Creation of atomic coherent superposition states via the technique of stimulated Raman adiabatic passage using a Λ-type system with a manifold of levels

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We propose a scheme for creating atomic coherent superposition states via the technique of stimulated Raman adiabatic passage in a Λ-type system where the final state has twofold levels. With the application of a control field, it is found that the presence of double dark states leads to two arbitrary coherent superposition states with equal amplitude but inverse relative phases, even though the condition of multiphoton resonance is not met. In particular, two orthogonal maximal coherent superposition states are created when the control field is resonant with the transition of the twofold levels. This scheme can also be extended to manifold Λ-type systems.

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Coherent superpositions of atomic or molecular states play a crucial role in contemporary quantum physics. Phenomena associated with coherent superpositions have attracted considerable attention and offer a variety of interesting potential applications, such as electromagnetically induced transparency and coherent population trapping [1,2], chemical reaction dynamics [3], quantum information and computing [4–6], quantum entanglement [7], and so on. Atoms or molecules prepared in specified quantum state can be realized by the π-pulse technique, stimulated Raman adiabatic passage [8,9], the chirped pulse method [10,11], etc. Although the π-pulse technique can prepare atoms in a particular state, this method is not robust because it is very sensitive to small variations of pulse areas. In contrast, the technique of stimulated Raman adiabatic passage (STIRAP) robustly allows a complete population transfer from an initial single state to the target single state. In addition to straightforward population transfer to a single quantum state, the STIRAP technique has been widely applied to create coherent superposition states. For instance, fractional STIRAP [12] has been introduced to construct coherent superposition states whose composition is determined by the intensity ratio of the pump and Stokes pulses. Moreover, a tripod linkage has been used for realization of superposition states and measurement of a coherent superposition state with equal amplitudes in metastable neon [13,14]. Nevertheless, in all the above schemes, the pump and Stokes fields must remain in two-photon resonance so as to eliminate the excited state from the transfer process and keep high transfer efficiency. On the other hand, in manifold level schemes, chirped adiabatic passage has been used to prepare a population in a superposition state of the final manifold levels [15]. Because of using chirped pulses, however, careful control of the chirp rate is required, and analytic solutions are difficult to achieve.

The existence of a dark state is the basis of electromagnetically induced transparency, coherent population trapping, adiabatic population transfer, etc. In manifold level systems [15,16], double dark states are present and their interaction has been studied by Lukin *et al.* [16]. Motivated by Refs. [15,16], in this paper we introduce a Λ-type system, where the final state has the structure of twofold closely spaced levels, to create atomic coherent superposition states. The application of a control field makes the condition of multiphoton resonance unnecessary. Because of the existence of double dark states, two arbitrary coherent superposition states with equal amplitude but inverse relative phases are constructed analytically. In the case of where the control field is resonant with the transition between the two closely spaced levels, two orthogonal maximally coherent superposition states are created simply.

Consider a four-level Λ-type system as shown in Fig. 1. The twofold levels, labeled 3 and 4, are coupled by a constant control field Ω_c, which can be a microwave or quasistatic field. Levels 1 and 2 and 2 and 3 are coupled by the pump laser pulse $\Omega_p(t)$ and the Stokes laser pulse $\Omega_s(t)$, respectively. The wave functions of the bare states are denoted by $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$. Then the time evolution of the probability amplitudes $C(t)=[C_1(t),C_2(t),C_3(t),C_4(t)]^T$ of the four states is described by the Schrödinger equation

$$i\hbar\frac{d}{dt}C(t)=H(t)C(t), \tag{1}$$

where the Hamiltonian of this system in the rotating-wave approximation is

$$H=\hbar\begin{bmatrix} 0 & \Omega_p(t) & 0 & 0 \\ \Omega_p(t) & -\Delta_1 & \Omega_s(t) & 0 \\ 0 & \Omega_s(t) & -\Delta & \Omega_c \\ 0 & 0 & \Omega_c & -(\Delta-\Delta_3) \end{bmatrix}. \tag{2}$$

