Laser-Cooled Cs Frequency Standard and a Measurement of the Frequency Shift due to Ultracold Collisions

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We demonstrate a laser-cooled Cs fountain which can be used as a frequency standard. The fountain has been used to measure the frequency shift due to ultracold Cs-Cs collisions at temperatures below 2.8 μK. The measured shift is -12.9 ± 0.7 mHz for a density of $(2.7 \pm 1.5) \times 10^9$ cm^{-3} beginning with an even distribution ($\pm 10\%$) among the $F=3$, m_F states. For a fountain with 95% of the atoms in the $m_F=0$ states, we measure a frequency shift of -5.5 ± 0.5 mHz at a density of $(3.5 \pm 2.0) \times 10^8$ cm^{-3}.

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The benefits of slow atoms for precision spectroscopy and atomic frequency standards have been recognized for many years. By using laser-cooled atoms at μK temperatures, it may be possible to improve the accuracy and stability of cesium frequency standards by 2–3 orders of magnitude to $\delta\nu/\nu \leq 10^{-16}$ [1]. This improvement is due to a longer interaction time which allows an ≈ 40-fold reduction in the transition linewidth and a reduction of nearly all of the known systematic frequency shifts.

However, the use of cold atoms may introduce new problems since atomic collisions are different at the μK temperatures achievable with laser cooling. While novel cold collisions between ground- and excited-state atoms have been studied by many groups [2], cold collisions between two ground-state atoms have only been studied in cryogenically cooled H and He [3], and during the thermalization of laser-cooled Cs in a magnetic trap [4]. At μK temperatures, collisions between ground-state alkali atoms are unusual since the atomic de Broglie wavelengths λ_{dB} are much larger than the scale of the interatomic potential. In this ultracold regime, the scattering is intrinsically quantum mechanical and is usually dominated by the lowest allowed partial angular momentum wave (*s* waves) [5]. These cross sections can be as large as λ_{dB}^2/π.

The ultracold collisions may also produce sizable frequency shifts in frequency standards. The frequency shift results from unequal collision-induced (*s*-wave) energy shifts of the two hyperfine states of a "clock transition." Recently, the frequency shift for the 9.2 GHz Cs transition has been calculated by Tiesinga *et al.* to be on the order of mHz (versus the previously expected μHz accuracy) for a Cs density of 10^9 cm^{-3} [6]. In this Letter, we describe our atomic fountain and measurements of the frequency shifts due to collisions of Cs atoms at temperatures below 2.8 μK.

Our atomic fountain, depicted in Fig. 1, extends the work of Kasevich *et al.* and Clairon *et al.* [7]. The lower vacuum chamber contains room-temperature Cs atoms at a density of $\approx 10^8$ cm^{-3}. Atoms with speeds less than 30 m/s are slowed by six 6 cm diam circularly polarized laser beams and captured in a magneto-optic trap [8]. With 160 mW in each beam, we load $\approx 10^9$ atoms into a 20 mm^3 volume in 0.4 s. Collisions between trapped atoms and room-temperature atoms produce a 1.2 s trap lifetime.

After loading the trap, the atoms are launched upwards by changing the frequency of the three (orthogonal) upward propagating laser beams [9] using an acousto-optic modulator driven by a frequency synthesizer. The 1.2 MHz frequency difference between the upper and lower laser beams cools the atoms into a frame moving upward at 0.9 m/s within 1 ms. The atoms are then further accelerated by increasing the frequency difference to 3.45 MHz. After 2 ms, the intensity of the beams is reduced and the detuning of the laser beams is changed from -22 to -65 MHz. In this way, the atoms are cooled using polarization gradients [10] to a temperature of 2.8 μK ($\Delta v_{HWHM} = 1.6$ cm/s) in a frame moving upwards at 2.5 m/s.

