Control of coherent population transfer via spontaneous decay-induced coherence

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We investigate the effect of the spontaneous decay-induced coherence on the control of coherent population transfer in a closed double Λ-type four-level system with the energy separation of the excited doublet comparable to their decay rates by using the stimulated Raman adiabatic passage technique. It is shown that when the two pulse laser fields are tuned to the particular frequency where the condition for quantum interference is satisfied, remarkable enhancement or suppression of population transfer can be realized through the spontaneous emission constructive or destructive quantum interference even with a finite pulse area, which is very favorable to coherent population transfer in quantum systems with weak oscillator strengths.

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I. INTRODUCTION

Coherent controlling of an atomic or a molecular system to a particular state has attracted considerable interest in recent years. Many techniques, such as π pulse, stimulated emission pumping, chirped pulse, and stimulated Raman adiabatic passage (STIRAP), have been proposed to realize population transfer [1–4]. Among these techniques, the STIRAP has proven an efficient and robust way for selective and complete coherent population transfer between two discrete atomic or molecular states. In the extensively studied Λ-type three-level system, three conditions should be satisfied for perfect population transfer. First, the initially populated state 1 and target state 3 should be coupled via an intermediate state 2 by the time-separated but partially overlapping pump and Stokes pulses in a counterintuitive order (i.e., the Stokes pulse precedes the pump pulse); second, the two-photon resonance between state 1 and state 3 should be maintained (while state 2 may be off resonance by a certain detuning); third, the time evolution of the atomic or molecular system should be adiabatic (i.e., large enough pulse area). In the adiabatic regime, the system would evolve solely along the dark state composed only of the lower states 1 and 3 at all times, and efficient and selective population transfer from state 1 to state 3 can be achieved. As the dark state has no component of the excited state, the properties of the latter nearly have no impact on the transfer efficiency. Nevertheless, as recently discussed by Vitanov *et al.* [5,6], large excited state spontaneous decay within or out of the quantum system would deteriorate the adiabaticity and subsequently lead to imperfect population transfer. Therefore, in order to realize complete population transfer in a decaying system, a larger pulse area is required. However, in a real atomic or molecular system, especially those with weak oscillator strengths, the pulse area cannot be as large as one would expect theoretically. In this paper, we demonstrate that even if the pulse area is not large enough to ensure perfect population transfer in an otherwise Λ-type three-level system, the population transfer can be enhanced or suppressed dramatically through the spontaneous decay-induced coherence in a closed double Λ-type four-level system with a closely separated excited doublet by using the STIRAP technique, where the energy separation of the doublet is comparable to their decay rates and the laser Rabi frequencies are far smaller than the doublet energy separation. This is especially favorable to population transfer in the quantum systems with weak oscillator strengths.

The four-level system is similar to that in Refs. [7–9]. In Ref. [7], Coulston and Bergmann studied the effect of the fourth level on the population transfer in a Λ-type three-level system and found that the fourth level near the intermediate level has little influence on the efficiency and selectivity of the transfer process as long as the adiabaticity is satisfied. Jin *et al.* [8] demonstrated that an arbitrary coherent superposition between the two states 1 and 3 can be established in the double Λ-type four-level system when the two laser fields are tuned to a particular frequency with the STIRAP technique. In Ref. [9], Zhu *et al.* showed that resonance fluorescence quenching and spectral line narrowing can be realized via spontaneous decay-induced quantum interference in the steady state regime. In this paper, in contrast to the general argument that the spontaneous decay is intrinsically incoherent in nature and detrimental to coherent population transfer, we show that population transfer can be controlled by the quantum coherence created from spontaneous emission interference with a finite pulse area when the two pulse laser fields are tuned to the particular frequency where the condition for quantum interference is fulfilled.

The paper is organized as follows. In Sec. II, we present the motion equation of the density matrix in the closed double Λ-type four-level system. In Sec. III, we show the numerical results about the enhanced or inhibited population transfer via the spontaneous emission-induced coherence by solving the time-dependent density matrix equation. The conclusions are given in Sec. IV.