Here, $\Delta_1=\omega_2-\omega_1-\omega_p$, $\Delta_2=\omega_2-\omega_3-\omega_s$, and $\Delta_3=\omega_3-\omega_4-\omega_c$ denote single-photon detunings. $\Delta=\Delta_1-\Delta_2$ means two-photon detuning. $\hbar\omega_i$ $(i=1–4)$ describes the energy of level $|i\rangle$, and ω_j $(j=p,s,c)$ is the carrier frequency of laser j. For simplicity, the above detunings are assumed time independent. Simple calculations show that one of the eigenvalues of Eq. (2) will be equal to zero if

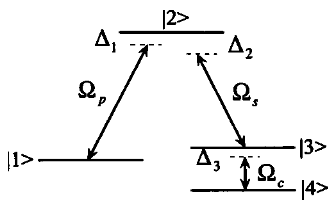

FIG. 1. Schematic diagram of a Λ-type system where the final state is a set of two of closely spaced levels.

$$\Delta = \frac{\Delta_3 \pm \sqrt{\Delta_3^2 + 4\Omega_c^2}}{2} \tag{3}$$

is satisfied. Then the corresponding eigenvector can be introduced as

$$|\psi_0\rangle = \cos\,\vartheta|1\rangle - \sin\,\vartheta(\cos\,\phi|3\rangle \pm \sin\,\phi|4\rangle), \tag{4}$$

with the mixing angles ϑ and ϕ defined to be

$$\tan\,\vartheta(t) = \frac{\Omega_p(t)}{\Omega_s(t)}\sqrt{1 + \frac{\Delta}{\Delta - \Delta_3}}, \quad \tan = \phi\frac{\Omega_c}{|\Delta - \Delta_3|}. \tag{5}$$

Here, the mixing angle ϑ is similar to the conventional one defined in the usual Λ-type system but is modified by a factor $a=\sqrt{1+\Delta/(\Delta-\Delta_3)}$. Certainly, since α is time independent, it will not alter the time evolution of the mixing angle ϑ. Meanwhile, ϕ defined here is an additional mixing angle related to detunings and the control field. It is obvious that, if the control field is removed, the adiabatic state will be rewritten as $|\psi_0\rangle=\cos\,\vartheta|1\rangle-\sin\,\vartheta|3\rangle$, and thus our system evolves into the normal Λ-type case.

From Eq. (4), one can see that the adiabatic state $|\psi_0\rangle$ associated with the null eigenvalue has no component of the excited state $|2\rangle$ and hence is immune to the specific properties of state $|2\rangle$ the main source of possible spontaneous emission to other states. So it is a dark state. Moreover, because the coupling of the state $|\psi_0\rangle$ to the other eigenstates is negligible in the adiabatic limit, we do not show the other three eigenstates here.

From Eqs. (3) and (4), we can see clearly that, corresponding to Δ_+ and Δ_-, there exist two dark states $|\psi_0\rangle_+$ and $|\psi_0\rangle_-$. [Here the subscripts correspond to the signs + and − in Eqs. (3) and (4), respectively.]. For delayed counterintuitive pump and Stokes pulses (the Stokes pulse precedes the pump pulse), the relations $0 \xleftarrow{t\to-\infty} \Omega_p(t)/\Omega_s(t) \xrightarrow{t\to\infty} \infty$ apply. As time progresses from $-\infty$ to ∞, the mixing angle ϑ rises from 0 to $\pi/2$. Consequently, the adiabatic state $|\psi_0\rangle_\pm$ starting in the bare state $|1\rangle$ will end in the coherent superposition state $|\Phi\rangle_+=\cos\,\phi|3\rangle+\sin\,\phi|4\rangle$ or $|\Phi\rangle_-=\cos\,\phi|3\rangle-\sin\,\phi|4\rangle$. These two superposition states have equal amplitude but inverse relative phases. Suitable manipulation of the detunings and control field permits any value of ϕ we desire. Therefore, at the end of the pump pulse, either a single state or an arbitrary superposition of $|3\rangle$ and $|4\rangle$ can be realized. It is important to note that this process needs neither two-photon nor three-photon resonance. Apparently, it is the control field that compensates the multiphoton detunings, and therefore the absolute transfer efficiency is kept without the condition of multiphoton resonance.

