Gain-Assisted Large and Rapidly Responding Kerr Effect using a Room-Temperature Active Raman Gain Medium

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A four-level N scheme with a two-mode active Raman gain core is investigated for large and rapidly responding Kerr effect enhancement at room temperature. The new scheme is fundamentally different from the electromagnetically induced transparency (EIT-)based ultraslow-wave Kerr effect enhancement scheme. It eliminates the requirement of group velocity matching and multispecies medium. It also eliminates significant probe field attenuation or distortion associated with weakly driven EIT-based schemes. We show that a probe field can acquire a large, frequency tunable, gain-assisted nonlinear phase shift and yet travel with gain-assisted *superluminal* propagation velocity. This raises the possibility of rapidly responding, frequency tunable nonlinear phase switching and phase gates for information science.

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Control of the slowly varying phase of a classical or quantum electromagnetic field has great technological importance in the field of information science. In a solid state medium such as an optical fiber, a propagation-dependent phase shift of an optical field can be produced using the Kerr effect [1] because very tight spatial confinement of the light mode results in high optical power in the fiber core. In glass-based optical fiber, however, the required low absorption characteristics necessarily implies nonlinear effects are very weak, requiring a very long propagation distance for the phase shift to accumulate. In addition, the lack of distinctive energy levels and transition selection rules also make the active control of nonlinear phase shifts difficult. In the field of information science, however, weak field, frequency tunability, active phase shift control, and compact size are important requirements that obviously cannot be met by glass-based or even highly doped nonlinear optical fibers. In gaseous phase media, a four-level N scheme has been proposed [2] to enhance nonlinear optical effects such as cross-phase modulation. The central point of this scheme, which is based on the principle of weakly driven electromagnetically induced transparency (EIT) [3], was to enhance the Kerr effect by significantly reducing the propagation velocity of a probe field. This N scheme and its variations have since been used in theoretical studies of quantum entanglement of ultraslow photons [4], single photon switching [5], nonlinear phase gates [6], and single photon propagation controls [7].

The weakly driven EIT-based nonlinear phase shift scheme [Fig. 1(a)], however, has significant problems. The two primary drawbacks of the scheme are the significant probe field attenuation and spreading at room temperature and the very long response time. Both of these difficulties arise from the very same key feature of the weakly driven EIT required for the significant enhancement of the Kerr nonlinearity, namely, the ultraslow propagation velocity. This is because the ultraslow propagation not only enhances the real part of the nonlinear index, but also significantly boosts the third-order attenuation. In addition, ultraslow propagation of the probe field necessarily implies slow system response time [8]. Currently, no significant nonlinear phase shift, pulsed or otherwise, has been demonstrated using weakly driven EIT-based ultraslow-wave schemes in room-temperature alkali atomic vapors.

In this Letter we propose a new scheme using warm atomic vapor that eliminates significant probe-field attenuation and distortion, and slow process response time using warm atomic vapors. The new scheme is based on gain-assisted or active Raman gain (ARG) configurations recently demonstrated [9–12] to be able to produce both superluminal and ultraslow propagations of optical waves. Contrary to all EIT-based schemes where the probe field operates in an absorption mode, the central idea of the ARG core-based scheme is that the probe field operates in a stimulated Raman emission mode. It is precisely this emission mode that leads to novel propagation characteristics. Indeed, we show that a probe field can acquire a large nonlinear phase shift, suffer no attenuation or distortion, yet travel with a superluminal group velocity. Furthermore, because of the large one-photon detuning

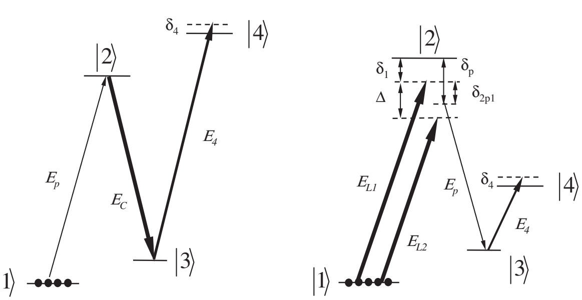

FIG. 1. Weakly driven EIT-based (a) and gain-assisted ARG core-based (b) schemes for enhancement of Kerr nonlinear phase shift.

the new scheme acquires important tunability for the probe field. These features are obviously unattainable with a weakly driven EIT scheme that relies on ultraslow propagation, where significant field attenuation and distortion, lack of tunability, and slow process response time are inherently unavoidable.

