Creation of coherent superposition states in inhomogeneously broadened media with relaxation

Nora Sandor, Joseph S. Bakos, Zsuzsa Sörlei, and Gagik P. Djotyan*

Research Institute for Particle & Nuclear Physics, Budapest, P.O. Box 49, Hungary
**Corresponding author: djotjan@rmki.kfki.hu*

Received June 22, 2011; accepted August 11, 2011;
posted September 22, 2011 (Doc. ID 149398); published October 26, 2011

We propose and analyze a scheme for creation of coherent superposition of metastable states in a tripod-structured atom using frequency-chirped laser pulses with negligible excitation of the atoms. The underlying physics of the scheme is explained using the formalism of adiabatic states. We show that the proposed scheme may be equally efficient in homogeneously and Doppler-broadened media. By numerically solving the master equation for the density matrix operator, we analyze the influence of relaxation processes on the efficiency of the creation of superposition states. We show that the proposed scheme is robust against small-to-medium variations of the laser field's parameters.

OCIS codes: 020.1670, 210.0210, 270.5585.

1. INTRODUCTION

Creation of coherent superposition of quantum states has attracted a great amount of attention in the last decade. The reason is a rich variety of possibilities for practical applications of coherent superposition states in different fields of optical science and technology. Preparation of atoms or molecules in coherent superposition states results in extreme changes in the optical (refractive and absorption) properties of a medium composed of such coherently prepared atoms or molecules. These changes may lead to dramatic variations of the propagation characteristics of laser pulses in such media due, for example, to the effect of electromagnetically induced transparency (see review papers [1–3] and references therein). Other effects and applications relating to the coherently prepared media include inversionless amplification of laser radiation [4], enhancement of the efficiency of nonlinear optical processes [5–7], quantum computing [8], optical information storage and processing using quantum states [9–15], and many others.

There is a large variety of techniques for creation of coherent superposition of atomic states. In a simplest case of a two-level model atom, for example, a coherent superposition of the two states may be created using a resonant laser pulse with the area of the Rabi frequency (integral of the Rabi frequency over time) not equal to an integer number of π. The value of the induced coherence, however, is sensitive even to relatively small variations of this area and to the resonance conditions between the interacting laser field and atomic transition [16].

Other, more robust schemes for creation of coherent superposition states have been proposed in the past decade in atomic and molecular systems with more complicated structures of working levels. Most of these schemes are based on the adiabatic following method, which is robust against small-to-medium variations of the parameters of the laser fields [17–28]. The most-used schemes of the adiabatic following include stimulated Raman adiabatic passage (STIRAP) [17–25], Stark chirped rapid-adiabatic passage (SCRAP) [26–28], and schemes involving frequency modulated (chirped) laser radiation [9,29–37].

The condition of two-photon resonance is crucial for the STIRAP scheme to be effective. Being sensitive to this condition, the STIRAP-based schemes allow negligible excitation of the atomic system. The SCRAP scheme is not too sensitive to the two-photon resonance condition but is accompanied by temporary excitation of the atom. The schemes based on frequency chirped (FC) laser pulses are free of the drawbacks of the other two schemes mentioned. Because of the frequency chirp, they are not sensitive to the resonance conditions (including the two-photon resonance condition) and allow transition of the populations between the metastable states of a multilevel atom without considerable excitation of the atom [30,33,32]. These features of the schemes based on the FC pulses are especially important for the coherent manipulation schemes of inner or (and) translational states of quantum systems, where excitation is prohibited to avoid the decoherence effects caused by the spontaneous decay of the excited states. Such elimination of the excitation in multilevel atoms is achieved through destructive interference of different quantum passes to the excited state.

In [30], a novel scheme of manipulation of atomic populations was proposed and a complete population transfer between metastable states of the Λ-type atom was demonstrated without considerable excitation of the atom by using a single FC laser pulse. This scheme may be considered a supplementary one to the STIPAP scheme. While the condition of Raman resonance is crucial for the STIRAP-based schemes, it *must be violated* in the scheme with a chirped laser pulse of [30] to avoid excitation of the atom. This is a major difference of this scheme compared with that of STIRAP. As our analysis shows, the initial mixture of the bright and dark superposition states when the atom is optically pumped into one of the

0740-3224/11/112785-12$15.00/0

ground states of the Λ-like atom in the scheme of [30] evolves into a completely dark state at the central part of the laser pulse and back to the same mixture of the dark and bright states as in the initial superposition at the end of the interaction but with additional phase difference of π. It results in complete transfer of the atomic population from the initially populated ground state to the other initially empty ground state without considerable population of the excited state. A similar scheme with a single FC pulse but in a tripod-type atom was used in [31] to transfer the atomic populations among the three metastable states and to create a coherent superposition of these states without considerable excitation of the atom. Note that, while the scheme proposed in [32] using STIRAP with one of the pulses having chirped frequency allows creation of an arbitrary coherence through variation of the speed of the chirp, it contains a nonadiabatic coupling between the dressed states and, hence, is not an adiabatic and robust one.

From the point of view of practical applications of the schemes for coherent manipulation of atomic or molecular quantum states, it is important to investigate their applicability in real media with inhomogeneously broadened transition lines typical for atomic gases or atoms in solid-state environments. Another important question to be addressed is the effect of the longitudinal and transverse relaxations on the efficiency of the schemes. Note that, in the case of STIRAP, the effect of dephasing was investigated in [38,39] and recently, in [40]. The influence of the dephasing and inhomogeneous line broadening due to solvent fluctuations on the efficiency of STIRAP in solute molecules was studied in [41].

In this paper, we generalize the scheme proposed in [30,31] by including three FC laser pulses (instead of a single one) and, thus, broadening the possibilities of the control of the atomic populations and created coherences. No time delay between the pulses is included, which reveals another distinction with the STIRAP-based schemes. Since the proposed scheme includes FC laser pulses and does not require strict resonance conditions, it seems to be prospective for applications in inhomogeneously broadened media. To explore the potential possibilities of the scheme, we investigate creation of coherent superposition of metastable (ground) states in a tripod-structured model atom with Doppler-broadened transition lines.

In the proposed scheme, three FC laser pulses, with two of them in Raman resonance and the third out of Raman resonance, perform coherent manipulation of the atomic populations and coherences. Similar to the case of a single FC laser pulse (see [30,31]), the proposed scheme allows suppressing the excitation of the atom to minimize the decoherence effects caused by the spontaneous decay of the excited states.

We investigate in detail the effect of Doppler broadening of transition lines on the efficiency of coherence creation between ground atomic states in the proposed scheme by considering an atomic gas of tripod-structured atoms at different temperatures.

We show that, while the proposed scheme is robust to variations of the parameters of the laser field out of Raman resonance, the value of the created coherence may be controlled by variation of amplitudes (Rabi frequencies) of the two laser pulses in Raman resonance. In the case when these two laser pulses originate from the same laser source, the induced coherence is immune to phase and amplitude fluctuations of these two fields.

The remainder of this paper is organized as follows. In Section 2, we present the mathematical formalism describing the interaction of FC laser pulses with the tripod-structured atom based on the master equations for the density matrix operator, taking into account longitudinal and transverse relaxation processes. In Section 3, creation of the coherent superposition states is discussed using dressed states analysis. The results of the numerical analysis are presented in Section 4. The effect of Doppler broadening of the transition lines in an atomic gas of tripod-structured atoms is analyzed in Section 5. In Section 6, the influence of longitudinal and transverse relaxation processes is discussed on the efficiency of the coherence creation. The robustness of the proposed scheme against variations of parameters of the acting laser pulses is analyzed in Section 7, and the obtained results are summarized in Section 8.

2. MATHEMATICAL FORMALISM

We consider interaction of three laser pulses with a model atom having a tripodlike structure of working levels; see Fig. 1. Each laser field is acting on the corresponding allowed electric-dipole transition in the atom [between a ground state $|n\rangle$ ($n = 1, 2, 3$) and the excited state $|0\rangle$] according to selection of the carrier frequencies or polarizations of the laser fields. Transitions between the ground states are forbidden in the electric-dipole approximation. The electric field strength of the laser field has the form $E(t) = \sum_{i=1}^{3} E_i(t) \times \cos(\int_{-\infty}^{t} \omega_i(t')\mathrm{d}t')$. An identical linear variation in time (linear

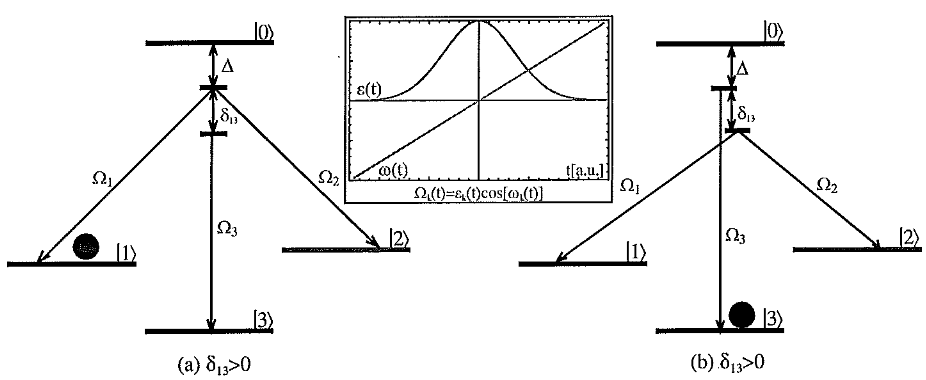


Fig. 1. (Color online) Level structure of the tripodlike atom and the interacting FC laser pulses for the cases of (a) positive and (b) negative Raman detuning and two different initial conditions for the atomic population. The inset shows a Gaussian laser pulse with positive linear chirp.

frequency chirp) of the carrier frequencies of all the interacting laser pulses is assumed in this paper: $\omega_k(t) = \omega_k^{(0)} + \beta t$, where $\omega_k^{(0)}$ is the central carrier frequency of the kth field and β is the speed of the chirp. The central frequency detuning δ_k from the resonance atomic frequency ω_{0k} is $\delta_k = \omega_{0k} - \omega_k^{(0)}$, $k \in \{1,2,3\}$. Since we assume Raman resonance between the fields E_1 and E_2, we have equal single-photon detuning for these waves: $\delta_1 = \delta_2 = \Delta$; see Fig. 1.

