

High-Precision Optical Measurement of the $2S$ Hyperfine Interval in Atomic Hydrogen

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(Received 3 June 2003; published 23 January 2004)

We have applied an optical method to the measurement of the $2S$ hyperfine interval in atomic hydrogen. The interval has been measured by means of two-photon spectroscopy of the $1S - 2S$ transition on a hydrogen atomic beam shielded from external magnetic fields. The measured value of the $2S$ hyperfine interval is equal to 177 556 860(16) Hz and represents the most precise measurement of this interval to date. The theoretical evaluation of the specific combination of $1S$ and $2S$ hyperfine intervals D_{21} is in fair agreement (within 1.4σ) with the value for D_{21} deduced from our measurement.

DOI: 10.1103/PhysRevLett.92.033003

PACS numbers: 32.10.Fn, 12.20.Fv, 32.30.Jc, 42.62.Fi

The frequency of the $2S$ hyperfine interval $f_{\text{HFS}}(2S)$ has been measured twice during the past 50 years by driving the magnetic-dipole radiofrequency transition in a hydrogen thermal beam [1,2]. The relative accuracy of these measurements (150–300 ppb) exceeds the accuracy of the theoretical prediction for the $2S$ hyperfine interval which is restricted by an insufficient knowledge of the proton structure. However, the specific combination of the $1S$ and $2S$ hyperfine intervals,

$$D_{21} = 8f_{\text{HFS}}(2S) - f_{\text{HFS}}(1S), \quad (1)$$

can be calculated with high precision due to significant cancellations of nuclear structure effects (see [3] and references therein). As the $1S$ hyperfine splitting in hydrogen $f_{\text{HFS}}(1S)$, known experimentally to several parts in 10^{13} (see, e.g., [4]), does not restrict the accuracy of (1), it is possible to compare the experimentally measured $2S$ hyperfine interval with $f_{\text{HFS}}(2S)$ deduced from the theoretical D_{21} value. The quantum-electrodynamics theory (QED) for the state-dependent contribution to the nS hyperfine splitting can thus be tested to the level up to α^4 and $\alpha^3 m_e/m_p$. This test is limited only by the experimental uncertainty.

The recent theoretical value of D_{21}^{theor} is equal to 48 953(3) Hz [3] and corresponds to a $2S$ hyperfine interval of $f_{\text{HFS}}^{\text{theor}}(2S) = 177 556 838.1(4)$ Hz. In 1956, Heberle, Reich, and Kusch measured $f_{\text{HFS}}(2S)$ for the first time [1]. Their result was equal to 177 556 860(50) Hz which is in agreement with $f_{\text{HFS}}^{\text{theor}}(2S)$. In 2000, Rothery and Hessels [2] improved the accuracy and obtained the value of 177 556 785(29) Hz. We have performed a totally independent optical measurement of $f_{\text{HFS}}(2S)$. The result of our measurement is 177 556 860(16) Hz which is until now the most precise value for the $2S$ hyperfine interval in atomic hydrogen. Both recent results are in a moderately good agreement (within 2σ) with the theoretical value.

For the measurement of the $2S$ hyperfine interval in atomic hydrogen, we have applied $1S - 2S$ two-photon spectroscopy to a cold hydrogen atomic beam which is

shielded from magnetic fields. Using a high-finesse cavity as a frequency flywheel, we deduce the $2S$ hyperfine interval as the frequency difference between two extremely stable laser light fields which excite the respective transitions between the different hyperfine sublevels of the $1S$ and $2S$ states in atomic hydrogen. The differential measurement cancels some important systematic effects. Applying this optical method, we achieve a level of accuracy which is nearly 2 times better than the accuracy of the recent radiofrequency measurement [2]. Along with the previous optical Lamb shift measurement [5], our present measurement demonstrates the perspectives of precision optical methods in fields where radiofrequency techniques have traditionally been used.

