Appl. Phys. B (2014) 114:183–187
DOI 10.1007/s00340-013-5679-8

Applied Physics B
Lasers and Optics

Toward a transportable microwave frequency standard based on laser-cooled $^{113}Cd^{+}$ ions

J. W. Zhang · S. G. Wang · K. Miao · Z. B. Wang · L. J. Wang

Received: 31 May 2013 / Accepted: 3 October 2013 / Published online: 20 October 2013

Abstract A transportable microwave frequency standard based on laser-cooled $^{113}Cd^{+}$ ions is introduced, and its working principle and potential performance are discussed. Based on the experimental setup developed at JMI, the frequency of the ground-state hyperfine splitting of $^{113}Cd^{+}$ is measured to within milliHertz uncertainty and its frequency stability is obtained.

1 Introduction

The three-dimensional (3D) quadrupole ion trap, known as the Paul trap, was invented by Wolfgang Paul in 1953. Since then, its use has rapidly expanded into many areas of application. Because the ions are confined in the ion trap and isolated from external fields, they provide an appropriate method for precision frequency metrology. Recently, optical clocks with a fractional frequency uncertainty of 8.6×10^{-18} and 2×10^{-17} based on Al^{+} [1] and $^{88}Sr^{+}$ [2], respectively, have been reported. Thus, a better performance from these optical clocks over cesium fountains, the current realization of the SI second, would enable the unit of time to be redefined in the future.

Ion traps are also applied in precision microwave spectroscopy owing to the long interaction time between the radiation field and the trapped ions. Several groups have successfully demonstrated microwave frequency standards based on trapped ions, including $^{199}Hg^{+}$, $^{171}Yb^{+}$, $^{137}Ba^{+}$, and $^{9}Be^{+}$ [3]. Most of the early ion microwave frequency standards are based on the hyperbolic Paul traps, spectral lamps, and buffer gas cooling. After the linear Paul trap was introduced to frequency standards [4] and the laser cooling was applied in ion traps [5], the performance of microwave frequency standards based on ions has been improved greatly. For instance, the frequency stability of the $^{199}Hg^{+}$ microwave frequency standard [6] was achieved at $3.3 \times 10^{-13}\tau^{-1/2}$ with the fractional frequency uncertainty to the level of 1×10^{-14}. And the stability of the $^{171}Yb^{+}$ microwave frequency standard [7] was predicted to be $5 \times 10^{-14}\tau^{-1/2}$ with its uncertainty of 4×10^{-15}. Furthermore, researchers at the Jet Propulsion Laboratory (JPL) have demonstrated an excellent stability of $5 \times 10^{-14}\tau^{-1/2}$ and a drift of $<2.7 \times 10^{-17}$/day [8] based on $^{199}Hg^{+}$ ions stored in a multi-pole linear ion trap [9] and excited by a discharge lamp. A very compact $^{199}Hg^{+}$ frequency standards for space applications has also been built at JPL [10].

Although the optical clocks based on trapped ions are extremely accurate, their use so far has been confined to laboratories. In many applications, e.g., precise frequency comparisons between different atomic clocks located in different laboratories using a transportable clock, the transportability of the transportable clock is of equal

J. W. Zhang · S. G. Wang · K. Miao · Z. B. Wang · L. J. Wang (✉)
NIM-THU Joint Institute for Measurement Science (JMI), Tsinghua University, Beijing 100084, China
e-mail: lwan@tsinghua.edu.cn

J. W. Zhang
e-mail: pooncn@gmail.com

J. W. Zhang · S. G. Wang · K. Miao · Z. B. Wang · L. J. Wang
Department of Precision Instrument, Tsinghua University, Beijing 100084, China

J. W. Zhang · Z. B. Wang · L. J. Wang
State Key Laboratory of Precision Measurement Technology and Instruments, Tsinghua University, Beijing 100084, China

L. J. Wang
Department of Physics, Tsinghua University, Beijing 100084, China

importance compared to the precision. One of the successful demonstrations of transportable frequency standards is the transportable cesium fountain clock developed by LNE-SYTRE in France [11]. Nevertheless, the microwave frequency standards based on trapped ions have superb performance in term of frequency stability and accuracy, and the ion traps are more accessible. Hence, a transportable microwave frequency standard based on trapped ions promises to be an excellent alternative to transportable fountain clocks.

