International Conference on Space Optics—ICSO 2006

Noordwijk, Netherlands

27-30 June 2006

Edited by Errico Armandillo, Josiane Costeraste, and Nikos Karafolas



Trapped strontium ion optical clock

G. P. Barwood, P. Gill, H. A. Klein, K. Hosaka, et al.



International Conference on Space Optics — ICSO 2006, edited by Errico Armandillo, Josiane Costeraste, Nikos Karafolas, Proc. of SPIE Vol. 10567, 105672U · © 2006 ESA and CNES CCC code: 0277-786X/17/\$18 · doi: 10.1117/12.2308177

TRAPPED STRONTIUM ION OPTICAL CLOCK

G P Barwood⁽¹⁾, P Gill⁽¹⁾, H A Klein⁽¹⁾, K Hosaka⁽¹⁾, G Huang⁽²⁾, S N Lea⁽¹⁾, H S Margolis ⁽¹⁾, K Szymaniec ⁽¹⁾, and B R Walton⁽¹⁾

⁽¹⁾ National Physical Laboratory, Hampton Road, Teddington, Middlesex TW11 0LW, UK. hugh.klein@npl.co.uk

⁽²⁾ Wuhan Institute of Physics and Mathematics (WIPM), 30 Xiao Hong Shan, Wuhan 430071 China.

Increasingly stringent demands on atomic timekeeping, driven by applications such as global navigation satellite systems (GNSS), communications, and very-long baseline interferometry (VBLI) radio astronomy, have motivated the development of improved time and frequency standards. There are many scientific applications of such devices in space [1]. The experiments that will form the next generation of space-based time and frequency programmes will use optical frequency devices. Such "space science" research will aim to answer the third question addressed by the ESA 2015-25 COSMIC VISION: What are the fundamental laws of the Universe? In Europe, a road map covering the space application of time and frequency metrology is being constructed under the auspices of iMERA [2]. Over the last decades several exciting fundamental space-based new experiments have emerged which will need very good optical clocks and frequency standards [3, 4]. An example of the trend to world-wide collaboration in this field is the "Laser Interferometer Space Antenna" (LISA). Aiming to detect gravitational waves using a space-based interferometer, with arm lengths of a few million km, LISA has very demanding requirements for laser stability. More recent mission proposals (eg [5]) suggesting very stringent multiple tests of both relativistic theory (special and general) and the stability of fundamental constants [6, 7], will require lasers that are both ultra-narrow (linewidth below one Hz) and very stable.

The accuracy and stability of any frequency standard is closely related to its quality factor Q, the ratio of its resonant frequency to the width of the resonant feature [8]. The best caesium primary standards have a Q of about 10^{10} . To achieve an uncertainty of better than one part in 10^{15} for the microwave standard, the centre of the resonant feature therefore has to be found to better than one part in 10^5 . This is achieved using the signal from many atoms. In contrast, a clock based on a weak optical transition between a long-lived metastable state and a lower ground state can have a $Q \ 10^5$ times higher than microwave standards [9]. Particular atoms and ions with appropriate metastable states have optical "clock" transitions with natural linewidths of below 1 Hz,

Proc. '6th Internat. Conf. on Space Optics', ESTEC, Noordwijk, The Netherlands, 27-30 June 2006 (ESA SP-621, June 2006)

corresponding to a Q of around 10^{15} or higher. Frequency standards based on optical clock transitions in a single ion have the advantage that the ion can be confined in a nearly perturbation-free environment [10]. Atom-based clocks may display better short-term stability by using a large ensemble of atoms [9].



Fig. 1. NPL endcap trap (schematic and photo)

The use of optical frequency standards as optical clocks became possible as a result of the development, by Ted Hänsch, Jan Hall and others, of widespan optical frequency combs of accurately-known and equally-spaced frequencies. Based on femtosecond lasers, combs enable the stability of an optical frequency standard to be transferred to the microwave region for comparison with the caesium primary frequency

standard, and made possible the first demonstration of a trapped-ion optical clock at the National Institute of Standards and Technology (NIST) [11].

