General method for complete population transfer in degenerate systems

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A simple theoretical solution to the design of a control field that generates complete population transfer from an initial state, via N nondegenerate intermediate states, to one arbitrary member of M $(M \leqslant N)$ degenerate states is constructed. The full control field exploits an $(M+N-1)$-node null adiabatic state, created by designing the relative phases and amplitudes of the component fields that together make up the full field. The solution found is universal in the sense that it does not depend on the exact number of the unwanted degenerate states or their properties. The results obtained suggest that a class of multilevel quantum systems with degenerate states can be completely controllable, even under extremely strong constraints, e.g., never populating a Hilbert subspace that is only a few dimensions smaller than the whole Hilbert space.

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I. INTRODUCTION

Coherent control, i.e., controlling atomic and molecular processes via quantum interference effects induced by external fields, has attracted great theoretical and experimental interest [1–3]. In particular, our ability to manipulate atomic and molecular excitation and therefore drive the microscopic motion to generate an arbitrarily selected target state is one key prerequisite for quantum information processing [4] in atoms and molecules.

One formal mathematical problem associated with coherent control is determining whether or not complete controllability is achievable. If, for a given system, it can be proved that there exists a control field (possibly composed of a number of component fields) that can yield complete population transfer to an arbitrary target state within a finite time, then the system is called completely controllable. For such systems, the primary task of coherent control studies is to design methods that are conceptually as simple as possible and also experimentally feasible. If the formal complete controllability of a quantum system is not established in general, then it is of great theoretical interest to construct specific control fields that can achieve, say, complete population transfer between two particular quantum states.

Some existence theorems that establish conditions for complete controllability of a quantum system have been proved [5–7]. The Huang-Tarn-Clark theorem [5] is believed to be the strongest result, but it applies only to systems with discrete and nondegenerate states. Ramakrishna *et al.* showed that for a quantum system with a Hilbert space of dimension L, the necessary and sufficient condition for complete controllability is that the field-free Hamiltonian and the interaction Hamiltonian induced by a control field generate a Lie algebra of dimension L^2 [6]. This criterion is applicable to multilevel systems that involve degenerate states [8,9], but the required computations to generate the commutators of the Lie algebra structure can be demanding for large L, vary drastically from system to system, and provide no hint as to how a control field can be designed. As such, the possibility for complete controllability in a variety of degenerate systems is still unknown and it is widely accepted that coherent control in degenerate systems is more difficult than in nondegenerate systems [10–13].

Even more challenging is to establish the complete controllability of a quantum system subject to restrictions on the population transfer pathway along which the system may evolve. Shapiro and Brumer [14] have shown that there is often a loss of controllability if the whole Hilbert space of a quantum system is partitioned and one wishes to generate an evolution that does not pass through some particular Hilbert subspace. Solá *et al.* [15] have proposed an optimal control theory to maximize the final population in the target state and minimize a time-integral of the population in all the unwanted states. Neither of these two previous studies has provided conditions for a class of quantum systems subject to strong constraints to be completely controllable.

Encouraged by recent progress in extending the stimulated Raman adiabatic passage (STIRAP) [16] method and its variations [13,17–19,21–25] for coherent control, we address the controllability of population transfer to one of M degenerate states in a system with $N \geqslant M$ nondegenerate intermediate states by invoking a general adiabatic passage scheme. In particular, we demonstrate that, under certain conditions, it is possible to realize complete population transfer from an initial state, via N nondegenerate intermediate states, to one arbitrary member of a set of M $(M \leqslant N)$ degenerate states, without ever populating any states other than the initial state and the target state. The solution is surprisingly simple and general in the sense that realizing such a population transfer pathway may not require our knowledge of the exact number of degeneracies or the properties of the M degenerate states except for the target state. The solution can also be easily adapted for the creation of an arbitrary superposition of the M degenerate states. The results we have obtained strongly suggest that a class of multilevel quantum systems with degenerate states should be completely controllable. The results also imply that complete controllability in a class of quantum systems may still be possible even when extremely strong constraints are applied, e.g., never populating a Hilbert subspace that is only dimension-two smaller than the whole Hilbert space.

The central idea of our approach is to manipulate the component fields of the full control field to create an $(M+N-1)$-node null eigenstate of the system dressed by the fields. This multinode null eigenstate is designed to fully correlate with the initial state at early times and then evolve to the target state as the component fields that have specified relative phases and amplitudes are turned on and off in a particular order. This scheme is a significant extension of our previous adiabatic passage method for the realization of complete control of the population transfer branching ratio between two degenerate states [19]. In addition, both the present study and our previous work [19] demonstrate a powerful marriage between weak-field coherent phase control and traditional adiabatic passage techniques [16] that are insensitive to the relative phases of the control fields.

This paper is organized as follows. In Sec. II we describe our model system. We then present in Sec. III our adiabatic passage scheme for the complete population transfer from an initial state to one arbitrary member of M degenerate states. Simple computational examples that support our theoretical results are described in Sec. IV. We discuss our results and conclude this paper in Sec. V.

II. THE MODEL SYSTEM

Consider a multilevel quantum system that has a nondegenerate initial state (denoted $|0\rangle$), and M degenerate orthogonal states (denoted $|f_1\rangle, |f_2\rangle, \ldots, |f_M\rangle$). Without loss of generality we denote one arbitrary member of the M degenerate states that we wish to transfer population to as $|f_M\rangle$. We assume that the M degenerate states cannot be directly coupled with the initial state and that there exist N nondegenerate intermediate states (denoted $|i_1\rangle, |i_2\rangle, \ldots, |i_N\rangle$) that can be coupled with the initial state and some of the M degenerate states. The question under consideration is the following: under what conditions can we, in principle, realize 100% population transfer from state $|0\rangle$ to state $|f_M\rangle$ without ever populating the intermediate states or any other degenerate states. From a mathematical point of view, this question is formidable because (i) the extent of controllability in degenerate systems without constraints is not yet resolved and (ii) the extent of controllability in systems subject to strong constraints has barely been studied.

