# Enhancing nonlinear frequency conversion using spatially dependent coherence

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We consider pulse propagation in a $\Lambda$-type medium with spatially dependent coherence. It has been shown in previous works that it is impossible to get complete nonlinear energy conversion between an injected pulse and a generated pulse for homogenous coherence distribution. The aim of our work is to achieve unity conversion efficiency. We show by analytic considerations and numerical simulations that this can be achieved only if the propagation satisfies the conditions of adiabaticity in the local frame on the position domain. We also derive an exact analytic model for pulse propagation in our $\Lambda$-type medium, which is valid even if adiabaticity is not satisfied.

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## I. INTRODUCTION

The interaction of laser fields with the three-level $\Lambda$-type system has revealed several interesting phenomena, for example, electromagnetically induced transparency [1], coherent population trapping [2], stimulated Raman adiabatic passage [3], lasing without inversion [4], and many others. Besides their fundamental interest, the above phenomena give rise to several useful applications ranging from enhanced nonlinear optics, where one can obtain generation of radiation with high efficiency in regimes where conventional methods fail [5], to practical schemes for quantum computation. In the latter area the $\Lambda$-type system has been, for example, used for creating quantum bits and for storing and transporting quantum information [6].

A particular series of studies using the $\Lambda$-type system focuses on the properties of the so-called phaseonium system [7]. The prototype model of the phaseonium system is a $\Lambda$-type system that is initially prepared in a coherent superposition of the two lower levels. When laser pulses propagate in this system then several novel phenomena can occur, such as enhancement of the index of refraction [7], creation of matched pulses in optically thick media [8–14], and occurrence of high efficiency nonlinear pulse generation [15–17,19,18,20–23]. In the latter area it has been shown that quantum coherence and interference can make a material to have an active role in nonlinear optical processes [22], in contrast to the traditional case of nonlinear optics where a material has a rather passive role [24]. Some interesting experiments verifying some of the theoretical predictions have already been conducted [25–30], showing the potential for useful applications of the predicted phenomena.

Grobe and co-workers [31] introduced a phaseonium medium where the coefficients of the initial coherent superposition, which of course are the probability amplitudes, are dependent on space. This system was termed as a system with spatial excitation distribution. As it was shown, in certain regimes the spatial excitation distribution can be written and read by laser fields. A similar system of phaseonium with spatial excitation distribution was also studied by Kazinets *et al.* [32]. There, a new type of transparency was recognized, which combines the properties of electromagnetically induced transparency and self-induced transparency.

It can be shown, through a straightforward calculation [33], that if the amplitudes in the initial coherent superposition of the phaseonium are constant, then complete nonlinear conversion from an incoming pulse to a generated pulse is impossible in general. Using a simplified analysis of a phaseonium medium with spatial excitation distribution we have recently shown that nonlinear conversion between two laser pulses with unity conversion efficiency is possible [33]. In this paper, we continue study of this problem and present a thorough theoretical analysis of nonlinear frequency conversion in a $\Lambda$-type medium with spatially dependent coherence. Our analysis contains results in the adiabatic regime, exact analytical results, and finally numerical results. Our findings show that complete nonlinear frequency conversion in a specifically designed phaseonium medium is possible for a wide range of system parameters.

This paper is organized as follows. In the following section we present the main equations that govern the propagation dynamics of laser pulses in our system. We assume that the system is initially prepared in a spatially dependent coherent superposition state and that the laser-matter interaction is weak. Following the standard approximation methods we obtain an equation for the propagation of laser pulses in the medium. In Sec. III we present a detailed study of the adiabatic behavior of the propagation equation, with emphasis to the case of nonlinear frequency conversion. Two different situations are analyzed, the case in which the generalized propagation constants are equal and the case in which they are different. The findings from the adiabatic analysis are also verified via numerical simulations. Then, in Sec. IV we present an analytical solution of the propagation equation, for a specific spatial excitation, that is based on a model for two-state level-crossing problems. Finally, a summary of our findings is given in Sec. V.

## II. THEORETICAL MODEL AND EQUATIONS OF MOTION

The quantum system under study is displayed in Fig. 1. Denoting the excited level by $|0\rangle$ and the lower levels by $|1\rangle$, $|2\rangle$ and allowing each laser pulse to drive only one transition, the Hamiltonian of this system in the interaction picture and in the rotating wave and dipole approximations is

 

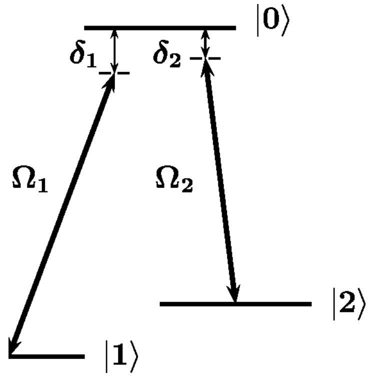


FIG. 1. Schematic diagram of the system studied.

given by (we use units such that $\hbar=1$)

$$\hat{H}(z,t)=\Omega_1(z,t)e^{-i\delta_1 t-ik_1z}|1\rangle\langle 0|+\Omega_2(z,t)e^{-i\delta_2 t-ik_2z}|2\rangle \times\langle 0|+\text{H.c.} \tag{2.1}$$

Here, $\Omega_n(z,t)=-\boldsymbol{\mu}_{n0}\cdot\boldsymbol{\varepsilon}_n\mathcal{E}_nf_n(z,t)$, with $n=1,2$, is the Rabi frequency of the transition $|n\rangle\leftrightarrow|0\rangle$, with $\boldsymbol{\mu}_{n0}$ being the associated dipole transition matrix element. Also, $\delta_n=\omega_0-\omega_n-\overline{\omega}_n$ is the laser field detuning from resonance for the transition $|n\rangle\leftrightarrow|0\rangle$, with the energies of the $n$th lower level and the upper level, respectively, being $\omega_n$ and $\omega_0$ and the angular frequency of the laser field being $\overline{\omega}_n$.

The laser field is described classically as a time-dependent and spatially dependent electric field,

$$\boldsymbol{E}(z,t)=\boldsymbol{\varepsilon}_1\mathcal{E}_1f_1(z,t)e^{i(\overline{\omega}_1t-k_1z)}+\boldsymbol{\varepsilon}_2\mathcal{E}_2f_2(z,t)e^{i(\overline{\omega}_2t-k_2z)}+\text{c.c.}, \tag{2.2}$$

where $k_n$, with $n=1,2$, is the wave number, $\boldsymbol{\varepsilon}_n$ the polarization vector, $\mathcal{E}_n$ the electric-field amplitude, and $f_n(z,t)$ the dimensionless pulse envelope of each laser pulse.

