Stimulated Raman Adiabatic Passage via bright state in $\Lambda$ medium of unequal oscillator strengths

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We consider the population transfer process in a $\Lambda$-type atomic medium of unequal oscillator strengths by stimulated Raman adiabatic passage via bright-state ($b$-STIRAP) taking into account propagation effects. Using both analytic and numerical methods we show that the population transfer efficiency is sensitive to the ratio $q_p/q_s$ of the transition oscillator strengths. We find that the case $q_p > q_s$ is more detrimental for population transfer process as compared to the case where $q_p \leq q_s$. For this case it is possible to increase medium dimensions while permitting efficient population transfer. A criterion determining the interaction adiabaticity in the course of propagation process is found. We also show that the mixing parameter characterizing the population transfer propagates superluminally.

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I. INTRODUCTION

Many branches of contemporary physics require atoms and molecules prepared in specified quantum states which is important for recently developing research areas of atom optics and quantum information. Furthermore with the growing interest in quantum information, there is also concern with creating and controlling specified coherent superpositions of quantum states. Therefore there has long been interest in finding techniques to control the transfer of population between quantum states.

A particularly interesting technique for population transfer is stimulated Raman adiabatic passage (STIRAP) $^{1,2}$ realized via so-called "dark" (or "trapped") states. The STIRAP method is a robust and powerful tool for coherent and complete population transfer between two (or more) quantum states. It has many applications in many domains such as atom optics $^{3,4}$, chemical-reactions $^{6}$, laser-induced cooling $^{7}$, etc.

In this paper we focus our attention on the alternative method involving rather "bright" $^{8,9}$ than "dark" state. The experimental realization of the method, called $b$-STIRAP, in a Pr:YSO crystal has been reported in $^{10}$. Unlike STIRAP, which is insensitive to the radiative losses from the excited state that is not populated, $b$-STIRAP stores some transient population in the excited state. Radiative losses are therefore possible resulting in a reduction in the transfer efficiency. Thus contrary to STIRAP, $b$-STIRAP should feature a sufficiently large one-photon detuning and sufficiently short interaction time in order to permit efficient population transfer. The effect of spontaneous decay from the intermediate state inside the system for $b$-STIRAP was studied in $^{11,12}$.

Due to the process of $b$-STIRAP, A-system is fully reversible in interactions with short laser pulses of durations much shorter than the relaxation times of the system and may hence serve for implementation of all-optical reversible processor $^{14}$. Recently, the combination of STIRAP and $b$-STIRAP have been used for the experimental implementation of optical logical gates in a solid memory $^{15}$.

The growing interest for $b$-STIRAP necessitates a further investigation when the propagation effects are taken into account. Note that the propagation effects for counterintuitive sequences of pulses and population transfer via STIRAP in media have been investigated in many papers (see for example Refs. $^{16,20–23}$). One of the main results of $^{10}$ is that during pulse propagation in the STIRAP regime the interaction adiabaticity as well as the spatial evolution of propagating pulses are strongly affected by the relationship between the oscillator strengths of the corresponding atomic transitions. In a recent work $^{17}$ a detailed theoretical analysis of the $b$-STIRAP process in media with equal oscillator strengths is presented. It is shown that there is some differences between STIRAP via dark state and STIRAP via bright state. The essential difference of the $b$-STIRAP method, as compared to the STIRAP method, is that $b$-STIRAP is a faster process that is realized with a superluminal velocity. Another difference is that the adiabaticity conditions are stronger in case of the $b$-STIRAP. For example, in the case where the oscillator strengths of both transitions are equal, the interaction adiabaticity, provided at the medium entrance, is broken down for $b$-STIRAP, while for STIRAP it is preserved during propagation. In this context the natural question arises whether the population transfer efficiency in a medium via $b$-STIRAP is also sensitive to the ratio of oscillator strengths.

To clarify the question addressed, we make a detailed theoretical study of nonlinear pulse propagation in a $\Lambda$-type three-level atomic system under the conditions of the "bright" state for various ratios of the medium coupling constants. Our results show that the population transfer dynamics strongly depends on the ratio of the oscillator strengths. We find that, depending on the ra-
tio, pulses propagating in a medium will maintain their capacity to produce efficient adiabatic population transfer for long distances in some cases and loose this property in other cases.

The paper is organized as follows. Section II A recalls the underlying physics of the b-STIRAP process. In Section II B we give the theoretical model and the governing equations. Section III contains the results of numerical calculations and their interpretation. In section IV the analytic solutions for explaining the numerical results presented in Section III are provided. Finally, we summarize the results obtained.

