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SPECTROSCOPY OF ATOMS AND MOLECULES

Deep Raman Cooling of Alkaline-Earth Atoms

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Abstract—We have analyzed deep Raman cooling of alkaline-earth atoms with two stable and two excited states in a field of three traveling light waves. It has been shown that the excitation of this system by Raman pulses applied between lowest states of the system can be used to deeply cool its atoms to a temperature that is on the order of or even lower than the temperature determined by the recoil energy.

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In the field of laser cooling of atoms, optical cooling of atoms of alkaline-earth metals is of special interest, since such atoms are used as a working medium for new optical frequency standards based on neutral atoms, with the stability of standards being at a level of 10^{-17} [1–5]. In this case, the clock transition the frequency of which is to be stabilized is a singlet–triplet intercombination transition, which is characteristic of all the alkaline-earth atoms, as well as the ytterbium atom. Clearly, in order to obtain record high stabilities, it is required to preliminarily cool these atoms at least to temperatures of alkali atoms in optical traps (10^{-4}–10^{-5} K). However, in this case, particular features of the energy-level diagram of quantum levels of alkaline-earth atoms do not make it possible to directly reach the required temperature. The reason for this is that, although the Doppler cooling based on a strong optical transition $^1S_0 \to {}^1P_1$ (in the case of the ^{40}Ca atom, the transition is $4s^2\,{}^1S_0 \to 4s4p\,{}^1P_1$) is efficient, it is still does not ensure the required cooling temperature, since this transition lies in the blue range of the spectrum and has a considerable width [6, 7]. At the same time, cooling based on a narrow line of an intercombination transition (e.g., the linewidth of the transition $4s^2\,{}^1S_0 \to 4s4p\,{}^3P_1$ in the ^{40}Ca atom is 400 Hz) can potentially ensure a low temperature. However, since the transition linewidth is small, only a small fraction of atoms from the entire velocity distribution will actively interact with the optical radiation, which sharply lowers the efficiency of the so-called Doppler cooling. In addition, the use of narrow lines leads to a situation in which the recoil frequency of the atom is high compared to the natural linewidth of the atomic transition. The latter circumstance does not allow one to apply the semiclassical model of the Doppler cooling.

One way to obviate a long lifetime upon cooling based on the narrow $4s^2\,{}^1S_0 \to 4s4p\,{}^3P_1$ transition is to use the method of controlling the spontaneous decay from the $^3P_1$ state, so-called quenched cooling (Fig. 1a) [6]. The idea is to facilitate the return of atoms to the ground $^1S_0$ state without waiting for the spontaneous decay due to the narrow (long-lived) transition to occur [6]. To do this, it is proposed to use additional optical radiation that is resonant to the $4s4p\,{}^3P_1 \to 4s5s\,{}^1S_0$ transition. As a result, the atom passes to the upper $4s5s\,{}^1S_0$ excited state with the subsequent decay to the $4s^2\,{}^1S_0$ ground state via the $^1P_1$ intermediate state. Then, if corresponding wave vectors and detunings of optical fields are chosen, the width of the initial momentum distribution will, on average, decrease due to the emission of spontaneous photons as a result of a cascade process $4s5s\,{}^1S_0 \to {}^1P_1 \to 4s^2\,{}^1S_0$.

In this work, we consider a method of deep Raman cooling for a four-level system of quantum states, with the occurrence of the $4s4p\,{}^3P_1$ long-lived lower state (state $|3\rangle$ in Fig. 1) that is necessary for this method. In this case, the Raman cooling proceeds in the same way as for an ordinary Λ atom with states $|1\rangle$, $|4\rangle$, and $|3\rangle$ (Fig. 1b). Unlike ordinary Raman cooling, two, rather than one, codirected traveling waves with different wavenumbers k_1 and k_2 are used in the left arm of the Λ system. Then, at the first stage, a velocity-selective π pulse acts on the atom in state $|1\rangle$ in a field of counterpropagating waves (with wavenumbers k_1, k_2, and k_3) for momentum p, the value of which is unambiguously determined by the choice of detunings δ_1 and δ_3.

