Efficient multiwave mixing in the ultraslow propagation regime and the role of multiphoton quantum destructive interference

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We analyze a lifetime-broadened four-state four-wave-mixing (FWM) scheme in the ultraslow propagation regime and show that the generated FWM field can acquire the same group velocity and pulse shape as those of an ultraslow pump field. We show that a new type of induced transparency resulted from multiphoton destructive interference that significantly reduced the pump field loss. Such induced transparency based on multphoton destructive interference may have important applications in other nonlinear optical processes.

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Multiwave mixing processes in the ultraslow propagation regime have been the focus of several recent studies.[1–16] The motivation for such a novel propagation technique lies in the potentially wide range of applications in diverse fields such as high-efficiency generation of short-wavelength coherent radiation at pump intensities approaching the single-photon level, nonlinear spectroscopy at very low light intensities, quantum single-photon nonlinear optics, and quantum information science.[1–17] A common feature of many of these schemes is a three-state electromagnetically induced transparency[18–21] method in which a strong, on-resonance control field effectively splits the terminal state of the one-photon transition for a pump laser. Such an Autler–Townes doublet results in destructive interference that reduces the absorption of the pump field. This destructive interference between two single-photon channels is a key element in these studies[4–9,17] and is especially important as a channel-opening technique in the case of efficient four-wave-mxing (FWM) schemes[6,7,10] in optically dense media.

In this Letter we analyze a lifetime-broadened four-state ladder system for FWM generation (Fig. 1). We show that, with two cw laser fields (a, b) and a weak pulsed pump field (p, pulse length τ), a pulsed FWM field (f) can be efficiently generated. We further show that, when the generated FWM field has become sufficiently intense, efficient backcoupling to the FWM generating state becomes important. This backcoupling pathway leads to competitive multiphoton excitation of the FWM generating state by three supplied and one internally generated field. We demonstrate that the competition is destructive in nature, resulting in a multiphoton destructive interference–based induced transparency that efficiently suppresses the amplitudes of the states involved.

We start with atomic equations of motion (assuming a nondepleted ground state, i.e., $A_0 \simeq 1$) and wave equations for electromagnetic fields $\Omega_{p,f}$:

$$\frac{\partial A_1}{\partial t} = i(\Delta_1 + i\gamma_1)A_1 + i\Omega_p A_0 + i{\Omega_b}^* A_2, \tag{1a}$$

$$\frac{\partial A_2}{\partial t} = i(\Delta_2 + i\gamma_2)A_2 + i{\Omega_a}^* A_3 + i\Omega_b A_1, \tag{1b}$$

$$\frac{\partial A_3}{\partial t} = i(\Delta_3 + i\gamma_3)A_3 + i\Omega_a A_2 + i\Omega_f A_0, \tag{1c}$$

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\Omega_{p(f)} = i\kappa_{01(03)} A_{1(3)} {A_0}^*, \tag{1d}$$

where $2\Omega_j$ and ω_j ($j = p, c, f, a$) are the Rabi and the optical frequencies of the relevant optical field; γ_k is the decay rate of state $|k\rangle$; and $\kappa_{01(03)} = 2N\omega_{p(f)}|D_{01(03)}|^2/(c\hbar)$, with N and $D_{01(03)}$ as the concentration and the dipole moment between state $|0\rangle$ and $|1\rangle$ ($|3\rangle$), respectively. In deriving Eqs. (1a)–(1d), we define $\Delta_1 = \omega_p - \varepsilon_1/\hbar$, $\Delta_2 = \omega_p + \omega_b - \varepsilon_2/\hbar$, $\Delta_3 = \omega_p + \omega_b + \omega_a - \varepsilon_3/\hbar$, with ε_j as the energy of state $|j\rangle$ ($\varepsilon_0 = 0$). We also take slowly varying amplitude and plane-wave approximations for the pulsed fields $\Omega_{p,f}$.

