

Large Cross-Phase Modulation between Slow Copropagating Weak Pulses in ^{87}Rb

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We propose a scheme to generate double electromagnetically induced transparency and optimal cross-phase modulation for two slow, copropagating pulses with matched group velocities in a single species of atom, namely ^{87}Rb . A single pump laser is employed and a homogeneous magnetic field is utilized to avoid cancellation effects through the nonlinear Zeeman effect. We suggest a feasible preparational procedure for the atomic initial state to achieve matched group velocities for both signal fields.

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Introduction.—The optical Kerr effect, $n = n^{(0)} + n^{(2)}I$ (for n the total refractive index, $n^{(0)}$ the linear refractive index, I the field intensity, and $n^{(2)}$ the optical Kerr coefficient), is invaluable for spectral broadening and self-focusing of laser pulses [1]. Large nonlinear interactions have also been used to generate single photons [2], enhanced refractive index [3], self-phase modulation [4], and Stokes and anti-Stokes generation [5,6]. In cross-phase modulation (XPM), I is the intensity of the other field; thus XPM enables the phase of each field to be controlled by the strength of the other, which is critical for applications such as deterministic optical quantum computation [7] and all-optical switching [8]. Unfortunately, $n^{(2)}$ is extremely small but can be effectively increased via electromagnetically induced transparency (EIT) [9–11], which yields a large optical nonlinearity [12–15]. Whereas these experiments demonstrated the promise of a large nonlinear coupling for cw fields, applicability is severely limited for propagating fields due to the different group velocities of the two pulses, which significantly reduces the interaction time.

Double EIT (DEIT) generalizes EIT for simultaneous action on two separate fields, and, for slow group velocities, can effect a large XPM [16–19]; however, DEIT has not yet been successfully realized. In this Letter we devise a method for achieving large XPM for two slow copropagating pulses with matched group velocities in a single species of atom, namely, using the $D1$ line of ^{87}Rb , by avoiding cancellation of nonlinearities near resonance and decoherence due to coupling bright states, plus employing population transfer to achieve matched group velocities for the two pulses.

Scheme.—Our scheme employs five atomic levels as shown in Fig. 1(a) and combines the advantageous properties of the N -type scheme [13], the tripod scheme [19], and the M -type scheme [18]. A single pump field drives the $|3\rangle \leftrightarrow |4\rangle$ transition with Rabi frequency Ω_p . If state $|2\rangle$ is omitted our scheme reduces to the N -type scheme: the ac Stark shift created by off-resonant coupling to state $|5\rangle$ creates a giant nonlinearity between signal fields 1 and 2, but EIT is only realized for signal field 1. Consequently,

due to the mismatch in the group velocities, the interaction time is greatly reduced. On the other hand, if state $|5\rangle$ is omitted we have a tripod scheme in which DEIT can be achieved but for which the nonlinearity disappears at exact two-photon resonance (as it does in the M -type scheme). The slight detuning that is therefore necessary to create nonlinear effects [18,19] also generates linear absorption, and for a given desired absorption rate the generated nonlinearity is suppressed by one to two orders of magnitude.

We will show that the scheme of Fig. 1(a) avoids the problems associated with the schemes discussed above. In addition to the pump field, two signal fields with slowly varying field amplitude \mathcal{E}_i ($i = 1, 2$) resonantly couple the $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |4\rangle$ transitions with Rabi frequencies $\Omega_i \equiv -|\mathbf{d}_{4i}|\mathcal{E}_i/\hbar$, with $\mathbf{d}_{ij} = \langle i|\mathbf{d}|j\rangle$ the matrix elements of the dipole moment operator \mathbf{d} . In addition, field 2 also off resonantly couples the $|3\rangle \leftrightarrow |5\rangle$ with Rabi frequency $\Omega'_2 \equiv -|\mathbf{d}_{53}|\mathcal{E}_2/\hbar$. If the atoms are initially prepared in a mixture [20] of state $|1\rangle$ and $|2\rangle$ with density matrix $\rho_{\text{mix}} = p_1|1\rangle\langle 1| + p_2|2\rangle\langle 2|$, the two Λ subsystems $|1\rangle \leftrightarrow |4\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |4\rangle \leftrightarrow |3\rangle$ induce EIT for both fields 1 and 2, i.e., DEIT. The real part of the corresponding index of refraction n_i for signal field i then has the approximate form [10]

