

Hertz-level measurement of the $^{40}\text{Ca}^+ 4s\ ^2S_{1/2}-3d\ ^2D_{5/2}$ clock transition frequency with respect to the SI second through the Global Positioning System

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We have measured the absolute frequency of a clock transition at the 10^{-15} level with a single, trapped, laser-cooled $^{40}\text{Ca}^+$ ion in a miniature Paul trap. In our measurement, an optical frequency comb is referenced to a hydrogen maser, which is calibrated to the SI second through the Global Positioning System (GPS). By analyzing the experimental data obtained within 32 days of a total averaging time $>2 \times 10^6$ s, the absolute frequency of the $^{40}\text{Ca}^+ 4s\ ^2S_{1/2}-3d\ ^2D_{5/2}$ clock transition is measured as 411 042 129 776 393.0 (1.6) Hz with a fractional uncertainty of 3.9×10^{-15} .

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Optical frequency standards have been developed rapidly thanks to recent techniques using cold atoms, optical frequency combs [1,2], and ultranarrow-linewidth lasers. Optical frequency standards are expected to take the place of the Cs primary microwave standard as the definition of the SI second in the near future. The absolute frequency measurement for the clock transition is an important step in achieving an optical frequency standard based on a single ion or neutral atoms. The clock transition frequency had been measured by using an optical frequency comb referenced to a Cs fountain and using the Global Positioning System (GPS) as a link to the SI second without a primary standard for direct calibration. The clock transition frequencies have been measured referenced to the Cs fountain at uncertainties on the order of 10^{-15} or even smaller with ^{87}Sr , ^{88}Sr , ^{171}Yb , ^{174}Yb , $^{88}\text{Sr}^+$, $^{171}\text{Yb}^+$, $^{27}\text{Al}^+$, $^{40}\text{Ca}^+$, and $^{199}\text{Hg}^+$ [3–11]. For an optical lattice clock, the clock frequency of Sr had been measured referenced to GPS as a link to the SI second by the National Metrology Institute of Japan—National Institute of Advanced Industrial Science and Technology (NMIJ/AIST) with uncertainties on the order of 10^{-14} level or smaller [12–14]. With the precise measurements of the optical frequency standards via single trapped ions or ultracold neutral atoms, frequencies of lasers (optical local oscillators) stabilized to the clock transitions of atoms and/or ions have been recommended by the International Committee for Weights and Measures (CIPM) as secondary representations of the SI second, contributing to International Atomic Time (TAI) [15].

Because there are commercially available lasers for photoionization, cooling, manipulation, and detection, a single Ca^+ ion has several technological advantages for building a practical optical clock, which has been proposed as an alternative candidate for the next definition of the SI second

[15]. The $4s\ ^2S_{1/2}-3d\ ^2D_{5/2}$ clock transition of $^{40}\text{Ca}^+$ at 729 nm has a natural linewidth of 0.2 Hz [16], which has good potential accuracy and low systematic shifts. The optical frequency standard based on $^{40}\text{Ca}^+$ is also being developed by the Quantum Optics and Spectroscopy Group in Innsbruck and the National Institute of Information and Communications in Japan (NICT). The Innsbruck group has measured the clock frequency as 411 042 129 776 393.2(1.0) Hz with a fractional uncertainty of 2.4×10^{-15} , which was referenced to the transportable Cs atomic fountain clock of LNE-SYRTE [10]. The NICT group has measured the clock frequency as 411 042 129 776 385(± 18) Hz with a fractional uncertainty of 10^{-14} level, which was referenced to the Cs atomic fountain clock [17].

