Creation of a coherent superposition of quantum states by a single frequency-chirped short laser pulse

G. P. Djotyan,* J. S. Bakos, G. Demeter, Zs. Sörlei, J. Szigeti, and D. Dzsotjan

KFKI Research Institute for Particle and Nuclear Physics, Budapest H-1525, POB 49, Hungary
**Corresponding author: djotjan@rmki.kfki.hu*

Received May 24, 2007; revised November 18, 2007; accepted November 19, 2007;
posted December 5, 2007 (Doc. ID 83012); published January 14, 2008

Traditional schemes for coherent population transfer or generation of coherent superposition states in multilevel atoms or molecules usually utilize two or more laser beams with radiation bandwidth smaller than the frequency interval between the working levels. We show the possibility of creation of the coherent superposition of three metastable states of a four-level atom with tripodlike level structure using a single short frequency-chirped laser pulse. The bandwidth of the pulse envelope (without chirp) must be comparable to or exceed the frequency distance between the two metastable levels. No appreciable excitation of the atom takes place during the creation of the coherent superposition state, thus diminishing significantly the effect of decoherence due to the spontaneous decay of the excited state. The proposed method of creation of superposition states is robust against variations in the laser pulse parameters. Since this method does not require maintaining steady resonance with the atomic transitions (owing to the frequency chirp of the laser pulse), it is effective both in homogeneously and inhomogeneously broadened media.

OCIS codes: 030.1670, 020.4180, 020.7010.

1. INTRODUCTION

Different schemes of coherent population transfer and coherent creation of superposition states have been investigated extensively in recent years [1–9] with a number of new applications in the fields of quantum optics [10], quantum information [11,12], generation of high harmonics and improving efficiency of nonlinear processes in resonant gases [13–18], control of chemical reactions [19], electromagnetically induced transparency [20–28], and others.

There is a well-known technique of coherent generation of superposition states using pulses with temporal area (time integral of the Rabi frequency) equal to $\pi/2$. This technique, similar to the π pulse technique for population transfer between quantum states is sensitive to the laser pulse area, the resonance conditions, and the transverse distribution of the laser beam intensity [29]. A technique of creation and measurement of a coherent superposition of two metastable states based on tripod-Stimulated Raman Adiabatic Passage (STIRAP) and on adiabatic evolution of the superposition states was developed in [2–4]. The drawback of the latter technique is the requirement of two-photon Raman resonance that is necessary to fulfill for the STIRAP scheme to be effective. While this condition may be achieved in atomic beams with narrow velocity distribution of the atoms (small Doppler shifts of the resonance lines), it cannot be fulfilled in inhomogeneously broadened quantum systems such as an ensemble of quantum dots (QD) or rare-earth-metal-ion-doped crystals [30] or when a single laser pulse is used for population transfer and creation of coherent superposition states in an atom with Λ structure of interaction linkages [31,32]. The same is true when a single laser pulse is applied for coherent population transfer between Rydberg states in potassium atoms [33]. Note that an extension of the STIRAP scheme applicable to degenerate levels allowed creation of predefined superposition of multiple magnetic sublevels in [34,35].

The laser pulses are assumed to be narrowband in the majority of the schemes dealing with creation of the coherent superposition of atomic or molecular quantum states or in the schemes of coherent control of populations of the quantum states, i.e., the bandwidth of the laser radiation is assumed to be narrower than the frequency separation between each pair of atomic (molecular) levels involved in the interaction. Usually it is also assumed that each laser pulse involved in the process interacts with a single quasi-resonant transition. The cases of simultaneous interaction of the laser pulse with multiple transitions in the atom due to the Rabi frequency exceeding the frequency spacing between the atomic levels are analyzed in a few papers for intense narrowband laser pulses (see, for example, [36,37] and for creation of coherent superposition of degenerate magnetic sublevels in [4]).

In the case of short laser pulses, the bandwidth of the laser pulse may exceed the spacing between some quantum levels involved in the interaction. This is the case of the short frequency-chirped laser pumping investigated for the Λ-linkage atom in [32].

The traditional adiabatic (dressed) states analysis cannot be applied directly to the quantum system in the case of the short-pulsed laser radiation with the bandwidth exceeding the separation of the involved "close" energy levels of the system, but it may be modified to treat the system of two close levels as a superposition state. This superposition state may be considered on the basis of "bright" and "dark" superpositions of the two states (see, for example, [1] with references therein [7–9]). While the bright component of the superposition effectively interacts with the laser field, the dark one is uncoupled from the laser field and is not affected by the interaction process. As a result of the interaction of the frequency-chirped short-pulsed laser radiation with the Λ-linkage atom in the adiabatic following regime, the bright and dark superposition components are separated from each other. The population of the bright component is removed from the initial state and transferred to the excited state, and the population of the dark component is left intact in the ground state (see [7–9]). This is a robust method for the creation of coherent superposition of metastable states by a single frequency-chirped laser pulse. The drawback of this method, however, is the excitation of the atom leading to the decoherence processes due to spontaneous decay of the excited state. Note that possibilities of population transfer by a single frequency-chirped laser pulse in an atomic system with Λ-interaction linkages were considered earlier in [38], where similarities and differences of producing complete population transfer using π pulses, STIRAP, or adiabatic passage by frequency-chirped laser pulses were examined.

At least two laser pulses are involved in the processes of creation of coherent superposition of metastable states in the majority of the previous considerations (see for example, [1–7,10,22–27,39,40]). The problem of synchronization of multiple pulses in time and space has to be addressed in all previous schemes of creation of coherent superposition states. This is especially important in optically thick media, where the synchronization inevitably will be disturbed during the propagation in the medium. A question arises: can a single laser pulse prepare a coherent superposition of ground states of a multilevel atom without excitation of the atom? While this question is important for simplification of the schemes of creation of coherent superpositions of quantum states, for practical applications creation of such states in a thick (in most cases inhomogeneously broadened) media using a single laser pulse with chirped frequency seems to be a problem of equal or greater importance.

In this paper, we show that such a preparation of the coherent superposition of metastable states $|1\rangle$, $|2\rangle$, and $|3\rangle$ in a four-level atom with tripod structure of the levels (see Fig. 1) is possible using a single short frequency-chirped laser pulse. An important additional condition is that the two states $|1\rangle$ and $|3\rangle$ (termed as close states) among the three metastable states have frequency separation smaller than the bandwidth of the laser pulse envelope, and the frequency distance of the third ("far") metastable state $|2\rangle$ from the two close ones exceeds the bandwidth of the laser pulse envelope. The two close metastable states ($|1\rangle$ and $|3\rangle$) may be considered as "quasi-degenerate" from the point of view of the (Fourier-transform limited) laser pulse having bandwidth exceeding the frequency separation of these states.

