Precision Atomic Spectroscopy for Improved Limits on Variation of the Fine Structure Constant and Local Position Invariance

T. M. Fortier,[1,2] N. Ashby,[2] J. C. Bergquist,[2] M. J. Delaney,[2,*] S. A. Diddams,[2,†] T. P. Heavner,[2] L. Hollberg,[2] W. M. Itano,[2] S. R. Jefferts,[2] K. Kim,[2,‡] F. Levi,[2,§] L. Lorini,[2] W. H. Oskay,[2] T. E. Parker,[2] J. Shirley,[2] and J. E. Stalnaker[2]

[1]P-23 Physics Division MS H803, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

[2]Time and Frequency Division MS 847, National Institute of Standards and Technology, Boulder, Colorado 80305, USA

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We report tests of local position invariance and the variation of fundamental constants from measurements of the frequency ratio of the 282-nm $^{199}Hg^+$ optical clock transition to the ground state hyperfine splitting in ^{133}Cs. Analysis of the frequency ratio of the two clocks, extending over 6 yr at NIST, is used to place a limit on its fractional variation of $<5.8 \times 10^{-6}$ per change in normalized solar gravitational potential. The same frequency ratio is also used to obtain 20-fold improvement over previous limits on the fractional variation of the fine structure constant of $|\frac{\dot{\alpha}}{\alpha}| < 1.3 \times 10^{-16}$ yr^{-1}, assuming invariance of other fundamental constants. Comparisons of our results with those previously reported for the absolute optical frequency measurements in H and $^{171}Yb^+$ vs other ^{133}Cs standards yield a coupled constraint of $-1.5 \times 10^{-15} < \dot{\alpha}/\alpha < 0.4 \times 10^{-15}$ yr^{-1} and $-2.7 \times 10^{-15} < \frac{d}{dt} \ln\frac{\mu_{Cs}}{\mu_B} < 8.6 \times 10^{-15}$ yr^{-1}.

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Measurements comparing the relative rates of two atomic clocks that are based on very narrow transitions between well-resolved and relatively unperturbed atomic energy levels currently report among the highest precision attained for any fundamental quantity. Such measurements, which can yield greater than 15 digits of frequency accuracy, permit tests of fundamental postulates of physics, including searches for variations of fundamental constants. In this Letter, we analyze measurements of the frequency ratio of the 282-nm optical transition in $^{199}Hg^+$ to that of the 9.193 GHz ground state hyperfine splitting of ^{133}Cs, taken over a 6 yr period. These measurements allow improved limits to be placed on violation of local position invariance (LPI) and on the temporal variations fundamental constants.

The possibility that cosmological changes might result in variations of physical constants constitutes a violation of Einstein's equivalence principle, one of the tenets of general relativity. Searches for such violations have received renewed attention partially because they are predicted in emerging theories aimed at unifying gravitation and quantum mechanics [1,2]. In particular, much interest has been focused on possible changes in the fine structure constant α, following the analyses of astronomical [3–7] and geochemical data [8–10]. These results have reported sensitivities to fractional variations of α at the level of $|\frac{\dot{\alpha}}{\alpha}| \simeq 10^{-16}$ yr^{-1} and $|\frac{\dot{\alpha}}{\alpha}| \simeq 10^{-17}$ yr^{-1}, respectively, assuming a linear drift over cosmological (10^{10} yr) and geological time scales (10^9 yr). The reported limits on α variation, however, are met by substantial disagreement between groups. Different analyses performed on data from the Oklo reactor yielded null results in Refs. [8,9] and a negative α variation in Ref. [10]. The analyses of quasar absorption spectra are also met with disagreement; null results are reported in Refs. [6,7], while a positive variation is concluded in Refs. [3,4]. Alternatively, the comparison of transition frequencies in different atomic species can provide highly sensitive and reproducible laboratory-based measurements of present-day changes in α. Recent improvements in the measurement of the Hg^+-clock transition frequency allow competitive limits to be placed on any time variations of α in our current epoch.

