Manipulation of two-level quantum systems with narrow transition lines by short linearly polarized frequency-chirped laser pulses

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We propose and investigate theoretically a novel scheme for transient slowing and cooling of two-level quantum systems with narrow transition linewidths by a sequence of counterpropagating, short, linearly polarized laser pulses with special frequency chirping. Both internal degrees of freedom and the motion of the center of mass of quantum systems are considered quantum mechanically. Interaction with a large number of laser pulses during the decay time permits a drastic decrease in the cooling time of such systems. © 1996 Optical Society of America.

1. INTRODUCTION

Lately there has been considerable progress in laser manipulation, including slowing, cooling, and trapping of atoms.[1–4] Important applications of laser cooling include high-resolution spectroscopy and frequency standards[1,2] with cooled and trapped atoms as well as the construction of novel forms of matter with cooled atoms trapped in the optical lattices induced by the interference of multiple laser beams.[5,6]

The spontaneous emission of the excited atoms provides a dissipation mechanism for the standard schemes of Doppler cooling by laser radiation.[7,8] This is true for more complicated schemes of laser cooling as well. With more complex schemes, both the Doppler, $T_D = \hbar\Gamma/(2k_B)$, and the recoil, $T_R = (\hbar k_L)^2/(Mk_B)$, limits for the laser (one-dimensional) cooling can be overcome (see, for example, Refs. 9 and 10). In these formulas Γ is the spontaneous emission rate from the excited atomic state, k_L is the wave number of the laser radiation, and k_B is the Boltzmann constant. It is important to note that spontaneous emission not only plays the role of an energy-dissipation mechanism leading to thermal equilibrium with a reservoir but, owing to a random walk in momentum space, provides a mechanism for the atoms to diffuse into the zero-velocity trapped state, as described in Refs. 9 and 10.

In some experimental situations, however, the spontaneous decay time of excited quantum systems (QS's) is too long, and the standard schemes for laser cooling cannot be readily applied. Such a situation takes place, for example in the case of QS's with narrow transition widths (metastable atomic states), for which the time of flight through the interaction region with the laser field is of the order of the spontaneous relaxation time. The use of trains of laser pulses with durations shorter than the relaxation time of the QS provides means for effective laser manipulation of the QS in such experimental situations.

Interaction of the laser pulses with the QS in the cases mentioned above has essentially a transient character and needs a full quantum-mechanical description of both the internal degrees of freedom and the motion of the center of mass of the QS.[1]

Splitting of the atomic wave packet in velocity space as the result of the action of an ultrashort laser pulse in the coherent regime of interaction was obtained in the research reported in Refs. 1, 11, and 12. A transient regime of cooling by a sequence of short laser pulses from a narrow-band traveling-wave field that is periodically turned on and off under the condition that the atoms decay completely between the pulses was investigated in the study cited in Ref. 13.

A novel scheme of transient laser cooling of a QS with a narrow transition linewidth by a sequence of counterpropagating short laser pulses with special frequency chirping is proposed and investigated in this paper.

The laser cooling scheme proposed consists of two steps. First a velocity-selective excitation of the ensemble of a two-level QS is produced by a laser pulse with a Gaussian envelope [shown in the Fig. 1(a), with frequency chirping presented in Fig. 1(b)]. This pulse is termed asymmetrically chirped. The asymmetrically chirped laser pulse excites simultaneously the part of the ensemble of the QS's that have velocities v, for example, $v > v_c$ (v_c is the central velocity of the Maxwellian velocity distribution of the ensemble of QS's), in the laboratory reference system opposite the pulse propagation direction and pushes these QS's toward the central velocity v_c by transmitting to them a momentum $\hbar\mathbf{k}_1$ ($\mathbf{k}_1$ is the wave vector of the laser pulse), leaving the other QS's in the ensemble (with $v < v_c$) in their ground states. This initial preparation of the ensemble of QS's by asymmetric excitation permits the shifting, afterward, of the velocity distributions of the excited and unexcited QS's toward each other in velocity space by acting with successive counterpropagating pulses. These pulses have frequency chirping [shown in Fig. 1(c)] and are termed symmetrically chirped pulses. More precisely, the second laser pulse

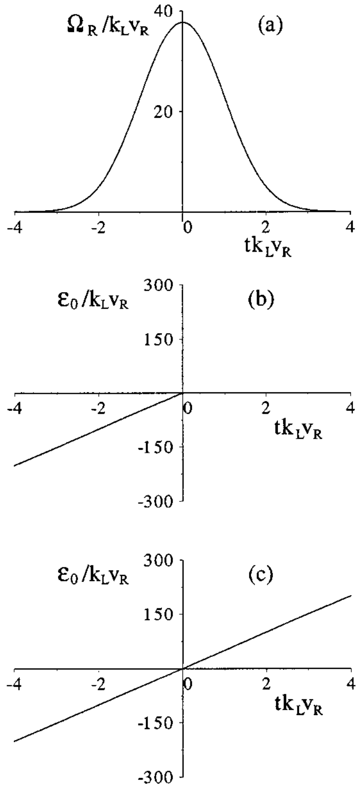

Fig. 1. (a) Time dependence of the normalized Rabi frequency Ω_R of a Gaussian laser pulse. (b) Time dependence of the normalized detuning $\epsilon(t) = \omega_L(t) - \omega_0$ for a QS having zero velocity in the laboratory reference frame in the case of an asymmetrically chirped Gaussian laser pulse. (c) Time dependence of the normalized detuning $\epsilon(t)$ in the case of a symmetrically chirped Gaussian laser pulse.

with symmetrical frequency chirping and counterpropagating to the first asymmetrically chirped one interacts with all velocity groups of QS's. This pulse excites the QS's with $v < v_c$ and pushes them toward the central velocity in the direction of wave vector $\mathbf{k}_2$ by transmitting to them a momentum $\hbar\mathbf{k}_2$. The same laser pulse simultaneously deexcites the QS's with $v > v_c$ previously excited by the first pulse and pushes them in the direction opposite that of $\mathbf{k}_2$ by transmitting to them a momentum $-\hbar\mathbf{k}_2$. In what follows, $|\mathbf{k}_1| = |\mathbf{k}_2| = k_L$ is assumed for the sake of simplicity.

