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Robust creation and phase-sensitive probing of superposition states via stimulated Raman adiabatic passage (STIRAP) with degenerate dark states

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Abstract

We describe a method for creating an arbitrary coherent superposition of two atomic states in a controlled and robust way by using a sequence of three pulses in a four-state system. The proposed technique is based on the existence of two degenerate dark states (i.e. states having no component of the excited state) and their interaction. The mixing of the dark states can be controlled by changing the relative delay of the pulses, and thus an arbitrary superposition state can be generated. It is shown that the method is robust against small variations of parameters (e.g. the area of the pulses) and is insensitive to radiative decay from the intermediate excited state. A time reversed version of the technique makes possible the determination of phase occurring in a superposition of two atomic states.

1. Introduction

An interesting and powerful application of coherent population trapping [1] in three-state atoms is the transfer of population with a counter-intuitive sequence of pulses [2]. This technique, which was experimentally rediscovered in Ref. [3] and named stimulated Raman adiabatic passage (STIRAP) allows, in principle, a complete population transfer from a single initial to a single final quantum state [4]. The underlying physical mechanism is the existence of an adiabatically decoupled (or dark) state, which at early and late times coincides with the initial and target quantum state respectively. When the pump and Stokes frequencies together maintain two-photon resonance, then the only conditions to be fulfilled for successful transfer are those of adiabaticity, i.e. large pulse area, and counterintuitive application of the pulses, i.e. Stokes before pump. The technique is robust: moderate variations of the form, the area or the delay between the pulses do not affect the process (in contrast to other techniques for population transfer noted below).

The present paper discusses an extension of STIRAP which will create or probe, in a robust way, a superposition between an initial state ψ_1 and a target state ψ_3. Specifically, our goal is to devise a pulse sequence which will, starting from the initial condition $\Psi(t) \to \psi_1$, produce a superposition parameterized by the mixing angle α and the relative phase γ,

$$\Psi(t) \to \cos\alpha\, \psi_1 + \sin\alpha\, e^{i\gamma}\, \psi_3. \tag{1}$$

Likewise we are interested in the reverse process, which undoes the superposition and should allow one to probe the

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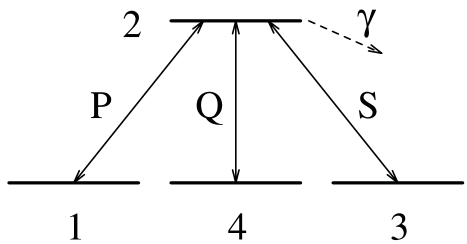

Fig. 1. Four-state system with tripod linkage produced by pulsed pump $P(t)$, Stokes $S(t)$, and control $Q(t)$ Rabi-frequencies. Dashed line indicates population loss.

coherence, in particular its phase. We anticipate interesting applications to quantum computing [5], preparation of cavity Fock states [6], atom optics and interferometry [7–9] and coherent control of chemical reactions [10].

As is well known, one can create an arbitrary superposition of two states, coupled by pulsed radiation, by adjusting the pulse area. A $\pi/2$ pulse, for example, creates equal amplitudes of the two states. However, this technique is not robust, because it requires very careful control of the duration of the interaction. In an atomic beam, only one velocity component will satisfy the necessary conditions. Spatial variation of the laser beam profile therefore poses problems. Furthermore, because the two states are connected directly via radiative transition (typically an electric-dipole transition involving opposite-parity states), spontaneous emission can take place from the excited state, thereby draining population away and altering the superposition.

Alternatively, as suggested in Refs. [7,8,11], one can modify the ordinary three-state STIRAP technique to create a definite superposition from a single quantum state: by maintaining a fixed ratio of Stokes and pump pulse amplitudes (rather than allowing the Stokes interaction to cease before that of the pump pulse) the state vector asymptotically coincides with a coherent mixture of initial and target states. Because the two states involved in the superposition can each be long lived (unlike the case of direct transition the states can have the same parity) there need be no change in the superposition caused by spontaneous emission. This method has drawbacks similar to the direct coupling, however, because the creation of a specific quantum superposition requires an accurate control of the relative strength of Stokes and pump pulses.

Here we proceed in a different way. We augment the usual three-state STIRAP scheme (wherein a pump pulse $P(t)$ couples states ψ_1 and ψ_2 and an earlier pulse $S(t)$ couples states ψ_2 and ψ_3) with a coherent coupling of the intermediate state (state ψ_2) to a fourth state (ψ_4, typically a metastable state) by means of a ''control'' pulse $Q(t)$. This interaction may be a static field or a microwave field, as well as an optical field. Fig. 1 sketches the various links. Under appropriate conditions (resonant tuning of carrier frequencies) there are, in this tripod-linked system, two degenerate null-eigenvalue adiabatic states, $\Phi_1(t)$ and $\Phi_2(t)$. Because these states have no component of the excited intermediate state ψ_2, they are immune to the spontaneous emission which characterizes this state; they are ''dark'' states (or ''trapped'' states). The pulse sequence can be set up in such a way that for $t \to \pm\infty$ one or both of the dark states asymptotically coincide with the bare atomic states ψ_1 and ψ_3. Due to the degeneracy of the dark states, the weak non-adiabatic coupling between them is important. It leads to a superposition, which asymptotically (after the pulses) turns into a coherent superposition of bare atomic states ψ_1 and ψ_3. We will show that the final admixture can be controlled by adjusting the timing of the pulses (the relative delay of the control pulse). The relative phase of the two components of the superposition is fixed by the relative phase of Stokes and pump pulses.

