Precision Measurement of the Hydrogen 1S-2S Frequency via a 920-km Fiber Link

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We have measured the frequency of the extremely narrow 1S-2S two-photon transition in atomic hydrogen using a remote cesium fountain clock with the help of a 920 km stabilized optical fiber. With an improved detection method we obtain $f_{1S-2S} = 2\,466\,061\,413\,187\,018\,(11)$ Hz with a relative uncertainty of 4.5×10^{-15}, confirming our previous measurement obtained with a local cesium clock [C. G. Parthey *et al.*, Phys. Rev. Lett. **107**, 203001 (2011)]. Combining these results with older measurements, we constrain the linear combinations of Lorentz boost symmetry violation parameters $c_{(TX)} = (3.1 \pm 1.9) \times 10^{-11}$ and $0.92c_{(TY)} + 0.40c_{(TZ)} = (2.6 \pm 5.3) \times 10^{-11}$ in the standard model extension framework [D. Colladay, V. A. Kostelecký, Phys. Rev. D **58**, 116002 (1998)].

Atomic hydrogen has played a central role in the history of quantum mechanics through ever-refined comparisons between experimental data and theoretical predictions. As the most simple atomic system it allowed these comparisons to reach an unprecedented accuracy, confirming quantum mechanics as the fundamental microscopic description of nature. However, recent spectroscopic investigations of muonic hydrogen [1] have resulted in inconsistencies between theoretical predictions and measured transition frequencies. To learn more about possible causes of this discrepancy it is pivotal, among other things, to further reduce experimental uncertainties.

For the last two decades, our group at the Max-Planck-Institut für Quantenoptik (MPQ) at Garching, Germany has been measuring the frequency of the narrow 1S-2S transition in atomic hydrogen with ever improving accuracy. This transition frequency is providing critical input data to the CODATA adjustment of fundamental constants [2]. The development of the frequency comb [3] has significantly simplified optical frequency measurements by providing a direct and phase coherent link to the radio frequency domain where the cesium atomic clock operates. In 1999 the accuracy of hydrogen spectroscopy surpassed the accuracy of even the best available commercial cesium clocks [4]. Only much more accurate cesium fountain clocks could provide the reference for further improvements. Just like the hydrogen spectrometer, these clocks are not readily transportable. The only exception up to this day has been the mobile cesium fountain clock FOM [5] built by the Observatoire de Paris, France (LNE-SYRTE) that was used in two subsequent improvements [6,7].

The recent implementation of a 920 km optical fiber link between Physikalisch-Technische Bundesanstalt (PTB) at Braunschweig, Germany and MPQ now allows us to compare the hydrogen 1S-2S transition frequency to the stationary cesium fountain clock CSF1 [8,9] at PTB. The link allows us to compare the two clocks directly without limiting their accuracy at any integration time [10]. In this Letter, we report on the first successful measurement of the hydrogen 1S-2S transition frequency against a remote clock. We further derive new constraints for the Lorentz boost symmetry violating parameters in the standard model extension framework [11,12].
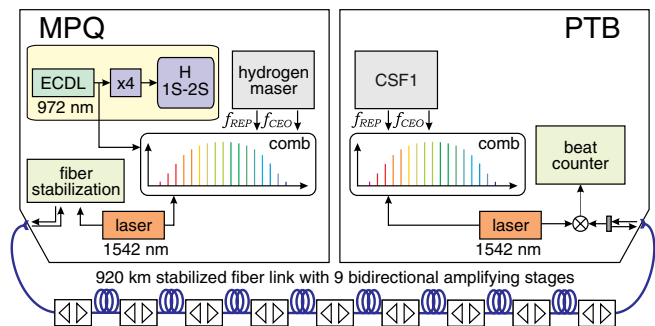

FIG. 1 (color online). The experimental setup. The frequency of the external cavity diode laser (ECDL) for hydrogen spectroscopy is measured using a frequency comb which is referenced to an active hydrogen maser. The maser is calibrated against the cesium fountain clock CSF1 via a 920 km long actively stabilized fiber link. $f_{\rm rep}$ denotes repetition rate and $f_{\rm CEO}$ the CEO frequency.