Transmission of mechanical momentum from the electromagnetic field to QS's in the manipulation scheme described above takes place during successive stimulated excitation and deexcitation of the QS's by counterpropagating laser pulses with durations of the trains of pulses shorter than the relaxation times of the QS's. It is necessary for our manipulation scheme that a QS being excited (deexcited) by a laser pulse transit into a quantum state in which the next laser pulse counterpropagating to the first one can deexcite (excite) this QS. This means that the QS's have to transit to the initial quantum state after each act of the excitation–deexcitation process.

The application of laser pulses linearly polarized in the same direction seems to be the most convenient laser configuration for the scheme of transient laser manipulation described above. In this configuration the laser fields provide transitions between the sublevels of QS's with the same magnetic numbers (having their quantization axes along the direction of the electric strength vectors of the laser fields). However, this configuration restricts use of the method of momentum families,[14–16] which essentially simplifies the analysis of the generalized Bloch equations in the case of the $\sigma^+ \leftrightarrow \sigma^-$ configuration of circularly polarized counterpropagating laser pulses. Note that the theory of laser cooling in the field of counterpropagating linearly polarized cw laser fields in the weak-field limit was developed in the research reported in Refs. 17 and 18. This approach corresponds to the assumption of independent action of the counterpropagating laser beams. As is shown in the present paper, the restriction of the weak-field limit is no longer important if the counterpropagating laser pulses with suitably chirped frequencies do not overlap and the conditions of the adiabatic passage (AP) regime[19] are fulfilled.

This paper is organized as follows: In Section 2 we derive the set of generalized Bloch equations in the momentum representation describing the behavior of two-level QS's in the field of short trains of linearly polarized counterpropagating laser pulses in the density-matrix formalism. The evolution of the density-matrix elements owing to spontaneous emission is discussed as well. The results of the laser manipulation of QS's are presented in Section 3, based on the numerical simulation of the equations derived. In Section 4 we summarize the results on laser slowing and cooling of two-level QS's with narrow transition lines obtained in the present paper. The theory of the AP regime is developed in Appendix A, which describes the interaction of a linearly polarized laser pulse with a two-level QS that is initially in an arbitrary superpositional quantum state.

2. MATHEMATICAL FORMALISM

We begin with the equation for the density matrix $\rho(\mathbf{r}\eta, \mathbf{r}'\eta')$ of the QS, with $\mathbf{r}$ being the coordinate of the center of mass and η describing the internal motion of the QS (Ref. 20):

$$i\hbar \frac{\partial}{\partial t} \rho = (\hat{H} - \hat{H}'^*)\rho + i\hbar\hat{R}\rho, \tag{1}$$

where $\hat{H} = \hat{H}_0(\mathbf{r}, \eta) + \hat{V}(\mathbf{r}, \eta)$, where $\hat{H}_0(\mathbf{r}, \eta)$ is the atomic Hamiltonian:

$$\hat{H}_0(\mathbf{r}, \eta) = \hat{H}_0(\eta) - \frac{\hbar^2}{2m}\Delta,$$

$$\hat{H}_0(\eta)\psi_n(\eta) = E_n\psi_n(\eta). \tag{2}$$

Here Δ is the Laplacian and the interaction Hamiltonian $\hat{V}$ in the dipole approximation is $\hat{V}(\mathbf{r}, \eta) = -\hat{d}(\eta)\mathbf{E}(r)$, where $\hat{d}(\eta)$ is the dipole moment operator of the QS and $\mathbf{E}(r)$ is the strength of the laser field. E_n is the energy of the nth eigenstate of the QS with wave function ψ_n, and m is the mass of the QS. The operator $\hat{R}$ in Eq. (1) describes interaction with the vacuum fluctuations that cause relaxation of the QS. The operator $\hat{H}'$ is the same as $\hat{H}$ but acts on the variables with the coordinates $\mathbf{r}'$ and η'.

The strength $\mathbf{E}(\mathbf{r}, t)$ of the field of two counterpropagating linearly polarized laser pulses is

$$\mathbf{E}(\mathbf{r}, t) = 1/2\hat{e}\{ \overset{+}{A}(t)\exp[i(\omega t - k_L z)] + \overset{-}{A}(t) \times \exp[i(\omega t + k_L z)] + \text{c.c.}\}, \tag{3}$$

where z is the coordinate along which one-dimensional slowing and cooling is under consideration. $\hat{e}$ is the unit vector of polarization of the fields, $\overset{+}{A}(t)$ and $\overset{-}{A}(t)$ are the amplitudes of the laser pulses with frequency ω and wave number k propagating in the positive $(+)$ and negative $(-)$ directions, respectively, of the Z axes.

We expand the density matrix $\rho(r\eta, r'\eta')$ over the discrete set of eigenfunctions $\psi_n(\eta)$ of the quantum states $|n\rangle$ of the internal motion of the QS and over the continuous spectrum of eigenfunctions $\exp(i\kappa z/\hbar)$ of the free motion of the QS with momentum κ:

$$\begin{aligned}\rho(r\eta, r'\eta') &= \int\int \mathrm{d}\kappa \mathrm{d}\kappa' \bigg\{ \sum_{m,n} a_{nm}(\kappa, \kappa', t)\psi_n{}^*(\eta)\psi_m(\eta') \\ &\quad \times \exp[-i/\hbar(E_n - E_m)t] \\ &\quad \times \exp[i/\hbar(\kappa z - \kappa' z')]\bigg\}. \end{aligned} \tag{4}$$

