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Multilevel adiabatic population transfer

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We present a theoretical description of a scheme in which coordinated laser pulses transfer population efficiently from any initially populated state (e.g., the ground state) to a multiply excited state of an atom or molecule without producing appreciable population in intermediate states. Our analytic results for multilevel excitation transfer provide a simple yet instructive extension to the counterintuitive pulse sequence studied previously for a three-state system.

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I. INTRODUCTION

For many applications in collision dynamics and spectroscopy, it is desirable to be able to transfer populations from an initial atomic or molecular state (e.g., ground state) to some specific excited state [1]. Coherent excitation by laser pulses [2,3] offers, in principle, a means of achieving complete population transfer. (Schemes based upon incoherent excitation will not be considered here.) The essential physics of such excitation derives from the time-dependent Schrödinger equation for a succession of nondegenerate levels (i.e., quantum states); inclusion of degeneracy (or Doppler broadening) requires a summation over atomic orientations (or detunings), and will not be considered in the present paper.

For a two-state system ($1 \leftrightarrow 2$) the desired population change ($1 \rightarrow 2$) may be produced by a resonant π pulse [4] or by a pulse with swept (chirped) frequency [3,5]. Although π pulses are simple to apply in theory, they are very sensitive to actual experimental conditions. In particular, averages over intensity profiles or atomic orientations (magnetic sublevels) make it impossible to induce the same excitation to each atom or molecule in a beam. Furthermore, it is difficult to prepare optical pulses that have precisely predetermined area. Chirped pulses do not have this disadvantage.

For a three-state chain ($1 \leftrightarrow 2 \leftrightarrow 3$) linked by two separate laser beams, complete transfer ($1 \rightarrow 3$) may be produced by a sequence of two resonant π pulses ($1 \rightarrow 2$ followed by $2 \rightarrow 3$). This procedure deals with multiple excitation as a sequence of independent two-level processes, and suffers from the same invalidating conditions. An alternative possibility, applicable when state 3 lies below state 2 (a λ system), is the stimulated emission process (SEP) [6], in which incoherent excitation into an excited state is accompanied or followed by deexcitation. It is now known that a very effective alternative to this "intuitive" pulse order is the "counterintuitve" scheme in which the first-step excitation pulse is applied after the second-step pulse [7]. This process, stimulated Raman scattering by delayed pulses (STIRAP) [1], is a unique combination of adiabatic following and a specific prescription for pulse timing that, taken together, move population between initial and final states without producing appreciable population in the intermediate state. Unlike SEP, the population transfer of STIRAP occurs coherently, and therefore the process requires the coherence of laser radiation.

Two particular three-state chains have been of interest: the simple ladder, in which each successive state lies higher in energy [8], and the Λ configuration [9], in which state 2 has greater energy than either state 1 or state 3. Ladder excitation has application to the excitation of Rydberg states or high-lying molecular vibrational states. A Λ configuration has application in transferring population into an excited molecular level, say, starting from level J'', moving into an electronic excited level $J'+1$, and subsequently returning to low-lying level $J''+2$. The λ configuration could also be used, e.g., to transfer molecular population to high-J states by successive cycles of $J_1 \rightarrow J_1+1 \rightarrow J_1+2$.

There are several possibilities for producing complete population transfer in an N-state ladder ($1 \leftrightarrow 2 \leftrightarrow \cdots \leftrightarrow N$). Conceptually the simplest is a succession of two-state π pulses that produces a succession of population inversions ($1 \rightarrow 2$, $2 \rightarrow 3, \ldots, N-1 \rightarrow N$). This coherent excitation procedure, as well as the corresponding scheme of excitation as a succession of three-

state systems, has the undesirable property of placing population into a succession of states, from each of which spontaneous emission may occur. Such emission acts as an effective loss mechanism and thereby diminishes the yield of the final-state population.

Another possibility is to employ simultaneous pulses of multiple nonresonant lasers, subject to the constraint of an (N-1)-photon resonance between states 1 and N. Such a procedure avoids populating intermediate states, but suffers from other disadvantages. In the limit of incoherent excitation (as described by rate equations), we can get, at most, only half the population into the excited state. In the alternative limit of coherent excitation, the population undergoes Rabi oscillations (at the multiphoton Rabi frequency) and the population transfer will be sensitive to the duration of the interaction (i.e., to the pulse area). In either case, the rate of transfer will be slow unless the laser intensity is very high: the atomic response is set by a multiphoton transition rate or a multiphoton Rabi frequency, and these are very small.

