New Measurement of the Rydberg Constant by Two-Photon Spectroscopy of Hydrogen Rydberg States

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We have made wavelength measurements of the three transitions 2S-8D, 2S-10D, and 2S-12D in atomic hydrogen and deuterium to determine a new value for the Rydberg constant, $R_\infty = 109\,737.315\,709(18)\ \mathrm{cm}^{-1}$, currently the most precise one. As reference we made use of an I_2-stabilized He-Ne laser, compared to the standards of the Bureau International des Poids et Mesures. The H-D isotopic shifts have also been measured and a value for the proton-to-electron mass ratio deduced: $m_p/m_e = 1836.152\,59(24)$.

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Precise knowledge of the Rydberg constant is important in many areas of physics and chemistry.[1] Several measurements of R_∞ have been made using high-resolution spectroscopy of different H and D transitions in the optical region and the most recent values[2–6] are in agreement. Here we report a new measurement of the Rydberg constant with an absolute accuracy of 1.7×10^{-10} and a precision relative to the standard laser of 4.3×10^{-11}. It is therefore the most precise measurement to date and, in fact, limited by the uncertainty in the realization of the wavelength and frequency standard in the optical domain (1.6×10^{-10}).

Our method is based on the measurements of the wavelengths of Doppler-free two-photon $2S_{1/2}$-$nD_{5/2}$ ($n \geq 8$) transitions in atomic hydrogen and deuterium.[2] These transitions permit R_∞ to be measured with higher precision than the one-photon $2S$-$3P$ and $2S$-$4P$ transitions used in other experiments,[3,4] because of the small natural widths of the upper levels (for example, ≈ 296 kHz for the $10D$ level). From this point of view the $1S$-$2S$ two-photon transition, with a natural linewidth of 1.3 Hz, seems the most favorable; but, since only the $2S$ Lamb shift has been measured with high enough precision,[7,8] the $1S$-$2S$ frequency provides a measurement of the $1S$ Lamb shift[5,6] rather than an independent value of the Rydberg constant.

In a previous paper,[2] we described the experimental setup used to perform a preliminary Rydberg constant measurement. For the work reported here, we have improved several parts of this basic apparatus. A thorough description of these improvements will be presented in a forthcoming paper.[9] To obtain very narrow linewidths we have developed a novel H($2S$) metastable beam source. The beam is collinear with two counterpropagating laser beams, a geometry that produces long laser-atom interaction times and then reduces line broadening. To enhance the two-photon absorption, the atomic beam is placed inside a Fabry-Perot cavity so that the atoms are subjected to a standing wave. The laser intensity is up to 50 W for each traveling wave and the beam waist is $w_0 \approx 600\ \mu$m. The optical excitation region is carefully shielded against stray electric and magnetic fields. The metastable beam is collimated by two apertures, 7 mm in diameter and 96 cm apart. After optical excitation, the nD states undergo a radiative cascade to the $1S$ state in a proportion of $\approx 95\%$ and we monitor the two-photon transitions by measuring the corresponding decrease in the $2S$ beam intensity. The $2S$ atoms are detected by applying a quenching electric field to mix the $2S$ with the $2P$ level and by measuring the resulting Lyman-α emission.

The laser used to excite the $2S$-nD transitions is a homemade, cw (≈ 1 W), single-mode (≈ 50-kHz jitter), ring dye (LD700) laser, pumped by a Kr^+ laser. Compared to our previous experiment,[2] two major improvements of our setup are an acousto-optic modulator used to precisely scan the dye-laser frequency in a reproducible way over a 250-MHz range centered at any desired frequency, and a microcomputer to monitor the frequency scanning and the data acquisition.

Figure 1 shows a recording of the metastable-atom

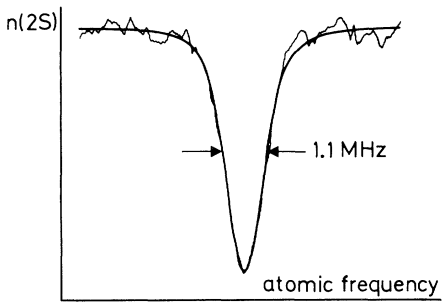

FIG. 1. Recording of the $2S_{1/2}(F=1)$-$10D_{5/2}$ two-photon transition profile in H, observed as a decrease of the metastable beam intensity $n(2S)$. The fit of the experimental profile with the calculated profile is also shown.

beam intensity as the laser wavelength is scanned through the $2S_{1/2}(F=1)$-$10D_{5/2}$ transition of the hydrogen atom (the signal, averaged over ten scans, is acquired during a 20-min run). The signal amplitude corresponds to a 9.4% decrease in the metastable beam intensity. The observed linewidth (in terms of total two-photon transition frequency) is 1.1 MHz, to be compared with the 296-kHz natural width. There are several possible causes for the broadening and shift of the signal:

(i) *The laser linewidth* is responsible for ≈ 100-kHz broadening.

