

Two-Photon Spectroscopy of Hydrogen 1S-2S

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1. Introduction and Overview

1.1 The interest in 1S - 2S; early experiments

In 1976, reviewing the field of two-photon spectroscopy, BLOEMBERGEN and LEVENSON wrote: " The 1S - 2S transition in atomic hydrogen is among the most important in physics, and it can be resolved by the Doppler-free two-photon technique. The experiment is a tremendous challenge....." [1]. More than a decade later, this remains a fair summary. The special nature of the 1S - 2S transition was pointed out independently by several authors, notably BAKLANOV and CHEBOTAYEV [2,3], who carried out a detailed study, and CAGNAC et al [4]. By a remarkable combination of properties it brings the ground state of hydrogen into the class of atomic levels which can be studied by the methods of high resolution laser spectroscopy. In recent years, experiments on the 1S - 2S transition in hydrogen and its isotopes have been used to test quantum electrodynamics, to determine the Rydberg constant and to give a value of the electron-proton mass ratio. It is true that so far all these objectives have been accomplished at least as well by other means; 1S - 2S experiments bristle with technical difficulties, and the first continuous wave (cw) measurements were reported only recently [5,6]. Now that this milestone has been passed, however, the potential for improvement is enormous.

To see the experiments in context, one must appreciate that high resolution spectroscopy of atomic hydrogen has largely been confined to transitions between excited states because of the experimental problems associated with studying the ground level. There is a 10 eV energy gap between the ground level and the nearest excited levels (in the $n = 2$ manifold); this corresponds to a wavelength of 121.5 nm, in the vacuum ultraviolet. To carry out a Doppler-free 1S - 2P experiment one would have to generate tunable narrow-band radiation at this wavelength; even then, the radiative lifetime of the upper level would give rise to a homogeneous linewidth of 100 MHz and one would still face the problem of finding a suitable frequency standard at 121.5 nm. The way in which measurements on the 1S - 2S transition can overcome these difficulties was shown in a remarkable series of experiments by HÄNSCH and his

coworkers [7,8,9] to measure the ground state Lamb shift. These were carried out during the period 1974–80 at Stanford. The use of two-photon spectroscopy takes the wavelength of the radiation required to 243 nm, into the range in which it is possible to generate tunable radiation from laser light by standard non-linear methods. As regards the line-width, the two-photon technique allows one to eliminate first-order Doppler broadening; but in addition, because the 2S level is metastable, the transition has a natural width of only 1.3 Hz, so that the resolution limit is of order 1 part in 10^{15} ! Of course, this limit has not yet been remotely approached; in these early Stanford experiments, the use of pulsed excitation led to linewidths (at 243 nm) of order 200 MHz. Finally, the problem of calibration was solved by using another transition in hydrogen, $H\beta$ ($n = 2 - n = 4$), as a reference. The 243 nm radiation required for the 1S – 2S excitation was generated by frequency doubling, so that the fundamental wavelength was at 486 nm, which coincides with $H\beta$. This transition was excited (using the fundamental laser radiation) in a separate hydrogen discharge, and the small frequency differences between one quarter of the 1S – 2S transition and components of $H\beta$ were measured. If Bohr theory were exact, these differences would be zero, so the uninteresting gross structure of hydrogen was eliminated from the measurements. This ingenious approach allowed the Lamb shift to be determined without the need for an accurate value of the Rydberg constant. Experiments were carried out in hydrogen and deuterium, eventually giving results with quoted uncertainties of about 30 MHz [9].