We consider an ARG core-based N scheme with two active Raman resonances [Fig. 1(b)], where a two-mode strong cw pump laser field $E_{L1(L2)}$ couples the ground state $|1\rangle$ to an excited state $|2\rangle$ with large one-photon detunings δ_1 and $\delta_1 + \Delta$ (with $|\Delta| \ll |\delta_1|$), respectively. A weak, pulsed (pulse length τ) probe field E_p couples state $|2\rangle$ to a lower excited state $|3\rangle$ with a large one-photon detuning $\delta_p \approx \delta_1$. Thus, the two Raman resonances have two-photon detunings $\delta_{2p1} = \delta_p - \delta_1$ and $\delta_{2p2} = \delta_p - (\delta_1 + \Delta) = \delta_{2p1} - \Delta$, respectively, for the two-photon transition $|1\rangle - |3\rangle$. A cw nonlinear phase-inducing laser field E_4 couples $|3\rangle - |4\rangle$ transition with a detuning δ_4. However, the $|4\rangle - |1\rangle$ transition back to the ground state is dipole forbidden, by choice.

We first work out the atomic response in the absence of the nonlinear phase control field E_4. The solution of this three-state two-mode ARG-core system is then used as the zeroth order solution to seek the nonlinear perturbation corrections to the probe field due to the presence of the nonlinear phase control field E_4 [13]. In the interaction picture the equations of motion describing the wave function amplitudes of this two-mode ARG scheme are given by

$$\dot{a}_2(z,t) = i\Omega_{21}^{(1)} e^{-i\delta_1 t} a_1 + i\Omega_{21}^{(2)} e^{-i(\delta_1+\Delta)t} a_1 + i\Omega_{23} e^{-i(\delta_1+\delta_{2p1})t} a_3 - \frac{\gamma_2}{2} a_2, \tag{1a}$$

$$\dot{a}_3(z,t) = i\Omega_{32} e^{i(\delta_1+\delta_{2p1})t} a_2 - \frac{\gamma_3}{2} a_3. \tag{1b}$$

Here, $\Omega_{21}^{(n)} = D_{21}E_{Ln}/(2\hbar)$ $(n = 1, 2)$ and $\Omega_{23} = D_{23}E_p/(2\hbar)$ are half-Rabi frequencies for $|1\rangle - |2\rangle$ and $|3\rangle - |2\rangle$ transitions (with dipole moments D_{21} and D_{23}, respectively) under excitations by laser fields E_{Ln} and E_p, and γ_j is the decay rate of state $|j\rangle$. We note that since $|\delta_1\tau| \gg 1$ and the probe laser is weak, the ground state population is not depleted ($a_1 \approx 1$). We also assume that δ_1 is much large than any Rabi frequencies, Doppler broadened linewidths, and frequency shifts induced by laser fields.

In order to predict the propagation of the probe field, Eqs. (1a) and (1b) must be solved simultaneously with the Maxwell equation for the probe field. Within the plane wave, slowly varying amplitude, and phase approximation, the Maxwell equation reads

$$\frac{\partial E_p^{(0)}}{\partial z} + \frac{1}{c}\frac{\partial E_p^{(0)}}{\partial t} = -i\kappa_{23} c_0 [a_3 a_2^* e^{-i(\delta_1+\delta_{2p1})t}]_{\mathrm{LFP}}, \tag{2}$$

where $E_p^{(0)} = E_p^{(0)}(z,t)$ is the negative frequency part of the probe field amplitude, $c_0 = 2\hbar/D_{32}$, and $\kappa_{32} = 2\pi\omega_p N|D_{32}|^2/(\hbar c)$, where N is the concentration and ω_p is the probe frequency. LFP, referring to low frequency part, indicates that only nonoscillating terms are kept.

