Coherent dynamics and complete population depletion of a special three-level quantum system

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By constructing a vector model we study the coherent dynamics of a special three-level quantum system—one with equal detunings and equal Rabi frequencies. This three-level problem can be fully related to a two-level problem. We find that complete population depletion can be achieved even in the case of constant nonzero detunings when the three-level system is prepared initially in the common level of the two transitions. Complete population depletion with constant detunings can also occur in other odd-number multilevel systems. An explanation in terms of an equivalent resonant excitation is presented.

I. INTRODUCTION

The problem of a two-level atom coupled to an external potential studied since the early years of quantum theory, continues to attract the attention of researchers. In spite of the fact that it represents the simplest nontrivial time-dependent problem, it is often a good approximation to real systems in which the field frequency nearly matches that of an atomic transition. In recent years some attention has been directed to the question of determining when such a system may be completely inverted by radiation pulses, namely, the population being transferred with 100% probability from the initially occupied to the initially empty level. Hioe[1] has addressed this matter by looking at amplitude and frequency modulation functions for which the equations of motion admit to exact solutions, and Robinson[2] has shown that the relevant features are related to temporal symmetries of the pulses. The specific result obtained indicated that a two-level system which is subject to a pulse with symmetric amplitude modulation and antisymmetric frequency modulation may be inverted provided the carrier frequency is exactly resonant with the atomic transition. (The probe amplitude is always taken to be positive.) If the atom-field detuning is a nonzero constant, complete inversion does not occur.

The purpose of the present paper is to analyze analogous effects in certain three-level systems. We assume one common level ($|2\rangle$) to be connected to the other ($|1\rangle$ and $|3\rangle$) levels, while $|1\rangle$ and $|3\rangle$ are not directly coupled. In addition, the atom-field detunings between $|2\rangle$ and the others are equal and the coupling strengths of the two atomic transitions are also equal (equal Rabi frequencies) (see Fig. 1).[3] These features can arise by illuminating the atoms either with two laser fields, or with one field[4] coupling $|2\rangle$, for example, to two Zeeman levels of a P_1 state as shown in Fig. 2. We find that, in contrast to the two-level problem, complete population depletion is possible for constant nonzero detunings in the three-level problem. Although this result is implicit in the work of Cook and Shore,[5] it was not discussed by these authors. We help to interpret this result by reducing the three-level equations of motion to a form that resembles an equivalent resonant coupled two-level problem. We note at this point that for three-level systems, complete population depletion (CPD) of a prepared state is *not*, in general, identical to complete population inversion (CPI), i.e., the transfer to a *single* initially empty level. CPI always implies CPD whereas CPD implies CPI only in a two-level system. Conditions for both CPD and CPI are discussed in Sec. III.

Some of our results for three-level systems also apply to systems with more levels, provided that the number of active states is odd. Cook and Shore[5] have already studied such N-level systems for constant detunings and Rabi frequencies. Our result generalizes theirs to time-dependent detunings and Rabi frequencies for $N=3$. The three-level system with equal detunings and equal Rabi frequencies and the Cook-Shore type N-level system have been studied by Hioe *et al.* in terms of a density matrix description,[3,6,7] which is more general than the probability amplitude (wave equation) description. For quantum systems in which decay, collisions, and incoherent pumping are absent, one can first solve for probability amplitudes and then combine probability amplitudes to get the corresponding solution for density matrix elements. Solving problems in this way has its advantages, for it is easier to solve the amplitude equations. When initial conditions for the density matrix elements of a quantum system can be rewritten in terms of probability amplitudes, one may simply apply a probability amplitude description to the

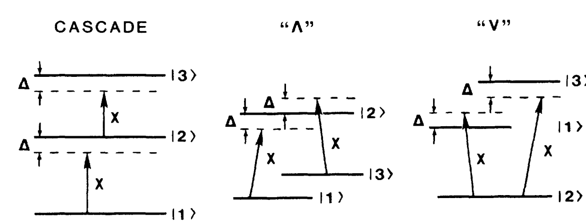

FIG. 1. Three types of three-level atoms driven by two laser fields such that the two transitions have equal detunings Δ and equal Rabi frequencies χ. Our analysis applies to all three types.

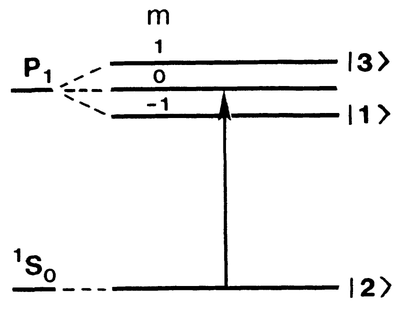

FIG. 2. A single laser field with linear polarization $\mathscr{E}$ which is perpendicular to an applied magnetic field **B** couples atomic state $^1S_0$ to two Zeeman levels ($m=\pm 1$) of a P_1 state. When the laser frequency Ω is tuned to the $^1S_0-P_1$ ($m=0$) transition frequency, the atom and laser field constitute a three-level system with equal detunings and equal Rabi frequencies.

whole process. In this work we shall employ probability amplitudes to describe the three-level system with equal detunings and equal Rabi frequencies and construct a vector model for it.

Due to their intuitive appeal, vector models are often constructed for atomic systems. The most familiar example of a vector model is one in which the components of this vector are combinations of density matrix elements, both in two-level Bloch equations[8] and in two-photon transitions in a three-level system.[9] A vector model is given in the Cook and Shore treatment of the N-level system.[5] An (N^2-1)-dimensional vector description in terms of density matrix elements applies to an N-level system.[10]

The particular vector model appropriate to the present problem will be described in Sec. II. In Sec. III we specialize to the case where the atom is prepared in the common level of two transitions and show that complete depletion with constant detunings can be realized with such initial conditions. The descriptions in terms of equivalent two-state equations with zero detuning is given also in this section. In Sec. IV we demonstrate that CPD with constant (nonzero) detuning can also occur in a Cook-Shore N-level system when N is odd.

II. DYNAMICS OF THE THREE-LEVEL SYSTEM WITH EQUAL DETUNINGS AND EQUAL RABI FREQUENCIES

A. Vector model

We consider a three-level atomic system which interacts with two laser fields. Nonzero dipole moments exist only between eigenstates $|1\rangle$ and $|2\rangle$, and $|2\rangle$ and $|3\rangle$. The electric fields can be both amplitude and frequency modulated and are of the form

$$\mathbf{E}(\mathbf{r},t)=\mathscr{E}_1(t)\cos[\Omega_1(t)t-\mathbf{k}_1\cdot\mathbf{r}+\phi_1(t)] + \mathscr{E}_2(t)\cos[\Omega_2(t)t-\mathbf{k}_2\cdot\mathbf{r}+\phi_2(t)] \ . \tag{2.1}$$

The laser field $(\mathscr{E}_1,\Omega_1)$ drives the 1-2 transition only, while the laser field $(\mathscr{E}_2,\Omega_2)$ drives the 2-3 transition only. The two atom-field detunings are

$$\Delta_{21}(t)=\omega_{21}-\mathrm{sgn}(\omega_{21})\frac{d}{dt}(\Omega_1 t+\phi_1) \ , \tag{2.2a}$$

$$\Delta_{32}(t)=\omega_{32}-\mathrm{sgn}(\omega_{32})\frac{d}{dt}(\Omega_2 t+\phi_2) \ , \tag{2.2b}$$

and the two Rabi frequencies (chosen real and positive) are

$$\chi_1(t)=\mathbf{p}_{21}\cdot\mathscr{E}_1/\hbar \ , \tag{2.3a}$$

$$\chi_2(t)=\mathbf{p}_{32}\cdot\mathscr{E}_2/\hbar \ , \tag{2.3b}$$

where ω_{jk} and $\mathbf{p}_{jk}$ are the frequency difference and the dipole moment between states $|j\rangle$ and $|k\rangle$, respectively.

