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Comments

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Three-state systems driven by resonant optical pulses of different shapes

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New analytic solutions to the problem of a three-state system driven simultaneously by resonant optical pulses of different shapes are presented. The solutions are useful for prescribing the conditions for complete population transfer from one state to another or for complete population return.

1. INTRODUCTION

The problem of atoms or molecules in which transitions are driven by laser beams has continued to receive a great deal of attention in recent years. A number of interesting analytic solutions to the problem have been derived not only for the two-state model,¹⁻⁵ in which only one transition is assumed to be driven by the applied laser, but also for the three-, four-, and some general N -state models in which more than one transition is assumed possible.⁶⁻¹⁹ In contrast to numerical solutions of the problem, analytic solutions can be effectively used for, among other things, predicting the conditions for complete (atomic) population transfer from one level to another, complete population return, population trapping, and dynamic constants of evolution. The analytic results on various dynamic symmetries,¹⁶ which the laser-driven systems may possess, also have interesting analogs in other physical systems.

Almost all of these analytic results, however, are for cases in which the applied lasers have the same time-dependent electric-field envelopes that propagate in the same phase, although they may have different amplitudes. The analytic solution of Gottlieb¹³ for a three-state model is one of the few exceptions. In his work the two driving lasers are assumed to have the same sinusoidal electric-field envelopes but with a $\pi/2$ phase difference.¹⁴

We shall present in this paper a number of analytic solutions for the three-state systems when they are driven by a variety of concurrent resonant laser pulses of different electric-field envelopes.

2. THREE-STATE SYSTEMS

We consider an atomic or molecular system driven by two or more lasers in which the major atomic transitions take place among only three of the many available states. We assume that the time-dependent Schrödinger equation (in units of $\hbar = 1$)

$$i \frac{\partial \Psi}{\partial t} = \hat{H}(t) \Psi \quad (2.1)$$

can be reduced to the form for which the Hamiltonian $\hat{H}(t)$ can be written as

$$\hat{H}(t) = \begin{bmatrix} 0 & \alpha_{12}(t) & -i\alpha_{13}(t) \\ \alpha_{12}(t) & 0 & \alpha_{23}(t) \\ i\alpha_{13}(t) & \alpha_{23}(t) & 0 \end{bmatrix}, \quad (2.2a)$$

where $\alpha_{12}(t)$, $\alpha_{23}(t)$, and $\alpha_{13}(t)$ are generally time-dependent real quantities involving the slowly varying electric-field envelopes of the applied lasers and where the zeros along the diagonal imply that the frequencies of the lasers are at resonance with the frequencies of the atomic transitions. The off-diagonal elements are often written as

$$\begin{aligned} \alpha_{12}(t) &= -\frac{1}{2}\Omega_1(t), & \alpha_{23}(t) &= -\frac{1}{2}\Omega_2(t), \\ \alpha_{13}(t) &= -\frac{1}{2}\Omega_3(t), \end{aligned} \quad (2.3)$$

where the Ω 's are referred to as the Rabi frequencies. $\hat{H}(t)$ can be written as

$$\hat{H}(t) = \begin{bmatrix} 0 & -\frac{1}{2}\Omega_1(t) & i\frac{1}{2}\Omega_3(t) \\ -\frac{1}{2}\Omega_1(t) & 0 & -\frac{1}{2}\Omega_2(t) \\ -i\frac{1}{2}\Omega_3(t) & -\frac{1}{2}\Omega_2(t) & 0 \end{bmatrix}. \quad (2.2b)$$

Level 1 is assumed to be the ground state, and levels 2 and 3 are labeled in such a way that the electric-dipole transitions between levels 1 and 2 and between levels 2 and 3 are permitted by the electric-dipole selection rule, so that $\Omega_1(t)$ and $\Omega_2(t)$ are generally not zero. Then Laporte's rule forbids the transition between levels 1 and 3 and thus α_{13} and Ω_3 should be zero. This may be the case, and it would not affect our discussion if $\Omega_3 = 0$; however, we want to include the possibility that the presence of a nonzero Ω_3 may represent some effective coupling between levels 1 and 3 because of other factors when the problem is reduced to a three-state problem represented by a Hamiltonian of the form of Eqs. (2.2). If all the Ω 's have the same time dependence such that Ω_2/Ω_1 and Ω_3/Ω_1 are constants, 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, and we shall assume that Ω_1 , Ω_2 , and Ω_3 have different time dependences.