The hydrogen spectrometer setup, described in detail elsewhere [6], has been modified by magnetic compensation and shielding systems and an optional differential pumping system [7]. A dye laser operating near 486 nm is locked to an ultrastable reference cavity made from ultra-low expansion glass by means of the Pound-Drever-Hall lock. The drift of the cavity, suspended in a vacuum chamber with a two-stage active temperature stabilization system, is typically 0.5 Hz/s. The frequency of the dye laser light is doubled in a BBO crystal, and the resulting UV radiation near 243 nm is coupled into a linear enhancement cavity inside a vacuum chamber. Atomic hydrogen, produced in a radiofrequency discharge at a pressure of around 1 mbar, flows through Teflon tubes to a copper nozzle cooled to 5 K. Hydrogen atoms thermalize in inelastic collisions with the cold walls of the nozzle. The atomic beam escapes from the nozzle coaxially to the mode of the enhancement cavity. On their way through the laser field, some atoms are excited via Doppler-free two-photon absorption from the ground state to the metastable $2S$ state. In the detection region, these atoms are quenched in a small electrical field and emit Lyman- α photons which are counted by a photomultiplier. Slow atoms are selected by time resolved spectroscopy [6] so that the second-order Doppler shift and the time-of-flight broadening are reduced, yielding typical linewidths around 2 kHz at 121 nm both for the

$1S(F=0) \rightarrow 2S(F=0)$ (singlet) and $1S(F=1) \rightarrow 2S(F=1)$ (triplet) transition lines.

A turbo pump evacuates the main volume of the vacuum system to 5×10^{-5} mbar. The excitation region, separated from the main volume by a nonmagnetic metal housing, is differentially pumped by a large cryopump. Two small holes in the front and back walls of the housing allow the excitation light to enter and exit this high vacuum zone and collimate the atomic beam. With hydrogen atoms escaping from the cold nozzle, the pressure in the excitation region is typically 3×10^{-8} mbar. An additional lockable opening in the housing allows the experiment to be performed at a higher pressure of 1.5×10^{-7} mbar. By increasing the temperature of the cryopump, it is also possible to work at even higher pressures up to 5×10^{-6} mbar. All parts adjacent to the hydrogen beam are covered with graphite to reduce stray electric fields in the excitation region.

To reduce the magnetic field along the excitation region, we use a two-stage magnetic shielding setup together with external compensation coils. We have measured the residual field inside the first shielding stage made from 100 μm thin Mumetal foil which encloses the entire excitation region, the detector, and the nozzle to be less than 20 mG. Inside this shielding, 1 mm thick Mumetal tubes located along the enhancement cavity axis cover about 90% of the whole excitation path of the hydrogen atoms. The evaluated averaged shielding factor of the second shielding stage is more than 20.

An external magnetic field shifts the magnetic sublevels of the hydrogen $1S_{1/2}$ and $2S_{1/2}$ states according to the Breit-Rabi equation. For two-photon processes, allowed transitions obey the selection rules $\Delta F = 0$ and $\Delta m_F = 0$. In our experiment, we excite two-photon transitions from different magnetic sublevels of the hydrogen ground state to corresponding sublevels of the $2S$ state. In small magnetic fields, when the triplet splitting vanishes, the $2S$ hyperfine interval is given by

$$f_{\text{HFS}}(2S) = f_{\text{HFS}}(1S) + f(1, \pm 1 \text{ or } 0) - f(0, 0), \quad (2)$$

where the symbol $f(F, m_F)$ denotes the transition frequency between sublevels with quantum numbers ($n = 1, F, m_F$) and ($n = 2, F, m_F$) at 121 nm. A magnetic field H shifts $f_{\text{HFS}}(2S)$ approximately as $10H^2$ kHz/G².

