ALL-OPTICAL ATOMIC CLOCKS

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Abstract: We have developed two all-optical clocks in which the rf output is generated directly from the optical process within the clockwork. 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 Hg^+ and Ca optical frequency standards with respect to the SI second as realized at NIST. We find the transition frequencies to be f_{Hg} = 1 064 721 609 899 143(10) Hz and f_{Ca} = 455 986 240 494 158(26) Hz, respectively. This work begins to reveal the high stability and potential for accuracy of optical atomic clocks. Furthermore, when combined with previous measurements, we find no time variations of these atomic frequencies within the uncertainties of $|(\partial f_{Hg}/\partial t)/f_{Hg}| = 2 \times 10^{-14}$ yr^{-1} and $|(\partial f_{Ca}/\partial t)/f_{Ca}| = 8 \times 10^{-14}$ yr^{-1}.

Introduction: Optical frequency standards based on laser-cooled atoms and ions promise superior stability and accuracy over existing microwave standards [1–6]. However, because of their high frequencies (~ 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 [7-13]. 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 Hg^+ or 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 [14]. Indeed, for the Hg^+ standard, the uncertainty in the measurement is essentially limited by our best realization of the SI second at ~2×10^{-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 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 Hg^+ and Ca systems have recently been described elsewhere [2,5,15-17], so we summarize only their basic features. The mercury optical frequency standard is based on a single, laser-cooled $^{199}Hg^+$ ion that is stored in a cryogenic, radio-frequency, spherical Paul trap. The ion is laser 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)

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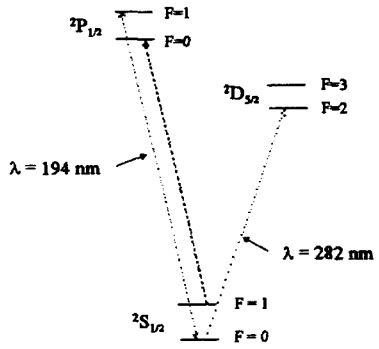

Figure 1. Partial level scheme for ^{199}Hg ion. The 194 nm radiation is used for Doppler cooling, state preparation and detection. The 282 nm transition from the ground state $^2S_{1/2}$ (F = 0, M_F = 0) to the metastable $^2D_{5/2}$ (F = 2, M_F = 0) state is the "clock" transition.

provides the reference for the optical standard [2]. We lock the frequency-doubled output of a 563 nm dye laser with subhertz linewidth [16] 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 [18,19]. 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.

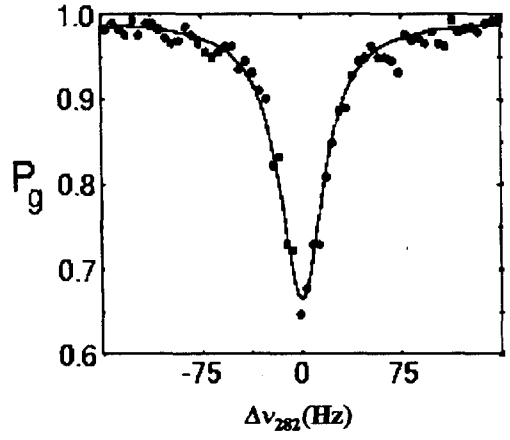

Figure 2. Typical spectrum of the 282 nm "clock" transition.

Most often, the frequency was locked to resonance with a 10 ms interrogation period, which should produce a fractional frequency stability of about $1 \times 10^{-14}\,\tau^{-1/2}$ for an averaging time τ measured in seconds [20].

The calcium standard starts with a collection of ~10^7 laser-cooled ^{40}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 ~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.

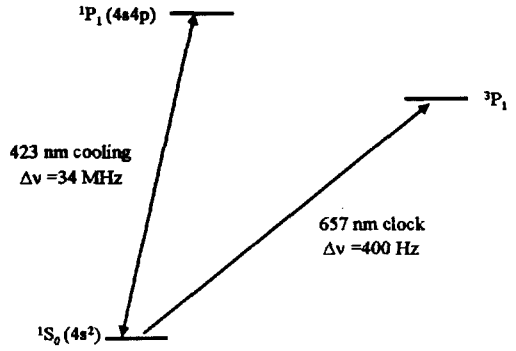

Figure 3. Simplified diagram of the relevant energy levels in the Ca standard.

