Radiative and collisional damping effects on efficient population transfer in a three-level system driven by two delayed laser pulses

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The effect of radiative decay and collisional damping on the delayed-field population transfer in the three-level system is investigated. The simple analytical steady-state solution of the three-level system, interacting with two intense delayed laser pulses in the dressed-state frame, is obtained and analyzed. The radiative relaxation transfers the system to the "trapped state," which is shown to be stable relative to both radiative decay and collisional damping. The effect of an additional level $|m\rangle$, situated close to the final level $|3\rangle$, on the population-transfer process is considered. The field-induced relaxation rate from intermediate level $|2\rangle$ to level $|3\rangle$ is shown to be a very easily controllable parameter of the field intensities and their relative delay. This result leads to selective population transfer to the desired level and may prevent leaking out of the three-level system. Theoretical analysis corresponds entirely to recent experimental data [U. Gaubatz *et al.*, J. Chem. Phys. **92**, 1 (1990); **92**, 5363 (1990)].

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I. INTRODUCTION

The problem of efficient and selective transfer of population to thermally unpopulated atomic or molecular levels is relevant to several important fields, including spectroscopy and collisional dynamics (by selective vibrational-level excitation), and optical control of chemical reactions.

This problem has been widely investigated and several techniques have been proposed: the π-pulse technique [1–3], adiabatic rapid passage by Stark switching and self-induced sweeping through the two-photon resonance[4,5], off-resonance Raman pumping [6,7], etc. Detailed analysis of these methods is made [8] and each of them is shown to have a serious disadvantage in order to become a practically applicable technique.

The delayed-field technique for efficient and selective population transfer (PT) from a thermally populated level $|1\rangle$ to level $|3\rangle$, via intermediate level $|2\rangle$ by means of interaction with two delayed pump E_p and Stokes E_S laser fields was proposed [9–11] and experimentally demonstrated [8]. The experiment was performed in a molecular sodium beam, interacting with two spatially delayed cw dye lasers and complete PT has been achieved.

The idea of the delayed-field technique is based on the population-trapping phenomena [12,13], which consists of the creation of the coherent superposition of two atomic ground levels connected by two-photon resonant laser fields through a common upper level. This "trapped state" [12,13] has no contribution of an upper atomic level and therefore is radiatively stable in contrast to other dressed states which decay to that one. Up to now the traditional way to reach the trapped state has been as follows. Because of two-photon resonance, switching on the interaction leads to a nonadiabatic transition from the atomic ground level to a mixture of dressed states describing the system. Then, due to radiative decay, after a few lifetimes the system goes over into the pure trapped state [13].

In the case of the delayed-field interaction the contribution of atomic levels into the trapped state (TS) becomes a function of the delay. The proper variation of delay may result in a complete transfer from the trapped state to the desired atomic level [8–11].

The purpose of our present work is to achieve efficient and selective population transfer by the delayed sequence of two laser pulses in a molecular bulk instead of in a beam, and with large enough one-photon detuning in order to escape the problems of nonhomogeneous Doppler broadening, laser linewidth, etc. Therefore the collisional damping effect together with radiation relaxation is to be carefully considered. The influence of additional (more than 3) levels on PT is also analyzed.

We use the set of the dressed states (DS) [14–16] as a basic set of eigenstates, which unifies and simplifies many calculations. The DS are adiabatic eigenstates of system exposed to two strong monochromatic external fields $E_{p,S}$ in a rotating-wave and slowly varying-field-envelope approximation. Light scattering is viewed as spontaneous emission causing transition among these states [16–20].

We start with consideration of adiabatic-following PT without any damping process. Next, the semiclassical approach [21] as well as the explicit solution of the density matrix expressed in the DS representation is performed. These solutions turn out to be quite simple, as the values of nondiagonal elements of the density matrix are equal to zero, therefore the equations for the diagonal elements go over into the "rate equations" [17,22]. In this representation, collisional damping effects, as well as

the problem of additional levels, can easily be included into consideration. More general analysis of the problem of additional levels, involving the interaction with a field is straightforward [23].

II. ADIABATIC-FOLLOWING POPULATION TRANSFER

A. Dressed states for a three-level system

Consider the interaction of a three-level system with two pump E_p and Stokes E_S laser pulses, Fig. 1,

$$E=E_p e^{-i\omega_p t}+E_S e^{-i\omega_S t}+\text{c.c.} \tag{1}$$

In the nondamping-regime approach (the pulse durations $\tau_{1,2}$ are much shorter than the radiative decay time τ_N). The wave functions of such coupled system are presented as follows [12–15] ($|i\rangle=\varphi_i e^{-iE_i t/\hbar}$, atomic states)

$$\Psi=e^{-i\Lambda t}\{C_1|1\rangle+C_2|2\rangle e^{i\Delta t}+C_3|3\rangle e^{i\delta t}\} . \tag{2}$$

Substituting Eq. (2) into the Schrödinger equation

$$ih\frac{\partial\Psi}{\partial t}=(H_0+\Omega_p+\Omega_S)\Psi , \tag{3}$$

one can obtain for C_i $(i=1,2,3)$ the following system of equations:

$$\begin{aligned} &I\dot{C}_1+\Lambda C_1=C_2\Omega_p^* , \\ &i\dot{C}_2+(\Lambda-\Delta)C_2=C_1\Omega_p+C_3\Omega_S , \\ &i\dot{C}_3+(\lambda-\delta)C_3=C_2\Omega_S^* , \end{aligned} \tag{4}$$

where $\Omega_{p,S}$ are the Rabi frequencies

$$\Omega_p=-\frac{d_{21}E_p}{\hbar} , \quad \Omega_S=-\frac{d_{23}E_S}{\hbar} , \tag{5}$$

and $\Delta=\omega_{21}-\omega_p$ is the one-photon detuning, $\delta=\omega_{31}-(\omega_p-\omega_S)$ is the two-photon detuning.

