Coherent population transfer and superposition of atomic states via stimulated Raman adiabatic passage using an excited-doublet four-level atom

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Coherent population transfer and superposition of atomic states via a technique of stimulated Raman adiabatic passage in an excited-doublet four-level atomic system have been analyzed. It is shown that the behavior of adiabatic passage in this system depends crucially on the detunings between the laser frequencies and the corresponding atomic transition frequencies. Particularly, if both the fields are tuned to the center of the two upper levels, the four-level system has two degenerate dark states, although one of them contains the contribution from the excited atomic states. The nonadiabatic coupling of the two degenerate dark states is intrinsic, it originates from the energy difference of the two upper levels. An arbitrary superposition of atomic states can be prepared due to such nonadiabatic coupling effect.

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I. INTRODUCTION

There has been a growing interest in target population transfer [1] over the past decade, because of its very interesting applications to chemical-reaction dynamics [2], laser-induced cooling [3], atomic optics [4], preparation of entanglement [5] and quantum computation [6]. Coherent population transfer between atomic ground-state levels originates from the concept of coherent population trapping [7]. The most robust technique for achieving efficient population transfer is stimulated Raman adiabatic passage (STIRAP) [1,8]. The technique of STIRAP allows, in principle, a complete population transfer from a single initial state to another single target state. Two different classical laser pulses are used in this model: the first (pump) laser pulse couples the ground state to the excited state which is connected by a second (Stokes) laser pulse to the final state. If the pump and Stokes frequencies maintain two-photon resonance, and if the Stokes laser pulse precedes the pump laser pulses (counterintuitively ordered pulses), then an efficient population transfer occurs when the evolution is adiabatic, that is, when an adiabatically decoupled (dark) state exists.

The simplest model of STIRAP is a three-level $\wedge$ system. Using this model, Kuklinski *et al.* [9] investigated the behavior of population transfer and gave the adiabatic following condition for generating such phenomenon. Later, it was found that four-level [10], five-level [11], multilevel [12] and autoionization [13] systems can also be used to realize coherent population transfer. An important feature of such schemes is that the adiabatic transformation is applied to the dark eigenstate of the system, i.e., the relevant eigenstate contains no contribution from the excited atomic state. Therefore the technique is immune to the detrimental consequence of atomic spontaneous emission.

It is well known, in a $\vee$-type system [14–16] or in an excited-doublet four-level system [17–19] with upper levels rapidly decaying to the lower state, that the dark states can also be exhibited because of the quantum interference effect. It was shown that the quantum interference effect can lead to depression or even cancellation of spontaneous emission from the excited doublet to the lower level, when an atom is initially in a superposition of upper levels [16] or the excited doublet is driven by a coherent field to an auxiliary level [17–21]. The theoretical prediction has been demonstrated experimentally in Ref. [20].

In this paper, we consider an excited-doublet four-level system and assume that the dipole moments between the two upper levels and each of the lower levels are parallel, in which the dark states can exist; even these states contain the contribution from the excited states. We investigate the behavior of adiabatic passage in this system, and find that this behavior is very closely related to the detunings between the laser frequencies and the atomic transition frequencies: a complete population transfer from an initial state to another target state can be realized in the excited-doublet four-level system, if both the pump and Stokes fields maintain two-photon resonance, but are not tuned at the middle point of the two upper levels; while an arbitrary superposition of atomic states can be prepared if both the fields are tuned to the center of the two upper levels.

The outline of this paper is as follows. In Sec. II, we present the dark states of the excited-doublet four-level atomic system. In Sec. III, we investigate the behavior of coherent population transfer and superposition of atomic states for this system in different frequency detunings of fields. Finally, we conclude in Sec. IV.

