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Optical Frequency Measurement of the 2S-12D Transitions in Hydrogen and Deuterium: Rydberg Constant and Lamb Shift Determinations

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We have performed a pure optical frequency measurement of the 2S-12D two-photon transitions in atomic hydrogen and deuterium. From a complete analysis taking into account this result and all other precise measurements (by ourselves and other authors), we deduce optimized values for the Rydberg constant, $R_\infty = 109\,737.315\,685\,16(84)\text{ cm}^{-1}$ (relative uncertainty of 7.7×10^{-12}) and for the 1S and 2S Lamb shifts $L_{1S} = 8172.837(22)\text{ MHz}$ and $L_{2S-2P} = 1057.8446(29)\text{ MHz}$ [respectively, $L_{1S} = 8183.966(22)\text{ MHz}$, and $L_{2S-2P} = 1059.2337(29)\text{ MHz}$ for deuterium]. These are now the most accurate values available. [S0031-9007(99)09458-2]

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For many years, Doppler free two-photon spectroscopy has been applied to the hydrogen atom in order to test quantum electrodynamics calculations and to improve the precision of the Rydberg constant R_∞ [1]. Recently, the uncertainty of the measurements has been reduced to a level below 10^{-11} thanks to optical frequency-multiplication chains, which link the measured frequency via intermediate standard lasers to the caesium clock. With such a chain, Hänsch and co-workers have taken advantage of the small natural width of the 1S-2S two-photon transition (1.3 Hz) to measure this frequency with an uncertainty of 3.4×10^{-13} [2]. In our group, we have made absolute frequency measurements of the 2S-8S/D transitions with an accuracy better than 8×10^{-12} [3]. In this last case, the precision was limited by the line shape analysis which becomes complicated by a large broadening (up to 1 MHz) due to the inhomogeneous light shift. The comparison of the 1S-2S and 2S-8S/D measurements has provided very precise determinations of R_∞ and of the Lamb shift [2,3]. Nevertheless, in order to confirm our 2S-8S/D frequency measurements, we have built a new chain to measure the frequencies of another transition, that is the 2S-12D transition. This transition yields complementary information to our study of the 2S- nS/nD transitions, because it is very sensitive to stray electric fields (the shift due to the quadratic Stark effect varies as n^7), and so such a measurement is a stringent test of Stark corrections to the Rydberg levels. In this Letter, we present these new results and make a complete analysis of the optical frequency measurements to determine the best values for R_∞ and the Lamb shifts.

Our new frequency chain uses two standard lasers, a laser diode stabilized on the 5S-5D two-photon transition of rubidium (LD/Rb laser, $\lambda = 778\text{ nm}$, $\nu = 385\text{ THz}$)

and a CO₂ laser stabilized to an osmium tetraoxyde line (labeled CO₂/OsO₄, $\lambda \approx 10\ \mu\text{m}$, $\nu \approx 29\text{ THz}$). In 1996, the frequencies of three LD/Rb lasers, one in the Laboratoire Kastler Brossel (LKB) and two in the Laboratoire Primaire du Temps et des Fréquences (LPTF), were measured with a frequency chain which connected the LD/Rb laser to the CO₂/OsO₄ standard [4]. More recently, the frequency measurement of this CO₂/OsO₄ standard has been remade with respect to the Cs clock with an uncertainty of 20 Hz (i.e., a relative uncertainty of 7×10^{-13}) [5]. This last measurement corrects the previous one of the CO₂/OsO₄ standard by -87 Hz . With this correction, the frequency of the LD/Rb standard laser working at the LKB is $385\,285\,142\,369.4(1.0)\text{ kHz}$. We have kept a conservative uncertainty of 1 kHz which takes into account the day-to-day repeatability and the long term stability of the LD/Rb standard and the accuracy of the CO₂/OsO₄ standard ($13 \times 20\text{ Hz}$).

