Implementation of stimulated Raman adiabatic passage in degenerate systems by dimensionality reduction

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We consider the problem of the implementation of stimulated Raman adiabatic passage (STIRAP) processes in degenerate systems, with a view to be able to steer the system wave function from an arbitrary initial superposition to an arbitrary target superposition. We examine the case of an N-level atomic system consisting of $N-1$ ground states coupled to a common excited state by laser pulses. We analyze the general case of initial and final superpositions belonging to the same manifold of states, and we cover also the case in which they are nonorthogonal. We demonstrate that for a given initial and target superposition, it is always possible to choose the laser pulses so that in a transformed basis the system is reduced to an effective three-level Λ system, and standard STIRAP processes can be implemented. Our treatment leads to a simple strategy, with minimal computational complexity, which allows us to determine the laser-pulse shape required for the wanted adiabatic steering.

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I. INTRODUCTION

Destructive quantum interference allows for the control of the properties of quantum systems as well as their evolution in time. Many important features can be understood by considering a three-level atom consisting of two ground states coupled to a common excited state by two laser fields. Whenever the detuning between the two laser fields matches the ground-state splitting, the system is prepared into a superposition of the ground state which is decoupled from the laser radiation—the *dark state* [1–3]. This also allows for the control of the absorptive and dispersive properties of a medium consisting of three-level atoms [4].

Additional interesting features appear for time-dependent laser fields. In this case, the dark state becomes time dependent, and this allows one to control the quantum state of the atom via adiabatic following of the dark state—the so-called stimulated Raman adiabatic passage (STIRAP) [5]. STIRAP is not directly applicable to degenerate systems, as in this case the system may have several dark states so that the nonadiabatic coupling between them is not negligible and the adiabatic theorem does not directly apply. Several strategies have been developed for the adiabatic steering of degenerate quantum systems in different configurations. This has led to a number of schemes for the creation and manipulation of superpositions [6–12], as well as schemes for the implementation of quantum gates based on STIRAP [13–15]. Of particular relevance for the work presented here is previous work dealing with an atomic system consisting of a multiplet of degenerate ground states coupled to a common excited state by laser pulses. Solutions for the steering of arbitrary superposition were identified by using numerical optimal control techniques [8]. Analytic solutions were also found for specific configurations [9]. Analytic solutions of the nondegenerate quantum control problem in the case of arbitrary initial and final superpositions belonging to different manifold of states were given in Ref. [10].

In this work, we consider the problem of steering the atomic wave function by STIRAP in an N-level atomic system consisting of $N-1$ ground states coupled to a common excited state by laser pulses. We analyze the general case of initial and final superpositions belonging to the same manifold of states, and we cover also the case in which they are nonorthogonal. We demonstrate that for a given initial and target superposition, it is always possible to choose the laser pulses so that in a transformed basis the system is reduced to an effective three-level system, and standard STIRAP processes can be implemented. Our treatment leads to a simple strategy, with minimal computational complexity, which allows us to determine the laser-pulse shapes required for the wanted adiabatic steering.

This work is organized as follows. In Sec. II, we define the system of interest and state the problem under consideration. In Sec. III, we derive the conditions for the reduction of the system to an effective three-level Λ system. We then specify the conditions on the laser pulses for the transfer from a given initial superposition to a wanted final superposition. In Sec. IV, we demonstrate the validity of our approach with numerical simulations. Conclusions are drawn in Sec. V.

II. STATEMENT OF THE PROBLEM

We consider an N-level atomic system with $N-1$ degenerate ground states coupled to a common excited state $|N\rangle$ by laser fields of equal frequency ω, taken to be equal to the atomic transition frequency. This is the same model considered in Refs. [8] and [9] to understand the mechanism of STIRAP processes in systems with a degenerate dark-state subspace. The scheme finds direct application in the creation and manipulation of atomic systems. For $N=4$, it directly describes an atomic system with three degenerate ground states coupled to a common excited state by fields of different polarizations. The procedure identified in this work also applies, for larger N, to level schemes including nondegenerate ground-state sublevels, e.g., sublevels of different hyperfine states. In this case, each level is individually resonantly coupled to a common excited state by a laser field of appropriate frequency and polarization. This gives rise to a degenerate dark space for

