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Recommended Citation

"New Limits on the Drift of Fundamental Constants from Laboratory Measurements," *Physical Review Letters*, vol. 92, no. 23, American Physical Society (APS), Jun 2004.

The definitive version is available at <https://doi.org/10.1103/PhysRevLett.92.230802>

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New Limits on the Drift of Fundamental Constants from Laboratory Measurements

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(Received 3 December 2003; published 10 June 2004)

We have remeasured the absolute $1S$ - $2S$ transition frequency ν_H in atomic hydrogen. A comparison with the result of the previous measurement performed in 1999 sets a limit of (-29 ± 57) Hz for the drift of ν_H with respect to the ground state hyperfine splitting ν_{Cs} in ^{133}Cs . Combining this result with the recently published optical transition frequency in $^{199}\text{Hg}^+$ against ν_{Cs} and a microwave ^{87}Rb and ^{133}Cs clock comparison, we deduce separate limits on $\dot{\alpha}/\alpha = (-0.9 \pm 2.9) \times 10^{-15} \text{ yr}^{-1}$ and the fractional time variation of the ratio of Rb and Cs nuclear magnetic moments $\mu_{\text{Rb}}/\mu_{\text{Cs}}$ equal to $(-0.5 \pm 1.7) \times 10^{-15} \text{ yr}^{-1}$. The latter provides information on the temporal behavior of the constant of strong interaction.

DOI: 10.1103/PhysRevLett.92.230802

PACS numbers: 06.30.Ft, 06.20.Jr, 32.30.Jc

In the era of a rapid development of precision experimental methods, the stability of fundamental constants becomes a question of basic interest. Any drift of non-gravitational constants is forbidden in all metric theories of gravity including general relativity. The basis of these theories is Einstein's Equivalence Principle (EEP) which states that weight is proportional to mass, and that in any local freely falling reference frame, the result of any nongravitational experiment must be independent of time and space. This hypothesis can be proven only experimentally as no theory predicting the values of fundamental constants exists. In contrast to metric theories, string theory models aiming to unify quantum mechanics and gravitation allow for, or even predict, violations of EEP. Limits on the variation of fundamental constants might therefore provide important constraints on these new theoretical models.

A recent analysis of quasar absorption spectra with redshifted UV transition lines indicates a variation of the fine structure constant $\alpha = e^2/4\pi\epsilon_0\hbar c$ on the level of $\Delta\alpha/\alpha = (-0.54 \pm 0.12) \times 10^{-5}$ for a redshift range $(0.2 < z < 3.7)$ [1]. On geological time scales, a limit for the drift of α has been deduced from isotope abundance ratios in the natural fission reactor of Oklo, Gabon, which operated about 2 Gyr ago. Modeling the processes which have changed the isotope ratios of heavy elements gives a limit of $\Delta\alpha/\alpha = (-0.36 \pm 1.44) \times 10^{-8}$ [2]. In these measurements, the high sensitivity to the time variation of α is achieved through very long observation times at moderate resolution for $\Delta\alpha$. Therefore, they are vulnerable to systematic effects [3].

Laboratory experiments can reach a 10^{-15} accuracy within years with better controlled systematics. This type of experiment is typically based on repeated absolute frequency measurements, i.e., comparison of a transition frequency with the reference frequency of the ground state hyperfine transition in ^{133}Cs . For an optical transition, the theoretical expression for the drift of its absolute frequency inevitably involves $\mu_{\text{Cs}}/\mu_{\text{B}}$ in addition to α , where μ_{Cs} is the magnetic moment of the Cs nucleus and μ_{B} is the Bohr magneton [4]. The microwave Rb and Cs clock comparison [5] even involves two nuclear moments. The magnitude of nuclear moments results from both the electromagnetic and the strong interaction.

Contributions from weak, electromagnetic, and strong interactions can be disentangled by combining several frequency measurements possessing a different sensitivity to the fundamental constants. In this Letter, we deduce separate stringent limits for the drifts of the fine structure constant α , $\mu_{\text{Cs}}/\mu_{\text{B}}$, and $\mu_{\text{Rb}}/\mu_{\text{Cs}}$ from combining the drifts of two optical frequencies in hydrogen and in the mercury ion with respect to the ground state hyperfine splitting in ^{133}Cs and the result of a microwave clock comparison [5]. Comparing measurements performed at different places and at different times we have to use the Lorentz and position invariance and we have to assume that the constants change linearly and do not oscillate on a year scale. With the exception of this, our results are independent of further assumptions about a particular drift model and possible correlated drifts of different constants [6].

