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Propagation of optical localized pulses in $\chi^{(2)}$ crystals: a $(3 + 1)$ -dimensional model and its reduction to the NLS equation

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Abstract. We derive the equations that describe the evolution of the modulation of a short localized optical pulse in a bulk medium presenting a nonzero second-order nonlinear susceptibility, far from the phase-matching. We give the equations in the general case, and for the symmetry classes of crystals $\bar{4}2m$ and $3m$. A wave interaction between the wave and some DC field, due to optical rectification and the electro-optic effect, strongly modifies the obtained system with regard to the three-dimensional NLS equation. An exhaustive list of the cases where the derived model can be reduced to the one-dimensional NLS equation is given. This reduction is possible for only a few special choices of the polarization and of the modulation direction, in a way corresponding to the symmetry of the crystal. The reduction of the same system to two coupled one-dimensional NLS equations is also investigated. With regard to the integrable systems of this type, an additional nonlinear term arises in all but a few particular cases.

1. Introduction

While the problem of soliton propagation in optical fibres has been extensively studied [1], in so far as industrial applications are now under investigation, the analogous problem in bulk media, and even in planar waveguides, is not yet completely solved. Indeed, it has been shown that, for a third-order Kerr nonlinearity, the propagation of a short localized pulse is described by the so-called nonlinear Schrödinger (NLS) equation [2]. The proper NLS equation is the $(1 + 1)$ -dimensional case, which is completely integrable by means of the inverse scattering transform (IST) method, and therefore admits soliton solutions. Zakharov *et al* [3] showed that the solutions are always unstable in more than $(1 + 1)$ dimensions: the input pulse either collapses or is spread out due to diffraction and dispersion. Is the same behaviour expected with second-order nonlinearities? It is well known that so-called cascading can lead to an analogue to the Kerr effect [4, 5]. At phase matching, or very close to it, a large conversion in second harmonic generation is possible, thus the corresponding effective Kerr coefficient is very large. It has been measured by several experimenters [6–9]. The obtained values are much larger than in any third-order material. This recently led to renewed interest in cascading, that showed the possibility of ‘soliton-like’ propagation [10–12], that has been experimentally observed [13]. As early as 1981, Kanashov and Rubenchik [14] showed that collapse never occurs through phase-matched cascading. Recent works taking into account a phase mismatch and

the effect of a cubic (Kerr) nonlinearity, together with the quadratic one, showed that either complete dispersion, collapse or soliton-like propagation may occur, depending on nonlinear coefficients and input power [15].

In this paper we are interested in modelling the behaviour of the wave far from the phase matching. There the existence of solitons is linked to the possibility to describe the propagation with the NLS equation. This possibility has already been demonstrated in the (1 + 1)-dimensional case [16]. A large part of the subject matter of this paper is the (3 + 1)-dimensional case: the case of a short pulse in a bulk medium. It is important from a fundamental point of view, but also for applications such as optical switching. Further, the models describing the physics in (2 + 1) or (1 + 1) dimensions can be derived from the (3 + 1)-dimensional model with great generality and rigor. In particular, the waves are always modulated in time: even if the coherence length is very large, it is finite. Further, a longitudinal (or temporal) modulation may appear during the propagation, even if the wave is continuous. Thus the temporal variable may not, in principle, be removed before checking that this well known Benjamin–Feir instability does not occur. Assuming that the wave is absolutely not modulated in time, or assuming that this modulation is very weak but not zero, yield different asymptotic models, because some wave interactions can be missed in the former case. The correct physical situation is obviously the latter case, thus correct low-dimensional models would better be derived by reduction of the (3 + 1)-dimensional asymptotic than directly. We derive these model equations in the first section of the paper; they describe some interaction between the wave and a DC electric field, through optical rectification and the electro-optic effect. Further, our study takes into account the tensorial structure of the linear and nonlinear susceptibilities, which was not accounted for in [16]. This is crucial in more than (1 + 1) dimensions, and also strongly modifies the behaviour in the low-dimensional case.

The second part of the paper deals with the reductions of the (3 + 1)-dimensional model to the (1 + 1)-dimensional NLS equation. The most important result is that this reduction is not obtained in any case. For example, spatial NLS solitons or dark solitons are obtained only when the polarization of the incident wave has some special direction with respect to the crystallographic axes of the nonlinear material. Then its reductions to a system of two coupled (1 + 1)-dimensional NLS equations is investigated. The resulting equations appear to be integrable only if several rather strong conditions are satisfied. Some particular analytical solutions are given in the nonintegrable case.

2. Derivation of the evolution equations in the general case

2.1. The model

Assuming that the magnetic susceptibility is negligible, or, at least, a scalar real constant, the Maxwell equations reduce to the following wave equation for the electric field \mathbf{E} :

$$\nabla \wedge (\nabla \wedge \mathbf{E}) = -\frac{1}{c^2} \partial_t^2 \mathbf{D} \quad (1)$$

where \mathbf{D} is the electric induction. We assume that \mathbf{D} can be described by the following standard model: it divides into the sum $\mathbf{D} = \mathbf{D}_l + \mathbf{P}_{nl}$ of a linear part \mathbf{D}_l satisfying,

$$\mathbf{D}_l = \chi^{(1)} * \mathbf{E} = \int_{-\infty}^t dt_1 \chi^{(1)}(t - t_1) \mathbf{E}(t_1) \quad (2)$$

and a nonlinear part \mathbf{P}_{nl} , corresponding to the nonlinear polarization such that

$$\begin{aligned} \mathbf{P}_{nl} &= \chi^{(2)} * (\mathbf{E}, \mathbf{E}) + \chi^{(3)} * (\mathbf{E}, \mathbf{E}, \mathbf{E}) \\ &= \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \chi^{(2)}(t - t_1, t - t_2) : \mathbf{E}(t_1)\mathbf{E}(t_2) \\ &\quad + \int_{-\infty}^t dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \chi^{(3)}(t - t_1, t - t_2, t - t_3) : \mathbf{E}(t_1)\mathbf{E}(t_2)\mathbf{E}(t_3) \end{aligned} \quad (3)$$

a priori, the nonlinear susceptibilities $\chi^{(1)}$, $\chi^{(2)}$ and $\chi^{(3)}$ are complex tensors of rank 2, 3 and 4, respectively. Expressions (2) and (3) describe an instantaneous response to the signal, linear, quadratic or cubic in the domain of the frequencies. Assuming that the electromagnetic wave is far enough from all absorption lines of the medium, the absorption can be completely neglected. Then $\chi^{(1)}$, $\chi^{(2)}$ and $\chi^{(3)}$ are real.

The $\chi^{(2)}$ -tensor is zero for any centrosymmetric crystal. Thus a simple isotropy hypothesis for $\chi^{(1)}$, $\chi^{(2)}$ and $\chi^{(3)}$ is not physically relevant. The second-order nonlinear materials have, from the crystallographic point of view, point-group symmetries of the classes $\bar{4}3m$ (e.g. GaAs), $\bar{4}2m$ (e.g. KDP), $3m$ (e.g. LiNbO₃), for the most common of them. Concerning the linear optical index, the first of these classes is isotropic, the others are uniaxial. Therefore we assume that $\chi^{(1)}$ corresponds to the uniaxial case. Nevertheless, the aim of this paper is not to study the effects of the linear anisotropy of the medium, but only to take a rigorous account of it in the study of the nonlinear behaviour. We shall impose that the propagation direction is parallel to the the optical axis, and then choose it as the z -axis. All polarizations are then transverse and propagate with the same velocity. The Fourier transform $\hat{\chi}^{(1)}$ of $\chi^{(1)}$ writes

$$\hat{\chi}^{(1)} = \begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix} \quad (4)$$

n_e and n_o are respectively the ordinary and extraordinary refractive indices of the medium. The isotropic case, an example of which is GaAs, will be recovered by setting $n_e = n_o$.

The structures of the $\chi^{(2)}$ and $\chi^{(3)}$ -tensors are more complicated, and depend on the point group symmetry. First we shall present the derivation of the evolution equations, under a form valid for any structure of these tensors, and thereafter, we will give the particular form of these equations in the two particular cases mentioned above. We will give the precise tensorial structure of the susceptibilities at this point. For simplicity, in what follows, subscripts l in \mathbf{D}_l and nl in \mathbf{P}_{nl} shall be omitted.

2.2. The multiscale expansion

The fields \mathbf{E} , \mathbf{D} (that is, \mathbf{D}_l) and \mathbf{P} (that is, \mathbf{P}_{nl}) are expanded simultaneously in a power series of some small parameter ε , and in a Fourier series with respect to some phase:

$$\varphi = kz - \omega t$$

$\omega/2\pi$ is the frequency of the wave, and $\mathbf{x} = (x, y, z)$ and t are respectively the three-dimensional space and the time variables. The propagation direction is chosen as the z -axis, which, as mentioned above, is assumed to be the optical axis of the crystal. This expansion reads

$$\mathbf{E} = \sum_{l \geq 1, p \in \mathbb{Z}} \varepsilon^l \mathbf{E}_l^p e^{ip\varphi} \quad (5)$$

with the reality condition $E_l^{-p} = E_l^{p,*}$ for each l and p ($*$ denotes complex conjugation). In fact, the E_l^p are all zero for $|p| > l$; this is set *a priori*, for physical reasons, at leading order, and follows from the resolution of the expansion equations at higher orders. The amplitudes E_l^p are functions of slow variables (τ, ξ, η, ζ) defined by,

$$\begin{cases} \tau = \varepsilon \left(t - \frac{z}{v} \right) \\ \xi = \varepsilon x \\ \eta = \varepsilon y \\ \zeta = \varepsilon^2 z. \end{cases} \quad (6)$$

v is a velocity to be determined, it will be found equal to the group velocity. τ is the variable that describes the shape of a given pulse, and ζ the variable that describes the evolution of this shape during the propagation of the pulse. Note that there exists an alternative definition of these slow variables. It reads,

$$\begin{cases} \zeta' = \varepsilon(z - vt) \\ \tau' = \varepsilon^2 t. \end{cases} \quad (7)$$

The variable ζ' is obviously equivalent to τ , and τ' is also equivalent to ζ . Thus both τ and ζ can in fact be considered as time variables as well as space variables.

Many terms in expansion (5) are zero. The choice of the leading order is a crucial part of the description of the physical problem under consideration. To fix which term is the first nonzero one in the expansion (5), that is, to determine the order of magnitude of the intensity of the input pulse, will complete the ansatz. Note that this intensity scale is defined in relation to the time- and spacescales, that is, the extension of the considered pulse. The scaling (6) assumes that this extension has the same order of magnitude in all directions.

Here we assume that the fundamental frequency wave (terms with $p = 1$ in equation (5) and their complex conjugate with $p = -1$) has an amplitude of order ε , and that the amplitude of the second harmonic ($p = \pm 2$ in equation (5)) is very small with respect to the fundamental, i.e. at most of order ε^2 . Thus,

$$\mathbf{E} = \varepsilon(\mathbf{E}_1^1 e^{i\varphi} + c.c.) + O(\varepsilon^2) \quad (8)$$

(*c.c.* means complex conjugate). Expansions (5) and (6) are substituted into the basic equations (1)(2)(3). Then we collect the coefficients of each power of ε , and obtain a set of equations, that we solve, order by order. Some technical details of the computation are given in the appendix.

