J. Phys. B: At. Mol. Opt. Phys. 36 (2003) 545-551

PII: S0953-4075(03)54352-8

# The <sup>199</sup>Hg<sup>+</sup> single ion optical clock: recent progress

U Tanaka<sup>1,2</sup>, S Bize<sup>1,3</sup>, C E Tanner<sup>1,4</sup>, R E Drullinger<sup>1</sup>, S A Diddams<sup>1</sup>, L Hollberg<sup>1</sup>, W M Itano<sup>1</sup>, D J Wineland<sup>1</sup> and J C Bergquist<sup>1</sup>

<sup>1</sup> Time and Frequency Division, National Institute of Standards and Technology, 325 Broadway, Boulder, CO 80305, USA

<sup>2</sup> Communications Research Laboratory, 588-2 Iwaoka, Iwaoka-cho, Nishi-ku,

Kobe 651-2492, Japan

<sup>3</sup> Laboratoire Systèmes de Référence Temps-Espace, 61, avenue de l'Observatoire, 75014 Paris, France

<sup>4</sup> University of Notre Dame, Department of Physics, 225 Nieuwland Science Hall, Notre Dame, IN 46556-5670, USA

Received 30 September 2002 Published 23 January 2003 Online at stacks.iop.org/JPhysB/36/545

### Abstract

We report on work directed toward the systematic evaluation of an optical frequency standard based on the  ${}^{2}S_{1/2}-{}^{2}D_{5/2}$  transition of a single, laser-cooled, trapped  ${}^{199}$ Hg<sup>+</sup> ion, whose resonance frequency is  $1.06 \times 10^{15}$  Hz. For the purpose of the evaluation, two  ${}^{199}$ Hg<sup>+</sup> standards have been constructed. In the cooling-laser system built for the second standard, an injection-locking scheme has been applied to a cw Ti:sapphire laser. We also report optical frequency measurements of the clock transition performed over the past 21 months with the first standard. During this term, the variation of the clock transition frequency is found to be less than one part in  $10^{14}$ .

(Some figures in this article are in colour only in the electronic version)

#### 1. Introduction

An optical clock based on a single trapped ion promises higher stability and accuracy than those of present-day time standards. Significant recent developments have made an optical clock more feasible. First, the realization of sub-Hertz linewidth laser systems [1, 2] and the development of mode-locked femtosecond lasers as coherent frequency-comb generators [3–5] have now made possible accurate measurements of absolute optical atomic frequencies [6, 7]. Second, an all-optical atomic clock comprising a femtosecond comb that is referenced to a narrow transition in a single trapped <sup>199</sup>Hg<sup>+</sup> ion has been demonstrated. A fractional frequency instability (Allan deviation) of  $7 \times 10^{-15}$  at 1 s was obtained for this clock [8]. The quantumlimited instability of a laser locked to the Hg<sup>+</sup> ion is expected to be about  $1 \times 10^{-15} \tau^{-1/2}$ , where  $\tau$  is the measurement period in seconds, with a fractional frequency uncertainty approaching  $10^{-18}$  [9, 10]. To achieve this goal, experimental evaluations of the uncertainty, which are assisted by comparing two independent systems, are indispensable. In this paper, we report progress in the development of a second Hg<sup>+</sup> ion frequency standard built for the purpose of

0953-4075/03/030545+07\$30.00 © 2003 IOP Publishing Ltd Printed in the UK

a systematic evaluation of the first frequency standard. In particular, we describe a second light system for laser cooling that uses a cw injection-locked Ti:sapphire laser. We also report optical-frequency measurements of the clock transition of the <sup>199</sup>Hg<sup>+</sup> ion, which have been performed over the past 21 months. During this period, the variation of the clock transition frequency was found to be less than  $1 \times 10^{-14}$ .

