

Precision Measurement of the Isotope Shift of the $1S$ - $2S$ Transition of Atomic Hydrogen and Deuterium

F. Schmidt-Kaler, D. Leibfried, M. Weitz, and T. W. Hänsch
Max-Planck-Institut für Quantenoptik, 8046 Garching, Germany
(Received 21 December 1992)

Using the extreme resolution of a cold hydrogen beam spectrometer and a novel sideband technique, the isotope shift of the $1S$ - $2S$ transition of hydrogen and deuterium has been directly measured to an accuracy of 3.7×10^{-8} . Our experimental result of 670994337(22) kHz is compared with the theoretical prediction of 670994414(22) kHz that can be calculated, if we assume the value of 1.9627(38) fm for the structure radius of the deuteron and 0.0189(4) fm² for the neutron-electron correction. The 2.5σ difference between the predicted and the measured values may be interpreted in terms of polarizability of the deuterium nucleus.

PACS numbers: 35.10.Bg, 21.10.Ft, 32.30.Jc

Recent advances in Doppler-free two-phonon spectroscopy of the $1S$ - $2S$ transitions in hydrogen and deuterium [1,2] with their extremely small natural linewidth of 1.3 Hz have opened a unique opportunity for very precise measurements of the 671 GHz isotope shift. We report here on a measurement of this shift to within 3.7 parts in 10^8 . Longitudinal excitation of the $1S$ - $2S$ transition in a liquid-nitrogen cooled atomic beam yields a resolution of 2.2 parts in 10^{11} . A novel electro-optic phase modulator [3] at 84 GHz permits a direct comparison of the isotope shift with the microwave standard frequency of an atomic clock. Our result, $\Delta\nu = 670994337(22)$ kHz, is 25 times more accurate than the best previous measurement [4].

The 671 GHz isotope shift can be ascribed almost entirely to the different masses of proton and deuteron. Nevertheless, nuclear volume effects more than 2 orders of magnitude smaller (about 1 MHz for H and -6 MHz for D) dominate the theoretical uncertainty. Quantum electro-dynamical effects cancel almost entirely in the isotope shift. The contribution of the deuteron polarizability was estimated to be approximately 20 kHz for the $1S$ - $2S$ transition [5]. Our experiment is the first of its kind that is sensitive even to these very small effects of hadronic dynamics.

Further improvements in measurement precision by 1 or 2 orders of magnitude are within reach of our present experimental setup. An accurate value of the mass ratio of electron and proton could be determined together with future isotope shift measurements of higher $2S$ - nS transitions, so that hadronic structure effects that scale with n^{-3} can be eliminated. We should mention that these hadronic effects could also be studied in separate experiments, such as Lamb-shift measurements in muonic hydrogen and deuterium [6]. Together with proper complementing experiments, accurate measurements of the $1S$ - $2S$ isotope shift will permit studies of hadronic structure and polarizability in a regime of very low relative momentum of electron and nucleus that is inaccessible to electron-nucleon colliders. If nuclear structure and the electron/proton mass ratio can be determined more accurately in independent experiments, then measurements of

the isotope shift can provide challenging tests of our understanding of relativistic two-body or even three-body systems.

Two-photon excitation of the $1S$ - $2S$ transition requires intense highly monochromatic light near 243 nm. We produce this light by doubling the frequency of a ring dye laser (Coherent 699) that is stabilized to an external optical resonator via a radio-frequency sideband modulation method.

The stability of the reference cavity is crucial for the present experiment. We use a pair of highly reflecting mirrors optically contacted to a Zerodur spacer of 45 cm length. The measured finesse is about 57000 and the free spectral range is 332 MHz. This cavity is freely suspended from soft springs in a temperature-stabilized vacuum chamber. A small part of the 486 nm laser light is phase modulated at 21.1 MHz and coupled into a TEM₀₀ mode of the reference cavity. Demodulating the photocurrent of the reflected light gives the error signal which is fed into the frequency control elements of the dye laser: For low frequencies we use a Brewster plate that can be rotated and a mirror that is mounted on a piezo translator. Frequency fluctuations up to 2 MHz are compensated with an electro-optic phase modulator inside the laser cavity. The frequency stability of the laser is limited by the reference cavity to about 1 kHz at 486 nm. An acousto-optic frequency shifter is employed for fine tuning of the dye laser with respect to the reference resonator.

