Quantum control of entanglement by phase manipulation of time-delayed pulse sequences. I

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We investigate coherent population transfer in double-Λ and diamond-type four-level systems driven by two pairs of time-delayed laser pulses. We show that the relative phase between the pulses induces interference so that the adiabatic passage dynamics is only possible for a particular phase. Both pulse area and phase are control knobs that can be used to prepare specific coherent superpositions using counterintuitive and intuitive pulse sequences. Since the diamond-type four-level system maps the Hamiltonian of two interacting two-level systems, the schemes can be used to manipulate entanglement. In particular we propose robust schemes to prepare specific entangled states by phase control with time-delayed pulse sequences.

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I. INTRODUCTION

Stimulated Raman adiabatic passage (STIRAP) is a powerful method of selective population transfer in quantum systems. The original idea is to use two time-delayed pulses for efficient population transfer between two lower states with no direct dipole-allowed transition in a Λ-type system [1,2]. It was found that a counterintuitive pulse sequence prepares the system in the so-called adiabatic dark state. 100% population transfer to the target state is achieved without populating the excited state, as long as the adiabatic condition is satisfied. The main feature of the method is the robustness with respect to moderate changes in the excitation parameters, which makes STIRAP a very attractive scheme for experimental implementations. Different generalizations of STIRAP for multilevel systems as well as for population transfer through the continuum have been proposed and tested [3].

Here we study population transfer by time-delayed pulse sequences, similar to those in the STIRAP scheme, in a four-level system with two intermediate levels coupled to both the initial and final states, which is called a double-Λ system [Fig. 1(b)]. It is known that in situations of incomplete control (for instance, if both intermediate states are addressed by the same pump and Stokes pulses) adiabatic passage is very sensitive to the signs (the phase) of the dipole matrix elements [4]. In this paper we assume complete controllability of the four transitions, so that the sensitivity of the adiabatic passage dynamics to the dipole moment is transferred to the relative phase of the pulses. We will show that phase-controlled interference can be used to prepare specific superposition states at the expense of reducing the stability of the dynamics to changes in the pulse area.

The influence of the phase on the dynamics may have very interesting applications. In the rotating wave approximation (RWA) the Hamiltonian of the double-Λ system is equivalent to the Hamiltonian of two interacting two-level systems if each of the two-level systems (the qubits) has a distinct transition frequency or, otherwise, if an external homogeneous field is applied to distinguish between the different qubits. In both cases the two-qubit system behaves as a four level system with closed-loop couplings. Four different pulses can then be applied to perform logic operations in the composite quantum system. We will show how the different entangled states can be prepared by manipulating the relative phase and the pulse areas. In fact, the wave function of any closed-loop system can be controlled by changing the relative phase of the external fields [5] and several interesting control scenarios are possible by mixing the properties of adiabatic pulse sequences and phase control [6]. In this contribution we concentrate on time-delayed pulse sequences, both in counterintuitive (the probe or Stokes pulse preceding the pump pulse) and intuitive sequences. In the accompanying paper [6] we study the effect of partially time-delayed pulse sequences. Other theoretical studies relevant to the present work include coherent population transfer in closed-

![FIG. 1. Scheme of a closed-loop four-level system in diamond (a) and double-Λ (b) configuration.](image-url)
loop systems [7] and preparation of entangled states by STIRAP using proper time delay and detuning [8].

II. GENERAL REMARKS ON PHASE CONTROL OF 2-QUBITS

We consider the Hamiltonian of two interacting two-level quantum systems (for example, two $1/2$ spin particles, two quantum dots, or two Rydberg atoms). Typically such a system is described by the four level system in a closed-loop configuration. We consider all transitions to be addressed and controlled independently by radio frequency or optical pulses. In the rotating wave approximation (RWA) this system is equivalent to a double-$\Lambda$ system in which each transition is driven by a distinct laser pulse. Both possible configurations are represented in Fig. 1. The independent control of the four transitions implies that either the energies of the four states are not degenerate (due, for instance, to the inter-system coupling) or that there are symmetry rules that allow one to distinguish the resonant transitions.

