Interferometric Measurement of the $1S_{1/2}$-$2S_{1/2}$ Transition Frequency in Atomic Hydrogen

J. R. M. Barr,(a) J. M. Girkin,(b) J. M. Tolchard, and A. I. Ferguson

Department of Physics, University of Southampton, Southampton S09 5NH, United Kingdom

(Received 24 October 1985)

The $1S_{1/2}$-$2S_{1/2}$ transition frequency in atomic hydrogen has been interferometrically measured by comparison with a reference line in $^{130}Te_2$ by Doppler-free two-photon laser spectroscopy. The measured $1S_{1/2}$-$2S_{1/2}$ transition frequency of 2 466 061 397(25) MHz is in good agreement with the theoretical predictions of Erickson modified to take account of recent measurements of the Rydberg constant. This measurement can be used to deduce a value for the ground-state Lamb shift and gives the result of 8182(25) MHz which compares with a value of 8173.248(81) MHz predicted by Erickson.

PACS numbers: 32.30.Jc, 06.20.Jr

In recent years atomic hydrogen has attracted the attention of laser spectroscopists in a number of experiments designed to measure fundamental constants[1-5] or to test theoretical calculations.[6] These experiments have been motivated by the existence of highly accurate theoretical calculations[7-9] and the development of new nonlinear laser-spectroscopic techniques which may enable discrepancies between these theoretical predictions to be tested. The wavelength of the Balmer-α transition has been determined many times[1-3] by Doppler-free techniques with the aim of measuring the Rydberg constant (R_∞). Although excellent accuracies were achieved (1 part in 10^9), the experimental resolution was limited to approximately 50 MHz. Narrower linewidths (~ 2 MHz) are expected to be achieved by use of double-quantum excitation of the $2S_{1/2}$-$3S_{1/2}$ transition[4] or two-photon excitation of the $2S_{1/2}$-$8S_{1/2}$, $8D_{3/2,5/2}$ transitions.[5] Previous investigations of the $1S_{1/2}$-$2S_{1/2}$ transition[6] have concentrated on measuring the ground-state Lamb shift by interferometrically comparing one quarter of the $1S_{1/2}$-$2S_{1/2}$ frequency with one of several Balmer-β components. In these experiments the ground-state Lamb shift was measured to an accuracy of 0.4%; however, no attempt to measure the transition energy of the lines involved was reported. In this Letter we report what we believe to be the first measurement of the absolute transition frequency of the $1S_{1/2}$-$2S_{1/2}$ transition in atomic hydrogen by Doppler-free techniques. If the current best value for the Rydberg constant is used to predict the $1S_{1/2}$-$2S_{1/2}$ energy separation, the measurement reported here can be interpreted as a measurement of the difference in Lamb shifts for the $1S_{1/2}$ and $2S_{1/2}$ levels. This can then be used to deduce a value for the ground-state Lamb shift. We refer to Lamb shifts as any correction, other than hyperfine structure and reduced-mass effects, which has to be applied to the Dirac expression for the hydrogen energy levels. If it is assumed that the theoretical predictions are correct, this measurement can be used as a measurement of the Rydberg constant. As such it is the first laser-spectroscopic measurement of the Rydberg constant on any transition other than Balmer-α. Further extensions of the techniques reported here will make possible improved measurements of the Rydberg constant, and may enable the ground-state Lamb shift to be measured to an accuracy which will distinguish between the different theoretical predictions.

The $1S_{1/2}$-$2S_{1/2}$ transition frequency has been measured by interferometrically comparing one quarter of the $1S_{1/2}$-$2S_{1/2}$ transition frequency with a reference line in $^{130}Te_2$ vapor which was previously calibrated by Doppler-free saturation spectroscopy to an absolute accuracy of 4 parts in 10^{10}.[10] The experimental system used in this measurement consisted of a coumarin 102 (C102) continuous-wave (cw) dye laser (Coherent 699-21), used in a standing-wave configuration. This laser was pumped by the violet lines of a krypton-ion laser (Coherent 3000K) and typically provided 10–70 mW in a 1-2-MHz linewidth near 486 nm. All frequency measurements were made in the blue so that the measured frequencies and hyperfine separations appeared to be one quarter of the atomic value.

