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Population transfer and coherence in the adiabatic limit by counterintuitive and intuitive pulse sequences*

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Two approaches of achieving population transfer and coherence are investigated for the three-level Λ system in the adiabatic limit. The effects of the laser pulse sequence on the population transfer efficiency and coherence are studied. Coherent control of quantum state and population is studied by numerical simulation based on self-consistent set of density matrix equations. It can be seen that the counterintuitive pulse sequence is more efficient in population transfer and coherence than the intuitive one.

Keywords: population transfer, quantum coherence, three-level Λ system, adiabatic
PACC: 9720T, 4225K, 3280B, 4250

1. Introduction

There has been an increasing interest in target population transfer[1,2] and quantum coherence[3,4] over the past decades due to their potential applications to chemical-reaction dynamics,[5] lasing without inversion (LWI),[6] electromagnetically induced transparency (EIT),[7−9] quantum computation[10] and spontaneous emission reduction and cancellation.[11,12]

The stimulated Raman adiabatic passage process (STIRAP) has recently been intensively studied.[13,14] In a three-level Λ system as shown in Fig.1, STIRAP indicates that the Stokes pulse $\Omega_{\rm s}$ that drives the transition between the initially unpopulated levels $|a\rangle$ and $|c\rangle$, precedes the pump pulse $\Omega_{\rm p}$ that drives the transition between the initially populated level $|b\rangle$ and the intermediate level $|a\rangle$. Additionally, the Stokes and pump pulses must be partially overlapped. This is the so-called counterintuitive pulse sequence, in contrast to the intuitive one in which the pump pulse $\Omega_{\rm p}$ precedes the Stokes pulse $\Omega_{\rm s}$.

In this paper, we demonstrate the effects of the laser pulse sequences on the population transfer efficiency[15] and coherence in a three-level atomic system. In Section 2, we present the basic equations. Section 3 describes the numerical simulation of population transfer and coherence in the adiabatic limit. Discussion and a brief summary are given in Section 4.

2. Theoretical basis

Consider the three-level Λ system shown schematically in Fig.1. Levels $|a\rangle$ and $|b\rangle$ are coupled by the pump laser pulse $\Omega_{\rm p}(t)$, levels $|a\rangle$ and $|c\rangle$ are coupled by the Stokes laser pulse $\Omega_{\rm s}(t)$. The transition between levels $|b\rangle$ and $|c\rangle$ is electric dipole forbidden. Two-photon resonance between levels $|b\rangle$ and $|c\rangle$ is maintained, while level $|a\rangle$ can be off resonance by a certain detuning Δ. $\Delta_{\rm p}$ and $\Delta_{\rm s}$ are the detunings of the pump and Stokes laser pulses, respectively. The laser pulse widths are supposed to be short compared to the lifetime of level $|a\rangle$.

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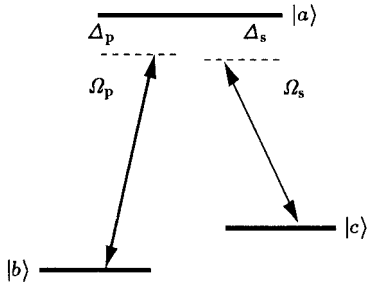


Fig.1. Schematic of the three-level Λ system.

The interaction Hamiltonian for the three-level system using the rotating wave approximation (RWA) can be written as

$$H_{\rm I} = -\frac{\hbar}{2}\begin{pmatrix} -2\Delta_{\rm p} & \Omega_{\rm p} & \Omega_{\rm s} \\ \Omega_{\rm p}^* & 0 & 0 \\ \Omega_{\rm s}^* & 0 & -2(\Delta_{\rm p}-\Delta_{\rm s}) \end{pmatrix}. \quad (1)$$

Consider the simple case of two-photon (Raman) resonance $\delta = \Delta_{\rm p} - \Delta_{\rm s} = 0$; the eigenvalues of the interaction Hamiltonian expressed by Eq.(1) are

$$\lambda_0 = 0, \quad \lambda_\pm = -\Delta_{\rm p} \pm \sqrt{\Delta_{\rm p}^2 + \Omega_{\rm p}^2 + \Omega_{\rm s}^2}. \quad (2)$$

