Propagation of laser pulses and coherent population transfer in dissipative three-level systems: An adiabatic dressed-state picture

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The interaction of a pair of copropagating pulses with three-level Λ-type atoms is discussed in terms of time-dependent coupled and decoupled superpositions $|\pm\rangle$ of the lower levels. Due to the explicit time dependence of these states there is a nonadiabatic coupling between the ''bright'' state $|+\rangle$ and the ''dark'' state $|-\rangle$ in addition to the strong coupling between $|+\rangle$ and the upper level $|a\rangle$. We show that under quasiadiabatic conditions and in the presence of decay from the upper level this coupling can be treated perturbatively and the Maxwell-Bloch equations can be solved analytically. With the help of such a perturbation approach, coherent population transfer and formstable laser pulse propagation are studied. [S1050-2947(96)06206-3]

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I. INTRODUCTION

The resonant interaction of time-dependent fields with three-level Λ-type atoms has attracted some attention in recent years. There are two aspects of the problem for which interesting effects have been predicted and observed. One is the coherent dynamical evolution of the atomic system and the other one is the loss-free propagation of fields.

For example, employing a so-called counterintuitive sequence of overlapping laser pulses [1], it is possible to transfer population between the lower levels in a very fast and effective way. Since the upper level remains virtually unpopulated throughout the process, decay out of this state does not affect the transfer. This method of stimulated Raman adiabatic passage (STIRAP) was first observed by Bergmann and co-workers [2]. Since then this phenomenon has been studied intensively both experimentally [3] and theoretically [4].

The mechanism of population transfer is understood as an adiabatic following of one instantaneous eigenstate of the Hamiltonian in the time-dependent fields. The nonadiabaticity of the process is described by a small parameter ϵ. A measure for the success of the transfer are the nonadiabatic losses that are, therefore, of particular theoretical interest. For special pulse shapes, exact analytic solutions of the Bloch equations have been found by Carrol and Hioe [5] and very recently by Laine and Stenholm [6]. The generalization to arbitrary pulse shapes is, however, not simple. It is well known from the theory of adiabatic processes in two-state systems [7] that the asymptotic nonadiabatic losses may be exponentially small in $1/\epsilon$ [$\sim\exp(-1/\epsilon)$]. For this reason a perturbation in ϵ fails to describe the asymptotic behavior. A method that can be applied to a broad class of smooth pulses was recently proposed by Elk [8]. It is based on the superadiabatic basis technique for two-state systems introduced by Berry [9].

In all previous studies, decay from the upper level was disregarded. In the present paper we show that a simpler description of the population transfer is possible when this decay is taken into account. For this we employ a picture which uses the adiabatic coupled (''bright'') and decoupled (''dark'') superpositions of the lower levels as a basis. In this basis there is a strong coupling between the bright state and the upper level with the total Rabi frequency Ω and a weak coupling between the bright and dark state characterized by a (formal) Rabi frequency Ω_- that is due to nonadiabatic corrections. We derive a simple analytical expression for the nonadiabatic losses in the limit $\epsilon\to 0$ when the ratio of the upper level decay rate γ to the total Rabi frequency Ω is fixed. This expression can be expanded in a Taylor series in ϵ showing that in the presence of decay the nonadiabatic losses are *not* exponentially small in $1/\epsilon$. Hence a perturbation approach can be used to describe the population transfer for finite ϵ. The nonadiabatic losses are calculated for finite values of ϵ and the influence of the pulse form and upper level decay is discussed.

Another interesting aspect of the interaction of time-dependent fields with Λ-type atoms is the quasiloss-free propagation of strong laser pulses in otherwise optically thick media. It was shown by Harris [10] that pulse pairs with arbitrary but identical envelopes (matched pulses) may propagate undisturbed, if the three-level atoms are prepared in the uncoupled coherent superposition of lower levels. For sufficiently strong pulses the preparation of the atoms is done at the front end of the pulses via coherent population transfer [11]. Furthermore as pointed out by Harris, matched pulses may also be generated by the matter-field interaction (pulse matching) for a variety of specific initial pulse shapes without coherent preparation of the atoms [12]. Konopnicki and Eberly showed that pulse pairs with the same hyperbolic secant envelope (simultons) and total pulse area 2π represent a soliton solution which remains formstable with very small energy losses [13]. Recently Grobe, Hioe, and Eberly predicted another class of formstable solutions which have

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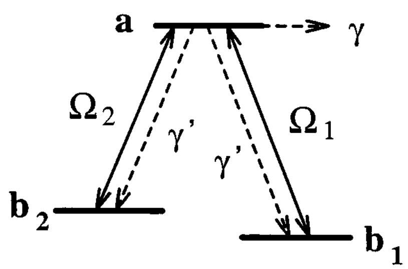

FIG. 1. Atomic system in bare-state representation. Two resonant pulses with Rabi-frequencies Ω_1 and Ω_2 couple upper level a to b_1 and b_2.

complementary pulse shapes (adiabatons) [14].

All these propagation phenomena have in common a quasiadiabatic nature of the matter-field interaction. Therefore the above mentioned approach of Maxwell-Bloch equations in the basis of adiabatic dark and bright states seems very appropriate to study pulse propagation phenomena as well. We will show that a perturbation in the nonadiabatic coupling allows an approximate analytical solution of the nonlinear dynamical equations and provides a simple physical explanation for the loss-free propagation of matched pulses, simultons, and adiabatons. We find that in contrast to matched pulses, adiabatons decay for long propagation distances and we point out the relation between adiabatons and the formation of matched pulses.

The paper is organized as follows. In Sec. II we derive the Maxwell-Bloch equations in terms of adiabatic dark and bright states. In Sec. III we analyze the dynamics of the atomic system for a given pair of pulses under quasiadiabatic conditions. In particular we study the dynamics of coherent population transfer (STIRAP). In Sec. IV we focus on the evolution of the fields and discuss the formation and propagation of matched pulses, simultons, and adiabatons.

II. MAXWELL-BLOCH EQUATIONS IN A BASIS OF ADIABATIC DARK AND BRIGHT STATES

We consider here the interaction of two copropagating pulses with three-level Λ-type atoms as shown in Fig. 1. The propagation direction is z. For the sake of simplicity and in order to obtain analytical results, we restrict our model to the bare essentials. We assume pulses with resonant carrier frequencies, ignore transverse effects and inhomogeneous broadening, and assume a symmetric situation of equal coupling strength and radiative decay rates. A pulse of Rabi frequency $\Omega_1(z,t)$ couples the transition $|a\rangle-|b_1\rangle$ and another one with Rabi frequency $\Omega_2(z,t)$ couples the $|a\rangle-|b_2\rangle$ transition. The population in the upper level $|a\rangle$ decays radiatively into levels $|b_{1,2}\rangle$ with a rate γ' and to some other states with rate γ. The decay rate of the optical coherences is denoted by $\gamma_\perp \geqslant \gamma' + \gamma/2$. The density matrix equations for the atomic system are given in a rotating frame by

$$\dot{\rho}_{aa} = -\Gamma\rho_{aa} - i(\Omega_1^*\rho_{ab_1} - \text{c.c.}) - i(\Omega_2^*\rho_{ab_2} - \text{c.c.}), \quad (1)$$

