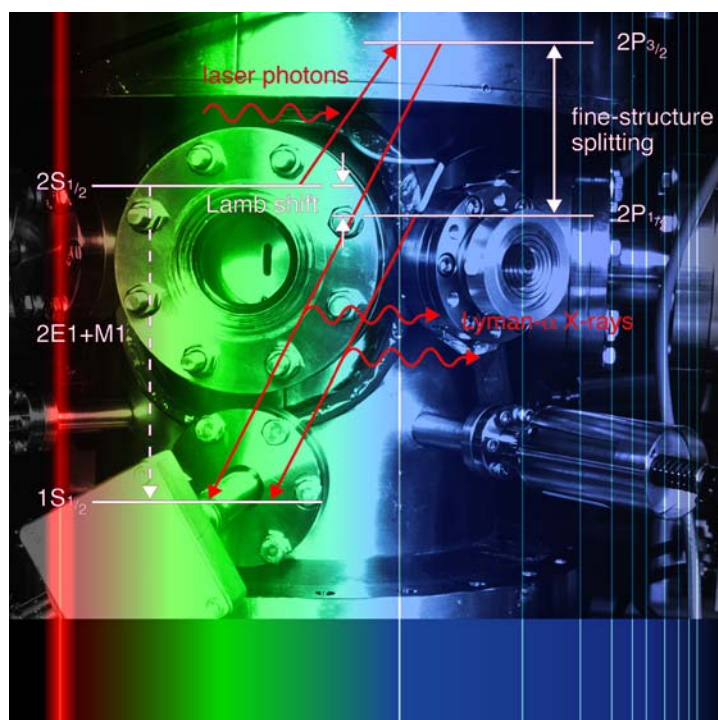


Hydrogenic Systems for Calculable Frequency Standards: Status and Options

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Abstract

The study of hydrogen and hydrogenic (one-electron) ions is an area of rapid progress and one of great potential for future frequency standards. In 1997, the two-photon 1S-2S transition in the hydrogen atom was included in the list of approved radiations for the practical realisation of the metre, and since then revolutions in optical frequency metrology have reduced the uncertainty in its frequency by more than an order of magnitude, to 1.8 parts in 10^{14} .

Hydrogenic systems are simple enough that the frequencies of their transitions can be calculated in terms of the Rydberg constant with an accuracy that can approach or exceed the measurement uncertainty. Transitions in such systems can be thought of as forming a natural frequency scale, and offer the prospect of a set of quantum frequency standards which are directly related to the fundamental constants.

The Rydberg constant is currently best determined from optical frequency measurements in hydrogen. However, to take full advantage of the recent high accuracy 1S-2S frequency measurement requires

- Improved measurements of other transition frequencies in the hydrogen atom;
- Reduced uncertainty in the quantum electrodynamics (QED) contributions to the energy levels, in particular the two-loop binding corrections;
- An improved value for the proton charge radius.

In He^+ and one-electron systems of higher atomic number Z , the two-loop binding corrections are a fractionally larger part of the Lamb shift due to their rapid scaling with Z . Measurements of the Lamb shift in medium- Z hydrogenic ions can therefore provide tests of these corrections, and feed in to the theoretical understanding of hydrogen itself. Although both theory and experiment are less accurate at higher Z , there is the potential for a new range of X-ray standards, providing that the QED corrections are well understood and new absolute measurement techniques can be developed.

A number of areas are suggested for future investigation:

- Improving the accuracy of hydrogen 2S- n S/D measurements, contributing to an improved Rydberg constant determination;
- Spectroscopy of cooled, trapped hydrogen atoms;
- Improved calculations of the two-loop binding corrections complete to all orders in $Z\alpha$.
- Lamb shift measurements in medium- Z hydrogenic ions by laser excitation, to check the two-loop binding corrections;
- Experiments to resolve discrepancies arising from the proton charge radius;
- Development of a small portable electron beam ion trap (EBIT) capable of producing high yields of hydrogenic ions;
- Development of techniques for absolute measurement of X-ray transitions observed in an EBIT.

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1. Introduction

The study of the hydrogen atom and hydrogenic (one-electron) ions is a fertile area of research and one of great importance for metrology [1]. In September 1997 the extremely narrow 1S-2S two-photon transition at 243 nm and other optical transitions in hydrogen were adopted by the *Comité International des Poids et Mesures (CIPM)* as new recommended radiations for the practical realisation of the metre [2]. As one-electron systems, hydrogen and other hydrogenic ions are of particular interest for frequency standards. The simplicity of their atomic structure means that their transition frequencies may be calculated (and related to one another) in terms of the Rydberg constant with an accuracy approaching or exceeding that with which they have been measured. Other transitions used as references for frequency standards from the rf to the X-ray region are in many-electron systems, where the complications introduced by the electron-electron interactions limit the accuracy with which the energy levels can be calculated.

In this report, we outline the status of theory and measurements for transitions in hydrogen and other one-electron systems which might have standards applications, and review the options for future work. We begin with a discussion of some theoretical aspects, concentrating on those which are currently limiting the comparison of experimental and theoretical results. The status and prospects for hydrogen itself are then considered, followed by sections on He^+ and hydrogenic ions of higher atomic number Z . Finally we review the options for future calculable frequency standards and make a number of recommendations for future work.

Whilst this report concentrates on one-electron systems, one should not overlook the work of Drake and others on helium-like (two-electron) systems (see e.g. [3-5]), which demonstrates that it is possible to make remarkably good calculations for such systems.

2. Theoretical aspects

An empirical formula for the wavelengths λ of transitions in hydrogen was suggested by Rydberg in 1889:

$$\frac{1}{\lambda} = R \left(\frac{1}{p^2} - \frac{1}{n^2} \right), \quad (1)$$

where p and n are integers and R is a constant now known as the Rydberg constant. Bohr's theory for the gross structure of the hydrogen atom led to a theoretical expression for the Rydberg constant:

$$R_{\infty} = \frac{m_e e^4}{8 \epsilon_0^2 h^3 c} = \frac{m_e c \alpha^2}{2h}, \quad (2)$$

where m_e and e are the mass and charge of the electron respectively, ϵ_0 is the permittivity of free space, h is Planck's constant, c is the speed of light in free space and α is the fine-structure

constant. The subscript ∞ denotes the Rydberg constant for a nucleus of infinite mass. The Rydberg constant as defined by equation 2 has units of inverse length and is usually quoted in reciprocal metres; however, the fixed speed of light means that the Rydberg constant can equivalently be expressed in frequency units with no loss of accuracy.

The Rydberg constant may be regarded as the natural scaling constant for $\Delta n > 0$ transitions in hydrogenic systems, where n is the principal quantum number. Transitions in hydrogen thus form a natural frequency scale ranging from the microwave to the UV [1]. For a higher- Z one-electron system the term R in equation 1 is replaced by $Z^2 R$, and the natural frequency scale extends into the VUV and X-ray region. At present the second is defined in terms of the 9.2 GHz hyperfine transition in the caesium atom. To measure an optical frequency it is necessary to multiply up from this frequency by about five orders of magnitude. In the past this could only be achieved using large and complex frequency multiplication chains, although the problem has now been greatly simplified by the development of new techniques using mode-locked femtosecond lasers [6,7]. A well understood hydrogen spectrum offers the possibility of a unit of time defined in terms of a hydrogen transition frequency which is calculable in terms of fundamental constants and a means of checking the methods used to link the optical and microwave regions of the spectrum.

The Rydberg constant is most accurately determined from spectroscopic measurements in hydrogen [8,9], rather than from the combination of fundamental constants of which it is composed (equation 2) [10]. To calculate transition frequencies in hydrogenic systems to an accuracy comparable with the experimental precision which has been achieved, it is necessary to include both relativistic and quantum electrodynamic (QED) effects. Using Dirac theory, the relativistically correct formulation of quantum mechanics, gives the main contributions to the energy levels. However, solutions to the Dirac equation do not give exact agreement with experiment; in particular they have no l dependence, states of the same n and j are degenerate. There are many corrections to the Dirac energies which need to be considered, such as one- and two-loop QED corrections, recoil corrections due to the finite nuclear mass and a correction for the non-zero nuclear volume. The evaluation of these corrections is an extremely challenging task and is described in more detail in, for example, the review of Pachucki [11] or that of Eides [12]. The difference between the actual binding energy of an atomic state and the Dirac-Coulomb energy is referred to as the Lamb shift (see Figure 1). For example, the ground state Lamb shift in hydrogen is 8173 MHz, which should be compared with the 1S-2S frequency of 2.5×10^{15} Hz. The Lamb shift scales approximately as n^{-3} and Z^4 , and is therefore largest for ground states and high- Z ions.

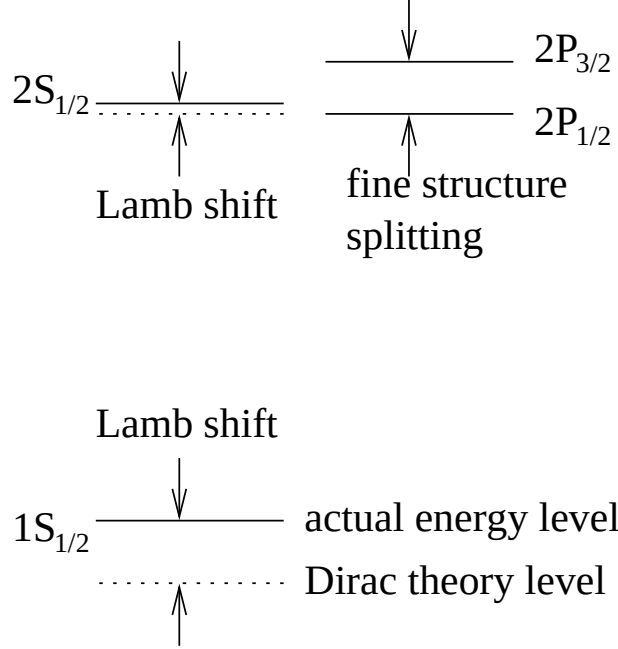


Figure 1. Term diagram of a hydrogen-like ion, showing the 1S and 2S Lamb shifts (not to scale).

The dominant one-loop radiative correction to a hydrogenic S-state may be written as

$$\Delta E_{\text{one-loop}} = \frac{\alpha}{\pi} \frac{(Z\alpha)^4}{n^3} F_n(Z\alpha) m_e c^2, \quad (3)$$

where $F_n(Z\alpha)$ is a slowly varying function of Z (see e.g. [11]). This consists of two terms: the vacuum polarization term and the larger self-energy term. The theoretical uncertainty arising from these terms is negligible compared with the current experimental level of precision; for example, the one-loop self-energy has recently been calculated with a numerical accuracy of 2.4 parts in 10^{16} for the 1S state of hydrogen [13].