It is straightforward to find the corresponding eigenstates of Eq.(1); the eigenfunction can be the linear combinations of the bare states $|a\rangle$, $|b\rangle$ and $|c\rangle$:

$$|\psi_0\rangle = \cos\theta|b\rangle - \sin\theta|c\rangle, \quad (3a)$$

$$|\psi_+\rangle = \cos\phi|a\rangle + \sin\phi\sin\theta|b\rangle + \sin\phi\cos\theta|c\rangle, \quad (3b)$$

$$|\psi_-\rangle = -\sin\phi|a\rangle + \cos\phi\sin\theta|b\rangle + \cos\phi\cos\theta|c\rangle; \quad (3c)$$

the angles θ and ϕ are determined by

$$\tan\theta = \frac{\Omega_{\rm p}}{\Omega_{\rm s}} \quad (4)$$

and

$$\tan 2\phi = -\frac{\sqrt{\Omega_{\rm p}^2 + \Omega_{\rm s}^2}}{\Delta_{\rm p}}, \quad (5)$$

where we have assumed Rabi frequencies $\Omega_{\rm p,s}$ real. If we initially prepare the system to be in one of the dressed states expressed by Eq.(3), and maintain the adiabatic condition at subsequent time, then the system remains in this state.

3. Numerical simulation of population transfer and coherence

In this section, we present numerical calculations of population transfer and coherence by counterintuitive and intuitive pulse sequences. To this end, we use two Gaussian pulses of different shapes separated by a pulse delay, as expressed by

$$\Omega_{\rm p}(t) = 3 \times {\rm e}^{-(\frac{t-\tau_{\rm p}}{T_{\rm p}})^2}, \quad \Omega_{\rm s}(t) = 3 \times {\rm e}^{-(\frac{t-\tau_{\rm s}}{T_{\rm s}})^2}, \quad (6)$$

where $\Omega_{\rm p}(t)$ and $\Omega_{\rm s}(t)$ are Rabi frequencies of the pump pulse and Stokes pulse respectively; $T_{\rm p}$ and $T_{\rm s}$ are the widths of the Gaussian pulses $\Omega_{\rm p}(t)$ and $\Omega_{\rm s}(t)$ respectively, $\tau_{\rm p}$ and $\tau_{\rm s}$ are their time delays. When $\tau_{\rm s} < \tau_{\rm p}$, $\Omega_{\rm s}(t)$ precedes $\Omega_{\rm p}(t)$, and this is the case of the counterintuitive pulse sequence; otherwise ($\tau_{\rm s} > \tau_{\rm p}$,$\Omega_{\rm p}(t)$ precedes $\Omega_{\rm s}(t)$), for the intuitive pulse sequence.

Next, we discuss the population transfer and coherence mechanisms for the two pulse sequences in the adiabatic limit. The adiabatic passage is based on the assumption that the Rabi frequencies $\Omega_{\rm s}(t)$ and $\Omega_{\rm p}(t)$ vary slowly with time, such that $|\dot{\theta}| \ll |\lambda_0 - \lambda_\pm|$.

3.1. Counterintuitive pulse sequence

The population transfer by the pulses in counterintuitive sequence, referred to as STIRAP,[1,16] exploits the existence of the adiabatic state $|\psi_0\rangle$ which is a time-dependent linear combination of states $|b\rangle$ and $|c\rangle$ only. The counterintuitive sequence means that

$$\lim_{t\to-\infty}\frac{\Omega_{\rm p}(t)}{\Omega_{\rm s}(t)} = 0, \quad \lim_{t\to+\infty}\frac{\Omega_{\rm p}(t)}{\Omega_{\rm s}(t)} = \infty, \quad (7)$$

or, in other words,

$$\theta(-\infty) = 0, \quad \theta(+\infty) = \frac{\pi}{2}.$$

Hence the adiabatic state $|\psi_0\rangle$ is equivalent to state $|b\rangle$ before excitation, and to state $|c\rangle$ after it, so that among the adiabatic states only state $|\psi_0\rangle$ is initially populated. If the excitation is adiabatic, the system will remain in this adiabatic state all the time, and eventually, the population will be completely transferred to state $|c\rangle$.

