OPTICAL CLOCKS WITH COLD ATOMS AND STABLE LASERS*

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The performance and prospects for neutral-atom optical frequency standards are discussed based on our recent progress with a calcium optical frequency standard. Second stage narrow-line cooling to microkelvin (and even 300 nK) temperatures, combined with launched atoms, should reduce Doppler frequency errors to about 1×10^{-16}. Advanced femtosecond optical frequency combs allow direct comparisons between the Ca optical standard, the Hg^+ optical standard and the Cs primary standard. These comparisons provide independent "reality checks" on both the stability and accuracy. Relative frequency measurements also constrain the possible time variation of atomic energy levels and fundamental constants.

1. Introduction

NIST is developing optical frequency standards based on laser-cooled and trapped calcium atoms, and also single Hg^+ ions.[1, 2] Both systems are promising candidates for the next generation of frequency standards and atomic clocks. In this paper we focus primarily on recent progress in laser-cooling and high precision spectroscopy of the calcium optical clock transition. The results are relevant to other neutral-atom frequency standards, particularly with respct to factors that affect stability and accuracy, which motivates much of our work. Precision spectroscopy of narrow transitions in cold (but moving) atoms deals with the questions of determining the exact lineshape and finding the center of the natural atomic resonance. The push toward atomic frequency standards based on optical transitions is inspired by the predicted improvement in stability and accuracy. Our work builds on the pioneering ideas and work of visionary scientists over more than 40 years.[3, 4] Just now, the ideas are coming to fruition because of combined advances in three technical areas of laser physics: laser cooling and trapping of atoms, highly stabilized cw-lasers, and a convenient method for optical synthesis using mode-locked lasers.

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1.1. Optical Frequency Standards: a Stability Advantage

It is clear that the use of higher-frequency oscillations for a clock will divide time into smaller units, and thus provide more precise timing and higher frequency stability. The fractional frequency fluctuations of an atomic frequency standard in the quantum projection noise limit, with N atoms and averaging time τ varies as $(\Delta\nu/\nu_0)(1/N\tau)^{1/2}$. A simplistic interpretation of the atomic frequency stability suggests that an optical standard with 10^6 atoms and a 1 Hz linewidth will give[5] $\sigma_y(\tau) \approx 2\times10^{-19}\ \tau^{-1/2}$. However, it might be foolish to extrapolate so far beyond our present experience. To reach the more fundamental atomic limits will require efficient use of the atoms, and extremely stable lasers as local-oscillators. Outstanding progress in that direction has been demonstrated with lasers locked to cavities at the mHz level,[6] and also with the stability of the combined laser-plus-cavity system at the $<5\times10^{-16}$ level for 0.5 to 200 s.[7]

In our present Ca frequency standard there are several factors that limit the short-term stability, including a natural linewidth of about 470 Hz, and a very poor ratio of "clock time" (about 1 ms) to cycle time ($\approx$ 30 ms). Nonetheless, an instability of about $2\times10^{-16}\ \tau^{-1/2}$ should be possible.[5, 8] Fortunately, the stability of our laser when locked to a short, high-finesse optical cavity ($\approx$ 2 Hz at 1 s) is reasonably well matched to the present Ca atomic stability.[2]

2. Calcium Optical Frequency Standard

The calcium inter-combination transition at 657 nm ($^1S_0 \leftrightarrow {}^3P_1$) was identified long ago for its potential use as an optical frequency standard/clock. Our work benefits notably from the calcium project at PTB (U. Sterr *et al.*, see this Proceedings) and in many ways is a collaborative effort.