II. THE THEORETICAL FRAMEWORK

A. Background

The b-STIRAP process is defined with respect to population transfer in a three level system, which we consider to be a ground state $|1\rangle$, an excited state $|2\rangle$, and a final state $|3\rangle$ in which we wish to maximize the population (see Fig. 1). These matter states are coupled by two laser fields: a field that is resonant with the transition from the ground to the excited state (the pump field) and a field that is resonant with the excited-to-final state transition (the Stokes field). The pump and Stokes pulses, detuned by $\Delta_{p,s}$ with respect to the corresponding resonances, have the respective Rabi frequencies $\Omega_p$ and $\Omega_s$. The dressed eigenstates of the matter-field system in the case of exact-two photon resonance are well known [18] and are given by

$$
|b_1\rangle = \cos \psi \sin \theta |1\rangle + \cos \psi \cos \theta |3\rangle + \sin \psi |2\rangle, \quad (1a)
$$
$$
|b_2\rangle = \sin \psi \sin \theta |1\rangle + \sin \psi \cos \theta |3\rangle - \cos \psi |2\rangle, \quad (1b)
$$
$$
|d\rangle = \cos \theta |1\rangle - \sin \theta |3\rangle, \quad (1c)
$$

where the time-dependent mixing angles are defined

$$
\psi(t) = \tan \rho(t) = \tan \theta(t) / \sin \theta(t), \quad \text{and} \quad \rho(t) = 2 \tan \theta(t),
$$

and $\Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}$ being the generalized Rabi frequency. We will consider without loss of generality $\Delta_p > 0$.

The central point of interest for b-STIRAP, among the dressed atomic eigenstates in Eqs. (1), is the bright state $|b_1\rangle$ which is a linear combination of all three bare states $|1\rangle$, $|2\rangle$ and $|3\rangle$. With $\Delta_p \neq 0$ and all population initially in $|1\rangle$, if the pump pulse precedes (but overlaps) the Stokes pulse, at time $t \to -\infty$ we have for the mixing angles $\theta = 90^\circ$ and $\psi = 0^\circ$, and the dressed state $|b_1\rangle$ corresponds to the bare, populated, state $|1\rangle$ (all population initially in the ground-state coincides with this particular eigenstate). At the end of the interaction, at time $t \to \infty$, where the Stokes pulse is applied after the pump pulse, we have mixing angles of $\theta = 0^\circ$ and $\psi = 0^\circ$ and the state $|b_1\rangle$ corresponds to the bare state $|3\rangle$ (all of the population projects onto the final state). If both dressing angles $\theta$ and $\psi$ are changed slowly, i.e. by ensuring an adiabatic evolution, all population remains in $|b_1\rangle$, and there is low probability that the system will make a non-adiabatic transition to another dressed state.

B. Description of the model

In the present paper we study the population transfer from state $|1\rangle$ to state $|3\rangle$ by means of b-STIRAP taking into consideration propagation effects. We consider two time-dependent laser fields propagation in a medium of three-level atoms in the lambda configuration as shown in Fig. 1. We assume that both fields propagate in the positive $x$ direction. Let

$$
E_p(x,t) = \xi_p(x,t) \cos(\omega_p t - k_p x - \varphi_p), \quad (2a)
$$
$$
E_s(x,t) = \xi_s(x,t) \cos(\omega_s t - k_s x - \varphi_s), \quad (2b)
$$

where $\xi_p$ and $\xi_s$ are the slowly varying envelopes of the electric fields of carrier frequencies $\omega_p$ and $\omega_s$, wave numbers $k_p$, $k_s$ and phases $\varphi_p$, $\varphi_s$. We assume that the temporal durations of both laser pulses are sufficiently short that we can neglect decay terms, such as arising from loss to other atomic states, spontaneous emission, or collisional dephasing effects.

The corresponding time-dependent Hamiltonian in the basis $\{|1\rangle, |2\rangle, |3\rangle\}$ in the Rotating Wave Approximation (RWA) reads [18]

$$
H = \hbar \begin{pmatrix}
0 & -\Omega_p & 0 \\
-\Omega_p & \Delta_p & -\Omega_s \\
0 & -\Omega_s & \delta
\end{pmatrix}, \quad (3)
$$

where the Rabi frequencies and the one- and two-photon detunings are defined as follows: $\Omega_{p,s} = |\xi_{p,s} d_{p,s}|/2\hbar$ with $d_{p,s}$ being the transition dipole moments, $\Delta_p = \omega_2 - \omega_1 - \omega_p + \phi_p$, and $\delta = \omega_3 - \omega_1 - \omega_p + \omega_s + \phi_p - \phi_s$,

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{The three-level Λ-type system coupled by two near resonant pulses with Rabi frequencies $\Omega_p$ and $\Omega_s$.}
\end{figure}
achieved by switching on the pulses in the intuitive order in the course of the evolution:

\[
\Delta p_a + \Delta p_a - \Omega_s a_3, \quad \frac{\partial p_2}{\partial t} = -\Omega p_1, \quad \frac{\partial a_3}{\partial t} = -\Delta a_2 + \delta a_3.
\]

All atoms are assumed to be initially in the ground state \( |1\rangle: a_1(\infty, x) = 1, a_2(\infty, x) = a_3(\infty, x) = 0 \).