The experimental setup is sketched in Fig. 1. The 4th harmonic of an ultrastable 972 nm external cavity diode laser (ECDL) [13] is used to excite the 1S-2S transition. The frequency of this laser is referenced to an active hydrogen maser via a femtosecond erbium-doped fiber frequency comb. The maser is then calibrated using the cesium atomic fountain clock CSF1 located at PTB. For this purpose the radiation of an ultrastable erbium-doped continuous wave (cw) fiber laser at 1542 nm referenced to the local hydrogen maser is sent from MPQ to PTB via the 920 km optical fiber link. Intermediate bidirectional optical amplifiers and fiber Brillouin amplification at the end points compensate for the fiber loss of 230 dB. A fraction of the light is sent back to MPQ and heterodyned with the light launched into the fiber to measure and compensate optical path length variations. At PTB the frequency of the MPQ laser is measured with respect to CSF1 using a second frequency comb. In essence this compares the frequencies of the microwave sources, the maser at MPQ and CSF1 at PTB, while the frequencies of the transfer lasers cancel out in the comparison. The link, as characterized in [10], does not pose a limit to this comparison for the hydrogen measurement since its fractional frequency instability (modified Allan deviation) is $5 \times 10^{-15} (\tau/\mathrm{s})^{-3/2}$ up to averaging times $\tau = 100$ s. The differential gravitational redshift between the distant hydrogen and cesium clocks is obtained from GPS measurements combined with the EGM2008 geoid model; its uncertainty is estimated from the tidal effects.

The current hydrogen beam apparatus has been described before [7]. In brief, the two-photon 1S-2S transition is driven in a beam of cold 6 K hydrogen atoms that travel collinearly within a standing wave of the exciting laser to cancel the first order Doppler effect. A small electric field (10 V/cm) at the end of the atomic beam deexcites the 2S state at a well-defined point and forces the release of a 121 nm Lyman-α photon. Previously, these photons were detected with a photomultiplier [4,6,7]. For this measurement we instead detect photoelectrons created when the Lyman-α photons hit the graphite coating covering the 2S detector inner walls. The photoelectrons are collected by a channeltron whose front face is biased at +270 V. The atomic beam is shielded from this voltage by a second Faraday cage to maintain a well localized deexcitation point. With this detector we obtain more than an order of magnitude larger signal due to its solid acceptance angle of almost 4π. The corresponding improvement of the statistics of observed lines centers is shown in Fig. 2.

During five days from November 22 to 26 of 2010, the 1S(F = 1)-2S(F = 1) hyperfine line component has been scanned 622 times. The data evaluation to find the line centers follows that of Ref. [7]. To reduce the second order Doppler effect we periodically block the excitation light and record photons from the excited state with certain time delays. This sets an upper limit on the atomic velocities.
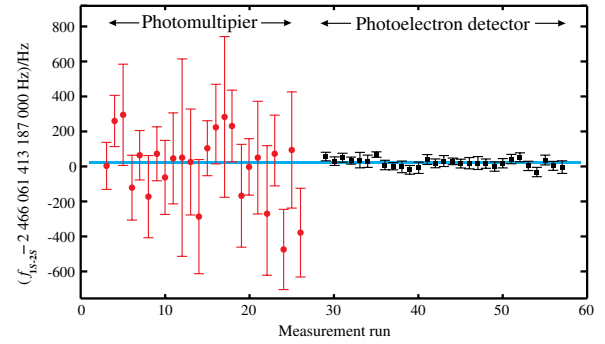

FIG. 2 (color online).   Comparison of the data scattering of the previous photomultiplier detector (red circles) with the photoelectron detector (black squares). Every point corresponds to one measurement run which typically consists of 7-10 line scans taken during ≈ 1 h. The blue line represents a weighted average.

The signal counts are sorted into time delay bins $\tau_1 = 10$–210 μs, $\tau_2 = 210$–410 μs, …, $\tau_8 = 1410$–1610 μs. We fit each hydrogen spectral line with a Lorentzian which is a good approximation of the line shape for delays $\tau_4 = 610$–810 μs and larger. The two dominating systematic effects are the ac Stark shift and the residual second order Doppler effect. A small quadratic contribution to the ac Stark shift is modelled and corrected with the help of a Monte Carlo simulation, that takes into account the full excitation geometry and the measured laser power. This leaves us with a purely linear ac Stark shift that we extrapolate to zero laser power. The remaining second order Doppler shift is corrected by using the measured velocity distribution of the 2S atoms (via the 2S-4P transition) in combination with Monte Carlo simulations that also takes into account a small laser power dependence [7].

