

Population transfer in three-level Λ atoms with Doppler-broadened transition lines by a single frequency-chirped short laser pulse

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We examine interaction of a single frequency-chirped laser pulse with three-level atoms that have a Λ configuration of levels. We show that it is possible to produce complete fast and robust population transfer of all atoms of the ensemble with Doppler-broadened transition lines from one ground state into the other ground state with negligibly small temporary population of the excited state by controlling the intensity of the laser pulse and the direction and speed of the frequency chirp. © 2000 Optical Society of America

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1. INTRODUCTION

In some important applications of the quantum optics, quantum chemistry, mechanical manipulation, and cooling of atoms or molecules by laser radiation it is desirable to produce samples of atoms or molecules whose population resides almost entirely in a particular quantum state. In the main, three effective methods are known for doing so in a coherent way.

The first method is the use of laser pulses with appropriate amplitude–time area, namely, π pulses.¹ For example, a laser pulse whose area (integral of the Rabi frequency over time) is equal to an odd number of π provides full inversion of a two-level atom that is initially in the ground state. This inversion, however, is highly sensitive to parameters of the laser pulse and to the resonance conditions.

The second method of complete population transfer in multilevel quantum systems is based on using the stimulated Raman adiabatic passage (STIRAP) scheme.^{2–6} STIRAP has been demonstrated⁵ (see also Ref. 6 and the references therein) to produce complete population transfer from the initially populated ground state to the second empty ground state without placing appreciable population into the excited state. This scheme of population transfer with frequency-chirped laser pulses (chirped STIRAP) was discussed in Ref. 7, and recently the possibility of coherent population transfer in quantum systems with a laser-induced continuum structure was analyzed in Ref. 8.

The condition of two-photon (Raman) resonance is a crucial one for effective population transfer to take place in the STIRAP and the chirped STIRAP techniques. This condition, however, may not be fulfilled simultaneously for all atoms of an atomic ensemble that has different Doppler shifts of transition lines because of the different motion velocities. Such a problem of controllable population transfer of atoms with Doppler-broadened

transition lines arises, for example, in the field of coherent control and vibrational state shaping in quantum chemistry,^{9–12} in laser manipulation and cooling of atoms (see, e.g., Refs. 13–17), and in other applications of the laser control of atomic or molecular dynamics¹⁸ and has to be investigated in detail.

The influence of the Doppler broadening of transition lines was discussed in Ref. 19 for the STIRAP scheme of population transfer, and it was shown that one can compensate for the effect of Doppler broadening on excitation of a ladder system by increasing the intensity of the pulses. Power broadening of the transition lines seems to be the main reason for such compensation. The Rabi frequency of the intense laser pulses, however, may exceed the frequency difference between adjacent eigenstates, leading to poor selectivity of the population transfer in the STIRAP scheme.

The third scheme for effective population transfer consists of sweeping the laser carrier frequency through resonance with the atomic (molecular) transitions during the laser pulse in such a way that the frequency of the laser radiation is far from resonance at the beginning of the interaction (at the leading edge of the laser pulse); then it is swept through resonance with the atomic transition at the center of the laser pulse when the laser amplitude is maximum and goes out of resonance at the end of the interaction (at the trailing part of the laser pulse). This is the so-called method of adiabatic following or adiabatic passage (AP).¹ There are two important conditions for AP of inversion with frequency-chirped pulses to take place. The first one is that the time variations of the frequency (chirp) and that of the envelope of the laser pulse must be sufficiently slow. The second condition is that the range of the frequency chirp must be larger than the peak Rabi frequency of the laser pulse (for exact conditions for AP to take place, see Ref. 1). One may satisfy both of these conditions by using sufficiently intense laser pulses. It is worth noting that there is no power broad-

ening of transition lines in the AP regime of interaction with the frequency-chirped laser pulses (see, e.g., Ref. 20) and, consequently, there are no losses in the selectivity of the population transfer in the case of high-intensity laser pulses. The AP with frequency-chirped laser pulses was successfully used for generation of narrow-band inversion with broadband picosecond laser pulses with chirped frequencies in Na atoms,²¹ for coherent control and vibrational state shaping in quantum chemistry,⁹⁻¹² and for phonon squeezing²² and for the fast laser cooling and manipulation (deflection and splitting of the beam) of atoms with narrow transition lines when ordinary laser cooling methods cannot be used effectively.¹³⁻¹⁵ Another important application of AP with chirped laser pulses was proposed for fast laser cooling by use of Dicke superradiance.¹⁶

An advantage of using the AP scheme with frequency-chirped pulses is the robustness of this scheme. That is, this scheme is insensitive to the precise duration and shape of the laser pulses as well as to the precise resonance conditions and hence to the Doppler broadening of the atomic lines. The last-named property of the AP scheme with frequency-chirped laser pulses can be used for population transfer in the quantum systems with Doppler-broadened transition lines.