In this paper, we report on measurements of the $^{40}\text{Ca}^+ 4s\ ^2S_{1/2}-3d\ ^2D_{5/2}$ transition frequency with an uncertainty level of 10^{-15} using an optical frequency comb referenced to a hydrogen maser (H maser), which is calibrated through GPS as a link to the SI second. To achieve the measurement at the 10^{-15} level, we have implemented the measurement for a very long time (7×10^5 s in May 2011 and 1.5×10^6 s in June 2011) to reduce the statistical uncertainty and the frequency transfer uncertainty. In addition to the absolute frequency measurement, the systematic uncertainties have been reduced by cooling the ion to lower temperatures and increasing the measurement precision on the electric quadrupole shift.

Our experiment is implemented with a single $^{40}\text{Ca}^+$ ion trapped and cooled in a miniature Paul ring trap. The trap has an endcap-to-center distance of $z_0 \approx 0.6$ mm with a center-to-ring electrode distance $r_0 \approx 0.8$ mm. Two electrodes perpendicular to each other are set in the ring plane to compensate for the ion's excess micromotion. A trapping rf voltage of ~ 580 V_{p-p} is applied to the ring at a frequency of 9.8 MHz. The excess micromotion is nulled by applying different dc bias voltages on the endcap electrodes and the compensation electrodes. The 729-nm laser is locked to the six chosen Zeeman components to cancel the linear Zeeman shift and the electric quadrupole shift. After that, the measurements of the clock transition frequency,

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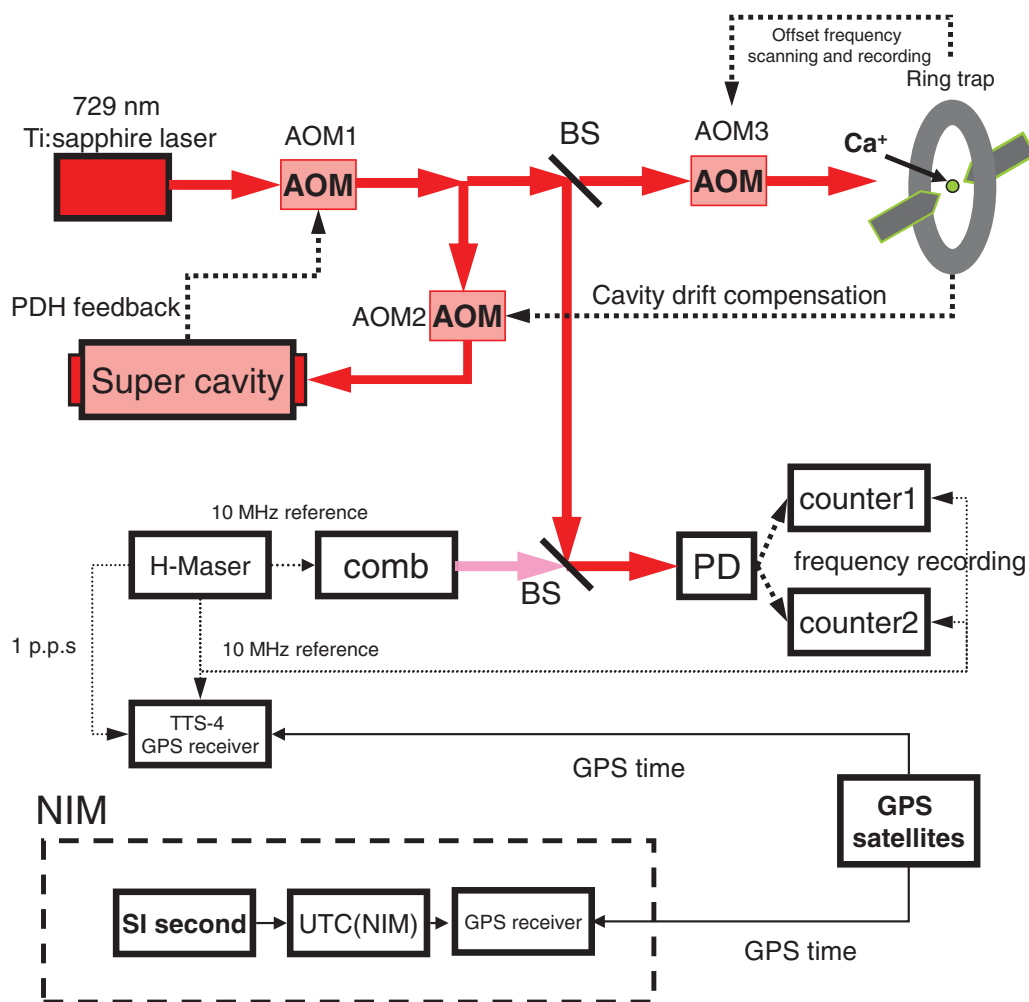