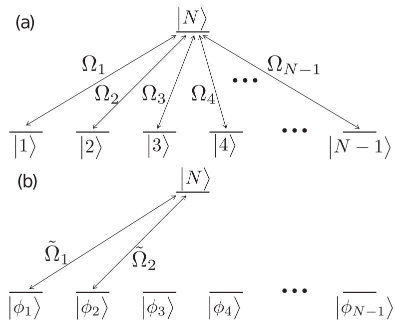

FIG. 1. N-level system consisting of $N-1$ ground states coupled to a common excited state. (a) Interaction scheme in the basis $\{|i\rangle\}$ with couplings Ω_i, as in Eq. (1). (b) Interaction scheme in the basis $\{|\phi_i\rangle\}$ with couplings $\tilde{\Omega}_i$, as in Eqs. (23).

which the procedure of implementation of STIRAP identified in this work applies.

In a frame rotating at frequency ω, the Hamiltonian in the rotating-wave approximation (RWA) can be written as

$$H = \sum_{i=1}^{N-1} \hbar\Omega_i(t)[|i\rangle\langle N| + |N\rangle\langle i|], \tag{1}$$

where $\Omega_i(t)$ is the time-dependent Rabi frequency for the transition $|i\rangle \to |N\rangle$. The Rabi frequencies are taken as real without loss of generality, as any complex phase can be reabsorbed into a redefinition of the basis states. The interaction scheme is represented in Fig. 1. The system has a subspace of superposition of ground states decoupled from the laser fields ("dark-state subspace") of dimension $N-2$ [8].

We aim to determine a set of laser pulses $\Omega_i(t)$ which drives the atomic system from an arbitrary initial ground-state superposition $|\psi_i\rangle$,

$$|\psi_i\rangle = \sum_{i=1}^{N-1} x_i|i\rangle, \tag{2}$$

to an arbitrary final target ground-state superposition $|\psi_f\rangle$,

$$|\psi_f\rangle = \sum_{i=1}^{N-1} y_i\,|i\rangle. \tag{3}$$

We restrict our analysis to the case of temporal evolution determined by the adiabatic following of a dark state, without any mixing with the excited state.

III. THEORETICAL ANALYSIS

For clarity, we consider separately the two cases of orthogonal and nonorthogonal initial and target states. We first discuss the orthogonal case in Sec. III A, while the general case is discussed in Sec. III B.

A. Case I: Orthogonal initial and target states

We consider the case of orthogonal initial and target states,

$$\langle\psi_f|\psi_i\rangle = 0. \tag{4}$$

We introduce an atomic basis for the ground-state subspace in which the first two states are the initial and the target states, $|\psi_i\rangle$ and $|\psi_f\rangle$. The basis is then completed by $N-3$ linear combinations of the original ground states, as can be obtained by the standard Gram-Schmidt orthogonalization procedure:

$$|\phi_1\rangle = |\psi_i\rangle, \tag{5}$$

$$|\phi_2\rangle = |\psi_f\rangle, \tag{6}$$

$$|\phi_3\rangle = \sum_i \alpha_3^i|i\rangle, \tag{7}$$

$$\vdots$$

$$|\phi_{N-1}\rangle = \sum_i \alpha_{N-1}^i|i\rangle, \tag{8}$$

where the coefficients α_j^i are determined by the orthogonalization procedure. For notational convenience, we rewrite this as

$$|\phi_i\rangle = \sum_j C_{ij}|j\rangle, \quad i,j = 1,\ldots,N-1. \tag{9}$$

The matrix C is orthogonal and the first two rows correspond to the coefficients x_i and y_i of the initial and the target superpositions, respectively.