After launching the atoms, a retroreflected laser beam

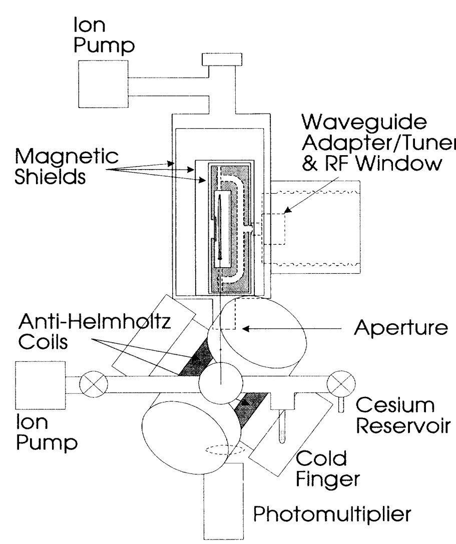

FIG. 1. Fountain schematic. Atoms are trapped in the lower vacuum chamber and launched through the microwave cavity in the upper chamber. Both chambers are pumped by 8 l/s pumps and are separated by a 1.1 cm diam aperture. Microwaves are coupled to the cavity through a coaxial cable connected to the waveguide adapter or tuner and a ceramic vacuum window.

tuned 75 MHz below the $6S_{1/2},F=4 \rightarrow 6P_{3/2},F'=4'$ transition optically pumps a variable fraction of the atoms from the $F=4$ ground state into the $F=3$ ground state. Any unwanted atoms which remain in the $F=4$ state are removed using ≈ 2000 photon recoils from a 1 ms pulse of a laser beam tuned to the $6S_{1/2},F=4 \rightarrow 6P_{3/2},F'=5'$ transition. Since the atoms optically pumped into the $F=3$ state scattered at most a few photons, we are able to vary the cold atom density without appreciably affecting the temperature or spatial distributions of atoms.

The atoms in the $F=3$ state continue to travel ballistically upwards and pass through one arm of the microwave cavity which excites the $\nu \equiv 9$, 192, 631, and 770 Hz ground-state hyperfine transition. The cavity and magnetic shields shown in Fig. 1 were obtained from a Hewlett-Packard 5061 high performance Cs frequency standard. The cavity has a loaded Q of 10^3 and is tuned with a dielectric stub and loss element in the coax-to-waveguide adapter or tuner. In our experiments, only the lower arm of the cavity is normally used.

The microwave frequency used to drive the cavity originates from a 5 MHz quartz oscillator which is phase locked to a hydrogen maser (Sigma Tau Corporation MHM-A1). The H maser has a short-term frequency stability of 2×10^{-13} after 1 s of averaging and 2.3×10^{-15} after 1000 s of averaging. The 9.192 GHz used to excite the Cs atoms is generated by phase coherently multiplying the 5 MHz maser output to 180 MHz. Using this, a step recovery diode generates a comb of coherent frequencies which are filtered to give a carrier at 9.180 GHz. This signal is mixed with 12.631... MHz from a HP 3335A frequency synthesizer which is also phase locked to the H maser. The synthesizer output level and attenuators are used to adjust the power at 9.192 GHz to ≈ -50 dBm (10 nW) so that the atoms receive a $\pi/2$ pulse in a transit through the lower cavity arm.

After the atoms travel to an apogee below the upper cavity, they return through the lower cavity. The atoms which complete the transition from the $F=3$ to the $F=4$ ground state are detected with a 500 μs pulse of a retroreflected circularly polarized, 0.4 cm diam, intensity stabilized laser beam tuned to the $6S_{1/2},F=4 \rightarrow 6P_{3/2},F'=5'$ "cycling" transition. The atomic fluorescence, detected with a photomultiplier tube, is normalized by dividing by the total number of atoms which return through the cavity. The total number of atoms (constant to within $\pm 2\%$) is determined by optically pumping the remaining $F=3$ atoms into the $F=4$ states with a laser tuned to the $6S_{1/2},F=3 \rightarrow 6P_{3/2},F'=4'$ transition and again detecting fluorescence with a second 500 μs pulse of light tuned to the cycling transition.