The total wave function can be written as $|\psi(t)\rangle = a_1(t)|00\rangle + a_2(t)|11\rangle + b_1(t)|01\rangle + b_2(t)|10\rangle$. Alternatively, in the product state basis (|ij⟩ = |i⟩|j⟩), where i, j = 0, 1, or ground state and excited state, respectively) the total wave function of the system is

$$|\psi(t)\rangle = a_1(t)|00\rangle + a_2(t)|11\rangle + b_1(t)|01\rangle + b_2(t)|10\rangle.$$ (1)

In what follows we mainly adopt the nomenclature of Quantum Information. The Schrödinger equation (in the RWA) for the case of two-photon resonance between the states $|00\rangle$ ($|11\rangle$) and $|10\rangle$ ($|01\rangle$) has the form

$$i\frac{\dot{a}_1(t)}{2} = \Omega_{p1}(t) a_1(t) + \Omega_{p2}(t) b_2(t)e^{-i\phi},$$

$$i\frac{\dot{a}_2(t)}{2} = \Omega_{p1}(t) a_2(t) + \Omega_{p2}(t) b_2(t)e^{-i\phi},$$

$$i\frac{\dot{b}_1(t)}{2} = \Omega_{s1}(t) b_1(t) + 2\Delta_1 a_1(t),$$

$$i\frac{\dot{b}_2(t)}{2} = \Omega_{s2}(t) b_2(t) e^{-i\phi} + 2\Delta_2 a_2(t),$$

where $\Omega_{p,i}(t)$ are the Rabi frequencies chosen to be real, $\Delta_{1,2}$ are one-photon detunings and we explicitly write the phase factor $e^{-i\phi}$. In the Appendix we show that $\phi$, the relative phase between both two-photon pathways, characterizes the dynamics in the closed-loop system. Therefore choosing all Rabi frequencies real involves no approximation. From the general form of the Hamiltonian it is clear that there are two possible pathways that connect state $|00\rangle$ to state $|11\rangle$, and

$$\Omega_{p1}(t) \Omega_{s1}(t)$$

$|00\rangle \rightarrow |01\rangle \rightarrow |11\rangle$

and

$$\Omega_{p2}(t) \Omega_{s2}(t)$$

$|00\rangle \rightarrow |10\rangle \rightarrow |11\rangle$.

The interference between these two channels is ultimately responsible for preparing any possible coherent superposition (entangled state) of the form $|\Psi^\beta\rangle = (|00\rangle + e^{i\beta}|11\rangle)/\sqrt{2}$. Moreover, since the system is closed, if initially the population is transferred, for instance, to state $|01\rangle$, then the two pathways,

$$\Omega_{p1}(t) \Omega_{s1}(t)$$

$|01\rangle \rightarrow |00\rangle \rightarrow |10\rangle$

and

$$\Omega_{p2}(t) \Omega_{s2}(t)$$

$|01\rangle \rightarrow |11\rangle \rightarrow |10\rangle$,

allow the preparation of any superposition (entangled state) of the form $|\Psi^\beta\rangle = (|01\rangle + e^{i\beta}|10\rangle)/\sqrt{2}$. Here and in the following paper [6] we will show that it is possible to prepare all types of entangled states starting from $|00\rangle$ by using simple sequences of pulses, typically designed for adiabatic passage of population, and controlling the phase $\phi$. In the proposed schemes we do not require independent control on the shape and amplitude of the different Rabi frequencies, so that we make hereafter $\Omega_{p1}(t) = \Omega_{p2}(t) = \Omega_p(t)$ and $\Omega_{s1}(t) = \Omega_{s2}(t) = \Omega_s(t)$ to simplify the equations. By defining

$$b_{\pm}(t) = [b_1(t) \pm e^{i\phi} b_2(t)]/\sqrt{2},$$

we obtain, for the new basis, the set of equations:

$$\dot{a}_1(t) = i\frac{\sqrt{2}}{2}\Omega_p b_{+}(t) \cos \frac{\phi}{2} b_+(t) + i \frac{\sqrt{2}}{2} \Omega_p a_1(t) a_2(t) + i \Delta_1 b_+(t)$$

$$\dot{a}_2(t) = i\frac{\sqrt{2}}{2}\Omega_p b_{-}(t) \sin \frac{\phi}{2} b_-(t)$$

$$\dot{b}_+(t) = \frac{\sqrt{2}}{2}\Omega_p (e^{i\phi/2} \Omega_{s2}(t) e^{-i\phi/2} \sin \frac{\phi}{2} a_1(t) + i \Delta_+ b_+(t) + i \Delta_- b_-(t).$$