In the late 1990s the European Space Agency (ESA) recognised the potential for various time and frequency applications in space arising from a technological shift from the microwave to the optical frequencies which was already underway on the ground. Optical frequency standards based on ultra-narrow transitions in trapped ions and atoms already offer better stability than terrestrial microwave fountains and maser devices. The new goal is now to extend this capability to spacebased applications. An ESA study [12] and presentation at the 13th EFTF and IFCS [13] highlighted three application areas: Communications, Navigation and Space Science. Following the 1st ESA International Workshop on Optical Clocks [3], a series of ESA commissioned studies are concentrating on different aspect of optical clocks for space. For example an ESA study on optical synthesizers for space-borne optical frequency metrology led by NPL in Teddington began in February 2006, with partners in Munich. Neuchätel and Düsseldorf.

There are a number of possible opportunities for conducting fundamental physics space-science activities using optical clocks. Optical frequency standards and femtosecond comb measurements now rival and in some cases exceed the capability of microwave frequency devices. Today the best primary caesium standards can realize the SI second with an accuracy of better than 1 part in 10¹⁵. However, it is getting increasingly difficult to improve on this value. Standards based on optical transitions are anticipated to achieve two or more orders of magnitude greater accuracy and stability. It is widely expected that the development of such optical clocks will lead to the redefinition of the SI second in terms of an optical transition in due course. As precision improves, it becomes difficult to compare frequency standards on the ground at different locations because of the gravitational red shift. Comparisons at the part in 10¹⁸ level require knowledge of altitude to 1 cm. To avoid local fluctuations of the geoid, which change local time, the possibility of locating a primary frequency standard in space has been suggested [14].

2. STRONTIUM ION END-CAP TRAP OPTICAL FREQUENCY STANDARD

Single strontium ions are created by electron beam ionization of a weak atomic beam and

confined in an end-cap trap [15] (see fig 1). The separation between the end-caps is 0.56 mm. Cooling of the ion, to a temperature of a few mK, is achieved by driving the strongly allowed $5s^{2}S_{1/2} - 5p^{2}P_{1/2}$ transition at 422 nm (Fig 2). This radiation is provided by a frequency-doubled 844 nm laser. When the ion is in the 5p ${}^{2}P_{1/2}$ state, there is a $\sim 1/13$ probability that it will end up in the long-lived 4d ${}^{2}D_{3/2}$ level and so an additional re-pumper laser, at 1092 nm, is required to maintain efficient cooling. Another laser, at 1033 nm (Fig 3) has recently been developed in order to drive the ion efficiently out of the ${}^{2}D_{5/2}$ level (Fig 2). Use of this laser will speed up the ion interrogation cycle and so should help improve the frequency stability of the standard.

The open geometry of the end-cap trap allows us to cool the ion in three non co-planar directions. Fluorescence from the 422 nm cooling transition is detected using a photomultiplier tube and micromotion can be reduced in three dimensions using rf photon correlation techniques [16, 17]. To keep the ion exactly at the trap centre, DC compensation voltages are applied to the outer endcaps and to compensation electrodes.



Fig 2: Partial term scheme for 88 Sr⁺, showing the cooling lasers at 422 nm and 1092 nm and also the optical clock transition at 674 nm

The optical frequency standard is the 5s ${}^{2}S_{1/2} - 4d$ ${}^{2}D_{5/2}$ quadrupole transition at 674 nm (fig. 2). This transition is interrogated with an extended cavity diode laser, locked to a high finesse ultra-lowexpansion (ULE) cavity. The frequency-stabilized

probe laser is shifted into resonance with the 674 nm clock transition by using a double-passed acousto-optic modulator (AOM), and the clock transition is observed using the quantum jump technique [18]. The Pound-Drever-Hall lock [19] to the cavity currently gives a linewidth of 1.4 Hz linewidth on a timescale of ~3 s. This was inferred from the measured beat frequency linewidth of 2 Hz between two similar laser systems (see Fig 3). On a timescale of ~30 s, the beat frequency linewidth increases to ~6 Hz, implying a single laser linewidth of 4 Hz, with possible cause of broadening due to weak parasitic etalon effects in the path between the laser and the ULE cavity. The beat frequency stability between the two lasers has an optimum value of around 2 parts in 10¹⁵ of the optical frequency at 1 s. On longer timescales, cavity drift rates of typically 0.2 Hz/s are observed.



