High Resolution Spectroscopy of Hydrogen

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1. Introduction

The hydrogen atom continues to hold fascinating challenges for spectroscopists. As the simplest of the stable atoms it has long permitted unique confrontations between basic theory and experiment. Very large further improvements in spectral resolution and accuracy appear feasible with the help of emerging new methods for stabilizing and measuring optical frequencies, together with advanced laser techniques for preparing, slowing, and manipulating atoms. Such prospects make more accurate quantum electrodynamic computations of hydrogen levels highly desirable. If theory is correct, measurements and comparisons of transition frequencies in hydrogen will yield precise new values of fundamental constants, in particular the Rydberg constant, the electron mass, and the charge radius of the proton. It would be much more exciting, of course, if we found out that theory fails at some level of scrutiny.

G. SERIES [1] has reminded us how spectroscopy of hydrogen has played a central role in the history of atomic physics and quantum mechanics. Seemingly minute discrepancies between theory and observation have more than once stimulated major conceptual breakthroughs. A good example is the spectral profile of the red Balmer-α line which presented one of the most tantalizing problems of atomic physics for more than a decade, in the 1930s and 40s. Unfortunately, the intricate fine structure always appeared blurred and smeared out by the Doppler-effect due to the random thermal motion of the light hydrogen atoms. Suspected deviations from the relativistic Dirac theory were only confirmed after the war with the observation of the 2S Lamb shift by radiofrequency spectroscopy of an atomic beam, a discovery that led to the development of quantum electrodynamics by Feynman, Schwinger, and Tomonaga.

2. Doppler-Free Spectroscopy and the Rydberg Constant

Major advances in optical high resolution spectroscopy became possible only in the early 1970s with the advent of monochromatic broadly tunable dye lasers [2] and powerful techniques of Doppler-free laser spectroscopy [3]. In the earliest successful method, saturation spectroscopy, the output of a tunable laser is split into a saturating beam and a probe beam which are sent in opposite directions through a cell with the absorbing gas. The saturating beam is strong enough to excite a noticeable fraction of the absorbing atoms so that it bleaches a path through the absorber. The probe beam can detect this bleaching if the laser is tuned to the center of a Doppler-broadened line so that both beams can interact with the same atoms, those that are standing still or are at most moving sideways.

Fig. 1 shows an early saturation spectrum of the hydrogen Balmer-α line, recorded in this way at Stanford [4], together with the seven theoretically predicted fine structure components and the Doppler-

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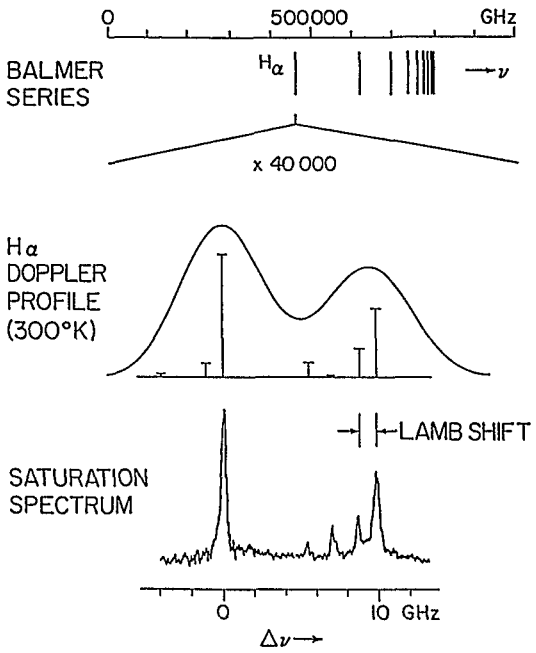

Fig. 1. Doppler-free saturation spectrum of the hydrogen Balmer-α line compared to theoretical fine structure and Doppler profile at room temperature [4].

broadened line profile at room temperature. We were thrilled to resolve the 2S Lamb shift for the first time directly in the Balmer-α spectrum. By measuring the absolute wavelength of the strongest $2P_{3/2}$-$3D_{5/2}$ component, we determined a new tenfold improved value of the Rydberg constant in 1974 [5]. This constant not only determines the binding energy between electron and nucleus, but plays an important role in its relationship to other fundamental physical constants [6].