We note that Eqs. (1) are a system of linear equations and can be solved formally by use of a standard Fourier-transform method. Applying this method, we obtain

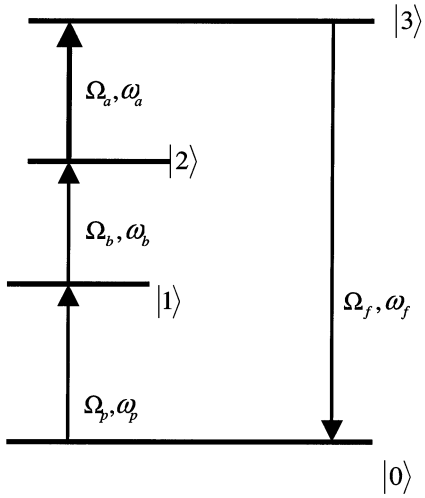

Fig. 1. Lifetime-broadened four-level atomic system interacting with two cw fields ($\Omega_{a,b}; \omega_{a,b}$), and a weak pulsed pump field ($\Omega_p; \omega_p$) to generate a FWM field ($\Omega_f; \omega_f$).

$$\Omega_p(z,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-i\omega t) \times \frac{\Lambda_p(0,\omega)[U_+ \exp(izK_-) - U_- \exp(izK_+)]}{U_+ - U_-}, \tag{2a}$$

$$\Omega_f(z,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-i\omega t) \times \frac{U_+ U_- \Lambda_p(0,\omega)[\exp(izK_-) - \exp(izK_+)]}{U_+ - U_-}, \tag{2b}$$

where $K_\pm = \omega/c + (\kappa_{03}D_f + \kappa_{01}D_p \pm G)/(2D)$, $U_\pm = (\kappa_{03}D_f - \kappa_{01}D_p \pm G)/(2\kappa_{01}\Omega_b{}^*\Omega_a{}^*)$, $G = [(\kappa_{03}D_f - \kappa_{01}D_p)^2 + 4\kappa_{03}\kappa_{01}|\Omega_b|^2|\Omega_a|^2]^{1/2}$, $D = |\Omega_b|^2(\omega + \Delta_3 + i\gamma_3) + |\Omega_a|^2(\omega + \Delta_1 + i\gamma_1) - (\omega + \Delta_1 + i\gamma_1)(\omega + \Delta_2 + i\gamma_2)(\omega + \Delta_3 + i\gamma_3)$, $D_p = (\omega + \Delta_2 + i\gamma_2)(\omega + \Delta_3 + i\gamma_3) - |\Omega_a|^2$, and $D_f = (\omega + \Delta_1 + i\gamma_1)(\omega + \Delta_2 + i\gamma_2) - |\Omega_b|^2$. In addition, we assume that at $z = 0$ the Fourier transforms of the pump and FWM fields are given by $\Lambda_p(0,\omega)$ and $\Lambda_f(0,\omega) = 0$, respectively.

Although Eqs. (2) are complex, one can gain much physical insight by seeking approximate solutions under suitable and realistic conditions. In the present study we focus on the adiabatic regime where the power series of $K_\pm$ and $U_\pm$ on ω converge rapidly. Specifically, we take $U_\pm = W_\pm + \mathcal{O}(\omega)$ and $K_\pm = (K_\pm)_{\omega=0} + \omega/V_{g\pm} + \mathcal{O}(\omega^2)$. We thus have

$$\Omega_p(z,t) = \frac{W_+\Omega_p(\eta_-)\exp(z\beta_-) - W_-\Omega_p(\eta_+)\exp(z\beta_+)}{W_+ - W_-}, \tag{3a}$$

$$\Omega_f(z,t) = \frac{W_+W_-[\Omega_p(\eta_-)\exp(z\beta_-) - \Omega_p(\eta_+)\exp(z\beta_+)]}{W_+ - W_-}, \tag{3b}$$

where $\eta_\pm = t - z/V_{g\pm}$, $\beta_\pm = i(K_\pm)_{\omega=0}$, $1/V_{g\pm} = \mathrm{Re}[(\partial K_\pm/\partial\omega)_{\omega=0}]$, $\beta_+ \simeq (-\kappa_{01}\kappa_{03}\gamma_2 + i\kappa_{01}\kappa_{03}\Delta_2)/(\kappa_{01}|\Omega_a|^2 + \kappa_{03}|\Omega_b|^2)$, $\beta_- \simeq -(\kappa_{01}|\Omega_a|^2 + \kappa_{03}|\Omega_b|^2) \times k_0{}^2(B_1 + iB_2)(B_1{}^2 + B_2{}^2)$, and $1/V_g \equiv 1/V_{g+} \simeq 1/c + \kappa_{01}\kappa_{03}/(\kappa_{01}|\Omega_a|^2 + \kappa_{03}|\Omega_b|^2)$. In deriving these results we define $B_1 \equiv |\Omega_b|^2\Delta_3 + \Delta_1|\Omega_a|^2$ and $B_2 \equiv |\Omega_b|^2\gamma_3 + \gamma_1|\Omega_a|^2$ and use conditions $|\Omega_b|^2, |\Omega_a|^2 \gg |(\Delta_2 + i\gamma_2)(\Delta_3 + i\gamma_3)|$. These conditions are consistent with the assumption that a well-behaved adiabatic process is required for rapid conversion of a power-series expansion.

