$$

$$\mathbf{E}^f(T, X, Y, Z) = \sum_{j=0}^{\infty} \varepsilon^j \mathbf{E}_j^f(T, X, Y, Z), \tag{7}$$

where $\mathbf{E}^f$ is the slowly varying envelope of the mode whose rapid phase is $(\omega_f, k^f)$. The functions $\mathbf{E}_j^f$ are smooth in all their arguments. $H$ denotes the set of the rapid phases $(\omega_f, k^f)$ which are contained in the field $\mathcal{E}$. In case of linear medium the modes propagate without interaction and the set of high frequencies $\{\omega_f, \exists\ k^f$ such that $(\omega_f, k^f) \in H\}$ is equal to $\Omega_{\mathcal{S}}$. In case of nonlinear medium, the generation of new phases (the so-called harmonics) is expected so that the series (6) may contain much more terms than in the source.

## 3. PROPAGATION IN A BIREFRINGENT CRYSTAL

We introduce the geometric framework (see Fig. 1b). We first define a reference frame $(x, y, z)$ associated with the pulse whose carrier wavevector $\mathbf{k}_0$ is collinear to the $z$-axis. We then introduce a reference frame $(1, 2, 3)$ associated with the optic axis of the crystal, where $\mathbf{e}_3$ is the main optic axis. We denote by $\theta$ the angle between the wavevector and the main optic axis. $\phi$ is the angle between the projection of the carrier wavevector onto the plane $(\mathbf{e}_1, \mathbf{e}_2)$ and the axis collinear to $\mathbf{e}_1$. The transition matrix between the reference frames $(x, y, z)$ and $(1, 2, 3)$ is denoted by $U$. The linear susceptibility is defined as the Fourier transform of the tensor $\chi^{(1)}$ defined by (2). It is a diagonal matrix $\hat{\chi}^{(1)}_{123}$ in the frame $(1, 2, 3)$, while in the reference frame $(x, y, z)$ the tensor $\hat{\chi}^{(1)}_{xyz}$ is $U^{-1}\hat{\chi}^{(1)}_{123}U$. In the following $\chi$ is a shorthand for the matrix $\hat{\chi}^{(1)}_{xyz} + I_d$.

### 3.1. Principle of the high-frequency expansion

We present the principle of the high-frequency expansion method. It can be applied if the source can be expanded as (5). We proceed to a priori expansions of the field inside the crystal of the kind (6-7). In linear media (or equivalently for evanescent sources $\alpha \gg 1$) all nonlinear phenomena can be neglected, and the set of the frequencies $\omega$ which are contained in $H$ is imposed by the source and is equal to $\Omega_S$. Otherwise the generation of harmonics should be taken into account so that the set $H$ could be much larger than in the linear case.

The establishment of the propagation equations for the slowly varying envelopes obeys the following scheme. The form (6-7) is substituted into Eq. (1). Collecting the terms with similar orders in $\varepsilon$ and the same rapid phases $(\omega_f, k^f)$, we get a family of equations. These equations can be decomposed into coupled systems of equations parametrized by the rapid phases. In linear media these systems are independent so that the envelopes of the different modes propagate independently, but in nonlinear media there are couplings between the propagation equations of the envelopes. If the form (6-7) is suitable, then the derived systems should have unique solutions. Actually we shall show the two following statements. First the rapid phases must satisfy dispersion relations which read as compatibility conditions for the existence of the high-frequency expansion (6). Second the leading order terms $\mathbf{E}^f_0$ are determined by compatibility conditions for the existence of the series expansion (7).

The form (6-7) is an ansatz, that is to say an a priori form of the solution which is valid in a given domain, here for $z \leq \varepsilon^{-1}$. It is compatible with the boundary conditions and the source. It is self-similar with respect to the operators that are encountered in the Maxwell equation.

### 3.2. The set of the rapid phases

We aim at showing here that the rapid phases $(\omega_f, k^f)$ of the set $H$ should fulfill the so-called dispersion equation. By substituting the ansatz (6-7) into Eq. (1) and collecting the coefficients with power $\varepsilon^\alpha$ and phases $(\omega_f, k^f)$, we get that the leading order term $\mathbf{E}^f_0$ should satisfy:

$$k^{f\,2} \begin{pmatrix} E^f_{0x} \\ E^f_{0y} \\ 0 \end{pmatrix} = \frac{\omega^2}{c^2}\chi \mathbf{E}^f_0. \tag{8}$$

For any $\omega$ there exist two positive real numbers $n_a$ and $n_b$ and two polarizations $\mathbf{s}_a$ and $\mathbf{s}_b$ so that $\mathbf{s}_a$ and $\mathbf{s}_b$ are unit vectors and $(n_a\omega c^{-1}, \mathbf{s}_a)$ and $(n_b\omega c^{-1}, \mathbf{s}_b)$ are solutions of the equations (8). The dispersion relation, the group velocity and the dispersion coefficient of the waves are defined as follows:

$$k_m(\omega) := \frac{\omega n_m(\omega)}{c}, \quad v_m(\omega) := \left(\frac{\partial k_m}{\partial \omega}\right)^{-1}, \quad \sigma_m(\omega) := k_m \frac{\partial^2 k_m}{\partial \omega^2}, \quad m = a, b. \tag{9}$$

