Laser Cooling below a Photon Recoil with Three-Level Atoms

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We demonstrate a new cooling technique which is used to cool sodium atoms in one dimension to an effective temperature of 100 nK, less than $\frac{1}{10}$ of the single photon recoil temperature $k_B T_{\rm rec} = (\hbar k)^2/2M$.

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The cooling of atoms with light has undergone remarkable advances in recent years. Highlights include the demonstration of optical molasses [1], the discovery of sub-Doppler temperatures [2], and the realization that the cooling mechanism is the result of optical pumping in light fields with polarization gradients [3]. The minimum achievable velocity spread for polarization-gradient cooling in one dimension has been calculated to be $v_{\rm rms} \sim 6\hbar k/M$ (Ref. [4]) (k the wave vector of the light and M the atomic mass) while the experiments in three dimensions have achieved temperatures as low as $v_{\rm rms} \sim 3.5\hbar k/M$.

Helium atoms have been cooled in one dimension by trapping them in a coherent state consisting of a superposition of the Zeeman ground-state sublevels $|m_F = +1, \delta p + \hbar k\rangle$ and $|m_F = -1, \delta p - \hbar k\rangle$ [5]. Note that the atomic states are described by both an internal state m_F, and the external momentum state of the atom. The remarkable feature of this work is that the rms momentum width $\langle \delta p \rangle$ about each of the two momentum peaks at $\pm \hbar k$ corresponds to an effective temperature $\langle \delta p \rangle^2/2M = 2\ \mu$K, a factor of 2 below the single photon recoil temperature $k_B T_r = (\hbar k)^2/2M$. In this scheme, atoms were cooled by allowing them to spontaneously scatter photons until they diffused into the desired coherent state. If atoms in that state interact weakly with the light field, they have a high probability of remaining in a "trapped" velocity state while additional atoms continue to be trapped as they randomly walk in velocity space.

In this paper we present a new method for velocity space trapping and demonstrate its effectiveness by cooling sodium atoms to a 1D effective temperature of 100 nK, a factor of 10 below the recoil limit. In our scheme, sequences of velocity sensitive stimulated Raman pulses between ground-state hyperfine levels and optical-pumping pulses on an allowed optical transition are used to create and irreversibly load, respectively, a velocity trapped state. In principle the range of the velocity trapped state may be arbitrarily narrow (thus narrower than that defined by a photon recoil), while in practice it is limited by the efficiency in which atoms are located into the state.

The philosophy of cooling below the recoil temperature by an optical-pumping technique was first discussed by Pritchard *et al.* in the context of atoms confined in a magnetic trap [6]. The proposed optical-pumping method was based on the spatial dependence of the rf transition frequency between ground states of an atom whereas the velocity selective coherent population trapping scheme and our method are based on optical pumping in the velocity space of free atoms.

Stimulated Raman transitions have been used to select an ensemble of atoms with a velocity spread equivalent to 24×10^{-12} K in one dimension, but this process does not increase the density of cold atoms within a given velocity class [7]. Other schemes for cooling below the photon recoil temperature have been proposed, but not demonstrated [8–10]. In the limit where the transition linewidth is narrower than the Doppler shift due to a photon recoil, the Doppler cooling limit has been calculated to be 0.53 times the recoil temperature [11].

In our scheme, an atom with two ground states $|1\rangle$ and $|2\rangle$ is irradiated by a pulse of light from two laser beams where the beam at frequency ω_1 propagates along a direction opposite to the second beam at frequency ω_2. When $\omega_1 - \omega_2$ is nearly equal to the frequency of the $|1\rangle \rightarrow |2\rangle$ transition (see Fig. 1) and if ω_1 and ω_2 are detuned sufficiently from any optical transition, the states $|1\rangle$ and $|2\rangle$ behave as a two-level system coupled by a two-photon Raman process of effective Rabi frequency $\Omega_{\rm eff}$ [12]. The $|1\rangle \rightarrow |2\rangle$ transition has twice the Doppler sensitivity of a single photon optical transition when ω_1 and ω_2 are counterpropagating. Thus, when the frequency difference $\omega_1 - \omega_2$ is tuned to the red of the two-photon resonance, an atom moving with velocity $+v$ will