FIG. 1. (Color online) Schematic diagram for the frequency measurement. AOM1 is used to lock the laser to the supercavity as the fast loop feedback of the PDH method, AOM2 is used to compensate the cavity drift, and AOM3 is used to scan and record the offset frequency. AOM denotes acousto-optic modulator; HWP half-wave-plate; PD photodetector; GPS global positioning system, NIM National Institute of Metrology of China.

the stability of the clock, and the evaluation of the systematic shifts are taken. The methods of trapping, cooling, probing, and locking schemes in this work have been used in our previous works [18–21]. The schematic diagram for our experimental setup is shown Fig. 1.

The probe laser at 729 nm is a commercial Ti:sapphire laser (MBR-110, Coherent), which is locked to a temperature-controlled high-finesse Fabry-Perot cavity (Zerodur material) supported on an active isolated platform (TS-140, Table Stable) using the Pound-Drever-Hall (PDH) technique [22]. A linewidth of ~ 13 Hz is measured from the heterodyne beat note with a homemade diode laser stabilized to another high-finesse ultralow-expansion (ULE) cavity. The long-term drift is measured to be ~ 3 Hz/s. An acousto-optic modulator (AOM) (80 MHz, Brimrose) driven by a sweeping function generator (2023A, IFR) is used to compensate for the long-term drift. After the compensation, we get a nonlinear drift less than 0.1 Hz/s. The offset frequency between the probe laser and the clock transition line center of the ion is achieved by an AOM, which shifts the laser frequency to match the transitions. The required AOM frequencies are updated every 40 cycles of pulses, which take ~ 1.1 s. By the “four points locking scheme”

[23,24], three pairs of the Zeeman transitions are interrogated, and the offset frequency between the probe laser and the clock transition could be obtained every ~ 13 s. In the mean time, a Ti:sapphire-based optical frequency comb (FC 8004, MenloSystems) is used to measure the 729-nm laser frequency. Both the repetition frequency and the offset frequency of the fs comb are locked to two individual synthesizers, which are referenced to a 10-MHz signal provided by an active H maser (CH1-75A) with an isolated splitter and a 60-m-long standard 50- Ω coaxial cable. Two individual counters are used to measure the beat frequency of the comb laser and the 729-nm laser simultaneously; if the difference of the readings of the two counters is larger than 1 Hz, we believe the measurement is not reliable; thus they are not taken into account. The probe laser frequency is measured every 1 s. With the above two parts of the measurement results, we can do the measurement of the clock transition frequency referenced to the H maser and calculate the frequency instability comparison of the $^{40}\text{Ca}^+$ optical clock versus the H maser (Fig. 2). Figure 2 shows the histogram of the clock transition frequency measurements referenced to the H maser on day MJD 55 726, which gives an averaged value of 411 042 129 776 490.7 Hz. The histogram