An inverted sequence of pulses leads to a complete return of the population into ψ_1, if the relative phase of Stokes and pump coincide with the phase of the superposition. In the case of a phase mismatch, some population will be transferred to state ψ_4. The amount of this population is a direct and sensitive measure of the phase mismatch. Thus the reverse process can be used to analyse the phase of an initial superposition state.

2. Degenerate dark states in four-level systems

We consider the four-state system shown in Fig. 1. Three resonant pulses with arbitrary but constant relative phase couple the lower states 1, 3 and 4 to an excited state 2. The atoms are assumed to be initially in state 1. The time-dependent Schrödinger equation for this system reads (in the rotating-wave approximation)

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{C}(t) = -i\,\mathbf{W}(t)\,\boldsymbol{C}(t), \tag{2}$$

where $\boldsymbol{C}(t)$ is a column vector, whose components are probability amplitudes $C_n(t)$, and the evolution matrix $\mathbf{W}(t)$ has the form

$$\mathbf{W}(t)=\frac{1}{2}\begin{bmatrix}0 & P(t) & 0 & 0\\ P(t) & 0 & S(t) & Q(t)\\ 0 & S(t) & 0 & 0\\ 0 & Q(t) & 0 & 0\end{bmatrix}. \tag{3}$$

Here $P(t)$, $S(t)$ and $Q(t)$ are the (time-dependent) Rabi-frequencies of the pump, Stokes and control pulses respectively. We assumed $P(t)$, $S(t)$ and $Q(t)$ to be real without loss of generality, since any *constant* phase factors $e^{i\phi_P}$, $e^{i\phi_S}$, $e^{i\phi_Q}$ can be absorbed in the definitions of the atomic basis states

$$\tilde{\psi}_1=\psi_1, \tag{4}$$

$$\tilde{\psi}_2=\mathrm{e}^{-i\phi_P}\psi_2, \tag{5}$$

$$\tilde{\psi}_3=\mathrm{e}^{i(\phi_S-\phi_P)}\psi_3, \tag{6}$$

$$\tilde{\psi}_4=\mathrm{e}^{i\phi_Q}\psi_4. \tag{7}$$

As an immediate consequence of this possibility, we see that the relative phase γ in the final admixture of states ψ_1 and ψ_3, Eq. (1), can be predetermined by adjusting the relative phase $\Delta\phi\equiv\phi_S-\phi_P$ between Stokes and pump pulses to have the value γ. Thus we will focus the following discussion on the question of how to control the mixing angle α.

The analysis of the system behavior is simplified by introducing a set of adiabatic (or dressed) states $\Phi_k(t)$, defined as instantaneous eigenstates of $\mathbf{W}(t)$. Two of these states are degenerate, with null eigenvalues, and have no component of atomic state ψ_2 (they are therefore termed ''trapped'' states, or ''dark'' states) :

$$\Phi_1(t)=\begin{bmatrix}\cos\vartheta(t)\\ 0\\ -\sin\vartheta(t)\\ 0\end{bmatrix},\quad \Phi_2(t)=\begin{bmatrix}\sin\varphi(t)\sin\vartheta(t)\\ 0\\ \sin\varphi(t)\cos\vartheta(t)\\ -\cos\varphi(t)\end{bmatrix}. \tag{8}$$

The remaining eigenvectors are

$$\Phi_3(t)=\frac{1}{\sqrt{2}}\begin{bmatrix}\cos\varphi(t)\sin\vartheta(t)\\ 1\\ \cos\varphi(t)\cos\vartheta(t)\\ \sin\varphi(t)\end{bmatrix},\quad \Phi_4(t)=\frac{1}{\sqrt{2}}\begin{bmatrix}\cos\varphi(t)\sin\vartheta(t)\\ -1\\ \cos\varphi(t)\cos\vartheta(t)\\ \sin\varphi(t)\end{bmatrix}. \tag{9}$$

The dynamical angles needed here are given by

$$\tan\vartheta(t)=\frac{P(t)}{S(t)},\quad \tan\varphi(t)=\frac{Q(t)}{\sqrt{P(t)^2+S(t)^2}}. \tag{10}$$

When the Q pulse is absent we have the usual three-state atomic system and the adiabatic states turn into the well-known adiabatic states for STIRAP [3,4]. However, the situation here is more complicated than the three-state STIRAP.