A. Evolution Owing to Stimulated Transitions

In what follows we neglect the relaxation processes during the interaction with laser pulses, assuming the duration of each laser train to be significantly shorter than the relaxation time of the QS. After substitution of Eqs. (3) and (4) into Eq. (1), and using the orthonormality of the functions ψ_n and the orthogonality conditions for the functions $\exp(i\kappa z/\hbar)$, we obtain

$$\begin{aligned}&\left[\frac{\partial}{\partial t} + \frac{i\hbar}{2m}(\kappa^2 - \kappa'^2)\right] a_{lq}(\kappa,\kappa') \\ &= \frac{i}{2\hbar}\bigg\{ \sum_{n,m} d_{ln}[a_{nm}(\kappa + k_L,\kappa')\overset{+}{A}\exp(i\omega t) \\ &\quad + a_{nm}(\kappa - k_L,\kappa')\overset{+}{A}{}^*\exp(-i\omega t) \\ &\quad + a_{nm}(\kappa - k_L,\kappa')\overset{-}{A}\exp(i\omega t) \\ &\quad + a_{nm}(\kappa + k_L,\kappa')\overset{-}{A}{}^*\exp(-i\omega t)] \\ &\quad - \sum_{n,m} d_{qm}^*[a_{nm}(\kappa,\kappa' + k_L)\overset{+}{A}{}^*\exp(-i\omega t) \\ &\quad + a_{nm}(\kappa,\kappa' - k_L)\overset{+}{A}\exp(i\omega t) \\ &\quad + a_{nm}(\kappa,\kappa' - k_L)\overset{-}{A}{}^*\exp(-i\omega t) \\ &\quad + a_{nm}(\kappa,\kappa' + k_L)\overset{-}{A}\exp(i\omega t)]\bigg\} \\ &\quad \times \exp\{it/\hbar[(E_l - E_q) - (E_n - E_m)]\}, \end{aligned} \tag{5}$$

where d_{ij} is the dipole-moment matrix element for the transition between the $|i\rangle$ and the $|j\rangle$ quantum states.

The equations obtained are valid for QS's with arbitrary numbers of levels in the field of two linearly polarized counterpropagating laser pulses. In the case of a two-level QS we have $n, m, l, q = 1, 2$.

The relation $d_{ij} = d_{jj}(1 - \delta_{ij})$, $j = 1, 2$, takes place for an isotropic QS, where δ_{ij} is Kronecker's delta function. We obtain from Eq. (5) in the momentum representation and in the resonant approximation (rotating-wave approximation) the following set of equations for the nondiagonal density-matrix elements $\rho_{12}(\kappa, \kappa') = a_{12}(\kappa, \kappa') \exp(-i\epsilon t)$ and for the elements $a_{11}(\kappa, \kappa') = n_1(\kappa, \kappa')$ and $a_{22}(\kappa, \kappa') = n_2(\kappa, \kappa')$:

$$\begin{aligned}&\left[\frac{\partial}{\partial t} + \xi_{12}(\kappa, \kappa')\right]\rho_{12}(\kappa, \kappa') \\ &= \frac{i}{2\hbar} d_{12}\{ \overset{+}{A}[n_{22}(\kappa + k_L, \kappa') - n_{11}(\kappa, \kappa' - k_L)] \\ &\quad + \overset{-}{A}[n_{22}(\kappa - k_L, \kappa') - n_{11}(\kappa, \kappa' + k_L)]\}, \end{aligned}$$

$$\begin{aligned}&\left[\frac{\partial}{\partial t} + \xi(\kappa, \kappa')\right] n_{11}(\kappa, \kappa') \\ &= \frac{i}{2\hbar}\{d_{12}[\rho_{21}(\kappa + k_L, \kappa')\overset{+}{A} + \rho_{21}(\kappa - k_L, \kappa')\overset{-}{A}] \\ &\quad - d_{12}{}^*[\rho_{12}(\kappa, \kappa' + k_L)\overset{+}{A}{}^* + \rho_{12}(\kappa, \kappa' - k_L)\overset{-}{A}{}^*]\}, \end{aligned}$$

$$\begin{aligned}&\left[\frac{\partial}{\partial t} + \xi(\kappa, \kappa')\right] n_{22}(\kappa, \kappa') \\ &= \frac{i}{2\hbar}\{d_{21}[\rho_{12}(\kappa - k_L, \kappa')\overset{+}{A}{}^* \\ &\quad + \rho_{12}(\kappa + k_L, \kappa')\overset{-}{A}{}^*] - d_{21}{}^*[\rho_{21}(\kappa, \kappa' - k_L)\overset{+}{A} \\ &\quad + \rho_{21}(\kappa, \kappa' + k_L)\overset{-}{A}]\}, \end{aligned} \tag{6}$$

where

$$\epsilon(t) = \omega(t) - \omega_0,$$

$$\omega_0 = (E_2 - E_1)/\hbar,$$

$$\xi_{12}(\kappa, \kappa') = i\epsilon + \frac{i\hbar}{2m}(\kappa^2 - \kappa'^2),$$

$$\xi(\kappa, \kappa') = \frac{i\hbar}{2m}(\kappa^2 - \kappa'^2).$$

The functions $n_1(\kappa) = n_{11}(\kappa, \kappa' = \kappa)$ and $n_2(\kappa) = n_{22}(\kappa, \kappa' = \kappa)$ are the probabilities of finding the QS with momentum $p = \hbar\kappa$ in the ground or the excited state, respectively, and describe the populations of these states. We have for these functions, putting $\kappa = \kappa'$ into Eqs. (6),

$$
\begin{aligned}
&\frac{\partial}{\partial t}\, n_1(\kappa) \\
&\quad = \frac{i}{2\hbar}\,\{d_{12}[\rho_{21}(\kappa + k_L,\ \kappa)\,\overset{+}{A} \\
&\qquad + \rho_{21}(\kappa - k_L,\ \kappa)\,\overset{-}{A} - A] - d_{12}{}^*[\rho_{12}(\kappa,\ \kappa + k_L)\,\overset{+}{A}{}^* \\
&\qquad + \rho_{12}(\kappa,\ \kappa - k_L)\,\overset{-}{A}{}^*]\}, \\
&\frac{\partial}{\partial t}\, n_2(\kappa) \\
&\quad = \frac{i}{2\hbar}\,\{d_{21}[\rho_{12}(\kappa - k_L,\ \kappa)\,\overset{+}{A}{}^* \\
&\qquad + \rho_{12}(\kappa + k_L,\ \kappa)\,\overset{-}{A} - A - A^*] \\
&\qquad - d_{21}{}^*[\rho_{21}(\kappa,\ \kappa - k_L)\,\overset{+}{A} + \rho_{21}(\kappa,\ \kappa + k_L)\,\overset{-}{A}, \\
&\left[\frac{\partial}{\partial t} + \xi_{12}(\kappa,\ \kappa + k_L)\right]\rho_{12}(\kappa,\ \kappa + k_L) \\
&\quad = \frac{i}{2\hbar}\, d_{12}\{\,\overset{+}{A}[n_2(\kappa + k_L) - n_1(\kappa)] \\
&\qquad + \,\overset{-}{A}[n_{22}(\kappa - k_L,\ \kappa + k_L) - n_{11}(\kappa,\ \kappa + 2k_L)]\}.
\end{aligned}
\tag{7}
$$

