Entangled-state preparation using adiabatic population transfer

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We describe an efficient and robust method for producing an entangled state of a two-spin system, using a sequence of two pulse pairs. We show that the mixing angle of the entangled state has a purely geometrical origin, so it is insensitive to small variations of the time-integrated pulse amplitude.

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I. INTRODUCTION

The recent developments in the theory of quantum information have stimulated interest in processes that could be used to entangle at least two quantum systems [1]. This paper presents a method for constructing a quantum superposition of two individual spin states,

$$|\Psi\rangle = \cos\Theta|\downarrow\rangle_1|\downarrow\rangle_2 + \sin\Theta|\uparrow\rangle_1|\uparrow\rangle_2, \qquad (1)$$

where $|\downarrow\rangle_j$ is the ''spin-down'' state and $|\uparrow\rangle_j$ is the ''spin-up'' state of the jth spin subsystem. If the mixing angle Θ is 0 or $\pi/2$ then the state $|\Psi\rangle$ is the product of two factors, each describing a definite state of one of the two spins. In general, the superposition state $|\Psi\rangle$ of Eq. (1) cannot be expressed as a single direct product of substates for the two spins, and so it is called an entangled state. When $\Theta=\pi/4$ the state $|\Psi\rangle$ is called the maximally entangled state of two quantum subsystems.

Contemporary work on quantum information processing [1] exploits the properties of coherent superpositions to form ''qubits,'' elements that offer information manipulation that is not possible with the conventional binary ''bits'' used in classical operations. When the superposed states are themselves composites of distinctly recognizable subsystems, such as is the case with the two spins discussed here, then the resulting entanglement of the probabilities of the two spins offers opportunities for applications that exploit the most remarkable features of quantum mechanics [1].

There are a few physical systems in which entanglement between quantum states of particles can be created and studied. Examples include entanglement of trapped ions [2–6], the atoms in a high-Q cavity [7] as well as nuclei entangled within molecules, as produced by nuclear-magnetic-resonance (NMR) methods [8].

These methods rely on one of the elementary properties of coherent excitation: the Rabi cycling of population between two states, as a result of a pulsed interaction [9]. The key parameter of such excitation is the Rabi frequency, essentially the interaction energy expressed in frequency units. By controlling the time integral of the pulsed Rabi frequency (the *pulse area*) one controls the number of Rabi cycles and thereby controls the mixing angle of the states. The mixing angle Θ is half the pulse area. When the pulse area is $\pi/2$, the mixing angle Θ is $\pi/4$, and the entanglement will be maximal.

Methods that rely on carefully controlled Rabi oscillations require careful control of pulse shape, as parametrized by the pulse area; in this sense they are not robust. Successful experimental realization of the basic components of quantum information processing devices will require fault-tolerant quantum logic gates [10,11]. Intensity-sensitive excitation can be avoided when one employs techniques that rely on another unusual property of quantum time evolution, the so-called Berry (or geometric) phase [12].

Berry [12] showed that when the Hamiltonian of a quantum system depends on a set of parameters (such as Rabi frequencies) which evolve along a closed curve in the parameter space, then a state vector corresponding to a simple nondegenerate eigenvalue develops a phase (the Berry phase) which depends only on the geometry of the curve in parameter space. The Berry phase was generalized to the case of degenerate levels by Wilczek and Zee [13].

Unlike the integral of the Rabi frequency (the dynamical phase), the geometrical phase does not depend on the duration of the interaction. It is therefore, for instance, independent of the speed with which an atom moves through an interaction region. If a quantum system undergoes random motion, due to some irregular interaction with the environment, the geometrical phase remains unchanged.

Recently Jones *et al.* [14] demonstrated experimentally a new approach to quantum computation with nuclear magnetic resonance. They implement a conditional Berry phase [12,15] and thus a controlled phase-shift gate. They used spin-half nuclei as an example to demonstrate this new geometrical approach, but the idea is more general.

In order to achieve excitation that does not depend on pulse area (i.e., a dynamical phase), we need to devise a procedure in which only the geometric phase occurs. To avoid any dynamical phase, we make the state vector a null-eigenvalue adiabatic state. Evolution of such a state has no dynamical phase. This idea was earlier used for the creation of an arbitrary superposition of two atomic states in a robust way using a sequence of three laser pulses in a four-state system [16–18]. That technique was based on the existence of two degenerate dark states (the null-eigenvalue adiabatic

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states) and their interaction. The mixing of the dark states [19] can be controlled by changing the relative delay of the pulses, and thus an arbitrary superposition state, can be generated. As suggested in Refs. [20], [21], and [22] one can modify the ordinary stimulated Raman adiabatic passage (STIRAP) [23] to create a superposition from a single quantum state by maintaining a fixed ratio of laser interactions. This method requires the existence of three quantum states.