One may adjust the lasers for equal detunings,

$$\Delta_{21}(t)=\Delta_{32}(t)\equiv\Delta(t) \ , \tag{2.4a}$$

and equal Rabi frequencies,

$$\chi_1(t)=\chi_2(t)\equiv\chi(t) \ . \tag{2.4b}$$

In this case the probability amplitudes $c_j(t)$ of the three states in the field-interaction representation obey the following coupled equations:

$$\frac{d}{dt}\begin{bmatrix} c_1 \\ c_2 \\ c_3 \end{bmatrix} = i \begin{bmatrix} \Delta & \frac{1}{2}\chi & 0 \\ \frac{1}{2}\chi & 0 & \frac{1}{2}\chi \\ 0 & \frac{1}{2}\chi & -\Delta \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \end{bmatrix} \ . \tag{2.5}$$

By making a linear transformation,

$$s_1=i(c_1-c_3)/\sqrt{2} \ , \tag{2.6a}$$

$$s_2=(c_1+c_3)/\sqrt{2} \ , \tag{2.6b}$$

$$s_3=ic_2 \ , \tag{2.6c}$$

one can recast Eq. (2.5) in the form

$$\frac{d}{dt}\begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix} = \begin{bmatrix} 0 & -\Delta(t) & 0 \\ \Delta(t) & 0 & \chi(t)/\sqrt{2} \\ 0 & -\chi(t)/\sqrt{2} & 0 \end{bmatrix} \begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix} \ , \tag{2.7}$$

or in a vector model form

$$\frac{d\mathbf{S}}{dt}=\mathbf{\Omega}_S\times\mathbf{S} \ , \tag{2.8}$$

with an amplitude vector

$$\mathbf{S}=\begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix} \tag{2.9a}$$

and driving field vector

$$\mathbf{\Omega}_S=\begin{bmatrix} -\chi/\sqrt{2} \\ 0 \\ \Delta \end{bmatrix} \ . \tag{2.9b}$$

The vector $\mathbf{\Omega}_S$ is real whereas the amplitude **S** is complex. Its magnitude (length) remains constant during the motion

$$||\mathbf{S}|| = (s_1 s_1^* + s_2 s_2^* + s_3 s_3^*)^{1/2} = \left[\sum_{j=1}^{3} |c_j|^2 \right]^{1/2} = 1 . \quad (2.10)$$

By separating $\mathbf{S}$ into real and imaginary parts $\mathbf{S} = \boldsymbol{\beta}_1 + i\boldsymbol{\beta}_2$, the complex vector equation (2.9) can be decomposed into two real vector equations

$$\dot{\boldsymbol{\beta}}_1 = \boldsymbol{\Omega}_S \times \boldsymbol{\beta}_1 , \quad (2.11a)$$

$$\dot{\boldsymbol{\beta}}_2 = \boldsymbol{\Omega}_S \times \boldsymbol{\beta}_2 , \quad (2.11b)$$

where

$$\boldsymbol{\beta}_1 = \begin{bmatrix} \frac{1}{\sqrt{2}} \mathrm{Im}(c_3 - c_1) \\ \frac{1}{\sqrt{2}} \mathrm{Re}(c_3 + c_1) \\ -\mathrm{Im} c_2 \end{bmatrix}, \quad \boldsymbol{\beta}_2 = \begin{bmatrix} \frac{1}{\sqrt{2}} \mathrm{Re}(c_1 - c_3) \\ \frac{1}{\sqrt{2}} \mathrm{Im}(c_1 + c_3) \\ \mathrm{Re} c_2 \end{bmatrix} . \quad (2.12)$$

Equations (2.11) indicate that the two real vectors $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$ rotate about the same driving field vector $\boldsymbol{\Omega}_S(t)$ as shown in Fig. 3.

By using Eqs. (2.11), one may easily show that $|\boldsymbol{\beta}_1|^2$, $|\boldsymbol{\beta}_2|^2$, and $\boldsymbol{\beta}_1 \cdot \boldsymbol{\beta}_2$ are three nonlinear constants of the motion. Geometrically, this means that the shape of the triangle spanned by the two vectors $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$ does not change during its motion; i.e., the triangle rotates rigidly about the instantaneous axis $\boldsymbol{\Omega}_S(t)$. Furthermore, one may deduce the following equation from Eqs. (2.11):

$$\frac{d}{dt}(\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2) = \boldsymbol{\Omega}_S \times (\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2) , \quad (2.13)$$

where

$$\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2 = \begin{bmatrix} \frac{1}{\sqrt{2}} \mathrm{Re}(c_1 c_2^* + c_2 c_3^*) \\ \frac{1}{\sqrt{2}} \mathrm{Im}(c_1 c_2^* + c_2 c_3^*) \\ \frac{1}{2}(|c_3|^2 - |c_1|^2) \end{bmatrix} . \quad (2.14)$$

Equation (2.13) demonstrates that $|\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2|^2$ is also a constant of the motion.[11] Moreover, certain results can be easily obtained by using Eq. (2.13) as will be seen in Sec. II B. When one of the $\boldsymbol{\beta}_i$ vectors is of unit length, the other, $\boldsymbol{\beta}_j$ ($j \neq i$, $i,j = 1,2$), shrinks to zero and the triangle reduces to the vector $\boldsymbol{\beta}_i$, since

$$|\boldsymbol{\beta}_1|^2 + |\boldsymbol{\beta}_2|^2 = \sum_{j=1}^{3} |c_j|^2 = 1 . \quad (2.15)$$

This situation can simplify the dynamics of the system and may be realized by preparing the system in state $|2\rangle$. A more detailed discussion for this initial condition is presented in Sec. III.