We first note that $\hat{H}(t)$ can be written in the form

$$\hat{H}(t) = \alpha_{12}(t)\mathcal{J}_1 + \alpha_{23}(t)\mathcal{J}_2 + \alpha_{13}(t)\mathcal{J}_3, \quad (2.4)$$

where

$$\mathcal{J}_1 = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \mathcal{J}_2 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix},$$

$$\mathcal{J}_3 = \begin{bmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{bmatrix}. \quad (2.5)$$

The matrices $\mathcal{J}_1, \mathcal{J}_2, \mathcal{J}_3$ satisfy the commutation relations

$$[\mathcal{J}_1, \mathcal{J}_2] = i\mathcal{J}_3, \quad [\mathcal{J}_2, \mathcal{J}_3] = i\mathcal{J}_1, \quad [\mathcal{J}_3, \mathcal{J}_1] = i\mathcal{J}_2 \quad (2.6)$$

and are therefore a representation of the angular-momentum operators for spin 1. Thus the three-level system belongs to the SU(2) model¹⁸ for $j = 1$, except that the angular-momentum operators that span the subspace in which $\hat{H}(t)$ lies are not in the more familiar representation in which \mathcal{J}_3 is diagonal. The eigenvectors of \mathcal{J}_3 can easily be found. We can then construct the following unitary matrix \hat{U} and its complex-conjugate transpose \hat{U}^\dagger :

$$\hat{U} = \begin{bmatrix} 1/\sqrt{2} & 0 & 1/\sqrt{2} \\ 0 & 1 & 0 \\ i/\sqrt{2} & 0 & -i/\sqrt{2} \end{bmatrix}, \quad \hat{U}^\dagger = \begin{bmatrix} 1/\sqrt{2} & 0 & -i/\sqrt{2} \\ 0 & 1 & 0 \\ 1/\sqrt{2} & 0 & i/\sqrt{2} \end{bmatrix}, \quad (2.7)$$

which transform the \mathcal{J} 's according to

$$\mathcal{J}_k' = \hat{U}^\dagger \mathcal{J}_k \hat{U}, \quad k = 1, 2, 3, \quad (2.8)$$

into a representation in which \mathcal{J}_3' is diagonal or

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

$$\mathcal{J}_3' = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix}. \quad (2.9)$$

Multiplying Eq. (2.1) from the left by \hat{U}^\dagger , we get

$$i \frac{\partial \Psi'}{\partial t} = \hat{H}'(t) \Psi', \quad (2.10)$$

where

$$\Psi' = \hat{U}^\dagger \Psi \quad (2.11)$$

and

$$\hat{H}'(t) = \hat{U}^\dagger \hat{H}(t) \hat{U} \quad (2.12)$$

or

$$\hat{H}'(t) = \alpha_{12}(t)\mathcal{J}_1' + \alpha_{23}(t)\mathcal{J}_2' + \alpha_{13}(t)\mathcal{J}_3'. \quad (2.13)$$

We can now make use of our results in Ref. 18 and obtain the analytic solution of Eq. (2.10) in terms of the solutions of the corresponding SU(2) model for $j = 1/2$, which satisfy

$$i \frac{\partial \phi}{\partial t} = \mathcal{H}(t) \phi, \quad (2.14)$$

where

$$\mathcal{H}(t) = \alpha_{12}(t)\hat{s}_1 + \alpha_{23}(t)\hat{s}_2 + \alpha_{13}(t)\hat{s}_3, \quad (2.15)$$

where $\hat{s}_1, \hat{s}_2,$ and \hat{s}_3 are the Pauli matrices multiplied by $1/2$, or

$$\mathcal{H}(t) = \begin{bmatrix} 1/2\Delta_0(t) & -1/2\Omega_0(t) \\ -1/2\Omega_0^*(t) & -1/2\Delta_0(t) \end{bmatrix}, \quad (2.16)$$

where

$$\Omega_0(t) = -\alpha_{12}(t) + i\alpha_{23}(t)$$

$$= 1/2[\Omega_1(t) - i\Omega_2(t)] \quad (2.17a)$$

and

$$\Delta_0(t) = \alpha_{13}(t) = -1/2\Omega_3(t). \quad (2.17b)$$

The quantities $\Omega_0(t)$ and $\Delta_0(t)$ are the Rabi frequency and detuning, respectively, of the two-level system whose probability amplitudes for the two states satisfy Eq. (2.14).