In order to fulfill condition (8), the set $(\omega_f, k^f, \mathbf{E}^f_0)$ must satisfy one of the three following alternatives:
- Either $k^f = k_a(\omega_f)$ and the components of $\mathbf{E}^f_0$ parallel to $\mathbf{s}_a(\omega_f)$ may be non-vanishing.
- Either $k^f = k_b(\omega_f)$ and the components of $\mathbf{E}^f_0$ parallel to $\mathbf{s}_b(\omega_f)$ may be non-vanishing.
- Or $k^f \notin \{k_a(\omega_f), k_b(\omega_f)\}$, and then necessarily $\mathbf{E}^f_0 \equiv 0$.

Note that the third option simply means that modes which are not phase-matched cannot have an envelope of leading order $\varepsilon^\alpha$.

We can now give a suitable description of the set $H$ of the rapid phases. The set $H$ in the general nonlinear framework should at least contain the rapid phases that directly originate from the source $\mathcal{S}$, that is to say:

$$H_{\mathcal{S}} = H_{a,\mathcal{S}} \cup H_{b,\mathcal{S}},$$

where $H_{a,\mathcal{S}}$ and $H_{b,\mathcal{S}}$ are the subsets of the rapid phases which satisfy either the $a$ dispersion relation or the $b$ relation:

$$H_{m,\mathcal{S}} = \{(\omega, k) \text{ such that } \omega \in \Omega_{\mathcal{S}} \text{ and } k = k_m(\omega)\}.$$

In a nonlinear medium the generation of new frequencies is expected. The choice of the ansatz should take into account this phenomenon and that is why the set $H$ of all possible rapid phases reads as the extended form:

$$H = \left\{ (\omega, k) \text{ such that } \exists n \in \mathbb{N},\ (\omega_i, k^i) \in H_{\mathcal{S}}, \omega = \sum_{j=1}^{n} \omega_i, \text{ and } k = \sum_{j=1}^{n} k_i \right\}.$$

The rapid phases $(\omega, k) \in H$ for which $\omega \notin \Omega_{\mathcal{S}}$ correspond to the so-called harmonic modes. Only the rapid phases of $H$ which satisfy one of the two phase matching conditions may possess a non-vanishing zero-th order component $\mathbf{E}_0^f$. That is why the physically relevant modes are those for which the rapid phases belong to $H_{ac}$:

$$H_{ac} = \{(\omega, k) \in H \text{ such that } k = k_a(\omega) \text{ or } k = k_b(\omega)\}.$$

The leading order terms for the other harmonic phases $(\omega_f, k^f) \in H \backslash H_{ac}$ are at most of order $\varepsilon^{2\alpha}$, that is to say $\mathbf{E}^f = \varepsilon^\alpha \mathbf{E}_\alpha^f + O(\varepsilon^{\alpha+1})$. Nevertheless it is technically necessary to take into account the harmonic modes that are not phase-matched so as to close the propagation equations. Furthermore these harmonic modes are fundamentally important because they may be intermediate factors for non-negligible cascaded $\chi^2$ nonlinear effects.

### 3.3. Boundary condition

If we assume that the source $\mathcal{S}$ can be expanded as (5), and accordingly that the field $\mathcal{E}$ inside the crystal is of the form (6-7), then collecting the coefficients with power $\varepsilon^\alpha$ and high carrier frequency $\omega_f$ establishes the continuity conditions which impose that the components parallel to the boundary surface of the input field $\mathcal{S}$ and of the field $\mathcal{E}$ should be equal, while there are no condition for the normal components. This imposes that the type $m$ mode $(m = a, b)$ with carrier frequency $\omega_f$ should be at $z = 0^+$:

$$\mathbf{E}_{0m}^f(\varepsilon t, \varepsilon x, \varepsilon y, z = 0^+) = \frac{2}{1 + n_m(\omega)} \frac{1}{s_{mx}^2 + s_{my}^2} \begin{pmatrix} s_{mx}^2 & s_{mx}s_{my} & 0 \\ s_{mx}s_{my} & s_{my}^2 & 0 \\ s_{mx}s_{mz} & s_{my}s_{mz} & 0 \end{pmatrix} \mathbf{v}^f(\varepsilon t, \varepsilon x, \varepsilon y).$$

## 4. GENERAL LINEAR CASE

We first address the case of evanescent sources so that the nonlinear polarization can be neglected. The following results hold true when the two eigenindices $(n_a, n_b)$ are different from each other. Note that the occurrence of the case $n_a = n_b$ corresponds to very particular configurations which will be addressed in Sections 6-7. Applying the methodology described in Section 3.1, we can show[4] that the input wave (5) breaks into the sum of modulations of high frequency signals, which also divide into two modes which all propagate independently. For each high frequency $\omega$ and mode $m = a, b$, the Poynting vector of the mode is collinear to the row vector:

$$\mathbf{u}_m(\omega) = \left( \frac{s_{mx}s_{mz}}{s_{mx}^2 + s_{my}^2}, \frac{s_{my}s_{mz}}{s_{mx}^2 + s_{my}^2}, -1 \right)^T. \qquad (10)$$

