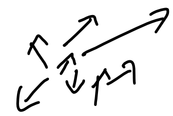
Optical Precision Measurements in Hydrogen and Determination of the Rydberg Constant

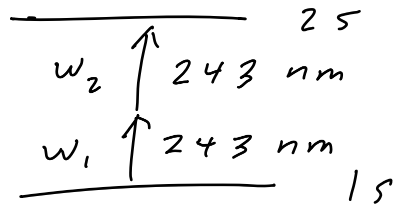


The primary problem is the Doppler shift: At 300K,

$$\langle v \rangle_H \simeq 2500 \text{ m/s, so}$$

$$\frac{\Delta \nu}{\nu} \simeq \frac{\Delta v}{v} = 8.4 \times 10^{-6}.$$

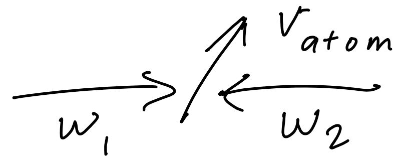
This can be eliminated by Doppler-free two-photon spectroscopy:



We will derive the transition rates in Physics 338. The required powers are in the kW range.

If the beams counterpropagate, the Doppler shifts cancel, as originally pointed out by Chebotaev. If the two frequencies ω_1 and ω_2 are equal,

$$\left(\frac{\Delta \nu}{\nu}\right)_1 = -\left(\frac{\Delta \nu}{\nu}\right)_2.$$



This leaves only the $(\Delta v / v)^2$ terms, which are much smaller and can be estimated to the required accuracy. (This is sometimes called the "time dilation" correction.)

Some recent results for H, D include:

(1) 1*S*-2*S*. The group of Theodor Hänsch has worked on this measurement for many years. The most recent result is given in Niering, *et al.,* Phys. Rev. Lett. **84**, 5496 (2000). The newest result, relative to the center of mass of the hyperfine structure, is

$$\nu_{1S\text{-}2S} = 2466061413187.103(46) \text{ kHz}.$$

(2) 2*S*-8*S,D* and 2*S*-12*D*. These measurements have been performed in a series of experiments by the group of Biraben in Paris. The *n*=8 experiment is described in de Beauvoir, *et al.,* Phys. Rev. Lett. **78**, 440 (1997). The result given there is

$$\nu_{2S_{1/2}-8D_{5/2}} = 770\ 649\ 561.5850\ (49) \text{ MHz}$$

A survey of their work, including their *n*=12 results as well as a comparison of 1*S*-3*S* with 2*S*-6*S/D*, is given in a 2000 review article (see below).

Measurement of the Hydrogen 1S-2S Transition Frequency by Phase Coherent Comparison with a Microwave Cesium Fountain Clock

M. Niering, R. Holzwarth, J. Reichert, P. Pokasov,* Th. Udem, M. Weitz, and T. W. Hänsch
Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, 85748 Garching, Germany

P. Lemonde,[1] G. Santarelli,[1] M. Abgrall,[1] P. Laurent,[1] C. Salomon,[2] and A. Clairon[1]
[1]*BNM-LPTF, Observatoire de Paris, 61 Avenue de l'Observatoire, 75014 Paris, France*
[2]*Laboratoire Kastler Brossel, ENS, 24 rue Lhomond, 75005 Paris, France*
(Received 6 March 2000)

We report on an absolute frequency measurement of the hydrogen 1S-2S two-photon transition in a cold atomic beam with an accuracy of 1.8 parts in 10^{14}. Our experimental result of 2 466 061 413 187 103(46) Hz has been obtained by phase coherent comparison of the hydrogen transition frequency with an atomic cesium fountain clock. Both frequencies are linked with a comb of laser frequencies emitted by a mode locked laser.

