Stark-chirped rapid adiabatic passage among degenerate-level manifolds

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We generalize Stark-chirped rapid adiabatic passage (SCRAP) to the case of two and three degenerate-level manifolds. The analysis of a degenerate-level system is facilitated by its subdivision into a set of smaller independently evolving subsystems corresponding to the minimal-sized invariant subspaces of the Hamiltonian. Population transfer from the starting to the final level is examined for different types of the invariant subspaces depending on the presence of dark states. It is shown that the complete transfer is feasible if the initial state is prepared into specific coherent superpositions. Our formalism is applicable to the general case of arbitrary numbers of degenerate states within each level and arbitrary couplings of the appropriate transitions. It represents a generalization of well-known Morris-Shore transformation to the case when the removed degeneracy of the sublevels leads to detuning from two-photon resonance. We give the application of the SCRAP formalism to the ^{87}Rb atom.

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I. INTRODUCTION

In a recently proposed technique of Stark-chirped rapid adiabatic passage (SCRAP), the population transfer among two atomic states is efficiently driven by the pump laser, while an intense, far off-resonant Stark laser modifies the transition probability between the two states by Stark-shifting their energies [1]. In this process the pump laser frequency is nearly resonant to the atomic transition so that ac Stark shift produces two separate crossings of diabatic energies during the Stark laser pulse. The essential element is that the pump Rabi frequency has to be appreciable only at one of the level crossings. It has been shown that the proper pump pulse intensity, duration, and partial overlap with the Stark pulse produce complete population transfer from one state to another [2]. Recently, SCRAP technique was extended for complete population transfer amongst three states. In the technique all three states have to be coupled simultaneously using three laser pulses. Near resonant Stokes laser pulse is applied in addition to the pump and the Stark laser pulses [3]. The population transfer is efficient if the population of an intermediate state is minimized by the proper laser pulse timing and by adiabatic evolution of the states. This technique was experimentally used for preparation of population inversion in NO molecules [4,5].

The SCRAP is proven to be a powerful alternative to another technique for selective population transfer, stimulated Raman adiabatic passage (STIRAP) [6–8]. In a typical STIRAP a pair of laser pulses in Raman resonance with the two ground states of a three level atomic Λ scheme is used to transfer population between two ground states. Two partially overlapping laser pulses produce dark state, i.e., a superposition state that is not coupled to the excited state. Population is transferred from initially populated ground state to another ground state by suitably varying the intensity of the two laser pulses, i.e., by adiabatically evolving the dark state from the initial ground state toward another ground state at the end of the transfer process. While the STIRAP is robust against the fluctuations of laser Rabi frequencies and temporal shape of the pump and Stokes pulses [9,10], it is unstable against Stark shifts of atomic levels. In systems with multiphoton transitions, SCRAP has the advantage over STIRAP because such transitions induce significant ac Stark shifts altering the two-photon Raman resonance that is substantial for the STIRAP population transfer. SCRAP is much more stable against unwanted Stark induced level shifts since it exploits the Stark shifts for producing energy crossings and does not rely on the exact two-photon resonance condition.

In this work we extend the SCRAP technique to a multilevel system consisting of two or three degenerate-level manifolds with arbitrary number of substates. The motivation of our study was the elegant generalization of STIRAP to a multilevel atom [11]. Our formalism relies on the decomposition of the Hilbert space of the system into minimal invariant subspaces to which the evolution of the system is restricted. It is a generalization of the Morris-Shore transformation [12,13] to the case when the removed degeneracy of the sublevels leads to detuning from two-photon resonance. We first study the possibility of SCRAP population transfer among two degenerate-level manifolds and afterward discuss the case of three degenerate-level manifolds. The method is applied to the adiabatic passage among two and three hyperfine levels in the Rb atom.

II. SCRAP IN A TWO-LEVEL ATOM

In this section we introduce the notation and set the stage for a general formalism. Consider SCRAP population transfer among two atomic degenerate-level manifolds (ground g, and final f), having energies $E_{g(f)}$. Transition $g-f$ is driven by the classical field pump pulse, while strong off-resonant Stark field pulse introduces dynamic Stark detunings. Let $\mathcal{G}=\{|g_i\rangle | i=1,\ldots,n_g\}$ and $\mathcal{F}=\{|f_j\rangle | j=1,\ldots,n_f\}$ be the bases of Hilbert spaces for manifolds g and f, respectively, consisting of bare atomic states. The state $|\Psi(t)\rangle$ of the system is represented in basis $\mathcal{F}\cup\mathcal{G}$ by the vector $\mathbf{C}(t)$ that incorporates explicit phases taken from frequency of the pump pulse, ω_p.

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We are concerned with coherent excitation so we will describe the dynamics by the time-dependent Schrödinger equation. In the rotating-wave picture and using rotating-wave approximation (RWA) we get the time-dependent Schrödinger equation for $\mathbf{C}(t)$,

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

Hamiltonian of the system is represented as

$$\mathbf{H}(t) = \hbar \begin{bmatrix} \boldsymbol{\Delta}_f + \mathcal{S}(t)\mathbf{S}_f & \frac{1}{2}\Omega_p(t)\mathbf{V}^\dagger \\ \frac{1}{2}\Omega_p(t)\mathbf{V} & \mathcal{S}(t)\mathbf{S}_g \end{bmatrix}, \tag{2}$$

where $\Omega_p(t)$ is the pump field Rabi frequency and $\mathbf{V}$ is the matrix representing the lowering operator that connects the states in manifold f to the states in manifold g. The n_f-dimensional diagonal matrix $\boldsymbol{\Delta}_f$ describes the static detuning of the pump frequency from the Bohr frequency of the transition $g-f$ and can be represented as $\boldsymbol{\Delta}_f = \Delta_f \mathbf{1}_{n_f}$, where $\mathbf{1}_{n_f}$ is n_f-dimensional unit matrix and $\Delta_f = (E_f - E_g)/\hbar - \omega_p$ is the common static detuning of all f states. The matrices $\mathbf{S}_f$ and $\mathbf{S}_g$ represent the Stark shift operators of the states in manifolds f and g, respectively. Their diagonal elements are proportional to the Stark shifts of the sublevels. All Stark shifts share the same time dependence, expressed by $\mathcal{S}(t)$, that arises from the laser Stark field variation in time. The quantity $\mathcal{S}(t)$ is proportional to the Stark pulse envelope and could be taken equal to the Stark shift of some chosen sublevel.

The structure of the RWA Hamiltonian of Eq. (2) is similar to that of the ordinary two-state SCRAP [1]. All time dependences are stored into $\Omega_p(t)$ and $\mathcal{S}(t)$, but instead of single ground and final states we now have degenerate manifolds of substates, and hence we have matrices $\mathbf{V}$, $\mathbf{S}_f$, $\mathbf{S}_g$, and $\boldsymbol{\Delta}_f$, instead of the single elements in ordinary two-state SCRAP case.