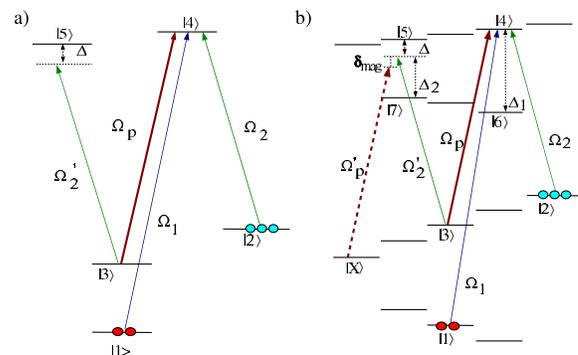


FIG. 1 (color online). Atomic level configuration for creating DEIT and large XPM between two weak signal fields 1 (Ω_1) and 2 (Ω_2, Ω'_2). (a) Simplified level configuration of the scheme. (b) Realization of the scheme using the $D1$ Line of ^{87}Rb .

$$n_i = 1 + \eta_i \delta_{pi}, \quad \eta_i \equiv \bar{\rho} p_i |\mathbf{d}_{4i}|^2 / (2\hbar \epsilon_0 |\Omega_p|^2) \quad (1)$$

for $i = 1, 2$ $\delta_{ij} \equiv \delta_j - \delta_i$, $\bar{\rho}$ the atomic number density, and p_i the initial population in state $|i\rangle$. For $E_{ij} \equiv E_j - E_i$, the detuning of the signal field frequency ω_i and the laser pump frequency ω_p are $\delta_i = \omega_i - E_{i4}/\hbar$ and $\delta_p = \omega_p - E_{34}/\hbar$, respectively. Because Eq. (1) varies strongly with the two-photon detuning δ_{pi} the group velocity is very small. Below we will show how the group velocities can be made equal by manipulating the populations p_i .

The off-resonant coupling for transition $|5\rangle \leftrightarrow |3\rangle$ caused by field 2 modifies the tripod scheme by producing an ac Stark shift for state $|3\rangle$ given by

$$\Delta E_3 = \hbar |\Omega'_2|^2 / \Delta, \quad \Delta = \omega_2 - E_{35}/\hbar \quad (2)$$

with Δ the (large) detuning of field 2 with respect to the $|3\rangle \leftrightarrow |5\rangle$ transition. This energy shift implies that the pump frequency ω_p is somewhat detuned from the $|3\rangle \leftrightarrow |4\rangle$ transition; hence the index of refraction for this EIT medium changes due to the dispersion relation of Eq. (1).

Replacing δ_{pi} by $\delta_{pi} - \Delta E_3/\hbar$ and inserting this into Eq. (1) yields

$$n_1 = 1 + \eta_1 (\delta_{p1} - \chi I_2), \quad \chi = |\mathbf{d}_{53}/\hbar|^2 / c \epsilon_0 \Delta \quad (3)$$

with intensity $I_i = |\mu_0^{-1} \mathbf{E}_i \times \mathbf{B}_i| = c \epsilon_0 |\mathcal{E}_i|^2$ corresponding to the modulus of the complex Poynting vector.