II. MOTION EQUATION OF THE DENSITY MATRIX

The closed double Λ-type four-level system, as shown in Fig. 1, interacts with two time-separated but partially over-

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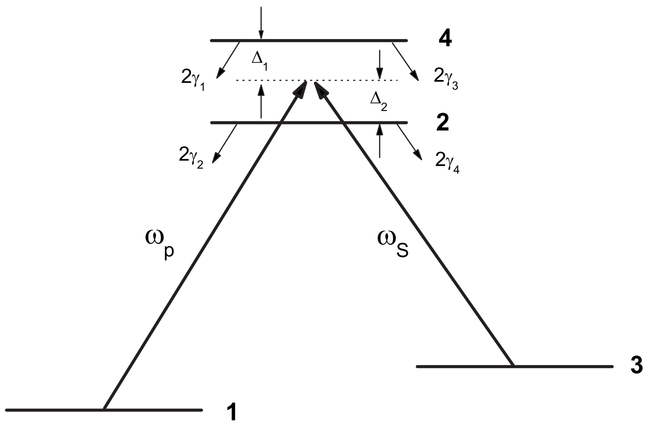

FIG. 1. The closed double Λ-type four-level system.

lapping laser pulses, where the level 1 and the closely separated doublet 4 and 2 are coupled by the pump laser with the Rabi frequencies $\Omega_1(t)=\mu_1 E_p(t)/\hbar$ and $\Omega_2(t)=\mu_2 E_p(t)/\hbar$, and the level 3 and doublet 4 and 2 are coupled by the Stokes laser with the Rabi frequencies $\Omega_3(t)=\mu_3 E_S(t)/\hbar$ and $\Omega_4(t)=\mu_4 E_S(t)/\hbar$, respectively. Here μ_1, μ_2, μ_3, and μ_4 are the dipole moments for the transitions 1-4, 1-2, 3-4, and 3-2, and $E_p(t)$ and $E_S(t)$ are the envelopes of the electric-field amplitudes, respectively. The Rabi frequencies of the pump and Stokes pulses are assumed to be Gaussian with the amplitude envelopes of the form $\Omega_{1,2}(t)=\Omega_{p1,p2}\exp[-(t-\tau)^2/T^2]$ and $\Omega_{3,4}(t)=\Omega_{S1,S2}\exp(-t^2/T^2)$, respectively, where T is the pulse duration, $\Omega_{p1,p2}$ and $\Omega_{S1,S2}$ are the peak values of the Rabi frequencies of the two pulses, and τ is the time delay between them. For simplicity, we assume that the four dipole moments are equal to each other, and the peak values of the two Rabi frequencies are also equal [we denote $\Omega_1(t)=\Omega_2(t)=\Omega_p(t)$, $\Omega_3(t)=\Omega_4(t)=\Omega_S(t)$]. The detunings of the pump laser from the resonant transitions 1-4 and 1-2 are given by $\Delta_1=\omega_p-\omega_{41}$ and $\Delta_2=\omega_p-\omega_{21}$, respectively, where $\omega_{ij}(i\neq j)$ is the resonant frequency between levels i and j, and ω_p is the pump laser frequency. We assume that the two-photon resonance between levels 1 and 3 is maintained, so the detunings of the Stokes laser with the frequency ω_S from the resonant transitions 3-4 and 3-2 are $\omega_S-\omega_{43}=\Delta_1$ and $\omega_S-\omega_{23}=\Delta_2$, respectively. The upper closely lying levels 4 and 2 are also coupled to the two lower levels 1 and 3 by the vacuum modes, and the spontaneous decay rates from the two upper levels to two lower levels are denoted as $2\gamma_1$, $2\gamma_2$, $2\gamma_3$, and $2\gamma_4$, respectively. Using the Weisskopf-Wigner approximation in the generalized reservoir theory, the equations for the density matrix elements can be written as follows [9–11]:

$$\dot{\rho}_{11}=2\gamma_2\rho_{22}+2\gamma_1\rho_{44}+i\Omega_1(\rho_{41}-\rho_{14})+i\Omega_2(\rho_{21}-\rho_{12})+2p_1\sqrt{\gamma_1\gamma_2}(\rho_{24}+\rho_{42}), \tag{1}$$