However, a significant source of theoretical uncertainty arises from higher-order QED terms, in particular the two-loop binding corrections, for which different calculations are currently in disagreement. The energy shift due to the two-loop corrections may be written in the form [11]:

$$\Delta E_{\text{two-loop}} = m_e c^2 \left(\frac{\alpha}{\pi} \right)^2 \frac{(Z\alpha)^4}{n^3} G_n(Z\alpha), \quad (4)$$

where $G_n(Z\alpha)$ is usually written as a double expansion in powers of $(Z\alpha)$ and $\ln(Z\alpha)$:

$$G_n(Z\alpha) = B_{40} + (Z\alpha)B_{50} + (Z\alpha)^2 \{ B_{63} \ln^3(Z\alpha)^{-2} + B_{62} \ln^2(Z\alpha)^{-2} + \dots \}. \quad (5)$$

The leading-order term in this expansion has been known for many years [14] and is 1.017 MHz for the 1S state of hydrogen. However, recent calculations have yielded a larger than expected value for the B_{50} term, of order $\alpha^2(Z\alpha)^5$ [15,16], which is -0.328 MHz for the ground state of hydrogen. Higher-order logarithmic terms (the B_{63} and B_{62} terms) may also be larger than anticipated [17-20], although there appears to be disagreement between these calculations which is not yet fully explained. There are indications that the convergence of the series expansion is very poor, and that it would be desirable to carry out a high-accuracy numerical calculation which avoids the expansion in $(Z\alpha)$, as has been done for the one-loop self energy. It is possible that this might be achieved using the partial wave mass renormalization technique of Quiney and Grant [21,22]. Clearly, however, experimental Lamb shift measurements accurate enough to be sensitive to these contributions would be most interesting. At higher Z the two-loop binding corrections are a fractionally larger part of the Lamb shift due to their rapid Z scaling. This is the motivation of the experimental work at NPL and Oxford University to measure the 2S Lamb shift in hydrogenic Si^{13+} , which forms part of the Foundation Programme “Hydrogenic Systems Calculable Frequency Standards” project 4.2. This work is discussed further in section 5 and in references [23] and [24] (included in this report as appendices 1 and 2).

A further source of theoretical uncertainty arises in the nuclear size correction to the energies of the S states. The nuclear size uncertainty is currently the most significant source of uncertainty for the hydrogen atom, for which there are two experimental values for the root mean square charge radius of the proton that disagree with each other [25,26], giving rise to a 149 kHz difference between the corresponding 1S ground state Lamb shift estimates [27]. This discrepancy may be clarified in time via further determinations of the proton size. In particular there is an experiment planned to measure the Lamb shift in muonic hydrogen, to which the proton size contributes about 2% [28]. This is expected to lead to a determination of the proton size to 1 part in 10^3 , which corresponds to an uncertainty of about 2 kHz in the 1S Lamb shift in hydrogen. Alternatively, given the new, precise spectroscopic data, and assuming the correctness of the QED analysis, the proton charge radius can be determined [29]. In the light of this calculation the results from the electron scattering data have been re-analysed [30] and better agreement with the spectroscopically derived result obtained. If the value of the proton size from the muonic hydrogen experiment turns out to be in agreement with this re-analysis then the situation would be much improved.

3. Hydrogen

Hydrogen, with its rich spectrum of resonances from the rf to the UV region of the spectrum (see Figure 2), has long provided a challenge to experimentalists as a testing ground for physical theories and a means of making precision measurements of fundamental constants. For more than a hundred years, spectroscopists have been working to measure and compare the frequencies of hydrogen with ever-increasing resolution and accuracy. The 1S-2S two-photon transition in particular, with its extremely narrow linewidth of 1.3 Hz, has inspired significant advances in high resolution spectroscopy and pushed the limits of optical frequency metrology (Figure 3).

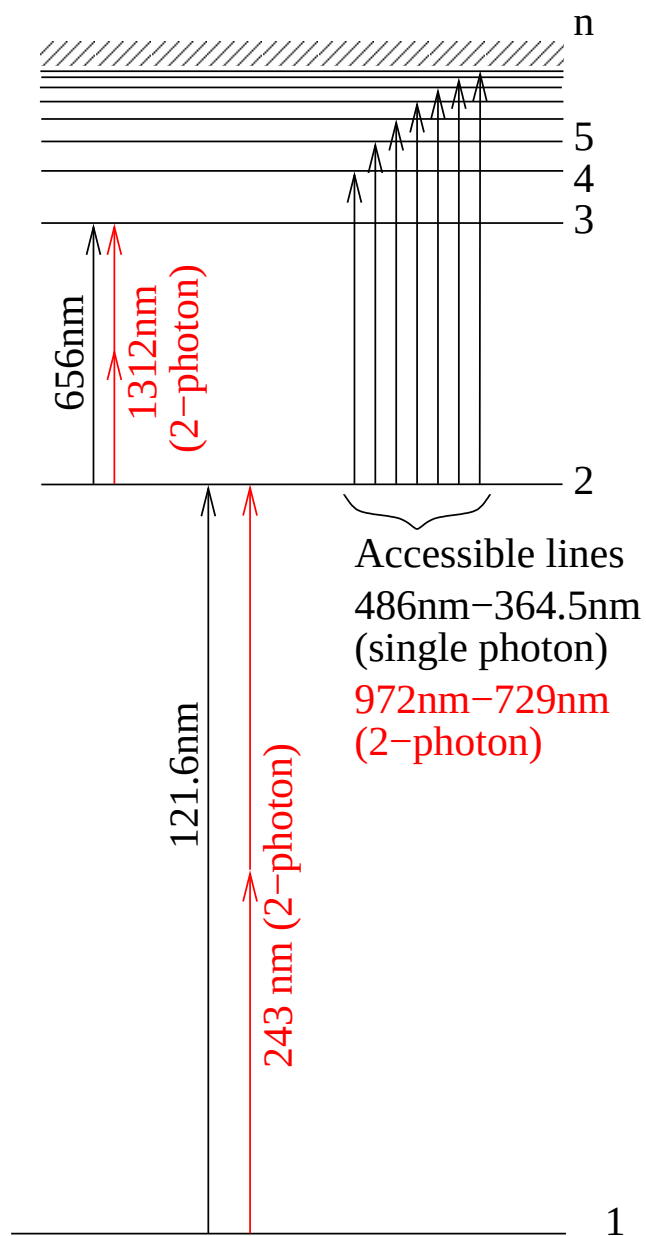


Figure 2. Principal energy levels of atomic hydrogen, showing selected transitions discussed and those accessible for study using a Ti:sapphire laser.

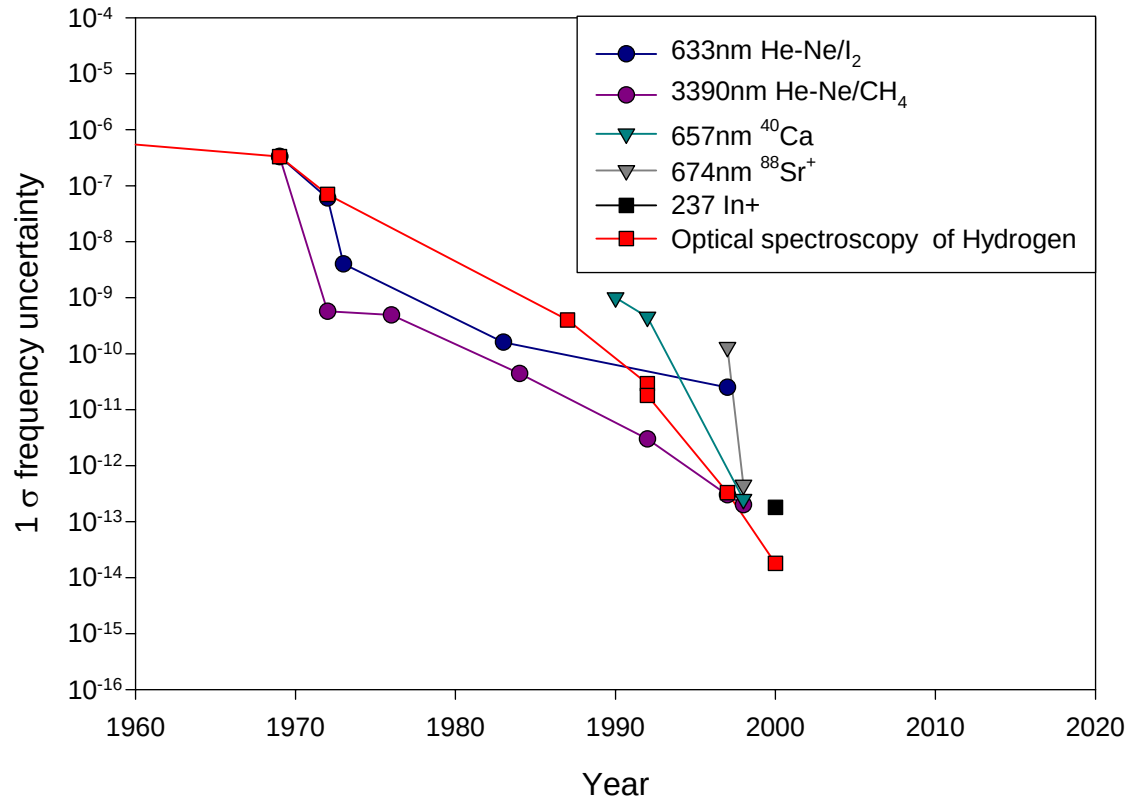


Figure 3. Improvements in optical frequency standards. The most precise measurements have been made by direct comparison with a caesium fountain using a femtosecond laser comb.

This section reviews some of the more recent measurements of transition frequencies in hydrogen, both optical and radiofrequency, and considers some of the recent developments which are likely to lead to further advances in the future.

3.1. Transitions with $\Delta n \geq 1$

3.1.1. Transitions starting from the 1S level

The technique of Doppler-free two-photon spectroscopy has led to impressive advances in the spectroscopy of hydrogen. Not only does it lead to an increase in resolution by eliminating the first-order Doppler shift, but it also enables transitions to be studied that are intrinsically narrower than those accessible using one-photon techniques, because it is possible to avoid the P states.

Spectroscopic studies of transitions starting from the 1S state fall into two categories. In the first, Lamb shifts have been deduced without the need for absolute frequency measurement by comparing transitions with a simple frequency ratio such as 4:1 [31-34]. The second category of experiment is that in which an absolute measurement of the transition wavelength or frequency is carried out either by comparison with an optical standard or using a frequency chain.

Since the advent of Doppler-free two-photon spectroscopy, which enabled the intrinsically narrow 1S-2S transition in hydrogen to be studied for the first time, impressive improvements in the measurement of its absolute frequency have been achieved (see Figure 4).

