Frequency ratio of an $^{115}$In$^+$ ion clock and a $^{87}$Sr optical lattice clock

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We report on the first frequency ratio measurement of an $^{115}$In$^+$ single ion clock and a $^{87}$Sr optical lattice clock. A hydrogen maser serves as a reference oscillator to measure the ratio by independent optical combs. Over more than 90 000 seconds of measurement time, the frequency ratio $f_{^{115}In^+}/f_{^{87}Sr}$ is determined to be 2.952 748 749 874 863 4(21) with relative uncertainty of $7.0 \times 10^{-16}$. The measurement creates a new connection in the network of frequency ratios of optical clocks.

Recent progress of optical clocks based on trapped ions and neutral atoms has made their uncertainties much smaller than those of the best cesium frequency standards [1–3]. Now a new definition of the SI unit of time using the optical clocks has become realistic. Consultative Committee for Time and Frequency (CCTF) drafted a roadmap to the redefinition of the SI second, where as a prerequisite to the new definition it requires measurements of frequency ratios between different atomic transitions as well as the reproducibility of the clocks in different laboratories both with uncertainty matching that of the best clocks. Frequency ratios have been reported (in references [4–10] among others) for comparisons of different combinations of ion clocks and optical lattice clocks, or even for different transitions in the same ion. However, a large number of frequency ratios has still never been measured directly.

The indium ion ($^{115}$In$^+$) is one of the original candidates in the first proposal for single ion clocks [1]. At 237 nm, it has the highest clock frequency of all presently investigated optical clocks [2], and high transition energies give it a very small sensitivity to blackbody radiation (BBR), comparable to that of $^{27}$Al$^+$ [12]. A small quadrupole moment enables multi-ion clocks with enhanced stability [13]. Despite the appeal of the $^{115}$In$^+$ ion, spectroscopic determination of the clock frequency [14–16] and clock operation by feedback to the clock laser [17] have been achieved only recently. A recommended value for the $^{115}$In$^+$ clock frequency has been included in the list of standard frequencies [2], but no frequency ratio to another optical clock is available so far. Here we report the first such measurement, with an uncertainty of $7.0 \times 10^{-16}$.

The measurement is performed using an $^{115}$In$^+$ optical clock [16, 17] and the $^{87}$Sr optical lattice clock NICT-Sr1 [18], located in the same building. The $^{115}$In$^+$ optical clock probes the $^1S_0^->^3P_0$, $m_F=\pm 7/2$ transitions. The $^{87}$Sr clock uses approximately 1000 $^{87}$Sr atoms loaded into a vertically oriented one-dimensional optical lattice after two-stage laser cooling and achieves a systematic uncertainty of $7 \times 10^{-17}$ in this measurement. The details of the clocks are described in the previous reports [14–18].

![FIG. 1. Schematic representation of the frequency ratio measurement. The optical frequencies of the $^{115}$In$^+$ clock and the $^{87}$Sr clock are recorded as beat notes with independent optical frequency combs locked to a common hydrogen maser (HM). Optical comb 2 is also used to stabilize the frequency of an auxiliary laser. SHG: second harmonic generation.](image)
The results are shown in Fig. 3 in terms of the relative deviation $\Delta y_{\text{Sr}}$ determined from the clock laser beat with the nearest comb line and binned over a 10 s interval. With a flicker-noise floor close to $2 \times 10^{-16}$, the HM then serves as a flywheel oscillator that can bridge even extended interruptions in Sr clock operation without degrading the stability. $\Delta y_{\text{In}^+}$ is determined in the same way for the four days of Mar 17–20 2020. The combined data is then used to determine $R(t)$.

