Measurement of the 1S-2S Frequency in Atomic Hydrogen

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We report on a first precise measurement of the 1S-2S energy interval in atomic hydrogen. Observing the 1S-2S transition in an atomic beam by pulsed Doppler-free two-photon spectroscopy and using an interferometrically calibrated absorption line of $^{130}Te_2$ at 486 nm as the reference, we measure the frequency $f(1S\text{-}2S) = 2\,466\,061\,395.6(4.8)$ MHz. Using the calculated 1S Lamb shift, we obtain a value for the Rydberg constant, $R_\infty = 109\,737.314\,92(22)$ cm^{-1}, which is not in good agreement with the most recent previous measurement.

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The 1S-2S two-photon transition in atomic hydrogen[1-3] with its extremely narrow natural linewidth of 1.3 Hz has long been recognized as one of the most intriguing transitions to be studied by Doppler-free high-resolution laser spectroscopy. Precision measurements of its frequency, isotope shift, and relation to other hydrogenic transitions can provide stringent tests of quantum electrodynamic theory and can yield accurate values for the Rydberg constant, the proton/electron mass ratio, and the charge radius of the proton.[3]

Here we report on a first precise measurement of the absolute frequency of the 1S-2S energy interval. Observing the transition by Doppler-free two-photon excitation in an atomic beam with a frequency-doubled pulsed dye laser, and using an interferometrically calibrated absorption line of $^{130}Te_2$ at the fundamental wavelength (486 nm) as the reference,[4] we obtain from the centroid of the two hyperfine components a frequency $f(1S\text{-}2S) = 2\,466\,061\,395.6(4.8)$ MHz. This frequency can be used to determine a new value of the Rydberg constant if we trust the theoretical value of the 1S Lamb shift. With the Rydberg constant as a scaling factor, we have calculated the hydrogen levels with Erickson's model,[5] using updated values for the speed of light[6] $c = 299\,792\,458$ m/s, the proton/electron mass ratio[7] $m_p/m_e = 1836.152\,701$ (37), and the fine structure constant[8] $\alpha = 1/137.035\,981(12)$. Adjusting the Rydberg constant to obtain the measured 1S-2S frequency, we obtain $R_\infty = 109\,737.314\,92(21)$ cm^{-1}. This result is not in good agreement with the most recent previous value, $R_\infty = 109\,737.315\,44(11)$ cm^{-1}, as obtained by Amin, Caldwell, and Lichten[9] from the wavelength of the Balmer-α line. The quoted value is that given in Ref. 9, corrected for the redefinition of the meter[6] and a sign error in the diffraction correction.[10] However, there is good agreement with an earlier value, $R_\infty = 109\,737.315\,04(32)$ cm^{-1}, as measured by Goldsmith, Weber, and Hänsch[11] and also adjusted for the redefinition of the meter.

On the other hand, taking the Rydberg constant as given, we can interpret our experiment as a new measurement of the Lamb shift of the hydrogen 1S ground state. In defining this Lamb shift, we follow here the convention of Mohr[12] rather than Erickson[5] and exclude the relativistic nuclear-recoil term (2.7) of Ref. 5, which represents a relativistic correction to the reduced-mass effect rather than a QED correction (-23.8127 MHz for $n = 1$). With R_∞ from Ref. 9 we obtain a 1S Lamb shift of 8184.8(5.4) MHz, in rather poor agreement with the theoretical value[5] of 8173.25(15) MHz. If we use R_∞ from Ref. 11, on the other hand, we obtain 8174.8(8.7) MHz. For comparison, the earlier measurement of Wieman and Hänsch[2] gave a value of 8175(30) MHz if we again exclude the relativistic nuclear-recoil correction. In that experiment, the 1S-2S transition frequency was compared with the $n = 2 \rightarrow 4$ Balmer-β frequency to within 1.2 parts in 10^8. However, measurements of the absolute Balmer-β frequency[13] have not yet been carried beyond an uncertainty of about 1 part in 10^7. A direct absolute wavelength measurement of the vacuum-ultraviolet Lyman-α line was reported by Herzberg[14] in 1956. It yielded the first experimental value of the 1S Lamb shift with a precision of about 15%.