2.3. The order by order resolution

- *order ε^1 .* The equations obtained at order ε^1 give the linear behaviour of the wave; it is seen that $E_1^{1,z} = 0$ (the wave is transverse up to this order) and that the wavenumber reads $k = n_o \omega / c$, where $n_o(\omega) = \sqrt{\hat{\chi}_o^{(1)}(\omega)}$ is the ordinary linear refractive index of the medium. We will write simply n_o for $n_o(\omega)$, where ω is the fundamental frequency of the wave, and mention explicitly the variable in the other cases. We make use of the notations n'_o, n''_o, \dots , for the ω -derivatives of $n_o, n_o^2(\omega)$ for $(n_o(\omega))^2$, and so on.

• *order ε^2* . Solving the system of the order ε^2 , we find first some compatibility condition, which shows that v must be the group velocity of the wave:

$$v = \frac{c}{n_o + \omega n'_o}.$$

The resolution yields a zero value for all E_2^p with $|p| \geq 3$, and the following nonzero term:

$$E_2^{1,z} = \frac{ic n_o}{\omega n_e^2} (\partial_\xi E_1^{1,x} + \partial_\eta E_1^{1,y}). \tag{9}$$

Thus the wave is no more transverse at this order. It should be noted that this longitudinal term is not caused by anisotropy or nonlinearity, but only by the scaling (6). The presence of the slow variables could represent a slight deviation in the propagation direction, of order ε . The dominant term of the second harmonic is obtained:

$$E_2^{2,s} = \frac{1}{n_o^2(\omega) - n_o^2(2\omega)} P_2^{2,s} \tag{10}$$

for $s = x, y$, and,

$$E_2^{2,z} = \frac{-1}{n_e^2(2\omega)} P_2^{2,z}. \tag{11}$$

The nonlinear polarization term is given by,

$$P_2^2 = \hat{\chi}^{(2)}(\omega, \omega) : E_1^1 E_1^1. \tag{12}$$

Equation (10) is valid under the assumption that $n_o(\omega) \neq n_o(2\omega)$; i.e. that there is no phase matching. If $n_o(2\omega)$ approaches $n_o(\omega)$, the term $E_2^{2,s}$ in equation (10) tends to infinity. At the limit, a term of order ε , very large with respect to ε^2 , must be introduced. This case where the fundamental and second harmonic have the same order of magnitude is the proper case of the second harmonic generation; we do not intend to study this here. Indeed, by phase matching, the resonant interaction can produce amplitudes of the same order of magnitude for both the fundamental and the second harmonic. Here, far from phase matching, the second harmonic is completely defined by the fundamental, in an algebraic way, and does not propagate with its own phase and group velocity, but with those of the fundamental. In counterpart, it stays very small. We do not have, properly speaking, a wave interaction, but a train of harmonics produced by the fundamental and following it.

For $p = 0$, the equations obtained at this order are trivial, and the equations that permit the computation of the term E_2^0 are obtained at order ε^4 . They show that E_2^0 cannot be zero. This is a term with no carrying wave, called ‘zero harmonic’ or ‘mean-value term’. It represents an electromagnetic wave whose typical wavelength is very large with regard to the optical ones, because it has the magnitude of the pulse length. Indeed, E_2^0 is a function of τ, ξ , and η , which are variables of order ε , that describe the pulse shape. (E_2^0 depends also on ζ , at next order). It corresponds to a zero value of the pulsation, that is the same, in the frame of an expansion in power series of ε , as a value of the order of magnitude of $\varepsilon\omega$ [16]. The importance of such terms has often be pointed out in several physical contexts. As an example, the present author recently published a study of the interactions between such terms and a fast oscillating wave of very high intensity in ferromagnetic media [17]. Their importance has been recognized in the optical frame too [16]. In the present problem, partial differential equations (PDEs), that describe an interaction between the mean-value

term and the fundamental, are obtained. They read,

$$\begin{aligned} [\alpha \partial_\xi^2 + \partial_\eta^2 + \rho \partial_\tau^2] E_2^{0,x} + (\alpha - 1) \partial_\xi \partial_\eta E_2^{0,y} &= \left(\frac{1}{c^2} \partial_\tau^2 - \frac{1}{n_e^2(0)} \partial_\xi^2 \right) P_2^{0,x} \\ &- \frac{1}{n_e^2(0)} \partial_\xi \partial_\eta P_2^{0,y} + \frac{1}{vn_e^2(0)} \partial_\xi \partial_\tau P_2^{0,z} \end{aligned} \quad (13)$$

$$\begin{aligned} [\partial_\xi^2 + \alpha \partial_\eta^2 + \rho \partial_\tau^2] E_2^{0,y} + (\alpha - 1) \partial_\xi \partial_\eta E_2^{0,x} &= \left(\frac{1}{c^2} \partial_\tau^2 - \frac{1}{n_e^2(0)} \partial_\eta^2 \right) P_2^{0,y} \\ &- \frac{1}{n_e^2(0)} \partial_\xi \partial_\eta P_2^{0,x} + \frac{1}{vn_e^2(0)} \partial_\eta \partial_\tau P_2^{0,z}. \end{aligned} \quad (14)$$

The constants α and ρ are given by

$$\rho = \frac{1}{v^2} - \frac{n_o^2(0)}{c^2} = \frac{(n_o + \omega n'_o)^2 - n_o^2(0)}{c^2} \quad (15)$$

$$\alpha = \frac{n_o^2(0)}{n_e^2(0)} \quad (16)$$

and the nonlinear polarization mean-value term P_2^0 is

$$P_2^0 = 2\hat{\chi}^{(2)}(\omega, -\omega) : \mathbf{E}_1^1 \mathbf{E}_1^{1,*}. \quad (17)$$

It is well known that this expression describes optical rectification. Because P_2^0 is nonzero in general, so is E_2^0 . On the other hand, $E_2^{0,z}$ can be written explicitly in terms of $E_2^{0,x}$ and $E_2^{0,y}$ (equation (108), see the appendix).

• *order ε^3* . At the following order ε^3 , another set of equations is found. We are interested in the two equations corresponding to the components x and y of the fundamental; using the dispersion relation, the terms in $E_3^{1,x}$ and $E_3^{1,y}$ cancel, and using the expression of the group velocity, so do the terms in $E_2^{1,x}$ and $E_2^{1,y}$. This yields, as a compatibility condition, the evolution equations for $E_1^{1,x}$ and $E_1^{1,y}$. They read,

$$[2ik\partial_\xi + \beta \partial_\xi^2 + \partial_\eta^2 - kk'' \partial_\tau^2] E_1^{1,x} + (\beta - 1) \partial_\xi \partial_\eta E_1^{1,y} = \frac{-\omega^2}{c^2} P_3^{1,x} \quad (18)$$

$$[2ik\partial_\xi + \partial_\xi^2 + \beta \partial_\eta^2 - kk'' \partial_\tau^2] E_1^{1,y} + (\beta - 1) \partial_\xi \partial_\eta E_1^{1,x} = \frac{-\omega^2}{c^2} P_3^{1,y}. \quad (19)$$

The constant β is given by

$$\beta = \frac{n_o^2}{n_e^2}$$

and reduces to 1 if the medium is isotropic. In this case, the crossed terms vanish, and the transverse derivative reduces to a simple Laplacian operator. However, for most of the common $\chi^{(2)}$ -materials, anisotropy cannot be neglected and $\beta \neq 1$. The nonlinear term reads,

$$P_3^1 = 2[\hat{\chi}^{(2)}(2\omega, -\omega) : \mathbf{E}_2^2 \mathbf{E}_1^{1,*} + \hat{\chi}^{(2)}(0, \omega) : \mathbf{E}_2^0 \mathbf{E}_1^1] + 3\hat{\chi}^{(3)}(\omega, \omega, -\omega) : \mathbf{E}_1^1 \mathbf{E}_1^1 \mathbf{E}_1^{1,*}. \quad (20)$$

We have derived the system of PDEs that describes the evolution of the modulation of a short localized pulse in a bulk second-order nonlinear medium; it is composed of four

coupled equations: equations (18), (19) describing the evolution of the amplitudes of the two wave polarizations, and two propagation equations (13), (14) for the slowly varying field E_2^0 . The fast oscillating wave yields a source term in these equations, through optical rectification. The nonlinear r.h.s. of equations (18), (19) given by equation (20), describes a self-interaction, but also interaction between the fast oscillating and the slowly varying wave: the electro-optic effect.

3. Evolution equations for some particular classes of crystals

3.1. Structure of the nonlinear susceptibilities

[18] gives the structure of the tensor, for all crystal classes. There are mainly two cases to be considered. The first one—and the simplest one—is the case of the $\bar{4}2m$ class. The $\chi^{(2)}$ -tensor has only six nonvanishing components, among which three are independent:

$$\chi_{xyz}^{(2)} = \chi_{yxz}^{(2)} \quad \chi_{xzy}^{(2)} = \chi_{yzx}^{(2)} \quad \chi_{zxy}^{(2)} = \chi_{zyx}^{(2)}. \quad (21)$$

This class contains in particular KH_2PO_4 (KDP), but also many other materials. The second case is this of the $3m$ symmetry class (trigonal and optically uniaxial). The $\hat{\chi}^{(2)}$ -tensor has the following structure: there are 11 nonvanishing components, among which five are independent:

$$\begin{aligned} \chi_{xzx}^{(2)} = \chi_{yzy}^{(2)} & \quad \chi_{xxz}^{(2)} = \chi_{yyz}^{(2)} & \quad \chi_{zxx}^{(2)} = \chi_{zyy}^{(2)} \\ \chi_{zzz}^{(2)} & \quad \chi_{yyy}^{(2)} = -\chi_{yxx}^{(2)} = -\chi_{xxxy}^{(2)} = -\chi_{xyx}^{(2)} \end{aligned} \quad (22)$$

(for some particular orientation of the crystal). An important example of material belonging to this class is the Lithium niobate LiNbO_3 .

