Electromagnetically-induced-transparency-enhanced Kerr nonlinearity: Beyond steady-state treatment

L. Deng,[1] M. G. Payne,[2] and W. R. Garrett[3,4]

[1]*Electron & Optical Physics Division, NIST, Gaithersburg, Maryland 20899-8410*

[2]*Department of Physics, Georgia Southern University, Statesboro, Georgia 30460*

[3]*Department of Physics, University of Tennessee, Knoxville, Tennessee 37996*

[4]*Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*

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A time-dependent perturbation calculation for a four-level system is presented. A resonantly enhanced Kerr nonlinearity is produced with a combination of long, short, and delayed laser pulses in the presence of electromagnetically induced transparency. We show the enhanced Kerr nonlinearity and vanishingly small linear susceptibility due to the induced transparency, both are favorable for cross-phase modulation. In addition, we show that possible constructive and destructive interference between different excitation pathways could also lead to enhancement and suppression of the Kerr nonlinearity.

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I. INTRODUCTION

The optical Kerr effect has been one of the most extensively studied phenomenon in the field of nonlinear optics because of its applications ranging from frequency conversions [1] to quantum nondemolition measurements [2]. Recently, Schmidt and co-workers [3] have proposed a nonlinear scheme based on electromagnetically induced transparency (EIT) [4] to enhance the magnitude of the cross-phase modulation. Their analysis, which is based on a steady-state solution with the wave-mixing process excluded, has predicted a dramatic increase of Kerr nonlinearities, making the EIT-assisted scheme a very attractive candidate for cross-phase modulation. Here, we present a time-dependent perturbation treatment on EIT-enhanced Kerr nonlinearity in the nonsteady-state regime (e.g., short-pulse excitation). Four features of the present work distinguish itself from the previous steady-state treatment: (1) The non-steady-state theory permits the study of the dynamics of the system, thereby providing an important extension to the strong, pulsed excitation regime (2) The delayed-pulse sequence eliminates the fast oscillation normally encountered when strong lasers are tuned on resonance, and therefore permits the acquisition of a reliable adiabatic solution to the problem (3) The allowed transition back to the ground state introduces rich dynamics, such as mixing-wave generation and quantum interference due to different excitation pathways. We note that the inclusion of the generated field, under suitable conditions, could lead to the enhancement or suppression of the Kerr nonlinearity, an effect that may have important consequences in other nonlinear processes such as spectral line narrowing (4) Selective injection of the source pulse at the optimized atomic coherence enhances the nonlinearity, and allows a clean analytical solution. To the best of our knowledge, the time-dependent EIT-assisted Kerr effect with a wave-mixing channel open has never been treated before. Our paper is organized in the following way: in Sec. II we present the model and solve both the atomic equation of motion and Maxwell equations for the mixing wave and source laser field. In Sec. III we calculate the EIT-enhanced Kerr coefficient. In Sec. IV a discussion of the features and limiting cases is given, and in Sec. V we present a summary.

II. THE MODEL AND SOLUTIONS OF EQUATIONS OF MOTION

The four-level system under investigation is depicted in Fig. 1(a). The system is assumed to first interact with two intense long pulse laser fields. A coupling laser (ω_{L2}) is tuned onto the $|1\Leftrightarrow|2\rangle$ transition. A weak probe laser (ω_{L1}) is tuned to the line center of the $|0\rangle\Leftrightarrow|2\rangle$ transition. Due to the fact that the contributions to the index from the members of the induced Autler-Townes doublet cancel each other out, the probe laser suffers no absorption.

Let us assume that both lasers are unfocused, have a transform-limited bandwidth, and have a pulse length of typically ~10 ns. We now introduce a weak short-pulse laser (ω_S), propagating colinearly with the two long-pulse lasers, to couple the states $|1\rangle$ and $|3\rangle$, which has a dipole-allowed transition back to the ground state. We point out that since the wave-mixing process can be highly efficient, one must treat the wave-mixing field and the source field on an equal basis. Therefore, in addition to four equations of motion for probability amplitudes of atomic wave function, there are two Maxwell equations for the field amplitudes that must be solved simultaneously. This is, at first glance, a formidable task. However, by carefully examining various terms we can simplify the calculation greatly. The first key point is that there are two very different time scales: the long-pulse ($\tau_L \sim 10$ ns) scale associated with the EIT process and the short-pulse scale ($\tau_S \sim 10$ ps) associated with the source laser for $|1\rangle\Leftrightarrow|3\rangle$ coupling [5]. The second key point is the use of a counterintuitive pulse sequence. The former allows one to freeze most of atomic parameters while calculating the nonlinear response of the system to the short pulse, whereas the latter ensures a robust adiabatic transfer leading to well-behaved atomic parameters that can be frozen out during the calculation of the nonlinear response of the system to the short-pulse excitation.