In the reference frame $(\varepsilon t, \varepsilon x, \varepsilon y, \varepsilon z)$ the slowly varying envelope $E_{0m}$ of the mode satisfies the transport equation with respect to the macroscopic variables $T = \varepsilon t$, $X = \varepsilon x$, $Y = \varepsilon y$, and $Z = \varepsilon z$:

$$\partial_Z E_{0m} + \left(-u_{mx}(\omega)\partial_X - u_{my}(\omega)\partial_Y + v_m(\omega)^{-1}\partial_T\right) E_{0m} = 0,$$

starting from $E_{0m}(T,X,Y,Z=0)=\frac{2}{1+n_m(\omega)}v_x(T,X,Y)$. In the moving reference frame $(\varepsilon(t-z/v_m),\varepsilon(x+u_{mx}z),\varepsilon(y+u_{my}z),\varepsilon^2 z)$ the slowly varying envelope $E_{0m}$ of the mode satisfies a Schrödinger-type equation with respect to the long scale variable $\zeta=\varepsilon^2 z$:

$$2ik_m(\omega)\partial_\zeta E_{0m}+\mathcal{L}_m(\omega)E_{0m}+\mathcal{K}_m(\omega)E_{0m}=0, \tag{11}$$

where the diffraction operator $\mathcal{L}_m(\omega)$ is anisotropic:

$$\begin{aligned}
\mathcal{L}_m(\omega) &= c_{m,xx}(\omega)\partial_{XX}+2c_{,xy}(\omega)\partial_{XY}+c_{m,yy}(\omega)\partial_{YY}, && (12)\\
c_{a,xx}(\omega) &= \frac{n_a^2}{n_b^2-n_a^2}\left(\frac{\chi_{zy}}{\chi_{zz}}+2u_{ay}\right)^2+\frac{n_a^2 s_{ax}^2}{\chi_{zz}(s_{ax}^2+s_{ay}^2)}+\frac{s_{ay}^2}{(s_{ax}^2+s_{ay}^2)^2} && (13)\\
c_{a,xy}(\omega) &= -\frac{n_a^2}{n_b^2-n_a^2}\left(\frac{\chi_{zy}}{\chi_{zz}}+2u_{ay}\right)\left(\frac{\chi_{zx}}{\chi_{zz}}+2u_{ax}\right)+\frac{n_a^2 s_{ax}s_{ay}}{\chi_{zz}(s_{ax}^2+s_{ay}^2)}-\frac{s_{ax}s_{ay}}{(s_{ax}^2+s_{ay}^2)^2} && (14)\\
c_{a,yy}(\omega) &= \frac{n_a^2}{n_b^2-n_a^2}\left(\frac{\chi_{zx}}{\chi_{zz}}+2u_{ax}\right)^2+\frac{n_a^2 s_{ay}^2}{\chi_{zz}(s_{ax}^2+s_{ay}^2)}+\frac{s_{ax}^2}{(s_{ax}^2+s_{ay}^2)^2} && (15)
\end{aligned}$$

and the dispersion operator $\mathcal{K}_m(\omega)$ contains crossed space-time derivatives:

$$\mathcal{K}_m(\omega)=-\sigma_m(\omega)\partial_{TT}+2k_m(\omega)(\partial_\omega u_{mx})(\omega)\partial_{TX}+2k_m(\omega)(\partial_\omega u_{my})(\omega)\partial_{TY}. \tag{16}$$

The expression of the diffraction operator is in agreement with the one derived by Dreger[5] where the author carries out an expansion of the explicit beam propagator in Fourier space. As pointed out in the introduction, the advantage of our approach is that it can still be applied when the beam propagator is not explicitly known and when nonlinearities arise.

The crossed space-time derivatives essentially originate from the fact that the Poynting vector of a monochromatic pulse is collinear to $\mathbf{u}_m(\omega)$ whose direction depends on the frequency $\omega$. Accordingly the different frequencies of a broadband pulse (or equivalently a short pulse) do not propagate exactly in the same direction which involves this additive "dispersion".

**Remark**: The expressions of the diffraction operator for a uniaxial crystal can then be rewritten in simple terms. Let us assume that $\chi_1=\chi_2:=\chi_o$ and $\chi_3=\chi_e$, with $\chi_e\neq\chi_o$. In the general framework $\theta\neq 0$ there are two distinct eigenindices, the so-called ordinary and extraordinary refractive indices:

$$n_o(\omega)=\chi_o(\omega)^{1/2},\qquad n_e(\omega)=\left(\frac{\chi_o(\omega)\chi_e(\omega)}{\cos^2\theta\chi_e(\omega)+\sin^2\theta\chi_o(\omega)}\right)^{1/2}. \tag{17}$$

