Transient laser cooling of two-level quantum systems with narrow natural linewidths

J. S. Bakos, G. P. Djotyan,* G. Demeter, and Zs. Sörlei
Research Institute for Particle and Nuclear Physics of the Hungarian Academy of Sciences, XII. Konkoly Thege út 29-33, P.O. Box 49, H-1525, Budapest, Hungary

(Received 11 September 1995)

We propose a scheme of transient laser manipulation and cooling of two-level quantum systems with narrow natural linewidths by a sequence of counterpropagating laser pulses with special frequency chirping. Interaction with a large number of laser pulses within the decay time decreases drastically the cooling time of such systems.

PACS number(s): 32.80.Pj, 42.50.Vk

Laser cooling of atoms has recently become a very important topic of investigation in the field of interaction of laser radiation with matter. Important applications of laser cooling include high-resolution spectroscopy and frequency standards [1,2] with cooled and trapped atoms, as well as construction of new forms of matter from cooled atoms trapped in the "optical lattices" created by the interference of multiple laser beams [3,4]. The simplest mechanism of cooling of neutral atoms by laser radiation is the so-called Doppler cooling, first proposed in [5,6]. Doppler cooling occurs when an atom is irradiated by counterpropagating laser beams tuned below the atomic resonance. Cooling in this scheme is achieved by the absorption of photons from the laser beams and the spontaneous emission of excited atoms. Spontaneous emission plays a vital role in other, more complicated schemes of 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 of laser cooling can be overcome (see, for example [7,8]). 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, but, due to a random walk in momentum space, provides a mechanism for atoms to diffuse into the zero-velocity trapped state, as in [7,8].

The standard schemes of laser Doppler cooling are not effective in the case of quantum systems (QS) with narrow widths of transitions (metastable atomic states). The main reason is that the average light pressure force in the case of Doppler cooling with cw laser beams is proportional to the linewidth of the quantum transition. Hence this force is weak, and the cooling time may be too large compared with the time of flight of the QS through the region of interaction with the laser beam. The usage of trains of laser pulses with duration shorter than the relaxation time of the QS may be a tool for effective laser cooling in the situation mentioned above. Interaction of laser pulses with QS in this case has an essentially transient character, and so a full quantum mechanical approach for the description of both the internal degrees of freedom, and the motion of the center of mass of the QS is needed [2].

Splitting of the atomic wave packet in velocity space due to the action of ultrashort laser pulses in the coherent regime of interaction was obtained in [2,9,10]. The transient regime of cooling by a sequence of short laser pulses of the narrow-band traveling-wave field under the condition that the atoms decay completely between pulses was investigated in [11]. It was shown in [12] that more deceleration of neutral atoms may be achieved with the train of ultrashort π pulses separated by a few radiative lifetimes than would be achived with the same average intensity used for cw generation. Laser cooling on transitions with a linewidth narrower than the recoil shift was studied by the Monte Carlo method using broadband excitation in [13]. The force of the stimulated light pressure on an atom arising from the action of two near-resonant standing waves with different frequencies modeling two counterpropagating waves with modulated in time amplitudes was studied in [14].

A scheme of transient laser cooling of QS with a narrow transition linewidth by a sequence of counterpropagating ultrashort laser pulses with special frequency chirping is proposed and investigated in this paper. The laser cooling scheme consists of two steps. First, a velocity-selective excitation of the ensemble of two-level QS is produced by a frequency chirped laser pulse, Fig. 1(a). The pulse has special frequency chirping, which is shown in Fig. 1(b) and is termed asymmetrically chirped pulse in the following. This pulse excites simultaneously the part of the ensemble of QS having velocity v; for example, $v>0$ in the laboratory reference system, which is opposite the pulse propagation direction. The QS are pushed towards the zero velocity state by the absorption of a momentum $\hbar k_1$ (k_1 is the wave vector of the laser pulse). The other part of the ensemble ($v<0$) remains in the ground state, as presented in Fig. 2(a). This initial preparation of the ensemble of QS by an asymmetrical excitation allows us, in the second phase, to move the distributions of the excited and unexcited QS toward each other in velocity space. This is achieved with the help of subsequent, counterpropagating laser pulses, with frequency chirping that is shown in Fig. 1(c) and is termed symmetrical chirping. Thus, the second laser pulse with symmetrical chirping and counterpropagating to the first one interacts with all velocity groups of the QS. This pulse excites QS with $v<0$, pushing them towards the zero velocity state in the direction of the wave vector k_2, by the absorption of a momentum $\hbar k_2$. The same laser pulse simultaneously deexcites the QS with $v>0$, previously excited by the first pulse, and pushes them in the direction opposite that of k_2, by the stimulated emis-

*Permanent address: NPO "Lazerayin Technika," Yerevan State University, Yerevan, Armenia.

