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# Direct derivation of a macroscopic NLS equation from the quantum theory

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## Abstract

The use of the matrix density formalism, and of a multiscale expansion, allows the derivation of a macroscopic nonlinear evolution equation for a short light pulse (a nonlinear Schrödinger equation), directly from the microscopic quantum mechanics equations.

We consider the simple case of a monochromatic plane wave, interacting with independent two-level atoms, to show that such a computation is possible. For their linear part, the results agree with that of the linear dispersion theory, but the obtained nonlinear coefficient differs appreciably from that derived from the computation of the so-called nonlinear susceptibilities, except in one simple particular situation.

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## 1. Introduction

Many experimental and theoretical works in nonlinear optics make use of the so-called 'nonlinear susceptibilities', that are mainly an expansion of a phenomenological response function in a power series of the electric field. Although it has given a very useful theoretical frame for the interpretation of many phenomena, and many quantitative measurements of the nonlinear properties of optical materials, this approach is not completely satisfactory from either the theoretical or the experimental viewpoints. Indeed, it is well known by researchers that the experimental values of the susceptibilities depend strongly on the experimental procedure, even if the theory states that they should not.

In other domains of nonlinear physics, such as hydrodynamics or the physics of waves in ferromagnetic media, the use of such susceptibilities has been avoided, the equations giving the evolution of a wavepacket are obtained directly from the basic equations of the Navier–Stokes or Maxwell–Landau type. The traditional frame for studying a water wave is the multiscale analysis, allowing the treatment of multi-dimensional problems, concerning not only wavepackets [1], but also solitary waves [2]. A Hamiltonian approach has given good results in the study of magnetostatic waves in magnetic films [3, 4]. Electromagnetic waves in magnetic media have also been studied using multiscale analysis [5–7]. To my knowledge, such an approach has never been attempted in nonlinear optics. The reason for this seems to

stem from the great complexity of the basic equations in this case. Indeed, as can be seen from the existing computations of the nonlinear susceptibilities [8, chapter 3], a quantum mechanical treatment seems to be necessary, which implies technical difficulties.

We will see that the use of the density matrix formalism, and of multiscale analysis, allows the direct derivation of a macroscopic nonlinear evolution equation for the light pulse, from a microscopic quantum mechanical model. For the sake of simplicity, we restrict the problem to the interaction between a plane wave and a range of independent identical two-level atoms. The aim of the paper is to give a new derivation method. The results are compared to those derived using nonlinear susceptibilities. We will conclude by discussing the possibility of applying this method to more realistic physical situations, in order to get a more precise description of the experimental features.

## 2. The Maxwell–Bloch equations

We consider an homogeneous medium, in which the dynamics of each atom is described by a two-level Hamiltonian:

$$H_0 = \hbar \begin{pmatrix} \omega_a & 0 \\ 0 & \omega_b \end{pmatrix}. \quad (1)$$

The atomic dipolar electric momentum is described by the operator  $\vec{\mu}$ , with

$$\mu_s = \begin{pmatrix} 0 & \mu_s \\ \bar{\mu}_s & 0 \end{pmatrix} \quad (s = x, y, z). \quad (2)$$

We denote the complex conjugate by an over bar, keeping the star for the Hermitian conjugate (so that  $(a_{ij})^* = (\bar{a}_{ji})$  for any matrix  $(a_{ij})$ ), as per the current notation found in mathematical papers. The electric field  $\vec{E}$  is governed by the Maxwell equations, that reduce to

$$\vec{\nabla}(\vec{\nabla} \cdot \vec{E}) - \Delta \vec{E} = \frac{-1}{c^2} \partial_t^2 (\vec{E} + 4\pi \vec{P}) \quad (3)$$

in the absence of magnetic effects.  $\vec{P}$  is the polarization density,  $c$  the velocity of light in a vacuum. We denote by  $\partial_t$  the derivative operator  $\frac{\partial}{\partial t}$  with regard to the time variable  $t$ , and  $\vec{\nabla}$  is the three-dimensional gradient operator. The coupling between the atoms and the electric field is taken into account by a coupling energy term in the total Hamiltonian  $H$ , that reads

$$H = H_0 - \vec{\mu} \cdot \vec{E} \quad (4)$$

and by the expression of  $\vec{P}$ :

$$\vec{P} = N \text{tr}(\rho \vec{\mu}) \quad (5)$$

where  $N$  is the number of atoms per volume unit. If  $\rho$  is the density matrix, the Schrödinger equation takes the form

$$i\hbar \partial_t \rho = [H, \rho]. \quad (6)$$

The set of equations (3)–(6) is sometimes called the Maxwell–Bloch equations, although this name usually denotes a reduction of this set of equations (see [8, section 5.4]).

The electric field will describe some quasi-monochromatic plane wave, slowly modulated along its propagation direction so that it yields a ‘temporal’ wavepacket, that propagates over distances which are very large with respect to its length. It can thus be written

$$\vec{E} = \sum_{n \geq 1, p \in Z} \varepsilon^n e^{ip\varphi} \vec{E}_n^p \quad (7)$$

where the  $\vec{E}_n^p$  are functions of the slow variables

$$\begin{cases} \tau = \varepsilon \left( t - \frac{z}{V} \right) \\ \zeta = \varepsilon^2 z \end{cases} \quad (8)$$

$\varepsilon$  is a small perturbative parameter,  $V$  is some velocity to be determined, which is actually the group velocity of the wave.  $\varphi$  is some fundamental phase  $\varphi = kz - \omega t$ . Thus the propagation direction is chosen to be the  $z$ -axis, and the problem is purely one dimensional.

We assume that the dominant term in (7) is

$$\vec{E} = \varepsilon (\vec{E}_1^1 e^{i\varphi} + \text{c.c.}) \quad (9)$$

(where c.c. denotes the complex conjugate), so that the incident wave contains only the frequency  $\omega/2\pi$  and its sidebands due to the finite pulse length. The scaling given by (7)–(9) is the one that is commonly used for the derivation of the nonlinear Schrödinger (NLS) equation in any frame [9, section 8.1]. The electric field is of order  $\varepsilon$ , thus ‘small’. When using phenomenological response functions, it is rather difficult to give a reference point for this smallness. From another viewpoint, the wave fields needed for nonlinear experiments are even very large, at least with regard to the fields attainable without using laser sources. In the present context, the field compares to the intra-atomic electric field, or more precisely, the electrostatic energy of the atomic dipole in the field compares to the difference  $\hbar\Omega = \hbar(\omega_b - \omega_a)$  between the energies of the atomic levels (we assume  $\omega_b > \omega_a$ ). The zero order in the space scales is that of the wavelength, which may seem strange for a theory written at the atomic scale (less than a nanometre), even though it concerns optical wavelengths (about a micrometre). But, considered from the energetic point of view, we must compare the energy  $\hbar\omega$  of a wave photon to the energy difference between the atomic levels  $\hbar\Omega$ . It is well known that only the transitions corresponding to frequencies of the same order of magnitude as the wave frequency appreciably affect the wave propagation. The energy scale fixes the frequency scale, then the timescale is that of the corresponding period, and the length scale can be deduced using the value of the light velocity.

