Hydrogen-Deuterium 1*S*-2*S* Isotope Shift and the Structure of the Deuteron

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We have determined the hydrogen-deuterium isotope shift of the 1*S*-2*S* two-photon resonance with a 150-fold increase in accuracy compared to previous measurements. Our experimental result of 670 994 334.64(15) kHz has been obtained with an optical frequency comb generator and a CH_4-stabilized He-Ne reference laser which provides the frequency stability of our spectrometer while switching between the isotopes. From this measurement we derive values for the difference of the mean square charge radii of the proton and deuteron $r_d^2 - r_p^2 = 3.8212(15)$ fm^2, and for the deuteron structure radius $r_{\mathrm{str}} = 1.975\,35(85)$ fm. [S0031-9007(97)05204-6]

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The deuteron is the simplest compound nucleus and provides an important testing ground for theories of nuclear few-body systems which are beginning to accurately predict the deuteron structure radius [1]. This radius has traditionally been determined from accelerator-based electron scattering experiments [2–4]. In 1993 optical spectroscopic measurements of the hydrogen-deuterium isotope shift of the 1*S*-2*S* resonance [5] reached a precision sufficient to derive a new value for the deuteron structure radius [6] which deviates significantly from the electron scattering results.

Here, we report on a new precision measurement of the H-D isotope shift of the 1*S*-2*S* interval, which exceeds the accuracy of our earlier experiment by more than 2 orders of magnitude. The high resolution and measurement accuracy in our experiment allows us to clearly observe nuclear size effects that contribute only at the level of about 10^{-5} to the total isotope shift of the 1*S*-2*S* transition. Because of the fact that many QED effects cancel in the isotope shift, the difference of the mean square charge radii between deuteron and proton can be accurately derived. However, this requires a careful theoretical analysis of nuclear structure effects. At this precision level we have to give a careful definition of the charge radius, since the description of the nucleus by an elastic form factor is no longer sufficient. The recent theoretical progress is described in the second part of this Letter. The complete theoretical description together with our tabletop atomic physics experiment allows us to reach much higher accuracy for the deuteron structure radius than past measurements with electron-nucleon colliders.

Our experimental setup is shown in Fig. 1. The hydrogen spectrometer, indicated at the top of this figure, has been described previously in detail [7]. The 1*S*-2*S* transition in both isotopes is driven in a cold atomic beam by longitudinal Doppler-free two-photon excitation ($F = 1 \rightarrow 1$, $m_F = \pm 1 \rightarrow \pm 1$ in hydrogen and $F = 3/2 \rightarrow 3/2$, $m_F = \pm 3/2 \rightarrow \pm 3/2$ in deuterium). The radiation of an ultrastable dye laser at 486 nm is frequency doubled. The resulting UV radiation at 243 nm is resonantly enhanced in a linear cavity inside a vacuum chamber. A mixture of hydrogen and deuterium atoms from a gas discharge emerges from a liquid helium cooled nozzle. Cold atoms, traveling along the laser field, are excited into the metastable 2*S* state. At the end of the interaction zone an electric quenching field forces the emission of Lyman-α

