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^{199}Hg$^+$ OPTICAL FREQUENCY STANDARD: PROGRESS REPORT*

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ABSTRACT

We are developing an optical frequency standard based on the narrow $^2S_{1/2}$–$^2D_{5/2}$ electric-quadrupole transition of a single trapped ^{199}Hg$^+$ ion. Small linear traps designed to operate at liquid helium temperatures have been constructed which will provide confinement in the Lamb-Dicke regime for the optical transition. Cryogenic operation yields long ion storage times and significantly suppresses frequency shifts due to collisions and blackbody radiation. A major step toward the completion of this standard has been the construction and evaluation of a well isolated, high-finesse, Fabry-Pérot cavity to stabilize the frequency of the interrogating laser. We report our cavity results and progress toward locking the laser frequency to the transition in the stored ion.

1 INTRODUCTION

Atomic ions stored in ion traps experience only very small perturbations of their internal energy levels, and hence make good candidates for stable and accurate frequency standards. Neutral atom and trapped ion standards based on microwave transitions have been developed that demonstrate fractional stabilities near or better than $10^{-13}\tau^{-1/2}$, where τ is the measurement averaging time. However, certain proposals such as the timing of millisecond pulsars and low-frequency gravitational wave detection require performance beyond that of present day standards [1, 2, 3]. The quantum-projection-noise-limited Allan variance of an atomic standard interrogated by the time-domain Ramsey method is given by the expression

$$\sigma_y(\tau) = \sqrt{\frac{\langle(\langle\omega_k\rangle_\tau - \langle\omega_{k+1}\rangle_\tau)^2\rangle_\tau}{2\omega_0^2}} = \frac{1}{\omega_0\sqrt{NT_R\tau}}. \quad (1)$$

In this expression for the two-sample Allan variance $\sigma_y(\tau)$, $\langle\omega_k\rangle_\tau$ corresponds to the kth measurement of the frequency of an oscillator locked to a reference consisting of an ensemble of N atoms each having transition frequency ω_0. The interrogation period is of length $T_R < \tau$. Although much performance improvement has been realized in microwave standards by increasing the number of atoms N in the probed sample and by extending the interrogation time T_R, gains in stability may be offset by losses in accuracy and *vice versa*. Such a degradation may arise, for example, from the introduction of uncontrolled perturbations such as collisional or Doppler shifts in dense or large samples, or reduction in signal when evolving for long T_R due to loss of atomic beam intensity. Increasing the frequency ω_0 by making use of long-lived optical transitions is perhaps the most direct way to improve the overall performance, but significant technical obstacles must be overcome. In particular, optimum performance requires a spectrally pure optical local oscillator possessing a linewidth less than the natural width of the atomic transition for times comparable to the interrogation time T_R.

We report on progress made toward the realization of such an optical frequency standard based on the narrow $5d^{10}6s\ ^2S_{1/2} - 5d^96s^2\ ^2D_{5/2}$ electric-quadrupole transition of a single laser-cooled ^{199}Hg$^+$ ion stored in a radiofrequency (Paul) trap [4]. The single-photon transition wavelength is 282 nm with a natural width of 1.8 Hz, corresponding to a line Q of greater than 5×10^{14}. We expect that it should be possible to stabilize the frequency of a laser oscillating in resonance with the atomic transition (or a subharmonic thereof) within $10^{-15}\tau^{-1/2}$, with an accuracy ultimately approaching 10^{-18}.

