

## Determination of the Rydberg Constant by Doppler-Free Two-Photon Spectroscopy of Hydrogen Rydberg States.

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**Abstract.** – Using the Doppler-free two-photon absorption technique, we observe the  $2S-8D$  and  $2S-10D$  transitions in atomic hydrogen and deuterium with a relative line width of  $1.8 \cdot 10^{-9}$ . The comparison of the wave-length of these transitions to the one of an iodine stabilized He-Ne laser provides a new determination of the Rydberg constant  $R_{\infty} = 109\,737.315\,69(6) \text{ cm}^{-1}$ . This measurement increases the precision on the Rydberg constant by a factor of two and gives a result slightly different from the preceding ones. A precise measurement of the  $2S-8D$  isotope shift is also performed.

### 1. Introduction.

The development of Doppler-free laser techniques has allowed substantial improvement of the resolution observed in optical spectroscopy. In recent years, these techniques have been applied to atomic hydrogen in order to increase the precision on the Rydberg constant  $R_{\infty}$ . A first method consists in studying the Balmer  $\alpha$  line and in deducing the Rydberg constant from wave-length measurements either of the  $2P-3D$  or of the  $2S-3P$  lines [1-3]. Up to now the most precise determination of  $R_{\infty}$  has been obtained with this method using a metastable atomic beam [3]. This measurement was limited to a precision of  $10^{-9}$  because of the natural width of the  $3P$  level (30 MHz).

Another method consists in studying the  $1S-2S$  two-photon transition whose natural line width is only 1.3 Hz. This transition has been induced with c.w. radiation and a resolution of  $5 \cdot 10^{-9}$  has been observed [4]. However, the two recently published measurements of the  $1S-2S$  wave-length [5, 6] have been performed with pulsed excitation so that the experimental relative line width is much larger. Up to now, the precision on the Rydberg constant deduced from the  $1S-2S$  transition experiment is then  $2 \cdot 10^{-9}$  only [5]. We note that the obtained value seems in slight disagreement with the value measured in [3]. Moreover,

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as the Lamb shift of the  $1S$  level is not experimentally known, the mere measurement of the  $1S$ - $2S$  frequency gives this Lamb shift rather than the Rydberg constant.

Our own way to measure the Rydberg constant is to study the Doppler-free two-photon  $2S$ - $nD$  transition ( $n \geq 8$ ) [7]. A first advantage of this method is that the  $2S$  Lamb shift has been measured with a high precision [8, 9]. Secondly, the natural widths of the Rydberg levels are small. The  $8D$  line width is, for example, 550 kHz and can then lead to a relative line width of  $7 \cdot 10^{-10}$ . In our experiment, a preliminary result gave a relative line width of  $1.8 \cdot 10^{-9}$  for the  $2S$ - $8D$  two photon line [10]. In the present paper we give a first Rydberg measurement on the  $2S$ - $8D$  and  $2S$ - $10D$  two-photon lines.

## 2. Observation of the $2S$ - $nD$ transitions.

The experimental set-up has already been described [7]. We induce the  $2S$ - $nS$  and  $2S$ - $nD$  Doppler-free two-photon transitions using a metastable atomic beam which is collinear with two counterpropagating laser beams. The line broadening due to the finite transit time of atoms in the laser beams is then very small.

The experimental set-up is shown in fig. 1. The metastable atomic beam is produced in two steps: molecular hydrogen is dissociated by a RF discharge. An effusing atomic beam flows into a first vacuum chamber where it is excited in the  $2S$  state by electronic bombardment. Because of the inelastic collisions with electrons, the metastable atomic beam makes an angle of  $20^\circ$  with the incident atomic beam and can thus be aligned with the two laser beams. The optical excitation takes place in the second vacuum chamber where electric