Figure 2 shows the numerical simulations of the control of population transfer and coherence by using the STIRAP technique in a three-level Λ system.

A remarkable characteristic of the three-level system with two-photon resonance is that the adiabatic state $|\psi_0\rangle$ does not involve the intermediate state $|a\rangle$. This implies that if the evolution is nearly adiabatic, the population of level $|a\rangle$ will remain negligible throughout the excitation. And thus, the specific properties of state $|a\rangle$, including a possible decay to other states, does not substantially affect the efficiency

of STIRAP. Secondly, the transfer efficiency does not depend on the single-photon detuning Δ, but rather on the two-photon detuning as shown below. Thirdly, it takes a great advantage of the fact that adiabatic population transfer is insensitive to the exact values of pulse areas and pulse shape,[17] which implies a vanishing small probability for the population of the unstable state $|\psi_+\rangle$ and $|\psi_-\rangle$. It is only through the decay of these unstable states that the population could leak out of the three-level system into other levels.

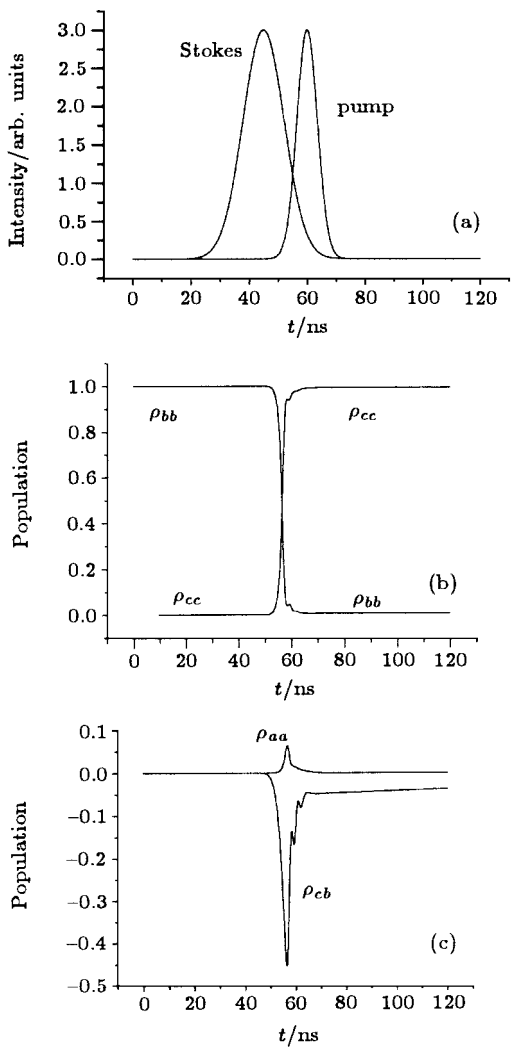


Fig.2. Numerical simulation of the control of population transfer and coherence using STIRAP. (a) The input Gaussian envelopes of the Stokes and the pump pulses with parameters τ_p=60ns, τ_s=45ns, $\tau = 1/\gamma$=1ns, T_p=5ns and T_s=10ns. (b) Population transfer between the ground states $|b\rangle$ and $|c\rangle$. (c) Transient population in the excited state $|a\rangle$ and coherence of ground states.

Figure 3 shows the population transfer efficiency as a function of two-photon detuning $(\Delta_p - \Delta_s)$, indicating that the population transfer efficiency strongly depends on the two-photon detuning. The transfer efficiency reaches its maximum at the two-photon resonance.

