Creation of arbitrary coherent superposition states by stimulated Raman adiabatic passage

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A technique for creation of well-defined preselected coherent superpositions of multiple quantum states is proposed. It is based on an extension of the technique of stimulated Raman adiabatic passage (STIRAP) to degenerate levels. As an example, the nine-state system composed of the magnetic sublevels of three levels with angular momenta \( J_g=0, J_e=1 \), and \( J_f=2 \) is studied in detail. Starting from the \( |J_g=0,M_g=0\rangle \) state, STIRAP can create an arbitrary preselected coherent superposition between the five \( M_f \) sublevels \( (M_f=-2,-1,0,+1,+2) \) of the \( J_f=2 \) level with 100% efficiency in the adiabatic limit. The populations and the phases of the \( M_f \) states in this superposition are determined entirely by the polarizations of the two laser fields. It is shown that this technique allows one to create any superposition of the five \( M_f \) states, that is, to reach any point in the Hilbert space of the \( J_f=2 \) manifold, and the corresponding recipes for choosing the laser polarizations are presented.

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I. INTRODUCTION

The technique of stimulated Raman adiabatic passage (STIRAP) has become a powerful and versatile tool for coherent and complete population transfer between two quantum states 1 and 3 via an intermediate state 2. STIRAP has several major advantages over techniques that use resonant pulses of precise areas (e.g., \( \pi \) pulses), which have stimulated its widespread applications. These range from population transfer between electronic states in atoms and rovibrational states in molecules to atom optics experiments, atom-photon interactions, and STIRAP to create well-defined preselected coherent superpositions of quantum states. As a case study we examine in detail the nine-state system composed of the magnetic sublevels of three levels with angular momenta \( J_g=0 \) (nondegenerate, initial/metastable or stable state), \( J_e=1 \) (degenerate, intermediate/decaying excited state), and \( J_f=2 \) (degenerate, final/metastable or stable state). Such a system can be found for instance in metastable neon atoms which have been used extensively in recent years in adiabatic population transfer and atom optics experiments [4–7]. This model system is supposed to be prepared initially in the \( |J_g=0,M_g=0\rangle \) state and the objective is to create, by a STIRAP-like process, an arbitrary preselected coherent superposition of the five \( M_f \) sublevels \( (M_f=-2,-1,0,+1,+2) \) of the \( J_f=2 \) level. Interestingly, this system possesses three dark states, two of which involve the five \( M_f \) sublevels only and are irrelevant for our purposes. The third dark state involves these five \( M_f \) sublevels and the single initial \( M_g=0 \) sublevel of the \( J_g=0 \) level and it is the population transfer vehicle in the present technique. We shall demonstrate that in the adiabatic limit the desired superposition is created with 100% efficiency and the populations and the phases of the \( M_f \) states in this superposition are determined entirely by the polarizations and phases

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of the two laser fields. Moreover, we shall show that this technique allows us to create any superposition of the five $M_f$ states, that is to reach any point in the Hilbert space of the $J_f=2$ manifold, and we shall prescribe the corresponding recipes for choosing the laser polarizations and phases.

The proposed technique follows and extends earlier proposals to use STIRAP for creation of superposition states. Marte et al. [8], Lawall and Prentiss [9], Weitz et al. [10], and Vitanov et al. [11] have proposed and implemented a variation of STIRAP, fractional STIRAP, in which the adiabatic evolution is interrupted, while both states 1 and 3 are still populated, which leaves the system in a superposition of states 1 and 3. This can be achieved either by truncating both the pump and Stokes pulses at the same instant of time [8–10] or by letting them vanish simultaneously maintaining a certain ratio [11]; the created superposition is determined by this ratio. In another extension of STIRAP, called tripod STIRAP [4,12], the three states in STIRAP are supplemented by an additional state 4, coupled to the intermediate state 2. By varying the amplitude and the timing of this additional coupling one can create various coherent superpositions of states 1, 3, and 4. An extension of tripod STIRAP, in which state 4 is replaced by $N$ states has been proposed by Kis and Stenholm [13]. Thanopoulos et al. [14] and Gong et al. [15] have studied other multistate extensions of STIRAP: a single state is coupled to a set of nondegenerate states (intermediate states) that are coupled further to a set of nearly degenerate [14] or degenerate states [15].

The proposed method for creation of coherent superpositions of magnetic sublevels can also be regarded as an extension of STIRAP to degenerate levels. The first proposal for the implementation of STIRAP in a system with level degeneracies both in the intermediate and final levels has been published by Karpati et al. [16]. The present work is a natural extension of our earlier study on degenerate levels [17] where the most general conditions for STIRAP between such levels have been derived. In the present paper, we use these earlier results and ideas more specifically, to the problem of creation of arbitrary coherent superposition states in systems of practical significance.

The present technique is an adiabatic alternative of the method proposed by Law and Eberly [18,19] for the creation of superpositions of magnetic sublevels of the same angular momentum state. The Law-Eberly proposal involves a suitable sequence of two-photon resonant Raman pulses of precise areas. As with any resonant $\pi$-pulse technique, small variations from the pulse areas or the carrier frequencies are detrimental and may lead to significant deviations from the target superposition state. In contrast, the present technique is adiabatic in nature and it is therefore robust against amplitude and frequency fluctuations of the external laser fields. In addition to sufficiently high laser intensity (needed to ensure adiabatic evolution) it only requires control of the polarizations and phases of the two laser fields (pump, linking levels $J_g=0$ and $J_f=1$, and Stokes, linking $J_g=1$ and $J_f=2$), which is significantly easier to achieve experimentally.