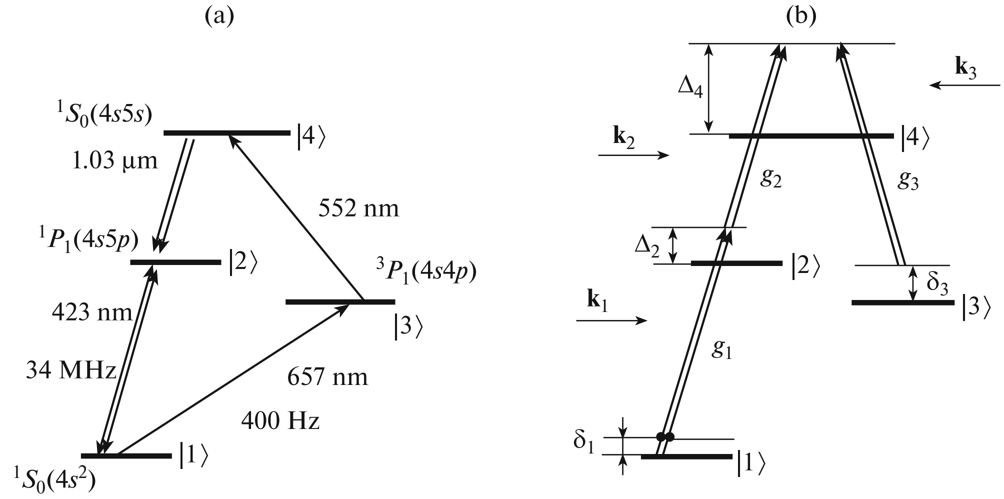

Fig. 1. (a) Scheme of energy levels of the ^{40}Ca atom. (b) A four-level atom in a field of three traveling light waves. The wave with frequency ω_1 is resonant to the optical transition $|1\rangle$–$|2\rangle$, the wave with frequency ω_2 is resonant to the optical transition $|2\rangle$–$|4\rangle$, and the wave with frequency ω_3 is resonant to the optical transition $|3\rangle$–$|4\rangle$, while the transition $|1\rangle$–$|3\rangle$ is a long-lived intercombination transition. The waves with wave vectors k_1 and k_2 are unidirectional, while the wave with wave vector k_3 propagates in the opposite direction.

Part of the atomic population is then transferred from level $|1\rangle$ to the second lowest level of the three-level system, becoming shifted by a value $\hbar(k_1 + k_2 + k_3)$ in the momentum space. The width of the momentum distribution formed in state $|3\rangle$ is determined by all the three Rabi frequencies and two detunings of light waves Δ_2 and Δ_4 from excited states $|2\rangle$ and $|4\rangle$ of the four-level system, while the position of this distribution on the momentum axis is determined by detunings δ_1 and δ_3 from the two-photon resonance. The next step in the Raman cooling implies the use of a resonant optical pumping, when the population from level $|3\rangle$ passes to excited state $|4\rangle$ under the action of the optical field and then reaches state $|1\rangle$ via intermediate state $|2\rangle$ by means of spontaneous decay. Upon passage from level $|3\rangle$ to state $|4\rangle$, the atomic distribution is shifted by a value of the recoil momentum k_3, while, upon emission of spontaneous photons from level $|4\rangle$ to level $|1\rangle$ via intermediate state $|2\rangle$, the momentum distribution is additionally shifted by a value of the algebraic sum of the projections of the recoil momenta of the atom from spontaneously emitted photons with wavenumbers k_1 and k_2 onto the direction of propagation of light rays. Because the direction of emission of spontaneous photons fluctuates, the projection of the recoil momentum of the atom also changes randomly in the interval $-\hbar(k_1 + k_2) \leqslant p_z \leqslant \hbar(k_1 + k_2)$. As a result, the peak of the velocity distribution of the population from level $|3\rangle$ returns to state $|1\rangle$ with a some random momentum shift, which leads to an increase in the number of atoms in state $|1\rangle$ for the initial value of the momentum p of the atom. After that, the cycle of cooling is repeated for other value of the atomic momentum.