The much-discussed "Mohr-Erickson" discrepancy[15,16] in the calculation of Lamb shifts remains unimportant at our level of experimental accuracy. If we compute contributions of order $R_\infty\alpha^5$ to the 1S Lamb shift with the help of Sapirstein's[16] more accurate coefficient $A_{60} = 33.2(1.2)$ rather than Erickson's $A_{60} = 25.8(7)$, the predicted 1S Lamb shift decreases by about 0.24 MHz, increasing the 1S-2S interval by about 1 part in 10^{10}.

In this experiment, we make use of a recent interferometric calibration[4] of a strategically located absorption line of $^{130}Te_2$, observed by Doppler-free saturation spectroscopy. When the $F = 1 \rightarrow 1$ hyperfine component of the 1S-2S transition is excited with a frequency-doubled dye laser, the fundamental laser frequency is only about 1.4 GHz away from the reference line, whose frequency has been measured to within 4 parts in 10^{10}. In addition, we take advantage of a weaker Doppler-free line in $^{130}Te_2$ (component i_2 of Ref. 4) which is found approximately 60 MHz higher than the hydrogen $F=1 \rightarrow 1$ resonance.

A scheme of the apparatus is shown in Fig. 1. A cw ring dye laser (Coherent 699-21) serves as the master oscillator for the pulsed two-photon spectrometer and as the source for saturation spectroscopy of $^{130}Te_2$. Operating with coumarin 102, pumped by a 413-nm Kr^+ laser, it provides about 100 mW in a single mode near 486 nm. To assure smooth linear scanning, we employ helium-gas–pressure tuning of the reference cavity rather than the factory-provided galvo-driven Brewster plate.

In order to obtain sufficient intensity for efficient frequency doubling, part of the cw output is sent through a three-stage pulsed dye-laser amplifier (Lambda Physik 2002EC without grating-tuned oscillator), pumped by a XeCl excimer laser (Lumonics TE860-2). The amplifier provides 10-ns light pulses of about 2.3 mJ energy, but broadens the linewidth to about 190 MHz. More seriously, it introduces uncontrolled frequency shifts due to rapid refractive-index changes in the amplifying medium. Such "chirping" was identified as the dominant source of uncertainty in earlier pulsed experiments.[2] To improve the resolution and minimize the effect of amplifier shifts, we send the amplifier output through a piezoelectrically tuned confocal filter interferometer (free spectral range 328 MHz), transmitting about 0.05 mJ through one of its narrow pass bands (< 15 MHz FWHM). The center frequency of the filter is actively servo locked to the frequency of the cw dye laser.

The filtered blue (486 nm) light pulses are focused to a waist of ≈ 25 μm radius inside the frequency doubler, a 10-mm-long angle-tuned urea crystal (Quantum Technology, $\theta = 75°$, $\Psi = 45°$) which produces 243-nm radiation with nearly 10% efficiency at our intensities. Astigmatism of the second-harmonic radiation due to walkoff is reduced with a cylindrical quartz collimating lens ($f = 5$ cm), placed 2 cm behind the crystal. The ultraviolet (243 nm) beam is extracted with a quartz prism and refocused to an oblong waist of 100 μm width and 250 μm height at the excitation region located 1.6 m from the final focusing lens. The use of the far field assures well-defined wave fronts, and a spherical reflecting mirror 25 cm behind the waist produces a matching counterpropagating beam as needed for Doppler-free two-photon excitation.