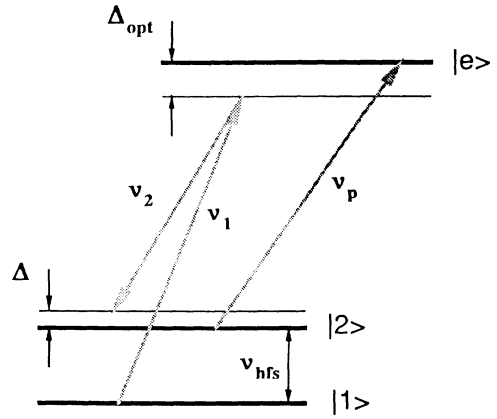

FIG. 1. Energy level diagram for Raman cooling.

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Doppler shift the transition into resonance and will receive a momentum kick of $2\hbar k$ towards $v=0$ as it makes the transition $|1\rangle \rightarrow |2\rangle$. On the other hand, if the directions of the two Raman beams are reversed, an atom moving with velocity $-v$ will Doppler shift the transition into resonance and receive a momentum kick towards $v=0$ as it makes the transition.

Using sequences of Raman pulses of varying frequency width, detuning, and propagation direction, a light field can be tailored which excites all atoms except those with a velocity near $v=0$ as shown in Fig. 2. Cooling is achieved by following the stimulated excitation with a light pulse of frequency ω_p tuned to the $|2\rangle \rightarrow |e\rangle$ resonance that optically pumps the atoms back to the $|1\rangle$ state. Each optical-pumping pulse randomizes the velocity of the atom so that a fraction of the atoms will acquire a velocity near $v=0$.

Each time a stimulated Raman-excitation–optical-pumping cycle is repeated, more atoms are pushed towards the $v=0$ state. There is a small probability that the atoms near $v=0$ will be excited by the applied laser pulses, and both the width of the velocity distribution and the number of cooled atoms will depend on the balance between the loading into the $v\sim 0$ state and the probability of unwanted excitation out of that state. If the process is to capture most of the atoms in the initial velocity distribution, atoms near $v=0$ must be able to survive the application of several hundred to several thousand pulses before being excited out of the velocity trapped state.

One of the attractive features of using Raman pulses to cool atoms is that both the *linewidth* and *line shape* of the Raman transition can be tailored to optimize the cooling. If pulses with different durations and frequencies are used, fast atoms can be pushed towards zero velocity using short pulses with large linewidths before using more velocity selective, longer duration pulses to get closer to $v=0$. Similarly, the frequency line shape of the resonance can be tailored by choosing the appropriate time evolution of the excitation pulses. For example, the square pulse excitation shown in Fig. 3 adds large sidelobes in the frequency spectrum that will produce unwanted off-resonant excitations. On the other hand, a Blackman pulse envelope [13] has a frequency spectrum with very little power away from the central excitation frequency, and the frequency spectrum is more desirable than either a $\sin^2 x/x^2$ or a Lorentzian function. We used an SRS DS 345 arbitrary wave-form generator to drive an acousto-optic modulator in order to generate a Blackman pulse intensity envelope from a cw laser beam. Figure 3 compares the measured atomic response to Blackman pulse excitation to square pulse excitation under Doppler-free conditions. The Blackman pulse decreases unwanted excitations by at least 3 orders of magnitude outside the central lobe.

