PHYSICAL REVIEW A **68**, 051801(R) (2003)

Three-photon destructive interference in ultraslow-propagation-enhanced four-wave mixing

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(Received 13 May 2003; revised manuscript received 11 August 2003; published 4 November 2003)

We show that, although the efficiency of a multiwave mixing process can be significantly enhanced by an ultra-slowly-propagating pump wave, a three-photon destructive interference between the generated wave and the pump wave limits the final achievable conversion efficiency.

DOI: 10.1103/PhysRevA.68.051801 PACS number(s): 42.50.Gy

Recent studies have shown that ultraslow propagation of optical waves may lead to significant enhancement of nonlinear optical processes [1] such as Kerr nonlinearity [2] and four-wave mixing (FWM) production [3,4]. In the latter case, it has been shown that an electromagnetically induced transparency (EIT) for the pump wave can open an efficient FWM channel that is otherwise prohibited by the strong absorption to the pump wave. The key point of such an operation is to manipulate the dispersion properties of the medium at the frequency of the pump wave so that the generated wave is phase matched at a small detuning with respect to the FWM producing state. It is this small phase-matching detuning that leads to the enhancement to the FWM production efficiency. Since such a small phase-matching detuning is closely tied to the group velocity of the phase-matched pump and generated waves, the alternate view is that the ultraslow propagation enhances the FWM production efficiency. Such an efficient nonlinear wave mixing process naturally leads to the following question: What will happen when the generated wave becomes sufficiently intense? The purpose of the present communication is to address this question. Here, we show that when the enhanced FWM field becomes sufficiently intense such a small detuning will also lead to strong absorption of the generated wave via a one-photon process. This process opens a second excitation pathway to the FWM producing state, which is π out of phase with respect to the excitation pathway provided by the laser fields. Consequently, deep in the medium where the FWM field is sufficiently intense, the two excitation pathways interfere destructively [5], resulting in no further excitation to the FWM generating state and therefore a saturated FWM production. Such a robust three-photon destructive interference thus inevitably limits the maximum enhancement achievable with the ultraslow-propagation technique.

We consider a four-level lifetime broadened atomic system that interacts with three laser fields (Fig. 1). Here, the lower three states (i.e., $|0\rangle$, $|1\rangle$, and $|2\rangle$) and a cw control laser field (Ω_c) form the usual EIT scheme. A weak CW laser field (Ω_s) provides a two-photon coupling ($|2\rangle \rightarrow |3\rangle$) to the FWM state $|3\rangle$ which is dipole coupled to the ground state $|0\rangle$. The novelty of this scheme is that with a sufficiently strong Ω_c, the $|0\rangle \rightarrow |2\rangle$ transition becomes transparent to the pump field (Ω_p). This opens an efficient FWM channel which otherwise produces no FWM because of the strong absorption of the pump wave tuned to one-photon resonance. In the Schrödinger picture the atomic equations of motion are given as [3]

$$\frac{\partial A_0}{\partial t} = i\Omega_p^* A_2 + i\Omega_m^* A_3, \tag{1a}$$

$$\frac{\partial A_1}{\partial t} = -\frac{\gamma_1}{2} A_1 + i\Omega_c^* A_2, \tag{1b}$$

$$\frac{\partial A_2}{\partial t} = -\frac{\gamma_2}{2} A_2 + i\Omega_p A_0 + i\Omega_c A_1 + i\Omega_s^{(2)*} A_3, \tag{1c}$$

$$\frac{\partial A_3}{\partial t} = i\left(\delta_m + i\frac{\gamma_3}{2}\right) A_3 + i\Omega_s^{(2)} A_2 + i\Omega_m A_0, \tag{1d}$$

