

# Nonadiabatic dynamics in the dark subspace of a multilevel stimulated Raman adiabatic passage process

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In this paper we consider a generalization of the three-level stimulated Raman adiabatic-passage process (STIRAP). In our scheme, there are  $N-1$  ground levels and one excited level. The ground levels are coupled with laser pulses through the excited level at multiphoton resonance. The ultimate aim of this scheme is to create coherent-superposition states on the ground levels. In a previous paper [J. Mod. Opt. (to be published), special issue on quantum interference] we have considered this problem from the optimal-control point of view. Here we apply a different approach: We reconsider the adiabatic approximation, which is commonly utilized to describing the STIRAP process. It is shown that in our case, in the adiabatic limit, the dark and bright subspaces of the Hamiltonian are decoupled; however, the nonadiabatic corrections influence significantly the dynamics in the dark subspace. An analytic solution is presented for the case of a five-level system. Moreover, we consider some special examples for the pulse sequences that effect prescribed final superposition states. The robustness of the scheme is studied, and the extension of our scheme is also considered.

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## I. INTRODUCTION

Adiabatic methods have been widely used in various fields of atomic and molecular physics. From the well-known applications of the Landau-Zener model, the approach has recently acquired topical interest in the form of stimulated Raman adiabatic passage (STIRAP). This has opened up new prospects in coherent laser control of atomic and molecular processes; for recent review see Refs. [1,2]. Most recently even cavity quantum electrodynamics (QED) has utilized adiabatic-transfer properties to achieve desired transfer of coherence [3–5].

In addition to straightforward population transfer, the STIRAP has been applied to the problem of manipulating and creating coherent state superpositions. Such superpositions are the desired initial states for many modern quantum applications including information processing and communication. The original STIRAP process has thus been utilized to create coherent superpositions in three- and four-level systems [6–9] and to prepare  $N$ -component maximally coherent-superposition states [10]. In extending the scheme to multilevel systems, one couples the atomic-energy levels in such a way that each one is connected to at most two other ones. Population transfer in a multistate chain has been studied theoretically [11,12] and also experimentally [13,14].

In most cases, the adiabatic transfers utilize the eigenstate corresponding to zero eigenenergy. If the system is prepared in this state at the initial time, it will remain there during the time evolution as long as adiabaticity prevails. If the adiabatic state is arranged to go over into the desired state at the final time, the process effects a smooth and efficient transfer between the states. The adiabaticity of the process guarantees its robustness with respect to fluctuations in the parameters

of the couplings. In the limit of ideal adiabatic transfer, the other eigenstates are not invoked in the evolution and their population remains zero throughout the whole time. The nonadiabatic corrections tend to involve these states, but the magnitudes of such couplings are supposed to be small compared with their energy separation from the adiabatic states.

However, when many levels get involved in the dynamics, the subspace with zero eigenvalues will contain several basis vectors. Then the nonadiabatic corrections that couple these cannot be neglected, because they effect transfer between the degenerate states of the zero-eigenvalue subspace; see, e.g., [7]. Still, the nonadiabatic coupling to states with nonzero eigenvalues does not have to be included. They are thus decoupled from the time evolution in the adiabatic subspace and, in the adiabatic limit, there appears an effective reduction of the full dynamics to the reduced adiabatic subspace where the eigenenergies are degenerate (at zero) and the couplings are the nonadiabatic corrections.

In this situation we thus see a clear example of the dynamical separation of different time scales. The states with nonzero eigenvalues carry out their evolution in a simple manner, whereas the adiabatic subspace offers a reduced dynamics occurring at the rate determined basically by the time scale of the time dependence of the Hamiltonian. In this subspace we have an effective Hamiltonian that can be investigated on its own right and the associated dynamic development in the original system can be determined afterwards. When the STIRAP method is applied to many-level systems, we encounter exactly this situation. The aim of the present paper is to continue our project with multilevel STIRAP to investigate the role of dynamics in the adiabatic subspace and its manifestations in the original system.

In our previous paper [15], we have studied a generalization of the three-level STIRAP; there we considered  $N-1$  lower-lying levels and one excited level. The lower-lying levels are coupled with resonant laser pulses through the excited level. The ultimate aim of this scheme is to create an arbitrary coherent-superposition state. It has been shown that this

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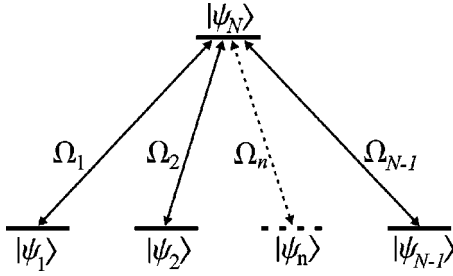


FIG. 1. Level scheme and couplings for the  $N$ -level STIRAP. There are  $N-1$  ground states and one excited state. Initially, only the state  $|\psi_1\rangle$  is populated, the others are empty. The aim is to create a coherent-superposition state of the ground levels.

model has a degenerate eigenvalue spectrum that yields a multidimensional dark subspace. A numerical approach, based on optimal control theory, has been applied to find the optimal pulse sequences that create a prescribed final superposition state. From the results of the numerical optimization it has been concluded that the transfer process occurs in the dark subspace. The robustness of the scheme has also been demonstrated.

The motivation of the present paper is twofold. On one hand, we provide an analytic solution for the population-transfer process in the multilevel atomic system, which in our previous work [15] was studied from the optimal-control point of view. On the other hand we show a rare example of nonadiabatic dynamics that is exactly solvable. In Refs. [16–18] the adiabatic corrections for the three-level STIRAP were studied. The importance of the nonadiabatic couplings was revealed for the “tripod” system [7] and for the “loop STIRAP” [19] as well. In our case, the adiabatic corrections prove to play an essential role in governing the dynamics; they determine the evolution of the system in the dark subspace. In this spirit the adiabaticity condition is carefully defined for our multilevel system and it is shown that in the adiabatic limit the population transfer takes place in the dark subspace. For a five-level system an explicit analytic solution is derived. The robustness of the transfer process is discussed and particular solutions for some special cases are obtained.

The organization of the paper is as follows. In Sec. II the physical model of our multilevel STIRAP is presented and the corresponding Hamiltonian is introduced. The eigensystem of the Hamiltonian is calculated and a condition for adiabatic evolution is introduced. In Sec. III an analytic solution for the transfer process is derived for a five-level system. Time evolution in the dark subspace is analyzed. The properties of the solution are illustrated through some special examples. We conclude the paper in Sec. IV.