The propagation of the pulses is governed by the Maxwell wave equations which in the slowly varying envelope approximation can be reduced to two independent first-order wave equations for each individual pulse that in terms of traveling coordinates \( z = x/c, \tau = t - x/c \) read [18]:

\[
\begin{align*}
\frac{\partial \Omega_p}{\partial z} &= -q_p N \text{Im}(a_1^* a_2), \\
\frac{\partial \Omega_s}{\partial z} &= -q_s N \text{Im}(a_3^* a_2), \\
\Omega_p \frac{\partial a_2}{\partial z} &= q_p N \text{Re}(a_1^* a_2), \\
\Omega_s \frac{\partial a_2}{\partial z} &= q_s N \text{Re}(a_3^* a_2).
\end{align*}
\]

Here \( q_p, q_s = 2\pi \omega_{p,s} d_{p,s} / \hbar c \) are the oscillator strengths of the atom-field couplings with \( N \) being the atomic number density.

The coupled equations [4] and [5] give a complete description of the problem we are considering. Analytical solution to the set of coupled equations for the case of equal oscillator strengths \( q_p = q_s \) was given and studied in the adiabatic following approximation in [17]. It was shown that the efficiency of the population transfer in the case of equal oscillator strengths decreases rapidly with the propagation length for small one-photon detunings, \( \Delta_p \sim \Omega \), meanwhile for large one photon detunings, \( \Delta_p \gg \Omega \), the population transfer process is more efficient in the medium and can occur for longer propagation distances.

However, in most practical cases, the oscillator strengths of the allowed transitions are not equal. We will analyze what happens when the oscillation strengths of the corresponding transitions are different.

We first solve numerically the set of coupled Schrödinger-Maxwell equations [4] and [5], and in Section IV we interpret them using approximate analytical solutions. We define the ratio of the transition strengths \( q = q_p / q_s \).

III. NUMERICAL RESULTS AND ANALYSIS

For the desired population transfer it is required that state vector \( |\Phi\rangle \) follow adiabatically the bright \( |b_1\rangle \) state in the course of the evolution: \( |\langle b_1 \vert z, \tau \vert \Phi \rangle| \approx 1 \). This is achieved by switching on the pulses in the intuitive order (the pump laser first) and by meeting the adiabaticity condition [17]

\[
|\lambda_{b_1} - \lambda_{b_2}| \gg |\langle b_2 \vert b_1 \rangle|, \quad |\lambda_{b_1} - \lambda_d| \gg |\langle d \vert b_1 \rangle|,
\]

where \( \lambda_{b_1}, \lambda_{b_2} \) and \( \lambda_d \) are the eigenvalues associated with the dressed states \( |b_1\rangle, |b_2\rangle \) and \( |d\rangle \), respectively. These adiabaticity conditions are generally satisfied, for smooth pulses, if:

\[
|\Delta_p T| \gg 1, \quad |\Delta_p| \psi^2 \sim \Omega^2 T / |\Delta_p| \gg 1. \quad (7)
\]

For the numerical investigation we consider Gaussian pulses at the medium entrance \( (z = 0) \) with equal durations and Rabi frequencies. The pulses should act in a way that eliminates transitions between different dressed states, i.e. they should satisfy conditions [7]. For that we choose the following parameters for the pulses:

\[
\Omega_0 T = \Delta_p T = 40, \quad \tau_d / T = 1.3, \quad \text{where } \tau_d \text{ is the time delay between the peaks of the pulses and } \Omega_0 \text{ is the peaks value of } \Omega.
\]

The chosen parameters correspond to the case \( \Delta_p \approx \Omega_0 \).

A. Case of equal oscillator strengths

We start with the example when the oscillator strengths of the corresponding transitions are equal, corresponding to \( q = 1 \). Figure 2 shows the time evolution of the propagating pulses (top left), atomic state populations (bottom left), state vector \( |\Phi\rangle \) projections onto the dressed states (top right) and mixing angles \( \theta \) and \( \psi \) (bottom right) at the entrance of
is not anymore a smooth decreasing monotonic function

FIG. 3: (Color online) The same dynamics as in Fig. 2 but at the propagation length: (a) \( q_s zNT = 7 \). The efficiency achieved for population transfer is \( \sim 95\% \); (b) \( q_s zNT = 20 \). The efficiency achieved for population transfer is \( \sim 2\% \).

