

# Improved Measurement of the $^{40}\text{Ca}^+$ Clock Transition Frequency towards an Uncertainty of $10^{-15}$ Level

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## Abstract

We report improved frequency measurement of a single  $^{40}\text{Ca}^+$  optical clock towards the  $10^{-15}$  uncertainty level. In contrast with our previous measurement, in which the accuracy was limited by the magnetic field fluctuation, the use of a magnetic shield significantly reduced the observed linewidth of its  $^2S_{1/2}$ - $^2D_{5/2}$  clock transition. From frequency comparison between the  $^{40}\text{Ca}^+$  clock and an  $^{87}\text{Sr}$  lattice clock, an Allan deviation of  $7 \times 10^{-16}$  at an averaging time of one thousand seconds was measured. We now measure the absolute frequency of the  $^{40}\text{Ca}^+$  clock transition with respect to the SI definition of the second.

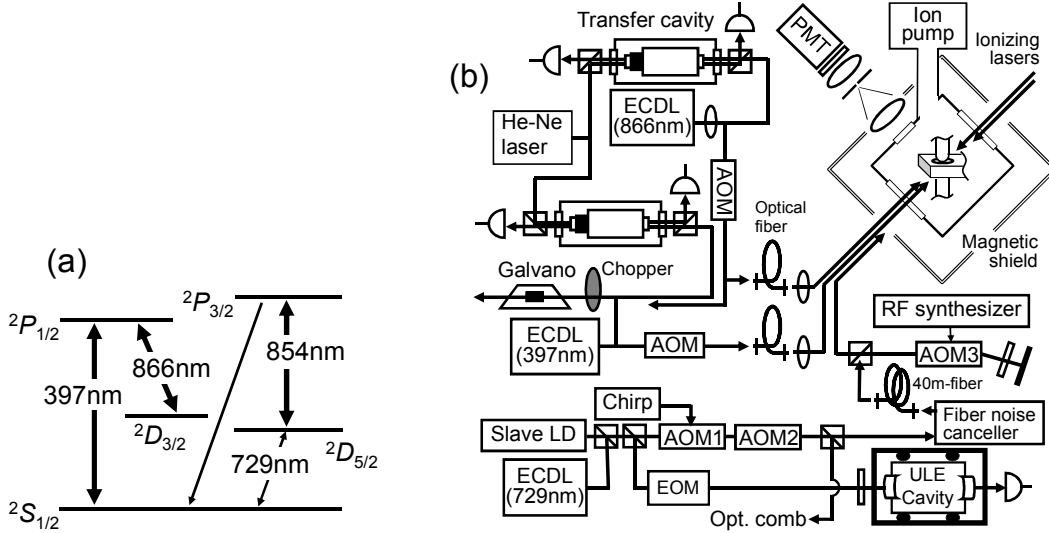
## 1. Introduction

As candidates for next generation definitions of the second, various efforts are being made to develop optical clocks with uncertainty better than that of the cesium standard. The best uncertainty of  $8.6 \times 10^{-18}$  has been reported in a composite quantum system using  $^{27}\text{Al}^+$  ions [1]. However, this requires sophisticated quantum logic operation and it is not easily accessible. Simpler clocks with modest accuracies have also been developed using Alkali-like ions ( $^{88}\text{Sr}^+$ ,  $^{177}\text{Yb}^+$ ). We are developing an optical clock using single  $^{40}\text{Ca}^+$  ions [2]. The energy levels of  $^{40}\text{Ca}^+$  that are relevant to the operation of the frequency standard are shown in Fig. 1(a). The lifetime of its  $^2D_{5/2}$  state is about 1.2 seconds, which gives a high line Q to the  $^2S_{1/2}$  -  $^2D_{5/2}$  transition at 729 nm (411 THz). In 2008 we reported the absolute frequency of this transition with an uncertainty of  $4 \times 10^{-14}$ . In the next year, it was also reported by M. Chwalla *et al.* with an improved uncertainty of  $10^{-15}$  level [3]. These two reports contributed to acceptance of this transition to the 2009 CCTF list of recommended radiation. After that, we measured the absolute frequency to be 411 042 129 776 395 ( $\pm 5$ ) Hz [4]. These measurements are consistent with each other within the uncertainties. This paper reports on our improved frequency measurement towards an uncertainty of  $10^{-15}$  level, as well as the frequency stability measurement of the clock laser stabilized by the  $^{40}\text{Ca}^+$  clock transition.