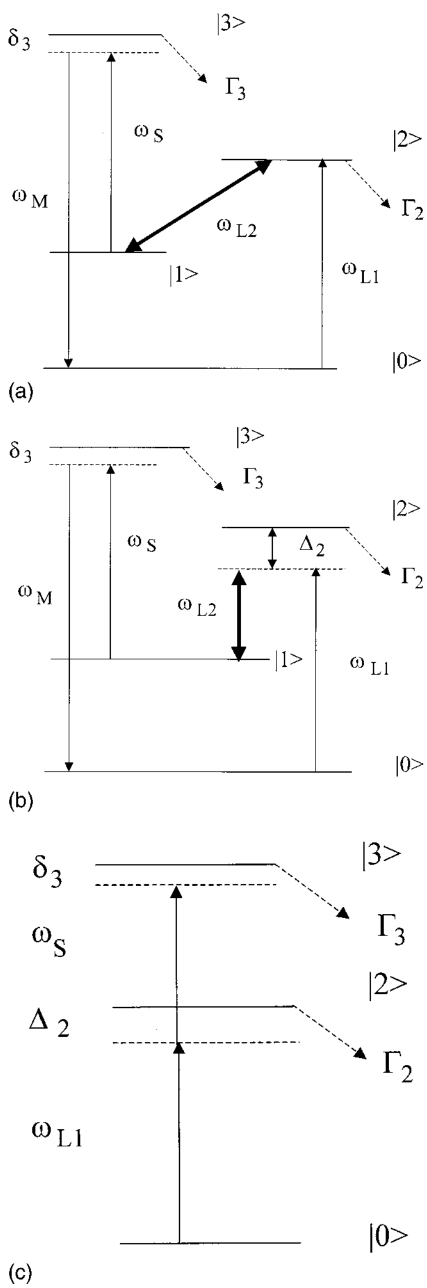

FIG. 1. (a) EIT-assisted four-level system. ω_{L1}, ω_{L2}, ω_S, and ω_M are probe, coupling, source, and mixing fields. (b) Non-EIT-assisted four-level system. Notations for various fields are that of (a). (c) Conventional three-level system for cross-phase modulation.

Let us assume the wave function of the form

$$|\Psi\rangle=\sum_{n=0}^{3} a_n(t)e^{-i\omega_n t}|n\rangle. \tag{1}$$

A set of equations of motion for the system depicted in Fig. 1(a) can be obtained as

$$\dot{b}_0=i\frac{\Omega_{02}}{2}b_2+i\frac{\Omega_{03}}{2}b_3, \tag{2a}$$

$$\dot{b}_1=i\frac{\Omega_{12}}{2}b_2+i\frac{\Omega_{13}}{2}b_3, \tag{2b}$$

$$\dot{b}_2=i\frac{\Omega_{20}}{2}b_0+i\frac{\Omega_{21}}{2}b_1-\frac{\Gamma_2}{2}b_2, \tag{2c}$$

$$\dot{b}_3=i\frac{\Omega_{30}}{2}b_0+i\frac{\Omega_{31}}{2}b_1+i\left(\delta_3+i\frac{\Gamma_3}{2}\right)b_3. \tag{2d}$$

As usual, $\Omega_{ij}=D_{ij}E/\hbar$ is the Rabi frequency for the relevant transition, and we have introduced $\delta_1=\omega_{L1}-\omega_{20}=0$, $\delta_2=\omega_{L2}-\omega_{21}=0$, and $\delta_3=\omega_{L3}-\omega_{31}=\omega_M-\omega_{30}$. We first solve Eqs. (2a)–(2c) by neglecting Ω_{31},Ω_{30}. This is because the source laser and the mixing fields are generally weak in comparison with the lasers used for creating EIT process, therefore they will be treated as perturbation. Based on this consideration, we have the zeroth-order equations of motion

$$\dot{b}_0^{(0)}=i\frac{\Omega_{02}}{2}b_2^{(0)}, \tag{3a}$$

$$\dot{b}_1^{(0)}=i\frac{\Omega_{12}}{2}b_2^{(0)}, \tag{3b}$$

$$\dot{b}_2^{(0)}=i\frac{\Omega_{20}}{2}b_0^{(0)}+i\frac{\Omega_{21}}{2}b_1^{(0)}. \tag{3c}$$

where we have neglected Γ_2 for the zeroth order due to the on resonance excitation. It is known that for a counterintuitive pulse sequence (i.e., laser ω_{L2} is applied first and laser ω_{L1} is applied at a delayed time) the solutions to Eqs. (3a)–(3c) can be obtained accurately to give the elements of density matrix as [6,7]

$$\rho_{00}^{(0)}=\frac{|\Omega_{12}|^2}{|\Omega_{12}|^2+|\Omega_{20}|^2}, \tag{4a}$$

$$\rho_{10}^{(0)}=-\frac{\Omega_{02}\Omega_{21}}{|\Omega_{12}|^2+|\Omega_{12}|^2}, \tag{4b}$$

$$\rho_{11}^{(0)}=\frac{|\Omega_{20}|^2}{|\Omega_{12}|^2+|\Omega_{20}|^2}, \tag{4c}$$

$$\rho_{20}^{(0)}=0,\ \ \rho_{12}^{(0)}=0,\ \ \rho_{22}^{(0)}=0, \tag{4d}$$

where $|\Omega|^2=|\Omega_{12}|^2+|\Omega_{20}|^2$. The fact that $\rho_{20}^{(0)}\approx 0$ and $\rho_{12}^{(0)}\approx 0$ implies that these laser pulses propagate with no distortion at a velocity nearly equal to the speed of light in vacuum, as expected for the EIT process. Of course, with the definition of $\kappa_{ji}=2\pi|D_{ji}|^2(\omega_i-\omega_j)/\hbar c$, the criteria

$$\frac{\kappa_{02}c}{|\Omega_{02}|^2+|\Omega_{12}|^2}\ll 1, \tag{5}$$

$$\frac{\kappa_{12}c}{|\Omega_{02}|^2+|\Omega_{12}|^2}\ll 1 \tag{6}$$

should be fulfilled in order to avoid significant group velocity mismatches between both long and short pulses.