(a)

(b)

FIG. 3: (Color online) The same dynamics as in Fig. 2 but at the propagation length: (a) \( q_s zNT = 7 \). The efficiency achieved for population transfer is \( \sim 95\% \); (b) \( q_s zNT = 20 \). The efficiency achieved for population transfer is \( \sim 2\% \).

the medium. The curves are obtained from a numerical solution of Eqs. (4)-(5) using the above mentioned parameters. The figure shows that the pulses induce a very efficient adiabatic population transfer. However, as the pulses propagate inside the medium the population transfer efficiency rapidly decreases. Indeed, as one can see from Fig. 3, at the propagation length \( q_s zNT = 7 \) (see Fig. 3(a)) the population transfer is already not complete (the achieved efficiency is \( \sim 95\% \)), and at \( q_s zNT = 20 \) (see Fig. 3(b)) the transfer efficiency is reduced by a factor of 50 (the achieved efficiency \( \sim 2\% \)). The reason for such a loss of the efficiency of the transfer process is that during propagation the adiabaticity of the interaction is more and more disturbed. Indeed, the time evolution of the mixing angle \( \theta(z, t) \) inside the medium is not anymore a smooth decreasing monotonic function (ensuring the evolution of the bright state \( |b_1\rangle \) from the bare state \( |1\rangle \) initially to the target state \( |3\rangle \) at the end of the interaction), but reveals some oscillating behavior, resulting in nonadiabatic couplings (proportional to \( \dot{\theta} \)) between the adiabatic states. The violation of the adiabaticity is more apparent when we look at the top right panels in Figs. 3(a)[b] presenting the populations of the dressed states. One can see that the bright state \( |b_1\rangle \) is not the only adiabatic state that is populated, as it is depopulated in the course of propagation and there appears a non-adiabatic coupling to the dark state \( |d\rangle \), resulting in loss of transfer efficiency. This is what we also see from the top left panels presenting the atomic state populations \( P_{1,3} \): the interference between different evolution paths leads to oscillations in the populations, rather than to complete population transfer.

B. Case of unequal oscillator strengths

In this subsection we proceed with the same input conditions, but we lift the requirement of equal coupling constants. Figure 4 shows the time evolution of the propagating pulses (top left), atomic state populations (bottom left), state vector \( |\Phi\rangle \) projections (top right) and dressing angles \( \theta \) and \( \psi \) (bottom right) at the entrance of a medium with \( q = 0.1 \). As seen, in this case the pulses also provide an adiabatic evolution and a very efficient population transfer at the entry of the medium. We will study whether this capacity of propagating pulses is maintained or not in the course of propagation.

In Figs. 4(a)[b] we report a numerical plot of the dynamics of the propagating pulses and of the atomic system at the propagation lengths \( q_s zNT = 7 \) and \( q_s zNT = 20 \), respectively. As seen from the figures, in the course of propagation the interaction adiabaticity is better preserved as compared to the case \( q = 1 \). Indeed, at the propagation length \( q_s zNT = 7 \) the population transfer is still complete (compare with Fig. 3(a) where at this propagation length the transfer is already not perfect). As to the propagation length \( q_s zNT = 20 \), the situation is not perfect. However, the time dependence of the mixing angle \( \theta \) is without pronounced peaks, its final value goes to zero, and the majority of population (about 87.5 %) is transferred to the final state \( |3\rangle \) (compare this result with 2% in case of equal oscillator strengths, see Fig. 3(b)). It is also notable that while in case of \( q = 1 \) the pulses are considerably distorted at this propagation length, in case of \( q = 0.1 \) the pulse distortion is much less (compare top left panels in Figs. 4(b) and 3(b)). Even though the pulses produce perfect adiabatic population transfer at the entry into the medium both for \( q = 1 \) and \( q = 0.1 \), in the course of propagation they maintain this ability much longer in case of \( q < 1 \) as compared to the case where \( q = 1 \).

Consider now a medium of unequal oscillator strengths such that \( q > 1 \). The dynamics of the trans-
fer process together with the pulse shapes for this case is shown in Figs. 4 and 5 for $q = 10$. It is seen that the population transfer process in the medium is less efficient compared to the cases $q = 0.1$ and $q = 1$. Indeed, by comparing Figs. 4(a) and 4(a) and 4(b) one can see that while for $q \leq 1$ at the propagation length $q_s z NT = 7$ the population transfer works quite well, in case of $q = 10$ the efficiency of the transfer already at this length is far from perfect ($\sim 25\%$). So, in this case in the course of propagation in the medium, the adiabaticity breaks down rather quickly, and both pulses undergo severe reshaping and, consequently, loose their capacity to produce an effective population transfer. Hence, the case $q > 1$ is more harmful to the transfer process than that of $q \leq 1$. In conclusion to this section one can see that the pulse dynamics depends on the ratio of the oscillator strengths as they characterize the speed of energy transfer from the pulses to the medium and vice-versus.

IV. ANALYTICAL SOLUTIONS

We now focus our attention on approximate analytical solutions that give an explanation for the above numerical results.

