Absolute Frequency Measurement of the $2S$-$8S/D$ Transitions in Hydrogen and Deuterium: New Determination of the Rydberg Constant

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We have performed a pure optical frequency measurement of the $2S$-$8S/D$ two-photon transitions in atomic hydrogen and deuterium. These frequencies are directly compared to a new frequency standard, a diode laser stabilized to a two-photon transition at 778 nm in rubidium. We deduce a new value for the Rydberg constant, $R_\infty = 109\,737.315\,685\,9(10)$ cm^{-1} with an uncertainty of 9×10^{-12}. From the isotope shift, we derive a precise value of the $2S$ Lamb shift in deuterium [$L_{2S\text{-}2P} = 1059.230(9)$ MHz] and the difference of the quadratic charge radii of deuteron and proton. [S0031-9007(96)02260-0]

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Hydrogen is the simplest atom, and its properties have been calculated very precisely: quantum electrodynamics (QED) calculations have continuously improved to achieve an impressive accuracy, currently of order 10^{-11} [1]. At the same time, experimental measurements in hydrogen have been performed at a comparable level of precision to deduce the Rydberg constant and to test the QED calculations [2]. Recently, the interferometric measurements have been superseded by accurate optical frequency ones. These measurements need frequency-multiplication chains which link the measured frequency via intermediate standard lasers to the cesium clock. With a frequency chain starting from the methane-stabilized helium-neon laser, Hänsch and co-workers measured the frequency of the $1S$-$2S$ two-photon transition with an uncertainty of 1.8×10^{-11} [3]. In our group, we built a frequency chain linking the frequencies of the $2S$-$8S/D$ two-photon transitions to two standard lasers (the iodine-stabilized and the methane-stabilized helium-neon lasers) and reached a precision of 1.3×10^{-11} [4]. Here we present a new optical frequency measurement of the $2S$-$8S/D$ transitions in hydrogen and deuterium with a frequency-multiplication chain. The relative uncertainty is reduced to about 6×10^{-12} and provides a more precise value of the Rydberg constant. In deuterium, we give a precise determination of the $2S$ Lamb shift and, from the isotope shift, we obtain the difference of the squared proton and deuteron charge radii $r_d^2 - r_p^2$.

Our frequency chain connects indirectly hydrogen frequencies to the cesium clock (see Fig. 1). The experiment is carried out at two different laboratories, the *Laboratoire Primaire du Temps et des Fréquences* (LPTF) at the *Observatoire de Paris* and the *Laboratoire Kastler Brossel* (LKB) in the *Université Pierre et Marie Curie*, which are linked by two, 3 km long, optical fibers. The cornerstone of this chain is a new standard, namely a laser diode at 778 nm (i.e., $\nu = 385$ THz) stabilized to the $5S_{1/2}$-$5D_{5/2}$ two-photon transition of rubidium (LD/Rb laser) [5]. The laser diode is used in an extended cavity configuration, the rubidium cell is placed inside an enhancement cavity, and the transition is detected by monitoring the fluorescence from the radiative cascade $5D$-$6P$-$5S$. The main metrological features of the LD/Rb laser are a 4×10^{-13} short term stability for $1s$ integration time and a day-to-day repeatability of 400 Hz. Two such lasers are operational at LPTF and one at LKB. The frequencies of these lasers are compared between the two laboratories using the optical fibers. To check the frequency shift due to the fiber, we have used a titanium-sapphire laser with a frequency jitter reduced to the few kHz level. After a round trip of 6 km through the optical fibers, we have observed a maximum frequency shift of 3 Hz. This shift is completely negligible for our frequency measurements. The frequencies of the LD/Rb lasers have been measured at the LPTF with a frequency chain which connects the LD/Rb laser

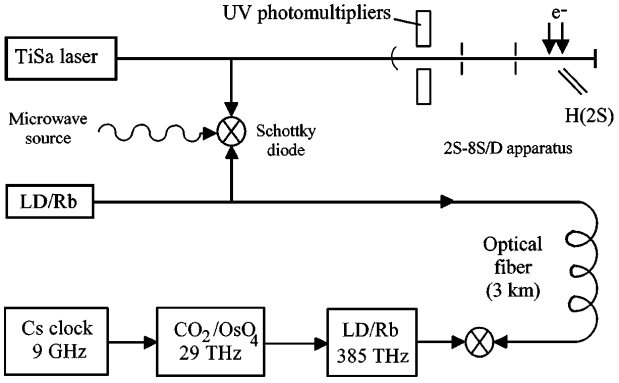

FIG. 1. Outline of the frequency chain between the $2S$-$8S/D$ hydrogen frequencies and the cesium clock.