## II. DEGENERATE STIRAP

The generalized STIRAP is displayed in Fig. 1:  $N-1$  atomic levels are coupled via a single atomic level with resonant laser pulses. In the rotating-wave approximation the Schrödinger equation of this system reads

$$\frac{d}{dt}\mathbf{C}(t) = -i\mathbf{H}(t)\mathbf{C}(t), \quad (1)$$

where the elements of the vector  $\mathbf{C}(t)$  correspond to the probability amplitudes associated with the atomic levels and the Hamiltonian is given by

$$\mathbf{H}(t) = \frac{1}{2} \begin{bmatrix} 0 & \cdots & \cdots & 0 & \Omega_1(t) \\ 0 & \cdots & \cdots & 0 & \Omega_2(t) \\ 0 & \cdots & \cdots & 0 & \vdots \\ \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & \cdots & \cdots & 0 & \Omega_{N-1}(t) \\ \Omega_1^*(t) & \Omega_2^*(t) & \cdots & \Omega_{N-1}^*(t) & 0 \end{bmatrix}. \quad (2)$$

In an atomic-superposition state, the individual atomic levels have definite phases. These phases can be adjusted by choosing the fixed phases of the laser pulses appropriately. However, they can be absorbed in the definitions of the atomic-basis states

$$|\tilde{\psi}_l\rangle = \exp(i\eta_l)|\psi_l\rangle \quad (1 \leq l \leq N-1) \quad (3)$$

in such a way that from now on it is assumed that all the field amplitudes  $\Omega_n$  are real.

In the following we will need the adiabatic states  $\chi_k(t)$ , which are defined as the instantaneous eigenstates of the Hamiltonian  $\mathbf{H}(t)$  in Eq. (2). The diagonalization of the Hamiltonian (2) is a special case of a more general system, which has been studied in [20]. The eigenvalue spectrum is highly degenerate,

$$E_0 = 0 \quad (\text{multiplicity } N-2)$$

$$E_{\pm} = \pm \frac{1}{2} \sqrt{\sum_{n=1}^{N-1} |\Omega_n|^2} = \pm \frac{1}{2} \Omega. \quad (4)$$

The Hilbert space in the adiabatic basis is decomposed into two subspaces: the eigenstates  $\chi_k(t)$  ( $k=1, \dots, N-2$ ), which belong to the zero eigenenergy are dark states, i.e., they do not involve the excited state  $|\psi_N\rangle$ . The dark states span an  $(N-2)$ -dimensional subspace. They are given by

$$\chi_1 = \frac{1}{\Theta_2} \begin{bmatrix} \Omega_2 \\ -\Omega_1 \\ 0 \\ 0 \\ \vdots \end{bmatrix}, \quad \chi_2 = \frac{1}{\Theta_2\Theta_3} \begin{bmatrix} \Omega_1\Omega_3 \\ \Omega_2\Omega_3 \\ -\Theta_2^2 \\ 0 \\ \vdots \end{bmatrix}, \quad (5)$$

$$\dots, \quad \chi_{N-2} = \frac{1}{\Theta_{N-2}\Theta_{N-1}} \begin{bmatrix} \Omega_1\Omega_{N-1} \\ \Omega_2\Omega_{N-1} \\ \vdots \\ -\Theta_{N-2}^2 \\ 0 \end{bmatrix},$$

where

$$\Theta_k = \sqrt{\sum_{n=1}^k \Omega_n^2}. \quad (6)$$

The two-dimensional bright subspace is spanned by the eigenstates

$$\chi_{\pm} = \frac{1}{\sqrt{2}\Theta_{N-1}} \begin{bmatrix} \Omega_1 \\ \Omega_2 \\ \vdots \\ \Omega_{N-1} \\ \pm \Theta_{N-1} \end{bmatrix}, \quad (7)$$

which belong to the eigenvalues  $E_{\pm}$  in Eq. (4). They include the excited state  $|\psi_N\rangle$ , hence the system can emit a photon when it is in these states. This is the origin of their name, “bright states.” The time dependence of  $\Omega_n(t)$  and  $\chi_l(t)$  is suppressed in these equations for brevity. In the new time-dependent basis  $\{\chi_1(t), \dots, \chi_{N-2}(t), \chi_{\pm}(t)\}$  the Schrödinger equation (1) has the form

$$\frac{d}{dt}\mathbf{B}(t) = -i\tilde{\mathbf{H}}(t)\mathbf{B}(t), \quad (8)$$

where the state vector  $\mathbf{C}(t)$  is transformed by the unitary operator

$$\mathbf{U}(t) = [\chi_1(t), \dots, \chi_{N-2}(t), \chi_+(t), \chi_-(t)] \quad (9)$$

according to

$$\mathbf{B}(t) = \mathbf{U}(t)^{-1}\mathbf{C}(t). \quad (10)$$

The transformed Hamiltonian  $\tilde{\mathbf{H}}(t)$  reads

$$\tilde{\mathbf{H}}(t) = \mathbf{U}(t)^{-1}\mathbf{H}(t)\mathbf{U}(t) + i\dot{\mathbf{U}}(t)^{-1}\mathbf{U}(t). \quad (11)$$

The first term on the right-hand side (rhs) is a zero matrix except for two nonzero elements on the diagonal in the lower right-hand corner. These two elements are the two nonzero eigenenergies  $\pm\Omega/2$  of the Hamiltonian  $\mathbf{H}(t)$  in Eq. (2). The second term is a matrix that has zero diagonal and nonzero imaginary off-diagonal elements. The off-diagonal elements are the nonadiabatic couplings in the time-dependent coordinate system. Its form allows an interpretation in terms of a gauge potential, which has made it possible to connect the associated time evolution to a topological phase as introduced by Berry; a detailed discussion can be found in [9].