In this basis, the Hamiltonian reads

$$H = \sum_{i=1}^{N-1} \tilde{\Omega}_i(t)[|\phi_i\rangle\langle N| + |N\rangle\langle\phi_i|], \tag{10}$$

where the transformed pulses are defined as

$$\tilde{\Omega}_i(t) = \sum_j C_{ij}\Omega_j(t). \tag{11}$$

Notice how the transformed Hamiltonian has the same structure as the initial one. We can thus easily find the dark states associated with (10). We first parametrize the $N-1$ laser pulses $\tilde{\Omega}_i$ in terms of a total amplitude Ω,

$$\Omega = \left(\sum_{i=1}^{N-1} \tilde{\Omega}_i^2\right)^{1/2}, \tag{12}$$

and $N-2$ angles $\theta_1,\ldots,\theta_{N-2}$ as

$$\tilde{\Omega}_1 = \Omega\sin\theta_{N-2}\sin\theta_{N-3}\cdots\sin\theta_2\sin\theta_1, \tag{13}$$

$$\tilde{\Omega}_2 = \Omega\sin\theta_{N-2}\sin\theta_{N-3}\cdots\sin\theta_2\cos\theta_1, \tag{14}$$

$$\tilde{\Omega}_3 = \Omega\sin\theta_{N-2}\sin\theta_{N-3}\cdots\cos\theta_2, \tag{15}$$

$$\vdots$$

$$\tilde{\Omega}_{N-3} = \Omega\sin\theta_{N-2}\sin\theta_{N-3}\cos\theta_{N-4}, \tag{16}$$

$$\tilde{\Omega}_{N-2} = \Omega\sin\theta_{N-2}\cos\theta_{N-3}, \tag{17}$$

$$\tilde{\Omega}_{N-1} = \Omega\cos\theta_{N-2}. \tag{18}$$

By forming the state

$$|\chi\rangle = \sum_j \tilde{\Omega}_j|\phi_j\rangle, \tag{19}$$

the $N-2$ dark states $|\chi_k\rangle$ can be easily obtained (apart from a normalization factor) as [9]

$$|\chi_k\rangle = \frac{\partial}{\partial\theta_k}|\chi\rangle. \tag{20}$$

Of particular relevance for the following is the first dark state, which reads

$$|\chi_1\rangle = \frac{\partial}{\partial\theta_1}|\chi\rangle \propto \cos\theta_1|\phi_1\rangle - \sin\theta_1|\phi_2\rangle. \tag{21}$$

In the basis $|\phi_i\rangle$ with couplings $\tilde{\Omega}_i$, it can immediately be seen that the system can be reduced by an appropriate choice of the laser pulses to a three-level Λ system consisting of the states $\{|\psi_i\rangle,|\psi_f\rangle,|N\rangle\}$, so that adiabatic transfer from $|\psi_i\rangle$ to $|\psi_f\rangle$ can be implemented.

In this subspace, the energy eigenstate corresponding to a eigenvalue $\lambda = 0$ is the first dark state, $|\Psi_0\rangle \propto \tilde{\Omega}_2(t)|\phi_1\rangle - \tilde{\Omega}_1(t)|\phi_2\rangle$. A sufficient condition to remain in this energy eigenstate throughout the evolution is (see, e.g., Eq. (6) in [16])

$$\left|\frac{\tilde{\Omega}_1(t)\dot{\tilde{\Omega}}_2(t) - \tilde{\Omega}_2(t)\dot{\tilde{\Omega}}_1(t)}{\sqrt{2}\left[\tilde{\Omega}_1^2(t) + \tilde{\Omega}_2^2(t)\right]^{3/2}}\right| \ll 1\ \forall t, \tag{22}$$

and may now serve to find optimized control functions $\tilde{\Omega}_{1/2}(t)$ with the side constraints that initially $\tilde{\Omega}_2 \gg \tilde{\Omega}_1$ and finally $\tilde{\Omega}_1 \gg \tilde{\Omega}_2$. It should be noted that fully adiabatic evolution may be a rather strict criterion, as it is for practical application only required that the final state of the system corresponds to the final dark state. In situations where the adiabatic preparation time is an issue, it may therefore be favorable to find more optimal control functions minimizing only the final occupations of the other two eigenstates.

Taking further into account typical experimental implementations, it is convenient to choose the transformed Rabi frequencies as

$$\tilde{\Omega}_1 \equiv f(t), \tag{23a}$$

$$\tilde{\Omega}_2 \equiv f(t+\tau), \tag{23b}$$

$$\tilde{\Omega}_j \equiv 0, \quad j > 2, \tag{23c}$$

where $f(t)$ has to be compatible with the conditions above. As it will be shown, this choice leads to physical laser pulses which are linear combinations of delayed pulses and are easy to implement. Condition (23c) determines the reduction of the system to an effective three-level Λ system, with the two states $|\phi_1\rangle,|\phi_2\rangle$ (i.e., $|\psi_i\rangle,|\psi_f\rangle$) coupled via a common excited state. The remaining $N-3$ states are spectator ground states not involved in the process. The reduction to an effective Λ system for an appropriate choice of laser pulses is shown in Fig. 1(b).