We use these dressed states to describe the state vector $\Psi(t)$ by writing

$$\Psi(t)=\sum_n B_n(t)\Phi_n(t). \tag{11}$$

The unitary transformation between the original (bare atomic) states and this alternative basis (adiabatic states) is through the matrix

$$\mathbf{U}(t)=\begin{bmatrix} \cos\vartheta(t) & \sin\varphi(t)\sin\vartheta(t) & \frac{1}{\sqrt{2}}\cos\varphi(t)\sin\vartheta(t) & \frac{1}{\sqrt{2}}\cos\varphi(t)\sin\vartheta(t) \\ 0 & 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \\ -\sin\vartheta(t) & \sin\varphi(t)\cos\vartheta(t) & \frac{1}{\sqrt{2}}\cos\varphi(t)\cos\vartheta(t) & \frac{1}{\sqrt{2}}\cos\varphi(t)\cos\vartheta(t) \\ 0 & -\cos\varphi(t) & \frac{1}{\sqrt{2}}\sin\varphi(t) & \frac{1}{\sqrt{2}}\sin\varphi(t) \end{bmatrix}. \tag{12}$$

Upon treating the expansion coefficients $B_n(t)$ as components of a vector $\boldsymbol{B}(t)$, through the transformation

$$\boldsymbol{C}(t)=\mathbf{U}(t)\boldsymbol{B}(t). \tag{13}$$

we obtain the Schrödinger equation as

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{B}(t)=-i\,\tilde{\mathbf{W}}(t)\boldsymbol{B}(t). \tag{14}$$

Because the transformation matrix $\mathbf{U}(t)$ is time-dependent, through the variation of both $\vartheta(t)$ and $\varphi(t)$, the transformation of the Hamiltonian has the form

$$\mathbf{W}(t)\rightarrow\tilde{\mathbf{W}}(t)=\mathbf{U}(t)^{-1}\mathbf{W}(t)\mathbf{U}(t)+i\,\dot{\mathbf{U}}(t)^{-1}\mathbf{U}(t), \tag{15}$$

The result reads

$$\tilde{\mathbf{W}}(t)=\begin{bmatrix} 0 & -i\dot{\vartheta}\sin\varphi(t) & -\frac{i}{\sqrt{2}}\dot{\vartheta}\cos\varphi(t) & -\frac{i}{\sqrt{2}}\dot{\vartheta}\cos\varphi(t) \\ i\dot{\vartheta}\sin\varphi(t) & 0 & \frac{i}{\sqrt{2}}\dot{\varphi} & \frac{i}{\sqrt{2}}\dot{\varphi} \\ \frac{i}{\sqrt{2}}\dot{\vartheta}\cos\varphi(t) & -\frac{i}{\sqrt{2}}\dot{\varphi} & \frac{1}{2}\Omega(t) & 0 \\ \frac{i}{\sqrt{2}}\dot{\vartheta}\cos\varphi(t) & -\frac{i}{\sqrt{2}}\dot{\varphi} & 0 & -\frac{1}{2}\Omega(t) \end{bmatrix}, \tag{16}$$

where

$$\Omega(t)=\sqrt{P(t)^2+S(t)^2+Q(t)^2} \tag{17}$$

is the total mean-square Rabi-frequency. Fig. 2 diagrams the relative positions of these eigenstates and shows the couplings between them.

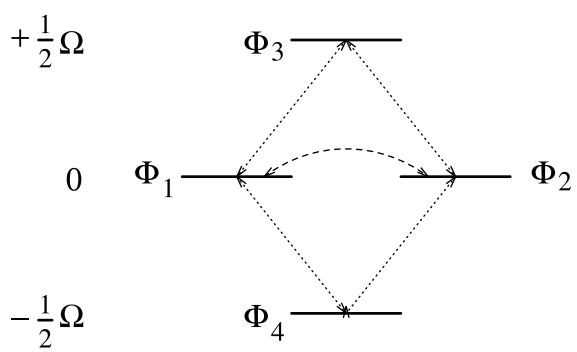

Fig. 2. Adiabatic states, ordered by eigenvalues. Dotted lines show nonadiabatic coupling between nondegenerate states. Dashed line shows coupling between degenerate (dark) states.

We note that our analysis below will also apply when there is one-photon detuning between state 2 and the states 1, 3 and 4, thereby placing a nonzero value into diagonal element $\mathbf{W}_{22}$ of Eq. (3). In this case the two dark states Φ_1 and Φ_2 are as shown in Eq. (8), although the remaining pair of eigenvectors differ from the construction of Eq. (9). This change has no effect on our results, which rely on adiabatic evolution to maintain the state vector as a combination of dark states.