#### 2. Atomic system and experimental setup

A partial energy-level diagram of <sup>199</sup>Hg<sup>+</sup> is shown in figure 1. The  ${}^{2}S_{1/2}(F = 0, M_{F} = 0) \rightarrow$  $^{2}D_{5/2}(F = 2, M_{F} = 0)$  electric-quadrupole transition at 282 nm provides the reference for the optical standard. The natural linewidth of the S-D transition is about 2 Hz at a resonance frequency of  $1.06 \times 10^{15}$  Hz. The 282 nm radiation used to drive the clock transition is generated in a nonlinear crystal as the second harmonic of a dye laser oscillating at 563 nm. A laser linewidth below 0.2 Hz for averaging periods from 1 to 10 s is realized by locking the frequency of the dye laser to a high-finesse Fabry-Pérot cavity that is temperature-controlled and supported on an isolation platform [1]. The  ${}^{2}S_{1/2}(F = 1) \rightarrow {}^{2}P_{1/2}(F = 0)$  transition at 194 nm is used for laser cooling. Another 194 nm light beam (the repumper) tuned to the  ${}^{2}S_{1/2}(F = 0) \rightarrow {}^{2}P_{1/2}(F = 1)$  transition is employed to return the ion to the ground state F = 1 level. These transitions are also used for state preparation and detection. A single, lasercooled <sup>199</sup>Hg<sup>+</sup> ion is confined in a cryogenic, spherical Paul trap. Under typical operating conditions, the radial secular frequency of the trapped ion is between 1.2 and 1.5 MHz. Staticpotential biasing electrodes are mounted near the trap electrodes, but outside the trapping volume, to cancel stray static-electric fields at the site of the ion. The trap electrodes are housed in a vacuum system held at liquid-helium temperature. At room temperature, collisions with background neutral mercury and hydrogen occasionally result in the formation of either Hg<sup>+</sup><sub>2</sub> or HgH<sup>+</sup> and the loss of the single Hg<sup>+</sup>. Use of a cryogenic system prevents this loss, and has made it possible to confine a single <sup>199</sup>Hg<sup>+</sup> ion continuously for more than 100 days. Transitions to the D state were detected using the technique of 'electron shelving' [11, 12], which infers the presence of the atom in the metastable level through the absence of scattering from the strong laser transition at 194 nm [9]. Laser-cooling, state preparation, excitation of the clock transition, and subsequent detection comprise one measurement cycle. After the ion was lasercooled to near the Doppler limit of 1.7 mK, the ion was prepared in the  ${}^{2}S_{1/2}(F = 0, M_{F} = 0)$ state by blocking the repumping laser. Following this, the cooling laser was also blocked, and in the absence of any 194 nm radiation, the ion was irradiated by the 282 nm light for a period between 10 and 120 ms. Finally, the 194 nm lasers were re-applied and the presence or absence of scattered light was used to determine whether the clock transition occurred. The next cycle commences when the ion is again in the ground state (determined by the presence of scattered light at 194 nm). The linewidths of the observed resonance are limited by the probe time of the 282 nm light. When the clock transition was probed for a period of 120 ms, a linewidth of 6.5 Hz was obtained [9]. For the purpose of locking the frequency of the laser to the clock transition of the single ion, the duration of the 282 nm light pulse was set typically between 20 and 40 ms, which gave a linewidth of approximately 40–20 Hz, respectively. Usually, 48 measurements were made on each side of the resonance before correcting the frequency of the laser toward line centre.

#### 3. Second Hg<sup>+</sup> optical frequency standard

In order to aid in the evaluation of the systematic shifts and their uncertainties, we built a second mercury optical frequency standard comprising a second cryogenic Hg<sup>+</sup> trap and a



Figure 1. Partial energy level diagram for <sup>199</sup>Hg<sup>+</sup>.



Figure 2. Schematic diagram of the second 194 nm light source for the cooling transition. ECDL: extended cavity diode laser; PD: photodetector; PZT: piezoelectric transducer; CL: cylindrical lens; BS: beam splitter.

second 194 nm light source. Narrow-band 563 nm radiation is provided to both systems from the same light source. The frequency of the 563 nm radiation is independently doubled and independently shifted into resonance with the clock transition of each standard by means of acousto-optic modulators.