Some of the ion species that are suitable for microwave frequency standards have their $^2D_{3/2}$ or $^2D_{5/2}$ long-lifetime states lying between the ground state $^2S_{1/2}$ and the excited states $^2P_{1/2}$ or $^2P_{3/2}$, such as $^{171}Yb^+$ and $^{137}Ba^+$. To speed up the cooling process for these ions, a repump laser, in addition to the cooling laser, is required to prevent the ions from being trapped in these metastable states. However, the $^{113}Cd^+$ ion is beyond this category, and its D states are more energetically favored over the P states. Hence, one laser is sufficient for efficient laser cooling. Moreover, its ground-state hyperfine splitting is about 15.2 GHz, larger than those of most of the other ion species. Hence, the $^{113}Cd^+$ ion is an ideal choice for building a transportable microwave frequency standard. A cadmium ion clock has been proposed before [12, 13]. In this paper, we report a transportable laser-cooled cadmium ion clock developed in our laboratory and discuss the working principle, the experimental setup, the measurement of the clock transition, and the current results of frequency stability results.

2 General descriptions

Figure 1 shows the energy levels of $^{113}Cd^+$ ions. The clock transition corresponds to the hyperfine splitting of the ground state, with frequency of $\nu_{00} \approx 15.2$ GHz. The 214.5 nm $^2S_{1/2}(F = 1, m_F = 1) \leftrightarrow {}^2P_{3/2}(F = 2,\ m_F = 2)$ transition is a cycling transition that can be used to cool and detect the ions. Although the circularly polarized cooling laser excites a cycling transition, ions can still leak to the $^2S_{1/2}(F = 0,\ m_F = 0)$ state via $^2P_{3/2}(F = 1)$ state resulting from the polarization impurity of the cooling laser. To speed up the cooling process, 20-dBm microwave radiation resonant with the ground-state hyperfine transition is applied during laser cooling [14]. The $^2S_{1/2}(F = 1) \leftrightarrow {}^2P_{3/2}(F = 1)$ transition is used to optically pump the ions to the $^2S_{1/2}(F = 0,\ m_F = 0)$ state. Because the hyperfine splitting of the $^2P_{3/2}$ state is about 800 MHz [12], the pump laser can be obtained by blue shifting the cooling laser with an acousto-optic modulator(AOM). Hence, only one laser is needed to realize laser cooling, ion detection,

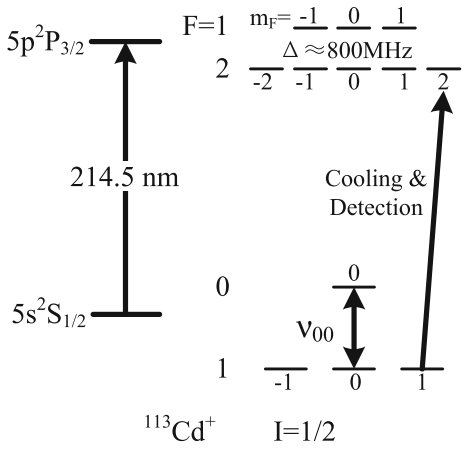

Fig. 1 Schematic of the energy levels of $^{113}Cd^+$ (not to scale). The cycling transition of $^2S_{1/2}(F = 1, m_F = 1) \leftrightarrow {}^2P_{3/2}(F = 2, m_F = 2)$ is used for laser cooling and ion detection, and the $^2S_{1/2}(F = 1) \leftrightarrow {}^2P_{3/2}(F = 1)$ transition is chosen for optical pumping

and optical pumping for the Cd^+ clock. This helps in making the Cd^+ clock transportable.