We show in this paper that a single laser pulse with frequency chirp may produce complete population transfer between two ground states in an ensemble of Λ atoms with different Doppler shifts of the transition lines without placing appreciable population in the intermediate excited state. The population transfer takes place in the intuitive order; that is, the frequency of the laser pulse is first swept through resonance with the initially populated ground state and the excited state of a moving atom and then through the transition between the initially empty ground and the excited states. We show that it is possible to avoid appreciable population of the excited state by use of sufficiently intense laser pulses with fast enough frequency chirp under the conditions when the adiabaticity of the population transfer still fulfilled. Unlike in the STIRAP scheme, the increase in the pulse intensity does not lead here to the loss of selectivity. The selectivity of the population transfer survives even when the Rabi frequency exceeds the frequency distance between the two ground states or the width of the Doppler-broadened transition lines of the atomic ensemble. It is worth noting that population transfer in the scheme with a single chirped laser pulse is robust: Moderate variations of the laser pulse shape, the Rabi frequency, and the parameters of the frequency chirp do not affect the process. Note that the negligibly small population of the excited state is temporary (i.e., it tends to the zero value at the end of the laser pulse). This means that the spontaneous decay of this state will be negligibly small for laser pulses with durations shorter than the decay time of the excited level.

When the population of the excited state is comparable in the STIRAP and the single frequency-chirped pulse schemes, the latter scheme seems to be simpler. The other, more important, advantage of the scheme with a single laser pulse is its insensitivity to Doppler broadening of the transition lines (see below). One has only to

use a frequency chirp that covers the width of the Doppler-broadened transition lines to compensate for this broadening and to provide the same population transfer in atoms that belong to different velocity groups. Because the duration of the laser pulse is assumed to be shorter than the decay time of the excited level, and because the population transfer in the AP regime is insensitive to small variations in the intensity, duration, and envelope function of the laser pulse, the population transfer is fast and robust in the scheme under consideration.

The possibility of nearly complete population transfer in a three-level chainwise (ladder) atomic system was shown in Ref. 8 in the AP regime of interaction with single laser pulses whose frequencies are chirped in the counterintuitive manner. The condition of two-photon resonance is crucial here also, just as in the case of STIRAP. It is easy to show, however, that the condition of two-photon resonance can be achieved only for the ladder systems in the case of a single laser pulse. This condition, however, is impossible to realize in the case of the Λ configuration of nondegenerate levels under consideration in this paper (see also Ref. 4).

Our main goal in this paper is to show that population transfer between two ground states of the Λ atom in the single frequency-chirped laser pulse scheme can be produced with nearly the same effectiveness as, for example, in the STIRAP scheme. The main advantage of the single-pulse scheme compared with the schemes of STIRAP and chirped STIRAP is its insensitivity to the Doppler shift of transition lines of the moving atoms.

In Section 2 we present and analyze the equations that describe the interaction of a three-level Λ atom with a single frequency-chirped laser pulse in the dressed-states picture. The results of numerical simulations are presented in Section 3. In Section 4 we discuss the results obtained and present the main conclusions of this paper.

2. MATHEMATICAL FORMALISM

We consider the interaction of a linearly polarized laser pulse with chirped carrier frequency with a three-level Λ atom moving with velocity component v along wave vector \mathbf{k} of the laser pulse; see Fig. 1. The pulse duration is assumed to be much shorter than all relaxation times of the

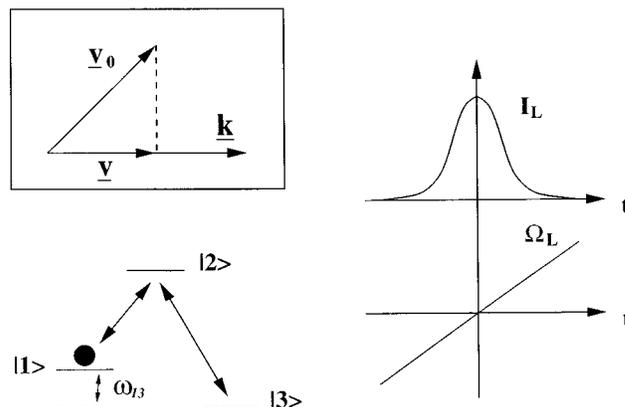


Fig. 1. Three-level Λ atom in the field of a single laser pulse with chirped frequency.

atom and allows us to deal with the Schrödinger equation for the probability amplitudes c_j of the atomic states $|j\rangle$, $j = 1, 2, 3$, (see Fig. 1) instead of with the Bloch equations for the density matrix elements^{23,24}:

$$\begin{aligned}\dot{c}_1 &= i\Omega_{12}(t)c_2 \exp[i\epsilon_{21}(t)t], \\ \dot{c}_2 &= i\{\Omega_{21}(t)c_1 \exp[-i\epsilon_{21}(t)t] \\ &\quad + \Omega_{23}(t)c_3 \exp[-i\epsilon_{23}(t)t]\}, \\ \dot{c}_3 &= i\Omega_{32}(t)c_2 \exp[i\epsilon_{23}(t)t],\end{aligned}\quad (1)$$

where $\Omega_{ij} = \Omega_{ji}^* = (1/2\hbar)d_{ij}A(t)$ ($i, j = 1, 2, 3$) is the Rabi frequency and d_{ij} is the dipole moment matrix element for laser-induced transition from state $|j\rangle$ to state $|i\rangle$ and $A(t)$ is the real envelope of the laser pulse. $\epsilon_{21} = \omega_L(t) - \mathbf{k}\mathbf{v} - \omega_{21}$ and $\epsilon_{23} = \omega_L(t) - \mathbf{k}\mathbf{v} - \omega_{23}$ are the detunings from one-photon resonance, where $\omega_L(t)$ is the time-dependent carrier frequency of the laser pulse, ω_{21} and ω_{23} are the resonant transition frequencies between the corresponding states, and $\mathbf{k}\mathbf{v}$ is the detuning that is due to the Doppler shift of the transition lines of the atom moving with velocity \mathbf{v} . In what follows, we assume linear chirp in time for the laser carrier frequency: $\omega_L(t) = \omega_{L0} + 2\beta t$, where ω_{L0} is the central frequency and 2β is the speed of the chirp.