Measurements of atomic frequency ratios can also be used to search for violations of LPI. Local position invariance requires that the outcome of any local nongravitational experiment is independent of where or when in the universe the experiment is performed. By looking for a variation in the ratio of two atomic frequencies that is correlated with changes in the gravitational potential caused by Earth's rotation about the Sun, we are able to provide additional limits to variations of LPI at levels exceeding those of previous clock comparisons [11].

The frequency ratio presented here is that of an optical electronic transition in Hg^+ to the microwave hyperfine transition of Cs used to define the SI second. Technical details pertaining to both the Cs and the Hg^+ clocks can be found in Ref. [12] and Refs. [13,14], respectively. Briefly, a single Hg^+ ion is confined in an rf Paul trap and laser cooled to ~1.7 mK via the strongly allowed first resonance line at 194 nm. The reference for the optical clock is the $^2S_{1/2}(F=0) \rightarrow {}^2D_{5/2}(F=2, M_F=0)$ electric quadrupole allowed transition at 282 nm with a natural lifetime of 86 ms. Transitions to the $^2D_{5/2}$ level are detected via electron shelving [15] using the 194-nm radiation. Spectroscopy of the narrow clock transition is performed by a frequency quadrupled fiber laser, where a portion of the light is used for stabilization to a low drift-rate (<1 Hz/s) high-finesse optical cavity at 563 nm [16].

The transition frequency of the Hg^+-ion clock is compared to the 9 192 631 770 Hz ground state hyperfine splitting of Cs as realized by a Cs-fountain clock. NIST-F1 has been in operation since 1998 and has undergone 15 formal accuracy evaluations that have been submitted to the Bureau International des Poids et Mesures (see Refs. [12,17]).

A Ti:sapphire based femtosecond laser frequency comb [18] performs the optical-to-microwave synthesis that compares the rates of the Cs and Hg^+ clocks (see Fig. 1). Stabilization of the comb offset frequency, f_0, via self-referencing, and locking of one of the comb modes to light at half the Hg^+-clock transition frequency produce a laser repetition rate, $f_{rep} \propto \nu_{Hg}/n$. Using a H maser as an intermediate reference, this signal is compared to the transition frequency of the Cs-fountain clock. The continuous comparison of the H maser and Cs-clock transition frequencies ensures that any drift from the H maser is eliminated in the final calculation of the Hg^+-clock transition frequency. Corrections due to systematic biases are also made to obtain the unperturbed Cs and Hg^+ transition frequencies. The dominant corrections for the Cs clock include second order Zeeman, black body, and collisional shifts [19]. Those for Hg^+ are dominated by the second order Zeeman shift. A fractional correction of $\approx 5 \times 10^{-16}$ is also included to account for the gravitational redshift resulting from the difference in elevation between the Cs-fountain clock and the Hg^+ clock.

