$^{88}Sr^{+}$ 445-THz Single-Ion Reference at the 10^{-17} Level via Control and Cancellation of Systematic Uncertainties and Its Measurement against the SI Second

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We describe experiments and measurements on a trapped and laser-cooled single ion of $^{88}Sr^{+}$ which, when probed on its reference $5s\,^2S_{1/2} \rightarrow 4d\,^2D_{5/2}$ transition at 445 THz, provides an optical frequency standard of evaluated accuracy outperforming the current realization of the SI second. Studies are presented showing that micromotion-associated shifts of the standard can be reduced to the 10^{-18} level and uncertainties in the blackbody-induced shifts for the current system are at the low 10^{-17} level due to the relatively well-known polarizability of the strontium ion system and careful choice of the trap structure. The current evaluated systematic shifts for the ion transition are at a fractional uncertainty of 2×10^{-17}. An absolute frequency measurement performed over a two-month period relative to a maser referenced to the SI second via Global Positioning System time transfer has determined the center frequency for the transition at $\nu_{SD} = 444\,779\,044\,095\,485.5 \pm 0.9$ Hz (1σ).

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From precision tests of relativity to the increased understanding of atomic physics and fundamental constants, atomic time standards have helped shed light on subtle physical phenomena that underlie fundamental concepts of physics [1–3]. In addition, these references underpin the SI system which forms the basis of physical measurement. New experimental systems employing dipole forbidden optical transitions in trapped and laser-cooled ions and neutral atoms have been shown to be excellent next-generation realizations of ultra-accurate frequency references [3–14]. Such systems now have evaluated accuracies that exceed the current realizations of the definition of the SI second as realized by a microwave transition in Cs atoms [15]. Among the trapped ion systems studied, the $^{88}Sr^{+}\,5s\,^2S_{1/2} \rightarrow 4d\,^2D_{5/2}$ electric dipole forbidden transition at 445 THz (674 nm) has a very high quality factor ($Q = \nu_0/\Delta\nu = 10^{15}$) and possesses a relatively simple electronic energy level scheme that is favorable for use with widely available solid state laser technology [16]. This ion system was the first to be directly measured against the Cs realization of the second [17] and has been established as one of the secondary representations of the SI second. This system has been primarily studied at the Frequency and Time Group, National Research Council of Canada (NRC) and the National Physical Laboratory (NPL) in the United Kingdom [7,11] with additional groups recently providing results [18]. We describe new results and methods by which the systematic shifts for the system have been reduced to the low 10^{-17} level via operation of the trap system under conditions which reduce and in some cases null out significant uncertainties which dominate many optical reference systems. The work also provides a new measurement of the transition center frequency relative to the SI second.

A single atomic ion of $^{88}Sr^{+}$ is held in an electrodynamic trapping field using a miniature radio frequency end-cap trap [19]. The trap has an end-cap separation $2z_0 = 0.541 \pm 0.015$ mm and radius $r_0 = 0.250 \pm 0.005$ mm with the end-cap electrodes made of molybdenum. The entire trap structure is housed in an ultrahigh vacuum chamber where the background gas pressure is 16 nPa composed primarily of H_2. At these pressures, the mean classical collisional time is on the order of 10^3 s [20]. Continuous storage times of over 1 week under laser cooling have been obtained thus suggesting that the $^{88}Sr^{+}$ system does not have a significant cross section for loss from the trap by collision with impurity gas partners. The ion trap is operated at a trap frequency of $\Omega = 2\pi \times 14.4$ MHz with a voltage amplitude of $V_0 = 212$ V. Under these conditions, the ion in the trap oscillates with motional secular frequencies of $\omega_Z/2\pi = 2.3$ MHz axial and $\omega_{X,Y}/2\pi = 1.2$ MHz in the radial direction. Loading of the ion trap is performed via photoionization of a weak Sr atomic beam using two-step excitation using radiations at 461 nm (S-P resonance line) and 404 nm (excitation into the continuum) [21]. The trapped ion is laser cooled on the $5s\,^2S_{1/2} - 5p\,^2P_{1/2}$ resonance line transition using a direct diode laser source stabilized to a nearby Rb saturated absorption line at 422 nm [22]. A diode pumped fiber laser provides a repump for the $4d\,^2D_{3/2} - 5p\,^2P_{1/2}$ transition at 1092 nm. Laser powers and detunings at 422 nm and 1092 nm were carefully adjusted to optimize the 422 nm fluorescence and laser cooling [18] such that the measured ion kinetic temperature was $T = 1.8 \pm 0.8$ mK (approaching the Doppler limit for this transition of 0.5 mK) [5]. Detected single-ion fluorescence rates of 10^4 counts per second are observed with a signal to background ratio of 70. The reference or clock transition used is the electric