This first laser measurement of the Rydberg constant has meanwhile been surpassed by numerous other experiments, studying several optical transitions in hydrogen and deuterium by different techniques of Doppler-free laser spectroscopy. Fig. 2 gives a summary of six recent measurements carried out at Yale [7,8], Paris [9,10], Stanford [11] and Oxford [12]. The accuracy of all these experiments is now limited by wavelength metrology. The most recent Rydberg measurement (D) has reached an accuracy of better than 2 parts in 10^{10}, determined only by the precision of the iodine-stabilized helium neon laser which is still the most accurate optical wavelength standard. This problem is the impetus behind recent efforts to measure the Rydberg constant by observing microwave transitions between circular Rydberg states [13]. Nonetheless, with its newly reached precision, the Rydberg constant is already the most accurately known of the fundamental constants except for the g-factor of the electron.

Other contributions in this volume give detailed accounts of the work at Yale, Paris, and Oxford. This article will therefore focus on the past experiments of our laboratory at Stanford and on our current and future program at the Max-Planck-Institute for Quantum Optics in Garching.

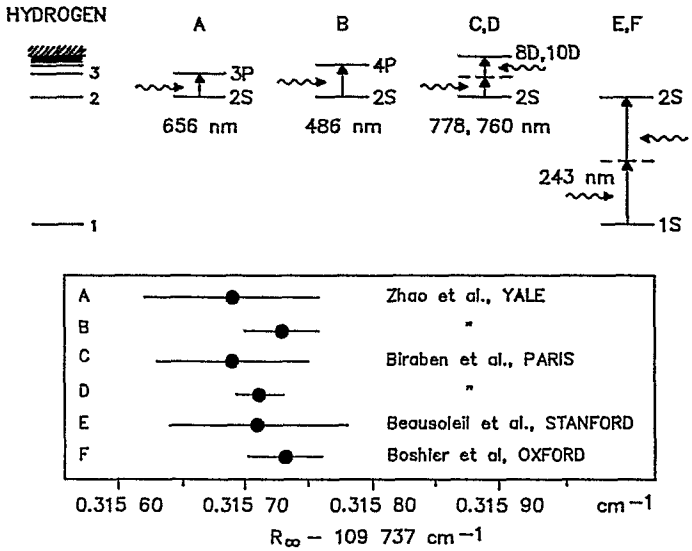

Fig. 2. Recent measurements of the Rydberg constant by laser spectroscopy of hydrogen and deuterium. Data points A and B are derived from the wavelength of the Balmer-α [7] or Balmer-ß line [8], observed by quenching of a beam of metastable 2S atoms with crossed dye laser beams. Data C and D are obtained by Doppler-free two-photon spectroscopy of 2S-8..12S transitions, recorded by quenching of a metastable atomic beam [9,10]. The Rydberg values E and F have been measured by Doppler-free two-photon spectroscopy of the 1S-2S transition in a gas cell [11,12].

3. Two-Photon Spectroscopy of the 1S-2S Transition

Even the earliest saturation spectra of the Balmer-α line approached the resolution limit imposed by the short lifetime of the excited levels. We have therefore long been concentrating our experimental efforts on the 1S-2S transition with its much narrower width [11,14-17]. The 1/7 second lifetime of the 2S state implies a natural line width of only about 1 Hz, or an ultimate resolution better than 10^{-15}.