By combining Maxwell and Schrödinger equations and differentiating the phase equations [4(a)] and [4(b)] with respect to time we obtain the following system of equations for the Rabi frequencies $\Omega_{p,s}$ and the one-photon detunings $\Delta_{p,s}$

$$\frac{\partial \Omega_{p}^2}{\partial z} = q_p N \frac{\partial}{\partial \tau} |a_1|^2,$$  \hspace{1cm} (8a)
$$\frac{\partial \Omega_{s}^2}{\partial z} = q_s N \frac{\partial}{\partial \tau} |a_3|^2,$$  \hspace{1cm} (8b)
$$\frac{\partial \Delta_{p}}{\partial z} = q_p N \frac{\partial}{\partial \tau} \frac{\text{Re}(a_1 \ast a_2)}{\Omega_p},$$  \hspace{1cm} (9a)
$$\frac{\partial \Delta_{s}}{\partial z} = q_s N \frac{\partial}{\partial \tau} \frac{\text{Re}(a_3 \ast a_2)}{\Omega_s}.$$  \hspace{1cm} (9b)

Equations (8a) describe the change in carrier frequencies of the pulses during propagation in a non-linear medium due to the parametric broadening of the pulse spectrum.
and propagation length \( Q \) defined as

\[
Q = \frac{Q_p}{Q_{ps}} = \frac{Q_p}{\sin^2 \theta + Q_p \cos^2 \theta}.
\]

(Phase-self modulation).

In the process we are concerned with the population of the level \(|1\rangle\) decreases \((\partial |a_1|^2 / \partial \tau < 0)\), while that of the third level increases \((\partial |a_3|^2 / \partial \tau > 0)\). Hence, the intensity of the pump pulse decreases proportionally to \(q_p\), and that of the Stokes one increases proportionally to \(q_s\).

Choosing a medium with a small value of \(q = q_p / q_s\), we slow down the process of pump pulse depletion (but not the process of Stokes pulse amplification), so one would expect to extend the population transfer process up to longer propagation lengths for such media.

From Eqs. (3) we obtain immediately the following equation of motion for the total photon number density \(n = \frac{\Omega_p^2}{\Omega_p^2 + \Omega_s^2}\), since the system is conservative \((|a_1|^2 + |a_2|^2 + |a_3|^2 = 1)\):

\[
\frac{\partial n}{\partial z} = -N \frac{\partial |a_2|^2}{\partial \tau}.
\]

(10) According to this equation, the total photon number density \(n(z, \tau)\) during the propagation of the pulses in the medium is not conserved if the intermediate level \(|2\rangle\) is populated, i.e. a part of the energy of the pulses is transferred to the medium.

We introduce now a quantity \(Q = \frac{\Omega_p^2}{n}\) that we call two-photon transition strength (similar to \(q_{ps} = \Omega_{ps}^2 / n_{ps}\)) defined as

\[
Q = \frac{q_s q_p}{q_s \sin^2 \theta + q_p \cos^2 \theta}.
\]

(11) Note that in case of equal oscillator strengths \((q_p = q_s = q)\) \(Q = q\), while for \(q_p \neq q_s\) \(Q\) is a function of both time and propagation length \(Q = Q(z, \tau)\).

FIG. 6: (Color online) The dynamics at the input face of the medium with unequal coupling constants, \(q = 10\). Top left: propagating fields; top right: projections of the state vector \(|\Phi\rangle\) onto dressed \(|b_1\rangle\) (red line), \(|d\rangle\) (black line) and \(|b_2\rangle\) (blue line) states; bottom left: state populations; bottom right: mixing angles \(\theta\) and \(\psi\).

From Eqs. (10) and (11), using the adiabatic approximation along the eigenstate \(|b_1\rangle\) (see Eq. (1)) and the definition of the two-photon detuning, we arrive at the following system of propagation equations for \(n\) and \(\delta\):

\[
\frac{\partial n}{\partial z} + \frac{N \Delta_p Q}{(\Delta_p^2 + 4nQ)^{3/2}} \frac{\partial n}{\partial \tau} = -\frac{N n \Delta_p}{(\Delta_p^2 + 4nQ)^{3/2}} \frac{\partial Q}{\partial \tau},
\]

(12)

\[
\frac{\partial \delta}{\partial z} = \frac{2(q_p - q_s)}{\Delta_p} \frac{\partial n}{\partial z}.
\]

(13) As can be seen from these equations, the evolution dynamics of both \(n\) and \(\delta\), during the pulse propagation in the medium is clearly dependent on the ratio of \(q_p\) and \(q_s\). Indeed, in case of equal transition strengths \((q_p = q_s)\) the condition of two-photon resonance \((\delta = 0)\) is kept...
automatically. However, for unequal oscillator strengths \((q_p \neq q_s)\), the two-photon detuning \(\delta\) is affected by the evolution dynamics of \(n(z, \tau)\), and the condition of two-photon resonance can be broken during the propagation of the pulses in the medium. So, self-phase modulation can start to develop as the pulses propagate inside the medium leading to a change in the spectra of both pulses, and consequently to the destruction of the bright state \(|b_1\rangle\).

