Single-Atom Optical Clock with High Accuracy

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For the past 50 years, atomic standards based on the frequency of the cesium ground-state hyperfine transition have been the most accurate time pieces in the world. We now report a comparison between the cesium fountain standard NIST-F1, which has been evaluated with an inaccuracy of about 4×10^{-16}, and an optical frequency standard based on an ultraviolet transition in a single, laser-cooled mercury ion for which the fractional systematic frequency uncertainty was below 7.2×10^{-17}. The absolute frequency of the transition was measured versus cesium to be 1 064 721 609 899 144.94 (97) Hz, with a statistically limited total fractional uncertainty of 9.1×10^{-16}, the most accurate absolute measurement of an optical frequency to date.

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Since the development of the first continuously operating cesium frequency standards in 1955 [1], the accuracy attainable with atomic clocks based on cesium has steadily increased. Now, a few of the recently developed cesium fountain frequency standards [2] have reported fractional frequency uncertainties below 10^{-15}. Even so, for more than 30 years it has been recognized that a frequency standard based on an optical transition in a trapped single ion [3] has the potential to reach a systematic fractional frequency uncertainty approaching 10^{-18}. Recently, direct measurement of optical frequencies has been made practical by the development of the femtosecond laser frequency comb [4,5]. This, combined with narrow linewidth lasers [6] has made possible the first generation of optical atomic frequency standards and clocks [7], based on both neutral atoms and trapped ions. Recent measurements [8–12] in single-ion clock systems based on Sr^+, Yb^+, and Hg^+ show notable progress toward the understanding and control of systematic frequency errors, such as the quadrupole shift [13], and pave the way for optical standards to begin to realize their long-anticipated accuracy. Here, we report a measurement of the absolute frequency of the Hg^+ optical clock versus the NIST-F1 cesium fountain standard, where the uncertainties of the systematic shifts of the optical standard are now smaller than those of the best cesium primary standards.

The mercury ion optical clock [14,15] has been described previously, and only a few of the central features are outlined here. A frequency-stabilized laser [6] at 563 nm is doubled and servo-locked to the 282 nm $5d^{10}6s\,^2S_{1/2}(F=0) \leftrightarrow 5d^9 6s^2\,^2D_{5/2}(F=2, m_F=0)$ electric-quadrupole ("clock") transition in a single laser-cooled $^{199}Hg^+$ ion, which is held in a cryogenic rf (Paul) trap, consisting of a ring electrode driven with rf, and two end cap electrodes held near dc ground. The strongly allowed $5d^{10}6s\,^2S_{1/2}(F=1) \leftrightarrow 5d^{10}6p\,^2P_{1/2}(F=0)$ transition at 194 nm is used for laser cooling, state preparation, and detection of the clock state via quantum jump spectroscopy [3] subsequent to each probe of the clock transition.

Earlier measurements of the Hg^+ optical clock transition frequency were systematically and statistically limited by the uncertainties of various perturbations to about ± 10 Hz, or fractionally 10^{-14}. We have now implemented several improvements that significantly reduce the systematic uncertainties. We first placed the standard within a single-layer magnetic shield and, with three pairs of computer-controlled coils, applied a small (8 μT) magnetic bias field. This produced a quadratic Zeeman shift of only -1.2 Hz, which reduced error due to uncertainty of the second-order Zeeman coefficient to below 5 mHz. We also constrained the uncertainty due to slow changes in $|B|$ by periodically measuring the frequency of the $5d^{10}6s\,^2S_{1/2}(F=0) \leftrightarrow 5d^9 6s^2\,^2D_{5/2}(F=2, m_F=2)$ transition, which has a linear field sensitivity of $\sim$39 kHz/μT. The largest field drifts observed were of order 60 nT, which leads to an uncertainty of ± 20 mHz. Secondly, the quadrupole shift was effectively eliminated by averaging the optical clock frequencies that were measured for three orthogonal orientations [13] of the 8 μT bias field. We operated the clock sequentially for these axes, typically spending 90 s for each orientation. The monitoring of $|B|$ ensured that the field was near its nominal value for each orientation, and that slow field drifts did not significantly change the orientation of the quantization axes. The amplitude of the clock transition, which depends upon the direction and polarization of the 282 nm probe light with respect to the quantization axis, was essentially equalized for the three field orientations by rotating the polarization of the probe light by use of a half-wave plate on a motorized rotation stage.