2 REFERENCE CAVITY

A two-step scheme is employed to stabilize a ring-dye laser operating at 563 nm, which lies at the heart of the local oscillator. The laser is first prestabilized by locking to a low-finesse ($\mathcal{F} \approx 800$) Fabry-Pérot cavity using the Pound-Drever-Hall reflected sideband technique [5]. High-frequency correction of the laser frequency noise is accomplished with an intracavity electro-optic modulator, and long term drifts are eliminated by steering a piezoelectric transducer (PZT) mounted behind one of the dye laser cavity mirrors. The prestabilization stage narrows the laser linewidth to ≈1 kHz for averaging times of about 1 s. The light is then transported through an optical fiber to a high-finesse ($\mathcal{F} > 150\,000$) cavity, and the reflected sideband technique is again used in conjunction with an acousto-optic modulator (AOM) to lock the laser to the cavity resonance. The short-term corrections are written directly onto the AOM drive frequency, while long-term drifts are fed back to a PZT on the prestabilization cavity to maintain frequency alignment with the high-finesse cavity.

Our requirement that the spectral purity of the laser be less than the 1.8 Hz natural linewidth of the ${}^2S_{1/2}$–${}^2D_{5/2}$ optical clock transition in ${}^{199}Hg^+$ places stringent demands on the physical stability of the high-finesse cavity. For this reason, the cavity spacer must have intrinsically low sensitivity to temperature variations and must be well isolated from the environment. The present apparatus uses mirrors optically contacted to the ends of a tapered cylindrical spacer 15 cm in diameter and 24 cm in length; a 1 cm diameter open bore forms the intracavity region. Both the spacer and mirror substrates are made of ULE glass [6], and the temperature is regulated near the point at which the temperature-dependent coefficient of thermal expansion is zero (≈30 °C). The cavity is isolated from the environment by mounting it in an evacuated chamber on an optical table which is passively isolated from seismic noise using a support system consisting of vertical strands of surgical tubing stretched to approximately 3 m. In the vertical, the fundamental vibrational mode of the suspended table has a frequency of ≈0.3 Hz, which provides an isolation from floor noise that exceeds a factor of 50 in noise amplitude at frequencies greater than 3 Hz. Some viscous damping is also employed, and the optical table is further enclosed in a wooden box lined internally with lead foam to reduce the coupling of acoustic noise into the cavity.

Dissipation of intracavity light in the dielectric coatings and radiation pressure on the mirrors shifts the cavity resonance. To stabilize this shift and hold it to an acceptable value, we couple only approximately 100 μW of 563 nm light into the cavity, and a servo actively controlling the radiofrequency power driving the AOM stabilizes the optical power exiting the cavity to ≈0.1 %.

To characterize the short term stability of the cavity without reference to the atomic ion, we utilize a second similar high-finesse cavity on a separate isolated table. Figure 1 shows the spectrum of the beat note obtained by heterodyning two laser beams derived from the beams stabilized to the two independent cavities. A nearly uniform relative cavity drift of ≈1 Hz/s is suppressed by mixing the beat note with a swept frequency synthesizer. A common-mode drift of similar size is also observed. The width of the spectrum at the half-power point is about 0.9 Hz (32 s averaging time). Assuming that the length fluctuations of the cavities are similar in frequency distribution and amplitude but otherwise independent implies that the frequency fluctuations of neither laser contributes a frequency width greater than 0.6 Hz at 563 nm. This corresponds to a fractional linewidth of only 1×10^{-15}. Time-domain measurements described in Ref. [7] were used to determine the fractional frequency instabilities $\sigma_y(\tau)$ for the 30 ms to several second time scale relevant for our proposed optical standard. These results, and the Allan deviations reported for a number of other stable laser systems, are compared in Figure 2.

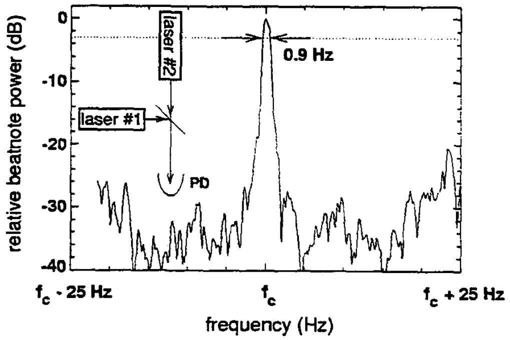

Figure 1: Power spectrum of the beat note between two laser beams stabilized to two independent cavities. The resolution bandwidth of the spectrum analyzer is 0.447 Hz, and the averaging time is 32 s.