Supposing the fields are varied very slowly (compared to carried frequency $\omega_{p,S}$), one can neglect $\dot{C}_i$ terms.

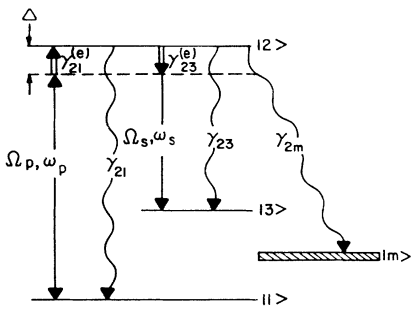

FIG. 1. Representative three-level system $|1\rangle,|2\rangle,|3\rangle$ coupled by two strong laser fields ω_p and ω_S. The relevant Rabi frequencies $\Omega_{p,S}$ and detunings Δ,δ of the laser frequencies from the transition frequencies as well as the radiative γ_{21},γ_{23} and optically induced elastic collisional rates [29] $\gamma_{21}^{(e)},\gamma_{23}^{(e)}$ are indicated, $\Delta=\omega_{21}-\omega_2$; $\delta=\omega_{31}-\omega_1+\omega_2$ (in the case shown, $\delta=0$). Level $|m\rangle$ summarizes all other levels accessible by dipole radiation from level $|2\rangle$.

Then, the equation for energies [12–15]] can be obtained from Eq. (4) (Λ_i is the eigenvalue of the eigenstate Ψ_i),

$$\Lambda(\Lambda-\Delta)(\Lambda-\delta)-\Lambda|\Omega_S|^2-(\Lambda-\delta)|\Omega_p|^2=0 . \tag{6}$$

So, instead of atomic states $|i\rangle$, with energies E_i, we obtain a new set of states Ψ_i with energies Λ_i. The dressed states Ψ_i are the coherent superposition of atomic levels. When one-photon Δ and two-photon δ detunings are large enough ($|\Delta|\tau_{1,2})\gg1$ and $|\delta|\tau_{1,2}\gg1$), Eqs. (2) with energies (6) describe such a type of adiabatic process, under which the atom remains in the initial atomic level $|1\rangle$, after switching off the interaction [14,15].

In the case of exact two-photon resonance $\delta=0$, one can obtain from (6)

$$\Lambda_3=0, \quad \Lambda_{1,2}=\frac{\Delta}{2}[1\mp(1+\Omega^2/\Delta^2)^{1/2}] , \tag{7}$$

where

$$\Omega=(\Omega_p^2+\Omega_S^2)^{1/2} . \tag{8}$$

The DS Ψ_i that correspond to these energies are, respectively,

$$\begin{aligned} \Psi_1&=e^{-i\Lambda_1 t}\left[\frac{a\Omega_p^*}{\Omega}|1\rangle+b|2\rangle e^{i\Delta t}+\frac{a\Omega_S^*}{\Omega}|3\rangle\right] , \\ \Psi_2&=e^{-i\Lambda_2 t}\left[\frac{-b\Omega_p^*}{\Omega}|1\rangle+a|2\rangle e^{i\Delta t}+\frac{-b\Omega_S^*}{\Omega}|3\rangle\right] , \\ \Psi_3&=-\frac{\Omega_S}{\Omega}|1\rangle+\frac{\Omega_p}{\Omega}|3\rangle . \end{aligned} \tag{9}$$

The amplitudes a,b describe the contribution of atomic states into the DS,

$$\begin{aligned} a&=\frac{1}{\sqrt{2}}\left[1+\frac{1}{(1+\Omega^2/\Delta^2)^{1/2}}\right]^{1/2} , \\ b&=\frac{1}{\sqrt{2}}\left[1-\frac{1}{(1+\Omega^2/\Delta^2)^{1/2}}\right]^{1/2} . \end{aligned} \tag{10}$$

The Ψ_3 state consists of coherent superposition of $|1\rangle$ and $|3\rangle$ atomic levels, only, and represents the "trapped state" [12,13]. Since the probabilities of spontaneous transitions $\langle\Psi_3|d|\Psi_{1,2}\rangle$ are equal to zero, this state is radiatively stable (suppose levels $|1\rangle$ and $|3\rangle$ are radiatively stable).

B. Criteria for adiabatic-following evolution

The criteria for adiabatic-following (AF) evolution of a three-level system driven by two quasiresonant laser fields, in the case of concurrent or delayed switching of interaction, is discussed. Obviously, the adiabatic-following evolution condition requires negligibly small coupling between the states Ψ_i, as the molecules interact with the laser pulses. The quantitative definition of adiabaticity is as follows [4,5,14,15,24–27]:

$$|\Lambda_i-\Lambda_j|\tau_{1,2}\gg1 \quad (i\neq j) \tag{11}$$

and has a very simple physical meaning, i.e., the energy splitting $|\Lambda_i-\Lambda_j|$ between the DS should be much larger than the spectral width of the pulses $\tau_{1,2}^{-1}$.