II. THE DARK STATES OF THE EXCITED-DOUBLET FOUR-LEVEL ATOMIC SYSTEM

We consider an excited-doublet four-level system as illustrated in Fig. 1. The transition from the upper levels $|2\rangle$ with energy $\hbar\omega_2$ and $|3\rangle$ with energy $\hbar\omega_3$ to the lower level $|1\rangle$ with energy $\hbar\omega_1$ is driven by the pump field with frequency ν_1. While the transition from the upper levels $|2\rangle$ and $|3\rangle$ to the lower level $|4\rangle$ with energy $\hbar\omega_4$ is driven by the Stokes field with frequency ν_2. The Hamiltonian for this system can be written as follows:

$$H = H_0 + H_1, \tag{1}$$

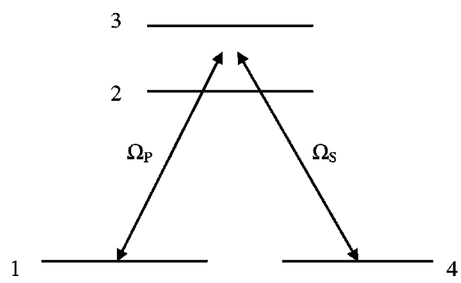

FIG. 1. An excited-doublet four-level atomic configuration with two lower states $|1\rangle$ and $|4\rangle$ and excited-doublet states $|2\rangle$ and $|3\rangle$. The transition from the upper levels $|2\rangle$ and $|3\rangle$ to the lower state $|1\rangle$ is driven by the pump pulse $\Omega_p(t)$; while the transition from the upper levels $|2\rangle$ and $|3\rangle$ to the lower state $|4\rangle$ is driven by the Stokes pulse $\Omega_s(t)$.

$$H_0=\hbar\Delta_1|2\rangle\langle 2|+\hbar\Delta_2|3\rangle\langle 3|+\hbar(\Delta_1-\delta)|4\rangle\langle 4|, \quad (2)$$

$$H_1=-\hbar[\Omega_{p1}(t)|2\rangle\langle 1|+\Omega_{p2}(t)|3\rangle\langle 1|] -\hbar[\Omega_{s1}(t)|2\rangle\langle 4|+\Omega_{s2}(t)|3\rangle\langle 4|]+\text{H.c.}, \quad (3)$$

where $\Omega_{p1}(t)$ and $\Omega_{p2}(t)$ are Rabi frequencies of the pump field coupling $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|1\rangle$, and $\Omega_{s1}(t)$ and $\Omega_{s2}(t)$ are Rabi frequencies of the Stokes field driving the transitions from $|2\rangle$ to $|4\rangle$ and $|3\rangle$ to $|4\rangle$, respectively. $\Delta_1=\omega_2-\omega_1-\nu_1$, $\Delta_2=\omega_3-\omega_1-\nu_1$, and $\delta=\omega_2-\omega_4-\nu_2$ are frequency detunings.

The time-dependent Schr̈odinger equation for this system is

$$\dot{c}(t)=-iw(t)c(t), \quad (4)$$

where $c(t)$ is a column vector, whose components are probability amplitudes $c_n(t)$, and the evolution matrix $w(t)$ has the form

$$w(t)=\begin{pmatrix} 0 & \Omega_{p1}(t) & \Omega_{p2}(t) & 0 \\ \Omega_{p1}(t) & -\Delta_1 & 0 & \Omega_{s1}(t) \\ \Omega_{p2}(t) & 0 & -\Delta_2 & \Omega_{s2}(t) \\ 0 & \Omega_{s1}(t) & \Omega_{s2}(t) & \delta-\Delta_1 \end{pmatrix}. \quad (5)$$