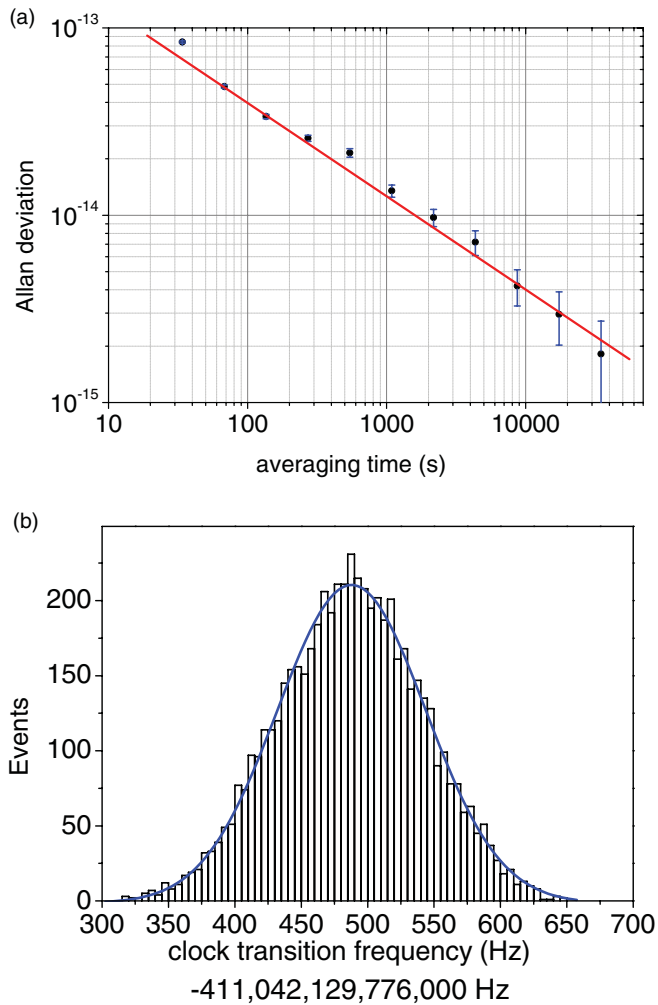


FIG. 2. (Color online) (a) The Allan deviation for the $^{40}\text{Ca}^+$ optical clock vs the H maser; the red line represents $4 \times 10^{-13} \tau^{-1/2}$. (b) Histogram of the frequency measurements of the clock transition calculated from the offset frequency and the comb measurements referenced to the H maser with a Gaussian fitting (blue curve).

follows a normal distribution and the standard deviation of the mean is 3.5 Hz. The longest continuous measurement is up to >50 h. As shown in Fig. 2, the Allan deviation for the ion transition versus the H maser comparison reaches the 10^{-15} level after >2000 s of averaging time, which could be limited by the stability of the H maser and the stability of the frequency transfer.

Frequency measurements were taken in 32 individual days, separated into two parts, one in May 2011 with 15 continuous days and the other in June 2011 with 17 continuous days (Fig. 3). Each filled circle in Fig. 3 represents a mean value of the measurement result based directly on the H maser. The error bars are given by the standard deviation of the mean. The former 15 days of measurements give a weighted averaged frequency of $411\,042\,129\,776\,489.7(0.9)$ Hz and the latter 17 days of measurements give a weighted averaged frequency of $411\,042\,129\,776\,489.1(0.4)$ Hz. In the first round, the laser locked to the clock transition in 1 day is about 12 h. More robustly, in the second round, the laser locked to the clock transition in 1 day is larger than 22 h. Thus, the clock in the second round works for $>90\%$ of the time and the statistical uncertainty for 1 day of the averaging time is expected to be smaller, yet the results showed the opposite. In fact, in the last 10 days, the Allan deviation only went down to 1×10^{-14} and then it showed a flicker floor. We think that the larger uncertainties are mainly limited by the H maser.

