

# PRIMARY FREQUENCY STANDARDS AT NIST\*

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## Abstract

*NIST has a more than 50-year history of developing ever more accurate atomic frequency standards. For most of that time, the technology was based on thermal atomic beams of cesium atoms, and the accuracy improvement was approximately a factor of 10 every 7 years. To put this into some kind of perspective, both the period of this history and the rate of improvement are almost identical to the development of information-storage density in magnetic media. Now, however, with the advent of laser-cooling schemes and optical-frequency metrology, the rate of progress is dramatically increasing. This paper discusses briefly the last of the thermal-beam standards and our first laser-cooled, atomic-fountain standard. It then goes into some detail about the newly developed all-optical standards that use an optical-frequency transition in a single, laser-cooled, trapped mercury ion or in an ensemble of laser-cooled and trapped calcium atoms. Based largely on the increased operating frequency of the “clock” transition, these new standards have the potential for several orders of magnitude improvement in stability and, in the case of the mercury standard, accuracy over today’s standards operating in the microwave region.*

## INTRODUCTION

NIST has the primary responsibility within the US for the accurate realization of the SI second, the national standard of time interval. In fact, however, time and its standard interval are coordinated around the world in such a way that the product delivered to the American people is not that from our standards laboratories, but rather that agreed upon from a world average of good standards. As such, the output from our national standard is reported to the BIPM office in Paris, where it is averaged together with that of all other good standards. In return, we get “corrections” to our time scale that result in the information that is disseminated to our customers.

As our part in the international community of timekeepers, NIST and its predecessor, NBS, has a 50-year history of developing evermore accurate atomic frequency standards. Until 1999 these standards were always based on an atomic beam of thermal cesium atoms. The last standard of this type, NIST-7 [1], produced an accuracy of about five parts in  $10^{15}$ , limited by the short interaction time with the atoms. A newer technology makes use of laser-cooled atoms [2]. After the atoms are collected and cooled by the interaction with three pairs of opposed laser beams, they are tossed up about 1 m through the apparatus by the same light forces that produce the cooling. As they travel upward and then fall back under the action of gravity in a fountain-like spray, they interact with the standard almost two orders of magnitude longer than in the thermal atomic beam predecessor. Unfortunately, new collision physics enters into this cold-atom clock and the apparent potential of the improved line Q is not fully realized. At present, this relatively new technology results in a realization of the SI second with an uncertainty in accuracy of about one part in  $10^{15}$ .

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Science has long understood that both accuracy and stability can, in many ways, be scaled as the frequency of the transition used for the standard. For this reason we have long sought ways to measure frequencies corresponding to the optical region of the spectrum. These frequencies, approximately  $10^{15}$  Hz or some *five orders of magnitude higher* than the microwave frequencies used in current standards, offer the potential for improvements in both stability and accuracy that is vastly beyond anything that could ever be achieved through future improvements to current microwave standards. For this reason, NIST has for years had a program to investigate this potential and prepare for the day when optical frequencies could be measured directly. That day arrived with the recent advent of the mode-locked-laser, optical-comb generator [3-9].

Here, we report on work toward all-optical clocks based on a femtosecond laser that phase-coherently divides the frequency of the visible radiation from either a  $\text{Hg}^+$  or a Ca optical-frequency standard down 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 [2]. Indeed, for the  $\text{Hg}^+$  standard, the uncertainty in the measurement is essentially limited by our best current realization of the SI second,  $\sim 2 \times 10^{-15}$ . 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 6-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 [10-14], 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 laser-cooled and detected by driving the  $^2\text{S}_{1/2} - ^2\text{P}_{1/2}$  cycling transition at 194 nm (Figure 1).

