JOURNAL OF MATHEMATICAL PHYSICS VOLUME 42, NUMBER 4 APRIL 2001

High-frequency asymptotics for Maxwell's equations in anisotropic media Part II: Nonlinear propagation and frequency conversion

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(Received 28 October 1999; accepted for publication 10 January 2001)

This paper is devoted to the derivation of the equations that govern the propagation and frequency conversion of pulses in noncentrosymmetric crystals. The method is based upon high-frequency expansions techniques for hyperbolic quasi-linear and semilinear equations. In the so-called geometric regime we recover the standard results on the frequency conversion of pulses in nonlinear crystals. In the diffractive regime we show that the anisotropy of the diffraction operator involves remarkable phenomena. In particular the phase matching angle of a divergent pulse depends on the distance between the waist and the crystal plate. Finally we detect a configuration where the beam propagation in a biaxial crystal involves the generation of spatial solitons thanks to an anomalous one-dimensional diffraction.
 [DOI: 10.1063/1.1354640]

I. INTRODUCTION

In 1961 Franken *et al.*[1] observed radiation at a double frequency when a ruby laser beam was directed into a quartz crystal. Unfortunately, because of phase mismatch of the fundamental and converted waves, the efficiency of conversion proved to be very low (about $10^{-10}\%$). The so-called phase matching condition which should be fulfilled for the second harmonic generation $\omega+\omega\mapsto 2\omega$ reads as $2\mathbf{k}(\omega)=\mathbf{k}(2\omega)$, or equivalently $n(2\omega)=n(\omega)$, where n is the refractive index. In the optical transparency region of isotropic crystals, and in anisotropic crystals for waves of identical polarizations, this condition is never fulfilled because of normal dispersion $(n(\omega)<n(2\omega))$. The use of anomalous dispersion is prohibited because the energy absorption is then very high. In 1962 Giordmaine[2] and Maker[3] simultaneously and independently proposed an ingenious method of matching the phase velocities of the fundamental and converted waves. The technique is based on the difference between the refractive indices of the waves with different polarizations in an anisotropic crystal. It is now current to reach efficiency of conversion of several ten percents.[4]

Anisotropy is a necessary condition for a medium to have a nonzero second order nonlinearity.[5] The χ^2-tensor is zero for any centrosymmetric crystal. The study of sum-frequency generation thus takes place in anisotropic media. The aim of this paper is to describe the effects of the anisotropy of the medium and to take a rigorous account of it in the study of the nonlinear regime and especially the frequency conversion phenomenon. The case of plane waves has been carefully studied in Ref. 6. We aim at deriving evolution equations for the slowly varying envelopes of broadband and divergent pulses by using a technique based on high-frequency expansions of the fields.[7]

The derived equations find practical applications in the framework of frequency conversion of high-power laser beams. Indeed the phase matching condition for efficient frequency doubling and tripling of laser beams is very drastic,[4] and it is therefore, necessary to detect the principal axis of the crystal with great precision. The standard process consists in observing the main output

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direction of the frequency converted pulse of a divergent pulse. This work was originally triggered by the experimental observation that the direction of the frequency converted pulse of a fundamental Gaussian pulse depends on the distance between the waist of the fundamental pulse and the crystal, even with a perfectly normal incidence. The departures for different distances exceed the high-precision level required for reaching the expected conversion performance. The results derived in this paper predict the phenomenon and allow to compute the direction of the phase matching angle as a function of the direction of the frequency converted pulse and the distance between the waist of the fundamental pulse and the crystal.

The results of this paper are also necessary for a careful treatment of the propagation and frequency conversion of partially coherent pulses. Indeed incoherent light with short coherence time is of interest for smoothing techniques for uniform irradiation in plasma physics.[8] If propagation of incoherent light in isotropic linear media is now rather well understood, the evolution of the statistical properties of incoherent pulses in anisotropic and/or nonlinear media has been insufficiently examined.[9] A high level of irradiation uniformity is required for both direct and indirect drive for Inertial Confinement Fusion.[8] This criterion can be reached by implementing active smoothing methods, such as Induced Spatial Incoherence with echelons,[10] Smoothing by Spectral Dispersion (SSD),[11] Smoothing by multimode Optical Fiber (SOF).[12] All these methods involve the illumination on the target with an intensity which is a time varying speckle pattern, so that the time integrated intensity averages towards a flat profile. As an unavoidable drawback the optical smoothing techniques also involve phase modulations in the amplifiers and frequency converters (SSD), or even intensity modulations (SOF).

The framework for high-frequency expansions of the solutions of Maxwell's equations follows from the appearance of the small parameter δ which has the order of magnitude of the carrier wavelength of light divided by the next smallest characteristic length present in the problem. If we assume that the carrier wavelength is 1, then we have seen in Ref. 13 that for propagation length of order δ^{-1}, which corresponds to the scales of the so-called geometric optics, evolution equations read as transport equations with constant velocity. Further, in the moving pulse-time frame (moving according to the velocity exhibited by the geometric transport equations), for propagation length of order δ^{-2}, which corresponds to the scales of diffractive optics, the evolution of the field is governed by a Schrödinger equation.

The first nonlinear effect we discuss in this paper is the sum frequency generation. A nonlinear χ^2-type function applied to expressions of the form $\Sigma_f \mathbf{E}_f^{\delta}(\delta t, \delta \mathbf{x}) \exp i(k^f z - \omega_f t)$ will produce harmonics, that is to say expression with phase $(k^{f_1}+k^{f_2})z-(\omega_{f_1}+\omega_{f_2})t$. If the couples (ω_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.

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 δ^{α}, then a p-wave interaction process will be noticeable for propagation length of order $\delta^{-(p-1)\alpha}$. Since we are mainly concerned in this paper with second-order nonlinearity, it means that the nonlinear effect will appear for propagation length of the order of $\delta^{-\alpha}$. Accordingly $\alpha=1$ will correspond to nonlinear geometric optics and $\alpha=2$ to nonlinear diffractive optics.

The paper is organized as follows. First we describe the general configuration at hand in Sec. II. Section III is devoted to the derivations of the dispersion relation, the phase matching and the suitable expanded form of the solution of the Maxwell equations. We address in Secs. IV and V the frequency conversion in birefringent crystals. In Sec. VI we derive the propagation equation of the slowly varying envelope of the field when the phase-matching conditions for frequency generation are not fulfilled. In Sec. VII we study a particular configuration which should allow the generation and propagation of spatial solitons.

II. FORMULATION AND SCALING

We consider an incident beam incoming from the left onto a nonmagnetic nonlinear crystal that occupies the domain $\mathbb{R}^3_+ := \{(x,y,z) \in \mathbb{R}^3, z>0\}$. 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

$$\mathbf{rot}\ \mathbf{rot}\ \mathcal{E} = -\mu_0 \partial_t^2 \mathcal{D}, \tag{1}$$

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

$$\mathcal{D}_l = \varepsilon_0 \mathcal{E} + \varepsilon_0 \chi^{(1)} * \mathcal{E}, \tag{2}$$

$$\mathcal{P}_{nl} = \varepsilon_0 \chi^{(2)} * (\mathcal{E},\mathcal{E}) + \varepsilon_0 \chi^{(3)} * (\mathcal{E},\mathcal{E},\mathcal{E}) + \cdots, \tag{3}$$

$$\chi^{(j)} * (\mathcal{E},\dots,\mathcal{E}) = \int_{-\infty}^{t} dt_1 \cdots \int_{-\infty}^{t} dt_j \chi^{(j)}(t-t_1,\dots,t-t_j) : \mathcal{E}(t_1) \cdots \mathcal{E}(t_j). \tag{4}$$

ε_0 and μ_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 Σ 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 Σ is assumed to be a modulation of a high-frequency signal whose carrier wavelength is λ_0, or the superposition of a finite number of such modes. From the characteristic spatial (resp. temporal) variations 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 $\delta := \min\{\lambda_0/R_0, \lambda_0/L_0\}$ is small. As pointed out in the introduction, the order of magnitude $\overline{\mathcal{S}}$ of the source also plays a crucial role in that it determines the strength of the nonlinear interaction. Let us denote by $\overline{\chi_1}$ (resp. $\overline{\chi_2}$) the typical value taken by the Fourier transforms of the components of the $\chi^{(1)}$-tensor (resp. $\chi^{(2)}$-tensor) evaluated at frequency $2\pi c/\lambda_0$. The characteristic nonlinear amplitude is defined by $\overline{E_{nl}} := \overline{\chi_1}/\overline{\chi_2}$. Our study takes place in the framework of weakly nonlinear waves, which reads as $\overline{\mathcal{S}}/\overline{E_{nl}} \ll 1$. This ratio may be related to the small parameter δ through a new parameter $\alpha > 0$ such that $\overline{\mathcal{S}}/\overline{E_{nl}} = \delta^\alpha$. Setting $\tilde{x} = x/\lambda_0$, $\tilde{y} = y/\lambda_0$, $\tilde{z} = z/\lambda_0$, $\tilde{t} = ct/\lambda_0$, $\tilde{\mathcal{D}} = \mathcal{D}/(\varepsilon_0 \overline{E_{nl}})$, and $\tilde{\mathcal{E}} = \mathcal{E}/\overline{E_{nl}}$ the dimensionless Maxwell equation reads as

$$\tilde{\mathbf{rot}}\ \tilde{\mathbf{rot}}\ \tilde{\mathcal{E}} = -\tilde{\mu}_0 \partial_{\tilde{t}}^2 \tilde{\mathcal{D}},$$

where $\tilde{\mu}_0 = \varepsilon_0 \mu_0 c^2 = 1$. The source $\tilde{\mathcal{S}}$ has a high-frequency expansion of the form

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

where *cc* is a shorthand for ''complex conjugate.'' $\Omega_{\mathcal{S}}$ is the collection of the high-carrier frequencies ω_f. $\mathbf{v}^f$ is the slowly varying envelope of the mode with carrier frequency ω_f. Note that a dimensionless propagation distance $\tilde{z}$ of the order of δ^{-1} corresponds to a physical distance of the order of R_0, while a dimensionless distance $\tilde{z}$ of the order of δ^{-2} corresponds to a physical distance of the order of R_0^2/λ_0 which is the well-known 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 δ and in a series with respect to a set of rapid phases $k^f z - \omega_f t$

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

$$\mathbf{E}^f(T,X,Y,Z)=\sum_{j=0}^{\infty}\delta^j\mathbf{E}_j^f(T,X,Y,Z), \tag{6b}$$

where $\mathbf{E}^f$ is the slowly varying envelope of the mode whose rapid phase is (ω_f,k^f). The functions $\mathbf{E}_j^f$ are smooth in all their arguments. H denotes the set of the rapid phases (ω_f,k^f) which are contained in the field $\mathcal{E}$. In case of linear medium[13] 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 (5).