Using the differential Fourier transform technique shown in Ref. [11] one can show that in the Fourier space Eq. (2) is given by

$$\frac{\partial \epsilon_p}{\partial z} - i\frac{\omega}{c}\epsilon_p = -i\kappa_{23}\epsilon_p W(\omega), \tag{3}$$

where $\epsilon_p = \epsilon_p(z,\omega)$ is the Fourier transform of $E_p^{(0)}(z,t)$, ω is the Fourier transform variable, and

$$W(\omega) = \frac{G_1}{\omega + \delta_{2p1} - i\gamma_3/2} + \frac{G_2}{\omega + \delta_{2p1} - \Delta - i\gamma_3/2}, \tag{4}$$

where $G_1 = |\Omega_{21}^{(1)}|^2/\delta_1^2$ and $G_2 = |\Omega_{21}^{(2)}|^2/(\delta_1 + \Delta)^2$. Equation (4) shows that there are two Raman resonances due to the two-mode pump field. One immediately recognizes that if $\delta_{2p1} = \Delta/2$ and the intensities of the two pump modes are adjusted so that $G_1 = G_2 = G$, then $\mathrm{Re}[W(\omega = 0)] = 0$. This choice of two-mode pump intensities and two-photon detunings removes a gain-dependent static phase [14]. Clearly, if $|\Delta\tau| \gg 1$ [15], Eq. (4) can be expanded in a rapidly converging McLaurin series with expansion parameter $1/|\Delta\tau| \ll 1$. Consequently, it is sufficiently accurate to keep only the first few terms in the series. Taking $W(\omega) = W^{(0)}(0) + W^{(1)}(0)\omega + O(\omega^2)$ and assuming a Gaussian input probe field $E_p^{(0)}(0,t) = E_p^{(0)}(0,0)e^{-t^2/\tau^2}$ so that $\epsilon_p(0,\omega) = (\tau/\sqrt{2})E_p^{(0)}(0,0)\times e^{-\omega^2\tau^2/4}$, one can solve Eq. (3) analytically. After inverse transform [16], we obtain

$$E_p^{(0)}(z,t) = E_p^{(0)}(0,0)\exp\left[G^{(1)}z - \left(\frac{t}{\tau} - \frac{z}{V_g\tau}\right)^2\right], \tag{5}$$

where $G^{(1)} = \kappa_{23}G\gamma_3/\Delta^2$ and group velocity is given by

$$V_g = \frac{c}{1 - c\kappa_{32}(8G/\Delta^2)} \approx -\frac{\Delta^2}{\kappa_{32}8G} \quad \left(\text{for } c\kappa_{32}\frac{8G}{\Delta^2} \gg 1\right). \tag{6}$$

Equations (5) and (6) clearly show that the probe field travels with a gain-assisted *superluminal* group velocity but without pulse attenuation, distortion, and gain-dependent phase shift [10,12,17].

Substituting Eq. (5) into Eq. (1b), using the adiabatic solution of Eq. (1a), i.e.,

$$a_2 \approx -\frac{\Omega_{21}^{(1)} e^{-i\delta_1 t}}{\delta_1 + i\gamma_2/2} - \frac{\Omega_{21}^{(2)} e^{-i(\delta_1+\Delta)t}}{\delta_1 + \Delta + i\gamma_2/2},$$

and integrating Eq. (1b) over time, we obtain a_3 in the absence of the nonlinear phase control field E_4,

$$a_3 \approx -i\frac{2\sqrt{G}}{c_0}\int_0^t dt' \cos\left(\frac{\Delta t'}{2}\right) E_p^{(0)}\left(z, t'\right). \tag{7}$$

