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The Mercury-Ion Optical Clock and the Search for Temporal Variation of Fundamental Constants

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***Abstract*— The repeated comparison of atomic frequency standards based upon different transitions enables the search for time variation of the fundamental constants that determine the transition frequencies. Over the course of two years we compared the frequency of the ^{199}Hg$^+$ $5d^{10}6s\ ^2S_{1/2}(F=0) \longleftrightarrow 5d^9 6s^2\ ^2D_{5/2}(F=2)$ electric-quadrupole transition at 282 nm with the frequency of the ground-state hyper¿ne splitting in neutral ^{133}Cs that de¿nes the SI second. These measurements constrain any fractional time variation of the ratio $\nu_{\mathrm{Cs}}/\nu_{\mathrm{Hg}}$ between the two frequencies to be less than $\pm\ 7\times10^{-15}$yr^{-1} (1σ uncertainty). According to recent atomic structure calculations, this sets an upper limit to a possible fractional time variation of the product $g_{\mathrm{Cs}}(m_e/m_p)\alpha^{6.0}$ at the same level.**

I. Introduction

A long-standing question in physics is whether the fundamental "constants" of nature are actually changing in time [1]. Recently, several factors have rekindled interest in searches for temporal variation of the fundamental constants. Foremost among these is the development of uni¿ed ¿eld theories (such as string theory and M theory) that allow for evolution in their coupling constants [2], [3]. Astrophysical data suggesting that such changes may have already occurred on the cosmological timescale [4] have also brought attention to the search. Finally, rapid progress development of in atomic frequency standards has recently made it possible to perform laboratory searches for present-day variation of fundamental constants [5].

Most searches for changes in fundamental constants have focused around the ¿ne-structure constant $\alpha = e^2/4\pi\varepsilon_0\hbar c$. The experiments in this area can be broken into two categories: historical tests based upon events in the distant past, and laboratory tests performed in the present. Limits to variation of the major constants from both types of experiments have recently been reviewed [6], [7].

One important historical test has been the spectroscopic examination of distant quasars and optical absorption in intervening dust clouds. Thiese data have been interpreted to show a change in α of $\Delta\alpha/\alpha = (-0.57 \pm 0.10)\times10^{-5}$ over the cosmological timescale (10^{10} yr) [4], [8]. A second test has been the examination of isotope ratios in the naturally occurring Oklo nuclear reactors [9], [10]. This analysis places a stringent limit to the possible variation of α over the geological timescale (10^9 yr), and if interpolated linearly, would constrain $|\dot{\alpha}/\alpha|$ to be less than 10^{-17} yr^{-1} [9]. Both of these results underscore the need for additional experimental work.

Historical results cannot tell us whether the natural constants are changing today, and so a separate avenue of research involves laboratory experiments. The repeated comparison of frequency standards based upon distinct atomic transitions provides one of the most promising settings for laboratory tests in this area due to the potentially high stability and accuracy of the standards. Here, we describe a comparison conducted over the course of two years between the frequency of an optical transition in a mercury ion and the ground-state hyper¿ne splitting of neutral cesium, which de¿nes the SI second. This comparison provides a new bound to a combination of fundamental constants.

II. Basis for comparison

The functional dependence of the mercury and cesium transitions on the relevant fundamental constants forms the theoretical basis for the frequency comparison. The optical transition is the $5d^{10}6s\ ^2S_{1/2}(F=0) \longleftrightarrow 5d^9 6s^2\ ^2D_{5/2}(F=2, m_F=0)$ electric-quadrupole transition at $\lambda = 282$ nm in ^{199}Hg$^+$, with frequency $\nu_{\mathrm{Hg}} \approx 1.06\times10^{15}$ Hz. Including relativistic and many-body effects, ν_{Hg} can be expressed as $\nu_{\mathrm{Hg}} \simeq R_\infty c\ F_{\mathrm{Hg}}(\alpha)$, where R_∞ is the Rydberg constant, and the relativistic Casimir factor $F_{\mathrm{Hg}}(\alpha)$ is a dimensionless function of the ¿ne-structure constant. Similarly, the frequency ν_{Cs} of the ground-state hyper¿ne transition $6S_{1/2}(F=3, m_F=0) \longleftrightarrow 6S_{1/2}(F=4, m_F=0)$ in neutral ^{133}Cs can be

