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Realization of adiabatic population transfer in a three-level system by using LR Hermitian invariants theory*

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We have studied the population transfer in a three-state system driven by laser beams in terms of the theory of Lewis–Riesenfeld Hermitian invariants for explicitly time-dependent Hamiltonian. The exact solutions obtained allow us to recover two existing schemes producing complete population transfer in the literature, i.e. the π-pulse and stimulated Raman adiabatic passage methods in our unified description. We present a strict formulation of adiabatic conditions and a more accurate expression of the π-pulse method according to the exact time-evolution of state vector.

Keywords: adiabatic population transfer, Lewis–Riesenfeld Hermitian invariant, π-pulse method, stimulated Raman adiabatic passage methods

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1. Introduction

Transitions in atoms or molecules driven by laser beams have continued to receive considerable attention in recent years.[1−4] Two well-known procedures can, in principle, lead to complete population transfer in a three-state system involving a constant frequency. The π-pulse method makes use of the Rabi population oscillations characterizing coherent excitation.[5,6] The stimulated Raman adiabatic passage (STIRAP[7,8]) technique is used to avoid intensity-sensitive excitation (i.e. dynamical factor) and to make the state vector a null-eigenvalue adiabatic state. According to Berry's work,[9] there is only the geometric phase occurring when one designs a procedure in which a system remains in a null-eigenvalue adiabatic state. The main feature of STIRAP is that the pulses are applied to the atom in the 'counterintuitive' order: first the Stokes transition (coupling the unpopulated state 2 and state 3) and then the pump transition (coupling the initial state 1 and the state 2). These two methods have been widely investigated and generalized in many papers.[10−17]

Lewis and Riesenfeld (LR) started the investigation of the dynamics and quantization of time-dependent systems long ago with the method of Hermitian invariants.[18,19] According to LR theory, Lai and Liang[20] investigated the time evolution of a class of exactly solvable time-dependent quantum systems with a time-dependent Hamiltonian given by a linear combination of $SU(1,1)$ and $SU(2)$ generators with the help of the invariant Hermitian operator. Exact solutions of the Schrödinger equation as well as the time-evolution operator are obtained in terms of eigenstates of the invariant operator. We study the time-evolution of a three-state system for which the Hamiltonian can be expressed in terms of the generators of the $SU(2)$ algebra. Using LR theory, we firstly obtain exact solutions of the system, then we recover both the π-pulse method based on time-evolution operator of the system and the STIRAP method in terms of instantaneous eigenstates with the help of time-dependent gauge transformation.[21] Finally, adiabatic conditions of these two schemes are discussed.

2. A three-state system driven by laser beams

We consider an atomic or molecular system driven by two lasers in which the atomic transitions take

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place among only three of the many available states. The time-dependent Schrödinger equation of the system (in units of $\hbar = 1$)

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\Psi(t) = \hat{H}(t)\Psi(t) \tag{1}$$

is reduced to the simple form of a three-level system with time-dependent Hamilton operator as [22–24]

$$\hat{H}(t) = \begin{bmatrix} 0 & \Omega_1(t) & 0 \\ \Omega_1(t) & 0 & \Omega_2(t) \\ 0 & \Omega_2(t) & 0 \end{bmatrix}, \tag{2}$$

where the coupling Rabi frequencies $\Omega_{1,2}$ are generally time-dependent real quantities involving the slowly varying electric-field envelopes of the applied lasers and where the vanishing of the diagonal elements implies that frequencies of the lasers are in resonance with the frequencies of the atomic transitions.[24] Level 1 is assumed to be the ground state, and levels 2 and 3 are labelled 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 $\Omega_2(t)$ generally do not vanish.