$$\dot{\rho}_{b_1b_1} = \gamma'\rho_{aa} + i(\Omega_1^*\rho_{ab_1} - \text{c.c.}), \quad (2)$$

$$\dot{\rho}_{b_2b_2} = \gamma'\rho_{aa} + i(\Omega_2^*\rho_{ab_2} - \text{c.c.}), \quad (3)$$

$$\dot{\rho}_{b_1b_2} = i\Omega_1^*\rho_{ab_2} - i\Omega_2\rho_{ab_1}^*, \quad (4)$$

$$\dot{\rho}_{ab_1} = -\gamma_\perp\rho_{ab_1} - i\Omega_1(\rho_{aa} - \rho_{b_1b_1}) + i\Omega_2\rho_{b_1b_2}^*, \quad (5)$$

$$\dot{\rho}_{ab_2} = -\gamma_\perp\rho_{ab_2} - i\Omega_2(\rho_{aa} - \rho_{b_2b_2}) + i\Omega_1\rho_{b_1b_2}, \quad (6)$$

where $\Gamma = 2\gamma' + \gamma$.

It is well established that in the case of cw-fields in two-photon resonance, the atom-field interaction is best described in terms of the coupled (''bright'' $|+\rangle$) and decoupled (''dark'' $|-\rangle$) superposition states of the lower levels:

$$|+\rangle = \frac{1}{\Omega}[\Omega_1^*|b_1\rangle + \Omega_2^*|b_2\rangle], \quad (7)$$

$$|-\rangle = \frac{i}{\Omega}[\Omega_2|b_1\rangle - \Omega_1|b_2\rangle], \quad (8)$$

where

$$\Omega(z,t) \equiv [|\Omega_1(z,t)|^2 + |\Omega_2(z,t)|^2]^{1/2} \quad (9)$$

is the total Rabi frequency. In terms of these states the atom-field interaction Hamiltonian reads

$$H = -\hbar\Omega|a\rangle\langle +| + \text{H.a.} \quad (10)$$

The state $|-\rangle$ is decoupled from the interaction for which reason it is called dark state.

In the case of pulses, $|+\rangle$ and $|-\rangle$ are generally time-dependent. A description in terms of these states is nevertheless useful. If we write the density matrix equations of the atomic system (which we call Bloch equations in the following) in terms of $|\pm\rangle$, new terms appear due to the explicit time-dependence. In a rotating frame we find

$$\dot{\rho}_{aa} = -\Gamma\rho_{aa} - i\Omega(\rho_{a+} - \text{c.c.}), \quad (11)$$

$$\dot{\rho}_{++} = \gamma'\rho_{aa} + i\Omega(\rho_{a+} - \text{c.c.}) + i(\Omega_-^*\rho_{-+} - \text{c.c.}), \quad (12)$$

$$\dot{\rho}_{--} = \gamma'\rho_{aa} - i(\Omega_-^*\rho_{-+} - \text{c.c.}), \quad (13)$$

$$\dot{\rho}_{+-} = 2i\Delta\rho_{+-} + i\Omega\rho_{a-} - i\Omega_-^*(\rho_{++} - \rho_{--}), \quad (14)$$

$$\dot{\rho}_{a+} = -(\gamma_\perp + i\Delta)\rho_{a+} - i\Omega(\rho_{aa} - \rho_{++}) - i\Omega_-\rho_{a-}, \quad (15)$$

$$\dot{\rho}_{a-} = -(\gamma_\perp - i\Delta)\rho_{a-} + i\Omega\rho_{+-} - i\Omega_-^*\rho_{a+}, \quad (16)$$

where we have introduced the ''Rabi frequency'' Ω_- of a formal nonadiabatic coupling between $|+\rangle$ and $|-\rangle$,

$$\Omega_- \equiv \frac{\dot{\Omega}_1^*\Omega_2^* - \dot{\Omega}_2^*\Omega_1^*}{\Omega^2} \quad (17)$$

and a (space and time-dependent) detuning

$$\Delta = \frac{\dot{\phi}_1|\Omega_1|^2 + \dot{\phi}_2|\Omega_2|^2}{\Omega^2}. \quad (18)$$

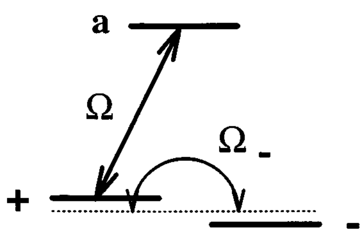

FIG. 2. Λ-system in dressed state representation.

Here $\Omega_j=|\Omega_j|e^{i\phi_j}$ $(j=1,2)$. The Bloch equations (11)–(16) correspond to the situation of a three-level system driven by two fields. One ''field'' with Rabi frequency Ω couples the upper level $|a\rangle$ to the bright state $|+\rangle$, and another ''field'' with Rabi frequency Ω_- couples the two superposition states $|+\rangle$ and $|-\rangle$. This is illustrated in Fig. 2.

A problem for the solution of the Bloch equations is the space and time-dependent detuning Δ. We note, however, that the Rabi-frequencies Ω_1 and Ω_2 of the two fields remain real throughout the interaction process, if they are real initially, and if the initial values of $\rho_{b_1b_2}$ and $i\rho_{ab_{1,2}}$ are real as well. Under these conditions we have

$$\Omega_-=\frac{\dot{\Omega}_1\Omega_2-\dot{\Omega}_2\Omega_1}{\Omega^2} \tag{19}$$

and

$$\Delta=0, \tag{20}$$

and the Bloch equations simplify considerably. This is the situation we will focus on in the following.

The Maxwell equations for the propagation of the two ''fields'' expressed in terms of the Rabi frequencies Ω and Ω_- read in the slowly-varying-amplitude-and-phase approximation

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial z}\right)\Omega(z,t)=-g^2N\mathrm{Im}[\rho_{a+}] \tag{21}$$

and

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial z}\right)\Omega_-(z,t)=g^2N\frac{\partial}{\partial t}\left(\frac{\rho_{a-}}{\Omega}\right). \tag{22}$$

Here $g=(\wp/\hbar)\sqrt{\hbar\nu/2\varepsilon_0}$ is the strength of the atom-field coupling; $\wp$ being the dipole moment, ν the transition frequency, and N is the density of atoms. As ususal for pulse-propagation problems, we introduce moving coordinates $\xi=z$ and $\tau=t-z/c$, such that the wave operator $\partial/\partial t+c\partial/\partial z$ becomes $c\partial/\partial\xi$.

The solution of the coupled system of dressed-state Bloch equations (11)–(16) and dressed-field Maxwell equations (21) and (22) is sufficient to determine the evolution of the two pulses. Ω and Ω_- are related to the Rabi-frequencies of the original pulses by

$$\Omega_1(\xi,\tau)=\Omega(\xi,\tau)\sin\left\{\int_{-\infty}^{\tau}d\tau'\,\Omega_-(\xi,\tau')\right\}, \tag{23}$$

$$\Omega_2(\xi,\tau)=\Omega(\xi,\tau)\cos\left\{\int_{-\infty}^{\tau}d\tau'\,\Omega_-(\xi,\tau')\right\}, \tag{24}$$

where we have assumed that $\Omega_1/\Omega_2\to 0$ for $t\to-\infty$.

