Analytic solutions for three-state systems with overlapping pulses

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Two classes of analytic solutions for three-state systems involving two overlapping laser pulses of different shapes, or of similar shapes but with their centers displaced, are presented. We find a remarkable connection between the order of arrival of the two overlapping pulses and the effectiveness of transfer from the ground state to the third state, and we find remarkable results for the maximum occupation probability of the intermediate state. The approach of these analytic solutions to the adiabatic-following process is also demonstrated.

I. INTRODUCTION

The problem of efficient transfer of population to thermally unpopulated atomic or molecular levels that are not accessible by one-photon transitions is of crucial importance in many atomic-physics projects.[1–4] Even though a three-state problem involving a two-photon transition of population can be readily solved numerically for any given condition, questions such as finding the optimum conditions for complete population transfer from state 1 to state 3, or for complete return of population from state 1 to state 1, are best analyzed and answered from solving the problem analytically. We have previously derived[5–8] a number of analytic solutions for the three-state systems which are applicable to a variety of pulse shapes. Of particular interest are the cases involving two delayed and overlapping incident laser pulses whose laser frequencies are at resonance with the transition frequencies from state 1 to state 2 and from state 2 to state 3, respectively.

In this paper, we present two additional analytic results for the three-state systems involving two overlapping laser pulses. A remarkable and unexpected conclusion emerges from these analytic results. To achieve an efficient transfer of population from state 1 to state 3 that would not be sensitively dependent on the input parameters such as the laser-pulse shapes and intensities, the atoms or molecules should interact first with the laser pulse for the $2\leftrightarrow3$ transition and then with the laser pulse for the $1\leftrightarrow2$ transition, the pulses being overlapped in time. We will refer to this procedure in which the laser pulses arrive in the counterintuitive order as a counterintuitive procedure.[9] We will show that as the pulse strengths are increased, (i) this procedure minimizes the occupation probability of the intermediate state (state 2) and thus makes the efficiency of population transfer from state 1 to 3 relatively unaffected by radiative or collisional damping of the intermediate state 2, and (ii) this procedure approaches, in the limit of very large pulse strengths, that of adiabatic rapid passage.[10–12]

Depending on the shapes of the pair of laser pulses considered, three different methods have been used by us for getting the analytic solutions for the three-state problem. The three methods will be referred to as (i) the direct method, (ii) the *uvw* method, and (iii) the SU(2) method, respectively, for easy reference. The latter two methods are generally useful because they enable one to make use of a large number of analytic solutions available for the two-state systems for getting the analytic solutions for the three-state systems. They will be described and presented in the Appendixes.

The analytic solutions giving the time-dependent and final probability amplitudes of the three states will be expressed generally in terms of transcendental functions many of whose properties are known and whose asymptotic properties are often expressible in terms of elementary functions. The points to be noted in every case are (i) the order of incidence of the two overlapping pulses, (ii) the final occupation probability of state 3 as a function of increasing pulse strengths, and (iii) the maximum occupation probability of state 2.

II. THREE-STATE SYSTEMS

We consider an atomic or molecular system driven by two laser beams in which the major atomic or molecular transitions take place among only three of the many available states. State 1 is assumed to be the ground state, and states 2 and 3 are labeled in such a way that the electric-dipole transitions between states 1 and 2 and between states 2 and 3 are permitted by the electric-dipole selection rule, and that the transition between states 1 and 3 is forbidden. After a time-dependent unitary transformation is used to remove optical-frequency terms from the Hamiltonian and wave function, the time-dependent Schrödinger equation in units of $\hbar=1$

$$i\frac{\partial\psi}{\partial t}=\hat{H}(t)\psi \tag{2.1}$$

is assumed to be expressible in the form for which the Hamiltonian $\hat{H}(t)$ can be written as

$$\hat{H}(t)=\begin{pmatrix} 0 & -\frac{1}{2}\Omega_1(t) & 0 \\ -\frac{1}{2}\Omega_1(t) & 0 & -\frac{1}{2}\Omega_2(t) \\ 0 & -\frac{1}{2}\Omega_2(t) & 0 \end{pmatrix}, \tag{2.2}$$

where $\Omega_1(t)$ and $\Omega_2(t)$ are generally time-dependent real quantities involving the slowly varying electric-field envelopes of the applied laser beams, and are referred to as the Rabi frequencies corresponding to the $1 \leftrightarrow 2$ and $2 \leftrightarrow 3$ transitions, respectively. Each of $\Omega_1(t)$ and $\Omega_2(t)$ is proportional to the amplitude of the applied oscillating electric field. Zeros appear on the diagonal of $\hat{H}(t)$ because the frequencies of the lasers are assumed to be at resonance with the frequencies of the atomic or molecular transitions. If the Ω's have the same time dependence such that Ω_2/Ω_1 is constant in time, then Eq. (2.1) can be reduced to one with a time-independent Hamiltonian by changing the time scale appropriately, and the solution can be expressed in terms of the eigenvalues and eigenvectors of the resulting time-independent Hamiltonian. We shall not consider such a case in this paper. The cases that are of interest to us and that we shall present in this paper are those for which Ω_2/Ω_1 is not constant in time; either because Ω_1 and Ω_2 are not of the same shape, or if they are of the similar shape, the incidence of one pulse is delayed with respect to the other. We assume that the two pulses overlap in time.

Following our previous paper,[5–8] the Rabi frequencies $\Omega_1(t)$ and $\Omega_2(t)$ of the two laser pulses will be expressed in terms of an arbitrary monotonic function $z(t)$ of the time t. This allows $\Omega_1(t)$ and $\Omega_2(t)$ to have an infinite variety of pulse shapes corresponding to the particular class of functions considered. The analytic solutions for $\psi(t)=(\psi_1(t),\psi_2(t),\psi_3(t))$, the probability amplitudes of states 1, 2, and 3, of Eq. (2.1), for three specific classes of functions representing the Rabi frequencies $\Omega_1(t)$ and $\Omega_2(t)$ will be presented in the following sections.