Close inspection of the expressions of $\beta_\pm$ indicates that under these conditions we have $\mathrm{Re}[\beta_\pm] < 0$ and $|\mathrm{Re}[\beta_\pm]| \ll |\mathrm{Re}[\beta_-]|$. The key consequence of this result is that under these conditions the η_- velocity component decays much faster than the η_+ component. Consequently, after a characteristic propagation distance one has

$$\Omega_p(z,t) = \frac{W_-\exp(z\beta_+)}{W_- - W_+}\,\Omega_p\!\left(t - \frac{z}{V_g}\right), \tag{4a}$$

$$\Omega_f(z,t) = \frac{W_+W_-\exp(z\beta_+)}{W_- - W_+}\,\Omega_p\!\left(t - \frac{z}{V_g}\right)\cdot \tag{4b}$$

Several interesting and important features in Eqs. (4) are worth noticing. First, it is possible, with experimentally achievable parameters, to obtain $V_g/c \equiv V_{g+}/c \ll 1$ (see the numerical example below). In fact, under the conditions specified, the generated FWM field travels with the same ultraslow group velocity as the pulsed pump field and also retains the same temporal profile. Second, from Eq. (4b) we can calculate the efficiency η at $z = L$:

$$\eta \simeq \frac{|W_+W_-|^2}{|W_- - W_+|^2}\exp(-\Gamma L), \tag{5}$$

where $\Gamma = -2\mathrm{Re}[\beta_+] \simeq 2\kappa_{01}\kappa_{03}\gamma_2/(\kappa_{01}|\Omega_a|^2 + \kappa_{03}|\Omega_b|^2)$. With the parameters given below and assuming $L = 2$ mm, we find $\eta \simeq 3 \times 10^{-4}$. Finally, but more important, is the existence of multiphoton destructive interference that leads to multiple induced transparencies. To see this we consider a propagation depth at which Eqs. (4) are valid. Taking the ratio of Eqs. (4) we obtain $\Omega_p(z,t)/\Omega_f(z,t) \simeq 1/W_+ \simeq \Omega_b{}^*/\Omega_a$. With this result it is straightforward to show that $\Omega_p + \Omega_b{}^*A_2 \simeq 0$, $\Omega_a{}^*A_3 + \Omega_bA_1 \simeq 0$, and $\Omega_aA_2 + \Omega_f \simeq 0$. When these results are used in Eqs. (1) (note $A_0 \simeq 1$), it can be seen that the amplitudes of all three upper atomic states $|j\rangle$ ($j = 1,2,3$) are strongly suppressed. Physically, when the FWM field is sufficiently intense an additional excitation channel to the state $|1\rangle$ occurs, i.e., $|0\rangle \to |1\rangle$ via $\Omega_f + \Omega_a{}^* + \Omega_b{}^*$. This excitation is 180° out of phase with respect to the excitation $|0\rangle \to |1\rangle$ provided by Ω_p, resulting in suppression of state $|1\rangle$, as indicated by $\Omega_p + \Omega_b{}^*A_2 = 0$. Extensive numerical calculations of Eqs. (2) have shown excellent agreement with the above-described analytical approximations under the conditions specified.