quadrupole allowed $5s^2S_{1/2} - 4d^2D_{5/2}$ transition whose natural line width is 0.4 Hz [16]. Detection of excitation on this narrow transition is made by observing the interruption of the strong 422 nm S-P fluorescence which indicates that the single-ion system has undergone excitation to the upper state of the S-D transition. The reference transition is excited using an ultrastable diode probe laser which employs a high finesse Corning Ultra-Low Expansion Glass (ULE) reference cavity thermally stabilized on its zero crossing of the material's thermal expansion coefficient. The probe system has been described in greater detail elsewhere [23] and consistently shows resolution of the ion reference transition equivalent to the Fourier transform of the pulse width down to line resolutions of 4 Hz. The drift rate of the cavity is extremely linear and stable over experimental periods of days with a typical drift rate of 10 mHz/s. In this work, excitation of the clock transition is typically obtained with a 100 ms pulse of 35 pW power focused to a spot size of $\omega = 25 \pm 2$ μm. The ion resides in a background magnetic field of 2.025(3) μT which serves to separate the individual Zeeman multiplet at an inner component separation of ± 11.3 kHz from line center. Stabilization to line center is achieved by measuring the transition rate at the half power points of pairs of symmetric Zeeman components [11]. By using different component pairs one can directly evaluate the electric-quadrupole shift and tensor component of the Stark effect on the transition and cancel these effects at the 10^{-18} level or better [11].

Reduction of the ion micromotion to extremely low levels is performed in two stages. Adjustment of DC trim electrodes in the radial plane and axial DC bias while observing the trap region with a CCD camera as the ion alternates between tight and weak trap confinement allows observation of a possible strong displacement by any residual trap asymmetry, patch potentials, stray fields, etc. [13]. This adjustment rapidly converges the system to a low state of micromotion. Final adjustment is achieved by observing the transition rate at the micromotion sideband relative to that obtained by the carrier [10,24]. This not only allows correction of the stray DC offsets of the ion but also precision correction of the phase shifts between the end-cap electrodes which also can result in significant micromotion [24]. Measurements were obtained by performing scans using probe laser radiation sent individually along three orthogonal directions to obtain the total magnitude of micromotion. In these measurements, carrier-to-sideband ratios of over 300:1 were observed for each direction probed.

The relative Stark shift arising from this micromotion is at the level of 2×10^{-17}. These low shifts are observed to be constant over several hour periods as evidenced by measurements of the micromotion sideband-to-carrier ratio for overnight experimental run periods. The $^{88}Sr^+$ system possesses another advantage in that the micromotion-induced scalar Stark effect and time dilation shifts are of opposite sign and comparable magnitude. As shown in [10,24], the total shift of the transition frequency due to micromotion effects can be expressed as the sum of the scalar Stark shift and time dilation shifts,

$$(\Delta\nu)_m = \left[2\gamma_s\left(\frac{M\Omega}{q}\right)^2 - \nu_0\left(\frac{1}{c}\right)^2\right]\frac{\Omega^2\sigma(1)}{(k\cos\phi)^2\sigma(0)}, \quad (1)$$

where ν_0 is the transition center frequency, Ω is the angular frequency of the trap field, k is the amplitude of the wave vector of the probe field, ϕ is the angle between the observation direction and the direction of micromotion, M and q are the ion's mass and charge, $\gamma_S = \partial\nu/\partial E^2$ is the Stark shift rate, and $\sigma(1)/\sigma(0)$ is the ratio of the first observed sideband intensity to the carrier line. Inspection of the above equation shows that with proper choice of the trap frequency Ω these two effects can be made to cancel each other. Based on previous calculations and more precise results reported by Jiang *et al.* [25] calculations of the scalar polarizabilities of the lower and upper states of the clock transition yield operation of the trap at a critical frequency of $\Omega/2\pi = 14.39$ MHz. Based on the uncertainties in these values, an assigned total uncertainty due to micromotion of 1×10^{-18} is determined.