The four states can be decoupled for $\phi = 0$ and $\phi = \pm\pi$. In the first case the equations simplify as follows:

$$\dot{a}_1(t) = i\frac{\sqrt{2}}{2}\Omega_p b_+(t),$$

$$\dot{a}_2(t) = i\frac{\sqrt{2}}{2}\Omega_p b_+(t),$$

$$\dot{b}_+(t) = i\frac{\sqrt{2}}{2}[\Omega_p(t)a_1(t) + \Omega_s(t)a_2(t)] + i \Delta_+ b_+(t) + i \Delta_- b_-(t),$$

$$\dot{b}_-(t) = i \Delta_+ b_+(t) + i \Delta_- b_-(t).$$

For equal detuning $\Delta_1 = \Delta_2 = \Delta$ ($\Delta_2 = 0$) we obtain

$$\dot{a}_1(t) = i\frac{\sqrt{2}}{2}\Omega_p b_+(t),$$

$$\dot{a}_2(t) = i\frac{\sqrt{2}}{2}\Omega_p b_+(t),$$

$$\dot{b}_+(t) = i\frac{\sqrt{2}}{2}[\Omega_p(t)a_1(t) + \Omega_s(t)a_2(t)] + i \Delta_+ b_+(t) + i \Delta_- b_-(t),$$

$$\dot{b}_-(t) = i \Delta_+ b_+(t) + i \Delta_- b_-(t).$$

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\[ \dot{a}_2(t) = \frac{i}{\sqrt{2}} \Omega_s b_s(t), \]

\[ \dot{b}_s(t) = \frac{i}{\sqrt{2}} [\Omega_p(t) a_1(t) + \Omega_s(t) a_2(t)] + i \Delta b_s(t), \]

\[ \dot{b}_- (t) = i \Delta b_-(t), \]

where the last equation is decoupled. In this three-level system, \( \pi \)-pulse or adiabatic passage strategies can be used to control the final population between states \( |0\rangle, |1\rangle \) and the entangled state \( |\Psi^\theta\rangle = (|01\rangle + |10\rangle)/\sqrt{2} \). On the other hand, the entangled state \( |\Psi^\pi\rangle = (|01\rangle - |10\rangle)/\sqrt{2} \) is inaccessible and cannot be prepared by any scheme for \( \phi = 0 \) in the absence of asymmetric detuning. Following the usual nomenclature for the Bell states we shall call \( |\Psi^0\rangle = |\Psi^\pi\rangle \), \( |\Psi^1\rangle = |\Psi^\pi\rangle \), \( |\Psi^2\rangle = |\Phi^+\rangle \), \( |\Phi^-\rangle \), and \( |\Phi^+\rangle = |\Phi^-\rangle \).

In the case \( \phi = \pm \pi \) the set of equations simplifies to

\[ \dot{a}_1(t) = \frac{i}{\sqrt{2}} \Omega_p b_-(t), \]

\[ \dot{a}_2(t) = \frac{i}{\sqrt{2}} \Omega_s b_+(t), \]

\[ \dot{b}_+ (t) = \frac{i}{\sqrt{2}} [\Omega_p(t) a_1(t) + \Omega_s(t) a_2(t)] + i \Delta b_+(t), \]

For equal detunings the system decouples into two independent two-level systems. Notice that according to Eq. (3), \( b_-(t) \) corresponds to the amplitude of \( |\Psi^\pi\rangle \) for \( \phi = \pm \pi \). Now the initial state \( |00\rangle \) is only coupled to the entangled state \( |\Psi^\theta\rangle \). In this scenario no adiabatic passage is possible using transformed limited pulses.

