Absolute frequency measurement of the 674-nm $^{88}Sr^+$ clock transition using a femtosecond optical frequency comb

H. S. Margolis,* G. Huang, G. P. Barwood, S. N. Lea, H. A. Klein, W. R. C. Rowley, and P. Gill
National Physical Laboratory (NPL), Teddington, Middlesex TW11 0LW, United Kingdom

R. S. Windeler
OFS, 700 Mountain Avenue, Murray Hill, New Jersey 070974
(Received 26 July 2002; published 10 March 2003)

The frequency of the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ electric quadrupole transition at 674 nm in a single, trapped, laser-cooled $^{88}Sr^+$ ion has been measured with respect to the Système International (SI) second using a femtosecond laser optical frequency comb. The measured frequency of 444 779 044 095.52 kHz, with an estimated standard uncertainty of 0.10 kHz, is more accurate than, and in agreement with, the value previously measured using a conventional frequency chain.

DOI: 10.1103/PhysRevA.67.032501 PACS number(s): 32.30.Jc, 32.80.Pj, 06.20.Fn, 06.30.Ft

I. INTRODUCTION

Highly stable and reproducible atomic frequency standards play a key role in the realization of the basic Système International (SI) units [1], tests of physical theories, and the measurement of fundamental physical constants [2,3]. It has long been recognized that lasers stabilized to narrow optical transitions in atomic systems offer the prospect of improved stability and accuracy compared to microwave standards due to the higher Q factors of the transitions involved. A forbidden transition in a single ion stored in a rf quadrupole trap is a promising reference for an optical frequency standard. By laser cooling, the ion can be confined in the Lamb-Dicke regime [4], eliminating the first-order Doppler effect. Careful control of the micromotion of the ion allows the second-order Doppler shift to be greatly reduced and the ion to be confined to the trap center where electric field perturbations are minimized [5]. Operating the trap under ultrahigh vacuum conditions ensures that collisional effects are negligible. Finally, the reference transition can be observed with high efficiency and signal-to-noise ratio using the quantum jump technique [6].

Over recent years there has been considerable progress toward the use of such references as optical frequency standards [7], with Fourier-transform limited linewidths as narrow as 6.7 Hz having been reported [8]. The measurement of optical frequencies has traditionally been a complex task using frequency chains designed specifically to link a particular optical frequency to the cesium primary standard at 9.2 GHz [9,10]. However, such measurements have been greatly simplified by the development of frequency comb generators based on Kerr-lens mode-locked femtosecond lasers [11], and it is now possible for the link between the optical and the microwave to be made in a single step [12]. Here we report the application of these methods to the measurement of the frequency of the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ electric quadrupole transition in $^{88}Sr^+$ at 445 THz (674 nm). A preliminary value for this transition frequency with a standard uncertainty of 300 Hz was reported previously [13]; here we present further measurements which lead to an improved result with a standard uncertainty of 100 Hz.

*Email address: Helen.Margolis@npl.co.uk

II. $^{88}Sr^+$ OPTICAL FREQUENCY STANDARD

A partial term diagram for $^{88}Sr^+$ is shown in Fig. 1. The $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ electric quadrupole transition has a natural linewidth of approximately 0.4 Hz, and was the first transition in a single trapped ion to be adopted by the Comité International des Poids et Mesures (CIPM) as a recommended radiation for the practical realization of the meter [1].