It is easy to show that

$$\rho_{12}(\kappa,\ \kappa') = \rho_{21}{}^*(\kappa',\ \kappa).$$

We assume that no overlap takes place between the counterpropagating pulses that we are considering: The ensemble of QS's interacts with only one laser pulse at a time. If, for instance, $\overset{-}{A} = 0$, then we have a closed set of equations for $n_1(\kappa)$, $n_2(\kappa + k_L)$, and $\rho_{12}(\kappa,\ \kappa + k_L)$, as follows from Eqs. (7). The same is true for the case of $\overset{+}{A} = 0$. In the latter case a closed set of equations for $n_1(\kappa)$, $n_2(\kappa - k_L)$, and $\rho_{12}(\kappa,\ \kappa - k_L)$ follows from Eqs. (7) by substitution: $k_L \rightarrow -k_L$, $\overset{+}{A} \rightarrow \overset{-}{A}$.

We have to use as the initial conditions for the solution of Eqs. (7) for each subsequent laser pulse the values of the QS parameters obtained after interaction with the previous pulse counterpropagating to the current one. So, for example, we need to know the value of $\rho_{12}(\kappa,\ \kappa + k_L)$ obtained after the interaction of the QS with the $\overset{-}{A}$ pulse to calculate the result of its interaction with the following $\overset{+}{A}$ pulse from Eqs. (7). To do this we have to solve Eqs. (7) for $\rho_{12}(\kappa,\ \kappa + k_L)$ in the field of the $\overset{-}{A}$ laser pulse when $\overset{+}{A} = 0$:

$$
\begin{aligned}
&\left[\frac{\partial}{\partial t} + \xi_{12}(\kappa,\ \kappa + k_L)\right]\rho_{12}(\kappa,\ \kappa + k_L) \\
&\qquad = i\bar{\Omega}_R[n_{22}(\kappa - k_L,\ \kappa + k_L) - n_{11}(\kappa,\ \kappa + 2k_L)],
\end{aligned}
\tag{8}
$$

$\overset{\pm}{\Omega}_R = (d_{12}/2\hbar)\,\overset{\pm}{A}$ are the Rabi frequencies.

The density-matrix elements $n_{22}(\kappa - k,\ \kappa + k_L)$ and $n_{11}(\kappa,\ \kappa + 2k_L)$ appear in Eq. (8). These elements are diagonal with respect to the internal transitions and nondiagonal with respect to the transitions between the quantum states of motion of the center of mass; they describe multiphoton processes of absorption (emission) of photons from one laser pulse and emission (absorption) into another counterpropagating one.

It is clear that the number of equations to be solved for the density-matrix elements grows with an increase in the number of successive pumping laser pulses, similarly to the case of a QS interacting with a standing electromagnetic wave.[1,16,17] The only difference between the cases discussed in those papers and the one in the present paper is the absence of a standing laser field periodically modulated in space owing to the lack of overlap of the counterpropagating laser pulses assumed in our consideration. Note that no localization effects of the QS take place because a standing laser field is absent from our manipulation scheme.

Assuming that the laser pulses are frequency chirped, we consider the case when the conditions of the AP regime are fulfilled (see Appendix A, where the theory of the AP regime is developed for interaction of the laser pulse with the QS initially in a superpositional quantum state).

It is important to note that the population difference $\overset{\pm}{Z}_{(0)}(t) = n_2(\kappa \pm k_L) - n_1(\kappa)$ at the end of the frequency-chirped laser pulse ($t \rightarrow +\infty$) does not depend on the initial values of the nondiagonal density-matrix elements in the first approximation of the AP approach, as follows from Eqs. (A6) of Appendix A. So we have to solve Eqs. (7) to determine the populations $n_1(\kappa)$ and $n_2(\kappa + k_L)$ [or $n_2(\kappa - k_L)$] after the action of an $\overset{+}{A}$ (or an $\overset{-}{A}$) pulse, using the values of $n_1(\kappa)$ and $n_2(\kappa + k_L)$ obtained after the action of the previous $\overset{-}{A}$ (or $\overset{+}{A}$) pulse as the initial conditions. The initial value of the nondiagonal matrix element $\rho_{12}(\kappa,\ \kappa + k_L)$ [or $\rho_{12}(\kappa,\ \kappa - k_L)$] does not significantly influence the values of the populations obtained after interaction with the laser pulses in the AP regime. So in the AP regime of interaction we can restrict our consideration to solving only Eqs. (7) instead of considering the set of Eqs. (8) and those generated from them and obeying the development of the nondiagonal density-matrix element $\rho_{12}(\kappa,\ \kappa + k_L)$ [or $\rho_{12}(\kappa,\ \kappa - k_L)$] in the field of the $\overset{-}{A}$ (or the $\overset{+}{A}$) pulse. This peculiarity of the AP regime in the case of suitably chirped laser pulses essentially simplifies the mathematical consideration of the interaction of QS's with counterpropagating trains of laser pulses and allows us to consider the actions of the counterpropagating laser pulses independently from each other with no restrictions on the intensities of the laser pulses as in the weak-intensity approach.[18] Also, the coincidence of the results of calculations for velocity distributions in the density-matrix formalism of the present paper, in which independence of actions of the counterpropagating pulses is assumed, and those of Ref. 21, in which the formalism of the wave functions was used, clearly shows the correctness of this statement.