Since we are interested in the effect of the relaxation processes on the dynamics of the atomic populations and coherences, we begin with analysis of the master equation for the density operator $\hat{\rho}$ of the system:

$$i\hbar\partial_t\hat{\rho} = [\hat{H},\hat{\rho}] + \hat{R}, \tag{1}$$

where the interaction Hamiltonian in the rotating wave approximation has the following form:

$$\hat{H} = \hbar\begin{pmatrix} 0 & \exp(i\Delta t)\Omega_1 & \exp(i\Delta t)\Omega_2 & \exp(i\delta_3 t)\Omega_3 \\ \exp(-i\Delta t)\Omega_1^* & 0 & 0 & 0 \\ \exp(-i\Delta t)\Omega_2^* & 0 & 0 & 0 \\ \exp(-i\delta_3 t)\Omega_3^* & 0 & 0 & 0 \end{pmatrix}, \tag{2}$$

and the relaxation operator is

$$\hat{R} = i\hbar\begin{pmatrix} -\Gamma\rho_{00} & (-\gamma_{01}+\Gamma/2)\rho_{01} & (-\gamma_{02}+\Gamma/2)\rho_{02} & (-\gamma_{03}+\Gamma/2)\rho_{03} \\ (-\gamma_{01}+\Gamma/2)\rho_{01}^* & \Gamma_1\rho_{00} & -\gamma_{12}\rho_{12} & -\gamma_{13}\rho_{13} \\ (-\gamma_{02}+\Gamma/2)\rho_{02}^* & -\gamma_{12}\rho_{12}^* & \Gamma_2\rho_{00} & -\gamma_{23}\rho_{23} \\ (-\gamma_{03}+\Gamma/2)\rho_{03}^* & -\gamma_{13}\rho_{13}^* & -\gamma_{23}\rho_{23}^* & \Gamma_3\rho_{00} \end{pmatrix}. \tag{3}$$

The time-dependent Rabi frequencies are $\Omega_j = \frac{1}{\hbar}E_j(t)d_{0j} \cdot \exp(i\beta t^2) = W_j f(t)\cdot\exp(i\beta t^2)$, with (in general, different) complex amplitudes W_j $(j = 1,2,3)$. The envelope function $f(t)$ is supposed to be the same for the three laser pulses. d_{0j} is the dipole moment matrix element for transition between the ground state $|j\rangle$ and the excited state $|0\rangle$. We assume that the laser pulses have the same Gaussian shape with duration $\tau_p = \tau_L/(2\ln 2)^{1/2}$, where τ_L is the full width at half-maximum of the pulse (intensity) envelope: $f(t) = \exp[-(t/\tau_p)^2]$. $\Gamma = \Gamma_{01} + \Gamma_{02} + \Gamma_{03}$ is the spontaneous decay rate of the excited state $|0\rangle$, with Γ_{0j} being the spontaneous relaxation rate to the ground state $|j\rangle$; γ_{0j} is the dephasing rate for optical coherence between the states $|0\rangle$ and $|j\rangle$ $(j = 1,2,3)$; and γ_{12}, γ_{13}, and γ_{23} are dephasing rates of coherences between the ground states. The following phase transformation of the density matrix operator: $\rho' = T^{-1}\cdot\rho\cdot T$, with

$$T = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \exp(-i\Delta) & 0 & 0 \\ 0 & 0 & \exp(-i\Delta) & 0 \\ 0 & 0 & 0 & \exp(-i\delta_3) \end{pmatrix},$$

allows us to eliminate the exponential phase factors in the right side of Eq. (1) and in the Hamiltonian in Eq. (2):

$$H' = T^{-1}HT + ih(\partial_t T^{-1})T = \begin{pmatrix} 0 & \Omega_1 & \Omega_2 & \Omega_3 \\ \Omega_1^* & -\Delta & 0 & 0 \\ \Omega_2^* & 0 & -\Delta & 0 \\ \Omega_3^* & 0 & 0 & -\delta_3 \end{pmatrix}. \tag{4}$$

This results in the following set of equations for the density matrix elements:

$$\begin{aligned}
\partial_\tau\rho_{00} &= -i\left(\sum_{i=1}^{3}\tilde{\Omega}_i\rho_{0i}^* - \tilde{\Omega}_i^*\rho_{0i}\right) - \tilde{\Gamma}\rho_{00}, \\
\partial_\tau\rho_{kk} &= i(\tilde{\Omega}_k\rho_{01}^* - \tilde{\Omega}_k^*\rho_{01}) + \tilde{\Gamma}_k\rho_{00}, \quad \text{where} \\
& k \in \{1,2,3\}, \\
\partial_\tau\rho_{01} &= -i[\tilde{\Omega}_2\rho_{12}^* + \tilde{\Omega}_3\rho_{13}^* + \tilde{\Omega}_1(\rho_{11}-\rho_{00}) + \tilde{\Delta}\rho_{01}] \\
&\quad - (\tilde{\Gamma}/2 + \tilde{\gamma}_{01})\rho_{01}, \\
\partial_\tau\rho_{02} &= -i[\tilde{\Omega}_1\rho_{12} + \tilde{\Omega}_3\rho_{23}^* + \tilde{\Omega}_2(\rho_{22}-\rho_{00}) + \tilde{\Delta}\rho_{02}] \\
&\quad - (\tilde{\Gamma}/2 + \tilde{\gamma}_{02})\rho_{02}, \\
\partial_\tau\rho_{03} &= -i[\tilde{\Omega}_1\rho_{13} + \tilde{\Omega}_2\rho_{23} + \tilde{\Omega}_3(\rho_{33}-\rho_{00}) + \tilde{\delta}_3\rho_{03}] \\
&\quad - (\tilde{\Gamma}/2 + \tilde{\gamma}_{03})\rho_{03}, \\
\partial_\tau\rho_{12} &= i[\tilde{\Omega}_2\rho_{01}^* - \tilde{\Omega}_1^*\rho_{02}] - \tilde{\gamma}_{12}\rho_{12}, \\
\partial_\tau\rho_{13} &= i[\tilde{\Omega}_3\rho_{01}^* - \tilde{\Omega}_1^*\rho_{03} + \tilde{\delta}_{13}\rho_{13}] - \tilde{\gamma}_{13}\rho_{13}, \\
\partial_\tau\rho_{23} &= i[\tilde{\Omega}_3\rho_{02}^* - \tilde{\Omega}_2^*\rho_{03} + \tilde{\delta}_{13}\rho_{23}] - \tilde{\gamma}_{23}\rho_{23},
\end{aligned} \tag{5}$$

where a dimensionless time variable $\tau = t/\tau_p$ is introduced and all the parameters are scaled with τ_p (or $1/\tau_p$):

$\tilde{\Gamma}_j = \Gamma_j \tau_p$, $\tilde{\gamma}_{kl} = \gamma_{kl}\tau_p$, and $\tilde{\Delta} = \Delta\tau_p$, $\tilde{\delta}_{13} = (\Delta - \delta_3)\tau_p$ is Raman detuning between the fields with Rabi frequencies $\Omega_{1,2}$ and Ω_3, $\tilde{\Omega}_k(\tau) = \tilde{W}_k \exp(-\tau^2)\exp(\tilde{\beta}\tau^2)$ with $\tilde{W}_k = W_k\tau_p$; $\tilde{\beta} = \beta\tau_p^2$. The diagonal density matrix elements ρ_{jj} ($j = 0,1,2,3$) are the populations of corresponding states; the nondiagonal elements ρ_{kj} ($k \neq j$) are the (complex) coherences between the corresponding states characterizing the coherent superposition of these states.

Before presenting and analyzing the results of the numerical simulation of Eqs. (5), and to gain an insight into the underlying physics of the processes in the atomic system, let us discuss the behavior of the system in the case when the relaxation processes may be neglected, assuming a duration of the pulses that is much shorter than the relaxation times scales.

At this point, it is convenient to perform a transformation from the bare state representation basis for the atomic state vector to the basis of "dark"–"bright" superposition states and perform the dressed states analysis in that basis.

