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Simple and robust extension of the stimulated Raman adiabatic passage technique to N-level systems

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STIRAP (stimulated Raman adiabatic passage) has proven to be an efficient and robust technique for transferring population in a three-level system without populating the intermediate state. Here we show that the counterintuitive pulse sequence in STIRAP, in which the Stokes pulse precedes the pump, emerges automatically from a variant of optimal control theory we have previously called ''local'' optimization. Since local optimization is a well-defined, automated computational procedure, this opens the door to automated computation of generalized STIRAP schemes in arbitrarily complicated N-level coupling situations. If the coupling is sequential, a simple qualitative extension of STIRAP emerges: the Stokes pulse precedes the pump as in the three-level system. But, in addition, spanning both the Stokes and pump pulses are pulses corresponding to the transitions between the $N-2$ intermediate states with intensities about an order of magnitude greater than those of the Stokes and pump pulses. This scheme is amazingly robust, leading to almost 100% population transfer with significantly less population transfer to the $N-2$ intermediate states than in previously proposed extensions of STIRAP. [S1050-2947(97)07611-7]

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I. INTRODUCTION

In recent years, the design of laser pulse sequences to achieve efficient and robust population transfer between quantum states has been the subject of many theoretical and experimental studies [1–24]. This problem is relevant to many applications, including spectroscopy, collision dynamics, and optical control of chemical reactions. A considerable number of studies have been devoted to the process of stimulated Raman adiabatic passage (STIRAP) in three-level [1–7] and multilevel [11–19]. systems. Recently, the STIRAP theory has been generalized to the case in which the intermediate state is autoionizing [21] or is described by a continuum of levels [22,23].

The STIRAP process provides the possibility of effective population transfer using relatively simple experimental setups. At the same time, it demonstrates a remarkable *counterintuitive* mechanism at work, in which the pump pulse, driving the transition between the initially populated level $|1\rangle$ and intermediate level $|2\rangle$ comes *after* the Stokes pulse, which drives the transition between the initially unpopulated levels $|2\rangle$ and $|3\rangle$ (Fig. 1). This ordering of pulses is both efficient and robust in achieving complete population transfer from state $|1\rangle$ to $|3\rangle$, while maintaining the population of state $|2\rangle$ at almost zero.

The properties of the STIRAP mechanism in a three-level system have been explored extensively, both numerically and analytically. For the most part the analytical studies have been performed in the adiabatic limit [1,4,9], although nonadiabatic effects in population transfer in three-level systems have been considered in [7]. Complete analytic results have been obtained only for specific pulse shapes [2,6]. Several extensions to N-level systems have been proposed [12,16–19], however, none to date appears to be definitive. For a review of the literature related to coherent population transfer in atomic and molecular systems, see [24].

Quite independent of the STIRAP literature, there has been a growing literature in recent years on the use of shaped optical pulse sequences to control atomic and molecular dynamics for various purposes [25–38]. These applications include laser heating [33] and cooling of molecules [34], preparation of specific electronic, vibrational or rotation states, and control of the products of chemical reactions [25,27,30,31]. One of the main computational tools brought to bear in these studies is optimal control theory (OCT) [27,28,31,37]. The closest application of optimal control to systems of the STIRAP type was a study of the design of optical pulse pairs to control the population transfer of three-level atoms in a medium [37]. However, the connection between STIRAP and the OCT literature has been elusive and explicit attempts to derive the STIRAP mechanism from optimal control have been unsuccessful [39]. This is not particularly surprising: (1) Adiabatic passage is generally energetically expensive relative to a Rabi pulse sequence, typically employing integrated pulse areas many times π.

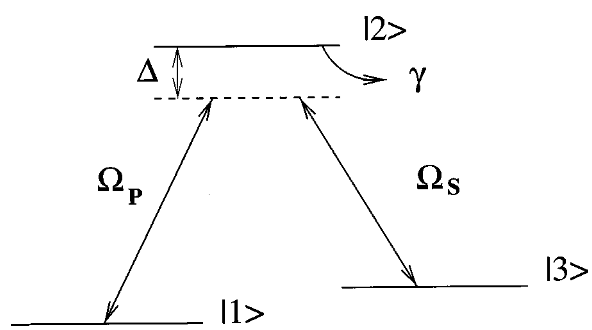

FIG. 1. The three-level Λ system. Level $|1\rangle$ is coupled to level $|2\rangle$, which in turn is coupled to level $|3\rangle$. There is assumed to be a two-photon resonance between levels $|1\rangle$ and $|3\rangle$, although there may not be a one-photon resonance with level $|2\rangle$. Levels $|1\rangle$ and $|2\rangle$ are coupled by a field with amplitude Ω_p; levels $|2\rangle$ and $|3\rangle$ are coupled by a field with amplitude Ω_s. Δ is the detuning of the intermediate level $|2\rangle$.

The penalty on the energy of the field that is routinely used in OCT calculations discriminates against pulses with large integrated areas. (2) The robustness of the STIRAP solution, which is perhaps its principle advantage, is difficult to quantify. Without robustness being incorporated explicitly into the objective functional in OCT there is no reason to expect STIRAP-type solutions to emerge from an OCT calculation.