III. POPULATION DYNAMICS

In the present section we study the dynamical evolution of the atomic system in a given time-dependent field. Although a generalization is straightforward, we disregard here spontaneous transitions into the lower levels, i.e., set $\Gamma=\gamma$ and assume $\gamma_\perp=\gamma/2$. In this case we may describe the atomic evolution with a Schrödinger-type equation for the state amplitudes $\{c_-,c_a,c_+\}$:

$$\frac{d}{dt}\begin{pmatrix}c_-\\ c_a\\ c_+\end{pmatrix}=\begin{pmatrix}0 & 0 & i\Omega_-\\ 0 & -\gamma_\perp & i\Omega\\ i\Omega_- & i\Omega & 0\end{pmatrix}\begin{pmatrix}c_-\\ c_a\\ c_+\end{pmatrix}. \tag{25}$$

We now introduce a dimensionless unit $\overline{t}=t/T$, where T is a characteristic time of the pulses, and separate an overall time dependence $\Omega(t)=\Omega_0f(t)$. With this we find

$$\epsilon\frac{d}{d\overline{t}}\begin{pmatrix}c_-\\ c_a\\ c_+\end{pmatrix}=\begin{pmatrix}0 & 0 & i\epsilon\overline{\Omega}_-\\ 0 & -\gamma_\perp/\Omega_0 & if(\overline{t})\\ i\epsilon\overline{\Omega}_- & if(\overline{t}) & 0\end{pmatrix}\begin{pmatrix}c_-\\ c_a\\ c_+\end{pmatrix}. \tag{26}$$

Here $\epsilon\equiv(\Omega_0T)^{-1}$ is a small parameter characterizing the nonadiabaticity of the interaction process and $\overline{\Omega}_-$ follows from Eq. (19) with d/dt replaced by $d/d\overline{t}$.

For the cases in which we are interested here, the adiabaticity parameter ϵ is small. For this reason one might try an approximate solution of Eq. (26) using a perturbation expansion in ϵ. It is well known, however, from the theory of adiabatic processes in two-state systems, that such a perturbation must be handled with care. If the system starts from one of the instantaneous eigenstates of the Hamiltonian, the nonadiabatic loss from this state is small beyond any order in ϵ and cannot be obtained from a perturbation expansion [7]. The same problem occurs here for $\gamma=0$, in which case the three-level system can be mapped onto a two-state system [5]. The presence of the decay term in Eq. (26) however, changes the situation substantially. If we let $\epsilon\to 0$ keeping $\gamma_\perp/\Omega_0$ fixed, the left-hand side (lhs) of the equation of motion of c_a vanishes:

$$\epsilon\frac{d}{d\overline{t}}c_a=-\frac{\gamma_\perp}{\Omega_0}c_a+if(\overline{t})c_+. \tag{27}$$

More precisely if $\epsilon\ll\gamma_\perp/\Omega_0$ or $T\gg\gamma_\perp^{-1}$ we may neglect the lhs of Eq. (27) as compared to the first term on the rhs since c_a changes on a time-scale of unity. This gives

$$c_a=\frac{i\Omega_0f(\overline{t})}{\gamma_\perp}c_+. \tag{28}$$

Substituting this result into the equation for c_+ we obtain

$$\epsilon \frac{d}{dt} c_+ = -\frac{\Omega_0 f^2(\overline{t})}{\gamma_\perp} c_+ + i\epsilon \overline{\Omega}_-(\overline{t}) c_- . \tag{29}$$

Now again in the limit $\epsilon \to 0$ the lhs is negligible as compared to the first term on the rhs yielding

$$c_+ = i\epsilon \frac{\gamma_\perp}{\Omega_0} \frac{\overline{\Omega}_-(\overline{t})}{f^2(\overline{t})} c_- . \tag{30}$$

Substituting this result into the equation for c_- we eventually arrive at

$$\frac{d}{dt} c_- = -\epsilon \frac{\gamma_\perp}{\Omega_0} \frac{\overline{\Omega}_-^2(\overline{t})}{f^2(\overline{t})} c_- , \tag{31}$$

that has the simple solution

$$c_-(\overline{t}) = c_-(-\infty) \exp\left\{ -\epsilon \frac{\gamma_\perp}{\Omega_0} \int_{-\infty}^{\overline{t}} d\overline{t}' \frac{\overline{\Omega}_-^2(\overline{t'})}{f^2(\overline{t'})} \right\}. \tag{32}$$

We point out that the nonadiabatic loss, $1 - |c_-(\overline{t})/c_-(-\infty)|^2$, can be expanded into a power series in ϵ and is *not* exponentially small in $1/\epsilon$. This particular property of the dissipative system makes it possible to study the quasiadiabatic situation using a perturbation approach in the nonadiabatic coupling. In fact the failure of this procedure for a three-level system without decay and inhomogeneous broadening seems to be an artifact of the idealization of the true situation. Recently Shapiro gave a description of coherent population transfer into a flat continuum beyond the adiabatic approximation [15]. Also in this case, the nonadiabatic losses can be expanded in powers of ϵ.

A. Coherent population transfer for $T \gg \gamma_\perp^{-1}$

We now discuss the coherent population transfer from level $|b_1\rangle$ to $|b_2\rangle$ by a counterintuitive pulse sequence for characteristic times large compared to the decay time from the excited state. If the atomic system is initially in $|b_1\rangle$ and the two overlapping pulses are applied such that Ω_2 is switched on and off first (counterintuitive sequence), $|-\rangle$ is identical to $|b_1\rangle$ for $t = -\infty$ and to $|b_2\rangle$ for $t = +\infty$. In the adiabatic limit $\epsilon \to 0$ the system stays in the dark state and all population is transferred from $|b_1\rangle$ to $|b_2\rangle$.

For $T \gg \gamma_\perp^{-1}$ the history of the transition process is given by Eqs. (28)–(32):

$$c_a(t) = -\frac{\Omega_-(t)}{\Omega(t)} c_-(t), \tag{33}$$

$$c_+(t) = \frac{i\gamma_\perp \Omega_-(t)}{\Omega^2(t)} c_-(t), \tag{34}$$

$$c_-(t) = \exp\left\{ -\gamma_\perp \int_{-\infty}^{t} dt' \frac{\Omega_-^2(t')}{\Omega^2(t')} \right\}. \tag{35}$$

The asymptotic nonadiabatic loss, i.e., the amount of population not transfered, is