3. Creation of superpositions via nonadiabatic mixing of dark states

The desired pulse sequence to create an arbitrary superposition will be designed such that the two degenerate dark states correspond, for $t \to \pm\infty$, to bare atomic states.

$$\Phi_1(t) \to \tilde{\psi}_1 \quad \text{for} \quad t \to \pm\infty, \tag{18}$$

$$\Phi_2(t) \to \tilde{\psi}_3 \quad \text{for} \quad t \to +\infty. \tag{19}$$

Without the control pulse $Q(t)$ these asymptotic conditions can be fulfilled by a pulse sequence whereby the Stokes pulse is turned on before the pump pulse arrives and is turned off after the pump pulse ceases. (Note that this is not the ''counterintuitive'' pulse sequence of STIRAP, which would require the Stokes pulse to cease before the pump pulse ceases.) This pulse sequence is indicated in the upper frame of Fig. 3. In this case the system is initially in the dark state $\Phi_1(t)$. The effect of the Stokes pulse, which arrives first, is to establish the connection between the initial atomic state and a particular adiabatic state $\Phi_1(t)$. The subsequent arrival of the pump pulse changes the composition of this dark state, but does not (if the evolution is adiabatic) change the identification of the actual system state vector $\Psi(t)$ with $\Phi_1(t)$. When pump and Stokes pulses have equal magnitudes, the adiabatic state is a 50–50 superposition of two atomic states. Since S and P are switched off in the opposite order to the order of turning on, the adiabatic evolution produced by these pulses alone would be completely reversible: without the Q pulse all population would return to the initial state.

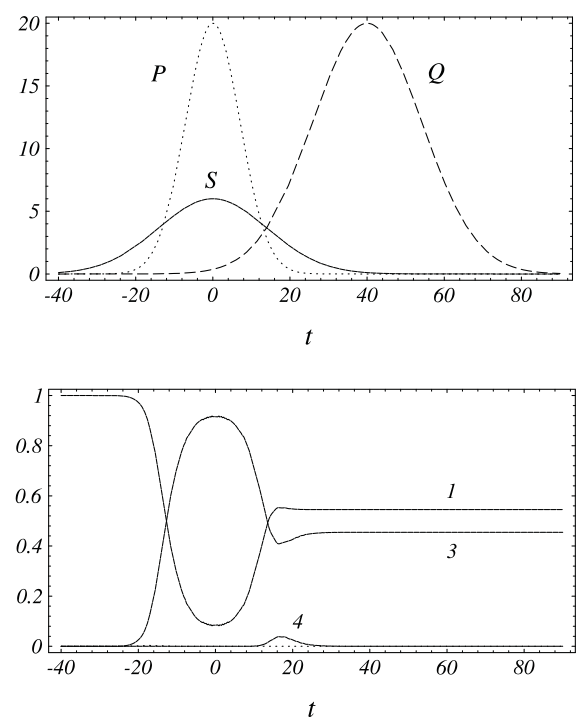

Fig. 3. Top: Example of pulses for coherence generation. Parameters: $A_P = A_Q = 20$, $A_S = 6$, $T_P = 10$, $T_S = T_Q = 20$, and $\tau_Q = 40$. Bottom: Time evolution of populations as produced by these pulses, obtained from numerical solution of the Schrödinger equation. Population of excited state 2 (dotted line) remains zero.

The time-reversal symmetry can be broken by the control pulse $Q(t)$, which introduces a coupling to a second dark state. As the following section shows (see Eq. (31)), this coupling leads to a mixture of dark states, described by a mixing angle Θ_∞, which asymptotically turns into a mixture of bare states (and this same mixing angle). Thus we have the sequence:

$$\begin{aligned} &\text{Initial stage:} \quad \tilde{\psi}_1 \to \Phi_1 \\ &\text{Interaction stage:} \quad \Phi_1 \to \cos\Theta_\infty\, \Phi_1 + \sin\Theta_\infty\, \Phi_2 \\ &\text{Final stage:} \quad \Phi_1 \to \tilde{\psi}_1, \quad \Phi_2 \to \tilde{\psi}_3. \end{aligned} \tag{20}$$

The result of this sequence is the state

$$\Psi(t) \to \cos\Theta_\infty\, \psi_1 + \sin\Theta_\infty\, \mathrm{e}^{i\Delta\phi}\psi_3. \tag{21}$$

To achieve the desired superposition of basis states it is only necessary that the pulse sequence produce an asymptotic mixing angle Θ_∞ equal to the angle α of Eq. (1),

$$\Theta_\infty = \alpha \tag{22}$$

and to fix the relative phase $\Delta\phi$ of pump and Stokes fields equal to the desired phase γ

$$\Delta\phi \equiv \phi_S - \phi_P = \gamma. \tag{23}$$

We illustrate the procedure, in Fig. 3, with an example of three Gaussian pulses,

$$P(t) = A_P\, f(t,T_P), \quad S(t) = A_S\, f(t,T_S), \quad Q(t) = A_Q\, f\big(t-\tau_Q,T_Q\big), \tag{24}$$