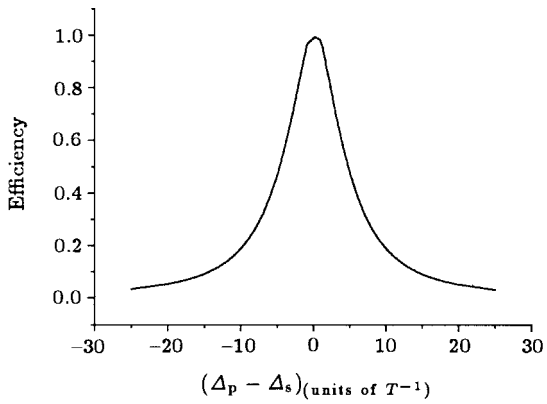


Fig.3. Numerically calculated transfer efficiency in stimulated Raman adiabatic passage plotted against the difference of the pump and Stokes detunings for the Gaussian pulse shape.

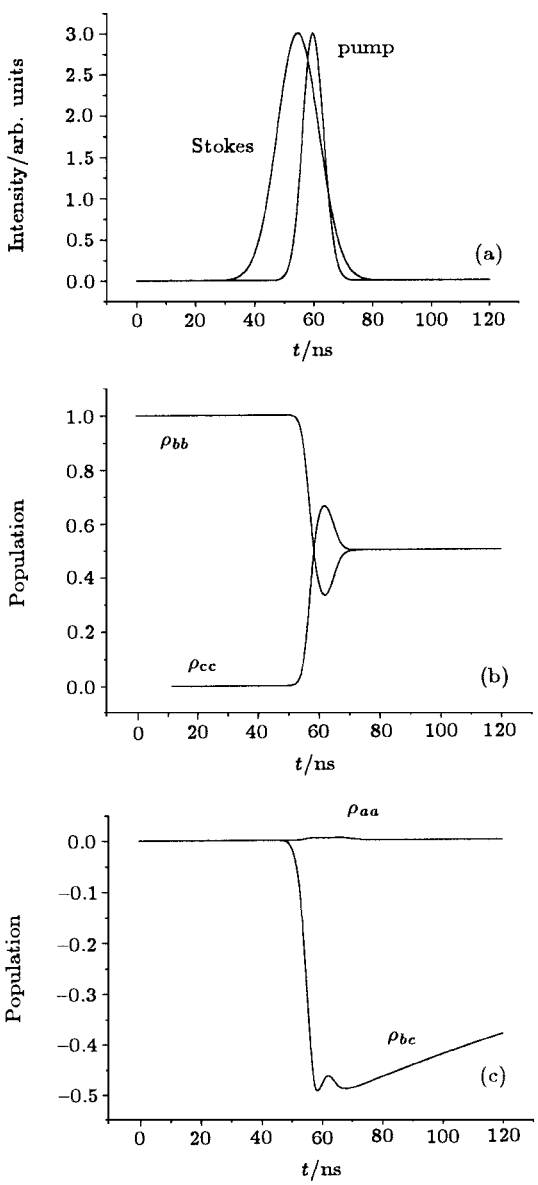


Fig.4. Numerical simulation of the control of coherence using partial STIRAP. The parameters are the same as in Fig.2 but τ_s=54.9ns.

Next, we present the numerical simulation shown in Fig.4 for the case of maximal atomic coherence by the technique of fractional STIRAP with Stokes pulse preceding pump pulse. The only difference from Fig.2 is the time delay between pulses of the pump and Stokes as shown in Fig.4(a), but in this case, we obtain very different results from the case of STIRAP for population transfer and coherence. Population transfer is not complete as shown in Fig.4(b). The states $|b\rangle$ and $|c\rangle$ are half/half populated, i.e $\rho_{bb} = \rho_{cc} = 0.5$; using the relation $|\rho_{bc}| \leq \sqrt{\rho_{bb}\rho_{cc}}$, one obtains $|\rho_{bc}| = 0.5$. The coherence term ρ_{bc}, as illustrated in the lower part of Fig.4(c), reaches its maximum value in an interval of time when the system is driven into trapping states. The residence of transient population in level $|a\rangle$ is few, as shown in the upper part of Fig.4(c).

Using counterintuitive pulse sequence, we have realized complete population transfer and maximal coherence by different time delay between pulses of the pump and Stokes. Next, we use intuitive pulse sequence to see how to realize complete population transfer and maximal coherence, also some differences between counterintuitive and intuitive pulse sequences.