After completion of the experiment we realized that the shielding factor of the Faraday cage protecting the atoms from the channeltron was insufficient. The additional field does not cause a significant dc Stark shift, since it is well separated from the excitation region by a 2.1 mm diam aperture. Most atoms contributing with delays larger than $\tau_3 = 410$–610 μs are excited at the first half of the excitation region at distances of a few centimeters from this aperture. However, this stray field increases the uncertainty about the location of the deexcitation point which translates into an additional uncertainty of the velocity distribution and hence the second order Doppler effect. We thus increased its contribution to the error budget from 5.1 to 8.0 Hz.

A summary of the contributing systematics can be found in Table I. It should be noted that the statistical uncertainty is smaller than in Ref. [7] due to the higher detection efficiency. Summing up all uncertainties in quadrature and taking into account frequency corrections [7] we find

$$f_{1S\text{-}2S} = 2\,466\,061\,413\,187\,018\,(11)\ \mathrm{Hz} \qquad (1)$$

for the hyperfine centroid computed with the same hyperfine constants as in the previous measurement [7]. For

TABLE I. Corrections and uncertainties for the hydrogen 1S-2S transition frequency. Corrections are given relative to the value obtained after statistical averaging $F_{stat} = 2466\,061\,102\,474\,893.1$ Hz. Δ denotes the correction and σ the uncertainty.

	Δ (Hz)	σ (Hz)
Statistics	0	3.3
Second order Doppler effect	+34.2	8.0
Line shape model	0	5.0
Quadratic ac Stark shift (243 nm)	−10.7	2.0
ac Stark shift, 486 nm quench beam	0	2.0
Hyperfine correction	+310 712 229.4	1.7
dc Stark effect	0	1.0
ac Stark shift, 486 nm scattered	0	1.0
Zeeman shift	0	0.93
Pressure shift	0	0.5
Blackbody radiation shift	+1.0	0.3
Power modulation AOM chirp	0	0.3
rf discharge ac Stark shift	0	0.03
Higher order modes	0	0.03
Line pulling by $m_F = 0$ component	0	0.004
Recoil shift	0	0.009
CSF1	0	1.87
Fiber link	0	0.025
Gravitational redshift	−128.70	0.11
Total	+310 712 125.2	10.8

comparison the current result is presented with the previous and other measurements from 1999 [4] and 2003 [6] in Fig. 3. The new measurement is in good agreement with the measurement [7] made with a local cesium fountain
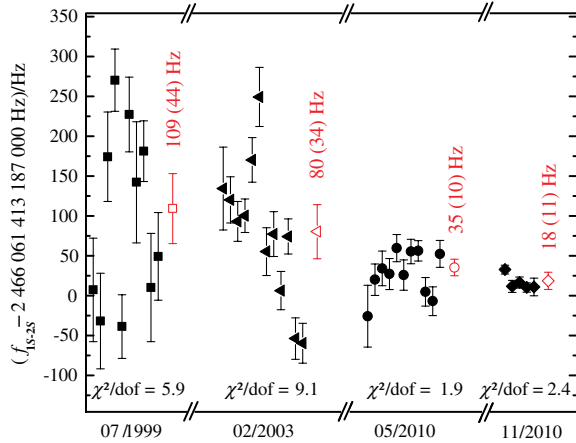

FIG. 3 (color online). Comparison of the hydrogen 1S-2S centroid frequency derived from the 1S(F = 1)-2S(F = 1) hyperfine component including the most recent November 2010 measurement [4,6,7]. Each black point represents a one-day average. The labeled red hollow points are the weighted mean values including systematic uncertainties. To measure the latter we also give χ^2 per degree of freedom (d.o.f.) of the computed average.

clock but has almost a factor of 2 smaller statistical uncertainty (3.3 Hz) as shown in Figs. 2 and 3 due to the higher detection efficiency.

As part of the search for a fundamental physical theory that can unify quantum mechanics and gravitation, tests for new physics beyond the standard model of particle physics are extremely important. Many candidate theories of quantum gravity, including loop quantum gravity [14,15] and string theory [16], may allow violations of Lorentz invariance and the combined charge-conjugation-parity-time-reversal invariance (CPT). The minimal standard model extension (SME) introduced by Kostelecký and co-workers [11,12] offers a systematic way of parameterizing such violations. The results of Lorentz and CPT violation tests—whether from astrophysical, accelerator, or nonrelativistic laboratory experiments—can be expressed in terms of the parameters of the SME.