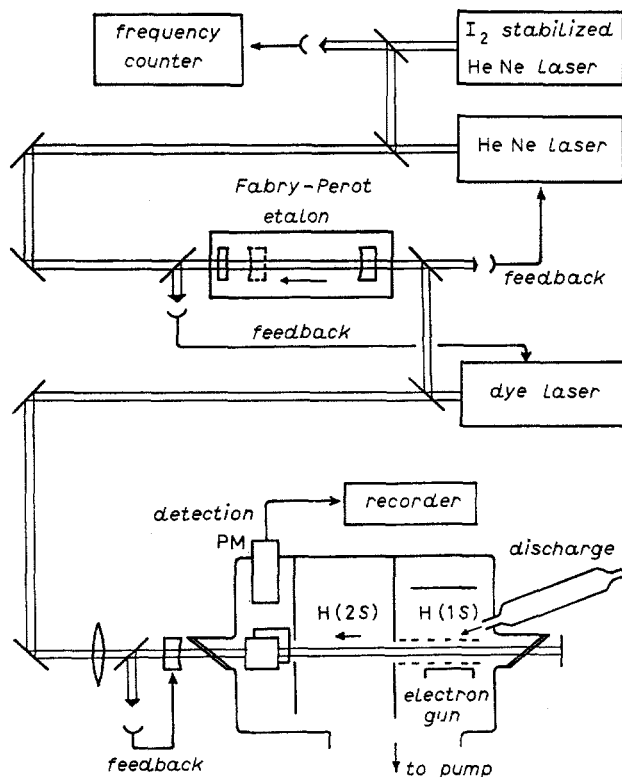


Fig. 1 - Experimental set-up.

and magnetic fields are reduced at best. The metastable atoms are detected in the third vacuum chamber. An applied electric field quenches the  $2S$  state and a photomultiplier detects the Lyman  $\alpha$  fluorescence. Measuring the photomultiplier current, we estimate the metastable beam intensity to be about  $10^7$  atoms  $s^{-1}$ .

The excitation beam is provided by a home-made c.w. ring LD700 dye laser [11]. At 778 and 760 nm (approximate wave-lengths of the  $2S$ - $8D$  and  $2S$ - $10D$  transitions) it can provide a power of 1 W. By locking it in an external Fabry-Perot cavity, we obtain a laser line width of about 150 kHz. To efficiently induce the two-photon transitions, the metastable atomic beam is placed inside a Fabry-Perot cavity [12]. The cavity length (50 cm) is locked on the laser frequency of monitoring the reflected beam polarization [13]. Inside the cavity the beam waist  $w_0$  is 570  $\mu\text{m}$  and the light power is about 40 W in each propagation direction.

After a two-photon excitation from the  $2S$  metastable state, the  $nD$  states undergo radiative cascade to the  $1S$  ground state in a proportion of about 90%. The two-photon transition can then be detected by observing the corresponding decrease of the  $2S$  beam intensity. In fig. 2 a typical recording shows the  $2S$ - $8D$  two-photon transition. The largest signal amplitude ( $2S_{1/2}(F=1) \rightarrow 8D_{5/2}$ ) corresponds to a 10 per cent decrease of the metastable beam intensity. The experimental line width (in terms of total two-photon transition frequency) is about 1.4 MHz. It corresponds to a relative line width of  $1.8 \cdot 10^{-9}$ . This result has to be compared with the natural width of the  $8D_{5/2}$  level which is 550 kHz. There are several reasons for the broadening and shift of the line:

i) *Second-order Doppler effect.*

For an atomic beam of 3.2 km/s mean velocity, the second-order Doppler effect decreases the line frequency by 44 kHz and broadens it by about 60 kHz.