(ii) *The second-order Doppler effect* both broadens and shifts the line. In order to evaluate the contribution of this velocity-dependent effect, we have measured the longitudinal velocity distribution of the metastable atoms in the beam by monitoring the absorption profile of the one-photon Balmer-α transition. The mean measured velocity is 3.05×10^3 m/s in hydrogen and 1.97×10^3 m/s in deuterium. For the transition of Fig. 1 the second-order Doppler effect shifts the line frequency to the red by 41 kHz and broadens it by ≈ 70 kHz.

(iii) *The finite transit time* causes broadening of ≈ 10 kHz for a hydrogen atom traveling with the mean velocity along a trajectory tilted at maximum angle with respect to the laser beams.[10]

(iv) *Saturation and light shift,* both power dependent, are the major sources of broadening and shifting. For an atom traveling exactly along the laser-beam axes, and for a one-way 50-W laser power, the excitation rate is $\approx 1.7\times10^5$ s^{-1} while the transit time is 3.3×10^{-4} s. The saturation is therefore large for this trajectory, while it can be small for trajectories which are parallel to it but offset with respect to the laser-beam axes. The light shift for an atom in the center of the laser beams is 560 kHz. Because the laser power seen by each atom depends on its trajectory and varies along it, the light shift also contributes to the line broadening. To account precisely for the combined effect of natural lifetime, light shift, and saturation we have performed calculations of the line profiles, taking into account all possible trajectories of metastable atoms. Details of this treatment will be given in Ref. 9. For the line of Fig. 1 we have calculated a 326-kHz shift and a 846-kHz width.

(v) *The contributions of other minor effects* (such as Stark and Zeeman effects due to residual stray fields) are not easily isolated. We can estimate the Stark effect because our observation of the $2S$-$20D$ transition gives an upper limit of 2 mV/cm for the residual electric field. Compared to our preliminary setup[2] where the stray electric field was ≈ 30 mV/cm, the observed improvement is due to the use of cryogenic pumps and of a new heated vacuum chamber. A residual 2-mV/cm field would cause a ≈ 100-kHz broadening and a ≈ 1-kHz Stark shift in the $2S$-$10D$ transition.

We have considered all possible broadening effects not taken into account in the above calculation [i.e., effects (i)–(iii) and (v)] by convoluting the line profile with a Gaussian curve. An example of a fit made with the theoretical line profiles obtained after this convolution is shown in Fig. 1. The fit parameters are line position, light power, and Gaussian broadening. We have studied the three $2S_{1/2}$-$nD_{5/2}$ ($n=8,10,12$) transitions in hydrogen and deuterium. For each transition, we have recorded 56 experimental line profiles for various laser powers. Each fit gives both the experimental line center (half-maximum center) and the light-shift–corrected line position. A careful study of these two series of data has been made and it has been checked that the extrapolation of the line center to zero light power gives a result consistent with the light-shift–corrected line position.

While the 1.1-MHz linewidth shown in Fig. 1 is typical of our data, we have measured linewidths as narrow as 700 kHz ($2S$-$12D$ transition), i.e., relative linewidth of 9×10^{-10}, which, to our knowledge, is the highest resolution so far achieved for an optical transition in hydrogen.

The transition wavelength is measured by interferometric techniques based on comparison of our dye-laser wavelength with an I_2-stabilized He-Ne laser at 633 nm. The key element for this comparison is nonconfocal Fabry-Perot etalon built with two silver-coated mirrors in optical adhesion on a Zerodur rod. We measure the Fresnel phase shift due to the wave-front curvature through the frequency interval between the TEM_{00} and TEM_{01} modes inside the cavity. In order to determine this Fresnel phase shift with a better accuracy than

TABLE I. Experimental determination of the $2S_{1/2}$-$nD_{5/2}$ transition frequency: example of the $n=8$ levels.

	Hydrogen	Deuterium
Resonance frequency (MHz)	385 324 758.668	385 429 619.684
Resonance frequency ×2	770 649 517.336	770 859 239.368
Second-order Doppler effect (MHz)	+0.040	+0.017
$2S_{1/2}$ hyperfine splitting (MHz)	+44.389	+13.641
$8D_{5/2}$ hyperfine splitting (MHz)	−0.028	−0.008
$2S_{1/2}$-$8D_{5/2}$ experimental value of the energy splitting (MHz)	770 649 561.737	770 859 253.018

TABLE II. Determination of the Rydberg constant.

Transition	Measured frequency (MHz)	R_∞ (cm^{-1})
H: $2S_{1/2}$-$8D_{5/2}$	770 649 561.737(49)	109 737.315 707 6(70)
D: $2S_{1/2}$-$8D_{5/2}$	770 859 253.018(43)	109 737.315 708 6(61)
H: $2S_{1/2}$-$10D_{5/2}$	789 144 886.593(35)	109 737.315 708 4(48)
D: $2S_{1/2}$-$10D_{5/2}$	789 359 610.420(34)	109 737.315 707 3(47)
H: $2S_{1/2}$-$12D_{5/2}$	799 191 727.573(47)	109 737.315 707 8(64)
D: $2S_{1/2}$-$12D_{5/2}$	799 409 185.184(42)	109 737.315 713 2(58)
Final result		109 737.315 708 8(47)

in our previous experiment,[2] we have carried out a very careful study[11] of this mode-frequency interval versus cavity optical alignment, so that effects of mirror curvature imperfections[12] are removed. The reflective phase shifts in the mirror coatings are eliminated by the method of virtual mirrors[13]: We use two rods of different lengths (first 10 cm, then 50 cm, and finally 10 cm again) and we take into account the slight aging of the mirrors due to the short air exposures during the two changes of the Fabry-Perot length.