We analyze the dynamics of the system using a density matrix approach. From the Liouville equation of motion we obtain the following equations for the density-matrix elements

$$i\frac{\partial}{\partial t}\rho_{00}(z,t)=-i(\Gamma_{01}+\Gamma_{02})\rho_{00}(z,t)+\Omega_1^*(z,t)\rho_{10}(z,t)-\Omega_1(z,t)\rho_{01}(z,t)+\Omega_2^*(z,t)\rho_{20}(z,t)-\Omega_2(z,t)\rho_{02}(z,t), \tag{2.3a}$$

$$i\frac{\partial}{\partial t}\rho_{11}(z,t)=i\Gamma_{01}\rho_{00}(z,t)+i\Gamma_{21}\rho_{22}(z,t)+\Omega_1(z,t)\rho_{01}(z,t)-\Omega_1^*(z,t)\rho_{10}(z,t), \tag{2.3b}$$

$$i\frac{\partial}{\partial t}\rho_{22}(z,t)=i\Gamma_{02}\rho_{00}(z,t)-i\Gamma_{21}\rho_{22}(z,t)+\Omega_2(z,t)\rho_{02}(z,t)-\Omega_2^*(z,t)\rho_{20}(z,t), \tag{2.3c}$$

$$i\frac{\partial}{\partial t}\rho_{10}(z,t)=-(\delta_1+i\gamma_{10})\rho_{10}(z,t)+\Omega_1(z,t)\rho_{00}(z,t)-\Omega_1(z,t)\rho_{11}(z,t)-\Omega_2(z,t)\rho_{12}(z,t), \tag{2.3d}$$

$$i\frac{\partial}{\partial t}\rho_{20}(z,t)=-(\delta_2+i\gamma_{20})\rho_{20}(z,t)+\Omega_2(z,t)\rho_{00}(z,t)-\Omega_1(z,t)\rho_{21}(z,t)-\Omega_2(z,t)\rho_{22}(z,t), \tag{2.3e}$$

$$i\frac{\partial}{\partial t}\rho_{12}(z,t)=(\delta_2-\delta_1-i\gamma_{12})\rho_{12}(z,t)+\Omega_1(z,t)\rho_{02}(z,t)-\Omega_2^*(z,t)\rho_{10}(z,t), \tag{2.3f}$$

with $\Sigma_n\rho_{nn}(z,t)=1$ and $\rho_{nm}(z,t)=\rho_{mn}^*(z,t)$. We have assumed a closed system, i.e., there is no decay to levels outside of the three-level manifold we study. For differences in the propagation dynamics of closed and open three-level systems see Ref. [34]. We denote by $\Gamma_{nm}$ the radiative decay rate of the populations from level $|n\rangle$ to level $|m\rangle$ and by $\gamma_{nm}$ the coherence decay rate between states $|n\rangle$ and $|m\rangle$, with

$$\gamma_{nm}=\frac{1}{2}\sum_k\Gamma_{nk}+\frac{1}{2}\sum_l\Gamma_{ml}+\gamma'_{nm}, \tag{2.4}$$

where indices $k,l$ correspond to the states $|k\rangle$ and $|l\rangle$ in which states $|n\rangle$ and $|m\rangle$, respectively, decay to. Also, $\gamma'_{nm}$ describes the decay due to dephasing processes. Examples of dephasing processes include inelastic collisions in atomic and molecular systems or electron-electron scattering, interface roughness, and phonon scattering in semiconductor quantum well systems. The effects of Doppler broadening will not be considered in this paper.

To complete the set of equations for the study of propagation of short laser pulses in this medium, the Maxwell wave equation is required, which in the slowly varying envelope approximation reads

$$\left[\frac{\partial}{\partial z}f_n(z,t)+\frac{1}{c}\frac{\partial}{\partial t}f_n(z,t)\right]\boldsymbol{\varepsilon}_n\mathcal{E}_ne^{i(\overline{\omega}_nt-k_nz)}=-\frac{2i\pi\overline{\omega}_n}{c}\boldsymbol{P}_n(z,t),\quad n=1,2. \tag{2.5}$$

As Doppler broadening has been ignored, the negative frequency part of the macroscopic polarization of the medium, $\boldsymbol{P}_n(z,t)$, is given by

$$\boldsymbol{P}_n(z,t)=\mathcal{N}\boldsymbol{\mu}_{0n}\rho_{n0}(z,t)e^{i(\overline{\omega}_nt-k_nz)},\quad n=1,2, \tag{2.6}$$

where $\mathcal{N}$ is the density of the particles. Substituting Eq. (2.6) into Eq. (2.5) we obtain the following equations for the propagation of the Rabi frequencies:

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\Omega_1(z,t)=ia_1\rho_{10}(z,t), \tag{2.7a}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\Omega_2(z,t)=ia_2\rho_{20}(z,t). \tag{2.7b}$$

Here, $a_n=2\pi\mathcal{N}|\boldsymbol{\mu}_{n0}|^2\overline{\omega_n}/c$, with $n=1,2$, is the propagation constant. It is convenient to transform Eqs. (2.3) and (2.7) in the local frame where $\tau=t-z/c$ and $\zeta=z$. In this frame Eq. (2.3) will be the same with the substitution $t\to\tau$ and $z\to\zeta$, while Eq. (2.7) reads

$$\frac{\partial}{\partial\zeta}\Omega_1(\zeta,\tau)=ia_1\rho_{10}(\zeta,\tau), \tag{2.8a}$$

$$\frac{\partial}{\partial\zeta}\Omega_2(\zeta,\tau)=ia_2\rho_{20}(\zeta,\tau). \tag{2.8b}$$

Equations (2.3)—written in the local frame—and (2.8) govern the spatiotemporal evolution of the laser pulses in the medium.

We assume that the system is initially prepared in a superposition of the lower levels with spatially dependent coefficients, such that

$$|\psi(\zeta,\tau=\tau_i)\rangle=b_1(\zeta)|1\rangle+b_2(\zeta)|2\rangle, \tag{2.9}$$

with $b_1(\zeta)$ and $b_2(\zeta)$ being, in general, complex, satisfying $|b_1(\zeta)|^2+|b_2(\zeta)|^2=1$. As has been shown in Ref. [31], any superposition with spatially dependent coefficients of the form of Eq. (2.9) can be created with the use of stimulated Raman adiabatic passage [3]. According to this method two laser pulses are applied to the medium and their shape and delay determine the spatial dependence of the coherent superposition of Eq. (2.9). This has been shown through theoretical analysis and detailed numerical simulations [31]. We note here that the effect of the initially applied or preparation laser pulses are simply to prepare the medium in the superposition of Eq. (2.9). At time $\tau=\tau_i$ the preparation laser pulses have already created the superposition and do not interact with the medium anymore.