The STIRAP method has been proposed for the creation of an entangled state for cavity QED two-atom systems [24] and for a Λ atomic systems [25].

In this paper we propose a simple method for entangling two subsystems. For definiteness we take these to be two spins. The method uses two-photon resonant excitation, and relies on the applicability of adiabatic evolution. It is similar to the tripod STIRAP method in the sense that it requires the existence of two degenerate dark states. However, the physical system differs from the tripod system. Here we consider a two-particle problem with strong interaction between them. We show that for a two-spin system the situation is more complicated than for a tripod system [16]. In the four-level tripod atomic system we have used sequences of absorption and emission processes in order to transfer part of the population from an initial state to a target state through an intermediate state. For a two-spin system two absorption processes are needed to create the entangled state defined by Eq. (1). As a consequence the final populations of the system will depend on a relative phase accumulated along excitation paths.

II. TWO-SPIN SYSTEM

Although the suggested method is applicable to any two-state system, we consider a simple model of two spins, denoted **I** and **S**, both of spin-1/2. We assume that the gyromagnetic ratios γ_S and γ_I are not the same.

As an example we can take a single ^{1}H nucleus as spin I, and a ^{13}C nucleus as spin S. These individual nuclear spins serve as the qubits. The spins are at rest in a static magnetic field of magnitude B_0 and interact weakly with each other via a scalar coupling $A\mathbf{I}\cdot\mathbf{S}$. This system has been used in an experiment [14]. The constant of the spin-spin interaction was $A\simeq 209.2$ Hz. Spin-spin relaxation times were 3.9 s for ^{1}H and 0.3 s for ^{13}C. The spin-lattice relaxation times were 7.6 s for ^{1}H and 25.3 s for ^{13}C. These relaxation times provide upper bounds for the time scale during which all processes must be completed.

We take the direction of the static magnetic field to define the z axis, which serves as the axis of quantization. The static Hamiltonian for this system, in the absence of pulsed excitation, is

$$H_{spin}=\gamma_S\hbar B_0 S_z+\gamma_I B_0 I_z+A\mathbf{I}\cdot\mathbf{S}. \tag{2}$$

We assume that the static magnetic field B_0 is strong enough so that we need to keep only AS_zI_z from the spin-spin coupling. In this approximation the Hamiltonian (2) can be represented as

$$H_{spin}=\gamma_S\hbar B_0 S_z+\gamma_I\hbar B_0 I_z+A\hbar I_z S_z. \tag{3}$$

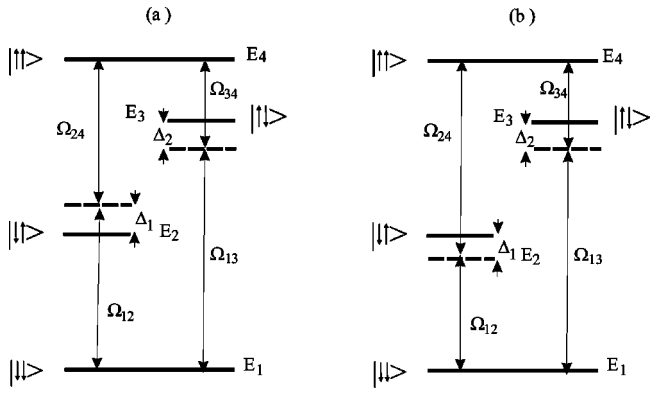

FIG. 1. Energy levels and linkages. (a) Opposite-sign detuning $\Delta_1=-\Delta_2$; (b) same-sign detuning $\Delta_1=\Delta_2$.

Because both I_z and S_z commute with the Hamiltonian of Eq. (3), their eigenvalues are constants of motion, given by

$$E_{m_S,m_I}=\gamma_S\hbar B_0 m_S+\gamma_I\hbar B_0 m_I+A\hbar m_S m_I \tag{4}$$

with the corresponding eigenvectors

$$|\Psi_{m_S,m_I}\rangle=|m_s\rangle|m_I\rangle. \tag{5}$$

We use uncoupled states as a basis with the following notation (see Fig. 1)

$$|\downarrow\rangle_S|\downarrow\rangle_I=|1\rangle,\quad E_1=-\frac{\hbar B_0(\gamma_S+\gamma_I)}{2}+\frac{A}{4}, \tag{6}$$

$$|\downarrow\rangle_S|\uparrow\rangle_I=|2\rangle,\quad E_2=-\frac{\hbar B_0(\gamma_S-\gamma_I)}{2}-\frac{A}{4},$$

$$|\uparrow\rangle_S|\downarrow\rangle_I=|3\rangle,\quad E_3=-\frac{\hbar B_0(\gamma_I-\gamma_S)}{2}-\frac{A}{4},$$

$$|\uparrow\rangle_S|\uparrow\rangle_I=|4\rangle,\quad E_4=\frac{\hbar B_0(\gamma_S+\gamma_I)}{2}+\frac{A}{4}.$$

We denote by E_j the static energy eigenvalue of state $|j\rangle$.

