Precision Measurement of the Hydrogen and Deuterium 1S Ground State Lamb Shift

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We report on new precision measurements of the 1S ground state Lamb shift in hydrogen and deuterium, based on a direct frequency comparison of the 1S-2S and 2S-4S two-photon resonances. By observing the 2S-4S transition via blue Balmer-β fluorescence we are now surpassing the accuracy of the 2S Lamb shift. Our results of 8172.86(6) MHz for hydrogen and 8184.00(8) MHz for deuterium are not in perfect agreement with the theoretical predictions of 8173.12(6) and 8184.13(6) MHz, respectively. We also derive values for the Rydberg constant and the electron-proton mass ratio.

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Recent advances in optical high resolution spectroscopy have opened the way for significant improvements in precision measurements of the hydrogen 1S Lamb shift, the Rydberg constant, and the hydrogen-deuterium 1S-2S isotope shift [1-4]. Here we report a new accurate measurement of the 1S Lamb shift in hydrogen and in deuterium. Since quantum electrodynamic effects scale as $1/n^3$, the Lamb shift is the largest for the 1S ground state. Similar to a previous experiment [1] we derive this shift from a radio-frequency measurement of the relatively small difference between the 2S-4S frequency and $\frac{1}{4}$ of the 1S-2S frequency. We achieve significant improvements in precision by observing the 2S-4S resonance via blue Balmer-β fluorescence rather than quenching of the metastable 2S atoms. By this method we have reached an uncertainty of 7.5 parts in 10^6 for hydrogen and 1.0 parts in 10^5 for deuterium. The accuracy for deuterium exceeds that of earlier experiments [5] by an order of magnitude and for hydrogen now surpasses that of the best radio-frequency measurements of the 2S-2P splitting [6], as well as that of quench asymmetry measurements of He^+ [7], so that our experiment may be considered as the most stringent test of QED for a bound atomic system to date. Exceptionally good agreement between quantum electrodynamic theory and experiment has been observed for the gyromagnetic ratio of the electron [8]. The development of QED was ushered in with the discovery of the "classic" 2S Lamb shift, but radio-frequency measurements of that shift have long reached the uncertainty limits imposed by the 100 MHz natural linewidth of the 2P state. The much narrower linewidths of optical two-photon transitions, as studied here, leave intriguing opportunities for further improvements in atomic systems.

Our experimental values differ from the theoretical predictions by 3.0σ for hydrogen and 1.3σ for deuterium if we base the calculations on a newer measurement of the proton charge radius [r_p =0.862(12) fm] [9]. For an older and contradicting measurement [r_p=0.805(11) fm] [10] agreement between experiment and theory improves for both isotopes. An alternative interpretation of the present measurement, assuming the "completeness" of quantum electrodynamic theory, would be a determination of the proton and deuteron charge radii. Reliable new measurements of the proton and deuteron charge radii would clearly have an important impact on future critical comparisons between spectroscopic experiments and quantum electrodynamic theory. Accuracies near 1 part in 10^3 might be feasible by Lamb shift measurements in muonic atoms. It will also be important to rule out that uncalculated higher order QED corrections give larger contributions than currently estimated. Two-loop binding corrections for hydrogen S states are now under investigation, and we hope that the calculations can soon be completed [11].

The 1S-2S spectrometer [12], the Ti:sapphire laser for excitation of the 2S-4S transition, and the frequency comparison are essentially the same as described previously [1]. For the 1S-2S spectrometer we couple the second harmonic of a frequency stabilized ring dye laser near 243 nm into a linear enhancement resonator inside a vacuum chamber. The standing UV wave excites 1S atoms from a collinear hydrogen beam emitted by a cooled nozzle. For simplicity we operate at liquid nitrogen temperature, so that the resolution is limited to 3 parts in 10^{11} by transit time broadening and the second order Doppler effect.