where $f(t,T) \equiv \exp[-(t/T)^2]$, and $T_S > T_P$. The upper frame of this figure shows the pulses and the lower frame shows the resulting populations. As can be seen the chosen pulse sequence accomplishes the desired task: The initial bare atomic state $\psi_1 = \tilde{\psi}_1$ is turned into a superposition $\cos\Theta_\infty\, \psi_1 + \sin\Theta_\infty\, \mathrm{e}^{i\Delta\phi}\psi_3$, where $\Delta\phi$ is the relative phase between Stokes and pump pulses, and the asymptotic mixing angle Θ_∞ is governed by the control pulse (see Eq. (31) below). Owing to the adiabaticity of the process the population of the excited state $\tilde{\psi}_2$ (dotted curve) remains always close to zero. Thus the coherence-generation is insensitive to spontaneous decay out of this state. We note, however, that a small transient population occurs in state $\tilde{\psi}_4$ (dash-dotted curve). Thus this state should be stable (or metastable).

By varying the delay τ_Q between the control pulse and the pump and Stokes pulses one produces a range of superpositions, as parameterized by the asymptotic mixing angle Θ_∞. Fig. 4 shows this mixing angle for the pulses of Fig. 3 as a function of the delay τ_Q. As can be seen, it is possible to choose the delay (in two ways) to produce any desired superposition.

The success of the method can be understood by considering the limiting case of adiabatic evolution. The adiabatic limit, defined by the conditions

$$\frac{\mathrm{d}\vartheta(t)}{\mathrm{d}t} \ll \Omega(t), \quad \frac{\mathrm{d}\varphi(t)}{\mathrm{d}t} \ll \Omega(t), \tag{25}$$

implies that the effective pulse area is very large

$$A \equiv \int_{-\infty}^{\infty} \mathrm{d}\tau\, \Omega(\tau) \gg 1. \tag{26}$$

For suitable systems this condition can be fulfilled even for pulses produced as a result of atoms moving across continuous-wave laser beams. For example, in atomic beam experiments A can reach values as large as 100 [12].

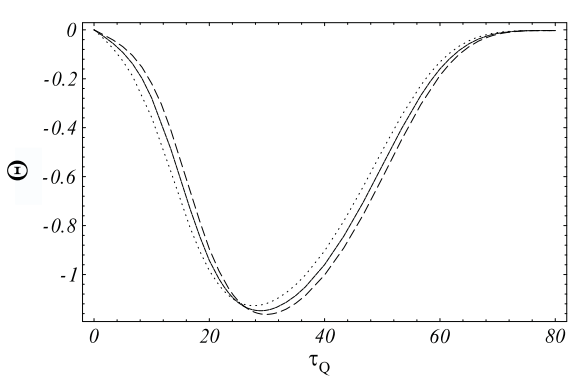

Fig. 4. Asymptotic mixing angle Θ_∞ as function of delay of control pulse for example of Fig. 3 (solid curve). The other curves show the sensitivity to variations of control-pulse amplitude: $A_Q / A_Q^{(0)} = 0.8$ (dotted curve) and $A_Q / A_Q^{(0)} = 1.2$ (dashed curve).

In this limiting situation the couplings of the dark states $\Phi_1(t)$ and $\Phi_2(t)$ to the bright states $\Phi_3(t)$ and $\Phi_4(t)$ can be disregarded. However, due to the degeneracy of $\Phi_1(t)$ and $\Phi_2(t)$, the coupling between these states cannot be neglected [13]. We shall assume that initially the system starts in a dark state and, by evolving adiabatically, remains in some superposition of the two degenerate dark states. Then we have $B_3(t) = B_4(t) = 0$ at all times. The amplitudes $B_1(t)$ and $B_2(t)$ are, however, affected by the pulses. To evaluate the behavior of these coefficients we consider the 2×2 submatrix of the Hamiltonian which connects these states. The equations of motion for the respective amplitudes are

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} B_1(t) \\ B_2(t) \end{bmatrix} = \begin{bmatrix} 0 & -\dot{\vartheta}\sin\varphi(t) \\ \dot{\vartheta}\sin\varphi(t) & 0 \end{bmatrix}\begin{bmatrix} B_1(t) \\ B_2(t) \end{bmatrix}. \tag{27}$$

The general solution to these equations, for arbitrary initial conditions, can be written as

$$B_1(t) = B_1(-\infty)\cos\Theta(t) - B_2(-\infty)\sin\Theta(t), \tag{28}$$

$$B_2(t) = B_2(-\infty)\cos\Theta(t) + B_1(-\infty)\sin\Theta(t), \tag{29}$$

where the phase $\Theta(t)$ is given by

$$\Theta(t) = \int_{-\infty}^{t} \mathrm{d}\tau\, \dot{\vartheta}(\tau)\sin\varphi(\tau). \tag{30}$$

The asymptotic superposition coefficients $B_n(\infty)$ are determined by the asymptotic mixing angle

$$\Theta_\infty \equiv \Theta(\infty) = \int_{-\infty}^{+\infty} \mathrm{d}\tau\, \dot{\vartheta}(\tau)\sin\varphi(\tau) = -\int_{-\infty}^{+\infty} \mathrm{d}\tau\, \vartheta(\tau)\,\frac{\mathrm{d}}{\mathrm{d}\tau}\sin\varphi(\tau). \tag{31}$$

Fig. 5 shows the difference of the bare-state populations obtained from the adiabatic approximation, Eqs. (28) and (29), and a numerical solution of the Schrödinger equation for the example discussed above. The agreement is excellent, with absolute errors less than 10^{-3}.

