Search for Variations of Fundamental Constants using Atomic Fountain Clocks

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Over five years, we have compared the hyperfine frequencies of ^{133}Cs and ^{87}Rb atoms in their electronic ground state using several laser-cooled ^{133}Cs and ^{87}Rb atomic fountains with an accuracy of $\sim 10^{-15}$. These measurements set a stringent upper bound to a possible fractional time variation of the ratio between the two frequencies: $\frac{d}{dt}\ln[(\nu_{\mathrm{Rb}})/(\nu_{\mathrm{Cs}})] = (0.2 \pm 7.0) \times 10^{-16} \ \mathrm{yr}^{-1}$ (1σ uncertainty). The same limit applies to a possible variation of the quantity $(\mu_{\mathrm{Rb}}/\mu_{\mathrm{Cs}})\alpha^{-0.44}$, which involves the ratio of nuclear magnetic moments and the fine structure constant.

Since Dirac's 1937 formulation of his large number hypothesis aimed at tying together the fundamental constants of physics [1], a large amount of work has been devoted to test if these constants were indeed constant over time [2,3].

In general relativity and in all metric theories of gravitation, variations with time and space of nongravitational fundamental constants, such as the fine structure constant $\alpha = e^2/4\pi\epsilon_0\hbar c$, are forbidden. They would violate Einstein's Equivalence Principle (EEP). EEP imposes the local position invariance stating that, in a local freely falling reference frame, the result of any local nongravitational experiment is independent of where and when it is performed. On the other hand, almost all modern theories directed toward unifying gravitation with the three other fundamental interactions predict violation of EEP at levels which are within reach of near-future experiments [4,5]. As the internal energies of atoms or molecules depend on electromagnetic as well as strong and weak interactions, comparing the frequency of electronic transitions, fine structure transitions, and hyperfine transitions as a function of time or gravitational potential provides an interesting test of the validity of EEP.

Until now, very stringent tests exist on geological and cosmological time scales. The analysis of the Oklo nuclear reactor showed that, 2×10^9 years ago, α did not differ from the present value by more than 10^{-7} of its value [6]. Light emitted by distant quasars has been used to perform absorption spectroscopy of interstellar clouds. For instance, measurements of the wavelengths of molecular hydrogen transitions test a possible variation of the electron to proton mass ratio m_e/m_p [7]. Comparisons between the gross structure and the fine structure of neutral atoms and ions would indicate that α for a redshift $z \sim 1.5$ (~ 10 Gyr) differed from the present value: $\Delta\alpha/\alpha = (-7.2 \pm 1.8) \times 10^{-6}$ [8]. Today this is the only claim that fundamental constants might change.

On much shorter time scales, laboratory tests using frequency standards have been performed [9–11]. These tests have a very high sensitivity to changes in fundamental constants. They are repeatable, systematic errors can be tracked as experimental conditions can be changed.

In this Letter, we present results that place a new stringent limit to the time variation of fundamental constants. By comparing the hyperfine energies of ^{133}Cs and ^{87}Rb in their electronic ground state over a period of nearly five years, we place an upper limit to the rate of change of the ratio of the hyperfine frequencies $\nu_{\mathrm{Rb}}/\nu_{\mathrm{Cs}}$. Our measurements take advantage of the high accuracy ($\sim 10^{-15}$) of several laser-cooled Cs and Rb atomic fountains. According to recent atomic structure calculations [11,12], these measurements are sensitive to a possible variation of the quantity $(\mu_{\mathrm{Rb}}/\mu_{\mathrm{Cs}})\alpha^{-0.44}$, where μ are the nuclear magnetic moments. We anticipate major advances in these tests using frequency standards, thanks to recent advances in optical frequency metrology using femtosecond lasers [13,14].

