Numerical studies of adiabatic population inversion in multilevel systems

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Received September 27, 1991; revised manuscript received January 13, 1992

A numerical analysis is presented of several potential pulsed-laser applications of adiabatic population inversion in atoms or molecules. A simple numerical method is demonstrated for evaluating adiabatic processes in multilevel systems for both swept-frequency and resonant, time-delayed pulses. Adiabatic inversion with time-delayed pulses is generalized to systems with any number of levels. The effect of Doppler broadening on time-delayed adiabatic pumping of a three-level system is investigated.

1. INTRODUCTION

In a resonantly driven two-level atom or molecule, it is possible to obtain complete population transfer from the lower to the upper energy level by the application of a driving pulse of area π.[1] This concept can be extended to multilevel systems. Pulses that are resonant at each step can be applied sequentially or simultaneously, and, if they have the correct areas, the population can be transferred completely from the initial to the final state. The drawback of this method is that it requires precise control of pulse areas. At the cost of somewhat increased driving intensity, the method of adiabatic pumping can accomplish the same thing with relaxed constraints on pulse areas. For a two-level system this is achieved if one sweeps the frequency of the driving field through the resonance slowly enough that its rate of change is small compared with the square of the Rabi frequency but fast enough that damping is insignificant. This is treated theoretically in Ref. 1, among many others, and has been experimentally demonstrated for optical transitions.[2–4] Recently the extension of adiabatic pumping methods to multilevel systems has received theoretical and experimental attention. Oreg *et al.*[5] generalized the rotating vector model that Feynman *et al.*[6] developed for driven, near-resonant, two-level systems to an N-level system with each step driven near resonance. Unlike the case in two-level systems, there are many adiabatic paths to inversion in such systems. For example, Oreg *et al.* showed that, for three levels, appropriate timing of constant-frequency pulses leads to adiabatic population transfer. Carroll and Hioe[7] developed analytic solutions for three-level systems, demonstrating this result in more detail. This method has been experimentally verified by the authors in Ref. 8 and by Liedenbaum *et al.*,[3] who used cw lasers pumping molecular beams. For certain systems of an arbitrary number of levels, Hioe and Carroll[9] showed that adiabatic pumping is possible and presented analytic solutions. There are many possible inversion pathways even for the three-level system, however, and identifying them is not trivial. A simple numerical procedure is presented here for assessing methods of pumping based on either swept frequencies or time delays for N-level systems, and several examples are given. Adiabatic behavior is verified by simulation of the time development of populations through numerical integration of multilevel, optical Bloch equations. Finally, the effects of Doppler broadening on adiabatic pumping with time-delayed pumping are considered, and limitations inherent in non-cascade-coupled systems are explored.

Continuing improvements in tunable, pulsed, visible-wavelength lasers have made it possible to generate temporally smooth and transform-limited-bandwidth light pulses suitable for adiabatically pumped short-lived electronic levels of atoms and molecules. Additionally, using semiconductor lasers as oscillators and pulsed, laser-pumped amplifiers, one should be able to obtain the required frequency sweep characteristics. Therefore adiabatic pumping using 5-ns Gaussian time-profile pulses is considered here. For this pulse length the pulse fluences required are in the range of microjoules per square centimeter for dipole allowed transitions. In the discussion in this paper it is assumed that the effect of the atoms or the molecules on the light pulses is negligible, and dampling resulting from finite lifetimes or dephasing of the induced coherence is neglected.

2. TWO-LEVEL SYSTEM

Before multilevel systems are discussed, it is instructive to consider adiabatic pumping in a two-level system. Figure 1 shows the evolution of the adiabatic, or dressed-state, energy levels of a two-level system as a constant-amplitude driving field is swept from red ($\Delta < 0$) to blue ($\Delta > 0$) through the resonance. The isolated atom has energies e_1 (lower) and e_2 (upper). The $|1\rangle$ and $|2\rangle$ labels will refer to the product states $|\text{lower}\rangle|n\rangle$ and $|\text{upper}\rangle|n-1\rangle$, respectively, where n is the number of photons in the driving wave.[10] Far from resonance the adiabatic eigenstates have energies $e_1 + nh\nu$ and $e_2 + (n-1)h\nu$ and are simply $|1\rangle$ and $|2\rangle$, respectively. Their energy difference is Δ, the detuning of the driving field. In the absence of coupling between the driving wave and the atom, the eigenstates would become degenerate when Δ reaches zero. With coupling, the adiabatic energy levels are the eigenvalues of the interaction Hamiltonian[1]

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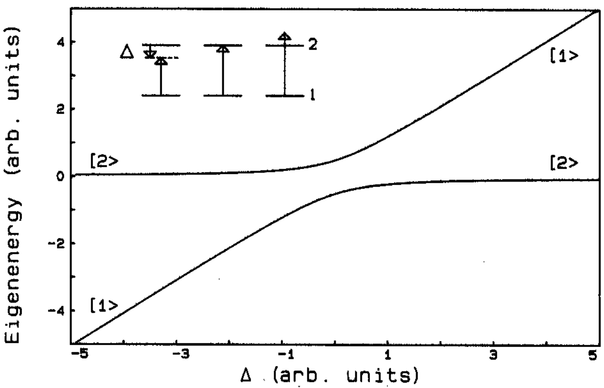

Fig. 1. Adiabatic energy levels for a two-level system as a function of the detuning Δ of the driving radiation from resonance.

