Opening Optical Four-Wave Mixing Channels with Giant Enhancement Using Ultraslow Pump Waves

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We show that by strongly modifying the dispersion properties of a four-level system, nonexisting wave mixing channels can be opened and significantly enhanced. Specifically, we show that coherent optical four-wave mixing with a pump wave mediated by electromagnetically induced transparency (thereby propagating with an extremely slow group velocity) will lead to many orders of magnitude enhancement in the amplitude of the generated wave. Contrary to common belief, a large transparency window, which causes a large propagation velocity, actually diminishes efficient mixing wave production.

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Since the first observation by Franken *et al.* [1], optical wave mixing has developed into one of the centerpieces of modern technology. In these highly nonlinear processes, mixing-wave production efficiency has always been of primary importance. However, recent works [2] on ultraslow optical-wave propagation have opened new possibilities for improvement, and perhaps even new research directions, in the field of nonlinear optics. A great majority of these works have focused on group velocity reduction of an optical pulse in the context of a three-level system. However, we believe that more emphasis should be given to the concept of dispersion/index manipulation. Indeed, it is dispersion/index manipulation that is at the heart of the electromagnetically induced transparency (EIT) process that has been exclusively used in all optical group velocity reduction experiments reported to date. The concept of dispersion/index manipulation is much broader than just group velocity reduction. Our research indicates that in the field of nonlinear optics it will have many profound consequences.

In this Letter, we report the first theoretical investigation of optical coherent four-wave mixing (FWM) with a weak pump wave that travels with an extremely slow group velocity. Two key contributions are presented in this study. First, we show that the EIT [3] window created by a coupling laser drastically modifies the dispersion properties for a pump wave traveling in the medium, and opens a coherent FWM channel that would otherwise not be possible. Correspondingly, the on-resonance pump wave propagates in this medium with an extremely slow group velocity and minimum absorption, thereby allowing efficient, coherent FWM production. Without the EIT process the on-resonance pump wave would be completely absorbed, leading to an incoherent FWM output. Second, we show that contrary to common belief, a wider transparency window, resulting in less loss and higher propagation velocity for the pump wave, actually significantly diminishes the coherent FWM efficiency. Specifically, we show that with an ultraslow group velocity the FWM process acquires many orders-of-magnitude enhancement. To the best of our knowledge, the results presented here have never been reported in the literature and represent a new research direction that may have profound technological applications.

Consider a typical four-level system interacting with one pulsed and two cw laser fields (Fig. 1). The pulsed pump field E_p (frequency ω_p, pulse length τ, tuned to $|0\rangle \rightarrow |2\rangle$ resonance) serves as the first step of the three-photon excitation of state $|3\rangle$. A cw field E_2 (frequency ω_2) provides two photons to complete the three-photon excitation of state $|3\rangle$. A second cw field E_c (frequency ω_c, tuned to resonance) couples states $|2\rangle$ and $|1\rangle$ to provide dispersion manipulation of state $|2\rangle$. With these definitions, a set of

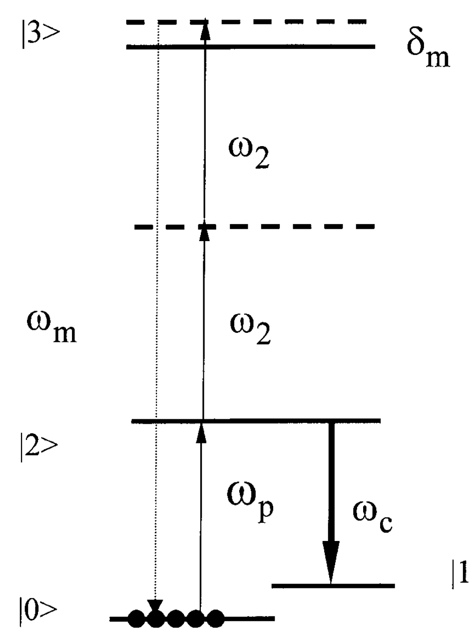

FIG. 1. Energy level diagram with relevant laser excitations for a scheme of four-wave mixing generation where the propagation velocity of the pump wave (ω_p) is modified strongly by the presence of a coupling laser (ω_c) in an EIT configuration.

