Multilevel inversion schemes in and beyond the adiabatic limit

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A systematic investigation of N-level inversion schemes, with use of "chirped frequency lasers," in and beyond the adiabatic limit is presented. (1) It is shown that, contrary to intuition, the favorable scheme for adiabatic inversion requires that, during the process, resonances occur in reversed order, i.e., first the $(N-1)$-N transition and then backward along the atomic-level ladder down to the transition 1-2. (2) A new multilevel inversion process at fast rates (far beyond the adiabatic limit) is found. Surprisingly, the process occurs for pulse area equal to π in spite of large (time-dependent) detunings. Full analytic explanation of this generalized π pulse inversion is presented. General new properties of the multilevel adiabatic approximation are also derived and discussed.

I. INTRODUCTION

The problem of achieving multilevel excitation is of theoretical and practical interest in optical resonance physics. The dynamical evolution of the three-level system particularly has been central to investigations[1] of two-photon coherence,[2] resonance Raman scattering, double optical resonance,[3] three-level superradiance,[4] coherent multistep photoionization and photodissociation,[5] and photon echoes.[6] Recently, specific experimental[7] and theoretical[8,9] attention was given also to N-level adiabatic inversion. This process consists of achieving N-level inversion by means of a continuous sweep of the field's frequencies and/or envelopes. The term adiabatic means that the field's variations are slow enough so that a quasi steady state is maintained all along the process. Of course, this rate must compete with the existing relaxation rates. Frequency-sweeping schemes may be found particularly efficient, compared to resonant excitation, when the level scheme contains splittings and all the sublevels need to be excited as, for example, is the case in laser isotope separation processes. For $N>2$, the adiabatic criterion allows for many possibilities of the ordering by which the various field-atom resonances (including multiphoton resonances) occur during the process.[9] It turns out that different ordering schemes lead to totally different evolution of the atomic variables and consequently to a different sensitivity of the adiabatic criterion to the sweeping rates. This may be decisive in the success of the inversion process. In the present work we present a systematic comparison between various inversion schemes for a three-level system, in a continuous range covering both adiabatic and nonadiabatic regimes. We establish a previous statement[9] that the favorable scheme for adiabatic inversion does not follow the "intuitive" order of resonances, i.e., the sequential order (1-2)—(1-3)—(2-3), but the opposite, backward order.

Next we show that for intermediate schemes in which all the three resonances occur more or less at the same time, there exists a wide range of very rapid rates, far away from the adiabatic limit (where the adiabatic inversion schemes collapse), in which revival of the inversion process occurs. Surprisingly, these fast inversions occur at pulse area equal to π, in spite of large time-dependent detunings. Full analytic explanation of this generalized π pulse is presented. The program of the paper is as follows. In Sec. II and Appendix A, we present definitions and basic equations. The various adiabatic inversion schemes are introduced and discussed in Sec. III. In Sec. IV a systematic numerical comparison between these schemes is presented. The behavior of the nonadiabatic inversions is analyzed in Sec. V, using a new appropriate integral representation of the Bloch equations. For completeness and convenient reference we present a general discussion of the adiabatic approximation including miscellaneous new results in Appendixes C and D.

II. DEFINITIONS AND BASIC EQUATIONS

The density-matrix equation of motion

$$i\hbar\dot{\rho}=[H,\rho] \tag{1}$$

for an N-level chainwise connected system in the rotating-wave approximation (RWA) takes the form

$$i\dot{\rho}_{ij}+\Delta_{ij}\rho_{ij}=F_{ij}(\rho),\quad 1\le i,j\le N \tag{2}$$

with

$$F_{ij}=\Omega_{i-1}\rho_{i-1,j}-\Omega_j\rho_{i,j+1}+\Omega_i\rho_{i+1,j}-\Omega_{j-1}\rho_{i,j-1}\;, \tag{3}$$

where Ω_i is the Rabi frequency of the ith transition

$$\Omega_i=2d_i\cdot\mathscr{E}_i/\hbar\;, \tag{4}$$

d_i is the dipole matrix element between the states of levels i and $i+1$, $\mathscr{E}_i$ and φ_i are the amplitude and phase of the field

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$$\mathbf{E}=\sum_{i=1}^{N-1}\mathscr{E}_i\exp(i\varphi_i)+\text{c.c.} \tag{5}$$

coupled to the ith transition, and Δ_{ij} is the cumulative detuning between levels i and j, e.g.,

$$\Delta_{ij}=\sum_{i=1}^{j-1}\Delta_{i,i+1}\ , \tag{6}$$

$$\Delta_{i,i+1}=\omega_i-\omega_i^a\ , \tag{7}$$

where

$$\omega_i(t)=-\dot{\varphi}(t) \tag{8}$$

and ω_i^a is the atomic transition frequency between levels i and $i+1$, and the overdot stands for time derivative.

For convenience, we will also make use of another representation of Eq. (2), i.e., we define the three $[N(N-1)/2]$-dimensional vectors

$$\mathbf{U}=\begin{bmatrix}u_{12}\\u_{23}\\\vdots\\u_{13}\\u_{24}\\\vdots\\u_{N-1,N}\end{bmatrix},\quad \mathbf{V}=\begin{bmatrix}v_{12}\\v_{23}\\\vdots\\v_{13}\\v_{24}\\\vdots\\v_{N-1,N}\end{bmatrix},\quad \mathbf{W}=\begin{bmatrix}w_1\\w_2\\\vdots\\w_{N-1}\\0\\0\\\vdots\\0\end{bmatrix}, \tag{9}$$

where

$$u_{ij}=\rho_{ij}+\rho_{ji}\ ,$$
$$v_{ij}=-i(\rho_{ij}-\rho_{ji})\ , \tag{10a}$$
$$w_j=[2/j(j+1)]^{1/2}\sum_{l=1}^{j}lw_{l,l+1}\ ,$$

and

$$w_{kl}=\rho_{kk}-\rho_{ll}\ . \tag{10b}$$

It is easily seen that Eq. (2) can be written as

$$\dot{\mathbf{U}}=-\underline{\Delta}\mathbf{V}\ ,$$
$$\dot{\mathbf{V}}=\underline{\Delta}\mathbf{U}+\underline{\Omega}\mathbf{W}\ , \tag{11}$$
$$\dot{\mathbf{W}}=-\underline{\Omega}\mathbf{V}\ .$$

Equations (11) have the same structure as the well-known two-level equations except that, for $N>2$, $\underline{\Delta}$ and $\underline{\Omega}$ are matrices (given explicitly in terms of Δ_{ij} and Ω_i, in Appendix A). The matrix $\underline{\Delta}$ is symmetric and nonsingular, containing the detunings Δ_{ij} along the diagonal and Rabi frequencies or zeros elsewhere. The matrix Ω involves only Rabi frequencies and zeros and is singular.

