Subrecoil cooling of atoms with a nondegenerate ground state

B. G. Matisov, I. A. Grigorenko, and I. E. Mazets
A. F. Ioffe Physicotechnical Institute, 194021 St. Petersburg, Russia

(Submitted 16 August 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **62**, No. 6, 466–471 (25 September 1995)

A new (double cascade) scheme is proposed for laser cooling of atoms by the method of velocity-selective, coherent, population trapping. This scheme makes it possible to cool the even-even isotopes of several elements to below the recoil limit. The atoms are subjected to a radiation-pressure force which precools the atomic ensemble and confines its cooled fraction. It is shown that there exists a regime in which the cooling efficiency approaches a constant nonzero value as the interaction time increases. © *1995 American Institute of Physics.*

The method of laser cooling, based on velocity-selective coherent population trapping (VSCPT), makes it possible to reach temperatures below the recoil limit in the case of atoms which possess a Λ-scheme of levels (in the experiment of Ref. 1—metastable ^{4}He in the triplet state). The main drawback of this cooling scheme is that there is no radiation-pressure force that precools the atomic ensemble and then prevents the cooled fraction of the atoms from diffusing into the region of higher momenta. In this connection, different improved schemes with precooling, for example, a standing-wave scheme,[2] where the polarization-gradient mechanism operates in addition to VSCPT, or a scheme incorporating the Doppler mechanism of precooling,[3] have been proposed. All of these methods are based on the trapping of ^{4}He atoms in a coherent superposition of different magnetic sublevels of the long-lived metastable state 2^3S^1.

In the present paper we propose a new scheme of subrecoil laser cooling of atoms with a nondegenerate (the total angular momentum $F_g=0$) ground state $|g\rangle$. The excited states $|e\rangle$ and $|e'\rangle$ are characterized by the angular momenta $F_e=1$ and $F_{e'}=2$ and excitation energies $E_e-E_g<E_{e'}-E_g$, respectively. An ensemble of such atoms is placed in the field of two oppositely propagating and oppositely circularly polarized traveling waves. Each wave consists of two components with the frequencies ω and ω' (the wave numbers are equal to $k=\omega/c$ and $q=\omega'/c$). The first component is resonant with the transition $|g\rangle-|e\rangle$ and the second component is resonant with the transition $|e\rangle-|e'\rangle$. We incorporate recoil in the detuning from resonance:

$$\Omega=\omega-\frac{E_e-E_g}{\hbar}-\frac{\hbar k^2}{2M},\quad \Omega'=\omega'-\frac{E_{e'}-E_e}{\hbar}-\frac{\hbar q^2}{2M}-\frac{\hbar kq}{M}, \tag{1}$$

where M is the mass of the atom.

Therefore the five states $|0,p\rangle=|g,\ p_z=0\rangle$, $|1,p\rangle=|e,\ p_z=p+\hbar k,\ m_z=1\rangle$, $|1',p\rangle=|e',\ p_z=p+\hbar(k+q),\ m_z=2\rangle$, $|2,p\rangle=|e,\ p_z=p-\hbar k,\ m_z=-1\rangle$, and $|2',p\rangle=|e',\ p_z=p-\hbar(k+q),\ m_z=-2\rangle$ form a closed family (p_z and m_z are the projections of the momentum and the total angular momentum of the atom, respectively, onto the z axis.

The equation describing the evolution of the atomic density matrix $\hat{\rho}$ is

$$\frac{\partial}{\partial t}\hat{\rho}=\frac{i}{\hbar}(\hat{\rho}\hat{H}-\hat{H}\hat{\rho})-(\hat{R}\hat{\rho}+\hat{\rho}\hat{R})+\hat{\Pi}(\hat{\rho}), \tag{2}$$

where the Hamiltonian $\hat{H}=\hat{H}_{at}+\hat{K}+\hat{H}_{int}$ is the sum of the unperturbed Hamiltonian which describes the internal degrees of freedom of the atom ($\hat{H}_{at}|s\rangle=E_s|s\rangle$, where $s=g$, e, e'), of the operator $\hat{K}$ representing the kinetic energy of the translational motion of the atom, and of the atom–field interaction operator $\hat{H}_{int}$, which can be expressed most compactly in the basis of the closed families of states:

$$\hat{H}_{\text{int}}=\int_{-\infty}^{\infty}dp[\hbar V\exp(-i\omega t)(|1,p\rangle\langle 0,p|+|2,p\rangle\langle 0,p|+\hbar u\exp(-i\omega' t)(|1',p\rangle$$
$$\times\langle 1,p|+|2',p\rangle\langle 2,p|)]+\text{H.c.} \tag{3}$$

Here V and u are the Rabi frequencies for the corresponding transitions.