We show in this paper that for sufficiently strong laser pulses with the peak Rabi frequency exceeding the largest

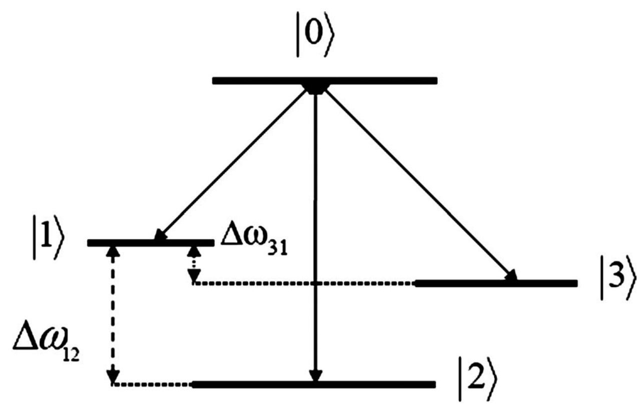

Fig. 1. Scheme of the tripod-linkage atom levels. Levels $|1\rangle$ and $|3\rangle$ are the close levels, and level $|2\rangle$ represents the far level.

frequency separation between the metastable states, the frequency-chirped laser pulse may effectively transfer the bright component of the superposition of the two close metastable states into the third far metastable state without appreciable excitation of the atom, leaving intact the dark component of the superposition of the close states.

If, for example, we assume that the atom is in a superposition of the states $|1\rangle$ and $|3\rangle$ initially, its state vector may be represented as a linear combination of the bright $|d_b\rangle$ and dark $|d_d\rangle$ superposition states: $|\Psi(-\infty)\rangle=\alpha_{01}|1\rangle+\alpha_{03}|3\rangle\equiv\alpha_b|d_b\rangle+\alpha_d|d_d\rangle$, with $|d_b\rangle=\beta_{b1}|1\rangle+\beta_{b3}|3\rangle$ and $|d_d\rangle=\beta_{d1}|1\rangle+\beta_{d3}|3\rangle$. Note, that the coefficients α and β in the case of a single laser pulse interacting with the atomic system depend only on relative values of the dipole moments of corresponding transitions and do not depend on the detuning and speed of the chirp (see below, Section 3). As a result of the interaction, the bright component of the superposition $|d_b\rangle$ will be transferred into state $|2\rangle$, and the original dark superposition $|d_d\rangle$ will be left unaffected by the laser pulse with negligible temporal excitation of the atom during the interaction. At the end of the interaction, a three-state superposition of the metastable states is created: $|\psi(\infty)\rangle=\alpha_d\beta_{1d}|1\rangle+\alpha_b|2\rangle+\alpha_d\beta_{3d}|3\rangle$. This is an effective method for creation of coherent superposition of metastable (ground) states without excitation of the atom, which is robust against variations of the parameters of the laser pulse such as its maximum intensity, duration, or speed of the chirp. Because there is no strict restriction on the resonance conditions due to the frequency chirp of the laser pulse, this method is effective both in homogeneously and inhomogeneously broadened quantum systems.

The remainder of this paper is organized as follows. In Section 2 we present the mathematical formalism for the analysis of the interaction of a frequency-chirped laser pulse with the four-level atom having tripod-linkage structure of the levels. The adiabatic (dressed) states analysis along with numerical simulation of the Schrödinger equation for the probability amplitudes of the states of the tripod-linkage atom with two close and one separated far metastable states in the field of the frequency-chirped laser pulse are presented in Section 3. Creation of dark superposition of the close metastable states, as well as of the coherent superposition of the three metastable states of the tripod-linkage atom without significant excitation of the atom is discussed in Section 4. The results of the investigation are summarized in Section 5.

2. MATHEMATICAL FORMALISM

We will assume that the duration of the laser pulse interacting with the atom is much shorter than any relaxation time of the atomic system, which allows us to deal with the Schrödinger equation for the probability amplitudes of the quantum states of the atom instead of the density matrix elements.

We use the following expansion of the state vector of the tripod-linkage atom (see Fig. 1):

$$|\Psi(t)\rangle = \exp[-iE_1 t/\hbar]\{a_0(t)\exp[-i\omega_L(t)t]|0\rangle + a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle\},$$

where the states $|n\rangle$ $(n=0,1,2,3)$ are the (bare) eigenstates of the atomic Hamiltonian in the absence of the external field, and $a_n(t)$ $(n=0,1,2,3)$ are phase-transformed probability amplitudes of the corresponding atomic states.

From the Schrödinger equation, we have the following equations for the probability amplitudes of the involved states of the atom interacting with a single laser field $E(z,t)=eA(t)\cos(\omega_L(t)-kz)$, where $A(t)$ is the (real) envelope of the laser pulse with chirped carrier frequency $\omega_L(t)=\omega_{L0}+\beta t$:

$$\dot{a}_0 - i a_0 \varepsilon_1(t) = -\frac{i}{2\hbar}A(t)[a_1 d_{10} + a_2 d_{20} + a_3 d_{30}],$$

$$\dot{a}_1 = -\frac{i}{2\hbar}A(t)d_{01}a_0,$$

$$\dot{a}_2 + i\Delta\omega_{21}a_2 = -\frac{i}{2\hbar}A(t)a_0 d_{02},$$

$$\dot{a}_3 + i\Delta\omega_{31}a_3 = -\frac{i}{2\hbar}A(t)a_0 d_{03}, \tag{1}$$

where k is the wavenumber of the wave propagating in the Z direction, ω_{L0} is the central carrier frequency of the laser pulse, and β is the speed of the chirp. $\varepsilon_1(t)=\omega_{L0}-\omega_{10}+2\beta t$ with $\omega_{10}=(E_0-E_1)/\hbar$ being the resonance frequency for transition between the states $|1\rangle$ and $|0\rangle$. E_n $(n=0,1,2,3)$ are energies of the corresponding states, and d_{ij} is the dipole moment matrix element for transition from state $|j\rangle$ to state $|i\rangle$ $(i,j=0,1,2,3)$ (see Fig. 1). The Rabi frequencies induced by the laser radiation are $\Omega_{ij}=\Omega^*_{ij}=(1/\hbar)d_{ij}A(t)$. The Raman detunings $\Delta\omega_{21}$ and $\Delta\omega_{31}$ are equal to the angular frequency intervals between the corresponding ground states: $\Delta\omega_{21}=(E_2-E_1)/\hbar$, $\Delta\omega_{31}=(E_3-E_1)/\hbar$.

