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Systematic shift evaluation of the NRC $^{88}\text{Sr}^+$ single-ion optical frequency standard to a few parts in 10^{-17}

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Abstract—A new strontium ion trap of the endcap design was built recently at the National Research Council of Canada for better control of the micromotion shifts. The uncertainty caused by these shifts has been reduced by more than four orders of magnitude compared to the original NRC trap, down to a level of $\approx 10^{-18}$. In this paper we discuss the evaluation of micromotion shifts and of several other sources of importance to optical frequency standards based on single ions. The total fractional frequency uncertainty of the new strontium ion trap system is estimated to be $\approx 2.2 \times 10^{-17}$, limited by the black-body radiation shift. A preliminary measurement of the clock transition frequency with a fractional uncertainty of 2×10^{-15} , limited by the accuracy of the maser reference, is also presented.

I. INTRODUCTION

Optical frequency standards based on either trapped ions or neutral atoms in optical lattices have made impressive progresses since the femtosecond laser frequency comb technology has become an accessible and reliable tool a little more than a decade ago [1]. The femto-comb technology breakthrough has overcome the major obstacles that used to burden optical frequency standards: the reliable comparison of optical frequencies from different atomic species and the measurement of the absolute frequency of an optical standard with respect to the microwave cesium clocks that define the SI second.

The rapid improvement of ultra-stable Fabry-Perot cavities in several labs during the last decade [2]–[7], with Hz and sub-Hz-level linewidths, has also been essential for the progress observed in the frequency stability of optical frequency standards. The improvements in stability and accuracy in frequency measurements have accelerated the ability to control and evaluate the systematic shifts that affect the various optical frequency standards. Unprecedented accuracy levels have been reported in the last few years by various groups with systematic shifts evaluations in the 10^{-15} to 10^{-17} range [8]–[11], and even below the 10^{-17} level in one case [12].

During the past year, we have reduced by a few orders of magnitude the total uncertainty on the systematic shifts of the NRC $^{88}\text{Sr}^+$ optical frequency standard, with a new uncertainty budget in the low 10^{-17} level. For comparison,

our previous trap system had an uncertainty of 3.4×10^{-14} [13], dominated by the micromotion shifts. In the following sections, we describe the new endcap trap that provides better control of micromotion and discuss the main shifts that are of concern for the $^{88}\text{Sr}^+$ ion. The small systematic shift uncertainties of our optical frequency standard has motivated a new measurement of the $5s\ ^2S_{1/2} - 4d\ ^2D_{5/2}$ transition frequency and preliminary results are reported in this paper.

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

Figure 1 shows a partial energy level diagram of $^{88}\text{Sr}^+$ with the transitions used for cooling and detection, for repumping, and for probing the clock transition. The fluorescence scattered by the ion at 422 nm is collected with a photon-counting photomultiplier tube to derive the quantum jump (electron-shelving) rate signal used in spectroscopic studies and for locking to the $5s\ ^2S_{1/2} - 4d\ ^2D_{5/2}$ clock transition. A high-sensitivity camera also collects the 422 nm fluorescence from the trap for imaging the ion motion. It is used as a first step to minimize micromotion as described in Section III.

The S – D clock transition has ten allowed transitions which we call Zeeman components. All of these are magnetic sen-

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

The micromotion shifts are Stark shifts and second-order Doppler shifts caused by the motion of the ion driven by the trap ac field. The main causes are displacement of the ion from the trap center, secular motion, and phase difference between the endcap electrodes [24].

Micromotion is first minimized with the camera. For this adjustment, the trap voltage is modulated at a rate of ≈ 1 Hz to create alternation between shallow and deep trapping potentials. The trim voltages are then adjusted until no discernable motion of the ion can be observed with the camera. The camera only provides a view of the ion in a plane perpendicular to the camera axis, but this step greatly reduces the adjustments required with the time-consuming sideband method.