We find that this intuitively appealing phenomenological derivation agrees with a rigorous calculation based on third-order time-dependent perturbation theory in the weak signal fields. Neglecting all decoherence effects and assuming a resonant pump field ($\delta_p = 0$), the Schrödinger equation yields the index of refraction

$$\begin{aligned} n_1 &= 1 + \eta_1 (\delta_1 - \chi I_2), \\ n_2 &= 1 + \eta_2 (\delta_2 - \chi I_2) - \eta_1 \chi I_1 \end{aligned} \quad (4)$$

with the self-phase modulation (SPM) coefficient for field 2 given by $-\eta_2 \chi$ and cross-phase modulation (XPM) coefficient for both fields given by $-\eta_1 \chi$. Both XPM and SPM coefficients are given to first order in $1/\Delta$ and to second order in $1/|\Omega_p|^2$. A more detailed analysis (see below) shows that this simple result is qualitatively correct if the detuning Δ is much larger than the decay rate, and the ac Stark shift (2) is smaller than the width of the EIT transparency window. As this scheme shares the advantages and avoids the disadvantages of the N -type, tripod, and M -type schemes, it yields the optimal XPM based on EIT techniques.

Implementation with ^{87}Rb .—Our scheme can be realized in a gas with a single species of atoms; a specific implementation using the $D1$ line of ^{87}Rb is illustrated in Fig. 1(b). A homogeneous magnetic field parallel to the laser propagation minimizes coupling to states that are not part of the scheme in Fig. 1(a). It is of particular importance to break two-photon resonance for the Λ subsystem $|3\rangle \leftrightarrow |5\rangle \leftrightarrow |X\rangle$. If this is not the case a further EIT

transition that transfers atoms from state $|3\rangle$ to state $|X\rangle$; this would remove the crucial energy shift (2) and hence destroy XPM. The linear Zeeman effect is not suitable to break two-photon resonance. However, for large enough magnetic fields the Zeeman splitting depends nonlinearly on the magnetic quantum number without affecting the corresponding selection rule [21]. Numerical diagonalization of the Hamiltonian for a magnetic field of $B = 150$ G shows that the energy differences E_{32} and E_{X3} differ by an amount $\delta_{\text{mag}} = (E_{32} - E_{X3})/\hbar = -12.9$ MHz. This is much larger than the EIT transparency window [13] $\delta\tau = \min\{\Omega'_p, |\Omega'_p|^2/\gamma\}$ and breaks two-photon resonance for the $|3\rangle \leftrightarrow |5\rangle \leftrightarrow |X\rangle$ transition. We therefore can neglect state $|X\rangle$ in our considerations; this approximation is confirmed by our numerical simulations (see below).

We analyzed this specific scheme in three ways: the simple analytical theory (SAT) described above yields Eq. (4) for the XPM coefficient, a more elaborate analytical theory (EAT), and a numerical simulation (NUM). The elaborate theory EAT is based on third-order time-dependent perturbation theory to solve the Schrödinger equation with the evolution restricted to states $|1\rangle \leftrightarrow |7\rangle$ of Fig. 1(b). Spontaneous emission is included in the non-Hermitian atomic Hamiltonian

$$H = \hbar \begin{pmatrix} \delta_1 & 0 & 0 & \Omega_1^* & 0 & \Omega_1'^* & 0 \\ 0 & \delta_2 & 0 & \Omega_2^* & 0 & \Omega_2''^* & 0 \\ 0 & 0 & \delta_p & \Omega_p^* & \Omega_2'^* & \Omega_p'^* & \Omega_2'''^* \\ \Omega_1 & \Omega_2 & \Omega_p & -i\frac{\gamma}{2} & 0 & 0 & 0 \\ 0 & 0 & \Omega_2' & 0 & \tilde{\Delta} & 0 & 0 \\ \Omega_1' & \Omega_2'' & \Omega_p' & 0 & 0 & \tilde{\Delta}_1 & 0 \\ 0 & 0 & \Omega_2'' & 0 & 0 & 0 & \tilde{\Delta}_2 \end{pmatrix}, \quad (5)$$