The 657 nm clock transition is spectrally sharp (470 Hz natural width) and can be detected with excellent signal-to-noise using shelving detection, factors which have led to demonstrated frequency instability[2] of $\approx 4\times10^{-15}\tau^{-1/2}$. Furthermore, the calcium transition is remarkably immune from perturbing effects due to external electric ($\approx 3\times10^{-6}$ Hz/(V/m)^2) and magnetic fields (0.5 Hz/G^2), and any cold-atom collision shift is so small that it proves difficult to detect. The Ca resonance line at 423 nm (34 MHz linewidth) allows rapid cooling, but is limited to simple Doppler cooling temperatures of $\approx$ 2 mK, corresponding to velocities of $\approx$70 cm/s. This significant thermal velocity and residual drift velocity when the atoms are released from the MOT, combined with experimental constraints on the geometry of the probe laser fields, put real limits on the accuracy that could be achieved. To date, the uncertainties in the clock frequency at 456 THz are 21 Hz at NIST, and 8 Hz at PTB, due primarily to 1st-order Doppler-related shifts.[9,10] While the Ca frequency standard has exceptional stability, it is not yet competitive in accuracy with the best microwave standards and single-ion optical standards that now show reproducibility at about 1×10^{-15} ($\approx$ 1 Hz at optical frequencies).

2.1. Doppler Problems

The seriousness and complexity of the first-order Doppler-related frequency shifts have been addressed in a number of papers, including seminal work by Bordé and Hall,[11] and recent papers focusing on cold atom clocks and interferometers.[12, 13] The basic problem is that time-dependent phases of optical fields seen by the atom appear as frequency shifts to the observed line center. For an atom in free-flight with gravity, and interacting with a laser field, the optical phase that the atom sees is a function of position $\boldsymbol{r}$ and interaction time $\boldsymbol{t_i}$, and is given approximately as

$$\Phi(\vec{r}(t_i)) = \vec{k}\cdot(\vec{r}_0 + \vec{v}_0\cdot t_i + \frac{1}{2}\vec{g}\cdot t_i^2) + k\,\frac{r_{perp}(t_i)^2}{2R}$$

where $\boldsymbol{v_0}$ is the initial atomic velocity, $\boldsymbol{g}$ is the gravitational acceleration, $\boldsymbol{r_{perp}}$ is the radial distance of the atom from the center of the laser beam and $\boldsymbol{R}$ is the radius of curvature of the fields.

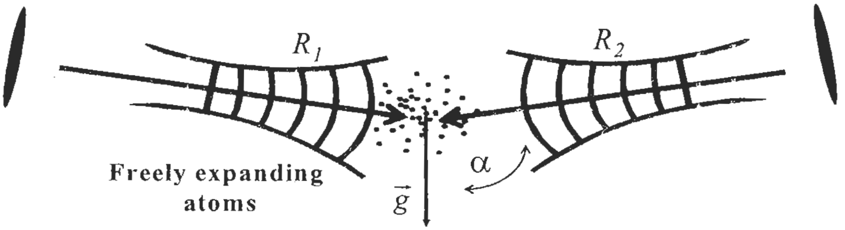

Figure 1. Geometry for saturated absorption spectroscopy of a ball of cold atoms.

In the present context – saturated absorption spectroscopy of a sample of cold atoms released from a MOT – the motional phase shifts can be separated into a number of different terms that have simple physical descriptions and specific functional dependencies on $\boldsymbol{R}$, $\boldsymbol{v}$, $\boldsymbol{t_i}$, and $\boldsymbol{g}$. For our situation the dominant terms are the atom velocity coupled to the imperfect cancellation of the forward and backward *k*-vectors, and the wavefront curvature of the probe fields. With single-stage cooling and mK atoms we typically have: $v_{thermal} \approx 70$ cm/s, $v_{drift} \approx 10$ cm/s, *k*-vector misalignment uncertainty ≈ 40 μrad, angle to gravity $(\alpha-\pi/2) < 1$ mrad, and $R \geq 40$ m. Using these parameters and interaction times of ≈ 1 ms, the dominant terms each result in frequency shifts of 1 to 10 Hz, fractionally a few $\times10^{-14}$! Thus, our recent efforts (and similarly those of PTB) have focused on advanced laser cooling schemes to reduce the velocity, combined with atom interferometry to improve the probe beams.

2.2. Quenched Narrow-Line Cooling

Since Ca lacks hyperfine structure that can provide "free" sub-Doppler cooling, we instead take advantage of the narrow clock transition that offers excellent velocity selectivity (0.3 mm/s for a 470 Hz linewidth). The 370 μs lifetime is too long to be useful for simple Doppler cooling of mK atoms in gravity. Nevertheless, an additional laser can be used to quench the $^3P_1$ excited state more rapidly and allow a reasonable cooling rate with the narrow clock transition. By simultaneously applying 657 nm and 552 nm light we observe temperatures of ≈10 μK in 3-dimensions while retaining ≈ 30 % of the atoms, as shown in Fig. 2.