The paper is organized as follows. In Sec. II we introduce the model system and define the problem. In Sec. III we derive the dark states of the system and determine the transfer dark state, which is used for the creation of the superposition state. In Sec. IV we discuss the phase relations between the driving fields and the superposition phases, which reveal interesting subtleties. In Sec. V we provide a mathematical proof of the completeness of the proposed technique, i.e., that any state in the Hilbert space of the final degenerate level can be reached by a suitable choice of laser polarizations; moreover, we prescribe the procedure for finding the polarizations and give numerical illustrations in Sec. VI. In Sec. VII we describe possible experimental implementations. In Sec. VIII we provide a summary of the results.

II. THE MODEL SYSTEM

We consider the nine-state coupled system shown in Fig. 1. The initial state is an angular momentum state with $J_g=0$ (single state), which is coupled to the sublevels of the excited state with $J_f=1$. The couplings are provided by the pump pulse that consists of $\sigma^+$ and $\pi$ polarization components. All components share the same time dependence. The excited states are coupled further to the final set of angular momentum states with $J_f=2$, hence it consists of five sublevels. The couplings are provided by the Stokes pulse that consists of $\sigma^-$ and $\pi$ polarization components, similarly to the pump pulse. This linkage can be realized experimentally in neon atoms [4–7].

The system Hamiltonian in the rotating-wave picture, with the rotating-wave approximation, is defined as

\[
H(t) = \begin{bmatrix}
0 & p(t)P & 0 \\
p(t)P^\dagger & \Delta & s(t)S \\
0 & s(t)S^\dagger & 0
\end{bmatrix},
\]

where the pump coupling matrix is given by

\[
P = \begin{bmatrix}
\sqrt{\frac{3}{2}}P_+ \\
\sqrt{\frac{1}{2}}P_- \end{bmatrix},
\]

and the Stokes coupling matrix reads

\[
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\]
we have already described in the Introduction, here the initial counterintuitive pulse sequence of STIRAP yields complete mulated in Ref. the final state space consists of five degenerate sublevels.

The time dependences of the pump and Stokes coupling pulses are described by the envelope functions \( p(t) \) and \( s(t) \), respectively, with unit peak amplitudes. The constant elements of the pump coupling matrix \( P \) are parametrized as

\[
P_\pm = P \exp(i\alpha_p)\sin(\eta_p)\cos(\varphi_p),
\]

(4a)

\[
P_\pi = P \exp(i\alpha_\pi)\sin(\eta_\pi)\cos(\varphi_\pi),
\]

(4b)

\[
P_s = P \exp(i\alpha_s)\sin(\eta_s)\sin(\varphi_s),
\]

(4c)

where \( \alpha_p, \pi \) defines the phase, and \( \eta_p, \eta_\pi, \eta_s \) define the ellipticity of the pump field \( E_p \). These phases and angles are assumed constant throughout the interaction. The peak amplitude of the Rabi frequency \( \Omega_p = 2P/\hbar \) is characterized by the constant \( P \), which in turn is proportional to the reduced matrix element of the dipole operator \( \mu \) between the \( g \) and \( e \) states, \( P = |\mu|e|E_p| \). Similarly, the constant elements of the Stokes coupling matrix \( S \) are parametrized as

\[
S_- = S \exp(i\alpha_-)\sin(\eta_-)\cos(\varphi_-),
\]

(5a)

\[
S_\pi = S \exp(i\alpha_\pi)\sin(\eta_\pi)\cos(\varphi_\pi),
\]

(5b)

\[
S_+ = S \exp(i\alpha_+)\sin(\eta_+)\sin(\varphi_+).
\]

(5c)

The parameters of the Stokes couplings are defined similarly to those of the pump couplings (4).

In a previous paper [17] it has been shown that in a degenerate three-level system, for a nondescending (viewed from the initial level) sequence of level degeneracies, the counterintuitive pulse sequence of STIRAP yields complete population transfer from the initial level to the final level. As we have already described in the Introduction, here the initial level consists of a single level, the excited (intermediate) state space consists of three degenerate sublevels, whereas the final state space consists of five degenerate sublevels. Therefore, the condition of complete population transfer formulated in Ref. [17], \( N_g \equiv N_e \equiv N_f \), is clearly met.

We are going to consider two coupling configurations: with and without the \( \pi \) fields. When only the \( \sigma^+ \) components of the coupling fields are present, one has a six-level coupled subsystem. Obviously, in this case we can create only superpositions of the \( M_f = -2,0,2 \) sublevels of the \( J_f = 2 \) level. Some considerations on the realization of STIRAP in this linkage have been published in Ref. [16]. Here we present a much more thorough analysis of this subsystem, and compare the results to those obtained for the nine-level fully coupled system, when both the \( \sigma^+ \) and \( \pi \) components of the coupling fields are present. In the latter case, all five sublevels \( M_f = -2,-1,0,1,2 \) of the \( J_f = 2 \) level are involved in the final coherent superposition state.

III. DARK STATES AND DYNAMICS

In Ref. [17] a general method has been developed to find those dark states of a three-level degenerate system, that can be used for population transfer in a STIRAP-like process. Although this method applies to any system that satisfies some basic conditions [17], for large number of states and couplings the calculations become too cumbersome.

We follow here a different strategy. We first note that a dark state \( \Psi_D(t) \) is an eigenstate of the Hamiltonian with null eigenvalue; hence

\[
H(t)\Psi_D(t) = 0.
\]

According to the general theory [17], the degenerate three-level system has \( N_g + N_e - N_f \) dark states; hence there are two dark states in the \( \sigma^+\sigma^- \)-coupled six-state (1-2-3) system and three dark states in the fully, \( \sigma^+\pi\sigma^- \)-coupled nine-state (1-3-5) system. Again according to the general theory [17], by performing a Morris-Shore transformation on the Stokes transition, we conclude that in the six-state (1-2-3) system there is one dark state \( \Psi_D^{(1)} \) composed of \( J_f = 2 \) sublevels only, and that in the nine-state (1-3-5) system there are two such dark states, \( \Psi_D^{(1)} \) and \( \Psi_D^{(2)} \). These dark states are decoupled from the rest of the system because all Stokes couplings have the same time dependence [17], and remain unpopulated. These dark states are irrelevant for STIRAP because they do not contain a contribution from the initial state \( |J_g = 0, M_f = 0 \rangle \) of the \( J_f = 0 \) level and hence they cannot connect adiabatically the \( J_g = 0 \) and \( J_f = 2 \) levels.