In order to understand the role of the nonadiabatic-coupling terms we recall the ordinary STIRAP process in a three-level system. There the dark subspace is one dimensional, i.e., there are only one dark state and two bright states. The off-diagonal terms in the transformed Hamiltonian couple the dark state to the two bright states. The so-called “adiabatic approximation” in this three-level system consists in neglecting the coupling terms, since they are much smaller than the nonzero eigenenergies, provided that the field amplitudes vary slowly enough with time. In our case the bright subspace is two dimensional as well, how-

ever, the dark subspace is  $N-2$  dimensional, i.e., there is an  $(N-2) \times (N-2)$  submatrix in  $\tilde{\mathbf{H}}(t)$  with zero diagonal and nonzero off-diagonal elements. In this block the off-diagonal nonadiabatic couplings cannot be neglected, since they are not small compared with the diagonal elements. We define the adiabatic approximation as follows: If the couplings between the dark and bright subspaces of the Hilbert space are small compared with the nonzero eigenenergies  $\pm\Omega/2$ ,

$$|\langle \chi_l(t) | \dot{\chi}_{\pm}(t) \rangle| \ll \frac{1}{2}\Omega, \quad l = 1 \dots N-2, \quad (12)$$

the population-transfer process is called adiabatic. In this case the dark subspace is decoupled from the bright subspace and a simplified Hamiltonian  $\tilde{\mathbf{H}}'(t)$  can be introduced,

$$\tilde{\mathbf{H}}'(t) = \begin{bmatrix} 0 & & i\Gamma(t) & 0 & 0 \\ & \ddots & & \vdots & \vdots \\ -i\Gamma^\dagger(t) & & 0 & 0 & 0 \\ 0 & \dots & 0 & +\frac{1}{2}\Omega(t) & 0 \\ 0 & \dots & 0 & 0 & -\frac{1}{2}\Omega(t) \end{bmatrix}, \quad (13)$$

where the upper-triangular matrix  $\Gamma(t)$  is defined as

$$\Gamma_{ij}(t) = \begin{cases} \sum_{k=1}^N \dot{U}_{ik}^{-1}(t)U_{kj}(t) & (i < j \leq N-2) \\ 0 & (\text{otherwise}). \end{cases} \quad (14)$$

The matrix  $\Gamma(t)$  describes the nonadiabatic coupling in the degenerate subspace. In the following sections we shall analyze in detail the impact of the nonadiabatic couplings on the population-transfer process.

We define the STIRAP process in this degenerate system as follows: transfer population from one of the ground states to several of them in such a way that the adiabaticity condition Eq. (12) is satisfied. In this way, an initial state, which resides in the dark subspace, will evolve to a superposition of the degenerate dark states. The nonadiabatic couplings mix only the dark eigenstates in the dark subspace. Their coupling to the bright states can be neglected. As a result, the simplified Hamiltonian in Eq. (13) is expected to describe the population transfer approximately correctly.

In the following sections, we address the problem of finding the field amplitudes  $\Omega_n(t)$  needed to achieve a predetermined superposition state  $\mathbf{C}(\infty)$ . As we will see, the nonadiabatic couplings play a principal role in the dynamics. An analytic approach is developed, which provides a solution for a restricted range of parameters. A subset of those superposition states that have solely dark state components can be created by this method.

### III. ANALYTIC APPROACH

In this section we derive a solution to the Schrödinger equation (8) with the simplified Hamiltonian  $\tilde{H}'(t)$ . We assume that the initial state is in the dark subspace. Therefore, in the adiabatic limit (12), it is enough to consider the time evolution in the dark subspace, so that the Schrödinger equation (8) reduces to

$$\frac{d}{dt}\tilde{\mathbf{B}}(t) = -i\mathbf{V}(t)\tilde{\mathbf{B}}(t), \quad (15)$$

where

$$\mathbf{V}(t) = \begin{bmatrix} 0 & & i\Gamma(t) \\ & \ddots & \\ -i\Gamma^\dagger(t) & & 0 \end{bmatrix} \quad (16)$$

and  $\tilde{\mathbf{B}}(t)$  is equal to  $\mathbf{B}(t)$  except for the last two elements, which are truncated out. In general, the matrix  $\mathbf{V}(t)$  consists of elements that are complicated functions of time. Therefore, one cannot hope that an analytic solution of Eq. (15) can be found for arbitrary field amplitudes  $\Omega_n(t)$  and in any dimension  $N-2$ . If the dark subspace is one-dimensional, we have the ordinary three-level STIRAP scheme. The Hamiltonian  $\mathbf{V}(t)$  is simply 0. The Schrödinger equation (15) implies that when the system is initially in a dark state, it will stay there through the entire time evolution provided that the process is adiabatic. In the case of a two-dimensional dark subspace, the “tripod” system is recovered, which has been studied in great details in Refs. [7,9]. We note that in this case the solution of Eq. (15) can be found straightforwardly for any time dependence of  $\mathbf{V}(t)$ , since we have essentially a two-level problem.

Here we are going to consider the solution of Eq. (15) in the case when  $\mathbf{V}(t)$  is a  $3 \times 3$  matrix. The solution derived will be nontrivial and it will enable us to discuss some essential effects that occur in adiabatic processes taking place in a degenerate adiabatic subspace.

Before giving the detailed derivation, we present here a simple description of the calculation carried out in detail in the Appendix B. Let us consider again the original Hamiltonian (2) for  $N=5$ ,

$$\hat{H}(t) = \frac{1}{2} \sum_{i=1}^4 [\Omega_i(t)|\psi_i\rangle\langle\psi_5| + \text{H.c.}] \quad (17)$$

Let us also assume that the Rabi frequencies  $\Omega_3(t)$  and  $\Omega_4(t)$  are proportional to each other,

$$\Omega_4(t) = \frac{1}{c}\Omega_3(t), \quad (18)$$

where  $c$  is a constant. Then, we define a coupled state as

$$|C\rangle = \frac{1}{\sqrt{1+c^2}}(c|\psi_3\rangle + |\psi_4\rangle). \quad (19)$$

The orthogonal decoupled state is given by

$$|D\rangle = \frac{1}{\sqrt{1+c^2}}(-|\psi_3\rangle + c|\psi_4\rangle), \quad (20)$$

see Ref. [21]. The Hamiltonian (17) can be rewritten in the form

$$\hat{H}(t) = \frac{1}{2}[\Omega_1(t)|\psi_1\rangle\langle\psi_5| + \Omega_2(t)|\psi_2\rangle\langle\psi_5| + \Omega_3(t)|C\rangle\langle\psi_5| + \text{H.c.}], \quad (21)$$

We see that our system reduces effectively to a four-level one, a “tripod” system, which has already been discussed in some detail in Ref. [7]. In the Appendix B we start by a systematic derivation of the solution of the Schrödinger equation (15). We will find that in order to get an exact analytic solution, we have to assume the relation (18) between the Rabi frequencies 3 and 4.