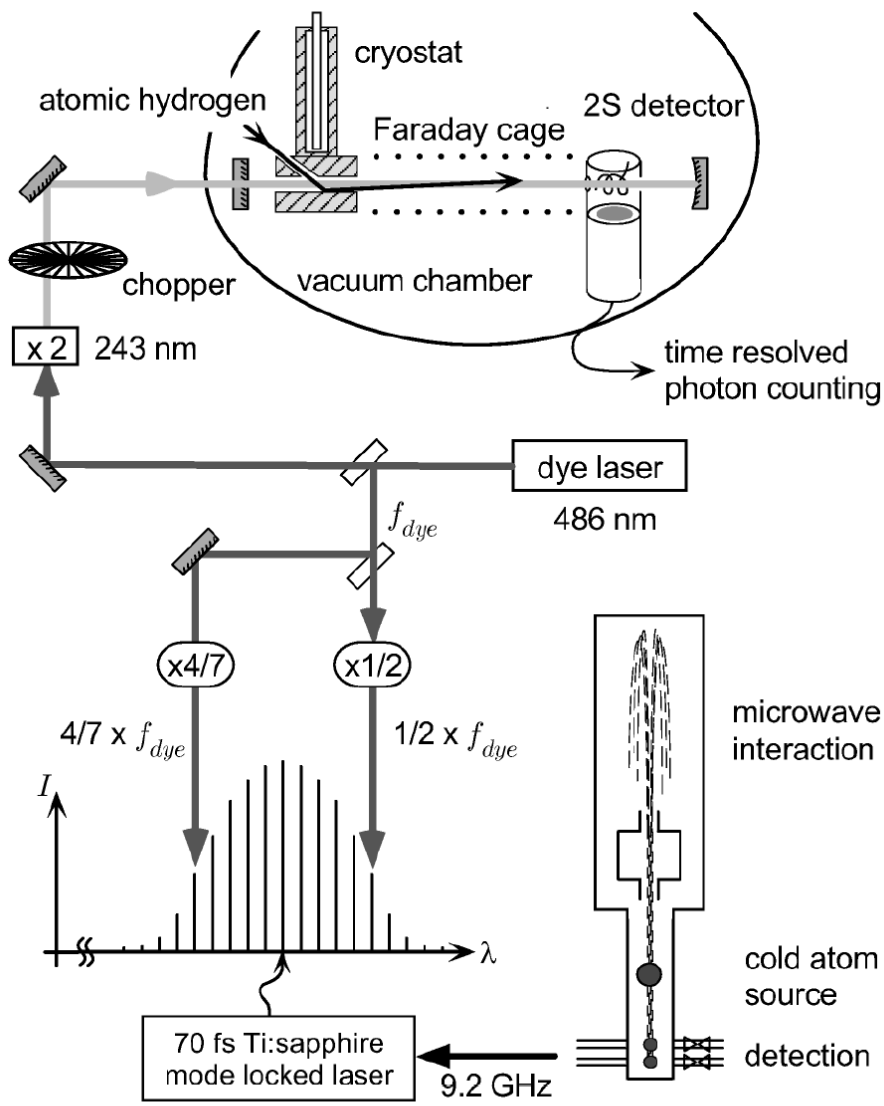


FIG. 1. Experimental setup for comparison of the hydrogen 1S-2S transition frequency with an atomic cesium fountain clock.

Here is the experimental scheme from de Beauvoir, *et al*., "Absolute Frequency Measurement of the 2*S*-8*S/D* Transitions in Hydrogen and Deuterium: New Determination of the Rydberg Constant," Phys. Rev. Lett. **78**, 440 (1997):