The exponential decay of the elements of the density matrix is described with the help of the operator

$$\hat{R}=\sum_{\mu}A_{\mu}\int_{-\infty}^{\infty}dp|\mu,p\rangle\langle\mu,p|, \tag{4}$$

where the relaxation constant is

$$A_{\mu}=\begin{cases}0, & \mu=0,\\ \gamma, & \mu=1,\ 2,\\ \Gamma, & \mu=1',\ 2'.\end{cases} \tag{5}$$

We assume that the relaxation rates satisfy the relation $\Gamma\ll\gamma$. Taking into account the fluctuations of the laser radiation[4] renormalizes the relaxation constants appearing in the equations for the off-diagonal elements of the matrix $\hat{\rho}$. Specifically, in these equations Γ must be replaced by $\Gamma'>\Gamma$. The laser line must be narrow enough so that $\Gamma'\ll\gamma$; i.e., VSCPT should be possible in the states $|0,p\rangle$, $|1',p\rangle$, and $|2',p\rangle$ with $p\approx 0$.

The last term in Eq. (2) describes the arrival of atoms in the lower levels as the upper levels relax:

$$\langle 0,p|\hat{\Pi}|0,p\rangle=2\gamma\int_{-1}^{1}d\zeta\Phi(\zeta)(\langle 1,p-\hbar k+\zeta\hbar k|\hat{\rho}|1,p-\hbar k+\zeta\hbar k\rangle+\langle 2,p+\hbar k$$
$$+\zeta\hbar k|\hat{\rho}|2,p+\hbar k+\zeta\hbar k\rangle),$$
$$\langle 1,p|\hat{\Pi}|1,p\rangle=2\Gamma\int_{-1}^{1}d\zeta\Phi(\zeta)\langle 1',p-\hbar q+\zeta\hbar q|\hat{\rho}|1',p-\hbar q+\zeta\hbar q\rangle, \tag{6}$$

$$\langle 2,p|\hat{\Pi}|2,p\rangle = 2\Gamma \int_{-1}^{1} d\zeta \Phi(\zeta) \langle 2',p+\hbar q+\zeta\hbar q|\hat{\rho}|2',p+\hbar q+\zeta\hbar q\rangle.$$

The remaining matrix elements of the operator $\hat{\Pi}$ are equal to zero. Here we disregard the difference of the wave numbers k and q from their exact resonance values. The kernel of the integral operator in Eq. (6) has the properties $\Phi(-\zeta)=\Phi(\zeta)$ and $\int_{-1}^{1}\Phi(\zeta)d\zeta=1$. The explicit form of $\Phi(\zeta)$ is presented, for example, in Ref. 5.

Our five-level scheme thus consists of two cascades[6] with a common lower level. A distinguishing feature of this scheme, in contrast to that of Ref. 6 , is that it includes atomic coherences which correspond to three- and four-photon transitions.

To solve Eq. (2) for long times we used a method similar to the one developed in Ref. 7 . It is based on the fact that the characteristic time of the translational dynamics of the atoms is much longer than the characteristic time of the dynamics of their internal degrees of freedom. We introduce the distribution function $w(p,t)$ of the atoms over closed families:

$$w(p,t)=\sum_{\mu} \langle \mu,p|\hat{\rho}|\mu,p\rangle,$$

where the summation extends over all values $\mu=0$, 1, 1′, 2, and 2′. Ignoring the nonadiabatic effects,[5] as done in Ref. 7, we express the population $\langle \mu,p|\hat{\rho}|\mu,p\rangle$ in terms of the function $w(p,t)$ as follows:

$$\langle \mu,p|\hat{\rho}|\mu,p\rangle = P_{\mu\mu}(p)w(p), \tag{7}$$

where the coefficients $P_{\mu\mu}$ are found from an algebraic system which the system (2) becomes in the indicated approximation. In what follows, we drop, for simplicity, the time argument of the function w. The true momentum distribution function $f(p)$ of the atoms can be expressed in terms of $w(p)$ as follows:

$$f(p)=P_{00}(p)w(p)+P_{11}(p-\hbar k)w(p-\hbar k)+P_{22}(p+\hbar k)w(p+\hbar k)+P_{1'1'}(p-\hbar(k+q))w(p-\hbar(k+q))+P_{2'2'}(p+\hbar(k+q))w(p+\hbar(k+q)). \tag{8}$$