Other possibilities can be based on N-level excitation with chirped pulses, in which rapid adiabatic passage occurs via a succession of single-photon transitions. As noted in previous work [5,7], it is desirable to proceed in a "counterintuitive" manner in which resonance conditions occur in reverse order.

Here we suggest another alternative, possible when N is an odd integer, that has the potential advantage of large transition rates while avoiding population transfer into some of the intermediate states. Our procedure is based on a straightforward generalization of the theory for three-state excitation [5,8,9], subsequently experimentally verified [1,10]. Specifically, we obtain a null-eigenvalue solution to the instantaneous Hamiltonian for an N-state excitation ladder (with N an odd integer), subject to two-photon resonance constraints. When experimental conditions are such that this eigenstate remains a good approximation to an actual state vector (i.e., the adiabatic approximation applies), then we show that it is possible to produce complete population inversion without precise control of pulse area. Unlike the three-state results, however, our solutions place population briefly into some of the intermediate states.

We demonstrate the method by restricting consideration to two pulses (a "bottom" or "late" or "pump" pulse, and a "top" or "early" or "Stokes" pulse). We exhibit the analytic expressions for probability amplitudes of three-, five-, and seven-state systems. We show examples of our analytic expressions for the time-dependent Schrödinger equation and, for comparison, numerical solutions of the same equations. Differences between the analytic and numerical results, attributable to the breakdown of the adiabatic approximation, diminish as the pulses become more intense.

II. EQUATIONS OF MOTION

Consider the multistate Schrödinger equation for pulsed excitation in the rotating-wave approximation (RWA) by a succession of lasers in the absence of spontaneous emission [2]:

$$\frac{d}{dt}C_n(t)=-i\sum_m W_{nm}(t)C_m(t)\ . \tag{1}$$

Here $\hbar W$ is the (Hermitian) RWA Hamiltonian matrix, and the absolute square of the complex-valued amplitude $C_n(t)$ is the probability $P_n(t)$ of finding the system in state n at time t

$$P_n(t)=|C_n(t)|^2\ . \tag{2}$$

When states couple only to adjacent states, as they do for electric dipole transitions, the matrix W becomes tridiagonal and the probability amplitude equations read

$$\begin{aligned}
&\frac{d}{dt}C_1(t)=-i\Delta_1(t)C_1(t)-i\tfrac{1}{2}\Omega_1(t)C_2(t)\ ,\\
&\qquad\vdots\\
&\frac{d}{dt}C_n(t)=-i\Delta_n(t)C_n(t)-i\tfrac{1}{2}\Omega_{n-1}(t)C_{n-1}(t)\\
&\qquad\qquad -i\tfrac{1}{2}\Omega_n(t)C_{n+1}(t)\ ,\\
&\qquad\vdots\\
&\frac{d}{dt}C_N(t)=-i\Delta_N(t)C_N(t)-i\tfrac{1}{2}\Omega_{N-1}(t)C_{N-1}(t)\ .
\end{aligned} \tag{3}$$

Here $2W_{n,n+1}(r)\equiv\Omega_n(t)$ is the Rabi frequency for the transition between states n and $n+1$, and $W_{n,n}(t)\equiv\Delta_n(t)$ is the frequency mismatch between the nth Bohr frequency and the associated laser carrier frequency. Without loss of generality we take $\Delta_1(t)=0$, thereby making $\Delta_n(t)$ the cumulative detuning after $n-1$ excitation steps. We desire solutions for which $C_1(0)=1$ is the initial condition and which, at some final time t_∞, have $C_N(t_\infty)=1$. Such solutions describe complete population transfer between state 1 and state N.