As an introduction to the general degenerate-level case, we examine the simplest case of equal sublevel Stark shifts

$$\mathbf{S}_f = s_f \mathbf{1}_{n_f}, \quad \mathbf{S}_g = s_g \mathbf{1}_{n_g}, \tag{3}$$

where s_f and s_g correspond to the common Stark shifts of the f and g substates, respectively. This will serve as a starting point for development of a degenerate-level formalism. The basic idea is to facilitate the analysis of a degenerate-level system by its subdivision into a set of smaller independently evolving subsystems. In the present case it is possible to find a suitable Morris-Shore (MS) transformation [12] of diabatic basis yielding a new adapted basis in which the dynamics of a coupled degenerate two-level system is reduced to a set of independently evolving nondegenerate two-state systems and a number of uncoupled (dark) states. It is easily seen that each two-state subsystem under SCRAP process evolves in a well-known manner [1,3]. Consequently, the case of a SCRAP population transfer between two atomic degenerate-level manifolds having equal sublevel Stark shifts is simply reduced to a set of independent nondegenerate two-state subsystems and dark states. Let us restate the above consideration from a more general point of view. Effectively, the MS transformation yields the decomposition of the state space into a set of minimal-sized subspaces to which the evolution is restricted. These subspaces correspond to minimal-sized invariant subspaces (hereafter, invariant subspaces) of the Hamiltonian $\mathbf{H}(t)$. Hence, to each independent nondegenerate two-state system and to each dark state corresponds an invariant subspace of the Hamiltonian. Concept of invariant subspaces extends the scope of the former approach based on MS transformation. Namely, two-photon resonance condition expressed by equal sublevel shifts in Eq. (3) is essential for the existence of MS transformation. Generally, the Stark field removes the sublevel degeneracy detuning the atomic sublevels from the two-photon resonance. The MS transformation does not exist in that case [12], but our concept of invariant subspaces is still applicable with likely altered size and number of subspaces. Therefore, the decomposition of the state space on the invariant subspaces is a generalization of MS transformation that is applicable in the general case of the removed sublevel degeneracy.

Analysis of a multilevel system is performed essentially by identifying the Hamiltonian invariant subspaces that depend substantially on couplings of the transitions and sublevel Stark shifts. Let $\mathcal{H}^{\text{inv}} = \mathcal{H}_f \oplus \mathcal{H}_g$ be such an invariant subspace formed by subspaces $\mathcal{H}_f$ and $\mathcal{H}_g$ corresponding to manifolds f and g, respectively. The defining condition $\mathbf{H}(t)\mathcal{H}^{\text{inv}} < \mathcal{H}^{\text{inv}}$ leads to the following requirements:

$$\boldsymbol{\Delta}_f \mathcal{H}_f < \mathcal{H}_f, \quad \boldsymbol{\Delta}_g \mathcal{H}_g < \mathcal{H}_g, \tag{4a}$$

$$\mathbf{S}_f \mathcal{H}_f < \mathcal{H}_f, \quad \mathbf{S}_g \mathcal{H}_g < \mathcal{H}_g, \tag{4b}$$

$$\mathbf{V}\mathcal{H}_f < \mathcal{H}_g, \quad \mathbf{V}^\dagger \mathcal{H}_g < \mathcal{H}_f. \tag{4c}$$

The conditions (4a) are trivially fulfilled and can be disregarded because the matrices $\boldsymbol{\Delta}_f$ and $\boldsymbol{\Delta}_g$ are constant multiples of appropriate unit matrices. Let $\mathcal{H}_g^d = \ker \mathbf{V}^\dagger$ be the subspace of states in manifold g that are dark to the transition $g \to f$, and let $\mathcal{H}_f^d = \ker \mathbf{V}$ be the subspace of states in manifold f that are dark to the transition $f \to g$. The conditions (4c) determine $\mathcal{H}_f$ ($\mathcal{H}_g$) up to a direct sum with some dark subspace from $\mathcal{H}_f^d$ ($\mathcal{H}_g^d$), and yield more gainful conditions

$$\mathbf{V}^\dagger\mathbf{V}\mathcal{H}_f < \mathcal{H}_f, \quad \mathbf{V}\mathbf{V}^\dagger\mathcal{H}_g < \mathcal{H}_g. \tag{5}$$

Refer briefly to the meaning of the operators involved in Eq. (5). Operator $\mathbf{V}$ couples the states from $\mathcal{H}_f$ to the states in $\mathbf{V}\mathcal{H}_f$ that belong to manifold g. On the other side, operator $\mathbf{V}^\dagger$ couples the states from $\mathbf{V}\mathcal{H}_f$ to the states in manifold f belonging to the subspace $\mathbf{V}^\dagger\mathbf{V}\mathcal{H}_f$ that may include the states external to $\mathcal{H}_f$. If one has to find the subsystems that evolve independently then the condition $\mathbf{V}^\dagger\mathbf{V}\mathcal{H}_f < \mathcal{H}_f$ naturally emerges because the interaction with the pump field must not drive the states out from $\mathcal{H}_f$. Therefore, $\mathcal{H}_f$ has to be invariant subspace of operator $\mathbf{V}^\dagger\mathbf{V}$, so that the evolution of the system is restricted within the subspace $\mathcal{H}_f \oplus \mathbf{V}\mathcal{H}_f$.

We are now ready to give an explicit construction of aforementioned invariant subspaces. Relations (4b) and (5) indicate that $\mathcal{H}_f$ should be common invariant subspace for $\mathbf{S}_f$ and $\mathbf{V}^\dagger\mathbf{V}$. Let $\mathcal{H}_{f,k}^{\mathrm{inv}}$, $k\in\{1,\ldots,n_f^{\mathrm{inv}}\}$, be minimal-sized common invariant subspaces of $\mathbf{V}^\dagger\mathbf{V}$ and $\mathbf{S}_f$. It is easily seen that the subspace $\mathcal{H}_{g,k}:=\mathbf{V}\mathcal{H}_{f,k}^{\mathrm{inv}}$ is invariant for $\mathbf{V}\mathbf{V}^\dagger$, but need not to be invariant for $\mathbf{S}_g$ due to the possibility that $\mathbf{S}_g$ couples the states from distinct subspaces $\mathcal{H}_{g,k}$ together with some states from dark subspace $\mathcal{H}_g^d$. Therefore, invariant subspaces $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$, common for both $\mathbf{S}_g$ and $\mathbf{V}\mathbf{V}^\dagger$, may be formed from several subspaces $\mathcal{H}_{g,k}$, $k\in I_\kappa$, accompanied with some subspace $\mathcal{H}_{g,\kappa}^d$ of $\mathcal{H}_g^d$, i.e., $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}:=\oplus_{k\in I_\kappa}\mathcal{H}_{g,k}\oplus\mathcal{H}_{g,\kappa}^d$, $\kappa\in\{1,\ldots,n_g^{\mathrm{inv}}\}$. It is worth noting that the set I_κ, containing the indices that label the subspaces $\mathcal{H}_{g,k}$ interlinked by $\mathbf{S}_g$, may be empty in the case that the corresponding invariant subspace entirely resides within an appropriate dark space. Finally, $\mathcal{H}_\kappa^{\mathrm{inv}}:=\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}\oplus\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ is invariant subspace for Hamiltonian $\mathbf{H}(t)$, including all subspaces $\mathcal{H}_{f,k}^{\mathrm{inv}}$ that are connected with $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$. The evolution during SCRAP process is restricted to $\mathcal{H}_\kappa^{\mathrm{inv}}$. Two different types of the invariant subspaces need to be considered.

First, if $\mathcal{H}_\kappa^{\mathrm{inv}}$ does not contain dark states from $\mathcal{H}_g^d$, it is possible to transfer all population from the subspace $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ into the subspace $\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}$, irrespective of the starting state. Namely, during SCRAP pulse sequence, all starting states in $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ are adiabatically connected to corresponding final states in $\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}$. This occurs because the evolution is decoupled from dark states that prohibit population transfer. The final states cannot be traced analytically unless related common invariant subspaces $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ and $\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}$ are one-dimensional, in which case there is one-to-one correspondence between starting and final states. In other cases the final states can be found only numerically because they depend on the parameters of the SCRAP process.