The relative frequency calibration was provided by the transmission fringes of a confocal interferometer with a quartz spacer and a measured free spectral range of 497.96(10) MHz. The interferometer temperature was actively stabilized to ±0.01 °C and was placed inside a second enclosure which was temperature stabilized to ±0.5 °C. The residual drift of the interferometer frequency was less than 20 MHz/h. The aluminum mirror coatings gave rise to a correction for the phase shift on reflection of less than 1 kHz near 486 nm for the tuning range used in these experiments.[11] Dispersion of air could be neglected since the interferometer was evacuated to 10^{-6} Torr and sealed off. A Doppler-free saturated-absorption experiment on $^{130}Te_2$ vapor enabled the absolute frequency to be determined under experimental conditions described by Barr *et al.*[10] In order to prevent optical feedback an acousto-optic modulator (Isomet) was inserted between the dye laser and the saturation spectrometer.

This shifted up the light used for this part of the experiment by 80.00 MHz.

The tunable 243-nm light required to excite the $1S_{1/2}$-$2S_{1/2}$ transition was generated by frequency doubling of the 486-nm radiation. In order to increase the doubling efficiency the low-power cw radiation was amplified with a pulsed, single-pass, four-stage, traveling-wave C102 dye amplifier pumped by a frequency-tripled Nd-doped yttrium-aluminum-garnet laser (Quanta Ray DCR 2). The output was 1.0–1.5 mJ in a 5-ns pulse with a 150–200-MHz linewidth which is approximately 2–2.5 times the Fourier-transform limit. The amplification process not only broadened the linewidth but also shifted the pulse spectrum up relative to the cw laser by 35–55 MHz due to pulse chirping.[6] This was observed with a confocal interferometer of 300-MHz free spectral range and 2-MHz linewidth.

Frequency doubling in urea (Quantum Technology $8\times8\times8$ mm^3) yielded up to 0.2 mJ at 243 nm. This was sufficient to enable the laser linewidth to be reduced by filtering with a confocal interferometer. The interferometer was 25 cm long with a mirror reflectivity of 90%. The linewidth was measured to be 25 MHz by use of a narrow-linewidth cw dye laser. A transmission maximum of the interferometer was maintained coincident with the cw-laser frequency by modulation of the interferometer length and use of standard first-derivative locking techniques. The length was modulated at 600 Hz over a length change corresponding to 7 MHz which broadened the linewidth of the interferometer to 30 MHz. The average interferometer pass-band frequency was locked to the cw frequency to better than 5×10^{-3} of the interferometer linewidth. The pulsed blue light was mode matched into the interferometer giving a free spectral range of 600 MHz. For mode matching, the interferometer must be aligned with its optic axis parallel to the laser beam to a measured angle of less than $\pm5\times10^{-4}$ rad. As the cw light and the pulsed light could be mode matched simultaneously, the angle between them must have been smaller than this limit. Consequently the difference in resonance frequency between the cw and pulsed light due to alignment effects could be made negligibly small. However, the frequency spectrum of the transmitted pulsed light was found to have a linewidth of 35–40 MHz and to be shifted up by 12.3(5.2) MHz relative to the cw frequency. This is related to the amplifier shift since the frequency spectrum of the light being filtered had a nonzero gradient at the cw frequency. The filtering process stretched the pulse length from 5 to 12 ns and changed the temporal profile from a Gaussian to a single-sided exponential. The peak power was reduced from 260 to 10 kW. High conversion to the uv was maintained by focusing to a beam radius of 50 μm in the urea crystal, yielding up to 20 μJ with a pulse duration of 6 ns.

Molecular hydrogen was obtained by electrolysis of a weak NaOH solution. Dissociation of H_2 was achieved in a microwave discharge (Microtron 200) with typical microwave powers of 6–10 W. The observation cell (50 mm$\times$25 mm diameter) was equipped with Spectrosil windows and two plane-parallel stainless-steel ionization detection plates (10×50 mm^2) separated by 15 mm. The pressure in the cell was monitored with a capacitive manometer (Baratron 222BA) and was varied in the region 0.1–1.5 Torr. Most measurements were taken at a pressure of 0.18 Torr. In recent high-resolution experiments, atomic beams have been used to avoid broadening and shifts due to discharge electric fields and pressure effects.[3,4] For the pressures and resolution used in this experiment it was found that the pressure shifts were negligible and so an atomic beam was not used.