In the absence of a strong magnetic bias field, cooling the ion with 194 nm radiation from a single arbitrarily polarized laser beam quickly pumps the ion into a dark state [16]. The dark state can be avoided by rapidly scrambling the polarization of the light field at the site of the ion, for example, by radiating the ion with three noncollinear beams with disparate frequencies. The three radiation components can be generated by using an acousto-optic modulator (AOM) that is driven with rf at three distinct frequencies. A crucial benefit of this arrangement is that it allows us to continually monitor and correct possible excess micromotion (motion at the rf trap frequency) along the three beam axes by use of a fluorescence modulation technique [17].

Table I presents an uncertainty budget for the Hg^+ optical frequency standard during its comparison with NIST-F1, showing the dominant sources of uncertainty. We correct for two frequency biases: the second-order Zeeman shift from the 8 μT bias field, and the gravitational redshift due to the altitude difference (about 4.5 m) between the mercury trap and NIST-F1.

By examining frequency differences between data taken at the different field axes, typical quadrupole shifts in the rf trap have been constrained to be 0.5 Hz or smaller. The error in the alignment of the magnetic fields [11] is less than $\pm 10°$, so averaging over the three orientations should constrain the total quadrupole shift to below 10 mHz. However, since we did not always spend equivalent time at each axis, the quadrupole shift is not averaged to zero for about 8% of our data, which leads to a fractional uncertainty of 5×10^{-17} due to the quadrupole shift. This uncertainty could be reduced by trimming the unbalanced portion of the data, but such trimming is currently unwarranted since the comparison with cesium is statistically limited at a higher level.

One important uncertainty is the magnitude of the quadratic Zeeman shift caused by ac magnetic fields at the drive frequency of the trap, near 12 MHz. The ac magnetic field at the site of the ion vanishes if the trap structure is physically and electrically symmetric. That is, if the ring electrode is centered between the end cap electrodes, their supporting rods are equally spaced and parallel, and if the capacitance of the rods and that of the trap is uniformly distributed. Because we have not yet measured the field at the site of the ion, we make a conservative estimate of its amplitude. The total capacitance of the trap structure, including the trap and the support rods, is calculated and measured to be approximately 1.5 pF. The support rods contribute the majority of the capacitance, while the ring and two end caps contribute less than 30 fF. The maximum amplitude of the oscillating electric potential applied to the ring electrode is 1150 V. Conservatively, if we assume that 75% of the total rf current flows through one end cap and supporting rod, then the maximum rms ac field at the site of the ion is less than 5×10^{-7} T. Hence, the maximum frequency shift from the ac field is less than 20 mHz, a fractional shift below 2×10^{-17}. We plan to measure the ac field in the future via microwave spectroscopy on the 40.5 GHz Hg^+ ground-state hyperfine transition, which is known to high accuracy [18].

TABLE I. Leading contributions to the systematic uncertainty budget of the Hg^+ optical clock, valid for the recent measurements of the absolute frequency versus NIST-F1. Corrections applied to the frequency of the mercury transition are shown along with the fractional frequency uncertainty due to each physical effect.

Physical Effect	Correction (Hz)	Fractional Uncertainty $\times 10^{-17}$
Quadrupole shift	0	5
Servo error	0	3
Zeeman shift (ac, trap)	0	2
Zeeman shift (dc)	1.203	2
Micromotion (Doppler, Stark)	0	2
Gravitation	0.524	1
Thermal motion (Doppler)	0	1
Other effects	0	2
Total	1.727	7.2

A digital servo system is used to steer the frequency of the 282 nm radiation source to that of the atomic transition, with a loop time constant near 10 s. Frequency bias due to nonoptimal steering, known as servo error, is dominated by incorrect anticipation of the drift rate of the reference optical cavity [6]. The conservative uncertainty reported for this comparison (± 30 mHz) is based on an examination of the servo record. Uncertainties due to second-order Doppler and ac Stark shifts from excess micromotion were constrained to contribute less than ± 20 mHz by examination of the fluorescence modulation data [17]. Thermal (secular) motion in the trap leads to an additional Doppler uncertainty of ± 10 mHz. There are a number of other potential sources of frequency errors ("Other effects" in Table I) that contribute, in aggregate, less than 2×10^{-17} to the fractional frequency uncertainty. These include, for example, ac Zeeman shift caused by possible line pickup at 60 Hz; Stark shifts due to light at 282, 563, and 194 nm, blackbody radiation, or the rf trapping field; Dick effect; acousto-optic modulator frequency chirp; uncertainty in the Zeeman coefficient; and high-order and ac quadrupole shifts.