These configurations correspond to an ordinary wave and an extraordinary wave, respectively. The unit polarization vector of an ordinary wave is simply $\mathbf{s}_o=(0,1,0)^T$, while the polarization vector of an extraordinary wave lies in the plane $(xz)$ and is given by $\mathbf{s}_e(\omega)=(\cos\beta,0,\sin\beta)^T$, where the angle $\beta(\omega)$ is:

$$\tan\beta(\omega)=\frac{\cos\theta\sin\theta(\chi_e(\omega)-\chi_o(\omega))}{\cos^2\theta\chi_e(\omega)+\sin^2\theta\chi_o(\omega)}.$$

**Ordinary wave.** The Poynting vector $\mathbf{u}_o$ which gives the direction of the rays along which the wave propagates in the geometric framework is simply $\mathbf{u}_o=(0,0,-1)^T$. The diffraction coefficients are $c_{o,xy}=0$, $c_{o,xx}=c_{o,yy}=1$, and $\partial_\omega\mathbf{u}_o=\mathbf{0}$. As a consequence an ordinary wave propagates according to the usual rules which govern the propagation of waves in linear isotropic media.

**Extraordinary wave.** The Poynting vector is $\mathbf{u}_e=(\tan\beta(\omega),0,-1)^T$. Thus $\beta$ is the walk-off angle, that is to say the angle between the carrier wavevector and the Poynting vector. The transverse diffraction coefficient $c_{e,xy}$ is zero while

$$c_{e,xx}(\omega)=\frac{\chi_o\chi_e}{\left(\cos^2\theta\chi_e+\sin^2\theta\chi_o\right)^2},\qquad c_{e,yy}(\omega)=\frac{\chi_o}{\cos^2\theta\chi_e+\sin^2\theta\chi_o}.$$

Note that the expressions of the diffraction coefficients $c_{m,..}$, $m=o,e$, are compatible with the asymptotic form of the Green's function in the case of a uniaxial medium given by Lax and Nelson.[6]

## 5. BIRADIAL WAVES IN BIAXIAL CRYSTALS

Let us assume $\chi_1 > \chi_2 > \chi_3$. We examine here the propagation in the particular configuration $\theta = \theta_r(\omega)$ and $\phi = 0$ with

$$\sin^2\theta_r = \frac{\chi_1 - \chi_2}{\chi_1 - \chi_3}.$$

The diffraction operator for the type $a$ wave is then degenerate, in the sense that there is no diffraction in the $y$-direction: $c_{a,yy} = c_{a,xy} = 0$, while $c_{a,xx} = 1$. The $a$ wave is polarized along the $y$-axis and its slowly varying envelope thus satisfies in the moving reference frame $(\varepsilon(t - z/v_a), \varepsilon x, \varepsilon y, \varepsilon^2 z)$ the Schrödinger equation:

$$2ik_a\partial_\zeta E_{0a} + \partial_{XX}E_{0a} - \sigma_a\partial_{TT}E_{0a} = 0,$$

where the macroscopic variables are $T = \varepsilon(t - z/v_a)$, $X = \varepsilon x$, $Y = \varepsilon y$, and $\zeta = \varepsilon^2 z$. Such an anomalous behavior is consistent with the results derived in Ref. 7 where the authors show that the asymptotic form of the Green's function is proportional to $z^{-1/2}$ instead of the standard $z^{-1}$-decay. This phenomenon is made transparent from our results, since the solution of a Schrödinger equation with a $d$-dimensional second-order operator spreads out as $z^{-d/2}$. This configuration could involve an interesting application in nonlinear optics. Consider high-intensity pulses $\alpha = 1$ so that the nonlinearity of the medium should be taken into account in the scales of diffractive optics. For the sake of simplicity we restrict ourselves to one of the three following classes[1]:

1) triclinic class with point group $\bar{1}$, such as Mica or $Al_2SiO_5$,
2) monoclinic class with point group $2/m$, such as $AgAuTe_4$ or $PbSiO_3$,
3) orthorombic class with point group $mmm$, such as $CaCl_2$, or $Al_2BeO_4$ (also called alexandrite).

The crystals of these classes are biaxial and have a vanishing $\chi^{(2)}$-tensor. This simplification allows us to get rid off the $\chi^{(2)}$-cascaded terms and to deal with a simple $\chi^{(3)}$-component which then reads as a simple Kerr effect. Assume that the incoming pulse is linearly polarized along the $y$-axis. Then the field stays linearly polarized along the $y$-axis and its slowly varying envelope satisfies in the moving reference frame $(\varepsilon(t - z/v_a), \varepsilon x, \varepsilon y, \varepsilon^2 z)$:

$$2ik_a\partial_\zeta E_0 + \partial_{XX}E_0 - \sigma_a\partial_{TT}E_0 + \gamma|E_0|^2E_0 = 0,$$

where $\gamma = (3/4)\hat{\chi}^{(3)}_{2222}(-\omega, \omega, \omega)$. We can also remove from this equation the group velocity dispersion term if there is no modulation of the input pulse at the time scale $\varepsilon^{-1}$, which is typically of the order of the picosecond, but only at scale $\varepsilon^{-2}$, which is typically of the order of the nanosecond. Then the slowly varying envelope of the field satisfies the standard one-dimensional Schrödinger equation:

$$2ik_a\partial_\zeta E_0 + \partial_{XX}E_0 + \gamma|E_0|^2E_0 = 0. \tag{18}$$

The one-dimensional nonlinear Schrödinger equation possesses the complete integrability property, which implies that stable solitons should be generated and propagate over large distances. If a pulse is focused onto a crystal plate according to the incident angle and polarization described here above, then in the $x$-transverse direction the profile of the pulse will not diffract and keep its original form while in the transverse $y$-direction the pulse will break into a soliton (or eventually several solitons) and radiation whose amplitude will decay as standard one-dimensional waves do in linear media, that is to say at rate $z^{-1/2}$.