The quantum kinetic equation for $w(p)$ will then have the form

$$\frac{\partial}{\partial t}w(p)=-2\gamma(P_{11}(p)+P_{22}(p))w(p)-2\Gamma(P_{1'1'}(p)+P_{2'2'}(p))w(p)$$
$$+2\gamma\int_{-1}^{1}d\zeta\Phi(\zeta)(P_{11}(p-\hbar k+\zeta\hbar k)w(p-\hbar k+\zeta\hbar k)+P_{22}(p+\hbar k+\zeta\hbar k)w(p+\hbar k+\zeta\hbar k))+2\Gamma\int_{-1}^{1}d\zeta\Phi(\zeta)(P_{1'1'}(p-\hbar q+\zeta\hbar q)w(p-\hbar q+\zeta\hbar q)+P_{2'2'}(p+\hbar q+\zeta\hbar q)w(p+\hbar q+\zeta\hbar q)). \tag{9}$$

The state of VSCPT with $p=0$ is valid if the following condition is satisfied:

$$\Omega+\Omega'=0. \tag{10}$$

This is the case that we shall examine below.

We introduce the quantities $F(p)$ and $D(p)$ which are defined as follows:

$$F(p)=2\gamma\hbar k(P_{11}(p)-P_{22}(p))+2\Gamma\hbar q(P_{1'1'}(p)-P_{2'2'}(p)), \tag{11}$$

$$D(p)=\left(1+\int_{-1}^{1}\zeta^2\Phi(\zeta)d\zeta\right)[\gamma(\hbar k)^2(P_{11}(p)-P_{22}(p))+\Gamma(\hbar q)^2(P_{1'1'}(p)$$
$$-P_{2'2'}(p))].$$

For $|p\gg\hbar k$ they represent the force and diffusion coefficients in the Fokker–Planck equation, into which the quantum kinetic equation (9) is transformed in the quasiclassical limit. Because of the symmetry of the double cascade scheme under study, we have $F(p)=-F(-p)$. At the same time, $D(p)=D(-p)$ and $D(p)>0$ for arbitrary p. It should be noted that $D(p)$ is proportional to the rate of the induced optical transitions in an atom with a given value of p, not only in the quasiclassical region but also near $p=0$, where the distribution function changes substantially over scales much smaller than $\hbar k$.

At long times, Eq. (9) can be solved by the method of matching of the asymptotic solutions in the outer (quasiclassical) and inner (quantum) regions of the momentum space.[7] The solution assumes an especially simple form in the case where the transition $|g\rangle-|e\rangle$ lies in the visible or ultraviolet range and the transition $|e'\rangle-|e\rangle$ lies in the infrared or microwave range, i.e., for $q\ll k$. Since the ratio $\Gamma P_{\mu'\mu'}(p)/\gamma P_{\mu\mu}(p)$, where $\mu=1,2$, never exceeds one in order of magnitude, for $q\ll k$ the terms associated with the excitation of atoms into the states $|1'\rangle$ and $|2'\rangle$ make a negligible contribution to F and D, and the second and fourth terms in Eq. (9) cancel one another. In other words, transitions induced by photons with the wave number q are essential for the internal coherent dynamics of an atom but have virtually no effect on the translational motion of the atom. The steady-state solution of Eq. (9) will then have the form

$$w(p)=\frac{C_0}{D(p)}\exp\left(\int_0^p\frac{F(p')dp'}{D(p')}\right), \tag{12}$$

where C_0 is the normalization constant. Of course, this solution is physically meaningful if the integral $\int_{-\infty}^{\infty}w(p)dp$ converges. If the integral $\int_{-\infty}^{\infty}p^2w(p)dp$ diverges, then Eq. (12) represents only a limit which the distribution function approaches as $t\to\infty$ and which cannot be reached over a finite time.

As $|p|\to\infty$, the asymptotic values of the force and the diffusion coefficients (11), respectively, are

$$F(p)\approx\frac{4\gamma\hbar k|V|^2\Omega}{(2\omega_R p/\hbar k)^3},\quad D(p)\approx\frac{14\gamma(\hbar k)^2|V|^2}{5(2\omega_R p/\hbar k)^2},$$

where $\omega_R=\hbar k^2/2M$ is the recoil frequency. The condition that the normalization integral converge requires that the laser frequency ω be offset from resonance by the amount (taking recoil into account)

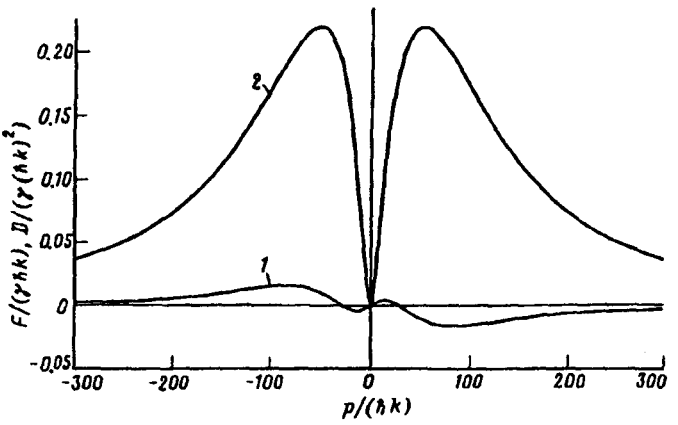

FIG. 1. Coefficients $F(p)$ and $D(p)$ for ^{200}Hg atoms cooled with use of the following interaction parameters: $V=0.15\times10^7$ s^{-1}, $u=0.15\times10^7$ s^{-1}, and $\Omega=-\Omega'=-0.5\times10^6$ s^{-1}.