The combined systematic fractional frequency uncertainty for the mercury system is less than 7.2×10^{-17}, which is smaller than that reported for NIST-F1. Straightforward improvements in several areas should reduce the fractional uncertainty to several times 10^{-18}, limited by the second-order Doppler shift due to residual micromotion and secular motion.

The NIST-F1 cesium fountain is the U.S. national frequency standard, and its uncertainty is among the lowest of any of the standards reporting to the BIPM. Its construction, operation, and accuracy have been thoroughly described [19–21]. In essential terms, a cloud of about 10^7 cesium-133 atoms is laser cooled to about 500 nK, launched vertically at a velocity of about 4 m/s, and prepared so that only about 10^6 atoms in the $|F, m_F\rangle = |3, 0\rangle$ state are present when the cloud reaches the Ramsey cavity. The Ramsey cavity, a microwave cavity resonant on the 9.2 GHz transition, subjects the atoms to a microwave pulse as they transit the cavity, preparing them in a coherent superposition of the $|4, 0\rangle$ and $|3, 0\rangle$ states. The atoms continue upward, decelerating continuously under the influence of gravity, stop, and accelerate downwards, nominally returning along the same path. Hence, the atoms transit the cavity twice with a time separation of about 0.5 s, and if the microwave frequency is exactly on resonance, the superposition evolves into the $|4, 0\rangle$ state. The average frequency of the microwaves is locked to the atomic resonance by alternately probing either side of the resonance and steering the frequency to balance the transition probability on the two sides of the resonance.

Cesium fountains, such as NIST-F1, have a number of systematic frequency shifts that must be corrected in order to accurately realize the SI definition of the second. In NIST-F1, the dominant uncertainties are a result of a spin-exchange frequency shift caused by very low energy collisions among the cesium atoms and by the blackbody shift. For the density and temperature during this comparison with the mercury standard, the fractional frequency uncertainty from spin-exchange was $\delta f/f = 2.7 \times 10^{-16}$, while that from the blackbody shift was 2.6×10^{-16}. Various other frequency shifts contribute, at a smaller level, to raise the combined systematic fractional frequency uncertainty of NIST-F1 during these comparisons to 4.1×10^{-16}, the quadrature sum of all the individual uncertainties in the frequency biases.

The comparison between mercury and cesium is mediated by a femtosecond laser frequency comb (FLFC) based on a Kerr-lens mode locked Ti:sapphire laser that operates at a 1 GHz repetition rate [15,22,23]. The FLFC acts as a divider that converts the optical stability and accuracy of the mercury clock transition to a stable microwave frequency that can be compared to cesium via a transfer hydrogen maser [14].

The FLFC is characterized by two frequency parameters, the laser carrier-envelope offset frequency, f_O, and the laser repetition rate, f_{rep}. The resultant frequency of the nth optical line in the comb is thus expressed as $f_n = f_O + nf_{rep}$. The offset frequency is measured in the heterodyne beat between the second and third harmonics of separated portions of the Ti:sapphire spectrum [23]. Once measured, f_O is phase locked to a stable rf reference frequency. The frequency f_{rep} is stabilized by phase locking the Nth mode of the FLFC to the Hg^+ clock laser [14]. The Hg-stabilized f_{rep} is then compared to NIST-F1 via the intermediate hydrogen maser. Microwave signals at 1 and 9.2 GHz derived from the same maser are simultaneously compared in two channels (using standard frequency synthesis, mixing and counting techniques) to f_{rep} and the cesium atomic transition in NIST-F1, respectively. Subtraction of the two data records eliminates the long-term fluctuations of the maser in the comparison of the Hg^+ and Cs standards.

There are several possible sources of frequency uncertainty in this optical-to-microwave comparison. The most significant are in the microwave domain due to temperature-driven fluctuations in the cable carrying the maser signal and the synthesis of the 1 GHz signal, which result in a fractional uncertainty of 2×10^{-16}. Sources of bias in the FLFC as employed here have been studied in detail [24], resulting in an upper fractional uncertainty limit of 1×10^{-16} that arises from excess noise in the photodetection of f_{rep}. Counting and possible cycle-slip errors are at a much lower level, yielding a systematic fractional uncertainty due to the comparison of 2.3×10^{-16}.