3.2. Intuitive pulse sequence

Consider now the intuitive pulse sequence in which the pump pulse $\Omega_{\rm p}(t)$ precedes the Stokes pulse $\Omega_{\rm s}(t)$. In other words,

$$\lim_{t\to-\infty}\frac{\Omega_{\rm p}(t)}{\Omega_{\rm s}(t)} = \infty, \quad \lim_{t\to+\infty}\frac{\Omega_{\rm p}(t)}{\Omega_{\rm s}(t)} = 0; \tag{8}$$

and this means that

$$\theta(-\infty) = \frac{\pi}{2}, \quad \theta(+\infty) = 0.$$

Unlike the counterintuitive pulse case, there is a substantial difference between the cases of $\Delta_{\rm p} = \Delta_{\rm s} = 0$ and $\Delta_{\rm p} = \Delta_{\rm s} \neq 0$. Here we only consider the case of $\Delta_{\rm p} = \Delta_{\rm s} \neq 0$.

For $\Delta_{\rm p} = \Delta_{\rm s} \neq 0$,

$$\lim_{t\to-\infty}\frac{\sqrt{\Omega_{\rm p}^2+\Omega_{\rm s}^2}}{\Delta_{\rm p}} = \lim_{t\to+\infty}\frac{\sqrt{\Omega_{\rm p}^2+\Omega_{\rm s}^2}}{\Delta_{\rm p}} = 0 \tag{9}$$

or, in other words,

$$\phi(-\infty) = \phi(+\infty) = 0;$$

which implies that only state $|\psi_-\rangle$ among the adiabatic states is populated initially as $|\psi_-\rangle = |b\rangle$. Furthermore, state $|\psi_-\rangle$ coincides with state $|c\rangle$ at $t \to +\infty$, thus if the excitation is adiabatic, then the system will remain in state $|\psi_-\rangle$, and the population will eventually be completely transferred to state $|c\rangle$.

Therefore, in the adiabatic limit, both the intuitive and counterintuitive pulse sequences produce complete population transfer for the case of $\Delta_{\rm p} = \Delta_{\rm s} \neq 0$. There is, however, a difference from the way that the population is transferred from state $|b\rangle$ to state $|c\rangle$. For the counterintuitive pulse, the population is transferred through the adiabatic state $|\psi_0\rangle$ which does not involve the intermediate state $|a\rangle$ and, thus, no population visits $|a\rangle$ at any time. For the intuitive pulse sequences, the population is transferred through the adiabatic state $|\psi_-\rangle$ which involves state $|a\rangle$. Thus, the intermediate state $|a\rangle$ is populated during the transfer. From Eq.(3c), the intermediate state $|a\rangle$ is populated during the transfer, and in the adiabatic limit its population is found to be

$$\begin{aligned} P_a(t) \approx & \sin^2\phi = \frac{1}{2}(1-\cos 2\phi) \\ = & \frac{1}{2}\left[1 - \frac{\Delta(t)}{\sqrt{\Omega_{\rm p}^2+\Omega_{\rm s}^2+\Delta^2}}\right]. \end{aligned} \tag{10}$$

Figure 5 depicts the dependence of population in the excited state $|a\rangle$ on time according to Eq.(10) in the case of intuitive pulse sequence.

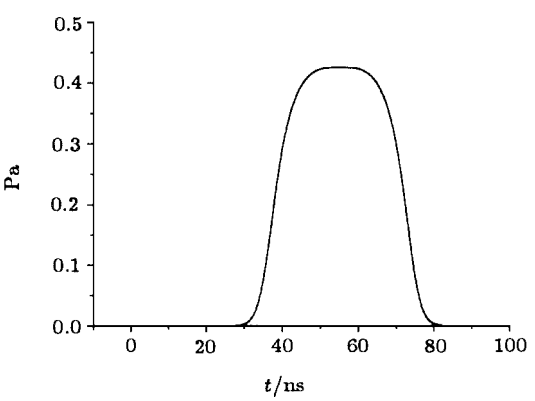


Fig.5. Dependence of population in excited state $|a\rangle$ on time using intuitive pulse sequence, with $\tau_{\rm p}$=50ns, $\tau_{\rm s}$=60ns, $\tau = 1/\gamma$=1ns $T_{\rm p}$=10ns and $T_{\rm s}$=10ns, $\Delta = 0.5$.