The set of equations for the probability amplitudes of quantum states of the tripod-linkage atom was discussed previously in a number of papers for generation of coherent superposition of ground states [2–4], with further applications in the construction of coherent beam splitters for atomic beams [39], and in adiabatic pulse propagation [40]. The condition of two-photon (Raman) resonance is an important condition to be fulfilled for these schemes to be effective. This is one reason why at least two narrowband laser pulses have been applied to generate the coherent superposition between the ground states in the previous investigations.

In the case of a single laser pulse interacting with the tripod-linkage atom, we have a simultaneous interaction of the laser pulse with all allowed transitions in the atom. The condition of two-photon (Raman) resonance cannot be fulfilled in this situation: the corresponding Raman detunings, as is mentioned above, are constants, equal to $\Delta\omega_{21}$ and $\Delta\omega_{31}$ in Eq. (1) representing the frequency separation between the corresponding ground states.

First we analyze the population dynamics of the states of the tripod-linkage atom during action of the frequency-chirped laser pulse with Gaussian envelope based on numerical simulation of Eq. (1). The results are presented in Fig. 2 for the case of the atom being in one of its metastable states ($|1\rangle$) initially. The Rabi frequency of the pulse is $\Omega_{01}(t)=\Omega_0\exp[-t^2/2\tau_p^2]$, with $2\tau_p$ being the duration of the pulse (intensity). In the simulations, we assume for simplicity that the Rabi frequencies are the same for all allowed transitions in the atom $\Omega_{01}=\Omega_{02}=\Omega_{03}$ with Ω_0 being the peak Rabi frequency of the pulse. As could be seen from Fig. 2, a part of the atomic population is transferred from the initially populated state $|1\rangle$ and distributed between the three metastable states $|1\rangle$, $|2\rangle$, and $|3\rangle$ as a result of the interaction with the frequency-chirped laser pulse. An important peculiarity of this population transfer is that only a negligibly small population is being transferred to the excited state of the atom $|0\rangle$ during the interaction. Such behavior is possible only when the bandwidth $\Delta\omega_L$ of the laser pulse envelope (without chirp) is larger than the frequency separation between the two close $|1\rangle$ and $|3\rangle$ states of the atom, but it is smaller than the separation between the states $|1\rangle$ ($|3\rangle$) and $|2\rangle$ (see Fig. 1):

$$\Delta\omega_L \gg |\Delta\omega_{13}|,$$

but

$$\Delta\omega_L < |\Delta\omega_{12}|. \tag{2}$$

Another condition for such population transfer is that the peak Rabi frequency of the laser pulse must be greater than the largest frequency distance between the ground states of the atom: $\Omega_0 \gg |\Delta\omega_{12}|$. The direction of the chirp also plays an important role in the population transfer without excitation of the atom: the resonance must be achieved first with the transition from the initially populated ground state to the excited one and only afterwards with the transitions from the other (initially empty) ground states.

3. ATOMIC POPULATION DYNAMICS: BRIGHT AND DARK SUPERPOSITION STATES AND ADIABATIC (DRESSED) STATES ANALYSIS

To explain the population dynamics in the tripod-linkage atom obtained above by numerical simulation of the

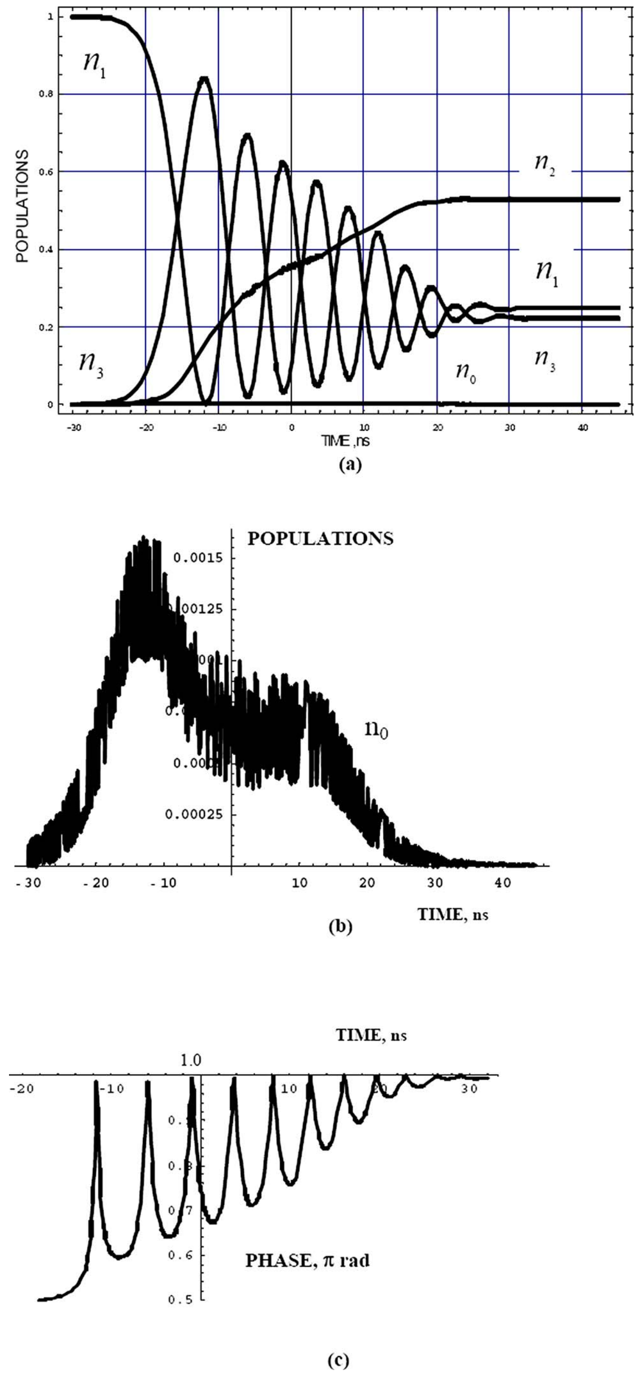

Fig. 2. (Color online) (a) Population dynamics of the states of the tripod-linkage atom during action of the frequency-chirped laser pulse. Parameters applied are duration of the pulse $\tau_{\mathrm{p}}=15$ ns, Rabi frequency for transitions from the metastable states to the excited state (the dipole moments for the allowed transitions are assumed equal to each other for simplicity) is $\Omega_{10}=\Omega_{20}=\Omega_{30}=15$ GHz, and the speed of the frequency chirp $\beta=3$ GHz/ns, the distance $\Delta\omega_{31}$ between the close levels is equal to 10 MHz and the distance between the close and the far levels is equal to 1 GHz. (b) Population of the excited state. (c) Evolution of the phase of the Raman coherence between states $|1\rangle$ and $|3\rangle$ in units of π.