The National Physical Laboratory (NPL) strontium ion standard is based on a single trapped $^{88}Sr^+$ ion confined in a miniature rf Paul trap which has a radius of approximately 0.5 mm [14]. An 844-nm diode laser is frequency doubled to 422 nm to laser cool the ion into the Lamb-Dicke regime in one dimension, and a distributed Bragg reflector (DBR) laser at 1092 nm is used to prevent optical pumping into the $4d\,^2D_{3/2}$ state. The $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition at 674 nm is

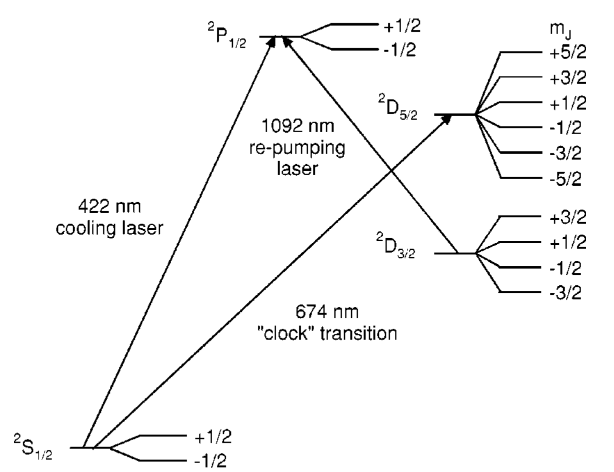

FIG. 1. Partial term diagram of $^{88}Sr^+$, showing the transitions used to cool and probe the trapped ion.

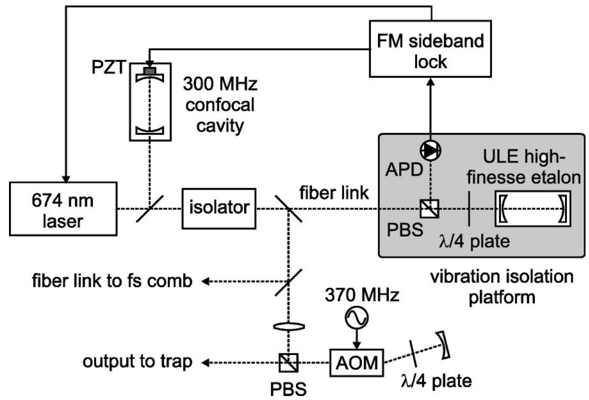

FIG. 2. Schematic of the probe laser system used to interrogate the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition in $^{88}Sr^+$ at 674 nm. (APD, avalanche photodiode; PBS, polarizing beam splitter; PZT, piezoelectric transducer; FM, frequency modulation).

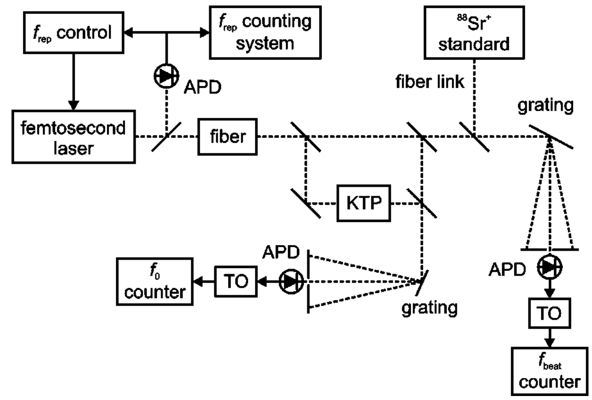

FIG. 3. The femtosecond comb setup used to measure the absolute frequency of the $^{88}Sr^+$ standard. (APD, avalanche photodiode; TO, tracking oscillator; KTP, potassium titanyl phosphate crystal.)

probed using an optically narrowed AlGaInP diode laser which is locked to a highly stable ultra-low expansion (ULE) high finesse cavity [15]. The linewidth of this laser source is about 200 Hz [16]. To bridge the frequency interval of about 740 MHz between the strontium transition frequency and the nearest mode of the cavity, the laser output is shifted by a double-passed acousto-optic modulator (AOM), as shown in Fig. 2.

In a dc magnetic field, the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition is split into ten Zeeman components. To reduce the effect of ambient laboratory magnetic fields, the ion trap is housed inside a μ-metal shield. Three pairs of coils mounted inside this shield are used to further reduce the magnetic field to about 4 μT at the trap center, which is sufficient to resolve the Zeeman components.

