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Adiabatic following in multilevel systems

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The problem of achieving population inversion adiabatically in an N-level system using one or more laser fields whose detunings and/or amplitudes are continuously varied is studied analytically and numerically. The SU(N) coherence vector picture is shown to suggest unexpected inversion procedures and also to give a generalized interpretation of adiabatic following. It is shown that the (N^2-1)-dimensional SU(N) space contains an $(N-1)$-dimensional steady-state subspace $\underline{\Gamma}(t)$ whose orthonormal basis vectors $\vec{\Gamma}_1, \ldots, \vec{\Gamma}_{N-1}$ are given explicitly in terms of the Hamiltonian matrix elements. The motion of the system can be interpreted as a "generalized precession" of $\vec{S}$ about $\underline{\Gamma}$. Multilevel adiabatic following occurs when the angle $\chi(t)$ between the coherence vector $\vec{S}$ and its projection onto $\underline{\Gamma}$ is very small. The multiple dimension of $\underline{\Gamma}$ is shown to provide a variety of paths for adiabatic inversion. The adiabatic solution is obtained by solving $N-1$ simple equations for the directional cosines of $\vec{S}$ on $\vec{\Gamma}_i$. The adiabatic solution and time scale and the state taken up by the atomic variable are discussed analytically and numerically for a three-level system.

I. INTRODUCTION

Multiphoton excitation of atomic and molecular systems with more than two levels is of considerable importance in many problems. In particular, three-level systems have been central to discussions[1–4] of two-photon coherence, Raman beats, three-level superradiance and echoes, off-resonance transient response, and coherent multistep photoionization and photodissociation. Some attention was given to special cases of three-level adiabatic rapid passage[5] and recently to coherent multilevel adiabatic excitation,[6] which is also the subject of the present work.

An idealized formulation of the N-level excitation process focuses on the problem of accomplishing complete inversion in an arbitrarily spaced N-level system that is chain-wise dipole connected by laser fields (Fig. 1), by means of a continuous sweep of the laser field frequencies and/or envelopes. The term adiabatic[7,8] means that the rate of change of the varying components is sufficiently small so that a quasi-steady-state is maintained all along the process. Of course this rate must be reasonably large compared to the natural decay rates of the system if the process is to be practical.

It is clear that the evolution of a coherently excited system in a pure state can be described by the Schrödinger equation with no need for the more complex density-matrix formalism. However, it was the density-matrix SU(2) coherence-vector theory which led to the complete description and analytic solution of the adiabatic process in two levels, termed adiabatic following.[8] Moreover, this solution is applicable also to systems in general mixed states, and lends itself naturally to situations involving relaxation.

The recent development of the SU(N) coherence-vector theory[2,3] of N-level systems raised the immediate question: Can this new picture provide a generalization of the two-level adiabatic following picture which will serve as a guide to the description of adiabatic processes and to analytic solutions in N-level systems?

We will show in this work that the answer is affirmative, and we also obtain a variety of unexpected results. The (N^2-1)-dimensional SU(N) space is shown to contain an $(N-1)$-dimensional steady-state subspace $\underline{\Gamma}(t)$ given explicitly in terms of the field detunings and envelopes. As $\underline{\Gamma}$ develops adiabatically, the SU(N) coherence vector is shown to follow it, and the conditions for adiabatic evolution and, in particular, adiabatic inversion are easily obtained. The problem is reduced to solving $N-1$ simple real equations for the directional cosines of the coherence vector referred to the basis vectors of $\underline{\Gamma}$.

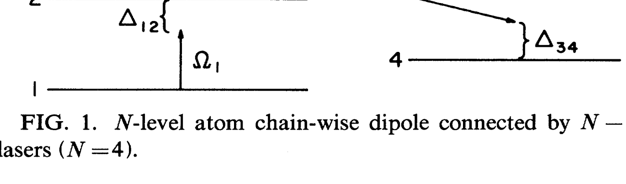

FIG. 1. N-level atom chain-wise dipole connected by $N-1$ lasers ($N=4$).

For a three-level system these equations are easily solved and we give an exact analytic solution for any shape and strength of the field envelopes and any time dependence of the detunings, all obeying the adiabatic conditions.

Other questions which we will answer include the following.

(i) What is the adiabatic time scale and what influences it? Since the adiabatic process sweeps through regions where no analytic guide was available and since the adiabatic rate must compete with the decay rates of the system, the answer to the question of time scale is of considerable theoretical and practical interest.

(ii) What are the "natural" intermediate states which the atomic variables would assume as the adiabatic inversion is performed? One of our unexpected findings is that even though the atomic levels are assumed to be only chain-wise dipole connected, the states involving only one-photon coherences are *not* necessarily the states taken up by the atomic variables (for $N>2$) as the adiabatic process evolves. We will give an explanation of this process.

