Enhanced frequency conversion of nonadiabatic resonant pulses in coherently prepared Λ systems

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We show that nonadiabatic, resonant amplitude- and phase-modulated pulses can be frequency converted with greater efficiency than adiabatic resonant pulses in a coherently prepared Λ system. Indeed, conversion efficiencies close to unity, similar to those achieved using highly detuned pulses, can been obtained by using highly nonadiabatic resonant pulses. Moreover, by solving the Maxwell-Bloch equations using Fourier transforms, we derive analytical expressions for the probe and the generated four-wave mixing (FWM) pulses as a function of time and propagation distance. From these expressions, which are valid for either adiabatic or nonadiabatic pulses, we derive the result that starting from a nonadiabatic probe pulse, an asymptotically matched probe-FWM pulse pair with the same shape as the initial probe pulse is obtained. In addition, we show that, starting with a nonadiabatic matched pulse pair or a pair of matched pulse trains, we obtain propagation of these pulses without either deformation or losses.

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I. INTRODUCTION

Coherent effects in the interaction between light and matter such as coherent population trapping (CPT) [1], electromagnetically-induced transparency (EIT) [2–4], and electromagnetically-induced absorption (EIA) [5–7] have spawned an enormous number of theoretical and experimental investigations, as well as many exciting applications in diverse fields such as "slow and fast light" [8,9], nonlinear optics [10], quantum information storage [9,11], frequency standards [12,13], and high-precision magnetometry [14]. In this paper, we discuss the interaction of *nonadiabatic* time-dependent fields with three-level Λ systems, shown in Fig. 1(a), which have been prepared in a coherent nonabsorbing superposition (dark state) of the lower levels. Such a medium has been called phaseonium by Scully [15,16] and Fleischhauer *et al.* [17], and can be prepared in several ways [18–20]. For convenience, we will use the four-level double Λ system, depicted in Figs. 1(b), where *cw* resonant fields interacting with the lower Λ system consisting of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$, prepare the system in such a superposition (see [21,22], and references therein). Once the coherent superposition is prepared, each leg of the upper Λ system consisting of the states $|1\rangle$, $|2\rangle$, and $|4\rangle$ interacts with a *nonadiabatic* time-dependent laser pulse which propagates in the atomic or solid-state medium.

Adiabatic or quasiadiabatic pulse propagation in simple Λ systems under EIT or CPT conditions, in double Λ systems under EIT conditions, and in coherently prepared Λ systems has been widely studied and a number of interesting phenomena identified (for a recent review, see [4]). Of particular interest, in the present context, is the phenomenon of *matched pulses*, first discovered by Harris [20,23], in which two pulses with initially identical time-dependent envelopes propagate at the vacuum speed of light, without changing their shape, in a coherently prepared Λ system. If the system is not coherently prepared but is initially in its ground state, the front edge of the pulses will coherently prepare the system [20], in a manner similar to stimulated Raman adiabatic passage (STIRAP, [18]), and the slightly deformed pulses will then propagate without further change in shape [24], and without losses. It has been shown in both the adiabatic [25] and quasiadiabatic [26] regimes that pulses with different initial time dependences, propagating in a coherently prepared medium, will eventually acquire the same shape [27]. Other form-stable pulse pairs, such as simultons which are analogous to self-induced transparency (SIT) in two-level systems [27–29], and adiabatons which have complementary pulse shapes [26,30], have also been identified.

If only the pulse with frequency ω_{41} and Rabi frequency V_{41} is initially present, it will be converted by four-wave mixing (FWM) into a pulse at the corresponding frequency ω_{42} with Rabi frequency V_{42} [see Fig. 1(b)]. In the adiabatic limit [25,31–35], for the case where $\omega_{41} \approx \omega_{42}$ and $\Gamma_{41} \approx \Gamma_{42}$, where Γ_{ij} is the transverse decay rate for the transition from state $|i\rangle$ to state $|j\rangle$, it can be shown (see Sec. II E,

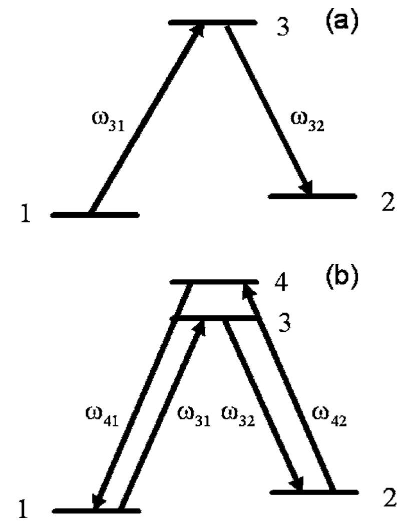

FIG. 1. Energy-level scheme for (a) single and (b) double Λ systems.