Fig 3: Beat between 2 independent probe lasers

The trap is mounted inside a mu-metal shield to reduce the effect of the Earth's magnetic field. Three pairs of coils are mounted in orthogonal orientations around the trap, and are used to null the field to a level of 70 nT as measured from the Zeeman splitting of the clock transition.



Fig 4: The 1033 nm extended cavity laser diode

A magnetic field of typically 1.4 µT is then applied in a known direction using one of the three coils. The ⁸⁸Sr⁺ ion is interrogated using a computer-controlled sequence of operations during which the ion is alternately cooled at 422 nm and then probed at 674 nm. During the probe laser pulses, the cooling and repumping lasers are blocked by a combination of acoustooptic modulators and a mechanical shutter, to prevent ac Stark shifts of the 674 nm clock transition. The probe laser pulses are typically 5 ms in duration, giving a Fourier-transformlimited linewidth of 200 Hz. The centre frequency of the 5s ${}^{2}S_{1/2}$ – 4d ${}^{2}D_{5/2}$ Zeeman structure is determined using a four-point servo scheme with a typical cycle time of 15 to 20 s to probe a pair of Zeeman components which are symmetrically placed around line centre. We use a proportional correction scheme similar to that described by Bernard et al. [20], combined with a "feedforward" correction to compensate for the drift of the ULE cavity [21].

Absolute frequency measurements of the clock transition frequency are performed by simultaneously recording the AOM offset frequency and measuring the frequency of the light locked to the ULE cavity with the use of a femtosecond optical frequency comb [22, 23]. Both the repetition rate and the carrier envelope offset frequency of the comb are stabilized to rf synthesizers which are referenced to the 10 MHz output of a hydrogen maser. The offset of the maser output frequency from 10 MHz can be determined by comparison with the NPL caesium fountain [24].

3 ELECTRIC QUADRUPOLE SHIFTS

The quadrupole shift due to the interaction between the electric quadrupole moment of the atomic state with any residual electric field gradient present at the position of the ion can give rise to potentially large systematic frequency uncertainties. The shift arises only in the ²D_{5/2} level, as the quadrupole shift for the ${}^{2}S_{1/2}$ state is zero. The resulting frequency shift can easily be several Hz or more, although it averages to zero for any three mutually perpendicular orientations of the magnetic field [25]. By varying the electric field gradient we were able to make an accurate measurement of the quadrupole moment of the strontium ion ${}^{2}D_{5/2}$ level [21]. The measurements showed that, in atomic units, this quadrupole moment, is 2.6(3), where the standard uncertainty is given in parenthesis. This can be compared with the value of 2.99 calculated in [26]. However, the quadrupole shift also averages to zero if measured for three Zeeman components, with $|m_j| = 1/2$, 3/2 and 5/2 [27]; this technique was used in the optical frequency measurements described in the next section.

4 OPTICAL FREQUENCY MEAUREMENTS

Absolute frequency measurements of the 5s ${}^{2}S_{1/2}$ – 4d ²D_{5/2} transition in ⁸⁸Sr⁺ were carried out over two separate periods in 2004, and also over one period in 2005. The femtosecond optical frequency comb was referenced to the NPL caesium fountain primary frequency standard throughout [28]. On the first six days (Fig. 5(a)) the quadrupole shift was nulled by selecting a particular pair of Zeeman components and carrying out frequency measurements for three mutually orthogonal orientations of the applied dc magnetic field, corresponding to angles β of about 11°, 101° and 90°, where β is the angle between the principal axis of the quadrupole field gradient and the magnetic axis. On the second period of five days (Fig. 5(b)), frequency measurements were made for three different pairs of Zeeman components in order to null the quadrupole shift. For two of these days, the magnetic field direction was such that $\beta \sim 10^\circ$, while for the other three days β was about 90°. Each data point shown in Fig. 4 corresponds to typically 10 000 s of data, and the error bars represent statistical uncertainties only. The unweighted mean frequencies of the two sets of data, before for systematic correcting errors. are 444 779 044 095 485.0 (1.3) Hz and 444 779 044 095 486.2 (1.2) Hz respectively, where the statistical uncertainties are shown in parentheses.