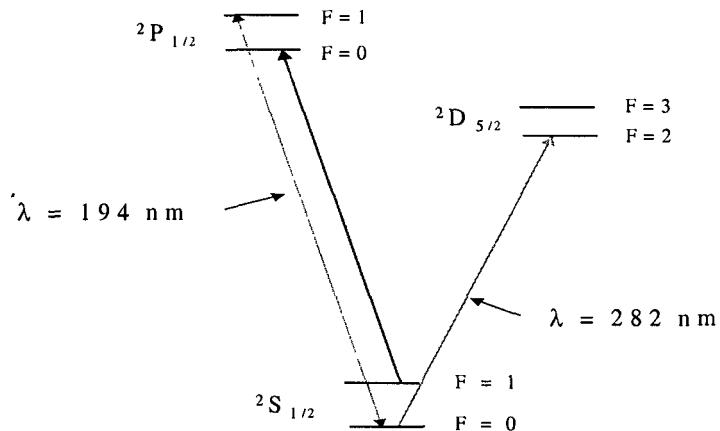


Figure 1. Partial level scheme for  $^{199}\text{Hg}^+$  ion. The 194 nm radiation is used for Doppler cooling, state preparation, and detection. The 282 nm transition from the ground state  $^2\text{S}_{1/2}(F = 0, M_F = 0)$  to the metastable  $^2\text{D}_{5/2}(F = 2, M_F = 0)$  state is the “clock” transition.

The  $^2\text{S}_{1/2}(F = 0, M_F = 0) - ^2\text{D}_{5/2}(F = 2, M_F = 0)$  electric-quadrupole transition at 282 nm provides the reference for the optical standard [10]. We lock the frequency-doubled output of a 563 nm dye laser with subhertz

linewidth [13] 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 [15,16]. Figure 2 shows an example of a normalized spectrum that was obtained from multiple, bidirectional scans through the resonance, where the probe time was 20 ms.

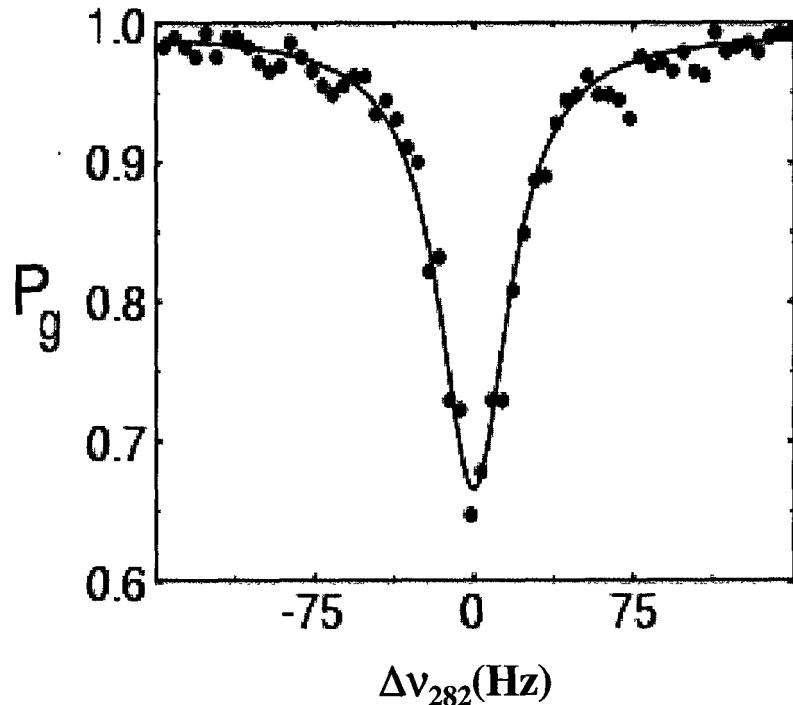


Figure 2. Typical spectrum of the 282 nm “clock” transition.

Most often, the frequency was locked to resonance with an interrogation period of 10 ms, which should produce a fractional frequency stability of about  $1 \times 10^{-14} \tau^{-1/2}$  for an averaging time  $\tau$  measured in seconds [17].

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  $^1S_0 - ^1P_1$  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 (Figure 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 [11]. 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 [3-9]. 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  $f_n = nf_r + f_0$  [18,19], where  $f_0$  is the frequency offset common to all modes. This offset is caused by the difference between the group and phase velocities inside the laser cavity.

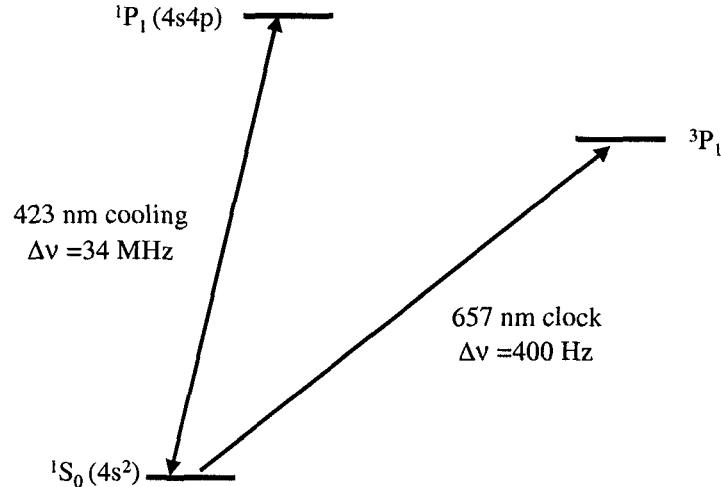


Figure 3. Simplified diagram of the relevant energy levels in the Ca standard.