The second-order nonlinearity acts on the fundamental only through cascaded terms, which are, from a mathematical point of view, of the same order as the term due to the $\chi^{(3)}$ Kerr nonlinearity. That is why the $\chi^{(3)}$ -tensor cannot be neglected. Its symmetry properties are found, for example, in [18]. For the $\bar{4}2m$ class, the $\chi^{(3)}$ -tensor has 21 nonzero elements, among which 11 are independent:

$$\begin{aligned} \chi_{xxxx}^{(3)} = \chi_{yyyy}^{(3)} = \chi_{zzzz}^{(3)} \\ \chi_{yyzz}^{(3)} = \chi_{xxzz}^{(3)} \quad \chi_{yzzy}^{(3)} = \chi_{xzxx}^{(3)} \quad \chi_{xxyy}^{(3)} = \chi_{yyxx}^{(3)} \\ \chi_{zzyy}^{(3)} = \chi_{zzxx}^{(3)} \quad \chi_{yzyz}^{(3)} = \chi_{xzxx}^{(3)} \quad \chi_{xyxy}^{(3)} = \chi_{yxyx}^{(3)} \\ \chi_{zyyz}^{(3)} = \chi_{zxxx}^{(3)} \quad \chi_{zyzy}^{(3)} = \chi_{zxzx}^{(3)} \quad \chi_{xyyx}^{(3)} = \chi_{yxyx}^{(3)}. \end{aligned} \quad (23)$$

Regarding the $3m$ class, we have 37 nonzero elements, among which 14 are independent:

$$\begin{aligned} \chi_{zzzz}^{(3)} \\ \chi_{xxxx}^{(3)} = \chi_{yyyy}^{(3)} = \chi_{xxyy}^{(3)} + \chi_{xyyx}^{(3)} + \chi_{xyxy}^{(3)} \\ \chi_{xxyy}^{(3)} = \chi_{yyxx}^{(3)} \quad \chi_{yyzz}^{(3)} = \chi_{xxzz}^{(3)} \quad \chi_{zzyy}^{(3)} = \chi_{zzxx}^{(3)} \\ \chi_{xyyx}^{(3)} = \chi_{yxxy}^{(3)} \quad \chi_{zyyz}^{(3)} = \chi_{zxxx}^{(3)} \quad \chi_{yzzy}^{(3)} = \chi_{xzxx}^{(3)} \\ \chi_{xyxy}^{(3)} = \chi_{yxyx}^{(3)} \quad \chi_{yzyz}^{(3)} = \chi_{xzxx}^{(3)} \quad \chi_{zyzy}^{(3)} = \chi_{zxzx}^{(3)} \\ \chi_{xxxz}^{(3)} = -\chi_{xyyz}^{(3)} = -\chi_{yxzy}^{(3)} = -\chi_{yyxz}^{(3)} \\ \chi_{xxzx}^{(3)} = -\chi_{xyzy}^{(3)} = -\chi_{yxzy}^{(3)} = -\chi_{yyzx}^{(3)} \\ \chi_{xzxx}^{(3)} = -\chi_{xzyy}^{(3)} = -\chi_{yzxy}^{(3)} = -\chi_{yzyx}^{(3)} \\ \chi_{zxxx}^{(3)} = -\chi_{zxyy}^{(3)} = -\chi_{zyxy}^{(3)} = -\chi_{zyyx}^{(3)}. \end{aligned} \quad (24)$$

The computation valid for the $\bar{4}2m$ class is in fact valid for any material for which the structure of the $\chi^{(1)}$, $\chi^{(2)}$ and $\chi^{(3)}$ -tensors satisfies equations (4), (21) and (23) respectively. The symmetry class $43m$ also satisfies equations (4) and (21), but with the particular property that $\hat{\chi}_{xyz}^{(2)} = \hat{\chi}_{xzy}^{(2)} = \hat{\chi}_{zxy}^{(2)}$ (and $n_e = n_o$). These crystals also satisfy equation (23), with the additional symmetry properties,

$$\begin{aligned}\chi_{xxxx}^{(3)} &= \chi_{zzzz}^{(3)} \\ \chi_{xxzz}^{(3)} &= \chi_{zzxx}^{(3)} = \chi_{xxyy}^{(3)} \\ \chi_{xzxx}^{(3)} &= \chi_{zxzx}^{(3)} = \chi_{xyxy}^{(3)} \\ \chi_{xzxz}^{(3)} &= \chi_{zxxz}^{(3)} = \chi_{xyyx}^{(3)}.\end{aligned}\quad (25)$$

Thus the results valid for the $\bar{4}2m$ class are also valid for the crystals of the $\bar{4}3m$ class. These are cubic crystals, optically isotropic (from a linear point of view). An important material belonging to this class is GaAs. If $\hat{\chi}_{yyy}^{(2)} = 0$, equation (22) describes the $\chi^{(2)}$ -structure for the $4mm$ and $6mm$ crystal classes. The $\chi^{(3)}$ -structure for the $4mm$ class differs from equation (24), but the $6mm$ class satisfies these conditions, with the additional requirements that the components $\chi_{xxxz}^{(3)}$, $\chi_{xxzx}^{(3)}$, $\chi_{xzxx}^{(3)}$, and $\chi_{zxxx}^{(3)}$ (and those that have the same value according to equation (24)) are zero. The corresponding computation will thus not be valid only for the $3m$, but also for the $6mm$ crystal class, that also contains many common $\chi^{(2)}$ -materials.

3.2. The $\bar{4}2m$ symmetry class

Now we have to compute the nonlinear terms, using the structure of the $\chi^{(2)}$ -tensor, that is given by equation (21) in this first particular case. This derivation is detailed in the appendix, and yields the following system:

$$\begin{aligned}[2ik\partial_\xi + \beta\partial_\eta^2 + \partial_\eta^2 - kk''\partial_\tau^2]E_1^{1,x} + (\beta - 1)\partial_\xi\partial_\eta E_1^{1,y} \\ = D_1 E_1^{1,x} |E_1^{1,x}|^2 + D_2 E_1^{1,x} |E_1^{1,y}|^2 + D_3 (E_1^{1,y})^2 E_1^{1,x,*} + E\Phi E_1^{1,y}\end{aligned}\quad (26)$$

$$\begin{aligned}[2ik\partial_\xi + \partial_\xi^2 + \beta\partial_\eta^2 - kk''\partial_\tau^2]E_1^{1,y} + (\beta - 1)\partial_\xi\partial_\eta E_1^{1,x} \\ = D_1 E_1^{1,y} |E_1^{1,y}|^2 + D_2 E_1^{1,y} |E_1^{1,x}|^2 + D_3 (E_1^{1,x})^2 E_1^{1,y,*} + E\Phi E_1^{1,x}\end{aligned}\quad (27)$$

$$[\alpha(\partial_\xi^2 + \partial_\eta^2) + \rho\partial_\tau^2]\Phi = \lambda(\partial_\xi^2 + \partial_\eta^2)(E_1^{1,x} E_1^{1,y,*} + E_1^{1,x,*} E_1^{1,y}).\quad (28)$$

The function Φ is some combination of $E_2^{0,x}$ and $E_2^{0,y}$, given by equation (119) in the appendix. The interaction constants read as follows:

$$D_1 = \frac{-3\omega^2}{c^2} \hat{\chi}_{xxxx}^{(3)}(\omega, \omega, -\omega)\quad (29)$$

$$\begin{aligned}D_2 = \frac{4\omega^2}{c^2} \frac{1}{n_e^2(2\omega)} \hat{\chi}_{xzy}^{(2)}(2\omega, -\omega) \hat{\chi}_{zxy}^{(2)}(\omega, \omega) + \frac{4\omega^2}{c^2} \frac{1}{n_e^2(0)} \hat{\chi}_{xzy}^{(2)}(0, \omega) \hat{\chi}_{zxy}^{(2)}(\omega, -\omega) \\ - \frac{3\omega^2}{c^2} (\hat{\chi}_{xxyy}^{(3)}(\omega, \omega, -\omega) + \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega))\end{aligned}\quad (30)$$

$$D_3 = \frac{4\omega^2}{c^2} \frac{1}{n_e^2(0)} \hat{\chi}_{xzy}^{(2)}(0, \omega) \hat{\chi}_{zxy}^{(2)}(\omega, -\omega) - \frac{3\omega^2}{c^2} \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega)\quad (31)$$

$$E = \frac{-2\omega^2}{c^2} \frac{vn_o^2(0)}{n_e^2(0)} \hat{\chi}_{xzy}^{(2)}(0, \omega)\quad (32)$$

$$\lambda = \frac{2}{vn_e^2(0)} \hat{\chi}_{zxy}^{(2)}(\omega, -\omega). \tag{33}$$

λ accounts for the interaction $\omega + (-\omega) \rightarrow 0$, that produces the zero harmonic or mean-value term (the rectified field). The constant E accounts for the interaction $\omega + 0 \rightarrow \omega$ of the latter with the fundamental, which is the electro-optic effect, and, in addition to the third-order Kerr effect, $D_3 = \frac{-1}{\alpha} E \lambda - \frac{3\omega^2}{c^2} \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega)$ describes the cumulative effect of the two preceding interactions, while $(D_2 - D_3)$ describes the cascaded interactions $\omega + \omega \rightarrow 2\omega$ and $2\omega + (-\omega) \rightarrow \omega$. Equations (17)–(27) have a form analogous to those of the NLS equation (47) in $(3 + 1)$ dimensions, coupled together, and also coupled to a third equation (equation (28)). The latter is (at least if the product $\alpha\rho$ is negative) a wave equation for the field Φ , with a source term function of the intensity of the fundamental. This system can be derived from the following Lagrangian density:

$$\begin{aligned} \mathcal{L} = & ik(UU_\zeta^* - U^*U_\zeta + VV_\zeta^* - V^*V_\zeta) + \beta U_\xi U_\xi^* + U_\eta U_\eta^* - kk''U_\tau U_\tau^* + V_\xi V_\xi^* \\ & + \beta V_\eta V_\eta^* - kk''V_\tau V_\tau^* + \frac{\beta - 1}{2}(V_\xi U_\eta^* + V_\eta U_\xi^* + V_\xi^* U_\eta + V_\eta^* U_\xi) \\ & + \frac{D_1}{2}(|U|^4 + |V|^4) + D_2|U|^2|V|^2 + \frac{D_3}{2}(V^2U^{*2} + U^2V^{*2}) \\ & + E(\Phi_{\xi\xi} + \Phi_{\eta\eta})(UV^* + VU^*) \\ & - \frac{E}{2\lambda}(\Phi_{\xi\xi} + \Phi_{\eta\eta})(\alpha(\Phi_{\xi\xi} + \Phi_{\eta\eta}) + \rho\Phi_{\tau\tau}). \end{aligned} \tag{34}$$

We use the shortened notation:

$$E_1^{1,x} = U \quad E_1^{1,y} = V \tag{35}$$

for the fields, and $\partial_\zeta U = U_\zeta$, etc for the derivatives. System (26)–(28) is derived from the Lagrangian density (34) by setting the total variation of the action equal to zero:

$$S = \int_{\mathbb{R}^4} \mathcal{L} d\xi d\eta d\zeta d\tau. \tag{36}$$

More precisely, equation (26) for $U = E_1^{1,x}$ is given by $\frac{\delta S}{\delta U^*} = 0$, equation (27) for $V = E_1^{1,y}$ by $\frac{\delta S}{\delta V^*} = 0$, and equation (28) for Φ by $\frac{\delta S}{\delta \Phi} = 0$. A Hamiltonian representation can be found using the density,

$$\mathcal{H} = \mathcal{L} - ik(UU_\zeta^* - U^*U_\zeta + VV_\zeta^* - V^*V_\zeta) \tag{37}$$

and:

$$H = \int_{\mathbb{R}^3} \mathcal{L} d\xi d\eta d\tau. \tag{38}$$

Then the equations (26)–(28) are obtained by,

$$\frac{\delta H}{\delta U^*} = 2ikU_\zeta \quad \frac{\delta H}{\delta V^*} = 2ikV_\zeta \quad \frac{\delta H}{\delta \Phi} = 0 \tag{39}$$