There is, however, one additional dark state in either cases, 1-2-3 and 1-3-5, which is composed of the sublevels of the \( J_f = 2 \) level and the initial state \( |0,0 \rangle \) of the \( J_g = 0 \) level. It can be written as

\[
\Psi_D(t) = \frac{1}{N_D(t)}[d_{0,0}(t),0,0,0,
\]

\[
d_{2,-2}(t),d_{2,-1}(t),d_{2,0}(t),d_{2,1}(t),d_{2,2}(t)]^T,
\]

(7)

where \( N_D(t) \) is a normalization factor. We shall therefore refer to this state as a transfer dark state because STIRAP proceeds through it.

It is very important, and fortunate, that the transfer dark state \( \Psi_D(t) \) is decoupled from the other dark states because the latter are constant,

\[
\langle \Psi_D^{(1)} | \Psi_D(t) \rangle = - \langle \Psi_D^{(2)} | \Psi_D(t) \rangle = 0,
\]

(8a)

\[
\langle \Psi_D^{(2)} | \Psi_D(t) \rangle = - \langle \Psi_D^{(1)} | \Psi_D(t) \rangle = 0,
\]

(8b)

provided \( \Psi_D(t) \) is orthogonal to \( \Psi_D^{(1)} \) and \( \Psi_D^{(2)} \),

\[
\langle \Psi_D^{(1)} | \Psi_D(t) \rangle = \langle \Psi_D^{(2)} | \Psi_D(t) \rangle = 0.
\]

(9)

If this was not the case, we would have had to face the formidable challenge of accounting for the resonant transitions between all three dark states induced by the nonadiabatic couplings.

In conclusion of this discussion, we have found that there is a time-dependent dark state \( \Psi_D(t) \), which, as we shall show below, can be used for STIRAP-like population transmion from \( J_g = 0 \) to \( J_f = 2 \). There are one (for the 1-2-3 system) or two (for the 1-3-5 system) other dark states, which are stationary. If the transfer dark state \( \Psi_D(t) \) is orthogonal to these states, it is decoupled from them and they can therefore
be discarded in the adiabatic limit, to which we adhere. However, these dark states are still significant as they define unambiguously the transfer dark state $\Psi_D(t)$ by means of the orthogonality condition.

We shall consider below in detail the two systems: the $\sigma^+\sigma^-$-coupled six-state (1-2-3) system and the full, $\sigma^+\pi\sigma^-$-coupled nine-state (1-3-5) system, and we shall present the explicit form of the corresponding transfer dark state.

A. The six-state subsystem

The $\sigma^+\sigma^-$-coupled six-state (1-2-3) system has two dark states [17]. The decoupled, stationary dark state, which contains only $J_z=2$ sublevels, reads

$$\Psi_D^{(1)} = \frac{1}{N}[0,0,0,0,0,0,-\sqrt{6}S_-S_+,0,0]^{T}. \quad (10)$$

The transfer dark state

$$\Psi_D = \frac{1}{N}[d_{0,0},d_{2,-2},0,d_{2,0},d_{2,2}]^{T} \quad (11)$$
can be derived by finding another linearly independent null-eigenvalue eigenstate of the Hamiltonian and then use Gramm-Schmidt orthogonalization. The nonzero components of $\Psi_D(t)$ are

$$d_{0,0}(t) = \frac{s(t)}{N(t)}(S_+^4 + S_-^4 + 6|S_-|^2|S_+|^2), \quad (12a)$$

$$d_{2,-2}(t) = \frac{p(t)\sqrt{2}}{N(t)}S_+^2(P_+^*S_-S_+ - P_-^*6|S_-|^2 + |S_+|^2), \quad (12b)$$

$$d_{2,0}(t) = -\frac{p(t)\sqrt{10}}{N(t)}(P_+^*|S_-|^2S_+^+ + P_-^*|S_-|^2S_-^+), \quad (12c)$$

$$d_{2,2}(t) = \frac{p(t)\sqrt{2}}{N(t)}S_+^2(P_+^*S_-S_+ - P_-^*6|S_-|^2 + |S_+|^2). \quad (12d)$$

B. The fully coupled nine-state system

The full, $\sigma^+\pi\sigma^-$-coupled nine-state (1-3-5) system, has three dark states [17]. As discussed above, two of them $\Psi_D^{(1)}$ and $\Psi_D^{(2)}$ are stationary and have only $J_z=2$ components. There is a considerable freedom in the choice of these two dark states because they are degenerate and define a two-dimensional dark subspace, in which any superposition of them is also a dark state. We use this leeway to an advantage because, as discussed above, in the adiabatic limit these dark states do not participate in the dynamics and there role is only to define unambiguously the transfer dark state $\Psi_D(t)$ through the orthogonality condition (9). Moreover, because of this latter argument, we do not have to orthonormalize them but only to ensure that they are linearly independent. We choose them as

$$\Psi_D^{(1)} = \begin{bmatrix} 0,0,0,0,0,0,0,0,0 \end{bmatrix}^{T} \quad (13a)$$

$$\Psi_D^{(2)} = \begin{bmatrix} 0,0,0,0,0,0,0,0,0 \end{bmatrix}^{T} \quad (13b)$$

The transfer dark state $\Psi_D(t)$, Eq. (7), is determined as follows: (i) the eigenvalue equation (6) provides three equations for the six components of $\Psi_D(t)$; (ii) the orthogonality conditions (9) provide two more equations; (iii) the normalization condition provides the last, sixth equation. This completes the unambiguous definition of the components of $\Psi_D(t)$, apart from an irrelevant common phase factor.