The Lagrange density of the SME includes operators than can be constructed from standard model fields, contracted with Lorentz-violating background tensors. Bounds on these background tensors are conventionally expressed in a sun-centered right-handed celestial equatorial coordinate system frame with coordinates (X, Y, Z, T). X points toward the vernal equinox and Z along the Earth's rotation axis; T is measured relative to the vernal equinox of the year 2000.

The tensor parameters that can be constrained with our measurements are conventionally denoted $c_{(TX)}$, $c_{(TY)}$, and $c_{(TZ)}$ [17]; they break isotropy and boost invariance. The effects they produce are spin independent but odd under parity, and this combination of features makes them more difficult to constrain than some other coefficients. There are constraints on them coming both from astrophysical [18] and accelerator [17,19] data; however, except for Ref. [19], the bounds were all based on reanalyses of preexisting data which had been taken for a different purpose. In contrast to that, dedicated experiments can be designed to maximize their sensitivity to certain parameters. Having four measurements of the 1S-2S transition conducted at different times of the year, we can constrain two linear combinations of the coefficients.

Our analysis of the experimental data is based on the work [20]. It was shown there that in case of nonzero $c_{(TJ)}$ parameters, the results of hydrogen 1S-2S frequency measurements should vary with the velocity of the frame in which the measurement is performed. Variation of the measured frequency can be expressed as

$$\frac{\delta f_{1S-2S}}{f_{1S-2S}} = \frac{5}{3}\frac{1}{c}[v_X c_{(TX)} + v_Y c_{(TY)} + v_Z c_{(TZ)}], \quad (2)$$

in terms of the laboratory's velocity $\vec{v} = (v_X, v_Y, v_Z)$. Neglecting the effects of the Earth's rotation, the laboratory's velocity is just that of the Earth moving around the Sun with magnitude $v \approx 10^{-4}c$, $\vec{v} = v(\sin\Omega_\oplus T, -\cos\eta\cos\Omega_\oplus T, -\sin\eta\cos\Omega_\oplus T)$, $\Omega_\oplus$ being

the orbital angular frequency and $\eta \approx 23.4°$ the Earth's axial tilt. Consequently, we can parametrize the expected annual variations of the 1S-2S frequency:

$$\frac{\delta f_{1S\text{-}2S}}{f_{1S\text{-}2S}} = \frac{5}{3}\frac{v}{c}\{c_{(TX)}\sin\Omega_\oplus T$$
$$- [c_{(TY)}\cos\eta + c_{(TZ)}\sin\eta]\cos\Omega_\oplus T\}. \quad (3)$$

It is clear that we can constrain only two linear combinations of coefficients; the combination $c_{(TY)}\sin\eta - c_{(TZ)}\cos\eta$ cannot be constrained with the present method because it requires change of velocity in the direction orthogonal to the Earth's orbital plane. From the measurement points taken in 2010 we can find a constraint for the following linear combination: $0.95c_{(TX)} - 0.28c_{(TY)} - 0.12c_{(TZ)} = (2.1 \pm 1.8) \times 10^{-11}$. This constraint is more precise by a factor of 4 than the bounds placed on another linear combination in Ref. [20].

Analysis of the 1S-2S frequency measurements made in 1999 and 2003 shows that the averaged value obtained during those measurements differs from the averaged 2010 value by 2.4 standard deviations. We believe that this difference is due to systematic uncertainties (second order Doppler shift and ac Stark shift) that were underestimated in 1999 and 2003 and a different model for analysis used then. Nevertheless, since there were no changes in the hydrogen beam apparatus between 1999 and 2003 we can assume that they have common systematic shifts. For 1999 and 2003 we therefore obtain $0.83c_{(TX)} + 0.51c_{(TY)} + 0.22c_{(TZ)} = (4 \pm 8) \times 10^{-11}$. From both linear combinations we can derive the following constraints:

$$c_{(TX)} = (3.1 \pm 1.9) \times 10^{-11},$$
$$0.92c_{(TY)} + 0.40c_{(TZ)} = (2.6 \pm 5.3) \times 10^{-11}. \quad (4)$$

The Lorentz violation coefficients for electrons cannot be measured in isolation. Only differences between these coefficients and the analogous coefficients in other sectors of the theory are observable. Because the 1S-2S spectroscopy experiment uses a cesium clock as a frequency reference, it ultimately compares coefficients between the electron and proton sectors. However, the proton coefficients that could potentially become involved have already been constrained extremely tightly by making comparisons between cesium Zeeman frequencies and the frequencies of electromagnetic oscillators composed of bulk matter [21].