The strontium ion is interrogated using a computer-controlled sequence of operations in which the ion is alternately cooled and then probed [17]. The number of quantum jumps to the $4d\,^2D_{5/2}$ state at a given probe laser frequency is counted for a particular number of interrogation cycles (typically 40). After each set of interrogation cycles the laser frequency is stepped. The probe laser frequency is locked to the center frequency of the Zeeman structure by a four-point locking scheme which uses the two $\Delta m=0$ Zeeman components symmetrically placed around the line center [16]. Typically, for the measurements presented here, the transition width was power broadened to 1–2 kHz, and the synthesizer was stepped by 0.7–1.0 kHz in order to sample the resonance curve on either side of a particular Zeeman component. The step correction frequency for the center frequency was typically 50 Hz.

III. FEMTOSECOND OPTICAL FREQUENCY COMB

The frequency measurements were carried out using a maser-referenced octave-span optical frequency comb [11,12]. This comb was generated by a Kerr-lens mode-locked Ti:sapphire laser with external spectral broadening in a microstructure fiber (see Fig. 3). The femtosecond Ti:sapphire laser has a linear cavity design with intracavity prisms for control of the group velocity dispersion [18], and is pumped with 4.5 W of single-frequency radiation at 532 nm. The pulse repetition rate, determined by the cavity length, is about 87 MHz, with fine control provided by translation of a piezo-mounted cavity fold mirror. The pulse duration is typically 10–15 fs and the average ouput power of the laser is about 600 mW. In the frequency domain, the output of the laser is a comb of modes, spaced by the repetition rate of the laser. This comb is centered at around 810 nm, and covers a spectral range of around 30–35 nm full width at half maximum. To broaden the frequency comb to span a full optical octave, about 30 mW of the laser output is coupled into a short length of microstructure fiber [19]. For the measurements reported here fibers of lengths between 11 cm and 25 cm were used.

The frequency of the mth mode of the comb is given by

$$f_m = m f_{\mathrm{rep}} + f_0, \tag{1}$$

where f_{rep} is the repetition rate of the laser and f_0 is the frequency offset of the whole comb with respect to the frequency origin. The frequency f_{rep} is determined by splitting off a small portion of the laser output and detecting the intermode beat spectrum using an avalanche photodiode. The intermode beat signal at the tenth harmonic of f_{rep} is frequency down-converted by mixing with the output of a Marconi 2024 rf synthesizer at frequency f_{s} and filtered to generate a signal at 9.8 MHz. This signal is used for two purposes. First, by mixing it with a reference 9.8-MHz signal from an IFR 2023A rf synthesizer in an analog phase comparator, it is used to generate an error signal which is fed back to the piezo-mounted fold mirror in the laser to stabilize the repetition rate. Secondly, it is mixed with a 10-MHz reference signal to produce a 200-kHz signal which is used to phase-lock a 200-MHz voltage-controlled oscillator (VCO) by means of a frequency divider with a division ratio of 1000. The output of this VCO is counted using a HP53132A frequency counter. From this counted frequency f_{c} and the synthesizer frequency f_{s}, the repetition rate of the laser can be determined:

$$f_{\mathrm{rep}} = \frac{1}{10}\left[f_{\mathrm{s}} - 10\ \ \mathrm{MHz} + \frac{f_{\mathrm{c}}}{1000} \right]. \tag{2}$$

The offset frequency f_0 is determined using the self-referencing technique [20,21]. Comb modes in a bandwidth of a few nanometers around 1060 nm are frequency doubled in a single pass through a KTP crystal, and recombined with modes in an equivalent bandwidth around 530 nm. The resultant beat frequency f_0 is detected using an avalanche photodiode after spectral filtering by a diffraction grating. The signal-to-noise ratio of this beat is insufficient for direct counting (around 20–25 dB in 100-kHz bandwidth), so it is filtered and amplified using an analog tracking oscillator to obtain a countable signal. The offset frequency can be controlled by adjusting the tilt of the end mirror in the laser cavity using a piezoelectric stack; however, it is not servoed but simply adjusted to set f_0 within the range of the tracking oscillator.

