Stimulated Raman adiabatic passage in a Λ system in the presence of quantum noise

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We exploit a microscopically derived master equation for the study of stimulated Raman adiabatic passage in the presence of spontaneous decay from the intermediate state toward the initial and final states and compare our results with the predictions obtained from a phenomenological model used previously [P. A. Ivanov, N. V. Vitanov, and K. Bergmann, Phys. Rev. A **72**, 053412 (2005)]. It is shown that our approach predicts a much higher efficiency for counterintuitively ordered pulses, while no significant difference between the two approaches is found for intuitively ordered pulses. These features are readily explained in the dressed-state picture.

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I. INTRODUCTION

Stimulated Raman adiabatic passage (STIRAP) [1,2] is a fruitful application of the adiabatic theorem [3] that allows the transfer of a population from a quantum state of a physical system toward another state, through an auxiliary intermediate state [4]. The passage occurs via a *dark* state, which aligns with the initial state in the beginning of the process and then gradually changes its structure until complete alignment with the target state occurs. The process ends when the dark and the target states coincide. Because for such an alignment it is necessary first to couple the final state with the intermediate one and then to couple the intermediate state with the initial one, this sequence of pulses is named counterintuitive.

With the intuitive sequence, corresponding to the opposite delay of the two couplings between the three states, and for nonzero single-photon detuning, it is possible to realize the bright-STIRAP (or simply b-STIRAP) process, which instead exploits the adiabatic change of a *bright* eigenstate of the Hamiltonian [5]. The main difference between STIRAP and b-STIRAP is that in the latter case the auxiliary state is effectively involved in the dynamics, because a certain amount of population is temporarily transferred to it during the process. This circumstance makes the b-STIRAP more sensitive than STIRAP to the presence of decay from the auxiliary level. In fact, while in the absence of environmental interaction and classical noise, the transfer from the initial state to the target state is predicted to be perfect, in the presence of dissipative dynamics the efficiency of the process is negatively affected.

Many models have been considered to study the effect of dephasing [6] and spontaneous emission from the auxiliary level, either toward external states [7] or toward internal states [8], that is, toward the initial or the target state. In all these models, the incoherent dynamics has been taken into account phenomenologically. Recently, in the case of time-independent Hamiltonian operators, discrepancies between phenomenological and microscopic models have been brought to light [9]. The term "microscopic" here means that the master equation is derived starting from the system Hamiltonian. Very recently, a microscopic model to describe the STIRAP and b-STIRAP processes under dissipation from the auxiliary state toward external states was presented [10]. The solution of the master equation derived from a model of interaction between the three-state system and a bosonic environment shows some interesting deviations from the predictions related to the phenomenological counterpart. In particular, the microscopic model predicts a much higher efficiency in the STIRAP scheme.

In this paper, by using a microscopic model in the spirit of Ref. [10], we investigate the effect of spontaneous decay from the intermediate state inside the Λ system. Because the initial and final states in STIRAP are usually ground or metastable, the intermediate state is necessarily an excited state, which may decay both inside and outside the system [11–15]. The loss of efficiency caused by external decay is more detrimental, for it leads to irreversible population loss from the system; it is also easier to describe and understand [7,10]. The effect of internal decay on STIRAP is a much more subtle effect because the loss of efficiency is compensated by the concomitant optical pumping [8]. Here we develop a rigorous microscopic theory of internal decay in STIRAP and b-STIRAP, which reveals some unexpected features compared to the phenomenological model [8].

Starting from a microscopic model of system-environment interaction, we derive a time-dependent master equation that describes the dynamics of our system. Then, after considering the resolution of the master equation, we compare the predictions coming from our model with the results coming from the phenomenological description of an analogous decay scheme [8]. We find that the efficiency of the STIRAP process is higher than predicted before. Exploiting the master equation at nonzero temperature, we also study the effects of temperature, showing that the thermal pumping dramatically and negatively affects the efficiency of the population transfer in the STIRAP process, while it has a slightly positive effect in b-STIRAP.

The paper is organized as follows. In Sec. II we present the derivation of the Markovian master equation of a system with a time-independent Hamiltonian and a time-dependent system-environment interaction term. In Sec. III we apply the result from the previous section and the general theory of Davies and Spohn [16,17] to derive the master equation of the three-state system. Then, in Sec. IV we present the results obtained at zero temperature and compare them with the results coming from the phenomenological model. In Sec. V we show the effects of temperature, and finally, in Sec. VI we make some concluding remarks.

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II. GENERAL FORMALISM

In this section we consider the general problem of the derivation of the master equation for a system whose Hamiltonian H_s is constant, while the system-environment interaction Hamiltonian contains oscillation terms. This is useful in the next section, where we deal with a system described (in a rotating frame) by a slowly varying Hamiltonian and interacting with a thermal bath through oscillating terms. According to the general theory of Davies and Spohn [16,17], under the hypothesis that the environmental correlation time is much smaller than the time scale of the Hamiltonian change, we can treat this system by assuming that the system Hamiltonian is time independent during the derivation of the master equation and putting the time dependence of the jump operators only after the derivation.

Therefore we start by considering a time-independent system Hamiltonian H_s and the following time-dependent system-bath interaction Hamiltonian:

$$H_I = \sum_\alpha (A_\alpha^+ e^{i\omega_\alpha t} + A_\alpha^- e^{-i\omega_\alpha t}) \otimes B_\alpha. \tag{1}$$

Following the approach presented in [19], let us introduce, for each Bohr frequency ω,

$$A_\alpha^\pm(\omega) = \sum_{\epsilon'-\epsilon=\omega} \Pi(\epsilon) A_\alpha^\pm \Pi(\epsilon'), \tag{2}$$

where $\Pi(\epsilon)$ is the projector on the subspace of the system Hilbert space corresponding to the energy eigenvalue ϵ and the sum is extended over all the couples of energies ϵ and ϵ' such that $\epsilon' - \epsilon = \omega$. The operators defined in this way satisfy both

$$[H_s, A_\alpha^\pm(\omega)] = -\omega_\alpha A_\alpha^\pm(\omega) \tag{3}$$

and

$$[A_\alpha^\pm(\omega)]^\dagger = A_\alpha^\mp(-\omega), \tag{4}$$

giving

$$e^{i H_s t} A_\alpha^\pm(\omega) e^{-i H_s t} = e^{-i\omega t} A_\alpha^\pm(\omega), \tag{5}$$

$$e^{i H_s t} [A_\alpha^\pm(\omega)]^\dagger e^{-i H_s t} = e^{i\omega t} [A_\alpha^\pm(\omega)]^\dagger. \tag{6}$$