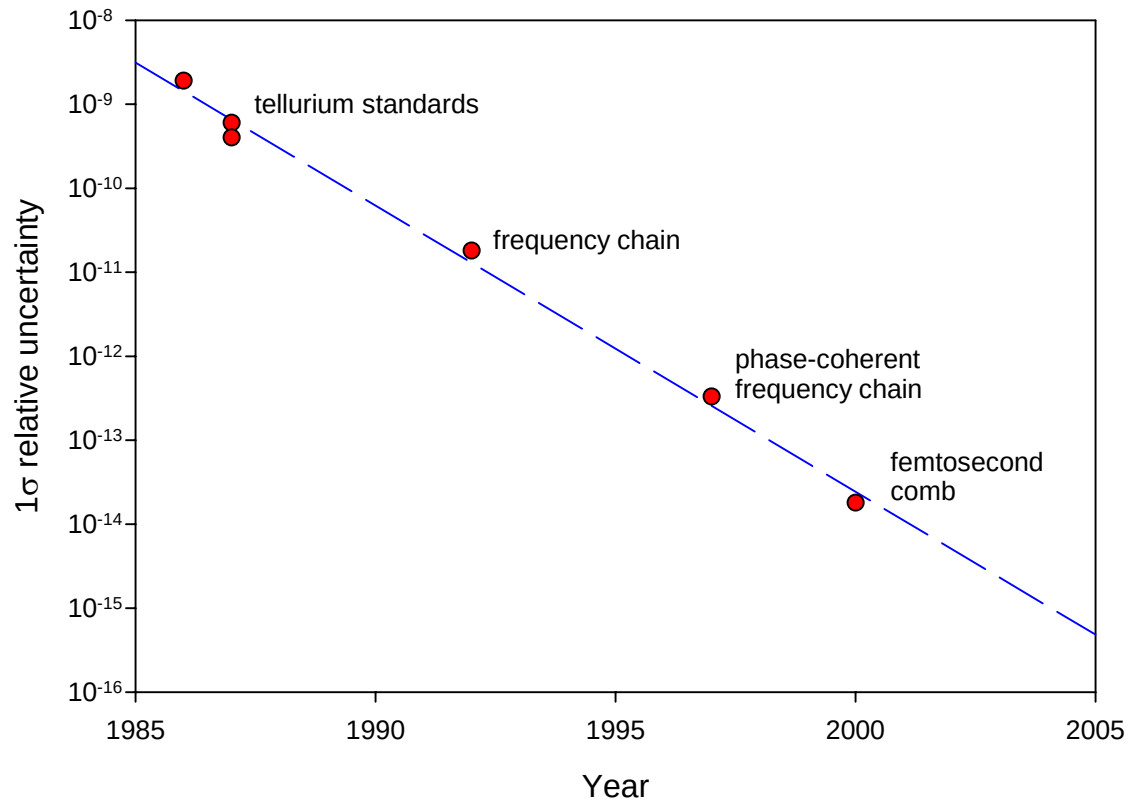


Figure 4. Accuracy of measurements of the 1S-2S two-photon transition in hydrogen.

The early absolute frequency measurements of this transition [35-37] used tellurium standards for calibration. A major improvement in accuracy was achieved by Hänsch and co-workers who developed a frequency chain to relate the 1S-2S interval to a methane-stabilized helium-neon laser standard (HeNe/CH₄) at 3.39 μ m [38]. This made use of the fortunate coincidence that the frequency of the laser at 486 nm, which is doubled to excite the two-photon transition, lies close to the 7th harmonic of the HeNe/CH₄ standard. Originally the frequency mismatch of 2.1 THz was bridged interferometrically, but in a later measurement of the 1S-2S transition frequency, it was determined using a phase-coherent chain of five cascaded optical frequency dividers [9]. This enabled the absolute frequency of the transition to be determined to 3.4 parts in 10^{13} , limited by the reproducibility of the HeNe/CH₄ standard.

Ideally, an optical frequency should be compared in a phase-coherent way with the 9.2 GHz hyperfine transition in the caesium atom which is currently used to define the second. In the past this could only be achieved by the development of very large and complex frequency multiplication chains. However novel techniques have recently been developed for bridging large frequency intervals in the optical region using mode-locked femtosecond lasers [6] and it is now possible for the link between the optical and microwave regions to be made in a single step [7]. These techniques have enabled the optical frequency of the 1S-2S transition to be compared directly with the microwave frequency of a caesium atomic clock. Coupled with higher spectral resolution and an improved theoretical model of the lineshape [39], this has resulted in a new measurement made using a caesium fountain reference with an uncertainty of 1.8 parts in 10^{14} or 46 Hz [40], compared with a natural linewidth of 1.3 Hz.

These experiments were carried out using a cooled beam of atoms, but technology is now being developed that may in the future allow such experiments to be done on trapped hydrogen atoms, with the possibility of bringing the resolution down to the natural linewidth. These developments are discussed further in section 3.2.

3.1.2. Other $\Delta n \geq 1$ transitions

The Rydberg constant is currently known to 8.3 parts in 10^{12} [10]. An absolute frequency measurement of the 1S-2S interval alone does not give the Rydberg constant because of the uncertainties in the QED corrections and the nuclear size. In order to separate out the Rydberg constant and the Lamb shift at least two, and preferably more, different transition frequencies must be measured [9]. Unfortunately, however, none of these have such a narrow natural linewidth as the 1S-2S transition (see Table 1).

n	Natural width / kHz		
	S	P	D
2	0.0013	100000	-
3	1000	30000	10000
4	680	13000	4400
5	430	6600	2300
6	280	3900	1300

Table 1. Natural widths of hydrogen atomic energy levels. For high n states, the widths scale approximately as n^{-3} .

Currently the best measurements of 2S- n S,D transition frequencies in hydrogen are those for 2S-8S,D [41] and 2S-12D [8], which were carried out by Biraben's group at the Laboratoire Kastler Brossel (LKB) in Paris using a metastable atomic beam. These transitions have been measured with an accuracy as high as 6 parts in 10^{12} using the frequency chain at LPTF, requiring the two laboratories to be linked by a 3 km long optical fibre. The 2S-8S,D measurements made use of

the close proximity of the transition frequencies to that of the rubidium two-photon standard, while the 2S-12D measurement required an optical frequency divider to relate the transition frequency to those of the rubidium standard and a CO₂/OsO₄ standard.

The laser system used in the Paris experiments to promote atoms from the metastable 2S state to higher-lying nS and nD levels consisted of a Ti:sapphire laser and a build-up cavity around the atomic beam. However, although the large tuning range of Ti:sapphire in principle allows any 2S- nS,D transition with $n \geq 4$ to be studied, the transitions which could be measured in practice were limited by the need for near-coincidences with suitable optical frequency standards. The development of new optical frequency metrology techniques based on femtosecond lasers has removed this restriction, and opens up the possibility of studying a number of different transitions under identical conditions. This should enable more stringent tests of systematic effects than in the previous experiments.

The principal sources of uncertainty in these measurements were the statistical uncertainty and the uncertainties in the theoretical model used to fit the line profiles, which were broadened by a large inhomogeneous light shift (see Table 2). To obtain improved accuracy, one can consider using optical excitation rather than electron bombardment to produce the metastable ions. This would provide the advantages of lower angular dispersion of the metastable beam, slower atoms and no electric field in the atom-laser interaction region. A more intense, slower beam would enable a lower laser power to be used to excite the 2S- nS,D transitions, reducing light shifts. Optical excitation is also likely to provide a better-defined metastable beam geometry and therefore to simplify the evaluation of the non-linear light shift. The Paris experiments also used electric field quenching to determine the number of metastable hydrogen atoms reaching the detection region; it is possible that fluorescence detection of the transitions could provide better signal-to-noise in addition to reducing the ambient electric field.

Source of uncertainty	Uncertainty / kHz	
	2S-8D _{5/2} [41]	2S-12D _{5/2} [8]
Rubidium two-photon standard	2	2
2 nd order Doppler shift	1	1
dc Stark effect	negligible	2.1
Theoretical lineshape model (including light shift)	3	4.5
Statistics	3.2	3.9
Total	4.9	6.7

Table 2. Uncertainties associated with atomic beam measurements of 2S- nD two-photon transition frequencies in hydrogen.

3.2. Cooling, trapping and antihydrogen

The work of Kleppner et al. at MIT on the cooling and trapping of atomic hydrogen has, after many years of effort, led to the achievement of Bose Einstein condensation [42,43]. In the process, a number of techniques which will be useful in the precision spectroscopy of atomic hydrogen have been developed. They have trapped atomic hydrogen in a static magnetic trap, loading with atoms which are pre-cooled in a low temperature source, and then further cooling by evaporation to the condensation transition temperature of 50 μ K. Recombination is suppressed by trapping atoms in only one of the ground state hyperfine levels. Evaporative cooling is used rather than laser cooling because a suitable laser source for laser cooling at the Lyman- α wavelength is not available. However advances are being made and recently the first generation of continuous wave radiation at this wavelength was reported [44]. Pulsed Lyman- α sources have previously been used for cooling and spectroscopy of atomic hydrogen.

The 1S-2S transition has already been studied in trapped hydrogen [45]. Indeed, the cold collisional frequency shift of this transition was used to diagnose the presence of the condensate [42]. The existence of this shift would mean that to be useful as an optical frequency standard the trap would have to be operated at low density ($<10^{10} \text{ cm}^{-3}$, even though densities of 10^{14} cm^{-3} are achievable) in order for the collisional shift to be of the same magnitude as the 1S-2S linewidth. However, this density would still give large signal rates [46]. Another possibility would be to use the condensate as a source for an atomic hydrogen fountain.

These techniques are also being applied and developed in the quest to produce cold atomic anti-hydrogen. One of the first experiments that is planned once this has been achieved is the comparison of the 1S-2S transition frequency with that in normal hydrogen, where any difference would imply a breaking of CPT invariance. In this case it is expected that the spectroscopy will have to be done on only 10 –1000 atoms, so efficient trapping and laser cooling are requirements and the large collaborations based at CERN are working towards those goals [47-50]. These techniques would add greatly to the performance of a hydrogen-based optical frequency standard.

Trapped hydrogen studies have also been made on circular Rydberg states, i.e. atoms excited to states of maximum orbital angular momentum $l = n-1$. Transitions between high n states around $n=30$ are studied because in these highly excited states the nuclear contributions to the energy levels are negligibly small [51]. For these states relativistic and radiative corrections are small and calculable although the energy levels are more sensitive to electric fields. These transitions occur in the microwave region of the spectrum and hence offer the possibility of calculable microwave frequencies if the systematic effects can be sufficiently well controlled. Since they are in the microwave region, it is also straightforward to measure the frequency of these transitions relative to the caesium standard.

3.3. Hyperfine structure transitions in hydrogen

Until recently the hydrogen maser had the best short-term stability of any frequency standard, and it is still in widespread use, although in the last few years microwave transitions in trapped ion standards have achieved even better stabilities (e.g. in Hg^+ a shot noise limited stability of $\sigma_y(\tau) = 6.5 \times 10^{-14}/\tau^{1/2}$ was achieved at 8 seconds [52]). The 1.42 GHz hydrogen-maser transition is between the $F = 1, m_F = 0$ and $F = 0, m_F = 0$ ground state hyperfine levels. Calculations of the interval are limited at the 10^{-6} level by a term depending on the proton form factor and polarisability [1].

Since the hyperfine transition can be measured far more precisely it cannot be regarded as a calculable frequency standard, but it is worthy of a diversionary consideration here due to its current widespread use in frequency metrology. At the time of the development of the hydrogen maser consideration was given to the use of hydrogen in a microwave beam clock, using the same 1.42 GHz hyperfine transition, but in a configuration more like that of a caesium beam system [53,54]. This proposed to overcome the problems of wall shifts in hydrogen masers, which prevent the unperturbed frequency from being obtained at a precision better than about 10^{-12} . This was one of the factors that prevented the hydrogen hyperfine transition being adopted as the defining transition of the SI second. Indeed it may have been adopted except that the caesium standard was better researched at the time. If uncomplicated by the wall shift, the relative simplicity of the hydrogen hyperfine spectrum should allow a better estimate of the unperturbed transition frequency. The energy levels in hydrogen are well separated compared with caesium and couple to different rf field polarities; they can therefore be isolated more successfully, making the difficult analysis needed to obtain the unperturbed frequency less critical.