We also assume that the two-photon resonance condition $\delta_1=\delta_2=\delta$ is satisfied and that the coherence decay rate between the lower levels is negligibly small, i.e., $\gamma_{12}\approx 0$. This condition implies that the radiative decay rates $\Gamma_{nm}$ between the lower levels are practically zero, which is quite plausible. The constants $\gamma'_{12}$ describing the dephasing contributions to broadening could be kept small by controling the experimental conditions. If the excited state $|0\rangle$ decays rapidly and the laser-matter interaction is weak, so that the following relations $|\Omega_n|\ll\gamma$, $\gamma\overline{\tau}\gg 1$, and $|\Omega_n|^2\overline{\tau}\ll\gamma$ are satisfied, with $\overline{\tau}$ being a characteristic pulse length and $\gamma=\min(\gamma_{10},\gamma_{20})$, the approximate solutions of Eqs. (2.3d) and (2.3e) are

$$\rho_{10}(\zeta,\tau)\approx-\frac{\Omega_1(\zeta,\tau)|b_1(\zeta)|^2+\Omega_2(\zeta,\tau)b_1(\zeta)b_2^*(\zeta)}{\delta+i\gamma_{10}}, \tag{2.10a}$$

$$\rho_{20}(\zeta,\tau)\approx-\frac{\Omega_1(\zeta,\tau)b_1^*(\zeta)b_2(\zeta)+\Omega_2(\zeta,\tau)|b_2(\zeta)|^2}{\delta+i\gamma_{20}}, \tag{2.10b}$$

and the propagation equation for the Rabi frequencies, Eqs. (2.8a) and (2.8b), reduce to [10,23]

$$\frac{\partial}{\partial\zeta}\boldsymbol{\Omega}(\zeta,\tau)=-i\boldsymbol{K}(\zeta)\boldsymbol{\Omega}(\zeta,\tau), \tag{2.11}$$

with

$$\boldsymbol{K}(\zeta)=\begin{bmatrix}\alpha_1|b_1(\zeta)|^2 & \alpha_1 b_1(\zeta)b_2^*(\zeta)\\ \alpha_2 b_2(\zeta)b_1^*(\zeta) & \alpha_2|b_2(\zeta)|^2\end{bmatrix}. \tag{2.12}$$

Here, $\alpha_n=a_n/(\delta+i\gamma_{n0}/2)$ are the generalized propagation constants and the vector of the Rabi frequencies is given by $\boldsymbol{\Omega}(\zeta,\tau)=[\Omega_1(\zeta,\tau),\Omega_2(\zeta,\tau)]^T$.

We note that in the case in which the probability amplitudes $b_1(\zeta)$ and $b_2(\zeta)$ are arbitrary spatially dependent functions, there is no general analytic solution of the propagation equation (2.11). This equation resembles the time-dependent Schrödinger equation with the replacement $\zeta\leftrightarrow t$, where the propagator $\boldsymbol{K}(\zeta)$, Eq. (2.12), plays the role of the Hamiltonian and is non-Hermitian in this case. In the case of a time-dependent Hamiltonian general solutions can be obtained if the dynamics satisfies adiabaticity [35]. Therefore, it is useful to study the adiabatic evolution of the system [36].

## III. ADIABATIC TREATMENT

In this section we study the behavior of the propagation, Eq. (2.11) in the adiabatic limit. As we have already mentioned, the propagation matrix $\boldsymbol{K}(\zeta)$ in Eq. (2.12) is non-Hermitian. Nevertheless, we can introduce an adiabatic basis, which consists of the right-hand eigenvectors of $\boldsymbol{K}(\zeta)$. In general, these vectors are nonorthogonal, although we can use them without any additional difficulty. After transforming the propagation equation (2.11) to the adiabatic basis, we can follow the standard adiabatic approximation methods to study the evolution of the system.

### A. The case when $\alpha_1\neq\alpha_2$

In principle, the generalized propagation constants $\alpha_1$ and $\alpha_2$ are not equal. This is the case when, e.g., the transition frequencies $\omega_1$ and $\omega_2$ and the relevant matrix elements are unrelated. The right-hand eigensystem of the propagation matrix $\boldsymbol{K}(\zeta)$ in Eq. (2.12) reads

$$\lambda_1=0,\quad \boldsymbol{s}_0=\begin{bmatrix}-b_2(\zeta)^*\\ b_1(\zeta)^*\end{bmatrix}, \tag{3.1a}$$

$$\lambda_2=\overline{\alpha},\quad \boldsymbol{s}_{\overline{\alpha}}=\frac{1}{N}\begin{bmatrix}\alpha_1 b_1(\zeta)\\ \alpha_2 b_2(\zeta)\end{bmatrix}, \tag{3.1b}$$

where $\overline{\alpha}=\alpha_1|b_1|^2+\alpha_2|b_2|^2$ and the normalization for the second adiabatic state is $N=\sqrt{|\alpha_1 b_1|^2+|\alpha_2 b_2|^2}$. One of the eigenstates, $\lambda_1$, is zero, whereas the other one $\lambda_2$ is nonzero.

The eigenvalue $\lambda_2$ is complex due to the fact that the propagator $\boldsymbol{K}(\zeta)$ is non-Hermitian. For the same reason, the two adiabatic states are not orthogonal. Nevertheless, we can use them as basis vectors, but we should keep in mind that we work in a nonorthogonal basis.

We now transform the propagation equation (2.11) to the adiabatic basis that is defined by Eq. (3.1). The transformation is performed by the nonunitary matrix

$$\boldsymbol{V}(\zeta)=\begin{bmatrix} -b_2(\zeta)^* & \dfrac{1}{N}\alpha_1 b_1(\zeta) \\ b_1(\zeta)^* & \dfrac{1}{N}\alpha_2 b_2(\zeta) \end{bmatrix}. \tag{3.2}$$

The transformed propagation equation (2.11) reads

$$\frac{\partial}{\partial\zeta}\tilde{\boldsymbol{\Omega}}(\zeta,\tau)=-i\tilde{\boldsymbol{K}}(\zeta)\tilde{\boldsymbol{\Omega}}(\zeta,\tau), \tag{3.3}$$

where

$$\tilde{\boldsymbol{\Omega}}(\zeta,\tau)=\boldsymbol{V}(\zeta)^{-1}\boldsymbol{\Omega}(\zeta,\tau), \tag{3.4}$$

$$\tilde{\boldsymbol{K}}(\zeta)=-i\boldsymbol{V}(\zeta)^{-1}\frac{\partial}{\partial\zeta}\boldsymbol{V}(\zeta)+\boldsymbol{V}(\zeta)^{-1}\boldsymbol{K}(\zeta)\boldsymbol{V}(\zeta).$$

The matrix elements of the transformed propagator $\tilde{\boldsymbol{K}}$ are given by

$$\tilde{K}_{11}=-i\frac{\overline{\alpha}'}{2\overline{\alpha}}, \tag{3.5a}$$

$$\tilde{K}_{12}=i\frac{\alpha_1\alpha_2}{\overline{\alpha}N}(b_1'b_2-b_1b_2'), \tag{3.5b}$$

$$\tilde{K}_{21}=-i\frac{N}{\overline{\alpha}}(b_1^{*\prime}b_2^*-b_1^*b_2^{*\prime}), \tag{3.5c}$$

$$\tilde{K}_{22}=i\left(\frac{N'}{N}-\frac{\overline{\alpha}'}{2\,\overline{\alpha}}\right)+\overline{\alpha}, \tag{3.5d}$$

where, for convenience, we have introduced the shorthand notation $'\equiv d/d\zeta$. We notice that $\tilde{K}_{11}$ is nonzero even though the matrix element $(\boldsymbol{V}^{-1}\boldsymbol{K}\boldsymbol{V})_{11}$ is zero. The nonzero value results entirely from the diabatic correction, i.e., from the first term in the definition of $\tilde{\boldsymbol{K}}$ in Eq. (3.4).