$\hat{H}(t)$ can be written in the form

$$\hat{H}(t) = \Omega_1(t)\hat{K}_1 + \Omega_2(t)\hat{K}_2, \tag{3}$$

where

$$\hat{K}_1 = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

$$\hat{K}_2 = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix}. \tag{4}$$

In order to have the 'standard' form of the $SU(2)$ model,[14] we transform $\hat{K}_1$ and $\hat{K}_2$ into the 'standard' representation $\hat{K}_1'$ and $\hat{K}_2'$, i.e.

$$\hat{K}_1' = \frac{1}{\sqrt{2}}\begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{bmatrix},$$

$$\hat{K}_2' = \frac{1}{\sqrt{2}}\begin{bmatrix} 0 & -\mathrm{i} & 0 \\ \mathrm{i} & 0 & -\mathrm{i} \\ 0 & \mathrm{i} & 0 \end{bmatrix}, \tag{5}$$

by means of the unitary transformation

$$\hat{K}_i' = \hat{M}^\dagger \hat{K}_i \hat{M}, \tag{6}$$

where the unitary matrix is

$$\hat{M} = \begin{bmatrix} \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ 0 & 1 & 0 \\ \frac{\mathrm{i}}{\sqrt{2}} & 0 & -\frac{\mathrm{i}}{\sqrt{2}} \end{bmatrix},$$

$$\hat{M}^\dagger = \begin{bmatrix} \frac{1}{\sqrt{2}} & 0 & -\frac{\mathrm{i}}{\sqrt{2}} \\ 0 & 1 & 0 \\ \frac{1}{\sqrt{2}} & 0 & \frac{\mathrm{i}}{\sqrt{2}} \end{bmatrix}. \tag{7}$$

The transformed Hamilton operator is

$$\begin{aligned}\tilde{H}(t) =& \hat{M}^\dagger \hat{H}(t)\hat{M} \\ =& \frac{\Omega_0(t)}{2}[\hat{K}_+ \mathrm{e}^{\mathrm{i}\varphi(t)} + \hat{K}_- \mathrm{e}^{-\mathrm{i}\varphi(t)}] + 0 * \hat{K}_0, \end{aligned} \tag{8}$$

expressed in terms of the generator of the $SU(2)$ algebra in the standard form

$$\hat{K}_+ = \hat{K}_1' + \mathrm{i}\hat{K}_2',$$
$$\hat{K}_- = \hat{K}_1' - \mathrm{i}\hat{K}_2',$$

$$\hat{K}_0 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix},$$

where

$$\Omega_1(t) = \Omega_0(t)\cos[\varphi(t)], \tag{9}$$
$$\Omega_2(t) = -\Omega_0(t)\sin[\varphi(t)], \tag{10}$$

and equivalently the angle

$$\varphi(t) = -\arctan\frac{\Omega_2(t)}{\Omega_1(t)} \tag{11}$$

determined by the ratio of $\Omega_2(t)$ and $\Omega_1(t)$, which, we will see, plays an important role in the scheme to have the adiabatic population transfer besides the norm of $\Omega_1(t)$ and $\Omega_2(t)$, i.e.

$$\Omega_0(t) = \sqrt{\Omega_1^2(t) + \Omega_2^2(t)}. \tag{12}$$

The transformed states are

$$\tilde{\Psi}(t) = \hat{M}^\dagger \Psi(t), \tag{13}$$

which satisfies the Schrödinger equation

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\Psi}(t) = \tilde{H}\tilde{\Psi}(t). \tag{14}$$

3. Solutions in terms of LR theory

In Ref.[20] constructions of the invariant Hermitian operator are given for $SU(1,1)$ and $SU(2)$ systems, and it is shown that invariant operator $\hat{I}$ can be obtained from the Hermitian operator $\hat{K}_0$ with a time-dependent unitary transformation

$$\hat{I}(t) = \hat{R}(t)\hat{K}_0\hat{R}^{\dagger}(t), \tag{15}$$

where

$$\hat{R}(t) = \exp\left[\frac{\gamma(t)}{2}\left(\hat{K}_+ e^{-i\beta(t)} - \hat{K}_- e^{i\beta(t)}\right)\right]. \tag{16}$$