III. PROPAGATION IN A BIREFRINGENT CRYSTAL

We introduce the geometric framework. We first define a reference frame (x,y,z) associated with the pulse whose carrier wave vector $\mathbf{k}_0$ 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 θ the angle between the wave vector and the main optic axis. ϕ is the angle between the projection of the carrier wave vector 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.

A. 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) with $\alpha>0$ (weak nonlinearity). In linear media (or equivalently for evanescent sources $\alpha\gg 1$) all nonlinear phenomena can be neglected, and the set of the frequencies ω which are contained in H is imposed by the source and is equal to $\Omega_{\mathcal{S}}$. 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 establishing of the propagation equations for the slowly varying envelopes obeys the following scheme. The form (6) is substituted into Eq. (1). Collecting the terms with similar orders in δ and the same rapid phases (ω_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,[13] but in nonlinear media there are coupling between the propagation equations of the envelopes. If the form (6) 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 (6a). Second, the leading order terms $\mathbf{E}_0^f$ are determined by compatibility conditions for the existence of the series expansion (6b).

The form (6) is an ansatz, that is to say an *a priori* form of the solution which is valid in a given domain, here for $z\lesssim\delta^{-1}$. 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. This fact was established in Ref. 13 for the linear operators, and we shall see in the following that the expansion (6) is also self-similar with respect to the nonlinear operators.

B. Expansions of the linear terms

The linear susceptibility is defined as the Fourier transform of the tensor $\chi^{(1)}$ defined by (2). It is a diagonal matrix $\hat{\chi}_{123}^{(1)}$ in the frame (1, 2, 3), while in the reference frame (x,y,z) the tensor $\hat{\chi}_{xyz}^{(1)}$ is $U^{-1}\hat{\chi}_{123}^{(1)}U$. In the following χ is a shorthand for the matrix $\hat{\chi}_{xyz}^{(1)}+I_d$. If $\mathcal{E}$ is of the form $\mathcal{E}=\frac{1}{2}\delta^{\alpha}(\mathbf{E}(\delta t,\delta x,\delta y,\delta z)e^{i(kz-\omega t)}+cc)$, then the contribution of the linear induction to the Maxwell equation (1) and the **rot rot** $\mathcal{E}$ term can be expanded as powers of δ. Denoting by $T=\delta t$, $X=\delta x$, $Y=\delta y$, and $Z=\delta z$ the slowly varying variables, we have on the one hand

$$-\mu_0\partial_t^2\mathcal{D}_l=\tfrac{1}{2}\delta^\alpha(\mathbf{D}_0(\mathbf{E})+\delta\mathbf{D}_1(\mathbf{E})+\delta^2\mathbf{D}_2(\mathbf{E})+O(\delta^3))e^{i(kz-\omega t)}+cc, \tag{7}$$

where the $\mathbf{D}_j(\mathbf{E})$ are linear functions of $\mathbf{E}$ given by

$$\mathbf{D}_0(\mathbf{E})=\frac{\omega^2}{c^2}\chi\mathbf{E},\quad \mathbf{D}_1(\mathbf{E})=\frac{i}{c^2}(\omega^2\chi)'\partial_T\mathbf{E},\quad \mathbf{D}_2(\mathbf{E})=-\frac{1}{2c^2}(\omega^2\chi)''\partial_T^2\mathbf{E}, \tag{8}$$

and the primes stand for partial derivatives with respect to ω. On the other hand the **rot rot** $\mathcal{E}$ term writes

$$\mathbf{rot}\ \mathbf{rot}\ \mathcal{E}=\tfrac{1}{2}\delta^\alpha(\mathbf{R}_0(\mathbf{E})+\delta\mathbf{R}_1(\mathbf{E})+\delta^2\mathbf{R}_2(\mathbf{E}))e^{i(kz-\omega t)}+cc, \tag{9}$$

where the mappings $\mathbf{R}_j(\mathbf{E})$ are sums of partial derivatives of E with respect to space coordinates of order j that are given in Ref. 13.

C. Dispersion relations for the rapid phases

We aim at showing here that the rapid phases (ω_f,k^f) of the set H should fulfill the so-called dispersion equation. By substituting the ansatz (6) into Eq. (1) and collecting the coefficients with power δ^α and phases (ω_f,k^f), we get by applying the identities (7) and (9) that the leading order term $\mathbf{E}_0^f$ should satisfy

$$\mathbf{R}_0(\mathbf{E}_0^f)=\mathbf{D}_0(\mathbf{E}_0^f), \tag{10}$$

similarly as in the linear case. This is of course expected, since the weakness of the amplitude of the pulse (of order δ^α with $\alpha>0$) prevents nonlinear terms from coming into the leading part of the expansion with respect to δ.

As established in Ref. 13 there exist two positive solutions 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 Eq. (10). We define the dispersion relationship, the group velocity and the dispersion coefficient of the waves as follows:

$$k_m(\omega):=\frac{\omega n_m(\omega)}{c},\quad v_m(\omega):=\left(\frac{\partial k_m}{\partial\omega}\right)^{-1},\quad \sigma_m(\omega):=k_m\frac{\partial^2 k_m}{\partial\omega^2},\quad m=a,b, \tag{11}$$

and we denote by β_m the angle between the polarization vector $\mathbf{s}_m$ and the z axis

$$\cos^2(\beta_m(\omega))=s_{mx}^2(\omega)+s_{my}^2(\omega).$$

In order to fulfill condition (10), the set $(\omega_f,k^f,\mathbf{E}_0^f)$ must satisfy one of the three following alternatives:

(i) Either $k^f=k_a(\omega_f)$ and the components of $\mathbf{E}_0^f$ parallel to $\mathbf{s}_a(\omega_f)$ may be nonvanishing;
(ii) either $k^f=k_b(\omega_f)$ and the components of $\mathbf{E}_0^f$ parallel to $\mathbf{s}_b(\omega_f)$ may be nonvanishing;
(iii) or $k^f\notin\{k_a(\omega_f),k_b(\omega_f)\}$, and then necessarily $\mathbf{E}_0^f\equiv 0$.

Note that the third option simply means that modes which are not phase-matched cannot have an envelope of order δ^α.

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 were exhibited in the linear framework for a given 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 formulation

$$H=\left\{(\omega,k)\ \text{ such that }\ \exists j_1,\ldots,j_n\in\mathbb{Z},(\omega_i,k^i)\in H_{\mathcal{S}},\omega=\sum_{j=1}^{n} j_i\omega_i,\ \text{ and }\ k=\sum_{j=1}^{n} j_ik_i\right\}.$$

The rapid phases $(\omega,k)\in H$ for which $\omega\notin\Omega_{\mathcal{S}}$ correspond to the so-called harmonic modes. Since the pairs (ω_f,k^f) are algebraic sums of adapted rapid phases which originate from $H_{\mathcal{S}}$, and since the media we usually consider have a normal dispersion, there is only two types of phase matching (assuming that $n_a<n_b$):

(i) Type I: Both fundamental modes are of type a and the harmonic mode is of type b with $\omega_h=\omega_p+\omega_q$ and $k_a(\omega_q)+k_a(\omega_p)=k_b(\omega_h)$;

(ii) Type II: One of the fundamental modes is of type a and the other one of type b, while the harmonic mode is also of type b with $\omega_h=\omega_p+\omega_q$ and $k_a(\omega_p)+k_b(\omega_q)=k_b(\omega_h)$.

Only the rapid phases of H which satisfy one of the two phase matching conditions may possess a nonvanishing zeroth-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 $\delta^{2\alpha}$, that is to say $\mathbf{E}^f\sim\delta^\alpha\mathbf{E}_\alpha^f+O(\delta^{\alpha+1})$. Nevertheless it is necessary to take into account the harmonic modes that are not phase-matched so as to close the propagation equations. To complete this section we would like to add that the phase matching condition is only required to be fulfilled at order 1. A phase matching condition satisfied up to a term of order δ:$k_b(\omega_h)-k^p-k^q\sim O(\delta)$ or $k_b(\omega_h)-k^p-k^q\sim O(\delta)$, is a sufficient condition for an harmonic phase to possess an envelope with a nonvanishing leading order term $\mathbf{E}_0^h$. We shall encounter such situations in the forthcoming sections.