The principal problems with the use of hydrogen in a beam system are the low mass of the atom (implying the need for cold sources), the inefficiency of detectors and the low frequency of the transition. Modern techniques can overcome these problems to some extent. Hydrogen beam sources at low temperatures have been developed [55] and bolometric detection promises great improvements over the ionisation based detectors. The transition frequency of course remains fixed unless an alternative transition is chosen, radically changing the situation.

Using the techniques outlined above it may well be possible to build a device with the anticipated performance for the hydrogen beam. However the competition would be a rubidium fountain where uncertainties approaching 10^{-16} are predicted [56]. Interestingly, the projections of stability performance made in the 1970s for the hydrogen beam system have, at the time of writing, already been exceeded by conventional active hydrogen masers, a warning for accuracy predictors.

A second exciting development is the cryogenic hydrogen maser. This is an extension to the conventional hydrogen maser suggested by Crampton et al. [57]. It was thought that the cryogenic hydrogen maser could have a frequency stability 2-3 orders of magnitude greater than the conventional hydrogen maser, and such devices were realised experimentally [58-60]. However, the study of hyperfine-induced frequency shifts in these devices led to the conclusion that the expected improvements in frequency stability may not be achievable, as the shift was of relatively large magnitude and was dependent on the atomic state distribution [61]. Analogous

shifts in caesium may limit the performance of atomic fountain clocks based on caesium and make rubidium, where the shifts are significantly smaller, more attractive [56,62,63].

4. He^+

The limitations in hydrogen due to the proton size uncertainty may be avoided in He^+ , where the nuclear size uncertainty (that of the alpha particle) is less of a problem. The uncertainty in the most accurate measurement of the alpha-particle charge radius leads to an uncertainty of 84 kHz in the 2S Lamb shift for He^+ , which is significantly smaller than the terms of the two-loop binding corrections [64]. This has motivated an experiment at the University of Sussex, which aims to measure the 2S-3S/D transitions in He^+ using two-photon spectroscopy with a frequency-doubled 656 nm dye laser source [64,65]. This will allow a determination of the 2S Lamb shift in He^+ , which is about 14 GHz. By comparing the results with the measurements in hydrogen, it should be possible to separate out the nuclear size and QED contributions to the Lamb shift. Absolute measurements of the 656 nm frequency should be possible to better than 100 kHz, making the experiment an interesting test of QED effects which should not be clouded by uncertainties in the nuclear size as in hydrogen.

For a number of years there was a nine standard deviation discrepancy between theoretical calculations and the best measurement of the 2S Lamb shift for He^+ , carried out using a quenching anisotropy method, which had an uncertainty of 160 kHz [66]. However, an unexpected polarization-related systematic effect in the experiment has recently been reported and is thought to resolve the situation [67].

A novel approach to He^+ might be to consider the two-photon transition from the ground state. He^+ might be interesting for the scheme proposed by Dehmelt for realising an optical frequency standard using electron shelving [68]. The scheme uses an allowed transition from the ground state for laser cooling and a slow “clock” transition from the ground state to a metastable state as a reference. In such experiments, the fluorescence photons from the cooling transition of a laser-cooled trapped ion are detected using a photomultiplier, with count rates typically upwards of a few kHz. When a photon is absorbed by the reference transition the fluorescence is extinguished until the metastable state decays and a “quantum jump” is observed. The 30.4 nm $1\text{S}_{1/2} - 2\text{P}_{1/2}$ Lyman- α transition in He^+ , which would have to be used for laser cooling, has a very short wavelength. The clock transition would be the $1\text{S}_{1/2} - 2\text{S}_{1/2}$ two-photon transition (analogous to the two-photon transition currently used in hydrogen), for which a laser at 60.8 nm would be required. At first sight the helium ion therefore looks like a very difficult candidate for the electron shelving scheme because of the very short wavelengths required; however, technological developments may make it more tractable in years to come.

For example, there has already been pulsed laser excitation of the 58 nm $1^1\text{S} - 2^1\text{P}$ transition in neutral helium with a measurement of the frequency to better than one part in 10^8 [69,70]. This experiment at the Vrije Universiteit Amsterdam used 584 nm from a ring dye laser which was doubled and focused into a gas jet of nitrogen for pulsed 5th harmonic generation. Such a pulsed high harmonic approach might be one way of driving the $1\text{S}_{1/2} - 2\text{P}_{1/2}$ transition in He^+ at 30.4

nm, and there is a facility at the European Laboratory for Nonlinear Spectroscopy (LENS) in Florence which might be capable of providing such radiation.

5. One-electron systems with $Z > 2$

The energy of gross structure transitions in hydrogenic systems scales as Z^2 . Thus as Z increases the Lyman- α transitions move from the UV to the X-ray region of the spectrum, e.g. at $Z = 12$ (magnesium) the Lyman- α wavelength is 0.84 nm. Highly-ionised one-electron systems can be made in a variety of ways. High velocity beams from heavy ion accelerators, with speeds 5-10 % of the speed of light, can be stripped by a thin foil or gas; alternatively, recoil ions can be excited by passing such a beam through a dilute gas of say neon or argon. One-electron ions can also be produced in plasma sources and electron beam ion traps [71,72]. Heavy ion storage rings (such as the ESR at GSI Darmstadt) have also been used to observe Lyman- α transitions in hydrogen-like uranium and other ions, using deceleration and electron capture in a gas jet target [73]. The accuracy of the best X-ray measurements of the Lyman- α wavelengths in one-electron ions for $Z = 12$ –92 ranges from 5 ppm to 130 ppm (see Table 3).

System and line	Wavelength or energy	Fractional uncertainty	Notes	Reference
Mg^{11+}	1S-2P _{1/2}	842.50(4) pm	4.7×10^{-5}	EBIT, limited by statistics
	1S-2P _{3/2}	841.90(2) pm		
Cl^{16+}	1S-2P _{1/2}	419.055(21) pm	5.1×10^{-5}	Decelerated + charge-transfer
	1S-2P _{3/2}	418.518(17) pm		
Ar^{17+}	1S-2P _{1/2}	373.6514(40) pm	1.1×10^{-5}	Plasma source
	1S-2P _{3/2}	373.1142(70) pm		
Ar^{17+}	1S-2P _{1/2}	373.6522(19) pm	5×10^{-6}	Recoil ion source
	1S-2P _{3/2}	373.1105(19) pm		
Fe^{25+}	1S-2P _{1/2}	178.364(19) pm	1.1×10^{-4}	Beam-foil source
	1S-2P _{3/2}	177.815(19) pm		
Fe^{25+}	1S-2P _{1/2}	178.347(15) pm	8.4×10^{-5}	Beam-foil source
	1S-2P _{3/2}	177.787(18) pm		
Ni^{27+}	1S-2P _{3/2}	153.0336(20) pm	1.3×10^{-5}	Decelerated + charge-transfer
U^{91+}	1S-2P _{3/2}	102.1707(132) keV	1.3×10^{-4}	Decelerated + charge transfer

Table 3. Measurement accuracies of 1S-2P transitions in selected hydrogenic ions.

An EBIT offers the advantage over fast beam sources of minimal Doppler shifts. There is evidence [81] that transitions from hydrogenic ions excited in an EBIT source do not suffer from contamination by spectator electron satellite transitions which limit fast beam and particularly

recoil ion sources. Low X-ray count rates have been a problem in many experiments; however Tarbutt et al [81] have recently demonstrated that high resolution spectra of Lyman- α transitions in medium- Z hydrogen-like ions such as Ar^{17+} and Ti^{21+} can be obtained with a statistical precision of a part per million using a curved-crystal Johann spectrometer on an EBIT, with integration times of a few hours. There remains the difficulty of calibrating such spectra to the same level of precision; methods by which this might be achieved are discussed, and these remain an area for future investigation.

The recent 24 ppm measurement of the $1\text{S}-2\text{P}_{3/2}$ transition in Mg^{11+} using an EBIT avoided the need for reference lines to determine the transition wavelength by using a quasimonolithic crystal arrangement, which was absolutely calibrated relative to optical standards [74]. In this arrangement a pair of parallel crystals Bragg reflect radiation to produce two images on a detector. The separation of the images is a function of the Bragg angle of reflection and the spacing between the crystals. Thus, by measuring the image separation and knowing the spacing, the Bragg angle and hence the wavelength of an X-ray line can be determined absolutely, providing the crystal lattice spacing is known, without the need for X-ray standards.

An alternative approach to absolute X-ray spectroscopy on an EBIT is described by Chantler et al. [82] and Paterson et al. [83]. These authors advocate using a curved crystal spectrometer, with Johann geometry, and describe an implementation at the NIST EBIT with a two-dimensional position-sensitive proportional counter. The calibration procedure uses a fluorescent source of K X-rays on the opposite port to the spectrometer port on the EBIT and involves mapping out the dispersion function of the spectrometer. Observations of Lyman- α in hydrogen-like vanadium ($Z=23$) are presented, but the statistics are poor. However, the same technique has been used to obtain measurements of transitions in helium-like vanadium with a precision as high as 27 ppm [84].

Another problem with current EBITs is their lack of portability. The development of a miniature, portable EBIT device would be very desirable. Some progress in this direction has been made with the demonstration of a small EBIT at the University of Dresden [85]. This is a room-temperature system which does not require cryogenic cooling, and uses permanent magnets; the overall dimensions of the vacuum assembly are less than 50 cm.

Conventional X-ray wavelength standards are K_α X-rays, with the lowest quoted uncertainty being 2.8×10^{-7} for the 8.05 keV Cu K_α X-ray [10]. If Lyman- α transitions in hydrogenic ions can be measured accurately enough relative to K_α wavelength standards, there is the prospect of an alternative validation of the standards via direct calculation of intervals in hydrogenic systems. Indeed if Lyman- α X-rays from hydrogenic systems trapped in an EBIT can be sufficiently well understood, and the limitations of low count rates can be overcome, then it is possible that they might one day be accepted as an alternative standards in the X-ray region of the spectrum.

Owing to the rapid scaling of QED effects with Z , transitions in highly charged ions are much more sensitive than hydrogen to the two-loop binding corrections to the energy levels. Measurements in highly charged hydrogenic ions could therefore test the calculations of these terms, and enable the viability of hydrogen and other hydrogenic systems as frequency standards to be assessed. For example, the $2\text{S}_{1/2}-2\text{P}_{3/2}$ transitions in a number of medium- Z hydrogenic

ions lie in a wavelength range which is accessible to high-power tuneable lasers. The most sensitive measurement to date in this range of Z is that for P^{14+} [86], which has a fractional precision of 0.14% of the 2S Lamb shift. This is about the same size as the self-energy correction of order $\alpha^2(Z\alpha)^5$ (see Figure 5), and hence improved precision is required to obtain a critical test of the theoretical calculations.

