

Continuous-wave measurement of the 1S Lamb shift in atomic hydrogen

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We have determined the 1S Lamb shift in atomic hydrogen with a precision of 2 parts in 10^4 by measuring the frequency of the 1S-2S transition observed by continuous-wave Doppler-free two-photon spectroscopy. We employ an interferometrically calibrated absorption line in $^{130}\text{Te}_2$ as our reference and obtain a 1S-2S interval of 2466061413.8(1.5) MHz. Choosing a value of the Rydberg constant measured independently by high-resolution spectroscopy of the hydrogen Balmer- β transition, we find the 1S Lamb shift to be 8173.3(1.7) MHz, in good agreement with the theoretical value of 8172.94(9) MHz.

The 1S-2S transition in atomic hydrogen¹⁻⁴ has an unusually narrow natural linewidth of 1.3 Hz, offering an eventual experimental resolution of 5 parts in 10^{16} . In addition, the extreme simplicity of the hydrogen atom allows highly accurate calculations of its energy levels using the modern formulation of quantum electrodynamics (QED). Hence, spectroscopic study of the hydrogen 1S-2S transition permits precise measurements of fundamental constants and stringent tests of QED. In the experiment reported here we have measured the 1S-2S transition frequency and have determined the hydrogen ground-state Lamb shift with a precision of 2 parts in 10^4 .

Until now measurements of the 1S-2S transition frequency have been severely instrument limited, since intense radiation at the required ultraviolet wavelength near 243 nm was available only from frequency doubling of pulsed laser sources.¹⁻³ Consequently, previous experiments suffered from large linewidths and frequency chirping in the pulsed laser amplifiers, effects which were compounded by nonlinear processes such as second harmonic generation and the two-photon excitation itself. A significant milestone was reached when Foot, Couillaud, Beausoleil, and Hänsch⁴ observed the 1S-2S transition using continuous-wave (cw) two-photon spectroscopy, thus laying the foundation for future precision measurements that completely avoid these systematic errors.

We have, for the first time, used cw Doppler-free two-photon spectroscopy to measure the 1S-2S transition frequency in atomic hydrogen gas with a precision of 6 parts in 10^{10} . We employ cavity-enhanced multimilliwatt radiation near 243 nm produced by sum-frequency generation, and we observe the 1S-2S transition in a low-pressure hydrogen-helium cell with a resolution of 3 parts in 10^9 . For a frequency comparison, we detect an optical heterodyne signal at the difference frequency between the 243-nm source and the second harmonic of a dye laser locked to an interferometrically calibrated $^{130}\text{Te}_2$ absorption line near 486 nm. Our result for the energy-level separation $f(1S-2S) = 2466061413.8(1.5)$ MHz disagrees with that of Ref. 2 by 3.8 standard deviations, suggesting that residual frequency chirping of the pulse amplified and filtered 486-nm radiation in that experiment was not sufficiently characterized to properly predict the resultant shift of the observed two-photon resonance at 122 nm.

Choosing a value of the Rydberg constant, $R_\infty = 109737.31573(3)$ cm^{-1} , measured independently by high-resolution spectroscopy of the hydrogen Balmer- β transition,⁵ we may use our determination of $f(1S-2S)$ to obtain an experimental value for the 1S Lamb shift. Here, with some modification, we follow the notation, conventions, and results of Johnson and Soff⁶ to compute contributions to hydrogen energy levels, with updated values for the speed of light,⁷ the fine-structure constant,⁸ and the proton-electron mass ratio.⁹ For purposes of comparison with theory,⁶ we adopt the rms charge radius of the proton given in Ref. 10 instead of the slightly larger value of Ref. 11. (Use of the later would raise the theoretical Lamb shift reported below by 0.15 MHz.) We incorporate the recent calculations of Bhatt and Grotch¹² and present the full details of our computation elsewhere.¹³ In order to extract the 1S Lamb shift from our value of $f(1S-2S)$ we must first compute the contributions to the 1S-2S energy interval obtained by solving the Dirac equation for an electron moving in the Coulomb field of an infinitely massive point nucleus. We add to this result theoretical reduced mass contributions (including the relativistic reduced mass correction) and then subtract our experimental value for the 1S-2S transition frequency to find the difference of the 1S and 2S Lamb shifts, $\Delta f_L(1S) - \Delta f_L(2S) = 7128.28(1.65)$ MHz. Next, we add to this number the experimentally known hydrogen 2S-2P (i.e., $2S_{1/2} - 2P_{1/2}$) frequency separation¹⁴

