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Ramsey Resonance in a Zacharias Fountain.

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Abstract. – We report a realization of Zacharias's 1953 proposal for observing a Ramsey resonance in an atomic fountain. Launched upward from a moving optical molasses where they have been cooled to $\sim 5\,\mu\text{K}$, cesium atoms pass once through a microwave cavity, continue to the summit of their trajectory, then fall again through the same cavity, completing the separated-fields interaction. The atoms spend 0.25 s in free flight above the cavity. Linewidth (2 Hz) and S/N imply a stability of $3 \cdot 10^{-12}\,\tau^{-1/2}$, at least as good as in existing Cs clocks, with eventual expected improvements of 10^2.

Shortly after Ramsey [1] introduced the method of separated oscillatory fields to produce narrow resonances, Zacharias [2] proposed an atomic fountain to achieve the separate interactions with a single microwave cavity; the atomic beam passes once through the cavity on the way up and once again on the way down. A long time between the two passages makes the Ramsey resonance very narrow, and thus suitable for high-performance atomic clocks or high-precision measurements. The lack of very slow atoms in thermal beams prevented Zacharias from operating an atomic fountain. Cs beams from a thermal oven typically move at a few hundred m/s, but a 30 cm high fountain, which gives a 1 Hz resonance width, requires a launch velocity of only 2.5 m/s and a much smaller transverse velocity to prevent significant spreading of the beam as it falls back.

Laser cooling of atoms [3] renewed interest in fountains [4-6] and the first atomic fountain was recently achieved [7]. In that pioneering experiment, Na atoms were captured in a magneto-optical trap [8], further cooled in optical molasses [9], and were launched with the radiation pressure of an upward-directed laser beam. They entered a microwave waveguide, reached their summit inside the waveguide and fell out into a detection region. 2 Hz wide Ramsey fringes were observed by twice pulsing the microwaves applied to the waveguide. A related experiment on free-falling Cs atoms was reported in [10]. Here we report a quite different sort of fountain Ramsey resonance, following Zacharias's prescription, which offers significant advantages and will likely lead to atomic clocks with unprecedented stability and accuracy.

The first major improvement in our method is launching the atoms. The radiation pressure launch of ref. [7] heated the atoms, largely negating the advantage of first cooling them with polarization gradient optical molasses [11-17]. In the present experiment, we use a launching method that introduces no extra heating and maintains the low molasses temperature, which is in the μK range for Cs. A low temperature is essential to minimize transverse spreading of the atoms during their trajectory.

This launching is done from a «moving» molasses [6, 12] whose use was first reported for an atomic funnel [18] and which is used here for producing a fountain (fig. 1) [19]. An upward-moving molasses is accomplished by shifting the frequency of the vertical beams while keeping the frequency ν_L of the horizontal beams unchanged. If the upward-traveling wave frequency is increased to $\nu_L + \Delta\nu_L$ and the downward one is decreased to $\nu_L - \Delta\nu_L$, this creates a moving standing wave at $v_z = \lambda\,\Delta\nu_L$. Because of the Doppler shift, atoms moving up at v_z see all beams at the same frequency ν_L: this realizes a static molasses in the frame moving at v_z. Now, if $\Delta\nu_L$ is slowly swept in time between 0 and a maximum value $\Delta\nu_L^f$, the atoms, initially at thermal equilibrium in the laboratory frame at a given temperature, will follow adiabatically the change of frequency and stay nearly at the same temperature in the moving frame. The allowed dv_z/dt is larger at high laser intensity and small detuning, *i.e.* at high molasses temperature. It is thus crucial to subsequently apply an additional cooling phase.