It is convenient to make the following transformations of the amplitudes:

$$\begin{aligned}c_1 &= a_1, & c_2 &= a_2 \exp[-i\epsilon_{21}(t)t], \\ c_3 &= a_3 \exp[-i(\epsilon_{21}(t) - \epsilon_{23}(t))t].\end{aligned}\quad (2)$$

We can rewrite Eqs. (1) in more-compact form by introducing the column vector $a = (a_1, a_2, a_3)$ and using Eqs. (2):

$$\frac{d}{dt}a = i\hat{H}a, \quad (3)$$

where the Hamiltonian \hat{H} in the rotating-wave-approximation is

$$\hat{H} = \begin{bmatrix} 0 & \Omega_{12} & 0 \\ \Omega_{21} & \epsilon_{21} + \frac{d}{dt}\epsilon_{21} & \Omega_{23} \\ 0 & \Omega_{32} & \omega_{13} \end{bmatrix}. \quad (4)$$

The detuning ϵ_R from the Raman resonance is $\epsilon_R = \epsilon_{21}(t) - \epsilon_{23}(t) = \omega_{23} - \omega_{21} = \omega_{13}$, where $\omega_{13} = (E_1 - E_3)/\hbar$ is the angular frequency interval between two ground states of the atom and E_1 and E_3 are the energies of these states; see Fig. 1. Note that the Raman detuning $\epsilon_R = \omega_{13}$ is a constant here that does not depend on time and on the atomic velocity. The solution $a(t)$ of Eq. (3) can be represented on the basis of the adiabatic dressed states $b^{(k)}(t)$:

$$a(t) = \sum_k r_k(t)b^{(k)}(t) \exp\left[-i\int_{-\infty}^t w_k(t')dt'\right], \quad (5)$$

with the initial condition at $t \rightarrow -\infty$: $a(-\infty) = \sum_k r_k(-\infty)b^{(k)}(-\infty)$, where $b^{(k)}(t)$ is the eigenvector that corresponds to the w_k eigenvalue of the Hamiltonian \hat{H} :

$$\hat{H}b^{(k)} = w_k b^{(k)}. \quad (6)$$

We obtain the following equation for the eigenvalues w_k , using Eqs. (4) and (6):

$$\begin{aligned}w^3 - w^2(\omega_{13} + \epsilon_{21}) + w[\epsilon_{21}\omega_{13} - (|\Omega_{23}|^2 + |\Omega_{12}|^2)] \\ + |\Omega_{12}|^2\omega_{13} = 0.\end{aligned}\quad (7)$$

It reduces to the following equation when the laser field is switched off ($\Omega_{12}, \Omega_{23} \rightarrow 0$):

$$w(\epsilon_{21} - w)(w - \omega_{13}) = 0.$$

A. Case of an Atom at Rest

The time dependencies of the three solutions $w_k^{(0)}$ ($k = 1, 2, 3$) of Eq. (7) form three straight lines in the case of linear frequency chirp assumed in our consideration. These lines, often termed diabatic curves, cross at some points. The lines $w^{(0)}_1 = 0$ and $w^{(0)}_3 = \omega_{13}$ cross the line $w^{(0)}_2(t) = \epsilon_{21}(t)$ but not each other because of the constant Raman detuning $\epsilon_R = \omega_{13}$ in our case of the single laser pulse, as is depicted by dashed lines in Figs. 2(a) and 3(a), which correspond to the case of an atom with zero velocity component ($v = 0$) in direction of laser pulse propagation.

Which of the two lines crosses the third one first depends on the sign of the slope of the line $w_2(t)$ (on the sign of the frequency chirp).