1.2 The drive towards higher precision

The precision achieved in the early experiments was limited primarily by the use of pulsed techniques. The fact that two non-linear processes – generation of 243 nm radiation and two-photon excitation – are involved makes it much more difficult to carry out a cw experiment; nevertheless, one must do so if one is to begin to realise the potential of the 1S – 2S transition. An uncertainty of tens of megahertz is much too large if the ground state Lamb shift is to provide a significant test of QED. The radio-frequency measurements of the $2^2S_{1/2} - 2^2P_{1/2}$ interval of about 1 GHz as first performed by LAMB and RETHERFORD [10] have now reached a quoted accuracy of the order of a few kilohertz [11,12,13]. The ground state Lamb shift is about 8 GHz, so one is aiming for considerably better than 100 kHz on a ground state result to provide a comparable test. It would be useful to carry out such measurements since there is little prospect of dramatic improvement in the 2S – 2P measurements; the uncertainty is already less than 10^{-4} of the 2P line-width.

However, there are other reasons for improving the precision of 1S – 2S measurements. For example, a measurement of the absolute frequency (in contrast to one in terms of $H\beta$) is sensitive to the value of the Rydberg constant. More generally, one can expect to get the most out of measurements of transition frequencies in

hydrogen-like systems by combining them both with each other and with data from other types of experiment. It is therefore likely that the study of the $1S - 2S$ transition will play an increasingly important role in understanding fundamental systems and the determination of fundamental constants.

For several years, considerable effort was put into attempts to develop a successful cw experiment [14,15]. Interest also developed in the coherent multiple pulse approach suggested by BAKLANOV and CHEBOTAYEV [16]. In the meantime, the objective of making absolute measurements of the transition frequency was also pursued, and in 1985 BARR et al [17] reported measurements of the frequencies of transitions in molecular tellurium in the vicinity of 486 nm which could act as references for the two-photon work. This was shortly followed by two papers from the Stanford and Southampton groups [18,19] in which the new tellurium standards were used in pulsed experiments to measure the absolute $1S - 2S$ frequency for the first time. However, even before these appeared, the first observation of a cw $1S - 2S$ signal was made at Stanford by FOOT et al [20]. This became the basis of a measurement in terms of the tellurium standard [5], and the cw experiment at Oxford gave its first results shortly afterwards [6]. The groups adopted different methods to generate the 243 nm radiation; Stanford chose sum frequency mixing, while at Oxford frequency doubling was used, a newly developed crystal (β -barium borate, or BBO) playing a crucial role. The advantage of the frequency doubling approach is that it opens the way to a very precise cw measurement of the $1S$ Lamb shift via a comparison with $H\beta$ as in the earlier pulsed experiments. Although this comparison has always been the main objective of the Oxford group, the work so far reported has been carried out using tellurium calibration. This limited the precision because the tellurium standards are simply not good enough to match the accuracy of a cw experiment. Nevertheless, the work at Oxford has given the most precise values of the Lamb shifts of the ground levels of hydrogen and deuterium, with uncertainties of less than 1 MHz. Alternatively, if one accepts that the calculation of the Lamb shift is reliable at this level of accuracy, the experiment can be interpreted as a measurement of the Rydberg constant, as accurate as the best of recent determinations based on other transitions [21].

1.3 The present position

No further measurements have been reported, though the research is being actively pursued at Munich, Novosibirsk, Oxford, Southampton and Yale. The frequency doubling scheme is basically simpler and more straightforward than other ways of producing radiation at 243 nm, and has the advantage that the fundamental is close to $H\beta$; it will probably become the standard method of exciting the transition. The results obtained so far are soon likely to be superseded as techniques are refined and frequency standards improve, so that this chapter is being written during an exciting period of rapid development. What the outcome will be in terms of physics it is too

early to tell. As a test of QED, hydrogen has the disadvantages of uncertainty in the nuclear size correction (some 50 kHz in the 1S Lamb shift) and a low value of the expansion parameter $Z\alpha$. This is underlined by recent work in He^+ [22] and highly ionized atoms [23]. Of course, high resolution spectroscopists dream of eventually realising experimentally the 1.3 Hz linewidth of the transition; however, major investments of time and effort would be needed to achieve this, and we are concerned here with the first steps along the way, and with more immediate objectives. In section 2 the experiments carried out to date are discussed, particularly that carried out at Oxford since it is the most recent and most accurate (details are from BOSHIER [24] and BOSHIER et al [25]). Section 3 contains concluding remarks.