D. 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), then collecting the coefficients with power δ^α and high carrier frequency ω_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. Since $\alpha>0$ these conditions are the same as in the linear configuration, so the type m mode $(m=a,b)$ with carrier frequency ω_f should be at $z=0^+$

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

E. Poynting vector and diffraction operator in the linear framework

In this section we remember the reader with the main results of Ref. 13 in the case when the nonlinear polarization is 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 were thoroughly studied in Ref. 13. In the framework of frequency conversion, these configurations are not interesting since we aim at using the existence of two different dispersion relations for the modes to fulfill the phase matching conditions. The input wave break into the sum of modulations of high-frequency signals, which also divide into two modes which all propagate independently. For each high frequency ω and mode $m=a,b,$ the Poynting vector of the mode is collinear to

$$\mathbf{u}_m(\omega)=\left(\frac{s_{mx}s_{mz}}{s_{mx}^2+s_{my}^2},\frac{s_{my}s_{mz}}{s_{mx}^2s_{my}^2},-1\right)^T. \tag{12}$$

In the reference frame $(\delta t,\delta x,\delta y,\delta z)$ the slowly varying envelope E_m^ω of the mode satisfies the transport equation $\partial_Z E_m^\omega+\mathcal{T}_m(\omega)E_m^\omega=0$ where

$$\mathcal{T}_m(\omega)=-u_{mx}(\omega)\partial_X-u_{my}(\omega)\partial_Y+v_m(\omega)^{-1}\partial_T. \tag{13}$$

In the moving reference frame $(\delta(t-z/v_m),\delta(x+u_{mx}z),\delta(y+u_{my}z),\delta^2 z)$ the slowly varying envelope E_m^ω of the mode satisfies a Schrödinger-type equation with respect to the long scale variable $\zeta=\delta^2 z$

$$2ik_m(\omega)\partial_\zeta E_m^\omega+\mathcal{L}_m(\omega)E_m^\omega+\mathcal{K}_m(\omega)E_m^\omega=0, \tag{14}$$

where the diffraction operator $\mathcal{L}_m(\omega)$ is anisotropic (see Ref. 13 for the complete expressions of the $c_{m,\ldots}$)

$$\mathcal{L}_m(\omega)=c_{m,xx}(\omega)\partial_X^2+2c_{m,xy}(\omega)\partial_X\partial_Y+c_{m,yy}(\omega)\partial_Y^2, \tag{15}$$

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

$$\mathcal{K}_m(\omega)=-\sigma_m(\omega)\partial_T^2+2k_m(\omega)(\partial_\omega u_{mx})(\omega)\partial_T\partial_X+2k_m(\omega)(\partial_\omega u_{my})(\omega)\partial_T\partial_Y. \tag{16}$$

F. Second-order nonlinear polarization

The nonlinear susceptibility is the Fourier transform of the tensor $\chi^{(2)}$ defined by (3)

$$\hat{\chi}^{(2)}(\omega_1,\omega_2):=\int_0^\infty dt_1\int_0^\infty dt_2\chi^{(2)}(t_1,t_2)e^{i\omega_1 t_1+i\omega_2 t_2}.$$

Time integration starts from 0 to satisfy the causality property. We introduce the projector Θ_{ω_c,k^c} acting on fields of the form $\mathcal{A}=\frac{1}{2}\Sigma_{(\omega_f,k^f)\in H}\mathbf{A}_\delta^f e^{i(k^f z-\omega_f t)}+cc$ by

$$\Theta_{\omega_c,k^c}(\mathcal{A})=\mathbf{A}_\delta^c \text{ if } (\omega_c,k^c)\in H, \text{ and } 0 \text{ otherwise.}$$

The (ω_h,k^h) component of the nonlinear polarization in the crystallographic frame then reads as

$$\Theta_{\omega_h,k^h}(\mathcal{P}_{nl})=\frac{\epsilon_0}{2}\sum_{p,q,\omega_p+\omega_q=\omega_h,k^p+k^q=k^h}\hat{\chi}^{(2)}(\omega_p,\omega_q):(\mathbf{E}^p,\mathbf{E}^q).$$

For frequencies ω_p and ω_q and for types m_1 and m_2 in $\{a,b\}$ we denote by $\mathbf{s}_{m_1,m_2}(\omega_p,\omega_q)$ the vector

$$\mathbf{s}_{m_1,m_2}(\omega_p,\omega_q):=\hat{\chi}^{(2)}(\omega_p,\omega_q):(\mathbf{s}_{m_1}(\omega_p),\mathbf{s}_{m_2}(\omega_q)). \tag{17}$$

We list in the following the processes which can give rise to the generation of new frequencies:

(i) Type I conversion.
Let us denote $\omega_h = \omega_p + \omega_q$, $k^p = k_a(\omega_p)$, $k^q = k_a(\omega_q)$, and $k^h = k^p + k^q$. Then $\mathcal{E}^p + \mathcal{E}^q$ is of the form $\frac{1}{2}E^p \mathbf{s}_a(\omega_p) e^{i(k^p z - \omega_p t)} + \frac{1}{2}E^q \mathbf{s}_a(\omega_q) e^{i(k^q z - \omega_q t)} + cc$. In the (xyz)-reference frame the nonlinear polarization reads $\mathcal{P}_{nl,xyz} = U^{-1}\mathcal{P}_{nl,123}U$ and the (ω_h, k^h) component is

$$\Theta_{\omega_h,k^h}(\mathcal{P}_{nl,xyz}) = \epsilon_0 \mathbf{s}_{a,a}(\omega_p,\omega_q) E^p E^q. \tag{18}$$

(i) Type II conversion.
Let us denote $\omega_h = \omega_p + \omega_q$, $k^p = k_a(\omega_p)$, $k^q = k_b(\omega_q)$, and $k^h = k^p + k^q$. Then $\mathcal{E}^p + \mathcal{E}^q$ is of the form $\frac{1}{2}E^p \mathbf{s}_a(\omega_p) e^{i(k^p z - \omega_p t)} + \frac{1}{2}E^q \mathbf{s}_b(\omega_q) e^{i(k^q z - \omega_q t)}$. In the (xyz)-reference frame the nonlinear polarization reads $\mathcal{P}_{nl,xyz} = U^{-1}\mathcal{P}_{nl,123}U$ and the (ω_h, k^h) component is

$$\Theta_{\omega_h,k^h}(\mathcal{P}_{nl,xyz}) = \epsilon_0 \mathbf{s}_{a,b}(\omega_p,\omega_q) E^p E^q. \tag{19}$$

In the case of the class $\overline{4}2m$ which contains in particular potassium dihydrogen phosphate (KDP) crystal, the $\chi^{(2)}$-tensor has only six nonvanishing components which are equal to the coefficient $2d$: $\hat{\chi}^{(2)}_{123} = \hat{\chi}^{(2)}_{132} = \hat{\chi}^{(2)}_{213} = \hat{\chi}^{(2)}_{231} = \hat{\chi}^{(2)}_{312} = \hat{\chi}^{(2)}_{321} = 2d$. The vectors $\mathbf{s}_{o,o}$ and $\mathbf{s}_{o,e}$ then read as

$$\mathbf{s}_{o,o} = 2d \begin{pmatrix} \sin\theta \sin(2\phi) \\ 0 \\ -\cos\theta \sin(2\phi) \end{pmatrix}, \quad \mathbf{s}_{o,e} = 2d \begin{pmatrix} \cos(2\phi)\sin(\beta - 2\theta) \\ -\sin(2\phi)\sin(\beta-\theta) \\ \cos(2\phi)\cos(\beta-2\theta) \end{pmatrix}.$$

IV. TYPE I PHASE-MATCHING

We assume that the incoming pulse consists of two modes with carrier frequencies ω_p and ω_q which are linearly polarized along the $\mathbf{s}_a(\omega_p)$-axis and $\mathbf{s}_a(\omega_q)$-axis, respectively

$$\mathcal{S} = \tfrac{1}{2}\delta^\alpha (v^p(\delta x, \delta y, \delta t)\mathbf{s}_a(\omega_p) e^{-i\omega_p t} + v^q(\delta x, \delta y, \delta t)\mathbf{s}_a(\omega_q) e^{-i\omega_q t}) + cc. \tag{20}$$

The type I phase matching conditions are assumed to be satisfied for the sum $\omega_p + \omega_q = \omega_h$

$$k_a(\omega_p) + k_a(\omega_q) = k_b(\omega_h). \tag{21}$$

We also assume that the pair $(\omega_h, k_b(\omega_h))$ is the only adapted harmonic phase, that is to say the phase matching condition is not fulfilled for a subtraction or a sum between ω_p, ω_q, and ω_h different from $\omega_h = \omega_p + \omega_q$. As a consequence $H_{ac} = \{(\omega_p, k^p), (\omega_q, k^q), (\omega_h, k^h)\}$ where $k^p = k_a(\omega_p)$, $k^q = k_a(\omega_q)$, and $k^h = k_b(\omega_h)$. The adapted ansatz is accordingly

$$\mathcal{E} = \mathcal{E}^p + \mathcal{E}^q + \mathcal{E}^h + \mathcal{R}, \tag{22a}$$

$$\mathcal{E}^f = \frac{1}{2}\,\delta^\alpha \left(\sum_{j=0}^{\infty} \delta^j \mathbf{E}^f_j(\delta t, \delta x, \delta y, \delta z) \right) e^{i(k^f z - \omega_f t)} + cc, \tag{22b}$$

where $\mathcal{R}$ indicates a series of harmonic modes whose leading order coefficients $\mathbf{E}^f_0$ are vanishing. The parameter α will then play a crucial part since the order of magnitude of the input pulse imposes the distance scale at which the nonlinear effects become noticeable.

A. Geometric optics $\alpha = 1$

We denote $T = \delta t$, $X = \delta x$, $Y = \delta y$, $Z = \delta z$.