The polarization density  $\vec{P}$  and the density matrix  $\rho$  are expanded in the same way as (7), except that  $\rho$  has a  $\varepsilon^0$  order term  $\rho_0$  that accounts for the initial state of the atoms. They are initially assumed to all be in their fundamental state  $a$ , so that  $\rho_0 = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$ . A population inversion is prohibited by this choice, but could be taken into account through a convenient modification of the ansatz. Recall that the density matrix accounts for the statistical distribution of the physical state of a large number of identical independent atoms among all their possible quantum states. This classical probability for some atom to be in some quantum state varies with space and time: its variations have the same space and time scales as those of the electric field. It varies spatially at distances which are very large with respect to the atomic scale. This ensures the validity of the statistical treatment yielded by the use of the density matrix formalism. The fact that the wavelength is very large with regard to the atomic scale also allows the use of the electric dipolar approximation, that justifies equations (4) and (5).

### 3. The resolution of the perturbative scheme

The expansion (7) is imported into the equations (3)–(5), and the coefficient of each power of  $\varepsilon$  equated. We denote the components of  $\rho$  by

$$\rho_n^p = \begin{pmatrix} \rho_{n,a}^p & \rho_{n,t}^p \\ \rho_{n,u}^p & \rho_{n,b}^p \end{pmatrix}. \quad (10)$$

Expression (5) for  $\vec{P}$  simply yields

$$P_n^{p,s} = N (\rho_{n,u}^p \mu^s + \rho_{n,t}^p \bar{\mu}^s) \quad (11)$$

for all  $n$ ,  $p$ , and  $s = x, y, z$ .

### 3.1. Order 0

In addition, we implicitly assumed that  $\rho_0^p = 0$  for any nonzero  $p$ . It is easily checked that this condition is necessary, except eventually, for the coherences  $\rho_{0,t}^p$  and  $\rho_{0,u}^p$ , if the corresponding harmonic of the incident wave oscillates at the exact resonance frequency of the atom  $p\omega = \pm\Omega$ . We assume that no such resonance exist in the present case.

### 3.2. Order 1

Because of the first order choice (9), the term of interest is  $p = 1$ . The Maxwell equation (3) at order  $\varepsilon^1$  for this term yields

$$\begin{aligned} E_1^{1,s} &= -\frac{4\pi}{\beta} P_1^{1,s} \quad \text{for } s = x, y \\ E_1^{1,z} &= -4\pi P_1^{1,z} \end{aligned} \quad (12)$$

with  $\beta = 1 - \frac{k^2 c^2}{\omega^2}$ . The Schrödinger equation (6) shows that the corrections to the populations at this order,  $\rho_{1,a}^1$  and  $\rho_{1,b}^1$ , are zero. The coherences are coupled with the electric field through

$$\hbar\omega\rho_{1,t}^1 = -\hbar\Omega\rho_{1,t}^1 + \sum_{s=x,y,z} \mu_s E_1^{1,s}. \quad (13)$$

Making use of equation (12) in (13) and in the similar equation for  $\rho_{1,u}^1$ , the following linear system is obtained:

$$\mathcal{L} \begin{pmatrix} \rho_{1,t}^1 \\ \rho_{1,u}^1 \end{pmatrix} = \begin{pmatrix} \hbar(\Omega + \omega) + NQ & NK \\ N\bar{K} & \hbar(\Omega - \omega) + NQ \end{pmatrix} \begin{pmatrix} \rho_{1,t}^1 \\ \rho_{1,u}^1 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \quad (14)$$

where

$$K = 4\pi \left( \frac{1}{\beta} (\mu_x^2 + \mu_y^2) + \mu_z^2 \right) \quad (15)$$

$$Q = 4\pi \left( \frac{1}{\beta} (|\mu_x|^2 + |\mu_y|^2) + |\mu_z|^2 \right). \quad (16)$$

Notice that  $(\rho_{1,t}^1, \rho_{1,u}^1) = (0, 0)$  solves equation (14). This yields a solution of the propagation problem in which the atomic dipoles are not excited by the wave. This is the case when the wave polarization is perpendicular to the direction of the polarization density defined by  $\vec{\mu}$  (assuming that the operator  $\vec{\mu}$ , in the two-level model, describes transverse linear oscillations of the atomic dipoles). Here we will only consider the waves that excite the atomic dipoles, e.g. for which the coherences are not both zero. The corresponding dispersion relation takes the form  $\det(\mathcal{L}) = 0$ , which yields

$$\hbar^2\omega^2 = (\hbar\Omega + NQ)^2 - N^2 K \bar{K}. \quad (17)$$

Then the solution can be written

$$\rho_1^1 = f \begin{pmatrix} 0 & -NK \\ \hbar(\Omega + \omega) + NQ & 0 \end{pmatrix} \quad (18)$$

$$\vec{E}_1^1 = \vec{e}_1^1 f \quad \text{with: } \vec{e}_1^1 = -N \begin{pmatrix} \theta_x/\beta \\ \theta_y/\beta \\ \theta_z \end{pmatrix} \quad (19)$$

where, for  $s = x, y, z$

$$\theta_s = 4\pi (\mu_s (\hbar (\Omega + \omega) + NQ) - \bar{\mu}_s NK). \quad (20)$$

For other values of  $p$ , the equations reduce to an analogous linear homogeneous system, whose determinant is deduced from  $\det \mathcal{L}$  by replacing  $\omega$  by  $p\omega$ , or when  $p = 0$ , replacing  $\omega$  by 0 and  $\beta$  by  $1 - c^2/V^2$ . Thus all the  $\rho_1^p$  and all the  $\bar{E}_1^p$  are zero, for  $p \neq \pm 1$ . Notice that this is necessarily true without any particular assumption for  $|p| > 1$ , while the same result for  $p = 0$  is closely related to the scaling, in which we imposed a unique velocity for all harmonics of the wave.