atomic equations of motion describing this system can be written as

$$\dot{A}_0 = i\Omega_{02}A_2 + i\Omega_{m03}A_3\,, \tag{1a}$$

$$\dot{A}_1 = \frac{\gamma_1}{2}A_1 + i\Omega_{12}A_2\,, \tag{1b}$$

$$\dot{A}_2 = \frac{\gamma_2}{2}A_2 + i\Omega_{21}A_1 + i\Omega_{20}A_0\,, \tag{1c}$$

$$\dot{A}_3 = i\left(\delta_m + i\,\frac{\gamma_3}{2}\right)A_3 + i\Omega_{m30}A_0 + i\Omega_{32}^{(2)}A_2e^{-i\Delta kz}\,. \tag{1d}$$

Here A_j and γ_j are the jth atomic wave function amplitude and decay rate, respectively. $\Omega_{20} = D_{20}E_p/(2\hbar)$, $\Omega_{21} = D_{21}E_c/(2\hbar)$, and $\Omega_{m30} = D_{30}E_m/(2\hbar)$ are one-half of the Rabi frequencies for the respective transitions, and $\Omega_{32}^{(2)}$ is the direct two-photon Rabi frequency from state $|2\rangle$ to $|3\rangle$. In deriving these equations, we have made the appropriate phase transformation to eliminate fast oscillatory factors, and introduced the notations $\Delta k = 3k_p - k_m$ and $\delta_m = \omega_m - \omega_{30}$. The key element in the present system is the Λ scheme established among the three lower states using the coupling laser and a weak pulsed laser that also serves as the first step pump for FWM. As we will show, when the group velocity of the pump field is reduced, a phase-matched FWM wave can be produced with high efficiency because its amplitude acquires a giant enhancement. Specifically, we show that when the group velocity of the pump wave is $V_g^{(p)} \approx 1.4 \times 10^{-6}c$, a phase-matched coherent FWM field propagating with $V_g^{(m)} \approx V_g^{(p)}$ can be produced efficiently in an otherwise poorly phase-matched system.

To proceed further, and to simplify the mathematics, we will neglect ground state and pump pulse depletion. Under these conditions Eq. (1) can be solved simultaneously with Maxwell equations that describe the propagation of the pump and FWM fields. Taking $A_0 \approx 1$, and Fourier-transforming Eq. (1), we immediately obtain, in dimensionless form,

$$\alpha_2 = \tau W_{20}D(\eta)\,, \tag{2a}$$

$$\alpha_3 = \tau W_{m30}D_m(\eta) + \tau W_{20}\tau\Omega_{32}^{(2)}D_m(\eta)D(\eta)e^{-i\Delta kz}\,. \tag{2b}$$

Here, $\eta = \omega\tau$ is the dimensionless Fourier transform parameter and α_2, α_3, W_{20}, W_{m30} are the Fourier transforms of A_2, A_3, $\Omega_{20}(z,t)$, $\Omega_{m30}(z,t)$, respectively. The dimensionless dispersion functions are given by

$$D(\eta) = \frac{(\eta + i\gamma_1\tau/2)}{|\Omega_{21}\tau|^2 - (\eta + i\gamma_1\tau/2)(\eta + i\gamma_2\tau/2)}$$

and

$$D_m(\eta) = -\frac{1}{(\eta + \delta_m\tau + i\gamma_3\tau/2)}\,.$$

Notice that W_{20} and W_{m30} obey Maxwell's equations for the pump and FWM fields,

$$\frac{\partial W_{20}}{\partial z} - i\,\frac{\eta}{c\tau}\,W_{20} = i\kappa_{02}\alpha_2\,,$$
$$\frac{\partial W_{m30}}{\partial z} - i\,\frac{\eta}{c\tau}\,W_{m30} = i\kappa_{03}\alpha_3\,. \tag{3}$$

Here $\kappa_{02(03)} = 2\tau N\omega_{p(m)}|D_{02(03)}|^2/(\hbar c)$, and N is the atomic concentration. Substituting Eq. (2) into Eq. (3), one immediately obtains, for the generated wave,

$$W_{m30}(z,\eta) = \kappa_{03}c\tau^2(\tau\Omega_{32}^{(2)})W_{20}(0,\eta) \times e^{i(z/c\tau)\eta[1+\kappa_{02}c\tau^2D(\eta)/\eta]}D(\eta)D_m(\eta) \times \left(\frac{1 - e^{-i(z/c\tau)B}}{B}\right), \tag{4}$$

where $W_{20}(0,\eta) = (\tau/\sqrt{2})\Omega_{20}(0,0)e^{-\eta^2/4}$ is the Fourier transform of $\Omega_{20}(z,t)$ with a Gaussian profile at the entrance of the medium, and $B = -\Delta kc\tau + \kappa_{02}c\tau^2D(\eta) - \kappa_{03}c\tau^2D_m(\eta)$.