Another important property is that summing over all the Bohr frequencies (both positive and negative), one reobtains the initial operators:

$$A_\alpha^\pm = \sum_\omega A_\alpha^\pm(\omega). \tag{7}$$

In the Schrödinger picture we thus have

$$H_I = \sum_{\alpha,\omega} [A_\alpha^+(\omega) e^{i\omega_\alpha t} + A_\alpha^-(\omega) e^{-i\omega_\alpha t}] \otimes B_\alpha, \tag{8}$$

which, in the interaction picture with respect to $H_s + H_B$, becomes

$$H_I = \sum_{\alpha,\omega} e^{-i\omega t} [A_\alpha^+(\omega) e^{i\omega_\alpha t} + A_\alpha^-(\omega) e^{-i\omega_\alpha t}] \otimes B_\alpha(t) \tag{9}$$

or, taking the Hermitian conjugate,

$$H_I = \sum_{\alpha,\omega} e^{i\omega t} \{[A_\alpha^+(\omega)]^i \dagger e^{-i\omega_\alpha t} + [A_\alpha^-(\omega)]^\dagger e^{i\omega_\alpha t}\} \otimes B_\alpha^\dagger(t). \tag{10}$$

The formal resolution of the Liouville equation gives

$$\dot\rho = \int_0^\infty ds \, \mathrm{tr}_B\{H_I(t-s)\rho(t)\rho_B H_I(t) - H_I(t)H_I(t-s)\rho(t)\rho_B\} + \text{H.c.}, \tag{11}$$

from which, substituting the expansions of H_I, one gets the following master equation:

$$\begin{aligned}\dot\rho = &\sum_{\omega,\omega'}\sum_{\alpha,\beta} e^{i(\omega-\omega'+\omega_\beta-\omega_\alpha)t}\,\Gamma_{\alpha\beta}^{++}(\omega)(A_\beta^+(\omega)\rho(A_\alpha^+(\omega'))^\dagger \\ &- (A_\alpha^+(\omega'))^\dagger A_\beta^+(\omega)\rho) + \sum_{\omega,\omega'}\sum_{\alpha,\beta} e^{i(\omega-\omega'+\omega_\beta+\omega_\alpha)t}\,\Gamma_{\alpha\beta}^{-+}(\omega) \\ &\times (A_\beta^+(\omega)\rho(A_\alpha^-(\omega'))^\dagger - (A_\alpha^-(\omega'))^\dagger A_\beta^+(\omega)\rho) \\ &+ \sum_{\omega,\omega'}\sum_{\alpha,\beta} e^{i(\omega-\omega'-\omega_\beta-\omega_\alpha)t}\,\Gamma_{\alpha\beta}^{+-}(\omega)(A_\beta^-(\omega)\rho(A_\alpha^+(\omega'))^\dagger \\ &- (A_\alpha^+(\omega'))^\dagger A_\beta^-(\omega)\rho) + \sum_{\omega,\omega'}\sum_{\alpha,\beta} e^{i(\omega-\omega'-\omega_\beta+\omega_\alpha)t}\,\Gamma_{\alpha\beta}^{--}(\omega) \\ &\times (A_\beta^-(\omega)\rho(A_\alpha^-(\omega'))^\dagger - (A_\alpha^-(\omega'))^\dagger A_\beta^-(\omega)\rho) + \text{H.c.},\end{aligned} \tag{12}$$

with

$$\Gamma_{\alpha\beta}^{++}(\omega) = \Gamma_{\alpha\beta}^{-+}(\omega) = \int_0^\infty ds e^{i(\omega-\omega_\beta)s}\langle B_\alpha^\dagger(t) B_\beta(t-s)\rangle \tag{13}$$

and

$$\Gamma_{\alpha\beta}^{+-}(\omega) = \Gamma_{\alpha\beta}^{--}(\omega) = \int_0^\infty ds\, e^{i(\omega+\omega_\beta)s}\langle B_\alpha^\dagger(t)\, B_\beta(t-s)\rangle. \tag{14}$$

This is the most general form of the Born-Markov master equation before a rotating wave approximation (RWA) is performed. Under the hypothesis that $\omega_\alpha, \omega_\beta \gg \omega, \omega'$, one can single out very clear conditions for RWA. The only terms that survive are those for which ω_α and ω_β appear in the combination $\omega_\alpha - \omega_\beta$, with $\alpha = \beta$, and $\omega = \omega'$:

$$\begin{aligned}\dot\rho = &\sum_\omega\sum_\alpha \Gamma_{\alpha\alpha}^{++}(\omega)(A_\alpha^+(\omega)\rho(A_\alpha^+(\omega))^\dagger - (A_\alpha^+(\omega))^\dagger A_\alpha^+(\omega)\rho) \\ &+ \sum_\omega\sum_\alpha \Gamma_{\alpha\alpha}^{--}(\omega)(A_\alpha^-(\omega)\rho(A_\alpha^-(\omega))^\dagger \\ &- (A_\alpha^-(\omega))^\dagger A_\alpha^-(\omega)\rho) + \text{H.c.},\end{aligned} \tag{15}$$

which, neglecting the Lamb shifts and coming back to the Schrödinger picture, becomes

$$\begin{aligned}\dot\rho = &-i[H_s,\rho] + \sum_\omega\sum_\alpha \gamma_{\alpha\alpha}^{++}(\omega)(A_\alpha^+(\omega)\rho(A_\alpha^+(\omega))^\dagger \\ &- \frac{1}{2}\{(A_\alpha^+(\omega))^\dagger A_\alpha^+(\omega),\rho\}) \\ &+ \sum_\omega\sum_\alpha \gamma_{\alpha\alpha}^{--}(\omega)(A_\alpha^-(\omega)\rho(A_\alpha^-(\omega))^\dagger \\ &- \frac{1}{2}\{(A_\alpha^-(\omega))^\dagger A_\alpha^-(\omega),\rho\}),\end{aligned} \tag{16}$$

where $\gamma_{\alpha\alpha}^{++}(\omega) = 2\mathrm{Re}\{\Gamma_{\alpha\alpha}^{++}(\omega)\}$ and $\gamma_{\alpha\alpha}^{--}(\omega) = 2\mathrm{Re}\{\Gamma_{\alpha\alpha}^{--}(\omega)\}$.