Let the two components of ϕ in Eq. (2.14) be ϕ_1 and ϕ_2 and let

$$\phi_1 = \phi_1' \exp\left[-i \frac{1}{2} \int^t \Delta_0(t) dt\right], \quad (2.18a)$$

$$\phi_2 = \phi_2' \exp\left[i \frac{1}{2} \int^t \Delta_0(t) dt\right]. \quad (2.18b)$$

Then Eq. (2.14) can be written as

$$i \frac{\partial \phi'}{\partial t} = \mathcal{H}'(t) \phi', \quad (2.19)$$

where

$$\mathcal{H}'(t) = \begin{bmatrix} 0 & -1/2\dot{A}(t)e^{-iB(t)} \\ -1/2\dot{A}(t)e^{iB(t)} & 0 \end{bmatrix}, \quad (2.20)$$

$$\dot{A}(t) = |\Omega_0(t)| = 1/2[\Omega_1^2(t) + \Omega_2^2(t)]^{1/2}, \quad (2.21a)$$

$$\dot{B}(t) = \frac{\Omega_1^2(t)}{\Omega_1^2(t) + \Omega_2^2(t)} \frac{d}{dt} \left[\frac{\Omega_2(t)}{\Omega_1(t)} \right] + 1/2\Omega_3(t). \quad (2.21b)$$

The quantities $\dot{A}(t)$ and $\dot{B}(t)$ are the equivalent Rabi frequency and detuning, respectively, for the two-level system, and both quantities are real here, which are in contrast to those given in Eqs. (2.16) and (2.17). The solution ϕ' given by Eq. (2.19) differs from the solution ϕ given by Eq. (2.14) only by an unimportant phase factor according to Eqs. (2.18) since $\Delta_0(t)$ or $\alpha_{13}(t)$ is assumed to be real. We shall use the solution ϕ' , which is available for a wide variety of cases, and express our three-level problem [Eq. (2.10)] and then will express the solution of Eq. (2.1) in terms of this solution.

Let $a(t)$ and $-b^*(t)$ denote the solutions for ϕ_1' and ϕ_2' , the two components of ϕ' , respectively, when the initial condition for Eq. (2.19) is $\phi_1'(0) = 1$ and $\phi_2'(0) = 0$. Then the solution of Eq. (2.19) for any arbitrary initial values of $\phi_1'(0)$ and $\phi_2'(0)$ is

$$\begin{bmatrix} \phi_1' \\ \phi_2' \end{bmatrix} = \begin{bmatrix} a & b \\ -b^* & a^* \end{bmatrix} \begin{bmatrix} \phi_1'(0) \\ \phi_2'(0) \end{bmatrix}. \quad (2.22)$$

According to the result obtained for the SU(2) model,¹⁸ the solution of Eq. (2.10) for our three-level problem can be expressed in terms of $a(t)$ and $b(t)$ obtained from Eq. (2.19) as

$$\Psi'(t) = \hat{D}^{(1)}(a, b)\Psi'(0), \tag{2.23}$$

or, more explicitly,

$$\begin{bmatrix} \Psi_1' \\ \Psi_2' \\ \Psi_3' \end{bmatrix} = \begin{bmatrix} a^2 & \sqrt{2}ab & b^2 \\ -\sqrt{2}ab^* & |a|^2 - |b|^2 & \sqrt{2}a^*b \\ b^{*2} & -\sqrt{2}a^*b^* & a^{*2} \end{bmatrix} \begin{bmatrix} \Psi_1'(0) \\ \Psi_2'(0) \\ \Psi_3'(0) \end{bmatrix}, \tag{2.24}$$

where $\hat{D}^{(1)}(a, b)$, shown explicitly in Eq. (2.24), is the three-dimensional representation of the unitary group. Using Eq. (2.11), we finally obtain the solution Ψ of Eq. (2.1) in terms of its initial value $\Psi(0)$ and in terms of the solutions $a(t)$ and $b(t)$ of the two-level system [Eq. (2.19)] as

$$\Psi(t) = \hat{U}\hat{D}^{(1)}(a, b)\hat{U}^\dagger\Psi(0), \tag{2.25}$$

where \hat{U} and \hat{U}^\dagger are given by Eqs. (2.7).

In particular, if the initial condition is

$$|\Psi_1(0)| = 1, \quad \Psi_2(0) = \Psi_3(0) = 0, \tag{2.26}$$

then we find, from Eqs. (2.23)–(2.25), that

$$\Psi_1(t) = \frac{1}{2}[a^2 + a^{*2} + b^2 + b^{*2}]\Psi_1(0), \tag{2.27a}$$

$$\Psi_2(t) = (a^*b - ab^*)\Psi_1(0), \tag{2.27b}$$

$$\Psi_3(t) = \frac{1}{2}i[a^2 - a^{*2} + b^2 - b^{*2}]\Psi_1(0). \tag{2.27c}$$

3. SOME APPLICATIONS

A number of analytic solutions for the two-level system [Eq. (2.19)] are known, and, as explained in Refs. 2 and 4, the variety of the time-dependent Rabi frequency and detuning, $\dot{A}(t)$ and $\dot{B}(t)$, can be significantly increased by changing the time variable from t to $z(t)$, where $z(t)$ can be an arbitrary nondecreasing function of the time. We shall consider a few specific cases of $\Omega_1(t)$, $\Omega_2(t)$, and $\Omega_3(t)$ in Eq. (2.2b) when neither of the ratios $\Omega_2(t)/\Omega_1(t)$ and $\Omega_3(t)/\Omega_1(t)$ is constant in general, for which the corresponding two-level problem [Eq. (2.19)] has been analytically solved.