$$\dot{\rho}_{22}=-(2\gamma_2+2\gamma_4)\rho_{22}+i\Omega_2(\rho_{12}-\rho_{21})+i\Omega_4(\rho_{32}-\rho_{23})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})(\rho_{24}+\rho_{42}), \tag{2}$$

$$\dot{\rho}_{33}=2\gamma_4\rho_{22}+2\gamma_3\rho_{44}+i\Omega_3(\rho_{43}-\rho_{34})+i\Omega_4(\rho_{23}-\rho_{32})+2p_2\sqrt{\gamma_3\gamma_4}(\rho_{24}+\rho_{42}), \tag{3}$$

$$\dot{\rho}_{44}=-(2\gamma_1+2\gamma_3)\rho_{44}+i\Omega_1(\rho_{14}-\rho_{41})+i\Omega_3(\rho_{34}-\rho_{43})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})(\rho_{24}+\rho_{42}), \tag{4}$$

$$\dot{\rho}_{12}=-(\gamma_2+\gamma_4+i\Delta_2)\rho_{12}+i\Omega_1\rho_{42}-i\Omega_4\rho_{13}+i\Omega_2(\rho_{22}-\rho_{11})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})\rho_{14}, \tag{5}$$

$$\dot{\rho}_{13}=i\Omega_1\rho_{43}+i\Omega_2\rho_{23}-i\Omega_3\rho_{14}-i\Omega_4\rho_{12}, \tag{6}$$

$$\dot{\rho}_{14}=-(\gamma_1+\gamma_3+i\Delta_1)\rho_{14}+i\Omega_2\rho_{24}-i\Omega_3\rho_{13}+i\Omega_1(\rho_{44}-\rho_{11})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})\rho_{12}, \tag{7}$$

$$\dot{\rho}_{23}=-(\gamma_2+\gamma_4-i\Delta_2)\rho_{23}+i\Omega_2\rho_{13}-i\Omega_3\rho_{24}+i\Omega_4(\rho_{33}-\rho_{22})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})\rho_{43}, \tag{8}$$

$$\dot{\rho}_{24}=-(\gamma_1+\gamma_2+\gamma_3+\gamma_4-i\omega_{42})\rho_{24}-i\Omega_1\rho_{21}+i\Omega_2\rho_{14}-i\Omega_3\rho_{23}+i\Omega_4\rho_{34}-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})(\rho_{22}+\rho_{44}), \tag{9}$$

$$\dot{\rho}_{34}=-(\gamma_1+\gamma_3+i\Delta_1)\rho_{34}-i\Omega_1\rho_{31}+i\Omega_4\rho_{24}-i\Omega_3(\rho_{33}-\rho_{44})-(p_1\sqrt{\gamma_1\gamma_2}+p_2\sqrt{\gamma_3\gamma_4})\rho_{32}. \tag{10}$$

In the above equations, the parameter $p_{1,2}$ denotes the alignment of the two spontaneous emission dipole matrix elements, which are defined as $p_{1,2}(t)=\vec{\mu}_{2;1,3}\cdot\vec{\mu}_{4;1,3}/|\vec{\mu}_{2;1,3}|\cdot|\vec{\mu}_{4;1,3}|$ with $\vec{\mu}_{ij}$ being the dipole moment for the transition from level i to level j, and the term $p_1\sqrt{\gamma_1\gamma_2}$ (or $p_2\sqrt{\gamma_3\gamma_4}$) represents the effect of quantum interference between the spontaneous emission pathways from level 2 to level 1 (or level 3) and from level 4 to level 1 (or level 3).

As discussed in Refs. [9–13], the parameters p_1 and p_2 play a very important role in the creation of coherence between the excited doublet. When the parameters p_1 and p_2 are positive (or negative), destructive (or constructive) quantum interference between the two spontaneous emission pathways can occur. If the alignment of the dipole moments between the two upper levels and each of the lower levels is perfectly parallel (or antiparallel), $p_1=p_2=1$ (or -1), complete destructive (or constructive) interference would take place, and the effect of the coherence between the doublet would be strongest; if the alignment of the two dipole moments is orthogonal (i.e., $p_1=p_2=0$), there will be no quantum interference. It should be noted that the interference of the spontaneous emission pathways can occur only if the transitions from the two closely-separated doublet share the same vacuum modes [9–12]. Hence, the effects of spontaneous decay-induced coherence are possible only if the two levels are close in energy compared to their decay rates. In the following, we set $\omega_{24}=2\gamma_1$ and $\gamma_1=\gamma_2=\gamma_3=\gamma_4=1$ for simplicity, and the other parameters are scaled with γ_1. We numerically integrate the density matrix equation by using the fourth-order Runge-Kutta integrator with the population initially in state 1 and study the effect of the coherence created from the spontaneous emission interference on the control of the population transfer with the STIRAP technique.

