rf Spectroscopy in an Atomic Fountain

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Laser-cooled sodium atoms pushed up on a vertical trajectory by radiation pressure are observed to turn around due to gravity. The relatively long time the atoms spent freely falling in this "atomic fountain" allowed the ground-state hyperfine splitting to be measured with a linewidth of 2 Hz. After a 1000-sec integration time, the center of the line was resolved to ± 10 mHz. The absolute splitting was measured to be 1771626129(2) Hz.

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Precise or sensitive measurements using atomic beams are often limited by the measurement time, and in situations where Ramsey's method of separated oscillatory fields is used, the measurement time is the transit time of the atoms through the apparatus.¹ In the early 1950's, Zacharias attempted to make a fountain of atoms by directing a thermal atomic beam upwards.² Slow atoms in the tail of the Boltzman distribution were expected to follow a ballistic trajectory, thus permitting a long measurement time. The experiment failed but nevertheless stimulated several important developments in atomic physics that led to the hydrogen maser and precise resonance experiments with bottled neutrons.³

Despite the failure, the lure of an atomic fountain has survived, and a detailed analysis of the Zacharias fountain for an improved time standard has been given recently.⁴ With the advent of laser cooling and trapping techniques,⁵ the feasibility of atomic fountains was reexamined. Analysis of fountains for precise two-photon spectroscopy⁶ was made, and the possibility of using atomic fountains for sensitive searches for electron or proton electric dipole moments and for "quantum reflection" studies was also suggested.⁷ Recent rf spectroscopy of atoms cooled in optical molasses^{8,9} and then dropped yielded a 44-Hz linewidth.¹⁰ We report here the construction of an atomic fountain, and show that narrow resonances can be observed. The 2-Hz linewidth of the resonance we excite is determined by the $\frac{1}{4}$ -sec measurement time. By comparison, the present U.S. time standard, NBS VI, has a 26-Hz linewidth.¹¹

The basic idea of our fountain was to first store and cool atoms in a optomagnetic trap, ¹² and then push them up with a pulse of laser light. Near the top of their trajectory, the atoms entered a waveguide where the microwave transition occurred. The returning atoms then fell into a detection region where they were resonantly ionized. A schematic of the experimental setup is shown in Fig. 1. The waveguide, trap, and detector were mounted inside a vacuum can where the pressure was typically 5×10^{-10} Torr. The cw dye laser used to excite the cooling transition was tuned ~20 MHz below the

Na $(3S_{1/2}, F=2) \rightarrow (3P_{3/2}, F=3)$ transition and an rf sideband provided by an electro-optic modulator was similarly tuned ~20 MHz below the $F=1 \rightarrow F=2$ transition to avoid optical pumping during the cooling process. The sideband had 10% of the carrier intensity.

The magnetic trap was formed by two 10-cm-diam coils, separated by 6 cm, with opposing currents and three orthogonal, 9-mm-diam standing laser fields. The magnetic field gradient was 1 G/mm. The trap was loaded from a thermal atomic Na beam that was slowed by a counterpropagating frequency-chirped laser beam.¹⁴ An equilibrium density corresponding to 5×10^7 trapped atoms was reached after ≈ 500 msec.¹⁵ The slowing laser beam was then turned off with an acousto-optic modulator. Simultaneously, the trapping magnet was



FIG. 1. Perspective view of the experimental setup for the atomic fountain. The atoms, initially confined to a small volume in the trap region, follow a ballistic trajectory through the waveguide and back to the detection region. A curved metal shield electrostatically focuses the photoionized atoms onto the detector. The waveguide is impedance matched to 50 Ω in both ends (Ref. 13). The distances from the trap to the detector is 14 and 31 cm to the top of the waveguide. The figure is roughly to scale.

turned off and the intensity in the molasses beam was reduced from 16 to 1.8 mW/cm² per beam. In the following 15 msec, the atoms were cooled from ≈ 1 mK to $\approx 50 \ \mu$ K, a temperature characteristic of low-intensity molasses.^{5,9,16} During the final cooling, the atoms expanded from an approximately 1-mm- to a 2-mm-diam sphere.

As soon as the molasses was turned off a short pulse from a vertical beam (730 mW/cm²) tuned in resonance with the $F=2 \rightarrow F=2$ transition launched the atoms up on their ballistic trajectories. In preparation for the interaction with the microwaves, the atoms were optically pumped to the F=1 ground state by turning off the laser rf sideband 3.6 µsec after the pushing pulse was turned on. This pulse of light gave the atoms an average vertical velocity of ≈ 240 cm/s. In the process, the ~ 80 scattered photons lead to an increase in the transverse velocity from 14 to ~ 22 cm/sec.