(but $\Phi_\zeta \neq 0$). Then the following conservation law holds:

$$\frac{dH}{d\zeta} = 0. \tag{40}$$

System (26)–(28) can be considered as a $(3 + 1)$ -dimensional form of the NLS equation, in the sense that it can be reduced to this completely integrable $(1 + 1)$ -dimensional equation

in several ways. These reductions will be studied in next section of this paper. Note that system (26)–(28) is not likely to possess the complete integrability property, even for some very special value of the coefficients, despite the fact that this feature is not proved. We will not make any attempt to solve this system in any way, but notice the following essential feature: the system (26)–(28), that describes the evolution of the modulation of a short pulse in a bulk second-order material of the $\bar{4}2m$ class, is not the simple so-called three-dimensional NLS equation:

$$(iA\partial_\zeta + \partial_\xi^2 + \partial_\eta^2 + B\partial_\tau^2)f + f|f|^2 = 0. \quad (41)$$

Therefore, the result by Zakharov *et al* [3], who proved that equation (41) has no stable solution, but leads for any initial data either to collapse or to complete dispersion, is not valid for the cascading of second-order optical nonlinearities in the materials of the $\bar{4}2m$ symmetry class, to which KDP belongs. Note also that system (26)–(28) is not the system that describes the evolution of a short localized pulse in a material with nonzero second-order nonlinear susceptibility, when the phase matching is realized. It has been proved [14] that collapse never occurs in the physical situation described by this latter system, and that it has soliton solutions, in the sense of stable localized pulses. Thus the problem of the existence of stable localized solutions of equations (26)–(28), that is, of optical light bullets in these materials, is still open.

The system (26)–(28) is analogous in its form to the completely integrable $(2+1)$ -dimensional Davey–Stewartson (DS) system [19]. More precisely, when reduced to a single polarization and $(2+1)$ dimensions, it has exactly the same form, but with other coefficients. The completely integrable case is reached if certain conditions between the coefficients are satisfied [20]. A remarkable feature is that the integrable reduction is the so-called DS I system, for which localized solitons exist [21]. This proves that system (26)–(28) can describe multidimensional solitons, at least in some particular two-dimensional case.

3.3. The $3m$ symmetry class.

For the $3m$ symmetry class, the structure of the $\chi^{(2)}$ -tensor is given by equation (22). The expressions are more complicated than in the previous case; we use the simplified notations (35). After some algebra, which is detailed in the appendix, we obtain the following partial differential system:

$$\begin{aligned} [2ik\partial_\zeta + \beta\partial_\xi^2 + \partial_\eta^2 - kk''\partial_\tau^2]U + (\beta - 1)\partial_\eta\partial_\xi V &= D_1U|U|^2 + D_2U|V|^2 \\ &+ D_3V^2U^* + F_1(E_2^{0,x}V + E_2^{0,y}U) + F_2U \int^\tau (\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) \\ &+ F_3U \int^\tau [\partial_\xi(UV^* + VU^*) + \partial_\eta(|U|^2 - |V|^2)] \end{aligned} \quad (42)$$

$$\begin{aligned} [2ik\partial_\zeta + \partial_\xi^2 + \beta\partial_\eta^2 - kk''\partial_\tau^2]V + (\beta - 1)\partial_\eta\partial_\xi U &= D_1V|V|^2 + D_2V|U|^2 \\ &+ D_3U^2V^* + F_1(E_2^{0,x}U - E_2^{0,y}V) + F_2V \int^\tau (\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) \\ &+ F_3V \int^\tau [\partial_\xi(UV^* + VU^*) + \partial_\eta(|U|^2 - |V|^2)] \end{aligned} \quad (43)$$

$$\begin{aligned} [\alpha\partial_\xi^2 + \partial_\eta^2 + \rho\partial_\tau^2]E_2^{0,x} + (\alpha - 1)\partial_\eta\partial_\xi E_2^{0,y} &= (\lambda_1\partial_\xi^2 + \lambda_3\partial_\tau^2)(UV^* + VU^*) \\ &+ \lambda_1\partial_\xi\partial_\eta(|U|^2 - |V|^2) + \lambda_2\partial_\xi\partial_\tau(|U|^2 + |V|^2) \end{aligned} \quad (44)$$

$$[\partial_\xi^2 + \alpha\partial_\eta^2 + \rho\partial_\tau^2]E_2^{0,y} + (\alpha - 1)\partial_\eta\partial_\xi E_2^{0,x} = (\lambda_1\partial_\eta^2 + \lambda_3\partial_\tau^2)(|U|^2 - |V|^2)$$

$$+\lambda_1 \partial_\xi \partial_\eta (UV^* + VU^*) + \lambda_2 \partial_\eta \partial_\tau (|U|^2 + |V|^2). \tag{45}$$

The constants are given in the appendix, formulae (129)–(140). In this case, the integro-differential four-fields system cannot be simplified, nor written in a Lagrangian form, as easily as in the previous one. But the observations concerning the previous case are still valid: the evolution of the amplitude modulation is neither governed by the three-dimensional NLS equation (41), nor by the system that describes cascaded propagation at phase matching, but by the complicated system (42)–(45) above. Thus the instability result of [3] does not apply here, and neither does the stability result of [14]. The question whether stable localized solutions of this system exist is thus still open. System (42)–(45), like system (26)–(28), is rather similar to the completely integrable DS system, and can be reduced to it in (2 + 1) dimensions, if some conditions on the coefficients are satisfied. Then the signs allow the existence of localized bidimensional solitons [20]. Thus the interaction with the rectified field can lead to the stabilization of the pulse, at least in this particular (2 + 1)-dimensional case.

3.4. Values of the interaction constants

The contribution of the DC field through the electro-optic effect and optical rectification is neglected by several authors. Therefore this point must be discussed carefully. Measured values of the linear electro-optic (Pockels) tensor r_{ijk} can be found in published matter [22]. This quantity is related to the second-order nonlinear susceptibility $d_{ijk} = \frac{1}{2} \hat{\chi}_{ijk}^{(2)}$ by the following relation [22, p 511]:

$$d_{ijk} = -\frac{1}{4} n_i^2 n_j^2 r_{ijk}.$$

Some values of the d_{ijk} , computed using the data given in [22], are listed in the following table, and compared with the values given by the same author for the same quantity in the case of second harmonic generation:

Material	ijk	$ d_{ijk}(0, \omega) $ (10^{-12} m V $^{-1}$)	$ d_{ijk}(\omega, \omega) $ (10^{-12} m V $^{-1}$)
LiNbO ₃	yyy	46.4	4.02
	zzz	181	33.9
	xzx	206	
	zxx	65.5	5.9
KDP	xyz	9.8	
	zxy	14.2	0.72
ADP	xyz	29.6	
	zxy	10.5	0.87

It can be seen that, far from being negligible, the electro-optic component is larger than the second harmonic generation component for a factor of about ten. The same holds for the optical rectification coefficient; although it is not directly measured, the so-called complete symmetry property of the $\chi^{(2)}$ tensor implies that it is equal to the electro-optic component, according to

$$\hat{\chi}_{ijk}^{(2)}(\omega, -\omega) = \hat{\chi}_{kij}^{(2)}(0, \omega).$$

However, these terms appear in the equations with some coefficients multiplying them, and it can be thought that these coefficients modify the orders of magnitude. The quantities

involved read, using the complete symmetry property,

$$C_{ijk}^{SHG} = \frac{(\hat{\chi}_{ijk}^{(2)}(\omega, \omega))^2}{n_e^2(2\omega)} \quad \text{and} \quad C_{ijk}^P = \frac{(\hat{\chi}_{ijk}^{(2)}(\omega, -\omega))^2}{n_e^2(0)}. \quad (46)$$

The square index $n_e^2(0)$ is the static relative dielectric permeability ϵ_r of the medium, along the extraordinary axis. According to the data of [22],

Material	ijk	C_{ijk}^P (10^{-24} m ² V ⁻²)	C_{ijk}^{SHG} (10^{-24} m ² V ⁻²)
LiNbO ₃	yyy	269	13.0
	zzz	4100	922
	zxx	536	27.9
KDP	zxy	38.4	0.94
ADP	zxy	29.4	1.33.

The electro-optic term is about 20 times the second harmonic generation term, despite the fact that the static index is larger than the optical one. Furthermore, in the (1 + 1)-dimensional reductions below, the electro-optic term appears with a factor

$$\frac{2}{(n_o + \omega n'_o)^2 - n_o^2(0)}$$

proportional to the inverse of the difference between the group velocity $v = c/(n_o + \omega n'_o)$ of the wave and the velocity $c/n_o(0)$ of the solitary wave of the DC field. Newell already noticed [2] that this could lead to a very large contribution of the electro-optic term, even for small electro-optic coefficients, if these two velocities were very close together. Although this is not the case in most practical situations, the contribution of the electro-optic effect is not negligible, because of the above mentioned values.

4. The (1 + 1)-dimensional reductions

4.1. The reduction problem

Both systems (26)–(28) and (42)–(45), that describe the evolution of a three-dimensional pulse in a bulk sample of some material belonging to the $\bar{4}2m$ class, or to the $3m$ class respectively, can be reduced to the NLS equation:

$$iA\partial_\zeta f + B\partial_X^2 f + Cf|f|^2 = 0 \quad (47)$$

in (1 + 1) dimensions, by several ways. In that sense, the above mentioned systems are (3 + 1)-dimensional generalizations of the NLS equation, but it is very unlikely that they also possess the property of being completely integrable by the IST method, even for very particular values of the coefficients.

Let us consider first the so-called ‘temporal case’, $X = \tau$. Recall that τ is not strictly speaking a time variable, but describes the longitudinal shape of the pulse, and could also be considered as a space variable, in units of length m/c . In the frame of the multiscale expansion, this means that the wave is not modulated at the spacescale described by the slow variables ξ and η of order ϵ , which is λ/ϵ . The order ϵ^0 in the expansion corresponds to the wavelength λ , and is thus about 1 μm for optical frequencies. A typical value for $1/\epsilon$ is the number of oscillations contained in a picosecond pulse, say a thousand. Thus $\lambda/\epsilon \sim 1$ mm.