approximated by $\nu_{\rm Cs} \simeq g_{\rm Cs}(m_e/m_p)\alpha^2 R_\infty c\ F_{\rm Cs}(\alpha)$, where $g_{\rm Cs}$ is the ^{133}Cs nuclear g-factor [7] and m_e/m_p is the ratio of the electron mass to the proton mass. The factors $F_{\rm Hg}(\alpha)$ and $F_{\rm Cs}(\alpha)$ can be calculated with suf¿cient precision to elucidate the constraints of the comparison [5], [11]. Using the results of one such calculation [11], we ¿nd that $\alpha \frac{\partial}{\partial \alpha} \ln F_{\rm Hg}(\alpha) \simeq -3.2$, and $\alpha \frac{\partial}{\partial \alpha} \ln F_{\rm Cs}(\alpha) \simeq +0.8$. Therefore, repeated comparisons of the mercury and cesium transition frequencies measure $\nu_{\rm Cs}/\nu_{\rm Hg}$, and provide a measure of the variation in the product of fundamental constants $U = g_{\rm Cs}(m_e/m_p)\alpha^{6.0}$.

The high sensitivity of this comparison to changes in the ¿ne-structure constant makes it attractive compared to other atomic-frequency ratios, especially if α alone is varying. However, there is little reason to assume that this is the case. It has been suggested within the framework of a uni¿ed theory that the variation of α is accompanied by an approximately 40-times-larger fractional change in the quantum chromodynamic scale parameter $\Lambda_{\rm QCD}$ [12]. The same fractional change should be expected in the proton mass, since $m_p \propto \Lambda_{\rm QCD}$. Furthermore, nuclear magnetic moments are also sensitive to changes in the quark masses m_q with respect to $\Lambda_{\rm QCD}$. Taking these factors into account, the more general dependence of the product U upon fundamental constants is estimated [13] to be $U = \alpha^{6.0}[m_e/\Lambda_{\rm QCD}][m_q/\Lambda_{\rm QCD}]^{0.1}$.

III. Experimental Methods

The mercury-ion optical-frequency clock has been described previously, and the reader is referred elsewhere [14]–[16] for more detailed information. A single $^{199}{\rm Hg}^+$ ion is con¿ned in a miniature spherical rf trap and laser-cooled to near the 1.7 mK Doppler limit on the $^2S_{1/2}(F=1) \longleftrightarrow {}^2P_{1/2}(F=0)$ transition at 194 nm. To probe the "clock" transition, the ion is ¿rst cooled and pumped into the $^2S_{1/2}(F=0)$ state. The ion is then exposed to the 282 nm probe light for a typical duration of 50 ms. Following this stage, the internal state of the ion is interrogated by using the method of electron shelving [17], [18]. Light from the cooling laser at 194 nm is directed onto the ion, and the absence or presence of photons scattered by the ion indicates that the ion has or has not been excited to the $^2D_{5/2}$ metastable state, which has a lifetime of approximately 90 ms. This experimental sequence is repeated, while stepping the probe light across the line center frequency. A digital servo loop then steers the average frequency of the probe laser light to the atomic resonance, with a time constant of $\tau_{\rm loop} \sim 15$ s.