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at 385 THz to a standard at 29 THz, which is a CO_2 laser stabilized to an osmium tetraoxyde line (CO_2/OsO_4). This standard had been previously measured with an uncertainty of 70 Hz [6]. The details of this frequency chain will be published shortly [7]. The frequency of the LD/Rb standard laser working at the LKB has been measured directly with respect to the CO_2/OsO_4 standard using the optical fiber and the frequency chain of the LPTF. Its frequency is

$$\nu_{\mathrm{Rb}}(\mathrm{LKB}) = 385\,285\,142\,370.5\ (1.0)\ \mathrm{kHz}\,.$$

The uncertainty is due to the CO_2/OsO_4 standard (13×70 Hz) and the day-to-day repeatability of the LD/Rb standard (about 400 Hz). Thanks to regular frequency comparisons between the LD/Rb standards, we have kept this precision throughout the measurements in hydrogen and deuterium (about four months).

The frequency comparison between the $2S$-$8S/D$ transitions and the LD/Rb standard is easy, thanks to the quasicoincidence between these frequencies (about 40 GHz in hydrogen and 144 GHz in deuterium). The LD/Rb laser and the titanium-sapphire laser (used for the observation of the hydrogen lines) are focused on a Schottky diode [8]. For the measurements in hydrogen, the Schottky diode is simultaneously irradiated with a microwave source at 13 GHz (48.4 GHz for deuterium) phase-locked to a low phase-noise quartz oscillator at 100 MHz. We detect the low-frequency beat note between the two optical frequencies and the third harmonic of the microwave source with a signal-to-noise ratio of 35 dB (resolution bandwidth of 300 kHz). A tracking oscillator is phase locked to this beat signal, and we count continuously this beat frequency. The frequency of the 100 MHz quartz oscillator has been continuously measured with respect to a high stability quartz oscillator at 10 MHz (stability of 2×10^{-9} during four months), which has been compared to a hydrogen maser several times. We estimate the uncertainty due to the Schottky diode frequency measurement to be about 15 Hz in hydrogen and 50 Hz in deuterium.

The hydrogen experiment has been described elsewhere [4,9]. The two-photon transition is induced with a highly stable titanium-sapphire laser. The frequency jitter is reduced to the level of 2 kHz by locking to a first cavity. The long term stability is obtained by a second reference cavity locked to an iodine stabilized He-Ne laser [10]. To reduce the collisional and transit time broadening, we use a metastable atomic beam collinear with the laser beams. The atomic beam is placed inside an enhancement cavity, where the optical power can be as much as 100 W in each direction with a beam waist of 660 μm. The laser-atom interaction region (56 cm long) is surrounded by a magnetic shield. To measure the metastable flux, we detect the Lyman-α fluorescence at the end of the beam, where an electric field quenches the metastable atoms. When the laser frequency is resonant with the $2S$-$8S/D$ transition, optical quenching of the metastable atoms occurs and we observe the corresponding decrease of the metastable yield (see Fig. 2). In this recording the linewidth is about 1.3 MHz (in terms of atomic frequency), including the natural width of the $8D$ level of 572 kHz. The broadening is mainly due to the inhomogenous light shift experienced by the atoms through the Gaussian profile of the laser beams. To evaluate the light shift, we record the atomic signal for different laser intensities and we extrapolate the line position to zero light power (Fig. 3). We have followed the same procedure as in Ref. [9]. For each recording, we fit a theoretical profile which takes into account the light shift and the saturation of the transition. Since our previous work, we have improved the calculation of the theoretical line shape: we have included the small hyperfine structure of the $8D$ levels (143 and 222 kHz for $8D_{5/2}$ and $8D_{3/2}$ in hydrogen), the photoionization of the excited levels, as well as the second-order Doppler shift. This effect is evaluated from the velocity distribution of the metastable atoms measured by monitoring the Doppler broadened $2S$-$6P$ transition at 410 nm with a collinear laser beam. We have also measured the population of the hyperfine levels of the $2S_{1/2}$ state (80% and 20% for the $F = 1$ and $F = 0$ levels in hydrogen), which are slightly different from the statistical weights (75% and 25%). Each fit gives both the experimental line center and the line position corrected for light shift and hyperfine structure of the $8D$ levels. An extrapolation of these data is given in Fig. 3. As we measure continuously the beat frequency between the titanium-sapphire laser and the LD/Rb standard laser, we deduce the absolute frequency of the line. Finally, from the linewidth of the $2S$-$15D$ transition, we find the stray electric field to be smaller than 3 mV/cm, corresponding to a Stark shift of about -400 Hz for the $8D_{5/2}$ level and 700 Hz for the $8S_{1/2}$. We have neglected these corrections in the data analysis.

