arXiv:quant-ph/9811007 3 Nov 1998

Coherent population transfer beyond the adiabatic limit: generalized matched pulses and higher-order trapping states

M. Fleischhauer[1], R. Unanyan[2,3], B.W. Shore[2,4], and K. Bergmann[2]
[1] *Sektion Physik, Ludwig-Maximilians Universität München, D-80333 München, Germany*
[2] *Fachbereich Physik, Universität Kaiserslautern, D-67663 Kaiserslautern, Germany*
[3] *Inst. for Physical Research of the Armenian Nat. Academy of Sciences, Ashtarak-2, 378410, Armenia*
[4] *Lawrence Livermore National Laboratory, Livermore CA 94550, USA*

(April 17, 2001)

We show that the physical mechanism of population transfer in a 3-level system with a closed loop of coherent couplings (loop-STIRAP) is not equivalent to an adiabatic rotation of the dark-state of the Hamiltonian but coresponds to a rotation of a *higher-order trapping state* in a generalized adiabatic basis. The concept of generalized adiabatic basis sets is used as a constructive tool to design pulse sequences for stimulated Raman adiabatic passage (STIRAP) which give maximum population transfer also under conditions when the usual condition of adiabaticty is only poorly fulfilled. Under certain conditions for the pulses (generalized matched pulses) there exists a higher-order trapping state, which is an exact constant of motion and analytic solutions for the atomic dynamics can be derived.

I. INTRODUCTION

The transfer of population in a multi-level atomic system from an initial to a target quantum state in a fast and effective way is currently a problem of practical importance as well as of substantial theoretical interest. If there is a dipole allowed transition between an initial and a target state, one can achieve the desired transfer by using either a constant-frequency π-pulse tuned to resonance, or an adiabatic process based on a swept carrier-frequency. Since a dipole-allowed transition implies radiative decay, one is however often interested in systems with two metastable states without a direct electric-dipole coupling. Whereas an extension of the two-state π-pulse approach to multistate excitation is possible, these techniques require careful control of the pulse areas. Adiabatic processes do not require such precise control, if the time-evolution is slow (meaning, generally, large pulse areas). In a three-state Raman-transition system, for example, it is possible to achieve adiabatic passage with the use of two constant-frequency pulses suitably delayed (counterintuitive order) [1]. The process of this stimulated Raman adiabatic passage (STIRAP) [2,3] can be represented by a slow rotation of a decoupled eigenstate of the H amiltonian (dark state) [4].

The disadvantage of STIRAP is the requirement for large pulse areas: to ensure adiabatic time evolution the effective average Rabi-frequency of the pulses must be large compared to the radiative decay rates of the intermediate level(s). Non-adiabatic corrections and the associated diabatic losses [5–7], scale with $1/\Omega T$ where $\hbar\Omega$ is a characteristic interaction energy and T is the effective time required for the transfer. In some potential applications, as for example the transfer of information in form of coherences [8], it is desirable to minimize these losses without the need of intense pulses or long transfer times. Intense fields induce time-varying ac-Stark shifts, which may be detrimental to the coherence transfer. Short times are required to minimize the effect of decoherence processes during the transfer [9].

An approach, which reduces non-adiabatic losses for pulses of moderate fluence in a three-state system, was recently introduced in Ref. [10]. In addition to the pair of Raman pulses ("pump pulse" and "Stokes pulse") which couple the initial and target state via a common upper level, a direct coupling (called "detuning pulse") between them is introduced. This scheme of loop-STIRAP does not require the usual adiabaticity conditions (of large pulse areas), nor is it of the π-pulse type (requiring specific pulse areas). Nevertheless, the scheme can produce complete population transfer.

In the present paper we show that the physical mechanism of loop-STIRAP is not an adiabatic rotation of the dark state, but the rotation of a *higher-order trapping state* in a generalized adiabatic basis. The concept of generalized adiabatic basis sets allows to rationalize many examples of population transfer even when the adiabaticity condition is poorly fulfilled. If pump and Stokes pulses fulfill certain conditions (they are then called generalized matched pulses), a higher-order trapping exists, which is an exact constant of motion. In this case analytic solutions for the atomic dynamics can be found which in contrast to the case of ordinary matched pulses with identical pulse shape [12] also include the possibility of population transfer. This can be exploited to design pulse sequences which give maximum population transfer. In contrast to techniques based on optimum control theory, which are used for such tasks, the generalized-dark-state concept provides a physical interpretation of the results. However, the design of pulse, which in some cases can lead to complete population transfer (i.e. without *any* diabatic losses) needs to respect more restrictive requirements for specific pulse properties similar to π-pulse techniques.

Our paper is organized as follows. In Sec.II we discuss the loop-STIRAP and propose a simple physical interpretation in terms of an adiabatic rotation of a generalized trapping state. In Sec.III we define generalized trapping states via an iterative partial diagonalization of the time-dependent Hamiltonian. In Sec.IV we derive conditions under which a higher-order trapping state is an exact constant of motion and thus allow for an analytic solution of the atomic dynamics. Finally, various examples of population and coherence transfer based on generalized trapping states are discussed in Sec.V.

II. LOOP-STIRAP

To set the stage we consider in the present section a three-state system driven by coherent fields in a loop configuration, as shown in Fig. 1. The bare atomic states ψ_1 and ψ_3 are coupled by a resonant Raman transition via the excited atomic state ψ_2 by a pump pulse and a Stokes pulse, having Rabi-frequencies $P(t)$ and $S(t)$, respectively, which are in general complex. In addition there is a direct coupling between states 1 and 3 by a coherent detuning pulse described by the (complex) Rabi-frequency $D(t)$. Before the application of the pulses the system is in state 1 and the goal is to transfer all population into the target state 3 by an appropriate sequence of pulses. For simplicity we assume that the carrier frequencies of the pulses coincide with the atomic transition frequencies and that the phases of the pulses are time-independent. Since the phases of pump and Stokes fields can be included into the definition of the bare atomic states ψ_1 and ψ_3, they can be set equal to zero without loss of generality. The phase of the detuning pulse is relevant and cannot be eliminated. The time-dependent Schrödinger equation for this system, in the usual rotating wave approximation, reads

$$\frac{d}{dt}\mathbf{C}(t) = -i\,\mathsf{W}(t)\mathbf{C}(t) \tag{1}$$

where $\mathbf{C}(t)$ is the column vector of probability amplitudes $C_n(t) = \langle n|\psi(t)\rangle$, $(|n\rangle \in \{\psi_1, \psi_2, \psi_3\})$. The evolution matrix $\mathsf{W}(t)$ has the form

$$\mathsf{W}(t) = \frac{1}{2}\begin{bmatrix} 0 & P(t) & D(t) \\ P(t) & 0 & S(t) \\ D^*(t) & S(t) & 0 \end{bmatrix}. \tag{2}$$

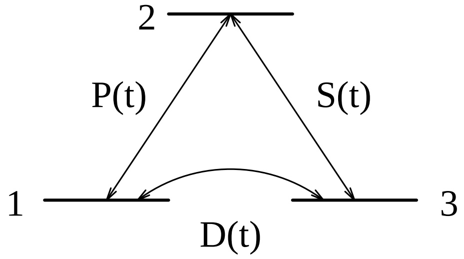

FIG. 1. Three-state system with loop linkage. $P(t)$, $S(t)$, $D(t)$ denote Rabi-frequencies of pump, Stokes and detuning pulse.