3 SINGLE-ION REFERENCE

We work with singly charged ions of ${}^{199}Hg$ because they offer transitions suitable for both optical [4] and microwave standards [14, 15]. Figure 3 shows the ${}^2S_{1/2}$–${}^2P_{1/2}$ electric-dipole transitions used for laser cooling, optical pumping, and state detection, and the ${}^2S_{1/2}$–${}^2D_{5/2}$ electric-quadrupole transition that is the reference for the optical standard. A number of technical barriers must be surmounted in working with Hg^+. One is associated with the presence of residual elemental Hg in the trap vacuum vessel. At room temperature, the storage lifetime is reduced to a few minutes through recombination of the trapped ions with neutral partners from the background. We minimize the ambient Hg by producing it only when needed via thermal disassociation of isotopically enriched HgO. To obtain the desired ion storage times

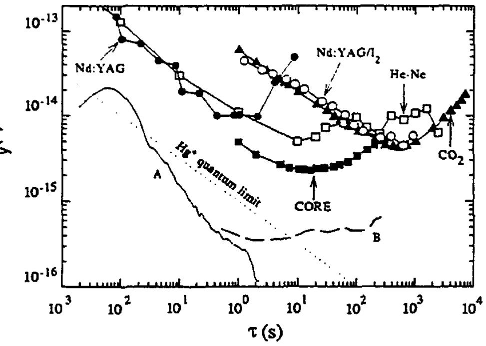

Figure 2: Allan deviation curves for stabilized lasers. We calculate $\sigma_y(\tau)$ for one of our sources from an analog-to-digital sample of the beat signal (curve A) and via a dual-mixer measurement system (curve B). The dotted line shows the quantum noise limit for a Hg^+ optical frequency standard with a single ion and $T_R = 30$ ms. Results for other stabilized lasers: Nd:YAG, Nd:YAG lasers locked to cavities [8]; Nd:YAG/I_2, iodine-stabilized Nd:YAG lasers [9]; He-Ne, methane-stabilized He-Ne lasers [10]; CO_2, CO_2 lasers locked to OsO_4 [11] (see comparable results in Ref. [12]); CORE, Nd:YAG lasers locked to cryogenic resonator oscillators [13].

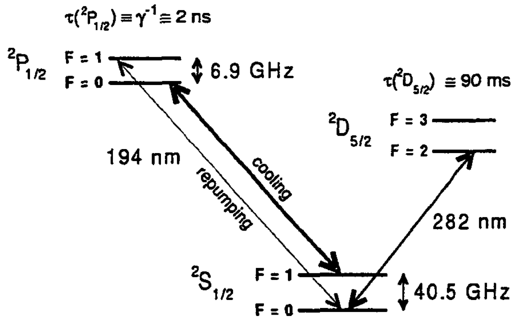

Figure 3: Simplified energy-level scheme for $^{199}Hg^+$. 194 nm radiation is used for Doppler cooling of the ion's motion; the long-lived optical clock transition is at 282 nm. The 40.5 GHz microwave clock transition is also shown.

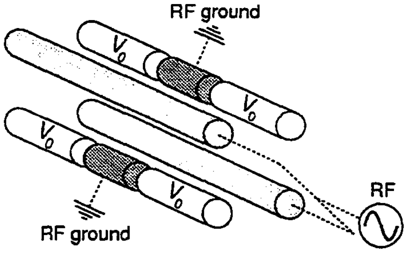

Figure 4: Basic geometry of the linear ion trap. Radial confinement is provided by a ponderomotive potential resulting from the RF voltage (several hundred volts at 10 MHz) applied to two opposing rods, with the complementary pair at RF ground. Trapping along the axis is provided by a DC potential V_0 (0-200 V) applied to the outer electrode segments.