In our experiments, three atomic fountains are compared to each other, using a hydrogen maser (H maser) as a flywheel oscillator (Fig. 1). Two fountains, a transportable fountain FOM and FO1 [15], are using cesium atoms. The third fountain is a dual fountain (DF) [16], operating alternately with rubidium (DF$_{\mathrm{Rb}}$) and cesium (DF$_{\mathrm{Cs}}$). These fountains have been continuously upgraded in order to improve their accuracy from 2×10^{-15} in 1998 to 8×10^{-16} for cesium and from 1.3×10^{-14} [17] to 6×10^{-16} for rubidium.

Fountain clocks operate as follows. First, atoms are collected and laser cooled in an optical molasses or in a magneto-optical trap in 0.3 to 0.6 s. Atoms are launched upwards, and selected in the clock level ($m_F = 0$) by a combination of microwave and laser pulses. Then, atoms interact twice with a microwave field tuned near the
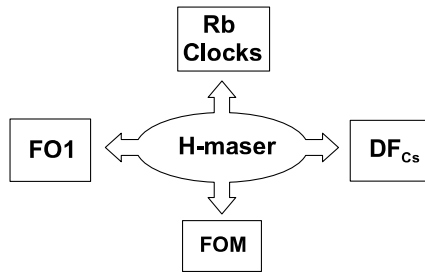
FIG. 1. BNM-SYRTE clock ensemble. A single 100 MHz signal from a H maser is used for frequency comparisons and is distributed to each of the microwave synthesizers of the ^{133}Cs (FO1, FOM, DFCs) and ^{87}Rb fountain clocks. In 2001, the Rb fountain had been upgraded and is now a dual fountain using alternately rubidium (DF$_{Rb}$) or cesium atoms (DF$_{Cs}$).

FIG. 2. The 2002 frequency comparison data. (a) H-maser fractional frequency offset versus FOM (■), and alternately versus DF$_{Rb}$ (○) and DF$_{Cs}$ (△ between dotted lines). (b) Fractional frequency differences. Between dotted lines, Cs-Cs comparisons, outside Rb-Cs comparisons. Error bars are purely statistical. They correspond to the Allan standard deviation of the comparisons and do not include contributions from fluctuations of systematic shifts of Table I. (MJD is Modified Julian Date. January 1, 2000 is MJD 51544.)

hyperfine frequency, in a Ramsey interrogation scheme. The microwave field is synthesized from a low phase noise 100 MHz signal from a quartz oscillator, which is phase locked to the reference signal of the H maser (Fig. 1). After the microwave interactions, the population of each hyperfine state is measured using light induced fluorescence. This provides a measurement of the transition probability as a function of microwave detuning. Successive measurements are used to steer the average microwave field to the frequency of the atomic resonance using a digital servo system. The output of the servo provides a direct measurement of the frequency difference between the H maser and the fountain clock.

The three fountains have different geometries and operating conditions: The number of detected atoms ranges from 3×10^5 to 2×10^6 at a temperature of $\sim 1~\mu$K, the fountain cycle duration from 1.1 to 1.6 s. The Ramsey resonance width is between 0.9 and 1.2 Hz. In measurements reported here, the fractional frequency instability is $(1-2) \times 10^{-13} \tau^{-1/2}$, where τ is the averaging time in seconds. Fountain comparisons have a typical resolution of $\sim 10^{-15}$ for a 12 h integration, and each of the four data campaigns lasts from one to two months during which an accuracy evaluation of each fountain is performed.