III. THE ROTATING WAVE APPROXIMATION SCHRÖDINGER EQUATION

In the absence of the spin-spin interaction, when $A=0$, the eigenvalue differences E_i-E_j yield two resonant frequencies,

$$\omega_S=\gamma_S B_0,\quad \omega_I=\gamma_I B_0.$$

With spin-spin coupling present each resonant frequency splits into two frequencies. The four frequencies are

$$\omega_S\pm\frac{A}{2},\quad \omega_I\pm\frac{A}{2}.$$

We assume that for each transition it is possible to apply a resonantly tuned radio frequency pulse, so that the Hamiltonian will include couplings between pairs of states as indicated in the loop configuration of Fig. 1.

Expanding the total wave function $|\Psi\rangle$ in the basis of the eigenstates given by Eqs. (6),

$$\begin{aligned}|\Psi(t)\rangle = & C_1(t)\exp[-iE_1t/\hbar]\exp[-i\omega_1t]|1\rangle \\ & + C_2(t)\exp[-iE_2t/\hbar]\exp[-i\omega_2t]|2\rangle \\ & + C_3(t)\exp[-iE_3t/\hbar]\exp[-i\omega_3t]|3\rangle \\ & + C_4(t)\exp[-iE_4t/\hbar]|4\rangle,\end{aligned}$$

and substituting it into the Schrödinger equation we obtain the set of differential equations for the amplitudes,

$$i\frac{d}{dt}\mathbf{C}(t) = \mathbf{W}(t)\mathbf{C}(t), \tag{7}$$

where, with the rotating wave approximation applied,

$$\mathbf{W} = \begin{bmatrix} 0 & \Omega_{12}(t)e^{i\chi} & \Omega_{13}(t) & 0 \\ \Omega_{12}(t)e^{-i\chi} & \Delta_1 & 0 & \Omega_{24}(t) \\ 0 & \Omega_{13}(t) & \Delta_2 & \Omega_{43}(t) \\ 0 & \Omega_{24}(t) & \Omega_{43}(t) & 0 \end{bmatrix}. \tag{8}$$

Here $\Delta_1 = \omega_{21} - \omega_1$, and $\Delta_2 = \omega_{31} - \omega_3$, $\omega_{nk} = (E_n - E_k)/\hbar$. We consider the case when the two-photon resonance condition holds between transitions 1 and 4,

$$E_1 + \omega_1 + \omega_3 = E_4 = E_1 + \omega_2 + \omega_4.$$

The Rabi frequencies $\Omega_{ij} = \gamma^{I,S}B_{ij}$, where B_{ij} are amplitudes of radio frequency pulses, have the following interpretation: Ω_{12} flips spin I when spin S is in the state $|\downarrow\rangle$, Ω_{43} flips spin I when spin S is in the state $|\uparrow\rangle$, Ω_{13} flips spin S when spin I is in the state $|\downarrow\rangle$, and Ω_{24} flips spin S when spin I is in the state $|\uparrow\rangle$. Without loss of generality we assume that Ω_{ij} are real and positive. Here $\chi = \chi_{12} - \chi_{24} + \chi_{43} - \chi_{13}$, where χ_{ij} are phases of the matrix elements of the transitions $i \leftrightarrow j$ $(i,j = 1,2,3,4)$, and is the relative phase between two paths of transitions from state 1 to the states 2 and 3 (absorption) and from state 4 to the same states 2 and 3 (emission). Most modern NMR spectrometers allow the control of the relative phase χ.

Using these equations we can restate the problem. We assume that spins are initially in a nonentangled state, specifically state $|1\rangle = |\downarrow\downarrow\rangle$, i.e., $C_1(-\infty) = 1$, and at the end of the interaction with the laser fields one wants to have state 1 entangled with state $|4\rangle = |\uparrow\uparrow\rangle$, i.e., $C_2(+\infty) = C_3(+\infty) = 0$ and $C_1(+\infty) \neq 0$, $C_4(+\infty) \neq 0$. A similar task was solved for a one-particle system using the tripod STIRAP method, Ref. [16], in which we applied three pulses under a two-photon resonance condition between initial and final states. We are here going to use this technique to create a superposition state for the four-level loop system (1) (see Fig. 1).