For frequency doubling, we use a nonlinear barium- β -borate crystal inside an external enhancement resonator. From up to 500 mW of fundamental power we generate about 2 mW UV power, which is coupled into a standing wave mode of a linear UV resonator (beam waist 140 μ m, circulating power 20 mW) inside the vacuum chamber of the atomic beam apparatus.

In our experiment (Fig. 1) the hydrogen or deuterium molecules are dissociated in a microwave discharge and guided with Teflon tubes to a nozzle cooled to liquid-nitrogen temperature. The cold atomic beam is traveling freely along a collinear 15 cm long excitation region until

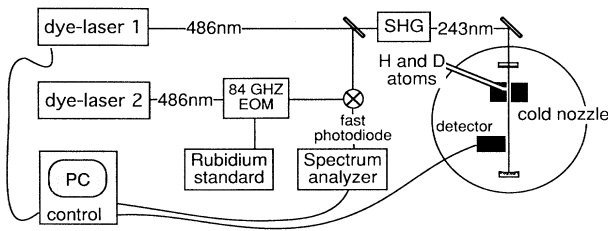


FIG. 1. Experimental setup.

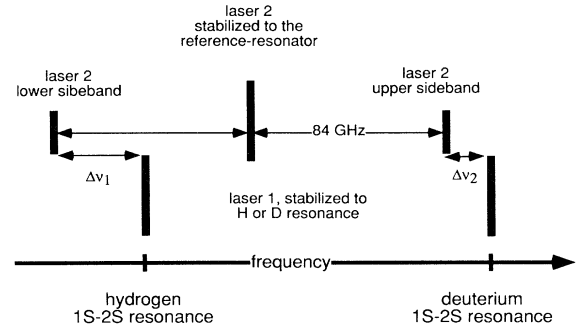
the atoms enter the detector where the excited metastable $2S$ state is quenched by a small electric field. The emitted Lyman- α photons are detected by a photomultiplier, followed by a pulse amplifier and discriminator, and recorded with the same personal computer that controls the frequency of the laser. A net coated with graphite works as an electric shield of the excitation region, while magnetic fields can be compensated with a pair of magnetic coils in Helmholtz geometry outside the vacuum chamber.

Cooling of the nozzle reduces the mean velocity of the atoms in the beam. At the temperature of liquid nitrogen the linewidths of the resonances are reduced to 35 kHz for hydrogen and 26 kHz for deuterium at the excitation wavelength of 243 nm [7]. Both relativistic Doppler shift and transit time broadening of the moving atoms in the beam determine the line shape of the $1S$ - $2S$ resonance. To lock the laser to the atomic $1S$ - $2S$ resonance of hydrogen or deuterium, a computer program shifts its frequency 5 kHz up and down and measures the difference in the count rate of metastable $2S$ atoms in the detector. This difference serves as an error signal to lock the laser to the maximum of the line shape.

To directly bridge the large frequency gap between the two resonances a novel electro-optic phase modulator is used [3]. The light of a similarly stabilized second dye laser (laser 2) is sent through a nonlinear LiTaO_3 crystal inside the focus of an open millimeter-wave Fabry-Pérot cavity. The standing millimeter wave in this cavity modulates the index of refraction for the laser light inside the crystal. The light is guided through the crystal in a zigzag path by multiple internal total reflections [3,8]. In this way phase matching of the optical and millimeter waves and a modulation index of about 5% are achieved. The millimeter-wave oscillator near 84 GHz is phase locked to a rubidium frequency standard with an accuracy of better than 10^{-10} . The sideband spacing is twice the modulation frequency Ω ,

$$2\Omega = 167\,347\,764.14 \pm 0.02 \text{ kHz}.$$

To move these sideband frequency markers near the hydrogen and deuterium resonances, the modulated laser carrier is tuned midway in between (Fig. 2). While the first laser excites the $1S$ - $2S$ transition of hydrogen or deuterium, the small residual beat frequencies $\Delta\nu_1$ and

FIG. 2. Scheme of the direct rf measurement of the isotope shift of the $1S$ - $2S$ transition in hydrogen and deuterium.