The experiments on the drift of the $^{199}\text{Hg} + 5d^{10}6s^2S_{1/2}(F=0) \rightarrow 5d^96s^2D_{5/2}(F'=2)$ electric quadrupole (“clock”) transition frequency ν_{Hg} were performed by the group of Bergquist at NIST between July 2000 and December 2002 [7]. These repeated measurements of ν_{Hg} limit the fractional time variation of the ratio $\nu_{\text{Cs}}/\nu_{\text{Hg}}$ to $(0.2 \pm 7) \times 10^{-15} \text{ yr}^{-1}$.

In 1999 [8] and 2003, we have phase coherently compared the frequency of the $(1S, F=1, m_F = \pm 1) \rightarrow (2S, F'=1, m'_F = \pm 1)$ two-photon transition in atomic hydrogen to the frequency of the ground state hyperfine splitting of ^{133}Cs using a frequency comb technique [9]. The 1999 setup of the hydrogen spectrometer has been described previously in [10], so we present only a brief description with an emphasis on the improvements since 1999. A sketch of the actual setup is shown in Fig. 1.

A cw dye laser emitting near 486 nm is locked to an external reference cavity. The new cavity for the 2003 measurement is more stable and drifts less than 0.5 Hz/s at 486 nm. The linewidth of the dye laser stabilized on the new cavity has been characterized as 60 Hz for an averaging time of 1 s. A small part of the laser light is transferred to a neighboring lab where its absolute frequency is measured. The main part is frequency doubled, and the resulting radiation near 243 nm (corresponding to half of the 1S-2S transition frequency) is coupled into a linear enhancement cavity inside the vacuum chamber of the hydrogen spectrometer.

Hydrogen atoms from a radio-frequency (rf) gas discharge are cooled to 5–6 K by collisions with the walls of

a copper nozzle. The nozzle forms a beam of cold atomic hydrogen which leaves the nozzle collinearly with the cavity axis and enters the interaction region between the nozzle and the detector shielded from stray electric fields by a Faraday cage. Some of the atoms are excited from the ground state to the metastable 2S state by Doppler-free absorption of two counterpropagating photons from the laser field in the enhancement cavity. The 1999 measurement was performed at a background gas pressure of around 10^{-6} mbar in the interaction region. In the meantime, we upgraded the vacuum system with a differential pumping configuration. This allows us to vary the background gas pressure in a range of 10^{-8} – 10^{-7} mbar and to reduce the background gas pressure shift and the corresponding uncertainty to 2 Hz.

Because of small apertures, only atoms flying close to the cavity axis can enter the detection region where the 2S atoms are quenched in a small electric field and emit L_{α} photons. The excitation light and the hydrogen beam are periodically blocked by two mutually phase locked choppers, and the L_{α} photons are counted time-resolved only in the dark part of a cycle. The delay τ between blocking the 243 nm radiation and the start of photon counting sets the upper limit for the atomic velocity of $v < l/\tau$, where l is the distance between nozzle and detector. With the help of a multichannel scaler, we count all photons and sort them into 12 adjacent time bins. From each scan of the laser frequency over the hydrogen 1S-2S resonance we therefore get up to 12 spectra measured with different delays. To correct for the second order Doppler shift, we use an elaborate theoretical model to fit all the delayed spectra of one scan simultaneously with a set of seven fit parameters [10]. The result of the fitting procedure is the 1S-2S transition frequency for a hydrogen atom at rest.

The transition frequency depends linearly on the excitation light intensity due to the dynamic ac-Stark shift. We vary the intensity and extrapolate the transition frequency to zero intensity to correct for this effect [8].

For an absolute measurement of the 1S-2S transition frequency, the dye laser frequency near 616.5 THz is phase coherently compared with an atomic cesium fountain clock. We take advantage of the frequency comb technique recently developed in Garching and Boulder [9] to bridge the large gap between the optical and radio-frequency domains. A mode-locked femtosecond (fs) laser emits a pulse train which equals a comb of laser modes in the frequency domain. The mode spacing corresponds to the repetition rate frequency f_{rep} of the fs laser, and the frequency of each mode can be written as $f_n = nf_{\text{rep}} + f_0$, where n is a large integer number and $f_0 < f_{\text{rep}}$ is the comb offset frequency. The repetition rate f_{rep} can easily be detected with a photodiode. For combs spanning more than one octave, f_0 can be determined by frequency doubling an infrared mode f_n in a nonlinear crystal to $2f_n$ and measuring the beat note with the appropriate