The 2002 measurements are presented in Fig. 2, which displays the maser fractional frequency offset, measured by the Cs fountains FOM and DF$_{Cs}$. Also shown is the H-maser frequency offset measured by the Rb fountain DF$_{Rb}$, where the Rb hyperfine frequency is conventionally chosen to be $\nu_{Rb}(1999) = 6\,834\,682\,610.904\,333$ Hz, our 1999 value. The data are corrected for the systematic frequency shifts listed in Table I. The H-maser frequency exhibits fractional fluctuations on the order of 10^{-14} over a few days, 10 times larger than the typical statistical uncertainty resulting from the instability of the fountain clocks. In order to reject the H-maser frequency fluctuations, the fountain data are recorded simultaneously (within a few minutes). The fractional frequency differences plotted in Fig. 2(b) illustrate the efficiency of this rejection. DF is operated alternately with Rb and Cs,
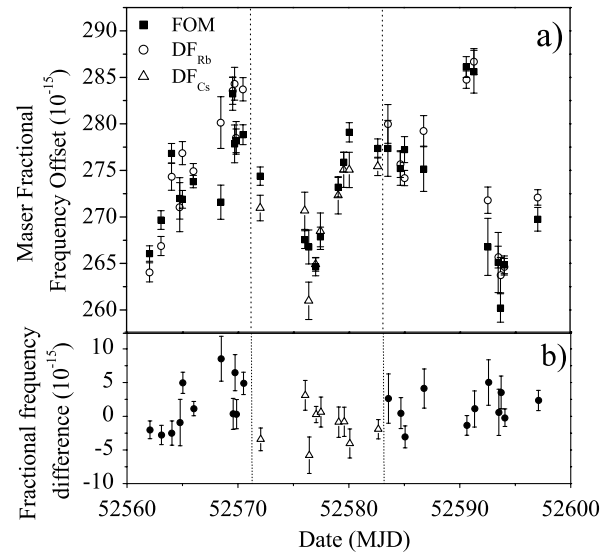
allowing both Rb-Cs comparisons and Cs-Cs comparisons (central part of Fig. 2) to be performed.

Systematic effects shifting the frequency of the fountain standards are listed in Table I. The quantization magnetic field in the interrogation region is determined with a 0.1 nT uncertainty by measuring the frequency of a linear field-dependent "Zeeman" transition. The temperature in the interrogation region is monitored with five platinum resistors and the uncertainty on the blackbody radiation frequency shift corresponds to temperature fluctuations of about 1 K [18]. Clock frequencies are corrected for the cold collision and cavity pulling frequency shifts using several methods [19,20]. For Rb, unlike [20], an optical molasses with a small number of atoms ($\sim 5.4 \times 10^6$) is used. We thus estimate that these two shifts are smaller than 5×10^{-17}. All other effects do not contribute significantly and their uncertainties are added quadratically. We searched for the influence of synchronous perturbations by changing the timing sequence and the atom launch height. To search for possible microwave leakage, we changed the power ($\times 9$) in the interrogation microwave cavity. No shift was found at a resolution of 10^{-15}. The shift due to residual coherences and populations in neighboring Zeeman states is estimated to be less than 10^{-16}. As shown in [21], the shift due to the microwave photon recoil is very similar for Cs and Rb and smaller than $+1.4 \times 10^{-16}$. Relativistic corrections (gravitational redshift and second order Doppler effect) contribute to less than 10^{-16} in the clock comparisons.

TABLE I.   Accuracy budget of the fountains involved in the 2002 measurements (DF and FOM).

Fountain	DF_{Cs}	DF_{Rb}	FOM
Effect	Value and uncertainty (10^{-16})		
Second order Zeeman	1773.0 ± 5.2	3207.0 ± 4.7	385.0 ± 2.9
Blackbody radiation	-173.0 ± 2.3	-127.0 ± 2.1	-186.0 ± 2.5
Cold collisions + cavity pulling	-95.0 ± 4.6	0.0 ± 1.0	-24.0 ± 4.8
Others	0.0 ± 3.0	0.0 ± 3.0	0.0 ± 3.7
Total uncertainty	8	6	8

For the Cs-Cs 2002 comparison, we find

$$\frac{\nu_{Cs}^{DF}(2002) - \nu_{Cs}^{FOM}(2002)}{\nu_{Cs}} = +12(6)(12) \times 10^{-16}, \quad (1)$$

where the first parentheses reflect the 1σ statistical uncertainty, and the second the systematic uncertainty, obtained by adding quadratically the inaccuracies of the two Cs clocks (see Table I). The two Cs fountains are in good agreement despite their significantly different operating conditions (see Table I), showing that systematic effects are well understood at the 10^{-15} level.