with detunings [see Fig. 1(b)] $\Delta_1 = \omega_1 - E_{16}/\hbar$, $\Delta_2 = \omega_2 - E_{37}/\hbar$, and phenomenological complex quantities $\tilde{\Delta}_i = -\Delta_i - i\frac{\gamma}{2}$ [22]. We obtained an accurate analytical result, but the expression is unwieldy and not displayed here. Instead we compare it with the numerical simulation NUM in which we solved the time evolution of the atomic density matrix based on the full Lindblad master equation for all 16 states of Fig. 1(b), including spontaneous emission based on all Clebsch-Gordan coefficients and 6- j symbols as specified by Steck [23]. Below we will also briefly discuss numerical results that include dephasing and transit broadening which is important for hot atomic gases. XPM between the two signal beams is proportional to that part of the atomic mean dipole moment which couples to signal field i and vanishes if the intensity of the other signal field is zero. To describe this we define a dimensionless mean cross dipole moment by

$$d_{\text{XPM}} = \frac{1}{e a_0} (|\text{Tr}(\rho \mathbf{d})|_{\mathcal{E}_{3-i} \neq 0} - |\text{Tr}(\rho \mathbf{d})|_{\mathcal{E}_{3-i} = 0}), \quad (6)$$

with e the electron's charge and a_0 the Bohr radius. d_{XPM} is a direct measure of the mutual interaction between the

signal pulses; in SAT and EAT it is proportional to the XPM terms, in NUM it also contains higher order terms. The results of NUM for d_{XPM} are shown in Fig. 2 and show excellent agreement with the analytical results EAT and qualitative agreement with the simple theory SAT. We have used Steck's spectroscopic data [23] and the parameters given in the figure caption.

The XPM phase shift can be identified as the difference between the phase factors acquired by signal field i if the second signal field is switched on and off, respectively, after it has been propagating for $L = 1.6$ mm (double the Rayleigh length). This definition amounts to

$$\phi_i^{\text{XPM}} = \frac{\omega_i}{c} L (n_i(\mathcal{E}_{3-i} \neq 0) - n_i(\mathcal{E}_{3-i} = 0)), \quad (7)$$

for $i = 1, 2$ and with the refractive index evaluated using the result of EAT for the steady state value of the atomic dipole moment. Figure 2 indicates that steady state is already achieved after $0.7 \mu\text{s}$. The stationary values are given by $\phi_1^{\text{XPM}} = 1.11$, corresponding to an effective XPM coefficient $(-\eta_1 \chi)_1^{\text{eff}} = 0.27 \text{ cm}^2/\text{W}$, and $\phi_2^{\text{XPM}} = 0.84$, corresponding to an effective XPM coefficient $(-\eta_1 \chi)_2^{\text{eff}} = 0.20 \text{ cm}^2/\text{W}$. The difference of XPM for field 1 and 2 is due to decoherence effects and coupling to off-resonant states. The pulses are attenuated by about 20% due to off-resonant coupling to states $|6\rangle$ and $|7\rangle$. In addition, the nonzero detuning associated with the energy shift (2) leads to an intensity-dependent attenuation of about 3%.

In summary, the three theoretical results SAT, EAT, and NUM indicate that the five-level scheme of Fig. 1(b) would work well even if all 16 hyperfine states are included. The off-resonant coupling to the additional states only results in