The precision to which the 2S-4S line center can be found limits the accuracy of the frequency comparison, since the natural linewidth of the 2S-4S transition (690 kHz) is much larger than that of the 1S-2S transition. Detection of the excited 4S fraction of atoms has a considerably higher potential accuracy than previous experiments monitoring the decrease in metastable 2S signal [1,3], which were limited by a large background of unexcited 2S atoms. In the present experiment we have implemented both detection methods. For excitation of the 2S-4S transition we use a frequency stabilized Ti:sapphire laser near 972 nm. The setup of the 2S-4S metastable atomic beam apparatus is shown in Fig. 1. Most of the Ti:sapphire radiation is coupled to a linear enhancement resonator (R_1=1 m, R_2=∞, L=71 cm) which is locked to the laser frequency, giving a circulating power of up to 40 W at an average beam radius of 0.5 mm. A beam of metastable hydrogen is produced in a first vacu-

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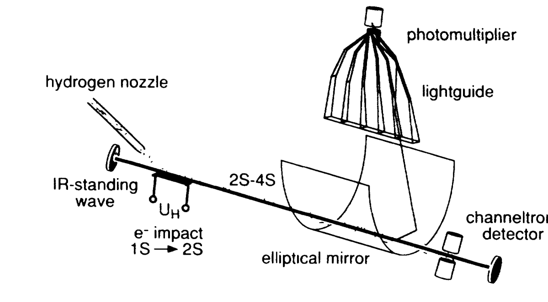

FIG. 1. Setup of the 2S-4S atomic beam apparatus.

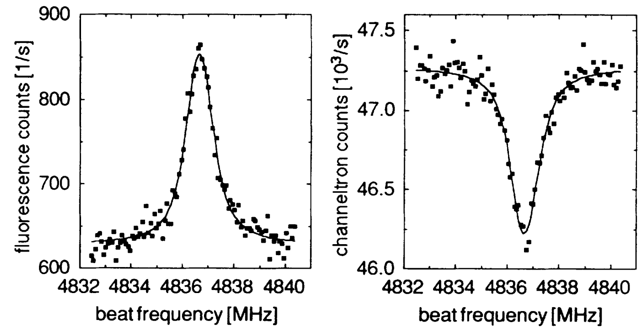

FIG. 2. Typical hydrogen $2S(F=1)$-$4S(F=1)$ spectra as detected by Balmer-β fluorescence (left) and by decrease in metastable $2S$ signal (right). The beat frequency has been measured with the blue dye laser locked to the $1S(F=1)$-$2S(F=1)$ transition.

um chamber by electron impact on hydrogen atoms. The metastable atoms leave this chamber through several apertures of about 4 mm diameter, enter a second vacuum chamber, and are detected after a 33 cm long interaction region via an electric quench field and two potassium iodine coated channeltrons sensitive to Lyman-α radiation. The interaction region is shielded against stray electric fields by a wire mesh tube coated with colloidial graphite. If the laser is in resonance with the $2S$-$4S$ transiton, a decrease of the metastable $2S$ count rate at the channeltron detector is monitored, since atoms decaying from the $4S$ state via an intermediate P state reach the $1S$ ground state with 95% probability.

At the same time, the blue $4S$-$2P$ Balmer-β fluorescence is monitored from a 15 cm long section of the atomic beam starting 15 cm behind the first aperture. This fluorescence near 486 nm is emitted with 58% probability during the decay of the $4S$ state. An elliptical mirror images the atomic beam through a vacuum window onto a focal line about 15 cm away outside the vacuum apparatus. Reflective end caps (not shown in Fig. 1) serve to let the mirror appear infinitely long. A Plexiglas lightguide transports the fluorescence light via multiple internal total reflections to the 21 mm diam entrance window of the photomultiplier (Hamamatsu R1924, quantum efficiency at 486 nm: 18%). The lightguide consists of five segments which do not touch each other. Subdividing the lightguide into segments increases its efficiency, since it increases the usable longitudinal acceptance angle. A numerical Monte Carlo simulation was used to successively optimize the geometry of this system. For reduction of stray light from the electron gun filament a combination of Schott color filters (FWHM 90 nm) was placed below the lightguide, which shielded the photomultiplier from most of the spectrally broad 1800 K thermal emission spectrum. The total detection efficiency of the fluorescence detector for Balmer-β photons is estimated to be 2%.