It should be emphasized that the difference between the methods of Raman and quenched coolings lies in different pathways of population of long-lived state $|3\rangle$. In the case of the quenched cooling, state $|3\rangle$ is populated because a π pulse is directly applied between states $|1\rangle$–$|3\rangle$. In the Raman cooling method, long-lived state $|3\rangle$ is also populated by the application of a π pulse between states $|1\rangle$–$|3\rangle$, but indirectly, via upper states. In this case, the coherent interaction regime can be realized only if detunings $\Delta_{2,4}$ of optical fields from excited states $|2\rangle$ and $|4\rangle$ are considerable. Therefore, the potential advantage of the use of the Raman cooling can consist of a more rapid (compared to the quenched cooling) transfer of part of the population from $|1\rangle$ to $|3\rangle$ because of the use of stronger optical transitions.

Consider now the realization of the Raman cooling mechanism of a four-level system where wave vectors are directed such as is shown in Fig. 1b. This choice of the directions of propagation of traveling waves is caused by the necessity of the velocity selection in the system of levels in Fig. 1b.

The system of equations for nonstationary probability amplitudes that describe the coherent dynamic of an atom in the field of three traveling waves for the interaction scheme under consideration can be represented as

$$\begin{aligned} &i\dot{a}_1(p - k_1 - k_2) \\ &= [(p - k_1 - k_2)^2 + \delta_1]a_1(p - k_1 - k_2) \\ &\quad - g_1 a_2(p - k_2), \end{aligned} \tag{1a}$$

$$i\dot{a}_2(p-k_2) = [(p-k_2)^2 - \Delta_2]a_2(p-k_2) - g_1 a_1(p-k_1-k_2) - g_2 a_4(p), \quad (1b)$$

$$i\dot{a}_3(p+k_3) = [(p+k_3)^2 + \delta_3]a_3(p+k_3) - g_3 a_4(p), \quad (1c)$$

$$i\dot{a}_4(p) = [p^2 - \Delta_4]a_4(p) - g_3 a_3(p+k_3) - g_2 a_2(p-k_2), \quad (1d)$$

where we have already passed to the momentum representation, introduced a resonant approximation, used the rotating wave approximation for the probability amplitudes $a_{2,4}(p)$, and normalized the momentum of the atom to quantity $\hbar k_2$. In Eqs. (1), g_i ($i = 1, 2, 3$) are the Rabi frequencies of light waves of the corresponding indices normalized to recoil frequency $\omega_R = \hbar k_2^2/2M$ (M is the mass of the atom), $\Delta_{2,4}$ are the frequency detunings of light waves from states $|2\rangle$ and $|4\rangle$ normalized to ω_R, and $\delta_{1,3}$ are similar detunings from states $|1\rangle$ and $|3\rangle$, which are also normalized to ω_R. In addition, in Eqs. (1), time is expressed in units of inverse recoil frequency ω_R^{-1}.

We emphasize that a particular feature of the interaction scheme presented in Fig. 1 is the fact that the wavenumbers of the exciting waves are considerably different. Correspondingly, the wavenumbers are also expressed in units of $k_2 = \omega_2/C$, and, in Eqs. (1), the wavenumbers are in fact written as $k_1 = k_1/k_2$, $k_2 = k_2/k_2 = 1$, and $k_3 = k_3/k_2$.

Then, assuming that $\Delta_{2,4} \gg g_{1,2,3}$, we eliminate adiabatically probability amplitudes $a_{2,4}(p)$. To do this, we equate the time derivatives on the left-hand sides of Eqs. (1b) and (1d) to zero, $\dot{a}_{2,4}(p) \approx 0$, find probabilities $a_{2,4}(p)$ from these equations, and substitute them into the two remaining equations. As a result, we obtain a system of two equations, which can be written in the form

$$i\dot{a}_1(p-k_1-k_2) = \left[(p-k_1-k_2)^2 + \delta_1 + \frac{g_1^2}{\Delta_2}\right]a_1(p-k_1-k_2) - \alpha a_3(p+k_3), \quad (2a)$$

$$i\dot{a}_3(p+k_3) = \left[(p+k_3)^2 + \delta_3 + \frac{g_3^2}{\Delta_4}\right]a_3(p+k_3) - \alpha a_1(p-k_1-k_2), \quad (2b)$$