In order to obtain a more detailed view of the dynamics in the dark subspace, it will prove convenient to express the elements of  $\mathbf{V}(t)$  as functions of time-dependent polar angles. To this end we make use of the derivation described in Appendix A and express the adiabatic states in Eqs. (5) and (7) in terms of polar coordinates. The dark states read

$$\chi_1 = \begin{bmatrix} \cos \theta \\ -\sin \theta \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \chi_2 = \begin{bmatrix} \sin \theta \sin \varphi \\ \cos \theta \sin \varphi \\ -\cos \varphi \\ 0 \\ 0 \end{bmatrix}, \quad (22)$$

$$\chi_3 = \begin{bmatrix} \sin \theta \cos \varphi \sin \delta \\ \cos \theta \cos \varphi \sin \delta \\ \sin \varphi \sin \delta \\ -\cos \delta \\ 0 \end{bmatrix}$$

and the two bright states are

$$\chi_+ = \frac{1}{\sqrt{2}} \begin{bmatrix} \sin \theta \cos \varphi \cos \delta \\ \cos \theta \cos \varphi \cos \delta \\ \sin \varphi \cos \delta \\ \sin \delta \\ 1 \end{bmatrix}, \quad (23)$$

$$\chi_- = \frac{1}{\sqrt{2}} \begin{bmatrix} \sin \theta \cos \varphi \cos \delta \\ \cos \theta \cos \varphi \cos \delta \\ \sin \varphi \cos \delta \\ \sin \delta \\ -1 \end{bmatrix}$$

where

$$\tan \theta = \frac{\Omega_1}{\Omega_2}, \quad \tan \varphi = \frac{\Omega_3}{\sqrt{\Omega_1^2 + \Omega_2^2}},$$

$$\tan \delta = \frac{\Omega_4}{\sqrt{\Omega_1^2 + \Omega_2^2 + \Omega_3^2}}. \quad (24)$$

In the adiabatic basis (22,23) the matrix  $V(t)$  has the form

$$V(t) = \begin{bmatrix} 0 & -i \dot{\theta} \sin \varphi & -i \dot{\theta} \cos \varphi \sin \delta \\ i \dot{\theta} \sin \varphi & 0 & i \dot{\varphi} \sin \delta \\ i \dot{\theta} \cos \varphi \sin \delta & -i \dot{\varphi} \sin \delta & 0 \end{bmatrix}. \quad (25)$$

In Appendix B we present a solution of the Schrödinger equation (15) with the operator  $V(t)$  given by Eq. (25). The solution is valid in the special case [Eq. (B12)]

$$\sin \varphi = c \tan \delta, \quad (26)$$

where  $c$  is an arbitrary real constant. This can be satisfied by a suitable choice of the laser-pulse envelopes, which will be

discussed below. If the initial state is  $\tilde{\mathbf{B}}(-\infty) = [1, 0, 0]^T$  then at any time  $t$ , the exact solution of Eq. (15) reads [Eq. (B16)]

$$\tilde{\mathbf{B}}(t) = \begin{bmatrix} \cos \beta \\ \frac{\sqrt{c^2 + \sin^2 \varphi}}{\sqrt{c^2 + 1}} \sin \beta \\ \frac{\cos \varphi \sin \beta}{\sqrt{c^2 + 1}} \end{bmatrix}, \quad (27)$$

where

$$\beta = \int_{-\infty}^t \dot{\beta} dt',$$

$$\dot{\beta} = |\dot{\theta}| \sqrt{\frac{c^2 + 1}{c^2 + \sin^2 \varphi}} \sin \varphi. \quad (28)$$

If the condition (26) applies to our system, then it implies that the Hamiltonian  $V(t)$  in Eq. (25) takes the form

$$V(t) = \begin{bmatrix} 0 & -i \dot{\theta} \sin \varphi & -i \dot{\theta} \frac{\sin \varphi \cos \varphi}{\sqrt{c^2 + \sin^2 \varphi}} \\ i \dot{\theta} \sin \varphi & 0 & i \dot{\varphi} \frac{\sin \varphi}{\sqrt{c^2 + \sin^2 \varphi}} \\ i \dot{\theta} \frac{\sin \varphi \cos \varphi}{\sqrt{c^2 + \sin^2 \varphi}} & -i \dot{\varphi} \frac{\sin \varphi}{\sqrt{c^2 + \sin^2 \varphi}} & 0 \end{bmatrix}. \quad (29)$$

There are two limiting values of the parameter  $c$ . If  $c=0$ , the Hamiltonian  $V(t)$  describes the “loop STIRAP” [19]. Here the pump and Stokes pulses are represented by the matrix elements  $V_{12}$  and  $V_{13}$ , respectively. The matrix element  $V_{23}$  corresponds to the detuning pulse. If  $c \rightarrow \infty$  then  $V(t)$  reduces to the Hamiltonian of a two-level system. In the general case one can choose the parameter  $c$  continuously from 0 to  $\infty$ , so that these two systems are connected smoothly.

In order to proceed further, we need to evaluate  $\beta$  in Eq. (28). As a concrete example, we assume the following time dependence of the angles  $\theta$  and  $\varphi$ ,

$$\theta(t) = \bar{\theta} + \frac{\Delta \theta}{2} f(t), \quad \varphi(t) = \bar{\varphi} + \frac{\Delta \varphi}{2} f(t), \quad (30)$$

where  $f(t)$  is a continuous monotonical growing function of time such that

$$\lim_{t \rightarrow \pm \infty} f(t) = \pm 1. \quad (31)$$

The parameters  $\bar{\theta}, \Delta \theta/2$ , and  $\bar{\varphi}, \Delta \varphi/2$  are constant. By the choice (30) one obtains

$$\beta = -\frac{\Delta \theta}{\Delta \varphi} \sqrt{c^2 + 1} \left[ \arctan \left( \frac{\cos \varphi}{\sqrt{c^2 + \sin^2 \varphi}} \right) - \arctan \left( \frac{\cos \varphi_i}{\sqrt{c^2 + \sin^2 \varphi_i}} \right) \right], \quad (32)$$

where  $\varphi_i$  is the initial value of  $\varphi$  at  $t = -\infty$ .