In summary, efficient population transfer via STIRAP is a possible solution with this Hamiltonian, using a counterintuitive pulse sequence with both Stokes pulses preceding both pump pulses and phase \( \phi = 0 \) [1,2]. However, the relative phase between two pathways in a closed-loop configuration is a more powerful control knob that determines the symmetry of the problem and enlarges the set of states that are accessible. We analyze the role of the phase in both counterintuitive (CIN) and intuitive (IN) type sequences.

### III. Dressed States Analysis

In order to understand the dynamics of the system as a function of phase and Rabi frequency we obtain the eigenvalues and eigenvectors of the Hamiltonian [Eq. (2)] for the case \( \Delta_1 = \Delta_2 = 0 \). The analytical expressions of the eigenvalues are \( \lambda_{1,2} = \mp \lambda_s(t)/2 \), \( \lambda_{3,4} = \mp \lambda_p(t)/2 \), where \( \lambda_p(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t) + \Omega_0^2} \) and \( \Omega_0^2 = \Omega_p^2(t) + \Omega_s^2(t) + 2 \cos \phi \Omega_p(t) \Omega_s(t) \). The analytical expressions of the dressed states are

\[ |c_1(t)\rangle = \frac{1}{\sqrt{2}} \left( \frac{\lambda_1 \Omega_p^2(t) - \Omega_s^2(t) + \Omega_0^2(t)}{\Omega_p(t) \Omega_s(t)} \right) |0\rangle + \frac{\Omega_0^2(t) + \Omega_p^2(t)}{\Omega_p(t) \Omega_s(t)} |1\rangle, \]

\[ |c_2(t)\rangle = \frac{1}{\sqrt{2}} \left( \frac{\lambda_1 \Omega_p^2(t) - \Omega_s^2(t) - \Omega_0^2(t)}{\Omega_p(t) \Omega_s(t)} \right) |0\rangle - \frac{\Omega_0^2(t) + \Omega_p^2(t)}{\Omega_p(t) \Omega_s(t)} |1\rangle, \]

\[ |c_3(t)\rangle = \frac{\xi}{\sqrt{2}} \left( \frac{\lambda_1 \Omega_p^2(t) + \Omega_s^2(t)}{\Omega_p(t) \Omega_s(t)} \right) |0\rangle + \frac{\Omega_0^2(t) - \Omega_p^2(t)}{\Omega_p(t) \Omega_s(t)} |1\rangle, \]

\[ |c_4(t)\rangle = \frac{\xi}{\sqrt{2}} \left( \frac{\lambda_1 \Omega_p^2(t) - \Omega_s^2(t)}{\Omega_p(t) \Omega_s(t)} \right) |0\rangle - \frac{\Omega_0^2(t) - \Omega_p^2(t)}{\Omega_p(t) \Omega_s(t)} |1\rangle, \]

where \( \xi = \sqrt{[\Omega_p^2(t) + \Omega_s^2(t)] [\Omega_p^2(t) + \Omega_s^2(t)]} \).

We consider excitation of the four-level system by CIN and IN sequences. In the first case, a pair of Stokes pulses precedes a pair of pump pulses and they overlap for some time, approximately equal to the half-width of the pulses. In the intuitive case, the pump and Stokes pulses exchange places in time.

Taking the limits for \( \Omega_p(t)/\Omega_s(t) \to 0, \infty \) we determine the correlation between the dressed states and the bare states at initial and final times. For the limit \( \Omega_p(t)/\Omega_s(t) \to 0 \) we obtain

\[ |c_1(t)\rangle \approx \left( \frac{1}{\sqrt{2}}, 0, -\frac{ie^{i\phi/2}}{2}, \frac{ie^{-i\phi/2}}{2} \right), \]
\[ |c_2(t)\rangle = \left( \frac{1}{\sqrt{2}}, 0 \right), \]
\[ |c_3(t)\rangle = \left( \frac{1}{\sqrt{2}}, 1 \right). \]

This limit provides the initial conditions for the CIN pulse sequence, \(|\psi(0)\rangle = |00\rangle = |c_1(0)\rangle + |c_2(0)\rangle| / \sqrt{2}.