To find the next-order correction we take $b_n = b_n^{(0)} + b_n^{(1)}$ and obtain the following equations of motion for the amplitudes that are in the first order of Ω_{31}, Ω_{30}:

$$\dot{b}_0^{(1)} = i\frac{\Omega_{02}}{2} b_2^{(1)} + i\frac{\Omega_{03}}{2} b_3^{(0)}, \tag{7a}$$

$$\dot{b}_1^{(1)} = i\frac{\Omega_{12}}{2} b_2^{(1)} + i\frac{\Omega_{13}}{2} b_3^{(0)}, \tag{7b}$$

$$\dot{b}_2^{(1)} = i\frac{\Omega_{20}}{2} b_0^{(1)} + i\frac{\Omega_{21}}{2} b_1^{(1)} - \frac{\Gamma_2}{2} b_2^{(1)}, \tag{7c}$$

$$\dot{b}_3^{(0)} = i\frac{\Omega_{30}}{2} b_0^{(0)} + i\frac{\Omega_{31}}{2} b_1^{(0)} + i\left(\delta_3 + i\frac{\Gamma_3}{2}\right) b_3^{(0)}. \tag{7d}$$

Since the pulse width of both the source laser and the mixing wave are much weaker and shorter than that of the probe and coupling lasers (i.e., $\tau_S \ll \tau_L$) we expect that the values of ρ_{00}, ρ_{10}, and ρ_{11} will remain nearly constant throughout the short pulse. This allows us to fix the time at, for instance, $t = t_d$ (where t_d is the delayed short-pulse injection time) for all zeroth-order solutions during the evaluation of the source and mixing fields.

We Fourier transform Eq. (7d) and obtain

$$\tilde{b}_3^{(0)}(\omega) = e^{ik_M z} \frac{M(z,\omega) b_0^{(0)}(t_d) + S(z,\omega) e^{i\Delta k z} B_1^{(0)}(t_d)}{2(\omega - \delta_3 - i\Gamma_3/2)}, \tag{8}$$

where

$$\tilde{b}_3^{(0)}(\omega) = \int_{-\infty}^{+\infty} \frac{dt}{\sqrt{2\pi}} b_3^{(0)}(t) e^{-i\omega t},$$

$$M(\omega) = \int_{-\infty}^{+\infty} \frac{dt}{\sqrt{2\pi}} \Omega_{30}(t) e^{-i\omega t},$$

$$S(\omega) = \int_{-\infty}^{+\infty} \frac{dt}{\sqrt{2\pi}} \Omega_{31}(t) e^{-i\omega t},$$

respectively. The Maxwell equations for the mixing wave and the source field take the form

$$\frac{\partial \Omega_{30}}{\partial z} + \frac{1}{c}\frac{\partial \Omega_{30}}{\partial t} = i\kappa_{03}\rho_{30} e^{-ik_M z} \approx i\kappa_{03} b_3^{(0)}(t) b_0^{(0)*}(t) e^{-ik_M z}, \tag{9a}$$

$$\frac{\partial \Omega_{31}}{\partial z} + \frac{1}{c}\frac{\partial \Omega_{31}}{\partial t} = i\kappa_{13}\rho_{31} e^{-ik_S z} \approx i\kappa_{13} b_3^{(0)}(t) b_1^{(0)*}(t) e^{-ik_S z}. \tag{9b}$$

Taking a fixed time $t = t_d$ for $b_0^{(0)}(t)$ and $b_1^{(0)}(t)$, carrying out Fourier transform on the both sides of Eq. (9), we obtain

$$\frac{\partial M}{\partial z} + i\frac{\omega}{c} M = i\kappa_{03} \tilde{b}_3^{(0)}(\omega) b_0^{(0)*} e^{-ik_M z} = \frac{i\kappa_{03}(M\rho_{00}^{(0)} + S e^{i\Delta k z}\rho_{10}^{(0)})}{2(\omega - \delta_3 - i\Gamma_3/2)}, \tag{10a}$$

$$\frac{\partial S}{\partial z} + i\frac{\omega}{c} S = i\kappa_{13} \tilde{b}_3^{(0)}(\omega) b_1^{(0)*} e^{-ik_S z} = \frac{i\kappa_{13}(M e^{-i\Delta k z}\rho_{01}^{(0)} + S\rho_{11}^{(0)})}{2(\omega - \delta_3 - i\Gamma_3/2)}. \tag{10b}$$

Under the condition of phase matching, $\Delta k = k_S - k_M + k_{L1} - k_{L2} = 0$, Eqs. (10a) and (10b) can be solved directly to yield solutions for the generated wave and attenuated source field. Assuming a Gaussian pulse profile for the source field at the entrance of the cell, we obtain from Eqs. (10a) and (10b)

$$M(z,\omega) = M_0 e^{-\omega^2 \tau_S^2/4 - i\omega z/c - i\omega t_d}(1 - e^{iK(\omega)z}), \tag{11a}$$

$$S(z,\omega) = S_0 e^{-\omega^2 \tau_S^2/4 - i\omega z/c - i\omega t_d}\left(1 + \frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}} e^{iK(\omega)z}\right), \tag{11b}$$

where

$$M_0 = -\frac{\sqrt{\pi}\kappa_{03}\tau_S\rho_{10}^{(0)}}{\kappa_{03}\rho_{00}^{(0)} + \kappa_{13}\rho_{11}^{(0)}} \Omega_{31}(0,0) \tag{12a}$$

and

$$S_0 = \frac{\sqrt{\pi}\kappa_{03}\tau_S\rho_{00}^{(0)}}{\kappa_{03}\rho_{00}^{(0)} + \kappa_{13}\rho_{11}^{(0)}} \Omega_{31}(0,0). \tag{12b}$$

