Efficient parametric amplification in double-Λ systems without maximal two-photon coherence

H. Shpaisman, A. D. Wilson-Gordon, and H. Friedmann
Department of Chemistry, Bar-Ilan University, Ramat Gan 52900, Israel
(Received 15 July 2004; published 22 December 2004)

We study four-wave mixing in a double-Λ system, for cw laser beams whose transverse intensity profiles (TIP's) are initially Gaussian, by solving the Maxwell-Bloch equations numerically. For systems where coherent population trapping (CPT) is initially absent, we show that efficient frequency conversion, without focusing or ring formation, can occur even at distances which are much shorter than those required to establish CPT. We also show, for certain configurations, that blue-detuned beams become focused on propagation so that very high frequency conversion, even exceeding 100% at the center of the profile, can occur. This focusing is, however, accompanied by ring formation. We show that focusing, without ring formation, can occur for identical blue-detuned beams when the initial relative phase is π so that CPT cannot be established on propagation. The behavior of the TIP's of the beams on propagation is explained by considering the effective linear and third-order contributions to the off-diagonal density-matrix elements.

DOI: 10.1103/PhysRevA.70.063814 PACS number(s): 42.65.Jx, 42.65.Ky, 42.50.Gy

I. INTRODUCTION

Modern nonlinear optics exploits quantum interference and coherence effects in order to control and modify the properties of the interacting optical fields and material systems. One of the most studied schemes is the double-Λ system, which consists of four atomic or molecular states interacting with four near-resonant laser beams so that a closed loop is formed. An important property of these systems is that both the initial relative phases and amplitudes of the electromagnetic fields determine the populations and coherences of the atoms [1–6] as well as the properties of the fields on propagation [6]. The double-Λ system has been investigated in the context of amplification without inversion [7–10], phase-sensitive laser cooling [11], the propagation of pairs of optical pulses [12], optical phase conjugation [13–16], phase control of photoionization [17], resonantly enhanced four-wave mixing (FWM) [6,13,18–24], cavity quantum electrodynamics (QED) [25], phase control of electromagnetically induced transparency [26,27] and coherent population trapping (CPT) [28], Ramsey fringes [29], light storing of a pair of pulses [30,31], and dynamic optical bistability [32]. Recently, Morigi *et al.* [33] have compared the phase-dependent properties of the ◇ (diamond) four-level system with those of the double-Λ system.

In this article, we discuss the behavior of the initially Gaussian transverse intensity profiles (TIP's) of cw copropagating beams that interact with a double-Λ system. Previous studies of propagation in four-level double-Λ systems [6,7,12,13,30,31,34] or in their five-level modified version [35], proposed by Johnsson and Fleischhauer [36–38], have considered either cw [6,13,35] or pulsed plane wave fields [12,24,30,31]. However, none of them have considered the role of the transverse intensity profiles of the interacting beams in enhancing FWM, although the potential significance of doing so was pointed out by Korsunsky and Kosachiov [6]. Harris and coworkers [39–43] have shown that if maximum two-photon (Raman) coherence can be established between the two lower states of the system, highly efficient FWM occurs within a coherence length, so that self-focusing and phase matching become irrelevant. (For earlier work on the role of maximal coherence in coherent anti-Stokes resonance Raman systems which are also double-Λ systems, see Refs. [44,45].) This is quite different from the situation in the pumped two-level system [46–49] where parametric amplification of the FWM and probe fields increases with the propagation length [49].

Here we demonstrate that for systems where CPT is initially absent, considerable enhancement of the FWM can occur even at distances which are an order of magnitude shorter than those required to establish CPT. Maximum frequency conversion can be achieved before the beams become focused, defocused, or develop a ring around the central peak which would be indicative of potential azimuthal-symmetry breakup [50]. We also show that when the beams are blue detuned and the nonlinearity of the medium is reduced considerably, self-focusing of the beams leading to very high frequency conversion can be obtained, near the axis of propagation of the copropagating beams. This self-focusing is, however, accompanied by ring formation which may lead to breakup [50]. Only by solving the Maxwell-Bloch equations in three dimensions, rather than in two dimensions as presented here, will it be possible to determine at what propagation length breakup will occur. In all these configurations, the laser at the FWM frequency is initially very weak. On propagation, FWM is generated with the correct phase so that CPT conditions can be established [6].

We also compare the propagation of four beams with equal Rabi frequencies and detunings to the blue, when $\Phi=0$ and $\Phi=\pi$. When $\Phi=0$, CPT exists from the outset and the beams propagate unchanged for a length that is short compared to the diffraction length. However, when $\Phi=\pi$ so that CPT is absent throughout propagation, and the nonlinearity sets in at a suitable length, all four beams can be focused without ring formation.

We show that the behavior of the transverse intensity profiles of the beams, on propagation, can be explained by studying the effective linear and third-order contributions to the off-diagonal density-matrix elements, as a function of the beam profile.

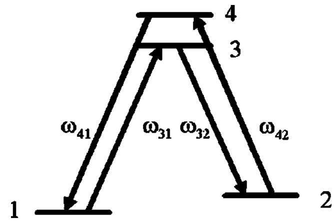

FIG. 1. Energy level scheme for the double-Λ system interacting with four beams.

II. THE MODEL

The four-level double-Λ system is depicted in Fig. 1. The lower Λ system consists of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$, and the upper Λ system consists of the states $|1\rangle$, $|2\rangle$, and $|4\rangle$. Each $|j\rangle \to |i\rangle$ transition (with $j=1$, 2 and $i=3,4$ throughout the paper) interacts with an electromagnetic field

$$\vec{E}_{ij}(\vec{r},t) = (1/2)\hat{x}_{ij}E_{ij}(r)\exp[-i(\omega_{ij}t - k_{ij}z + \varphi_{ij})] + \text{c.c.}, \tag{1}$$

with unit polarization vector $\hat{x}_{ij}$, frequency ω_{ij}, wave vector k_{ij}, and initial phase φ_{ij}, whose detuning from the transition frequency ω'_{ij} is $\Delta_{ij}=\omega'_{ij}-\omega_{ij}$ and whose Rabi frequency is $2V_{ij}(r)=\mu_{ij}E_{ij}(r)/\hbar$.