The probe light on the 282 nm clock transition is the frequency-doubled output of a dye laser operating at 563 nm. The laser is stabilized to a resonance of a thermally and vibrationally isolated high-¿nesse ($\mathcal{F} \sim 200\ 000$) Fabry-Pérot cavity. After removing the predictable linear frequency drift of the cavity with an acousto-optic modulator, the light is frequency-doubled in a deuterated ammonium dihydrogen phosphate (AD*P) crystal and directed into the ion trap. The fractional frequency instability is 3×10^{-16} between 1 s and 10 s, corresponding to a 640 mHz linewidth at 563 nm [14]. For a 50 ms probe time, the observed linewidth is Fourier-transform-limited with a full width at half maximum of about 16 Hz, which is large compared to the ¡uctuations of the probe laser frequency during the probe time. The fractional frequency stability of the optical standard is then approximately $5 \times 10^{-15}\tau^{-1/2}$ [16].

Thus far, we have described an optical frequency standard. To compare the optical frequency with the cesium frequency standard, we must phase-coherently divide the optical frequency down to produce a microwave-frequency signal that can be compared to the cesium transition frequency. A portion of the stable 563 nm light locked to the atomic resonance is sent through an optical ¿ber 180 m long to a separate laboratory for the frequency comparison. An active noise-cancellation scheme is used to remove phase noise induced by ¡uctuations in the optical path length of the ¿ber [19], [20], and thereby preserve the coherence of the light. The frequency of the 563 nm light is measured with an octave-spanning optical frequency comb [21]–[24]. The comb is generated by the output of a femtosecond Ti:Sapphire laser, spectrally broadened in a microstructure ¿ber. The resulting spectrum consists of a series of equally spaced, phase-coherent modes with frequencies $f_n = nf_r + f_0$, where the repetition rate f_r of the mode-locked laser is near 1 GHz, n is an integer, and f_0 is an adjustable frequency offset that is common to all of the modes. The spectrum spans a wavelength range of roughly 520 nm to 1170 nm. The offset f_0 is measured with a heterodyne beatnote between a frequency-doubled mode from the infrared end of the spectrum $2f_n = 2nf_r + 2f_0$ and a mode from the blue end of the spectrum, $f_{2n} = 2nf_r + f_0$. A second beatnote f_b is measured between the 563 nm light and the nearest element of the optical comb, $f_m = 2mf_r + f_0$. By controlling the pump power and cavity length of the femtosecond laser, the two beatnote frequencies f_0 and f_b are phase-locked to be constants. Finally, the repetition rate f_r is detected on a fast photodiode and serves as the radio-frequency output of the optical clock system. The output stability of f_r is determined largely by the frequency stability of the 563 nm light, as the noise and inaccuracy added by the frequency comb system are negligible at the level of our comparison with the cesium frequency [25], [26].

The actual comparison with the cesium transition frequency is carried out through a reference hydrogen maser. The synthesizers and counters that control the experiment are referenced to the 5 MHz output signal of the maser, which has a typical fractional frequency instability near $2\times10^{-13}\tau^{-1/2}$ for measurement times 1 s $< \tau <$ 10^5 s. This maser is periodically calibrated by the NIST-F1 cesium fountain primary standard [27], as well as international cesium standards. The frequency of the reference maser is known at a given time to within four parts in 10^{15} with respect to the SI second.

The systematic frequency shifts of the mercury-ion optical clock have recently been discussed in detail [16]. The most signi¿cant sources of uncertainty are the second-order Zeeman shift of the clock transition and the electric quadrupole shift. The second-order Zeeman shift is signi¿cant because the clock has thus far been operated with a magnetic bias ¿eld and in an

