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Dubé, P.; Madej, A. A.; Bernard, J. E.; Humphrey, G.; Vainio, M.; Jiang, J.; Jones, D. J.

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Recent Progress on the NRC $^{88}\text{Sr}^+$ Single-Ion Optical Frequency Standard

P. Dubé, A.A. Madej,
J.E. Bernard and G. Humphrey
Institute for National Measurement Standards
NRC, Ottawa, ON, Canada
Email: pierre.dube@nrc-cnrc.gc.ca

M. Vainio
Center for Metrology and
Accreditation (MIKES)
Espoo, Finland

J. Jiang and D.J. Jones
Department of Physics and Astronomy
University of British Columbia
Vancouver, BC, Canada

Abstract—We report our recent progress made with the NRC $^{88}\text{Sr}^+$ single-ion optical frequency standard. The long-term operation of the standard was improved by actively stabilizing the cooling, repump and clearout laser sources, and by using a femtosecond fiber laser frequency comb to link the probe laser frequency to the microwave time standards. With the femtosecond fiber comb, we have demonstrated continuous operation for a period of eight days and continuous measurement of the probe laser source for three days. Micromotion shifts have been the dominant source of uncertainty in our rf Paul trap because laser beam access is only possible along one axis in the current design. With the aim of reducing to a minimum these shifts, we have constructed an endcap trap designed for minimization of micromotion along three orthogonal axes. We report on the successful trapping of single ions with this endcap trap. Since electrode contamination during trap loading can also induce micromotion shifts as a result of evolving patch potentials, we have increased the trap loading efficiency with photo-ionization.

I. INTRODUCTION

Optical frequency standards based on a trapped and laser-cooled single ion approximate closely the ideal of an isolated and unperturbed quantum system and are promising candidates for attaining the highest frequency accuracies [1]. Several ion systems have surpassed the microwave time standards in terms of both stability and reproducibility [2]–[4] and have the potential of becoming the next generation realization of the second.

At NRC we have developed for a number of years an optical frequency standard based on the trapped and laser-cooled $^{88}\text{Sr}^+$ ion. With the advent of the femtosecond laser frequency comb technology a decade ago, rapid progress was made in the characterization, measurement, and reduction of the systematic shifts that limit the ultimate accuracy of this frequency standard. For example, we found a method that cancels the electric quadrupole shift and the tensor parts of the quadratic Stark shifts [5]. On the other hand, we also found that micromotion-related shifts, namely the second-order Doppler shift and the scalar Stark shift, dominate the error budget of the $^{88}\text{Sr}^+$ ion in our current rf Paul trap apparatus [5]. These shifts are a consequence of micromotion minimization along only a single trap axis.

For the purpose of minimizing the micromotion shifts, we have assembled an endcap trap [6], designed with enough optical ports for laser beam access along three orthogonal axes and with trim electrodes for precise ion positioning. This new trap is presented in Section VI.

The stability and accuracy of any ion frequency standard depends crucially on the linewidth and stability of its probe laser system because a single-ion oscillator combined with Hz-level linewidths imposes severe limitations on the data collection rate for probe frequency corrections. The 674 nm probe laser performance is discussed in Section III.

Another important aspect of an optical frequency standard for its operation as a clockwork and for high-accuracy frequency measurements is its long-term operation. This requires reliable operation of the probe, cooling, repump, and clearout lasers, and of the femto-second laser frequency comb if a link between microwave and optical frequencies is used. We have demonstrated operation of a fiber comb for a duration of eight days and have made a new measurement of the $^{88}\text{Sr}^+$ ion frequency to confirm the stability of the optical standard over a period of several years. Stabilization of the trapping laser sources is described in Section IV and the femto-second fiber comb measurements are presented in Section V.

Finally, the expected attainable uncertainty in the frequency of the $^{88}\text{Sr}^+$ ion is discussed briefly in Section VI in the context of the new endcap trap.

II. $^{88}\text{Sr}^+$ OPTICAL FREQUENCY STANDARD

The energy levels and laser wavelengths of interest for the $^{88}\text{Sr}^+$ ion frequency standard are illustrated in the partial energy level diagram of Fig. 1. This system is described in detail elsewhere [7], [8], so only a brief overview is given here.

