

## High-accuracy frequency standards using laser-cooled $\text{Hg}^+$ ions\*

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We discuss frequency standards based on laser-cooled  $^{199}\text{Hg}^+$  ions confined in cryogenic rf (Paul) traps. In one experiment, the frequency of a microwave source is servoed to the ions' ground-state hyperfine transition at 40.5 GHz. For seven ions and a Ramsey free precession time of 100 s, the fractional frequency stability is  $3.3 (2) \times 10^{-13} \tau^{-1/2}$  for measurement times  $\tau < 2$  h. The ground-state hyperfine interval is measured to be 40 507 347 996.841 59 (14) (41) Hz, where the first number in parentheses is the uncertainty due to statistics and systematic errors, and the second is the uncertainty in the frequency of the international time scale to which the standard is compared. In a second experiment under development, a strong-binding cryogenic trap will confine a single ion for use in an optical frequency standard based on a narrow electric quadrupole transition at 282 nm.

### I. INTRODUCTION

Frequency standards based on trapped and laser-cooled ions can achieve both high accuracy and high stability [1, 2, 3].  $^{199}\text{Hg}^+$  offers a microwave clock transition at 40.5 GHz and an optical clock transition at  $1.06 \times 10^{15}$  Hz (see Fig. 1). To first order, both transitions are insensitive to magnetic and electric fields at zero fields. Using a linear trap, we expect to reduce all systematic frequency shifts of the  $^{199}\text{Hg}^+$  40.5 GHz transition to less than a part in  $10^{16}$ . Using a spherical quadrupole trap and a single ion, we expect to reach a relative uncertainty smaller than  $10^{-17}$  for the optical transition.

If the fluctuations of the atomic signal are due only to quantum statistics, then the stability of a frequency source servoed to the atomic transition is given by [4, 5]

$$\sigma_y(\tau) = \frac{1}{\omega_0 \sqrt{N T_R}} \tau^{-1/2} \quad (1)$$

where  $\omega_0$  is the frequency of the atomic transition,  $N$  is the number of ions,  $T_R$  is the Ramsey interrogation time, and  $\tau$  is the averaging time of the measurement. For the ground-state hyperfine transition,  $\omega_0/2\pi = 40.5$  GHz. It appears feasible to use  $N = 100$  ions and  $T_R = 100$  s, which gives  $\sigma_y(\tau) \approx 4 \times 10^{-14} \tau^{-1/2}$ . For the 282 nm  $^2S_{1/2} \rightarrow ^2D_{5/2}$  electric quadrupole transition,  $\omega_0/2\pi = 10^{15}$  Hz, so that using  $N = 1$  and  $T_R = 25$  ms gives  $\sigma_y(\tau) \approx 10^{-15} \tau^{-1/2}$ .

## II. CRYOGENIC LINEAR rf TRAP

Figure 2 shows the linear rf trap used in the 40.5 GHz microwave frequency standard. Two diagonally opposite rods are grounded, while the potential of the other two rods is  $V_0 \cos(\Omega t)$ , where  $V_0 = 150$  V and  $\Omega/2\pi = 8.6$  MHz. The resulting pseudopotential confines the ions radially in a well with a secular frequency  $\omega_r/2\pi = 230$  kHz. To reduce patch fields and remove electrical charge that otherwise leaves the rods slowly in the cryogenic environment, we thread insulated resistive wires through the rods to heat them. Two cylindrical sections at either end of the trap are held at a potential of approximately +10 V, confining the ions axially. The ions are laser-cooled using the 194 nm  $^2S_{1/2} \rightarrow ^2P_{1/2}$  electric dipole transitions shown in Fig. 1. Typically, a string of approximately ten ions is confined near the trap axis. By minimizing the ion micromotion in all three dimensions, we assure that the laser-cooled ions lie along the rf nodal line [6]. Here, the atomic motion driven by the oscillating electric field of the trap (the micromotion) is also minimized, so Doppler shifts and ac Stark shifts are negligible. Furthermore, rf heating is minimized, so the cooling laser radiation, which perturbs the clock states, can be removed during the long probe periods of the clock transition:

We place the trap in a liquid He cryogenic environment. At 4 K, Hg and most other background gases are cryopumped onto the walls of the chamber. This greatly reduces the ion loss rate due to collisions with the background gas. In addition, at this low background pressure, pressure shifts should be negligible. Operation at 4 K also reduces the frequency shifts due to blackbody radiation of the  $^{199}\text{Hg}^+$  ground-state hyperfine transition. At  $T = 4$  K, the fractional blackbody Zeeman shift is  $-2 \times 10^{-21}$ , and the fractional blackbody Stark shift is  $-3 \times 10^{-24}$  [7].

## III. LASER-ATOM INTERACTIONS

Laser beams at 194 nm [8] are used for cooling, state preparation, and determining the internal states of the ions. To cool the ions, we tune the frequency of a primary laser slightly below resonance with transition  $p$ .

Although this is a cycling transition, the laser can off-resonantly excite an ion into the  ${}^2P_{1/2}$ ,  $F = 1$  level, from which the ion can decay into the  ${}^2S_{1/2}$ ,  $F = 0$  level. To maintain fluorescence, a repumping laser beam, resonant with transition  $r$ , is overlapped collinearly with the primary laser beam. To prevent optical pumping into the dark states of the ground state  $F = 1$  level, the beams  $r$  and  $p$  cross the ions at  $+20^\circ$  and  $-20^\circ$  relative to the trap axis. The polarization of the beam at  $+20^\circ$  is in the plane of the 194 nm beams, while the polarization of the remaining light is continuously modulated between left and right circular.

The ions are prepared in the  ${}^2S_{1/2}$ ,  $F = 0$  state by turning the repumping beam off. We determine the atomic state after the clock radiation is applied by pulsing on the primary beam for a time comparable to the time necessary to pump the ions from the  ${}^2S_{1/2}$ ,  $F = 1$  to the  ${}^2S_{1/2}$ ,  $F = 0$  level (typically 10 ms). If the ion is found in the  ${}^2S_{1/2}$ ,  $F = 1$  level, it will scatter about  $10^4$  photons before it optically pumps into the  ${}^2S_{1/2}$ ,  $F = 0$  level. We detect and count approximately 150 of these photons. If the ion is found in the  ${}^2S_{1/2}$ ,  $F = 0$  level, it will scatter only a few photons.

#### IV. THE 40.5 GHZ MICROWAVE CLOCK

For the first part of our measurement cycle, we cool the ions by pulsing on both the primary and repumping 194 nm laser beams for 300 ms. Next, we turn off the repumping beam for about 90 ms, so that essentially all of the ions are optically pumped into the  ${}^2S_{1/2}$ ,  $F = 0$  level. We probe the clock transition using the Ramsey technique of separated oscillatory fields [9]. Both beams are blocked during the Ramsey microwave interrogation period, which consists of two 250 ms microwave pulses separated by the free precession period  $T_R$ . We vary  $T_R$  from 2 to 100 s. Finally, we turn on the primary beam for about 10 ms while counting the number of detected photons. This determines the ensemble average of the atomic state population and completes one measurement cycle.

We derive the microwave frequency from a low-noise crystal quartz oscillator offset-locked to a reference hydrogen maser [10]. Stepping the synthesizer frequency between measurement cycles produces a set of Ramsey fringes. A digital servo locks the average synthesizer frequency to the central Ramsey fringe. For  $N = 7$  and  $T_R = 100$  s, the fractional frequency stability of the synthesizer is  $\sigma_y(\tau) \cong 3.3 (2) \times 10^{-13} \tau^{-1/2}$ , for  $\tau \leq 2$  h. Consistently, the measured  $\sigma_y(\tau)$  is about twice the value expected from Eq. (1), primarily because of laser intensity fluctuations at the site of the ions. Our measured frequency stability is comparable to those of the Cs beam standard NIST-7, for which  $\sigma_y(\tau) \cong 8 \times 10^{-13} \tau^{-1/2}$  [11], and the Cs fountain standard, for which  $\sigma_y(\tau) \cong 2 \times 10^{-13} \tau^{-1/2}$  [12].