III. STATIONARY ADIABATIC SOLUTION

Let us regard the Rabi frequencies $\Omega_n(t)$ and detunings $\Delta_n(t)$ as slowly varying functions of time (we clarify below the conditions that must be met for this adiabatic approximation to hold), and determine the stationary solutions to these equations at an arbitrary time t. That is, we set $(d/dt)C_n(t)=0$ for all n. This solution is a null-eigenvalue eigenstate of the RWA Hamiltonian evaluated at time t

$$\sum_n W_{nm}(t)C_m(t)=0\ , \tag{4a}$$

or

$$\begin{bmatrix}
0 & \Omega_1 & 0 & 0 & 0 & \cdots\\
\Omega_1 & 2\Delta_2 & \Omega_2 & 0 & 0 & \cdots\\
0 & \Omega_2 & 2\Delta_3 & \Omega_3 & 0 & \cdots\\
0 & 0 & \Omega_3 & 2\Delta_4 & \Omega_4 & \cdots\\
0 & 0 & 0 & \Omega_4 & 2\Delta_5 & \cdots\\
\cdots & \cdots & \cdots & \cdots & \cdots & \cdots
\end{bmatrix}
\begin{bmatrix} C_1\\ C_2\\ C_3\\ C_4\\ C_5\\ \cdots \end{bmatrix}
=0\ . \tag{4b}$$

We desire a solution for which the population remains as

small as possible in the intermediate states, $1<n<N$. It is evident that there exist solutions that place no population into state 2 at any time, $C_2(t)=0$. This property, which is well known for three-state systems, readily extends to all of the even n states: We can require solutions such that

$$C_{2m}(t)=0 \quad \text{for all } t \ . \tag{5}$$

It follows from this requirement that the odd n detunings Δ_n must vanish (those for even n remain arbitrary):

$$\Delta_{2m+1}(t)=0 \ . \tag{6}$$

That is, we require that the N-state ladder should behave like a sequence of two-photon resonances. This constraint ensures that the (virtual) populations in intermediate states (even n) will be negligible. Evidently N must be an odd integer for such a solution to be valid.

The relevant equations for odd-integer n are

$$\Omega_n(t)C_n(t)=-\Omega_{n+1}(t)C_{n+2}(t) \ . \tag{7}$$

From this formula we deduce that the succession of odd n amplitudes is given by the pattern

$$\begin{aligned} C_1(t)&=\Omega_2(t)\Omega_4(t)\Omega_6(t)\cdots\Omega_{N-1}(t)/S(t) \ , \\ C_3(t)&=-\Omega_1(t)\Omega_4(t)\Omega_6(t)\cdots/S(t) \ , \\ C_5(t)&=+\Omega_1(t)\Omega_3(t)\Omega_6(t)\cdots/S(t) \ , \\ &\vdots \end{aligned} \tag{8}$$

ending with a final state amplitude $C_N(t)$ that is the product of all odd n Rabi frequencies

$$C_N(t)=\pm\Omega_1(t)\Omega_3(t)\Omega_5(t)\cdots\Omega_{N-1}(t)/S(t) \ . \tag{9}$$

The number $S(t)$ is the normalization, obtained by summing the squares of the various combinations of Rabi frequencies.

$$\begin{aligned} S=&|\Omega_2\Omega_4\Omega_6\cdots|^2 \\ &+|\Omega_1\Omega_4\Omega_6\cdots|^2+\cdots|\Omega_1\Omega_3\Omega_5\cdots|^2 \ . \end{aligned} \tag{10}$$

The succession of nonzero components can also be written as

$$\begin{aligned} &C_1(t) \ , \\ &C_3(t)=\mathcal{R}_1(t)C_1(t) \ , \\ &C_5(t)=\mathcal{R}_3(t)\mathcal{R}_1(t)C_1(t) \ , \\ &\vdots \end{aligned} \tag{11}$$

where

$$\mathcal{R}_n(t)=-\Omega_n(t)/\Omega_{n+1}(t) \ . \tag{12}$$

These odd n amplitudes, combined with null even n amplitudes, provide components of a null-eigenvalue eigenstate of the RWA Hamiltonian $\hbar W(t)$. For the three-state Λ system, this eigenstate is the well-studied "population-trapping" state [11], a coherent superposition state that has no admixture of the population-losing amplitude $C_2(t)$. This state is selectively populated in the experiments that have demonstrated complete population transfer [8,10].

IV. PROCEDURE FOR POPULATION TRANSFER

Our generalization of the Oreg-Bergmann method for complete population transfer consists of devising a set of slowly varying (adiabatic) pulses that have the following properties.

(i) Prior to the initial time $t=0$, all Rabi frequencies vanish.