Equation (4) is the main result of the present work. It contains rich dynamics enabled by the strong dispersion manipulation proposed here. Close inspection of Eq. (4) reveals four main contributions of our work. (1) A coherent FWM channel which otherwise would not exist is opened by the EIT process. (2) The pump wave preserves its Gaussian pulse shape, travels with an ultraslow group velocity, and acquires negligible pulse distortion. (3) The FWM wave travels with an ultraslow group velocity that matches the ultraslowly propagating pump wave. (4) Significant enhancement to the FWM is possible by reducing the group velocity of the pump wave.

To make these results more understandable, let us consider the case where $|\Omega_{21}\tau^2| > \max(\gamma_L\tau, \gamma_2\tau)$ (γ_L is the pump wave linewidths). Under this condition the dispersion function $D(\eta)$ can be expanded and further truncated to give $D(\eta) \approx D_0 + D_1\eta + D_2\eta^2$. This leads to an analytical solution for both the pump and generated fields, thereby providing much insight into the propagation effect. If one neglects D_2, which is on the order of $1/|\Omega_{21}\tau|^4$, one finds that the pump wave preserves its Gaussian shape with negligible pulse broadening, and travels with a group velocity given by $V_g^{(p)} = c/(1 + \kappa_{02}c\tau^2\mathrm{Re}[D_1])$, where $\mathrm{Re}[D_1] \approx 1/|\Omega_{21}\tau|^2$. If the driving field can be made sufficiently weak while still preserving the conditions described above, the group velocity will be much less than the speed of light in vacuum for a sufficiently high atomic vapor concentration. A similar consideration for the dispersion function $D_m(\eta)$ indicates that the group velocity of the mixing wave is mainly determined by $V_g^{(m)} = c/[1 + \kappa_{03}c\tau^2/(\delta_m\tau)^2]$. This means that group velocity matching between the two waves is possible. Notice that to achieve phase matching $\delta_m\tau$ must be small (typically $\kappa_{03} < \kappa_{02}$). Therefore, a significant enhancement to the coherent FWM process is possible.