Now we transform back the state vector  $\tilde{\mathbf{B}}(t)$  to the original bare-state basis. This can easily be done by appending two zeros to the end of  $\tilde{\mathbf{B}}(t)$  and multiplying it by the unitary matrix  $\mathbf{U}(t)$ , Eq. (9), composed of the adiabatic states (22) and (23). Finally one finds

$$\mathbf{C}(t) = \begin{bmatrix} \cos \theta \cos \beta + \frac{\sqrt{c^2+1}}{\sqrt{c^2+\sin^2\varphi}} \sin \theta \sin \beta \sin \varphi \\ -\sin \theta \cos \beta + \frac{\sqrt{c^2+1}}{\sqrt{c^2+\sin^2\varphi}} \cos \theta \sin \beta \sin \varphi \\ -\frac{c^2 \cos \varphi \sin \beta}{\sqrt{c^2+1} \sqrt{c^2+\sin^2\varphi}} \\ -\frac{c \cos \varphi \sin \beta}{\sqrt{c^2+1} \sqrt{c^2+\sin^2\varphi}} \\ 0 \end{bmatrix}. \quad (33)$$

It is important to notice that the final value of  $\mathbf{C}(t)(t \rightarrow \infty)$  does not depend on the precise form of the time dependence of the angles  $\theta$  and  $\varphi$ , only the initial and final values matter, provided that they vary in time in the same manner according to Eq. (30). We interpret this feature as robustness; a particular final state of the system can be achieved by following several different paths in the parameter space. However, the rate of change must be slow enough so that the adiabaticity condition in Eq. (12) is satisfied.

The condition (26) has its implication for the field amplitudes  $\Omega_n$  as well. The amplitudes can be represented in the following form:

$$\begin{aligned} \Omega_1 &= A \cos \varphi \sin \theta, \\ \Omega_2 &= A \cos \varphi \cos \theta, \\ \Omega_3 &= A \sin \varphi, \\ \Omega_4 &= \frac{1}{c} A \sin \varphi, \end{aligned} \quad (34)$$

which satisfy both Eqs. (24) and (26). Now we can discuss the physical meaning of the different choices of the parameter  $c$ . First we note that the amplitudes  $\Omega_3$  and  $\Omega_4$  are proportional to each other. This could be relevant in an experimental realization, since one needs essentially three distinct pulses only, even if originally we required four.

If  $c \rightarrow \infty$ , then the fourth field  $\Omega_4$  is switched off. One has a ‘‘tripod’’ system, where three ground levels are coupled via one excited level by three laser pulses [7]. We have already seen that in this case the Hamiltonian  $\mathbf{V}(t)$  describes a two-level system since the dark subspace is two dimensional. Let us assume that  $\theta_i = 0$  and  $\theta_f = \pi/2$ . It follows that the initial state is  $\mathbf{C}(-\infty) = [1, 0, 0, 0]^T$ . The final state reads

$$\mathbf{C}(\infty) = \begin{bmatrix} \sin \beta_f \sin \varphi_f \\ -\cos \beta_f \\ -\sin \beta_f \cos \varphi_f \\ 0 \\ 0 \end{bmatrix} \quad (35)$$

and

$$\beta_f = -\frac{\pi}{2(\varphi_f - \varphi_i)} (\cos \varphi_f - \cos \varphi_i). \quad (36)$$

The components  $C_1, C_2, C_3$  of  $\mathbf{C}(\infty)$  form a unit vector in a three-dimensional polar coordinate system. For a given  $\varphi_f$  and  $\beta_f$  one should find a solution of Eq. (36) for  $\varphi_i$ . That solution may or may not exist. As an example we look for the result to have equal populations on the levels  $|\psi_1\rangle$ ,  $|\psi_2\rangle$ , and  $|\psi_3\rangle$ . The angles

$$\varphi_f = \arccos \frac{1}{\sqrt{2}}, \quad \beta_f = \arccos \frac{1}{\sqrt{3}}, \quad \varphi_i = 0.5264 \quad (37)$$

offer us such a distribution. The angle  $\varphi_i$  was found by solving Eq. (36) numerically.

The other limit of the parameter  $c$  is the value zero. In practice this means that the field amplitude  $\Omega_4$  significantly exceeds the other ones. Starting from the state vector  $\mathbf{C}(-\infty)$  as in the previous paragraph at time  $t$ , one has

$$\mathbf{C}(t) = \begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}. \quad (38)$$

This is a surprising result, it emerges as follows: In the Schrödinger equation (1), the coupling between levels  $|\psi_4\rangle$  and  $|\psi_5\rangle$  dominates for large  $\Omega_4$ . This interaction can be treated as the main part of the Hamiltonian and the rest may be considered as a perturbation. The superposition states

$$|\psi_+\rangle = \frac{1}{\sqrt{2}}(|\psi_4\rangle + |\psi_5\rangle), \quad |\psi_-\rangle = \frac{1}{\sqrt{2}}(|\psi_4\rangle - |\psi_5\rangle) \quad (39)$$

oscillate at the frequencies  $\pm \Omega_4$ , which is much larger than the other Rabi frequencies. As a result, the further ground states cannot interact resonantly with the states  $|\psi_\pm\rangle$ , so that the population transfer is blocked.

In general, the choice of  $c$  depends on the state we want to create in the population-transfer process. Let us assume that the target superposition state is characterized by the population distribution  $(P_1, P_2, P_3, P_4)$ . It follows from Eq. (33) that the ratio of  $P_3$  versus  $P_4$  provides  $c^2$ . The probability  $P_2$  determines the angle  $\beta_f$ . Let the initial state be  $\mathbf{C}(-\infty) = [1, 0, 0, 0]^T$ . This condition fixes  $\theta_i = 0$  since  $\beta_i = 0$  always. Moreover, let us choose  $\theta_f = \pi/2$ . Then, the ratio of  $P_1$  versus  $P_3$  or  $P_4$  gives  $\varphi_f$ . However, the values of  $c$  and  $\varphi_f$  almost entirely determine  $\beta_f$  [Eq. (32)], which is already set. There is one more freedom, the initial angle  $\varphi_i$ . If a value of  $\varphi_i$  can be found that satisfies Eq. (32), then the target population distribution  $(P_1, P_2, P_3, P_4)$  can be obtained by the method above. If such a solution does not exist, one can still vary  $\theta_f$  and try to find such parameters that do not lead to a contradiction. In general, it is not guaranteed that there exists