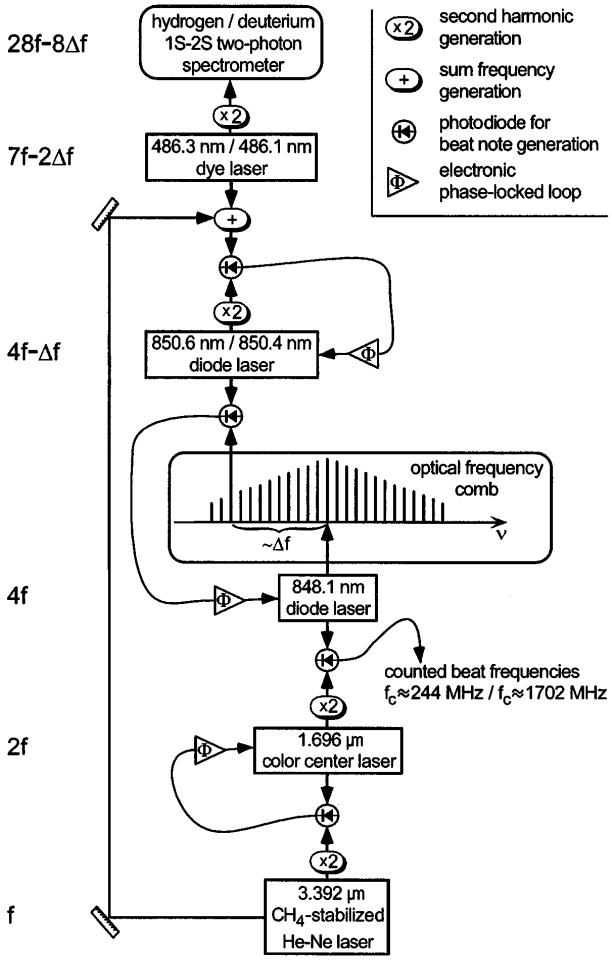

FIG. 1. Phase-coherent frequency chain for the comparison of the 1*S*-2*S* transition frequencies in hydrogen and deuterium.

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photons, which are detected by a photomultiplier. We apply a time-delayed measurement technique selecting slow atoms to reduce transit time broadening and the relativistic Doppler shift [7].

Figure 1 also shows the frequency chain that has been described in part in [8], where an absolute measurement of the $1S$-$2S$ frequency has been reported. A new element, the optical frequency comb (OFC) generator [9] is added to allow a rapid variation of the bridged frequency gap in order to switch between hydrogen and deuterium. This device creates a broad comb of equally spaced frequencies by efficient electro-optic modulation in a monolithic optical resonator. With a modulation frequency $f_{\mathrm{OFC}} = 6.3418$ GHz, our OFC has a width of 3.5 THz. As in our previous experiments, we exploit the close coincidence of the 28th subharmonic of the $1S$-$2S$ transition frequency with the absolute frequency of the CH_4-stabilized He-Ne laser. However, this laser now serves only as a flywheel to maintain the position of the OFC in frequency space, while the dye laser frequency is switched between the excitation wavelengths of the two isotopes. In this way, the absolute frequency of the He-Ne standard cancels in our result, while its frequency stability determines the uncertainty. The typical time for measuring and switching between isotopes is about 1000 s. The Allan variance for this time has been determined to be around 10^{-13} using a frequency chain at the Physikalisch Technische Bundesanstalt (PTB) in Braunschweig [10]. To transfer the stability of the He-Ne laser to the 850 nm region where the OFC operates we create its fourth harmonic with the help of a NaCl:OH^- color center laser. The frequency doubled output of this laser provides the carrier frequency for the OFC generation. Intermediate lasers are used after each nonlinear optical device to regain sufficient power for the next step. To phase-coherently connect the lasers we use optical phase-locked loops [11]. The frequency chain relates the difference in the counted radio frequency Δf_c to the isotope shift $(f^D_{1S\text{-}2S} - f^H_{1S\text{-}2S})$ according to

$$\Delta f_{\mathrm{exp}} = 8\Delta f_c + 104 f_{\mathrm{OFC}} - \Delta f_{\mathrm{HFS}}\,, \qquad (1)$$

where $\Delta f_{\mathrm{HFS}} = 215\,225\,585(14)$ Hz accounts for the well known hyperfine structure in both isotopes [12].

Figure 2 shows $1S$-$2S$ excitation spectra recorded versus the detuning from selected modes of the OFC generator. The detuning is $8 \times (f_c - 244.306\ \mathrm{MHz})$ for hydrogen and $8 \times (f_c - 1702.1\ \mathrm{MHz})$ for deuterium. We determine the line center by fitting a Lorentzian to the experimental data. Numerical simulations [13] confirm that this procedure is sufficient at the present level of accuracy, even though the expected line shape is not exactly Lorentzian. From the radio frequency f_c of the line center we calculate the H-D isotope shift according to Eq. (1).