Let

$$\begin{aligned} \Omega_1(t) &= [f(z) \cos \theta(z)]\dot{z}, & \Omega_2(t) &= [f(z) \sin \theta(z)]\dot{z}, \\ \Omega_3(t) &= g(z)\dot{z}, \end{aligned} \tag{3.1}$$

where $\dot{z}(t)$ is the time derivative of $z(t)$, which is arbitrary, and $f(z)$ is assumed to be ≥ 0 . Then, from Eqs. (2.21), we find

$$\dot{A}(t) = \frac{1}{2}f(z)\dot{z}, \tag{3.2a}$$

$$\dot{B}(t) = \left[\frac{d\theta(z)}{dz} + \frac{1}{2}g(z) \right] \dot{z}. \tag{3.2b}$$

We now consider some specific cases of $f(z)$, $g(z)$, and $\theta(z)$. Notice that if $f(z)$ and $\theta(z)$ are constants, $g(z) = 0$, and $z = t$, Eqs. (3.1) and (2.2b) reduce to a case considered by Gottlieb,¹³ by Hioe,¹⁴ and by Pegg.¹⁵

(1) Let

$$f(z) = \frac{2\alpha}{\pi[z(1-z)]^{1/2}}, \quad g(z) = \frac{2\gamma}{\pi z(1-z)}, \tag{3.3a}$$

and

$$\theta(z) = \theta_0 - \frac{\beta}{\pi} \ln(1-z), \tag{3.3b}$$

where $\alpha, \beta, \gamma, \theta_0$ are constants and where the time interval $-\infty \leq t \leq +\infty$ is mapped into $0 \leq z(t) \leq 1$. Then Eqs. (3.2) give

$$\dot{A}(t) = \frac{\alpha}{\pi} \frac{1}{[z(1-z)]^{1/2}} \dot{z} \tag{3.4a}$$

and

$$\dot{B}(t) = \frac{\beta z + \gamma}{\pi z(1-z)} \dot{z}. \tag{3.4b}$$

From Appendix 2.2 of Ref. 4, the solutions $a(t)$ and $b(t)$ of Eq. (2.19) are given by

$$a = {}_2F_1 [R - i\beta/(2\pi), -R - i\beta/(2\pi); \frac{1}{2} + i\gamma/\pi; z], \tag{3.5a}$$

$$b = \frac{-\alpha}{(2\gamma + i\pi)} z^{[1/2 - (i\gamma/\pi)]} {}_2F_1 \left[\frac{1}{2} + R - \frac{i(\beta + 2\gamma)}{2\pi}, \frac{1}{2} - R - \frac{i(\beta + 2\gamma)}{2\pi}; \frac{3}{2} - \frac{i\gamma}{\pi}; z \right], \tag{3.5b}$$

where ${}_2F_1(\alpha', \beta'; \gamma'; z)$ is the Gauss hypergeometric function²⁰ and

$$R = (2\pi)^{-1}(\alpha^2 - \beta^2)^{1/2}. \tag{3.6}$$

Substituting Eqs. (3.5) into Eqs. (2.23)–(2.25) or into Eq. (2.27) gives us the analytic solution of the three-state system [Eq. (2.1)] whose Rabi frequencies given by Eqs. (3.1) and (3.3) have different time-dependent envelopes. The arbitrary time-dependent function $z(t)$, which is a nondecreasing function of t , can be used to give an infinite variety of shapes, and Ref. 4 listed a number of them, an example of which will be given in Section 4.

If, instead of Eqs. (3.3), we let

$$f(z) = \frac{2\alpha}{\pi[z(1-z)]^{1/2}}, \quad g(z) = \frac{2\beta}{\pi(1-z)}, \tag{3.7a}$$

$$\theta(z) = \theta_0 - \frac{2\gamma}{\pi} \tanh^{-1}(1-2z), \tag{3.7b}$$

then Eqs. (3.2) give exactly the same $\dot{A}(t)$ and $\dot{B}(t)$ as those given by Eqs. (3.4). Thus we obtain exactly the same result.