The amplitudes of the quantum states at the end of the interaction are obtained by numerically solving Eq. (7) with the initial conditions

$$C_1(-\infty) = 1, \quad C_2(-\infty) = 0, \quad C_3(-\infty) = 0, \quad C_4(-\infty) = 0 \tag{9}$$

for a model case assuming Gaussian pulses,

$$\begin{aligned}\Omega_{12}(t) &= \alpha_{12}\exp[-(t-\tau)^2/T^2], \\ \Omega_{13}(t) &= \alpha_{13}\exp[-(t-\tau)^2/T^2], \\ \Omega_{24}(t) &= \beta_{24}\exp[-(t+\tau)^2/T^2], \\ \Omega_{34}(t) &= \beta_{34}\exp[-(t+\tau)^2/T^2].\end{aligned} \tag{10}$$

We consider two general pulse sequences: ''intuitively'' ordered, in which initially populated states are accessed by the initial radiation, and ''conterintuitively'' ordered, in which only unoccupied states are initially linked. Figure 2 shows these sequences. We also consider two classes of detunings: opposite signed, $\Delta_1 = -\Delta_2$ [see Fig. 1(a)] and same signed, $\Delta_1 = \Delta_2$ [see Fig. 1(b)]. We will show below that in order that there be two dark states we require $\chi = 0$ [see Eq. (8)]. Therefore we have assumed $\chi = 0$ in the modeling. Without loss of generality we consider $\alpha = \alpha_{12} = \alpha_{13}$ and $\beta = \beta_{24} = \beta_{34}$.

Figure 3 shows the probabilities obtained from numerical solution of Eqs. (7) with the initial conditions (9) as function of delay for the opposite-signed detunings $\Delta_1 = -\Delta_2 = 5/T$ [see Fig. 1(a)]. The peak Rabi frequencies are $\alpha = \beta = 10/T$, where T characterizes the pulse duration, see Eq. (10). Negative delays τ correspond to counterintuitive ordering of pulses and positive τ corresponds to intuitive ordering of pulses. We see from this figure that the final populations of the states 1 and 4 are the same for either ordering of the pulses.

Furthermore we can control the final state of the quantum system by changing the parameter τ/T. For example, when $|\tau/T| \approx 0.25$ or 1 we have a maximal entanglement between two spins. Of particular interest is the observation, for the same-signed detunings [see Fig. 1(b)] $\Delta_1 = \Delta_2 = 5/T$ (see Fig. 4), that the final populations of the states 1 and 4 are tolerant to (small) change of the parameter τ/T near $|\tau/T| \approx 0.6$. Because the population is either in state 4 or in state 1, this choice of the detunings does not lead to an entangled state. This behavior of the population is similar to what occurs with STIRAP [23], but here we have a robust population transfer from the initial state to the target state for both the intuitive and counterintuitive pulse orderings. The insensitivity to the pulse ordering is due to large intermediate detunings, $\Delta T \gg 1$.

This method differs from the well-known quantum Rabi nutation method [9] as shown in Fig. 5, which displays how the atomic populations at the end of the interaction depend on the pulse area for the counterintuitive sequences of pulses. It is well known that with the area method the

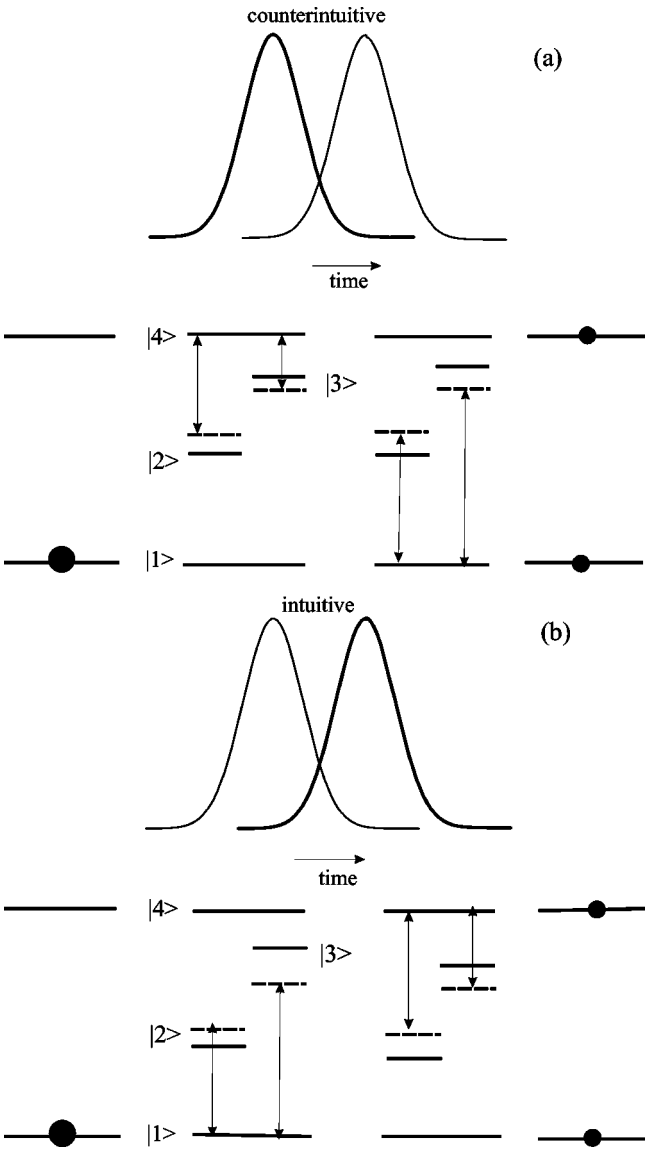

FIG. 2. Pulse sequence and linkages for (a) counterintuitive and (b) intuitive pulse orderings.