The second method, based on sideband intensity, is used for an accurate and quantitative evaluation of the shifts. The micromotion is determined by selecting a single Zeeman component of the clock transition and by measuring the sideband-to-carrier intensity ratios along three orthogonal axes. Neutral density filters are used to match approximately the carrier and sideband signals in each direction. The sum of the ratios is proportional to the total micromotion and is used to calculate the Stark and second-order Doppler shifts.

Initially, the micromotion shifts were dominated by a 0.75° phase difference between the endcap voltages. A series of experiments was then conducted to determine the optimum phase shift correction to apply, by trimming wire lengths, to minimize the phase difference. Once this step was successfully completed, we could reach levels of typically 2×10^{-17} for both scalar Stark shifts and second-order Doppler shifts. This phase adjustment only needs to be performed once.

Ion centering in the trap potential is achieved by adjusting the voltages on the trim electrodes to minimize the sidebands. This exercise is repeated every time the trap is loaded. Micromotion is sometimes measured at the end of a long measurement run to confirm that it has not changed appreciably.

The ion temperature was measured by the sideband intensity method described above, but this time using the secular sidebands. Averaged over the three principal axes of secular motion, the ion temperature in the endcap trap is $1.8(7)$ mK. The secular motion at this temperature causes micromotion shifts of magnitude 3×10^{-18} .

The scalar Stark shift and the second-order Doppler shift are correlated as they both depend on the rms value of the trap electric field [13], [24]. With the recent improvement in the accuracy of the polarizabilities of $^{88}\text{Sr}^+$ [25], it is now possible to reduce the uncertainty on those combined micromotion shifts by another factor of 25. The residual shifts of 2×10^{-17} obtained with our trap are therefore further reduced to a level of 1×10^{-18} .

The tensor part of the Stark shift is also reduced to very low levels by the minimization of micromotion. This shift varies with the magnetic sublevel quantum number; it is largest for the symmetric Zeeman pair that connects to the $m_J = \pm 5/2$ sublevels of the $^2D_{5/2}$ state. For this Zeeman pair, the tensor

sitive and a symmetrically-shifted pair of such components

must be measured to cancel the first order Zeeman shift [14]. As described in Section IV-C, by measuring multiple pairs of symmetric components connecting to different $^2D_{5/2}$ magnetic sub-levels, the electric quadrupole shift and the tensor part of the Stark shift can also be cancelled [13], [15]. The highest Fourier-transform-limited resolution we have achieved on a Zeeman component is $4.1(3)$ Hz, corresponding to a line Q of 1×10^{14} [5]. When the laser is locked to the ion clock transition linecenter for a frequency measurement, the linewidth is increased to ≈ 10 Hz by shortening the pulse length to 100 ms. The shorter pulses allow for an increase in the quantum jump rate and a faster servo loop response for tracking the mHz/s-level drift rate changes of the reference ULE cavity during lock runs.

For a more detailed description of the frequency standard and experimental apparatus, the reader is referred to earlier work [5], [16]–[19].

III. ENDCAP TRAP

The previous NRC ion trap system was based on the Paul design and the vacuum chamber had only one optical access port for monitoring the ion motion [16]. This has limited our ability to measure and minimize micromotion and has resulted in large uncertainties on the frequency shifts [13]. A new trap of the endcap design [20] and a vacuum chamber with three orthogonal optical access ports were therefore built recently for proper control of the ion micromotion [19].

The trap is built with endcap electrodes made with 0.50 mm diameter molybdenum wires. Their end faces, polished to a mirror finish, are separated by 0.54 mm. The shield electrodes slid over the endcap wires were made from 2.0 mm diameter tantalum rods. Alumina tubes are inserted between the shields and endcaps for electrical insulation and mechanical centering. The micromotion is minimized by controlling the ion position in the trap with voltages on trim electrodes. Two are orthogonal to the trap with voltages on the radial plane and the other is realized by applying a differential voltage on the endcap electrodes for axial control.