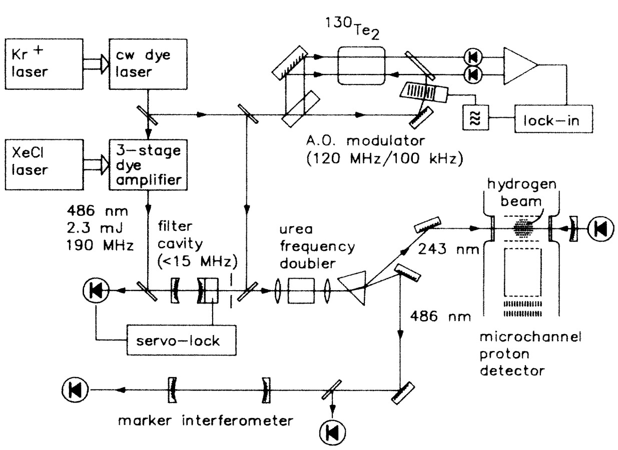

FIG. 1. Scheme of experimental setup.

The blue light is sent into an evacuated, temperature-stabilized, confocal marker interferometer[17] with a free spectral range of 75.688 864 2(57) MHz. Separate marker fringes for cw and pulsed light are recorded simultaneously by suitable electronic signal processing.

All earlier experiments[1-3] observed the hydrogen atoms in a gas cell via collision-induced Lyman-α emission. Here we employ an atomic-beam apparatus similar to equipment used for photodissociation studies[18] in order to avoid pressure shifts and problems associated with resonance trapping. Hydrogen atoms are produced in a dc gas-discharge tube operating near 0.7 Torr, and escape through a Teflon-lined metal nozzle (0.3 mm diameter, 1.3 cm long) into a cryogenic pump directly below. Shielding electrodes connected to the nozzle keep dc electric fields in the excitation region below 0.1 V/cm. The degree of dissociation in the interaction region reaches about 30%, and the atomic density is estimated as $\approx 10^{13}/\mathrm{cm}^3$. The ultraviolet light enters the apparatus through quartz windows and is focused about 1 mm below the nozzle. Ground-state hydrogen atoms are excited to the metastable $2S$ level by two 243-nm photons and then photoionized by a third 243-nm photon. A pulsed extraction field applied after the light pulse directs the photoions towards a two-stage microchannel plate detector, followed by a gated integrator. A short drift region permits differentiation between protons, other ionic species, and scattered light by time-of-flight mass spectrometry.

The tellurium reference lines are recorded simultaneously by Doppler-free saturation spectroscopy, using two probe beams in a differential detection scheme. The dimensions of the tellurium cell (Opthos, 75 mm long) and the beam parameters are similar to those used in the calibration.[4] However, we understand that the cell temperature during the calibration was overestimated by about 100 °C.[16] If we heat our cell until the linear single-pass absorption of an isolated Doppler-broadened line 9 GHz below the reference line reaches 23%, as has been reported for the calibration cell,[19] we measure a temperature of 519 ± 5 °C. The pump beam is chopped at 100 kHz with an acousto-optic modulator, and the resulting probe modulation is filtered with a 50-kHz high-pass filter and detected with a lock-in amplifier. The modulator shifts the pump frequency by 120 MHz, so that the tellurium lines appear red-shifted by 60 MHz, resulting in an almost perfect coincidence between the hydrogen $F=1 \rightarrow 1$ resonance and the $^{130}Te_2$ i_2 line.[4]

All signals (the hydrogen photoion signal, the tellurium lock-in amplifier signal, the ultraviolet intensity, and cw and pulsed blue marker signals) are digitized and processed by a microcomputer. Several hundred spectra have been recorded and evaluated in order to explore and reduce various systematic corrections.

Figure 2 shows the hydrogen $1S$-$2S$ spectrum with its two hyperfine components together with a simultaneously recorded saturation spectrum of $^{130}Te_2$. Letter symbols indicate the calibrated line b_2 and the auxiliary reference line i_2. The hydrogen photoion signal has been divided by the cube of the ultraviolet intensity in order to reduce the corresponding hydrogen signal fluctuations.

Table I gives a summary of our experimental results. The hydrogen two-photon line centers are found by fitting the experimental data with Gaussian line profiles. Tellurium line centers are found by fitting the experimental data with asymmetric Lorentzian line profiles, even though the line shifts caused by dispersive light-induced lens effects remain below 0.1 MHz. The quoted statistical errors give single standard deviations derived from the scatter of the fitted line centers in a final series of 26 spectra.