### 3.3. Order 2

Nonlinear terms appear at this order in the Schrödinger equation (6). They read

$$\sum_{s=x,y,z} \sum_{q+r=p} E_1^{q,s} [\mu_s, \rho_1^r] \quad (21)$$

and are *a priori* not zero for  $p = 0$  and  $\pm 2$ . For  $p = 0$ , there are two terms in the sum, that are Hermitian conjugates. Due to the fact that the commutator is an antisymmetric product, the terms are antihermitic. But they are also real, and thus cancel each other. Only one nonlinear term is left at this order:

$$\sum_{s=x,y,z} E_1^{1,s} [\mu_s, \rho_1^1] = -2\hbar\omega NK (\hbar (\Omega + \omega) + NQ) f^2 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (22)$$

As usual and as mentioned above for the first order, the second harmonic is defined in a unique way. A remarkable feature is that the corrections to the field and polarization density at this order are zero ( $\bar{E}_2^2 = \bar{0}$ ,  $\bar{P}_2^2 = \bar{0}$ ), while the density matrix is not:

$$\rho_2^2 = NK (\hbar (\Omega + \omega) + NQ) f^2 \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (23)$$

The nonvanishing components of  $\rho_2^2$  are the populations ( $a$  and  $b$  components); while the coherences  $\rho_{2,t}^2$  and  $\rho_{2,u}^2$  are zero, which implies the vanishing of the electric field. All other harmonics (i.e. all terms but those corresponding to  $p = \pm 2$  and  $\pm 1$ ) are zero, due to the absence of the nonlinear term. Justification is the same as at first order.

The fundamental term ( $p = 1$ ), at order  $\varepsilon^2$ , is treated as follows. The Maxwell equation (3) gives a relation between  $\bar{E}_2^1$  and  $\bar{P}_2^1$  analogous to (12) but involving the previous order:

$$\begin{aligned} E_2^{1,s} &= -\frac{4\pi}{\beta} P_1^{1,s} + i\hbar N \Lambda \theta_s \partial_\tau f & \text{for } s = x, y \\ E_2^{1,z} &= -4\pi P_2^{1,z} \end{aligned} \quad (24)$$

with

$$\Lambda = \frac{-2c}{\hbar\omega\beta^2 V} \left( \frac{V}{c} (\beta - 1) + \frac{kc}{\omega} \right). \quad (25)$$

The corrections to the populations  $\rho_{2,a}^1$  and  $\rho_{2,b}^1$  are zero, and the coherences are related through

$$\mathcal{L} \begin{pmatrix} \rho_{2,t}^1 \\ \rho_{2,u}^1 \end{pmatrix} = i\hbar \left( \frac{NK + N\Lambda \sum_{s=x,y} \theta_s \mu_s}{(\hbar (\Omega + \omega) + NQ) + N\Lambda \sum_{s=x,y} \theta_s \bar{\mu}_s} \right) \partial_\tau f. \quad (26)$$

(Notice that the index  $s$  takes the three values  $x, y$  or  $z$  in some cases, and only the two values  $x$  and  $y$  in others.) The solvability condition follows, it gives the value of the velocity  $V$ :

$$V = \frac{kc^2}{\omega} \frac{4\pi N [2\hbar\Omega (|\mu_x|^2 + |\mu_y|^2) - 4\pi N (\sigma_x^2 + \sigma_y^2 + 2\sigma_z^2/\beta)]}{\hbar^2 (k^2 c^2 - \omega^2) - \frac{k^2 c^2}{\omega^2} 4\pi N [2\hbar\Omega (|\mu_x|^2 + |\mu_y|^2) - 4\pi N (\sigma_x^2 + \sigma_y^2 + 2\sigma_z^2/\beta)]}. \quad (27)$$

The  $\sigma_j$  are defined by

$$\vec{\sigma} = \vec{\mu} \wedge \bar{\vec{\mu}}. \quad (28)$$

It is checked by direct computation that  $V = \frac{d\omega}{dk}$ . The complete solution at this order then reads

$$\rho_2^1 = \begin{pmatrix} 0 & -NKg \\ (\hbar(\Omega + \omega) + NQ)g + i\hbar\Xi\partial_\tau f & 0 \end{pmatrix} \quad (29)$$

$$\bar{E}_2^1 = -N \begin{pmatrix} \theta_x/\beta \\ \theta_y/\beta \\ \theta_z \end{pmatrix} g + i\hbar N \begin{pmatrix} \Lambda\theta_x - 4\pi\mu_x\Xi/\beta \\ \Lambda\theta_y - 4\pi\mu_y\Xi/\beta \\ -4\pi\mu_z\Xi \end{pmatrix} \partial_\tau f \quad (30)$$

where  $\Xi$  reads

$$\Xi = 1 + \sum_{s=y,z} \theta_s \mu_s \Lambda / K. \quad (31)$$

### 3.4. Order 3

The Maxwell equations at order  $\varepsilon^3$ , for the fundamental  $p = 1$ , yield, for  $s = x, y$ :

$$\begin{aligned} E_3^{1,s} &= -\frac{4\pi}{\beta} P_3^{1,s} + i\hbar N \Lambda \theta_s \partial_\tau g + \frac{2ikc^2}{\beta^2 \omega^2} N \theta_s \partial_\zeta f + U_s \partial_\tau^2 f \\ E_3^{1,z} &= -4\pi P_3^{1,z} \end{aligned} \quad (32)$$

with, for  $s = x, y$ :

$$\begin{aligned} U_s &= \frac{2N\hbar}{\beta\omega} \left( \Lambda\theta_s + \left(1 - \frac{1}{\beta}\right) 4\pi\mu_s \Xi \right) + \frac{1}{\beta\omega^2} \left(1 - \frac{1}{\beta}\right) N\theta_s \\ &\quad - \frac{2\hbar kc^2 N}{\omega^2 \beta V} \left( \Lambda\theta_s - \frac{4\pi\mu_s \Xi}{\beta} \right) + \frac{c^2 N \theta_s}{\omega^2 \beta^2 V^2}. \end{aligned} \quad (33)$$

The Schrödinger equation (6) involves only one nonlinear term, which reads

$$\sum_{s=x,y,z} E_1^{-1,s} [\mu_s, \rho_2^2] = \eta f |f|^2 \begin{pmatrix} 0 & v_t \\ v_u & 0 \end{pmatrix} \quad (34)$$

with

$$\eta = -2NK(\hbar(\Omega + \omega) + NQ) \quad (35)$$

$$v_t = \hbar(\Omega - \omega)(\hbar(\Omega + \omega) + NQ) \quad (36)$$

$$v_u = \hbar(\Omega + \omega) N\bar{K}. \quad (37)$$

The coherences obey the system

$$\mathcal{L} \begin{pmatrix} \rho_{3,t}^1 \\ \rho_{3,u}^1 \end{pmatrix} = \begin{pmatrix} W_t \\ W_u \end{pmatrix} \quad (38)$$

where the right-hand side member can be written, for  $j = t, u$ :

$$W_j = W_j^g \partial_\tau g + W_j^p \partial_\zeta f + W_j^d \partial_\tau^2 f + W_j^{nl} f |f|^2. \quad (39)$$

