Kerr-free propagation of a near-resonant laser in an atomic vapor

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In this paper, we address the propagation of a near resonant laser inside an atomic vapor in the case when, due to the Kerr effect, the laser beam would either self-focus or self-defocus. We show, both theoretically and experimentally, how to get rid of such an alteration in the transverse beam profile without changing any of the characteristics of the laser light under consideration (wavelength, intensity, etc.), nor of the atomic vapor. Moreover, our proposed method offers a lot of control on the beam profile, whose transverse size after propagation may be chosen at will by making use of a second, copropagating laser, whose required wavelength and intensity may be derived analytically.

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I. INTRODUCTION

The optical Kerr effect [1], which results from the intensity dependence of the index of refraction, $n \equiv n_0 + n_2 I$, where I is the light intensity, is ubiquitous in laser-matter interaction and may lead to light self-defocusing (if $n_2 < 0$) or self-focusing (when $n_2 > 0$). This effect may be detrimental for various applications, e.g., isotope separation [2,3], when one needs to let a laser light with a specific wavelength, close to that of a spectral line, propagate inside a given atomic vapor. In addition to the constraints on the vapor composition and on the laser frequency, and because of technological issues or needs to "optimize" a given process, one often has to also specify the laser intensity, the vapor density, and the length of laser propagation inside the vapor. It may then happen that, for all these carefully chosen parameters, the space profile of the laser field may be significantly modified due to the Kerr effect, sometimes up to the point that the laser can no longer propagate, thus making the chosen application ineffective.

In this paper we provide a theoretical analysis, confirmed experimentally, showing that it is possible to get rid of the Kerr effect for the laser light of interest (henceforth called laser 1) by using a second copropagating laser (henceforth called laser 2), as is illustrated in Fig. 1. Indeed, the second laser provides an additional term in the index of refraction, which now writes $n = n_0 + n_2 I_1 - n_2' I_2$, where I_1 and I_2 are, respectively, the intensities of laser 1 and laser 2, and we show here that it is possible to choose the frequency and intensity of laser 2 so that $n_2 I_1 - n_2' I_2 \approx 0$. The Kerr effect induced by the second laser cancels out that due to the first one, which can then propagate inside the atomic vapor regardless of the constraints imposed on the laser or vapor properties. Moreover, it is clear that, with an appropriate choice of I_2, one may change the sign of $n_2 I_1 - n_2' I_2$ compared to that of n_2, thus leading to the focusing of a laser light that would naturally self-defocus or vice versa. Hence, our proposed scheme allows one to precisely monitor the propagation of virtually any (low intensity) laser light inside any atomic vapor.

Nevertheless, several conditions need to be satisfied for our scheme to be effective. In particular, the atoms need to be treated as three-level systems, levels $|0\rangle$, $|1\rangle$, and $|2\rangle$, as illustrated in Fig. 2 (levels $|0\rangle$, $|1\rangle$, and $|2\rangle$ may be split into hyperfine sublevels with no significant change to our basic results). The three-level approximation is well grounded if laser 1 (respectively, laser 2) is nearly resonant with transition $|0\rangle \to |1\rangle$ (respectively, $|1\rangle \to |2\rangle$) but way off resonance with any other transition, if the Doppler broadening of the spectral lines is much less than the detunings Δ_1 and Δ_2 defined in Fig. 2 and if the lifetimes of levels $|1\rangle$ and $|2\rangle$ are long enough for such effects as Raman scattering to be negligible. Moreover, in order to write the laser index of refraction as $n = n_0 + n_2 I_1 - n_2' I_2$, we need to use a perturbative expansion of the atoms' polarization up to third order in the field amplitudes, and the atoms' response to the laser electric fields needs to be nearly adiabatic. The latter condition is fulfilled if, for $j = 1$ or 2, $\Delta_j \tau_j \gg 1$, τ_j being the time duration of laser j, while the perturbative value of polarization is accurate if $\Omega_j / \Delta_j \ll 1$, where Ω_1 (respectively, Ω_2) is the Rabi frequency of transition $|0\rangle \to |1\rangle$ (respectively, $|1\rangle \to |2\rangle$).

Usually, the previous conditions are easily met, and they were indeed satisfied when we performed the experiment on an atomic vapor of barium, described in Sec. III, which yielded the results illustrated in Figs. 1 and 6 showing the actual effectiveness of our proposed scheme. Moreover, the values for the intensity I_2 and the detuning Δ_2 of laser 2, which were found experimentally to lead to an output profile of laser 1 that best matched its input profile, are very close to those predicted theoretically for the Kerr effect cancellation. This validates our theoretical analysis of the phenomenon.

The paper is organized as follows. In Sec. II, we detail the theoretical analysis that lets us derive the conditions that must be met by laser 2 in order to cancel out the Kerr effect on laser 1. The calculations are first performed for a three-level atom, and then generalized to allow for a hyperfine structure. Section III is devoted to the experimental evidence of our proposed scheme in an atomic vapor of barium, while Sec. IV concludes and summarizes this work.

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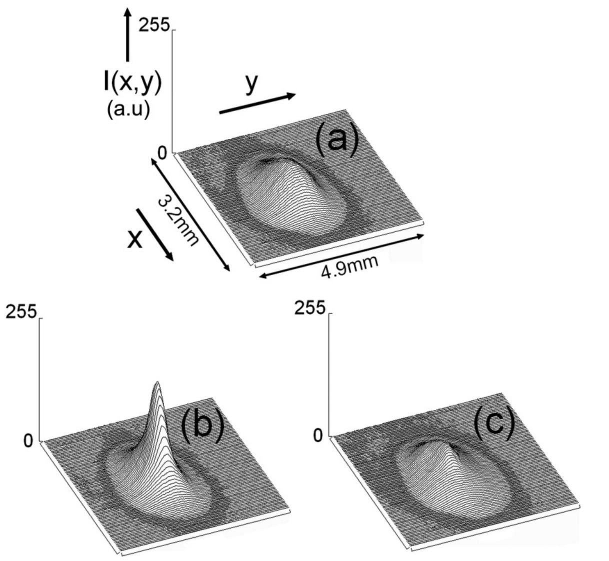

FIG. 1. XY surface plots of laser 1 at the output of a 1-m-long barium vapor in three situations illustrating the control of the Kerr effect by a copropagating laser, laser 2. (a) Laser 1 is alone and its frequency is far enough from the atomic transition for its space profile to remain nearly unperturbed. (b) Laser 1 is still sent alone in the vapor but its frequency is tuned near the atomic resonance (its detuning is $\Delta_1 = -25.84$ GHz, and its intensity is $I_1 = 3.2 \times 10^3$ W/cm^2) which leads to self-focusing due to the Kerr effect. (c) The input characteristics of laser 1 are the same as those in panel (b), but the Kerr effect is almost canceled out by making use of a second copropagating laser whose detuning with respect to resonance and intensity are, respectively, $\Delta_2 = -34.17$ GHz and $I_2 = 1.2 \times 10^6$ W/cm^2.

II. THEORETICAL ANALYSIS

In our analysis of the Kerr effect cancellation, we assume that no atom is ionized, and consider two linearly polarized laser fields with parallel polarizations, propagating along the z direction, so that the total electric field amplitude can be written as

$$E = E_1(x,y,z,t)e^{i(k_{l_1}z-\omega_{l_1}t)} + E_2(x,y,z,t)e^{i(k_{l_2}z-\omega_{l_2}t)} + \text{c.c.}, \tag{1}$$

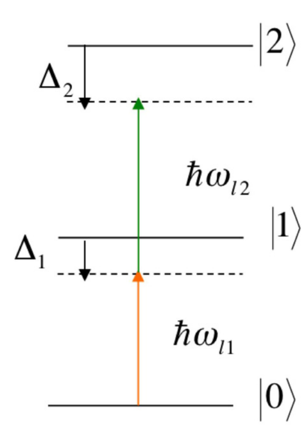

FIG. 2. (Color online) Energy ladder of the three-level atom.

where $\omega_{l_j} = k_{l_j}c$ ($j = 1,2$) and where E_1 and E_2 are, respectively, the slowly varying amplitudes of laser fields 1 and 2, which are such that, for $j = 1$ or 2, $|E_j^{-1}\partial_z E_j| \ll k_{l_j}$ and $|E_j^{-1}\partial_t E_j| \ll \omega_{l_j}$.