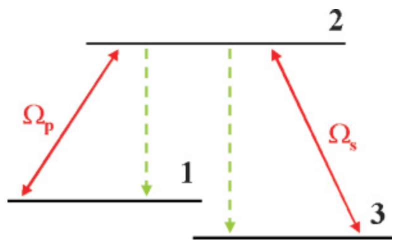

FIG. 1. (Color online) States $|1\rangle$ and $|3\rangle$ are coherently coupled to level $|2\rangle$ [solid (red) arrows]. State $|2\rangle$ is coupled to the other two states by a dipolar system-environment interaction [dashed (green) arrows].

III. OUR MODEL

A. The system

We consider a three-level system in the Λ configuration (shown in Fig. 1) whose Hamiltonian is

$$H_{\rm sys}(t) = \begin{bmatrix} \omega_1 & \Omega_p(t)e^{i(\omega_{21}-\Delta)t} & 0 \\ \Omega_p(t)e^{-i(\omega_{21}-\Delta)t} & \omega_2 & \Omega_s(t)e^{-i(\omega_{23}-\Delta)t} \\ 0 & \Omega_s(t)e^{i(\omega_{23}-\Delta)t} & \omega_3 \end{bmatrix}. \tag{17}$$

The system interacts with a bosonic bath. The free bath is described by

$$H_B = \sum_k \omega_k b_k^\dagger b_k, \tag{18}$$

while the system-bath interaction Hamiltonian is

$$H_{\rm int} = \sum_k g_k^{(12)}(|1\rangle\langle 2| + |2\rangle\langle 1|)(b_k + b_k^\dagger) + \sum_k g_k^{(32)}(|3\rangle\langle 2| + |2\rangle\langle 3|)(b_k + b_k^\dagger). \tag{19}$$

In the rotating frame associated with the transformation $T(t) = e^{i\omega_1 t}|1\rangle\langle 1| + e^{i(\omega_2-\Delta)t}|2\rangle\langle 2| + e^{i\omega_3 t}|3\rangle\langle 3|$, the total Hamiltonian,

$$H = H_s(t) + H_B + H_I(t), \tag{20}$$

with

$$H_s(t) = \begin{bmatrix} 0 & \Omega_p(t) & 0 \\ \Omega_p(t) & \Delta & \Omega_s(t) \\ 0 & \Omega_s(t) & 0 \end{bmatrix}, \tag{21}$$

and

$$H_I(t) = \sum_k g_k^{(12)}(e^{i(\omega_1-\omega_2+\Delta)t}\,|1\rangle\langle 2| + e^{-i(\omega_1-\omega_2+\Delta)t}\,|2\rangle\langle 1|)(b_k + b_k^\dagger) + \sum_k g_k^{(32)}(e^{i(\omega_3-\omega_2+\Delta)t}\,|3\rangle\langle 2| + e^{-i(\omega_3-\omega_2+\Delta)t}\,|2\rangle\langle 3|)(b_k + b_k^\dagger). \tag{22}$$

The eigenstates and eigenvalues of $H_s(t)$ are

$$\omega_+ = \Omega_0 \cot\varphi, \quad \omega_0 = 0, \quad \omega_- = -\Omega_0 \tan\varphi, \tag{23a}$$

$$|+\rangle = \sin\varphi\sin\theta|1\rangle + \cos\varphi|2\rangle + \sin\varphi\cos\theta|3\rangle, \tag{23b}$$

$$|0\rangle = \cos\theta|1\rangle - \sin\theta|3\rangle, \tag{23c}$$

$$|-\rangle = \cos\varphi\sin\theta|1\rangle - \sin\varphi|2\rangle + \cos\varphi\cos\theta|3\rangle, \tag{23d}$$

where

$$\tan\theta(t) = \frac{\Omega_p(t)}{\Omega_s(t)}, \tag{24a}$$

$$\tan 2\varphi(t) = \frac{2\Omega(t)}{\Delta(t)}, \tag{24b}$$

$$\Omega(t) = \sqrt{\Omega_p^2(t) + \Omega_s^2(t)}. \tag{24c}$$

It is well known [5] that, for the intuitive sequence of pulses, in which the probe pulse $\Omega_p(t)$ precedes the Stokes pulse $\Omega_s(t)$, and for any nonvanishing detuning $\Delta \neq 0$, one has $|-\rangle = |1\rangle$ for $t \to -\infty$ and $|-\rangle = |3\rangle$ for $t \to \infty$. Therefore, if all the pulses vary adiabatically, the population of state $|1\rangle$ can be transferred to state $|3\rangle$: this process is called b-STIRAP. In contrast, if $\Omega_s(t)$ precedes $\Omega_p(t)$, independently of the detuning, one has $|0\rangle = |1\rangle$ for $t \to -\infty$ and $|0\rangle = |3\rangle$ for $t \to \infty$: in this counterintuitive sequence the population from $|1\rangle$ to $|3\rangle$ is adiabatically transferred through state $|0\rangle$ and the process is called STIRAP.