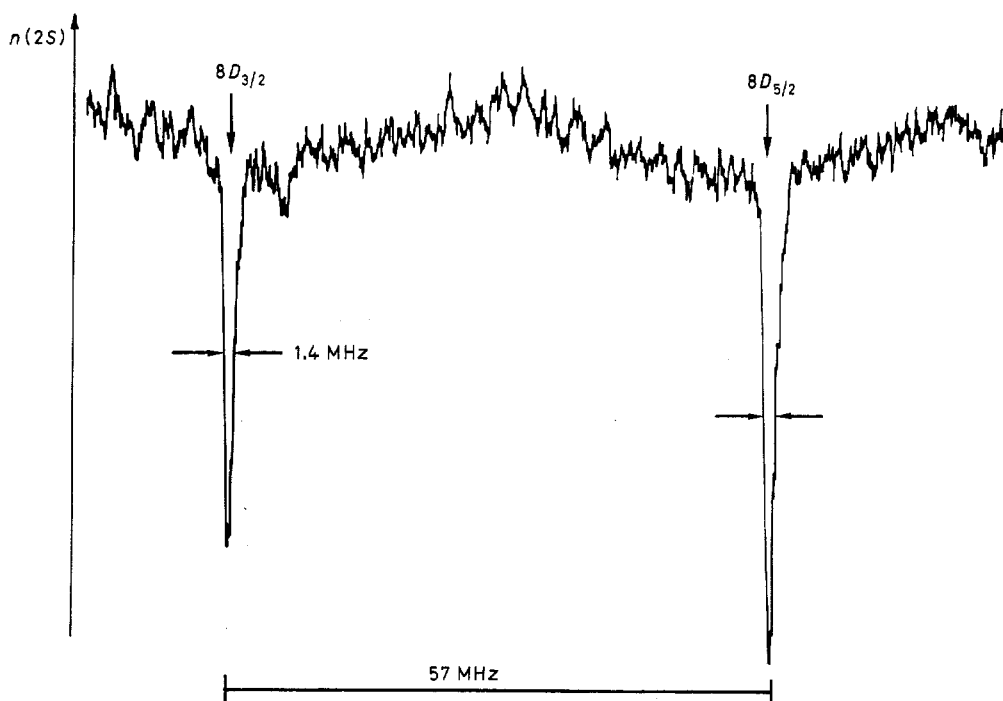


Fig. 2. - Recording of the  $2S_{1/2}(F=1)$ - $8D_J$  two-photon transition in hydrogen;  $n(2S)$  is the metastable beam intensity. The  $2S_{1/2} \rightarrow 8D_{5/2}$  line corresponds to a ten per cent decrease of  $n(2S)$ . The frequency scale is in term of the total two-photon frequency.

ii) *Finite transit time* [14, 15].

For atomic trajectories making the largest possible angle with respect to the laser beam, the line-broadening would be 90 kHz.

iii) *Saturation of the two-phonon excitation.*

For a metastable atom travelling along the beams axis, the excitation rate is about  $1.3 \cdot 10^5 \text{ s}^{-1}$  (for a 40 W light power in each propagation direction) when the transit time is  $6.2 \cdot 10^{-5} \text{ s}$ .

iv) *Light shift.*

With a 40 W light power in each direction, the light shift for an atom at rest in the centre of the laser beams is about 480 kHz.

To precisely evaluate broadening and shift of the  $2S_{1/2}-8D_{5/2}$  line due to the last two effects, it is useful to calculate the line profile by summing the contributions to the signal of all the possible atomic trajectories. With the experimental parameters quoted above, we calculate a line shift of +300 kHz and a line width of 780 kHz.

Other stray effects (as Stark effect and pressure broadening) may perturb the measurement. For example, an electric field of 30 mV/cm is sufficient to induce a Stark splitting of 1 MHz in the  $8D_{5/2}$ -level and a shift of -45 kHz. However, even in our experimental conditions, the relative line width of the lines observed is less than  $2 \cdot 10^{-9}$  and allows the measurement of the  $2S_{1/2}-8D_{5/2}$  transition frequency with a very high precision.

### 3. Measurement of the transition wave-lengths.

Our measurement is based on the comparison between the two-photon line wave-length and the one of an iodine-stabilized helium-neon laser at 633 nm. In order to determine the two-photon line position, the dye laser frequency is modulated with a 525 Hz sinusoidal wave giving a 1 MHz peak-to-peak shift. The  $2S$  metastable beam intensity is monitored at the same frequency. The two-photon line shape becomes a derivative trace and we record the zero position of this trace.

Our iodine-stabilized helium-neon laser has been compared with the one of the «Institut National de Métrologie». Taking into account the frequency measurement precision of the  $I_2$  stabilized He-Ne laser ( $1.6 \cdot 10^{-10}$ ) [16], we know the frequency of our He-Ne laser with a precision of  $2 \cdot 10^{-10}$ .

