Precise Optical Lamb Shift Measurements in Atomic Hydrogen

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The 1S ground-state Lamb shift in atomic hydrogen has been measured to an accuracy of 1.3 parts in 10^5 by directly comparing the optical frequencies of the 1S-2S and the 2S-4S,4D two-photon transitions. The result, 8172.82(11) MHz, agrees with the theoretical prediction of 8172.94(9) MHz and rivals measurements of the 2S Lamb shift as a test of QED for a bound system. A comparison of the 2S-4S and 2S-4D intervals yields a 4S Lamb shift of 131.66(4) MHz.

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We report on a new optical measurement of the hydrogen 1S ground-state Lamb shift, based on a direct frequency comparison of the two-photon transitions 1S-2S and 2S-4S,4D. With an uncertainty of 1.3 parts in 10^5, our optical measurement exceeds the accuracy of earlier experiments [1,2] by an order of magnitude and begins to rival and complement radio-frequency measurements of the 2S Lamb shift [3,4] as one of the most stringent tests of quantum electrodynamics (QED) for a bound system. Our result L_{1S}=8172.82(11) MHz agrees with the theoretical value of 8172.94(9) MHz, which is predicted if the proton charge radius is taken to be 0.805(11) fm [5]. With a more recent measurement [6] of 0.862(12) fm, however, the theoretical value increases by 150 kHz, lessening the agreement between experiment and theory.

QED, despite its unsettling concepts perhaps the most successful theory in physics, was ushered in with the discovery of the 2S Lamb shift in the hydrogen atom [3]. Refined rf measurements of the $2S_{1/2}$-$2P_{1/2}$ interval have since been pushed to the limits imposed by the 100-MHz natural width of the 2P state [4]. The 8 times larger Lamb shift of the hydrogen 1S ground state is not accessible to rf spectroscopy, since there is no nearby P state. However, it has long been recognized that two-photon spectroscopy of the extremely sharp 1S-2S transition near 243 nm could permit a very accurate optical measurement of the 1S Lamb shift. Unfortunately, such an experiment is technically very difficult, and past measurements have been handicapped by the lack of a suitable cw laser source [7] and later by the lack of sufficiently reproducible optical wavelength standards [1,2].

The new measurement reported here takes advantage of recent dramatic advances in high-resolution laser spectroscopy of atomic hydrogen. Doppler-free two-photon spectroscopy of the 1S-2S transition in a cold atomic beam [8,9] has reached a resolution of 1 part in 10^{11}. By locking a 486-nm dye laser via its second harmonic to 1S-2S, and using it as a frequency reference for 972-nm Ti:sapphire laser, we have been able to record Doppler-free two-photon spectra of the transitions 2S-4S and 2S-4D in a second beam of metasable atomic hydrogen, as illustrated schematically in Fig. 1. A fast photodiode monitors the rf beat signal between the second harmonic of the infrared laser and the dye laser. If the simple Bohr model of the hydrogen atom was correct, this beat frequency at resonance would be exactly zero. From the measured nonzero beat frequency and the known relativistic corrections, the 1S Lamb shift can be derived, without any need for an absolute optical frequency standard, in contrast to the two preceding measurements of the 1S Lamb shift [1,2]. Our accuracy is limited by the ability to locate the center of the 4S and 4D states, whose natural widths of 0.69 and 4.4 MHz, respectively, are substantially less than the 12.8 MHz of the 4P state which would ultimately limit the accuracy of a comparison of 1S-2S with the Balmer-β frequency [7]. A detailed description of our measurements and data analysis will be published later.

The 1S-2S spectrometer is similar to that described previously [8]. The frequency of the ring dye laser is stabilized with the help of a radio-frequency sideband technique to a reference cavity with gyroscope-quality mirrors optically contacted to a 45-cm-long Zerodur spacer suspended in a temperature-controlled vacuum chamber. A barium beta borate frequency-doubler crystal inside an external buildup cavity produces about 2 mW of 243-nm radiation which is coupled into a further linear buildup cavity for Doppler-free two-photon excitation of a col-