When the field E_4 is present, a driving term $i\Omega_{34}e^{i\delta_4 t}a_4$ is added to the right-hand side of Eq. (1b), and the equation of motion for the amplitude a_4 and its correction to the amplitude a_3 must be considered. It is this correction that leads to the Kerr nonlinear phase shift of the probe field. The equation of motion for a_4 is given by

$$\dot{a}_4 = i\Omega_{43}e^{-i\delta_4 t}a_3 - \frac{\gamma_4}{2}a_4, \tag{8}$$

where $\Omega_{43} = D_{43}E_4/(2\hbar)$. Typically $|\delta_4| \gg |\Omega_{43}|$ and $|\delta_4\tau| \gg 1$; thus, we obtain an adiabatic following solution $a_4 \approx -\Omega_{43}e^{-i\delta_4 t}a_3/(\delta_4 + i\gamma_4/2)$ where a_3 is given by Eq. (7). The correction to a_3, referred to as A_3, can be expressed as

$$A_3 = -i\frac{|\Omega_{43}|^2}{\delta_4 + i\gamma_4/2}\int_0^t dt' a_3(t'). \tag{9}$$

The nonlinear polarization at the probe field frequency can now be constructed to give [18]

$$P_{\mathrm{NL}}^{(0)}(\omega_p) = [ND_{23}A_3a_2^* e^{-i(\delta_1+\delta_{2p1})t}]_{\mathrm{LFP}}. \tag{10}$$

Using the differential Fourier transform technique described in Ref. [11], we can show that the LFP contribution to the nonlinear polarization is given by

$$\begin{aligned}\text{nonlinear term} &= -i\frac{4\pi\omega_p}{c}P_{\mathrm{NL}}^{(0)}(\omega_p)\\ &= (i\Phi_{\mathrm{XPM}} + G^{(3)})E_p^{(0)},\end{aligned} \tag{11}$$

where we have defined the nonlinear phase shift (by cross-phase modulation) per unit length and the third-order nonlinear gain coefficient at the probe field frequency as

$$\Phi_{\mathrm{XPM}}(\omega_p) = \left(-\kappa_{23}\frac{8G}{\Delta^2}\right)\frac{|\Omega_{43}|^2\delta_4}{\delta_4^2 + \gamma_4^2/4}, \tag{12a}$$

$$G^{(3)}(\omega_p) = \left(\kappa_{23}\frac{8G}{\Delta^2}\right)\frac{|\Omega_{43}|^2\gamma_4/2}{\delta_4^2 + \gamma_4^2/4} > 0. \tag{12b}$$

We note that the quantity in the parenthesis in Eq. (12a) is exactly the inverse of the superluminal group velocity shown in Eq. (6). We further note that Eq. (12b) demonstrates a third-order gain, as expected, from a simulated emission process [19].

It is now appropriate to comment on the procedures that lead to analytical results Eqs. (12a) and (12b). In deriving nonlinear atomic responses using the differential Fourier transform method, one encounters a common denominator $f(\omega) = -\omega^2\tau^2 \pm \omega\tau\Delta\tau - \Delta^2\tau^2/4$ in all terms as the result of Fourier transform of the double integral in A_3. This is also the case of all EIT-based schemes if applied to short-pulsed operation. In an ARG core-based system a two-photon detuning such that $|\Delta\tau| \gg 1$, which is exactly the condition for rapid converging power series expansion of Eq. (4) and probe field superluminal propagation, also makes the ω dependence of $f(\omega)$ very weak [15]. This condition results in, for a Gaussian pulse envelope, an analytical solution with very good accuracy. Without this condition it is, in general, incorrect to make a steady-state approximation in a short-pulsed operation. This conclusion applies equally to all weakly driven EIT-based schemes where $|\Omega_C\tau| \approx 1$ [20].