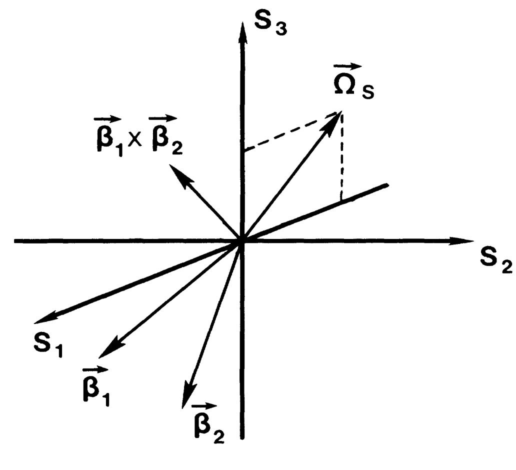

FIG. 3. The vector model of the three-level system with equal detunings and equal Rabi frequencies. Three real vectors $\boldsymbol{\beta}_1$, $\boldsymbol{\beta}_2$, and $\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2$ rotate about the same driving field vector $\boldsymbol{\Omega}_S(t)$. Note that $\boldsymbol{\beta}_1 \times \boldsymbol{\beta}_2$ is perpendicular to both $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$.

B. Connection with the two-state problem

Equations (2.8) and (2.11) have the same form as the Bloch equation describing a two-level problem with detuning $\Delta(t)$ and the Rabi frequency $\chi(t)/\sqrt{2}$, in which the Bloch vector has the components u, v, and w. The quantity ic_2 is analogous to the population difference w between the upper and lower levels of the two-level system, while $i(c_1 - c_3)/\sqrt{2}$ and $(c_1 + c_3)/\sqrt{2}$ are analogous to the real and imaginary parts (u and v, respectively) of the coherence in the field-interaction representation, respectively. As suggested in the Introduction, the two-level system is the simplest quantum system and consequently many results have been obtained for it.[1,2,8,12–19] In particular, there are modulation functions for which the two-level problem has been solved analytically.[1,8,12–14,18] These conclusions may be taken over to the special three-level system under consideration.

For a two-level atom which is initially in the ground state and excited by a nearly resonant laser field, some of the known results are (a) a hyperbolic secant coupling pulse without frequency or phase modulation of area $2m\pi$ (m integral) leads to zero transition probability to the excited state, namely complete population return (CPR), regardless of detuning,[12,14] (b) asymmetric pulses with constant detuning do not lead to CPR in general (i.e., except under accidental, overdetermined circumstances),[14,15] (c) it is impossible to get complete population depletion by any pulse with constant (nonzero) detuning, and (d) CPD can be achieved by a resonant pulse with temporally symmetric amplitude modulation and antisymmetric frequency modulation.[1,2,18] (In terms of the vector model of the two-level system, CPD implies a rotation of the Bloch vector from an initially downward orientation to an upward orientation, whereas CPR implies a rotation in which the Bloch vector returns to its initially downward orientation.)

TABLE I. Some results about complete population return, complete population depletion, and complete population inversion.

Pulse features	Two-level system initially in ground state	Three-level system ($\Delta_{21}=\Delta_{32}$, $\chi_1=\chi_2$) initially in state $\|2\rangle$	state $\|1\rangle$ or $\|3\rangle$
Hyperbolic secant without frequency modulation, area $2\eta m\pi$[a]	CPR	CPR	CPR
Asymmetric, constant nonzero detuning	no CPR	no CPR	no CPR
Constant nonzero detuning	no CPD (no CPI)	CPD possible no CPI	no CPD no CPI
$\chi(t)$ symmetric and $\Delta(t)$ antisymmetric, suitable areas such as those prescribed in Eqs. (2.16)	CPD (CPI)	CPR	CPD CPI[b]

[a] $\eta=1$ in the two-level problem whereas $\eta=\sqrt{2}$ in the three-level problem under discussion.
[b] CPI occurs only between states $|1\rangle$ and $|3\rangle$.

Viewing the three-level system with equal detunings and equal Rabi frequencies in terms of its vector model, it is straightforward to apply these results of the two-level problem to the three-level problem under discussion and arrive at the following conclusions.

(1) When the three-level system is initially in state $|1\rangle$, which corresponds to $\boldsymbol{\beta}_1\times\boldsymbol{\beta}_2$ being downward initially, two hyperbolic secant pulses each of area $\sqrt{2}m\pi$ lead to CPR for any constant detuning Δ, while it is impossible, in general, to obtain CPR by using asymmetric pulses with constant (nonzero) detuning. Moreover, complete population inversion between states $|1\rangle$ and $|2\rangle$ is impossible for any pulse, owing to the conservation of $|\boldsymbol{\beta}_1\times\boldsymbol{\beta}_2|^2$. Similar results can be reached for the three-level system initially in state $|3\rangle$. In addition, CPI between states $|1\rangle$ and $|3\rangle$ can be achieved by resonant pulses with temporally symmetric amplitude modulation and antisymmetric frequency modulation, but not by pulses with constant detuning.[20] For example, CPI between the states $|1\rangle$ and $|3\rangle$ occurs for pulses with

$$\chi(t)=\frac{\eta[(2m-1)^2+(2T\delta_0)^2]^{1/2}}{2T}\,\mathrm{sech}\left[\frac{t}{2T}\right], \quad m=1,2,3,\ldots \tag{2.16a}$$

$$\Delta(t)=\delta_0\tanh(t/2T), \tag{2.16b}$$

where $\eta=\sqrt{2}$ in the three-level problem under discussion (while $\eta=1$ in the two-level problem). Note that $\int_{-\infty}^{+\infty}\Delta(t)dt=0$.

(2) When the three-level system is initially in state $|2\rangle$, which may correspond to $\boldsymbol{\beta}_1$ being either downward or upward and $\boldsymbol{\beta}_2$ vanishing initially, CPR occurs for hyperbolic secant pulses each of area $2\sqrt{2}m\pi$ and of arbitrary constant detuning Δ; CPR can also be achieved by resonant pulses with symmetric amplitude modulation and antisymmetric frequency modulation. The pulse shape prescribed in Eqs. (2.16) is an example. Some simple analytic solutions for these CPR pulses are presented in the Appendix. On the other hand, CPR is impossible, in general, for asymmetric pulses with constant detuning. These results and some others given in Sec. III are summarized in Table I.

III. THREE-LEVEL SYSTEM INITIALLY IN STATE $|2\rangle$

A. Complete population depletion

The initial condition that the system be in state $|2\rangle$ leads to interesting results. Such an initial condition is satisfied in a "V"-type level scheme, such as that found in Na with $3\,^2P_{1/2}$, $3\,^2S_{1/2}$, and $3\,^2P_{3/2}$ corresponding to levels 1, 2, and 3, respectively. Under such initial conditions, the populations of states $|1\rangle$ and $|3\rangle$ are always the same, i.e.,

$$|c_1(t)|^2=|c_3(t)|^2, \tag{3.1}$$

which follows from the fact that $|\boldsymbol{\beta}_1\times\boldsymbol{\beta}_2|^2$ is a constant of the motion with zero initial value. Equation (3.1) tells one that the maximum possible population of state $|1\rangle$ or $|3\rangle$ is one-half, implying that CPI is impossible with the given initial condition, even for resonant fields $\Delta=0$.[21]

