Quenched narrow-line laser cooling of ⁴⁰Ca to near the photon recoil limit

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We present a cooling method that should be generally applicable to atoms with narrow optical transitions. This technique uses velocity-selective pulses to drive atoms towards a zero-velocity dark state and then quenches the excited state to increase the cooling rate. We demonstrate this technique of quenched narrow-line cooling by reducing the 1D temperature of a sample of neutral ⁴⁰Ca atoms. We cool selected velocities with the ${}^{1}S_{0}(4s^{2}) \rightarrow {}^{3}P_{1}(4s4p)$ 657-nm intercombination line and quench with the ${}^{3}P_{1}(4s4p) \rightarrow {}^{1}S_{0}(4s5s)$ intercombination line at 553 nm, which increases the cooling rate eightfold. Limited only by available quenching laser power, we have transferred 18% of the atoms from our initial 2-mK velocity distribution and achieved temperatures as low as 4 μ K, corresponding to a v_{rms} of 2.8 cm/s or 2 recoils at 657 nm. This cooling technique, which is closely related to Raman cooling, can be extended to three dimensions.

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Following the great success of sub-Doppler laser cooling techniques with alkali-metal atoms, several groups are now striving to achieve similar cooling results for the alkalineearth-metal atoms, with applications ranging from Bose-Einstein condensation to atomic interferometry and optical frequency standards. Unfortunately, the lack of magnetic and hyperfine substructure in the ground states of the predominant alkaline-earth-metal isotopes precludes the usual sub-Doppler mechanisms such as polarization-gradient cooling [1,2], velocity-selective coherent population trapping [3], or Raman cooling [4]. Furthermore, the broad cooling transitions for the alkaline-earth-metal atoms have large Doppler limits, and typically yield magneto-optic trap (MOT) temperatures of more than 1 mK [5-7]. These complications have necessitated the development of new cooling strategies to reach the microkelvin temperatures "routinely" achieved with several alkali-metal species. This paper presents an alternative approach, termed "quenched narrow-line laser cooling" (QNLC), which uses a narrow optical transition to provide increased velocity selectivity for laser cooling, and quenching of the upper state of this transition to enhance the cooling efficiency. To demonstrate the potential of this technique, we have used repeated velocity-selective stepwise excitation with two intercombination lines of neutral ⁴⁰Ca to transfer as much as 18% of the atoms in an initial velocity distribution with a 2 mK temperature into a narrow peak. The lowest one-dimensional (1D) temperature observed was 4 μ K with a corresponding transfer efficiency of 7%. Moreover, these results can be significantly improved simply by increasing the quenching power, and can be readily extended to two or three dimensions. These greatly reduced temperatures should significantly reduce the uncertainty in Ca-based optical frequency standards, for which a recent measurement of the absolute frequency was principally limited by residual atomic velocity [8].

Our approach to second-stage cooling takes advantage of the two-electron structure of these atoms, which gives rise to

narrow intercombination lines. In principle, these transitions can serve as excellent frequency or velocity (via the firstorder Doppler shift) discriminators, which are ideal for subrecoil laser cooling. The spectacular results achieved with the intercombination line of ⁸⁸Sr, for which temperatures of <700 nK have been attained, confirm the potential of these transitions [9,10]. Unfortunately, for several species, such as Ca and Mg, the intercombination line is too weak to effectively cool a distribution with an initial temperature of several millikelvins. Nonetheless, Binnewies et al. [11] were able to use the velocity selectivity of the Ca intercombination line at 657 nm to demonstrate a new cooling mechanism, Maxwell-demon cooling. They were able to achieve a net transfer of 5% of the initial distribution into a peak with a 1D temperature of $<10 \ \mu$ K. However, this technique does not seem to lend itself to three-dimensional (3D) cooling, which is critical for many applications.

A more versatile approach is to increase the cooling efficiency by quenching the upper level of the narrow transition through excitation to another state that decays more quickly to the ground state. In Fig. 1 we show how we quench the metastable ${}^{3}P_{1}$ state in Ca (lifetime=400 μ s) through excitation to the ${}^{1}S_{0}(4s5s)$ level (lifetime=30 ns) via the intercombination line at 553 nm. The enhancement of narrow-line cooling rates through quenching was first demonstrated in the context of sideband cooling of trapped ions [12,13]. When applied to neutral atoms, quenching can increase the



FIG. 1. Relevant energy level diagram for QNLC in ⁴⁰Ca. The 553-nm quenching beam transfers population from the long-lived (400 μ s) ³*P*₁(4*s*4*p*) state to the ground state via the short-lived (30 ns) ¹*S*₀(4*s*5*s*) state.

