

Three-state model driven by two laser beams

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We use a three-state model for an atom or molecule in which two transitions are simultaneously driven by the oscillating electric fields of two laser beams. The amplitudes and detunings of the applied oscillating fields vary during the optical pulse, which can cause transitions from one state to another. For some special cases not previously known, the transition probabilities and probabilities of no transition are obtained analytically. We give conditions for complete transfer of the atomic population from one state to another, and for complete return to the initial state.

Atoms or molecules in which transitions are driven by laser beams are often treated by using a classical description of the applied oscillating field and a quantum description of the state of the atom or molecule. The two-state model, in which one transition is driven by the applied oscillating field, has been much studied.¹ The dynamics of the three-state model, with two driven transitions, deserves some attention. The two transitions are driven by concurrent optical pulses, so that the work on the two-state model is not directly relevant. In two symmetric cases, the equations of motion for the driven three-state system can be simplified,² and analytic solutions of the time-dependent Schrödinger equation can then be found. This paper treats less symmetric cases in which analytic solutions can be found only by use of Clausen's special function.³ We shall find simple expressions for the occupation probabilities of the three states at the end of the concurrent optical pulses. These formulas can easily give conditions for complete transfer of the occupation probability to any desired state, or complete return to the initial state.

We write the Schrödinger equation for the atom or molecule as

$$i\hbar \frac{d}{dt} \psi = H(t) \psi, \quad (1)$$

where ψ is a vector with three complex components and $H(t)$ is a 3×3 matrix that depends on the energies of the three states and on the amplitudes and frequencies of the two applied oscillating fields. Suppose that $H(t)$ is a diagonal matrix in the absence of the applied fields. This means that E_1 , E_2 , and E_3 , the energies of the three states, appear as diagonal elements of $H(t)$. We do not assume anything about the order of E_1 , E_2 , and E_3 on the energy scale. We use the electric dipole approximation in describing effects of the oscillating fields. This means that

products of electric fields and transition dipole moments appear in off-diagonal elements of $H(t)$. We assume that states 1 and 3 have the same parity, so that Laporte's rule⁴ gives $H_{13} = H_{31} = 0$. We assume that each laser beam drives only its own transition, so that the amplitude and frequency of oscillations in H_{12} and H_{23} can be varied independently. These off-diagonal elements of $H(t)$ contain oscillating terms of both positive and negative frequencies, unless circularly polarized light propagating along a magnetic field is used. Frequencies of the wrong sign are far from resonance, and dropping terms containing them is consistent with the assumption that each transition is driven by only one of the applied oscillating fields. The neglect of such terms is the rotating-wave approximation, and it makes H_{12} and H_{23} proportional to $\exp(-i\omega_{12}t)$ and $\exp(-i\omega_{23}t)$, where ω_{12} and ω_{23} are applied frequencies having the same signs as $E_1 - E_2$ and $E_2 - E_3$. These exponential factors in off-diagonal elements of $H(t)$ can be removed by a time-dependent unitary transformation; the general formulation is given by Einwohner, Wong, and Garrison.⁵ Let $\hbar = 1$; the two detunings become

$$\Delta_1(t) = E_1 - E_2 - \omega_{12}$$

and

$$\Delta_2(t) = E_2 - E_3 - \omega_{23},$$

and their signs follow from the convention mentioned above and used earlier.⁶ These detunings appear in diagonal elements of the transformed Hamiltonian matrix. Their time dependence can be produced by use of Stark or Zeeman effects if frequency modulation of the applied oscillating fields is inconvenient. The time-dependent unitary transformation eliminates all optical frequencies from (1), and gives

$$i \frac{d}{dt} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} = \begin{pmatrix} (2\Delta_1 + \Delta_2)/3 & -\frac{1}{2}\Omega_1(t) & 0 \\ -\frac{1}{2}\Omega_1(t) & (-\Delta_1 + \Delta_2)/3 & -\frac{1}{2}\Omega_2(t) \\ 0 & -\frac{1}{2}\Omega_2(t) & -(\Delta_1 + 2\Delta_2)/3 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix}, \quad (2)$$