2. Review of 1S - 2S Experiments

2.1 Basic Structure

A 1S - 2S experiment can be roughly divided up as follows:

- Production of ground state hydrogen atoms, and their transport to an interaction region.
- Generation of 243 nm radiation, beam handling, enhancement/amplification.
- Excitation and detection system.
- Calibration.

The way in which 243 nm radiation was produced - specifically, the distinction between pulsed and cw experiments - represents the most obvious subdivision of work reported so far, and we shall discuss these categories separately. However, future cw experiments are likely to be primarily distinguished by the methods adopted to detect and calibrate the two-photon signals. We consider calibration separately at the outset.

2.2 Calibration

As has already been pointed out, two different methods of calibration have been used in the work so far reported. One can measure in terms of $H\beta$, and in this way determine the ground state Lamb shift. If instead one uses a frequency standard, one can determine the 1S Lamb shift by allowing for the other contributions to the measured 1S - 2S interval; this requires a value of the Rydberg constant from another source. Alternatively, if one assumes the 1S Lamb shift known from theory at the level of accuracy of the experiment, one can regard the measurement as a determination of the Rydberg constant. All the most recent work has been in terms of the external standards provided by the tellurium transitions, but these introduce an uncertainty which in the case of the cw experiments dominates the final error. This situation must be

avoided in future work, probably by a change in calibration technique. A frequency doubled cw experiment offers the possibility of returning to a comparison with $H\beta$ or making measurements in terms of an improved external standard at 486 nm, and work towards both these objectives is in progress.

2.3 Pulsed experiments

These cannot compete with cw experiments in terms of the accuracy of measurement, so we shall discuss them very briefly, and primarily insofar as they have a bearing on cw work. With pulsed excitation, there is no difficulty in generating enough 243 nm radiation to observe strong signals with easily realizable number densities of atomic hydrogen. The problem is the uncertainty introduced by the distorting effects of pulsed dye amplifiers which affect the shape and position of the observed 1S - 2S resonance. These effects have been responsible for the main contribution to the error in all the pulsed experiments except the first (the dominant uncertainty in this case was due to the Doppler broadening of $H\beta$; in subsequent work, the Doppler-free techniques of saturation or polarization spectroscopy were used).

The detection scheme for the earlier pulsed experiments in which the atoms were excited in a cell [7,8,9] made use of collisional transfer of excitation between the 2S and 2P levels. The intensity of Lyman- α radiation (at 121.5 nm) resulting from the subsequent decay to the ground level was measured as a function of laser frequency. This is a convenient method, but not suitable for the highest precision; by its nature it prevents the experiment from being carried out in a collision-free environment. Also, its efficiency is hard to estimate; signal levels in these experiments were significantly lower than expected, and it is possible that radiative trapping of Lyman- α allowing other relaxation mechanisms to depopulate the excited levels may have been responsible. The simplicity of the method has led to its being used also for both cw experiments carried out so far, but there are obvious advantages in using excitation from a ground state beam with subsequent detection of metastables. In one of the two most recent pulsed experiments [18] the excitation occurred in a beam, and in both [18,19] a new detection system was adopted. The Doppler-free two-photon excitation to the 2S level was followed by ionization by a further 243 nm photon, and the ions could be detected with high efficiency.