Typical Balmer-β fluorescence and channeltron $2S$ quench hydrogen $2S$-$4S$ spectra fitted with theoretical line shapes are shown in Fig. 2. They were obtained averaging forty scans during 20 min of measuring time, while the blue laser is locked to the maximum of the $1S$-$2S$ resonance and serves as a frequency reference. For the $2S$-$4S$ spectra observed in fluorescence the signal at full laser power is about 300 Balmer-β counts/s with a background of 600 counts/s. Typical detection rates for the channeltron detector are 50000 counted atoms per second for hydrogen and a signal decrease of 2.5% on resonance. The signal to noise ratio for the fluorescence signals is thus about a factor of 2 better than that of the channeltron signals, which could be further improved if the background radiation from the metastable beam source could be reduced.

For a frequency comparison of the $1S$-$2S$ and $2S$-$4S$ signals, we double the frequency of part of the infrared light in the $KNbO_3$ crystal and mix the resulting blue light with blue light from the dye laser on a fast photodiode. The frequency of the beat signal is measured both by a radio-frequency counter and a spectrum analyzer locked to a rubidium standard.

We have calculated $2S$-$4S$ theoretical line shapes which include the ac Stark shift, ionization from the $4S$ state by absorption of a further 972 nm photon, and the second order Doppler shift, using a numerical Monte Carlo method exploring all possible atomic trajectories through the 972 nm standing wave and integrating over the atomic velocity distribution. By fitting our experimental $2S$-$4S$ channeltron and fluorescence spectra with theoretical line shapes similar to the method described previously [1,3], light shift corrected line positions were determined. A small residual dependence of the fitted center frequency on the light power was accounted for by recording spectra at different light powers and extrapolating to zero power. The obtained beat frequencies $(2S$-$4S)-\frac{1}{4}(1S$-$2S)$ for the hydrogen are 4836.136(10) MHz and 4836.146(16) MHz for the fluorescence and channeltron signals (statistical errors only). For deuterium, we obtained 4813.660(12) and 4813.586(28) MHz, respectively. The necessary corrections to these values are listed in Table I for the fluorescence signals. The velocity distribution of the atoms in both beams has been measured by excitation of the Doppler-broadened $2S$-$4P$ transiton. The obtained second order shifts are given in Table I. Residual electric fields are estimated to be less than 50 mV/cm for the channeltron signals and negligible (< 10 mV/cm) for the fluorescence signals due to the large distance of the observed atoms from the electron gun. For the channeltron signals we obtain an uncertainty of 1.5 kHz at 486 nm for dc Stark effect. After completion of the measurements it was discovered that an uncompensated inhomogeneous magnetic field of about 400 mG rms was present along the beam axis. For deuterium with its small hyperfine splitting in the $4S$ state [5.11554(3) MHz] such a field introduces a non-negligible quadratic Zeeman shift due to hyperfine decoupling. Table I lists the necessary corrections, as computed by a Monte Carlo simulation incorporating the measured field values.