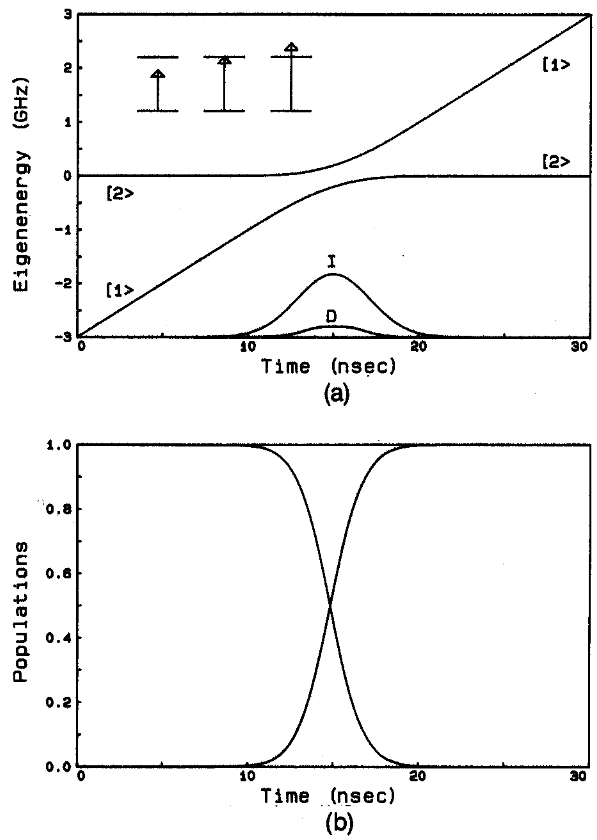

Fig. 2. (a) Adiabatic energy levels as a function of time for a 5-ns FWHM driving pulse frequency swept to the blue at 200 MHz/ns with a pulse area of 6π. Curve I is the time profile of the drive pulse plotted in arbitrary units. Curve D is explained in the text. (b) Population in the upper and lower levels as a function of time for the same conditions as in (a).

$$H = \hbar \begin{bmatrix} \Delta & \Omega/2 \\ \Omega/2 & 0 \end{bmatrix}, \tag{1}$$

where $|\Omega|$ is the Rabi frequency. For convenience, the energy of $|2\rangle$ is defined as the zero of energy here and throughout this paper. For electric dipole coupling of the field to the atom, $\Omega \equiv \boldsymbol{\mu} \cdot \boldsymbol{\epsilon}/\hbar$, where μ is the atomic dipole matrix element and ϵ is defined by the expression for the driving electric field, $\mathbf{E} = 1/2[\boldsymbol{\epsilon} \exp(-i\omega t) + \boldsymbol{\epsilon}^* \exp(i\omega t)]$. For the purpose of this discussion, Ω may be taken to be real without loss of generality. Thus, for $\Delta = 0$, the coupling removes the degeneracy and splits the eigenenergies by the Rabi frequency Ω, and the eigenstates corresponding to the upper and lower eigenenergies are $(|1\rangle + |2\rangle)/\sqrt{2}$ and $(|1\rangle - |2\rangle)/\sqrt{2}$, respectively.

From Fig. 1 it is apparent that, if the atom is initially in level $|1\rangle$ and adiabatically follows the lower curve as the driving field is scanned through the resonance, it will finish in level $|2\rangle$. From the Landau–Zener[11] theory of avoided crossings, the probability of transferring from the lower to the upper curve during the sweep through the transition is $\exp(-2\pi\gamma)$, where $\gamma = (\Omega/2)^2(\mathrm{d}\Delta/\mathrm{d}t)^{-1}$. Thus, if the sweep rate $\mathrm{d}\Delta/\mathrm{d}t$ is much less than the square of the Rabi frequency, the system will follow the lower curve, and complete population inversion will be achieved.

For a swept-frequency driving wave that is pulsed rather than constant in amplitude, the situation is substantially the same. If the detuning is large enough that the product state $|1\rangle$ is a good approximation to the lower-energy eigenstate when the light pulse turns on, and if the tuning rate is slow, one can expect adiabatic inversion. Figure 2 shows an example of a pulse with a Gaussian time profile (5-ns FWHM) and a frequency sweep rate of 200 MHz/ns. Exact resonance and peak intensity both occur at a time of 15 ns. The trace labeled I represents the intensity profile of the pulse whose area is chosen to be 6π, i.e., $\int \mathrm{d}t \mu\epsilon(t)/\hbar = 6\pi$. While analytic expressions can be derived for the dressed states and the eigenenergies as a function of Δ or time, numerical procedures are used here because they are quite simple to implement and they

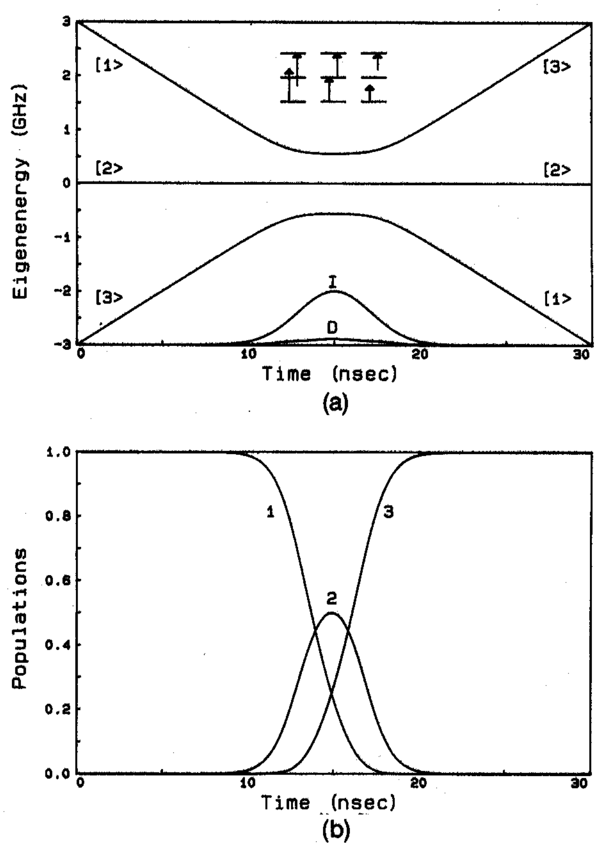

Fig. 3. (a) Adiabatic energy levels as a function of time for 5-ns FWHM synchronous drive pulses frequency swept to the red at 200 MHz/ns for both transitions with pulse areas of 12π. D is the diabatic term defined in the text. (b) Populations as a function of time for the same conditions as in (a).