We can then implement a standard STIRAP process by taking a pair of pulses $f(t), f(t+\tau)$ which satisfy the standard requirements of STIRAP in terms of smoothness, duration, strength, and overlap. The system has a dark state, given by Eq. (21). The temporal dependence of the angle θ_1 is determined by the temporal dependence of $\tilde{\Omega}_1$, $\tilde{\Omega}_2$. In the specific case,

$$\tan\theta_1 = \frac{\tilde{\Omega}_1}{\tilde{\Omega}_2} = \frac{f(t)}{f(t+\tau)}, \tag{24}$$

which implies that $\theta_1(t \to -\infty) = 0$ and $\theta_1(t \to +\infty) = \pi/2$. The dark state thus has the properties

$$|\chi_1(-\infty)\rangle = |\psi_i\rangle, \tag{25}$$

$$|\chi_1(+\infty)\rangle = |\psi_f\rangle. \tag{26}$$

Therefore, as in standard STIRAP in a three-level system, adiabatic evolution along the dark state $|\chi_1\rangle$ will lead to the transfer of the system from $|\psi_i\rangle$ to $|\psi_f\rangle$.

The conditions (23) for the transformed pulses are translated for the physical laser pulses as

$$\begin{pmatrix} C_{11} & \cdots & C_{1,N-1} \\ C_{21} & \cdots & C_{2,N-1} \\ \vdots & \vdots & \vdots \\ C_{N-1,1} & \cdots & C_{N-1,N-1} \end{pmatrix} \begin{pmatrix} \Omega_1 \\ \Omega_2 \\ \vdots \\ \Omega_{N-1} \end{pmatrix} = \begin{pmatrix} f(t) \\ f(t+\tau) \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \tag{27}$$

which is a linear system easily solvable because the coefficient matrix is orthogonal. We stress that each pulse Ω_i is a linear combination of pulses $f(t)$ and $f(t+\tau)$. Specific examples will be given in Sec. IV, which is devoted to numerical solutions of the adiabatic evolution. We also notice that our derivation remains valid for more general parametrizations of transformed pulses, e.g., $\tilde{\Omega}_1(t) = \Omega_0(t)\sin[\phi(t)]$, $\tilde{\Omega}_2(t) = \Omega_0(t)\cos[\phi(t)]$, in which case the evolution would remain adiabatic whenever $|\phi'(t)/\Omega(t)| \ll 1$.

B. Case II: Nonorthogonal initial and target states

We consider the case in which the initial and target states are not orthogonal:

$$\langle\psi_f|\psi_i\rangle = \cos\alpha \neq 0, \quad \alpha < \pi/2. \tag{28}$$

We introduce a basis $\{|\phi_i\rangle\}$ $(i = 1,\ldots,N-1)$ for the ground-state subspace, with the first two basis vectors defined as

$$|\phi_1\rangle = |\psi_i\rangle, \tag{29}$$

$$|\phi_2\rangle = \frac{1}{\sin\alpha}[-|\psi_f\rangle + \cos\alpha|\psi_i\rangle], \tag{30}$$

and the remaining $N-3$ basis states determined by the standard Gram-Schmidt orthogonalization procedure, so as to complete the basis. In this basis, the final target state is expressed as

$$|\psi_f\rangle = \cos\alpha|\phi_1\rangle - \sin\alpha|\phi_2\rangle. \tag{31}$$

As for the case analyzed previously, we rewrite this as

$$|\phi_i\rangle = \sum_j C_{ij}|j\rangle, \quad i,j = 1,\ldots,N-1, \tag{32}$$

where the matrix C is orthogonal. We proceed as before and introduce the set of laser couplings $\tilde{\Omega}_i$ given by Eq. (11), and we parametrize them in terms of a total amplitude Ω given by Eq. (12) and angles θ_i given by Eq. (13). The expressions for the dark states in terms of the parameters θ_i, given by Eqs. (20) and in particular Eq. (21), remain valid.