The maximum value of $P_a(t)$ is 50%, and it is determined by the particular case considered. This means that, in the case of strong decay from the intermediate state, the counterintuitive pulse sequence is more effective than the intuitive one.

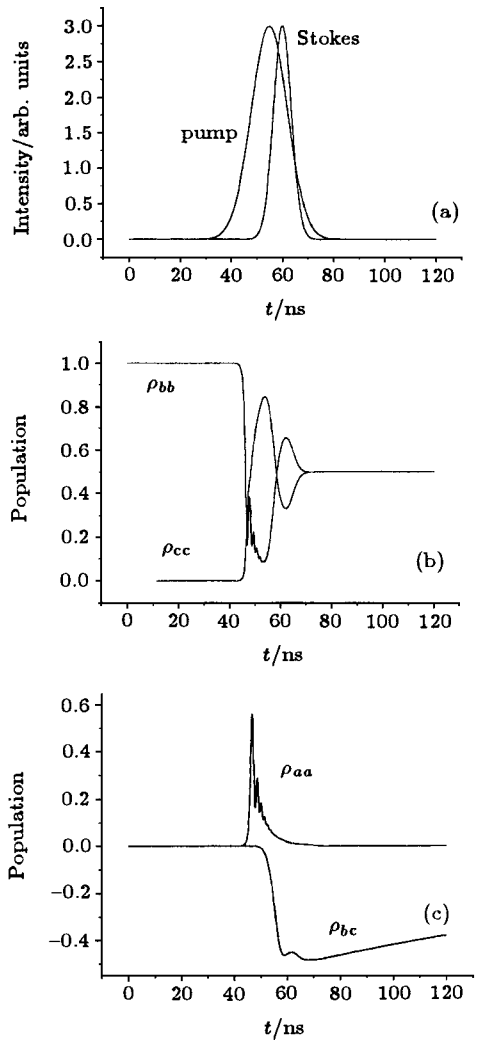


Fig.6. Numerical simulation of control of coherence using Intuitive pulse sequences. The parameters are τ_p=54.9ns, τ_s=60ns, $\tau = 1/\gamma$=1ns T_p=10ns and T_s=5ns.

Then we present the numerical simulation, as shown in Fig.6, for the case of maximal atomic coherence using the intuitive pulse sequences. It seems that the plots of Figs.4(a) and 6(a) are of the same envelopes, but we have exchanged their parameters. States $|b\rangle$ and $|c\rangle$ are also half/half populated as shown in Fig.6(b). Though the final results are the same in Figs.4(b) and 6(b), there are more Rabi oscillations in the populations of levels $|b\rangle$ and $|c\rangle$ in Fig.6(b) than in Fig.4(b). When using intuitive pulse sequences, level $|a\rangle$ is populated during the process and the maximal value reaches 0.5, as shown in Fig.6(c). In Fig.4(c), there are few populations in level $|a\rangle$; this is an advantage to realize maximal coherence in practice, for no population can leak out of the system through level $|a\rangle$. From Figs.4(c) and 6(c), we can see that maximal coherence can be realized in both cases theoretically. In practice, because of the population leaking out of the system through level $|a\rangle$ in Fig.6(c), counterintuitive pulse sequences is more perfect than intuitive one in realizing maximal coherence.

4. Conclusion

The three-level Λ system is the simplest model of STIRAP. It was found that four-level,[18] five-level,[19] and multilevel[20] systems can also be used to realize coherent population transfer. We have discussed the properties of population transfer and coherence by delayed pulses in a three-level Λ system in the adiabatic limit. Special attentions have been paid to the effect of the pulse sequence on the population transfer efficiency and coherence. There are some differences between the two pulse sequences, since the population transfers in the two sequences are realized via different adiabatic states. Both the intuitive and the counterintuitive pulse sequences can realize maximal coherence in theory, but there are some differences in practice. It has been shown that counterintuitive sequence is more efficiency and easy to realize than the intuitive one in transferring population and coherence.

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