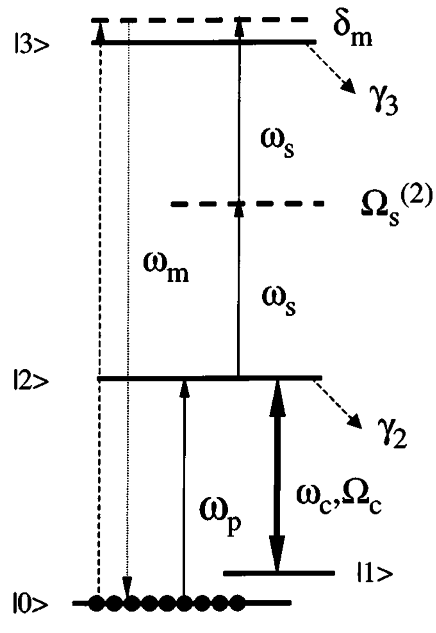

FIG. 1. Energy level diagram showing relevant laser couplings. A cw control field Ω_c creates a transparency window to reduce the absorption to the pulsed pump field Ω_p. The power of the control field is kept relatively low to significantly reduce the propagation velocity of the pump wave. A weak cw field Ω_s provides the two-photon coupling $|2\rangle \rightarrow |3\rangle$ with a FWM detuning defined as $\delta_m = \omega_m - \omega_{30}$. The dashed line represents the absorption of the generated wave, which opens a second excitation pathway to the same FWM producing state $|3\rangle$. When an alkali metal vapor is used as the medium, states $|0\rangle$ and $|1\rangle$ can be chosen from the ground state hyperfine manifold, whereas states $|2\rangle$ and $|3\rangle$ can be chosen from upper level P states.

where $\Omega_{p,c,m}$ are the half one-photon Rabi frequencies for the laser-driven transitions shown in Fig. 1. $\Omega_s^{(2)}$ is the half effective two-photon Rabi frequency for the transition $|2\rangle \rightarrow |3\rangle$. γ_j ($j=1,2,3$) is the decay rate of the state $|j\rangle$. In the following calculation, we neglect both the ground state ($A_0 \simeq 1$) and pump field depletions. We further assume that $|\Omega_c| \gg |\Omega_p|$ and the two-photon coupling is relatively weak so that the $i\Omega_s^{(2)*}A_3$ term in Eq. (1c) can be neglected. The latter step is justified since the dominant behavior of the level $|2\rangle$ is due to the EIT process maintained by the control field Ω_c [6] (also see below). With these assumptions we obtain the following adiabatic solution [7]:

$$A_2 \simeq \frac{i}{|\Omega_c|^2} \frac{\partial \Omega_p(0, t-z/V_g)}{\partial t}, \tag{2}$$

where

$$\frac{1}{V_g} = \frac{1}{c} + \frac{\kappa_{02}}{|\Omega_c|^2}. \tag{3}$$

The propagation of the FWM field at ω_m is described by

$$\frac{\partial \Omega_m}{\partial z} + \frac{1}{c}\frac{\partial \Omega_m}{\partial t} = i\kappa_{03} A_3 A_0^*, \tag{4}$$

where $\kappa_{03} = 2\pi\omega_m N |D_{03}|^2/(\hbar c)$ with N and D_{03} being the concentration and the dipole moment for the transition $|3\rangle \rightarrow |0\rangle$, respectively. Taking the Fourier transform of Eqs. (1d), (2), and (4), we obtain

$$\alpha_2 = \frac{\eta\tau}{|\Omega_c\tau|^2} \Lambda_p e^{i\eta z/(V_g\tau)}, \tag{5a}$$

$$\left(\eta + \delta_m\tau + i\frac{\gamma_3\tau}{2}\right)\alpha_3 = -\Lambda_m\tau - \Omega_s^{(2)}\tau\alpha_2, \tag{5b}$$

$$\frac{\partial \Lambda_m}{\partial z} - \frac{i\eta}{c\tau}\Lambda_m = i\kappa_{03}\alpha_3, \tag{5c}$$

where $\eta = \omega\tau$ is the dimensionless transform variable with τ being the probe pulse length at the entrance of the medium. Λ_p, Λ_m, α_2, and α_3 are the Fourier transforms of Ω_p, Ω_m, A_2, and A_3, respectively. From Eq. (5), one immediately obtains

$$\Lambda_m = -\frac{\kappa_{03}\tau}{(\eta + \delta_m\tau + i\gamma_3\tau/2)} \frac{\Omega_s^{(2)}\tau}{|\Omega_c\tau|^2} \eta\Lambda_p(0,\eta) \times e^{i\eta z/(V_g\tau)}\left(\frac{1-e^{-iKz}}{K}\right), \tag{6}$$

where

$$K = \frac{\eta}{V_g\tau} - \frac{\eta}{c\tau} + \frac{\kappa_{03}\tau}{\eta + \delta_m\tau + i\gamma_3\tau/2}.$$