The auxiliary parameters $\gamma(t)$ and $\beta(t)$ are related to $\Omega_0(t)$ and $\varphi(t)$ by the following auxiliary equations respectively:

$$\dot{\gamma}(t) = \Omega_0(t)\sin(\varphi(t) + \beta(t)) \tag{17}$$

and

$$\dot{\beta}(t) = \Omega_0(t)\cot\gamma(t)\cos(\varphi(t) + \beta(t)), \tag{18}$$

derived [20] from equation $\frac{d\hat{I}}{dt} = 0$. It is easy to find a new Hamilton operator using time-dependent gauge transformation [21] with the help of unitary operator $\hat{R}$, which is given by

$$\begin{aligned}\hat{H}' =& \hat{R}^{\dagger}(t)\tilde{H}(t)\hat{R}(t) - i\hat{R}^{\dagger}(t)\frac{\partial}{\partial t}\hat{R}(t)\\ =& 2\Omega(t)\hat{K}_0,\end{aligned} \tag{19}$$

which is equivalent to $\tilde{H}(t)$, where

$$\begin{aligned}\Omega(t) =& \dot{\beta}(t)\sin^2\frac{\gamma(t)}{2}\\ &- \frac{\Omega_0(t)}{2}\sin\gamma(t)\cos(\beta(t) + \varphi(t)).\end{aligned} \tag{20}$$

The time-dependent Schrödinger equation is covariant

$$i\frac{d}{dt}|\Psi'(t)\rangle = \hat{H}'(t)|\Psi'(t)\rangle, \tag{21}$$

where

$$|\Psi'(t)\rangle = \hat{R}^{\dagger}(t)|\tilde{\Psi}(t)\rangle = \hat{R}^{\dagger}(t)\hat{M}^{\dagger}|\Psi(t)\rangle. \tag{22}$$

3.1. Time evolution of quantum states

Assume that $|n\rangle$, which is time-independent, are the eigenstates of $\hat{K}_0$ with eigenvalue K_n. The eigenstates of $\hat{I}(t)$ with the same eigenvalue K_n are obviously given by

$$|n,t\rangle = \hat{R}(t)|n\rangle. \tag{23}$$

According to Ref.[20] the general solution of the Schrödinger equation (14) is obtained as a compact form in terms of time-evolution operator

$$|\tilde{\Psi}(t)\rangle = \hat{U}(t,0)|\tilde{\Psi}(0)\rangle, \tag{24}$$

where the time-evolution operator is seen to be [20]

$$\hat{U}(t,0) = \hat{R}(t)e^{-i\epsilon(t)\hat{K}_0}\hat{R}^{\dagger}(0), \tag{25}$$

and

$$\epsilon(t) = \int_0^t 2\Omega(t')\,dt'. \tag{26}$$

We assume that only level 1 is initially populated, i.e. $|\Psi(0)\rangle = |1\rangle$, and the initial condition in $K_{\pm,0}$ representation is expressed as

$$\tilde{\Psi}(0) = \hat{M}^{\dagger}\Psi(0) = \frac{1}{\sqrt{2}}\begin{pmatrix}1\\0\\1\end{pmatrix}. \tag{27}$$

Then we obtain

$$|\tilde{\Psi}(t)\rangle = \tilde{C}_1(t)|1\rangle + \tilde{C}_2(t)|2\rangle + \tilde{C}_3(t)|3\rangle, \tag{28}$$