The solvability condition of system equation (38) reads

$$\begin{vmatrix} W_t & NK \\ W_u & \hbar(\Omega - \omega) + NQ \end{vmatrix} = 0. \quad (40)$$

According to (39), it can be expanded as follows:

$$R\partial_\tau g + iA\partial_\zeta f + B\partial_\tau^2 f + Cf|f|^2 = 0 \quad (41)$$

with

$$R = \begin{vmatrix} W_t^g & NK \\ W_u^g & \hbar(\Omega - \omega) + NQ \end{vmatrix} \quad (42)$$

and so on. We notice that the solvability condition of the system (26) at order 2, that gives  $V$ , is exactly  $R = 0$ , thus  $g$  vanishes from the equation. It is easily checked that  $A$ ,  $B$  and  $C$  are real constants, and condition (41) becomes the NLS equation

$$iA\partial_\zeta f + B\partial_\tau^2 f + Cf|f|^2 = 0. \quad (43)$$

#### 4. The coefficients of the NLS equation

##### 4.1. Preliminary

The coefficients of the NLS equation (43) are defined by, for  $j = p, d, nl$ :

$$X^j = \begin{vmatrix} W_t^j & NK \\ W_u^j & \hbar(\Omega - \omega) + NQ \end{vmatrix} \quad (44)$$

with  $X^p = iA$ ,  $X^d = B$ ,  $X^{nl} = C$ . A comparison with the coefficients given by other theories necessitates the presentation of equation (43) under some standard form (see e.g. [8], 6.5.32). This form reads

$$i\partial_\zeta \mathcal{E} - \frac{1}{2}k_2\partial_\tau^2 \mathcal{E} + \gamma \mathcal{E} |\mathcal{E}|^2 = 0. \quad (45)$$

The variable used in (45) is some component  $\mathcal{E}$  of the electric field amplitude, so that  $\vec{E}_1^1 = \mathcal{E}\vec{u}$ , where  $\vec{u}$  is a unitary polarization vector. Thus  $\mathcal{E} = \frac{1}{\|\vec{e}_1^1\|} f$ . The coefficients are identified as follows:

$$-\frac{1}{2}k_2 = \frac{B}{A} \quad \text{and} \quad \gamma = \frac{C}{A \|\vec{e}_1^1\|^2}. \quad (46)$$

The remainder of the paper is devoted to the comparison between the values of  $k_2$  and  $\gamma$  obtained in [8] and in this paper, respectively. Our purpose is to compare our results with the most commonly admitted theory. Therefore, we restrict our references to this often quoted book, despite the fact that many others exist including a lot of more recent works. The explicit expressions for  $A$  and  $\|\vec{e}_1^1\|$  are needed. The former follows from (44), it reads

$$iA = c^2 \begin{vmatrix} \frac{2ik}{\beta^2\omega^2} N \sum_{s=x,y} \theta_s \mu_s & NK \\ \frac{2ik}{\beta^2\omega^2} N \sum_{s=x,y} \theta_s \bar{\mu}_s & \hbar(\Omega - \omega) + NQ \end{vmatrix} \quad (47)$$

and thus reduces to

$$A = \frac{16\pi kc^2 N^2 K}{\beta^2 \omega^2} \left( 2\pi N \left( \sigma_x^2 + \sigma_y^2 + \frac{2\sigma_z^2}{\beta} \right) - \hbar\Omega (|\mu_x|^2 + |\mu_y|^2) \right) \quad (48)$$

$\vec{e}_1^1$  is given by (19), its squared norm reads

$$\|\vec{e}_1^1\|^2 = N^2 \left( \frac{1}{\beta^2} (|\theta_x|^2 + |\theta_y|^2) + |\theta_z|^2 \right). \quad (49)$$



#### 4.2. The dispersion coefficient

Note first that the dispersion relation (17) exactly coincides with that found from the linear susceptibility  $\overset{\leftrightarrow(1)}{\chi}(\omega)$  computed in [8], for the density matrix description of a two-level model, as here. An expression for  $\overset{\leftrightarrow(1)}{\chi}(\omega)$  is given by formula 3.5.15 in [8]. For a two-level Hamiltonian, and using the present notations, it reduces to

$$\chi_{ij}^{(1)}(\omega) = \frac{N}{\hbar} \left( \frac{\mu_i \bar{\mu}_j}{\Omega - \omega} + \frac{\bar{\mu}_i \mu_j}{\Omega + \omega} \right). \quad (50)$$

Seeking for a monochromatic plane wave solution of the Maxwell equations (3) with

$$\vec{P} = \overset{\leftrightarrow(1)}{\chi}(\omega) \cdot \vec{E} \quad (51)$$

a dispersion relation is obtained, that exactly coincides with (17). The derivative  $\frac{d\omega}{dk}$  is computed from this dispersion relation: the obtained value of the group velocity exactly coincides with that of expression (27).

The following point is to compute explicitly coefficient  $B$  of the dispersion term in equation (43). It reads

$$B = \begin{vmatrix} \sum_{s=x,y} \mu_s U_s & NK \\ \sum_{s=x,y} \bar{\mu}_s U_s - \hbar^2 \Xi & \hbar(\Omega - \omega) + NQ \end{vmatrix}. \quad (52)$$

After some computation, we get

$$B = B_{\Xi} \hbar^2 \Xi + B' \quad (53)$$

with

$$B_{\Xi} = NK - 4\pi N\Lambda (\hbar(\Omega - \omega)(\mu_x^2 + \mu_y^2) + 4\pi N\mu_z(\mu_y\sigma_x - \mu_x\sigma_y)) \quad (54)$$

$\Lambda$  is defined by equation (25).

$$B' = \frac{c^2 4\pi N^2 K}{\omega^2 \beta^2 V^2} \left( 1 + \frac{V^2}{c^2} (\beta - 1) - 2 \frac{V}{c} \beta \Lambda \hbar \omega \left( \frac{kc}{\omega} - \frac{V}{c} \right) \right) \times \left( 4\pi N \left( \sigma_x^2 + \sigma_y^2 + \frac{2\sigma_z^2}{\beta} \right) - 2\hbar\Omega (|\mu_x|^2 + |\mu_y|^2) \right). \quad (55)$$

It can be checked by formal and numerical computation that

$$\frac{B}{A} = \frac{-1}{2} \frac{d^2 k}{d\omega^2}. \quad (56)$$

Thus the expression of the dispersion coefficient found using the present multiscale expansion coincides with the value given in the literature, that is usually derived using the linear dispersion theory. Thus, with respect to its linear part, the present computation agrees perfectly with the already known results.