II. THEORY

We begin with the equation of motion for the SU(N) coherence vector[2] $\vec{S}=(S_1,S_2,\ldots,S_{N^2-1})$. It is simply a generalized rotation in $N-1$ dimensions,

$$\dot{S}_i=\sum_{j,k} f_{ijk}\Gamma_j S_k \equiv \Gamma \otimes S \ . \tag{1}$$

Equation (1) is an alternative form of the Liouville equation for the density matrix $\hat{\rho}$ and Hamiltonian $\hat{H}$,

$$i\hbar\frac{\partial\hat{\rho}}{\partial t}=[\hat{H},\hat{\rho}] \ . \tag{2}$$

For an N-level system of unperturbed energies $E_i, i=1,2,\ldots,N$, irradiated by a laser field

$$\vec{E}(z,t)=\sum_{j=1}^{N-1}\vec{\epsilon}_{j,j+1}(t)\exp[-i(\nu_{j,j+1}t-K_{j,j+1}z)]+\text{c.c.} \ , \tag{3}$$

we have in the rotating-wave approximation

$$\vec{S}=(u_{12},u_{23},\ldots,u_{13},u_{24},\ldots,v_{12},v_{23},\ldots,v_{13},v_{24},\ldots,w_1,w_2,\ldots,w_{N-1}) \ , \tag{4}$$

$$\vec{\Gamma}=(-\Omega_1,-\Omega_2,\ldots,0,0,\ldots,0,0,\ldots,\Delta_1,\Delta_2,\ldots,\Delta_{N-1}) \ , \tag{5}$$

where

$$u_{jk}=\rho_{jk}+\rho_{kj} \ , \quad v_{jk}=-i(\rho_{jk}-\rho_{kj}) \ , \quad 1\le j<k\le N \tag{6}$$

$$w_l=[2/l(l+1)]^{1/2}(w_{12}+2w_{23}+\cdots+lw_{l,l+1}) \ , \quad w_{jk}=\rho_{jj}-\rho_{kk} \tag{7}$$

and where $\Omega_j(t)$ is the Rabi frequency defined as usual in terms of the atomic dipole moments $\vec{d}_{j,j+1}$ and field envelope $\vec{\epsilon}_{j,j+1}(t)$ by

$$\Omega_j(t)=2\vec{d}_{j,j+1}\cdot\vec{\epsilon}_{j,j+1}(t)/\hbar \tag{8}$$

and where $\Delta_l(t)$ is defined in terms of the detunings

$$\Delta_{jk}(t)=\nu_{jk}(t)-(E_j-E_k)/\hbar \ ,$$

by (9)

$$\Delta_l=[2/l(l+1)]^{1/2}(\Delta_{12}+2\Delta_{23}+\cdots+l\Delta_{l,l+1}) \ .$$

The antisymmetry of the SU(N) structure constants f_{ijk} in Eq. (1) guarantees that the length of $\vec{S}(t)$ is constant. Equation (1) contains, as its simplest case when $N=2$, the optical Bloch equation expressed as a single vector precession equation[8]

$$\frac{d\vec{S}}{dt}=\vec{\Gamma}\times\vec{S} \ . \tag{10}$$

Equation (10) has the interpretation that the coherence vector $\vec{S}$ is precessing about the torque vector $\vec{\Gamma}$ at a precession frequency $\Gamma=|\vec{\Gamma}|$. Under the condition that the torque vector $\vec{\Gamma}$, initially nearly parallel to $\vec{S}$, changes its direction slowly enough, the quasi-steady-state solution

$$\vec{S}=\frac{S}{\Gamma}\vec{\Gamma} \tag{11}$$

is maintained and the coherence vector is said to be adiabatically following $\vec{\Gamma}(t)$. In the following sections we extend this picture to an N-level system.

III. THE STEADY-STATE SUBSPACE OF AN N-LEVEL SYSTEM

The striking property of N-level systems with $N>2$ is that the steady-state subspace $\underline{\Gamma}$ of Eq. (1) contains $N-2$ solutions in addition to $\vec{\Gamma}$ (see Appendix). The basis vectors $\vec{\Gamma}_1,\vec{\Gamma}_2,\ldots,\vec{\Gamma}_{N-1}$ of $\underline{\Gamma}$ can be given explicitly in terms of $\Omega_i(t)$ and $\Delta_{ij}(t)$ and are chosen to be orthonormal with $\vec{\Gamma}_1=\vec{\Gamma}/\Gamma$. General properties of $\underline{\Gamma}$ are easily obtained from the equation of motion (1) by the definition,

$$\vec{\Gamma}\otimes\vec{\Gamma}_i=0 \ , \tag{12}$$

orthonormalization,

$$\vec{\Gamma}_i\cdot\vec{\Gamma}_j=\delta_{ij} \ , \tag{13}$$

and precession,

$$\dot{\vec{S}}\cdot\vec{\Gamma}_i=0 \ . \tag{14}$$

Further, the directional cosines of $\vec{S}$ on Γ_i

$$D_i=\vec{S}\cdot\vec{\Gamma}_i/S \tag{15}$$

become constants of motion if $\dot{\vec{\Gamma}}_i=0$. This can be interpreted as a generalized precession of $\vec{S}$ about $\underline{\Gamma}$. The relative position of $\vec{S}$ with respect to $\underline{\Gamma}$ can be measured by the angle χ between $\vec{S}$ and its projection $\vec{S}_p$ on to $\underline{\Gamma}$. Both $\vec{S}_p$ and χ are given in terms of the directional cosines $D_i = \cos\theta_i$ as follows:

$$\vec{S}_p = S \sum_i D_i \vec{\Gamma}_i \;, \tag{16}$$

$$\cos\chi = \left[\sum_i D_i^2 \right]^{1/2} . \tag{17}$$

The evolution of the system is determined by the angle χ. Adiabatic motion is characterized by small-angle "precession" $\chi(t) \sim 0$, but for completeness we will discuss later also a nonadiabatic case with large-angle precession.