A. Transformation to the "Dark"–"Bright" Superposition States Basis

For the analysis provided below, it is convenient to introduce new basis functions consisting of the following superpositions of the bare atomic states of the tripodlike atom:

$$|db_1\rangle = \frac{W_2^*|1\rangle - W_1^*|2\rangle}{\sqrt{|W_1|^2+|W_2|^2}}, \qquad |db_2\rangle = \frac{W_1|1\rangle + W_2|2\rangle}{\sqrt{|W_1|^2+|W_2|^2}},$$
$$|db_3\rangle = \frac{W_3|3\rangle}{|W_3|}, \qquad |db_4\rangle \equiv |0\rangle. \tag{6}$$

In the new basis, the Hamiltonian of Eq. (4) can be written as

$$\hat{H}_{db} = \hbar\begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & f(t)\sqrt{|W_1|^2+|W_2|^2} \\ 0 & 0 & \delta_{13} & f(t)|W_3| \\ 0 & f(t)\sqrt{|W_1|^2+|W_2|^2} & f(t)|W_3| & 2t\beta+\Delta \end{pmatrix}. \tag{7}$$

$$|\psi(t)\rangle = \sum_k r_k(t)\vec{b}_k(t)\cdot\exp\left(\int_{-\infty}^{t}\lambda_k(t_1)\mathrm{d}t_1\right), \tag{8}$$

with initial condition $|\psi(-\infty)\rangle = \sum_k r_k(-\infty)\vec{b}_k(-\infty)$, where λ_k and $\vec{b}_k$ are the eigenvalues and eigenvectors of the Hamiltonian $\hat{H}_{db}$:

$$\hat{H}_{db}\vec{b}_k = \lambda_k\vec{b}_k. \tag{9}$$

According to the adiabatic theorem (see [42,43]) in the adiabatic regime of interaction $r_k(t) \equiv r_k(-\infty)$, $k \in \{1,2,3,4\}$, which means that, if the atom is in one (or superposition) of its eigenstates initially (before the interaction process, at $t \to -\infty$), it remains in the same eigenstate (or the superposition of the eigenstates) during the interaction process.

We obtain the following equation for the eigenvalues (quasienergies) $\lambda = \lambda_k$ from Eqs. (7)–(9):

$$\begin{aligned}&\lambda(\lambda^3 + \lambda^2(\delta_{13} - 2\beta t - \Delta)\\ &\quad - \lambda[(|W_1|^2+|W_2|^2+|W_3|^2)f(t)^2 + \delta_{13}(2\beta t+\Delta)]\\ &\quad - (|W_1|^2+|W_2|^2)\delta_{13}f(t)^2)\\ &\quad = 0,\end{aligned} \tag{10}$$

and the following expressions for the corresponding dressed eigenvectors $\vec{b}_j$:

$$\vec{b}_k = \begin{pmatrix} 0 & \frac{f(t)\sqrt{|W_1|^2+|W_2|^2}}{\sqrt{N}} & \frac{\lambda_k(\lambda_k-[\Delta+2\beta t])-f(t)^2(|W_1|^2+|W_2|^2)}{f(t)|W_3|\sqrt{N}} & \frac{\lambda_k}{\sqrt{N}}\end{pmatrix}^{(T)},$$
$$k \in \{2,3,4\}. \tag{11}$$

In addition, $\vec{b}_1 = (1 \quad 0 \quad 0 \quad 0)^{(T)}$, with the normalization factor

$$N = \sqrt{f(t)^2(|W_1|^2+|W_2|^2) + \left|\frac{\lambda_k(\lambda_k-[\Delta+2\beta t]) - f(t)(|W_1|^2+|W_2|^2)}{f(t)|W_3|}\right|^2 + \lambda_k^2}.$$

3. DRESSED STATES ANALYSIS

The state vector of the atom in the dark–bright basis can be represented on the basis of the eigenfunctions of the interaction Hamiltonian $\hat{H}_{db}$ as

As it follows from Eq. (11), the following relations take place between the components of the dressed state vector $\vec{b}_k$, $k \in \{2,3,4\}$:

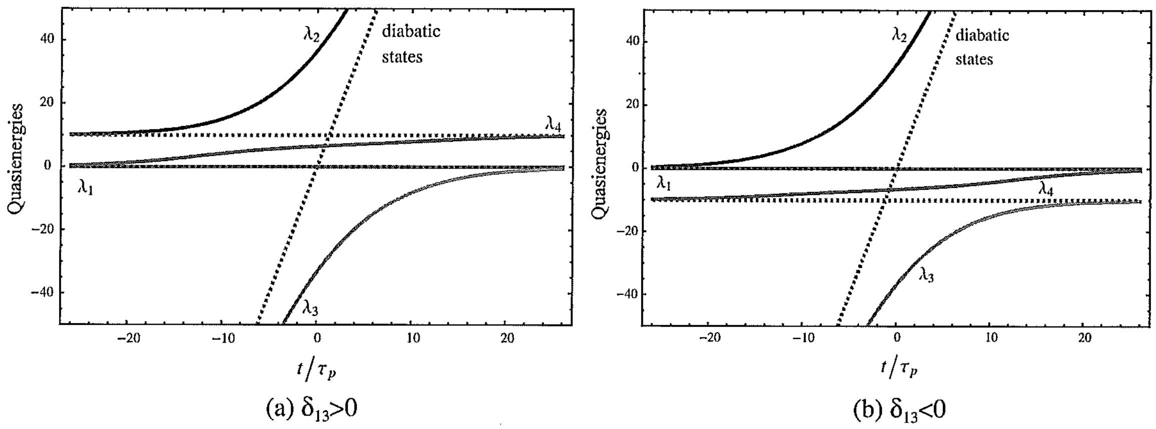


Fig. 2. (Color online) Time dependence of the eigenvalues (quasi-energies) of the Hamiltonian equation [Eq. (14)] in cases of (a) positive and (b) negative Raman detuning; $|\delta_{13}|\cdot\tau_p = 250$ in both cases. All parameters are normalized to the pulse duration τ_p.

$$\frac{b_k^{(3)}}{b_k^{(2)}} = \frac{\lambda_k(\lambda_k - [\Delta + 2\beta t]) - f(t)(|W_1|^2 + |W_2|^2)}{f(t)^2 |W_3| \sqrt{|W_1|^2 + |W_2|^2}},$$
$$\frac{b_k^{(4)}}{b_k^{(2)}} = \frac{\lambda_k}{f(t)\sqrt{|W_1|^2 + |W_2|^2}}. \tag{12}$$

The dynamics of the four eigenvalues (quasi-energies) is shown in Fig. 2 for Gaussian-shaped laser pulses with positive ($\beta > 0$) linear chirp for two cases of Raman detuning δ_{13}: positive ($\delta_{13} > 0$) and negative ($\delta_{13} < 0$).

Below we consider two different cases of creation of coherent superposition states that differ by the initial preparation of the atom (see Fig. 1). In the first case, before the interaction process (at $t \to -\infty$), the entire atomic population is optically pumped into one of the two ground states connected by the pulses in Raman resonance [see Fig. 1(a)]. In the second case, the atom is prepared in the ground state $|3\rangle$ initially, [see Fig. 1(b)]. We will refer to the cases of positive and negative Raman detuning supposing that the atom is prepared in the ground state $|1\rangle$ (positive Raman detuning) or in the state $|3\rangle$ (negative Raman detuning) with the same positive chirp ($\beta > 0$).

While in the first case of ,positive Raman detuning ($\delta_{13} > 0$), at the positive chirp ($\beta > 0$) the pulses in Raman resonance (Ω_1 and Ω_2) reach the single-photon resonances with corresponding transitions earlier than the third pulse (Ω_3) out of Raman resonance, in the second case of negative Raman detuning (and positive chirp), the resonances occur in reversed order. As it will be shown below, in both cases of the initial preparation of the atom, only a negligible (and temporary) excitation of the atom takes place during the interaction.

As it can be seen in Fig. 2, in both cases of positive and negative Raman detuning, there is a quasi-energy of the system (λ_1) that has zero value at the beginning and during the entire interaction process: $\lambda_1 = 0$. This quasi-energy corresponds to the dark superposition state $|db_1\rangle$ [see Eqs. (6)]. At the same time, there is another quasi-energy, λ_4 (see Fig. 2), whose value, in contrast to the other two quasi-energies λ_2 and λ_3, does not depend strongly on the Rabi frequencies and is restricted by the value of the Raman detuning δ_{13}:

$$0 \le |\lambda_4| \le |\delta_{13}|. \tag{13}$$

Let us assume at this point that, before the interaction process, the entire atomic population is distributed between the ground states $|1\rangle$ and $|2\rangle$ and is prepared in the bright superposition $|db_2\rangle$ [see Eqs. (6)]. As it follows from Fig. 2(a), in the case of positive Raman detuning, only the quasi-energy λ_4 coincides at the beginning of the interaction ($t \to -\infty$) with the energy of the (bare) state $|db_2\rangle$; and the corresponding adiabatic (dressed) state $\vec{b}_4$ coincides with the state $|db_2\rangle$. This means that the dressed state $\vec{b}_4$ is one that has to be identified with the assumed initial state vector of the atom in the absence of the laser field. At the end of the interaction, this quasi-energy coincides with the energy of the bare state $|db_3\rangle$, which, with a phase factor (equal to $W_3/|W_3|$), coincides with the bare ground state $|3\rangle$ in the initial (energy) representation. It means that the atomic population of the bright superposition of two ground states $|1\rangle$ and $|2\rangle$ is adiabatically transferred into the third ground state of the atom as a result of interaction with the three FC laser pulses. The same scenario will take place in the case of negative Raman detuning if initially the atom is localized in the state $|3\rangle$, as in the second scheme of coherence creation [see Fig. 2(b)].