In Eqs. (11) and (12), we have also introduced

$$K(\omega) = \frac{\kappa_{03}\rho_{00}^{(0)} + \kappa_{13}\rho_{11}^{(0)}}{2(\omega - \delta_3 - i\Gamma_3/2)}. \tag{13}$$

The frequency dependence in $K(\omega)$ implies that pulse distortions have occurred, and both the generated wave and the source filed are not pure Gaussian. However, if $e^{ik(\omega)z}$ oscillates fast, its contribution to the inverse Fourier transform integral would be small. In this limit, Eqs. (11) and (12) will lead to a mixing wave and source field traveling with the speed of light in vacuum with undistorted pulse profile equals to that of the source field at the entrance of the cell. This is the ‘‘matched pulse’’ limit [4].

Using Eqs. (8), (11), and (12), we now solve

$$\dot{b}_0^{(1)} = i\frac{\Omega_{02}}{2} b_2^{(1)} + i\frac{\Omega_{03}}{2} b_3^{(0)}, \tag{14a}$$

$$\dot{b}_1^{(1)} = i\frac{\Omega_{12}}{2} b_2^{(1)} + i\frac{\Omega_{13}}{2} b_3^{(0)}, \tag{14b}$$

$$\dot{b}_2^{(1)} = i\frac{\Omega_{20}}{2}b_0^{(1)} + i\frac{\Omega_{21}}{2}b_1^{(1)} - \frac{\Gamma_2}{2}b_2^{(1)}. \quad (14c)$$

A simple perturbation analysis indicates that only $b_2^{(1)}(t)$ is needed for the calculation of Kerr nonlinearity. Taking the Fourier transform on both sides of Eqs. (14a)–(14c), solving for $\tilde{b}_2^{(1)}(\omega)$, and then taking the inverse Fourier transform, we obtain

$$\rho_{20}^{(1)} = b_2^{(1)}(t)b_0^{(0)*}(t_d) = \int_{-\infty}^{+\infty}\frac{d\omega}{\sqrt{2\pi}}e^{i\omega t}\int_{-\infty}^{+\infty}\frac{d\omega'}{\sqrt{2\pi}}\alpha(\omega,\omega')\beta(\omega,\omega'), \quad (15)$$

where we have defined

$$\alpha = \frac{\Omega_{20}(t_d)M^*(\omega') + \Omega_{21}(t_d)S^*(\omega')}{(4\omega^2 - |\Omega|^2 - i2\omega\Gamma_2)},$$

$$\beta = \frac{\rho_{00}^{(0)}(t_d)M(\omega'+\omega) + \rho_{10}^{(0)}(t_d)S(\omega'+\omega)}{2(\omega'+\omega-\delta_3-i\Gamma_3/2)}. \quad (16)$$

In deriving this result, we have taken $\Omega_{20}(t_d)$ and $\Omega_{21}(t_d)$ as constants while evaluating first-order corrections to the atomic wave function. This is justified because these quantities vary slowly in comparison with the short-pulse source laser.

An important feature is immediately noticed from Eq. (16). That is, the inclusion of the wave-mixing field could lead to the cancellation or enhancement of $\rho_{20}^{(1)}$ when $\Omega_{20}M^*(\omega) = \pm\Omega_{21}S^*(\omega)$ and/or $\rho_{00}^{(0)}M(\omega) = \pm\rho_{10}^{(0)}S(\omega)$ are satisfied. The situation leading to cancellation should be avoided for cross-phase modulation, but it may have applications in other Kerr-related nonlinear processes, such as spectral line narrowing and elimination.

III. ELECTROMAGNETICALLY-INDUCED-TRANSPARENCY-ENHANCED KERR NONLINEARITY

To evaluate Kerr nonlinearity, we calculate the polarization for the probe frequency as follows. Using Eqs. (11), (12), (15), and (16), we calculate the polarization responsible for the probe absorption

$$\begin{aligned}P^{\rm NL}(\omega_p=\omega_{L1}) &= ND_{02}\rho_{20}^{(1)} \\ &= \frac{\pi\tau_S^2\kappa_{03}^2\rho_{00}^{(0)}|\rho_{10}^{(0)}|^2}{(\kappa_{03}\rho_{00}^{(0)}+\kappa_{13}\rho_{11}^{(0)})^2}\frac{N|D_{20}|^2|D_{31}|^2}{\hbar^3}|E_S|^2E_{L1}^{(+)}E^{ik_{L1}z} \\ &\times\int_{-\infty}^{+\infty}\frac{d\omega}{\sqrt{2\pi}}e^{i\omega t}\int_{-\infty}^{+\infty}\frac{d\omega'}{\sqrt{2\pi}}\frac{\left(m^*(\omega') - \dfrac{\rho_{00}^{(0)}\Omega_{21}(t_d)}{\rho_{01}^{(0)}\Omega_{20}(t_d)}s^*(\omega')\right)[m(\omega'+\omega)-s(\omega'+\omega)]}{2(\omega'-\delta_3-i\Gamma_3/2)(4\omega^2-|\Omega|^2-i2\Gamma_2)} \\ &= 3\chi^{(3)}(\omega_{L1})|E_S|^2E_{L1}^{(+)}e^{ik_{L1}z},\end{aligned} \quad (17)$$

where we have introduced two dimensionless quantities for a Gaussian source laser pulse

$$m(z,\omega) = e^{-\omega^2\tau_S^2/4 - i\omega z/c - i\omega t_d}(1-e^{iK(\omega)z}), \quad (18a)$$

$$s(z,\omega) = e^{-\omega^2\tau_S^2/4 - i\omega z/c - i\omega t_d}\left(1 + \frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}}e^{iK(\omega)z}\right). \quad (18b)$$