As an example, a series of alternating measurements in hydrogen and deuterium is shown in Fig. 3. The residual frequency uncertainty of the measured line centers is

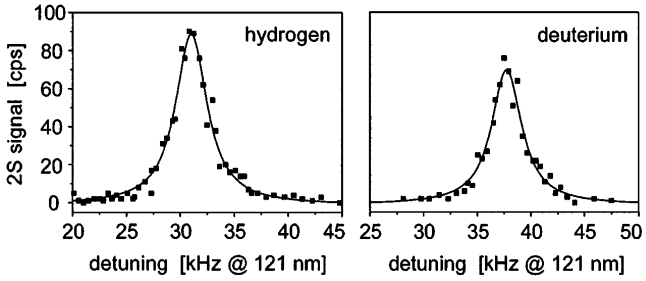

FIG. 2. Experimental $1S$-$2S$ spectra in hydrogen and deuterium as measured relatively to selected modes of the optical comb generator.

limited by the short term frequency stability of the He-Ne standard. We determine the isotope shift by fitting a pair of parallel lines to the hydrogen and deuterium data, thus accounting for a linear frequency drift of the standard. Since the coefficient of the ac Stark shift is the same in hydrogen and deuterium [14], we kept the power of the exciting UV light at 243 nm constant. Further systematic effects such as dc Stark and pressure shifts do not play a significant role at the present level of accuracy. A realistic model for the line shape [13] predicts that the relativistic Doppler shift of the line center is about 80 Hz in hydrogen for the chosen delay time. Even though we expect the same Doppler shift for deuterium due to our time-delayed signal detection, we allow for an uncertainty of 20 Hz to account for a possible difference. After averaging the results of 10 measurements like the one shown in Fig. 3, we obtain the experimental result for the $1S$-$2S$ H-D isotope shift,

$$\Delta f_{\mathrm{exp}} = 670\,994\,334.64(15)\ \mathrm{kHz}\,. \qquad (2)$$

The uncertainty of 150 Hz is dominated by the frequency fluctuations of the CH_4-stabilized He-Ne standard.

At this precision level the theoretical contributions to the isotope shift must be reanalyzed. Although the majority of QED effects are the same for hydrogen and deuterium, an accurate determination of the difference in the nuclear charge radii from the H-D isotope shift depends on both QED and non-QED effects. Therefore, a large

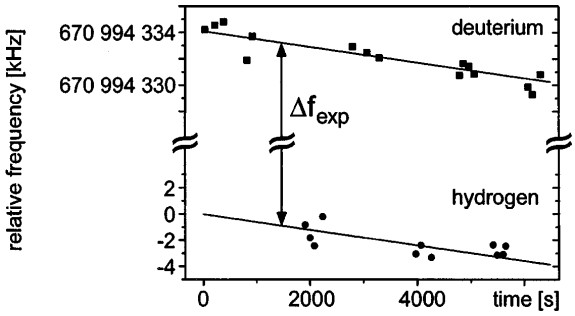

FIG. 3. Alternating measurements of the $1S$-$2S$ transition frequency in hydrogen and deuterium. The dots stand for line centers of single spectra as shown in Fig. 2.

effort was undertaken to recalculate higher order recoil corrections, resulting in the identification of some new effects. In the following we present a short description of all known contributions to the H-D isotope shift which contribute at the level of 1 kHz. For details we refer to the review of Sapirstein and Yennie in [15] and the recent update in [6].

