CHIN. PHYS. LETT. Vol. 35, No. 7 (2018) 074202

Improvement of Stability of $^{40}Ca^{+}$ Optical Clock with State Preparation *

Meng-Yan Zeng(曾孟彦)[1,2,3,4], Yao Huang(黄垚)[1,2,3], Hu Shao(邵虎)[1,2,3,4], Miao Wang(王淼)[1,2,3,4], Hua-Qing Zhang(张华青)[1,2,3,4], Bao-Lin Zhang(张宝林)[1,2,3,4], Hua Guan(管桦)[1,2,3], Ke-Lin Gao(高克林)[1,2,3]**

[1]*State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071*

[2]*Key Laboratory of Atomic Frequency Standards, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071*

[3]*Center for Cold Atom Physics, Chinese Academy of Sciences, Wuhan 430071*

[4]*University of Chinese Academy of Sciences, Beijing 100049*

(Received 26 March 2018)

Stability is one of most important performances of an atomic clock. Here we describe our recent work on improving the stability of our $^{40}Ca^{+}$ optical clock. State preparation is adopted to transfer the ion to the ground-state magnetic sublevel of the clock transition, after the quenching laser transfers the ion to the ground state at each cycle. Using this method, the stability for $^{40}Ca^{+}$ optical clock is improved to about $6.3 \times 10^{-15}/\sqrt{\tau}$. Compared with $1.0 \times 10^{-14}/\sqrt{\tau}$ in previous work, the averaging time is decreased to reach a given level of statistical uncertainty in a clock comparison.

PACS: 42.62.Fi, 37.10.Ty, 95.55.Sh DOI: 10.1088/0256-307X/35/7/074202

Optical clocks based on narrow linewidth forbidden transitions have demonstrated great improvements in recent years, with both single-ion optical clocks[1–5] and optical lattice clocks.[6–13] Up to now, some of the optical clocks have already achieved uncertainties reaching the 10^{-17} or even 10^{-18} level in fractional frequency units, which is smaller than the state-of-the-art Cs primary standard.[14,15] Moreover, the stabilities of some optical clocks exceed the Cs fountains.

For optical clocks based on laser-cooled trapped ions, different clock transition frequencies have been measured in various laboratories around the world, such as Ba^{+}, Sr^{+}, Hg^{+}, In^{+}, Al^{+}, Yb^{+}, and Ca^{+}.[1–5,16–21] For the ion-based optical clocks, the smallest uncertainty achieved is 3×10^{-18}[17] based on the Yb^{+} ion. On the other hand, great improvements on clock stabilities have also been demonstrated recently. Optical lattice clocks based on numerous neutral atoms, which can achieve better stabilities with the higher signal-to-noise ratio than single-ion optical clocks. To date, the clocks with the best stability are based on Sr atoms in JILA and Yb atoms in NIST with a few parts in the 10^{-18} level at ∼10000 s.[6,7,12] With quantum-degenerate atoms frozen into a 3D lattice, a synchronous clock comparison between two regions of the 3D lattice yields an instability of $3.1 \times 10^{-17}/\sqrt{\tau}$ and reach 5×10^{-19} in 1 h of averaging time.[30] Stability of $2.0 \times 10^{-15}/\sqrt{\tau}$ has been achieved based on the comparison of two Al^{+} quantum logic clocks in NIST.[1]

Optical clocks based on the $4s^2S_{1/2}$–$3d^2D_{5/2}$ electric quadrupole transition of a single trapped $^{40}Ca^{+}$ ion have been studied by our group in WIPM and other laboratories around the world.[18,19,31,32] In comparison with optical clocks in the previous statement, the $^{40}Ca^{+}$ optical clock has its own advantages. Its simplicity on the energy-level scheme (Fig. 1) and ultra-narrow natural linewidth of 0.2 Hz[23] on the $4s^2S_{1/2}$–$3d^2D_{5/2}$ clock transition allow the possibility of making an optical clock with good potential accuracy and low systematic shifts. What is more, there is a good prospect for a low-cost, compact, and robust $^{40}Ca^{+}$ optical clock due to its commercially available diode laser system.