A second class of techniques that have been developed for control of atomic and molecular dynamics, in parallel to OCT, is called ''local optimization'' [33–36], or tracking [38]. In these methods, at every instant in time the control field is chosen to achieve a monotonic increase in the desired objective. Typically in these methods two conditions are used at each time step, one to determine the phase of the field and one to determine the amplitude. In contrast with OCT, which incorporates information on later time dynamics through forward-backward iteration, these methods use only information on the current state of the system and the yields are in principle lower than in OCT. However, there are several attractive features to the local methods. (1) Since the increase in yield is monotonic these fields are often amenable to immediate interpretation. (2) Since these methods use information on the current state of the system only, they could in principle be adapted for laboratory implementation. (3) Because these methods differ so radically from OCT, they may identify different classes of solutions from OCT. Although not necessarily optimal, these solutions may be appealing because of other properties, e.g., their robustness.

In recent studies, we showed how a local optimization scheme could be used to lock population on an intermediate level or levels, while increasing the energy in another part of the system [33–36]. Specifically, we showed that the phase of the field could be used to lock the population in a manifold of excited levels while the sign of the amplitude could be used to achieve ground-state vibrational heating. Further reflection on these studies suggests some parallels with the STIRAP mechanism: (1) the locking of population on the intermediate level is the analog of avoiding population transfer to level $|2\rangle$ in STIRAP; (2) the monotonic increase in energy in the ground vibrational manifold is identical, in the case of only two participating vibrational levels in the ground state, to the monotonic transfer of population from level $|1\rangle$ to $|3\rangle$ in STIRAP; (3) the role of the phase is crucial in the local scheme, as it is in STIRAP; (4) in both schemes the specific features of the amplitude function have a great deal of flexibility.

The goal of this paper is, first of all, to show that there is indeed a rigorous relationship between local optimization methods and STIRAP in the limit of three-level systems and long times. Once this relationship is established, a great many things become possible. Since local optimization is a flexible method and can be applied to arbitrarily complicated situations, e.g., to multilevel systems, to systems with detuning and radiative decay, and for times shorter than those required for the adiabatic limit, this raises the possibility of finding numerical solutions of the STIRAP type for all these situations. Several extensions of STIRAP to N-level systems have been proposed recently [12,16–19]. Generally, these extended STIRAP schemes employ a set of $N-1$ pulses, one pulse for each of the transition frequencies between the coupled states. In one proposal, the envelopes progress in reverse order from the last to the first transition [14]. In a second proposal the envelopes are grouped into two overlapping sets, with the even transitions coming before the odd transitions [12]. Here we find a third strategy in which the Stokes pulse, resonant with the transition $|N-1\rangle \rightarrow |N\rangle$ comes before the pump pulse, resonant with the transition $|1\rangle \rightarrow |2\rangle$; the envelopes for all the other transition frequencies are about an order of magnitude more intense than either the Stokes or the pump pulses and span *both* the pump and the Stokes pulse.

In Sec. II, we present the basic description of the control scheme by applying it to population transfer in a three-level system. The role of detuning and decay is also explored. In Sec. III, we analyze control of the population transfer in four- and five-level systems. In Sec. IV we abstract the key features of the pulse sequences found in Sec. III and apply them to a nine-level system. Section V is a conclusion.

II. THREE-LEVEL SYSTEM

Consider the interaction of a three-level Λ system with a pump $E_p(t)$ and a Stokes $E_s(t)$ laser pulse:

$$E = E_p(t)\cos\omega_p t + E_s(t)\cos\omega_s(t), \tag{1}$$

where $E_p(t)$ and $E_p(t)$ are the envelopes of the pulses. In the interaction representation and the rotating-wave approximation, the Schrödinger equation takes the form

$$\begin{pmatrix} \dot{a}_1(t) \\ \dot{a}_2(t) \\ \dot{a}_3(t) \end{pmatrix} = \begin{pmatrix} 0 & i\Omega_p(t)\exp\{i\Delta\omega_{21}t\} & 0 \\ i\Omega_p(t)\exp\{-i\Delta\omega_{21}t\} & 0 & i\Omega_s(t)\exp\{-i\Delta\omega_{23}t\} \\ 0 & i\Omega_s(t)\exp\{i\Delta\omega_{23}t\} & 0 \end{pmatrix} \begin{pmatrix} a_1(t) \\ a_2(t) \\ a_3(t) \end{pmatrix}, \tag{2}$$

where a_1, a_2, and a_3 are the probability amplitudes of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$, $\Omega_p(t) = \mu_{12}E_p(t)/2\hbar$ and $\Omega_s(t) = \mu_{23}E_s(t)/2\hbar$ are the pump and Stokes Rabi frequencies, respectively, and $\Delta\omega_{21} = \omega_p - \omega_{21}$ and $\Delta\omega_{23} = \omega_s - \omega_{23}$ are the detunings of the laser frequencies $\omega_{p,s}$ from the transition frequencies $\omega_{21,23}$.

In the assumption of a Raman resonance, one can rewrite the Schrödinger equation in the following form:

$$\begin{pmatrix} \dot{a}_1(t) \\ \dot{a}_2(t) \\ \dot{a}_3(t) \end{pmatrix} = \begin{pmatrix} 0 & i\Omega_p(t) & 0 \\ i\Omega_p(t) & i\Delta & i\Omega_s(t) \\ 0 & i\Omega_s(t) & 0 \end{pmatrix} \begin{pmatrix} a_1(t) \\ a_2(t) \\ a_3(t) \end{pmatrix}, \tag{3}$$

where $\Delta = \Delta\omega_{21} = \Delta\omega_{23}$.