The second case is when the dark states are present in $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ due to interaction with the Stark field. Because the dark states suppress transfer of population to final level, not all states from $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ have the corresponding final states in $\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}$. Generally, $\oplus_{k\in I_\kappa}\dim\mathcal{H}_{g,k}$ states are adiabatically connected to final states in $\oplus_{k\in I_\kappa}\mathcal{H}_{f,k}^{\mathrm{inv}}$ enabling the population transfer, while $\dim\mathcal{H}_{g,\kappa}^d$ states do not contribute to the population transfer and preserve the population within ground level. In order to obtain the complete population transfer, it is necessary to prepare the initial state into specific coherent superpositions. Subspaces corresponding to each of the two groups of superpositions cannot be determined without knowing the parameters of the SCRAP process, apart from the trivial case $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}=\mathcal{H}_{g,\kappa}^d$. In the following subsection we give an illustrative example.

SCRAP among two hyperfine levels in ^{87}Rb

Here we apply the above formalism to the SCRAP between two hyperfine levels $5S_{1/2}$, $F_g=2$ and $5P_{1/2}$, $F_f=1$ of ^{87}Rb that are coupled by classical field with corresponding atomic lowering operator given by

$$\hat{V}=\hat{\mathbf{V}}\cdot\hat{\boldsymbol{\epsilon}}_F, \tag{6}$$

where $\hat{\boldsymbol{\epsilon}}_F$ is the polarization of the light field. The vector operator $\hat{\mathbf{V}}$ is defined by

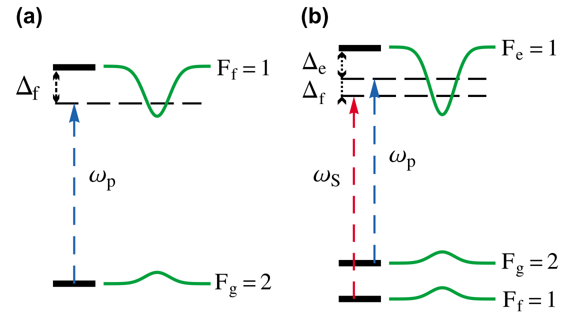

FIG. 1. (Color online) Atomic hyperfine level diagrams for (a) two-level and (b) three-level SCRAP. Solid curves schematically represent atomic dynamic level detuning induced by the Stark laser pulse. ω_p and ω_S are frequencies of the pump and Stokes laser fields, respectively. $\Delta_{f(e)}$ are static level detunings.

$$\hat{\mathbf{V}}=(-1)^{F_f+J_g+I+1}\sqrt{(2F_f+1)(2J_g+1)}\begin{Bmatrix}J_f & J_g & 1\\ F_g & F_f & I\end{Bmatrix}\times\sum_{q=-1}^{1}\sum_{m_g,m_f}\langle F_g,m_g|F_f,m_f;1,q\rangle|F_g,m_g\rangle\langle F_f,m_f|\mathbf{e}_q^*, \tag{7}$$

where $I=3/2$ is the nuclear quantum number of ^{87}Rb, $\{:::\}$ is Wigner 6j-symbol and $\langle F_g,m_g|F_f,m_f;1,q\rangle$ is the Clebsch-Gordan coefficient that connects the final level state $|F_f,m_f\rangle$ to the ground level state $|F_g,m_g\rangle$ via polarization $\mathbf{e}_q^*$,

$$\mathbf{e}_{\pm1}=\mp\frac{1}{\sqrt{2}}(\mathbf{e}_x\pm i\mathbf{e}_y),\quad \mathbf{e}_0=\mathbf{e}_z, \tag{8}$$

given in some orthonormal basis of polarization vectors. We choose the coordinate system such that the field propagates along the z axis, and define a basis of Zeeman states relative to this quantization axis. Bases of Hilbert spaces for manifolds f and g are

$$\mathcal{F}=\{|1,-1\rangle_f,|1,0\rangle_f,|1,1\rangle_f\}, \tag{9a}$$

$$\mathcal{G}=\{|2,-2\rangle_g,|2,-1\rangle_g,|2,0\rangle_g,|2,1\rangle_g,|2,2\rangle_g\}. \tag{9b}$$

Generally, depending on the Stark field polarization, Stark shifts of magnetic hyperfine sublevels have scalar, vector, and tensor part, having none, linear, and quadratic dependence on magnetic quantum number m_F, respectively. Hence, Stark field can remove the degeneracy of hyperfine levels through the linear and quadratic dependence of shift on m_F. In this example we assume that the Stark field is linearly polarized so that the linear dependence vanishes. Alkali ground hyperfine sublevels gain the shift that does not have tensor part, so the degeneracy of ground sublevels is preserved. Excited hyperfine sublevels gain both scalar and tensor shifts. We will assume that pump frequency is chosen such that $\Delta_f>0$, and that the Stark field frequency is such that the Stark shifts of the f (g) sublevels are negative (positive) [see Fig. 1(a)]. Off-diagonal elements of the Stark shift operators will be neglected for simplicity. In that manner we have the following structure of Stark shifts:

$$\mathbf{S}_g = s_g \,\mathrm{diag}\{\underbrace{1, \ldots, 1}_{2F_g+1}\}, \tag{10a}$$

$$\mathbf{S}_f = \mathrm{diag}\{-(1 + s_f m_f^2/F_f^2)|m_f = -F_f, \ldots, F_f\}, \tag{10b}$$

where s_g and s_f are constants arbitrarily chosen in this example. $\mathcal{S}(t)$ is chosen equal to the absolute value of the Stark shift of the final sublevel $|1,0\rangle_f$. For numerical calculations we will use Gaussian shapes for the laser pulses, yielding

$$\Omega_p(t) = \Omega_0 e^{-(t-\tau_p)^2/T_p^2}, \tag{11a}$$

$$\mathcal{S}(t) = S_0 e^{-t^2/T^2}. \tag{11b}$$

The Stark pulse center defines the time $t=0$. Relative to this, the peak of the pump pulse is at time τ_p, chosen to correspond to the first intersection of diabatic energies of ground level and final sublevel $|1,0\rangle_f$. We will use T_p as the unit of time and $1/T_p$ as the unit of frequency. We assume that the Stark pulse has duration $T=2T_p$. The polarization of the pump field is chosen to be linear along x axis, so the matrix representing the lowering operator is

$$\mathbf{V} = \begin{bmatrix} \frac{1}{2} & 0 & 0 \\ 0 & \frac{1}{\sqrt{8}} & 0 \\ -\frac{1}{\sqrt{24}} & 0 & \frac{1}{\sqrt{24}} \\ 0 & -\frac{1}{\sqrt{8}} & 0 \\ 0 & 0 & -\frac{1}{2} \end{bmatrix}. \tag{12}$$

Ground level dark subspace determined as kernel of $\mathbf{V}^\dagger$ is

$$\mathcal{H}_g^d = \mathrm{span}\{1/\sqrt{2}|2,-1\rangle_g + 1/\sqrt{2}|2,1\rangle_g, 1/\sqrt{8}|2,-2\rangle_g + \sqrt{3}/2|2,0\rangle_g + 1/\sqrt{8}|2,2\rangle_g\}. \tag{13}$$