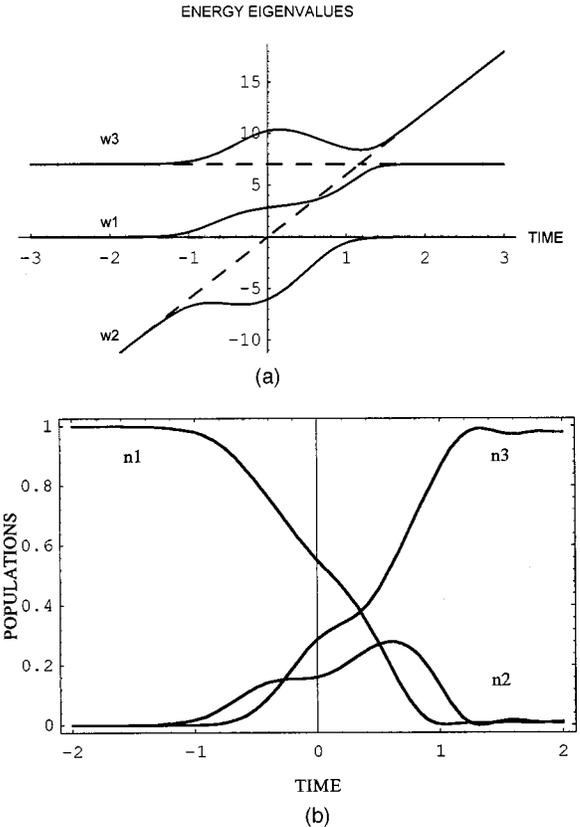


Fig. 2. Time dependence of the energy eigenvalues and the populations of the adiabatic dressed states. Positive chirp: population transfers from one ground state into the other ground state. (a) The adiabatic solution. Dashed lines are the diabatic lines. (b) The result of the numerical simulation. The parameters applied are $\beta\tau_L^2 = 3$, $\Omega_{21}\tau_L = \Omega_{23}\tau_L = 5$, and $\omega_{13}\tau_L = 7$.

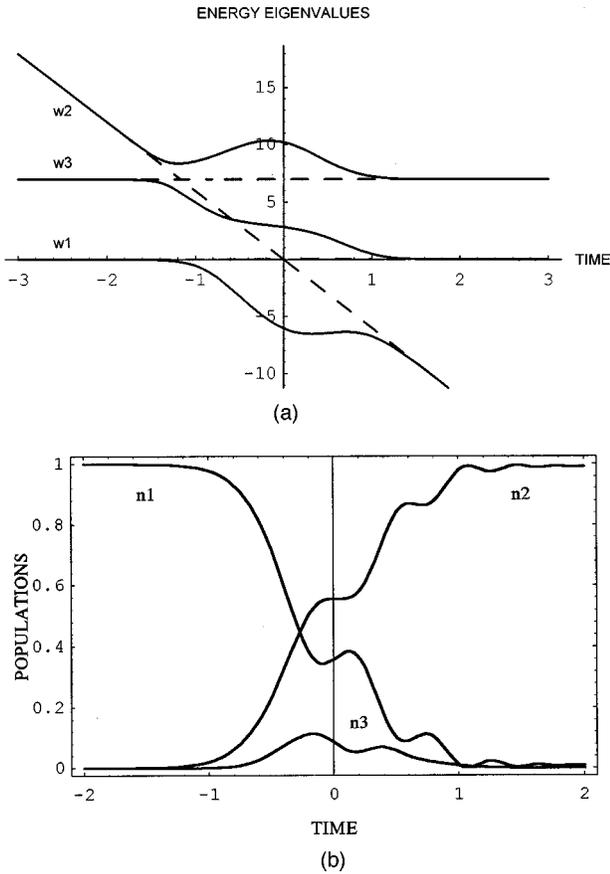


Fig. 3. Time dependence of the energy eigenvalues and the populations of the adiabatic dressed states. Negative chirp; excitation of the atom. (a) The adiabatic solution. Dashed lines are the diabatic lines. (b) The result of the numerical simulation. The parameters applied are $\beta\tau_L^2 = -3$, $\Omega_{21}\tau_L = \Omega_{23}\tau_L = 5$, and $\omega_{13}\tau_L = 7$.

We obtain for the components $b^{(k)}_i$ ($k, i = 1, \dots, 3$) of the adiabatic dressed-state vector $b^{(k)}(t)$ from Eq. (7), using Eq. (5):

$$\begin{aligned}
 b_1^{(k)} &= \frac{\Omega_{12}(w_k - \omega_{13})}{\sqrt{N}}, \\
 b_2^{(k)} &= \frac{w_k(w_k - \omega_{13})}{\sqrt{N}}, \\
 b_3^{(k)} &= \frac{\Omega_{32}w_k}{\sqrt{N}},
 \end{aligned} \tag{8}$$

where the normalization factor N is $N = \Omega_{12}^2(w_k - \omega_{13})^2 + w_k^2(w_k - \omega_{13})^2 + \Omega_{32}^2w_k^2$, with w_k ($k = 1, 2, 3$) being the solution of Eq. (7).

We have to choose that one among the state vectors $b^{(k)}(t)$ that tends to the initial (bare) state vector of the atom in the absence of the laser field at $t \rightarrow -\infty$.

Let us assume that the atom is in the ground state $|1\rangle$ initially (at $t \rightarrow -\infty$) with the state-vector components $a_1(-\infty) = 1$ and $a_2(-\infty) = a_3(-\infty) = 0$. It is easy to show from Eqs. (8) that, only for $w = w_1$, $b^{(1)}_1 \rightarrow 1$, $b^{(1)}_2 \rightarrow 0$, and $b^{(1)}_3 \rightarrow 0$ at $t \rightarrow -\infty$. So dressed state

$b^{(1)}(t)$ is that state that has to be identified with the assumed initial (bare) state of the atom in the absence of the laser field:

$$b^{(1)}(t \rightarrow -\infty) \rightarrow \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} = a(t \rightarrow -\infty).$$

This means that we have to choose the solution $w = w_1$ [$w(t \rightarrow -\infty) = w_1^{(0)}$] of Eq. (7) in the case of the initial conditions assumed. According to the adiabatic theorem,²³ the population of the atom will remain in this dressed state if the conditions for adiabatically slow variation of the laser field have been fulfilled (see, for example, Ref. 1).