Although one-photon transitions from 1S to 2S are forbidden, the 2S level can be excited by simultaneous absorption of two photons of 243 nm wavelength. If these photons come from opposite directions, first-order Doppler-broadening cancels without any need to select slow atoms, as first recognized by V.P. Chebotaev and coworkers [18]. The main difficulty in two-photon spectroscopy of hydrogen 1S-2S has long been the lack of a good tunable laser near 243 nm. Until recently, such experiments had to rely on frequency doubled pulsed dye laser systems with rather large instrumental linewidth and cumbersome frequency chirping problems [14-17].

In all but one [17] of the 1S-2S experiments to date, the atoms have been observed in a gas cell. Molecular hydrogen is dissociated in a gas discharge, and the atoms travel by gas flow and diffusion into the observation cell. The signal is observed by monitoring collision induced vacuum ultraviolet Lyman-α photons.

In 1S-2S spectra of hydrogen and deuterium, recorded by C. WIEMAN [16] in this way, the observed linewidth remained as large as 100 MHz. Nonetheless, the isotope shift could be measured well enough to yield first experimental evidence for a relativistic correction to the nuclear recoil effect. This correction was known theoretically, but was considered too small to be observable.

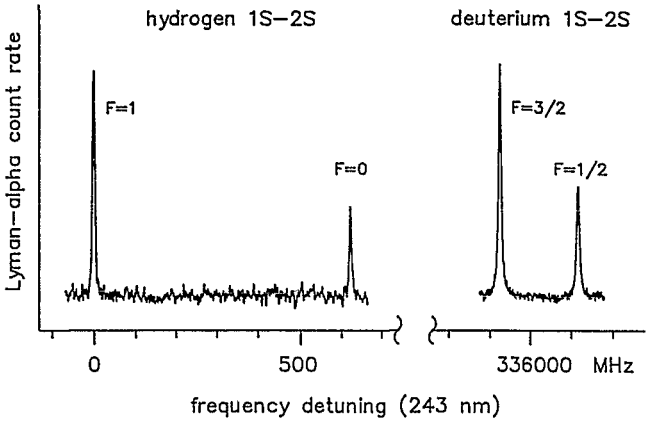

Fig. 3. Continuous wave Doppler-free two-photon spectra of hydrogen and deuterium 1S-2S [11].

Despite such successes, it was obvious that only a continuous wave laser source can do justice to the extremely sharp 1S-2S transition. Production of intense cw radiation near 243 nm remained long an elusive goal. Satisfactory power levels of several mW were first achieved by B. COUILLAUD et al. [19] by summing the frequency of a 351 nm argon laser and a 790 nm dye laser in a crystal of KDP. In the first cw experiment with this source [11], the power in the observation cell was further enhanced with a standing wave build-up cavity. Fig. 3 shows two-photon spectra recorded in this way. Although the resolution is much superior to the earlier pulsed spectra, it remains limited to a few MHz by laser frequency jitter, collision effects, and transit-time broadening. A further at least millionfold improvement in resolution should ultimately be achievable.

In 1986, R. BEAUSOLEIL and D. McINTYRE completed their thesis research at Stanford with an absolute frequency measurement of the F = 1 component of hydrogen 1S-2S [11,20]. As frequency reference they employed a 486 nm cw dye laser, locked to a narrow absorption line of $^{130}Te_2$ vapor. This line was chosen near a reference line, calibrated to within 4 parts in 10^{10} by A. FERGUSON et al. [21]. Its second harmonic coincides very nearly with the resonance frequency of the hydrogen two-photon transition, so that the frequencies can be precisely compared by observing a radio frequency beat signal.

With a careful study of pressure shifts in pure hydrogen and in a dilute mixture of hydrogen in helium, the absolute wavelength was determined to within 7 parts in 10^{10}. The corresponding value of the Rydberg constant (data point E in Fig. 2) is in good agreement with the other recent Rydberg measurements.