There are three common invariant subspaces of $\mathbf{V}^\dagger\mathbf{V}$ and $\mathbf{S}_f$

$$\mathcal{H}_{f,1}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f - \frac{1}{\sqrt{2}}|1,1\rangle_f\right\}, \tag{14a}$$

$$\mathcal{H}_{f,2}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f + \frac{1}{\sqrt{2}}|1,1\rangle_f\right\}, \tag{14b}$$

$$\mathcal{H}_{f,3}^{\mathrm{inv}} = \mathrm{span}\{|1,0\rangle_f\}, \tag{14c}$$

and five common invariant subspaces of $\mathbf{V}\mathbf{V}^\dagger$ and $\mathbf{S}_g$

$$\mathcal{H}_{g,1}^{\mathrm{inv}} = \mathrm{span}\left\{\sqrt{\frac{3}{8}}|2,-2\rangle_g - \frac{1}{2}|2,0\rangle_g + \sqrt{\frac{3}{8}}|2,2\rangle_g\right\}, \tag{15a}$$

$$\mathcal{H}_{g,2}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-2\rangle_g - \frac{1}{\sqrt{2}}|2,2\rangle_g\right\}, \tag{15b}$$

$$\mathcal{H}_{g,3}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-1\rangle_g - \frac{1}{\sqrt{2}}|2,1\rangle_g\right\}, \tag{15c}$$

$$\mathcal{H}_{g,4}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{8}}|2,-2\rangle_g + \frac{\sqrt{3}}{2}|2,0\rangle_g + \frac{1}{\sqrt{8}}|2,2\rangle_g\right\}, \tag{15d}$$

$$\mathcal{H}_{g,5}^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-1\rangle_g + \frac{1}{\sqrt{2}}|2,1\rangle_g\right\}. \tag{15e}$$

Note that $\mathcal{H}_{g,4}^{\mathrm{inv}}$ and $\mathcal{H}_{g,5}^{\mathrm{inv}}$ are dark subspaces of $\mathcal{H}_g^d$. Combining results (14) and (15) we get five invariant subspaces to which the evolution is restricted

$$\mathcal{H}_1^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f - \frac{1}{\sqrt{2}}|1,1\rangle_f, \sqrt{\frac{3}{8}}|2,-2\rangle_g - \frac{1}{2}|2,0\rangle_g + \sqrt{\frac{3}{8}}|2,2\rangle_g\right\}, \tag{16a}$$

$$\mathcal{H}_2^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f + \frac{1}{\sqrt{2}}|1,1\rangle_f, \frac{1}{\sqrt{2}}|2,-2\rangle_g - \frac{1}{\sqrt{2}}|2,2\rangle_g\right\}, \tag{16b}$$

$$\mathcal{H}_3^{\mathrm{inv}} = \mathrm{span}\left\{|1,0\rangle_f, \frac{1}{\sqrt{2}}|2,-1\rangle_g - \frac{1}{\sqrt{2}}|2,1\rangle_g\right\}, \tag{16c}$$

$$\mathcal{H}_4^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{8}}|2,-2\rangle_g + \frac{\sqrt{3}}{2}|2,0\rangle_g + \frac{1}{\sqrt{8}}|2,2\rangle_g\right\}, \tag{16d}$$

$$\mathcal{H}_5^{\mathrm{inv}} = \mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-1\rangle_g + \frac{1}{\sqrt{2}}|2,1\rangle_g\right\}. \tag{16e}$$

Subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, $\kappa \in \{1,2,3\}$, do not contain dark states, thus it is possible to transfer all of the population from $\mathcal{H}_{g,\kappa}^{\mathrm{inv}}$ to $\mathcal{H}_{f,\kappa}^{\mathrm{inv}}$ for $\kappa \in \{1,2,3\}$. It is worth noting that the complete population transfer requires the starting states to be particular coherent superpositions. Contrary, population remains trapped within dark subspaces $\mathcal{H}_{g,4}^{\mathrm{inv}}$ and $\mathcal{H}_{g,5}^{\mathrm{inv}}$. Previous conclusions can be depicted by plotting the adiabatic energies corresponding to aforementioned invariant subspaces. Figure 2 shows time dependence of the pump and Stark pulse envelopes (topmost part) and adiabatic and diabatic energies versus time (other parts). First three plots of energies (from top to bottom) correspond to invariant subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, $\kappa \in \{1,2,3\}$. It can be seen that starting from appropriate ground state, all the population transfers into the related final state. Note that the application of SCRAP for the complete population transfer requires the preparation of the initial state into the specific coherent superpositions of magnetic ground hyperfine substates. The opposite situation is shown in the two lowest plots where the population rests within ground level subspaces $\mathcal{H}_{g,4}^{\mathrm{inv}}$ and $\mathcal{H}_{g,5}^{\mathrm{inv}}$, respectively.

III. SCRAP IN A THREE-LEVEL ATOM

We now analyze the SCRAP process between three atomic degenerate-level manifolds (ground g, excited e, and

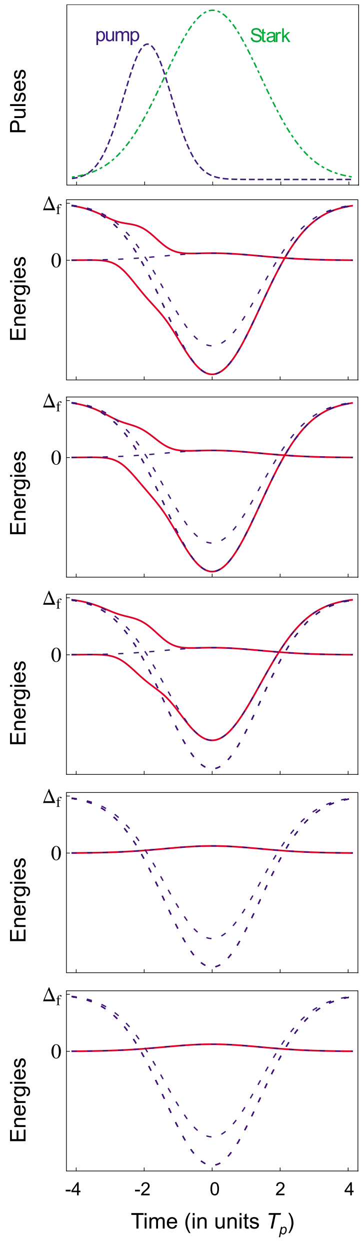

FIG. 2. (Color online) SCRAP among two hyperfine levels $5S_{1/2}$, F_g=2 and $5P_{1/2}$, F_f=1 in ^{87}Rb. Topmost: time dependence of the pump and Stark pulse envelopes (arbitrary scaled). Other: adiabatic (solid lines) and diabatic (dashed lines) energies versus time, related to the invariant subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, κ=1–5 (top—bottom). The dashed line starting from energy 0 corresponds to degenerate g states. The two dashed lines originating from Δ_f correspond to the states $|1,0\rangle_f$ (smaller shift) and $|1,\pm 1\rangle_f$ (larger shift). Used parameters are Δ_f=200/T_p, S_0=500/T_p, Ω_0=4/5S_0, s_g=1/20, s_f=1/5.