Table 1 shows the most important corrections we make to the average frequency for each run [13]. The fractional Zeeman shift due to the static magnetic field is  $+0.24 B_{static}^2$ , where  $B_{static}$  is in teslas. The measured Zeeman splitting of the ground state hyperfine transitions gives  $B_{static} \equiv 3 \times 10^{-7}$  T, with fluctuations of  $1 \times 10^{-8}$  T. Thus the fractional uncertainty in this Zeeman shift is  $1.4 \times 10^{-15}$ .

We also correct for an ac Zeeman shift that depends linearly on the rf power  $P_{rf}$  delivered to the trap. This shift is caused by magnetic fields due to asymmetric currents at frequency  $\Omega$  in the trap electrodes. These currents are caused by stray capacitance to the ground plane. Considering that the current distribution may vary from load to load, we measure the average transition frequency versus  $P_{rf}$  for each ion crystal. A fit to these data gives the frequency shift  $P_{rf} (d\omega/dP_{rf})/\omega_0$  and the extrapolated frequency at zero rf power  $\omega_0$ , for that crystal. Typically,  $(d\omega/dP_{rf})/\omega_0 \equiv (2.5 \pm 2.1) \times 10^{-16}$  /mW; within the uncertainty, this value is the same for each crystal. In 18 days, we measured  $\omega_0$  five times, using 42 runs and five different ion crystals. The normalized  $\chi^2$  for these five frequency measurements is 0.77, and  $\omega_0$  is independent of time within our uncertainty. The uncertainty in the extrapolated frequency averaged over the five ion crystals used in the frequency measurement is  $3.2 \times 10^{-15}$ .

The magnitudes of several frequency shifts scale with the free precession time as  $1/T_R$ . These include shifts due to the phase chirp of the microwaves as they are switched on and off, any leakage microwave field present during the free precession time  $T_R$ , and asymmetries in the microwave spectrum. By varying  $T_R$ , we measure the fractional shift from these combined effects to be  $-3(3) \times 10^{-14}/T_R$  (where  $T_R$  is in seconds).

We determine the frequency of our reference maser relative to that of the primary standards by comparing it to that of the International Atomic Time (TAI) [14]. This determines the average frequency of our  $Hg^+$  standard to be  $\omega_0 = 2\pi \cdot 40\,507\,347\,996.841\,59(14)(41)$  Hz. Here, the first uncertainty is due to the systematic shifts shown in Table 1. The second uncertainty is due to the quoted uncertainty in the frequency of TAI. Our uncertainty from systematic effects (3.4 parts in  $10^{15}$ ) is approximately equal to the best values reported, from a cesium beam clock (5 parts in  $10^{15}$ ) [15], and a cesium fountain clock (2 parts in  $10^{15}$ ) [16].

Our main uncertainties can be reduced in future work. Better magnetic shielding will reduce fluctuations in the static magnetic field. The magnetic field at frequency  $\Omega$  can be reduced by decreasing  $\Omega$  and the trap dimensions. Finally, by monitoring each ion individually, we can determine their internal states with negligible uncertainty, which will eliminate noise due to frequency and intensity fluctuations of the laser.

## V. OPTICAL FREQUENCY STANDARD

We are now developing a frequency standard based on the  $^{199}\text{Hg}^+$ , 282 nm electric quadrupole transition, which has a natural linewidth of 1.7 Hz. Since the clock transition frequency  $\omega_0$  is high ( $\omega_0/2\pi = 1.1 \times 10^{15}$  Hz), the fractional stability can be very high, as seen in Eq. (1). For example, for a single ion probed using the Ramsey technique with a free precession time of 25 ms (limited by the  $^2\text{D}_{5/2}$ -state lifetime), the fractional frequency instability is about  $1 \times 10^{-15} \tau^{-1/2}$ , two orders of magnitude lower than that of our microwave standard.