Efficient FWM generation requires that the pump and the generated waves travel with closely matched group velocities. This is possible if the FWM phase-matching point is chosen properly. Notice that, when $V_g \ll c$, $|\delta_m\tau| \gg 1$ and $|\delta_m| \gg \gamma_3$, we have

$$K \simeq \frac{\eta}{V_g\tau} + \frac{\kappa_{03}\tau}{\delta_m\tau + i\gamma_3\tau/2}\left(1 - \frac{\eta}{\delta_m\tau + i\gamma_3\tau/2}\right) = K_0 + \eta\left(\frac{1}{V_g\tau} - \frac{\kappa_{03}\tau}{(\delta_m\tau + i\gamma_3\tau/2)^2}\right), \tag{7}$$

where $K_0 = \kappa_{03}\tau/(\delta_m\tau + i\gamma_3\tau/2)$. To match the group velocities of the two waves, we require

$$\frac{1}{V_g\tau} = \frac{\kappa_{03}\tau}{(\delta_m\tau)^2}.$$

Thus, a properly chosen small FWM detuning can lead to a phase-matched FWM field that propagates with an ultraslow group velocity. Insert Eq. (7) into Eq. (6), we obtain

$$\Lambda_m(z,\eta) \simeq -\frac{\Omega_s^{(2)}\tau}{|\Omega_c\tau|^2}\eta\Lambda_p(0,\eta)e^{i\eta z/(V_g\tau)} \times \left(1 - e^{-i\kappa_{03}\tau z/(|\Omega_c\tau|\sqrt{\kappa_{03}/\kappa_{02}} + i\gamma_3\tau/2)}\right), \tag{8}$$

which, after inverse transform, yields

$$\Omega_m(z,t) \simeq -i\frac{\Omega_s^{(2)}\tau}{|\Omega_c\tau|^2}\tau\frac{\partial\Omega_p(0,t-z/V_g)}{\partial t} \times \left(1 - e^{-i\kappa_{03}\tau z/(|\Omega_c\tau|\sqrt{\kappa_{03}/\kappa_{02}} + i\gamma_3\tau/2)}\right). \tag{9}$$

Notice that Eqs. (8) and (9) indicate that the generated field is inversely proportional to the intensity of the control field. Since the control field must be reduced in order to achieve ultraslow propagation, this implies the possibility of enhancement to FWM production with velocity matched ultraslow pump and generated waves. More importantly, Eqs. (8) and (9) also indicate that, once the real part of the exponent in the second term in parentheses becomes large enough, this term becomes negligible. This will always happen for large enough concentration with a given medium length. When this occurs Eq. (9) can be recast into

$$\Omega_m(z,t) = -\Omega_s^{(2)}A_2, \tag{10}$$

where we have used Eq. (2) for A_2. If we insert Eq. (10) into Eq. (1d), we find that at a sufficient depth into the medium where Eq. (10) is valid

$$\frac{\partial A_3}{\partial t} = i\left(\delta_m + i\frac{\gamma_3}{2}\right)A_3. \tag{11}$$

This indicates that from this point in the medium the two coupling terms in Eq. (1d) interfere destructively, and no further excitation can be produced to the state $|3\rangle$. Physically, when the generated wave is sufficiently intense, the absorp-

