Agreement between two ⁸⁸Sr⁺ optical clocks to 4 parts in 10¹⁷

G. P. Barwood,* G. Huang, H. A. Klein, L. A. M. Johnson, S. A. King, H. S. Margolis, K. Szymaniec, and P. Gill

National Physical Laboratory, Teddington, TW11 0LW, United Kingdom

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The frequencies of two nominally identical ⁸⁸Sr⁺ trapped single ion optical clocks, based on the 674 nm $5s^{2}S_{1/2}$ -4 $d^{2}D_{5/2}$ electric quadrupole clock transition, have been compared over a period of nine months. The frequencies of the two clocks were found to agree within a total uncertainty of 4×10^{-17} , demonstrating that the individual ⁸⁸Sr⁺ optical clocks are reproducible at the 3×10^{-17} level. The absolute frequency of the clock transition was measured to be f = 444779044095486.71(24) Hz using an optical frequency comb referenced to a cesium fountain primary frequency standard. The standard uncertainty of 0.24 Hz (5.3×10^{-16} of the optical frequency) is dominated by measurement statistics and cesium fountain systematics and is around four times lower than previously published.

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During the last few years, there have been rapid advances in the performance of optical atomic clocks based on various cold trapped ions [1-9] and neutral atoms [10,11]. The frequencies of a number of cold ion and atom-based standards, including the ⁸⁸Sr⁺ clock studied here, are now known with sufficient accuracy to be used as secondary representations of the second [12]. The estimated systematic uncertainties of the best optical clocks are now below 1 part in 10^{17} [2,11] compared to ~2 parts in 10^{16} [13] for the best cesium primary standards. However, to date, there have only been a few comparisons between optical clocks that demonstrate agreement below 1 part in 10^{16} [2,11]. Such measurements are essential to verify the estimated uncertainties of the optical clocks. For ⁸⁸Sr⁺, the National Research Council (NRC) in Canada recently estimated the uncertainty of their (single) standard to be at the low parts in 10^{17} level [6,14]. In this Rapid Communication, we report an uncertainty budget at a similar level, but also include experimental two-trap comparison data to demonstrate this reproducibility over a period of several months. This comparison shows a null offset to within a total relative uncertainty of 4×10^{-17} , from which we deduce a singletrap reproducibility of 3×10^{-17} . Furthermore, an absolute frequency measurement of the ⁸⁸Sr⁺ optical clock transition relative to the National Physical Laboratory (NPL) cesium fountain primary frequency standard is reported, significantly better than previously published.

The two ⁸⁸Sr⁺ optical clocks are based on the 5*s* ²S_{1/2}– 4*d* ²D_{5/2} electric quadrupole transition at 674 nm in independently trapped and cooled strontium ions. The ions are separately confined in a pair of nominally identical Paul traps of an endcap design [15,16]. The traps were driven with an rf voltage at a frequency of either 17 or 14.2 MHz to give radial secular frequencies of between 1.1 and 1.8 MHz. Three orthogonal pairs of coils are placed around each trap to control the dc magnetic field. The Earth's field was nulled and then a horizontal bias field of 7 μ T applied using low-noise current drivers. Mu-metal shields surrounding the trap reduce the effect of ac and transient magnetic fields generated by nearby power supplies at the UK mains frequency of 50 Hz and a local train line.

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Trapped single strontium ions are cooled using light at 422 nm from a frequency doubled diode laser at 844 nm and tuned to the $5s^2S_{1/2}-5p^2P_{1/2}$ transition. Clear-out from the metastable $4d^2D_{3/2}$ level is achieved via a 1092 nm distributed feedback (DFB) laser. A 1033 nm DFB laser is used to drive the $4d^2D_{5/2}-5p^2P_{3/2}$ transition. This laser is normally shuttered "off" but admitted to the trap when the ion has been driven into the $4d^2D_{5/2}$ level by the probe laser to reduce dead time in the measurement cycle. A more complete description of the overall experimental arrangement for a single trap is given in [17]. The 674 nm probe laser is an extended cavity diode laser locked to an ultralow expansion (ULE) optical cavity with a finesse of $\sim 200\,000$. For the results presented here, 50 ms rectangular probe pulses are used to synchronously probe two ions, which lead to a 20 Hz Fourier transform-limited spectroscopic linewidth in both traps. The probe laser has a linewidth of ~ 4 Hz at 20 s and is similar to that reported in [18] except that a 5 mW slave laser is now injection locked to the light transmitted through the ULE cavity. This filters out high frequency laser noise at ~400 kHz corresponding to the servo bandwidth, which is outside the cavity transmission window. The output from this injection-locked laser is split three ways: two outputs are directed via independent maser-referenced acousto-optic modulators (AOMs) to the two traps and a third beam is delivered to the femtosecond comb laboratory via a 40 m optical fiber. The output for the comb is frequency shifted via a third maser-referenced AOM to provide light at the center frequency of the 674 nm transition. This AOM is also used for cancellation of the environmentally induced phase noise in the fiber link.