In the case of positive Raman detuning, the atom is prepared in the atomic bare state $|1\rangle$, which is a superposition of the dark state $|db_1\rangle$ (which remains unchanged during the interaction), and the bright state $|db_2\rangle$, which evolves to ground state $|3\rangle$. Consequently, the atom ends up in a superposition of the dark state $|db_1\rangle$ and the state $|3\rangle$.

In order to provide coherent population transfer, it is important to avoid excitation of the atom. As it follows from Eqs. (6), the state $|db_4\rangle$ coincides with the excited atomic bare state $|0\rangle$. Considering Eqs. (11)–(13) and taking into account that the contribution of the excited state into the atomic state vector is described by the component $b_4^{(4)}$ of the dressed state vector $\vec{b}_k$ corresponding to the quasi-energy λ_4, the relative contribution of the excited state $|db_4\rangle$ may be estimated as

$$\left|\frac{b_4^{(4)}}{b_4^{(2)}}\right| \le \left|\frac{\delta_{13}}{\sqrt{|W_1|^2 + |W_2|^2}}\right|. \tag{14}$$

As it follows from the obtained relation, the population of the excited state may be suppressed by increasing the amplitudes W_1 or/and W_2 of the Rabi frequencies of the laser pulses in Raman resonance. Obviously, the excited state also remains unpopulated when the system evolves in the $\vec{b}_1 = |db_1\rangle$ dark state.

A. Creation of Coherent Superposition States

Let us now discuss creation of coherent superposition of ground states in the tripodlike atom. Considering the case of the positive Raman detuning along with the positive frequency chirp ($\beta > 0$), let us assume that the population of the atom is initially optically pumped into the one of the ground states, e.g., into the state $|1\rangle$. Taking into account the dark–bright superposition basis of Eqs. (6), the initial atomic state may be written as $|\psi(-\infty)\rangle = |1\rangle = \frac{W_2|db_1\rangle + W_1^*|db_2\rangle}{\sqrt{|W_1|^2+|W_2|^2}}$. For this initial condition, we have for the coefficients $r_k(t) \cong r_k(-\infty)$ describing the statistical weights of the eigenstates in the dressed states representation of the atomic wave function in Eq. (8):

$$r_1 = \frac{W_2}{\sqrt{|W_1|^2+|W_2|^2}}, \qquad r_2 = \frac{W_1^*}{\sqrt{|W_1|^2+|W_2|^2}},$$
$$r_3 = 0, \qquad r_4 = 0. \tag{16}$$

As it was discussed above, the dressed state $\vec{b}_1$, corresponding to the eigenvalue $\lambda_1 = 0$, coincides with the dark state $|db_1\rangle$ and does not change during the interaction. At the same time, as a result of the interaction, the dressed state $\vec{b}_4$ evolves from the bright state $|db_2\rangle$ to the state $|db_3\rangle$, with an additional π phase factor. The resulting state vector will have the following form in the dark–bright superposition basis and the bare states basis:

$$|\psi(t\to+\infty)\rangle = \frac{1}{\sqrt{|W_1|^2+|W_2|^2}}(W_2|db_1\rangle - W_1^*|db_3\rangle)$$
$$= \frac{1}{\sqrt{|W_1|^2+|W_2|^2}} \times \left(\frac{|W_2|^2|1\rangle - W_2W_1^*|2\rangle}{\sqrt{|W_1|^2+|W_2|^2}} - \frac{W_1^*W_3}{|W_3|}|3\rangle\right). \tag{17}$$

As it follows from the obtained equation, the final atomic wave function is a coherent superposition of the three ground states of the tripodlike atom. The contribution of different ground states into the obtained admixture is governed by the Rabi frequencies (intensities) of the two laser pulses in Raman resonance. Note that the excited state is absent in the final atomic wave function. During the interaction process there may be some temporary excitation of the atom, which, however, may be successfully suppressed by increasing the intensity of the pulses in Raman resonance [see Eq. (14)].

A similar consideration may be provided for the case of negative Raman detuning and positive frequency chirp assuming that the tripodlike atom is prepared in the state $|3\rangle$ [see Fig. 1(b)]. In this case, the initial wave function of the atom may be written as $|\psi(-\infty)\rangle = |3\rangle = W_3^*/|W_3||db_3\rangle$, with coefficients $r_k(t) \cong r_k(-\infty)$: $r_1 = 0$, $r_2 = 0$, $r_3 = |W_3|/W_3$, and $r_4 = 0$. In this case, the dressed state $\vec{b}_4$ evolves from the state $|db_3\rangle$ to the state $|db_2\rangle$ with an additional π phase factor, so the final state vector becomes

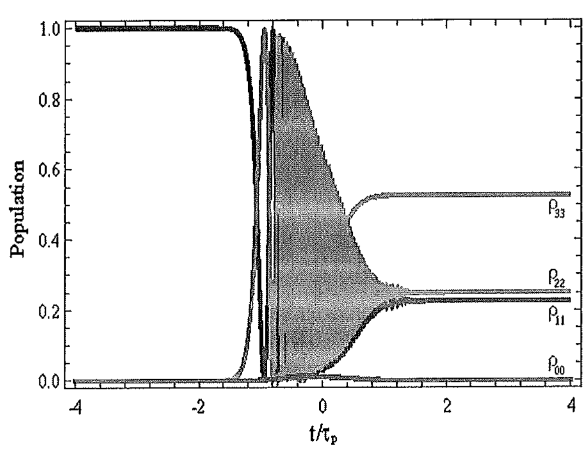
$$|\psi(t\to+\infty)\rangle = -\frac{W_3^*}{|W_3|}|db_2\rangle$$
$$= -\frac{W_3^*}{|W_3|\sqrt{|W_1|^2+|W_2|^2}}(W_1|1\rangle + W_2|2\rangle). \tag{18}$$

As it follows from Eq. (18), a coherent superposition of two ground states connected by the two laser pulses in Raman resonance is created. As in the previous case, the intensities of the pulses in Raman resonance govern the contribution of the two ground states in the coherent superposition. It is worth noting that, if $W_1 = W_2$, a maximum coherence of 0.5 is achieved in this scheme.

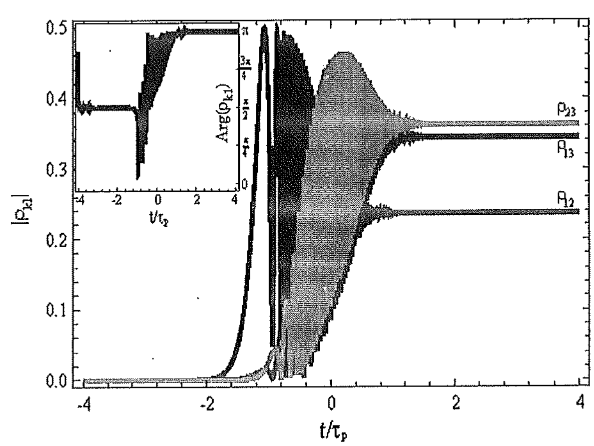
4. RESULTS OF THE NUMERICAL SIMULATIONS

In this section, we analyze the scheme of creation of coherent superposition states by numerical simulation of the system of Eqs. (5) for the density matrix elements. At this point, we take the duration of the pulses shorter than the longitudinal and transverse relaxation times of the atomic system to confirm the conclusions based on the dressed state analysis of Section 3.

The results of the numerical simulations are presented in Figs. 3 and 4 for the cases of positive and negative Raman detuning. As it follows from the numerical solutions, the

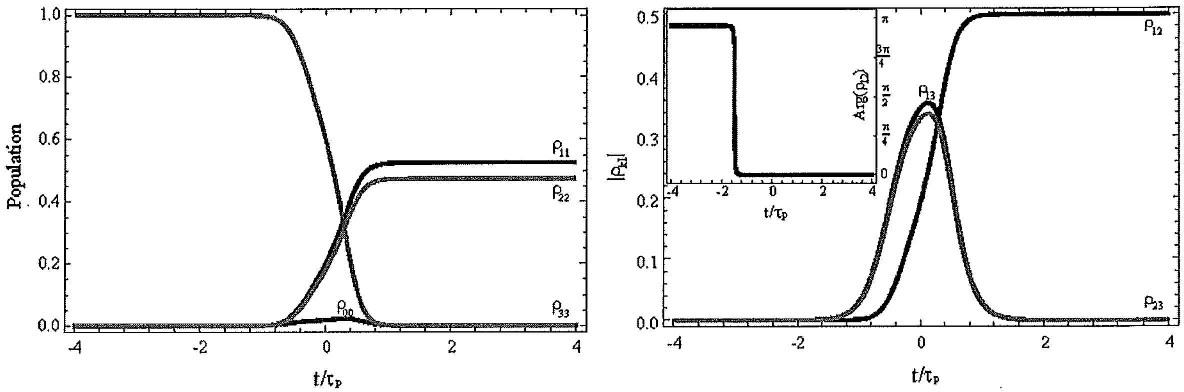


(a) Populations



(b) Coherences

Fig. 3. (Color online) Time evolution of the populations and coherences in the case of positive Raman detuning. Inset: the phase of the coherence ρ_{12}. The parameters applied are $W_1\tau_p = 500$, $W_2\tau_p = 475$, $W_3\tau_p = 525$, $\beta\tau_p^2 = 2500$, and $|\delta_{13}|\tau_p = 250$.