We recently reduced the uncertainty of our $^{40}Ca^{+}$ optical clocks to 5×10^{-17}.[18] With the improvement of the uncertainty, the demands on clock stability have also increased dramatically. Limited by the stability of the clock laser and control of the optical and electromagnetic field geometry and laboratory conditions, it took 20000 s of averaging time for the stability to reach 7×10^{-17} in previous work. In this study, we present recent results of the optimization of the frequency stability of our optical clock. State preparation is adopted to transfer the ion to the ground-state magnetic sublevel of the clock transition, after the quenching laser transfers the ion to the ground state at each cycle. Compared with our previous work with a fractional stability of $1.0 \times 10^{-14}/\sqrt{\tau}$,[18] a fractional stability of 6.3×10^{-17} at ∼10000 s is achieved. Details of the sequencing of cooling and probe pulses, state preparation step, comparison of the two clocks and stability measurements are presented.

*Supported by the National Key R&D Program of China under Grant No 2017YFA0304401, the National Natural Science Foundation of China under Grant Nos 91336211, 11634013, 11474318 and 11622434, the Strategic Priority Research Program B of Chinese Academy of Sciences under Grant No XDB21030100, the Hubei Provincial Science Fund for Distinguished Young Scholars under Grant No 2017CFA040, and the Youth Innovation Promotion Association of Chinese Academy of Sciences under Grant No 2015274.

**Corresponding author. Email: klgao@wipm.ac.cn

© 2018 Chinese Physical Society and IOP Publishing Ltd

Our experiment is implemented with a laser cooled single $^{40}Ca^{+}$ ion, which is trapped in miniature Paul ring traps–both are driven at $\sim 2\pi \times 24.7$ MHz at which frequency the micromotion-induced Stark shifts and 2nd-order Doppler shifts cancel each other.[2,22] For each trap, the ambient magnetic field is reduced by two layers of magnetic shielding and then compensated by adjusting the current on three pairs of coils. For our experimental conditions, eventually the background magnetic field is set to be $\sim$0.5 μT.

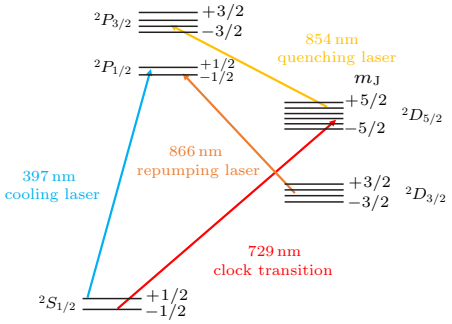

Fig. 1. The energy-level diagram of the $^{40}Ca^{+}$ ion, with the transitions and laser wavelengths used to operate the optical frequency standard. The optical clock is realized by probing the $5s^2S_{1/2}$–$4d^2D_{5/2}$ electric-quadrupole transition at 729 nm.

After loading the ion with a two-step photoionization scheme on the neutral atoms of calcium emitted from a resistively heated oven, two laser beams (397 nm and 866 nm) enter through a viewport of the vacuum chamber to achieve the Doppler cooling of the ion. The ion's kinetic motion is cooled using a frequency-stabilized diode laser red detuned from the $4s^2S_{1/2}$–$4p^2P_{1/2}$ dipole transition at 397 nm. A repumping laser at 866 nm prevents the ion from decaying to the metastable $^2D_{3/2}$ state to maintain the cooling cycle. Both the 397-nm and the 866-nm lasers are stabilized to an ultra-stable laser source at 729 nm using transfer cavities. The clock transition Zeeman spectrum is then probed using an ultra-stable laser source at 729 nm. A more detailed description of the ion trap and the associated laser systems for photon ionization, cooling, quenching, detection, and probing has been given in previous works.[25–27]

Recent work has focused on optimizing the frequency stability of our $^{40}Ca^{+}$ ion frequency standard. It is convenient to measure the stability of our two optical clocks through the clock comparison. For cancellation of the linear Zeeman shift, the quadrupole shift, and the tensor part of the Stark shifts,[24] the two clocks are running synchronously by measuring the six chosen Zeeman transitions with $M_J = \pm 1/2$, $M_J = \pm 3/2$ and $M_J = \pm 5/2$ for the $D_{5/2}$ level one after another. Due to the equal ion populations of the $M_J = \pm 1/2$ ground-state magnetic sublevels, the probe laser can excite the ion with 50% probability for interrogation of each Zeeman transition, thus degrading the lock stability. It means that the stability of our optical clocks could be improved by a factor of $\sqrt{2}$ as the consequence of the binomial statistics of the quantum jumps in case, preparing the $^{40}Ca^{+}$ ion to the relevant ground-state magnetic sublevel before interrogation.