IV. N-LEVEL ADIABATIC FOLLOWING

Clearly, as we sweep the detunings Δ_{ij} and Rabi frequencies Ω_i, the whole steady-state subspace moves with time. Our generalization of adiabatic following is that under the adiabatic condition (to be specified later), the coherence vector $\vec{S}(t)$ follows $\underline{\Gamma}(t)$ with $\chi(t) \sim 0$. The angles θ_i between $\vec{S}$ and $\vec{\Gamma}_i$ may, on the other hand, change with time so that $\vec{S}$ might move within $\underline{\Gamma}$ during the adiabatic process. It is the detail of this motion which determines completely the adiabatic solution, since $\chi(t)=0$ implies

$$\vec{S}(t) = \vec{S}_p(t) = s \sum_i D_i(t) \vec{\Gamma}_i(t) \;, \tag{18}$$

where the $\vec{\Gamma}_i(t)$ are known.

In principle the directional cosines of the adiabatic solution can be determined by the N conservation laws[2] $\mathrm{Tr}(\rho^n) = \mathrm{const}$ of the Bloch equation. The problem is then reduced to solving $N-1$ coupled polynomials in the $N-1$ variables. Alternately, one can introduce $N-1$ simple differential equations for the adiabatic D_i where most of the properties are apparent from the simple form of the coefficients. These differential equations for the directional cosines of the adiabatic following process are easily obtained from Eqs. (1) and (18), giving the $N-1$ simple real equations

$$\dot{D}_i = \sum_{j=1}^{N-1} g_{ij} D_j \;, \quad i = 1, 2, \ldots, N-1 \tag{19}$$

with

$$\sum_{i=1}^{N-1} D_i^2 = 1 \;, \quad g_{ij} = \dot{\vec{\Gamma}}_i \cdot \vec{\Gamma}_j \;. \tag{20}$$

Detailed solutions of these equations for $N=3$ are presented and discussed in Secs. V and VI.

The adiabatic conditions under which $\vec{S}$ will indeed follow $\underline{\Gamma}$ are twofold.

(1) The initial detunings $\Delta_{ij}(0)$ and Rabi frequencies $\Omega_j(0)$ must be chosen so that the initial-state vector $\vec{S}(0)$ is in steady state, namely,

$$\chi(0) = 0 \;. \tag{21}$$

(2) The term "sufficiently slow" for the sweeping rate is determined by

$$|\dot{\vec{\Gamma}}_i| \ll \Omega_0 \;, \tag{22}$$

that is, the rate of change of direction of all the $\underline{\Gamma}$ basis vectors must be small compared to the precession rate Ω_0. For two levels the precession rate is well known to be $\Omega_0 = \Gamma$. In general, for $N>2$, Ω_0 is not known, but, as will be shown in the next section, it can be bounded so that Eq. (22) becomes a practical criterion for the adiabatic rate of the $N-1$ level system.

Adiabatic following is most useful if one wishes to lead the atomic state steadily from some initial state $\vec{S}(0)$ to a definite final state $\vec{S}(T)$. Of course, not all pairs of states can be connected. The initial and final states must obey all the conservation laws compatible with the equation of motion.[2] The simplest example is adiabatic inversion, where the initial ground state is led adiabatically to the state where all population is in level N. The general procedure to follow is first determine initial and final values for the detunings Δ_{ij} and the Rabi frequencies Ω_i, so that

$$\chi(T) = \chi(0) = 0 \;, \tag{23}$$

and then sweep adiabatically from the initial to the final values.

The striking result of this scheme for $N>2$ is the multiplicity of possible paths for adiabatic following. Because of the multiple dimension of $\underline{\Gamma}$, the adiabatic condition $\chi(0) = \chi(T) = 0$ for any given atomic states $\vec{S}(0)$ and $\vec{S}(T)$ can be satisfied by many choices of Ω_i and Δ_{ij}. This point will be discussed in detail in connection with three-level adiabatic inversion in Sec. VI.

V. ADIABATIC FOLLOWING IN A THREE-LEVEL SYSTEM

As an example, we will now discuss the adiabatic following process and its solutions for a three-level system.

The steady-state subspace $\underline{\Gamma}$ is two dimensional with the orthonormal basis given by (see Appendix)

$$\vec{\Gamma}_1 = \vec{\Gamma}/\Gamma \;, \tag{24}$$

$$\vec{\Gamma}_2 = (\vec{\Gamma}' - \alpha \vec{\Gamma}_1)/(\Gamma'^2 - \alpha^2)^{1/2} \;, \tag{25}$$

with $\alpha = (\vec{\Gamma}' \cdot \vec{\Gamma}_1)$ and

$$\vec{\Gamma} = \begin{bmatrix} -\Omega_1 \\ -\Omega_2 \\ 0 \\ \Delta_{12} \\ (\Delta_{12} + 2\Delta_{23})/\sqrt{3} \end{bmatrix} , \quad \vec{\Gamma}' = \begin{bmatrix} -\Omega_1(\Omega_1^2 + \Omega_2^2 - 4\Delta_{23}\Delta_{13}) \\ -\Omega_2(\Omega_1^2 + \Omega_2^2 - 4\Delta_{12}\Delta_{13}) \\ \Omega_1\Omega_2(\Delta_{12} - \Delta_{23}) \\ \Omega_1^2\Delta_{12} + \Omega_2^2\Delta_{23} - 4\Delta_{12}\Delta_{23}\Delta_{13} \\ \sqrt{3}(\Omega_1^2\Delta_{12} + \Omega_2^2\Delta_{23} - 4\Delta_{12}\Delta_{23}\Delta_{13}) \end{bmatrix} . \tag{26}$$

We notice two distinct points. First, three components were omitted. These are the v_{ij} coherences which vanish in the steady-state solutions. The second point is the appearance of the two-photon coherence u_{13} in Γ_2. This will lead to the evolution of u_{13} during the adiabatic process.