![FIG. 3. Frequency measurements of $^{115}\text{In}^+$ and $^{87}\text{Sr}$ clocks. Results are shown as fractional deviation after subtracting a common baseline of $1.2 \times 10^{-12}$ to accommodate the mean HM frequency $y_{\text{HM}} = 1.2001 \times 10^{-12}$. The blue '+' symbols represent $\Delta y_{\text{In}^+}$ while the red '-' symbols represent $\Delta y_{\text{Sr}}$.](image)

The systematic uncertainty evaluation of the In$^+$ clock is largely identical to our previous report [16]. However, the clock frequency is now determined by actively stabilizing the clock laser to represent the center of two resolved transitions between Zeeman sublevels where it was previously determined from spectra obtained by laser frequency scans across unresolved transitions over degenerate Zeeman sublevels. A dynamic application of magnetic field during the clock laser irradiation periods [17] serves to separate the transitions between the Zeeman sublevels ($^1S_0 \ m_F = \pm 9/2$ to $^3P_0 \ m_F = \pm 7/2$). This simultaneously allows precise determination of the magnetic field using the known $g$-factors of the upper and lower energy levels [13]. The quadratic Zeeman shift is estimated to be 8 mHz using the magnetic field value and the known coefficient [13]. Another improvement is a reduction of the 2nd order Doppler shift due to secular motion. An auxiliary laser derived from a fundamental wavelength of 922 nm addresses the $^1S_0 - ^3P_1$ transition at 230 nm. Normally used for optical pumping and state readout, we inserted a pulse of this laser for direct cooling of the In$^+$-Ca$^+$ motion along the linear trap axis just before the clock interrogation period. The natural linewidth of 360 kHz allows a much lower temperature than the cooling of the $^{40}\text{Ca}^+$ ion on the 22 MHz wide $^2S_{1/2} - ^2P_{1/2}$ transition. The effect is confirmed by the observed suppression of the red motional sideband. The
temperature of the in-phase mode estimated from the ratio of the blue and red sidebands \( \frac{\Delta f_{\text{blue}}}{\Delta f_{\text{red}}} \) is about 70 \( \mu \)K. Although the implementation of the cooling is currently limited to one degree of motional freedom, the uncertainty is reduced from 0.7 Hz to 0.5 Hz with this improvement. The BBR shift is estimated to be \(-34\) mHz by temperature measurement of the trap electrode during operation. The overall systematic uncertainty budget is listed in Table I, with a total fractional uncertainty of \( \frac{\Delta f_{\text{Sr}}}{f_{\text{Sr}}} = 5.0 \times 10^{-16} \) corresponding to 0.6 Hz. The Sr clock contributes a systematic uncertainty of \( \Delta f_{\text{Sr}} = 7.4 \times 10^{-17} \) as described in reference [13]. Combined with \( u_a = 4.9 \times 10^{-16} \), this yields an overall fractional uncertainty of \( 7.0 \times 10^{-16} \), and we determine the optical frequency ratio to be \( R = 2.952 748 749 874 863 4 (21) \). Since the reference HM is a part of the Japan Standard Time System (JST) at NICT [21], its frequency can also be traced to the SI second, allowing for an absolute measurement of the \( ^{115}\text{In}^+ \) clock frequency. This begins with the fractional frequency deviation of the international timescale UTC, determined by the International Bureau of Weights and Measures (BIPM), and published for the period of Feb 25 to Mar 31, 2020 in volume 387 of its monthly Circular T. Based on measurements of multiple primary and secondary frequency standards, the reported value corresponds to \( \frac{\Delta f_{\text{UTC}}}{f_{\text{UTC}}} = 5.2(1.3) \times 10^{-16} \). We then use the stability of the international timescale to extrapolate this value to a shorter 25 day interval, and determine the frequency difference of UTC(NICT) relative UTC from the time delays published in the same Circular T. From the 25 day interval, the stability of the JST HM ensemble allows us to determine the mean frequency of the reference HM for the time of the \( ^{115}\text{In}^+ \) measurements with an overall uncertainty of \( 4.0 \times 10^{-16} \) [21]. Table I lists the individual uncertainty contributions along with the overall uncertainty of the absolute frequency measurement. The absolute \( ^{115}\text{In}^+ \) clock frequency is determined as \( 1 267 402 452 901 040.1(1.0) \) Hz, representing a comparison to the SI second. With a fractional uncertainty of \( 8.1 \times 10^{-16} \), this is a six-fold improvement over our previous measurement based on \( ^{115}\text{In}^+ \) spectra with unresolved Zeeman sublevels. The result falls outside the previously estimated uncertainty \( 10 \) by about \( 1.4 \sigma \). While the source of the discrepancy is not clearly identified, the possibility of errors from spectral distortions has been the driving motivation in implementing the new scheme of field application during spectroscopy. This technique has eliminated this possible source of uncertainty.