To get the final absolute frequency of the clock transition from the averaged frequency, systematic shifts and the calibration of the reference must be taken into account. There are several sources of systematic shifts, which might be associated with the quadrupole $729\text{-nm } 4s \ ^2S_{1/2} - 3d \ ^2D_{5/2}$ clock transition in a laser-cooled trapped $^{40}\text{Ca}^+$ ion. The details of evaluating the systematic shifts have been given in Ref. [18]. Here, systematic shifts of our system are reevaluated. We have made some improvements on the evaluation of the systematic uncertainty with the following procedures. After the reminimization of the micromotion, the ion temperature is estimated to be at $3(3)$ mK from the intensity of the Secular motion sidebands relative to the carrier (normally about 0.2–0.4) [25]. With the estimated ion temperature, the second-order

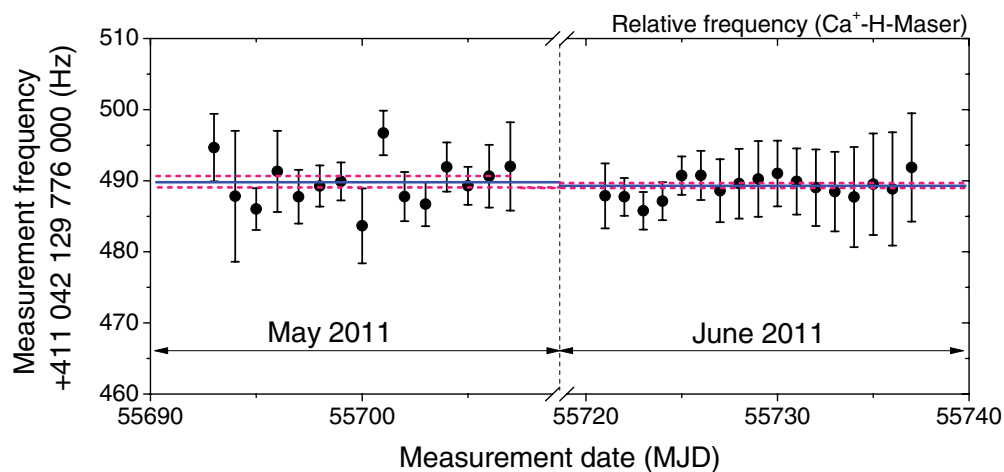


FIG. 3. (Color online) Frequency measurement of the $4s \ ^2S_{1/2} - 3d \ ^2D_{5/2}$ transition of a laser-cooled trapped single $^{40}\text{Ca}^+$ ion referenced to the H maser. Blue solid lines and red dashed lines represent the weighted mean and the uncertainty of the frequency measurements for each month, respectively. Data shown in this figure do not include systematic corrections.

TABLE I. The systematic frequency shifts and their uncertainties of the evaluation of the clock. Shifts and uncertainties given are in fractional frequency units ($\Delta\nu/\nu$).

Effect	Measurements in May		Measurements in June	
	Shift (units of 10^{-16})	Uncertainty (units of 10^{-16})	Shift (units of 10^{-16})	Uncertainty (units of 10^{-16})
Second-order Doppler shift due to thermal motion	-0.10	0.10	-0.10	0.10
Second-order Doppler shift due to micromotion	-0.49	0.49	-0.49	0.49
Stark shift due to thermal motion and micromotion	0	0.04	0	0.04
ac Stark shift due to 397, 866, and 854 nm	0	0.04	0	0.04
ac Stark shift due to 729 nm	0.97	1.46	0.97	1.46
Blackbody radiation shift	8.51	0.27	8.51	0.27
Linear Zeeman shift	0	4.52	0	6.23
Second-order Zeeman shift	0	0.01	0	0.01
Electric quadrupole shift	0	0.72	0	0.51
Gravitational shift	38.44	1.10	38.44	1.10
Total shift	47.4	5.0	47.4	6.5

Doppler shift caused by thermal kinetic energy is calculated to be $-0.004(0.004)$ Hz [26]. By locking the probe laser to the six different chosen Zeeman transitions ($M_J = \pm 1/2$, $M_J = \pm 3/2$, and $M_J = \pm 5/2$) one after another with an interrogation time of 11 ms, we achieve the transition linewidth of ~ 80 Hz. By averaging the center frequency of the three pairs of the components, we can null the quadrupole shift [23,26,27]. By averaging the difference of center frequency for different components, the uncertainty is obtained. For the much longer averaging time, the fractional uncertainty is smaller than the previous work [18]. Using the new factor value given in Ref. [28], the evaluation of the blackbody radiation shift uncertainty is reduced. For our $^{40}\text{Ca}^+$, the altitude was measured to be $35.2(1.0)$ m; thus the gravitational shift for the clock is estimated to be $1.583(0.045)$ Hz.