In 2002, the ^{87}Rb frequency measured with respect to the average ^{133}Cs frequency is found to be

$$\nu_{Rb}(2002) = 6\,834\,682\,610.904\,324(4)(7) \text{ Hz} \quad (2)$$

where the error bars now include DF_{Rb}, DF_{Cs}, and FOM uncertainties. This is the most accurate frequency measurement until now.

In Fig. 3 are plotted all our Rb-Cs frequency comparisons. Except for the less precise 1998 data [17], two Cs fountains were used together to perform the Rb measurements. The uncertainties for the 1999 and 2000 measurements were 2.7×10^{-15}, because of lower clock accuracy and lack of rigorous simultaneity in the earlier frequency comparisons [16]. A weighted linear fit to the data in Fig. 3 determines how our measurements constrain a possible time variation of ν_{Rb}/ν_{Cs}. We find

$$\frac{d}{dt} \ln\left(\frac{\nu_{Rb}}{\nu_{Cs}}\right) = (0.2 \pm 7.0) \times 10^{-16} \text{ yr}^{-1}, \quad (3)$$

which represents a fivefold improvement over our previous results [16] and a 100-fold improvement over the Hg^+-H hyperfine energy comparison [11].

We now examine how this result constrains possible variations of fundamental constants. For an alkali with atom number Z, the hyperfine transition frequency can be approximated by

$$\nu \propto \alpha^2 \frac{\mu}{\mu_N} \left(\frac{m_e}{m_p}\right) R_\infty c F_{rel}(Z\alpha), \quad (4)$$

where R_∞ is the Rydberg constant, c the speed of light, μ the magnetic moment of the nucleus, and μ_N the nuclear magneton. $F_{rel}(Z\alpha)$ is a relativistic function which strongly increases with Z [11,22]. For ^{133}Cs, this Casi-

mir relativistic contribution amounts to 40% of the hyperfine splitting and $\alpha(\{\partial \ln[F_{rel}(Z\alpha)]\}/(\partial\alpha)) = 0.74$. For ^{87}Rb, this quantity is 0.30 [23]. Following [11] and neglecting possible changes of the strong and weak interactions affecting μ_{Rb} and μ_{Cs}, the sensitivity of the ratio ν_{Rb}/ν_{Cs} to a variation of α is simply given by

$$\frac{\partial}{\partial \ln\alpha} \ln\left(\frac{\nu_{Rb}}{\nu_{Cs}}\right) \simeq (0.30\text{–}0.74) = -0.44. \quad (5)$$

Using Eqs. (3) and (5), we thus set the new limit: $\dot\alpha/\alpha = (-0.4 \pm 16) \times 10^{-16} \text{ yr}^{-1}$.

In contrast with [11], Ref. [22] argues that a time variation of the nuclear magnetic moments must also be considered in a comparison between hyperfine frequencies. The magnetic moments μ can be calculated using the Schmidt model. For atoms with odd A and Z such as ^{87}Rb and ^{133}Cs, the Schmidt magnetic moment $\mu^{(s)}$ is found to depend only on g_p, the proton gyromagnetic ratio. With this simple model, Ref. [22] finds

$$\frac{\partial}{\partial \ln g_p} \ln\left(\frac{\nu_{Rb}}{\nu_{Cs}}\right) \simeq \frac{\partial}{\partial \ln g_p} \ln\left(\frac{\mu_{Rb}^{(s)}}{\mu_{Cs}^{(s)}}\right) \simeq 2.0. \quad (6)$$