The first step is to write the Bloch equations [51] for the double-Λ system [6]. It should be pointed out that the Bloch equations for the off-diagonal elements of the density matrix are the same for all four-level systems that interact with four fields so that a loop is formed. The equations for the diagonal elements differ only in the decay terms. The Bloch equations are given by

$$\dot{\rho}_{11} = i(V_{13}\rho'_{31} + V_{14}\rho'_{41} - V_{31}\rho'_{13} - V_{41}\rho'_{14}) - \gamma_{12}\rho_{11} + \gamma_{21}\rho_{22} + \gamma_{31}\rho_{33} + \gamma_{41}\rho_{44}, \tag{2}$$

$$\dot{\rho}_{22} = i(V_{23}\rho'_{32} + V_{24}\rho'_{42} - V_{32}\rho'_{23} - V_{42}\rho'_{24}) + \gamma_{12}\rho_{11} - \gamma_{21}\rho_{22} + \gamma_{32}\rho_{33} + \gamma_{42}\rho_{44}, \tag{3}$$

$$\dot{\rho}_{33} = i(V_{31}\rho'_{13} + V_{32}\rho'_{23} - V_{13}\rho'_{31} - V_{23}\rho'_{32}) - \gamma_{3}\rho_{33} + \gamma_{43}\rho_{44}, \tag{4}$$

$$\dot{\rho}_{44} = i(V_{41}\rho'_{14} + V_{42}\rho'_{24} - V_{14}\rho'_{41} - V_{24}\rho'_{42}) - \gamma_{4}\rho_{44}, \tag{5}$$

$$\dot{\rho}'_{21} = i(V_{23}\rho'_{31} + aV_{24}\rho'_{41} - V_{31}\rho'_{23} - aV_{41}\rho'_{24}) - (\Gamma_{21} + i\Delta_{21})\rho'_{21}, \tag{6}$$

$$\dot{\rho}'_{31} = i(V_{31}\rho_{11} + V_{32}\rho'_{21} - V_{31}\rho_{33} - V_{41}\rho'_{34}) - (\Gamma_{31} + i\Delta_{31})\rho'_{31}, \tag{7}$$

$$\dot{\rho}'_{32} = i(V_{32}\rho_{22} + V_{31}\rho'_{12} - V_{32}\rho_{33} - a^{*}V_{42}\rho'_{34}) - (\Gamma_{32} + i\Delta_{32})\rho'_{32}, \tag{8}$$

$$\dot{\rho}'_{41} = i(V_{41}\rho_{11} + a^{*}V_{42}\rho'_{21} - V_{31}\rho'_{43} - V_{41}\rho_{44}) - (\Gamma_{41} + i\Delta_{41})\rho'_{41}, \tag{9}$$

$$\dot{\rho}'_{42} = i(V_{42}\rho_{22} + aV_{41}\rho'_{12} - aV_{32}\rho'_{43} - V_{42}\rho_{44}) - (\Gamma_{42} + i\Delta_{42})\rho'_{42}, \tag{10}$$

$$\dot{\rho}'_{43} = i(V_{41}\rho'_{13} + a^{*}V_{42}\rho'_{23} - V_{13}\rho'_{41} - a^{*}V_{23}\rho'_{42}) - (\Gamma_{43} + i\Delta_{43})\rho'_{43}, \tag{11}$$

where $a=\exp(i\Phi)$ and $\Phi=\varphi_{31}-\varphi_{32}+\varphi_{42}-\varphi_{41}$ is the initial relative phase, γ_{kl} is the longitudinal decay rate from state $|k\rangle \to |l\rangle$, γ_i is the total decay rate from state $|i\rangle$, and $\Gamma_{kl}=0.5(\gamma_k+\gamma_l)+\Gamma^{*}_{kl}$ is the transverse decay rate of the off-diagonal density-matrix element ρ'_{kl}, where Γ^{*}_{kl} is the rate of phase-changing collisions. The rapidly oscillating terms have been eliminated by the substitutions

$$\rho'_{ij} = \rho_{ij}\exp[-i(\Delta_{ij}t + k_{ij}z - \varphi_{ij})], \tag{12}$$

where $i=1$, 2, and $j=3$, 4, and

$$\rho'_{21} = \rho_{21}\exp\{-i[(\Delta_{31}-\Delta_{32})t + (k_{31}-k_{32})z - (\varphi_{31}-\varphi_{32})]\}, \tag{13}$$

$$\rho'_{43} = \rho_{43}\exp\{-i[(\Delta_{41}-\Delta_{31})t + (k_{41}-k_{31})z - (\varphi_{41}-\varphi_{31})]\}. \tag{14}$$

It is only possible to write the Bloch equations in this form when the multiphoton resonance condition $\omega_{31}-\omega_{32}+\omega_{42}-\omega_{41}=0$ is satisfied. This condition can be rewritten in terms of the one-photon detunings as $\Delta_{31}-\Delta_{32}=\Delta_{41}-\Delta_{42}=\Delta_{21}$, where Δ_{21} is the two-photon or Raman detuning or, alternatively, $\Delta_{41}-\Delta_{31}=\Delta_{42}-\Delta_{32}=\Delta_{43}$.

In addition to solving the steady-state Bloch equations numerically, we have also obtained analytical formulae which express the off-diagonal density matrix elements in terms of the populations of the states. These formulas are a generalization of those previously developed for the three-level Λ system [51] and for the double-Λ system [21] in the case where V_{31} and V_{42} are strong, and V_{32} and V_{41} are weak (strong-weak-strong-weak configuration), and it is assumed that the strong fields remain constant. We find that the analytical expression for ρ'_{ij} can be decomposed into a sum of terms that are linear and third order in the Rabi frequency,

$$\rho'_{ij} = \rho'^{(1)}_{ij} + \rho'^{(3)}_{ij}, \tag{15}$$

or, more explicitly, as

$$\rho'_{31} = \tilde{\chi}^{(1)}_{31}V_{31} + a\tilde{\chi}^{(3)}_{31}V_{32}V_{24}V_{41}, \tag{16}$$

$$\rho'_{32} = \tilde{\chi}^{(1)}_{32}V_{32} + a^{*}\tilde{\chi}^{(3)}_{32}V_{31}V_{14}V_{42}, \tag{17}$$

$$\rho'_{41} = \tilde{\chi}^{(1)}_{41}V_{41} + a^{*}\tilde{\chi}^{(3)}_{41}V_{42}V_{23}V_{31}, \tag{18}$$

$$\rho'_{42} = \tilde{\chi}^{(1)}_{42}V_{42} + a\tilde{\chi}^{(3)}_{42}V_{41}V_{13}V_{32}, \tag{19}$$

where $\tilde{\chi}^{(1)}_{ij}$ and $\tilde{\chi}^{(3)}_{ij}$ are proportional to the effective linear and third-order susceptibilities $\chi^{(1,3)}_{ij}$ [52]. The real and imaginary parts of the effective linear susceptibility $\chi^{(1)}_{ij}$ are proportional to the refraction and absorption of the field that interacts with the $|i\rangle \to |j\rangle$ transition, and the effective third-order susceptibility $\chi^{(3)}_{ij}$ gives the contribution to the nonlinear polarization at ω_{ij} from FWM. The role played by the phase is clearly seen in Eqs. (16)–(19). If $\Phi=0$, the two contributions to ρ'_{ij} interfere, either constructively or destructively, depending on their relative signs. If the phase is switched to $\Phi=\pi$, the constructive interference is replaced by destructive interference, or vice versa. It is important to realize that $\chi_{ij}^{(1,3)}$ are themselves phase dependent, since they can be expressed in terms of the populations whose phase dependence arises from that of the coherences [see Eqs. (2)–(11)]. Unfortunately, the analytical expressions for the susceptibilities are too unwieldy to reproduce here. However, as we show in Sec. III, their numerical evaluation gives important physical insight into the behavior of the system. We can calculate the phase mismatch introduced by interaction with the medium from the expression