The 243-nm light was focused onto a mirror 1–2 cm behind the cell with a 10-cm focal length fused-silica lens and a 15-cm focal length cylindrical fused-silica lens. The latter compensated for the distorted uv beam profile caused by walkoff in the doubling crystal. The spot radius of the uv was 100 μm and energies in the range 0.7–3 μJ were used. The ions produced by Doppler-free two-photon–resonant three-photon ionization were collected by a small electric field (5 V cm^{-1}) between the metal plates. The Stark shift caused by this field can be calculated to be less than 8 kHz.[12] The collected current was amplified by a detector with a sensitivity of 13 V pC^{-1} and a noise level corresponding to 10^4 electrons/pulse. Typical signal levels were several volts corresponding to the production of 10^6 electron-proton pairs per pulse.

The spectra shown in Fig. 1 were obtained by scanning the dye laser over a 2-GHz interval in 200 s. The

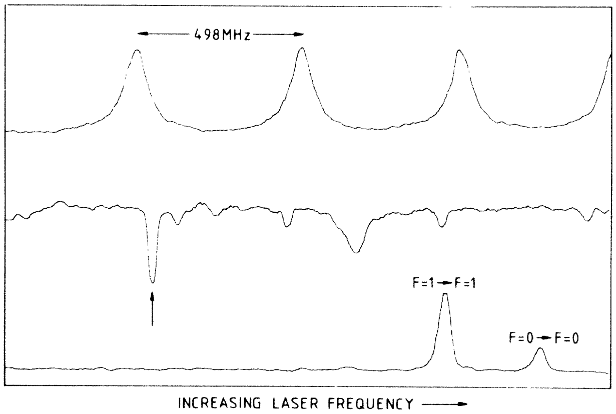

FIG. 1. A typical series of spectra. The top trace shows the interferometer fringes used as frequency markers. The middle trace shows a portion of the $^{130}Te_2$ spectrum, where the intermediate transition used as a transfer line is indicated by an arrow. The bottom trace shows the hydrogen $1S_{1/2}$-$2S_{1/2}$ spectrum.

data from the pulsed part of the experiment were collected on a pulse-to-pulse basis with no averaging while the data from the saturation spectrometer were smoothed by the 0.1-s time constant on a phase-sensitive detector. The confocal filter could only be locked over a limited scan range and so it was convenient to use another $^{130}\mathrm{Te}_2$ line as a transfer standard.

The data were analyzed as follows. The centers of the interferometer peaks were found on the computer by numerical differentiation of data which had been smoothed by the carrying out of a moving average over thirty points. The zero crossing gave the positions of the extrema. For the hydrogen data and the $^{130}\mathrm{Te}_2$ data the smoothing was over ten points. Only in the case of the hydrogen data did varying of the number of points cause a change in the centroid of each component. However, the shift was smaller than the statistical uncertainties and could be neglected. The separations of the various lines were found in terms of the reference interferometer by interpolation. It was found that small nonlinearities in the laser scan produced slight variations in the interferometer peak separations which required a quadratic fit to be taken into account. The success of this correction was observed by our measuring the known hyperfine splitting in hydrogen and ensuring that it was within the experimental error of the accepted value. The magnitude of the correction was about ±1% of the interferometer free spectral range. Eleven scans were used to obtain the separation between the calibrated $^{130}\mathrm{Te}_2$ line and the transfer line with use of the cw laser. The transfer line was found to be 549.1(1.7) MHz higher in frequency than the calibrated line. Thirty scans were used to obtain the $^{130}\mathrm{Te}_2$ line to H $F=1\rightarrow 1$ hyperfine-component separation.

To arrive at the final energy measurement several corrections have to be taken into account. These are shown in Table I and are explained as follows, where all corrections are applied in terms of the frequency scale of the 486-nm radiation.

(1) The amplifier shift was measured at the start and finish of each sequence of spectra and used to correct the average value of the separation between the transfer $^{130}\mathrm{Te}_2$ line and the H $F=1\rightarrow 1$ hyperfine component. The error of 5.9 MHz represents one standard deviation and includes the uncertainty of the amplifier shift (5.2 MHz) and statistical uncertainties (2.8 MHz).