It is well known that the counterintuitive pulse sequence (Stokes puls precedes pump pulse, without a detuning pulse) leads to an almost complete population transfer, if the adiabaticity condition $\Omega T \gg 1$ is fulfilled. Here T is the characteristic time for the transfer, given by the interval where $S(t)$ and $P(t)$ overlap, and Ω the effective total Rabi-frequency averaged over the interval T

$$\Omega = \frac{1}{T}\int_{-\infty}^{\infty} dt \sqrt{P(t)^2 + S(t)^2}. \tag{3}$$

As shown in [10] an almost perfect transfer is also possible when pump and Stokes alone do not fulfill the adiabaticity condition by applying an additional detuning pulse. Fig. 2 illustrates an example of ramped pump and Stokes pulses intersected by a hyperbolic-secant detuning pulse,

$$P(t) = A_P \,\sin\left[\frac{1}{2}\arctan\left(t/T_P\right) + \frac{\pi}{4}\right], \tag{4}$$

$$S(t) = A_S \,\cos\left[\frac{1}{2}\arctan\left(t/T_S\right) + \frac{\pi}{4}\right], \tag{5}$$

$$D(t) = A_D \,\mathrm{sech}\left[t/T_D\right]. \tag{6}$$

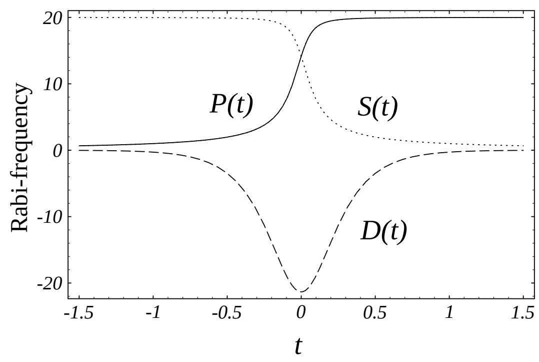

FIG. 2. Pair of ramped pump (line) and Stokes (dotted) pulses with $A_P = A_S = 20$ and $T_P = T_S = 0.1$ applied in counterintuitive order (Stokes precedes pump) with additional hyperbolic secant detuning pulse (dashed) with $A_D = -13.4i$ and $T_D = 0.2$

Fig. 3 shows examples of population histories for these pulses. When only the pump and Stokes pulses are present, the population transfer is rather poor, since the pulse areas are small ($\Omega T \sim |A_P|T_P = |A_S|T_S = 2$). As can be seen from the upper part of Fig.3, only about 70% of the initial population ends up in state 3.

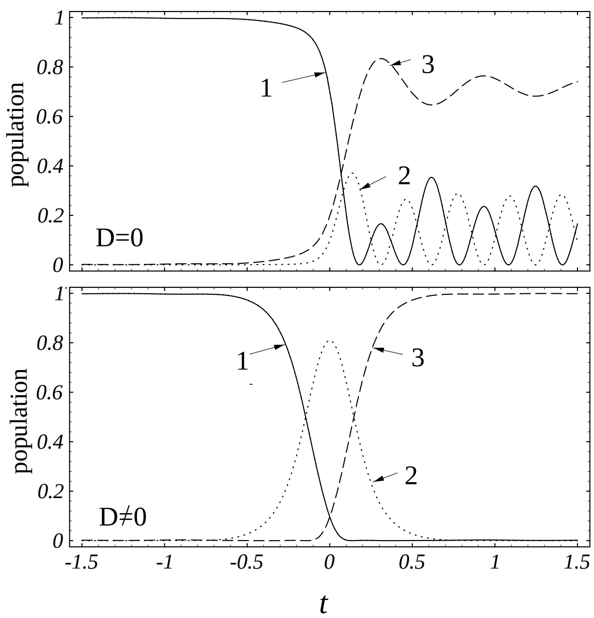

FIG. 3. Populations of states ψ_1 (line), ψ_2 (dotted) and ψ_3 (dashed) for pulse sequence of Fig.2. The upper picture shows population when only pump and Stokes pulses are applied, and the lower one if the detuning pulse is added.

The situation is remarkably different when a detuning pulse with $|A_D|T_D \approx 2.7$ and a phase factor of $e^{-i\pi/2}$ is applied; see the lower part of Fig.3. With a detuning pulse present all the population is transfered from the initial to the target state. This result is relatively insensitive to changes in the amplitude (or the shape of the detuning pulse) if the phase is $-\pi/2$.

We note that in contrast to ordinary STIRAP there is (for a short time) a substantial intermediate population of state 2. This indicates that the transfer does not occur as adiabatic rotation of the dark state from ψ_1 to ψ_3.

For our present discussion it is useful to describe ordinary STIRAP in terms of the following set of adiabatic superposition states

$$\begin{bmatrix} \Phi_1(t) \\ \Phi_2(t) \\ \Phi_3(t) \end{bmatrix} = \mathsf{U}(t)^* \begin{bmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \end{bmatrix} \tag{7}$$

with the unitary matrix

$$\mathsf{U}(t) = \begin{bmatrix} 0 & 1 & 0 \\ \sin\theta_0(t) & 0 & \cos\theta_0(t) \\ i\cos\theta_0(t) & 0 & -i\sin\theta_0(t) \end{bmatrix}. \tag{8}$$

The dynamical angle θ_0 is defined by

$$\tan\theta_0(t) = \frac{P(t)}{S(t)}. \tag{9}$$

The vector of probability amplitudes in the bare atomic basis $\mathbf{C}(t)$ and a corresponding vector $\mathbf{B}(t)$ in the superposition basis (7) are related through the transformation

$$\mathbf{B}(t) = \mathsf{U}(t)\mathbf{C}(t). \tag{10}$$

Since $\mathsf{U}(t)$ is time-dependent, the transformed evolution matrix has the form

$$\mathsf{W}(t) \to \widetilde{\mathsf{W}}(t) = \mathsf{U}(t)\mathsf{W}(t)\mathsf{U}(t)^{-1} + i\,\dot{\mathsf{U}}(t)\mathsf{U}(t)^{-1}. \tag{11}$$