A concern for the accuracy of ion optical clocks is the evolution of patch potentials on the electrodes surfaces, created at loading time, which may lead to micromotion changes during a measurement. Deposition of strontium on the electrodes is minimized by using photo-ionization loading [21], [22].

IV. FREQUENCY SHIFTS

There are many sources of frequency shifts in ion optical frequency standards such as Stark shifts, second-order Doppler shifts, electric-quadrupole shifts, blackbody radiation shifts, light shifts, collisional shifts, etc. [13], [16], [23]. These shifts must be evaluated for each trap system and vary greatly depending on the ion species. Except for the electric quadrupole shift, the shifts discussed below are the main contributors to the uncertainty budget of the strontium ion in the NRC trap.

Stark shift is 2.2×10^{-17} for the level of minimization that we achieve. Although this shift cannot be compensated by the second-order Doppler shift because of its dependence on $m_{J'}^2$ and quantization axis, it is cancelled by the method described in Section IV-C.

B. Blackbody Radiation Shift

The blackbody radiation (BBR) shift is the dominant source of uncertainty for the $^{88}\text{Sr}^+$ ion. The uncertainty on this shift depends on the accuracy of the static scalar polarizabilities of the clock transition and on the accuracy of the model that estimates the fields at the ion.

The BBR shift is proportional to the difference between the scalar polarizabilities of the upper and lower states of the clock transition. This difference was recently calculated with an accuracy of 3.6% [25].

For modeling the blackbody radiation field at the ion, we have first measured experimentally the temperature increase of the endcap electrodes with trap voltage. This was achieved by using a second trap in vacuum, almost identical to the trap used for the optical frequency standard. A small thermistor (1 mm diameter) inserted between, and in contact with, the endcaps was then used to monitor the increase in electrode temperature with applied voltage. For a voltage of 214 V amplitude, typical of the experiments reported here, it was found that the temperature increased by 32°C above room temperature. We give a generous uncertainty of 10°C to this measurement because we didn't correct for a possible bias caused by the thermistor.

The model for the BBR field at the ion takes into account the radiation from the chamber walls, measured with calibrated thermistors, the radiation from the electrodes including multiple reflections off the electrode surfaces, and finally the radiation from the vacuum chamber reflected off the electrode surfaces. As mentioned earlier, the molybdenum endcap electrodes which present a large solid angle to the ion, were polished to a mirror finish. For this material, the surface emissivity near room temperature is 0.06(3) [26].

The model is preliminary as some sources of heat were neglected in the estimate. For example, the cavity between the endcap and shield electrodes was considered to be at room temperature in the current model. However, it is warmer than room temperature because of heat transfer and direct heating of the alumina material due to its loss tangent parameter. Although there is no direct optical path between that cavity and the ion, multiple reflections off the endcaps surfaces allow for a small solid angle to reach the ion.

Table I summarizes the contributions to the BBR shift uncertainty budget. The low endcap emissivity greatly reduces the sensitivity to endcap temperature. The budget is dominated by the uncertainty in the difference between the scalar polarizabilities of the upper and lower states of the clock transition, $\Delta\alpha_0 = \alpha_0(D_{5/2}) - \alpha_0(S_{1/2})$. The estimated BBR shift is 0.243(9) Hz. Refinements to the model will be considered although they are not expected to make significant changes to the total BBR uncertainty.

TABLE I
BBR SHIFT UNCERTAINTY BUDGET

Parameter	Value	Unc.	$(\Delta f_{\text{BBR}}/f) \times 10^{-18}$
Endcaps emissivity	0.06	0.03	2.5
Endcaps temp. (°C)	55	10	2.1
Wall temp. (°C)	23.0	0.4	2.9
$\Delta\alpha_0$ (a.u.)	-29.3	1.0	19
Total			20

C. Electric Quadrupole Shift

The electric quadrupole moment interacts with the electric field gradient to cause the largest frequency shifts in our $^{88}\text{Sr}^+$ ion trap when one considers only individual pairs of symmetric components. Fortunately, these shifts can be “perfectly” cancelled by the method described below when two or three symmetric pairs of components are measured [13], [15].

