Population transfer in multilevel system through modified stimulated Raman adiabatic passage

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Stimulated Raman adiabatic passage (STIRAP) has been successfully extended to multilevel system. During the STIRAP process, the intermediate levels have notable population which is detrimental if these levels could decay to other levels through spontaneous emission. This paper proposes a novel method to reduce the intermediate level population during the STIRAP process. A complete population transfer can be achieved in this modified STIRAP even if the intermediate level decays to other levels. OCIS codes: 020.0020, 020.1670.

Stimulated Raman adiabatic passage (STIRAP) is an efficient and robust technique for population transfer between states and has been extensively studied both experimentally and theoretically for years^[1]. STIRAP can be applied in many fields, such as laser $\operatorname{cooling}^{[2]}$, atom optics^[3], quantum computing^[4], etc.. Atoms and molecules prepared in specified quantum state can be realized by the π -pulse technique, STIRAP^[1,5], the chirped pulse method^[6,7], amplitude modulated method^[8] and so on. The technique of STIRAP robustly allows a complete population transfer from an initial single state to the target state in N-level system^[9]. But the electronic radiation loss is not considered in the Hamiltonian in Ref. [9], the transfer population is very high. Contrarily, the population in the target state is very low if one includes the radiant loss of the intermediate states. The way to solve this problem is to reduce the population of the intermediate states in the adiabatic passage, and the local optimization technique overcame the problem^[10]. By requiring the intermediate level population to be zero through adiabatic passage, one can numerically get the laser pulse orders and pulse intensities. This method requires control of pulse intensities and delay times. In this paper, we propose a novel method to solve this problem by adjusting the order of laser pulses only. Our method is simple and even more robust, one only has to control the delay time of the laser pulse.

The five level system is shown in Fig. 1. Our purpose is to transfer from level 1 to level 5. Levels 1 and 2, 3 and 4 are respectively coupled by the pump laser pulses



Fig. 1. Five-level system, $\Delta_{\rm p1} = \omega_2 - \omega_1 - \omega_{\rm p1}$, $\Delta_{\rm p2} = \omega_4 - \omega_3 - \omega_{\rm p2}$, $\hbar\omega_i$ (i = 1 - 5) describes the energy of level $|i\rangle$, and ω_j $(j = {\rm p1}, {\rm p1}, {\rm s1}, {\rm s2})$ is the carrier frequency of the corresponding laser.

 $\Omega_{p1}(t)$ and $\Omega_{p2}(t)$; levels 2 and 3, 4 and 5 are respectively coupled by the Stokes laser pulses $\Omega_{s1}(t)$ and $\Omega_{s2}(t)$. The wave functions of the bare states are denoted by $|1\rangle$, $|2\rangle$, $|3\rangle$, $|4\rangle$ and $|5\rangle$. The time evolution of the probability amplitudes $C(t) = [C_1(t), C_2(t), C_3(t), C_4(t), C_5(t)]^T$ of the five states is described by the Schödinger equation

$$ih\frac{\mathrm{d}}{\mathrm{d}t}C(t) = H(t)C(t). \tag{1}$$

Since $A_{\rm p} = A_{\rm p0} \cos(\omega_{\rm p} t - k_{\rm p} x + \phi_{\rm p})$ and $A_{\rm si} = A_{\rm si0} \cos(\omega_{\rm s} t - k_{\rm s} x + \phi_{\rm si})$, the Hamiltonian reads

$$H(t) = \frac{h}{2} \begin{bmatrix} 0 & \Omega_{\rm p1}(t) & 0 & 0 & 0\\ \Omega_{\rm p1}(t) & 2V_{\rm p1} & \Omega_{\rm s1}(t) & 0 & 0\\ 0 & \Omega_{\rm s1}(t) & 0 & \Omega_{\rm p2}(t) & 0\\ 0 & 0 & \Omega_{\rm p2}(t) & 2V_{\rm p2} & \Omega_{\rm s2}(t)\\ 0 & 0 & 0 & \Omega_{\rm s2}(t) & 0 \end{bmatrix}.$$

$$(2)$$

The adiabatic state with null eigenvalue (it is on twophoton resonance, i.e., $\Delta_{p1} - \Delta_{s1} = 0$, $\Delta_{p2} - \Delta_{s2} = 0$) is

$$|\psi\rangle = \frac{\Omega_{s1}\Omega_{s2}}{\Omega'}|1\rangle - \frac{\Omega_{p1}\Omega_{s2}}{\Omega'}|3\rangle + \frac{\Omega_{p1}\Omega_{p2}}{\Omega'}|5\rangle, \quad (3)$$

$$\Omega' = \sqrt{\Omega_{s1}^2 \Omega_{s2}^2 + \Omega_{p1}^2 \Omega_{s2}^2 + \Omega_{p1}^2 \Omega_{p2}^2}.$$
 (4)

In traditional scheme, both of the Stokes pulses precede the two pump lasers by 0.2T with T being the period of the pulse.