III. ADIABATIC INVERSION SWEEPING SCHEMES IN A THREE-LEVEL SYSTEM

The adiabatic evolution of a general mixed state follows the quasisteady-state equation (see Appendix C)

$$[H(t),\rho(t)]=0 \tag{12}$$

and is consequently determined completely by the eigenvalues of the Hamiltonian (Appendix D).

For adiabatic inversion this requirement must be satisfied in particular for the ground state (at $t=0$), and for level N $[\rho_{33}(T)=1]$ at the end of the process ($t=T$). For $N>2$ Eq. (12) allows many possible paths for adiabatic inversion. In this work we investigate possible inversion schemes for a three-level system.

The conditions necessary for full adiabatic inversion at $t=T$ are[9]

$$\Delta_{12}(T)=-\Delta_{23}(0)\ , \tag{13a}$$

$$\Delta_{23}(T)=-\Delta_{12}(0)\ , \tag{13b}$$

$$\Omega_1(0)\ll\Delta_{12}(0),\quad \Omega_2(T)\ll\Delta_{23}(T)\ , \tag{13c}$$

i.e., Δ_{13} is to be swept through the point of two-photon resonance and if the Rabi frequencies Ω_i are not small enough they must be pulse shaped. In particular, we choose linear sweepings for Δ_{ij} and symmetrical pulses:

$$\Delta_{12}(t)=a-\lambda t\ , \tag{14a}$$

$$\Delta_{23}(t)=b-\lambda t\ , \tag{14b}$$

$$\Omega_i(t)=\Omega(t)\equiv\Omega_p\sin^2(\pi t/T) \tag{14c}$$

with

$$T=(a+b)/\lambda\ . \tag{14d}$$

For simplicity, we also assumed positive initial detunings a,b and equal Rabi frequencies with zero initial slope.

Our analysis considers three typical schemes.

(1) "Forward sweeping," $a<b$; the resonances occur in the intuitively favorable order (1-2)—(1-3)—(2-3).

(2) "Backward sweeping," $a>b$; the order of resonances is reversed, i.e., (2-3)—(1-3)—(1-2).

(3) "Intermediate sweeping scheme," $a\sim b$; all the resonances occur approximately at the same time.

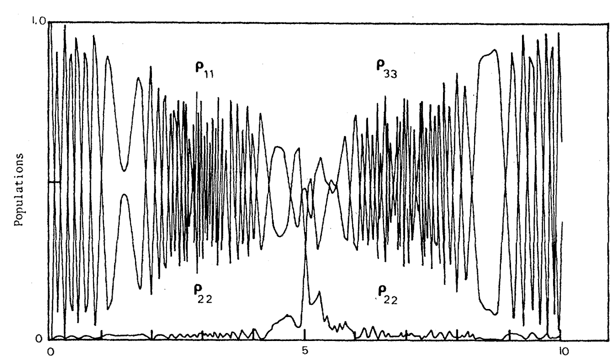

FIG. 1. Nonadiabatic evolution of populations of a three-level system, for $\Omega_1(t)=\Omega_2(t)=1$ and $\Delta_{12}(0)=0$ $[\Delta_{12}(t)=-\lambda t$, $\Delta_{23}(t)=10-\lambda t$, $t^*=10/\Delta_{13}(0)]$.

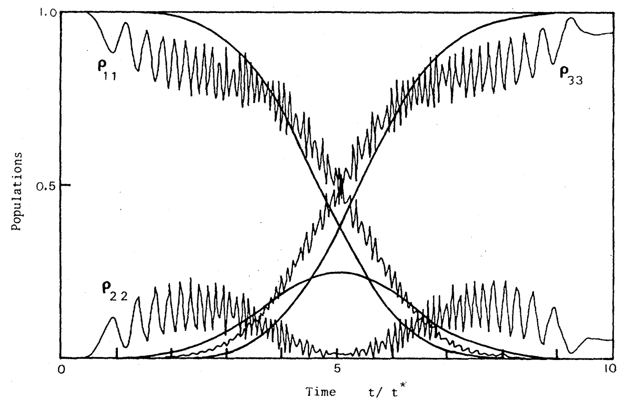

FIG. 2. Adiabatic inversion in a three-level system for the field of Eqs. (14) with $\Omega_p=6$, $a=0$, $b=10$, and $\lambda=0.1$ (oscillatory lines) and with $\Omega_p=6$, $a=10$, $b=0$, and $\lambda=0.5$ (smooth lines).

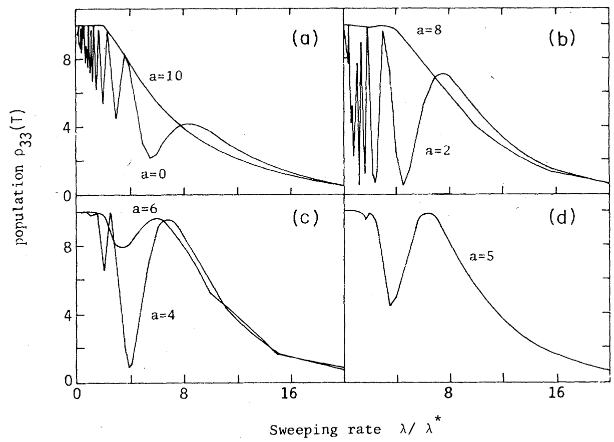

FIG. 3. Final population of level 3 vs sweeping rate λ, for various sweeping schemes (Eqs. (14), $\lambda^*=[\Delta_{13}(0)/10]^2$).