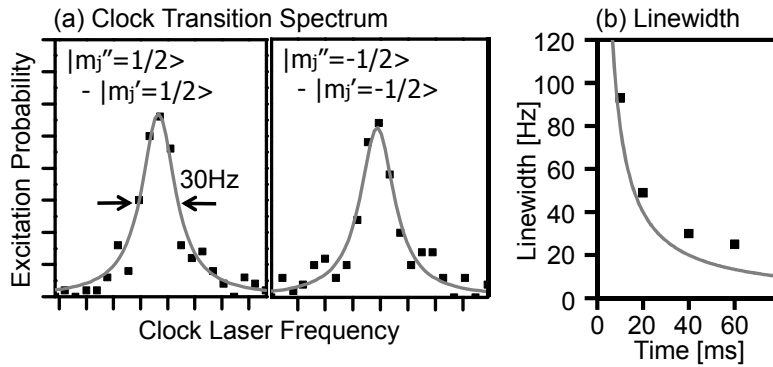
In this paper, we first outline the experimental setup, then explain the clock transition measurement, frequency lock to the clock transition, and absolute frequency measurement. Finally, a conclusion is described.

## 2. Experimental setup

The experimental setup is shown in Fig. 1(b), which is based on the setup described in our previous paper [2]. The output of the 729-nm clock laser is distributed to the ion-trap chamber by a 40-m PM optical fiber. To minimize the phase noise caused by the fiber transfer, we employed a noise cancellation system [5]. The clock transition spectrum is observed by the electron shelving method [6]. A stable magnetic field is used to resolve the clock transition into ten Zeeman components. In our previous measurement, the observed linewidth was broadened to a few hundred hertz mainly due to the ambient magnetic field fluctuation. We therefore developed an ion trap chamber equipped with a two-layer magnetic shield. This shield decreases the amplitude of the magnetic field fluctuation by a factor of more than 20. A typical clock transition spectrum is shown in Fig. 2(a). It shows a pair of the Zeeman components of  $|^2S_{1/2}, M_J=1/2\rangle$  -  $|^2D_{5/2}, M_J=1/2\rangle$  and  $|^2S_{1/2}, -1/2\rangle$  -  $|^2D_{5/2}, -1/2\rangle$  transitions. Averaging these two transition frequencies, the first order Zeeman shift is cancelled. The linewidth is about 30 Hz when the interrogation time between the ion and the 729-nm light is 40 ms. The linewidth observed at various interrogation times is shown in Fig. 2(b). The linewidth was limited by the transit-time broadening using a rectangular light pulse. Introducing the magnetic shield, the observed linewidth has reduced to about one-tenth of that previously observed.



**Figure 1** (a) Partial term energy diagram of  $^{40}\text{Ca}^+$  ions. (b) Experimental setup for  $\text{Ca}^+$  ions. AOM: acousto-optic modulator; EOM: electro-optic modulator; PMT: photo-multiplier tube; Galvano: Galvano tube. Roles of AOM1~3 are described in the text.



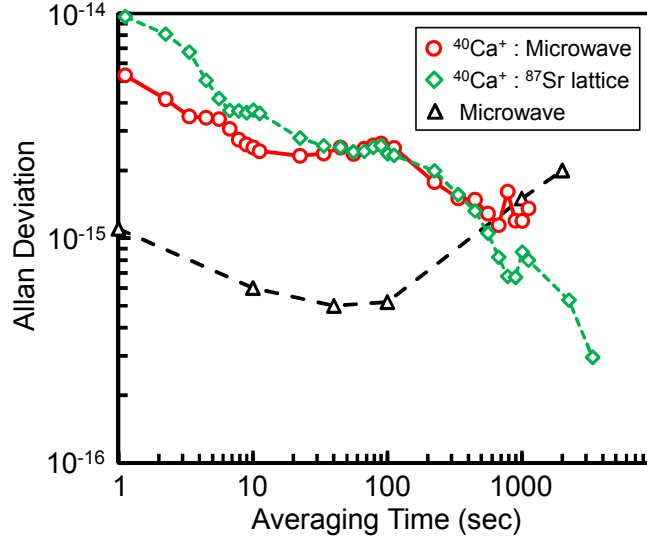
**Figure 2** (a) Typical clock transition spectra and their Lorentz fitting. (b) Linewidth as the function of interrogating time of the clock laser. Gray line shows theoretical transit-time broadening using rectangular light pulses.

### 3. Frequency Stability Measurement

In order to measure the frequency stability of the  $^{40}\text{Ca}^+$  optical clock, the 729-nm laser frequency was stabilized by the clock transition. The laser frequency is locked to the average of the Zeeman component pairs shown in Fig. 2(a) to cancel the first order Zeeman shift. An optical comb measures the laser frequency using a cryogenic sapphire oscillator (CSO) [7] as the microwave frequency reference. The 729-nm clock frequency is stabilized by an ultra-low-expansion (ULE) optical cavity, which is shown in Fig. 1(b), in the short term. However, the resonance frequency of the ULE cavity has a long-term drift at a slow rate of about 0.03 Hz/s. In addition, very small thermal instability of the cavity also causes a frequency drift. The long-term linear drift is reduced by frequency chirp using an acousto-optic modulator (AOM1 in Fig. 1(b)). The residual drift is detected by measuring the clock transition using the second AOM (AOM2) for frequency scanning. This drift is compensated by the third AOM (AOM3) for frequency scanning. The measured stability is shown in Fig. 3 (circles). We believe that the Allan deviation of  $1 \times 10^{-15}$  at an averaging time of 1000 seconds is limited by the frequency stability of the microwave reference.