For the three-level system prepared initially in state $|2\rangle$, one may choose

$$c_j(t_0)=i\delta_{j2}, \tag{3.2}$$

where t_0 is the initiation time of the pulses. This corresponds to $\boldsymbol{\beta}_1=(0,0,-1)$ and $\boldsymbol{\beta}_2=\mathbf{0}$ initially [see Eqs. (2.12)]. Since $|\boldsymbol{\beta}_1|^2$ and $|\boldsymbol{\beta}_2|^2$ are constants of motion, one further obtains from Eqs. (2.12) that

$$\boldsymbol{\beta}_2(t)=\mathbf{0},\quad c_1(t)=c_3^*(t),\quad \mathrm{Re}c_2(t)=0\ ,$$
$$\boldsymbol{\beta}_1(t)=\begin{bmatrix}-\sqrt{2}\,\mathrm{Im}c_1(t)\\ \sqrt{2}\,\mathrm{Re}c_1(t)\\ -\mathrm{Im}c_2(t)\end{bmatrix}. \tag{3.3}$$

Equations (3.3) show that $c_1(t)$ and $c_3(t)$ are not mutually independent with our choice of initial condition.[22] Knowledge of one implies knowledge of the other. The projection of the vector $\boldsymbol{\beta}_1$ on the s_3 axis uniquely determines the probability amplitude $c_2(t)$. Consequently, when $\boldsymbol{\beta}_1$ lies in the $s_1 s_2$ plane, we have CPD since

$$c_2=0\ . \tag{3.4}$$

In terms of the vector model, CPD corresponds to an initially downward orientation of the $\boldsymbol{\beta}_1$ vector with respect to the horizontal $s_1 s_2$ plane, followed by a rotation into that plane. (In the two-level problem, the excitation of *half* the population from the initially populated lower state to the upper state likewise corresponds to an initially downward orientation of the Bloch vector with respect to the horizontal uv plane, followed by a rotation into that plane.) Obviously, there are many ways to accomplish this, including the use of fields with constant detunings $\Delta=\text{const}\neq 0$. This is a striking and surprising result. No matter how one gets CPD, the reorientation is accomplished as long as $\boldsymbol{\beta}_1$ is rotated into the $s_1 s_2$ plane from its initial downward position.

That CPD in state $|2\rangle$ is possible in a field with constant detuning is very interesting, since it is impossible in a two-level system. In order to understand this phenomenon, we give an explanation in terms of an equivalent resonant excitation in a reduced two-state problem in Sec. III B. Usually, CPD or CPI can be achieved in an N-level system when each of the laser fields with constant frequency and phase are resonant with the respective atomic transitions [$(N-1)$ one-photon resonances].[5,23,24] If frequency modulation is allowed, CPD can also be realized in a three-level atomic system coupled by two nearly resonant laser fields, for example[6] with detunings $\Delta_{21}(t)=-\Delta_{32}(t)\propto\tan(t/2T)$ [note this differs from condition (2.4a)] and Rabi frequencies $\chi_1(t)\propto\chi_2(t)\propto\mathrm{sech}(t/2T)$. We can show that CPI can occur in such a three-level system (i.e., with $\Delta_{21}+\Delta_{32}=0$) when $\Delta_{21}=\text{const}\neq 0$. Inversion in a multilevel system with[25] or without[4] frequency modulation through adiabatic[26] following has been studied. This does not lead to CPI or CPD beyond the adiabatic limit. The fact that CPD with constant nonzero detunings can occur in odd N-level systems is implicitly contained in the expressions given by Cook and Shore.[5] We consider this result explicitly and write the equations for $N=3$ in a form where the CPD can be explained in terms of an equivalent two-level problem *on resonance*.

B. An explanation for CPD in state $|2\rangle$

Using the relation $c_1(t)=c_3^*(t)$ from Eq. (3.3), one may write

$$c_1(t)=b(t)e^{i\psi(t)}/\sqrt{2}\ , \tag{3.5a}$$
$$c_3(t)=b(t)e^{-i\psi(t)}/\sqrt{2}\ , \tag{3.5b}$$

with $b(t)$ real and smooth. Substituting Eqs. (3.5) into Eq. (2.5), one obtains

$$\dot{c}_2=i(\chi/\sqrt{2})(\cos\psi)b\ , \tag{3.6a}$$
$$\dot{b}=i(\chi/\sqrt{2})(\cos\psi)c_2\ , \tag{3.6b}$$
$$\dot{\psi}=\Delta(t)-i(\chi/\sqrt{2})(\sin\psi)(c_2/b)\ . \tag{3.6c}$$

Equations (3.6a) and (3.6b) are the same as those for a two-state problem with coupling potential $(\chi/\sqrt{2})\cos\psi$ (i.e., Rabi frequency $\sqrt{2}\chi\cos\psi$) and zero detuning. Since $b(t)$ is real, $c_2(t)$ is pure imaginary. The formal solutions of Eqs. (3.6a) and (3.6b) are readily obtained by the use of an effective pulse area up to time t,

$$A(t)=\int_{t_0}^{t}\sqrt{2}\chi(t')\cos\psi(t')dt'\ . \tag{3.7}$$

For the initial condition (3.2), the solution is

$$c_2(t)=i\cos\tfrac{1}{2}A(t)\ , \tag{3.8a}$$
$$b(t)=-\sin\tfrac{1}{2}A(t)\ . \tag{3.8b}$$

CPD of state $|2\rangle$ can occur if the pulse area $|A|\geq\pi$, but not otherwise. Equation (3.8a) demonstrates that the evolution of the amplitude c_2 is uniquely determined by the effective pulse area A, which, in turn, is determined by reduced Rabi frequency $\sqrt{2}\chi\cos\psi$. Quantitatively the behavior of $\sqrt{2}\chi\cos\psi$ is not known unless the phase $\psi(t)$ of c_1 or c_3 is known, which, however, requires solving the complicated nonlinear equations (3.6). That is, solving Eqs. (3.6) directly is very difficult. Alternatively, one may be willing to return to the linear equations (2.11) and (3.3) for solving $\mathrm{Re}c_1$ and $\mathrm{Im}c_1$ first. This may be done by actually solving the corresponding two-state problem. Thus, for those Rabi frequencies $\chi(t)$ and detunings $\Delta(t)$ whose solutions for the two-state problem are known, one knows the solution of $\mathrm{Re}c_1(t)$ and $\mathrm{Im}c_1(t)$. These determine $\psi(t)$. We give two examples below to illustrate the explanation using the effective pulse area A.

C. Rectangular pulse

We first consider the stepwise excitation with a pulse of constant amplitude and frequency—a rectangular pulse. In this case, the orientation of the driving vector $\boldsymbol{\Omega}_S=(-\chi/\sqrt{2},0,\Delta)$ is fixed and $\boldsymbol{\beta}_1$ precesses about $\boldsymbol{\Omega}_S$ with angular frequency

$$\sigma\equiv|\boldsymbol{\Omega}_S|=(\Delta^2+\tfrac{1}{2}\chi^2)^{1/2} \tag{3.9}$$

and fixed angle $\arctan|(\chi/\sqrt{2})/\Delta|$ starting from its initially downward $(0,0,-1)$ orientation. Thus $\boldsymbol{\beta}_1$ can be rotated to the $s_1 s_2$ plane only when $\chi/\sqrt{2}\geq|\Delta|$. It should be noted that no matter how large $|\Delta|$ is, one can always get CPD by applying strong enough laser fields; i.e., there is no upper limit for $|\Delta|$ beyond which CPD in state $|2\rangle$ does not occur for the rectangular pulse with constant frequency. This feature can be explained by the discussion of the effective pulse area A.