Condition (13c) must be interpreted in the sense that for adiabatic inversion the relation $\Omega_1(t) \ll \Delta_{12}(t)$ must hold as long as the ground state is almost completely populated. Clearly for a given value of $\Delta_{13}(0)=a+b$, this condition is better fulfilled by the backward-sweeping scheme ($a>b$). For the extreme case of forward sweeping with $a=0$, the laser pulse must not "start" $[\Omega_1(t)\sim 0]$ until $\Delta_{12}(t)$ acquires significant size. Otherwise, $[H(t\sim 0),\rho(t\sim 0)]\neq 0$ and the atomic state does not belong to the steady-state subspace of the Bloch equations. The atom then evolves in a nonadiabatic oscillatory manner as demonstrated by numerical solutions for $\Omega_i(t)=1$ in Fig. 1. Our sine squared pulse (14c), however, may have a very slow start and allows adiabatic inversion also for $\Delta_{12}(0)=0$. But, as seen in Fig. 2, the inversion is less stable and slower than in the backward-sweeping scheme. The conclusion is that the forward-sweeping scheme is indeed less favorable for adiabatic inversion. This statement is supported further in Sec. IV.

IV. NUMERICAL COMPARISON BETWEEN THE VARIOUS INVERSION SWEEPING SCHEMES NEAR AND BEYOND THE ADIABATIC LIMIT

In this section we summarize the behavior of the various inversion sweeping schemes (14) as a function of the sweeping rate λ, in and beyond the adiabatic limit. We have solved numerically the Bloch equations for various values of initial detunings a,b with $a+b=10$ and $\Omega_p=6$. The results, though specific, demonstrate the general features of each sweeping scheme. In Fig. 3, the final population of level 3, $\rho_{33}^f=\rho_{33}(T)$, is drawn versus the sweeping rate λ for the various schemes.

For backward sweeping with $a=10$ ($b=0$) we see in Fig. 3(a) that the inversion is complete for $\lambda<2$ and decreases gradually with λ for $\lambda>2$. As we increase "b" in favor of "a" [Figs. 3(b)—3(d)] we notice the development of a "dip" in the full inversion region. This "dip" increases with "b" and the peak on its right-hand side (rhs) moves towards larger-λ regions. As shown later on, the process then becomes less and less adiabatic. As "b" approaches "a" we enter the intermediate region $a\sim b$ [Fig. 3(d)] characterized by a large "dip" followed by a wide range of complete and almost-complete inversion. This revival of the inversion process at very short pulses (large λ) has the characteristics of a π pulse in spite of the existence of large detunings. This new inversion process will be analyzed in Sec. V.

For $b>a$ (forward sweeping) new "dips" appear in the shrinking adiabatic region and the peak on its rhs decreases. When "a" approaches zero [$b=10$, Fig. 3(a)] the adiabatic region becomes completely oscillatory with λ. The new unexpected result of the systematic investigation is the possibility of fast inversion in a continuous wide range of sweeping rates λ in the intermediate scheme. The nonadiabatic nature of this range is shown in Fig. 4, for $a=b=5$ and $\Omega_p=6$. Figure 4(a) presents the evolution of the v_{ij} coherences which constitute the absorbtive part of the atomic polarization. The adiabaticity condition (12) is characterized by $v_{ij}=0$.[9] In contrast, here v_{ij} are not small relative to u_{ij}, in fact in this case $v_{ij}>u_{ij}$. The degree of adiabaticity is best measured by the adiabatic invariants which have a clear geometrical representation using the vector model description of the Bloch equation (see Appendix B). In Fig. 4(b) the directional cosines $\cos\alpha_i$ and cosine of the "following angle" χ between the **S** vector and the $\underline{\Gamma}$ plane are drawn in comparison with the adiabatic cosines (the straight lines). This is another manifestation of the nonadiabatic nature of the fast inversion case ($\lambda\sim 7$). Further, while $\cos\chi=1$ is a general adiabatic invariant, the directional cosines $\cos\alpha_i$ become adiabatic invariants only for $a=b$, and $\Omega_1=\Omega_2$. This peculiar property is a result of a more general feature of the intermediate scheme with $a=b$ and $\Omega_1=\Omega_2$ (Cook-Shore condition[10]), which enables further detailed investigation of the fast inversion regions beyond the adiabatic limit, presented in Sec. V.