According to the Allan deviation formula for passive atomic clocks

$$\sigma_y(\tau) = \frac{1}{2\pi\nu_{00} \cdot \Delta t_R \cdot SNR}\sqrt{\frac{T_c}{\tau}} \quad (\tau \le T_c \le 2\Delta t_R), \tag{1}$$

the limit for the frequency stability of the Cd^+ clock is estimated to be $\sigma_y(\tau) = 2 \times 10^{-14}\tau^{-1/2}$ assuming $\Delta t_R = 1$ s and $SNR = 1{,}000$, where Δt_R is the time interval between the two microwave pulses in Ramsey's method of separated oscillation fields, SNR the signal-to-noise ratio, T_c the cycle time required to make a single interrogation of the error signal, and τ the sampling time. To achieve this objective, a cloud of at least 10^6 ions has to be trapped. To trap such a large number of ions, a linear quadrupole trap was applied.

The second-order Doppler frequency shift (SODFS), due to the motion of ions in the linear ion trap, is one of the main systemic frequency shifts. The total SODFS in a linear quadrupole trap is given by [9]

$$\frac{\Delta\nu_{00}^D}{\nu_{00}} = -\frac{3k_BT}{2mc^2} - \alpha_m, \tag{2}$$

where k_B is the Boltzmann constant, T the temperature of the ions, m the mass of the ion, and c the speed of light. The first term on the right-hand side of equation is the SODFS due to the thermal secular motion of ions, and the second term is due to ion micromotion. α_m is obtained by averaging the micromotion shifts across the ion cloud and can be expressed as

$$\alpha_m = N_d\frac{k_BT}{mc^2}. \tag{3}$$

In the linear quadrupole trap with only a small amount of non-interacting ions, N_d is 1 [9]. For a large cloud of 10^6 ions in a properly designed linear quadrupole trap with temperature around 1 K through laser cooling, N_d can be controlled to <3 according to our previous calculation [15]. Hence, the total SODFS is of order -3×10^{-15}. Assuming the temperature uncertainty is around 0.5 K, the fractional frequency uncertainty of SODFS is estimated to be 1.5×10^{-15}.

Another limitation of the frequency uncertainty for most microwave atomic clocks is the blackbody radiation shift (BBRS) which can be presented in the form [16, 17]

$$\frac{\Delta\nu_{00}^{BBR}}{\nu_{00}} = -\beta\left(\frac{T_a}{300}\right)^4\left[1+\epsilon\left(\frac{T_a}{300}\right)\right]^2, \tag{4}$$

where β is the fractional BBRS coefficient of a given species of atoms or ions, T_a the ambient temperature in K, and ϵ a small correction arising from the frequency distribution. β for $^{199}Hg^+$ and $^{171}Yb^+$ is $1.02(5) \times 10^{-16}$ and $9.4(5) \times 10^{-16}$, respectively [17]. Although there are no experimental or theoretical data about the BBRS of the ground-state hyperfine transition for $^{113}Cd^+$, a rough estimate of its magnitude can be derived from reported data of other ions. Comparing $^{199}Hg^+$, $^{171}Yb^+$, and $^{113}Cd^+$, we find that the D lines of these ions, the biggest contributors in the ac Stark shift owing to blackbody radiation, are all in the ultraviolet(uv) band with wavelengths of 194 (165) nm, 369 (329) nm, and 227 (214) nm for the D_1 (D_2) lines, respectively. Thus, a fair estimate of β for $^{113}Cd^+$ is $<1 \times 10^{-15}$, and the fractional frequency uncertainty from the BBRS is of order 1×10^{-16} when the uncertainty of T_a is <10 K.

The static Zeeman frequency shift is

$$\frac{\Delta\nu_{00}^{Z}}{\nu_{00}} = \frac{2KB\Delta B}{\nu_{00}}, \tag{5}$$

where $K = 2.582 \times 10^{10}$Hz/T^2 [15, 18] and B is the magnetic field induction in Tesla. By measuring the frequencies associated with the $^2S_{1/2}(F=1, m_F=\pm1) \leftrightarrow {}^2S_{1/2}(F=0,\ m_F=0)$ magnetically sensitive transitions, ΔB can be determined to $<1 \times 10^{-9}$ T. Hence, the fractional uncertainty in the frequency shift due to the Zeeman effect is $<3.5 \times 10^{-16}$.