$$\Delta k \simeq (\omega/c)(n_{31} - n_{32} + n_{42} - n_{41}), \tag{20}$$

where [52] $n_{ij}^2 = 1 + 4\pi \,\mathrm{Re}\, \chi_{ij}^{(1)}$, and the copropagating laser beams are assumed to be close in frequency. This assumption also allows us to neglect Doppler broadening. As $\chi_{ij}^{(1)}$ depends on the intensity of all four beams, Δk varies as a function of both the beam radius and the propagation length. In our work on parametric amplification (PA) in the two-level system interacting with a strong pump and weak probe, we found that the phenomenon of electromagnetically induced phase matching (EIPM), in which Δk becomes zero for certain pump intensities on propagation [49], plays a crucial role in determining the magnitude of the PA. However, we show here that the distance at which PA occurs in the double-Λ system is generally very short, so that EIPM is unimportant.

In order to study the propagation of the beams, we solve the Maxwell-Bloch equations, in the paraxial approximation, which may be written in the form [46–49]

$$\frac{\partial}{\partial z} V'_{ij} = \frac{i}{4L_D} \nabla_T^2 V'_{ij} + \frac{i}{L_{ij}} \rho'_{ij}, \tag{21}$$

where

$$\nabla_T^2 = \partial^2/\partial\xi^2 + (1/\xi)\partial/\partial\xi + (1/\xi^2)\partial^2/\partial\theta^2 \tag{22}$$

is the transverse Laplacian in dimensionless cylindrical coordinates, $\xi = r/\sqrt{2} w_{31}(0)$, where $w_{31}(0)$ is the initial spot size of the field at frequency ω_{31}, $V'_{ij} = V_{ij}/\Gamma_{31}$ is the dimensionless Rabi frequency, the parameter $L_D = k[w_{31}(0)]^2$ is the diffraction length, and the parameter $L_{ij} = \hbar \Gamma_{31}/\pi k N \mu_{ij}^2 = 4/\alpha_{ij}(0)$, where $\alpha_{ij}(0)$ is the unsaturated line-center absorption coefficient for the $|j\rangle \rightarrow |i\rangle$ transition. In the calculations we assume that $L_{ij} = L_{NL}$ (NL stands for nonlinear) for all the transitions. The ratio $L_{rel} = L_{NL}/L_D$ expresses the propagation distance at which the nonlinearity becomes important, relative to the length at which diffraction becomes important. Thus for a constant value of L_D, decreasing the value of L_{rel} ensures that the nonlinearity takes effect at a shorter propagation distance.

We solve the Maxwell-Bloch equations numerically for both plane waves (PW's) and beams whose initial transverse intensity profiles are Gaussian with the same waist sizes:

$$V'_{ij} = V'_{ij}(0)\exp(-\xi^2). \tag{23}$$

In all the calculations presented here, we assume that $\Gamma'_{ij} = \Gamma_{ij}/\Gamma_{31} = 1$ for all four one-photon transitions, $\gamma_{43} = 0$, $\gamma'_{21} = \gamma'_{12} = \gamma_{12}/\Gamma_{31} = 10^{-5}$, and $\Gamma^*_{ij} = 0$. In order to compare PW's and Gaussian beams, we assume that the initial Rabi frequencies of the beams in the PW approximation are equal to the initial values of $V'_{ij}(0)$, the on-axis Rabi frequencies of the Gaussian TIP's of the beams.

III. NUMERICAL RESULTS

A. CPT and maximal two-photon coherence

In this section, we examine the well-known situation where CPT with maximal two-photon coherence ($|\rho'_{21}|^2 = \rho_{11}\rho_{22} = 1/4, V'_{31}/V'_{32} = V'_{41}/V'_{42}$) either already exists at $z=0$, or is achieved after a short propagation distance [6,39–43]. We show that maximum conversion of $V'_{42}(0)$ to $V'_{41}(0)$ can be achieved before focusing or ring formation sets in, and even before CPT is achieved. In the examples discussed below, the beams that interact with the lower Λ system have equal Rabi frequencies $[V'_{31}(0) = V'_{32}(0) = 8]$ and are equally detuned to the red or blue ($\Delta'_{31} = \Delta'_{32} = \pm 4$ where $\Delta'_{ij} = \Delta_{ij}/\Gamma_{31}$), so that the two-photon detuning $\Delta_{21} = 0$. If we were dealing with a single-Λ system, we would obtain CPT with $|\rho'_{21}|^2$ remaining constant at its maximum value of $1/4$ throughout propagation, and $\chi_{ij}^{(1)} = 0$.

When $V'_{41}(0)$ and $V'_{42}(0)$ are very weak, only minor deviations from this situation are expected. We first consider such a case where, initially, $V'_{41}(0) = 0.001$, $V'_{42}(0) = 1$, with $\Delta'_{ij} = \pm 4$ and $L_{rel} = 1.66 \times 10^{-4}$. In Fig. 2(a), we plot the two-photon coherences, $|\rho'_{21}|^2$ and $|\rho'_{43}|^2$, and the populations at the center ($\xi = 0$) of the Gaussian beams. As expected, the on-axis two-photon coherence $|\rho'_{21}|^2$ rapidly achieves its maximum value. In Fig. 2(b), the TIP's are plotted as a function of z/L_D, and we see that they remain Gaussian on propagation. In Fig. 2(c), we compare the amplitudes of the PW beams with those of the Gaussian beams at line center. We see that the strong fields $V'_{31}(0)$ and $V'_{32}(0)$ remain almost constant, whereas $V'_{42}(0)$ is strongly converted to $V'_{41}(0)$, with maximum conversion of 74% at $z/L_D = 0.002$. The behavior of $V'_{41}(0)$ and $V'_{42}(0)$, on propagation, is the same for both PW and Gaussian beams with either positive or negative detuning.