The $^{88}\text{Sr}^+$ optical frequency standard is based on the $5s\ ^2S_{1/2} - 4d\ ^2D_{5/2}$ electric-quadrupole allowed transition at 674 nm (445 THz). This clock transition has a natural linewidth of 0.407 Hz and its $D_{5/2}$ metastable state has a lifetime of 0.391 s [9]. The quality factor is thus $Q = 1.1 \times 10^{15}$. The fractional frequency stability of a laser locked to this $S-D$ transition can in principle reach a level of $\approx 1 \times 10^{-17}$ (5 mHz) in less than a day averaging provided that quantum

Fig. 1. Energy level diagram of $^{88}\text{Sr}^+$.

projection noise (QPN) is the dominant noise process (see Fig. 2). The narrow linewidth of the clock transition and the slow data collection rate from the single ion require an ultra-stable and ultra-narrow laser source to probe the clock transition without degradation of the ion stability.

The laser at 422 nm, red-detuned from linecenter by about half the 21.7 MHz linewidth of the $5s\ ^2S_{1/2} - 5p\ ^2P_{1/2}$ transition, is used for cooling the ion. The upper state of the cooling transition decays to the metastable $D_{3/2}$ state with a branching ratio of 1:13 [10]. To prevent interruption of the cooling process, laser radiation at 1092 nm illuminates the ion during the 422 nm cooling period to repump the ion back into the $P_{1/2}$ state.

A typical measurement cycle consists of a cooling pulse of 20 ms followed by an probe pulse of 100 ms. The fluorescence at 422 nm is monitored with a photo-multiplier tube during the cooling period to detect whether the ion was promoted or not to the $D_{5/2}$ metastable state by the preceding probe laser pulse. When the ion is promoted to the metastable state, the fluorescence is interrupted. We refer to these events as quantum jumps. They are used to either record spectra of the clock transition or to lock the 674 nm laser frequency to the clock transition linecenter [11].

The spectrum of the S–D transition is composed of ten magnetic sensitive Zeeman components, split symmetrically from the clock transition linecenter ν_0 . The linear Zeeman shift is cancelled by measuring a symmetric pair of components and by taking the average frequency. The electric quadrupole shift and the tensor parts of the quadratic Stark shifts can also be cancelled if the average of transitions that sample all of the Zeeman sub-levels of the $D_{5/2}$ state are averaged [5], [12]. Practically this is achieved by averaging the frequencies from 3 pairs of Zeeman components. We use this method when making measurements of the ion frequency.

The frequency stability of the ion is slightly degraded by the long natural lifetime of the metastable $D_{5/2}$ state. As soon as a quantum jump is detected by the fluorescence signal, a

laser at 1033 nm can be used to return the ion back to the ground state via the short-lived $P_{3/2}$ state. Although we have tested the clearout laser, this feature was not used in the data presented in this paper.

All our measurements so far have been performed with an rf Paul trap apparatus [5], [7]. Laser beam access is along a single axis which prevents complete cancellation of the micromotion. The residual micromotion shifts are -6 ± 13 Hz (3×10^{-14}). They were evaluated in an extensive series of measurements where the shifts were measured as a function of the quantization axis defined by an applied magnetic field [5]. A new endcap trap described in Section VI was built to reduce these micromotion shifts by several orders of magnitude.

Small micromotion shifts can also appear after minimization because of patch potentials created by strontium atoms deposited on the trap electrodes during loading. These patch potentials can be reduced significantly if the loading process is made more efficient. We have found that photo-ionization loading allowed for a decrease in the strontium flux by three orders of magnitude compared to electron bombardment loading. Photo-ionization is realized with a two-step process where the neutral strontium atoms are first excited to the $5p\ ^1P_1$ state from the $5s\ ^1S_0$ ground state with laser radiation at 461 nm, followed by excitation to an auto-ionizing state with 405 nm laser radiation [13].

III. PROBE LASER SYSTEM

The probe laser light at 674 nm is produced with a commercial extended-cavity diode laser head driven with low-noise home-made electronics. The linewidth of the free-running laser is on the order of 1 MHz. The laser is pre-stabilized on a Fabry-Perot cavity that has a linewidth of 110 kHz. The error signal between the laser and the cavity is derived using the Pound-Drever-Hall (PDH) technique [14], and applied, after some signal conditioning, to both the current and the piezo-electric translator (PZT) of the diode laser. This first stage reduces the laser linewidth to $\simeq 50$ Hz relative to a resonance of the cavity.