The validity of the fitting procedures is substantiated by the excellent accuracy with which we are able to measure the separation between the two hydrogen hyperfine components. Without applying any corrections, we find an experimental splitting of 310.76(23) MHz. The expected value is $\frac{1}{4} \times \frac{7}{8}$ of the ground-state hyperfine splitting[20] or 310.713 758 3 MHz. Likewise,

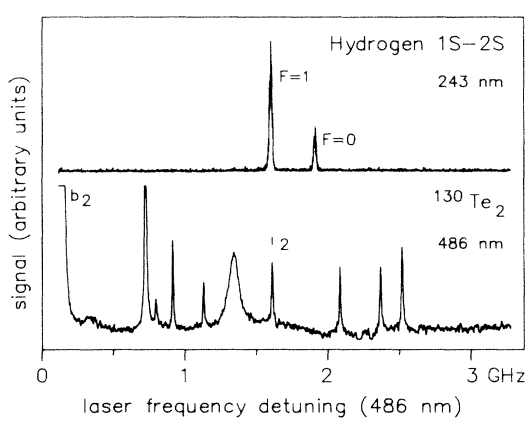

FIG. 2. Doppler-free two-photon spectrum of the hydrogen $1S$-$2S$ transition with simultaneously recorded saturation spectrum of $^{130}Te_2$. The tellurium reference spectrum appears shifted towards lower frequencies by 60 MHz as a result of the acousto-optic modulator.

we are able to measure the separation between the calibrated $^{130}Te_2$ reference line b_2 and the auxiliary line i_2 with good reproducibility, yielding the result shown in Table I.

The comparison of hydrogen and tellurium is, unfortunately, complicated by a number of systematic corrections. The largest of these, aside from the trivial acousto-optic shift, is a consistent blue shift of the filtered 486-nm pulses relative to the cw laser by an average amount of close to 4 MHz, as revealed by a comparison of the marker etalon fringes for pulsed and cw light. A locking error in the servo system may be responsible for much of this "filter shift." Any relative misalignment of the pulsed and cw beams in the marker etalon is less than 200 μrad and cannot be responsible for the observed shift. We also have ruled out any measurable contribution due to self-phase modulation in the urea crystal, by simultaneously recording marker fringes at two different intensities with the help of a rotating attenuator. However, a small contribution (≈ 0.5 MHz) is expected from a blue shift of the amplified pulses relative to the cw oscillator and filter pass band. An additional contribution can arise from the excitation of high-order transverse modes in the filter cavity which are blue-shifted as a result of spherical aberrations.[21] In order to minimize this effect, a circular aperture was placed immediately behind the filter cavity during the final measurements. Recording twenty spectra with an aperture of 3.5 mm diameter and six spectra with 2.5 mm, we observed a reduction of about 0.8 MHz in the filter shift for the smaller aperture relative to that for the larger one.

Unfortunately, any residual chirping or spatial frequency variation in the blue light can give rise to shifts and asymmetries in the second-harmonic spectrum and two-photon excitation spectrum which need not be the same as the measured shift between pulsed and cw marker fringes. A proper analysis would require accurate knowledge of the actual time- and space-dependent electric field.[2] The recorded hydrogen spectra in fact show a noticeable "tail" towards lower frequencies. As a result, a nonlinear least-squares fit by a Gaussian line profile gives a slightly lower center frequency than a computation of the center of gravity of the upper half of the line, with an average difference of 0.3(5) MHz (at 486 nm). We therefore add a "chirp correction" of 0.3 MHz to the measured filter shift to obtain the "relative pulse shift" listed in Table I, with an error of 25% or ± 1 MHz estimated in the absence of a rigorous analysis.

The hydrogen lines are also shifted towards higher frequencies by the ac Stark effect.[22] With effective ultraviolet pulse powers ranging from 65 to 170 W, the predicted shifts range from 0.2 to 0.5 MHz (at 486 nm), with an uncertainty of about 50% allowing for errors in the measurements of intensity distribution and

TABLE I. Summary of results.