Software controls the frequencies of AOM1 and AOM2 which are used to bridge the frequency interval between the ULE cavity resonance and the clock transition in the two traps and thus records the frequency difference between the two clocks. The quantum jump probability is recorded with the laser stepping between the high and low frequency half-maxima of a pair of Zeeman components symmetrically placed around line center [19]. First, a pair of $\Delta m_J =$ 0 transitions is interrogated, then $\Delta m_J = \pm 1$, and finally $\Delta m_J = \pm 2$ with upper state magnetic quantum numbers $m_J = \pm 1/2, \pm 3/2, \text{ and } \pm 5/2,$ respectively. The average of these three transition frequencies is independent of the linear Zeeman shift, quadrupole shift, and tensor components

^{*}geoffrey.barwood@npl.co.uk

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	Frequency shift $\times 10^{-18}$	Measurement uncertainty $\times 10^{-18}$	Comparison uncertainty $\times 10^{-18}$
Blackbody shift due to 295(1) K ambient temperature	494	7	10
Blackbody shift uncertainty due to Stark constant		34	
Second-order Zeeman shift	2	1	1
Residual first order Zeeman shift	<1	7	10
Stark shift (secular motion/ heating)	9	9	13
Doppler shift due to secular motion	<1	4	6
674 nm laser Stark shift	3	3	<1
Cooling laser Stark shifts due to shutter leakage	3	3	<1
Servo errors	56	28	18
Stark shifts due to micromotion	31	7	10
Doppler shifts due to micromotion	-24	7	10
Residual quadrupole shifts	<1	4	3
Uncompensated fiber links	<1	11	6
"Line pulling" due to neighboring lines	<1	2	3
AOM frequency chirping	<1	1	1
Collisional	<1	7	10
Subtotal	574	49	32
Measurement statistics		482	24
Gravitational redshift	-63	16	<1
Fiber noise cancellation		4	
NPL-CsF2 systematics		200	
10 MHz distribution and rf synthesizer		100	
Frequency comb		10	
Total	511	534	40

TABLE I. Uncertainty budget for the 2012 two-trap comparisons and absolute frequency measurements.

of the Stark shift [1,20]. To reduce any systematic effects caused by slowly drifting magnetic field directions, the order of these interrogations is reversed in alternate measurements (i.e., the interrogation sequence is then $\Delta m_J = \pm 2, \pm 1$, and 0). Following a set of six measurements of pairs of components, a new trap center frequency is recorded. The laser interrogates each pair of Zeeman components for approximately equal time periods. Typically, Zeeman switching intervals of 2 or 3 minutes were chosen for the data presented here.

To estimate shifts and uncertainties associated with the ⁸⁸Sr⁺ optical clock, we adopt a similar approach to [14], but include discussion specific to our system to explain the uncertainty budget in Table I. This includes estimates relating both to the absolute frequency measurement and to the twotrap comparison. For most entries, the two-trap comparison uncertainty is $\sqrt{2}$ times the uncertainty for the single-trap optical frequency measurement. However, other entries are smaller for the two-trap comparison as some contributions are common mode between the traps. For example, there are short fiber links between the 674 nm laser and both traps that are in the same temperature-controlled laboratory but which have no phase noise cancellation. It is expected that frequency shifts arising from any changes in the temperature of these fiber links will be largely common mode for the two traps. Both traps are in close proximity and at the same height above the geoid to within 1 cm. In contrast, the NPL cesium fountain is located in a second laboratory and the time-averaged height of the cesium atoms in the fountain is 0.58(0.15) m higher than the

two ⁸⁸Sr⁺ ion traps. For the absolute frequency measurement we therefore need to take into account a gravitational redshift of -6.3(1.6) parts in 10^{17} .

