Cs-Based Frequency Measurement of a Single, Trapped Ion Transition in the Visible Region of the Spectrum

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(Received 7 December 1998)

An optical frequency chain referenced to a Cs atomic clock has been used to measure directly the frequency of the electric quadrupole allowed $5s\,^2S_{1/2} - 4d\,^2D_{5/2}$ transition at 445 THz in a single, trapped, and laser cooled $^{88}Sr^+$ ion. A transition frequency, $f_{S\text{-}D} = 444\,779\,044\,095.4$ kHz with an estimated standard uncertainty of 0.2 kHz has been determined. Intrinsic offsets of the probed ion transition in the current experiment are calculated to be at the 10^{-15} level. [S0031-9007(99)08942-5]

PACS numbers: 32.30.Jc, 06.20.–f, 06.30.Ft, 32.80.Pj

Over the years, atomic frequency standards have been a key element in advancing our knowledge in the fields of precision physical measurement, primary realizations of base units in the SI, and tests of theory at the highest level of accuracy [1]. In the optical region of the spectrum, precision stabilization of highly coherent laser sources to high-Q transitions in atoms and molecules together with the development of methods to accurately "count" such extremely high frequencies (10^{14}–10^{15} Hz) have opened the opportunity for accurate measurement of a fundamental nature. In the mid-1970s a new type of atomic frequency standard was proposed by Dehmelt [2] based on the probing of a narrow dipole forbidden transition in a single ion held in a small rf quadrupole trap and brought to near rest [3]. Single ions can be held almost indefinitely in such small electrodynamic traps and isolated from collisions. Through laser cooling, the ion's velocity can be reduced so that the amplitude of its oscillation inside the trap is less than the wavelength of the probe radiation. Under such conditions, the first order Doppler shift disappears (Lamb-Dicke regime [4]), the second order, relativistic Doppler effect is very small, and the ion is restricted to the center of the trap where the confining electric field vanishes and external field gradients can be reduced to low levels. In addition, for a suitable ion, the resulting narrow spectral line can be measured with a high signal-to-noise (S/N) ratio through the quantum jump technique which permits the detection of single transitions with almost 100% efficiency [2].

The accurate measurement of an optical frequency of several hundred terahertz is accomplished by means of a frequency chain connecting the unknown optical frequency to known transfer standards or directly to the SI realization of the second, a cesium atomic clock. To date, most optical frequency measurements have employed transfer standards in the optical or infrared region [5–7]. At visible frequencies, a Cs-based optical frequency chain measurement has been reported by a group at the Physikalisch-Technische Bundesanstalt (PTB) [8] who have measured the frequency of a many-atom calcium standard at 657 nm with a fractional uncertainty of $<10^{-12}$. In a previous experiment at the National Research Council of Canada (NRC) [9], the transition frequency of a single trapped barium ion at 12.5 μm was measured with a Cs-based frequency chain with an uncertainty of 4×10^{-11}, limited by the stability of the probe laser. We report here Cs-based frequency chain measurements of an optical transition at 674 nm in a single, trapped $^{88}Sr^+$ ion with a fractional uncertainty of $<10^{-12}$. These results are, to our knowledge, the first Cs-based chain measurements and the most accurate of any kind to date of a visible transition frequency in a single, trapped ion or atom.

The electric quadrupole allowed $5s\,^2S_{1/2} - 4d\,^2D_{5/2}$ transition at 445 THz (674 nm) in the single trapped $^{88}Sr^+$ ion has a natural linewidth of approximately 0.4 Hz and was recently selected by the Comité International des Poids et Mesures (CIPM) as a recommended optical frequency for the realization of the meter [10]. Previously, the best value for the frequency of this transition was $444\,779\,044\,093 \pm 20$ kHz [11].

In our experiment, a frequency-doubled diode laser at 422 nm is used for laser cooling and fluorescence detection on the $5s\,^2S_{1/2} - 5p\,^2P_{1/2}$ resonance transition [12]. An auxiliary diode-pumped Nd-fiber laser at 1092 nm is used to pump the ion out of the $4d\,^2D_{3/2}$ state since the $5p\,^2P_{1/2}$ state has a 1:13 branching ratio to this level. With a dc bias magnetic field, the $5s\,^2S_{1/2} - 4d\,^2D_{5/2}$ "clock" transition at 674 nm (445 THz) is split into 10 Zeeman components [13] and is probed by a portion of the output of an ultrastable diode probe laser [14] which is shifted by a double-passed, acousto-optic modulator (AOM) to allow scanning of the S-D spectrum. By observing the number of quantum jumps in the 422-nm fluorescence due to shelving of the single ion in the upper, $4d\,^2D_{5/2}$ level, an efficient detection of each S-D transition is obtained. The single Sr^+ ion is held in an rf Paul trap [14,15]. Briefly, the ion is confined within a spherical rf quadrupole trap of endcap to center distance 0.5 mm with an applied 12-MHz rf trapping potential amplitude of 260 V. A single-layer magnetic shield (CO-NETIC AA) reduces the effects of slowly varying laboratory dc magnetic fields and ac broadening on the positions and widths

 0031-9007/99/82(16)/3228(4)$15.00 © 1999 The American Physical Society

of the Zeeman components. A small magnetic field of approximately 14 μT is applied in the region of the trap by pairs of coils driven by a stable, low-noise current source resulting in a separation of the two inner Zeeman components of approximately 156 kHz.