As follows from analysis of the solutions of Eq. (7), the solution $w = w_1$ tends to the diabatic line $w^{(0)}_3 = \omega_{13}$ at the end of the laser pulse (at $t \rightarrow \infty$) in the case of positive slope of variation in time of the frequency chirp [$\beta > 0$; Fig. 2(a)] and tends to the diabatic curve $w_2(t) = \epsilon_{21}(t)$ in the case of negative slope of the frequency variation [$\beta < 0$; Fig. 3(a)].

The Raman detuning ω_{13} has been assumed to be positive: $\omega_{13} > 0$ ($\omega_{23} > \omega_{21}$) for both positive and negative β . Note that the time behavior of the eigenvalue $w = w_1$ at $\beta > 0$ mentioned above must be replaced by the behavior that corresponds to the case of $\beta < 0$ and vice versa when $\omega_{13} < 0$.

The solution of the Eqs. (8) that corresponds to the eigenvalue $w = w_3$ is $b^{(3)}(t)$. So, in the case of $\beta > 0$ (when the laser carrier frequency is growing in time), $w_1 \rightarrow w_3^{(0)}$ at $t \rightarrow +\infty$ and, correspondingly,

$$b^{(1)}(t \rightarrow +\infty) \rightarrow b^{(3)}(t \rightarrow +\infty) \rightarrow \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}.$$

This dressed state coincides with the bare ground state $|3\rangle$ at the end of the laser pulse. It means that the population of the atom has been completely transferred from the initially populated ground state $|1\rangle$ to the initially empty ground state $|3\rangle$.

The solution of Eqs. (8) that corresponds to the eigenvalue $w = w_2$ is $b^{(2)}(t)$. We have from Eqs. (8) that $w_1 \rightarrow w^{(0)}_2$ at $t \rightarrow +\infty$ in the case of $\beta < 0$ (when the laser carrier frequency is decreasing in time) and, correspondingly,

$$b^{(1)}(t \rightarrow +\infty) \rightarrow b^{(2)}(t \rightarrow +\infty) \rightarrow \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}.$$

This dressed state coincides with excited (bare) state $|2\rangle$ of the atom at the end of the laser pulse.

So the population of the initially populated state $|1\rangle$ has been transferred into the second ground state $|3\rangle$ in the case of positive frequency chirp ($\beta > 0$ and $\omega_{13} > 0$) when the frequency of the laser has been swept first through resonance with the $|1\rangle \leftrightarrow |2\rangle$ transition and after that through resonance with the transition $|2\rangle \leftrightarrow |3\rangle$. Correspondingly, the diabatic line of the excited state crosses first the diabatic line of the initially populated state and only afterward the diabatic line of the second (initially empty) ground state.

In the case of negative chirping ($\beta < 0$) in the same atom ($\omega_{13} > 0$), the crossing of the diabatic lines takes place in the opposite order: The diabatic line of the excited state crosses first the diabatic line of the initially empty ground state $|3\rangle$ and only after that the diabatic line of the initially populated state $|1\rangle$. The result is excitation of the atom at the end of the laser pulse.

B. Case of Moving Atoms

The above analysis for an atom with zero velocity component ($v = 0$) along the direction of laser pulse propagation can be applied directly to the case of a moving atom with $v \neq 0$. The diabatic line $w = w_2^{(0)} = \epsilon_{21}(t, v)$ is a function of velocity v . In this case, and in crossing the axes of the ordinates in point $w_2^{(0)}(t = 0, v) = \epsilon_{21}(t = 0, v) = -\mathbf{k}\mathbf{v}$, the effective detuning $\epsilon_{\epsilon\phi\phi} = \epsilon_{21} + (d/dt)\epsilon_{21}$ in Eq. (3) is $\epsilon_{\epsilon\phi\phi} = \omega_{L0} - \omega_{21} - \omega_{L0}v/c + 4\beta t(1 - v/c)$ in the case of the moving atom. The effect of the motion of the atom is the change of the frequency detuning $\epsilon_{\epsilon\phi\phi}$ owing to Doppler frequency shift.

In a real experimental situation we deal with an ensemble of atoms that have some distribution of the velocity and hence different resonance frequencies in the laboratory reference frame owing to the Doppler effect. As is shown below in Section 3, the range of the frequency chirping of the laser frequency has to exceed the width of the Doppler-broadened transition lines of the atomic ensemble for achieving complete population in all atoms of the ensemble.

3. RESULTS OF THE NUMERICAL SIMULATIONS

We have numerically solved the set of Eqs. (3) to affirm the results of Section 2 obtained in the model of the adiabatic dressed states. The time variable in our simulations is normalized by the duration τ_L of the laser pulse, whose envelope $A(t)$ is a Gaussian one: $A(t) = A_0 \exp(-t^2/2\tau_L^2)$. The Rabi frequencies and the detunings are normalized by the laser pulse envelope bandwidth $1/\tau_L$.