Our goal, as in STIRAP, is to find envelopes of the pump and Stokes pulses that lead to complete and robust population transfer from state $|1\rangle$ to the final state $|3\rangle$, while keeping the population of the intermediate state $|2\rangle$ as small as possible. Here we propose to do this using our earlier local optimization scheme [36], modified to suit the three-level configuration. Our strategy is first to find a condition on the phase of the laser fields that keeps the population on level $|2\rangle$ locked and then to find conditions on the amplitude of the laser fields that lead to monotonic increase in the population in level $|3\rangle$. If the population in level $|2\rangle$ is locked at a small value and if complete population transfer from level $|1\rangle$ to level $|3\rangle$ is achieved then the procedure has achieved the same objective as in STIRAP. In fact, as we shall now see, for the particular case of a three-level system the STIRAP solution emerges automatically.

Using the Schrödinger equation, Eq. (3), we can develop expressions for the time derivative of the populations in each of the three levels:

$$\frac{d|a_1(t)|^2}{dt}=2\,\mathrm{Re}\{a_1^*(t)\dot{a}_1(t)\}=-2\Omega_p(t)\mathrm{Im}\{a_1^*(t)a_2(t)\},$$

$$\frac{d|a_2(t)|^2}{dt}=2\,\mathrm{Re}\{a_2^*(t)\dot{a}_2(t)\}=-2[\Omega_p(t)\mathrm{Im}\{a_2^*(t)a_1(t)\}+\Omega_s(t)\mathrm{Im}\{a_2^*(t)a_3(t)\}],$$

$$\frac{d|a_3(t)|^2}{dt}=2\,\mathrm{Re}\{a_3^*(t)\dot{a}_3(t)\}=-2\Omega_s(t)\mathrm{Im}\{a_3^*(t)a_2(t)\}. \tag{4}$$

Locking population in level $|2\rangle$ amounts to the condition that

$$\frac{d|a_2(t)|^2}{dt}=0. \tag{5}$$

We note that this condition is satisfied if we choose Ω_p and Ω_s as

$$\Omega_p=-\Omega_0(t)\mathrm{Im}\{a_3^*(t)a_2(t)\}, \tag{6}$$

$$\Omega_s=\Omega_0(t)\mathrm{Im}\{a_1^*(t)a_2(t)\}, \tag{7}$$

The magnitude of $\Omega_0(t)$ is an arbitrary envelope function that may be chosen on physical grounds to satisfy reasonable conditions of switching on and off. The sign of $\Omega_0(t)$, however, is crucial: the choice of sign is used to satisfy the condition that

$$\frac{d|a_1(t)|^2}{dt}<0. \tag{8}$$

$$\frac{d|a_3(t)|^2}{dt}>0. \tag{9}$$

At first glance this looks like two conditions that must be satisfied with only one unknown, Ω_0; note, however, that if $|a_2(t)|^2$ is locked then the decrease in $|a_1(t)|^2$ guarantees the increase in $|a_3(t)|^2$. This observation generalizes to N levels: if the population in the $N-2$ intermediate levels is locked a decrease in $|a_1(t)|^2$ guarantees a decrease in $|a_N(t)|^2$. For the calculations shown below, the envelope of the locking pulses was chosen as

$$\Omega_0(t)=\frac{\Omega_0}{\cosh^2(t-t_d)}, \tag{10}$$

where Ω_0 is an overall amplitude factor and t_d is the delay time of the locking pulses relative to the seed pulse (see below). We arbitrarily chose $t_d=6$ for both the pump and Stokes pulses, just to get a smooth shape for both the beginning and end of the pulses.

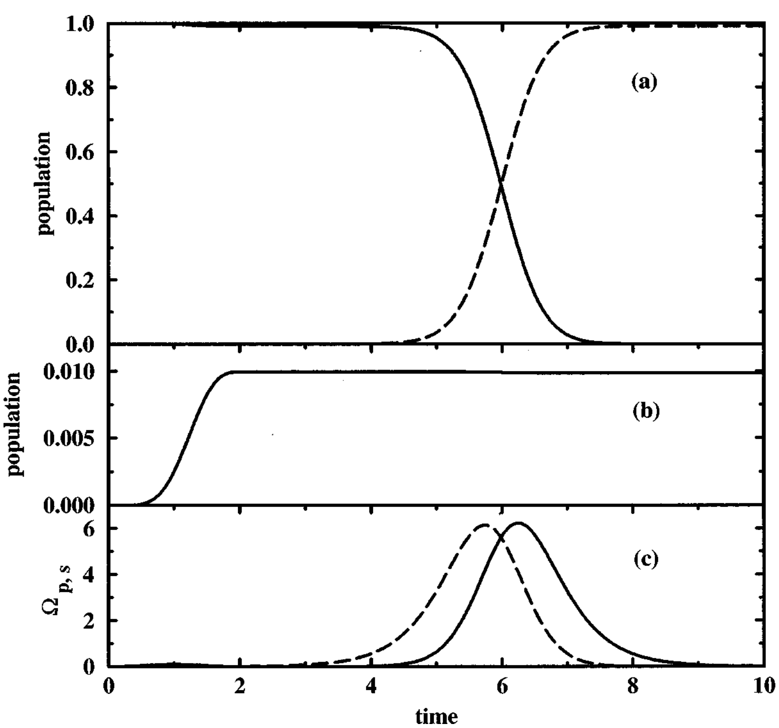

FIG. 2. Population transfer in the three-level Λ system using a sequence of two optical pulses with local optimization (see text). (a) The population of $|1\rangle$ (solid line) and $|3\rangle$ vs time (dashed line). (b) Population of the intermediate level $|2\rangle$ vs time. (c) The sequence of two optical pulses found using the local optimization method. Note that the counterintuitive sequence of Stokes pulse (dashed line) followed by pump pulse (solid line) emerges automatically in the method.