In the adiabatic limit, the second term can be disregarded and we are left with the first one, which for ordinary STIRAP, i.e. without the detuning pulse, reads

$$\mathsf{U}(t)\mathsf{W}(t)\mathsf{U}(t)^{-1} = \frac{1}{2}\begin{bmatrix} 0 & \Omega(t) & 0 \\ \Omega(t) & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \tag{12}$$

where $\Omega(t) = \sqrt{P(t)^2 + S(t)^2}$. One recognizes that the superposition state $\Phi_3(t)$ is decoupled from the coherent interaction in this limit. Moreover, because $\Phi_3(t)$ does not contain the excited atomic state ψ_2, it does not spontaneously radiate and is therefore called a dark state [4]. For a counterintuitive sequence of pulses the angle $\theta_0(t)$ vanishes initially and approaches $\pi/2$ for $t \to \infty$. Thus $\Phi_3(t)$ asymptotically coincides with the initial and target states for $t \to \pm\infty$ respectively. Therefore ordinary STIRAP can be understood as a rotation of the adiabatic dark state $\Phi_3(t)$ from the initial to the target bare atomic state [3]. Non-adiabatic corrections are contained in the second contribution to $\widetilde{\mathsf{W}}(t)$

$$i\,\dot{\mathsf{U}}(t)\mathsf{U}(t)^{-1} = \frac{1}{2}\begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 2\dot{\theta}_0(t) \\ 0 & 2\dot{\theta}_0(t) & 0 \end{bmatrix}. \tag{13}$$

They give rise to a coupling between the dark state $\Phi_3(t)$ and the so-called bright state $\Phi_2(t)$.

Let us now apply the same transformation to the loop-STIRAP system, i.e. including the detuning pulse. We find:

$$\widetilde{\mathsf{W}}(t) = \frac{1}{2}\begin{bmatrix} 0 & \Omega(t) & 0 \\ \Omega(t) & \mathrm{Re}\big[D(t)\big]\sin 2\theta_0(t) & 2\dot{\theta}_0(t) + i\big[D(t)\sin^2\theta_0(t) - D^*(t)\cos^2\theta_0(t)\big] \\ 0 & 2\dot{\theta}_0(t) - i\big[D^*(t)\sin^2\theta_0(t) - D(t)\cos^2\theta_0(t)\big] & -\mathrm{Re}\big[D(t)\big]\sin 2\theta_0(t) \end{bmatrix}. \tag{14}$$

If $D(t)$ is real or complex but not strictly imaginary, there is a time dependent energy shift of the superposition states $\Phi_2(t)$ and $\Phi_3(t)$ and the detuning pulse adds an imaginary part to the nonadiabatic coupling. If $D(t)$ is imaginary, as in the example discussed above, there is no detuning but a *real* contribution to the nonadibatic coupling. Let us now assume an imaginary detuning pulse, i.e. $D(t) = i\widetilde{D}(t)$, with $\widetilde{D}(t)$ being real. In this case the transformed evolution matrix simplifies to

$$\widetilde{\mathsf{W}}(t) = \frac{1}{2}\begin{bmatrix} 0 & \Omega(t) & 0 \\ \Omega(t) & 0 & 2\dot{\theta}_0(t) - \widetilde{D}(t) \\ 0 & 2\dot{\theta}_0(t) - \widetilde{D}(t) & 0 \end{bmatrix}. \tag{15}$$

If the amplitude of the detuning pulse matches the non-adiabatic term, i.e. if $\widetilde{D}(t) = 2\dot{\theta}_0(t)$, the dark state Φ_3 is exactly decoupled even if the adiabaticity condition for pump and Stokes alone ($\Omega(t)$ being much larger than $\dot{\theta}_0(t)$) is not fulfilled. However, since $\theta_0(t)$ rotates from 0 to $\pi/2$, the detuning pulse would have to be exactly a π-pulse in such a case.

$$\int_{-\infty}^{\infty} dt\, \widetilde{D}(t) = \int_{-\infty}^{\infty} dt\, 2\dot{\theta}_0(t) = 2\theta_0(t)\Big|_{-\infty}^{+\infty} = \pi \tag{16}$$

Furthermore no pump or Stokes pulses were required for population transfer to begin with, since at any time the entire population is kept in the dark state by the action of the detuning pulse and thus pump and Stokes would not interact with the atoms. This is consistent with the observation that an exactly decoupled state Φ_3 implies exactly vanishing (not only adiabatically small!) probability amplitude of the excited bare state ψ_2 for all times. Since the origin of population transfer in this case is the well-known phenomenon of π-pulse coupling, which requires a careful control of the area and the shape of the detuning pulse, the case $\widetilde{D}(t) = 2\dot{\theta}_0(t)$ is of no further interest here.

On the other hand, if $\widetilde{D}(t)$ is negative, as in the example of Fig.2, the non-adiabatic coupling is effectively increased by the detuning pulse (note that $d\theta_0(t)/dt > 0$). Thus the success of population transfer in Fig.3 cannot be understood as dark-state rotation. This is illustrated in Fig.4, which shows the populations of the superposition states $\Phi_1 = \psi_2$, Φ_2, and Φ_3 for the above example. One clearly sees that about 80% of the population is driven out of the dark state during the interaction.

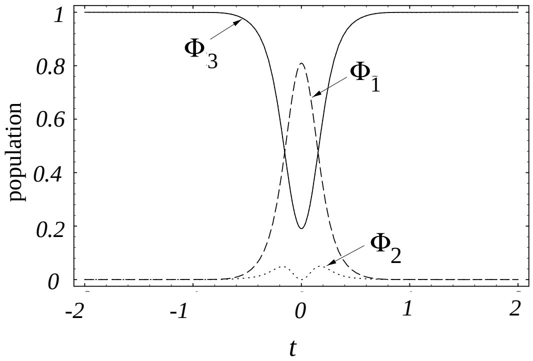

FIG. 4. Population of superposition states Φ_1 (dashed), Φ_2 (dotted), and the dark state Φ_3 (line). Parameters are that of Fig.2

It is worth noting, however, that Φ_2 remains almost unpopulated during the interaction and all population exchange happens between states Φ_1 and Φ_3. This suggests an interpretation of the process as *adiabatic population return between the superposition states* Φ_1 and Φ_3. In fact comparing the dressed-state evolution matrix $\widetilde{\mathsf{W}}(t)$, Eq.(15), with the bare-state evolution matrix $\mathsf{W}(t)$, Eq.(2) (without detuning pulse), one recognizes a formal agreement with the correspondence $P(t) \leftrightarrow \Omega(t)$ and $S(t) \leftrightarrow 2\dot{\theta}_0(t) - \widetilde{D}(t)$. That is there exists a *generalized trapping state* which is a superposition of the states Φ_1 and Φ_3. Since here $\Omega(t) = \text{const.}$ and $2\dot{\theta}_0(t) - \widetilde{D}(t)$ vanishes in the asymptotic limits $t \to \pm\infty$, this generalized trapping state coincides with Φ_3 for $t \to \pm\infty$, which in turn coincides with ψ_1 and ψ_3 in the respective limits.