The efficiency of the cooling process can be crudely estimated by the following argument. Assume the excitation line shape of a set of Raman pulses is a square well for which atoms with velocity $|v| < v_0$ are trapped and atoms with velocity $|v| > v_0$ are excited with probability p_e to the $|2\rangle$ state. The atoms' steady-state velocity spread σ (symmetric about zero velocity) is determined by the extent to which their velocity is randomized during the optical-pumping cycle and by the size of the stimulated kick. When $v_0 \ll \sigma$ the fraction of atoms caught in the trapped state after each Raman-pulse–optical-pumping pulse cycle is roughly given by the ratio of the velocity width of the trapped state to the width of the untrapped distribution. Thus, the number of atoms in the untrapped state exponentially decays with time constant $(t_{\rm eq}/\tau)^{-1} \approx p_e(v_0/\sigma)^n$, where τ is the duration of the set of Raman pulses shown in Fig. 2, and $n=1$, 2, or 3 is the number of dimensions for the trapping state.

As a numerical example, we insert the parameters of our one-dimensional experiment, $p_e \sim 0.7$, $\sigma \sim 2\hbar k/M$, and we cooled atoms to a velocity width of $0.3\hbar k/M$. The total time used for each sixteen-pulse cycle was $\tau = 300$ μsec, giving a predicted equilibrium time of 3 msec. The optical-pumping time of an atom is limited by the lifetime of the optical transitions (16 nsec for Na) and thus contributes negligibly to the total cooling time.

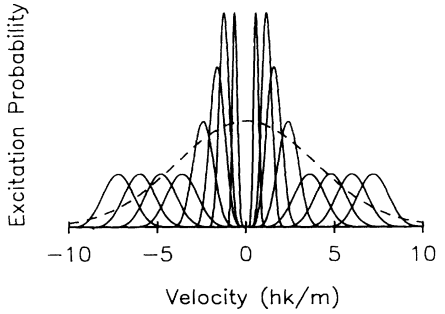

FIG. 2. The probability of excitation vs velocity for each of the sixteen sequentially applied stimulated Raman cooling pulses (solid lines) and the initial velocity distribution of atoms cooled in the polarization-gradient molasses (dashed line).

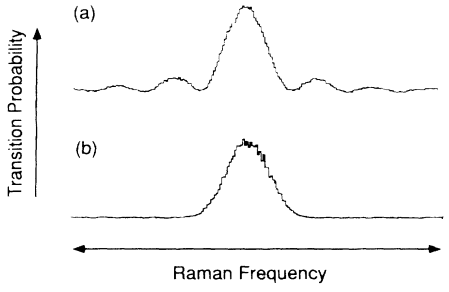

FIG. 3. Measured excitation probability vs frequency detuning for both (a) a square wave and (b) a Blackman pulse (defined as $\Omega(t) = \Omega_{\rm eff}\{0.5\cos(\pi[2t/\tau - 1]) + 0.08\cos(2\pi[2t/\tau - 1]) + 0.42\}$ for $0 \le t \le \tau$).

In reality, there will be a small probability that atoms in the velocity trapped state can be ejected from that state into the untrapped distribution. The above model can be readily extended to include this effect by adding an extra term to account for the residual excitation rate, with the result that in steady state a fraction of the initial distribution resides in the velocity trapped state. The ejection might be due to off-resonant stimulated excitation from the Raman pulses, excitation to the $P_{3/2}$ state by either of the two Raman beams or the optical-pumping pulse, or absorption of a spontaneous Raman photon emitted during the optical-pumping cycle. None of these detrimental effects presents an insurmountable experimental obstacle. For example, off-resonant excitation during the Raman pulses can be reduced by increasing the detuning from the intermediate level.

The cooling experiment was done as follows: $\sim 10^7$ atoms were loaded into an optomagnetic trap [14] from a slowed Na beam. The atomic beam was slowed by a counterpropagating, frequency chirped laser. After collecting atoms for 160 msec the magnetic field used to trap the atoms was shut off, and the atoms were further cooled in polarization-gradient optical molasses to a temperature of ~ 35 μK and then optically pumped into the $F=1$ ground state of sodium. 5 msec after extinguishing the molasses light and approximately 25 msec after the field coils were switched off, the Raman cooling cycle was initiated. The velocity distribution of the atoms was typically sampled 10 msec later as described below.