The dye laser is locked to a TEM₀₀ mode of the reference cavity. Its frequency can be changed by means of a double-pass broadband acousto-optic modulator (AOM) placed between the laser and the cavity. Because of the AOM, the optical frequency doubling, and the two-photon excitation of the hydrogen atoms, the frequency shift corresponding to 121 nm is 8 times higher in absolute value than the frequency shift of the synthesizer driving the AOM. The intensity of the light used to lock the laser to the reference cavity is stabilized. All synthesizers providing the radio frequencies in our experiment

are locked to the 10 MHz signal of a commercial HP5071A cesium frequency standard (specified Allan standard deviation 5×10^{-12} within 1 s). The standard introduces a negligible error to the measured value.

During 16 days of measurements, we have recorded about 2000 hydrogen spectra for the triplet and singlet transitions. A single spectrum consists of about 30 data points, each measured for 1 s. For our fitting procedure, we have chosen the spectra recorded at a delay time of 810 μs (corresponding to velocities of the atoms ≤ 230 m/s), for which the spectrum asymmetry is considerably reduced. A typical count rate for the triplet transition in its maximum is 350, while the averaged ratio between triplet and singlet count rates is 3.25 ± 0.03 . We ascribe the deviance of this value from 3 to different recombination rates in the nozzle for hydrogen atoms in the singlet and triplet ground states.

One measurement run consists of 2–6 hydrogen spectra recorded one after another within approximately 5 min. After each run, we change the frequency of the laser light to excite the other transition. During a measurement day, we have switched about 50 times between the triplet and singlet transitions. The intensity of the excitation light is monitored after the enhancement cavity and has been kept as constant as possible during the whole day of measurement.

As the laser is locked to the same mode of the cavity, the cavity drift is the same for both singlet and triplet transitions. To determine the drift, we have fitted each hydrogen spectrum with a Lorentzian function in the time and frequency domains. A part of a day drift data set is shown in Fig. 1. To reduce the effect of a nonlinear cavity drift, the drift data are fitted stepwise within the short time periods (about 20 min each) covering two singlet and two triplet runs. During these time periods, the drift is closely linear, and we have fitted each group of four consecutive runs with two parallel lines separated by an offset frequency as shown in Fig. 1. The fit procedure delivers the slope (cavity drift), offset frequency, and corresponding errors. According to the hydrogen level

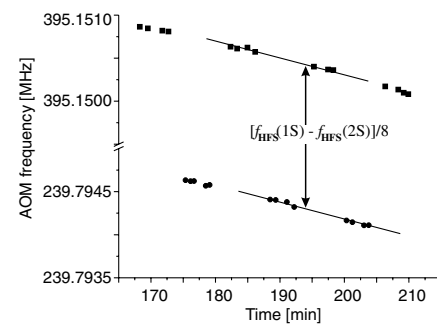


FIG. 1. AOM frequencies corresponding to the singlet (circles) and triplet (squares) transitions. The frequency dependence of the data is due to the cavity drift. The linear fit with the same slope of four data runs is shown.

scheme and the measurement technique, the offset frequency is equal to $[f_{\text{HFS}}(1S) - f_{\text{HFS}}(2S)]/8$. From this, the $2S$ hyperfine interval and the D_{21} difference can be calculated using the precise experimental value for $f_{\text{HFS}}(1S)$.

A numerical simulation of the two-photon excitation process in the hydrogen beam shows that the maximum of the excitation probability distribution for the delayed atoms is shifted in space towards the first half of the excitation region where the residual magnetic fields are the smallest. A conservative estimation of a shift arising from the less shielded paths in the direct vicinity of the nozzle and the detector gives a value of $0.5(0.5)$ Hz.