For the limit \(\Omega_p(t) / \Omega(t) \to \infty\) we obtain
\[ |c_1(t)\rangle = \left( \frac{1}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}} \right), \]
\[ |c_2(t)\rangle = \left( \frac{1}{\sqrt{2}}, 0, -\frac{1}{\sqrt{2}} \right), \]
\[ |c_3(t)\rangle = \left( \frac{1}{\sqrt{2}}, 0, -\frac{1}{\sqrt{2}} \right). \]

\[ |c_4(t)\rangle = \left( \frac{1}{\sqrt{2}}, 0, -\frac{1}{\sqrt{2}} \right), \] (10)

so that \(|\psi(0)\rangle = |00\rangle = |c_1(0)\rangle + |c_4(0)\rangle| / \sqrt{2} is the initial condition for the IN pulse sequence.

Therefore we observe that the system is initially in a superposition of the \(|c_1(t)\rangle\) and \(|c_2(t)\rangle\) dressed states for the CIN sequence and in a superposition of \(|c_3(t)\rangle\) and \(|c_4(t)\rangle\) dressed states for the IN sequence. According to the adiabatic theorem these are the only dressed states that are populated during all the time evolution if adiabatic conditions are satisfied. In the adiabatic representation spanned by the dressed states, the Schrödinger equation reads
\[ i\dot{\Phi}(t) = D(t)\Phi(t) + U_N(t)\Phi(t), \]

where \(D(t) = U(t)H(t)U^{-1}(t)\), \(U(t)\) is the transformation matrix that diagonalizes the Hamiltonian, and \(U_N(t) = -iU(t)U^{-1}(t)\) is the nonadiabatic coupling matrix. For the phase dependent four-level Hamiltonian, \(U_N(t)\) has the form
\[ U_N(t) = \frac{\hat{\Omega}(t)}{\hat{\Omega}^2(t)} \]
\[ = \frac{\Omega_p(t)}{\Omega^2(t)} \Omega(t) \sin \phi - \frac{\Omega_p(t)\Omega(t)\sin \phi}{\Omega^2(t)} 0 - \frac{\Omega_p(t)\Omega(t)\sin \phi}{\Omega^2(t)} 0 0 - \frac{\Omega_p(t)\Omega(t)\sin \phi}{\Omega^2(t)} \]

where \(\hat{\Omega}(t) = \Omega_p(t)\Omega(t) - \Omega_p(t)\Omega(t)\sin \phi/2\).

\[ \hat{\Omega}(t) = \Omega_p(t)\Omega(t) - \Omega_p(t)\Omega(t)\sin \phi/2 \]

In general, the nonadiabatic terms induce transitions among the dressed states that can affect the dynamic evolution. According to Eq. (12) there are nonzero nonadiabatic couplings between the set \(\{ |c_1(t)\rangle, |c_2(t)\rangle \}\) (which transports the population in the CIN case), and the set \(\{ |c_3(t)\rangle, |c_4(t)\rangle \}\) (which transports the population in the IN case). This implies that the dressed states that carry the population are coupled to the remaining states. The nonadiabatic terms depend on \(\hat{\Omega}^{-2}(t)\). Therefore they become smaller for increasing Rabi frequencies (pulse areas). The adiabatic requirements also depend on the phase \(\phi\), and they become stronger as \(\phi\) departs from zero.

The strength of the nonadiabatic couplings must be compared with the energy difference between the eigenvalues. In Fig. 2 we show the time evolution of the eigenvalues for different phases. We observe that the energy difference between \(\lambda_1(t)\) and \(\lambda_3(t)\) (or \(\lambda_2(t)\) and \(\lambda_4(t)\)) is maximal at \(\phi = 0\), Fig. 2(a), and becomes smaller as \(\phi \to \pm \pi\), Figs. 2(b) and 2(c). Since the strength of the nonadiabatic couplings increases when the distance between eigenstates decreases,
the separation between the dressed states can be used solely as a measure of adiabaticity.