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cooling efficiency not only by speeding up the rate at which one repeats cooling cycles, but also by giving the atoms additional momentum kicks towards zero velocity. These advantages were used to increase the force on a metastable He atomic beam by excitation of a two-photon transition with simultaneous light fields [14].

To take advantage of the velocity selectivity of the narrow transition, one approach is to excite the atoms in a stepwise manner with time-separated pulses. One can then implement the velocity-selective excitation strategies that have been used with great success in Raman cooling. In the first demonstration of Raman cooling, Kasevich and Chu used a Raman transition between hyperfine ground states to cool Na atoms to a 1D temperature of 100 nK and a 3D temperature of 4.3 μ K [4,15]. Later Reichel *et al.* used square Ramancooling pulses to achieve a 1D temperature of 3 nK for Cs [16].

For quenching times short compared to excitation times used for the narrow transition, we propose the following QNLC procedure and illustrate it with the Ca system. We start by pumping a velocity slice towards zero velocity using light whose frequency is tuned slightly red of the ${}^{1}S_{0} \rightarrow {}^{3}P_{1}$ intercombination line at 657 nm (see Fig. 1). Due to the narrow linewidth of this transition (400 Hz), the spectra of our red pulses are Fourier-transform limited, so we can easily control the width and shape of the velocity group excited. We then quench the ${}^{3}P_{1}$ population with a 553-nm light pulse (copropagating with the red pump beam), which moves the population to the ${}^{1}S_{0}(4s5s)$ state and gives the velocity-selected atoms a second momentum kick towards zero velocity. From this state the atoms quickly return to the ground state via two cascaded decays (see Fig. 1) with their associated (randomly directed) recoils. Next we use a similar sequence of red and green pulses from the opposite direction to pump a second velocity slice, symmetrically located on the opposite side of the distribution, towards zero. A single cooling cycle can thus reduce atomic velocities on both sides of the distribution by two-photon momenta (analogous to the case of Raman cooling). We repeat this cooling cycle many times, driving the atoms towards zero velocity, which coincides with a zero in our sinc² Fourier-transform-limited excitation function. In this way, atoms that accumulate around zero velocity have only a small probability of being pumped away, so in principle, extremely low temperatures can be achieved. As in the case of Raman cooling, the cooling limit is set by the cooling time and the width of the small transition-probability region around zero velocity, but not by the random recoils. Narrower sinc² functions (i.e., longer red pulses in the time domain) yield colder temperatures, but they address a smaller range of velocities, so there is a compromise between temperature and number, unless cooling with a sequence of pulse widths is used [4,15,16].

In our experimental demonstration of this technique, inadequate light power at 553 nm (limited by what could be transferred through a 180-m fiber) led to quenching times that were an order of magnitude longer than the velocityselective cooling pulses. Under these conditions it was more efficient to use a slightly modified version of the procedure just described, in which we use a standing-wave pulse of 553





FIG. 2. Timing diagram for our experimental realization of QNLC. Arrows represent the relative direction of the k vector for each pulse. For short quenching times it would be more efficient to use a copropagating 553-nm pulse after each 657-nm pulse.

nm light to quench the ${}^{3}P_{1}$ state after each pair of counterpropagating red pulse excitations. The drawback to this approach is the randomness introduced in the direction of the momentum kick from the quenching pulse. This loss, however, is more than offset in our case by the increased intensity of a standing (rather than traveling) wave that reduces the quenching time and the number of quenching pulses (by two) used per cooling cycle.