Most of the H-D isotope shift of the $1S$-$2S$ interval is caused by the different masses of the nuclei. The leading effects are obtained in the Breit treatment,

$$E = \mu[\mathcal{E}(n,j) - 1] - \frac{\mu^2}{2(M+m)}[\mathcal{E}(n,j) - 1]^2, \tag{3}$$

and amount to 671 004 073.56 kHz. Here $\mathcal{E}(n,j)$ denotes the dimensionless Dirac energy, and m, M, and μ are the electron, nucleus, and reduced mass, respectively. This formula does not account for the fact that the deuteron has spin 1. The relativistic corrections may depend on the spin, and in our case one has to subtract the so-called Darwin-Foldy correction $\Delta E = -\mu^3\alpha^4/2M^2n^3$, which is included by convention [16] in the deuteron charge radius, but not in the proton charge radius. It contributes 11.37 kHz to the $1S$-$2S$ frequency in deuterium. There are many other effects that result from the nuclear mass dependence of the Lamb shift called recoil corrections. They are first calculated under the assumption that the nucleus is a pointlike particle with spin $1/2$. Corrections beyond this assumption are considered separately. The recoil corrections give $-4\,536.16$ kHz to the isotope shift. The uncertainty due to yet unknown corrections of the order $m^2/M\alpha^7\ln^2(\alpha^{-2})/\pi$ are estimated to be 1 kHz by assuming that the coefficient is equal to 1 (we correct our previous estimate in [6]).

There are conceptual problems with the proton self-energy correction because it modifies the proton form factors. To incorporate this correction unambiguously we must precisely specify the meaning of the nuclear mean square charge radius. The standard definition is based on the Sachs form factor G_E which, in the nonrelativistic limit, is the Fourier transform of the charge density distribution

$$\frac{\langle r^2\rangle}{6} = \frac{\partial G_E(q^2)}{\partial(q^2)}\bigg|_{q^2=0}. \tag{4}$$

This definition is not appropriate at our precision level, because radiative corrections to G_E are infrared divergent. We follow the enhanced definition proposed in [17], which is based on the forward scattering amplitude described by

$$T^{\mu\nu}(x - x') = -i\langle t|Tj^\mu(x)\,j^\nu(x')\,|t\rangle, \tag{5}$$

where $t = (M,0,0,0)$ and j^μ is the 4-vector current of the nucleon components. In the nonrelativistic limit the T^{00} component contains a logarithmic singularity. This term occurs because the nucleus has a charge and does not depend on other details, such as spin. Once it is subtracted from T^{00} the remainder has a proper analytical behavior. At low momentum transfer it could be described by one quantity, the nuclear charge radius. The associated correction to the energy for S states has the form

$$\Delta E = \frac{2}{3n^3}(Z\alpha)^4\mu^3\langle r^2\rangle + \frac{4(Z^2\alpha)(Z\alpha)^4}{3\pi n^3}\frac{\mu^3}{M^2} \times\left[\ln\left(\frac{M}{\mu(Z\alpha)^2}\right) - \ln k_0(n)\right], \tag{6}$$

where $\ln k_0(n)$ is the Bethe logarithm and Z denotes the nucleus charge. The first term is due to the finite nuclear size, while the second term comes from the proton self-energy and contributes 2.98 kHz to the H-D isotope shift.

The correction of the order of $(Z\alpha)^2$ to the finite size effect is also significant due to the logarithmic singularity of the relativistic wave function at the origin and amounts to -2.79 kHz for the H-D isotope shift [16]. The combination of radiative corrections and finite size gives a small contribution of 0.60 kHz. It is obtained from the radiatively corrected electron charge density at the origin [18]. The nuclear structure effects beyond Eq. (6) are well described by the two-photon scattering amplitude at zero momentum

$$\Delta E = -\frac{e^4}{2}\phi^2(0)\int\frac{d^4q}{(2\pi)^4 i}\frac{1}{q^4} \times[T^{\mu\nu} - t^{\mu\nu}(M)]\,t_{\mu\nu}(m), \tag{7}$$