(ii) Subsequent to the initial time $t=0$, the odd-n Rabi frequencies $\Omega_1, \Omega_3, \ldots$ remain zero, while each of the even-n Rabi frequencies $\Omega_2, \Omega_4, \ldots$ becomes nonzero. Symbolically we require

$$\Omega_{\text{odd}} \ll \Omega_{\text{even}} \ .$$

(iii) At some later time, all even-n Rabi frequencies vanish while the odd-n Rabi frequencies do not. Symbolically we require

$$\Omega_{\text{odd}} \gg \Omega_{\text{even}} \ .$$

(iv) Eventually, at some large time $t=t_\infty$, the odd-n Rabi frequencies also vanish; there are no excitation fields present.

(v) The pulses must at all times maintain connection to permit population flow $1\rightarrow 3\rightarrow 5\rightarrow\cdots\rightarrow N$; i.e., there must be an interval of adequate length during which the product of all Rabi frequencies is not zero. Ideally Ω_{odd} will be diminishing while Ω_{even} is rising.

If such a pulse sequence can be constructed while maintaining conditions of adiabatic change, then the initial-state vector has the single component $C_1(t)$ and the final-state vector has the single component $C_N(t)$; components $C_{2m}(t)$ will remain zero at all times. At each step of the excitation the rate of upward population flow is governed by a single Rabi frequency, not by a two-photon (or more general multiphoton) Rabi frequency.

V. EXAMPLES

A simple example of such a pulse sequence is one for which all even-n Rabi frequencies have a common time dependence $f(t)$, while all odd-n Rabi frequencies have a common time dependence $g(t)$,

$$\Omega_2(t)=\Omega_4(t)=\cdots=\Omega_0 f(t), \quad f(0)\neq 0, \quad f(t_\infty)=0 \ , \tag{13a}$$

$$\Omega_1(t)=\Omega_3(t)=\cdots=\Omega_0 g(t), \quad g(0)=0, \quad g(t_\infty)\neq 0 \ . \tag{13b}$$

Such a scheme allows complete population inversion, bypassing even-n intermediate states but briefly populating odd-n intermediate states.

For three states the amplitudes are those considered in earlier works,

$$C_1(t)=\frac{f(t)}{S(t)}, \quad C_3(t)=-\frac{g(t)}{S(t)} \ , \tag{14a}$$

$$S(t)^2=|f(t)|^2+|g(t)|^2 . \tag{14b}$$

This solution predicts population flow from $P_1=1$ when $g=0$, through $P_1=P_3$ when $f=g$, to a final asymptote $P_3=1$ as $f\to 0$. This is the "counterintuitive" three-state excitation, in which the second-step excitation pulse is applied *before* the first-step excitation pulse. The ratio $f(t)/S(t)$ may be viewed as an angle, the mixing angle.

For five states the amplitudes are

$$C_1(t)=\frac{f(t)^2}{S(t)},\quad C_3(t)=-\frac{g(t)f(t)}{S(t)},\quad C_5(t)=\frac{g(t)^2}{S(t)}, \tag{15a}$$

$$S(t)^2=|f(t)|^4+|f(t)|^2g(t)|^2+|g(t)|^4 . \tag{15b}$$

This solution requires two mixing angles. For seven states the amplitudes are

$$C_1(t)=\frac{f(t)^3}{S(t)},\quad C_3(t)=-\frac{g(t)f(t)^2}{S(t)},$$
$$C_5(t)=\frac{g(t)^2f(t)}{S(t)},\quad C_7(t)=-\frac{g(t)^3}{S(t)}, \tag{16a}$$

$$S(t)^2=|f(t)|^6+|f(t)|^4|g(t)|^2 +|f(t)|^2|g(t)|^4+|g(t)|^6 . \tag{16b}$$

In both these latter cases, as well as for larger systems, appreciable population will briefly appear in an intermediate state. This population will be greatest when $f=g$. For the five-state system the intermediate-state population is largest, $P_3=\frac{1}{3}$, when $f=g$, while for the seven-state system the intermediate states have the greatest populations $P_3=P_5=\frac{1}{4}$. Whether the population present in these intermediate states poses a practical limitation on achievable population transfer will depend on the rate at which radiative or collisional processes remove population from these states. The time scale for such detrimental processes should be much longer than the population transfer time.