A. Evolution Owing to Spontaneous Emission

Spontaneous emission is assumed to take place mainly after the action of the counterpropagating trains of laser pulses and not during their interaction. So we can consider that this process occurs in the absence of laser radiation. This essentially simplifies the theoretical consideration. We have to solve the following equations describing the evolution of the populations of the QS's that is due to spontaneous emission:

$$\frac{\partial}{\partial t}\, n_1(\kappa) = \frac{\langle n_2(\kappa)\rangle}{T_1},$$

$$\frac{\partial}{\partial t}\, n_2(\kappa) = -\frac{n_2(\kappa)}{T_1}; \tag{9}$$

$$\langle n_2(\kappa)\rangle = \int_{-\hbar k_L}^{\hbar k_L} F(\kappa')n_2(\kappa+\kappa')\mathrm{d}k', \tag{10}$$

where T_1 is the spontaneous decay time and $F(\kappa')\mathrm{d}\kappa'$ is the probability that the spontaneously emitted photon with linear polarization will have its momentum between κ' and $\kappa' + \mathrm{d}\kappa'$ along the Z-space axis of the laser manipulation[17]:

$$F(\kappa) = \frac{3}{4k_L}\left(1 - \frac{\kappa^2}{k_L^2}\right).$$

We obtain for the population of the ground state after relaxation of the QS from the excited state [see Eqs. (9)]

$$n_1(\kappa,\, t \to +\infty)$$
$$= n_1(\kappa, 0) + \int_{-\hbar k_L}^{\hbar k_L} F(\kappa')n_2(\kappa+\kappa', 0)\mathrm{d}\kappa', \tag{11}$$

where $n_1(\kappa,0)$ and $n_2(\kappa,0)$ are the values of the populations obtained at the end of the previous cycle of laser manipulation.

3. RESULTS AND DISCUSSION

We consider laser pulses that have linear frequency chirp in the process of cooling described above. The frequency detuning $\epsilon_{1,2}(t)$ from resonance for such a laser pulse is

$$\epsilon_{1,2}(\kappa,t) = \omega_L - \omega_0 + 2\gamma t \mp \hbar k\kappa/m, \tag{12}$$

with + corresponding to the $\overset{-}{A}$ pulse with detuning ϵ_1 and − to the $\overset{+}{A}$ pulse with detuning ϵ_2. Here the frequency shifts $\mp \hbar k\kappa/m$ that are due to the Doppler effect have been included in the detunings $\epsilon_{1,2}$.

The scheme of manipulation is as follows: An ensemble of QS's which are initially in the ground state: $n_2(\kappa,\, t = -\infty) = 0$, with equilibrium Maxwellian distribution of the ground-state population $n_1(v/v_R,\, t = -\infty) = n_0(v/v_R)$ on the dimensionless velocity $v/v_R = \hbar\kappa/(mv_R)$ with the maximum at $v = v_c$:

$$n_0(v/v_R) = \frac{1}{\pi^{1/2}v_R q}\exp[-(v/v_R - v_c/v_R)^2/q^2], \tag{13}$$

is irradiated by subsequent laser pulses $\overset{+}{A}$ and $\overset{-}{A}$ that have symmetrical frequency chirps [see Fig. 1(c)]. $v_R = \hbar k_L/m$ is the recoil velocity and $q = v_T/v_R$, where v_T is the equilibrium thermal velocity of the ensemble.

Gaussian intensity envelope $I_L(t)$ is assumed for laser pulses with Rabi frequency $\Omega_R(t)$ and duration T_0:

$$\Omega_R(t) = \Omega_R^{(m)} \exp\left[-\frac{1}{2}\left(\frac{t/\tau_R}{\tau_L/\tau_R}\right)^2\right], \tag{14}$$

where $\tau_R = (k_L v_R)^{-1}$.

The parameters of the laser pulses are chosen so that the conditions of AP are fulfilled and the spontaneous decay of the excited QS is negligible during the trains of counterpropagating laser pulses. Thus, slowing of the ensemble takes place and the initial velocity distribution is pushed to one having a central velocity $v_c = 0$. The resulting velocity distribution of the ensemble after the action of 10 (an even number) laser pulses $\overset{+}{A}$ and $\overset{-}{A}$ is shown in Fig. 2. This figure clearly demonstrates the process of slowing by frequency chirped pulses in AP regime.

Let us now treat cooling of the ensemble with the velocity distribution obtained by considering this distribution as an initial one.

The first laser pulse applied for cooling (the 11th from the beginning of the manipulation process) is assumed to be an asymmetrically chirped $\overset{-}{A}$ pulse [see Fig. 1(b)]. The detuning $\epsilon_1(\kappa, t)$ passes through zero during the frequency chirping of the laser pulse, depending on the velocity of the QS. The chirping can be performed in such a way that $\epsilon_1(\kappa, t)$ passes through zero only for a QS moving from the left to the right (with $\kappa > 0$) in the laboratory reference frame. Transition to the excited state takes place for these QS's, which are initially in the ground state. At the same time, the QS's moving from the right to the left (with $\kappa < 0$) stay in their ground states, far from resonance with the laser field [Fig. 3(a)]. Then we use laser pulses with symmetrical frequency chirping to push the distributions of the excited and unexcited QS's toward each other. The resulting velocity distributions of QS's in the excited and the ground states after the action of 15 laser pulses (with the first pulse being asymmetrically chirped and with the other 14 being symmetrically chirped and propagating in the positive and the negative directions of the Z axes along which the cooling is being performed) are shown in Fig. 3(b).

At this point of interaction, relaxation of the excited

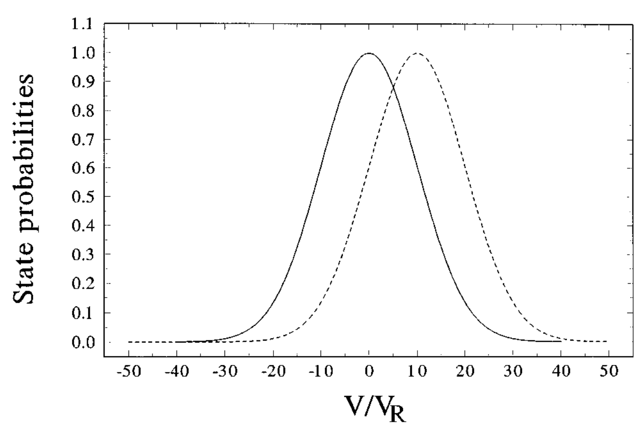

Fig. 2. Velocity distribution of the ensemble of two-level QS's after the action of 10 symmetrically chirped counterpropagating laser pulses with Gaussian envelopes [see Eq. (14)] and with linear frequency chirp: $\omega_L(t) = \omega_0 + 2c_1k_Lv_Rt/\tau_R$. The dashed curve is the initial Maxwellian velocity distribution of the ensemble, described by Eq. (13). The parameters applied are $c_1 = 8\pi$; $\Omega_R{}^{(m)}\tau_R = 12\pi$, $\tau_L = \tau_R$, $q = 10$, and $v_c/v_R = 10$.