For a two-level system initially in the lower level and excited at $t=0$ by a stepwise external field with Rabi frequency $\chi/\sqrt{2}$ and detuning Δ, the u, v, and w components of the Bloch vector are

$$u=2\,\mathrm{Re}\left\{-i\frac{\chi}{\sqrt{2}\sigma}\sin\left[\frac{\sigma t}{2}\right]\left[\cos\left[\frac{\sigma t}{2}\right]+i\frac{\Delta}{\sigma}\sin\left[\frac{\sigma t}{2}\right]\right]\right\}, \quad (3.10a)$$

$$v=2\,\mathrm{Im}\left\{-i\frac{\chi}{\sqrt{2}\sigma}\sin\left[\frac{\sigma t}{2}\right]\left[\cos\left[\frac{\sigma t}{2}\right]+i\frac{\Delta}{\sigma}\sin\left[\frac{\sigma t}{2}\right]\right]\right\}, \quad (3.10b)$$

$$w=-\frac{\Delta^2}{\sigma^2}-\frac{\chi^2}{2\sigma^2}\cos(\sigma t)\ . \quad (3.10c)$$

Applying this result to the three-level problem under discussion and using the correspondence relation $u\to-\sqrt{2}\,\mathrm{Im}c_1$, $v\to\sqrt{2}\,\mathrm{Re}c_1$, and $w\to-\mathrm{Im}c_2$, we get

$$c_1=c_3^*=-\frac{\chi}{\sigma}\sin\left[\frac{\sigma t}{2}\right]\left[\cos\left[\frac{\sigma t}{2}\right]+i\frac{\Delta}{\sigma}\sin\left[\frac{\sigma t}{2}\right]\right], \quad (3.11a)$$

$$c_2=i\left[\frac{\Delta^2}{\sigma^2}+\frac{\chi^2}{2\sigma^2}\cos(\sigma t)\right]. \quad (3.11b)$$

Equation (3.11b) indicates that CPD ($c_2=0$) can be achieved only when $\chi/\sqrt{2}\geq|\Delta|$, which confirms the same conclusion made previously according to the vector model. Using Eqs. (3.5) and (3.11a), one obtains the reduced Rabi frequency

$$\sqrt{2}\chi\cos\psi=\frac{\sqrt{2}\chi\cos(\frac{1}{2}\sigma t)}{[\cos^2(\frac{1}{2}\sigma t)+(\Delta/\sigma)^2\sin^2(\frac{1}{2}\sigma t)]^{1/2}}\ , \quad (3.12)$$

which oscillates with period $4\pi/\sigma$ (see Fig. 4). Consequently, the effective pulse area is obtained by substituting Eq. (3.12) into Eq. (3.7),

$$A(t)=4\arcsin\left[\frac{\chi}{\sqrt{2}\sigma}\sin\left[\frac{\sigma t}{2}\right]\right]. \quad (3.13)$$

The maximum value of $|A|$ is $4\arcsin(\chi/\sqrt{2}\sigma)$, which is just the area OPQ of Fig. 4, and can be made to exceed π for any given Δ, no matter how large, by increasing χ. This explains our previous conclusion which is based on the vector model. In addition, if $|A(t)|=\pi$, then $c_2(t)=0$, in agreement with Eq. (3.8a).

D. Smooth pulses

Bambini and Berman[14] have found analytic solutions to the two-state problem for a class of pulse functions with constant detuning. This class of pulses extended from $t=-\infty$ to $t=+\infty$ and lead to an excitation probability P_2 at $t=+\infty$, which can vary from zero to a maximum value

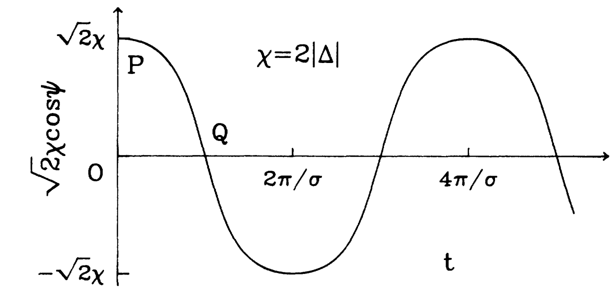

FIG. 4. Reduced Rabi frequency in an equivalent resonant two-state problem when the three-level system with equal detunings and equal Rabi frequencies is prepared initially in state $|2\rangle$ and excited by stepwise is external fields of Rabi frequency χ. The reduced Rabi frequency $\sqrt{2}\chi\cos\psi(t)$ has a period $4\pi/\sigma$, and the pulse area for the first half-period cancels exactly, as does that for second half-period, the third half-period, etc. The figure is plotted for $\chi=2|\Delta|$.

$$P_{2\max}=\mathrm{sech}(\pi\alpha)\mathrm{sech}(\pi\alpha+\pi\alpha\lambda)\cosh^2(\tfrac{1}{2}\pi\alpha\lambda) \quad (3.14)$$

depending on the pulse area. [The quantity $\alpha=\Delta T$ is the detuning parameter, T a time-scale parameter of the pulses, and λ parameter characterizing the shape of pulses $(-1<\lambda<\infty)$.] (The case $\lambda=0$ is the hyperbolic secant pulse, for which $P_{2\max}$ is reached whenever the pulse area is an odd multiple of π.) The pulse areas of these pulses are proportional to their maximum Rabi frequencies.

Applying such pulses to the three-level system with equal detunings and equal Rabi frequencies, one may determine that, for any given parameters α and λ, there is an infinite number of pulse areas which cause CPD for state $|2\rangle$, if $P_{2\max}\geq\frac{1}{2}$. No such pulses exist if $P_{2\max}<\frac{1}{2}$. From Eq. (3.14), one easily sees that the condition $P_{2\max}\geq\frac{1}{2}$ is satisfied provided

$$\cosh^2(\tfrac{1}{2}\pi\alpha\lambda)\geq\sinh^2[\pi\alpha(1+\tfrac{1}{2}\lambda)]\ . \quad (3.15)$$

This shows that there is an upper limit about $|\alpha|=|\Delta|T$ for any given λ, such that when $|\Delta|T$ exceeds this limit, $P_{2\max}$ cannot be larger than one-half. We thus conclude that a necessary condition for CPD in state $|2\rangle$ with these pulses with fixed λ is

$$|\alpha|=|\Delta|T\leq 2y(\lambda)/\pi\ , \quad (3.16a)$$

where

$$\cosh(\lambda y)=\sinh[(\lambda+2)y]\ . \quad (3.16b)$$

For a hyperbolic secant pulse ($\lambda=0$) $\chi(t)\propto\mathrm{sech}(t/2T)$, Eqs. (3.16) become

$$|\alpha|=|\Delta|T\leq\pi^{-1}\ln(\sqrt{2}+1)\approx 0.28\ . \quad (3.17)$$

For given α and λ, one cannot get CPD by increasing the maximum Rabi frequency when $|\alpha|$ exceeds $2y(\lambda)/\pi$. This feature of these smooth pulses is different from that of the rectangular pulse discussed previously.