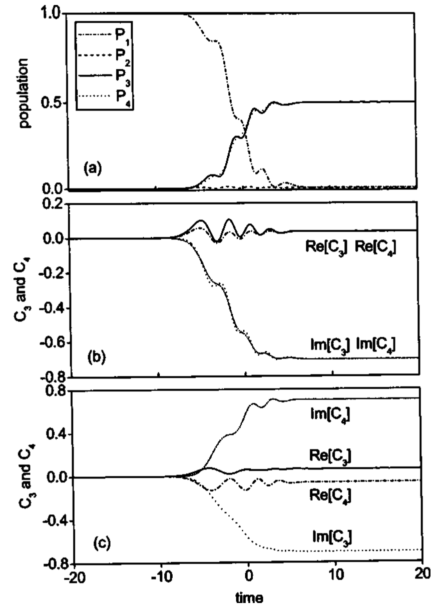

FIG. 2. The time evolutions of (a) populations $P_i=|C_i|^2$ ($i=1-4$), and (b) and (c) amplitudes C_3 and C_4. Parameters are $\Omega_p=\Omega_s=4.0$, $\Omega_c=2.5$, $\Delta_1=3.5$, and $\Delta_3=0$. (a) $\Delta_2=1.0$ or 6.0. (b) $\Delta_2=1.0$. (c) $\Delta_2=6.0$. All the parameters are scaled by T_0.

It is noteworthy that when the control field is on resonance, i.e., $\Delta_3=0$, the ultimate superposition state $|\Phi\rangle_\pm$ has the form of $|\Phi\rangle_+=(|3\rangle+|4\rangle)/\sqrt{2}$ for $\Delta_+=\Omega_c$ and $|\Phi\rangle_-=(|3\rangle-|4\rangle)/\sqrt{2}$ for $\Delta_-=-\Omega_c$. A special feature here is that the target state is now a maximally coherent superposition of the twofold levels, and $|\Phi\rangle_+$ is orthogonal to $|\Phi\rangle_-$. They are possible states of the quantum bits and hence have essential applications in quantum phase gates [5,17].

To illustrate the preceding analytic solutions, we present numerical simulations in the case of Gaussian pulses with the same duration T for the pump and Stokes lasers, $\Omega_p(t)=\Omega_p\exp[-(t-\tau)^2/T^2]$ and $\Omega_s(t)=\Omega_s\exp[-(t+\tau)^2/T^2]$. The Stokes pulse precedes the pump pulse, and the delay time between them is 2τ. Ω_s and Ω_p are the peak values of the two pulses. During our numerical simulations, Eq. (3) and the condition $\Omega_s(\Omega_p)T \gg 1$ (adiabatic criterion) are satisfied. Without loss of generality, the system is assumed to be initially in state $|1\rangle$, i.e., $C_1(-\infty)=1$, $C_{2,3,4}(-\infty)=0$.

By solving the Schrödinger equation (1), we show the time evolutions of the populations. Here $T=5T_0$ and $\tau=2.5T_0$. We scale all parameters in terms of T_0. As Fig. 2(a) reveals, when $\Delta_3=0$, the twofold levels $|3\rangle$ and $|4\rangle$ obtain equal population at the end of the pump pulse while levels $|1\rangle$ and $|2\rangle$ are empty. Figures 2(b) and 2(c) show the time evolutions of probability amplitudes C_3 and C_4. After all the interactions are over, $C_3=C_4$ when $\Delta=\Omega_c$ while $C_3=-C_4$ when $\Delta=-\Omega_c$. As discussed above, these two cases correspond to two orthogonal coherent superposition states $|\Phi\rangle_+$ and $|\Phi\rangle_-$. We find that under the condition of adiabatic evolution the resonant control field ($\Delta_3=0$) guarantees the creation of two precisely orthogonal maximal coherent superposition states $|\Phi\rangle_+$ and $|\Phi\rangle_-$.