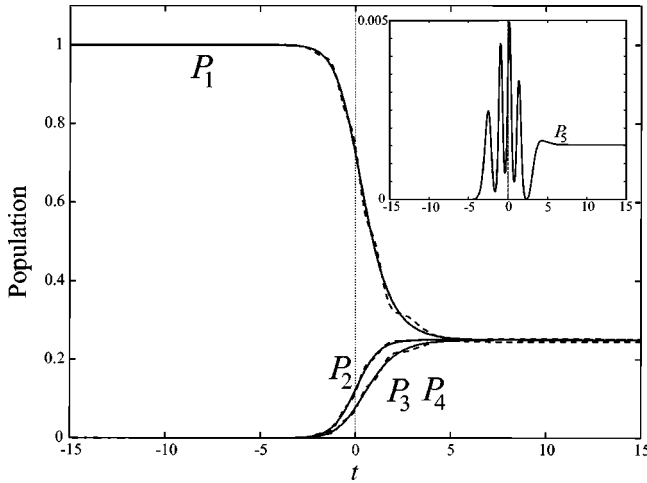


FIG. 2. Time evolution of the populations  $P_1(t) - P_5(t)$  for a five-level STIRAP. Starting from the single populated state  $|\psi_1\rangle$  the system smoothly evolves to the prescribed superposition states of equal populations on the ground levels. The solid curves show the analytic solution, whereas the dashed ones represent the result of the numerical integration of the Schrödinger equation. Time is measured in arbitrary time units.

a solution. Intuitively this is acceptable, since the four laser pulses are not independent of each other, as we have discussed above [see Eq. (34)].

To illustrate our method we consider the case of equal populations on the lower-lying states, i.e.,  $P_n = 1/4$  ( $n = 1, \dots, 4$ ). The initial state is  $\mathcal{C}(-\infty) = [1, 0, 0, 0]^T$ , as usual. The parameter  $c$  must be 1. The initial and final values of the angle  $\theta$  are set again as  $\theta_i = 0$  and  $\theta_f = \pi/2$ . The other angles are evaluated following the method described in the previous paragraph. At the end we have

$$\beta_f = \frac{\pi}{3}, \quad \varphi_i = 0.670\,22, \quad \varphi_f = 0.463\,65. \quad (40)$$

This analytic solution is obtained in the adiabatic limit. We emphasize that we did not make any special assumption about the precise shape of the function  $f(t)$  in Eq. (30). In order to compare the above result with the “exact” solution, we choose a specific form for the time dependence of the angles  $\theta$  and  $\varphi$

$$\theta(t) = \frac{\pi}{4} \left( 1 + \tanh \frac{t}{\tau} \right), \quad (41)$$

$$\varphi(t) = 0.566\,94 - 0.103\,28 \tanh \frac{t}{\tau},$$

where  $\tau$  is an arbitrary constant that must be chosen so large that the adiabaticity condition (12) is fulfilled. In Fig. 2 the time evolution of the populations  $P_1(t) - P_4(t)$  are shown. The solid lines represent the analytic solution Eq. (33) while the dashed ones result from the numerical integration of the Schrödinger equation (1). The analytical and numerical solutions agree very well. Finally, in Fig. 3 the pulses corresponding to Eq. (41) are displayed.

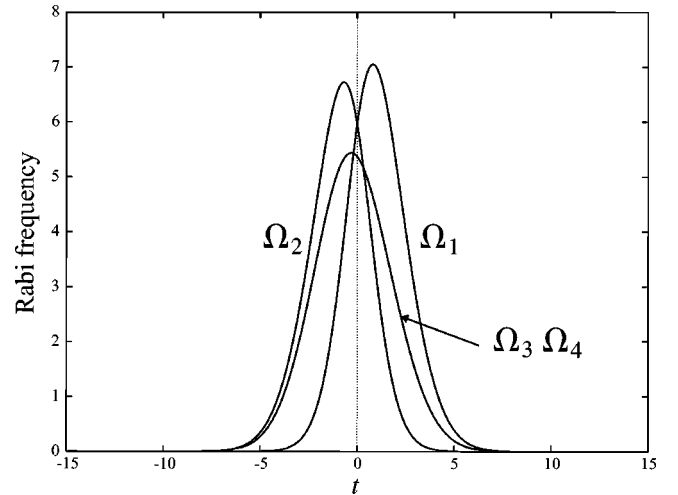


FIG. 3. Analytically calculated envelopes of the Rabi frequencies  $\Omega_1(t) - \Omega_4(t)$ , which create the equal superposition state of Fig. 2. Time and frequency are measured in arbitrary time and frequency units.

#### IV. CONCLUSIONS

We have studied an adiabatic population-transfer scheme in a multilevel system. The linking pattern of the levels resembles that of STIRAP, however, here we have several low-lying levels and a single excited level. One of our purposes has been to find such a pulse sequence, which, starting from a single populated low level, effects a prescribed final superposition state. It has been shown that the Hamiltonian describing this system has a degenerate eigensystem: It has two bright states and  $N - 2$  dark states. We have required that the excited level be populated only minimally during the population transfer. Minimal involvement of the excited level implies that the time evolution of the system takes place in the dark subspace. This condition, however, is equivalent to requiring adiabatic evolution. Here, adiabaticity means that the the dark and bright subspaces are decoupled from one another. As a result, for our purposes it is enough to restrict the description of the dynamics to the dark subspace. This has been achieved by transforming the Schrödinger equation into the time-dependent basis, which is formed by the bright and dark eigenstates of the Hamiltonian. In this representation, the size of the system can be reduced by 2, the dimension of the bright subspace. Then we have looked for analytic solutions of the reduced Schrödinger equation. In general, such a solution is impossible to obtain. However, we have worked out one for a five-level system. In order to find an analytic solution we had to impose an extra condition on the system: Two of the coupling fields have to vary in the same way with time, only their maximal amplitudes may differ. Our solution is an example for such adiabatic dynamics where the nonadiabatic couplings in the Hamiltonian have a substantial influence on determining the time evolution of the system. This is due to the special feature of the Hamiltonian that its dark subspace is degenerate. Even if we have found a solution in a restricted parameter space, it turns out that the required pulse sequences can be obtained for several interesting superposition states. We have also proved that our scheme is robust for a special choice of the time variation of the mixing

angles: They must vary with time in the same way, however, their initial and final values are freely adjustable. Their common time dependence can be arbitrary, provided the adiabaticity condition is satisfied. In that case only their initial and final values determine the final superposition state.