The cooling and trapping beams were supplied by one cw dye laser while the two Raman laser beams were derived from a second dye laser tuned ~ 30 GHz below the $F=2 \rightarrow 3$ resonance. The ~ 1-W output beam of the Raman laser was divided into two beams of nearly equivalent power with an ~ 30-MHz acousto-optic modulator. One of these beams passed through an ~ 1.7-GHz electro-optic phase modulator to generate the second Raman frequency. The two beams were then made to nearly copropagate through a second acousto-optic modulator which was used to control the amplitude (hence Raman envelope) of the light. The Raman frequency was switched by changing the frequency used to drive the electro-optic modulator. Finally, the beams were separated, collimated to an ~ 4-mm $1/e^2$ diameter and then directed along counterpropagating paths to the ~ 4-mm $1/e$-diam trap [15]. The peak Rabi frequency for the Raman transitions was $\Omega_{\text{eff}} \sim 50$ kHz.

Each Raman beam was linearly polarized, with the polarization axes taken to be orthogonal to one another. This ensured roughly equal ac Stark shifts for all hyperfine Zeeman sublevels [16]. The magnetic field during the Raman cooling phase was less than 5 mG, verified by driving the Doppler-free transitions. Both of the above measures were taken to suppress shifts of the Zeeman states which might lead to unwanted residual excitation from the trapped state.

The optical-pumping beam was derived from the molasses laser, and was frequency shifted to be approximately resonant with the $3S_{1/2}$, $F=2 \rightarrow 3P_{3/2}$, $F=2$ transition. An acousto-optic modulator switched on the optical-pumping beam. The beam intensity was ~ 100 mW/cm^2 in the interaction region. The propagation direction of the pumping beam was switched from copropagating to counterpropagating with a Pockels cell and polarizer and intersected the Raman beam axis at a 5° angle.

A sequence of eight Raman plus optical-pumping pulses was directed along a horizontal axis x and tuned to excite atoms with velocities $v_x < 0$ so that they would be pushed towards $v_x = 0$. Each pulse in the sequence was tuned closer to the resonance line and the pulse widths of the last four pulses were increased in time so that the spectral width of each pulse was narrowed as shown in Fig. 2. A similar eight-pulse sequence was then applied with the propagation directions of the Raman and optical-pumping beams reversed in order to push atoms with $v_x > 0$ towards $v_x = 0$. When the Raman pulses were tuned more than $2\hbar k$ away from the line, the optical-pumping pulse was directed in the same direction as the velocity kick of the Raman pulse. For smaller Raman detunings, we found that the best results were obtained when the direction of the optical-pumping pulse momentum kick was reversed with respect to the direction of the Raman momentum kick.

After a selected number of pulse cycles (each cycle consisting of the sixteen-pulse sequence described above), the number of atoms at a given velocity was measured by selectively exciting the $3S_{1/2}$, $F=1 \rightarrow 3P_{3/2}$, $F=2$ transition with another velocity selective Raman pulse and then observing the number of atoms making the transition to the $F=2$ state by measuring the fluorescence from a short pulse of light resonant with the $3S_{1/2}$, $F=2 \rightarrow 3P_{3/2}$, $F=3$ transition. The velocity distribution was determined by scanning the frequency of the Raman pulse across the Doppler broadened line for a number of identically prepared sets of atoms [7]. The velocity resolution of this detection scheme was $0.1\hbar k/M$. The results of such a frequency scan after nineteen pulse cycles (5.7 msec) are shown in Fig. 4. Approximately 50% of the originally trapped atoms with a velocity spread of $\Delta v_{\text{rms}} \sim 4\hbar k/M$ have been compressed into a single velocity spike characterized by an rms velocity of $\Delta v_{\text{rms}} \sim 0.2\hbar k/M$. This corresponds to an effective temperature of ~ 100 nK, a factor of ~ 10 below the single photon recoil limit and a factor of 400 below the temperature obtained with polarization-gradient molasses. The $1/e$ equilibrium time for loading atoms into the trapped state for these data was measured to be ~ 1.5 msec.