The frequency gap between the 2S-12D lines ($\lambda \approx 750\text{ nm}$, $\nu \approx 399.5\text{ THz}$) and the LD/Rb standard is almost equal to half of the CO₂/OsO₄ standard frequency. We have built an optical frequency divider to reduce this frequency by a factor of 2 [2,6]. The different parts of the experiment are carried out simultaneously at the LKB and LBTF (see Fig. 1). The two laboratories are linked by two, 3 km long, optical fibers which are used, via a phase coherent chain, to transfer the CO₂/OsO₄ frequency reference from LPTF to LKB. The frequency shift introduced by these optical fibers is at most 3 Hz [7]. We use an auxiliary laser at 809 nm ($\nu \approx 370.5\text{ THz}$) and the laser frequencies satisfy the equations,

$$\nu(2S-12D) + \nu(809) = 2\nu_{\text{Rb}},$$

$$\nu(2S-12D) - \nu(809) = \nu(\text{CO}_2).$$

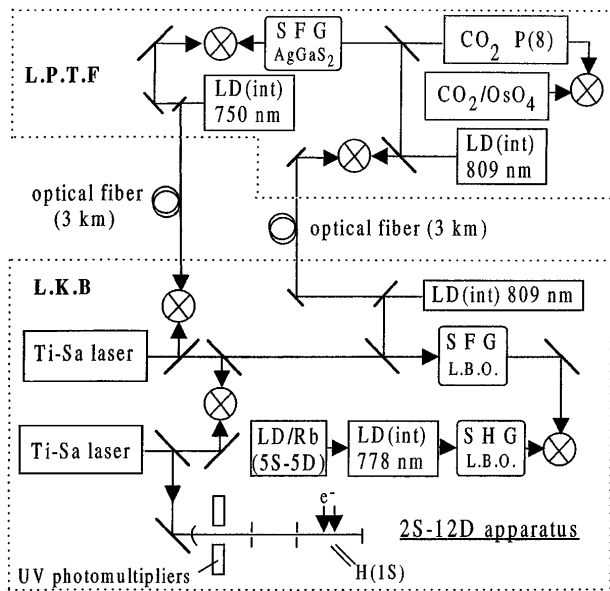


FIG. 1. Outline of the frequency chain between the $2S$ - $12D$ hydrogen frequencies and the LD/Rb and CO_2/OsO_4 standards. The details are explained in the text.

A laser diode (power of 50 mW) is injected by the LD/Rb standard at 778 nm and frequency doubled in a LiB_3O_5 (LBO) crystal placed in a ring cavity. The generated UV beam is frequency compared to the frequency sum (made also in an LBO crystal) of the 750 and 809 nm radiations produced by a first titanium-sapphire laser (about 300 mW) and a laser diode (about 30 mW). One part of the 809 nm radiation is sent via one fiber to the LPTF. There, a 809 nm local laser diode is phase locked on the LKB one. A frequency sum of this 809 nm laser diode and of an intermediate CO_2 laser in an AgGaS_2 crystal generates a wave at 750 nm. This wave is used to phase lock a laser diode at 750 nm which is sent back to the LKB by the second fiber. In such a way, the two equations are simultaneously satisfied and all the frequency counting is performed in the LKB. A second titanium-sapphire laser induces the two-photon transitions. For the hydrogen measurements, the CO_2 intermediate laser uses the $P(8)$ line [respectively, $\text{CO}_2 R(4)$ line for deuterium], and the residual frequency difference between the two titanium-sapphire lasers is about 2.5 GHz (respectively, 41.3 GHz for deuterium). These frequency beat notes are detected with a photodiode or a Schottky diode.