Figure 2 shows a schematic diagram of the second 194 nm light source. The 194 nm radiation is produced by sum-frequency generation with a beta-barium borate (BBO) crystal, where the wavelengths of the fundamental beams are 257 and 792 nm [13]. The 792 nm light is enhanced in a resonant cavity, while the 257 nm light is single passed through the BBO. The 194 nm beam used as the repumper is also generated in the same BBO crystal

by simultaneously locking a second extended-cavity diode laser to the resonant cavity. The 257 nm radiation is generated by frequency-doubling the 515 nm light from a single-frequency argon-ion laser and is similar to that described in [13]. The frequency of the 515 nm light is locked to a hyperfine component of molecular iodine by feedback to a piezo-mounted cavity mirror. Some of the output power of the stabilized argon-ion laser is guided into a confocal Fabry–Pérot cavity that is 10 cm long and whose mirrors are coated for both 515 and 792 nm. The long-term drift of this cavity is reduced by locking it to the stable 515 nm source. When the frequency of the 792 nm source is then locked to a fringe of the cavity, the stability of the argon-ion laser is transferred to the 792 nm laser and thereby to the 194 nm radiation. The main difference between the two 194 nm sources is that the 792 nm radiation in the second system is produced by an injection-locked Ti:sapphire laser [14]. This choice was primarily dictated by experimental convenience given available equipment, but it nonetheless forms an interesting laser. This method requires only simple optics in the Ti:sapphire laser cavity (the four cavity mirrors plus Ti:sapphire crystal). The master laser at 792 nm is an extendedcavity diode laser in the Littman configuration. The power of the zero-order reflection beam is 11 mW, and the continuous scanning range is about 2 GHz. This beam is mode-matched to the Ti:sapphire cavity with two lenses followed by two successive isolators that give an isolation greater than 60 dB. Because of losses through the isolators, only about 8 mW of power from the master laser reaches the Ti:sapphire cavity. The Ti:sapphire cavity consists of a 10 mm-long, Brewster-cut Ti:sapphire crystal, two mirrors with a common radius of curvature of 15 cm, a flat mirror and a flat output coupler with a reflectivity of 97.6%. All of the other mirrors have a reflectivity exceeding 99.9%. The round-trip length is 157 cm and the minimum beam waist, located in the Ti:sapphire crystal, is 33  $\mu$ m. No other optical element is necessary since the master laser forces single-frequency and unidirectional operation. A commercial frequencydoubled Nd:YVO4 laser at 532 nm is used as the pump laser. As in [14], the frequency of the extended-cavity diode laser is locked to the resonance of the Ti:sapphire cavity by the Pound–Drever–Hall technique [15]. The current of the diode laser is modulated at 16 MHz. A beam splitter is inserted in the Ti:sapphire laser beam such that 0.5% of the power is monitored with a photo-detector. The ac component of the monitored signal is demodulated and used as the error signal. The feedback circuit consists of a fast and slow loop. The fast component of the error signal controls the master laser current and the slow component is used to correct the extended-cavity diode laser cavity length. When free-running, the resonance of the Ti:sapphire laser cavity is driven by acoustical noise and fluctuates about 20 MHz. We reduce this jitter by locking the frequency of the Ti:sapphire laser to the reference cavity that is stabilized to the argon-ion laser. With the servo loop closed, the jitter can be reduced below 1 MHz.

Figure 3 shows the output power of the injection-locked Ti:sapphire laser as a function of pump power for a constant master-laser power of 8 mW. The output power is measured following the beam splitter used to derive the error signal. At pump powers higher than 3 W, the injection lock becomes unstable. The locking range has been shown to depend on the ratio between the power of the master laser and that of the slave laser [14, 16]. Either by using a master laser with more power or by better coupling of the master laser into the Ti:sapphire cavity, we could make the injection-locked laser stable at higher pump powers [14, 16]. A power-enhancement cavity for the 792 nm radiation is used for the sum-frequency generation of the 194 nm radiation. The 792 nm enhancement cavity is formed by two 10 cm radius of curvature mirrors and two flat mirrors. The round-trip length of the cavity is 72.5 cm, and the minimum cavity waist size is approximately  $31 \ \mu$ m. A  $3 \ mm \times 3 \ mm \times 6 \ mm$ , Brewster-cut and angle-tuned BBO crystal is placed at this waist. For this configuration we can obtain a power-enhancement factor of about 110. The cavity length is locked to the frequency of the 792 nm source by use of the Hänsch–Couillaud method [17]. The 257 nm beam is focused