Let $\eta = \omega\tau_S$ and we immediately obtain

$$\frac{\chi^{(3)}(\omega_{L1})}{\chi_0^{(3)}} = \int_{-\infty}^{+\infty}\frac{d\eta}{\sqrt{2\pi}}e^{i\eta t/\tau_S}\int_{-\infty}^{+\infty}\frac{d\eta'}{\sqrt{2\pi}}\frac{\left(m^*(\eta') - \dfrac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}s^*(\eta')\right)}{(\eta'+\eta-\delta_3\tau_S-i\Gamma_3\tau_S/2)}\frac{m(\eta'+\eta)-s(\eta'+\eta)}{(4\eta^2-|\Omega|^2\tau_S^2-i2\eta\Gamma_2\tau_S)}, \quad (19)$$

where

$$\chi_0^{(3)} = \frac{\pi\tau_S^3\rho_{00}^{(0)}}{6}\frac{|\rho_{10}^{(0)}|^2}{\left(\rho_{00}^{(0)}+\dfrac{\kappa_{13}}{\kappa_{03}}\rho_{11}^{(0)}\right)^2}\frac{N|D_{20}|^2|D_{31}|^2}{\hbar^3}. \quad (20)$$

Equation (19) is the main result of the present study, which is very different from that obtained with a steady-state theory. In particular, it allows the possibility of enhancing and canceling the Kerr nonlinearity via constructive or destructive interference. We also notice important features of the EIT-assisted four-level system. We see that the denominator in Eq. (19) contains the Rabi frequency of the coupling laser $|\Omega_{12}|$ rather than a one-photon detuning Δ_2 for a non-EIT-assisted four-level system [Fig. 1(b)]. The latter usually is much larger than $|\Omega_{12}|$ in order to avoid significant absorption to the probe laser. Therefore, the EIT-assisted four-level system will have significant enhancement to the Kerr nonlinearity with negligible one-photon absorption, particularly in the cases where EIT process can be created at fairly low laser intensity.

IV. DISCUSSIONS

Now we are ready to see some simple cases. First, we neglect the wave-mixing field by choosing the angular momentum of the level $|3\rangle$ such that the coupling to the ground state is defeated. Taking $m(\eta)=0$, from Eq. (19) we have

$$\frac{\chi^{(3)}(\omega_{L1})}{\chi_0^{(3)}}=\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}\int_{-\infty}^{+\infty}\frac{d\eta}{\sqrt{2\pi}}e^{i\eta t/\tau_S}\int_{-\infty}^{+\infty}\frac{d\eta'}{\sqrt{2\pi}}\frac{s^*(\eta')}{(4\eta^2-|\Omega|^2\tau_S^2-i2\eta\Gamma_2\tau_S)}\frac{s(\eta'+\eta)}{(\eta'+\eta-\delta_3\tau_S-i\Gamma_3\tau_S/2)}. \tag{21}$$

We see that if the source field is a plane wave of frequency ω (therefore one can apply the steady-state treatment), then $s(\omega)=\delta(\omega)$, and Eq. (19) reduces to that of Ref. [3],

$$\chi^{(3)}(\omega_{L1})\propto\frac{1}{|\Omega|^2(\delta_3+i\Gamma_3/2)}\approx\frac{1}{|\Omega_{12}|^2(\delta_3+i\Gamma_3/2)}\quad\text{for }|\Omega_{12}|>|\Omega_{02}|\text{ and }|\Omega|>\Gamma_2. \tag{22}$$

For the short-pulse operation, however, even at the nondepleted pump regime, Eq. (19) will yield a result that is very different from the steady-state treatment since the latter cannot provide any dynamic information of the system.

For a Gaussian source field, Eqs. (18) and (19) lead to

$$\frac{\chi^{(3)}(\omega_{L1})}{\chi_0^{(3)}}=\int_{-\infty}^{+\infty}\frac{d\eta}{\sqrt{2\pi}}e^{i\eta\zeta}X(\eta),$$

where

$$X(\eta)=\int_{-\infty}^{+\infty}\frac{d\eta'}{\sqrt{2\pi}}\frac{e^{-\eta'^2/2-\eta'\eta/2-\eta^2/4}}{2(\eta'+\eta-\delta_3\tau_S-i\Gamma_3\tau_S/2)(4\eta^2-|\Omega|^2\tau_S^2-i2\eta\Gamma_2\tau_S)}e^{iK(\eta'+\eta)\xi}\left[\left(\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}-1\right)+\left(1+\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}\frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}}\right)e^{-iK(\eta')\xi}\right]\left(1+\frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}}\right).$$

In deriving these results, we have used dimensionless quantities $\xi=z/c\tau_S$, $\zeta=(t-t_d-z/c)/\tau_S$.