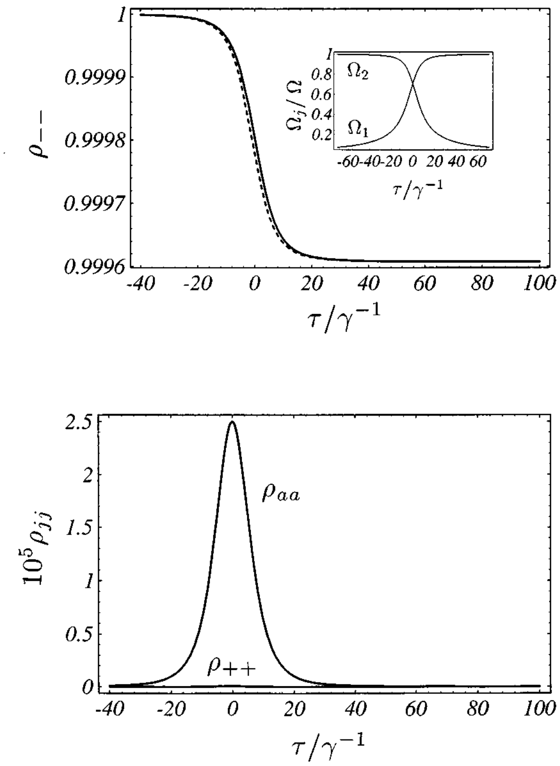

FIG. 3. Coherent population transfer for $T \gg \gamma_\perp^{-1}$. (a) Dark-state population from analytical approximation (dark line) and numerical calculation (dashed line). (b) Population in $|+\rangle$ and $|a\rangle$. The analytical approximation and exact numerical results are almost indistinguishable. The inset in (a) shows the form of the incident ramped pulses.

$$1 - |c_-(\infty)|^2 = 1 - \exp\left\{ -\gamma \int_{-\infty}^{\infty} dt' \frac{\Omega_-^2(t')}{\Omega^2(t')} \right\}. \tag{36}$$

We note that according to Eqs. (23) and (24) for a complete population transfer from $|b_1\rangle$ to $|b_2\rangle$ with a counterintuitive pulse sequence,

$$\int_{-\infty}^{\infty} dt \Omega_-(t) = \frac{\pi}{2}. \tag{37}$$

One recognizes from Eq. (35) that in the case of a population transfer slower than the coherence decay, the population in the dark state decreases smoothly and monotonically from unity to its final value. During this process, population is built up in the excited and bright states proportional to the Rabi frequency of the nonadiabatic coupling. We have illustrated this in Fig. 3 for the example of ramped pulses $\Omega_1 = \Omega \sin[\frac{1}{2}\arctan(\gamma t/10) + \pi/4]$, $\Omega_2 = \Omega \cos[\frac{1}{2}\arctan(\gamma t/10) + \pi/4]$, such that $\Omega = 10\gamma$ and $\Omega_- = 0.05\gamma/[1 + (0.1\gamma t)^2]$. Shown are the populations in all levels according to Eqs. (33)–(35) and following from a numerical calculation.

B. Coherent population transfer for $T \leqslant \gamma_\perp^{-1}$

If the characteristic pulse time is on the order of, or short compared to, the upper level decay time, we can no longer neglect the time-derivatives in Eq. (26). We may, however, solve the equations of motion perturbatively in the nonadiabatic coupling. In lowest order and with the initial condition $c_-(-\infty)=1$, we find

$$c_-(t)=1+i\int_{-\infty}^{t} dt'\,\Omega_-(t')c_+(t'), \tag{38}$$

where c_+ follows from

$$\frac{d}{dt}\begin{pmatrix} c_a \\ c_+ \end{pmatrix}=\begin{pmatrix} -\gamma_\perp & i\Omega(t) \\ i\Omega(t) & 0 \end{pmatrix}\begin{pmatrix} c_a \\ c_+ \end{pmatrix}+i\Omega_-(t)\begin{pmatrix} 0 \\ 1 \end{pmatrix}. \tag{39}$$

These equations can be solved analytically only for $\Omega=\text{const}$. We will therefore restrict ourselves in the following to this case. We find for the amplitudes of the adiabatic dressed states

$$c_a(t)=-\frac{\Omega}{\Omega'}\int_0^\infty d\xi\,\Omega_-(t-\xi)e^{-\gamma_\perp/2\,\xi}\sin(\Omega'\xi), \tag{40}$$

$$c_+(t)=i\int_0^\infty d\xi\,\Omega_-(t-\xi)\times e^{-\gamma_\perp/2\,\xi}\left[\cos(\Omega'\xi)+\frac{\gamma_\perp}{2\Omega'}\sin(\Omega'\xi)\right], \tag{41}$$

$$c_-(t)=1-\int_{-\infty}^{t} d\tau\,\Omega_-(\tau)\int_0^\infty d\xi\,\Omega_-(\tau-\xi)\times e^{-\gamma_\perp/2\,\xi}\left[\cos(\Omega'\xi)+\frac{\gamma_\perp}{2\Omega'}\sin(\Omega'\xi)\right], \tag{42}$$

where $\Omega'=[\Omega^2-(\gamma_\perp/2)^2]^{1/2}$. To discuss the history of the population transfer we consider the example $\Omega=10$, $\Omega_-=0.5/(1+t^2)$, and $\gamma=10^{-2}$ (now all in arbitrary time units). In Fig. 4 we have plotted the population in the adiabatic dark and bright states and in the upper level according to the results from first order perturbation and following from an exact numerical calculation. In contrast to the case studied in the preceding subsection, the population now undergoes a large excursion away from the dark state and eventually returns to it. One also recognizes that almost all of this population is driven into level $|a\rangle$ while the bright state remains virtually empty. The physical mechanism of this process is a Raman-type transition between $|-\rangle$ and $|a\rangle$ via the two "fields" Ω and Ω_-. Most of the population is thereby trapped in a superposition state $\sim(\Omega|-\rangle-\Omega_-|a\rangle)$ that represents the uncoupled dressed state in a (first-order) superadiabatic basis [16].

Expanding $\Omega_-(\tau-\xi)$ in Eqs. (40)–(42) into powers of ξ one can evaluate the integrals. A scaling analysis shows that all but the first few terms are higher order in the adiabaticity parameter ϵ. Keeping only the lowest order terms we find the simple analytic expressions for the history of the dressed-state amplitudes:

$$c_a(t)=-\frac{\Omega_-(t)}{\Omega}+\frac{\gamma_\perp}{\Omega}\frac{\dot\Omega_-(t)}{\Omega^2}, \tag{43}$$

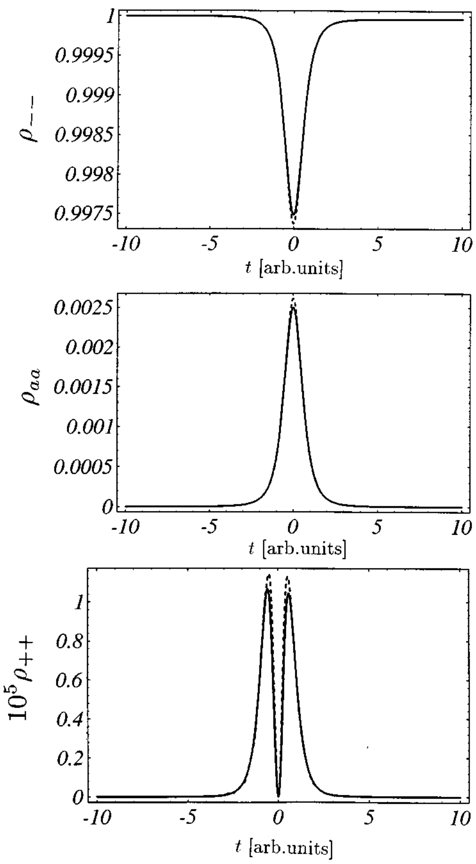

FIG. 4. Coherent population transfer for $T\leq\gamma^{-1}$. Shown are the populations in the dark state (a), the upper level (b), and the bright state (c) following from analytical approximation (dark line) and numerical calculation (dashed line).