3 Experimental setup

Because the detailed design of the ion trap has been reported elsewhere [19, 20], here we only provide a brief introduction to the design architecture (see Fig. 2). The electrodes of the linear quadrupole trap are made of oxygen-free copper, the radius of each being $R = 7.11$ mm;

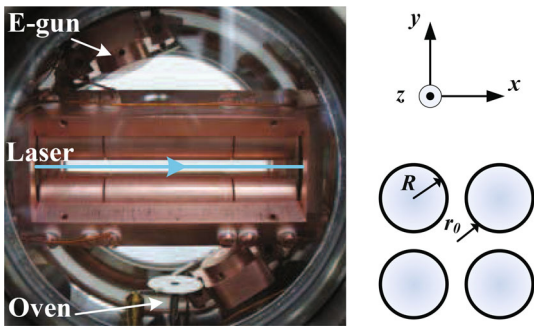

Fig. 2 Photograph of the linear ion trap installed in the vacuum chamber showing the homemade e-gun and cadmium oven (*left*), and a schematic section view of the linear ion trap (*right*)

the radius from the trap axis to the electrodes is $r_0 = 6.2$ mm, the ratio R/r_0 being optimized numerically [21]. Every electrode is cut into three segments. A dc endcap voltage is applied to the outer segments to confine the ions in the axial direction; a 1.25 MHz radio frequency voltage is applied to the inner segments of the diagonal electrodes to confine the ions in the radial direction. Small dc voltages are added on the inner segments to compensate the scattering electric fields. The ion trap is installed in an ultra-high-vacuum chamber. Silica windows are installed on the chamber to enable the laser beam to pass through the trap center along the axis of the trap and to allow the fluorescence spectra radiated by the excited ions in the x-direction to be collected. The fluorescence is detected by an electron-multiplying CCD (EMCCD) and a photon-counting photomultiplier tube (PMT). The vacuum chamber is surrounded by three Helmholtz coils, two of which are used to cancel the ambient magnetic fields in the x- and y-directions, and the third is employed to create a static magnetic field along the z-direction. To improve the long-term stability of the magnetic field, the whole physical assembly is shielded by a silicon-steel sheet box, and the current of the third Helmholtz coil is actively controlled via a fluxgate magnetometer with a 1 nT resolution. The cadmium atomic vapor is generated by slightly heating the cadmium reservoir, and the atoms given off are ionized by electron bombardment.

The 214.5 nm frequency-quadrupled diode laser system is a TA-FHG pro from Toptica Inc., Germany. An external cavity diode laser working at 858 nm produces the seed and then frequency-quadrupled to 214.5 nm by two cascade "bow-tie" doubling cavities. During the clock operation, the laser frequency must be stabilized to within a few MHz. To achieve this, the transfer cavity technique is applied to lock the 858-nm seed laser to the D_2 lines of the cesium atom [22].

The local oscillator of the Cd^+ clock is a BVA SC-cut oven-controlled crystal oscillator (OCXO) oscillating at

10 MHz. This 10 MHz reference signal is synthesized to 15.2 GHz by a commercial microwave synthesizer. The synthesizer is controlled remotely using a personal computer to set the output frequency to within a resolution of 1 mHz and the power to within a resolution of 0.1 dBm. The ions are subjected to microwave radiation applied using a horn antenna through one of the silica windows on the vacuum chamber.

4 Current results of the clock

While the red-detuned cooling laser, the e-gun, and the oven are turned on, a cloud of $^{113}Cd^{+}$ ions is trapped in approximately 2 min. The inset of Fig. 3 shows a photograph of trapped ions captured by the EMCCD. By measuring the Doppler-broadened width of the $^{2}S_{1/2}(F=1,m_F=1) \leftrightarrow {}^{2}P_{3/2}(F=2,m_F=2)$ transition, the temperature of the ions is found to be approximately 1 K. Even after the cooling laser is blocked for 10 s, the measured temperature is <5 K.