Our experimental realization of this cooling method (see Fig. 2) begins with an \sim 7-ms loading cycle, during which we load 10^7 atoms from a Ca beam into a MOT (see Ref. [17] for details of our diode-laser based MOT) using the 423-nm ${}^{1}S_{0} \leftrightarrow {}^{1}P_{1}$ cooling transition (see Fig. 1). After this first cooling stage the temperature of the atomic sample is about 2 mK (corresponding to a v_{rms} of 0.6 m/s), slightly above the 0.8-mK Doppler limit. To avoid large light shifts of the ground state that would compromise the velocity selectivity of the red cooling transition, we turn off the blue trapping light for the duration of the second-stage cooling. We then commence QNLC with a pair of counterpropagating red pulses (2.5 μ s square-shaped π pulses), separated by 2 μ s in time. This 657-nm light is spatially filtered with an optical fiber and collimated to a diameter of 4 mm (8 mW in each beam). In order to excite the $m=0 \rightarrow m=0$ transition, the light is linearly polarized parallel to the dominant B-field direction (due to a trap imbalance, the atoms rest at a 6-G point in our magnetic-field gradient, which remains on during the entire measurement). We tune the laser to a frequency \sim 350 kHz (corresponding to a velocity of 23 cm/s) red of resonance so that the first zero of the sinc² frequency spectrum is within 5 kHz of resonance for atoms at rest. The frequency of the red light is stabilized to an environmentally isolated, high-finesse Fabry-Perot cavity resonance, whose drift is cancelled to less than 0.3 kHz/minute (typical data averaging times were about 1 minute). After the first pair of counterpropagating red pulses, atoms that were transferred to the excited state are pumped to the ${}^{1}S_{0}(4s5s) m=0$ state by a 553-nm pulse (duration of \sim 50 μ s) with an efficiency of \sim 50%. As a result of the small transition rate (we estimate $\Gamma[{}^{1}S_{0}(4s5s) \rightarrow {}^{3}P_{1}(4s4p)] \sim 2440(600) \text{ s}^{-1}), \text{ we could}$

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FIG. 3. Velocity distribution of 40 Ca atoms before (dotted line) and after (solid line) 15 second-stage cooling cycles. Each cycle consists of two 2.5- μ s red pulse pairs each followed by 50 μ s of 553-nm quenching light. Finally, a 553-nm light pulse (duration of 150 μ s) is used to pump remaining atoms back to the ground state. The red velocity probe pulse is 10 μ s long. We transfer about 15% of our atoms into a narrow peak with a v_{rms} of 5.9 cm/s, corresponding to 4 recoils or a temperature of 17 μ K. This width represents a convolution of the probe and the true velocity distribution.

only attain a 1/e quenching time of 55 μ s, with 17 mW of green light collimated to a standing wave diameter of 3 mm. The frequency-stabilized green light is generated by a dye laser in another laboratory and sent to us via optical fiber. This light intersects the atoms at an angle of $\sim 8^{\circ}$ relative to the red cooling beams. From the ${}^{1}S_{0}(4s5s)$ state the atoms decay rapidly (<35 ns) to the ground state by way of the ${}^{1}P_{1}$ state. Then follows a second pair of counterpropagating red pulses, but this time in reverse order (see Fig. 2) to make the overall cooling process more symmetric. Due to the lobes of the sinc² frequency spectrum, there is some probability that the second pulse of a pulse pair can transfer atoms excited by the first red pulse back to the ground state, making the cooling due to the first pulse slightly less efficient. After the second pair of red pulses, a second green quenching pulse follows to complete the cooling cycle. We then repeat the cooling cycle (consisting of two pairs of counterpropagating pulses, each followed by a standing-wave quenching pulse) eight to twenty times. At the end of this sequence we clean out the ${}^{3}P_{1}$ state with an extra green pulse lasting 100 to 150 μ s that leaves less than 3% of the population in the excited state. We then measure the velocity distribution.

Our velocity probe consists of a single 657-nm pulse (10 or 20 μ s in duration, depending on our desired spectral resolution) that excites a narrow velocity slice of atoms to the excited state. It excites the $m=0 \rightarrow m=0$ transition, and is collinear with the red cooling beams. While continuously cycling the complete measurement sequence, we slowly sweep the frequency of the velocity probe over the atomic velocity range to generate our distributions. We typically average 60 sweeps of 0.5 s duration to generate a data set. In order to achieve a good signal-to-noise ratio for these velocity distribution measurements, we use two 423-nm probe pulses in a normalized shelving detection scheme that we developed for our optical clock (see Fig. 2) [17].

In Fig. 3 we show the effect of 15 cooling cycles taken with a red pulse length of 2.5 μ s and a green pulse length of 50 μ s. We see that the QNLC process transfers most of the



FIG. 4. Velocity distribution as in Fig. 3 except with ten secondstage cooling cycles with increased cooling pulse duration (5 μ s) and a higher resolution velocity probe (20- μ s duration). A post cooling pulse of 553-nm light (duration of 100 μ s) is used to pump remaining atoms back to the ground state. The narrow peak has a v_{rms} of 3.3 cm/s, corresponding to ~2 recoils or 5.3 μ K. This width represents a convolution of the probe with the actual velocity distribution.

atoms in the range of 30 cm/s into the narrow peak at zero velocity, consistent with the expected momentum transfer from our cooling pulses. A Gaussian fit to the central peak yielded a v_{rms} of 6 cm/s, or about 4 recoils at 657 nm, corresponding to a 1D temperature of 17 μ K. The transfer efficiency can be estimated by comparing the area of the peak to the area of the initial distribution-in this case we find 30% transfer. However, since approximately half of the atoms have escaped the interaction region since we first turned off the trapping beams, we have a net efficiency closer to 15% (we have seen net efficiencies as high as 18%). Fast ballistic expansion due to our warm initial temperature causes the atoms to move into weaker parts of the laser beams during the cooling and probe periods. Because of laser power constraints, we are unable to increase the beam sizes, thus the number of cooling cycles we can use is severely limited.