(days), it is necessary to further refrigerate the trap enclosure to liquid He temperatures, so that any remaining free Hg is sequestered by cryopumping on the vessel walls. Cryogenic operation, however, introduces a host of other challenges, most prominently the accumulation and "freezing out" of patches of charge on the trap electrodes. The resulting stray electric fields can add enough additional bias to prevent the trapping of ions. Therefore, heaters must be incorporated into the trap structure so that its temperature may be elevated enough to permit the dissipation of any charge accumulated during the time the Hg oven and ionizing electron beam are activated. After loading, the trap is returned to cryogenic operation without further evidence of fluctuating bias fields.

We are experimenting with a number of heated trap structures, all variations of the linear geometry shown schematically in Figure 4. The operating principles of the linear Paul trap and laser cooling are described in detail elsewhere [16], but a few comments relevant to the present discussion are in order. Ions stored in radiofrequency traps exhibit a driven micromotion at the trap drive frequency if they are not confined to nodal points of the radiofrequency trapping field. Even small amounts of micromotion can lead to parametric heating of the secular motion of the trapped ions. For a single ion, it is possible to minimize this micromotion with the application of static bias fields which move the ion to the minimum of the ponderomotive potential. The amplitude of micromotion can be directly measured by correlating the phase of the ion scattered-light signal (which is modulated by the Doppler-shifted atomic absorption) with that of the trap drive [17]. By observing the correlation and applying static voltages to compensate for the existing bias, the driven motion can be reduced to negligible levels.

The transition strength of the optical carrier is sensitively dependent on the vibration amplitude, and uncontrolled fluctuations in transition strength may result if the mean excitation number of the harmonic motion is large or changing. This effect can be overcome by strongly confining the ion so that its maximum excursions are much less than a wavelength of the 282 nm interrogating radiation, the Lamb-Dicke regime. The desire for rigid confinement requires that linear traps capable of supporting large electric potentials ($\approx$1-2 kV) with electrode dimensions and spacings $\leq$1 mm be realized in the laboratory. Presently, we have trapped and cooled ions to crystallization in one geometry which is capable of Lamb-Dicke confinement at secular frequencies of $1-2$ MHz during cryogenic operation, and have constructed a smaller, tighter trap which should yield much stronger confinement for identical applied voltages. In the future, we may move to micro-miniature lithographic traps similar to those used in our group at the National Institute of Standards and Technology in Boulder for quantum-state engineering investigations, modified for cryogenic operation.

Another difficulty specific to the Hg^+ experiments is the necessity of a reliable and economical source of narrow-band 194 nm radiation for laser Doppler cooling. Currently, the cooling light is generated using a single-mode Ar^+ laser oscillating at 515 nm that is frequency-doubled in β-barium borate (BBO) to 257 nm. The second-harmonic radiation is then sum-frequency mixed in BBO with the output of diode sources at 792 nm to produce light at 194 nm. We have constructed a Yb:YAG laser at 1.03 μm that is frequency-doubled in $KNbO_3$ to obtain over 400 mW of power at 515 nm, which is sufficient to replace the Ar^+ laser. Because the harmonic generation efficiency is limited by blue-light-induced infrared absorption at high powers in $KNbO_3$, we anticipate even better conversion efficiency by frequency-doubling with lithium triborate in the future. We have also constructed a Nd:FAP laser to replace the 563 nm dye laser and its Ar^+ pump. The inherent frequency stability and low maintenance of these all-solid-state systems make them very attractive for metrological applications and high-precision spectroscopy.