The slow transverse variables ξ and η vanish completely in a waveguide or a fibre, with a transverse extent of an order of magnitude midway between λ and λ/ε , e.g. $\lambda/\sqrt{\varepsilon}$. Indeed, if $x \leq \lambda/\sqrt{\varepsilon}$, then $\xi = \varepsilon x \leq \sqrt{\varepsilon}\lambda$, which tends to 0 with ε . Therefore, x cannot take values as large as λ/ε , and there is no ξ -dependency. Further, if x varies up to $\lambda/\sqrt{\varepsilon}$, this upper bound tends to infinity as ε tends to zero. Thus, this describes a plane wave, truly infinite at the x -scale. Even with this, the boundary conditions at infinity imposed by the guide properties for the variables x and y might be different from the chosen conditions (a bounded field), and care must be taken in applications. Note that, for a waveguide width comparable with the wavelength, the boundary conditions corresponding to the waveguide are no longer negligible.

When considering some spatial transverse variable $X = r\xi + s\eta$, one of the transverse variables and the longitudinal—or time—variable τ are removed. The transversal variable is suppressed using some planar waveguide, as was done previously. The removal of the time variable τ is rather different: we must assume that there is no modulation at the timescale $1/\varepsilon\omega$, typically a picosecond, but only at $1/\varepsilon^2\omega$, typically a nanosecond. This means not only that the pulse length must be at least one nanosecond, but also that no faster longitudinal modulation arises inside the pulse during the propagation. Note further that the reduction of the (3 + 1)-dimensional system to (1 + 1) dimensions describes the limit when the time modulation becomes negligible, while a direct derivation of the (1 + 1)-dimensional model would describe the unphysical case where there is no time modulation at all.

From a mathematical point of view, the ‘spatio-temporal’ variable $X = r\xi + s\eta + d\tau$ is totally analogous to the previous ones. We consider it here mainly in order to be exhaustive. In a bulk medium, the temporal case corresponds in principle to a plane wave. The spatio-temporal wave would correspond to such a planar pulse, but whose plane is not orthogonal to the propagation direction. This would be realized, if we could realize a planar pulse in a bulk medium, simply using a prism. The physical meaning of these variables is thus clear, even if it is not commonly realized in experiments.

4.2. Reduction to the nonlinear Schrödinger equation.

We consider a single polarization, defined by

$$E_1^{1,x} = U = af \quad E_1^{1,y} = V = bf \tag{48}$$

where the complex constants a and b are normalized as: $|a|^2 + |b|^2 = 1$. If they are real, we set:

$$a = \cos \theta \quad b = \sin \theta.$$

Using expression (48) in the evolution system yields two evolution equations simultaneously satisfied by the single function f . The requirement that these two equations are identical yields constraints on a , b and on the definition of the variable X . The results are summarized below.

4.2.1. The $\bar{4}2m$ symmetry class.

- *Temporal case.* Let us assume that the structure of the $\chi^{(2)}$ -tensor is given by equation (21), and that the amplitudes E_1^1 and E_2^0 do not depend on ξ and η . Then equation (28) has only the trivial solution $\Phi = 0$. The NLS equation (47) is obtained only if $b = \pm a$, or $b = \pm ia$, or $b = 0$, or $a = 0$. The variable X is τ , the constants are given

by $A = 2k$, $B = -kk''$, and the nonlinear self-interaction constant reads,

$$\begin{aligned} \text{--for } b = \pm a & \quad C = -\frac{1}{2}(D_1 + D_2 + D_3) \\ \text{--for } b = \pm ia & \quad C = -\frac{1}{2}(D_1 + D_2 - D_3) \\ \text{--for } a \text{ or } b = 0 & \quad C = -D_1. \end{aligned}$$

The coefficients D_1 , D_2 and D_3 are given by equations (30) and (31). Note that for the circular polarizations ($b = \pm ia$), the factor proportional to $\hat{\chi}_{xzy}^{(2)}(0, \omega)\hat{\chi}_{zxy}^{(2)}(\omega, -\omega)$ in D_3 , that accounts for optical rectification and the electro-optic effect, vanishes, while it must be taken into account for the linear polarizations. Note also that the linear polarizations that make an angle of $\pm\pi/4$ with the crystallographic axes, or parallel to one of them, are the only ones that can support solitons of the NLS equation. In the case of the polarizations parallel to the axes, the self-modulation of the wave is purely due to the third-order tensor.

• *Spatial (and spatio-temporal) case.* We assume now that the amplitudes depend only on the single variable $X = r\xi + s\eta + d\tau$. If the medium is optically isotropic ($\beta = 1$), as is, for example, GaAs, the NLS equation (47) is obtained for any linear (a, b real) or circular ($b = \pm ia$), but not for elliptical polarization. In the anisotropic case ($\beta \neq 1$), the reduction is possible only if the polarization is linear and makes an angle $\theta = \pm\pi/4$ with the crystallographic axis, and if the direction of the modulation (or of the planar guide) is either parallel or perpendicular to the polarization: $(r, s) = (a, b)$ or $(-b, a)$. The dispersion-diffusion constant of the NLS equation (47) reads $B = \beta - kk''d^2$ in the former case, and $B = 1 - kk''d^2$ in the latter, and for circular polarizations. The nonlinear constant is:

$$C = -\frac{1}{2} \left(D_1 + D_2 + D_3 \pm \frac{2E\lambda}{\alpha + \rho d^2} \right) \tag{49}$$

for linear polarizations $b = \pm a$, and reduces to

$$C = -\frac{1}{2}(D_1 + D_2 - D_3) \tag{50}$$

in the isotropic case $\beta = 1$, for circular polarizations. The purely spatial case, obtained by setting $d = 0$, is not a special case here.

4.2.2. *The 3m symmetry class.*

• *Temporal case.* Now we assume that the $\chi^{(2)}$ -tensor has the structure given by equation (22), which is valid for the 3m class of crystals, and that the amplitudes E_1^1 and E_2^0 do not depend on ξ and η . Equations (44)–(45) are solved to yield

$$E_2^{0,x} = \frac{\lambda_3}{\rho}(UV^* + VU^*) \tag{51}$$

$$E_2^{0,y} = \frac{\lambda_3}{\rho}(|U|^2 - |V|^2). \tag{52}$$

Making use of equations (51), (52), equations (42), (43) reduce to the NLS equation (47) if either $b = \pm a$, or $b = \pm ia$, or $b = 0$, or $a = 0$. The nonlinear constant then reads, for the linear polarizations $((a, b) \propto (1, 1), (1, -1), (1, 0), \text{ or } (0, 1))$,

$$C = - \left(D_1 + \frac{F_1\lambda_3}{\rho} \right) \tag{53}$$

i.e. using the complete symmetry property of the $\chi^{(2)}$ -tensor:

$$C = \frac{2\omega^2}{c^2} \left[\frac{(\hat{\chi}_{yyy}^{(2)}(\omega, \omega))^2}{n_o^2 - n_o^2(2\omega)} - \frac{(\hat{\chi}_{zxx}^{(2)}(\omega, \omega))^2}{n_e^2(2\omega)} + \frac{2(\hat{\chi}_{yyy}^{(2)}(\omega, -\omega))^2}{(n_o + \omega n'_o)^2 - n_o^2(0)} \right]$$

$$-\frac{2(\hat{\chi}_{zxx}^{(2)}(\omega, -\omega))^2}{n_e^2(0)} + \frac{3}{2}\hat{\chi}_{xxxx}^{(3)}(\omega, \omega, -\omega) \Big]. \tag{54}$$

For the circular polarizations $b = \pm ia$, $C = -D_2$, with D_2 given by equation (130).

• *Spatial (and spatio-temporal) case.* We consider now the one-dimensional variable $X = r\xi + s\eta + d\tau$. System (42)–(45) can be reduced to the NLS equation (47) only if

$$\begin{aligned} \theta = -\frac{\pi}{2}, \frac{\pi}{6}, \frac{5\pi}{6} \quad \text{and} \quad & \begin{cases} r = \cos \theta \\ s = \sin \theta \end{cases} \\ \text{or} \quad \theta = 0, \pm\frac{\pi}{3} \quad \text{and} \quad & \begin{cases} r = -\sin \theta \\ s = \cos \theta. \end{cases} \end{aligned} \tag{55}$$

The coefficients then reads,

$$B = \beta - kk''d^2 \quad \text{if } \theta = -\frac{\pi}{2}, \frac{\pi}{6}, \frac{5\pi}{6} \tag{56}$$

$$B = 1 - kk''d^2 \quad \text{if } \theta = 0, \pm\frac{\pi}{3}$$

$$C = -D_1 - F_1 \frac{\lambda_1 + \lambda_2 d + \lambda_3 d^2}{\alpha + \rho d^2} - F_2 \frac{\alpha \lambda_2 + (\alpha \lambda_3 - \rho \lambda_1) d}{\alpha(\alpha + \rho d^2)}. \tag{57}$$

In the purely spatial case $d = 0$, it reduces to

$$C = -D_1 - \frac{F_1 \lambda_1}{\alpha} - F_2 \frac{\lambda_2}{\alpha}. \tag{58}$$

That is:

$$C = \frac{2\omega^2}{c^2} \left[\frac{(\hat{\chi}_{yyy}^{(2)}(\omega, \omega))^2}{n_o^2 - n_o^2(2\omega)} - \frac{(\hat{\chi}_{zxx}^{(2)}(\omega, \omega))^2}{n_e^2(2\omega)} - \frac{2(\hat{\chi}_{yyy}^{(2)}(\omega, -\omega))^2}{n_e^2(0)} + \frac{3}{2}\hat{\chi}_{xxxx}^{(3)}(\omega, \omega, -\omega) \right]. \tag{59}$$

It is remarkable that, among the two terms that describe the optical rectification and electro-optic effect, only the yyy -component of the $\chi^{(2)}$ -tensor has a nonvanishing contribution. If we consider the $4mm$ and $6mm$ classes of crystals, this component is zero, thus the influence of optical rectification completely disappears from the spatial case, although it is non-negligible in the temporal case, and for the $3m$ class.

There exists another case where the reduction is possible, for some very particular value of d . We make no further mention of it because it is of little concrete interest.