B. Master equation

Since in the rotating frame we have a slowly varying system Hamiltonian and a time-dependent system-environment interaction term, we can use the general formalism presented before to derive the master equation that describes the dynamics of our system. We obtain the following master equation (for details, see Appendix A):

$$\begin{aligned}
\dot\rho = &-i[H_s,\rho] + [\gamma_{aa}^{++}(\omega_{+0})\cos^2\theta\cos^2\varphi + \gamma_{bb}^{++}(\omega_{+0})\sin^2\theta\cos^2\varphi]\big(|0\rangle\langle +|\rho|+\rangle\,\langle 0| - \tfrac{1}{2}\,\{|+\rangle\langle +|,\rho\}\big) \\
&+ [\gamma_{aa}^{--}(\omega_{0-})\cos^2\theta\sin^2\varphi + \gamma_{bb}^{--}(\omega_{0-})\sin^2\theta\sin^2\varphi]\,\big(|-\rangle\,\langle 0|\,\rho|0\rangle\,\langle -| - \tfrac{1}{2}\,\{|0\rangle\,\langle 0|\,,\rho\}\big) \\
&+ [\gamma_{aa}^{++}(\omega_{+-})\sin^2\theta\cos^4\varphi + \gamma_{aa}^{--}(\omega_{+-})\sin^2\theta\sin^4\varphi + \gamma_{bb}^{++}(\omega_{+-})\cos^2\theta\cos^4\varphi + \gamma_{bb}^{--}(\omega_{+-})\cos^2\theta\sin^4\varphi] \\
&\times\,\big(|-\rangle\langle +|\rho|+\rangle\,\langle -| - \tfrac{1}{2}\,\{|+\rangle\langle +|,\rho\}\big) \\
&+ [(\gamma_{aa}^{++}(0) + \gamma_{aa}^{--}(0))\sin^2\theta\sin^2\varphi\cos^2\varphi + (\gamma_{bb}^{++}(0) + \gamma_{bb}^{--}(0))\cos^2\theta\sin^2\varphi\cos^2\varphi] \\
&\times\big[(|+\rangle\langle +| - |-\rangle\,\langle -|)\,\rho\,(|+\rangle\langle +| - |-\rangle\,\langle -|) - \tfrac{1}{2}\,\{|+\rangle\langle +| + |-\rangle\,\langle -|\,,\rho\}\big] \\
&+ [\gamma_{aa}^{--}(\omega_{0+})\cos^2\theta\cos^2\varphi + \gamma_{bb}^{--}(\omega_{0+})\sin^2\theta\cos^2\varphi]\,\big(|+\rangle\,\langle 0|\,\rho|0\rangle\langle +| - \tfrac{1}{2}\,\{|0\rangle\,\langle 0|\,,\rho\}\big)
\end{aligned}$$

$$+ [\gamma_{aa}^{++}(\omega_{-0})\cos^2\theta\sin^2\varphi + \gamma_{bb}^{++}(\omega_{-0})\sin^2\theta\sin^2\varphi]\left(|0\rangle\langle-|\rho|-\rangle\langle 0| - \tfrac{1}{2}\{|-\rangle\langle-|,\rho\}\right)$$
$$+ [\gamma_{aa}^{--}(\omega_{-+})\sin^2\theta\cos^4\varphi + \gamma_{aa}^{++}(\omega_{-+})\sin^2\theta\sin^4\varphi + \gamma_{bb}^{--}(\omega_{-+})\cos^2\theta\cos^4\varphi + \gamma_{bb}^{++}(\omega_{-+})\cos^2\theta\sin^4\varphi]$$
$$\times\left(|+\rangle\langle-|\rho|-\rangle\langle+| - \tfrac{1}{2}\{|-\rangle\langle-|,\rho\}\right), \tag{25}$$

where $\omega_{nm} = \omega_n - \omega_m$.

From (13) and (14) one gets that the decay rates are given by a spectral density $J_j(\omega)$ multiplied by a factor depending on the photon population $N(\omega)$ of the bath modes at the relevant frequency corrected with $\pm\omega_a$ or $\pm\omega_b$ depending on the case; that is,

$$\begin{cases} \gamma_{jj}^{++}(\omega) = J_j(\omega-\omega_j)\,(1+N(\omega-\omega_j)), & \omega-\omega_j > 0,\\ \gamma_{jj}^{--}(\omega) = J_j(\omega+\omega_j)\,(1+N(\omega+\omega_j)), & \omega+\omega_j > 0,\\ \gamma_{jj}^{++}(\omega) = J_j(|\omega-\omega_j|)\,N(|\omega-\omega_j|), & \omega-\omega_j < 0,\\ \gamma_{jj}^{--}(\omega) = J_j(|\omega+\omega_j|)\,N(|\omega+\omega_j|), & \omega+\omega_j < 0, \end{cases} \tag{26}$$

with $j = a,b$ and

$$\omega_a = \omega_1 - \omega_2 + \Delta, \tag{27a}$$
$$\omega_b = \omega_3 - \omega_2 + \Delta. \tag{27b}$$

The zero-temperature spectral density $J_j(\omega)$ for a general bosonic reservoir is given by [18,19]

$$J_j(\omega) = d(\omega)\,|g_j(\omega)|^2, \tag{28}$$

where $g_a(\omega)$ [$g_b(\omega)$] is the system-reservoir coupling constant $g_k^{(12)}$ [$g_k^{(32)}$] in the continuum limit, and $d(\omega)$ is the reservoir density of states at frequency ω.

It is important to note that, under the hypothesis that $\omega_j \gg \omega$ for any Bohr frequency ω between the dressed states in the rotating frame (which is the usual case since ω_j's are optical frequencies associated with the atomic transitions, while ω's are of the order of magnitude of the coupling terms Ω's), and taking into account that the frequencies in (27a) are negative, the only conditions satisfied are $\omega - \omega_j > 0$ and $\omega + \omega_j < 0$. Therefore, in (26), only the rates of the first and fourth classes are possible. Moreover, at zero temperature, only the rates of the first class survive, since the number of photons in the reservoir is 0. In such a case the master equation becomes