4. The Ground State Lamb Shift

If the Rydberg constant is considered known, a measurement of the 1S-2S frequency permits a different interpretation: it can yield an experimental value for the Lamb shift of the 1S ground state, or, more precisely, for the difference of the Lamb shifts of the 1S and 2S levels. Measurements of the 2S Lamb shift have been carried to very high precision [22] in order to test quantum

electrodynamic theory. The Lamb shift of the 1S ground state is eight times larger, but cannot be measured by radio-frequency techniques, since there is no nearby P state.

Wieman was able to determine the 1S Lamb shift to within a few parts per thousand by comparing the 1S-2S frequency with the n = 2-4 Balmer-ß transition, recorded simultaneously with the blue fundamental dye laser radiation [16]. By comparing 1S-2S, in effect, with the Balmer-ß measurement at Yale, the new Stanford experiment [11] gave a 1S Lamb shift of 8173.3(1.7) MHz, in good agreement with the theoretical value of 8172.94(9) MHz. With an uncertainty of 2 parts in 10^4, it is about an order of magnitude away from challenging the best current calculations. Unlike measurements of the 2S Lamb shift, comparisons of optical transition frequencies in hydrogen need not be limited by poor resolution due to a short-living P state, and they may soon provide the most stringent tests of quantum electrodynamics for a bound system.

5. Towards Higher Resolution

At Garching, R. Kallenbach and C. Zimmermann have begun work on a new experiment, aimed at an initial thousandfold improvement of the resolution of the 1S-2S transition. Together with J. Sandberg, they have developed a new highly monochromatic ultraviolet light source, based on second harmonic generation in a crystal of barium beta borate [23]. Light from a 486 nm ring dye laser is injected into a doubly resonant build-up cavity surrounding the crystal. This cavity enhances the intensities of fundamental and second harmonic radiation, and it provides a clean TEM_{00} output mode despite a considerable walk-off in the angle tuned crystal. Several mW have in this way been generated with an input power of 300 mW.

The rms linewidth of the dye laser has so far been reduced to 300 Hz relative to a reference cavity with the help of an intracavity ADP phase modulator and a fast servo system which compensates for small rapid optical path fluctuations in the liquid dye jet [24]. A perhaps even more elegant alternative is the external laser frequency stabilizer [25] which compensates for phase and frequency noise after the light has left the laser cavity. J. Hall and coworkers [26] have recently reduced the linewidth of a commercial ring dye laser to sub-Hz levels with such a device.

Even with such a highly monochromatic laser, the natural line width of the 1S-2S transition can only be approached if the atom can interact with the light field for a sufficiently long time and without being perturbed by collisions. As a first step towards this end we have constructed a hydrogen atomic beam apparatus. The atoms are cooled to a temperature of about 6 K with the help of an escape nozzle mounted on a liquid helium cryostat [27], as illustrated in Fig. 4. A coaxial ultraviolet standing wave field is maintained inside a build-up cavity surrounding the beam source. Excitation over an interaction length of several cm reduces transit broadening to a few kHz. Atom cooling also reduces relativistic second order Doppler-shifts from 70 kHz at room temperature to about 1 kHz, and it increases the excitation probability per atom in proportion to the square of the mean interaction time.

We are exploring other means of slowing the atoms for future experiments with even higher resolution. One promising approach may be laser radiation pressure cooling [28]. Unfortunately, such cooling requires vacuum ultraviolet Lyman-α radiation near 121.5 nm. On the other hand, a hydrogen atom can be slowed from 6 K to a few mK by resonant scattering of only about 100 photons. A train of suitable pulsed Lyman-α radiation could, for instance, be produced by injecting 364.5 nm light from