The particular specificity of the 1S-2S measurements for constraining these particular Lorentz violation is based on several different factors. Because both the 1S and 2S states are spherically symmetric, the difference in the electron energies between these states is insensitive to most other SME coefficients. The hyperfine energies depend in principle on several other SME coefficients, but these have already been sufficiently strongly constrained that they may safely be ignored in this analysis [20]. The

measurements of the spin-dependent SME terms were made at low laboratory energies, just like the 1S-2S measurement.

The fact that the present measurements were made at low energies is significant for two reasons. First, a laser spectroscopy experiment presents a very clean environment. Bounds based on astrophysical observations frequently rely on having an accurate understanding of the extremely energetic phenomena that are occurring inside distant objects. Accelerator experiments are better understood but still lack the well controlled environment of a precision frequency measurement.

The second reason is somewhat more subtle. The SME is an effective field theory, and it is only expected to be valid up to some energy scale Λ. As this scale Λ is approached, new forms of Lorentz violation may become important. Bounds based on the observed absence of a process such as vacuum Cherenkov radiation ($e^- \rightarrow e^- + \gamma$) or photon decay ($\gamma \rightarrow e^+ + e^-$) up to some energy E naively indicate bounds at the $m_e^2 c^4/E^2$ level. However, the threshold energy for such processes is exactly the same energy at which new forms of Lorentz violation may be expected to become important [22]. This means that the threshold bounds are actually rather strongly model dependent. This model dependence does not exist for bounds obtained with experiments performed at lower energies. The hydrogen 1S-2S result also gives the only strong terrestrial bound involving $c_{(TZ)}$ that is not based on the absence of model-dependent reaction thresholds. This gives our measurement a unique sensitivity not found elsewhere.

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[1] R. Pohl et al., Nature (London) **466**, 213 (2010).
[2] P. J. Mohr, B. N. Taylor, and D. B. Newell, Rev. Mod. Phys. **84**, 1527 (2012).
[3] Th. Udem, R. Holzwarth, T. W. Hänsch, Nature (London) **416**, 233 (2002).
[4] M. Niering et al., Phys. Rev. Lett. **84**, 5496 (2000).

[5] J. Guéna *et al.*, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **59**, 391 (2012).

[6] M. Fischer *et al.*, Phys. Rev. Lett. **92**, 230802 (2004).

[7] C. G. Parthey *et al.*, Phys. Rev. Lett. **107**, 203001 (2011).

[8] S. Weyers, U. Hübner, R. Schröder, Chr. Tamm, and A. Bauch, Metrologia **38**, 343 (2001).

[9] S. Weyers, A. Bauch, R. Schröder, and Chr. Tamm, *Proceedings of the 6th Symposium on Frequency Standards and Metrology*, edited by P. Gill (World Scientific, Singapore, 2001), p. 64.

[10] K. Predehl *et al.*, Science **336**, 441 (2012).

[11] V. A. Kostelecký and R. Potting, Nucl. Phys. **B359**, 545 (1991).

[12] D. Colladay and V. A. Kostelecký, Phys. Rev. D **58**, 116002 (1998).

[13] N. Kolachevsky, J. Alnis, C. G. Parthey, A. Matveev, R. Landig, and T. W. Hänsch, Opt. Lett. **36**, 4299 (2011).

[14] R. Gambini and J. Pullin, Phys. Rev. D **59**, 124021 (1999).

[15] J. Alfaro, H. A. Morales-Técotl, and L. F. Urrutia, Phys. Rev. D **65**, 103509 (2002).

[16] V. A. Kostelecký and S. Samuel, Phys. Rev. D **39**, 683 (1989).

[17] B. Altschul, Phys. Rev. D **82**, 016002 (2010).

[18] B. Altschul, Phys. Rev. D **74**, 083003 (2006).

[19] J.-P. Bocquet *et al.*, Phys. Rev. Lett. **104**, 241601 (2010).

[20] B. Altschul, Phys. Rev. D **81**, 041701(R) (2010).

[21] P. Wolf, F. Chapelet, S. Bize, and A. Clairon, Phys. Rev. Lett. **96**, 060801 (2006).

[22] V. A. Kostelecký and R. Lehnert, Phys. Rev. D **63**, 065008 (2001).