The adiabaticity condition should be maintained during all the interaction processes. The last one may be divided into three ranges (Fig. 2): 1 and 3—the range of switching on and off; 2—the quasistationary range, where the field variation can be omitted.

In a resonant two-level system ($\Delta=0$) the adiabaticity condition cannot be satisfied as far as the absorption processes and the atomic-states mixture occurs. In a two-photon resonant three-level system ($\delta=0$) the adiabaticity condition (11) may be realized within some interval $t_1<t<t_2$ (range 2), supposing the intensity of fields to be strong enough. The probability of nonadiabatic transitions $\Psi_i\leftrightarrow\Psi_j$ inside this interval will be negligibly small [25,26], if

$$w(\Psi_i\leftrightarrow\Psi_j)=|(\Lambda_i-\Lambda_j)\tau_{1,2}|^{-2}\ll 1 \ . \tag{12}$$

However, in the ranges 1 and 3 the condition (11) is violated in the case of concurrent switching ($\Omega_{p,S}=\Omega_{p,S}^{(0)}e^{-(t/\tau)^2}$). As one can see, Eq. (7) at $t\rightarrow\pm\infty$, the energy splitting tends to zero, $(\Lambda_1-\Lambda_3)\rightarrow 0$. The parameters $\Omega_{p,S}/\Omega$ [Eq. (9)] being independent of time, lead to the nonadiabatic mixture both at the beginning of the interaction, when the population of level $|1\rangle$ transfers to the mixture of the states Ψ_1 and Ψ_3 and at the end of the interaction, when the population of the $\Psi_{1,3}$ states transfers to the mixture of atomic $|1\rangle$ and $|3\rangle$ levels.

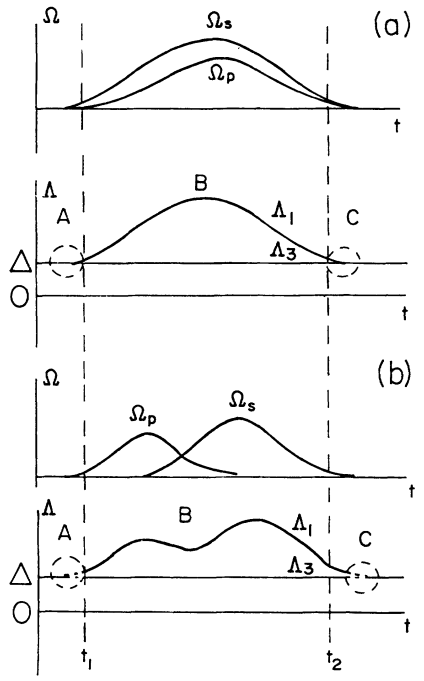

FIG. 2. The time evolution of DS energy levels $\Lambda_{1,3}$ in the case of concurrent (a) and delayed (b) switching of interaction (A,C denote the ranges of the interaction switching; B is the range of adiabatic evolution). (a) A plus C is the range of nonadiabatic transition $|1\rangle\rightarrow|3\rangle$. (b) In ranges A,C there are no transitions, since now at $t<t_1$, $\Omega_S=0$ and at $t>t_2$, $\Omega_p=0$.

The great advantage of the delayed-field idea consists of the possibility to overcome the problem of such nonadiabatic transitions and to treat the three-level system adiabatically both under $\delta=0$ and $\Delta=0$ conditions.

When the Ω_S field precedes Ω_p and $\delta=\Delta=0$, the nonadiabatic transitions between $|2\rangle$ and $|3\rangle$ do not occur, as long as both states are empty. However, these states are shifted by means of the Stark effect. Effective detuning between $|1\rangle$ and $|2\rangle$ become, respectively, $\Delta_{\text{eff}}=\Lambda_1$. Therefore the following switching on of the Ω_p field occurs adiabatically. Suppose the intensity of fields within $t_1<t<t_2$ is large enough until the moment t_2, when the Ω_S is switched off. The population of level $|1\rangle$ transfers through the trapped state Ψ_3 to level $|3\rangle$ (see Sec. II C). Thus, the Ω_p field can be switched off also adiabatically, as long as levels $|1\rangle$ and $|2\rangle$ become empty. When the Ω_p field precedes Ω_S only the two-photon resonant AF procedure can be performed.

C. Time evolution of atomic-states population

The advantages of the delayed-field technique to provide adiabatic population transfer are immediately seen when considering the time evolution of atomic states.

1. *Concurrent switching of interaction* ($\Omega_{p,S}=\Omega_{p,S}^{(0)}e^{-(t/\tau)^2}$)

In accordance with (7) and (9) at $t\rightarrow-\infty$ the population of the ground level $|1\rangle$ is transferred to the mixture of the DS Ψ_1 and Ψ_3. So, under concurrent switching and the short time of interaction the system cannot be transferred completely to the trapped state Ψ_3.

In the steady-state regime complete transfer becomes possible due to the radiative decay process $\Psi_{1,2}\rightarrow\Psi_3$. The following switching off of the interaction at $t\rightarrow\infty$ transfers the Ψ_3 state population to the mixture of atomic levels $|1\rangle$ and $|3\rangle$.

