Absolute Frequency Measurement of the Hydrogen 1S-2S Transition and a New Value of the Rydberg Constant

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(Received 15 July 1992)

We have compared the frequency of the hydrogen 1S-2S transition (resonance width $\Delta\nu/\nu = 2.8 \times 10^{-11}$) with a reference frequency synthesized from an 88-THz infrared CH_4-stabilized HeNe laser. In this first measurement of an optical frequency that overcomes the uncertainty limits of the visible I_2-stabilized He-Ne laser standard, we find a 1S-2S frequency of 2466061413.182(45) MHz with an 18-fold improved precision of 1.8 parts in 10^{11}. A new value of the Rydberg constant, $R_\infty = 109\,737.315\,684\,1(42)$ cm^{-1}, is deduced.

PACS numbers: 42.50.Wm, 06.20.Jr, 06.30.Ft, 42.62.Fi

Precision spectroscopy of the hydrogen atom has long provided a unique tool to measure fundamental constants and to test basic physics laws. The 1S-2S transition [1] with a natural linewidth of 1.3 Hz is perhaps the most tantalizing hydrogen resonance. A linewidth of $\Delta\nu/\nu = 2.8 \times 10^{-11}$ is observed [2] by Doppler-free two-photon spectroscopy of an atomic beam cooled to 80 K. We have measured the absolute optical frequency of this transition with an uncertainty of 1.8 parts in 10^{11} by comparison with a reference frequency synthesized with a phase-locked laser frequency chain from an 88-THz infrared CH_4-stabilized He-Ne laser, which has been calibrated against a microwave Cs frequency standard. Our result, $f(1S\text{-}2S) = 2466061413.182(45)$ MHz, is 18 times more accurate than the best [3] of the previous measurements [3-5].

From a comparison with theory we deduce a new Rydberg value, $R_\infty = 109\,737.315\,684\,1(42)$ cm^{-1}, somewhat below the 1989 measurement [6] of 109737.315709(18) cm^{-1} by Biraben *et al.*. The Rydberg constant is an important cornerstone in the system of fundamental constants and the principal scaling factor for any spectroscopic transition. With an uncertainty of 3.8 parts in 10^{11}, our new value represents the most accurate measurement of any fundamental constant.

The present experiment is the first of its kind that is not limited by the systematic errors of optical wavelength metrology, and the first to overcome the uncertainty limit of 1.6 parts in 10^{10} imposed by the internationally accepted visible wavelength standard, an I_2-stabilized He-Ne laser at 633 nm. The frequency of the relevant iodine resonance (power broadened linewidth $\Delta\nu/\nu \gtrsim 10^{-8}$) has been determined only once relative to the CH_4-stabilized helium-neon infrared frequency standard in a direct (but not yet phase-coherent) measurement of a visible optical frequency [7].

The new hydrogen frequency measurement not only improves our knowledge about the relationship between the standard of time and the system of fundamental constants, but it establishes a new set of reference frequencies throughout the ultraviolet, visible, infrared, and microwave regions, because the frequencies of all other hydrogen transitions may now be predicted with improved accuracy, provided the theory of quantum electrodynamics is correct.

The hydrogen spectrometer [2] has evolved from a setup described previously [1]. Hydrogen atoms emerge from a nozzle cooled to liquid-nitrogen temperature. While traveling 15 cm collinearly along the uv-standing wave field inside a 243-nm enhancement resonator, the atoms are excited by Doppler-free two-photon absorption to the 2S metastable state. To detect the metastable atoms, a small electric field inside the detector quenches the 2S state, and the emitted Lyman-α photons at 121 nm are detected by a photomultiplier. The 243-nm light is generated as the second harmonic of a 486-nm dye laser locked to a highly stable external resonator. The $1S(F=1) \rightarrow 2S(F'=1)$ resonance has a linewidth of about 35 kHz at 243 nm and appears redshifted by 24 kHz due to the second-order Doppler effect (Fig. 1). The laser frequency can be locked to the line maximum with the help of a personal computer.

For the frequency measurement of the 1S-2S transition we take advantage of its close coincidence with the 28th harmonic of the CH_4-stabilized HeNe laser frequency at 88 THz. The absolute frequency of the CH_4 line has been measured with respect to the Cs atomic clock in several different laboratories [8-10], making it the best-known midinfrared frequency and the highest frequency that has been compared to the Cs standard with a phase-locked frequency chain.