After the ions are loaded and cooled, the clock transition frequency can be measured by Ramsey's method of separated oscillatory fields; the detailed measurement sequence is given in Ref. [23, 20]. Figure 3 shows a typical measured Ramsey fringe, where the full width at half maximum (FWHM) of the center fringe is $2\delta = 0.25$ Hz. By measuring the clock transition frequencies at different magnetic field intensities, the clock transition frequency at zero field can be obtained. For this method, the ground-state hyperfine splitting frequencies of $^{111}Cd^{+}$ and $^{113}Cd^{+}$ were measured in early work [23]. Recently, the experimental

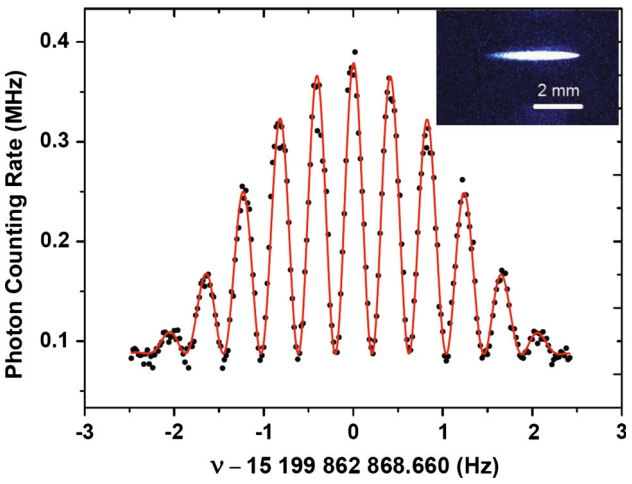

Fig. 3 Typical Ramsey fringe with a corresponding time interval of 2 s between two microwave pulses. The *solid circles* are measured data, and the *curve* is fitted to the Ramsey fringe formula. The *inset* is a photograph of ions captured by the EMCCD with an exposure time of 0.2 s

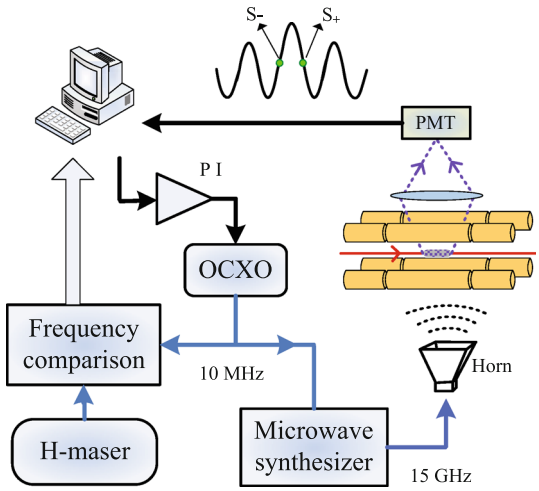

Fig. 4 Schematic illustration of the frequency stability measurement setup. *PMT* photomultiplier tube, *PI* proportional-integral controller, *OCXO* oven-controlled crystal oscillator, *H-maser* hydrogen maser. S_+ and S_- are the signals measured at the microwave frequencies of $\nu_{00} \pm \delta$, where ν_{00} is the midpoint frequency of the center Ramsey fringe, and δ is half of the FWHM

setup was upgraded to provide greater magnetic field stability and measurement sequence control. Accordingly, the clock transition frequency of $^{113}Cd^{+}$ was measured as 15,199,862,855.0125(87) Hz [15], which is limited mainly by the uncertainty of the frequency reference used in the measurement.

Recently, we performed some preliminary measurements on this newly upgraded experimental setup (see Fig. 4) and obtained a preliminary result. The error signal, $error = S_- - S_+$, was obtained by measuring the difference in the ion transition probabilities in two microwave radiation fields with the same power and the same magnitude of δ for the red- and blue-shifted frequencies from the center Ramsey fringe. A feedback voltage from a proportional-integral controller (PI) is used to correct the OCXO's frequency to lock the error signal to zero with a time constant of approximately 100 s. The frequency stability of the experimental setup is measured by comparing the OCXO output with the signal from an active hydrogen maser (H-maser) located in National Institute of Metrology of China(NIM) about 40 km away; the signal is transmitted over an urban fiber with active phase compensation [24].