#### 4.3. The nonlinear coefficient

The coefficient  $C$  of the nonlinear term in equation (43) reads

$$C = \begin{vmatrix} -\eta v_t & NK \\ \eta v_u & \hbar(\Omega - \omega) + NQ \end{vmatrix}. \quad (57)$$

It reduces to

$$C = 4N^3 K^2 \bar{K} \hbar \Omega (\hbar(\Omega + \omega) + NQ). \quad (58)$$

It is useful, in order to give some physical interpretation of  $C$ , to simplify this expression, restricting it to some more specific physical situation. We assume that the polarization operator  $\bar{\mu}$ , in the two-level model, describes oscillations of the molecular dipole along some direction, making an angle  $\alpha$  with the propagation direction  $z$ . Thus

$$\bar{\mu} = \begin{pmatrix} \cos \alpha \\ 0 \\ \sin \alpha \end{pmatrix} \mu. \quad (59)$$

The quantities involved in the computation of the nonlinear coefficient  $\gamma$  then reduce to

$$C = \frac{4(4\pi)^3 N^3 \mu^4 \bar{\mu}^2}{\beta^4} \hbar \Omega (\cos^2 \alpha + \beta \sin^2 \alpha)^3 [\beta \hbar (\Omega + \omega) + 4\pi N \mu \bar{\mu} (\cos^2 \alpha + \beta \sin^2 \alpha)] \quad (60)$$

$$A = \frac{-4(4\pi)^2 N^2 \mu^3 \bar{\mu} \hbar \Omega k c^2}{\beta^3 \omega^2} \cos^2 \alpha (\cos^2 \alpha + \beta \sin^2 \alpha) \quad (61)$$

$$\|\bar{e}_1^1\|^2 = \frac{(4\pi)^2 N^2 \mu \bar{\mu} \hbar^2 (\Omega + \omega)^2}{\beta^2} (\cos^2 \alpha + \beta^2 \sin^2 \alpha) \quad (62)$$

and the nonlinear coefficient itself reads

$$\gamma = \frac{-\beta \omega^2 (\cos^2 \alpha + \beta \sin^2 \alpha)^2 (\beta \hbar (\Omega + \omega) + 4\pi N \mu \bar{\mu} (\cos^2 \alpha + \beta \sin^2 \alpha))}{4\pi c^2 \hbar^2 k N (\Omega + \omega)^2 \cos^2 \alpha (\cos^2 \alpha + \beta^2 \sin^2 \alpha)}. \quad (63)$$

The dispersion relation can be explicitly solved:

$$k = \frac{\omega}{c} \sqrt{\frac{\omega^2 - \omega_2^2}{\omega^2 - \omega_1^2}} \quad (64)$$

where

$$\omega_1^2 = \Omega \left( \Omega + \frac{8\pi N \mu \bar{\mu}}{\hbar} \sin^2 \alpha \right) \quad (65)$$

$$\omega_2^2 = \Omega \left( \Omega + \frac{8\pi N \mu \bar{\mu}}{\hbar} \right). \quad (66)$$

It is seen that the wave cannot propagate if  $\omega_1 \leq \omega \leq \omega_2$ . The nonlinear coefficient then reads

$$\gamma = \frac{-8\pi N \mu^2 \bar{\mu}^2 \omega \Omega (\omega^2 - \Omega^2)^2 \cos^4 \alpha}{\hbar^3 c \sqrt{(\omega^2 - \omega_1^2)^3 (\omega^2 - \omega_2^2)} \left[ (\omega^2 - \omega_1^2)^2 - (\Omega^2 - \omega_1^2) (\omega_2^2 - \omega_1^2) \right]}. \quad (67)$$

The nonlinear coefficient presents not only resonance terms for the linear resonance frequency  $\omega_1$ , but also a weaker divergence for the frequency  $\omega_2$  at which the wavevector  $k$  is zero. The resonance frequency  $\omega_1$  depends on the propagation direction. If it is assumed that the atomic dipoles are excited perpendicular to the propagation direction ( $\alpha = 0$ ), then  $\omega_1 = \Omega$  is the frequency corresponding to the difference between the atomic levels, and the nonlinear coefficient reduces to

$$\gamma = \frac{-8\pi N \mu^2 \bar{\mu}^2 \omega \Omega}{\hbar^3 c \sqrt{(\omega^2 - \Omega^2)^3 (\omega^2 - \omega_2^2)}}. \quad (68)$$

#### 4.4. The case of a circular polarization

Consider the particular situation where  $\mu_x = \mu$ ,  $\mu_y = i\mu$  and  $\mu_z = 0$ . This dipolar momentum can be excited by circularly polarized light (indeed, when  $\mu_z = 0$ , the polarization vector  $\vec{e}_1^1$  given by equation (19) is collinear to the dipolar momentum  $\vec{\mu}$ ) therefore this situation will be referred to as that of a circular polarization. It appears from (15) that the quantity  $K$  vanishes, thus so does the nonlinear coefficient  $C$  in the NLS equation (41). Care must be taken before we conclude. Indeed, the other coefficients  $A$  and  $B$  of equation (41) also vanish: the computation is in fact not completely valid in this case. The special case of the circular polarization can be considered as a limiting case of elliptic polarization. Computation shows that the corresponding limit of the ratio  $C/A$  is zero. This way we can conclude that the Kerr effect does not exist in this situation. However, it is preferable to check this statement by direct consideration of the equations. We must consider the equations obtained at first order for the density matrix: the equation (14) for the term  $\rho_1^1$ . For a circular polarization, with  $K = 0$ , the matrix of the linear system (14) is diagonal. The solution is either

$$\rho_1^1 = \begin{pmatrix} 0 & \rho_{1,t}^1 \\ 0 & 0 \end{pmatrix} \quad \text{if } \hbar(\Omega + \omega) + NQ = 0 \quad (69)$$

or

$$\rho_1^1 = \begin{pmatrix} 0 & 0 \\ \rho_{1,u}^1 & 0 \end{pmatrix} \quad \text{if } \hbar(\Omega - \omega) + NQ = 0. \quad (70)$$

Note that expression (19), (20) for the polarization vector is valid only for the latter solution. Then we pursue the resolution of the perturbative scheme. There is only one nonlinear term appearing in the Schrödinger equation (6) at order 2, it is given by equation (22). According to (12) and (11), the electric field components can be written

$$E_1^{1,s} = \frac{-4\pi N}{\beta} (\rho_{1,u}^1 \mu_s + \rho_{1,t}^1 \bar{\mu}_s). \quad (71)$$