Also in this case it is possible to reduce the system to an effective three-level Λ system and implement the adiabatic evolution from $|\psi_i\rangle$ to $|\psi_f\rangle$ as a standard STIRAP process. The required form for the transformed pulses is

$$\tilde{\Omega}_1 \equiv f(t), \tag{33}$$

$$\tilde{\Omega}_2 \equiv \frac{1}{\tan\alpha} f(t) + f(t+\tau), \tag{34}$$

$$\tilde{\Omega}_j \equiv 0, \quad j > 2\,. \tag{35}$$

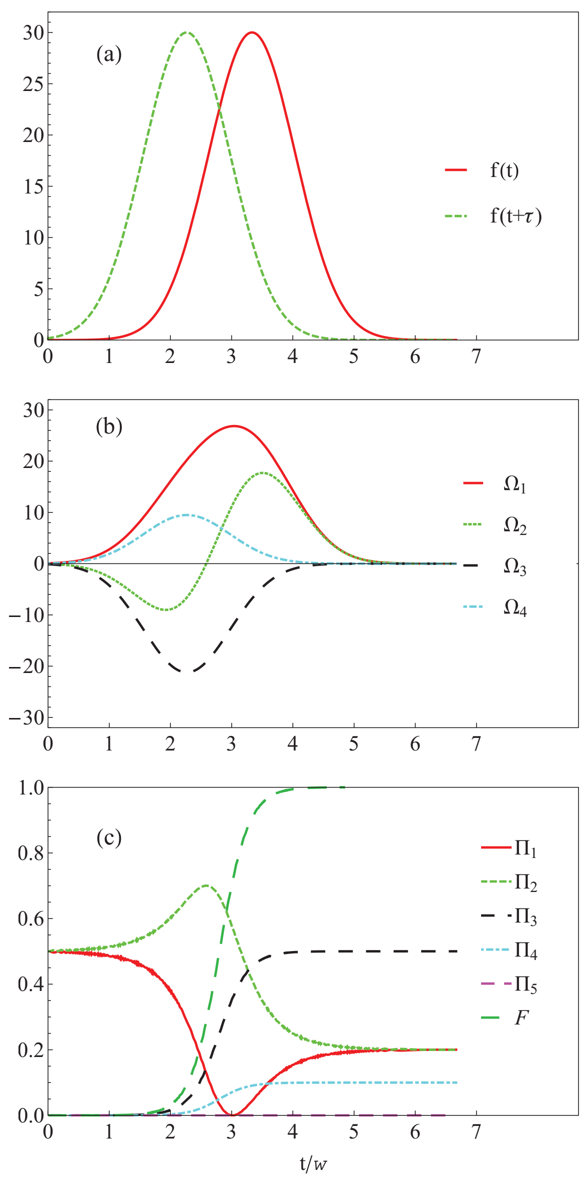

FIG. 2. (Color online) Numerical solutions for the time evolution of the five-level system starting from the initial state $|\psi_i\rangle = (|1\rangle + |2\rangle)/\sqrt{2}$. The pulses are determined, following the procedure outlined in the text, to transfer the system into the state $|\psi_f\rangle = \sqrt{2/10}|1\rangle - \sqrt{2/10}|2\rangle - \sqrt{5/10}|3\rangle + \sqrt{1/10}|4\rangle$. (a) Pulse shapes for the Rabi frequencies $\tilde{\Omega}_1$, $\tilde{\Omega}_2$ in the transformed basis $\{|\phi_i\rangle\}$. (b) Pulse shapes for the Rabi frequencies Ω_i in the atomic basis $\{|i\rangle\}$. (c) Population of the ground and excited states in the atomic basis, and fidelity F of preparation of the target state. The parameters of the simulation are $\Omega_0 = 30$, $\tau = 160$, $w = 150$, $t_0 = 500$.

