1S–2S Transition-Frequency Calibration

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1. INTRODUCTION

Recent experiments on atomic hydrogen have shown that it is now possible to make measurements with a reproducibility far exceeding the accepted absolute precision of the available visible frequency standards. For example, the recent work of ALLEGRINI et al. [1] has shown that the overall systematic error of their measurements of the Rydberg states in hydrogen is about 4 parts in 10^{11}. Their measurements have had to be referred to the visible iodine stabilised HeNe laser which has an internationally accepted absolute accuracy of only 1.6 parts in 10^{10}. If any progress is to be made in making precision measurements of fundamental atomic constants and in tests of QED, new visible frequency standards and new ways of comparing optical frequencies will have to be developed.

In this contribution we speculate on some new techniques of optical frequency comparison by use of modulated lasers, and on a possible new optical frequency standard based on the methane stabilised HeNe laser. We start by reviewing our own work in providing a convenient secondary frequency standard based on $^{130}Te_2$ transitions in the vicinity of Balmer β in hydrogen and deuterium. We also report on a new and improved measurement of the hydrogen 1S to 2S transition frequency using a pulse amplified laser.

2. TELLURIUM CALIBRATION AT 486 nm

The $^{130}Te_2$ spectrum is extremely rich in the blue region of the spectrum. In collaboration with the NPL we have calibrated two reference lines in $^{130}Te_2$ using Doppler-free saturation spectroscopy [2]. These transitions have been referred to the iodine stabilised HeNe laser using the NPL 1m plane-plane interferometer. A typical $^{130}Te_2$ spectrum in the region of Balmer β in hydrogen is shown in figure 1. The reference line is indicated in this figure. This line lies some 1.4 GHz below one quarter the centroid of the 1S to 2S transition frequency. The lines are approximately 20MHz wide. Some of the problems in using this standard and in

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using interferometry include phase shift corrections on coatings, illumination effects, prismatic imbalances and diffraction effects. Furthermore the cell has to be used at a carefully controlled temperature in order to control the pressure shift. Care has to be taken in using this reference since, for example, we have found that one cell (belonging to Oxford) from the same source has a blue shift of some 1 MHz. This is thought to be due to a leak caused by heating the cell too rapidly. The $^{130}Te_2$ calibration is probably usable to an accuracy of about 4 parts in 10^{10}. We understand that the NPL are going to undertake a more careful study of the $^{130}Te_2$ spectrum which may improve the accuracy somewhat [3].

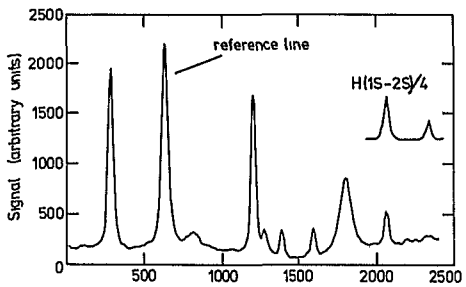

Fig 1.

Doppler-free saturation spectrum of $^{130}Te_2$ in the region of Balmer β in hydrogen shown as a scan over 2.5 GHz. Also shown is an inserted spectrum of the hydrogen 1S to 2S transition at one quarter of the transition frequency.

3. PULSED SPECTROSCOPY OF 1S-2S

The earliest approaches to the observation of the 1S to 2S transition in atomic hydrogen relied on the use of pulsed light sources. This was first done by Hänsch and his co-workers who used pulsed oscillators and amplifiers [4]. This was later improved by amplifying a continuous-wave dye laser [5]. One of the problems of the use of pulsed amplifiers has been the frequency chirp associated with the transient nature of the gain. This effect has been reduced by spectrally narrowing the amplified radiation in a confocal filter at both Stanford [6] and Southampton [7]. In these experiments a continuous-wave C102 dye laser was amplified in a three or four stage amplifier pumped by either an excimer laser [6] or a frequency tripled Nd:YAG laser [7]. The resulting linewidth of a few hundred MHz was spectrally filtered to about 30 - 40 MHz using a confocal filter. The hydrogen 1S to 2S transition frequency was compared with the $^{130}Te_2$ transitions which were calibrated by Southampton/NPL.

The results of these experiments were in reasonable agreement although the Stanford group reported rather smaller errors. Subsequent, continuous-wave experiments at both Stanford and Oxford show poor agreement with the Stanford pulsed result. This has led to speculation that the frequency chirp in pulse amplified experiments is so difficult to characterise that pulse amplifiers cannot be used for precision measurements.