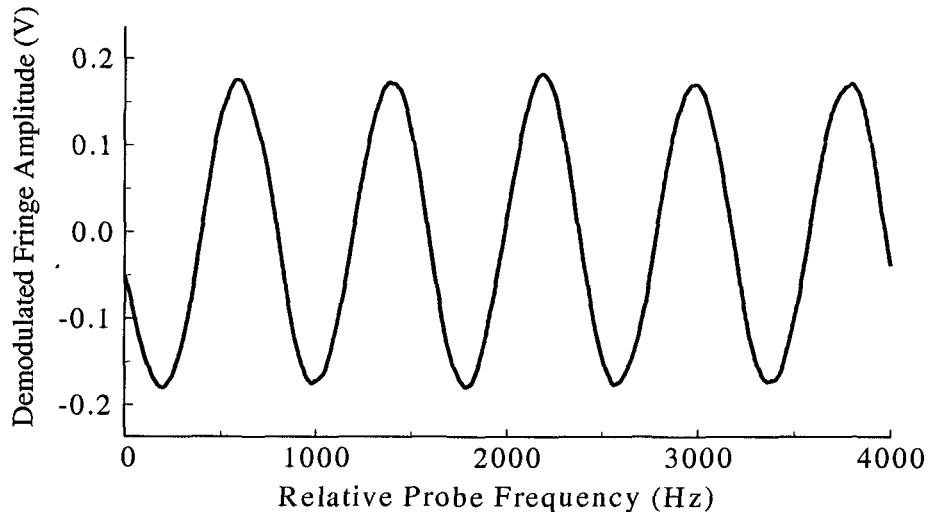


Figure 4. Optical Bordé-Ramsey fringes with a 960 Hz (FWHM) resolution. The total averaging time to generate the data for this figure was 60 s.

$f_r$  can be measured by direct detection of the laser's output with a photodiode.  $f_0$  is measured by heterodyning the blue side of the comb, as shown in Figure 5. This self-referenced technique [7,8] 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 [20], launching the longer pulses from a commercially available femtosecond laser into an air-silica microstructure fiber [21,22] 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].

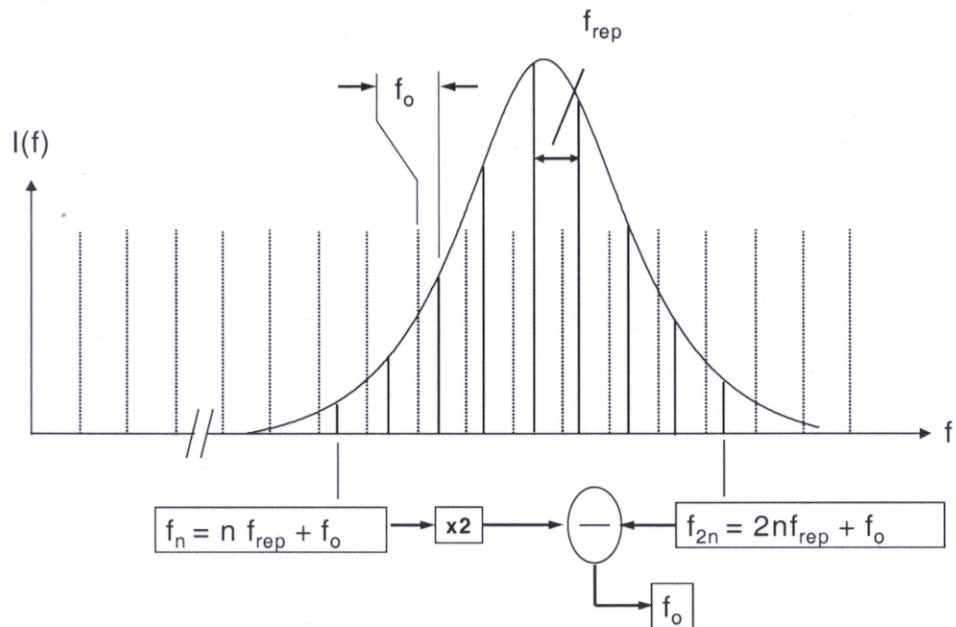


Figure 5. Schematic of the “self referencing” technique used to generate a signal at the offset frequency  $f_0$ .