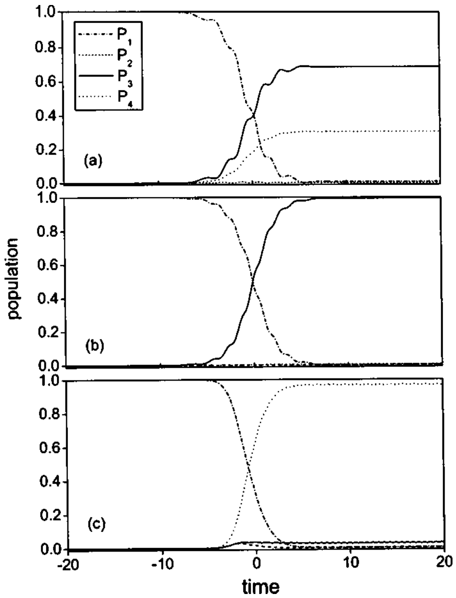

FIG. 3. The time evolutions of populations $P_i=|C_i|^2$ $(i=1-4)$. Parameters are (a) $\Omega_p=\Omega_s=4.0$, $\Omega_c=1.5$, $\Delta_1=2.0$, and $\Delta_2=1.0$; (b) $\Omega_p=\Omega_s=4.0$, $\Omega_c=3.0$, $\Delta_1=0.0$, and $\Delta_2=0.2$; (c) $\Omega_p=2.0$, $\Omega_s=9.0$, $\Omega_c=1.7$, $\Delta_1=11$, and $\Delta_2=1.0$. All the parameters are scaled by T_0.

From Eq. (4) one can derive the final population probability ratio of level $|3\rangle$ to $|4\rangle$: $R=(\cos\,\phi/\sin\,\phi)^2$. Figure 3 illustrates some examples. As is well known, the STIRAP technique applied in normal Λ-type systems can transfer population from the initial state to the final one completely, but it is essential that the condition of two-photon resonance be met. Otherwise, the excited level will be populated during the transfer process and thus its decay will sharply deteriorate the transfer efficiency. However, in the present case, an efficient population transfer without any population in the excited level can be realized although the two-photon detuning Δ is not equal to zero. Therefore, we can say that a preselected atomic single or superposition quantum state can be realized through a proper choice of parameters according to the analytic expression of the dark state $|\psi_0\rangle_\pm$ without any resonance conditions being satisfied. These parameters, i.e., the strength of the constant control field and the detunings Δ_3 and Δ, require precise control according to the expression of the additional mixing angle ϕ. Nevertheless, it is well known that the frequency fluctuations are negligible, and the constant control field, which can be a microwave field, has a power stability of better than 1%. Therefore, the desired superposition state in the present case should be influenced only slightly by the fluctuations.

As for the adiabatic criterion, we display the other three nonzero eigenvalues (solid line) and the rate of change in the mixing angle $\dot{\vartheta}$ (dotted line) numerically in Fig. 4. One can see that $\dot{\vartheta}$ is very small compared with the separations of the eigenvalues. Hence, nonadiabatic coupling between the eigenstates is negligible and the adiabatic condition can be satisfied.

Additionally, this Λ-type system with twofold levels can be extended to manifold Λ-type systems, just as shown in

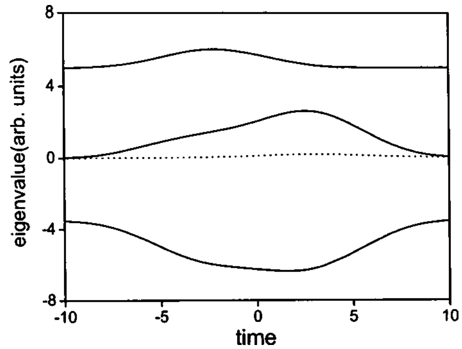

FIG. 4. The three nonzero eigenvalues (solid line) and the rate of change in the mixing angle $\dot{\vartheta}$ (dotted line). Parameters are $\Omega_p=\Omega_s=4.0$, $\Omega_c=2.5$, $\Delta_1=3.5$, $\Delta_2=6.0$, and $\Delta_3=0.0$. All the parameters are scaled by T_0.

