Phase-Coherent Measurement of the Hydrogen 1*S*-2*S* Transition Frequency with an Optical Frequency Interval Divider Chain

Th. Udem, A. Huber, B. Gross, J. Reichert, M. Prevedelli,* M. Weitz, and T. W. Hänsch
Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, 85748 Garching, Germany
(Received 21 May 1997)

We have measured the absolute frequency of the hydrogen 1*S*-2*S* two-photon resonance with an accuracy of 3.4 parts in 10^{13} by comparing it with the 28th harmonic of a methane-stabilized 3.39 μm He-Ne laser. A frequency mismatch of 2.1 THz at the 7th harmonic is bridged with a phase-locked chain of five optical frequency interval dividers. From the measured frequency $f_{1S\text{-}2S} = 2\,466\,061\,413\,187.34(84)$ kHz and published data of other authors we derive precise new values of the Rydberg constant, $R_\infty = 10\,973\,731.568\,639(91)$ m^{-1} and of the Lamb shift of the 1*S* ground state, $L_{1S} = 8172.876(29)$ MHz. These are now the most accurate values available. [S0031-9007(97)04182-3]

PACS numbers: 31.30.Jv, 06.20.Jr, 21.10.Ft

For almost three decades, the 1*S*-2*S* two-photon transition in atomic hydrogen with its natural linewidth of only 1.3 Hz has inspired advances in high resolution laser spectroscopy and optical frequency metrology [1]. This resonance has become a *de facto* optical frequency standard. More importantly, it is providing a cornerstone for the determination of fundamental physical constants and for stringent tests of quantum electrodynamic theory. In the future, it may unveil conceivable slow changes of fundamental constants or even differences between matter and antimatter.

Here, we report on a new precise measurement of the absolute frequency of the 1*S*-2*S* interval which exceeds the accuracy of the best previous measurement [2] by almost 2 orders of magnitude. The 1*S*-2*S* resonance is observed by longitudinal Doppler-free two-photon spectroscopy of a cold atomic beam. The resonance frequency is compared with the frequency of a cesium atomic clock with the help of a phase-coherent laser frequency chain, using a transportable CH_4-stabilized He-Ne laser at 3.39 μm as an intermediate reference. In this way, we have determined a 1*S*-2*S* interval of $f_{1S\text{-}2S} = 2\,466\,061\,413\,187.34(84)$ kHz with an uncertainty of 3.4 parts in 10^{13}, limited by the reproducibility of the He-Ne reference laser. This represents now the most accurate measurement of any optical frequency in the ultraviolet and visible region. Together with the results of other authors, in particular, the recent precision measurements of the $2S_{1/2}$-$8D_{5/2}$ transition frequency in hydrogen by the group of Biraben [3], we derive new and more precise values for both the Rydberg constant and the 1*S* Lamb shift. This Lamb shift provides now the best test of quantum electrodynamics for an atom.

As in our earlier experiment [2] we are taking advantage of the near coincidence between the 1*S*-2*S* interval and the 28th harmonic of the frequency of a CH_4-stabilized 3.39 μm He-Ne laser. However, a frequency mismatch of 2.1 THz near the 7th harmonic is no longer bridged interferometrically but is now measured with the help of a phase-locked chain of five frequency interval dividers. We are thus demonstrating the viability of a new approach to measuring the frequency of light. The cascaded bisection of optical frequency intervals has long been proposed [1], but only single divider stages have been demonstrated before [4].

As illustrated in Fig. 1, such a divider stage receives two input laser frequencies f_1 and f_2, and it forces a third laser to oscillate at the precise midpoint $f_3 = (f_1 + f_2)/2$, by electronically phase locking the second harmonic $2f_3$ to the sum frequency $f_1 + f_2$ via a low-frequency beat signal. In this way the original frequency gap is divided by two. With a chain of n cascaded divider stages, a given frequency interval can be divided