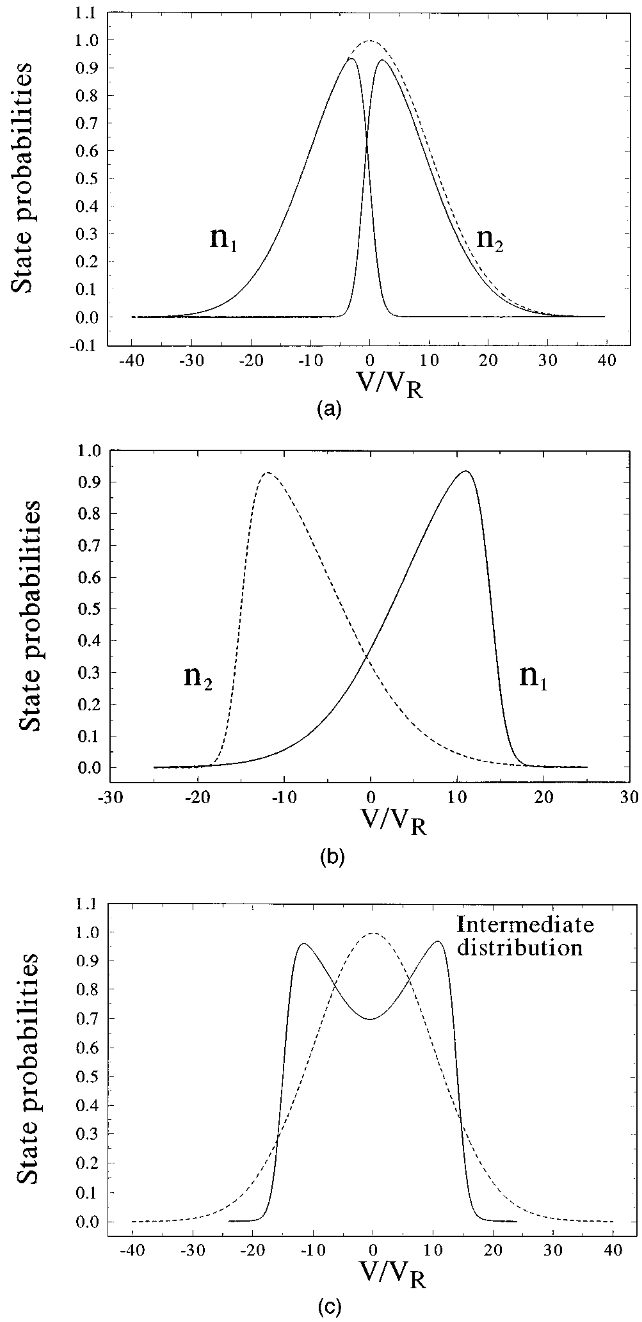

Fig. 3. Velocity distribution function of the ensemble of two-level QS's in the ground (n_1) and in the excited (n_2) states: (a) After the action of the first asymmetrically chirped laser pulse [see Fig. 1(b)]. The dashed curve represents the velocity distribution of the QS obtained after the slowing process (see Fig. 2). (b) After the action of 14 subsequent counterpropagating symmetrically chirped laser pulses. (c) The intermediate quasi-equilibrium velocity distribution function of the ensemble after the first relaxation to the ground state. The dashed curve is the same as the solid curve in Fig. 2.

QS's to their ground states is assumed to take place. The resulting equilibrium velocity distribution obtained by integration according Eq. (11) is shown in Fig. 3(c).

In the second step, as in the first one, we perform an excitation of nearly the half of the ensemble of QS's by an asymmetrically frequency-chirped laser pulse [Fig. 4(a)] and use laser pulses with symmetrical frequency chirping to push the distributions of the excited and unexcited QS's toward each other [Fig. 4(b)]. The final resulting velocity distribution after the second relaxation process is shown in Fig. 5. It displays narrowing of the velocity distribution, i.e., cooling of the ensemble of QS's. This is the

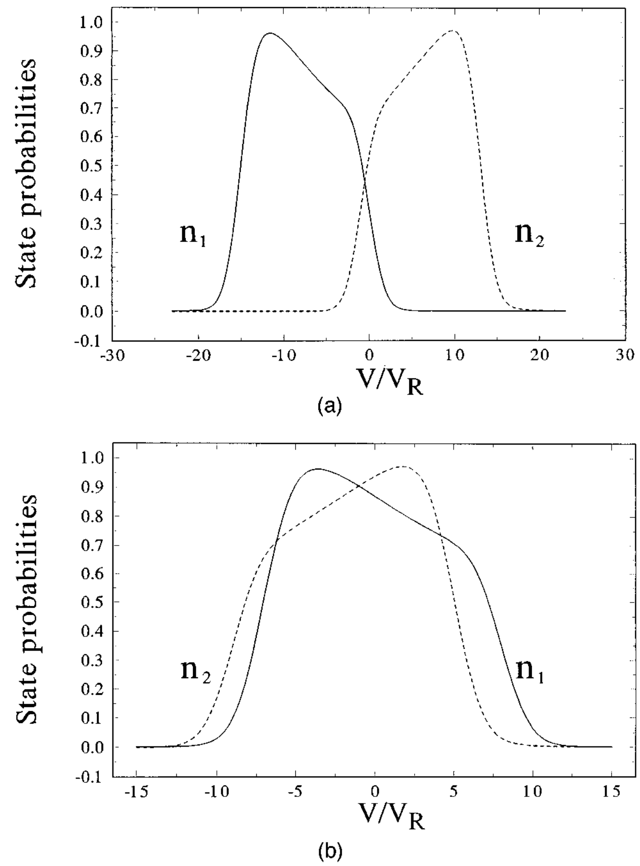

Fig. 4. Velocity distribution function of the ensemble of two-level QS's in the ground (n_1) and in the excited (n_2) states: (a) After the action of an asymmetrically chirped laser pulse applied after the first relaxation process, the 16th from the beginning of the cooling process. (b) After the action of 8 symmetrically chirped laser pulses applied after the first relaxation process, the 24th ones from the beginning of the laser cooling, with the 1st and the 16th asymmetrically chirped.