$$\Delta f_L(2S) - \Delta f_L(2P) = 1057.845(9) \text{ MHz},$$

or¹⁵

$$\Delta f_L(2S) - \Delta f_L(2P) = 1057.8514(19) \text{ MHz},$$

giving

$$\Delta f_L(1S) - \Delta f_L(2P) = 8186.13(1.65) \text{ MHz}.$$

Finally, we can confidently add to this difference the theoretical 2P Lamb shift,¹³ $\Delta f_L(2P) = -12.8367(3)$ MHz, to obtain a ground-state Lamb shift of $\Delta f_L(1S) = 8173.3(1.7)$ MHz, in good agreement with the theoretical prediction¹³ of 8172.94(9) MHz. Our value represents a factor of 18 improvement in precision over the previous result of Ref. 1 [8175(30) MHz], a factor of

15 improvement in precision over the experiment of Ref. 3, which gave a result of 8190(25) MHz if we use the latest value of the Rydberg constant,⁵ and a factor of 10 improvement in resolution over Ref. 2.

On the other hand, if we trust the theoretical determination of the 1S Lamb shift, then we can interpret our experimental result as a measurement of the Rydberg constant. We obtain $R_\infty = 109737.31571(7) \text{ cm}^{-1}$, in good agreement with the result of Ref. 5 and the result $R_\infty = 109737.31569(6) \text{ cm}^{-1}$ of Biraben, Garreau, and Julien.¹⁶

A simplified scheme of this experiment is shown in Fig. 1. A detailed description of the cw 243-nm source can be found elsewhere.^{4,13} We employ efficient 90° phase matching to mix the outputs of a 790-nm ring dye laser (Coherent 699-21) and a 351-nm argon-ion laser (frequency-stabilized Coherent I-100) in a potassium dihydrogen phosphate (KDP) crystal. The 790-nm laser intensity in the crystal is enhanced in a servolocked ring cavity, providing sum-frequency output power at 243 nm ranging from 2–5 mW.

The 243-nm radiation is mode-matched into a servolocked standing-wave cavity which provides an enhancement of 8–16 for the ultraviolet light intensity circulating through the Pyrex observation cell. Atomic hydrogen is produced by microwave dissociation of H₂ in a flowing-gas discharge, and the pressure in the cell is measured with a Baratron pressure gauge. The discharge is operated alternately with 100% H₂ and a 0.7%-H₂-99.3%-He mixture in order to investigate the pressure shift of the 1S-2S transition. The resonance signal is detected by counting collision-induced Lyman- α photons.

Figure 2 shows a two-photon spectrum of the $F=1$ component of the hydrogen 1S-2S transition, obtained by frequency scanning the 790-nm dye laser. For this particular trace, the cell contained 0.225 Torr of atomic and molecular hydrogen, the circulating 243-nm intensity was 65 W/cm² at the interaction region, and the count rate at line center was 4000 counts/s. As described below, the “marker fringes” shown in Fig. 2 are obtained by a heterodyne comparison of the 243-nm radiation with the reference laser.

The measurement of an absolute frequency in this experiment relies on a previous interferometric calibration¹⁷ of an absorption line (component b_2 of Ref. 17) near 486 nm in molecular tellurium vapor (¹³⁰Te₂). We stabilize a dye laser to a nearby ¹³⁰Te₂ line (component i_2 of Ref. 17) which has a frequency 57 MHz greater than one-fourth of the hydrogen 1S-2S $F=1$ transition frequency.

Laser radiation near 486 nm is generated by an additional cw ring dye laser (Coherent 699-21). The tellurium is contained within a sealed cell that is heated to 513(5)°C, and Doppler-free signals are observed by frequency modulation saturation spectroscopy.¹⁸ The pump beam is chopped by an acousto-optic modulator that also shifts the light frequency upward by 120 MHz, while the probe beam is phase modulated at 20 MHz. The resultant dispersion-shaped resonance signal is used to frequency stabilize the dye laser.