(a) Populations (b) Coherences

Fig. 4. (Color online) Time evolution of the populations and coherences in the case of negative Raman detuning. Inset: the phase of the coherence ρ_{12}. The parameters used are the same as in Fig. 3.

dynamics of the states' populations and coherences confirm the results of Section 3 based on the adiabatic consideration of the problem. Accordingly, in the case of the positive Raman detuning (with initial preparation of the atom in the state $|1\rangle$), all three ground states are populated at the end of the interaction: the population of the bright state is transferred into state $|3\rangle$, and the population of the dark superposition is left intact in the ground state [see Fig. 3(a)]. The time evolution of the atomic (bare) energy eigenstate $|3\rangle$ corresponds to the dynamics of the single dressed state $(\vec{b}_4)$. Consequently, no oscillations occur in its time evolution. In contrast, the bare states $|1\rangle$ and $|2\rangle$ can be described as a superposition of the dressed states $\vec{b}_1$ and $\vec{b}_4$ with related *nonequal* eigenvalues (quasi-energies) λ_1 and λ_4. Because of this superposition of the dressed states, the time evolution of populations of the bare states displays an oscillatory character [see Fig. 3(a)].

In the case of negative Raman detuning, population of the initially populated state $|3\rangle$ is transferred into the bright superposition of the states $|1\rangle$ and $|2\rangle$. The time evolution of the populations in this case is connected to a single dressed state $\vec{b}_4$. As a result, no oscillations occur in the dynamics of the populations [see Fig. 4(a)]. It is important to note that, in both cases, the population of the excited state is negligible (and temporal), reaching merely 1%–2% of the all atomic population.

We obtain the following expressions for the final values of the density matrix elements $\rho_{kl}^{\text{fin}} = \rho_{kl}(t \to +\infty)$ $(k, l = 0, \ldots 3)$ from Eqs. (17) and (18) corresponding to the cases of the positive $(\delta_{13} > 0)$ and negative $(\delta_{13} < 0)$ Raman detuning, respectively:

$$\delta_{13} > 0\colon \rho_{00}^{\text{fin}} = 0, \qquad \rho_{11}^{\text{fin}} = \frac{|W_2|^4}{(|W_1|^2 + |W_2|^2)^2},$$
$$\rho_{22}^{\text{fin}} = \frac{|W_1|^2|W_2|^2}{(|W_1|^2 + |W_2|^2)^2}, \qquad \rho_{33}^{\text{fin}} = \frac{|W_1|^2}{|W_1|^2 + |W_2|^2},$$
$$\rho_{12}^{\text{fin}} = \frac{|W_2|^2 W_1 W_2^*}{(|W_1|^2 + |W_2|^2)^2}, \qquad \rho_{13}^{\text{fin}} = \frac{|W_2|^2 W_1 W_3^*}{|W_3|^2(|W_1|^2 + |W_2|^2)},$$
$$\rho_{23}^{\text{fin}} = \frac{|W_1|^2 W_2 W_3^*}{|W_3|^2(|W_1|^2 + |W_2|^2)}; \tag{19}$$

$$\delta_{13} < 0\colon \rho_{00}^{\text{fin}} = 0, \qquad \rho_{11}^{\text{fin}} = \frac{|W_1|^2}{|W_1|^2 + |W_2|^2},$$
$$\rho_{22}^{\text{fin}} = \frac{|W_2|^2}{|W_1|^2 + |W_2|^2}, \qquad \rho_{33}^{\text{fin}} = 0,$$
$$\rho_{12}^{\text{fin}} = \frac{W_1 W_2^*}{|W_1|^2 + |W_2|^2}, \qquad \rho_{13}^{\text{fin}} = 0, \qquad \rho_{23}^{\text{fin}} = 0. \tag{20}$$

Dependence of the absolute value of the final induced coherence $|\rho_{12}^{\text{fin}}|$ on the ratio of the peak Rabi frequencies of the pulses in Raman resonance is presented in Fig. 5 for both cases of positive and negative Raman detuning. The results show an excellent agreement of the predictions based on the dressed states analysis (solid curves) with results of the numerical simulation of Eqs. (5) for the density matrix elements (dots).

5. CREATION AND CONTROL OF THE COHERENT SUPERPOSITION STATES IN DOPPLER-BROADENED MEDIA

An inhomogeneous (e.g., Doppler) broadening of the atomic transition lines may limit the potential of schemes based on STIRAP to create coherent superposition states. We show in this section that using FC laser pulses in the proposed scheme allows one to create and control the coherent superposition states equally efficiently in homogeneously broadened and Doppler-broadened media.

A Doppler-broadened medium of a gas of tripod-structured atoms is modeled by averaging the created coherence over distribution of the resonance frequencies (velocities) of atoms in the gas at different values of temperature T assuming that all three FC laser pulses propagate in a same direction.

Considering a gas of ^{87}Rb atoms at temperature T and assuming Maxwell–Boltzmann distribution for the velocities of the atoms, we have for the (normalized) probability distribution $P(\tilde{\Delta})$ for an atom to have single-photon detuning $\tilde{\Delta} = \Delta\tau_p$:

$$P(\tilde{\Delta}) = \sqrt{\frac{mc^2}{(2\pi)^3 kT(f_0\tau_p)^2}} \exp\left[-\frac{mc^2(\tilde{\Delta})^2}{8\pi^2 kT(f_0\tau_p)^2}\right], \tag{21}$$

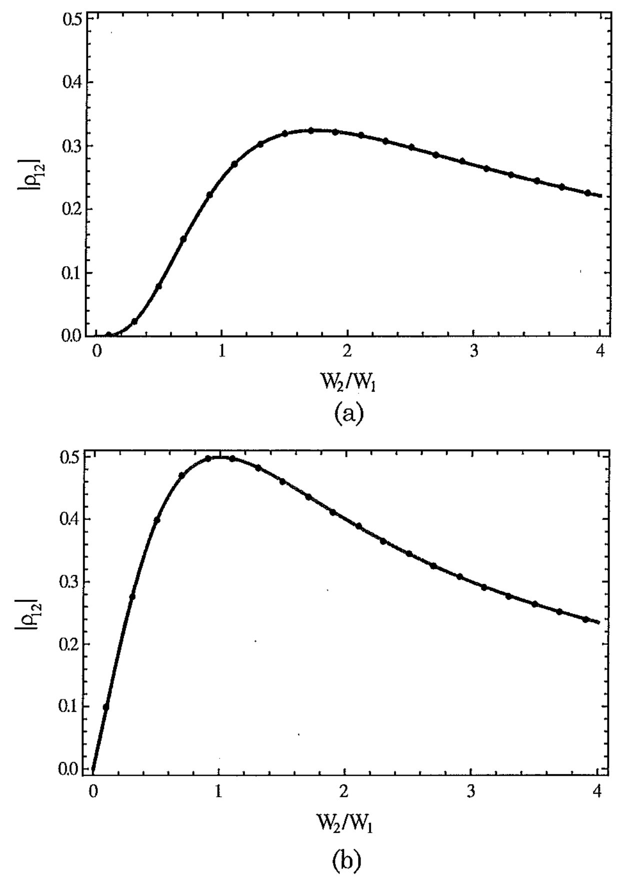


Fig. 5. (Color online) Resulting absolute values of the coherence ρ_{12} versus the ratio of the peak values of the Rabi frequencies W_2/W_1 for the cases of (a) positive and (b) negative Raman detuning calculated from the dressed states analysis. The dots are the results of the numerical solution of the master equation, Eqs. (5), in the absence of relaxation processes. The following values of the parameters are applied: $W_1\tau_p = 250$, $W_3\tau_p = 275$, $\beta\tau_p^2 = 2500$, $|\delta_{13}|\tau_p = 250$, and $W_2\tau_p$ is varying.

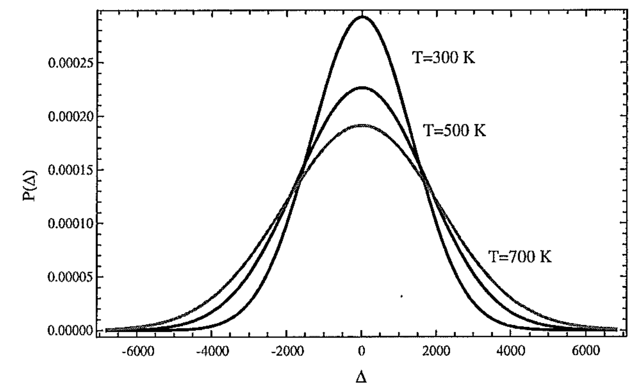


Fig. 6. (Color online) Probability distribution (normalized to unity) for atomic gas at temperatures equal to 300, 500, and 700 K.

where k is the Boltzmann constant, $m = 86.909u$ is the mass of ^{87}Rb (u being the atomic unit) and $f_0 = 384.230$ THz is the frequency distance between the excited and the ground states ($F = 1$ and $F' = 0$ hyperfine states in the D_2 line of ^{87}Rb); see Fig. 6.