final f), having energies $E_{g(e,f)}$. The transitions $g-e$ and $f-e$ are driven by classical field pump and Stokes pulses, respectively. Strong off-resonant Stark field pulse is used to introduce dynamic Stark detunings. Similar to Section II, let $\mathcal{G}=\{|g_i\rangle | i=1,\ldots,n_g\}$, $\mathcal{F}=\{|f_j\rangle | j=1,\ldots,n_f\}$, and $\mathcal{E}=\{|e_k\rangle | k=1,\ldots,n_e\}$, denote the bases of Hilbert spaces for manifolds g, f, and e, respectively, consisting of bare atomic states. In the same manner, the state $|\Psi(t)\rangle$ of the system is now represented in basis $\mathcal{E}\cup\mathcal{G}\cup\mathcal{F}$ by the vector $\mathbf{C}(t)$ incorporating explicit phases taken from frequency of the pump and Stokes pulses, ω_p and ω_S. In the rotating-wave picture and using RWA we get the time-dependent Schrödinger equation analog to Eq. (1). Matrix representation $\mathbf{H}(t)$ of the system Hamiltonian has the form

$$\mathbf{H}(t)=\hbar\begin{bmatrix} \mathbf{\Delta}_e+\mathcal{S}(t)\mathbf{S}_e & \frac{1}{2}\Omega_p(t)\mathbf{V}_g^\dagger & \frac{1}{2}\Omega_S(t)\mathbf{V}_f^\dagger \\ \frac{1}{2}\Omega_p(t)\mathbf{V}_g & \mathcal{S}(t)\mathbf{S}_g & \mathbf{0} \\ \frac{1}{2}\Omega_S(t)\mathbf{V}_f & \mathbf{0} & \mathbf{\Delta}_f+\mathcal{S}(t)\mathbf{S}_f \end{bmatrix}, \quad (17)$$

where $\Omega_p(t)$ and $\Omega_S(t)$ are Rabi frequencies of the pump and Stokes field, respectively, and $\mathbf{V}_g$ ($\mathbf{V}_f$) is the matrix representing the lowering operator that connects the states in manifold e to the states in manifold g (f). The zeros $\mathbf{0}$ denote null rectangular matrices of appropriate dimensions. The diagonal matrices $\mathbf{\Delta}_e$ and $\mathbf{\Delta}_f$ describe static detunings and can be represented as $\mathbf{\Delta}_e=\Delta_e\mathbf{1}_{n_e}$ and $\mathbf{\Delta}_f=\Delta_f\mathbf{1}_{n_f}$, where common static detunings Δ_e and Δ_f for one-photon transitions are given by

$$\Delta_e=(E_e-E_g)/\hbar-\omega_p, \quad (18a)$$

$$\Delta_f=(E_f-E_g)/\hbar+\omega_S-\omega_p. \quad (18b)$$

The matrices $\mathbf{S}_e$, $\mathbf{S}_g$, and $\mathbf{S}_f$ correspond to the Stark shift operators of the states in manifolds e, g and f, respectively. Again, the quantity $\mathcal{S}(t)$ is proportional to the Stark pulse envelope and is chosen to introduce a referent Stark shift. The structure of the RWA Hamiltonian of Eq. (17) is similar to that of the conventional three-state SCRAP with single elements replaced by the matrices [3].

As in the Sec. II, we first inspect the case when Stark shifts of the sublevels are equal

$$\mathbf{S}_e=s_e\mathbf{1}_{n_e}, \quad \mathbf{S}_g=s_g\mathbf{1}_{n_g}, \quad \mathbf{S}_f=s_f\mathbf{1}_{n_f}, \quad (19)$$

where s_e, s_g, and s_f correspond to the common Stark shifts of the e, g, and f states, respectively. Again, we can utilize three-level MS transformation [13] to obtain sets of independently evolving nondegenerate three-state and two-state systems and a set of uncoupled (dark) states, provided the following condition is fulfilled

$$[\mathbf{V}_g^\dagger\mathbf{V}_g,\mathbf{V}_f^\dagger\mathbf{V}_f]=0. \quad (20)$$

To each such independently evolving nondegenerate system corresponds an invariant subspace of the Hamiltonian, as is already mentioned in Sec. II. The origin of operators involved in Eq. (20) has been addressed above, and we will briefly discuss the commutation condition. Consider some subspace $\mathcal{H}_e$ of states in manifold e. Following the discussion in Sec. II, if one has to find the subsystems that are dynamically independent, then $\mathcal{H}_e$ has to be common invariant subspace of operators $\mathbf{V}_g^\dagger\mathbf{V}_g$ and $\mathbf{V}_f^\dagger\mathbf{V}_f$, so that the evolution of the system is restricted to the subspace $\mathcal{H}_e\oplus\mathbf{V}_g\mathcal{H}_e\oplus\mathbf{V}_f\mathcal{H}_e$. The condition (20) assures that all minimal-sized common invariant subspaces are one-dimensional, i.e., that the corresponding three-state and two-state systems are nondegenerate. We note that two-state subsystems arise when one of subspaces $\mathbf{V}_g\mathcal{H}_e$ or $\mathbf{V}_f\mathcal{H}_e$ contains only null vector, i.e., when the states from $\mathcal{H}_e$ are dark to one of transitions $e\to g$ or $e\to f$. Conditions (19) and (20) that are essential for the MS transformation only affect the size and number of independently evolving invariant subspaces. Therefore, as in the two-level SCRAP case, the decomposition of the state space on the invariant subspaces generalizes MS transformation.

Let $\mathcal{H}^{\text{inv}}=\mathcal{H}_e\oplus\mathcal{H}_g\oplus\mathcal{H}_f$ be an invariant subspace for the Hamiltonian. The necessary condition $\mathbf{H}(t)\mathcal{H}^{\text{inv}}<\mathcal{H}^{\text{inv}}$ yields the following requirements:

$$\mathbf{\Delta}_e\mathcal{H}_e<\mathcal{H}_e,\quad \mathbf{\Delta}_g\mathcal{H}_g<\mathcal{H}_g,\quad \mathbf{\Delta}_f\mathcal{H}_f<\mathcal{H}_f, \tag{21a}$$

$$\mathbf{S}_e\mathcal{H}_e<\mathcal{H}_e,\quad \mathbf{S}_g\mathcal{H}_g<\mathcal{H}_g,\quad \mathbf{S}_f\mathcal{H}_f<\mathcal{H}_f, \tag{21b}$$

$$\mathbf{V}_g^\dagger\mathcal{H}_g<\mathcal{H}_e,\quad \mathbf{V}_f^\dagger\mathcal{H}_f<\mathcal{H}_e, \tag{21c}$$

$$\mathbf{V}_g\mathcal{H}_e<\mathcal{H}_g,\quad \mathbf{V}_f\mathcal{H}_e<\mathcal{H}_f. \tag{21d}$$

The conditions (21a) can be disregarded as trivially fulfilled. Let $\mathcal{H}_{ge}^d=\ker\mathbf{V}_g^\dagger$ ($\mathcal{H}_{fe}^d=\ker\mathbf{V}_f^\dagger$) be the subspace of states in manifold g (f) that are dark to the transition $g\to e$ ($f\to e$), and let $\mathcal{H}_{ef}^d=\ker\mathbf{V}_f$ be the subspace of states in manifold e that are dark to the transition $e\to f$. The conditions (21c) determine $\mathcal{H}_g$ ($\mathcal{H}_f$) up to a direct sum with some dark subspace of $\mathcal{H}_{ge}^d$ ($\mathcal{H}_{fe}^d$), and together with Eq. (21d) yield more useful conditions

$$\mathbf{V}_g^\dagger\mathbf{V}_g\mathcal{H}_e<\mathcal{H}_e,\quad \mathbf{V}_f^\dagger\mathbf{V}_f\mathcal{H}_e<\mathcal{H}_e, \tag{22a}$$

$$\mathbf{V}_g\mathbf{V}_g^\dagger\mathcal{H}_g<\mathcal{H}_g,\quad \mathbf{V}_f\mathbf{V}_f^\dagger\mathcal{H}_f<\mathcal{H}_f. \tag{22b}$$