For the excitation of the three-level system by hyperbolic secant pulses of area $(2m-1)\eta\pi$ $(\eta=\sqrt{2})$, for which $P_2=P_{2\max}=\mathrm{sech}^2(\pi\alpha)$ in the corresponding two-state problem $(\eta=1)$, we plot the reduced Rabi frequency $\sqrt{2}\chi(t)\cos\psi(t)$ and effective area $A(t)$ in Fig. 5. The reduced Rabi frequency oscillates between positive and negative values and, consequently, the effective pulse area is not large even if the input pulse area is very large. The total effective area $A(+\infty)$ reaches its maximum value $2\arccos(1-2P_{2\max})$ for input pulse area $\sqrt{2}(2m-1)\pi$, which can be obtained from Eq. (3.8a) and the correspondence relation. Consequently, for $|\Delta|T=0.2<0.28$ [Eq. (3.17)], $A(+\infty)>\pi$ with an input pulse area $\sqrt{2}(2m-1)\pi$, so there exists an infinite number of input pulse areas which lead to $A(+\infty)=\pi$. On the other hand, for $|\Delta|T=0.3>0.28$, $A(+\infty)<\pi$ and it is impossible to get $A(+\infty)=\pi$ for any input pulse area. Note $A(+\infty)\neq\pi$ for most hyperbolic secant pulses, however, $A(t)$ might pass the value π (i.e., CPD) during the pulse process (see Fig. 5). This suggests another way of obtaining CPD: Apply truncated hyperbolic secant pulses.

The effective pulse area A can also explain CPR in state $|2\rangle$ caused by hyperbolic secant pulses of area $2\sqrt{2}m\pi$ and constant detuning, a result stated in Sec. II B. We show in the Appendix that $\sqrt{2}\chi\cos\psi$ is an odd func-

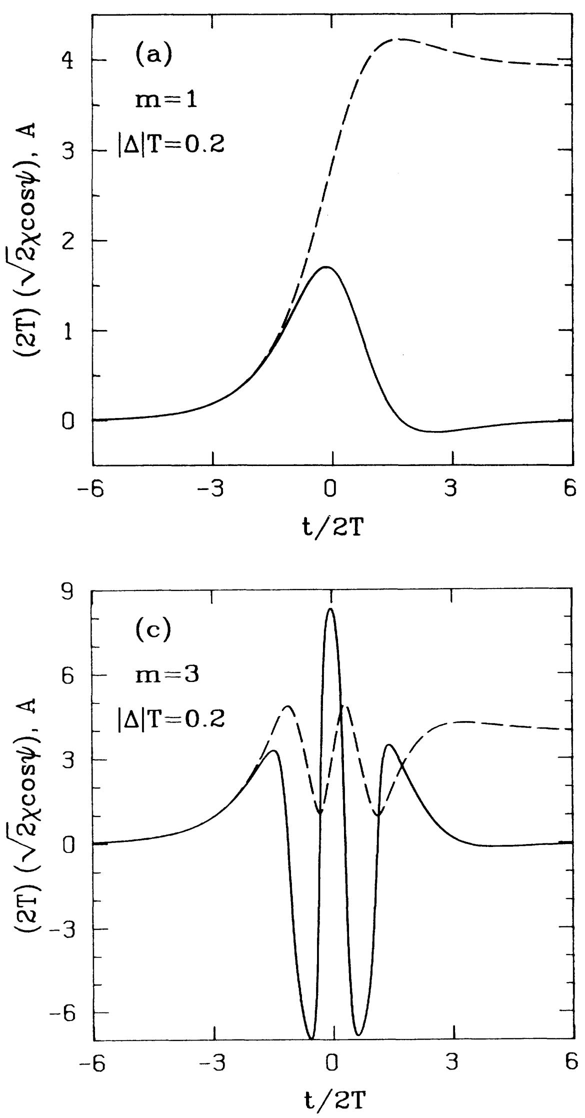

FIG. 5. Reduced Rabi frequency $\sqrt{2}\chi(t)\cos\psi(t)$ (solid line) and effective pulse area $A(t)$ (dashed line) when the three-level system prepared initially in state $|2\rangle$ is excited by the hyperbolic secant pulses of area $\sqrt{2}(2m-1)\pi$ for (a) $|\Delta|T=0.2$, $m=1$; (b) $|\Delta|T=0.3$, $m=1$; (c) $|\Delta|T=0.2$, $m=3$; and (d) $|\Delta|T=0.3$, $m=3$. The reduced Rabi frequency is plotted in units of $(2T)^{-1}$ while the pulse area is dimensionless. Total effective area $A(+\infty)$ is independent of m and is a function of $|\Delta|T$ only.

tion of t for $m=1$ and $m=2$. Generally, it is easy to show that $w(t)$ in the two-level problem is an even function of t for any m. One can further determine, through the two-level Bloch equation, that u and v components are even and odd functions of t, respectively. Correspondingly, in the three-level system, $\cos\psi(t)$ and $\sqrt{2}\chi\cos\psi$ are odd functions of t. According to Eq. (3.7), $A(t)$ is an even functions of t and $A(+\infty)$ vanishes. This, together with Eq. (3.8a), explains why the three-level system finally returns to its initial value (CPR) for the hyperbolic secant pulses of area $2\sqrt{2}m\pi$.

IV. COMPLETE POPULATION DEPLETION IN AN N-LEVEL SYSTEM

We consider N-level systems with energy levels $1,2,\ldots,N$ driven by $N-1$ laser fields such that each neighboring pair of levels l and $l+1$ is nearly resonant with one laser field; i.e., they form a chain type coupled N-level system. Cook and Shore[5] have studied the system for the case that the $N-1$ laser fields are equally detuned from the respective transitions

$$\Delta_{21}=\Delta_{32}=\cdots=\Delta_{N,N-1}\equiv\Delta=\text{const} , \tag{4.1}$$

and the corresponding Rabi frequencies have the relation

$$\chi_l=\sqrt{l(N-l)/2}\chi, \quad l=1,2,\ldots,N-1 \tag{4.2}$$

where χ is constant in time. Note that $N=3$ is a special case of the three-level system with equal detunings and equal Rabi frequencies and has been studied in Sec. III C. By comparing with a spin-J system ($N=2J+1$) in a constant magnetic field, Cook and Shore gave explicit general solutions for the probability amplitudes $c_l(t)$ in the field-interaction representation for constants Δ and χ. (χ can be time dependent when $\Delta=0$.) In addition, they presented a vector model for such an N-level system. They showed that CPI and CPR are possible for resonant excitation $\Delta=0$, but only between levels l and $N-l+1$. Using their result, we point out that CPD can occur in an odd N-level system in the case of constant nonzero detunings.