About 100 μW of the ULE-stabilized 674 nm radiation from the ion trap laboratory was transferred to the frequency comb laboratory using a single-mode fiber. The beat frequency f_{beat} between this light and the comb was detected by an avalanche photodiode after spectral filtering using a grating in a similar manner to the offset frequency f_0. The signal-to-noise ratio in this beat is about 20 dB in 100-kHz bandwidth, so again a tracking oscillator is locked to the beat to obtain a countable signal. To bring the beat frequency f_{beat} into the range of the tracking oscillator, the repetition rate of the laser is adjusted by altering the synthesizer frequency f_s.

The frequency f_s is recorded and the frequencies f_c, f_0, and f_{beat} are counted by HP53132A frequency counters which are synchronously gated and read by PC-based software via a GPIB interface. The rf synthesizers and frequency counters are all referenced to the 10-MHz output of a hydrogen maser which forms part of the clock ensemble used to generate the time scale UTC (NPL), which is the U.K.'s realization of Coordinated Universal Time. During the period of the measurements reported here, the maser reference provided traceability to the SI second to 2.5 parts in 10^{14}. The 10-MHz signal from the maser is transmitted to the femtosecond laser laboratory by 250 m of RG213 coaxial cable, which is mainly underground. The round trip delay time measured using the 1 pulse per second output of the maser showed a maximum relative diurnal sinusoidal variation of about 0.4 ns peak-to-peak amplitude, superimposed on a longer-term drift. Both these effects were strongly correlated to external temperature and were consequently largest for the measurements taken during the summer, when the temperature changes were largest. The maximum rate of change of the delay implies a frequency pulling of 8×10^{-15}, and for most of the data is significantly less than this, because the temperature changes were smaller or because data were taken close to a turning point. This frequency pulling effect is therefore negligible compared to the other sources of uncertainty in the present measurement.

The frequency f_{Sr} of the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition in $^{88}\mathrm{Sr}^+$ is given by

$$f_{\mathrm{Sr}}=mf_{\mathrm{rep}}\pm f_0\pm f_{\mathrm{beat}}+f_{\mathrm{offset}} \tag{3}$$

where f_{offset} is the frequency offset between the ULE cavity frequency and the trapped ion transition frequency. It is straightforward to determine the mode number m and the signs of the beats in this case, because the transition frequency is already known with an uncertainty of far better than 1 MHz [10,22]. The synthesizer used to drive the AOM is referenced to an off-air frequency standard (a tracking receiver which is locked to the 198-kHz Droitwich radio transmitter signal). The accuracy of this standard is better than 1 part in 10^9, and the frequency $f_{\mathrm{offset}}/2$, which is periodically stepped, is recorded by the ion trap software. The transition frequency is determined by summing the mean value of the ULE cavity frequency as measured using the femtosecond comb and the mean value of f_{offset} recorded over the same time interval.