We have undertaken an experiment to try to improve the performance of pulse amplifier experiments. The system is shown schematically in figure 2. It consisted of a continuous-wave C102 dye laser amplified in three stages by a frequency tripled Q-switched Nd:YAG laser. The output energy was approximately 2.0 mJ in a 150 MHz linewidth and was up-shifted from the continuous-wave laser by 60 MHz caused by the frequency chirp. This light was then spectrally filtered in a confocal interferometer with a finesse of 40 and a free spectral range of 300 MHz. The linewidth of the filtered radiation was approximately 16 MHz.

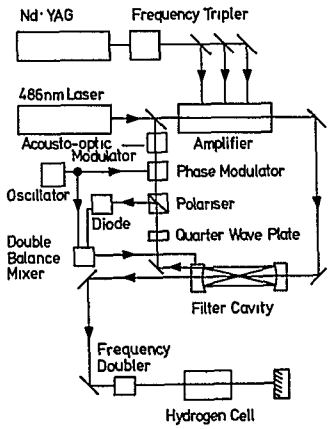

Fig 2 Schematic diagram of the apparatus used to observe the 1S to 2S transition in hydrogen using a pulse amplified laser.

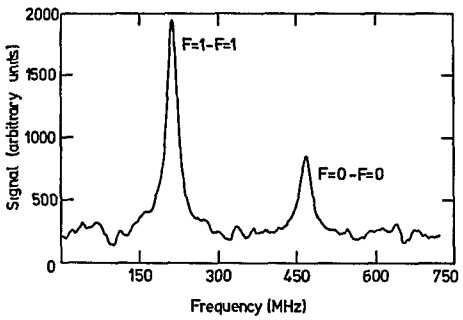

Fig 3 Spectrum of the hydrogen 1S to 2S transition taken using pulse amplified radiation.

The novel aspect of this experiment was that the confocal cavity was locked to continuous-wave radiation which was frequency shifted by an acousto-optic modulator such as to centre the filtering cavity onto the chirped amplified radiation. This reduced the residual amplifier shift to -2(1) MHz. The dominant contribution to this shift resulted from the cw light being injected off-axis into the cavity. Because the filter cavity had a high finesse we used a phase modulation scheme for locking. Indeed, we normally locked the dye laser to the filtering cavity and scanned the spectrum by scanning the filter cavity.

The filtered radiation was then frequency doubled in urea and the 1S to 2S signal was detected by two-photon resonant, three-photon ionisation. A typical spectrum is shown in figure 3 and shows a linewidth of 18 MHz at 486 nm. This spectrum has been compared with the $^{130}Te_2$ reference lines and we have measured a 1S to 2S transition frequency of 2466061407(9) MHz. This compares with 2466061414.13(79) MHz reported for the best continuous-wave measurement [8]. A full report on this measurement is in preparation.

It is clear that the best measurements on the 1S to 2S transition in hydrogen will be accomplished using continuous-wave lasers. However, there is a range of exotic atoms such as muonium, positronium and anti-hydrogen where high energy, high resolution pulsed laser sources will be needed. We believe that with care, precise measurements can be made with pulse amplified radiation. We hope to apply some of these techniques to a study of muonium at the Rutherford Appleton Laboratory (RAL) in a collaboration led by Zu Putlitz and Jungmann at Heidelberg, Hughes at Yale, Baird at Oxford and Barr and myself at Southampton. The RAL muon source is some two orders of magnitude more intense than that used by Chu and collaborators at KEK in Japan [9].

4. COHERENT MULTIPLE PULSE SPECTROSCOPY

The idea of using a train of coherent pulses for the observation of the 1S to 2S transition in hydrogen was first suggested by BAKLANOV et al. in 1976 [10]. The observation of Doppler-free spectra using a coherent pulse train from a synchronously pumped dye laser was demonstrated by ECKSTEIN et.al [11].

In this scheme a mode-locked synchronously pumped dye laser is used to provide a train of tunable light pulses at a precise repetition rate in the region of 100 MHz. The output from the dye laser is passed through the sample and then retro-reflected from a mirror. The distance between the sample and the mirror is adjusted such that a pulse passing through the sample in one direction collides with its neighbour in the train going in the opposite direction. This produces a standing-wave field where a Doppler-free excitation can take place.

The Doppler-free excitation can best be understood in the frequency domain where the train of pulses appears as a comb of modes, all equally spaced in frequency and covering a spectral region which is roughly equal to the inverse of the pulse duration of a single pulse in the train. For a typical dye laser this might be 500 GHz. If one of the modes is in two-photon resonance with the sample

then the adjacent modes higher in frequency and lower in frequency will also be in resonance, as will the modes next higher and lower in frequency, and so on. Thus when one mode is in resonance many of the modes will contribute to the signal. It is easy to show that the signal strength that is expected is the same as that obtained from a single frequency laser of the same average power. A resonance condition also exists when the sum frequency of two adjacent modes equals the atomic resonance frequency.