Except for $\phi \sim \pm \pi$, STIRAP-type adiabatic conditions $[\int_0^t dt \lambda_\pm(t) > 1]$ apply to the closed-loop or double-$\Lambda$ system, where the threshold of adiabaticity will increase for increasing phase. At $\phi = \pm \pi$, Fig. 2(d), the difference between the pairs of dressed states is zero and the nonadiabatic coupling is infinite. This implies that the probability of diabatic crossing between the dressed states is one and, in fact, the system follows the two pathways shown by dark lines in Fig. 2(d). For this particular case the true dressed states can be obtained by directly diagonalizing the Hamiltonian with $\phi = \pm \pi$. It turns out that the dressed states are time independent:

\[
|c_1\rangle = (1/\sqrt{2},0,1/2,1/2), \\
|c_2\rangle = (1/\sqrt{2},0,1/2,1/2), \\
|c_3\rangle = (0,1/\sqrt{2},1/2,−1/2), \\
|c_4\rangle = (0,1/\sqrt{2},1/2,−1/2),
\]

with $\lambda_{1,2} = \mp \Omega_p(t)/\sqrt{2}$, $\lambda_{3,4} = \pm \Omega_\perp(t)/\sqrt{2}$. The dynamics proceeds, both in the CIN and IN sequences, through the superposition of $|c_1\rangle$ and $|c_2\rangle$, with zero nonadiabatic couplings. The dynamics implies Rabi oscillations between $|00\rangle$ and $|\Psi^+\rangle$, while the $|11\rangle$ state is dark.

In summary, we conclude that the system approximately follows two dressed states, $|c_1(t)\rangle$ and $|c_2(t)\rangle$ in the CIN pulse sequence, and $|c_3(t)\rangle$ and $|c_4(t)\rangle$ in the IN sequence (see Fig. 2). The nonadiabatic coupling between both sets of states is very small for reasonably large Rabi frequencies, except for the relative phase near $\pm \pi$, when the adiabatic requirements are very demanding.

IV. PREPARATION OF ENTANGLEMENT

In the CIN sequence, the initial wave function in the adiabatic representation is $[|c_1(0)\rangle+|c_2(0)\rangle]/\sqrt{2}$. If the nonadiabatic couplings can be neglected (that is, in the adiabatic limit) the wave function will evolve as

\[
|\psi(t)\rangle \approx |c_1(t)\rangle + \exp \left[-i \int_0^t dt' [\lambda_2(t') - \lambda_1(t')] \right] |c_2(t)\rangle,
\]

leading to interference between the paths at the end of the process. The interference depends on the dynamical phase, which is the effective pulse area $[\lambda_- = \lambda_2(t) - \lambda_1(t)]$.

\[
S_- = -\int_0^\infty dt \lambda_-(t).
\]

The outcome of the dynamics therefore depends on the characteristic of the dressed states at the end of the process $[|c_1(\infty)\rangle$ and $|c_2(\infty)\rangle$], which correspond to the limit $\Omega_p(t)/\Omega_\perp(t) \rightarrow \infty$ and on the relative phase and peak Rabi frequency that control the interference term. Inserting Eqs. (10) into Eq. (14) we obtain

\[
|\Psi(\infty)\rangle = \cos S_-/2 |4\rangle - e^{-i\phi_2}/\sqrt{2} \sin S_-/2 |2\rangle |3\rangle = \cos S_-/2 |11\rangle \\
- e^{-i\phi_2} \sin S_-/2 |\Psi^-\rangle.
\]

At $\phi = 0$, $S_- = 0$ and $|\Psi(\infty)\rangle = |11\rangle$. This corresponds to the STIRAP behavior. For any other relative phase, the dynamics, even in the adiabatic limit, is no longer an adiabatic passage: it oscillates as a function of the effective pulse area $S_-$ between the final state $|11\rangle$ ($|4\rangle$) and the entangled state $|\Psi^-\rangle$. Finally, at $\phi = \pm \pi$, Eq. (16) is no longer valid, since the true dressed states are given by Eq. (13). In this case the outcome of the dynamics implies Rabi oscillations between $|00\rangle$ and $|\Psi^+\rangle$ as a function of pulse area. The fidelity of the entangled state preparation depends quadratically on the Rabi frequency. However, since $S_-$ is still close to zero for small $\phi$, we will show that the present scheme is still more stable than the proposed entangled state preparation by Rabi pulses [6].