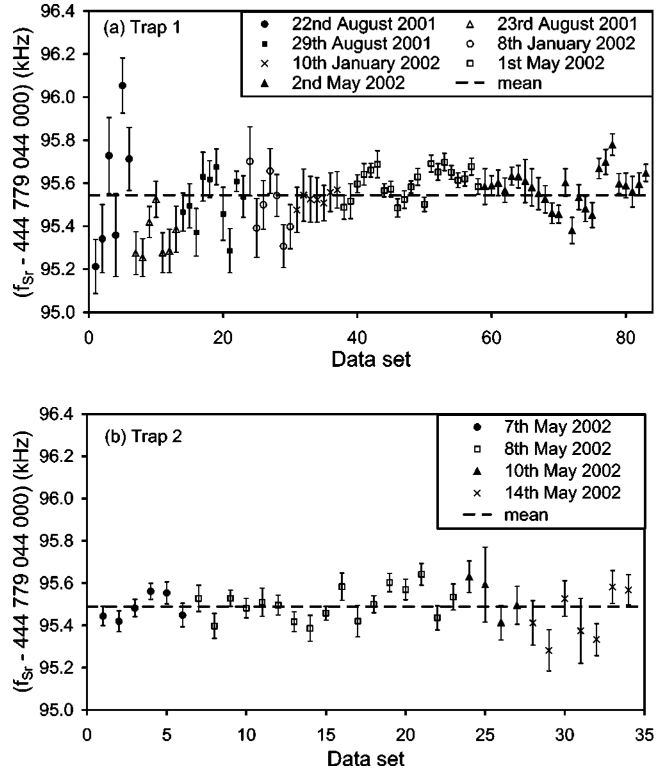

FIG. 4. The frequency of the $^{88}\mathrm{Sr}^+$ $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition, measured in two ion traps of similar design. The dashed lines represent the average measured value for each trap.

IV. RESULTS OF FREQUENCY MEASUREMENTS

Frequency measurements of the $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ transition in $^{88}\mathrm{Sr}^+$ were made on 11 separate days, and included measurements on two ion traps of similar design. Each data set shown in Fig. 4 is the mean of typically around 300 1-s measurements, and the error bar shown is the standard error of the mean of the data set. For the early measurements, the standard deviation of a set of 1-s measurements of the ULE cavity-stabilized laser was typically about 1 kHz, while for the later measurements this was reduced to around 400 Hz. The Allan deviation of the measured frequency at 1 s was therefore $(1–2)\times10^{-12}$. Between successive data sets the sign of one or both of the beats f_0 or f_{beat} was changed, to check that the tracking oscillators were not introducing any

TABLE I. Systematic frequency shifts in the frequency measurements and their estimated uncertainties (in Hz).

Source	Trap 1 Correction	Trap 1 Uncertainty	Trap 2 Correction	Trap 2 Uncertainty
Reproducibility		152		104
422 nm ac Stark shift	−48	60	0	0
1092 nm ac Stark shift	0	<3	0	<3
Servo errors	−3	3	−12	12
Other frequency shifts	0	<1	0	<1
Maser frequency	0	11	0	11
Total uncertainty for each trap		164		105

significant offset. The synthesizer frequency f_s was also adjusted between most measurement periods, to compensate for a long-term mechanical drift of the femtosecond laser cavity length.

Each data point shown in Fig. 4 has been corrected to allow for small offsets of the locked frequency from the actual center of the Zeeman pattern, which result from drifts of the ULE cavity frequency [23]. The drift rate was up to $\approx 1\ \mathrm{Hz\,s^{-1}}$ for the periods when frequency measurements were made. The corrections were estimated from the expected relationship between the offset and the mean quantum jump imbalance; the mean correction applied to the data for trap 1 was -3 Hz, and for the trap 2 data it was -12 Hz. The data points for trap 1 have also been corrected to allow for an ac Stark shift arising from the 422 nm cooling light. This must be switched off during the periods when the ion is probed using the 674-nm laser to avoid broadening and shifting of the reference transitions. In the earlier measurements, this switching was performed using only an AOM, leading to a small amount of 422-nm leakage to the trap when this beam was nominally switched off. In the later measurements, a combination of a mechanical shutter and the AOM were used. By deliberately increasing the amount of leakage through the AOM during some of the measurements on trap 2, the 422-nm ac Stark shift was measured. Applying an appropriately scaled correction to the results for trap 1 led to a mean correction of $-48(60)$ Hz. For trap 2, only data sets for which the mechanical shutter was used are included in Fig. 4.