We have studied three transitions in hydrogen and deuterium: $2S_{1/2}(F = 1 \text{ or } \frac{3}{2})$-$8S_{1/2}$, -$8D_{3/2}$, and -$8D_{5/2}$.

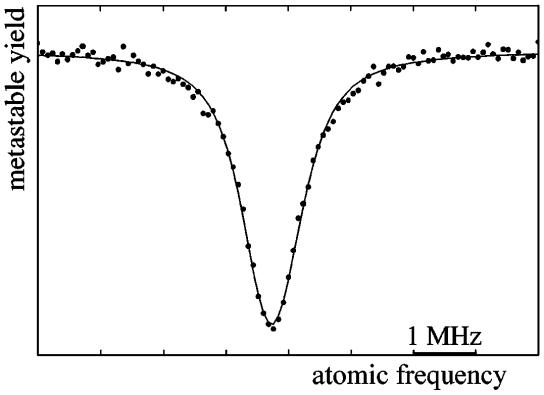

FIG. 2. Hydrogen two-photon spectra of the $2S_{1/2}(F = 1)$-$8S_{5/2}$ transition.

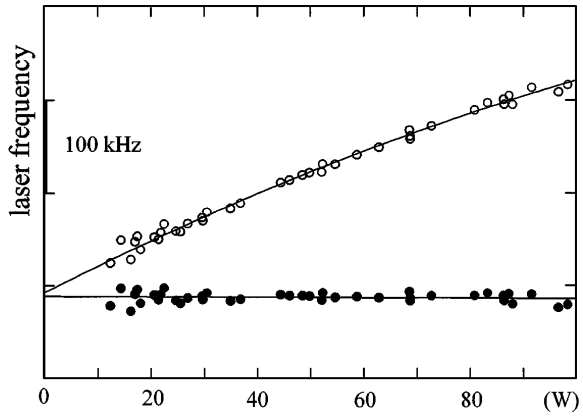

FIG. 3. Extrapolation of the line-center frequency (∘) and of the line position corrected for light-shift and $8D$ hyperfine structure (•) versus the light power in the case of the $2S_{1/2}$-$8D_{5/2}$ transition of deuterium.

In hydrogen, the $2S_{1/2}$-$8D_{5/2}$ transition was measured twice, at the beginning and at the end of the experiment. For each extrapolation we recorded the signal for about forty different light powers. Each signal is the average over ten scans during a 20-min run. We have tried to observe the line for very low light power. In this case we make the mean of several runs to improve the signal-to-noise ratio. In total we have performed 378 runs for data collection. Table I gives the measured frequencies after correction of the hyperfine structure of the S levels. The quoted uncertainties are only statistical. These experimental values can be intercompared using the theoretical value of the fine structure and of the Lamb shifts in the $n = 8$ levels. We have taken into account all the recent calculations of the high-order terms following Ref. [1]. Using $r_p = 0.862$ fm and $r_d = 2.115$ fm for the nucleus charge radii [11,12], the values of the Lamb shifts are $L_{8S_{1/2}} = 16.5008(3)$ MHz, $L_{8D_{3/2}} = -0.0607(2)$ MHz, and $L_{8D_{5/2}} = 0.0714(2)$ MHz [16.5223(3) MHz, −0.0607(2) MHz, and 0.0715(2) MHz, respectively, in deuterium]. We can thus deduce several independent values of the $2S_{1/2}$-$8D_{5/2}$ interval (see Table I). These values are in good agreement for both hydrogen and deuterium. The mean values are, respectively, 770 649 561 585.0(4.9) kHz and 770 859 252 851.5(4.4) kHz. The 4.9 and 4.4 kHz uncertainties (one standard deviation) come from the statistics, the frequency of the LD/Rb laser (2 kHz), the evaluation of the second-order Doppler effect (1 kHz), and the imperfections of the theoretical model (3 kHz).