The three-level adiabatic solution is

$$\vec{S}(t)=S[D_1(t)\vec{\Gamma}_1(t)+D_2(t)\vec{\Gamma}_2(t)] , \quad (27)$$

where the directional cosines $D_i(t)$ obey the simple equations

$$\dot{D}_1=gD_2 , \quad (28)$$

$$\dot{D}_2=-gD_1 , \quad (29)$$

with

$$D_1^2+D_2^2=1 , \quad (30)$$

and

$$g=\dot{\vec{\Gamma}}_1\cdot\vec{\Gamma}_2 , \quad (31)$$

giving

$$D_1=\cos\theta , \quad D_2=\sin\theta , \quad (32)$$

$$\theta(t)=\theta_0-\int_0^t g\,dt . \quad (33)$$

We note the following interesting relations:

$$\int_0^t g\,dt=\int_0^t \vec{\Gamma}_2\cdot\dot{\vec{\Gamma}}_1 dt$$
$$=-\int_0^t \vec{\Gamma}_1\cdot\dot{\vec{\Gamma}}_2 dt , \quad (34)$$

and it can be shown that $\int \vec{\Gamma}_2\cdot d\vec{\Gamma}_1$ or $\int \vec{\Gamma}_1\cdot d\vec{\Gamma}_2$ is independent of the path and hence

$$\oint \vec{\Gamma}_2\cdot d\vec{\Gamma}_1=0 . \quad (35)$$

Equations (33) and (35) imply that $\theta(t)$, and therefore the adiabatic solution $\vec{S}(t)$ of Eq. (26), is determined by the values of Δ_{ij} and Ω_i only at t and t_0, independent of their values in between. We also notice that the g function is invariant under the exchange of the level indices 1 and 3 so that, for inversion, Eq. (35) leads to

$$\int_{\vec{\Gamma}_1(0)}^{\vec{\Gamma}_1(T)} \vec{\Gamma}_2\cdot d\vec{\Gamma}_1=0 . \quad (36)$$

We find especially interesting the case of equal detunings and Rabi frequencies, namely,

$$\Delta_{12}(t)=\Delta_{23}(t) ,$$
$$\Omega_1(t)=\Omega_2(t) . \quad (37)$$

It is easy to see that, in this case, $g(t)=0$ and the directional cosines $D_i(t)$ become constants of motion. The $\vec{S}$ vector then depends on time only through $\vec{\Gamma}_1$ and $\vec{\Gamma}_2$.

VI. THREE-LEVEL ADIABATIC INVERSION BY SWEEPING THROUGH A TWO-PHOTON RESONANCE POINT

Once we specify the initial and final atomic state, we can use the adiabatic condition (23) to trace the various possible paths for adiabatic passage between them. The initial state (level 1) and the final state (level 3) are represented by

$$\vec{S}(0)=[0,0,0,1,1/\sqrt{3}] ,$$
$$\vec{S}(T)=[0,0,0,0,-2/\sqrt{3}] . \quad (38)$$

We have again omitted the three v components, which all vanish.

The adiabatic condition (23) implies that both states must be expressed as a linear combination of $\vec{\Gamma}_1$ and $\vec{\Gamma}_2$, or $\vec{\Gamma}$ and $\vec{\Gamma}'$ of Eqs. (24) and (25). It is easily seen that the first three components are satisfied by imposing $\Omega_i(0)=\Omega_i(T)=0$, namely pulse-shaped Rabi frequencies. The last two components are then compatible with any detunings except that we must have both

$$|\Delta_{12}(0)|\neq 0 \quad \text{and} \quad |\Delta_{23}(T)|\neq 0 . \quad (39)$$

These conditions (39) should be considered carefully because they are somewhat unexpected, even anti-intuitive. The natural way to excite the atom is to bring it first to level 2 by resonant excitation of the 1-2 transition and then proceed to level 3 by sweeping into 2-3 resonance. We see, however, that (39) implies something close to the opposite order of resonances. In fact the natural excitation sequence, starting on 1-2 resonance with the first laser, removes the ground state out of the steady-state subspace, which results in large-angle precession. Even after avoiding this possibility by ensuring that (39) is satisfied, we are still left with a lot of freedom for our initial and final detunings.

If we restrict ourselves to processes where

$$D_i(T)=D_i(0) , \quad (40)$$

we obtain the following condition on the detunings

$$\Delta_{12}(T)=-\Delta_{23}(0)=\Delta_{32}(0) ,$$
$$\Delta_{23}(T)=-\Delta_{12}(0)=\Delta_{21}(0) . \quad (41)$$

We notice that the final detunings can be obtained from the initial values by exchanging the indices 1 and 3 consistent with Eq. (36).