A histogram and stability plot of the data collected versus cesium are shown in Fig. 1. These data were taken over seven days between June 15, 2005 and July 1, 2005. The arithmetic mean frequency of the mercury standard for these runs was 1 064 721 609 899 144.98 Hz. The total deviation [25] (similar to the Allan deviation, but a better predictor of long-term fractional frequency instability) is computed from 13 612 frequency readings, each with a 10 s gate time. The values at longer times are computed by averaging the appropriate number of 10 s intervals. The fit to the total deviation is $3.38 \times 10^{-13}/\sqrt{\tau}$, where τ is the averaging time in seconds. The time series of frequency readings has nonuniformly distributed dead time. However, the slope of the total deviation plot, along with the Gaussian distribution in the inset, indicate a white frequency noise process. Consequently, the presence of nonuniform dead time does not cause a problem. The instability of the mercury standard is low enough [14] that the instability of the comparison is dominated by that of the cesium fountain. Extrapolating to the full length of the data set gives the statistical measurement uncertainty of 0.97 Hz.

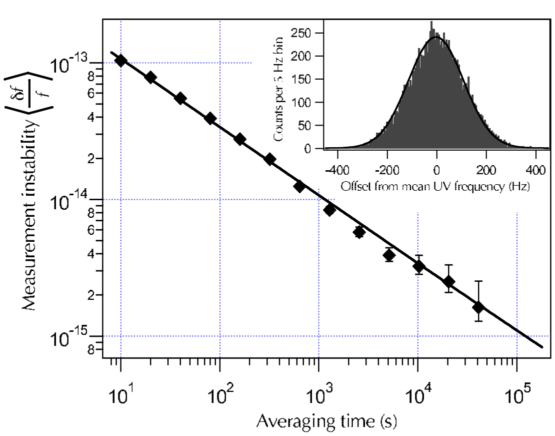

FIG. 1 (color online). Fractional frequency instability, as represented by the total deviation, for the comparison of the mercury and cesium standards. The data set consists of 13 612 frequency readings, each taken with 10 s gate time. A histogram of the frequency values (inset) is shown with a Gaussian fit.

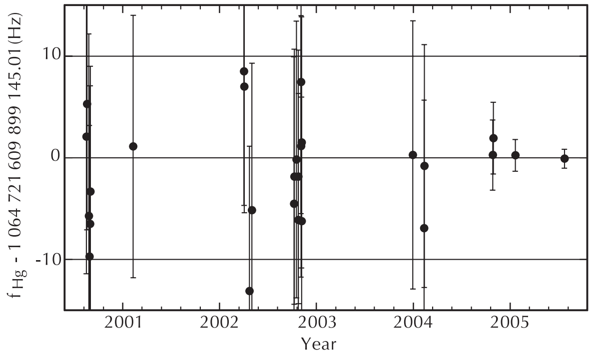

FIG. 2. Record of all absolute frequency measurements of the mercury optical clock transition frequency. The data are plotted as deviations from their weighted mean, 1 064 721 609 899 145.01 Hz. The error bars represent 1 standard deviation of total statistical and systematic uncertainty. The last point in this data set, taken in June 2005, corresponds to the measurement described in this Letter.

During the course of the multi-day frequency comparison of the two standards, NIST-F1 was offline for approximately 1 d. Nevertheless, a set of data points was taken over this time interval versus the same maser used for each of the measurement sets. The frequency of the maser was calibrated versus NIST-F1 immediately before and after this data set. The average Hg^+ frequency for this data set was $f_{Hg} = 1064[\ldots]144.82(1.60)$ Hz. Taking the weighted mean of this set and that taken directly versus cesium yields the final frequency $f_{Hg} = 1064[\ldots]144.94(82)$ Hz, where the uncertainty is purely statistical. Adding in quadrature the statistical uncertainty and the systematic uncertainty due to cesium (4.1×10^{-16}), Hg^+ (7.2×10^{-17}), and the comparison (2.3×10^{-16}) yields a fractional frequency uncertainty of 9.1×10^{-16}, for a final result of $f_{Hg} = 1\,064\,721\,609\,899\,144.94(97)$ Hz.

This comparison between mercury and cesium frequency standards represents the most accurate absolute measurement of an optical frequency to date and is made possible by the high accuracy of the two standards. Recent precision optical frequency measurements reported by other laboratories include the measurement of the Sr^+ optical clock transition [8] with a fractional uncertainty of 3.4×10^{-15}, and a preliminary measurement of the Yb^+ clock transition at 3.0×10^{-15} [26].

The record of all absolute frequency measurements of the mercury optical clock frequency f_{Hg} is shown in Fig. 2. The smaller uncertainties of the last few measurements are partially due to (a) a better evaluation of the systematic frequency shifts of the Hg^+ optical clock that are described in this paper and (b) the reduction in the maser frequency uncertainty that is gained by running the Hg^+ and Cs standards simultaneously. The most recent measurement, described in this Letter, will serve as an accurate baseline for improved tests of the stability of fundamental constants [15,27].

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