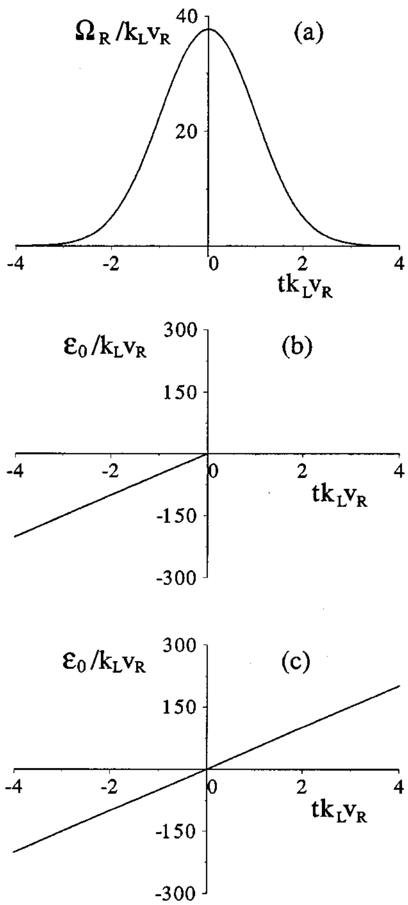

FIG. 1. (a) Time dependence of the normalized Rabi frequency $\Omega_R = |A^{(+)}d_{12}/(\hbar)|$ of the Gaussian laser pulse. (b) Time dependence of the normalized frequency detuning $\varepsilon_0 = \omega_L(t) - \omega_0$ of an asymmetrically chirped Gaussian laser pulse. (c) Time dependence of the normalized frequency detuning ε_0 of the symmetrically chirped Gaussian laser pulse.

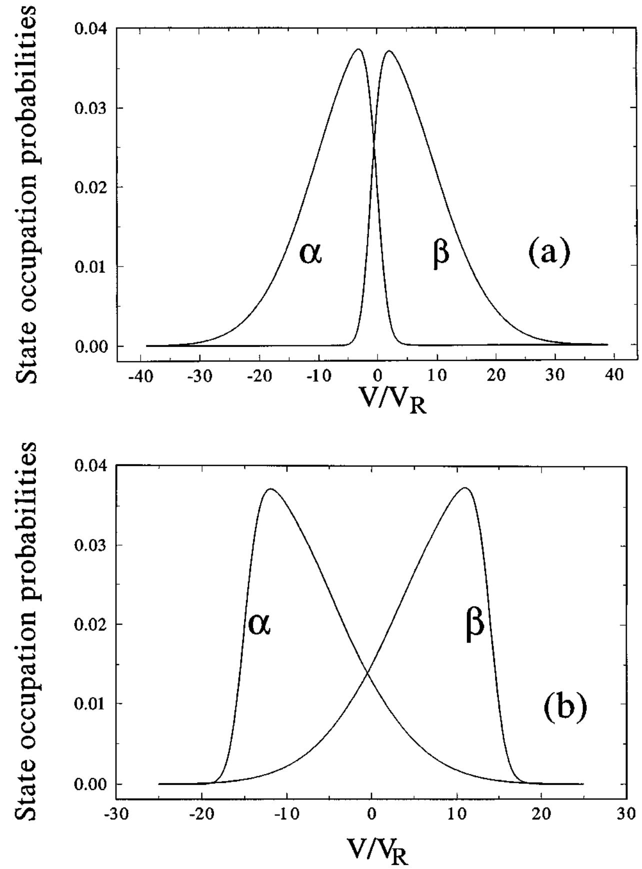

FIG. 2. Velocity distribution function of the ensemble of two-level QS in the ground (α) and in the excited (β) states. The velocity is normalized by the recoil velocity v_R. All laser pulses applied are identical to Gaussian intensity distribution in time. The maximum Rabi frequency, normalized by $k_L v_R$ is equal to 12π. The normalized duration of the pulse $\tau_L k_L v_R = 1$ and the linearly chirped frequency $\omega_L(t) = \omega_0 + 2c_1 k_L v_R t$ with $c_1 = 8\pi$. The width σ of the initial Maxwellian velocity distribution in the ensemble is put equal to $10v_R$. (a) After the action of the first asymmetrically chirped laser pulse [see Fig. 1(b)]. (b) After the action of 14 subsequent, counterpropagating symmetrically chirped laser pulses [see Fig. 1(c)].

sion of a momentum $\hbar k_2$. From now on, $|k_1| = |k_2| = k_L$ is assumed for the sake of simplicity.