Similarly, we assume that the atom polarization, P, can be written as

$$P = P_1(x,y,z,t)e^{i(k_{l_1}z-\omega_{l_1}t)} + P_2(x,y,z,t)e^{i(k_{l_2}z-\omega_{l_2}t)} + \text{c.c.}, \tag{2}$$

where P_1 and P_2 are slowly varying envelopes. Then, within the paraxial approximation [4], we easily find the following equations of propagation for E_j ($j = 1,2$),

$$\frac{\partial E_j}{\partial z} + \frac{1}{c}\frac{\partial E_j}{\partial t} - \frac{i}{2k_{l_j}}\Delta_\perp E_j = \frac{ik_{l_j}}{2\varepsilon_0}P_j, \tag{3}$$

where $\Delta_\perp E_j \equiv \partial^2 E_j/\partial x^2 + \partial^2 E_j/\partial y^2$.

A. Three-level atom

For the sake of clarity, we first calculate the atom polarization by making use of the so-called three-level atom approximation, illustrated in Fig. 2, which amounts to assuming that the atom only has three levels of energy, $|0\rangle$, $|1\rangle$, and $|2\rangle$, so that the wave function may be written as

$$|\Psi\rangle = c_0|0\rangle + c_1e^{-i\omega_{l1}t}|1\rangle + c_2e^{-i(\omega_{l1}+\omega_{l2})t}|2\rangle. \tag{4}$$

This approximation is well grounded if laser 1 and 2 are, respectively, nearly resonant with the transitions $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$, but are way off resonance with any other atomic transition, and if spontaneous emission from levels $|1\rangle$ or $|2\rangle$ is negligible, so that such phenomena as stimulated Raman scattering are not to be accounted for. Moreover, the ground atomic level has to be nondegenerate; in particular there must not be any hyperfine structure (a theoretical analysis accounting for the hyperfine structure is given in Sec. II B), and the Doppler broadening of the spectral lines has to be small compared to the frequency detuning of the laser fields. Then, if we denote by $\hbar\omega_1$ and $\hbar\omega_2$ the energy difference between levels $|1\rangle$ and $|0\rangle$ and levels $|2\rangle$ and $|1\rangle$, respectively, and if we denote $\Delta_1 \equiv \omega_1 - \omega_{l1}$ and $\Delta_2 \equiv (\omega_1 + \omega_2) - (\omega_{l1} + \omega_{l2})$ (see Fig. 2), the Schrödinger equation readily yields the following:

$$\frac{dc_0}{dt} = i(\Omega_1^* e^{i\omega_{l1}t} + \Omega_2 e^{-i\omega_{l2}t} + \text{c.c.})c_1e^{-i\omega_{l1}t}, \tag{5}$$

$$\frac{dc_1}{dt} = -i\Delta_1c_1 + i(\Omega_1e^{-i\omega_{l1}t} + \Omega_2e^{-i\omega_{l2}t} + \text{c.c.})(c_0e^{i\omega_{l1}t} + c_2e^{i\omega_{l2}t}), \tag{6}$$

$$\frac{dc_2}{dt} = -i\Delta_2c_1 + i(\Omega_2e^{-i\omega_{l2}t} + \Omega_1e^{-i\omega_{l1}t} + \text{c.c.})c_1e^{i\omega_{l2}t}, \tag{7}$$

where Ω_1 and Ω_2 are the Rabi frequencies for lasers 1 and 2, $\Omega_{1,2} \equiv \mu_{1,2}E_{1,2}/\hbar$, with μ_1 being the dipole moment of transition $|0\rangle \to |1\rangle$ and μ_2 that of transition $|1\rangle \to |2\rangle$.

We henceforth make use of the so-called rotating-wave approximation (see Ref. [5]) that amounts to neglecting in Eqs. (5)–(7) the nonresonant terms, which are those whose phases vary very rapidly with time. This leads to the following

Bloch equations for $\sigma_{ij} \equiv c_i^* c_j$:

$$\frac{d\sigma_{01}}{dt} = -i\Delta_1\sigma_{01} + i\Omega_1(\sigma_{00} - \sigma_{11}) + i\Omega_2^*\sigma_{02}, \quad (8)$$

$$\frac{d\sigma_{02}}{dt} = -i\Delta_2\sigma_{02} + i\Omega_2\sigma_{01} - i\Omega_1\sigma_{12}, \quad (9)$$

$$\frac{d\sigma_{12}}{dt} = -i(\Delta_2 - \Delta_1)\sigma_{12} + i\Omega_2(\sigma_{11} - \sigma_{22}) - i\Omega_1^*\sigma_{02}. \quad (10)$$

Spontaneous emission has been neglected in the previous equations, which is valid if the lifetimes, T_1 and T_2, of levels $|1\rangle$ and $|2\rangle$ are such that $\Delta_{1,2}T_{1,2} \gg 1$. If this condition were not fulfilled, one would just need to replace Δ_1 by $\Delta_1 + i/T_1$ and Δ_2 by $\Delta_2 + i/T_2$, which would not greatly change the resolution of Eqs. (8)–(10).

Let us now solve Eqs. (8)–(10) by perturbation. At 0 order in the field amplitudes, $\sigma_{00} = 1$ while $\sigma_{ij} = 0$ whatever $(i,j) \neq (0,0)$. Hence, at first order in the field amplitudes, the equation for σ_{01} reads

$$\frac{d\sigma_{01}}{dt} = -i\Delta_1\sigma_{01} + i\Omega_1, \quad (11)$$

which yields, when $\Omega_1^{-1} d\Omega_1/dt \gg \Delta_1^{-1}$,

$$\sigma_{01} = \int_{-\infty}^{t} i\Omega_1(t')e^{i\Delta_1(t'-t)}dt' \quad (12)$$

$$= \Omega_1/\Delta_1 - \frac{1}{\Delta_1}\int_{-\infty}^{t} \frac{d\Omega_1}{dt'}e^{i\Delta_1(t'-t)}dt' \quad (13)$$

$$\approx \Omega_1/\Delta_1. \quad (14)$$

From the latter *adiabatic* [6] value for σ_{01}, we find that, at lowest order in the field amplitudes,

$$\sigma_{11} \approx |\Omega_1|^2/\Delta_1^2. \quad (15)$$

Plugging the value Eq. (14) for σ_{01} into Eq. (9) for σ_{02}, and accounting for the fact that $|\sigma_{01}| \gg |\sigma_{12}|$, one finds

$$\frac{d\sigma_{02}}{dt} \approx -i\Delta_2\sigma_{02} + i\frac{\Omega_1\Omega_2}{\Delta_1}, \quad (16)$$

which yields, using the same *adiabatic* approximation as for σ_{01},

$$\sigma_{02} \approx \frac{\Omega_1\Omega_2}{\Delta_1\Delta_2}, \quad (17)$$

from which one gets

$$\sigma_{22} \approx \frac{|\Omega_1|^2|\Omega_2|^2}{\Delta_1^2\Delta_2^2}. \quad (18)$$

Plugging the values we found for σ_{11}, σ_{22}, and σ_{02} into Eq. (10) for σ_{12}, and making use of the same adiabatic calculation as that employed to derive σ_{01} and σ_{02}, we find, at lowest order in the field amplitudes,

$$\sigma_{12} \approx \frac{\Omega_2|\Omega_1|^2}{\Delta_2\Delta_1^2}. \quad (19)$$

Since, from Eq. (18), σ_{22} is of order 4 in the field amplitudes, at third order, the identity $\sigma_{00} + \sigma_{11} + \sigma_{22} = 1$ simply amounts to $\sigma_{00} + \sigma_{11} = 1$, which, from Eq. (15) for σ_{11} yields

$$\sigma_{00} \approx 1 - |\Omega_1|^2/\Delta_1^2. \quad (20)$$

Using this value for σ_{00}, together with Eq. (15) for σ_{11} and Eq. (17) for σ_{02}, in Eq. (8) for σ_{01}, we find the following adiabatic expression for σ_{01}, at third order in the field amplitudes:

$$\sigma_{01} = \frac{\Omega_1}{\Delta_1}\left(1 - 2\frac{|\Omega_1|^2}{\Delta_1^2} + \frac{|\Omega_2|^2}{\Delta_1\Delta_2}\right). \quad (21)$$