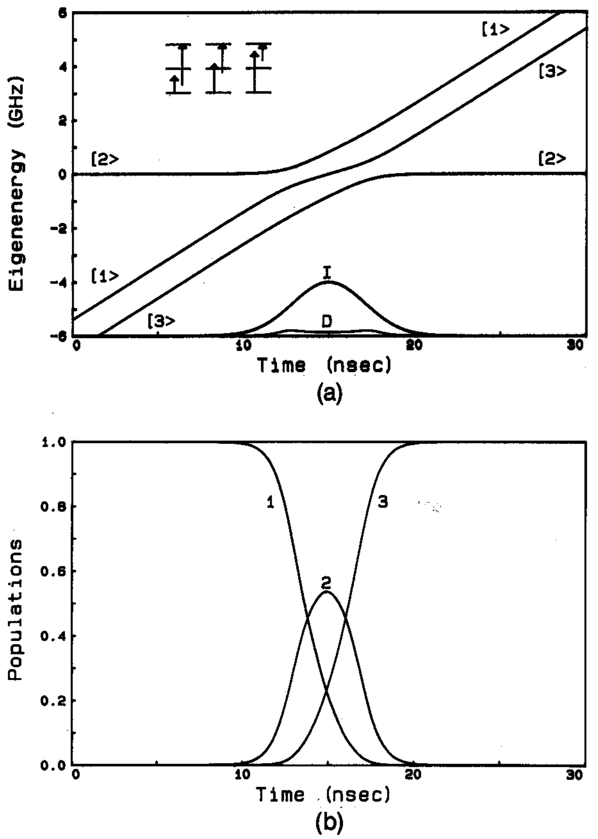

Fig. 4. (a) Adiabatic energy levels as a function of time for 5-ns FWHM synchronous drive pulses of area 12π frequency swept to the blue at 400 MHz/ns for the lower transition and to the red at 400 MHz/ns for the upper transition, with the lower transition reaching resonance first. D is the diabatic term defined in the text. (b) Populations as a function of time for the same conditions as in (a).

can be extended to more-complex multilevel systems. The upper plot of Fig. 2(a) shows the numerically calculated eigenenergies of the Hamiltonian of Eq. (1). Here Δ is varied linearly in time to describe the frequency chirp,[12] and Ω has the Gaussian time profile of the pulsed light field. The lower plot shows the time development of the populations in the two product states $|1\rangle$ and $|2\rangle$ computed by numerical integration of the optical Bloch equation[13]

$$\mathrm{d}\rho/\mathrm{d}t = -(i/\hbar)[H, \rho], \tag{2}$$

where ρ is the atomic density matrix and H is the interaction Hamiltonian. For the conditions chosen here, the value of γ in the Landau–Zener formula has a maximum value of ~10 at $t = 15$ ns, so the behavior should be adiabatic. However, that formula does not strictly apply to this case of time-varying coupling strength Ω. The more general requirement[14] for the adiabatically following eigenstate Ψ_1 is that

$$D = \sum_n \left[\frac{(\mathrm{d}\langle\Psi_1|/\mathrm{d}t)|\Psi_n\rangle}{e_1 - e_n}\right]^2 \ll 1, \tag{3}$$

where e_n is the eigenenergy of the nth eigenstate Ψ_n. The diabatic factor D is plotted as the curve labeled D in Fig. 2(a). Here and throughout this paper it is multiplied by 100 and shifted from zero to the lower axis of the figure for clarity. It has a maximum value that is less than 0.01.

From the analytic form for the adiabatic eigenstates for this two-level system, it is easy to show that inequality (3) reduces to

$$\mathrm{d}\Delta/\mathrm{d}t \ll 2(\Omega^2 + \Delta^2)^{3/2}/\Omega. \tag{4}$$

For our example, D is maximum when Δ is zero, in which case inequality (4) becomes $\mathrm{d}\Delta/\mathrm{d}t \ll 2\Omega^2$, in agreement with the Landau–Zener formula.

3. THREE-LEVEL SYSTEMS

For multilevel systems, adiabatic pathways exist for various combinations of frequency sweep rates and directions and for various pulse sequences. The methods presented in Section 2 for the two-level system provide a convenient procedure for evaluating the possibilities. For example, for a three-level system driven by two light pulses the interaction Hamiltonian is

$$H = \hbar\begin{bmatrix} \Delta_1 & \Omega_1/2 & 0 \\ \Omega_1/2 & 0 & \Omega_2/2 \\ 0 & \Omega_2/2 & \Delta_2 \end{bmatrix}, \tag{5}$$

where a subscript 1 refers to the lower transition and a subscript 2 refers to the upper transition. Figures 3–5 show three examples of adiabatic pumping with swept-frequency light and synchronous pulses. In Fig. 3 the two

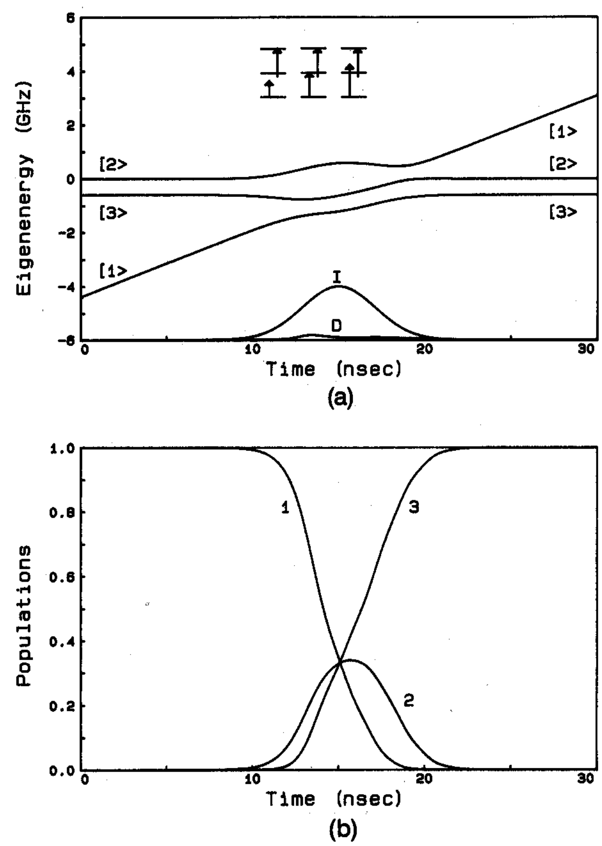

Fig. 5. (a) Adiabatic energy levels as a function of time for 5-ns FWHM synchronous drive pulses of area 18π, with the lower drive frequency swept at 250 MHz/ns to the blue and the upper drive frequency fixed at 600 MHz blue. D is the diabatic term defined in the text. (b) Populations as a function of time for the same conditions as in (a).