where

$$\begin{aligned}\tilde{C}_1(t) =& \frac{1}{\sqrt{2}}e^{-i\int_0^t\dot{\beta}(t')dt'}\left[e^{-\frac{i}{2}\delta(t)}\cos\frac{\gamma(t)}{2}\cos\frac{\gamma(0)}{2} + e^{\frac{i}{2}\delta(t)}\sin\frac{\gamma(t)}{2}\sin\frac{\gamma(0)}{2}\right]^2\\ &+ \frac{1}{\sqrt{2}}e^{-i\beta(0)-i\beta(t)}\left[e^{-\frac{i}{2}\delta(t)}\cos\frac{\gamma(t)}{2}\sin\frac{\gamma(0)}{2} - e^{\frac{i}{2}\delta(t)}\sin\frac{\gamma(t)}{2}\cos\frac{\gamma(0)}{2}\right]^2,\end{aligned} \tag{29}$$

$$\begin{aligned}\tilde{C}_2(t) =& i\Big\{\sin\beta(0)\sin\gamma(0)\cos\gamma(t) + \sin[\delta(t) - \beta(0)]\sin\gamma(t)\cos^2\frac{\gamma(0)}{2}\\ &+ \sin[\delta(t) + \beta(0)]\sin\gamma(t)\sin^2\frac{\gamma(0)}{2}\Big\},\end{aligned} \tag{30}$$

$$\begin{aligned}\tilde{C}_3(t) =& \frac{1}{\sqrt{2}}e^{i\int_0^t\dot{\beta}(t')dt'}\left[e^{\frac{i}{2}\delta(t)}\cos\frac{\gamma(t)}{2}\cos\frac{\gamma(0)}{2} + e^{-\frac{i}{2}\delta(t)}\sin\frac{\gamma(t)}{2}\sin\frac{\gamma(0)}{2}\right]^2\\ &+ \frac{1}{\sqrt{2}}e^{i\beta(0)+i\beta(t)}\left[e^{\frac{i}{2}\delta(t)}\cos\frac{\gamma(t)}{2}\sin\frac{\gamma(0)}{2} - e^{-\frac{i}{2}\delta(t)}\sin\frac{\gamma(t)}{2}\cos\frac{\gamma(0)}{2}\right]^2;\end{aligned} \tag{31}$$

and

$$\delta(t) = \int_0^t [2\Omega(t') - \dot{\beta}(t')]\mathrm{d}t'$$
$$= -\int_0^t [\dot{\beta}(t')\cos\gamma(t') + \Omega_0(t')\sin\gamma(t')\cos(\beta(t') + \varphi(t'))]\mathrm{d}t' \quad (32)$$

is the pulse area, which is the key quantity in the π-pulse scheme. It is easy to verify from Eqs.(29)–(31) that the initial condition (27) is satisfied. When the parameters in Eqs.(29)–(32) are adjusted such that

$$\tilde{C}_1(t) = -\frac{\mathrm{i}}{\sqrt{2}},$$
$$\tilde{C}_2(t) = 0$$
$$\tilde{C}_3(t) = \frac{\mathrm{i}}{\sqrt{2}}, \quad (33)$$

we then achieve the complete population transfer $|\Psi(t)\rangle = |3\rangle$ according to Eq.(13), since

$$\Psi(t) = \hat{M}\widetilde{\Psi}(t) = \hat{M}\begin{pmatrix} -\frac{\mathrm{i}}{\sqrt{2}} \\ 0 \\ \frac{\mathrm{i}}{\sqrt{2}} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \quad (34)$$

In the following our main goal is to demonstrate the explicit scheme that the condition of complete population transfer Eq.(33) can be fulfilled.