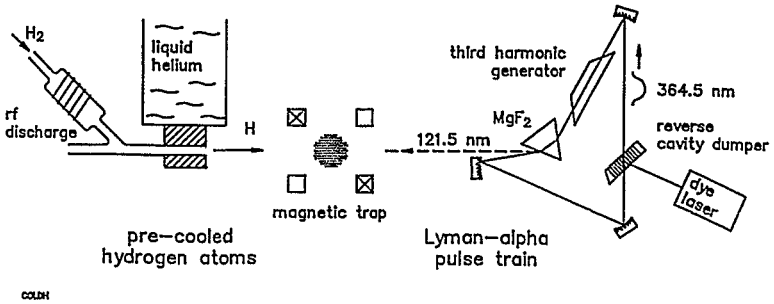

Fig. 4. Hydrogen atomic beam apparatus and scheme for laser cooling and magnetic trapping of hydrogen atoms.

a dye laser into an optical ring resonator, so that it passes repeatedly through a gas cell which produces some third harmonic radiation, as illustrated in Fig. 4.

A magnetic trap can be employed to suspend the cold atoms so that they do not escape or fall down due to gravity. Hydrogen atoms cooled with a dilution refrigerator have recently been trapped magnetically by N. MASUHARA et al. [29] and by R. VAN ROIJEN et al. [30], as also reported elsewhere in this volume. Even though trapped hydrogen atoms are perturbed by the magnetic field, high resolution two-photon spectroscopy of 1S-2S should still be possible, since upper and lower levels have very nearly equal magnetic moments so that first- and second-order Zeeman shifts remain small.

For ultimate precision one could let the cold atoms escape from the trap so that they can be observed in free fall due to gravity. Two-photon optical Ramsey spectroscopy of an "atomic fountain" has been analyzed in a semiclassical model [31]. On their parabolic trajectories the atoms are passing twice through the same standing wave laser field, so that the total excitation probability shows interference effects between the two excitation amplitudes. In contrast to ordinary Ramsey spectroscopy, the fastest atoms in a fountain spend the longest time between the two interactions and therefore produce the narrowest interference fringes. This difference has an interesting consequence when averaging over a broad atomic velocity distribution: The central interference signal appears very nearly as a simple Lorentzian resonance with just the natural linewidth. Such an experiment can hence reach the same resolution as if the atoms were standing still in the laser field. A linewidth of 1 Hz requires a fountain of just a few cm height.

6. Optical Frequency Metrology

Long before such ultimate resolution is reached, new approaches for measuring and comparing optical frequencies are urgently needed.

One interesting reference standard may be the methane stabilized helium neon laser at 3.39 μm. Its infrared frequency can be compared directly with the microwave cesium frequency standard with the help of a relatively short frequency chain [32]. An accuracy of 1 part in 10^{12} or better appears feasible for a transportable secondary standard.

At Garching, we are exploring several different approaches for synthesizing the seventh harmonic of a 3.39 μm helium neon laser, which coincides within about 2 THz with the 0.486 μm fundamental

wavelength for spectroscopy of hydrogen 1S-2S. The most straightforward scheme is a laser frequency chain employing tunable solid state lasers and nonlinear optical crystals for frequency mixing.

An alternative, proposed by R. Kallenbach, employs two tunable dye lasers, operating at the fourth and sixth harmonic of the 3.39 μm helium neon laser frequency f. The frequency ratio of 3:2 is assured by comparing the second harmonic of the laser at 6f with the third harmonic of the laser at 4f. The frequency difference is simultaneously maintained at 2f by monitoring a beat signal between the sum frequency 4f+f and the difference frequency 6f-f. Summing the frequencies 6f and f finally produces 0.485 μm radiation at 7f.

A conceptually even simpler approach uses just one optical parametric oscillator, pumped by a dye laser or diode laser at 4f and oscillating at the two frequencies f and 3f. The signal frequency f is enforced by injection locking with light from the 3.39 μm reference laser. The pump frequency is adjusted so that the idler frequency agrees with the third harmonic of the reference laser. The seventh harmonic is then generated by simply summing idler and pump frequency.

A much more general and appealing scheme would be a true optical frequency divider, as could be realized by synchronizing the cyclotron motion of an electron with a laser field [33,34].