One needs N laser fields to resonantly couple the initial state to the N intermediate states, with the corresponding Rabi frequencies denoted by $2\Omega_{P_1}, 2\Omega_{P_2}, \ldots, 2\Omega_{P_N}$, and additional N laser fields to resonantly couple the intermediate states with the M degenerate states, with the Rabi frequency associated with state $|i_k\rangle$ $(k=1,2,\ldots,N)$ and state $|f_j\rangle$ $(j=1,2,\ldots,M)$ represented by $2\Omega_{S_{kj}}$. A schematic diagram of the energy levels and the corresponding parameters for the Rabi frequencies is shown in Fig. 1. As in the simplest version of STIRAP [16], we assume that the laser fields are Gaussian pulses and that they are counterintuitively ordered. Specifically, the electric field E_{P_k} that couples state $|0\rangle$ with state $|i_k\rangle$ is given by

$$E_{P_k}(t) = \tilde{E}_{P_k}\cos(\omega_{P_k}t + \phi_{P_k})\exp[-(t-T)^2/T^2], \tag{1}$$

and the electric field E_{S_k} that couples state $|i_k\rangle$ with the M degenerate states is given by

$$E_{S_k}(t) = \tilde{E}_{S_k}\cos(\omega_{S_k}t + \phi_{S_k})\exp[-t^2/T^2]. \tag{2}$$

Here t is the time variable, T is the pulse width, $\tilde{E}_{P_k}$ and $\tilde{E}_{S_k}$ are the peak amplitudes of the electric fields, ω_{P_k} and ω_{S_k} are the laser carrier frequencies, and ϕ_{P_k} and ϕ_{S_k} are the laser phases. Note that $E_{P_k}(t)$ is delayed by T relative to $E_{S_k}(t)$. The Rabi frequencies are then given by

$$2\Omega_{P_k} = 2\tilde{\Omega}_{P_k}\exp[-(t-T)^2/T^2], \tag{3}$$

$$2\Omega_{S_{kj}} = 2\tilde{\Omega}_{S_{kj}}\exp[-t^2/T^2], \tag{4}$$

where the peak Rabi frequencies $2\tilde{\Omega}_{P_k}$ and $2\tilde{\Omega}_{S_{kj}}$ are

$$2\tilde{\Omega}_{P_k} = \mu_{0k}\tilde{E}_{P_k}\exp(i\phi_{P_k}), \tag{5}$$

$$2\tilde{\Omega}_{S_{kj}} = \mu_{kj}\tilde{E}_{S_k}\exp(i\phi_{S_k}). \tag{6}$$

Here $\mu_{0k}\equiv\langle 0|\hat{\mu}|i_k\rangle$ and $\mu_{kj}\equiv\langle i_k|\hat{\mu}|f_j\rangle$ are the transition dipole moments.

In the rotating-wave approximation and in the interaction representation, the Hamiltonian of the system plus the component fields of the full control field is given by

$$\mathbf{H} = \begin{bmatrix} 0 & \Omega_{P_1} & \Omega_{P_2} & \cdots & \Omega_{P_N} & 0 & 0 & \cdots & 0 \\ \Omega^*_{P_1} & 0 & 0 & \cdots & 0 & \Omega_{S_{11}} & \Omega_{S_{12}} & \cdots & \Omega_{S_{1M}} \\ \Omega^*_{P_2} & 0 & 0 & \cdots & 0 & \Omega_{S_{21}} & \Omega_{S_{22}} & \cdots & \Omega_{S_{2M}} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \Omega^*_{P_N} & 0 & 0 & \cdots & 0 & \Omega_{S_{N1}} & \Omega_{S_{N2}} & \cdots & \Omega_{S_{NM}} \\ 0 & \Omega^*_{S_{11}} & \Omega^*_{S_{21}} & \cdots & \Omega^*_{S_{N1}} & 0 & 0 & \cdots & 0 \\ 0 & \Omega^*_{S_{12}} & \Omega^*_{S_{22}} & \cdots & \Omega^*_{S_{N2}} & 0 & 0 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & \Omega^*_{S_{1M}} & \Omega^*_{S_{2M}} & \cdots & \Omega^*_{S_{NM}} & 0 & 0 & \cdots & 0 \end{bmatrix} \tag{7}$$

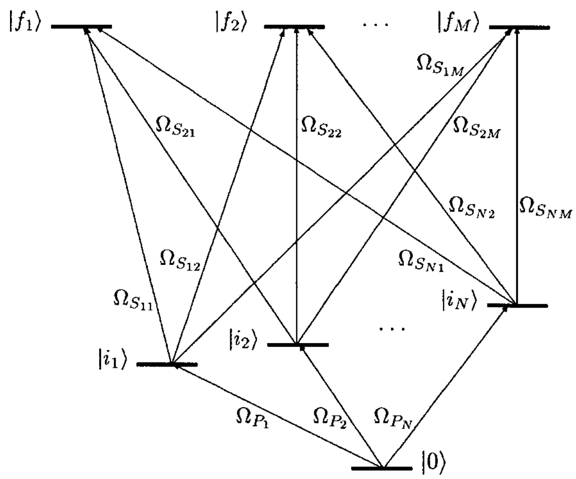

FIG. 1. A schematic diagram of an adiabatic passage scheme for the realization of complete population transfer from an initial state $|0\rangle$, via N nondegenerate intermediate states $|i_1\rangle, |i_2\rangle, \ldots, |i_N\rangle$, to state $|f_M\rangle$ that is degenerate with other $(M-1)$ states $|f_1\rangle, |f_2\rangle, \ldots, |f_{M-1}\rangle$. Under certain conditions 100% population can be transferred from state $|0\rangle$ to state $|f_M\rangle$ without ever populating any of the intermediate states or any of the undesired degenerate states.

As suggested by this Hamiltonian, we have assumed that the N nondegenerate intermediate states will not be coupled to one another by the external fields.

III. ADIABATIC PASSAGE IN DEGENERATE SYSTEMS

Our previous work on complete control of the population transfer from an initial state to one of two degenerate target states [19] suggests that the desired complete population transfer from the initial state $|0\rangle$ to the target state $|f_M\rangle$ may be achieved by using an eigenstate dressed by the external fields that has nodes on all the states that we do not wish to transfer population to. If, by choosing the component fields of the full control field so that a multinode dressed eigenstate is created that correlates first with state $|0\rangle$ and then with the target state $|f_M\rangle$ as the fields evolve in time, then in the adiabatic limit, i.e., in cases where the dynamics of the system can adiabatically follow the evolution of the multinode dressed eigenstate, this control field will generate complete population transfer from state $|0\rangle$ to state $|f_M\rangle$.