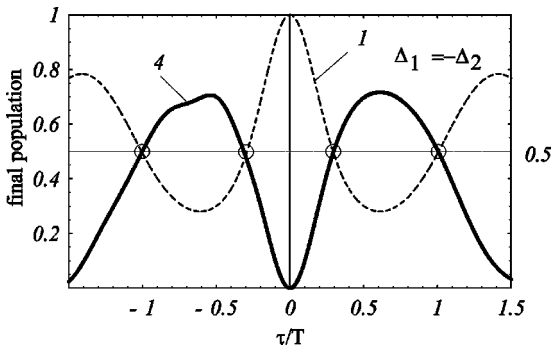

FIG. 3. Probability for finding the population in state 1 (dashed line) or state 4 (solid line) after the interaction with the radiation field as a function of τ/T. The parameters are $\Delta_1=-\Delta_2=5/T$, $\alpha=\beta=10/T$. Negative τ correspond to the intuitive sequences of pulses, and for positive case counterintuitive order. Maximum entanglement is reached for several values of τ/T indicated by the circles.

populations oscillate with increasing area of the laser pulse. However, we see from Fig. 5 that a change of αT causes a monotonic variation of the populations. Thus for the opposite-signed detunings $\Delta_1=-\Delta_2$ we can control the final populations of the state $|1\rangle$ and $|4\rangle$ by changing external parameters, for instance τ/T. In the next section we develop, for this case, an analytical solution of the Schrödinger equation (7) in the adiabatic limit.

IV. ADIABATIC APPROXIMATION

A. Complete population transfer

In order to understand the physical mechanism of the robust population transfer from the initial state $|1\rangle=|\downarrow\downarrow\rangle$ to the target state $|4\rangle=|\uparrow\uparrow\rangle$, or the creation of a controlled superposition of these states, we consider in this section the same-signed $\Delta_1=\Delta_2=\Delta$ in the adiabatic approximation. We consider the case when $\Omega_{12}(t)=\Omega_{13}(t)=\Omega_1(t)$ and $\Omega_{24}(t)=\Omega_{34}(t)=\Omega_4(t)$.

For this particular case the eigenvalues of the Hamiltonian $\mathbf{W}(t)$ have the following form

$$\lambda_{1,2}(t)=\tfrac{1}{2}\left(\Delta\pm\sqrt{\Delta^2+4\Omega_4^2(t)+4\Omega_1^2(t)+4\sqrt{\Omega_4^4(t)+\Omega_1^4(t)+2\Omega_1^2(t)\Omega_4^2(t)\cos\chi}}\right),$$

$$\lambda_{3,4}(t)=\frac{1}{2}\left(\Delta\pm\sqrt{\Delta^2+4\Omega_4^2(t)+4\Omega_1^2(t)-4\sqrt{\Omega_4^4(t)+\Omega_1^4(t)+2\Omega_1^2(t)\Omega_4^2(t)\cos\chi}}\right).$$

All eigenvalues of the matrix $\mathbf{W}(t)$ are different except for the case when $\chi=\pi$ and $\Omega_4(t)=\Omega_1(t)$. The formulas for eigenvectors of the Hamiltonian $\mathbf{W}(t)$ in the general case of same-signed when $\Delta_1=\Delta_2=\Delta$, are lengthy. We limit the discussion to the case when $\chi=0$. For this case the following eigenvectors are

$$\Phi_1(t)=\begin{bmatrix}\cos\vartheta(t)\\0\\0\\-\sin\vartheta(t)\end{bmatrix}, \tag{11}$$

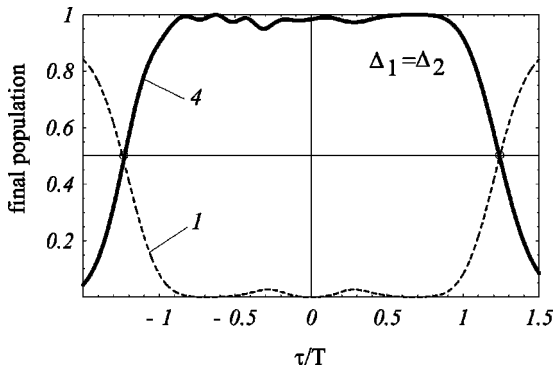

FIG. 4. The same as for Fig. 3, but $\Delta_1=\Delta_2=5/T$.