We now consider the case of large detuning $|\Delta'_{41}| = |\Delta'_{42}| = 100$, with $V'_{41}(0) = 0.001$, and $L_{rel} = 1.66 \times 10^{-4}$, as in the previous example. However, in this case we take a larger initial value for $V'_{42}(0)$, namely, $V'_{42}(0) = 8$. In Fig. 3(a), we plot the two-photon coherences, and the populations at the center ($\xi = 0$) of the Gaussian beams. We see that the two-photon coherence and populations of the lower levels oscillate as a function of the propagation length z/L_D, while maintaining $|\rho'_{21}|^2 = \rho_{11}\rho_{22}$ which can deviate strongly from its maximum value [6]. In Fig. 3(b), the TIP's are plotted as a function of z/L_D and we see that the they remain Gaussian on propagation. In Fig. 3(c), we compare the amplitudes of the PW beams with those of the Gaussian beams at line center, and note that there is very little difference between

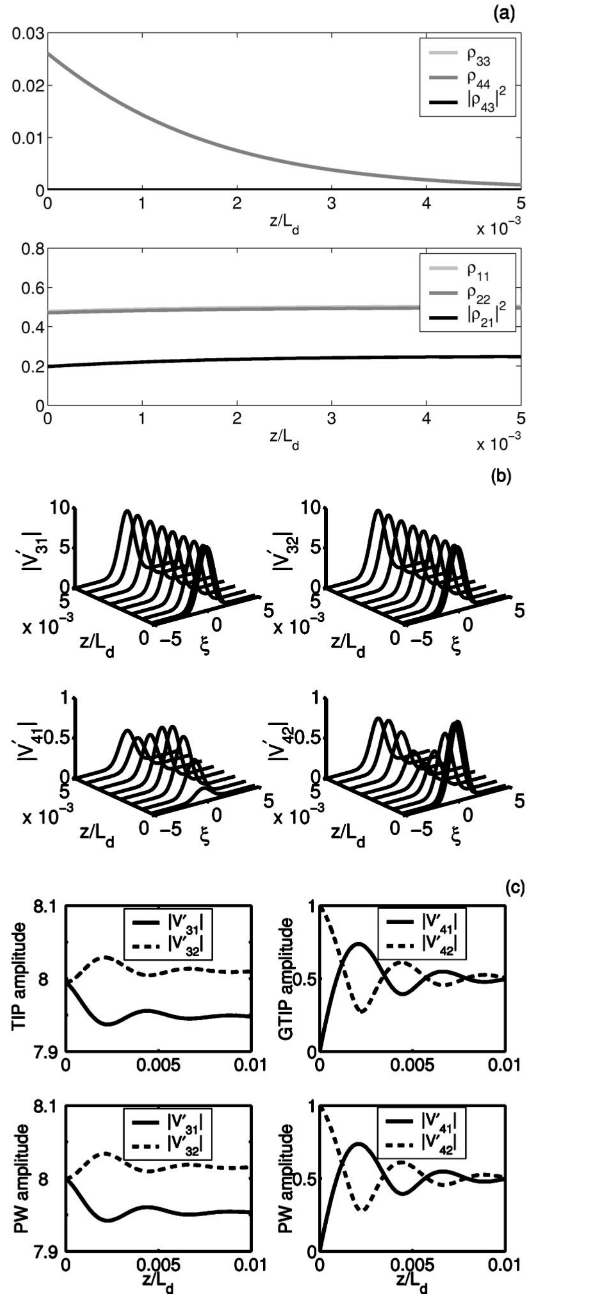

FIG. 2. Double-Λ system in which CPT with maximal two-photon coherence exists at $z \simeq 0$. (a) Populations and two-photon coherences of Gaussian beams at $\xi = 0$, as a function of z/L_D; (b) TIP's (V'_{ij} vs ξ) of propagating beams as a function of z/L_D; (c) comparison between Rabi frequencies of Gaussian (at $\xi = 0$) and PW beams as a function of z/L_D. Initial Rabi frequencies are $V'_{31}(0) = V'_{32}(0) = 8$, $V'_{41}(0) = 0.001$, and $V'_{42}(0) = 1$. Detunings are $\Delta'_{ij} = \pm 4$, and $L_{rel} = 1.66 \times 10^{-4}$.

the two cases. The maximum conversion of $V'_{42}(0)$ to $V'_{41}(0)$ is 87%, and occurs at $z/L_D = 0.047$, at the height of the first oscillation. This distance, which is an order of magnitude larger than in the previous case, can be shortened by reducing the detuning in the upper Λ system. However, as the detuning decreases, the conversion becomes less efficient. For example, when $|\Delta'_{41}| = |\Delta'_{42}| = 50$, the maximum conversion is 85% at $z/L_D = 0.024$ and when $|\Delta'_{41}| = |\Delta'_{42}| = 25$, the maximum conversion is 82% at $z/L_D = 0.014$. As the detuning decreases, the initial deviation from CPT ($|\rho_{21}|^2 = \rho_{11}\rho_{22}$) increases. Although this deviation decreases on propagation, it may still be significant at the point where maximum conversion takes place.

In order to reinforce this point, let us decrease the detuning even further to $|\Delta'_{41}| = |\Delta'_{42}| = 10$. The maximum conversion of 73% is achieved at $z/L_D = 0.009$ where the deviation from CPT is -0.074, $|\rho_{21}|^2 = 0.057 \ll 0.25$, and $\rho_{33} \simeq \rho_{44} = 0.1$. CPT only occurs at $z/L_D \simeq 0.1$ which is an order of magnitude greater than the distance where maximum conversion is achieved. As can be seen from Fig. 3(d), maximum conversion is achieved before the beams become focused, defocused or develop a ring around the central peak which would be indicative of potential azimuthal-symmetry breakup [50]. Thus it is possible to obtain significant conversion in the absence of CPT at very small propagation distances. Of course, one cannot reduce the detuning indefinitely. By $|\Delta'_{41}| = |\Delta'_{42}| = 6$, ring formation, but not focusing, occurs before maximum conversion is achieved.

B. Focusing in the absence of CPT

The question now arises as to whether it is possible to choose parameters that will lead to focusing of the beams and hence to enhanced conversion of $V'_{42}(0)$ to $V'_{41}(0)$. It turns out that it is possible, provided one chooses all the beams to be detuned to the blue, and increases the value of L_{rel} so that the nonlinearity sets in at a distance that is sufficiently long for focusing to build up. We discuss two possible configurations for achieving focusing.

1. Three strong fields

We first consider the case in which three of the beams are equally strong, $V'_{31}(0) = V'_{32}(0) = V'_{42}(0) = 8$, while $V'_{41}(0) = 0.001$, all the beams are equally detuned to the blue, $\Delta'_{ij} = -4$ and L_{rel} is increased to 1.52×10^{-3}. In Fig. 4(a), we compare the amplitudes of the PW beams with those of the initially Gaussian beams on axis, and note that the maximum conversion of $V'_{42}(0)$ to $V'_{41}(0)$ for the case of the Gaussian beams exceeds 100%, reaching 122% at $z/L_D = 0.12$, as opposed to only 60% for the PW beams. For these parameters, CPT with $|\rho'_{21}|^2 = 0.2$ is established on propagation at $z/L_D \simeq 0.24$ which is approximately twice the length at which maximum conversion takes place. Thus, in this case, focusing leads to very high on-axis frequency conversion well before CPT is established. In Fig. 4(b), we plot the TIP's of the initially Gaussian beams, as they propagate. In contrast to the configurations discussed in Sec. III A, the TIP's do not retain their Gaussian shape but develop into a central focused beam surrounded by a much weaker ring. If we follow the changes in V'_{41} on propagation, we see that at small values of z/L_D, there are two peaks in the wings of the TIP which grow on propagation, and eventually form the ring around the central peak. The origin of the two peaks can be traced to the behavior of ρ'_{41} as a function of ξ at $z = 0$. In Figs. 4(c) and 4(d), we plot the real and imaginary parts of the contributions to ρ'_{41} at $z = 0$ [see Eqs. (15)]. We see that the contri-