$$\alpha = g_1 g_2 g_3/\Delta_2\Delta_4.$$

Solutions of Eqs. (2) can be represented as

$$a_1(p-k_1-k_2) = \left[a_1^{(0)}(p-k_1-k_2)\cos D(p) + \frac{\alpha}{D(p)}\left(\frac{\beta_3-\beta_1}{2\alpha}a_1^{(0)}(p-k_1-k_2) + a_3^{(0)}(p+k_3)\right) \times \sin D(p)\right]\exp(-i\xi(p)t), \quad (3a)$$

$$a_3(p+k_3) = \left[a_3^{(0)}(p+k_3)\cos D(p) + \frac{\alpha}{D(p)}\left(\frac{\beta_3-\beta_1}{2\alpha}a_3^{(0)}(p+k_3) + a_1^{(0)}(p-k_1-k_2)\right) \times \sin D(p)\right]\exp(-i\xi(p)t), \quad (3b)$$

where

$$D(p) = (1/2)\sqrt{[\beta_1(p) - \beta_3(p)]^2 + 4\alpha^2},$$

$$\beta_1(p) = (p-k_1-k_2)^2 + \delta_1 + \alpha_1,$$

$$\beta_3(p) = (p+k_3)^2 + \delta_3 + \alpha_3,$$

$$\xi(p) = (1/2)[\beta_1(p) - \beta_3(p)], \quad \alpha_1 = \frac{g_1^2}{\Delta_2}, \quad \alpha_3 = \frac{g_3^2}{\Delta_4}.$$

The probabilities of population of lowest states under the initial conditions

$$a_1(p, t=0) = a_1^{(0)}(p) = \exp(-p^2/(\Delta p)^2),$$
$$a_3(p, t=0) = 0$$

have the form

$$W_1(p) = \left|a_1^{(0)}(p)\right|^2 \times \left[1 - \frac{\alpha^2}{D(p+k_1+k_2)^2}\sin^2(D(p+k_1+k_2)t)\right], \quad (4a)$$

$$W_3(p) = \left|a_1^{(0)}(p-k_3-k_1-k_2)\right|^2 \frac{\alpha^2}{D(p-k_3)^2} \times \sin^2(D(p-k_3)t), \quad (4b)$$

where Δp is the momentum width of the initial distribution of atoms. Transition probabilities (4) from state $|1\rangle$ to state $|3\rangle$ obtained above show how the intercombination state is populated. As can be seen from expressions (4), the population of state $|3\rangle$ depends both on the momentum of the atom and on characteristics of all the applied optical fields. Here, a maximal population of state $|3\rangle$ is achieved at $D(p - k_3) = \alpha$ during the time of the π pulse $t_\pi = \pi/2\alpha$, which is possible only if the equality $\beta_1(p - k_3) = \beta_3(p - k_3)$ holds. The latter condition directly defines the value of the

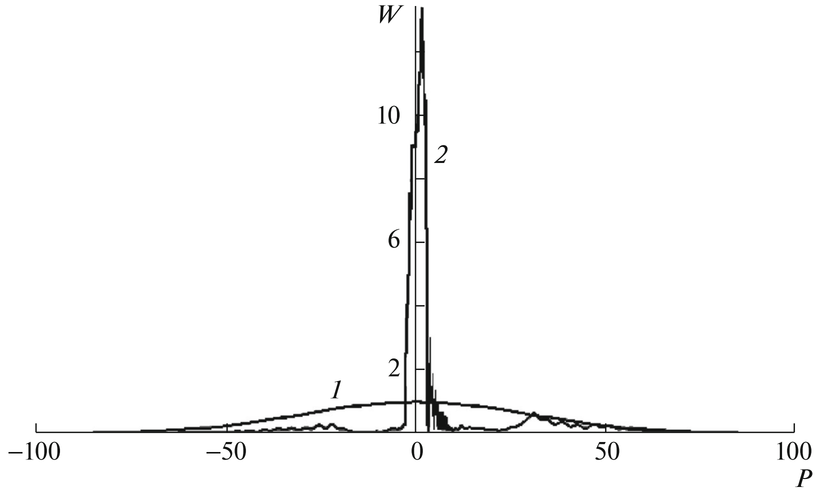

Fig. 2. (*1*) Initial velocity distribution of atoms with a width of $\Delta p \cong 70$. (*2*) An average of over 40 realizations: a narrow peak of cooled atoms with a width of $\Delta p \cong 5$, which was obtained as a result of the action of 74 pairs of pulses of laser radiation (each pair consists of π pulses applied between states $|1\rangle$ and $|3\rangle$ and pulses of pumping from level $|4\rangle$ to level $|3\rangle$) with subsequent spontaneous decay of the population transferred from level $|4\rangle$ to ground state $|1\rangle$ via intermediate state $|2\rangle$. In this case, in each realization, the value of α decreases from 20 to 1.56 upon approaching zero on the momentum axis, while durations of corresponding π pulses increase from 0.08 to 1.