The solution of Eqs. (6) and (9) is rather tedious but straightforward. The resulting components are presented in the Appendix.

In either case, for the full nine-state system and the reduced six-state system, it is easy to show from the explicit forms of the transfer dark states (12) and (A1) that in the adiabatic limit a STIRAP-like population transfer is achieved from the single $J_z=0$ level to the $J_z=2$ manifold. Indeed, for counterintuitively ordered pulses (Stokes before pump), we have $p(t)/s(t)\to0$ initially and hence, $\Psi_D(t\to-\infty)\to|J_z=0,M_z=0\rangle$. At large times, the opposite relation $s(t)/p(t)\to0$ applies and hence $\Psi_D(t\to+\infty)$ is a superposition of $|J_z=2,M_z\rangle$ states. The composition of this superposition is determined by Eqs. (A1b)–(A1f), without the time-dependent factors $p(t)/N(t)$.

IV. PHASE ANALYSIS

In the traditional STIRAP process the phases of the laser pulses enter the final superposition state in a trivial manner: the pulse phases determine only the phases of the probability amplitudes but they do not affect their magnitudes. This property holds also for the degenerate systems studied in Refs. [4,12,13]. The common property of these systems is that there are no closed loops in the coupling configuration: in all of the above cited works there is a single pathway from the single initial state to each final state. However, in our case there are multiple interfering pathways between the initial state and at least some of the final states (see Fig. 1). There is a phase associated with each pathway that depends on the phase of the laser pulses. The pathways interfere throughout the population transfer process, hence the final populations will depend not only on the relative amplitudes of the laser pulses, but on the relative phases as well.

In this section we have a closer look at our system from the phase point of view. To this end, we perform a phase transformation on the Hamiltonian of Eq. (1). The diagonal unitary transformation matrix is defined by
\[ V = \text{diag}[1, \exp(i \theta_{1}^{p}), \exp(i \theta_{0}^{p}), \exp(i \theta_{1}^{s}), \exp(i \theta_{2}^{s})] \]  

(14)

The phases of the matrix elements of the transformed Hamiltonian \( VH^{l} \) can be varied by changing the phases in \( V \). We can eliminate most of them by choosing \( \theta_{l}^{p} \) appropriately, although some of the phases persist, and they will contribute to not only the phase but the magnitude of the probability amplitudes of the created final state as well.

**A. The six-state subsystem**

The Hamiltonian of the \( \sigma^{+} \sigma^{-} \)-coupled six-state subsystem is given by Eqs. (1)-(3), except that the \( \pi \) components of both the pump and Stokes pulses are missing. We can eliminate all, but one phase in this Hamiltonian by choosing the phase parameters of \( V \) in Eq. (14) as

\[ \theta_{1}^{p} = \alpha_{p}^{+}, \]  

(15a)

\[ \theta_{0}^{p} = \alpha_{p}^{-}, \]  

(15b)

\[ \theta_{2}^{p} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(15c)

\[ \theta_{0}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(15d)

\[ \theta_{2}^{s} = \alpha_{p}^{-} + \alpha_{s}^{-}, \]  

(15e)

where the rest of the phases \( \theta_{l}^{p} \) are set to zero. The phase factor that cannot be eliminated from the Hamiltonian is given by

\[ \rho = \Delta \alpha_{p} - \Delta \alpha_{s}, \]  

(16)

where \( \Delta \alpha_{p} = \alpha_{p}^{+} - \alpha_{s}^{-} \) is the phase difference between the \( \sigma^{+} \) and \( \sigma^{-} \) components of the pump and Stokes pulses. As can be easily seen, the phase parameter of Eq. (16) persists; it cannot be eliminated by rotating the quantization axis, although its numerical value depends on the choice of the quantization axis.

Let us assume that in the phase-transformed basis we can find the polarizations and phases of the coupling fields that produce a prescribed final superposition state. The phases will determine the value of the phase \( \rho \) of Eq. (16). The created superposition state can be described by three phases \( \varphi_{p}^{p}, \varphi_{0}^{p}, \varphi_{p}^{s} \) in the phase-transformed basis, which contribute to two relevant phase differences \( \varphi_{p}^{p} - \varphi_{0}^{p} \) and \( \varphi_{p}^{s} - \varphi_{0}^{s} \), since the global phase is irrelevant. However, we require the phase differences \( \varphi_{p} - \varphi_{0} \) for the final state in the initial, bare atomic basis. Using the phase relations between the bare and phase-transformed bases one can easily derive the relation

\[ \Delta \alpha_{p} + \Delta \alpha_{s} = (\varphi_{p}^{p} - 2 \varphi_{0}^{p} + \varphi_{p}^{s}) - (\varphi_{p}^{p} - 2 \varphi_{0}^{p} + \varphi_{2}). \]  

(17)

We have two linear equations (16) and (17) for the sum and the difference of the phase differences \( \Delta \alpha_{p} \) and \( \Delta \alpha_{s} \). They can be easily solved and, therefore, the relative phases of the created final state can be adjusted independently of the populations.