2. *Delayed switching of interaction, Ω_p precedes Ω_S*

The sequental switching on Ω_p and Ω_S pulses transfers the system to the Ψ_1 state, which in the case of large Δ plays a role of trapped state Ψ_3. The sequental turning off of Ω_p and Ω_S transfers the system, respectively, $\Psi_1\rightarrow|3\rangle$. Thus, the complete transfer process during the sequental switching of the interaction is the following:

$$|1\rangle\rightarrow\Psi_1\rightarrow|3\rangle \ . \tag{13}$$

3. *Delayed switching of interaction, Ω_S precedes Ω_p*

This case is the most important. The population of level $|1\rangle$ transfers to the trapped-state at the beginning of the interaction and cannot leave it in the future,

$$|1\rangle\rightarrow\Psi_3\rightarrow|3\rangle \ . \tag{14}$$

The unique properties of the counterintuitive sequence of switching can be seen explicitly. It is very important for application that the trapped state can be achieved through the adiabatic following passage when the interaction time $\tau_{1,2}$ is much shorter than decay time τ_N.

The efficiency of the PT does not depend on one-photon detuning Δ [see Eq. (9)]. The relaxation processes do not disturb the PT efficiency. Moreover, because of radiative decay all the population goes over into the Ψ_3 state.

All these results lead to the following conclusion. In the AF regime efficient PT occurs for proper delay and any sequence of pulses. The partial contradiction of our results with the experiment lies in the steady-state regime being provided in the experiment [8].

III. STEADY-STATE REGIME

A. Semiclassical approach. Probability of transitions between dressed states

The expressions (9) are the adiabatic solutions, and valid only when the pulse duration $\tau_{1,2}$ is much less than decay time τ_N. In the opposite case $\tau_{1,2} > \tau_N$, radiative as well as collisional damping mix these states and should be taken into account.

The quantized radiation field causes spontaneous transitions between $\Psi_i \rightarrow \Psi_j$, with emission of frequencies $\omega_{ij}^{(p,S)} = \omega_{p,S} + (\Lambda_i - \Lambda_j)$. The probability of such spontaneous transitions can be calculated on the basis of semiclassical radiation theory [21]. For example, the probability of the $\Psi_1 \rightarrow \Psi_2$ transition, with emission of ω_T frequency, is $[\omega_T = \omega_p + (\Lambda_1 - \Lambda_2)]$

$$w_{(\omega_T)} = \frac{4\omega_T^3}{3\hbar c^3} |D_{21}(\omega_T)|^2 = \gamma_{21} \left| \frac{\Omega_p}{\Omega} b^2 \right|^2 , \tag{15}$$

with $\gamma_{21} = 4\omega_{21}^3 d_{21}^2 / 3\hbar c^3$. The dipole matrix element for the $\Psi_1 \rightarrow \Psi_2$ transition is determined as

$$D_{2 \leftarrow 1} = \int \Psi_2^* \hat{d} \Psi_1 d^3 r . \tag{16}$$

The full transition probabilities $\Psi_i \rightarrow \Psi_j$ are as follows:

$$\begin{aligned} &w_{1\rightarrow 2} = \Gamma_1 |b|^4, \quad w_{2\rightarrow 1} = \Gamma_1 |a|^4, \quad w_{3\rightarrow 1} = 0 , \\ &w_{1\rightarrow 3} = \Gamma_2 |b|^2, \quad w_{2\rightarrow 3} = \Gamma_2 |a|^2, \quad w_{3\rightarrow 2} 0 , \end{aligned} \tag{17}$$

where $\Gamma_{1,2}$ are the field-induced rates of relaxation $(\gamma_{21} \rightarrow \Gamma_1; \gamma_{23} \rightarrow \Gamma_2)$

$$\begin{aligned} \Gamma_1 &= \gamma_{21} \left| \frac{\Omega_p}{\Omega} \right|^2 + \gamma_{23} \left| \frac{\Omega_S}{\Omega} \right|^2 , \\ \Gamma_2 &= \gamma_{21} \left| \frac{\Omega_S}{\Omega} \right|^2 + \gamma_{23} \left| \frac{\Omega_p}{\Omega} \right|^2 . \end{aligned} \tag{18}$$

We have to emphasize that $w_{3\rightarrow 1} = w_{3\rightarrow 2} = 0$.

B. Quantization of field: strong solution

The description of weak mixing of the states Ψ_i by spontaneous emission using the semiclassical approach is correct only at $w_{i\rightarrow j} t \ll 1$. At times $t \gg \omega_{i\rightarrow j}^{-1}$ spontaneous emission leads to sufficiently strong dressed-states mixing. The "atom plus fields" system in this case must be described by a noncoherent mixture,

$$\Psi = \sum_{i=1,2,3} C_i(t) \Psi_i . \tag{19}$$

In order to find $C_i(t)$ values and to extend the method of DS to times $t \gtrsim \gamma^{-1}$, we consider the exact equations for the density matrix [28], valid for an arbitrary dependence of the external fields on time,

$$i \frac{\partial}{\partial t} \hat{\rho} = \{\hat{\rho}, \hat{H}\} + i \Gamma \hat{\rho} , \tag{20}$$

where $\hat{H}$ is the Hamiltonian of the system atom plus intense fields, and the term $\Gamma\hat{\rho}$ is expressed exactly by rigorously taking into account the interaction with the quantized field. The nonzero elements of the matrix Γ are [17]

$$\begin{aligned} &\Gamma_{12,12} = \Gamma_{21,21} = \Gamma_{23,23} = \Gamma_{32,32} = -\Gamma/2 , \\ &\Gamma_{22,22} = -\Gamma, \quad \Gamma_{11,22} = \gamma_{21}, \quad \Gamma_{33,22,} = \gamma_{23} , \end{aligned} \tag{21}$$

where $\Gamma = \gamma_{21} + \gamma_{23} = \Gamma_1 + \Gamma_2$ in the collisionless regime.