$$\dot\rho = -i[H_s,\rho] + [\gamma_{aa}^{++}(\omega_{+0})\cos^2\theta\cos^2\varphi + \gamma_{bb}^{++}(\omega_{+0})\sin^2\theta\cos^2\varphi]\left(|0\rangle\langle+|\rho|+\rangle\langle 0| - \tfrac{1}{2}\{|+\rangle\langle+|,\rho\}\right)$$
$$+ [\gamma_{aa}^{++}(\omega_{-0})\cos^2\theta\sin^2\varphi + \gamma_{bb}^{++}(\omega_{-0})\sin^2\theta\sin^2\varphi]\left(|0\rangle\langle-|\rho|-\rangle\langle 0| - \tfrac{1}{2}\{|-\rangle\langle-|,\rho\}\right)$$
$$+ [\gamma_{aa}^{++}(\omega_{+-})\sin^2\theta\cos^4\varphi + \gamma_{bb}^{++}(\omega_{+-})\cos^2\theta\cos^4\varphi]\times\left(|-\rangle\langle+|\rho|+\rangle\langle-| - \tfrac{1}{2}\{|+\rangle\langle+|,\rho\}\right)$$
$$+ [\gamma_{aa}^{++}(\omega_{-+})\sin^2\theta\sin^4\varphi + \gamma_{bb}^{++}(\omega_{-+})\cos^2\theta\sin^4\varphi]\times\left(|+\rangle\langle-|\rho|-\rangle\langle+| - \tfrac{1}{2}\{|-\rangle\langle-|,\rho\}\right)$$
$$+ [\gamma_{aa}^{++}(0)\sin^2\theta\sin^2\varphi\cos^2\varphi + \gamma_{bb}^{++}(0)\cos^2\theta\sin^2\varphi\cos^2\varphi]$$
$$\times\left[(|+\rangle\langle+| - |-\rangle\langle-|)\rho(|+\rangle\langle+| - |-\rangle\langle-|) - \tfrac{1}{2}\{|+\rangle\langle+| + |-\rangle\langle-|,\rho\}\right]. \tag{29}$$

This equation shows that at zero temperature there are the following processes: transitions from $|+\rangle$ to $|-\rangle$, and vice versa, transitions from $|+\rangle$ to $|0\rangle$ and from $|-\rangle$ to $|0\rangle$, and a dephasing process involving levels $|+\rangle$ and $|-\rangle$ (see Fig. 2). This suggests the idea that the damping can help to transfer the population to level $|0\rangle$, so that the efficiency of the counterintuitive sequence should be positively affected by the dissipation.

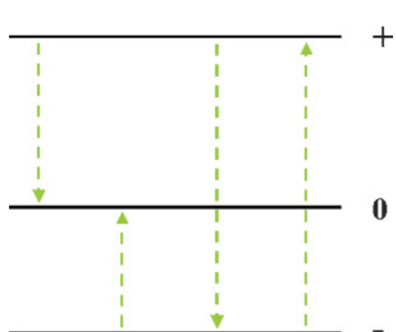

FIG. 2. (Color online) Scheme of the decays for the dressed states. There are transitions from $|+\rangle$ to $|-\rangle$, and vice versa. Both $|+\rangle$ and $|-\rangle$ decay toward $|0\rangle$. The dephasing between state $|+\rangle$ and state $|-\rangle$ is not represented.

IV. ANALYSIS OF THE EFFICIENCY AT ZERO TEMPERATURE

In this section we analyze the efficiency of both the STIRAP and the b-STIRAP processes, by numerically studying the postpulse population of the target state $|3\rangle$ and compare the prediction of our model with the predictions of a phenomenological model introduced in Ref. [8].

We consider Gaussian edges for the laser pulses,

$$\Omega_1 = \frac{\Omega_0}{2}\,e^{-(t-\tau/2)^2/T^2}, \tag{30a}$$
$$\Omega_2 = \frac{\Omega_0}{2}\,e^{-(t+\tau/2)^2/T^2}, \tag{30b}$$

taking into account that we have the so-called intuitive sequence (which corresponds to b-STIRAP) when $\Omega_p = \Omega_2$

and $\Omega_s = \Omega_1$, while we get the counterintuitive sequence (which corresponds to STIRAP) when $\Omega_p = \Omega_1$ and $\Omega_s = \Omega_2$. The adiabatic condition is fulfilled when the amplitude and the characteristic duration of the pulses are such that $\Omega_0 T \gg 1$ (see Appendix B).

The phenomenological model which we compare with our microscopic model corresponds to the following master equation:

$$\dot{\rho} = -i[H,\rho] - \tfrac{1}{2}D, \tag{31}$$

with

$$D = \begin{pmatrix} -2\Gamma_1\rho_{22} & (\Gamma_1+\Gamma_3)\rho_{12} & 0 \\ (\Gamma_1+\Gamma_3)\rho_{21} & 2(\Gamma_1+\Gamma_3)\rho_{22} & (\Gamma_1+\Gamma_3)\rho_{23} \\ 0 & (\Gamma_1+\Gamma_3)\rho_{32} & -2\Gamma_3\rho_{22} \end{pmatrix}, \tag{32}$$

which describes spontaneous emission from level 2 to levels 1 and 3 at rates Γ_1 and Γ_3, respectively. Such a master equation is related to the bare states and then turns out to be time independent.

Concerning the microscopic model, we assume a flat spectrum for both the transition $2 \to 1$, corresponding to $J_a(\omega)$, and the transition $2 \to 3$, corresponding to $J_b(\omega)$. In particular, we assume $J_a(\omega) \equiv \Gamma$ and $J_b(\omega) \equiv \alpha\Gamma$. This may come, for instance, from the assumption that the dipole moments between state $|1\rangle$ and state $|2\rangle$ and between state $|3\rangle$ and state $|2\rangle$ are proportional, so that $g_k^{(12)} = \alpha g_k^{(32)}$ for any k in Eq. (19).

A. The counterintuitive sequence

We first analyze the counterintuitive sequence, where the population is carried by the dark state $|0\rangle$. Figure 3 shows the comparison between the microscopic and the phenomenological models, with $\alpha = 1$ and $\Gamma_1 = \Gamma_2 = \Gamma$. It is evident that the microscopic model predicts a very high efficiency (essentially 1) for a wider range of Γ. This can be explained on the basis of the decay scheme in Fig. 2: all the decay processes describe jumps either toward $|0\rangle$ or toward states that, in turn, decay toward $|0\rangle$, so that the dark state is robust against zero-temperature dissipation.