Schrödinger equation, we show in this section that the four-level atom with a tripod structure of the levels may be considered as an equivalent three-state atom with Λ-interaction linkages when the condition [Eq. (2)] takes place: one of the metastable states of such a three-state atom is a superposition state composed of the two close states of the original tripod-linkage atom and the other metastable state and the excited state are the far metastable state and the excited one of the original atom.

The condition [Eq. (2)] allows us to neglect the term $\Delta\omega_{31}a_3$ compared with the term $\dot{a}_3$ in the last equation in Eq. (1): $\Delta\omega_{31} \ll |\dot{a}_3/a_3|$. It is convenient to rewrite Eq. (1) using the basis of the bright $|d_b\rangle$ and dark $|d_d\rangle$ superposition of the states $|1\rangle$ and $|3\rangle$ (see also [7–9]):

$$|d_b\rangle=\frac{1}{\sqrt{1+\gamma_{31}^2}}(|1\rangle+\gamma_{31}|3\rangle), \quad |d_d\rangle=\frac{1}{\sqrt{1+\gamma_{31}^2}}(\gamma_{31}|1\rangle-|3\rangle),$$

where $\gamma_{31}=\Omega_{03}/\Omega_{01}=d_{03}/d_{01}$.

The Schrödinger equation (1) for the probability amplitudes d_b and d_d of these superposition states has a form:

$$\dot{d}_b=-\frac{i}{2}a_0F_1, \quad \dot{a}_2+i\Delta\omega_{21}a_2=-\frac{i}{2}a_0F_2,$$

$$\dot{d}_d=0,$$

$$\dot{a}_0-ia_0\varepsilon_1=-\frac{i}{2}[d_bF_1^*+a_2F_2^*], \tag{3}$$

where $F_1=\Omega_{01}\sqrt{1+\gamma_{31}^2}$, $F_2=\Omega_{02}\gamma_{21}$, and $\gamma_{21}=\Omega_{02}/\Omega_{01}=d_{02}/d_{01}$. Equation (3) is identical to the Schrödinger equation of an equivalent three-level atom with Λ-interaction linkages, one of the metastable states of which is replaced by the bright component of the superposition of the atomic states $|1\rangle$ and $|3\rangle$ of the original tripod-linkage atom. As it follows from Eq. (3), the probability amplitude d_d of the dark superposition of the atomic states $|1\rangle$ and $|3\rangle$ does not change during the interaction. Note that such a behavior of the dark component was obtained when the term proportional to $\Delta\omega_{31}$ was neglected in the last equation of Eq. (1). This term would provide interaction between the dark and bright components forcing the dark state to be "not so dark." However, the first condition in Eq. (2), which may be rewritten in the form $\Delta\omega_{31}\tau_p \ll 1$, means that evolution of the dark state will be negligible under the relatively short time of the action of the laser pulse. This can be seen also in Fig. 2(a), where $\Delta\omega_{31}\tau_p=0.15$. The results of simulations in this case differ slightly from the results corresponding to $\Delta\omega_{31}\tau_p=0$. In the latter case one expects the final population of the far state to be 0.5 and the populations of the close states to be same and equal to 0.25 each for the case of the assumed equal dipole transition elements.

The amplitudes d_b, a_0, and a_2 may be represented as components of a column-state vector $\underset{\sim}{c}=(d_b,a_0,a_2)^T=d_b(1\ 0\ 0)^T+a_0(0\ 1\ 0)^T+a_2(0\ 0\ 1)^T$ of the equivalent effective Λ-linkage atom. In this representation, Eq. (3) will have the following form:

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\underset{\sim}{c}=\hat{H}_b\underset{\sim}{c}, \tag{4}$$

where column vectors $(1\ 0\ 0)^T$, $(0\ 1\ 0)^T$, and $(0\ 0\ 1)^T$ stand for the states $|d_b\rangle$, $|0\rangle$, and $|2\rangle$, respectively (see Fig. 3). The Hamiltonian $\hat{H}_b$ has the following form:

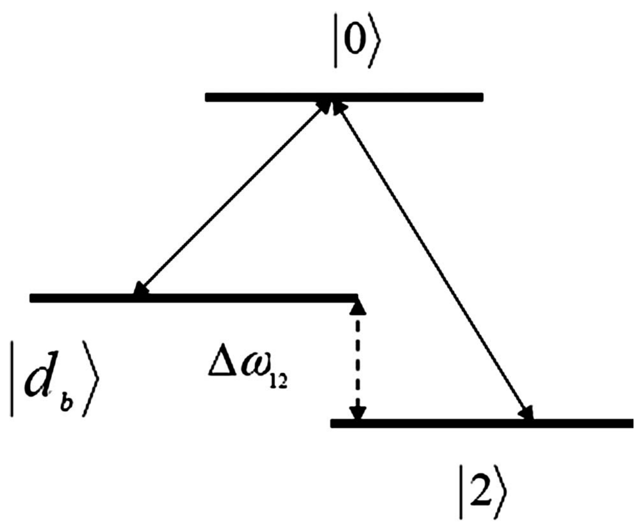

Fig. 3. Structure of the quantum states of the equivalent-linkage atom with one of the ground states, $|d_b\rangle$ being a superposition of the close metastable states of the tripod atom.

$$\hat{H}_b = -\hbar \begin{bmatrix} 0 & F_1/2 & 0 \\ F_1^*/2 & \varepsilon_1 & F_2^*/2 \\ 0 & F_2/2 & -\Delta\omega_{21} \end{bmatrix}. \tag{5}$$

The solution of Eq. (4) can be represented on the basis of the adiabatic states $\underset{\sim}{b}^{(k)}(t)$:

$$\underset{\sim}{c}(t) = \sum_k r_k(t)\underset{\sim}{b}^{(k)}(t)\exp\left[-i\int_{-\infty}^{t} w_k(t')\mathrm{d}t'/\hbar\right], \tag{6}$$

with the initial condition at $\mathbf{t}\to-\infty$:

$$\underset{\sim}{c}(-\infty) = \sum_k r_k(-\infty)\underset{\sim}{b}^{(k)}(-\infty), \tag{7}$$

where $\underset{\sim}{b}^{(k)}(t)$ is the adiabatic-state eigenvector, corresponding to the kth adiabatic eigenvalue (quasienergy) w_k of the Hamiltonian $\hat{H}_b$:

$$\hat{H}_b\underset{\sim}{b}^{(k)} = w_k\underset{\sim}{b}^{(k)}. \tag{8}$$

The quantity $r_k(t)$ in Eq. (7) is the probability amplitude of the adiabatic-state vector $\underset{\sim}{b}^{(k)}(t)$. According to the adiabatic theorem [41], $r_k(t)\equiv r_k(-\infty)$.