Another key systematic shift dominating many current optical frequency standards is the blackbody-radiation-induced Stark shift of the reference transition due to the room temperature thermal environment of the enclosure and immediate environment surrounding the ion. The $^{88}Sr^+$ ion possesses no electric dipole allowed transitions from the $5s^2S_{1/2}$ or $4d^2D_{5/2}$ states at frequencies below 290 THz (1.03 μm). Thus the dynamic (resonant) corrections due to the thermal radiation are very small and the shifts are dominated by the static scalar polarizability. Calculations of blackbody shift coefficients yield low uncertainties due to the preferential energy level arrangement of the strontium ion [25]. We next evaluate the nonuniformity of the thermal environment of the objects surrounding the ion including the ion trap electrodes. An identical trap structure was constructed and fitted with precision thermistors so that the actual equilibrium temperature could be determined on several critical surfaces. It was found that the end-cap electrode faces and surrounding materials increased in temperature relative to the background chamber environment by up to 32 °C when operated at the typical trap voltages. The effect of this nonuniform thermal distribution on the net blackbody shift is strongly suppressed by our choice of trap electrode material. The electrode faces are made from molybdenum that was polished to better than $\lambda/2$ surface finish at visible wavelengths and whose emissivity is very low ($\epsilon = 0.06$). The shift arising from the heated trap structure only contributes a displacement of 0.013 Hz to the total shift (0.249 Hz) which is dominated by the walls of the chamber. Simulations of single and multiple reflections of thermal radiation of the nearby trap structure and environment have been performed and show that the total uncertainty added by the heated electrode

surfaces are small compared to the current uncertainties assigned to the scalar Stark shift coefficients. Precision calibrated thermistors located on the trap chamber measure the wall temperature and thus allow accurate determination of the blackbody Stark shift to the level of 2.2×10^{-17} fractional uncertainty.

The $^{88}Sr^{+}$ system has a nuclear spin of $I = 0$ and thus possesses a linear Zeeman shift for all components of the reference transition. The current experimental arrangement employs a double magnetic shield and the linear shift is rapidly compensated via the method of measuring symmetric Zeeman pairs [11]. This compensation system is very robust and magnetic field drifts and fluctuations remain within the few nT level over several hours. AC magnetic field penetration within the shield environment can lead to shifts of the line center, however the spectral resolution of $\delta\nu < 10$ Hz used places any dominant AC-induced features (such as the 60 Hz sidebands due to power-line frequencies) well away and isolated from the studied carrier. Any of these features are less than $1/50$ of the intensity of the central transition carrier and thus negligible in the current analysis. The atomic system possesses a very small second-order Zeeman shift at low field intensities. For the average of all pairs of components with different upper state m_J, the field displacement is 3.1 μHz/μT^2 [10]. Table I lists the principal systematic uncertainties discussed and yields a total estimated systematic uncertainty of 2×10^{-17} which compares favorably to the best high-quality performance achieved by other optical clock candidate systems and exceeds the current performance of Cs atomic frequency standards [15].

A key goal in the development of optical atomic clocks is the relation of the optical transition frequency relative to the best current realizations of the SI second. A mode-locked erbium-doped fiber laser frequency comb has been developed which provides continuous and long term linkage between the ion optical frequency and the NRC ensemble of rf standards [26]. The comb is stabilized to a reference frequency from a precision synthesizer whose signal is referenced from an NRC active H maser. Measurements were performed over a two month period with data collected relative to the maser frequency. The maser is calibrated in its absolute offset from the SI second through international GPS satellite intercomparison using the Natural Resources Canada Precise Point Positioning (PPP) data-processing technique [27] and using the Bureau International des Poids et Mesures (BIPM) circular-T reports. Using such corrections together with NRC inhouse frequency standards, it was possible to obtain uncertainties of the measured ion center frequency to a resulting final value of 0.9 Hz. Compensation of the servo error due to the drift rate of the laser was addressed by using the experimental drift rates of the probe versus the ion and calculating the resulting servo offsets. Corrections of the servo offset postprocessed by this method yielded uncertainties in the line center due to servo error of below 0.03 Hz. These servo offset corrections have been applied to the data shown in Fig. 1 and the final frequency determination. Stabilities in the ion versus NRC H4 maser data followed the maser stability with an Allan standard deviation of 2×10^{-14} at 100 s averaging to below 5×10^{-16} at 10^4 s. The stability of the probe laser lock to the ion resonance was estimated by fitting a third-order polynomial to the probe laser ULE reference cavity drift and analyzing the offsets from this drift. It was found that the servo provided effective stabilization at the 200 s level with the stability improving as $1.0 \times 10^{-14}/\sqrt{\tau}$ for longer times until the cavity thermal noise limit of the ULE reference cavity is encountered at 2.6×10^{-16} [23]. Typical measurement periods varied from 5 hours to over 2.5 days. Evaluations of the systematic shifts such as micromotion levels were performed at the beginning and end of such runs to confirm the ion system remained within the low uncertainty limits as specified in Table I. The mean of the