Now the correct procedure of taking the collisional damping into account can be provided by the following substitution into the atomic rate, $\Gamma = \gamma_{21} + \gamma_{23} + \gamma_{21}^{(e)} + \gamma_{23}^{(e)}$. The terms $\gamma_{21}^{(e)} + \gamma_{23}^{(e)}$ describe the "elastic" collisionally induced optical transitions [29] from a virtual to a real level (Fig. 1). We neglect very small terms $\gamma_{ij}^{(I)}$ that are responsible for inelastic collisionally induced transitions $|2\rangle \leftrightarrow |1\rangle$ and $|2\rangle \leftrightarrow |3\rangle$.

The transformation of Eq. (20) into the DS representation [16–18] is as follows:

$$\hat{\rho} = \Psi\Psi^\dagger = \sum_{i,j} C_i C_j^* \Psi_i \Psi_j^\dagger = \sum Q_{ij} \varphi_i \varphi_j^* , \tag{22}$$

making the substitutions [see Eq. (9)] $Q_{ij} = C_i C_j^* e^{-i(\Lambda_i - \Lambda_j)t}$, $\Psi_j = \varphi_j e^{-i\Lambda_j t}$. Equation (20) can be rewritten as

$$i \frac{dQ_{\alpha\beta}}{dt} + i(\Lambda_\alpha - \Lambda_\beta) Q_{\alpha\beta} = \sum_{\mu,\nu=1,2,3} \gamma_{\alpha\beta,\mu,\nu} Q_{\mu\nu} , \tag{23}$$

where

$$\gamma_{\alpha\beta,\mu\nu} = \sum_{i,j,k,l} (\varphi_\alpha^\dagger)_i (\varphi_\beta)_j \Gamma_{ij,kl} (\varphi_\mu)_k (\varphi_\nu^\dagger)_l \tag{24}$$

and $(\varphi_\alpha)_i$ is the ith component of the spinor φ_α [see Eq. (9)]. In the case of monochromatic external fields the values $\gamma_{\alpha\beta,\mu\nu}$ are time independent. The nonzero transition rates are compiled in Table I. Substituting the data from Table I into Eq. (23) one obtains

$$\begin{aligned} \frac{dQ_{11}}{dt} &= Q_{22} w_{2\rightarrow 1} - Q_{11}(w_{1\rightarrow 2} + w_{1\rightarrow 3}) \\ &\quad - \gamma_{11,12}(Q_{12} + Q_{21}) , \\ \frac{dQ_{22}}{dt} &= Q_{11} w_{1\rightarrow 2} - Q_{22}(w_{2\rightarrow 1} + w_{2\rightarrow 3}) \\ &\quad - \gamma_{22,12}(Q_{12} + Q_{21}) , \\ \frac{dQ_{12}}{dt} &+ i(\Lambda_1 - \Lambda_2) Q_{12} = \gamma_{12,12}(Q_{12} + Q_{21}) \\ &\quad + \gamma_{12,11} Q_{11} + \gamma_{12,22} Q_{22} , \end{aligned} \tag{25}$$

TABLE I. Relaxation rates of transition between the DS Ψ_i ($i=1,2,3$).

$\gamma_{11,11}=-(\Gamma_1\lvert b\rvert^4+\Gamma_2\lvert b\rvert^2)$	$\gamma_{22,22}=-(\Gamma_1\lvert a\rvert^4+\Gamma_2\lvert a\rvert^2)$
$\gamma_{11,22}=\Gamma_1\lvert a\rvert^4=w_{2\to1}$	$\gamma_{22,11}=\Gamma_1\lvert b\rvert^4=w_{1\to2}$
$\gamma_{11,12}=\gamma_{11,21}=ab\left[\frac{\Gamma}{2}-\Gamma_1\lvert a\rvert^2\right]$	$\gamma_{22,12}=\gamma_{22,21}=ab\left[\frac{\Gamma}{2}-\Gamma_1\lvert b\rvert^2\right]$
$\gamma_{21,21}=\gamma_{12,12}=\left[\frac{\Gamma}{2}+\Gamma_1\lvert ab\rvert^2\right]$	$\gamma_{33,11}=\Gamma_2\lvert b\rvert^2\equiv w_{1\to3}$
$\gamma_{21,12}=\gamma_{12,21}=-\Gamma_1\lvert ab\rvert^2$	$\gamma_{33,22}=\Gamma_2\lvert a\rvert^2\equiv w_{2\to3}$
$\gamma_{21,11}=\gamma_{12,11}=ab\left[\frac{\Gamma}{2}+\Gamma_1\lvert b\rvert^2\right]$	$\gamma_{33,12}=\gamma_{33,21}=\Gamma_2 ab$
$\gamma_{21,22}=\gamma_{12,22}=ab\left[\frac{\Gamma}{2}+\Gamma_1\lvert a\rvert^2\right]$	$\gamma_{13,13}=\gamma_{31,31}=\frac{\Gamma}{2}\lvert b\rvert^2;\ \gamma_{32,32}=\gamma_{23,23}=\frac{\Gamma}{2}\lvert a\rvert^2$

$$\frac{dQ_{33}}{dt}=-\frac{d}{dt}(Q_{11}+Q_{22})=Q_{11}w_{1\to3}+Q_{22}w_{2\to3}+\Gamma_2 ab(Q_{12}+Q_{21})\ .$$