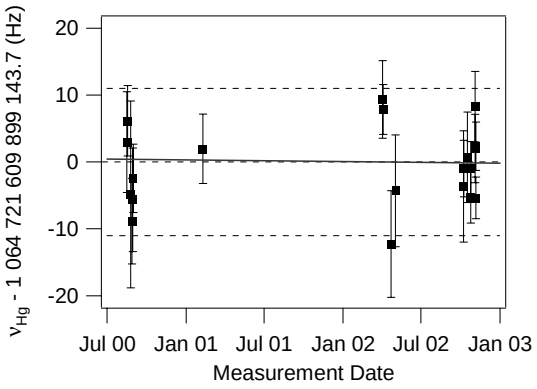

Fig. 1. Absolute frequency measurements of the $^{199}\mathrm{Hg}^{+}$ $^{2}S_{1/2}(F=0) \longleftrightarrow {}^{2}D_{5/2}(F=2)$ transition with respect to the ^{133}Cs ground state hyper¿ne splitting that de¿nes the SI second. The plot shows the deviation of each measurement from the weighted average value with its statistical $\pm 1\sigma$ error bar. The total systematic uncertainty is represented by the dashed lines at ± 11 Hz. The linear ¿t (solid line) has a slope of -0.24 ± 1.3 Hz yr $^{-1}$.

environment that has been prone to magnetic-¿eld ¡uctuations. The daily ¡uctuations in the ambient magnetic ¿eld lead to an uncertainty of 2.2 Hz on the clock frequency, and the uncertainty on the coef¿cient of the second-order Zeeman shift leads to an additional uncertainty of 2.6 Hz. The quadrupole shift arises from the interaction of stray electric-¿eld gradients in the ion trap with the atomic quadrupole moment in the $^{2}D_{5/2}$ excited state [28]. While we have not yet performed the evaluation of this shift, we expect that it will be below 1 Hz in magnitude. However, in the absence of the evaluation, we have placed conservative bounds of ± 10 Hz on the uncertainty in the shift. The total systematic (Type B [29]) uncertainty is ± 11 Hz, which is obtained by adding in quadrature the systematic uncertainties of the mercury clock and the hydrogen maser.

IV. Results and Conclusions

The results of twenty absolute measurements of the frequency of the Hg^{+} optical transition with respect to the cesium hyper¿ne splitting are shown in Fig. 1. These measurements, conducted over the course of two years, show reproducibility better than 10 Hz at 1.06×10^{15} Hz [16]. The weighted average of the frequency measurements give the absolute frequency $\nu_{\mathrm{Hg}} = 1\ 064\ 721\ 609\ 899\ 143.7\ (1.1)$ Hz, where the total statistical (Type A) uncertainty is only 1.0 Hz. A linear ¿t to the data set gives a slope of -0.24 ± 1.3 Hz yr^{-1}. Combining this result with the systematic fractional uncertainty of 10^{-14}, this measurement constrains the possible variation of $\nu_{\mathrm{Cs}}/\nu_{\mathrm{Hg}}$ at $\pm 7 \times 10^{-15}$ yr^{-1}. Equivalently, the product $U = g_{\mathrm{Cs}}(m_e/m_p)\alpha^{6.0}$ is constrained at the same level. If we assume that no factors besides α are varying, we can place a limit on the possible present-day linear variation in α, $|\dot{\alpha}/\alpha| < 1.2 \times 10^{-15}$ yr^{-1}, an improvement by a factor of 30 over [5].

The limits that can be set on the variation of fundamental constants are of course limited by the uncertainties of the clock system. It is hoped that in the near future we will be able to signi¿cantly lower our systematic uncertainties by completing the evaluation of the quadrupole shift and reducing the effects of stray magnetic ¿elds. Beyond simply improving the present tests, it is important to consider what other tests can be performed that will constrain the variation of different combinations of fundamental constants. The direct comparison of two optical frequency standards may test the stability of α alone. A promising example is the comparison between the mercury optical transition to the 657 nm $^{1}S_0(m=0) \longleftrightarrow {}^{3}P_1(m=0)$ transition in neutral ^{40}Ca [21], [30]. Other complementary work presently underway, such as the ongoing comparison between the hyper¿ne transitions in cesium and rubidium [31], [32], will test the stability of the strong and electroweak interactions and provide independent constraints to theory.

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