where $|a_1|^2$, $|a_2|^2$, and $|a_3|^2$ are time-dependent occupation probabilities for the three states, and $\Omega_1(t)$, $\Omega_2(t)$ are the so-called Rabi frequencies. The applied electric fields oscillate with time-dependent amplitudes, and each of these Rabi frequencies is proportional to the amplitude of the corresponding applied field. We assume that $\Omega_1(t)$ and $\Omega_2(t)$ are positive functions of t , and that their ratio is constant. This suggests that the two oscillating fields could be derived from the same laser. The pulse area of McCall and Hahn⁷ is replaced by two dimensionless areas

$$\alpha_1 = \int_{-\infty}^{\infty} \Omega_1(t) dt, \quad \alpha_2 = \int_{-\infty}^{\infty} \Omega_2(t) dt.$$

A preliminary step toward writing solutions of (2) in terms of one of the special functions is a change of independent variable. We introduce $x(t)$, a nondecreasing function of t that varies from 0 to 1. The two Rabi frequencies are given by

$$\Omega_1(t) = \frac{\alpha_1}{\pi} [x(1-x)]^{-1/2} \frac{dx}{dt}, \quad (3a)$$

$$\Omega_2(t) = \frac{\alpha_2}{\pi} [x(1-x)]^{-1/2} \frac{dx}{dt},$$

and the two detunings are given by

$$\Delta_1(t) = \left[\frac{\beta_1}{x} + \frac{\gamma_1}{1-x} \right] \frac{dx}{dt}, \quad \Delta_2(t) = \left[\frac{\beta_2}{x} + \frac{\gamma_2}{1-x} \right] \frac{dx}{dt}. \quad (3b)$$

Specific choices of the function $x(t)$ are listed elsewhere.⁸ Here we mention the function of Rosen and Zener:⁹

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

where τ is the time constant. The resulting Rabi frequencies and detunings are

$$\Omega_j(t) = \frac{\alpha_j}{\tau} \operatorname{sech} \left[\frac{\pi t}{\tau} \right]$$

and

$$\Delta_j(t) = \frac{\pi}{\tau} \left[\beta_j + \gamma_j - (\beta_j - \gamma_j) \tanh \left[\frac{\pi t}{\tau} \right] \right],$$

where $j=1$ or 2 . An example is shown in Fig. 1.

Analytic solutions of (2) can be written in terms of Clausen's function if

$$\beta_1 + \beta_2 = 0 \quad \text{or} \quad \gamma_1 + \gamma_2 = 0.$$

This condition should be distinguished from the condition of two-photon resonance,² which is

$$\beta_1 + \beta_2 = 0 \quad \text{and} \quad \gamma_1 + \gamma_2 = 0.$$

Clausen's brief paper³ introduces the series

$$1 + \frac{\alpha' \beta'}{1! \gamma' \epsilon'} x + \frac{\alpha'(\alpha'+1) \beta'(\beta'+1) \delta'(\delta'+1)}{2! \gamma'(\gamma'+1) \epsilon'(\epsilon'+1)} x^2 + \dots \quad (4)$$

and gives a third-order differential equation satisfied by this function of x , which is $F(\alpha', \beta', \delta'; \gamma', \epsilon'; x)$ in