We obtain the following equation for the quasienergies w_k, and the components $b_i^{(k)}$ of the corresponding dressed eigenstates using Eqs. (5)–(8):

$$w^3 + w^2\hbar[\Delta\omega_{21} - \varepsilon_1] - w\hbar^2[\varepsilon_1\Delta\omega_{21} + (|F_1|^2 + |F_2|^2)] - \hbar^3|F_1|^2\Delta\omega_{21} = 0, \tag{9}$$

$$b_1^{(k)} = \frac{F_1\hbar(w_k + \hbar\Delta\omega_{21})}{\sqrt{N}}, \quad b_2^{(k)} = \frac{w_k(w_k + \hbar\Delta\omega_{21})}{\sqrt{N}}, \quad b_3^{(k)} = \frac{F_2\hbar w_k}{\sqrt{N}}, \tag{10}$$

where the normalization factor is

$$N = F_1^2\hbar^2(w_k + \hbar\Delta\omega_{21})^2 + w_k^2(w_k + \hbar\Delta\omega_{21})^2 + F_2^2\hbar^2 w_k^2.$$

The dynamics of the three quasienergies w_k ($k=1,2,3$), the solutions of Eq. (9), is shown in Fig. 4 for a laser pulse with Gaussian envelope $A(t)$ and a positive (from red to blue) linear frequency chirp: $\omega_L(t)=\omega_{L0}+\beta t$, $\beta>0$. As it can be seen, the value of one of the quasienergies referred to as w_1 is restricted between zero and the value of the frequency distance $\Delta\omega_{21}$ between the ground states $|d_b\rangle$ and $|2\rangle$ of the equivalent Λ-linkage atom independently of the value of the Rabi frequency of the laser pulse:

$$0 \leqslant w_1 \leqslant \hbar|\Delta\omega_{21}|. \tag{11}$$

Let us assume at this point that the atom is in the metastable state $|d_b\rangle$ initially (at $t\to-\infty$) with the state vector components $d_b(-\infty)=1$ and $a_0(-\infty)=a_2(-\infty)=0$: the initial state vector $\underset{\sim}{c}(t\to-\infty)=(1\ 0\ 0)^T$. As follows from Eqs. (9) and (10), only for $w=w_1$ will $b_1^{(1)}\to1$, $b_2^{(1)}\to0$, and $b_3^{(1)}\to0$ at $t\to-\infty$. So the adiabatic state

$$\underset{\sim}{b}^{(1)}(t) = b_1^{(1)}(1\ 0\ 0)^T + b_2^{(1)}(0\ 1\ 0)^T + b_3^{(1)}(0\ 0\ 1)^T \tag{12}$$

is the one that has to be identified with the assumed initial-state vector of the atom in the absence of the laser field: $\underset{\sim}{b}^{(1)}(t\to-\infty)=\underset{\sim}{c}(t\to-\infty)$.

According to the adiabatic theorem [41], the population of the atom will remain in this adiabatic state if the conditions of the adiabaticity are fulfilled (see [1]): the laser pulse has to act on the atom in a way that eliminates transitions between different dressed states. In the case under consideration the laser pulse envelope has to be sufficiently long for its bandwidth (without chirp) to be smaller than the frequency interval $\Delta\omega_{21}$ between the two metastable states $|d_b\rangle$ and $|2\rangle$; see Eq. (2). We have the following relations between the adiabatic-state vector components $b_i^{(k)}$ from Eq. (10):

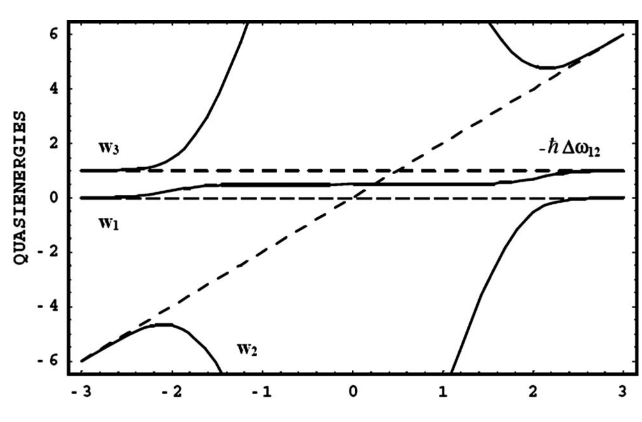

Fig. 4. Evolution of the quasienergies of the equivalent Λ-linkage atom during the action of the frequency-chirped laser pulse corresponding to the case of the quantum states population dynamics shown in Fig. 2. Dashed lines are the diabatic lines. Time is normalized by τ_p.

$$b_2^{(k)}/b_1^{(k)} = \frac{w_k}{\hbar F_1}, \quad b_3^{(k)}/b_1^{(k)} = \frac{F_2 w_k}{F_1(w_k - \hbar\Delta\omega_{21})} \quad (k=1,2,3). \tag{13}$$

As it follows from these relations for $w_k = w_1$, and taking into account Eq. (11), the relative contribution of the (excited) state $|0\rangle$ [described by the component $b_2^{(1)}$; see Eqs. (12) and (13)] into the atomic adiabatic state is of the order of magnitude of $\Delta\omega_{21}/F_1$: $|b_2^{(1)}/b_1^{(1)}| \leqslant (\Delta\omega_{21}/F_1)$. This contribution decreases when the generalized Rabi frequency F_1 increases. It is negligibly small when the Rabi frequency exceeds considerably the frequency distance between the two ground states: $F_1 \gg \Delta\omega_{21}$. As the analysis of Eq. (9) for the quasi-energies shows, the solution for w_1 does not depend on the Rabi frequency at all and can be approximated as

$$w_1 \cong -\frac{d_{01}^2 + d_{03}^2}{d_{01}^2 + d_{03}^2 + d_{02}^2} \hbar\Delta\omega_{21}, \tag{14}$$

in the region of sufficiently large Rabi frequencies, when

$$|F_1|^2 + |F_2|^2 \gg |\varepsilon_1 \Delta\omega_{21}|, \quad |F_1| \gg |\Delta\omega_{21}|. \tag{15}$$