To quantify this statement let us introduce a basis of *second-order adiabatic states*. Using now the first-order states Φ_1, Φ_2, and Φ_3 as a basis set instead of the bare atomic states, we introduce in analogy to Eq.(7)

$$\begin{bmatrix} \Phi_1^{(2)}(t) \\ \Phi_2^{(2)}(t) \\ \Phi_3^{(2)}(t) \end{bmatrix} = \mathsf{U}_1(t)^* \begin{bmatrix} \Phi_1(t) \\ \Phi_2(t) \\ \Phi_3(t) \end{bmatrix} = \mathsf{U}_1(t)^* \cdot \mathsf{U}(t)^* \begin{bmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \end{bmatrix}. \tag{17}$$

The unitary transformation matrix is given by

$$\mathsf{U}_1(t) = \begin{bmatrix} 0 & 1 & 0 \\ \sin\theta_1(t) & 0 & \cos\theta_1(t) \\ i\cos\theta_1(t) & 0 & -i\sin\theta_1(t) \end{bmatrix}. \tag{18}$$

which has the same form as $\mathsf{U}(t)$, Eq.(2) but here the dynamical angle $\theta_1(t)$ is defined by

$$\tan\theta_1(t) = \frac{\Omega(t)}{2\dot{\theta}_0(t) - \widetilde{D}(t)}. \tag{19}$$

Denoting the vector of probability amplitudes in these generalized adiabatic states by $\mathbf{B}^{(2)}(t)$ we find the relation

$$\mathbf{B}^{(2)}(t) = \mathsf{U}_1(t)\mathbf{B}(t). \tag{20}$$

One easily verifies that for the above example more than 95% of the population remains in the generalized trapping state $\Phi_3^{(2)}(t)$. Thus the success of the population transfer in loop STIRAP can be understood as a rotation of the second-order decoupled state $\Phi_3^{(2)}(t)$ – which is an approximate constant of motion – from the initial to the target bare atomic state.

III. GENERALIZED ADIABATIC BASIS AND GENERALIZED TRAPPING STATES FOR STIRAP

We now return to the case of ordinary STIRAP, i.e. without a detuning pulse D. The formal equivalence of $\mathsf{W}(t)$ and $\widetilde{\mathsf{W}}(t)$ suggest an iteration of the procedure introduced in the last section. We define an nth order generalized adiabatic basis by the iteration:

$$\begin{bmatrix} \Phi_1^{(n)}(t) \\ \Phi_2^{(n)}(t) \\ \Phi_3^{(n)}(t) \end{bmatrix} = \mathsf{U}_{n-1}(t)^* \begin{bmatrix} \Phi_1^{(n-1)}(t) \\ \Phi_2^{(n-1)(t)} \\ \Phi_3^{(n-1)}(t) \end{bmatrix} = \mathsf{U}_{n-1}(t)^* \cdot \mathsf{U}_{n-2}(t)^* \cdots \mathsf{U}_0(t)^* \begin{bmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \end{bmatrix}. \tag{21}$$

Correspondingly we obtain for the vector of probability amplitudes in the nth order basis

$$\mathbf{B}^{(n)} = \mathsf{U}_{n-1}\mathbf{B}^{(n-1)} = \mathsf{U}_{n-1} \cdot \mathsf{U}_{n-2} \cdots \mathsf{U}_0 \mathbf{C} \equiv \mathsf{V}_n \mathbf{C} \tag{22}$$

where we have dropped the time dependence. The nth order transformation matrix is defined as

$$\mathsf{U}_n(t) \equiv \begin{bmatrix} 0 & 1 & 0 \\ \sin\theta_n(t) & 0 & \cos\theta_n(t) \\ i\cos\theta_n(t) & 0 & -i\sin\theta_n(t) \end{bmatrix}, \tag{23}$$

with

$$\sin\theta_0(t) = \frac{P(t)}{\Omega_0(t)}, \qquad \cos\theta_0(t) = \frac{S(t)}{\Omega_0(t)}, \qquad \Omega_0(t) = \sqrt{P(t)^2 + S(t)^2}, \tag{24}$$

$$\sin\theta_n(t) = \frac{\Omega_{n-1}(t)}{\Omega_n(t)}, \qquad \cos\theta_n(t) = \frac{2\dot{\theta}_{n-1}(t)}{\Omega_n(t)}, \qquad \Omega_n(t) = \sqrt{\Omega_{n-1}(t)^2 + 4\dot{\theta}_{n-1}(t)^2}. \tag{25}$$

The iteration is illustrated in Fig.5.

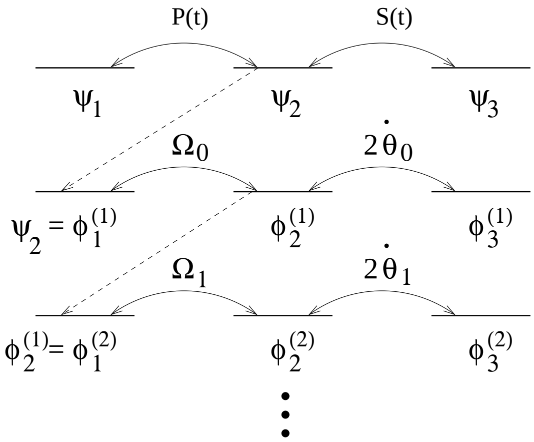

FIG. 5. Iterative definition of nth order adiabatic basis

In the nth-order basis, the equation of motion has then the form

$$\frac{d}{dt}\mathbf{B}^{(n)}(t) = -i\mathsf{W}_n(t)\mathbf{B}^{(n)}(t), \tag{26}$$

with

$$\begin{aligned} \mathsf{W}_n(t) &\equiv \frac{1}{2}\begin{bmatrix} 0 & \Omega_n(t)\sin\theta_n(t) & 0 \\ \Omega_n(t)\sin\theta_n(t) & 0 & \Omega_n(t)\cos\theta_n(t) \\ 0 & \Omega_n(t)\cos\theta_n(t) & 0 \end{bmatrix} \\ &= \frac{1}{2}\begin{bmatrix} 0 & \Omega_{n-1}(t) & 0 \\ \Omega_{n-1}(t) & 0 & 2\dot{\theta}_{n-1}(t) \\ 0 & 2\dot{\theta}_{n-1}(t) & 0 \end{bmatrix}. \end{aligned} \tag{27}$$

If $\cos\theta_k(t)$ vanishes, which implies that θ_{k-1} is time-independent, the state $\Phi_3^{(k)}$ decouples from the interaction. In this case exact analytic solutions of the atomic dynamics can be found as discussed in the next section. The analytic solutions also include cases of population or coherence transfer. If $\cos\theta_k(t)$ does not vanish but is small, the corresponding coupling in the evolution matrix can be treated perturbatively. In such a situation we have a *generalized adiabatic dynamics*.

In conclusion of this section it should be noted, that the iterative definition of a generalized adiabatic basis is conceptually very similar to the superadiabatic approach of Berry [11] introduced for two-level systems.