To obtain the adiabatic limit the coupling between the adiabatic basis vectors should be negligible [35], therefore, we must have

$$|\tilde{K}_{12}|,|\tilde{K}_{21}|\ll|\tilde{K}_{11}-\tilde{K}_{22}|. \tag{3.6}$$

This requirement imposes certain conditions on the spatial dependence of the probability amplitudes $b_1(\zeta)$ and $b_2(\zeta)$: they must vary with position slowly enough so that their derivatives are so small that the adiabaticity condition is fulfilled.

In this model there are two possibilities to fulfill the adiabaticity condition, Eq. (3.6): the first one is the proper choice for the position dependence of the probability amplitudes $b_i(\zeta)$. We can simply say that the wider the transition region is, the smaller the derivatives of $b_i(\zeta)$ are. Certainly, it is assumed that $b_i(\zeta)$ is smooth. The second possibility is to vary the particle density $\mathcal{N}$ of the medium. It is easy to see that the propagator matrix elements $\tilde{K}_{12}$ and $\tilde{K}_{21}$ are independent of the density. On the other hand, the difference $\tilde{K}_{11}-\tilde{K}_{22}$ depends linearly on $\mathcal{N}$. Therefore, the adiabaticity conditions can be satisfied by increasing the particle density. However, the density cannot be increased arbitrarily: In the case of too large density the dephasing effects increase as well, which ruin the coherent superposition state, Eq. (2.9), of the medium. In the master equation (2.3) these sorts of decays are accounted for by the decay rates $\gamma'_{nm}$ defined in Eq. (2.4). In addition, if the particle density becomes high enough, then near dipole-dipole interactions (local-field effects) arise [37]. This leads to a modification of the dynamics of the laser-matter interaction and, in that case, Eqs. (2.3) and (2.8) are no longer adequate to describe the system.

In the adiabatic limit, the transformed Rabi frequencies $\tilde{\Omega}_l(\zeta,\tau)$ evolve according to

$$\tilde{\Omega}_0(\zeta,\tau)=\exp\left(-i\int_{\zeta_i}^{\zeta}\tilde{K}_{11}(\xi)d\xi\right)\tilde{\Omega}_0(\zeta_i,\tau), \tag{3.7a}$$

$$\tilde{\Omega}_{\overline{\alpha}}(\zeta,\tau)=\exp\left(-i\int_{\zeta_i}^{\zeta}\tilde{K}_{22}(\xi)d\xi\right)\tilde{\Omega}_{\overline{\alpha}}(\zeta_i,\tau). \tag{3.7b}$$

These equations are always valid in the adiabatic limit. We have not made any specific assumptions about the spatial dependence of the probability amplitudes $b_1(\zeta)$ and $b_2(\zeta)$, except that the adiabaticity conditions should be fulfilled. Let us consider the exponential factors in Eqs. (3.7a) and (3.7b). The first one yields

$$\exp\left(-i\int_{\zeta_i}^{\zeta}\tilde{K}_{11}(\xi)d\xi\right)=\sqrt{\frac{\overline{\alpha}_i}{\overline{\alpha}}} \tag{3.8}$$

and the second is

$$\exp\left(-i\int_{\zeta_i}^{\zeta}\tilde{K}_{22}(\xi)d\xi\right)=\frac{N}{N_i}\sqrt{\frac{\overline{\alpha}_i}{\overline{\alpha}}}\exp\left(-i\int_{\zeta_i}^{\zeta}\overline{\alpha}(\xi)d\xi\right), \tag{3.9}$$

where the subscript $i$ means that the quantity should be evaluated at the entry of the medium $\zeta_i$. In Eq. (3.9) the exponential factor on the right-hand side (rhs) describes attenuation, therefore, for a sufficiently long propagation distance the exponential factor in Eq. (3.7b) becomes zero. This means that the component of the field along the eigenvector $\boldsymbol{s}_\alpha$ vanishes. Here we see a clear advantage of adiabatic evolution: if adiabaticity prevails throughout the propagation, the injected pulse propagates without energy loss. If adiabaticity is violated, then part of the injected energy is absorbed by the medium.

In the adiabatic limit the solution of the propagation equation (2.11) is given by

$$\mathbf{\Omega}(\zeta,\tau)=\boldsymbol{W}(\zeta,\zeta_i)\mathbf{\Omega}(\zeta_i,\tau), \tag{3.10}$$

with

$$\boldsymbol{W}(\zeta,\zeta_i)=\boldsymbol{V}(\zeta)\begin{bmatrix} e^{-i\kappa_1(\zeta)} & 0 \\ 0 & e^{-i\kappa_2(\zeta)} \end{bmatrix}\boldsymbol{V}(\zeta_i)^{-1}, \tag{3.11}$$

where $\kappa_l(\zeta)=\int_{\zeta_i}^{\zeta}\widetilde{K}_{ll}(\xi)d\xi$, with $l=1,2$. As we have shown, for sufficiently long propagation distance $|\zeta-\zeta_i|$ the factor $\exp[-i\kappa_2(\zeta)]$ goes to zero. In this limit the transition matrix, Eq. (3.11), reads

$$\boldsymbol{W}(\zeta,\zeta_i) =\frac{e^{-i\kappa_1(\zeta)}}{\overline{\alpha_i}}\begin{bmatrix} \alpha_2 b_2(\zeta_i)b_2(\zeta)^* & -\alpha_1 b_1(\zeta_i)b_2(\zeta)^* \\ -\alpha_2 b_2(\zeta_i)b_1(\zeta)^* & \alpha_1 b_1(\zeta_i)b_1(\zeta)^* \end{bmatrix}. \tag{3.12}$$

Let us assume that at the entry of the medium the probability amplitudes take the values $b_1(\zeta_i)=0$ and $b_2(\zeta_i)=1$. We require that the occupations between the two ground states change completely in the course of the propagation, i.e., at the end of the medium $\zeta_f$ we have $|b_1(\zeta_f)|=1$ and $b_2(\zeta_f)=0$. For the field we choose such an initial condition so that it is decoupled from the system:

$$\Omega_1(\zeta_i,\tau)=\Omega_i(\tau),\quad \Omega_2(\zeta_i,\tau)=0. \tag{3.13}$$