Now, from the definition, Eq. (2), of P_1 and P_2, from Eq. (4) for $|\Psi\rangle$, and from the definition $P = N\langle\Psi|\hat{\mu}|\Psi\rangle$, one finds

$$P_1 = N\mu_1\sigma_{01}, \quad (22)$$

$$P_2 = N\mu_2\sigma_{12}. \quad (23)$$

Using the Eq. (21) for σ_{01} in Eq. (22) for P_1, and plugging this value of P_1 into the field equation, Eq. (3), one finds the following wave equation for E_1,

$$\frac{\partial E_1}{\partial z} + \frac{1}{c}\frac{\partial E_1}{\partial t} - \frac{i}{2k_1}\Delta_\perp E_1 = ik_1E_1\left[\frac{N\mu_1^2}{2\varepsilon_0\hbar\Delta_1} - \frac{N\mu_1^4|E_1|^2}{\varepsilon_0\hbar^3\Delta_1^3} + \frac{N\mu_1^2\mu_2^2|E_2|^2}{2\varepsilon_0\hbar^3\Delta_1^2\Delta_2}\right], \quad (24)$$

which straightforwardly shows that the index of refraction, $n^{(1)}$, for the field E_1 reads $n^{(1)} \equiv n_0^{(1)} + n_2^{(1)}I_1 - n_2'^{(1)}I_2$, with

$$n_0^{(1)} = 1 + \frac{N\mu_1^2}{2\varepsilon_0\hbar\Delta_1}, \quad (25)$$

$$n_2^{(1)} = \frac{-2N\mu_1^4}{\varepsilon_0^2c\hbar^3\Delta_1^3}, \quad (26)$$

$$n_2'^{(1)} = \frac{-N\mu_1^2\mu_2^2}{\varepsilon_0^2c\hbar^3\Delta_1^2\Delta_2}. \quad (27)$$

From these results, it is clear that there is no Kerr effect on laser 1 if laser 2 is chosen, so that

$$\frac{I_2}{I_1} = \frac{2\mu_1^2\Delta_2}{\mu_2^2\Delta_1}, \quad (28)$$

provided that laser 2 is not subject to focusing, or defocusing, due to a Kerr-like effect induced by laser 1. From Eq. (19) for σ_{12} it is clear that the index of refraction for the field E_2 is $n^{(2)} \equiv 1 + n_2^{(2)}I_1$, with

$$n_2^{(2)} = \frac{N\mu_1^2\mu_2^2}{\varepsilon_0^2c\hbar^3\Delta_1^2\Delta_2}. \quad (29)$$

Hence, when condition (28) is fulfilled,

$$n_2^{(2)}I_1 = \frac{\Delta_1\mu_2^2}{2\Delta_2\mu_1^2}n_2^{(1)}I_2 \equiv n'^{(2)}I_2. \quad (30)$$

From Eq. (30), a necessary condition for laser 2 to be very little affected by the Kerr effect is $(\mu_2^2\Delta_2)/(\mu_1^2\Delta_1) \ll 1$. More precisely, the power of laser 2 needs to be less than

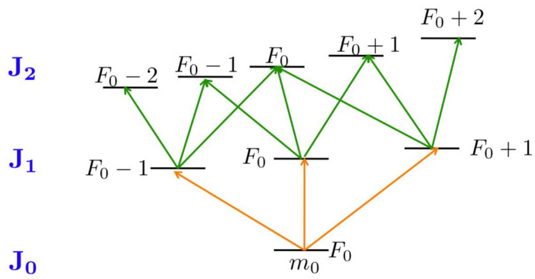

FIG. 3. (Color online) Hyperfine levels coupled to the ground level $|0,F_0,m_0\rangle$ in the case when $J_0=0$. Arrows show which levels are coupled.

the critical one, which scales like $\lambda_2^2/n'^{(2)}$ [5]. In particular, when designing our experiment on the Kerr-effect cancellation detailed in Sec. III, we were careful to choose our parameters so that the power of laser 2 was indeed less than the critical one.

B. Hyperfine structure and multi-isotope atoms

In this section, we show that our proposed scheme to cancel the Kerr effect by making use of a second, copropagating, laser still works when the atoms possess a hyperfine structure and when the atomic vapor is composed of several isotopes.

1. Hyperfine structure

Atoms with an odd number of nucleons possess a nonzero nuclear spin, and therefore a hyperfine structure, i.e., a fine level $|i\rangle$ ($|i\rangle=|0\rangle,|1\rangle$, or $|2\rangle$ for the three-level atom of Sec. II A), with kinetic momentum J_i, is split into hyperfine sublevels denoted by $|J_i,F_i,m_i\rangle$, where F_i is the total kinetic momentum including the nuclear spin ($F_i\equiv J_i+\mathcal{I}$, $\mathcal{I}$ being the nuclear spin) and where m_i is the projection of F_i along the direction of the laser's electric fields.

We still assume, in this section, that laser 1 is tuned close enough to the transition between fine levels $|0\rangle$ and $|1\rangle$ that no other fine levels are to be accounted for. Similarly, with regard to laser 2, we assume that only the transition between fine levels $|1\rangle$ and $|2\rangle$ is to be considered. However, within one given fine level, the energy difference between hyperfine sublevels is so small that one has to account for all allowed transitions, $|J_i,F_i,m_i\rangle\rightarrow|J_j,F_j,m_j\rangle$, that satisfy the selection rules $F_i-F_j=0,\pm1$ and $m_i=m_j$ (because we assume that the lasers are linearly polarized). The latter condition makes it possible to consider transitions for a given m independently of those with $m'\neq m$. An example of all transitions to be accounted for between hyperfine sublevels, when the kinetic momentum of the fine level $|0\rangle$ is $J_0=0$, and for a given m, is illustrated in Fig. 3.

The derivation of P_1 and P_2 when account is made of the atom hyperfine structure is reported in the Appendix. The result for P_1 is

$$P_1=N\frac{E_1}{\hbar}\left[\alpha\bar{\mu}_1^2-\frac{\beta_1\bar{\mu}_1^4|E_1|^2}{\hbar^2\Delta_1^3}+\frac{\beta_2\bar{\mu}_1^2\bar{\mu}_2^2|E_2|^2}{\hbar^2\Delta_1^2\Delta_2}\right], \quad (31)$$

where

$$\alpha\equiv\sum_{F_0}\sum_{m_0=-F_0}^{F_0}\sum_{j=-1}^{1}\frac{\mu^2_{1,F_0,F_0+j,m_0}}{g_0\Delta_1\bar{\mu}_1^2} \quad (32)$$

and where

$$\beta_1\equiv\frac{1}{g_0\bar{\mu}_1^4}\sum_{F_0}\sum_{m_0=-F_0}^{F_0}\left\{\left(\sum_{j=-1}^{1}\mu^2_{1,F_0,F_0+j,m_0}\right)^2+\sum_{j=-1}^{1}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\left[\mu_{1,F_0,F_0+j,m_0}\mu_{1,F_0,F_0+l,m_0}\mu_{1,F_0+j+l_1,F_0+l,m_0}\mu_{1,F_0+j+l_1,F_0+j,m_0}\right]\right\}, \quad (33)$$

$$\beta_2\equiv\frac{1}{g_0\bar{\mu}_1^2\bar{\mu}_2^2}\sum_{F_0}\sum_{m_0=-F_0}^{F_0}\sum_{j=-1}^{1}\left\{\mu_{1,F_0,F_0+j,m_0}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\left[\mu_{2,F_0+j,F_0+j+l,m_0}\mu_{2,F_0+j+l+l_1,F_0+j+l,m_0}\mu_{1,F_0,F_0+j+l+l_1,m_0}\right]\right\}. \quad (34)$$

In the expressions above, $\bar{\mu}_1$ and $\bar{\mu}_2$ are, respectively, the mean dipole moments of transitions $|0\rangle\rightarrow|1\rangle$ and $|1\rangle\rightarrow|2\rangle$, $\sum_{F_0}$ denotes the sum over all values of the total kinetic momentum, F_0, of level $|0\rangle$, and $g_0\equiv\sum_{F_0}(2F_0+1)$ is the number of hyperfine sublevels within level $|0\rangle$. Moreover μ_{i,F_i,F_i+j,m_i} is the dipole moment of the transition $|J_i,F_i,m_i\rangle\rightarrow|J_{i+1},F_i+j,m_i\rangle$, which, by convention, is zero if the transition does not exist [e.g., $F_i+j<0$, or $|F_i+j|>\max(F_{i+1})$]. As for Δ_1 and Δ_2, they are defined as in Fig. 2; in our derivation of P_1 we neglected the energy difference between hyperfine sublevels compared to Δ_1 and Δ_2.