The strengths $E^{(\pm)}$ of the electromagnetic fields of the laser pulses, linearly polarized along the unit vector e, with amplitudes $A^{(+)}(t)$ and $A^{(-)}(t)$, propagating in the positive and negative directions of the z axes are

$$E^{(\pm)} = \tfrac{1}{2} e\{A^{(\pm)} \exp(i\omega_L t \mp ik_L z) + \text{c.c.}\}.$$

The internal motion of the QS and the motion of its center of mass, both quantum mechanicaly considered, are described in the density-matrix formalism by the set of generalized Bloch equations (see, for example [2]). The number of equations in this set is infinite in the case of linearly polarized, counterpropagating laser pulses under consideration. Note that the choice of $\sigma_+ \leftrightarrow \sigma_-$ configuration of circularly polarized laser beams allows the simplification of the set of generalized Bloch equations [7,15]. These difficulties are circumvented in our case of laser pulses with durations shorter than the decay time of the QS, since it is possible to describe the interaction of the QS with the laser pulses in terms of the atomic probability amplitudes instead of using the density-matrix formalism. The approximation that decay can be ignored during the action of a group of pulses but not between the different pulse groups simplifies the mathematical treatment of the problem. The influence of the relaxation to the ground state may then be analyzed by the generalized Bloch equations in the absence of the laser field.

The equations for the probability amplitudes of the ground $a(\kappa,t)$ and the excited, $b(\kappa,t)$ states of QS in the $E^{(-)}$ field in the momentum representation are [2]

$$\frac{\partial a(\kappa+\kappa_L/2)}{\partial t} + i\varepsilon_1(\kappa)a(\kappa+\kappa_L/2) = \frac{i}{2\hbar}A^{(-)}d_{12}b(\kappa-k_L/2), \tag{1}$$

$$\frac{\partial b(\kappa-k_L/2)}{\partial t} - i\varepsilon_2(\kappa)b(\kappa-k_L/2) = \frac{i}{2\hbar}A^{(-)*}d_{21}a(\kappa+k_L/2),$$

where

$$\varepsilon_1(\kappa)=\frac{\omega_L-\omega_0}{2}+\frac{k_L\kappa\hbar}{2M}-\frac{\hbar k_L^2}{8M},$$

$$\varepsilon_2(\kappa)=\frac{\omega_L-\omega_0}{2}+\frac{k_L\kappa\hbar}{2M}+\frac{\hbar k_L^2}{8M}.$$

ω_0 is the eigenfrequency of QS moving with normalized velocity $v=\hbar k/M$ in the laboratory reference system, with M being the mass of the QS. $\omega_R=\hbar k_L^2/8M$ represents the change of the resonance frequency of the QS due to the recoil after the absorption or emission of a photon with momentum $\hbar k_L$. The substitutions $A^{(-)}\rightarrow A^{(+)}$ and $k_L\rightarrow -k_L$ have to be made in Eq. (1) to describe interaction of QS with the $E^{(+)}$ field.

The value $|a(\kappa,t)|^2$ gives the probability of the QS moving with a velocity $v=\hbar k/M$ and being in the ground state $|a\rangle$ at time t. The value $|b(\kappa,t)|^2$ has the same meaning for the excited state $|b\rangle$.

We introduce the function $f(k)$ in such a way that $|f(k)|^2$ is the probability of QS having normalized velocity k and being in either the ground state with amplitude $a(k\pm k_L/2)$ or the excited state with amplitude $b(k\mp k_L/2)$, with the upper (lower) sign corresponding to the interaction with the $E^{(-)}$ ($E^{(+)}$) field.

Initially, all QS with quantum-mechanically well-defined velocities within the ensemble are assumed to be in their ground states, with the Maxwellian velocity distribution

$$|a(\kappa,t=0)|^2=1,$$

$$|b(\kappa,t=0)|^2=0, \tag{2}$$

$$|f(k)|^2=\frac{1}{\sigma\sqrt{\pi}}\exp\left(-\frac{k^2}{\sigma^2}\right),$$

where $\sigma=v_TM/\hbar$, with v_T being the equilibrium thermal velocity of the QS. The velocity-dependent probabilities $\alpha(k,t)=|f(k)a(k,t)|^2$ and $\beta(k,t)=|f(k)b(k,t)|^2$ of occupying the ground (α) and excited (β) states at an arbitrary instant of time t are normalized in such a way that the integral of $\alpha(k,t)+\beta(k,t)$ over the velocity space is equal to unity.