Expressions (25) can be simplified, using the "strong-interaction" condition, which requires that the radiative decay rate Γ, including the collisional width $\gamma^{(e)}=\gamma_{21}^{(e)}+\gamma_{23}^{(e)}$ be much smaller than the energy splitting. This is equivalent to an adiabaticity condition Γ, $\gamma^{(e)}\ll|\Lambda_i-\Lambda_j|$. Then, we can neglect the time derivatives $\dot{Q}_{12,21}\sim\Gamma Q_{12,21}$ compared to the $(\Lambda_1-\Lambda_2)Q_{12,21}$ terms. Thus, the value of nondiagonal $Q_{12,21}$ elements can be estimated as $Q_{12,21}\sim\Gamma/(\Lambda_1-\Lambda_2)$, allowing them to be ignored in Eq. (25) relative to diagonal elements Q_{ii}. Consequently, Eq. (25) is converted to the rate equations [17,22]

$$\frac{dQ_{ii}}{dt}=\sum_k \gamma_{ii,kk}Q_{kk}\ . \tag{26}$$

We stress, that collision-dependent terms $\gamma_{21}^{(e)},\gamma_{23}^{(e)}$ are included in the nondiagonal rate coefficients $\gamma_{ij,kl}$ only (see Table I). Consequently, the collisional damping process does not affect the population of the DS Ψ_i.

Inspection of Eqs. (25) shows that under $t\gg w_{i\to j}^{-1}$, the system evolves completely to the trapped state Ψ_3, e.g.,

$$Q_{33}=1,\quad Q_{11}=Q_{22}=Q_{21}=Q_{12}=0\ . \tag{27}$$

Thus, the state of the system in the steady-state regime is described by the wave function (19)

$$\Psi(t\gg w_{i\to j}^{-1})=\Psi_3\ . \tag{28}$$

Delayed switching off the $\Omega_{p,S}$ fields leads the system either to level $|3\rangle$ when Ω_S precedes Ω_p, or to level $|1\rangle$ when Ω_p precedes Ω_S.

C. Effects of additional levels $|m\rangle$ on population-transfer selectivity and efficiency

Suppose that we have the set of additional levels $|m\rangle$ situated close to level $|3\rangle$. They are not coupled to the laser field and can be reached by dipole-allowed transitions from level $|2\rangle$. Under these conditions Eqs. (25) are converted to the following form:

$$\begin{aligned}
\frac{dQ_{11}}{dt}&=Q_{22}w_{2\to1}-Q_{11}\left[w_{1\to2}+w_{1\to3}+\sum_m w_{1\to m}\right],\\
\frac{dQ_{22}}{dt}&=Q_{11}w_{1\to2}-Q_{22}\left[w_{2\to1}+w_{2\to3}+\sum_m w_{2\to m}\right],\\
\frac{dQ_{33}}{dt}&=Q_{11}w_{1\to3}+Q_{22}w_{2\to3}=\Gamma_2(Q_{11}|b|^2+Q_{22}|a|^2)\ ,\\
\frac{dQ_{mm}}{dt}&=Q_{11}w_{1\to m}+Q_{22}w_{2\to m}\\
&=\gamma_{2m}(Q_{11}|b|^2+Q_{22}|a|^2)\ .
\end{aligned} \tag{29}$$

Here, we used the results of Secs. III A and III B in order to obtain the transition probabilities $\omega_{1,2\to m}$,

$$w_{1\to m}=\gamma_{2m}|b|^2,\quad w_{2\to m}=\gamma_{2m}|a|^2\ . \tag{30}$$

Substituting expressions (30) into Eq. (29) we obtain

$$\frac{d}{dt}\left[\frac{Q_{33}}{\Gamma_2}-\frac{Q_{mm}}{\gamma_{2m}}\right]=0\ . \tag{31}$$

The solution of Eq. (31) is

$$\frac{Q_{33}}{\Gamma_2}=\frac{Q_{mm}}{\gamma_{2m}} \tag{32}$$

because at $t\to-\infty$ the system occupied only the ground level $|1\rangle$. Taking into account the normalization procedure, one can obtain from (32)

$$\begin{aligned}
Q_{33}&=\frac{\Gamma_2}{\Gamma_2+\sum_m\gamma_{2m}}\ ,\\
Q_{m'm'}&=\frac{\gamma_{2m'}}{\Gamma_2+\sum_m\gamma_{2m}}\ .
\end{aligned} \tag{33}$$

Expressions (33) show the effect of additional levels $|m\rangle$ located near level $|3\rangle$ on the PT process. In the steady-state regime part of population leaves state Ψ_3 and

goes over into the levels $|m\rangle$, which decreases the efficiency of the PT. Comparison of the field-induced rate Γ_2 with γ_{2m} leads to the following conclusion. Usually $\gamma_{2m} \sim \gamma_{23}$. If $\gamma_{21} \geq \gamma_{23}$ (also a very typical situation) and $\Omega_s \geq \Omega_p$, we have $\Gamma_2 \sim \gamma_{21} \gg \gamma_{2m}$. As one can see, varying of the $\Omega_{p,S}$ field intensities may increase Γ_2, the field-induced rate of population transfer to level $|3\rangle$ in comparison with the radiative decay rate γ_{2m}.