One of the characteristic features of the local optimization method is the need for a ''seed'' pulse that puts at least some small amount of population on all intermediate states that will be used in the transfer process. The need for such a seed population can be seen from Eqs. (6) and (7), which shows that to get nonzero fields from the algorithm the amplitudes $a_j(t)$ must be nonzero. To prepare this seed population we use two pulses, which are resonant with the transitions $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively. For all the simulations shown below the following functional form was used for the envelope of the seed pulses:

$$\Omega_{\text{seed}} = A_0 \sin \frac{\pi t}{2\tau_s}. \tag{11}$$

The amplitude A_0 controls the amount of the intermediate level population prepared during the seed and is typically much smaller than Ω_0. The dimensionless pulse duration τ_s was taken to be 2 in all simulations.

Figure 2 displays the time evolution of the population in a three-level system. All parameters are normalized by the width of the seed pulse [Eq. (11)]. During the seed pulse, population is exchanged freely between the states. At the end of the seed period the locking pulses are applied. Note the monotonic transfer of population from level $|1\rangle$ to $|3\rangle$ in Fig. 2(a). The locking of the population in the intermediate level $|2\rangle$ at a value of around 1% is shown in Fig. 2(b). The envelopes of the pulses, Ω_p and Ω_s, that emerge from this procedure are shown in Fig. 2(c). Note the counterintuitive ordering of Stokes pulse before pump pulse, characteristic of STIRAP, emerging automatically in this calculation. To understand this, note that the first factor in Eqs. (6) and (7), $\Omega_0(t)$, is identical for both pulses; the second factor is different, however, and turns on faster in Eq. (7) than in Eq. (6), since level $|1\rangle$ is populated at $t=0$ and level $|3\rangle$ is not.

In Fig. 3, the final-state population is plotted as a function of time delay between pulses and as a function of effective Rabi frequency, $\sqrt{[\Omega_p^{\max}]^2+[\Omega_s^{\max}]^2}$, where $\Omega_p^{\max}$ and $\Omega_s^{\max}$ are maximum values of the field amplitudes of the pump and Stokes pulses. It is seen that beyond a critical value of field intensity and time delay, the solutions are robust with respect to change in parameters. This behavior is a well-known feature of the STIRAP mechanism.

The effect of decay of the intermediate level $|2\rangle$ may be explored by adding the term $-\gamma a_2(t)$ into the right-hand side of the second equation of Eq. (3). The result is illustrated in Fig. 4. It is seen that complete population transfer is still achieved except for the part of the population that was in the intermediate level immediately after the seed pulse. Note again the counterintuitive order of the pulses.

It is interesting to consider the effect of detuning from the intermediate level $|2\rangle$ while maintaining the two-photon resonance condition between levels $|1\rangle$ and $|2\rangle$. It may be seen from Eq. (4) that the detuning Δ does not enter into the equations for the population changes although it does enter into the equations of motion, Eq. (3). Thus, in the rotating frame the solutions are identical with and without detuning, but after transforming back to the original representation the solutions are significantly different. The envelopes for the pulses with detuning are now oscillatory and quite complicated, as compared with the smooth envelopes for the resonant intermediate case.

III. CONTROL OF POPULATION TRANSFER IN FOUR- AND FIVE-LEVEL SYSTEM

In the interaction representation and the rotating-wave approximation, the Schrödinger equation for a system of N states with sequential coupling takes the form

$$\begin{pmatrix} \dot a_1(t) \\ \dot a_2(t) \\ \dot a_3(t) \\ \dot a_4(t) \\ \dot a_5(t) \\ \vdots \\ \dot a_{N-1}(t) \\ \dot a_N(t) \end{pmatrix} = \begin{pmatrix} 0 & i\Omega_1(t) & 0 & 0 & 0 & \cdots & 0 & 0 \\ i\Omega_1(t) & i\Delta_1 & i\Omega_2(t) & 0 & 0 & \cdots & 0 & 0 \\ 0 & i\Omega_2(t) & i\Delta_2 & i\Omega_3(t) & 0 & \cdots & 0 & 0 \\ 0 & 0 & i\Omega_3(t) & i\Delta_3 & i\Omega_4(t) & \cdots & 0 & 0 \\ 0 & 0 & 0 & i\Omega_4(t) & i\Delta_4 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & 0 & \cdots & i\Delta_{N-1} & i\Omega_{N-1}(t) \\ 0 & 0 & 0 & 0 & 0 & \cdots & i\Omega_{N-1}(t) & 0 \end{pmatrix} \begin{pmatrix} a_1(t) \\ a_2(t) \\ a_3(t) \\ a_4(t) \\ a_5(t) \\ \vdots \\ a_{N-1}(t) \\ a_N(t) \end{pmatrix}, \tag{12}$$

where $\Omega_j(t)(j=1,2,\ldots,N-1)$ are the Rabi frequencies of the first, second, and $N-1$ transitions, respectively, and Δ_j are the detunings of the corresponding transitions.