A detailed description of our laser frequency chain will be published elsewhere [11]. To transfer the full accuracy of the HeNe/CH_4 to the optical domain we phase coherently generate an auxiliary reference frequency at the eighth harmonic at 424 nm in three successive steps (left-hand side of Fig. 2). In each stage a 20-MHz heterodyne beat signal between a laser sufficiently powerful to drive the next nonlinear process and the second harmonic of its predecessor is generated with at least 40

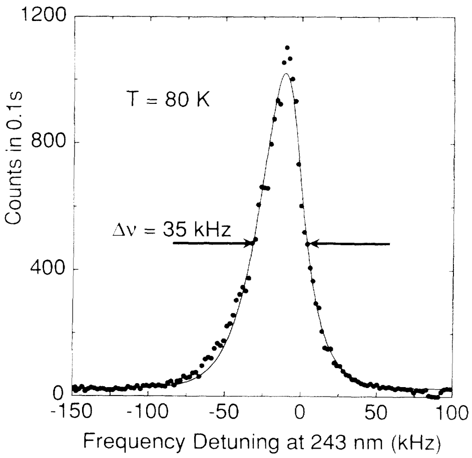

FIG. 1. Line profile of the $1S$-$2S$ two-photon resonance of the hydrogen atomic beam cooled to 80 K. The solid curve is a theoretical fit to the experimental points taking the second-order Doppler effect and time-of-flight broadening into account. The position of the true line center was deduced from the asymmetry of the experimental curve.

dB signal-to-noise ratio in a 100 kHz bandwidth. To achieve phase synchronization this signal is phase locked [12] to a stable rf oscillator controlled by a commercial rubidium standard which is long-term stabilized to the German national time standard via radio transmission. During our measurements the phase errors in each stage are monitored and found to be less than 1.5 rad at all times, thus assuring full coherence of the violet reference frequency with the HeNe/CH_4 standard.

Our transportable HeNe/CH_4 standard was built in the Institute of Laser Physics, Novosibirsk, Russia [13]. The linewidth of the inverted Lamb dip is 100 kHz (FWHM). To lock the laser to the molecular transition it is frequency modulated across the resonance, its first derivative yielding the error signal for the servo system. During ten days in November 1991, the HeNe/CH_4 standard was calibrated at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig by comparison with the Cs atomic clock using the PTB frequency chain [9]. The measured frequency was 88 376 181 599.67(15) kHz. We also measured the dependence of the absolute frequency on laser power and modulation amplitude. To increase the capture range of the CH_4 frequency lock during the hydrogen measurement the modulation amplitude was 3 times higher than during calibration at the PTB. This introduces a redshift of the HeNe/CH_4 frequency of 400 ± 50 Hz. Therefore the total uncertainty of our secondary frequency standard is 160 Hz and the relative uncertainty is 1.8×10^{-12}. The stability in terms of the Allan parameter [14] $\sigma(2,\tau,\tau)/\nu$ for averaging times 10 s $< \tau <$ 100 s is less than 6×10^{-14}.

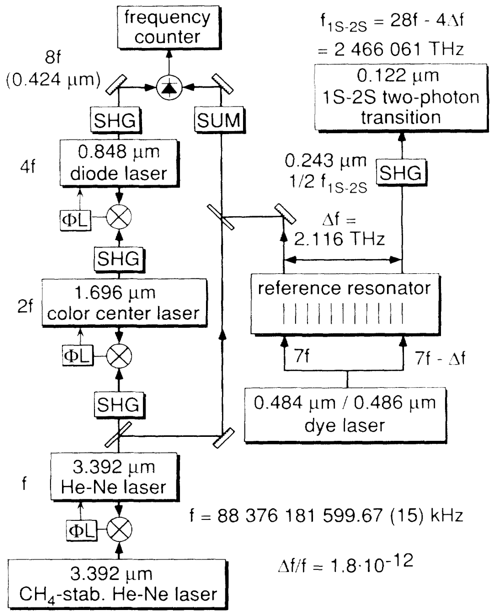

FIG. 2. Experimental setup for the frequency measurement of the $1S$-$2S$ transition in atomic hydrogen (ΦL, phase-locked loop: SHG, second harmonic generation).