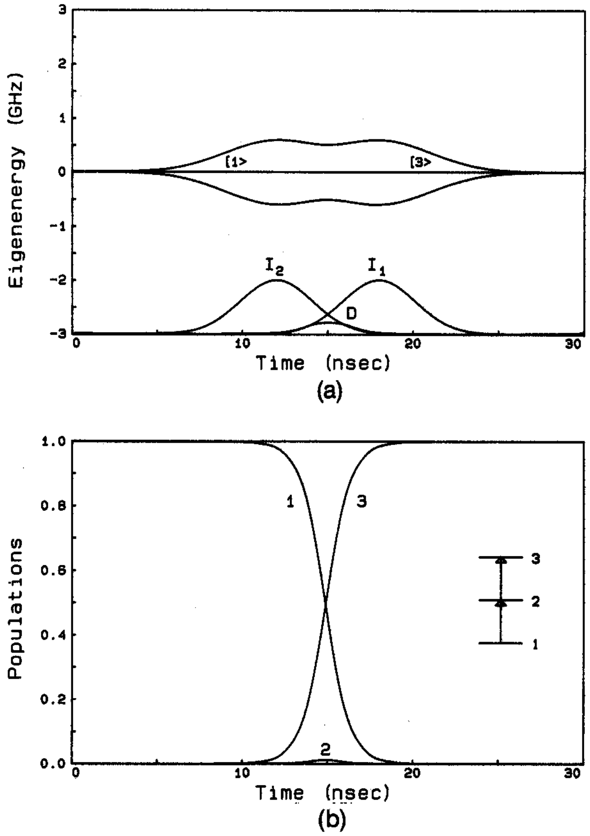

Fig. 6. (a) Adiabatic energy levels as a function of time for 5-ns FWHM asynchronous, resonant drive pulses of area 18π. The time delay is 6 ns, with the upper transition (I_2) driven first. D is the diabatic term defined in the text. (b) Populations as a function of time for the same conditions as in (a).

light frequencies are swept in the same direction at a rate of 200 MHz/ns, so they reach resonance simultaneously at the center of the pulses. Curve I shows the shape of the light pulses, both with an area of 12π. These conditions are chosen so that the maximum value for the diabatic factor D satisfies the condition $D \leq 0.01$, as in the two-level case. Empirically, it is found that compliance ensures adiabatic behavior.

To identify the energy levels at late time with the unperturbed state $|1\rangle$, $|2\rangle$, or $|3\rangle$, one makes the linear continuation of the early-time curves. Thus the adiabatic curves of Fig. 3 show that atoms initially in the ground state ($|1\rangle$) are pumped to the highest energy state ($|3\rangle$), those in the intermediate state remain there, and those in the highest state are transferred to the ground state. This procedure of computing eigenenergies as a function of time and associated initial and final eigenstates with the product states $|1\rangle$, $|2\rangle$, etc., along with evaluation of the diabatic term D, permits a quick assessment of candidate pumping schemes. Figure 3(b) confirms the desired adiabatic population transfer. It shows the time evolution of the population under the assumption that the atoms are all initially in the ground state. In Fig. 4 the pulses are coincident in time and the light frequencies are scanned in opposite directions at 400 MHz/ns, with the first resonance reached before the second resonance. The pulse areas are given 12π. In this case, $|1\rangle$ goes to $|3\rangle$ as before, but $|2\rangle$ goes to $|1\rangle$, while $|3\rangle$ goes to $|2\rangle$.

The process of Fig. 5 also takes the population from $|1\rangle$ to $|3\rangle$. Here only one frequency is swept at 250 MHz/ns, and the pulse areas are 18π. The fixed driving frequency for the upper transition is 600 MHz blue of resonance, while at the maximum intensity point the swept frequency is 650 MHz red of resonance with the lower transition. As Fig. 5(b) shows, the result is again well-behaved adiabatic transfer of the ground-state population to level 3.

These three examples illustrate the general process of adiabatic transfer for swept frequencies, which is a subset of the more general process of adiabatic transfer as described by Oreg *et al.*[5] One alternative that they pointed out is the use of time-delayed pulses. This method is attractive because it does not require frequency-chirped lasers. Figure 6 shows an example. Here the resonant, Gaussian profile (5-ns FWHM) pulses are offset by 6 ns, with the *upper* transition driven first. Each pulse has an area of 18π. As has been pointed out,[7,8] one of the eigenenergies for this system is zero at all times and is associated with the eigenstate

$$\Psi_0 = (\Omega_2|1\rangle - \Omega_1|3\rangle)/(\Omega_1^2 + \Omega_2^2)^{1/2}. \tag{6}$$

At early time, when only the first pulse is on, $|2\rangle$ and $|3\rangle$ are coupled to form the two states shifted up and down in energy, respectively, while the zero-energy eigenstate is $|1\rangle$. For late time, states $|1\rangle$ and $|2\rangle$ are coupled to form the shifted states and the zero-energy eigenstate is $|3\rangle$. Thus, in passing through the pulse sequence, the population in $|1\rangle$ is transferred to $|3\rangle$. It is interesting to note that $|2\rangle$ does not appear in Ψ_0, so $|2\rangle$ is never populated for a perfectly adiabatic transition. Figure 6(b) shows a small $|2\rangle$ population, owing to the slightly diabatic nature of the transition under these conditions.

This method has been demonstrated experimentally[3,8] with the use of cw lasers to excite molecules in a beam. The laser foci were spatially displaced, so the flight of the molecules provided the proper time delays.