A specific scheme satisfying $\Omega_i(0)=\Omega_i(T)=0$ in Eq. (41) is

$$\Omega_i(t)=A_i\sin^2\left[\frac{\pi\lambda}{a+b}t\right] , \quad (42)$$

$$\Delta_{12}(t)=a-\lambda t ,$$
$$\Delta_{23}(t)=b-\lambda t , \quad (43)$$

where the initial detunings a and b are arbitrary except that $a\neq 0$. Clearly at $t=T=(a+b)/\lambda$, the desired final parameters are reached and, if λ is sufficiently small, complete inversion will be achieved. The adiabatic restriction on the sweeping rate λ will be discussed later.

We notice three interesting points regarding the procedure sketched above.

(1) This proposed scheme can be achieved by a single laser interacting with the two transitions.

(2) At $t=T/2$, the Rabi frequencies reach their peak and the detunings are at two-photon resonance:

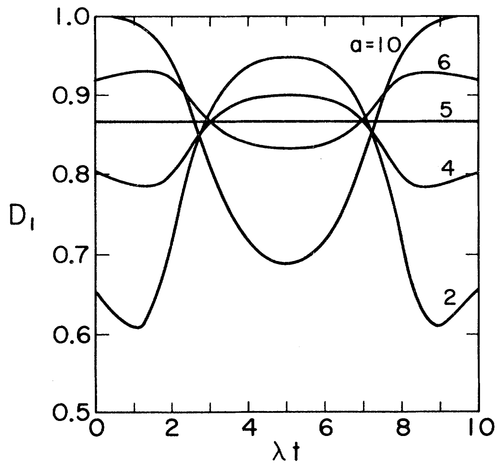

FIG. 2. Quasienergy D_1 vs λt for various paths (initial conditions a,b). For all paths we chose $a+b=10$ and $A_i=6$.

$$\dot{\Omega}_i(T/2)=0 , \tag{44}$$

$$\Delta_{13}(T/2)=\Delta_{12}(T/2)+\Delta_{23}(T/2)=0 . \tag{45}$$

(3) From Eqs. (44), (45), and (31) we obtain $g(T/2)=0$, so that at $t=T/2$, the D_i have an extremum. Particularly for $a>b$, $D_1(T/2)$ has a minimum point and for $b>a$, it has a maximum point. The opposite is true for $D_2(T/2)$.

It should be pointed out that $a>b$ implies that the order of resonances (which correspond to the Landau-Zener level crossings) is $\Delta_{23}=0$, $\Delta_{13}=0$, $\Delta_{12}=0$, and the only resonance which involves population is the two-photon resonance, leading to a smooth and steady population inversion as described later on. For $a<b$ the order of resonances is reversed and we obtain the natural excitation order mentioned before. It can be shown that as a is decreased in favor of b and we are getting closer to the natural scheme, the motion of the vector $\vec{S}$ becomes more sensitive to changes of Δ_{ij} and Ω_i and a slower sweeping rate is needed to maintain the adiabatic process. When a approaches zero, the adiabatic process collapses completely and 1-2 oscillations followed by 2-3 oscillations occur. This emphasizes our somewhat unexpected conclusion that the "antinatural" case $a>b$ is more favorable for adiabatic inversion.

Our solution for $D_1(t)$ for various initial conditions a,b are shown in Fig. 2. These solutions are indistinguishable from our numerical solutions of the full Bloch equation (1). We notice at $t=T/2$ the points of minimum and maximum for $a>b$ and $a<b$, respectively. The border straight line for $a=b$ is the case of equal detunings [Eq. (37)], where D_i become constants of motion. We also notice that $D_i(T)=D_i(0)$. In addition, for sufficiently slow rates λ, we found that $D_i(\tau=\lambda t)$ is independent of λ, consistent with Eq. (34).

It is easily seen that D_1 is related to the quasienergy of the system

$$D_1=\frac{\vec{S}\cdot\vec{\Gamma}}{S\Gamma}=\frac{1}{2S\Gamma}\langle\psi|H|\psi\rangle . \tag{46}$$

Clearly D_i measures the relative contribution of $\vec{\Gamma}_i$ to the atomic state $\vec{S}$ [Eq. (27)]. We expect that, for $a>b$, as D_2 approaches its maximum value, the contribution of $\vec{\Gamma}_2$ is increased and we will see the development of the atomic two-photon coherence u_{13}. This is shown in Fig. 3. We notice the smooth and steady flow of the atomic coherence typical of a very small-angle precession. This characterizes the evolution of all variables, coherences, populations, and directional cosines. The smooth and steady flow of populations towards complete inversion is shown in Fig. 4. Since this case is for a relatively strong field, for which $\Omega_i(T/2)\sim\Delta_{12}(T/2)$, we also see a power broadening effect as $\rho_{22}(t)$ rises and falls during the inversion process. For a weak field, for which $\Omega_i(T/2)<\Delta_{12}(T/2)$, we obtain a similar behavior except that $\rho_{22}(t)$ remains zero all along the process, with $\rho_{11}(T/2)=\rho_{33}(T/2)=\frac{1}{2}$.