III. RESULTS AND DISCUSSIONS

It is well known that the mechanism of STIRAP [1–4] is based on coherent population trapping (CPT), which has been originally studied by Alzetta *et al.* [14], Arimondo and Orriols [15], and Stroud *et al.* [16], and been recently reviewed by Arimondo [17]. In the extensively studied Λ-type three-level system, when two-photon resonance between state 1 and state 3 is satisfied, there exists a time-dependent trapped state (i.e., time-dependent dark state), which is a coherent superposition of the two lower states 1 and 3. In the adiabatic limit, if the time-separated but partially overlapping pump and Stokes pulses are applied in the counterintuitive order, then the system would evolve solely along the stark state at all times, which coincides with state 1 initially and state 3 in the end, and perfect population transfer from state 1 to state 3 without ever actually populating state 2 can be realized. In the present double Λ-type four-level system, without the consideration of the spontaneous decays of the excited doublet, as analyzed in Refs. [8,9], when the pump and Stokes fields keep two-photon resonance, but are not tuned at the midpoint of the doublet, there only exists one eigenstate (adiabatic state) of the time-dependent Hamiltonian of the atom-field system with zero eigenvalue, which is

$$|\phi_0\rangle = \cos\theta|1\rangle - \sin\theta|3\rangle, \tag{11}$$

where $\tan\theta=\Omega_p(t)/\Omega_S(t)$, and θ is the mixing angle used in standard STIRAP. Obviously, the adiabatic state φ_0 has no component of the bare states 2 and 4 and is therefore the dark state, as studied in CPT in the 1970s [14–17]. In the adiabatic regime, the system would evolve solely along the dark state φ_0 and complete population transfer from state 1 to state 3 can be obtained with the counterintuitively ordered pulses, just as in the isolated three-level system formed by states 1, 2 (or 4), and 3, which has been studied in Refs. [7,8]. However, when the two lasers are tuned to the specific one-photon detuning where the condition for quantum interference is satisfied, i.e., $\mu_1^2\Delta_2+\mu_2^2\Delta_1=0$ and $\mu_3^2\Delta_2+\mu_4^2\Delta_1=0$ [9,11,12], there exist two degenerate adiabatic states with the eigenvalues equal to zero; one is the trapped state of Eq. (11), and the other is

$$|\phi_1\rangle = \sin\theta\sin\varphi|1\rangle + \frac{\cos\varphi}{\sqrt{2}}(|2\rangle - |4\rangle) + \cos\theta\sin\varphi|3\rangle, \tag{12}$$

where $\tan\varphi=\omega_{42}/2/\sqrt{2[\Omega_S^2(t)+\Omega_p^2(t)]}$, and φ is an additional mixing angle related to the energy separation of the doublet. Due to the nonadiabatic coupling between the two degenerate states φ_0 and φ_1, the outcome of the system state vector $\Psi(\infty)$ is a mixture of the two bare states 1 and 3, which can be described as

$$|\Psi(\infty)\rangle = \sin\gamma_f(\infty)|1\rangle - \cos\gamma_f(\infty)|3\rangle, \tag{13}$$

where $\gamma_f(t)=\int_{-\infty}^{t}(d\theta/dt')\sin\varphi dt'$ is called the Berry phase [8,18]. It can be seen from Eq. (13) that if the energy separation of the doublet ω_{42} is far smaller than the peak Rabi frequencies Ω_p and Ω_S, then φ is nearly equal to zero, and the population transfer behaves almost in the same manner as that in the Λ-type three-level system; if ω_{42} is much larger than Ω_p and Ω_S, then φ nearly equals $\pi/2$, and almost no population transfer can occur. However, when ω_{42} is comparable to Ω_p and Ω_S, only a part of populations can be transferred from state 1 to state 3, and a coherent superposition between them is established.