To confirm the link to the SI second, we can also calculate the \( ^{115}\text{In}^+ \) frequency from \( R \) and the CIPM recommendation for \( f_{\text{Sr}} \) as \( f_{\text{Sr}}' = R 	imes f_{\text{Sr}} \) to find \( 1 267 402 452 901 040.1(1.0) \) Hz, also with a relative uncertainty of \( 8.1 \times 10^{-16} \). The difference of the \( ^{115}\text{In}^+ \) clock frequencies determined in these ways is \( 0.2 \) Hz \( (1.6 \times 10^{-16}) \), and it is well within the additional uncertainty of the frequency link to the SI second.

While the Doppler shifts from ion motion limit this first optical ratio measurements of the \( ^{115}\text{In}^+ \) clock to a fractional uncertainty of \( 7 \times 10^{-16} \), extending the motional cooling scheme to all degrees of freedom together with a more accurate evaluation of the residual micro-motion will further reduce the uncertainty. Other uncertainty contributions represent the statistical uncertainty of in-situ measurements. These, along with the statistical contribution in any clock comparison, are expected to dramatically improve with increased stability of the clock laser. The current measurements only probe the clock transition for less than 20 % of the cycle time. The duty cycle of the clock interrogation will also increase with a new photo-counting setup that will decrease the time dedicated to state detection. With future improvements, we hope to demonstrate the \( ^{115}\text{In}^+ \) clock as a competitor to established candidates [22, 23], combining the insensitivity to environmental effects found in \( ^{27}\text{Al}^+ \) with an interrogation scheme that does not require the complexities of quantum logic techniques.

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**Table I.** Shifts and uncertainties in \( ^{115}\text{In}^+ \) system as well as frequency link to the SI second. BBR; blackbody radiation, S.M.; secular motion, M.M.; micromotion.

| Effects | Shift (10^{-16}) | Uncertainty (10^{-16}) |
|---------|-----------------|------------------------|
| 2nd order Zeeman shift | 0.06 | < 0.1 |
| Clock laser Stark shift | −0.03 | < 0.1 |
| BBR shift | −0.27 | < 0.1 |
| 2nd order Doppler (S.M.) | −0.004 | 3.9 |
| 2nd order Doppler (M.M.) | 0 | 3.1 |
| \( ^{115}\text{In}^+ \) systematics | −0.24 | 5.0 |
| \( ^{115}\text{In}^+ \) statistics | 4.9 |
| \( ^{115}\text{In}^+ \) total | 7.0 |
| Gravitational shift | 82.7 | 0.2 |
| HM extrapolation | 2.4 |
| UTC(NICT)−UTC extrapolation | 2.3 |
| UTC extrapolation (25 days) | 1.8 |
| UTC−SI second | 1.3 |
| Overall total | 8.1 |

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1. F.-L. Hong, Optical frequency standards for time and length applications, Measurement Science and Technology 28, 012002 (2016).
2. F. Riehle, P. Gill, F. Arias, and L. Robertsson, The CIPM list of recommended frequency standard values: guidance, and procedures, Metrologia 55, 188 (2018).
3. J. Lodewyck, On a definition of the SI second with a set of optical clock transitions, Metrologia 56, 655009 (2019).
4. T. Rosenband, D. B. Hume, P. O. Schmidt, C. W. Chou, A. Brusch, L. Lorini, W. H. Oskay, R. E. Drullinger,
[54x-1730][12] M. S. Safronova, M. G. Kozlov, and C. W. Clark, Precision calculation of blackbody radiation shifts for optical frequency metrology, Phys. Rev. Lett. 107, 143006 (2011).