In Fig. 2, when t = t1, $\Omega_{\text{p1,p2}} = \Omega_{\text{s1,s2}}$, the population of $|3\rangle$ is 33%. The radiant loss of level 3 could reduce the population transfer efficiency. How can we reduce this transient intermediate state population? Through optimal control theory (OCT), Malinovsky *et al.* proposed a pulse order which greatly reduces the transient intermediate population transfer^[10]. But this method requires control of pulse intensities (some pulse intensities should be ten times larger than the other intensities). Here, we propose a simple and robust way to reduce transient intermediate population. We only adjust the delay time of laser to reduce the population of the intermediate states. The population of $|3\rangle$ is $|c_3|^2 = \frac{1}{\frac{\Omega_{s1}^2}{\Omega_{s2}^2} + 1 + \frac{\Omega_{s2}^2}{\Omega_{s2}^2}}$. We can adjust

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Fig. 2. Traditional time evolution of pulses in five level system.



Fig. 3. (a) Time evolution of pulses and (b) time evolution of populations produced by these pulses. Populations of excited states $|2\rangle$ and $|4\rangle$ remain zero.

the pulses to make $\frac{\Omega_{s1}^2}{\Omega_{p1}^2} + 1 + \frac{\Omega_{p2}^2}{\Omega_{s2}^2} \gg 1$ at any crossing. To illustrate the preceding analytic solutions, we present numerical simulation in the case of the pulses with the same duration T (T = 170 ns, the period of the pulse) for the pump and Stokes lasers $\Omega_{pi}(t) = \Omega_0 \sin^4(\pi t/T)$ and $\Omega_{si}(t) = \Omega_0 \sin^4(\pi t/T)$ ($i = 1, 2; \Omega_0 = 7$ GHz). Using the STIRAP technique, the Stokes pulse precedes the corresponding pump pulse by 0.2T; the second pump laser precedes the first one by 0.35T, then the second Stokes laser also precedes the first one by 0.35T. Obviously, the adiabatic condition $\Omega T \gg 1$ is satisfied, the dark states will not transfer to other dressed states.

In Fig. 3(a) at t1, t2, t3, t4 and t5, we can see $\frac{\Omega_{s1}^2}{\Omega_{p1}^2} + 1 + \frac{\Omega_{p2}^2}{\Omega_{s2}^2} \gg 1$ (i.e., $|c_3|^2$ is nearly zero) through simple calculation. In the simulation, the population of $|3\rangle$ is only 1.9% versus 33% in the traditional order, as shown in Fig. 3(b).

From Eqs. (3) and (4), for adjusting pump and Stokes lasers, the relation $\frac{\Omega_{p1}(t)\Omega_{p2}(t)}{\Omega'(t)}$ is applied. As time progresses from $-\infty$ to ∞ , the adiabatic state $|\psi\rangle$ starting in the bare state $|1\rangle$ will end in the target state $|5\rangle$. In the adiabatic passage, the denominator of expression $|c_3|^2$ (the population of $|3\rangle$) is much larger than 1, so the population of $|3\rangle$ is very small.

In the seven level system, levels 5 and 6, 6 and 7 are respectively coupled by the pump laser $\Omega_{p3}(t)$ and Stokes laser $\Omega_{s3}(t)$ in the foundation of five level system. We insert one pair of Stokes and pump pulses between two pairs of those, i.e., the third pump laser $(\Omega_{p3}(t))$ precedes the first one $(\Omega_{p1}(t))$ by 0.35*T* and the second one $(\Omega_{p2}(t))$ precedes the first one $(\Omega_{p1}(t))$ by 0.18*T*. Each pair of pulses are in STIRAP order, i.e., each Stokes pulse precedes the corresponding pump pulse by 0.2*T*. In the traditional order, the population of $|3\rangle$ and $|5\rangle$ is almost 28% versus 1.8% (see Fig. 4) in the new scheme.

In the nine level system, levels 7 and 8, 8 and 9 are respectively coupled by the pump laser $\Omega_{p4}(t)$ and Stokes laser $\Omega_{s4}(t)$ in the foundation of seven level system. The fourth pump laser ($\Omega_{p4}(t)$) precedes the first one ($\Omega_{p1}(t)$) by 0.35*T*, both of the second ($\Omega_{p4}(t)$) and third ($\Omega_{p3}(t)$) ones precede the first one by 0.18*T*. Each pair of pulses are in STIRAP order, i.e., each Stokes pulse precedes the corresponding pump pulse by 0.2*T*. In the new scheme the populations of $|3\rangle$, $|5\rangle$, and $|7\rangle$ are respectively 1.8% versus 26% (see Fig. 5), 1.5% versus 19%, and 3.5% versus 25% in the traditional scheme.

We have explored analytically and numerically to reduce the population of the intermediate in N-level systems to realize complete population transfer from the initial state to the target state. By controlling the pulse delay time between the pairs of the pump and Stokes pulses properly, the population of the intermediate states is reduced to be very low in the adiabatic passage. In order to achieve this purpose, the usual adiabatic condition for STIRAP should be satisfied. In addition, one should keep each Stokes laser precede the corresponding pump laser by 0.2T.



Fig. 4. Time evolution of populations produced by the three pairs of Stokes and pump pulses. Populations of excited states $|2\rangle$, $|4\rangle$, and $|6\rangle$ remain zero.



Fig. 5. Time evolution of populations produced by the four pairs of Stokes and pump pulses.

In brief, our scheme demonstrates the possibility of reducing the population of the intermediate states to realize complete population transfer from the initial state to the target state with two or even more than two intermediate states systems.

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