As only $\tilde{\Omega}_1$, $\tilde{\Omega}_2$ are nonzero, the system is reduced to a three-level Λ system, as in the case analyzed previously. Furthermore, the specific choice of the pulse form, given by Eqs. (33) and (34), leads to the transfer of the atomic system from $|\phi_1\rangle$ to $\cos\alpha|\phi_1\rangle - \sin\alpha|\phi_2\rangle$, i.e., from $|\psi_i\rangle$ to $|\psi_f\rangle$. This can be shown by noticing that

$$\tan\theta_1(t) = \frac{\tilde{\Omega}_1}{\tilde{\Omega}_2} = \frac{f(t)}{f(t+\tau)} \to \begin{cases} 0 & t \to -\infty \\ \tan\alpha & t \to +\infty \end{cases}. \tag{36}$$

Thus, $\theta_1(t \to -\infty) = 0$ and $\theta_1(t \to +\infty) = \alpha$. Therefore, the dark state $|\chi_1\rangle$ has the properties

$$|\chi_1(-\infty)\rangle = |\psi_i\rangle, \tag{37}$$

$$|\chi_1(+\infty)\rangle = |\psi_f\rangle, \tag{38}$$

and the adiabatic following of the dark state leads to the evolution of the system from $|\psi_i\rangle$ to $|\psi_f\rangle$.

The physical laser pulses are obtained by solving the system of Eqs. (27), and again each Ω_i is a linear combination of $f(t)$ and $f(t+\tau)$.

IV. NUMERICAL ANALYSIS

In this section, we prove the validity of our approach with numerical simulations. We numerically study the time evolution of the atomic system to verify that our choice for the pulse sequence does indeed lead to adiabatic transfer from the initial to the target state. We consider a five-level system, with four ground states and one excited state. As already stressed, the procedure to identify the required pulse shape for the wanted transfer has minimal computational complexity, as it simply requires the inversion of an orthogonal matrix, which corresponds to a transposition. Thus, the same procedure can be applied to larger atomic systems, with the same coupling structure, without any computational difficulty.

For fully adiabatic evolution [cf. Eq. (22)], the evolution of the atomic system does not involve populating the atomic excited state. Thus, we can study the time evolution of the atomic system by solving the Schrödinger equation. In all numerical simulations presented here, we will take the transformed pulses $\tilde{\Omega}_1 = f(t)$, $\tilde{\Omega}_2 = f(t+\tau)$ to have Gaussian shape,

$$f(t) = \Omega_0 \exp[-(t-t_0)^2/w^2], \tag{39}$$

where t_0 is the pulse center. The pulse delay τ between the delayed pulses is chosen so as to guarantee an overlap, an essential condition for the STIRAP process. The amplitude of the pulses Ω_0 and its width w are chosen so as to guarantee the adiabaticity of the process. We notice that we chose the same amplitude Ω_0 for the pulses for simplicity. However, this is not a requirement for the STIRAP process in the effective three-level Λ system, and any combination of amplitudes for the two pulses $\tilde{\Omega}_1$, $\tilde{\Omega}_2$, such that the adiabaticity condition is satisfied, will lead to the correct implementation of the STIRAP process.

Figure 2 shows the solution of the time-dependent Schrödinger equation for the case of orthogonal initial and target states. Figure 2(a) reports the pulse shapes $f(t)$, $f(t+\tau)$ in the transformed basis, while in the initial basis the required laser pulses to obtain the wanted transfer are then determined via Eq. (27), and are reported in Fig. 2(b). We notice that