Note that in [2], two dark superposition of metastable states were obtained in the case when three interacting laser waves were in exact resonance with corresponding transitions (or in Raman resonance with each other). In contrast with that case, only one dark superposition of the close (quasi-degenerate) states $|1\rangle$ and $|3\rangle$ may be defined in the situation discussed in this paper, which is uncoupled from the interaction with the laser field [see Eq. (3)]. The second dark superposition state, which includes the separated (far) ground state $|2\rangle$ is not completely dark: there is interaction of this superposition at the leading and rear fronts of the laser pulse accompanied by some temporary excitation of the atom [see Eq. (11)]. This superposition is getting completely dark at sufficiently high values of the Rabi frequency exceeding the frequency separation between the far and the close states [see Eq. (14) and condition (15)].

We have a "trapped" superposition of two metastable states of the equivalent Λ-linkage atom in the region of constant quasienergy w_1 in Fig. 4. This trapped (dark) superposition state is composed of the bright superposition $|d_b\rangle$ state and the metastable state $|2\rangle$ of the original tripod-linkage atom. There is no excitation of the atom in this region. The quasienergy w_1 tends to the diabatic line $w_3^0 = -\hbar\Delta\omega_{21}$ at the end of the interaction with the pulse (at $t \to \infty$) in the case of a positive slope of the time variation of the frequency chirp (positive chirp, $\beta > 0$; see Fig. 3). Otherwise, it tends to the diabatic curve $w_2(t) = \hbar\varepsilon_1(t)$ in the case of negative chirp ($\beta < 0$). The latter corresponds to the quasi-energy of the excited state of the equivalent Λ-linkage atom, which is the same as the excited state of the original tripod-linkage atom.

The adiabatic states picture shows the possibility of transfer of the complete population of the superposition state $|d_b\rangle$ into state $|2\rangle$ with an appropriate sign of the frequency chirp. For the original tripod-linkage atom, this means the transfer of the bright superposition of the close metastable states $|1\rangle$ and $|3\rangle$ into the other far metastable state $|2\rangle$ without considerable excitation of the atom while the dark component of the superposition is being left without any variation during the interaction. In the case of the atom in the state $|1\rangle$ initially, the following superposition of the three metastable states of the atom will be created as a result of interaction with the laser pulse:

$$|\Psi(\infty)\rangle = \frac{\gamma_{31}^2}{1+\gamma_{31}^2}|1\rangle + \frac{1}{\sqrt{1+\gamma_{31}^2}}|2\rangle - \frac{\gamma_{31}}{1+\gamma_{31}^2}|3\rangle.$$

As it follows from the consideration presented in this section, the adiabatic-state analysis allowed to explain the results of the population dynamics and creation of the coherent superposition of the metastable states of the tripod-linkage atom obtained by the numerical simulation of the Schrödinger equation (see Fig. 2).

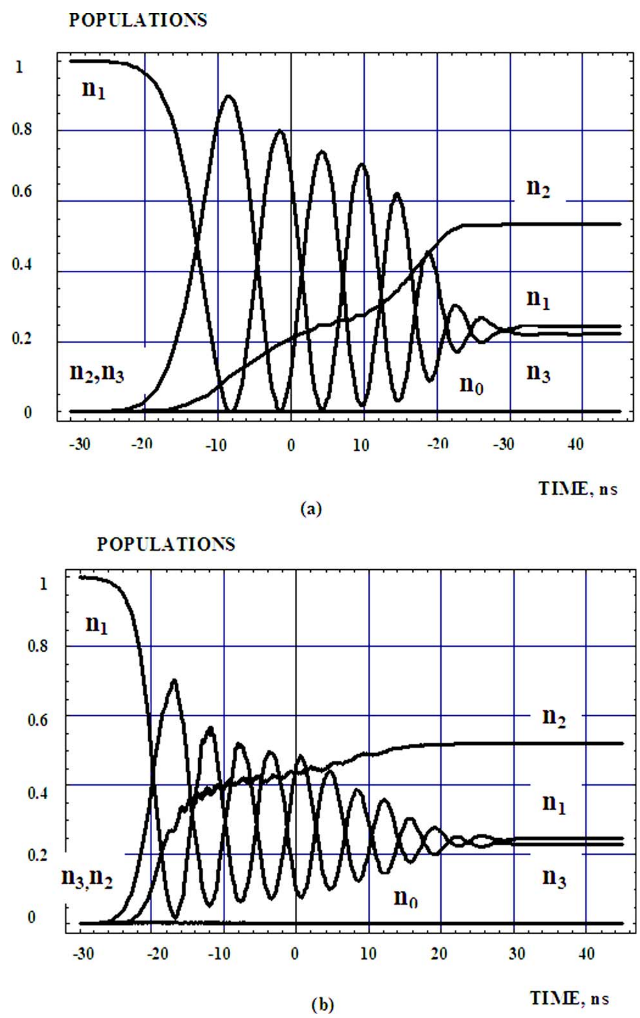

Fig. 5. (Color online) Population dynamics in the tripod-linkage atom for two atoms moving with different velocities (different values of the Doppler shift of the transition lines). The central frequency of the laser pulse has Doppler shift (a) $\omega_D = -2\beta\tau_p$; (b) $\omega_D = 2\beta\tau_p$.