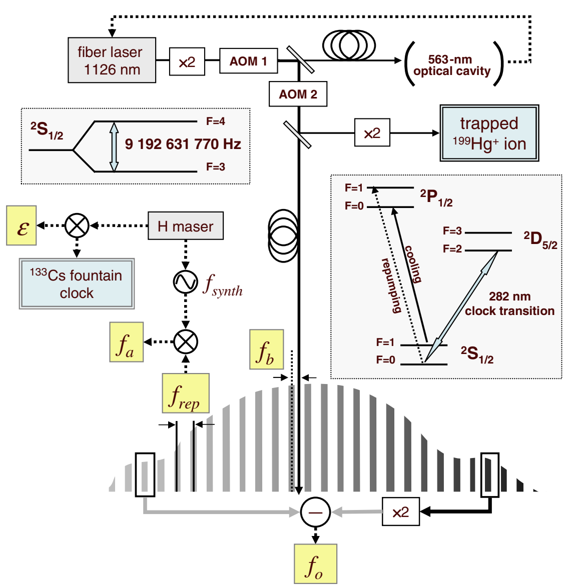

FIG. 1 (color online). Simplified experimental setup for comparison of the single Hg^+-clock transition to the Cs microwave clock transition via an octave-spanning frequency comb. Acousto-optic modulators, AOM1 and AOM2, stabilize the fiber laser to a high-finesse optical cavity and to the Hg^+ transition, respectively. The frequency comb repetition rate, f_{rep}, is measured relative to the Cs-fountain clock via a synthesizer stabilized to a H maser. The absolute Hg^+-transition frequency is calculated from $\nu_{Hg} = 2 \times [n \times (f_a + f_{synth}) - f_0 - f_b](1 + \epsilon)$, where f_0 is the comb offset frequency, f_b is the beat between comb line n and the Hg^+-clock laser, $f_a = f_{rep} - f_{synth}$, and ϵ is the fractional frequency deviations between the H maser and the Cs-clock transition frequencies.

Past measurements that make up the 6 yr time record (Fig. 2) can be found in Refs. [20] (prior to January 2001), [21] (January 2001 to January 2003), and [22] (January 2004 to July 2005). The observed decrease of the error bars as seen in Fig. 2 is primarily due to improved measurements of the Hg^+-clock systematic shifts, which had previously limited the measurement uncertainties from 2000 to 2004. Specifically, the results from 2000 to 2004 had been artificially limited by a conservative estimate of 10 Hz uncertainty in the electric quadrupole shift of the Hg^+ $^2D_{5/2}(F = 2, M_F = 0)$ state [21]. A recent measurement of the quadrupole moment by Oskay *et al.* [23] along with some earlier measurements of the trap secular frequencies (giving an upper bound to the static trap electric field gradients) has facilitated a revision of the previously published systematic uncertainty assigned to measurements of the Hg^+-clock transition frequency. Consequently, the total uncertainty in the first 25 data points in Fig. 2 reflects the reduction in the electric quadrupole uncertainty from 10 to 1 Hz. With decreasing systematic uncertainties over the years, the statistical uncertainty in later years began to dominate the total uncertainty of the measurements. As a result of the reduction in the systematic uncertainties, the last five measurements (from 2005 to 2006) were obtained with longer averaging times, with the statistical uncertainty limiting the measurement error.

The most recent measurement of the Hg^+-clock transition (the last data point in Fig. 2) gave $\nu_{Hg} = 1\,064\,721\,609\,899\,145.9 \pm (1.1)$ Hz, obtained from 10^5 s of data between 8 March 2006 and 10 March 2006. The

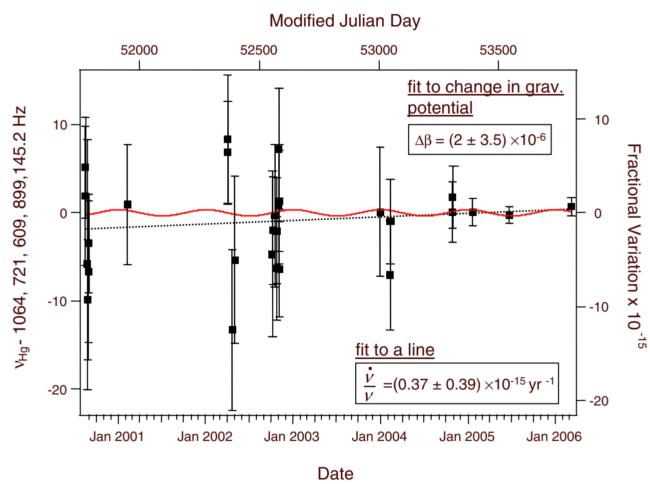

FIG. 2 (color online). Six year time record of the absolute frequency measurements of the clock transition in Hg^+. The offset is the weighted mean of the Hg^+-clock transition frequency of all the data. Shown is a weighted fit of the data to a line (dotted line). Also shown is a weighted fit of the data to changes in the normalized gravitational solar potential (solid line) to search for any violations of local position invariance.

total measurement uncertainty of 1×10^{-15} is the quadrature sum of the total statistical uncertainty (0.9×10^{-15}) and the uncertainties from the optical-to-microwave synthesis process (0.3×10^{-15}), and the uncertainties of the systematic biases of the Cs clock (0.41×10^{-15}) and the Hg^+ clock (0.03×10^{-15}). Details pertaining to the most recent measurement can be found in Ref. [19].