2.4 CW Experiments

2.4.1 Feasibility

In contrast to the pulsed experiments, the main problem for many years in cw experiments was to observe any signal at all. However, this is not apparent from simple feasibility estimates, as we now show. We assume for simplicity that one excites the atoms using two counter-propagating plane-polarized gaussian beams each with the

same power P , and take the bandwidth of the light to be small compared with the width Γ of the resonance. Let the waist size of the beam be w_0 , and suppose that we detect atoms excited over a length L of the beam much less than the Rayleigh length. Then if the atomic number density is N , the peak excitation rate in the detection volume is

$$R \approx \frac{1}{\pi \epsilon_0^2 c^2 \hbar^4} \frac{L}{w_0^2} \frac{4}{\Gamma} N M^2 P^2$$

where

$$M^2 = \left| \sum_j \frac{\langle i | e z | j \rangle \langle j | e z | f \rangle}{\omega_{ji} - \omega} \right|^2,$$

$|i\rangle$ and $|f\rangle$ respectively representing the 1S and 2S states. Evaluating this expression, one finds

$$R \approx 2.7 \times 10^{-7} (L/w_0^2 \Gamma) N P^2 W^{-2} \text{ m}^4 \text{ s}^{-2}.$$

Practical considerations limit the choice of w_0 , L and N . We take the values appropriate to the Oxford experiment [6], $w_0 = 100 \mu\text{m}$, $L = 10 \text{ mm}$, with a typical pressure in the interaction region of around 100 mtorr. Then with $\Gamma/2\pi \sim 10 \text{ MHz}$ we have a signal count rate of

$$S = \eta R \sim 1.3 \times 10^{10} P^2 W^{-2} \text{ s}^{-1}.$$

for fully dissociated hydrogen, assuming that every excitation is followed by a detectable Lyman- α photon. The efficiency η of the detection system (solid angle, filter) has been taken to be 10^{-3} . From the evidence of the pulsed work, we can expect our estimate of S to be too large (and with hindsight, from the results of both cw experiments, it appears that the factor under some conditions can be as much as 1000). Even so, we can hope to generate 1 mW or so of 243 nm radiation and use an enhancement cavity to give a value of $P \sim 10 \text{ mW}$ and a signal $S \sim 1000 \text{ counts s}^{-1}$.

The real difficulty lay elsewhere. Until quite recently, the problem with the frequency doubling approach was the lack of suitable crystals. Lithium formate, the material used in the early pulsed work, is inefficient and subject to uv damage. Urea appeared on the scene around 1979, and was reported to have much superior properties [26]; however, it proved enormously difficult to obtain a crystal of good enough optical quality. The material is fragile, hard to grow and to polish. It also suffers surface damage when generating uv. Nevertheless, the first observation of a cw two-photon signal by frequency doubling was made with urea, following significant advances in polishing techniques at the Clarendon. However, urea was soon abandoned in favour of

the newly-developed material BBO [27,28], which has about the same efficiency as urea and is now available in crystals of excellent quality with no degrading of performance in use. We now describe the Oxford experiments, which were carried out with a single crystal of BBO, since they are the most accurate yet reported.

2.4.2 The Oxford Experiment

Flowing molecular hydrogen or deuterium was dissociated in a radio-frequency discharge operating at about 30 MHz. For some of the experiments, a 5% H, 5% D, 90% He mixture was used to reduce pressure shifts as in [5]; measurements were made in the range 20 to 300 mtorr and extrapolated to zero pressure. Efficient transport of dissociated hydrogen atoms from a discharge to an interaction region is important in a cw experiment, but despite much experimentation it remains a somewhat empirical science and recombination may be responsible for some of the apparent loss in signal discussed earlier. A short length of Teflon tubing was used, looped to prevent discharge light from reaching the interaction region.