The hydrogen experiment has been described elsewhere [3,8]. To reduce the collisional and transit time broadening, the two-photon transitions are induced in a metastable atomic beam collinear with the laser beams. We use a highly stable titanium-sapphire laser with a frequency jitter and a long term stability of a few kHz [9]. Thanks to an enhancement cavity surrounding the atomic beam, the optical power can be as much as 50 W in each direc-

tion with a beam waist of $646 \mu\text{m}$. At the end of the atomic beam, an electric field quenches the metastable atoms and we detect the Lyman- α fluorescence to measure the metastable yield. As the excited $12D$ states decay preferentially to the $1S$ ground state in a proportion of 95%, the optical excitation can be detected via the corresponding decrease of the $2S$ beam intensity. The acquisition and analysis of the experimental data follow the same procedure as in Ref. [8]. To evaluate the light shift, which is the major source of shift and broadening, we record the atomic signal for different laser intensities, and we extrapolate the line position to zero light power. For each recording, a theoretical profile is fitted to the experimental curve. This theoretical line shape takes into account the light shift, the saturation of the transition, the small hyperfine structure of the $12D$ levels, the photoionization, the small deviation of the atomic trajectories due to the light forces, as well as the second-order Doppler shift. The velocity distribution is measured by monitoring the Doppler shifted $2S$ - $6P$ transition. Each fit gives both the experimental line center and the line position corrected for light shift and hyperfine structure of the $12D$ level. As we measure continuously the various beat frequencies of our frequency chain, we can deduce the absolute frequency of the line. Finally, the result is corrected for the shift due to black-body radiation (2.1 kHz for the $12D$ levels at 330 K [10]) and for the Stark shift due to stray electric fields. To evaluate these fields, we have studied the $2S$ - $20D$ transitions. In this case, the broadening due to the linear Stark effect is large (it varies as n^2) and, from the line shape of the $2S$ - $20D$ transitions, we deduce a mean value of the stray electric fields of 2.0 (1.0) mV/cm. A careful analysis of the Stark effect, including the mixing of the quasidegenerate levels ($D_{5/2}$ - $F_{5/2}$ and $D_{3/2}$ - $P_{3/2}$), gives a Stark shift of -2.1 (1.2) kHz and -6.0 (4.9) kHz for the $12D_{5/2}$ and $12D_{3/2}$ levels.

We have studied the $2S_{1/2}(F=1 \text{ or } 3/2)$ - $12D_{5/2}$ and $2S_{1/2}(F=1 \text{ or } 3/2)$ - $12D_{3/2}$ transitions in hydrogen and deuterium. For each transition, the atomic signal is recorded for at least 50 light powers, and, in total, we have used 237 runs (20 min long) for data collection. The measured frequencies (after correction of the hyperfine structures) are reported in Table I. The quoted uncertainties are due only to the statistics. The two experimental results for the $12D_{5/2}$ and $12D_{3/2}$ levels can be compared by taking into account the theoretical value of the fine structure and the Lamb shift of the $12D$ levels. Following recent calculations of the Bethe logarithm [11], these Lamb shifts are $L_{12D_{3/2}} = -17.6(2)$ kHz and $L_{12D_{5/2}} = 21.5(2)$ kHz in hydrogen and deuterium. Finally, we obtain two independent values of the $2S_{1/2}$ - $12D_{5/2}$ interval (see Table I) which are in fair agreement for hydrogen and deuterium. The average values are, respectively, $799\,191\,727\,402.8(6.7)$ kHz and $799\,409\,184\,967.6(6.5)$ kHz. These uncertainties (1 standard deviation) are due to the statistical error, the

TABLE I. Frequencies of the $2S$ - $12D$ two-photon transitions.