$$c_+(t)=i\frac{\gamma_\perp}{\Omega}\frac{\Omega_-(t)}{\Omega}+i\frac{\dot\Omega_-(t)}{\Omega^2}\left(1-\frac{\gamma_\perp^2}{\Omega^2}\right), \tag{44}$$

$$c_-(t)=1-\gamma_\perp\int_{-\infty}^{t} d\tau\frac{\Omega_-^2(\tau)}{\Omega^2}-\frac{\Omega_-^2(t)}{2\Omega^2}\left(1-\frac{\gamma_\perp^2}{\Omega^2}\right). \tag{45}$$

In the limit $t\to+\infty$ only the first two terms in c_- survive. This yields an expression for the asymptotic nonadiabatic loss which is identical to the result of the last subsection in lowest order of the nonadiabatic coupling:

$$c_-(\infty)=1-\gamma_\perp\int_{-\infty}^{\infty} dt\frac{\Omega_-^2(t)}{\Omega^2}. \tag{46}$$

In Table I we have compared the asymptotic values of c_- according to Eq. (46) with exact numerical results for the example of Fig. 4 for values of γ/Ω ranging from 0.1 to 10^{-6}. We note the excellent agreement even for very small damping rates. A substantial deviation from the exact result occurs only if the nonadiabatic loss according to Eq. (46) is less than the value following from the superadiabatic approach of Ref. [8] for $\gamma=0$. The exponentially small diabatic loss in this case is $(8-4\sqrt{2})e^{-20}\approx4.83\times10^{-9}$. The regime of very small decay, such that $\epsilon\gamma_\perp\leq\Omega\exp\{-2/\epsilon\}$, cannot be

TABLE I. $1-c_-(\infty)$.

γ/Ω	analytical from Eq. (46)	numerical
10^{-1}	1.9635×10^{-3}	1.9788×10^{-3}
10^{-2}	1.9635×10^{-4}	1.9807×10^{-4}
10^{-3}	1.9635×10^{-5}	1.9819×10^{-5}
10^{-4}	1.9635×10^{-6}	1.9919×10^{-6}
10^{-5}	1.9635×10^{-7}	2.0926×10^{-7}
10^{-6}	1.9635×10^{-8}	3.1009×10^{-8}

described by the present approach, since then the major contributions to the nonadiabatic losses are nonpolynomial in ϵ. For most practical applications the upper level decay is, however, larger than the critical value at which the validity of the perturbation approach breaks down.

IV. PULSE PROPAGATION

A. Adiabatic limit, $\Omega_-=0$

The dark state $|-\rangle$, Eq. (8), is truly decoupled from the laser fields if it is not explicitly time-dependent, that is, if $\Omega_-=0$. One can easily see that this is the case when

$$\frac{\dot{\Omega}_1}{\Omega_1}=\frac{\dot{\Omega}_2}{\Omega_2}. \tag{47}$$

This condition implies that the two pulses can have arbitrary strength but need to have identical shapes.

If the medium is initially prepared in a coherent superposition of lower levels, such that $\rho_{--}(0)=1$, pulse pairs that fulfill the condition $\Omega_-(0,\tau)=0$ will propagate loss free through the medium which is optically thick for each of the individual pulses. Since the system stays in the dark state $|-\rangle$ we have $\rho_{a+}=\rho_{a-}=0$ and hence

$$c\frac{\partial}{\partial\xi}\Omega(\xi,\tau)=0, \tag{48}$$

$$c\frac{\partial}{\partial\xi}\Omega_-(\xi,\tau)=0. \tag{49}$$

This is the situation of *matched pulses* discovered by Harris [10]. It was also shown by Harris and Luo [11] that the pulses self-generate the required atomic coherence if the atom is initially in one of the lower states, $|b_{1,2}\rangle$. The physical mechanism of this preparation is a coherent population transfer at the front end of the pulses.

For a pulse pair with identical shapes $[\Omega_-(0,\tau)=0]$ and all initial population in states $|a\rangle$ and $|+\rangle$ instead of $|-\rangle$, the propagation problem reduces to that of a two-level atom in a single field of Rabi frequency Ω. Since the dark state is decoupled from the coherent interaction and can only be reached by spontaneous transitions from $|a\rangle$, we have

$$\rho_{a-}\equiv0 \tag{50}$$

and

$$c\frac{\partial}{\partial\xi}\Omega_-(\xi,\tau)=0. \tag{51}$$

Hence the dark state remains decoupled throughout the interaction. The reduced set of Bloch equations for the nonvanishing density matrix elements reads

$$\dot{\rho}_{aa}=-\gamma\rho_{aa}-i\Omega(\rho_{a+}-\text{c.c.}), \tag{52}$$

$$\dot{\rho}_{++}=+i\Omega(\rho_{a+}-\text{c.c.}), \tag{53}$$

$$\dot{\rho}_{a+}=-\gamma_\perp\rho_{a+}-i\Omega(\rho_{aa}-\rho_{++}), \tag{54}$$

and the total Rabi frequency evolves according to

$$c\frac{\partial}{\partial\xi}\Omega(\xi,\tau)=-g^2N\,\text{Im}[\rho_{a+}]. \tag{55}$$

For a two-level system interacting with a single field, a couple of soliton solutions that preserve the initial pulse shape and display anomalously small energy loss are known. The most famous one is the McCall-Hahn 2π-hyperbolic secant pulse of self-induced transparency [17]:

$$\Omega(\xi,\tau')=\frac{1}{T}\,\text{sech}\left[\frac{\tau'}{T}\right], \tag{56}$$

where $\xi=z$ and $\tau'=t-z/v$. v is the group velocity of the pulses

$$\frac{1}{v}=\frac{1}{c}(1+g^2NT^2). \tag{57}$$

This means, if the atomic system is initially prepared in the bright state $|+\rangle$, a pair of pulses with the same hyperbolic secant shape and a total pulse area of 2π will propagate formstable through the three-level medium. The corresponding dynamics of the atomic system in the absence of decay is given by

$$\rho_{aa}-\rho_{++}=2\,\text{sech}^2\left[\frac{\tau'}{T}\right]-1, \tag{58}$$

$$\rho_{a+}=-i\,\text{sech}\left[\frac{\tau'}{T}\right]\tanh\left[\frac{\tau'}{T}\right]. \tag{59}$$