where $t_{\mu\nu}(m)$ is a bare amplitude (without radiative corrections) for a pointlike spin $1/2$ particle of mass m. This amplitude requires subtraction of the singularities connected with the terms already included: relativistic, self-energy, and finite size corrections. Since recoil effects have been calculated with the assumption that the nucleus has spin $1/2$, we subtracted the corresponding amplitude for a pointlike particle with the mass equal to the deuteron mass in Eq. (7). The main contribution comes from small momenta and is associated with the deuteron polarizability. The binding energy of the deuteron of 2.2 MeV is comparable to the electron rest energy, giving rise to a polarizability correction. The most recent analysis of Friar *et al.* [16] gives 18.58(7) kHz for the $1S$-$2S$ frequency in deuterium. The corrections beyond this polarizability term, i.e., the elastic part, have been estimated in the static nucleus limit [16], and they amount to 0.46 kHz. The yet uncalculated contributions coming from relativistic corrections to the proton-neutron interaction and the deuteron recoil enter at similar order of magnitude as retardation terms in the polarizability contribution. The second uncertainty is due to an additional photon exchange in the inelastic amplitude. We estimate the total uncertainty of Eq. (7) to 0.5 kHz. A detailed study of the deuteron structure corrections as described by Eq. (7) would be very desirable.

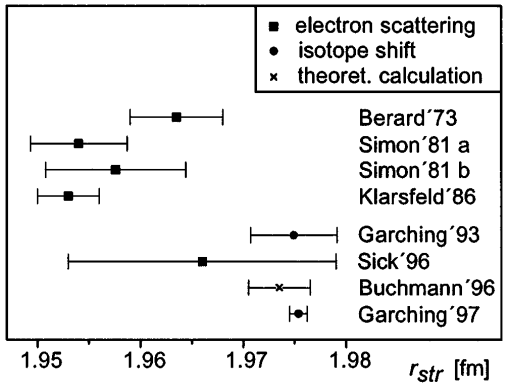

FIG. 4. Values for the deuteron structure radius. See Refs. [2,3] (*a* and *b* stand for a fit with third and fourth order polynomials) [4,5,23,24].

We calculate the theoretical isotope shift *excluding* nuclear size effects as

$$\Delta f_{\mathrm{th}} = 670\,999\,568.6(1.5)\,(1.5)\ \mathrm{kHz}, \tag{8}$$

where the first uncertainty comes from the electron-proton mass ratio and the second is the theoretical uncertainty. In this calculation we have used the constants given in references as follow: α [19], M_p/m [20], M_d/M_p [21], and R_∞ [8]. The difference

$$\Delta f_{\mathrm{th}} - \Delta f_{\mathrm{exp}} = 5234.0(2.1)\ \mathrm{kHz} \tag{9}$$

between the theoretical and the experimental values is caused by the nuclear finite size effect, which is given by the first term of Eq. (6). From this we derive the difference of the deuteron-proton mean square charge radii

$$r_d^2 - r_p^2 = 3.8212(15)\ \mathrm{fm}^2. \tag{10}$$

The deuteron structure radius is $r_{\mathrm{str}}^2 = (r_d^2 - r_p^2) - r_n^2 - 3/4M_p^2$ [4], where $r_n^2 = -0.114(3)\ \mathrm{fm}^2$ [22] is the neutron charge radius and M_p the proton mass. From this equation one obtains

$$r_{\mathrm{str}} = 1.975\,35(85)\ \mathrm{fm}. \tag{11}$$

The precision of atomic spectroscopy measurements of the deuteron radius is thus exceeding the accuracy of previous measurements based on elastic electron scattering by an order of magnitude. Furthermore, our result is in disagreement with these measurements as can be seen from Fig. 4. The recent reanalysis of scattering data by Sick [23] indicates a possible agreement with our data, when including the second order Born approximation. We emphasize again at this point that our determination of the deuteron structure radius is requiring a careful analysis of the nuclear self-energy. We have incorporated some corrections beyond the Sachs form factor. The recent prediction of the deuteron structure radius by Buchmann *et al.* [24] is in very good agreement with the new value presented here.

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