The complete transfer of the atomic populations from one ground state into the other ground state in the case of positive chirp, or into the excited state in the case of negative chirp, is shown in Figs. 2(b) and 3(b) for an atom that has a zero-velocity component along the direction of laser pulse propagation.

The frequency chirp in our numerical simulations is chosen in such a way that there is resonance with the transition $|1\rangle \leftrightarrow |2\rangle$ at $t \rightarrow 0$ when the Rabi frequency of the laser pulse reaches its maximum value. As the analysis shows, the conditions for adiabatic passage from state $|1\rangle$ to state $|2\rangle$ and further to state $|3\rangle$ are fulfilled well for the positive chirp ($\beta > 0$, $\omega_{13} > 0$) when the atom is in the ground state $|1\rangle$ initially; see Fig. 2(b). In the case of negative chirp ($\beta < 0$, $\omega_{13} > 0$), the frequency of the laser pulse passes first through resonance with the empty transition $|2\rangle \leftrightarrow |3\rangle$ and afterwards with the populated transition $|1\rangle \leftrightarrow |2\rangle$. The temporary population of state $|3\rangle$ takes place after some population of excited state $|2\rangle$ from state $|1\rangle$. At this time, however, the frequency of the laser pulse is far from resonance with the $|2\rangle \leftrightarrow |3\rangle$

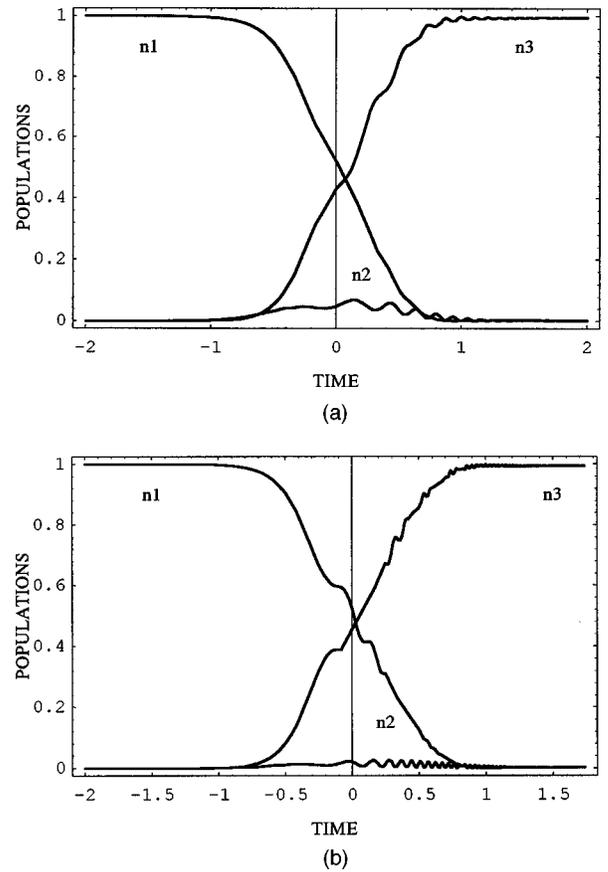


Fig. 4. Time dependence of the atomic populations in the case of higher intensities and faster frequency chirp of the laser pulse. The parameters applied are (a) $\beta\tau_L^2 = 15$, $\Omega_{21}\tau_L = \Omega_{23}\tau_L = 10$, and $\omega_{13}\tau_L = 7$; (b) $\beta\tau_L^2 = 50$, $\Omega_{21}\tau_L = \Omega_{23}\tau_L = 20$, and $\omega_{13}\tau_L = 7$.

transition. It results in disturbance of the AP conditions for the $|2\rangle \leftrightarrow |3\rangle$ transition and leads to Rabi oscillations of the atomic populations of states $|3\rangle$ and $|2\rangle$ [and, hence, to oscillations of the population of state $|1\rangle$; see Fig. 3(b)]. This is the reason for the asymmetry of the time behavior of the atomic populations in Figs. 2(b) and 3(b).

One has to minimize the population of the excited state of the Λ atoms to avoid the loss of coherence owing to the spontaneous decay of the excited state. We may successfully perform this task by increasing the peak intensity of the laser pulses and the speed of the frequency chirp, as we illustrate in Fig. 4, for which a substantial decrease in the value of the temporary population of the excited state has been achieved by an increase of the Rabi frequency and the speed of the chirp (still under the conditions of the AP regime of interaction).

In the case of the moving atoms, the conditions of the AP regime of interaction have to be fulfilled for all atoms of the ensemble for the complete population transfer to be successful for the all-atomic ensemble. The atoms that belong to different velocity groups have different Doppler shifts of the transition lines. This means that resonance with different velocity groups of atoms with the frequency-chirped laser pulse will be achieved at different times and, hence, at different parts of the laser pulse that have different instantaneous values of intensity. This

means that a laser pulse has to be intense enough or the speed of the chirp has to be high enough for the resonances with different velocity groups of atoms take place in the central part of the laser pulse where the conditions of AP are fulfilled.