The characteristic equation of this system is

$$\lambda^4+a_1\lambda^3+a_2\lambda^2+a_3\lambda+a_4=0, \quad (6)$$

where

$$a_1=(\Delta_1+\Delta_2)-(\delta-\Delta_1), \quad (7)$$

$$a_2=-[\Omega_{p1}^2(t)+\Omega_{p2}^2(t)+\Omega_{s1}^2(t)+\Omega_{s2}^2(t)+(\Delta_1+\Delta_2)\times(\delta-\Delta_1)-\Delta_1\Delta_2], \quad (8)$$

$$a_3=-[\Omega_{p1}^2(t)\Delta_2-\Omega_{p1}^2(t)(\delta-\Delta_1)+\Omega_{p2}^2(t)\Delta_1-\Omega_{p2}^2(t)\times(\delta-\Delta_1)+\Omega_{s1}^2(t)\Delta_2+\Omega_{s2}^2(t)\Delta_1+\Delta_1\Delta_2(\delta-\Delta_1)], \quad (9)$$

$$a_4=[\Omega_{p1}^2(t)\Delta_2+\Omega_{p2}^2(t)\Delta_1](\delta-\Delta_1)+[\Omega_{p1}(t)\Omega_{s2}(t)-\Omega_{p2}(t)\Omega_{s1}(t)]^2. \quad (10)$$

We can easily find from Eqs. (6)–(10), that one or two of eigenvalues of the characteristic equation (6) is zero under the condition

$$\frac{\Omega_{p1}(t)}{\Omega_{p2}(t)}=\frac{\Omega_{s1}(t)}{\Omega_{s2}(t)}=C \quad (11)$$

(for simplicity, we choose C=1) and for suitable detunings. They are as follows:

(1) When only one of the fields is tuned to the center of the two upper levels, one of the eigenvalues of Eq. (6) will be zero. For $\Delta_1=-\Delta_2=\omega_{32}/2=\omega_0$, the corresponding eigenvector is

$$|E_1\rangle=\frac{1}{\sqrt{2\Omega_p^2(t)+\omega_0^2}}[\omega_0|1\rangle+\Omega_p(t)|2\rangle-\Omega_p(t)|3\rangle] \quad (12)$$

for the opposed case, i.e., $\delta=\omega_0$, but $\Delta_1\neq\omega_0$, the corresponding eigenvector is

$$|E_2\rangle=\frac{1}{\sqrt{2\Omega_s^2(t)+\omega_0^2}}[\Omega_s(t)|2\rangle-\Omega_s(t)|3\rangle+\omega_0|4\rangle]. \quad (13)$$

(2) When $\Delta_1=\delta\neq\omega_0$, i.e., the pump and the Stokes fields maintain two-photon resonance, but are not tuned at the middle point of the two upper levels. In this case, Eq. (6) has one zero eigenvalue, the corresponding eigenstate is

$$|E_3\rangle=\frac{1}{\sqrt{\Omega_p^2(t)+\Omega_s^2(t)}}[\Omega_s(t)|1\rangle-\Omega_p(t)|4\rangle]. \quad (14)$$

(3) When $\Delta_1=\delta=\omega_0$, i.e., both the pump and Stokes field are tuned to the center of the two upper levels, Eq. (6) has two zero eigenvalues, and the system has two degenerate eigenstates: one is Eq. (14), and the other is

$$|E_4\rangle=\frac{\sqrt{\Omega_p^2(t)+\Omega_s^2(t)}}{\sqrt{2(\Omega_p^2(t)+\Omega_s^2(t))+\omega_0^2}}\left[|2\rangle-|3\rangle+\frac{\omega_0}{\sqrt{\Omega_p^2(t)+\Omega_s^2(t)}}(\Omega_p(t)|1\rangle+\Omega_s(t)|4\rangle)\right]. \quad (15)$$