The commutator involved by equation (22) reads

$$[\mu_s, \rho_1^1] = (\mu_s \rho_{1,u}^1 - \bar{\mu}_s \rho_{1,t}^1) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (72)$$

And thus the nonlinear term reads

$$\sum_{s=x,y,z} E_1^{1,s} [\mu_s, \rho_1^1] = \frac{-4\pi N}{\beta} \left( (\mu_x^2 + \mu_y^2) (\rho_{1,u}^1)^2 - \overline{(\mu_x^2 + \mu_y^2)} (\rho_{1,t}^1)^2 \right) \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} = 0. \quad (73)$$

Thus  $\rho_2^2$  vanishes. Because the zero harmonic of the density, and both the zero and second harmonics of the field already vanish at this order, there is no longer a nonlinear term at order 2. Therefore there is no self-interaction at order 3, because the nonlinearity is quadratic. This shows that the evolution equation derived at order 3 will be linear. In other words, the Kerr effect does not arise for circular polarizations, in the frame of a two-level model.

## 5. Comparison with the standard theory

### 5.1. A simple case

In order to compare the result of this paper to previous calculations, we can compute the nonlinear coefficient  $\gamma$  by means of the nonlinear susceptibilities. According to [8]

(equations (6.5.31) and (4.1.19)), the nonlinear coefficient  $\gamma$  is related to the nonlinear susceptibility  $\chi^{(3)} = \chi^{(3)}(\omega = \omega + \omega - \omega)$  through

$$\gamma = \frac{6\omega\pi}{nc} \chi^{(3)} \tag{74}$$

where  $n$  is the linear refractive index.  $\chi^{(3)}$  can be computed from formula (3.7.14) of [8]. We rewrite down this formula with slight modifications in the notations, as follows:

$$\chi^{(3)}(\omega_p + \omega_q + \omega_r, \omega_r, \omega_q, \omega_p) = \mathcal{P}_I \chi^{(3)'}(\omega_p + \omega_q + \omega_r, \omega_r, \omega_q, \omega_p) \tag{75}$$

where  $\mathcal{P}_I$  is the averaging operator on the permutations of the indices  $(p, q, r)$ , and  $\chi^{(3)'}$  $(\omega_r, \omega_q, \omega_p)$  is a primitive expression of the susceptibility, that does not possess the required symmetry properties. It reads

$$\begin{aligned} \chi_{kjih}^{(3)'}(\omega_t, \omega_r, \omega_q, \omega_p) &= \frac{N}{\hbar^3} \sum_{vnml} \rho_{ll}^{(0)} \\ &\times \left\{ \frac{\mu_{lv}^k \mu_{vn}^j \mu_{nm}^i \mu_{ml}^h}{[\omega_{vl} - \omega_t][\omega_{nl} - \omega_p - \omega_q][\omega_{ml} - \omega_p]} \right. \\ &+ \frac{\mu_{lv}^h \mu_{vn}^k \mu_{nm}^j \mu_{ml}^i}{[\omega_{nv} - \omega_r][\omega_{mv} - \omega_p - \omega_q][\omega_{vl} + \omega_p]} \\ &+ \frac{\mu_{lv}^i \mu_{vn}^k \mu_{nm}^j \mu_{ml}^h}{[\omega_{nv} - \omega_t][\omega_{vm} + \omega_p + \omega_q][\omega_{ml} - \omega_p]} \\ &+ \frac{\mu_{lv}^h \mu_{vn}^i \mu_{nm}^k \mu_{ml}^j}{[\omega_{mn} - \omega_t][\omega_{nl} + \omega_p + \omega_q][\omega_{vl} + \omega_p]} \\ &+ \frac{\mu_{lv}^j \mu_{vn}^k \mu_{nm}^i \mu_{ml}^h}{[\omega_{vn} + \omega_t][\omega_{nl} - \omega_p - \omega_q][\omega_{ml} - \omega_p]} \\ &+ \frac{\mu_{lv}^h \mu_{vn}^j \mu_{nm}^k \mu_{ml}^i}{[\omega_{nm} + \omega_t][\omega_{mv} - \omega_p - \omega_q][\omega_{vl} + \omega_p]} \\ &+ \frac{\mu_{lv}^i \mu_{vn}^j \mu_{nm}^k \mu_{ml}^h}{[\omega_{nm} + \omega_t][\omega_{vm} + \omega_p + \omega_q][\omega_{ml} - \omega_p]} \\ &\left. + \frac{\mu_{lv}^h \mu_{vn}^i \mu_{nm}^j \mu_{ml}^k}{[\omega_{ml} + \omega_t][\omega_{nl} + \omega_p + \omega_q][\omega_{vl} + \omega_p]} \right\}. \tag{76} \end{aligned}$$

We have set  $\omega_t = \omega_p + \omega_q + \omega_r$  for simplicity. The matrix  $(\rho_{lm}^{(0)})$  is the same as denoted  $\rho_0$  above, and the matrix  $(\mu_{lm}^j)$  is the same as  $\mu^j$  defined by (2). Relative to the original formula, we have dropped the imaginary terms  $\pm i\gamma_{lm}$  in the denominators. Indeed, these terms account for the damping, which is neglected in the present model, and therefore must be taken as zero for the comparison.

When considering the Kerr effect, or self-interaction of one wave, formulas (75), (76) present a divergence. Therefore we write

$$\chi_{kjih}^{(3)}(\omega, \omega, \omega, -\omega) = \lim_{\delta\omega \rightarrow 0} \chi_{kjih}^{(3)}(\omega + \delta\omega, \omega, \omega, -\omega + \delta\omega). \tag{77}$$

The terms  $\chi_{kjih}^{(3)'}(\omega + \delta\omega, \omega, -\omega + \delta\omega, \omega)$  and  $\chi_{kjih}^{(3)'}(\omega + \delta\omega, \omega, \omega, -\omega + \delta\omega)$  cancel each other and finally

$$\chi_{kjih}^{(3)}(\omega, \omega, \omega, -\omega) = \frac{N}{3\omega\hbar^3} \left[ \frac{\mu_h \bar{\mu}_k \mu_j \bar{\mu}_i}{(\Omega + \omega)^2} - \frac{\mu_k \bar{\mu}_j \mu_i \bar{\mu}_h}{(\Omega - \omega)^2} + \frac{\mu_h \bar{\mu}_i \mu_k \bar{\mu}_j - \mu_i \bar{\mu}_k \mu_j \bar{\mu}_h}{\Omega^2 - \omega^2} \right]. \tag{78}$$

Consider first a polarization operator  $\vec{\mu} = (\mu, 0, 0)$  parallel to the  $x$ -axis. This means that the transition considered in the two-level model corresponds to oscillations of the charge along the  $x$ -axis, and can be excited by light linearly polarized along the  $x$  direction. Then the only component of  $\chi^{(3)}$  to be considered is  $\chi_{xxxx}^{(3)}$ , computed from equation (78):

$$\chi_{xxxx}^{(3)}(\omega, \omega, \omega, -\omega) = \chi_0^{(3)} = -\frac{4}{3} \frac{N}{\hbar^3} \frac{\Omega |\mu|^4}{(\omega^2 - \Omega^2)^2}. \quad (79)$$

Then making use of (74), we find exactly the same value of  $\gamma$  as in (68). Thus the present theory completely agrees with previous calculations in this simple case.

