A ^{40}Ca OPTICAL FREQUENCY STANDARD AT 657 NM: FREQUENCY MEASUREMENTS AND FUTURE PROSPECTS

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Due to higher transition frequencies, optical frequency standards promise significantly improved stability over that of their microwave counterparts. Our laser-cooled ^{40}Ca optical frequency standard is based on the $^1S_0$ (m=0) $\rightarrow$ $^3P_1$ (m=0) intercombination line at 657 nm, and has demonstrated a fractional frequency instability of 4×10^{-15} at 1 s. Using a mode-locked femtosecond-laser-based frequency comb referenced to the primary Cs standard, we have made an absolute frequency measurement of this transition with an uncertainty of ±26 Hz (1σ). A discussion of the major systematic effects in our system is included. In an effort to reduce our largest systematic uncertainty, we have investigated second-stage cooling in one dimension on the narrow "clock" transition, through quenching of the excited state. We have transferred 45 % (22 % net efficiency) of the atoms into a narrow distribution with a temperature of 4 μK, representing a reduction in temperature by a factor of 500.

1 Introduction

Interest in optical frequency standards has increased dramatically over the past few years as the result of two major developments. First, the standards themselves have improved to the point where they are now beginning to be competitive with their microwave counterparts. Second, the development of mode-locked femtosecond-laser-based frequency combs has enabled direct connections between the optical and microwave domains, which greatly simplifies the daunting task of measuring optical frequencies.[1,2] The importance of optical frequency standards results simply from their high line Q's, which can potentially lead to extremely stable and accurate standards. The stability of a frequency standard can be expressed as:

$$\text{Fractional frequency instability} = \frac{\Delta\nu}{\nu_0 \times S/N}, \qquad (1)$$

where $\Delta\nu$ and ν_0 are the linewidth and frequency of the transition, respectively, and S/N is the spectroscopic signal-to-noise ratio. Here we see the advantages of working at high frequency as well as with a large number of atoms. In our work, we have focused on the development of a standard based on an optical transition in neutral Ca. The advantage of working with neutral atoms is that one can readily trap and cool large numbers, ideally without large associated frequency shifts. The downside of neutral atom-based standards is that the residual velocity of the absorbers can lead to significant systematic shifts. This paper will first focus on the present performance of our standard and then discuss some remedies for the limitations we face.

2 NIST Ca optical frequency standard

The transition at 657 nm in Ca has a narrow linewidth (400 Hz), convenient wavelength, and is insensitive to external perturbations (see Fig. 1). In order to achieve sub-kHz spectroscopic linewidths and to reduce systematic uncertainty due to atomic velocity, it is necessary to work with laser-cooled atoms. To this end we constructed a compact magneto-optic trap that we load from an atomic beam. With 40 mW of 423 nm light from a frequency-doubled diode-laser, we can load ~10^7 atoms into the trap in 10 ms, with a Doppler-limited atom temperature of 2 mK.[3] We probe the clock transition with a diode laser locked tightly via the Pound-Drever-Hall technique to an environmentally isolated optical cavity. To generate narrow features with high S/N, we use an optical version of a Ramsey sequence consisting of two counter-propagating pairs of co-propagating travelling-wave pulses, as first proposed by Bordé.[4] To improve our S/N further, we modified the technique of electron shelving, which was first used to detect weak transitions in trapped ions.[3] Our scheme uses near-resonant pulses (10 μs duration) on the 423 nm cycling transition to measure the ground-state population before and after red excitation. This approach normalizes the measured excitation relative to trap number and has improved our S/N by a factor of 20 over the technique of red fluorescence detection.

We begin the measurement cycle with 7 ms of trapping, after which we turn off the trapping beams and turn on the first blue probe pulse. Our imaging system collects 7 % of the induced fluorescence, which is sent to a photomultiplier tube. Next follows the Bordé-Ramsey excitation sequence of 4 red pulses (each generated

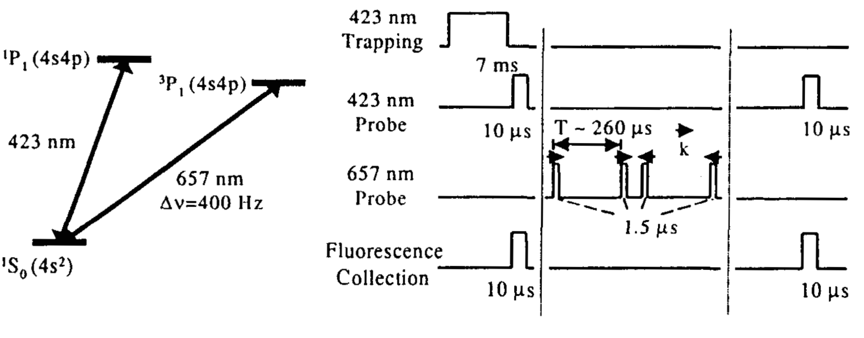


Figure 1. (a) Relevant Ca energy levels. (b) Timing diagram for spectroscopy on the "clock" transition.