As others have pointed out,[3,7,8] the intermediate level can be slightly detuned from the one-photon level without a qualitative change in this behavior. If two-photon resonance is maintained, but the first photon is blue of resonance, the upper curve and the line of Fig. 6(a) will be displaced upward by an amount equal to the detuning. In this case the upper curve can be associated with $|3\rangle$ at early time and $|1\rangle$ at late time, while the intermediate line is associated with $|1\rangle$ early and $|3\rangle$ late. It is interesting to note that, if the pulse sequence is reversed, $|1\rangle$ still goes to $|3\rangle$, so adiabatic transfer from the lowest to the highest energy state can be realized with either pulse sequence for certain values of one-photon detuning. However, when the lower transition is pumped first, the population of the intermediate level is not suppressed as it is when the upper transition is pumped first.

4. *N*-LEVEL SYSTEMS

For systems of more than three levels the number of adiabatic pathways increases, of course. Using the methods outlined in Section 3 for the three-level system, one can find numerous examples. For example, the method of Fig. 3 above can be generalized to scanning the lowest and highest transitions in the same direction, with all the intermediate transitions resonantly pumped or the method

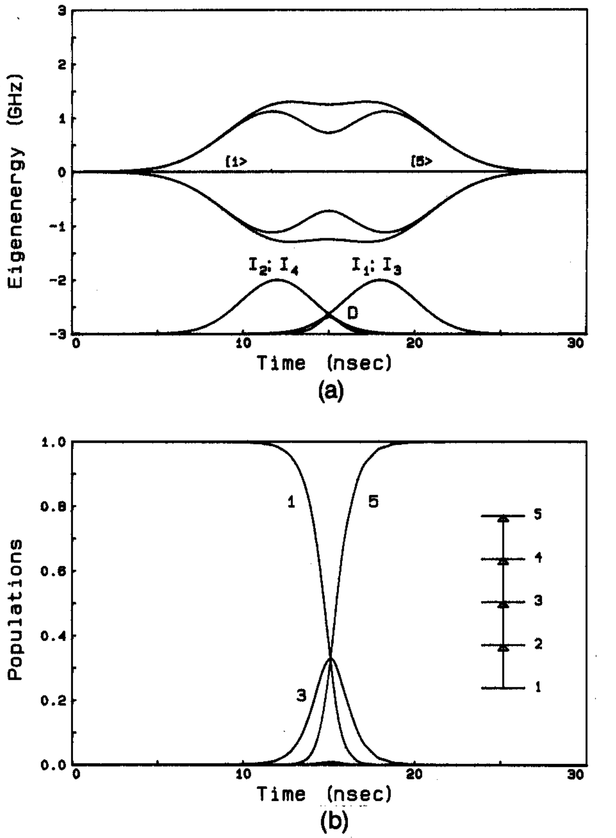

Fig. 7. (a) Adiabatic energy levels of a five-level system as a function of time for 5-ns FWHM asynchronous, resonant drive pulses of area 36π. The time delay is 6 ns, the even-numbered transitions ($I_{2,4}$) are driven first, followed by the odd transitions. D is the diabatic factor defined in the text. (b) Populations as a function of time for the same conditions as in (a).

of Fig. 5 can be generalized to scanning the lowest transition in the red-to-blue direction, with the last transition pumped blue of resonance and all intermediate transitions resonantly pumped. Clearly, there are many possibilities with swept-frequency light.

The delayed-pulse method can also be generalized. The Hamiltonian of interest has the tridiagonal form of Eq. (5), with detunings along the diagonal (with $\Delta_2 = 0$ owing to our selection of the zero of energy) and coupling terms off the diagonal. If all the Δ's are zero and if the number of coupled states is odd, one of the eigenenergies is zero and is associated with an eigenstate composed only of the odd levels $|1\rangle$, $|3\rangle$, ...[9] For example, for a five-level system the zero-energy eigenstate is

$$\Psi_0 = \frac{\Omega_2\Omega_4|1\rangle - \Omega_1\Omega_4|3\rangle + \Omega_1\Omega_3|5\rangle}{N}, \tag{7}$$

where N is the normalized factor. Similarly, for a seven-level system the zero-energy eigenstate is

$$\Psi_0 = \frac{\Omega_2\Omega_4\Omega_6|1\rangle - \Omega_1\Omega_4\Omega_6|3\rangle + \Omega_1\Omega_3\Omega_6|5\rangle - \Omega_1\Omega_3\Omega_5|7\rangle}{N}. \tag{8}$$

Note that $|1\rangle$ has as its coefficient the even-numbered driving terms, while the highest level, $|3\rangle$, $|5\rangle$, or $|7\rangle$, has as its coefficient the odd-numbered driving terms. Further, the intermediate-level coefficients progress from predominantly even to predominantly odd terms. Thus synchronous application of all the even driven pulses followed by synchronous application of all the odd drive pulses can adiabatically change this eigenstate from the lowest to the highest level. Of course, other sequences of temporally overlapped pulses are possible, but the preceding one is probably the most efficient.

Figure 7 provides a demonstration for a five-level system. Four Gaussian (5-ns FWHM) pulses are applied, with pulses 2 and 4 preceding pulses 1 and 3 by 6 ns. All the pulses have an area of 36π. Note that only the odd levels acquire a significant population during the transfer.