IV. GENERALIZED MATCHED PULSES AND ANALYTIC SOLUTION OF ATOMIC DYNAMICS

If a dynamical angle θ_{n-1} is a constant, the time-dependent state $\Phi_3^{(n)}(t)$ is decoupled from the interaction (constant of motion). In this case the dynamical problem reduces to that of a two-state system interacting via a *real* resonant coherent coupling plus a decoupled state.

$$\frac{d}{dt}\begin{bmatrix} B_1^{(n)}(t) \\ B_2^{(n)}(t) \\ B_3^{(n)}(t) \end{bmatrix} = -\frac{i}{2}\begin{bmatrix} 0 & \Omega_{n-1}(t) & 0 \\ \Omega_{n-1}(t) & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}\begin{bmatrix} B_1^{(n)}(t) \\ B_2^{(n)}(t) \\ B_3^{(n)}(t) \end{bmatrix} \tag{28}$$

This equation can immediately be solved

$$B_1^{(n)}(t) = B_1^{(n)}(0)\,\cos\phi(t) - iB_2^{(n)}(0)\,\sin\phi(t), \tag{29}$$

$$B_2^{(n)}(t) = B_2^{(n)}(0)\,\cos\phi(t) - iB_1^{(n)}(0)\,\sin\phi(t), \tag{30}$$

$$B_3^{(n)}(t) = B_3^{(n)}(0), \tag{31}$$

where

$$\phi(t) = \frac{1}{2}\int_0^t d\tau\, \Omega_{n-1}(\tau). \tag{32}$$

In particular if the atom is initially in the trapping state, it will stay in that state.

For example if θ_0 does not depend on time, the usual dark state $\Phi_3^{(1)}$ is an exact constant of motion. As can be seen from Eq.(9), for θ_0 to be time-independent, Stokes and pump need to be either cw fields or need to have the same envelope function, i.e. have to be *matched pulses* [12],

$$S(t) = \Omega_0(t)\cos\theta_0, \tag{33}$$

$$P(t) = \Omega_0(t)\sin\theta_0, \tag{34}$$

where $\Omega_0(t)$ can be an arbitrary function of time and θ_0 =const. The atomic dynamics is trivial in this case. Since $\Phi_3^{(1)}$ is time-independent, the trapping state is a constant superposition of the bare atomic states 1 and 3.

On the other hand, if some higher-order dynamical angle θ_n is constant, the system remains in a generalized trapping state if initially prepared in it. The projection of this state onto the bare atomic basis is in general time-dependent, and one can have a substantial rearrangement of atomic level population including population transfer. If a higher-order dynamical angle is constant we will call pump and Stokes pulses *generalized matched pulses*.

To obtain an explicit condition for generalized matched pulses in terms of $P(t)$ and $S(t)$ we successively integrate relations (25). This leads to the iteration

$$\begin{aligned}\theta_{k-1}(t) &= \frac{1}{2}\int_{-\infty}^t dt'\,\Omega_k(t')\cos\theta_k(t') + \theta_k^0,\\ \Omega_{k-1}(t) &= \Omega_k(t)\sin\theta_k(t),\end{aligned} \tag{35}$$

starting with some $\theta_n(t) = \theta_n$ = const. and $\Omega_n(t)$ as an arbitrary function of time. Each iteration leads to one constant θ_k^0, which can be freely chosen. The application of generalized matched pulses to coherent population transfer will be discussed in the next section.

As noted before there may be cases, where for some number n the dynamical angle $\theta_n(t)$ does depend on time but its time-derivative is much smaller than the corresponding generalized Rabi-frequency $\Omega_n(t)$, while the same is not true for all $k < n$. In this case the state $\Phi_3^{(n)}(t)$ is an approximate constant of motion and we have an nth order adiabatic process. The example of loop-STIRAP discussed in the last section is a realization of a higher-order adiabatic process, which is non-adiabatic in the first-order basis.

V. APPLICATION OF GENERALIZED MATCHED PULSES TO POPULATION- AND COHERENCE TRANSFER

In the following we discuss several examples for a coherent transfer of population from one non-decaying state to the other or to the excited state using generalized matched pulses. We furthermore discuss the possibility to transfer coherence, for example from the ground state transition to an optical transition. Since in all cases there exist a generalized trapping state which is an exact constant of motion, we can obtain exact analytic results for the atomic dynamics.

A. Population and coherence transfer with second-order generalized matched pulses

1. Complete transfer of coherence from a ground-state doublet to an optical transition

First we discuss the case when $\Phi_3^{(2)}$ is an exact constant of motion, i.e. a trapping state. Furthermore we assume that the state vector Ψ coincides with this trapping state at $t = -\infty$. Then the system will remain in the trapping state at later times. Therefore θ_1 =const. and it is clear from Fig.5 that Ψ is a time independent superposition of states $\Phi_1^{(1)}$ and $\Phi_3^{(1)}$ and thus has at all times a constant probability amplitude of the bare atomic state 2. In fact from

$$\mathbf{C} = \mathsf{V}_2^{-1}\mathbf{B}^{(2)} = \mathsf{V}_2^{-1}\begin{bmatrix}0\\0\\1\end{bmatrix} \tag{36}$$

we find

$$\begin{bmatrix} C_1(t) \\ C_2(t) \\ C_3(t) \end{bmatrix} = -i\cos\theta_1 \begin{bmatrix} i\tan\theta_1\ \cos\theta_0(t) \\ 1 \\ -i\tan\theta_1\ \sin\theta_0(t) \end{bmatrix}. \tag{37}$$

We now identify state 2 with a lower i.e. non-decaying level and state 3 with an excited states. The pump pulse $P(t)$ then couples two ground states which could be realized for example by a magnetic coupling. The Stokes pulse, which couples states 2 and 3 is considered an optical pulse. Due to the finite and constant admixture of state 2 to the trapping state, second-order generalized matched pulses are best suited to transfer coherence for example from the 1-2 transition to the 3-2 transition.

We now want to construct pulses, that would lead to the desired complete coherence transfer. To achieve this we have to satisfy the initial and final conditions

$$\theta_0(-\infty) = 0, \tag{38}$$
$$\theta_0(+\infty) = \pi/2. \tag{39}$$

On the other hand, the iteration equation (35) requires for second-order matched pulses that

$$\theta_0(t) = \frac{1}{2}\int_{-\infty}^{t} dt'\,\Omega_1(t')\cos\theta_1 + \theta_0^0, \tag{40}$$
$$\Omega_0(t) = \Omega_1(t)\sin\theta_1, \tag{41}$$

where θ_1 and θ_0^0 are arbitrary constants and $\Omega_1(t)$ an arbitrary positive function of time. To fulfill the initial condition (38) we set $\theta_0^0 = 0$. In order to satisfy the final condition (39) we then have to adjust the total pulse area (see Eq.(40))

$$A_0 = \int_{-\infty}^{\infty} dt\,\Omega_0(t) = \pi\tan\theta_1. \tag{42}$$

Thus pump and Stokes pulses have the form

$$P(t) = \Omega_0(t)\sin\left[\frac{\pi A\,(t)}{2A_0}\right], \tag{43}$$
$$S(t) = \Omega_0(t)\cos\left[\frac{\pi A\,(t)}{2A_0}\right]. \tag{44}$$

with

$$A\,(t) = \int_{-\infty}^{t} dt'\,\Omega_0(t') \tag{45}$$

With this choice an initial coherent superposition of states 2 and 1

$$\Psi(-\infty) = -i\cos\theta_1\ \psi_2 + \sin\theta_1\ \psi_1 \tag{46}$$

can be completely mapped into a coherent superposition of states 2 and 3

$$\Psi(+\infty) = -i\cos\theta_1\ \psi_2 - \sin\theta_1\ \psi_3. \tag{47}$$

In order to transfer a given ground-state coherence to an optical transition the pulse area A_0 should be chosen according to (42), $A_0 = |C_1(-\infty)/C_2(-\infty)|$. The shape of $\Omega(t)$ is otherwise arbitrary. It should be noted that Eq.(46) requires a certain fixed phase of the initial coherent superposition. The phase of the pump pulse, which is included in the definition of ψ_1 (cf. Sec.II), may need adjustment to satisfy this condition.