To measure the frequency stability of the  $^{40}\text{Ca}^+$  clock at more than 1000 seconds we compared the 729-nm laser frequency locked by the  $^{40}\text{Ca}^+$  transition with a 698-nm laser frequency locked by the  $^{87}\text{Sr}$  clock transition. The  $^{87}\text{Sr}$

transition is measured using the optical lattice technique [8]. The measured Allan deviation is also shown in Fig. 3. An Allan deviation of  $7 \times 10^{-16}$  at 1000 s was measured, an improvement of about one digit over previous measurements limited to about  $5 \times 10^{-15}$  at 1000 s without magnetic shielding.



**Figure 3** Frequency stability evaluated by Allan deviation between the  $^{40}\text{Ca}^+$  clock, the  $^{87}\text{Sr}$  lattice clock and the microwave frequency standard. Circles (red): Allan deviation between the  $^{40}\text{Ca}^+$  clock and the microwave standard; Squares (green): between the  $^{40}\text{Ca}^+$  and the  $^{87}\text{Sr}$  clock; Triangles (black): Allan deviation of the cryogenic sapphire oscillator measured by the developer (University of West Australia).

#### 4. Absolute Frequency Measurement

The absolute frequency of the clock transition is evaluated using the laser frequency locked to the average of the Zeeman component pairs, such as the pair shown in Fig. 2(a). It is measured by an optical comb using a hydrogen maser as the frequency reference. Here, the reference frequency from the Hydrogen maser is evaluated every second by the Coordinated Universal Time (UTC) maintained at NICT. The time difference between the UTC(NICT) and the SI definition of the second on the geoid surface (TAI) is corrected using the Circular T published by the BIPM.

Systematic frequency shifts are evaluated as follows. A faint leakage of 397-nm light from the AOM crystal at the interrogation between the ion and clock laser causes an ac Stark shift, and to correct this we measure the transition frequency with various 397-nm cooling laser powers. Black body radiation (BBR) shift is calculated from the estimation of the temperature of the ion trap. From the measurement of the geographical height of the ion trap from the geoid surface, the gravitational shift is calculated to be 3.4 Hz [9].

The electric quadrupole moment of the  $D_J$  state interacts with the electric field gradient, causing an additional systematic shift of a few hertz [10]. The value of the quadrupole shift ( $\Delta\nu_Q$ ) is expressed at the  $M_J$  sublevel of the  $D_J$  state using

$$\hbar\Delta\nu_Q = \frac{dE_z}{dz} \Theta(D, j) \frac{J(J+1) - 3M_J^2}{4J(2J-1)} (3 \cos^2 \beta - 1) \quad (1)$$

where  $dE_z/dz$  is the electric field gradient along the symmetry axis of the trap potential ( $z$ ),  $\beta$  is the angle between the axis  $z$  and the magnetic field, and  $\Theta(D, j)$  is the strength of the quadrupole moment in terms of a reduced matrix element. From Eq. (1), the ratio of the quadrupole shift of the  $|^2D_{5/2}, M_J = \pm 1/2\rangle$  levels to the  $|^2D_{5/2}, \pm 3/2\rangle$  levels is 4:1. Therefore, we measure two Zeeman component pairs whose excited levels are  $|^2D_{5/2}, \pm 1/2\rangle$  and  $|^2D_{5/2}, \pm 3/2\rangle$  pairs. When  $f_{3/2}$  represents the average of the  $|^2S_{1/2}, 1/2\rangle - |^2D_{5/2}, 3/2\rangle$  and  $|^2S_{1/2}, -1/2\rangle - |^2D_{5/2}, -3/2\rangle$  transition frequencies and  $f_{1/2}$  represents the average of the  $|^2S_{1/2}, 1/2\rangle - |^2D_{5/2}, 1/2\rangle$  and  $|^2S_{1/2}, -1/2\rangle - |^2D_{5/2}, -1/2\rangle$  frequencies, we calculate the transition frequency corrected for the quadrupole shift by using the following equation,