We list all significant frequency shifts in Table I. Taking all of them into account we get a total fractional shift of 4.74×10^{-15} with a fractional uncertainty of 5.0×10^{-16} from the data obtained in May 2011 and a total fractional shift of 4.74×10^{-15} with a fractional uncertainty of 6.5×10^{-16} for the data obtained in June 2011. We find that the uncertainty of linear Zeeman shift is in fact limiting the final systematic uncertainty. The linear Zeeman effect uncertainty is mainly caused by the fluctuation of the magnetic field, which was measured by calculating the variance of the Zeeman splitting of the Zeeman transitions. According to the variance of the Zeeman splitting, the uncertainty was calculated from statistics. To reduce the systematic uncertainties in the future, one has to increase the stability of the magnetic field.

In our measurement, the largest frequency correction comes from the calibration of the frequency of the H maser. To calibrate the frequency of the H maser, a GPS time and frequency transfer receiver with an antenna (TTS-4, PikTime Systems) has been used [29]. The receiver with reference to the 10 MHz and one pulse per second (pps) signals from the H maser receives the GPS signals from six to ten GPS satellites on average and generates and records the GPS measurement data. In the meantime, another receiver with reference to the 10 MHz and the one pps signals of the UTC (NIM) in the National Institute of Metrology (NIM) of

China has made a similar measurement. Using the two sets of data from the two institutes, we calculate the frequency difference of the H maser from the UTC (NIM). Considering the frequency difference of UTC (NIM) and SI second, the H maser we used for frequency measurement can be calibrated. Based upon the calculation with the GPS precise point positioning (PPP) technique [30], we achieve a frequency transfer uncertainty of $\sim 1 \times 10^{-14}$ with an averaging time of 1 day. In Fig. 4, we show the time difference between the H maser and the UTC (NIM). The weighted average of the frequency offset between the H maser and the UTC (NIM) is then calculated to be $-2.3649(0.0337) \times 10^{-13}$ for the data obtained in May 2011 and $-2.3582(0.0083) \times 10^{-13}$ for the data obtained in June 2011. Meanwhile, the frequency difference between the UTC (NIM) and the SI second from the primary standard can be calculated from the data reported on the BIPM website [31]. We found the computed values of [UTC-UTC(NIM)] and uncertainties for the two months of our measurements in Circular T No. 281 and No. 282; the results published for the two months were $0.0(2.1) \times 10^{-15}$ in May and $0.0(2.1) \times 10^{-15}$ in June, respectively. We

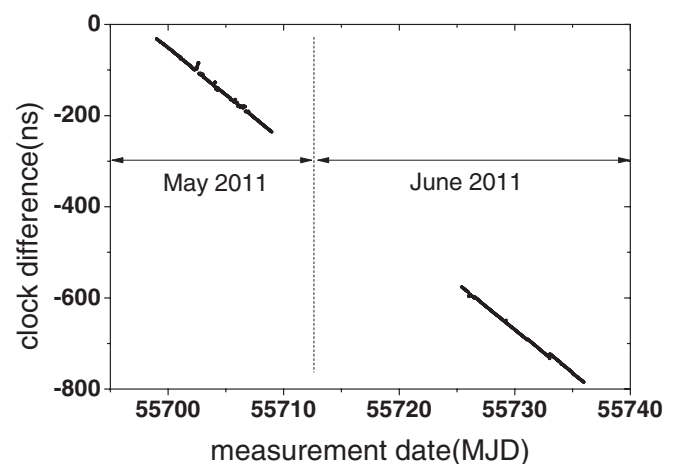


FIG. 4. Time difference between the H maser and the UTC (NIM) calculated using PPP technique.