(2) Let

$$f(z) = \frac{2\alpha}{\pi(z^2 + 1)}, \quad g(z) = \frac{2\gamma z}{\pi(z^2 + 1)}, \tag{3.8a}$$

and

$$\theta(z) = \theta_0 + \frac{\beta}{\pi} \tan^{-1} z, \tag{3.8b}$$

where $\alpha, \beta, \gamma, \theta_0$ are again constants. Here $z(t)$ is assumed to increase from $-\infty$ to $+\infty$ as t increases from $-\infty$ to $+\infty$, and thus the range of $\theta(z)$ is between $\theta_0 - \frac{1}{2}\beta$ and $\theta_0 + \frac{1}{2}\beta$. From Eq. (3.2) we find that

$$\dot{A}(t) = \frac{\alpha}{\pi} \frac{1}{z^2 + 1} \dot{z} \tag{3.9a}$$

and

$$\dot{B}(t) = \frac{\beta + \gamma z}{\pi(z^2 + 1)} \dot{z}. \tag{3.9b}$$

For Rabi frequency and detuning of these forms, the solutions $a(t)$ and $b(t)$ of Eq. (2.19) are given in Appendix 2.1 of

Ref. 4, where $a(t)$ can be identified with a_1 and $b(t)$ with $-a_2^*$. The solutions are again expressed in terms of the Gauss hypergeometric function. In the special case of $\gamma = 0$ or $\Omega_3(t) = 0$, $A(t)$ and $B(t)$ given by Eqs. (3.9) have the same time dependence, and the two-level problem [Eq. (2.19)] can be readily solved, although in the corresponding three-level problem the ratio of $\Omega_2(t)/\Omega_1(t)$ is not constant. In this case we find that, as $t \rightarrow +\infty$,

$$a(+\infty) = \left(\cos r + i \frac{\beta}{2r} \sin r \right) \exp(-i\beta/2), \quad (3.10a)$$

$$b(+\infty) = i \frac{\beta}{2r} \sin r, \quad (3.10b)$$

where

$$r = 1/2(\alpha^2 + \beta^2)^{1/2}. \quad (3.11)$$

If the initial value of Ψ in Eq. (2.1) is given by Eqs. (2.26), then from Eqs. (2.27) we find that

$$|\Psi_1(+\infty)|^2 = \left[\left(\cos^2 r - \frac{\beta^2}{4r^2} \sin^2 r \right) \cos \beta + \frac{\beta}{2r} \sin(2r) \sin \beta - \frac{\alpha^2}{4r^2} \sin^2 r \right]^2, \quad (3.12a)$$

$$|\Psi_2(+\infty)|^2 = \left(\frac{\alpha}{r} \sin r \right)^2 \left(\cos r \cos \frac{1}{2}\beta + \frac{\beta}{2r} \sin r \sin \frac{1}{2}\beta \right)^2, \quad (3.12b)$$

$$|\Psi_3(+\infty)|^2 = \left[\left(\cos^2 r - \frac{\beta^2}{4r^2} \sin^2 r \right) \sin \beta - \frac{\beta}{2r} \sin(2r) \cos \beta \right]^2, \quad (3.12c)$$

which give the final occupation probabilities of the three levels.

4. SOLUTIONS IN TERMS OF CLAUSEN FUNCTION

We have shown in the previous sections that the solution of Eq. (2.1) with $\hat{H}(t)$ given by Eqs. (2.2) can always be expressed in terms of the solution of the two-level system [Eq. (2.19)], where $\hat{H}'(t)$ is given by Eq. (2.20). We have given examples in which the corresponding two-level system is of the analytically solvable type. In this section we shall consider some examples in which the analytic solutions of the corresponding two-level problem do not appear obvious. Instead, solving Eq. (2.1) directly by eliminating two of the

Table 1. Cases of Complete Transfer of the Occupation Probability from State 1 to State 3^a

α_1	α_2	z_f	$\int \Omega_1(t) dt$
2π	π	1	2π
4π	$3 \times 7^{-1/2}\pi$	4/7	2.182516 π
6π	1.393889 π	1	6π
6π	1.165659 π	0.302727	2.225412 π
8π	1.177527 π	0.181497	2.241378 π
8π	1.483642 π	0.862208	6.063121 π
10π	1.612837 π	1	10π

^a We assume that the optical pulse amplitudes are given by Eqs. (4.2); $z(t)$ is an arbitrary function that increases from 0 to z_f .