	Frequency (MHz)	Error (MHz)
Measured H-$^{130}Te_2$ separation		
$f_{H1}-f_{i_2}$ [a]	−6.34	0.28[b,c]
Systematic corrections		
Acousto-optic modulator shift	−60.000	0.003
Relative pulse shift	4.2	1.0
ac Stark shift error	0	0.17
First-order Doppler shift	0	0.4
Second-order Doppler shift	0.016	0.003
Corrected separation		
$f_{H1}-f_{i_2}$	−62.1	1.1
Tellurium frequencies		
$f_{i_2}-f_{b_2}$	1437.0	0.3[b]
f_{b_2}	616 513 896.3	0.25[d]
Hydrogen ($F=1\rightarrow 1$) frequency		
f_{H1}	616 515 271.2	1.2
Measured hydrogen hfs		
$f_{H0}-f_{H1}$	310.76	0.23
Hydrogen $1S$-$2S$ centroid		
$f(1S\text{-}2S)=3f_{H1}+f_{H0}$	2 466 061 395.6	4.8

[a] $f_{H1}=\frac{1}{4}f(1S\text{-}2S)_{F=1\rightarrow 1}$, $f_{H0}=\frac{1}{4}f(1S\text{-}2S)_{F=0\rightarrow 0}$. f_{i_2}, f_{b_2} are the frequencies of components i_2, b_2 of $^{130}Te_2$ (Ref. 4).
[b] One standard deviation.
[c] Includes ac Stark corrections.
[d] Reference 4.

spot size.

Any vertical misalignment of the counterpropagating ultraviolet beams will give rise to residual first-order Doppler shifts. Optimization of the beam overlap, as monitored via the excitation signal, assures that such errors remain below 0.1 mrad, corresponding to shifts of less than 0.4 MHz (at 486 nm). Second-order Doppler red shifts at room temperature are smaller than 15 kHz.

With all these corrections, we find the hydrogen $F=1\rightarrow 1$ hyperfine resonance when the blue cw laser is tuned 62.1(1.1) MHz below the $^{130}Te_2 i_2$ component, which is 1437.0(3) MHz above the calibrated b_2 reference line. Together with the measured hydrogen hyperfine splitting, we obtain the centroid of the $1S$-$2S$ frequency as listed in Table I.

In summary, this paper reports a first precise measurement of the hydrogen $1S$-$2S$ energy interval. A Rydberg constant derived from this result is in poor agreement with the most recent previous measurement. In the light of this discrepancy, an independent confirmation of the $^{130}Te_2$ reference wavelength appears very desirable. On the other hand, the experimental uncertainties quoted here (4.8 MHz) reflect systematic corrections (ac Stark effect, amplifier chirping, etc.) which resist accurate characterization. These effects place a practical limit on further improvements of pulsed techniques in determining the absolute $1S$-$2S$ transition energy. With the recent development of a narrow-band tunable source of cw 243-nm radiation,[3] these effects are no longer important, and we look for spectacular advances in the precision of this measurement.

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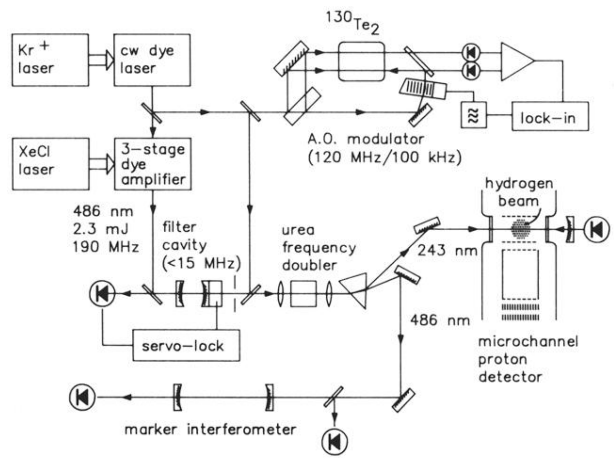

FIG. 1. Scheme of experimental setup.