For the sake of simplicity we will address the resonant case; we therefore set $\Delta_j=0$ for all j. We again seek the condition to lock the population on the intermediate levels while transferring population from the initial to the final level. For a four-level system these conditions are

$$\Omega_1 = -\Omega_0(t)a_3(t)\text{Im}\{a_3^*(t)a_2(t)\}/a_1(t),$$

$$\Omega_2 = \Omega_0(t)\text{Im}\{a_3^*(t)a_4(t)\}, \tag{13}$$

$$\Omega_3 = -\Omega_0(t)\text{Im}\{a_3^*(t)a_2(t)\}.$$

For a five-level system we add one more equation:

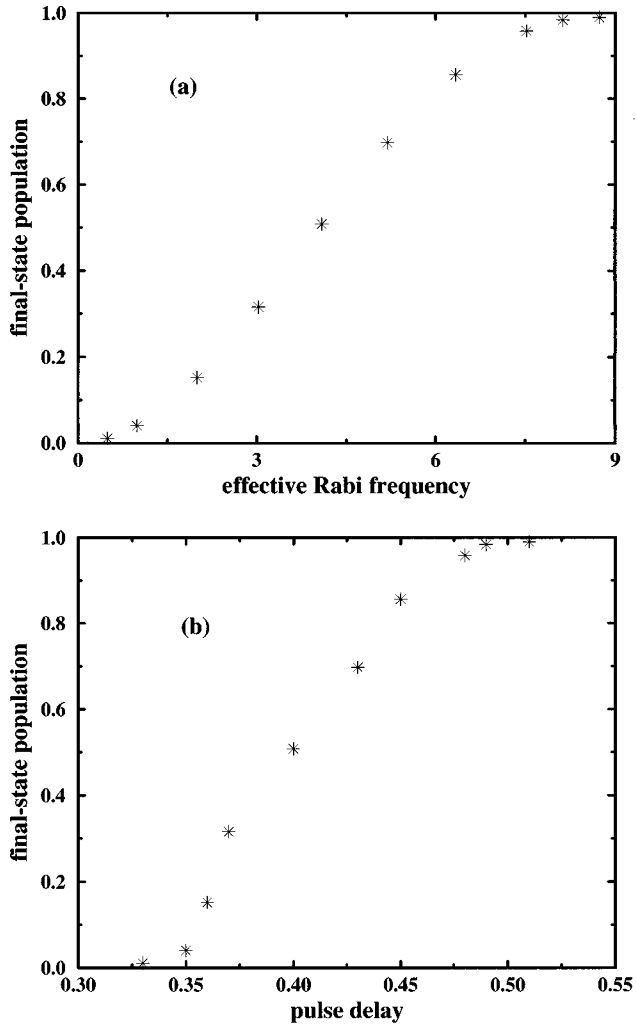

FIG. 3. (a) Population in level $|3\rangle$ as a function of the effective Rabi frequency, $\Omega_e = \sqrt{\Omega_s^2 + \Omega_p^2}$. (b) Population in level $|3\rangle$ as a function of the delay between the Stokes and pump pulses.

$$\Omega_4 = \Omega_0(t) a_3(t) \mathrm{Im}\{a_3^*(t) a_2(t)\}/a_5(t), \tag{14}$$

where $\Omega_0(t)$ is an overall amplitude factor for the control pulses which may be chosen the same way as in previous section, Eq. (10).

The results of the simulations for population transfer in the four- and five-level systems are shown in Figs. 5 and Fig. 6, respectively. Three and four seed pulses were used, respectively, with the shapes and durations of the pulses chosen in the same way as in the three-level system, Eq. (11). Qualitatively, the evolution of the population in the four- and five-level systems is similar to that in the three-level system: there is monotonic transfer of population from the initial to the final state [Figs. 5(a) and 6(a)] with almost no population in the intermediate levels [Figs. 5(b) and 6(b)]. Again, the counterintuitive sequence of the pulses emerges for both these cases [Figs. 5(c), 5(d) and Figs. 6(c), 6(d)]. However, there is an interesting new twist, in that the resonant frequencies connecting the intermediate levels among themselves now appear, with envelopes that straddle both the Stokes and the pump pulse, and with intensities significantly higher than that of either the Stokes or pump pulses. We will refer to this general pattern as a ''straddling'' STIRAP sequence (S-STIRAP) and we show below that it is an efficient and robust extension of STIRAP to generic N-level systems.

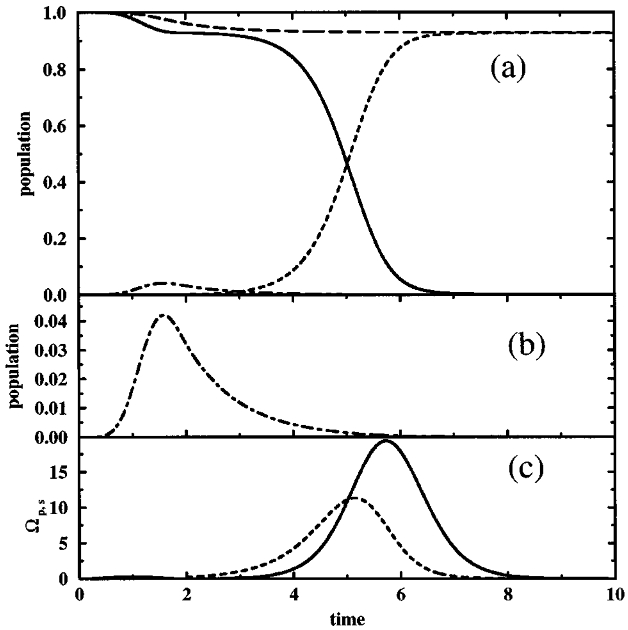

FIG. 4. (a) Population transfer in a Λ system in the presence of decay from the intermediate level $|2\rangle$, using local optimization. $\gamma\tau = 0.5$. The population of $|1\rangle$ (solid line) and $|3\rangle$ (dashed line), total population (long-dashed line). (b) Population of the intermediate level $|2\rangle$ vs time. (c) The sequence of two optical pulses found using the local optimization method. Note that the counterintuitive sequence of Stokes pulse (dashed line) followed by pump pulse (solid line) again emerges automatically in the method.