One can see from Eq. (14) that this eigenvector contains no contribution from the excited levels $|2\rangle$ and $|3\rangle$. Hence this is a dark state of the excited-doublet four-level atomic system. Although other eigenstates (12), (13), and (15) contain the contribution from the excited levels $|2\rangle$ and $|3\rangle$, it has been shown that these eigenstates are still dark states, as long as certain conditions are satisfied [14–21]. For this system the conditions are as follows [21]: (1) the dipole moments between the two upper levels and each of the two lower levels are parallel and (2) the spontaneous emission rates from $|2\rangle$ and $|3\rangle$ to $|1\rangle$ (or $|4\rangle$) are symmetric, i.e., $\gamma_{21}=\gamma_{31}$ and $\gamma_{24}=\gamma_{34}$. Under these conditions, the spontaneous emission pathways from $|2\rangle$ to $|1\rangle$ (or $|4\rangle$) and from $|3\rangle$ to $|1\rangle$ (or $|4\rangle$) display a destructive interference and cancellation really happens, which make the atom decouple from the vacuum model and the fluorescence depressed. So these eigenstates can become nondecaying in the vacuum, that is, these eigenstates are dark states when these conditions are met, even both the upper levels rapidly decay to the lower levels.

III. ADIABATIC PASSAGE IN THE EXCITED-DOUBLET FOUR-LEVEL SYSTEM

In the preceding section, we gave the dark states, Eqs. (12)–(15), corresponding to zero eigenvalues under different frequency detunings. It is known that the existence of the trapped state is necessary but not sufficient to guarantee the population transfer. Obviously, Eqs. (12) and (13) cannot be used to transfer the population from initial single state to another target state, because only one pump (or Stokes) field exists in these two dark states. However, coherent population transfer can occur via Eqs. (14) and (15).

When the pump and Stokes fields keep two-photon resonance, but are not tuned at the middle point of the two upper levels, the four-level system has only one dark state $|\phi_1\rangle=|E_3\rangle$, i.e., Eq. (14). This dark state is completely same as that of three-level $\wedge$ system. Therefore we can say under this condition, that the behavior of adiabatic passage for this four-level system is same as that of $\wedge$ system. The familiar form of $|\phi_1\rangle$ make possible the counterintuitive excitation scheme in which the Stokes pulse precedes the pump pulse. In this arrangement, all population initially in state $|1\rangle$ projects into $|\phi_1\rangle(\Omega_p(t)\ll\Omega_s(t))$, and at the final time all population in $|\phi_1\rangle$ projects onto $|4\rangle(\Omega_p(t)\gg\Omega_s(t))$.

For the two degenerate upper levels, and when both the pump and Stokes fields are resonance with their corresponding transition frequencies ($\delta=\Delta_1=\omega_0=0$). In this case, the system has two degenerate eigenstates: $|\phi_1\rangle$ and $|\phi_2\rangle=|E_4\rangle=1/\sqrt{2}(|2\rangle-|3\rangle)$. Because $\langle\phi_2|\dot{\phi}_1\rangle=\langle\dot{\phi}_2|\phi_1\rangle\equiv0$, there is no transition between these two dark states. If all population is initially in state $|1\rangle$, this system can adiabatically transfer the population from $|1\rangle$ to $|4\rangle$, which is unaffected by the presence of $|\phi_2\rangle$. That is, under this condition, a complete population transfer can also be realized via the STIRAP technique, which is same as that of Ref. [10]; while if all population is initially in $|\phi_2\rangle$ (spontaneously induced coherence can be used to generate such superposition state), this system stays in this dark state as long as the fields are turned on, which is also unaffected by the presence of $|\phi_1\rangle$.

The situation is more complicated and more interesting when both the pump and Stokes fields are tuned to the center of the two nondegenerate upper levels. In this case, the interaction Hamiltonian has two zero eigenvalues, the corresponding two degenerate dark states are Eqs. (14) and (15). Setting

$$\tan\theta=\frac{\Omega_p(t)}{\Omega_s(t)}, \tag{16}$$

$$\tan\varphi=\frac{\omega_0}{\sqrt{2(\Omega_p^2(t)+\Omega_s^2(t))}}, \tag{17}$$

Eqs. (14) and (15) can be rewritten as

$$|\phi_1\rangle=\cos\theta|1\rangle-\sin\theta|4\rangle, \tag{18}$$

$$|\phi_2\rangle=\sin\theta\sin\varphi|1\rangle+\frac{1}{\sqrt{2}}\cos\varphi(|2\rangle-|3\rangle)+\cos\theta\sin\varphi|4\rangle, \tag{19}$$