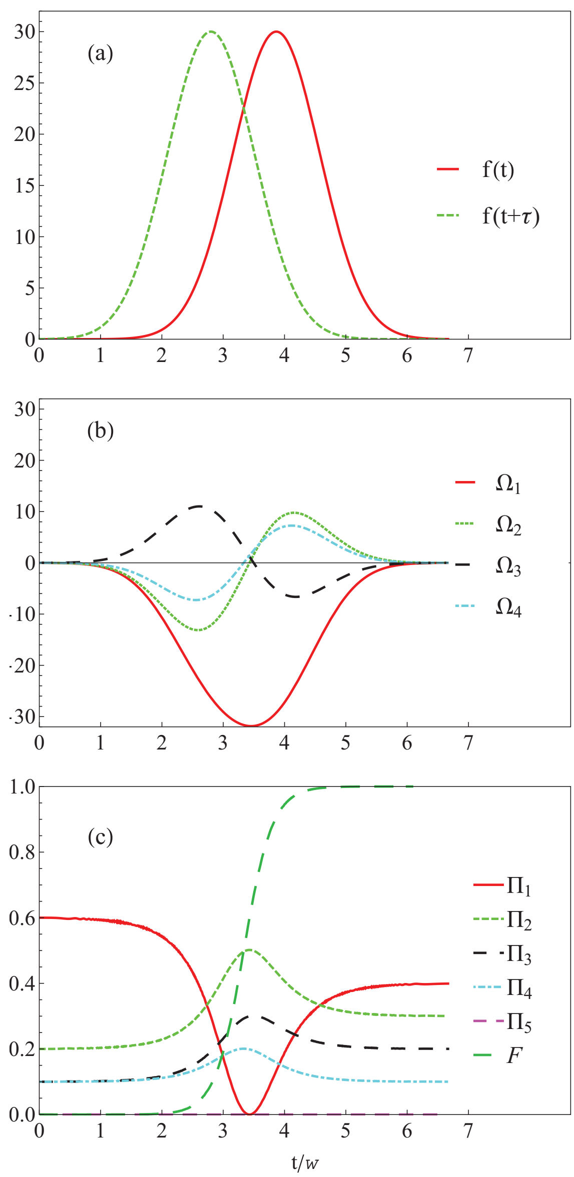

FIG. 3. (Color online) Numerical solutions for the time evolution of the five-level system starting from the initial state $|\psi_i\rangle = -\sqrt{3/5}|1\rangle + \sqrt{1/5}|2\rangle - \sqrt{1/10}|3\rangle + \sqrt{1/10}|4\rangle$. The pulses are determined, following the procedure outlined in the text, to transfer the system into the state $|\psi_f\rangle = \sqrt{2/5}|1\rangle + \sqrt{3/10}|2\rangle - \sqrt{1/5}|3\rangle + \sqrt{1/10}|4\rangle$. (a) Pulse shapes for the Rabi frequencies $\tilde{\Omega}_1$, $\tilde{\Omega}_2$ in the transformed basis $\{|\phi_i\rangle\}$. (b) Pulse shapes for the Rabi frequencies Ω_i in the bare atomic basis $\{|i\rangle\}$. (c) Population of the ground and excite states, and fidelity F of preparation of the target state. The parameters of the simulation are $\Omega_0 = 30$, $\tau = 160$, $w = 150$, $t_0 = 500$.

the required relative sign between Rabi frequencies can experimentally be easily implemented by introducing a relative phase between the corresponding electric fields. The resulting time-dependent populations Π_i $(i = 1-5)$ of the different atomic states are reported in Fig. 2(c), together with the fidelity of preparation of the wanted state, $F = |\langle\psi_f|\psi(t)\rangle|^2$, where $|\psi(t)\rangle$ describes the state of the system at time t. Our numerical results show that the fidelity approaches unity after the pulse sequence, i.e., the system is effectively prepared in the wanted state $|\psi_f\rangle$.

An analogous numerical analysis was also carried out for the case of nonorthogonal initial and target states. The procedure differs from the case analyzed previously only in the definition of the transformed laser pulses. The nonorthogonality of the initial and target superpositions require a different transformation for $\tilde{\Omega}_2$, as given by Eq. (34). Our numerical results for this case, presented in Fig. 3, confirm the validity of our approach: the process leads to a complete transfer from $|\psi_i\rangle$ to $|\psi_f\rangle$, without populating the excited state. Also in this case the amplitudes of the transformed fields were taken to be equal for simplicity. However, this is not a requirement for the STIRAP process and arbitrary amplitudes can be used, provided that they are large enough to guarantee the adiabaticity of the process.

V. CONCLUSIONS

In this work, we considered the problem of the implementation of stimulated Raman adiabatic passage processes in degenerate systems, with a view to be able to steer the system wave function from an arbitrary initial superposition to an arbitrary target superposition. We examined the case of an N-level atomic system consisting of $N-1$ ground states coupled to a common excited state by laser pulses. We analyzed the general case of initial and final superpositions belonging to the same manifold of states, and we covered also the case in which they are nonorthogonal. We demonstrated that for a given initial and target superposition, it is always possible to choose the laser pulses so that in a transformed basis the system is reduced to an effective three-level Λ system, and standard STIRAP applies. Our treatment leads to a simple strategy, with minimal computational complexity, which allows us to determine the laser-pulse shape required for the wanted adiabatic steering.

ACKNOWLEDGMENTS

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