Indeed, when more cooling cycles are used, we see increased transfer efficiency and narrower distributions (as was demonstrated in Raman cooling experiments [4,15,16]). However the longer cooling time required means more atoms are lost transverse to the cooling direction. We experimented with using more cooling cycles with less efficient quenching and found that the central peak was fairly insensitive over the range from 18 cycles with 30- μ s quenching time to eight cycles with 70- μ s quenching time (keeping the total cooling time approximately fixed at 1.5 ms).

The 14-cm/s width of the central peak in Fig. 3 is a result of the convolution of the velocity distribution and the velocity probe. Data taken at higher probe resolution indicate that v_{rms} for these conditions is less than 4 cm/s (7.5 μ K). To strive for colder temperatures, we increased the red pulse length to 5 μ s to give a sharper velocity discriminator. Figure 4 shows a velocity distribution resulting from ten cooling cycles using these longer cooling pulses. To keep the 657-nm pulse area constant for these measurements, we chose to double the size of the red beams rather than reduce their power in an effort to increase their pumping efficiency. We also adjusted the detuning to ~ -173 kHz (corresponding to 11.4 cm/s) to place the zero of the excitation function at zero velocity. Indeed, we see narrower distributions under these conditions (we estimate a deconvolved value of $v_{rms} < 2.8$ cm/s, corresponding to temperature of 3.7 μ K) and the expected smaller fraction of atoms in the peak (7% net efficiency) due to the reduced range of velocities covered. We also see quite clearly several other nodes of the sinc² function that serve as additional dark velocities where the atoms can accumulate. We fully expect that at higher resolutions we can achieve subrecoil temperatures, as was seen in the Raman cooling experiments. Monte Carlo simulations of the cooling process (including the recoils at 553 nm, 1.03 μ m, and 423 nm) support this expectation.

From these results, a strategy that can cool a large fraction of the initial velocity distribution to very cold temperatures becomes evident, namely, to use a series of pulses at different resolutions and detunings [4,15,16]. To be able to exploit such a strategy, however, we need to accelerate the cooling process. With simple scaling of the 553-nm power, we calculate that a fifteenfold increase (dye lasers regularly supply 20 times the power we currently send to our atoms) would enable us to cool the atoms nine times faster. Results from a Monte Carlo simulation assuming a 1/e pumping time of 4 μ s and using multiple red cooling pulses ranging from 2.5 to 5 μ s in duration indicate that we can expect to transfer 50% of the atoms into a peak with subrecoil temperature.

Moreover, one can envision a feasible 3D strategy in which one cools alternately in three dimensions [15], with each reduction in temperature effectively increasing the available cooling time. Due to the multirecoils involved in this experiment, it would be hard to achieve 3D subrecoil temperatures due to recoil heating in transverse dimensions. However, with appropriate shaping of our red pulses [15,16] a limit of several recoils seems quite feasible, especially in light of the near-recoil temperatures achieved with 3D Raman cooling [15]. Furthermore, as one nears the recoil limit and presumably has increased the cooling time available, one could remove the quenching and use the decay of the ${}^{3}P_{1}$ state itself to reduce the recoil effect. Regardless, 3D temperatures of a few microkelvins rather than millikelvins would greatly benefit the Ca optical frequency standard, atomic interferometry, and other applications.

In conclusion, we have proposed a second-stage cooling scheme based on quenched narrow-line laser cooling. We have used this technique to reduce the temperature of a ⁴⁰Ca atomic cloud in 1D by a factor of 500 in a cooling time of 1.5 ms. With ample quenching power, near subrecoil temperatures in 3D should be possible with greater than 10% net transfer efficiency. Moreover, this technique should be applicable to other atoms (such as Mg) with narrow optical transitions and available quenching transitions.

Note added. Recently, a different approach to quenched narrow line cooling was demonstrated. In this experiment a 3D temperature of 6 μ K was achieved for Ca in a cooling time of 25 ms [18].

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