Temperature measurement confirms that the launch works as predicted. With the molasses laser at a detuning $\delta = -2\Gamma = -10$ MHz below the $6S_{1/2}$, $F=4 \rightarrow 6P_{3/2}$, $F'=5$ transition and an intensity $I = 5\ \mathrm{mW/cm^2}$, $\Delta\nu_L$ can be swept between 0 and 1.6 MHz in 1 ms,

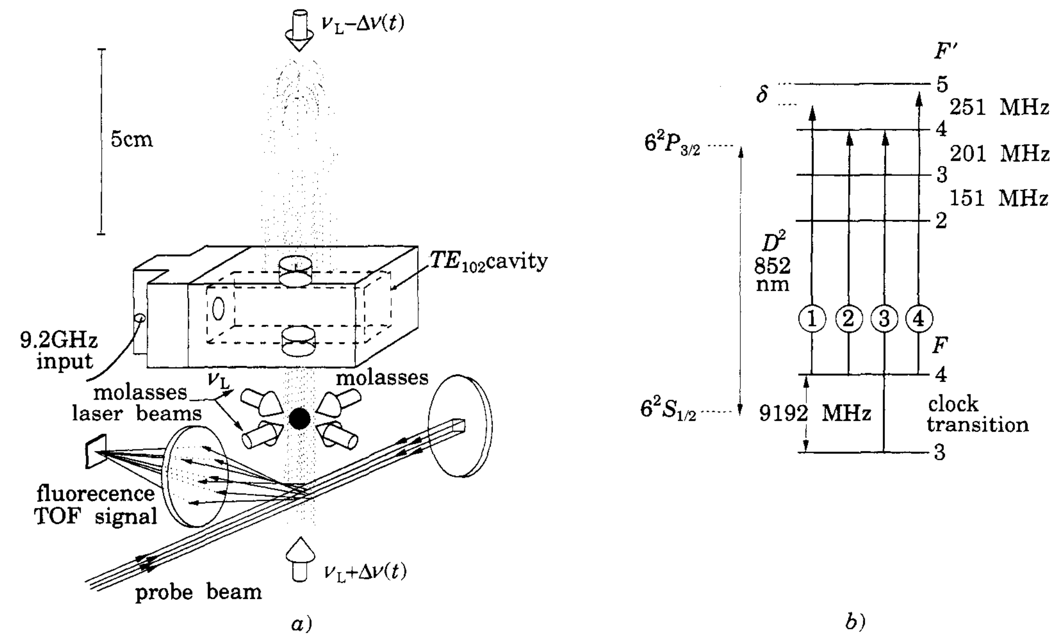

Fig. 1. – *a*) The Zacharias-style fountain. Cs atoms at 5.5 μK are launched from a «moving» molasses through a microwave cavity. The change of hyperfine state, due to the Ramsey resonance after passing twice through the cavity, is detected by state-selective resonance fluorescence in the probe beam. *b*) Relevant Cs transitions. Molasses loading and launching: 1 + 3. Hyperfine pumping: 2. Detection: 4.

accelerating nearly all the atoms to 1.4 m/s. Then a 1.6 ms cooling time with fixed $\Delta\nu_L^f$ (1.6 MHz), I divided by four and δ increased to -70 MHz, reduces the temperature from $\sim 60\,\mu$K to $(5.5 \pm 1)\,\mu$K. This temperature is identical to that of a static molasses with the same intensity and detuning [14]. All of the temperatures (before launch, just after launch and following the cooling period) are measured by time of flight (TOF) as in our previous work [14]. For measuring the temperature of launched atoms, we simply include their initial velocity in the calculation of the TOF spectrum to which the data are fit. After a 0.3 s fountain time, about 20% of the atoms of the molasses are still detected in a 0.7 cm^2 horizontal area, a value consistent with the thermal spreading at 5.5 μK.

A second major difference in our experiment compared to the first fountain is that we use a continuously fed microwave cavity rather than a pulsed traveling wave and that the atoms pass completely out of the cavity and then fall back through it. This is important for several reasons. First, in an atomic clock there are certain errors (cavity phase shift errors) connected to the motion of the atoms with respect to the residual running microwave in the cavity. In conventional clocks, these errors are evaluated by reversing the direction of the atomic beam. In a fountain, that reversal occurs automatically but only if the atoms pass completely through the cavity. This phase shift error is the major source of inaccuracy in present Cs clocks. Beyond this, the fact that the atoms pass completely out of the cavity gives free access to a region where one can induce small phase shifts and perform, for instance, QND measurements of small (~ 1) photon numbers, atomic-electric-dipole moment measurements, or study long-range atom-atom collisions and atom-surface interactions.

