A MERCURY-ION OPTICAL CLOCK*

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We have developed an optical clock based on a laser whose frequency is locked to a single, laser-cooled $^{199}Hg^+$ ion and that uses a femtosecond laser and microstructure fiber to phase-coherently divide the optical frequency to a countable microwave frequency. The measured short-term stability in the optical domain is about an order of magnitude higher than the best cesium-fountain clock. The estimated value of the electric-quadrupole frequency shift of the $^2S_{1/2}$ (F = 0, m_F = 0) - $^2D_{5/2}$ (F = 2, m_F = 0) clock transition is given.

1. Introduction

An important measure of the performance of an atomic clock is the frequency stability of its locked reference oscillator. For a given linewidth and atom number, the stability of an oscillator that is steered to resonance with an atomic transition is directly proportional to the frequency of the oscillator. Hence, a clock operating at a frequency in the optical region of the electromagnetic spectrum ($\approx 10^{15}$ Hz) could be many orders of magnitude more stable than clocks operating at a frequency in the microwave region of the spectrum ($\approx 10^{10}$ Hz). In fact, it is possible to exchange some of the potential gain in stability for higher accuracy by using only a small number of atoms that are confined to a small volume in space. However, only recently has there been a practical device that is capable of faithfully "counting" each cycle of the optical radiation in order to generate time. In these Proceedings (see also the contributions by E. Curtis *et al.* and S. Diddams *et al.*), we report on research at NIST toward the realization of highly stable and accurate optical timepieces [1] that use a femtosecond laser and a microstructure fiber as the "clockwork" to phase-coherently divide down the optical frequency to a countable microwave frequency [2-9]. Here, we concentrate on the description of the mercury optical standard, where the frequency of an optical oscillator is short-term stabilized by locking to the resonance of a high-finesse cavity [10] and long-term stabilized by locking its harmonic to the narrow resonance of the $^2S_{1/2}$ (F = 0, m_F = 0) - $^2D_{5/2}$ (F =

2, $m_F = 0$) electric-quadrupole-allowed transition (282 nm or 1.064 PHz) of a single, laser-cooled $^{199}Hg^+$ ion [11]. This system has demonstrated better stability (and anticipates higher accuracy) than that of the microwave cesium-fountain standards [1,11]. We describe its present and anticipated performance, and briefly present its main limitation.

2. Stability of atomic clocks

The Allan deviation $\sigma_y(\tau)$ provides a convenient measure of the fractional frequency instability of a clock as a function of averaging time τ [12]. For an oscillator locked to an atomic transition of frequency f_0 and linewidth Δf,

$$\sigma_y(\tau) = \left\langle \frac{\Delta f_{rms}}{f_0} \right\rangle_\tau \approx \frac{\Delta f}{\pi f_0}\sqrt{\frac{T}{N\tau}} \quad , \tag{1}$$

where Δf_{rms} is the measured frequency fluctuation, N is the number of atoms, and T is the cycle time (the time required to make a single determination of the line center) with $\tau > T$. This expression assumes that technical noise is reduced to a sufficiently small level such that the quantum-mechanical atomic projection noise is the dominant stability limit [13,14] and that the signal contrast is 100 %. If we further assume that the signal linewidth $\Delta f = 1/2T$ (i.e., Ramsey interrogation and zero dead time), then [15]

$$\sigma_y(\tau) \approx \frac{1}{2\pi f_0}\sqrt{\frac{1}{NT\tau}} \; . \tag{2}$$

In both cases, the fractional frequency instability increases if the signal contrast is less than 100 %, which could be caused, for example, by technical noise that is comparable to, or greater than the quantum projection noise, or a laser linewidth that exceeds the inverse probe time. The contrast can also be degraded if the ion is not strongly confined in the Lamb-Dicke limit where the average quantum-occupation number $\langle n \rangle \leq 1$ [16]. If $\langle n \rangle > 1$, then fluctuations in n from measurement cycle to measurement cycle can cause variations in the transition probability of the ion [17], which reduce the signal contrast.