We excite the transition with a four-pulse Bordé-Ramsey sequence (pulse duration =1.5 μs) with light from a continuous-wave (CW), frequency-stabilized diode laser. Using a shelving detection technique similar to that employed in the Hg^+ system, near-resonant 423 nm pulses (5 μs 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.

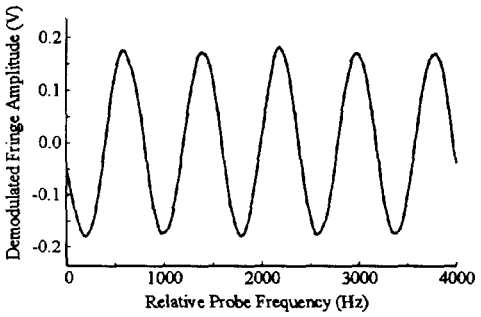

Figure 4. Optical Bordé-Ramsey fringes with a 960 Hz (FWHM) resolution. The total averaging time to generate this figure was 60 s.

This system has demonstrated a fractional frequency stability of $4 \times 10^{-15}\ \tau^{-1/2}$ when probing sub-kilohertz linewidths [5]. 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 [7-13]. 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^{th} mode of the comb is given by $f_n = nf_r + f_0$ [21, 22], 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. f_r can be measured by direct detection of the laser's output with a photodiode. f_o is measured by heterodyning the doubled frequency of mode $f_n = nf_r + f_0$ from the infrared wing of the comb with mode $f_{2n} = 2nf_r + f_0$ from the blue side of the comb as shown in figure 5. This self-referenced technique [11,12] 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 [23], launching the longer pulses

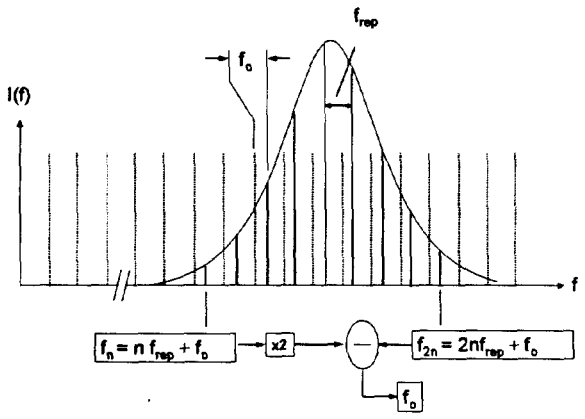

Figure 5. Schematic of the "self referencing" technique to generate a signal at the offset frequency f_0.

from a commercially-available femtosecond laser into an air-silica microstructure fiber [24,25] 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} [12].

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 μm core and group-velocity dispersion that vanishes near 770 nm [24]; Fig. 6.

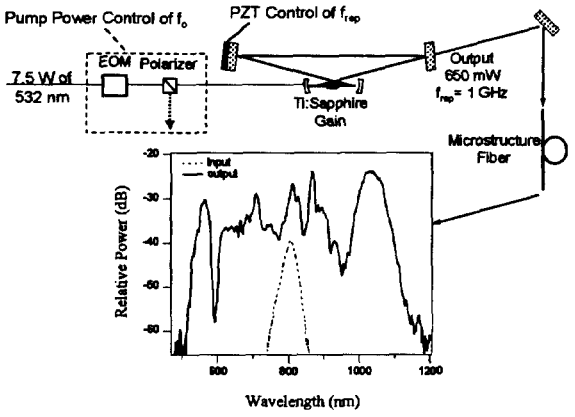

Figure 6. Schematic of Ti:Sapphire, mode-locked laser coupled to the microstructure fiber showing the spectral braodening produced by the fiber.

This power density is sufficient to increase the spectral width of the laser from 13 THz to more than 300 THz, spanning from ~520 nm to ~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 [12]. 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 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.

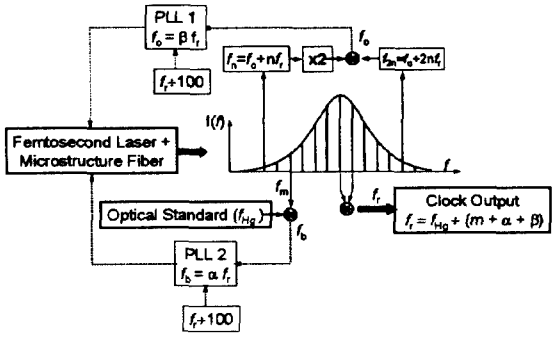

Figure 7. Schematic of an arrangement to use an optical frequency standard in conjunction with the optical comb generator to make an all-optical atomic clock.