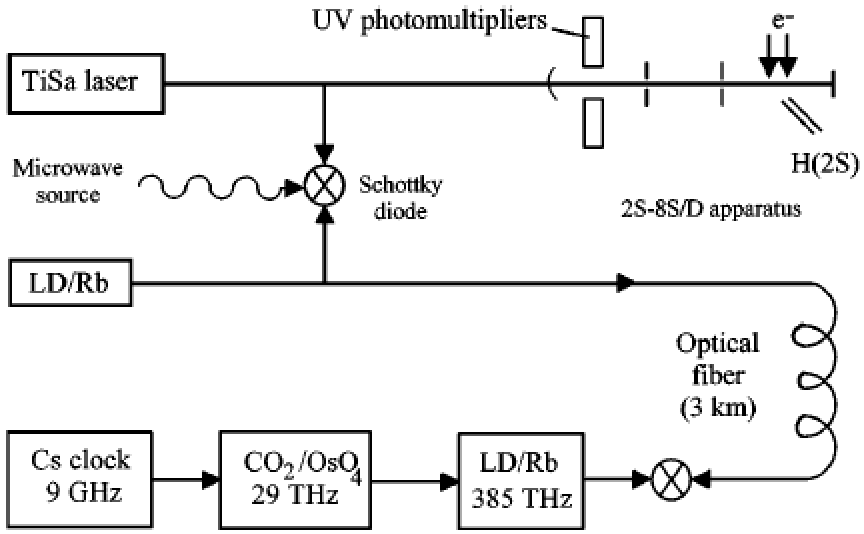


FIG. 1. Outline of the frequency chain between the 2*S*-8*S*/*D* hydrogen frequencies and the cesium clock.

The same group has measured the 2*S*-12*D* transition, and they have also compared 1*S*-3*S* with 2*S*-6*S*/6*D*. The scheme for the 2*S*-12*D* measurement is shown in their 2000 review article (see next page for the title and reference information):

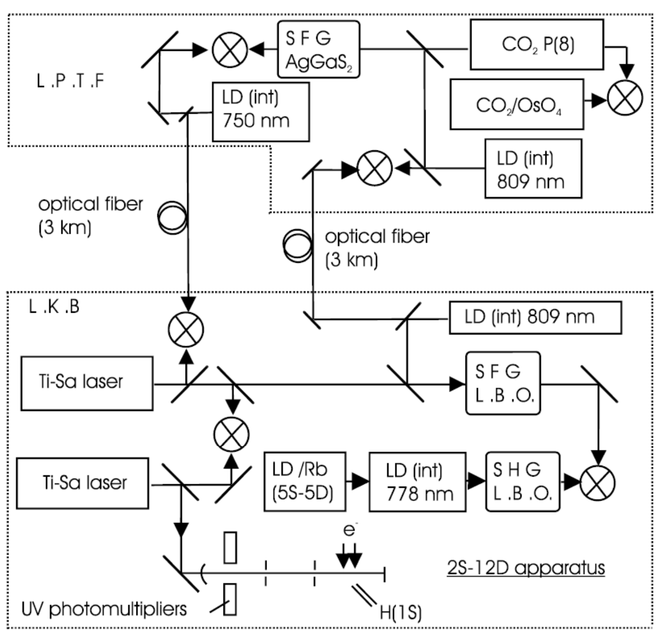


Fig. 17. Outline of the frequency chain between the 2S–12D hydrogen frequencies and the LD/Rb and CO_2/OsO_4 standards. The details are explained in the text (Ti-Sa: titanium sapphire laser, LD/Rb: rubidium stabilized laser diode, LD(int): intermediate laser diode, CO_2/OsO_4: osmium tetraoxyde stabilized CO_2 laser, SHG: second harmonic generation, SFG: sum frequency generation).

An excellent overview of modern hydrogen metrology was published by Biraben's group:

Eur. Phys. J. D **12**, 61–93 (2000)

THE EUROPEAN PHYSICAL JOURNAL D
EDP Sciences

Metrology of the hydrogen and deuterium atoms: Determination of the Rydberg constant and Lamb shifts

B. de Beauvoir[1], C. Schwob[1], O. Acef[2], L. Jozefowski[1], L. Hilico[1,a], F. Nez[1], L. Julien[1], A. Clairon[2], and F. Biraben[1,b]