From Eq. (31), it is clear that it is still possible to cancel out the Kerr effect for a given laser beam by making use of a second, copropagating laser, provided that the intensities I_1 and I_2 of lasers 1 and 2 are such that

$$\frac{I_2}{I_1}=\frac{\beta_1\bar{\mu}_1^2\Delta_2}{\beta_2\bar{\mu}_2^2\Delta_1}. \quad (35)$$

One can easily check that the result obtained in Sec. II A is recovered since, when the atom has no hyperfine structure, the sums $\sum_{F_0}$ in Eqs. (33) and (34) only contain one term, so that $\beta_1=2$ and $\beta_2=1$.

2. Multi-isotope atomic vapor

Let us now consider the situation when the lasers propagate inside an atomic vapor composed of a fraction, η_e, of isotopes with an even number of nucleons, and, therefore, no hyperfine structure, and of a fraction, η_o, of isotopes with an odd number of nucleons which, for the sake of simplicity, are assumed to all have the same nuclear spin, $\mathcal{I}$. Then, clearly, the polarization, P_1, is just

$$P_1 = N\frac{E_1}{\hbar}\left[(\eta_e+\eta_o\alpha)\bar{\mu}_1^2 - \frac{(\eta_e+\eta_o\beta_1)\bar{\mu}_1^4|E_1|^2}{\hbar^2} + \frac{(\eta_e+\eta_o\beta_2)\bar{\mu}_1^2\bar{\mu}_2^2|E_2|^2}{\hbar^2}\right], \quad (36)$$

so that the Kerr effect cancellation is obtained provided that the condition

$$\frac{I_2}{I_1} = \frac{(\eta_e+\eta_o\beta_1)\bar{\mu}_1^2\Delta_2}{(\eta_e+\eta_o\beta_2)\bar{\mu}_2^2\Delta_1} \quad (37)$$

is fulfilled. If the odd isotopes do not all have the same nuclear spin, and if the fraction of odd isotopes with nuclear spin, $\mathcal{I}_j$, is η_j, then, in Eq. (37), the terms $\eta_o\beta_1$ and $\eta_o\beta_2$ only need to be replaced by $\sum_j \eta_j\beta_1^{(j)}$ and $\sum_j \eta_j\beta_2^{(j)}$, where $\beta_1^{(j)}$ and $\beta_2^{(j)}$ are just the values assumed by β_1 and β_2 for the corresponding nuclear spin, $\mathcal{I}_j$.

C. Application to barium

Let us now make an explicit calculation, for an atomic vapor of barium, of the condition that must be fulfilled by I_2/I_1 in order to obtain the cancellation of the Kerr effect on laser 1 by making use of laser 2. The atomic vapor of barium was chosen for the experiment detailed in Sec. III so that the theoretical condition for the Kerr cancellation derived here can be directly compared with that found experimentally.

In its natural state, barium is composed of seven different isotopes; five of them (^{132}Ba, ^{134}Ba, ^{136}Ba, and ^{138}Ba), representing about 82% of its composition, have an even number of nucleons and therefore a 0 nuclear spin [7,8]. The isotopic abundance of ^{133}Ba is very small, less than 0.1%, while ^{135}Ba and ^{137}Ba have the same nuclear spin, $\mathcal{I} = 3/2$. Hence, the condition, Eq. (37), for the Kerr effect cancellation may be directly applied with $\eta_e = 0.82$ and $\eta_o = 0.18$.

The ground state of barium (level $|0\rangle$) is $|6s^2,{}^1S_0\rangle$ (hence $J_0 = 0$), and we choose for level $|1\rangle$ the level $|6s6p,{}^1P_1\rangle$ (hence, $J_1 = 1$), whose energy is 18 060.261 cm^{-1}, and for level $|2\rangle$ the level $|6s8s,{}^1S_0\rangle$ (hence, $J_2 = 0$), whose energy is 34 371.002 cm^{-1} [9]. All the allowed transitions between hyperfine levels we have to account for are given in Fig. 4.

Because $J_0 = 0$, there is only one possible value for F_0, $F_0 = \mathcal{I} = 3/2$, so that $g_0 = 4$. Moreover, because $J_2 = 0$, so that $F_2 = F_0$, the general expressions, Eqs. (33) and (34) respectively, for β_1 and β_2 can be cast in the more simple forms

$$\beta_1 = \frac{2}{4\bar{\mu}_1^4}\sum_{m=-F}^{F}\left\{\sum_{j=-1}^{1}\mu_{1,F,F+j,m}^2\right\}^2,$$
$$\beta_2 = \frac{1}{4\bar{\mu}_1^2\bar{\mu}_2^2}\sum_{m=-F}^{F}\left\{\sum_{j=-1}^{1}\mu_{1,F,F+j,m}\mu_{2,F+j,F,m}\right\}^2, \quad (38)$$

with $F = 3/2$.

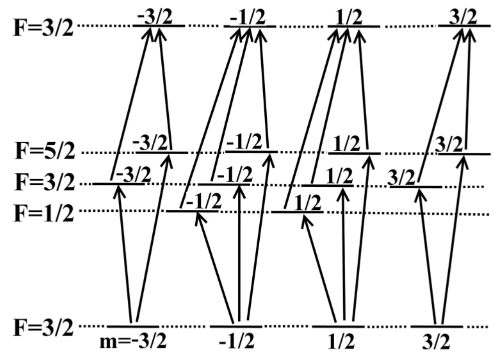

FIG. 4. Hyperfine structure levels in the case of the chosen transitions in ^{135}Ba or ^{137}Ba.

In order to derive β_1 and β_2, we need to calculate all the dipole moments, $\mu_{1,F,F+j,m}$ and $\mu_{2,F+j,F,m}$, for the transitions between hyperfine levels. To do so, we use the formula that can be found in Ref. [10], relating the dipole moment $\mu_{J,J',F,F',m}$ for the transition $|J,F,m\rangle \to |J',F',m\rangle$ to the mean value $\bar{\mu}$ of the dipole moment for the transition $|J\rangle \to |J'\rangle$:

$$\mu_{J,J',F,F',m} = (-1)^{\mathcal{I}+J+F'+F+1-m}\sqrt{3(2F+1)(2F'+1)} \times \begin{pmatrix} F & 1 & F' \\ -m & 0 & m \end{pmatrix}\begin{Bmatrix} J & J' & 1 \\ F' & F & \mathcal{I} \end{Bmatrix}, \quad (39)$$

where $\begin{pmatrix} F & 1 & F' \\ -m & 0 & m \end{pmatrix}$ is a three-j symbol and $\begin{Bmatrix} J & J' & 1 \\ F' & F & \mathcal{I} \end{Bmatrix}$ a six-j symbol (see Refs. [11,12]). For the barium, the six-j symbol needs to be calculated with $\mathcal{I} = 3/2$ and, for the chosen fine levels $|1\rangle$ and $|2\rangle$, its squared value is always $1/12$. Then, using the values of the 3-j symbols that can be found in Ref. [11], one easily finds that Eq. (38) yields $\beta_1 = 2$ while Eq. (39) yields $\beta_2 = 1$, i.e., exactly the same values as for an atom with no hyperfine structure. We therefore conclude that, for the chosen transitions in an atomic vapor of barium, Eq. (28) yields the condition for the cancellation of the Kerr effect on laser 1 by laser 2 provided that μ_1 and μ_2 are, respectively, the mean dipole moments of transitions $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$.