In hydrogen, there have been several precise measurements of the $2S$ Lamb shift (in fact, the difference $L_{2S\text{-}2P}$ of $2S_{1/2}$ and $2P_{1/2}$ Lamb shifts) by microwave [13] or by optical spectroscopy [14]. This latter method used the $1/n^3$ scaling law of the Lamb shift [15]. Using the weighted mean value of these results [$L_{2S\text{-}2P} = 1057.8400(56)$ MHz] and the theoretical value of the Lamb shifts of the $2P$ and $8D$ levels [$L_{2P} = -12.8356(20)$ MHz], we can extract the Rydberg constant from the hydrogen $2S_{1/2}$-$8D_{5/2}$ interval. The result is $R_\infty = 109\,737.315\,685\,6(11)$ cm^{-1} with an uncertainty of 1 part in 10^{11}. This uncertainty arises mainly from the frequency measurement (6.4×10^{-12}), the Lamb shifts (7.7×10^{-12}), and the proton-to-electron mass ratio (1.3×10^{-12}) [16].

In deuterium, there exists no precise determination of the $2S$ Lamb shift, so we cannot deduce directly R_∞. On the other hand, using the value of R_∞ given above, we deduce the $2S$ Lamb shift $L_{2S\text{-}2P} = 1059.230(9)$ MHz. This value is in good agreement with a previous, less precise determination [1059.240(100) MHz [17]] but disagrees with the theoretical value [1059.210(7) MHz [12]] calculated with the deuteron charge distribution $r_d = 2.115$ fm. From another point of view, we can consider the isotope shift of the $2S$-$8D$ interval. This

TABLE I. Frequencies of he $2S$-$8S/D$ two-photon transitions.

Transition	Measured frequency (MHz)	$2S_{1/2}$-$8D_{5/2}$ deduced frequency (MHz)
Hydrogen		
$2S_{1/2}$-$8D_{5/2}$	770 649 561.5866 (58)	770 649 561.5866
$2S_{1/2}$-$8D_{3/2}$	770 649 504.4535 (66)	770 649 561.5826
$2S_{1/2}$-$8D_{1/2}$	770 649 350.0163 (75)	770 649 561.5784
$2S_{1/2}$-$8D_{5/2}$	770 649 561.5886 (54)	770 649 561.5886
Mean value of the $2S_{1/2}$-$8D_{5/2}$ measurements:		770 649 561.5850 (49)
Deuterium		
$2S_{1/2}$-$8D_{5/2}$	770 859 252.8523 (32)	770 859 252.8523
$2S_{1/2}$-$8D_{3/2}$	770 859 195.7044 (38)	770 859 252.8492
$2S_{1/2}$-$8D_{1/2}$	770 859 041.2511 (52)	770 859 252.8538
Mean value of the $2S_{1/2}$-$8D_{5/2}$ measurements:		770 859 252.8515 (44)

isotope shift is mainly a mass effect. Thanks to the precise determination of the mass ratios m_p/m_e and m_d/m_p, the uncertainty in the mass effect is only 0.6 kHz. As this isotope shift is very sensitive to the nuclear volume effect, we can calculate the difference of the quadratic charge radii of the deuteron and the proton. We obtain $r_d^2 - r_p^2 = 3.827(32)$ fm^2, in good agreement with the result of Weitz *et al.* deduced from the $1S$-$2S$ isotope shift [3.822(16) fm^2 [12]]. Using $r_p = 0.862$ fm, we deduce a value of the deuteron charge distribution $r_d = 2.138(9)$ fm.