Fig. 5(a). Here we put forward the threefold system depicted in Fig. 5(b). Two resonant control fields Ω_c and Ω_d are applied, and the other parameters have the same meaning as in the above twofold system. Provided that $\Delta=\pm\sqrt{\Omega_c^2+\Omega_d^2}$, two dark states will come into existence. Set φ to be $\tan^{-1}(\Omega_s\Omega_c/\sqrt{2}\Delta\Omega_p)$; then the adiabatic state

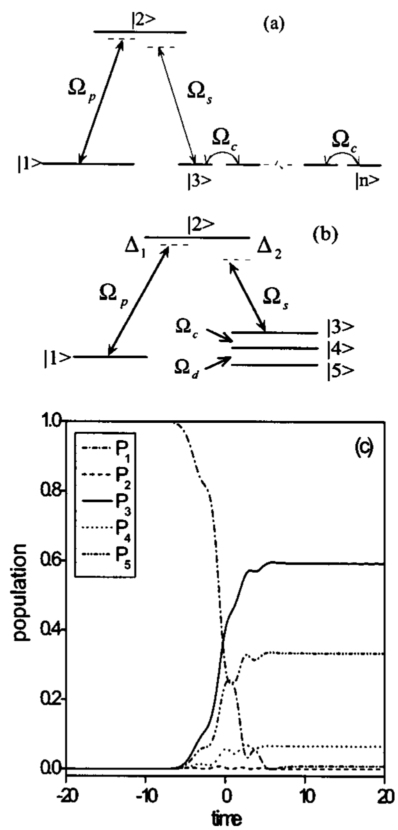

FIG. 5. (a), (b) The extended manifold level system. (c) The time evolutions of populations corresponding to the threefold level system as shown in (b). Parameters are $\Omega_p=\Omega_s=4.0$, $\Omega_c=3.0$, $\Omega_d=4.0$, $\Delta_1=4.0$, $\Delta_2=5.0$, and $\Delta_3=-1.0$. All the parameters are scaled by T_0.

$$|\psi\rangle_{\pm} = \sin\varphi|1\rangle - \cos\varphi(\Omega_c|3\rangle \pm |\Delta||4\rangle + \Omega_d|5\rangle)/\sqrt{\Delta^2 + \Omega_c^2 + \Omega_d^2}$$

can be realized. By adjusting the field parameters and the detunings, the desired single quantum state or superposition state can be constructed. Figure 5(c) presents an example of a numerical simulation. In a similar manner, *N*-component coherent superposition states can be created in the manifold system with a proper choice of parameters.

In summary, we have explored both analytically and numerically the creation of superpositions in a Λ-type system where the final state has two closely spaced levels. As long as Eq. (3) is satisfied, two arbitrary coherent superposition states with equal amplitude but inverse relative phases can be prepared without multiphoton resonance. Special attention has been paid to the case of the control field resonant with the corresponding transition frequency. It can be seen that under this condition $|\Phi\rangle_+$ and $|\Phi\rangle_-$ are orthogonal maximally coherent superposition states. A proper choice of parameters provides more freedom in manipulating the atomic coherent superposition states. Our scheme is usually feasible in a realistic experimental situation [18]. For example, one can place the sample cell in a microwave cavity to allow the interaction with a microwave control field. As for the adiabatic condition, it requires the strong coupling regime, that is, $\Omega_c T \gg 1$, which is not too difficult to reach in the microwave domain [19]. In brief, our scheme demonstrates the possibility of creating preselected atomic coherent superposition states.

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