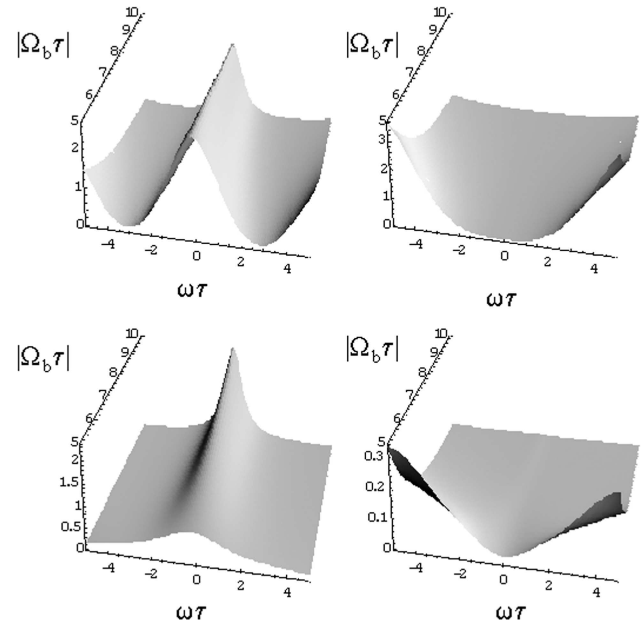

Fig. 2. Surface plots of $10^{-10}|\alpha_1/(\Lambda_p\tau)|^2$ (top) and $10^{-11}|\alpha_3/(\Lambda_p\tau)|^2$ (bottom) versus $\omega\tau$ and $|\Omega_b\tau|$ for both small z (left, destructive interference is ineffective) and for large z (right, destructive interference is effective). Parameters: $\tau = 10^{-6}$/s, $\Omega_a\tau = 3$, $\gamma_1\tau = 5.9$, $\gamma_2\tau = 0.8$, $\gamma_3\tau = 0.09$, $\Delta_1\tau = \Delta_2\tau = 0$, and $\Delta_3\tau = 0.2$. These parameters are experimentally achievable with typical magneto-optically trapped rubidium atoms.

An experimental candidate for the proposed system is ^{85}Rb atoms[4] (for instance, $|0\rangle = |5S_{1/2}\rangle$, $|1\rangle = |5P_{1/2}\rangle$, $|2\rangle = |5D_{3/2}\rangle$, and $|3\rangle = |nP_{3/2}\rangle$ with $n > 10$). The respective transitions are $|0\rangle \to |1\rangle$ at 795 nm, $|1\rangle \to |2\rangle$ at 762 nm, and $|2\rangle \to |3\rangle$ at 1.3–1.5 μm. We take, at $z = 0$, $\tau = 10^{-6}$/s, $\kappa_{01} = 100\kappa_{03} = 10^9/(\text{s cm})$, $\gamma_1\tau \simeq 5.9$, $\gamma_2\tau \simeq 0.8$, $\gamma_3\tau \simeq 0.09$, $\Delta_1\tau = \Delta_2\tau \simeq 0$, $\Delta_3\tau \simeq 0.2$, $\Omega_b\tau \simeq 5$, and $\Omega_a\tau \simeq 3$.

Figure 2 shows surface plots of $|\alpha_1/\Lambda_p|^2$ (top) and $|\alpha_3/\Lambda_p|^2$ (bottom) as a function of $\omega\tau$ and $|\Omega_b\tau|$ for $z \simeq 0$ (left, destructive interference is ineffective) and for large z (right, destructive interference is effective). Here α_j ($j = 1, 3$) are the Fourier transforms of A_j, which can be obtained directly from Eqs. (1). It can be seen that, for small z, the field Ω_a has introduced loss within the transparency window (a sizable α_1 near $\omega\tau \simeq 0$, top left). When the multiphoton destructive interference with state $|1\rangle$ is effective, however, high transparency is achieved (top right). Correspondingly, at small z the generated field grows linearly (near $\omega\tau \simeq 0$, bottom left), whereas when the destructive interference is effective the generated field ceases to grow (bottom right). These calculations agree well with the analytical solutions in Eqs. (3) and (4) under the conditions specified. The typical difference between the two methods is <5%.

In summary, we have analyzed an efficient FWM scheme in the ultraslow propagation regime. We have shown a new type of induced transparency based on highly efficient and multiphoton destructive interference that can lead to effective suppression of excitations of all upper atomic states. Consequently, the generated FWM field and the pump field propagate with the same ultraslow group velocity and retain the same pulse profiles. Our theory and results may offer new possibilities for future applications of the index manipulation technique, which is particularly important for nonlinear optical processes in the ultraslow propagation regime.

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