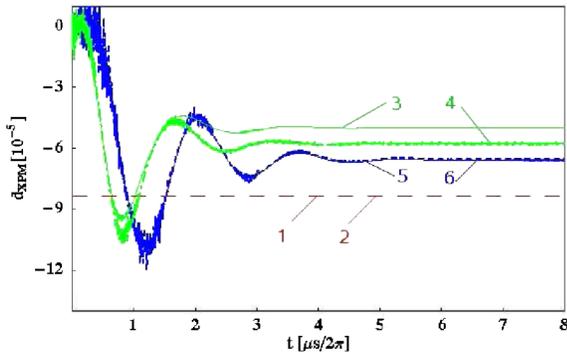


FIG. 2 (color online). Mean cross dipole moment d_{XPM} of Eq. (6) as a function of time for signal field 1 and 2. Lines 1, 2 represent the result of SAT [Eq. (4)], which is identical for both signal fields. Lines 3, 4 (5, 6) represent the results of EAT and NUM for signal field 1 (2), respectively. The parameters used for these solutions are $\Omega_1 = 0.68$ MHz, $\Omega_2 = -0.55$ MHz, $\Omega_p = 4.06$ MHz, $\Delta = -134.58$ MHz, $\Delta_1 = 894.93$ MHz, $\Delta_2 = 621.85$ MHz, $\delta_{\text{mag}} = -12.93$ MHz, and $\gamma = 5.73$ MHz, $\bar{\rho} = 10^{14} \text{ cm}^{-3}$, and $B = 150$ G, with initial atomic state $\rho_{\text{mix}} = 0.4|1\rangle\langle 1| + 0.6|2\rangle\langle 2|$.

the small, very rapidly oscillating perturbations displayed in Fig. 2 as an increased width of lines 4 and 6.

Preparation of the initial state.—For pulsed signal fields maximum interaction time is achieved for DEIT with equal group velocities. From Eq. (4) one can infer that the group velocity v_i of signal field i is proportional to $v_i \propto p_i |\mathbf{d}_{4i}|^2$. It is therefore possible to achieve $v_1 = v_2$ by preparing suitable populations p_i of the initial density matrix $\rho_{\text{mix}} = p_1|1\rangle\langle 1| + p_2|2\rangle\langle 2|$. We propose the following procedure. We first pump all atoms to state $|4\rangle$ whence they decay to states $|1\rangle$ and $|2\rangle$ plus other states. To maximize the EIT effect one may repump atoms in all states but $|1\rangle$ and $|2\rangle$ into state $|4\rangle$ until the atomic state is well approximated by ρ_{mix} with $p_i = \gamma_{4i}/(\gamma_{41} + \gamma_{42}) \propto |\mathbf{d}_{4i}|^2$, where γ_{4i} is the decay rate from state $|4\rangle$ to state $|i\rangle$. With these populations the group velocities would be proportional to $v_i \propto |\mathbf{d}_{4i}|^4$ and thus differ significantly. We therefore induce as a last step in the atomic state preparation a Raman transition between $|1\rangle$ and $|2\rangle$. Ideally this exchanges the populations, $p_1 \leftrightarrow p_2$, resulting in $p_i = |\mathbf{d}_{4,3-i}|^2 / (|\mathbf{d}_{41}|^2 + |\mathbf{d}_{42}|^2)$ so that both group velocities are now proportional to $|\mathbf{d}_{41}|^2 |\mathbf{d}_{42}|^2$ and therefore equal. We remark that the Raman transition is not required to work perfectly. An exchange of only 90% of the populations would lead to a difference of 10% in the group velocities, for instance.

Maximum phase shift for pulses at single-photon level.—One prominent application of XPM would be the creation of a controlled phase gate for photonic qubits [24]. We therefore estimate the maximal XPM phase shift achievable for two Gaussian pulses at single-photon level propagating through DEIT media. To do so we generalize the expression of XPM Kerr coefficient (4) by replacing η_1 by the standard EIT expression that includes spontaneous emission [9,10] for Λ atoms in a resonant pump beam, $\tilde{\eta}_1 = \eta_1 |\Omega_p|^2 / (|\Omega_p|^2 - \delta_1(\delta_1 + i\gamma/2))$. To first order in the detuning δ_1 we have $\tilde{\eta}_1 \approx \eta_1 + i\tau\delta_1$ with $\tau \equiv \eta_1 \gamma / (2|\Omega_p|^2)$. Within the paraxial approximation, the slowly varying electric field amplitude of a Gaussian pulse of duration T and minimum ($1/e$ intensity) waist w_0 at single-photon level in a medium that is described by $\tilde{\eta}_1$ propagates according to

$$\mathcal{E} = E_{\text{max}} \frac{w_0^2}{\Theta} \frac{T}{\Xi} \exp\left\{-\frac{x^2 + y^2}{2\Theta} - \frac{(\frac{z}{v_{\text{gr}}} - t)^2}{\Xi^2}\right\} \quad (8)$$