3.2. Instantaneous eigenstates

The π-pulse method that relies on Rabi oscillations requires careful control of pulse shape, as parametrized by the pulse area. Experimental realization of complete population transfer is very difficult. In order to achieve the complete population transfer that is independent of pulse area, i.e. the dynamical phase, we need to devise a procedure in which only the geometric phase occurs. To avoid any dynamical phase, we make the state vector a null-eigenvalue state. This idea can be realized in the gauge of diagonalized Hamiltonian $\hat{H}'(t)$, the basis vectors of which are $|\Psi'_+(t)\rangle$, $|\Psi'_0(t)\rangle$ and $|\Psi'_-(t)\rangle$, corresponding to the eigenvalues $\{E_+(t), E_0(t), E_-(t)\}$ respectively, where

$$E_+(t) = 2\Omega(t), \quad (35)$$
$$E_0(t) = 0,$$
$$E_-(t) = -2\Omega(t). \quad (36)$$

$|\Psi'_0(t)\rangle$ is the null-eigenvalue state desired, which satisfies

$$\hat{H}'(t)|\Psi'_0(t)\rangle = 0. \quad (37)$$

We initially prepare the system as a null-eigenvalue state $|\Psi'_0(0)\rangle$ and from Eqs.(22), (24) and (25) we obtain

$$|\tilde{\Psi}_0(0)\rangle = \hat{R}(0)|\Psi'_0(0)\rangle, \quad (38)$$

and the null-eigenvalue instantaneous eigenstate (noticing that the null-eigenvalue is $\hat{K}_0|\Psi'_0\rangle = 0$)

$$|\Psi_0(t)\rangle = \hat{M}\hat{U}(t,0)\hat{R}(0)|\Psi'_0(0)\rangle$$
$$= \hat{M}\hat{R}(t)|\Psi'_0(0)\rangle, \quad (39)$$

which is expressed as

$$|\Psi_0(t)\rangle = \mathrm{i}\sin\gamma(t)\sin\beta(t)|1\rangle + \cos\gamma(t)|2\rangle$$
$$-\mathrm{i}\sin\gamma(t)\cos\beta(t)|3\rangle. \quad (40)$$

This is a state without a dynamic phase, and our task is to adjust the time-dependent parameters to realize population transfer, i.e. to make

$$|\langle 1|\Psi_0(t)\rangle| = 0,$$
$$|\langle 2|\Psi_0(t)\rangle| = 0, \quad (41)$$

and

$$|\langle 3|\Psi_0(t)\rangle| = 1, \quad (42)$$

in order to achieve the complete population transfer.

4. Adiabatic approximation

We now discuss the explicit conditions to achieve complete population transfer, i.e. to have Eqs.(33), (41) and (42) be satisfied separately. We consider here the adiabatic approximation in which the terms containing time derivatives $\dot{\gamma}$ and $\dot{\beta}$ in the auxiliary equations (17) and (18) can be neglected.

If the auxiliary parameters $\gamma(t)$ and $\beta(t)$ vary with time slowly enough, i.e.

$$\dot{\gamma}(t) \simeq 0, \qquad \dot{\beta}(t) \simeq 0, \quad (43)$$

we have [20]

$$\beta(t) = 2n\pi - \varphi(t),$$
$$\tan\gamma(t) \to \infty. \quad (44)$$

Then we assume

$$\gamma(t) = \frac{\pi}{2}. \tag{45}$$

4.1. Scheme A: modified π-pulse method

Under the approximation of Eqs.(44) and (45), the simplified form of $\tilde{C}_2(t)$ is seen to be

$$\tilde{C}_2(t) = -\mathrm{i}\cos\beta(0)\sin\delta(t), \tag{46}$$

with the pulse area (Eq.(32)) reducing to

$$\delta(t) = \int_0^t \Omega_0(t')\,\mathrm{d}t'. \tag{47}$$

When $\delta(t) = n\pi$, we have $\tilde{C}_2(t) = 0$, and

$$\begin{aligned}\tilde{C}_1(t) =& \frac{1}{2\sqrt{2}}\mathrm{e}^{\mathrm{i}\beta(0)-\mathrm{i}\beta(t)}\left[1+\cos\delta(t)\right] \\ &- \frac{1}{2\sqrt{2}}\mathrm{e}^{-\mathrm{i}\beta(0)-\mathrm{i}\beta(t)}\left[1-\cos\delta(t)\right], \quad (48)\\ \tilde{C}_3(t) =& \frac{1}{2\sqrt{2}}\mathrm{e}^{-\mathrm{i}\beta(0)+\mathrm{i}\beta(t)}\left[1+\cos\delta(t)\right] \\ &- \frac{1}{2\sqrt{2}}\mathrm{e}^{\mathrm{i}\beta(0)+\mathrm{i}\beta(t)}\left[1-\cos\delta(t)\right]. \quad (49)\end{aligned}$$