If the number of coupled levels is even, the algebra becomes difficult, and the analytic results have not been derived here. Instead a search has been made for adiabatic pathways by using numerical methods for a four-level system and for delayed-pulse pumping. One successful method uses a long-duration pulse or cw light to couple $|2\rangle$ and $|3\rangle$. This produces dressed states $|2\rangle \pm |3\rangle$ separated by a Rabi frequency. If the light waves coupling these dressed states to $|4\rangle$ and $|1\rangle$ are both resonant with either the upper or the lower dressed level, the system behaves like the three-level system, in that driving first the upper transition ($|3\rangle \to |4\rangle$) and then the lower transition ($|1\rangle \to |2\rangle$) results in a complete population transfer from $|1\rangle$ to $|4\rangle$ with a suppressed intermediate-level population. Increasing the coupling of the intermediate levels, $|2\rangle$ and $|3\rangle$, makes the system more like the three-level system. In practice, often the pulse durations must all be comparable. In this case similar results can be obtained if the $|2\rangle \to |3\rangle$ transition is pumped at an intermediate time between the other two pulses, as shown in Fig. 8. The first transition is pumped by light shifted to the blue (or red) by an amount approximately equal to half the maximum Rabi splitting of the $|2\rangle \pm |3\rangle$ dressed states, the second transition is pumped resonantly, and the third is pumped by red (or blue) shifted light; so the overall process is three-photon resonant. At early and late times, this splits the lowest and highest levels apart from the intermediate levels, as Fig. 8 shows. The pulses are applied in reverse order, so initially $|4\rangle$ is mixed with $|3\rangle$. Thus, at early time, the unperturbed second-highest energy state in Fig. 8 must be predominantly $|1\rangle$. At late time $|1\rangle$ is mixed with $|2\rangle$, so that the second-highest state must be $|4\rangle$. Thus $|1\rangle$ transfers adiabatically to $|4\rangle$. Because the optimum detunings of lasers 1 and 3 are dependent on the intensity of laser 2, this method is not so forgiving as the all-resonant pumping of systems with an odd number of levels, but it does achieve adiabatic transfer with fixed-frequency lasers.

The above scheme was tested by a numerical integration of Eq. (2) by using Gaussian (5-ns FWHM) pulses centered at times of 17, 15, and 13 ns, as shown in Fig. 8. Each pulse has an area of 18π. The detuning of 500 MHz is chosen to match approximately the energy splittings at $t = 15$ ns. As Fig. 8(b) shows, adiabatic transfer occurs. Note that the population of the two intermediate levels is considerably suppressed.

It is expected that, in a generalization to systems with an even number of levels greater than 4, the intermediate transitions should be pumped by resonant light with the

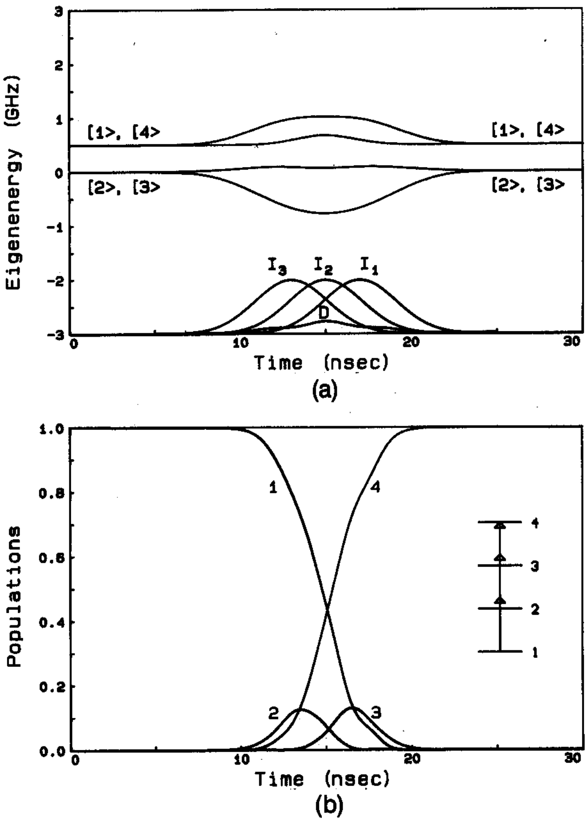

Fig. 8. (a) Adiabatic energy levels of a four-level system as a function of time for 5-ns FWHM asynchronous drive pulses of area 18π. The pulses are centered at 17, 15, 13 ns. The lowest transition is driven by 500-MHz blue-shifted light, the middle transition is driven by resonant light, and the upper transition is driven by 500-MHz red-shifted light. D is the diabatic term defined in the text. (b) Populations as a function of time for the same conditions as in (a).

time profile I_2 of Fig. 8, while the first and the last transitions should be pumped off-resonance with the time profiles I_1 and I_3, respectively, with overall resonance between the lowest and highest levels.

5. NONCASCADE SYSTEMS

In any application of adiabatic pumping the degeneracy of the states involved must be considered. If the degeneracy increases in each step, for example in a $J = 1 \rightarrow 2 \rightarrow 3$ sequence, the results given above are applicable. Each m_J sublevel connects to one in the next level, assuming parallel linear polarization of all the light. Other sequences of J values generally involve systems with more-complicated connections between levels, so that a sublevel may be coupled with more than one sublevel of another level. Figure 9 shows some examples. If the sequence of levels is $J = 0 \rightarrow 1 \rightarrow 1$, dipole selection rules require the use of crossed polarizations for copropagating pump beams, since $m_J = 0 \rightarrow m_J = 0$ is not allowed for $\Delta J = 0$. This corresponds to the Y-shaped configuration of Fig. 9(a) with an interaction Hamiltonian

$$H = \hbar \begin{bmatrix} \Delta_1 & \Omega_1/2 & 0 & 0 \\ \Omega_1/2 & 0 & \Omega_2/2 & \Omega_3/2 \\ 0 & \Omega_2/2 & \Delta_2 & 0 \\ 0 & \Omega_3/2 & 0 & \Delta_3 \end{bmatrix}. \quad (9)$$

The lowest and intermediate levels have $m_J = 0$, and the upper levels have $m_J = \pm 1$. For zero detunings and $\Omega_2 = \Omega_3$, applying the upper pulse (Ω_2 and Ω_3) ahead of the lower pulse partitions all the level $|1\rangle$ population equally between $|3\rangle$ and $|4\rangle$. More generally, if the fluences associated with Ω_2 and Ω_3 are not equal, the final populations of levels 3 and 4 are proportional to the fluences (fluence $\equiv \int I dt$) F_2 and F_3, respectively. The level 2 population is strongly suppressed as in the cascade three-level case.

If the Y shape is inverted as in Fig. 9(b), so that the lowest levels are $J = 1$ and the highest is $J = 0$, assuming that $|1\rangle$ and $|2\rangle$ are initially equally populated, the usual sequence of driving the upper and then the lower transition results in redistribution of the population such that half is in $|4\rangle$ and half is distributed between $|1\rangle$ and $|2\rangle$ in proportion to F_2 and F_1, respectively.