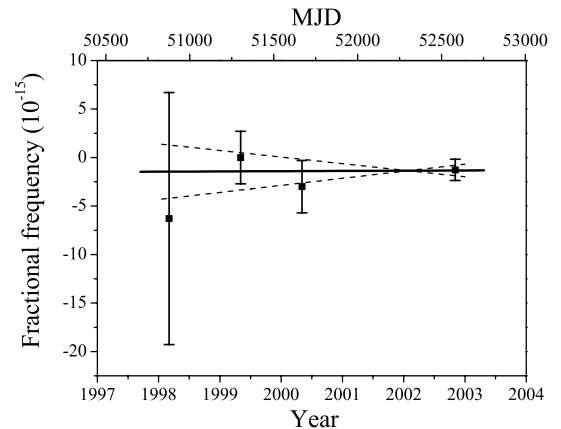Attributing any variation of ν_{Rb}/ν_{Cs} to a variation of

FIG. 3.   Measured ^{87}Rb frequencies referenced to the ^{133}Cs fountains over 57 months. The 1999 measurement value $[\nu_{Rb}(1999) = 6\,834\,682\,610.904\,333$ Hz] is conventionally used as reference. A weighted linear fit to the data gives $\frac{d}{dt}\ln[(\nu_{Rb})/(\nu_{Cs})] = (0.2 \pm 7.0) \times 10^{-16} \text{ yr}^{-1}$. Dotted lines correspond to the 1σ slope uncertainty.

g_p, Eqs. (3) and (6) lead to $\dot{g_p}/g_p = (0.1 \pm 3.5) \times 10^{-16}$ yr^{-1}. However, it must be noted that the Schimdt model is oversimplified and does not agree very accurately with the actual magnetic moment.

Moreover, attributing all the time variation of ν_{Rb}/ν_{Cs} to either g_p or α independently is somewhat artificial. Theoretical models allowing for a variation of α also allow for variations in the strength of the strong and electroweak interactions. For instance, Ref. [5] argues that Grand unification of the three interactions implies that a time variation of α necessarily comes with a time variation of the coupling constants of the other interactions. Reference [5] predicts that a fractional variation of α is accompanied with a ~40 times larger fractional change of m_e/m_p. In order to independently test the stability of the three fundamental interactions, several comparisons between different atomic species and/or transitions are required. For instance and as illustrated in [14], absolute frequency measurements of an optical transition are sensitive to a different combination of fundamental constants: $(\mu_{Cs}/\mu_N)(m_e/m_p)\alpha^x$, where x depends on the particular atom and/or transition.

A more complete theoretical analysis going beyond the Schmidt model would clearly be very useful to interpret frequency comparisons involving hyperfine transitions. This is especially important as most precise frequency measurements, both in the microwave and the optical domain [14,24,25], are currently referenced to the ^{133}Cs hyperfine splitting, the basis of the SI definition of the second. The H hyperfine splitting, which is calculable to a high accuracy, had already been considered as a possible reference several decades ago. Unfortunately, despite numerous efforts, the H hyperfine splitting is currently measured to only seven parts in 10^{13} (using H masers), almost 3 orders of magnitude worse than the results presented in this Letter.

In summary, by comparing ^{133}Cs and ^{87}Rb hyperfine energies, we have set a stringent upper limit to a possible fractional variation of the quantity $(\mu_{Rb}/\mu_{Cs})\alpha^{-0.44}$ at $(-0.2 \pm 7.0) \times 10^{-16}$ yr^{-1}. In the near future, accuracies near one part in 10^{16} should be achievable in microwave atomic fountains, improving our present Rb-Cs comparison by 1 order of magnitude.

We anticipate that comparisons between rapidly progressing optical and microwave laser-cooled frequency standards currently developed in several laboratories will bring orders of magnitude gain in sensitivity. In order to have the full benefit of these advances, frequency comparisons with improved accuracy between these distant clocks will be necessary. Serving this purpose, a new generation of time/frequency transfer at the 10^{-16} level is currently under development for the European Space Agency space mission ACES which will fly ultrastable clocks on board the international space station in 2006 [26]. These comparisons will also allow for a search of a possible change of fundamental constants induced by the annual modulation of the Sun gravitational potential due to the elliptical orbit of Earth [27].

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