There are a few ways to improve the ion clock stability, including obtaining narrower clock transitions using better clock lasers: increasing the duty cycle,[1] using the Ramsey scheme instead of the Rabi scheme when probing,[17,38] and using the state preparation technique.[34–38] The method chosen to achieve the state preparation depends on the energy-level structure of the ion and the experimental requirements. For instance, it is easy to achieve the state-preparation of $^{171}Yb^{+}$ by turning the repumping laser off before the cooling laser.[34] The $^{88}Sr^{+}$ and several other ions are state prepared in other ways such as frequency-resolved optical pumping on the clock transition[35] or optical pumping using circularly polarized light on the cooling transition.[36] A quantum logic method has also been adopted to achieve the state preparation of the $^{27}Al^{+}$ ion.[37]

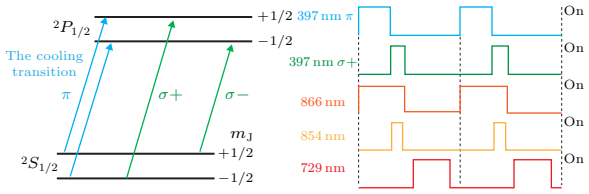

Fig. 2. (a) The Doppler cooling cycle of the $^2S_{1/2}$–$^2P_{1/2}$ transition is chosen to achieve the state-preparation of the $^{40}Ca^{+}$ ion. (b) A simplified pulse sequence for the interrogation cycle. A diode laser at 854 nm called the quenching laser is used to clear out the $^2D_{5/2}$ metastable state at each cycle, and reset the ion to the ground state before the next interrogation pulse. The polarization of the 397 nm cooling laser is briefly switched from linear to circular at the end of the cooling pulse for state preparation.

The state-preparation method implemented in our current setup is using circularly polarized light on the cooling transition. Interacting with the linearly polarized cooling laser, the $^{40}Ca^{+}$ ion can be populated from two ground-state magnetic sublevels with equal probabilities and decay to either Zeeman ground state from the upper $^2P_{1/2}$ level rapidly. However, the cooling laser only drives the $^2S_{1/2}(m = -1/2)$–$^2P_{1/2}(m = +1/2)$ transition when it is switched to the $\sigma+$ polarized light, leaving the ion in the $^2S_{1/2}(m = +1/2)$ ground state. The $\sigma-$ cooling laser prepares the $^{40}Ca^{+}$ ion into the $^2S_{1/2}(m = -1/2)$ ground state in a similar way. When the cooling laser is circularly polarized, the fluorescence of detection vanishes immediately, indicating that the state preparation has been achieved.

In our system, the $^{40}Ca^{+}$ state preparation is achieved by an electro-optic modulator (EOM) followed by a quarter-wave plate providing a voltage-controlled wave plate on the 397 nm beam path. The dc drive voltage of the EOM is output through a bleeder circuit controlled by three TTL input lines, which provide three different output voltages to the

modulator. The voltage applied determines whether the polarization of cooling laser at the ion is linear, $\sigma-$, or $\sigma+$. After the fluorescence vanished it takes <0.1 ms for state preparation typically, therefore state preparation starts 1 ms before the 397 nm laser is turned off to ensure optical pumping of the ion into the ground-state magnetic sublevel of the correlative probed transition.

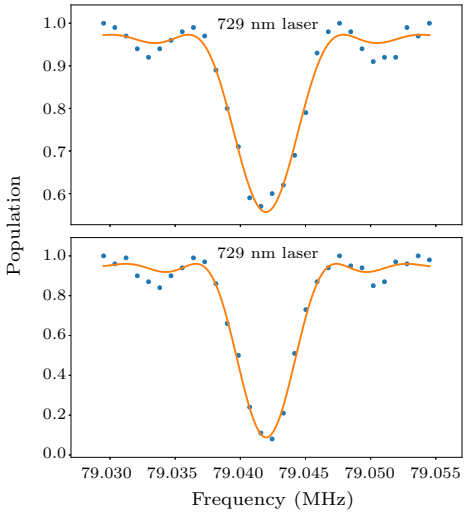

Fig. 3. The polarization of cooling laser required for ground state selection is different for the higher and lower frequency Zeeman components of clock transitions. During a full servo cycle to the ion, the quantum jump rate is sampled on the chosen three pairs of symmetrical components. The first picture shows the population of $^2S_{1/2}(m=-1/2)$–$^2D_{5/2}(m=-1/2)$ clock transition without state preparation. The second picture shows the effect of using state preparation with $\sigma-$ light on the transition. In the latter situation, the software changes the polarization of the cooling laser accordingly. The ion is transferred to $^2S_{1/2}(m=-1/2)$ with ≈92% efficiency.