In case of biaxial crystals with non-vanishing $\chi^{(2)}$-tensor the result is qualitatively the same, in the sense that the diffraction operator still reads as a one-dimensional second-order derivative, but $\chi^{(2)}$-cascaded terms make the nonlinear term more complicated.

## 6. CRITICAL CONFIGURATIONS FOR STRONGLY BIAXIAL CRYSTALS

If $\chi_1 > \chi_2 > \chi_3$, then a simple study of the matrix $\chi$ shows that the event $n_a = n_b$, corresponds to the configurations when $\phi = 0$ and $\theta = \pm\theta_c(\omega)$ where $\theta_c(\omega)$ is defined by:

$$\sin^2\theta_c = \frac{\chi_1/\chi_2 - 1}{\chi_1/\chi_3 - 1}, \tag{19}$$

which defines the two optic axes of the biaxial crystal. In such a configuration $n_a = n_b = \chi_2^{1/2}$, and the two mutually orthogonal polarization vectors are:

$$\mathbf{s}_a = (\cos\beta_c, 0, \sin\beta_c)^T, \quad \mathbf{s}_b = (0, 1, 0)^T,$$

where the angle $\beta_c(\omega)$ between the polarization vector $\mathbf{s}_a$ and the propagation axis $z$ is given by:

$$\tan\beta_c = -\sqrt{\left(1-\frac{\chi_2}{\chi_1}\right)\left(\frac{\chi_2}{\chi_3}-1\right)}.$$

Substituting the ansatz (6-7) with $H = \{(\omega, n_a(\omega))\}$ into Eq. (1) and collecting the coefficients of each power of $\varepsilon$, we get the following result. Let us denote by $E_{0a} = \mathbf{s}_a.\mathbf{E}_0$ and $E_{0b} = \mathbf{s}_b.\mathbf{E}_0$ the projections of the field $\mathbf{E}_0$ onto the eigenvectors $\mathbf{s}_a$ and $\mathbf{s}_b$. The scalar fields $E_{0a}$ and $E_{0b}$ satisfy the coupled equations in the reference frame $(\varepsilon(t - z/v_a), \varepsilon x, \varepsilon y, \varepsilon z)$:

$$\partial_Z E_{0a} - \tan\beta_c \partial_X E_{0a} = \frac{\tan\beta_c}{2\cos\beta_c}\partial_Y E_{0b}, \tag{20}$$

$$\partial_Z E_{0b} = \frac{\sin\beta_c}{2}\partial_Y E_{0a}, \tag{21}$$

starting from $E_{0a}(T,X,Y,Z=0) = \frac{2}{1+n_a(\omega)}v_x(T,X,Y)$ and $E_{0b}(T,X,Y,Z=0) = \frac{2}{1+n_a(\omega)}v_y(T,X,Y)$.

In Fourier space Eqs. (20-21) can be solved analytically. In the physical space, using the stationary phase method, we get that the field is concentrated on the circle with center $Z(\tan\beta_c)/2$ and radius $Z(\tan\beta_c)/2$ if the input field is localized around 0. In other words the wave surface has the shape of a cone. This conical refraction is a well-known phenomena which was predicted in 1832 by Hamilton and observed thereafter by Lloyd. Historical references and an elementary study of conical refraction can be found in Ref. 8. More advanced treatments are devoted to the subject.[9,10] In particular Warnick and Arnold[11] predicts additional fringes by computing the asymptotic form of the Green's function. To recover precisely these results one should take into account the second-order derivatives, which makes the evolution of the field more complicated. The strategy is still the same as in the other configurations. It consists in looking at the evolution of the field at the long scale $\varepsilon^2 z$ around the points defined by the transport equation, that is to say the cone defined by $(x - z(\tan\beta_c)/2)^2 + y^2 = (z(\tan\beta_c)/2)^2$. This specific study will be carried out elsewhere. Nevertheless, we would like to add the following comment that gives a new insight into the phenomena that govern conical refraction. We would like to show that the additional fringes arise in the scales of geometric optics, and that there is no need to introduce diffractive effects to explain the observed complex structures. In the moving reference frame $(\varepsilon(t - z/v_a), \varepsilon(x + z(\tan\beta_c)/2), \varepsilon y, \varepsilon z)$ the equations (20-21) read as:

$$\partial_Z E_{0a} = \frac{\tan\beta_c}{2}\partial_X E_{0a} + \frac{\tan\beta_c}{2\cos\beta_c}\partial_Y E_{0b}, \tag{22}$$

$$\partial_Z E_{0b} = \frac{\sin\beta_c}{2}\partial_Y E_{0a} - \frac{\tan\beta_c}{2}\partial_X E_{0b}, \tag{23}$$

where the macroscopic variables are $T = \varepsilon(t - z/v_a)$, $X = \varepsilon(x + z(\tan\beta_c)/2)$, $Y = \varepsilon y$, and $Z = \varepsilon z$. Substituting the second equation into the first one, and vice-versa, establishes that the modes $E_{0m}$ for $m = a$ and $b$ obey the standard wave equations with uniform "velocity" $(\tan\beta_c)/2$:

$$\partial_{ZZ} E_{0a} = \frac{\tan^2\beta_c}{4}(\partial_{XX} + \partial_{YY})E_{0a}, \tag{24}$$

$$\partial_{ZZ} E_{0b} = \frac{\tan^2\beta_c}{4}(\partial_{XX} + \partial_{YY})E_{0b}, \tag{25}$$

where $Z$ plays the role of the usual time. The initial conditions are imposed by $E_{0m}(Z = 0)$ and $\partial_Z E_{0m}(Z = 0)$. As is well-known the solution of the wave equation $u_{tt} = c^2\Delta u$ satisfies the Huygens principle which states that, if the Laplacian acts on a space with odd dimension $d$, then the solution $u(x,t)$ depends only on the initial data at $t = 0$ for $x_0 \in \{x_0 \in \mathbb{R}^d, |x_0 - x| = ct\}$. Thus an initial delta-like pulse at $t = 0$, $x = 0$ will give rise at time $t$ to a pulse concentrated on the circle with center 0 and radius $ct$. This justifies the accuracy of geometric optics since the propagation of a pulse is then accurately described by rays. However this property does not hold true for even dimension, since the solution $u(x,t)$ then depends on the initial data at $t = 0$ in the whole cone $x_0 \in \{x_0 \in \mathbb{R}^d, |x_0 - x| \leq t\}$. In the standard wave equation, the space has dimension $d = 3$ and the Huygens principle is satisfied. In our case, $Z$ plays the role of $t$ and $d = 2$, which proves that complex structure inside the main cone can be generated during the propagation, even in the scales of geometric optics where simple ray transport is expected. The evolution of an input Gaussian beam governed by Eqs. (24-25) is plotted in Fig. 2. We refer to the standard literature on the wave equation for a description of the different phenomena that can arise.[12,13]

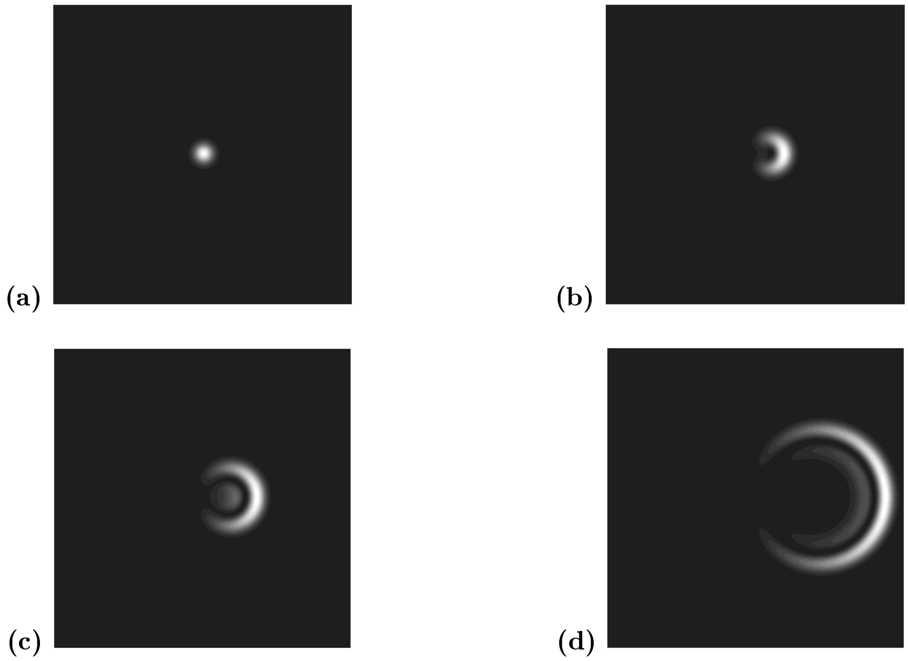



**Figure 2.** Conical refraction of a Gaussian beam polarized along the $x$-axis for different values of $Z$. $Z = 0$ (a), $Z = 4.13$ (b), $Z = 8.26$ (c), and $Z = 20.65$ (d). Here $\cos\beta_c = 0.9$ or equivalently $(\tan\beta_c)/2 = 0.24$.