The time record in Fig. 2 can be used to search for time-dependent variations of the fine structure constant by looking for changes in the frequency ratio of $\nu_{\rm Hg}/\nu_{\rm Cs}$ (this ratio is implied in any absolute frequency measurement since the Cs transition frequency defines the SI second). In atoms, the fine structure constant $\alpha = e^2/4\pi\epsilon_o\hbar c \approx 1/137$ appears as a scaling factor that relates the relative energy scales of the electronic, fine, and hyperfine structures. The absolute measurement of an optical transition frequency permits a comparison of the electronic structure of one atom to the hyperfine structure of Cs. The α dependence of the optical transition of atom j yields $\nu_j \propto R_y F_j(\alpha)$, whereas the hyperfine interval of Cs exhibits a dependence $\nu_{\rm Cs} \propto \alpha^2 R_y (\frac{\mu_{\rm Cs}}{\mu_B}) F_{\rm Cs}(\alpha)$ (see, for example, Ref. [2]). Here, R_y is the Rydberg constant, μ_B is the Bohr magneton, and $\mu_{\rm Cs}$ is the magnetic moment of Cs. The factor $F_j(\alpha) \propto \alpha^{N_j}$ expresses the relativistic correction to the transition frequency as a power dependence on α, with the value of N_j differing for different atomic transitions.

Such a comparison also depends on the ratio of the magnetic moments $\frac{\mu_{\rm Cs}}{\mu_B}$. Because this ratio will likely accompany any changes in α, we employ an analysis similar to that used in Ref. [24] to determine the relationship between their coupled fractional variations. This is accomplished by analyzing the time dependence of the natural logarithm of the ratio of the optical and microwave frequencies:

$$\frac{d}{dt}\ln\left(\frac{\nu_j}{\nu_{\rm Cs}}\right) = \frac{d}{dt}\ln\left[\frac{\alpha^{N_j - N_{\rm Cs} - 2}}{(\mu_{\rm Cs}/\mu_B)}\right]$$
$$\text{or} \quad \frac{\frac{d}{dt}(\nu_{\rm Hg}/\nu_{\rm Cs})}{\nu_{\rm Hg}/\nu_{\rm Cs}} = \frac{\dot{\alpha}}{\alpha}N - \frac{d}{dt}\ln\frac{\mu_{\rm Cs}}{\mu_B}, \tag{1}$$

where $N = [N_j - N_{\rm Cs} - 2]$.

Dzuba *et al.* have calculated the relativistic correction factors for the Cs- and Hg^+-clock transitions to be $F_{\rm Cs}(\alpha) \propto \alpha^{0.8}$ and $F_{\rm Hg}(\alpha) \propto \alpha^{-3.2}$ [25]. Substituting these into Eq. (1), we obtain the dependence $\frac{d}{dt}\ln\frac{\nu_{\rm Hg}}{\nu_{\rm Cs}} \propto -6 \times \frac{\dot{\alpha}}{\alpha}$. Any drift in the ratio of the clock frequencies is determined by a weighted least squares linear fit to the historical time record (Fig. 2), which gives 0.39 ± 0.42 Hz yr^{-1}, or fractionally, $\frac{d}{dt}\ln\frac{\nu_{\rm Hg}}{\nu_{\rm Cs}} \approx (0.37 \pm 0.39) \times 10^{-15}$ yr^{-1}. Assuming that $\frac{d}{dt}\ln\frac{\mu_{\rm Cs}}{\mu_B} = 0$, this yields a 1-$\sigma$ limit of $\dot{\alpha}/\alpha = (-6.2 \pm 6.5) \times 10^{-17}$ yr^{-1}, which is 20 times more stringent than our previously reported limit [21].