Let $\mathcal{H}_{e,k}^{\text{inv}}$, $k\in\{1,\ldots,n_e^{\text{inv}}\}$, be common invariant subspaces of $\mathbf{V}_g^\dagger\mathbf{V}_g$, $\mathbf{V}_f^\dagger\mathbf{V}_f$, and $\mathbf{S}_e$. It is trivial to see that the subspace $\mathcal{H}_{g,k}:=\mathbf{V}_g\mathcal{H}_{e,k}^{\text{inv}}$ ($\mathcal{H}_{f,k}:=\mathbf{V}_f\mathcal{H}_{e,k}^{\text{inv}}$) is invariant for $\mathbf{V}_g\mathbf{V}_g^\dagger$ ($\mathbf{V}_f\mathbf{V}_f^\dagger$), but need not to be invariant for $\mathbf{S}_g$ ($\mathbf{S}_f$) due to the possibility that it links the states from different subspaces $\mathcal{H}_{g,k}$ ($\mathcal{H}_{f,k}$) together with some states from dark subspace $\mathcal{H}_{ge}^d$ ($\mathcal{H}_{fe}^d$). Hence, invariant subspaces $\mathcal{H}_{g,\kappa'}^{\text{inv}}$, common for both $\mathbf{S}_g$ and $\mathbf{V}_g\mathbf{V}_g^\dagger$ and connected with some of the subspaces $\mathcal{H}_{e,k}^{\text{inv}}$, may be formed from several subspaces $\mathcal{H}_{g,k}$ accompanied with some subspace $\mathcal{H}_{ge,\kappa'}^d$ of dark space $\mathcal{H}_{ge}^d$, i.e., $\mathcal{H}_{g,\kappa'}^{\text{inv}}:=\oplus_{k\in I_{g,\kappa'}}\mathcal{H}_{g,k}\oplus\mathcal{H}_{ge,\kappa'}^d$, $\kappa'\in\{1,\ldots,n_g^{\text{inv}}\}$, where the set $I_{g,\kappa'}$ contains the indices k labeling the subspaces $\mathcal{H}_{g,k}$ that are interconnected by $\mathbf{S}_g$. Analogously, $\mathcal{H}_{f,\kappa''}^{\text{inv}}:=\oplus_{k\in I_{f,\kappa''}}\mathcal{H}_{f,k}\oplus\mathcal{H}_{fe,\kappa''}^d$, $\kappa''\in\{1,\ldots,n_f^{\text{inv}}\}$. The sets of indices $I_{g,\kappa'}$ or $I_{f,\kappa''}$ may be empty in case that related invariant subspace entirely belongs to the appropriate dark subspace. Some of the nonempty sets $I_{g,\kappa'}$ may have a nonempty intersection with exactly one corresponding set $I_{f,\kappa''}$, because for at least one $k\in I_{g,\kappa'}\cap I_{f,\kappa''}$ the relations $\mathbf{V}_g\mathcal{H}_{e,k}^{\text{inv}}<\mathcal{H}_{g,\kappa'}^{\text{inv}}$ and $\mathbf{V}_f\mathcal{H}_{e,k}^{\text{inv}}<\mathcal{H}_{f,\kappa''}^{\text{inv}}$ may hold. Such subspaces $\mathcal{H}_{g,\kappa'}^{\text{inv}}$ and $\mathcal{H}_{f,\kappa''}^{\text{inv}}$ are then dynamically connected via the excited level subspace $\mathcal{H}_{e,k}^{\text{inv}}$ and the invariant subspace for Hamiltonian is composed as $\mathcal{H}_\kappa^{\text{inv}}:=\oplus_{k\in I_{g,\kappa'}\cup I_{f,\kappa''}}\mathcal{H}_{e,k}^{\text{inv}}\oplus\mathcal{H}_{g,\kappa'}^{\text{inv}}\oplus\mathcal{H}_{f,\kappa''}^{\text{inv}}$ including all subspaces $\mathcal{H}_{e,k}^{\text{inv}}$ that are connected with $\mathcal{H}_{g,\kappa'}^{\text{inv}}$ and $\mathcal{H}_{f,\kappa''}^{\text{inv}}$. If some of the nonempty sets $I_{g,\kappa'}$ does not have a nonempty intersection with any of the sets $I_{f,\kappa''}$, then the invariant subspace is constructed solely from subspaces related to g and e manifolds, i.e., $\mathcal{H}_\kappa^{\text{inv}}:=\oplus_{k\in I_{g,\kappa}}\mathcal{H}_{e,k}^{\text{inv}}\oplus\mathcal{H}_{g,\kappa}^{\text{inv}}$. That occurs if $\oplus_{k\in I_{g,\kappa}}\mathcal{H}_{e,k}^{\text{inv}}<\mathcal{H}_{ef}^d$. Similar situation may involve final end excited level resulting in $\mathcal{H}_\kappa^{\text{inv}}:=\oplus_{k\in I_{f,\kappa}}\mathcal{H}_{e,k}^{\text{inv}}\oplus\mathcal{H}_{f,\kappa}^{\text{inv}}$. Distinct types of the invariant subspaces depending of the presence of dark states need to be examined.

First, if $\mathcal{H}_\kappa^{\text{inv}}$ does not contain any dark state from $\mathcal{H}_{ge}^d$ nor from $\mathcal{H}_{ef}^d$, it is possible to transfer all the population from $\mathcal{H}_{g,\kappa'}^{\text{inv}}$ into $\mathcal{H}_{f,\kappa''}^{\text{inv}}$, irrespective of the starting state. All ground starting states from the subspace $\mathcal{H}_{g,\kappa'}^{\text{inv}}$ are adiabatically connected to the related ending states in the subspace $\mathcal{H}_{f,\kappa''}^{\text{inv}}$ enabling the complete population transfer. Exact ending states cannot be known in advance, unless the aforementioned invariant subspaces are one-dimensional. In such case there is one-to-one correspondence between the states at the beginning of the SCRAP process to the appropriate states at the end. In all other cases the ending states can be determined only numerically because of their dependence on the particular parameters of the SCRAP process.

The situation changes when the dark states are present in $\mathcal{H}_\kappa^{\text{inv}}$. The states from $\mathcal{H}_{ge}^d$ prevent population transfer from the level g to the level e, while the states from $\mathcal{H}_{ef}^d$ obstruct transfer of population from the level e toward the level f. Due to the presence of dark states from $\mathcal{H}_{ge}^d$ (or $\mathcal{H}_{fe}^d$), part of the starting population remains trapped within these states. If a number of dark states from $\mathcal{H}_{ef}^d$ are contained within appropriate excited level subspace of $\mathcal{H}_\kappa^{\text{inv}}$, there is the same number of ground starting states that are adiabatically connected to the states in the excited level subspace of $\mathcal{H}_\kappa^{\text{inv}}$. The rest of ground starting states are adiabatically connected to the ending states in the final level. Thus, it is required to prepare the starting state into specific coherent superpositions in order to perform the complete population transfer. Exact starting and ending superpositions cannot be found in advance, except in the case of one-dimensional adiabatically connected starting and ending subspaces. Otherwise, one must resort to numerics for particular choice of SCRAP parameters. In the next subsection we demonstrate previous considerations on the real atomic system.