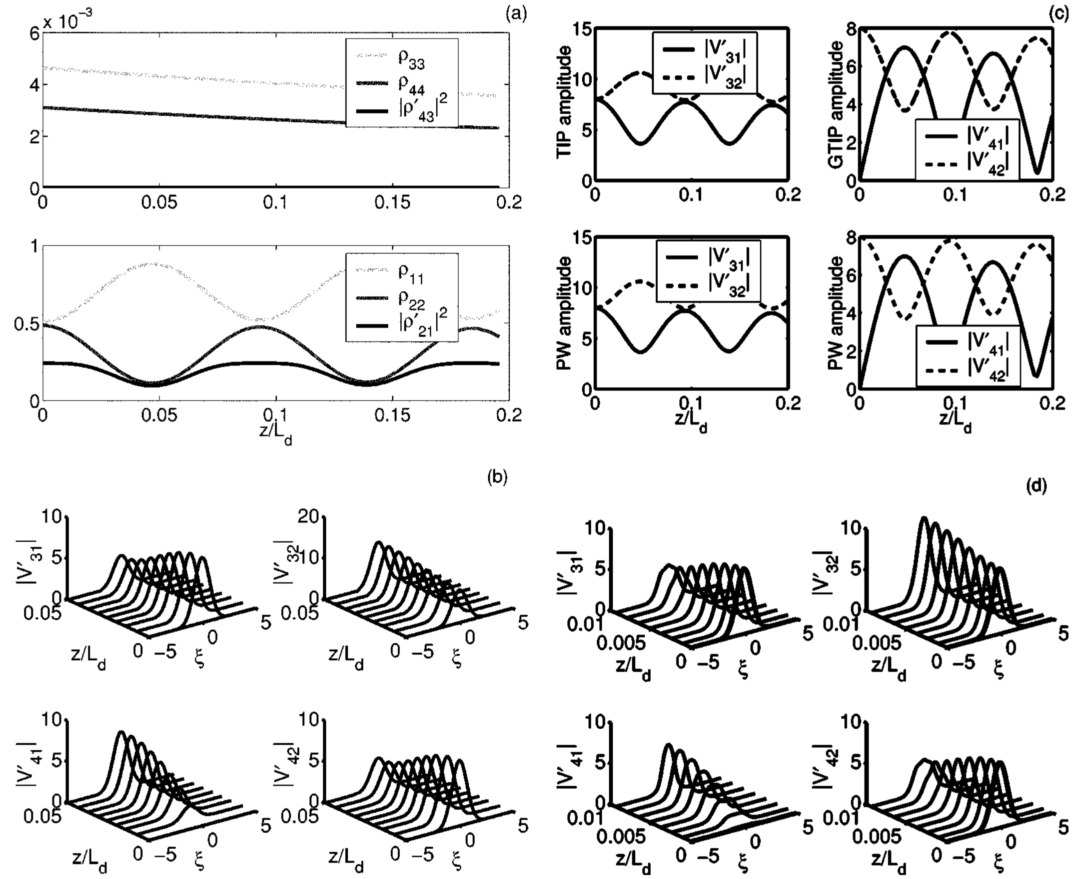

FIG. 3. Double-Λ system in which detuning of upper Λ system is varied. In (a), (b), and (c), $\Delta'_{41}=\Delta'_{42}=\pm 100$, and in (d), $\Delta'_{41}=\Delta'_{42}=\pm 10$. (a) Populations and two-photon coherences of Gaussian beams at $\xi=0$, as a function of z/L_D; (b) and (d) TIP's (V'_{ij} vs ξ) of propagating beams as a function of z/L_D; and (c) comparison between Rabi frequencies of Gaussian beams (at $\xi=0$) and PW beams as a function of z/L_D. Initial Rabi frequencies are $V'_{31}(0)=V'_{32}(0)=V'_{42}(0)=8$, and $V'_{41}(0)=0.001$. Detunings are $\Delta'_{31}=\Delta'_{32}=\pm 4$, and $L_{rel}=1.66\times 10^{-4}$.

bution from $\rho'^{(3)}_{41}$ is much greater than that from $\rho'^{(1)}_{41}$ which is to be expected for a field that is initially much weaker than the other fields. In addition, it can be seen that the contribution from FWM is greater off axis than on axis, leading to the formation of a ring.

2. Two strong fields: Strong-weak-strong-weak configuration

We now consider the strong-weak-strong-weak configuration which has been studied extensively by Babin *et al.* [20–23]. We solve the Maxwell-Bloch equations for the parameters $V'_{31}(0)=V'_{42}(0)=4$, $V'_{32}(0)=0.1$, $V'_{41}(0)=0.001$, $\Delta'_{ij}=-4$, and $L_{rel}=1.66\times 10^{-3}$. In Fig. 5(a), we compare the amplitudes of the PW's with those of the initially Gaussian beams on axis, and note the maximum conversion of $V'_{42}(0)$ to $V'_{41}(0)$ and $V'_{31}(0)$ to $V'_{32}(0)$ for the case of the Gaussian beams is approximately twice that of the PW beams (41% against 19%) at a length $z/L_D=0.17$. Thus focusing leads to much higher frequency conversion for this case. In addition, the fields $V'_{31}(0)$ and $V'_{42}(0)$ which are initially strong, undergo focusing on propagation which prevents the steep drop in intensity experienced by the PW beams. We note that when the beams are detuned to the red, $\Delta'_{ij}=4$, all the initially Gaussian beams become strongly defocused on propagation, so that there is almost no difference between the behavior of the initially Gaussian and the PW beams. In this case, CPT is not present initially but, in contrast to the previous case, is achieved on propagation at a length which is approximately equal to that at which maximum conversion occurs. At $z/L_D=0.17$, the deviation from CPT is -0.05 and $|\rho_{21}|^2=0.19$. In Fig. 5(b), we plot the TIP's of the blue-detuned initially Gaussian beams, as they propagate. As in the previous case, the TIP's do not retain their Gaussian shape but develop into a central focused beam surrounded by a weaker ring. If we follow the changes in V'_{32} and V'_{41} on propagation, we see that at small values of z/L_D, they each acquire two peaks in the wings which grow on propagation, and eventually form the ring around the central peak. The origin of the rings can again be explained by considering the behavior of $\rho'^{(1,3)}_{41}$ and $\rho'^{(1,3)}_{32}$ as a function of ξ at $z=0$.