In particular, we examine here the existence and properties of the null eigenstate (denoted as $|\Lambda\rangle$) of $\mathbf{H}$. By definition, one has

$$\mathbf{H}|\Lambda\rangle = 0. \tag{8}$$

In the same representation of $\mathbf{H}$, $|\Lambda\rangle$ can be written in terms of its $(1+N+M)$ components, i.e., $(z_0; x_1, x_2, \ldots, x_N; y_1, y_2, \ldots, y_M)^T$. Then Eqs. (7) and (8) give

$$\sum_{k=1}^{N} \Omega_{P_k} x_k = 0, \tag{9}$$

$$\begin{pmatrix} \Omega^*_{S_{11}} & \Omega^*_{S_{21}} & \cdots & \Omega^*_{S_{N1}} \\ \Omega^*_{S_{12}} & \Omega^*_{S_{22}} & \cdots & \Omega^*_{S_{N2}} \\ \vdots & \vdots & \vdots & \vdots \\ \Omega^*_{S_{1M}} & \Omega^*_{S_{2M}} & \cdots & \Omega^*_{S_{NM}} \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_N \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \tag{10}$$

and

$$\begin{pmatrix} \Omega_{S_{11}} & \Omega_{S_{12}} & \cdots & \Omega_{S_{1M}} \\ \Omega_{S_{21}} & \Omega_{S_{22}} & \cdots & \Omega_{S_{2M}} \\ \vdots & \vdots & \vdots & \vdots \\ \Omega_{S_{N1}} & \Omega_{S_{N2}} & \cdots & \Omega_{S_{NM}} \end{pmatrix} \begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_M \end{pmatrix} = -z_0 \begin{pmatrix} \Omega^*_{P_1} \\ \Omega^*_{P_2} \\ \vdots \\ \Omega^*_{P_N} \end{pmatrix}. \tag{11}$$

Below we examine the solution to Eqs. (9)–(11) in three different cases, specifically, $M=N$, $M<N$, and $M>N$.

A. The $M=N$ case

Let $M=N$. We denote the $M\times M$ matrix S_{kj} $(k=1,2,\ldots,M, j=1,2,\ldots,M)$ by $\mathbf{S}$,

$$\mathbf{S} \equiv \begin{bmatrix} \Omega_{S_{11}} & \Omega_{S_{12}} & \cdots & \Omega_{S_{1M}} \\ \Omega_{S_{21}} & \Omega_{S_{22}} & \cdots & \Omega_{S_{2M}} \\ \vdots & \vdots & \vdots & \vdots \\ \Omega_{S_{M1}} & \Omega_{S_{M2}} & \cdots & \Omega_{S_{MM}} \end{bmatrix}. \tag{12}$$

Clearly, if

$$\det(\mathbf{S}) \neq 0, \tag{13}$$

then Eq. (10) has only one solution

$$x_1 = x_2 = \cdots = x_N = 0. \tag{14}$$

With this solution for x_k $(k=1,2,\ldots,N)$, Eq. (9) is automatically satisfied. Further, under condition (13), one obtains the only solution to Eq. (11), namely

$$\begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_M \end{pmatrix} = -z_0 \mathbf{S}^{-1} \begin{pmatrix} \Omega^*_{P_1} \\ \Omega^*_{P_2} \\ \vdots \\ \Omega^*_{P_N} \end{pmatrix}, \tag{15}$$

where the value of z_0 is determined by the normalization requirement and $\mathbf{S}^{-1}$ denotes the inverse of $\mathbf{S}$. Note that $\mathbf{S}^{-1}$ can be explicitly written as follows:

$$\mathbf{S}^{-1} = \frac{1}{\det(\mathbf{S})} \begin{pmatrix} A_{11} & A_{21} & \cdots & A_{M1} \\ A_{12} & A_{22} & \cdots & A_{M2} \\ \vdots & \vdots & \vdots & \vdots \\ A_{1M} & A_{2M} & \cdots & A_{MM} \end{pmatrix}, \tag{16}$$

where A_{kj} is the cofactor of $\Omega_{S_{kj}}$,

$$A_{kj} = (-1)^{k+j}\Delta_{kj}, \tag{17}$$

with Δ_{kj} being the complementary minor of $\Omega_{S_{kj}}$, i.e., the determinant of an $(M-1)\times(M-1)$ submatrix of $\mathbf{S}$ obtained by deleting the kth row and jth column of $\mathbf{S}$.

It should be pointed out that whether or not det($\mathbf{S}$) can be zero is a time-independent property inherent to the system itself. Using Eqs. (4) and (6) one obtains

$$\det(\mathbf{S}) = \left(\frac{1}{2}\right)^M \left[\prod_{k=1}^{M} \tilde{E}_{S_k} \exp(i\phi_{S_k})\right] \exp(-Mt^2/T^2) \times \det\begin{pmatrix} \mu_{11} & \mu_{12} & \cdots & \mu_{1M} \\ \mu_{21} & \mu_{22} & \cdots & \mu_{2M} \\ \vdots & \vdots & \vdots & \vdots \\ \mu_{M1} & \mu_{M2} & \cdots & \mu_{MM} \end{pmatrix}. \tag{18}$$

Thus, provided that

$$\det\begin{pmatrix} \mu_{11} & \mu_{12} & \cdots & \mu_{1M} \\ \mu_{21} & \mu_{22} & \cdots & \mu_{2M} \\ \vdots & \vdots & \vdots & \vdots \\ \mu_{M1} & \mu_{M2} & \cdots & \mu_{MM} \end{pmatrix} \neq 0, \tag{19}$$

det($\mathbf{S}$) will be nonzero at all times. Unless there are some particular system symmetries, it is rare that condition (19) will be violated.

Thus, under condition (19), $\mathbf{H}$ has only one null eigenvector with N nodes on the intermediate states and its other $(M+1)$ components $z_0, y_1, y_2, \ldots, y_M$ satisfying Eq. (15). Due to the ordering of the laser fields [see Eqs. (3) and (4)], this null eigenvector initially correlates with state $|0\rangle$ and then correlates with a superposition of the M degenerate states, $\Sigma_{k=1}^{M} y_k|f_k\rangle$. So if the system remains in this null eigenstate, the population will be transferred from state $|0\rangle$ to state $\Sigma_{k=1}^{M} y_k|f_k\rangle$ without populating any of the N intermediate states as time passes. We must still examine whether or not it is possible to realize complete population transfer to one arbitrary member of the M degenerate states without ever populating the other $(M-1)$ degenerate states, and if yes, under what conditions.