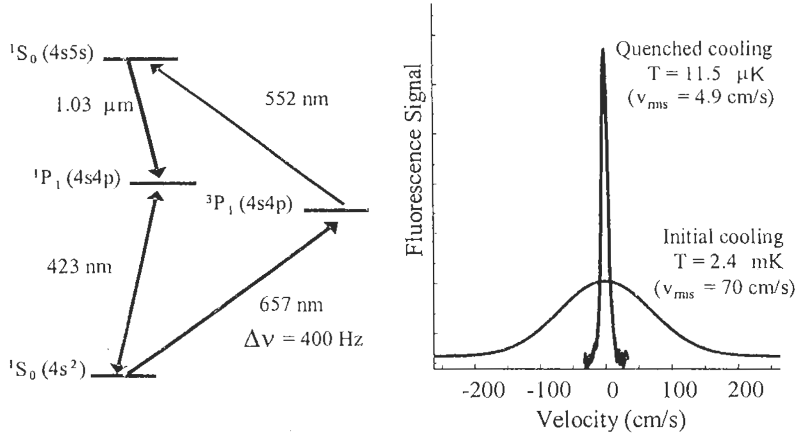

Figure 2. Quenched narrow line cooling of calcium atoms using 657 nm plus 552 nm. The right panel shows the velocity distribution with just 423 nm cooling, and with additional second stage cooling.

A pulsed variant of the quenched-cooling method reduces the velocity by another factor of five and produces sub-recoil 1-D temperatures ≈ 300 nK, $v \approx$ 1 cm/s.[9] This pulsed narrow-line cooling could be done in 3-D but it would be somewhat complex.

With the Ca thermal velocity reduced by a factor of fifteen, the velocity-dependent systematic errors are correspondingly reduced and the signal contrast is increased (Fig. 3). Unfortunately, even velocities of 5 cm/s will not be good enough to reach the accuracies that we strive for, and we are left with several Doppler frequency shifts of about 200 mHz, fractionally 5×10^{-16}. Obviously, it would be advantageous to have the atoms confined to the Lamb-Dicke limit, as is possible with ions. Several people have proposed using optical lattices for this purpose.[14] Optical clocks based on these concepts look promising, but additional complications such as magnetic structure need to be worked out. In the mean time, substantial further progress can be made with our existing cold neutral atoms.

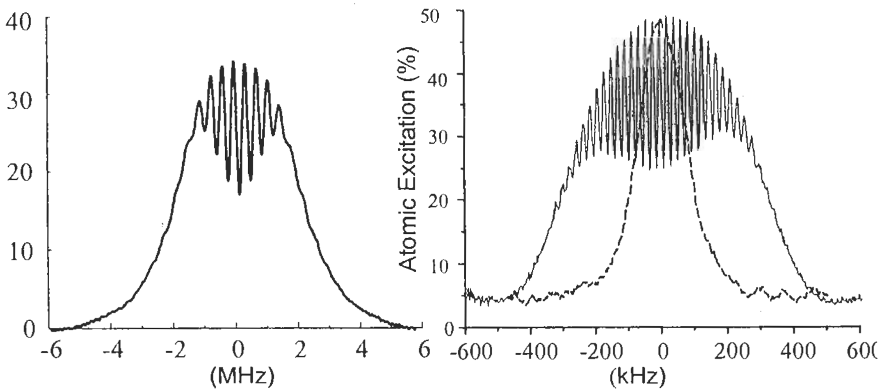

Figure 3. Saturated absorption optical Ramsey fringes on the 657 nm calcium clock transition created by the four-pulse Bordé method. Both vertical axes are in percentage of atoms excited, and the horizontal axes show laser frequency minus calcium frequency (but note the different scales). The left-hand panel shows a low-resolution scan of the fringes taken with 2.5 mK atoms, while the graphs on the right use 10 μK atoms. The dashed trace without fringes shows the Doppler width of the 10 μK atoms. The underlying width of the Ramsey pedestal results from the Doppler width in the case of the mK atoms, while in the case of the 10 μK atoms it is the Fourier-transform limit of the probe pulses.