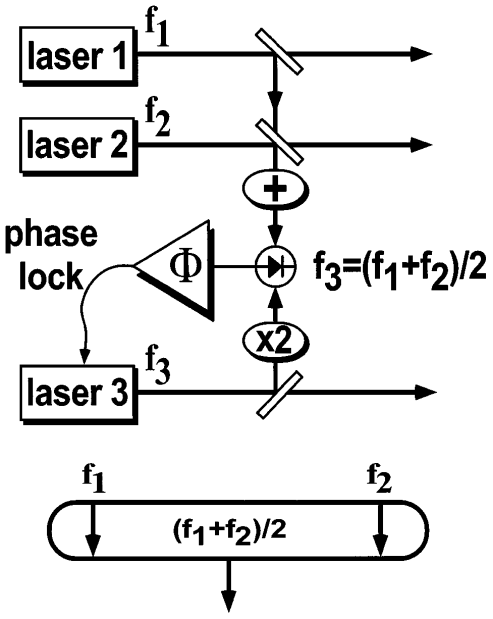

FIG. 1. Principle of an optical frequency interval divider and symbol as used in Fig. 2.

 0031-9007/97/79(14)/2646(4)$10.00

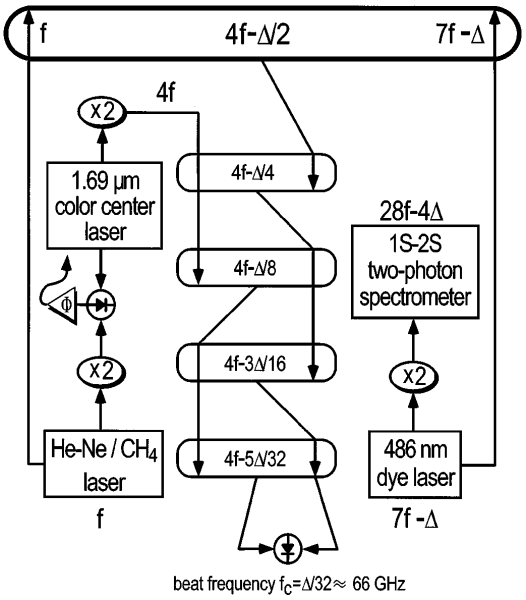

FIG. 2. Experimental setup for the comparison of the $1S$-$2S$ two-photon resonance at 243 nm with a 3.39 μm methane-stabilized He-Ne laser. The large-gap divider on top finds the midpoint between f and $7f - \Delta$: $4f - \Delta/2$. The difference to $4f$ is then further reduced using the cascaded divider stages.

by 2^n. In order to avoid $1/f$ noise it is advantageous to lock the beat signal not to zero but to some offset radio frequency f_{LO} of a local oscillator, so that $f_3 = (f_1 + f_2)/2 - f_{\mathrm{LO}}/2$ is no longer at the exact midpoint, but still precisely known. This kind of heterodyning allows us to monitor the performance of the phase-locked loops by simply counting the beat frequency, confirming that the uncertainty introduced by cycle slipping is absolutely negligible at the present level of precision.

An overview of our experiment is shown in Fig. 2. The reference frequency f is provided by a transportable CH_4-stabilized He-Ne laser that has been built by the group of Bagayev at the Institute of Laser Physics in Novosibirsk, Russia [5]. Since our earlier experiment [2] this laser has been somewhat improved, and its output frequency is now reproducible within 2.6×10^{-13}, as confirmed by several calibrations at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig before and during the $1S$-$2S$ measurement. The comparisons with the German atomic cesium time standard with the help of a harmonic frequency chain at the PTB [6] give a frequency $f = 88\,376\,182\,599\,937(23)$ Hz.

The frequency of the He-Ne laser is doubled in an angle-tuned $AgGaSe_2$ crystal, and a $NaCl{:}OH^-$ color center laser is electronically phase locked to the second harmonic $2f$. It acts as a transfer oscillator and provides sufficient power for further frequency doubling in a $LiIO_3$ crystal so that we produce the 4th harmonic $4f$ of the He-Ne standard.