momentum of the atom for which the population is maximal,

$$p_\pi = \left[\left(\sum_{i=1}^{3} k_i\right)^2 + \delta_1 - \delta_3 + \alpha_1 - \alpha_3\right] \Bigg/ 2\sum_{i=1}^{3} k_i. \quad (5)$$

We note that, in accordance with (5), the value of resonant momentum of the atom p_π is determined by all three wavenumbers, detunings δ_1 and δ_3 from levels $|1\rangle$ and $|3\rangle$ and effective Rabi frequencies α_1 and α_3.

In order to calculate the final velocity distribution of atoms due to the Raman cooling, we will use the initial velocity distribution of atoms with a width of $\Delta p \cong 70$. This value of the velocity scatter corresponds to a temperature of 2.4 mK and is characteristic of the Doppler cooling on the basis of transition $|1\rangle$–$|2\rangle$ [7]. At the first stage, we used solution (4) of the equation in order to determine the probability of excitation of the atom. In this case, the Rabi frequencies and the detunings for the numerical simulation were chosen such that to obtain π pulses between states $|1\rangle$ and $|3\rangle$ for the value of the resonant momentum given by (5). As a result, part of the population passed to state $|3\rangle$, becoming shifted in the momentum space by a value $k_1 + k_2 + k_3$ from the initial momentum. By varying parameters δ_1, δ_3, α_1, and α_3, one can choose any value of the resonant momentum. Therefore, in the calculation, we acted alternately on the momentum distribution by positive and negative momenta. Then, under the action of the resonant radiation, the distribution of atoms was moved from level $|3\rangle$ to state $|4\rangle$, shifting along the momentum axis by $-k_3$ and, after that, all atoms experienced a spontaneous decay to ground state $|1\rangle$ via intermediate state $|2\rangle$. The modeling of the spontaneous decay consisted of that the population arrived from the excited state could have any random value of the momentum in the interval between $-k_1 - k_2$ and $k_1 + k_2$. As a result, we obtained a new momentum distribution, which, at the next step, was again used as an initial distribution.

Results of numerical simulation are presented in Fig. 2, which shows the shapes of the initial velocity distribution and the distribution that was obtained after the action of 74 π pulses applied between states $|1\rangle$ and $|3\rangle$ and pulses of pumping from level $|4\rangle$ to level $|3\rangle$ (with subsequent spontaneous decay to ground state $|1\rangle$ via intermediate state $|2\rangle$), which alternate pairwise between the positive and negative sides of the initial momentum distribution. Rabi frequencies and durations of π pulses of the laser radiation were chosen such that, in the range distant from zero pulses, the population with a rather wide momentum distribution would be captured (the greater the value of α, the shorter the duration of the π pulse and the wider the peak) and, the closer the zero pulse range, the narrower the peaks of the transferred population (the smaller the value of α, the longer the duration of the π pulse and the narrower the peak). Upon approaching the zero-point of the momentum axis, the value of α decreases from 20 to 1.56, while the duration of corresponding π pulses increases from 0.08 to 1. Because the shape of the final distribution is random, in order for the description to be correct, we present the resultant value that was averaged over 40 realizations. It can be seen that a narrow peak of cooled atoms is formed indeed, the width of which is $\Delta p \cong 5$, which corresponds to an effective temperature of atoms on the order of ≅12 μK. It should be noted that not all the atoms gather in a narrow peak, and a part of them acquire a velocity that is higher than the initial velocity as a result of random fluctuations of the direction of emission of spontaneous photons. However, the amount of these atoms is small (<20%) and the main part of the initial momentum distribution is efficiently cooled. This method of cooling of alkaline-earth atoms can also be efficient in the case of the use of dynamical traps [8].

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