If we transform Eqs. (56)–(59) back into the original basis we obtain the *simulton* solutions first found by Konopnicki and Eberly [13],

$$\Omega_1(\xi,\tau')=\frac{\alpha_1}{T}\text{sech}\left[\frac{\tau'}{T}\right], \tag{60}$$

$$\Omega_2(\xi,\tau')=\frac{\alpha_2}{T}\text{sech}\left[\frac{\tau'}{T}\right], \tag{61}$$

where $\alpha_{1,2}$ are arbitrary real constants with $\alpha_1^2+\alpha_2^2=1$. The corresponding solutions of the Bloch equations are

$$\rho_{ab_{1,2}}=-i\,\alpha_{1,2}\text{sech}\left[\frac{\tau'}{T}\right]\tanh\left[\frac{\tau'}{T}\right], \tag{62}$$

$$\rho_{b_\mu b_\mu}=\alpha_\mu^2\left(1-\text{sech}^2\left[\frac{\tau'}{T}\right]\right), \tag{63}$$

$$\rho_{b_1b_2}=\alpha_1\alpha_2\left(1-\mathrm{sech}^2\left[\frac{\tau'}{T}\right]\right). \tag{64}$$

We have seen that the dark state of the Λ-system is decoupled from the interaction with the fields if both pulses have the same shape. There are two cases in which a formstable pulse propagation is possible. If initially all population is in the dark state it is completely hidden from the fields and the pulses ''see'' no atoms at all. The strength and the form of the pulses are irrelevant as long as they are matched. The propagation of these *matched pulses* is completely unaffected by losses from the excited state. In the complementary case where all initial population is in the subsystem formed by the bright state $|+\rangle$ and the upper state $|a\rangle$, the system reduces to a two-level–single-field problem. For such a system formstable propagation is possible, if the conditions for self-induced transparency in the adiabatic dressed state basis are fulfilled. This means both pulses must have a hyperbolic secant shape and the total field must have a pulse area of 2π. In contrast to matched pulses, these *simultons* are affected by the decay from the upper level. Eventually all population will end up in the dark state due to spontaneous transitions and the matched-pulse situation is reached.

B. Quasiadiabaticity, $|\Omega_-|\ll\Omega$

We now consider the case of a nonvanishing but small nonadiabatic coupling. For slowly varying pulse shapes, such that

$$|\Omega_-|\ll\Omega \quad \text{or} \quad |\dot{\Omega}_1\Omega_2-\dot{\Omega}_2\Omega_1|\ll\Omega^3, \tag{65}$$

the coupling between the dark and bright dressed states is weak compared to the coupling of $|+\rangle$ to the upper level $|a\rangle$. As shown in Sec. III, we may treat the Ω_--coupling perturbatively in this case. We recognize from Eqs. (11)–(16) and (22) that condition (65) can only be maintained, if initially all population is in the decoupled state:

$$\rho_{--}(0)=1. \tag{66}$$

This can be realized either by preparing the atoms in a coherent superposition of the bare states with weights given by the Rabi-frequencies or by adjusting the pulse shapes at the front end according to the initial atomic configuration. The second situation is the favorable one, if the energy splitting between the lower states is larger than the thermal energy, such that only one of them, say $|b_1\rangle$, is populated initially. In this case one has to ensure that Ω_2 is switched on first since then $|-\rangle$ is identical to $|b_1\rangle$. Note that in any case the atoms must be initially in a pure state.

If the conditions given in Eqs. (65) and (66) are fulfilled, the set of coupled nonlinear Maxwell-Bloch equations may be solved analytically by a perturbation expansion in $\epsilon\sim|\Omega_-|/\Omega$. Since initially all population is in $|-\rangle$, only coherences between the states $|+\rangle$ and $|-\rangle$ and between $|a\rangle$ and $|-\rangle$ are built up in first order of ϵ. We therefore have

$$\rho_{a+}^{(1)}=0, \tag{67}$$

$$\rho_{aa}^{(1)}=0, \tag{68}$$

$$\rho_{++}^{(1)}=0. \tag{69}$$

Equation (67) has the immediate consequence that the total Rabi frequency is undisturbed,

$$c\frac{\partial}{\partial\xi}\Omega(\xi,\tau)=0. \tag{70}$$

This means that under quasiadiabatic conditions, the total ''energy'' of the pulses remains the same for propagation distances large compared to the one-photon absorption length of the medium. In this limit the photons are redistributed between the fields by stimulated Raman scattering and the effect of dissipation on the total field is negligible.

In order to determine the field dynamics we have to solve the remaining equations of motion for Ω_-, ρ_{a-}, and ρ_{+-}:

$$c\frac{\partial}{\partial\xi}\Omega_-(\xi,\tau)=g^2N\frac{\partial}{\partial\tau}\left(\frac{\rho_{a-}^{(1)}}{\Omega}\right) \tag{71}$$

and

$$\frac{\partial}{\partial\tau}\rho_{a-}^{(1)}(\tau)=-\gamma_\perp\rho_{a-}^{(1)}+i\Omega\rho_{+-}^{(1)}, \tag{72}$$

$$\frac{\partial}{\partial\tau}\rho_{+-}^{(1)}(\tau)=i\Omega\rho_{a-}^{(1)}+i\Omega_-. \tag{73}$$

As we have shown in Sec. III, the optical coherence can be eliminated adiabatically if the characteristic time of changes in the fields, in particular in Ω_-, is long compared to the decay time of the upper level. Note that no assumption about the strength of the coupling is required. This yields

$$\rho_{a-}^{(1)}=\frac{i\Omega}{\gamma_\perp}\rho_{+-}^{(1)}. \tag{74}$$

Second, we introduce a nonlinear time-stretch similar to that used in Ref. [14],

$$T\equiv\frac{1}{\Omega_0^2}\int_{-\infty}^{\tau}d\tau'\,\Omega^2(0,\tau'), \tag{75}$$

where Ω_0 is some appropriately chosen average value of the total Rabi frequency. If the total Rabi frequency is constant for a long time period, one could take this value for Ω_0. T is a nonlinear but monotonic function of τ. Using (70) and (74) the equations of motion in terms of T can be written as

$$c\frac{\partial}{\partial\xi}F(\xi,T)=i\frac{g^2N}{\Omega_0^2\gamma_\perp}\frac{\partial}{\partial T}\rho_{+-}^{(1)}(T), \tag{76}$$

$$\frac{\partial}{\partial T}\rho_{+-}^{(1)}(T)=-\frac{\Omega_0^2}{\gamma_\perp}\rho_{+-}(T)+i\Omega_0^2F(\xi,T), \tag{77}$$

where $F(\xi,T)\equiv\Omega_-(\xi,T)/\Omega^2(0,T)$. Thus by applying a perturbation approach in the nonadiabatic coupling and a nonlinear time-stretch we have transformed the original set of coupled nonlinear equations into a pair of linear, first-order, partial differential equations with constant coefficients. We can very easily solve these equations by a Fourier-transformation with respect to T. Note that F as well as ρ_{+-} are quadratically integrable in T. We eventually arrive at

$$F(\xi,\omega)=F(0,\omega)\exp\left\{-g^2N\left[\frac{i\omega\Omega_0^2}{\Omega_0^4+\omega^2\gamma_\perp^2}+\frac{\omega^2\gamma_\perp}{\Omega_0^4+\omega^2\gamma_\perp^2}\right]\frac{\xi}{c}\right\}. \tag{78}$$