For the IN pulse sequence, at initial times $|\psi(0)\rangle = [(|c_3(0)\rangle+|c_4(0)\rangle)/\sqrt{2}]$ in the adiabatic representation. At the end of the process, assuming adiabatic evolution, the wave function will exhibit interference properties $[\lambda_- = \lambda_4(t) - \lambda_3(t)]$.

\[
|\psi(\infty)\rangle \approx |c_3(\infty)\rangle + \exp \left[-i \int_0^\infty dt \lambda_3(t) \right] |c_4(\infty)\rangle,
\]

that depend on the effective pulse area $S_+ = \int_0^\infty dt \lambda_3(t)$. Using Eq. (9) in Eq. (17) we obtain the form of the wave function in the original basis (except for $\phi = \pm \pi$).

\[
|\Psi(\infty)\rangle = \cos S_+/2 |4\rangle - i \sqrt{2} \sin S_+/2 |2\rangle + e^{-i\phi}|3\rangle = \cos S_+/2 |11\rangle \\
- i \sin S_+/2 |\Psi^-\rangle.
\]

Again, the dynamics in the adiabatic limit implies at final
time oscillations between the final state $|11\rangle$ (dashed line) and an entangled state or coherent superposition. In this case the relative phase of the superposition is directly controlled by the relative phase between the pulses. The frequency of oscillation depends on the pulse area and the relative phase of the pulses. Figure 3 shows several examples of population dynamics and final state populations as a function of the relative phase and area of the pump pulse. In Figs. 3(a) and 3(e) we show the population dynamics for the CIN and IN pulse sequences. The numerical results are obtained solving the Schrödinger equation using Gaussian shaped pulses $[Figs. 3(b) and 3(f)]$ with equal pulse areas $S_p=S_s[S_p,t_s \equiv \int_0^t d\Omega_p(t)]$ and time delays equal to the pulse width (time is scaled to the time width).

In the lowest four frames of Fig. 3 we show the dependence of the population at final time as a function of the pulse area (with fixed relative phase) and as a function of relative phase (with fixed pulse area). There is a clear difference in the sensitivity of the final state population for the CIN and IN pulse sequences. In the counterintuitive case the population is more stable as a function of the pulse area $[Fig. 3(c)]$ and more sensitive to the relative phase $[Fig. 3(d)]$. In contrast, the population is less sensitive to the phase while it oscillates rapidly with respect to the pulse area for the IN pulse sequence $[Figs. 3(g) and 3(h)]$. This qualitative difference is clearly revealed on the contour plots of the final population as a function of both pulse area and phase, see Fig. 4.

The complementary behavior of the final state sensitivities in the CIN and IN sequences can be easily explained by analyzing the functional dependence of the dynamical phases, $S_\pm$, on the corresponding parameters. $S_-$ and $S_+$ are...
defined by the different eigenvalues, $\lambda_1(t)$ and $\lambda_2(t)$ [Eq. (4)]. At $\phi=0$ we have $\lambda_1(t) = 0$, so that the dynamical phase is zero. As a result, the final state population is not sensitive to the pulse area for the CIN sequence. This is the well-known case of adiabatic passage following STIRAP. In contrast, $|\lambda_2(t)|$ is maximal at $\phi=0$. This gives maximal dynamical phase and fast oscillations of the final state, which is the expected outcome of a $\pi$ pulse dynamics. When the relative phase $\phi$ increases, $\lambda_1(t)$ grows almost linearly while $\lambda_2(t)$ is almost constant [6]. Thus the final state preparation is more sensitive to phase in the CIN dynamics and most sensitive to pulse area in the IN dynamics. In the worse case (for $\phi = \pm \pi$) the sensitivity is like the $\pi$ pulse dynamics. In the best case (for $\phi=0$) the sensitivity is like STIRAP. The fidelity of the entanglement preparation depends quadratically on the Rabi frequency, but the coefficient of this dependence rises from zero at $\phi=0$ to the highest value at $\phi = \pm \pi$. A more quantitative discussion is shown in [6].