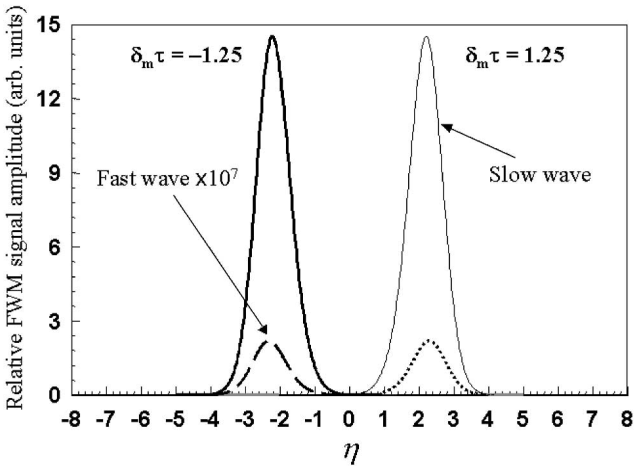

FIG. 2. Plot of the dimensionless quantity $|\frac{W_{m30}}{\tau\Omega_{32}^{(2)}\tau\Omega_{20}(0,0)/\sqrt{2}}|$ as a function of η. The thick and thin solid lines are for the ultraslow wave scheme. The dashed and dotted lines are for the fast wave scheme magnified 10^7 times. For the thick solid line and dashed line, the detuning is $\delta_m\tau = -1.25$, whereas for the thin solid line and dotted line the detuning is $\delta_m\tau = 1.25$. The parameters for the slow wave scheme are $\gamma_1\tau = 0.01$, $\gamma_2\tau = 500$, $\gamma_3\tau = 5$, $\Omega_{21}\tau = 65$, $\kappa_{02}c\tau^2 = 3 \times 10^9$, and $\kappa_{03}c\tau^2 = 3 \times 10^7$, and the phase matched group velocities are $V_g^{(p)} = V_g^{(m)} \approx 1.4 \times 10^{-6}c$. For the fast wave scheme, $\gamma_1\tau = 0.01$, $\gamma_2\tau = 500$, $\gamma_3\tau = 5$, $\Omega_{21}\tau = 650\,000$, $\kappa_{02}c\tau^2 = 3 \times 10^9$, and $\kappa_{03}c\tau^2 = 3 \times 10^7$, and the phase matched group velocities are $V_g^{(p)} \approx V_g^{(m)} \approx 0.99c$.

In Fig. 2, we have plotted the scaled Fourier transform of the mixing wave as a function of η for two sets of parameters. The first set of parameters is chosen such that a significant propagation velocity reduction to the pump wave is achieved. With these parameters, the group velocity of the pump and mixing waves are well matched at $V_g^{(p)} = V_g^{(m)} \approx 1.4 \times 10^{-6}c$. For comparison, we also show the result when the EIT process is achieved with a much wider transparency window. In this case the group velocity of the pump wave is nearly the speed of light in vacuum ($V_g^{(p)} = V_g^{(m)} \approx 0.99c$). Notice that the field amplitude for the generated wave is much smaller than that of the ultraslow pump wave case. Direct comparison of these two results shows that a giant enhancement to the coherent FWM generation can be achieved with ultraslow propagation. We point out that Eq. (4) indicates that there are two generated waves that travel with apparently different velocities. One wave is generated locally, acquiring the velocity of the pump wave $V_g^{(p)}$. The other wave was generated early in the medium, and therefore has the velocity of $V_g^{(m)}$. Close inspection of Eq. (4) shows that when phase matching is achieved, both waves are combined into one FWM wave that travels with the same group velocity as that of the ultraslow pump wave. Of course this must be the case in order to have the waves constructively interfere, resulting in a coherent FWM amplitude buildup. This result can also be clearly seen in Fig. 3, where the real part of B is plotted. This parameter contains information about the difference in the group velocity of the pump and generated wave. It is clearly seen that at the phase matched points, where $\Delta k = 0$, this velocity difference can be reduced to zero.

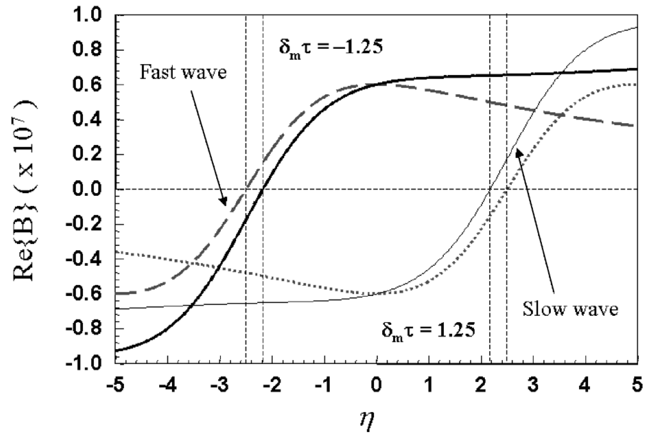

FIG. 3. Plot of the real part of the dimensionless quantity B as a function of η. Phase matching can be achieved near where $\mathrm{Re}\{B\} \approx 0$. The thick and thin solid lines are for the ultraslow pump wave case. The dashed and dotted lines are for the fast pump wave case. The detuning associated with each curve is the same as in Fig. 2.

The general features of wave mixing with extremely slow fundamental waves achieved by the dispersions/index manipulation process discussed in the present work are also expected in many other nonlinear processes [4,5]. Such significant enhancement may lead to new compact, efficient, narrow-bandwidth coherent light devices. Since the choice of state $|3\rangle$ is quite arbitrary, the significant enhancement reported here can be used to generate coherent radiation in a wavelength region where no coherent source is currently available. With this method it is also possible to vary the coupling wave Rabi frequency, thereby producing a highly efficient, yet tunable, coherent FWM output.

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