To first order in perturbation theory, the frequency shift Δf_Q of a magnetic sublevel $m_{J'}$ is given by [15], [23], [27]:

$$\Delta f_Q = \frac{1}{4} \nu_Q (3 \cos^2 \theta - 1) \left[m_{J'}^2 - \frac{J'(J'+1)}{3} \right]. \quad (1)$$

ν_Q is a characteristic frequency proportional to the electric field gradient and the electric quadrupole moment of the $D_{5/2}$ state. θ is the angle between the electric field gradient and the quantization axis defined by the applied magnetic field. For the $D_{5/2}$ state, $J' = 5/2$ and the right-hand term in the square brackets evaluates to 35/12. Δf_Q also describes the linecenter shifts of symmetric Zeeman pairs because the two components of any symmetric pair connect to magnetic quantum numbers $m_{J'}$ that have the same magnitude but opposite signs in the $D_{5/2}$ state.

The cancellation method uses the dependence of the frequency shift on the magnetic sublevels. It is easy to see that $\Delta f_Q = 0$ in (1) for $m_{J'}^2 = J'(J'+1)/3$. This suggests a method where the quadrupole-shift-free ion frequency at $m_{J'}^2 = J'(J'+1)/3$ is calculated by interpolation from the frequencies of two pairs of components that connect to different values of $m_{J'}^2$. Alternatively, one notes that the sum of the term in square brackets in (1) over all the sublevels evaluates to zero:

$$\sum_{m_{J'}=-J'}^{J'} \left[m_{J'}^2 - \frac{J'(J'+1)}{3} \right] = 0. \quad (2)$$

For $^{88}\text{Sr}^+$, three pairs of components are required to connect to all of the upper state magnetic sublevels (see Fig. 1). The electric-quadrupole-shift-free clock transition frequency is simply obtained by averaging the three linecenters. Both methods give the same results, but one may be preferred over the other for various reasons. For example, the cycling time through the components is faster with the interpolation method as only two pairs of components need to be measured. On the other hand, the measurement of three pairs of components has

Fig. 2. Electric quadrupole shift from the unperturbed S - D line center, Δf_Q , as a function of the upper state magnetic sub-level quantum number m_J^2 . The small error bars represent the statistical uncertainty in the line center of each symmetric pair of components. For $^{88}\text{Sr}^+$, the electric quadrupole shift is cancelled at the intercept of $m_J^2 = 35/12$. The labels Z , B_0 , B_1 , and B_2 indicate different magnetic field directions. The trap is usually operated at the magnetic field B_0 . The largest shifts are obtained with B_2 , a magnetic field aligned with the electric field gradient of the ion trap.

the advantage of providing a diagnostic for parasitic shifts, such as line contamination from secular sidebands.

Figure 2 is a plot of Δf_Q as a function of m_J^2 . The lines with different slopes illustrate the effect of changing the angle θ by means of the magnetic field direction. The data labeled B_0 , B_1 , and B_2 were measured with three pairs of components. For those the average was used to determine the clock transition line center, $\Delta f_Q = 0$ on the graph. For the line labeled Z , only two pairs were measured and the interpolation method was used.

The electric quadrupole shift cancellation is performed continuously in our experiments; the servo system measures each pair of components in a continuously repeated sequence. Unless there are important drifts in the electric quadrupole shift from cycle-to-cycle that could challenge the locking servo, the error is insignificant. We have analyzed some data to estimate the drift rate of the electric quadrupole shift in the new trap and found it undetectable at the $\mu\text{Hz/s}$ level. Therefore we consider the uncertainty in the cancellation to be negligible ($> 10^{-18}$). The measurement uncertainties on the different pairs of components does not appear in this analysis as they are reflected in the line center measurement uncertainty. The tensor part of the Stark shift has the same dependence on m_J^2 as the electric quadrupole shift [13], [15]. Consequently, it is also cancelled by the methods described above and its uncertainty is also considered negligible.