The robustness of the counterintuitive scheme according to the microscopic model is not related to the special choice $\alpha = 1$. Indeed, we have evaluated the efficiency of the transfer for Γ and α spanning the ranges $10^{-2} \leqslant \Gamma \leqslant 10^4$ and $10^{-2} \leqslant \alpha \leqslant 10^4$ and have always found the maximum of population transmission. Therefore, we can assert that the efficiency of the scheme is not affected by a discrepancy in the decay rates.

It is interesting to compare the parameter range of high decay rates, where the difference between the phenomenological and the master equation approaches appears, to the existing experiments. In the implementation of STIRAP with continuous-wave (cw) lasers, the atomic beam crosses two spatially displaced and partially overlapping cw laser beams at right angles [1,15]. The time it takes for the atoms or molecules to cross the laser beams is 1 to 2 orders of magnitude longer than the lifetime of the excited state, which implies $\Gamma T = 10$ to 100. These values are on the edge of the regime ($\Gamma T \gtrsim 100$) where the difference between the two approaches emerges. Therefore, although the foregoing experiments do not allow discrimination between the phenomenological and the microscopic models, this effect can easily be demonstrated in a dedicated experiment.

B. The intuitive sequence

Concerning the b-STIRAP process (i.e., in the intuitive sequence), we find that the two models predict very similar results. In particular, Fig. 4 shows perfect coincidence of the predictions of the two models in the case $\alpha = 1$ and $\Gamma_1 = \Gamma_2 = \Gamma$. Moreover, from Fig. 4 one can see that (for both models) the efficiency is very sensitive to the presence of decays, so that it drops to almost 0 at $\Gamma T = 1$. The

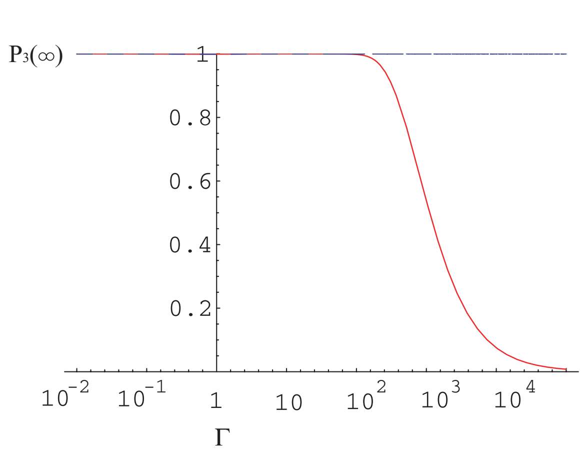

FIG. 3. (Color online) Counterintuitive sequence. Final population of the target state vs Γ (in units of T^{-1} and on a logarithmic scale) according to the microscopic [dashed (blue) line] and phenomenological [solid (red) line] models. The relevant parameters are $\Omega_0 = 25\,T^{-1}$, $\tau = 1.5\,T^{-1}$, $T\,\Delta = 1$, and $\alpha = 1$.

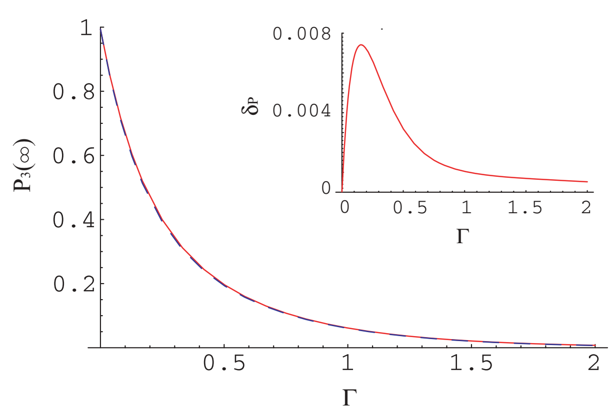

FIG. 4. (Color online) Intuitive sequence. Final population of the target state vs Γ (in units of T^{-1}) according to microscopic [dashed (blue) line] and phenomenological [solid (red) line] model. The relevant parameters are $\Omega_0 = 25\,T^{-1}$, $\tau = 1.5\,T^{-1}$, $T\,\Delta = 1$, and $\alpha = 1$. The two curves are essentially coincident. Inset: Difference δ_P of the postpulse populations related to the phenomenological model and the microscopic one vs. Γ (in units of T^{-1}). This difference is always smaller than 8×10^{-3}.

reason for the fragility of the efficiency in this scheme is that, while all the populations are guided by the decay toward state $|0\rangle$, population transfer is instead carried on by state $|-\rangle$.

It is worth noting that the result of the comparison is qualitatively quite similar to the result of the comparison we made in connection with the scheme with external decay [10]. Indeed, in both cases the predictions from the microscopic and the phenomenological models are almost coincident for the intuitive sequence, while for the counterintuitive sequence we find that the microscopic model predicts a higher efficiency. Nevertheless, we stress here the fact that the enhancement of efficiency in STIRAP in the strong damping limit is due to very different mechanisms in the two cases of external and internal decay. In fact, while for external decay a very strong damping is responsible for a dynamical decoupling of the dark state, which then is protected against losses, in the case of internal decay the dissipation is instead responsible for transitions toward the state that carries the population, therefore protecting the process of population transfer.

V. ANALYSIS OF THE EFFICIENCY AT NONZERO TEMPERATURE

In this section we consider the effects of nonzero temperature. Looking at (26), we see that the $N(\omega)$'s are evaluated at very close frequencies, which are essentially $\omega \approx \omega_{21} \approx \omega_{23}$. For this reason, we described temperature by a single number N, which is the number of photons in the reservoir modes of frequencies close to the bare atomic transitions.

We have seen that at zero temperature, for increasing Γ (and even for quite small values of the decay constant), the efficiency falls to 0 when the intuitive sequence is applied. When temperature is nonvanishing all the transitions included in (25) but not present in (29) must be considered. In particular, transitions from $|0\rangle$ to $|-\rangle$ should increase the efficiency of the b-STIRAP process, since in this scheme the population is transferred through state $|-\rangle$. In contrast, in the counterintuitive sequence we should get a lower transfer efficiency, since thermal terms are responsible for loss of population from state $|0\rangle$ during the STIRAP process.