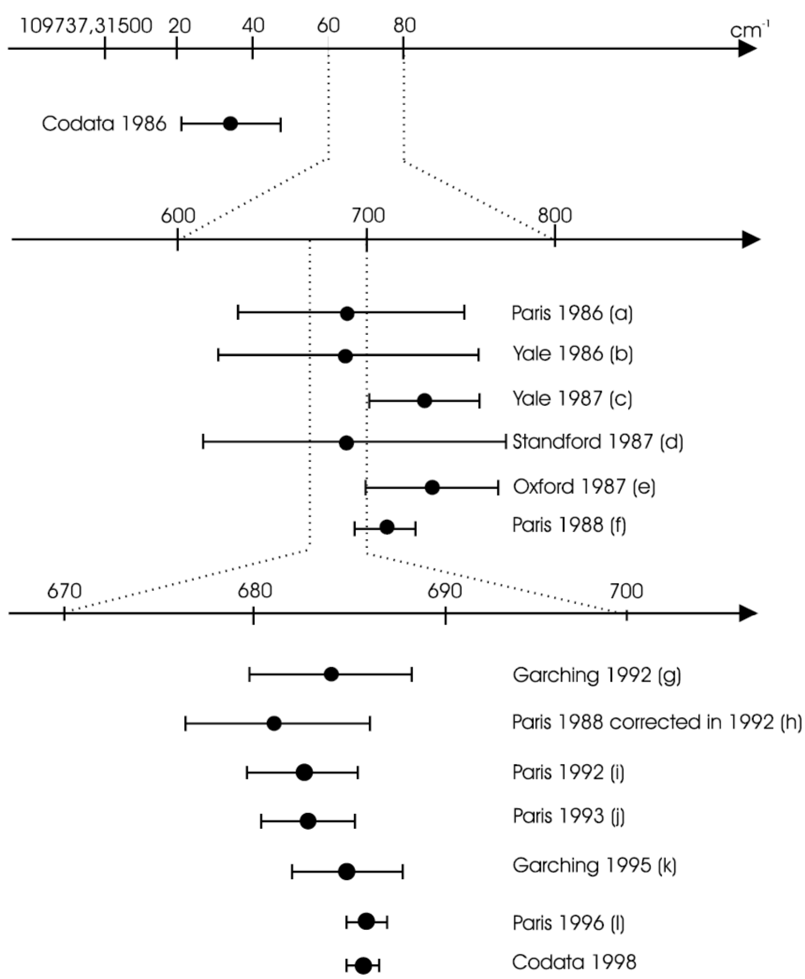


Fig. 22. Comparison of various determinations of the Rydberg constant since the 1986 adjustment of the fundamental constants; Codata 1986 [70], a [71], b [72], c [73], d [74], e [48], f [75], g [76], h: reference [75] corrected for the new measurement of the He–Ne/I_2 standard laser [33], i [4], j [5], k [42], l [6], Codata 1998 [44].