VI. NUMERICAL EXAMPLES

To illustrate the preceding solutions we consider the pulse shape

$$p(t)=\begin{cases}0 & \text{for } t<0\\ \sin^4(\pi t/T) & \text{for } 0<t<T\\ 0 & \text{for } t>T ,\end{cases} \tag{17}$$

and we choose the two pulses $f(t)$ and $g(t)$ to be offset in time by an increment τ,

$$f(t)=p(t),\quad g(t)=p(t+\tau) . \tag{18}$$

This shape turns on and off smoothly (as contrasted with linear ramps) and has finite duration (as contrasted with a Gaussian).

To parametrize the field strength one might choose peak pulse intensity or total pump fluence, together with a specification of the dipole moment. It proves more convenient for present purposes to specify the traditional pulse area,

$$A=\int_0^\infty dt\,\Omega_0 p(t)=\frac{3\pi}{8}\Omega_0 T . \tag{19}$$

For a single-photon transition this area is the traditional tipping angle: it has the value π for a pulse that produces complete two-state inversion (a π pulse). With these choices our numerical model depends on only two param-

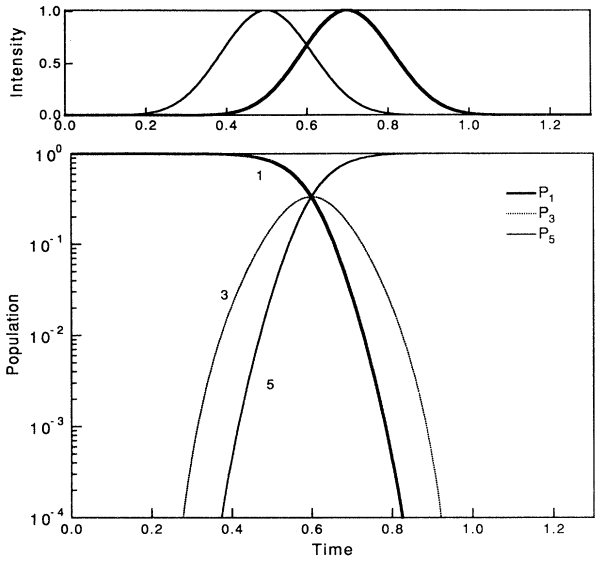

FIG. 1. Analytic solutions for five-state excitation by two pulses, each of duration $T=1$, whose delay is $\tau=0.2T$. Top frame: intensities of the two pulses vs time, normalized to unit peak value. Bottom frame: populations vs time.

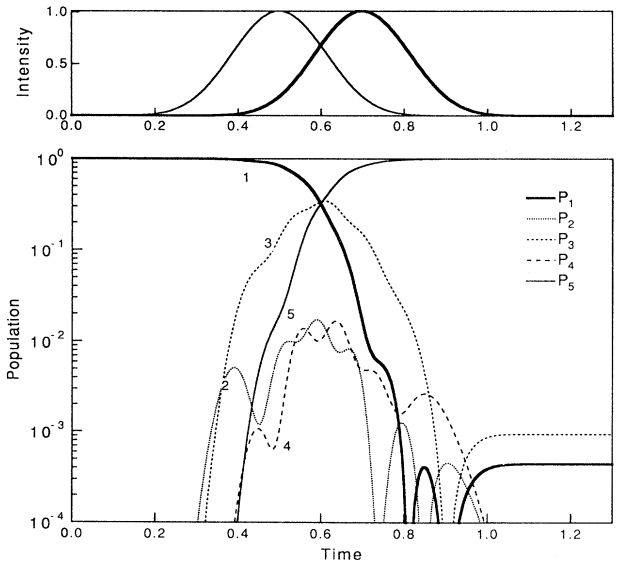

FIG. 2. Numerical solutions for five-state excitation, as in Fig. 1. The pulse area is $A=12\pi$.