Our reference laser has been compared to the standard lasers of the Bureau International des Poids et Mesures (BIPM), via an intermediate standard He-Ne laser of the Institut National de Métrologie. The frequency of our reference laser relative to the BIPM standard is known with a precision of 10^{-11}. Therefore, we may assume for our absolute frequency reference the precision of the primary wavelength and frequency standard in the visible (1.6×10^{-10}).[14]

For each of the six studied transitions we have corrected the measured frequency for the second-order Doppler effect and for hyperfine splittings.[15] Table I gives details of these corrections for the $n=8$ levels. We have used the experimental value of the hydrogen $2S$ Lamb shift[8] and the theoretical value (1057.229 MHz) of the deuterium one, calculated from Ref. 16, to deduce the $2P_{1/2}$-$nD_{5/2}$ frequencies. We have then compared these frequencies to those reported by Erickson[17] (corrected for the 1986 recommended values of the fine-structure constant, the proton/electron, and the deuteron/proton mass ratios[18]), using the Rydberg constant as a scaling factor. The measured frequencies lead to six independent determinations of the Rydberg constant reported in Table II. The quoted errors do not include the reference-laser uncertainty. As may be seen from this table our six independent measurements of R_∞ are quite self-consistent. Taking into account the reference-laser uncertainty, our final result is $R_\infty = 109\,737.315\,709(18)$ cm^{-1}. The precision is 1.7×10^{-10} and only 4.3×10^{-11} with respect to our reference laser (see Table III). In this table, the first two items do not come from our experiment and give the present limitation to the method. The uncertainty of the theoretical line shape takes into account the dependence of the fit results on the spatial distribution of metastable atomic trajectories. The statistical error is the standard deviation of our six independent measurements. The mirror-aging error is deduced from the observed drift between the two sets of measurements with the 10-cm spacer. Compared to Ref. 2, the

TABLE III. Error budget (parts in 10^{11}).

Electron-to-proton mass ratio	1.1
Lamb-shift and energy-level calculations	1.5
Stark effect	0.5
Second-order Doppler effect	0.5
Uncertainty of the theoretical line shape	2.0
Statistical	2.0
Aging of the mirrors	2.0
Fresnel phase-shift measurement	1.2
Comparison between the He-Ne lasers	1.0
Root-mean-square error	4.3
Standard laser	16.0
Final error	16.6

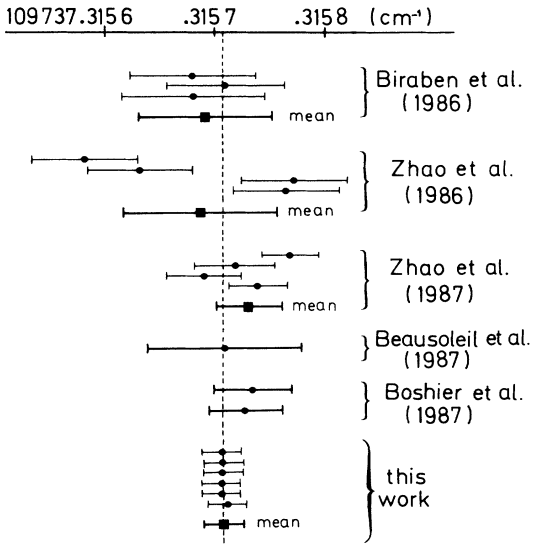

FIG. 2. Comparison of our result with recent high-precision measurements of the Rydberg constant.

improvements in these two items are due to the new scanning system of the laser frequency and to the better precision in the Fresnel phase-shift measurement. In Fig. 2, our results are compared to our previous one[2] and to those obtained by different groups since 1986 from cw excitation either of the Balmer-α and -β transitions[3,4] or of the $1S$-$2S$ transition.[5,6]

Our experiment also provides three H-D isotopic shifts from which we can deduce the proton/electron mass ratio: $m_p/m_e = 1836.15259(24)$, in excellent agreement with the more precise measurement $m_p/m_e = 1836.152701(37)$ of Van Dyck *et al.*[19] The uncertainty of our measurement is the rms sum of the following contributions: statistical (1.1×10^{-7}), proton/deuteron mass ratio (3×10^{-9}),[18] nuclear size (6.3×10^{-8}),[16,17] and second-order Doppler effect (2×10^{-8}).

In conclusion, we have made a new measurement of the Rydberg constant that is not limited by uncertainties inherent in the experiment, but rather by the precision of the wavelength and frequency standard in the visible range. In fact, our result shows that the present realization of the meter is no longer satisfactory in the optical domain and that more precise optical frequency standards are required.

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