The dependence of the final population n_3 of the initially empty ground state $|3\rangle$ of an atom moving with velocity component v on the maximum Rabi frequency of the laser pulse is depicted in Fig. 5. As follows from this dependence, a complete population transfer to the initially empty ground state takes place also for atoms whose Doppler frequency shifts are larger than the maximum Rabi frequency of the applied laser pulse.

The dependence of population n_3 on the normalized width of the Doppler-broadened transition line is depicted in Fig. 6. An almost complete population transfer to the initially empty ground state has been shown for all atoms of the atomic ensemble with the full width of the Doppler-broadened transition line three times larger than the maximum Rabi frequency of the laser pulse. Note that the range of the frequency chirping of the laser frequency in Fig. 7 exceeds the width of the Doppler-broadened transition line.

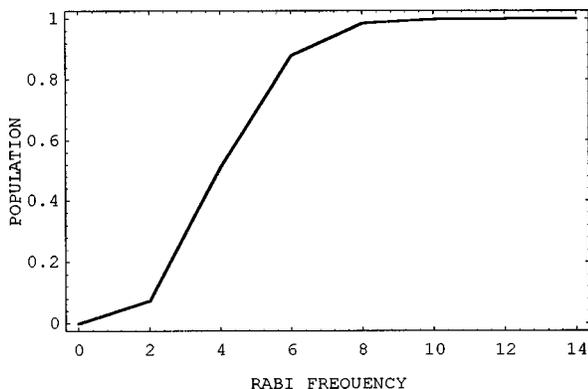


Fig. 5. Dependence of the final population n_3 of the initially empty ground state $|3\rangle$ of an atom moving with normalized velocity $k_L v \tau_L = 30$ on the normalized Rabi frequency $\Omega_R \tau_L = \Omega_{21} \tau_L = \Omega_{23} \tau_L$. The parameters applied are $\omega_{13} \tau_L = 20$ and $\beta \tau_L^2 = 15$.

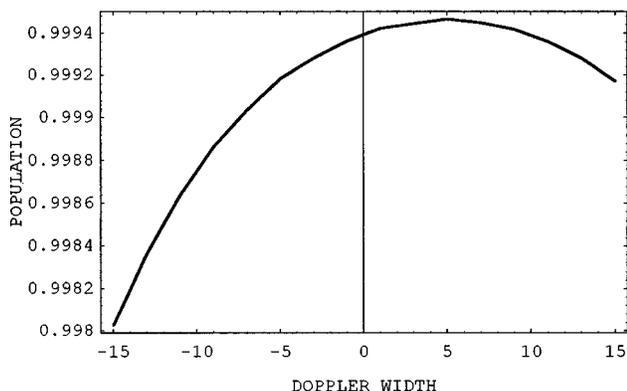


Fig. 6. Dependence of the final population n_3 of the initially empty ground state $|3\rangle$ on the normalized width $k_L v \tau_L$ of the Doppler-broadened transition line. The parameters applied are $\omega_{13} \tau_L = 10$, $\beta \tau_L^2 = 15$, and $\Omega_{21} \tau_L = \Omega_{23} \tau_L = 10$.

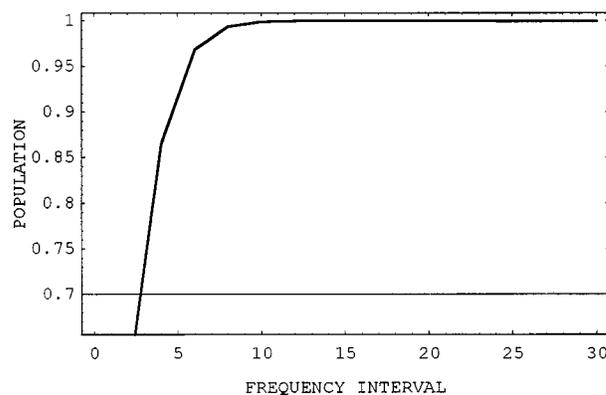


Fig. 7. Dependence of the final population n_3 of the initially empty ground state $|3\rangle$ on the normalized angular frequency distance $\omega_{13} \tau_L$ between two ground states of the Λ atom. The parameters applied are $k_L v \tau_L = 0$, $\beta \tau_L^2 = 15$, and $\Omega_{21} \tau_L = \Omega_{23} \tau_L = 20$.

The spectral selectivity of the population transfer scheme by a single frequency-chirped laser pulse considered in this paper is extremely high. It is illustrated in Fig. 7, where a complete population transfer from a one ground state of the Λ atom into the other one is shown to be successful also for the case when the Rabi frequency is much larger than the frequency distance between the ground states.

4. CONCLUSIONS

In conclusion, the results of analysis of the interaction of a single laser pulse with a linearly chirped frequency with three-level Λ atoms have been presented. We have shown that such a pulse can make a complete population transfer from one ground state of the atom into the other ground state or into the excited state, depending on the direction of the frequency chirp. The population transfer takes place in an intuitive manner. When enough intense laser pulses with enough high-speed frequency chirping are used, the temporary population of the excited state may be negligibly small.

We have shown that the scheme with single-frequency chirped laser pulses is effective for complete population transfer in the atoms that are moving with different velocities and hence have different Doppler shifts of the transition lines. The range of the frequency chirping of the laser frequency in this case has to exceed the width of the Doppler-broadened transition line.