The modified Allan deviations of the $^{113}Cd^{+}$ frequency standard and the free-running OCXO compared with the H-maser are shown in Fig. 5. The measured frequency stability of the clock is 3×10^{-13} and 2.3×10^{-14} for the average time of 1 s and 4,000 s, respectively. Because the time constant of the controller is 100 s, the Allan deviation of <100 s is mainly determined by the OCXO, which also can be deduced from consistency checks of measurements between the Allan deviations of the $^{113}Cd^{+}$ clock and the

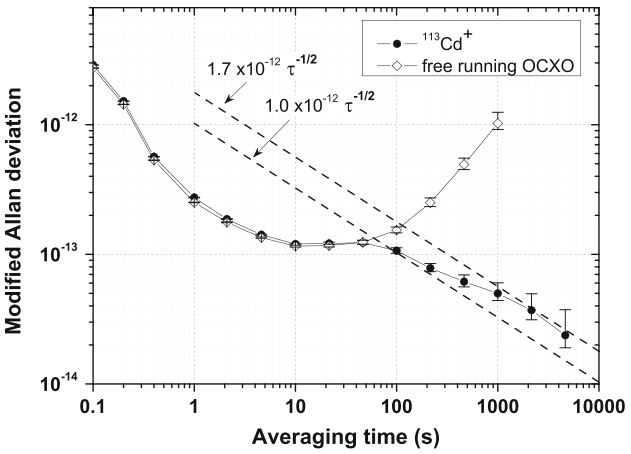

Fig. 5 Modified Allan deviations of the $^{113}Cd^+$ frequency standard and the free-running OCXO. The frequency reference during the measurements is produced by a H-maser

free-running OCXO for averaging times shorter than 100 s. For averaging times longer than 100 s, the Allan deviation varies between $1.0 \times 10^{-12}\tau^{-1/2}$ and $1.7 \times 10^{-12}\tau^{-1/2}$, which is mainly determined by the ions. This measured frequency stability is worse than the calculated limit in Sect. 2 because the small number of trapped ions (approximately 10^4) limits the SNR of the clock signal. In addition, fluctuations in laser power and frequency also introduce additional technical noise. To improve the SNR, a series of upgrades will be carried out for the next round of measurements.

5 Conclusions and outlook

According to our calculation, the microwave frequency standard based on laser-cooled $^{113}Cd^+$ ions has great performance potential with a frequency stability expected to be $2 \times 10^{-14}\tau^{-1/2}$ and a frequency uncertainty of 1.5×10^{-15}. Meanwhile, this clock can be designed to be transportable because of the special energy-level structure of the cadmium ions. With the experimental setup of $^{113}Cd^+$ clock, we have demonstrated a cloud of laser-cooled $^{113}Cd^+$ ions, measured the ground-state hyperfine splitting frequency of $^{113}Cd^+$, and obtained the preliminary data of the frequency stability. Based on the work carried out on the experimental setup, a new compact setup with a multi-layer magnetic shield is under development, and results better than the current measurements are expected. Such a truly transportable microwave frequency standard based on laser-cooled $^{113}Cd^+$ ions will provide a promising solution in applications requiring comparisons between atomic clocks.

Acknowledgments This work was supported by 973 Program (Grant No. 2010CB922901), Tsinghua University Scientific Research Initiative Program (Grant No. 20131080063), and National Natural Science Foundation of China (Grand No. 11304177).