$$E_{\text{max}} = \sqrt{\frac{\hbar\omega}{\sqrt{2\pi^3} \epsilon_0 c T w_0^2}} \quad (9)$$

with $\Theta \equiv w_0^2 + i\frac{z}{k}$ and $\Xi \equiv \sqrt{T^2 + 4kz\tau}$. As for any Gaussian beam the field intensity is only close to its maximal value over a propagation length $L = 2z_R$, with $z_R = kw_0^2$ the Rayleigh length. In addition, the EIT transparency window attenuates the pulse through the factor Ξ . Requiring that Ξ changes little over z_R implies a minimum pulse duration of $T = 2\sqrt{\tau k z_R}$. Using these values we can

estimate the maximum phase shift ϕ^{\max} as the product of L and the XPM induced change in the wave vector $k = n\omega/c$,

$$\begin{aligned}\phi^{\max} &= L \frac{\omega}{c} \chi^{\text{XPM}} c \varepsilon_0 E_{\max}^2 = -\frac{3\sqrt{6}}{4\pi} \frac{\lambda}{w_0} \frac{\gamma}{\Delta} \sqrt{\rho k^{-3}} \\ &\approx 0.58 \frac{\lambda}{w_0} \frac{\gamma}{\Delta} \sqrt{\rho k^{-3}}.\end{aligned}\quad (10)$$

To avoid decoherence we need $\gamma/\Delta \ll 1$. The diffraction limit [17] implies $\lambda/w_0 < 1$ and we estimate that resonant dipole-dipole interaction would modify ϕ^{\max} for $\bar{\rho}k^{-3} \simeq 1$. Thus, the maximum XPM phase shift between two photons would in this approach be of the order of 0.1 rad. This is considerably smaller than previous estimates of more than 1 rad [14,16] but may be overcome by combining our scheme with the ideas presented in Ref. [25]. The difference arises from imposing different constraints (dipole-dipole interaction) and our neglect of ground-state decoherence, which is reasonable for trapped ultracold atoms.

Limitations.—The numerical example presented above is based on the parameters for a dense ultracold gas such as a trapped elongated Bose-Einstein condensate. A mutual phase shift of the order of π could then be obtained for pulses containing hundreds of photons. In a hot atomic gas (e.g., $T = 440$ K and $\bar{\rho} \sim 10^{14}$ cm $^{-3}$), the efficiency of this scheme is mainly limited by the atomic motion. Although the Doppler effect is canceled in the two-photon detuning $\delta_i - \delta_p$ for copropagating light fields, the atomic motion narrows the transparency window according to $\delta_\tau \leq \Omega_p^2/\Delta_D$ [5,26], where $\Delta_D \approx 500$ MHz is the mean Doppler detuning. The XPM phase shift would then be 2 orders of magnitude smaller than the optimal value. Transit broadening and ground-state dephasing [27] due to the atomic motion in and out of the laser fields and collisions between atoms are also detrimental. For laser pulses with a width of 2 mm in a hot atom setup using buffer gas [6], rates less than 1 kHz are possible. Numerical simulations indicate that our scheme would produce 0.4 rad XPM (with 40% attenuation) for two weak pulses with 10^2 photons, a transit broadening rate of 1 kHz, and a ground-state dephasing rate of 1 kHz.

Conclusion.—Cross-phase modulation between weak signal fields is one of the most significant challenges for nonlinear optical switching and quantum information. We have proposed an approach to achieve strong cross-phase modulation under realistic conditions for ^{87}Rb in which matched group velocities for two interacting pulses can be realized. Our approach incorporates all of the following desirable conditions: copropagating laser beams to avoid Doppler shifts, a single atomic species, and only a single pump field. We show that our scheme yields optimal XPM for pump schemes based on DEIT, and we estimate the maximum phase shift to be 0.1 rad for Gaussian pulses at single-photon level. To avoid decoherence effects Bose-

condensed gases would be preferred, but we expect the scheme also to be effective for thermal gases if sufficiently strong signal pulses are used.

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