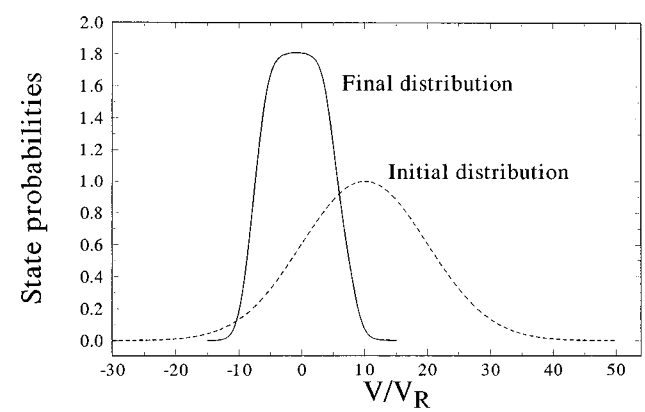

Fig. 5. Final quasi-equilibrium velocity distribution of the ensemble after the second relaxation process (the first cooling cycle).

first cycle of laser cooling. Repetition of this cycle can lead to significant cooling of the ensemble of QS's. The analysis shows that the slope of the final velocity distribution depends on the duration and the peak intensity of the laser pulse.

4. CONCLUSIONS

To summarize, we have presented the results obtained by fully quantum consideration of both the internal degrees of freedom and the motion of the center of mass of a QS with a narrow transition linewidth in our novel scheme of transient slowing and cooling by frequency-chirped counterpropagating short laser pulses.

The theory of AP has been developed in this paper for QS's initially in an arbitrary superpositional quantum state. Based on this theory, it has been demonstrated that in the case of short counterpropagating frequency-chirped laser pulses in the AP regime of interaction there is no need for restrictions such as those of the weak-intensity approach. This simplifies a fully quantum description of the motion of a QS in a field of short trains of counterpropagating frequency-chirped laser pulses.

Slowing of two-level QS's has been demonstrated by successive action with short, symmetrically chirped counterpropagating laser pulses. Cooling of the ensemble of QS's with near-zero central velocity obtained after the slowing process was accomplished by application of asymmetrically and successive symmetrically chirped laser pulses. The former, applied first to the ensemble of QS's in the ground state, excites only the half of the ensemble of QS's with positive (or negative) velocities in the laboratory reference frame and leaves the other half of the ensemble in the ground state. Subsequent application of symmetrically chirped pulses results the shifting of the excited and the unexcited parts of the velocity distributions of the QS's toward each other. Cooling of the ensemble takes place after the spontaneous transition of the excited part of the ensemble to the ground state. The method proposed can provide effective cooling of the ensemble of QS's during a few cooling cycles.

If, for an estimation of the pulse duration, we use parameters corresponding to fine-structure transitions of heavy ions[13]: $m = 200$ amu, $T_1 = 10^{-4}$ s, and $2\pi/k_L = 350$ nm, we obtain $\tau_L = \tau_R = m/(\hbar k_L^2) = 9.8 \times 10^{-6}$ s for the duration of the laser pulses used in our simulations. The number of symmetrically chirped pulses of nanosecond duration applied after the asymmetric excitation and before the relaxation of the QS's takes place is of the order of 10^3. This is also the factor by which the cooling time is reduced, compared, for example, to the transient cooling applied in a storage ring.[13] So the interaction of a QS with a large number of pulses during the decay time greatly reduces the cooling time by providing effective cooling of the QS within a few cycles.

It is important to note that the bandwidth of the asymmetrically chirped pulse has to be restricted when selective excitation is to take place in an ensemble of QS's. That is, the bandwidth of the asymmetrically chirped laser pulse has to be less than the width of the Doppler-broadened absorption line of the QS that is being cooled.

The minimum temperature obtainable by the cooling scheme proposed in the present paper, with the asymmetric laser pulses of duration $\tau_L = \tau_R$ used in our simulations (see also the estimations above) is limited by the temperature T_R, which corresponds to the recoil limit of the laser cooling. The latter one coincides in our case with the temperature $T_L = \hbar/(\tau_L k_B)$ determined by the bandwidths of the laser pulses. The temperature T_L will be the limiting temperature for our cooling scheme in the case of asymmetric laser pulses with shorter durations when $\tau_L < \tau_R$.

Note that the above-mentioned restriction on the bandwidth (duration) of the asymmetric pulses is not important in the case of symmetrically chirped pulses, which excite and deexcite all the QS's in the ensemble in the AP regime under consideration. The only additional requirement is that the frequency chirping of these pulses be produced to include not only the Doppler-broadened absorption line of the QS but the bandwidth of the laser pulse. This situation is similar to the one described in Ref. 22 for a homogeneously broadened atomic system, in which the picosecond frequency-chirped laser pulses create an inversion profile that is far narrower than the pulse spectrum.

APPENDIX A: THEORY OF ADIABATIC PASSAGE WITH ARBITRARY INITIAL CONDITIONS

It is convenient in the AP regime to use the effective spin vector formalism in which the dynamic response of a two-level QS to a driving electromagnetic field, for example $\overset{+}{A}$ ($\overset{-}{A}$), apart from relaxation processes is described by the generalized Bloch equations for the vector $\overset{+}{\mathbf{R}}$ ($\overset{-}{\mathbf{R}}$):

$$\partial \overset{\pm}{\mathbf{R}}/\partial t = \overset{\pm}{\mathbf{\Omega}} \times \overset{\pm}{\mathbf{R}}, \tag{A1}$$

where

$$\overset{\pm}{\mathbf{R}}(t,\kappa) = \hat{e}_1 \overset{\pm}{X} + \hat{e}_2 \overset{\pm}{Y} + \hat{e}_3 \overset{\pm}{Z},$$

$$\overset{\pm}{\mathbf{\Omega}}(t) = -\hat{e}_1 \overset{\pm}{\Omega}\Omega_R(t) + \hat{e}_3 \overset{\pm}{\theta}(t),$$