The key of the wave-length comparison is a nonconfocal Fabry-Perot etalon. This etalon is built with two silver-coated mirrors, one flat and the other one spherical (radius of curvature  $R = 60 \text{ cm}$ ). The finesse of the cavity is about 60 at 633 nm and 100 at 778 nm. The etalon is placed inside a box evacuated to less than  $10^{-6} \text{ mbar}$ . For the wave-length measurement we use the interferometric procedure described in ref. [17]. In order to eliminate the effects due to reflective phase shifts in the mirror coatings, the method of virtual mirrors is used. Two etalon spacings are alternately employed (10 cm and 50 cm).

The measurement scheme is represented in fig. 1. An auxiliary He-Ne laser is mode-matched into the etalon cavity. The difference between the incident intensity and the Fabry-Perot transmission is considered to prevent a possible shift due to the variation of the laser gain with frequency. The beat frequency between this He-Ne laser and the reference one is measured by a frequency counter. The dye laser is also mode-matched into the etalon cavity and locked on it. It is brought into atomic resonance through thermal sweeping of the etalon.

The frequencies of the two radiations inside the cavity agree with a basic resonance condition [17]:

$$\nu = \frac{c}{2L} (N + \Phi + \Psi) ,$$

where  $L$  is the cavity length,  $N$  an integer number,  $\Phi$  the Fresnel phase shift

$$\Phi = \frac{1}{\pi} \cos^{-1}(1 - L/R)^{1/2}$$

and  $\Psi$  the reflective phase shift for light of frequency  $\nu$ .

The contribution of  $\Phi$  is measured by comparison of the modes TEM00 and TEM01 frequencies for each radiation in the cavity. The contribution of  $\Psi$  is eliminated in three successive steps, where the etalon spacings are alternately 10 cm, 50 cm and 10 cm. A drift of the silver coating has been observed during the time of our measurement (approximately 4 months). This aging effect results in an increase of about 0.8 Å of the apparent etalon length at 633 nm with respect to the same length at 778 nm. This effect can easily be taken into account in our results. The numbers  $N$  are deduced from etalon transmission recordings for various wave-lengths.

For each transition, the beat frequency between the two He-Ne lasers has to be extrapolated at null light power to eliminate systematic effects due to two-photon light shift. Such an extrapolation is shown in fig. 3, where the transition involved is the  $2S_{1/2}(F=1)-8D_{5/2}$  transition in hydrogen. Each dot is obtained as the average of ten measurements of the beat frequency at the centre of the atomic resonance. With a 40 W light power, the light shift is about 330 kHz. Taking into account the imprecision of the light power scale, this experimental value is in good agreement with the theoretical one (300 kHz). Figure 3 clearly shows that this extrapolation quite eliminates the two-photon light shift.

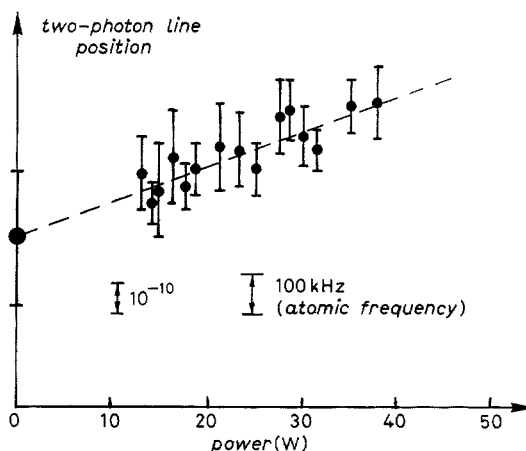


Fig. 3. – Extrapolation of the two-photon line position *vs.* the light power. The dashed line denotes a linear fit and the extrapolation result is indicated on the *y*-axis.

