

**ALL OPTICAL ATOMIC CLOCKS BASED ON  
A SINGLE MERCURY ION AND CALCIUM ATOMS**

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**ABSTRACT**

The frequency comb created by a femtosecond mode-locked laser and a microstructure fiber is used to phase-coherently measure the frequencies of both the  $\text{Hg}^+$  and Ca optical frequency standards with respect to the SI second as realized at NIST. We find the transition frequencies to be  $f_{\text{Hg}} = 1\,064\,721\,609\,899\,143(10)$  Hz and  $f_{\text{Ca}} = 455\,986\,240\,494\,158(26)$  Hz, respectively. This work begins to reveal the high stability and accuracy potential of optical atomic clocks based on the  $\text{Hg}^+$  and Ca standards. Furthermore, when combined with previous measurements, we find no time variations of these atomic frequencies within the uncertainties of  $|\partial f_{\text{Hg}}/\partial t|/f_{\text{Hg}} < 2 \times 10^{-14} \text{ yr}^{-1}$  and  $|\partial f_{\text{Ca}}/\partial t|/f_{\text{Ca}} < 8 \times 10^{-14} \text{ yr}^{-1}$ .

Keywords: Optical, Atomic Clock, femtosecond, comb

**INTRODUCTION**

Optical frequency standards based on laser-cooled atoms and ions promise superior stability and accuracy over existing microwave standards [1–5]. However, because of their high frequencies ( $\sim 10^{15}$  Hz or PHz), it has proven difficult to count cycles as required for building functioning clocks. Only recently, a reliable and convenient clockwork fast enough to count optical oscillations has been realized [6–9]. Here, we report on work toward an all-optical clock based on a femtosecond laser that phase-coherently divides down the frequency of the visible radiation from either a  $\text{Hg}^+$  or Ca optical-frequency standard to a countable radio frequency. We have measured the absolute frequencies of these optical transitions in terms of the SI second as realized at NIST [10]. Indeed, for the  $\text{Hg}^+$  standard, the uncertainty in the measurement is essentially limited by our knowledge of the SI second at  $\sim 2 \times 10^{-15}$ . Additionally, the comparison of atomic frequencies over time provides constraints on the possible time variation of fundamental constants. We now have measurements of the absolute  $\text{Hg}^+$  frequency taken over a six month interval that differ by less than their statistical uncertainty and much less than their systematic uncertainty.

**THE OPTICAL STANDARDS**

The  $\text{Hg}^+$  and Ca systems have recently been described elsewhere [1,4, 11–13], so we summarize only their basic features. The mercury optical frequency standard is based on a single, laser-cooled  $^{199}\text{Hg}^+$  ion that is stored in a cryogenic, radio-frequency, spherical Paul trap. The ion is cooled and detected by driving the  $^2S_{1/2} - ^2P_{1/2}$  cycling transition at 194 nm. The  $^2S_{1/2}$  ( $F=0, M_F=0$ ) -  $^2D_{5/2}$  ( $F=2, M_F=0$ ) electric-quadrupole transition at 282 nm [Fig.1] provides the reference for the optical standard [1]. We lock the frequency-doubled output of a 563 nm dye laser with

sub-hertz linewidth[12] to the quadrupole resonance. Transitions to the metastable  $^2D_{5/2}$  state are detected with near unit efficiency since the absorption of a single 282 nm photon suppresses the scattering of many 194 nm photons on the strongly allowed  $^2S_{1/2} - ^2P_{1/2}$  transition [14, 15]. In figure 2 we show an example of a normalized spectrum that was obtained from multiple, bidirectional scans through the resonance, where the probe time was 20 ms. Most often, the frequency was locked to resonance with a 10 ms interrogation period, which yielded a fractional frequency stability of  $2 \times 10^{-15} \tau^{-1/2}$  for an averaging time  $\tau$  measured in seconds [16].

The calcium standard starts with a collection of  $\sim 10^7$  laser-cooled  $^{40}\text{Ca}$  atoms held in a magneto-optic trap. The 423 nm  $S_0 - P_1$  transition is used for trapping and Doppler-cooling the atoms to a residual temperature of  $\sim 2$  mK. The  $^1S_0$  ( $M_J=0$ ) -  $^3P_1$  ( $M_J=0$ ) weakly allowed electric-dipole transition (400 Hz natural linewidth) at 657 nm is the “clock” transition for this frequency standard [Fig. 3]. We excite the transition with a four-pulse Bordé-Ramsey sequence (pulse duration = 1.5  $\mu\text{s}$ ) with light from a continuous-wave (CW), frequency-stabilized diode laser. Using a shelving detection technique similar to that employed in the  $\text{Hg}^+$  system, near-resonant 423 nm pulses (5  $\mu\text{s}$  duration) are used before and after the 657 nm excitation to determine the fraction of atoms transferred from the ground state. Figure 4 shows the Bordé-Ramsey fringes taken at a resolution of 960 Hz. This system has demonstrated a fractional frequency stability of  $4 \times 10^{-15} \tau^{-1/2}$  when probing sub-kilohertz linewidths[4]. For the measurements presented here the Ca spectrometer was operated with linewidths ranging from 0.96 to 11.55 kHz, which are integer submultiples of the recoil splitting.