By making use of the solution given by Eq. (3.10) with the transition matrix, Eq. (3.12), we find for $\mathbf{\Omega}(\zeta_f,\tau)$,

$$\Omega_1(\zeta_f,\tau)=0, \tag{3.14a}$$

$$\Omega_2(\zeta_f,\tau)=-\sqrt{\frac{\alpha_2}{\alpha_1}}e^{-i\arg b_1(\zeta_f)}\Omega_i(\tau). \tag{3.14b}$$

Now let us consider the energy conservation in this system: The total energy density of the two pulses is given by a bilinear form $\mathcal{P}(\zeta,\tau)$ defined as

$$\mathcal{P}(\zeta,\tau)=\mathbf{\Omega}^{\dagger}(\zeta,\tau)\boldsymbol{D}\mathbf{\Omega}(\zeta,\tau), \tag{3.15}$$

where $\boldsymbol{D}$ is a diagonal matrix with constant elements that fix the dimension of the energy density. The derivative of $\mathcal{P}$ with respect to $\zeta$ should vanish if the energy is conserved

$$\frac{\partial\mathcal{P}(\zeta,\tau)}{\partial\zeta}=i(\mathbf{\Omega}^{\dagger}\boldsymbol{K}^{\dagger}\boldsymbol{D}\Omega-\mathbf{\Omega}^{\dagger}\boldsymbol{D}\boldsymbol{K}\mathbf{\Omega}). \tag{3.16}$$

On the rhs the components of the vector $\mathbf{\Omega}$ are the Rabi frequencies at the position $\zeta$. If this vector belongs to the zero-eigenvalue subspace of the propagator $\boldsymbol{K}$, then both scalar products vanish. Therefore, the energy is conserved in our system if the evolution is adiabatic, and initially the vector $\mathbf{\Omega}_i$ was in the zero-eigenvalue subspace of the propagator. Adiabatic evolution implies that the transition matrix of the system is given by Eq. (3.11). If the previous conditions are not met, then the rhs of Eq. (3.16) is nonzero and the equation describes attenuation.

In the example that leads to Eq. (3.14) we satisfied the conditions of energy conservation, hence the result shows that complete nonlinear conversion between two laser pulses is possible in our model.

The process of nonlinear frequency conversion in our system is illustrated in Fig. 2, where the spatiotemporal evolution of the normalized intensities of the laser pulses is displayed. The results have been obtained from a numerical solution of Eq. (2.11). The initial spatial distributions are chosen as

$$b_1(\zeta)=\sqrt{\frac{1}{1+e^{-(\zeta-\zeta_0)/\zeta'}}}, \tag{3.17a}$$

$$b_2(\zeta)=\sqrt{\frac{1}{1+e^{(\zeta-\zeta_0)/\zeta'}}}, \tag{3.17b}$$

and the incoming pulse has a sin-squared shape. It is clear that the incoming laser pulse is completely converted to a new laser pulse. The accuracy of the approximations that lead to Eq. (2.11) has been assessed by comparing the numerical solution of Eqs. (2.3) and (2.8) with that of Eq. (2.11). These calculations verify the validity of Eq. (2.11) for describing the propagation of pulses in our system.

The validity of the adiabatic approximation method is demonstrated in Fig. 3. The agreement is very good between the numerical solution of Eq. (2.11) and the result obtained by using the analytic form of the transition matrix, Eq. (3.12), the maximum difference between the two results is about $5\times10^{-3}$, implying that the adiabatic approximation is valid for the chosen parameter set.

### B. The case when $\alpha_1=\alpha_2$

A physically interesting limit appears when $\alpha_1=\alpha_2=\alpha$. This is quite common, since the two ground states can be the magnetic sublevels of a state with $J=1$ and $M=\pm1$ and the excited state has $J=0$. Now the eigensystem of the propagation matrix $\boldsymbol{K}(\zeta)$ in Eq. (2.12) reads

$$\lambda_1=0,\quad \boldsymbol{s}_0=\begin{bmatrix} -b_2(\zeta)^* \\ b_1(\zeta)^* \end{bmatrix},$$

$$\lambda_2=\alpha,\quad \boldsymbol{s}_\alpha=\begin{bmatrix} b_1(\zeta) \\ b_2(\zeta) \end{bmatrix}. \tag{3.18}$$

The two eigenstates $\boldsymbol{s}_0$ and $\boldsymbol{s}_\alpha$ are orthogonal. We form a matrix $\boldsymbol{U}$ from these vectors that is unitary now,

$$\boldsymbol{U}(\zeta)=\begin{bmatrix} -b_2(\zeta)^* & b_1(\zeta) \\ b_1(\zeta)^* & b_2(\zeta) \end{bmatrix}. \tag{3.19}$$