We produce approximately 1 nW of the second harmonic of this reference laser in an angle-tuned urea crystal. We then compare the frequency of this light to that of the 243-nm sum-frequency generator by overlapping the two wave fronts on a photomultiplier. The resulting beat signal is monitored with a spectrum analyzer that acts simply as a tuned receiver with a resolution bandwidth of 1 MHz. A “beat note” is observed whenever the receiver frequency is equal to the absolute value of the difference frequency of the two 243-nm light sources. By changing the receiver frequency in discrete steps as the 790-nm dye laser is scanned, we observe several beat notes at 10-MHz intervals as shown in Fig. 2.

Inference of the frequency of the stabilized laser requires several steps which are summarized in Table I and presented in detail elsewhere.¹³ By using a precision marker interferometer we measure a frequency separation between the i_2 and b_2 components of 1436.58(26) MHz. Consideration of systematic effects in the laser stabilization scheme results in an offset of 0.03(5) MHz of the stabilized laser frequency with respect to the i_2 line center. The 60-MHz red shift of the reference laser shown in

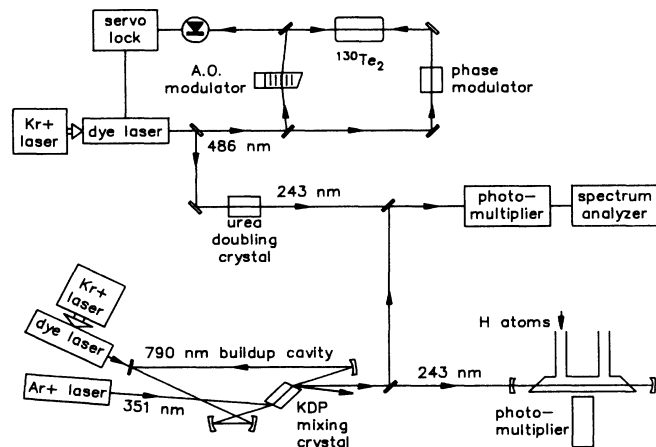


FIG. 1. Schematic of the experimental apparatus.

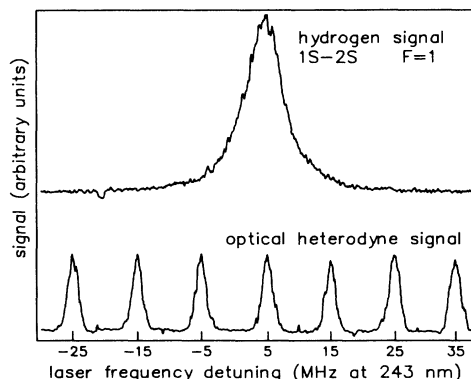


FIG. 2. Continuous-wave two-photon spectrum of the $F=1$ component of the hydrogen 1S-2S transition. The peaks in the simultaneously recorded spectrum analyzer output correspond to different values of the beat frequencies between the two 243-nm sources. These “beat notes” are obtained by changing the frequency to which the analyzer is set in 10-MHz steps.

TABLE I. Summary of results.

	Frequency (MHz)	Error ^a (MHz)
486.3 nm		
¹³⁰ Te ₂ reference frequency ^b		
f_{b_2}	616 513 896.30	0.25
Measured frequency separation		
$f_{i_2} - f_{b_2}$	1436.58	0.26
Servo control offset	0.03	0.05
Acousto-optic shift	-60.00	0.00
Stabilized laser frequency		
f_{ref}	616 515 272.91	0.36
243.1 nm		
Hydrogen ($F=1$) offset ^c		
$f_{H_1} - 2f_{ref}$	5.71	0.23
121.6 nm		
Hydrogen hyperfine splitting ^d		
$\Delta f_{hfs}(1S-2S) = \frac{1}{4} [\Delta f_{hfs}(1S) - \Delta f_{hfs}(2S)]$	310.71	0.00
Hydrogen 1S-2S centroid		
$f(1S-2S) = 2f_{H_1} + \Delta f_{hfs}(1S-2S)$	2 466 061 413.77	1.51

^aOne standard deviation. ^c $f_{H_1} = \frac{1}{2} f(1S-2S)_{F=1}$.
^bReference 17. ^dReferences 19 and 20.

Table I results from the 120-MHz blue shift of the pump beam frequency by the acousto-optic modulator. The sum of all these effects results in a stabilized laser frequency of 616 515 272.91(36) MHz.