However, the analysis of Eq. (12) shows that \(\frac{\partial n}{\partial z} \approx 0\) at the propagation length satisfying the condition

\[
\frac{\Delta_p Q N}{(\Delta_p^2 + 4nQ)^{3/2}} z \ll 1, \tag{14}
\]

which at large one-photon detunings reduces to

\[
\frac{QN}{\Delta_p^2} z \ll 1. \tag{15}
\]

Under this condition the two-photon resonance is preserved: \(\frac{\partial \delta}{\partial z} \approx 0\). Condition (14) is similar to that of the generalized adiabaticity for a simple two-level system when replacing the two-photon oscillator strength by a one-photon oscillator strength.

Note that when deriving Eq. (13) we neglected the time dependence of the one-photon detuning \(\Delta_p\), which is valid for large initial values of this parameter and at the propagation lengths satisfying the condition (15).

Thus, provided that one remains in the regime given by the condition (14), the conservation law of the total photon number density is guaranteed in case of different transition strengths, i.e. \(n(z, \tau) = n_0(\tau)\), and one can neglect small deviations from the two-photon resonance condition.

In case of equal oscillator strengths \((q_p = q_s)\) the dynamics of the photon number density \(n\) in a medium coincides with that of the generalized Rabi frequency \(\Omega\) and is studied in [17]. As shown in this work, the photon number density \(n\) propagates in a medium with a non-linear group velocity less than \(c\). In this case the condition (14) means that the group delay in the medium is negligibly small.

### A. Equations and solutions for the mixing angle \(\theta\):
Superluminal population transfer.

As seen from the numerical study performed in Sec. III, the mixing angle \(\theta(z, \tau)\) appears to be the key dynamical parameter in the interaction between the atoms and the fields. In order to follow the propagation dynamics of the angle \(\theta(z, \tau)\) we will derive an evolution equation for \(\theta(z, \tau)\).

Using the definitions of \(\theta\), \(\Omega\) and \(Q\), we obtain the following expressions for \(\Omega_{p,s}^2\)

\[
\Omega_{p}^2(z, \tau) = nQ(\theta(z, \tau)) \sin^2(\theta(z, \tau)), \tag{16a}
\]

\[
\Omega_{s}^2(z, \tau) = nQ(\theta(z, \tau)) \cos^2(\theta(z, \tau)). \tag{16b}
\]

A suitable combination of Eqs. (16a) and (16b), yields the desired evolution equation for \(\theta(z, \tau)\):

\[
\sin 2\theta(z, \tau) \left[ \frac{\partial \theta(z, \tau)}{\partial z} - \frac{q_p q_s N}{Q^2(\theta(z, \tau))} \frac{\cos^2 \psi \partial \theta(z, \tau)}{\partial \tau} \right] = 0. \tag{17}
\]

This equation is a central equation of our study that helps to understand the main properties and limitations for population transfer process during propagation in a medium.

The analytical solution to Eq. (17) is complicated in the general case, so for simplicity we consider the case of large one-photon detunings \((\Delta_p T \gg 1)\) where \(\cos^2 \psi \sim 1\). In this case Eq. (17) can be solved analytically by the method of characteristics as in [16], and the solution reads

\[
\theta(z, \tau) = \theta_0(\xi), \tag{18}
\]

where \(\theta_0(\xi) \equiv \theta(z = 0, \tau = \xi)\) is the function given at the medium entrance, \(z = 0\). Here \(\xi(z, \tau)\) is an implicit function governing the nonlinear propagation of the pulses and determined from the following integral equation

\[
\int_{-\infty}^{\xi} n_0(\tau')d\tau' = \int_{-\infty}^{\tau} n_0(\tau')d\tau' + \frac{q_p q_s}{Q^2(\xi(z, \tau))} N z. \tag{19}
\]

Equation (19) defines the "nonlinear" time \(\xi = \tau - z/u(z, \tau)\) with \(u(z, \tau)\) being the "nonlinear" velocity at which the mixing angle \(\theta\) propagates. As seen from this equation, at the medium entrance \((z = 0)\) \(\xi = \tau\), while inside the medium \((z \neq 0)\) the nonlinear time \(\xi\) is larger than \(\tau\): \(\xi > \tau - x/c\). This means that the mixing angle \(\theta(z, \tau)\) propagates with a velocity exceeding the light speed in vacuum \(c\), i.e. superluminally.