IV. THE STRADDLING STIRAP SEQUENCE

Stimulated by the simple, general form of the pulse sequences that emerged in the calculations described in the previous section, we conjecture that the S-STIRAP strategy is robust and that the seed pulse and local optimization condition can be abandoned. For an N-level system one simply applies the counterintuitive Stokes pump pulse sequence for the $|N-1\rangle \rightarrow |N\rangle$ and $|1\rangle \rightarrow |2\rangle$ transitions, respectively, and a set of intense pulses corresponding to all intermediate transition frequencies with envelopes that span both the Stokes and pump pulses. Figure 7 shows that this scheme is successful in transferring about 99% of the population from level $|1\rangle$ to level $|9\rangle$ in a nine-level system. At intermediate times, the largest amount of population to any intermediate level is less than 3%. We emphasize that in these calculations there are neither control conditions nor seed pulses: the simple general pattern of the previous section, which we call S-STIRAP, was used without any attempt at optimization. The envelope for all straddling pulses was taken as

$$\Omega_i(t) = \frac{\Omega_i}{\cosh^2(t - t_d^i)}. \tag{15}$$

An alternative generalized STIRAP scheme has been proposed recently by Oreg *et al.* In that scheme, the steps in the population transfer process $|1\rangle \rightarrow |2\rangle \cdots \rightarrow |N\rangle$ are alternately classified as either odd or even, i.e., the $|1\rangle \rightarrow |2\rangle$ transition

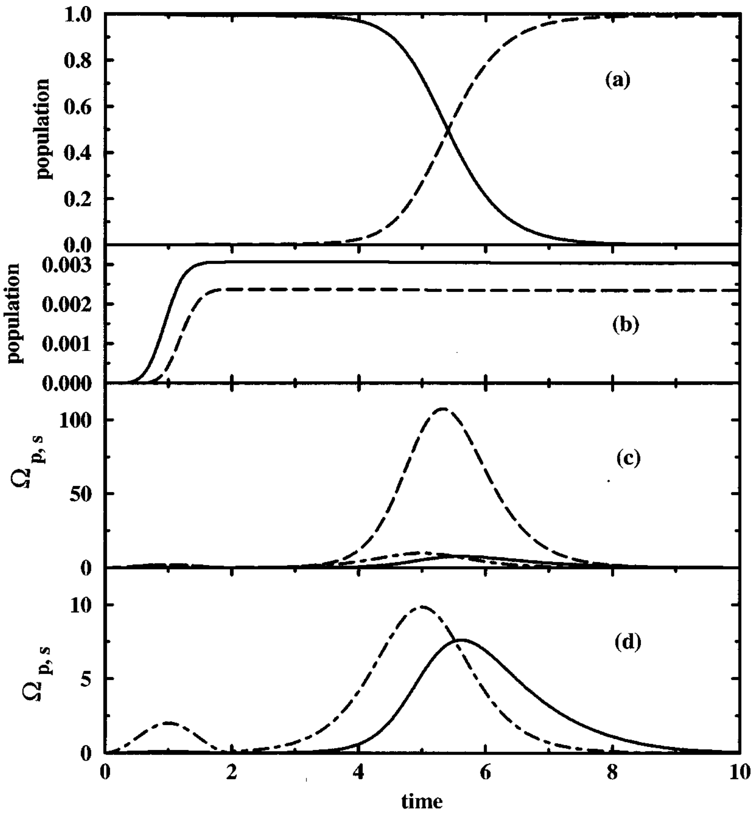

FIG. 5. Population transfer in a four-level, sequentially coupled system, using local optimization. (a) The population of $|1\rangle$ (solid line) and $|4\rangle$ (dashed line) vs time. (b) The population of the intermediate levels $|2\rangle$ (solid line) and $|3\rangle$ (dashed line) vs time. (c) The sequence of optical pulses found using the local optimization method. Note that the counterintuitive sequence of Stokes pulse (dot-dashed line) followed by pump pulse (solid line) again emerges automatically. However, now there is a third pulse, resonant with the $|2\rangle \rightarrow |3\rangle$ transition, which envelopes both these other pulses, and is about 10 times more intense. (d) Expanded trace of the Stokes and pump pulses. On this scale, the seed pulse at $t \approx 1$ is visible (see text).

is odd, the $|2\rangle \rightarrow |3\rangle$ transition is even, the $|3\rangle \rightarrow |4\rangle$ transition is odd, etc. The envelopes of all odd transitions are superimposed, and delayed relative to the envelopes for all even transitions; clearly, this is a natural generalization of the Stokes pump sequence in a three-level system. This alternating STIRAP (A-STIRAP) strategy works quite well; Fig. 8 shows that it is successful in transferring about 99% of the population from level $|1\rangle$ to level $|9\rangle$ in the nine-level system. However, note that the largest population in the intermediate levels rises to almost 30% in the A-STIRAP se-