by an rf pulse sent to an acousto-optic modulator (AOM)), which transfers ~20 % of the atoms to the long-lived excited state. Before the excited state decays significantly, we read out the remaining ground-state population with the second blue probe pulse. The ratio of the integrated fluorescence signals induced by the blue probe pulses provides our spectroscopic signal. To generate the Bordé-Ramsey fringes, we scan the 657 nm probe frequency while continuously repeating the measurement cycle. Figure 2 shows a fringe pattern taken at the natural linewidth of the transition. We usually operate the standard with a fringe resolution of 960 Hz since it represents a good compromise between linewidth and S/N (recall Eq. 1).

As a first evaluation of the performance of our standard, we tuned the laser to the side of a high-resolution fringe and measured the short-term stability of our system relative to our optical cavity (see Fig. 2).[5] The resulting upper limit on the short-term instability of the standard was more than an order of magnitude less than that of the best microwave standards. Good short-term stability is critical to a high performance standard in order to reduce averaging times needed for its evaluation. More recently a second test of the short-term stability was performed when we used a mode-locked laser to directly connect the Ca standard to an optical clock based on a single Hg^+ ion.[6] These measurements placed an upper limit of $< 7\times10^{-15}$ at 1 s for a version of the Ca standard with a slightly reduced S/N. While these results are encouraging, we note that this transition is capable of achieving significantly higher stability; currently our contrast is severely reduced (our fringe amplitude is 5 % of the maximum possible) primarily due to the high temperature of our cooled atoms.

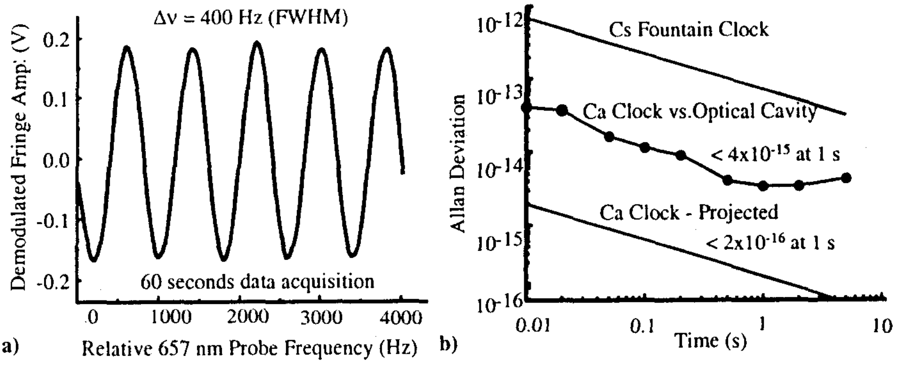
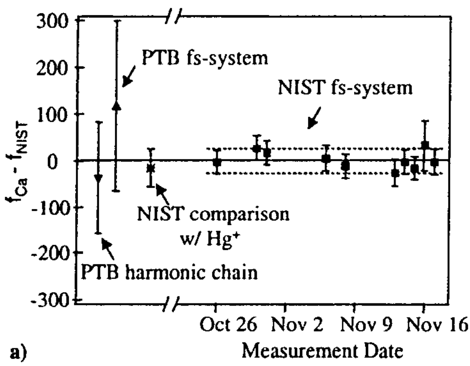


Figure 2. (a) Bordé-Ramsey fringes at the natural linewidth of the "clock" transition. (b) Ca short-term instability measured against an optical cavity.

3 Frequency measurements of the clock transition with a femtosecond-laser-based frequency comb

Using an optical frequency comb based on a mode-locked femtosecond laser, we have recently measured the frequency of the Ca clock transition relative to the Cs fountain standard.[7-8] To prepare our Ca system for an absolute frequency measurement, we use square wave frequency modulation of our 657 nm light at 70 Hz and lock the probe laser to the central Ca fringe. A daily evaluation of the systematic effects (including the magnetic field, red probe beam overlap and symmetry of excitation, and released atom drift velocity) was recorded for use in the final frequency evaluation.