If we have a two-level sample we expect to see a series of resonances separated by half the inverse of the repetition rate of the laser as the carrier frequency is scanned. If a second transition is within the bandwidth of the laser then this too will give rise to a series of resonances. The resulting spectrum is rather like that obtained from a Fabry-Perot interferometer with overlapping orders. However, in the mode-locked case the modes are precisely equally spaced in frequency.

The mode-locked laser can be used for comparing frequency differences. For example the hydrogen-deuterium isotope shift can be accurately measured using this technique. In this scheme the 671GHz shift can be spanned by a mode-locked dye laser. By scanning a frequency interval of only one mode spacing all the information on the isotope shift can be obtained. An alternative method of using this technique is to lock the mode-locked laser carrier frequency to the 1S to 2S transition frequency. At 486 nm the dye laser will appear as a comb of modes. These modes can be heterodyned against a single frequency laser locked to Balmer β or to a $^{130}Te_2$ reference.

The main features of the coherent multiple pulse technique are: that it is self-calibrating in frequency; that the signal strength is the same as that obtained with a single mode laser of the same average power and that frequency doubling efficiency can be good.

These features are exploited in an experiment which is taking place in our laboratory. A schematic diagram of this experiment is shown in figure 4. The system consists of an all-lines violet mode-locked Kr^+ ion laser operating at a repetition rate of about 250 MHz which synchronously pumps a C102 dye laser. The dye laser typically produces about 300 mW of average power and pulse durations of about 3 psec. This is frequency doubled to 243 nm in a crystal of β-barium borate to produce in excess of 2 mW average power. The output from the second harmonic crystal is then mode-matched into an ultra-violet enhancement cavity. The free

spectral range of this cavity is adjusted to match the repetition rate of the pulse train in such a way that the pulses collide in the centre of the cavity. We have obtained enhancements in excess of x20.

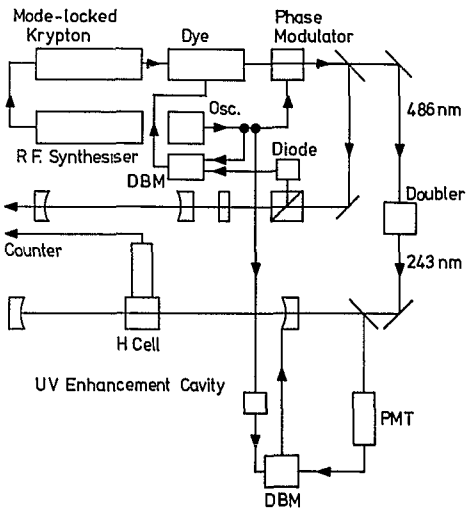

Fig 4 Diagram of the apparatus being used to observe the 1S to 2S transition using a train of ultrashort pulses from a mode-locked dye laser.

Frequency stabilisation and scanning is accomplished by use of a confocal cavity of free spectral range matched to the dye laser repetition rate. Phase modulated sidebands are put on to the mode spectrum of the mode-locked pulse train and used to lock the laser to the reference cavity. The frequency modulation technique is also used to lock the ultra-violet enhancement cavity to the mode-locked pulse train.

Two-photon absorption is detected by observing collisionally induced Lyman α fluorescence in a cell containing atomic hydrogen. This experiment has been set up and we shall report on the results soon.

The mode-locked pulse train is one of a range of ways of comparing optical frequencies. A second technique which we have been investigating is the use of a frequency modulated (FM) dye laser. This has similarities to the mode-locked laser in that we are using the precise nature of the mode spacing when intracavity modulation is applied. In the case of the FM laser phase modulation is applied and in the case of the mode-locked laser amplitude modulation is applied.

5. FM LASER SPECTROSCOPY

An ideal FM laser is a laser which produces an output of constant amplitude but whose instantaneous frequency is sinusoidally modulated about a central carrier frequency. Thus the electric field can be described as

$$E(t) = (E_0/2)\exp(i\omega_0 t + i\Gamma \sin\Omega t) + c.c. \qquad (1)$$

In this case ω_0 is the carrier frequency, Ω is the modulation frequency and Γ is the modulation index which describes the region over which the modulation takes place.