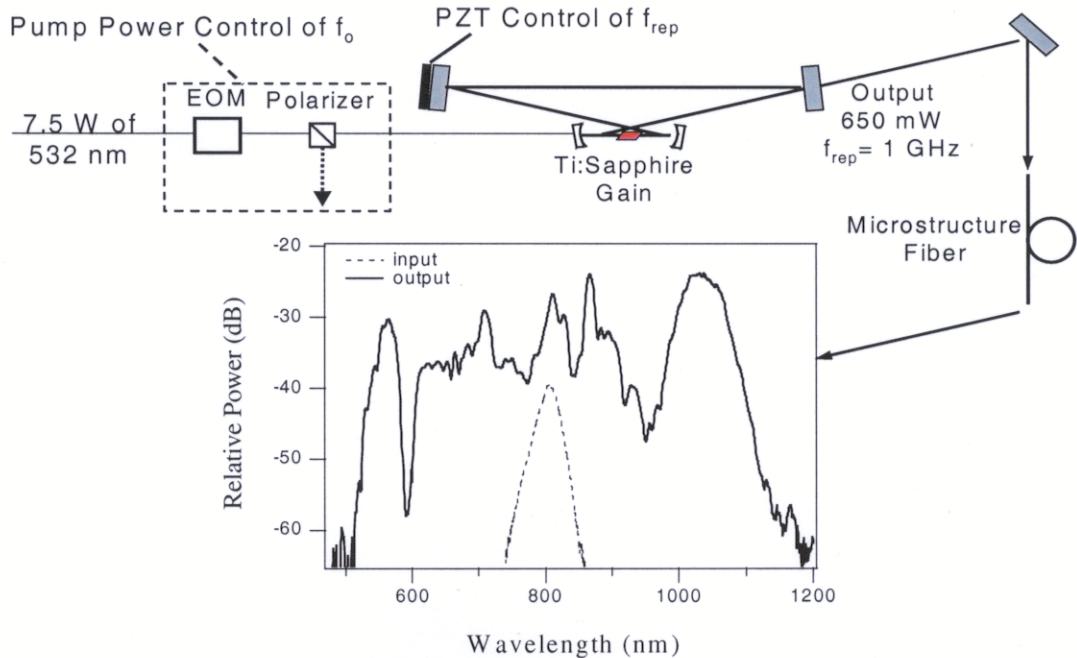


Figure 6. Schematic of Ti:sapphire, mode-locked laser coupled to the microstructure fiber showing the spectral broadening produced by the fiber.

We couple approximately 200 mW of 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 a group-velocity dispersion that vanishes near 770 nm [21] (Figure 6). 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. Control of  $f_r$  is achieved with a piezo-transducer driving a cavity mirror, while  $f_0$  is controlled by adjusting the 532 nm pump beam's intensity with an electro-optic modulator [8]. When both  $f_0$  and  $f_r$  are phase-locked, the frequency of every mode in the comb is known with the same accuracy as that of the optical standard.

An example of an arrangement for an all-optical clock is shown in Figure 7 for the case of the  $\text{Hg}^+$  standard. Some of the light from the optical comb is focused onto a fast photo-diode to generate  $f_r$ , the mode spacing of the comb and the main rf output of the clock. The CW light from the  $\text{Hg}^+$  (563 nm) or Ca (657 nm) standard is transferred to the mode-locked laser system via single-mode optical fibers that are 130 m and 10 m long, respectively. Approximately 2 mW of CW light from the fiber is mode-matched with the appropriate spectral region of the frequency comb to generate a beat signal  $f_b$  with a nearby mode. A phase-locked loop is used to stabilize this beat signal in units of  $f_r$ . The self-referenced technique is then used to generate the offset frequency,  $f_0$ , which is also controlled in units of  $f_r$ . In this way, the output frequency,  $f_r$ , is phase-related to the frequency of the optical standard by the relationship  $f_r = f_{\text{Hg}} / (m + \alpha + \beta)$ , where  $m$  is an integer and  $\alpha$  and  $\beta$  are integer ratios.