To obtain a constraint on the coupled variation of α and $\mu_{\rm Cs}/\mu_B$, we compare the limit on the drift of the absolute frequency from our Hg^+ measurements with those of the hydrogen $1S \rightarrow 2S$ transition [24] and the $^{171}Yb^+$ $^2S_{1/2}(F=0) \rightarrow {}^2D_{3/2}(F=0, M_F=0)$ transition [26]. Because of its very light nucleus, the relativistic corrections for hydrogen are negligible; hence $N_H' = 0$. Based on calculations in Ref. [25], the relativistic α dependence for the Yb^+-clock transition is $F_{\rm Yb}(\alpha) \propto \alpha^{0.9}$. Returning to Eq. (1), we perform the substitution $y = \frac{d}{dt}\ln\frac{\mu_{\rm Cs}}{\mu_B}$ and $x = \dot{\alpha}/\alpha$ to obtain $y = Nx - \frac{d}{dt}\ln\frac{\nu_j}{\nu_{\rm Cs}}$, which relates the possible linear drift in $\mu_{\rm Cs}/\mu_B$ to that of α. Using the observed fractional frequency drift rates of $\frac{d}{dt}\ln\frac{\nu_{\rm Yb}}{\nu_{\rm Cs}} = (-1.2 \pm 4.4) \times 10^{-15}$ yr^{-1} [26] and $\frac{d}{dt}\ln\frac{\nu_{\rm H}}{\nu_{\rm Cs}} = (-3.2 \pm 6.3) \times 10^{-15}$ yr^{-1} [24], we obtain a plot for the constraints from the drift in the time records of absolute optical frequency measurements (see Fig. 3). The 1-σ limits on the possible drift rates of α and $\frac{\mu_{\rm Cs}}{\mu_B}$ are determined by minimizing the function $\chi^2(x, y) = \sum_{j=1}^{3} \frac{1}{\sigma_j^2}(y_j + \frac{d}{dt} \times \ln\frac{\nu_j}{\nu_{\rm Cs}} - Nx_j)^2$. The 1-$\sigma$ error ellipse is bounded by $\chi^2_{\rm min} + M = \chi^2(x, y)$, where $M = 1$ defines the contour. For the data in Fig. 3 we find $\chi^2_{\rm min} = 0.11$. The projection of the error ellipse onto the x and y axes yields the coupled constraints,

$$-1.5 \times 10^{-15} < \dot{\alpha}/\alpha < 0.4 \times 10^{-15} \text{ yr}^{-1},$$
$$-2.7 \times 10^{-15} < \frac{d}{dt}\ln\frac{\mu_{\rm Cs}}{\mu_B} < 8.6 \times 10^{-15} \text{ yr}^{-1}.$$

A decrease in the uncertainty of future absolute optical measurements, or the direct comparison of two optical clocks, will yield significantly better constraints on $\dot{\alpha}/\alpha$.

The time record of the frequency comparison of the optical-to-microwave transitions also allows for tests of