The CW light from the 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 $f_r = f_{Hg}/ (m + \alpha + \beta)$, where m is an integer and α and β are integer ratios.

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

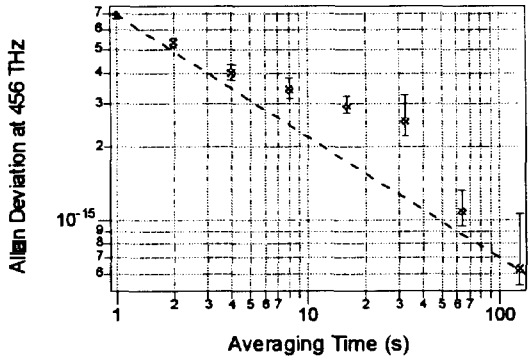

Figure 8. Stability of one of our optical standards measured against an optical comb which is referenced to the other optical standard.

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 phase corrected for the environment.

Figure 9 summarizes the frequency measurements of Hg^+ between August 2000 and February 2001 while figure 10 summarizes the Ca measurements made from Oct. 26 to Nov. 17, 2000. The frequency of the maser was calibrated by comparing to the local NIST time scale (5 hydrogen masers and 3 commercial cesium clocks), which in turn was calibrated by the local cesium fountain standard (NIST-F1 [14]). The fractional frequency uncertainty of the reference maser

relative to the SI second was about 1.8×10^{-15} for these measurements.

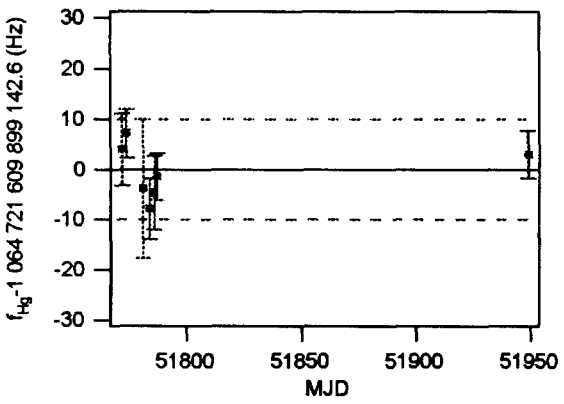

Figure 9. Absolute frequency measurements of our Hg^+ standard relative to the NIST realization of the SI second. The error bars represent the statistical uncertainty while the dotted lines at +/- 10 Hz represent our estimate of systematic errors.

The weighted mean of our measurements of the Hg^+ clock transition is f_{Hg} =1 064 721 609 899 143 Hz. The statistical uncertainty ($\approx 4\times10^{-15}$) of our limited duration measurements is essentially the result of the reference-maser's short-term stability ($\sim2\times10^{-13}\tau^{-1/2}$). Because we have not made a full experimental evaluation of the 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 1v would be less than 1 Hz (or fractional frequency shift $<10^{-15}$) [26]. 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 Hg^+ system can be reduced to values approaching 1×10^{-18} [2,26].

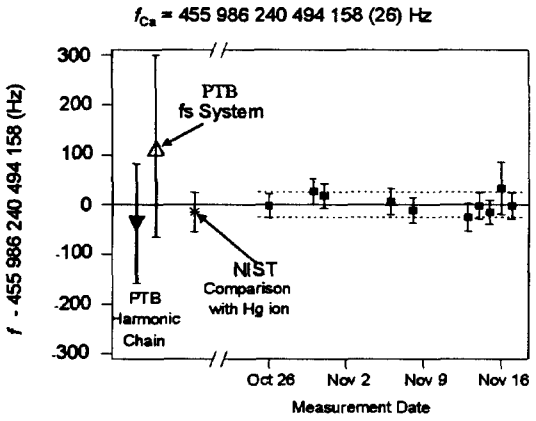

Figure 10. Absolute frequency measurements of our Ca optical standard.

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 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 ~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 three days of measurements, we were able to reduce this shift by a factor of ~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 November 16, where a large drift velocity led to an uncertainty of ~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 ~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 [27], 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 [28]. 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 [29]. 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.

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 [30]. 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 [27] 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.

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