It is possible to generalize the method of reducing the effective dimension of the dark subspace to 2, for which case the solution is known: Assume that we have two sets of mutually coherent pulses. In both of them the pulses vary with time in the same way. These two sets of pulses play the role of Stokes pulses in a STIRAP process. It is easy to show, similarly as we have done in this work, that they define two coupled and several decoupled states. The population-transfer process populates only the two coupled states, the ratio is determined by the time integral of the coupling in the effective two-dimensional Hamiltonian, which acts on the reduced two-dimensional subspace.

We have assumed that each level pair can be addressed individually by suitably chosen laser fields. In atomic systems this implies utilization of laser tuning and polarization control. Experimentally this imposes strict restrictions on the coherence between the various pulses. In molecular systems the range of level spacings and the different selection rules make the analysis more delicate. We are, however, convinced that the efficiency and flexibility of STIRAP schemes are such that they will be of utility in a broad range of systems, which includes also the utilization of systems with many coupled levels.

#### ACKNOWLEDGMENTS

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#### APPENDIX A: DIAGONALIZATION OF THE HAMILTONIAN

In this appendix we present a simple way to find the eigenstates of the Hamiltonian (2). First we introduce polar angles  $\theta, \varphi, \dots, \xi, \zeta$  with which the field amplitudes are parametrized as

$$\begin{aligned}\Omega_1 &= \Omega \sin \theta \cos \varphi \cdots \cos \xi \cos \zeta, \\ \Omega_2 &= \Omega \cos \theta \cos \varphi \cdots \cos \xi \cos \zeta, \\ \Omega_3 &= \Omega \sin \varphi \cdots \cos \xi \cos \zeta, \\ &\vdots \\ \Omega_{N-2} &= \Omega \sin \xi \cos \zeta, \\ \Omega_{N-1} &= \Omega \sin \zeta,\end{aligned}\tag{A1}$$

where  $\Omega = \sqrt{\sum_{n=1}^{N-1} \Omega_n^2}$ . In this way the amplitudes satisfy the equations

$$\begin{aligned}\tan \theta &= \frac{\Omega_1}{\Omega_2}, \\ \tan \varphi &= \frac{\Omega_3}{\sqrt{\Omega_1^2 + \Omega_2^2}}, \\ &\vdots \\ \tan \zeta &= \frac{\Omega_{N-1}}{\sqrt{\sum_{n=1}^{N-2} \Omega_n^2}}.\end{aligned}\tag{A2}$$

Let us introduce the set of orthogonal transformations

$$\begin{aligned}\mathbf{O}_1 &= \begin{bmatrix} \cos \theta & \sin \theta & & \mathbf{0} \\ -\sin \theta & \cos \theta & & \\ & & 1 & \\ & & & \ddots \\ \mathbf{0} & & & & 1 \end{bmatrix}, \\ \mathbf{O}_2 &= \begin{bmatrix} 1 & & & \mathbf{0} \\ & -\sin \varphi & \cos \varphi & \\ & \cos \varphi & \sin \varphi & \\ & & & 1 \\ \mathbf{0} & & & & \ddots \end{bmatrix}, \\ &\vdots \\ \mathbf{O}_{N-2} &= \begin{bmatrix} 1 & & & \mathbf{0} \\ & \ddots & & \\ & & -\sin \zeta & \cos \zeta \\ & & \cos \zeta & \sin \zeta \\ \mathbf{0} & & & & 1 \end{bmatrix}, \\ &\vdots \\ \mathbf{O}_{N-1} &= \begin{bmatrix} 1 & & & \mathbf{0} \\ & \ddots & & \\ & & 1 & \\ & & & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \\ \mathbf{0} & & & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \end{bmatrix}.\end{aligned}\tag{A3}$$

The transformation  $\mathbf{O}_1$  corresponds to a pure rotation  $\mathbf{R}_1$ . The transformations  $\mathbf{O}_n (n=2, \dots, N-2)$  can be decomposed into a product of a pure rotation and a flip,



$$\mathbf{O}_n = \mathbf{F}_n \mathbf{R}_n = \begin{bmatrix} 1 & & & \mathbf{0} \\ & \ddots & & \\ & & 0 & 1 \\ & & 1 & 0 \\ \mathbf{0} & & & \ddots \end{bmatrix} \times \begin{bmatrix} 1 & & & \mathbf{0} \\ & \ddots & & \\ & & \cos \chi & \sin \chi \\ & & -\sin \chi & \cos \chi \\ \mathbf{0} & & & \ddots \end{bmatrix}. \quad (\text{A4})$$

Note that the rotations  $\mathbf{R}_n$  can be expressed in terms of generators

$$\mathbf{R}_n = \exp(-i\chi \boldsymbol{\sigma}_n),$$

$$\boldsymbol{\sigma}_n = \begin{bmatrix} \ddots & & \mathbf{0} \\ & 0 & i \\ & -i & 0 \\ \mathbf{0} & & \ddots \end{bmatrix}. \quad (\text{A5})$$

Defining the unitary transformation  $\mathbf{U}(t)$  as

$$\mathbf{U}(t) = \mathbf{R}_1 \mathbf{F}_2 \mathbf{R}_2 \cdots \mathbf{F}_{N-2} \mathbf{R}_{N-2} \mathbf{O}_{N-1}, \quad (\text{A6})$$

the Hamiltonian (2) is diagonalized through the transformation

$$\mathbf{U}^T(t) \mathbf{H}(t) \mathbf{U}(t) = \frac{1}{2} \begin{bmatrix} & & \mathbf{0} \\ & & \Omega \\ \mathbf{0} & & -\Omega \end{bmatrix}. \quad (\text{A7})$$

Since the matrix  $\mathbf{U}(t)$  is unitary, its column vectors are orthogonal to each other and so they can be chosen as the dark and bright basis vectors.