### B. The adiabaticity criterion

The obtained analytical result (18) relies on the adiabaticity condition (6) requiring the energy spacing between the eigenvalues to be much larger than the dynamic coupling term (given by \(b\)) which ensures the adiabatic following of the bright state \(|b_1\rangle\) during the propagation of the pulses. Let us see whether the adiabaticity condition satisfied at the medium entrance, remains valid in the course of propagation.

With propagation effects taken into account, the time derivative \(\dot{\theta}\) takes the form \(d\theta_0/d\tau = (d\theta_0/d\xi)\partial \xi/d\tau\). So, even though we impose at the medium entrance a small derivative \(d\theta_0/d\xi\), the adiabaticity condition can break down, since during the propagation process the derivative \(\partial \xi/d\tau\) can become considerably large. Indeed, differentiating equation (19) with respect to \(\tau\), we obtain
for the derivative $\partial \xi / \partial \tau$

\[
\frac{\partial \xi}{\partial \tau} = \frac{n_0(\tau)}{n_0(\xi)} A^{-1},
\]

(20)

with

\[
A = 1 - \frac{2(q_s - q_p)}{\Omega^2_s(\xi)} N z \frac{d\theta_0(\xi)}{d\xi} \sin 2\theta,
\]

(21)

where $\Omega_0(\xi)$ is the function given at the medium entrance, $z = 0$. As seen from the obtained equation, at small values of $A$ the derivative $\partial \xi / \partial \tau$ becomes large leading to the violation of the adiabaticity condition. The adiabaticity condition is satisfied if $A \geq 1$, namely under the following condition

\[
(q_s - q_p) \frac{d\theta_0}{d\xi} \sin 2\theta_0 \leq 0.
\]

(22)

In particular, for a medium with equal transition strengths ($q_s = q_p$) we have $A = 1$. For an intuitive pulse sequence the angle $\theta$ changes from $\pi/2$ to $0$, so the derivative $\frac{d\theta_0(\xi)}{d\xi} \leq 0$ throughout the interaction. Hence, in the case where $q_s \geq q_p$ the factor $A$ is always more than 1, and consequently, the adiabaticity condition in principle never breaks down during the propagation process. In the opposite case of $q_s < q_p$, at the propagation lengths defined by the following condition

\[
z \approx - \frac{\Omega^2_s T}{2(q_s - q_p)}.
\]

(23)

the factor $A \to 0$, and consequently $\partial \xi / \partial \tau \to \infty$ ($\dot{\theta} \to \infty$). The condition $q_s < q_p$ means that the probability of the transition $|1\rangle \to |2\rangle$ is greater than that of the transition $|2\rangle \to |3\rangle$ and, thus, the population transfer $|1\rangle \to |2\rangle$ dominates the depletion of level $|2\rangle$, i.e., the interaction adiabaticity breaks down, and the state $|b_1\rangle$ does not carry the dynamics anymore.

Thus, the generalized condition for the interaction adiabaticity is very sensitive to the ratio of the transition strengths on the adjacent transitions. Note, that the increase in the derivative $\partial \theta / \partial \tau$ during the propagation process (i.e. increase of the influence of superadiabatic corrections) is a property of media consisting of atoms with unequal transition strengths.

The above arguments are illustrated in Fig. 8 presenting the time evolution of the mixing parameter $\theta(z, \tau)$ as given by Eq. (19) for different values of the parameter $q$ at the propagation length $q_s z NT = 20$. Identifying the slope of $\theta$ as the measure of nonadiabaticity, we can see from this figure that the evolution of the mixing parameter is more adiabatic in the case where $q = 0.1$ (dashed curve), while for $q = 14$ (full curve) the slope becomes steeper in the course of propagation, implying that the adiabaticity condition breaks down. So, the population trans diarric process is more stable against the nonadiabaticity caused by nonequality of coupling constants in case of $q_p \leq q_s$.

The analytical solution given above is valid in the region where both pulses overlap, and where the two-photon resonance is physically significant. The overlapping region is defined by $\Omega_p \Omega_s = 0.4 \sin 2\theta \neq 0$. Note, that outside the overlapping region where $\Omega \to 0$, even for an isolated atom the adiabaticity condition can not be satisfied.
C. Population transfer in the adiabatic limit

We now investigate the possibility of a complete population transfer during the pulse propagation in the medium in the adiabatic limit. In this limit the population of the final level evolves as

\[ P_3(z, \tau) = \cos^2 \psi(z, \tau) \cos^2 \theta_0(\xi(z, \tau)). \]  \tag{24}