Proposition 1: If the source can be expanded as (20), then the fundamental modes are of type a for the carrier frequencies ω_p and ω_q while the harmonic mode at frequency ω_h is of type b. By denoting $E^p_0 = \mathbf{s}_a(\omega_p).\mathbf{E}^p_0$, $E^q_0 = \mathbf{s}_a(\omega_q).\mathbf{E}^q_0$, and $E^h_0 = \mathbf{s}_b(\omega_h).\mathbf{E}^h_0$ the projections of the modes onto their respective unit polarization vectors, the slowly varying envelopes E^f_0 satisfy the following coupled equations:

$$\partial_Z E_0^p + \mathcal{T}_a(\omega_p)(E_0^p) = \frac{i\omega_p d^{(I)}}{n_a(\omega_p)\cos^2(\beta_a(\omega_p))} E_0^{q*} E_0^h, \tag{23a}$$

$$\partial_Z E_0^q + \mathcal{T}_a(\omega_q)(E_0^q) = \frac{i\omega_q d^{(I)}}{n_a(\omega_q)\cos^2(\beta_a(\omega_q))} E_0^{p*} E_0^h, \tag{23b}$$

$$\partial_Z E_0^h + \mathcal{T}_b(\omega_h)(E_0^h) = \frac{i\omega_h d^{(I)}}{n_b(\omega_h)\cos^2(\beta_b(\omega_h))} E_0^p E_0^q, \tag{23c}$$

starting from $E_0^p(T,X,Y,Z=0) = (2/[1+n_a(\omega_p)])v^p(T,X,Y)$, $E_0^q(T,X,Y,Z=0) = (2/[1+n_a(\omega_q)])v^q(T,X,Y)$ *and* $E_0^h(T,X,Y,Z=0)=0$, *where the transport operator* $\mathcal{T}_m(\omega)$ *is given by* (13) *and*

$$d^{(I)} = \frac{\mathbf{s}_a(\omega_p).\mathbf{s}_{a,b}(\omega_q,\omega_h)}{2c}.$$

Proof: The strategy is formally the same as in linear configurations. We substitute the ansatz (22a) and (22b) with $\alpha=1$ into Eq. (1) and we collect the coefficients with the same power of δ and the same carrier frequency. At order δ we find the dispersion relation and phase matching conditions discussed in Sec. III C. At order δ^2, we project the equation onto the three axes. For the frequency ω_p (resp. $-\omega_p$), denoting $k^p = k_a(\omega_p)$

$$\mathbf{R}_0(\mathbf{E}_1^p) + \mathbf{R}_1(\mathbf{E}_0^p) = \mathbf{D}_0(\mathbf{E}_1^p) + \mathbf{D}_1(\mathbf{E}_0^p) + \mu_0\omega_p^2 \Theta_{\omega_p,k^p}(\mathcal{P}_{nl}(\mathcal{E}_0,\mathcal{E}_0)), \tag{24}$$

where we retain only the terms of order δ in $\mathcal{P}_{nl}$, which are the ones that give a contribution of the nonlinear polarization of order δ^2. Further in the nonlinear term $\mathcal{P}_{nl}(\mathcal{E}_0,\mathcal{E}_0)$ we only retain the coefficients with rapid phase $+(k^p z - \omega_p t)$. Only the frequencies ω_h and $-\omega_q$ can generate ω_p (resp. $-\omega_h$ and ω_q for $-\omega_p$). We, therefore, compute the sum of a type *a* wave at frequency $-\omega_q$ with a type *b* wave at frequency ω_h. By applying (19) we get in the reference frame (*x*,*y*,*z*):

$$\Theta_{\omega_p,k^p}(\mathcal{P}_{nl}(\mathcal{E}_0,\mathcal{E}_0)) = \epsilon_0 \mathbf{s}_{a,b}(\omega_q,\omega_h) E_0^{q*} E_0^h.$$

The projection of Eq. (24) onto $\mathbf{s}_a(\omega_p)$ then provides the compatibility condition which reads as Eq. (23a).

For the frequency ω_q, the situation is similar. We get Eq. (23b) with an expression of $d^{(I)}$ which is $d^{(I)'} = (\mathbf{s}_a(\omega_q).\mathbf{s}_{a,b}(\omega_p,\omega_h))/(2c)$, and using the symmetry properties of $\hat{\chi}^{(2)}$[14] it is easy to prove that $d^{(I)'} = d^{(I)}$.

For the frequency ω_h, denoting $k^h = k_b(\omega_h)$:

$$\mathbf{R}_0(\mathbf{E}_1^h) + \mathbf{R}_1(\mathbf{E}_0^h) = \mathbf{D}_0(\mathbf{E}_1^h) + \mathbf{D}_1(E_0^h) + \mu_0\omega_h^2 \Theta_{\omega_h,k^h}(\mathcal{P}_{nl}(\mathcal{E}_0,\mathcal{E}_0)), \tag{25}$$

where we retain only the terms of order δ in $\mathcal{P}_{nl}$, which are the ones that give a contribution of the nonlinear polarization of order δ^2. Further in the nonlinear term $\mathcal{P}_{nl}(\mathcal{E}_0,\mathcal{E}_0)$ we only retain the coefficients with rapid phase $+(k^h z - \omega_h t)$. Only the frequencies ω_p and ω_q can generate ω_h (resp. $-\omega_p$ and $-\omega_q$ for $-\omega_h$). We therefore compute the sum of a type *a* wave at frequency ω_p with a type *a* wave at frequency ω_q. By applying (18), we get in the reference frame (*x*,*y*,*z*)

$$\Theta_{\omega_h,k^h}(\mathcal{P}_{nl,xyz}) = \epsilon_0 \mathbf{s}_{a,a}(\omega_p,\omega_q) E_0^p E_0^q.$$

The projection of Eq. (25) onto $\mathbf{s}_b(\omega_h)$ then provides the compatibility condition which reads as Eq. (23c) with $d^{(I)''} = (\mathbf{s}_b(\omega_h).\mathbf{s}_{a,a}(\omega_p,\omega_q))/(2c)$, and using the symmetry properties of $\hat{\chi}^{(2)}$ it is easy to prove that $d^{(I)''} = d^{(I)}$. □

B. Approximate phase matching

In the above section we have considered a perfect phase matching condition. This condition is indeed very stringent and should be fulfilled at the leading order. Nevertheless it is barely possible in realistic experimental configurations to reach such a level of perfection. It is therefore relevant to address the case of a slight perturbation of the ideal case $\theta=\theta_{pm}$ where θ_{pm} is the angle which satisfies (21). We consider in this section that the phase matching condition is fulfilled up to a term of order δ and we set $\theta=\theta_{pm}+\delta\eta$. The corresponding propagation equations read

$$\partial_Z E_0^p+\mathcal{T}_a(\omega_p)E_0^p=\frac{i\omega_p d^{(I)}}{n_a(\omega_p)\cos^2(\beta_a(\omega_p))}E^{q*}\tilde{E}_0^h,$$

$$\partial_Z E_0^q+\mathcal{T}_a(\omega_q)E_0^q=\frac{i\omega_q d^{(I)}}{n_a(\omega_q)\cos^2(\beta_a(\omega_q))}E_0^{p*}E_0^h,$$

$$\partial_Z E_0^h+\mathcal{T}_b(\omega_h)E_0^h=\frac{i\omega_h d^{(I)}}{n_b(\omega_h)\cos^2(\beta_b(\omega_h))}E_0^pE_0^q+i\eta_k E_0^h,$$

where $\eta_k=\eta[\partial k_b(\omega_h)/\partial\theta]|_{\theta=\theta_{pm}}$. By setting $\tilde{E}_0^h=E_0^h e^{-i\eta_k Z}$, this system reduces:

$$\partial_Z E_0^p+\mathcal{T}_a(\omega_p)E_0^p=\frac{i\omega_p d^{(I)}}{n_a(\omega_p)\cos^2(\beta_a(\omega_p))}E_0^{q*}\tilde{E}_0^h e^{i\eta_k Z},$$

$$\partial_Z E_0^q+\mathcal{T}_a(\omega_q)E_0^q=\frac{i\omega_q d^{(I)}}{n_a(\omega_q)\cos^2(\beta_a(\omega_q))}E_0^{p*}\tilde{E}_0^h e^{i\eta_k Z},$$

$$\partial_Z \tilde{E}_0^h+\mathcal{T}_b(\omega_h)\tilde{E}_0^h=\frac{i\omega_h d^{(I)}}{n_b(\omega_h)\cos^2(\beta_b(\omega_h))}E_0^pE_0^q e^{-i\eta_k Z}.$$

It appears that it is necessary to add a phase $-\eta_k Z$ to the harmonic field so as to make the frequency conversion equations into a standard form. It shows that the rapid phase of the harmonic is imposed by the product of the phases of the fundamental modes: $\exp i((k^p+k^q)z-(\omega_p+\omega_q)t)$, which is different from the ''natural'' type b phase $\exp i(k_b(\omega_h)z-\omega_h t)$, with $\omega_h=\omega_p+\omega_q$.