TABLE I. Contributions to the systematic shifts and fractional uncertainty for the $^{88}Sr^{+}$ ion system.

Effect	Shift (Hz)	Uncertainties [u/$\nu_0(10^{-18})$]
Micromotion effects	0	1
Blackbody AC Stark shift	0.249	22
1092 nm AC Stark shift	−0.002	2
674 nm AC Stark shift	0.000 014	0.02
Electric quadrupole + Tensor Stark effect	0	≤ 1
Residual second-order Doppler + Stark effect (thermal)	−0.0013	1.3
Quadratic Zeeman shift	0.000 037	0.002
Total	0.246	22

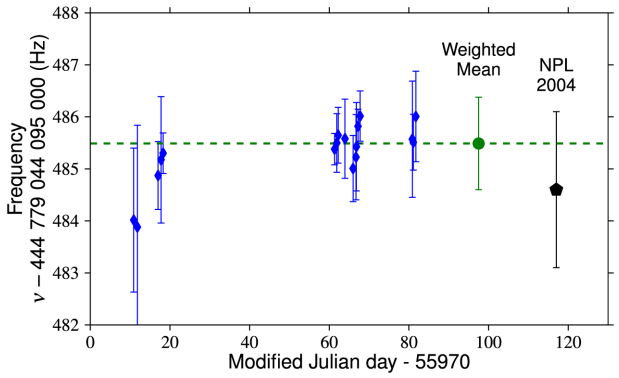

FIG. 1 (color online). Summary of absolute frequency measurements of *S-D* transition during a two-month period showing weighted mean value with maser uncertainty included (circle) and comparison with measurement performed in Ref [7] (pentagon). The dates of measurement for the individual runs are given on the horizontal axis using the modified Julian day subtracted by 55970.

measurements presented in Fig. 1 determined an unperturbed center frequency for the $5s\,^2S_{1/2} \rightarrow 4d\,^2D_{5/2}$ transition to be 444 779 044 095 485.5(0.9) Hz. The total fractional contribution to the uncertainty arises primarily from the rf time reference which gives 2×10^{-15}. In addition, the weighted mean statistical uncertainty of the sets of data yielded 2×10^{-16} together with the current systematic uncertainties of 2×10^{-17} and provided a minor contribution to the total uncertainty. The current result represents an order of magnitude reduction in the evaluated uncertainty of the ion compared to [7] and the absolute frequency measurement is in agreement with the given absolute frequency of the NPL group of 444 779 044 095 484.6(1.5) Hz [7].

We have shown low systematic uncertainties for the $^{88}Sr^+$, $5s\,^2S_{1/2} \rightarrow 4d\,^2D_{5/2}$ optical frequency reference below that of the current realization of the SI second and have provided an improved value for the transition frequency. The small systematic shifts together with the inherent intrinsic advantages of using a relatively simple system operating for several days make it a very strong candidate for wide use as a realization of the second and a potential probe for sensitive phenomena such as the precision measurement of gravitational redshift [2], precision atomic physics [3], and to improve worldwide international atomic time (TAI). Improvements in the current systematic shifts of the $^{88}Sr^+$ ion standard will occur primarily through improved knowledge of the scalar Stark shift parameters of the lower and upper state of the clock transition. Experimental comparisons between similar trap systems will also be able to further quantify and reduce the uncertainties associated with the blackbody radiation shift. It is quite possible that through improved experimental measurement and theoretical calculation such improvements can be achieved to readily reach total uncertainties below the 10^{-17} level.

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