If we use the $1/n^3$ scaling law of the Lamb shift [15], we can make another analysis of the measurements in deuterium. If we consider the frequencies $\nu_{1S\text{-}2S}$ and $\nu_{2S\text{-}8D}$ of the $1S$-$2S$ and $2S$-$8D$ intervals, we can form the linear combination $7\nu_{2S\text{-}8D}$-$\nu_{1S\text{-}2S}$. In this way, the theoretically well-known quantity L_{1S}-$8L_{2S}$ appears [15]. If we apply this method to the precise measurements of the $1S$-$2S$ [3,18] and $2S$-$8D$ deuterium frequencies, we obtain $R_\infty = 109\,737.315\,687\,1(22)$ cm^{-1}, in excellent agreement with the hydrogen determination. The weighted mean of the hydrogen and deuterium values is $R_\infty = 109\,737.315\,685\,9(10)$ cm^{-1}. This result, with a relative uncertainty of 9×10^{-12}, is the most precise available. It is in fair agreement with both our result of 1993 [$R_\infty = 109\,737.315\,683\,4(24)$ cm^{-1} [4]] and that of Hänsch and co-workers [$R_\infty = 109\,737.315\,684\,9(30)$ cm^{-1} [12]]. Finally, using the $1S$-$2S$ isotope shift [18], we can also calculate the isotope shift of the quantity $7\nu_{2S\text{-}8D}$-$\nu_{1S\text{-}2S}$, which is independent of the nuclear effects and mainly sensitive to the mass ratio m_p/m_e. In this way we derive a proton-to-electron mass ratio of $m_p/m_e = 1836.152\,68(10)$ in agreement with the far more precise value $m_p/m_e = 1836.152\,666\,5(40)$ obtained by van Dyck *et al.* [16].

To conclude, we have determined the Rydberg constant with an uncertainty of 9×10^{-12}. For the future, thanks to the $1/n^3$ scaling law of the Lamb shifts, the comparison of different optical frequencies provides a new method to reduce further the uncertainties in both the Rydberg constant and the Lamb shifts. In this paper, we have provided a demonstration of this method in the case of deuterium. To exploit the potential of this approach, one needs very precise optical frequency measurements. For this reason, we intend to measure the optical frequencies of the $2S$-$12S/D$ and $1S$-$3S$ transitions with respect to the cesium clock. The comparison of these measurements will reduce the uncertainty in the Rydberg constant to a few parts in 10^{12}.

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[1] See, for example, J. R. Sapirstein and D. R. Yennie, in *Quantum Electrodynamics*, edited by T. Kinoshita (World Scientific, Singapore, 1990); K. Pachucki *et al.*, J. Phys. B **29**, 177 (1996).
[2] See, for example, B. Cagnac, M. D. Plimmer, L. Julien, and F. Biraben, Rep. Prog. Phys. **57**, 853 (1994).
[3] T. Andreae *et al.*, Phys. Rev. Lett. **69**, 1923 (1992).
[4] F. Nez *et al.*, Europhys. Lett. **24**, 635 (1993).
[5] Y. Millerioux *et al.*, Opt. Commun. **108**, 91 (1994).
[6] A. Clairon *et al.*, IEEE Trans. Instrum. Meas. **34**, 265 (1985); Metrologia **25**, 9 (1988).
[7] D. Touahri *et al.*, Opt. Commun. (to be published).
[8] O. Acef *et al.*, Opt. Commun. **109**, 428 (1994).
[9] J. C. Garreau, M. Allegrini, L. Julien, and F. Biraben, J. Phys. **51**, 2263 (1990); **51**, 2275 (1990); **51**, 2293 (1990).
[10] S. Bourzeix, M. D. Plimmer, F. Nez, L. Julien, and F. Biraben, Opt. Commun. **99**, 89 (1993).
[11] G. G. Simon, C. Schmitt, F. Borkowski, and V. H. Walther, Nucl. Phys. **A333**, 381 (1980).
[12] For the value of r_d, see an analysis by M. Weitz *et al.*, Phys. Rev. A **52**, 2664 (1995).
[13] S. R. Lundeen and F. M. Pipkin, Phys. Rev. Lett. **46**, 232 (1981); E. W. Hagley and F. M. Pipkin, Phys. Rev. Lett. **72**, 1172 (1994).
[14] S. Bourzeix *et al.*, Phys. Rev. Lett. **76**, 384 (1996).
[15] S. G. Karshenboim, J. Phys. B **29**, L31 (1996).
[16] D. L. Farnham, R. S. Van Dyck, and P. B. Schwinberg, Phys. Rev. Lett. **75**, 3598 (1995).
[17] B. L. Cosens, Phys. Rev. **173**, 49 (1968).
[18] F. Schmidt-Kaler, D. Leibfried, M. Weitz, and T. W. Hänsch, Phys. Rev. Lett. **70**, 2261 (1993).