**THE OPTICAL COMB**

The recent introduction of mode-locked lasers to optical frequency metrology greatly simplifies the task of optical-frequency measurements [6–8, 17–19]. The spectrum emitted by a mode-locked laser consists of a comb of regularly spaced continuous waves that are separated by the pulse repetition rate  $f_r$ . The frequency of the  $n^{\text{th}}$  mode of the comb is given by  $f_n = nf_r + f_0$  [20, 21], where  $f_0$  is the frequency offset common to all modes. This offset is caused by the difference between the group- and the phase-velocity inside the laser cavity.  $f_r$  can be measured by direct detection of the laser’s output with a photodiode.  $f_0$  is measured by heterodyning the 2<sup>nd</sup> harmonic of mode  $f_n = nf_r + f_0$  from the infrared wing of the comb with mode  $f_m = 2mf_r + f_0$  from the blue side of the comb [7, 8]. This self-referenced technique requires that the optical comb span at least an octave in frequency space.

While an octave-spanning comb can be produced directly from a mode-locked laser [22], launching the longer pulses from a

commercially-available femtosecond laser into an air-silica microstructure fiber [23, 24] also produces a frequency comb that spans an octave. Nonlinear processes in the fiber produce the additional equally spaced and phase-coherent modes to the transmitted light. It has been demonstrated that this process of spectral broadening preserves the uniformity of spacing and spectral fidelity of the comb to at least a few parts in  $10^{16}$  [8].

We couple approximately 200 mW average power from a femtosecond, Ti:sapphire ring laser ( $f_r \approx 1$  GHz) through a 15 cm piece of microstructure fiber that has a 1.7  $\mu\text{m}$  core and group-velocity dispersion that vanishes near 770 nm [23]. This power density is sufficient to increase the spectral width of the laser from 13 THz to more than 300 THz, spanning from  $\sim 520$  nm to  $\sim 1170$  nm. The infrared part of the comb from the fiber ( $\lambda \sim 1060$  nm) is split off by a dichroic mirror and frequency-doubled into the green portion of the visible spectrum by means of a  $\text{KNbO}_3$  crystal 2 mm long. Following an adjustable delay line that matches the optical path lengths, the frequency-doubled light is spatially combined with the green part of the original comb to produce a beat note at  $f_o$ . We phase-lock both  $f_o$  and  $f_r$  to synthesized frequencies derived from a cavity-tuned hydrogen maser that acts as the transfer standard to the NIST realization of the SI second [10]. Control of  $f_r$  is achieved with a piezo transducer driving a cavity mirror, while  $f_o$  is controlled by adjusting the 532 nm pump beam's intensity with an electro-optic modulator [8]. When both  $f_o$  and  $f_r$  are phase-locked, the frequency of every mode in the comb is known with the same accuracy as that of the reference maser.

The CW light from the  $\text{Hg}^+$  (563 nm) and Ca (657 nm) standards is transferred to the mode-locked laser system via two single-mode optical fibers that are 130 m and 10 m long, respectively. Approximately 2 mW of CW light from each fiber is mode-matched with the appropriate spectral region of the frequency comb to generate a beat signal  $f_b$  with a nearby mode. This beat note is amplified and measured with a counter. The optical frequency is then expressed as  $f_{\text{opt}} = mf_r + f_o + f_b$ , where  $m$  is a large integer uniquely determined for each system from previous coarse measurements of  $f_{\text{opt}}$ .

## RESULTS

Figure 5 summarizes the frequency measurements of  $\text{Hg}^+$  made between August 2000 and February 2001, while figure 6 summarizes the Ca measurements made from October 26 to November 17, 2000. The frequency of the maser is calibrated by comparing to the local NIST time scale (5 hydrogen masers and 3 commercial cesium clocks), which in turn is calibrated by the local cesium fountain standard (NIST-F1 [10]). The fractional uncertainty in the frequency of the reference maser relative to the SI second was about  $1.8 \times 10^{-15}$  for these measurements.