## 7. CRITICAL CONFIGURATIONS FOR WEAKLY BIAXIAL CRYSTALS

This section addresses the case of weakly biaxial or uniaxial crystals in the case when $n_a = n_b$. More exactly we assume here that the $\mathbf{e}_3$-axis and the propagation axis $z$ of the input pulse are collinear $\theta = 0$. Furthermore the susceptibilities $\chi_1$ and $\chi_2$ are close to each other so that they can be written as: $\chi_1 = \chi_o$, $\chi_2 = \chi_o - \varepsilon^2\eta_\chi$, and $\chi_3 = \chi_e < \chi_o$. We shall neglect in this section the nonlinear terms. The leading order term $\mathbf{E}_0$ of the slowly varying envelope is then transverse and it satisfies the coupled Schrödinger equations in the moving reference frame $(\varepsilon(t - z/v_o), \varepsilon x, \varepsilon y, \varepsilon^2 z)$:

$$2ik_o\partial_\zeta E_{0x} + \rho\partial_{XX}E_{0x} + \partial_{YY}E_{0x} + (\rho-1)\partial_{XY}E_{0y} - \sigma_o\partial_{TT}E_{0x} = k_o^2\eta_\chi\left(\sin^2(\phi)E_{0x} + \cos(\phi)\sin(\phi)E_{0y}\right), \quad (26)$$

$$2ik_o\partial_\zeta E_{0y} + \partial_{XX}E_{0y} + \rho\partial_{YY}E_{0y} + (\rho-1)\partial_{XY}E_{0x} - \sigma_o\partial_{TT}E_{0y} = k_o^2\eta_\chi\left(\cos(\phi)\sin(\phi)E_{0x} + \cos^2(\phi)E_{0y}\right), \quad (27)$$

starting from $\mathbf{E}_0(T, X, Y, \zeta = 0) = \frac{2}{1+n_o(\omega)}\mathbf{v}(T, X, Y)$, where $\rho = \chi_o/\chi_e$ and the macroscopic variables are $T = \varepsilon(t - z/v_o)$, $X = \varepsilon x$, $Y = \varepsilon y$, and $\zeta = \varepsilon^2 z$. Taking the spatial Fourier transform with respect to the transverse coordinates, this system reduces to a system of ordinary differential equations that can be easily solved.

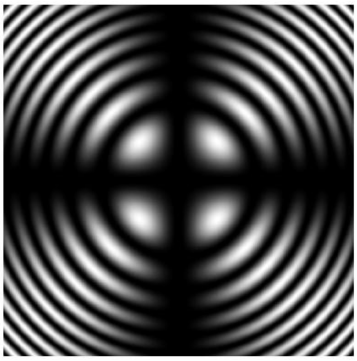
Consider the experimental set up where a linearly polarized divergent light beam emerging from a polarizer $P_x$ is normally incident onto a plane parallel crystal plate, and the interference pattern at the output of the crystal plate is analyzed. The initial pulse is polarized along the $x$-axis, and we retain only the component of the output pulse which is $y$-polarized. Neglecting group velocity dispersion we have (in Fourier space or equivalently in the far field):

$$|\hat{E}_y(\zeta, k_x, k_y)|^2 = F_\phi\left(k_x\rho_c, k_y\rho_c, \eta_\chi k_o\zeta/4\right)|\hat{E}_x(0, k_x, k_y)|^2,$$

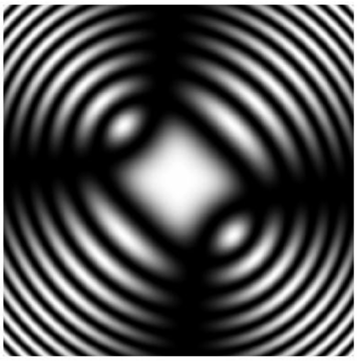
$$F_\phi(u, v, \eta) = (2uv + \eta\sin(2\phi))^2\mathrm{sinc}^2\left(\sqrt{(u^2+v^2)^2 + \eta^2 + 2\eta((v^2-u^2)\cos(2\phi) + 2uv\sin(2\phi))}\right),$$

where $\mathrm{sinc}(s) = \sin(s)/s$ and $\rho_c^2 = (\chi_o/\chi_e - 1)z_c/(4k_o)$.

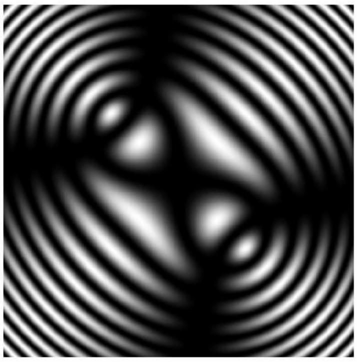
Fig. 3 plots the function $(u, v) \mapsto F_\phi(u, v, \eta)$ for different values of the parameters $\phi$ and $\eta$. Comparisons with experimental observations show excellent agreement. See Figure 14.26 in Ref. 8 for an observation of Fig. 3f, Figures 465-466 in Ref. 14 for observations of Fig. 3d and Fig. 3e, and Figure 5.30 in Ref. 15 for another observation of