IV. DISCUSSION AND COMPARISON WITH EXPERIMENT

The above results can be compared with the experiments performed by Gaubatz *et al.* (see Fig. 3 in comparison with Fig. 10 of Ref. [8]). Analyzing the experimental data ($\tau_{\text{int}} = \tau_{1,2} = 170$ ns , $\tau_N = 12$ ns) we see that the population transfer in a Na_2 molecular beam interacting with two spatially delayed cw laser beams has been performed either in the steady-state regime or in the transient regime, respectively, when $D > 0$ (Ω_p precedes Ω_S) and $D = 0$ (Ω_p and Ω_S are completely overlapped); D describes the displacement between spatially separated Ω_p and Ω_S beams.

In the case $\omega_{ij}\tau_{\text{int}} \gg 1$ the relaxation processes stir DS Ψ_i. However, and it is crucially important that the Ψ_3 state does not contain intermediate level $|2\rangle$. It means that the Ψ_3 state is radiatively stable. Thus, due to spontaneous transition $\Psi_{1,2} \rightarrow \Psi_3$, the system transfers to Ψ_3 in a few lifetimes τ_N, independently of the initial distribution of population.

A. $D < 0$, Ω_S precedes Ω_p

The system transfers directly to the trapped state Ψ_3, which transfers correspondingly to $|3\rangle$, after switching off the interaction. The efficiency of the process is close to $S = 100\%$, and does not depend on the value of one-photon detuning (really, a weak dependence on detuning, related to the violence of the adiabaticity condition should appear). In the experiment $S = 95\%$ under $\Delta = 0$ and it decreases slightly to $S = 75\%$ under $\Delta = 2.5 \times 10^9$ s^{-1}.

B. $D = 0$; $\Delta = 0$

The steady-state requirement $\gamma_{21}\tau_{\text{int}} \gg 1$ is fulfilled. Thus, the system should be located in the Ψ_3 state completely. The subsequent switching off of the interaction would transfer 50% of population into level $|3\rangle$, Eq. (9). (In the experiment $S = 20\%$, the theoretical simulation leads to $S \simeq 25\%$, without taking the decay process into account.)

C. $D = 0$; $\Delta = 2.5 \times 10^9$ s^{-1}

The switching on of the interaction will stir and equally populate only Ψ_1 and Ψ_3 states (since $\Delta\tau_N \gg 1$). Using expression (18) and substituting the experimental data $\gamma_{21}^{-1} = 12$ ns; $\Omega_p = 1.3 \times 10^9$ s^{-1}; $\Omega_S = 0.87 \times 10^9$ s^{-1}; $\Delta = 2.5 \times 10^9$ s^{-1} into Eqs. (10) and (17), one can obtain $|b|^2 \simeq 1/8$; $|V_{1,2}/\Delta|^2 \simeq \frac{1}{4}$; $\omega_{1\rightarrow 3} \simeq 7 \times 10^6$ s^{-1}; $\omega_{1\rightarrow 2} \simeq 8.5 \times 10^5$ s^{-1}. The transition probability $w_{1\rightarrow 2}$ is considerably smaller compared to $w_{1\rightarrow 3}$, which implies that the Ψ_2 state remains empty during the interaction process. When the interaction is switched off the expressions for Ψ_1 and Ψ_3 become identical, i.e., at $t \rightarrow \infty$, $\Psi_1 = \Psi_3 = 2^{-2}$ ($|1\rangle + |3\rangle$). Thus, like the previous case ($\Delta = 0$), the population of level $|3\rangle$, until the end of the interaction, would be also $\simeq 50\%$ (in the experiment $S \simeq 20\%$, the theoretical value is $S \simeq 25\%$).

$D > 0$ (Ω_p precedes Ω_S); $\Delta = 0$

The rates of decay $w_{1\rightarrow 2,1\rightarrow 3} \sim \gamma_{21}$ and full decay to Ψ_3, during the interaction time, has to occur. Switching off the interaction transfers the population from the Ψ_3 state to level $|1\rangle$, leading to $S = 0$ (which completely coincides with the experiment).

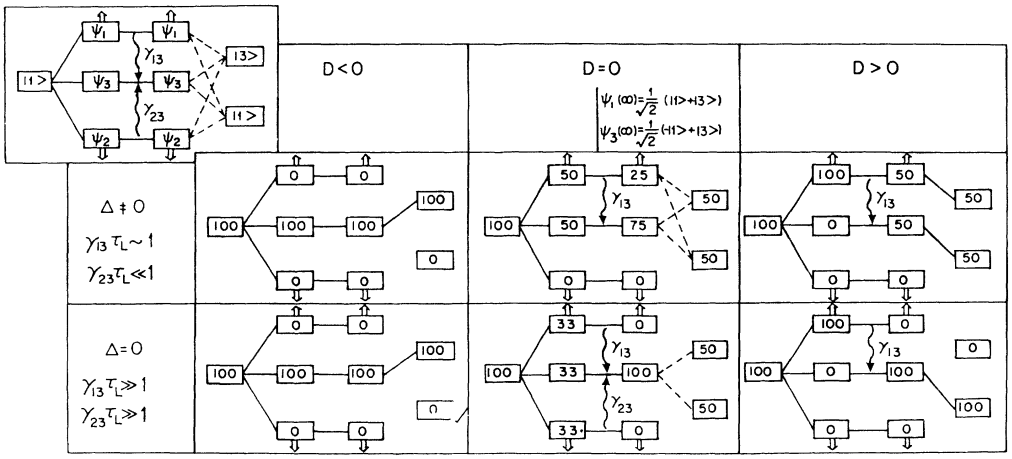

FIG. 3. The flow of the population from initially populated level $|1\rangle$ ($n_0 = 100$) via the states $\Psi_{1,2,3}$ to levels $|1\rangle$ and $|2\rangle$ for characteristic geometric arrangements ($D > 0$, $D = 0$, $D < 0$) as well as characteristic detunings ($\Delta = 0$; $\Delta \neq 0$, $\delta = 0$). The diagrams are explained in the upper left panel. The wavy arrows indicate the radiative decay from $\Psi_{1,2}$ states to the trapped state Ψ_3. The wide straight arrows indicate the leaking out of the population from the three-level system.