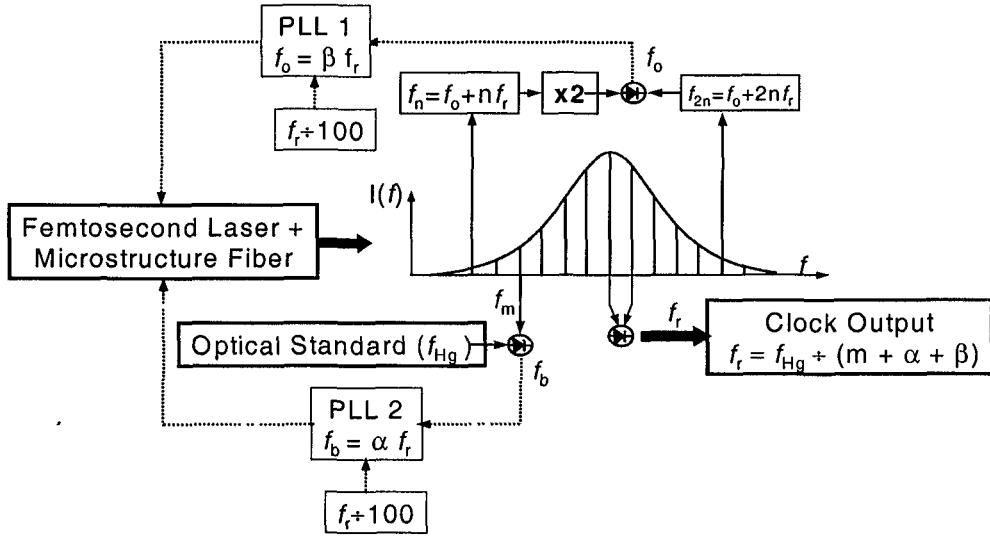


Figure 7. Schematic of an arrangement to use an optical frequency standard in conjunction with the optical comb generator to make an all-optical atomic clock.

## RESULTS

Even at this early state of development, the results from our optical clocks are very exciting. Figure 8 shows the stability between our two optical standards related through an optical comb referenced to one of them. The short-term stability is characterized by a  $\sigma_y(\tau) \approx 7 \times 10^{-15} \tau^{-1/2}$ . These data begin to suggest the tremendous potential for stability available through this technology. The slight loss of stability around 30 s is the result of the fact that one of the optical fibers was not yet phase-corrected for the environment.

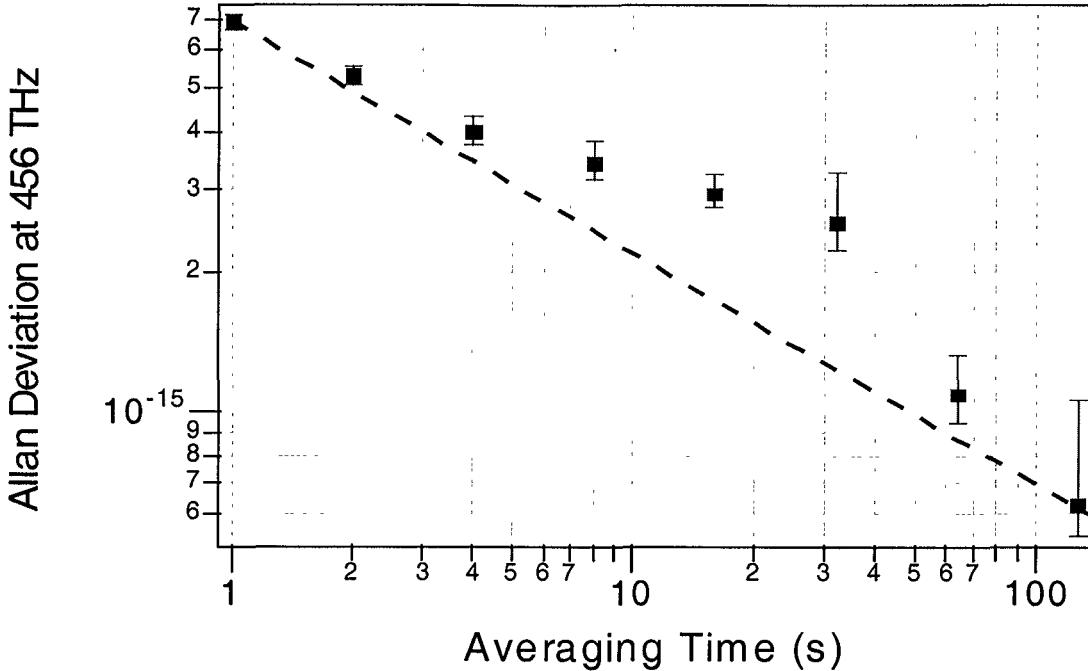


Figure 8. Stability of one of our optical standards measured against an optical comb that is referenced to the other optical standard.