To further reduce the Doppler and beam geometry problems we are now using cold atoms to diagnose the quality of the probe laser fields. The idea is simple but powerful: by launching balls of cold atoms with m/s velocities, rather than cm/s, we amplify by ≈ 200 times the problems associated with non-ideal optical probe beams. To do this, we use a 3-pulse collinear non-frequency selective atom interferometer described by Trebst.[13] Launching the atoms through the probe laser fields, we enhance the phase-shifts resulting from wavefront errors and can use the results to adjust the wavefronts of the laser fields. Similarly, the 4-pulse Ramsey-Bordé frequency-sensitive interferometer is used with launched atoms to adjust the probe beam alignment. Controlling both the launch velocity and the interaction time allows us to separate the wavefront errors from those associated with gravity and spatial offset. Preliminary results indicate that these methods allow correction of the laser wavefront to an effective radius of > 300 m, and the beam alignment to about 5 microradians. This implies that we should be able to reduce the velocity-dependent frequency shifts to a level of $\approx 1\times10^{-16}$. If so, they will be well below other frequency uncertainties. We must now more seriously address effects such as the 300 K blackbody radiation induced Stark shift, the Stark shift due to the clock laser itself, any collision shift, the residual phase-shifts in the probe fields due to AOMs and switches, inhomogeneity in optical probe fields, and other mechanical-, thermal- and gravity-induced optical phase changes. At this point, we don't really know what the ultimate limitation in accuracy will be for the calcium optical frequency standard.

3. Optical Frequency Combs and Comparisons of Atomic Standards

Optical frequency combs based on femtosecond mode-locked lasers are used with our optical frequency standards to realize optical atomic clocks, to coherently connect optical and microwave frequencies, and to make intercomparisons between the Ca, Hg^+ and Cs standards at NIST. Recent advances in comb technology greatly extend their useful spectral coverage and make the systems quite reliable, allowing long-term operation. In particular, a 2/3-octave frequency comb is created with a broadband 1 GHz Ti:Sapphire laser that is self-referenced using the 2f-3f method without any microstructure fiber.[15] Coherently linking this broadband laser with a ≈ 500 MHz mode-locked Cr:Forsterite laser (1.3 μm) provides a comb of modes from 570 to 1450 nm, which can be referenced to the optical standards or a microwave source.[16]

In both the optical and microwave regions, we have explored the time- and frequency-domain characteristics of optically-controlled femtosecond combs, and find that they have remarkably high fidelity in accuracy and stability. In fact, many of our results are still limited by our measurement systems rather than by the performance of the combs. At the present time, the optically-referenced frequency combs at NIST show:

- Frequency reproducibility optical-to-optical $\leq 4\times10^{-17}$
- Frequency instability optical-to-optical $\leq 6.3\times10^{-16}\ \tau^{-1}$
- Repetition rate instability by optical cross-correlation $\leq 2\times10^{-15}\ \tau^{-1}$
- Timing jitter detected by optical cross-correlation ≤ 0.4 fs (1-100 Hz BW)
- Photodiode-generated microwave instability $\leq 1\times10^{-14}\tau^{-1}$
- Phase-noise on photodiode-generated 1 GHz < -125 dBc/Hz (100 Hz offset)
- Reproducibility of repetition-rate detected on photodiodes $< 2\times10^{-16}$.

These results indicate that femtosecond optical frequency combs are not a limitation to the performance of the current generation of optical atomic clocks.

If we use atomic transitions as our references for time, frequency, and length measurements, we must be confident that these do not vary with the operating conditions (other than the effects predicted by Einstein's relativity). There is always the fundamental question that Dirac raised, of whether the forces of nature and structure of atoms evolve with time; and thus whether our reference of time depends on time. Renewed interest in this topic comes from three areas: an increasing body of astronomical data suggesting[17] that about 10 billion years ago the fine structure constant was different from the present value by 1 part in 10^5, new test theories that are being formulated that parameterize searches for physics beyond relativity and the standard model,[18, 19] and that now there is more than one type of atomic frequency standard that can provide accuracy at the 10^{-14} to 10^{-15} level.