$$\overset{\pm}{\Omega}{}^2(t) = \overset{\pm}{\Omega}{}_R^2(t) + \overset{\pm}{\theta}{}^2(t);$$

$$\overset{\pm}{X}(\kappa,\ t) = \rho_{12}(\kappa,\ \kappa \pm k) + \rho_{12}{}^*(\kappa,\ \kappa \pm k),$$

$$\overset{\pm}{Y}(\kappa,\ t) = i[\rho_{12}{}^*(\kappa,\ \kappa \pm k) - \rho_{12}(\kappa,\ \kappa \pm k)],$$

$$\overset{\pm}{Z}(\kappa,\ t) = n_2(\kappa \pm k) - n_1(\kappa),$$

where the effective detuning $\overset{\pm}{\theta}(t)$ is

$$\overset{\pm}{\theta} = \xi_{12}(\kappa,\ \kappa \pm k) \cong i[\epsilon(t) \mp kv], \qquad v = \hbar\kappa/m.$$

We have the following equations from Eq. (A1) for the functions $\overset{\pm}{X}$, $\overset{\pm}{Y}$, and $\overset{\pm}{Z}$:

$$\frac{\partial}{\partial t}\ \overset{\pm}{X} = \overset{\pm}{\theta}\ \overset{\pm}{Y},$$

$$\frac{\partial}{\partial t}\ \overset{\pm}{Y} = -\overset{\pm}{\theta}X + \overset{\pm}{\Omega}_R\overset{\pm}{Z},$$

$$\frac{\partial}{\partial t}\ \overset{\pm}{Z} = -\ \overset{\pm}{\Omega}_R\ \overset{\pm}{Y}. \tag{A2}$$

The assumption of slow variation in time of the angle formed by the vector of the generalized Rabi frequency $\mathbf{\Omega}$ (from now on we drop the superscripts for the sake of simplicity) with the Z axis in abstract space with respect to Rabi frequency Ω_R forms the basis of the AP approximation[18,21,23]:

$$|\nu| = \frac{|\dot{\theta}\Omega - \theta\dot{\Omega}|}{\Omega^2\Omega_R} \ll 1, \tag{A3}$$

where the overdot denotes time derivation.

Another assumption that generally was made in the earlier considerations of AP[19,24,25] was that the QS is in a pure state initially, before the interaction with the laser pulse. However, the situation when the QS is initially in a superpositional state is the case that often occurs in the schemes of interaction with trains of laser pulses. We present in this appendix the results of the theory of AP developed for interaction of laser pulses with QS's initially in arbitrary superpositional states. The laser pulses are assumed to be frequency chirped.

It is convenient to transform the reference system in abstract space into one obtained by rotation of the coordinate system around the Y axis with the new Z' axis coinciding with the vector of the generalized Rabi frequency $\mathbf{\Omega}$. This transformation has the following form:

$$X' = \pm X\cos[\alpha(t)] \mp Z\sin[\alpha(t)],$$

$$Z' = \pm X\sin[\alpha(t)] \pm Z\cos[\alpha(t)],$$

$$Y' = Y, \tag{A4}$$

where $\alpha(t)$ is the angle between the instantaneous direction of the vector $\mathbf{\Omega}(t)$ and that at the beginning of the interaction at $t \to -\infty$. The upper sign corresponds to the case when the initial effective detuning $\theta(t = -\infty) > 0$. The lower sign is valid for negative initial values of the detuning $\theta(t = -\infty) < 0$.

The set of Eqs. (A2) has the following form in the new coordinate system after transformation into the new time variable

$$\beta = \int_{t_0}^{t}\Omega(t')\mathrm{d}t':$$

$$\dot{X}' + \nu(\beta)Z' = \mp Y',$$

$$\dot{Y}' = \pm X',$$

$$\dot{Z}' = \nu(\beta)X', \tag{A5}$$

where the overdot denotes derivation over the time variable β.

The small parameter $\nu \ll 1$ [see inequality (A3)] appears in the equations obtained. The smallness of this parameter allows us to use perturbative methods for approximate solution of Eqs. (A5):

$$X' = x_0 + x_1 + x_2 + \ldots,$$

$$Y' = y_0 + y_1 + y_2 + \ldots,$$

$$Z' = z_0 + z_1 + z_2 + \ldots,$$

where c_n $(c = x, y, z)$ is of the order of ν^n and $n = 0, 1, 2, \ldots$.

The solution of Eqs. (A5) in the zero-order ($n = 0$) approximation after their transformation into the old nonrotating coordinate system is as follows:

$$X_0(t) = \pm\frac{\theta(t)}{\Omega(t)}[X(-\infty)\cos(\beta) \mp Y(-\infty)\sin(\beta)]$$

$$+ \mp\frac{\Omega_R(t)}{\Omega(t)}Z(-\infty),$$

$$Y_0(t) = \pm X(-\infty)\sin(\beta) + Y(-\infty)\cos(\beta),$$

$$Z_0(t) = \pm\frac{\theta(t)}{\Omega(t)}Z(-\infty) \pm \frac{\Omega_R(t)}{\Omega(t)}[X(-\infty)\cos(\beta)$$

$$\mp Y(-\infty)\sin(\beta)]. \tag{A6}$$

The relations $X(-\infty) = Y(-\infty) = 0$ take place when the QS is in the ground or in the excited state initially, and we obtain the well-known solutions for the population difference $Z(t)$ and the nondiagonal matrix element $\rho = (X + iY)/2$.[11,24,25]

As follows from Eqs. (A4), corresponding to the zero-order approximation (AP regime), the influence of the initial values $X(-\infty)$ and $Y(-\infty)$ of the nondiagonal elements of the density matrix is negligible at the end of the laser pulse when $\Omega_R(t \to +\infty) = 0$.

Note that new features of the process of interaction of the laser pulse with the QS appear when the latter is initially in a superpositional state, i.e., $X(-\infty) \neq 0$, $Y(-\infty) \neq 0$, or both. For example, an oscillation regime can be predicted in this case that is absent in the case of $X(-\infty) = Y(-\infty) = 0$. This can influence the behavior of the internal motion of the QS as well as the motion of its center of mass. One can predict other interesting features of the propagation effects by taking into account the variation of the laser pulse that is due to interaction with the QS.[26]

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