Transition	Measured frequency (MHz)	$2S_{1/2}$ - $12D_{5/2}$ Deduced frequency (MHz)
Hydrogen		
$2/S_{1/2}$ - $12D_{5/2}$	799 191 727.4037 (47)	799 191 727.4037
$2S_{1/2}$ - $12D_{3/2}$	799 191 710.4727 (62)	799 191 727.3999
Mean value of the $2S_{1/2}$ - $12D_{5/2}$ measurements:		799 191 727.4028 (67)
Deuterium		
$2/S_{1/2}$ - $12D_{5/2}$	799 409 184.9668 (45)	799 409 184.9668
$2S_{1/2}$ - $12D_{3/2}$	799 409 168.0380 (44)	799 409 184.9698
Mean value of the $2S_{1/2}$ - $12D_{5/2}$ measurements:		799 409 184.9676 (65)

Stark effect (2.1 kHz), the second-order Doppler effect (1 kHz), the stability and the measurement of the LD/Rb standard laser (2 kHz), and the imperfections of the theoretical model (4.5 kHz). These measurements are slightly less precise than our previous ones of the $2S$ - $8S/D$ transitions [3,12], because of the smaller signal-to-noise ratio and the larger quadratic Stark effect.

Table II gives the values of the Rydberg constant deduced from different transitions by several methods. It is convenient to express an energy level in hydrogen as the sum of two terms: the first, given by the Dirac equation and by the first relativistic correction due to the recoil of the proton, is known exactly, apart from the uncertainties in the physical constants involved (mainly R_∞). The second term is the Lamb shift, which contains all the other corrections, i.e., the QED corrections, the other relativistic corrections, and the effect of the proton charge distribution. Consequently, to extract R_∞ from the accurate measurements one needs to know the Lamb shifts. For this analysis, the theoretical values of the Lamb shifts are sufficiently precise, except for those of the $1S$ and $2S$ levels. In hydrogen, there have been several precise determinations of the $2S_{1/2}$ - $2P_{1/2}$ splitting

by microwave spectroscopy [13] and by the anisotropy method [14]. Using the mean value of these results [$L_{2S-2P} = 1057.8454(65)$ MHz], we can deduce from our $2S_{1/2}$ - $12D_{5/2}$ determination a value of the Rydberg constant $R_\infty = 109\,737.315\,684\,5(13)$ cm^{-1} . The uncertainty (1.2×10^{-11}) comes from the frequency measurement (8.4×10^{-12}), the $2S$ Lamb shift (8.1×10^{-12}), and the proton-to-electron mass ratio (1.3×10^{-12}) [15]. To compare our present result with our earlier one [3], Table II gives the Rydberg constant value deduced from the $2S$ - $8D$ measurement with the same procedure. These two values are in acceptable agreement (they differ by about 1 standard deviation). Although slightly less precise, our new result confirms our previous one and shows that the correction due to the quadratic Stark effect is well analyzed (this correction is 17 times larger for the $12D$ than for the $8D$ levels). Table II gives the average of these two results with an uncertainty of 10^{-11} . This result is the most precise if we do not make theoretical assumptions concerning the $1S$ and $2S$ Lamb shifts.

The other methods to determine R_∞ use the $1/n^3$ scaling law of the Lamb shift which gives the theoretical value of the linear combination of the Lamb shifts

TABLE II. Determination of the Rydberg constant R_∞ .

Method and transitions involved	$(R_\infty - 109737)$ cm^{-1}
Determination of R_∞ from the $2S$ - nD and $2S$ - $2P$ measurements	
$2S$ - $12D$ and $2S$ - $2P$ in hydrogen	0.315 6845 (13)
$2S$ - $8D$ and $2S$ - $2P$ in hydrogen	0.315 6858 (13)
$2S$ - $12D$, $2S$ - $8D$, and $2S$ - $2P$ in hydrogen	0.315 6852 (11)
Determination of R_∞ without the $2S$ - nS/D measurements	
$1S$ - $2S$, $2S$ - $2P$, and $1/n^3$ law in hydrogen	0.315 6854 (20)
Determination of R_∞ from linear combination of optical frequencies measurements	
$2S$ - $12D$, $1S$ - $2S$, and $1/n^3$ law in hydrogen	0.315 6838 (17)
$2S$ - $12D$, $1S$ - $2S$, and $1/n^3$ law in deuterium	0.315 6838 (16)
$2S$ - $12D$, $1S$ - $2S$, and $1/n^3$ law in hydrogen and deuterium	0.315 6838 (13)
$2S$ - $8D$, $1S$ - $2S$, and $1/n^3$ law in hydrogen and deuterium	0.315 6861 (12)
$2S$ - $8D$, $2S$ - $12D$, $1S$ - $2S$, and $1/n^3$ law in hydrogen and deuterium	0.315 6850 (10)
General least squares adjustment in hydrogen and deuterium	0.315 685 16 (84)