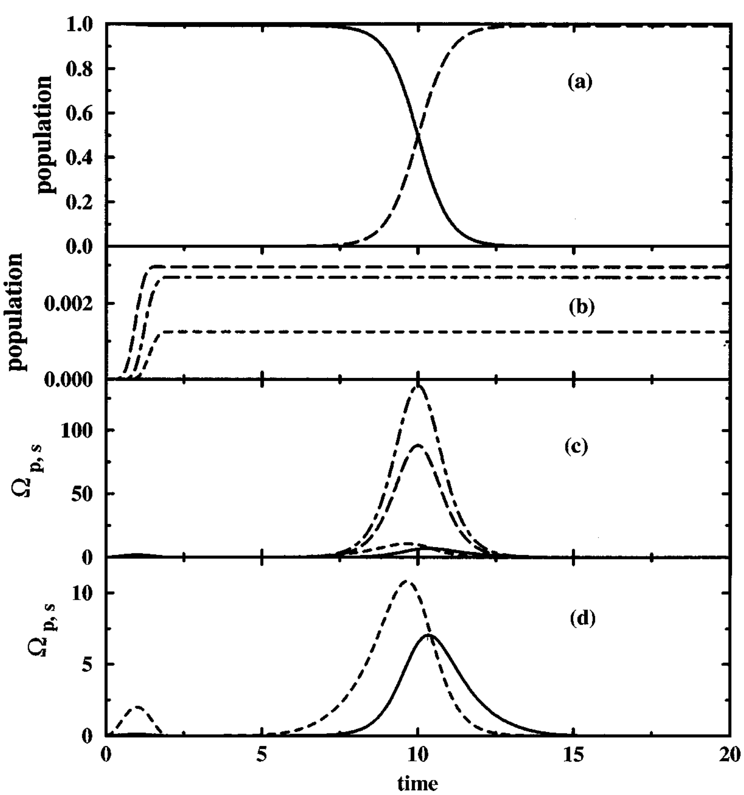

FIG. 6. Population transfer in a five-level, sequentially coupled system, using local optimization. (a) The population of $|1\rangle$ (solid line) and $|5\rangle$ (dashed line) vs time. (b) The population of the intermediate levels $|2\rangle$ (long-dashed line), $|3\rangle$ (dot-dashed line), and $|4\rangle$ (dashed line) vs time. (c) The sequence of optical pulses found using the local optimization method. The counterintuitive sequence of Stokes pulse (dashed line) followed by pump pulse (solid line) again emerges automatically. However, now there are two additional pulses, resonant with the $|2\rangle \rightarrow |3\rangle$ and $|3\rangle \rightarrow |4\rangle$ transition, which envelope the Stokes and pump pulses, and are about 10 times more intense. (d) Expanded trace of the Stokes and pump pulses. Again, the seed pulse at $t \approx 1$ is visible.

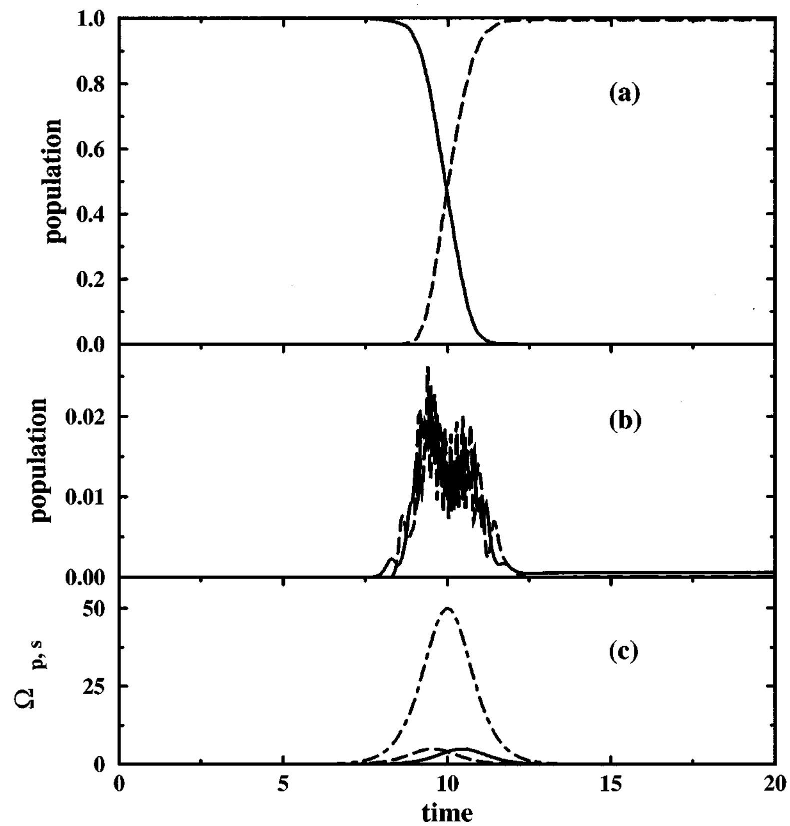

FIG. 7. Population transfer in a nine-level system with sequential coupling, using a straddling, or S-STIRAP pulse sequence. The properties of the S-STIRAP pulse sequence were abstracted from the local optimization of the four- and five-level systems: (1) the Stokes precedes the pump pulse, (2) pulses corresponding to transitions between all intermediate states straddle both the Stokes and pump pulse, and are given about 10 times the intensity of the latter. Note that there is no longer any seed pulse or attempt at local optimization. (a) The population of $|1\rangle$ (solid line) and $|9\rangle$ (dashed line) vs time. (b) The population of the intermediate levels $|4\rangle$ (solid line) and $|8\rangle$ (dot-dashed line), which receive the most population at intermediate times. Note that the population never exceeds 3% in any of the intermediate states. (c) The sequence of optical pulses in the S-STIRAP scheme. The intense straddling pulse actually consists of a superposition of 6 pulses with frequencies resonant with the intermediate transitions.

quence, as compared with 3% in the S-STIRAP sequence. In addition, the original A-STIRAP scheme works only when the number of levels N is odd; modifications must be made for each different even value of N on a case-by-case basis. For example, a recent paper by those authors dealt exclusively with the four-level system, and found that detuning from resonance with the $|2\rangle \rightarrow |3\rangle$ transition was essential [14]. To our knowledge, no general extension to higher val-