(2) +77.678 MHz is the separation between the $F=1\rightarrow 1$ hyperfine component and the centroid of the hyperfine levels.

(3) −80.00 MHz comes from the frequency shift caused by the acousto-optic modulator.

(4) The pressure shift was monitored over the range 0.2–1.5 Torr. The measured shift was +3(6) MHz/Torr to higher frequencies. Hence a correction of −0.6(1.2) MHz at the operating pressure of 0.2 Torr is included.

TABLE I. Theoretical predictions of the $1S_{1/2}$-$2S_{1/2}$ energy separation using the calculations of Erickson (Ref. 7) modified by use of the weighted mean value of the Rydberg constant [see Taylor (Ref. 13)] together with the predicted $1S_{1/2}$ Lamb shift. The measured frequencies and corrections to these frequencies are also shown in the table, and refer to the visible laser frequency. The final $1S_{1/2}$ to $2S_{1/2}$ transition frequency is 4 times the laser frequency. All values are in megahertz.

Theory	
Dirac contribution	−2 467 411 574.203
Reduced mass	1 343 062.867
Nuclear recoil	−22.325
$1S_{1/2}$ Lamb shift	8173.248(81)
$1S_{1/2}$-$2S_{1/2}$ frequency	2 466 061 405.5(2.9)
Experiment	
Calibrated $^{130}\mathrm{Te}_2$ frequency[a]	616 513 896.30(25)
Intermediate $^{130}\mathrm{Te}_2$ offset	+549.1(1.7)
$^{130}\mathrm{Te}_2-H(F=1\rightarrow 1)$	+907.4(5.9)
$\mathrm{H}(F=1\rightarrow 1)$, centroid	+77.678
Acousto-optic shift	−80.00
Pressure shift	−0.6(1.2)
ac Stark shift	−0.6(0.6)
Interferometer free spectral range	0.0(0.1)
Total	616 515 349.3(6.3)
Measured $1S_{1/2}$-$2S_{1/2}$ frequency	2 466 061 397(25)
Deduced $1S_{1/2}$ Lamb shift	8182(25)

[a] Reference 8.

(5) The ac stark shift can be calculated[6] to be 0.6(0.2) MHz, and increases the separation between the levels. This shift was below the resolution of this experiment and is included as a correction of −0.6(0.6) MHz.

The predicted value of the $1S_{1/2}$-$2S_{1/2}$ transition frequency is a sensitive function of the assumed value of the Rydberg constant. There have been several recent measurements of the Rydberg constant. These are shown in Table II where the data have been modified to take account of changes in these measurements due to the redefinition of the meter, small changes in the fine-structure constant, and the electron-to-proton mass ratio.[13] The prediction of the Erickson[7] theory for the $1S_{1/2}$ to $2S_{1/2}$ transition frequency using these values for the Rydberg constant is also shown in Table II together with our experimental measurement of this transition frequency.

If the Dirac and relativistic nuclear-recoil contributions calculated with the weighted mean value of the Rydberg constant in Table II are subtracted from the measured value of the $1S_{1/2}$-$2S_{1/2}$ transition frequency a value of 7137(25) MHz is obtained for the difference

TABLE II. Comparison of some recently measured values of the Rydberg constant adjusted by Taylor (Ref. 13) together with the weighted mean. The resulting $1S_{1/2}$ to $2S_{1/2}$ transition frequency based on Erickson's theory is also shown. Our experimental measurement of this frequency and the Rydberg constant are shown for comparison. The tabulated quantities are $\Delta R := R_\infty - 10\,973\,731\ \mathrm{m}^{-1}$ and $\Delta E := E_{1S\text{-}2S} - 2\,466\,061\,000$ MHz.

	ΔE	ΔR
Goldsmith *et al.*[a]	0.500(32)	397.8(7.2)
Petley *et al.*[b]	0.521(64)	402.6(14.4)
Amin *et al.*[c]	0.539(12)	406.6(2.7)
Weighted mean[d]	0.534(13)	405.5(2.9)
This work	0.50(11)	397(25)

[a] Reference 1.
[b] Reference 2.
[c] Reference 3.
[d] Reference 13.

in $1S_{1/2}$ and $2S_{1/2}$ Lamb shifts. This compares with a theoretical value of 7128.171(81) MHz predicted by Erickson.[7] If the theoretical value for the $2S_{1/2}$ Lamb shift predicted by Erickson (excluding recoil) of 1045.077(0.010) MHz is accepted then we obtain an experimental value for the ground-state Lamb shift of 8182(25) MHz compared with a theoretical value of 8173.248(81) MHz.