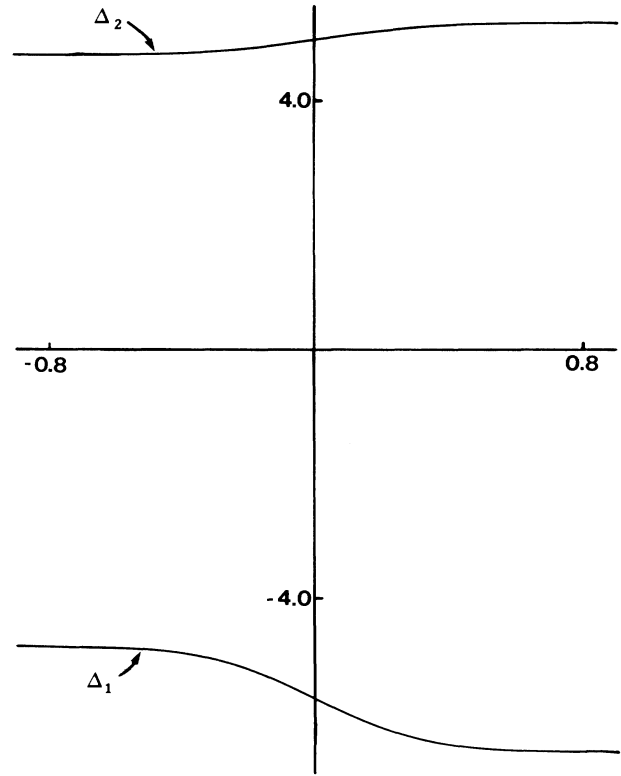


FIG. 1. Detuning functions that go with hyperbolic-secant pulse shape. Here we assume $\beta_1 = -0.7540$, $\gamma_1 = -1.0287$, $\beta_2 = 0.7540$, $\gamma_2 = 0.8350$, and $\tau = 1$.

Pochhammer's semicolon notation.¹⁰ Other work on this function is reviewed by Erdélyi *et al.*¹¹ and by Slater.¹²

After writing the general solution of (2) in terms of Clausen's function, we impose the initial condition that only one of the three states is occupied at $t = -\infty$, before the concurrent optical pulses act on the three-state system. This is easily done if $\gamma_1 + \gamma_2 = 0$, whereas a discussion of time reversal is needed if $\beta_1 + \beta_2 = 0$. Furthermore, a discussion of the behavior of Clausen's series near $x=1$ is needed in either case. If (4) diverges at $x=1$, we can find the behavior of Clausen's function when $1-x$ is small and positive, and apply the result to this problem. In certain special cases, we can use the work of Watson and Whipple¹³ to find the final occupation probabilities given in Tables I and II. Time reversal connects the probabilities of no transition given in Tables I and II, but not the transition probabilities. In other special cases, the series (4) terminates, and evaluation of Clausen's function at $x=1$ is straightforward. The integer n , which is approximately the number of terms in (4), appears in the resulting formulas. For $n < 2$, the final occupation probabilities are given in Table III. The appearance of this integer, and the restriction of our tabulated results to cases that give simple formulas, act to impose various conditions on

$$\alpha_1, \alpha_2, \beta_1, \beta_2, \gamma_1, \gamma_2, \quad (5)$$

TABLE I. Final occupation probabilities for the three states in the special case of $\beta_1 + \beta_2 = 0$, $\beta_1 = -\frac{1}{2}(\gamma_1 - \gamma_2)$, and $\alpha_1^2 - \alpha_2^2 = 2\pi^2(\gamma_1^2 - \gamma_2^2)$. To simplify the formulas, we define $\Phi = [\frac{1}{4}(\alpha_1^2 + \alpha_2^2) - \frac{1}{2}\pi^2(\gamma_1^2 + \gamma_2^2)]^{1/2}$; this angle can be positive, zero, or imaginary.

Initial occupation probabilities	Final occupation probabilities
1	$\frac{[\cosh(\pi\gamma_1) + \cos\Phi][\cosh(\pi\gamma_2) + \cos\Phi]}{4 \cosh(\pi\gamma_1)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$
0	$\frac{[\cosh(\pi\gamma_1) - \cos\Phi][\cosh(\pi\gamma_2) + \cos\Phi]}{2 \cosh(\pi\gamma_1)\cosh(\pi\gamma_2)}$
0	$\frac{[\cosh(\pi\gamma_1) - \cos\Phi][\cosh(\pi\gamma_2) - \cos\Phi]}{4 \cosh(\pi\gamma_2)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$
0	$\frac{[\cosh(\pi\gamma_1) + \cos\Phi][\cosh(\pi\gamma_1) - \cos\Phi]}{2 \cosh(\pi\gamma_1)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$
1	$\frac{\cos^2\Phi}{\cosh(\pi\gamma_1)\cosh(\pi\gamma_2)}$
0	$\frac{[\cosh(\pi\gamma_2) + \cos\Phi][\cosh(\pi\gamma_2) - \cos\Phi]}{2 \cosh(\pi\gamma_2)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$
0	$\frac{[\cosh(\pi\gamma_1) - \cos\Phi][\cosh(\pi\gamma_2) - \cos\Phi]}{4 \cosh(\pi\gamma_1)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$
0	$\frac{[\cosh(\pi\gamma_1) + \cos\Phi][\cosh(\pi\gamma_2) - \cos\Phi]}{2 \cosh(\pi\gamma_1)\cosh(\pi\gamma_2)}$
1	$\frac{[\cosh(\pi\gamma_1) + \cos\Phi][\cosh(\pi\gamma_2) + \cos\Phi]}{4 \cosh(\pi\gamma_2)\cosh[\frac{1}{2}\pi(\gamma_1 + \gamma_2)]\cosh[\frac{1}{2}\pi(\gamma_1 - \gamma_2)]}$