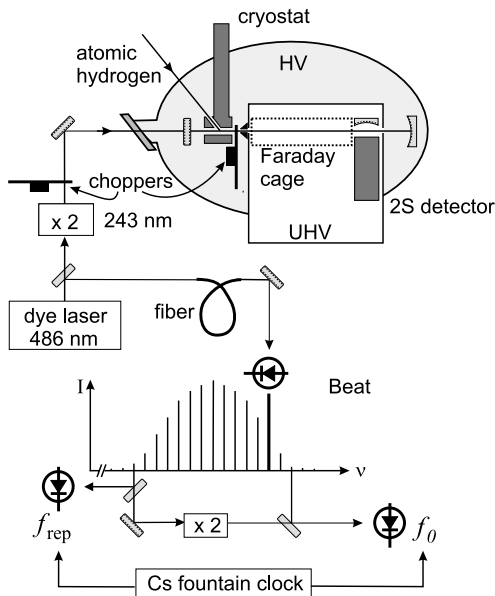


FIG. 1. Simplified experimental setup for comparison of the hydrogen 1S-2S transition frequency with a primary frequency standard (HV, high vacuum; UHV, ultrahigh vacuum). A mode-locked femtosecond laser with repetition rate f_{rep} emits the frequency comb which is broadened in a photonic crystal fiber. f_0 is the carrier-envelope offset frequency.

mode f_{2n} of the blue part of the comb, yielding $f_0 = 2f_n - f_{2n}$.

In our experiment, the frequency comb from a mode-locked fs Ti:sapphire laser with a repetition rate f_{rep} of 800 MHz is broadened in a photonic crystal fiber to cover more than an optical octave. We phase lock both degrees of freedom of the comb (f_{rep}, f_0) to the radio-frequency output of the Cs fountain clock to get a frequency comb with optical modes whose frequencies are known with the accuracy of the primary frequency standard. Any of these modes are available for optical measurements.

For both the 1999 and 2003 measurements, the transportable Cs fountain clock FOM has been installed at MPQ. Its stability is $1.8 \times 10^{-13} \tau_{\text{ave}}^{-1/2}$ for an averaging time of τ_{ave} , and its accuracy has been evaluated to 8×10^{-16} [11] at BNM-SYRTE. During the experiments in Garching, only a verification at the level of 10^{-15} has been performed. Consequently we attribute a conservative FOM accuracy of 2×10^{-15} for these measurements.

We have measured the $1S$ - $2S$ transition in atomic hydrogen during 10 d in 1999 and during 12 d in 2003. For comparability, both data sets have been analyzed using the same theoretical line-shape model [10]. In Fig. 2, the results of the extrapolation to zero excitation light intensity and the respective statistical error bars for each day are presented. The statistical uncertainty was significantly reduced compared to the 1999 measurements due to the narrower laser linewidth and a better signal-to-noise ratio, but the scatter of the day averages did not reduce accordingly. We ascribe this effect to a residual uncompensated first-order Doppler shift. Later measurements performed without the fountain clock with a deliberately introduced asymmetry in the 243 nm cavity indicate an adjustment-dependent frequency shift (the elimination of this additional systematic shift should be of high concern in future measurements). The effect should average out after multiple readjustments of the spectrometer which have been typically performed twice a day. It is impossible to correct the data *a posteriori* because such details of the spectrometer adjustment were not recorded during the phase-coherent measurement.

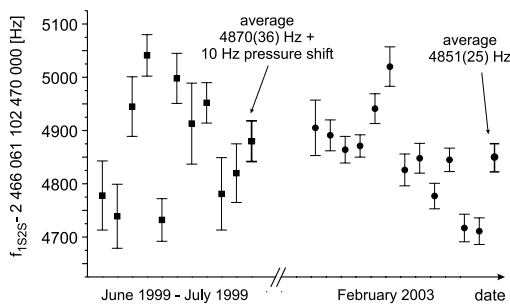


FIG. 2. Experimental results and averages for the 1999 and 2003 measurements of the absolute ($1S, F=1, m_F=\pm 1$) \rightarrow ($2S, F'=1, m'_F=\pm 1$) transition frequency in atomic hydrogen. The background pressure shift of +10 Hz as explained in [8] is added to the 1999 value.

Other effects which can cause the systematic shift (intra-beam pressure shift, background gas pressure shift, Stark shift of the hyperfine levels induced by the rf gas discharge, stray electric fields) have been checked and can be excluded on the level of the observed scatter.