The fact that we were able to load only half of the initial set of atoms into the velocity trapped state indicates a residual loss rate from the trapped state. We believe off-resonant excitation during the optical-pumping pulses to

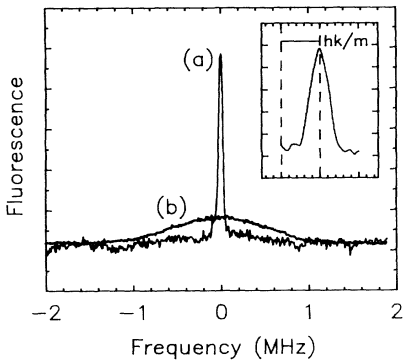

FIG. 4. (a) The velocity distribution after application of the stimulated Raman cooling pulses. The inset, showing a high resolution scan of the central velocity spike, compares the velocity distribution to the velocity change $\Delta v = 3$ cm/sec from the recoil of a single photon. (b) The initial velocity distribution of sodium atoms due to polarization-gradient cooling. A uniform background signal ~ 3 times the size of the peak signal for curve *b* has been subtracted from curve *a*. The background was due to incomplete optical pumping from $F=2 \rightarrow F=1$ during the Raman cooling sequence, and is responsible for the increased noise on curve *a*.

be the dominant loss mechanism, and we are currently working to eliminate this effect. By increasing the extent of the velocity trapping region to yield an effective temperature of T_{rec} we were able to trap nearly all of the atoms from the initial distribution.

This cooling scheme can also be extended to two and three dimensions. It is possible to cool in several dimensions simultaneously with Raman beams from all directions pulsed on simultaneously despite the occurrence of multibeam diffraction effects. The details of both our analytic calculations and Monte Carlo simulations and the comparison of these calculations with our experimental results will be presented elsewhere. The simulations and our experiments suggest that we should be able to cool atoms in two dimensions to an effective temperature of $0.06T_{\text{rec}}$ and in three dimensions to less than $0.25T_{\text{rec}}$ in less than 10 msec.

The Raman cooling technique may provide a powerful method of achieving high densities of cold atoms. For example, sodium atoms initially precooled and trapped in a optomagnetic trap can provide an initial sample with a density in excess of 10^{11} atoms/cm^3 and a temperature of 35 μK. Subsequent Raman cooling could reduce the temperature to ~ 300 nK (in three dimensions) in 10 msec, leaving 50% of the initial distribution atoms in the same internal quantum state with a negligible reduction in spatial density.

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[15] The trap diameter was measured 5 msec after the molasses light was turned off.
[16] This was verified experimentally by driving the Doppler-free transitions. The level shifts were measured to be less than 0.125 of the effective Raman Rabi frequency. In the limit of detuning much larger than the ground-state and excited-state hyperfine splittings, all level shifts are equal.

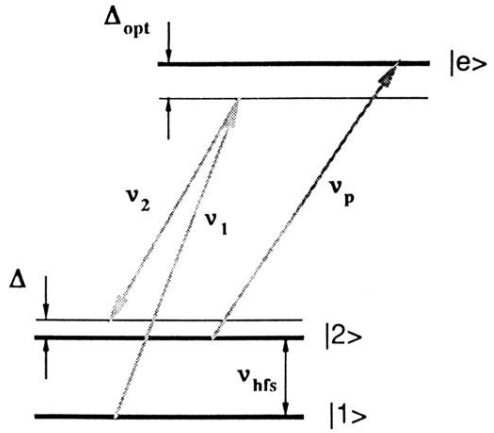

FIG. 1. Energy level diagram for Raman cooling.