Rabi Frequency

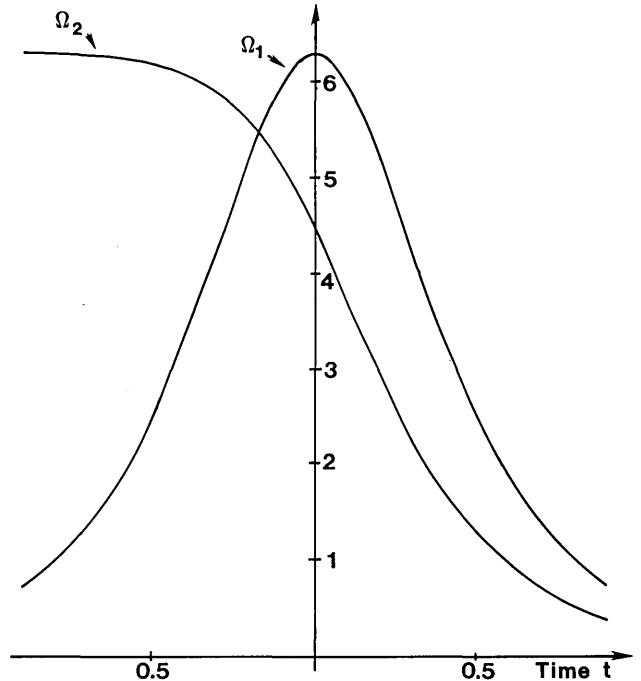


Fig. 1. Optical pulse shapes given by Eqs. (4.2) and (4.5) with $\tau = 1$. This implies that $z(t)$ runs from zero to one and that $\Omega_1(t)$ has a hyperbolic-secant shape. Also we use $\alpha_1 = 2\pi$ and $\alpha_2 = \pi$, which gives complete transfer of the occupation probability from state 1 to state 3 as t increases from $-\infty$ to $+\infty$.

three components of Ψ from the coupled equations leads to a known type of third-order equation²¹ satisfied by the Clausen function ${}_3F_2(\alpha', \beta', \gamma'; \delta', \epsilon'; z)$, which has many known properties. In particular, ${}_3F_2(\alpha', \beta', \gamma'; \delta', \epsilon'; z)$ has a series representation given by

$$1 + \frac{\alpha' \beta' \gamma'}{1! \delta' \epsilon'} z + \frac{\alpha'(\alpha'+1) \beta'(\beta'+1) \gamma'(\gamma'+1)}{2! \delta'(\delta'+1) \epsilon'(\epsilon'+1)} z^2 + \dots \quad (4.1)$$

We let, in Eq. (2.2b),

$$\Omega_1(t) = \frac{\alpha_1}{\pi[z(1-z)]^{1/2}} \dot{z}, \quad \Omega_2(t) = \frac{\alpha_2}{\pi z(1-z)^{1/2}} \dot{z}, \quad \Omega_3(t) = 0, \quad (4.2)$$

where α_1 and α_2 are nonzero constants. As before, $z(t)$ is an arbitrary function of time, and z increases from 0 to $z_f \leq 1$ as t increases from $-\infty$ to $+\infty$. Assuming that states 2 and 3 are unoccupied at $z = 0$, which corresponds to the initial time, the solutions for $\Psi_1(z)$, $\Psi_2(z)$, and $\Psi_3(z)$ of Eq. (2.1) are

$$\Psi_1 = {}_3F_2 \left(\frac{1}{2}, \frac{\alpha_1}{2\pi}, -\frac{\alpha_1}{2\pi}; \frac{1}{2} + \frac{i\alpha_2}{2\pi}, \frac{1}{2} - \frac{i\alpha_2}{2\pi}; z \right), \quad (4.3a)$$

$$\Psi_2 = \frac{i\alpha_1/\pi}{1 + \alpha_2^2/\pi^2} [z(1-z)]^{1/2} {}_3F_2 \left(\frac{3}{2}, 1 + \frac{\alpha_1}{2\pi}, 1 - \frac{\alpha_1}{2\pi}; \frac{3}{2} + \frac{i\alpha_2}{2\pi}, \frac{3}{2} - \frac{i\alpha_2}{2\pi}; z \right), \quad (4.3b)$$

$$\Psi_3 = \frac{-\alpha_1\alpha_2/\pi^2}{1 + \alpha_2^2/\pi^2} z^{1/2} {}_3F_2\left(\frac{1}{2}, 1 + \frac{\alpha_1}{2\pi}, 1 - \frac{\alpha_1}{2\pi}; \frac{3}{2} + \frac{i\alpha_2}{2\pi}, \frac{3}{2} - \frac{i\alpha_2}{2\pi}; z\right). \quad (4.3c)$$

If we assume that α_1 is an even integral multiple of π , then the Clausen series ${}_3F_2$ terminates, and Ψ_1 , Ψ_2 , and Ψ_3 are proportional to polynomials. Evaluation of $|\Psi_1|^2$, $|\Psi_2|^2$, and $|\Psi_3|^2$, the occupation probabilities, is now quite straightforward.