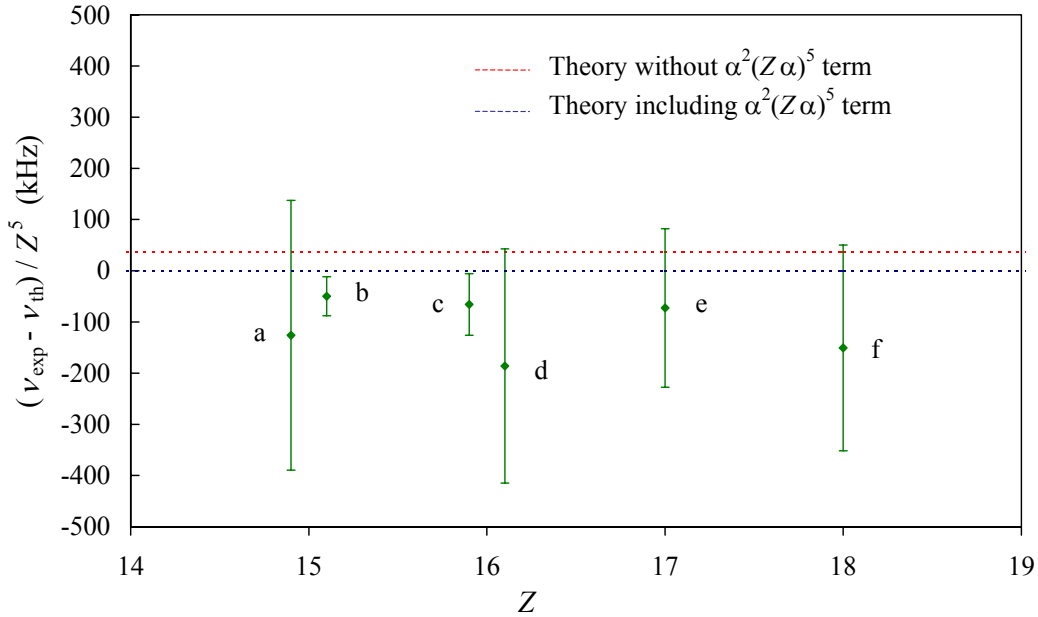


Figure 5. Comparison of experiment and theory for the 2S Lamb shift in hydrogen and hydrogenic ions. The difference between the experimental and theoretical results ($v_{\text{exp}} - v_{\text{th}}$) has been scaled with Z^5 , indicating to what extent each measurement can be interpreted as a test of the two-loop binding corrections of order $\alpha^2(Z\alpha)^5$. The theoretical values are taken from [87], except for the order $\alpha^2(Z\alpha)^5$ term, which is taken from [15]. The experimental values are a: Pellegrin et al., 1985 [88]; b: Pross et al., 1993 [86]; c: Zacek et al., 1984 [89]; d: Georgiadis et al., 1986 [90]; e: Wood et al., 1982 [91]; f: Gould and Marrus, 1983 [92].

The technique for studying the $2S_{1/2}$ - $2P_{3/2}$ transitions in medium- Z hydrogenic ions by laser spectroscopy is illustrated in Figure 6. Laser radiation is used to excite ions from the metastable $2S_{1/2}$ state to the $2P_{3/2}$ state, from which they rapidly decay to the ground state via an allowed electric dipole transition. The resonance is monitored by observing the rate of emission of Lyman- α photons as a function of the laser frequency. The 2S Lamb shift may be deduced from such a measurement of the $2S_{1/2}$ - $2P_{3/2}$ interval because the $n=2$ fine structure splitting is more accurately known theoretically.

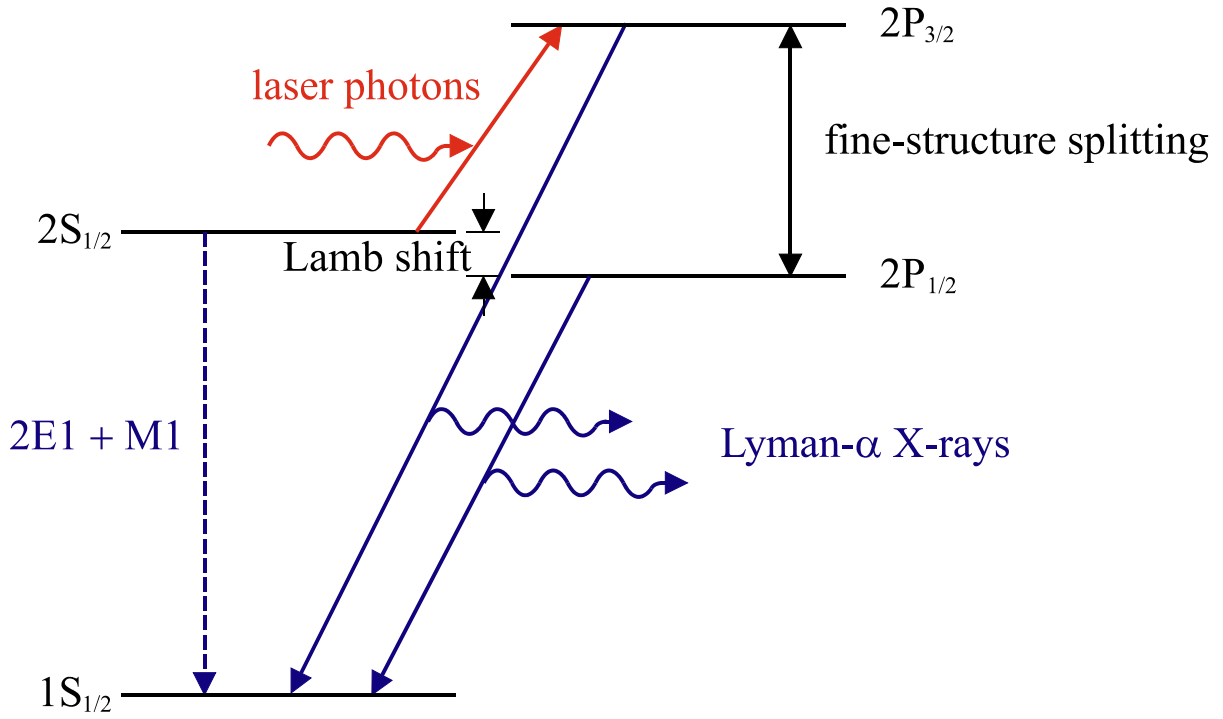


Figure 6. Partial term diagram of a hydrogenic ion, showing the transition induced in an $n=2$ laser resonance experiment. (Not to scale).

All previous 2S Lamb shift measurements for medium- Z hydrogen-like ions have been carried out using fast ion beams, and uncertainties associated with Doppler shifts form a significant source of error in all these experiments. Various methods have been employed or suggested for reducing the sensitivity of fast beam experiments to Doppler corrections [93-95], and a measurement of the $2S_{1/2}$ - $2P_{3/2}$ transition frequency in N^{6+} is currently under way at Florida State University [96]. An alternative approach is to reduce such systematic errors by using slow ions trapped in an EBIT, which have no net centre-of-mass motion. Experiments are in progress to measure the $2S_{1/2}$ - $2P_{3/2}$ transitions in N^{6+} and Si^{13+} using the Oxford EBIT. The Si^{13+} experiment is described in more detail in references [23] and [24], which are included as appendices 1 and 2 of this report.

6. Discussion

This report has considered a number of options for calculable frequency standards. These are summarised in Table 4.

Hydrogenic system and transition	Present best accuracy	Comments
H 1S-2S (two-photon)	1.8×10^{-14}	Currently the best measured optical frequency in a hydrogenic system [40]
H 2S- n S/D (two-photon)	6×10^{-12}	Paris measurements [8,41]
He ⁺ 1S – 2S (two-photon)	-	Not yet studied (see section 4)
He ⁺ 2S – n S/D (two-photon)	-	Experiment underway at the University of Sussex [64]
High-Z Lyman- α	parts in 10^5	See Table 3

Table 4. Options for calculable frequency standards.

The most promising system as a calculable optical frequency standard currently appears to be hydrogen itself; however as laser wavelengths in the UV become more easily available, it may be that He⁺ will become more attractive. At present the wavelengths of Lyman- α transitions in mid-to high-Z hydrogenic systems are significantly less well known than K $_{\alpha}$ wavelengths. However, with the development of new X-ray wavelength measurement techniques and small but intense sources of low energy highly charged ions it is possible that these transitions could one day become an alternative to K $_{\alpha}$ X-ray wavelength standards.

The Rydberg constant has been determined from spectroscopic measurements in hydrogen with an accuracy of 8.3 parts in 10^{12} . Unfortunately a measurement of the narrow 1S-2S interval alone does not give the Rydberg constant because of the uncertainties in the QED corrections and the corrections due to the non-zero nuclear size. To take full advantage of the recent high accuracy 1S-2S frequency measurement requires:

- Improved measurements of other transition frequencies in the hydrogen atom;
- Reduced uncertainty in the QED contributions to the energy levels, in particular the two-loop binding corrections;
- An improved value for the proton charge radius.

In addition to their potential as future X-ray wavelength standards, medium-Z hydrogenic ions are also useful testing grounds for the QED contributions to the energy levels. For example, measurements of the 2S Lamb shift in N⁶⁺ and Si¹³⁺ are underway, with the aim of testing calculations of the two-loop binding corrections.

There are a number of areas which might be considered for future study:

- Improving the accuracy of hydrogen $2S$ - nS/D measurements, which would contribute to an improved Rydberg constant determination;
- Spectroscopy of cooled, trapped hydrogen atoms;
- Improved calculations of the two-loop binding corrections complete to all orders in $Z\alpha$;
- Continuation of experiments to measure Lamb shifts in medium- Z hydrogenic systems by laser excitation, to check higher-order QED corrections;
- Experiments to resolve discrepancies arising from the proton charge radius;
- Development of a small portable EBIT capable of producing high yields of hydrogenic ions;
- Development of techniques for absolute measurement of X-ray transitions observed in an EBIT.

In summary, transitions in hydrogenic systems with $\Delta n \geq 1$ could be useful as calculable frequency standards, providing that QED effects can be sufficiently well understood. In particular, the $1S$ - $2S$ two-photon transition in the hydrogen atom is likely to remain one of the most attractive long-term optical frequency standards.

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Appendix 1

“Laser spectroscopy of the 2S Lamb shift in hydrogenic silicon”

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Laser Spectroscopy of the 2S Lamb Shift in Hydrogenic Silicon

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Abstract. Transitions in highly charged ions are particularly sensitive to QED effects, which scale rapidly with atomic number, Z . An experiment to determine the 2S Lamb shift in hydrogenic silicon, using ions trapped in the Oxford electron beam ion trap (EBIT) is in progress. The laser system required for the experiment is currently under development at the National Physical Laboratory (NPL); this involves locking a frequency-stabilised Ti:sapphire laser operating at 734 nm to a high finesse build-up cavity. This will be used to drive and measure the $^2S_{1/2}$ - $^2P_{3/2}$ transition in Si^{13+} . The transition is much more sensitive to two-loop binding corrections in Si^{13+} than in lower- Z systems. Thus this measurement offers the opportunity to test the uncertainty of theoretical contributions which presently limit the ability of transitions in hydrogen and He^+ to serve as calculable frequency standards. A better understanding of QED effects could also pave the way for calculable X-ray standards based on $\Delta n > 0$ transitions in high- Z hydrogenic systems.