TABLE II. Final occupation probabilities for the three states in the special case of $\gamma_1 + \gamma_2 = 0$, $\gamma_1 = -\frac{1}{2}(\beta_1 - \beta_2)$, and $\alpha_1^2 - \alpha_2^2 = 2\pi^2(\beta_1^2 - \beta_2^2)$. We define $\Phi = [\frac{1}{4}(\alpha_1^2 + \alpha_2^2) - \frac{1}{2}\pi^2(\beta_1^2 + \beta_2^2)]^{1/2}$.

Initial occupation probabilities	Final occupation probabilities
1	$\frac{[\cosh(\pi\beta_1) + \cos\Phi][\cosh(\pi\beta_2) + \cos\Phi]}{4 \cosh(\pi\beta_1)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_1 - \beta_2)]}$
0	$\frac{[\cosh(\pi\beta_1) + \cos\Phi][\cosh(\pi\beta_1) - \cos\Phi]}{2 \cosh(\pi\beta_1)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_1 - \beta_2)]}$
0	$\frac{[\cosh(\pi\beta_1) - \cos\Phi][\cosh(\pi\beta_2) - \cos\Phi]}{4 \cosh(\pi\beta_1)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_1 - \beta_2)]}$
0	$\frac{[\cosh(\pi\beta_1) - \cos\Phi][\cosh(\pi\beta_2) + \cos\Phi]}{2 \cosh(\pi\beta_1)\cosh(\pi\beta_2)}$
1	$\frac{\cos^2\Phi}{\cosh(\pi\beta_1)\cosh(\pi\beta_2)}$
0	$\frac{[\cosh(\pi\beta_1) + \cos\Phi][\cosh(\pi\beta_2) - \cos\Phi]}{2 \cosh(\pi\beta_1)\cosh(\pi\beta_2)}$
0	$\frac{[\cosh(\pi\beta_1) - \cos\Phi][\cosh(\pi\beta_2) - \cos\Phi]}{4 \cosh(\pi\beta_2)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_1 - \beta_2)]}$
0	$\frac{[\cosh(\pi\beta_2) + \cos\Phi][\cosh(\pi\beta_2) - \cos\Phi]}{2 \cosh(\pi\beta_2)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_1 - \beta_2)]}$
1	$\frac{[\cosh(\pi\beta_1) + \cos\Phi][\cosh(\pi\beta_2) + \cos\Phi]}{4 \cosh(\pi\beta_2)\cosh[\frac{1}{2}\pi(\beta_1 + \beta_2)]\cosh[\frac{1}{2}\pi(\beta_2 - \beta_2)]}$

TABLE III. Examples of final occupation probabilities for the three states obtained by assuming $\gamma_1 + \gamma_2 = 0$ and that the series terminate. The pulse areas α_1 and α_2 are given in Table IV along with applicable inequalities. For any value of n , the final occupation probability of state 2 is zero.