Fig. 5 illustrates a less elegant but perhaps more feasible scheme for an optical difference frequency divider and frequency synthesizer [35]. Each stage can be compared to a differential gear box: It produces a new frequency f_3 exactly halfway in between two input frequencies f_1 and f_2. To this end, it contains a tunable laser, whose second harmonic is phase locked to the sum frequency f_1+f_2. By connecting several such stages in cascade, the original frequency difference is successively halved until it becomes accessible to a microwave frequency counter. If a laser frequency f and its second harmonic are used as the starting frequencies, a beat signal at $f/2^n$ can be observed after n stages. Only 16 stages would be sufficient to link 486 nm radiation to the 9 GHz cesium frequency standard.

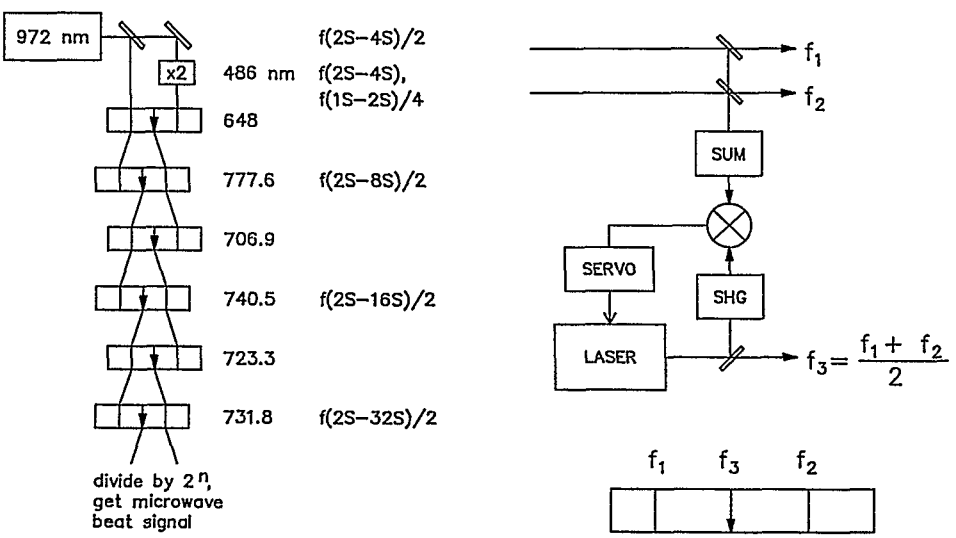

Fig. 5. Optical difference frequency divider and synthesizer of hydrogen transition frequencies [35].

The exact path can be chosen for convenience or so as to synthesize a desired optical frequency. Very few stages are sufficient to synthesize a number of hydrogen transition frequencies as illustrated in Fig. 5. Such a scheme represents then an "artificial" hydrogen atom which can be used to compare different hydrogen transition frequencies with extreme precision, and without the need to make absolute frequency measurements.

7. Comparing Experiment and Theory

Such comparisons promise interesting tests of QED. Unfortunately, however, the theory of hydrogen is no longer simple, once we try to predict its energy levels with adequate precision [36]. The quantum electrodynamic corrections to the Dirac energy of the 1S state, for instance, have an uncertainty of about 35 kHz, caused by numerical approximations in the calculation of the one-photon self-energy of a bound electron, and 50 kHz due to uncalculated higher order QED corrections.

The accuracy of computed energy levels is further limited by uncertainties of fundamental constants and by nuclear size and structure effects. The dominant contributor to the uncertainty of the 1S-2S frequency ($\simeq$500 kHz) is still the Rydberg constant. However, the electron/proton mass ratio, known within $2 \cdot 10^{-8}$ [37], introduces an additional uncertainty of about 30 kHz. The rms charge radius of the proton contributes at least another 50 kHz [38], assuming that it has been measured to within 2.5%; estimates of this accuracy still differ widely.