Let us first examine the "matched pulse" regime [4]. From Eqs. (18a) and (18b) we see that if $K(\omega)z\gg1$ after a characteristic propagation distance, the terms involving $e^{iK(\psi)z}$ will oscillate very fast and contribute negligibly, on the average, to the integral. If we neglect these terms, then both of the generated and the source fields are given as

$$m(\eta)=s(\eta)=\exp\left[-\frac{\eta^2}{4}-i\eta\left(\frac{z/c+t_d}{\tau_S}\right)\right].$$

This is to say that both the mixing wave and the source field travel with the speed of light in vacuum, without any pulse shape distortion due to dispersion, and therefore will have an identical "matched pulse" shape. This immediately leads to a conclusion of zero Kerr nonlinearity. This is because those two fields interfere destructively as they propagate in the medium. Of course, one should keep both the concentration and the power of the source laser to a level that no significant population will be put in state $|3\rangle$.

Next, we examine the large detuning regime, i.e., the regime where $\delta_3\tau_S\gg1$. This is the regime where it is known to give high conversion efficiency for the mixing wave [8]. In this regime, $K(\omega)z$ is small yet the frequency dependence can be removed since most of the contribution to the integral by the Gaussian envelope occurs when $|\eta|=|\omega\tau_S|\leqslant4$. In this limit, we have

$$K(\omega)z=\frac{\kappa_{03}\rho_{00}^{(0)}+\kappa_{13}\rho_{11}^{(0)}}{2(\omega-\delta_3-i\Gamma_3/2)}z\approx-\frac{\kappa_{03}c\tau_S^2\rho_{00}^{(0)}+\kappa_{13}c\tau_S^2\rho_{11}^{(0)}}{2(\delta_3\tau_S+i\Gamma_3\tau_S/2)}\frac{z}{c\tau_S}=K_0\xi$$

and

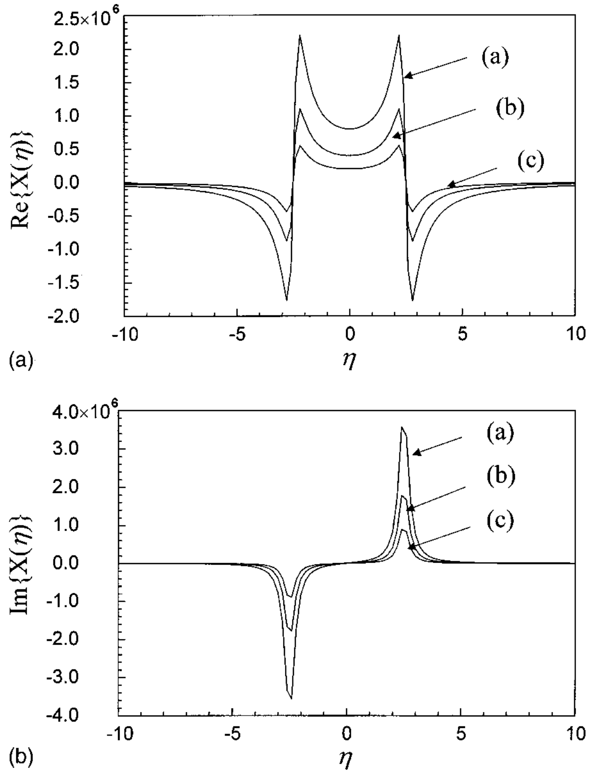

FIG. 2. (a) A plot of the dimensionless quantity Re$\{X(\eta)\}$ as a function of the transform parameter η for different atomic coherences: (a) $\rho_{00}\approx 0.8$, $\rho_{10}\approx 0.4$; (b) $\rho_{00}\approx 0.5$, $\rho_{10}\approx 0.5$; (c) $\rho_{00}\approx 0.2$, $\rho_{10}\approx 0.4$. (b) A plot of the dimensionless quantity Im$\{X(\eta)\}$ as a function of the transform parameter η for different atomic coherences: (a) $\rho_{00}\approx 0.8$, $\rho_{10}\approx 0.4$; (b) $\rho_{00}\approx 0.5$, $\rho_{10}\approx 0.5$; (c) $\rho_{00}\approx 0.2$, $\rho_{10}\approx 0.4$.