Our consideration is based on the relation

$$A_{1k}\Omega_{S_{1M}} + A_{2k}\Omega_{S_{2M}} + \cdots + A_{Nk}\Omega_{S_{NM}} = \delta_{kM}\det(\mathbf{S}), \tag{20}$$

which leads to

$$\mathbf{S}^{-1}\begin{pmatrix} \Omega_{S_{1M}} \\ \Omega_{S_{2M}} \\ \vdots \\ \Omega_{S_{NM}} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ \vdots \\ 1 \end{pmatrix}. \tag{21}$$

Hence if the laser fields are designed such that

$$\begin{pmatrix} \Omega_{P_1}^* \\ \Omega_{P_2}^* \\ \vdots \\ \Omega_{P_N}^* \end{pmatrix} = \xi(t)\begin{pmatrix} \Omega_{S_{1M}} \\ \Omega_{S_{2M}} \\ \vdots \\ \Omega_{S_{NM}} \end{pmatrix}, \tag{22}$$

where $\xi(t)$ is any function of time, then Eqs. (15), (21), and (22) yield

$$\begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_M \end{pmatrix} = -z_0\xi(t)\begin{pmatrix} 0 \\ 0 \\ \vdots \\ 1 \end{pmatrix}. \tag{23}$$

That is, by manipulating the component fields of the total control field we can guarantee that the only null eigenvector of $\mathbf{H}$ will have additional $(M-1)$ nodes on states $|f_1\rangle, |f_2\rangle, \ldots, |f_{M-1}\rangle$. The total number of nodes of $|\Lambda\rangle$ is then given by $(M+N-1)$. It follows that if the dynamics of population transfer adiabatically follows the evolution of $|\Lambda\rangle$, complete population transfer from state $|0\rangle$ to one arbitrary member of the M degenerate states can be achieved without ever populating the other $(M-1)$ degenerate states or the N intermediate states.

For the specific pulse shape functions given in Eqs. (3) and (4), condition (22) is equivalent to

$$\begin{pmatrix} \tilde{\Omega}_{P_1}^* \\ \tilde{\Omega}_{P_2}^* \\ \vdots \\ \tilde{\Omega}_{P_N}^* \end{pmatrix} = \eta\begin{pmatrix} \tilde{\Omega}_{S_{1M}} \\ \tilde{\Omega}_{S_{2M}} \\ \vdots \\ \tilde{\Omega}_{S_{NM}} \end{pmatrix}, \tag{24}$$

where η is a nonzero constant. Using Eqs. (5) and (6), the above condition can be translated to

$$\begin{pmatrix} \mu_{01}^*\tilde{E}_{P_1}\exp(-i\phi_{P_1}) \\ \mu_{02}^*\tilde{E}_{P_2}\exp(-i\phi_{P_2}) \\ \vdots \\ \mu_{0N}^*\tilde{E}_{P_N}\exp(-i\phi_{P_N}) \end{pmatrix} = \eta\begin{pmatrix} \mu_{1M}\tilde{E}_{S_1}\exp(i\phi_{S_1}) \\ \mu_{2M}\tilde{E}_{S_2}\exp(i\phi_{S_2}) \\ \vdots \\ \mu_{NM}\tilde{E}_{S_N}\exp(i\phi_{S_N}) \end{pmatrix}. \tag{25}$$

Surprisingly, Eq. (25) makes no reference to the transition dipole moments that are related to states $|f_1\rangle, |f_2\rangle, \ldots, |f_{M-1}\rangle$. Then, even if one has no knowledge about the properties of states $|f_1\rangle, |f_2\rangle, \ldots, |f_{M-1}\rangle$ beforehand, it is still possible, by use of adiabatic passage, to completely suppress the population transferred to these states. Note also that condition (25) requires a definite phase relationship between the component fields of the full control field, which is a consequence of the fact that quantum interference effects are directly responsible for the appearance of the additional $(M-1)$ nodes of $|\Lambda\rangle$.

There is another interesting implication of Eq. (25). Let us assume that among the N transition dipole moments related to state $|f_M\rangle$, l of them, say, $\mu_{1M}, \mu_{2M}, \ldots, \mu_{lM}$, happen to be zero. To still have an $(M+N-1)$-node null eigenvector of $\mathbf{H}$, Eq. (25) requires $\tilde{E}_{P_1}=\tilde{E}_{P_2}=\cdots=\tilde{E}_{P_l}=0$, i.e., removing the l laser fields that couple state $|0\rangle$ to the intermediate states $|i_1\rangle, |i_2\rangle, \ldots, |i_l\rangle$. This clearly results in a simplification of our adiabatic passage scheme, but it does not suggest that in these cases the intermediate states $|i_1\rangle,|i_2\rangle,\ldots,|i_l\rangle$ are no longer important or can be removed from the system, because they are still coupled with the other unwanted degenerate states. These intermediate states in this situation can be regarded as a high-dimensional analog of the so-called branch state in a five-level extended STIRAP scheme [10], for which case it has been shown that increasing the coupling between an unwanted state and the branch state will enhance the yield of the desired state.

B. The $M<N$ case

In this case Eq. (11) requires

$$\mathbf{S}\begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_M \end{pmatrix} = -z_0 \begin{pmatrix} \Omega^*_{P_1} \\ \Omega^*_{P_2} \\ \vdots \\ \Omega^*_{P_N} \end{pmatrix}, \qquad (26)$$

and

$$\sum_{j=1}^{M} \Omega_{S_{kj}} y_j = -z_0 \Omega^*_{P_k}, \; k=M+1,\; M+2,\ldots,N. \qquad (27)$$

Under condition (22), Eq. (26) still gives the result of Eq. (23), which can also ensure that Eq. (27) is satisfied. It is then clear that the null eigenvector, if it exists, will still have $(M-1)$ nodes on the unwanted degenerate states.

Next we examine Eqs. (9) and (10) for $M<N$. Note first that under condition (22), Eq. (9) is included in Eq. (10). Equation (10) indicates that now there are N variables $x_1,x_2,\ldots,x_N$ and yet the number of the constraints is only M. Hence there is more than one solution to Eq. (10), i.e., $\mathbf{H}$ can have multiple null eigenvectors if $M<N$.