We now consider the case of room-temperature atomic vapor. In a warm vapor large Doppler broadened linewidths degrade the effectiveness of EIT-based schemes. It is, however, much less important in an ARG-based scheme because of the large one-photon detunings. In Fig. 2 we compare the performance of EIT-based [Fig. 1(a)] [21] and ARG-based [Fig. 1(b)] schemes using experimentally realistic parameters.

In the EIT-based scheme both probe and control fields are on resonance. For $\tau = 10\ \mu$s, we take $\kappa_{12} \approx 10^{11}/(\mathrm{cm\ s})$, $\gamma_3 \approx 300$ Hz, $\gamma_2 \approx 500$ MHz, $\gamma_4 \approx 500$ kHz [21,22], $|\delta_4| = 10\gamma_4$, and $|\Omega_{43}| = 700$ kHz. We obtain the third order probe field attenuation coefficient $\alpha^{(3)} \approx 0.1\ \mathrm{cm}^{-1}$ (thin dashed line) for $\Phi_{\mathrm{XPM}}^{(\mathrm{EIT})} \approx 1$ rad (thick dashed line) [Fig. 2(a)].

In the ARG-based scheme, we take $\kappa_{32} \approx 10^{11}/(\mathrm{cm\ s})$, $\delta_1 = 1.9994$ GHz, $\delta_{2p1} = \Delta/2 = 600$ kHz, and $\Omega_{21}^{(1)} = \Omega_{21}^{(2)} = \Omega_{21}$. Other parameters are the same as before. With these parameters, $|\Delta\tau| = 12 \gg 1$, $|\delta_4\tau| = 50 \gg 1$, and all conditions required are satisfied and our analytical results are valid. We have probe field gain $G^{(3)} \approx 0.1\ \mathrm{cm}^{-1}$ (thin solid line) for $\Phi_{\mathrm{XPM}}^{(\mathrm{ARG})} \approx 1$ rad (thick solid line) [Fig. 2(a)].

In Fig. 2(b) we plot the probe field nonlinear phase shift, absorption (EIT), and gain (ARG) coefficients as functions of nonlinear phase shift field Rabi frequency $|\Omega_{43}|$. Here, we have chosen $|\Omega_{21}^{(1)}| = |\Omega_{21}^{(2)}| = 10$ MHz for the ARG scheme. For the EIT scheme we take $|\Omega_C| = 100$ MHz in order to limit the attenuation $\alpha^{(3)} \approx 0.1\ \mathrm{cm}^{-1}$. All other parameters are as in Fig. 2(a). Clearly, the ARG-based

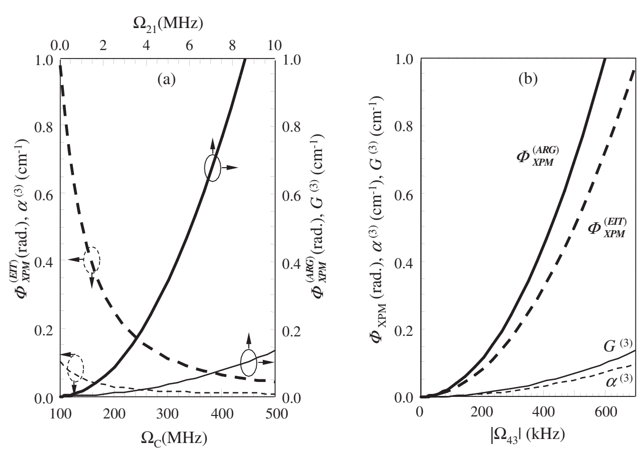

FIG. 2. (a) Probe field nonlinear phase shifts, attenuation coefficient $\alpha^{(3)}$ (thick and thin dashed lines, for the EIT scheme), and gain coefficient $G^{(3)}$ (thick and thin solid lines for the ARG scheme). (b) Probe field nonlinear phase shifts, attenuation (EIT scheme), and gain (ARG scheme) coefficients as functions of $|\Omega_{43}|$.