The uncertainty budget for our frequency measurement has been discussed in more detail elsewhere [28]. The largest contributions to the frequency measurement uncertainty are briefly discussed here. Although the quadrupole shift of the transition frequency is nominally nulled for both sets of data, the cancellation of the shift in the first method requires the three magnetic field orientations used to be exactly orthogonal. With an estimated 10° uncertainty in the magnetic field directions chosen, the residual uncertainty due to the quadrupole shift is around 10% of the mean magnitude of the shift. In the second method, the cancellation of the quadrupole shift relies only on the angle between the magnetic field axis and the quadrupole axis being stable over the course of a day's measurements, and so the residual uncertainty is negligible in this case.

ULE cavity frequency drift can lead to servo errors in the lock to the centre of the Zeeman structure. As a result, we induced deliberate changes in both the magnitude and the direction of the cavity drift rate by adjusting the temperature control system, leading to larger than normal imbalances between the quantum jump rates on each side of the line. The relationship between measured frequency and quantum jump imbalance determined in this way was consistent with the relationship predicted on the basis of the observed linewidth and servo parameters.



Fig. 5. Frequency of the 5s ${}^{2}S_{1/2} - 4d {}^{2}D_{5/2}$ transition in ${}^{88}Sr^{+}$, determined from (a) the average of measurements made in three nominally orthogonal magnetic field directions and (b) the average of measurements carried out by using three different pairs of Zeeman components corresponding to transitions for which $|m_{j}'| = 1/2$, 3/2 and 5/2. The solid lines show the average measured value using each technique. Corrections have been applied for systematic shifts [28].

During the 2004 measurements, the largest source of systematic uncertainty was the ac Stark shift arising from the 422 nm cooling radiation. Although this was nominally blocked during the probe laser interrogation periods by a combination of an AOM and a mechanical shutter, in practice it was not perfectly extinguished due to a small amount of light scattered past the small shutter blade. Since then, we have significantly improved the extinction of the cooling laser radiation during the probe laser periods, reducing the 422 nm ac Stark shift to a negligible level. Further frequency measurements carried out in 2005 after this change confirm our previous frequency result, although with a three times larger error bar because significantly less data was taken and the caesium fountain was only run during the last part of the data-taking period.

The uncertainty in the maser reference frequency mainly arises from the uncertainty in the caesium fountain frequency (1 part in 10^{15} statistical over the timescale of these measurements and 1 part in 10^{15} systematic) [24] but also has a contribution from the uncertainty in correcting for the frequency shifts in the cables used to transfer the maser signal to the femtosecond comb laboratory.

Correcting for the systematic shifts gives frequency values of 444 779 044 095 484.3 (1.9) Hz and 444 779 044 095 484.8 (1.6) Hz using the respective methods of nulling the electric quadrupole shift The unweighted mean of the two values gives a final value for the 674 nm electric quadrupole clock transition frequency in ⁸⁸Sr⁺ of 444 779 044 095 484.6 (1.5) Hz [28]. For the calculation of the final uncertainty, only the statistical error is reduced, because the systematic uncertainties are mostly common to the two measurements. This result is in good agreement with other measurements elsewhere of the strontium ion clock transition frequency [29], and when both statistical and systematic errors are considered, its fractional uncertainty of 3.4 x 10^{-15} is within a factor of three of that of the NPL primary caesium standard.

5. FUTURE ⁸⁸Sr⁺ TRAPPED ION CLOCK PERFORMANCE AND STAND-ALONE OPERATION

The recent improvements to the probe laser linewidth and ULE cavity drift rate should lead to significant reductions in servo errors. A second endcap trap has been developed, which will allow a more detailed experimental investigation of systematic errors in the near future by means of two-trap comparisons. Recent results of such a comparison between Yb⁺ ion trap standards at PTB [30] has demonstrated reproducibility at better than a part in 10^{15} . With these improvements, we anticipate to be able to achieve an optical frequency measurement of the strontium clock transition that is limited by the accuracy of the caesium fountain. As well as being of interest for a possible future redefinition of the second, measurements of this and other optical frequency standards over timescales of a few years will provide increasingly sensitive laboratory tests of the time invariance of fundamental constants [31, 32].