Figure 5(a) shows the dependence of the postpulse population of state $|3\rangle$ on Γ and temperature (through the number of photons N in the relevant reservoir modes), for the intuitive sequence. It is easily seen that the efficiency, which goes to 0 for large Γ in the zero-temperature regime, reaches nonzero values for nonvanishing temperature. This efficiency reaches a maximum value for intermediate values of temperature. As an example, Fig. 5(b) shows the temperature dependence (in a wider range) of the efficiency for $\Gamma = 1$: in this case the optimal point is reached at $N \simeq 10$. The asymptotic value, corresponding to a very high temperature, is essentially $1/3$, which is traceable back to an equipartition of the population in the three levels, irrespective of the interactions.

Figure 6 shows the postpulse population of state $|3\rangle$ as a function of both Γ and temperature, for the counterintuitive

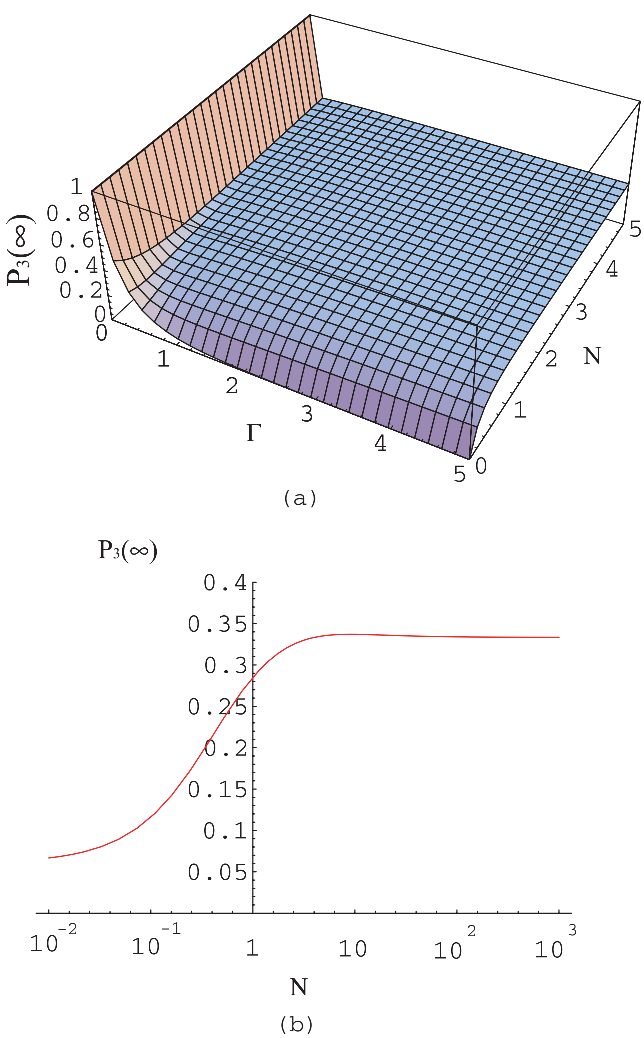

FIG. 5. (Color online) Intuitive sequence. (a) Final population of the target state vs Γ (in units of T^{-1}) and the number of photons N. (b) Final population vs the number of photons N (on a logarithmic scale) for $\Gamma = 1$. In both cases, the relevant parameters are $\Omega_0 = 25\,T^{-1}$, $\tau = 1.5\,T^{-1}$, $T\,\Delta = 1$, and $\alpha = 1$.

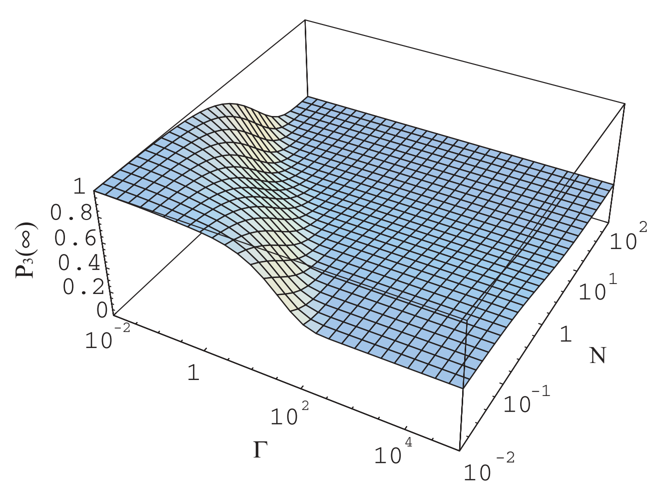

FIG. 6. (Color online) Counterintuitive sequence. Final population of the target state vs Γ (in units of T^{-1} and on a logarithmic scale) and the number of photons N (on a logarithmic scale). The relevant parameters are $\Omega_0 = 25\,T^{-1}$, $\tau = 1.5\,T^{-1}$, $T\,\Delta = 1$, and $\alpha = 1$.

sequence. In this case it is clearly shown that the temperature negatively affects the efficiency of the population transfer. Indeed, even a very small number of thermal photons is responsible for a significant diminishing of the postpulse population, which instead approaches unity at rigorously zero temperature.

VI. DISCUSSION AND CONCLUDING REMARKS

In this paper we have analyzed the effects on a STIRAP scheme of the losses of the auxiliary level to the two metastable states, examined by means of the numerical resolution of a master equation which has been microscopically derived. It is worth noting that, to remove the rapid oscillations in the system Hamiltonian, we are forced to describe the system in a rotating frame, where the system-bath interaction term turns out to be time dependent. Therefore, when deriving the master equation, we have to deal both with a slowly varying system Hamiltonian, which can be treated following the general theory of Davies and Spohn, and a rapidly oscillating system-environment interaction term. The latter point makes the final master equation different from what one usually gets. Indeed, the zero-temperature transitions are guiding the system not toward the dressed (rotating) ground state $|-\rangle$ but, instead, toward state $|0\rangle$. In this sense, the oscillating terms in the system-environment interaction act like a pumping that makes the counterintuitive sequence much more robust than the intuitive sequence.