The most obvious solution to Eq. (10) is given by $x_1=x_2=\cdots=x_N=0$. Under condition (22) the associated null eigenvector is then given by

$$|\Lambda\rangle = |\Lambda_1\rangle \equiv [z_0;0,0,\ldots,0;0,0,\ldots,-z_0\xi(t)]^T. \qquad (28)$$

This eigenvector is analogous to the only null eigenvector in the $M=N$ case. Let the other null eigenvectors be denoted $|\Lambda_2\rangle$. Under condition (22) $|\Lambda_2\rangle$ can be written as

$$|\Lambda_2\rangle \equiv [z_0;x_1,x_2,\ldots,x_N;0,0,\ldots,-z_0\xi(t)]^T, \qquad (29)$$

with $x_1,x_2,\ldots,x_N$ satisfying Eq. (10) and at least one of them being nonzero. Since any linear combination of $|\Lambda_1\rangle$ and $|\Lambda_2\rangle$ is still a null eigenvector, we rewrite the other null eigenvectors as

$$|\Lambda_3\rangle \equiv |\Lambda_2\rangle - |\Lambda_1\rangle = (0;x_1,x_2,\ldots,x_N;0,0,\ldots,0)^T. \qquad (30)$$

Note that $|\Lambda_3\rangle$ is orthogonal to Λ_1. As such, at early times state $|\Lambda_1\rangle$ fully correlates with the initial state $|0\rangle$, but all the other null eigenvectors represented by $|\Lambda_3\rangle$ have zero overlap with state $|0\rangle$.

The important question is whether or not one can avoid populating states $|\Lambda_3\rangle$ and therefore avoid populating the intermediate states during the population transfer. At first glance it seems plausible that since states $|\Lambda_3\rangle$ are degenerate with $|\Lambda_1\rangle$, nonadiabatic transitions from $|\Lambda_1\rangle$ to $|\Lambda_3\rangle$ will be unavoidable. Interestingly, this is not the case. Specifically, the strength of the nonadiabatic coupling between $|\Lambda_3\rangle$ and $|\Lambda_1\rangle$ is proportional to $|\langle\Lambda_3|d\Lambda_1/dt\rangle|$, which turns out to be zero at all times due to the $(M+N-1)$-node structure of $|\Lambda_1\rangle$ and the $(M+1)$-node structure of $|\Lambda_3\rangle$ [see Eqs. (28) and (30)]. Hence, only the null eigenvector $|\Lambda_1\rangle$ that has $(M+N-1)$ nodes is relevant to the dynamics of population transfer.

It is interesting to note that if $M=1$ (i.e., there is only one target state) then our model system becomes a collection of N lambda systems. In this extreme case condition (25) may be regarded as a simple extension of a previous result for "double-lambda" systems [20]. It is then surprising that under the same condition complete population transfer to one target state can still be achieved even when this target state is degenerate with other states. The analysis here leads to a significant prediction. That is, under condition (25) and for $M\leqslant N$, we can always achieve adiabatic passage via the null eigenvector $|\Lambda_1\rangle$, irrespective of the actual value of M and the values of the transition dipole moments between the intermediate states and the unwanted degenerate states. As a result, during the entire process of population transfer the undesired degenerate states and all the intermediate states can never be populated, even if the exact number of the unwanted degenerate states and their properties are unknown to us.

Of course, if the exact number of degeneracies $M<N$ is given, it may be unnecessary to still use the $2N$ component fields to achieve complete population transfer. As indicated in the preceding section, one may remove $2(N-M)$ laser fields and consider only M intermediate states to realize the same control.

C. The $M>N$ case

In this case Eqs. (9) and (10) in general have only one solution given by Eq. (14). So the null eigenvector, if it exists, should generically have N nodes on all the intermediate states. However, there are multiple solutions to Eq. (11) since there are M unknown variables $y_1,y_2,\ldots,y_M$ while the number of the constraints is only N. Therefore $\mathbf{H}$ again has multiple null eigenvectors. Denote two orthogonal null eigenvectors of $\mathbf{H}$ by $|\Lambda'\rangle$ and $|\Lambda''\rangle$,

$$|\Lambda'\rangle = (z_0';0,0,\ldots,0;y_1',y_2',\ldots,y_M')^T,$$

$$|\Lambda''\rangle = (z_0'';0,0,\ldots,0;y_1'',y_2'',\ldots,y_M'')^T. \qquad (31)$$

The strength (denoted χ) of the nonadiabatic coupling between states $|\Lambda'\rangle$ and $|\Lambda''\rangle$ is proportional to $|(z_0')^*dz_0''/dt+\sum_{j=1}^{M}(y_j')^*dy_j''/dt|$. Due to the normalization requirement, $z_0'=y_1'=y_2'=\cdots=y_M'=0$ is not an acceptable solution to Eq. (11). As such, there is no general reason why χ should be zero. In particular, under condition (22), one finds that one null eigenvector can be analogous to that in the $M\leqslant N$ case, i.e., $|\Lambda'\rangle=[z_0;0,0,\ldots,0;0,0,\ldots,-z_0\xi(t)]^T$. Then $\chi \propto |z_0^* dz_0''/dt - z_0^*\xi^*(t)dy_M''/dt|$, which is nonzero in general. This being the case, state $|\Lambda''\rangle$ and therefore some of the states $|f_1\rangle,|f_2\rangle,\ldots,|f_{M-1}\rangle$ will be populated during the population transfer. This is true no matter how slowly the laser fields are turned on or off, as states $|\Lambda''\rangle$ and $|\Lambda'\rangle$ are degenerate.

Clearly, then, in the $M>N$ case, our scheme cannot guarantee complete population transfer to state $|f_M\rangle$. However, the results above suggest that this problem can be easily fixed if there are more intermediate states available. That is, using additional $(M-N)$ intermediate states and adding $2(M-N)$ laser fields will recover the desired complete population transfer.