The output from this first stage is then stabilized using an ultra-stable cavity made with ultra-low thermal expansion coefficient glass (ULE). The spacer length is 25 cm, the cavity finesse is 160 000 and the resonance linewidths are 3.7 kHz wide. The light emerging from the first cavity is locked to the ultra-stable cavity using the PDH technique. The conditioned error signal between the two cavities is fed back to an acousto-optic modulator (AOM) located after the first cavity output for fast frequency corrections (150 kHz), and to a PZT-actuated mirror of the first cavity for drift corrections. The linewidth of the laser radiation relative to the reference ULE cavity is less than 1 mHz. The actual linewidth is much larger as a result of thermal noise in the mirrors and mirror coatings [15], and mechanical noise from the environment. We have observed 5 Hz Fourier-transform-limited linewidths in high-resolution scans of a Zeeman component of the clock transition [16].

As a measure to reduce to a minimum the frequency drifts, the reference ULE cavity is stabilized to its temperature of

zero thermal expansion coefficient of $5.45(1)^\circ\text{C}$. The present drift rate of this cavity is only 12 mHz/s (see Fig. 3). This rate is still slowing down with time.

The stability of the probe laser locked to the ULE cavity was studied by measuring its frequency with the ion clock transition. Figure 2 shows an Allan deviation plot of the data. The stability reaches a level of 5×10^{-16} after 3000 s of averaging time, following closely the expected QPN of the ion for the present experimental conditions. Note that the relative stability between the probe laser frequency and the ion transition is nearly an order of magnitude better than the best $^{88}\text{Sr}^+$ ion frequency measurement [17]. With the present QPN limit, we can expect to find the ion linecenter with a precision of 1×10^{-16} after 19 hours of averaging. The ion stability can be improved by increasing the quantum jump rate. In our experiment this can be achieved by selectively pumping the ion into the lower level of the probed transition, by using the clearout laser, and by cooling the ion to a lower temperature.

Fig. 2. Allan deviation of the probe laser measured by using the ion S-D transition as the frequency reference. The *short-dashed line* is a $1/\sqrt{\tau}$ fit through the data ($2.6 \times 10^{-14}/\sqrt{\tau}$), the *long-dashed line* is the theoretical dependence of the Allan deviation for the present experimental conditions assuming a purely quantum-projection-noise (QPN) limited measurement ($1.9 \times 10^{-14}/\sqrt{\tau}$). The *solid line* represents the Allan deviation limited by the QPN when all the parameters are optimized ($2.6 \times 10^{-15}/\sqrt{\tau}$). The *horizontal dotted line* is the estimated Allan deviation caused by the thermal noise level of the 25 cm long Fabry-Perot cavity.

The reader is referred to the literature for a more complete description of this laser system and its performance [16].

IV. TRAP LASERS STABILIZATION

The lasers at 422 nm, 1092 nm and 1033 nm can be operated free-running for short periods of time because the linewidths of the transitions they drive are 22 or 24 MHz wide. For reliable, long-term operation, and for optimum performance it is necessary to lock each of these lasers to obtain MHz-level stability over days. In this Section, we describe the stabilization methods used to control these laser sources.

A. 422 nm laser

The 422 nm source is a commercial extended cavity GaN diode laser. It has a free-running linewidth of ≈ 10 MHz. The linewidth is reduced to 2.4 MHz with stabilization onto a

low-finesse Fabry-Perot cavity. The spectrally-narrowed light is then stabilized to the Doppler-free $5s^2S_{1/2}(F'' = 2) - 6p^2P_{1/2}(F' = 3)$ hyperfine transition in ^{85}Rb [18]. This transition is red-detuned from the $^{88}\text{Sr}^+$ cooling transition by 440 MHz. A double-pass AOM provides precise tuning of the 422 nm frequency with respect to the cooling transition. The 422 nm laser routinely stays locked to ^{85}Rb for a few days, until a mode-hop occurs.

B. 1092 nm and 1033 Lasers

The 1092 nm radiation is produced with an ytterbium-doped, diode-pumped fiber laser while the 1033 nm radiation is from an extended cavity diode laser. Both lasers are stabilized to a polarization-stabilized He-Ne laser using a Fabry-Perot transfer cavity [8]. The infrared lasers remain locked for several days with this robust system which has a capture range of several hundred MHz. The rms frequency fluctuations of the stabilized 1092 nm and 1033 nm lasers are, respectively, 0.5 MHz and 1 MHz.