**B. The fully coupled nine-state system**

In the fully coupled nine-state system both the \( \sigma^{\pm} \) and the \( \pi \) components are present in the model Hamiltonian of Eqs. (1)-(3). We can eliminate all, but four phases in the Hamiltonian by choosing

\[ \theta_{1}^{p} = \alpha_{p}^{+}, \]  

(18a)

\[ \theta_{0}^{p} = \alpha_{p}^{-}, \]  

(18b)

\[ \theta_{2}^{p} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18c)

\[ \theta_{2}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18d)

\[ \theta_{0}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18e)

\[ \theta_{2}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18f)

\[ \theta_{2}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18g)

\[ \theta_{2}^{s} = \alpha_{p}^{+} + \alpha_{s}^{-}, \]  

(18h)

for the phase transformation \( V \) in Eq. (14). The four phases that cannot be eliminated are associated with \( 2 \times 4 \) elements of the phase-transformed Hamiltonian (the factor 2 results from the Hermicity of the Hamiltonian). These phases are given by

\[ \varrho_{1} = - \Delta \alpha_{p}^{+} + \Delta \alpha_{s}^{-}, \]  

(19a)

\[ \varrho_{2} = - \Delta \alpha_{p}^{+} + \Delta \alpha_{s}^{-}, \]  

(19b)

\[ \varrho_{3} = \Delta \alpha_{p}^{-} - \Delta \alpha_{s}^{-}, \]  

(19c)

\[ \varrho_{4} = - \Delta \alpha_{p}^{+} + \Delta \alpha_{s}^{0} + \Delta \alpha_{s}^{-} - \Delta \alpha_{s}^{-}, \]  

(19d)

where \( \Delta \alpha_{x}^{q} = \alpha_{x}^{q} - \alpha_{s}^{q} \) for \( q = +, - \) and \( x = p, s \). Note that \( \varrho_{4} = \varrho_{1} + \varrho_{3} \); therefore, instead of six there are only three different phase factors that determine the magnitudes of the probability amplitudes in the final state of the system, beside the polarizations and the shapes of the pulses. The equations (19a)-(19c) provide three restrictions for the phases of the six polarization components of the pump and Stokes pulses. Consequently, there are only three phases that can be chosen arbitrarily. However, in a five-component final state, there are four relative phases, plus an unimportant common phase. Hence, it follows that in order to reach all possible states in the final state space with a prescribed population distribution and the associated phases it is necessary to vary the phases \( \varrho_{1}, \varrho_{2}, \varrho_{3} \) as well. Comparing the six-state subsystem to the nine-state fully coupled system we conclude that in the latter case the more complex linkage results in the loss of independent control of the populations and phases in the final superposition state in the most general case.

**V. COMPLETENESS AND THE INVERSE PROBLEM**

When we consider state preparation in a degenerate system it is necessary to examine whether the complete target
These dark states form the last system final state space is hold. Let the dimension of the two components where \( v \) state condition of Eq. (5) to express the components of the dark state \( \Psi_D \), to obtain
\[
e^{-2\alpha_x}v_2 - \sqrt{6}e^{-i(a_x' + \alpha_x)} \tan(\varphi) v_0 + e^{-2\alpha_x^x} \tan^2(\varphi) v_2 = 0,
\]
where we have set \( \eta_x = \pi/2 \), because the component \( S_x \) of the Stokes field of Eq. (5) is missing. The solution of Eq. (24) is trivial for \( v_2 = 0 \). For nonzero \( v_2 \) the solution for \( \tan(\varphi) \) reads
\[
\tan(\varphi) = e^{i(a_x' - a_x)} \sqrt{6v_0 + \sqrt{6v_0^2 - 4v_2v_2}} \frac{v_2}{v_2}.
\]
One can always set the phases \( a_x' \) so that the right-hand side of Eq. (25) is real. In this way we have shown that for the six-level subsystem the entire final state space can be reached by the transfer dark state (11) by a suitable choice of the pump and Stokes field polarizations and phases.

### B. The fully coupled nine-level system

Now the excited state space is \( N_e = 3 \) dimensional, and the final state space is \( N_f = 5 \) dimensional. In this case Eq. (22) implies
\[
\Psi_D^{(1)} = 0, \\
\Psi_D^{(2)} = 0.
\]
where \( \Psi_D^{(1)} \) and \( \Psi_D^{(2)} \) are defined by Eqs. (13a) and (13b), respectively. One can always adjust the three components \( P_+ \) and \( P_- \) of the pump coupling matrix so that Eq. (26a) is satisfied. By making use of the notation \( a = S_x'/S_x^2 \) and \( b = S_y'/S_y^2 \), Eqs. (26b) and (26c) take the forms
\[
\Psi_D^{(1)} = 0, \\
\Psi_D^{(2)} = 0,
\]
and the solutions of these equations can be expressed as
\[
b = v_2a^3 + 3v_0a^2 + 2v_1a + \sqrt{2}v_2, \\
0 = \frac{1}{(v_1a + \sqrt{2}v_2)a} \sum_{i=0}^{6} Q_i a^i.
\]
\[ Q_0 = 2v^3, \]  
\[ Q_1 = 6 \sqrt{2}v_1v_2^2, \]  
\[ Q_2 = 2(6v_0v_2^2 + 12v_1^2v_2), \]  
\[ Q_3 = 4(2v_3 + 8\sqrt{3}v_0v_1v_2), \]  
\[ Q_4 = 2v_-v_1v_2 + 4\sqrt{6}v_0v_2^2 + 3v_0^2v_2 - 2v_-v_2^2, \]  
\[ Q_5 = 3[2v_3^2v_1 - 2(\sqrt{6}v_2v_1 + 2\sqrt{2}v_-v_1^2, \quad Q_6 = -v_3^2v_2 - v_-v_2^2 + \sqrt{6}v_-v_0v_1, \]  
Equation (28b) is an algebraic equation for \( a \). According to Gauss’s fundamental theorem of algebra [20], every polynomial equation of degree \( n \) with complex coefficients has \( n \) roots in the complex numbers. Therefore, Eq. (28b) has six roots for \( a \), which implies at least one root (if all roots are equal) and at most six roots (if they are all different). These solutions can be obtained by numerical methods. Inserting the values of \( a \) and the components of \( \mathbf{v} \) into Eq. (28a), we obtain the parameter \( b \). These last steps complete the proof that in the nine-level fully coupled system the entire final state space can be reached by the transfer dark state.