We determine the line center of the $F=1$ component of the hydrogen 1S-2S transition by fitting the recorded signal to a Lorentzian. The resulting 1S-2S linewidth is pressure-broadened by 15.1(6) MHz/Torr (at 243 nm) for 100% H₂ and by 9.2(5) MHz/Torr (at 243 nm) for the 0.7%-H₂-99.3%-He mixture. The zero-pressure linewidth, limited by the linewidths of the fundamental lasers and by transit-time broadening, is 3.7(3) MHz. The center of each beat note is determined as the center of gravity of the top half of the peak.

Figure 3 displays the frequency offset (at 243 nm) of the $F=1$ component of the hydrogen 1S-2S transition relative to the second harmonic of the reference laser frequency as a function of the cell pressure for both 100% H₂ and the 0.7%-H₂-99.3%-He mixture. The error bars on all data points represent contributions from a 200-kHz fitting uncertainty and statistical uncertainties. When the cell contains pure hydrogen, the observed pressure shift is $-5.52(48)$ MHz/Torr (at 243 nm). However, the accuracy of an extrapolation of the line of best fit to zero pressure is limited by the changing gas composition as the cell pressure approaches 0.04(1) Torr, the residual cell pressure when there is no gas flow.¹³ The uncertainty in the pressure at which the shift vanishes can cause an error as large as 300 kHz for the extrapolated frequency offset in pure hydrogen. On the other hand, when the cell contains

the 0.7%-H₂-99.3%-He mixture, the pressure shift is reduced by almost an order of magnitude to $-0.65(33)$ MHz/Torr (at 243 nm). Thus, since the uncertainty of the H₂-He "zero pressure" point can cause an additional error of only 50 kHz, we rely on this second data set to obtain the frequency offset of 5.71(23) MHz listed in Table I, a result consistent with the corresponding value of 5.93(34) MHz for the 100% H₂ data.

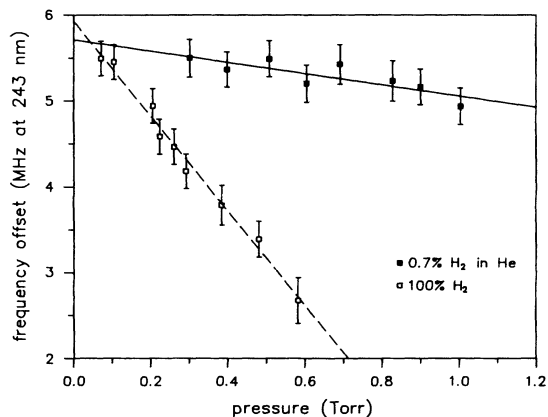


FIG. 3. Pressure shift of the hydrogen 1S-2S $F=1$ resonance relative to the second harmonic of the reference laser. The displayed lines are the best weighted fits to the corresponding data sets.

Finally, we must account for the hyperfine interaction to arrive at an experimental value for the centroid of the 1S-2S energy interval. As shown in Table I we must add one-quarter of the difference between the ground-state hyperfine splitting¹⁹ $\Delta f_{\text{hfs}}(1S)$ and the 2S value²⁰ $\Delta f_{\text{hfs}}(2S)$ to the observed frequency of the $F=1$ component to obtain the 1S-2S centroid frequency of 2466061413.77(1.51) MHz.

An order of magnitude improvement in the precision of this experiment can be achieved readily by reducing the 790-nm dye laser scan nonlinearity and by employing a better frequency reference. Much narrower experimental linewidths can be expected from the use of a cold hydrogen beam and reduction of the bandwidth of the fundamental lasers.⁴ However, the determination of the 1S Lamb shift from an absolute frequency measurement of the 1S-2S energy interval will soon be limited in precision by the uncertainty of the Rydberg constant. It then becomes more interesting to compare the 1S-2S transition with another narrow transition in hydrogen, such as a two-photon 2S- n S transition. As discussed previously,⁴ such a comparison permits the measurement of a small

frequency difference that depends critically on the Lamb shifts of the participating levels. In this way one can perform Lamb-shift measurements beyond the level of precision set by the Rydberg constant and other optical frequency standards. Ultimately, one can take advantage of gravitational cooling and a hydrogen fountain scheme²¹ to observe the 1S-2S transition with a resolution approaching the natural linewidth of 1.3 Hz.

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