$\Delta\nu_2$ with the sidebands of the reference laser are recorded with a fast photodiode, followed by a spectrum analyzer. Laser 1 is alternately stabilized to the $1S$ - $2S$ resonances of hydrogen (hyperfine level $F=1$) and deuterium (hyperfine level $F=\frac{3}{2}$, $m_F=\pm\frac{3}{2}$), while the beat note between laser 1 and the sidebands of laser 2 is recorded (Fig. 2). The frequency difference between the two-photon resonances at 243 nm is then given by

$$\Delta = 2(2\Omega + \Delta\nu_1 - \Delta\nu_2),$$

where the factor of 2 arises from the frequency doubling of the 486 nm light. Because we cannot excite both resonances at the same time, and the reference resonator frequency of laser 2 is slowly drifting (approximately 15 Hz/s), the frequency marks are also drifting. To correct for this effect, we record the drifts of both beat frequencies with time. The drift of $\Delta\nu_1(t)$ is fitted with a polynomial of fourth order. The difference between measured $\Delta\nu_2(t)$ and interpolated $\Delta\nu_1(t)$ at the same times is constant within the measurement accuracy. The mean value has an uncertainty of 5 kHz due to the interpolation procedure described above.

Before we can evaluate the isotope shift, we have to discuss possible sources of systematic errors. Residual electric fields shift the $1S$ - $2S$ transition frequency by nearly the same amount for hydrogen and deuterium, while the absolute shift can be estimated to be less than 1.5 kHz [7]. For deuterium the hyperfine components with $m_F=\pm\frac{1}{2}$ are separated from the unshifted $m_F=\pm\frac{3}{2}$ components by a magnetic field of 2.2 G during the recording of this transition. In hydrogen the Zeeman splitting of the $F=1$ hyperfine components in the residual field of less than 0.3 ± 0.1 G is not resolved. Since the $F=1$, $m_F=0$ component is blueshifted, while the $F=1$, $m_F=\pm 1$ components are unshifted, we can give a total shift of the center of the $1S$ - $2S$ $F=1$ resonance of 0.22 ± 0.17 kHz [7]. Because the thermal velocities of the hydrogen and the deuterium atoms are different at the same temperature due to their different masses, their relativistic Doppler shifts are not the same. We have calculated these shifts from the asymmetry of the observed

TABLE I. Determination of the isotope shift.

	Frequency (kHz)	Error (kHz)
Sideband spacing	$2 \times 167\,347\,764.14$	± 0.02
Beat difference	$2 \times 454\,628.6$	± 2.5
Rel. Doppler shift for H, $T=80$ K	-11.8	± 3
Rel. Doppler shift for D, $T=80$ K	7.3	± 3
Residual magnetic field effect	0.22	± 0.17
Difference of 1S-2S two-photon transition frequencies for hydrogen and deuterium at 243 nm	$335\,604\,781.2$	± 10.9
Total hyperfine structure correction at 121 nm	-215 225.595	± 0.014
Isotope shift of the 1S-2S energy difference (centroids)	$670\,994\,337$	± 22

resonance lines, which is in good agreement with theoretical predictions. They are 11.8 ± 3.0 kHz for hydrogen and 7.3 ± 3.0 kHz for deuterium at the excitation wavelength of 243 nm, when the nozzle is cooled to 80 K.

We correct for the hyperfine structure [9] to obtain the line centroids. Table I summarizes all corrections at the excitation wavelength of 243 nm. Our final result of $670\,994\,337 \pm 22$ kHz is in good agreement with the last measurement at Oxford, $670\,994.33 \pm 0.64$ MHz [4] (Fig. 3).

The larger isotope shift is mainly a mass effect. The reduced mass correction for hydrogen and deuterium, including relativistic nuclear recoil effects, can be calculated with a precision of 12 kHz, because the ratios of electron mass and proton mass, respectively, deuteron mass are known with the accuracy of 2.0×10^{-8} (Ref. [10]). A much smaller part of the isotope shift, about 5 MHz, is a volume effect due to the difference in the charge radii of the two nucleons. Both the uncertainty of mass ratios and the measurement accuracy are included in the given error contribution. Thus from our measurement of the isotope shift we can calculate the difference of the quadratic charge radii of the proton and the deuteron [11]. We find $\langle r_D^2 \rangle - \langle r_p^2 \rangle = 3.795 \pm 0.019$ fm², if we only assume first-order nuclear size corrections.