Stability is a crucial performance parameter of an optical clock that determines the averaging time required to reach a given level of statistical uncertainty in a frequency measurement. The stability measurement result of the clock can be given after the comparison experiments of optical clocks. Such work has been performed with the same kind of atoms or ions[1,3,7,8,12,28] as well as different kinds.[16,19,29] In our experiment, the two clocks share the same lasers, including the cooling laser, the repumping laser, the quenching laser, and the clock laser. The clock laser is split into two beams passing through two individual AOMs and then sent into the two ion traps for detection. The servos provide feedback with the two AOMs to keep the laser frequency locked to the six chosen clock transitions.

For the longest continuous data sets (> 2 d), the Allan deviation of the frequency difference between two traps was calculated, and the mean result is shown in Fig. 4. Considering that the common mode contribution for both clocks is small relative to the total uncertainty, the Allan deviation for the frequency difference between the two $^{40}Ca^+$ clocks is simply divided by $\sqrt{2}$ to reflect the performance of the single clock. For this comparison, the pulse time chosen for the probe laser was 80 ms. The stability of our $^{40}Ca^+$ single-ion standard is $6.3\times10^{-15}/\sqrt{\tau}$. Compared with our previous value of $1.0\times10^{-14}/\sqrt{\tau}$, the averaging time is decreased for a given statistical uncertainty level.

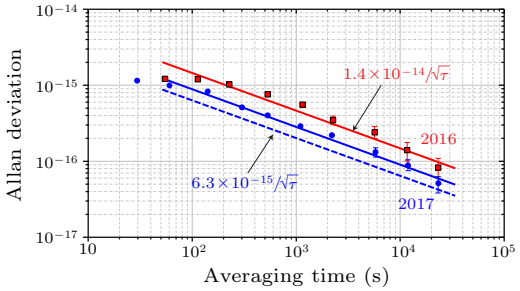

Fig. 4. Allan deviation as a function of averaging time obtained from comparison experiments between two $^{40}Ca^+$ ion clocks. Compared with the results of our previous work in 2016 shown with the red line, the solid blue line representing the data of comparison measurements has been improved by a factor of $\sqrt{2}$ using state preparation in our recent work in 2017. The Allan deviation was divided by $\sqrt{2}$ to obtain the single-clock performance (dashed blue line).

In summary, we have presented a method of optimization for the frequency stability of our $^{40}Ca^+$ optical clock. The results of measurement show that the two clocks have both reached the stability of $6.3\times10^{-15}/\sqrt{\tau}$, which is a factor of $\sqrt{2}$ better than our previous result.[18] The improvements are made by preparing the ion in the ground-state magnetic sublevel of the probed transition before each probing cycle. Compared with the best stability demonstrated for a single-ion clock of $2.0\times10^{-15}/\sqrt{\tau}$,[1] the stability of the $^{40}Ca^+$ clock achieves the same order of magnitude. However, with higher clock transition frequency and narrower linewidth about 1 Hz of the clock laser, the stability of the $^{27}Al^+$ clock is approximately three times better than our measurement result.[1] Considering that the clock transition frequency is determined by the reference ion alone and the observed clock transition linewidth is 10 Hz, further optimization will work on obtaining the narrower linewidth 729 nm clock laser with an order of magnitude decrease. What is more, the maximum probe time and thus the stability of the clocks are limited by the coherence time of the laser and the atom. The coherence time of the atom is limited by the heating rate of the ions in the trap and ac magnetic field fluctuations.