4.3. (1 + 1)-dimensional reductions for two interacting polarizations

4.3.1. *The $\bar{4}2m$ class.* We consider the two variables ζ and $X = r\xi + s\eta + d\tau$. We use the normalization condition $r^2 + s^2 = 1$, so that $r = \cos \theta$ and $s = \sin \theta$, except in the purely temporal case: $r = s = 0$. In this latter case we will set $d = 1$. The equation for the quantity Φ that describes the DC rectified field is solved as,

$$\Phi = \frac{\lambda}{\alpha + \rho d^2}(UV^* + VU^*). \tag{60}$$

In the purely temporal case, the solution is $\Phi = 0$. It can be recovered from (60) by taking the limit $d \rightarrow \infty$. The effect of the optical rectification and electro-optic effect

never expresses as a supplementary PDE in (1 + 1) dimensions, in contrast to the (2 + 1)- and (3 + 1)-dimensional cases. The reduction yields the following system of two coupled one-dimensional NLS equations:

$$2ik\partial_\zeta U + B_1\partial_X^2 U + (\beta - 1)rs\partial_X^2 V = D_1U|U|^2 + D_2'U|V|^2 + D_3'V^2U^* \quad (61)$$

$$2ik\partial_\zeta V + B_2\partial_X^2 V + (\beta - 1)rs\partial_X^2 U = D_1V|V|^2 + D_2'V|U|^2 + D_3'U^2V^* \quad (62)$$

with

$$B_1 = \beta r^2 + s^2 - kk''d^2 \quad (63)$$

$$B_2 = r^2 + \beta s^2 - kk''d^2$$

$$D_j' = D_j + \frac{\lambda}{\alpha + \rho d^2} E \quad (64)$$

for $j = 1, 2$. Notice the presence of the nonlinear terms $D_3'V^2U^*$ and $D_3'U^2V^*$, that do not appear in the system describing the propagation of two optical polarizations in a birefringent monodimensional Kerr medium [25]. Furthermore, an off-diagonal differential term due to the anisotropy appears. This term obviously vanishes in the purely temporal case ($r = s = 0$). Otherwise it is removed by the following rotation:

$$\begin{aligned} U' &= rU + sV \\ V' &= -sU + rV. \end{aligned} \quad (65)$$

Then, r times equation (61) plus s times equation (62), and $-s$ times equation (61) plus r times equation (62) yield the following equations for U' and V' respectively:

$$\begin{aligned} 2ik\partial_\zeta U' + (\beta - kk''d^2)\partial_X^2 U' &= D_1''U'|U'|^2 + D_2''U'|V'|^2 + D_3''V'^2U'^* \\ &+ D_4 \left[U'^2V'^* + 2V'|U'|^2 - V'|V'|^2 \right] \end{aligned} \quad (66)$$

$$\begin{aligned} 2ik\partial_\zeta V' + (1 - kk''d^2)\partial_X^2 V' &= D_1''V'|V'|^2 + D_2''V'|U'|^2 + D_3''U'^2V'^* \\ &- D_4 \left[V'^2U'^* + 2U'|V'|^2 - U'|U'|^2 \right]. \end{aligned} \quad (67)$$

The constants are given by

$$\begin{aligned} D_1'' &= D_1 + K \sin^2 2\theta \\ D_2'' &= D_2 + \frac{E\lambda}{\alpha + \rho d^2} - 2K \sin^2 2\theta \end{aligned} \quad (68)$$

$$D_3'' = D_3 + \frac{E\lambda}{\alpha + \rho d^2} - K \sin^2 2\theta$$

$$D_4 = \frac{1}{2} K \sin 4\theta$$

$$K = \frac{E\lambda}{\alpha + \rho d^2} + \frac{1}{2}(D_2 + D_3 - D_1). \quad (69)$$

Notice the appearance of three additional nonlinear terms. They vanish with the coefficient D_4 , that is, when the susceptibilities satisfy $K = 0$, or for the particular choice of the modulation direction: $\theta = 0, \frac{\pi}{4}, \frac{\pi}{2}, \frac{3\pi}{4}$.

The following system, describing the propagation of vector solitons in Kerr birefringent unidimensional media, has been studied by several authors,

$$i\partial_\zeta f + a_1\partial_X^2 f + b_1f|f|^2 + c_1f|g|^2 = 0 \quad (70)$$

$$i\partial_\zeta g + a_2\partial_X^2 g + b_2g|g|^2 + c_2g|f|^2 = 0. \quad (71)$$

Manakov showed [23] that this system is completely integrable by the IST method when

$$a_1 = a_2 \quad b_1 = c_2 \quad b_2 = c_1 \quad \text{and} \quad b_1 b_2 > 0. \quad (72)$$

Zakharov and Schulman showed [24] that (72) is the only integrable case, together with the following:

$$a_1 = -a_2 \quad b_1 = -c_2 \quad b_2 = -c_1 \quad \text{and} \quad b_1 b_2 > 0. \quad (73)$$

Kaup has studied the same system [25] in the more general, but not integrable case where:

$$a_1 = a_2 \quad a_1, b_1, b_2 \text{ have the same sign} \quad \text{and} \quad b_1 c_1 = b_2 c_2. \quad (74)$$

It also describes vector solitons.

System (66), (67) coincides with (70), (71) only if $D_4 = 0$ and $D_3'' = 0$. The former condition can be realized by a convenient choice of the angle θ , and is satisfied in the temporal case. The latter condition is not so easily satisfied. In the purely temporal case, it reduces to $D_3 = 0$, that reads,

$$3\hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega) = \frac{4}{n_e^2(0)} \hat{\chi}_{xzy}^{(2)}(0, \omega) \hat{\chi}_{zxy}^{(2)}(\omega, -\omega). \quad (75)$$

This condition can be interpreted as an equilibrium between some electro-optic and Kerr contributions. It also would be satisfied if both the electro-optic and Kerr effect were negligible, which is not the case in second-order media, far from phase matching. For optically anisotropic media ($\beta \neq 1$), Manakov's integrability case is reached only in the purely temporal case ($X = \tau$), if the following conditions are satisfied:

$$D_3 = 0 \quad \text{and} \quad D_1 = D_2.$$

If the medium is isotropic, the integrability condition is:

$$D_1 + D_3 = D_2 \quad \text{and} \quad D_3 + \frac{E\lambda}{\alpha + \rho d^2} = 0.$$

It holds for any choice of the variable X . The purely temporal case is recovered by setting d to infinity, the condition is then the same as for anisotropic media. That system (66)–(67) belongs to Zakharov–Schulman's integrability case necessitates a normal dispersion ($k'' > 0$), and a particular spatio-temporal variable X , such that,

$$d = \sqrt{\frac{\beta + 1}{2kk''}}.$$

Furthermore, one of the following conditions must be satisfied:

- $\theta = 0, \frac{\pi}{2}, \pi$ $D_1 + D_2 = D_3$ and $D_3 + \frac{E\lambda}{\alpha + \rho d^2} = 0$
- $\theta = \frac{\pi}{4}, \frac{3\pi}{4}$ $D_1 = 0$ $D_2 = D_3$ and $D_3 + \frac{E\lambda}{\alpha + \rho d^2} \neq 0.$

System (66), (67) is the system studied by Kaup in [25] if

$$X = \tau \quad \text{and} \quad D_3 = 0$$

or, if the medium is optically isotropic ($\beta = 1$), if one of the following conditions is satisfied:

- $\theta = 0, \frac{\pi}{2}, \pi$ and $D_3 = \frac{E\lambda}{\alpha + \rho d^2}$
- $\theta = \frac{\pi}{4}, \frac{3\pi}{4}$ and $D_2 = D_1 + D_3$
- $D_2 + \frac{E\lambda}{\alpha + \rho d^2} = D_1$ and $D_3 + \frac{E\lambda}{\alpha + \rho d^2} = 0$.

Furthermore, $1 - kk''d^2$ and $D_1'' = D_1 + K \sin^2 2\theta$ must have the same sign. This condition writes $-k''D_1 > 0$ in the temporal case. The appearance of particular values of the angle θ in the case called ‘isotropic’ comes from the anisotropy of the nonlinear part, that still exists when the linear part is isotropic. System (66), (67) that describes the propagation of two interacting polarizations in a crystal of the $\bar{4}2m$ class with a nonvanishing $\chi^{(2)}$ is very rarely completely integrable, and does not often have the same form as the system that describes the same physical problem in Kerr media.

4.3.2. *The 3m class.* An integrability case of Manakov’s type is obtained in the purely temporal case $X = \tau$, if:

$$D_3 + \frac{F_1\lambda_3}{\rho} = 0 \quad \text{and} \quad D_2 = D_1 + \frac{F_1\lambda_3}{\rho}. \tag{76}$$

For a purely spatial variable $X = \cos \theta \xi + \sin \theta \eta$, both Kaup’s and Manakov systems require optical isotropy ($\beta = 1$). Equations analogous to equations (66), (67) are obtained, with complicated coefficients. The system (66), (67) with $D_4 = 0$ is obtained for three special values of the angle θ :

$$\theta = 0 \quad \text{and} \quad \cos 2\theta = \pm \frac{1}{8} \sqrt{16 + \frac{\alpha^2(D_1 - D_2 - D_3)^2}{F_1^2\lambda_1^2}} + \frac{\alpha(D_1 - D_2 - D_3)}{F_1\lambda_1}.$$

The system then has Kaup’s form if $D_3'' = 0$, that writes

$$D_3 + \frac{\lambda_1 F_1}{\alpha} = 0$$

in the former case, and

$$\frac{3D_1 - 3D_2 + 5D_3}{8} + \frac{\lambda_1 F_1}{2\alpha} \mp \frac{1}{8} \sqrt{(D_1 - D_2 - D_3)^2 + \frac{16\lambda_1^2 F_1^2}{\alpha^2}} = 0 \tag{77}$$

in the latter cases. The system is integrable if the following additional condition is satisfied:

$$D_2 = D_1 + D_3 \tag{78}$$

for $\theta = 0$, and

$$\frac{-D_1 + D_2 + 9D_3}{8} + \frac{\lambda_1 F_1}{2\alpha} \pm \frac{3}{8} \sqrt{(D_1 - D_2 - D_3)^2 + \frac{16\lambda_1^2 F_1^2}{\alpha^2}} = 0 \tag{79}$$

for the other values of the angle θ . In the particular case where $D_1 = D_2 + D_3$, this value is $\pi/6$, and the conditions on the coefficients reduce to $D_3 = 0$ to obtain Kaup’s system,

and $F_1\lambda_1 = 0$ for the integrability. There is no integrable case of Manakov's type for a spatio-temporal variable.

On the other hand, it has been seen in the case of the $\bar{4}2m$ class that the integrable system of Zakharov and Schulman can be reached only for a particular spatio-temporal variable. This holds for the $3m$ class too. However, as written above, the reduction of the nonlinear part of the equations to that of system (61), (62) is not possible for spatio-temporal variables. Thus there is no integrability case of Zakharov–Schulman's type for the $3m$ class.