The 1999 and 2003 d -dependent data were averaged without weighting [12]. After accounting for the total systematic uncertainties of 28 Hz (1999) and 23 Hz (2003) in the mutually equivalent evaluation processes of both measurements we deduce a difference equal to (-29 ± 57) Hz over a time interval of 44 months. This is equivalent to a fractional time variation of the ratio $\nu_{\text{Cs}}/\nu_{\text{H}}$ equal to $(3.2 \pm 6.3) \times 10^{-15} \text{ yr}^{-1}$.

The frequency of any optical transition can be written as $\nu = A \text{Ry} F_{\text{rel}}(\alpha)$ [4,13], where Ry is the Rydberg energy and the relativistic correction $F_{\text{rel}}(\alpha)$ takes into account relativistic and many-body effects. F_{rel} depends on the transition in the system considered and embodies the dependence on α , while the parameter A is independent of fundamental constants. For absolute frequency measurements Ry always cancels out.

Numerical calculations of the dependence of $F_{\text{rel,Hg}}(\alpha)$ for ν_{Hg} on the fine structure constant α yield [13]

$$\alpha \frac{\partial}{\partial \alpha} \ln F_{\text{rel,Hg}}(\alpha) \approx \frac{\alpha}{\Delta \alpha} \frac{\Delta F_{\text{rel}}}{F_{\text{rel}}} = -3.2. \quad (1)$$

The corresponding expression for ν_{H} gives a very weak dependence on α :

$$\alpha \frac{\partial}{\partial \alpha} \ln F_{\text{rel,H}}(\alpha) \approx 0. \quad (2)$$

The frequency of the ground state hyperfine transition in ^{133}Cs is given by

$$\nu_{\text{Cs}} = A' \text{Ry} \alpha^2 \frac{\mu_{\text{Cs}}}{\mu_{\text{B}}} F_{\text{rel,Cs}}(\alpha). \quad (3)$$

Following [13], the relativistic correction $F_{\text{rel,Cs}}(\alpha)$ is

$$\alpha \frac{\partial}{\partial \alpha} \ln F_{\text{rel,Cs}}(\alpha) \approx +0.8. \quad (4)$$

Therefore, the comparison of the clock transition in Hg^+ against a primary frequency standard tests a drift of [7]

$$\frac{\partial}{\partial t} \ln \frac{\nu_{\text{Cs}}}{\nu_{\text{Hg}}} = \frac{\partial}{\partial t} \left(\ln \frac{\mu_{\text{Cs}}}{\mu_{\text{B}}} + (2.0 + 0.8 + 3.2) \ln \alpha \right), \quad (5)$$

whereas the $1S$ - $2S$ experiment tests the drift of

$$\frac{\partial}{\partial t} \ln \frac{\nu_{\text{Cs}}}{\nu_{\text{H}}} = \frac{\partial}{\partial t} \left(\ln \frac{\mu_{\text{Cs}}}{\mu_{\text{B}}} + (2.0 + 0.8) \ln \alpha \right). \quad (6)$$

For clarity, we set $x \equiv (\partial/\partial t) \ln \alpha$, $y \equiv (\partial/\partial t) \times \ln(\mu_{\text{Cs}}/\mu_{\text{B}})$ and get as experimental constraints

$$y + 6x = (0.2 \pm 7) \times 10^{-15} \text{ yr}^{-1} (\text{Hg}^+), \quad (7a)$$

$$y + 2.8x = (3.2 \pm 6.3) \times 10^{-15} \text{ yr}^{-1} (\text{H}). \quad (7b)$$

Solving for x and y yields the separate restrictions for the drifts of α and $\mu_{\text{Cs}}/\mu_{\text{B}}$ without any assumptions of conceivable mutual correlations (see, e.g., [6]). In this