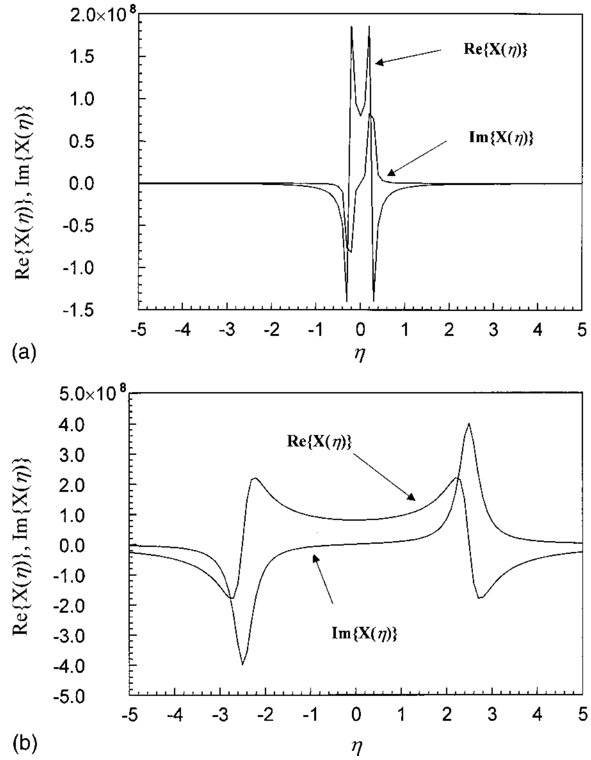

FIG. 3. (a) A plot of the dimensionless quantities Re$\{X(\eta)\}$ and Im$\{X(\eta)\}$ as a function of the transform parameter η. Parameter used: $\rho_{00}\approx 0.8$, $\Omega_{21}\approx 500$ MHz, $\Omega_{20}\approx 10$ MHz, $\Gamma_2\approx 100$ MHz, $\Gamma_3\approx 100$ MHz. (b) A plot of the dimensionless quantities Re$\{X(\eta)\}$ and Im$\{X(\eta)\}$ as a function of the transform parameter η. Parameter used: $\rho_{00}\approx 0.8$, $\Omega_{21}\approx 50$ MHz, $\Omega_{20}\approx 1$ MHz, $\Gamma_2\approx 10$ MHz, $\Gamma_3\approx 10$ MHz.

$$\frac{1}{(\eta'-\delta_3\tau_S-i\Gamma_3\tau_S/2)}\approx-\frac{1}{\delta_3\tau_S}\left(1+\frac{\eta'-i\Gamma_3\tau_S/2}{\delta_3\tau_S}\right),$$

therefore

$$X(\eta)=\frac{C_0 e^{-\eta^2/8}}{(4\eta^2-|\Omega|^2\tau_S^2-i2\eta\Gamma_2\tau_S)}\left(1-i\frac{\Gamma_3}{2\delta_3}+\frac{\eta}{2\delta_3\tau_S}\right),$$

where

$$C_0=-\frac{1}{2\delta_3\tau_S}\left[\left(\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}-1\right)e^{iK_0\xi}+\left(1+\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}\frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}}\right)\right]\left(1+\frac{\kappa_{13}\rho_{11}^{(0)}}{\kappa_{03}\rho_{00}^{(0)}}\right).$$

Although the above expression is given in the Fourier-transform domain, the behavior of this Kerr nonlinearity in the time domain can be deduced easily. In Fig. 2 we have plotted both the real and imaginary parts of $X(\eta)$ as a function of the Fourier transform parameter η for different atomic coherence. It can be seen that the large enhancement does not occur at the maximum atomic coherence. In Fig. 3 we plot the real and imaginary parts of $X(\eta)$ as a function of η for different pump-probe intensities. It is clear that the enhancement is achieved with relative lower pump and probe intensities, as expected from Eq. (22).

Before ending the discussion here, let us look one example where we take $\tau_L=50$ ns and $\tau_S=1$ ns therefore, $\tau_S/\tau_L=1/50\ll 1$. (Note that with this choice of pulse lengths, we expect the leading term of the result to be identical to that of steady-state treatment.) For a modest transition, a combined Rabi frequency $|\Omega|=10\,\mathrm{cm}^{-1}$ would be sufficient to make $|\Omega|\tau_S=300\gg(2\eta)\approx 10$. However, when $\eta^2/8>4$ the contribution from the Gaussian envelope to the integral is negligible. This permits a Taylor expansion of the denominator in the expression of $X(\eta)$, thereby allowing an analytical result of Eq. (10) to be obtained. We therefore have

$$X(\eta)=-\frac{1}{|\Omega|^2\tau_S^2}\frac{C_0 e^{-\eta^2/8}}{1-\dfrac{4\eta^2-i2\eta\Gamma_2\tau_S}{|\Omega|^2\tau_S^2}}\left(1-i\frac{\Gamma_3}{2\delta_3}+\frac{\eta}{2\delta_3\tau_S}\right)$$

$$\approx-\frac{C_0 e^{-\eta^2/8}}{|\Omega|^2\tau_S^2}\left(1+\frac{4\eta^2-i2\eta\Gamma_2\tau_S}{|\Omega|^2\tau_S^2}\right)\times\left(1-i\frac{\Gamma_3}{2\delta_3}+\frac{\eta}{2\delta_3\tau_S}\right)$$

$$=e^{-\eta^2/8}(A_0+A_1\eta+A_2\eta^2+A_3\eta^3),$$

where

$$A_0 = -\frac{C_0}{|\Omega|^2\tau_S^2}\left(1 - i\frac{\Gamma_3}{2\delta_3}\right),$$

$$A_1 = -\frac{C_0}{|\Omega|^2\tau_S^2}\left[\frac{1}{2\delta_3\tau_S} - i\frac{2\Gamma_2\tau_S}{|\Omega|^2\tau_S^2}\left(1 - i\frac{\Gamma_3}{2\delta_3}\right)\right],$$

$$A_2 = -\frac{C_0}{|\Omega|^2\tau_S^2}\left[\frac{4}{|\Omega|^2\tau_S^2}\left(1 - i\frac{\Gamma_3}{2\delta_3}\right) - i\frac{2\Gamma_2\tau_S}{|\Omega|^2\tau_S^2}\frac{1}{2\delta_3\tau_S}\right],$$

$$A_3 = -\frac{C_0}{|\Omega|^2\tau_S^2}\frac{1}{2\delta_3\tau_S}\frac{4}{|\Omega|^2\tau_S^2}.$$