This wavelength region near 848 nm is convenient for implementing an optical frequency interval divider chain with readily available grating-tuned diode lasers. In order to measure the frequency $7f - \Delta$ of the dye laser used in the hydrogen spectrometer we employ a first large-gap divider stage to bisect the interval between the dye laser frequency and the He-Ne reference, producing a frequency $4f - \Delta/2$. We then compare this frequency with the 4th harmonic of the standard using 4 additional cascaded divider stages to reduce the frequency gap $\Delta/2$ by a factor of 16. The final beat frequency of $f_c \approx 66$ GHz is accessible to a commercial fast photodiode, followed by a harmonic mixer and a synthesizer for down conversion and a radio frequency counter. Since our diode lasers have a relatively broad linewidth we use a digital phase locking scheme [7] that keeps track of the accumulated optical phase, making the divider chain rather immune to external perturbations so that it can remain locked for hours. As long as the rms phase fluctuations stay below 1 cycle, a measuring time of 1 s is sufficient to transport an optical frequency accuracy of better than one part in 10^{14}.

In the phase-locked condition our frequency chain relates the frequency of the observed $1S$-$2S$ hyperfine component f^F_{1S-2S} to the measured microwave frequency f_c through

$$f^F_{1S\text{-}2S} = 28f - f_{\mathrm{LO}} - 128f_c\,. \qquad (1)$$

The local oscillator frequencies used in the frequency chain are summed up in $f_{\mathrm{LO}} = 3$ GHz. In all the measurements, the local oscillators and the rf counter have been referenced to a local cesium clock.

The hydrogen spectrometer has been described before in detail [8]. The hydrogen $1S(F = 1, m_F = \pm 1) \rightarrow 2S(F' = 1, m_{F'} = \pm 1)$ transition is driven in a cold atomic beam by longitudinal Doppler-free two-photon excitation with light at 243 nm that is generated by doubling the frequency of an ultrastable dye laser at 486 nm in a crystal of beta barium borate, as indicated on the right side of Fig. 2. The uv radiation is resonantly enhanced in a linear cavity inside the vacuum chamber of the atomic beam apparatus. Hydrogen atoms from a gas discharge escape from a nozzle cooled with a liquid helium flow-through cryostat. Cold atoms traveling along the laser field are excited to the metastable $2S$ state. An electric quenching field a distance $l \simeq 10$ cm downstream from the nozzle forces the emission of 121 nm Lyman-α photons which are detected with a solar blind photomultiplier.

In order to reduce transit time broadening and second order Doppler shifts we select the slowest atoms by turning the laser light periodically off with a chopper and counting signal photons only if they arrive after a time delay τ that was chosen between 0.5 and 1.5 ms. An upper limit for the second order Doppler shift is given by $\Delta\nu_{\mathrm{max}} = -1/2(l/\tau c)^2 f^{\mathrm{F}}_{1S\text{-}2S}$. We have taken into account only those measurements for which $\Delta\nu_{\mathrm{max}} \leq 350$ Hz. A realistic model for the line shape [9] predicts that the actual

shift of the line center is approximately 4 times smaller than $\Delta\nu_{\max}$, since the slowest atoms which spend the longest time in the light field contribute a disproportionately large part to the signal. Other uncertainties of the hydrogen spectrometer such as dc and ac Stark shifts are estimated to contribute less than 325 Hz [8].

Figure 3 shows an example of a $1S$-$2S$ excitation spectrum, recorded with a delay time $\tau = 1.5$ ms. We determine the line center by fitting a Lorentzian to the experimental data. Simulations [9] confirm that this procedure is easily sufficient at the present level of accuracy, even though the expected line shape is not exactly Lorentzian. Evaluating the line centers of 41 spectra with $\Delta\nu_{\max} \leq 350$ Hz we obtain a mean value of the measured microwave frequency $f_c = 66\,164\,143\,149.4(2.0)$ Hz. We determine the optical frequency $f_{1S\text{-}2S}^{F=1}$ according to the prescription (1) and add a correction for the well-known hyperfine splitting of the $1S$ and $2S$ [10] levels, $f_{\rm hf} = 310\,712\,223(13)$ Hz, to find the frequency of the hyperfine centroid,

$$f_{1S\text{-}2S} = 2\,466\,061\,413\,187.34(84)\ \text{kHz}\,.$$

The uncertainty of 3.4 parts in 10^{13} is dominated by the CH_4-stabilized He-Ne laser reference. The $1S$-$2S$ resonance line shape in our experiment is now well enough understood [9] that we could determine the line center to within 1.5 parts in 10^{14} if a sufficiently accurate optical frequency standard were available.