where

$$\tan\vartheta(t)=\frac{\Omega_1(t)}{\Omega_4(t)} \tag{12}$$

and

$$\Phi_2(t)=\frac{1}{N(t)}\begin{bmatrix}\Omega_1(t)\\ \frac{1}{4}(\Delta-\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})\\ \frac{1}{4}(\Delta-\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})\\ \Omega_4(t)\end{bmatrix}, \tag{13}$$

$$\Phi_3(t)=\frac{1}{N(t)}\begin{bmatrix}\Omega_1(t)\\ \frac{1}{4}(\Delta+\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})\\ \frac{1}{4}(\Delta+\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})\\ \Omega_4(t)\end{bmatrix}, \tag{14}$$

where $N(t)$ is a normalizing function. The fourth eigenstate remains fixed at all times, and

$$\Phi_4(t)=\frac{1}{\sqrt{2}}\begin{bmatrix}0\\1\\-1\\0\end{bmatrix}. \tag{15}$$

The eigenvalues are 0, $\frac{1}{2}(\Delta-\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})$, $\frac{1}{2}(\Delta+\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})$ and Δ, respectively. Next we show that for both intuitive and counterintuitive pulse ordering robust population transfer from the initial state 1 to the target state 4, occurs in the adiabatic limit $\Delta T\gg 1$ and $\max\{\Omega_1(t),\Omega_4(t)\}T\gg 1$.

The time evolution of the system for counterintuitive pulse ordering is the same as for STIRAP. The system remains, at all times, in the trapped state equation (11) and the state vector of the system rotates from coincidence with state 1 to coincidence with state 4. For the intuitive sequences of pulses the state vector of the system coincides with the vector of Eq. (13) and robust population transfer occurs from the initial state 1 to the state 4. However, during the time of evolution the final state 4 will accumulate an additional phase

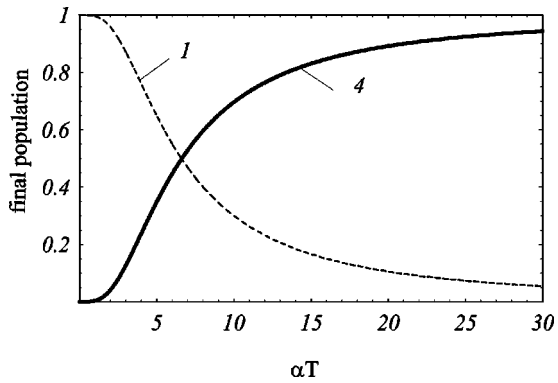

FIG. 5. The final population of the states 1 (dashed line) and 4 (solid line) as a function of the area of laser pulse αT. The parameters are $\Delta_1=-\Delta_2=5/T$, $\tau/T=0.5$, $\alpha=\alpha_{12}=\alpha_{13}=\beta_{24}=\beta_{34}$.

$$\frac{1}{2}\int_{-\infty}^{+\infty}(\Delta-\sqrt{\Delta^2+8\Omega_1^2(t)+8\Omega_4^2(t)})dt.$$

This latter case is of interest when population transfer from the initial state 1 to the target state 4 is wanted, but is not suitable for the creation of the coherent-superposition problem of Eq. (1).

B. Partial population transfer: Creation of an entangled state

Next, we consider the opposite-signed detuning case when $\Delta_1=-\Delta_2=\Delta$. For this case we find the following four different eigenvalues of the Hamiltonian $\mathbf{W}(t)$:

$$\lambda_{1,2}(t)=\pm\frac{1}{\sqrt{2}}\sqrt{\Omega_0^2(t)-\sqrt{\Omega_0^4(t)-16\Omega_1^2(t)\Omega_4^2(t)\sin^2\frac{\chi}{2}}}, \tag{16}$$

$$\lambda_{3,4}(t)=\pm\frac{1}{\sqrt{2}}\sqrt{\Omega_0^2(t)+\sqrt{\Omega_0^4(t)-16\Omega_1^2(t)\Omega_4^2(t)\sin^2\frac{\chi}{2}}},$$

where

$$\Omega_0(t)=\sqrt{2\Omega_1^2(t)+2\Omega_4^2(t)+\Delta^2}. \tag{17}$$

For vanishing accumulated relative phase of the excitation, $\chi=0$, the Hamiltonian has two degenerate eigenvalues $\lambda_1(t)=\lambda_2(t)=0$. In this case it is easy to show that the matrix $\mathbf{W}(t)$ is unitarily equivalent to the tripod Hamiltonian $\tilde{\mathbf{W}}(t)$ of Refs. [16] and [17]