Atoms with a vertical velocity of 240 cm/sec and less than \sim 4 cm/sec transverse velocity turned around near the top of the waveguide and fell back into the detection region ~ 450 msec after the push. The atoms were detected by resonant ionization at a distance 14 cm above the center of the trap. A 1- μ sec dye-laser pulse (275 mW/cm²) tuned to the $F=2 \rightarrow F=3$ transition first excited the atoms from $3S_{1/2}$ to $3P_{3/2}$, and a 10nsec, 25-mJ pulse from a frequency-tripled Nd-doped yttrium-aluminum-garnet (Nd:YAlG) laser at 355 nm ionized the excited atoms ≈ 250 nsec after the cw laser was turned on. Ions created in a ~ 2 -cm³ region where the two laser beams crossed were focused onto a microchannel-plate detector. The detection efficiency was linear with count rate to within 10% for count rates below 200 ions per pulse. 4 msec after the photoionization a new cycle began by loading the trap.

The distribution of atoms detected in the fountain was recorded as function of flight time (time between pushing and detection pulses). The signal shown in Fig. 2(a) was deliberately made small to avoid saturating the detector and the detection cw laser beam had the rf sideband left on. Evidence for the atoms following ballistic trajectories was obtained by using a pulse from the cw laser to deflect the atoms out of the fountain on their first passage through the detection region. The descending atoms showed a corresponding notch in the time-offlight distribution. Figure 2(b) shows an example where atoms are deflected out around 105 msec after the pulse. In agreement with what is expected for atoms accelerated by gravity, the time-of-flight signal drops to zero around 270 msec.

Atoms near the top of their trajectory enter an rf waveguide as shown in Fig. 1. A traveling-wave TE_{10} mode¹³ was excited in a 46-cm-long, 11.4-cm-square aluminum tube closed on both ends. Atoms enter the waveguide through a 2.5×10-cm² hole in the bottom, and feel an oscillating magnetic field with approximately uniform magnitude and fixed polarization. A Hewlett-



FIG. 2. (a) Number of detected ions as a function of time between pushing and detection. Atoms detected after 169 msec must have turned around above the detection region. (b) Atoms passing the detection region between 102 and 109 msec are blasted out of the fountain. The corresponding notch for falling atoms is observed. The signal drops to zero (curve offset) at \sim 450 msec because atoms that would have arrived later hit the top of the waveguide.

Packard 8662A synthesizer slaved to a temperaturestabilized FTS 1050A quartz oscillator provided an rf frequency stable to 5 parts in 10^{12} for a 1000-sec interval. The absolute calibration of the crystal oscillator was provided by timing signals received from the Naval Observatory via a Loran C frequency monitor.

The only transition in the $F=1 \rightarrow F=2$ manifold that is insensitive to a dc magnetic field to first order is the $m_F=0 \rightarrow m_F=0$ transition. In order to define the quantization axis and to Zeeman shift the other transitions out of resonance, a bias magnetic field of 15-20 mG was applied along the polarization axis of the rf field.

We excited the transition using Ramsey's method of separated oscillatory fields.¹ A first " $\pi/2$ " pulse is used to create an equal superposition of the ground and excited states of the atom. The rf power is then turned off in 10 nsec with an isolation of better than 50 dB. Some time Δt later, a second $\pi/2$ pulse is applied, and if the phase of the freely precessing atom is equal to the phase of the microwave signal modulo 2π , the second pulse will put the atom completely in the excited state. We chose a measurement time $\Delta t = 255$ msec (limited by the flight time through the waveguide) by detecting 446 msec after the push. Operating with $t_p = 3.2$ -msec $\pi/2$ pulses (B_{rf} $\approx 61.4 \,\mu\text{G}$) the observed central fringe [Fig. 3(b)] has a FWHM of 2.0 Hz. In the limit of small detunings $\Delta v \ll v_{\text{Rabi}} = 43$ Hz, the theoretical line shape is $\infty 1 + \cos(\Delta v \Delta t)$ and a fit to the data yields an interaction time of 253 ± 2 msec. A full Ramsey line shape is shown in Fig. 3(a) for a 32-msec $\pi/2$ pulse and 125 msec interaction time. The fit for the theoretical line shape gives estimates for the Rabi frequency and $\pi/2$ pulse width that are better than 10% of the experimental values.

At 446 msec after the pushing pulse, 60-100 atoms



FIG. 3. (a) The observed Ramsey line shape for two 32-msec $\pi/2$ pulses separated by 125 msec. (b) The central fringe obtained with two 3.2-msec $\pi/2$ pulses separated by 255 msec.

per toss are detected with a signal-to-noise ratio of $\sim 4:1$. Sources of noise included (i) stray thermal Na atoms in the photoionization region at the time of detection (we estimate 10-30 detected atoms at a sodium partial pressure of 2×10^{-11} Torr), (ii) Nd:YAlG-laser-intensity fluctuations ($\sim 30\%$), and (iii) fluctuations in the number of trapped atoms ($\sim 10\%$). Each data point in Fig. 3(b) represents an integration over 20 tosses at 1 toss/sec. After an integrating time of 1000 sec, the line center had a fitted uncertainty ± 10 mHz.