The laser pulses in our cooling scheme are assumed not to overlap each other in the medium consisting of the two-level QS. Thus, we can neglect the standing-wave effects responsible for the multiphoton processes of stimulated absorption and emission from one laser beam into another.

The interactions of the two-level QS with the laser pulses have been analyzed by the numerical simulation of Eq. (1), with the initial conditions (2) for the first pulse. The initial conditions for each subsequent pulse are given by the solution of Eq. (1) after the action of the previous pulse. Laser pulses with Gaussian intensity distributions in time, Fig. 1(a), are assumed to be applied to the ensemble of QS in the numerical simulation. During the first step of cooling, 15 laser pulses are applied to the ensemble, the first of which is asymmetrically chirped; the remaining 14 are symmetrically chirped and propagating in alternating directions along the z axes. The resulting velocity distributions for the QS from the ensemble in the excited and in the ground states are shown in Fig. 2(b). At this point, relaxation of excited QS to the ground state is assumed to take place. The resulting quasiequilibrium velocity distribution (here we neglect collisions between QS) after the spontaneous transition of the excited QS to their ground states is shown on Fig. 3.

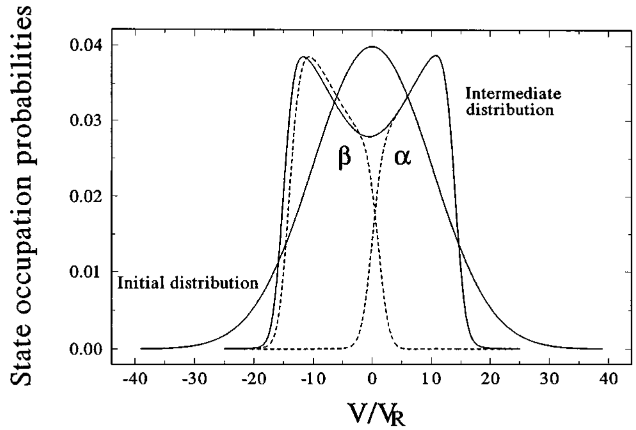

FIG. 3. Intermediate quasiequilibrium velocity distribution function of the ensemble after the relaxation to the ground state. All parameters are the same as in Fig. 2. Dotted lines: the state occupation probabilities α and β after the action of an asymmetrically chirped laser pulse [see Fig. 1(b)], the 16th from the beginning of the cooling process.

During the second step, we excite about one-half of the ensemble of QS with an asymmetrically chirped laser pulse, just as before (see Fig. 3). Afterwards, laser pulses with symmetrical frequency chirping are applied to push the distributions of excited and unexcited QS towards each other. The resulting velocity distributions of QS in the ground (α) and the excited (β) states are shown in Fig. 4 by dotted lines. The final quasiequilibrium velocity distribution after the spontaneous transition of the excited part β of the ensemble of QS to the ground state is shown in the same Fig. 4. It

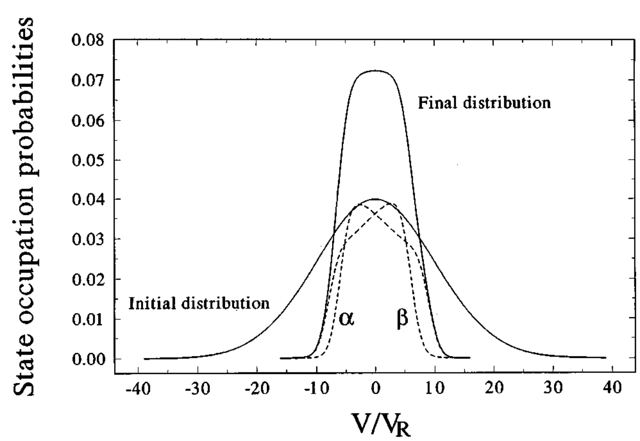

FIG. 4. The final velocity distribution function of the ensemble after the second relaxation process (the first cooling cycle). All parameters are the same as in Fig. 2. Dotted lines: the state occupation probabilities α and β after the action of 8 subsequent, counterpropagating symmetrically chirped laser pulses, the 24th from the beginning of the cooling process.

clearly displays the narrowing of the velocity distribution obtained compared with the initial one, i.e., the cooling of the ensemble of QS. This is the first cycle of cooling. Repetition of this cycle may lead to efficient cooling of the ensemble of QS. The analysis shows that the slopes of the final velocity distribution depend on the asymmetrically chirped laser pulse duration and peak intensity.