To compare the measured isotope shift with a theoretical prediction we have to calculate the difference in charge radii, given from elastic nucleon-electron scatter-

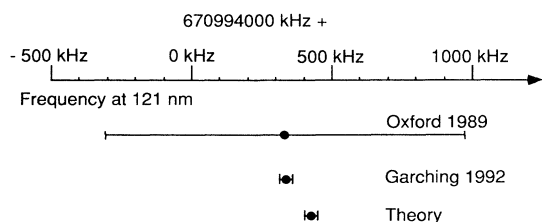


FIG. 3. Comparison of recent isotope shift measurements of the 1S-2S transition with the theoretical prediction.

ing experiments. Measurements of the ratio of e -H and e -D scattering [12] at low momentum transfer ($q=0.05$ to 0.5 fm⁻²) have the advantage of avoiding problems due to quark resonances [13,14] and of reducing systematic uncertainties of the experimental apparatus. The experimental result, the ratio of deuteron and hydrogen structure factor G_{ED}/G_{EP} , can be written as a product of terms:

$$G_{ED}/G_{EP} = (G_{EN} + G_{EP})(G_{EP})^{-1} C_E(q)(1 - q^2/8M_p^2),$$

with the structure factor of neutron-electron scattering G_{EN} , the deuteron structure factor $C_E(q)$, and the last term as a relativistic correction. This equation in momentum space corresponds to an equation for the charge radii:

$$\langle r_{D,\text{charge}}^2 \rangle = \langle r_{D,\text{structure}}^2 \rangle + \langle r_{H,\text{charge}}^2 \rangle - 6 \frac{\partial G_{EN}}{\partial q^2} \Big|_{q=0}.$$

Thus the difference in quadratic charge radii of deuterium and hydrogen depends only on the structure radius of the deuteron and the neutron-electron correction. If we take 1.9627(38) fm for the deuteron structure radius [15] and $0.0189(4)$ fm² for $\partial G_{EN}/\partial q^2$ (Ref. [16]), we calculate $3.739(17)$ fm² for the difference in quadratic charge radii. From this value we arrive at $670\,994\,414(22)$ kHz for the theoretical hyperfine corrected isotope shift (Fig. 4). The difference of $-77(31)$ kHz between our experimental value and the theoretical value can be interpreted as an energy shift of the 1S deuterium state due to a nuclear electric polarizability in the electric field of the 1S electron. Experiments of elastic deuteron-²⁰⁸Pb scattering have measured the deuteron polarizability $\alpha=0.70(5)$ fm³ (Ref. [17]), in fairly good agreement with theoretical predictions.

With a simple theoretical model that assumes for the neutron-proton interaction a square well potential, the second-order correction to the 1S energy was estimated to be of the order of -20 kHz. The main uncertainties of the calculation are caused by the relatively crude model for the deuteron. Details of this calculation will be given

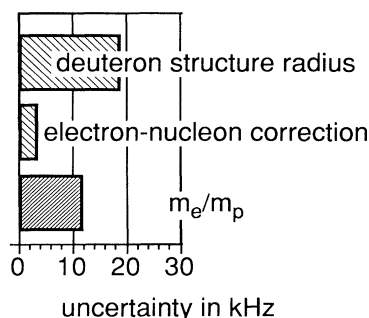


FIG. 4. Uncertainties in the theoretical prediction of the isotope shift.

elsewhere [5].

Several improvements now in preparation should soon reduce the experimental uncertainty of the $1S$ - $2S$ isotope shift to about 1 kHz. The present experimental error is mainly due to the drifts of the reference cavity. This drift can be avoided by locking laser 2 to the output frequency of a recently completed laser frequency chain that transports the accuracy of the cesium standard up to the blue laser frequency of 661 THz [18]. The uncertainties due to the second-order Doppler shifts can be significantly reduced by cooling the atomic beam to lower temperatures. In recent experiments near liquid-helium temperature, the resolution of the $1S$ - $2S$ transition has reached $\Delta\nu/\nu=7\times 10^{-12}$.