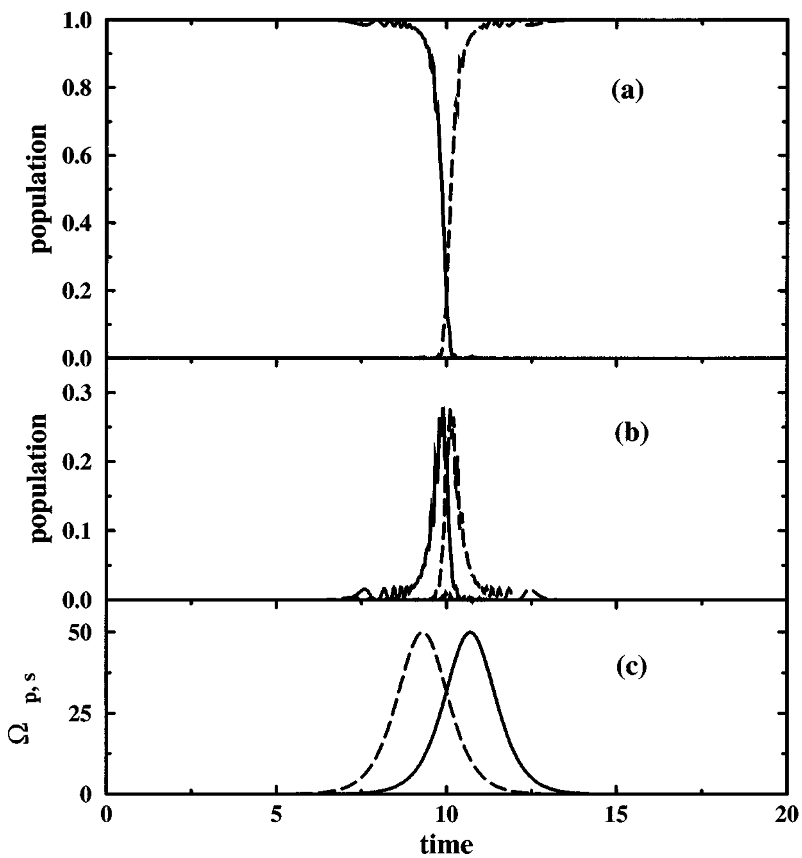

FIG. 8. Population transfer in a nine-level system with sequential coupling, using the A-STIRAP (alternating STIRAP) scheme of Ref. [12]. (a) The population of $|1\rangle$ (solid line) and $|9\rangle$ (dashed line) vs time. (b) The population of the intermediate levels $|3\rangle$ (solid line) and $|7\rangle$ (dot-dashed line), which receive the most population at intermediate times. Note that the population in these intermediate states approaches 30%, ten times more than in the S-STIRAP scheme. (c) The sequence of optical pulses in the A-STIRAP scheme. In the A-STIRAP sequence the pulses corresponding to all odd transitions $(|1\rangle \rightarrow |2\rangle, \ldots, |7\rangle \rightarrow |8\rangle)$ are given overlapping envelopes and delayed relative to the pulses corresponding to all even transitions $(|2\rangle \rightarrow |3\rangle, \ldots, |N-1\rangle \rightarrow |N\rangle)$. Thus, each of the envelopes in the figure is in fact 4 superimposed pulses.

ues of even N has been proposed. This underscores a significant advantage of the S-STIRAP sequence, which works equally well for odd or even N.

V. CONCLUSIONS

We have established a rigorous connection between stimulated Raman adiabatic passage (STIRAP) and local optimization of the time-dependent Schrödinger equation for 3 coupled levels. This is the first rigorous bridge between the tools being used in the growing literature of control theory applied to quantum mechanical systems and the efficient and robust STIRAP scheme. Since the local optimization technique is straightforward to implement for arbitrarily complicated N-level systems (with radiative decay, detuning, non-nearest-neighbor couplings, nonadiabatic evolution) one now has an automated method for computing robust STIRAP-type pulse sequences for arbitrary systems. We demonstrated this capability on a three-level system with radiative decay and on a four- and five-level system. The pulse sequences that emerge show the pump pulse is delayed relative to the Stokes pulse, but in addition, there are pulses corresponding to transitions between the $N-2$ intermediate states, with envelopes that span both the Stokes and pump pulse and are about an order of magnitude more intense. We call this a ''straddling'' STIRAP (or S-STIRAP) pulse sequence. Since the pattern is so general we tested it on a nine-level system without any optimization of parameters. Population transfer was 99%, with less than 3% in any of the intermediate states at any time during the process. This is about an order of magnitude less population on the intermediate states than in a recently proposed alternating STIRAP (A-STIRAP) scheme. Moreover, the S-STIRAP scheme works equally well for odd or even N, whereas the A-STIRAP works, in general, for odd N only. Thus, it seems that the S-STIRAP method is a promising candidate for a general and robust extension of STIRAP to sequentially coupled N-level systems.

We anticipate that there may be different generalizations of STIRAP that apply in more complicated situations. For example, in an N-level system with non-nearest-neighbor couplings, or with radiative decay, detuning, or nonadiabatic evolution, qualitatively different extensions of STIRAP may apply. To find these alternative generalizations, if they exist, the same general methodology as used in this paper may be followed, i.e., (1) apply the local optimization method to obtain a numerical solution to the problem; (2) test the robustness of the local optimization solution with respect to moderate changes in the pulse characteristics; (3) abstract the salient features of the local optimization pulse sequence and use these features to design pulse sequences *de novo*, abandoning any connection with the original optimization procedure.

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