Eq. (31) allows a qualitative discussion of Fig. 4. As can be seen from Eq. (10), $\vartheta(t)$ is nonzero only when the pump pulse is on. On the other hand, $(\mathrm{d}/\mathrm{d}t)\sin\varphi(t)$ is, to a first approximation, proportional to the slope of the control pulse. Thus Θ_∞ is roughly proportional to this slope averaged over the duration of the pump pulse, a quantity that depends on the control-pulse delay τ_Q. The angle Θ_∞ vanishes for zero delay and increases monotonically with τ_Q until it reaches a maximum. Further increasing the delay reduces the average slope again and thus Θ_∞ decreases. Although quantitatively the mixing angle depends on the actual pulse shape, the qualitative behaviour is similar for a large class of pulse forms.

The proposed scheme is very robust. Since the excited state ψ_2 remains unpopulated (in lowest order of the weak non-adiabatic coupling), the effect of spontaneous emission can be neglected in the adiabatic limit. Satisfying the asymptotic conditions (18) and (19) is not critical, since they only require a certain pulse sequence. No careful control of relative intensities is required. Finally as shown by the dotted and dashed curves in Fig. 4, the asymptotic mixing angle Θ_∞ is relatively insensitive to variations of the control pulse amplitude (as long as the time evolution is adiabatic). Although not shown here, a similar conclusion holds for variations of the pump and Stokes amplitudes. For application to experiments involving atomic beams moving transversely across laser beams, it is important that the coherence generation be insensitive to the speed at which the atoms traverse the laser beams. If transverse Doppler-shifts can be neglected – as is often the case in these kinds of experiments – the effect of the velocity distribution is just a change in the interaction time of the individual atoms with the laser beams. This amounts to a rescaling of the time unit. The asymptotic mixing angle is not affected by

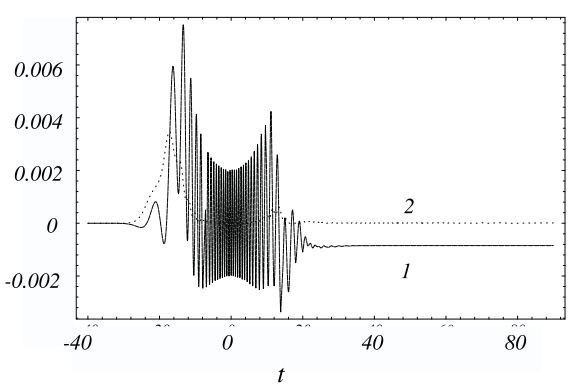

Fig. 5. Difference of adiabatic and exact results for populations of state 1 (solid line) and state 2 (dotted line) for the example of Fig. 3.

such a rescaling of time units (as long as the evolution is adiabatic), as can be seen from Eq. (31). Thus the suggested method is insensitive to the longitudinal velocity distribution of an atomic beam.

4. Reversal and phase-sensitive probing of superpositions

Let us now consider a pulse sequence which reverses a superposition and returns population into state ψ_1. More generally, let us start from an initial superposition of states 1 and 3,

$$\Psi(-\infty) = \cos\alpha\,\psi_1 + \sin\alpha\,\mathrm{e}^{i\gamma}\psi_3 = \cos\alpha\,\tilde{\psi}_1 + \sin\alpha\,\mathrm{e}^{i(\gamma-\Delta\phi)}\tilde{\psi}_3 \tag{32}$$

where α is the initial mixing angle and γ the initial relative phase of the states ψ_1 and ψ_3 (absolute phases are physically irrelevant). Obviously if we apply a pulse sequence which is time reversed to the one that created the superposition we will