C. Diffractive optics $\alpha=2$

In this configuration the nonlinear effects are weaker, of the order of $\delta^{2\alpha}=\delta^4$, so that they can show themselves only after a longer propagation distance z of the order of δ^{-2}. The technique is the same as for the derivations of the propagation equations in the linear framework. The final result is expressed in terms of the original variables x,y,z,t:

Proposition 2: Let us assume that the input field $\mathcal{S}$ consists of two modes with carrier frequencies ω_p and ω_q which are linearly polarized along the $\mathbf{s}_a(\omega_p)$-axis and $\mathbf{s}_a(\omega_q)$-axis, respectively. Then the fundamental modes are of type a and the harmonic mode is of type b ($\omega_h=\omega_p+\omega_q$). We denote $k^p=k_a(\omega_p)$, $k^q=k_a(\omega_q)$, $k^h=k_b(\omega_h)$, $\eta_k=k^h-k^p-k^q$, and $\tilde{\mathbf{E}}^h=\mathbf{E}^h e^{-i\eta_k z}$. We introduce the projections of the envelopes of the modes onto their respective unit polarization vectors: $E^p=\mathbf{s}_a(\omega_p).\mathbf{E}^p$, $E^q=\mathbf{s}_a(\omega_q).\mathbf{E}^q$, and $\tilde{E}^h=\mathbf{s}_b(\omega_h).\tilde{\mathbf{E}}^h$. The system which governs the propagation and conversion is:

$$\partial_z E^p+\mathcal{T}_a(\omega_p)E^p-\frac{i}{2k^p}\mathcal{L}_a(\omega_p)E^p-\frac{i}{2k^p}\mathcal{K}_a(\omega_p)E^p=\frac{i\omega_p d^{(I)}}{n_a(\omega_p)\cos^2(\beta_a(\omega_p))}E^{q*}\tilde{E}^h e^{i\eta_k z},$$

$$\partial_z E^q+\mathcal{T}_a(\omega_q)E^q-\frac{i}{2k^q}\mathcal{L}_a(\omega_q)E^q-\frac{i}{2k^q}\mathcal{K}_a(\omega_q)E^q=\frac{i\omega_q d^{(I)}}{n_a(\omega_q)\cos^2(\beta_a(\omega_q))}E^{p*}\tilde{E}^h e^{i\eta_k z},$$

$$\partial_z\tilde{E}^h+\mathcal{T}_b(\omega_h)\tilde{E}^h-\frac{i}{2k^h}\mathcal{L}_b(\omega_h)\tilde{E}^h-\frac{i}{2k^h}\mathcal{K}_b(\omega_h)\tilde{E}^h=\frac{i\omega_h d^{(I)}}{n_b(\omega_h)\cos^2(\beta_b(\omega_h))}E^pE^qe^{-i\eta_k z},$$

where the transport operator $\mathcal{T}_m$ and the diffraction-dispersion operators $\mathcal{L}_m$ and $\mathcal{K}_m$ are given by (13), (15), and (16), respectively (where small letters should be substituted for capital letters).

V. TYPE II PHASE-MATCHING

We assume that the source can be expanded as

$$\mathcal{S}=\frac{1}{2}\delta^2\begin{pmatrix} v_x(\delta x,\delta y,\delta t)e^{-i\omega_q t}\\ v_y(\delta x,\delta y,\delta t)e^{-i\omega_p t}\\ 0\end{pmatrix}+cc, \tag{26}$$

and that the type II phase matching condition is almost satisfied by the sum $\omega_p+\omega_q=\omega_h$:

$$k_b(\omega_h)-k_a(\omega_p)-k_b(\omega_q)=O(\delta).$$

We also assume that the pair $(\omega_h,k_b(\omega_h))$ is the only adapted harmonic phase, that is to say the phase matching condition is not fulfilled for a subtraction or a sum between ω_p, ω_q, and ω_h different from $\omega_h=\omega_p+\omega_q$, so that the suitable ansatz is (22a) and (22b) with $H_{ac}=\{(\omega_p,k^p),(\omega_q,k^q),(\omega_h,k^h)\}$ where $k^p=k_a(\omega_p)$, $k^q=k_b(\omega_q)$, and $k^h=k_b(\omega_h)$. The main result is obtained by using the very same techniques as in the previous sections so that we only state it in the original variables *x,y,z,t*. We denote the phase mismatch by $\eta_k:=k^h-k^p-k^q$.

Proposition 3. Let us assume that the input field $\mathcal{S}$ consists of two modes with carrier frequencies ω_p and ω_q which are linearly polarized along the y axis and x axis, respectively. Then the modes of the field $\mathcal{E}$ are ordinary for the fundamental ω_p and extraordinary for the fundamental ω_q and harmonic $\omega_h=\omega_p+\omega_q$. By introducing $\tilde{\mathbf{E}}^h=\mathbf{E}^he^{-i\eta_k z}$ and denoting $E^p=\mathbf{s}_a(\omega_p).\mathbf{E}^p$, $E^q=\mathbf{s}_b(\omega_q\mathbf{E}^q)$. and $\tilde{E}^h=\mathbf{s}_b(\omega_h).\tilde{\mathbf{E}}^h$ the projections of the envelopes of the modes onto their respective unit polarization vectors, the system which governs the propagation and conversion is:

$$\partial_zE^p+\mathcal{T}_a(\omega_p)E^p-\frac{i}{2k^p}\mathcal{L}_a(\omega_p)E^p-\frac{i}{2k^p}\mathcal{K}_a(\omega_p)E^p=\frac{i\omega_p d^{(II)}}{n_a(\omega_p)\cos^2(\beta_a(\omega_p))}E^{q*}\tilde{E}^he^{i\eta_k z},$$

$$\partial_zE^q+\mathcal{T}_b(\omega_q)E^q-\frac{i}{2k^q}\mathcal{L}_b(\omega_q)E^q-\frac{i}{2k^q}\mathcal{K}_b(\omega_q)E^q=\frac{i\omega_q d^{(II)}}{n_b(\omega_q)\cos^2(\beta_b(\omega_q))}E^{p*}\tilde{E}^he^{i\eta_k z},$$

$$\partial_z\tilde{E}^h+\mathcal{T}_b(\omega_h)\tilde{E}^h-\frac{i}{2k^h}\mathcal{L}_b(\omega_h)\tilde{E}^h-\frac{i}{2k^h}\mathcal{K}_b(\omega_h)\tilde{E}^h=\frac{i\omega_h d^{(II)}}{n_b(\omega_h)\cos^2(\beta_b(\omega_h))}E^pE^qe^{-i\eta_k z},$$

where

$$d^{(II)}=\frac{\mathbf{s}_a(\omega_p).\mathbf{s}_{b,b}(\omega_q,\omega_h)}{2c}.$$

VI. PROPAGATION FAR FROM PHASE-MATCHING

In the two previous sections we have examined the two cases corresponding to phase-matching for sum-frequency generation. In this section we consider the general situation where the phase-matching condition is not fulfilled so as to derive the propagation equation of the fundamental wave, and also some information for the different harmonic waves. Such a work has been performed by Leblond[15] who considered the propagation of a pulse along the principal axis of a uniaxial crystal. In this section we consider the general case of biaxial crystals in the configuration when the two eigenindices are different from each other. We assume a source of the form

$$\mathcal{S}=\tfrac{1}{2}\delta v(\delta x,\delta y,\delta t)(s_{ax}(\omega),s_{ay}(\omega),0)^T e^{-i\omega t}+cc, \tag{27}$$

and we shall see that the suitable ansatz for the scales corresponding to diffractive optics is:

$$\mathcal{E}=\mathcal{E}^{0\omega}+\mathcal{E}^{1\omega}+\mathcal{E}^{2\omega}+\mathcal{R}, \tag{28a}$$

$$\mathcal{E}^{l\omega}=\frac{1}{2}\delta\sum_{j=0}^{\infty}\delta^j \mathbf{E}_j^{l\omega}(\delta(t-z/v_a),\delta(x+u_{ax}z),\delta(y+u_{ay}z),\delta^2 z)e^{il(k_a(\omega)z-\omega t)}+cc, \tag{28b}$$

where $\mathcal{R}$ is a sum of harmonic waves whose leading order term is of order δ^3 or smaller, and v_a, $\mathbf{u}_a$ are shorthands for $v_a(\omega)$, $\mathbf{u}_a(\omega)$, respectively.

Proposition 4: If we assume that the input field $\mathcal{S}$ consists of one mode with carrier frequency ω which is linearly polarized along the $\mathbf{s}_a(\omega)$-axis, then the slowly varying envelope of the field $\mathcal{E}$ is of type a and $E_0=\mathbf{s}_a(\omega).\mathbf{E}_0^{1\omega}$ satisfies the nonlinear Schrödinger equation

$$2ik_a\partial_\zeta E_0+\mathcal{L}_a(\omega)E_0+\mathcal{K}_a(\omega)E_0+P_3(E_0)=0, \tag{29}$$

starting from $E_0(T,X,Y,\zeta=0)=(2/[1+n_a(\omega)])v(T,X,Y)$, where $P_3(E_0)=(\gamma_1+\gamma_2)|E_0|^2E_0+\mathbf{s}_a.\hat{\chi}^{(2)}(0,\omega)(\mathbf{E}_1^{0\omega *},\mathbf{E}_0^{1\omega})$,

$$\gamma_1=\tfrac{3}{4}\mathbf{s}_a(\omega).\hat{\chi}^{(3)}(\omega,\omega,-\omega):(\mathbf{s}_a(\omega),\mathbf{s}_a(\omega),\mathbf{s}_a(\omega)),$$

$$\gamma_2=\tfrac{1}{2}\mathbf{s}_a(\omega).\hat{\chi}^{(2)}(2\omega,-\omega):((n_a^2(\omega)J-\chi(2\omega))^{-1}\mathbf{s}_{a,a}(\omega,\omega),\mathbf{s}_a(\omega)),$$

where J is the 3×3 matrix whose entries are vanishing but $J_{11}=J_{22}=1$. The second harmonic is of order δ^2 and its leading order term $\mathbf{E}_1^{2\omega}$ is given by (30). The zero harmonic is of order δ^2 and its leading order term $\mathbf{E}_1^{0\omega}$ is given by (32). All other harmonic are of order smaller than δ^3.

Note that far from phase matching, the second harmonic does not propagate with its natural phase velocity and group velocity, but with those of the fundamental. But it is smaller by an order of magnitude. The same holds true for the zero-harmonic term. By ''zero-harmonic'' we mean an electromagnetic wave whose wavelength is of the order of δ^{-1}.

Proof:

(i) Computation of the leading order term at 2ω.
Collecting the terms of order δ^2 at frequency 2ω provides an explicit representation for $\mathbf{E}_1^{2\omega}$:

$$\mathbf{E}_1^{2\omega}=\tfrac{1}{2}(n_a^2(\omega)J-\chi(2\omega))^{-1}\hat{\chi}^{(2)}(\omega,\omega):(\mathbf{E}_0^{1\omega},\mathbf{E}_0^{1\omega}). \tag{30}$$

Note that the matrix $n_a^2(\omega)J-\chi(2\omega)$ is invertible since we assume that there is no phase-matching for second harmonic generation.