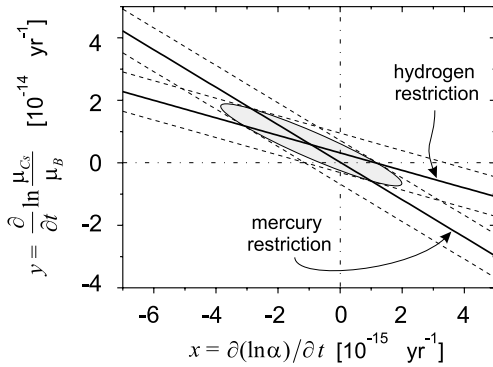


FIG. 3. Possible values for the relative drift rates x and y and their experimental restrictions (dashed lines) as stated by Eqs. (7). The probability density for x and y to be the true values is given by $P(\Delta x, \Delta y) \propto \exp[-R(\Delta x, \Delta y)/2]$ with the distances from the crossing point Δx and Δy and $R(\Delta x, \Delta y) = (\Delta y + 6\Delta x)^2/\sigma_{\text{Hg}}^2 + (\Delta y + 2.8\Delta x)^2/\sigma_{\text{H}}^2$. The resulting elliptical uncertainty region, defined by $R(\Delta x, \Delta y) = 1$, gives the standard deviation when projected on either axis by integration over the other. The uncertainties of the measured values are $\sigma_{\text{H}} = 6.3 \times 10^{-15} \text{ yr}^{-1}$ and $\sigma_{\text{Hg}} = 7 \times 10^{-15} \text{ yr}^{-1}$.

sense, this evaluation is model independent. Figure 3 represents both equations and their solution graphically.

For the relative drift of the fine structure constant at the end of the second millennium, we deduce the limit of

$$x = \frac{\partial}{\partial t} \ln \alpha = (-0.9 \pm 2.9) \times 10^{-15} \text{ yr}^{-1}. \quad (8)$$

The limit on the relative drift of $\mu_{\text{Cs}}/\mu_{\text{B}}$ is

$$y = \frac{\partial}{\partial t} \ln \frac{\mu_{\text{Cs}}}{\mu_{\text{B}}} = (0.6 \pm 1.3) \times 10^{-14} \text{ yr}^{-1}. \quad (9)$$

We deduce the uncertainties in expressions (8) and (9) as projections of the ellipse (Fig. 3) on the corresponding axes. This is equivalent to performing Gaussian propagation of uncertainties when resolving Eqs. (7) for x and y . Here, the measurement results for Hg^+ and H are treated as uncorrelated even though the drift rates x and y may be correlated [6]. The given 1σ uncertainties for x and y in Eqs. (8) and (9) incorporate both the statistical and systematic uncertainties of the hydrogen and the mercury measurements. Both limits (8) and (9) are consistent with zero. Meanwhile Peik and co-workers have added a precise drift measurement on a single trapped Yb ion to push the overall limit even further [14].

These results allow us to deduce a restriction for the relative drift of the nuclear magnetic moments in ^{87}Rb and ^{133}Cs . From 1998 to 2003, the drift of the ratio of the ground state hyperfine frequencies in ^{87}Rb and ^{133}Cs has been measured to be [5]

$$\frac{\partial}{\partial t} \ln \frac{\nu_{\text{Rb}}}{\nu_{\text{Cs}}} = (0.2 \pm 7.0) \times 10^{-16} \text{ yr}^{-1}. \quad (10)$$

Substituting the corresponding dependencies $F_{\text{rel}}(\alpha)$ for these transitions [5,13] yields

$$\frac{\partial}{\partial t} \ln \frac{\nu_{\text{Rb}}}{\nu_{\text{Cs}}} = \frac{\partial}{\partial t} \left(\ln \frac{\mu_{\text{Rb}}}{\mu_{\text{Cs}}} - 0.53 \ln \alpha \right). \quad (11)$$

Combining (8), (10), and (11) we get

$$\frac{\partial}{\partial t} \ln \frac{\mu_{\text{Rb}}}{\mu_{\text{Cs}}} = (-0.5 \pm 1.7) \times 10^{-15} \text{ yr}^{-1}, \quad (12)$$

where the same procedure as in Fig. 3 was used with a diagram covering x and $z \equiv (\partial/\partial t) \ln(\mu_{\text{Rb}}/\mu_{\text{Cs}})$.

In conclusion, we have determined separate limits for the drift of α , $\mu_{\text{Cs}}/\mu_{\text{B}}$ and $\mu_{\text{Rb}}/\mu_{\text{Cs}}$ from laboratory experiments without assumptions of conceivable correlations among them. All these limits are consistent with zero. Quasar absorption spectra measured with the Keck/HIRES spectrograph show a significant deviation between the values of α today and 10 Gyr ago [1]. A corresponding linear drift of α is smaller than the uncertainty of our result, therefore it cannot be excluded.

We thank S. G. Karshenboim for fruitful discussions of this work. N. K. acknowledges support from the AvH Stiftung. The work was partly supported by DFG (Grant No. 436RUS113/769/0-1) and RFBR. The development of the FOM fountain was supported by Centre National d'études spatiales and Bureau National de Météologie.

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