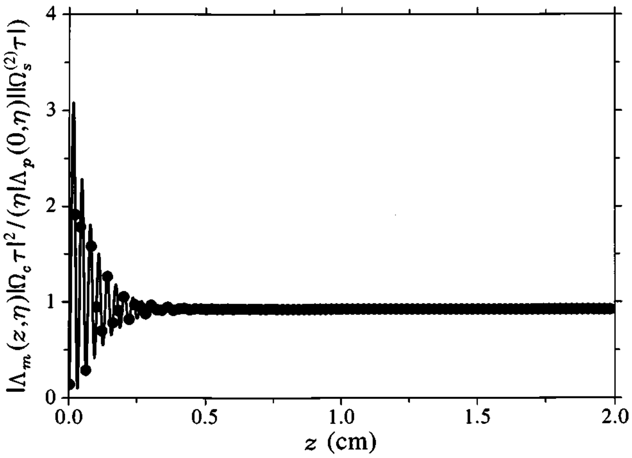

FIG. 2. Plot of dimensionless quantity $|\Lambda_m(z,\eta)|\Omega_c\tau|^2/\eta\Lambda_p(0,\eta)\Omega_s^{(2)}\tau|^2$ as a function of the propagation distance z. Parameters used are $\tau=10\ \mu$s, $\kappa_{02}=10^{11}\ \mathrm{cm}^{-1}\,\mathrm{s}^{-1}$, $\kappa_{03}=10^9\ \mathrm{cm}^{-1}\,\mathrm{s}^{-1}$, $\Omega_c\tau=628$, $\gamma_1\tau=10^{-4}$, $\gamma_2\tau=628$, $\gamma_3\tau=6.28$, and $\delta_m\tau=62.8$. With these parameters, the pump and the generated waves are group-velocity-matched at $V_g^p=V_g^m\simeq400$ m/s (these parameters are similar to those of a typical alkali metal vapor). In the plot we took $\eta=2.2$ which corresponds to the location of one of the FWM peaks (see Fig. 3) in the Fourier space. The solid curve is obtained from the exact solution [Eq. (6)], whereas the solid circles are obtained from the approximate solution [Eq. (8)]. At sufficient propagation distance the FWM field ceases growing as the three-photon destructive interference becomes effective.

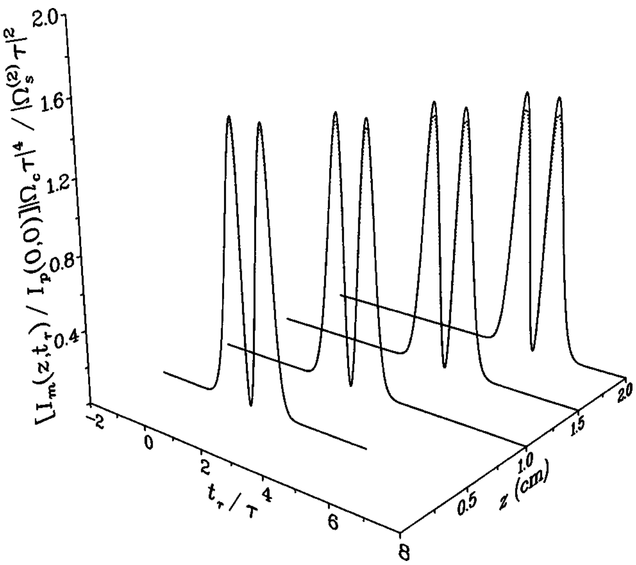

FIG. 3. Plot of dimensionless quantity $|\Omega_m(z,t)|^2|\Omega_c\tau|^4/|\Omega_p(0,0)|^2\Omega_s^{(2)}\tau|^2$ as a function of t/τ and z with a Gaussian input probe pulse. Parameters are those in Fig. 2. The dotted lines are obtained from the approximate solution [i.e., Eq. (9)] whereas the solid lines are the results of a full numerical evaluation of Eqs. (1a)–(1d) and Eq. (4) with the only assumption $|\Omega_c|\gg|\Omega_p|$. Two FWM peaks are the result of simultaneous group velocity matching of the two components due to the splitting of the level $|2\rangle$ [notice the $(\delta_m\tau)^2$ in the group velocity matching condition]. The small differences at each peak indicate the level of accuracy of our treatment.