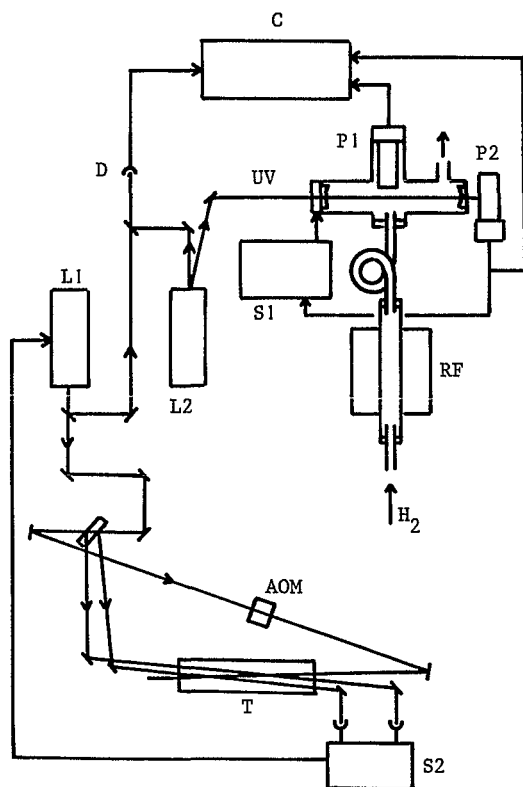


Figure 1 Apparatus of Oxford experiment [6]. L1, L2: tunable dye lasers. UV: ultra violet radiation (243 nm). RF: radio-frequency dissociation of flowing molecular hydrogen. P1: signal photomultiplier (Lyman- α detector). P2: photomultiplier for cavity locking and signal normalisation. S1: cavity length servo-control. C: computer. AOM: acousto-optic modulator. T: heated quartz cell containing tellurium. S2: laser frequency servo-control. D: fast photodiode

The BBO crystal was mounted intra-cavity in a krypton-pumped Coumarin 102 ring dye laser, and produced typically 2 mW at 243 nm. The crystal was phase-matched by angle tuning. The large walk-off angle of about 5° gave a beam with a very elongated intensity profile (aspect ratio 19:1); however, an optical system was devised which transferred this non-gaussian beam into the TEM₀₀ mode of an enhancement cavity in the hydrogen cell well enough to give intracavity powers P greater than 20 mW. Signals were up to 25,000 counts s⁻¹, depending on conditions, with background (largely scattered 243 nm light) typically 1000 s⁻¹. Signal/noise was thus never a problem in the measurements. Profiles were recorded of the stronger hyperfine component of the transition for both hydrogen and deuterium; a typical recording is shown in figure 2.

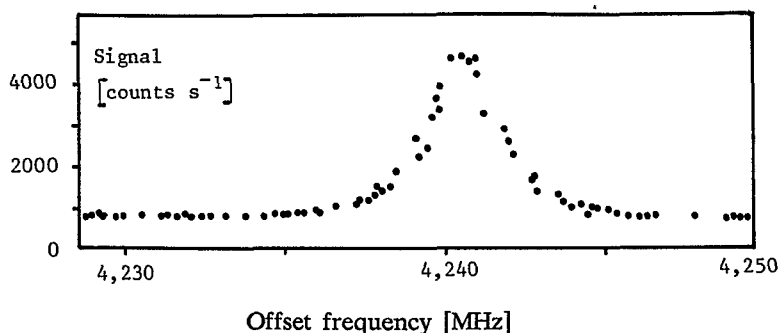


Figure 2 The stronger component of the 1S-2S two photon transition in deuterium. The signal is the normalised Lyman- α fluorescence observed as a function of the frequency difference between lasers L1 and L2 (fig. 1) when L1 is locked to the appropriate transition (b_2) in $^{130}\text{Te}_2$. The measured offset frequency is 20 MHz greater than the true value because of the shift introduced by the acousto-optic modulator. The pressure in the deuterium cell was 270 mtorr

It was found that the normal method of scanning the frequency doubling laser by rotating the tipping Brewster plate in the reference cavity was insufficiently smooth over the small ranges required; a satisfactory alternative was found to be to shift electronically the reference point used for locking the reference cavity. Further refinements to the lasers were unnecessary, because their performance did not limit the accuracy of the measurements. The calibration procedure was responsible for most of the uncertainty, and was the least satisfactory aspect of the experiment as we now discuss.