The largest frequency correction arises from the Stark shift due to blackbody radiation. In the absence of any heat sources, the ion trap is in thermal equilibrium and so the ion experiences blackbody radiation. However, the inner endcap electrodes, which subtend 29% of the solid angle viewed by the ion, are at an elevated temperature when rf is supplied to the trap. This causes a perturbation to the room temperature blackbody environment, the size of which must be estimated.

To evaluate the temperature rise of the inner electrodes, a dummy trap was assembled that could be driven with similar drive circuits to the operational traps. Temperature measurements were made of the tantalum endcap electrodes using a Cedip thermal imaging camera through a MgF₂ window. With rf applied to the endcaps, inner endcap temperature rises of 4(2) K were observed. The uncertainty arises from uncertainties in the emissivity of the electrodes and rf drive levels in the real and dummy traps. The temperature of the trap vacuum enclosures was monitored using two calibrated thermistors within the mu-metal shields. This temperature was 0.6(0.1) K higher for the 2013 data. The small 2012 to 2013 differences in blackbody radiation shifts are taken into account for the frequency measurements although only the 2012 correction is shown in Table I. For the absolute frequency measurements, uncertainties related to the published calculations for the blackbody Stark shift are included. Two recent calculations give slightly different values for the total blackbody shift from absolute zero to 300 K of 0.250(9) Hz [21] and 0.22(0.01) Hz [22]. For the absolute frequency measurements, we assume a total average blackbody frequency shift of 0.235(15) Hz which we then correct to our ambient temperature of 295 K. This uncertainty is higher than that reported in [6] as we have included a more recent blackbody shift calculation [22] in our estimate.

The lasers used for cooling and probing the ion can cause Stark shifts but the 422, 1092, and 1033 nm lasers are shuttered when the 674 nm laser is interrogating the ion. Values for the Stark shift from these lasers are calculated from the measured extinction ratios and probe laser power in our system.

We have estimated Stark shifts due to the secular motion in both traps from the ion heating rates measured via the Rabi oscillation decay on the clock transition [16]. In our traps, the two heating rates are slightly different; one trap displays a heating rate of ~ 1 motional quantum/ms, whereas in the second trap it is ~ 4 quanta/ms. The higher of these two heating rates is allowed for in Table I. During the probe pulse when the cooling laser is off, this heating will cause the ion to have increased motional systematic shifts including exposure to rf electric fields that are present away from the ac center of the trap. The associated frequency shift and uncertainty is given in Table I. Finally, the software-controlled servo, centering the 674 nm laser frequency to the ${}^{2}S_{1/2} - {}^{2}D_{5/2}$ transition line center, has small residual errors in the quantum jump imbalances which result in the frequency offsets and uncertainties shown in Table I.

For 3 minute Zeeman switching times, a new frequency difference between the traps is calculated every 1080 s. Individual data sets lasted between 1 hour and 38 hours. For the two longest continuous data sets the Allan deviation of the frequency difference between two traps was calculated and the mean result is shown in Fig. 1. The fitted instability of $3.0 \times 10^{-14} \tau^{-1/2}$ corresponds to a single-trap instability of $2.2 \times 10^{-14} \tau^{-1/2}$. This result is slightly higher than our previously observed single-trap instability [17] of $1.4 \times 10^{-14} \tau^{-1/2}$, when locking only to the $\Delta m_J = 0$ transition,



FIG. 1. (Color online) Overlapping Allan deviation of the frequency difference between the two NPL strontium ion trap optical clocks. A straight line fit shows an instability of $\sigma = 3.0 \times 10^{-14} \tau^{-1/2}$, corresponding to $2.2 \times 10^{-14} \tau^{-1/2}$ for each trap.



FIG. 2. (Color online) Two-trap frequency comparison data from October 2012 to July 2013 (~19 days of measurements). The mean frequency difference between the traps of -4(11) mHz (dashed line) is consistent with zero to within the measurement uncertainties.

probably because Zeeman component switching necessarily involves the $\Delta m_J = \pm 2$ transitions that are the most sensitive to magnetic field. We observe some day-to-day variation in the short-term frequency instability that modeling suggests arises from different magnetic field noise levels. Analysis of the 2012 data, which extends over a discontinuous period of $\sim 10^6$ s, indicates a dominant white frequency noise characteristic over time scales to $\tau > 200\,000$ s.