In Fig.6 we have shown the populations of the bare atomic states for the example $\Omega_0(t) = \sqrt{\pi}\exp(-t^2)$ ($A = \pi$) and $\Psi(-\infty) = 1/\sqrt{2}(\psi_1 - i\psi_2)$ from a numerical solution of the Schrödinger equation. One clearly sees that all population from state 1 is transferred to state 3. This transfer happens without diabatic losses despite the fact that $A = \pi$ and thus the usual adiabaticity condition is only poorly fulfilled.

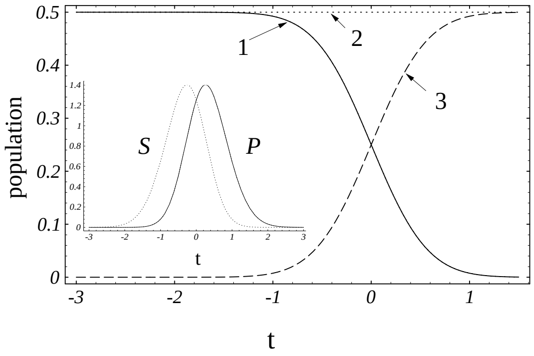

FIG. 6. Example of complete coherence transfer from $1-2$ to $3-2$ with second-order generalized matched pulses. Plotted are the populations of bare atomic states for a pair of pulses as shown in the insert. One recognizes constant population in state 2 and complete transfer of population in 1 to 3.

The process discussed here may have some interesting applications, since it allows to transfer coherence from a robust and long-lived ground state transition to an optically accessible transition.

The population transfer from 1 to 3 with finite constant state amplitude in 2 discussed here coincides with the solution found by Malinovsky and Tannor [13] with numerical optimization techniques. Assuming a finite constant amplitude in state 2, these authors numerically optimized the peak Rabi-frequency (which in this case is the only remaining free parameter) to achieve maximum population transfer. They found, that in order to maximize the final amount of population in state 3, the peak Rabi-frequency has to be larger than a certain critical value. This can very easily be verified from the generalized matched-pulse solutions (42,47).

$$|C_3(\infty)|^2 = \sin^2\theta_1 = \frac{A^2}{\pi^2+A^2} \tag{48}$$

$$|C_2(\infty)|^2 = \cos^2\theta_1 = \frac{\pi^2}{\pi^2+A^2}. \tag{49}$$

In the limit $\theta_1 \to \pi/2$, which implies $A \to \infty$, the admixture of level 2 vanishes and we essentially have population transfer from state 1 to state 3.

2. Population transfer from 1 to 3 and non-exponential diabatic losses

We have seen in the last subsection that second-order matched pulses can be used to effectively transfer population from state 1 to 3, if there is an initial admixture of the excited state. This amplitude is inversely proportional to the square of the pulse area A. Therefore one could expect a good transfer for large A also if all population is initially in state 1. In this case there is some finite amount of population which is not trapped in the generalized dark state $\Phi_3^{(2)}$. Clearly in order to achieve maximum population transfer, pump and Stokes pulse should be in counterintuitive order and hence conditions (38) and (39) should be fulfilled. Since the pulses are assumed to be second-order matched pulses, the dynamical problem with the initial condition

$$\begin{bmatrix} B_1^{(2)}(-\infty) \\ B_2^{(2)}(-\infty) \\ B_3^{(2)}(-\infty) \end{bmatrix} = \mathsf{U}_1 \cdot \mathsf{U}_0 \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix} = \begin{bmatrix} 0 \\ i\cos\theta_1 \\ \sin\theta_1 \end{bmatrix} \tag{50}$$

can easily be solved (see Eq.(29-31)). From Eqs.(25) we find $\Omega_1(t) = \Omega_0(t)/\sin\theta_1$. Thus

$$B_1^{(2)}(\infty) = \frac{\pi}{\sqrt{\pi^2+A^2}} \sin\left[\frac{1}{2}\sqrt{\pi^2+A^2}\right], \tag{51}$$

$$B_2^{(2)}(\infty) = i\frac{\pi}{\sqrt{\pi^2+A^2}} \cos\left[\frac{1}{2}\sqrt{\pi^2+A^2}\right], \tag{52}$$

$$B_3^{(2)}(\infty) = \frac{A}{\sqrt{\pi^2+A^2}}, \tag{53}$$

where A is the total pulse area defined in (42). From this we find the asymptotic populations of the bare atomic states

$$\left|C_1(\infty)\right|^2 = \frac{\pi^2}{\pi^2+A^2}\sin^2\left(\frac{1}{2}\sqrt{\pi^2+A^2}\right), \tag{54}$$

$$\left|C_2(\infty)\right|^2 = \frac{4\pi^2A^2}{\left(\pi^2+A^2\right)^2}\sin^4\left(\frac{1}{4}\sqrt{\pi^2+A^2}\right), \tag{55}$$

$$\left|C_3(\infty)\right|^2 = \frac{1}{\left(\pi^2+A^2\right)^2}\left[A^2+\pi^2\cos\left(\frac{1}{2}\sqrt{\pi^2+A^2}\right)\right]^2. \tag{56}$$

Thus the diabatic losses scale in general with $1/A^2$, i.e. non-exponentially with A. Furthermore for

$$\frac{1}{2}\sqrt{\pi^2+A^2} = 2n\pi \qquad \text{or} \qquad A = \pi\sqrt{16n^2-1} \tag{57}$$

with $n = 1, 2, \ldots$ the population transfer is complete. We show in Fig. 7 the final population in state 3 as a function of A/π.