4.3.3. *A system describing the interaction of two polarizations.* In summary, the integrability of the model describing the propagation of two interacting polarizations in $(1 + 1)$ dimensions is very rare. This model coincides very rarely with the model that describes the same physical problem in Kerr media, studied by Kaup. The obtained equations are rather complicated in the general case, but for a temporal variable, or some particular choice of the spatial variable, they reduce to the following relatively simple system:

$$2ik\partial_\zeta U + B_1\partial_X^2 U = D_1U|U|^2 + D_2U|V|^2 + D_3V^2U^* \tag{80}$$

$$2ik\partial_\zeta V + B_2\partial_X^2 V = D_1V|V|^2 + D_2V|U|^2 + D_3U^2V^* \tag{81}$$

where the B_j and D_j are reals constants, depending on the specific case. System (80), (81) can be derived, using equation (39), from the following Hamiltonian density:

$$\mathcal{H} = B_1U_XU_X^* + B_2V_XV_X^* + \frac{D_1}{2}(|U|^4 + |V|^4) + D_2|U|^2|V|^2 + \frac{D_3}{2}(V^2U^{*2} + U^2V^{*2}). \tag{82}$$

For a spatial variable X , if the anisotropy is not negligible ($B_1 \neq B_2$), if B_1, B_2 and $(-D_1)$ have the same sign, and if the additional condition $D_2 = \pm D_3$ is satisfied, the system (80), (81) possesses the following particular solution:

$$U = \frac{U_0e^{i\psi}}{\cosh \Omega} \tag{83}$$

$$V = \frac{V_0e^{i\psi}}{\cosh \Omega'} \tag{84}$$

with

$$\psi = pX + q\zeta. \tag{85}$$

p, q are real with $p^2 > \frac{-2k}{B_1}q, \frac{-2k}{B_2}q$,

$$\Omega = \sqrt{p^2 + \frac{2k}{B_1}q} \left(X - \frac{B_1}{k}p\zeta \right) \tag{86}$$

$$\Omega' = \sqrt{p^2 + \frac{2k}{B_2}q} \left(X - \frac{B_2}{k}p\zeta \right) \tag{87}$$

$$|U_0|^2 = \frac{-2B_1}{D_1} \left(p^2 + \frac{2k}{B_1}q \right) \tag{88}$$

$$|V_0|^2 = \frac{-2B_2}{D_1} \left(p^2 + \frac{2k}{B_2}q \right) \tag{89}$$

and $U_0V_0^*$ purely imaginary if $D_2 = +D_3$, or real if $D_2 = -D_3$. This particular solution corresponds to two solitons propagating simultaneously without seeing each other.

In the temporal, and in the spatial isotropic ($B_1 = B_2 = B$) cases, a vector soliton solution can be written. It has a linear or circular polarization defined by $V = \pm U$, or $V = \pm iU$ respectively. U expresses as in equations (83), (85) and (86) above, with

$$|U_0|^2 = \frac{-2(Bp^2 + 2kq)}{D_1 + D_2 \pm D_3}. \quad (90)$$

The sign $+$ is valid when $V = \pm U$ and the sign $-$ when $V = \pm iU$. Due to expressions (86) and (90), the sign conditions $B(D_1 + D_2 \pm D_3) < 0$ and $p^2 + 2kq/B > 0$ must be satisfied. Each of these solutions satisfy one the NLS equations for a single polarization derived in section 4.2. An elliptically polarized soliton solution of system (80), (81), not described by a single NLS equation, can be written if the following additional condition is satisfied:

$$D_1 = D_2 + \epsilon D_3 \quad \text{with} \quad \epsilon = \pm 1. \quad (91)$$

This solution writes

$$\begin{aligned} U &= \frac{U_0 e^{i\psi}}{\cosh \Omega} & V &= \frac{\pm V_0 e^{i\psi}}{\cosh \Omega} & \text{for } \epsilon = +1 \\ \text{or} & & V &= \frac{\pm i V_0 e^{i\psi}}{\cosh \Omega} & \text{for } \epsilon = +1 \end{aligned} \quad (92)$$

ψ and Ω are as previously given by equations (85) and (86). U_0 and V_0 are positive real numbers related by

$$U_0^2 + V_0^2 = \frac{-2(Bp^2 + 2kq)}{D_1}. \quad (93)$$

Note that BD_1 must be negative. These particular solutions generalize the integrable case. The existence of other analytic solutions, as well as an analytical or numerical study of system (80), (81), are left for further investigation.

5. Conclusion

We derived the equations that describe the evolution of the modulation of a short localized optical pulse in a bulk medium presenting a nonzero second-order nonlinear susceptibility, far from the the phase matching corresponding to the second harmonic generation. The equations have been given in the general case, and for the particular symmetry classes of crystals $42m$ and $3m$. The nonlinear effects have three different physical origins: the Kerr effect, the cascading, i.e. the second harmonic generation, and back-conversion to the fundamental, and the combination of optical rectification and electro-optic effect. Far from phase matching, the amplitude, time- and spacescales are the same for these three effects. However, while the cascading operates simply as an effective Kerr nonlinearity, a proper wave interaction occurs through optical rectification and the electro-optic effect, between the wave and some DC field, in fact a solitary wave with a typical length of the same order of magnitude as the pulse length.

We thus obtain a new $(3 + 1)$ -dimensional model, for which no stability result exists at this time. In the absence of a mathematical study, a numerical resolution of the derived model would show if stable localized pulses can be expected. They exist at least for some particular $(2 + 1)$ -dimensional reduction of the system.

The obtained model can be reduced to the completely integrable (1+1)-dimensional NLS equation; but not so easily as expected at first glance. The reduction describing temporal solitons is possible for only a few polarizations: circular, or linear, with a polarization direction parallel to one of the crystallographic axes, or making a $\pi/4$ angle with it, while the reduction describing spatial solitons is possible only for some special choices of the polarization and of the modulation direction (that is the orientation of the waveguide). The polarization must be either orthogonal or parallel to the waveguide and the orientation of the guide corresponds to the symmetry of the crystal; for the $42m$ symmetry class, which is tetragonal, it must make an angle of $\pm\pi/4$ with the crystallographic axes, and in the case of $3m$ class, which is trigonal, an angle of $\pm\pi/3$. The expression for the nonlinear coefficient in the NLS equation is not the same in all cases. As an example, the effect of optical rectification and electro-optic effect disappears for circular polarization, although it is essential in other cases. Spatio-temporal reductions to NLS have also been given, in order to make an exhaustive list of the completely integrable reductions of the system.

The ability of reducing this system to two coupled NLS equations, describing the evolution of the modulation of two coupled polarizations, has also been investigated. The integrable models of Manakov or Zakharov and Schulman are reached only if some restrictive conditions on the coefficients are satisfied. An alternative model has been given, for which a few particular analytic solutions can be written. On the other hand, the reduction to the completely integrable (2 + 1)-dimensional DS system have been achieved; its publication is in preparation [20].

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Appendix

In this appendix, we give some technical details of the perturbative expansions.

A.1. The treatment of the convolutions

The vectors \mathbf{D} and \mathbf{P} are given by the convolution products (2) and (3). Let us examine how they expand in a power series of ε . First we make the change of integration variable $t' = t - t_1$ in (2), then we expand \mathbf{E} as in (5), to obtain

$$\mathbf{D} = \chi^{(1)} * \mathbf{E} = \sum_{l \geq 1, p \in \mathbb{Z}} \int_0^\infty dt' \chi^{(1)}(t') \varepsilon^l \mathbf{E}_l^p \left(\varepsilon \left(t - t' - \frac{z}{V} \right), \varepsilon x, \varepsilon y, \varepsilon^2 z \right) e^{ip(kz - \omega(t-t'))}. \quad (94)$$

The argument of the amplitude function \mathbf{E}_l^p can be rewritten as $(\tau - \varepsilon t', \xi, \eta, \zeta)$. The slow variables τ, ξ, η, ζ and the integration variable t' are assumed to have the order of magnitude of unity, and we can expand \mathbf{E}_l^p in a Taylor series. Then \mathbf{D} reduces to

$$\mathbf{D} = \sum_{l \geq 1, p \in \mathbb{Z}, j \geq 0} \frac{\varepsilon^{l+j} (-1)^j}{j!} e^{ip\varphi} \left(\int_0^\infty dt' t'^j \chi^{(1)}(t') e^{ip\omega t'} \right) \partial_\tau^j \mathbf{E}_l^p. \quad (95)$$

The integral coefficient is, apart from a multiplicative constant, the j th ω' -derivative $\chi^{(1,j)}$ of the Fourier transform of $\chi^{(1)}$:

$$\hat{\chi}^{(1)}(\omega') = \int_0^\infty dt' \chi^{(1)}(t') e^{ip\omega't'}. \tag{96}$$

Thus,

$$D = \sum_{l \geq 1, p \in \mathbb{Z}} \varepsilon^l e^{ip\varphi} D_l^p \tag{97}$$

with

$$D_l^p = \sum_{j=0}^l \frac{i^j}{j!} \hat{\chi}^{(1,j)}(p\omega) \partial_\tau^j E_{l-j}^p. \tag{98}$$

Note that expression (98) is valid for any tensor $\chi^{(1)}$, not only a diagonal one.

A similar proof gives for P ,

$$P = \sum_{l \geq 1, p \in \mathbb{Z}} \varepsilon^l e^{ip\varphi} P_l^p \tag{99}$$

with

$$P_l^p = P_l^{(2)p} + P_l^{(3)p}. \tag{100}$$

The contribution $P_l^{(2)p}$ due to the second-order susceptibility reads,

$$P_l^{(2)p} = \sum_{\substack{j_1, j_2 \geq 0, l_1, l_2 \geq 1 \\ p_1, p_2 \in \mathbb{Z} \\ j_1 + j_2 + l_1 + l_2 = l \\ p_1 + p_2 = p}} \frac{i^{j_1 + j_2}}{j_1! j_2!} \hat{\chi}^{(2, j_1, j_2)}(p_1\omega, p_2\omega) \partial_\tau^{j_1} E_{l_1}^{p_1} \partial_\tau^{j_2} E_{l_2}^{p_2} \tag{101}$$

where:

$$\hat{\chi}^{(2, j_1, j_2)}(\omega_1, \omega_2) = \frac{\partial^{j_1 + j_2}}{\partial \omega_1^{j_1} \partial \omega_2^{j_2}} \hat{\chi}^{(2)}(\omega_1, \omega_2) \tag{102}$$

is the partial derivative of the two-dimensional Fourier transform $\hat{\chi}^{(2)}$ of $\chi^{(2)}$:

$$\hat{\chi}^{(2)}(\omega_1, \omega_2) = \int_0^\infty dt_1 \int_0^\infty dt_2 \chi^{(2)}(t_1, t_2) e^{i(\omega_1 t_1 + \omega_2 t_2)}. \tag{103}$$

The contribution $P_l^{(3)p}$ due to the third-order susceptibility has an expression analogous to equation (101), but with three indices of each type j_s, l_s, p_s , and three fields. It uses the three-dimensional Fourier transform $\hat{\chi}^{(3)}$ of $\chi^{(3)}$:

$$\hat{\chi}^{(3)}(\omega_1, \omega_2, \omega_3) = \int_0^\infty dt_1 \int_0^\infty dt_2 \int_0^\infty dt_3 \chi^{(3)}(t_1, t_2, t_3) e^{i(\omega_1 t_1 + \omega_2 t_2 + \omega_3 t_3)}. \tag{104}$$

A.2. The equations for the zero harmonic

The equations of the multiscale expansion, written at order ε^4 , for the zero harmonic ($p = 0$), read,

$$[\partial_\eta^2 + \rho \partial_\tau^2] E_2^{0,x} - \partial_\eta \partial_\xi E_2^{0,y} + \frac{1}{v} \partial_\xi \partial_\tau E_2^{0,z} = \frac{1}{c^2} \partial_\tau^2 P_2^{0,x} \tag{105}$$

$$[\partial_\xi^2 + \rho \partial_\tau^2] E_2^{0,y} - \partial_\eta \partial_\xi E_2^{0,x} + \frac{1}{v} \partial_\eta \partial_\tau E_2^{0,z} = \frac{1}{c^2} \partial_\tau^2 P_2^{0,y} \tag{106}$$

$$\left[\partial_\xi^2 + \partial_\eta^2 - \frac{n_e^2(0)}{c^2} \partial_\tau^2 \right] E_2^{0,z} + \frac{1}{v} \partial_\tau (\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) = \frac{1}{c^2} \partial_\tau^2 P_2^{0,z}. \tag{107}$$

The constant ρ is given by equation (15) and the second-order polarization term P_2^0 by equation (17). Taking the derivative of equations (105), (106) and (107) with respect to ξ , η and τ respectively, multiplying the third equation by $-1/v$ and summing up, we can integrate to compute $E_2^{0,z}$:

$$E_2^{0,z} = \int^\tau \frac{v}{n_e^2(0)} \left[n_o^2(0)(\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) + \partial_\xi P_2^{0,x} + \partial_\eta P_2^{0,y} - \frac{1}{v} \partial_\tau P_2^{0,z} \right] \quad (108)$$

where the notation $\int^\tau f$ designates some primitive of f with respect to τ , vanishing at infinity. Making use of equation (108) with equations (105) and (106), we find the evolution equation for $E_2^{0,x}$ and $E_2^{0,y}$ (equations (13) and (14)).