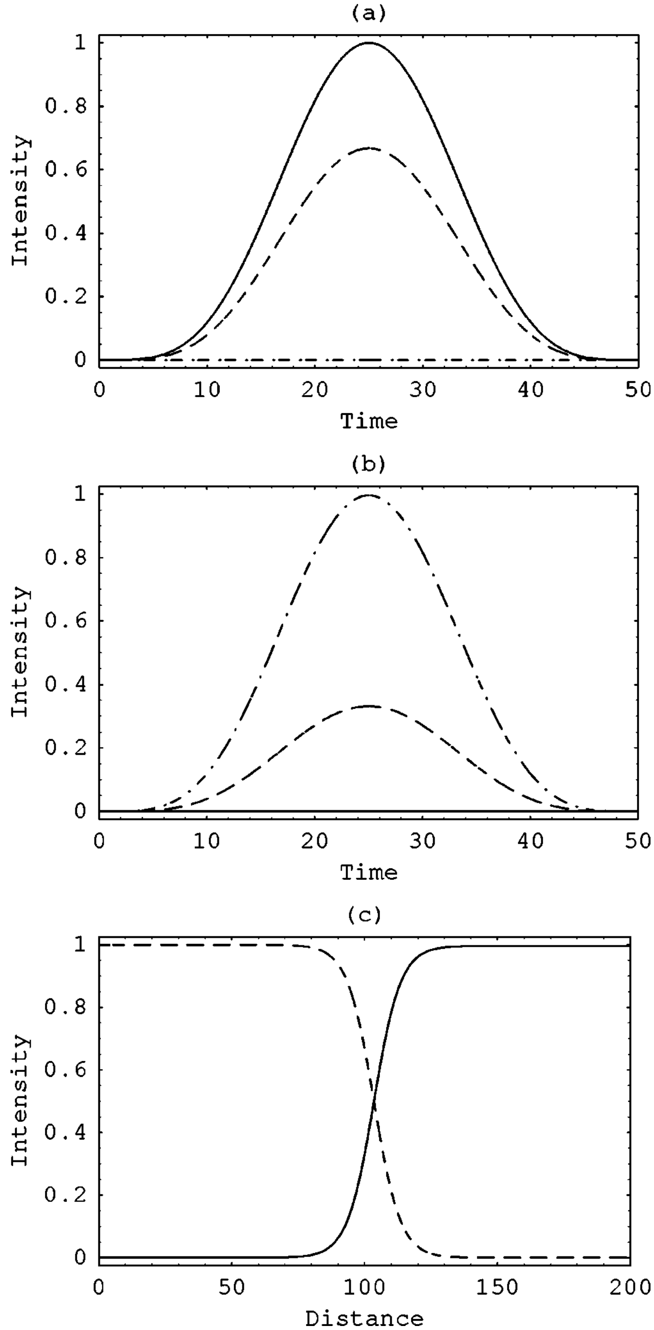


FIG. 2. Plots of $|\Omega_1(\zeta,\tau)|^2/|\Omega_1|^2$ for (a) and $|\alpha_1/\alpha_2||\Omega_2(\zeta,\tau)|^2/|\Omega_1|^2$ for (b) as a function of $\tau$ for different values of $\zeta$, with $\zeta=0$ (solid curves), $\zeta=100$ (dashed curves), and $\zeta=200$ (dot-dashed curves). In (c), we present the maximum of the normalized field intensities as a function of $\zeta$ for the incoming field (dashed curve) and the generated field (solid curve). The figures were obtained with the spatial distribution of Eq. (3.17) with $\zeta_0=100$ and $\overline{\zeta}=5$. The incident pulse is $\Omega(\tau)=\Omega_1\sin^2(\tau\pi/\tau_p)$, with $0\leqslant\tau\leqslant\tau_p$. The parameters used in the calculations are $a_1=1000$, $a_2=2000$, $\Omega_1=0.01$, $\tau_p=50$, $\delta=0$, $\gamma_{10}=\gamma_{20}=100$, $\zeta_0=100$, and $\overline{\zeta}=5$. All quantities are in arbitrary units.

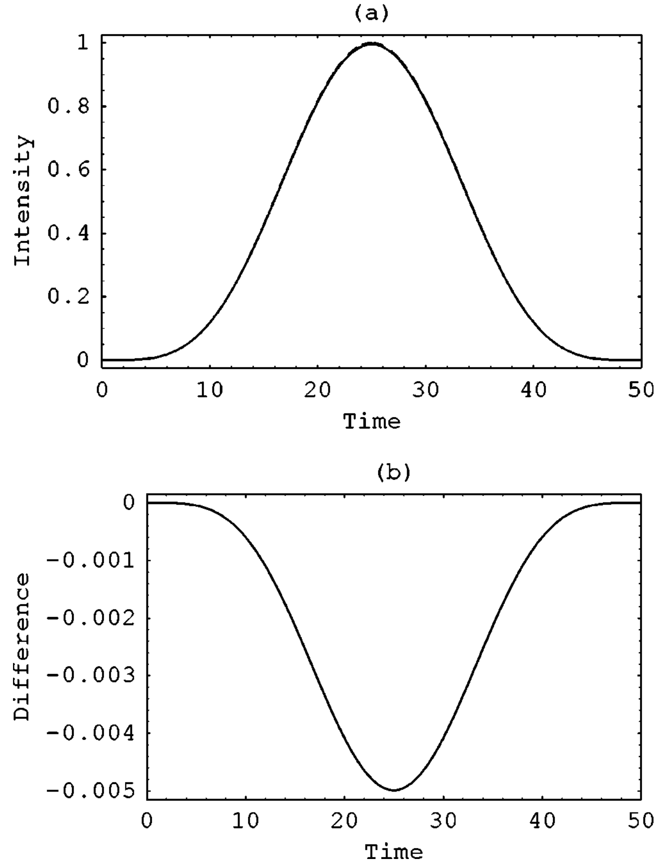


FIG. 3. In (a) we present numerical and analytical results for $|\alpha_1/\alpha_2||\Omega_2(\zeta,\tau)|^2/|\Omega_1|^2$ at position $\zeta=200$ in the medium. The numerical and analytical results are practically indistinguishable in the figure. We also plot the difference between the numerical and analytical results in (b). The medium and pulse parameters are as in Fig. 2. Similar agreement between the analytical and numerical results is obtained for $|\Omega_1(\zeta,\tau)|^2/|\Omega_1|^2$.

This matrix transforms the propagation equation (2.11) to the adiabatic basis, Eq. (3.18), according to Eq. (3.4). The transformed propagation matrix takes the form

$$\tilde{\boldsymbol{K}}(\zeta)=\begin{bmatrix} 0 & -i(b_1b_2'-b_1'b_2) \\ i(b_1^*b_2^{*\prime}-b_1^{*\prime}b_2^*) & \alpha \end{bmatrix}. \tag{3.20}$$

The eigenvalue $\alpha$ in the diagonal of $\tilde{\boldsymbol{K}}$ describes attenuation, therefore, the field component along the eigenvector $\boldsymbol{s}_\alpha$ vanishes for sufficiently long propagation distance. In the adiabatic limit the off-diagonal elements of $\tilde{\boldsymbol{K}}$ are negligible compared with the difference of the diagonal ones, such that

$$|b_1b_2'-b_1'b_2|\ll|\alpha|. \tag{3.21}$$

In this case the above condition should be fulfilled in order to have adiabatic evolution of the system.

If adiabaticity is fulfilled and we study the field in the long distance limit ($\zeta-\zeta_i\gg|\alpha|^{-1}$), the transition matrix reads

$$\boldsymbol{W}(\zeta,\zeta_i)=\boldsymbol{U}(\zeta)\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\boldsymbol{U}(\zeta_i)^{-1}$$
$$=\begin{bmatrix} b_2(\zeta_i)b_2(\zeta)^* & -b_1(\zeta_i)b_2(\zeta)^* \\ -b_2(\zeta_i)b_1(\zeta)^* & b_1(\zeta_i)b_1(\zeta)^* \end{bmatrix}. \tag{3.22}$$

For the same choice of the boundary conditions for the probability amplitudes $b_{1,2}(\zeta)$ and initial conditions for the field $\boldsymbol{\Omega}(\zeta_i,\tau)$ as in Sec. III A, we find that at the end of the medium the field is given by

$$\Omega_1(\zeta_f,\tau)=0, \tag{3.23a}$$

$$\Omega_2(\zeta_f,\tau)=-e^{-i\arg b_1(\zeta_f)}\Omega_i(\tau). \tag{3.23b}$$

The energy conservation is obvious in this case, hence, complete nonlinear conversion between the incoming and the outgoing laser pulses is possible. We note that we have verified numerically that the findings of this section are valid.