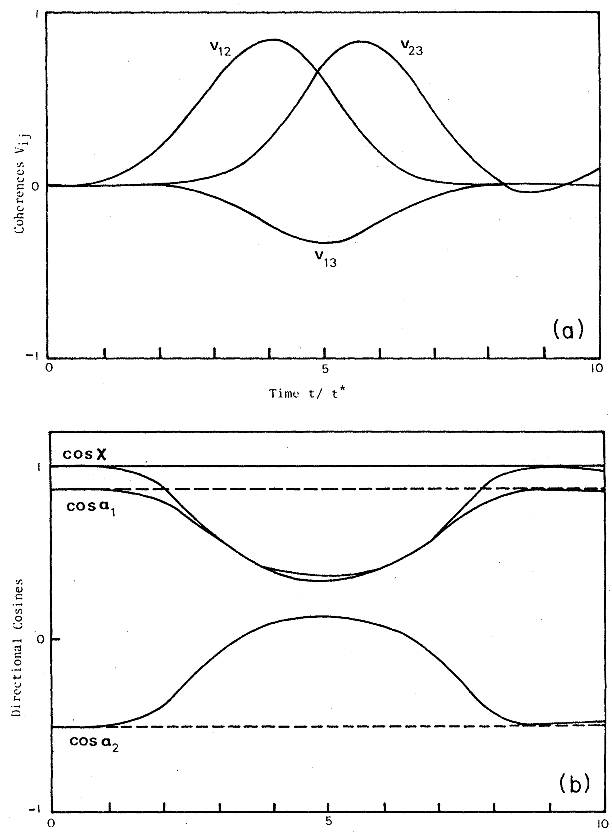

FIG. 4. (a) Evolution of the v_{ij} coherences in a nonadiabatic pulse [Eqs. (14) with $a=b=5$, $\Omega_p=6$, and $\lambda=6.7$] [for the full inversion point of Fig. 3(d)]. (b) Evolution of the directional cosines $\cos\alpha_i$ and cosine of the "following angle" χ, for adiabatic pulse [Eqs. (14) with $a=b=5$, $\Omega_p=6$, and $\lambda=0.1$ (the straight lines)], and for nonadiabatic pulse (same pulse with $\lambda=6.7$).

V. INVERSION BEYOND THE ADIABATIC LIMIT

The conditions $a=b$ and $\Omega_1=\Omega_2$ of the intermediate-sweeping scheme are a special case (for $N=3$) of the Cook-Shore conditions[10] for N-level systems:

$$\Delta_{i,i+1}=\Delta_0 ,\qquad \Omega_i=[i(N-i)]^{1/2}\Omega_0 , \tag{15}$$

for $i=1,2,\ldots,N-1$.

It was shown[11,12] that under these conditions, the dynamics of the N-level system breaks down to N modes which evolve independently. It was also shown[12] that one of these modes contains three components and behaves like a two-level system. Specifically, for our three-level system with $\Omega_1=\Omega_2=\sqrt{2}\Omega_0$, and $\Delta_{12}=\Delta_{23}=\Delta_0$, the three combinations

$$U=\tfrac{1}{2}(u_{12}+u_{23}) ,\quad V=\tfrac{1}{2}(v_{12}+v_{23}) ,\quad W=w_{13} , \tag{16}$$

obey the familiar two-level-like equations of motion:

$$\dot U=-\Delta_0 V ,\quad \dot V=\Delta_0 U+\Omega_0 W ,\quad \dot W=-\Omega_0 V . \tag{17}$$

The inversion w_{13} of our three-level system in the intermediate-sweeping scheme can therefore be analyzed within the framework of the two-level-like equations (17) with

$$\Delta_0=a-\lambda t ,\quad \Omega_0=\Omega_i/\sqrt{2}=\Omega_p'\sin^2[\pi\lambda t/(2a)] ,\quad \Omega_p'=\Omega_p/\sqrt{2},\ a=5 . \tag{18}$$

We start the analysis of the nonadiabatic fast inversion process by noticing that it occurs at rate λ [$\lambda=6.7$, Fig. 3(d)], for which the pulse area

$$\vartheta(T)=\int_0^T \Omega_0 dt=a\Omega_p'/\lambda \tag{19}$$

is equal to π. This is rather surprising, since in the absence of sweeping, full inversion occurs at $\vartheta(T)=\pi$ only for resonant excitation ($\Delta_0=0$), whereas in our case $\Delta_0(t)$ is large ($a\sim\Omega_p'$). In order to reconcile this rather peculiar finding we introduce a new integral representation of the Bloch equation (17). Specifically, we used the Green's-function technique in a similar manner to Appendix C, only here the source term is not F_{ij} as in Eq. (C1), but $\Delta_{ij}\rho_{ij}$. The result is the following exact integral equation:

$$\mathbf{S}=\begin{bmatrix}U(t)\\V(t)\\W(t)\end{bmatrix}=\mathbf{S}_1+\mathbf{S}_2 , \tag{20}$$

where $\mathbf{S}_1$ is the usual on-resonance π-pulse solution (in the absence of the source term)

$$\mathbf{S}_1=\begin{bmatrix}0\\ \sin\vartheta(t)\\ \cos\vartheta(t)\end{bmatrix} , \tag{21}$$

and $\mathbf{S}_2$ is the correction due to nonvanishing Δ_0,

$$\mathbf{S}_2=\begin{bmatrix}-\int_0^{\vartheta}(\Delta_0/\Omega_0)V\,d\vartheta'\\ \int_0^{\vartheta}(\Delta_0/\Omega_0)U\cos(\vartheta'-\vartheta)d\vartheta'\\ \int_0^{\vartheta}(\Delta_0/\Omega_0)U\sin(\vartheta'-\vartheta)d\vartheta'\end{bmatrix} . \tag{22}$$

In Eqs. (21) and (22)

$$\vartheta(t)=\int_0^t \Omega_0(t')dt' . \tag{23}$$

The variables U,V,W,Δ_0,Ω_0 in Eq. (23) depend on ϑ'. In general $|\mathbf{S}(t)|=|\mathbf{S}_1(t)|=1$, so that both $\mathbf{S}$ and $\mathbf{S}_1$ rotate on the unit sphere with $2\mathbf{S}_1\cdot\mathbf{S}_2=-\mathbf{S}_2^2$. From this representation it is clear that if at the end of the pulse ($t=T$), $\vartheta(T)=\pi$ and $\mathbf{S}_2(t)=0$, full inversion occurs inspite of large detunings.

The consistency of these conditions is easily verified. Since Δ_0/Ω_0 is antisymmetric around $t=T/2$, if W is also antisymmetric then it follows from Eq. (17) that U and V are both symmetric and therefore $\mathbf{S}_2(T)$ vanishes yielding full inversion at $\vartheta(T)=\pi$.