The inclusion of the nonzero temperature terms in the master equation partially modifies these conclusions. In fact, thermal photons switch on different transitions, which make the postpulse population of state $|-\rangle$ different from 0. As a consequence the efficiency of the counterintuitive sequence is reduced, while, in contrast, the efficiency of the intuitive sequence increases, due to the fact that at very high temperatures the three states are equally populated. However, this increase is not great enough to make the intuitive sequence preferable to the counterintuitive one.

Though the analysis of this STIRAP scheme (with the same decay channels), performed in previous papers by means of phenomenological dissipative terms, has shown a great robustness of the counterintuitive sequence with respect to losses, our analysis shows that the scheme at zero temperature is much more robust for very high decay rates. We conclude by noting that the difference between the values of the postpulse population in the two models is traceable back to different phenomena occurring in the two models: in the phenomenological model strong dissipation causes dynamical decoupling, which forbids the transition from level 1 to level 3 (according to the analysis in [8]), while in the microscopic model the dissipation assists the population transfer, since all the zero-temperature jumps guide the system toward the dark state.

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF THE MASTER EQUATION

In this appendix we give some more details about the derivation of the master equation in our model. In our case we consider

$$A_a^+ = |1\rangle\langle 2|, \tag{A1a}$$
$$A_a^- = |2\rangle\langle 1|, \tag{A1b}$$
$$A_b^+ = |3\rangle\langle 2|, \tag{A1c}$$
$$A_b^- = |2\rangle\langle 3|, \tag{A1d}$$

$$B_a = \sum_k g_k^{(12)}(b_k + b_k^\dagger), \tag{A2a}$$
$$B_b = \sum_k g_k^{(32)}(b_k + b_k^\dagger), \tag{A2b}$$

from which we obtain the following jump operators:

$$\begin{aligned}
&A_a^+(\omega_{+0}) = \cos\theta\cos\varphi|0\rangle\langle +|, && A_a^-(\omega_{+0}) = 0,\\
&A_b^+(\omega_{+0}) = -\sin\theta\cos\varphi|0\rangle\langle +|, && A_b^-(\omega_{+0}) = 0,\\
&A_a^+(\omega_{0-}) = 0, && A_a^-(\omega_{0-}) = -\cos\theta\sin\varphi|-\rangle\langle 0|,\\
&A_b^+(\omega_{0-}) = 0, && A_b^-(\omega_{0-}) = \sin\theta\sin\varphi|-\rangle\langle 0|,\\
&A_a^+(\omega_{+-}) = \sin\theta\cos^2\varphi|-\rangle\langle +|, && A_a^-(\omega_{+-}) = -\sin\theta\sin^2\varphi|-\rangle\langle +|,\\
&A_b^+(\omega_{+-}) = \cos\theta\cos^2\varphi|-\rangle\langle +|, && A_b^-(\omega_{+-}) = -\cos\theta\sin^2\varphi|-\rangle\langle +|,\\
&A_a^+(0) = A_a^-(0) = \sin\theta\sin\varphi\cos\varphi\,(|+\rangle\langle +| - |-\rangle\langle -|)\,,\\
&A_b^+(0) = A_b^-(0) = \cos\theta\sin\varphi\cos\varphi\,(|+\rangle\langle +| - |-\rangle\langle -|)\,.
\end{aligned} \tag{A3}$$

Putting all the jump operators inside the Lindblad form in (16), we get the master equation in (25).

APPENDIX B: ADIABATIC APPROXIMATION

In this appendix we prove that with the pulses in (30a) and (30b), the adiabatic approximation holds, provided $\Omega_0 T \gg 1$.

The condition that guarantees the validity of the adiabatic approximation is

$$\frac{|\langle m|\dot{H}|n\rangle|}{(E_m(t)-E_n(t))^2} \ll 1, \quad \forall m,n, \qquad \text{(B1)}$$

where $E_k(t)$ is the kth instantaneous eigenvalue of the Hamiltonian H, while $|k\rangle$ is the corresponding instantaneous eigenstate. In passing, we mention that recently the efficacy of this condition has been criticized [20] and that, more recently, its sufficiency has been proven, provided that the Hamiltonian contains only real and nonoscillating terms [21]. The necessity always holds [22].

The numerator is a linear combination of the derivatives of the two pulse edges: $\langle m|\dot{H}|n\rangle = \alpha_s\dot{\Omega}_s(t) + \alpha_p\dot{\Omega}_p(t)$. Now, for $\Delta = 0$ one has that the three eigenvalues of H are $\omega_- = -\Omega$, 0, and $\omega_+ = \Omega$, with $\Omega = \sqrt{\Omega_s^2(t)+\Omega_s^2(t)}$, so that the absolute values of all three Bohr frequencies are either Ω or 2Ω. Therefore, the left-hand side of Eq. (B1) is of the order $t/\Omega_0 T^2$, and since in a real experiment t spans values in a range whose amplitude is of the order of T, one has $t/\Omega_0 T^2 \lesssim (\Omega_0 T)^{-1} \ll 1$, and the condition is fulfilled.

Let us now consider the case $\Delta \neq 0$. The numerator is again a linear combination of $\dot{\Omega}_s(t)$ and $\dot{\Omega}_p(t)$, hence keeping its order of magnitude. Concerning the denominator, the three eigenvalues are $\omega_- = (\Delta - \sqrt{\Delta^2 + 4\Omega^2})/2$, 0, and $\omega_+ = (\Delta + \sqrt{\Delta^2 + 4\Omega^2})/2 > \Omega$, so that the only Bohr frequency that has possibly changed its order of magnitude (still nonvanishing, anyway) is that related to the transitions $|-\rangle \leftrightarrow |0\rangle$. Now, since it is easy to verify that $\dot{H}|0\rangle = 0$, which implies $\langle -|\dot{H}|0\rangle = 0$, the condition in Eq. (B1) is also fulfilled in this case.

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