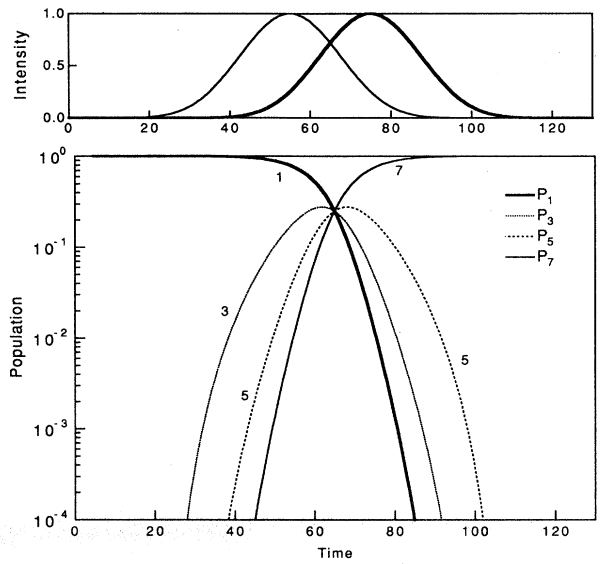

FIG. 3. Analytic solutions for seven-state excitation for the pulses of Fig. 1. Top frame: intensities of the two pulses vs time. Bottom frame: populations vs time.

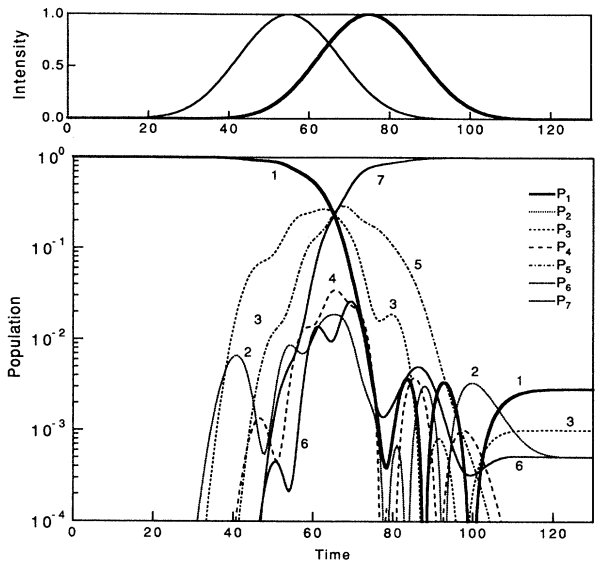

FIG. 4. Numerical solutions for seven-state excitation for pulses with area $A = 12\pi$, as in Fig. 3.

eters: pulse delay τ and pulse area A. We take the pulse duration as our unit of time, by setting $T = 1$.

Figure 1 shows an example, for a five-state system, of the analytic solutions for excitation by these pulses, in the adiabatic approximation. The upper frame shows the pulse intensities, $|f(t)|^2$ and $|g(t)|^2$, while the lower frame shows the several populations $P_n(t)$. In the upper frame the thick line indicates the pulses that couple states 1 and 2 as well as states 3 and 4 (the "bottom" pulse); the thin line refers to the radiation that couples states 2 and 3 as well as states 4 and 5 (the earlier "top" pulse). In the lower frame the thick line denotes the population in the ground state (state 1).

For comparison, Fig. 2 shows the solutions obtained by numerical integration of the Schrödinger equation for intensity such that the pulse area is 12π. The populations of odd-n states are nearly the same in the two figures, but the numerical solutions show small populations in even-n states. These deviations from the adiabatic approximation will be present in the actual response of a five-level system to such pulses; they represent the failure of the analytic solutions to mimic faithfully the excitation produced by these pulses. For the computations displayed here, these even-state populations remain, at all times, less than 1% of the total population. The population in state 3 is always less than $\frac{1}{3}$. More intense pulses produce excitation that follows the analytic expressions more closely (when $A = 20\pi$ the largest even-n populations are about one-third as large as shown here, and when $A = 40\pi$ the even-n populations are less than 0.1%). With appreciably weaker pulses the adiabatic approximation fails and the population transfer becomes incomplete (when $A = 4\pi$ the even-n populations are as large as 10% and the population transfer is only 87%). Failure of the adiabatic approximation means that the actual state vector is a combination of dressed states, rather than a single dressed state. The oscillatory behavior of the numerical solutions is the expected interference between their components.

Figure 3 shows the analytic solutions for a seven-state atom, in the adiabatic approximation, for these same pulses. Figure 4 shows the numerical results (again for $A = 12\pi$). Again the differences between these two figures, particularly the occurrence of small portions of population in even-n states, indicates the inaccuracy of the adiabatic approximation as a faithful description of the actual excitation. It will be observed that the 12π pulses with a seven-state system (Fig. 4) are noticeably less adiabatic than are 12π pulses with a five-state system (Fig. 2). As the number of states increase it is necessary to increase the pulse areas if one wishes to maintain the same small value of intermediate-state population. For the example of Fig. 4, an increase of pulse area to 20π will produce results comparable to Fig. 2.