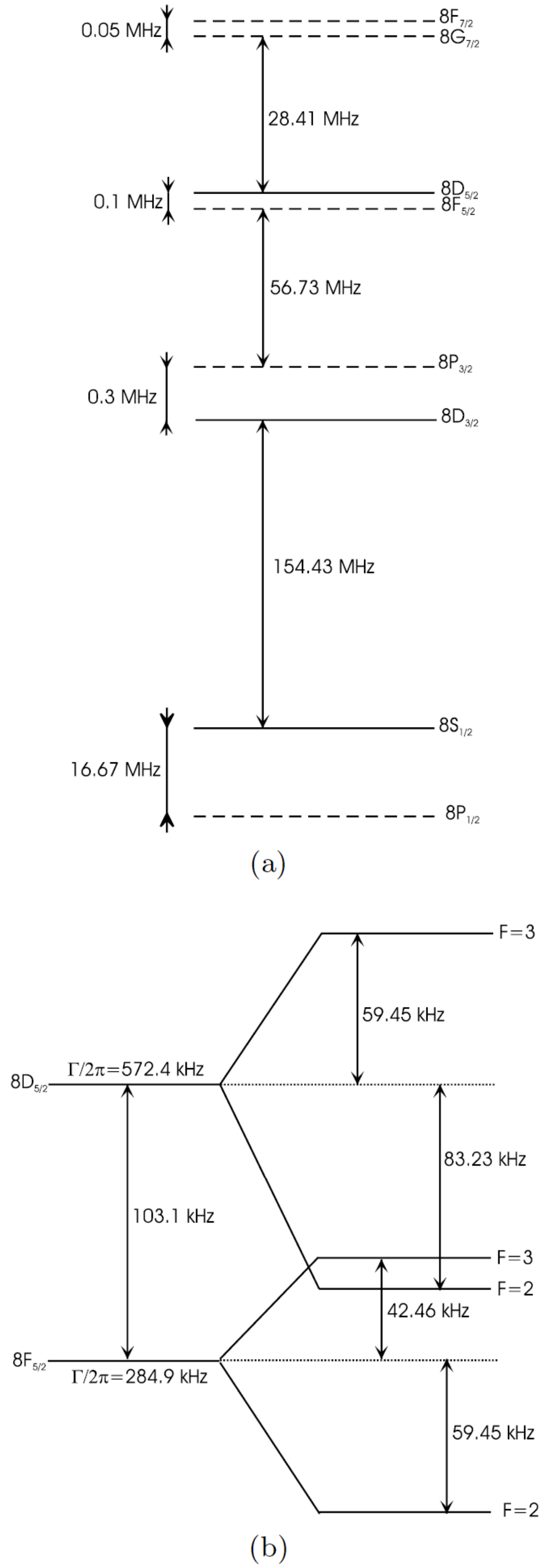


Fig. 12. (a) Fine structure and Lamb shifts of the 8S, 8P, 8D and 8F levels. The solid line corresponds to the levels which are excited with a two-photon transition from the metastable state. (b) Hyperfine structure of the $8D_{5/2}$ and $8F_{5/2}$ levels in hydrogen.

To see how the Rydberg constant can be obtained from the several recent spectroscopic results, the relationships between the various energy intervals are needed. The procedure is reviewed in detail in the Eur. Phys. J. D review, and is briefly summarized here. The energy $E_{\mathrm{H}}(nLJ)$ of a level $|jnLJ\rangle$ of hydrogen is:

$$E_{\mathrm{H}}(nLJ) = d_{\mathrm{H}}(nLJ)hcR_\infty + r_{\mathrm{H}}(n)hcR_\infty + hL_{\mathrm{H}}(nLJ) \tag{35}$$

where $d_H(nLJ)hcR_\infty$ and $r_H(n)hcR_\infty$ are the Dirac and recoil energies, which can be written in exact form in terms of the fine structure constant α and the electron:proton mass ration m_e/m_p. The Lamb shift $L_{\mathrm{H}}(nLJ)$ must be determined from experimental data, together with the Rydberg constant R_∞. The key relationships, which can be used in various combinations, are:

From theoretical work,

$$L_{\mathrm{H}}(1\mathrm{S}_{1/2}) - 8L_{\mathrm{H}}(2\mathrm{S}_{1/2}) = -187.232(5)\ \mathrm{MHz} \qquad (36)$$

$$L_{\mathrm{D}}(1\mathrm{S}_{1/2}) - 8L_{\mathrm{D}}(2\mathrm{S}_{1/2}) = -187.225(5)\ \mathrm{MHz} \qquad (37)$$

From the Paris 2*S*-8*D*, 12*D* experiments, where we define for hydrogen (H) or deuterium (D), $\nu_{H,D}(2S_{1/2} - nD_{5/2}) \equiv a_{H,D}(2S_{1/2} - nD_{5/2})cR\infty + L_{H,D}(nD_{5/2}) - L_{H,D}(2S_{1/2})$, we have,

$$\nu_A(2\mathrm{S}_{1/2}{-}n\mathrm{D}_{5/2}) = a_A(2\mathrm{S}_{1/2}{-}n\mathrm{D}_{5/2})cR_\infty + L_A(n\mathrm{D}_{5/2}) - L_A(2\mathrm{S}_{1/2}) \qquad (4\ \text{equations}). \quad (38)$$

and from the Paris 1*S*-3*S* and 2*S*-6*S*,6*D* comparisons,

$$\nu_{\mathrm{H}}(2\mathrm{S}_{1/2}{-}6\mathrm{D}_{5/2}) - \frac{1}{4}\nu_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}3\mathrm{S}_{1/2}) = \left[a_{\mathrm{H}}(2\mathrm{S}_{1/2}{-}6\mathrm{D}_{5/2}) - \frac{1}{4}a_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}3\mathrm{S}_{1/2})\right] cR_\infty + L_{\mathrm{H}}(6\mathrm{D}_{5/2}) - L_{\mathrm{H}}(2\mathrm{S}_{1/2}) - \frac{1}{4}\left(L_{\mathrm{H}}(3\mathrm{S}_{1/2}) - L_{\mathrm{H}}(1\mathrm{S}_{1/2})\right). \tag{39}$$