tion of the FWM opens the second excitation pathway to the state $|3\rangle$. This excitation is π out of phase with respect to the laser excitation (i.e., $\omega_p+2\omega_s$) to the same state, resulting in a destructive interference that suppresses further excitation of the state $|3\rangle$ [5]. Therefore, the production of the FWM field saturates. Notice that Eq. (9) is independent of the density of the medium. These are the characteristics of a destructive interference [8]. This result indicates that, although a significant enhancement to the FWM generation can be obtained with an ultraslow pump wave, the three-photon destructive interference poses a limit to the overall FWM conversion efficiency.

We have carried out extensive numerical calculations to test the validity of the treatment described above. In Fig. 2, we compare, in the Fourier space, the exact solution [Eq. (6)] and the approximate solution [Eq. (8)] as a function of propagation distance. The agreement between the two methods is excellent. As predicted by Eq. (8), deep inside the medium the effective destructive interference limits the further production of the FWM field. In Fig. 3, we demonstrate the agreement between the approximate solution Eq. (9) and the full numerical solution obtained by solving Eqs. (1a)–(1d) and (4) simultaneously. Extensive and systematic numerical simulations have shown very good agreement between the two methods in the region discussed. With the parameters used in Fig. 2, the maximum difference (at the peak) between the two methods is less than 2% for $z=0.5$ cm and less than 6% for $z=2$ cm. We have therefore established, both analytically and numerically, the validity of our treatment leading to Eqs. (8) and (9).

The present study is based on the validity of the adiabatic treatment which ensures the applicability of Eq. (2). This requirement should always be observed when reducing the group velocity of the pump wave. Failure to observe this requirement can lead to significant probe pulse spread and attenuation. This is because, when the driving field Ω_c becomes very weak, as required to achieve ultraslow propagation of the probe field at a few m/s level, the transparency window becomes much narrower, resulting increased absorption to the probe field. Under this circumstance, the spontaneous emission from level $|2\rangle$ becomes significant and nonadiabatic contributions become important. Consequently, further corrections to Eq. (2) must be sought. These corrections lead to appreciable pump wave attenuation and spread, effects that must be avoided for high-efficiency and high-quality wave mixing processes. In general, for $\gamma_2\tau>1$ taking $|\Omega_c|\geqslant\gamma_2$ is sufficient to maintain a robust and effective adiabatic process. This will keep both probe field attenuation and spread at a negligible level (as seen in Fig. 3), yet still permit more than five orders of magnitude reduction of the probe field propagation velocity, and, therefore, lead to significant enhancement of the FWM production. Of course, in order to fully utilize the advantages of the EIT-assisted channel opening technique for ultraslow propagation enhanced nonlinear processes, it is necessary to delay and inhibit the onset of the three-photon destructive interference [9] discussed in the present work.

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[6] With the conditions that $|\Omega_c|\gg|\Omega_p|$ and the two-photon coupling is weak ($|\Omega_s^{(2)}\tau|<1$), a nondepleted ground state and a nondepleted pump beam are ensured even when the generated wave is absorbed efficiently. The inclusion of the two-photon back-coupling term $i\Omega_s^{(2)*}A_3$ in Eq. (1c) produces a negligible effect in the region discussed. This is because, compared with the strong excitation of state $|2\rangle$ by the control field Ω_c, such a two-photon back-coupling produces a negligible effect to the evolution of the state $|2\rangle$. In fact, for $|\Omega_s^{(2)}|\ll|\delta_m|$ this back-coupling term produces no appreciable effect at all.

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[8] This is because the generated field depends on the product of the concentration and propagation distance (i.e., Nz). Therefore, an increase in concentration will reduce the onset propagation distance for the destructive interference to take effect.

[9] In the case of the commonly known FWM scheme employing high-energy, short-pulsed lasers, the only way of delaying the onset of this detrimental effect is to either reduce the concentration or shorten the propagation distance, both significantly reducing the production of the generated wave.