### 5.2. An anisotropic situation

Two other situations will be considered. First, we assume that

$$\vec{\mu} = \begin{pmatrix} \mu \cos \alpha \\ 0 \\ \mu \sin \alpha \end{pmatrix}. \quad (80)$$

This corresponds to an anisotropic situation, where the charge oscillations taken into account by the two-level model can only be excited in a fixed direction that makes an angle  $\alpha$  with the transverse direction  $x$ . The exciting light is still linearly polarized. The polarization is directed by the vector  $\vec{e}_1^1$  given by equation (19), with

$$\vec{e}_1^1 \propto \vec{u} = \begin{pmatrix} \cos \alpha / (1 - n^2) \\ 0 \\ \sin \alpha \end{pmatrix}. \quad (81)$$

We assume that the electric field  $\vec{E}$  takes the form

$$\vec{E} = \vec{E}(\omega) e^{i\varphi} + \text{c.c.} = \mathcal{E} e^{i\varphi} \frac{\vec{u}}{\|\vec{u}\|} + \text{c.c.} \quad (82)$$

The component at frequency  $\omega$  and along the  $j$ -axis of the nonlinear polarization  $\vec{P}_{nl}$  writes, according to [8], equation (4.1.5):

$$\begin{aligned} P_{nl}^j(\omega) &= 3 \left( \chi^{(3)}(\omega, \omega, \omega, -\omega) \cdot \vec{E}(\omega) \vec{E}(\omega) \vec{E}(-\omega) \right)_j \\ &= 3 \frac{\mathcal{E} |\mathcal{E}|^2}{\|\vec{u}\|^3} \left\{ \chi_{xxxx}^{(3)} u_x^3 + \left( \chi_{jxxz}^{(3)} + \chi_{jxzx}^{(3)} + \chi_{jzxx}^{(3)} \right) u_x^2 u_z \right. \\ &\quad \left. + \left( \chi_{jxzz}^{(3)} + \chi_{jzxx}^{(3)} + \chi_{jzxx}^{(3)} \right) u_x u_z^2 + \chi_{jxxx}^{(3)} u_z^3 \right\}. \end{aligned} \quad (83)$$

On the other hand, making use of the expression (80) of  $\mu$  in the expression (78) of  $\chi^{(3)}$  yields

$$P_{nl}^j(\omega) = 3 \frac{\mathcal{E} |\mathcal{E}|^2}{\|\vec{u}\|^3} \chi_0^{(3)} \left( \frac{\cos^2 \alpha}{1 - n^2} + \sin^2 \alpha \right)^3 \begin{pmatrix} \cos \alpha \\ 0 \\ \sin \alpha \end{pmatrix} \quad (84)$$

where  $\chi_0^{(3)}$  is the value of  $\chi_{xxxx}^{(3)}$  obtained when  $\alpha = 0$  (cf equation (79)). The first observation is that  $\vec{P}_{nl}(\omega)$  is not collinear to  $\vec{E}(\omega)$ . Thus equation (6.5.32) of [8] cannot be used directly. A naive generalization of this equation could take the form

$$\begin{aligned} i \partial_\zeta E^x - \frac{k''}{2} \partial_\tau^2 E^x + \frac{2\pi\omega}{nc} P_{nl}^x &= 0 \\ i \partial_\zeta E^z - \frac{k''}{2} \partial_\tau^2 E^z + \frac{2\pi\omega}{nc} P_{nl}^z &= 0. \end{aligned} \quad (85)$$

Together with the value (84) of  $\vec{P}_{nl}$ , this yields two different NLS equations for a single wave amplitude  $\mathcal{E}$ , instead of a single one according to the result of this paper. When considering two independent polarizations, a set of two coupled NLS equations is a well known model, that accounts for their interaction. But here  $E^x$  and  $E^z$  are not independent. Thus the nonlinear susceptibilities seem to fail to describe this particular situation. Although it is not correct from the mathematical point of view, we can solve this problem by simply dropping one of the two equations in (85). This yields a single NLS equations for the amplitude  $\mathcal{E}$ , whose nonlinear coefficient reads as follows. If we keep the second equation, the nonlinear coefficient is  $\gamma_z$ , such that

$$\gamma_z = \frac{6\omega\pi}{nc} \chi_0^{(3)} \frac{\left(\frac{\cos^2 \alpha}{1-n^2} + \sin^2 \alpha\right)^3}{\left(\frac{\cos^2 \alpha}{(1-n^2)^2} + \sin^2 \alpha\right)}. \quad (86)$$

If we keep the first equation, we get a different coefficient,  $\gamma_x$ , with

$$\gamma_x = \gamma_z(1 - n^2). \quad (87)$$

Because  $\gamma_x$  tends to the value of  $\gamma$  given by (74) as  $\alpha$  tends to zero, and  $\gamma_z$  does not, the former is likely a better approximate value of the coefficient. Using the dispersion relation (64), we can rewrite the expression for  $\gamma_x$ . We have the identities

$$\cos^2 \alpha + (1 - n^2) \sin^2 \alpha = \frac{\omega^2 - \Omega^2}{\omega^2 - \omega_1^2} \cos^2 \alpha \quad (88)$$

$$\cos^2 \alpha + (1 - n^2)^2 \sin^2 \alpha = \frac{\cos^2 \alpha}{(\omega^2 - \omega_1^2)^2} \left[ (\omega^2 - \omega_1^2)^2 + (\omega_2^2 - \omega_1^2) (\omega_1^2 - \Omega^2) \right]. \quad (89)$$

Thus  $\gamma_x$  takes the form

$$\gamma_x = \frac{8\pi\omega\Omega N |\mu|^4}{c\hbar^3} \frac{(\omega^2 - \Omega^2) \cos^4 \alpha}{\sqrt{(\omega^2 - \omega_2^2) (\omega^2 - \omega_1^2) \left[ (\omega^2 - \omega_1^2)^2 + (\omega_2^2 - \omega_1^2) (\omega_1^2 - \Omega^2) \right]}}. \quad (90)$$

This expression can be compared to the expression (67) of the nonlinear coefficient  $\gamma$  found by the multiscale approach. Both expressions are comparable, but not identical: the resonance at  $\omega = \omega_1$  is much weaker in the formula computed from the susceptibilities:  $\gamma_x \propto (\omega - \omega_1)^{-1/2}$ , instead of  $(\omega - \omega_1)^{-3/2}$  in (67). In the same way, the coefficient  $\gamma$  vanishes as  $(\omega - \Omega)^2$  in (67), while  $\gamma_x$  is proportional to  $(\omega - \Omega)^1$  only.