**(a)** $\eta = 0$ and any $\phi$

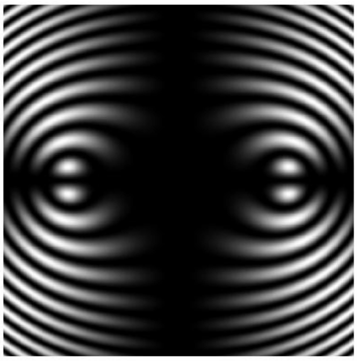


**(b)** $\eta = \pi/2$ and $\phi = \pi/4$

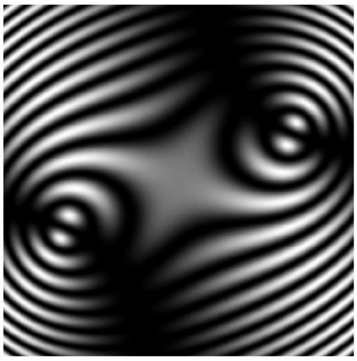


**(c)** $\eta = \pi$ and $\phi = \pi/4$

**(d)** $\eta = 3\pi/2$ and $\phi = \pi/4$



**(e)** $\eta = 2\pi$ and $\phi = \pi$



**(f)** $\eta = 5\pi/2$ and $\phi = 7\pi/8$

**Figure 3.** Interference patterns from biaxial crystal plates between two crossed polarizers. Functions $(u, v) \mapsto F_\phi(u, v, \eta)$ over the domain $(-4, 4) \times (-4, 4)$ for different values of $\eta$ and $\phi$.

Fig. 3e. In particular Fig. 3f is the theoretical counterpart of the cover of the sixth edition of the book "Principles of Optics" by Born and Wolf[8] ! Let us briefly discuss the main properties of the functions $F_\phi$. If the initial polarization vector is collinear to one of the axis of the crystal ($\phi = 0$ or $\pi$), then $F_0$ has a factor $u^2v^2$, which shows that there is a centered dark cross, whatever $\eta_\chi$. If the initial polarization vector is collinear to the bisecting line of the axes of the crystal ($\phi = \pi/4$), then we have:

$$F_{\frac{\pi}{4}}(u,v,\eta) = (2uv+\eta)^2 \mathrm{sinc}^2\left(\sqrt{(u^2+v^2)^2+\eta^2+4\eta uv}\right), \quad (28)$$

which shows that the transmission at the center becomes nonzero when $\eta$ increases and may even be 1 for particular values of $\eta$. Indeed, whatever $\phi$, the transmission at $u = v = 0$ is $F_\phi(0,0,\eta) = \sin(\eta)^2\sin(2\phi)^2$. If $\phi = 0$, it is always 0, but if $\phi = \pi/4$, it is equal to $\sin(\eta)^2$ which is maximal and equal to 1 when $\eta = \pi/2 \bmod \pi$. This implies that a plane wave is fully transmitted in this configuration.

The results derived in this section provide the principle and the precise characterization of electro-optic switching devices of the family of Pockels cells,[16] consisting of a KDP crystal plate between two crossed polarizers. KDP is uniaxial, that is to say $\eta_\chi = 0$. The output interference pattern is then the well-known Maltese cross (Fig. 3a). Applying an electric field between two faces of the KDP crystal involves an alteration of the distribution of the electric charges of the atoms and molecules which constitute the crystal, which affect the optical properties of the medium. The theory of electro-optics is well-known, and we refer for instance to Ref. 17 for a survey. In the case of point group $\bar{4}2m$ to which KDP crystal belongs, it is known that the the crystals become biaxial while they are uniaxial in the absence of external electric field, that is to say $\eta_\chi$ takes nonzero values which are imposed by the applied electric field. By applying the tension from 0 to the value corresponding to $\eta_\chi = 2\pi/(k_o(\omega)z_c)$, the transmittivity goes from 0 to 1 for an input plane wave with carrier frequency $\omega$. Finally note also that the transfer function $(u,v) \mapsto F_{\pi/4}(u,v,\pi/2)$ possesses a flat top hat. This configuration could then be used as a spatial filter as well.

## 8. CONCLUSION

We have derived the equations which govern the propagation of the slowly varying envelopes of pulses in a bulk medium presenting anisotropic properties. The strategy mainly consists in two steps. We first consider the Maxwell equations in the scales of optic geometric, that is to say for propagation distance of the same order as the radius of the beam or the duration of the pulse times the light velocity. In this framework the propagation equations read as transport equations, which actually give the propagation of the rays according to the law of geometric optics. Second we revisit the Maxwell equations in the moving frame indicated by the above-derived transport equations. In the scales of the geometric optics, the propagation equations are then trivial, which allows to consider larger propagation distances, of the order of the Rayleigh distance or the dispersion distance. In this framework the propagation equations read as Schrödinger-like equations, which actually give the propagations of the slowly varying envelope according to the law of diffractive optics.

By applying this methodology we have put into evidence that we can deal with many situations. We have recovered well-known results, but we have also exhibited closed form expressions for the diffraction operator which has led to original results regarding an anomalous diffraction in a very particular configuration. Another advantage of this method is that it can still be applied when we take into account the nonlinear susceptibility of the medium.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