The signal contrast and clock stability is also fundamentally diminished as the probe period approaches the natural lifetime τ_D of the metastable state. In this case, the signal strength decreases because the ion can decay during the probe and detection period. A quantitative treatment that includes decay shows that the optimum probe time to minimize $\sigma_y(\tau)$ for Ramsey excitation is nominally the same as the metastable lifetime [18]. For this case, $\sigma_y(\tau)$ is degraded by nearly a factor 2 compared to the case of an equivalent probe time without atomic decay. Thus, assuming zero dead time and <n> < 1, the optimum stability is approximately given by

$$\sigma_y(\tau) \approx \frac{1}{\pi f_0}\sqrt{\frac{1}{NT\tau}} \quad , \tag{3}$$

where $T = \tau_D$.

3. The optical frequency standard

Our optical frequency standard is based on a single, nearly motionless and unperturbed $^{199}Hg^+$ ion that is confined in a cryogenic, spherical Paul trap [8]. The ion is cooled, state-prepared and detected by light that is scattered on the strongly allowed $^2S_{1/2}$ - $^2P_{1/2}$ transition at 194 nm. The $^2S_{1/2}$ ($F = 0$, $m_F = 0$) - $^2D_{5/2}$ ($F = 2$, $m_F = 0$) electric-quadrupole allowed transition at 282 nm ($\tau_D \approx 0.09$ s) provides the reference for the optical standard (Fig.1). The natural linewidth of the S-D resonance is about 2 Hz at 1.064 PHz, and recently a Fourier-transform-limited linewidth of only 6.7 Hz (Q=1.5×10^{14}) was observed [11]. We lock the frequency-doubled output of a narrowband and stable 563 nm dye laser [10] to the narrow S-D resonance by the method of electron shelving, whereby each transition to the metastable D-state is detected by the suppression of the scattering of many 194 nm photons on the strongly allowed S-P transition [19,20]. The short-term (1-10 s) fractional frequency instability of the probe laser is ≤ 5×10^{-16}, which matches well to the best stability that is predicted by the shot-noise-limited detection of the S-D transition of a single mercury ion. From Eq. 3, the lowest fractional frequency instability of the single-ion mercury standard is expected to be about 1×10^{-15} $\tau^{-1/2}$ with a fractional frequency uncertainty that approaches 10^{-18} [11]. However, in our present realization of the Hg optical standard, the stability obtained from locking to the ion was degraded primarily because the signal contrast was less than 100 % (see the spectra in Fig. 1), the probe time was only 20 ms, and there was substantial dead time between probes of the S-D resonance. Hence, $\sigma_y(\tau) \leq 2\times10^{-15}$ for averaging times up to ~ 30 s (where the laser stability is dominated by the cavity), at which point $\sigma_y(\tau)$ began to average down as $\tau^{-1/2}$ (as the long-term stability was transferred to the ion) [1,11].

In our next version of the mercury optical standard, we anticipate increasing the signal contrast by cooling the ion to the zero-point energy of the confining potential either with sideband cooling [16] or electromagnetically-induced-transparency (EIT) cooling [21]. We also will minimize the dead time by more rapidly pumping (≈ 1 μs) into the $^2S_{1/2}$, F = 0 hyperfine state prior to each probe of the clock transition and by including a short pulse (≈ 1 ms) of radiation at 398 nm connecting the $^2D_{5/2}$ ($F = 2$, $m_F = 0$) state to the $^2P_{3/2}$ ($F = 3$) manifold with