below) that the maximum conversion on propagation in the z direction is

$$C_{\max} = |V_{41}(t',z)/V_{42}(t',0)|_{\max} \simeq |\rho_{21}|, \tag{1}$$

when the initial pulse is resonant with the $|2\rangle$ to $|4\rangle$ transition at frequency ω'_{42}, that is, $\Delta_{42}=\omega'_{42}-\omega_{42}=0$, and the two-photon detuning $\Delta_{21}=\Delta_{31}-\Delta_{32}=\Delta_{41}-\Delta_{42}$ is zero. [We note that Eq. (1) and all subsequent time-dependent equations are written in the local frame where $t'=t-z/c$]. When, however, $\Delta_{42} \gg \Gamma_{42}$

$$C_{\max} = |V_{41}(t',z)/V_{42}(t',0)|_{\max} \simeq 2|\rho_{21}|. \tag{2}$$

It can be seen from Eqs. (1) and (2), that for maximum two-photon coherence $|\rho_{21}|=1/2$, $C_{\max}=1/2$ for *resonant* pulses whereas $C_{\max}=1$ for *far-detuned* pulses. This value of $|\rho_{21}|$ is achieved by applying cw beams with equal Rabi frequencies to the lower Λ system, and is maintained provided the Rabi frequencies applied to the upper Λ system are either much weaker than the cw beams, or initially matched such that $V_{41}(t',0)=V_{42}(t',0)$. However, the distance at which maximum conversion is achieved is longer for far-detuned pulses than for resonant pulses by a factor of approximately Δ_{42}/Γ_{42} so that the two-photon coherence has to be maintained at its maximum value for a far longer distance, which is not easy to achieve experimentally [33,35]. Here we show that it is possible to achieve high values of $C_{\max}$, *even at resonance*, for nonadiabatic, amplitude- and phase-modulated pulses, at distances that are shorter than those required for far-detuned adiabatic pulses. Moreover, we show that in the asymptotic limit, the probe pulse and the generated FWM signal exhibit matched pulse behavior. We also generalize the lossless and shape-conserving propagation of adiabatic matched pulses to the case of nonadiabatic matched pulses or pulse trains. It should be pointed out that Paspalakis and Kis [36] and Kis and Paspalakis [37] have recently proposed an alternative method of achieving unit conversion efficiency using resonant pulses in the adiabatic limit in a medium with position-dependent two-photon coherence [38,39].

Highly efficient frequency conversion has been observed experimentally by Jain *et al.* [31], Harris and coworkers [32], and Merrian *et al.* [33,34] in atomic Pb and by Hakuta *et al.* [40] in solid H_2. Recently, Bennink *et al.* [35] have reported experiments in a coherently prepared double Λ system in Na vapor, in which the incident optical field was amplitude and phase modulated. They found that the field generated at the FWM frequency, which was different from the initial frequency, had almost the identical amplitude and phase modulation as the incident field. This seems to be an example of "pulse matching"[23,27].

II. THE MODEL

A. The double Λ system

Let us first consider the double Λ system. Each $|j\rangle \to |i\rangle$ transition (with $j=1,2$ and $i=3,4$) interacts with an electromagnetic field

$$\vec{E}_{ij}(\vec{r},t) = (1/2)\hat{x}_{ij}E_{ij}(r)\exp[-i(\omega_{ij}t - k_{ij}z + \varphi_{ij})] + \text{c.c.}, \tag{3}$$

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 is Δ_{ij} and whose Rabi frequency is $2V_{ij}(r) = \mu_{ij}E_{ij}(r)/\hbar$.

The first step is to write the Bloch equations for the double Λ system [21,41] which reduce to those of the single Λ system [42] when $V_{4j}=0$. The Bloch equations are given by

$$\dot{\rho}_{11} = i(V_{13}\rho'_{31} + V_{14}\rho'_{41} - V_{31}\rho'_{13} - V_{41}\rho'_{14}) - \gamma_{12}\rho_{11} + \gamma_{21}\rho_{22} + \gamma_{31}\rho_{33} + \gamma_{41}\rho_{44}, \tag{4}$$

$$\dot{\rho}_{22} = i(V_{23}\rho'_{32} + V_{24}\rho'_{42} - V_{32}\rho'_{23} - V_{42}\rho'_{24}) + \gamma_{12}\rho_{11} - \gamma_{21}\rho_{22} + \gamma_{32}\rho_{33} + \gamma_{42}\rho_{44}, \tag{5}$$

$$\dot{\rho}_{33} = i(V_{31}\rho'_{13} + V_{32}\rho'_{23} - V_{13}\rho'_{31} - V_{23}\rho'_{32}) - \gamma_3\rho_{33} + \gamma_{43}\rho_{44}, \tag{6}$$

$$\dot{\rho}_{44} = i(V_{41}\rho'_{14} + V_{42}\rho'_{24} - V_{14}\rho'_{41} - V_{24}\rho'_{42}) - \gamma_4\rho_{44}, \tag{7}$$

$$\dot{\rho}'_{21} = i(V_{23}\rho'_{31} + aV_{24}\rho'_{41} - V_{31}\rho'_{23} - aV_{41}\rho'_{24}) - (\Gamma_{21} + i\Delta_{21})\rho'_{21}, \tag{8}$$

$$\dot{\rho}'_{31} = i(V_{31}\rho_{11} + V_{32}\rho'_{21} - V_{31}\rho_{33} - V_{41}\rho'_{34}) - (\Gamma_{31} + i\Delta_{31})\rho'_{31}, \tag{9}$$

$$\dot{\rho}'_{32} = i(V_{32}\rho_{22} + V_{31}\rho'_{12} - V_{32}\rho_{33} - a^*V_{42}\rho'_{34}) - (\Gamma_{32} + i\Delta_{32})\rho'_{32}, \tag{10}$$