Taking \( \cos^2 \psi \sim 1 \) (which is valid for large single-photon detunings), we see that a complete population transfer in the medium at a given propagation length \( z \) occurs when \( \theta_0(\xi) = 0 \) which is realized at the times \( \xi(z, \tau) \to \infty \). Setting \( \xi(z, \tau) \) equal to \( \infty \) in the analytical solution \( \psi \), we can obtain from the curve \( z(\tau) \) defined from the following equation

\[ \int_\tau^\infty n_0(\tau')d\tau' = (q_p/q_s)\Omega z(\tau), \]  \tag{25}

a set of points (located on \( z(\tau) \)) at which the population transfer is complete. The question is whether for each given value of \( z \) there exists \( \tau \) such that equation (25) is satisfied. As one can see, at the medium entrance (\( z = 0 \)) a complete population transfer is realized at the end of the interaction (\( \tau \to \infty \)), while for atoms located at \( z \neq 0 \), a complete transferring process occurs at earlier times \( \tau \) (before the interaction is switched off). As compared to an isolated atom, the population transfer process via b-STIRAP in a medium is a faster process, that is, a superluminal population transfer, as long as the distortion of the pulses do not prevent adiabatic passage.

In principle, Eq. (24) leads to the maximal propagations length \( z_{\text{max}} \) defined by the equation

\[ \int_{-\infty}^{\infty} n_0(\tau')d\tau' = (q_p/q_s)\Omega z_{\text{max}}, \]  \tag{26}

beyond which population transfer cannot occur. This means in terms of energy that population transfer can in principle lasts until the transfer process uses all the photon available in the pulses.

Figure 9 shows the normalized value of \( z_{\text{max}} \) as given by solution (26) for different values of the ratios between \( q_p \) and \( q_s \). The analytical solution obtained can not be applied beyond the regions delimited by the presented curves. Besides condition (14), we thus obtain a second limitation on propagation lengths at which our solution is valid.

The physical meaning of the maximal propagation length \( z_{\text{max}} \) given by Eq. (26) becomes more clear in case of equal oscillator strengths. Indeed in this case on the left-hand side of Eq. (26), we have the total number of photons in both pulses \( N_{\text{ph}} \) passing through the unit area, and on the right-hand side of this equation we have the total number of atoms \( N_{z_{\text{max}}} \) interacting with the radiation in the unit area. So, in completely symmetric case where the number of photons in the pump and Stokes pulses are equal, it has a trivial meaning: for each atom in a medium where population transfer occurs at the two-photon resonance correspond two photons. In the asymmetric case, considered in the present paper, we have "effective photon number" given by

\[ (q_p/q_s)N_{\text{ph}} = N_{\text{atoms}}. \]  \tag{27}

As follows from the above theoretical analysis, for small propagation lengths at which pulse deformations are still negligible, the population transfer process does not differ from that of an isolated atom. However, at large propagation lengths the population transfer process becomes faster. The transfer process is restricted up to certain propagation lengths determined from the conditions (14) and (26) that should be met for a successful transfer. In case of \( q_p/q_s > 1 \) there is an additional limitation on the propagation length given by the adiabaticity condition (23). The propagation length \( z_{\text{max}} \) [as given by Eq. (26)] at which a complete population transfer is possible in a medium is the smallest length among those defined by the conditions (14) and (26). To estimate propagation distances \( z \), we consider a set of parameters relevant to a typical alkali atom vapor: \( N = 10^{13} \) atoms/cm\(^2\), \( \omega = 10^{15} \) s\(^{-1}\), \( d \approx 0.8 \times 10^{-17} \) SGS units, \( T = 10^{-9} \) s. Estimations show that \( q_{\omega}zNT = 1 \) corresponds to 0.05 cm. For a medium with \( q = 0.5 \) an efficient b-STIRAP transfer is possible, in principle, up to \( z = 300 \) cm, while for \( q = 5 \) it is limited up to \( z = 13 \) cm.

V. CONCLUSION

In this paper we have presented a detailed study of population transfer process via b-STIRAP in a medium of three-level \( \Lambda \)-atoms with unequal oscillator strengths of corresponding atomic transitions. The propagation equations describing the dynamics of the process have been derived and approximate analytical solutions have been obtained. It is shown that the population transfer efficiency is sensitive to the ratio of the oscillator strengths, \( q = q_p/q_s \), and can be increased by a proper choice of this parameter. In particular, we find that the transfer efficiency is severely affected in case of \( q > 1 \) and rapidly decreases with propagation length, while in case of \( q \leq 1 \) propagating pulses maintain their capacity to produce a complete population transfer over larger propagation lengths. The analytical solution obtained has allowed us to investigate in detail the adiabaticity condition in a medium and the fact the transfer can occur superluminaly. The results show that the adiabaticity requirements fulfilled at the medium entrance are better maintained during propagation when \( q \leq 1 \), while for \( q > 1 \) the adiabaticity breaks down rather quickly in the course of propagation, and the propagating pulses undergo severe distortions. The conditions restricting the propagation length at which a complete population transfer via b-STIRAP in a medium occurs are derived.
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