III. ANALYTIC SOLUTION FOR CLASS 1 PULSES

The first class of laser pulses we shall consider consists of a pair of pulses whose Rabi frequencies are given by

$$\Omega_1(t)=\frac{\alpha_1}{[z(1-z)]^{1/2}}\dot{z}, \quad \Omega_2(t)=\frac{\alpha_2}{z(1-z)^{1/2}}\dot{z}, \tag{3.1}$$

where $z(t)$ is an arbitrary monotonic function of the time t, $\dot{z}$ its time derivative, and α_1 and α_2 are arbitrary positive dimensionless constants. An example of Ω_1 and Ω_2 is given by setting

$$z=\frac{1}{2}\left[1+\tanh\frac{t}{\tau}\right], \tag{3.2}$$

where τ corresponds to an arbitrary pulse length, and where $t=-\infty$ to $+\infty$ corresponds to $z=0$ to 1. It gives

$$\Omega_1(t)=\frac{\alpha_1}{\tau}\operatorname{sech}\frac{t}{\tau}, \quad \Omega_2(t)=\frac{\sqrt{2}\alpha_2}{\tau}\left[1-\tanh\frac{t}{\tau}\right]^{1/2}, \tag{3.3}$$

which are plotted in Fig. 1 as functions of t. The plot emphasizes that the two pulses are applied in the counterintuitive order, since we assume that the entire population is initially in state 1. The pair of pulses given by (3.1) was considered in Ref. 6 and the analytic solution for $\psi(t)$ was obtained by us by identifying the third-order differential equation for $\psi_1(t)$, after eliminating $\psi_2(t)$ and $\psi_3(t)$ from Eq. (2.1), with the known differential equation satisfied by the Clausen function[13] ${}_3F_2(\alpha',\beta',\gamma';\delta',\epsilon';z)$. This is what we called the direct method and a brief description of it is given in Appendix A. The probability amplitudes of the three states at time t are found to be given by

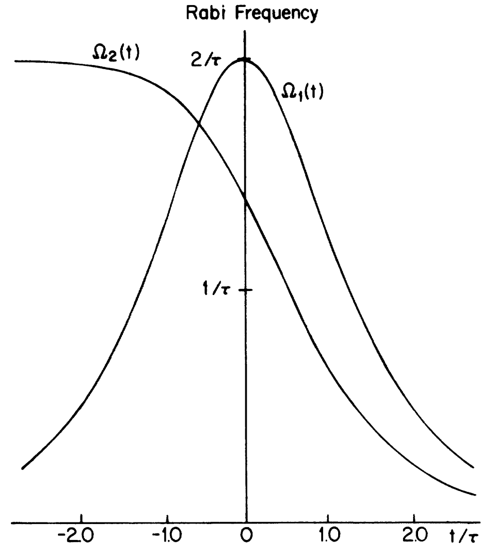

FIG. 1. Optical pulse shapes given by Eq. (3.3).

$$\psi_1(t)={}_3F_2\left[\frac{1}{2},\frac{\alpha_1}{2},-\frac{\alpha_1}{2};\frac{1+i\alpha_2}{2},\frac{1-i\alpha_2}{2};z\right],$$

$$\psi_2(t)=\frac{i\alpha_1}{1+\alpha_2^2}[z(1-z)]^{1/2} \times {}_3F_2\left[\frac{3}{2},1+\frac{\alpha_1}{2},1-\frac{\alpha_1}{2};\frac{3+i\alpha_2}{2},\frac{3-i\alpha_2}{2};z\right],$$

$$\psi_3(t)=\frac{-\alpha_1\alpha_2}{1+\alpha_2^2}z^{1/2} \times {}_3F_2\left[\frac{1}{2},1+\frac{\alpha_1}{2},1-\frac{\alpha_1}{2};\frac{3+i\alpha_2}{2},\frac{3-i\alpha_2}{2};z\right]. \tag{3.4}$$

The Clausen function ${}_3F_2(\alpha',\beta',\gamma';\delta',\epsilon';z)$ has many known properties. In particular, it has a series representation given by

$$1+\frac{\alpha'}{1!}\frac{\beta'}{\delta'}\frac{\gamma'}{\epsilon'}z+\frac{\alpha'(\alpha'+1)}{2!}\frac{\beta'(\beta'+1)}{\delta'(\delta'+1)}\frac{\gamma'(\gamma'+1)}{\epsilon'(\epsilon'+1)}z^2+\cdots \tag{3.5}$$

It is seen that if α_1 is a nonzero even integer, the Clausen series in (3.4) terminate. Complete transfer of the occupation probability can be quite easily obtained, and it is given, for several cases, in Table I, where, for each case, the maximum occupation probability $|\psi_2(t)|^2_{max}$ of state 2 is also given.

TABLE I. Cases of complete transfer of the occupation probability from state 1 to state 3 for class 1 pulses.

α_1	α_2	$\lvert\psi_2(t)\rvert^2_{max}$
2	1	0.250 000
6	1.393 889	0.139 490
10	1.612 837	0.100 605

It is to be noted that as the pulse amplitudes or strengths α_1 and α_2 are increased, the maximum occupation probability of state 2 decreases. In fact, the maximum occupation probability of state 2 is already small even for α_1 and α_2 that are not very large. This is the case even though the laser frequencies are assumed to be at resonance with the $1\leftrightarrow 2$ as well as with the $2\leftrightarrow 3$ transitions. The counterintuitive interaction order has the effect of minimizing the maximum occupation probability of state 2, and increasing the pulse amplitudes when the pulse order is counterintuitive has the effect of further decreasing the maximum occupation probability of state 2. This general feature will be further exemplified in the following sections.

IV. ANALYTIC SOLUTION FOR CLASS 2 PULSES

The second class of laser pulses we shall consider consists of a pair of pulses whose Rabi frequencies are given by

$$\Omega_1(t)=\frac{\alpha_1}{z^{1/2}}\dot{z},\quad \Omega_2(t)=\frac{\alpha_2}{z}\dot{z}\ . \tag{4.1}$$

For $z(t)$ given by Eq. (3.2) for example, Ω_1 and Ω_2 become

$$\Omega_1(t)=\frac{\alpha_1}{\sqrt{2}\tau}\left[1-\tanh\frac{t}{\tau}\right]^{1/2}\operatorname{sech}\frac{t}{\tau}\ ,$$
$$\Omega_2(t)=\frac{\alpha_2}{\tau}\left[1-\tanh\frac{t}{\tau}\right]\ , \tag{4.2}$$

and they are plotted in Fig. 2 as functions of t for the specific values of $\alpha_1=3.554$ and $\alpha_2=1.193$. It should be noted again that the two pulses will interact with the atoms or molecules in the counterintuitive order.

The analytic solution for $\psi(t)$ for this case has been obtained in two ways with the use of the direct method and the *uvw* method which are described in Appendixes A and B. Using the direct method, the third-order differential equation satisfied by ψ_1, after eliminating ψ_2 and ψ_3 from Eq. (2.1), is identified with the known differential equation satisfied by[13] ${}_1F_2(\alpha';\beta',\gamma';z)$. Using the *uvw* method, the solutions for ψ_1, ψ_2, and ψ_3 are expressed in terms of the products of Bessel functions[14] of complex orders, $J_\nu(z)$. We present both forms of solutions in the following:

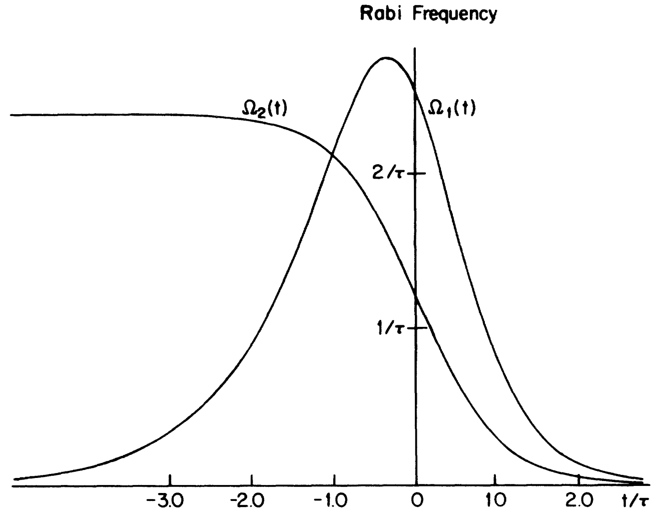

FIG. 2. Optical pulse shapes given by Eq. (4.2) for $\alpha_1=3.554$ and $\alpha_2=1.193$.

$$\psi_1(t)={}_1F_2\left[\frac{1}{2};\frac{1+i\alpha_2}{2},\frac{1-i\alpha_2}{2};\frac{-\alpha_1^2}{4}z\right],$$
$$\psi_2(t)=\frac{i\alpha_1}{1+\alpha_2^2}z^{1/2}\,{}_1F_2\left[\frac{3}{2};\frac{3+i\alpha_2}{2},\frac{3-i\alpha_2}{2};-\frac{\alpha_1^2}{4}z\right],$$
$$\psi_3(t)=\frac{-\alpha_1\alpha_2}{1+\alpha_2^2}z^{1/2}\,{}_1F_2\left[\frac{1}{2};\frac{3+i\alpha_2}{2},\frac{3-i\alpha_2}{2};-\frac{\alpha_1^2}{4}z\right]; \tag{4.3}$$

or

$$\psi_1(t)=|a|^2-|b|^2\ ,$$
$$\psi_2(t)=a^*b-ab^*\ , \tag{4.4}$$
$$\psi_3(t)=-(a^*b+ab^*)\ ,$$

where

$$a=\Gamma\left[\frac{1+i\alpha_2}{2}\right]\left[\frac{\alpha_1 z^{1/2}}{4}\right]^{1/2}J_{\frac{1}{2}(-1+i\alpha_2)}\left[\frac{1}{2}\alpha_1 z^{1/2}\right],$$
$$b=i\Gamma\left[\frac{1+i\alpha_2}{2}\right]\left[\frac{\alpha_1 z^{1/2}}{4}\right]^{1/2}J_{\frac{1}{2}(1+i\alpha_2)}\left[\frac{1}{2}\alpha_1 z^{1/2}\right]. \tag{4.5}$$

Notice that the solution depends on $\alpha_1 z^{1/2}$, not on α_1 and $z(t)$ separately. The functions ${}_1F_2(\alpha';\beta',\gamma';z)$ and $J_\nu(z)$ have series representations given by

$${}_1F_2(\alpha';\beta',\gamma';z)=1+\frac{\alpha'}{1!\beta'\gamma'}z+\frac{\alpha'(\alpha'+1)}{2!\beta'(\beta'+1)\gamma'(\gamma'+1)}z^2+\cdots\ , \tag{4.6}$$

$$J_\nu(z)=\sum_{m=0}^{\infty}(-1)^m(\tfrac{1}{2}z)^{2m+\nu}/[m!\Gamma(m+\nu+1)]\ . \tag{4.7}$$

When $\alpha_1 z^{1/2}$ is large, the asymptotic series

$$a \sim \frac{1}{\pi^{1/2}}\Gamma\left[\frac{1+i\alpha_2}{2}\right] \times\left[\cos\left[\frac{\alpha_1 z^{1/2}}{2}-\frac{i\pi\alpha_2}{4}\right] +\frac{2i\alpha_2+\alpha_2^2}{4\alpha_1 z^{1/2}}\sin\left[\frac{\alpha_1 z^{1/2}}{2}-\frac{i\pi\alpha_2}{4}\right]+\cdots\right]$$

and

$$b \sim \frac{i}{\pi^{1/2}}\Gamma\left[\frac{1+i\alpha_2}{2}\right] \times\left[\sin\left[\frac{\alpha_1 z^{1/2}}{2}-\frac{i\pi\alpha_2}{4}\right] +\frac{2i\alpha_2-\alpha_2^2}{4\alpha_1 z^{1/2}}\cos\left[\frac{\alpha_1 z^{1/2}}{2}-\frac{i\pi\alpha_2}{4}\right]+\cdots\right] \quad (4.8)$$

are useful. Complete transfer of the occupation probability can be obtained, and it is given, for several cases, in Table II, where for each case, the maximum occupation probability, $|\psi_2(t)|^2_{\max}$, of state 2 is also given. For large values of α_1, the asymptotic series give the approximate conditions for complete transfer of population,

$$\alpha_1 \simeq (2n-1)\pi, \quad \alpha_2\sinh(\tfrac{1}{2}\pi\alpha_2)\simeq(2n-1)\pi \ , \quad (4.9)$$

where n is a positive integer. The corresponding estimate of the maximum occupation probability of state 2 is

$$|\psi_2(t)|^2_{\max}\simeq \text{sech}^2(\tfrac{1}{2}\pi\alpha_2) \ . \quad (4.10)$$

It is to be noted again that as the pulse amplitudes α_1 and α_2 are increased, the maximum occupation probability of state 2 decreases, and that the maximum occupation probability is already small even for α_1 and α_2 that are not very large.

The solution given in the form of Eq. (4.3) may be compared with that given in Ref. 7 in terms[13] of ${}_2F_2(\alpha',\beta';\gamma',\delta';z)$ for the case of the same pair of pulses given by (4.1) but for it a nonzero one-photon detuning is assumed, a two-photon resonance condition being assumed to be satisfied. The form of the solution given by Eq. (4.3) also bears some resemblance to that given by Eq. (3.4) for the previous case.

TABLE II. Cases of complete transfer of the occupation probability from state 1 to state 3 for class 2 pulses.