The suggested scheme for population transfer has high spectral selectivity. That is, the complete population transfer from a one ground state of the Λ atom into the second, initially empty, state is successful also when the corresponding Rabi frequency is of order of or even larger than the frequency distance between the ground states. The spectral selectivity survives also when the Rabi frequency is of the order of the width of the Doppler broadening of the transition lines of the atomic ensemble.

It is worth noting that the population transfer in the scheme with a single chirped laser pulse is robust: Moderate variations of the laser pulse shape, the Rabi frequency, and the parameters of the frequency chirp do not affect the process.

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REFERENCES

1. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, New York, 1987).
2. C. E. Carrol and F. T. Hioe, "Three-state system driven by resonant optical pulses of different shapes," *J. Opt. Soc. Am. B* **5**, 1335–1340 (1988).
3. J. R. Kuklinski, U. Gaubatz, F. T. Hioe, and K. Bergmann, "Adiabatic population transfer in a three-level system driven by delayed laser pulses," *Phys. Rev. A* **40**, 6741–6744 (1989).
4. B. W. Shore, K. Bergmann, A. Kuhn, S. Schieman, J. Oreg, and J. H. Eberly, "Laser-induced population transfer in multistate systems: a comparative study," *Phys. Rev. A* **45**, 5297–5300 (1992).
5. U. Gaubatz, P. Rudecki, S. Schieman, and K. Bergmann, "Population transfer between molecular vibrational levels by stimulated Raman scattering with partially overlapping laser fields. A new concept and experimental results," *J. Chem. Phys.* **92**, 5363–5376 (1990).
6. 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).
7. Y. B. Band and O. Manges, "Chirped adiabatic passage with temporally delayed pulses," *Phys. Rev. A* **50**, 584–594 (1994).
8. E. Paspalakis, M. Protopapas, and P. L. Knight, "Population transfer through the continuum with temporally delayed chirped laser pulses," *Opt. Commun.* **142**, 34–40 (1997).
9. B. Armstrup, J. D. Doll, R. A. Sauerbrey, G. Szabó, and A. Lőrincz, "Optimal control of quantum system by chirped pulses," *Phys. Rev. A* **48**, 3830–3836 (1993).
10. M. Sterling, R. Zadoyan, and V. A. Apkarian, "Interrogation and control of condensed phase chemical dynamics with linearly chirped pulses: I_2 in solid Kr," *J. Chem. Phys.* **104**, 6497–6506 (1996).
11. J. Janszky, P. Adam, A. V. Vinogradov, and T. Kobayashi, "Vibrational state shaping for selective laser chemistry," *Chem. Phys. Lett.* **213**, 368–372 (1993).
12. J. Janszky, A. V. Vinogradov, T. Kobayashi, and Z. Kis, "Vibrational Schrödinger-cat states," *Phys. Rev. A* **50**, 1777–1784 (1994).
13. J. S. Bakos, G. P. Djotyan, G. Demeter, and Zs. Sörlei, "Transient laser cooling of two-level quantum systems with narrow natural linewidths," *Phys. Rev. A* **53**, 2885–2888 (1996).
14. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Manipulation of two-level quantum systems with narrow transition lines by short linearly polarized frequency-chirped laser pulses," *J. Opt. Soc. Am. B* **13**, 1697–1705 (1996).
15. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Theory of the adiabatic passage in two-level quantum systems with superpositional initial states," *J. Mod. Opt.* **44**, 1511–1523 (1997).
16. G. P. Djotyan, J. S. Bakos, G. Demeter, and Zs. Sörlei, "Transient cooling of atoms with narrow transition lines using Dicke superradiance," in *Ultracold Atoms and Bose-Einstein Condensation*, Trends in Optics and Photonics (Optical Society of America, Washington, D.C., 1996), pp. 66–71.
17. C. S. Adams and E. Riis, "Laser cooling and trapping of neutral atoms," *Prog. Quantum Electron.* **21**, 1–79 (1997).
18. J. Oreg, F. T. Hioe, and J. H. Eberly, "Adiabatic following in multilevel systems," *Phys. Rev. A* **29**, 690–697 (1984).
19. A. V. Smith, "Numerical studies of adiabatic population inversion in multilevel systems," *J. Opt. Soc. Am. B* **9**, 1543–1551 (1992).
20. C. Liedenbaum, S. Stolte, and J. Reuss, "Inversion produced and reversed by adiabatic passage," *Phys. Rep.* **178**, 3–24 (1989).
21. J. S. Mellinger, S. R. Gandhi, A. Hariharan, J. X. Tull, and W. S. Warren, "Generation of narrowband inversion with broadband laser pulses," *Phys. Rev. Lett.* **68**, 2000–2003 (1992).
22. J. Janszky, T. Kobayashi, and A. V. Vinogradov, "Phonon squeezing in chirped pulse pump and probe experiments," *Opt. Commun.* **76**, 30–33 (1990).
23. P. Meystre and M. Sargent III, *Elements of Quantum Optics* (Springer-Verlag, New York), 1991.
24. A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1962), Vol. II.