Figure 9 summarizes the frequency measurements of  $\text{Hg}^+$  between August 2000 and February 2001, while Figure 10 summarizes the Ca measurements made from 26 Oct. to 17 Nov. 2000. The frequency of the maser was calibrated by comparing to the local NIST time scale (five hydrogen masers and three commercial cesium clocks), which in turn was calibrated by the local cesium-fountain standard (NIST-F1 [2]). The fractional frequency uncertainty 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  ${}^2D_{5/2}$  state arising from coupling with possible static potentials of the trap. In our spherical Paul trap, where the confinement of the ion uses no applied static fields, the maximum quadrupole shift for an uncontrolled patch charge of 1 V would be less than 1 Hz (or fractional frequency shift  $< 10^{-15}$ ) [23]. 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 as yet 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  $1 \times 10^{-18}$  [10,23].

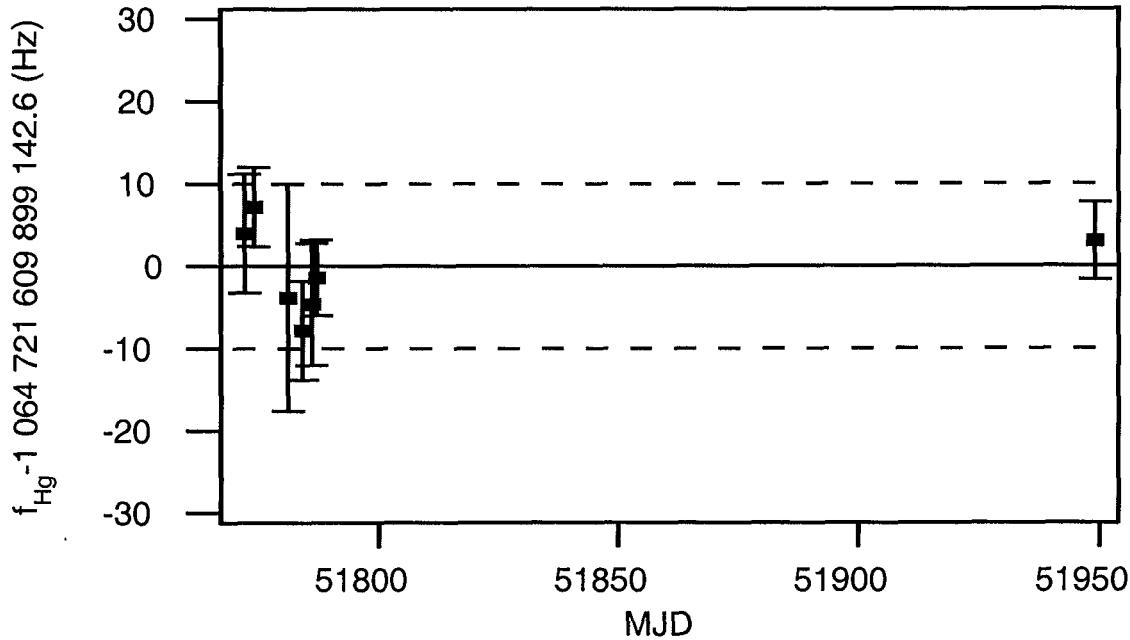


Figure 9. Absolute frequency measurements of our  $\text{Hg}^+$  standard relative to the NIST realization of the SI second. The error bars represent the statistical uncertainty while the dotted lines at  $\pm 10$  Hz represent our estimate of systematic errors.

For the Ca data shown in Figure 10, a correction was applied to the data to account for frequency shift caused by residual phase chirping on the optical Ramsey pulses produced by amplitude modulating an acousto-optic 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 was 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 led to a residual first-order Doppler shift with magnitude  $< 15$  Hz (except on 16 November, 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 10.