Theoretical values for the Lamb shifts have been calculated as described recently [1], but using the new results of Pachucki [13] for binding corrections to the one-loop self-energy of the states $1S$ and $2S$ and of Mohr [14] for the $4S$ state. Unless otherwise noted, we base our theoretical values (in contrast to Ref. [1]) on a newer measurement of the proton (rms) charge radius, $r_p = 0.862(12)$ fm [9], which is believed to be more reliable than a contradicting older measurement [10] since the scattering experiments were performed at lower electron-proton momentum transfers reducing the effects of disturbing quark resonances. The deuteron nuclear charge radius can be calculated from the proton charge radius with the deuteron structure radius and neutron-electron correction. In contrast to Ref. [4], we use here values of a more recent careful analysis by Klarsfeld *et al.* [15], although this increases slightly the disagreement between the theoretical value of the hydrogen deuterium isotope shift of the $1S$-$2S$ transiton and its experimental value [4]. We note that a very recent analysis [16] of an electron-deuteron scattering experiment in Saclay suggests a higher value for the deuteron structure radius which would lead to a better agreement. We do not include estimations for the effect of nuclear polarizability [17] on the deuterium energy levels (< 20 kHz for the $1S$ level), since these effects also have been neglected in the determination of the deuteron charge radius. With $r_p = 0.862(11)$ fm we obtain a deuteron charge radius $r_d = 2.115(6)$ fm and theoretical Lamb shift values $L_{1S_{1/2}} = 8173.12(6)$ MHz, $L_{2S_{1/2}} = 1045.043(7)$ MHz, $L_{2P_{1/2}} = -12.8354(20)$ MHz, $L_{4S_{1/2}} = 131.6804(11)$ MHz for hydrogen and $L_{1S_{1/2}} = 8184.13(6)$ MHz, $L_{2S_{1/2}} = 1046.418(8)$ MHz, $L_{2P_{1/2}} = -12.8343(10)$ MHz, $L_{4S_{1/2}} = 131.8524(12)$ MHz for deuterium.

TABLE I. Calculation of the hydrogen and deuterium $1S$ Lamb shift from the measured beat frequencies (fluorescence signal data only).

	Hydrogen (MHz)	Deuterium (MHz)
Extrapolated beat frequency	4836.136(10)	4813.660(12)
Corrections:		
line shape	0.000(4)	0.000(5)
reference cavity drift	0.000(3)	0.000(3)
second order Doppler shift		
$1S$-$2S$ (80 K line)	−0.003(1)	−0.002(1)
$2S$-$4S$	0.046(4)	0.015(2)
Zeeman shift	−0.005(2)	−0.032(16)
Hyperfine structure	−38.837(0)	−11.935(0)
$(E_{4S}-E_{2S}) - \frac{1}{4}(E_{2S}-E_{1S})$ (hyperfine centroid)	4797.337(12)	4801.706(21)
Dirac and reduced mass	−3928.707(0)	−3931.867(0)
$\frac{1}{4}L_{1S} - \frac{5}{4}L_{2S} + L_{4S}$	868.630(12)	869.839(21)
$\frac{5}{4}(L_{2S_{1/2}} - L_{2P_{1/2}})^{a}$	1322.306(11)	
Theoretical Lamb shifts[b]	−147.725(3)	1176.170(10)
$\frac{1}{4}L_{1S}$ Lamb shift	2043.211(17)	2046.009(23)

[a]Experimental hydrogen $2S_{1/2}$-$2P_{1/2}$ Lamb shift [6].
[b]For hydrogen: $\frac{5}{4}L_{2P_{1/2}} - L_{4S}$; for deuterium: $\frac{5}{4}L_{2S} - L_{4S}$.

From the measured beat frequencies we determine the difference frequencies of the hyperfine centroids of $2S$-$4S$ and a quarter of $1S$-$2S$. The calculation is shown in Table I for the fluorescence signals. If we include results from the $2S$ quench signals [4797.347(18) and 4801.635(32) MHz], we obtain weighted mean values of 4797.340(11) and 4801.694(20) MHz for hydrogen and deuterium, respectively. These values are not in perfect agreement with the theoretical values of 4797.364(6) and 4801.729(7) MHz, which were obtained assuming the newer measurement [9] of the proton charge radius. With the older measurement of the proton charge radius [10], the theoretical values decrease by 16 kHz and the agreement becomes satisfactory.