$$\mathbf{W}(t)=\mathbf{U}\tilde{\mathbf{W}}(t)\mathbf{U}^{\dagger}, \tag{18}$$

where

$$\mathbf{U}=\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}.$$

According to Refs. [16] and [17] the matrix

$$\tilde{\mathbf{W}}(t)=\begin{bmatrix} 0 & \sqrt{2}\Omega_1(t) & 0 & 0 \\ \sqrt{2}\Omega_1(t) & 0 & \Delta & \sqrt{2}\Omega_4(t) \\ 0 & \Delta & 0 & 0 \\ 0 & \sqrt{2}\Omega_4(t) & 0 & 0 \end{bmatrix}$$

has two degenerated eigenvalues. Using the transformation of Eq. (18) we find the eigenvectors of the matrix $\mathbf{W}(t)$ in terms of two time-dependent angles $\vartheta(t)$ Eq. (12) and $\varphi(t)$:

$$\tan\varphi(t)=\frac{\Delta}{\sqrt{2\Omega_1^2(t)+2\Omega_4^2(t)}}. \tag{19}$$

The two degenerate null-eigenvalue eigenvectors are [16,17]

$$\Phi_1(t)=\begin{bmatrix} \cos\vartheta(t) \\ 0 \\ 0 \\ -\sin\vartheta(t) \end{bmatrix},$$
$$\Phi_2(t)=\frac{1}{\sqrt{2}}\begin{bmatrix} \sqrt{2}\sin\vartheta(t)\sin\varphi(t) \\ -\cos\varphi(t) \\ \cos\varphi(t) \\ \sqrt{2}\cos\vartheta(t)\sin\varphi(t) \end{bmatrix} \tag{20}$$

while the remaining eigenvectors are

$$\Phi_3(t)=\frac{1}{\sqrt{2}}\begin{bmatrix} \sin\vartheta(t)\cos\varphi(t) \\ \frac{1}{\sqrt{2}}(1+\sin\varphi(t)) \\ \frac{1}{\sqrt{2}}(1-\sin\varphi(t)) \\ \cos\vartheta(t)\cos\varphi(t) \end{bmatrix},$$
$$\Phi_4(t)=\frac{1}{\sqrt{2}}\begin{bmatrix} \sin\vartheta(t)\cos\varphi(t) \\ \frac{1}{\sqrt{2}}(\sin\varphi(t)-1) \\ -\frac{1}{\sqrt{2}}(1+\sin\varphi(t)) \\ \cos\vartheta(t)\cos\varphi(t) \end{bmatrix}. \tag{21}$$

The analytic solution of the Schrödinger equation is found for the limiting case of adiabatic evolution defined by the conditions [16,17]

$$\frac{d\vartheta(t)}{dt}\ll\Omega_0(t), \quad \frac{d\varphi(t)}{dt}\ll\Omega_0(t). \tag{22}$$

The angles $\vartheta(t)$ and $\varphi(t)$, defined in Eqs. (12) and (19), reach the asymptotic values $\vartheta(-\infty)=0$, $\varphi(-\infty)=\pi/2$ for the counterintuitive sequence of pulse [the quantum system interacts first with the pulse $\Omega_1(t)$ and then with $\Omega_4(t)$]. In this case the vectors Φ_n $n=1,2,3,4$ have the components

$$\Phi_1(-\infty)=\begin{bmatrix} 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad \Phi_2(-\infty)=\begin{bmatrix} 0 \\ 0 \\ 0 \\ 1 \end{bmatrix},$$
$$\Phi_3(-\infty)=\begin{bmatrix} 0 \\ 1 \\ 0 \\ 0 \end{bmatrix}, \quad \Phi_4(-\infty)=\begin{bmatrix} 0 \\ 0 \\ 1 \\ 0 \end{bmatrix}. \tag{23}$$

For applications to quantum gates it is necessary to consider all possible transitions. When the system is in the adiabatic states Φ_3 or Φ_4, the spin system is prepared initially in the state $|2\rangle=|\uparrow\rangle_S|\downarrow\rangle_I$ or $|3\rangle=|\downarrow\rangle_S|\uparrow\rangle_I$. The radiative interaction induces the following transformations at the end of the interaction:

$$|2\rangle\rightarrow|2\rangle e^{-i\delta} \quad \text{and} \quad |3\rangle\rightarrow-|3\rangle e^{i\delta}, \tag{24}$$

where

$$\delta=\int_{-\infty}^{+\infty}\Omega_0(t)dt$$

is a dynamical phase. The same transformation applies for the intuitively ordered sequence of pulses $[\vartheta(-\infty)=\pi/2,\varphi(-\infty)=\pi/2]$.