Because of systematic shifts in the resonance frequency, we did not determine the absolute hyperfine interval to this accuracy. The most significant uncertainty is our lack of knowledge of the bias field. For a field of 15 mG, the quadratic Zeeman shift is $\alpha B^2 \simeq 0.5$ Hz, where α is calculated to be $\simeq 2.21 \text{ mHz/mG}^2$. By measuring the shift of the resonance as a function of bias field strength, we found the quadratic Zeeman shift to be 40% higher than predicted, suggesting a 20% calibration error in our magnetic field measurements. Another serious systematic effect is the first-order Doppler shift, which is exacerbated in our traveling-wave geometry. A given atomic trajectory may carry the atom longitudinally in the waveguide, and hence to regions of different phase with respect to the position of the upward-traveling atoms. Atoms traveling more than ~ 3 cm laterally between pulses will miss the detection region, so the worse-case phase shift is $\Delta \phi = 20^{\circ}$, corresponding to the frequency shift of ~ 0.25 Hz. A phase offset introduces an asymmetry in the full Ramsey line shape which we observed by shifting the photoionization region to the side of the detection region. However, when the ionization volume was centered we observed a symmetric waveform, indicating an average shift of less than 0.1 Hz. Correcting for the Zeeman shift, and accounting for the above uncertainties, our value for the transition frequency is 1771626129 ± 2 Hz.

It is an instructive exercise to examine the major systematic effects that limit the accuracy of the present Cs time standard^{11,17} and compare them to what can be ob-

tained with a Cs fountain designed specifically for an atomic clock. The long-term accuracy of the current Cs-beam frequency standard is limited primarily by cavity phase shift $(\Delta v/v = 0.80 \times 10^{-13})$, the second-order Doppler shift (0.10×10^{-13}) , pulling from neighboring transitions (0.20×10^{-13}) , and magnetic field inhomogeneities and offsets $(0.03 \times 10^{-13} \text{ and } 0.02 \times 10^{-13})$. The uncertainty due to the second-order Doppler shift for a fountain clock will be on the order of $\Delta v/v \approx 10^{-18}$. The low rf power requirements necessary to flip the spin of the atoms greatly decreases the frequency pulling from neighboring transitions. Thus, the clock could be run at much lower bias fields. For $B_0 \simeq 1$ mG the quadratic Zeeman shift is $(414 \text{ Hz/G}^2)B_0^2 \simeq 0.4 \text{ mHz}$, leading to a fractional uncertainty of 10^{-15} for a 1% field inhomogeneity. At this bias level, off-resonant transitions lead to an uncertainty of $\Delta v/v \simeq 10^{-16}$.

Cavity-phase-shift effects are the most serious systematics for a conventional atomic clock. The small traveling-wave component of the electromagnetic field in a microwave cavity with a finite Q will cause the firstorder Doppler shift that we previously discussed, but this effect can be greatly reduced with the use of high-Q cavities.¹⁷ The other phase-shift effect that plagues conventional clocks is that two different microwave regions are used and the two cavities may have a phase shift with respect to each other. A traditional method of dealing with this effect is to periodically reverse the direction of the atomic beam. A fountain clock that uses only one cavity will be free of this systematic effect.

The short-term frequency stability obtained with the fountain is 300 mHz/ $\sqrt{\tau}$ (τ is the integration time measured in seconds) as compared with 18 mHz/ $\sqrt{\tau}$ for the NBS VI and 45 mHz/ $\sqrt{\tau}$ for the Hewlett-Packard 5061B commercial standard with low-noise option. However, our fountain clock holds the promise for improved stability. We estimate that a compact cesium clock using atoms cooled to 20 μ K¹⁸ (corresponding to an rms velocity in one dimension of 3.7 cm/sec) will have a counting rate 3-4 orders of magnitude higher than our "first try" experiment. With this increase in signal-to-noise ratio and smaller systematic shifts, an atomic-fountain microwave clock will compete favorably with clocks based on conventional beams.

A single ion stored in an rf trap is in an almost ideal environment for an atomic clock, but measurements on single ions are hampered by signal-to-noise problems. Larger signals may be obtained by storing more ions in the trap, but at the cost of larger systematic shifts.¹⁹ The linewidth of atoms in a fountain can never compete with that of stored ions, but for atomic clocks locked to optical transitions,²⁰ a \sim 1-Hz linewidth is not a major liability, and the signal-to-noise ratio of an atomicfountain clock can be better than an ion-trap clock. Furthermore, neutral atoms have narrow lines at more convenient optical frequencies.

Finally, we note that atomic fountains have other uses.

For example, we are currently trying to produce atoms with a velocity spread on the order of hundreds of nanokelvins to be used in a surface scattering experiment.⁷ High-density atomic fountains will also be used in a variety of extremely sensitive atomic physics measurements.

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