We can either fit the 2*S* Lamb shift or take the best previous experimental value,

$$L_{\mathrm{H}}(2\mathrm{S}_{1/2}) = 1\,045.009\,4(65)\ \mathrm{MHz}. \qquad (40)$$

From the work of Hänsch's group,

$$\nu_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2}) = a_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2})cR_\infty + L_{\mathrm{H}}(2\mathrm{S}_{1/2}) - L_{\mathrm{H}}(1\mathrm{S}_{1/2}), \quad (41)$$

$$\nu_{\mathrm{D}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2}) - \nu_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2}) = \left[a_{\mathrm{D}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2}) - a_{\mathrm{H}}(1\mathrm{S}_{1/2}{-}2\mathrm{S}_{1/2})\right] cR_\infty + L_{\mathrm{D}}(2\mathrm{S}_{1/2}) - L_{\mathrm{H}}(2\mathrm{S}_{1/2}) - L_{\mathrm{D}}(1\mathrm{S}_{1/2}) + L_{\mathrm{H}}(1\mathrm{S}_{1/2}). \tag{42}$$

Also from Hänsch's group, as well as related work by Boshier and Hinds, we have the following Lamb shift relationships (if the theoretical Lamb shifts are used for n=4):

$$L_{\mathrm{H}}(1\mathrm{S}_{1/2}) - 5L_{\mathrm{H}}(2\mathrm{S}_{1/2}) = 2\,947.831(37)\ \mathrm{MHz} \quad (1\mathrm{S}-2\mathrm{S}\ \text{ and }\ 2\mathrm{S}-4\mathrm{S/D}\ \text{comparison}), \qquad (43)$$

$$L_{\mathrm{H}}(1\mathrm{S}_{1/2}) - 5L_{\mathrm{H}}(2\mathrm{S}_{1/2}) = 2\,947.787(34)\ \mathrm{MHz} \quad (1\mathrm{S}-2\mathrm{S}\ \text{ and }\ 2\mathrm{S}-4\mathrm{P}\ \text{comparison}). \qquad (44)$$

Putting this all together, the Rydberg constant can be evaluated using various combinations of the available optical data:

Table 20. Determination of the Rydberg constant.

method and transitions involved	equations	$(R_\infty - 109\,737)$ cm^{-1}
determination of R_∞ from the 2S–nD and 2S–2P measurements		
2S–2P and 2S–8S/D in hydrogen	(38, 40)	0.315 6861(13)
2S–2P and 2S–12D in hydrogen	(38, 40)	0.315 6848(13)
2S–2P, 2S–8S/D and 2S–12D in hydrogen	(38, 40)	0.315 6855(11)
determination of R_∞ from linear combination of optical frequencies measurements		
2S–8S/D, 1S–2S and $1/n^3$ law in hydrogen	(36, 38, 41)	0.315 6865(16)
2S–12D, 1S–2S and $1/n^3$ law in hydrogen	(36, 38, 41)	0.315 6842(17)
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen	(36, 38, 41)	0.315 6854(13)
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in deuterium	(37, 38, 41, 42)	0.315 6854(12)
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen and deuterium	(36–38, 41, 42)	0.315 6854(10)
general least squares adjustment in hydrogen and deuterium		
2S–2P, 2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law	(36–44)	0.315 685 50(84)

The "$1/n^3$ law" presumes the predicted scaling with n of many of the low-order terms in the *ns* Lamb shift. Thus the corresponding determinations are not purely experimental, although their reliance on theory is only on well-established principles.