The striking difference between the weak- and strong-field adiabatic inversions is the adiabatic rate. For our strong-field case, the adiabatic rate is by 2 orders of magnitude greater than the adiabatic rate of the weak-field case. This is shown in Figs. 5 and 6. Here we see the collapse of the adiabatic process with increasing rates λ. As λ becomes small, the lines converge to the adiabatic solution, which is identical to our analytical results. We find that the weak-field adiabatic rate is $\lambda\sim0.01\lambda_0^2$, whereas

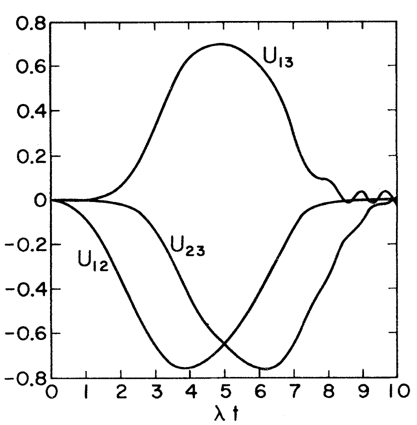

FIG. 3. Evolution of the coherences u_{12}, u_{23}, and u_{13} during the adiabatic process. ($A_i=8, a=10, b=0$.)

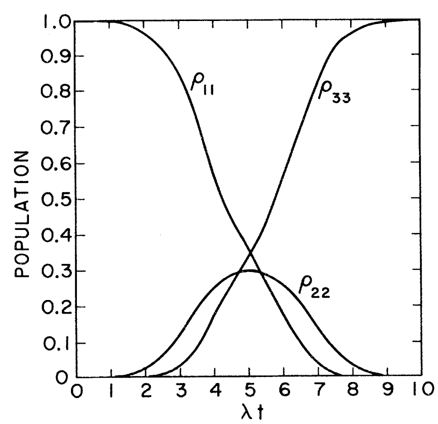

FIG. 4. Adiabatic population inversion in a strong-field laser ($A_i=8, a=10, b=0$).

the strong-field adiabatic rate is $\lambda \sim 1\Delta_0^2$. In all our calculations we use the frequency unit $\Delta_0 = [\Delta_{12}(0) + \Delta_{23}(0)]/10$. These rates can be predicted by our adiabatic condition $|\dot{\vec{\Gamma}}_i| \ll \Omega_p$ [Eq. (22)].

Although the small-angle precession rate Ω_p is not known, it can be bounded. We notice in Fig. 5 that the bottleneck of the adiabatic process is the point of two-photon resonance at $t = T/2$. The natural frequencies of a three-level system at two-photon resonance are well known (Brewer and Hahn of Ref. 9) and we take the smallest natural frequency

$$\nu_{\min} = [(\Omega_1^2 + \Omega_2^2 + \Delta_{12}^2)^{1/2} - \Delta_{12}]/2 \tag{47}$$

as a lower bound for Ω_p.

In the limits of weak ($\Omega_i \ll \Delta_{12}$) and strong ($\Omega_i > \Delta_{12}$) fields, we obtain immediately

$$\Omega_0 \sim \begin{cases} \Omega_1^2/2\Delta_{12} & \text{for } \Omega_i \ll |\Delta_{12}| \\ \Omega_1 & \text{for } \Omega_i > |\Delta_{12}| \ . \end{cases} \tag{48}$$

Namely, the precession rate Ω_0 is bounded by the two-photon Rabi frequency, $\Omega_e = \Omega_1^2/2\Delta_{12}$, for the weak field and by the single-photon Rabi frequency Ω_i for the strong field. The left-hand side of the adiabatic condition is easily calculated at $t = T/2$ and we obtain

$$|\dot{\Gamma}_1| < |\dot{\vec{\Gamma}}_2| \sim \begin{cases} |\dot{\Delta}_{12}|/\Omega_e & \text{for } \Omega_i \ll |\Delta_{12}| \\ \dot{\Delta}_{12}|/\Omega_1 & \text{for } \Omega_i > |\Delta_{12}| \ . \end{cases} \tag{49}$$

The adiabatic condition therefore becomes

$$|\dot{\Delta}_{12}| \ll \begin{cases} \Omega_e^2 & \text{for } \Omega_i \ll |\Delta_{12}| \\ \Omega_1^2 & \text{for } \Omega_i > |\Delta_{12}| \end{cases} \tag{50}$$

consistent with our numerical results.

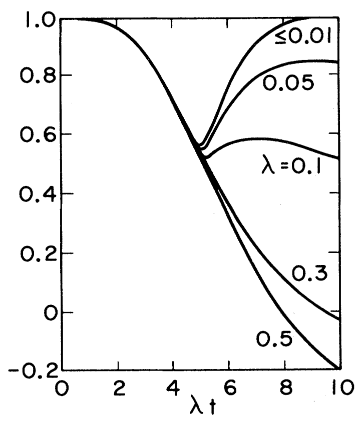

FIG. 5. Collapse of the adiabatic process for a weak-field case ($A_i = 2, a = 10$), D_1 vs λt.

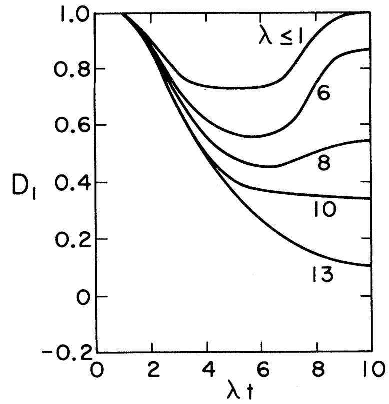

FIG. 6. Collapse of the adiabatic process for a strong-field case ($A_i = 8, a = 10$), D_1 vs λt.