This example points out a limitation of adiabatic pumping. There seems to be no way to transfer population totally from one level to another of lower degeneracy. Of course, the initial level could be prepared by optical pumping so that only a single sublevel is populated or so that the initial state has total coherence among sublevels before adiabatic pumping.

A lower degeneracy level can be used as the intermediate state, however. For example, Fig. 9(c) shows the example of $J = 1$, $m_J = \pm 1 \rightarrow J = 0 \rightarrow J = 1$, $m_J = \pm 1$. Using time-delayed pumping and controlling the light polarizations so that only one populated lower level and one unpopulated upper level are coupled to the intermediate level on each pump pulse pair, one can sequentially transfer the population from the lower to the upper sublevels. Alternatively, the left legs of the X shape in Fig. 9(c) can be detuned to the red of the intermediate level, while the right legs are detuned to the blue. Then the usual sequence of driving first the upper and then the lower transitions produces complete inversion. Otherwise, if both legs are resonant with the intermediate state and this sequence is used, only half the population will be transferred to the upper pair of levels.

6. DOPPLER-BROADENED SYSTEMS

For many applications the sample will have a range of Doppler shifts and hence of detunings. This section examines the effect of Doppler broadening on the adiabatic pumping of a three-level system by time-delayed pulses. The two light pulses are tuned to the center of the Doppler profiles, and it is assumed that there are no collisions dur-

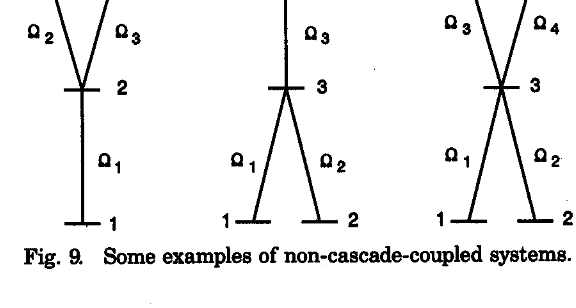

Fig. 9. Some examples of non-cascade-coupled systems.

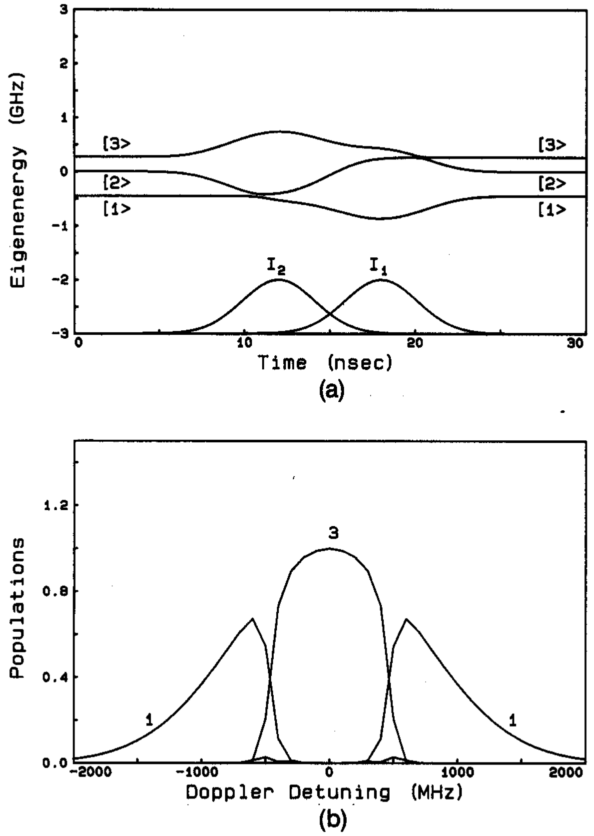

Fig. 10. (a) Adiabatic energy level of a three-level system as a function of time for 5-ns FWHM asynchronous, resonant drive pulses of area 18π. The time delay is 6 ns, the Doppler shift of the lower transition is -450 MHz, and the Doppler shift of the upper transition is -270 MHz. (b) Populations as a function of the Doppler shift of the lower transition. The Doppler profile for the lower transition has a FWHM of 1.66 GHz. The Doppler shift of the upper transition is 0.6 of the shift of the lower transition.

ing the pumping process. The two transitions are permitted to have different frequencies and thus different Doppler shifts. As an illustration of the nature of the atomic response with Doppler shifts, atoms moving with one particular longitudinal velocity are considered. The signs of the shifts for this group of atoms will be the same for copropagating pump waves and opposite for counterpropagating waves. The adiabatic energy levels with copropagating waves are shown in Fig. 10(a), where the Doppler shift for the lower transition is -0.45 GHz and that for the upper transition is -0.27 GHz. Both pulses have an area of 18π. As the adiabatic curves show, the transfer from $|1\rangle$ to $|3\rangle$ involves two curve crossings. The first is the crossing of $|1\rangle$ with a state that is a combination of $|2\rangle$ and $|3\rangle$. The second is the crossing of $|3\rangle$ with a combination of $|1\rangle$ and $|2\rangle$. Thus I_1 must be small at the first crossing to permit crossover, and I_2 must be small at the second crossing. The crossing points move outward with increasing Doppler shift and with increasing pump strength. Thus, for weak pumping, the range of velocities effectively pumped will be smaller than that for strong pumping. For sufficiently strong pumping, all the velocity groups should be effectively pumped. Figure 10(b) shows the populations in the three levels for each Doppler detuning after pumping by 18π pulses. All the atoms with shifts less than 500 MHz are effectively pumped, while those with larger shifts remain in the ground state. Few are left in $|2\rangle$.

Figure 11 shows the corresponding adiabatic energy levels for counterpropagating pump waves. In this case there is only one crossing and the two-photon detuning is smaller, so the pumping is effective to approximately 1 GHz for 18π pulses. Doubling the pulse area in either case approximately doubles the pumped width, so copropagating waves pump to 1 GHz with 36π pulses and counterpropagating pulses pump the entire Doppler profile.