Many details of the apparatus have been previously described [14]. Cesium atoms are caught in optical molasses (fig. 1) with $\delta = -10$ MHz and $I = 5$ mW/cm^2. Horizontal beams are ~ 10 mm in diameter with an intensity uniformity of $\pm 15\%$, whereas vertical beams are limited to a 8.5 mm diameter by the holes in the microwave cavity. The vertical and one of the horizontal beam pairs consist of linearly and orthogonally polarized counterpropagating waves. The other horizontal standing wave has a linear vertical polarization to allow Zeeman or hyperfine optical pumping (see below). A repumping diode laser beam tuned to the $F = 3 \rightarrow F' = 4$ transition is combined with the two horizontal molasses beams. After loading the molasses for 0.5 s, about $7 \cdot 10^6$ atoms are confined and cooled in the ~ 0.5 cm^3 molasses volume. A TE_{102} microwave cavity made of a standard X band waveguide shorted at both ends is located 2.7 cm above the molasses. The two holes have been drilled in the narrow side of the waveguide half a guide wavelength away from the shorts. The microwave magnetic field is vertical. The cavity is overcoupled so that $Q = 1000$. A weak ($B = 3 \cdot 10^{-6}$ T) vertical bias magnetic field is applied to resolve the hyperfine transitions and record only the $6S_{1/2}$, $F = 3$, $m_F = 0 \rightarrow F = 4$, $m_F = 0$ clock transition sensitive to B only at second order. The maximum field inhomogeneity over the whole fountain is made smaller than $\pm 10^{-6}$ T with field compensating coils.

Once the atoms have been launched and cooled to the desired temperature, a 2 ms long laser pulse, tuned to the $F = 4 \rightarrow F' = 4$ transition and having both σ and π polarization with respect to the guiding magnetic field, pumps all the atoms into the $F = 3$ level, with negligible heating. After their two transits through the cavity, the atoms fall through a (10×10) mm^2 cross-section probe beam centered 25 mm below the molasses center. The probe, tuned to $F = 4 \rightarrow F' = 5$, efficiently detects those atoms making the clock transition. Their fluorescence ($\sim 10^3$ detected photons/atom) appears as a TOF signal against a dark background. In order not to be limited by shot-to-shot fluctuations in the number of atoms in the molasses ($\pm 30\%$), we, as in [10], normalize the number of detected $F = 4$ atoms to the total $F = 3 + F = 4$ atoms in each fountain cycle. Let $A_{F=4}$ be the area of the first half of the TOF signal. During the second half, a repumping beam, superimposed on the probe, is quickly ($< 100\,\mu$s) made resonant with the $F = 3 \rightarrow F' = 4$ transition so that the area

$A_{F=3+F=4}$ of the second half of the TOF signal is a measure of the total number of atoms. The normalized clock signal is $A_{F=4}/A_{F=3+F=4}$. With no Zeeman optical pumping, one expects that $\sim 1/2F+1 = 1/7$ of the atoms in the molasses contribute to the clock signal, in good agreement with the measured value ($(11 \pm 2)\%$).

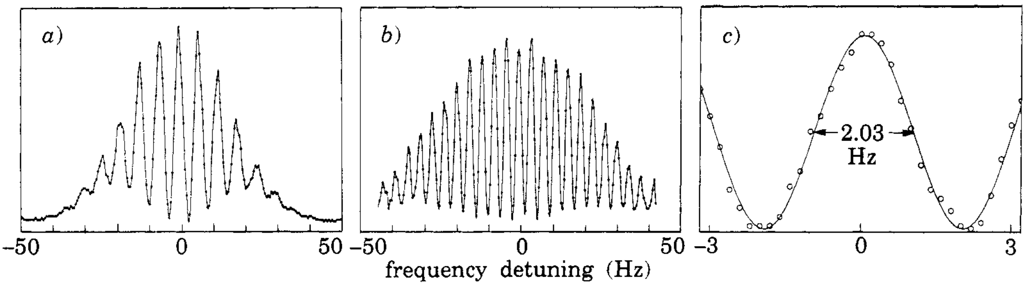

Fig. 2. - Ramsey fringes for two launch conditions. Each dot is the average of 5 fountain cycles. *a)* $v_z = 1.19$ m/s, $T = 26$ μK. Atoms reach 3.4 cm above the cavity for a 3.0 Hz wide fringe. Frequency step: 0.5 Hz. *b)* $v_z = 1.41$ m/s, $T = (5.5 \pm 1)$ μK. Fringe width 2.0 Hz. *c)* Central fringe of *b)* at 0.2 Hz frequency step. The curve is a least-square fit to a cosine lineshape. The standard deviation is 2.8%. The frequency standard deviation is 50 mHz and is dominated by the instability of the 9.2 GHz source.