III. EXPERIMENTAL EVIDENCE

A. Setup

In order to check our theoretical prediction of the Kerr-free laser propagation, an experiment was led on an atomic vapor of barium using the atomic transitions specified in Sec. II C. Level $|1\rangle$ $(6s6p,{}^1P_1)$ corresponds to an energy of 18 060.261 cm^{-1}, while level $|2\rangle$ $(6s8s,{}^1S_0)$ corresponds to an energy of 34 371.002 cm^{-1}. The mean dipole moment for the transition $|0\rangle \to |1\rangle$ is well known to be $\mu_1 \approx 8$ D. As regards the dipole moment for the transition $|1\rangle \to |2\rangle$, we chose the value published in Ref. [13], $\mu_2 \approx 0.7$ D, which seems quite accurate [14].

In our Kerr-free experiment, the barium temperature is close to 900 K, so that for our laser wavelengths, $\lambda_1 \approx 553$ nm and $\lambda_2 \approx 613$ nm, the Doppler broadening is (FWHM) $\Delta\nu_D = \sqrt{8\ln(2)T/M}/\lambda \sim 1$ GHz.

The lifetimes of levels $|1\rangle$ and $|2\rangle$ are not known perfectly but may be estimated to be $T_1 \approx 20$ ns and $T_2 \approx 4$ μs [15].

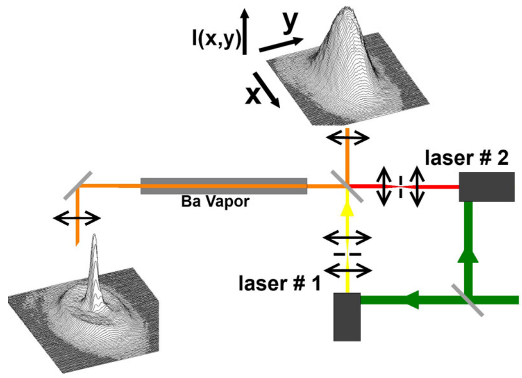

FIG. 5. (Color online) Experimental setup.

Figure 5 shows our experimental setup. The laser source is composed of a frequency-doubled single-mode-pulsed YAG:Nd3+ laser (Quanta-Ray DCR3) amplifying the output of two Coherent Inc. continuous-wave (cw)-stabilized single-mode ring dye oscillators pumped by two diode pumped solid state laser (DPSSL) Verdi 5W (Coherent Inc.). The absolute frequencies of the amplified laser fields are measured with cw laser wavelength meters (Burleigh WA1000). After spatial filtering, the transverse distribution is checked by a charge-coupled device (CCD) camera exhibiting a near transverse Gaussian profile.

The overlapping of the two beams is carefully adjusted with the help of a second CCD camera at the exit of the oven. Temporal pulse profiles ($\tau_l = 6$ ns duration total pulse width at half maximum with a nearly Gaussian distribution) as well as energy per pulse measurements are done before and after propagation in the atomic vapor with respectively high-speed photodiodes on a Lecroy 1-GHz bandwidth scope and calibrated Laser Precision RJ-7200 energy meters. With the same pump being used for lasers 1 and 2, there is no time jitter between the two lasers. This laser system is characterized by a reproducible output spatial distribution. Shot-to-shot variations of the temporal profile and of the energy per pulse are lower than a few percent.

The atomic medium is obtained using vapor pressure of solid barium heated in a temperature-controlled oven of $L =$ 1 m effective length. A vapor pressure of argon is applied to ensure window protection. Less than 1 Torr was used in order to obtain a dephasing collision time $T_{\rm col}$ as long as possible. We measured that a pressure of 1 Torr of argon leads to a time $T_{\rm col}$ of the order of 200 ns.

Temperatures in the oven are measured by a thermal sensor but do not have sufficient reliability to be used for deducing the barium atomic density. As a consequence, the NL product in the oven is determined for different temperatures between 870 K and 960 K using the method described in Ref. [16]. At $T = 900$ K, which is the temperature corresponding to the experiments described here, we find $NL = 5.44 \times 10^{15}\mathrm{cm}^{-2}$.

B. Kerr effect cancellation

The main parameters of the two lasers are summed up in Table I. From these data, the inverse Fresnel number is $\lambda L/\pi w^2 \approx 0.3$, so that the space profiles are almost not affected by free propagation (the beam waist increases by about 4.5% after 1 m of propagation for a Gaussian spatial profile).

TABLE I. Main experimental parameters for laser 1 and laser 2.

	Laser 1	Laser 2
Energy $\mathcal{E}$ (μJ)	0.25	120
Beam waist[a] along x, w_x (mm)	0.76	0.86
Beam waist[a] along y, w_y (mm)	0.88	0.96
Pulse duration (ns)	6	6
Wavelength (nm)	553	613

[a]The waist is the radius of $1/e$ irradiance.

The energy and detuning of laser 1 were chosen so as to obtain, without laser 2, a clear single filament at the output of the barium vapor. With laser 2 copropagating, we sought the parameters (laser 2 energy and frequency detuning) yielding an output profile for laser 1 which best agreed with the input one. Figure 1 shows one result of such an experimental procedure which corresponds to $\Delta_1 = -25.84$ GHz and $\Delta_2 = -34.17$ GHz, with an energy per pulse $\mathcal{E}_1 \approx 0.25$ μJ for laser 1 and $\mathcal{E}_2 \approx 120$ μJ for laser 2. One can easily check that the power of laser 1, $\mathcal{P}_1 \approx 39$ W, is way above the critical one, $\mathcal{P}_{c_1} \approx 0.4$ W [5,17], so that laser 1 alone is expected to self-focus after propagating over $L \approx 1$ m of barium vapor, as experimentally observed. By contrast, the power of laser 2, $\mathcal{P}_2 \approx 1.9 \times 10^4$ W, is less than the critical one, $\mathcal{P}_{c_2} \approx 6 \times 10^4$ W, so that the transverse profile of laser 2 is nearly not affected by its propagation in the atomic vapor of barium, as required for our process to be effective.

Both Δ_1 and Δ_2 are much larger than the Doppler broadening, than $1/T_1$ and $1/T_2$, and than $1/\tau_l$, so that the analysis of the Sec. II holds. In particular, we showed in Sec. II C that our theoretical condition to cancel out the Kerr effect on laser 1 by making use of laser 2 is given by Eq. (28), which yields

$$\left.\frac{I_2}{I_1}\right|_{\rm th} \approx 345, \tag{40}$$

while, experimentally, we find

$$\left.\frac{I_2}{I_1}\right|_{\rm exp} = \frac{\mathcal{E}_2 w_x^1 w_y^1}{\mathcal{E}_1 w_x^2 w_y^2} \approx 389. \tag{41}$$

The theoretical predictions are, therefore, in very good agreement with the experimental results. The relative discrepancy between the theoretical and experimental values of I_2/I_1 is close to 12%, which is within the uncertainties induced by those on the knowledge of μ_2 and on the measurements of the lasers' energies and waists. Note moreover that, as may be seen in Fig. 1, even if they are close, the output profile of laser 1, when laser 2 is copropagating, does not exactly match the out of resonance profile. This is mainly because the input profile of laser 2 does not exactly match that of laser 1 (see Table I).