The first term in the exponent of Eq. (78) describes a dispersive propagation of the ''pulse'' F. The second term describes an absorption of the high-frequency components of F. If all relevant Fourier-frequencies are sufficiently small, such that $\omega \ll \Omega_0^2/\gamma_\perp$, the absorption term and the nonlinear correction to the dispersion can be neglected. In this case, Eq. (78) describes the propagation of the ''pulse'' F in a linear dispersive medium. Hence F propagates formstable with a group velocity

$$\frac{1}{v}=\frac{1}{c}\left(1+\frac{g^2N}{\Omega_0^2}\right), \tag{79}$$

$$F(\xi,T)=F\left(0,T-\frac{g^2N}{\Omega_0^2}\frac{\xi}{c}\right). \tag{80}$$

These solutions of the nonlinear Maxwell-Bloch equations in the quasiadiabatic limit are the *adiabatons* introduced by Grobe, Hioe, and Eberly in Ref. [14]. They are not restricted to certain types of pulse shapes as long as condition (65) is fulfilled. It should be noted, however, that Eq. (80) describes formstable propagation with respect to T and not to the actual time τ. Only if Ω is constant over a period of time long compared to the extention of the adiabaton is the propagation formstable in the usual sense. In this case, the pulse envelopes have a complementary shape $\Omega_1^2=\text{const}-\Omega_2^2$.

For $\Omega=$ const, an analytic solution of the Maxwell-Bloch equations (71)–(73) is also possible for characteristic pulse times of the order of, or short compared to, the upper level decay time. In this case, we can simply Fourier-transform the corresponding equations and find

$$\Omega_-(\xi,\omega)=\Omega_-(0,\omega)\exp\left\{-g^2N\left[\frac{i\omega(\Omega^2-\omega^2)}{(\Omega^2-\omega^2)^2+\omega^2\gamma_\perp^2}+\frac{\omega^2\gamma_\perp^2}{(\Omega^2-\omega^2)^2+\omega^2\gamma_\perp^2}\right]\right\}. \tag{81}$$

We again recognize the presence of a dispersive and a high-frequency-absorption term. The formstable adiabaton solution is obtained when the relevant Fourier-frequencies are small compared to Ω and $\Omega^2/\gamma_\perp$.

In Fig. 5, an example of an adiabaton with complementary pulse envelopes is displayed. We show the analytical result according to Eq. (81) and a numerical calculation. One can recognize a formstable propagation over many one-photon absorption lengths $\xi_a \equiv \gamma_\perp c/g^2N=(\gamma_\perp/\Omega_0)\xi_0$.

There are two mechanisms that limit the lifetime of an adiabaton. First, since its group velocity is smaller than c but the total field propagates with the speed of light, the adiabaton will eventually reach the back end of the pulses where the adiabatic condition (65) is violated. Second, for longer

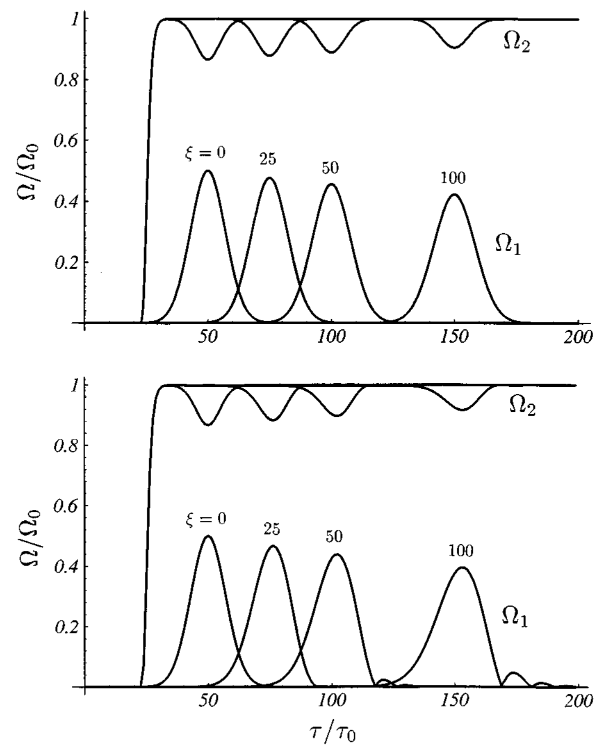

FIG. 5. Propagation of an adiabaton for different propagation distances within the medium in units of $\xi_0=(g^2N/\Omega_0c)^{-1}$ for $\gamma_\perp/\Omega_0=0.1$. The time τ is measured in units of $\tau_0=\Omega_0^{-1}$. $\Omega_1(0,\tau)=0.5\Omega_0\exp[-(\tau-50)^2/100]$, $\Omega_2^2=\Omega^2-\Omega_1^2$, and $\Omega(0,\tau)=\Omega_0\exp[-((\tau-125)/100)^{80}]$. Analytical results are shown in (a), numerical results in (b).

propagation distances the absorption term in Eq. (78) cannot be neglected and the adiabaton decays. Eventually the ''fields'' F or Ω_- die away, such that

$$\frac{\dot\Omega_1}{\Omega_1}\rightarrow\frac{\dot\Omega_2}{\Omega_2}\quad\text{for}\quad\xi\rightarrow\infty. \tag{82}$$

This means that the interaction with the medium generates pulses with identical pulse shapes — a process first discovered by Harris [12]. This correlation phenomenon has interesting consequences also for the quantum fluctuations of the fields [18].

In Fig. 6, we illustrate the long time behavior of the adiabaton for the example discussed in Fig. 5 but for a larger decay rate. Again, the solution from Eq. (81) and from a numerical beam-propagation code are shown.

We have seen that a perturbative solution of the nonlinear Maxwell-Bloch equations for quasiadiabatic fields predicts the existence of quasiformstable solutions for a constant total Rabi frequency. The question arises, however, what is the origin of this formstable propagation? We have already seen that from the point of view of the dressed-state Bloch equations, the original system is identical to a three-level Λ-system driven by the two fields Ω and Ω_-. We now note that the first-order field equations also correspond to this situation, if $\Omega=$const, and if losses are neglected. In this case, the loss-free propagation [for $\gamma_\perp=0$ the loss term in Eq. (78) vanishes] of Ω_- can be understood as electromag-

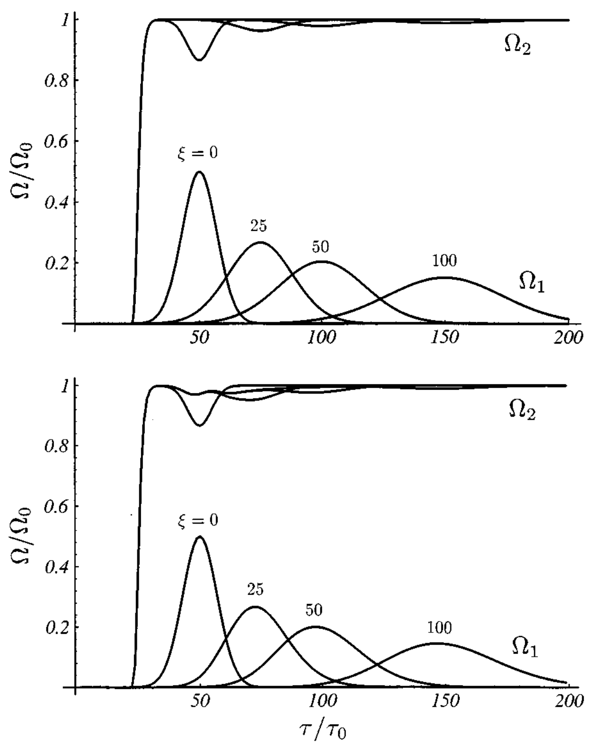

FIG. 6. Decay of adiabaton of Fig. 5 for large upper level decay $\gamma_\perp/\Omega_0 = 2.5$. Analytical results are shown in (a) and numerical results in (b).