V. CONCLUSION

We have presented a phase dependent scheme of population transfer in a four-level closed-loop system. Adding the knob of the relative phase of the pulses on the control scheme, the final state preparation depends on the square of the pulse area, that is, the properties of adiabatic passage are lost. However, both counterintuitive and intuitive sequences can be used to prepare specific superposition states. The most sensitive parameter of excitation varies depending on the chosen sequence. In particular, for a system of two interacting qubits, a simple STIRAP type sequence provides a very interesting scheme to prepare entangled states, since the excitation is more robust to the pulse area than any $\pi$ pulse sequence or than the intuitive time-delayed pulse sequence.

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APPENDIX

The general time-dependent Schrödinger equation for the 4-level system in closed-loop configuration for the case of two-photon resonance (see Fig. 1) has the form

\[
\begin{pmatrix}
\dot{a}_1(t) \\
\dot{a}_2(t) \\
\dot{b}_1(t) \\
\dot{b}_2(t)
\end{pmatrix} = -\frac{1}{2} \begin{pmatrix}
0 & 0 & \Omega_{p1}(t)e^{i\phi_1} & \Omega_{p2}(t)e^{i\phi_2} \\
0 & 0 & \Omega_{s1}(t)e^{-i\phi_1} & \Omega_{s2}(t)e^{-i\phi_2} \\
\Omega_{p1}(t)e^{-i\phi_2} & \Omega_{p2}(t)e^{-i\phi_1} & 2\Delta_1 & 0 \\
\Omega_{s1}(t)e^{i\phi_2} & \Omega_{s2}(t)e^{i\phi_1} & 0 & 2\Delta_2
\end{pmatrix} \begin{pmatrix}
a_1(t) \\
a_2(t) \\
b_1(t) \\
b_2(t)
\end{pmatrix},
\]

where $\Omega_{ij}$ are the Rabi frequencies, $\phi_i$ are the initial phases of the laser fields, $\Delta_1 = \omega_{01,00} - \omega_{p1} = \omega_{2} - \omega_{p2}$, $\Delta_2 = \omega_{10,00} - \omega_{s2} = \omega_{11,10} - \omega_{s1} = (E_{ij} - E_{ik})/\hbar$, and $E_{ij}$ is the energy of the $|ij\rangle$ state including interaction shifts (see, for example, Ref. [8]).

By choosing the phases, $a_1 \rightarrow a_1 \cdot e^{i\phi_1}$ and $a_2 \rightarrow a_2 \cdot e^{i\phi_2}$, we obtain

\[
\begin{pmatrix}
\dot{a}_1(t) \\
\dot{a}_2(t) \\
\dot{b}_1(t) \\
\dot{b}_2(t)
\end{pmatrix} = -\frac{1}{2} \begin{pmatrix}
0 & 0 & \Omega_{p1}(t) & \Omega_{p2}(t)e^{i\phi_2} \\
0 & 0 & \Omega_{s1}(t) & \Omega_{s2}(t)e^{i\phi_2} \\
\Omega_{p1}(t)e^{-i\phi_2} & \Omega_{p2}(t)e^{-i\phi_2} & 2\Delta_1 & 0 \\
\Omega_{s1}(t)e^{-i\phi_2} & \Omega_{s2}(t)e^{-i\phi_2} & 0 & 2\Delta_2
\end{pmatrix} \begin{pmatrix}
a_1(t) \\
a_2(t) \\
b_1(t) \\
b_2(t)
\end{pmatrix}.
\]

So, in fact there are only two essential phases: $\phi_2 - \phi_1$ and $\phi_1 - \phi_3$. If $\phi_2 - \phi_1 = \phi_1 - \phi_3$ there is no phase dependence at all in the dynamics. This is the situation most typically considered in the literature. If $\phi_2 - \phi_1 = \phi_1 - \phi_3$ we can redefine $b_2 \rightarrow b_2 e^{i(\phi_2 - \phi_3)}$ so that we obtain Eq. (2), where only one phase, the overall phase difference between both 2-photon paths, $\phi = \phi_1 - \phi_2 + \phi_4 - \phi_5$, is important.