**C. The inverse problem**

In the previous two subsections we have shown for the six-level subsystem and for the nine-level fully coupled system that the entire final state space can be covered with the transfer dark state by a suitable choice of the pump and Stokes field phases and ellipticities. Moreover, the presented proofs provide a constructive recipe to determine the field parameters that produce a prescribed final superposition state. Let us consider first the six-level subsystem: one can use Eq. (25) to obtain the relative phase and amplitude of the \( \sigma^k \) components of the Stokes field, i.e., the elements of the Stokes field coupling matrix \( S \), by making use of the components of the required final state \( \mathbf{v}_f \). Then, using the equivalent form of the relation (22),

\[ P^* = -N_d^fS\mathbf{v}_f, \]

one can easily determine the phase and the ellipticity of the pump field. In the case of the nine-level fully coupled system one can follow the same procedure, but start from Eq. (27).

**VI. EXAMPLES**

To demonstrate the efficiency of our method for finding the field parameters for the prescribed final states we consider some numeric examples. We choose three different target states in the \( J_f = 2 \) manifold \( \{ J_f = 2, M_f \} \) in the nine-level fully coupled system defined as

\[ \Psi_1 = \frac{1}{\sqrt{5}}[e^{-i\pi/4}, e^{-i\pi/6}, 1, e^{i\pi/12}, e^{i\pi/8}]^T, \]

\[ \Psi_2 = \frac{1}{2}[e^{-i\pi/7}, 0, 0, \sqrt{2}e^{i2\pi/7}, e^{i2\pi/7}]^T, \]

\[ \Psi_3 = \frac{1}{\sqrt{3}}[0, 1, e^{i\pi/5}, e^{i4\pi/5}]^T. \]

The necessary parameters of the pump and Stokes fields are calculated as described in the preceding section. The obtained values are listed in Table I. Note that for \( \Psi_3 \), Eq. (31c), the roots of Eq. (28b) are degenerate; hence there are only three different solutions. We have found excellent agreement between the final states obtained by the numeric integration of the Schrödinger equation using the parameters of Table I, and the prescribed final states of Eq. (31).

In Fig. 2 we show the time evolution of the populations of the \( J_f = 2 \) sublevels, using the parameters in the first line for \( \Psi_1 \) of Table I. The evolution of the populations is nearly the same for the following five parameter sets for \( \Psi_1 \) too.

**VII. IMPLEMENTATION**

We now turn to the discussion of the experimental feasibility of the proposed technique. As a specific example, we consider implementation in a crossed-beam experiment with metastable neon atoms, a setup that has been used extensively by Bergmann and co-workers [4–7]. In this setup, the Ne\(^*\) atoms are produced in a gas discharge and form an atomic beam. The atoms are prepared initially by optical pumping in the metastable state \(^3P_0\), which corresponds to the initial state \( J_f = 0 \) in our model system. The excited level \(^3P_1\) serves as the intermediate level with \( J_f = 1 \) and the metastable level \(^3P_2\) represents the final target level with \( J_f = 2 \), where the desired superposition state is to be created. The atoms enter a main chamber where the collinear pump laser driving the transition \(^3P_0 \leftrightarrow ^3P_1\), and Stokes laser driving the transition \(^3P_2 \leftrightarrow ^3P_0\), cross the atomic beam at right angles. An appropriate displacement of the two laser beams produces the time delay of the two interactions in the atomic rest frame and the typical counterintuitive pulse sequence of STIRAP. Each laser beam is elliptically polarized such that it can be decomposed into right-handed and left-handed circularly polarized fields. The quantization axis is chosen to be parallel to the wave vector \( \mathbf{k} \) of the (collinear) laser beams. This arrangement realizes the reduced, six-state \( \sigma^+\sigma^- \)-coupled system, in which the proposed technique can produce a coherent superposition of the magnetic sublevels with \( M = -2, 0, +2 \) of the \( J_f = 2 \) level.

The typical experimental parameters are as follows: the longitudinal velocity of the atoms in the atomic beam \( \nu \sim 800\pm 150 \text{ m/s} \), the diameters of the laser beams \( d \sim 2 \) mm, and the power of the lasers \( W \sim 150 \text{ mW} \) [4–7]. Therefore, the interaction time of the atoms with each laser field is \( 2\pi \sim 2.5 \) \( \mu \)s. The laser beams have nearly Gaussian intensity profiles. For such laser intensities the peak Rabi frequencies are about \( \Omega_p \sim 600 \text{ MHz} \) and \( \Omega_i \sim 500 \text{ MHz} \), which, combined with the interaction times, imply pulse areas of over 100\( \pi \); these areas are well above the required ones (10\( \pi \sim 30\pi \)) for adiabatic evolution [2].
These considerations show that the proposed state-preparation technique can be realized with the presently available experimental facilities in its reduced, $\sigma^+\sigma^-$-coupled six-state version, which produces a coherent superposition of the magnetic sublevels $M_f=-2,0,+2$ of the $J_f=2$ level.