(ii) Computation of the leading order term at 0ω.
Collecting the terms of order δ^4 at frequency 0ω yields:

$$\mathbf{R}_2(\mathbf{E}_1^{0\omega})=\mathbf{D}_2(\mathbf{E}_1^{0\omega})-c^{-2}\partial_T^2\mathbf{P}_2,$$

where $\mathbf{P}_2$ is given by

$$\mathbf{P}_2=\hat{\chi}^{(2)}(\omega,-\omega)(\mathbf{E}_0^{1\omega},\mathbf{E}_0^{1\omega *}).$$

Note that $\mathbf{D}_2(\mathbf{E}_1^{0\omega})=-c^{-2}\chi(0\omega)\partial_T^2\mathbf{E}_1^{0\omega}$. The computation of $\mathbf{E}_1^{0\omega}$ is formally identical as for the second-harmonic wave, that is to say the zero-harmonic wave is obtained by applying an inversion operator [here $(\mathbf{R}_2-\mathbf{D}_2)^{-1}$] to a functional of the leading-order term of the fundamental wave (here $-c^{-2}\partial_T^2\mathbf{P}_2$). But the inversion is a little more elaborate, since it requires to apply the Green function of a linear nondispersive Maxwell equation. We give in what follows an explicit formulation of this inversion. First denote by **Rot** the standard ''**rot**'' operator operating on the macroscopic variables (X,Y,Z). Consider the problem of finding the solution **E** of the following Maxwell equation with a source:

$$\mathbf{Rot}\ \mathbf{Rot}\ \mathbf{E} = -c^{-2}\chi(0\omega)\partial_T^2\mathbf{E} - c^{-2}\partial_T^2\mathbf{P},$$

where $\mathbf{P}(T,X,Y,Z)$ is the polarization induced by the source. Taking the Fourier transform with respect to time and space

$$(|\mathbf{K}|^2 I_d - \mathbf{K}\otimes\mathbf{K} - \nu^2 c^{-2}\chi(0\omega))\hat{\mathbf{E}}(\nu,\mathbf{K}) = \nu^2 c^{-2}\hat{\mathbf{P}}(\nu,\mathbf{K}), \tag{31}$$

where ν is the frequency, $\mathbf{K}$ the wave number, and $\mathbf{S}\otimes\mathbf{U}$ is the matrix whose entries are $S_i U_j$. We denote by N_a and N_b the two solutions of the Fresnel equation associated with the tensor $\chi(0\omega)$:

$$\det(N^2 I_d - N^2|\mathbf{K}|^2\mathbf{K}\otimes\mathbf{K} - \chi(0\omega)) = 0,$$

and by $\mathbf{S}_a$ and $\mathbf{S}_b$ the corresponding unit eigenvectors. The Green function $\hat{g}(\nu,\mathbf{K})$ corresponding to Eq. (31) is defined by the equation

$$\hat{\mathbf{E}}(\nu,\mathbf{K}) = \hat{g}(\nu,\mathbf{K})\hat{\mathbf{P}}(\nu,\mathbf{K}).$$

It was shown by Lax and Nelson that the Green function can be written in the form[16]

$$\hat{g}(\nu,\mathbf{K}) = \frac{\nu^2}{c^2}\left(\frac{e_a}{|\mathbf{K}|^2/N_a^2 - \nu^2/c^2 - io} + \frac{e_b}{|\mathbf{K}|^2/N_b^2 - \nu^2/c^2 - io}\right) - \frac{\mathbf{K}\otimes\mathbf{K}}{\mathbf{K}^T\chi(0\omega)\mathbf{K}},$$

$$e_m = \frac{\mathbf{S}_m\otimes\mathbf{S}_m}{\mathbf{S}_m^T\chi(0\omega)\mathbf{S}_m}.$$

If $\nu<0$, then the term $-io$ should be replaced by $+io$. We then introduce the auxiliary function $\hat{G}$ which is the projection of $\hat{g}$ onto the characteristic equation satisfied by $\mathbf{P}_2$

$$\hat{G}(\nu,K_x,K_y) = \hat{g}(\nu,K_x,K_y,K_x u_{ax}(\omega) + K_y u_{ay}(\omega) - \nu/v_a(\omega)),$$

$$G(T,X,Y) = \frac{1}{(2\pi)^3}\int \hat{G}(\nu,K_x,K_y)e^{i(K_xX+K_yY-\nu T)}dXdYdT.$$

We finally define the convolution operator Ψ which associates to any pair of vector-valued functions $\mathbf{A}_1(T,X,Y)$ and $\mathbf{A}_2(T,X,Y)$ the vector-valued function

$$\Psi*(\mathbf{A}_1,\mathbf{A}_2)(T,X,Y) := \int G(T-s,X-u,Y-v)[\hat{\chi}^{(2)}(\omega,-\omega):(\mathbf{A}_1,\mathbf{A}_2)(s,u,v)]dsdudv.$$

The leading order term $\mathbf{E}_1^{0\omega}(T,X,Y,\zeta)$ of the zero-harmonic wave can then be expressed for every ζ as the application of the Ψ-operator to the pair $(\mathbf{E}_0^{1\omega}(.,.,.,\zeta),\mathbf{E}_0^{1\omega *}(.,.,.,\zeta))$:

$$\mathbf{E}_1^{0\omega}(T,X,Y,\zeta) = \Psi*(\mathbf{E}_0^{1\omega}(.,.,.,\zeta),\mathbf{E}_0^{1\omega *}(.,.,.,\zeta))(T,X,Y). \tag{32}$$

(iii) Equation for the corrective term at ω.
Collecting the terms of order δ^2 at frequency ω we get an explicit form for the first corrective term $\mathbf{E}_1^{1\omega}$ of the fundamental wave. No nonlinear term is coming to at this order, so the expression is identical to the linear framework (see Ref. 13).

(iv) Equation for the leading order term at ω.
Collecting the terms of order δ^3 at frequency ω we get

$$\mathbf{R}_2(\mathbf{E}_0^{1\omega}) + \mathbf{R}_1(\mathbf{E}_1^{1\omega}) + \mathbf{R}_0(\mathbf{E}_2^{1\omega}) = \mathbf{D}_2(\mathbf{E}_0^{1\omega}) + \mathbf{D}_1(\mathbf{E}_1^{1\omega}) + \mathbf{D}_0(\mathbf{E}_2^{1\omega}) - c^{-2}\mathbf{P}_3,$$

where the contribution of the nonlinear polarization is:

$$\mathbf{P}_3 = (\tfrac{3}{4})\hat{\chi}^{(3)}(\omega,\omega,-\omega):(\mathbf{E}_0^{1\omega},\mathbf{E}_0^{1\omega},\mathbf{E}_0^{1\omega *}) + \hat{\chi}^{(2)}(2\omega,-\omega):(\mathbf{E}_1^{2\omega},\mathbf{E}_0^{1\omega *})$$
$$+\hat{\chi}^{(2)}(0\omega,\omega):(\mathbf{E}_1^{0\omega},\mathbf{E}_0^{1\omega}).$$

Projecting onto $\mathbf{s}_a(\omega)$ we get

$$\mathbf{s}_a(\omega).\mathbf{R}_2(\mathbf{E}_0^{1\omega})+\mathbf{s}_a(\omega).\mathbf{R}_1(\mathbf{E}_1^{1\omega})=\mathbf{s}_a(\omega).\mathbf{D}_2(\mathbf{E}_0^{1\omega})+\mathbf{s}_a(\omega).\mathbf{D}_1(\mathbf{E}_1^{1\omega})-c^{-2}\mathbf{s}_a(\omega).\mathbf{P}_3.$$

Substituting the expression of $\mathbf{E}_1^{1\omega}$ establishes the result.

Note that the long-scale variable ζ plays the role of a parameter in the expression (32) of $\mathbf{E}_1^{0\omega}$. Consequently Eq. (29) reads as a simple first-order evolution equation with respect to the ζ-variable for the envelope E_0. This provides a simple numerical scheme to compute $E_0(.,.,.,\zeta+\Delta\zeta)$ from $E_0(.,.,.,\zeta)$.

VII. SPATIAL SOLITON PROPAGATION IN BIAXIAL CRYSTALS

We examine in this section the propagation in biaxial crystals of the modulation of a high-frequency signal with frequency ω in the particular configuration $\theta=\theta_r(\omega)$ and $\phi=0$ or π where

$$\sin^2\theta_r(\omega)=\frac{1-\chi_2(\omega)/\chi_1(\omega)}{1-\chi_3(\omega)/\chi_1(\omega)}.$$

Computing all relevant quantities according to the general formulas we have found in particular that the type a eigenindex and unit polarization eigenvector are $n_a^2(\omega)=\chi_2(\omega)$ and $\mathbf{s}_a(\omega)=(0,1,0)^T$, respectively (for the type b we refer to Ref. 13). The diffraction coefficients for the a-mode are $c_{a,xx}(\omega)=1$, $c_{a,xy}(\omega)=0$, and $c_{a,yy}(\omega)=0$, while the dispersion operator reads $\mathcal{K}_a(\omega)=-\sigma_a(\omega)\partial_T^2$. The striking point is that the diffraction operator for the type a wave is degenerate, in the sense that there is no diffraction in the y-direction. Let us assume that the carrier frequency ω of the input pulse is such that the phase matching condition for the second-harmonic generation is not fulfilled. For the sake of simplicity we first restrict ourselves to one of the three following classes:[17]

(1) triclinic class with point group $\overline{1}$, such as Mica or Al_2SiO_5;
(2) monoclinic class with point group $2/m$, such as $AgAuTe_4$ or $PbSiO_3$;
(3) orthorhombic 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 of the $\chi^{(2)}$-cascaded terms and to deal with a simple $\chi^{(3)}$-component which then reads as a simple Kerr effect. The result is the following:

Proposition 5. In cases 1, 2, 3, if we assume that the input field $\mathcal{S}$ consists of one mode with carrier frequency ω which is linearly polarized along the y axis, then the slowly varying envelope of the field $\mathcal{E}$ is polarized along the y axis and $E_0=(0,1,0).\mathbf{E}_0^{1\omega}$ satisfies the nonlinear Schrödinger equation

$$2ik_a\partial_\zeta E_0+\partial_X^2E_0-\sigma_a\partial_T^2E_0+\gamma|E_0|^2E_0=0, \tag{33}$$

starting from $E_0(T,X,Y,\zeta=0)=(2/[1+n_a(\omega)])v(T,X,Y)$, where $\gamma=(\frac{3}{4})\hat{\chi}^{(3)}_{2222}(-\omega,\omega,\omega)$.