References

1. C.W. Chou, D.B. Hume, J.C.J. Koelemeij, D.J. Wineland, T. Rosenband, Phys. Rev. Lett. **104**(7), 070802 (2010)
2. A.A. Madej, P. Dubé, Z. Zhou, J.E. Bernard, M. Gertsvolf, Phys. Rev. Lett. **109**(20), 203002 (2012)
3. P.T.H. Fisk, Rep. Prog. Phys. **60**(8), 761 (1997)
4. J.D. Prestage, G.J. Dick, L. Maleki, J. Appl. Phys. **66**(3), 1013 (1989)
5. W.M. Itano, J.C. Bergquist, J.J. Bollinger, D.J. Wineland, Phys. Scr. T **59**, 106 (1995)
6. D.J. Berkeland, J.D. Miller, J.C. Bergquist, W.M. Itano, D.J. Wineland, Phys. Rev. Lett. **80**(10), 2089 (1998)
7. S.J. Park, P.J. Manson, M.J. Wouters, R.B. Warrington, M.A. Lawn, P.T. Fisk, in *Proceedings of the 2007 Joint Meeting of EFTF-IEEE IFCS* (2007), pp. 613–616. doi:10.1109/FREQ.2007.4319145
8. E. Burt, W. Diener, R. Tjoelker, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **55**(12), 2586 (2008)
9. J. Prestage, R. Tjoelker, L. Maleki, in *Proceedings of the 1999 Joint Meeting of EFTF-IEEE IFCS* (1999), pp. 121–124. doi:10.1109/FREQ.1999.840723
10. J. Prestage, S. Chung, T. Le, L. Lim, L. Maleki, in *Proceedings of the 2005 IEEE IFCS* (2005), pp. 472–476. doi:10.1109/FREQ.2005.1573976
11. J. Guena, M. Abgrall, D. Rovera, P. Laurent, B. Chupin, M. Lours, G. Santarelli, P. Rosenbusch, M. Tobar, R. Li, K. Gibble, A. Clairon, S. Bize, IEEE Ttrans. Ultrason., Ferroelect., Freq. Cont. **59**(3), 391 (2012)
12. U. Tanaka, H. Imajo, K. Hayasaka, R. Ohmukai, M. Watanabe, S. Urabe, Phys. Rev. A **53**(6), 3982 (1996)
13. B.M. Jelenkovic, S. Chung, J.D. Prestage, L. Maleki, Phys. Rev. A **74**(2), 022505 (2006)
14. U. Tanaka, S. Urabe, M. Watanabe, Appl. Phys. B **78**, 43 (2004)
15. S.G. Wang, J.W. Zhang, Z.B. Wang, K. Miao, L.J. Wang, Opt. Express **21**(10), 12434 (2013)
16. W.M. Itano, L.L. Lewis, D.J. Wineland, Phys. Rev. A **25**(2), 1233 (1982)
17. E.J. Angstmann, V.A. Dzuba, V.V. Flambaum, Phys. Rev. A **74**(2), 023405 (2006)
18. J. Vanier, C. Audoin, The Quantum Physics of Atomic Frequency Standards, vol. 1, (Adam Hilger, Bristol, 1989) pp. 37.
19. S.G. Wang, J.W. Zhang, K. Miao, Z.B. Wang, L.J. Wang, Chin. Phys. Lett. **30**(1), 013703 (2013)
20. J.W. Zhang, Z.B. Wang, S.G. Wang, K. Miao, B. Wang, L.J. Wang, in *Proceedings of the 2012 IEEE IFCS* (2012). doi:10.1109/FCS.2012.6243602
21. J.J. Everdij, A. Huijser, N.F. Verster, Rev. Sci. Instrum. **44**(6), 721 (1973)
22. S.G. Wang, J.W. Zhang, Z.B. Wang, B. Wang, W.X. Liu, Y.Y. Zhao, L.J. Wang, Chin. Opt. Lett. **11**(3), 031401 (2013)
23. J.W. Zhang, Z.B. Wang, S.G. Wang, K. Miao, B. Wang, L.J. Wang, Phys. Rev. A **86**(2), 022523 (2012)
24. B. Wang, C. Gao, W.L. Chen, J. Miao, X. Zhu, Y. Bai, J.W. Zhang, Y.Y. Feng, T.C. Li, L.J. Wang, Sci. Rep. **2**, 556 (2012)