Complete transfer of the occupation probability from state 1 to state 3 by the two concurrent, resonant optical pulses is obtained in several cases, which we list in Table 1. Because the series terminates, $|\Psi_2|^2$ vanishes as $z \rightarrow 1$; hence, if $z_f = 1$, the condition for complete transfer is

$${}_3F_2\left(\frac{1}{2}, \frac{\alpha_1}{2\pi}, -\frac{\alpha_1}{2\pi}; \frac{1}{2} + \frac{i\alpha_2}{2\pi}, \frac{1}{2} - \frac{i\alpha_2}{2\pi}; 1\right) = 0. \quad (4.4)$$

However, either $z_f = 1$ or $z_f < 1$ may lead to an algebraic equation for α_2^2 for complete transfer. Solving such equations gives the numbers in the second column of Table 1. The simplest case appears as the first line in Table 1; here, $\Psi_1 = 1 - z$, $\Psi_2 = i[z(1 - z)]^{1/2}$, and $\Psi_3 = -z^{1/2}$. The area of pulse 1, defined by

$$\int \Omega_1(t) dt,$$

where the integral runs from $-\infty$ to $+\infty$ or over the whole of the optical pulse, appears to be a significant number for characterizing the pulse and is included in Table 1. If we define the area of pulse 2 in a similar way, the integral would

Occupation Probability

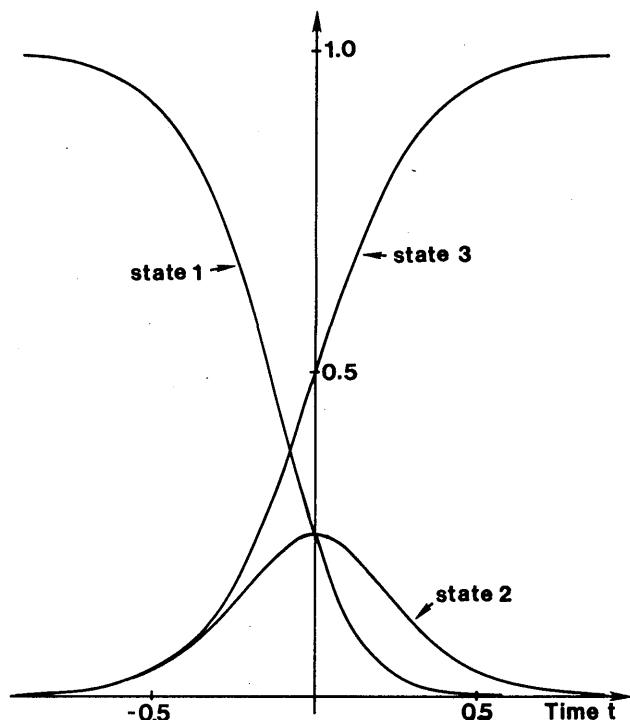


Fig. 2. Time-dependent occupation probabilities corresponding to the first line of Table 1; we used Eq. (4.5) with $\tau = 1$.

Occupation Probability

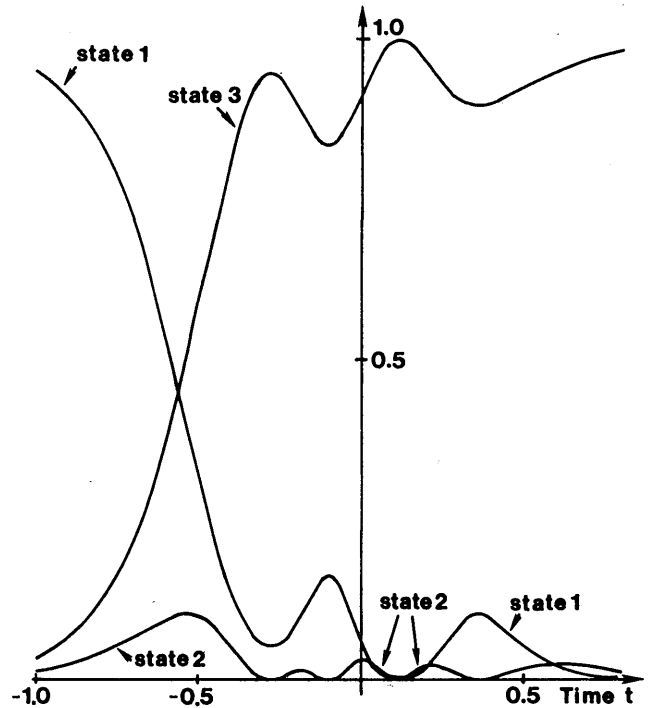


Fig. 3. Time-dependent occupation probabilities corresponding to the last line of Table 1; we used Eq. (4.5) with $\tau = 1$.

diverge. The divergence, which comes from the neighborhood of $z = 0$ or from early times, poses no problem physically since we assume that states 2 and 3 are unoccupied at the start of the two concurrent optical pulses.