Operation of the strontium-ion optical frequency standard in optical clock mode has potential as a

reference clock for use eg in ESA ground station T&F facilities. For the strontium ion clock, there is considerable scope for miniaturisation of the trap and all the required optical frequencies can be generated using laser diodes. Currently, the 1092 nm radiation is provided by a DFB laser, and tunes reliably to the same frequency from day to day at the same current and temperature, which can be read and controlled by a PC. The 422 nm cooling radiation, 1033 nm and 674 nm clock light, although currently provided by extended cavity lasers, could also be provided by DFB laser sources. Integration of a compact trap with DFB laser light delivered by fibre to the trap, together with an increased level of opto-electronic and computer control, could provide the basis for a remotely operated frequency standard. Such a single ion trap standard together with a suitable eg fibre frequency comb could form a future miniature optical clock with both high accuracy and stability.

Valuable discussions with Lute Maleki and colleagues at JPL together with ESA staff and study partners are acknowledged. This work was partially funded by ESA and the UK NMSD Quantum Metrology and Length programmes.

1. Maleki L ed. *Proceedings of workshop on the scientific application of clocks in Space* JPL Publication 97-15 (1997).

2. http://www.euromet.org/projects/imera/

- 3. Proceedings of the 1st ESA International Workshop on Optical Clocks (8-10 June 2005). 4.Karshenboim SG and Peik E eds., Astrophysics, Clocks and Fundamental Constants Lecture Notes in Physics **648** 297-307 Springer-Verlag Berlin Heidelberg (2005).
- 5. Schiller S et al., *Proc. 39th ESLAB Symposium* 19-21 F Favata, A Gimenez. eds. (April 2005).
 6. Lea SN, Flowers JF and Gallop JC "Laboratory limits to time-variation of the fundamental constants" to be submitted to *Rep*.

Prog. Phys.
7. Flowers JL, Klein HA & Margolis HS, *Contemporary Physics* 45, 123-145 (2004).
8. Riehle F, *Frequency Standards* WILEY-VCH

(2004) and references therein. 9. Metrologia **42** No 3 Special Issue *50 years of atomic time-keeping 1955 to 2005* (June 2005),

with particular reference to Gill P, *Metrologia* **42** S125-S137 (2005).

10. Lea SN & Klein HA, "Trapped Ion Optical Clocks", *Yearbook of Science and Technology*, McGraw-Hill (2006).

11. Diddams SA et al. Science 293, 825 (2001).12. Klein HA and Knight DJE, "Ultra stable optical clocks, frequency sources and standards

for space applications", Final Report to ESA under contract 13290/98/NL/MV (1999).

13. Hodge CC et al. *Ultra-stable optical*

frequencies for space, Proc. 13th EFTF and IFCS, Besancon (April 1999).

14. Kleppner D "Time Too Good to Be True",

Physics Today **59**, 10 (2006)

15. Schrama CA, Peik E, Smith WW and. Walther H, *Opt. Commun.* **101**, 32 (1993).

16. Siemers I et al., *Europhys. Lett.* 18, 139 (1992).

17. Berkeland DJ et al., J. Appl. Phys. 83, 5025 (1998).

18. Dehmelt H, *IEEE Trans. Instrum. Meas.* **31**, 83 (1982).

19. Drever RWP et al., Appl. Phys. B**31**, 97 (1983).

20. Bernard JB et al., *Opt. Commun.* **150**, 170 (1998).

21. Barwood GP et al, PRL 93 133001 (2004)

22. Margolis HS et al., *Phys. Rev.* A **67**, 032501 (2003).

23 Lea SN et al., *Metrologia* **40**, 84 (2003).

24. Szymaniec K et al., Metrologia 42, 49 (2005).

25. Itano WM, "External field shifts of the ¹⁹⁹Hg⁺ frequency standard", *J. Res. Natl. Inst. Stand. Technol.* **105**, 829 (2000).

26. Oskay W, Itano WM and Bergquist JC, *Phys Rev Lett*, **94**, 163001 (2005)

27. Dube P et al., Phys. Rev. Lett. 95, 033001 (2005)

28. Margolis HS et al, Science **306** 1355-1358 (2004).

29. Madej AA et al., *Phys. Rev.* A**70**, 012507 (2004).

30. Schneider T, Peik E and Tamm Chr., *Phys. Rev. Lett.*, **94**, 230801 (2005).

31. Fischer M et al., *Phys. Rev. Lett.* **92**, 230802 (2004).

32. Peik E et al., *Phys. Rev. Lett.* **93**, 170801 (2004).

© Crown Copyright 2006