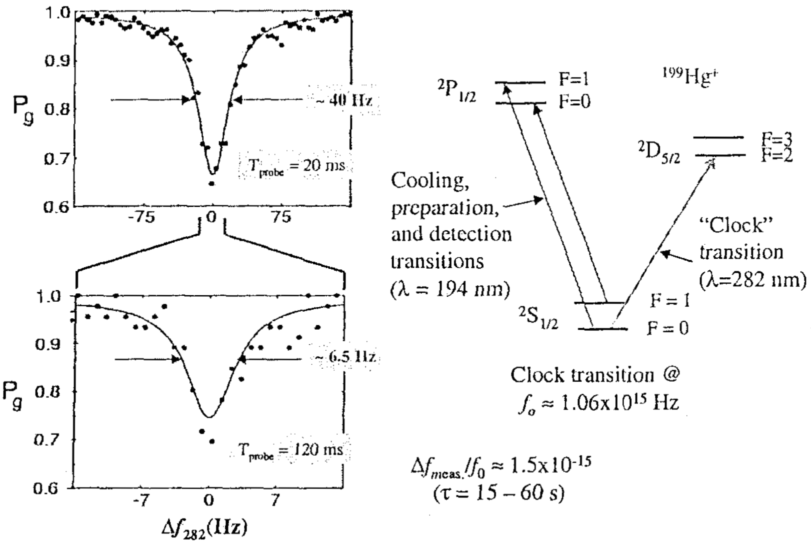

Figure 1. Quantum-jump absorption spectra of the $^2S_{1/2}(F=0) \leftrightarrow {}^2D_{5/2}(F=2)$ $\Delta m_F = 0$ electric-quadrupole transition and a partial energy-level diagram of $^{199}Hg^+$ with the relevant transitions indicated. Δf_{282} is the frequency detuning of the 282 nm probe laser, and P_g is the probability of finding the atom in the ground state. The upper spectrum is obtained with a Rabi excitation pulse 20 ms long (averaged over 292 sweeps) and the lower spectrum corresponds to an excitation pulse 120 ms long (averaged over 46 sweeps). The linewidths are consistent with the Fourier-transform limit of the respective pulse times [10].

circularly polarized light to clear out the $^2D_{5/2}$ state after each measurement cycle [16]. In our present configuration, we wait for the ion to return to the ground state by spontaneous decay from the metastable D-state. Additionally, we currently probe the ion's return to the ground state by looking for 194 nm light that is scattered on the S-P transition. By sweeping out the D-state population after each measurement cycle, this step can be eliminated. Hence, we expect a dead time per measurement cycle < 5 ms (dominated by the state detection period). With these improvements, we expect to reach the fractional frequency instability $\sigma_y(\tau) \approx 1\times10^{-15}\tau^{-1/2}$.

4. The frequency divider

The Hg^+ standard provides high stability and, potentially, high accuracy, but to realize a countable clock output we must phase-coherently convert the optical signal to a lower frequency. The clockwork that divides the 1.064 PHz optical frequency to a countable microwave frequency f_r is discussed in more detail in these Proceedings (see S. Diddams *et al.*) and in Ref. 1. Briefly, our clockwork is based on a femtosecond laser and a microstructure optical fiber [22,23]. The Ti:sapphire femtosecond ring laser emits a train of pulses at the nominal repetition rate of f_r = 1 GHz [24]. The frequency-domain spectrum of the pulse train is a uniform comb of phase-coherent continuous waves separated by f_r. The frequency of the n^{th} mode of this comb is $f_n = nf_r + f_o$ [25,26], where f_o is the frequency offset common to all modes that results from the difference between the group and phase velocities inside the laser cavity [27]. The frequencies f_o and f_r can be linked to the 563 nm laser oscillator such that every element of the femtosecond comb, as well as their frequency separation f_r, is phase-coherent with the laser locked to the Hg^+ standard. Thus, with no other frequency references as an input, we realize all aspects of a high-stability, optical atomic clock: a stable laser that is locked to a narrow atomic reference, and whose frequency is phase-coherently divided down to give a microwave output that can be recorded with a counter [1].

5. High-stability clock output

The stability of the 532 THz laser should be transferred to each element of the femtosecond comb, and, thus, to the inter-comb spacing f_r. We have compared the stability of f_r to the output of a synthesizer that is referenced to a hydrogen maser for which $\sigma_y(1\text{s}) \approx 2.5\times10^{-13}$ and demonstrated that the 1-s instability of f_r is no worse than that of the hydrogen maser [1]. Before we can claim that a microwave signal with stability matching that of the optical standard can be obtained from the optical clock, f_r needs to be compared to an oscillator with stability significantly better than that of the hydrogen maser. Nonetheless, we can measure the stability of the comb in the optical domain, and thereby infer the expected stability of f_r, by comparing one element of the optical comb to a second optical standard with high stability. For example, we detected and counted the heterodyne beat signal between a single element of the comb at 456 THz and a frequency-stabilized diode laser locked to the ${}^1S_0$ - ${}^3P_1$ intercombination transition of a laser-cooled ensemble of calcium atoms [28,29, and see E. Curtis *et al.* in these Proceedings]. The Allan deviation of the heterodyne signal between the Hg^+ stabilized comb and the Ca-stabilized optical standard averages down roughly as $9\times10^{-15}\ \tau^{-1/2}$, which is consistent with the expected fractional frequency instability of the Ca standard in its present configuration. For $\tau > 30$ s the instability of the Hg^+ standard in its present configuration contributes to the measured instability at approximately the same level as the Ca standard. Hence, the measured fractional frequency instability reaches about 1.5×10^{-15} at 100 s [1].