The average values of the density matrix elements: populations and coherences $\langle\rho_{kl}\rangle$ ($k, l = 0, 1, 2, 3$) are calculated numerically. First, the master equation [Eqs. (5)] is solved numerically for the values of $\tilde{\Delta}$ corresponding to nonzero probability values [see Eq. (21) and Fig. 6] in order to obtain the density matrix elements $\rho_{kl}^{\mathrm{final}}(\tilde{\Delta})$ at the end of the interaction ($t \to \infty$). The resulting populations and induced coherences are presented in Fig. 7 as functions of the single-photon detuning (Doppler shift) $\tilde{\Delta}$. As it is seen from Fig. 7, there is a range of values of the Doppler shift $\tilde{\Delta}$, where the final coherences (populations) are independent on $\tilde{\Delta}$. This feature is due to the frequency chirping of the laser pulses: as long as the Doppler shift is smaller than the frequency range $[-4\beta\tau_p, 4\beta\tau_p]$ covered by the chirp span during the interaction time (approximately equal to $4\tau_p$), the velocity of motion of the atoms does not have an impact on the resulting population and coherence distribution.

The averaging of the obtained solutions for the Doppler-broadened atomic gas is produced by numerically evaluating the integral

$$\langle\rho_{kl}\rangle = \int_{-\infty}^{\infty} P(\tilde{\Delta})\rho_{kl}^{\mathrm{fin}}(\tilde{\Delta})\mathrm{d}\tilde{\Delta}.$$

The absolute value of the average induced coherence $|\langle\rho_{12}\rangle|$ established after the interaction with the laser field is presented in Fig. 8 as a function of the normalized frequency span $\beta\tau_p^2$ of the laser pulses for different values of the gas temperature. As it can be seen from this figure, the average value of the induced coherence does not depend on the Doppler broadening for sufficiently large frequency span of the pulses during the interaction time due, for example, to a sufficiently high speed of the frequency chirp.

6. EFFECT OF THE RELAXATION PROCESSES

In this section, we discuss the influence of the relaxation processes on the efficiency of creation of the superposition states. As a first step, we analyze the influence of the spontaneous decay of the excited state. The dependence of the states' final populations and of the phase of the created coherence established after the interaction with the laser pulses is shown in Fig. 9 as a function of the longitudinal relaxation rate. One could anticipate a negligible influence of the spontaneous relaxation processes on the populations and coherences of the atom when no considerable excitation of the atom takes place. However, the results of the numerical simulations show that, even for the negligible excitation of the atom, the final populations and induced coherences depend on the longitudinal relaxation rate. The reason is linked to the optical coherences ρ_{0k} ($k = 1, 2, 3$) that are not negligibly small and depend on the longitudinal relaxation rate Γ [see Eqs. (5)]. As can be seen from Fig. 9, the larger the longitudinal relaxation rate (or the longer the laser pulses), the more of the atomic population is transferred (optically pumped) into states $|1\rangle$ and $|2\rangle$ connected by the laser pulses in Raman resonance. The insets in the figures show behavior of the phase of the induced coherence, which implies that, for longer laser pulses, the atom ends up in the dark superposition in both cases of the positive and negative Raman detuning.

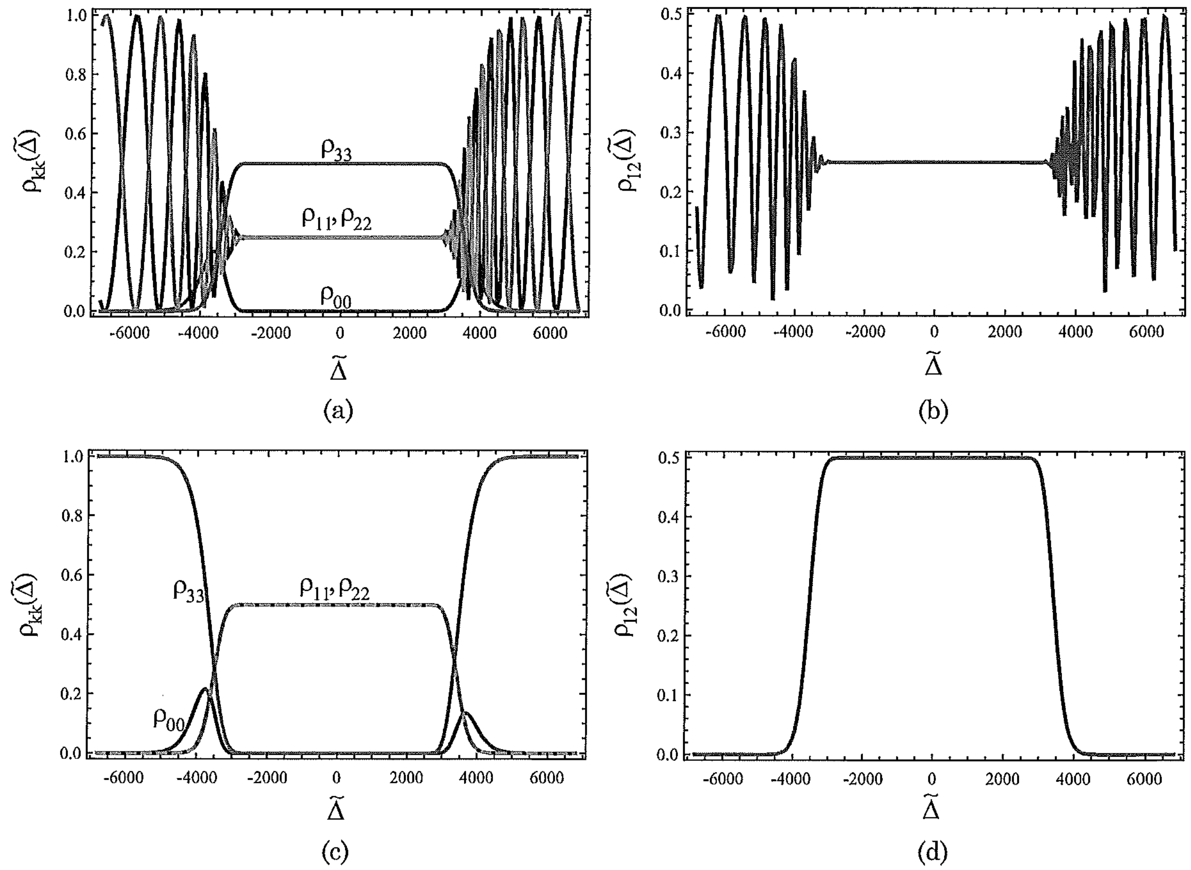


Fig. 7. (Color online) Final populations and coherences established in the atoms at the end of interaction with laser pulses as a function of the normalized single-photon detuning $\tilde{\Delta} = \Delta\tau_p$ for the cases of the (a) and (b) positive and (c) and (d) negative Raman detuning. The applied parameters are $W_1\tau_p = W_2\tau_p = 250$, $W_3\tau_p = 275$ $\beta\tau_p^2 = 1060$, $\delta_{13}\tau_p = 100$, and $\tau_p = 10^{-6}$ s.

Since the quantum interference processes are the basis for the schemes of creation of coherent superposition states, the phase relations between the states' probability amplitudes (the values and the phases of the corresponding coherencies) must play an important role in the considered processes. That is why a strong effect of the transverse relaxation (dephasing) processes may be anticipated on creation and control of the coherent superposition states, as well as on the population transfer between the atomic states.