In order to illustrate the flexibility of the fountain method, Ramsey fringes at different resolutions and molasses temperatures are presented in fig. 2. In fig. 2*a*), the atoms are relatively hot ($T = 26$ μK) and the center of the atom cloud reaches only 3.4 cm above the cavity at the top of its trajectory. The fringe width is 3.0 Hz and the Rabi pedestal is about 50 Hz wide. In fig. 2*b*), atoms are launched at the maximum height allowed by our apparatus at a temperature of 5.5 μK. The atoms reach 6.4 cm above the cavity, the Ramsey interrogation time T_f is 0.25 s, giving a fringe width of 2.0 Hz. The small velocity spread of the launched atoms ($\delta v_z/v_z = \pm 1.5\%$) enables a large number of side fringes to be present (the slight decrease of the central fringe comes from the microwave power being slightly above the optimum value). Figure 2*c*) presents the central fringe of fig. 2*b*), recorded with a 0.2 Hz frequency step. Each data point is the average of 5 (molasses loading + fountain) cycles (5.5 s). The continuous lines is a least-square fit to the cosine transition probability valid for monokinetic atoms. The r.m.s. vertical deviation of the points from the fit is 2.8% of the full fringe amplitude and is a measure of the actual S/N including both the fluctuations in the normalized fluorescence signal and the effect of the frequency noise of the 9.2 GHz source which is dominant here. The frequency (horizontal) standard deviation is indeed 50 mHz or a relative deviation of $5.5 \cdot 10^{-12}$ which is comparable to the measured Allan variance of our quartz crystal reference oscillator ($5 \cdot 10^{-12}$ between 1 and 100 s) from which the 9.2 GHz frequency is generated. In addition, when we increased the width of the fringes (short fountain times) or decreased the fringe contrast (by deteriorating the d.c. $\boldsymbol{B}$ field homogeneity) in order to reduce the influence of the quartz frequency noise, the S/N increased to 30 for one fountain cycle (1.1 s), an improvement of about a factor of 2. Thus it is clear that the present limitation to the S/N in fig. 2*c*) is the quartz frequency instability. Obviously a very good fly-wheel oscillator is needed for these high-resolution fountain experiments.

The stability of a pulsed clock can be evaluated by the square root of the Allan variance [20]:

$$\sigma(\tau) = (\delta\nu/\pi\nu) \cdot N/S \cdot (T_{\text{cycle}}/\tau)^{+1/2}, \tag{1}$$

where $\delta\nu$ is the fringe width at half maximum, T_{cycle} the time for a complete molasses loading and fountain cycle ($T_{cycle} = T_{load} + T_f$) and τ the integration time in s. With S/N = 30 for $T_{cycle} = \tau = 1.1$ s, one gets a short-term stability of $\sigma(\tau) = 3 \cdot 10^{-12} \tau^{-12}$ which is, to our knowledge, comparable to or better than any presently operating Cs standard. This stability is not limited by the atom shot noise (about $2 \cdot 10^5$ atoms per toss contribute to the Ramsey fringe signal), but rather by technical fluctuations in the normalization procedure. When these fluctuations are sufficiently reduced (by about a factor of 10), atoms shot noise-limited detection should be achieved and the short-term stability of an oscillator locked to our Cs fountain would be $3 \cdot 10^{-13} \tau^{-1/2}$ or 10^{-15} per day. This would outperform by more than a factor 10 all presently operating Cs clocks. Furthermore the number of atoms involved in the clock signal is susceptible of considerable improvements by producing a more intense atomic beam using atoms funnels [18, 21] and/or using a magneto-optical trap to collect the slow atoms [7, 8, 10]. Zeeman optical pumping can also improve the clock signal and with pumping we did observe a threefold increase in fringe signal. However, this pumping requires a few tens of fluorescence cycles, atoms heat up and no large improvement is expected for high fountains.