C. Control of the output profile of laser 1 using laser 2

Clearly, using laser 2, it is possible to change the sign of the Kerr index of refraction of laser 1, i.e., make it that $(n_2^{(1)} I_1 - n_2^{\prime(1)} I_2) n_2^{(1)} < 0$, and make laser 1 focus while it

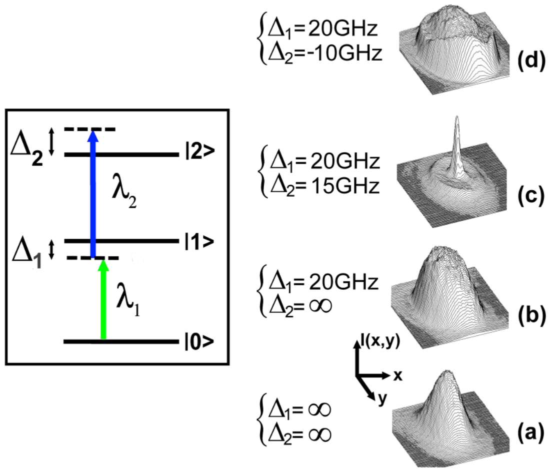

FIG. 6. (Color online) Example of the monitoring of the space profile of laser 1 obtained by tuning the frequency of laser 2. The other laser characteristics are identical to those of Fig. 1.

would have defocused when propagating alone, or vice versa. Actually, the value of $(n_2^{(1)}I_1 - n_2'^{(1)}I_2)$ can be chosen at will, so that the atomic vapor can be used as an adaptable lens that allows one to control the output profile of laser 1 using laser 2.

The monitoring of the space profile of laser 1, by making use of laser 2, is illustrated in Fig. 6, where, at constant energy per pulse, depending on the tuning of laser 2 frequency, we could make laser 1 focus inside the vapor while it would have defocused when propagating alone or, conversely, we could increase the defocussing. For instance, Fig. 6(a) displays the transverse profile of laser 1 at the exit of the vapor when laser 1 and laser 2 frequencies are far from the atomic resonance, i.e., when no interaction exists with the atomic vapor. Figure 6(b) represents the output transverse profile of laser 1 for a detuning $\Delta_1 = +20$ GHz (self-defocusing) and when laser 2 does not interact with the atomic medium (frequency of laser 2 far from the resonance). Figures 6(c) and 6(d) show the output transverse profile of laser 1 for two values of Δ_2, the first one leading to focusing ($\Delta_2 = +15$ GHz) and the second one to an increased defocusing ($\Delta_2 = -10$ GHz).

IV. CONCLUSION

In this paper, we showed that, when two lasers copropagate inside an atomic vapor, with laser 1 nearly resonant with the transition $|0\rangle \to |1\rangle$ and laser 2 nearly resonant with the transition $|1\rangle \to |2\rangle$, as shown in Fig. 2, then the index of refraction of laser 1 is $n = n_0 + n_2 I_1 - n_2' I_2$, where I_1 and I_2 are, respectively, the intensities of lasers 1 and 2. Hence, by choosing I_2 and the detuning to resonance, Δ_2, of laser 2, so that $n_2 I_1 = n_2' I_2$, it is possible to cancel out the Kerr effect on laser 1, which can then propagate with very little distortion of its transverse profile.

Theoretically, this result is obtained through a resolution of the Bloch equations using both a perturbative analysis and the adiabatic approximation. Such approximations are well grounded provided that, for $j = 1$ or 2, $\Omega_j/\Delta_j \ll 1$, and $\Omega_j \tau_j \gg 1$, where τ_j is the time duration of laser j, and where Ω_1 (Ω_2) is the Rabi frequency of transition $|0\rangle \to |1\rangle$ ($|1\rangle \to |2\rangle$).

Moreover, our result on the Kerr effect cancellation mainly applies when the atoms may be considered as three-level systems, which leads to the condition Eq. (28) for an effective cancellation, while accounting for the atoms' hyperfine structure (when the nuclear spin is nonzero) is only a technical difficulty that changes Eq. (28) to Eq. (35). It is noteworthy that, for the chosen example of barium atoms of Sec. II C, Eqs. (28) and (35) are actually the same.

In order to prove the effectiveness of our proposed scheme, we performed an experiment in an atomic vapor of barium. Experimentally, we chose the intensity, I_1, and detuning, Δ_1, of laser 1 so as to obtain, when this laser propagates by itself, a clear single filament at the output of the barium vapor, illustrated in Fig. 1(b). We then let lasers 1 and 2 copropagate and adjusted the intensity, I_2, and detuning, Δ_2, of laser 2 so as to make the output profile of laser 1 as close as possible to its input profile, as may be seen by comparing Fig. 1(c) with Fig. 1(a). Hence, we indeed showed experimentally that, using laser 2, we could strongly reduce the alteration of the transverse profile of laser 1 due to its interaction with the barium vapor. Moreover, the values found for I_2 and Δ_2 are very close to those predicted theoretically (the discrepancy between the experimental and theoretical values is within the uncertainties due to the lack of precision on the value of the dipole moment of the transition $|1\rangle \to |2\rangle$), which validates our theoretical analysis.

By changing the value of Δ_2, we also showed that we could make laser 1 focus while, when propagating alone, it would have self-defocused, or, conversely, increase the defocussing. Using laser 2 we can therefore choose, at will, the output profile of laser 1.

In conclusion, we proposed in this paper quite a general method to cancel out the Kerr effect on a near-resonnant laser propagating inside an atomic vapor by using the crossed-Kerr effect induced by a second, copropagating, laser light. The method is quite efficient, as demonstrated on a barium vapor, and easy to implement. It moreover allows quite a precise control of the laser beam propagation whose transverse size at the vapor output may be chosen at will.

ACKNOWLEDGMENTS

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APPENDIX: THREE-LEVEL SYSTEM WITH HYPERFINE STRUCTURE.

Let us now address the derivation of the polarization, P, when the atoms have a hyperfine structure. As explained in Sec. IIB1, a given hyperfine level, denoted by $|J_i, F_i, m_i\rangle$, is identified by the kinetic momentum, J_i, of its fine level $|i\rangle$, by the value of its total kinetic momentum, $F_i = J_i + \mathcal{I}$, where $\mathcal{I}$ is the nuclear spin, and by the projection, m_i, of F_i along the direction of the laser's electric fields. Because hyperfine sublevels are very close to each other, one needs to account for all allowed transitions, $\Delta F = 0, \pm 1$, between two given fine levels $|i\rangle$ and $|j\rangle$, which makes the derivation of P more complicated than when $\mathcal{I}=0$. However, when the lasers are linearly polarized, as assumed in this paper, the selection rule $\Delta m=0$ makes it possible to consider transitions for a given value of m independently of those for $m'\neq m$. An example of all transitions one has to account for when the ground level is such that $J_0=0$ (as is the case for barium), and for one given value of m, is illustrated in Fig. 3.

In order to account for the atom hyperfine structure, we now write the wave function, $|\Psi^n\rangle$, of atom n,

$$|\Psi^n\rangle=\sum_{F_0}\sum_{m_0=-F_0}^{F_0}c^n_{0,F_0,m_0}|J_0,F_0,m_0\rangle+\sum_{F_1}\sum_{m_1=-F_1}^{F_1}c^n_{1,F_1,m_1}e^{-i\omega_{l_1}t}|J_1,F_1,m_1\rangle+\sum_{F_2}\sum_{m_2=-F_2}^{F_2}c^n_{2,F_2,m_2}e^{-i(\omega_{l_1}+\omega_{l_1})t}|J_2,F_2,m_2\rangle,\quad \text{(A1)}$$

where $\sum_{F_i}$ represents the sum over all possible values of the total kinetic momentum, F_i, of level $|i\rangle$. As for the atom polarization, it is

$$P=\frac{1}{\Delta V}\sum_{n=1}^{\mathcal{N}}\langle\Psi^n|\hat{\mu}|\Psi^n\rangle,\quad \text{(A2)}$$

where ΔV is a small volume of laser-atom interaction (which nevertheless contains a large number, $\mathcal{N}$, of atoms) about a given space location $\vec{r}$.