D. Gravitational Redshift

The height of a benchmark above the geoid, on the floor near the ion trap, was determined recently by Natural Resources Canada (NRCAN) with an accuracy of 5 cm [28]. Adding the vertical distance between the benchmark and the ion, the total height of the $^{88}\text{Sr}^+$ ion frequency standard above the geoid is

95.36(5) m, giving a gravitational redshift of 4.6279(24) Hz. This measurement provides a knowledge of the gravitational redshift with a fractional frequency uncertainty of 5.4×10^{-18} . Since the measurement was made with respect to a reference GPS antenna permanently mounted on the roof of the building, future knowledge of the height of the ion above the geoid can be updated with revised heights of the antenna. Also, when a more accurate geoid model becomes available, it will be a simple matter to revise the ion gravitational redshift. The antenna has been measured to move upwards with a velocity of approximately 3 mm per year due to local ground movement.

E. Uncertainty Budget

A simplified uncertainty budget for the NRC $^{88}\text{Sr}^+$ ion optical frequency standard is presented in Table II. The sources of uncertainty discussed so far are included, except for the electric quadrupole shift and the tensor Stark shift as they are considered to have negligible contributions.

TABLE II
 $^{88}\text{Sr}^+$ UNCERTAINTY BUDGET

Source	Shift (Hz)	Unc. (Hz)
Black body shift	0.243	0.009
Gravitational redshift	4.6279	0.0024
Micro-motion shifts	0.0	0.001
1092-nm ac Stark shift	-0.002	0.001
2nd-order Doppler (Thermal)	-0.0013	0.0005
Total	4.868	0.010

Also included in the table is the ac Stark shift caused by the 1092 nm repumper light that is incompletely turned off by the AOM used as a chopper (30 dB attenuation). If that source of uncertainty were to become an issue, a mechanical chopper could be used instead of the AOM. The ac Stark shift from the 674 nm laser light is 0.013(6) mHz, and the second order Zeeman shift is 95.51(3) μHz for a magnetic field of 2.025(3) μT . The equations used to calculate these shifts are given in [16]; the frequency shifts that depend on the magnetic quantum numbers must be averaged over all the possible m_J^2 to account for the averaging method now used to cancel the electrical quadrupole shift and the tensor Stark shift. The total estimated fractional frequency uncertainty for the $^{88}\text{Sr}^+$ ion in the new trap is thus 2.2×10^{-17} .

V. FREQUENCY MEASUREMENT

The very low shift uncertainty of our new $^{88}\text{Sr}^+$ ion frequency standard system has motivated a new absolute frequency measurement of the ion clock transition. An example labeled “Maser vs Ion” is the Allan deviation obtained by comparing the ion to the maser with a femto-second laser frequency comb [30]. The curve labeled “ULE vs Ion” shows the Allan deviation of the probe laser (ULE cavity) compared to the ion line center during a frequency measurement. It is

Fig. 3. Allan deviations as a function of averaging time τ . The curve labeled “Maser vs Ion” is the Allan deviation of the ion frequency measured with a hydrogen maser. It is determined by the stability of the maser source. The curve labeled “ULE vs Ion” is the Allan deviation of the reference ULE cavity frequency compared with the ion frequency during a lock run. The drift of the cavity was fitted with a third-order polynomial to reveal the lock stability, $1 \times 10^{-14}/\sqrt{\tau}$, shown by the black dashed line. The estimated ULE cavity thermal noise is shown as the horizontal dashed line (2.6×10^{-16}).

to be noted here that a third-order polynomial was used to compensate for the changes in the cavity drift rate to reveal a lock stability of $1 \times 10^{-14}/\sqrt{\tau}$.