netically induced transparency (EIT) [19] on the $|+\rangle - |-\rangle$ transition due to the strong coupling of the ''upper'' level $|+\rangle$ to the additional level $|a\rangle$. In Fig. 7, we show a typical linear susceptibility spectrum for EIT in a Λ-system with a strong constant driving field. One recognizes a transparency dip on resonance and a corresponding large linear dispersion. The absence of absorption and the linear dispersion are responsible for the formstable propagation of Ω_- with a group velocity $v < c$. The perfect transparency is associated with a population trapping in a coherent superposition of the ''lower'' levels that in our case is

$$|-2\rangle = \frac{i}{\sqrt{\Omega^2 + \Omega_-^2}} (\Omega |-\rangle - \Omega_- |a\rangle), \tag{83}$$

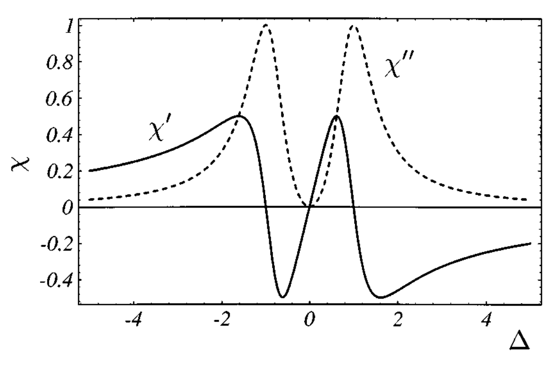

FIG. 7. Typical spectrum of real (dark line) and imaginary (dashed line) part of linear probe-field susceptibility for EIT in three-level Λ-system. The driving field Rabi frequency is equal to the upper level decay rate γ.

$|-2\rangle$ is the first-order superadiabatic dark state of the system. One easily sees that in this state, the source terms in the equations of motion of Ω and Ω_- vanish. Using the perturbative results of Sec. III, (43)–(45), and letting $\gamma \to 0$ we find

$$c_{-2} = \frac{1}{\sqrt{\Omega^2 + \Omega_-^2}} \left(\Omega + \frac{\Omega_-^2}{2\Omega} \right) \approx 1 - \mathcal{O}\left[\left(\frac{\Omega_-}{\Omega} \right)^4 \right], \tag{84}$$

which is indeed unity up to corrections of fourth order in Ω_-/Ω. Thus we can understand the formstable propagation of adiabatons as electromagnetically induced transparency in the adiabatic dressed basis.

V. SUMMARY

In the present paper, we have discussed the interaction of a pair of pulses with a resonant three-level Λ-system under conditions of quasiadiabaticity. For the description of the interaction process, we introduce the basis of adiabatic dark and bright states. This basis transformation turns the original three-level system with bichromatic fields into a different three-level system with two field couplings. Here one coupling, characterized by the total Rabi frequency of the original fields, is strong, while the other coupling is weak since it is due to nonadiabatic corrections. We show that in the presence of decay from the upper level, this nonadiabatic coupling can be treated perturbatively, which allows for an approximate analytical solution of the nonlinear Maxwell-Bloch equations.

In the first part of the paper, we present the dynamics of the coherent population transfer between the lower levels by a counterintuitive sequence of pulses. We show that in the adiabatic limit $\epsilon \to 0$, where ϵ is the small adiabaticity parameter, the nonadiabatic loss from the dark state scales like $\epsilon\gamma_\perp/\Omega$, where $\gamma_\perp$ is the decay rate of the optical coherence. In contrast to a purely Hamiltonian system, here the nonadiabatic loss is not exponentially small in $1/\epsilon$ and hence a perturbation approach is feasible. We give simple analytical expressions for the dynamical behavior of the state amplitudes as well as for the asymptotic nonadiabatic losses. For transfer times long compared to the upper level decay, the dark state population decreases smoothly and monotonically from unity to its asymptotic value. For shorter transfer times the dark state amplitude undergoes a large excursion. This behavior can be understood as a Raman-transition between the adiabatic dressed-states. When the nonadiabatic coupling is turned on, population is driven out of the dark state into the upper level. Since the transfer time is short compared to the decay from this state, the population has no time to decay and will be driven back to the dark state when the nonadiabatic coupling is turned off while the total field is still present. The perturbation approach breaks down for relatively large adiabaticity parameters, namely if $(1/\epsilon)\exp\{-2/\epsilon\}$ becomes comparable to $\gamma_\perp/\Omega$. We note that the validity condition is practically always fulfilled for $\epsilon < 0.1$.

Next we study the propagation of pulse pairs in three-level Λ-media. The adiabatic dark state is truly decoupled from the interaction with the two fields if the two pulses are in two-photon resonance and have identical shapes (i.e., are matched). The nonadiabatic coupling vanishes in this case and the problem reduces to a two-level system interacting with one field plus one uncoupled state. There are then two possibilities for a formstable loss-free propagation of the pulse pair. If all population is initially in the dark state, it will remain there and the *matched pulses* [10] do not interact with the atoms. If in the opposite case no population is initially in the dark state, a formstable propagation with very small losses is possible if the conditions for self-induced transparency [17] of the total field Ω are fulfilled. The corresponding solutions are the *simultons* found in Ref. [13].

When the relative change of the two pulse shapes is slow on a time scale set by the inverse total Rabi frequency, there is a nonvanishing but weak nonadiabatic coupling. When the pulse shapes are such that the atoms are initially in the dark state, the nonadiabatic ''field'' Ω_- propagates quasiformstable over many one-photon absorption lengths. These are the *adiabaton* solutions introduced in Ref. [7]. The physical origin of the quasiformstable propagation of adiabatons is electromagnetically induced transparency [19] in adiabatic dressed states. The (positive) linear dispersion associated with the induced transparency results in a group velocity for Ω_- that can be much less than c [20]. Since we are dealing with pulses, $\Omega_-(t)$ is not monochromatic. Its off-resonant components experience an absorption that increases quadratically with the detuning. This absorption process eventually leads to the decay of the adiabatons leaving the pulses in a configuration with matched pulse shapes.

In conlcusion, we show that the adiabatic dressed state picture is an appropriate tool to obtain analytic results for the interaction of time dependent fields with three-level atoms under quasiadiabatic conditions. It also provides simple explanations for several quasiadiabatic phenomena, such as the coherent population transfer and its limitations or the formstable propagation of matched pulses, simultons, and adiabatons.

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