Since the output power of the HeNe/CH_4 clock laser of only 0.4 mW is not sufficient to drive two nonlinear processes, a second HeNe laser with an output power of 20 mW is phase locked to the CH_4-stabilized laser. Half of the HeNe power is used to generate 10 nW of the second harmonic (SHG) in a $AgGaSe_2$ crystal.

For the next stage we have constructed a highly stable NaCl:OH^- color-center laser (based on a design by Beigang *et al.* [15]) which is phase locked to the second harmonic of the HeNe laser at 1.696 μm. To provide the link to the diode laser, a crystal of $LiIO_3$ generates 0.4 μW of second-harmonic light at 848 nm.

A 50-mW laser diode (STC LT50A-03U) is prestabilized by optical feedback from a confocal Fabry-Pérot etalon [16]. A $KNbO_3$ frequency doubler crystal produces more than 6 μW at 424 nm, providing the violet reference frequency for the $1S$-$2S$ measurement.

To measure the frequency of the dye laser at the seventh harmonic we sum its blue light and 10 mW of the fundamental HeNe/CH_4 power in a $LiNbO_3$ crystal (center of Fig. 2). The rf beat signal of the 200 nW output and the violet reference frequency at 424 nm is fed into a tracking filter. The filter output is counted with 1 Hz resolution and transferred to a microcomputer for further processing.

The right-hand side of Fig. 2 shows the dye laser of the $1S$-$2S$ spectrometer. Since the frequency ratio of 1:28 does not hold exactly, we have to bridge a difference frequency of $\Delta f = 2.1$ THz between 617 THz (486 nm) and 619 THz (484 nm). To measure Δf we use the transmission fringes of the stable reference resonator (finesse 70000, resonance width 5 kHz) as frequency markers.

For comparison with the 424-nm reference the dye laser is locked to a fringe nearest to the seventh harmonic, while for excitation of the $1S$-$2S$ transition another fringe separated by the difference frequency is chosen. Hence a measurement of the resonator mode spacing and a count of the number of spanned modes allows us to measure the difference frequency.

For the measurement of the mode spacing two dye lasers were stabilized to the cavity. The light of laser 1 was sent through an 84-GHz modulator [17], also controlled by the rubidium standard. A 20-Hz filter-limited beat linewidth was observed between the carrier of laser 2 and the first-order sideband of laser 1 with both lasers locked. At 84 GHz 253 free spectral ranges are spanned in this measurement. At a frequency of 616.561 THz, the measured FSR was 332383433.1(4) Hz. Measurements at several frequencies between 617 and 619 THz reveal systematic variations in the mode spacing no larger than 3 Hz, in agreement with theory [18].

In our current setup we cannot measure the hydrogen transition and count the beat frequency simultaneously. Instead, we first locked the dye laser to the hydrogen transition and recorded the slow drift of the resonator mode (approximately 15 Hz/s) with respect to the atomic transition. Next we locked the dye laser to another mode of the resonator such that we could count a low-frequency beat with the violet reference. Here we recorded the changing beat frequency, i.e., the drift of the second resonator mode with respect to the CH_4 transition. We repeated this cycle several times during a measurement day. The obtained drift curves were interpolated by a third-order polynomial and subtracted from each other. The resulting frequency difference was independent of time within our measurement precision. This difference plus 6372 modes of the reference cavity corresponds to the frequency difference Δf. The number of modes was extracted from a previous measurement of the $1S$-$2S$ transition frequency [3] that had an accuracy of 800 kHz (i.e., much less than one mode spacing).

We have carried out fifteen measurements during four nights in April and May 1992. From the distribution of our measurements we evaluate a mean of $f = 2466061102.470$ MHz for the $1S(F=1) \rightarrow 2S(F'=1)$ transition and a standard deviation of 42.5 kHz. We attribute this uncertainty to the interpolation procedure for the resonator drift. All significant remaining uncertainties are of systematic nature. The uncertainty in the resonator free spectral range of 0.4 Hz has to be multiplied by the number of modes, contributing $4 \times 6372 \times 0.4$ Hz$=10.2$ kHz at the $1S$-$2S$ frequency. The contributions to the uncertainty due to the $1S$-$2S$ spectrometer are the following: the $1S$-$2S$ line center lock point (5 kHz), the uncertainty of the second-order Doppler shift (6 kHz), and the dc Stark shift (1.8 kHz). Other sources of errors like ac Stark shift, magnetic fields, and the shift due to the wave-front curvature in our uv enhancement cavity used to excite the atoms are below 1 kHz. All these uncertainties add up quadratically to an error of 8.1 kHz.