E. $D>0$ (Ω_p precedes Ω_S); $\Delta=2.5\times10^9\ s^{-1}$

The system is driven via the Ψ_1 state. Because of long interaction time ($\tau_{int}\sim170$ ns), the Ψ_1 state decays to the Ψ_3 state. The number of atoms remaining in Ψ_1 could be simply estimated, $n(\Psi_1)\sim n_0 e^{-\omega_{1\to3}\tau}$. Using expression (18) and experimental data one can obtain for $\omega_{1\to3}\tau_{int}\sim0.9$, and for $n(\Psi_1)\sim0.5n_0$. According to Eq. (9), in the case $D>0$ Ψ_1 transfers completely to level $|3\rangle$, while Ψ_3 transfers completely to level $|1\rangle$, so $S\simeq50\%$ (in the experiment $S\simeq42\%$, the theoretical value is $S\simeq100\%$).

Resume. The complete agreement of our theoretical calculation with the experimental data [8] in the case $D>0$ and $D<0$, as well as a considerable discrepancy in the case $D=0$, indicates explicitly the remarkable loss of the population $\sim50\%$ from the considered three-level system to some buffer level $|4\rangle$ that is not coupled by fields to the treated system. It should be mentioned that in the case $D=0$, the estimation [8] is not valid because it has been made with a violation of the population-conservation law, $\rho_{11}+\rho_{22}+\rho_{33}=1$. The loss occurs only from the intermediate level $|2\rangle$, i.e., from the $\Psi_{1,2}$ states. (Also in the experiment Fig. 10 in Ref. [8], the existence of such a loss is implied, but is not clearly emphasized.) Therefore in the cases $D>0$, $D<0$ this loss does not affect the population transfer. The strong effect on the results occurs only in the case $D=0$.

V. APPLICATION OF THE PULSED DELAYED-FIELD TECHNIQUE TO REAL EXPERIMENTAL SITUATIONS

The developed theory of the pulsed delayed-field technique has a lot of advantages. It can be used in molecular or atomic bulk, for extremely simple and effective PT. High peak intensity of pulsed lasers may simplify the requirements, which had to be provided for the experimental observation of PT by cw lasers.

Possibility to use the large one-photon detuning. Under typical condition (for ordinary dye laser) the Rabi frequency can be estimated as $\Omega_{p,S}=d_{1,2}E_{p,S}/\hbar\sim1\ cm^{-1}$ if $d_{1,2}$ are atomic transitions and the duration of pulses $\tau_{1,2}\sim20$ ns. Suppose we claim the PT efficiency $S\sim90\%$ is realized. It means the probability of the nonadiabatic transition (12) should be less than $P<0,1$. The estimation for Δ ($\Delta\le\Omega^2\tau_{1,2}/10$) turns out to be as follows: $\Delta\le400\ cm^{-1}$ for atomic dipole transition and $\Delta\le4\ cm^{-1}$ for molecular transition, assuming $d_{mol}/d_{atom}\sim0,1$. In the case of the broadband laser, the time-limited broadening $\tau_{1,2}^{-1}$ has to be substituted by real laser width $\delta\omega_L$. In this case Δ can be estimated as $\Delta\le1\ cm^{-1}$ (if $\delta\omega_L\ge0,1\ cm^{-1}$).

The possibility to operate far from resonance allows one to neglect the influence of such effects as laser-field fluctuations and broadband laser widths. The Doppler-broadening effect is negligible, because of a Raman-type interaction, $\delta\gamma_D=(k_1-k_2)v_0$.

Naturally, the large detuning may involve into the interaction more than three levels. This question is considered in a separate article [23]. The preliminary results have shown that in the four-level system (an additional level can be sited close to the $|2\rangle$ or $|3\rangle$) the effective PT to the chosen level $|3\rangle$ can be achieved, only in the case of counterintuitive sequence of switching (Ω_S precedes Ω_p).

All the advantages mentioned above lead to much easier realization of the PT with delayed-laser pulses in molecular bulk than with cw lasers in molecular beam.

VI. SUMMARY AND OUTLOOK

Our study analyzes the process of delayed-field population transfer both in adiabatic-following and steady-state regimes. We pointed out that the atomic system can be converted to the trapped state due to adiabatic following in a time period shorter than the decay time. Moreover, in the AF regime the complete population transfer occurs for proper time delay, independently of the pulses sequence.

The trapped-state Ψ_3 is shown to be stable both relative to radiative decay and to collisional damping. Therefore, under counterintuitive switching Ω_S precedes Ω_p and both radiative and collisional damping processes do not affect the PT efficiency. The system is transferred to Ψ_3 at the beginning of the interaction and cannot leave it in the future.

The effect of additional level $|m\rangle$ located close to the level $|3\rangle$ on the PT process is considered. The field-induced relaxation rate Γ_2 to the state Ψ_3 being a function of input intensities, may exceed the rate γ_{2m}, and prevent leaking from the three-level system. The theoretical results are shown to satisfy the experimental data.

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