V. FEMTO-SECOND FIBER FREQUENCY COMB

The femto-second fiber comb used in the present series of measurements is a stretched-pulse erbium-doped ring fiber oscillator that uses nonlinear polarization rotation as the mode-locking mechanism. It produces 120-fs pulses at a rate of 100 MHz. There are two external branches, each with a chirped-pulse erbium-doped fiber amplifier and with a highly nonlinear fiber. One of the branches is used for offset frequency locking. The other branch is optimized for super-continuum generation at 1348 nm for frequency-doubling in a periodically-poled MgO:LiNbO_3 crystal to 674 nm. A detailed description of this fiber comb is given elsewhere [19], [20].

The operation of the fiber comb as a clockwork linking the microwave time standards to the ion optical frequency was tested by measuring the 674 nm probe laser frequency over a period of several days. The results are shown in Fig. 3. The fiber comb remained locked without adjustment (repetition rate and offset frequency) to the microwave standards for the entire duration of the measurement period of eight days. Figure 4 shows the Allan deviation of the probe laser calculated with the last three days of Fig. 3. The $1/\tau$ dependence reflects the white phase noise behavior of the maser. Since the measurement of the probe laser has a $1/\tau$ dependence with values similar to those of the maser, the probe laser can be considered to be phase stable relative to the stability level of the maser. Variations in the ULE drift rate account for the rise in the Allan deviation at averaging times longer than 1000 s. The Allan deviation measured with the $^{88}\text{Sr}^+$ clock transition as reference shown in Fig. 2 reached lower levels for two reasons; the ion is a more stable reference than the maser and the ULE drift rate was removed in the analysis with a piecewise linear function [16].

The $^{88}\text{Sr}^+$ clock transition frequency was measured with the femtosecond fiber comb in another series of measurements. As in the long-term measurement described above, the fiber comb measures the absolute frequency of the probe laser

Fig. 3. Probe laser frequency measured with the erbium-doped fiber laser frequency comb referenced to the NRC rf atomic clocks. The observed linear drift of 11.9 mHz/s is caused by the isothermal creep of the reference cavity ULE spacer. The missing data is due to a loss of lock of the probe laser to its reference cavity.

Fig. 4. The solid line is the Allan deviation of the probe laser calculated from the final three days of the data shown in Fig. 3 after removal of the linear drift. The Allan deviation of the reference maser is shown as solid dots for comparison. Both curves follow the characteristic $1/\tau$ dependence of the maser phase noise up to an averaging time of $\tau \approx 200$ s.

with respect to the microwave time signal from a maser source. Simultaneously, the offset of the probe laser frequency from three pairs of Zeeman components is measured. The average of these component linecenters results in an ion frequency measurement that is corrected for the linear Zeeman shift, the electric quadrupole shift and the tensor terms of the quadratic Stark shifts [5], [7], [12]. The absolute ion frequency is found by combining the comb measurements with the offset between the probe laser and the ion. A value of $444\,779\,044\,095\,477.6$ Hz is obtained, with a statistical uncertainty of 0.8 Hz. The maser uncertainty was 1.1 Hz for this measurement and the uncertainty associated with the transfer of the maser signal to the comb is estimated at 0.3 Hz. The total standard uncertainty estimate is 1.4 Hz [20]. The data is shown in Fig. 5.

This result obtained in 2009 can be compared to a mea-

Fig. 5. Measurement of the $^{88}\text{Sr}^+$ ion S-D clock transition frequency with an erbium-doped fiber laser frequency comb. A drift of +14 mHz/s, due to the probe laser reference cavity, was subtracted. The resulting frequency is $444\,779\,044\,095\,477.6 \pm 1.4$ Hz. Note that this frequency is not corrected for ion systematic shifts except the electric quadrupole shift and the tensor terms of the quadratic Stark shifts.

surement made in 2005 under similar conditions, that is before it was corrected for micromotion shifts and blackbody radiation shifts [5]. This value is $f_{2005}(\text{not corrected}) = 444\,779\,044\,095\,478.0 \pm 4.3$ Hz. The excellent agreement with the 2009 result, $f_{2009} - f_{2005} = -0.4 \pm 4.5$ Hz, demonstrates a high level of stability of the standard over a period of several years despite net micromotion shifts of 6 Hz.

VI. NEW ION TRAP

An endcap trap [6] was built to give access to the cooling laser beam along three orthogonal axes for minimization of the micromotion in three dimensions. The endcap electrodes are made with 0.50 mm diameter molybdenum wire and are separated by 0.54 mm. Their end faces were polished to a mirror finish. The shield electrodes were made from 2.0 mm diameter tantalum rods. Trim electrodes, including the endcap electrodes for the axial direction, are provided to control the ion position along three orthogonal axes. The micromotion signal will be obtained from the modulation of the 422 nm fluorescence due to the first order Doppler shift at the rf potential applied to the electrodes [21]. It is expected that micromotion shifts will be reduced to a level of 10^{-17} or less [17].