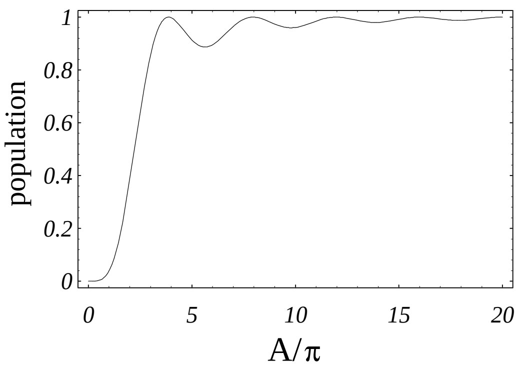

FIG. 7. Final population in state 3 as a function of total pulse area A/π for population transfer from state 1 with second-order matched pulses. For $A/\pi = \sqrt{16n^2-1}$ the transfer is complete (100.00%)

A special case of the population transfer with second-order matched pulses discussed in the present section is the analytical model discussed by Vitanov and Stenholm in [14]. These authors considered a pulse sequence with

$$\Omega_0(t) = \frac{\alpha}{2T}\text{sech}^2\left(\frac{t}{T}\right), \qquad \theta_0(t) = \frac{\pi}{4}\left[\tanh\left(\frac{t}{T}\right)+1\right] \tag{58}$$

and thus $\tan\theta_1 = \Omega_0(t)/2\dot{\theta}_0(t) = \alpha/\pi = \text{const}$.

B. Population transfer via large-area third-order matched pulses

Next we analyze the possibility of population transfer when $\Phi_3^{(3)}$ is exactly trapped. In order for $\Phi_3^{(3)}$ to be a constant of motion or equivalently to have third-order matched pulses θ_2 =const. We assume again that the system state vector Ψ is initially in the trapping state in which it will remain for all times. In order to realise population transfer from state 1 to state 2 or 3 in this case, we furthermore must satisfy the initial conditions

$$C_1(-\infty) = 1, \quad C_2(-\infty) = 0, \quad C_3(-\infty) = 0. \tag{59}$$

This can be translated into a condition for the initial values of the dynamical phases θ_0 and θ_1 using Eq.(22). In fact from

$$\mathbf{C} = \mathsf{V}_3^{-1}\mathbf{B}^{(3)} = \mathsf{V}_3^{-1}\begin{bmatrix}0\\0\\1\end{bmatrix} \tag{60}$$

we find

$$C_1(t) = i\left(\cos\theta_0(t)\sin\theta_1(t)\sin\theta_2 - \sin\theta_0(t)\cos\theta_2\right) \tag{61}$$
$$C_2(t) = -i\cos\theta_1(t)\sin\theta_2 \tag{62}$$
$$C_3(t) = -i\left(\cos\theta_0(t)\cos\theta_2 + \sin\theta_0(t)\sin\theta_1(t)\sin\theta_2\right). \tag{63}$$

The initial condition it is fulfilled when

$$\cos\theta_0\,(-\infty)\sin\theta_1\,(-\infty)\sin\theta_2 - \cos\theta_2\sin\theta_0\,(-\infty) = 1, \tag{64}$$
$$\sin\theta_2\cos\theta_1\,(-\infty) = 0, \tag{65}$$
$$\cos\theta_0\,(-\infty)\cos\theta_2 + \sin\theta_2\sin\theta_0\,(-\infty)\sin\theta_1\,(-\infty) = 0. \tag{66}$$

The result is

$$\theta_1\,(-\infty) = \frac{\pi}{2}, \quad \theta_0\,(-\infty) = \theta_2 + \frac{\pi}{2}. \tag{67}$$

From Eq.(25) we find the following differential equation

$$2\frac{d\theta_1(t)}{dt} = \alpha\,\Omega_1(t), \qquad \text{where} \qquad \alpha = (\tan\theta_2)^{-1} = \text{const}. \tag{68}$$

Introducing

$$x(t) = \tan\theta_1(t) \tag{69}$$

we find furthermore

$$\frac{2x\,\dot{x}}{(1+x^2)^{3/2}} = \alpha\Omega_0(t), \tag{70}$$
$$2\frac{d\theta_0(t)}{dt} = \frac{\Omega_0(t)}{x(t)}. \tag{71}$$

Integrating these equations and taking into account the initial conditions (67) yields

$$\tan\theta_1(t) = x(t) = \sqrt{\frac{1}{f^2(t)} - 1}, \tag{72}$$
$$\theta_0(t) = \theta_2 + \frac{\pi}{2} + \frac{1}{\alpha}\left[1 - \sqrt{1 - f^2(t)}\right], \tag{73}$$

where

$$f(t) = \frac{\alpha}{2}\int_{-\infty}^{t} dt'\,\Omega_0\,(t'). \tag{74}$$

$\Omega_0(t)$ is an arbitrary smooth function which we assume to vanish at infinity, $\Omega_0\,(\pm\infty) = 0$. We still have one free constant α, which we can choose. As we will show now, we can choose α such that the efficiency of the transfer from state 1 to states 3 or 2 approaches unity.

1. Population transfer from ground state to state 3

In order to transfer the initial population from state 1 to the target state 3, it is necessary to satisfy the final conditions

$$\theta_1\,(+\infty) = \frac{\pi}{2}, \quad \theta_0\,(+\infty) = \theta_2 \tag{75}$$

which implies

$$\tan\theta_1(\infty) = \sqrt{\frac{4}{\alpha^2 A^2} - 1} \to \infty, \tag{76}$$

$$\theta_0(\infty) = \theta_2 + \frac{\pi}{2} + \frac{1}{\alpha}\left[1 - \sqrt{1 - \alpha^2 A^2/4}\right] = \theta_2, \tag{77}$$

where

$$A = \int_{-\infty}^{\infty} dt\, \Omega_0(t) \tag{78}$$

is the pulse area. From these condition one finds the constraint

$$\alpha = -\frac{4\pi}{\pi^2 + A^2}, \qquad A \gg \pi. \tag{79}$$

The diabatic losses in the limit $A \gg 1$ are

$$1 - \left|C_3(\infty)\right|^2 \approx \frac{4\pi^2}{2\pi^2 + A^2} \tag{80}$$

and thus in the adiabatic limit we have essentially complete population transfer from state 1 to state 3.

Fig.8 shows an example of population transfer with third-order matched pulses. Here $\Omega_0(t) = A/2\,\mathrm{sech}^2(t)$ and $A = 20\pi$. Pump and Stokes pulses are shown in the upper frame and the population histories in the lower one. We see that the amplitudes of the Stokes and pump pulses are unequal. As in ordinary STIRAP the population of the state 2 is small during the evolution.

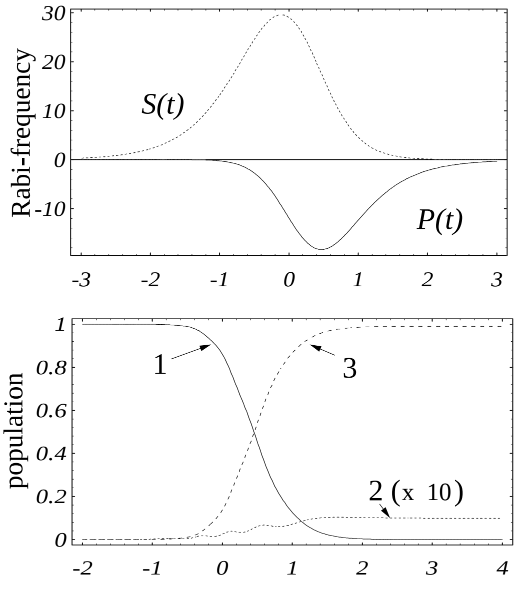

FIG. 8. Population transfer from 1 to 3 with third-order matched pulses. Upper frame shows pulses, lower frame population dynamics. Here $\Omega_0(t) = A/2\,\mathrm{sech}^2(t)$ and $A/\pi = 20$.