Now the inverse Fourier transform can be carried out analytically, yielding

$$\frac{\chi^{(3)}(\omega_{L1})}{\chi_0^{(3)}} = \int_{-\infty}^{+\infty}\frac{d\eta}{\sqrt{2\pi}}e^{i\eta\zeta}X(\eta)$$
$$\approx \frac{e^{-2\zeta^2}}{\sqrt{2}}\left\{A_0 + iA_1\sqrt{2}\zeta + A_2\frac{1-4\zeta^2}{2} + iA_3\frac{\zeta(-3+4\zeta^2)}{\sqrt{2}}\right\}$$
$$= e^{-2\zeta^2}(B_0 + B_1\zeta + B_2\zeta^2 + B_3\zeta^3),$$

where

$$B_0 = \frac{1}{\sqrt{2}}\left(A_0 + \frac{A_3}{2}\right), \quad B_1 = i\left(A_1 - \frac{3A_3}{2}\right),$$

$$B_2 = -\sqrt{2}A_2, \quad B_3 = i2A_3.$$

As can be seen, the dominate term is the first term in the expression of B_0 which is identical to that obtained with a steady-state treatment, as expected. The remaining terms are the corrections obtained with the time-dependent perturbation treatment. The main point of this approximate, however, is that the Kerr coefficient, under the condition given, can be expressed as a product of a Gaussian of width of the short pulse and a time-dependent polynomial.

Finally, let us examine the phase matching for the generated wave and the EIT-assisted enhancement to the Kerr nonlinearity. Indeed, the conventional phase matching for wave mixing is ensured in the present treatment. To see this, we start with the indices for the source and wave mixing fields, i.e.,

$$n_S = 1 - \frac{2\pi N\rho_{11}|D_{13}|^2}{\delta_3\hbar} \quad \text{and} \quad n_M = 1 - \frac{2\pi N\rho_{00}|D_{03}|^2}{\delta_3\hbar}.$$

For parallel laser beams we have, for the conservation of momentum and energy, $k_M = k_{L1} - k_{L2} + k_S$ and $\omega_M = \omega_{L1} - \omega_{L2} + \omega_S$. By combining these relations, one immediately obtains $\kappa_{03}\rho_{00} = \kappa_{13}\rho_{11}$. From Eq. (12a) we see that this is precisely the condition that maximizes the amplitude of the generated field. Therefore, for a phase-matched operation we have

$$C_0 = -\frac{1}{\delta_3\tau_S}\left[\left(\frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}} - 1\right)e^{iK_0\xi} + \left(1 + \frac{\rho_{00}^{(0)}\Omega_{21}}{\rho_{01}^{(0)}\Omega_{20}}\right)\right],$$

where $K_0 \approx -\kappa_{03}\rho_{00}^{(0)}/(\delta_3 + i\Gamma_3/2)$. In general, there is a second enhancement factor due to the exponential term in Eq. (11a). The optimum output for the generated wave will be the combination of the conventional phase matching and the constructive interference achieved with the exponential term in Eq. (11a).

The EIT-assisted enhancement can be seen from the expression of C_0 and $X(\eta)$. It is easily seen that for the EIT-assisted four-level system we have $\mathrm{Re}[\chi_{\mathrm{EIT}}^{(3)}(\omega_{L1})] \propto 1/|\Omega|^2\delta_3$ and $\mathrm{Im}[\chi_{\mathrm{EIT}}^{(3)}(\omega_{L1})] \propto \Gamma_3/|\Omega|^2\delta_3^2$. As has been pointed out before [3], compared with the conventional three-level Λ-scheme [Fig. 1(c)] for cross-phase modulation where the Kerr nonlinearity and two-photon limited absorption are given by

$$\mathrm{Re}[\chi_{3\text{-level}}^{(3)}(\omega_{L1})] \propto \frac{|\Omega_{20}|^2|\Omega_S|^2}{\Delta_2^2\delta_3}$$

and

$$\mathrm{Im}[\chi_{3\text{-level}}^{(3)}(\omega_{L1})] \propto \frac{|\Omega_{20}|^2}{\Delta_2^2},$$

a large phase shift per unit absorption length can be achieved with the EIT scheme at a low intensity level. We emphasize that the improvement is due to the significant difference between the large one-photon detuning in the case of a conventional three-level Λ scheme and the one-photon coupling Rabi frequency $|\Omega_{21}|$ in the EIT-assisted case, and the corresponding difference in the absorption coefficients. In the EIT-assisted scheme negligible probe laser absorption is ensured whereas in the conventional three-level Λ scheme, the absorption of the probe laser always poses a significant limitation.

V. SUMMARY

In summary, we have presented a time-dependent perturbation treatment on EIT-enhanced Kerr nonlinearity where the wave-mixing field is also taken into consideration. The inclusion of the mixing field opens possibilities for the suppression and enhancement of the Kerr nonlinearity under suitable conditions, therefore adding rich dynamics to the system. Furthermore, we have shown that the EIT-enhanced scheme is superior in comparison to the conventional three-level scheme for cross-phase modulation. The time-dependent treatment allows one to examine the dynamic response of the system to the source laser, particularly the suppression and enhancement of Kerr effect due to the internally generated field. The latter may find applications in processes such as coherent control.

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[5] In principle, by neglecting dephasing effects any pulse combinations such that $\tau_S/\tau_L \ll 1$ may be treated according to the method presented here.

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[7] Note that, in general, the pulse duration for the wave-mixing field is different from that of the source field since the pulse bandwidth narrowing may happen. However, we assume that such pulse stretching in the time domain is still shorter in comparison to that of the probe and coupling laser. This is a valid assumption because of the criteria Eqs. (5) and (6).

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