The assumption that $\mathbf{W}(t)$ varies adiabatically implies that if the system is initially in one of the states Φ_1 or Φ_2 it will not acquire any component Φ_3 or Φ_4. However, due to the degeneracy of $\Phi_1(t)$ and $\Phi_2(t)$, the transitions between these two states cannot be neglected. For the second case, when the system is in the state $|1\rangle=|\downarrow\rangle_I|\downarrow\rangle_S$ or $|4\rangle=|\uparrow\rangle_I|\uparrow\rangle_S$ before the interaction, the situation is more complicated. The initial state coincides with the adiabatic state $\Phi_1(-\infty)$ or $\Phi_2(-\infty)$ [see Eq. (23)] depending on the ordering of pulses. Using the transformation (18) and the method that has been used in Refs. [16] and [17], we can solve the Schrödinger equation (7) in the adiabatic limit (22). For the initial conditions (9), the solution to these equations at the end of the interactions reads

$$C_1(+\infty)=\cos\Theta, \quad C_4(+\infty)=\sin\Theta, \tag{25}$$

where

$$\Theta = \int_{-\infty}^{\infty} d\tau \frac{d\vartheta}{d\tau} \sin\varphi(\tau) \tag{26}$$

for the counterintuitive sequence of pulses. Thus the evolution is described by the transformation

$$|1\rangle = |\downarrow\rangle_S|\downarrow\rangle_I \rightarrow \cos\Theta|\downarrow\rangle_S|\downarrow\rangle_I + \sin\Theta|\uparrow\rangle_S|\uparrow\rangle_I . \tag{27}$$

If the system is initially in the state $|4\rangle = |\uparrow\rangle_S|\uparrow\rangle_I$, it transits to an entangled state

$$|4\rangle = |\uparrow\rangle_S|\uparrow\rangle_I \rightarrow \cos\Theta|\downarrow\rangle_S|\downarrow\rangle_I - \sin\Theta|\uparrow\rangle_S|\uparrow\rangle_I . \tag{28}$$

The intermediate states $|\uparrow\rangle_S|\downarrow\rangle_I$ and $|\downarrow\rangle_S|\uparrow\rangle_I$ are crucial as intermediate states during the adiabatic process but remain unpopulated for $t = +\infty$.

It has been proved [26] that any transformation involving a number of qubits can be constructed using only single-qubit operations and a two-qubit operation. Thus with the operations described by Eqs. (24), (27), and (28) plus single-qubit operations, it is possible to construct any unitary transformation.

It is instructive to compare the formulas, Eqs. (27) and (28), with the usual formula expanding two-state excitation probabilities as a function of pulse area [5]. As was already noted above, the present atomic system is always in a superposition of states Φ_1 or Φ_2, so in the adiabatic limit, the dynamical phase is absent.

V. GEOMETRIC INTERPRETATION

As shown in Ref. [17] the phase Θ has a geometrical interpretation. We consider a three-dimensional space in which the coordinates are the values of Rabi frequencies $\Omega_1(t)$ and $\Omega_4(t)$ and detuning. At any time instant of time the Hamiltonian is represented by a vector in this space, $\mathbf{R} = (\Omega_1, \Omega_4, \Delta)$. As time increases, the vector $\mathbf{R}$ traces out a trajectory. The integral (26) can be evaluated using Stokes' theorem. After simple calculations the integral can be presented in the form

$$\Theta = \int_S \mathbf{V} d\mathbf{S}, \tag{29}$$

where

$$\mathbf{V} = \frac{\mathbf{R}}{R^3} \tag{30}$$

and S is an oriented surface that is bounded by a closed curve in the parameter space. Thus Eq. (29) expresses the phase Θ as the flux through S of the field of a monopole with unit strength located at the point of degeneracy $(\Omega_1 = 0, \Omega_4 = 0, \Delta = 0)$ in the parameter space. It is easy to show that Θ depends only on ratios $\Delta/\Omega_{1,4}$ and τ/T, where τ is an effective delay between pulses and T is the duration of the interaction. The phase Θ is independent of how the process is carried out as long as it is slow enough to satisfy to the adiabatic conditions (22). The formulas (29) and (30) are analogous to those of Berry for the spin system in the external magnetic field [12].

VI. CONCLUSION

In this paper we have shown that, in the adiabatic limit and with two-photon resonant excitation, it is possible to prepare an entangled state of two two-level systems in a robust and controlled way. When the pulses are appropriately delayed in time, efficient population transfer from the initial state $|1\rangle = |\downarrow\downarrow\rangle$, to the state $|4\rangle = |\uparrow\uparrow\rangle$, is achieved if the detuning of the relevant transition frequencies (see Fig. 4) has the same sign. When that detuning is of opposite sign, a two-particle entangled state is formed.

There are many ways to realize a loop (see Fig. 1) linkage pattern within quantum systems examined in studies of NMR, such as in experiment [14], where 500-MHz radiation with the maximum radio frequency field strength (in Hz) up to 774 Hz was used. The entangled state does not depend on the exact values of the Rabi frequencies as long as the conditions for adiabatic following are satisfied.

We have shown that the superposition angle has a geometrical interpretation. Thus, our method may open new possibilities for robust geometric quantum information processing without dynamical phases. It is important to note that the proposed method is quite general and it can be applied on the two-level atomic system for generation of entangled states.

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