The stability of the lock to the ion linecenter is almost ten times better than that of the maser, so the “Maser vs Ion” curve is determined by the maser stability. It reaches a level of 6×10^{-16} after 10 000 s.

The lock stability has been improved by a reduction of the ion temperature from ≈ 14 mK in the previous trap to 1.8(7) mK in the new endcap trap. Figure 4 shows the increase in excitation probability at lower temperatures. Further improvements are expected with state-preparation of the ion and with a laser to “clear-out” the $^2D_{5/2}$ state before each probe pulse.

Fig. 4. Excitation probability of a magnetic sublevel in the $^2D_{5/2}$ state as a function of probe pulse length. The filled circles are data points. The solid green line is a simulation obtained by solving the optical Bloch equations for an ion temperature of 1.8 mK. The red line is a simulation for a 14 mK temperature showing the decrease in the maximum excitation probability with temperature due to a loss of coherence. The maximum possible excitation probability is 0.5 for the current data as the ion was not state-prepared into the probed magnetic sub-level of the $^2S_{1/2}$ state before each probe pulse.

Figure 5 is a plot of recent absolute frequency measurements of the $^{88}\text{Sr}^+$ ion S - D transition as a function of the Modified Julian Day (MJD). The maser offsets from the SI second were calculated from the *Circular T* reports of BIPM. The uncertainty on the mean frequency of 0.9 Hz (2×10^{-15}) is that of the maser with respect to the SI second.

The ion frequency uncertainty is negligible at that level. Nevertheless, it should be mentioned that there is an additional source of uncertainty, not discussed in Section IV-E, at the $\approx 10^{-16}$ level associated with the current lock algorithm. It is caused by the assumption that the cavity drift rate is constant during a measurement run. A modified algorithm [31] should be able to reduce this effect to $< 10^{-17}$ given the small and predictable drift rate of our ULE cavity of typically (10 ± 2) mHz/s.

Fig. 5. Preliminary measurement of the $^{88}\text{Sr}^+$ S - D clock transition frequency. The blue diamonds are absolute frequency measurements corrected for the systematic shifts reported in Table II and the maser offset from the SI second. The error bars were derived from the stability (Allan deviation) of each measurement. The green dashed line, the green circle and the red square are located at the weighted mean of the data. The uncertainty in the maser frequency is shown with the error bars on the green circle (0.9 Hz), while the uncertainty in the mean by the error bars on the red square (0.14 Hz). The frequency measurement made by the National Physical Laboratory in 2004 [8] is shown as a black pentagon.

A preliminary value for our new measurement of the $^{88}\text{Sr}^+$ ion S - D transition frequency is:

$$f_0 = 444\,779\,044\,095\,485.6 \pm 0.9 \text{ Hz.}$$

This new value is in good agreement with a measurement made by National Physical Laboratory (NPL) with a cesium fountain [8] as shown in Fig. 5.

VI. CONCLUSION

The progress made in the control and understanding of the systematic shifts in the newly built NRC $^{88}\text{Sr}^+$ ion trap has allowed for a decrease in the total uncertainty by three orders of magnitude compared to our previous trap system. The main improvements were realized after the micromotion shifts were decreased to levels of 10^{-18} . Combined with an improved knowledge of the BBR shift and the cancellation method that eliminates the electric quadrupole shift and the

tensor Stark shift, the total uncertainty budget for the $^{88}\text{Sr}^+$ ion is now estimated at 2.2×10^{-17} , an order of magnitude better than a state-of-the-art cesium fountain clock. A preliminary measurement of the clock transition frequency with the new trap was also presented, limited in accuracy by the uncertainty on the maser frequency to a level of 2×10^{-15} . The value obtained is in good agreement with previous measurements from the NPL [8] and from the NRC [13]. It is expected that this new measurement will help reduce the uncertainty on the recommended value for the $S-D$ transition frequency of $^{88}\text{Sr}^+$ as a secondary realization of the second.

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