With the measured value for the $2S$-$2P$ splitting in hydrogen $L_{2S_{1/2}} - L_{2P_{1/2}} = 1057.845(9)$ MHz [6], the theoretical prediction for $L_{2P_{1/2}}$ and $L_{4S_{1/2}}$ for hydrogen and $L_{2S_{1/2}}$ and $L_{4S_{1/2}}$ for deuterium and including earlier results [18] we derive a $1S$ Lamb shift of 8172.86(6) MHz for hydrogen and 8184.00(8) MHz for deuterium. These measurements are currently the most precise ones of these quantities. Compared to our earlier published results [1] for hydrogen this new measurement represents an about twofold improvement in accuracy. A precise measurement of the hydrogen deuterium isotope shift of the $1S$-$2S$ transiton [4] permits an indirect frequency comparison of deuterium $2S$-$4S$ with a quarter of hydrogen $1S$-$2S$. In this way we can derive another value for the hydrogen $1S$ Lamb shift of 8172.87(8) MHz. From both measurements we obtain a slightly more accurate weighted mean value of 8172.86(5) MHz for the hydrogen ground state Lamb shift.

Our measured values for the $1S$ ground state Lamb shift can be compared with the theoretical predictions of 8173.12(6) and 8184.13(6) MHz for hydrogen and deuterium, respectively. Assuming the older measurement of the proton charge radius [10], the theoretical values decrease by 150 kHz and the agreement between theory and experiment improves. Disagreement between theory based on the newer measurement of the proton charge radius [9] and experiment is also observed for earlier determinations of the hydrogen $1S$ Lamb shift [1,3] and for the $2S$ Lamb shift [6].

Based on a recent measurement of the hydrogen $1S$-$2S$ absolute frequency [2] we can give an improved value for the Rydberg constant of $R_\infty = 109\,737.315\,684\,4(31)$ cm^{-1}. This result agrees with the value given by Biraben and co-workers [3], $R_\infty = 109\,737.315\,683\,0(31)\ \text{cm}^{-1}$.

Further improvement in precision for the ground state Lamb shift is hindered by the uncertainty in the $2S$ Lamb shift. However, the observable difference frequency between $2S$-$4S$ and a quarter of $1S$-$2S$ can serve as a direct test of QED without this limitation. (It is a measure for a compound "hyper Lamb shift" $\frac{1}{4}L_{1S} - \frac{5}{4}L_{2S} + L_{4S}$). Future experiments may reach still higher precision by using the optically excited cold metastable beam for excitation into the $4S$, or even a higher Rydberg nS state with smaller natural linewidth. Detection of the nS-$2P$ fluorescence similar to the present experiment appears especially well suited for spectroscopy of an optically excited metastable hydrogen beam, since there is no stray light at the detection wavelength.

The present experiment also allows simultaneous determination of the electron proton mass ratio and nuclear size or structure effects. Terms scaling as $1/n^2$ and $1/n^3$ can be eliminated from the measured hydrogen deuterium isotope shift of $(E_{4S}\text{-}E_{2S}) - \frac{1}{4}(E_{2S}\text{-}E_{1S})$ together with the $1S$-$2S$ isotope shift [4]. From our current results, we calculate an electron proton mass ratio of $1/1836.153\,13(40)$, which is an order of magnitude less accurate than that obtained by van Dyck *et al.* [19] via mass spectroscopy in a Penning trap. Improvement in precision opens the possibility for a more accurate determination of the electron proton mass ratio.

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$$4\,797\,322(25) \text{ and } 4\,801\,674(74)\ \text{MHz},$$

$(E_{4D_{5/2}}\text{-}E_{2S}) - \frac{1}{4}(E_{2S}\text{-}E_{1S})$:

$$6\,490\,144(24) \text{ and } 6\,494\,841(41)\ \text{MHz}.$$

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