$$\Omega<-\frac{21}{5}\omega_R, \tag{13}$$

i.e., into the red direction. According to Eq. (10), the frequency ω' should be offset from resonance by an amount of the same magnitude but in the blue direction.

While the exponential function in Eq. (12) causes the wings of the distribution function to decay, the pre-exponential factor $1/D(p)$ determines the effective temperature T of the cooled fraction of the atoms: $T=\delta p^2/k_BM$, where k_B is Boltzmann's constant, and δp is the half-width at half-maximum of the peak of the function $1/D(p)$ near $p=0$. Analysis shows that just as in the case of the Λ scheme,[7] for $\Gamma'<\omega_R$ it is possible to obtain temperatures below the recoil limit, $T_R=2\hbar\omega_R/k_B$. The condition $\Gamma'<\omega_R$ imposes a limit on the noise of the two independent lasers which excite the transitions $|g\rangle-|e\rangle$ and $|e\rangle-|e'\rangle$. We believe, however, that the modern experimental technology[8] makes it possible to overcome such difficulties.

The most important feature of our double cascade scheme is that it is possible in this case to obtain at long interaction times a nondecreasing efficiency, i.e., the cooled fraction of the atoms relative to the total number of atoms, despite the finite relaxation rate Γ' of the atomic coherences corresponding to four- and two-photon transitions. This circumstance is attributable to the existence of the Doppler precooling mechanism in the case considered by us. It should be noted that in the two-dimensional cooling scheme[3] the Doppler force is not confining in all directions of the momentum of the atom.

Finally, we call attention to Ref. 9 , where a five-level scheme, but of a different, inverted-W type, which uses the hyperfine structure of the ground state, was proposed for cooling ^{23}Na. In this scheme the filling of the $m_z=\pm2$ magnetic sublevels of the hyperfine component with a total angular momentum of 2 in the ground state leads to a substantial decrease in the maximum light-pressure force and, consequently, to a sharply lower limiting value of the cooling efficiency.

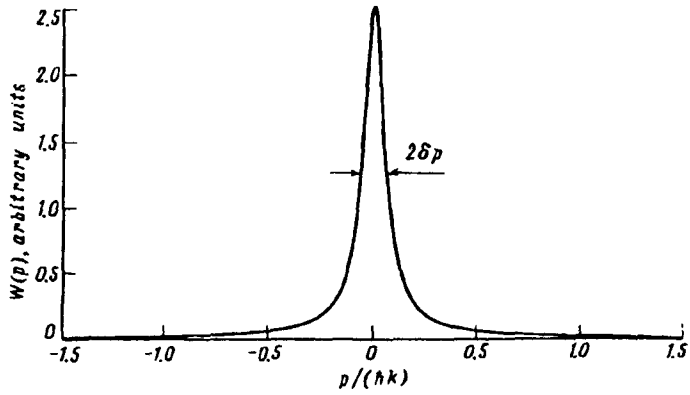

FIG. 2. Stationary distribution $w(p)$ of ^{200}Hg atoms over the closed families with the interaction parameters shown in Fig. 1. The total width $2\delta p$, which determines the effective temperature, is indicated.

Figure 1 shows the momentum dependences of the coefficients F and D for the case of the cooling of ^{200}Hg atoms. The difference of $F(p)$ from the standard Doppler force for small values of p is due to the induction of long-lived atomic coherences. Here the double cascade scheme is realized on the states 6^1S_0, $6^3P_1^0$, and $6^3P_2^0$. The spectroscopic parameters of mercury are presented, for example, in Ref. 10. Note the very short lifetime (about 10^{-7} s) of the $6^3P_1^0$ state, which decays into the nondegenerate ground state by an intercombination transition. Figure 2 shows the corresponding distribution $w(p)$ of the atoms over closed families. The effective temperature, which corresponds to the value δp found for the momentum spread, is $\sim 10^{-2} T_R \approx 10^{-8}$ K. Estimates also give typical values of 10–20% for the efficiency of cooling of the atoms by the method proposed in the present paper.

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Translated by M. E. Alferieff