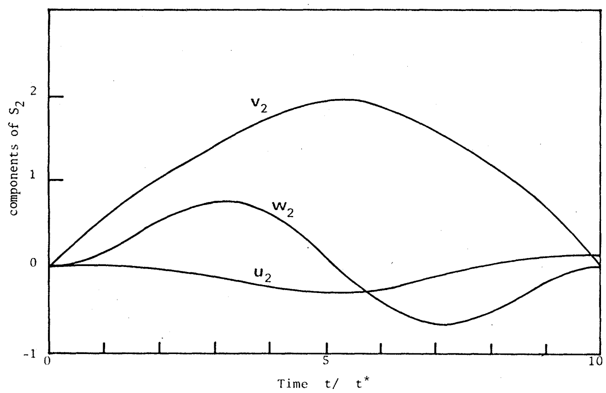

FIG. 5. Evolution of $\mathbf{S}_2=\mathbf{S}-\mathbf{S}_1$ at the fast inversion point of Fig. 3(d) ($a=b=5$, $\Omega_p=6$, $\lambda=6.7$).

This is demonstrated in Fig. 5 in which the evolution of the solution for $\mathbf{S}_2$ is shown to fulfill the required symmetry (obeyed always for $\mathbf{S}_1$). Figure 5 also exposes the dominant role of $\mathbf{S}_2(t)$ during the process so that apart from the end points $\mathbf{S}_2(t)\neq 0$ and the behavior of the atomic variables is completely different from that of a π pulse.

It should be noticed that this new inversion process lies in between the two limits $\Omega_p \gg a$ (π-pulse regime) and $\Omega_p \ll a$ (the adiabatic regime). A comparison between the various regimes is presented in Fig. 6 which further supports the above analysis. All the full inversion peak points occur at λ values $\lambda_n = a\Omega_p'(2n-1)\pi$ for which the pulse area is $\vartheta(T)=(2n-1)\pi$. The upper dashed line corresponds to the case $\Omega_p' > a$ for which the usual π-pulse characteristics are predominant. The solid line corresponds to moderate $\Omega_p' \sim a$ where the new inversion points, discussed in detail above, occur. The lower dashed line refers to weak $\Omega_p' < a$ for which the pulse area is smaller than π for all rates λ beyond the adiabatic regime and no λ oscillations occur.

VI. SUMMARY AND REMARKS

Resonant excitation may not be efficient in realistic situations where the atomic-level system is characterized by large splittings. For such systems inversion schemes in which the laser's frequency is swept through the various possible resonances may be advantageous. With this main motivation in mind we have presented in this work a systematic analysis of various inversion sweeping schemes in and beyond the adiabatic limit, for a three-level system. We have shown that for the adiabatic case the intuitive sweeping scheme for which the order of resonances occurs in succession along the atomic-levels ladder is very sensitive to the sweeping rates and is therefore less favorable for inversion. Further, we have found a set of conditions under which unexpectedly nonadiabatic inversion takes place in a large range of fast sweeping rates avoiding the necessity to compete with relaxations. This process was examined and explained both by numerical solution of the Bloch equations and analytically by a new integral representation of the Bloch equation. For completeness we have presented in the Appendixes C and D general new properties of multilevel adiabatic approximation, namely, the following: (1) Perturbation expansion which allows first-order corrections to the adiabatic limit, and the derivation of closed expressions for the first nonzero contributions to the v_{ij} coherence which govern the propagation of the laser pulses; (2) the connection between the constants of motion of the Bloch equation and the adiabatic limit is discussed yielding closed expressions for the adiabatic solution for a general mixed-state case.

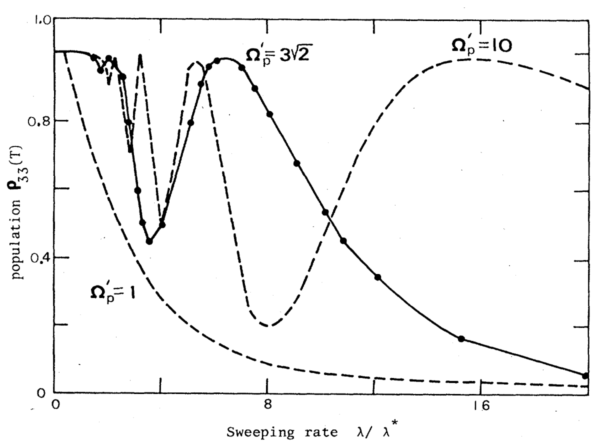

FIG. 6. Final population of level 3 for the Cook-Shore system [Eqs. (14), with $a=b=5$] vs sweeping rate λ for various field intensities Ω_p'.

ACKNOWLEDGMENTS

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APPENDIX A: MATRIX REPRESENTATION OF THE N-LEVEL BLOCH EQUATIONS

The N-level Bloch equations (2) may be written in matrix notation as follows:

$$\dot{\mathbf{U}} = -\underline{\Delta}\mathbf{V}\,,$$
$$\dot{\mathbf{V}} = \underline{\Delta}\mathbf{U} + \underline{\Omega}\mathbf{W}\,, \tag{A1}$$
$$\dot{\mathbf{W}} = -\underline{\Omega}\mathbf{V}\,.$$

The vectors $\mathbf{U},\mathbf{V},\mathbf{W}$ were defined in Eq. (9), and elements of the matrices $\underline{\Delta}$ and $\underline{\Omega}$ are given by

$$\Delta_{jk,j'k'} = \delta_{jj'}\delta_{kk'}\Delta_{jk} + \delta_{j'j}(\delta_{k',k-1}\Omega_{k-1} + \delta_{k',k+1}\Omega_k) - \delta_{k'k}(\delta_{j',j-1}\Omega_{j-1} + \delta_{j',j+1}\Omega_j) \tag{A2}$$

and

$$\Omega_{jk,j'k'} = \delta_{k',j'+1}\delta_{k,j+1}(-\delta_{j'j}\alpha_j + \delta_{j',j-1}\beta_j) \tag{A3}$$

with

$$\alpha_j = [2(j+1)/j]^{1/2}, \quad \beta_j = [2(j-1)/j]^{1/2}\,,$$

and where δ_{ij} is the Kronecker delta function.