A generic impediment to the realization of any optical frequency standard is the delivery of the phase-stable radiation from the local oscillator to the atom. We transport the 563 nm light via optical fiber from the suspended high-finesse cavity table to the optical table supporting the trap cryostat. Doppler shifts due to the relative motion of the two tables, as well as vibration and insertion noise from the fiber itself, contaminate the phase of the interrogating radiation. This additive noise is sensed by heterodyning light on the cavity table with light which has made a round-trip to the trap table. Phase-locking this beat signal to a stable RF source actively reduces the frequency noise acquired during the transport of the light from the cavity to the ion trap [4, 18]; hence the phase purity of the cavity-stabilized laser is restored at the site of ion. The 563 nm light is frequency-doubled in deuterated ammonium dihydrogen phosphate to near resonance with the ion at 282 nm near the trap. Measurement or locking of the 282 nm light to the Hg^+ transition begins with laser cooling of the ion to near the Doppler limit of 1.7 mK with a pair of copropagating 194 nm beams: a strong (several hundred microWatts) cooling beam, and a weaker ($\approx$1 μW) repumping beam which prevents the ion from becoming trapped in the $^2S_{1/2}$ $F = 0$ level. Turning off the repumping radiation prepares the ion in the lower ($F = 0$) hyperfine level of the ground state. The clock transition is then probed with the time-domain Ramsey method in the absence of any 194 nm radiation. Finally, transitions to the $^2D_{5/2}$ level are detected using optical-optical double resonance ("electron shelving") [19], in which we detect light scattered from the strong cycling $^2S_{1/2}$–$^2P_{1/2}$ electric-dipole transition at 194 nm. If the atom is in the $^2D_{5/2}$ state, no scattering above background is observed. If the atom remains in the ground state, we detect strong scattering as the ion is driven up to the $^2P_{1/2}$ state and decays spontaneously back to the ground state. For a single ion starting in the ground state, a peak fluorescence count rate of tens of kiloHertz has been observed using a photomultiplier. This quantum amplification enables state detection with nearly unit efficiency. A digital integral servo loop will adjust the average frequency of the 282 nm radiation by means of an AOM to match the center of the detected atomic resonance. The values of the steered frequency will be recorded after each measurement cycle for evaluation [4, 20].

4 PERFORMANCE

Single laser-cooled ions stored in traps offer significant advantages over other atomic references in terms of relative immunity from systematic frequency shifts. For $^{199}Hg^+$ the fractional magnitude of the second order Doppler shift is approximately 2×10^{-18} at the Doppler cooling limit. We have chosen an isotope with nonzero nuclear spin which makes available transitions which are first-order independent of magnetic field strength at zero field. Hence the first-order Zeeman shift can be made very small. In our trap, collisional and blackbody radiation shifts are minimized by the cryogenic operation required for long storage times. Although light shifts from the strong cooling radiation can broaden the narrow reference transition, we shutter the cooling beams during the probe cycle to prevent this from occurring. In probing the $^2S_{1/2}$–$^2D_{5/2}$ resonance, AC Stark effects are negligible as we rely on a single-photon quadrupole transition. Because we use an optical transition to a state with a nonvanishing electronic quadrupole moment, the limiting accuracy of our standard may arise from the uncertainty in the interaction of the $^2D_{5/2}$ atomic moment with the electric fields of the trap. While the quadrupole shift from the AC trapping fields can be precisely calibrated since they affect the motion of the ion in a known way, static fields or patches of charge on the trap electrodes may be difficult to control. Shifts larger than 1 Hz may be expected. We can significantly reduce the uncertainty in these shifts by measuring the quadrupole transition frequencies for three mutually orthogonal orientations of a quantizing magnetic field, in which case the quadrupole shift averages to zero.

From Eqn. (1) with $T_R = 0.03$ s, we estimate a projection noise-limited measurement stability for a single ion of approximately 1 part in 10^{15}, or 1 Hz, for averaging times of 1 s. We have demonstrated an optical local oscillator with a linewidth of less than 0.6 Hz for averaging times up to 32 s, which is sufficient for such an Hg^+ optical standard. We are hopeful that the system we are assembling, in combination with a frequency synthesis chain connecting the optical transition to microwave frequencies [21], might eventually provide a time standard with an accuracy near 10^{-18}, and stability surpassing the best present-day clocks.

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