1 Introduction

Transitions in hydrogen and hydrogenic (one-electron) ions are being studied in several laboratories with the aim of improving optical and X-ray frequency standards [1]. Transitions with $\Delta n > 0$ in hydrogen span a large frequency range from radiofrequencies into the vacuum ultraviolet. For hydrogenic ions of higher nuclear charge Z , this range extends into the X-ray region of the spectrum due to the Z^2 scaling of the gross energy level structure. These transitions can be regarded as forming a “natural” frequency scale governed by the Rydberg constant $R_\infty = m_e e^4 / (8\epsilon_0^2 h^3 c)$. Thus hydrogenic systems offer the possibility of a set of frequency standards extending from radiofrequencies into the X-ray region, which are directly related to the fundamental constants via the Rydberg constant.

The two-photon 243 nm $1S_{1/2}$ - $2S_{1/2}$ transition and other transitions in the hydrogen atom have recently been included in a new list of approved radiations for the practical realisation of the metre [2]. The particular interest of these transitions lies in the fact that, uniquely among present optical frequency standards, they may be calculated in terms of the Rydberg constant with an accuracy approaching that with which they have been measured. The latest measurement of

the 1S-2S transition frequency by phase coherent comparison with a microwave Cs fountain clock reports an uncertainty of 46 Hz [3]. However the determination of the 1S Lamb shift in hydrogen is limited by the knowledge of other hydrogenic transitions, e.g. the 2S- n S or the 2S- n D ($n = 8, 12, \dots$) transitions, to 22 kHz [4].

Theoretical uncertainties in the calculation of hydrogen and hydrogenic transition frequencies arise from a number of sources. Of greatest significance for this work are the two-loop QED corrections to the energy levels which are considered in detail later. Other sources of uncertainty not considered further here are: (i) uncertainties which depend on the knowledge of values of the fundamental constants such as the fine structure constant and the electron to proton mass ratio, and (ii) nuclear size corrections which are particularly severe in hydrogen itself because the two most precise measurements of the root mean square charge radius of the proton disagree [5,6]. A new determination of the proton size is underway via a measurement of the Lamb shift in muonic hydrogen [7].

Owing to the rapid scaling of QED effects with atomic number Z , transitions in highly charged ions are much more sensitive than hydrogen to the two-loop binding corrections to the energy levels. Measurements in highly charged hydrogenic ions could therefore test the calculations of these terms, and enable the viability of hydrogen and other hydrogenic systems as calculable frequency standards to be assessed. For example, the $2S_{1/2}$ - $2P_{1/2}$ and $2S_{1/2}$ - $2P_{3/2}$ transitions in a number of medium- Z hydrogenic ions lie in a wavelength range that is accessible to high-power tuneable lasers. The most sensitive measurement to date in this range of Z is that for P^{14+} [8], which has a fractional precision of 0.14% of the 2S Lamb shift. This is about the same size as the self-energy correction of order $\alpha^2(Z\alpha)^5$, and hence improved precision is really required to obtain a critical test of theory.

This paper describes the progress of a laser resonance experiment which aims to measure the Lamb shift in hydrogenic silicon with an accuracy that will allow it to test the two-loop binding corrections mentioned above. This in turn should allow the viability of calculable frequency standards, based on transitions in lower- Z one-electron systems such as hydrogen and He^+ , to be assessed. Following a review of some theoretical contributions to hydrogenic energy levels, the details of the laser resonance experiment are outlined.

2 Theoretical Contributions to Hydrogenic Energy Levels

There have been a number of recent reviews of hydrogenic systems and QED [9]–[12]; these proceedings contain the most extensive and recent information. To calculate transition frequencies in hydrogen to an accuracy comparable with the experimental precision which has been achieved [3], it is necessary to take into account a large number of corrections to the values obtained using the Dirac equation. These include quantum electrodynamic (QED) corrections, pure and radiative recoil corrections arising from the finite nuclear mass, and a correction due to the non-zero volume of the nucleus. The evaluation of these corrections is an extremely challenging task.

Here we briefly mention theoretical sources of uncertainty which arise from higher-order QED terms which were previously neglected or which are in unresolved disagreement. Whilst the one-loop electron self-energy has recently been calculated with high numerical accuracy (2.4 parts in 10^{16} for the 1S state of hydrogen) [13], the situation for the two-loop binding corrections is less satisfactory. The energy shift due to the two-loop corrections may be written in the form [9,11]

$$\Delta E_n = m_e c^2 (\alpha/\pi)^2 [(Z\alpha)^4/n^3] G_n(Z\alpha), \quad (1)$$

where $G_n(Z\alpha)$ is usually written as a double expansion in powers of $(Z\alpha)$ and $\ln(Z\alpha)$,

$$G_n(Z\alpha) = B_{40} + (Z\alpha)B_{50} + (Z\alpha)^2 \{ B_{63} \ln^3(Z\alpha)^{-2} + B_{62} \ln^2(Z\alpha)^{-2} + \dots \}. \quad (2)$$

The leading-order term in this expansion has been known for many years [14], but recent calculations have yielded a larger than expected value for the term B_{50} , of order $\alpha^2(Z\alpha)^5$ [15,16]. Higher-order logarithmic terms (B_{63} and B_{62}) may also be larger than anticipated [17]–[20]; there appears to be disagreement between these calculations which is not yet fully explained. There are indications that the convergence of the series expansion is very poor, and that it would be desirable to carry out a high-accuracy numerical calculation which avoids the expansion in $Z\alpha$, as has been done for the one-loop self energy. Clarification of the theoretical situation would be welcome; meanwhile experimental Lamb shift measurements which are accurate enough to be sensitive to these contributions are most interesting. This is the motivation of the experimental work described below. The approximate size of some of the different contributions to the 2S Lamb shift in Si^{13+} is given in Table 1.

3 Experiment

The technique for studying the $2S_{1/2}$ – $2P_{3/2}$ transitions in Si^{13+} by laser spectroscopy is illustrated in Fig. 1. Laser radiation at 734 nm is used to excite ions from the metastable $2S_{1/2}$ state to the $2P_{3/2}$ state, from which they rapidly decay to the ground state via an allowed electric dipole transition. The resonance is monitored by observing the rate of emission of 2 keV Lyman- α X-ray photons as a function of the laser frequency. The 2S Lamb shift may be deduced from such a measurement of the $2S_{1/2}$ – $2P_{3/2}$ interval because the $n = 2$ fine structure splitting is more accurately known theoretically.

All previous 2S Lamb shift measurements for medium- Z hydrogen-like ions have been carried out using fast ion beams, and uncertainties associated with Doppler shifts form a significant source of error in all these experiments. Various methods have been employed or suggested for reducing the sensitivity of fast beam experiments to Doppler corrections [22]–[24]. A measurement of the $2S_{1/2}$ – $2P_{3/2}$ transition frequency in N^{6+} using a fast ion beam is currently under way at Florida State University [25]. Our approach, however, is to reduce such

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Table 1. Size of some contributions to the Lamb shift of the $2S_{1/2}$ state in Si^{13+} . All values are taken from reference 21 except the order $\alpha^2(Z\alpha)^5$ term which is from reference 9. The two-loop logarithmic terms are not included due to the current discrepancy in the values obtained from different calculations (see text)

	Energy shift (meV)
Order α self-energy	67.681
Vacuum polarization	-3.984
Finite nuclear size	0.328 (± 0.005)
Relativistic recoil	0.019 (± 0.019)
Relativistic reduced mass	-0.003 (± 0.003)
Two-loop radiative corrections:	
$\alpha^2(Z\alpha)^4$	0.021
$\alpha^2(Z\alpha)^5$	-0.083
Total	63.97

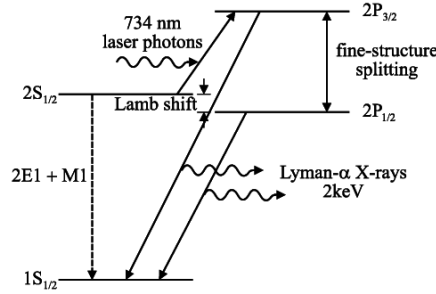


Fig. 1. Partial term diagram of the Si^{13+} ion, showing the transition that would be induced in an $n = 2$ laser resonance experiment. (Not to scale)

systematic errors by using slow ions trapped in the Oxford electron beam ion trap (EBIT) [26], which have no net centre-of-mass motion.

The operation of an EBIT is described in detail elsewhere [27]. Briefly, neutral atoms or low charge state ions are injected into an electron beam which is compressed to high current density by an axial magnetic field, where they are stripped to high charge states by sequential electron impact ionization. The resulting ions are confined radially by a combination of the space charge of the electron beam and the axial magnetic field, whilst axial trapping is achieved using potentials applied to a series of cylindrical electrodes. In this way, the ions are

confined to a cylindrical volume about $70\text{ }\mu\text{m}$ in diameter and 2 cm in length, where they are further excited by electron impact for spectroscopic measurements. The ionization balance obtained within the trap depends on parameters such as the electron beam energy and background gas pressure.

The $2S_{1/2}-2P_{3/2}$ interval in hydrogen-like Si^{13+} has previously been observed using a fast ion beam [28] but there has been no precision measurement of its frequency to date. Our choice of Si^{13+} is based on the availability of high-power lasers at the appropriate wavelength, the ease of ion production in an EBIT, and the efficiency with which the Lyman- α X-rays can be detected. The $2S_{1/2}-2P_{3/2}$ transition in Si^{13+} is at a wavelength of approximately 734 nm , and has a natural width of about 7 nm , so can be studied using a Ti:sapphire laser. Silicon ions have been injected into the Oxford EBIT using a metallic vapour vacuum arc (MEVVA) ion source [29], and an electron beam energy of $3\text{--}4\text{ keV}$ has been shown to be sufficient to produce the hydrogen-like charge state [30]. The Lyman- α X-rays have an energy of about 2 keV , and may conveniently be observed using a lithium-drifted silicon ($\text{Si}(\text{Li})$) detector.

Although the lifetime of the $2S_{1/2}$ state in Si^{13+} is much longer than that of the $2P_{3/2}$ state, it is still only 16 ns . For this reason, very high laser power (several kW or more) is required to obtain a reasonable transition rate to the $2P_{3/2}$ state [31]. The required intensities may be obtained by using an extremely high finesse enhancement cavity to build up the output power from a frequency-stabilized laser [32].