Figure 10 also shows the good agreement between our measurement and the most recent value measured with a harmonic-frequency chain [24], which provides a degree of confidence in the reproducibility of the Ca standards. It is also in reasonable agreement with the very recent measurements made by the PTB with a femtosecond comb [25]. An additional measure of the Ca frequency can be made by using the present absolute

measurement of  $Hg^+$  and our earlier measurement of the 76 374 564 455 429(40) Hz gap between  $f_{Hg}$  and the Ca standard [26]. This yields a value of  $f_{Ca} = 455\ 986\ 240\ 494\ 143(40)$  Hz, in good agreement with the value from the present direct measurement.

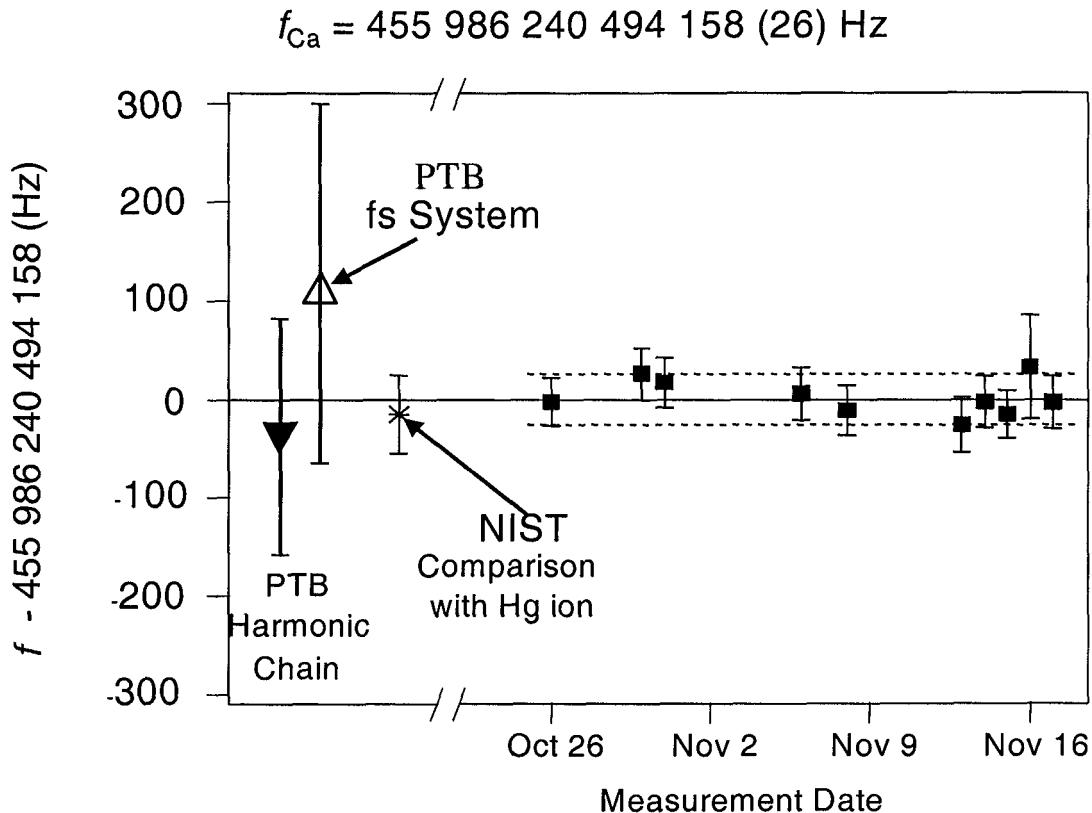


Figure 10. Absolute frequency measurements of our Ca optical standard.

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 [27]. 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 the PTB in 1997 [24] gives  $|(\partial f_{Ca}/\partial t)/f_{Ca}| = 8 \times 10^{-14} \text{ yr}^{-1}$ . Similarly, our measurements on  $f_{Hg}$  from August 2000 to February 2001 provides an initial baseline constraint on the time variation of  $|(\partial f_{Hg}/\partial t)/f_{Hg}| \leq 2 \times 10^{-14} \text{ yr}^{-1}$ . 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.

## ACKNOWLEDGMENTS

We acknowledge the support of the Office of Naval Research. This work was also supported through a Cooperative Research and Development Agreement with Timing Solutions Corp., Boulder, Colorado.

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