**Figure 3.** Output power of the injection-locked Ti:sapphire laser. The pump power is corrected for the transmission of the 532 nm light through the cavity mirror, which is highly reflecting at 792 nm. The master-laser injection power is held constant at 8 mW.

through the BBO crystal with a highly reflective mirror that has a radius of curvature of 10 cm. The incident angle is adjusted so that both fundamental beams are collinear inside the crystal. With 14 mW of power at 257 nm and 400 mW of power at 792 nm, 48  $\mu$ W of output power at 194 nm are obtained. For the Hg<sup>+</sup> experiments, we operate with a power of about 25  $\mu$ W in order to avoid saturation of the  ${}^{2}S_{1/2}(F = 1)-{}^{2}P_{1/2}(F = 0)$  transition. The second Hg<sup>+</sup> trap has the same dimensions as the first one [7] and is also placed in a cryogenic vessel. Small clouds of Hg<sup>+</sup> ions have been loaded and confined in our second cryogenic system, and fluorescence from these ions has been observed. Measurements of the frequency of the clock transition with the two independent systems are now in progress.

#### 4. Frequency measurements of the clock transition

Optical frequency measurements of the clock transition are made by use of an optical frequency comb [3–6]. The 563 nm light is transferred to the comb through an optical fibre. If we choose the *n*th mode of the comb whose frequency is lower than the fundamental laser frequency  $f_{563}$ , the laser frequency is given by  $f_{563} = f_b + f_0 + nf_r$ , where  $f_0$  is the frequency offset common to all modes of the comb,  $f_r$  is the repetition frequency of the comb (which is about 1 GHz), and  $f_b$  is a beat frequency between  $f_{563}$  and the *n*th mode of the comb. The offset frequency  $f_0$  and the beat frequency  $f_b$  are locked to a hydrogen maser by controlling the pump power and the cavity length of the mode-locked femtosecond laser, respectively. To obtain the value of  $f_{563}$ , we count  $f_r$  with a high-resolution counter that is also referenced to the hydrogen maser. The frequency of the maser [18] is calibrated by comparing it to the local NIST primary standard NIST-F1 [19]. The fractional uncertainty in the frequency of the reference maser reaches  $1.8 \times 10^{-15}$  for these measurements.

Figure 4 summarizes the frequency measurements of Hg<sup>+</sup> made between August 2000 and May 2002. The second-order Zeeman shift was corrected for all values. The statistical uncertainty about  $1.6 \times 10^{-15}$  is essentially the result of the reference maser's short-term



Figure 4. A chronological record of the average daily frequency of the clock transition measured since August 2000.

stability, which is about  $2 \times 10^{-13}$  at 1 s. The weighted mean of our measurements of the Hg<sup>+</sup> clock transition is  $f_{282} = 1064721609899145.9(1.6)(10)$  Hz, where 1.6 Hz is the statistical uncertainty and 10 Hz is the systematic uncertainty. It should be possible to reduce the uncertainties of all systematic shifts to values approaching  $10^{-18}$  [10]. The  ${}^{2}S_{1/2}(F = 0, M_{F} = 0) - {}^{2}D_{5/2}(F = 2, M_{F} = 0)$  transition has no linear Zeeman shift, but it does have a quadratic Zeeman shift, a second-order Stark shift (such as the shift due to the blackbody radiation), and a shift due to the interaction between the electric-field gradient and the atomic electric-quadrupole moment. None of these shifts has yet been measured accurately but their values have recently been calculated by one of us [10]. The largest uncertainty is expected to arise from the quadrupole shift, which is caused by the interaction between the atomic quadrupole-moment of the  ${}^{2}D_{5/2}$  state and a static electric field gradient. In spherical Paul traps, no static field gradient is deliberately applied; however, a gradient can exist due to patch potentials that are randomly distributed on the trap electrodes. For example, in our trap, a static potential of 1 V between ring and endcaps could give a quadrupole shift as large as 1 Hz [10]. This shift can be eliminated by averaging the S–D transition frequency over three mutually orthogonal, equal-magnitude magnetic field orientations [10].