#### 4. Results.

The previous experimental method has been applied to three different transitions:  $2S_{1/2} \rightarrow 8D_{5/2}$  in *H* and *D* and  $2S_{1/2} \rightarrow 10D_{5/2}$  in *H*. The respective wave-lengths in air are 777.8 nm, 777.6 nm and 759.6 nm. Table I gives the experimental frequency measurements after extrapolation to null light power and the corrections due to the second-order Doppler effect and to the hyperfine structure [18, 19]. Using the theoretical work of Erickson [20], and taking into account the recently measured value of  $m_p/m_e$  [21], we can deduce the

TABLE I. - *Experimental results.*

	Hydrogen $8D_{5/2}$	Hydrogen $10D_{5/2}$	Deuterium $8D_{5/2}$
Experimental result (MHz)	385 324 758.54(20)	394 572 421.06(19)	385 429 619.56(23)
$\times 2$	770 649 517.08(40)	789 144 842.12(38)	770 859 239.12(46)
Second-order Doppler effect (MHz)	+ 0.044	+ 0.045	+ 0.022
$2S_{1/2}$ hyperfine splitting (MHz)	+ 44.389	+ 44.389	+ 13.641
$nD_{5/2}$ hyperfine splitting (MHz)	- 0.028	- 0.014	- 0.008
$2S_{1/2}$ - $nD_{5/2}$ energy splitting (MHz)	770 649 561.49(40)	789 144 886.54(38)	770 859 252.78(46)
$R_\infty - 109\,737$ ( $\text{cm}^{-1}$ )	0.315 682(57)	0.315 711(53)	0.315 682(65)
Final result	$R_\infty = 109\,737.315\,692(60) \text{ cm}^{-1}$		

Rydberg constant. The three values of  $R_\infty$  are in good agreement. Our final result is  $R_\infty = 109\,737.315\,692(60) \text{ cm}^{-1}$ .

The various experimental errors are evaluated in table II in the typical example of the  $2S_{1/2}$ - $8D_{5/2}$  transition in  $H$ . In table III our result is compared with other recent measurements of the Rydberg constant. Our measurement improves the precision on  $R_\infty$  by a factor of about 2 and gives a value a little larger than the preceding ones: a slight disagreement with the most precise one [3] can be noticed.

Moreover, our experiment also provides a measurement of the isotopic shift between  $H$  and  $D$  in the  $8D_{5/2}$  level. We obtain

$$\Delta_{\text{exp}} = 209\,691.29(7) \text{ MHz} .$$

TABLE II. - *Error Budget for the Rydberg constant measurement.*

Component	Precision (parts in $10^{10}$ )
Extrapolation of the two-photon line position	2.6
Reflection phase shift measurement and aging of coatings	2.5
Fresnel phase shift measurement	3.0
$I_2$ stabilized He-Ne laser	2.0
Stark effect	1.0
r.m.s. sum	5.2

TABLE III. - *Comparison with recent measurements.*

	$(R_\infty - 109\,737) \text{ cm}^{-1}$
GOLDSMITH <i>et al.</i> [1]	0.315 00(32)
PETLEY <i>et al.</i> [2]	0.315 21(64)
AMIN <i>et al.</i> [3]	0.315 44(11)
HILDUM <i>et al.</i> [5]	0.314 92(21)
BARR <i>et al.</i> [6]	0.315 00(110)
Present result	0.315 69(6)

These values are all corrected using the new definition of  $c$  and the experimental ratio  $m_e/m_p$  [21].

The above precision is much smaller than the precisions on each  $2S_{1/2}$ - $8D_{5/2}$  measurement, since various systematic errors cancel. This result is in good agreement with the theoretical value of Erickson [20] when modified to reflect the most precise determination of the proton-to-electron mass ratio  $m_p/m_e$  [21]:

$$\Delta_{\text{theor}} = 209\,691.32 \text{ MHz} .$$

If this measurement is considered as a way to measure  $m_p/m_e$ , the Rydberg constant value allows us to deduce

$$\frac{m_p}{m_e} = 1836.152\,72(64) ,$$

which agrees with the last result of Van Dyck *et al.* [21]

$$\frac{m_p}{m_e} = 1836.152\,701(37) .$$

We notice that, up to now, this method cannot give a precision better than  $10^{-7}$  on the  $m_p/m_e$  ratio because of uncertainties due to nuclear-size effects.

In conclusion, the measurements reported here have allowed us to improve the experimental precision on the Rydberg constant, because of the very small line width of the  $2S_{1/2}$ - $nD_{5/2}$  lines observed in hydrogen. Elimination of residual stray electric field and other causes of broadening will allow us to obtain optical relative line widths smaller than  $10^{-9}$ . In a near future, precision of the order of  $10^{-10}$  on the Rydberg constant will thus be achieved.

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