The full clock spectrum of the seven $\Delta m_F=0$ transitions is shown in Fig. 2(a). Transitions with $\Delta m_F=0$ are favored since the oscillating magnetic field is largely parallel to an applied 860 μG dc magnetic field generated by passing a current through twelve turns of wire wrapped around the microwave cavity structure. While the contrast of the $m_F=0$ fringes is $\approx 90\%$, the fringe contrast for the $m_F=\pm 1$ field sensitive transitions in Fig. 2(a) is 25% and no fringes are clearly visible with the $m_F=\pm 3 \rightarrow m_F=\pm 3$ transitions. This reduction in contrast is due to magnetic field inhomogeneities on the order of 3 μG for the different trajectories of the atoms in the fountain. In Fig. 2(a), the baseline structure between the large peaks is caused by the Fourier spectrum of frequencies seen by an atom during a transit through the cavity and also by the small amplitude for $\Delta m_F=\pm 1$ transitions.

The central Ramsey fringes for the $F=3,m_F=0 \rightarrow F=4,m_F=0$ clock transition are shown in Fig. 3. The FWHM linewidth is $\Delta\nu=1.41$ Hz for a mean time of 0.354 s between microwave field interactions in the cavi-

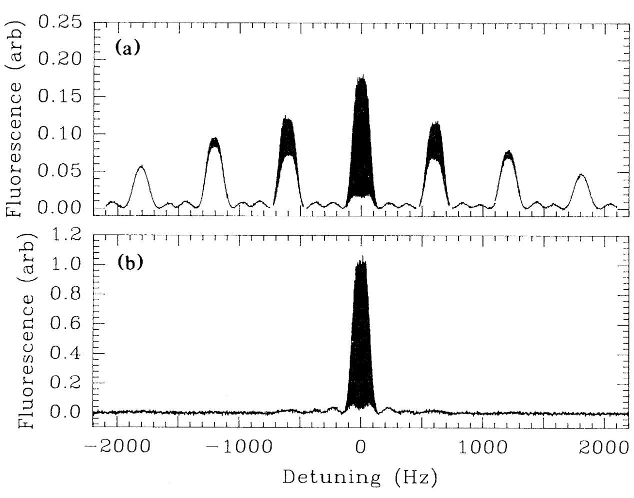

FIG. 2. (a) Clock spectrum of the 7 $\Delta m_F=0$ transitions from the $F=3$, m_F ground-state hyperfine level to the $F=4$, m_F ground state and (b) the spectrum for 95% of the atoms initially prepared in the $F=3$, $m_F=0$ state. The degeneracy of the m_F levels is removed by an 860 μG magnetic field.

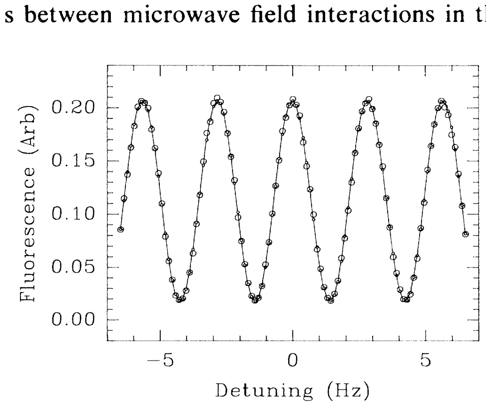

FIG. 3. The central Ramsey fringes of the $F=3,m_F=0 \rightarrow F=4,m_F=0$ transition. The FWHM linewidth is 1.41 Hz for a fountain height of 15 cm. The large circles are the data (1 launch per point, 0.933 s per launch) and the small circles and line represent the fit to the data.

ty. The center frequency is determined with a precision of 1.5 mHz after 100 launches. For our present noise level of 2%, taking data only at the half height point on either side of the central Ramsey fringe determines the center frequency to ± 11 mHz/$\tau^{1/2}$ or $\delta\nu/\nu = \pm 1.2 \times 10^{-12}/\tau^{1/2}$ where τ is the averaging time in s.

To measure any frequency shift due to the ultracold collisions, we vary the laser-cooled Cs density and record the apparent transition frequency. The frequency of the central Ramsey fringe versus density is shown in Fig. 4. The frequency shift appears linear in cold atom density and has a value of -12.9 ± 0.7 mHz or -1.4×10^{-12} at a density of $(2.7 \pm 1.5) \times 10^9$ cm^{-3}. Data at each density are taken by changing the density of each successive launch in order to eliminate errors due to any frequency drift of the H maser. The error bars indicate the statistical uncertainty of the center frequency and demonstrate that the H-maser frequency and the frequency shift due to the cold atom density were constant to ± 0.37 mHz.