An important complement to our $1S$-$2S$ frequency measurement is the recent precise measurement of the hydrogen $2S_{1/2}$-$8D_{5/2}$ frequency $f_{2S\text{-}8D} = 770\,649\,561\,585.0(4.9)$ kHz with an uncertainty of 6 parts in 10^{12} by the group of Biraben [3]. The Rydberg constant R_∞ and the ground state Lamb shift L_{1S} can be determined from these two measurements by solving the set of linear equations,

$$f_{1S\text{-}2S} = R_\infty[e(2S) - e(1S)] + L_{2S} - L_{1S}\,, \quad (2)$$

$$f_{2S\text{-}8D} = R_\infty[e(8D) - e(2S)] + L_{8D} - L_{2S}\,. \quad (3)$$

Here, the term $R_\infty e(nS/D)$ gives the Dirac energy (in Hz) of the corresponding state including the recoil corrections due to the finite nuclear mass that are not considered to be part of the Lamb shift [11]. They are calculated with the most recent value of the fine structure constant $\alpha^{-1} = 137.035\,999\,44(57)$ [12] and the electron-proton mass ratio 1836.152 666 5(40) [13]. For the small $8D_{5/2}$ Lamb shift we can use the theoretical value $L_{8D_{5/2}} = 71.4(2)$ kHz. In the past, before optical frequency measurements had reached the present level of accuracy, it would have been best to use a value of the $2S$ Lamb shift as derived from radio frequency measurements of the hydrogen $2S$-$2P$ splitting. Using the weighted average of Refs. [14,15], where the result of [15] is recalculated according to [16] using a new value for the fine structure splitting, and with the theoretical values for the small Lamb shifts $L_{2P_{1/2}}$ and $L_{2P_{3/2}}$, we arrive at $L_{2S} = 1045.0079(72)$ MHz. In the future, when optical frequency measurements reach even higher accuracy, it will be better to replace L_{2S} in both equations by $(L_{1S} - L')/8$, where $L' = L_{1S} - 8L_{2S} = -187.232(5)$ MHz can be calculated much more accurately than the Lamb shifts of the individual levels [17]. At present, both approaches give results with comparable accuracies, and smaller uncertainties are obtained if we take the arithmetic averages of L_{1S} and R_∞ as derived by these two methods, as shown in Table I for L_{1S}. Note that both approaches involve some small theoretical uncertainties (in $L_{8D_{5/2}}$, $L_{2P_{1/2}}$, and especially in L') due to uncalculated higher order QED terms.

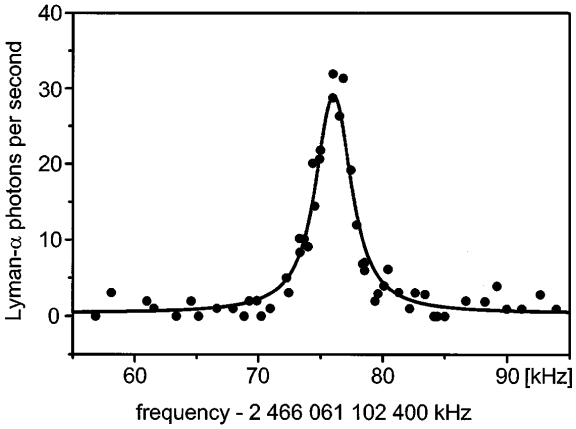

FIG. 3. Hydrogen $1S(F = 1) \rightarrow 2S(F' = 1)$ resonance as measured with the optical interval divider chain. There is an uncertainty in the absolute frequency scale of 640 Hz.