An external electrical field E mixes the $2S_{1/2}$ level with the adjacent $2P_{1/2}$ and $2P_{3/2}$ levels, shifting its energy. While in first order perturbation theory the $1S$ level is not shifted in a DC electrical field, the $2S$ $F = 0$ and $F = 1$ levels are shifted differently because of their different energy spacing from the $2P$ levels. According to a calculation of the DC Stark shift with the hyperfine structure taken into consideration, the shift of the $2S$ hyperfine interval is equal to $1100E^2$ Hz cm²/V². The stray electrical fields within the excitation region of our setup are estimated to be below 30 mV/cm [6] corresponding to a shift of -1 Hz.

The AC Stark shift of a two-photon transition scales inversely to the energy difference between real levels (in our case $1S$ and $2S$ levels) and virtual levels [8]. The hyperfine intervals are on the order of 1 GHz, while virtual energy levels are about $3/8$ Ry away from both $1S$ and $2S$ levels. Therefore, the differential AC Stark shift of the hyperfine components in the hydrogen atom is about 10^{-6} of the AC Stark shift of the $1S - 2S$ transition frequency. The latter is typically on the level of 500 Hz in our experiment, corresponding to a negligible differential shift of the $2S$ hyperfine interval assumed that the light intensity is constant. However, inevitable small fluctuations of the 243 nm light intensity cause different AC Stark shifts of each hydrogen spectrum. We have corrected for the intensity fluctuations, using the experimental value of 2.6 Hz/mW for the $1S - 2S$ AC Stark shift [9], which shifts the final value of the $2S$ interval by 2 Hz. Besides correction, we have added a conservative 2 Hz error to the error budget, which may arise from the evaluation of the light intensity circulating in the enhancement cavity.

Because of the second-order Doppler effect, the measured line shape of the two-photon transition is not symmetric, and the line center is shifted. For both singlet and triplet transitions, the excited atoms are from the same atomic beam, and the same velocity class is selected by the precisely defined delay time. Therefore, the second-order Doppler effect cancels for the differential measurement of the $2S$ hyperfine interval. We have evaluated different velocity classes of hydrogen atoms corresponding to different delay times and observe essentially no

effect on the evaluated $2S$ hyperfine interval. As an independent test we have fitted a theoretically simulated lineform for the delay time of 810 μ s with a Lorentzian function and found that the possible error of the line center definition is less than 2 Hz. This error is also added to the error budget.

According to [10], the interaction cross section for atomic hydrogen in the $2S$ state is different for triplet and singlet states, and the pressure shift of the $2S$ hyperfine interval is comparable to the pressure shift of the $2S$ triplet level. The previous $2S$ hyperfine interval measurement [2] indicates for a pressure shift of $-31(24)$ MHz/mbar, which is of the same order of magnitude as the pressure shift of $f(1, \pm 1)$ in hydrogen which is $-8(2)$ MHz/mbar [11,12]. In [2], the data was not corrected for the shift, but the error budget was increased.

We have performed the measurements at four different background gas pressures. The data are plotted in Fig. 2, each point representing the weighted average of many spectra. Points a , b , and c have been measured with approximately the same hydrogen flow through the nozzle, whereas point d is the result of a one day measurement with doubled hydrogen flow where we have investigated the effect of a pressure shift in the beam. To exclude possible long-term drifts of the experiment, we have switched 3 times between the configurations of points a and b . The data of point c have been measured half a year later. Within the available range of pressures, we observe no clear systematic dependence of the $2S$ hyperfine interval frequency on the background gas pressure. However, there is some scatter of the data. In the final averaging of the data points a , b , and c , we correct for the pressure shift taken from [11,12] for the $2S$ triplet level, but add the biggest correction of 10 Hz to the error budget.