We consider, in particular, the initial condition that all population is in the middle level (level $J+1$) of the N-level system ($N=2J+1$), i.e.,

$$c_l(0)=\delta_{l,J+1} . \tag{4.3}$$

Using the solution for constant detunings and Rabi frequencies given by Cook and Shore,[5] one obtains

$$\begin{aligned} c_{J+1}(t)&=\sum_{M=-J}^{J} d^J_{M0}(\theta)[d^J_{M0}(\theta)]^* e^{-iM\sigma t} \\ &=\frac{4\pi}{2J+1}\sum_{M=-J}^{J} |Y_{JM}(\theta,0)|^2 e^{-iM\sigma t} \\ &=P_j(\cos^2\theta+\cos(\sigma t)\sin^2\theta) , \end{aligned} \tag{4.4}$$

where P_J is the Legendre polynomial of order J, σ is given in Eq. (3.9), and

$$\theta=\arctan|\chi/(\sqrt{2}\Delta)| . \tag{4.5}$$

Setting $J=1$ in Eq. (4.4) leads to a result similar to that in Eq. (3.11b). Since the argument of the Legendre polynomials $y(t)\equiv\cos^2\theta+\cos(\sigma t)\sin^2\theta$ is a periodic function of time t with period $\tau=2\pi/\sigma$, $c_{J+1}(t)$ is likewise a periodic function with the same τ. One may note that y varies from its initial value $y(0)=1$ to its minimum value $\cos(2\theta)$ and then goes back to $y=1$. Thus when there are m nodes of P_J, $y_J^{(i)}$ ($i=1,2,\ldots,m\le J$), lying between $\cos(2\theta)$ and 1, there exist m t_J within each half-period π/σ such that

$$\begin{aligned} &\cos^2\theta+\cos(\sigma t_J)\sin^2\theta=y_J^{(i)} , \\ &c_{J+1}(t_J)=P_J(y_J^{(i)})=0, \quad i=1,2,\ldots,m . \end{aligned} \tag{4.6}$$

Consequently, a condition for CPD to be possible is that

$$\cos(2\theta)=\frac{\Delta^2-\frac{1}{2}\chi^2}{\Delta^2+\frac{1}{2}\chi^2}\le y_J^{(1)} . \tag{4.7}$$

For any given Δ and J, one can always force the inequality (4.7) to hold by increasing χ, similar to the three-level system discussed in Sec. III C. From the ordering of $y_J^{(1)}$,

$$y_1^{(1)}=0<y_2^{(1)}<\cdots<y_J^{(1)}<y_{J+1}^{(1)}<\cdots , \tag{4.8a}$$

$$y_J^{(1)}\to 1 \quad \text{for } J\to\infty , \tag{4.8b}$$

one knows that for any given Δ and χ, there are an infinite number of odd N-level systems in which one can generate CPD. Also, it is easier to make CPD for larger than for smaller J, since smaller χ is required.

In terms of the vector model of Cook and Shore, initial condition (4.3) corresponds to a flat disc of radius $[J(J+1)]^{1/2}=\frac{1}{2}(N^2-1)^{1/2}$ and the stepwise excitation of the system corresponds to the precession of the disc about a fixed driving vector with angular frequency σ and cone angle 2θ. When Eqs. (4.6) hold, there exist $2m$ orientations of the disc which correspond to $c_{J+1}=0$.

For the initial condition (4.3), we can show by means of a perturbation expansion that $c_l(t)=(-1)^{J-1+l}c^*_{N+1-l}(t)$ for all $l\le J+1$. Thus, we can transform the original $N=2J+1$ equations to another set of $2J+1$ in which $J+1$ of the equations describe motion of a ($J+1$)-level system with all resonant excitation and oscillating Rabi frequencies modulated by J phases of c_l ($l\le J$). [These are true as long as the relations $\Delta_{l+1,l}(t)=\Delta_{N-l+1,N-l}(t)$ and $\chi_l(t)=\chi_{N-l}(t)$ are satisfied.] Consequently, CPD with constant detunings in Cook-Shore N-level systems can be understood in terms of such resonant excitation. In the more general case in which $\Delta_{l+1,l}(t)=\Delta_{N-l+1,N-l}(t)$ and $\chi_l(t)=\chi_{N-l}(t)$, that $c_{J+1}=0$ with $c_l(0)=\delta_{l,J+1}$ (i.e., CPD) may also be possible since the transformation of the equations of motion to a resonant excitation also holds in this case.

V. DISCUSSION

For a three-level system in which the frequencies of two dipole-allowed transitions are very close, one can use a *single* laser field to excite the two transitions. In the case of one laser field[4] with frequency Ω and polarization $\hat{\mathcal{E}}$ coupling level $|2\rangle$, for example, to two Zeeman levels of a P_1 state (see Fig. 2), a three-level system with equal detunings and equal Rabi frequencies is obtained when the conditions $\Omega=\frac{1}{2}(\omega_{32}+\omega_{12})$ and $\mathbf{p}_{21}\cdot\hat{\mathcal{E}}=\mathbf{p}_{32}\cdot\hat{\mathcal{E}}$ are satisfied. In this case, the frequency and phase of the laser field must be constants. Those results obtained in Secs. II and III with restrictions $\Delta=$const applies to this quantum system. In particular, CPD in state $|2\rangle$ with constant detuning can occur in it. For an equally spaced upward cascade three-level system excited by one nearly resonant laser with polarization $\hat{\mathcal{E}}$, it becomes a three-level system with equal detunings and equal Rabi frequencies if $\mathbf{p}_{21}\cdot\hat{\mathcal{E}}=\mathbf{p}_{32}\cdot\hat{\mathcal{E}}$. In this case, the atom-field detunings may be time dependent. All conclusions obtained in Secs. II and III apply to this system.

It is worthwhile to note that neither equal detunings $\Delta_{21}(t)=\Delta_{32}(t)$ nor equal Rabi frequencies $\chi_1(t)=\chi_2(t)$ are a necessary condition for obtaining CPD in a three-level atom coupled to one or two laser fields. In fact, it is easy to show that, for stepwise excitation with constant detunings, CPD in state $|2\rangle$ can be obtained under conditions $\Delta_{32}=\xi\Delta_{21}$ and $\chi_2=\sqrt{\xi}\chi_1$ ($\xi>0$) for any ξ if the inequalities $4\xi/(\xi+1)\le(\chi_1/\Delta_{21})^2\le 4\xi(\xi+1)/(\xi-1)^2$ are satisfied.