α_1	α_2	$\lvert\psi_2(t)\rvert^2_{\max}$
3.554 272	1.193 434	0.199 771
9.653 358	1.598 851	0.103 957
15.873 647	1.824 197	0.075 379

V. ANALYTIC SOLUTION FOR CLASS 3 PULSES

The third class of laser pulses we shall consider consists of a pair of pulses whose Rabi frequencies are given by

$$\Omega_1(t)=(2\alpha/\beta)[\cos\theta(t)]\dot{\theta} \ , \quad \Omega_2(t)=(2\alpha/\beta)[\sin\theta(t)]\dot{\theta} \ , \quad (5.1)$$

where α and β are arbitrary dimensionless constants and $\theta(t)$ is an arbitrary function of the time. This is a generalization of the case first considered by Gottlieb,[15] Hioe,[16] and Pegg.[17]

The analytic solution for $\boldsymbol{\psi}(t)$ for this case has been obtained with the use of the SU(2) method described in Appendix C. The probability amplitudes $\psi_1(t)$, $\psi_2(t)$, and $\psi_3(t)$ of the three states are given by

$$\psi_1(t)=\tfrac{1}{2}[a^2+(a^*)^2+b^2+(b^*)^2] \ , \quad \psi_2(t)=a^*b-ab^* \ , \quad \psi_3(t)=\tfrac{1}{2}i[a^2-(a^*)^2+b^2-(b^*)^2] \ , \quad (5.2)$$

where

$$a(t)=\left[\cos\frac{r}{\beta}(\theta-\theta_0+\tfrac{1}{2}\beta)+\frac{i\beta}{2r}\sin\frac{r}{\beta}(\theta-\theta_0+\tfrac{1}{2}\beta)\right] \times\exp\left[-\frac{i}{2}(\theta-\theta_0+\tfrac{1}{2}\beta)\right] ,$$

$$b(t)=\frac{i\alpha}{2r}\sin\frac{r}{\beta}(\theta-\theta_0+\tfrac{1}{2}\beta)\exp\left[-\frac{i}{2}(\theta+\theta_0-\tfrac{1}{2}\beta)\right] ,$$

$$r\equiv\tfrac{1}{2}(\alpha^2+\beta^2)^{1/2} \ , \quad (5.3)$$

and θ_0 is an arbitrary constant. We assume that laser pulses are applied to the atom or molecule when t lies in the interval from 0 to T, for which the range of $\theta(t)$ is between $\theta_0-\frac{1}{2}\beta$ and $\theta_0+\frac{1}{2}\beta$. The final values of $a(t)$ and $b(t)$ are

$$a(T)=\left[\cos r+\frac{i\beta}{2r}\sin r\right]\exp(-i\beta/2)$$

and

$$b(T)=\frac{i\alpha}{2r}\sin r\,\exp(-i\theta_0) \ , \quad (5.4)$$

and the final probability amplitudes are

$$\psi_1(T)=\cos^2 r\cos\beta+\frac{\beta}{r}\sin r\cos r\sin\beta -\frac{\beta^2\sin^2 r}{\alpha^2+\beta^2}\cos\beta-\frac{\alpha^2\sin^2 r}{\alpha^2+\beta^2}\cos 2\theta_0 ,$$

$$\psi_2(T)=i\left[\frac{\alpha}{r}\sin r\cos r\cos(\theta_0-\tfrac{1}{2}\beta) -\frac{2\alpha\beta}{\alpha^2+\beta^2}\sin^2 r\sin(\theta_0-\tfrac{1}{2}\beta)\right], \quad (5.5)$$

$$\psi_3(T)=\cos^2 r\sin\beta-\frac{\beta}{r}\sin r\cos r\cos\beta -\frac{\beta^2\sin^2 r}{\alpha^2+\beta^2}\sin\beta-\frac{\alpha^2\sin^2 r}{\alpha^2+\beta^2}\sin 2\theta_0 .$$

A simple example of (5.1) is given by

$$\Omega_1(t)=\frac{2\alpha}{T}\cos\theta(t),\quad \Omega_2(t)=\frac{2\alpha}{T}\sin\theta(t) \quad (5.6)$$

for $0\le t\le T$, and $\Omega_1(t)=\Omega_2(t)=0$ for $t<0$ or $t>T$. Here

$$\theta(t)=\frac{\beta}{T}t+\theta_0-\tfrac{1}{2}\beta . \quad (5.7)$$

A useful feature of our example and solution given in this section is that it includes both the counterintuitive and intuitive interaction sequences as special cases. Let us fix

$$\theta_0=\frac{\pi}{4} , \quad (5.8)$$

then

$$\beta=\begin{cases}\pi/2\\ -\pi/2\end{cases} \quad (5.9)$$

give the intuitive and counterintuitive interaction sequences, respectively, since it is seen from (5.6)–(5.8) that at $t=0$, $\Omega_1(0)=2\alpha/T$ and $\Omega_2(0)=0$ for $\beta=\pi/2$, so that the atoms or molecules interact first with the laser pulse for the $1\leftrightarrow 2$ transition; whereas $\Omega_1(0)=0$ and $\Omega_2(0)=2\alpha/T$ for $\beta=-\pi/2$, so that the atoms or molecules interact first with the laser pulse for the $2\leftrightarrow 3$ transition, while all the population of the atoms or molecules is in state 1 initially. The pulses given by Eq. (5.6) are shown, for the intuitive interaction order, in Fig. 3. For the counterintuitive order, the labels Ω_1 and Ω_2 for the two pulses are simply interchanged. The final occupation probability $|\psi_3(T)|^2$ for state 3 for the intuitive and counterintuitive interaction sequences as a function of increasing amplitude α can now be compared and it is plotted in Fig. 4 as a solid curve and a dotted curve, respectively. It is seen that while $|\psi_3(T)|^2$ oscillates between 0 and 1 for the intuitive interaction order, it approaches, with smaller and smaller oscillations, to the value 1 for the counterintuitive interaction order. This can be clearly seen also from Eq. (5.5) that as $|\alpha|$ becomes large,

$$|\psi_3(T)|^2\rightarrow\begin{cases}(\cos^2 r-\sin^2 r)^2=\cos^2 2r\\ (\cos^2 r+\sin^2 r)^2=1 ,\end{cases} \quad (5.10)$$

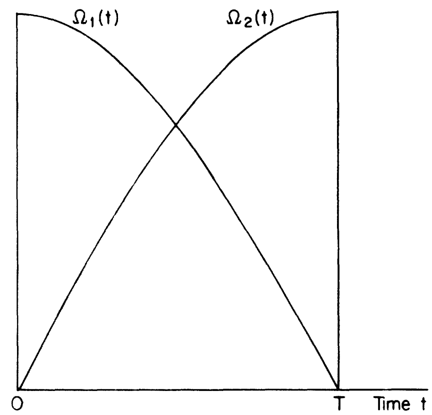

FIG. 3. Optical pulse shapes given by Eqs. (5.6)–(5.9) for $\beta=\pi/2$.

for the intuitive and counterintuitive interaction sequences, respectively. The advantage of using the counterintuitive interaction sequence for getting as much population as possible to state 3 from state 1 is clearly exhibited.