The results for the frequency comparison between the two traps are shown in Fig. 2. A total of 34 data files were taken at different times between October 2012 and July 2013. These represent a total of over 19 days of comparison data, some taken simultaneously with femtosecond comb measurements. The pause in data taking between December 2012 and June 2013 was partly to allow the implementation of a system for automatic monitoring and minimization of the excess micromotion in the two traps. New rf trap drive circuits were also introduced during this period that operated at 14.2 MHz, closer to the frequency where the rf-induced Stark shifts and second-order Doppler shifts cancel [14]. As a result of these changes, the 2012 and 2013 comparison results were analyzed separately. The automatic micromotion minimization operates via computer-controlled voltages on an endcap and two compensation electrodes. A full evaluation of the arrangement to reduce micromotion automatically is still under way; in 2013 a high gain was used for the endcap correction that was later found to be nonoptimal. This caused increased instability in the 2013 two-trap comparisons giving larger statistical uncertainties (shown by the error bars in Fig. 2) and corresponding scatter. This resulted from larger quadrupole shift variations due to computer-generated endcap voltage changes. The difference between the 2012 and 2013 results is 48(34) mHz, consistent with zero.

The weighted average frequency differences between the two traps in 2012 and 2013 were -10(12) mHz and +38(32) mHz, respectively. Combining these figures and weighting according to the two uncertainties gives a mean



FIG. 3. (Color online) Absolute optical frequency measurements and associated uncertainties for 113 hours of data taken between December 2012 and July 2013. The mean frequency is indicated by the dashed green line and the standard uncertainties by dotted lines.

difference of 4 mHz with a statistical (type A) standard error of the mean of 11 mHz (2.4×10^{-17}). Adding this in quadrature with the systematic (type B) uncertainty of 3.2×10^{-17} shown in Table I (right-hand column) gives a total standard uncertainty for the comparison of 4×10^{-17} (18 mHz). Assuming similar contributions from each trap and so dividing this number by $\sqrt{2}$ gives a reproducibility of 13 mHz or 3 parts in 10^{17} of the optical frequency for a single trap.

Absolute frequency measurements of the ⁸⁸Sr⁺ clock transition were made during December 2012 and June to July 2013 totaling a period of 113 hours (4.7 days) of data taking (Fig. 3). Measurements were made simultaneously using both a Ti:sapphire comb [23,24] and a fiber comb [25]. Both combs were referenced to a hydrogen maser, the frequency of which was calibrated throughout the measurement period using the local cesium fountain primary standard NPL-CsF2 [26,27]. The values obtained using the two combs agreed to within one part in 10¹⁷, demonstrating that the combs themselves introduced negligible uncertainty. Uncertainties arising from the rf frequency distribution and synthesizers are also addressed [4] and found to be less than one part in 10¹⁶ for the period of the measurements. The systematic uncertainty

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arising from the cesium fountain primary standard itself is 2.0 $\times 10^{-16}$ [13]. After making the necessary corrections and including all uncertainties, the transition frequency is determined to be f = 444779044095486.71(24) Hz. This new measurement is in reasonable agreement with both the recent NRC value of f = 444779044095485.5(0.9) Hz [6] and our previous measurement [1]. The absolute frequency measurement uncertainty of 5.3 parts in 10^{16} (Table I; column labeled "Measurement uncertainty") is limited by statistics and uncertainties associated with the cesium fountain and is almost four times better than previously published. The largest correction to the strontium optical clock relates to the blackbody shift.

In summary, we have made a high accuracy frequency comparison between two strontium ion optical clocks. This comparison shows that the two standards agree to within a total relative uncertainty of 4 parts in 10^{17} . This corresponds to a single-trap reproducibility of 3 parts in 10^{17} , well below the reproducibility of cesium fountain primary frequency standards. Finally, an absolute frequency measurement with a relative standard uncertainty of 5.3 parts in 10^{16} has also been presented. This is expected to lead to a reduced uncertainty for the internationally agreed value for this transition frequency as an optical secondary representation of the second.

Future work will concentrate on improving laser reliability and implementation of a new trap design to reduce motional heating rates. Lower heating rates, together with optical ground state preparation into the appropriate Zeeman level, will provide coherent excitation into the $4d^2D_{5/2}$ level and allow use of longer interrogation times. The resulting narrower clock transition linewidth and increased signal-to-noise ratio is expected to significantly improve frequency stability, reducing the time taken to reach a given uncertainty level for two-trap comparisons and allow improved evaluation of frequency shifts.

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