In this way we find a hydrogen $1S$ ground state Lamb shift

$$L_{1S} = 8172.876(29)\ \text{MHz} \quad \text{(hydrogen)}\,.$$

This result is in agreement with earlier determinations of the $1S$ Lamb shift [18–20]. However, it is in only moderate agreement with the theoretical value $L_{1S,\text{theo}} = 8172.797(40)$ MHz using a rms proton charge radius $r_p = 0.862(12)$ fm [21] and a new value of the electron-proton mass ratio [13]. The use of other proton charge radii, as stated in the literature [22,23], further reduces this moderate agreement. Alternatively our experiment can be interpreted as a measurement of the proton rms charge radius, yielding $r_p = 0.890(14)$ fm, provided that the theoretical calculations are correct. Independent more accurate measurements of the proton charge radius, for instance, via the $2S$ Lamb shift of muonic hydrogen, would permit stringent new tests of bound state QED.

We can use an earlier measurement of the hydrogen-deuterium isotope shift of the $1S$-$2S$ transition [670 994 337(22) kHz [24]] to derive the absolute frequency of the $1S$-$2S$ interval in deuterium. Inserting this value and the absolute frequency of the deuterium $2S$-$8D$ splitting $f_{2S-8D_{5/2}} = 770\,859\,252\,851.5(4.4)$ kHz

TABLE I. Results of L_{1S} and dominating contributions to the total uncertainties (in kHz).

Source of uncertainty	Using rf L_{2S} measurements	Using theoretical $L' = L_{1S}$-$8L_{2S}$	Mean
L_{2S}	30.2	...	15.1
$f_{2S\text{-}8D}$	15.7	33.0	24.4
$f_{1S\text{-}2S}$	0.8	1.8	1.3
$L_{8D_{5/2}}$	0.6	1.4	1.0
L'	...	5.5	2.8
Results (L_{1S})	8 172 860(34)	8 172 892(34)	8 172 876(29)

[3] into the set of Eqs. (2) and (3) we obtain a deuterium ground state Lamb shift

$$L_{1S} = 8184.011(55)\ \text{MHz}\ \text{(deuterium)}.$$

Here, we have replaced L_{2S} in both equations by $(L_{1S} - L')/8$, with $L' = -187.225(5)$ MHz [17], since we do not know of any sufficiently precise measurements of the $2S$-$2P$ splitting in deuterium.

Finally, the outlined method yields a Rydberg constant $R_\infty = 10\,973\,731.568\,70(11)\ \text{m}^{-1}$ with the hydrogen data, and $R_\infty = 10\,973\,731.568\,72(14)\ \text{m}^{-1}$ with the data for deuterium. A slightly more accurate Rydberg value is obtained, if we use as additional input the results of three independent recent hydrogen $1S$ Lamb shift measurements based on a comparison of the hydrogen $1S$-$2S$ interval with the $2S$-$4S/4D$ [18] and $2S$-$4P$ [19] intervals as well as a comparison of the $1S$-$3S$ interval with the $2S$-$6S/6D$ interval [20]. In all three experiments, a combination of Lamb shifts is determined from a measured difference of optical frequencies of a few GHz. We can now replace Eq. (3) by analogous equations for the measured beat frequencies, such as the $f_{2S\text{-}4S/D} - 1/4 f_{1S\text{-}2S}$. We have solved these by the method outlined above, using subsequently the hydrogen $2S$-$2P$ splitting determined by rf measurements and $(L_{1S} - L')/8$, as well as analogous combinations to replace the $3S$, $4S$, and $6S$ Lamb shifts [17]. We use theoretical values for the smaller and better known Lamb shifts of the higher excited states as stated by the authors of the corresponding measurement [18–20]. Our final result for the Rydberg constant is the average weighted by the squared uncertainties of individual results for R_∞

$$R_\infty = 10\,973\,731.568\,639(91)\ \text{m}^{-1}.$$

This value is in good agreement with the result $R_\infty = 10\,973\,731.568\,59(10)\ \text{m}^{-1}$ recently determined in Paris [3]. The main uncertainties are due to the rf L_{2S} measurements ($59 \times 10^{-6}\ \text{m}^{-1}$), the optical $1S$ Lamb shift measurements ($46 \times 10^{-6}\ \text{m}^{-1}$), and the $2S_{1/2}$-$8D_{5/2}$ frequency ($48 \times 10^{-6}\ \text{m}^{-1}$). For future improvements precise frequency measurements of the $2S$-nS/D transitions in hydrogen are desirable.