This ideal FM spectrum can be Fourier transformed into the frequency domain to give a spectrum of equally spaced modes with a Bessel function amplitude distribution. These equally spaced modes can be used for comparing optical frequencies by heterodyning a reference laser, unknown laser and FM laser on a nonlinear detector. Three beats can be observed ie the beats between the reference laser and one of the modes of the FM laser, the beats between the unknown laser and one of the modes of the FM laser and the mode spacing of the FM laser. The separation between the reference and unknown laser can hence be deduced.

We have developed FM lasers based on a commercial ring laser (Coherent 699-21). In this case all the intracavity etalons are removed and replaced by a lithium niobate phase modulator. This modulator can be resonantly driven at a frequency close to the cavity mode spacing. A simple theory of FM operation of a laser suggests that the modulation index is given by [12]

$$\Gamma = \left(\frac{\delta}{\pi}\right)\left(\frac{\Omega}{|\omega_m - \Omega|}\right), \qquad (2)$$

where δ is the single pass phase retardation of the phase modulator, ω_m is the cavity mode spacing.

In figure 5 we plot the measured FM bandwidth of a dye laser with a three plate birefringent tuning element as a function of the modulation frequency in the region of matching modulation frequency to cavity mode spacing. There is reasonable agreement between the results and the predictions of equation (2). Bandwidths in excess of 500 GHz have been achieved with this system. For smaller detuning the FM spectrum becomes distorted and FM mode-locking is observed. Wider bandwidth operation has been observed by using a two plate birefringent filter as a tuning element. Bandwidths in excess of 2 THz have been obtained in this case.

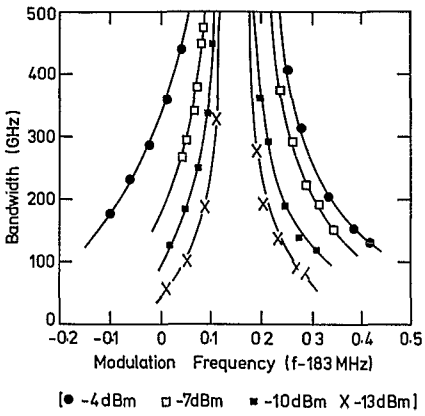

Fig 5 Bandwidth of an FM dye laser tuned by a three plate birefringent filter as a function of the detuning between the passive mode spacing and the driving frequency applied to the phase modulator.

It is important to be able to test the FM laser spectrum to see how closely it approaches that of an ideal FM oscillator. Mode analysis of the FM spectrum is too tedious. We have developed methods of testing the quality of the FM spectrum which involve nonlinear mixing of the FM beams. If we phase shift a part of the FM beam by Θ we obtain a modified field,

$$E'(t) = (E_0/2)\exp(i\omega_0 t + i\Gamma \sin(\Omega t + \theta)) + c.c.$$

If we have a nonlinear process which mixes the beam represented by $E(t)$ and $E'(t)$ we get

$$E(t)E'(t) \propto E_0^2 \exp(i2\omega_0 t + i2\Gamma \cos(\theta/2) \sin(\Omega t + \theta/2)) + c.c. \qquad (3)$$

This represents another FM oscillation centred at a carrier frequency $2\omega_0$ with modulation index $2\Gamma\cos(\theta/2)$. When θ is chosen to equal π we obtain a single frequency at twice the carrier frequency.

We have demonstrated this effect in two experiments [13,14]. One is in single frequency ultraviolet generation by sum frequency mixing and the other is in Doppler-free two-photon spectroscopy, with the FM laser.

Schematic diagrams of the two experiments are shown in figure 6. In the case of the sum frequency experiment a delay is introduced by splitting part of a beam, going through a delay line and recombining the beams with a lens in a nonlinear

crystal. The sum frequency is generated at the bisector of the two beams and is mode analysed by a UV interferometer. In figure 7 we plot some UV spectra taken for an FM bandwidth of about 10 GHz for different values of the phase change $\Delta\theta = \theta - \pi$. When $\Delta\theta = 0$ a single frequency UV spectrum is obtained indicating a clean FM spectrum from the dye laser. As $\Delta\theta$ is increased sidebands appear at the modulation frequency. We have assigned an effective modulation index Γ' which agrees very well with the expression given in equation (3).