In conclusion, recent advances in the accuracy of spectroscopic measurements are providing new probes for the structure of the deuteron. They rival the accuracy of some measurements performed with electron-nucleon colliders and allow studies of nuclear features in the low energy domain. An accurate mass ratio of electron and proton can be determined together with other complementing experiments. If the ultimate resolution of 1.3 Hz could be reached, for instance in an atomic fountain experiment [19], the $1S$ - $2S$ isotope shift could, in principle, be determined to within better than 1 part in 10^{14} .

We want to thank K. Pachucki for essential help in the calculation of the energy shift due to the deuteron polarizability. We also acknowledge valuable discussions with C. Zimmermann and expert technical assistance by W. Simon and H. Mader. This work was supported in part by the Deutsche Forschungsgemeinschaft.

-
- [1] C. Zimmermann, R. Kallenbach, and T. W. Hänsch, *Phys. Rev. Lett.* **65**, 571 (1990).
 - [2] F. Schmidt-Kaler, D. Leibfried, C. Zimmermann, and T. W. Hänsch (to be published).
 - [3] D. Leibfried, F. Schmidt-Kaler, M. Weitz, and T. W.

- Hänsch, *Appl. Phys. B* **56**, 65 (1993).
- [4] M. G. Boshier, P. E. G. Baird, C. J. Foot, E. A. Hinds, M. D. Plimmer, D. N. Stacey, J. B. Swan, D. A. Tate, D. M. Warrington, and G. K. Woodgate, *Phys. Rev. A* **40**, 6169 (1989).
- [5] K. Pachucki *et al.* (to be published).
- [6] D. Taqqu (private communication).
- [7] F. Schmidt-Kaler, Ph.D. thesis, LMU-München, 1992 (unpublished).
- [8] R. Kallenbach, B. Scheumann, C. Zimmermann, D. Meschede, and T. W. Hänsch, *Appl. Phys. Lett.* **54**, 1622 (1989).
- [9] For the hydrogen $1S$ state: H. Hellwig, R. F. C. Vessot, M. W. Levine, P. W. Zizewitz, D. W. Allan, and D. J. Glaze, *IEEE Trans. Instrum. Measure.* **19**, 200 (1970); L. Essen, R. W. Donaldson, M. J. Bangham, and E. G. Hope, *Nature (London)* **229**, 110 (1971); for the hydrogen $2S$ state: D. J. Wineland and N. F. Ramsey, *Phys. Rev. A* **5**, 821 (1972); for the deuterium $1S$ state: J. Heberle, H. A. Reich, and P. Kusch, *Phys. Rev.* **101**, 612 (1956); for the deuterium $2S$ state: H. A. Reich, J. Heberle, and P. Kusch, *Phys. Rev.* **104**, 1585 (1956).
- [10] E. R. Cohen and B. N. Taylor, in *Codata Bulletin* (Perгамon, New York, 1986), Vol. 63.
- [11] M. Weitz, Ph.D. thesis, LMU-München, 1992 (unpublished).
- [12] R. W. Berard, F. R. Buskirk, E. B. Dally, J. N. Dyer, X. K. Maruyama, R. L. Topping, and T. J. Traverso, *Phys. Lett.* **47B**, 355 (1973).
- [13] L. H. Hand, D. J. Miller, and R. Wilson, *Rev. Mod. Phys.* **35**, 335 (1963).
- [14] G. Simon, Ch. Schmitt, F. Borkowski, and V. H. Walther, *Nucl. Phys.* **A333**, 381 (1980).
- [15] T. E. O. Ericson, *Nucl. Phys.* **A416**, 281c (1984).
- [16] V. E. Krohn and G. R. Ringo, *Phys. Rev.* **148**, 1303 (1966).
- [17] N. L. Rodning, L. D. Knutson, W. G. Lynch, and M. B. Tsang, *Phys. Rev. Lett.* **49**, 909 (1982).
- [18] T. Andreae, W. König, R. Wynands, D. Leibfried, F. Schmidt-Kaler, C. Zimmermann, D. Meschede, and T. W. Hänsch, *Phys. Rev. Lett.* **69**, 1923 (1992).
- [19] R. G. Beausoleil and T. W. Hänsch, *Opt. Lett.* **10**, 547 (1985).