Spectral widths of 0.5–1 kHz were observed, reflecting the linewidth of the probe laser system. The frequency of the probe laser is referenced to the clock transition frequency by locking the AOM-shifted portion of its output to the two inner Zeeman components, $(m(j'), m(j'')) = (\pm 1/2, \pm 1/2)$ using the technique described in [16] and recording the frequency applied to the AOM. Four discrete frequencies, one on each side of each of the two Zeeman components, are scanned during each 20-s lock cycle. The uncertainty in the laser-referenced center frequency of the S-D manifold, after accounting for the observed laser drift of up to 5 Hz/s, is calculated to be less than 100 Hz.

The frequency chain shown in Fig. 1 has been developed at NRC in order to measure the frequency of the Sr^+ S-D transition with respect to a Cs standard. The bottom of the chain is formed by the NRC phase-locked infrared chain [17] which links a 5-MHz reference signal (R) through two microwave oscillators (X and V) to four CO_2 lasers (A, B, C, and D). The 5-MHz reference is provided by a hydrogen maser which is continuously compared to two primary and one commercial Cs standard and had a 1-s Allan deviation, σ_y of better than 3×10^{-13} and a frequency offset of $(-9 \pm 1) \times 10^{-14}$ for all measurements relative to proper time. This frequency offset was taken into account in the calculation of the S-D transition frequency. Tungsten-nickel, metal-insulator-metal (MIM), point-contact diodes are used to produce harmonics of the laser and microwave radiation and produce the heterodyne beats shown in Fig. 1. The infrared chain is locked by first phase locking laser D to laser A so that the beat $5D - 4A$ is exactly 1125 MHz. Then laser C is phase locked to lasers A and D through the beat shown, and similarly for laser B and microwave oscillators V and X. The infrared chain is closed by mixing down the X oscillator to 35 MHz, phase locking a 5-MHz voltage controlled oscillator (VCO) (not shown) to this beat, and controlling the frequency of laser A so that the VCO is phase locked to the 5-MHz reference. When all the servo loops are closed, each of the CO_2 lasers and microwave oscillators has the same long-term stability as the reference standard.

The top part of the chain consists of an optical divide-by-3 system [18] to phase lock a Tm:YAG transfer laser (laser F) [19] at 148 THz (2.02 μm) to the probe laser (laser G) so that the heterodyne beat, $G - 3F$ is exactly 55 MHz. A counter monitors the phase lock through the 55-MHz beat. The frequency of the Tm:YAG laser is then measured by mixing its output with that of an auxiliary CO_2 laser (laser E) on a MIM diode to produce the beat, $F - 5E \simeq 1525$ MHz [19]. A tracking oscillator is