An aspherical lens with a numerical aperture of 0.7 was mounted inside the vacuum chamber for efficient collection of the fluorescence sent to a photo-multiplier tube (PMT). Opposite the PMT, another port is reserved for a photon-counting camera which will be used for trap diagnostic and studies involving ion clouds which are not efficiently detected by the restricted field of view of the PMT. The light collection systems of the camera and PMT are equipped with a band-pass interference filter at 422 nm for optimum rejection of background light from the other laser sources.

We have successfully trapped single ions with this new trap. Figure 6 shows quantum jumps from a single ion observed in the fluorescence signal detected by the PMT. The high contrast between the bright and dark periods ensures that the quantum

jumps are detected with nearly 100% efficiency. Figure 7 is a spectrum of the S-D transition that shows the ten Zeeman components. The splitting observed is caused by the ambient magnetic field as there were no magnetic shields around the trap for these initial tests. The linewidth of the probe laser was purposely broadened to record this spectrum.

Fig. 6. $^{88}\text{Sr}^+$ fluorescence at 422 nm observed with the photo-multiplier tube. The dark periods correspond to times when the ion is in the metastable $^2D_{5/2}$ state following excitation by a probe laser pulse at 674 nm.

Fig. 7. Low-resolution quantum jump spectrum of the S-D transition in $^{88}\text{Sr}^+$ recorded with the new end-cap trap. An ambient magnetic field of about 25 μT caused the observed splittings in the Zeeman components.

With the new trap, the micromotion shifts are expected to be reduced from 3×10^{-14} to 10^{-17} or less. The remaining dominant source of uncertainty will likely be the blackbody-radiation (BBR) shift. According to a recent theoretical calculation, the BBR shift for the $5s\ ^2S_{1/2} - 4d\ ^2D_{5/2}$ transition is 0.250 ± 0.009 Hz at 300 K [22]. The fractional frequency uncertainty, limited by the knowledge of the BBR shift, is thus 2×10^{-17} . It should therefore be possible to reduce the total $^{88}\text{Sr}^+$ ion systematic shifts to a fractional frequency uncertainty of a few parts in 10^{17} .

VII. CONCLUSION

We have reported in this paper several improvements made to the $^{88}\text{Sr}^+$ ion optical frequency standard aimed at increasing its accuracy, stability, and long-term operation. All the lasers sources are now frequency-stabilized. The main limitation to long-term operation at present is the lifetime of the ion in the

trap. This lifetime varies from a few hours to a day and is thought to be limited by collisions with background hydrogen gas even though the background pressure is only 10^{-8} Pa in the vacuum chamber.

We have demonstrated operation of the femto-second fiber comb for a period of eight days in a measurement of the probe laser frequency. These results confirm the suitability of the fiber comb as a clockwork to link optical and rf frequencies. The fiber comb was also used to make a new measurement of the ion frequency in the rf Paul trap apparatus. The systematic shifts were not evaluated. The frequency obtained was found to be in excellent agreement (-0.4 ± 4.5 Hz) with a previous measurement made four years earlier. This comparison shows that the rf Paul trap apparatus provides stable operation at the 10^{-14} level over long periods of time despite the micromotion shifts.

The probe laser stability, measured using the ion as a reference, was found to reach a stability level of 5×10^{-16} after 3000 s. The Allan deviation followed closely the stability expected for the QPN of the measurement. The actual performance of the probe laser system is suitable to measure the ion linecenter with an uncertainty of 10^{-16} or better. Improvements to the ion quantum jump rate are expected to make feasible the determination of the ion linecenter with a precision of 10^{-17} with an averaging time on the order of one day.

We have successfully trapped single ions in a new endcap trap designed for the minimization of micromotion along three orthogonal axes. This modification compared to our rf Paul trap apparatus is expected to reduce the micromotion-related shifts by more than three orders of magnitude, to a level of 10^{-17} or less. Finally, a new determination of the BBR shift with a fractional frequency uncertainty of 2×10^{-17} suggests that the total $^{88}\text{Sr}^+$ systematic shifts can be reduced to a fractional frequency uncertainty of a few parts in 10^{17} .

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