An alternative interpretation of this experiment is that it is a measurement of the Rydberg constant. As such it is the first measurement of this important constant in a transition other than Balmer-α. The present work gives a value for the Rydberg constant of $10\,973\,731.50(11)\ \mathrm{m}^{-1}$ which is consistent with other measurements but with a somewhat larger uncertainty.

The methods described in this paper offer a potential for significant improvements in the measurement of the Rydberg constant and the Lamb shifts in atomic hydrogen. The principal uncertainties are due to linewidths of the resonances obtained and uncertainties in the frequency shift between the cw dye laser and the amplified beam. Both of these problems could be reduced by reducing the linewidth of the filter interferometer. We estimate that the linewidth could be realistically reduced to the region of 3 MHz without significant loss of signal-to-noise ratio. We would then expect the accuracy of our measurements to improve to about 300 kHz. At this level of accuracy an improved measurement of the Rydberg constant should prove possible. This will make an interesting test of systematic errors since all other accurate measurements of the Rydberg constant have been on Balmer-α. It should also be possible to make an improved measurement of the $1S_{1/2}$ Lamb shift by comparing the $1S_{1/2}$-$2S_{1/2}$ transition frequency directly with a component in Balmer-β. A metastable beam suitable for making this measurement has been constructed in our laboratory. This next generation of experiment may enable the differences between the Erickson,[7] Mohr,[8] and Sapirstein[9] theories, which amount to about 300 kHz in the ground-state Lamb shift, to be tested.

The authors gratefully acknowledge the support of Science and Engineering Research Council for this work in the form of research grants, an Advanced Fellowship (A.I.F.), research studentships (J.R.M.B., J.M.T.), and a CASE studentship (J.M.G.) in conjunction with the National Physical Laboratory. We also thank Dr. B. W. Petley for many useful discussions.

(a)Present address: Central Laser Facility, Rutherford Appleton Laboratory, Oxford, United Kingdom.

(b)Present address: Photon Control Limited, Cambridge, United Kingdom.

[1] J. E. M. Goldsmith, E. W. Weber, and T. W. Hänsch, Phys. Rev. Lett. **41**, 1525 (1978).

[2] B. W. Petley, K. Morris, and R. E. Schawyer, J. Phys. B **13**, 3099 (1980).

[3] S. R. Amin, C. D. Caldwell, and W. Lichten, Phys. Rev. Lett. **18**, 1234 (1981).

[4] D. Shiner and C. Wieman, in *Precision Measurement and Fundamental Constants II,* edited by B. N. Taylor and W. D. Phillips, National Bureau of Standards Special Publication No. 617 (U.S. GPO, Washington, D.C., 1984), p. 123.

[5] F. Biraben and L. Julien, Opt. Commun. **53**, 319 (1985).

[6] C. Wieman and T. W. Hänsch, Phys. Rev. A **22**, 192 (1980).

[7] G. W. Erickson, J. Phys. Chem. Ref. Data **6**, 831 (1977).

[8] P. J. Mohr, Ann. Phys. (N.Y.) **88**, 52 (1974).

[9] J. Sapirstein, Phys. Rev. Lett. **47**, 1723 (1981).

[10] J. R. M. Barr, J. M. Girkin, A. I. Ferguson, G. P. Barwood, P. Gill, W. R. C. Rowley, and R. C. Thomson, Opt. Commun. **59**, 217 (1985).

[11] J. M. Bennet, J. Opt. Soc. Am. **54**, 612 (1964).

[12] J. A. Blackman and G. W. Series, J. Phys. B **6**, 1090 (1973).

[13] B. N. Taylor, private communication of work carried out in connection with the 1985 least squares adjustment of the fundamental constants. Other values used are $\alpha^{-1} = 137.035\,988(<10)$ and $m_p/m_e = 1836.152\,701(37)$.