The quadratic sum of the reproducibility and the systematic errors of the frequency standard of 160 Hz in the infrared contribute to the $1S$-$2S$ uncertainty with a factor of 28, i.e., 4.5 kHz. The total uncertainty then is 44.7 kHz. Taking into account the corrections for the hyperfine splitting of the $1S$ and $2S$ level we obtain a centroid frequency separation $f_{1S\text{-}2S} = 2466061413.182(45)$ MHz. A comparison with previous measurements is shown in Fig. 3.

The relationship between this frequency and the Rydberg constant is given by the Dirac theory, corrected for the finite nuclear mass and Lamb shifts. The Lamb shift of the $1S$ state, including the proton charge radius effect, has recently been measured [19,20] to a part in 10^5 by directly measuring the ratio of the $1S$-$2S$ and $2S$-$4S/4D$ two-photon transition frequencies. To make the error budget slightly smaller, we use this frequency ratio in combination with $f_{1S\text{-}2S}$ to determine the absolute frequency of the $2S$-$4S$ and $2S$-$4D$ transitions. Next we correct for the hyperfine splittings of the $2S$, $4S$, and $4D$ levels. We take the measured $2S$-$2P_{1/2}$ splitting and the theoretical values for the $2P_{1/2}$, $4S$, and $4D_{5/2}$ Lamb shifts from the literature as cited in Ref. [19] and obtain adjusted $2S$-$4S/2D$ transition frequencies which do not contain the Lamb shift or hyperfine structure contributions any more and can be compared with the predictions according to the Dirac theory including relativistic reduced mass contributions. Solving for the Rydberg constant we obtain $R_\infty = 109737.3156841(42)$ cm^{-1}. The uncertainty of 3.8×10^{-11} is more than a factor of 4 smaller than the one of the most recently published value of this fundamental constant [6] and is largely due to the uncertainties in the beat frequency measurement between $f_{2S\text{-}4S/4D}$ and $\frac{1}{4} f_{1S\text{-}2S}$. With this measurement we have reached a state where the uncertainties of small corrections [19] such as the Lamb shift, the electron-proton mass ratio, and the proton charge radius are becoming more important than the experimental accuracy of the

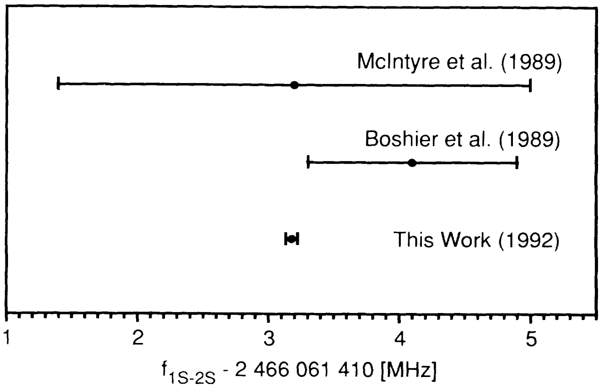

FIG. 3. Comparison of our value for the $1S$-$2S$ transition frequency with those of previous measurements [3-5].

$1S$-$2S$ energy separation.

The accuracy of the $1S$-$2S$ frequency can be further improved with a phase-coherent measurement of the remaining frequency mismatch of 2.1 THz. One could for instance use a recently demonstrated divider scheme [21,22] to repeatedly halve the frequency difference until the beat frequency can be counted. With an upgraded HeNe/CH_4 laser system which can resolve the hyperfine structure of the CH_4 the reproducibility of our standard could be increased by a factor of 5. Finally, one can cool the atomic beam to liquid-helium temperature which will further increase the resolution of the $1S$-$2S$ transition.

We gratefully acknowledge much help and advice from V. P. Chebotayev, G. Kramer, H. R. Telle, and J. L. Hall. We also thank M. Weitz for his essential assistance with the determination of the Rydberg constant. This work has been supported in part by the Deutsche Forschungsgemeinschaft.

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