In considering the ultimate stability of a fountain clock, it is interesting to discuss further expression (1) in the limit where $T_f \gg T_{load}$ and for atom shot noise-limited detection. If we detect all the atoms after the fountain (for instance by using an open Fabry-Perot cavity and a large-area probe beam), $\sigma(\tau)$ decreases as $T_f^{-1/2}$. However, the Doppler effect will tend to wash out the fringes after velocity integration if the atoms move horizontally more than $\sim \lambda_{\mu w}$. One then needs sub-Doppler techniques, for instance two-photon Ramsey fringes in the optical domain [5, 6].

More serious in the microwave domain is the role of the residual Doppler effect on the clock accuracy: the unavoidable traveling-wave component of the microwave field in the cavity induces a Doppler shift of the fringes (distributed cavity phase shift). Increasing the Q factor of the cavity would be a solution but one then approaches the maser threshold and cavity pulling effects become severe [22]. Alternatively, one may reduce the interaction volume in the microwave cavity by having small holes ($r \leqslant 5$ mm) defining a region where the phase is as constant as possible. In this case, the transverse temperature determines the number of detected atoms and $\sigma(\tau)$ varies as $(r^2 T T_f/\tau)^{1/2}$ if we neglect the initial molasses size and assume $v_{r.m.s.} T_f \gg r$. As an example, for a 1 Hz wide fringe, $r = 3$ mm, and with presently achievable Cs atom number (10^7 in $F = 3$, $m_F = 0$) and temperature (5 μK), we find $\sigma(\tau) = 2 \cdot 10^{-14} \tau^{-1/2}$ or 10^{-16} per day of integration. This stability would challenge that of the best H masers.

Not only is the stability potentially very high, but also the potential accuracy of such a Cs fountain is very attractive. Most of the limiting factors of a conventional clock are considerably reduced [4]: the second-order Doppler shift and the gravitational red shift are small ($\approx 10^{-17}$) and calculable. The residual first-order Doppler effect is proportional to the fringe width. Having a phase shift as small as that in [23] would give an effect of about 10^{-16}. For $Q_{cav} = 10^4$, 10^7 atoms and a 1 Hz fringe width, the power dissipated by the cavity is only 10^4 larger than the power absorbed by the atoms in the cavity. As a consequence, the cavity pulling effect is no longer proportional to $(Q_{cav}/Q_{at})^2$ as usual in Cs clocks, but tend to be proportional to Q_{cav}/Q_{at}. The relative cavity frequency detuning must then be controlled to better than 10^{-7} to reach a 10^{-17} accuracy level [22]. This cavity pulling effect could also be reduced by the techniques discussed by Wineland in [24]. Interestingly, maser oscillation could easily be achieved with a superconducting cavity. Finally our fountain allows a magnetic-field map with a 10^{-10} T field resolution and a $\sim 1\ \text{cm}^3$ volume resolution to be recorded by measuring the frequency of a field-dependent transition as a function of the height of the fountain. With a constant $\boldsymbol{B}$ field of 10^{-7} T, the resulting error would be less than 10^{-16}. In such a clock no laser light-shift is present. On the other hand, the black-body shift [25] must be considered ($-1.69 \cdot 10^{-14}$ at 300 K) and the temperature of the fountain region must be known to within ± 0.5 °C to allow correction at a 10^{-16} uncertainty level. Collisions could also become a problem at this level of precision. Changing the initial atomic density over a broad range is easy with such a fountain, allowing measurement of collisional shifts. Whatever the ultimate source of limiting systematic errors, the excellent linewidth and frequency stability of a cold atom fountain clock should allow unprecedently accurate evaluation of these errors. We anticipate $\sim 10^{-16}$ accuracy, about 10^2 better than any existing clock.

Finally, most of the atomic beam manipulation techniques presented here can be extended to optical frequency standars in the visible or UV domain [5, 6], with perhaps even more frequency stability and accuracy capabilities.

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