We thank A. K. Dmitriyev of the Institute of Laser Physics in Novosibirsk, Russia, for his help with the CH_4-stabilized He-Ne laser standard. We are also indebted to G. Kramer and B. Lipphardt of the PTB in Braunschweig, Germany, and D. A. Tyurikov of the P. N. Lebedev Institute, Moscow, for their essential support in the calibration of this standard. This work has been supported in part by the Deutsche Forschungsgemeinschaft.

*Present address: LENS, Largo E. Fermi 2, Florence, Italy.

[1] T. W. Hänsch, in *The Hydrogen Atom*, edited by G. F. Bassani, M. Inguscio, and T. W. Hänsch (Springer-Verlag, Berlin, 1989), p. 93.
[2] T. Andreae *et al.*, Phys. Rev. Lett. **69**, 1923 (1992).
[3] B. de Beauvoir *et al.*, Phys. Rev. Lett. **78**, 440 (1997).
[4] H. R. Telle, D. Meschede, and T. W. Hänsch, Opt. Lett. **15**, 532 (1990); R. Wynands, T. Mukai, and T. W. Hänsch, Opt. Lett. **17**, 1749 (1992).
[5] S. N. Bagayev *et al.*, Laser Phys. **7**, 1 (1997).
[6] C. O. Weiss *et al.*, IEEE J. Quantum Electron. **24**, 1970 (1988).
[7] M. Prevedelli, T. Freegarde, and T. W. Hänsch, Appl. Phys. B **60**, S241 (1995).
[8] C. Zimmermann, R. Kallenbach, and T. W. Hänsch, Phys. Rev. Lett. **65**, 571 (1990); F. Schmidt-Kaler *et al.*, Phys. Rev. A **51**, 2789 (1995).
[9] A. Huber *et al.* (to be published).
[10] L. Essen *et al.*, Nature (London) **229**, 110 (1971); J. W. Heberle, H. A. Reich, and P. Kusch, Phys. Rev. **101**, 612 (1956).
[11] K. Pachucki *et al.*, J. Phys. B **29**, 177 (1996).
[12] T. Kinoshita, Phys. Rev. Lett. **75**, 4728 (1995).
[13] D. L. Farnham, R. S. Van Dyck, Jr., and B. P. Schwinberg, Phys. Rev. Lett. **75**, 3598 (1995).
[14] S. R. Lundeen and F. M. Pipkin, Phys. Rev. Lett. **46**, 232 (1981).
[15] E. W. Hagley and F. M. Pipkin, Phys. Rev. Lett. **72**, 1172 (1994).
[16] U. Jentschura and K. Pachucki, Phys. Rev. A **54**, 1853 (1996).
[17] S. G. Karshenboim, Z. Phys. D **39**, 109 (1997).
[18] M. Weitz *et al.*, Phys. Rev. A **52**, 2664 (1995).
[19] D. J. Berkeland, E. A. Hinds, and M. G. Boshier, Phys. Rev. Lett. **75**, 2470 (1995).
[20] S. Bourzeix *et al.*, Phys. Rev. Lett. **76**, 384 (1996).
[21] G. G. Simon *et al.*, Nucl. Phys. **A333**, 381 (1980).
[22] L. N. Hand, D. J. Miller, and R. Wilson, Rev. Mod. Phys. **35**, 335 (1963).
[23] P. Mergell, U. G. Meißner, and D. Drechsel, Nucl. Phys. **A596**, 367 (1996).
[24] F. Schmidt-Kaler *et al.*, Phys. Rev. Lett. **70**, 2261 (1993).