2. Population transfer from ground state to the state 2

In order to transfer the initial population from state 1 to state 2, it is necessary to satisfy the conditions

$$\theta_1(+\infty) = 0, \quad \theta_2 = \frac{\pi}{2} \tag{81}$$

In this case we have to fix α to be

$$\alpha = \frac{2}{A}, \qquad A \gg 1. \tag{82}$$

Fig.9 shows the pulses $P(t)$ and $S(t)$ and the evolution of the atomic populations. Here $\Omega_0(t) = A/2\,\text{sech}^2(t)$ and $A = 16$. We see that the Stokes and pump pulses are in a counterintuitive sequence. At first the atomic population oscillates between state 1 and 3, but as the pulse sequence proceeds the whole population is transfered into state 2. In other words, during the full pulse sequence there occur several STIRAP transitions, but due to the large nonadiabatic coupling the population accumulates in state 2.

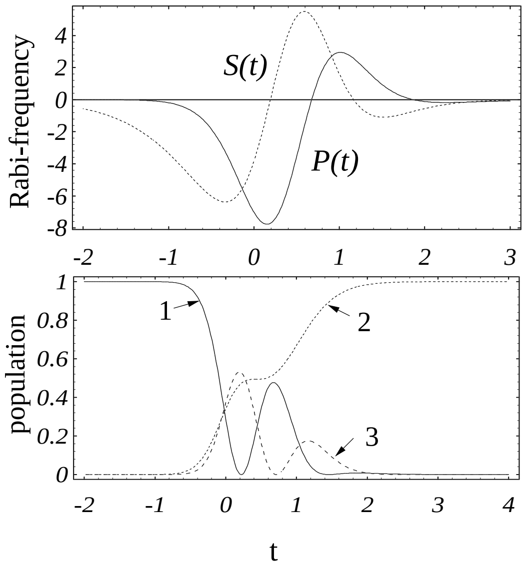

FIG. 9. Population transfer from 1 to 2 with third-order matched pulses. Upper frame shows pulses, lower frame population dynamics. Here $\Omega_0(t) = A/2\,\text{sech}^2(t)$ and $A = 16$.

VI. SUMMARY

We have introduced the concept of generalized dressed states in order to explain the success of population transfer in stimulated Raman adiabatic passage with a loop coupling. If the interaction of a three-level system with a pair of time-dependent pump and Stokes pulses is described in terms of the so-called dark and bright states instead of the instantaneous eigenstates of the Hamiltonian, the original three-state–two-field system is transformed into a system of three states coupled by two effective interactions [15,7]. This allows for an iteration procedure leading to higher-order adiabatic basis sets [11]. We showed that in the case of loop-STIRAP there is a higher-order trapping state, which is an approximate constant of motion even when the usual adiabaticity condition is not fulfilled. This state adiabatically rotates from the initial to the target quantum state of the atom and thus leads to efficient population transfer, however, at the expense of placing some population into the decaying atomic state.

The concept of generalized trapping states allows the construction of pulse sequences which lead to an optimum population or coherence transfer also for small pulse areas and allows for solutions for the atomic dynamics. If pump and Stokes pulses fulfill certain conditions (so-called generalized matched pulses) the effective 3×3 coupling matrix factorizes at a specific point of the iteration. The trapping state of the corresponding nth-order adiabatic basis is then an exact constant of motion. In this case the atomic dynamics reduces to a two-level problem with a real coupling which can be solved analytically.

For ordinary matched pulses, i.e. if pump and Stokes have the same shape, the atomic dynamics is rather limited. The corresponding dark state is a constant superposition of states 1 and 3. In the case of generalized matched pulses, however, the trapping state has a time-dependent overlap with the bare atomic states and thus population or coherence transfer is possible. We have discussed with specific example population transfer with second and third-order matched pulses. We found that for certain values of the pulse areas complete population or coherence transfer is possible. In the general case the diabatic losses scale non-exponentially with the inverse pulse area.

ACKNOWLEDGEMENTS

The work of RU is supported by the Alexander von Humboldt Foundation. BWS thanks the Alexander von Humboldt Stiftung for a Research Award; his work is supported in part under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48. Partial support by the EU Network ERB-CHR-XCT-94-0603 is also acknowledged.

---

[1] J. Oreg, F. T. Hioe, and J. H. Eberly, Phys. Rev. **A** 29, 690 (1984).

[2] U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988); for reviews on the experimental developments of STIRAP see: K. Bergmann and B. W. Shore, in *Molecular Dynamics and Spectroscopy by Stimulated Emission Pumping*, ed. by H.L. Dai and R.W. Field (World Scientific, Singapore, 1995) pp. 315-373;

[3] K. Bergmann, H. Theuer, and B. W. Shore, *"Coherent Population Transfer Among Quantum States of Atoms and Molecules"* Rev. Mod. Phys. **70**,1003 (1998).

[4] for a review on coherent population trapping see: E. Arimondo, Prog. in Optics **35**, 259 (1996).

[5] M. Elk, Phys. Rev. A **52**, 4017 (1995).

[6] T.A. Laine and S. Stenholm, Phys. Rev. A **53**, 2501 (1996).

[7] M. Fleischhauer and A.S. Manka, Phys. Rev. A **54**, 794 (1996).

[8] A.S. Parkins, P. Marte, P. Zoller, and H.J. Kimble, Phys. Rev. Lett. **71**, 3095 (1993); T. Pellizzari, S.A. Gardiner, J.I. Cirac, and P. Zoller, Phys. Rev. Lett. **75**, 3788 (1995).

[9] In adiabatic population transfer involving atoms coupled to a common cavity mode as used in [8], the transfer time needs to be much shorter than the cavity decay time.

[10] R.G. Unanyan, L.P. Yatsenko, K. Bergmann, and B.W. Shore, Opt. Comm. **139**, 48 (1997).

[11] M.V. Berry, Proc. R. Soc. A **414**, 31 (1987); *ibid* **429**, 61 (1990); R. Lim and M.V. Berry, J. Phys. A **24**, 3255 (1991).

[12] S.E. Harris, Phys. Rev. Lett. **72**, 52 (1994); J.H. Eberly, M.L. Pons, and H.R. Haq, Phys. Rev. Lett. **72**, 56 (1994).

[13] V.S. Malinovsky and D.J. Tannor, Phys. Rev. A **56**, 4929 (1997).

[14] N.V. Vitanov and S. Stenholm, Phys. Rev. A **55**, 648 (1997).

[15] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, and C. Cohen-Tannoudji, J. Opt. Soc. Am. **6**, 2112 (1989).