scheme is superior to the EIT-based scheme. We note that it is always possible to choose parameters such that $|V_g^{(\mathrm{ARG})}| = |V_g^{(\mathrm{EIT})}|$. This leads to $|\Phi_{\mathrm{XPM}}^{(\mathrm{ARG})}| = |\Phi_{\mathrm{XPM}}^{(\mathrm{EIT})}|$ and $|G^{(3)}| = |\alpha^{(3)}|$. However, the ARG scheme has the advantages of no loss, no distortion, and fast response time, features that are unmatchable by weakly driven EIT-based nonlinear phase shift schemes.

In summary, we have presented a novel scheme to achieve a large and rapidly responding nonlinear phase shift to a probe field. Contrary to a weakly driven EIT-based scheme, which is absorptive, the new scheme is based on the key idea of the probe field being operated in a stimulated Raman emission mode. It is precisely this emission process that leads to new features such as large nonlinear phase shifts with nondistorted gain and superluminal propagation velocities. We note that this latter feature is entirely consistent with the gain-assisted propagation and is in agreement with the theoretical and experimental studies reported in Refs. [10–12]. This feature leads to the possibility of fast nonlinear phase switching and nonlinear phase gates for information science. We further note that since it is not based on ultraslow propagation, an ARG-based scheme eliminates the necessity of a two-species medium and the requirement of group velocity matching between an ultraslow probe field and a nonlinear phase control field [4]. In addition, the far-off resonance operation mode brings about important tunability. These are significant and key features that may have important technological implications. Using experimentally realistic parameters we have shown that ARG-based schemes are superior to weakly driven EIT-based nonlinear phase shift schemes in room-temperature vapors. We finally note that the theory can be easily generalized to describe a quantum field at a single photon level. In theory an ARG scheme permits the possibility of single photon nonlinear phase shift and phase gates without requiring ultraslow group velocity matching and multispecies medium. Full quantum mechanical generalizations and experimental demonstration of the scheme proposed are under way.

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[13] We have neglected very weak spontaneous Raman emission near the frequency of the probe field.
[14] It eliminates a static phase shift that is independent of the nonlinear phase control field E_4. In principle, one can also use a single mode pump field, where the similar gain-assisted nonlinear phase shift occurs in the presence of a static (Ω_{43} independent) linear phase shift.
[15] In EIT-based schemes a two-photon detuning such as $|\Delta\tau| > 1$, however, results in severe penalties.
[16] For a Gaussian pulse the dominant contributions come from when $|\omega\tau| < 4$. Thus, for $|\delta_{2p}\tau| > 5$, the $\omega\tau$ in the denominator of the integrand can be neglected.
[17] We have assumed $|\Delta| \gg \gamma_3$; therefore, $W^{(0)}(0) \simeq 0$ and $W^{(1)}(0) \simeq 8G/\Delta^2$. Small residual due to γ_3 leads to amplification of the probe field but not to phase shift.
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[21] We state that results of the weakly driven EIT-based scheme shown here for comparison are obtained using a cold vapor formula with known room-temperature relaxation constants for γ_j ($j = 2, 3, 4$) (see also Ref. [22]).
[22] To avoid larger Doppler broadening of high-lying levels at room temperature, state $|4\rangle$ (regardless of EIT-based or ARG-based schemes) should be a member of the ground state manifold, and E_4 is therefore a circularly polarized microwave field. Our parameters are directly relevant to a ^{87}Rb system where $|1\rangle = |5S_{1/2}, F = 1, m_F = -1\rangle$, $|2\rangle = |5P_{1/2}, F' = 1, m_{F'} = 0\rangle$, $|3\rangle = |5S_{1/2}, F = 1, m_F = +1\rangle$, and $|4\rangle = |5S_{1/2}, F = 2, m_F = +2\rangle$ with a magnetic field of a few Gauss and a microwave field frequency of 6.8 GHz.