In summary, we have studied the dynamics of a three-level atom interacting with radiation fields for equal atom-field detunings $\Delta(t)$ and equal Rabi frequencies $\chi(t)$ and have constructed a vector model [Eqs. (2.11)] for the probability amplitudes. The vector model enables us to make a connection with the corresponding two-level problem for which many results have been obtained previously. In this manner we found analytic solutions for the three-level problem for those coupling pulses for which closed-form solutions to the two-level problem are known. Through the vector model, we have shown that one can obtain CPD for the common level $|2\rangle$ of the two transitions even in the case of constant nonzero detunings. To explain this somewhat surprising result, we recast the equations of motion (2.11) into Eqs. (3.6), which correspond to the equations of motion of a resonant coupled two-level system. The realization of CPD is interpreted in terms of an effective pulse area A of the equivalent two-level equations. Finally, we have shown that a three-level system is not the only quantum system in which CPD with constant detunings can occur. It also can be achieved for the middle level of an odd N-level system, provided that all detunings are equal and a certain relation is satisfied among $(N-1)$ Rabi frequencies.

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APPENDIX: SIMPLE ANALYTIC SOLUTIONS FOR HYPERBOLIC SECANT PULSES

For a two-level atom initially in the ground state excited by a hyperbolic secant pulse which leads to CPR (area $2m\pi$ and constant detuning $\Delta=\alpha/T$) or to CPD (area $[(2m-1)^2+(2T\delta_0)^2]^{1/2}\pi$ with frequency modulation, Eqs. (2.16)), the Bloch vector can be expressed in terms of elementary functions for all t. For instance, in the constant detuning case, for $m=2$,

$$u=\frac{8\alpha\,\mathrm{sech}(t/2T)}{1+4\alpha^2}\left[1-\frac{6\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right], \tag{A1a}$$

$$v=\frac{4\,\mathrm{sech}(t/2T)\tanh(t/2T)}{1+4\alpha^2}\left[1-\frac{18\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right], \tag{A1b}$$

$$w=\frac{8\,\mathrm{sech}^2(t/2T)}{1+4\alpha^2}\left[1-\frac{9\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right]-1, \tag{A1c}$$

and, in the frequency modulation case [see Eqs. (2.16)], for $m=2$,

$$u=-\frac{2T\delta_0\mathrm{sech}(t/2T)}{[9+(2T\delta_0)^2]^{1/2}}\left[1+\frac{8-4\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right], \tag{A2a}$$

$$v=-\frac{\mathrm{sech}(t/2T)}{[9+(2T\delta_0)^2]^{1/2}}\left[1+\frac{8-12\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right], \tag{A2b}$$

$$w=\tanh\frac{t}{2T}\left[1-\frac{4\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right]. \tag{A2c}$$

Such results for the two-level problem can be taken over to the three-level system with equal detunings and equal Rabi frequencies. For such a three-level system initially prepared in state $|2\rangle$,

$$c_j(-\infty)=i\delta_{j2}, \tag{A3}$$

one can use the correspondence relation $u\to-\sqrt{2}\,\mathrm{Im}c_1$, $v\to\sqrt{2}\,\mathrm{Re}c_1$, and $w\to-\mathrm{Im}c_2$ to get $c_1=c_3^*$ and c_2 [see Eqs. (3.3)].

For hyperbolic secant pulses of Rabi frequency

$$\chi(t)=\frac{\sqrt{2}m}{T}\mathrm{sech}\frac{t}{2T}, \tag{A4}$$

(area $2\sqrt{2}m\pi$), and constant detunings $\Delta=\alpha/T$, one may obtain the amplitudes $c_j(t)$ as $m=1$,

$$c_1=c_3^*=\frac{\sqrt{2}}{1+4\alpha^2}\mathrm{sech}\frac{t}{2T}\left[\tanh\frac{t}{2T}-i2\alpha\right], \tag{A5a}$$

$$c_2=i\left[1-\frac{2}{1+4\alpha^2}\mathrm{sech}^2\frac{t}{2T}\right], \tag{A5b}$$

and for $m=2$,

$$c_1=c_3^*=\frac{2\sqrt{2}\,\mathrm{sech}(t/2T)}{1+4\alpha^2}\left[\left[1-\frac{18\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right]\tanh\frac{t}{2T}-i2\alpha\left[1-\frac{6\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right]\right], \tag{A6a}$$

$$c_2=i\left[1-\frac{8\,\mathrm{sech}^2(t/2T)}{1+4\alpha^2}\left[1-\frac{9\,\mathrm{sech}^2(t/2T)}{9+4\alpha^2}\right]\right] . \tag{A6b}$$

The two amplitudes $c_2(t)$ are even functions of t and $c_j(+\infty)=c_j(-\infty)=i\delta_{j2}$. Comparing Eqs. (A5a) and (A6a) with Eq. (3.5), we identify for $m=1$,

$$\cos\psi=\frac{-\tanh(t/2T)}{[4\alpha^2+\tanh^2(t/2T)]^{1/2}} , \tag{A7}$$

and for $m=2$,

$$\cos\psi=\frac{-[9+4\alpha^2-18\,\mathrm{sech}^2(t/2T)]\tanh(t/2T)}{\{[9+4\alpha^2-18\,\mathrm{sech}^2(t/2T)]^2\tanh^2(t/2T)+4\alpha^2[9+4\alpha^2-6\,\mathrm{sech}^2(t/2T)]^2\}^{1/2}} , \tag{A8}$$

both of which are odd functions of t. Consequently, the reduced Rabi frequency $\sqrt{2}\chi\cos\psi$ is an odd function of t for $m=1$ and $m=2$. Substituting Eq. (A7) into Eq. (3.7), one obtains the pulse area for $m=1$

$$A(t)=2\pi-4\,\mathrm{arcsec}[(4\alpha^2+1)^{1/2}\cosh(t/2T)] , \tag{A9}$$

which is an even function of t and vanishes at $t=+\infty$.

For the hyperbolic secant pulses with frequency modulation—Eqs. (2.16)—one may get the amplitudes $c_j(t)$ as $m=1$,

$$c_1=c_3^*=\frac{-1+i2T\delta_0}{\{2[1+(2T\delta_0)^2]\}^{1/2}}\mathrm{sech}\frac{t}{2T} , \tag{A10a}$$

$$c_2=-i\tanh(t/2T) , \tag{A10b}$$

$m=2$,

$$c_1=c_3^*=\frac{-\mathrm{sech}(t/2T)}{\{2[9+(2T\delta_0)^2]\}^{1/2}}\left[\left[1+\frac{8-12\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right]-i2T\delta_0\left[1+\frac{8-4\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right]\right] , \tag{A11a}$$

$$c_2=-i\tanh\frac{t}{2T}\left[1-\frac{4\,\mathrm{sech}^2(t/2T)}{1+(2T\delta_0)^2}\right] . \tag{A11b}$$

Note $c_j(+\infty)=-c_j(-\infty)=-i\delta_{j2}$. The quantities $\cos\psi$ and $A(t)$ for $m=1$ are readily obtained as before,

$$\cos\psi=[1+(2T\delta_0)^2]^{-1/2} , \tag{A12}$$

$$A(t)=\pi+2\arcsin[\tanh(t/2T)] , \tag{A13}$$

which satisfies $A(+\infty)=2\pi$. One sees that CPR in state $|2\rangle$ occurs for all these pulses, which agrees with our statement made in Sec. II B.

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