For the intuitive interaction sequence, complete transfer of population from state 1 to state 3 occurs at the values of α in (5.6) given by

$$\alpha=\frac{\pi}{2}(4n^2-1)^{1/2},\quad n=1,2,\ldots ; \quad (5.11)$$

and for the counterintuitive sequence, the corresponding values of α are given by

$$\alpha=\frac{\pi}{2}(16n^2-1)^{1/2},\quad n=1,2,\ldots . \quad (5.12)$$

For the values of α given by Eq. (5.12) which give complete transfer from state 1 to state 3 for both interaction sequences, the maximum occupation probability of state 2 is

$$1-\frac{1}{16n^2},\quad \frac{1}{4n^2}-\frac{1}{16n^4} , \quad (5.13)$$

respectively, for the intuitive and counterintuitive in-

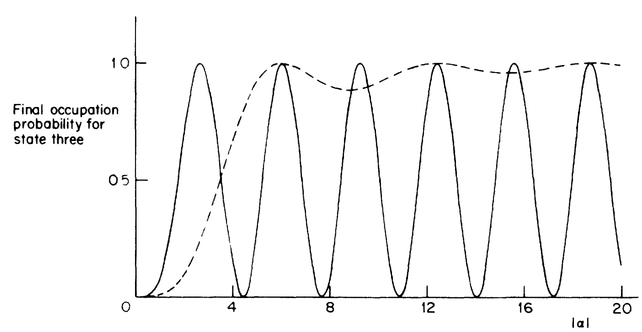

FIG. 4. Final occupation probability of state 3 plotted as a function of dimensionless amplitude for the pulses given by Eqs. (5.6)–(5.9). The solid curve is for $\beta=\pi/2$, and the dotted curve is for $\beta=-\pi/2$.

teraction sequences.[8] Thus it is seen that as the amplitudes for both pulses are increased, while the maximum occupation probability for state 2 approaches one for the intuitive interaction order, it quickly diminishes and approaches zero for the counterintuitive interaction order.

VI. ADIABATIC FOLLOWING

A remarkable common result that emerges from all the preceding analytic solutions is that sending two overlapping laser pulses in the counterintuitive interaction sequence with large pulse amplitudes provides an efficient way for population transfer in a three-state system that is quite independent of the laser-pulse shapes, and that minimizes the occupation probability of the intermediate state 2, making the efficiency of population transfer from state 1 to 3 relatively unaffected by radiative or collisional damping of the intermediate state. In the limit of very large pulse amplitudes, the process described approaches that of adiabatic rapid passage or adiabatic following for which a simple analytic solution was recently given.[12]

Consider a three-state system interacting with a pair of laser pulses of arbitrary shapes shown in Fig. 5 incident in the counterintuitive interaction sequence such that

$$\left.\frac{\Omega_1(t)}{\Omega_2(t)}\right|_{t\to-\infty}\to 0,\quad \left.\frac{\Omega_2(t)}{\Omega_1(t)}\right|_{t\to+\infty}\to 0\ , \tag{6.1}$$

where $t\to-\infty$ and $t\to+\infty$ correspond to times before and after the interaction with the lasers, respectively. If T denotes the pulse length or the interaction time, and Ω_{eff} denotes $\frac{1}{2}(\Omega_1^2+\Omega_2^2)^{1/2}$, it was pointed out[12] that the adiabatic following process can be achieved if

$$\Omega_{\text{eff}}T \gg 1\ , \tag{6.2}$$

that is, for given T, the process can be approached by increasing Ω_{eff}. Equation (6.2) is the dimensionless criterion for adiabatic following.[10] Assuming that (6.1) and (6.2) are satisfied, the probability amplitudes of states 1, 2, and 3 were shown[12] to be approximately given by

$$\begin{bmatrix}\psi_1(t)\\ \psi_2(t)\\ \psi_3(t)\end{bmatrix} \simeq \begin{bmatrix}\cos\phi(t)\\ 0\\ -\sin\phi(t)\end{bmatrix}\ , \tag{6.3}$$

where $\cos\phi(t)$ denotes $\Omega_2(t)[\Omega_1(t)^2+\Omega_2(t)^2]^{-1/2}$ and $\sin\phi(t)$ denotes $\Omega_1(t)[\Omega_1(t)^2+\Omega_2(t)^2]^{-1/2}$, or

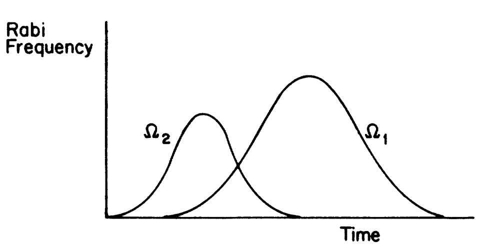

FIG. 5. Two overlapping pulses where pulse 2 precedes pulse 1.

$$\tan\phi(t)=\frac{\Omega_1(t)}{\Omega_2(t)}\ . \tag{6.4}$$

Using Eq. (6.1), it is seen that the adiabatic-following solution (6.3) gives complete transfer of population from state 1 to state 3 without populating state 2 to any significant extent at any time, even though the laser frequencies of the two incident laser pulses may be at resonance with the $1\leftrightarrow 2$ and $2\leftrightarrow 3$ transitions, respectively. Our exact analytic solutions given in the three preceding sections clearly demonstrate how this process is approached in the limit of high laser intensity, where (6.2) is satisfied, for the various specific pulse shapes. Conditions (6.1) and (6.2) can be applied to each pulse shape considered above and to other pulse shapes. This reduction in the population of state 2 by use of the counterintuitive pulse order can be understood physically as the result of mixing of states 2 and 3 by driving the $2\leftrightarrow 3$ transition strongly. The oscillating electric field gives an Autler-Townes[18] splitting of state 2 into two components that differ in energy by the Rabi frequency, and this changes the effect of the resonant pulse that drives the $1\leftrightarrow 2$ transition.

The high efficiency of population transfer using the counterintuitive interaction sequence in a three-level system was experimentally confirmed by Gaubatz *et al.*[19] recently. They performed experiments in which the laser frequencies of the pump and stimulating lasers were tuned to be on resonance with two specific transitions between the electronic states of sodium molecules which were sent to cross the two laser beams at right angles. The centers of the two beams could be shifted relative to each other. The maximum population in level 3 was observed when the two laser beams were partially overlapped in the counterintuitive interaction sequence.

VII. CONCLUSION

We have presented three classes of analytic solutions, (3.4), (4.3) or (4.4), and (5.4), that clearly demonstrate efficient transfer of population from state 1 to state 3 with minimum occupation probability of state 2 using the counterintuitive interaction sequence with high intensity laser pulses. They also give us a better understanding on how the limiting case of the so-called adiabatic following process is approached, for which the approximate solution is given by Eq. (6.3). Our conclusion is supported by the experimental confirmation given by Gaubatz *et al.*

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APPENDIX A: THE DIRECT METHOD

In this appendix, we outline the method and steps that led us to the analytic solutions (3.4) and (4.3) for pulses of the shapes given by (3.1) and (4.1), respectively.