The cold atom density in the fountain is determined by measuring the optical thickness of the ballistically traveling sample of atoms as a function of time. The thickness is measured using 1 ms pulses of a 6 nW vertical laser beam which is centered on the openings of the microwave cavity. During a launch, the 1 mm diam laser beam is pulsed 10 times and the optical thickness is measured for seven laser detunings spanning 13 MHz. The time dependence of the thickness is consistent with an initially uncorrelated Gaussian velocity and spatial distributions which we assume in order to calculate the density. With this assumption, the $1/e$ column height of the sample increases as $(z_0^2 + 4u^2t^2)^{1/2}$, where $u^2 = 2kT/M$ and $z_0 = 0.2$ cm is the initial column height measured with a charge-coupled-device camera. The final sample column height of 1.9 cm is measured by detecting the fluorescence as a function of time when the atoms return to the detection region 0.522 s after launching and yields $u = 1.9$ cm/s. Using the resonant photon absorption cross section of 1.4×10^{-9} cm^2, we calculate a density versus time and the time-averaged density between the microwave pulses.

Calculating a frequency shift cross section from the data is not necessarily straightforward since the collision energy of the detected atoms with other atoms in the sample decreases by an order of magnitude during the fountain time. However, s-wave frequency shifts are usually independent of temperature since the cross section scales as $T^{-1/2}$ and the relative velocity as $T^{1/2}$ [6]. Therefore, the average fountain density is relevant if s waves dominate [11].

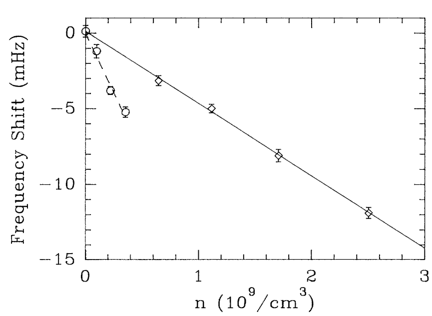

FIG. 4. Frequency shift vs ultracold Cs density for atoms evenly distributed among the $F=3$, m_F states (solid line) and for atoms in only the $m_F=0$ states (dashed line). For a density of $(1.0 \pm 0.6) \times 10^9$ cm^{-3}, the shifts are -4.8 ± 0.2 and -15.8 ± 1.4 mHz.

To preserve the short term stability, as many $m_F=0$ atoms as possible should be used and atoms in $m_F \neq 0$ states should be removed (or optically pumped into the $m_F=0$ state) since they do not contribute to the signal but do cause a frequency shift. We have removed these atoms by launching the atoms in the $F=4$ state as in the above, using an additional microwave source to transfer atoms from the $F=4$, $m_F=0$ state to the $F=3$, $m_F=0$ state, and then again pushing the $F=4$ atoms out of the way. The full clock spectrum for this case is shown in Fig. 2(b). Here, 95% of the atoms in the fountain are in the $m_F=0$ state and 2% are in the $m_F=\pm 1$ states. By varying the population transferred to the $F=3$, $m_F=0$ state, we have measured the frequency shift due to collisions between atoms in the $m_F=0$ states to be -5.5 ± 0.5 mHz at a density of $(3.5 \pm 2.0) \times 10^8$ cm^{-3}.

The measured density shift for atoms initially distributed among all of the $F=3$, m_F states in Fig. 4 is of the order of the shift estimated by Tiesinga *et al.* [6]. At 2.8 μK, they calculated a frequency shift of -2.8 mHz (-1.6 mHz for s waves and -1.2 mHz for p waves) at a density of 10^9 cm^{-3} which is about 40% smaller but within the density uncertainty of the -4.8 mHz measured shift. For the $m_F=0$ states, p waves do not contribute due to Bose symmetry and Tiesinga *et al.* estimated the shift to be -2.2 mHz whereas we measure a shift of -15.8 mHz for a density of 10^9 cm^{-3}. Since the Cs-Cs potentials used in the calculations have large uncertainties, this discrepancy is not unreasonable. In fact, future measurements of the frequency shifts for each F, m_F state might be used to improve the accuracy of the potentials. With improved accuracy, the location of elastic collision resonances, useful for evaporative cooling of Cs [4], may be accurately predicted [12]. The present and future measurements also determine the frequency shifts for measurements with laser-cooled interferometers [13].