## APPENDIX B: SOLUTION OF THE SCHRÖDINGER EQUATION IN THE DARK SUBSPACE

In accordance with Eqs. (15) and (25) we are going to find the unitary time-evolution operator  $\mathcal{U}(t)$ , which satisfies

$$\frac{d}{dt} \mathcal{U}(t) = -i\mathbf{V}(t) \mathcal{U}(t). \quad (\text{B1})$$

The matrix  $\mathbf{V}(t)$  can be decomposed into a weighted sum of operators, which admit the algebra of the angular momentum operators,

$$\mathbf{V}(t) = \dot{\theta} \sin \varphi \mathbf{J}_1 + \dot{\theta} \cos \varphi \sin \delta \mathbf{J}_2 - \dot{\varphi} \sin \delta \mathbf{J}_3, \quad (\text{B2})$$

where

$$[\mathbf{J}_m, \mathbf{J}_n] = i\epsilon_{mnk} \mathbf{J}_k. \quad (\text{B3})$$

The matrices  $\mathbf{J}_k$  are defined as

$$\mathbf{J}_1 = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \mathbf{J}_2 = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix},$$

$$\mathbf{J}_3 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{bmatrix}. \quad (\text{B4})$$

In order to solve Eq. (B1) we start with the ansatz [22]

$$\mathcal{U}(t) = \mathbf{D}^j(\alpha(t), \beta(t), \gamma(t))$$

$$= \exp[-i\alpha(t)\mathbf{J}_3] \exp[-i\beta(t)\mathbf{J}_2] \exp[-i\gamma(t)\mathbf{J}_3]. \quad (\text{B5})$$

The Wigner rotation matrix  $\mathbf{D}^j(\alpha, \beta, \gamma)$  satisfies a set of partial differential equations

$$i \frac{\partial}{\partial \alpha} \mathbf{D}^j(\alpha, \beta, \gamma) = \mathbf{J}_3 \mathbf{D}^j(\alpha, \beta, \gamma),$$

$$i \frac{\partial}{\partial \beta} \mathbf{D}^j(\alpha, \beta, \gamma) = (-\mathbf{J}_1 \sin \alpha + \mathbf{J}_2 \cos \alpha) \mathbf{D}^j(\alpha, \beta, \gamma),$$

$$i \frac{\partial}{\partial \gamma} \mathbf{D}^j(\alpha, \beta, \gamma) = (\mathbf{J}_1 \cos \alpha \sin \beta + \mathbf{J}_2 \sin \alpha \sin \beta + \mathbf{J}_3 \cos \beta) \mathbf{D}^j(\alpha, \beta, \gamma). \quad (\text{B6})$$

In order to find the time-dependent angles  $\alpha$ ,  $\beta$ , and  $\gamma$  in Eq. (B5) we take the total time derivative of the matrix  $\mathbf{D}^j(\alpha, \beta, \gamma)$ ,

$$i \frac{d}{dt} \mathbf{D}^j(\alpha, \beta, \gamma) = i \left( \dot{\alpha} \frac{\partial}{\partial \alpha} + \dot{\beta} \frac{\partial}{\partial \beta} + \dot{\gamma} \frac{\partial}{\partial \gamma} \right) \mathbf{D}^j(\alpha, \beta, \gamma). \quad (\text{B7})$$

We set  $\gamma$  identically to zero. Now we compare Eqs. (B1) and (B2) with Eq. (B7) and identify the angle  $\alpha$  by

$$\tan \alpha = -\frac{\tan \varphi}{\sin \delta}. \quad (\text{B8})$$

We also define the angle  $\beta$  as

$$\beta = \int_{-\infty}^t \dot{\beta} dt',$$

$$\dot{\beta} = |\dot{\theta}| (\sin^2 \varphi + \cos^2 \varphi \sin^2 \delta)^{1/2}. \quad (\text{B9})$$

The partial derivative with respect to  $\beta$  in Eq. (B7) gives the first two terms in Eq. (B2). The third term should result from the partial derivative with respect to  $\alpha$ . However, the condition

$$\dot{\alpha} = \frac{-\sin \delta \dot{\varphi} + \sin \varphi \cos \varphi \cos \delta \dot{\delta}}{\sin^2 \varphi + \cos^2 \varphi \sin^2 \delta} = -\sin \delta \dot{\varphi} \quad (\text{B10})$$

must be fulfilled. After some algebra one arrives at a differential equation that connects  $\varphi$  and  $\delta$ ,

$$\cos \delta \sin \delta \dot{\varphi} = \tan \varphi \dot{\delta}. \quad (\text{B11})$$

The solution to this equation reads

$$\sin \varphi = c \tan \delta, \quad (\text{B12})$$

where  $c$  is an arbitrary real constant. From Eq. (B12) we express  $\sin \delta$  and insert it into Eq. (B8) to obtain

$$\tan \alpha = -\frac{\sqrt{c^2 + \sin^2 \varphi}}{\cos \varphi}. \quad (\text{B13})$$

The equation for  $\beta$ , Eq. (B9), also transforms to

$$\beta = \int_{-\infty}^t \dot{\beta} dt', \quad (\text{B14})$$

$$\dot{\beta} = |\dot{\theta}| \sqrt{\frac{c^2 + 1}{c^2 + \sin^2 \varphi}} \sin \varphi.$$

Now we are in a position to furnish explicitly the time-evolution matrix  $\mathcal{U}(t)$ . We claim that it is given by

$$\mathcal{U}(t) = \exp(-i\alpha \mathbf{J}_3) \exp(-i\beta \mathbf{J}_2) \exp(i\alpha_0 \mathbf{J}_3), \quad (\text{B15})$$

where  $\alpha$  and  $\beta$  are given by Eqs. (B13) and (B14), respectively. The last term on the rhs results from the requirement that  $\mathcal{U}(t)$  be a unit matrix at  $t = -\infty$  [since  $\beta(-\infty) = 0$  but  $\alpha_0 \equiv \alpha(-\infty) \neq 0$ , in general]. It can be readily verified that the matrix  $\mathcal{U}(t)$  in Eq. (B15) satisfies the Schrödinger equation (B1) by taking into account Eqs. (B6), (B7), (B12), (B13), and (B14). If the initial state is  $\tilde{\mathbf{B}}(-\infty) = [1, 0, 0]^T$  then

$$\tilde{\mathbf{B}}(t) = \mathcal{U}(t) \tilde{\mathbf{B}}(-\infty) = \begin{bmatrix} \cos \beta \\ \frac{\sqrt{c^2 + \sin^2 \varphi}}{\sqrt{c^2 + 1}} \sin \beta \\ \frac{\cos \varphi \sin \beta}{\sqrt{c^2 + 1}} \end{bmatrix}, \quad (\text{B16})$$

where we used the definition of  $\alpha$  from Eq. (B13).

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