The final populations of the atomic states as a function of the transverse relaxation rate are presented in Fig. 10 as a result of numerical simulation of the master equation, Eqs. (5). It can be seen from the behavior of the populations that the effect of the transverse relaxation begins to be imperative at $\tau_p \approx 1/(\gamma \cdot 10^{1/2})$. This is true for both cases of the positive and negative Raman detuning. If the interacting pulses are longer, the transverse relaxational processes destroy the adiabatic transfers. At large transverse relaxation rates, all the

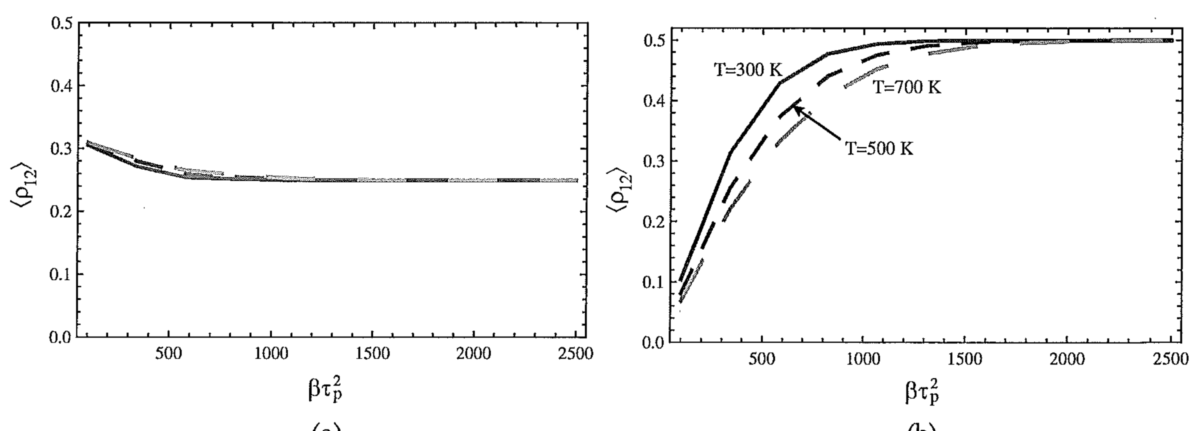
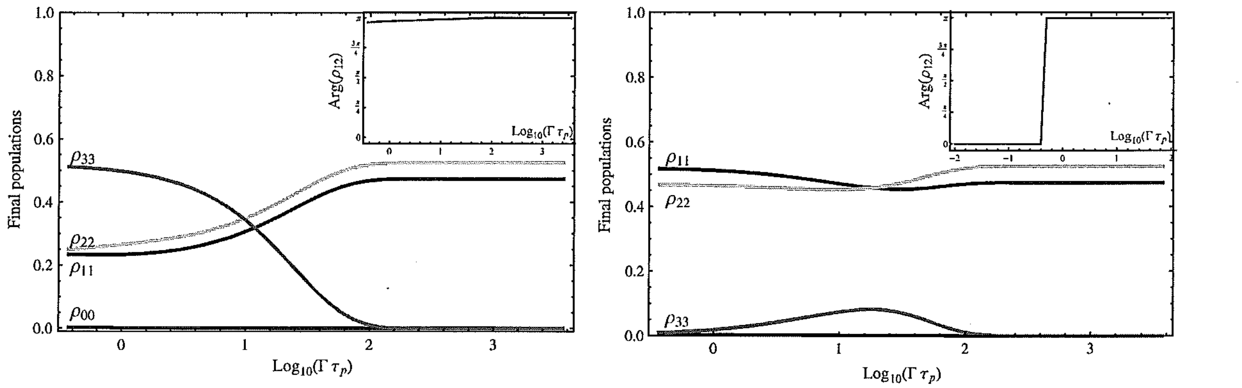
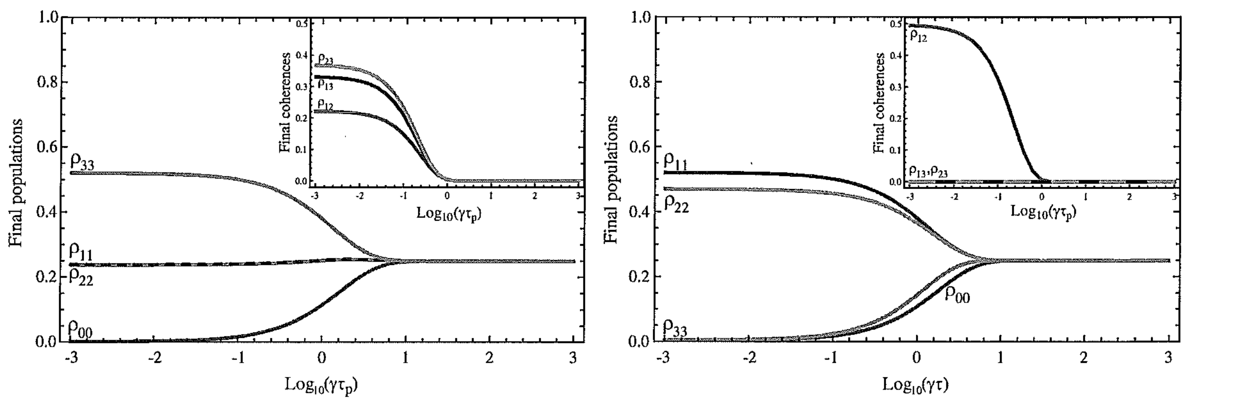


Fig. 8. (Color online) Average value of the final coherence between states $|1\rangle$ and $|2\rangle$ in the cases of (a) $\delta_{13} > 0$ and (b) $\delta_{13} < 0$ as a function of the speed of chirp for the temperatures of the gas equal to 300 (red), 500 (blue), and 700 K (green). The applied parameters are the same as in Fig. 7.



(a) $\delta_{13}>0$ (b) $\delta_{13}<0$

Fig. 9. (Color online) Final populations of the atomic states in the case of (a) positive and (b) negative Raman detuning as a function of the dimensionless longitudinal relaxation rate in the absence of transverse relaxation processes. Insets: the phase of the created coherence ρ_{12}. The parameters used are $W_1\tau_p = 500$, $W_2\tau_p = 475$, $W_3\tau_p = 525$, $\beta\tau_p^2 = 2500$, and $|\delta_{13}|\tau_p = 250$.



(a) (b)

Fig. 10. (Color online) Final populations of the atomic states in the case of (a) positive and (b) negative Raman detuning as a function of the dimensionless transverse relaxation rate in the absence of longitudinal relaxation. Insets: the phase of the created coherence ρ_{12}. The parameters applied are $W_1\tau_p = 500$, $W_2\tau_p = 475$, $W_3\tau_p = 525$, $\beta\tau_p^2 = 2500$, and $|\delta_{13}|\tau_p = 250$.

states are equally populated and there is no coherent superposition created as a result of the interaction (see insets in Fig. 10).

7. ROBUSTNESS OF THE PROCESS

Let us now discuss the robustness of the proposed scheme of creation of superposition states against variation of the main parameters of the laser radiation. As it follows from Fig. 5 and Eqs. (19) and (20), the absolute value of the created coherence $|\rho_{12}|$ depends on the ratio of the peak Rabi frequencies of the laser pulses in Raman resonance and does not depend on the phase relations between the laser pulses. One can control the induced coherence for example, by varying the intensity of one of the laser pulses in Raman resonance, while leaving fixed the intensity of the second pulse in both cases of positive and negative Raman detuning. Note that the induced coherence is robust against changes in the intensity of the laser pulse out of Raman resonance as soon as these changes do not violate the adiabaticity conditions (see, for example, [42,43]).

Robustness of the scheme against variations of the parameters of the pulses in Raman resonance may be provided by utilizing laser pulses from the same source. In this case, at variation δW of the peak Rabi frequencies of the pulses, the variation of the ratio of the two peak Rabi frequencies may be estimated as $\delta(W_1/W_2) \approx W_1/W_2[1 + \delta W(W_2 - W_1)/(W_1 W_2)]$. As it follows from this relation, the dependence of the variation $\delta(W_1/W_2)$ on the parameter δW is weak and, hence, the robustness of the process is especially high in the case of close values of the peak Rabi frequencies of the pulses in Raman resonance: $W_1 \approx W_2$. In this case, the maximum value of the induced coherence $|\rho_{12}|$ equal to 0.5 is achieved for the negative Raman detuning [see Fig. 5(b)]. Note that the considered schemes are also extremely robust against variations of the speed of the chirp, as usually takes place in the case of the adiabatic processes.

8. CONCLUSIONS

In conclusion, we have presented and investigated a scheme for creation of arbitrary coherent superposition of two and three ground states in a tripodlike atom without considerable excitation of the atom using FC laser pulses. The created coherent superposition of the ground states is controlled by the ratio of the peak intensities of the laser pulses in Raman resonance.

We have analyzed the applicability of the scheme in a Doppler-broadened medium of a gas composed of tripod-structured atoms by averaging the induced coherence over the velocity distribution of the atoms in the gas at different temperatures. The results show that the scheme is effective even for relatively large widths of the Doppler-broadened transition lines if the frequency span of the laser pulses due to the chirp exceeds the width of the Doppler broadening.

We have analyzed the influence of the relaxation processes on the population transfer and creation of the superposition states by numerical simulation of the master equation for the density matrix elements. We have shown that the considered scheme allows minimizing the effect of the spontaneous decay of the excited states by suppression of the population of the excited state. However, even under the condition of negligible excitation of the atom, for longer laser pulses, the longitudinal relaxation may influence the induced coherences and the resulting population distribution among the ground states. For laser pulses longer that the decay time of the excited state, optical pumping of the atom by a pair of the pulses in Raman resonance results in accumulation of the atomic population in dark superposition of the ground states linked by the laser pulses in Raman resonance. It is worth noting at this point that the influence of the spontaneous decay may be minimized by increasing the speed of the population transfer. As our analysis (not presented here) shows, it may be achieved by increasing the speed of the frequency chirp, yet in the region where the adiabatic conditions are not violated.

As it may be anticipated, the transverse relaxation (dephasing) has a strong destructive effect in the considered scheme. The numerical simulation of the master equation has shown that, already at duration of the laser pulses close to the dephasing constant of the medium, the transverse relaxation processes destroy the adiabatic transfers. At larger transverse relaxation rates or longer laser pulses, all the states are equally populated as a result of interaction and there is no coherent superposition state created. While the detrimental effect of the transverse relaxation may be avoided by utilizing sufficiently short laser pulses, this effect may also be diminished by increasing the chirp speed of the FC pulses. A detailed analysis of possibilities to minimize the influence of the relaxation processes by varying the speed and form of the frequency chirp will be provided in our upcoming paper.