There are a number of potential sources for systematic errors in Cs frequency standards. Differences in our magnetic bias field versus the height of the fountain cause a $\pm 2 \times 10^{-15}$ uncertainty due to the quadratic Zeeman shift ($+316$ μHz for $B_c = 860$ μG). A first-order Doppler shift arises due to a small traveling wave component in the microwave cavity caused by the Ohmic losses in the Cu cavity walls. Using a measured value by Bauch, Heindorff, and Schroeder [14], we estimate a worst case frequency shift of $\pm 2 \times 10^{-14}$. The adjacent $m_F = \pm 1$ states may also cause an apparent shift of the $m_F=0 \rightarrow m_F=0$ transition frequency (Rabi and Ramsey line pulling) [15]. The line pulling increases with increasing microwave power and by driving $3\pi/2$ vs $\pi/2$ excitations in each cavity passage, the line pulling is determined to be less than 1×10^{-14}. Because scattered laser light produces ac Stark shifts of the ground states, all laser beams are blocked by mechanical shutters while the atoms are in the fountain and no shift is present at the 4×10^{-14} level. The power absorbed by the atoms is not a negligible fraction of the microwave power in the cavity and the apparent microwave transition frequency is pulled proportionally to any cavity mistuning and the cold atom density [16]. With a 2 MHz upper limit on the mistuning, the cavity pulling is estimated to be less than $\pm2\times10^{-16}$ and no shifts were observed for several cavity mistunings.

For future frequency standards where nearly all of the atoms are prepared in an $m_F=0$ state, it should be possible to reduce the population asymmetry in the $m_F=\pm1$ states to less than 0.1% of the $m_F=0$ population in order to eliminate any line pulling effects. This will allow a 3 times smaller magnetic bias field which can be reduced a factor of 5 more by using a cylindrical cavity which has a longer interaction time (and a smaller first-order Doppler shift) [17]. With a 60 μG bias field, the fractional frequency shift of the $M_F=0$ transition is $+1.7\times10^{-16}$ and the sensitivities to magnetic field inhomogeneities and changes in the external field will be reduced by a factor of 15.

Our short term stability is already a factor of 2 below that of many primary Cs standards [18] and can be improved by at least a factor of 10. Presently, atoms completing the clock transition are detected in the lower vacuum chamber where there are also background Cs atoms at room temperature. These background atoms comprise $\approx20\%$ of the maximum detected fluorescence from atoms in the $F=4$, $m_F=0$ state and can be eliminated by inserting an additional detection chamber between the trap and the microwave cavity. Improved solid-angle collection and a more efficient detector will increase the signal size by more than a factor of 100.

Further reduction of the ultracold frequency shift will be made by eliminating atoms in the $m_F=0$ state which have *large* transverse velocities ($T_{\rm eff}>400$ nK) that prevent them from returning through the 6 mm$\times$5 mm aperture of the cavity. This is easily realizable by driving velocity-selective stimulated Raman transitions [19] with phase-locked diode lasers [20]. With our present geometry, the shift can be reduced by a factor of 10 to -5×10^{-14} while maintaining the above gain in the short term stability.

Finally, we emphasize that the density-dependent frequency shift can be extrapolated to $n=0$ even if only density ratios are well known. With our present data, the $n\rightarrow0$ frequency is determined to $\delta\nu/\nu=\pm4\times10^{-14}$ even though the absolute density is only certain within a factor of 2. By continuously performing a density extrapolation, an accuracy approaching 10^{-16} could be obtained while maintaining a short term stability of $3\times10^{-14}/\tau^{1/2}$.

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