## IV. EXACT ANALYTIC TREATMENT

In the preceding section we considered adiabatic pulse propagation and we derived approximate analytic solutions for the propagation equation (2.11). For practical applications probably this is the most important case because the injected light pulse can be converted to a new pulse without energy loss. However, it is interesting to study that situation where the adiabaticity conditions are not satisfied. One possibility is to solve numerically the propagation equation (2.11). Another one is to look for some special choice for the probability amplitudes $b_1(\zeta)$ and $b_2(\zeta)$ for which exact analytic solution of the propagation equation can be obtained. In the following we will consider such an example.

Let us choose the probability amplitudes of the atomic coherent superposition state, Eq. (2.9), as

$$b_1(\zeta)=\sqrt{\frac{1}{2}\left(1+\tanh\frac{\zeta}{\Delta\zeta}\right)}e^{i\varphi_1},$$
$$b_2(\zeta)=\sqrt{\frac{1}{2}\left(1-\tanh\frac{\zeta}{\Delta\zeta}\right)}e^{i\varphi_2}. \tag{4.1}$$

The position $\zeta$ varies from $-\infty$ to $+\infty$. The limiting values $\pm\infty$ correspond to $|\zeta_{i,f}|\gg\Delta\zeta$ in practice. For the spatial dependence of Eq. (4.1) the propagation matrix, Eq. (2.12), becomes

$$\boldsymbol{K}(\zeta)=\frac{1}{2}\begin{bmatrix}\alpha_1\left(1+\tanh\frac{\zeta}{\Delta\zeta}\right) & \alpha_1 e^{i\varphi}\mathrm{sech}\frac{\zeta}{\Delta\zeta}\\ \alpha_2 e^{-i\varphi}\mathrm{sech}\frac{\zeta}{\Delta\zeta} & \alpha_2\left(1-\tanh\frac{\zeta}{\Delta\zeta}\right)\end{bmatrix}, \tag{4.2}$$

where $\varphi=\varphi_1-\varphi_2$. Inserting this propagator to the propagation equation (2.11) we obtain a system of differential equations that can be solved analytically.

Apart from some minor differences, the matrix $\boldsymbol{K}(\zeta)$ in Eq. (4.2) defines a famous level-crossing problem, first studied by Demkov and Kunike [38]. For equal and real $\alpha_1$, $\alpha_2$ the solution is well known [38–40]. However, to the best of our knowledge, the more general case in which $\alpha_1$, $\alpha_2$ are different and complex has not been studied yet. Therefore, we present here the solution for that case. We follow the derivation of Ref. [39], however, as the propagation constants $\alpha_l$ are complex in our case we have a non-Hermitian propagator. For brevity we introduce the notations

$$\overline{A_1}(\zeta)=\frac{\alpha_1}{2}e^{i\varphi}\mathrm{sech}\frac{\zeta}{\Delta\zeta}, \tag{4.3a}$$

$$\overline{A_2}(\zeta)=\frac{\alpha_2}{2}e^{-i\varphi}\mathrm{sech}\frac{\zeta}{\Delta\zeta}, \tag{4.3b}$$

$$\overline{B_1}(\zeta)=\frac{\alpha_1}{2}\left(1+\tanh\frac{\zeta}{\Delta\zeta}\right), \tag{4.3c}$$

$$\overline{B_2}(\zeta)=\frac{\alpha_2}{2}\left(1-\tanh\frac{\zeta}{\Delta\zeta}\right), \tag{4.3d}$$

The diagonal terms of $\boldsymbol{K}(\zeta)$ in Eq. (4.2) can be eliminated by the transformation

$$\boldsymbol{U}(\zeta)=\begin{bmatrix}\exp-iB_1(\zeta,\zeta_i) & 0\\ 0 & \exp-iB_2(\zeta,\zeta_i)\end{bmatrix}, \tag{4.4}$$

where $B_l(\zeta,\zeta_i)=\int_{\zeta_i}^{\zeta}\overline{B_l}(\xi)d\xi$. The propagation equation (2.11) is transformed by this transformation in the same way, as in Eq. (3.4). The transformed equation reads

$$\frac{\partial}{\partial\zeta}\tilde{\Omega}_1(\zeta,\tau)=-i\overline{A_1}(\zeta)e^{-iB(\zeta,\zeta_i)}\tilde{\Omega}_2(\zeta,\tau), \tag{4.5a}$$

$$\frac{\partial}{\partial\zeta}\tilde{\Omega}_2(\zeta,\tau)=-i\overline{A_2}(\zeta)e^{iB(\zeta,\zeta_i)}\tilde{\Omega}_1(\zeta,\tau), \tag{4.5b}$$

where $B(\zeta,\zeta_i)=B_2(\zeta,\zeta_i)-B_1(\zeta,\zeta_i)$. Then we eliminate $\tilde{\Omega}_2(\zeta,\tau)$ from Eq. (4.5a) and replace the variable $\zeta$ with

$$q=\frac{1}{2}\left(1+\tanh\frac{\zeta}{\Delta\zeta}\right). \tag{4.6}$$

The new variable $q$ varies from 0 to 1 as $\zeta$ goes from $-\infty$ to $\infty$. Finally we arrive at the hypergeometric differential equation

$$q(1-q)\frac{\partial^2\tilde{\Omega}_1}{\partial q^2}+[\nu-(\lambda+\mu+1)q]\frac{\partial\tilde{\Omega}_1}{\partial q}-\lambda\mu\tilde{\Omega}_1=0. \tag{4.7}$$

The constants $\lambda,\mu,\nu$ are defined as

$$\lambda=i\frac{\alpha_1\Delta\zeta}{2},\quad \mu=i\frac{\alpha_2\Delta\zeta}{2}, \tag{4.8}$$

$$\nu=\frac{1}{2}(1+i\alpha_2\Delta\zeta).$$

The general solution of Eq. (4.7) reads

$$\tilde{\Omega}_1 = aF(\lambda,\mu,\nu;q) + bq^{1-\nu}F(\lambda+1-\nu,\mu+1-\nu,2-\nu;q), \tag{4.9}$$

where the parameters $a$ and $b$ are determined from the initial conditions and $F(\lambda,\mu,\nu;q)$ is the hypergeometric function. Note that $a$ and $b$ may depend on $\tau$ in general.

The solution for the other field $\tilde{\Omega}_2$ can be obtained by using Eqs. (4.5a) and (4.9)

$$\tilde{\Omega}_2 = \frac{2ie^{i(B-\varphi)}\sqrt{q(1-q)}}{\alpha_1 \Delta\zeta}\bigg[ a\frac{\lambda\mu}{\nu}F(\lambda+1,\mu+1,\nu+1;q) + b\,(1-\nu)z^{-\nu}F(\lambda+1-\nu,\mu+1-\nu,1-\nu;q)\bigg]. \tag{4.10}$$

The solution of the original propagation equation (2.11) is given by

$$\mathbf{\Omega} = \boldsymbol{U}\tilde{\mathbf{\Omega}}, \tag{4.11}$$

where $\tilde{\mathbf{\Omega}} = [\tilde{\Omega}_1, \tilde{\Omega}_2]^T$.