In principle, the full $\sigma^+\sigma^-$-coupled nine-state version of the proposed technique, which can produce a coherent superposition of all five magnetic sublevels $M_f=-2,-1,0,+1,+2$ of the $J_f=2$ level, is difficult to be realized in a crossed-beam experiment. The reason is that the $\sigma^-$-polarized laser beams must cross both the atomic beam and the elliptically polarized laser beams at right angles. Because the width of the atomic beam (a few millimeters) greatly exceeds the laser wavelengths, the relative phase between the $\sigma^-$-polarized laser beams and the elliptically polarized beams is not well defined. Hence we are left with only two relative laser phases, instead of four, and we cannot access the entire Hilbert space of the $J_f=2$ manifold. This technique nevertheless allows one to reach a vast portion of this space, for example, any population distribution.

The full $\sigma^+\sigma^-$-coupled nine-state technique is relevant in well-localized systems, where the relative phases of all laser fields can be well defined, for example, in ion traps. It may also be possible to implement the full scheme in the neon experiment by using a rotation of the quantization axis to provide the two missing parameters; such an implementation, however, lies beyond the scope of the present paper.

### VIII. SUMMARY

In this paper we have proposed and analyzed a technique for efficient and robust preparation of preselected coherent superpositions of the $2J+1$ magnetic sublevels of a degenerate level with an angular momentum $J$. We have illustrated this technique by considering in detail the important special case $J_g=0\rightarrow J_e=1\rightarrow J_f=2$. This technique represents an extension of STIRAP to degenerate levels, in which a pump pulse drives the transition $J_g=0\rightarrow J_e=1$ and a Stokes pulse drives the transition $J_e=1\rightarrow J_f=2$, with the Stokes coming first. This technique, therefore, shares all the advantages of STIRAP in terms of efficiency, robustness and immunity to decay from the intermediate level $J_e=1$, which remains unpopulated throughout the interaction. We have demonstrated that, starting in the $J_g=0$ level, one can create any desired superposition of the $J_f=2$ sublevels by using elliptically po-

![Image](https://example.com/image.png)

**FIG. 2.** (Color online) Time evolution of the populations in the STIRAP process characterized by the parameters in the first line of Table I. The target state is given by $\Psi_1$ in Eq. (31). The populations $P(2,M), M=-2,\ldots,2$, all coincide.

---

**TABLE I.** The calculated field parameters that produce the prescribed final states of Eq. (31).

<table>
<thead>
<tr>
<th>$\eta_p$</th>
<th>$\phi_p$</th>
<th>$\Delta\alpha_0^-$</th>
<th>$\Delta\alpha_0^+$</th>
<th>$\eta_3$</th>
<th>$\phi_3$</th>
<th>$\Delta\alpha_3^-$</th>
<th>$\Delta\alpha_3^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\eta_1$</td>
<td>$\phi_1$</td>
<td>$\Delta\alpha_1^-$</td>
<td>$\Delta\alpha_1^+$</td>
<td>$\eta_2$</td>
<td>$\phi_2$</td>
<td>$\Delta\alpha_2^-$</td>
<td>$\Delta\alpha_2^+$</td>
</tr>
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<td>-1.0813</td>
<td>-0.9161</td>
<td>0.44</td>
<td>0.2638</td>
<td>-0.8253</td>
<td>-0.5662</td>
</tr>
<tr>
<td>0.2946</td>
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<td>0.9702</td>
<td>1.0665</td>
<td>0.3661</td>
<td>0.1981</td>
<td>0.7215</td>
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<tr>
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<td>0.3217</td>
<td>0.3019</td>
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<td>0.9546</td>
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<td>0.9114</td>
<td>-0.9727</td>
</tr>
<tr>
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<td>0.8315</td>
<td>1.2292</td>
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<td>0.8055</td>
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<td>1.0129</td>
<td>1.0397</td>
<td>0.2776</td>
<td>0.2531</td>
<td>0.6189</td>
<td>-0.567</td>
</tr>
<tr>
<td>$\Psi_1$</td>
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<td>0.1172</td>
<td>-1.0638</td>
<td>-0.1291</td>
<td>0.3717</td>
<td>0.1563</td>
<td>-0.8941</td>
</tr>
<tr>
<td>0.4085</td>
<td>0.1893</td>
<td>-1.0915</td>
<td>-0.0722</td>
<td>0.2757</td>
<td>0.3455</td>
<td>0.9366</td>
<td>0.5802</td>
</tr>
<tr>
<td>0.4054</td>
<td>0.194</td>
<td>0.8166</td>
<td>0.5577</td>
<td>0.2857</td>
<td>0.3437</td>
<td>0.7926</td>
<td>-0.1936</td>
</tr>
<tr>
<td>0.4247</td>
<td>0.1773</td>
<td>0.8499</td>
<td>1.2336</td>
<td>0.2428</td>
<td>0.3637</td>
<td>0.847</td>
<td>-0.8151</td>
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<tr>
<td>0.3042</td>
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<td>0.7458</td>
<td>0.5007</td>
<td>0.3658</td>
<td>0.1545</td>
<td>0.5095</td>
<td>-0.4198</td>
</tr>
<tr>
<td>0.1875</td>
<td>0.2394</td>
<td>0.9095</td>
<td>-0.805</td>
<td>0.3582</td>
<td>0.1363</td>
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<td>0.1849</td>
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<td>0.2325</td>
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<td>-1.505</td>
<td>-0.865</td>
<td>0.2991</td>
<td>0</td>
<td>0.9423</td>
</tr>
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<td>0.3437</td>
<td>0.3766</td>
<td>0.4</td>
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</tr>
<tr>
<td>0.312</td>
<td>0.0603</td>
<td>0.705</td>
<td>1.5823</td>
<td>0.1713</td>
<td>0</td>
<td>0.2577</td>
<td>0</td>
</tr>
</tbody>
</table>
larized light for both fields. The parameters of the created superposition state are determined solely and unambiguously by the polarizations and the phases of the pump and Stokes fields.