## 5. Conclusion

We have measured the optical clock transition of the <sup>199</sup>Hg<sup>+</sup> ion on several occasions spanning more than 21 months. The weighted average frequency for these measurements is  $f_{282} = 1064721609899145.9(1.6)(10)$  Hz. We anticipate that the largest uncertainty in the clock transition frequency is due to the atomic quadrupole shift, which is dependent on the orientation of the applied magnetic field, and which we have not corrected so far. To assist in the evaluation of the systematic uncertainty experimentally, we built a second Hg<sup>+</sup> standard. We have applied an injection-locking scheme to generate high-power 792 nm radiation as one of the fundamental sources used in sum-frequency-generation of 194 nm light. With 400 mW of 792 nm light enhanced in a cavity and 14 mW of 257 nm light, 48  $\mu$ W of the 194 nm radiation is obtained.

## Acknowledgments

The authors are grateful to T Parker, S Jefferts and T Heavner for providing the absolute frequency calibration, and to A Bartels of Giga-Optics GmbH for his valuable assistance with the femtosecond laser. We also thank R Windeler of Lucent Technologies for providing the microstructure optical fibre. UT acknowledges the support of the former Science and Technology Agency of Japan (presently Ministry of Education, Culture, Sports, Science and Technology). This work was partially supported by the Office of Naval Research.

## References

- [1] Young B C, Cruz F C, Itano W M and Bergquist J C 1999 Phys. Rev. Lett. 82 3799
- [2] Bergquist J C, Rafac R J, Young B C, Beall J A, Itano W M and Wineland D J 2001 Proc. SPIE 4269 1-7
- [3] Udem T, Huber A, Gross B, Reichert J, Prevedelli M, Weitz M and Hänsch T 1999 Phys. Rev. Lett. 82 3568
- [4] Diddams S A, Jones D J, Ye Y, Cundiff S T, Hall J L, Ranka J K, Windeler R S, Holzwarth R, Udem T and Hänsch T 2000 Phys. Rev. Lett. 84 5102
- [5] Jones D J, Diddams S A, Ranka J K, Stentz A, Windeler R S, Hall J L and Cundiff S T 2000 Science 288 635
- [6] Vogel K R, Diddams S A, Oates C W, Curtis E A, Rafac R J, Itano W M, Bergquist J C, Fox R W, Lee W D, Wells J S and Hollberg L 2001 Opt. Lett. 26 102
- [7] Udem Th, Diddams S A, Vogel K R, Oates C W, Curtis E A, Lee W D, Itano W M, Drullinger R E, Bergquist J C and Hollberg L 2001 Phys. Rev. Lett. 86 4996
- [8] Diddams S A, Udem Th, Bergquist J C, Curtis E A, Drullinger R E, Hollberg L, Itano W M, Lee W D, Oates C W, Vogel K R and Wineland D J 2001 Science 293 825
- [9] Rafac R J, Young B C, Beall J A, Itano W M, Wineland D J and Bergquist J C 2000 Phys. Rev. Lett. 85 2462
- [10] Itano W M 2000 J. Res. Natl Inst. Stand. Technol. 105 829
- [11] Dehmelt H G 1975 Bull. Am. Phys. Soc. 20 60
- [12] Bergquist J C, Itano W M and Wineland D J 1987 Phys. Rev. A 36 428
- [13] Berkeland D J, Cruz F C and Bergquist J C 1997 Appl. Opt. 36 4159
- [14] Cummings E A, Hicken M S and Bergeson S D 2001 Preprint physics/0112014
- [15] Drever R W P, Hall J L, Kowalski F B, Hough J, Ford G M, Munley A J and Ward H 1983 Appl. Phys. B 31 97
- [16] Siegman A E 1986 Lasers (Mill Valley, CA: University Science Books) pp 1129-70
- [17] Hänsch T W and Couillaud B 1980 Opt. Commun. 35 441
- [18] Parker T E 1999 Proc. 1999 Joint European Mtg on Freq. and Time Forum and the IEEE Int. Freq. Control Symp. (Switzerland: Neuchâtel) p 173
- [19] Jefferts S R, Shirley J, Parker T E, Heavner T P, Meekhof D M, Nelson C, Levi F, Costanzo G, DeMarchi A, Drullinger R, Hollberg L, Lee W D and Walls F L 2002 *Metrologia* 39 321