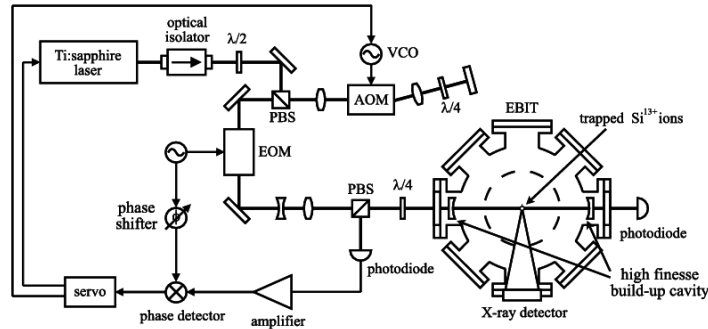


Fig. 2. Schematic diagram of the laser system to be used for a measurement of the $2S_{1/2}-2P_{3/2}$ transition frequency in Si^{13+} . ($\lambda/2$: half wave plate, $\lambda/4$: quarter wave plate, PBS: polarizing beamsplitter, VCO: voltage-controlled oscillator, AOM: acousto-optic modulator, EOM: electro-optic modulator)

Radiation at 734 nm from a continuous-wave, single-frequency Ti:sapphire laser, will be coupled into a high finesse cavity constructed around the Oxford

EBIT as shown in Fig. 2. The trapped Si^{13+} ions will lie at the laser beam waist within the enhancement cavity, and to keep the finesse of the cavity as high as possible it is necessary for the high reflectivity mirrors to lie within the EBIT vacuum chamber. The Ti:sapphire laser will be locked to the high finesse cavity using the rf sideband locking technique [33]. Fast frequency fluctuations will be corrected using an acousto-optic modulator in a double-pass configuration, whilst the slower branch of the servo loop will use a piezo-mounted mirror in the laser cavity.

Construction of a prototype enhancement cavity and the associated locking electronics is currently underway. This demonstration cavity will initially operate in air and consists of two identical mirrors with 20 ppm absorption plus scattering loss, and 200 ppm transmission. The cavity dimensions are somewhat constrained by the ion trap geometry; we intend to use mirrors with 0.5 m radius of curvature, positioned in a near-confocal arrangement. This will give a beam waist radius of around $250\text{ }\mu\text{m}$, sufficiently large to avoid serious difficulties in aligning the laser beam to the trapped ions. Such a cavity has a finesse of around 1.4×10^4 , which for an incident laser power of 100 mW would give an average intensity of about 4 kW mm^{-2} at the beam waist, sufficient to obtain a reasonable $2\text{S}_{1/2}-2\text{P}_{3/2}$ transition rate.

4 Hydrogenic Systems for Calculable Standards

The Si^{13+} Lamb shift experiment described above is designed to test QED two-loop binding corrections which presently limit the ability of transitions in hydrogen and He^+ to serve as calculable frequency standards. The limitations in hydrogen due to the proton size uncertainty may be avoided in He^+ , where the uncertainty in the nuclear size correction is significantly smaller than the two-loop binding corrections. This has motivated a project at the University of Sussex, which aims to determine the 2S Lamb shift in He^+ via measurements of two-photon $2\text{S}-n\text{S}, n\text{D}$ transitions [11]. By comparing the results with the measurements in hydrogen, it should be possible to separate out the nuclear size and QED contributions to the Lamb shift. There was also a nine standard deviation discrepancy between the latest theoretical calculations and the best previous measurement of the 2S Lamb shift for He^+ , carried out using a quenching anisotropy method [34]. However, an unexpected polarization-related systematic effect in that experiment has been reported and may resolve the situation [35].

A novel approach to He^+ might be to consider the two-photon transition from the ground state. He^+ might be interesting for the scheme proposed by Dehmelt for realising an optical frequency standard using electron shelving [36]. The scheme uses an allowed transition from the ground state for laser cooling and a slow “clock” transition from the ground state to a metastable state as a reference. In such experiments, the fluorescence photons from the cooling transition of a laser-cooled trapped ion are detected using a photo-multiplier, with count rates typically upwards of a few kHz. When a photon is absorbed by the reference transition the fluorescence is extinguished until the metastable state

decays and a “quantum jump” is observed. The 30.4 nm $1\text{S}_{1/2}$ – $2\text{P}_{1/2}$ Lyman- α transition in He^+ which would have to be used for laser cooling is very short. The “clock” transition would be the $1\text{S}_{1/2}$ – $2\text{S}_{1/2}$ two-photon transition (analogous to the two-photon transition presently used in hydrogen) for which a 60.8 nm laser would be required. At first sight the helium ion looks like a very difficult candidate for the above scheme because of the very short wavelengths required, however technological developments may make it more tractable in years to come.

Hydrogen and He^+ offer a number of transitions which might serve as calculable frequency standards. At higher Z there are also opportunities to consider. The best measurements of Lyman- α wavelengths in one-electron ions in the range $Z=12$ – 28 have reported accuracies of up to 5 ppm [37]–[39] and measurements of lower accuracy have been made right up to U^{91+} [40], for which the energy of the Lyman- α X-rays is about 100 keV. If the measurements are improved and the QED contributions can be understood with sufficient accuracy, such Lyman- α transitions could one day become calculable frequency standards in the X-ray region.

Measurements of transitions in hydrogenic systems are important both as tests of fundamental physics and for obtaining the Rydberg constant from precision spectroscopy of atomic hydrogen. If the QED corrections to the energy levels can be calculated with sufficient accuracy, then hydrogenic systems offer the prospect of “natural” or calculable frequency standards which are directly related to fundamental constants. This paper has reported the progress of a laser resonance experiment which aims to measure the Lamb shift in hydrogenic silicon with an accuracy that will allow it to test QED two-loop binding corrections. This should allow the viability of calculable frequency standards, based on transitions in lower- Z one-electron systems such as hydrogen and He^+ , to be assessed.

Acknowledgements

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Appendix 2

“Hydrogenic systems: calculable frequency standards”

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HYDROGENIC SYSTEMS: CALCULABLE FREQUENCY STANDARDS

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ABSTRACT

Hydrogenic systems are simple enough that it is possible to calculate the energies of their transitions with an accuracy approaching or exceeding that with which they can be measured. In this paper we discuss the status of measurements and theory for such systems, and consider the current limitations to the accuracy achieved. One source of theoretical uncertainty arises from two-loop quantum electrodynamic corrections to the energy levels. We describe an experiment designed to test these corrections: a measurement of the $2S_{1/2}$ - $2P_{3/2}$ transition in Si^{13+} by laser spectroscopy.

1. INTRODUCTION

The study of the hydrogen atom and hydrogenic (one-electron) ions has emerged as a fertile area for research into future frequency standards [1]. Indeed, the two-photon 243 nm $1S_{1/2}$ - $2S_{1/2}$ transition in the hydrogen atom has recently been included in a new list of approved radiations for the practical realisation of the metre [2]. The particular interest of this interval lies in the fact that, uniquely among present optical frequency standards, it may be calculated with an accuracy approaching that with which it has been measured.

Transitions with $\Delta n > 0$ in hydrogen span a large frequency range from radiofrequencies into the vacuum ultraviolet. For hydrogenic ions of higher nuclear charge Z , this range extends into the X-ray region of the spectrum due to the Z^2 scaling of the gross energy level structure. These transitions can be regarded as forming a “natural” frequency scale governed by the Rydberg constant $R_\infty = m_e e^4 / (8 \epsilon_0^2 h^3 c)$. Thus hydrogenic systems offer the possibility of a set of frequency standards extending from radiofrequencies into the X-ray region, which would be closely linked to fundamental constants via the Rydberg constant.

2. CURRENT STATUS OF HYDROGEN AS A CALCULABLE FREQUENCY STANDARD

2.1 Experiment

Since the advent of Doppler-free two-photon spectroscopy, which enabled the intrinsically narrow $1S_{1/2}$ - $2S_{1/2}$ transition to be studied, impressive improvements in the measurement of its frequency have been achieved (see figure 1).

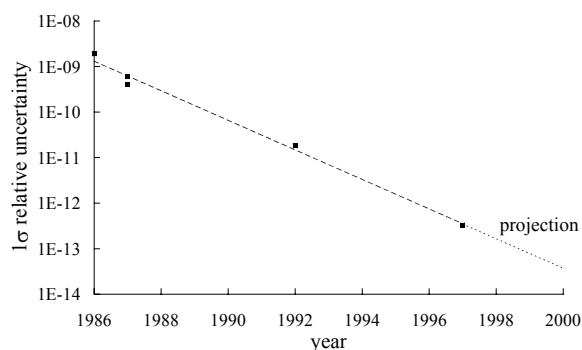


Fig. 1. Accuracy of measurements of the $1S_{1/2}$ - $2S_{1/2}$ two-photon transition in hydrogen.

The early absolute frequency measurements of this transition [3-5] used tellurium standards for calibration. A major improvement in accuracy was achieved by Hänsch and co-workers who developed a frequency chain to relate the $1S_{1/2}$ - $2S_{1/2}$ interval to a methane-stabilized helium-neon laser standard (HeNe/CH_4) at 3.39 μm [6]. This made use of the fortunate coincidence that the frequency of the laser at 486 nm, which is doubled to excite the two-photon transition, lies close to the 7th harmonic of the HeNe/CH_4 standard. Originally the frequency mismatch of 2.1 THz was bridged interferometrically, but in the most accurate measurement of the $1S_{1/2}$ - $2S_{1/2}$ transition frequency to date, it was determined using a phase-coherent chain of five cascaded optical frequency dividers [7]. This enabled the absolute frequency of the transition to be determined to 3.4 parts in 10^{13} , limited by the reproducibility of the HeNe/CH_4 standard. Recently,

techniques have been developed for bridging large frequency intervals in the optical region using mode-locked femtosecond lasers [8], enabling the optical frequency of the $1S_{1/2}$ - $2S_{1/2}$ transition to be compared directly with the microwave frequency of a caesium atomic clock. Coupled with higher spectral resolution and an improved theoretical model of the lineshape which allows the unperturbed transition frequency to be determined to an accuracy of 1.5 parts in 10^{14} [9], a significantly improved absolute frequency measurement is anticipated in the near future.

2.2 Theory

To calculate transition frequencies in hydrogen to an accuracy comparable with the experimental precision which has been achieved, it is necessary to take into account a large number of corrections to the values obtained using the Dirac equation. These include quantum electrodynamic (QED) corrections, pure and radiative recoil corrections arising from the finite nuclear mass, and a correction due to the non-zero volume of the nucleus. The evaluation of these corrections is an extremely challenging task (see for example the review of Pachucki *et al.* [10]).

Uncertainties in the calculations arise from a number of sources. Firstly, the calculation of these corrections depends on the values of certain fundamental constants, namely the electron to proton mass ratio m_e/m_p and the fine structure constant α . The values of m_e/m_p and α in the most recent adjustment of the fundamental constants [11] have relative standard uncertainties of 2.1×10^{-9} and 3.7×10^{-9} respectively. The largest correction depending on m_e/m_p is of order $m_e/m_p \approx 5 \times 10^{-4}$, and so is known with an accuracy of around 10^{-12} . The largest term depending on α is included in the Dirac energy, and is of order $\alpha^2 \approx 5 \times 10^{-5}$; the uncertainty of this term is therefore around 2×10^{-13} . Further refinement of the theoretical calculations beyond these levels will therefore require improved measurements of both these fundamental constants.

Secondly, the nuclear size correction to the energy of the S states currently has a large uncertainty because the two most precise measurements of the root mean square charge radius of the proton disagree [12,13]. This uncertainty comes in at the level of 5×10^{-11} for the 1S state of hydrogen, and is reduced by a factor of n for higher-lying levels. Although the Rydberg constant has been determined with an accuracy of 8.3 parts in 10^{12} by considering other transitions in addition to the $1S_{1/2}$ - $2S_{1/2}$ transition [7,14], a new measurement of the proton radius is greatly to be desired.