When the spontaneous decays of the upper doublet within the studied system are taken into account, the time evolution of the quantum system could be readily treated with the density matrix equation. We consider the situation that when the pulse area is not large enough to ensure adiabaticity in an otherwise Λ-type three-level system, how the spontaneous decay-induced coherence can control the population transfer with the energy separation of the excited doublet comparable to their decay rates and the laser Rabi frequencies much smaller than the doublet energy separation.

Figure 2 shows the time evolution of the populations in the four states with the laser fields tuned midway between the doublet 2 and 4 under different values of the parameters p_1 and p_2, where the pulse area is finite (only equal to 6). In all figures, the time origin is chosen at the peak of the Stokes

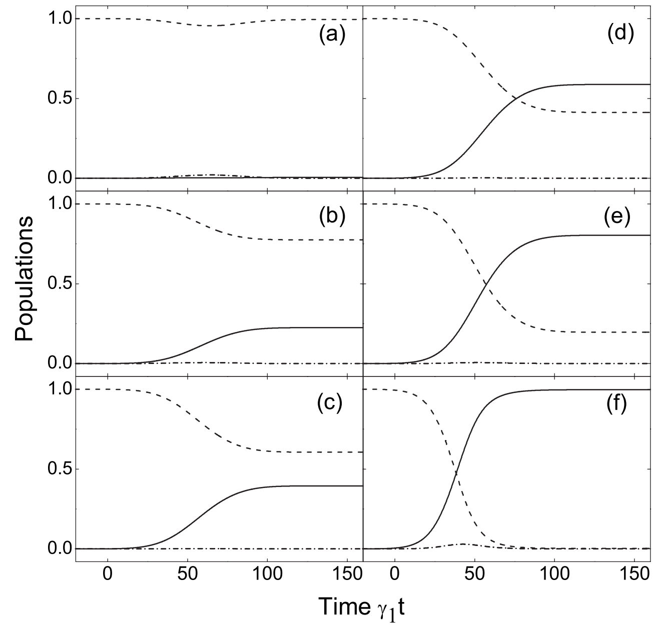

FIG. 2. The time evolution of the populations in the four states ρ_{11} (dashed line), ρ_{33} (solid line), and ρ_{22} and ρ_{44} (dot-dashed lines) with the laser fields tuned at the middle point of the doublet under different values of the parameter p $(p=p_1=p_2)$ with $\Omega_{p1,p2}=\Omega_{S1,S2}=0.15$, $T=40$, $\tau=60$, $\omega_{42}=2$, $\gamma_2=\gamma_3=\gamma_4=\gamma_1=1$, in corresponding units of γ_1. (a) $p=1$, (b) $p=0.9$, (c) $p=0.6$, (d) $p=0$, (e) $p=-0.5$, and (f) $p=-1$.

pulse. It can be seen that the population transfer is dramatically modified by the coherence between the doublet resulting from the quantum interference between the two spontaneous emission pathways. In the absence of the quantum interference (i.e., $p_1=p_2=0$), only a part of populations (about sixty percent) can be transferred from state 1 to state 3 due to the limited pulse area and large energy separation and decay rates of the doublet [see Fig. 2(d)]. When the dipole moments between the two upper levels and each of the two lower levels are exactly parallel (i.e., $p_1=p_2=1$), the population would be trapped in state 1, and almost no population transfer can take place. This is due to the fact that the maximal quantum coherence of the doublet created from the complete destructive interference between the spontaneous pathways prevents the population in state 1 from being excited by the pump field. Similar zero absorption from spontaneous decay-induced coherence has been studied in a V-type system [19–21] and in an excited-doublet four-level system [9,11,13] in the steady state regime. With the deviation of the parameters p_1 and p_2 from the maximum values (unity), as the destructive interference is incomplete, which implies weak coherence between the doublet, the population in state 3 increases from zero to a certain steady value and the population in state 1 decreases from unity to a steady value, whereas the populations in the doublet 2 and 4 (the doublet has equal populations) nearly remain zero [see Figs. 2(b) and 2(c)]. When the parameters p_1 and p_2 further decrease and become negative, the population in state 3 increases further due to the coherence from the constructive quantum interference. If the alignment of the two matrix elements is antiparallel (i.e., $p_1=p_2=-1$), almost complete population transfer can be realized due to the strongest quantum coherence induced from the maximal constructive quantum interference, as seen in Fig. 2(f), even though the pulse area is not large enough to ensure perfect population transfer in an otherwise Λ-type three-level system. This can be well understood from the final populations as a function of the one-photon detunings of the pump and stokes lasers while keeping the two-photon resonance satisfied under different values of the parameters p_1 and p_2, as shown in Fig. 3.