The removal of the time variable is involved by the assumption that there is no modulation of the input pulse at the time scale δ^{-1}, which is typically of the order of the picosecond, but only at scale δ^{-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

$$2ik_a\partial_\zeta E_0+\partial_X^2E_0+\gamma|E_0|^2E_0=0. \tag{34}$$

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 $z^{-1/2}$. The incident pulse must at least fulfill a well-known power criterion so that a soliton can be generated[18]

$$\int_{-\infty}^{\infty} |E_0| dx \geqslant 1.279\gamma^{-1/2},$$

which simply means that the incident pulse should be sufficient focused so that its power $\int |E_0|^2 dx$ be concentrated on a small segment. Nevertheless one should still remain in the domain where Eq. (33) holds true, which is basically the paraxial approximation.

In case of biaxial crystals with nonvanishing $\chi^{(2)}$-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. We aim in the following proposition at generalizing Proposition 5 to any biaxial crystal.

Proposition 6. For any biaxial crystal in the configuration $\theta=\theta_r$ and $\phi=0$, if we assume that the input field $\mathcal{S}$ consists of one mode with carrier frequencies ω which is linearly polarized along the y axis, then the slowly varying envelope of the field $\mathcal{E}$ is polarized along the y axis and $E_0=(0,1,0).\mathbf{E}_0$ satisfies the nonlinear Schrödinger equation

$$2ik_a\partial_\zeta E_0+\partial_X^2E_0-\sigma_a\partial_T^2E_0+P_3(E_0)=0, \tag{35}$$

where $P_3(E_0)=(\gamma_1+\gamma_2)|E_0|^2E_0+\Sigma_{j,l=1}^3\gamma_{3_{jl}}\Phi_{jl}(|E_0|^2(.,.,.,\zeta))E_0$, with

$$\gamma_1=\tfrac{3}{4}\hat{\chi}^{(3)}_{2222}(\omega,\omega,-\omega),$$

$$\gamma_2=\frac{1}{2}\sum_{j=1}^{3}\hat{\chi}^{(2)}_{2j2}(2\omega,-\omega)[(n_a^2(\omega)J-\chi(2\omega))^{-1}\chi^{(2)}_{.22}(\omega,\omega)]_j,$$

$$\gamma_{3_{jl}}=\hat{\chi}^{(2)}_{2j2}(0,\omega)\hat{\chi}^{(2)}_{l22}(\omega,-\omega),$$

$$\Phi_{jl}(I(.,.,.))(T,X,Y)=\sum_{j',l'=1}^{3}U_{jj'}U_{ll'}\int G_{j'l'}(T-s,X-u,Y-v)I(s,u,v)dsdudv.$$

G is the Green function whose Fourier transform is $\hat{G}(\nu,K_x,K_y)=\hat{g}(\nu,K_x,K_y,-\nu/v_a(\omega))$ with:

$$\hat{g}(\nu,\mathbf{K})=\frac{\nu^2}{c^2}\left(\frac{e_a}{|\mathbf{K}|^2/n_a(0\omega)^2-\nu^2/c^2-io}+\frac{e_b}{|\mathbf{K}|^2/n_b^2(0\omega)-\nu^2/c^2-io}\right)-\frac{\mathbf{K}\otimes\mathbf{K}}{\mathbf{K}^T\chi(0\omega)\mathbf{K}},$$

$$e_m=\frac{\mathbf{s}_m(0\omega)\otimes\mathbf{s}_m(0\omega)}{(\mathbf{s}_m(0\omega)^T)\chi(0\omega)\mathbf{s}_m(0\omega)},$$

where $n_m(0\omega)$ and $\mathbf{s}_m(0\omega)$ are the eigenindices and unit eigenvectors of the Fresnel equation corresponding to the tensor $\chi(0\omega)$ at angles $\theta_r(\omega)$, $\phi=0$

$$(n^2I_d-n^2|\mathbf{K}|^{-2}\mathbf{K}\otimes\mathbf{K}-\chi(0\omega,\theta_r(\omega),0))\mathbf{s}=0.$$

All terms in P_3 are proportional to $|E_0|^2E_0$ or a product of three terms proportional to E_0. Note that the only coefficient of the $\chi^{(3)}$-tensor which plays a role is $\hat{\chi}^{(3)}_{2222}(\omega,\omega,-\omega)$. The coefficients of the $\chi^{(2)}$-tensor which play a role are the ones with at least two indices equal to 2. In case of orthorhombic class with point group 222, the only non-vanishing coefficients of the $\chi^{(2)}$-tensor are the ones with three different indices. Consequently all components of P_3 vanish but $\gamma_1|E_0|^2E_0$ so that we get back the result of Proposition 5.

VIII. SPATIAL SPECTRUM OF THE SECOND HARMONIC PULSE

In this section we aim at giving the explanation of a recent experimental observation. The framework is the following. In the context of Inertial Confinement Fusion, many high-power laser beams are focused onto a spherical target composed of a mixture of deuterium–tritium so as to compress it and to obtain density and pressure conditions which involve thermonuclear burning. The laser energy production is based on the amplification of an infrared pulse in glass amplifiers, which are the only ones capable to deliver an energy of the order of 1 to 2 megajoules. Nevertheless it is necessary to frequency convert the pulse in the ultraviolet (UV) domain so as to optimize the plasma-laser interaction between the laser beams and the inertial confinement-fusion (ICF) target. Thus the frequency tripling performance conditions the feasibility of the project. Two successive KDP crystals, which can be produced in large dimensions, are used for the frequency doubling and summing operations. In order to get a high tripling rate, it is necessary to adjust the positions of the KDP crystals in the laser chain with very high accuracy, since a precision of the order of 15 μrad is required. The method consists in focusing a fundamental beam and to detect the main output angle of the second harmonic pulse (''test'' configuration), which should correspond to the optimal frequency conversion angle. However it appears that the direction of the frequency converted pulse of a fundamental Gaussian pulse depends on the distance between the waist of the fundamental pulse and the crystal. The departures for different distances far exceed the high precision level required for reaching the expected conversion performance for applications to ICF. It is, therefore, necessary to give a precise account of this unexpected phenomenon.

We assume that the fundamental pulse has Gaussian shape in the waist plane $z=0$. We denote by z_0 the distance from the waist plane to the crystal plate, and by z_c the thickness of the plate. If w_0 is the beam radius in the waist plane, A_0 is its maximal amplitude, and $\mathbf{e}$ is its unit polarization vector, then in the plane just before the plate the input field writes

$$\hat{\mathbf{E}}(z=z_0^-,k_x,k_y)=A_0\pi w_0^2\exp\left(-\frac{(k_x^2+k_y^2)w_0^2}{4}-i\frac{(k_x^2+k_y^2)z_0}{2k}\right)\mathbf{e},$$

where k is the free wave number and we have performed a Fourier transform with respect to the transverse coordinates $(x,y)\mapsto(k_x,k_y)$. By continuity of the tangential components of the electric field, the field just inside the plate is the sum of an ordinary wave and an extraordinary wave

$$\hat{E}_{o,\omega}(z=z_0^+,k_x,k_y)=A_0\pi w_0^2 e_y\exp\left(-\frac{(k_x^2+k_y^2)w_0^2}{4}-i\frac{(k_x^2+k_y^2)n_o(\omega)z_0}{2k_o(\omega)}\right),$$

$$\hat{E}_{e,\omega}(z=z_0^+,k_x,k_y)=\frac{A_0\pi w_0^2 e_x}{\cos(\beta(\omega))}\exp\left(-\frac{(k_x^2+k_y^2)w_0^2}{4}-i\frac{(k_x^2+k_y^2)n_e(\omega)z_0}{2k_e(\omega)}\right).$$

For the type I configuration we consider the case $e_x=0$, $e_y=1$. For the type II configuration we choose an equiphotonic repartition $e_x=1/\sqrt{2}$, $e_y=1/\sqrt{2}$.