In both the cases discussed in this section, ring formation occurs before focusing, at a distance which is either shorter than (case of three strong beams) or similar to (strong-weak-strong-weak configuration) that at which CPT is established. When propagation is continued beyond the distance where focusing occurs, the TIP's eventually regain their Gaussian behavior. In order to establish at which stage, in the propagation, azimuthal-symmetry breakup [50] will occur, it is

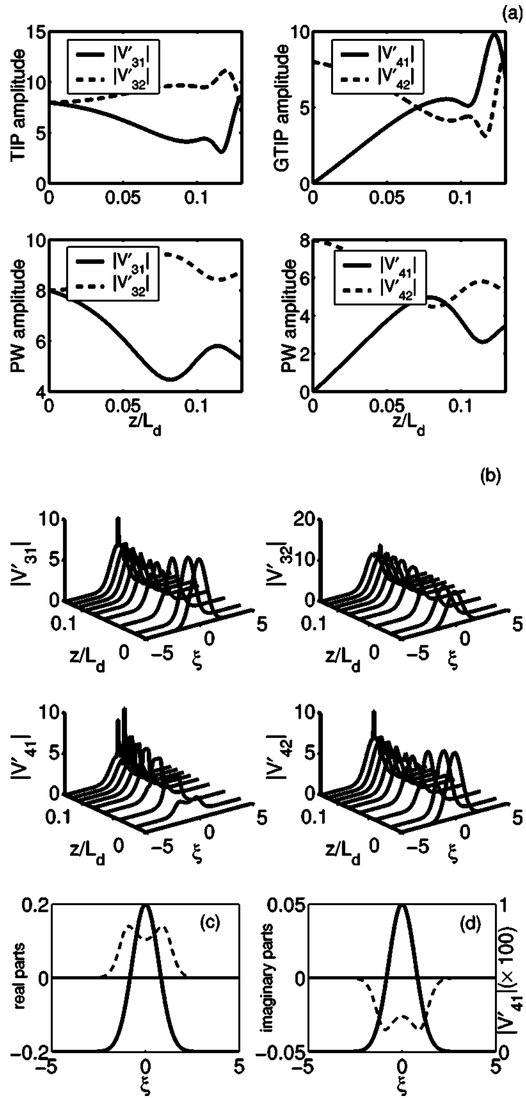

FIG. 4. Double-Λ system with three strong fields. (a) Comparison between Rabi frequencies of initially Gaussian beams (at $\xi=0$) and PW beams as a function of z/L_D; (b) TIP's (V'_{ij} vs ξ) of propagating beams as a function of z/L_D; (c) $\mathrm{Re}\rho'^{(1)}_{41}$ (thin solid line), $\mathrm{Re}\rho'^{(3)}_{41}$ (dashed line), and V'_{41} (thick solid line) as a function of ξ; (d) $\mathrm{Im}\rho'^{(1)}_{41}$ (thin solid line), $\mathrm{Im}\rho'^{(3)}_{41}$ (dashed line), and V'_{41} (thick solid line) as a function of ξ. Initial Rabi frequencies are $V'_{31}(0)=V'_{32}(0)=V'_{42}(0)=8$, and $V'_{41}(0)=0.001$. Detunings are $\Delta'_{ij}=-4$, and $L_{rel}=1.52\times10^{-3}$.

necessary to solve the Maxwell-Bloch equations in three dimensions, rather than the two dimensions studied here.

In the following section, we compare a case of perfect CPT with one where CPT cannot occur, and show that in the latter case, the beams achieve focusing without ring formation. We do this by considering the propagation of four beams which have the same Rabi frequencies and detunings from their respective transitions, for the case where $\Phi=0$. In this case CPT with maximal coherence is established from the outset [6]. We then switch the relative phase to $\Phi=\pi$ where CPT cannot occur.

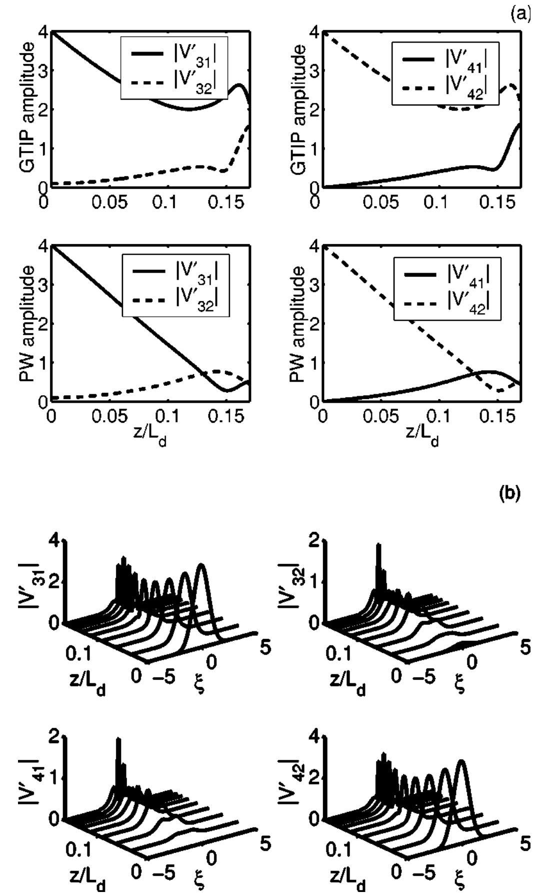

FIG. 5. Double-Λ system with two strong fields. (a) Comparison between Rabi frequencies of initially Gaussian beams (at $\xi=0$) and PW beams as a function of z/L_D; (b) TIP's (V'_{ij} vs ξ) of propagating beams as a function of z/L_D. Initial Rabi frequencies are $V'_{31}(0)=V'_{42}(0)=4$, $V'_{32}(0)=0.1$, and $V'_{41}(0)=0.001$. Detunings are $\Delta'_{ij}=-4$, and $L_{rel}=1.66\times10^{-3}$.