If we eliminate ψ_2 and ψ_3 from the coupled differential equations

$$i\frac{d}{dz}\begin{bmatrix}\psi_1\\ \psi_2\\ \psi_3\end{bmatrix}=\begin{bmatrix}0 & f_1(z) & 0\\ f_1(z) & 0 & f_2(z)\\ 0 & f_2(z) & 0\end{bmatrix}\begin{bmatrix}\psi_1\\ \psi_2\\ \psi_3\end{bmatrix}, \tag{A1}$$

we get the following third-order differential equation for ψ_1:

$$\frac{1}{f_1 f_2}\psi_1''' + \left[\left(\frac{1}{f_2}\right)'\left(\frac{1}{f_1}\right)+\frac{2}{f_2}\left(\frac{1}{f_1}\right)'\right]\psi_1'' + \left[\left(\frac{1}{f_1}\right)'\left(\frac{1}{f_2}\right)'+\frac{1}{f_2}\left(\frac{1}{f_1}\right)''+\frac{f_1}{f_2}+\frac{f_2}{f_1}\right]\psi_1' + \left(\frac{f_1}{f_2}\right)'\psi_1=0 , \tag{A2}$$

where the prime denotes the derivative with respect to z.

Substituting (3.1) into (2.1) and (2.2), or substituting

$$f_1=-\frac{\alpha_1}{2[z(1-z)]^{1/2}},\quad f_2=-\frac{\alpha_2}{2z(1-z)^{1/2}} \tag{A3}$$

into (A2) gives the following differential equation for ψ_1:

$$\frac{d^3\psi_1}{dz^3}+\left[\frac{2}{z}+\frac{3/2}{z-1}\right]\frac{d^2\psi_1}{dz^2} + \left[\frac{1}{4}(1+\alpha_2^2)\frac{1}{z^2}+\frac{1}{4}(5-\alpha_1^2-\alpha_2^2)\frac{1}{z(z-1)}\right]\frac{d\psi_1}{dz} - \frac{\alpha_1^2}{8}\frac{1}{z^2(z-1)}\psi_1=0 . \tag{A4}$$

By comparing Eq. (A4) with the equation satisfied by $F={}_3F_2(\alpha',\beta',\gamma';\delta',\epsilon';z)$,

$$\frac{d^3F}{dz^3}+\frac{(3+\alpha'+\beta'+\gamma')z-(1+\delta'+\epsilon')}{z(z-1)}\frac{d^2F}{dz^2} + \frac{(1+\alpha'+\beta'+\gamma'+\alpha'\beta'+\alpha'\gamma'+\beta'\gamma')z-\delta'\epsilon'}{z^2(z-1)}\frac{dF}{dz} + \frac{\alpha'\beta'\gamma'}{z^2(z-1)}F=0 , \tag{A5}$$

we find ψ_1 given by Eq. (3.4) that satisfies the initial condition. To find ψ_2, we use the first equation of (A1) and write $\psi_2=f_1^{-1}i\,d\psi_1/dz$, and use the differentiation formula for a generalized hypergeometric function

$$\frac{d}{dz}\,{}_pF_q(a_1,\dots,a_p;b_1,\dots,b_q;z) = \frac{a_1\cdots a_p}{b_1\cdots b_q} \times {}_pF_q(a_1+1,\dots,a_p+1;b_1+1,\dots,b_q+1;z) . \tag{A6}$$

To find ψ_3, we use the second equation of (A1) and write $\psi_3=f_2^{-1}(i\,d\psi_2/dz-f_1\psi_1)$ and use the differentiation formulas given by Rainville[20] to simplify the resulting expression. A simpler method is to start from the third-order differential equation for ψ_3 which can be derived from (A2) by simply interchanging f_1 and f_2 and substituting ψ_3 for ψ_1. We then substitute $\psi_3=z^{1/2}y$ and identify the equation satisfied by y with the equation satisfied by ${}_3F_2$. The appropriate constant factor for ψ_3 can be determined from $i\,d\psi_3/dz=f_z\psi_z$.

Similarly, to find the analytic solution (4.3), we first substitute (4.1) into (2.1) and (2.2), or substitute

$$f_1=-\frac{\alpha_1}{2z^{1/2}},\quad f_2=-\frac{\alpha_2}{2z} \tag{A7}$$

into (A2), giving the following differential equation for ψ_1:

$$z^2\psi_1'''+2z\psi_1''+[\tfrac{1}{4}\alpha_1^2 z+\tfrac{1}{4}(\alpha_2^2+1)]\psi_1'+\tfrac{1}{8}\alpha_1^2\psi_1=0 . \tag{A8}$$

By comparing (A8) with the equation satisfied by $F={}_1F_2(\alpha';\beta',\gamma';x)$:

$$x^2\frac{d^3F}{dx^3}+(\beta'+\gamma'+1)x\frac{d^2F}{dx^2}+(\beta'\gamma'-x)\frac{dF}{dx}-\alpha'F=0 , \tag{A9}$$

we find ψ_1 given by Eq. (4.3) that satisfies the initial condition. We then proceed in a similar way to find ψ_2 and ψ_3.

APPENDIX B: THE *uvw* METHOD

In this Appendix, we describe the method that led us to the analytic solution (4.4) and (4.5) for pulses of the shapes given by (4.1). The method is quite general and can be applied to other pulse shapes.

If in Eq. (2.1), we let

$$\psi_1=u,\quad \psi_2=iv,\quad \psi_3=-w , \tag{B1}$$

then the three-state time-dependent Schrödinger equation at resonance is equivalent to the undamped Bloch equation[21] given by

$$\frac{d}{dt}\begin{bmatrix}u\\ v\\ w\end{bmatrix}=\begin{bmatrix}0 & -\frac{1}{2}\Omega_1(t) & 0\\ \frac{1}{2}\Omega_1(t) & 0 & -\frac{1}{2}\Omega_2(t)\\ 0 & \frac{1}{2}\Omega_2(t) & 0\end{bmatrix}\begin{bmatrix}u\\ v\\ w\end{bmatrix} . \tag{B2}$$

Equation (B2) is equivalent to the two-state Schrödinger equations if we set

$$u=|a|^2-|b|^2\ ,$$
$$v=-i(a^*b-ab^*)\ , \qquad \text{(B3)}$$
$$w=a^*b+ab^*\ ,$$

where a and b satisfy

$$i\frac{d}{dt}\begin{bmatrix} a \\ b \end{bmatrix}=\begin{bmatrix} -\frac{1}{4}\Omega_2(t) & -\frac{1}{4}\Omega_1(t) \\ -\frac{1}{4}\Omega_1(t) & \frac{1}{4}\Omega_2(t) \end{bmatrix}\begin{bmatrix} a \\ b \end{bmatrix}\ . \qquad \text{(B4)}$$

Thus from Eq. (B1), ψ_1, ψ_2, and ψ_3 are given in terms of a and b by Eq. (4.4), provided that the solutions a and b from Eq. (B4) are chosen so that they satisfy the initial conditions for ψ_1, ψ_2, and ψ_3.

For Ω_1 and Ω_2 given by (4.1), the solutions of (B4) that satisfy the initial conditions for ψ_1, ψ_2, and ψ_3 can be shown to be given by Eq. (4.5), and hence from Eqs. (B1) and (B3) we arrive at the analytic solution (4.4).