Since each component of the vectors $\mathbf{U}$, $\mathbf{V}$, and $\mathbf{W}$ of Eq. (10) is specified by a pair of indices jk satisfying $1 < j < k < N$, i.e., $j,k = 1,2;2,3;\ldots;1,3;2,4;\ldots$, these pairs serve also for indexing rows as well as columns of the matrices $\underline{\Delta}$ and $\underline{\Omega}$.

For $N=3$ we have

$$\underline{\Delta}=\begin{bmatrix}\Delta_{12} & 0 & \Omega_2\\ 0 & \Delta_{23} & -\Omega_1\\ \Omega_2 & -\Omega_1 & \Delta_{13}\end{bmatrix} \tag{A4}$$

and

$$\underline{\Omega}=\begin{bmatrix}2\Omega_1 & 0 & 0\\ \Omega_2 & -\sqrt{3}\Omega_2 & 0\\ 0 & 0 & 0\end{bmatrix}. \tag{A5}$$

APPENDIX B: THE GEOMETRICAL PICTURE OF N-LEVEL ADIABATIC FOLLOWING (REF. 9)

Equation (10) defines the N^2-1 components of the SU(N) coherence vector **S**,

$$\mathbf{S}=(u_{12},u_{23},\ldots,u_{13},u_{24},\ldots,v_{12},v_{23},\ldots,w_1,w_2,\ldots,w_{N-1})\ . \tag{B1}$$

The Hamiltonian is represented by the vector $\mathbf{\Gamma}$:

$$\mathbf{\Gamma}=(-\Omega_1,-\Omega_2,\ldots,-\Omega_{N-1},0,0,0,\ldots,0,\Delta_1,\Delta_2,\ldots,\Delta_{N-1})\ . \tag{B2}$$

The Bloch equation (1) takes the form of a generalized cross-product equation:

$$[\dot{\mathbf{S}}]_i=[\mathbf{\Gamma}\times\mathbf{S}]_i=\sum f_{ijk}\Gamma_j S_k\ , \tag{B3}$$

where f_{ijk} are the completely antisymmetric structure constants of the SU(N) group and the Γ_j and S_k are the components of $\mathbf{\Gamma}$ and **S**, respectively. The steady-state subspace $\underline{\Gamma}$ of Eq. (B3) contains $N-1$ orthonormal solutions $\mathbf{\Gamma}_1,\mathbf{\Gamma}_2,\ldots,\mathbf{\Gamma}_{N-1}$ of the equation

$$\mathbf{\Gamma}\times\mathbf{X}=\mathbf{0}\ . \tag{B4}$$

These basis vectors are explicit in terms of the field parameters Δ_{ij} and Ω_i with zero v_{ij} components. The motion of the vector **S** is a generalized precession around the whole steady-state subspace, i.e.,

$$\dot{\mathbf{S}}\times\mathbf{\Gamma}_j=\mathbf{0}\ \text{ for all } j\ . \tag{B5}$$

The angle $\mathcal{X}$ between **S** and its projection on $\underline{\Gamma}$ is given in terms of the directional cosines $\cos\alpha_i=\mathbf{S}\cdot\mathbf{\Gamma}_i/|S|$ by

$$\cos\mathcal{X}=\left[\sum_{i=1}^{N-1}(\cos\alpha_i)^2\right]^{1/2}\ . \tag{B6}$$

The adiabatic solution is characterized by the adiabatic invariant $\mathcal{X}(t)=0$.

The directional cosines $\cos\alpha_i$ which are not adiabatic invariants determine the adiabatic solution:

$$\mathbf{S}=\sum_{i=1}^{N-1}\cos\alpha_i(t)\mathbf{\Gamma}_i(t)\ . \tag{B7}$$

For the Cook-Shore system the directional cosines $\cos\alpha_i$ become additional adiabatic constants.

APPENDIX C: PERTURBATION EXPANSION OF THE N-LEVEL BLOCH EQUATIONS NEAR THE ADIABATIC LIMIT

The criterion $[H(t),\rho(t)]=0$ valid in the adiabatic limit was obtained from geometrical considerations intuitively.[9] However, a rigorous expansion of the Bloch equation near the adiabatic limit, yielding the quasisteady-state solution as a zeroth-order approximation, was given only for the two-level system.[13] This expansion allowed an extension of the solution beyond the adiabatic limit by taking higher orders in the expansion giving also the validity criterion for the adiabatic approximation.

In this appendix we present a similar treatment for the N-level Bloch equations. As in the two-level case the form appropriate for expansion near the adiabatic limit is obtained using the Green's function of the operator on the left-hand side of Eq. (2) which includes only the field detunings. The Rabi frequencies are all included in the "source" term F_{ij}. The result is

$$i\rho_{ij}(t)=\int_0^\infty \exp\{i[\phi_{ij}(t)-\phi_{ij}(t-\tau)]\}F_{ij}(t-\tau)d\tau\ , \tag{C1}$$

where

$$\phi_{ij}(0)=0 \text{ and } \dot{\phi}_{ij}(t)=\Delta_{ij}(t)\ . \tag{C2}$$

Equation (C1) is similar to the integral form used by Crisp[13] for the two-level system. It should be noticed, however, that our $\Delta_{ij}(t)$ include the phase modulation terms. We now expand $\phi_{ij}(t-\tau)$ and $F_{ij}(t-\tau)$ for $i=j$ around $\tau=0$ (for convenience we occasionally omit the indices ij), i.e.,

$$\begin{aligned} i\rho &= \int_0^\infty \exp[i\Delta(t)\tau][1-(i/2)\dot{\Delta}(t)\tau^2]\\ &\quad\times[F(t)-\dot{F}(t)\tau+\cdots]d\tau\\ &=-i\sum_{n=0}^{\infty} a_n(t)n!/[i\Delta(t)]^{n+1}\ . \end{aligned} \tag{C3}$$

The coefficient $a_n(t)$ is explicit in terms of the few derivatives of F and Δ which contribute to the term τ^n. When phase modulation is excluded $a_n(t)=[(-1)^n/n!](d/dt)F$. In general for $i\neq j$ we obtain

$$\rho_{ij}=(1/\Delta_{ij})\{F_{ij}(t)+(d/dt)[F_{ij}(t)/i\Delta_{ij}(t)]+R\}\ , \tag{C4}$$

where R involves higher derivatives of F_{ij} and Δ_{ij}.