D. Complete population transfer to arbitrary superpositions of M degenerate states

We note that states $|f_1\rangle,|f_2\rangle,\ldots,|f_M\rangle$ are just one particular choice of the basis states of an M-dimensional degenerate subspace. Indeed, any arbitrary superposition of states $|f_1\rangle,|f_2\rangle,\ldots,|f_M\rangle$ can be used as one of the basis states of the same degenerate subspace. Consider now a target superposition state

$$|f_M'\rangle = \sum_{j=1}^{M} c_j |f_j\rangle, \tag{32}$$

where c_j $(j=1,2,\ldots,M)$ are arbitrary coefficients in the superposition state. Let $|f_M'\rangle$ be the last new basis state of the same degenerate subspace and let the other new orthogonal basis states be $|f_1'\rangle,|f_2'\rangle,\ldots,|f_{M-1}'\rangle$. The transition dipole moment between state $|i_k\rangle$ and state $|f_j'\rangle$ is represented by μ_{kj}'.

We now apply the above general solution to complete population transfer in degenerate systems to state $|f_M'\rangle$ instead of $|f_M\rangle$. Then, for $M\leqslant N$ and

$$\det\begin{pmatrix} \mu_{11}' & \mu_{12}' & \cdots & \mu_{1M}' \\ \mu_{21}' & \mu_{22}' & \cdots & \mu_{2M}' \\ \vdots & \vdots & \vdots & \vdots \\ \mu_{M1}' & \mu_{M2}' & \cdots & \mu_{MM}' \end{pmatrix} \neq 0, \tag{33}$$

all population can be transferred from state $|0\rangle$ to the superposition state $|f_M'\rangle$ if the component fields satisfy

$$\begin{pmatrix} \mu_{01}^*\tilde{E}_{P_1}\exp(-i\phi_{P_1}) \\ \mu_{02}^*\tilde{E}_{P_2}\exp(-i\phi_{P_2}) \\ \vdots \\ \mu_{0N}^*\tilde{E}_{P_N}\exp(-i\phi_{P_N}) \end{pmatrix} = \eta \begin{pmatrix} \mu_{1M}'\tilde{E}_{S_1}\exp(i\phi_{S_1}) \\ \mu_{2M}'\tilde{E}_{S_2}\exp(i\phi_{S_2}) \\ \vdots \\ \mu_{NM}'\tilde{E}_{S_N}\exp(i\phi_{S_N}) \end{pmatrix}, \tag{34}$$

where

$$\mu_{kM}' = \sum_{j=1}^{M} c_j \mu_{kj}, \quad k=1,2,\ldots,N. \tag{35}$$

Since the transition dipole moments μ_{kj}' can be obtained from μ_{kj} by considering a unitary transformation between two different sets of basis states of the same degenerate subspace, it can be easily proved that condition (33) is equivalent to condition (19). Note also that there is no need to construct, from superpositions of states $|f_1\rangle,|f_2\rangle,\ldots,|f_M\rangle$, the explicit forms of states $|f_1'\rangle,|f_2'\rangle,\ldots,|f_{M-1}'\rangle$, as their properties are not required to predict that they can never be populated during the population transfer.

IV. NUMERICAL EXAMPLE

To confirm the validity and feasibility of our general solution to the realization of complete population transfer to one arbitrary member of a set of M degenerate states, we present in this section some numerical examples. In particular, we consider a system that has $N=7$ nondegenerate states as intermediate states and $M\leqslant N$ degenerate orthogonal states. We define the sum of population in all the intermediate states as P_x, the sum of population in all the degenerate states except for state $|f_M\rangle$ as P_y, and the population in the target state $|f_7\rangle$ as P_f. For convenience all the transition dipole moments are assumed to be real and all the laser phases are set to zero. Hence all the Rabi frequencies take real values. To fulfill condition (24), we assume $\tilde{\Omega}_{P_k}=\tilde{\Omega}_{S_{k7}}$, for $k=1,2,\ldots,N$.

Figure 2 displays an example of population transfer in the case of $M=N$. It is seen that during the population transfer, the maximum value of P_x is less than 0.3%, and the maximum value of P_y is less than 0.1%. The final P_f is extremely close to 100%. We then arbitrarily alter some of the peak Rabi frequencies that are related to states $|f_1\rangle,|f_2\rangle,\ldots,|f_6\rangle$. The associated results are shown in Fig. 3. Clearly, population transfer is still almost complete and P_x and P_y again remain negligible at all times. This demonstrates that totally suppressing the population transferred to states $|f_1\rangle,|f_2\rangle,\ldots,|f_6\rangle$ does not require us to know their properties beforehand. Similar results are obtained in numerous other numerical experiments.

Figure 4 displays analogous results in a case where two transition dipole moments μ_{17} and μ_{27} (and therefore the two peak Rabi frequencies $\tilde{\Omega}_{S_{17}}$ and $\tilde{\Omega}_{S_{27}}$) that are related to the target state $|f_7\rangle$ happen to be zero. As predicted by Eq. (25), this requires us to remove two laser fields that connect the initial state to two intermediate states. Accordingly we set $\tilde{\Omega}_{P_1}$ and $\tilde{\Omega}_{P_2}$ to zero. As seen from Fig. 4, almost complete population transfer with P_x and P_y negligible at all times is also achieved.

Figure 5 shows a numerical example where all the peak Rabi frequencies that are related to states $|f_1\rangle$ and $|f_2\rangle$ are set to zero, while keeping other peak Rabi frequencies the same as those used in Fig. 2. Due to this procedure, $|f_1\rangle$ and $|f_2\rangle$ are totally decoupled from the system and effectively there are only five degenerate states. As seen from Fig. 5, almost 100% population transfer to state $|f_7\rangle$ with P_x and P_y negli-