It can be seen from Fig. 3(a) that when $p_1=p_2=1$, the population in state 3 appears as a narrow dip at the middle point of the two one-photon resonances; the largest population transfer occurs at the two one-photon resonances, whereas no population transfer can take place at the middle point of the doublet due to the maximal destructive quantum interference. As both the spontaneous decay rates and energy separation of the doublet are much larger than the laser field Rabi frequency, which would deteriorate the adiabaticity, and the pulse area is limited, the transfer efficiency at the two one-photon resonances is very low (only about 43%). This also accounts for the rapid decrease of the transfer efficiency with respect to the one-photon detuning. With the decrease of the parameters p_1 and p_2, the depth of the dip in the state 3 population becomes smaller and its width becomes wider, and the transfer efficiency at the middle point increases faster than that at the two one-photon resonances [see Figs. 2(b) and 2(c)]. When $p_1=p_2=0$, almost equal population transfer can be realized at the two one-photon resonances and the midway between them, as shown in Fig. 3(d). Nearly complete population transfer to the target state 3 at the middle point can be achieved with the antiparallel alignment of the two matrix elements due to maximum constructive quantum interference [see Fig. 3(f)]. Further calculations show that the transfer efficiency would decrease with the decrease of the Rabi frequencies of the two pulses and the increase of the energy separation of the excited doublet.

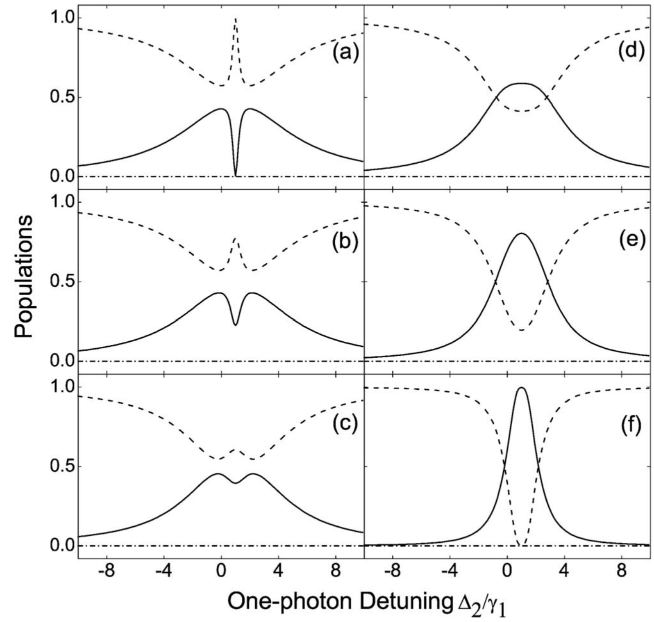

FIG. 3. The final populations in the four states ρ_{11} (dashed line), ρ_{33} (solid line), and ρ_{22} and ρ_{44} (dot-dashed lines) as a function of the one-photon detunings of the pump and stokes lasers under different values of the parameter p. (a) $p=1$, (b) $p=0.9$, (c) $p=0.6$, (d) $p=0$, (e) $p=-0.5$, and (f) $p=-1$, and the other parameters are the same as those in Fig. 2.