It should be noted that for $10 < \tau < 30$ s frequency and phase fluctuations introduced by the 180 m long optical fiber that transmits the 532 THz light to the femtosecond system partially degraded the stability observed between the Ca standard and the Hg-referenced comb. We therefore implemented active cancellation [30,31] of this fiber noise and additionally further improved the signal-to-noise in the Ca standard. Data taken under these conditions reveal a fractional frequency instability of 7×10^{-15} at $\tau = 1$ s [1]. Similar stability in the ~1 GHz clock output remains to be verified.

Finally, when f_r and f_0 are detected and counted with respect to the frequency of the hydrogen maser (which acts as a transfer standard to the NIST realization of the SI second [32]), an absolute measurement of the $^{199}Hg^+$ clock transition can be made [33]; f_{Hg} = 1 064 721 609 899 143 (10) Hz. The statistical uncertainty of our measurements is about +/- 2 Hz, which is due in part to the fractional frequency instability of the maser at our measurement times and in part by the accuracy determination of the cesium standard. The systematic uncertainty of +/- 10 Hz assigned to f_{Hg} is based on theoretical arguments (see below) in lieu of a full experimental evaluation. A second Hg standard has been constructed toward making a full evaluation.

6. External field shifts to the clock transition

While the $^2S_{1/2}$ ($F = 0$, $m_F = 0$) - $^2D_{5/2}$ ($F = 2$, $m_F = 0$) hyperfine component has no linear Zeeman shift, it does have a quadratic Zeeman shift. In addition there is a second-order Stark shift and a shift due to the interaction between a static electric-field gradient and the D-state atomic electric-quadrupole moment. None of these shifts has yet been measured accurately but their values have recently been calculated [34]. The fractional frequency uncertainties that might be reasonably expected for the quadratic Zeeman shift and the second-order Stark shift should be $\leq 10^{-18}$. The fractional frequency uncertainty associated with the electric-quadrupole shift is more difficult to reduce to a negligible value. The atomic quadrupole moment arises from the departure of the electronic charge distribution of an atom from spherical symmetry. The interaction of the atomic quadrupole moment with static external field gradients, such as those that might be generated by static potentials on the electrodes of an ion trap, is analogous to the interaction of a nuclear quadrupole moment with the electric-field gradients due to the atomic electrons. In principle, a spherical Paul trap should produce no quadrupole shift since the ion is bound in the pseudo-potential of an inhomogeneous oscillating electric field and no static electric field is applied. However, patches of stray charges can give rise to field gradients that might be as large as 10^3 V/cm^2 for a Paul trap, such as ours, where the ring diameter is about 1 mm. For the S-D transition in $^{199}Hg^+$, a field gradient of that magnitude would give a maximum quadrupole shift on the order of 1 Hz. It was shown in Ref. 34 that the average value of the diagonal matrix elements of the Hamiltonian describing the interaction of external electric-field gradients with the atomic quadrupole moment is zero when the values correspond to any three mutually perpendicular orientations of the laboratory quantization axis. Hence the quadrupole shift can be eliminated from the observed transition frequency by sequentially orienting a magnetic field of constant magnitude in each of three mutually orthogonal directions and averaging the transition frequencies that are measured for each orientation. We also note that states with zero total angular momentum have no quadrupole shift. For example, the $^2S_{1/2}$ (F = 2, m_F = 0) - $^2D_{3/2}$ (F = 0, m_F = 0) hyperfine component in $^{201}Hg^+$, for which the nuclear spin I = 3/2, has no quadrupole shift [35]. Quadrupole transitions without a DC quadrupole shift can be found in several other ions that are being studied as potential references for optical clocks [36].

7. Conclusions

In conclusion, we have constructed an optical clock based on the 1.064 PHz (282 nm) electric-quadrupole transition in a laser-cooled, single $^{199}Hg^+$ ion. The optical frequency is phase-coherently divided down to provide a microwave output using a mode-locked femtosecond laser and a microstructure optical fiber. The short-term (1 s) instability of the optical output of the clock is measured against an independent optical standard to be $\leq 7 \times 10^{-15}$. This optically-referenced femtosecond comb provides a countable output at 1 GHz, which should ultimately be usable as a higher-accuracy reference for timescales, synthesis of frequencies from the RF to the UV, comparison to other atomic standards, and tests of fundamental properties of nature.

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