where θ is the mixing angle used in standard STIRAP [1], while φ is an additional mixing angle related to the energy difference of the two upper levels. If the two upper levels are degenerate, i.e., $\omega_0=0$, then $\varphi=0$. In this case there is no adiabatic coupling between the two dark states. While for $\omega_0\neq0$, i.e., for nondegenerate case, $\langle\phi_2|\dot{\phi}_1\rangle=-d\theta/dt\sin\phi$, a nonadiabatic transition between the two degenerate dark states may occur. Therefore we can say that the nonadiabatic coupling of the two dark states is intrinsic, it only depends on the energy difference of the two upper levels. In the adiabatic limit, the nonadiabatic coupling between these states cannot be neglected due to the degeneracy of the two dark states [22,23]. The behavior of the four-level system will be significantly influenced by such nonadiabatic coupling of the two dark states.

Consider the case where initially $|\Psi\rangle$ is either one of these two, by defining the state vector $|\Psi^i\rangle$ to be the state vector which evolves from the initial condition

$$|\Psi^i(-\infty)\rangle=|\phi_i(-\infty)\rangle. \tag{20}$$

At a later time, due to the coupling of the two degenerate eigenstates, $|\Psi^i\rangle$ acquires a component along $|\phi_j\rangle(j\neq i)$. The state vector then takes the form

$$|\Psi^i(t)\rangle=\sum_j B_{ij}(t)|\phi_j(t)\rangle(i,j=1,2), \tag{21}$$

where the matrix element B_{ij} is given by

$$\frac{d}{d(t)}B_{ij}(t)=-\sum_k A_{ik}(t)B_{kj}(i,j,k=1,2), \tag{22}$$

and

$$A_{jk}=\left\langle\phi_j\left|\frac{d}{d(t)}\right|\phi_k\right\rangle. \tag{23}$$

After some simple calculations, the matrix B_{ij} at the end of the interaction can be derived as follows

$$B(\infty)=\begin{pmatrix}\cos\gamma_f & \sin\gamma_f\\ -\sin\gamma_f & \cos\gamma_f\end{pmatrix}, \tag{24}$$

where

$$\gamma_f=\oint_{-\infty}^{t}\frac{d\theta}{dt'}\sin\varphi dt' \tag{25}$$

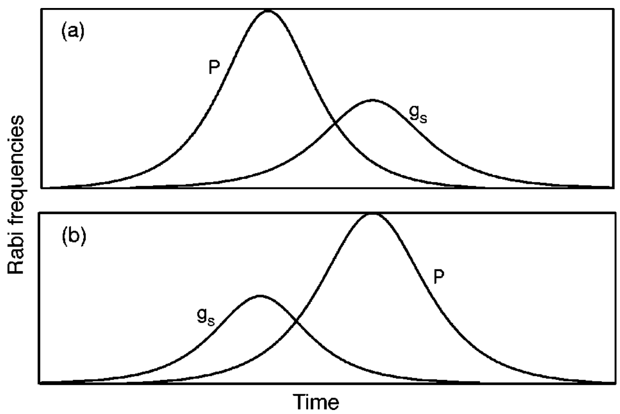

FIG. 2. (a) Intuitively ordered pulses (i.e., the Stokes pulse precedes the pump pulse) while (b) counterintuitively ordered pulses (i.e., the pump pulse precedes the Stokes pulse).

is called the geometric phase [24]. Comparing our scheme with the scheme of Ref. [22], we notice that there is an essential distinction between them. In Ref. [22], the additional mixing angle φ comes from the control field $Q(t)$. The nonadiabatic coupling of the two dark states is external, it depends on the coherent control field $Q(t)$. If $Q(t)=0$, the nonadiabatic coupling disappears and the system will reduce to the usual three-level system. While here the additional mixing angle comes from the energy difference of the two upper energy levels, the nonadiabatic coupling of the two dark states is intrinsic, it will disappear only if $\omega_0=0$.