A. Type I conversion

Applying Proposition 2 the system which governs the second harmonic generation in the type I configuration is:

$$\partial_z E_{o,\omega}-\frac{i}{2k_o(\omega)}\partial_x^2 E_{o,\omega}-\frac{i}{2k_o(\omega)}\partial_y^2 E_{o,\omega}=\frac{i2\omega d^{(I)}}{n_o(\omega)}E_{o,\omega}^*E_{e,2\omega}e^{i\eta_k z},$$

$$\partial_z E_{e,2\omega}-\eta\partial_x E_{e,2\omega}-\frac{ic_x(2\omega)}{2k_e(2\omega)}\partial_x^2 E_{e,2\omega}-\frac{ic_y(2\omega)}{2k_e(2\omega)}\partial_y^2 E_{e,2\omega}=\frac{i2\omega d^{(I)}}{n_e(2\omega)\cos^2\beta(2\omega)}E_{o,\omega}^2 e^{-i\eta_k z},$$

where $\eta_k=k_e(2\omega)-2k_o(\omega)$, $\eta=\tan\beta(2\omega)$, and

$$c_x(2\omega)=\frac{\chi_o(2\omega)\chi_e(2\omega)}{(\cos^2(\theta)\chi_e(2\omega)+\sin^2(\theta)\chi_o(2\omega))^2},\quad c_y(2\omega)=\frac{\chi_o(2\omega)}{\cos^2(\theta)\chi_e(2\omega)+\sin^2(\theta)\chi_o(2\omega)}.$$

We assume that the frequency conversion rate is low. Taking the Fourier transform with respect to the spatial transverse coordinates $(x,y)\mapsto(k_x,k_y)$:

$$\partial_z\hat{E}_{o,\omega}+\frac{ik_x^2}{2k_o(\omega)}\hat{E}_{o,\omega}+\frac{ik_y^2}{2k_o(\omega)}\hat{E}_{o,\omega}=0, \tag{36a}$$

$$\partial_z\hat{E}_{e,2\omega}+ik_x\eta\hat{E}_{e,2\omega}+\frac{ik_x^2c_x(2\omega)}{2k_e(2\omega)}\hat{E}_{e,2\omega}+\frac{ik_y^2c_y(2\omega)}{2k_e(2\omega)}\hat{E}_{e,2\omega}=id^I_{e,2\omega}\hat{E}_{o,\omega}*\hat{E}_{o,\omega}e^{i(2k_o-k_e)z}, \tag{36b}$$

where $*$ stands for the convolution operation. It is easy to find the explicit form of $\hat{E}_{o,\omega}$ from Eq. (36a) by a simple exponentiation, and convoluting this expression with itself

$$\hat{E}_{o,\omega}*\hat{E}_{o,\omega}(z_0+z,k_x,k_y)=\frac{A_0\pi w_0^2}{\frac{w_0^2}{2}+i\frac{n_o(\omega)z_0+z}{k_o(\omega)}}\exp-\frac{k_x^2+k_y^2}{2}\left(\frac{w_0^2}{4}+i\frac{n_o(\omega)z_0+z}{2k_o(\omega)}\right).$$

In order to compute $|\hat{E}_{e,2\omega}|$ we set:

$$\hat{E}_{e,2\omega}=\hat{E}_{e,2\omega}\exp iz\left(k_x\eta+\frac{k_x^2c_x(2\omega)}{2k_e(2\omega)}+\frac{k_y^2c_y(2\omega)}{2k_e(2\omega)}\right),$$

whose modulus is equal to the modulus of $\hat{E}_{e,2\omega}$ and which satisfies

$$\partial_z\overline{E}_{e,2\omega}=id^{II}_{e,2\omega}\hat{E}_{o,\omega}*\hat{E}_{o,\omega}\exp iz\left(k_x\eta+\frac{k_x^2c_x(2\omega)}{2k_e(2\omega)}+\frac{k_y^2c_y(2\omega)}{2k_e(2\omega)}+2k_o(\omega)-k_e(2\omega)\right).$$

The right-hand side is known, so by a simple exponentiation we get that, up to a multiplicative constant

$$|\hat{E}_{e,2\omega}|^2(z_0+z_c,k_x,k_y)=e^{-\gamma(k_x,k_y)k_ow_0^2}|E_i(-\gamma(k_x,k_y)(z_1+iz_c))-E_i(-\gamma(k_x,k_y)z_1)|^2, \tag{37}$$

where E_i is the integral exponential function: $E_i(x):=\int_1^\infty[\exp(-xt)/t]dt$ and

$$z_1=\frac{k_o(\omega)w_0^2}{2}+iz_0n_0(\omega),$$

$$\gamma(k_x,k_y)=k_x\eta+k_x^2\left(\frac{c_x(2\omega)}{2k_e(2\omega)}-\frac{1}{4k_o(\omega)}\right)+k_y^2\left(\frac{c_y(2\omega)}{2k_e(2\omega)}-\frac{1}{4k_o(\omega)}\right)+2k_o(\omega)-k_e(2\omega).$$

A study of the function (37) proves that the locations of the minima of the spectral intensity (which are experimentally detectable with high precision) do not depend on the waist distance z_0.

B. Type II conversion

We still assume that the frequency conversion rate is low. Taking the Fourier transform for the spatial transverse coordinates and applying Proposition 3, the system which governs the second harmonic generation in the type II configuration is:

$$\partial_z \hat{E}_{o,\omega} + \frac{ik_x^2}{2k_o(\omega)} \hat{E}_{o,\omega} + \frac{ik_y^2}{2k_o(\omega)} \hat{E}_{o,\omega} = 0, \tag{38a}$$

$$\partial_z \hat{E}_{e,\omega} + ik_x \eta_1 \hat{E}_{e,\omega} + \frac{ik_x^2 c_x(\omega)}{2k_e(\omega)} \hat{E}_{e,\omega} + \frac{ik_y^2 c_y(\omega)}{2k_e(\omega)} \hat{E}_{e,\omega} = 0, \tag{38b}$$

$$\partial_z \hat{E}_{e,2\omega} + ik_x \eta_2 \hat{E}_{e,2\omega} + \frac{ik_x^2 c_x(2\omega)}{2k_e(2\omega)} \hat{E}_{e,2\omega} + \frac{ik_y^2 c_y(2\omega)}{2k_e(2\omega)} \hat{E}_{e,2\omega}$$
$$= \frac{2i\omega d^{(II)}}{n_e(2\omega)\cos^2 \beta(2\omega)} \hat{E}_{o,\omega} * \hat{E}_{e,\omega} e^{-i\eta_k z}, \tag{38c}$$

where $\eta_k = k_e(2\omega) - k_e(\omega) - k_o(\omega)$, $\eta_1 = \tan\beta(\omega)$, and $\eta_2 = \tan\beta(2\omega)$. The calculations are identical to the ones performed in the type I configuration. One first compute the closed-form expressions of the fields $\hat{E}_{o,\omega}$ and $\hat{E}_{e,\omega}$ from Eqs. (38a) and (38b). These expressions are then substituted into the right-hand side of Eq. (38c) which can then be solved. We have found that, up to a multiplicative constant

$$|\hat{E}_{e,2\omega}|(2k\theta_x, 2k\theta_y) = \Bigg| \int_0^{z_c} \frac{1}{(\bar{\alpha} + iC_x z)^{1/2}} \exp k \frac{A_x(\theta_x) z^2 + i\bar{\alpha} B_x(\theta_x) z - \bar{\alpha}^2 \theta_x^2}{\bar{\alpha} + iC_x z}$$
$$\times \frac{1}{(\bar{\alpha} + iC_y z)^{1/2}} \exp k \frac{A_y(\theta_y) z^2 + i\bar{\alpha} B_y(\theta_y) z - \bar{\alpha}^2 \theta_y^2}{\bar{\alpha} + iC_y z} \times \exp(-i\eta_k z) dz \Bigg|, \tag{39}$$

where $\bar{\alpha} = z_r + iz_0$, $z_r = kw_0^2/2$, $k = 2\pi/\lambda = \omega/c$, and

$$A_x(\theta_x) = \theta_x^2 \left(\frac{c_x(\omega)}{n_o(\omega) n_e(\omega)} - \frac{c_x(2\omega)}{n_e(2\omega)} C_x \right) + \theta_x \left(\frac{\eta_1}{n_o(\omega)} - 2\eta_2 C_x \right) - \frac{\eta_1^2}{4},$$

$$B_x(\theta_x) = \theta_x^2 \left(\frac{c_x(2\omega)}{n_e(2\omega)} - 2C_x \right) + \theta_x (2\eta_2 - \eta_1),$$

$$C_x = \frac{1}{2} \left(\frac{1}{n_o(\omega)} + \frac{c_x(\omega)}{n_e(\omega)} \right),$$

$$A_y(\theta_y) = \theta_y^2 \left(\frac{c_y(\omega)}{n_o(\omega) n_e(\omega)} - \frac{c_y(2\omega)}{n_e(2\omega)} C_y \right),$$

$$B_y(\theta_x) = \theta_y^2 \left(\frac{c_y(2\omega)}{n_e(2\omega)} - 2C_y \right),$$

$$C_y = \frac{1}{2} \left(\frac{1}{n_o(\omega)} + \frac{c_y(\omega)}{n_e(\omega)} \right).$$

In the type II configuration the positions of the minima of the spectral intensity depend on the waist distance z_0.

IX. CONCLUSION

In this paper we have derived the equations which govern the evolutions of the slowly varying envelopes of pulses in a bulk medium presenting anisotropic properties and nonlinear susceptibilities. In case of phase-matching we have derived the equations that govern the frequency conversion of the source. In case of no phase matching we have derived the nonlinear Schrödinger-type equation that governs the evolution of the pulse. We have shown that the diffraction operator is anisotropic, and that the nonlinear term may be more complicated than the standard Kerr effect due to $\chi^{(2)}$-cascaded effects. We have in particular detected a configuration where stable solitons should be naturally generated since the equation then reads as the standard one-dimensional Schrödinger equation with Kerr nonlinearity. As a natural extension of this work we may also think at the propagation of partially coherent light in a linear or nonlinear anisotropic medium. Indeed the intensity profiles of the speckle spots along the propagation axis are imposed by the diffraction. So an anisotropic diffraction should involve interesting and original characteristics of the speckle spots.

ACKNOWLEDGMENTS

We thank L. Videau, C. Sauteret, C. Rouyer, and A. Migus for useful and stimulating discussions. This work was performed under the auspices of the Laser MegaJoule Program of Commissariat à l'Energie Atomique/Direction des Applications Militaires (Grant No. CEA/DAM N° 0V 1751 CE F7).

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