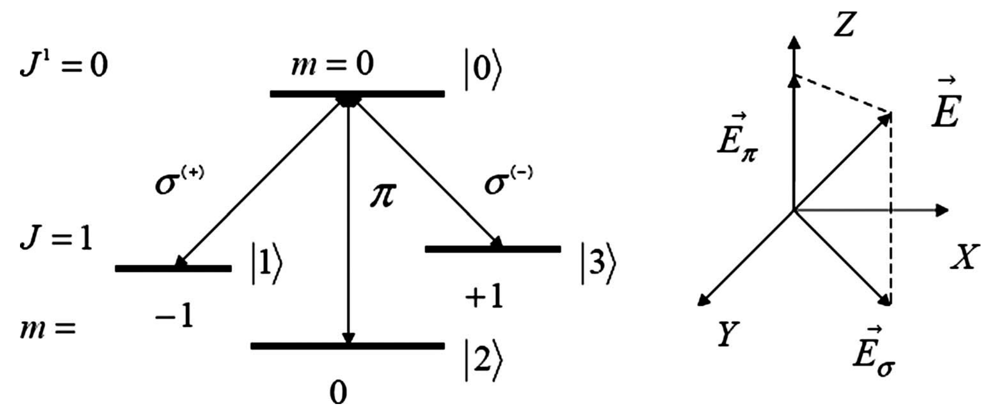

Fig. 6. Energy scheme of the $J=1 \leftrightarrow J^1=0$ transition of a Sm atom in a magnetic field with additionally applied nonresonant π-polarized laser field performing Stark shift of the sublevels with $m=0$. The electric strength vector $\vec{E}$ of the frequency-chirped laser pulse is shown in the right part of the figure with σ and π components. The quantizing magnetic field is directed along the Z axis.

4. DISCUSSION

As was mentioned in the Introduction, quantum superposition states play an important role in numerous applications in quantum and nonlinear optics and related fields. The creation of the coherent superposition of the metastable states considered above is accompanied by negligible excitation of the atom. Because no strict resonance conditions are needed for the laser pulse in this process, this method may be successfully applied also to inhomogeneously broadened media. Here the limitation on the width $\Delta\omega_{\mathrm{inh}}$ of the inhomogeneous broadening is that it must be smaller than the overall chirp of the pulse carrier frequency during the duration of the pulse: $\Delta\omega_{\mathrm{inh}} < \beta\tau_p$.

To verify the efficiency of the method in an inhomogeneously broadened medium, we present the population dynamics in the atom with tripod interaction linkages simulated for two identical atoms moving with two different velocities inducing Doppler shifts $\Delta\omega_D^{(1,2)} = \Delta\omega_{\mathrm{inh}}^{(1,2)}$ of the resonance lines in the atoms. These shifts correspond to the limiting values for the laser pulse frequency detuning: $\Delta\omega_D^{(1,2)} = \pm\beta\tau_p$ (see Fig. 5). The Doppler shift is included into the parameter ε_0 describing the detuning of the central frequency of the frequency-chirped laser pulse from the resonance frequency of the $|1\rangle \leftrightarrow |0\rangle$ transition. As it follows from Fig. 5, the proposed scheme of creation of the coherent superposition state is valid even for these two limiting values of the Doppler shift.

The proposed scheme of creation of coherent superposition states is robust against variations of the laser pulse peak intensity (Rabi frequency) and of the speed of the chirp. To verify the robustness, we introduced harmonic modulation of the laser pulse amplitude and (or) the speed of the chirp. The limitation on the parameters of this modulation, the depth and the frequency of the modulation, is that the spectrum broadening of the laser pulse induced by the modulation must be less than the frequency interval between the close and the far states to fulfill the second condition in Eq. (2). With this limitation on the modulation frequency, as our simulations showed, the proposed scheme was effective even at modulation of the laser pulse amplitude and the speed of the chirp with up to 50% depth.

As an appropriate quantum system for realization of the above-considered scheme for coherent superposition state creation may be considered the manifold of magnetic sublevels of $J=1 \leftrightarrow J'=0$ transition in, for example, the Sm atom, where the total momentum numbers $J=1$ and $J'=0$ stand for the ground and the excited states, respectively (see Fig. 6). Note that this quantum system was used in [42] to demonstrate experimentally inversionless amplification of picosecond laser pulses due to Zeeman coherence between the magnetic sublevels of the ground state.

To have simultaneous transitions between all working levels of the tripod-linkage atom, we need both π and σ polarization components to be present in the laser radiation. It may be realized, for example, by applying a linearly polarized laser pulse with the electric strength vector being at some angle to the quantizing magnetic field (see inset in Fig. 6). One has to apply an additional π polarized nonresonant laser radiation, which, due to the Stark effect, will shift the magnetic sublevel $m=0$ to realize the separated far ground state. The role of the close metastable states of the tripod-linkage atom will be played by the magnetic sublevels with quantum numbers $m=-1$ and 1. While transitions from these states to the excited state with magnetic quantum number $m=0$ will be provided by the $\sigma^{(+)}$ and $\sigma^{(-)}$ components of the E_σ projection of the electric field of the laser pulse, transitions between the states $|J=1, m=0\rangle$ and $|J'=0, m=0\rangle$ will be provided by the E_π polarization component of the laser field (see Fig. 6).

Variation of orientation of the electric field strength vector $\vec{E}$ will change the relative amplitudes of the E_σ and E_π polarization components and correspondingly, the Rabi frequencies for corresponding transitions. As a consequence, different compositions of the probability amplitudes of the states $|1\rangle$, $|2\rangle$, and $|3\rangle$ in the final superposition will be created. In this way, a desired superposition of these states may be created just by variation of the orientation of the electric vector.

Note that the above-considered scheme may be realized also by using magnetic sublevels of different hyperfine levels of the ground states of alkali metals (for example, Rb atoms). In the latter case one needs no additional nonresonant electric field for shifting of one of the levels. Spatially quantized systems, such as QD [43], including coupled QD [44], may be considered as other candidates of quantum systems appropriate for realization of the proposed scheme.

A QD with tripod linkage of the states with an appropriate frequency distance between the ground states (e.g., with two close and one far ground state with given distances between the states) may be calculated and also constructed experimentally. In the latter case too, one needs no additional nonresonant laser field to produce a far state.

5. CONCLUSIONS

In conclusion, we have proposed and analyzed a scheme of creation of coherent superposition of metastable states in an atom with a tripod-linkage of the levels using a single frequency-chirped laser pulse. The analysis was based on numerical solution of the Schrödinger equation for the probability amplitudes of the quantum states, and the results were verified and understood by using bright and dark superposition states along with the adiabatic state analysis. An important peculiarity of the scheme is that no appreciable excitation of the atom takes place during the interaction process, providing immunity to the decoherence effects connected with the spontaneous decay of the excited state. Our analysis has shown that the scheme under consideration is robust against variations in the laser pulse amplitude and the speed of the chirp: modulation of the laser pulse amplitude and the speed of the chirp with even 50% depth does not change significantly the efficiency of the scheme.

An important property of the scheme is its applicability to inhomogeneously broadened media due to the absence of strict resonance conditions. The limitation here is that the range of the frequency chirp during the laser pulse must exceed the width of the inhomogeneously broadened transition lines of the atomic ensemble. This property of the scheme is important for applications in quantum computing schemes based on quantum dots or the rare-earth-metal-ion-doped crystals.