A.3. The (3 + 1)-dimensional evolution system for the $\bar{4}2m$ class

Using the structure (21) of the $\chi^{(2)}$ -tensor in the present case, we can compute the nonlinear polarization terms. The term that yields the second harmonic reads

$$P_2^2 = 2\hat{\chi}_{zxy}^{(2)}(\omega, \omega) E_1^{1,x} E_1^{1,y} \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad (109)$$

while the term that produces the zero harmonic is

$$P_2^0 = 2\hat{\chi}_{zxy}^{(2)}(\omega, -\omega)(E_1^{1,x} E_1^{1,y,*} + E_1^{1,x,*} E_1^{1,y}) \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \quad (110)$$

The ‘cascaded’ term $P_3^{(2)1}$ reads

$$P_3^{(2)1,x} = 2[\hat{\chi}_{xzy}^{(2)}(2\omega, -\omega) E_2^{2,z} E_1^{1,y,*} + \hat{\chi}_{xzy}^{(2)}(0, \omega) E_2^{0,z} E_1^{1,y}] \quad (111)$$

$$P_3^{(2)1,y} = 2[\hat{\chi}_{xzy}^{(2)}(2\omega, -\omega) E_2^{2,z} E_1^{1,x,*} + \hat{\chi}_{xzy}^{(2)}(0, \omega) E_2^{0,z} E_1^{1,x}]. \quad (112)$$

The Kerr term $P_3^{(3)1}$ reads

$$P_3^{(3)1,x} = 3[\hat{\chi}_{xxx}^{(3)}(\omega, \omega, -\omega) E_1^{1,x} |E_1^{1,x}|^2 + (\hat{\chi}_{xxyy}^{(3)}(\omega, \omega, -\omega) + \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega)) \times E_1^{1,y} |E_1^{1,y}|^2 + \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega) (E_1^{1,y})^2 E_1^{1,x,*}] \quad (113)$$

$$P_3^{(3)1,y} = 3[\hat{\chi}_{xxx}^{(3)}(\omega, \omega, -\omega) E_1^{1,y} |E_1^{1,y}|^2 + (\hat{\chi}_{xxyy}^{(3)}(\omega, \omega, -\omega) + \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega)) \times E_1^{1,x} |E_1^{1,x}|^2 + \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega) (E_1^{1,x})^2 E_1^{1,y,*}]. \quad (114)$$

Equations (18), (19), (13), (14) yield respectively,

$$[2ik\partial_\zeta + \beta\partial_\xi^2 + \partial_\eta^2 - kk''\partial_\tau^2] E_1^{1,x} + (\beta - 1)\partial_\xi\partial_\eta E_1^{1,y} = D_1 E_1^{1,x} |E_1^{1,x}|^2 + D_2 E_1^{1,x} |E_1^{1,y}|^2 + D_3 (E_1^{1,y})^2 E_1^{1,x,*} + E E_1^{1,y} \int^\tau (\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) \quad (115)$$

$$[2ik\partial_\zeta + \partial_\xi^2 + \beta\partial_\eta^2 - kk''\partial_\tau^2] E_1^{1,y} + (\beta - 1)\partial_\xi\partial_\eta E_1^{1,x} = D_1 E_1^{1,y} |E_1^{1,y}|^2 + D_2 E_1^{1,y} |E_1^{1,x}|^2 + D_3 (E_1^{1,x})^2 E_1^{1,y,*} + E E_1^{1,x} \int^\tau (\partial_\xi E_2^{0,x} + \partial_\eta E_2^{0,y}) \quad (116)$$

$$[\alpha\partial_\xi^2 + \partial_\eta^2 + \rho\partial_\tau^2] E_2^{0,x} + (\alpha - 1)\partial_\xi\partial_\eta E_2^{0,y} = \lambda\partial_\xi\partial_\tau (E_1^{1,x} E_1^{1,y,*} + E_1^{1,x,*} E_1^{1,y}) \quad (117)$$

$$[\partial_\xi^2 + \alpha\partial_\eta^2 + \rho\partial_\tau^2] E_2^{0,y} + (\alpha - 1)\partial_\xi\partial_\eta E_2^{0,x} = \lambda\partial_\eta\partial_\tau (E_1^{1,x} E_1^{1,y,*} + E_1^{1,x,*} E_1^{1,y}). \quad (118)$$

The constants are given by equations (30)–(33). In equations (115), (116), we see that the zero harmonic intervenes only through the combination

$$\Phi = \int^{\tau} (\partial_{\xi} E_2^{0,x} + \partial_{\eta} E_2^{0,y}). \quad (119)$$

A single evolution equation for Φ is found in the following way: we take the derivative of equation (117) with respect to ξ , and of equation (118) with respect to η , and sum up. After some cancellations, we can integrate with respect to τ and obtain equation (28). This way, the evolution system (26)–(28) is obtained.

A.4. Evolution equations for the 3m class

Now we examine the case of the 3m class of crystals, in which the structure of the $\chi^{(2)}$ -tensor is given by equation (22). At order ε^2 , the second harmonic reads

$$E_2^{2,x} = 2\kappa_2 UV \quad (120)$$

$$E_2^{2,y} = 2\kappa_2(U^2 - V^2) \quad (121)$$

$$E_2^{2,z} = 2\kappa_2(U^2 + V^2) \quad (122)$$

where we use the simplified notation $E_1^{1,x} = U$, $E_1^{1,y} = V$, and the constants κ_2 , κ_3 are defined as

$$\kappa_2 = \frac{-\hat{\chi}_{yyy}^{(2)}(\omega, \omega)}{n_o^2 - n_o^2(2\omega)} \quad (123)$$

$$\kappa_3 = \frac{-\hat{\chi}_{zxx}^{(2)}(\omega, \omega)}{n_e^2(2\omega)}. \quad (124)$$

The polarization term responsible for the zero harmonic is

$$P_2^{0,x} = -2\hat{\chi}_{yyy}^{(2)}(\omega, -\omega)(UV^* + VU^*) \quad (125)$$

$$P_2^{0,y} = -2\hat{\chi}_{yyy}^{(2)}(\omega, -\omega)(|U|^2 - |V|^2) \quad (126)$$

$$P_2^{0,z} = 2\hat{\chi}_{zxx}^{(2)}(\omega, -\omega)(|U|^2 + |V|^2). \quad (127)$$

The r.h.s. of equation (18) is computed by substituting these expressions, and the expression (108) of $E_2^{0,z}$, in the expression of the polarization at the order ε^3 . The ‘cascaded’ part reads,

$$P_3^{(2)1,x} = 2[\hat{\chi}_{xzx}^{(2)}(2\omega, -\omega)E_2^{2,z}E_1^{1,x,*} - \hat{\chi}_{yyy}^{(2)}(2\omega, -\omega)(E_2^{2,x}E_1^{1,y,*} + E_2^{2,y}E_1^{1,x,*}) + \hat{\chi}_{xzx}^{(2)}(0, \omega)E_2^{0,z}E_1^{1,x} - \hat{\chi}_{yyy}^{(2)}(0, \omega)(E_2^{0,x}E_1^{1,y} + E_2^{0,y}E_1^{1,x})]. \quad (128)$$

The y component has a symmetrical expression, and the Kerr part $P_3^{(3)1,s}$ ($s = x, y$) has the same expression as for the $\bar{4}2m$ class (equations (113), (114)). After some algebra, we obtain equation (42), and in the same way, equations (43)–(45). The constants are given by the following formulae:

$$D_1 = K_1 + K_2 + K_3 - \frac{3\omega^2}{c^2} \hat{\chi}_{xxx}^{(3)}(\omega, \omega, -\omega) \quad (129)$$

$$D_2 = 2K_2 + K_3 - \frac{3\omega^2}{c^2} (\hat{\chi}_{xxy}^{(3)}(\omega, \omega, -\omega) + \hat{\chi}_{xyy}^{(3)}(\omega, \omega, -\omega)) \quad (130)$$

$$D_3 = K_1 - K_2 - \frac{3\omega^2}{c^2} \hat{\chi}_{xyyx}^{(3)}(\omega, \omega, -\omega) \quad (131)$$

$$K_1 = \frac{-2\omega^2}{c^2} \hat{\chi}_{xzx}^{(2)}(2\omega, -\omega) \kappa_3 \quad (132)$$

$$K_2 = \frac{2\omega^2}{c^2} \hat{\chi}_{yyy}^{(2)}(2\omega, -\omega) \kappa_2 \quad (133)$$

$$K_1 = \frac{4\omega^2}{c^2 n_e^2(0)} \hat{\chi}_{xzx}^{(2)}(0, \omega) \hat{\chi}_{zxx}^{(2)}(\omega, -\omega) \quad (134)$$

$$F_1 = \frac{2\omega^2}{c^2} \hat{\chi}_{yyy}^{(2)}(0, \omega) \quad (135)$$

$$F_2 = \frac{-2\omega^2}{c^2} \frac{v n_o^2(0)}{n_e^2(0)} \hat{\chi}_{xzx}^{(2)}(0, \omega) \quad (136)$$

$$F_3 = \frac{4\omega^2}{c^2} \frac{v}{n_e^2(0)} \hat{\chi}_{xzx}^{(2)}(0, \omega) \hat{\chi}_{yyy}^{(2)}(\omega, -\omega) \quad (137)$$

$$\lambda_1 = \frac{2}{n_e^2(0)} \hat{\chi}_{yyy}^{(2)}(\omega, -\omega) \quad (138)$$

$$\lambda_2 = \frac{2}{v n_e^2(0)} \hat{\chi}_{zxx}^{(2)}(\omega, -\omega) \quad (139)$$

$$\lambda_3 = \frac{-2}{c^2} \hat{\chi}_{yyy}^{(2)}(\omega, -\omega) = \frac{-n_e^2(0)}{c^2} \lambda_1. \quad (140)$$

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