The velocity distribution in the hydrogen beam [6] originates from a thermal velocity distribution. Atoms belonging to the slow wing of the distribution interact

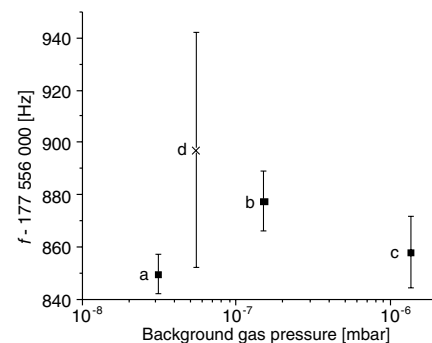


FIG. 2. Averaged results of measurements of the $2S$ hyperfine interval at different background gas pressures (logarithmic pressure scale, error bars give statistical error). The cross represents a measurement with doubled hydrogen flow. a , nine days of measurement; b , four days; c , two days; d , one day.

TABLE I. Summary of systematic errors and the final result for the $2S$ hyperfine interval.

	Frequency [Hz]	Error [Hz]
Averaged interval frequency	177 556 860	6
Residual magnetic fields	0.5	0.5
DC Stark shift	-1	1
AC Stark shift	0	2
Line shape effects	0	2
Pressure shift (background gas)	0	10
Pressure shift in the beam	0	10
Final result	177 556 860	16

more frequently with the rest of the beam than atoms traveling at the most probable thermal velocity, therefore a pressure shift due to intrabeam interaction should be larger for them. We have evaluated the $2S$ hyperfine interval of each group of four runs (see Fig. 1) contributing to point d at different delay times (including all atoms, atoms with velocities ≤ 900 m/s, ≤ 450 m/s, ≤ 300 m/s, ≤ 230 m/s, ≤ 190 m/s), and find some scatter on the level of 5 Hz for different delays of the same group (the error bars in Fig. 2 are dominated by the scatter between different groups). For regular experimental conditions without doubled hydrogen flow, the effect should be smaller. Accounting for the low statistics of this single day, we have added an error of 10 Hz for the possible pressure shift in the beam.

One of the main processes causing the data scattering is a nonlinear drift of the laser frequency on the time scale of 30 min. This process does not cancel in our fit procedure, and has to be averaged. During the 16 days of measurement, the cavity drift can be considered as random and we expect no systematic shift due to it. The distribution of the evaluated values for the $2S$ hyperfine interval without the data of point d is symmetrical and can be approximated with a Gauss function of 140 Hz width. The weighted average for $f_{\text{HFS}}(2S)$ is 177 556 860(6) Hz.

All contributions to the $2S$ hyperfine interval in our measurement are summarized in Table I. In Fig. 3, we compare the D_{21} values corresponding to the $2S$ hyperfine interval measurements in atomic hydrogen with the present theoretical value [3]. Applying a pressure shift correction to the data of [2] reduces the 2.3σ disagreement with our result, but it might also be interpreted as an indication for an unknown systematic effect. Both recent results enclose the theoretical value. We confirm theory on a level of 0.12 ppm.

Our current measurements along with other precision experiments on the hyperfine structure of $1S$ and $2S$ levels in hydrogen and the $^3\text{He}^+$ ion [13] offer a test of QED on a level of accuracy comparable to tests on pure leptonic

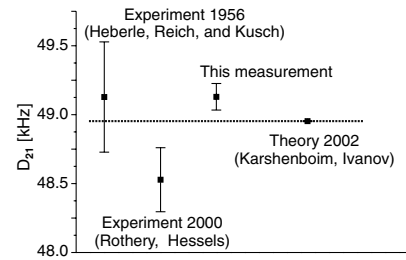


FIG. 3. D_{21} values corresponding to the several $2S$ hyperfine interval measurements. The dashed lines represent the error bar of the theoretical value.

atoms such as muonium and positronium [3]. The 16 Hz error of our measurement rivals the 20 Hz error of the radiofrequency $2S$ hyperfine interval measurement in deuterium [14], which can also be performed optically.

N. K. acknowledges support from the AvH-Stiftung. The work was supported in part by DFG (Grant No. 436RUS113/769/0-1) and RFBR (Grant No. 03-02-16843). The authors thank A. Pahl for calculations concerning the DC Stark effect and Eric Hessels for useful discussions.

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