Equation (C4) resembles Crisp's result, except here we deal with an N-level system and our first-order term includes phase modulation $[\Delta_{ij}=\Delta_{ij}(t)]$. It is seen from

Eq. (C3) that the series converges rapidly for large Δ_{ij} ($\Delta_{ij} \gg \Omega_i$) or if higher derivatives of Δ_{ij} and F_{ij} are also smaller. Intuitively, the first-order approximation (neglecting R) is valid if

$$|(d/dt)(F_{ij}/\Delta_{ij})| \ll |F_{ij}| \ . \quad \text{(C5)}$$

Equation (C5) is the adiabatic condition leading to the N-level adiabatic solution.

The equations for the two leading orders of Eq. (29) are

$$\Delta_{ij}\rho_{ij}^{(0)} - F_{ij}^{(0)} = 0, \quad i \neq j \quad \text{(C6)}$$

and

$$\Delta_{ij}(\rho_{ij}^{(0)} + \rho_{ij}^{(1)}) = F_{ij}^{(0)} + F_{ij}^{(1)} - i(d/dt)(F_{ij}^{(0)}/\Delta_{ij}) \ , \quad \text{(C7)}$$

where $F_{ij}^{(k)} = F_{ij}(\rho^{(k)})$. Derivatives of $(F_{ij}^{(1)}/\Delta_{ij})$ belong to higher orders and were omitted.

It should be noticed that F_{ij} with $i \neq j$ depends also on populations ρ_{kk} which must be solved directly from Eq. (2) in consistency with Eq. (C6). It is readily seen that $\rho_{ij}^{(0)}$ is the steady-state solution of the Bloch equations (2), i.e.,

$$[H(t), \rho^{(0)}] = 0 \ . \quad \text{(C8)}$$

Combining Eqs. (C6) and (C7), we obtain also

$$i(d/dt)\rho^{(0)} = [H(t), \rho^{(1)}] \ . \quad \text{(C9)}$$

Equations (C8) and (C9) of the two leading orders define the adiabatic following approximation. In addition, these equations also determine the first nonzero contribution to the v_{ij} coherences which are vanishingly small in an adiabatic process, but govern the propagation of the laser fields. The explicit expressions for v_{ij} in terms of the zeroth-order solution $\rho_{ij}^{(0)}$ are obtained easily using the notation (11) of the Bloch equations. The equations for the two leading orders [viz., (C8) and (C9)] in this notation are written in terms of the $\underline{\Delta}$ and $\underline{\Omega}$ matrices as follows:

$$\mathbf{V}^{(0)} = \mathbf{0} \ , \quad \text{(C10)}$$

$$\underline{\Delta}\mathbf{U}^{(0)} + \underline{\Omega}\mathbf{W}^{(0)} = \mathbf{0} \quad \text{(C11)}$$

and

$$\dot{\mathbf{U}}^{(0)} = -\underline{\Delta}\mathbf{V}^{(1)} \ , \quad \text{(C12)}$$

$$\dot{\mathbf{V}}^{(0)} = \underline{\Delta}\mathbf{U}^{(1)} + \underline{\Omega}\mathbf{W}^{(1)} = \mathbf{0} \ , \quad \text{(C13)}$$

$$\dot{\mathbf{W}}^{(0)} = -\underline{\Omega}\mathbf{V}^{(1)} \ . \quad \text{(C14)}$$

From Eqs. (C11)–(C13) we get

$$\mathbf{V}^{(1)} = (\underline{\Delta}^2 + \underline{\Omega}\,\underline{\tilde{\Omega}})(\underline{\dot{\Delta}}\mathbf{U}^{(0)} + \underline{\dot{\Omega}}\mathbf{W}^{(0)})$$
$$= (\underline{\Delta}^2 + \underline{\Omega}\,\underline{\tilde{\Omega}})^{-1}(\underline{\dot{\Delta}}\,\underline{\Delta}^{-1}\underline{\Omega} + \underline{\Delta}^{-1}\underline{\Omega}\,\underline{\dot{\Omega}})\mathbf{W}^{(0)} \ . \quad \text{(C15)}$$

The matrix $(\underline{\Delta}^2 + \underline{\Omega}\,\underline{\Omega})$ is nonsingular since $\underline{\Delta}^2$ involves the detunings Δ_{ij} in all its diagonal elements and $\underline{\Omega}\,\underline{\tilde{\Omega}}$ does not involve Δ_{ij} at all. Since the matrices $\underline{\Delta}$ and $\underline{\Omega}$ are explicit (Appendix B), the $v_{ij}^{(1)}$ coherences are determined by the zeroth-order adiabatic solution.

Finally, we now introduce two by-products valid in the adiabatic limit [Eqs. (C10)–(C15)], i.e., the population's rate equation

$$\dot{\mathbf{W}}^{(0)} = -\underline{Z}\mathbf{W}^{(0)} \ , \quad \text{(C16)}$$

where

$$\underline{Z} = (\underline{I} + \underline{\tilde{M}}\,\underline{M})\underline{\tilde{M}}\,\underline{\dot{M}} \quad \text{(C17)}$$

with

$$\underline{M} = \underline{\Delta}^{-1}\underline{\Omega} \ , \quad \text{(C18)}$$

and the population's constant of motion

$$(d/dt)[\mathbf{W}(\underline{I} + \underline{\tilde{M}}\,\underline{M})\mathbf{W}] = \mathbf{0} \ . \quad \text{(C19)}$$

For a two-level system, $M = \Omega/\Delta$ is a scalar and Eq. (C16) can be easily integrated analytically yielding the well-known solution $W^{(0)} = \Delta/(\Delta^2 + \Omega^2)^{1/2}$. For $N > 2$, however, the matrices $\underline{\tilde{M}}$ and $\underline{\dot{M}}$ do not commute and direct integration of Eq. (C19) is not possible.