We choose the initial condition to be given by Eq. (3.13), as this field is decoupled from the quantum system at the initial position $\zeta_i$. Therefore, according to Eqs. (4.9)–(4.11), $a=\Omega_i(\tau)$ and $b=0$ because $F(\lambda,\mu,\nu;0)=1$. The solution then reads

$$\Omega_1(\zeta,\tau) = \Omega_i(\tau)e^{-iB_1}F(\lambda,\mu,\nu;q), \tag{4.12a}$$

$$\Omega_2(\zeta,\tau) = \Omega_i(\tau)\frac{2\,i\,e^{-i(B_1+\varphi)}\sqrt{q(1-q)}}{\alpha_1\Delta\zeta}\,\frac{\lambda\mu}{\nu} \times F(\lambda+1,\mu+1,\nu+1;q). \tag{4.12b}$$

As an example we compare the results of the analytic solution of Eq. (4.12) with those obtained from the numerical solution of Eqs. (2.3) and (2.8) and the adiabatic solution of Sec. III A. The agreement between the three results is very good as can be seen in Fig. 4. In detail, numerical and analytical solutions are practically indistinguishable in Fig. 4(a), whereas the adiabatic results give a good approximate solution to the system. For clarity we also plot the difference between the numerical and analytical or adiabatic results in Fig. 4(b).

Finally, we demonstrate that the exact analytic solution, Eq. (4.12), describes the dynamics correctly even if we are out of the adiabatic limit. We take equal propagation constants $\alpha_1=\alpha_2$ for the two modes. Then, for the probability amplitudes in Eq. (4.1) the adiabaticity condition, Eq. (3.21), yields

$$\frac{1}{\Delta\zeta} \ll |\alpha|. \tag{4.13}$$

We choose $\Delta\zeta = |\alpha|^{-1}$ which clearly violates the previous inequality. We compared the numerical solution of Eqs. (2.3) and (2.8) with the analytic solution, Eq. (4.12), and with the approximate adiabatic solution, Eq. (3.22). We have found that according to our expectations, the exact analytic and the numerical solutions agree very well, whereas the adiabatic

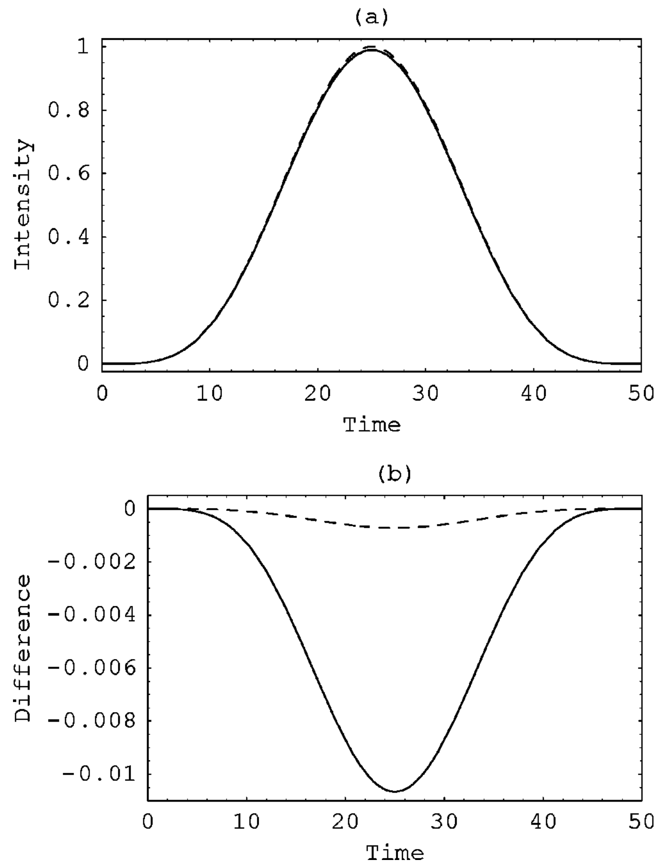


FIG. 4. Numerical (solid curve), analytical (dash-dotted curve), and adiabatic (dashed curve) for $|\alpha_1/\alpha_2||\Omega_2(\zeta,\tau)|^2/|\Omega_1|^2$ at position $\zeta=200$ in the medium are presented in (a), using the spatial distribution of Eq. (4.1) with $\Delta\zeta=5$. In (b) we plot the difference between the numerical and the analytical results (dashed curve) and the difference between the numerical and the adiabatic results (solid curve). Rest of the medium and pulse parameters are as in Fig. 2. Similar agreement is obtained for $|\Omega_1(\zeta,\tau)|^2/|\Omega_1|^2$.

approximation yields a significantly different result. We conclude that the exact analytic solution describes correctly the pulse-propagation dynamics as long as the conditions that led to the propagation equation (2.11) are satisfied.

## V. SUMMARY

In this paper we have presented a thorough theoretical analysis of nonlinear frequency conversion in a $\Lambda$-type medium with spatially dependent coherence. The medium was a phaseonium system, in which the two lower states were prepared initially in a coherent superposition state. The third excited state was empty initially. We have studied pulse propagation in this system assuming that the coherent superposition state of the medium is position dependent.

In the first part of the paper we have considered adiabatic pulse propagation: here adiabaticity is considered in the local frame on the position domain. Starting from a linear propagation equation for the pulses we have derived the conditions for adiabatic propagation. We have found explicit expressions for the matrix elements of the propagator that should be fulfilled. However, since the propagator is formed of the position-dependent coefficients of the atomic superposition state and the propagation constants of the medium, the adiabaticity conditions pose restrictions on these quantities. We have analyzed in detail two different cases: (a) the propagation constants are not equal for the two modes; and (b) the propagation constants are equal. We have shown that in both cases the efficiency of energy transfer between an injected pulse and the generated pulse is unity. Moreover, unit transfer efficiency can be achieved only if the propagation satisfies the conditions of adiabaticity in the local frame on the position domain. We have tested the results by comparing the approximate adiabatic solutions with numerical ones. We have found very good agreement for the chosen parameter sets.

In the second part of the paper we have worked out an exact analytic solution for pulse propagation in our system. For a special choice of the coefficients of the atomic superposition state we have derived an exact analytic solution of the propagation equation. We have realized that this special model is closely related to the Demkov-Kunike model of driven two-level systems. We have tested the exact analytic solution versus numerical simulations in cases when adiabaticity is satisfied or it is not satisfied, and have verified that the analytic solution is correct. Therefore, it can be used to study further pulse-propagation effects in a $\Lambda$-type medium with spatially dependent coherence.

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