The reduced scatter observed in the later measurements is due to improvements in the femtosecond comb performance, resulting in an increase in the signal-to-noise ratio in the beat signals. However, the points are not randomly distributed; for example, on the 1st and 2nd of May the measured frequency of trap 1 showed slow variations over a total range of about 400 Hz. There is also a slight asymmetry (biased to lower frequency) in the distribution of the frequency values about the mean for either trap, which is suspected to be the result of transverse second-order Doppler shifts. Stray dc electric fields can cause the ion to be displaced from the trap center, leading to both a second-order Doppler effect from residual motion of the ion and a smaller dc Stark shift [24]. The size of these effects can be estimated from the intensity of the rf and secular motion sidebands [5]. For the observed modulation index of $\beta<1$ in the case of micromotion along the direction of the laser propagation, we calculate a longitudinal second-order Doppler shift of less than 0.1 Hz, comparable with previous estimates [10]. However, there is currently no diagnostic on the NPL strontium traps for monitoring micromotion transverse to the laser propagation direction, and so larger transverse second-order Doppler shifts could easily be present. These shifts are hard to estimate, but will always shift the reference transition to lower frequency, and are therefore a likely explanation for the slight asymmetry observed in the frequency distribution.

Further contributions to the observed frequency variations could be the electric quadrupole shift of the $4d\,^2D_{5/2}$ level and servo errors due to slowly varying magnetic fields. Another frequency shift that arises is due to the presence of the 1092-nm laser, which in the earlier measurements was not switched off during the probe periods. This can cause a small ac Stark shift which for our system is estimated to be no more than 3 Hz. Other effects, for example due to the blackbody ac Stark shift and background gas collisions, are estimated to cause frequency shifts of less than 1 Hz.

Since successive results, particularly in trap 1, appear correlated, it is not appropriate to use normal statistics and assume that the standard uncertainty scales as $N^{-1/2}$, for N data sets. As already discussed, the most likely causes for frequency shifts in our traps are servo errors caused by magnetic fields or varying transverse second-order Doppler shifts. A straightforward statistical approach to dealing with these effects is to assume that for each trap they give rise to a rectangular distribution of frequency values [25]. In this case the standard uncertainty is given by the semirange of the frequency spread divided by $\sqrt{3}$. For trap 1, the data points from the first day are ignored in determining the frequency spread, because the larger spread observed on this day was due mainly to poor signal-to-noise ratio in the beat signals for the earliest measurements. The other sources of uncertainty are summarized in Table I. The frequency values obtained for the 674-nm reference transition in the two traps are $f_{\mathrm{Sr}}=444\,779\,044\,095.54(16)$ kHz for trap 1 and $f_{\mathrm{Sr}}=444\,779\,044\,095.49(11)$ kHz for trap 2. To obtain the final value of the measured frequency we take the unweighted mean of these two values, which gives $f_{\mathrm{Sr}}=444\,779\,044\,095.52(10)$ kHz. The difference between the measured frequencies for the two traps is 60(190) Hz, consistent with previous reproducibility measurements on three similar traps [15].

V. CONCLUSION

The $5s\,^2S_{1/2}$–$4d\,^2D_{5/2}$ electric quadrupole transition in $^{88}\mathrm{Sr}^+$ is determined to be 444 779 044 095.52(10) kHz. This value is a factor of 2 more accurate than, and in good agreement with, the best previous measurement of this frequency, which was performed using a conventional frequency chain [10]. The good agreement between these two measurements, carried out using ion traps in different laboratories and using different methods, provides evidence for both the accuracy of the femtosecond laser comb approach to optical frequency measurement and the reproducibility of ion trap standards at the 5 parts in 10^{13} level. The current reproducibility of our measurements is believed to be limited by transverse second-order Doppler shifts of the strontium ion transition. Further reduction in the uncertainty will require the use of techniques for minimizing the micromotion of the ion in three dimensions, and with additional improvements it is expected that it should be possible to reduce all trap systematics to below 1 part in 10^{15}.

ACKNOWLEDGMENT

We would like to acknowledge advice and assistance from D. Henderson and P. B. Whibberley, and technical assistance from N. J. Moore. This work was supported by the U.K. National Measurement System Length Program under Contract No. LE99/A01. It was also part of the U.K.-Japan ''Cold Trapped Ions'' International Collaborative Research Project supported by the Japan Science and Technology Corporation.

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