[13] J. Keller, T. Burgermeister, D. Kalincev, A. Didier, A. P. Kolumb, T. Nordmann, J. Kiethe, and T. E. Mehlstäubler, Controlling systematic frequency uncertainties at the 10^{-19} level in linear coulomb crystals, Phys. Rev. A 99, 013405 (2019).

[14] J. von Zanthier, T. Becker, M. Eichenseer, A. Y. Nevesky, C. Schwedes, E. Peik, H. Walther, R. Holzwarth, J. Reichert, T. Udem, T. W. Hänsch, P. V. Pokasov, M. N. Skvortsov, and S. N. Bagayev, Absolute frequency measurement of the In^+ clock transition with a mode-locked laser, Opt. Lett. 25, 1729 (2000).

[15] Y. Wang, R. Dumke, T. Liu, A. Stejskal, Y. Zhao, J. Zhang, Z. Lu, L. Wang, T. Becker, and H. Walther, Absolute frequency measurement and high resolution spectroscopy of \(^{115}\text{In}^+\) 5s\(^2\) 1S_0\text{S}3\text{S}^2P_0\) narrowline transition, Optics Communications 273, 526 (2007).

[16] N. Ohtsubo, Y. Li, K. Matsubara, T. Ido, and K. Hayasaka, Frequency measurement of the clock transition of an indium ion sympathetically-cooled in a linear trap, Optics Express 25, 11725 (2017).

[17] N. Ohtsubo, Y. Li, N. Nemitz, H. Hachisu, K. Matsubara, T. Ido, and K. Hayasaka, Optical clock based on a sympathetically-cooled indium ion, Hyperfine Interact. 240, 39 (2019).

[18] H. Hachisu, F. Nakagawa, Y. Hanado, and T. Ido, Months-long real-time generation of a time scale based on an optical clock, Sci. Rep. 8, 4243 (2018).

[19] U. Tanaka, T. Kitanaka, K. Hayasaka, and S. Urabe, Sideband cooling of a Ca\(^+\) In\(^+\) ion chain toward the quantum logic spectroscopy of In\(^+\), Applied Physics B 121, 147153 (2015).

[20] Y. Hanado, K. Imamura, N. Kotake, F. Nakagawa, Y. Shimizu, R. Tabuchi, Y. Takahashi, M. Hosokawa, and T. Morikawa, The new generation system of Japan standard time at nict, International Journal of Navigation and Observation 2008, 841672 (2007).

[21] N. Nemitz, T. Gotoh, F. Nakagawa, H. Ito, Y. Hanado, T. Ido, and H. Hachisu, Absolute frequency of \(^{87}\text{Sr}\) at 1.8 \times 10^{-16} uncertainty by reference to remote primary frequency standards (2020), arXiv:2008.00723.

[22] N. Huntemann, C. Sanner, B. Lipphardt, C. Tamm, and E. Peik, Single-ion atomic clock with 3 \times 10^{-18} systematic uncertainty, Phys. Rev. Lett. 116, 063001 (2016).

[23] S. M. Brewer, J.-S. Chen, A. M. Hankin, E. R. Clements, C. W. Chou, D. J. Wineland, D. B. Hume, and D. R. Leibrandt, \(^{27}\text{Al}^+\) quantum-logic clock with a systematic uncertainty below 10^{-18}, Phys. Rev. Lett. 123, 033201 (2019).