Because the laser pulse with chirped frequency does not appreciably excite the atom in the scheme under consideration, we anticipate lossless propagation of the pulse in the otherwise absorbing medium similarly with the lossless propagation in the medium composed of atoms with Λ-interaction linkages (see [32]). An important difference here is that while the total population is transferred from one ground state to another one in the Λ-linkage atoms as the result of interaction with the frequency-chirped laser pulse, a coherent superposition of the metastable (ground states) is created in the case of the tripod-linkage atoms. It means that an optically thick medium consisting of such atoms may be prepared in the coherent superposition of metastable states along the trace of propagation of the frequency-chirped laser pulse. Such preparation of the atomic medium may find important applications in the nonlinear and quantum optics.

ACKNOWLEDGMENTS

This work was supported by the Research Fund (OTKA) of the Hungarian Academy of Sciences under contracts T038274, 034141, T031981, T42773, and GVOP-KMA230.

REFERENCES

1. 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).
2. 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).
3. H. Theuer, R. Unanyan, C. Habscheid, K. Klein, and K. Bergmann, “Novel laser controlled variable matter wave beamsplitter,” Opt. Express **4**, 77–83 (1999).
4. 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).
5. 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).
6. 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).
7. 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).
8. 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).
9. 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 lambda-atoms: population trapping and coherent optical pumping,” Laser Phys. **10**, 355–359 (2000).
10. 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).
11. C. P. Williams and S. H. Clearwater, *Explorations in Quantum Quantum Computing* (Springer-Verlag, 1997).
12. D. Bouwmeester, A. Ekert, and A. Zeilinger, *The Physics of Quantum Information: Quantum Cryptography, Quantum Teleportation, Quantum Computation* (Springer-Verlag, 2000).
13. 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).
14. M. Jain, Hui 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).
15. A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. F. Harris, “Raman generation by phased and antiphased molecular states,” Phys. Rev. Lett. **85**, 562–565 (2000).
16. 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).
17. E. Korsunsky, T. Halfmann, J. P. Marangos, M. Fleischhauer, and K. Bergmann, “Analytical study of four-wave mixing with large atomic coherence,” Eur. Phys. J. D **23**, 167–180 (2003).
18. T. Rickes, J. P. Marangos, and T. Halfmann, “Enhancement of third-harmonic generation by Stark-chirped rapid adiabatic passage,” Opt. Commun. **227**, 133–142 (2003).
19. P. Brumer and M. Shapiro, “Laser control of molecular processes,” Annu. Rev. Phys. Chem. **43**, 257–282 (1992).
20. K.-J. Boller, A. Imamoglu, and S. E. Harris, “Observation of electromagnetically induced transparency,” Phys. Rev. Lett. **66**, 2593–2596 (1991).
21. J. E. Field, K. H. Hahn, and S. E. Harris, “Observation of electromagnetically induced transparency in collisionally broadened lead vapor,” Phys. Rev. Lett. **67**, 3062–3065 (1991).

22. S. E. Harris, "Normal modes for electromagnetically induced transparency," Phys. Rev. Lett. **72**, 52–55 (1994).
23. S. E. Harris, and Z.-F. Luo, "Preparation energy for electromagnetically induced transparency," Phys. Rev. A **52**, R928–R931 (1995).
24. S. E. Harris, "Electromagnetically induced transparency," Phys. Today **50**, 36–42 (1997).
25. R. Grobe, F. T. Hioe, and J. H. Eberly, "Formation of shape-preserving pulses in a nonlinear adiabatically integrable system," Phys. Rev. Lett. **73**, 3183–3186 (1994).
26. F. T. Hioe and R. Grobe, "Matched optical solitary waves for three- and five-level systems," Phys. Rev. Lett. **73**, 2559–2562 (1994).
27. J. H. Eberly, M. L. Pons, and H. R. Haq, "Dressed-field pulses in an absorbing medium," Phys. Rev. Lett. **72**, 56–59 (1994).
28. J. Marangos, "Optics-focusing light with light," Nature **372**, 679–680 (1995).
29. L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Dover, 1987).
30. L. Rippe, M. Nilsson, S. Kröll, R. Klieber, and D. Suter, "Experimental demonstration of efficient and selective population transfer and qubit distillation in a rare-earth-metal-ion-doped crystal," Phys. Rev. A **71**, 062328 (2005).
31. 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).
32. G. P. Djotyan, J. S. Bakos, Zs. Sörlei, and J. Szigeti, "Coherent control of atomic quantum states by single frequency-chirped laser pulses," Phys. Rev. A **70**, 063406 (2004).
33. X. Z. Zhang, Q. Z. Ma, and X. H. Li, "Coherent population transfer in Rydberg potassium atom by a single frequency-chirped laser pulse," Chin. Phys. **15**, 1497–1501 (2006).
34. Z. Kis, A. Karpati, B. W. Shore, and V. V. Vitanov, "Stimulated Raman adiabatic passage among degenerate-level manifolds," Phys. Rev. A **70**, 053405 (2004).
35. 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).
36. S. Menon and G. S. Agarwal, "Gain from cross talk among optical transitions," Phys. Rev. A **59**, 740–749 (1999).
37. R. Unanyan, S. Guérin, B. W. Shore, and K. Bergmann, "Efficient population transfer by delayed pulses despite coupling ambiguity," Eur. Phys. J. D **8**, 443–449 (2000).
38. B. W. Shore, K. Bergmann, A. Kuhn, S. Schiemann, J. Oreg, and J. H. Eberly, "Laser-induced population transfer in multistate systems: a comparative study," Phys. Rev. A **45**, 5297–5300 (1992).
39. 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).
40. I. E. Mazets, "Adiabatic pulse propagation in coherent atomic media with the tripod level configuration," Phys. Rev. A **71**, 023806 (2005).
41. A. Messiah, *Quantum Mechanics* (North-Holland, 1962), Vol. II.
42. A. Nottelmann, C. Peters, and W. Lamge, "Inversionless amplification of picosecond pulses due to Zeeman coherence," Phys. Rev. Lett. **70**, 1783–1786 (1993).
43. L. Jacak, P. Hawrylak, and A. Wójs, *Quantum Dots* (Springer, 1997).
44. A. O. Orlov, I. Amlani, G. H. Bernstein, C. S. Lent, and G. L. Snider, "Realization of a functional cell for quantum-dot cellular automata," Science **277**, 928–930 (1997).