The minimum temperature obtainable by the cooling scheme proposed in the present paper, with the duration of the laser pulses applied, $\tau_L=1/k_L v_R$, is limited by T_R, corresponding to the recoil limit of the laser cooling, which in this case coincides with the temperature $T_L=\hbar/(\tau_L k_B)$ determined by the bandwidth of the laser pulses. The latter will be the limiting temperature for our cooling scheme for shorter laser pulses with $\tau_L<1/k_L v_R$. Note that the constraint on the duration of the asymmetrically chirped pulse is important if selective excitation is to take place in the ensemble of QS, namely, the bandwidth of the asymmetrically chirped laser pulse has to be less than the width of the Doppler broadened absorption line of the QS being cooled. This restriction, however, is not important in the case of the symmetrically chirped pulses, which excite and deexcite all the QS in the ensemble in the adiabatic passage regime [16] under consideration. The only additional requirement is that the frequency chirping of these pulses must be produced to include not only the Doppler broadened absorption line of QS but the bandwidth of the laser pulse. This situation is similar to the one described for a homogeneously broadened system in [17], where the picosecond frequency-chirped laser pulses create an inversion profile that is far narrower than the pulse spectrum.

If, for an estimation of the pulse duration, we use parameters corresponding to fine-structure transitions of heavy ions [11]: $M=200$ amu, $T_s=10^{-4}$ s, and $2\pi/k_L=350$ nm, we obtain $\tau_L=1/k_L v_R=M/\hbar k^2=9.8\times10^{-6}$ s for the duration of the asymmtrically chirped pulse 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 takes place is of the order of 10^3. This is also the factor by which cooling time is reduced compared to, for example, the transient cooling applied in a storage ring [11]. The interaction of QS with a large number of pulses during the decay time greatly reduces the cooling time by providing an effective cooling of the QS within a few cycles of the method proposed. It is important to note that we have obtained the same results as ones presented in this paper by using the density-matrix formalism in the adiabatic passage regime.

In conclusion, note that some modifications of our method of transient laser manipulation and cooling may be proposed by introducing a cutoff in the chirping process of the laser pulses at the resonance of QS belonging to the narrow velocity group near the zero velocity or utilizing population trapping mechanisms.

This work was supported by the Research Fund (OTKA) of the Hungarian Academy of Sciences under Contract No. T-4089.

[1] Special issue on laser cooling and trapping of atoms, J. Opt. Soc. Am. B **6**, 2020 (1989).
[2] V. G. Minogin and V. S. Letokhov, *Laser Light Pressure on Atoms* (Gordon & Breach, New York, 1987).
[3] G. Grynberg *et al.*, Phys. Rev. Lett. **70**, 2249 (1993).
[4] A. Hemmerich *et al.*, Europhys. Lett. **22**, 89 (1993).
[5] T. W. Hansch and A. Schawlow, Opt. Commun. **13**, 68 (1975).
[6] D. Wineland and H. Dehmelt, Bull. Am. Soc. **20**, 699 (1975).
[7] A. Aspect *et al.*, Phys. Rev. Lett. **61**, 826 (1988).
[8] M. Kasevich *et al.*, Phys. Rev. Lett. **69**, 1741 (1992).
[9] A. P. Kazantsev, Usp. Fiz. Nauk **124**, 113 (1978) [Sov. Phys. Usp. **21**, 58 (1978)].
[10] R. J. Cook, Phys. Rev. Lett. **41**, 1788 (1978).
[11] K. Molmer, Phys. Rev. Lett. **66**, 2301 (1991).
[12] P. D. Lett *et al.*, Hyperfine Interact. **44**, 335 (1988).
[13] H. Wallis and W. Ertmer, J. Opt. Soc. Am. B **6**, 2211 (1989).
[14] V. S. Voitsekhovich *et al.*, Pis'ma Zh. Éksp. Teor. Fiz. **49**, 138 (1989) [JETP Lett. **49**, 161 (1989)].
[15] Y. Castin, H. Wallis, and J. Dalibard, J. Opt. Soc. Am. B **6**, 2046 (1989), pp. 3–24.
[16] C. Liedenbaum *et al.*, Phys. Rep. **178**, No. 1 (1989).
[17] J. S. Melinger *et al.*, Phys. Rev. Lett. **68**, 2000 (1992).