Initial occupation probabilities	Integer n	Final occupation probabilities
1	1	$\frac{4\beta_1^2(\beta_1 + \beta_2)^2 + (3\beta_1 + \beta_2 - 2\gamma_1)^2}{(4\beta_1^2 + 1)(\beta_1 + \beta_2)^2}$
0		0
0		$\frac{-4(\beta_1 - \gamma_1)(2\beta_1 + \beta_2 - \gamma_1)}{(4\beta_1^2 + 1)(\beta_1 + \beta_2)^2}$
0	0	$(\beta_2 - \gamma_1)/(\beta_1 + \beta_2)$
1		0
0		$(\beta_1 + \gamma_1)/(\beta_1 + \beta_2)$
0	1	$\frac{(\beta_2 - \gamma_1)[(\beta_1^2 + \frac{1}{4})(\beta_2^2 + \frac{1}{4}) + 2\beta_1(\beta_1 - \beta_2) + 2(3\beta_1 - \beta_2)\gamma_1 + 4\gamma_1^2]}{(\beta_1 + \beta_2)(\beta_1^2 + \frac{1}{4})(\beta_2^2 + \frac{1}{4})}$
1		0
0		$\frac{(\beta_1 + \gamma_1)[(\beta_1^2 + \frac{1}{4})(\beta_2^2 + \frac{1}{4}) - 2\beta_2(\beta_1 - \beta_2) + 2(\beta_1 - 3\beta_2)\gamma_1 + 4\gamma_1^2]}{(\beta_1 + \beta_2)(\beta_1^2 + \frac{1}{4})(\beta_2^2 + \frac{1}{4})}$
0	1	$\frac{-4(\beta_2 + \gamma_1)(\beta_1 + 2\beta_2 + \gamma_1)}{(4\beta_2^2 + 1)(\beta_1 + \beta_2)^2}$
0		0
1		$\frac{4\beta_2^2(\beta_1 + \beta_2)^2 + (\beta_1 + 3\beta_2 + 2\gamma_1)^2}{(4\beta_2^2 + 1)(\beta_1 + \beta_2)^2}$

the six parameters of our three-state model. These various conditions are listed in Table IV. In each section of Table IV, and for each value of n , three of the parameters (5) vary independently, and the other three are given in

terms of them. Since the formulas of Watson and Whipple can be applied to terminating series as well as convergent infinite series, there are some small areas of overlap between Tables II and III, and between different sections

TABLE IV. Conditions for obtaining final occupation probabilities in closed form. The six parameters of the model were introduced in (3), and the integer n appears if the series terminate.

Initial occupation probabilities	Conditions on pulse parameters	Final occupation probabilities
Arbitrary	$\beta_1 + \beta_2 = 0, \beta_1 = -\frac{1}{2}(\gamma_1 - \gamma_2)$ $\alpha_1^2 - \alpha_2^2 = 2\pi^2(\gamma_1^2 - \gamma_2^2)$	Results in Table I
Arbitrary	$\gamma_1 + \gamma_2 = 0, \gamma_1 = -\frac{1}{2}(\beta_1 - \beta_2)$ $\alpha_1^2 - \alpha_2^2 = 2\pi^2(\beta_1^2 - \beta_2^2)$	Results in Table II
1	$\gamma_1 + \gamma_2 = 0, n > 0, 0 < \alpha_1 < 2\pi n$ $\alpha_1^2 = 4\pi^2 n^2 [1 + (\beta_1 - \gamma_1)/(\beta_1 + \beta_2)]$ $\alpha_2^2 = -4\pi^2(\beta_1 - \gamma_1)[\beta_1 + \beta_2 + n^2/(\beta_1 + \beta_2)]$	Example in Table III
0		Examples in Table III
0		
0	$\gamma_1 + \gamma_2 = 0, n \geq 0, (\beta_1 + \gamma_1)(\beta_2 - \gamma_1) > 0$ $\alpha_1^2 = 4\pi^2[\beta_1^2 + (n + \frac{1}{2})^2](\beta_2 - \gamma_1)/(\beta_1 + \beta_2)$ $\alpha_2^2 = 4\pi^2[\beta_2^2 + (n + \frac{1}{2})^2](\beta_1 + \gamma_1)/(\beta_1 + \beta_2)$	Examples in Table III
1		
0		
0	$\gamma_1 + \gamma_2 = 0, n > 0, 0 < \alpha_2 < 2\pi n$ $\alpha_1^2 = -4\pi^2(\beta_2 + \gamma_1)[\beta_1 + \beta_2 + n^2/(\beta_1 + \beta_2)]$ $\alpha_2^2 = 4\pi^2 n^2 [1 + (\beta_2 + \gamma_1)/(\beta_1 + \beta_2)]$	Example in Table III
0		
1		

TABLE V. Conditions for complete transfer of the occupation probability from one state to another, or complete return to the initial state. Here we assume $\gamma_1 + \gamma_2 = 0$. Since the series all terminate, the conditions listed in the appropriate section of Table IV must also be satisfied.