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

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

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

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

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

and

$$\rho'_{21} = \rho_{21}\exp\{-i[(\Delta_{31}-\Delta_{32})t + (k_{31}-k_{32})z - (\varphi_{31}-\varphi_{32})]\}, \tag{15}$$

$$\rho'_{43} = \rho_{43} \exp\{-i[(\Delta_{41} - \Delta_{31})t + (k_{41} - k_{31})z - (\varphi_{41} - \varphi_{31})]\}. \tag{16}$$

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.

B. The coherently prepared Λ system

Let us consider the special case where resonant strong fields interact with the lower Λ system, preparing the system coherently. The resonant or detuned laser pulses then interact with the $|1,2\rangle \to |4\rangle$ transitions. We are interested in studying the propagation of laser pulses with initial Rabi frequencies $V_{41}(t,z=0)$ and $V_{42}(t,z=0)$. Assuming the populations ρ_{11}, ρ_{22}, and the Raman coherence ρ'_{21} to be unchanged on propagation, ρ_{33}, ρ_{44}, and ρ'_{43} to be negligible, and $\Phi = 0$, we find from Eqs. (11) and (12) that

$$\dot{\rho}'_{41} = i(V_{41}\rho_{11} + V_{42}\rho'_{21}) - (\Gamma_{41} + i\Delta_{41})\rho'_{41}, \tag{17}$$

$$\dot{\rho}'_{42} = i(V_{42}\rho_{22} + V_{41}\rho'_{12}) - (\Gamma_{42} + i\Delta_{42})\rho'_{42}, \tag{18}$$

where

$$\rho_{11} = \frac{|V_{32}|^2}{|V_{31}|^2 + |V_{32}|^2}, \tag{19}$$

$$\rho_{22} = \frac{|V_{31}|^2}{|V_{31}|^2 + |V_{32}|^2}, \tag{20}$$

$$\rho'_{21} = -\frac{V_{31}V_{32}^*}{|V_{31}|^2 + |V_{32}|^2}. \tag{21}$$

This is just one of several ways of clamping the Raman coherence ρ'_{21} to a particular value for a time that is long compared to the other lifetimes in the system [18,19]. Its maximum magnitude ($|\rho'_{21}| = \rho_{11} = \rho_{22} = 1/2$) is achieved when $|V_{31}| = |V_{32}|$. Such a medium has been called phaseonium by Scully [15,16] and Fleischhauer and coworkers [17] although its importance was noticed earlier in connection with coherent Raman spectroscopy [43,44].

C. Maxwell-Bloch equations for coherently prepared system

In order to study pulse propagation, we solve the Maxwell-Bloch equations, in the paraxial approximation, which may be written in the form [45]

$$\left(\frac{d}{dz} + \frac{1}{c}\frac{d}{dt}\right)\tilde{V}_{ij} = i\alpha^0_{ij}\rho'_{ij}, \tag{22}$$

where $j = 1,2$, $i = 3,4$, $\tilde{V}_{ij} = V_{ij}/\Gamma_{31}$ is the dimensionless Rabi frequency, and $\alpha^0_{ij} = \pi\omega_{ij}N\mu^2_{ij}/c\hbar\Gamma_{31}$ is four times the unsaturated line-center absorption coefficient for the $|j\rangle \to |4\rangle$ transition [46]. In our numerical work, we solve either the full Maxwell-Bloch equations [Eqs. (4)–(13) and Eq. (22)] for all four fields, or the restricted set of Maxwell-Bloch equations [Eqs. (17), (18), and (22)] for only the upper Λ system. The restricted set of Maxwell-Bloch equations (or the equivalent Maxwell-Schrodinger equations) have been solved analytically in the steady-state [31,33–35], adiabatic [25–27,47], or quasiadiabatic [26,47] approximation, in the bare-state [25,31,33–35] or dressed-state representation [26,27,47], with [25,27,31,33,34] or without [26,35,47] Doppler broadening. The case where the Raman coherence is position dependent has also been considered [36–39]. In addition to the analytical solutions to these equations, there have been a number of numerical studies in a variety of parameter regimes [48,49]. According to Eberly and Kozlov [27], this work has led to a consensus that the final pulses must be matched in their temporal shape [23]. In addition, the adiabatic approximation predicts that if the initial temporal shapes of the pulses are identical, they will remain so throughout the propagation [23,25].