Over a period of several weeks, we made a series of frequency measurements of our Ca standard relative to the NIST time scale, which is referenced to the Cs fountain. The resulting value, 455 986 240 494 158(26) Hz (1σ), represents a reduction in the uncertainty by nearly a factor of five over previous results. This value is in good agreement with past measurements at the Physikalisch-Technische Bundesanstalt (PTB) and NIST using both traditional and femtosecond-based frequency chains (see Fig. 3).[8-11] This good reproducibility highlights one aspect that distinguishes Ca from other optical standards: it is thus far the only optical standard at different laboratories to be evaluated and show agreement at the $\sim 10^{-13}$ level.



a)

Effect	Shift	Uncertainty
1st Order Doppler		20 Hz
Locking Errors		12.5
Collision Shift		10
RF offset	~10 Hz	4
AC Stark Shifts		3
Recoil Overlap		2
Blackbody Shift		1
Quadratic Zeeman	~23	1
Quadratic Stark		0.1
2nd Order Doppler		0.003
Total:		**26 Hz**

b)

Figure 3. (a) Comparison of Ca absolute frequency measurements. (b) List of systematic uncertainties.

Systematic uncertainties (~26 Hz) were much greater than the statistical uncertainties (~5 Hz) and determine the daily measurement error bars. This typical daily error bar was taken as the final measurement uncertainty since it was not clear that the data represented a random sampling of systematic shifts. The greatest systematic uncertainty in this measurement (20 Hz) was a consequence of the large residual drift velocity (~15 cm/s), which resulted from an asymmetry in the atom release from the trapping beams. Other important systematic effects included uncertainty in the lock point, collisional shifts, and an offset due to residual phase ringing on the rf pulses that drive the AOM's. In order to understand these effects and to see how they can be significantly reduced to improve the future performance of the standard, it is instructive to consider them more closely.

For our frequency standard, shifts due to the first-order Doppler effect can appear in several guises. First, if the red probe beams have a residual vertical component, a shift occurs when the released atoms fall through them. We corrected this shift by averaging pairs of measurements with the Ramsey-Bordé pulse directions reversed. Second, atoms moving through curved wavefronts can lead to phase chirps resulting in frequency shifts, so the use of high quality collimating optics and optical fibers (for spatial filtering) is essential. Finally, the combination of imperfect angular overlap of the counter-propagating probe beams and a transverse drift velocity of the atomic cloud leads to a shift as well. We attempted to minimize this effect by requiring that each probe beam couple into its opposing optical fiber. We then put an upper limit on this shift by directly measuring the transverse velocity of the atomic cloud. See the paper by Riehle *et al.* in these proceedings for an original approach to reducing these Doppler shifts.

The uncertainty in the lock point resulted from our square-wave modulation and 1-f detection method, which exposed us to shifts due to background asymmetries. We evaluated possible shifts due to our modulation scheme by using different modulation depths and were able to set a conservative upper limit. With more complex modulation/detection schemes, these shifts can be greatly reduced, although they will remain an issue as we attempt more accurate measurements.

One technical issue proved to be quite important in the course of this measurement. We discovered a systematic shift that depended on the Bordé-Ramsey fringe resolution (see Fig. 4), which was larger than our uncertainty budget. After an extensive search, which allowed us to rule out many parasitic effects, we found the dominant contributor to be related to the switches that controlled the rf signal going to our AOM's. As the switches turned the modulators on and off (the red pulses had a duration of 1.5 μs), they produced a "ringing" on our otherwise square pulses. This ringing led to a net phase shift of the fringe pattern, which in turn led to a resolution-dependent frequency shift. By improving the switching

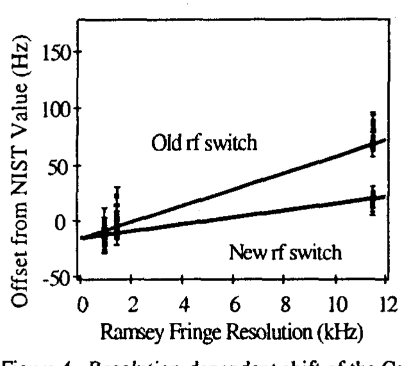


Figure 4. Resolution-dependent shift of the Ca standard frequency. This shift is reduced with improved switching electronics.

electronics, we reduced the shift by more than a factor of three. Nonetheless, this led to a correction of the data of 3 to 10 Hz. In future work, the use of direct digital synthesis waveforms should reduce this effect further, although frequency chirping in the modulators themselves may still present problems.