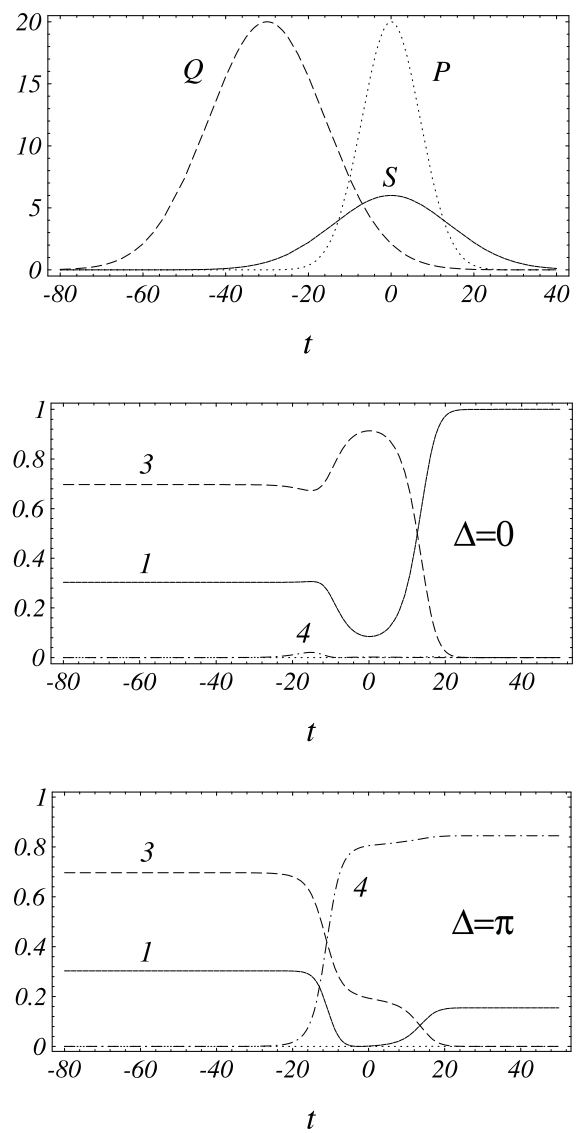

Fig. 6. Top: Example of pulses for coherence reversal. The initial mixing angle is $\alpha = -0.9877$. Parameters: $A_P = A_Q = 20$, $A_S = 6$, $T_P = 10$, $T_S = T_Q = 20$, and $\tau_Q = -30$. Middle and bottom: Time evolution of the populations, as produced by these pulses for $\Delta = 0$ and $\Delta = \pi$.

exactly undo the superposition and return all population into state ψ_1. However, the reversal is also possible under less stringent conditions. If we apply a pulse sequence where the Stokes pulse is again switched on before and switched off after the pump pulse, and if the control pulse precedes the other two, the whole population can be returned to state ψ_1 if two conditions are satisfied: Firstly, the relative phase $\Delta\phi$ between Stokes and pump has to match the phase γ of the initial coherent superposition. Secondly, the time delay between the control pulse $Q(t)$ and the other pulses must be chosen to exactly undo the mixing of the dark states.

To prove this claim we can apply the preceding analysis of the adiabatic evolution. We apply a reversed pulse-sequence, such as shown in the top frame of Fig. 6, to a four-level system whose initial state is described by Eq. (32), and we determine the populations as $t \to \infty$. In the dark-state basis (8), (9) the initial state has the following components,

$$B_1(-\infty) = \cos\alpha,$$

$$B_2(-\infty) = \mathrm{e}^{i\Delta}\sin\alpha,$$

$$B_3(-\infty) = B_4(-\infty) = 0,$$

where $\Delta = \gamma - \Delta\phi = \gamma - (\phi_S - \phi_P)$. After the interaction these amplitudes become

$$B_1(\infty) = \cos\alpha\cos\Theta_\infty - \mathrm{e}^{i\Delta}\sin\alpha\sin\Theta_\infty,$$

$$B_2(\infty) = \mathrm{e}^{i\Delta}\sin\alpha\cos\Theta_\infty + \cos\alpha\sin\Theta_\infty,$$

$$B_3(\infty) = B_4(\infty) = 0,$$

where Θ_∞ is the asymptotic mixing angle for the reverse pulse-sequence. From these expressions we find the final populations of the atomic bare states to be

$$P_1(\infty) = \left|\cos\alpha\cos\Theta_\infty - \sin\alpha\sin\Theta_\infty\,\mathrm{e}^{i\Delta}\right|^2, \tag{33}$$

$$P_2(\infty) = P_3(\infty) = 0, \tag{34}$$

$$P_4(\infty) = \left|\cos\alpha\sin\Theta_\infty + \sin\alpha\cos\Theta_\infty\,\mathrm{e}^{i\Delta}\right|^2. \tag{35}$$

If the phase difference between Stokes and pump matches that of the initial coherence, i.e. if $\Delta = 0$, then the final populations are

$$P_1(\infty) = \cos^2(\Theta_\infty + \alpha), \tag{36}$$

$$P_2(\infty) = P_3(\infty) = 0, \tag{37}$$

$$P_4(\infty) = \sin^2(\Theta_\infty + \alpha). \tag{38}$$

If the reversed-pulse angle Θ_∞ is the negative of the original angle α, then the population is found entirely in state ψ_1 at the end of the reversing sequence. The middle frame of Fig. 6. shows the evolution of the atomic bare-state populations for the