D. Analytical solution

In order to obtain an analytical solution for the case where the pulses propagate either adiabatically or *nonadiabatically*, we take the Fourier transforms of Eqs. (17), (18), and (22), and obtain [23]

$$\frac{d}{dz}\hat{\tilde{V}}_{41} = -\hat{\alpha}_{41}\hat{\tilde{V}}_{41} + \hat{\kappa}_{41}\hat{\tilde{V}}_{42}, \tag{23}$$

$$\frac{d}{dz}\hat{\tilde{V}}_{42} = -\hat{\alpha}_{42}\hat{\tilde{V}}_{42} + \hat{\kappa}_{42}\hat{\tilde{V}}_{41}, \tag{24}$$

where $\tilde{x} = x/\Gamma_{31}$, $\hat{x}$ indicates the Fourier transform of x, and

$$\hat{\alpha}_{4j} = -i\alpha^0_{4j}\left(\frac{\rho_{jj}}{\tilde{\Delta}_{4j} - \tilde{\omega} - i\tilde{\Gamma}_{4j}}\right) - i\frac{\omega}{c}, \tag{25}$$

$$\hat{\kappa}_{41} = i\alpha^0_{41}\left(\frac{\rho'_{21}}{\hat{\tilde{\Delta}}_{41} - \tilde{\omega} - i\tilde{\Gamma}_{41}}\right), \tag{26}$$

$$\hat{\kappa}_{42} = i\alpha^0_{42}\left(\frac{\rho'_{12}}{\tilde{\Delta}_{42} - \tilde{\omega} - i\tilde{\Gamma}_{42}}\right), \tag{27}$$

and ω is the Fourier variable.

The solution to these ordinary differential equations is given by [46]

$$\hat{\tilde{V}}_{41}(z) = \frac{1}{\hat{g}_+ - \hat{g}_-}\{[\hat{\kappa}_{41}\hat{\tilde{V}}_{42}(0) - (\hat{g}_- + \hat{\alpha}_{41})\hat{\tilde{V}}_{41}(0)]\exp(\hat{g}_+z) - [\hat{\kappa}_{41}\hat{\tilde{V}}_{42}(0) - (\hat{g}_+ + \hat{\alpha}_{41})\hat{\tilde{V}}_{41}(0)]\exp(\hat{g}_-z)\}, \tag{28}$$

$$\hat{\tilde{V}}_{42}(z) = \frac{1}{\hat{g}_+ - \hat{g}_-}\{[\hat{\kappa}_{42}\hat{\tilde{V}}_{41}(0) - (\hat{g}_- + \hat{\alpha}_{42})\hat{\tilde{V}}_{42}(0)]\exp(\hat{g}_+z) - [\hat{\kappa}_{42}\hat{\tilde{V}}_{41}(0) - (\hat{g}_+ + \hat{\alpha}_{42})\hat{\tilde{V}}_{42}(0)]\exp(\hat{g}_-z)\}, \tag{29}$$

where

$$\hat{g}_{\pm} = -\frac{1}{2}(\hat{\alpha}_{41} + \hat{\alpha}_{42}) \pm \frac{1}{2}[(\hat{\alpha}_{41} - \hat{\alpha}_{42})^2 + 4\hat{\kappa}_{41}\hat{\kappa}_{42}]^{1/2}. \tag{30}$$

Assuming that that the two-photon coherence ρ'_{21} is determined solely by the lower Λ system so that $\rho_{11}\rho_{22}=|\rho'_{21}|^2$, and substituting Eqs. (25)–(27) and (30) in Eqs. (28) and (29), we find that

$$\hat{g}_{+} = i\omega/c, \tag{31}$$

$$\hat{g}_{-} = -(\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22}) + i\omega/c, \tag{32}$$

[or vice versa, since Eqs. (28) and (29) are symmetrical with respect to $g_{\pm}$], with

$$\bar{\alpha}^0_{4j} = \alpha^0_{4j}/(\tilde{\Gamma}_{4j} - i\tilde{\omega} + i\tilde{\Delta}_{4j}), \tag{33}$$

and

$$\hat{\tilde{V}}_{41}(z) = \frac{e^{i\omega z/c}}{\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22}}(-\hat{\tilde{V}}_{42}(0)\rho'_{21}\bar{\alpha}^0_{41}\{1 - \exp[-(\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22})z]\} + \hat{\tilde{V}}_{41}(0)\{\bar{\alpha}^0_{42}\rho_{22} + \bar{\alpha}^0_{41}\rho_{11}\exp[-(\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22})z]\}), \tag{34}$$

$$\hat{\tilde{V}}_{42}(z) = \frac{e^{i\omega z/c}}{\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22}}(-\hat{\tilde{V}}_{41}(0)\rho'_{12}\bar{\alpha}^0_{42}\{1 - \exp[-(\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22})z]\} + \hat{\tilde{V}}_{42}(0)\{\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22}\exp[-(\bar{\alpha}^0_{41}\rho_{11} + \bar{\alpha}^0_{42}\rho_{22})z]\}). \tag{35}$$

Equations (34) and (35) are crucial analytical results. On substituting Eqs. (19)–(21), we see that that a pair of pulses will propagate unchanged provided they are initially matched according to the expression

$$V_{42}(t',0)/V_{41}(t',0) = V_{32}/V_{31}. \tag{36}$$

In addition, we see that if only one of the pulses is initially present, the incident and generated pulses will be matched asymptotically according to

$$V_{42}(t',z)/V_{41}(t',z) \to V_{32}/V_{31}. \tag{37}$$

These results are well-known for adiabatic pulses [25,34] but here we see that they are also true for *nonadiabatic* pulses, even if the transition frequencies and dipole moments are unequal.