Just like when the atom has no hyperfine structure, we write the polarization as

$$P=P_1e^{-i\omega_{l_1}t}+P_2e^{-i\omega_{l_2}t},\quad \text{(A3)}$$

and, if we denote

$$\sigma^n_{ij,F_i,F_j,m}\equiv c^n_{j,F_j,m}\left(c^n_{i,F_i,m}\right)^*,\quad \text{(A4)}$$

and if let $\mu_{i,F_i,F_i+l,m}$ be the dipole moment of the transition $|J_i,F_i,m\rangle\to|J_j,F_i+l,m\rangle$, which, by convention, is zero if the transition does not exist [i.e., if $F_i+l<0$, or $F_i+l>\max(F_1)$ or $|m|>\max(F_i+l)$], then

$$P_1=\frac{1}{\Delta V}\sum_{n=1}^{\mathcal{N}}\sum_{F_0}\sum_{m_0=-F_0}^{F_0}\sum_{j=-1}^{1}\mu_{1,F_0,F_0+j,m_0}\sigma^n_{01,F_0,F_0+j,m_0},\quad \text{(A5)}$$

$$P_2=\frac{1}{\Delta V}\sum_{n=1}^{\mathcal{N}}\sum_{F_1}\sum_{m_1=-F_1}^{F_1}\sum_{j=-1}^{1}\mu_{2,F_1,F_1+j,m_1}\sigma^n_{12,F_1,F_1+j,m_1}.\quad \text{(A6)}$$

Using the rotating-wave approximation, neglecting the energy difference between hyperfine sublevels compared to the detunings Δ_1 and Δ_2 of Fig. 2, assuming that the lifetimes of these levels are infinite, and denoting

$$\Omega_{i,F_i,F_i+j,m_i}\equiv\frac{\mu_{i,F_i,F_i+j,m_i}E_i}{\hbar},\quad \text{(A7)}$$

the Rabi frequency of the transition $|J_i,F_i,m_i\rangle\to|J_{i+1},F_i+j,m_i\rangle$, one easily finds the Bloch equations

$$\frac{d\sigma^n_{01,F_0,F_0+j,m}}{dt}=-i\Delta_1\sigma^n_{01,F_0,F_0+j,m_0}+i\sum_{l=-1}^{1}\Omega_{1,F_0+j+l,F_0+j,m_0}\sigma^n_{00,F_0,F_0+j+l,m_0}-i\sum_{l=-1}^{1}\Omega_{1,F_0,F_0+l,m_0}\sigma^n_{11,F_0+l,F_0+j,m_0}+i\sum_{l=-1}^{1}\Omega^*_{2,F_0+j,F_0+j+l,m_0}\sigma^n_{02,F_0,F_0+j+l,m_0},\quad \text{(A8)}$$

$$\frac{d\sigma^n_{12,F_1,F_1+j,m_1}}{dt}=i(\Delta_1-\Delta_2)\sigma^n_{12,F_1,F_1+j,m_1}+i\sum_{l=-1}^{1}\Omega_{2,F_1+j+l,F_1+j,m_1}\sigma^n_{11,F_1,F_1+j+l,m_1}-i\sum_{l=-1}^{1}\Omega^*_{1,F_1+l,F_1,m_1}\sigma^n_{02,F_1+l,F_1+j,m_1}-i\sum_{l=-1}^{1}\Omega_{2,F_1,F_1+l,m_1}\sigma^n_{22,F_1+l,F_1+j,m_1},\quad \text{(A9)}$$

$$\frac{d\sigma^n_{02,F_0,F_0+j,m_0}}{dt}=-i\Delta_2\sigma^n_{02,F_0,F_0+j,m_0}+i\sum_{l=-1}^{1}\Omega_{2,F_0+j+l,F_0+j,m_0}\sigma^n_{01,F_0,F_0+j+l,m_0}-i\sum_{l=-1}^{1}\Omega_{1,F_0,F_0+l,m_0}\sigma^n_{12,F_0+l,F_0+j,m_0},\quad \text{(A10)}$$

where we recall that $\sigma^n_{ij,F_i,F_j,m}\equiv(c^n_{i,F_i,m})^*c^n_{j,F_j,m}$. Just like when the atom has no hyperfine structure, the Bloch equations are solved by perturbation. At 0 order in the field amplitudes, and when the thermal energy is much larger than the energy difference between hyperfine levels, we find,

$$\sigma_{00,F_0,F_0,m_0}=1/g_0,\quad \text{(A11)}$$

where $g_0\equiv\sum_{F_0}(2F_0+1)$ is the number of hyperfine sublevels within the ground level. Then,

$$c^n_{0,F_0,m}=\frac{e^{i\varphi^n_{0,F_0,m}}}{\sqrt{g_0}},\quad \text{(A12)}$$

where $\varphi^n_{0,F_0,m}$ is a phase that depends on the considered atom, n, and that varies randomly from one atom to the other.
By making use of the adiabatic approximation, one finds from Eq. (A8) that, at lowest order in the fields amplitudes,

$$\sigma^n_{01,F_0,F_0+j,m_0}\approx\sum_{l=-1}^{1}\frac{\Omega_{1,F_0+j+l,F_0+j,m_0}\sigma^n_{00,F_0,F_0+j+l,m_0}}{\Delta_1}.\quad \text{(A13)}$$

Plugging Eq. (A12) into Eq. (A13) yields

$$\sigma^n_{01,F_0,F_0+j,m_0} \approx \frac{1}{g_0\Delta_1}\sum_{l=-1}^{1}\Omega_{1,F_0+j+l,F_0+j,m_0}\exp\left[i\left(\varphi^n_{0,F_0+j+l,m_0}-\varphi^n_{0,F_0,m_0}\right)\right]. \tag{A14}$$

Similarly, solving Eq. (A10) by making use of the adiabatic approximation yields, at lowest order in the fields amplitudes,

$$\begin{aligned}\sigma^n_{02,F_0,F_0+j,m_0} &\approx \frac{1}{\Delta_2}\sum_{l=-1}^{1}\Omega_{2,F_0+j+l,F_0+j,m_0}\sigma^n_{01,F_0,F_0+j,m_0}\\ &\approx \frac{1}{g_0\Delta_1\Delta_2}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\left\{\Omega_{2,F_0+j+l,F_0+j,m_0}\Omega_{1,F_0+j+l+l_1,F_0+j+l,m_0}\exp\left[i\left(\varphi^n_{0,F_0+j+l+l_1,m_0}-\varphi^n_{0,F_0,m_0}\right)\right]\right\}.\end{aligned} \tag{A15}$$

From Eqs. (A12), (A14), and (A15) we then find

$$c^n_{1,F_0,m_0} \approx \frac{1}{\sqrt{g_0}\Delta_1}\sum_{l=-1}^{1}\Omega_{1,F_0+l,F_0,m_0}e^{i\varphi^n_{0,F_0+l,m_0}}, \tag{A16}$$

$$c^n_{2,F_0,m_0} \approx \frac{1}{\sqrt{g_0}\Delta_1\Delta_2}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\Omega_{2,F_0+l,F_0,m_0}\Omega_{1,F_0+l+l_1,F_0+l,m_0}e^{i\varphi^n_{0,F_0+l+l_1,m_0}}, \tag{A17}$$

so that

$$\sigma^n_{11,F_1,F_1+j+l,m_1} \approx \frac{1}{g_0\Delta_1^2}\sum_{l_1=-1}^{1}\sum_{l_2=-1}^{1}\left\{\Omega^*_{1,F_1+l_1,F_1,m_1}\Omega_{1,F_1+j+l_1+l_2,F_1+j+l,m_1}\exp\left[i\left(\varphi^n_{0,F_1+j+l+l_2,m_1}-\varphi^n_{0,F_1+l_1,m_1}\right)\right]\right\}, \tag{A18}$$

$$\sigma^n_{12,F_1,F_1+j,m_1} \approx \frac{1}{g_0\Delta_1^2\Delta_2}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\sum_{l_2=-1}^{1}\left\{\Omega_{2,F_1+j+l,F_1+j,m_1}\Omega^*_{1,F_1+l_1,F_1,m_1}\Omega_{1,F_1+j+l+l_2,F_1+j+l,m_1}e^{i\left(\varphi^n_{0,F_1+j+l+l_2,m_1}-\varphi^n_{0,F_1+l_1,m_1}\right)}\right\}, \tag{A19}$$

while $\sigma^n_{22,F_0,F_0+j+l,m_1}$, whose expression will not be given here, is clearly of order 4 in the field amplitudes.