SCRAP among three hyperfine levels in ^{87}Rb

As an example we will analyze SCRAP in ^{87}Rb from the ground hyperfine level $5S_{1/2}$, $F_g=2$ to the final level $5S_{1/2}$, $F_f=1$ via the excited level $5P_{1/2}$, $F_e=1$. Transitions $g-e$ and $f-e$ are driven by classical fields, pump and Stokes respectively, with corresponding atomic lowering operators given by

$$\hat{V}_g=\hat{\mathbf{V}}_g\cdot\hat{\boldsymbol{\epsilon}}_p,\quad \hat{V}_f=\hat{\mathbf{V}}_f\cdot\hat{\boldsymbol{\epsilon}}_S, \tag{23}$$

where $\hat{\boldsymbol{\epsilon}}_p$ and $\hat{\boldsymbol{\epsilon}}_S$ are the polarizations of the pump and Stokes field. The vector operators $\hat{\mathbf{V}}_g$ and $\hat{\mathbf{V}}_f$ are defined in analogy with Eq. (7). We choose the coordinate system such that the fields propagate along the z axis, and define a basis of Zeeman states relative to this quantization axis. Bases of Hilbert spaces for manifolds e, g, and f are

$$\mathcal{E}=\{|1,-1\rangle_e,|1,0\rangle_e,|1,1\rangle_e\}, \tag{24a}$$

$$\mathcal{G}=\{|2,-2\rangle_g,|2,-1\rangle_g,|2,0\rangle_g,|2,1\rangle_g,|2,2\rangle_g\}, \tag{24b}$$

$$\mathcal{F}=\{|1,-1\rangle_f,|1,0\rangle_f,|1,1\rangle_f\}. \tag{24c}$$

Similar to Sec. II, we assume that the Stark field is linearly polarized, so the degeneracy of ground levels is preserved. Excited hyperfine sublevels gain both scalar and tensor shifts. To assure necessary conditions for adiabatic connection between ground and final level [3], we choose pump and Stokes frequencies such that $\Delta_f<0$ and $\Delta_e>0$, and take Stark field frequency so that the Stark shifts of the e (g and f) sublevels are negative (positive) [see Fig. 1(b)]. Off-diagonal elements of the Stark shift operators are again neglected for simplicity. Sublevel Stark shifts have the form

$$\mathbf{S}_g=s_g\,\mathrm{diag}\{\underbrace{1,\ldots,1}_{2F_g+1}\},\quad \mathbf{S}_f=s_f\,\mathrm{diag}\{\underbrace{1,\ldots,1}_{2F_f+1}\}, \tag{25a}$$

$$\mathbf{S}_e=\mathrm{diag}\{-(1+s_e m_e^2/F_e^2)|m_e=-F_e,\ldots,F_e\}, \tag{25b}$$

where s_g, s_f and s_e are constants arbitrarily chosen in this example. $\mathcal{S}(t)$ is taken equal to the absolute value of the Stark shift of the excited sublevel $|1,0\rangle_e$. For numerical calculations we shall assume Gaussian shapes for all pulses, and take the pump and Stokes Rabi frequencies to have identical peak values Ω_0, obtaining

$$\Omega_p(t)=\Omega_0 e^{-(t-\tau_p)^2/T_p^2}, \tag{26a}$$

$$\Omega_S(t)=\Omega_0 e^{-(t-\tau_S)^2/T_S^2}, \tag{26b}$$

$$\mathcal{S}(t)=S_0 e^{-t^2/T^2}. \tag{26c}$$

We will also take equal pump and Stokes durations, $T_p=T_S$ and $T=2T_p$. Stark field peak value is taken large enough to assure necessary diabatic energy crossings. The timings τ_p and τ_S of pulses are chosen to correspond to appropriate first crossings of diabatic energies of ground and final level with diabatic energy of excited sublevel $|1,0\rangle_e$, in the "counterintuitive" order [3]. The polarizations of the pump and Stokes field are both chosen to be linear along x axis, so the matrices representing lowering operators are

$$\mathbf{V}_g=\begin{bmatrix}\frac{1}{2}&0&0\\0&\frac{1}{\sqrt{8}}&0\\\frac{1}{\sqrt{24}}&0&\frac{1}{\sqrt{24}}\\0&\frac{1}{\sqrt{8}}&0\\0&0&\frac{1}{2}\end{bmatrix},\quad \mathbf{V}_f=\begin{bmatrix}0&-\frac{1}{\sqrt{24}}&0\\\frac{1}{\sqrt{24}}&0&-\frac{1}{\sqrt{24}}\\0&\frac{1}{\sqrt{24}}&0\end{bmatrix}, \tag{27}$$

Dark subspaces are the following:

$$\mathcal{H}_{ge}^d=\mathrm{span}\{1/\sqrt{2}|2,-1\rangle_g+1/\sqrt{2}|2,1\rangle_g,1/\sqrt{8}|2,-2\rangle_g+\sqrt{3}/2|2,0\rangle_g+1/\sqrt{8}|2,2\rangle_g\}, \tag{28}$$

$$\mathcal{H}_{fe}^d=\mathrm{span}\{1/\sqrt{2}|1,-1\rangle_f-1/\sqrt{2}|1,1\rangle_f\}, \tag{29}$$

$$\mathcal{H}_{ef}^d=\mathrm{span}\{1/\sqrt{2}|1,-1\rangle_e-1/\sqrt{2}|1,1\rangle_e\}. \tag{30}$$

There are three common invariant subspaces for $\mathbf{V}_g^\dagger\mathbf{V}_g$, $\mathbf{V}_f^\dagger\mathbf{V}_f$, and $\mathbf{S}_e$

$$\mathcal{H}_{e,1}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_e-\frac{1}{\sqrt{2}}|1,1\rangle_e\right\}, \tag{31a}$$

$$\mathcal{H}_{e,2}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_e+\frac{1}{\sqrt{2}}|1,1\rangle_e\right\}, \tag{31b}$$

$$\mathcal{H}_{e,3}^{\mathrm{inv}}=\mathrm{span}\{|1,0\rangle_e\}, \tag{31c}$$

five invariant subspaces for $\mathbf{V}_g\mathbf{V}_g^\dagger$ and $\mathbf{S}_g$

$$\mathcal{H}_{g,1}^{\mathrm{inv}}=\mathrm{span}\left\{\sqrt{\frac{3}{8}}|2,-2\rangle_g-\frac{1}{2}|2,0\rangle_g+\sqrt{\frac{3}{8}}|2,2\rangle_g\right\}, \tag{32a}$$

$$\mathcal{H}_{g,2}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-2\rangle_g-\frac{1}{\sqrt{2}}|2,2\rangle_g\right\}, \tag{32b}$$

$$\mathcal{H}_{g,3}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-1\rangle_g-\frac{1}{\sqrt{2}}|2,1\rangle_g\right\}, \tag{32c}$$

$$\mathcal{H}_{g,4}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{2\sqrt{2}}|2,-2\rangle_g+\frac{\sqrt{3}}{2}|2,0\rangle_g+\frac{1}{2\sqrt{2}}|2,2\rangle_g\right\}, \tag{32d}$$

$$\mathcal{H}_{g,5}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|2,-1\rangle_g+\frac{1}{\sqrt{2}}|2,1\rangle_g\right\}, \tag{32e}$$

and three invariant subspaces for $\mathbf{V}_f\mathbf{V}_f^\dagger$ and $\mathbf{S}_f$

$$\mathcal{H}_{f,1}^{\mathrm{inv}}=\mathrm{span}\{|1,0\rangle_f\}, \tag{33a}$$

$$\mathcal{H}_{f,2}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f+\frac{1}{\sqrt{2}}|1,1\rangle_f\right\}, \tag{33b}$$

$$\mathcal{H}_{f,3}^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_f-\frac{1}{\sqrt{2}}|1,1\rangle_f\right\}. \tag{33c}$$