C. Role of the relative phase

In order to demonstrate the effect of switching the relative phase from $\Phi=0$ to $\Phi=\pi$, we solve the Maxwell-Bloch equations for the parameters $V_{ij}(0)=4$, $\Delta_{ij}=-4$, and $L_{rel}=1.11\times10^{-3}$. When the initial relative phase $\Phi=0$, perfect CPT with maximal two-photon coherence is obtained [6], and all the beams propagate without changing their shape up to $z/L_D\simeq0.22$. This distance is proportional to L_{rel} and can therefore be modified by changing L_{rel}. The fact that the beams propagate unchanged can be explained by considering the contributions to ρ'_{ij} [see Eqs. (15)]. In Figs. 6(a) and 6(b), we plot the real and imaginary parts of $\rho'^{(1)}_{41}$ and $\rho'^{(3)}_{41}$ at $z=0$, as a function of ξ. We see that $\rho'^{(1)}_{ij}$ and $\rho'^{(3)}_{ij}$ are of equal amplitude but opposite sign, so that their sum $\rho'_{ij}=0$, and the beams propagate unchanged apart from the effect of diffraction. However, when the phase is switched to $\Phi=\pi$, the two-photon coherence becomes zero [6] and CPT no longer holds. Furthermore, we see from Eqs. (16)–(19) that the sign of $\rho'^{(3)}_{ij}$ is switched so that the contributions to ρ'_{ij} are now equal in both magnitude and sign, as shown in Figs. 6(c) and 6(d). Consequently, they interfere constructively. From Figs. 6(c) and 6(d), we see that the incident and generated fields at

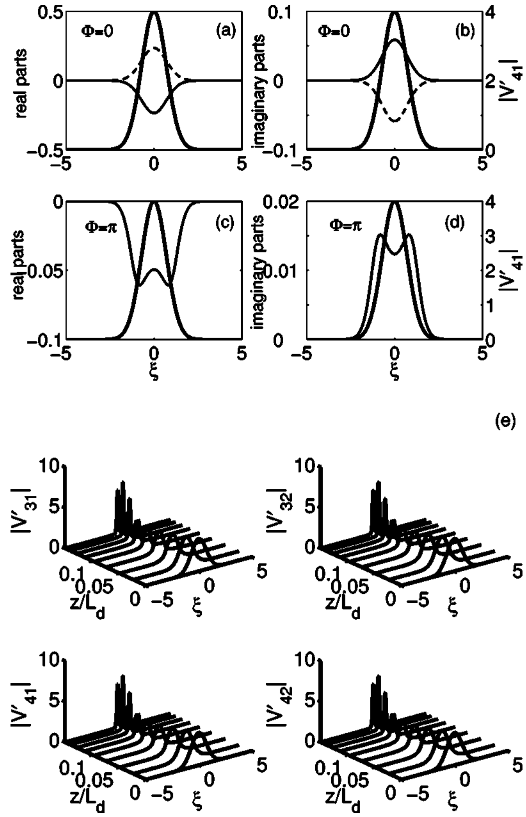

FIG. 6. Double-Λ system with $\Phi=0$ and $\Phi=\pi$. In (a) and (b), $\Phi=0$, and in (c), (d), and (e), $\Phi=\pi$. (a) and (c) $\mathrm{Re}\rho_{41}^{\prime(1)}$ (thin solid line), $\mathrm{Re}\rho_{41}^{\prime(3)}$ (dashed line), and V_{41}' (thick solid line) as a function of ξ; (b) and (d) $\mathrm{Im}\rho_{41}^{\prime(1)}$ (thin solid line), $\mathrm{Im}\rho_{41}^{\prime(3)}$ (dashed line), and V_{41}' (thick solid line) as a function of ξ; and (e) TIP's (V_{ij}' vs ξ) of propagating beams as a function of z/L_D for $\Phi=\pi$. Initial Rabi frequencies are $V_{ij}'(0)=4$, detunings are $\Delta_{ij}'=-4$, and $L_{rel}=1.11\times10^{-3}$.

ω_{ij} are absorbed more strongly at the wings than at the center, and are focused near the center as a result of the positive slope, and defocused at the wings due to negative slope. As a result of both these effects [53,54], we see in Fig. 6(e) that the amplitudes of the beams decrease at first and then gradually increase as the beams become more focused on propagation. After reaching a maximum, the focused beams then decay completely. The maximum amplitude is approximately twice the initial amplitude and there is no sign of ring formation. If the strength of the nonlinearity is too high, the decay is so rapid that focusing does not take place. In the PW approximation, the beams are rapidly absorbed on propagation.

It should be noted that as the relative phase Φ increases from zero, the length at which CPT is achieved increases. For the parameters of Fig. 6 and $\Phi\leqslant 3\pi/4$, the beams rapidly regain their Gaussian profiles when CPT is established and then propagate smoothly. As Φ increases further ($\Phi\to\pi$), focusing occurs before CPT is established, and is followed by breakup.

In discussing phase dependence in the double-Λ system, and indeed in any system which forms a loop, it is important to distinguish between cases where the field at the FWM frequency is initially absent (or very small) and those where it is present. In the former case, examples of which are discussed in Secs. III A and III B, the initial phase is irrelevant since, on propagation in the nonlinear medium, FWM is generated with the correct phase. Only when the FWM frequency is present at the outset, as in the case discussed in this section, is the initial phase important.

IV. CONCLUSIONS

In this article, we discuss the behavior of the TIP's of cw four copropagating beams interacting with a double-Λ system. We first study two well-known cases where CPT is established on the lower Λ system and then perturbed by the upper Λ system. In the first, the fields that interact with the upper Λ system are very weak, and in the second, one of them is stronger but detuned far from resonance. We show that there is considerable transfer of energy from the strong field to the weak field due to FWM and that it occurs at a very short propagation length. CPT exists in both these configurations from the outset. The two-photon coherence is constant at its maximum value in the former case, but oscillates with distance in the latter case. When the detuning is reduced considerably so that the initial deviation from CPT is large, we find that efficient frequency conversion takes place, without focusing or ring formation, at a distance which is an order of magnitude smaller than that at which CPT is established. This is contrary to conventional wisdom which holds that efficient conversion only takes place when CPT holds.

We also show that when the beams are blue detuned and the nonlinearity of the medium is reduced considerably, self-focusing of the beams leading to very high frequency conversion can be obtained, near the axis of propagation of the copropagating beams. This self-focusing is, however, accompanied by ring formation which may lead to breakup. Only by solving the Maxwell-Bloch equations in three dimensions, rather than in two dimensions as presented here, will it be possible to determine at what propagation length breakup will occur. In order to compare a system that has CPT at the outset with one that has no possibility of establishing CPT on propagation, we calculate the propagation of four beams with equal Rabi frequencies and detunings to the blue, when $\Phi=0$ and $\Phi=\pi$. When $\Phi=0$, the beams propagate unchanged for a length that is short compared to the diffraction length. However, when $\Phi=\pi$ and L_{NL} is sufficiently large, all four beams can be focused without ring formation.

The behavior of the transverse intensity profiles of the beams on propagation are explained by studying the effective linear and third-order contributions to the off-diagonal density-matrix elements, as a function of the beam profile.

ACKNOWLEDGMENT

We wish to thank Dr. Dafna Bortman-Arbiv for her help at the early stages of the research.