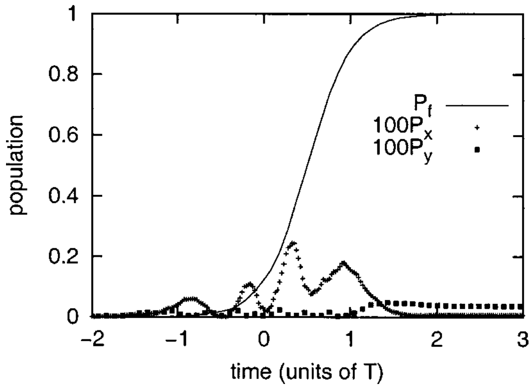

FIG. 2. A numerical example of adiabatic passage through a 13-node null eigenstate for the complete population transfer from an initial state to one arbitrary member of $M=7$ degenerate states. The number of intermediate states is given by $N=7$. P_x represents the population in all the intermediate states, P_y represents the population in all the degenerate states except for the target state, and P_f represents the population in the target state $|f_7\rangle$. The parameters for the peak Rabi frequencies are, in units of $1/T$, given by $(\tilde{\Omega}_{P_1},\tilde{\Omega}_{P_2},\tilde{\Omega}_{P_3},\tilde{\Omega}_{P_4},\tilde{\Omega}_{P_5},\tilde{\Omega}_{P_6},\tilde{\Omega}_{P_7})=(60,90,60,120,90,99,135)$, $(\tilde{\Omega}_{S_{11}},\tilde{\Omega}_{S_{12}},\tilde{\Omega}_{S_{13}},\tilde{\Omega}_{S_{14}},\tilde{\Omega}_{S_{15}},\tilde{\Omega}_{S_{16}},\tilde{\Omega}_{S_{17}})=(90,15,0,150,36,18,60)$, $(\tilde{\Omega}_{S_{21}},\tilde{\Omega}_{S_{22}},\tilde{\Omega}_{S_{23}},\tilde{\Omega}_{S_{24}},\tilde{\Omega}_{S_{25}},\tilde{\Omega}_{S_{26}},\tilde{\Omega}_{S_{27}})=(90,57,24,45,69,78,90)$, $(\tilde{\Omega}_{S_{31}},\tilde{\Omega}_{S_{32}},\tilde{\Omega}_{S_{33}},\tilde{\Omega}_{S_{34}},\tilde{\Omega}_{S_{35}},\tilde{\Omega}_{S_{36}},\tilde{\Omega}_{S_{37}})=(90,75,39,36,39,78,60)$, $(\tilde{\Omega}_{S_{41}},\tilde{\Omega}_{S_{42}},\tilde{\Omega}_{S_{43}},\tilde{\Omega}_{S_{44}},\tilde{\Omega}_{S_{45}},\tilde{\Omega}_{S_{46}},\tilde{\Omega}_{S_{47}})=(60,18,24,75,66,48,120)$, $(\tilde{\Omega}_{S_{51}},\tilde{\Omega}_{S_{52}},\tilde{\Omega}_{S_{53}},\tilde{\Omega}_{S_{54}},\tilde{\Omega}_{S_{55}},\tilde{\Omega}_{S_{56}},\tilde{\Omega}_{S_{57}})=(39,27,93,15,66,78,90)$, $(\tilde{\Omega}_{S_{61}},\tilde{\Omega}_{S_{62}},\tilde{\Omega}_{S_{63}},\tilde{\Omega}_{S_{64}},\tilde{\Omega}_{S_{65}},\tilde{\Omega}_{S_{66}},\tilde{\Omega}_{S_{67}})=(93,69,18,87,72,78,99)$, and $(\tilde{\Omega}_{S_{71}},\tilde{\Omega}_{S_{72}},\tilde{\Omega}_{S_{73}},\tilde{\Omega}_{S_{74}},\tilde{\Omega}_{S_{75}},\tilde{\Omega}_{S_{76}},\tilde{\Omega}_{S_{77}})=(36,54,48,57,96,78,135)$. Note that condition (24) is satisfied since we have chosen $\tilde{\Omega}_{P_k}=\tilde{\Omega}_{S_{k7}}$, for $k=1,2,\ldots,7$. At all times $P_x<0.3\%$ and $P_y<0.05\%$.

gible at all times is also obtained. This clearly confirms our previous prediction that our adiabatic passage method works as well even if the exact number of degeneracies (e.g., $M=5$ or $M=7$) is unknown to us.

To end this section, we note that in general nonadiabatic effects in many-level systems are expected to be stronger than those in few-level systems. Admittedly, it is not an easy

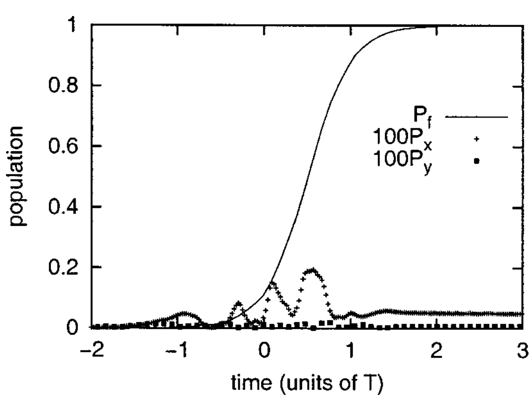

FIG. 3. Same as in Fig. 2 except $\tilde{\Omega}_{S_{14}}=39$, $\tilde{\Omega}_{S_{25}}=39$, $\tilde{\Omega}_{S_{26}}=48$, $\tilde{\Omega}_{S_{32}}=45$, $\tilde{\Omega}_{S_{43}}=81$, $\tilde{\Omega}_{S_{52}}=57$, $\tilde{\Omega}_{S_{56}}=48$, $\tilde{\Omega}_{S_{62}}=39$, $\tilde{\Omega}_{S_{72}}=24$. At all times $P_x<0.2\%$ and $P_y<0.03\%$.

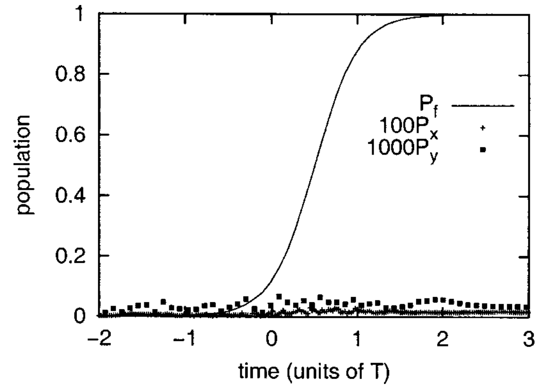

FIG. 4. Same as in Fig. 2 except $\tilde{\Omega}_{S_{17}}=\tilde{\Omega}_{S_{27}}=0$ and $\tilde{\Omega}_{P_1}=\tilde{\Omega}_{P_2}=0$. At all times $P_x<0.03\%$ and $P_y<0.01\%$.

task to analytically expose the nonadiabatic effects associated with our model system. However, as seen in the numerical examples, nonadiabatic effects here are small even when the field strength is still not very strong. We have also checked that by increasing the pulse width or the field strength, or optimizing the time delay between the laser fields and the ratio of the peak Rabi frequencies (e.g., $\tilde{\Omega}_{P_k}/\tilde{\Omega}_{S_{kM}}$), one can always further suppress the nonadiabaticity of the dynamics and therefore the population transfer can be made even closer to the adiabatic limit.