VII. ADIABATIC CONDITION

Conditions for applicability of the adiabatic approximation in these kind of problems have been discussed previously [1]. A sufficient condition is the so-called adiabatic criterion: the rate of change of the mixing angle is less than the separation of the dressed eigenvalues. Although the adiabatic criterion is a sufficient condition, it is not also a necessary condition, and so the breakdown of the criterion does not always mean the breakdown of the adiabatic approximation. In particular, the criterion is most restrictive at the beginning and end of the pulse sequence (where it is known to fail), but at those times the breakdown has the least effect on population transfer. Generalization of this to multiple mixing angles (i.e.,

multiple ratios of pulse products) offers a possible guide to the applicability of the adiabatic approximation to larger systems: we require that none of the amplitudes change rapidly compared with the separation of instantaneous dressed states.

VIII. SUMMARY

The present paper examines analytical and numerical results for a class of solutions to the nondegenerate time-dependent Schrödinger equation. The essence of the proposed scheme is the timing of coherent excitation pulses to create dressed states, by application of a first set of fields, and the subsequent transfer of population by application of a second set of fields. It is not difficult to produce complete population transfer, just as in traditional two-state adiabatic-following processes. In all cases the two pulses should be such that the earlier "top" pulse is declining while the later "bottom" pulse is rising. The procedure does not require precise timing nor does it require restrictive conditions on pulse area; the amount of population transfer is not sensitive to moderate changes in pulse fluence or pulse shape. These properties are important for experimental implementation of the procedure. The pulses may be made resonant with the transitions, but this condition is not necessary. The simple analytic solutions, valid in the adiabatic approximation, provide an interesting class of multistate solutions to the time-dependent Schrödinger equation.

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[1] U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, J. Chem. Phys. **92**, 5363 (1990); U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988); B. W. Shore, *The Theory of Coherent Atomic Excitation* (Ref. [2]), Sec. 1.2.

[2] B. W. Shore, *The Theory of Coherent Atomic Excitatin* (Wiley, New York, 1990).

[3] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[4] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Ref. [2]), Sec. 5.1; L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Ref. [3]), Sec. 3.2.

[5] J. Oreg, F. T. Hioe and J. H. Eberly, Phys. Rev. A **29**, 690 (1984); B. W. Shore, *The Theory of Coherent Atomic Excitation* (Ref. [2]), Sec. 5.6.

[6] C. Kitrell, E. Abramson, J. L. Kinsey, S. A. McDonald, D. E. Reisner, R. W. Field, and D. H. Katyama, J. Chem. Phys. **75**, 2056 (1981); C. Hamilton, J. L. Kinsey, and R. W. Field, Ann. Rev. Phys. Chem. **37**, 493 (1986); X. Yang and A. M. Wodtke, J. Chem. Phys. **92**, 116 (1990); See also J. Opt. Soc. Am. B **7**, 9 (1990), devoted to molecular spectroscopy and dynamics by stimulated-emission pumping; G. Z. He, A. Kuhn, S. Schiemann, and K. Bergmann, J. Opt. Soc. Am. B **7**, 1960 (1990); A. Kuhn, G. W. Coulston, G. Z. He, S. Schiemann, K. Bergmann, and W. S. Warren (unpublished).

[7] J. Oreg, G. Hazak, and J. H. Eberly, Phys. Rev. A **32**, 2776 (1985).

[8] C. Liedenbaum, S. Stolte, and J. Reuss, Phys. Rep. **178**, 1 (1989); J. Reuss and N. Dam, in *Applied Laser Spectroscopy,* edited by W. Demtroeder and M. Inguscio (Plenum, New York, 1990); N. Dam, S. Stolte, and J. Reuss, Chem. Phys. **135**, 437 (1989).

[9] J. R. Kuklinski, U. Gaubatz, F. T. Hioe, and K. Bergmann, Phys. Rev. A **40**, 6741 (1989).

[10] H. G. Rubahn, E. Konz, S. Schiemann, and K. Bergmann, Z. Phys. D (to be published).

[11] H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (1978); P. M. Radmore and P. L. Knight, J. Phys. B **15**, 561 (1982).