A measurement of the 1S-2S frequency alone can therefore give only a modest further improvement of the Rydberg constant before its interpretation is limited by other uncertainties. At one time it has been suggested to determine the proton-electron mass ratio from the 1S-2S isotope shift. However, after the most recent measurements by VAN DYCK et al. [37], the uncertainty of the isotope shift is now dominated by nuclear size effects.

Fortunately, some of these troublesome uncertainties scale in a known way with the principle quantum number n, and they can be eliminated by combining measurements of different transitions. To give an example, we could measure the isotope shifts Δf of two separate transitions such as 1S-2S and 2S-nS. By forming the linear combination $7\Delta f(2S\text{-}nS) - (1 - 8/n^3) \cdot \Delta f(1S\text{-}2S)$, we obtain a new composite frequency which no longer contains terms proportional to $1/n^3$. It is thus independent of nuclear size and can be calculated much more precisely than its constituents. A comparison of experiment and theory can then yield an improved electron-proton mass ratio.

Following the same strategy, we can find a composite optical frequency, $7f(2S\text{-}nS) - (1 - 8/n^3) \cdot f(1S\text{-}2S)$, which does not depend on the nuclear charge radius or on higher order QED corrections scaling with $1/n^3$. With improvements in the computation of the self energy and with an improved measurement of the electron mass, this frequency can provide a very precise Rydberg value. If Coulomb's law is valid within atomic dimensions, exactly the same Rydberg value must be obtained from any 2S-nS transition as well as from microwave spectroscopy of high Rydberg states, provided perturbing external fields can be sufficiently controlled.

A dimensionless frequency ratio, such as $f(1S\text{-}2S)/f(2S\text{-}nS)$, on the other hand, is independent of the Rydberg constant. Its measurement can serve as a sensitive test of quantum electrodynamic level shifts and as a means to determine the size of the proton or deuteron, provided QED is correct.

8. Positronium, Muonium, and Anti-Hydrogen

Additional tests of fundamental theory become possible if transitions in hydrogen are compared with those in other hydrogen-like atoms. The exotic positronium (e^+e^-) and muonium (μ^+e^-) are particularly interesting since they consist of leptons so that no cumbersome hadron structure corrections are necessary. Unfortunately, both atoms are unstable so that they cannot be studied with the same high resolution as hydrogen.

Positronium still presents rather formidable theoretical challenges [39], since it is a relativistic two-body system which cannot be approximated by the motion of a particle of reduced mass in a fixed Coulomb potential. In addition, QED calculations must include annihilation terms. First laser measurements of the 1S-2S two-photon transition frequency in positronium [40] give a result about five standard deviation lower than the theoretical predictions, after taking a recalibration of the tellurium reference line into account [41]. On the other hand, as yet uncalculated higher order terms could well account for this discrepancy.

Recently, first experimental results have also been reported for the 1S-2S two-photon transition of muonium [42]. With the relatively heavy muon mass this atom is much more similar to hydrogen so that its energy levels can be computed more readily, and the 2.2 μsec lifetime of the muon should permit a resolution of better than 100 kHz.

Another most interesting atom that should permit spectroscopy with extreme resolution is anti-hydrogen (p^-e^+). After the recent successful trapping of low energy anti-protons by G. Gabrielse and coworkers [43] it appears conceivable to produce and magnetically trap slow anti-hydrogen atoms. High resolution spectroscopy of anti-hydogen 1S-2S two-photon transitions should require just a few atoms inside the trap. By observing simultaneously hydrogen and anti-hydrogen, one can investigate whether there are any spectroscopic differences between matter and anti-matter. At very low temperature and with a vertical trap of sufficient length it might even be possible to determine whether anti-hydrogen senses a stronger gravitational force than hydrogen, as recently suspected [44].

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"Ultra-spectroscopy" of atomic hydrogen as envisioned here can provide a strong motivation to develop and refine our spectroscopic instruments and techniques far beyond the present state of the art. If past history is any guide such efforts are likely to be rewarded by new applications and unexpected discoveries.

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