If the pulse area is adjusted such that

$$\delta(t) = 2n\pi, \tag{50}$$

we have

$$\begin{aligned}\tilde{C}_1(t) &= \frac{1}{\sqrt{2}}\mathrm{e}^{\mathrm{i}\beta(0)-\mathrm{i}\beta(t)}, \\ \tilde{C}_3(t) &= \frac{1}{\sqrt{2}}\mathrm{e}^{-\mathrm{i}\beta(0)+\mathrm{i}\beta(t)}. \end{aligned}\tag{51}$$

We assume the order of laser beams to be: intuitive order $\left(\beta(t)-\beta(0) = -\frac{\pi}{2}\right)$, whereas counterintuitive order $\left(\beta(t)-\beta(0) = \frac{\pi}{2}\right)$,[14,15] then

$$\begin{aligned}\tilde{C}_1(t) &= \frac{\mathrm{i}}{\sqrt{2}}, \\ \tilde{C}_3(t) &= \frac{-\mathrm{i}}{\sqrt{2}}, \end{aligned}\tag{52}$$

for the intuitive order; but

$$\begin{aligned}\tilde{C}_1(t) &= \frac{-\mathrm{i}}{\sqrt{2}}, \\ \tilde{C}_3(t) &= \frac{\mathrm{i}}{\sqrt{2}}, \end{aligned}\tag{53}$$

for the counterintuitive order. In both cases the complete population transfer Eq.(34) is achieved.

For the case

$$\delta(t) = (2n+1)\pi, \tag{54}$$

the state coefficients are

$$\begin{aligned}\tilde{C}_1(t) &= -\frac{1}{\sqrt{2}}\mathrm{e}^{-\mathrm{i}\beta(0)-\mathrm{i}\beta(t)}, \\ \tilde{C}_3(t) &= -\frac{1}{\sqrt{2}}\mathrm{e}^{\mathrm{i}\beta(0)+\mathrm{i}\beta(t)}. \end{aligned}\tag{55}$$

For both the intuitive order with $\beta(t) = -\frac{\pi}{2}$, $\beta(0) = 0$, and the counterintuitive order with $\beta(t) = 0$, $\beta(0) = -\frac{\pi}{2}$, we have

$$\begin{aligned}\tilde{C}_1(t) &= -\frac{\mathrm{i}}{\sqrt{2}}, \\ \tilde{C}_3(t) &= \frac{\mathrm{i}}{\sqrt{2}}. \end{aligned}\tag{56}$$

Summing up all these situations, we obtain what we call the 'modified π -pulse method', which includes two conditions to achieve complete population transfer: (a) the pulse area Eq.(47) should be controlled to have $\delta(t) = n\pi$, and (b)

$$\begin{aligned}\beta(t)-\beta(0) =& \varphi(0)-\varphi(t) \\ =& \arctan\frac{\Omega_2(0)}{\Omega_1(0)} - \arctan\frac{\Omega_2(t)}{\Omega_1(t)} \\ =& \pm\frac{\pi}{2}, \end{aligned}\tag{57}$$

obtained from Eqs.(11) and (45).

This is different from the 'π-pulse method',[5,6] which demands that the pulse area be equal to π or any odd-multiple of π only. Our condition shows that not only the pulse area Eq.(47) is determined mainly by the norm Ω_0, but also the ratio $\frac{\Omega_2}{\Omega_1}$ plays a role in the scheme of adiabatic transfer.