Now we investigate the behavior of adiabatic passage in the four-level atomic system. We consider two different ordering of pulses: counterintuitively ordered pulses [i.e., the pump pulse precedes the Stokes pulse, Fig. 2(a)] and intuitively ordered pulses [i.e., the Stokes pulse precedes the pump pulse, Fig. 2(b)]. We assume that initially all population is in state $|1\rangle$. For the counterintuitively ordered pulses, we have initially $|\theta|=0$. Because ω_0 is a constant, we have initially and finally $\omega_0^2 \gg \Omega_p^2(t)+\Omega_s^2(t)$ leading to $|\varphi| = \pi/2$. Obviously the dark state $|\phi_1\rangle$ satisfies the initial condition. So the state vector will begin as the adiabatic state $|\phi_1\rangle$. After the end of interaction, the mixing angles are $\theta = \varphi = \pi/2$ so that $|\phi_1(\infty)\rangle = -|4\rangle$ and $|\phi_2(\infty)\rangle = |1\rangle$. According to Eq. (21), we have

$$|\Psi(\infty)\rangle_{cin} = \sin\gamma_f|1\rangle - \cos\gamma_f|4\rangle. \tag{26}$$

As for the intuitively ordered pulses, in the same way, we can easily obtain the results as follows

$$|\Psi(\infty)\rangle_{in} = -\sin\gamma_f|1\rangle + \cos\gamma_f|4\rangle. \tag{27}$$

Here the subscript "in" ("cin") denotes the ordering scheme, intuitively (counterintuitively) ordered pulses. Obviously, an arbitrary superposition states of atom can be prepared by both the conterintuitive and the intuitive ordering of pulses if the atom is initially in its state $|1\rangle$. The comparison of Eqs. (26) and (27) reveals that the superposition states emerging from $|1\rangle$ are almost same in both cases except for a difference in sign.

We can see from above results, that the behavior of adiabatic passage of the excited-doublet four-level system depends sensitively on the detunings between the laser frequencies and the atomic transition frequencies: if both the pump and the Stokes fields keep two-photon resonance, but are not tuned at the middle point of the upper levels, this system can be used to transfer the population from initial state to target state as three-level $\wedge$ system does; while if both the fields are tuned to the center of the two upper levels, the four-level system can be used to prepare an arbitrary superposition of atomic states. We can get our desired results by adjusting the detunings in the adiabatic limit. As for the adiabatic passage condition, it is same with three-level $\wedge$ atoms, which should be satisfied in the four-level atomic system.

IV. SUMMARY AND DISCUSSION

In this paper, we investigated the behavior of adiabatic passage in the excited-doublet four-level system, in which the parallel dipole moments between the two upper levels and each of the lower levels are considered. We found that the behavior of STIRAP is very closely related to the frequency detunings between the laser frequencies and the atomic transition frequencies: the behavior of coherent population transfer for the excited-doublet four-level system is same as that of three-level $\wedge$ system when both the pump and the Stokes fields keep two-photon resonance, but are not tuned at the middle point of the upper levels; while for both the fields tuned to the center of the two upper levels, the four-level system has two degenerate dark states, although one of them contains the contribution from the excited atomic states. The nonadiabatic coupling effect between the two dark states is intrinsic, it originates from the energy difference of the two upper levels. An arbitrary atomic superposition states can be prepared due to the nonadiabatic coupling of the two degenerate dark states. For an experimental realization of this proposal, it needs the conditions of adiabatic passage limit as well as complete cancellation of spontaneous emission in the four-level atomic system. The adiabatic condition is similar to $\wedge$ models [1], which should be easily satisfied; as for the condition of total cancellation emission, such experiment has been demonstrated from the mixing of the levels arising from internal fields [20]. Thus our scheme gives one way to achieve a complete population transfer and to prepare an arbitrary superposition of atomic states by using the excited-doublet four-level atom.

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