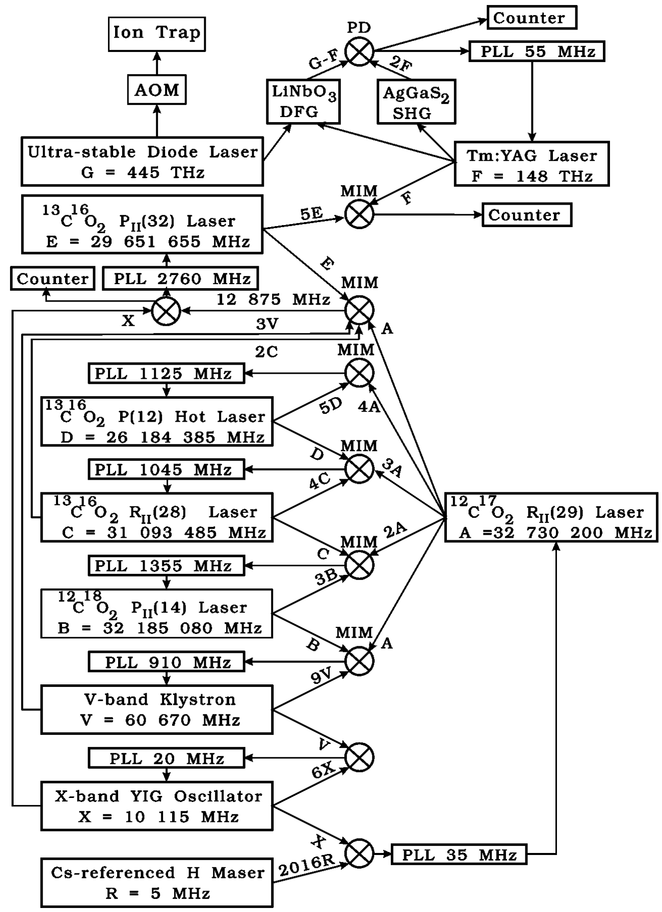

FIG. 1. The frequency chain used in the measurement of the frequency of the $5s\,^2S_{1/2} - 4d\,^2D_{5/2}$ transition in the $^{88}Sr^+$ ion with respect to a Cs primary time standard.

offset phase locked to the beat $F - 5E$ and the frequency counted. Laser E is locked to the infrared frequency chain through the beat $E + A - 2C - 3V - X = 2760$ MHz as shown in Fig. 1. A tracking oscillator is phase locked to the beat and the frequency counted. The typical sample standard deviation of the 1-s count is less than 50 Hz and drifts are below 100 Hz/min.

The frequency of the unshifted output from the 445-THz ultrastable laser is determined from the known frequencies of the oscillators in the IR chain and the 1-s readings from the counters measuring the beats $F - 5E$ and $E + A - 2C - 3V - X$. The counters are synchronized through software triggering and have a dead time of approximately 0.2 s per reading. Because of the low systematic drift rates of only a few Hz/s, better synchronization of the counters is not necessary for the present level of precision.

Chain measurements were performed on four separate days. 11 August 1997: with laser E phase locked to the IR chain for an integrated measurement period of 21 s; and 1 September 1998, 3 September 1998, 11 November 1998: with laser E frequency locked to the IR chain and the beat frequency counted for integrated measurement periods of 3260, 6880, and 1100 s, respectively. The duration of some experimental runs exceeded 10 min. Chain measurements during and directly adjacent to samples where the beat frequency, $G - 3F$, differed by more than 2 Hz from 55 MHz, indicating cycle slips in the divide by three system [18], were rejected.

The observed 1-s Allan deviation of the chain-measured probe laser frequency ranged from 3×10^{-12} on 11 November 1998 to 1×10^{-11} on 1 September 1998 and depended on the S/N ratio of the various heterodyne beats and the environmental noise levels. Examination of the Allan deviation showed that the fluctuations decreased as $\tau^{-1/2}$ for averaging times, τ up to 100 s. The chain measurements were averaged in bins corresponding to the 20-s measurement cycles of the lock to the ion transition and, together with the AOM-shifted frequency offset of the probe from the ion transition, an absolute determination of the center ion transition frequency was determined for each 20-s sample. A correction was applied to the AOM-shifted offset to account for drifts in the ultrastable laser and the resulting lag in the lock to the ion transition [16]. Such corrections never exceeded 100 Hz and were more typically only a few tens of hertz. Drifts in the ultrastable laser were clearly detected and followed by the chain measurements. The distribution of samples for each particular day was observed to be Gaussian having HWHM's of 1300 Hz for 1 September 1998, 680 Hz for 3 September 1998, and 450 Hz for the 11 November 1998 data. For the 11 August 1997 data, the final uncertainty of 3000 Hz is due to statistical fluctuations and the small number of measurements. For the other days, the quantity of data was much larger and the uncertainty due to the statistical fluctuations was exceeded by that due to the limited long-term stabilities of the measurement apparatus and the lock of the AOM-shifted probe laser to the ion. For example, by observing the stability of the determined Zeeman splitting of the ion inner transitions, it was observed that the probe laser lock servo reached a stability floor of 2×10^{-13}. In addition, when the Allan deviation of the determined ion frequency values were examined over greater averaging times on long continuous runs, stability floors of 1.4×10^{-12}, 6×10^{-13}, and 4.5×10^{-13} were observed for 1 September 1998, 3 September 1998, and 11 November 1998, respectively.

The average transition frequency for each day was determined by the weighted mean of the 20-s samples and the limiting stabilities were utilized as the current uncertainty in the mean value. These results are plotted in Fig. 2. The dashed line represents the weighted mean of these four values and is equal to $f_{S\text{-}D} = 444\,779\,044\,095.4 \pm 0.2$ kHz (1σ).