We have shown that the proposed scheme is robust against small-to-medium variations in the intensities of the laser pulses. A possibility of robust creation of maximum coherence of 0.5 is demonstrated by using two laser pulses in Raman resonance with close peak Rabi frequencies from a common laser source.

The presented adiabatic scheme of coherence creation may find applications in the fields of quantum and nonlinear optics, atomic interferometry, and in mapping and long-term storage of optical information in populations and coherences of metastable atomic states.

ACKNOWLEDGMENTS

This work was funded by the Research Fund of the Hungarian Academy of Sciences (OTKA) under contracts K 68240 and NN 78112, and by the National Development Agency of Hungary under contract ELI-09-1-2010-0010.

REFERENCES

1. M. D. Lukin, "Trapping and manipulating photon states in atomic ensembles," Rev. Mod. Phys. **75**, 457–472 (2003).
2. M. Fleischhauer, A. Imamoglu, and J. P. Marangos, "EIT: optics in coherent media," Rev. Mod. Phys. **77**, 633–673 (2005).
3. P. Král, I. Thanopulos, and M. Shapiro, "Coherently controlled adiabatic passage," Rev. Mod. Phys. **79**, 53–77 (2007).
4. E. Kuznetsova, O. Kocharovskaya, P. Hemmer, and M. O. Scully, "Atomic interference phenomena in solids with a long-lived spin coherence," Phys. Rev. A **66**, 063802 (2002).
5. J. B. Watson, A. Saprera, X. Chen, and K. Burnett, "Harmonic generation from a coherent superposition of states," Phys. Rev. A **53**, R1962–R1965 (1996).
6. M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, "Efficient nonlinear frequency conversion with maximal atomic coherence," Phys. Rev. Lett. **77**, 4326–4329 (1996).
7. M. D. Lukin, P. R. Hemmer, M. Loeffler, and M. Scully, "Resonant enhancement of parametric processes via radiative interference and induced coherence," Phys. Rev. Lett. **81**, 2675–2678 (1998).
8. C. P. Williams and S. H. Clearwater, *Explorations in Quantum Computing* (Springer-Verlag, 1997).
9. G. P. Djotyan, J. S. Bakos, and Zs. Sörlei, "Coherent writing and reading of information using frequency-chirped short bichromatic laser pulses," Opt. Express **4**, 113–120 (1999).
10. G. P. Djotyan, J. S. Bakos, and Zs. Sörlei, "Three-level Λ-atom in the field of frequency-chirped bichromatic laser pulses: writing and storage of optical phase information," Phys. Rev. A **64**, 013408 (2001).
11. A. Mair, J. Hager, D. F. Phillips, R. L. Walsworth, and M. D. Lukin, "Phase coherence and control of stored photonic information," Phys. Rev. A **65**, 031802 (2002).
12. A. E. Kozhekin, K. Molmer, and E. Polzik, "Quantum memory for light," Phys. Rev. A **62**, 033809 (2000).
13. M. Fleischhauer and M. D. Lukin, "Quantum memory for photons: dark-state polaritons," Phys. Rev. A **65**, 022314 (2002).
14. C. Monroe, "Quantum information processing with atoms and photons," Nature **416**, 238–246 (2002).
15. G. P. Djotyan, N. Sandor, J. S. Bakos, and Zs. Sörlei, "Optical phase information writing and storage in populations of metastable quantum states," J. Opt. Soc. Am. B **26**, 1959–1966 (2009).
16. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, 1987).
17. A. S. Parkins, P. Marte, P. Zoller, and H. J. Kimble, "Synthesis of arbitrary quantum states via adiabatic transfer of Zeeman coherence," Phys. Rev. Lett. **71**, 3095–3098 (1993).
18. K. Bergmann, H. Theuer, and B. W. Shore, "Coherent population transfer among quantum states of atoms and molecules," Rev. Mod. Phys. **70**, 1003–1025 (1998).
19. R. Unanyan, M. Fleischhauer, B. W. Shore, and K. Bergmann, "Robust creation and phase-sensitive probing of superposition states via STIRAP with degenerate dark states," Opt. Commun. **155**, 144–154 (1998).
20. H. Theuer, R. G. Unanyan, C. Habscheid, K. Klein, and K. Bergmann, "Novel laser controlled variable matter wave beamsplitter," Opt. Express **4**, 77 (1999).
21. F. Vewinger, M. Heinz, R. G. Fernandez, N. V. Vitanov, and K. Bergmann, "Creation and measurement of a coherent superposition of quantum states," Phys. Rev. Lett. **91**, 213001, (2003).
22. N. Sangouard, S. Guérin, L. P. Yatsenko, and T. Halfmann, "Preparation of coherent superposition in a three-state system by adiabatic passage," Phys. Rev. A **70**, 013415 (2004).
23. R. G. Unanyan, M. E. Pietrzyk, B. W. Shore, and K. Bergmann, "Adiabatic creation of coherent superposition states in atomic beams," Phys. Rev. A **70**, 053404 (2004).
24. Z. Kis, N. V. Vitanov, A. Karpati, C. Barthel, and K. Bergmann, "Creation of arbitrary coherent superposition states by stimulated Raman adiabatic passage," Phys. Rev. A **72**, 033403 (2005).
25. N. V. Vitanov, K. A. Suominen, and B. W. Shore, "Creation of coherent atomic superpositions by fractional stimulated Raman adiabatic passage," J. Phys. B **32**, 4535–4546 (1999).
26. L. P. Yatsenko, B. W. Shore, T. Halfmann, K. Bergmann, and A. Vardi, "Source of metastable H(2s) atoms using the Stark chirped rapid-adiabatic-passage technique," Phys. Rev. A **60**, R4237–R4240 (1999).

27. L. Yatsenko, N. V. Vitanov, B. W. Shore, T. Rickes, and K. Bergmann, "Creation of coherent superpositions using Stark-chirped rapid adiabatic passage," Opt. Commun. **204**, 413–423 (2002).
28. A. A. Rangelov, N. V. Vitanov, L. P. Yatsenko, B. W. Shore, T. Halfmann, and K. Bergmann, "Stark-shift-chirped rapid-adiabatic-passage technique among three states," Phys. Rev. A **72**, 053403 (2005).
29. G. Djotyan, J. Bakos, Zs. Sörlei, J. Szigeti, P. Ignacz, and Z. Toth, "Interaction of a sequence of frequency-chirped bichromatic laser pulses with an ensemble of Λ-atoms: population trapping and coherent optical pumping," Laser Phys. **10**, 355–359 (2000).
30. G. P. Djotyan, J. S. Bakos, and Zs. Sörlei, "Coherent control of atomic quantum states by single frequency-chirped laser pulses," Phys. Rev. A **70**, 063406 (2004).
31. G. P. Djotyan, J. S. Bakos, Zs. Sörlei, J. Szigeti, and D. Dzsotjan, "Creation of a coherent superposition of quantum states by a single frequency-chirped short laser pulse," J. Opt. Soc. Am. B **25**, 166–174 (2008).
32. L. Deng, Y. Nie, X. Yang, J. Shiqi, and S. Gong, "Creation of an arbitrary coherent superposition state with chirped delayed pulses," J. Phys. B **43**, 035401 (2010).
33. J. S. Melinger, S. R. Gandhi, A. Hariharan, J. X. Tull, and W. S. Warren, "Generation of narrowband inversion with broadband laser pulses," Phys. Rev. Lett. **68**, 2000 (1992).
34. S. Chelkowsky, A. D. Bandrauk, and P. B. Corcum, "Efficient molecular dissociation by a chirped ultrashort infrared laser pulse," Phys. Rev. Lett. **65**, 2355–8 (1990).
35. Y. B. Band and O. Magnes, "Chirped adiabatic passage with temporally delayed pulses," Phys. Rev. A **50**, 584–594 (1994).
36. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Population transfer in three-level L atoms with Doppler-broadened transition lines by a single frequency-chirped short laser pulse," J. Opt. Soc. Am. B **17**, 107–113 (2000).
37. V. S. Malinovsky and J. I. Krause, "Efficiency and robustness of coherent population transfer with intense, chirped laser pulse," Phys. Rev. A **63**, 043415 (2001).
38. P. A. Ivanov, N. V. Vitanov, and K. Bergmann, "Effect of dephasing on stimulated Raman adiabatic passage," Phys. Rev. A **70**, 063409 (2004).
39. Q. Shi and E. Geva, "Stimulated Raman adiabatic passage in the presence of dephasing," J. Chem. Phys. **119**, 11773 (2003).
40. C. Lazarou and N. V. Vitanov, "Dephasing effects on stimulated Raman adiabatic passage in tripod configurations," Phys. Rev. A **82**, 033437 (2010).
41. M. Demirplak and S. T. Rice, "Optical control of molecular dynamics," J. Chem. Phys. **116**, 8028–8035 (2002).
42. A. Messiah, *Quantum Mechanics* (North-Holland, 1962), Vol. 2.
43. B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, 1990).