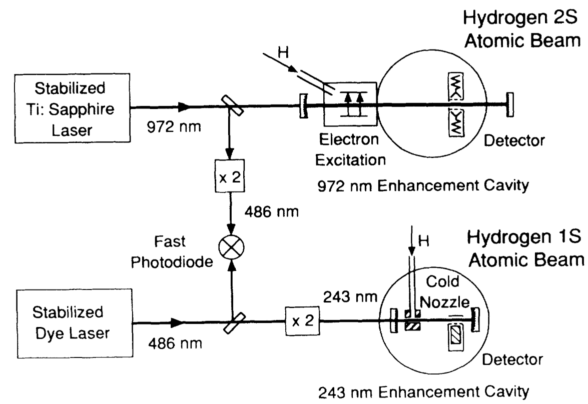

FIG. 1. Experimental setup for optical measurement of the hydrogen 1S Lamb shift.

linear beam of hydrogen atoms in the $1S$ ground state. The frequency of the blue dye laser is locked to the maximum of the $1S$-$2S$ signal with the help of a personal computer.

The Ti:sapphire laser for excitation of the $2S$-$4S$ and $2S$-$4D$ transitions is also servolocked to a highly stable external reference cavity of finesse 2000, with mirrors mounted on the same Zerodur spacer as used for the dye laser reference cavity. Stabilization has previously been achieved simply with a laser mirror mounted on a piezo transducer [10]. To compensate for high-frequency noise at larger pump powers (≈ 10 W) we feed error signal frequency components between 5 and 100 kHz to a voltage-controlled oscillator which drives an external acousto-optic frequency shifter. In this way, we achieve a linewidth of less than 50 kHz at 1 W output power, as measured from the beat signal. To tune the frequency we change the drive frequency of a second acousto-optic modulator between laser and reference cavity. The infrared light is mode matched into a standing-wave buildup cavity around a second atomic beam apparatus. When the cavity is locked to the laser frequency the circulating power reaches 50 W with a mean waist of 0.5 mm.

The beam of metastable atoms is produced in two steps, similar to the method of Biraben *et al.* [11]. Hydrogen molecules are dissociated in a microwave discharge. In a first vacuum chamber (pressure about 10^{-4} mbar) the atoms are then excited to the $2S$ state by electron impact and directed along the laser axis. The atoms leave the first chamber through a 4-mm aperture and enter a second vacuum chamber (pressure about 10^{-6} mbar) where they travel along a 33-cm-long interaction region collinear with the standing-wave laser field.

The $2S$ atoms are detected shortly behind a second aperture of 2.5 mm diameter: An electric quench field of about 50 V/cm mixes $2S$ and $2P$, forcing the emission of Lyman-α photons which are detected by two channeltrons coated with potassium iodide. Typical detection rates reach 20000 per second. This count rate is reduced if the laser is on resonance, since the excited $4S$ and $4D$ atoms cascade with 95% probability into the ground state via an intermediate P state. Typical spectra are shown in Fig. 2, fitted with theoretical line shapes. The maximum signal decrease is about 5% for the $2S$-$4S$ transition and 20% for $2S$-$4D$. For atoms which travel directly along the laser axis, the transitions are highly saturated.

The interaction region between the two apertures is shielded from electric and magnetic fields by means of a Mumetal tube coated on the inside with colloidal graphite. To first order, magnetic fields cause only a broadening of the $2S$-$4D$ resonance; we measured less than 10 mG within $\frac{4}{5}$ of the interaction length, corresponding to a broadening of about 100 kHz. Residual electric fields are estimated to be less than 50 mV/cm [12].

To compare the $1S$-$2S$ and $2S$-$4S$,$4D$ transition frequencies, we produce 500 nW of blue second-harmonic

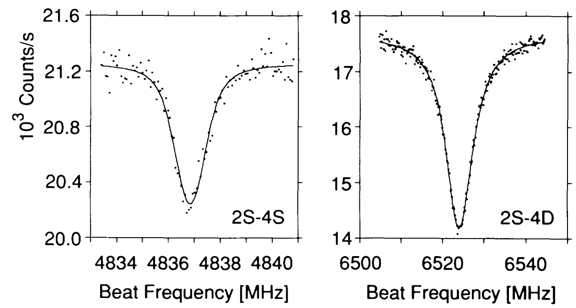

FIG. 2. Hydrogen two-photon spectra $2S_{1/2}(F=1)$-$4S_{1/2}(F=1)$ (left) and $2S_{1/2}(F=1)$-$4D_{5/2}$ (right). The beat frequency has been measured with the blue laser locked to the $1S_{1/2}$-$2S_{1/2}(F=1)$ transition.

radiation by sending part of the infrared light into a $KNbO_3$ crystal. This light is mixed with radiation from the dye laser on a fast photodiode. The beat signal is monitored with a radio-frequency spectrum analyzer and also with a microwave counter, both of which are locked to a rubidium frequency standard.