In order to simplify Eqs. (34) and (35), let us assume that $\tilde{\Delta}_{41}=\tilde{\Delta}_{42}=\tilde{\Delta}$, $\tilde{\Gamma}_{41}=\tilde{\Gamma}_{42}=\tilde{\Gamma}$, $\alpha^0_{41}=\alpha^0_{42}=\alpha^0$, and $\rho_{11}+\rho_{22}=1$. It can then be shown that

$$\hat{\tilde{V}}_{41}(z) = -\hat{\tilde{V}}_{42}(0)\rho'_{21}e^{i\omega z/c}\{1 - \exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta} - i\tilde{\omega})]\} + \hat{\tilde{V}}_{41}(0)e^{i\omega z/c}\{\rho_{22} + \rho_{11}\exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta} - i\tilde{\omega})]\} \tag{38}$$

$$\hat{\tilde{V}}_{42}(z) = -\hat{\tilde{V}}_{41}(0)\rho'_{12}e^{i\omega z/c}\{1 - \exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta} - i\tilde{\omega})]\} + \hat{\tilde{V}}_{42}(0)e^{i\omega z/c}\{\rho_{11} + \rho_{22}\exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta} - i\tilde{\omega})]\}. \tag{39}$$

E. The adiabatic approximation

The adiabatic approximation holds when $\tilde{\omega}$ can be neglected in the exponentials, either because the initial pulse is very long or because it is very detuned. In this approximation, inverting the Fourier transformations in Eqs. (38) and (39) leads to

$$\tilde{V}_{41}(t',z) = -\tilde{V}_{42}(t',0)\rho'_{21}\{1 - \exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta})]\} + \tilde{V}_{41}(t',0)\{\rho_{22} + \rho_{11}\exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta})]\}, \tag{40}$$

$$\tilde{V}_{42}(t',z) = -\tilde{V}_{41}(t',0)\rho'_{12}\{1 - \exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta})]\} + \tilde{V}_{42}(t',0)\{\rho_{11} + \rho_{22}\exp[-\alpha^0 z/(\tilde{\Gamma} + i\tilde{\Delta})]\}. \tag{41}$$

Let us consider two cases of frequency conversion from $\tilde{V}_{42}$ to $\tilde{V}_{41}$: large detuning and zero detuning. When the detuning $\tilde{\Delta} \gg \tilde{\Gamma}$, Eqs. (40) and (41) reduce to

$$\tilde{V}_{41}(t',z) = \tilde{V}_{42}(t',0)\rho'_{21}[\exp(-\alpha^0\tilde{\Gamma}z/\tilde{\Delta}^2)\exp(i\alpha^0 z/\tilde{\Delta}) - 1], \tag{42}$$

$$\tilde{V}_{42}(t',z) = \tilde{V}_{42}(t',0)[\rho_{22}\exp(-\alpha^0\tilde{\Gamma}z/\tilde{\Delta}^2)\exp(i\alpha^0 z/\tilde{\Delta}) + \rho_{11}], \tag{43}$$

so that for $\alpha^0 z \ll \tilde{\Delta}^2/\tilde{\Gamma}$,

$$|\tilde{V}_{41}(t',z)/\tilde{V}_{42}(t',0)| = 2|\rho'_{21}||\sin(\alpha^0 z/2\tilde{\Delta})|, \tag{44}$$

$$|\tilde{V}_{42}(t',z)/\tilde{V}_{42}(t',0)| = [1 - 4\rho_{11}\rho_{22}\sin^2(\alpha^0 z/2\tilde{\Delta})]^{1/2}. \tag{45}$$

It can be seen from Eqs. (42)–(45), that the Rabi frequencies oscillate [21,22,41], and that the maximum conversion

$$C_{\max} = |\tilde{V}_{41}(t',z)/\tilde{V}_{42}(t',0)|_{\max} \simeq 2|\rho_{21}|, \tag{46}$$

occurs on the first oscillation at a distance

$$\alpha^0 z_{\max} = \pi\tilde{\Delta}. \tag{47}$$

We now consider the case of zero detuning. For this case

$$\tilde{V}_{41}(t',z)/\tilde{V}_{42}(t',0) = -\rho_{21}[1 - \exp(-\alpha^0 z/\tilde{\Gamma})], \tag{48}$$

$$\tilde{V}_{42}(t',z)/\tilde{V}_{42}(t',0) = [\rho_{11} + \rho_{22}\exp(-\alpha^0 z/\tilde{\Gamma})], \tag{49}$$

so that

$$C_{\max} = |\tilde{V}_{41}(t',z)/\tilde{V}_{42}(t',0)|_{\max} \simeq |\rho_{21}|, \tag{50}$$

and $\alpha^0 z_{\max}$ is of the order of $\tilde{\Gamma}$ which is much shorter than in the case of a far detuned field [see Eq. (47)]. However, even when the Raman coherence is maximal, the maximum conversion is only 50%, as opposed to almost 100% in the case of large detuning.