Initial occupation probabilities		Final occupation probabilities		Condition for this
1	0	1	0	$F(1+n, 1-n, 1-i(2\beta_1+\beta_2-\gamma_1); \frac{3}{2}-i\beta_1, 2-i(\beta_1+\beta_2); 1)=0$
0	0	0		
0	0	0		
1	0	0	0	$F(n, -n, -i(2\beta_1+\beta_2-\gamma_1); \frac{1}{2}-i\beta_1, -i(\beta_1+\beta_2); 1)=0$
0	0	0		
0	0	1		
0	1	1	0	$F(-n, n+1, \frac{1}{2}+i(\beta_1-\beta_2+\gamma_1); \frac{1}{2}+i\beta_1, \frac{3}{2}-i\beta_2; 1)=0$
1	0	0		
0	0	0		
0	0	1	0	$F(n, -n, i(\beta_1+2\beta_2+\gamma_1); \frac{1}{2}+i\beta_2, i(\beta_1+\beta_2); 1)=0$
0	0	0		
1	0	0		

of Table IV.

After finding simple formulas for the final occupation probabilities, we can set two of the final occupation probabilities equal to zero. This gives cases of complete transfer of the atomic population to a specific state, or complete return to the initial state. When such cases are obtained from Tables I or II, we find that the series (4) terminates, which simplifies the calculations. We can now find more general conditions for complete transfer or complete return, by starting with the assumption that (4) terminates. If $\gamma_1 + \gamma_2 = 0$, we find conditions for complete transfer or complete return that are listed in Table V. Each equation in Table V amounts to two conditions on the parameters (5) because it has real and imaginary parts. Also, the assumption that all series terminate leads to the

conditions listed in Table IV. Thus for each initial condition and each value of n , we have five conditions on the parameters (5), and a one-parameter family of cases of complete transfer or complete return. If n is less than about 3 or 4, all these conditions can be written out and incorporated in formulas for the parameters (5). Some specific examples are given in Table VI.

If we assume $\beta_1 + \beta_2 = 0$, the conditions for complete transfer or complete return can now be obtained easily by using the time-reversal argument mentioned above. Since two of the initial occupation probabilities and two of the final occupation probabilities are zero, time reversal is more useful here than in the general treatment of (2). An example with $\beta_1 + \beta_2 = 0$ is included in Table VI. If the hyperbolic-secant pulse shape is used, the detunings for

TABLE VI. Specific cases of complete transfer and complete return. The six parameters of the model were introduced in (3), and are here determined explicitly.

Initial occupation probabilities		Final occupation probabilities		n	α_1	α_2	β_1	β_2	γ_1	γ_2
1	0	1	0	3	2.2637	20.7819	0.3043	-1.7534	-1.1239	1.1239
0	0	0								
0	0	0								
1	0	0	0	2	8.7074	9.0630	0.8642	-0.8182	0.8881	-0.8881
0	0	0								
0	0	1								
0	1	1	0	2	8.8677	14.9498	3.5659	0.2748	-0.1285	0.1285
1	0	0								
0	0	0								
1	0	0	0	2	8.1665	9.5834	-0.7540	0.7540	-1.0287	0.8350
0	0	0								
0	0	1								

this example are those shown in Fig. 1. The precise formulation of time reversal, and other details of our calculations, will be submitted for publication elsewhere.

This treatment of complete transfer and complete return shows how analytic solutions of (2) give information that would be quite difficult to extract from numerical solutions. We believe that our analytic solutions contribute to improved understanding of quantum dynamics of

three-state systems.

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