VII. THREE-LEVEL ADIABATIC INVERSION BY DELAYED PULSES

The paths for adiabatic inversion discussed so far do not exhaust all the possibilities. A completely different adiabatic inversion scheme is obtained by imposing two-photon resonance at all times, $\Delta_{13}(t) = 0$. Under this condition the adiabatic restrictions (23) and (40) lead to the following conditions:

$$|\Omega_i| \ll |\Delta_{12}| \ , \tag{51}$$

$$2\Omega_1\Omega_2 \ll |\Omega_1^2 - \Omega_2^2| \ , \quad \text{at } t = 0 \text{ and } T \tag{52}$$

$$|\Omega_1^2(T) - \Omega_2^2(T)| = -|\Omega_1^2(0) - \Omega_2^2(0)| \ . \tag{53}$$

The first inequality is immediately recognized as the well-known condition[9] for elimination of level 2 and reduction to an effective two-level atom (levels 1 and 3), with the effective Rabi frequency

$$\Omega_e = \frac{\Omega_1\Omega_2}{2\Delta_{12}} \tag{54}$$

and the effective detuning

$$\Delta_e = \frac{\Omega_1^2 - \Omega_2^2}{4\Delta_{12}} \ . \tag{55}$$

Conditions (52) and (53) can now be written in terms of these effective parameters as

$$|\Omega_e| \ll |\Delta_e| \ , \quad \text{at } t = 0 \text{ and } T \tag{56}$$

$$\Delta_e(T) = -\Delta_e(0) \ . \tag{57}$$

These conditions are the well-known conditions for two-level adiabatic inversion and could be predicted by the SU(2) vector theory applied to the "effective" two-level system. If we wish to work out a practical version of this scheme, we must sweep the Rabi frequencies in opposite directions, for example,

$$\Omega_1(t) = \lambda t \ , \quad \Omega_2(t) = a - \lambda t \ , \tag{58}$$

as shown by the solid lines in Fig. 7. The effective detun-

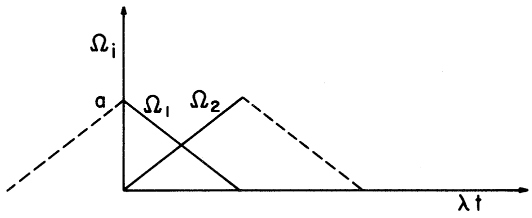

FIG. 7. Sweeping scheme of the effective detuning $\Delta_e=(\Omega_1^2-\Omega_2^2)/4\Delta_{12}$. $\Omega_1=\lambda t$, $\Omega_2=a-\lambda t$ (solid lines). The extension of the scheme to two delayed pulses is drawn by the dashed lines.

ing will reverse its sign at $T=a^2/\lambda$ and inversion is then reached.

This scheme can be extended to two delayed pulses as shown by the dotted lines in Fig. 7, giving a smooth adiabatic inversion with $\chi(t)=0$ all along the process. We mention this example to demonstrate the fact that adiabatic inversion can be obtained simply by designing a proper delay between the pulses. It is interesting to note that without a delay the process becomes completely nonadiabatic. In particular, if $\Omega_1=\Omega_2$, under the same conditions $[\Delta_{13}(t)=0,\Omega_i\ll\Delta_{12}]$, the effective detuning vanishes and we obtain on-resonance effective two-level Rabi oscillations. In the SU(3) space we found that the motion of the system is then a large-angle precession with $\chi(t)=60°$. This is consistent with the 120° inversion angle characteristic of three levels.[2]

VIII. SUMMARY

In this work we solved the problem of N-level adiabatic following. It was shown that the (N^2-1)-dimensional SU(N) space contains an $(N-1)$-dimensional steady-state subspace $\underline{\Gamma}$ which governs the generalized precessional motion of the system. Our generalization of adiabatic following is that the coherence vector $\vec{S}$ follows the whole steady-state subspace $\underline{\Gamma}$. The adiabatic solution was shown to be determined completely by the motion of $\vec{S}$ within $\underline{\Gamma}$, namely by the time dependence of the directional cosines D_i of $\vec{S}$ on $\underline{\Gamma}$. $N-1$ simple equations for D_i were presented and complete solutions were obtained for three-level systems with any time shape of the detunings and Rabi frequency obeying the adiabatic following conditions. The adiabatic rate conditions were formulated in the geometrical terms of the adiabatic following picture.

The multiple dimension of $\underline{\Gamma}$ (for $N>2$) was shown to allow a lot of freedom for the detunings, leading to a variety of possible paths for the adiabatic process and, in particular, for N-level adiabatic inversion. Two specific schemes for three-level adiabatic inversion were discussed. The first one sweeps the detunings Δ_{12} and Δ_{23} linearly from their arbitrary initial values a and b, and can be achieved by a single laser. It was found that for $a>b$ where the three resonances (two one-photon resonances and one two-photon resonance) occur in a nonintuitive order, the quasienergy of the system D_i reaches a minimum at the point of two-photon resonance, and the adiabatic rate can be made faster than in the case of the intuitive resonance order ($a<b$). It was shown that the fastest adiabatic rate is achieved for $a>b$ (nonintuitive detuning sweep) in the strong-field limit when the peaks of the Rabi frequencies are comparable to the intermediate detuning Δ_{12} at the point of two-photon resonance. We also noticed and explained the evolution of the two-photon coherence u_{13} during the adiabatic process, even though the levels are only chain-wise connected.

The second inversion scheme was shown to consist of a properly arranged delay between the two laser pulses with $\Delta_{13}(t)=0$ and $\Delta_{12}(t)\gg\Omega_i(t)$ all along. We have shown that, without the proper delay, the process ceases to be adiabatic, providing an example of a large-angle generalized precession of $\vec{S}$ in the SU(3) space.