[1] S. P. Krinitzky and D. T. Pegg, Phys. Rev. A **33**, 403 (1986).
[2] S. J. Buckle, S. M. Barnett, P. L. Knight, M. A. Lauder, and D. T. Pegg, Opt. Acta **33**, 1129 (1986).
[3] D. Kosachiov, B. G. Matisov, and Y. V. Rozhdestvensky, Opt. Commun. **85**, 209 (1991).
[4] D. Kosachiov, B. G. Matisov, and Y. V. Rozhdestvensky, J. Phys. B **25**, 2473 (1992).
[5] W. Maichen, R. Gaggl, E. Korsunsky, and L. Windholz, Europhys. Lett. **31**, 189 (1995).
[6] E. A. Korsunsky and D. V. Kosachiov, Phys. Rev. A **60**, 4996 (1999).
[7] O. A. Kocharovskaya and P. Mandel, Phys. Rev. A **42**, 523 (1990).
[8] O. A. Kocharovskaya, G. Mauri, and E. Arimondo, Opt. Commun. **84**, 393 (1991).
[9] C. H. Keitel, O. A. Kocharovskaya, L. M. Narducci, M. O. Scully, S.-Y. Zhu, and H. M. Doss, Phys. Rev. A **48**, 3196 (1993).
[10] P. Doug, S. H. Tang, W. N. Man, and J. Y. Gao, J. Phys. B **34**, 2851 (2001).
[11] D. Kosachiov, B. G. Matisov, and Y. V. Rozhdestvensky, Europhys. Lett. **22**, 11 (1993).
[12] E. Cerboneschi and E. Arimondo, Phys. Rev. A **52**, R1823 (1995).
[13] M. D. Lukin, P. R. Hemmer, M. Loffler, and M. O. Scully, Phys. Rev. Lett. **81**, 2675 (1998).
[14] P. R. Hemmer, D. P. Katz, J. Donoghue, M. Cronin-Golomb, M. S. Shahriar, and P. Kumar, Opt. Lett. **20**, 982 (1995).
[15] T. T. Grove, M. S. Shahriar, P. R. Hemmer, P. Kumar, V. S. Sudarshanam, and M. Cronin-Golomb, Opt. Lett. **22**, 769 (1997).
[16] T. T. Grove, E. Rousseau, X.-W. Xia, D. S. Hsiung, and M. S. Shahriar, Opt. Lett. **22**, 1677 (1997).
[17] Y. F. Li, J. F. Sun, X. Y. Zhang, and Y. C. Wang, Opt. Commun. **202**, 97 (2002).
[18] B. L. Lu, W. H. Burkett, and M. Xiao, Opt. Lett. **23**, 804 (1998).
[19] S. Barreiro and J. W. R. Tabosa, Opt. Commun. **233**, 383 (2004).
[20] S. A. Babin, U. Hinze, E. Tiemann, and B. Wellegehausen, Opt. Lett. **21**, 1186 (1996).
[21] S. A. Babin, E. V. Podivilov, D. A. Shapiro, U. Hinze, E. Tiemann, and B. Wellegehausen, Phys. Rev. A **59**, 1355 (1999).
[22] U. Hinze, B. N. Chichkov, E. Tiemann, and B. Wellegehausen, J. Opt. Soc. Am. B **17**, 2001 (2000).
[23] S. A. Babin, S. I. Kablukov, U. Hinze, E. Tiemann, and B. Wellegehausen, Opt. Lett. **26**, 81 (2001).
[24] M. G. Payne and L. Deng, Phys. Rev. A **65**, 063806 (2002).
[25] J. H. Eberly, Philos. Trans. R. Soc. London, Ser. A **355**, 2387 (1997).
[26] E. A. Korsunsky, N. Leinfellner, A. Huss, S. Baluschev, and L. Windholz, Phys. Rev. A **59**, 2302 (1999).
[27] A. F. Huss, E. A. Korsunsky, and L. Windholz, J. Mod. Opt. **49**, 141 (2002).
[28] W. Maichen, F. Renzoni, I. Mazets, E. Korsunsky, and L. Windholz, Phys. Rev. A **53**, 3444 (1996).
[29] A. S. Zibrov, I. Novikova, and A. B. Matsko, Opt. Lett. **26**, 1311 (2001).
[30] A. Raczynski and J. Zaremba, Opt. Commun. **209**, 149 (2002).
[31] A. Raczynski, J. Zaremba, and S. Zielindka-Kaniasty, Phys. Rev. A **69**, 043801 (2004).
[32] I. Novikova, A. S. Zibrov, D. F. Phillips, A. Andre, and R. L. Walsworth, Phys. Rev. A **69**, 061802(R) (2004).
[33] G. Morigi, S. Franke-Arnold, and G.-L. Oppo, Phys. Rev. A **66**, 053409 (2002).
[34] E. A. Korsunsky and M. Fleischhauer, Phys. Rev. A **66**, 033808 (2002).
[35] F. L. Kien and K. Hakuta, Phys. Rev. A **69** 043811 (2004).
[36] M. T. Johnsson, E. Korsunsky, and M. Fleischhauer, Opt. Commun. **212**, 335 (2002).
[37] M. T. Johnsson and M. Fleischhauer, Phys. Rev. A **66**, 043808 (2002).
[38] M. T. Johnsson and M. Fleischhauer, Phys. Rev. A **67**, 061802(R) (2003).
[39] M. Jain, A. J. Merriam, A. Kasapi, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **75**, 4385 (1995).
[40] M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996).
[41] S. Harris, G. Y. Yin, M. Jain, H. Xia, and A. J. Merriam, Philos. Trans. R. Soc. London, Ser. A **355**, 2291 (1997).
[42] A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, Opt. Lett. **24**, 625 (1999).
[43] A. J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **84**, 5308 (2000).
[44] A. D. Wilson-Gordon, R. Klimovsky-Barid, and H. Friedmann, Phys. Rev. A **25**, 1580 (1982).
[45] L. J. Rothberg and N. Bloembergen, Phys. Rev. A **30**, 820 (1984).
[46] D. Bortman-Arbiv, A. D. Wilson-Gordon, and H. Friedmann, Phys. Rev. A **58**, R3403 (1998).
[47] D. Bortman-Arbiv, A. D. Wilson-Gordon, and H. Friedmann, Phys. Rev. A **61**, 033806 (2000).
[48] D. Bortman-Arbiv, A. D. Wilson-Gordon, and H. Friedmann, Phys. Rev. A **63**, 031801(R) (2001).
[49] D. Bortman-Arbiv, A. D. Wilson-Gordon, and H. Friedmann, Opt. Commun. **204**, 371 (2002).
[50] J. M. Soto-Crespo, E. M. Wright, and N. N. Akhmediev, Phys. Rev. A **45**, 3168 (1992).
[51] S. Boublil, A. D. Wilson-Gordon, and H. Friedmann, J. Mod. Opt. **38**, 1739 (1991).
[52] R. W. Boyd, *Nonlinear Optics*, 2nd ed. (Academic, San Diego, 2003).
[53] R. R. Moseley, S. Shepherd, D. J. Fulton, B. D. Sinclair, and M. H. Dunn, Phys. Rev. Lett. **74**, 670 (1995).
[54] R. R. Moseley, S. Shepherd, D. J. Fulton, B. D. Sinclair, and M. H. Dunn, Phys. Rev. A **53**, 408 (1995).