APPENDIX D: CONSTANTS OF MOTION AND MULTILEVEL ADIABATIC FOLLOWING

The propagator of the density matrix $\rho(t)$ is given by

$$\rho(t) = U(t)\rho(0)U^{\dagger}(t) \ , \quad \text{(D1)}$$

where $U(t)$ is the propagator of the Schrödinger state vector. It should be noticed that this propagator is valid for the general mixed-state case and also for $\rho(t)$ in the rotating-wave approximation.

For Hermitian Hamiltonians (no decays) Eq. (D1) is a similarity transformation which leaves the eigenvalues r_i of ρ invariant even though the Hamiltonian is time dependent and does not commute with ρ. It is easy to see that the set of constants of motion r_i is equivalent to the set of constants $\mathrm{Tr}[\rho(t)]^n$ presented recently,[14,15] since

$$\mathrm{Tr}[\rho^n] = \sum_{i=1}^{N} r_i^n \quad \text{(D2)}$$

for all $n = 1, 2, \ldots, N$. Equation (D1) may thus be written as

$$A^{\dagger}(t)\rho(t)A(t) = A^{\dagger}(0)\rho(0)A(0) = R \ , \quad \text{(D3)}$$

where $\underline{R}$ is a constant diagonal matrix with $R_{ij} = \delta_{ij} r_i$.

Equations (D1) and (D3) are just formal representations of the Bloch equation since solving for $U(t)$ or $A(t)$ is equivalent to solving for $\rho(t)$. However, for the adiabatic process $[H(t), \rho(t)] = 0$ and both H and ρ are diagonalized by the same matrix (if H is nondegenerate). The matrix $A(t)$ of Eq. (D3) is therefore constructed simply by the eigenvectors of the Hamiltonian. In this case $U(t) = A(t)A^{\dagger}(0)$ and Eq. (D1) gives an explicit solution for $\rho(t)$ in terms of the components of the Hamiltonians eigenvectors. For the pure state $\rho_{11}(0) = 1$ this solution coincides with the dressed states of the Schrödinger equation.[8]

The quasienergy of the adiabatic system is given in terms of the constants of motion r_i and the eigenvalues of the Hamiltonian λ_i by

$$\bar{E}(t) \equiv \langle \psi | H | \psi \rangle = \mathrm{Tr}[\rho H] = \sum_{i=1}^{N} r_i(0)\lambda_i(t) \ . \quad \text{(D4)}$$

This quasienergy plays an interesting role in the geometrical interpretation of the Bloch equations. Specifically, it is equal to twice the scalar product $\mathbf{S}\cdot\mathbf{\Gamma}$ (Ref. 9) and the adiabatic directional cosine $\cos\alpha_1$ is therefore related to the Hamiltonian eigenvalues λ_i by

$$\cos\alpha_1(t)=\tfrac{1}{2}\sum_i r_i(0)\lambda_i(t)/|S|\,|\Gamma| \; . \tag{D5}$$

For the three-level system the characteristic equation is

$$\begin{aligned}&\sum_{i=0}^{3} a_i\lambda^i=0 \; ,\\ &a_0=\Omega_1^2\Delta_{12}/4 \; ,\\ &a_1=\Delta_{12}\Delta_{23}-(\Omega_1^2+\Omega_2^2)/4 \; ,\\ &a_2=-(\Delta_{12}+\Delta_{23}) \; ,\\ &a_3=1 \; ,\end{aligned} \tag{D6}$$

and the eigenvectors belonging to λ_m, $m=1,2,3$ are

$$\begin{bmatrix}A_{1m}\\A_{2m}\\A_{3m}\end{bmatrix}=\frac{1}{D_m}\begin{bmatrix}-\Omega_1(\lambda_m-\Delta_{23})\\2\lambda_m(\lambda_m-\Delta_{23})\\-\Omega_2\lambda_m\end{bmatrix} \; , \tag{D7}$$

where D_m is the normalization factor.

If $\rho(0)$ is diagonal, $A(0)$ is the unit matrix and the adiabatic solution is given by

$$\begin{aligned}&\rho_{11}=\Omega_1^2\sum_m(\lambda_m-\Delta_{23}^2)^2\sigma_m \; ,\\ &\rho_{22}=4\sum_m\lambda_m^2(\lambda_m-\Delta_{23})^2\sigma_m \; ,\\ &\rho_{33}=\Omega_2^2\sum_m\lambda_m^2\sigma_m \; ,\\ &\rho_{12}=\rho_{21}=-2\Omega_1\sum_m\lambda_m(\lambda_m-\Delta_{23})^2\sigma_m \; ,\\ &\rho_{23}=\rho_{32}=-2\Omega_2\sum_m\lambda_m^2(\lambda_m-\Delta_{23})\sigma_m \; ,\\ &\rho_{13}=\rho_{31}=\Omega_1\Omega_2\sum_m\lambda_m(\lambda_m-\Delta_{23})\sigma_m \; ,\end{aligned} \tag{D8}$$

where

$$\sigma_m=\rho_{mm}(0)/D_m^2 \; .$$

The explicit expressions for the roots λ_m of Eq. (D6) are

$$\begin{aligned}&\lambda_m=2\sqrt{q}\,\cos[\vartheta+2m\pi/3]-a_2/3 \; ,\\ &q=(a_2^2-3a_1)/9 \; ,\\ &r=(9a_1a_2-27a_0-2a_2^3)/54 \; ,\\ &\vartheta=\cos^{-1}(r/q) \; .\end{aligned} \tag{D9}$$

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[1]The amount of work already published on the three-level system is, of course, far too great to cite. In Refs. 2–6 we mention only representative works. A wide list of relevant literature was presented in F. T. Hioe and J. H. Eberly, Phys. Rev. Lett. **47**, 838 (1981).

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