As we will show in Sec. III, higher conversion can be achieved *at resonance* using nonadiabatic pulses. The conversion can be improved even more by using nonadiabatic pulse trains [50]. This occurs because for nonadiabatic pulses

$$V_{41}(t',z)/V_{42}(t',0) \to V_{31}/V_{32}, \tag{51}$$

at much shorter distances than those required for pulse matching.

F. The quasiadiabatic approximation

When the detuning is zero and there are small deviations from adiabaticity, it is possible to expand the exponentials in Eqs. (34) and (35) or in the simpler Eqs. (38) and (39) in a Taylor series in $\tilde{\omega}$ [51]. For example,

$$\exp[-\alpha^0 z/(\tilde{\Gamma} - i\tilde{\omega})] \simeq \exp[-\alpha^0 z/\tilde{\Gamma}]\{1 - i\alpha^0 z\tilde{\omega}/\tilde{\Gamma}^2 + (\alpha^0 z\tilde{\omega}^2/2i\tilde{\Gamma}^3)(2 - \alpha^0 z/\tilde{\Gamma}) + \cdots\}. \tag{52}$$

The inverse Fourier transforms can then be evaluated analytically term by term.

III. NUMERICAL RESULTS FOR NONADIABATIC GAUSSIAN AND SINUSOIDAL PULSES

We now present some numerical results for nonadiabatic pulses. We consider two cases of frequency conversion: one in which the incident short pulse is amplitude modulated (a Gaussian pulse), and one in which the incident pulse is both amplitude and phase modulated (a short sinusoidal pulse). The results reported here are obtained by solving numerically either the full or restricted set of Maxwell-Bloch equations for the double Λ system, interacting with two strong cw lasers with equal Rabi frequencies ($\tilde{V}_{31} = \tilde{V}_{32}$) that are resonant with the lower Λ system, and a pulsed field with either Gaussian or combined sinusoidal-Gaussian time dependence, interacting resonantly with the $|2\rangle \to |4\rangle$ transition. When solving the full set of Maxwell-Bloch equations, the fields interacting with the upper system are switched on only after the populations of the lower states and the Raman coherence achieve their steady-state values. Our calculations show that these values are not modified in either time or space by introducing the pulsed fields. For simplicity, we assume that $\tilde{\Gamma}_{4j} = \tilde{\Gamma}$, and $\alpha^0_{41} = \alpha^0_{42} = \alpha^0$.

The Gaussian time dependence is given by

$$\tilde{V}_{42}(t',0) = \tilde{V}_{42}(0,0)\exp[(t' - t'_0)^2/\tau^2], \tag{53}$$

and the sinusoidal time dependence is given by

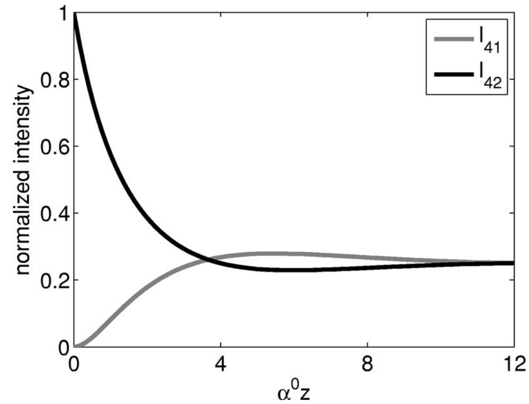

FIG. 2. Time-integrated relative intensities $I_{41,42}$ of the generated and converted pulses as a function of $\alpha^0 z$, for an incident, resonant Gaussian pulse of length $\Gamma_{31}\tau = 1$ and initial Rabi frequency $\tilde{V}_{42}(0,0) = 0.01$.

$$\tilde{V}_{42}(t',0) = \tilde{V}_{42}(0,0)\sin[2n\Gamma_{31}(t' - t'_0)],$$
$$0 \leq \Gamma_{31}(t' - t'_0) \leq \pi, \tag{54}$$

where the time t'_0 is some arbitrary time after the populations and coherence of the lower Λ system achieve the steady-state values given in Eqs. (19)–(21). In Eq. (53), $\Gamma_{31}\tau$ is the dimensionless pulse width and in Eq. (54), n is the number of cycles within the time interval of length $\Gamma_{31}t = \pi$. In order to avoid oscillations that derive from sudden turning on and off [52,53] of the sinusoidal pulse, we contain the sine pulse in a Gaussian envelope which would display adiabatic behavior if it were on its own.