Several other parameters can also be extracted from the analysis (in the generalized fit, five parameters are extracted from twelve results). Perhaps the most important is the ground-state Lamb shift (see next page):

Table 21. Determination of the $1S_{1/2}$ Lamb shift in hydrogen.

method and transitions involved	equations	$L_H(1S_{1/2})$ (MHz)
comparison of transition frequencies lying in a ratio 4:1		
2S–2P, 1S–3S and 2S–6S/D	(39, 40)	8 172.825(47)
2S–2P, 1S–2S and 2S–4S/D	(43, 40)	8 172.878(51)
2S–2P, 1S–2S and 2S–4P	(44, 40)	8 172.834(48)
comparison of the 1S–2S and 2S–nD frequencies using the $2S_{1/2}$ Lamb shift		
2S–2P, 1S–2S and 2S–8S/D	(38, 40, 41)	8 172.854(33)
2S–2P, 1S–2S and 2S–12D	(38, 40, 41)	8 172.825(34)
2S–2P, 1S–2S, 2S–8S/D and 2S–12D	(38, 40, 41)	8 172.840(31)
comparison of the 1S–2S and 2S–nD frequencies using the $1/n^3$ scaling law		
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen	(36, 38, 41)	8 172.837(32)
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen and deuterium	(36–38, 41, 42)	8 172.837(26)
general least squares adjustment in hydrogen and deuterium		
2S–2P, 2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law	(36–44)	8 172.840(22)
theory $r_p = 0.862(12)$ fm [56]		8 172.731(40)
theory $r_p = 0.805(11)$ fm [56]		8 172.582(40)

Note the discrepancy of at least 2.4 σ. This could indicate either (1) significant errors in the Lamb shift theory, perhaps because of still-uncalculated "two-loop" corrections, or (2) that the proton charge radius is larger than either of the existing measurements indicate; a value of $r_p =$ 0.901(16) fm would bring the Lamb shift into agreement with experiment. For deuterium, the results from Hänsch's group definitely provide the best value of the deuteron structure radius. Their value of 1.97535(85) fm (Phys. Rev. Lett. **80**, 468 (1998) is considerably larger than results from electron scattering, and about five times more accurate.

It is also possible to obtain an improved value for the classic Lamb shift interval in the $2S_{1/2}$ state from purely optical data, although the new result then depends heavily on the $1/n^3$ scaling law:

Table 23. Determination of the $2S_{1/2}$ Lamb shift in hydrogen.

method and transitions involved	equations	$\nu_H(2S_{1/2}-2P_{1/2})$ (MHz)
direct measurement of the $2S_{1/2}$–$2P_{1/2}$ splitting		
$2S_{1/2}$–$2P_{1/2}$, Newton *et al.* [77]		1 057.862(20)
$2S_{1/2}$–$2P_{1/2}$, Lundeen *et al.* [61]		1 057.845(9)
$2S_{1/2}$–$2P_{3/2}$, Hagley *et al.* [62,63]		1 057.842(12)
2S–2P, Wijngaarden et al. [64]		1 057.852(15)
comparison of transition frequencies lying in a ratio 4:1		
1S–3S, 2S–6S/D and $1/n^3$ scaling law	(36, 39)	1 057.841(10)
1S–2S, 2S–4S/D and $1/n^3$ scaling law	(36, 43)	1 057.857(12)
1S–2S, 2S–4P and $1/n^3$ scaling law	(36, 44)	1 057.842(11)
comparison of the 1S–2S and 2S–nD frequencies using the $1/n^3$ scaling law		
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen	(36, 38, 41)	1 057.8446(42)
2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law in hydrogen and deuterium	(36–38, 41, 42)	1 057.8447(34)
general least squares adjustment in hydrogen and deuterium		
2S–2P, 2S–8S/D, 2S–12D, 1S–2S and $1/n^3$ law	(36–44)	1 057.8450(29)
theory $r_p = 0.862(12)$ fm [56]		1 057.836(6)
theory $r_p = 0.805(11)$ fm [56]		1 057.812(6)