After initial experiments with a tellurium cell which later turned out to be contaminated, we borrowed the actual cell which had been calibrated and reproduced the conditions under which it had been operated at the NPL. Even with this system, the three main contributions to the uncertainty in measuring the 1S - 2S frequencies in

hydrogen and deuterium were attributable to the method of calibration, as follows (all in kHz):

	H	D
Statistical uncertainty	152	140
Uncertainty in locking point on Te line	320	160
Uncertainty in Te frequencies	660	660

Other effects are negligible compared with these, but are discussed in [24] and [25]. The statistical uncertainty is mainly due to small temperature fluctuations of the tellurium cell; these had periods of order minutes, and gave corresponding shifts of the calibration frequencies. The tellurium lines are relatively wide (~ 20 MHz), but even so the uncertainty in the locking point could be reduced considerably by the use of a more sophisticated locking system. However, the main problem is the uncertainty in the frequencies of the transitions themselves. The final results for the 1S - 2S frequencies are:

$$f(1S - 2S) = 2,466,061,414.12(75) \text{ MHz (H);}$$

$$2,466,732,408.45(69) \text{ MHz (D).}$$

In deriving values of the Lamb shifts of the 1S levels from these results, one needs to subtract the Dirac energy. To calculate this requires in turn a value of the Rydberg constant. We use that of ZHAO et al [21], who measured on H β ; the reference frequency in their work was a He-Ne laser which has been directly compared with that employed in the NPL calibration of the tellurium lines. Some of the errors in the two measurements are therefore systematic; effectively, the 1S - 2S frequency is being compared with H β as in the early pulsed experiments, albeit via three steps. We obtain the following values for the 1S Lamb shifts, which we take to be the sum of all QED contributions plus the correction for finite nuclear size [29]:

$$\text{H: } \Delta\nu = 8,172.94(85) \text{ MHz (theory } 8,173.05(10) \text{ MHz),}$$

$$\text{D: } \Delta\nu = 8,184.10(80) \text{ MHz (theory } 8,184.09(12) \text{ MHz).}$$

The theoretical results quoted here and below make use of the most recent calculations of the reduced mass and recoil corrections [30,31,32] and values of the fundamental constants (see [24,25]). We stress once more that systematic and random uncertainties due to the calibration procedure completely dominate the quoted experimental errors; detailed examination of the data suggests that the total contribution from other sources is less than 50 kHz, despite the use of cell excitation and the need to extrapolate to

zero pressure. Even though the results so far do not provide a critical test of QED, therefore, there is the prospect of considerable improvement. However, if we accept the evidence provided by the $n = 2$ Lamb shift experiments that the theory is very unlikely to be incorrect at the level of our experimental error, we can interpret the $1S - 2S$ frequency measurements as determinations of the Rydberg constant; we obtain

$$R_{\infty} = 109,737.31573(3) \text{ cm}^{-1}.$$

This is as precise as the best previous measurement [21]. Finally, we can derive the isotope shift in the $1S - 2S$ transition; this is

$$\delta\nu (\text{D} - \text{H}) = 670,994.33(64) \text{ MHz (theory } 670,994.39(12) \text{ MHz)}.$$

Work is in progress at Oxford to improve this result by wide-band heterodyning techniques which avoid the need for frequency standards.

2.4.3 Comparison with Sum-frequency Mixing

In the Stanford experiment [5,20], 243 nm radiation was generated by mixing 351 nm light from an argon ion laser with a dye laser output at 790 nm in a crystal of KDP. This gave a little more uv output power than the Oxford frequency doubling system, but the method has the disadvantage that calibration of the two-photon signal is less straightforward. In the Oxford experiment, it was possible to measure the frequency difference between the fundamental from the frequency doubling laser and the tellurium calibration lines simply by optical heterodyning at 486 nm. The tellurium cell was placed in a saturation spectrometer, a second dye laser being locked to the appropriate line for hydrogen or deuterium. The method lends itself readily to a comparison with $H\beta$. At Stanford, calibration was carried out at 243 nm, so the light from the reference laser had to be frequency doubled. The difficulties raised by this procedure prevented any results being obtained for deuterium. However, the experiment did give the first cw measurement of the hydrogen ground state Lamb shift; the result was 8173.3(1.7) MHz.