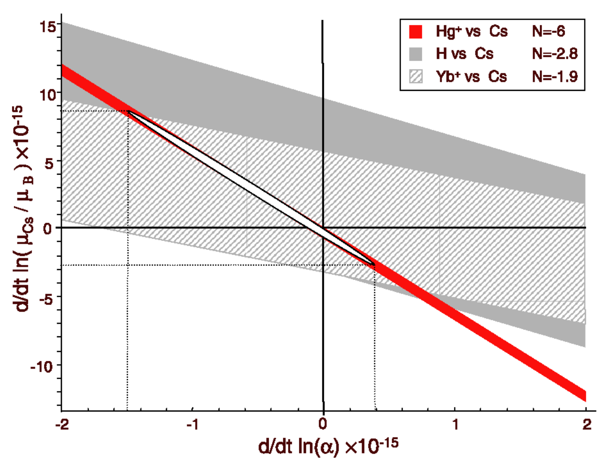

FIG. 3 (color online). Constraints on $|\dot{\alpha}/\alpha|$ and $|\frac{d}{dt}\ln\frac{\mu_{\rm Cs}}{\mu_B}|$ deduced from the measured fractional drift rates of the H, Yb^+, and Hg^+ absolute optical frequencies are determined by the projection of the 1-σ error ellipse on the x and y axes.

LPI. Local position invariance implies that the frequencies of two atomic clocks of different structure should suffer identical redshifts as they move together through a changing gravitational potential, ΔU [27]. If LPI is violated, the gravitational redshift of a clock frequency, $\frac{\Delta \nu}{\nu} = (1 - \beta) \times \frac{\Delta U}{c^2}$, will include a constant term, β, that is dependent on the internal structure of a particular clock (in general relativity $\beta = 0$). This would yield a change in the fractional difference of the two clocks of

$$\frac{\Delta(\nu_A/\nu_B)}{\nu_A/\nu_B} = (\beta_B - \beta_A)\frac{\Delta U}{c^2}. \quad (2)$$

To look for violations of LPI, we perform a weighted fit of the time record to look for deviations in the frequency ratio between the Hg^+- and Cs-clock transitions that follow changes in the gravitational potential as Earth orbits the Sun. The calculated potential is dominated by that of the Sun, but includes the effects of other celestial bodies in the solar system as well [28]. In Fig. 2, the only free fit parameter determines the amplitude of the maximum fractional frequency deviation to be $\frac{\Delta(\nu_{Hg}/\nu_{Cs})}{\nu_{Hg}/\nu_{Cs}} = 0.7 \pm 1.2 \times 10^{-15}$. This value is used in conjunction with the peak-to-peak change in the solar potential of order, $\frac{\Delta U}{c^2} \approx \frac{GM_{\odot}e}{c^2 a^2} \approx 3.3 \times 10^{-10}$, to place a limit on $(\beta_{Hg} - \beta_{Cs}) \leq 2 \pm 3.5 \times 10^{-6}$.

In summary, we have presented the most recent measurements of the absolute optical frequency of the Hg^+-clock transition. The reduction in the total uncertainty of the latest measurements and 6 yr time record have allowed for a more stringent limit to be placed on variations of fundamental constants, giving a 20-fold improvement over our previous limit of $|\frac{\dot{\alpha}}{\alpha}|$ [21]. The time record is also used to search for violations of LPI resulting from changes in the solar potential. Our limits are nearly ~8 times [29] more stringent than previous results presented by Bauch and Weyers [11], which put limits on variations between the hyperfine splitting of H relative to that of Cs. By comparing our result with previously reported absolute optical measurements of Yb^+ and H, we obtain a limit for $|\frac{\dot{\alpha}}{\alpha}|$ and $|\frac{d}{dt}\ln\frac{\mu_{Cs}}{\mu_B}|$. The comparison between two optical atomic clocks where the fractional uncertainties are projected to achieve the 10^{-17} level will yield much stricter limits on $|\frac{\dot{\alpha}}{\alpha}|$.

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*Current address: Physics Department, University of Wisconsin—Madison, 1150 University Avenue, Chamberlin Hall, Room 2320, Madison, WI 53706-1390, USA.

†Electronic address: sdiddams@boulder.nist.gov

‡Current address: School of Mechanical Engineering, Yonsei University, 134 Shinchon-dong, Seodaemum-gu, Seoul 120-749, Korea.

§Current address: Istituto Nazionale di Ricerca Metrologica, Strada delle Cacce 91, I-10135 Torino, Italy.

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