$$f = (4/3)f_{3/2} - (1/3)f_{1/2}. \quad (2)$$

We also consider other contributions to the systematic frequency shift, such as the quadratic Zeeman shift. In our previous measurement, the total uncertainty of  $\pm 5$  Hz ( $1.2 \times 10^{-14}$ ) is limited mainly by its statistical uncertainty (type A error) because of the observed linewidth of about 300 Hz. After introducing the magnetic shield, the linewidth was decreased to about 30 Hz, and the Allan deviation of the frequency stability was improved by about one digit. From this stability improvement, it is estimated that the  $^{40}\text{Ca}^+$  clock monitors the frequency change of the UTC(NICT) with an uncertainty smaller than  $1 \times 10^{-15}$  level. In the absolute frequency measurement, therefore, we can expect the maximum improvement in the total uncertainty of about one digit. Making full use of the advantage of the improved stability, we are trying to re-evaluate the systematic shifts, such as the quadrupole shift and the ac Stark shifts. The link uncertainty between the UTC(NICT) and TAI might be also a problem. Taking all effects into account, we will evaluate the absolute transition frequency, aiming at a total uncertainty low parts in  $10^{-15}$ .

## 5. Conclusion

We have improved our experimental setup using an ion trap equipped with a magnetic shield. The linewidth of the  $^2S_{1/2} - ^2D_{5/2}$  clock transition was decreased to about 30 Hz. Locking the clock laser frequency to the clock transition, the Allan deviance of  $7 \times 10^{-16}$  at 1000 s was measured. We now measure the transition frequency in order to evaluate the absolute frequency with an uncertainty of  $10^{-15}$  level.

## 6. Acknowledgments

The  $^{87}\text{Sr}$  optical lattice clock at NICT is developed by Dr. Atsushi Yamaguchi and Dr. Tetsuya Ido. The CSO was developed at the University of West Australia [7] and it is maintained by Dr. Motohiro Kumagai. The authors would like to express their appreciation to these collaborators.

## 7. References

1. C.-W. Chou, D. B. Hume, J. C. J. Koelemeij, D. J. Wineland, and T. Rosenband, "Frequency Comparison of Two High-Accuracy  $\text{Al}^+$  Optical Clocks," *Phys. Rev. Lett.*, **104**, 2010, 070802.
2. K. Matsubara, K. Hayasaka, Y. Li, H. Ito, S. Nagano, M. Kajita, and M. Hosokawa, "Frequency Measurement of Optical Clock Transition of  $^{40}\text{Ca}^+$  Ions with an Uncertainty of  $10^{-14}$  Level," *Appl. Phys. Express*, **1**, 2008, 067011.
3. M. Chwalla, J. Benhelm, K. Kim, G. Kirchmair, T. Monz, M. Riebe, P. Schindler, A. S. Villar, W. Hänsel, C. F. Roos, R. Blatt, M. Abgrall, G. Santarelli, G. D. Rovera, and Ph. Laurent, "Absolute Frequency Measurement of the  $^{40}\text{Ca}^+ 4s^2S_{1/2} - 3d^2D_{5/2}$  Clock Transition," *Phys. Rev. Lett.* **102**, 2009, 023002.
4. K. Matsubara, Y. Li, S. Nagano, H. Ito, M. Kajita, R. Kojima, K. Hayasaka, and M. Hosokawa, "Frequency Stability Measurement of a  $^{40}\text{Ca}^+$  Optical Clock," *Asia-Pacific Radio Science Conference*, Toyama, Japan, 2010, A3a-3.
5. L.-S. Ma, Z. Y. Bi, A. Bartels, L. Robertsson, M. Zucco, R. S. Windeler, G. Wilpers, C. Oates, L. Hollberg, and S. A. Diddams, "Optical Frequency Synthesis and Comparison with Uncertainty at the  $10^{-19}$  Level," *Science*, **303**, 2004, pp. 1843–1845.
6. H. Dehmelt, "Mono-Ion Oscillator as Potential Ultimate Laser Frequency Standard," *IEEE Trans. Instrum. Meas.*, IM-31, 1982, pp. 83–87.
7. C. R. Locke, E. N. Ivanov, J. G. Hartnett, P. L. Stanwix, and M. E. Tobar, "Design techniques and noise properties of ultrastable cryogenically cooled sapphire-dielectric resonator oscillators" *Rev. Sci. Instrum.*, **79**, 2008, 051301.
8. M. Takamoto, F. Hong, R. Higashi, and H. Katori, "An optical lattice clock," *Nature*, **435**, 2005, pp. 321-324
9. M. Kumagai, H. Ito, M. Kajita, and M. Hosokawa, "Evaluation of caesium atomic fountain NICT-CsF1," *Metrologia*, **45**, 2008, pp. 139–148.
10. C. F. Roos, M. Chwalla, K. Kim, M. Riebe, and R. Blatt, "Designer atoms for quantum metrology," *Nature*, **443**, 2006, pp. 316–319.