The weighted mean of our measurements of the  $\text{Hg}^+$  clock transition is  $f_{\text{Hg}} = 1\,064\,721\,609\,899\,143$  Hz. The statistical uncertainty ( $\approx 4 \times 10^{-15}$ ) of our limited-duration measurements is essentially the result of the reference-maser's short-term stability ( $\sim 2 \times 10^{-13} \tau^{-1/2}$ ). Because we have not made a full experimental evaluation of the  $\text{Hg}^+$  standard, we assign a very conservative value of 10 Hz for the total systematic uncertainty. The

dominant contribution to the uncertainty of the  $S$ - $D$  transition frequency is the electric-quadrupole shift of the  $^2D_{3/2}$  state arising from coupling with the static potentials of the trap. In our spherical Paul trap, where the confinement of the ion uses no static applied fields, the maximum quadrupole shift should be less than 1 Hz (or fractional frequency shift  $< 10^{-15}$ ) [25]. In principle, it is possible to eliminate the quadrupole shift by averaging the  $S$ - $D$  transition frequencies for three mutually orthogonal orientations of quantizing magnetic field of constant magnitude. In the present experiment, we have measured the  $S$ - $D$  frequency for various field values, but we have made no attempt to eliminate the quadrupole shift by using three orthogonal fields of constant magnitude. No shift of the resonance frequency is observed within the precision of these measurements even under strongly varying conditions of magnetic field. We anticipate that the uncertainties of all systematic shifts in the  $\text{Hg}^+$  system can be reduced to values approaching  $1 \times 10^{-18}$  [1, 25].

For the Ca data shown [Fig. 6], an additional correction is applied each day to account for frequency shift caused by residual phase chirping on the optical Ramsey pulses produced by amplitude modulating an acoustooptic modulator (AOM). The phase chirping produced resolution-dependent frequency shifts on the order of 100 Hz for fringes 11.5 kHz wide but of only 10 Hz for fringes 0.96 kHz wide. On each day, the Ca frequency was measured for  $\sim 30$  minutes at each of several fringe resolutions, and the zero-intercept of a linear fit to the data was used as the corrected frequency. On the last 3 days of measurements, we were able to reduce this shift by a factor of  $\sim 3$  with improvements to the RF pulses that drive the AOMs. The statistical uncertainty (typically 8 Hz) for each day's measurement is smaller than the uncontrolled systematic uncertainties in the Ca frequency. The largest systematic uncertainty stems from incomplete knowledge of the angular overlap of the counter-propagating beams in the Ca spectrometer, combined with transverse drift velocity of the cold Ca ensemble. This leads to residual first-order Doppler shift with magnitude  $< 15$  Hz (except on November 16, where a large drift velocity led to an uncertainty of  $\sim 52$  Hz). Other significant uncertainties include our lack of knowledge or control of electronic offsets and baseline symmetries ( $< 12$  Hz), wavefront curvature ( $< 10$  Hz), and cold-atom collisional shifts ( $< 10$  Hz). Taking all known systematic uncertainties in quadrature gives a confidence level of  $\sim 26$  Hz for the measured mean values indicated by the dashed lines in figure 6.

Figure 6 also shows the good agreement between our measurement and the most recent value measured with a harmonic frequency chain [26], which provides a degree of confidence in the reproducibility of the Ca standards. It is also in reasonable agreement with the very recent PTB measurements made with a femtosecond comb [27]. An additional measure of the Ca frequency can be made by using the present absolute measurement of  $\text{Hg}^+$  and our earlier measurement of the 76 374 564 455 429(40) Hz gap between  $f_{\text{Hg}}$  and the Ca standard [28]. This yields a value of  $f_{\text{Ca}} = 455\,986\,240\,494\,143(40)$  Hz in good agreement with the value from the present direct measurement.

Finally, these results also provide data on the relative time variability of atomic frequencies. S. Karshenboim has recently reviewed the implications of such comparisons and their

contribution toward constraining the possible time variation of fundamental constants [29]. In this regard  $Hg^+$  and Ca are two of the most interesting cases to study. Comparing our present measurement of  $f_{Ca}$  to measurements made by PTB in 1997 [26] gives  $(\partial f_{Ca}/\partial t)/f_{Ca} < 8 \times 10^{-14} \text{ yr}^{-1}$ . Similarly, our August 2000 to February 2001 measurements on  $f_{Hg}$  provides an initial baseline constraint on the time variation of  $(\partial f_{Hg}/\partial t)/f_{Hg} \leq 2 \times 10^{-15} \text{ yr}^{-1}$ . Here we use the defined unit of time based on the frequency of the Cs hyperfine interval and assume that any time dependence is slow and dominantly linear over the relevant time scale. We believe this represents the tightest laboratory test yet of the time variability of these disparate transitions.

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