A typical spectrum is recorded by averaging over 40 scans during 20 min. A slow drift of the infrared laser's reference cavity (about 1 kHz/min) is compensated by observing the rf beat frequency. The precision of this method was deduced from independent spectrum-analyzer and counter measurements to be about 4 kHz at 486 nm.

The expected line shapes are somewhat asymmetric and their ac Stark shifts are no longer growing strictly linearly at high laser power, since atoms which travel directly along the laser axis are significantly depleted. Therefore, our spectra have been fitted by line profiles calculated by Garreau *et al.* [12] by numerically summing over the contributions from all possible atom trajectories. An additional convolution with a Gaussian profile takes into account random broadening effects such as laser frequency and intensity fluctuations, or stray electric and magnetic fields. The adjustable fit parameters were unshifted center frequency, metastable flux, laser power, and Gaussian broadening. A slight problem arises because the dark count rate could not be measured directly. A small uncontrolled electric field in the detector close to the shielding mesh of the channeltrons quenches an estimated 25% of the metastable flux even without applied quench voltage. During the data analysis, we estimated the true dark count rate by comparing the signal saturation with and without applied quench voltage. The entire fitting procedure is designed to correct for the ac Stark effect. A remaining small dependence of the fitted line center on laser power is accounted for by recording spectra at different laser intensities and extrapolating to zero power.

The extrapolated beat frequencies are listed at the top of Table I. The quoted systematic corrections for uncertainties in the theoretical line shape account for the

TABLE I. Determination of the 1S Lamb shift.

	$2S_{1/2}$-$4S_{1/2}$ (MHz)	$2S_{1/2}$-$4D_{5/2}$ (MHz)
Extrapolated beat frequency $(2S\text{-}4S/4D)-\frac{1}{4}(1S\text{-}2S)$	4836.136(28)	6523.623(28)
Corrections:		
(1) line shape	0.000(12)	0.000(11)
(2) dc Stark effect	0.000(2)	0.000(3)
(3) reference cavity drift	0.000(4)	0.000(4)
(4) second-order Doppler shift:		
1S-2S (room temp.)	−0.016(1)	−0.016(1)
2S-4S/4D	0.036(2)	0.036(2)
Hyperfine structure $\Delta_{\rm HFS}$	−38.837(3)	−33.511(0)
Dirac and rel. reduced mass contributions Δ_0	−3928.707(0)	−5752.645(1)
$\frac{1}{4}L_{1S}-\frac{5}{4}L_{2S}+L_{4S,4D}$	868.612(31)	737.487(31)
$\frac{5}{4}(L_{2S_{1/2}}-L_{2P_{1/2}})$ [a]	1322.306(11)	1322.306(11)
$\frac{5}{4}L_{2P_{1/2}}-L_{4S,4D}$	−147.722(5)	−16.581(3)
$\frac{1}{4}L_{1S}$ Lamb shift	2043.196(33)	2043.212(33)

[a]Experimental $2S_{1/2}$-$2P_{1/2}$ Lamb shift [4].

dependence of the result on the estimated effective aperture of the metastable beam at the source (2.5 mm), the estimated dark count rate, and the not exactly linear relation between measured and fitted laser power. If we simply fit our spectra by Lorentzian line shapes and linearly extrapolate the center frequencies to zero power, we arrive at beat frequencies of 4836.159(24) MHz for $2S_{1/2}$-$4S_{1/2}$ and 6523.722(28) MHz for $2S_{1/2}$-$4D_{5/2}$ (statistical errors only). For the stronger 2S-4D transition, the nonlinear effective light shift due to saturation clearly has to be taken into account.

We have measured the velocity distribution of the metastable atoms by exciting the Doppler-broadened 2S-4P transition with a collinear blue dye laser beam. Assuming a $v^4\exp(-v^2/v_0^2)$ velocity distribution [13], v_0 is found to be 2295(50) m/s, and the calculated second-order Doppler shift is given in Table I. The also listed relativistic Doppler shift of the 1S-2S transition, as derived from the observed line asymmetry, is the main source of frequency locking error for this line.