Previously at NIST, the frequency of a stabilized laser at 563 nm was doubled and then locked to this transition in a single ion confined near the Lamb-Dicke limit [17]. Periodically, the laser frequency was unlocked and scanned to obtain a laser-broadened line shape approximately 80 Hz wide at 563 nm [18]. However, in the room temperature environment, collisions with the background gas limited the storage time of the ion to about ten minutes. To thoroughly investigate such a frequency standard, we require the long-observation times provided in the cryogenic environment. We have trapped crystals of mercury ions in a second cryogenic system that houses a quadrupole rf trap with a ring diameter of 932  $\mu\text{m}$ . During loading, the trap electrodes are heated by beams of light from an argon ion laser to prevent charge from building up on the surfaces. We have also improved the stabilized laser system that generates the 282 nm radiation [18, 19].

## VI. SUMMARY

We have evaluated a frequency standard based on the 40.5 GHz transition in laser-cooled strings of  $^{199}\text{Hg}^+$  ions in a linear cryogenic rf trap. The stability and accuracy of this standard are comparable to those of the best cesium standards. The fractional uncertainty in  $\omega_0$  is limited primarily by Zeeman shifts, which should be significantly reduced in future experiments. We are also developing a 282 nm optical frequency standard, whose potential stability is  $\sigma_y(\tau) \approx 10^{-15} \tau^{-1/2}$ .

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## VIII. REFERENCES

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Table 1: Systematic shifts of the average clock transition frequency. Typical values are for an rf power  $P_{rf} = 20mW$ , a Ramsey interrogation time  $T_R = 100s$ , and a static magnetic field  $B_{static} = 3 \cdot 10^{-7}$  T. Here,  $B_{\Omega}$  is the magnetic field component at frequency  $\Omega$

| Shift  | scaling                          | Magnitude of effect (typical) | Uncertainty in effect |
|--|----------------------------------|-------------------------------|-----------------------|
| Quadratic Zeeman (static)                          | $+ \langle B_{static}^2 \rangle$ | $2 \cdot 10^{-14}$            | $1.4 \cdot 10^{-15}$  |
| Quadratic Zeeman ( $\Omega$ )                      | $+ \langle B_{\Omega}^2 \rangle$ | $5 \cdot 10^{-15}$            | $3.2 \cdot 10^{-15}$  |
| Microwave chirp, leakage, and spectrum asymmetries | $1/T_R$                          | $3 \cdot 10^{-15}$            | $8 \cdot 10^{-16}$    |

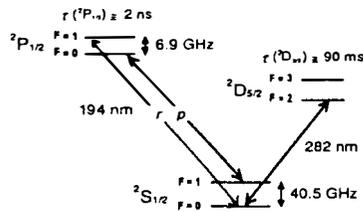


Figure 1: Partial energy diagram of  $^{199}\text{Hg}^+$

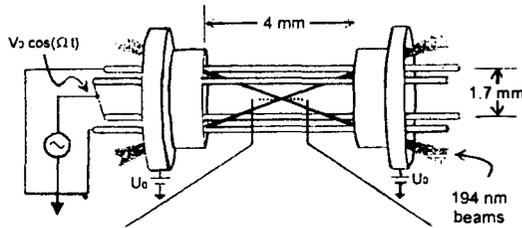


Figure 2: Linear rf trap, and an image of a linear ion crystal. The ions are spaced approximately 10 nm apart, and the gaps in the ion crystal are due to ions other than  $^{199}\text{Hg}^+$ , which do not fluoresce at the frequencies of the 194 nm laser beams. The spatial extent of the ions is exaggerated for clarity, in reality the laser beams simultaneously overlap all the ions.