TABLE II. The absolute frequency measurement budget table. The unit of shifts and uncertainties are given in Hz.

Contributor	Measurements in May		Measurements in June	
	Shift (Hz)	Uncertainty (Hz)	Shift (Hz)	Uncertainty (Hz)
Systematic shift (Table I)	1.95	0.21	1.95	0.27
Statistical	0	0.90	0	0.40
H maser reference calibrated with UTC (NIM)	97.21	1.39	96.93	0.34
UTC (NIM) reference	-2.3	0.9	-2.5	0.9
Total	96.9	1.9	96.4	2.6

also found the estimation of the UTC accuracy by comparison of the TAI frequency with that of the given individual primary frequency standards (PFS). In these two months, the results published in Circular T were $5.5(0.3) \times 10^{-15}$ in May and $6.0(0.4) \times 10^{-15}$ in June, respectively. With the results of the UTC-UTC(NIM) and TAI-PFS, we calculated the frequency difference between the UTC (NIM) and the SI second was $5.5(2.1) \times 10^{-15}$ for the data obtained in May 2011 and $6.0(2.1) \times 10^{-15}$ for the data obtained in June 2011.

In Table II, we give our estimation for several possible corrections to the absolute frequency of the $^{40}\text{Ca}^+$ clock transition.

Based upon the data listed in Table II, we determine the total correction for the frequency measurement shown in Fig. 3 is -96.9 Hz for the data obtained in May 2011 and -96.4 Hz for the data obtained in June 2011. The combined fractional uncertainty of the absolute frequency measurement is 4.6×10^{-15} for the data obtained in May 2011 and 2.6×10^{-15} for the data obtained in June 2011. The corrected absolute frequency of the $^{40}\text{Ca}^+ 4s^2S_{1/2}-3d^2D_{5/2}$ clock transition is $411\,042\,129\,776\,393.3(1.9)$ Hz for the data obtained in May 2011 and $411\,042\,129\,776\,392.7(1.1)$ Hz for the data obtained in June 2011, the two measurements agrees with each other within their uncertainties. The unweighted mean of the above two values gives a final result of $411\,042\,129\,776\,393.0(1.6)$ Hz; the final uncertainty is calculated by considering both statistical and systematic uncertainties. The result is in agreement with the previous measurements [10,17] and the recommended frequency value [15] within their uncertainties. The agreement among the standards in multiple laboratories at the level of 4×10^{-15} represents the good reproducibility of the Ca^+ optical frequency standard. The systematic uncertainties for the clock transition frequency (Table I) discussed above are all smaller than 1×10^{-15} .

In summary, the absolute frequency measurement of the $^{40}\text{Ca}^+ 4s^2S_{1/2}-3d^2D_{5/2}$ clock transition has been implemented at the 10^{-15} level. This is a milestone towards a practical optical clock. It is important to increase the measurement precision in the precision spectroscopy research area. Better precision could be achieved if we implemented the measurement referenced to GPS as a link to the SI second for a longer time. We shall do a comparison of the $^{40}\text{Ca}^+$ clocks between Wuhan, Tokyo, and Innsbruck, with respect to the SI second through GPS in the future. Moreover, we suppose the stability of the H maser and the transfer cable might cause the problem of the reproducibility that is not good enough and need to be improved. By introducing cryogenic sapphire oscillators and using optical fibers as in the frequency transfer, it is possible to achieve a measurement reproducibility of 10^{-16} level instead of 10^{-15} . To achieve a smaller uncertainty in the future, we need to use a more stable reference such as a Cs fountain instead of the GPS system to achieve better results with smaller measurement uncertainty or to reduce the averaging time achieving the same uncertainty.

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