7. DAMPING AND TIME SCALING

It is well known[1,13] that, in two-level systems, damping reduces the maximum inversion attainable. The damping can arise either from population transfer between levels or from dephasing collisions. The effect of dephasing on the three-level delayed-pulse process of Fig. 3 above is examined here under the restrictions that

$$d\rho_{ij}/dt = -\gamma_{ij}\rho_{ij} - (i/\hbar)[H,\rho]_{ij},$$
$$\gamma_{13} = \gamma_{12} + \gamma_{23},$$
$$\gamma_{12} = \gamma_{23}. \tag{10}$$

With these assumptions, it is found that the fraction of

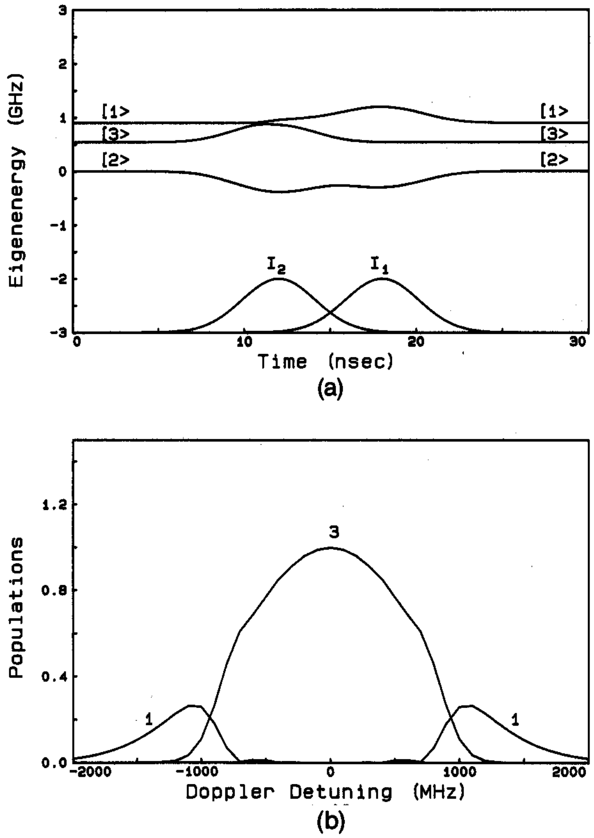

Fig. 11. (a) Adiabatic energy level of a three-level system as a function of time for 5-ns FWHM asynchronous, resonant drive pulses of area 18π. The time delay is 6 ns, the Doppler shift of the lower transition is -900 MHz, and the Doppler shift of the upper transition is $+480$ MHz. (b) Populations as a function of the Doppler shift of the lower transition. The Doppler profile for the lower transition has a FWHM of 1.66 GHz. The Doppler shift of the upper transition is 0.6 of the shift of the lower transition.

atoms pumped to the upper level is

$$\eta = 0.333 + 0.667 \exp(-4.3 \times 10^9 \gamma_{12}). \qquad (11)$$

This is a good approximation for the Doppler-broadened cases of Figs. 10 and 11 as well. The effect of damping resulting from population transfer is comparable, with the exception that a short lifetime of $|2\rangle$ does not degrade the process, because $|2\rangle$ is never significantly populated. Otherwise, the transfer must take place in a time that is short compared with the shortest damping time if high efficiency is to be obtained.

This raises the question of how the examples here can applied in situations in which the pulse duration τ is greater or less than the value of 5 ns that has been used in all the calculations presented here. For resonant, time-delayed pumping of the three-, five-, and seven-level systems, keeping the pulse areas constant while changing the time scale should maintain adiabatic behavior as long as damping is unimportant. For time-delay pumping of the four-level system the detuning must be scaled as $1/\tau$ because the Rabi frequencies scale as $1/\tau$ for a fixed pulse area. For swept-frequency pumping of the two-level system, inequality (4) indicates that keeping the pulse area constant while increasing the frequency sweep rate proportionally to $1/\tau$ will maintain the adiabatic condition. For the swept-frequency three-level systems, analytic results have not been derived here. Instead, the method described above can be used to evaluate these cases. Clearly, the sweep rates must be increased for shorter pulses, and the pulse areas probably must be increased as well.

Of course, the fluence for a given pulse area is proportional to $1/\tau$, so the intensity is proportional to $1/\tau^2$. Thus the kilowatt-per-square-centimeter intensities characteristic of nanosecond pulses become gigawatt-per-square-centimeter intensities for picosecond pulses. This is high enough that ac Stark shifts and competing multiphoton processes must be considered.

8. CONCLUSION

This paper has presented a simple way to identify and characterize methods of adiabatically inverting multilevel atomic or molecular systems. By numerically diagonalizing the interaction Hamiltonian, one can quickly trace the time development of adiabatic eigenstates. This also permits computation of the diabatic term defined in Eq. (3) above. It is found from numerical integration of the Bloch equations that, if this term is always less than 0.01, the system exhibits adiabatic behavior. In all the examples given here, pulse energies near the minimum required to achieve adiabatic behavior were used.

The use of time delays with fixed-frequency pump light in adiabatic inversion has been generalized to systems of any number of levels. The case of an odd number of coupled levels is particularly simple. Synchronously pumping the even-numbered transitions and then synchronously pumping the odd-numbered transitions can effectively invert the population. Also, it was shown how the time-delayed method can be applied to a four-level system.

Obviously, adiabatic pumping of multilevel systems could be accomplished by sequentially adiabatically pumping up the ladder of states. This would actually require smaller pulse areas than the single-step pumping described here. However, damping might make this unattractive owing to the longer time required. In addition, single-step pumping can be accomplished with fixed-frequency light for systems of three or more levels, and in this case some of the intermediate states are only slightly populated, so their radiative decay does not contribute significantly to the damping of the overall process.

Any modification of the driving waves by the sample was neglected. This is valid only for optically thin samples. The study of optically thick samples for two-level systems driven near resonance[1] has shown that the coupling of the radiation and the sample as described by the Maxwell–Bloch equations leads to a variety of interesting phenomena such as self-induced transparency and free-induction decay. Extensions of these ideas to multilevel systems are explored in several recent papers.[15]

For convenience, Gaussian time-profile pulses were used in all the present computations. Other shapes are also capable of adiabatic pumping as long as the pump pulses are smooth, with gradual turn-on and turn-off.

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