The origin of the discrepancy seems to stem from the fact that the computation of the nonlinear susceptibilities does not take the propagation into account. Indeed, no mention of a propagation direction, not even of a space variable appears in the derivation of the susceptibilities [8]. But the propagation affects the nonlinear effect. The shift of the resonance frequency can be recovered when determining the dispersion relation from the linear susceptibility. The use of the obtained polarization vector partly corrects the expression for  $\gamma_x$ , but not completely. The considered situation, of a two-level model involving a non-transverse polarization with regard to the propagation direction, can be considered as the most simple model that gives an account of anisotropy. It appears that in such a situation, the nonlinear susceptibilities do not yield the nonlinear evolution equation in a way completely coherent from the mathematical point of view, while the multiscale expansion approach is mathematically totally coherent, and that the derived nonlinear coefficient can be erroneous. The same kind of discrepancy between the results obtained by the two approaches can be expected to arise when considering light propagation in an anisotropic medium in a more realistic way.

### 5.3. Circular polarization

Now we choose  $\mu_x = \mu$ ,  $\mu_y = i\mu$ , and  $\mu_z = 0$ . This corresponds to a transition which can only been excited by a circularly polarized wave. This is, for example, the case for the transition between the states  $1s$  and  $2p, m = +1$  of an hydrogenoid atom, where  $m$  is the quantum number that refers to the  $z$ -component of the orbital kinetic momentum. The dipolar momentum is  $\vec{\mu} = -e\vec{x}$ , where  $e$  is the absolute value of the electronic charge, and  $\vec{x} = (x, y, z)$ . The matrix of the  $\vec{\mu}$  components in the subspace of the space of the atomic states generated by the two states  $1s$  and  $2p, m = +1$ , can be computed from the explicit expressions for the atomic orbitals, which can be found in any elementary book about quantum mechanics. These matrices read

$$\mu_x = \frac{128}{243}a_0e \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \mu_y = \frac{128}{243}a_0e \begin{pmatrix} 0 & i \\ -i & 0 \end{pmatrix} \quad \mu_z = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}. \quad (91)$$

This corresponds to our assumption ( $a_0$  is the Bohr radius). This computation is not intended to prove that the considered two-level model will actually describe the behaviour of hydrogenoid atoms in some conveniently polarized light, but only that the above assumption on  $\vec{\mu}$  has some physical relevance.

In this case the electric wave field reads

$$\vec{E} = \frac{\mathcal{E}}{\sqrt{2}} \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix} + \text{c.c.} \quad (92)$$

The nonlinear polarization reads then, in a way analogous to (83):

$$P_{nl}^x(\omega) = \frac{3}{2\sqrt{2}} \mathcal{E} |\mathcal{E}|^2 \left\{ \chi_{xxxx}^{(3)} - i\chi_{xxxy}^{(3)} + i(\chi_{xxyx}^{(3)} + \chi_{xyxx}^{(3)}) \right. \\ \left. + (\chi_{xxyy}^{(3)} + \chi_{xyxy}^{(3)}) - \chi_{xyyx}^{(3)} + i\chi_{xyyy}^{(3)} \right\} \quad (93)$$

$$P_{nl}^y(\omega) = iP_{nl}^x(\omega). \quad (94)$$

The  $\chi^{(3)}$  components are computed from (78), and then  $P_{nl}^x(\omega)$  is computed explicitly. The vectorial NLS equation (85) yields a single NLS equation for the amplitude  $\mathcal{E}$ , with the nonlinear coefficient

$$\gamma = \frac{8\pi}{nc\hbar^3} \frac{N|\mu|^4}{(\Omega^2 - \omega^2)}. \quad (95)$$

Thus the nonlinear coefficient  $\gamma$  computed from the nonlinear susceptibilities is not zero, while the multiscale approach has shown that the Kerr effect does not occur in this case. The reason is, that the approach that uses nonlinear susceptibilities decomposes the circular polarization into the sum of two linear polarizations, computes a nonlinear effect for each component, and again combines the results. Such a method uses a summability property that is properly linear, and is not true for a nonlinear problem.

## 6. Conclusion

A NLS equation, describing the evolution of some temporally localized pulse of some plane wave in a nonlinear optical medium, has been derived directly from a quantum mechanical model. The Kerr effect is mainly due to oscillations of the populations of the two levels at the second-harmonic frequency, while the oscillations of the wave field and of the polarization density at the second harmonic frequency are negligible.

The linear properties (the dispersion relation, the values of the group velocity and of the dispersion coefficient in the NLS equation) have been found again, in complete accordance

with other computations. The nonlinear coefficient in the NLS equation completely agrees with the result given by less rigorous theories, in the simple case of a linearly polarized transverse wave. But in more complicated situations that yield a simplified anisotropic model, there is an appreciable discrepancy. This is, due to the fact that the computation of the nonlinear susceptibilities does not make any reference to the propagation direction. The shift of the linear resonance that occurs when the atomic dipoles are not perpendicular to the propagation direction is therefore not correctly accounted for by the nonlinear susceptibilities, and the strength of the corresponding resonance is under-evaluated. In contrast, the viewpoint adopted by this paper gives a correct account of the propagation. When considering a transition involving a circular polarization, the two approaches disagree totally. Indeed, the multiscale expansion shows that no self-interaction occurs, while the nonlinear polarization computed with the help of the nonlinear susceptibilities is not zero.

Although this paper is restricted to the academic problem of a two-level atom, it will be possible to generalize this approach to more realistic situations. We have thus given a way to derive the equations governing light propagation over relatively large distances in a nonlinear medium. The nonlinear coefficient obtained may differ appreciably from the standard values. The latter are only valid for the propagation through very thin samples, because they are derived neglecting retardation.

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