The largest source of possible systematic uncertainty in the measurements is cycle slips in the phase-locked loops of the IR chain or tracking oscillators. Since the tracking oscillators use relatively broad bandwidth servos to lock

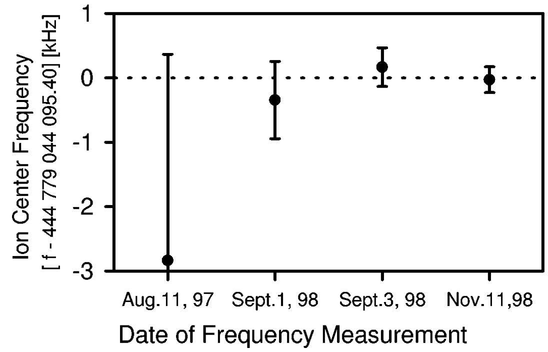

FIG. 2. The frequency of the Sr^+ ion S-D transition frequency as measured on each of the experimental days. The dashed line represents the average value of $f_{S\text{-}D} = 444\,779\,044\,095.4$ kHz.

voltage controlled oscillators to moderately stable beat signals, it is unlikely that slips occurred in the tracking oscillators, except possibly on 11 August 1997 when the S/N ratio of the beatnote, $F - 5E$ was below 10 dB. A previous search for cycle slips in the IR chain [17], found that if cycle slips do occur, they are so rare that they are of no importance at the present level of precision. The S/N ratios of the chain beats during the present experiment were as good as or better than those used in the previous study and an analysis of the 1-s counter readings indicates that the IR chain lasers were more tightly locked. No systematic shifts in the measured ion transition frequency were detectable as the bias magnetic field was varied between 5 and 19 μT.

Systematic shifts in the measured frequency due to the ion itself are much smaller than the measurement uncertainty of 200 Hz. Table I summarizes the calculated sources of systematic uncertainty affecting the ion in the current experiment. From measurements of the spectral sideband amplitudes at the trap's secular frequencies, an ion kinetic temperature of $T = 15 \pm 5$ mK has been calculated [20]. Similar measurements of the sidebands at the rf drive frequency indicate that the ion is displaced by stray electric fields out of the rf trapping node. The second-order Doppler and Stark shifts have been estimated from this motion [21] and the electric fields to be 0.13 and 0.2 Hz, respectively. Since the $4d\,^2D_{5/2}$ level possesses a quadrupole moment [22] of $Q = 1.43 \times 10^{-20}$ m^2, this moment can interact with the electric field gradient within the trap producing a level shift. Estimates of the field gradient arising mainly due to coated Sr metal on the trap structure indicate that the electric quadrupole shift is <0.5 Hz. The ac Stark shift due to perturbation of the S and D levels by blackbody radiation has been calculated [23] to be 0.16 Hz. An upper limit on the magnitude of stray ac magnetic fields has been determined from measurements with and without the magnetic shielding of the linewidths for a number of Zeeman components having different shift sensitivities.

TABLE I. Ion related sources of systematic uncertainty and their calculated values in the present experiment.

Source	Shift of line center	Magnitude
Second order Doppler effect	0.13 Hz	3×10^{-16}
Quadratic Stark shift	0.2 Hz	5×10^{-16}
Electric quadrupole shift of $4d\,^2D_{5/2}$ level	$<$0.5 Hz	$<1 \times 10^{-15}$
Blackbody ac Stark shift	0.16 Hz	4×10^{-16}
ac magnetic fields	$<$0.2 Hz	$<5 \times 10^{-16}$
Quadratic Zeeman shift (static field)	15 mHz	3×10^{-17}
Collisions	$<$10 mHz	$<2 \times 10^{-17}$

In summary, we have performed the first Cs-based frequency measurement of a visible transition in a single trapped and laser-cooled ion standard. A measurement of the diode probe laser light stabilized to the 445-THz electric quadrupole allowed transition yielded a determination of the absolute frequency with a standard uncertainty of 0.2 kHz ($\delta\nu/\nu = 4.5 \times 10^{-13}$). The present measurement represents a factor of 100 improvement in the knowledge of the single Sr^+ ion reference frequency. From a consideration of the trapped ion operating parameters and environment, the systematic shifts associated with the trapped single-ion system are on the 10^{-15} level and could be directly improved by further reduction of ion micromotion, reduction of stray fields in the trap structure, and further shielding. The accuracy limitation of the present measurement is limited by the stabilities of the laser lock to the ion transition and the chain measurement apparatus. Improvements to the probe laser linewidth and drift would serve to improve the stability of the laser lock to the ion transition. Nevertheless, the present measurement makes the $^{88}Sr^+$ transition one of the best known in the visible region of the electromagnetic spectrum.

We would like to acknowledge the important contributions of G. R. Hanes for the early supervision of the single ion research program at NRC and helpful discussions with J. S. Boulanger during the present measurements. The excellent technical support provided by R. Pelletier, B. Hoger, W. Cazemier, W. Boland, and T. Cassidy in the construction of the apparatus is gratefully appreciated.

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