A potential shift worthy of discussion here is due to collisions between cold atoms. Such shifts are always a worry for neutral atom standards (they present one of the fundamental limits to the primary Cs standard) and their magnitude is hard to predict a priori. At our present performance we see no evidence of a collision-induced frequency shift for the clock transition. We were able to put an upper limit on this shift by changing the density of our sample and looking for a shift relative to the Cs-referenced frequency comb. For our estimated density of $3\times10^9/cm^3$ and atom temperature of 2 mK, we were able to rule out a shift at the 10 Hz (2.5×10^{-14}) level. As the standard evolves and we strive for measurements at the Hz level and below, a more careful search for shifts will be required.

4 Summary and future prospects

It seems appropriate to conclude with some general comments on our experience with the Ca standard and the improvements we need to implement. We have found the setup to be quite robust; with routine maintenance, the standard has been available as needed almost continuously over the past year. We were able to achieve good short-term stability as well as make a determination of the absolute frequency of the transition with a fractional uncertainty of 7×10^{-14}. When combined with earlier measurements of this transition frequency, our value sets an upper limit on the drift of this frequency (relative to the Cs standard's hyperfine transition) of $+2\pm8\times10^{-14}$/yr. This could useful in studying the possible drift of physical constants, a topic currently of great interest.[12]

However, it is clear that to improve the short-term stability and reduce the frequency uncertainty in the standard we will need to reduce the 3-D velocity of the atoms. One interesting option is to use the clock transition itself for cooling since its narrow frequency width can offer high velocity selectivity. In fact, the corresponding transition in Sr has been used to cool atoms to temperatures as low as 400 nK.[13,14] However, we are unable to implement the identical approach in Ca due to its weaker intercombination transition, which is too slow to cool the atoms before they expand out of the trapping beams. One solution is to quench the long-lived excited state by exciting another transition that pumps the atoms back to the ground state much faster than they otherwise naturally decay. This approach, which was first demonstrated for sideband cooling of trapped ions [15], enables one to take advantage of the velocity selectivity of the transition while still achieving sufficient cooling rates. Different implementations of this approach have recently been

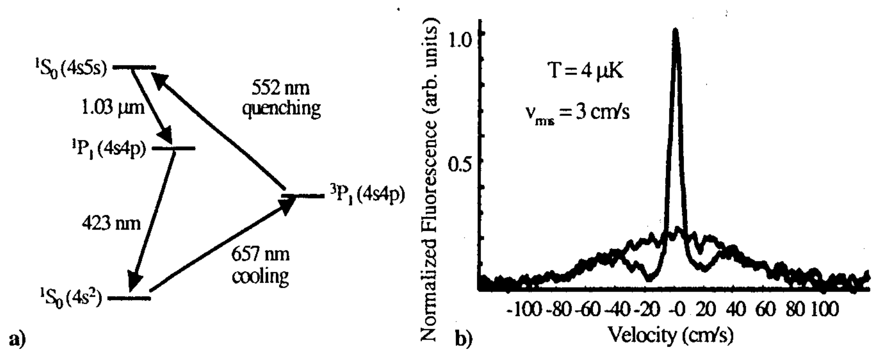


Figure 5. (a) Relevant energy levels for cooling. (b) Velocity distribution of atoms before and after 15 chirped and 10 stationary second-stage cooling cycles. Each cycle consisted of two counter-propagating, temporally separated, 5 μs red pulses followed by 20 μs of 552 nm light. A 40 μs post cooling pulse of 552 nm light was used to pump the remaining atoms back to the ground state. See Ref. [16].

demonstrated in 1-D here at NIST [16] and in 3-D at PTB [17] (in collaboration with W. Ertmer's group at Hannover). Figure 5 summarizes our recent results, in which we attained a 1-D temperature of 4 μK. Quenched cooling on the intercombination line in 3-D should enable us to reach temperatures of a few microkelvin, which in turn should enable frequency measurements in the low 10^{-15} range. However, we should emphasize that the current primary standards based on Cs fountains are poised to move below 10^{-15}, so we will need to develop techniques to move the Ca system into the 10^{-16} domain as well. One exciting possibility has been investigated by Katori *et al.*; they have trapped sub-microkelvin Sr atoms in an optical lattice, and have chosen the lattice wavelength to produce a near-zero relative Stark shift of levels of the clock transition. (See the paper by Katori *et al.* in these proceedings). While many experimental details must be worked out, this approach could be viable for Ca as well. Certainly the idea of having ten million little "clocks", each cooled to near zero motion inside its own potential well, is tantalizing and could well represent the future of neutral atom-based optical clocks.

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