$L_{1S}-8L_{2S}$ [16]. Using this relation and the measurements of the $2S$ Lamb shift, it is possible to extract a value of R_∞ from the very precise measurement of the $1S-2S$ transition [2]. The result, independent of our $2S-nD$ measurements, is given in the second part of Table II and agrees perfectly with the value deduced from the $2S-nD$ frequencies.

Finally, in the third part of Table II, we give the values of R_∞ obtained from the linear combination of several optical frequencies. For example, if we consider the frequencies ν_{1S-2S} and ν_{2S-12D} of the $1S-2S$ and $2S-12D$ intervals, we can form the linear combination $7\nu_{2S-12D}-\nu_{1S-2S}$, where the theoretically well-known quantity $L_{1S}-8L_{2S}$ appears. This method is independent of the microwave measurements of the $2S$ Lamb shift and is relevant for both hydrogen and deuterium. If we consider the $1S-2S$ and $2S-12D$ transitions, the values obtained for hydrogen and deuterium are in excellent agreement (see Table II). With a least squares procedure, it is possible to apply this method to several transitions. If we use all the precise optical frequency measurements in hydrogen and deuterium (transitions $1S-2S$, $2S-8D$, and $2S-12D$), we obtain a value of R_∞ more precise than the previous ones. This method also provides very accurate determination of the $1S$ and $2S$ Lamb shifts, $L_{1S} = 8172.834(26)$ MHz and $L_{2S-2P} = 1057.8442(34)$ MHz [respectively, $8183.963(26)$ MHz and $1059.2333(34)$ MHz for deuterium]. This result for the $2S$ Lamb shift is independent and more precise than the direct determinations made by microwave spectroscopy.

To make an average of these various determinations of R_∞ , we have performed a least squares adjustment which takes into account the measurements of the $2S$ Lamb shift [13,14], the optical frequency measurements of the $1S-2S$ [2], $2S-8D$ [3], and $2S-12D$ transitions in hydrogen and deuterium, and also the measurements of the $1S$ Lamb shift made by frequency comparison of the $1S-2S$ and $2S-4S/P/D$ transitions [17,18] or of the $1S-3S$ and $2S-6S/D$ ones [19]. We obtain the values $R_\infty = 109\,737.315\,685\,16(84)$ cm⁻¹, $L_{1S} = 8172.837(22)$ MHz, and $L_{2S-2P} = 1057.8446(29)$ MHz [respectively, $8183.966(22)$ MHz and $1059.2337(29)$ MHz for deuterium]. These values, which take into account all the most recent results, are the most precise to date. The $1S$ Lamb shift value is in poor agreement with the more recent calculation in hydrogen [$L_{1S} = 8172.731(40)$ MHz with the proton radius $r_p = 0.862(12)$ fm [20]]. Assuming the validity of these QED calculations, we deduce $r_p = 0.900(16)$ fm.

To conclude, we have made an optical frequency measurement of the $2S-12D$ transition in hydrogen and deuterium. This result confirms our previous work on the $2S-8S/D$ transitions and, furthermore, we have reduced the uncertainty in the Rydberg constant to less than $7.7 \times$

10^{-12} . We present also an exhaustive analysis of the most accurate measurements in hydrogen and deuterium which shows that the optical frequency measurements have superseded the microwave determination of the $2S$ Lamb shift. The precision is now limited by the uncertainties of the $2S-nD$ frequencies.

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