Finally, we consider the transfer efficiency at the middle point of the doublet as a function of the time delay between the two pulses under different values of the parameters p_1 and p_2. As displayed in Fig. 4, when the alignment of the two matrix elements is exactly parallel, the transfer efficiency remains equal to zero irrespective of time delay. With the decrease of the parameters p_1 and p_2, the transfer efficiency increases as the time delay increases and then reaches a steady value. When the alignment of the two matrix elements is perfectly antiparallel, the transfer efficiency increases faster to a steady value of nearly 100% and would not decrease even as the time delay increases indefinitely. The behavior has also been studied in the three-level Λ-type system with a large decay rate of the excited state [6]. The independence of the transfer efficiency on large time delay is due to the fact that the system is closed and the population transfer dynamics is determined by optical pumping with large time delay, where the time delay is irrelevant. It should be noted that the population transfer through the optical pumping is incoherent, whereas it is coherent through the STIRAP process.

Obviously, as seen in Figs. 2–4, the coherent population transfer can be efficiently controlled by the quantum coherence created from spontaneous emission interference even with a finite pulse area when the two pulse laser fields are

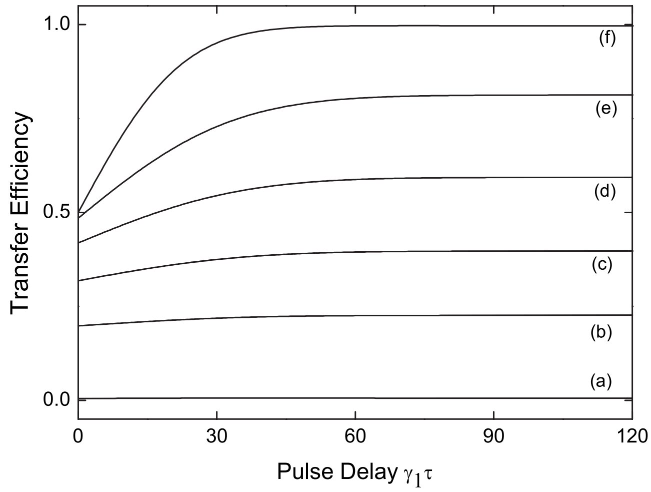

FIG. 4. The transfer efficiency as a function of the time delay between the two pulses with the laser fields tuned at the middle point of the doublet under different values of the parameter p. (a) $p=1$, (b) $p=0.9$, (c) $p=0.6$, (d) $p=0$, (e) $p=-0.5$, and (f) $p=-1$, and the other parameters are the same as those in Fig. 2.

tuned to the particular frequency where the condition for quantum interference is satisfied. The control of the population transfer via the spontaneous decay-induced coherence can also be realized even for the case when the excited doublets are well separated. However, in this case, higher Rabi frequencies of the laser fields are needed in order to get the same transfer efficiency. This clearly indicates that it is no longer true that the spontaneous decay would always be detrimental to coherent population transfer, at least not in general.

In order to experimentally observe the effects, one needs two closely separated levels with parallel or antiparallel dipole moments. This can be experimentally obtained by using the sodium dimer, as suggested in Refs. [22–24], where the mixture of the triplet and singlet g-parity Rydberg states by spin-orbit interaction in the sodium dimer can be used to form the two closely separated levels, and the alignment of the dipole moments between the upper doublet and a lower singlet state (or a triplet state) are parallel $p=1$ (or antiparallel $p=-1$). Therefore, by tuning the frequencies of the two pulse lasers and the time delay between them, the predicted effects under the maximal quantum coherences from the complete destructive or constructive interferences between the spontaneous pathways could be experimentally observed.

IV. CONCLUSIONS

In conclusion, in contrast to the general thought that the spontaneous decays are dephasing in nature and detrimental to coherent population transfer in the Λ-type three-level system [5,6], we demonstrate that the population transfer can be enhanced or inhibited remarkably through the spontaneous decay-induced coherence in a double Λ-type four-level system with the energy separation of the excited doublet comparable to their decay rates when the lasers are tuned to the particular one-photon detuning where the quantum interference can take place. In the double Λ-type four-level scheme, almost perfect population transfer can be realized by using the STIRAP technique with a relatively small pulse area compared with the case of the Λ-type three-level scheme, due to the constructive quantum interference between the two spontaneous emission pathways. This is particularly favorable to coherent population transfer in the quantum systems with weak oscillator strengths.

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