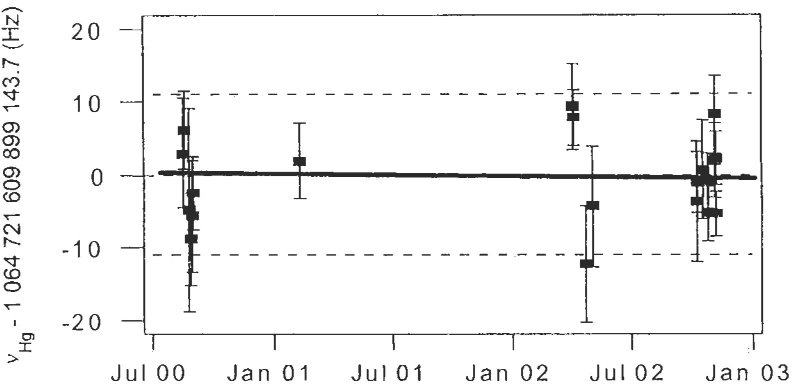

Figure 4. Hg^+ optical frequency relative to the cesium primary frequency standard.[21]

Using optical frequency combs, we have made several comparisons of the relative frequency of Ca, Hg^+ and Cs standards. Published frequency measurements of Hg^+ relative to the NIST-F1 primary cesium standard are shown in Fig. 4. The average of all the data gives the Hg^+ optical frequency with uncertainties of ±1.1 Hz statistical and ±10 Hz systematic for the electric quadrupole shift not yet evaluated. A linear fit using the total uncertainty gives a slope of -0.24 ± 1.3 Hz/yr, indicated by the bold line. Following the Prestage-Dzuba[17, 20] model for evaluating frequency comparisons in terms of a possible time variation of the fine structure constant α, and assuming that $g_{Cs}(m_e/m_p)$ is constant, gives $(1/\alpha)(d\alpha/dt) \leq 0.5 \pm 1.1 \times 10^{-15}$/year. Similarly, measurements of calcium in terms of cesium have been made at PTB through the years using harmonic frequency chains, and more recently at NIST and PTB using optical combs as shown in Fig. 5.

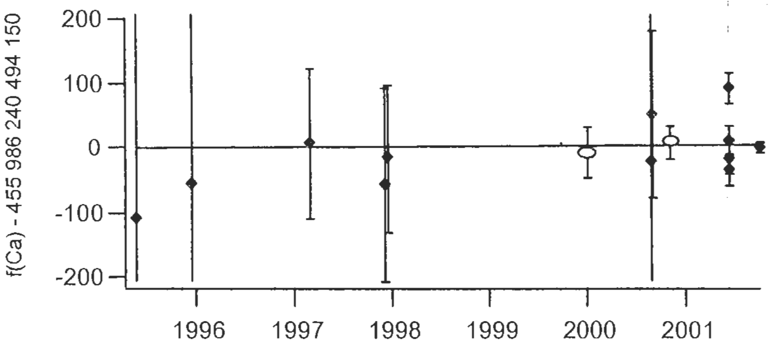

Figure 5. Ca optical frequency in Hz from published results from NIST (symbol-O) and PTB (symbol-♦) for the past 6 years. The line represents the weighted linear fit.

The data show steady improvement in accuracy of the Ca standard over time, but no obvious temporal dependence. A weighted linear fit to the data gives a slope of 0.8 ±12 Hz/yr. At the present accuracy, we find no systematic temporal variation of either the Hg^+ or Ca optical frequencies relative to cesium.

4. Summary

Optical atomic clocks are no longer just a promise of the future, but are here today. The reported reproducibility for some the ion-based optical standards are similar to the best microwave atomic frequency standards, and optical neutrals look as though they can also be competitive. Short-term instabilities of optical standards are already orders of magnitude better than other sources. With rapid improvements in all the atomic standards due to cold atoms, better lasers, and microwave sources, we anticipate more stringent tests of fundamental physics, such as time variation of fundamental constants, Einstein's relativity, symmetry postulates, and searches for other forces, as well as new technical capabilities and applications.

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