To see specific examples of the pulses [Eqs. (4.2)] and solution [Eqs. (4.3)], let

$$z(t) = \frac{1}{2}[1 + \tanh(\pi t/\tau)], \quad (4.5)$$

where τ is the time constant. It gives a hyperbolic-secant pulse shape, first used by Rosen and Zener,²² for $\Omega_1(t)$. The pulse shape for $\Omega_2(t)$ is quite different, and it has an infinite area. The two pulse shapes are shown in Fig. 1. The time-dependent occupation probabilities obtained from our solution (4.3) are shown in Figs. 2 and 3 for two different sets of values for α_1 and α_2 .

We can obtain further new results by combining the results of this section with those of the previous section. Let

$$\begin{aligned} \Omega_1(t) &= [f(z)\cos\theta(z)]\dot{z}, & \Omega_2(t) &= [f(z)\sin\theta(z)]\dot{z}, \\ \Omega_3(t) &= 0, \end{aligned} \quad (4.6a)$$

where

$$\begin{aligned} f(z) &= \frac{\alpha}{\pi z(1-z)^{1/2}}, \\ \theta(z) &= \frac{\beta}{\pi} \sin^{-1}(1-2z) + \frac{\pi-\beta}{2}, \end{aligned} \quad (4.6b)$$

where α and β are positive constants. We assume that states 2 and 3 are unoccupied at $z = 0$ or $t = -\infty$, the initial time. The solution of Eq. (2.1) is

$$\Psi_1 = (\sin \theta) {}_3F_2 \left(\frac{1}{2}, \frac{\beta}{\pi}, -\frac{\beta}{\pi}; \frac{1}{2} + \frac{i\alpha}{2\pi}, \frac{1}{2} - \frac{i\alpha}{2\pi}; z \right) + \frac{2\beta \cos \theta}{\pi(1 + \alpha^2/\pi^2)} [z(1-z)]^{1/2} {}_3F_2 \left(\frac{3}{2}, 1 + \frac{\beta}{\pi}, 1 - \frac{\beta}{\pi}; \frac{3}{2} + \frac{i\alpha}{2\pi}, \frac{3}{2} - \frac{i\alpha}{2\pi}; z \right), \quad (4.7a)$$

$$\Psi_2 = \frac{2i\alpha\beta/\pi^2}{1 + \alpha^2/\pi^2} z^{1/2} {}_3F_2 \left(\frac{1}{2}, 1 + \frac{\beta}{\pi}, 1 - \frac{\beta}{\pi}; \frac{3}{2} + \frac{i\alpha}{2\pi}, \frac{3}{2} - \frac{i\alpha}{2\pi}; z \right), \quad (4.7b)$$

and

$$\Psi_3 = \frac{2\beta \sin \theta}{\pi(1 + \alpha^2/\pi^2)} [z(1-z)]^{1/2} {}_3F_2 \left(\frac{3}{2}, 1 + \frac{\beta}{\pi}, 1 - \frac{\beta}{\pi}; \frac{3}{2} + \frac{i\alpha}{2\pi}, \frac{3}{2} - \frac{i\alpha}{2\pi}; z \right) - (\cos \theta) {}_3F_2 \left(\frac{1}{2}, \frac{\beta}{\pi}, -\frac{\beta}{\pi}; \frac{1}{2} + \frac{i\alpha}{2\pi}, \frac{1}{2} - \frac{i\alpha}{2\pi}; z \right). \quad (4.7c)$$

If β is an integral multiple of π , all the series appearing here terminate, and the condition for complete transfer of population can be readily obtained as before.

5. SUMMARY

We have presented new analytic solutions to the problem of a three-state system driven by two or more resonant laser pulses of different shapes. The three-state system, whose Hamiltonian is given by Eqs. (2.2), is shown to be reducible to a two-state system whose Hamiltonian is given by Eq. (2.20). We have presented specific examples in which the time-dependent envelopes of the laser pulses assume various different forms, each consisting of an infinite variety, for which the solutions are expressed in terms of known functions. Our solutions can be used to give useful information, such as the conditions for complete population transfer or return.

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