When summing $\sigma^n_{12,F_1,F_1+j,m_1}$ over all atoms, as in Eq. (A6) to find P_2 then, because the phases φ^n_{0,F_0,m_0} vary randomly from one to atom to the other, only terms such that $j+l+l_2=l_1$ give a nonzero contribution. Hence, if we denote by N the atom density and

$$\Omega_1 \equiv \bar{\mu}_1 E_1/\hbar, \tag{A20}$$

$$\Omega_2 \equiv \bar{\mu}_2 E_2/\hbar, \tag{A21}$$

where $\bar{\mu}_1$ and $\bar{\mu}_2$ are, respectively, the mean dipole moments of transitions $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$, then

$$P_2 = N\frac{\Omega_2|\Omega_1|^2}{g_0\Delta_2^2\Delta_1}\sum_{F_1}\sum_{m_1=-F_1}^{F_1}\sum_{j=-1}^{1}\sum_{l=-1}^{1}\sum_{l_1=-1}^{1}\Pi_{F_1,m_1,j,l_1,l_2}, \tag{A22}$$

where

$$\Pi_{F_1,m_1,j,l_1,l_2} = \frac{\mu_{2,F_1,F_1+j,m_1}\mu_{2,F_1+j+l,F_1+j,m_1}\mu_{1,F_1+l_1,F_1,m_1}\mu_{1,F_1+l_1,F_1+j+l,m_1}}{\bar{\mu}_1^2\bar{\mu}_2}. \tag{A23}$$

Let us now calculate P_1, and, therefore, $\sigma^n_{01,F_0,F_0+j,m_0}$, at third order in the field amplitudes. Solving Eq. (A8) by making use of the adiabatic approximation yields

$$\begin{aligned}\Delta_1\sigma^n_{01,F_0,F_0+j,m_0} =& \sum_{l=-1}^{1}\Omega_{1,F_0+j+l,F_0+j,m_0}\sigma^n_{00,F_0,F_0+j+l,m_0}\\ &-\sum_{l=-1}^{1}\Omega_{1,F_0,F_0+l,m_0}\sigma^n_{11,F_0+l,F_0+j,m_0}\\ &+\sum_{l=-1}^{1}\Omega^*_{2,F_0+j,F_0+j+l,m_0}\sigma^n_{02,F_0,F_0+j+l,m_0},\end{aligned} \tag{A24}$$

where $\sigma^n_{11,F_0+l,F_0+j,m_0}$ and $\sigma^n_{02,F_0,F_0+j+l,m_0}$ are, respectively, given by Eqs. (A18) and (A15). Since we are only interested in P_1, we will only calculate $\bar{\sigma}_{01,F_0,F_0+j,m_0}$, which results from the averaging of $\sigma^n_{01,F_0,F_0+j,m_0}$ over all the atoms contained in the considered volume ΔV,

$$\bar{\sigma}_{01,F_0,F_0+j,m_0} \equiv \frac{1}{\mathcal{N}}\sum_{n=1}^{\mathcal{N}}\sigma^n_{01,F_0,F_0+j,m_0}. \tag{A25}$$

From Eq. (A12), it is clear that

$$\bar{\sigma}_{00,F_0,F_0+j,m_0} \equiv \frac{1}{\mathcal{N}}\sum_{n=1}^{\mathcal{N}}\sigma^n_{00,F_0,F_0+j,m_0} \tag{A26}$$

is zero if $j \neq 0$ so that Eq. (A24) yields

$$\Delta_1 \bar{\sigma}_{01,F_0,F_0+j,m_0} = \Omega_{1,F_0,F_0+j,m_0} \bar{\sigma}_{00,F_0,F_0,m_0} - \sum_{l=-1}^{1} \Omega_{1,F_0,F_0+l,m_0} \bar{\sigma}_{11,F_0+l,F_0+j,m_0} + \sum_{l=-1}^{1} \Omega^*_{2,F_0+j,F_0+j+l,m_0} \bar{\sigma}_{02,F_0,F_0+j+l,m_0}, \quad \text{(A27)}$$

and, from Eq. (A18),

$$\bar{\sigma}_{11,F_0+l,F_0+j,m_0} \equiv \frac{1}{\mathcal{N}} \sum_{n=1}^{\mathcal{N}} \sigma^n_{11,F_0+l,F_0+j,m_0} = \frac{1}{g_0 \Delta_1^2} \sum_{l_2=-1}^{1} \Omega^*_{1,F_0+j+l_2,F_0+l,m_0} \Omega_{1,F_0+j+l_2,F_0+j,m_0}, \quad \text{(A28)}$$

while, from Eq. (A15), one finds,

$$\bar{\sigma}_{02,F_0,F_0+j+l,m_0} \equiv \frac{1}{\mathcal{N}} \sum_{n=1}^{\mathcal{N}} \sigma^n_{02,F_0,F_0+j+l,m_0} = \frac{1}{g_0 \Delta_1 \Delta_2} \sum_{l_1=-1}^{1} \Omega_{2,F_0+j+l+l_1,F_0+j+l,m_0} \Omega_{1,F_0,F_0+j+l+l_1,m_0}. \quad \text{(A29)}$$

In order to conclude the derivation of P_1, there only remains to evaluate $\bar{\sigma}_{00,F_0,F_0,m_0}$ at second order in the field amplitudes. This estimate of $\bar{\sigma}_{00,F_0,F_0,m_0}$ can be obtained by considering only the transitions from level $|0\rangle$ to level $|1\rangle$ due to laser 1 and by neglecting the transitions from level $|1\rangle$ to level $|0\rangle$ due to the same laser, because the corresponding contributions to $\bar{\sigma}_{00,F_0,F_0,m_0}$ will be of higher order. Then,

$$\bar{\sigma}_{00,F_0,F_0,m_0} \approx \frac{1}{g_0} \left[1 - \sum_{j=-1}^{1} \frac{|\Omega_{1,F_0,F_0+j,m_0}|^2}{\Delta_1^2} \right]. \quad \text{(A30)}$$

Plugging Eqs. (A28)–(A30) into Eq. (A27) and using the value thus found for $\bar{\sigma}_{01,F_0,F_0+j,m_0}$ into Eq. (A5) for P_1, one finds

$$P_1 = N \frac{E_1}{\hbar} \left[\alpha \bar{\mu}_1^2 - \frac{\beta_1 \bar{\mu}_1^4 |E_1|^2}{\hbar^2 \Delta_1^3} + \frac{\beta_2 \bar{\mu}_1^2 \bar{\mu}_2^2 |E_2|^2}{\hbar^2 \Delta_1^2 \Delta_2} \right], \quad \text{(A31)}$$

where

$$\alpha \equiv \sum_{F_0} \sum_{m_0=-F_0}^{F_0} \sum_{j=-1}^{1} \frac{\mu^2_{1,F_0,F_0+j,m_0}}{g_0 \Delta_1 \bar{\mu}_1^2}, \quad \text{(A32)}$$

and where

$$\beta_1 \equiv \frac{1}{g_0 \bar{\mu}_1^4} \sum_{F_0} \sum_{m_0=-F_0}^{F_0} \left\{ \left(\sum_{j=-1}^{1} \mu^2_{1,F_0,F_0+j,m_0} \right)^2 + \sum_{j=-1}^{1} \sum_{l=-1}^{1} \sum_{l_1=-1}^{1} \left[\mu_{1,F_0,F_0+j,m_0} \mu_{1,F_0,F_0+l,m_0} \mu_{1,F_0+j+l_1,F_0+l,m_0} \mu_{1,F_0+j+l_1,F_0+j,m_0} \right] \right\}, \quad \text{(A33)}$$

$$\beta_2 \equiv \frac{1}{\bar{\mu}_1^2 \bar{\mu}_2^2} \sum_{F_0} \sum_{m_0=-F_0}^{F_0} \sum_{j=-1}^{1} \left\{ \mu_{1,F_0,F_0+j,m_0} \sum_{l=-1}^{1} \sum_{l_1=-1}^{1} \left[\mu_{2,F_0+j,F_0+j+l,m_0} \mu_{2,F_0+j+l+l_1,F_0+j+l,m_0} \mu_{1,F_0,F_0+j+l+l_1,m_0} \right] \right\}. \quad \text{(A34)}$$

The condition for a Kerr-free propagation of laser 1 can thus now be written

$$\frac{I_2}{I_1} = \frac{\beta_1 \bar{\mu}_1^2 \Delta_2}{\beta_2 \bar{\mu}_2^2 \Delta_1}. \quad \text{(A35)}$$

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