In the future, a better design of the magnetic shielding and traps with lower heating rates will increase the coherence time, and thus will improve the stability. Other avenues such as Ramsey pulse interrogation, reduction of frequency noise, choosing a longer interrogation time, and a clock laser with better stability than the current work could make it possible to reduce the stability of the clock even further, and the stability can be further improved by making a clock

based on multiple ions.[33]

References

[1] Chou C W, Hume D B, Koelemeij J C J et al 2010 *Phys. Rev. Lett.* **104** 070802
[2] Madej A A, Dube P, Zhou Z et al 2012 *Phys. Rev. Lett.* **109** 203002
[3] Barwood G P, Huang G, Klein H A et al 2014 *Phys. Rev.* A **89** 050501
[4] Huntemann N, Okhapkin M, Lipphardt B et al 2012 *Phys. Rev. Lett.* **108** 090801
[5] King S A, Godun R M, Webster S A et al 2012 *New J. Phys.* **14** 013045
[6] Bloom B J, Nicholson T L, Williams J R et al 2014 *Nature* **506** 71
[7] Nicholson T L, Campbell S L, Hutson R B et al 2015 *Nat. Commun.* **6** 6896
[8] Ushijima I, Takamoto M, Das M et al 2015 *Nat. Photon.* **9** 185
[9] Falke S, Lemke N, Grebing C et al 2014 *New J. Phys.* **16** 073023
[10] Targat R Le, Lorini L, Coq Y Le et al 2013 *Nat. Commun.* **4** 2109
[11] McFerran J J, Yi L, Mejri S et al 2014 *Phys. Rev.* A **89** 043432
[12] Hinkley N, Sherman J A, Phillips N B et al 2013 *Science* **341** 1215
[13] Beloy K, Hinkley N, Phillips N B et al 2014 *Phys. Rev. Lett.* **113** 260801
[14] Li R, Gibble K and Szymaniec K 2011 *Metrologia* **48** 283
[15] Weyers S, Gerginov V, Nemitz N et al 2012 *Metrologia* **49** 82
[16] Rosenb, T, Hume D B, Schmidt P O et al 2008 *Science* **319** 1808
[17] Huntemann N, Sanner C, Lipphardt B et al 2016 *Phys. Rev. Lett.* **116** 063001
[18] Huang Y, Guan H, Liu P et al 2016 *Phys. Rev. Lett.* **116** 013001
[19] Matsubara K, Hachisu H, Li Y et al 2012 *Opt. Express* **20** 22034
[20] Sherman J A, Koerber T W, Markhotok A et al 2005 *Phys. Rev. Lett.* **94** 243001
[21] Hayasaka K 2012 *Appl. Phys.* B **107** 965
[22] Berkel, D J, Miller J D, Bergquist J C et al 1998 *J. Appl. Phys.* **83** 5025
[23] Barton P A, Donald C J S, Lucas D M et al 2000 *Phys. Rev.* A **62** 032503
[24] Dubé P, Madej A A, Zhou Z et al 2013 *Phys. Rev.* A **87** 023806
[25] Huang Y, Liu Q, Cao J et al 2011 *Phys. Rev.* A **84** 053841
[26] Huang Y, Cao J, Liu P et al 2012 *Phys. Rev.* A **85** 030503
[27] Huang Y, Liu P, Bian W et al 2014 *Appl. Phys.* B **114** 189
[28] Schneider T, Peik E and Tamm C 2005 *Phys. Rev. Lett.* **94** 230801
[29] Ludlow A D, Zelevinsky T, Zawada M et al 2008 *Science* **319** 1805
[30] Campbell S L, Hutson R B, Marti G E et al 2017 *Science* **358** 90
[31] Chwalla M, Benhelm J, Kim K et al 2009 *Phys. Rev. Lett.* **102** 023002
[32] Champenois C, Houssin M, Lisowski C et al 2004 *Phys. Lett.* A **331** 298
[33] Keller J, Burgermeister T, Kalincev D et al 2016 *J. Phys.: Conf. Ser.* **723** 012027
[34] Tamm C, Engelke D, Buhner V et al 2000 *Phys. Rev.* A **61** 053405
[35] Roos C F, Chwalla M, Kim K et al 2006 *Nature* **443** 316
[36] Barwood G P, Gill P, Huang G et al 2012 *Meas. Sci. Technol.* **23** 055201
[37] Schmidt P O, Rosenb, T, Langer C et al 2005 *Science* **309** 749
[38] Dube P, Madej A A, Shiner A et al 2015 *Phys. Rev.* A **92** 042119