Six invariant subspaces of the Hamiltonian can be constructed using results (31a)–(31c), (32a)–(32e), and (33a)–(33c)

$$\mathcal{H}_1^{\mathrm{inv}}=\mathrm{span}\left\{\frac{1}{\sqrt{2}}|1,-1\rangle_e-\frac{1}{\sqrt{2}}|1,1\rangle_e,\ \sqrt{\frac{3}{8}}|2,-2\rangle_g-\frac{1}{2}|2,0\rangle_g+\sqrt{\frac{3}{8}}|2,2\rangle_g\right\}, \tag{34a}$$

$$\mathcal{H}_2^{\mathrm{inv}} = \mathrm{span}\left\{ \frac{1}{\sqrt{2}}|1,-1\rangle_e + \frac{1}{\sqrt{2}}|1,1\rangle_e, \frac{1}{\sqrt{2}}|2,-2\rangle_g - \frac{1}{\sqrt{2}}|2,2\rangle_g, |1,0\rangle_f \right\}, \tag{34b}$$

$$\mathcal{H}_3^{\mathrm{inv}} = \mathrm{span}\left\{ |1,0\rangle_e, \frac{1}{\sqrt{2}}|2,-1\rangle_g - \frac{1}{\sqrt{2}}|2,1\rangle_g, \frac{1}{\sqrt{2}}|1,-1\rangle_f + \frac{1}{\sqrt{2}}|1,1\rangle_f \right\}, \tag{34c}$$

$$\mathcal{H}_4^{\mathrm{inv}} = \mathrm{span}\left\{ \frac{1}{2\sqrt{2}}|2,-2\rangle_g + \frac{\sqrt{3}}{2}|2,0\rangle_g + \frac{1}{2\sqrt{2}}|2,2\rangle_g \right\}, \tag{34d}$$

$$\mathcal{H}_5^{\mathrm{inv}} = \mathrm{span}\left\{ \frac{1}{\sqrt{2}}|2,-1\rangle_g + \frac{1}{\sqrt{2}}|2,1\rangle_g \right\}, \tag{34e}$$

$$\mathcal{H}_6^{\mathrm{inv}} = \mathrm{span}\left\{ \frac{1}{\sqrt{2}}|1,-1\rangle_f - \frac{1}{\sqrt{2}}|1,1\rangle_f \right\}. \tag{34f}$$

Subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, $\kappa \in \{2,3\}$, do not contain dark states, therefore it is possible to obtain complete population transfer from $\mathcal{H}_{g,\kappa'}^{\mathrm{inv}}$ to $\mathcal{H}_{f,\kappa''}^{\mathrm{inv}}$ for pairs $(\kappa',\kappa'') \in \{(2,1),(3,2)\}$. Note that the complete population transfer requires the starting states to be particular coherent superpositions. Conversely, the subspace $\mathcal{H}_1^{\mathrm{inv}}$ contains the dark state from $\mathcal{H}_{ef}^d$, so that the population transfers exclusively to the excited level, not to the final. The subspaces $\mathcal{H}_{f,4}^{\mathrm{inv}}$ and $\mathcal{H}_{f,5}^{\mathrm{inv}}$ ($\mathcal{H}_{f,6}^{\mathrm{inv}}$) are dark for transition from ground (final) to excited level and retain the initial population during the SCRAP process. Previous results can be illustrated by plotting the adiabatic energies corresponding to above-mentioned invariant subspaces. Figure 3 shows time dependence of the pump, Stokes and Stark pulse envelopes (topmost part) and adiabatic and diabatic energies versus time (lower parts). First plot of energies is related to $\mathcal{H}_1^{\mathrm{inv}}$ and shows that the population adiabatically transfers to the excited level. Second and third energy plots correspond to invariant subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, $\kappa \in \{2,3\}$. It is obvious that starting from appropriate ground state, all the population transfers into the related final state. Similar to the two-level case in Sec. II, the total population transfer requires that the initial ground state is prepared into the specific coherent superpositions. Different situation is shown in the three lowest plots where the population rests within ground (final) level subspaces $\mathcal{H}_{g,4}^{\mathrm{inv}}$ and $\mathcal{H}_{g,5}^{\mathrm{inv}}$ ($\mathcal{H}_{f,3}^{\mathrm{inv}}$), respectively.

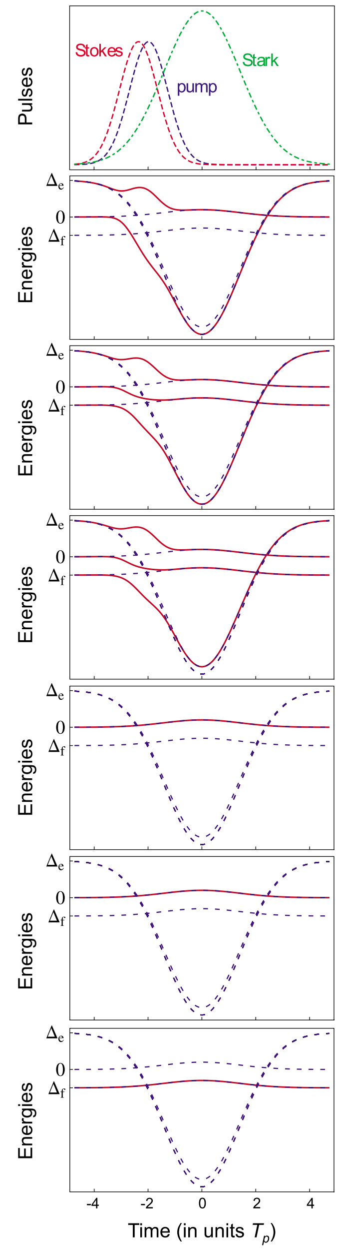

FIG. 3. (Color online) SCRAP in ^{87}Rb among hyperfine levels $5S_{1/2}$, F_g=2 and $5S_{1/2}$, F_f=1 via $5P_{1/2}$, F_e=1. Topmost: time dependence of the Stokes, pump, and Stark pulse envelopes (arbitrary scaled). Other: adiabatic (solid lines) and diabatic (dashed lines) energies versus time, related to the invariant subspaces $\mathcal{H}_\kappa^{\mathrm{inv}}$, κ =1–6 (top—bottom). The dashed line starting from energy 0 (Δ_f) corresponds to the degenerate g (f) states. The two dashed lines originating from Δ_e correspond to the states $|1,0\rangle_e$ (smaller shift) and $|1,\pm 1\rangle_e$ (larger shift). Used parameters are Δ_e=100/T_p, Δ_f =−1/2Δ_e, S_0=400/T_p, Ω_0=4/5S_0, s_g=s_f=1/20, s_e=1/20.

IV. CONCLUSION

We have presented a general formalism for describing Stark-chirped rapid adiabatic passage among degenerate-level manifolds. Cases of two and three degenerate manifolds are considered. Analysis of a degenerate-level system is facilitated by its subdivision into a set of smaller independently evolving subsystems that are related to the minimal-sized invariant subspaces of the Hamiltonian. The evolution is restricted within such invariant subspaces enabling separate analysis of each subsystem. Population transfer from the ground to the final level is considered for different types of invariant subspaces depending on the presence of dark states. The complete transfer is feasible if the initial state is prepared into specific coherent superpositions. Our formalism is applicable to the general case of arbitrary numbers of degenerate states within each level and arbitrary couplings of the appropriate transitions. It represents a generalization of Morris-Shore transformation to the case when the removed degeneracy of the sublevels leads to detuning from two-photon resonance. Applying the general formalism, we examined SCRAP among two and three hyperfine levels in the rubidium atom. The formalism gives a full description of the SCRAP population transfer process and should be useful for analyzing adiabatic passage in a wide variety of atomic and molecular systems.

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