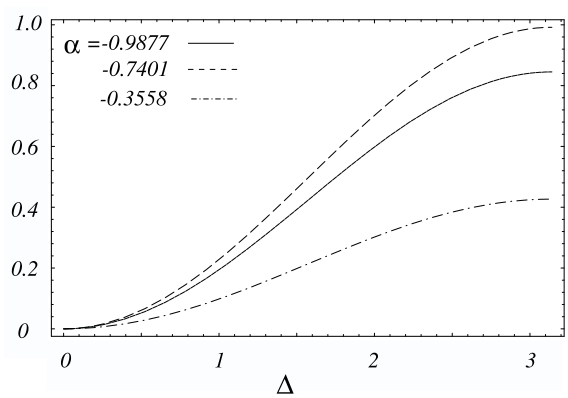

Fig. 7. Final population of state 4 as function of phase mismatch $\Delta \equiv \gamma - \Delta\phi$ for $\alpha = -0.9877$ (solid line), $\alpha = -0.7401$ (dashed line), and $\alpha = -0.3558$ (dash-dotted line). Time delay τ_Q is chosen such that $\Theta_\infty = -\alpha$. The results shown correspond to the numerical solution of the Schrödinger equation for Gaussian pulses of Fig. 6, which are however indistinguishable from the analytic adiabatic results according to Eq. (39).

sequence of Gaussian pulses shown in the upper frame for $\Delta = 0$. The time delay τ_Q is chosen such that $\Theta_\infty = -\alpha = -0.9877$. The entire population is driven back to state 1.

Eqs. (33) and (35) give an interesting possibility to detect the phase of a superposition state. If $\Theta_\infty = -\alpha$ but $\Delta \neq 0$, i.e. $\Delta\phi \neq \gamma$, some population is transferred to state ψ_4 as $t \to +\infty$. This is shown in the bottom frame of Fig. 6 for the out-of phase case $\Delta = \pi$. The final amount of population in state ψ_4 depends on the phase mismatch,

$$P_4(\infty) = \sin^2(2\alpha)\sin^2(\Delta/2). \tag{39}$$

Thus a measurement of the population in state ψ_4, and knowledgement of the phase difference between pump and Stokes pulses, gives the original phase $\gamma = \Delta + (\phi_S - \phi_P)$. Fig. 7 shows the population in state ψ_4 as a function of Δ for different values of α corresponding to the Gaussian pulse sequence of Fig. 6. The curves are obtained from a numerical evaluation of the Schrödinger equation. (The corresponding curves for the analytic results in the adiabatic limit are indistinguishable from these on the scale of the plot.) Thus one can determine the phase of the initial superposition state by varying the phase difference between Stokes and pump and measuring the population in state ψ_4.

5. Summary and conclusions

In the present paper we suggested a method to create a quantum superposition of metastable states out of a single initial state in a robust and controlled way. In order to achieve robustness against variations of pulse amplitudes we applied the technique of stimulated adiabatic Raman passage (STIRAP) known from three-level systems and we extended it by another coherent coupling by means of a control pulse. As a consequence of this additional coupling there are two degenerate adiabatic dark or trapped states. The underlying mechanism of our proposed technique is the weak non-adiabatic interaction between these, which remains important even in the adiabatic limit due to the degeneracy of the dark states. With an appropriately chosen sequence of pump and Stokes pulses, the two dark states can be made to coincide with bare atomic states for $t \to \pm\infty$. Thus the initial state vector of the atom is identical to one of the dark states. Without the presence of the control pulse the dark state would adiabatically rotate either back into the initial quantum state or into another single atomic state. The control pulse disturbs this evolution in a well-defined way so the outcome is a mixture of two bare atomic states. We have shown that the mixing angle Θ_∞ of this superposition can be adjusted by changing the delay of the control pulse with respect to the other pulses. The relative phase of the two amplitude components is determined by the difference phase between pump and Stokes pulses.

The nonadiabatic mixing of the degenerate dark states can also be used to reverse the superposition or to probe the superposition including its phase. A time reversed sequence of pulses with an appropriately chosen delay of pump and Stokes with respect to the preceding control pulse returns all population to state ψ_1, if the relative phase of Stokes and pump matches that of the superposition. If there is a difference in these phases, some population will be transferred into state 4, which gives a sensitive means to probe the phase of an initial superposition state. A combination of coherence creation and detection in an atomic beam set up with the proposed STIRAP technique opens new possibilities for Ramsey-type interferometry.

As opposed to other seemingly simpler methods, such as π-pulse techniques or ordinary three-level STIRAP, the proposed technique is robust against amplitude variations. In particular, no careful control of the pulse area or the relative intensities of the pulses is needed. As is typical for schemes based upon population trapping, our method places no population into the intermediate excited state and is therefore insensitive to possible radiative decay from that level. As with other Raman-interaction excitation, it establishes coherence on a dipole-forbidden transition between metastable states. We anticipate interesting applications in quantum computing, optical memory storage, interferometry, and coherent control of reaction dynamics.

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