After these corrections we can determine the 1S Lamb shift from the measured beat frequency Δ:

$$\Delta=(E_{4S,4D}-E_{2S})-\tfrac{1}{4}(E_{2S}-E_{1S})$$
$$=\Delta_0+\Delta_{\rm HFS}+\tfrac{1}{4}L_{1S}-\tfrac{5}{4}L_{2S}+L_{4S,4D}\,,$$

where L denotes the Lamb shift of the corresponding state. Dirac and relativistic reduced mass corrections are contained in Δ_0, and hyperfine structure corrections in $\Delta_{\rm HFS}$.

The 1S and 2S hyperfine splittings have been measured very accurately, but for the 4S and $4D_{5/2}$ states we have to rely on extrapolated values [14] of 22.1938(12) and 1.1414(1) MHz, respectively. Since the hyperfine splitting of the latter state is smaller than the natural linewidth, a correction [12] to the line center becomes necessary which takes the relative excitation probabilities into account. For the difference $L_{2S_{1/2}}-L_{2P_{1/2}}$ we choose the value 1057.845(9) MHz obtained by radio-frequency measurements [4]. For the small Lamb shifts 2P, 4S, and 4D states we rely upon calculated values. For this calculation, we follow Johnson and Soff [15], and include self-energy contributions according to Mohr [16] and additional recoil corrections calculated by Bhatt and Grotch [17] and by Doncheski, Grotch, and Ericson [18]. Since we do not know of any recent calculations for Mohr's constant $G_{\rm SE}$ for the 4S state, and since this constant has only a small dependence on the principal quantum number, we take the value of 32.1 for the 2S state [19] and assume an error of 5. For the even smaller Lamb shift of the 4D state we follow Ericson [20]. A detailed description of our calculations will be published later. Assuming the smaller value for the proton charge radius [5], we obtain $L_{1S}=8172.94(9)$ MHz, $L_{2S}=1045.019(11)$ MHz, $L_{2P_{1/2}}=-12.836(2)$ MHz, $L_{4S}=131.677(4)$ MHz, and $L_{4D_{5/2}}=0.5364(1)$ MHz.

Our final experimental result for the 1S Lamb shift is $L_{1S}=8172.82(11)$ MHz, which is the weighted average from the frequency comparisons of the 1S-2S with the 2S-4S and 2S-4D transitions. Figure 3 compares this result with two earlier measurements [1,2] and with the theoretical predictions based on the two different charge radii of the proton [5,6].

From the difference of the two beat frequencies we can determine the interval $4S_{1/2}$-$4D_{5/2}$ yielding, to our knowledge, the first optical measurement of the 4S Lamb shift. The result, $L_{4S}=131.66(4)$ MHz, is in good agreement with the theoretical prediction and less precise radio-frequency measurements [21].

When future measurement precision exceeds the accuracy of radio-frequency Lamb shift measurements, it will

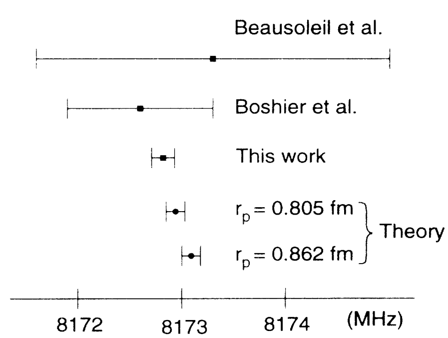

FIG. 3. Comparison of the measured hydrogen 1S Lamb shift with other recent experimental values and with theoretical predictions assuming two different proton charge radii as given in Refs. [5,6].

be reasonable to compare the measured value of $\frac{1}{4}L_{1S} - \frac{5}{4}L_{2S} + L_{4S}$ with theory directly. Our current measurement gives 868.61(3) MHz, which is in agreement with the theoretical value of 868.64(2) MHz assuming the earlier measurement of the proton charge radius [5]. Taking the latter measurement [6] changes the theoretical value to 868.66(2) MHz. Substantial further improvements in precision seem possible by using the optically excited slow metastable beam of the $1S$-$2S$ spectrometer for excitation into the $4S$ state. Direct frequency comparisons of $1S$-$2S$ with $2S$-nS,nD transitions to higher states of narrower width will also become feasible.

Future improved measurements of the proton charge radius could thus have an important impact on our ability to test QED. In addition, a direct calculation of self-energy corrections is highly desirable, in order to avoid the present extrapolation [16] from highly charged nuclei to $Z=1$.

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