In Figs. 2–4, we plot the relative integrated intensities

$$I_{4j} = \frac{\int_{-\infty}^{\infty} |\tilde{V}_{4j}(t',z)|^2 \, dt'}{\int_{-\infty}^{\infty} |\tilde{V}_{42}(t',0)|^2 \, dt'}, \tag{55}$$

of the pulses with frequencies ω_{4j}, as a function of the propagation distance $\alpha_0 z$, for incident resonant pulses. In Fig. 2,

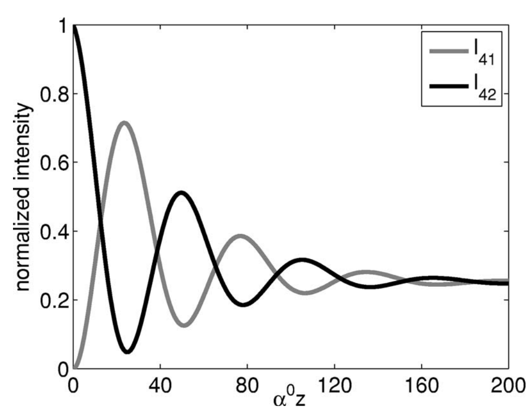

FIG. 3. Time-integrated relative intensities $I_{41,42}$ of the generated and converted pulses as a function of $\alpha^0 z$, for an incident four-cycle sinusoidal pulse of length $\Gamma_{31}t = \pi$ with a Gaussian envelope and initial Rabi frequency $\tilde{V}_{42}(0,0) = 0.01$.

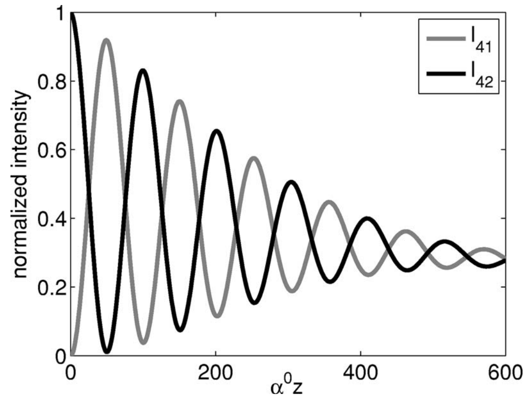

FIG. 4. Time-integrated relative intensities $I_{41,42}$ of the generated and converted pulses as a function of $\alpha^0 z$, for an incident, eight-cycle sinusoidal pulse of length $\Gamma_{31}t=\pi$ with a Gaussian envelope and initial Rabi frequency $\widetilde{V}_{42}(0,0)=0.01$.

the incident pulse is Gaussian with width $\Gamma_{31}\tau=1$; in Fig. 3, the incident pulse is a four-cycle sinusoidal pulse ($n=4$) with a Gaussian envelope; and in Fig. 4, the incident pulse is a eight-cycle sinusoidal pulse with a Gaussian envelope ($n=8$). We first note that, for these examples of nonadiabatic time dependence, the pulse intensities oscillate on propagation, even although they are at resonance with the upper Λ system. This is quite different from the adiabatic, resonant case [see Eqs. (48) and (49)] where the pulse intensities vary exponentially until pulse matching is achieved asymptotically. In addition, the maximum intensity conversion, achieved on the first oscillation, is greater than the 25% achievable in the adiabatic case. The maximum intensity conversion is even greater for the sinusoidal pulses and increases as the number of oscillations increases. However, the distance at which maximum conversion is achieved also increases.

In Figs. 5–7, we compare the temporal shapes of the pulse generated at the FWM frequency, and the pulse that has been depleted, with the original pulse, at the propagation distance where maximum intensity conversion occurs. The Gaussian case is shown in Fig. 5, the four-cycle pulse in Fig. 6, and the eight-cycle pulse in Fig. 7. The first point to note is the oscillatory behavior at the pulse tail. This is due to the nonadiabatic nature of the incident pulses [51]. At greater propagation distances, the oscillations are damped out until pulse matching is achieved. The oscillatory behavior can be reproduced for Gaussian pulses by expanding the exponentials in Eqs. (38) and (39) [see Eq. (52)] and performing the inverse Fourier transformation analytically for the first few terms in the expansion. Recently, Cheng *et al.* [54] have shown that the inclusion of these terms is essential if the noninstantaneous response of the nonlinear polarization is to be treated properly. The second point to note is the increasing resemblance of the generated pulse to the incident pulse, as we proceed from the Gaussian to the four-cycle to the eight-cycle pulses. This near-matching which leads to a high degree of conversion, occurs at distances which are much shorter than those required for full pulse matching. However, the distance at which maximum conversion occurs increases as the pulses become more nonadiabatic (compare Figs. 5–7). It should be emphasized that the results presented here