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APPENDIX

The equations of motion for the $N-1$ components of the generalized Bloch vector in the rotating-wave approximation are

$$\dot{u}_{j,j+1}=-\Delta_{j,j+1}v_{j,j+1}-\Omega_{j+1}v_{j,j+2}+\Omega_{j-1}v_{j-1,j+1}\,,$$

$$\dot{v}_{j,j+1}=\Delta_{j,j+1}u_{j,j+1}+\Omega_{j+1}u_{j,j+2}-\Omega_{j-1}u_{j-1,j+1}+\left[\frac{2(j+1)}{j}\right]^{1/2}\Omega_j w_j-\left[\frac{2(j-1)}{j}\right]^{1/2}\Omega_j w_{j-1}\,, \tag{A1}$$

and for $k>j+1$

$$\begin{aligned}\dot{u}_{jk}&=-\Delta_{jk}v_{jk}-\Omega_{k-1}v_{j,k-1}-\Omega_k v_{j,k+1}+\Omega_{j-1}v_{j-1,k}+\Omega_j v_{j+1,k}\,,\\ \dot{v}_{jk}&=\Delta_{jk}u_{jk}+\Omega_{k-1}u_{j,k-1}+\Omega_k u_{j,k+1}-\Omega_{j-1}u_{j-1,k}-\Omega_j u_{j+1,k}\,,\end{aligned} \tag{A2}$$

and

$$\dot{w}_{j-1}=-\left[\frac{2j}{j-1}\right]^{1/2}\Omega_{j-1}v_{j-1,j}+\left[\frac{2(j-1)}{j}\right]^{1/2}\Omega_j v_{j,j+1}\,, \tag{A3}$$

where variables with indices less than 1 or greater than N should be set equal to zero.

The steady-state equations are obtained by setting all derivatives to zero. From Eqs. (A1)–(A3) it is seen immediately that all the v_{ij} components must vanish and we are left with equations involving only u_{ij} and w_j. Specifically,

$$\sum_{i'<j'} a_{ij,i'j'} u_{i'j'} = b_{ij} \ , \quad 1 < i < j < (N-1) \tag{A4}$$

$$b_{ij} = 2\delta_{j,i+1}\Omega_i w_{i,i+1} \ ,$$
$$a_{ij,i'j'} = \delta_{ii'}\delta_{jj'}2\Delta_{ij} + \delta_{ii'}\delta_{j',j-1}\Omega_{j-1} - \delta_{ii'}\delta_{j',j+1}\Omega_j - \delta_{i',i-1}\delta_{j'j}\Omega_{i-1} - \delta_{i',i+1}\delta_{j'j}\Omega_i \ . \tag{A5}$$

It can be seen that $\det|a| \neq 0$, and there exists a unique solution for any independent choice of $w_{i,i+1}$. Since there are exactly $N-1$ independent such choices, we obtain $N-1$ independent steady-state solutions. For $N=3$ Eq. (A4) reduces to

$$\begin{aligned} &2\Delta_{12}u_{12} + \Omega_2 u_{13} = -2\Omega_1 w_1 \ , \\ &2\Delta_{23}u_{23} - \Omega_1 u_{13} = \Omega_2 w_1 - \sqrt{3}\Omega_2 w_2 \ , \\ &\Omega_2 u_{12} - \Omega_1 u_{23} + \Delta_{13} u_{13} = 0 \ , \end{aligned} \tag{A6}$$

leading to

$$\begin{aligned} u_{12} &= \Omega_1[(\Omega_1^2 - 4\Delta_{23}\Delta_{13})w_{12} + \Omega_2^2 w_{23}]/D \ , \\ u_{23} &= \Omega_2[(\Omega_2^2 - 4\Delta_{12}\Delta_{13})w_{23} + \Omega_1^2 w_{12}]/D \ , \\ u_{13} &= 2\Omega_1\Omega_2(\Delta_{23}w_{12} - \Delta_{12}w_{23})/D \ , \end{aligned} \tag{A7}$$

where

$$D = \Delta_{12}(4\Delta_{23}\Delta_{13} - \Omega_1^2) - \Omega_2^2\Delta_{23} \ . \tag{A8}$$

If we take $w_{12} = \Delta_{12}$ and $w_{23} = \Delta_{23}$, the solution is $\vec{\Gamma}$. A second independent solution is obtained by taking $w_{12} = w_{23} = D$, and the solution is then

$$\vec{\Gamma}' = \begin{bmatrix} -\Omega_1(\Omega_1^2 + \Omega_2^2 - 4\Delta_{23}\Delta_{13}) \\ -\Omega_2(\Omega_1^2 + \Omega_2^2 - 4\Delta_{12}\Delta_{13}) \\ 2\Omega_1\Omega_2(\Delta_{12} - \Delta_{23}) \\ \Omega_1^2\Delta_{12} - \Omega_2^2\Delta_{23} - 4\Delta_{12}\Delta_{23}\Delta_{13} \\ \sqrt{3}(\Omega_1^2\Delta_{12} - \Omega_2^2\Delta_{23} - 4\Delta_{12}\Delta_{23}\Delta_{13}) \end{bmatrix} . \tag{A9}$$

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[1]The amount of work already published on the three-level system is far too great to cite, of course. In Ref. 2 a wide representation of the relevant earlier literature was attempted. In Refs. 3 and 4 we list the more recent papers that have come to our attention.

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