4.2. Scheme B: STIRAP

Our starting point of the STIRAP scheme is the null-eigenvalue instantaneous eigenstate Eq.(40) which, under adiabatic approximation, is reduced to

$$|\Psi_0(t)\rangle = -\mathrm{i}\sin\beta(t)\,|1\rangle + \mathrm{i}\cos\beta(t)\,|3\rangle. \tag{58}$$

If we assume the counterintuitive pulse sequence in this scheme to be

$$\beta(0) = -\arctan\frac{\Omega_2(0)}{\Omega_1(0)} = -\frac{\pi}{2} \tag{59}$$

and

$$\beta(t) = -\arctan\frac{\Omega_2(t)}{\Omega_1(t)} \simeq 0, \tag{60}$$

we obtain obviously complete population transfer, i.e. at initial time ($t = 0$), $|\Psi_0(0)\rangle = i\,|1\rangle$; and at time t, $|\Psi_0(t)\rangle = \mathrm{i}\,|3\rangle$. The population transfer can

be asymptotically achieved if the ratio $\dfrac{\Omega_2(t)}{\Omega_1(t)}$ is very large initially and then becomes small enough after a long time of adiabatic control.

The mechanism to produce population transfer in our case is the same as that in STIRAP—the pulses are applied to the atom in the 'counterintuitive' order: the transition occurs between the two unpopulated states 2 and 3 firstly and then between the initial state 1 and state 2.[14] The adiabatic conditions (Eq.(44) and subsequently Eq.(45)) are necessary for obtaining the complete population transfer.

4.3. Conditions of population transfer in comparison with the original π-pulse and the STIRAP schemes

For both of the above two schemes, the adiabatic condition Eq.(44) is required. We use

$$\dot{\beta}\,(t) = -\dot{\varphi}\,(t) = -\frac{\mathrm{d}}{\mathrm{d}t}\left(\arctan\frac{\Omega_2(t)}{\Omega_1(t)}\right) \simeq 0, \qquad (61)$$

along with Eq.(45) to produce the transfer. Because of the very small variation rate of Eq.(61) the interaction time must be long enough so that we can have the finite angle change Eq.(57). In other words, the adiabatic time-evolution is required in the modified π-pulse method based on our exact solution and the population transfer can be obtained only asymptotically, while the original π-pulse method, in which the only condition is the pulse area being an odd-multiple of π, is not an adiabatic process. The conditions in the STIRAP scheme discussed in Ref.[14] are given by

$$\frac{1}{2}\Omega_0 t \gg 1, \qquad (62)$$

which can be approached by increasing Ω_0 in a finite time period t, or in other words, the complete population transfer can be achieved with pulses strong enough even for a short pulse duration. This is very different from our adiabatic conditions (Eqs.(57) and (61)), which require a long interaction time.

5. Conclusion

We have studied the time evolution of a three-state system in terms of exactly solvable time-dependent Hamiltonian given by a linear combination of $SU(2)$ generators with the help of the invariant Hermitian operator. With the exact solutions we recover the both schemes producing complete population transfer under different initial conditions: for the modified π-pulse method the system is prepared initially at ground state, and for the STIRAP method the system is maintained at a special instantaneous eigenstate of null-eigenvalue. The key point for the modified π-pulse method to have a complete population transfer is to control the pulse area and the phase of laser beams at the same time, which can be achieved in the adiabatic approximations resulting from exact solutions. In the STIRAP scheme, we apply the counterintuitive pulse order to drive the system under the same adiabatic condition as in the π-pulse method. The STIRAP scheme is experimentally more robust since the difficult procedure to adjust the pulse area is avoided. In these two schemes the strict adiabatic conditions $\gamma = \dfrac{\pi}{2}$ and $\dot{\beta}(t) \to 0$ (namely, Eq.(61)) with a long pulse duration ($t \to \infty$) should be satisfied.

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