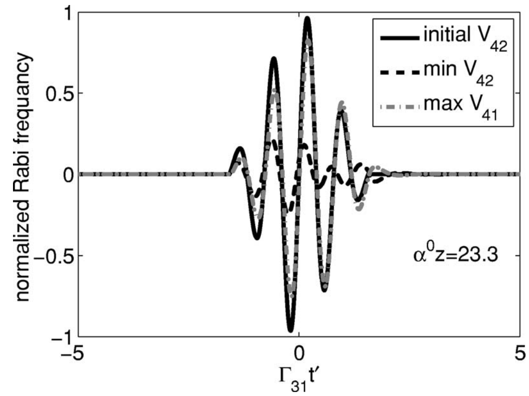

FIG. 6. Comparison of the initial four-cycle pulse amplitude $\widetilde{V}_{42}$ with the pulse amplitudes $\widetilde{V}_{41}$ and $\widetilde{V}_{42}$ at the value of $\alpha_0 z$ where the integrated intensity of $\widetilde{V}_{41}$ reaches its maximum value and $\widetilde{V}_{42}$ its minimum value. Parameters as in Fig. 3.

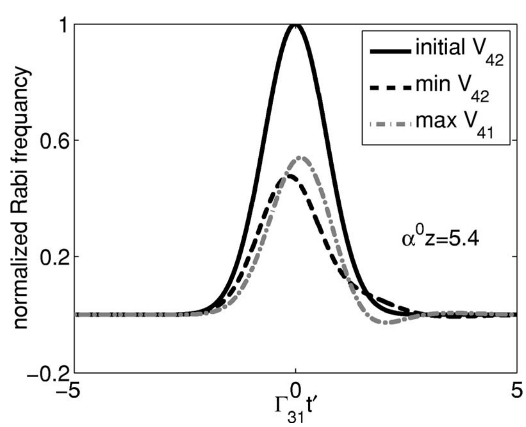

FIG. 5. Comparison of the initial Gaussian pulse amplitude $\widetilde{V}_{42}$ with the pulse amplitudes $\widetilde{V}_{41}$ and $\widetilde{V}_{42}$, at the value of $\alpha_0 z$ where the integrated intensity of $\widetilde{V}_{41}$ reaches its maximum value and $\widetilde{V}_{42}$ its minimum value. Parameters as in Fig. 2.

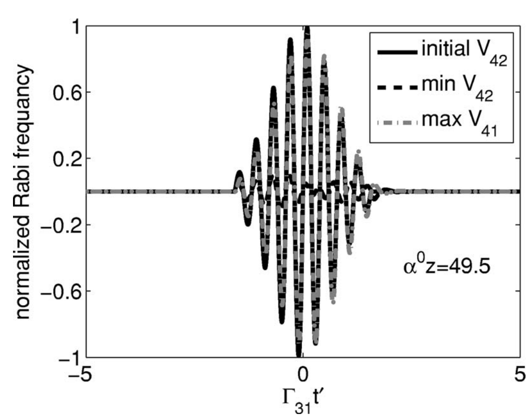

FIG. 7. Comparison of the initial eight-cycle pulse amplitude $\widetilde{V}_{42}$ with the pulse amplitudes $\widetilde{V}_{41}$ and $\widetilde{V}_{42}$ at the value of $\alpha_0 z$ where the integrated intensity of $\widetilde{V}_{41}$ reaches its maximum value and $\widetilde{V}_{42}$ its minimum value. Parameters as in Fig. 4.

for a weak probe pulse remain valid until $|V_{42}(0,0)|/|V_{3j}| \simeq 0.1$.

IV. CONCLUSIONS

We have shown that nonadiabatic, resonant amplitude- and phase-modulated pulses can be converted with greater efficiency than adiabatic resonant pulses in a coherently prepared Λ system (see Figs. 2–4). Indeed, conversion efficiencies close to unity can been obtained by using either strongly detuned probe pulses [see Eq. (46)] or highly nonadiabatic resonant ones. The reason for the higher efficiency is that in both cases, dissipation is limited by avoiding occupation of the decaying upper state due either to the large detuning of the nonresonant probe pulse, or to the short interaction time with the atom of the nonadiabatic probe pulse. For the same reason, the optical length required for the high conversion efficiency is much larger than that needed to achieve maximum efficiency with adiabatic pulses.

In this paper, we have compared adiabatic and nonadiabatic pulse propagation for the case where the Raman coherence ρ_{21} is constant in space and time. If the atomic coherence is position dependent, the conversion efficiency can be unity for adiabatic propagation at zero or small detunings [36,37]. Note that in our method, the Raman coherence is created by two cw laser beams, whereas the space-dependent atomic coherence is created using STIRAP [38,39].

Moreover, by solving the Maxwell-Bloch equations using Fourier transforms, we have derived analytical expressions for the probe and the generated FWM pulses as a function of time and propagation distance. From these generally valid expressions we derived the result that starting from a nonadiabatic probe pulse, an asymptotically matched probe-FWM pulse pair with the same shape as the initial probe pulse is obtained. In addition, we showed that, starting with a nonadiabatic matched pulse pair or a pair of matched pulse trains, we obtain propagation of these pulses without either deformation or losses.

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