Comparisons of the results of the work at Oxford, Stanford and elsewhere are shown in figs. 3 and 4.

3. Conclusion

The first point to make is that the results quoted above show that the present priority is to improve the comparison of the $1S - 2S$ frequency with a reference. This alone could bring the ground state Lamb shift into serious contention as a critical test of QED. Three ways suggest themselves, all of which are being pursued:

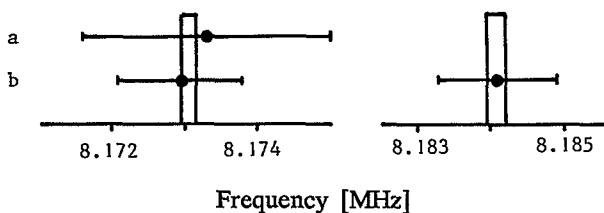


Figure 3 Comparison of measured 1S Lamb shifts with theory [24,25], the limits of which are shown as vertical lines. (a) Reference [5], (b) present work [6]. No earlier work of comparable precision exists for deuterium

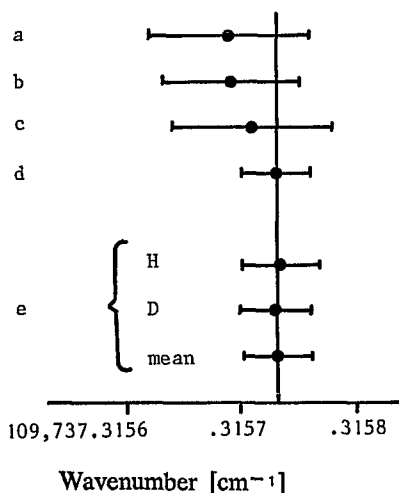


Figure 4 The present results for the Rydberg constant compared with other recent values. (a) Reference [36], (b) reference [35], (c) reference [5], (d) reference [21], (e) present work [6]. The vertical line is drawn through the mean of the new results

1. More refined work on tellurium lines. This is not a route towards much higher precision, but a more thorough study of lines currently limiting the accuracy of 1S - 2S experiments is clearly desirable.
2. A frequency chain providing an accurate standard in the vicinity of 486 nm without the need for interferometry.
3. Direct comparison of 1S - 2S with other hydrogen transitions, particularly H β as in the earlier pulsed work. This is the immediate goal of the Oxford group.

There are other obvious refinements. The Oxford experiments were carried out using commercial lasers, with frequency jitter around the 1 MHz level; there are well established techniques which can reduce this substantially. Cell excitation must ultimately be abandoned; although it is not at present a limiting factor, it becomes

increasingly unsatisfactory as techniques develop and precision improves. For a comparison with $H\beta$, it would be desirable to work with cold beams for both parts of the experiment. In Oxford, we are building a system in which the metastables created in the $1S - 2S$ excitation are used in a measurement of the sharp $2S - 4S$ transition. Such an approach should permit a very direct comparison of frequencies.

The main significance of the work so far carried out at Oxford is that it demonstrates the feasibility of making measurements of the $1S - 2S$ transition with a frequency doubled cw system. This opens the way to considerable improvements in precision, but the interpretation of the measurements will depend on what other information is available – the transition frequency is sensitive to QED effects, the proton size, and the values of fundamental constants. Cynics might point out that after well over a decade of work on the $1S - 2S$ transition, the results have produced no real surprises. A more optimistic view is that it is only over the last year or two that it has become possible to study the transition with the standard techniques of high resolution cw laser spectroscopy, and the next decade will bring dramatic developments. We shall see.

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