The final type of uncertainty arises from higher-order terms which are neglected in the calculations. For example, whilst the one-loop electron self-energy has recently been calculated with high numerical accuracy (2.4 parts in 10^{16} for the 1S state of hydrogen) [15], the situation for the two-loop binding corrections is less

satisfactory. The energy shift due to the two-loop corrections may be written in the form $\Delta E_n = m_e c^2 (\alpha/\pi)^2 [(Z\alpha)^4/n^3] G_n(Z\alpha)$ where $G_n(Z\alpha)$ is usually written as a double expansion in powers of $(Z\alpha)$ and $\ln(Z\alpha)$. The leading-order term in this expansion has been known for many years [16], but recent calculations have yielded a larger than expected value for the term of order $\alpha^2(Z\alpha)^5$ [17,18], and suggested that higher-order terms may also be larger than anticipated [19]. All the indications are that the convergence of this series expansion is very poor, and that it would be desirable to carry out a high-accuracy numerical calculation which avoids the expansion, as has been done for the one-loop self energy.

3. SINGLY IONIZED HELIUM AND HIGHER-Z ONE-ELECTRON SYSTEMS

3.1 He^+

The theoretical limitations of the proton size uncertainty may be avoided in He^+ , where the nuclear size (that of the alpha particle) is well known. This has motivated a project at the University of Sussex, which aims to determine the 2S Lamb shift in He^+ via measurements of two-photon $2S$ - nS, nD transitions [20]. By comparing the results with the measurements in hydrogen, it should be possible to separate out the nuclear size and QED contributions to the Lamb shift. There is also a nine standard deviation discrepancy between the latest theoretical calculations and the best previous measurement of the 2S Lamb shift for He^+ , carried out using a quenching anisotropy method [21]. However, an unexpected polarization-related systematic effect in that experiment is currently under investigation, and may resolve the situation [22].

3.2 Higher-Z hydrogenic ions

The energy of gross structure transitions in hydrogenic systems scales as Z^2 . Thus as Z increases the Lyman- α transitions move from the UV to the X-ray region of the spectrum. Highly charged one-electron ions can be made in a variety of ways. High velocity beams from heavy ion accelerators with velocities 5-10% of the speed of light can be stripped by a thin foil or gas; alternatively recoil ions can be excited by passing such a beam through a dilute gas, e.g. neon or argon. One-electron ions can also be produced in plasma sources or electron beam ion traps.

The best measurements of Lyman- α wavelengths in one-electron ions in the range $Z=12$ -28 have reported accuracies of up to 5 ppm [23-25]. Measurements of lower accuracy have been made right up to U^{91+} [26], for which the energy of the Lyman- α X-rays is about 100 keV. These transitions could one day serve as calculable frequency standards in the X-ray region, if the QED contributions can be calculated with sufficient accuracy.

4. LASER SPECTROSCOPY OF THE 2S LAMB SHIFT IN HYDROGENIC SILICON

4.1 Motivation

Owing to the rapid scaling of QED effects with atomic number Z , transitions in highly charged ions are much more sensitive than hydrogen to the two-loop binding corrections to the energy levels. Measurements in highly charged hydrogenic ions could therefore test the calculations of these terms, and enable the viability of hydrogen and other hydrogenic systems as frequency standards to be assessed. For example, the $2S_{1/2}$ - $2P_{3/2}$ transitions in a number of medium- Z hydrogenic ions lie in a wavelength range which is accessible to high-power tuneable lasers. The most sensitive measurement to date in this range of Z is that for P^{14+} [27], which has a fractional precision of 0.14% of the 2S Lamb shift. This is about the same size as the self-energy correction of order $\alpha^2(Z\alpha)^5$, and hence improved precision is really required to obtain a critical test of theory.

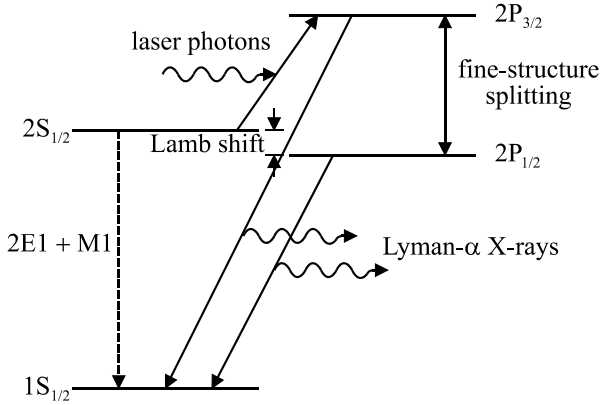


Fig. 2. Partial term diagram of a hydrogenic ion, showing the transition that would be induced in an $n=2$ laser resonance experiment. (Not to scale.)

4.2 Principle of the experiment

The technique for studying the $2S_{1/2}$ - $2P_{3/2}$ transitions in medium- Z ions by laser spectroscopy is illustrated in figure 2. Laser radiation is used to excite ions from the metastable $2S_{1/2}$ state to the $2P_{3/2}$ state, from which they rapidly decay to the ground state via an allowed electric dipole transition. The resonance is monitored by observing the rate of emission of Lyman- α photons as a function of the laser frequency. The 2S Lamb shift may be deduced from such a measurement of the $2S_{1/2}$ - $2P_{3/2}$ interval because the $n=2$ fine structure splitting is more accurately known theoretically.

4.3 Source of highly charged ions

All previous 2S Lamb shift measurements for medium- Z hydrogen-like ions have been carried out using fast ion beams, and uncertainties associated with Doppler shifts form a significant source of error in all

these experiments. Various methods have been employed or suggested for reducing the sensitivity of fast beam experiments to Doppler corrections [28-30]. A measurement of the $2S_{1/2}$ - $2P_{3/2}$ transition frequency in N^{6+} using a fast ion beam is currently under way at Florida State University [31]. Our approach, however, is to reduce such systematic errors by using slow ions trapped in the Oxford electron beam ion trap (EBIT) [32], which have no net centre-of-mass motion.

The operation of an EBIT is described in detail elsewhere [33]. Briefly, neutral atoms or low charge state ions are injected into an electron beam which is compressed to high current density by an axial magnetic field, where they are stripped to high charge states by sequential electron impact ionization. The resulting ions are confined radially by a combination of the space charge of the electron beam and the axial magnetic field, whilst axial trapping is achieved using potentials applied to a series of cylindrical electrodes. In this way, the ions are confined to a cylindrical volume about 70 μm in diameter and 2 cm in length, where they are further excited by electron impact for spectroscopic measurements. The ionization balance obtained within the trap depends on parameters such as the electron beam energy and background gas pressure.

4.4 Choice of ion

We have decided to begin our studies by measuring the $2S_{1/2}$ - $2P_{3/2}$ interval in hydrogen-like Si^{13+} . This transition has previously been observed using a fast ion beam [34] but there has been no precision measurement of its frequency to date. Our choice of Si^{13+} is based on the availability of high-power commercial lasers at the appropriate wavelength, the ease of ion production in an EBIT, and the efficiency with which the Lyman- α X-rays can be detected. The $2S_{1/2}$ - $2P_{3/2}$ transition in Si^{13+} is at a wavelength of approximately 734 nm, and has a natural width of about 7 nm, so can be studied using a Ti:sapphire laser. Silicon ions have been injected into the Oxford EBIT using a metallic vapour vacuum arc (MEVVA) ion source [35], and an electron beam energy of 3-4 keV has been shown to be sufficient to produce the hydrogen-like charge state [36]. The Lyman- α X-rays have an energy of about 2 keV, and may conveniently be observed using a lithium-drifted silicon (Si(Li)) detector.

4.5 Experimental arrangement

Although the lifetime of the $2S_{1/2}$ state is much longer than that of the $2P_{3/2}$ state, it is still only 16 ns. For this reason, very high laser power (several kW or more) is required to obtain a reasonable transition rate to the $2P_{3/2}$ state [37]. Such intensities may be obtained by using an extremely high finesse enhancement cavity to build up the output power from a frequency-stabilized laser [38].

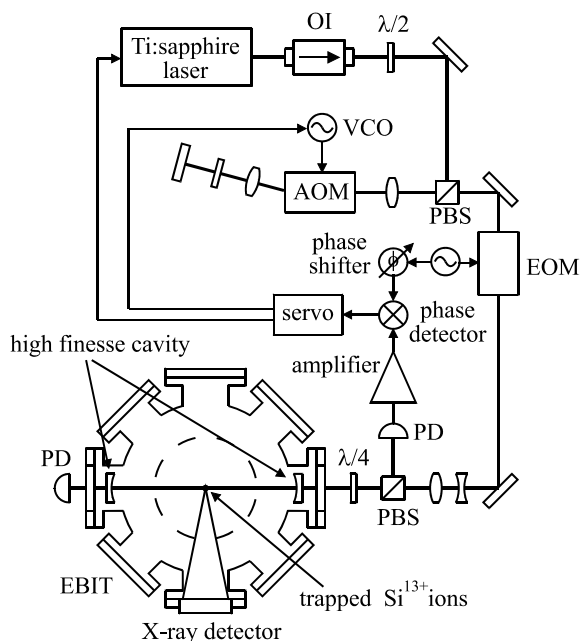


Fig. 3. Schematic diagram of the laser system to be used for a measurement of the $2S_{1/2}$ - $2P_{3/2}$ transition frequency in Si^{13+} . (OI: optical isolator, $\lambda/2$: half wave plate, $\lambda/4$: quarter wave plate, PBS: polarizing beamsplitter, VCO: voltage-controlled oscillator, AOM: acousto-optic modulator, EOM: electro-optic modulator, PD: photodiode.)

The source of 734 nm radiation is a continuous-wave, single-frequency Ti:sapphire laser which will be coupled into a high finesse cavity constructed around the Oxford EBIT as shown in figure 3. The trapped Si^{13+} ions will lie at the laser beam waist within the enhancement cavity, and to keep the finesse of the cavity as high as possible it is necessary for the high reflectivity mirrors to lie within the EBIT vacuum chamber. The Ti:sapphire laser will be locked to the high finesse cavity using the rf sideband locking technique [39]. Fast frequency fluctuations will be corrected using an acousto-optic modulator in a double-pass configuration, whilst the slower branch of the servo loop will use a piezo-mounted mirror in the laser cavity.

Construction of a prototype enhancement cavity and the associated locking electronics is currently underway. This cavity, which will initially operate in air, consists of two identical mirrors with 20 ppm absorption plus scattering loss, and 200 ppm transmission. The cavity dimensions are somewhat constrained by the ion trap geometry; we intend to use mirrors with 0.5 m radius of curvature, positioned in a near-confocal arrangement. This will give a $1/e^2$ beam waist radius of around 250 μm , sufficiently large to avoid serious difficulties in aligning the laser beam to the trapped ions. Such a cavity has a finesse of around 1.4×10^4 , which for an incident laser power of 100 mW would give an average intensity

of about 4 kW mm^{-2} at the beam waist, sufficient to obtain a reasonable $2S_{1/2}$ - $2P_{3/2}$ transition rate.

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