V. DISCUSSION AND CONCLUSION

Controlled population transfer from a selected initial state to a manifold of degenerate states has previously been examined in the context of atomic processes involving magnetic sublevels [21–25]. Our results are much more powerful and general than those previously established. A key difference between this work and previous studies is that we have exploited the properties of particular coherent excitations of manifolds with multiple nondegenerate intermediate states. In particular, with N nondegenerate intermediate states it becomes possible to adjust, with $2N$ laser fields, all the Rabi frequencies involved in Eq. (22). Hence Eq. (22) can always

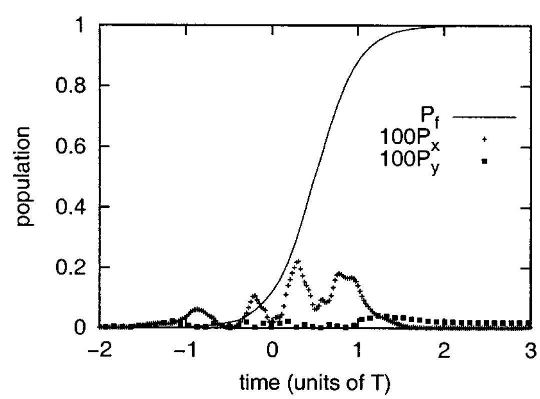

FIG. 5. Same as in Fig. 2 except that all the peak Rabi frequencies that are related to states $|f_1\rangle$ and $|f_2\rangle$ are set to zero, i.e., $\tilde{\Omega}_{S_{k1}}=\tilde{\Omega}_{S_{k2}}=0$, for $k=1,2,\ldots,7$. At all times $P_x<0.3\%$ and $P_y<0.05\%$.

be satisfied by designing the laser fields, providing thereby a systematic solution to the design of complete population transfer from an initial state to an arbitrary superposition of $M \leqslant N$ degenerate states. It is also straightforward to determine if the requirements [i.e., condition (19) and $M \leqslant N$] are met for our general solution to be applicable.

Most coherent control approaches based on STIRAP-like dynamics are independent of the relative phases of the control laser fields [17]. As shown in this work and in Refs. [19,20,25], this is not true for all STIRAP-like control schemes. On one hand, the phase-dependence of the control process causes even more experimental difficulties; on the other hand the phase-dependence of our adiabatic passage scheme clearly demonstrates that laser phases can be also important in STIRAP-like dynamics and may bring about dramatic opportunities for controlling atomic and molecular processes.

One great advantage of our adiabatic passage scheme is its adaptability in cases where the intermediate states or the unwanted degenerate states are not bound states. That is, since the $(M+N-1)$-node null eigenvector does not overlap with any of the intermediate states or any of the unwanted degenerate states, the structure of this multinode eigenvector survives if these states actually decay. As long as the associated decay rate constants are much smaller than the peak Rabi frequencies of the component fields [19], the nonadiabaticity induced by the decaying states will be negligible and the dynamics of population transfer would be still given by the evolution of the $(M+N-1)$-node null eigenvector.

This work extends our previous adiabatic passage method for the realization of complete control of the population transfer branching ratio between two degenerate states [19]. The extension is from a five-level and four-pulse scheme to a $(1+N+M)$-level and $(2N)$-pulse scheme. While the five-level and four-pulse scheme should be experimentally achievable, directly applying the present work to realistic systems would be demanding for $M \geqslant 3$, as it would in general require a significant number of laser fields. However, from the point of view of understanding the limits to complete controllability in degenerate systems subject to strong constrains and/or with unknown parameters, this work is of great theoretical interest. In particular, we have shown that it is possible to realize complete population transfer without ever populating a Hilbert subspace that is only dimension-two smaller than the whole Hilbert space, even when the exact number of degeneracies and the properties of the unwanted degenerate states are unknown to us. We hope that the results of this study will motivate future theretical work on coherent control in degenerate systems. For example, it is interesting to consider variations on our adiabatic passage scheme, and to seek alternative, and experimentally more feasible, control scenarios that can provide the same type of population transfer pathway that is already shown to exist.

Although our adiabatic passage solution offers a general method for complete population transfer from an initial state to an arbitrary member of a set of M degenerate states, there are still two subtle differences between this solution and an actual proof of complete controllability of the degenerate system. First, a completely controllable degenerate system will have a complete population transfer pathway within a finite time, whereas the complete population transfer in our solution, in the most strict sense (i.e., P_f is exactly 100%), can only be realized in the adiabatic limit, e.g., for infinitely large pulse width. Second, we have used the rotating-wave approximation in constructing our solution. Mathematically speaking, such an approximation inevitably modifies the issue of complete controllability. It is unclear to what physical extent this approximation changes the controllability of degenerate systems. For the above two reasons we regard our adiabatic passage method as a specific physical, but not mathematical, solution to the complete control of degenerate systems.

One potential application of this study is to provide a useful guide for understanding the extremely complex laser fields obtained by genetic algorithms in adaptive feedback control of quantum systems [1,2,26]. Suppose an adapative feedback control experiment is carried out in a degenerate system and the control goal is to maximize the population in only one of M degenerate states and minimize the population in all the unwanted degenerate states and the intermediate states. It would be of great interest to see if the optimized control field suggested by a genetic algorithm can be decomposed into $2N \geqslant 2M$ laser fields with their frequencies, amplitudes, and relative phases close to what is suggested by our adiabatic passage scheme. If this is the case, then such a feedback control experiment becomes a realization of our general solution to complete population transfer in degenerate systems and more applications of this work can be expected.

To summarize, we have shown that under certain conditions complete population transfer from an initial state, via N nondegenerate intermediate states, to an arbitrary superposition of M $(M \leqslant N)$ degenerate states is achievable by adiabatically following an $(M+N-1)$-node null eigenstate that is created by designing the relative phases and amplitudes of the component fields of a control field. The results may find applications in quantum information processing in atoms and molecules and shed considerable light on the issue of complete controllability in degenerate systems under strong constraints and/or with unknown parameters.

Note added in proof. Recently a similar idea with a specific molecular application was published in Ref. [27].

ACKNOWLEDGMENT

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