In the most general case, each field contains $\sigma^+$, $\sigma^-$, and $\pi$ polarization components, which can be produced by two mutually perpendicular and fully overlapping linearly polarized laser beams: two pump beams and two Stokes beams. In this case all nine sublevels of the $J_f=0\rightarrow J_e=1 \rightarrow J_f=2$ system are coupled. We have shown that the eight independent parameters characterizing the pump and Stokes fields suffice to create any superposition from the five magnetic sublevels of the $J_f=2$ level, which is characterized by eight independent parameters as well: four populations and four relative phases. In other words any point in the Hilbert space of the $J_f=2$ manifold can be reached by a suitable choice of polarizations and phases. Moreover, we have not only proved mathematically this assertion, but have prescribed a procedure for the choice of polarizations and phases. We have illustrated this procedure with examples and have shown that in general, there are multiple sets of polarizations and phases, which create a specific superposition.

Because of practical considerations, we have in parallel studied the simpler case when only $\sigma^+$ and $\pi$ polarizations are present. In this case only six sublevels of the $J_f=0\rightarrow J_e=1 \rightarrow J_f=2$ system are coupled. This arrangement allows one to create an arbitrary preselected superposition of the magnetic sublevels $M_f=-2,0,2$ of the $J_f=2$ level. In this case only two elliptically polarized lasers are needed: one pump and one Stokes.

We have discussed the experimental feasibility of this techniques by considering specifically neon atoms, in which a number of STIRAP related experiments have been carried out. We have concluded that, besides a careful control of polarizations, the present technique does not pose further challenges and should be feasible with present laboratory resources.

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APPENDIX: DARK-STATE COMPONENTS FOR THE NINE-LEVEL FULLY COUPLED SYSTEM

The nonvanishing components of the transfer dark state for the nine-level system read

\[ d_{2,0}(t) = -\frac{p(t)}{N(t)} \sqrt{5} S'_e (P'_e [2S^2_S S^2_\pi + 2S^2_\pi S^2_e + (4|S|^2] - [S]^2 S_S^* S_e^*] - P'_e([3|S|^2 + 3|S|^2] S_S^* S_e^* + (2|S|^2 + 4|S|^2] S_S^* S_e^* + [8|S|^2 S^2_\pi + 3|S|^2 S_S^* S_e^* + 7|S|^2 S^2_e - 2S_S^2 S_e^* + c.c.)}, \]

\[ d_{2,1}(t) = -\frac{p(t)}{N(t)} \frac{\sqrt{10}}{3} (P'_e[-S^2_\pi S^2_S S_e^* + S^2_\pi S^2_e - 2S_S^2 S_e^* S_e^* - 3S_S^* S_e^* S_e^* - P'_e([S]^2 S_S^* S_e^* + (3|S|^2 S^2_S + 6|S|^2 S_S^* S_e^* - 2S_S^2 S_e^* S_e^* + [2|S|^2 S_S^* S_e^* + 2|S|^2 S^2_S + 3|S|^2 S_S^* S_e^* + 2|S|^2 S^2_S - 2S_S^2 S_e^* S_e^* - 2S_S^2 S_e^* S_e^*] + 4|S|^2 S^2_S S_e^* S_e^* + S^2_\pi S^2_e + 2S^2_\pi S^2_e - 2S^2_\pi S^2_e - 3S^* S^* S^* S_e^* S_e^*}, \]

\[ d_{2,2}(t) = -\frac{p(t)}{N(t)} \frac{\sqrt{5}}{3} S'_e (P'_e [6|S|^4 + 4|S|^4 + 8|S|^2 |S|^2] S_e^2 + 2|S|^2 |S|^2] S_e^2 + 3|S|^2 |S|^2] S_e^2 + 4|S|^2 |S|^2] S_e^2 + 2|S|^2 |S|^2] S_e^2 + (4|S|^2] S_e^2 - [S]^2 S_S^2 S_e^2 + 7|S|^2 S^2 S_e^2 - 2S^2 S_S^2 S_e^2 + c.c.)], \]

\[ d_{2,3}(t) = -\frac{p(t)}{N(t)} \frac{\sqrt{10}}{3} (P'_e[-2S_\pi S^2_S S_e^* + 2|S|^4 S_S^* + 3|S|^4] S_e^* + 4|S|^4 S_S^* + 2|S|^4 S_e^* + 5|S|^4] S_e^* S_S^* - 2S^2 S_S^* S_e^* - 2S^2 S_S^* S_e^* + 4|S|^2 S^2 S_S^* S_e^* + P'_e([S]^2 S_S^* S_e^* + (3|S|^2 S_S^* S_e^* + 2|S|^2 S_S^* S_e^* + 2|S|^2 S^2 S_S^* S_e^* + 3|S|^2 S^2 S_S^* S_e^* + 2|S|^2 S^2 S_S^* S_e^* + 2|S|^2 S^2 S_S^* S_e^*] + 3|S|^2 S^2 S_S^* S_e^* - 3S^* S^* S^* S_S^* S_e^*), \]

with $|S|^2 = |S|_e^2 + |S|_\pi^2 + |S|_e^2$. The structure of the components
$d_{2,M}(t)$ is quite complicated, but we can make some general observations. The components $d_{2,M}(t)$ and $d_{2,-M}(t)$, for $M = 1, 2$, are related by exchanging the terms $S_+ \leftrightarrow S_-$ and $P_+ \leftrightarrow P_-$. Furthermore, the expression for the component $d_{2,0}(t)$ is invariant upon the exchange of the subscripts $+\text{ and } -$. It is also readily verified that for $P_\pi = S_\pi = 0$ (no π-polarized pump and Stokes lasers), Eq. (A1) reduces to Eq. (12) for the $\sigma^+\sigma^-$-coupled six-state system.