It is interesting to note that by equating Eqs. (4.4) and (4.5) with Eq. (4.3), we have the following three identities between ${}_1F_2$ and the sum of products of the corresponding Bessel functions of complex orders:

$${}_1F_2\left[\frac{1}{2};\frac{1+i\nu}{2},\frac{1-i\nu}{2};-z^2\right]=\frac{-\pi z}{2\cosh(\frac{1}{2}\pi\nu)}[J_{\frac{1}{2}(1+i\nu)}(z)J_{\frac{1}{2}(1-i\nu)}(z)-J_{\frac{1}{2}(-1-i\nu)}(z)J_{\frac{1}{2}(-1+i\nu)}(z)]\ , \qquad \text{(B5)}$$

$${}_1F_2\left[\frac{3}{2};\frac{3+i\nu}{2},\frac{3-i\nu}{2};-z^2\right]=\frac{\pi(1+\nu^2)}{4\cosh(\frac{1}{2}\pi\nu)}[J_{\frac{1}{2}(1+i\nu)}(z)J_{\frac{1}{2}(-1-i\nu)}(z)+J_{\frac{1}{2}(1-i\nu)}(z)J_{\frac{1}{2}(-1+i\nu)}(z)]\ , \qquad \text{(B6)}$$

and

$${}_1F_2\left[\frac{1}{2};\frac{3+i\nu}{2},\frac{3-i\nu}{2};-z^2\right]=\frac{i\pi(1+\nu^2)}{4\nu\cosh(\frac{1}{2}\pi\nu)}[J_{\frac{1}{2}(1+i\nu)}(z)J_{\frac{1}{2}(-1-i\nu)}(z)-J_{\frac{1}{2}(1-i\nu)}(z)J_{\frac{1}{2}(-1+i\nu)}(z)]\ . \qquad \text{(B7)}$$

The identities (B6) and (B7) can be independently shown to hold by using the following known identity:

$${}_1F_2(\tfrac{1}{2};\nu+1,-\nu+1;-z^2)=\frac{\pi\nu}{\sin(\pi\nu)}J_\nu(z)J_{-\nu}(z)\ . \qquad \text{(B8)}$$

Identity (B5) requires a lengthier independent proof. We may first use the following known identity:[14]

$${}_2F_3(\tfrac{1}{2}+\tfrac{1}{2}\mu+\tfrac{1}{2}\nu,1+\tfrac{1}{2}\mu+\tfrac{1}{2}\nu;1+\mu,1+\nu,1+\mu+\nu;-z^2)=(\tfrac{1}{2}z)^{-\mu-\nu}\Gamma(\mu+1)\Gamma(\nu+1)J_\mu(z)J_\nu(z)\ , \qquad \text{(B9)}$$

for the first product of Bessel functions in (B5). For the second product of Bessel functions in (B5), on the other hand, since the orders of the Bessel functions add up to -1, we need to first make use of a recurrence relation for the Bessel functions to express one of the Bessel functions in terms of Bessel functions of the contiguous orders, before using the identity (B9). We then make use of the contiguous function relations for the generalized hypergeometric functions given by Rainville[20] that simplify the results to the simple identity (B5).

Three classes of analytic solutions for the two-state Schrödinger equation were given previously by the authors.[22,23] Some of these solutions can be used through Eqs. (B1)–(B3) for obtaining the corresponding solutions for the three-state Schrödinger equation (2.1) at resonance.

In Ref. 22, two classes of analytic solutions are given for the two-state Schrödinger equation in the form

$$i\frac{d}{dt}\begin{bmatrix} a_1 \\ a_2 \end{bmatrix}=\begin{bmatrix} 0 & -\frac{1}{2}\Omega(t)e^{-iB} \\ -\frac{1}{2}\Omega(t)e^{iB} & 0 \end{bmatrix}\begin{bmatrix} a_1 \\ a_2 \end{bmatrix}\ . \qquad \text{(B10)}$$

Let us restrict ourselves to the cases in which B is bounded. Then we need to set $\gamma=0$ for both classes of solutions given in Ref. 22. Thus we have

$$\Omega(t)=\begin{cases} \dfrac{\alpha}{\pi[z(1-z)]^{1/2}}\dot{z} & \text{for class 1} \\ \dfrac{\alpha}{\pi(z^2+1)}\dot{z} & \text{for class 2}\ , \end{cases} \qquad \text{(B11)}$$

$$B(t)=\begin{cases} -\dfrac{\beta}{\pi}\ln(1-z) & \text{for class 1} \\ \dfrac{\beta}{\pi}\tan^{-1}z & \text{for class 2}\ , \end{cases} \qquad \text{(B12)}$$

where $z(t)$ can be suitably chosen so that the time interval may correspond to $z=0$ to $\frac{1}{2}$ for class 1 or $z=-\infty$ to $+\infty$ for class 2.

However, class 2 can be simplified by using $\tan^{-1}z$ as a new independent variable, and this leads to (5.3). Then we may relate the solutions and the parameters in Eq. (B4) to those in Eq. (B10) by

$$\begin{aligned} \tfrac{1}{2}\Omega_1(t) &= \Omega(t) , \\ \tfrac{1}{2}\Omega_2(t) &= \frac{dB}{dz}\frac{dz}{dt} , \\ a &= a_1 \exp\left[\tfrac{1}{4} i \int^t \Omega_2 dt\right] , \\ b &= a_2 \exp\left[-\tfrac{1}{4} i \int^t \Omega_2 dt\right] , \end{aligned} \tag{B13}$$

where a_1 and a_2 are the solutions for (B10) given in Ref. 22 (with γ set equal to 0). The solutions of the corresponding three-state problem, (2.1) and (2.2), are then given by Eqs. (B1) and (B3).

APPENDIX C: THE SU(2) METHOD

In this Appendix, we describe the method that led us to the analytic solution (5.4) for pulses of the shapes given by (5.1). The method was given in Ref. 6 but unfortunately it contained some errors. Since it is quite a general and useful method, we shall outline it here.