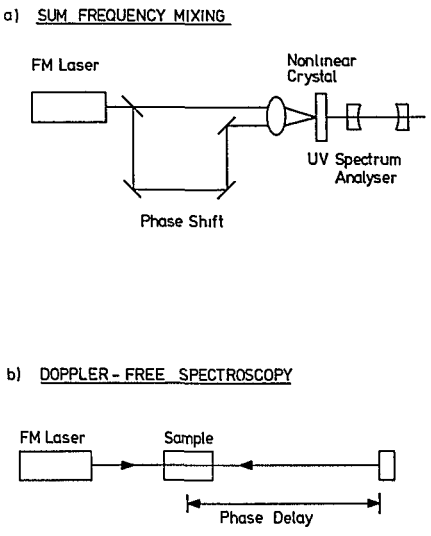

Fig 6 Two experimental arrangements for demonstrating the quality of an FM spectrum a) sum frequency generation in a nonlinear crystal followed by mode analysis of the generated ultraviolet b) Doppler-free two-photon spectroscopy.

The experimental arrangement for the demonstration of Doppler-free two-photon spectroscopy is shown in figure 6 (b). In this case the phase delay is accomplished by adjusting the distance between the sample and the retro-reflecting mirror. In figure 8 (a) we show a Doppler-free two-photon spectrum of the 3S to 4D transition in sodium taken with a single frequency dye laser. Figure 8 (b) shows the identical spectrum obtained with an FM laser. It can be seen that the two sets of spectra are almost identical except that the Doppler background is smaller in the case of the FM laser. This is explained by the fact that the FM bandwidth exceeds the Doppler width and hence only a small part of the FM laser can contribute to the Doppler background.

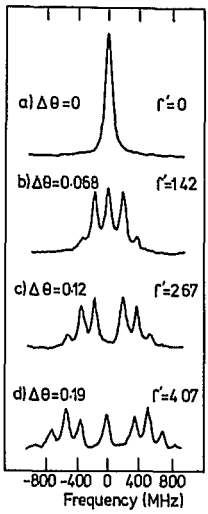

Fig 7 The spectrum of the sum frequency of the FM dye laser for different values of the phase delay.

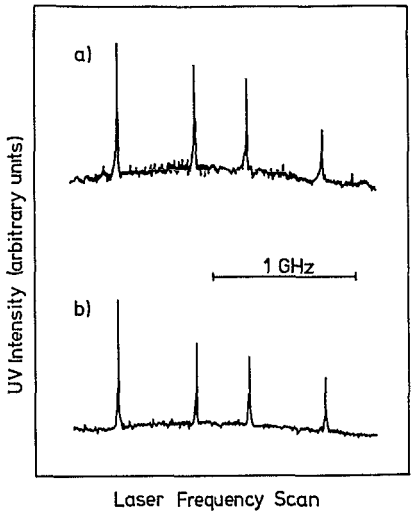

Fig 8 a) Doppler-free two-photon spectrum of the sodium 3S to 4D transition taken with a single frequency dye laser.
b) The same transition observed using an FM dye laser.

6. ABSOLUTE FREQUENCY CALIBRATION

We have developed techniques for the comparison of frequency intervals in excess of 2 THz using modulated dye lasers: either mode-locked or FM. This has still not addressed the problem of obtaining an absolute frequency standard in the visible which can be referred back to the Cs standard of time.

An exciting proposal for generating a more stable visible reference has been to use the harmonics of the methane stabilised HeNe laser. This laser has been developed to a stage where it is stable to better than a few parts in 10^{12} and can be easily referred to the Cs standard[15]. The harmonics of the methane stabilised laser lie in the visible region. For example the seventh harmonic lies at 485 nm. There are a number of schemes for generating the seventh harmonic of the methane stabilised laser. If this could be done it would be only 2.1 THz away from one quarter the 1S to 2S transition frequency in hydrogen. This opens up the possibility of using an FM dye laser to compare a laser locked to the seventh harmonic of the methane stabilised HeNe and one locked to the 1S to 2S transition.

Indeed lasers locked to the harmonics of the methane stabilised HeNe laser may prove to be of importance in the next generation of fundamental measurements in the visible region. It is possible to envisage schemes of harmonic generation and mixing covering the region from 848 nm down to 212 nm with the reference frequencies separated by 88 THz. This would mean that there would be nowhere in the visible or ultraviolet region of the spectrum more than 44 THz away from a reference laser.

7. CONCLUSION

The rapid progress in recent years in the spectroscopy of the hydrogen atom has renewed pressure for a much better optical frequency standard. This in itself would not be enough to solve the measurement problem. New techniques of comparing optical frequencies are needed. We have developed methods of modulating lasers which can be used for frequency differences in excess of 2THz.

New optical frequency standards based on harmonics of methane stabilised lasers will mean that we will never be more than 44 THz away from a reference frequency. New techniques of making frequency interval measurements of this magnitude will then be needed.

8. ACKNOWLEDGEMENTS

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