The first crucial point is the observation that the Hamiltonian $\hat{H}(t)$ given by Eq. (2.2) can be written as

$$\hat{H}(t) = -\tfrac{1}{2}\Omega_1(t)\hat{J}_1 - \tfrac{1}{2}\Omega_2(t)\hat{J}_2 - (0)\hat{J}_3 , \tag{C1}$$

where

$$\hat{J}_1 = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} , \quad \hat{J}_2 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix} , \quad \hat{J}_3 = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix} , \tag{C2}$$

satisfy the commutation relations

$$[\hat{J}_1, \hat{J}_2] = i\hat{J}_3, \quad [\hat{J}_2, \hat{J}_3] = i\hat{J}_1, \quad [\hat{J}_3, \hat{J}_1] = i\hat{J}_2 . \tag{C3}$$

When the generally time-dependent Hamiltonian $\hat{H}(t)$ can be expressed in terms of the generators of the SU(2) algebra, it is said to belong to the SU(2) model,[24] and the solutions of the time-dependent Schrödinger equation were shown to be expressible in terms of sums of products of the solutions of the corresponding two-state problem. In order to make use of the "standard" representation of the SU(2) group, we transform $\hat{J}_1$, $\hat{J}_2$, and $\hat{J}_3$ given in (C2) into the "standard" representation $\hat{J}_1{}'$, $\hat{J}_2{}'$, and $\hat{J}_3{}'$ given by

$$\hat{J}_1{}' = \frac{1}{\sqrt{2}}\begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix} , \quad \hat{J}_2{}' = \frac{1}{\sqrt{2}}\begin{bmatrix} 0 & -i & 0 \\ i & 0 & -i \\ 0 & i & 0 \end{bmatrix} , \quad \hat{J}_3{}' = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix} , \tag{C4}$$

through the unitary matrix $\hat{U}$ according to

$$\hat{J}_k{}' = \hat{U}^\dagger \hat{J}_k \hat{U}, \quad k = 1,2,3 \tag{C5}$$

where

$$\hat{U} = \begin{bmatrix} 1/\sqrt{2} & 0 & 1/\sqrt{2} \\ 0 & 1 & 0 \\ i/\sqrt{2} & 0 & -i/\sqrt{2} \end{bmatrix} , \quad \hat{U}^\dagger = \begin{bmatrix} 1/\sqrt{2} & 0 & -i/\sqrt{2} \\ 0 & 1 & 0 \\ 1/\sqrt{2} & 0 & i/\sqrt{2} \end{bmatrix} . \tag{C6}$$

We define

$$\hat{H}'(t) = \hat{U}^\dagger \hat{H}(t) \hat{U} , \tag{C7}$$

and

$$\psi' = \hat{U}^\dagger \psi , \tag{C8}$$

then Eq. (2.1) can be written as

$$i\frac{\partial \psi'}{\partial t} = \hat{H}'(t)\psi' , \tag{C9}$$

where the transformed Hamiltonian

$$\hat{H}'(t) = -\tfrac{1}{2}\Omega_1(t)\hat{J}_1{}' - \tfrac{1}{2}\Omega_2(t)\hat{J}_2{}' - (0)\hat{J}_3' \tag{C10}$$

is now expressed in terms of the generators of the SU(2) algebra in the standard form.

The corresponding Hamiltonian for the two-state problem when it is expressed in terms of the generators of the SU(2) algebra in the standard form is

$$\begin{aligned} \hat{\mathcal{H}}'(t) &= -\tfrac{1}{2}\Omega_1(t)\hat{\sigma}_1 - \tfrac{1}{2}\Omega_2(t)\hat{\sigma}_2 - (0)\hat{\sigma}_3 \\ &= \begin{bmatrix} 0 & -\tfrac{1}{4}(\Omega_1 - i\Omega_2) \\ -\tfrac{1}{4}(\Omega_1 + i\Omega_2) & 0 \end{bmatrix} , \end{aligned} \tag{C11}$$

where

$$\hat{\sigma}_1 = \tfrac{1}{2}\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} , \quad \hat{\sigma}_2 = \tfrac{1}{2}\begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} , \quad \hat{\sigma}_3 = \tfrac{1}{2}\begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} . \tag{C12}$$

Let the solutions ϕ' of the two-state problem

$$i\frac{\partial \phi'}{\partial t} = \hat{\mathcal{H}}'(t)\phi' \tag{C13}$$

be given by

$$\begin{bmatrix}\phi_1'\\ \phi_2'\end{bmatrix}=\begin{bmatrix}a & b\\ -b^* & a^*\end{bmatrix}\begin{bmatrix}\phi_1'(0)\\ \phi_2'(0)\end{bmatrix}, \tag{C14}$$

i.e., $a(t)$ and $-b^*(t)$ denote the solutions for ϕ_1' and ϕ_2', the two components of $\boldsymbol{\phi}'$. Then according to the result obtained for the SU(2) model, the solution of Eq. (C9) for our three-state problem can be expressed in terms of $a(t)$ and $b(t)$ obtained from Eq. (C13) as

$$\boldsymbol{\psi}'(t)=\hat{D}^{(1)}(a,b)\boldsymbol{\psi}'(0), \tag{C15}$$

or more explicitly,

$$\begin{bmatrix}\psi_1'\\ \psi_2'\\ \psi_3'\end{bmatrix}=\begin{bmatrix}a^2 & \sqrt{2}ab & b^2\\ -\sqrt{2}ab^* & |a|^2-|b|^2 & \sqrt{2}a^*b\\ (b^*)^2 & -\sqrt{2}a^*b^* & (a^*)^2\end{bmatrix}\times\begin{bmatrix}\psi_1'(0)\\ \psi_2'(0)\\ \psi_3'(0)\end{bmatrix}, \tag{C16}$$

where $D^{(1)}(a,b)$, shown explicitly in (C16), is the three-dimensional representation of the SU(2) group.[25] Using Eq. (C8), we finally obtain the solution $\boldsymbol{\psi}$ in terms of its initial value $\boldsymbol{\psi}(0)$ and in terms of the solutions $a(t)$ and $b(t)$ of the corresponding two-state problem as

$$\boldsymbol{\psi}(t)=\hat{U}\hat{D}^{(1)}(a,b)\hat{U}^{\dagger}\boldsymbol{\psi}(0), \tag{C17}$$

where $\hat{U}$ and $\hat{U}^{\dagger}$ are given by Eq. (C6). We find that (5.2) is the solution of the three-state problem. The equation corresponding to (5.2) is given correctly in Ref. 6, but (2.18) is a superfluous transformation. This causes errors in Sec. 3 of Ref. 6. If the initial condition is $\psi_1(0)=1$ and $\psi_2(0)=\psi_3(0)=0$, we use $\phi_1'(0)=1$ and $\phi_2'(0)=0$ as the initial condition in (C13). Solution of (C13) gives a and b as functions of t or z; (4.5) and (5.3) are two specific examples.

The solutions and the parameters of Eq. (C13), where $\hat{\mathcal{H}}'(t)$ is given by (C11), can be related to those given in Ref. 22 by

$$\begin{aligned}&\tfrac{1}{2}(\Omega_1^2+\Omega_2^2)^{1/2}=\Omega(t),\\ &\tan^{-1}\frac{\Omega_2}{\Omega_1}=\theta_0+B(t),\\ &a=a_1,\\ &b=-a_2^*e^{-i\theta_0}\end{aligned} \tag{C18}$$

where θ_0 is any arbitrary constant, and a_1 and a_2 are the solutions given in Ref. 22. The parameter θ_0 appears in b, and we set $\gamma=0$, as noted in Appendix B; Sec. 3 of Ref. 6 should be corrected accordingly. Finally, the solution of the three-state problem is given by (5.2) and (5.3).

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